

Supporting Information

for

An Interrupted Corey-Chaykovsky Reaction of Designed Azaarenium Salts: Synthesis of Complex Polycyclic Spiro- and Fused Cyclopropanoids

Bara Singh,[#] Arshad J. Ansari,[#] Nirmal Malik and S. S. V. Ramasastry*

Department of Chemical Sciences, Indian Institute of Science Education and Research (IISER)
Mohali, Sector 81, S A S Nagar, Manauli PO, Punjab 140 306, India

[#]Contributed equally to this work

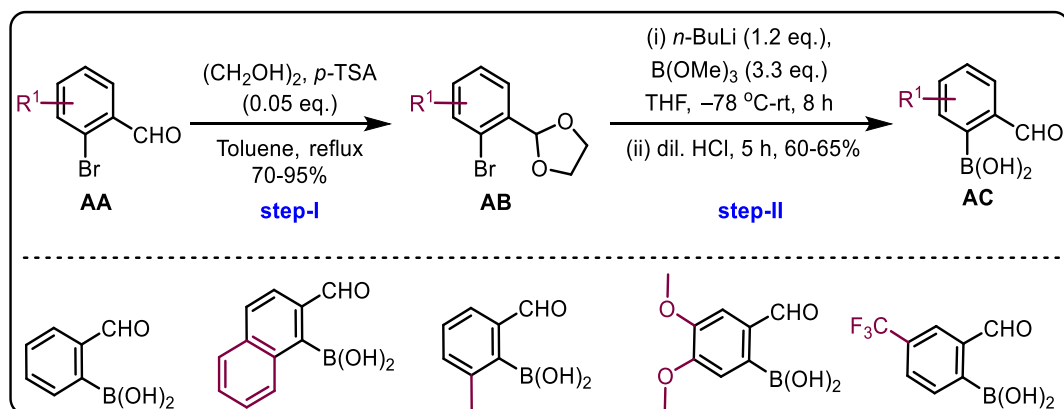
E-mail: ramsastry@iisermohali.ac.in

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General experimental methods: All the reagents, solvents, and catalysts employed in this study were procured from Sigma-Aldrich and were used without further purification. For thin-layer chromatography (TLC), silica aluminum foils with fluorescent indicator 254 nm (from Aldrich) were used, and compounds were visualized by irradiation with UV light and/or by treatment with a solution of *p*-anisaldehyde (23 mL), conc. H₂SO₄ (35 mL) and acetic acid (10 mL) in ethanol (900 mL) followed by heating. Column chromatography was performed using SD Fine silica gel 60-120 mesh (approximately 15–20 g per 1 g of the crude product). Dry THF was obtained by distillation over sodium and stored over sodium wire. As indicated, IR spectra were recorded on a Perkin–Elmer FT IR spectrometer as thin films or KBr pellets, with ν_{max} in inverse centimeters. Melting points were recorded on a digital melting point apparatus Stuart SMP30. ¹H NMR, ¹³C NMR, and ¹⁹F NMR spectra were recorded on a 400 and 500 MHz Bruker Biospin Avance III FT-NMR spectrometer. NMR shifts are reported as delta (δ) units in parts per million (ppm), and coupling constants (*J*) are reported in Hertz (Hz). The following abbreviations are utilized to describe peak patterns when appropriate: br=broad, s=singlet, d=doublet, t=triplet, q=quartet, and m=multiplet. Proton chemical shifts are given in δ relative to tetramethylsilane (δ 0.00 ppm) in CDCl₃ (δ 7.26 ppm) or in (CD₃)₂SO (δ 2.50 ppm). Carbon chemical shifts are internally referenced to the deuterated solvent signals in CDCl₃ (δ 77.1 ppm) or ((CD₃)₂SO (δ 39.5 ppm). Single crystal X-ray analysis was carried out on a Rigaku XtaLAB mini diffractometer. High-resolution mass spectra were recorded on a Waters QTOF mass spectrometer.

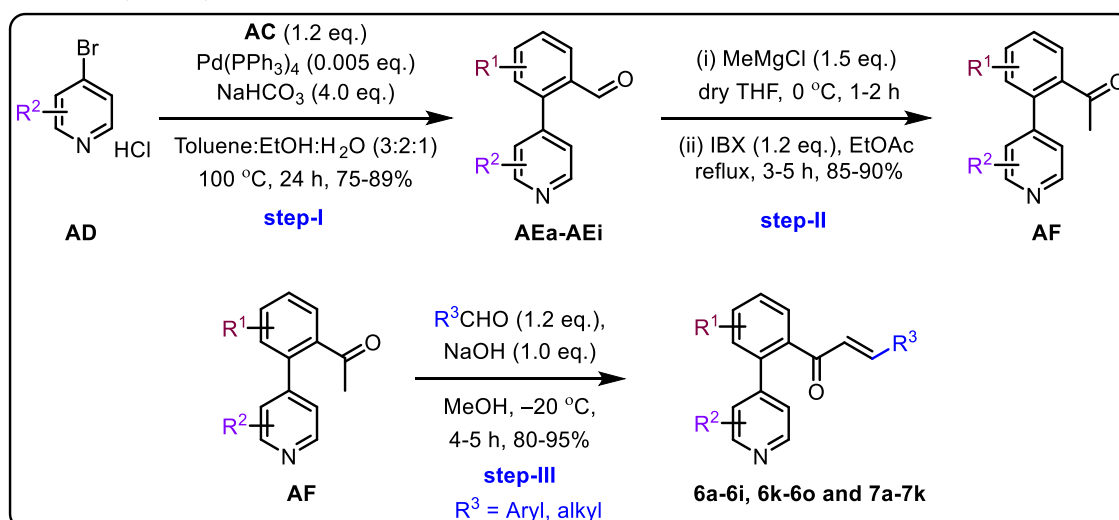
General procedure-1: Synthesis of 2-formyl boronic acids (AC)

All 2-formyl boronic acids were synthesized according to the reported literature.¹



Scheme S1: Synthesis of 2-formyl boronic acids AC

General procedure-2: Synthesis of enone-tethered pyridines (6a-6i, and 6k-6o)- and quinolines (7a-7k)



Scheme S2: General representation of the synthesis of enone-tethered pyridines and quinolines (6a-6i, 6k-6o and 7a-7k)

A representative procedure for the synthesis of AEa-AEi (Scheme S2, step I): Pd(PPh₃)₄ (0.005 mmol), NaHCO₃ (4.0 mmol), 4-bromopyridine hydrochloride or 4-bromoquinolines AD (1.0 mmol), corresponding boronic acid (1.2 mmol), and toluene (3.0 mL), EtOH (2.0 mL) and H₂O (2.0 mL) were added to a sealed tube. The reaction mixture was degassed with nitrogen, and the resulting solution was stirred at 100 °C for 24 h. After the reaction completed (TLC), the reaction was quenched with saturated aq. NH₄Cl and extracted using ethyl acetate. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane-ethyl acetate to afford biaryl aldehydes AEa-AEi (yield 75-89%).

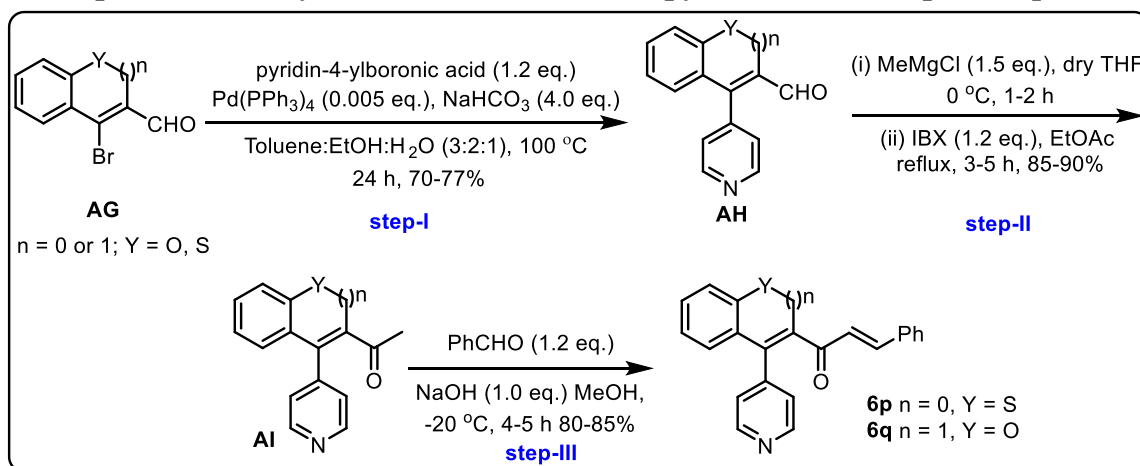
¹ a) Adamczyk-Woźniak, A. A.; Ejsmont, K.; Gierczyk, B.; Kaczorowska, E.; Matuszewska, A.; Schroeder, G.; Sporzynski, A.; Zarychta, B. *J. Organomet. Chem.* **2015**, 788, 36. b) Tseng, N. -W.; Lautens, M. *J. Org. Chem.* **2009**, 74, 1809.

A representative procedure for the synthesis of AF (Scheme S2, step II): An oven-dried 25 mL RB flask was charged with biaryl aldehydes **AE** in 10 mL dry THF and placed at 0 °C under an N₂ atmosphere. Then, methyl magnesium chloride (3.0 M in THF, 1.5 eq.) was added drop wise at the same temperature and stirred for 1 h. Upon completion, the reaction mixture was quenched with water (~2-3 mL) and extracted with ethyl acetate (2x5 mL). The organic extracts were combined, dried over anhydrous Na₂SO₄, and concentrated under reduced pressure. The crude product was forwarded to the next step without any purification.

The crude product was dissolved in ethyl acetate and added IBX (1.2 eq.), and stirred at 80 °C. The reaction progress was monitored by TLC, and on completion, the reaction mixture was filtered through a celite pad and washed with ethyl acetate (2x3 mL). The combined organic layers were washed with brine, dried over anhydrous Na₂SO₄, and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using hexane/ethyl acetate (5:1) as eluent to afford biaryl ketones **AF** (yield 85-90%).

A representative procedure for the synthesis of 6a-6i, 6k-6o, and 7a-7k (Scheme S2, step III): The biaryl ketones **AF** and the corresponding aldehydes (1.2 eq.) were dissolved in MeOH, and KOH (1.0 eq.) was introduced at -20 °C for 2 h. Then, the reaction mixture was shifted to room temperature and stirred for 30 min, monitored the reaction (by TLC) till the complete consumption of the starting material. The reaction mixture was quenched with saturated aqueous NH₄Cl solution (~2-3 mL) and extracted with ethyl acetate (2x5 mL). The combined organic layers were dried over anhydrous Na₂SO₄ and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using hexane/ethyl acetate (5:1) as eluent to afford biaryl ketone **6a-6i**, **6k-6o**, and **7a-7k** (yield 80-95%).

General procedure-3: Synthesis of enone-tethered pyridinium salts (**6p** and **6q**)

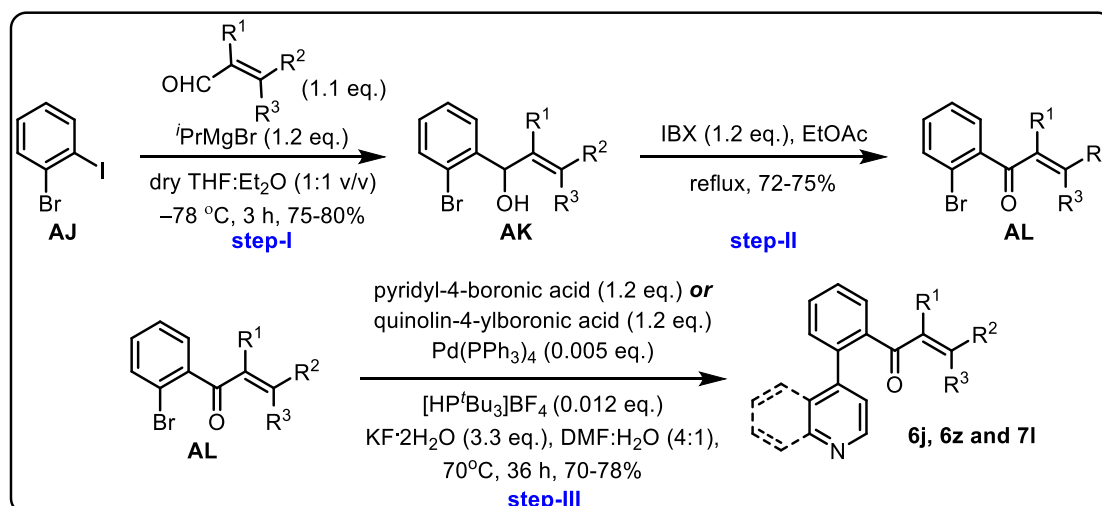


Scheme S3: General representation for the synthesis of **6p** and **6q**

A representative procedure for the synthesis of AH (Scheme S3, step I): Pd(PPh₃)₄ (0.005 mmol), NaHCO₃ (4.0 mmol), bromides **AG** (1.0 mmol), pyridin-4-ylboronic acids (1.2 mmol), and toluene (3.0 mL), EtOH (2.0 mL) and H₂O (2.0 mL) were added to a sealed tube. The reaction mixture was degassed with nitrogen, and the resulting solution was stirred at 100 °C for 24 h. After the reaction completed (by TLC), the reaction mixture was quenched

with saturated aq. NH_4Cl solution and extracted using ethyl acetate. The organic extracts were combined, dried over anhydrous Na_2SO_4 , and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford biaryl aldehydes **AH** (yield 70-77%). The biaryl aldehydes **AH** were subjected to further transformation to obtain corresponding enone **6p** and **6q**, as mentioned in Scheme S2.

General procedure 4: Synthesis of enone-tethered pyridinium salts (**6j**, **6z**, and **7l**)



Scheme S4: General representation for the synthesis of **6j**, **6z**, and **7l**

A representative procedure for the synthesis of AL (Scheme S4, steps I and II): All the 2-bromo-enones were synthesized according to the reported literature.²

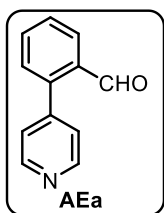
A representative procedure for the synthesis of 6j, 6z, and 7l (Scheme S4, step III): $\text{Pd}(\text{PPh}_3)_4$ (0.005 mmol), $[\text{HP}^t\text{-Bu}_3]\text{BF}_4$ (0.012 mmol), enone (1.0 mmol), pyridin-4-ylboronic acids (1.2 mmol), KF (3.3 mmol) and H_2O (60.0 mmol) were added to a sealed tube. The reaction tube was degassed with nitrogen, DMF (2.0 mL) was added using a syringe, and the resulting solution was stirred at 70 °C for 36 h. After the completion of the starting material, the reaction mixture was quenched with saturated aq. NH_4Cl solution and extracted using ethyl acetate. The organic extracts were combined, dried over anhydrous Na_2SO_4 , and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford enone-tethered pyridines **6j**, **6z**, and enone-tethered quinoline **7l**.

Spectral data of aldehydes and enones reported in this study

2-(Pyridin-4-yl)benzaldehyde (AEa).

This compound was isolated as pale-yellow semi-solid by following the general procedure-2. 1000 mg of **AD** afforded 848 mg of **AEa** (89% yield). $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\text{max}}/\text{cm}^{-1}$ 2854, 1691, 1592, 1541,

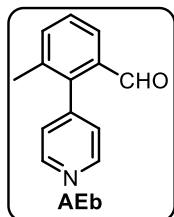
² a) Patel, K.; Mishra, U. K.; Mukhopadhyay, D.; Ramasastry, S. S. V. *Chem. Asian J.* **2019**, *14*, 4568. b) Diemer, V.; Berthelot, A.; Bayardon, J.; Jugé, S.; Leroux, F. R.; Colobert, F. *J. Org. Chem.* **2012**, *77*, 14, 6117.



1474, 1407, 1257, 990, 830, 764, 642, 628. **¹H NMR (500 MHz, CDCl₃):** δ 9.98 (s, 1H), 8.72 (dt, *J* = 4.2, 1.5 Hz, 2H), 8.05 (dd, *J* = 7.8, 1.4 Hz, 1H), 7.69 (dd, *J* = 7.6, 1.3 Hz, 1H), 7.62-7.54 (m, 1H), 7.43 (dd, *J* = 7.8, 1.3 Hz, 1H), 7.33 (dt, *J* = 4.5, 1.3 Hz, 2H). **¹³C NMR (125 MHz, CDCl₃):** δ 191.08, 149.80 (2C), 145.82, 142.67, 133.88, 133.42, 130.36, 129.01, 128.35, 124.72 (2C). **HRMS (ESI):** *m/z* calcd for C₁₂H₁₀NO (M+H)⁺: 184.0762, found: 184.0767.

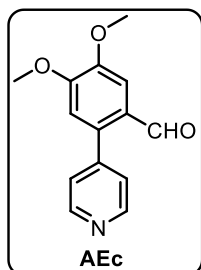
3-Methyl-2-(pyridin-4-yl)benzaldehyde (AEb).

This compound was isolated as pale-yellow thick oil following the general procedure-2. 300 mg of **AD** afforded 246 mg of **AEb** (82% yield). *R_f* = 0.4 (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** *v*_{max}/cm⁻¹ 2929, 1690, 1675, 1593, 1459, 1430, 1383, 1209, 912, 766. **¹H NMR (400 MHz, CDCl₃):** 9.69 (s, 1H), 8.78-8.70 (m, 2H), 7.88 (dd, *J* = 7.7, 1.4 Hz, 1H), 7.55 (d, *J* = 7.6 Hz, 1H), 7.47 (t, *J* = 7.7 Hz, 1H), 7.25-7.19 (m, 2H), 2.13 (s, 3H). **¹³C NMR (100 MHz, CDCl₃):** δ 191.43, 149.91, 145.52, 142.00, 136.56, 135.75, 133.71, 132.15, 132.05, 128.62, 125.64, 124.88, 19.92. **HRMS (ESI):** *m/z* calcd for C₁₃H₁₂NO (M+H)⁺ 198.0919, found: 198.0922.



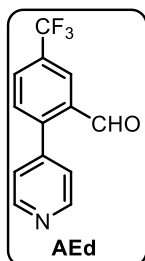
4,5-Dimethoxy-2-(pyridin-4-yl)benzaldehyde (AEc).

This compound was isolated as pale-yellow semisolid by following the general procedure-2. 250mg of **AD** afforded 258 mg of **AEc** (82% yield). *R_f* = 0.3 (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** *v*_{max}/cm⁻¹ 2930, 1659, 1594, 1507, 1438, 1355, 1223, 1094, 1064, 920, 887, 831, 724, 659. **¹H NMR (400 MHz, CDCl₃):** δ 9.82 (s, 1H), 8.78-8.63 (m, 2H), 7.56 (s, 1H), 7.37-7.26 (m, 2H), 6.83 (s, 1H), 4.00 (s, 6H). **¹³C NMR (100 MHz, CDCl₃):** δ 189.91, 153.64, 149.78 (2C), 149.55, 145.61, 138.13, 126.76, 125.00 (2C), 112.05, 109.05, 56.38, 56.24. **HRMS (ESI):** *m/z* calcd for C₁₄H₁₄NO₃ (M+H)⁺: 244.0974, found: 244.0981.



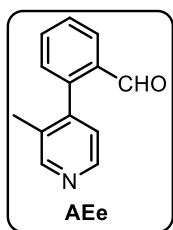
2-(Pyridin-4-yl)-5-(trifluoromethyl)benzaldehyde (AEd).

This compound was isolated as pale-yellow oil by following the general procedure-2. 200 mg of **AD** afforded 210 mg of **AEd** (81% yield), *R_f* = 0.4 (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** *v*_{max}/cm⁻¹ 2851, 1687, 1591, 1573, 1498, 1460, 1239, 957, 807. **¹H NMR (400 MHz, CDCl₃):** δ 9.99 (s, 1H), 8.82-8.73 (m, 2H), 8.38-8.28 (m, 1H), 7.95 (ddd, *J* = 8.1, 1.9, 0.7 Hz, 1H), 7.60 (dt, *J* = 7.9, 0.7 Hz, 1H), 7.39-7.32 (m, 2H). **¹³C NMR (100 MHz, CDCl₃):** δ 189.64, 150.11 (2C), 145.67 (apparent q, *J*_{C-F} = 0.92 Hz), 144.45, 133.74, 131.59 (q, *J*_{C-F} = 33.4 Hz), 131.18, 130.18 (q, *J*_{C-F} = 3.5 Hz), 125.48 (q, *J*_{C-F} = 3.7 Hz), 124.45 (2C), 123.34 (q, *J*_{C-F} = 270.9 Hz). **¹⁹F NMR (376 MHz, CDCl₃):** δ -62.98. **HRMS (ESI):** *m/z* calcd for C₁₃H₉F₃NO (M+H)⁺ 252.0636 found: 252.0654.



2-(3-Methylpyridin-4-yl)benzaldehyde (AEe).

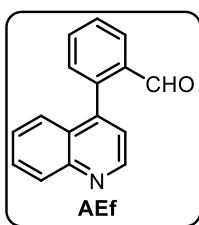
This compound was isolated as pale-yellow semi-solid by following the general procedure-2.



260 mg of **AD** afforded 225 mg of **AEe** (75% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 1693, 1591, 1571, 1544, 1474, 1403, 1361, 1297, 1061, 992, 836, 813. **^1H NMR (500 MHz, CDCl_3):** δ 9.76 (s, 1H), 8.56 (s, 1H), 8.52 (dd, $J = 5.1, 2.2$ Hz, 1H), 8.05 (d, $J = 7.8$ Hz, 1H), 7.71-7.68 (m, 1H), 7.60-7.56 (m, 1H), 7.27 (d, $J = 7.5$ Hz, 1H), 7.15 (dd, $J = 5.0, 1.9$ Hz, 1H), 2.11 (s, 3H). **^{13}C NMR (125 MHz, CDCl_3):** δ 190.98, 150.92, 147.16, 145.91, 142.08, 134.05, 133.18, 131.57, 129.93, 128.80, 128.23, 124.35, 17.01. **HRMS (ESI):** m/z calcd for $\text{C}_{13}\text{H}_{12}\text{NO}$ ($\text{M}+\text{H}$)⁺ 198.0919, found: 198.0923.

2-(Quinolin-4-yl)benzaldehyde (AEf).

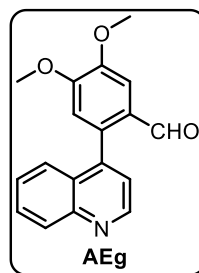
This compound was isolated as yellowish-brown oil by following the general procedure-2.



500 mg of **AD** afforded 439 mg of **AEf** (78% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2085, 1687, 1589, 1564, 1427, 1370, 1245, 1218, 1018, 821, 757. **^1H NMR (400 MHz, CDCl_3):** δ 9.62 (s, 1H), 8.97 (d, $J = 4.3$ Hz, 1H), 8.20 (d, $J = 8.4$ Hz, 1H), 8.10 (dd, $J = 7.7, 0.8$ Hz, 1H), 7.72-7.67 (m, 2H), 7.60 (t, $J = 7.5$ Hz, 1H), 7.50-7.42 (m, 2H), 7.39 (d, $J = 7.3$ Hz, 1H), 7.33 (d, $J = 4.2$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** 190.83, 149.62, 148.15, 144.52, 140.93, 134.22, 133.92, 130.97, 129.97, 129.84, 129.14, 127.88, 127.73, 127.43, 125.54, 122.42. **HRMS (ESI):** m/z calcd for $\text{C}_{16}\text{H}_{12}\text{NO}$ ($\text{M}+\text{H}$)⁺ 234.0919 found 234.0918.

4,5-Dimethoxy-2-(quinolin-4-yl)benzaldehyde (AEg).

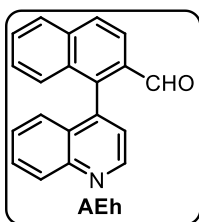
This compound was isolated as pale-yellow sticky oil following general procedure-2. 220 mg



of **AD** afforded 252 mg of **AEg** (81% yield), $R_f = 0.4$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2923, 1712, 1678, 1594, 1566, 1513, 1503, 1462, 1384, 1280, 1218, 1137, 1098, 875, 766. **^1H NMR (400 MHz, CDCl_3):** δ 9.47 (s, 1H), 8.99 (d, $J = 4.3$ Hz, 1H), 8.21 (dt, $J = 8.4, 0.9$ Hz, 1H), 7.76 (ddd, $J = 8.4, 6.6, 1.6$ Hz, 1H), 7.63 (s, 1H), 7.60-7.56 (m, 1H), 7.54-7.51 (m, 1H), 7.38 (d, $J = 4.3$ Hz, 1H), 6.86 (s, 1H), 4.04 (s, 3H), 3.95 (s, 3H). **^{13}C NMR (100 MHz, CDCl_3):** δ 189.6, 153.6, 149.6, 149.5, 148.2, 144.2, 136.1, 129.9, 129.8, 128.1, 127.6, 127.4, 125.6, 122.7, 112.7, 108.5, 56.4, 56.2. **HRMS (ESI):** m/z calcd for $\text{C}_{18}\text{H}_{16}\text{NO}_3$ ($\text{M}+\text{H}$)⁺ 294.1130, found: 294.1139.

1-(Quinolin-4-yl)-2-naphthaldehyde (AEh).

This compound was isolated as pale-yellow semi-solid by following the general procedure-2.

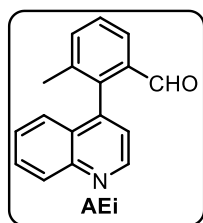


240 mg of **AD** afforded 279 mg of **AEh** (85% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3060, 2863, 1686, 1616, 1589, 1564, 1371, 1331, 1218, 913, 821, 778. **^1H NMR (400 MHz, CDCl_3):** δ 9.67 (d, $J = 0.9$ Hz, 1H), 9.08 (d, $J = 4.3$ Hz, 1H), 8.33 – 8.24 (m, 1H), 8.17 (d, $J = 8.7$ Hz, 1H), 8.08 (d, $J = 8.7$ Hz, 1H),

7.99 (dt, $J = 8.3, 1.0$ Hz, 1H), 7.74 (ddd, $J = 8.4, 6.8, 1.5$ Hz, 1H), 7.62 (ddd, $J = 8.2, 6.7, 1.3$ Hz, 1H), 7.47 (d, $J = 4.3$ Hz, 1H), 7.37 (dtd, $J = 8.3, 6.8, 1.3$ Hz, 2H), 7.27 (ddd, $J = 16.8, 8.5, 1.3$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ 191.1, 149.5, 148.0, 142.5, 141.4, 136.0, 131.9, 131.5, 130.1, 129.9, 129.5, 129.2, 128.5, 128.4, 127.6, 127.5, 127.1, 125.9, 123.6, 122.2. HRMS (ESI): m/z calcd for $\text{C}_{20}\text{H}_{14}\text{NO}$ ($\text{M}+\text{H}^+$) 284.1075 found: 284.1081.

3-Methyl-2-(quinolin-4-yl)benzaldehyde (AEi).

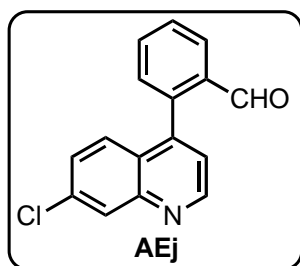
This compound was isolated as yellowish-brown oil by following the general procedure-2.



240 mg of **A** afforded 244 mg of **AEi** (85% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2851, 1687, 1591, 1573, 1498, 1460, 1239, 957, 807. ^1H NMR (400 MHz, CDCl_3): δ 9.48 (s, 1H), 8.97 (dd, $J = 4.2, 1.7$ Hz, 1H), 8.26 – 8.20 (m, 1H), 7.96 (dd, $J = 7.7, 1.5$ Hz, 1H), 7.83 (dd, $J = 8.6, 7.0$ Hz, 1H), 7.67 – 7.58 (m, 2H), 7.54 (t, $J = 7.7$ Hz, 1H), 7.45 (dd, $J = 7.0, 1.2$ Hz, 1H), 7.35 (dd, $J = 8.5, 4.2$ Hz, 1H), 1.98 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 191.95, 150.76, 148.21, 141.93, 138.34, 135.66, 135.21, 134.83, 133.78, 129.88, 128.94, 128.51, 128.33, 127.61, 125.13, 121.79, 19.65. HRMS (ESI): m/z calcd for $\text{C}_{17}\text{H}_{14}\text{NO}$ ($\text{M}+\text{H}^+$) 248.1075 found: 248.1073.

2-(7-Chloroquinolin-4-yl)benzaldehyde (AEj).

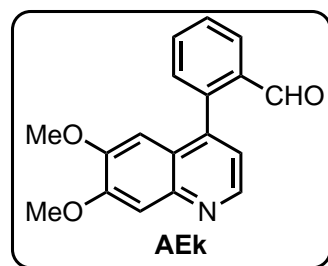
This compound was isolated as yellow solid by following the general procedure-2. 500 mg of



AD afforded 409 mg of **AEj** (88% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). M.P = 110-113 °C. IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3061, 2924, 1696, 1597, 1584, 1499, 1416, 1260, 1197, 1070, 882, 827, 766. ^1H NMR (400 MHz, CDCl_3): δ 9.62 (s, 1H), 8.96 (d, $J = 4.3$ Hz, 1H), 8.17 (d, $J = 1.0$ Hz, 1H), 8.10 (d, $J = 7.7$ Hz, 1H), 7.73 (t, $J = 7.4$ Hz, 1H), 7.65 (t, $J = 7.7$ Hz, 1H), 7.41-7.38 (m, 3H), 7.33 (d, $J = 4.3$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 190.60, 150.71, 148.58, 144.79, 140.19, 135.81, 134.20, 134.02, 130.93, 129.41, 128.91, 128.43, 128.39, 126.94, 126.17, 122.49. HRMS (ESI): m/z calcd for $\text{C}_{16}\text{H}_{11}\text{ClNO}$ ($\text{M}+\text{H}^+$) 268.0529 found 268.0550.

2-(6,7-Dimethoxyquinolin-4-yl)benzaldehyde (AEk).

This compound was isolated as pale-yellow solid by following the general procedure-2. 500

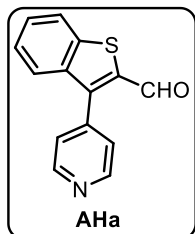


mg of **AD** afforded 424 mg of **AEk** (77% yield), $R_f = 0.4$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). M.P = 141-143 °C. IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3061, 2928, 1694, 1619, 1595, 1494, 1432, 1351, 1293, 1216, 1120, 1090, 861, 766. ^1H NMR (400 MHz, CDCl_3): δ 9.62 (s, 1H), 8.76 (d, $J = 4.4$ Hz, 1H), 8.10 (dd, $J = 7.8, 1.2$ Hz, 1H), 7.72 (t, $J = 7.4$ Hz, 1H), 7.61 (t, $J = 7.8$ Hz, 1H), 7.48 (s, 1H), 7.42 (d, $J = 7.5$ Hz, 1H), 7.18 (d, $J = 4.4$ Hz, 1H), 6.64 (s, 1H), 4.02 (s, 3H), 3.71 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 191.15, 152.58, 150.45, 147.41, 145.50, 142.38, 141.60, 134.06, 130.79, 129.08, 127.66, 123.17,

120.93, 108.41, 102.77, 102.75, 56.23, 55.92. **HRMS (ESI):** m/z calcd for $C_{18}H_{16}NO_3$ ($M+H^+$) 294.1130 found 294.1143.

3-(Pyridin-4-yl)benzo[*b*]thiophene-2-carbaldehyde (AHa).

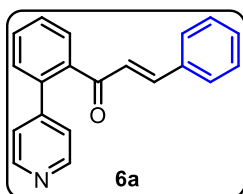
This compound was isolated as reddish-brown sticky oil by following the general procedure-



3. 300 mg of **AG** afforded 284 mg of **AHa** (82% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2845, 2830, 1713, 1660, 1591, 1520, 1410, 1351, 1263, 1161, 989, 857, 764, 733. **1H NMR (500 MHz, $CDCl_3$):** δ 9.94 (s, 1H), 8.87-8.76 (m, 2H), 7.98-7.90 (m, 1H), 7.74 (dd, $J = 8.3, 1.1$ Hz, 1H), 7.56 (ddd, $J = 8.2, 7.1, 1.2$ Hz, 1H), 7.49-7.41 (m, 3H). **^{13}C NMR (125 MHz, $CDCl_3$):** δ 184.72, 150.37 (2C), 143.87, 141.96, 140.47, 139.84, 138.44, 128.76, 125.71, 125.10 (2C), 124.81, 123.45, 123.45, 150.3 (2C), 143.8, 141.9, 140.4, 139.8, 138.4, 128.7, 125.7, 125.1 (2C), 124.8, 123.4. **HRMS (ESI):** m/z calcd for $C_{14}H_{10}NOS$ ($M+H^+$)⁺ 240.0483, found: 240.0479.

(*E*)-3-Phenyl-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (6a).

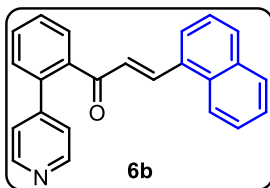
This compound was isolated as pale-yellow sticky oil following general procedure-2. 500 mg



of **AF** afforded 643 mg of **6a** (89% yield), $R_f = 0.4$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2921, 2852, 1667, 1593, 1331, 1209, 1027, 983, 827, 760. **1H NMR (400 MHz, $CDCl_3$):** δ 8.59 (d, $J = 6.1$ Hz, 2H), 7.66 (dd, $J = 7.5, 1.5$ Hz, 1H), 7.60 (td, $J = 7.5, 1.5$ Hz, 1H), 7.53 (td, $J = 7.5, 1.4$ Hz, 1H), 7.45 (dd, $J = 7.5, 1.4$ Hz, 1H), 7.40-7.29 (m, 8H), 6.74 (d, $J = 16.0$ Hz, 1H). **^{13}C NMR (100 MHz, $CDCl_3$):** δ 195.74, 149.91 (2C), 148.32, 145.62, 139.52, 138.19, 134.22, 130.95, 130.83, 130.04, 128.95 (2C), 128.93, 128.65, 128.34 (2C), 126.57, 123.79 (2C). **HRMS (ESI):** m/z calcd for $C_{20}H_{16}NO$ ($M+H^+$)⁺ 286.1232 found 286.1245.

(*E*)-3-(Naphthalen-1-yl)-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (6b).

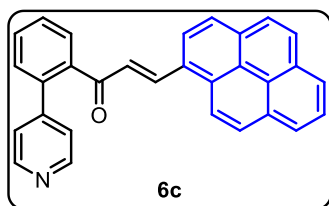
This compound was isolated as pale-yellow oil by following the general procedure-2. 150 mg



of **AF** afforded 204 mg of **6b** (80% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 3068, 1664, 1641, 1595, 1509, 1396, 1347, 1311, 1289, 1214, 1027, 979, 827, 779, 762. **1H NMR (500 MHz, $CDCl_3$):** δ 8.65-8.59 (m, 2H), 8.21 (d, $J = 15.7$ Hz, 1H), 7.91-7.86 (m, 1H), 7.81 (ddd, $J = 8.2, 7.1, 1.9$ Hz, 2H), 7.73 (dd, $J = 7.6, 1.5$ Hz, 1H), 7.60 (td, $J = 7.5, 1.5$ Hz, 1H), 7.55 (td, $J = 7.5, 1.4$ Hz, 1H), 7.51-7.44 (m, 3H), 7.39-7.31 (m, 4H), 6.74 (d, $J = 15.8$ Hz, 1H). **^{13}C NMR (125 MHz, $CDCl_3$):** δ 195.37, 149.92 (2C), 148.48, 142.02, 139.74, 138.19, 133.65, 131.64, 131.44, 131.13, 131.01, 130.06, 129.13, 129.04, 128.83 (2C), 127.01, 126.30, 125.44, 125.33, 124.00, 123.11. **HRMS (ESI):** m/z calcd for $C_{24}H_{18}NO$ ($M+H^+$)⁺ 336.1388 found: 336.1379.

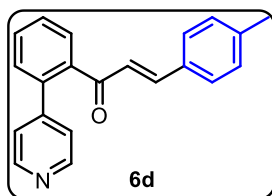
(*E*)-3-(Pyren-1-yl)-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (6c).

This compound was isolated as yellowish-orange solid by following the general procedure-2.



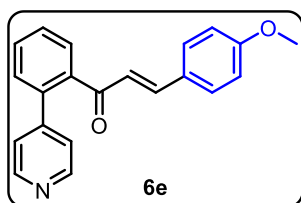
100 mg of **AF** afforded 166 mg of **6c** (80% yield), M.P = 163-167 °C. $R_f = 0.3$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 1659, 1637, 1592, 1580, 1317, 1286, 1214, 1023, 977, 844. **$^1\text{H NMR}$ (500 MHz, CDCl_3):** δ 8.66-8.59 (m, 2H), 8.49 (d, $J = 15.7$ Hz, 1H), 8.15-8.09 (m, 3H), 8.03-7.98 (m, 2H), 7.97-7.88 (m, 3H), 7.78 (td, $J = 6.0, 5.6, 3.0$ Hz, 2H), 7.60 (ddd, $J = 14.7, 7.4, 1.5$ Hz, 2H), 7.47 (dd, $J = 7.5, 1.5$ Hz, 1H), 7.40-7.34 (m, 2H), 6.88 (d, $J = 15.7$ Hz, 1H). **$^{13}\text{C NMR}$ (125 MHz, CDCl_3):** δ 195.16, 149.92 (2C), 148.59, 141.69, 139.96, 138.28, 132.97, 131.20, 131.11, 130.54, 130.11, 130.06, 129.18, 128.84, 128.81, 128.72, 128.28, 127.99, 127.25, 126.30, 126.16, 125.94, 125.02, 124.76, 124.44, 124.15, 124.03 (2C), 122.11. **HRMS (ESI):** m/z calcd for $\text{C}_{30}\text{H}_{20}\text{NO}$ ($\text{M}+\text{H}$)⁺ 410.1545, found: 410.1555.

(*E*)-1-(2-(Pyridin-4-yl)phenyl)-3-(*p*-tolyl)prop-2-en-1-one (**6d**).



This compound was isolated as pale-yellow oil by following the general procedure-2. 100 mg of **AF** afforded 137 mg of **6d** (90% yield), $R_f = 0.5$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2921, 2851, 1666, 1642, 1595, 1408, 1326, 1208, 1024, 983, 826, 760. **$^1\text{H NMR}$ (400 MHz, CDCl_3):** δ 8.62-8.52 (m, 2H), 7.64 (dd, $J = 7.5, 1.5$ Hz, 1H), 7.58 (dd, $J = 7.5, 1.6$ Hz, 1H), 7.53 (dd, $J = 7.5, 1.4$ Hz, 1H), 7.45 (dd, $J = 7.6, 1.4$ Hz, 1H), 7.34 (d, $J = 16.1$ Hz, 1H), 7.31-7.27 (m, 2H), 7.23 (d, $J = 7.9$ Hz, 2H), 7.13 (d, $J = 7.9$ Hz, 2H), 6.69 (d, $J = 16.0$ Hz, 1H), 2.33 (s, 3H). **$^{13}\text{C NMR}$ (101 MHz, CDCl_3):** δ 195.89, 149.85 (2C), 148.38, 145.86, 141.47, 139.63, 138.13, 131.47, 130.85, 130.02, 129.72 (2C), 128.90, 128.62, 128.39 (2C), 125.67, 123.80 (2C), 21.54. **HRMS (ESI):** m/z calcd for $\text{C}_{21}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$)⁺ 300.1388 found 300.1386.

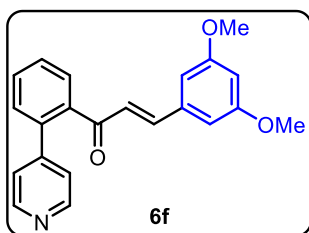
(*E*)-3-(4-Methoxyphenyl)-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (**6e**).



This compound was isolated as pale-yellow sticky oil following general procedure-2. 150 mg of **AF** afforded 227 mg of **6e** (95% yield), $R_f = 0.3$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3054, 3025, 2839, 1663, 1636, 1592, 1569, 1251, 1172, 1025, 828, 775. **$^1\text{H NMR}$ (400 MHz, CDCl_3):** δ 8.57-8.56 (m, 2H), 7.63 (dd, $J = 7.4, 1.1$ Hz, 1H), 7.60-7.49 (m, 1H), 7.53-7.49 (m, 1H), 7.44 (d, $J = 7.4$ Hz, 1H), 7.34-7.28 (m, 5H), 6.83 (d, $J = 8.6$ Hz, 2H), 6.61 (d, $J = 16.0$ Hz, 1H), 3.79 (s, 3H). **$^{13}\text{C NMR}$ (100 MHz, CDCl_3):** δ 195.85, 161.89, 149.82 (2C), 148.44, 145.69, 139.78, 138.07, 130.73, 130.18 (2C), 129.99, 128.85, 128.60, 126.88, 124.44, 123.80 (2C), 114.44 (2C), 55.40. **HRMS (ESI):** m/z calcd for $\text{C}_{21}\text{H}_{18}\text{NO}_2$ ($\text{M}+\text{H}$)⁺ 316.1338, found: 316.1348.

(*E*)-3-(3,5-Dimethoxyphenyl)-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (**6f**).

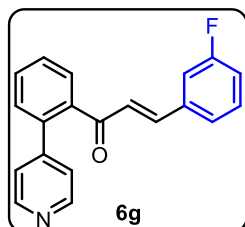
This compound was isolated as pale-yellow sticky oil following general procedure-2. 120 mg



of **AF** afforded 170 mg of **6f** (81% yield), $R_f = 0.3$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2923, 2839, 1666, 1641, 1589, 1542, 1456, 1426, 1284, 1203, 1154, 1064, 989, 828. **^1H NMR (400 MHz, CDCl_3):** δ 8.59-8.51 (m, 2H), 7.63 (dd, $J = 7.6, 1.5$ Hz, 1H), 7.56 (td, $J = 7.5, 1.6$ Hz, 1H), 7.50 (td, $J = 7.5, 1.5$ Hz, 1H), 7.41 (dd, $J = 7.6, 1.4$ Hz, 1H), 7.31-7.20 (m, 3H), 6.63 (d, $J = 16.0$ Hz, 1H), 6.42 (s, 3H), 3.71 (d, $J = 1.6$ Hz, 6H). **^{13}C NMR (100 MHz, CDCl_3):** δ 195.47, 160.94 (2C), 149.74 (2C), 148.44, 145.47, 139.38, 138.18, 136.07, 131.02, 130.05, 128.99, 128.67, 126.95, 123.86 (2C), 106.10 (2C), 103.08, 55.38, 55.35. **HRMS (ESI):** m/z calcd for $\text{C}_{22}\text{H}_{20}\text{NO}_3$ ($\text{M}+\text{H}^+$) 346.1443 found 346.1449.

(*E*)-3-(3-Fluorophenyl)-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (**6g**).

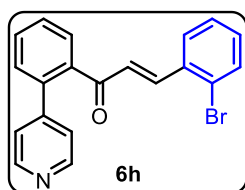
This compound was isolated as pale-yellow oil by following the general procedure-2. 140 mg



of **AF** afforded 172 mg of **6g** (80% yield), $R_f = 0.4$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2924, 2853, 1667, 1639, 1593, 1406, 1321, 1209, 1008, 981, 824, 762. **^1H NMR (400 MHz, CDCl_3):** δ 8.59 (m, 2H), 7.66 (d, $J = 7.5, 1\text{H}$), 7.62 (t, $J = 7.5$ Hz, 1H), 7.54 (t, $J = 7.4$ Hz, 1H), 7.46 (d, $J = 7.5$ Hz, 1H), 7.33-7.28 (m, 4H), 7.09 (d, $J = 7.6$ Hz, 1H), 7.06-6.99 (m, 2H), 6.67 (d, $J = 16$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 195.34, 162.92 (d, $J_{\text{C-F}} = 245.6$ Hz), 149.97 (2C), 148.26, 143.74 (d, $J_{\text{C-F}} = 2.8$ Hz), 139.32, 138.25, 136.47 (d, $J_{\text{C-F}} = 7.6$ Hz), 131.16, 130.50 (d, $J_{\text{C-F}} = 8.1$ Hz), 130.06, 128.99, 128.74, 127.53, 124.26 (d, $J_{\text{C-F}} = 2.8$ Hz), 123.79 (2C), 117.68 (d, $J_{\text{C-F}} = 21.3$ Hz), 114.47 (d, $J_{\text{C-F}} = 21.9$ Hz). **^{19}F NMR (376 MHz, CDCl_3):** δ -112.26. **HRMS (ESI):** m/z calcd for $\text{C}_{20}\text{H}_{15}\text{FNO}$ ($\text{M}+\text{H}^+$) 304.1138 found 304.1187.

(*E*)-3-(2-Bromophenyl)-1-(2-(pyridin-4-yl)phenyl)prop-2-en-1-one (**6h**).

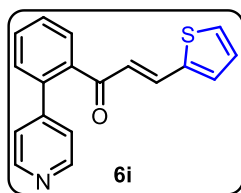
This compound was isolated as pale-yellow oil by following the general procedure-2. 125 mg



of **AF** afforded 195 mg of **6h** (85% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 1666, 1644, 1597, 1465, 1440, 1408, 1207, 1025, 828, 760. **^1H NMR (500 MHz, CDCl_3):** δ 8.64-8.58 (m, 2H), 7.71-7.64 (m, 2H), 7.61 (td, $J = 7.6, 1.5$ Hz, 1H), 7.54 (tt, $J = 7.8, 1.2$ Hz, 2H), 7.47 (dd, $J = 7.6, 1.3$ Hz, 1H), 7.33-7.29 (m, 2H), 7.25-7.21 (m, 2H), 7.19-7.14 (m, 1H), 6.59 (d, $J = 16.0$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 195.78, 149.92 (2C), 148.27, 143.82, 139.22, 138.17, 134.27, 133.43, 131.58, 131.09, 129.95, 129.15, 129.07, 128.73, 127.77, 127.71, 125.69, 123.96 (2C). **HRMS (ESI):** m/z calcd for $\text{C}_{20}\text{H}_{15}\text{BrNO}$ ($\text{M}+\text{H}^+$) 364.0337, found: 364.0351.

(*E*)-1-(2-(Pyridin-4-yl)phenyl)-3-(thiophen-2-yl)prop-2-en-1-one (**6i**).

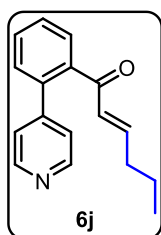
This compound was isolated as reddish-brown oil by following the general procedure-2. 150 mg of **AF** afforded 195 mg of **6i** (88% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by



254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3105, 3063, 3026, 1658, 1636, 1583, 1474, 1409, 1363, 1282, 1209, 1078, 1023, 827, 761. **^1H NMR (500 MHz, CDCl_3):** δ 8.63-8.56 (m, 2H), 7.68-7.63 (m, 1H), 7.59 (td, $J = 7.6, 1.5$ Hz, 1H), 7.53 (dd, $J = 7.5, 1.3$ Hz, 1H), 7.52-7.48 (m, 1H), 7.45-7.42 (m, 1H), 7.35 (dt, $J = 5.0, 1.0$ Hz, 1H), 7.30-7.27 (m, 2H), 7.18-7.15 (m, 1H), 7.00 (dd, $J = 5.1, 3.6$ Hz, 1H), 6.51 (d, $J = 15.7$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 194.82, 149.78 (2C), 148.43, 139.66, 139.48, 138.22, 137.55, 132.04, 130.95, 130.06, 129.57, 128.90, 128.66, 128.37, 125.28, 123.83 (2C). **HRMS (ESI):** m/z calcd for $\text{C}_{18}\text{H}_{14}\text{NOS}$ ($\text{M}+\text{H}$)⁺ 292.0796, found: 292.0789.

(E)-1-(2-(Pyridin-4-yl)phenyl)hex-2-en-1-one (6j).

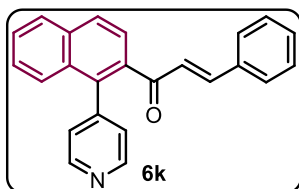
This compound was isolated as pale-yellow oil by following the general procedure-4. 150 mg



of **AL** afforded 155 mg of **6j** (81% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3064, 2959, 2871, 1650, 1617, 1594, 1209, 1118, 970, 804, 762. **^1H NMR (400 MHz, CDCl_3):** δ 8.61-8.57 (m, 2H), 7.56-7.49 (m, 3H), 7.41 (dd, $J = 7.6, 1.3$ Hz, 1H), 7.25-7.22 (m, 2H), 6.54 (dt, $J = 15.8, 6.9$ Hz, 1H), 6.07 (dt, $J = 15.7, 1.5$ Hz, 1H), 2.02 (qd, $J = 7.2, 1.5$ Hz, 2H), 1.31-1.23 (m, 2H), 0.78 (t, $J = 7.4$ Hz, 3H). **^{13}C NMR (100 MHz, CDCl_3):** δ 196.70, 151.59, 149.83, 148.34 (2C), 139.51, 137.81, 130.85, 130.60, 129.74, 128.76, 128.55, 123.80, 123.75, 34.51, 21.08, 13.59. **HRMS (ESI):** m/z calcd for $\text{C}_{17}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$)⁺ 252.1388 found: 252.1385.

(E)-3-Phenyl-1-(1-(pyridin-4-yl)naphthalen-2-yl)prop-2-en-1-one (6k).

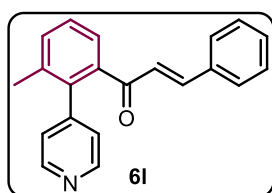
This compound was isolated as pale-yellow oil by following the general procedure-2. 120 mg



of **AF** afforded 163 mg of **6k** (80% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 1658, 1642, 1598, 1340, 1207, 1051, 978, 824, 766, 750. **^1H NMR (400 MHz, CDCl_3):** δ 8.71-8.68 (m, 2H), 8.04 (d, $J = 8.5$ Hz, 1H), 8.00 (dd, $J = 8.1, 1.4$ Hz, 1H), 7.71 (d, $J = 8.5$ Hz, 1H), 7.67-7.61 (m, 2H), 7.56-7.52 (m, 1H), 7.38-7.34 (m, 7H), 7.30 (d, $J = 9.9$ Hz, 1H), 6.75 (d, $J = 16.1$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 196.28, 149.57 (2C), 146.04, 137.05, 135.47, 134.23, 134.21, 131.13, 130.85, 128.97 (2C), 128.89, 128.62, 128.38 (2C), 128.36 (2C), 127.46, 127.41, 126.80, 126.39, 125.77, 124.58. **HRMS (ESI):** m/z calcd for $\text{C}_{24}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$)⁺ 336.1388 found 336.1375.

(E)-1-(3-Methyl-2-(pyridin-4-yl)phenyl)-3-phenylprop-2-en-1-one (6l).

This compound was isolated as pale-yellow oil by following the general procedure-2. 100 mg

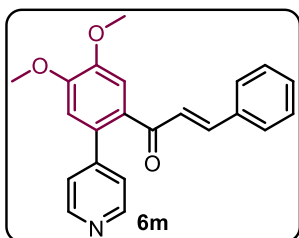


of **AF** afforded 138 mg of **6l** (91% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3062, 2931, 1668, 1643, 1599, 1574, 1450, 1331, 1286, 1232, 1134, 1083, 1054, 981, 764. **^1H NMR (500 MHz, CDCl_3):** δ 8.61-8.57 (m, 2H), 7.47-7.40 (m, 3H), 7.38-7.33 (m, 5H), 7.29 (d, $J = 16.1$ Hz, 1H), 7.18-7.14 (m, 2H), 6.70 (d, $J = 16.0$ Hz, 1H), 2.18 (s, 3H). **^{13}C NMR (125**

MHz, CDCl₃): δ 195.93, 149.60 (2C), 147.77, 145.74, 140.02, 137.17, 136.30, 134.32, 132.48, 130.74, 128.94 (2C), 128.33 (2C), 128.08, 126.77, 125.79, 124.73(2C), 20.44. **HRMS (ESI):** m/z calcd for C₂₁H₁₈NO (M+H)⁺ 300.1388 found: 300.1403.

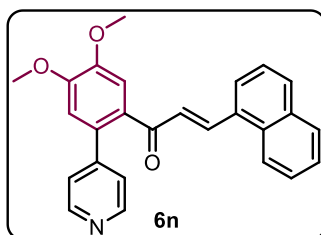
(E)-1-(4,5-Dimethoxy-2-(pyridin-4-yl)phenyl)-3-phenylprop-2-en-1-one (6m).

This compound was isolated as an off-white solid by following the general procedure-2. 150 mg of **AF** afforded 231 mg of **6m** (88% yield), M.P = 155-158 °C, R_f = 0.3 (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2935, 2849, 1660, 1637, 1596, 1351, 1244, 1156, 1029, 989, 831. **¹H NMR (400 MHz, CDCl₃):** δ 8.64-8.56 (m, 2H), 7.41 (d, *J* = 15.9 Hz, 1H), 7.38-7.27 (m, 6H), 7.26-7.21 (m, 2H), 6.91 (s, 1H), 6.56 (d, *J* = 15.9 Hz, 1H), 4.00 (s, 3H), 3.99 (s, 3H). **¹³C NMR (100 MHz, CDCl₃):** δ 194.15, 151.10, 149.92 (2C), 149.12, 148.53, 144.09, 134.40, 132.19, 132.13, 130.59, 128.90 (2C), 128.20 (2C), 126.45, 124.02 (2C), 112.41, 112.19, 56.25 (2C). **HRMS (ESI):** m/z calcd for C₂₂H₂₀NO₃ (M+H)⁺ 346.1443 found: 346.1426.



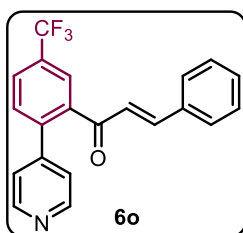
(E)-1-(4,5-Dimethoxy-2-(pyridin-4-yl)phenyl)-3-(naphthalen-1-yl)prop-2-en-1-one (6n).

This compound was isolated as a reddish-orange solid by following the general procedure-2. 150 mg of **AF** afforded 246 mg of **6n** (82% yield), M.P = 95-96 °C. R_f = 0.3 (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3058, 3009, 2930, 1709, 1659, 1595, 1542, 1516, 1462, 1439, 1413, 1395, 1350, 1322, 1272, 1243, 1218, 1203, 1154, 1026, 788. **¹H NMR (400 MHz, CDCl₃):** 8.68 – 8.61 (m, 2H), 8.27 (d, *J* = 15.6 Hz, 1H), 7.99 – 7.93 (m, 1H), 7.87 – 7.81 (m, 2H), 7.52 (ddd, *J* = 7.2, 4.9, 1.6 Hz, 2H), 7.41 – 7.34 (m, 4H), 7.16 (dd, *J* = 7.2, 1.1 Hz, 1H), 6.93 (s, 1H), 6.62 (d, *J* = 15.6 Hz, 1H), 4.02 (s, 3H), 4.01 (s, 3H). **¹³C NMR (100 MHz, CDCl₃):** δ 193.91, 151.21, 150.01 (2C), 149.21, 148.57, 140.65, 133.60, 132.35, 132.18, 131.84, 131.45, 130.78, 129.04, 128.77, 126.93, 126.25, 125.44, 125.18, 124.20 (2C), 123.20, 112.40, 112.31, 56.27, 56.23. **HRMS (ESI):** m/z calcd for C₂₆H₂₂NO₃ (M+H)⁺ 396.1600 found 396.1605.



(E)-3-Phenyl-1-(2-(pyridin-4-yl)-5-(trifluoromethyl)phenyl)prop-2-en-1-one (6o).

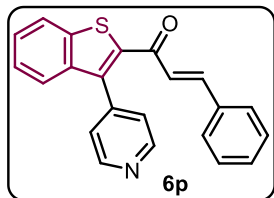
This compound was isolated as pale-yellow liquid by following the general procedure-2. 120 mg of **AF** afforded 174 mg of **6o** (81% yield), R_f = 0.3 (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3061, 2957, 2873, 1650, 1614, 1595, 1108, 973, 801, 757. **¹H NMR (400 MHz, CDCl₃):** δ 8.70-8.62 (m, 2H), 7.93 (d, *J* = 1.8 Hz, 1H), 7.88 (dd, *J* = 8.0, 1.9 Hz, 1H), 7.62 (d, *J* = 8.0 Hz, 1H), 7.43-7.29 (m, 8H), 6.71 (d, *J* = 16.1 Hz, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 194.19, 150.18 (2C), 146.91, 146.55, 141.47, 140.15, 133.87, 131.21, 130.88, 130.62, 129.04 (2C), 128.48 (2C), 127.50 (apparent q, *J*_{C-F} = 3.6 Hz), 125.86 (apparent q, *J*_{C-F} = 3.7 Hz),



125.77, 123.54 (2C), 123.50 (q, $J_{C-F} = 270.9$ Hz). ^{19}F NMR (376 MHz, CDCl_3): δ -62.69. HRMS (ESI): m/z calcd for $\text{C}_{21}\text{H}_{15}\text{F}_3\text{NO}$ ($\text{M}+\text{H}$) $^+$ 354.1106 found: 354.1120.

(E)-3-Phenyl-1-(3-(pyridin-4-yl)benzo[b]thiophen-2-yl)prop-2-en-1-one (6p).

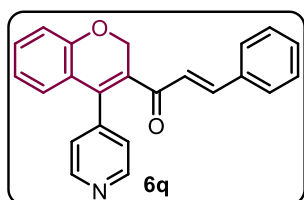
This compound was isolated as reddish-brown solid by following the general procedure-3.



100 mg of **AI** afforded 138 mg of **6p** (80% yield), M.P. = 149.6-151.2 °C. $R_f = 0.4$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 1640, 1594, 1573, 1522, 1483, 1357, 1210, 1179, 991, 974, 762. ^1H NMR (500 MHz, CDCl_3): δ 8.84-8.78 (m, 2H), 7.97-7.91 (m, 1H), 7.68 (d, $J = 15.6$ Hz, 1H), 7.52 (td, $J = 8.0, 1.0$ Hz, 2H), 7.46-7.39 (m, 3H), 7.38-7.30 (m, 3H), 7.23-7.15 (m, 2H), 6.72 (d, $J = 15.5$ Hz, 1H). ^{13}C NMR (125 MHz, CDCl_3): δ 184.52, 150.26 (2C), 144.24, 143.68, 141.59, 140.83, 139.52, 137.93, 134.32, 130.85, 129.01 (2C), 128.39 (2C), 127.69, 125.42, 125.08 (2C), 124.82, 123.50, 122.89. HRMS (ESI): m/z calcd for $\text{C}_{22}\text{H}_{16}\text{NOS}$ ($\text{M}+\text{H}$) $^+$ 342.0953, found: 342.0941.

(E)-3-Phenyl-1-(4-(pyridin-4-yl)-2H-chromen-3-yl)prop-2-en-1-one (6q).

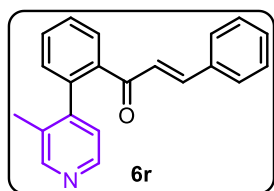
This compound was isolated as pale-yellow solid by following the general procedure-3.



150 mg of **AI** afforded 125 mg of **6q** (83% yield), M.P. = 87-90 °C. $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3036, 1645, 1591, 1574, 1481, 1449, 1373, 1277, 1239, 1204, 1115, 988, 823, 762. ^1H NMR (500 MHz, CDCl_3): δ 8.74-8.65 (m, 2H), 7.45 (d, $J = 15.7$ Hz, 1H), 7.32-7.25 (m, 6H), 7.08 (dt, $J = 6.8, 1.6$ Hz, 2H), 6.99 (dd, $J = 8.1, 1.2$ Hz, 1H), 6.90 (td, $J = 7.6, 1.2$ Hz, 1H), 6.75 (dd, $J = 7.8, 1.6$ Hz, 1H), 6.20 (d, $J = 15.8$ Hz, 1H), 5.07 (s, 2H). ^{13}C NMR (125 MHz, CDCl_3): δ 190.76, 155.55, 150.08 (2C), 144.93, 143.34, 138.95, 134.29, 131.99, 130.65, 129.75, 128.93 (2C), 128.18 (2C), 127.77, 124.90, 124.86 (2C), 122.93, 121.95, 116.76, 66.40. HRMS (ESI): m/z calcd for $\text{C}_{23}\text{H}_{18}\text{NO}_2$ ($\text{M}+\text{H}$) $^+$ 340.1338 found 340.1333.

(E)-1-(2-(3-Methylpyridin-4-yl)phenyl)-3-phenylprop-2-en-1-one (6r).

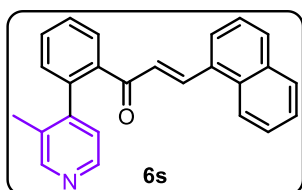
This compound was isolated as pale-yellow oil by following the general procedure-2.



150 mg of **AF** afforded 187 mg of **6r** (82% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3062, 2931, 1666, 1638, 1599, 1574, 1454, 1331, 1288, 1232, 1134, 1092, 1054, 983, 764. ^1H NMR (500 MHz, CDCl_3): δ 8.45 (d, $J = 1.8$ Hz, 1H), 8.42 (dd, $J = 5.0, 1.8$ Hz, 1H), 7.77-7.74 (m, 1H), 7.60-7.56 (m, 1H), 7.53 (tt, $J = 7.5, 1.7$ Hz, 1H), 7.42 (dd, $J = 16.0, 1.9$ Hz, 1H), 7.36-7.32 (m, 5H), 7.26 (dd, $J = 7.8, 1.6$ Hz, 1H), 7.09 (dd, $J = 5.0, 1.8$ Hz, 1H), 6.79 (dd, $J = 15.9, 2.0$ Hz, 1H), 2.14 (s, 3H). ^{13}C NMR (125 MHz, CDCl_3): δ 194.10, 150.84, 148.63, 146.96, 145.11, 139.12, 138.03, 134.33, 131.22, 130.96, 130.74, 130.03, 128.96 (2C), 128.77, 128.33 (2C), 128.22, 125.49, 123.99, 17.02. HRMS (ESI): m/z calcd for $\text{C}_{21}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$) $^+$

300.1388 found 300.1399. **(E)-1-(2-(3-Methylpyridin-4-yl)phenyl)-3-(naphthalen-1-yl)prop-2-en-1-one (6s)**.

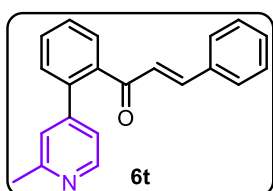
This compound was isolated as yellowish-brown oil by following the general procedure-2.



120 mg of **AF** afforded 189 mg of **6s** (89% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2921, 2851, 1666, 1642, 1595, 1408, 1326, 1208, 1024, 983, 826, 760. **$^1\text{H NMR}$ (500 MHz, CDCl_3):** δ 8.48 (s, 1H), 8.46 (d, $J = 5.0$ Hz, 1H), 8.30 (d, $J = 15.6$ Hz, 1H), 8.01-7.97 (m, 1H), 7.87-7.81 (m, 3H), 7.60 (td, $J = 7.5, 1.5$ Hz, 1H), 7.57-7.53 (m, 1H), 7.50 (td, $J = 7.6, 1.5$ Hz, 2H), 7.43-7.36 (m, 2H), 7.28 (dd, $J = 7.5, 1.4$ Hz, 1H), 7.13 (d, $J = 4.9$ Hz, 1H), 6.85 (d, $J = 15.6$ Hz, 1H), 2.15 (s, 3H). **$^{13}\text{C NMR}$ (125 MHz, CDCl_3):** δ 193.73, 151.05, 148.66, 147.18, 141.65, 139.25, 138.15, 133.68, 131.72, 131.54, 131.29, 131.15, 130.99, 130.13, 128.95, 128.82, 128.33, 127.79, 127.02, 126.30, 125.44, 125.24, 124.08, 123.17, 17.06. **HRMS (ESI):** m/z calcd for $\text{C}_{25}\text{H}_{20}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 350.1545 found 350.1529.

(E)-1-(2-(2-Methylpyridin-4-yl)phenyl)-3-phenylprop-2-en-1-one (6t).

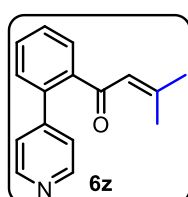
This compound was isolated as pale-yellow oil by following the general procedure-2. 120



mg of **AF** afforded 136 mg of **6t** (80% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 1669, 1643, 1603, 1573, 1545, 1494, 1448, 1331, 1261, 1209, 763, 752. **$^1\text{H NMR}$ (400 MHz, CDCl_3):** δ 8.47 (d, $J = 5.1$ Hz, 1H), 7.66 (dd, $J = 7.5, 1.5$ Hz, 1H), 7.60 (td, $J = 7.6, 1.5$ Hz, 1H), 7.53 (td, $J = 7.5, 1.4$ Hz, 1H), 7.45 (dd, $J = 7.6, 1.4$ Hz, 1H), 7.39-7.32 (m, 6H), 7.17 (d, $J = 1.7$ Hz, 1H), 7.11 (dd, $J = 5.1, 1.7$ Hz, 1H), 6.71 (d, $J = 16.0$ Hz, 1H), 2.54 (s, 3H). **$^{13}\text{C NMR}$ (100 MHz, CDCl_3):** δ 195.89, 158.64, 149.20, 148.69, 145.43, 139.49, 138.49, 134.29, 130.89, 130.77, 129.96, 128.94 (2C), 128.90, 128.51, 128.31 (2C), 126.63, 123.39, 120.96, 24.45. **HRMS (ESI):** m/z calcd for $\text{C}_{21}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 300.1388 found 300.1376.

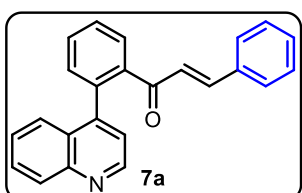
3-Methyl-1-(2-(pyridin-4-yl)phenyl)but-2-en-1-one (6z).

This compound was isolated as yellowish-red oil by following the general procedure-4. 160



mg of **AL** afforded 150 mg of **6z** (78% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2925, 1665, 1608, 1583, 1437, 1409, 1225, 1101, 831, 750. **$^1\text{H NMR}$ (400 MHz, CDCl_3):** δ 8.63 (d, $J = 5.1$ Hz, 2H), 7.63 (dd, $J = 7.4, 1.6$ Hz, 1H), 7.58-7.48 (m, 2H), 7.39 (dd, $J = 7.3, 1.5$ Hz, 1H), 7.28 (d, $J = 4.2$ Hz, 2H), 6.03 (s 1H), 2.09 (s 3H), 1.74 (s 3H). **$^{13}\text{C NMR}$ (100 MHz, CDCl_3):** δ 195.17, 156.97, 149.51 (2C), 148.86, 141.54, 137.76, 130.58, 129.90, 128.65, 128.50 (2C), 124.95, 123.94, 27.67, 20.92. **HRMS (ESI):** m/z calcd for $\text{C}_{16}\text{H}_{16}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 238.1232 found: 238.1231.

(E)-3-Phenyl-1-(2-(quinolin-4-yl)phenyl)prop-2-en-1-one (7a).

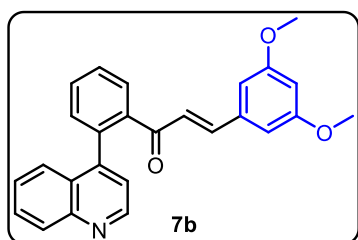


This compound was isolated as yellowish-brown semi-solid by following the general procedure-2. 200 mg of **AF** afforded 250 mg

of **7a** (92% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 1667, 1642, 1604, 1331, 1277, 1209, 1017, 981, 959, 764, 750. **^1H NMR (400 MHz, CDCl_3):** δ 8.87 (d, $J = 4.3$ Hz, 1H), 8.13 (dd, $J = 8.6, 1.3$ Hz, 1H), 7.85-7.81 (m, 1H), 7.76 (dd, $J = 8.4, 1.4$ Hz, 1H), 7.69 (ddd, $J = 8.4, 6.8, 1.5$ Hz, 1H), 7.62 (td, $J = 6.8, 1.7$ Hz, 2H), 7.50 (ddd, $J = 8.3, 6.9, 1.3$ Hz, 1H), 7.47-7.44 (m, 1H), 7.30-7.24 (m, 3H), 7.23-7.17 (m, 2H), 7.05-7.00 (m, 2H), 6.59 (d, $J = 16.0$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 194.40, 149.70, 148.34, 147.32, 144.76, 140.32, 136.61, 134.09, 130.98, 130.78, 130.54, 129.91, 129.52, 129.01, 128.76 (2C), 128.73, 128.14 (2C), 127.07, 127.04, 125.68, 125.51, 122.01. **HRMS (ESI):** m/z calcd for $\text{C}_{24}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 336.1388 found 336.1376.

(E)-3-(3,5-Dimethoxyphenyl)-1-(2-(quinolin-4-yl)phenyl)prop-2-en-1-one (7b).

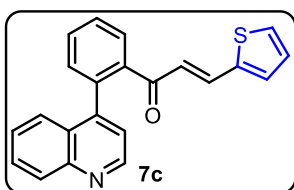
This compound was isolated as pale-yellow solid by following the general procedure-2. 150 mg of **AF** afforded 210 mg of **7b** (88% yield), M.P = 113.4-116 °C. $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3060, 2933, 2838,



1667, 1642, 1586, 1507, 1457, 1425, 1284, 1203, 1154, 1061, 1020, 979, 925, 843, 767, 734. **^1H NMR (400 MHz, CDCl_3):** δ 8.88 (d, $J = 4.4$ Hz, 1H), 8.17-8.09 (m, 1H), 7.86-7.80 (m, 1H), 7.79 (dd, $J = 8.4, 1.4$ Hz, 1H), 7.70 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.66-7.60 (m, 2H), 7.55-7.43 (m, 2H), 7.28 (d, $J = 4.9$ Hz, 1H), 7.21 (d, $J = 15.8$ Hz, 1H), 6.50 (d, $J = 15.8$ Hz, 1H), 6.38 (t, $J = 2.2$ Hz, 1H), 6.17 (d, $J = 2.2$ Hz, 2H), 3.65 (d, $J = 1.2$ Hz, 6H). **^{13}C NMR (100 MHz, CDCl_3):** δ 194.23, 160.78 (2C), 149.74, 148.40, 147.18, 144.60, 140.29, 136.52, 135.97, 131.05, 130.81, 129.98, 129.53, 129.17, 128.76, 127.03, 126.97, 125.82, 125.70, 122.16, 105.80 (2C), 103.15, 77.45, 77.13, 76.81, 55.34, 55.31. **HRMS (ESI):** m/z calcd for $\text{C}_{26}\text{H}_{22}\text{NO}_3$ ($\text{M}+\text{H}$) $^+$ 396.1600 found 396.1597.

(E)-1-(2-(Quinolin-4-yl)phenyl)-3-(thiophen-2-yl)prop-2-en-1-one (7c).

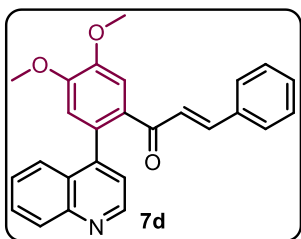
This compound was isolated as reddish-brown oil by following the general procedure-2. 180



mg of **AF** afforded 194 mg of **7c** (78% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2954, 2924, 2853, 1660, 1639, 1582, 1508, 1420, 1387, 1364, 1280, 1209, 1018, 969, 853, 766, 709. **^1H NMR (400 MHz, CDCl_3):** δ 8.89 (d, $J = 4.4$ Hz, 1H), 8.17-8.09 (m, 1H), 7.86-7.80 (m, 1H), 7.75-7.67 (m, 2H), 7.62 (td, $J = 6.4, 1.3$ Hz, 2H), 7.50 (ddd, $J = 8.5, 7.0, 1.4$ Hz, 1H), 7.47-7.43 (m, 1H), 7.39 (d, $J = 15.6$ Hz, 1H), 7.30-7.26 (m, 1H), 7.24 (d, $J = 5.1$ Hz, 1H), 6.96 (d, $J = 3.6$ Hz, 1H), 6.90 (ddd, $J = 4.9, 3.6, 1.1$ Hz, 1H), 6.40 (d, $J = 15.5$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 193.5, 149.7 (2C), 148.3, 147.2, 140.2, 139.5, 136.8, 136.6, 131.8, 131.0, 130.8, 129.9, 129.5, 129.2, 128.9, 128.7, 128.1, 127.0, 125.6, 124.1, 121.9. **HRMS (ESI):** m/z calcd for $\text{C}_{22}\text{H}_{16}\text{NOS}$ ($\text{M}+\text{H}$) $^+$ 342.0953 found 342.0949.

(E)-1-(4,5-Dimethoxy-2-(quinolin-4-yl)phenyl)-3-phenylprop-2-en-1-one (7d).

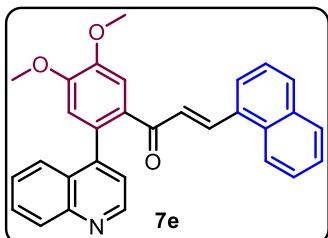
This compound was isolated as pale-yellow solid by following the general procedure-2. 150 mg of **AF** afforded 170 mg of **7d** (88% yield), M.P = 158-161 °C. $R_f = 0.3$ (4:6 EtOAc:



Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3073, 2959, 2929, 2851, 1705, 1658, 1594, 1573, 1514, 1464, 1440, 1346, 1263, 1248, 1199, 1145, 1027, 991, 805. **^1H NMR (400 MHz, CDCl_3):** δ 8.91 (d, $J = 4.4$ Hz, 1H), 8.16 (dd, $J = 8.6, 1.3$ Hz, 1H), 7.82 (dd, $J = 8.5, 1.4$ Hz, 1H), 7.74 (ddd, $J = 8.3, 6.8, 1.4$ Hz, 1H), 7.56 (ddd, $J = 8.3, 6.8, 1.4$ Hz, 1H), 7.45 (s, 1H), 7.32 (d, $J = 4.4$ Hz, 1H), 7.27 – 7.20 (m, 2H), 7.20 – 7.13 (m, 2H), 6.92 (s, 1H), 6.86 (dd, $J = 7.1, 1.8$ Hz, 2H), 6.44 (d, $J = 15.8$ Hz, 1H), 4.05 (s, 3H), 3.95 (s, 3H). **^{13}C NMR (100 MHz, CDCl_3):** δ 192.75, 151.09, 149.65, 149.06, 148.23, 147.79, 143.38, 134.23, 132.93, 130.53, 130.29, 129.77, 129.68, 128.68, 127.95, 127.34, 127.30, 125.70, 125.35, 122.32, 113.26, 112.08, 56.28. **HRMS (ESI):** m/z calcd for $\text{C}_{26}\text{H}_{21}\text{NO}_3$ ($\text{M}+\text{H}$)⁺ 396.1600 found 396.1590.

(E)-1-(4,5-dimethoxy-2-(quinolin-4-yl)phenyl)-3-(naphthalen-1-yl)prop-2-en-1-one (7e).

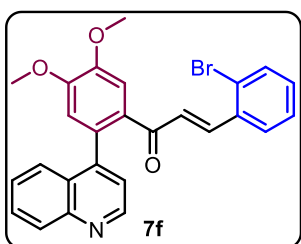
This compound was isolated as a yellowish-brown solid by following the general procedure-



2. 125 mg of **AF** afforded 136 mg of **7e** (75% yield), M.P = 157-159.4 °C. $R_f = 0.3$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2955, 2931, 2852, 1654, 1596, 1570, 1513, 1464, 1348, 1274, 1265, 1248, 1213, 1147, 1088, 1028, 976, 804, 777, 749. **^1H NMR (400 MHz, CDCl_3):** δ 8.91 (dd, $J = 4.4, 1.6$ Hz, 1H), 8.20-8.12 (m, 2H), 8.10 (d, $J = 15.5$ Hz, 1H), 7.86-7.80 (m, 1H), 7.80-7.72 (m, 3H), 7.61-7.54 (m, 1H), 7.52 (s, 1H), 7.48-7.40 (m, 2H), 7.38 (dd, $J = 8.5, 4.2$ Hz, 1H), 7.21 (t, $J = 7.8$ Hz, 1H), 6.94 (s, 1H), 6.62 (d, $J = 7.3$ Hz, 1H), 6.42 (d, $J = 15.4$ Hz, 1H), 4.07 (s, 3H), 3.97 (s, 3H). **^{13}C NMR (100 MHz, $\text{DMSO}-d_6$):** δ 192.49, 151.22, 149.86, 149.10, 148.49, 147.64, 140.07, 133.47, 133.08, 131.65, 131.32, 130.66, 130.53, 129.94, 129.68, 128.63, 127.79, 127.37, 127.33, 126.75, 126.11, 125.73, 125.25, 124.82, 123.15, 122.39, 113.36, 112.17, 56.32, 56.29. **HRMS (ESI):** m/z calcd for $\text{C}_{30}\text{H}_{24}\text{NO}_3$ ($\text{M}+\text{H}$)⁺ 446.1756 found 446.1757.

(E)-3-(2-Bromophenyl)-1-(4,5-dimethoxy-2-(quinolin-4-yl)phenyl)prop-2-en-1-one (7f).

This compound was isolated as yellowish-brown semi-solid by following the general

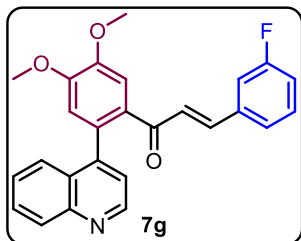


procedure-2. 120 mg of **AF** afforded 166 mg of **7f** (90% yield), $R_f = 0.3$ (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2954, 2923, 2853, 1656, 1593, 1564, 1513, 1464, 1439, 1345, 1275, 1200, 1145, 1100, 1026, 805, 757, 733. **^1H NMR (400 MHz, CDCl_3):** δ 8.90 (d, $J = 4.3$ Hz, 1H), 8.14 (dt, $J = 8.5, 0.9$ Hz, 1H), 7.79 (dd, $J = 8.4, 1.4$ Hz, 1H), 7.71 (ddd, $J = 8.4, 6.9, 1.4$ Hz, 1H), 7.60 (d, $J = 15.8$ Hz, 1H), 7.53 (ddd, $J = 8.3, 6.8, 1.3$ Hz, 1H), 7.46 (d, $J = 1.7$ Hz, 1H), 7.44 (d, $J = 1.4$ Hz, 1H), 7.32 (d, $J = 4.4$ Hz, 1H), 7.08 (td, $J = 7.6, 1.8$ Hz, 1H), 7.02 (td, $J = 7.6, 1.5$ Hz, 1H), 6.92 (s, 1H), 6.43 (dd, $J = 7.7, 1.8$ Hz, 1H), 6.31 (d, $J = 15.8$ Hz, 1H), 4.05 (s, 3H), 3.95 (s, 3H). **^{13}C NMR (100 MHz, CDCl_3):** δ 192.56, 151.22, 149.82, 149.03, 148.43, 147.47, 141.49, 134.31, 133.19, 132.65, 131.08, 130.71, 129.88, 129.65, 128.01, 127.46, 127.34, 127.29, 125.72, 125.39, 122.43,

113.27, 113.25, 112.20, 56.31, 56.27. **HRMS (ESI):** m/z calcd for $C_{26}H_{21}BrNO_3$ ($M+H$)⁺ 474.0705 found 474.0692.

(E)-1-(4,5-Dimethoxy-2-(quinolin-4-yl)phenyl)-3-(3-fluorophenyl)prop-2-en-1-one (7g).

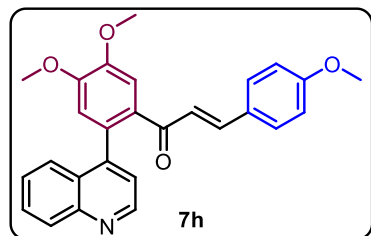
This compound was isolated as yellowish-brown oil by following the general procedure-2.



180 mg of **AF** afforded 190 mg of **7g** (78% yield). R_f = 0.3 (4:6 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2990, 2933, 1661, 1592, 1515, 1447, 1348, 1275, 1248, 1147, 806, 750. **¹H NMR (400 MHz, CDCl₃):** δ 8.92 (dd, J = 4.2, 1.7 Hz, 1H), 8.13-8.05 (m, 2H), 7.75 (dd, J = 8.5, 7.1 Hz, 1H), 7.53 (dd, J = 7.1, 1.2 Hz, 1H), 7.45 (s, 1H), 7.40 (dd, J = 8.5, 4.2 Hz, 1H), 7.17-7.06 (m, 2H), 6.92 (d, J = 3.4 Hz, 2H), 6.61 (dt, J = 7.7, 1.3 Hz, 1H), 6.47 (d, J = 9.4 Hz, 1H), 6.28 (d, J = 15.8 Hz, 1H), 4.04 (s, 3H), 3.95 (s, 3H). **¹³C NMR (100 MHz, CDCl₃):** δ 192.64, 162.66 (d, J_{C-F} = 245.3 Hz), 151.36, 150.55, 148.83, 148.20, 140.70 (d, J_{C-F} = 2.9 Hz), 139.00, 136.62 (d, J_{C-F} = 7.7 Hz), 134.08, 133.17, 132.06, 130.10 (d, J_{C-F} = 8.2 Hz), 129.77, 128.93, 128.23, 127.46, 126.48, 123.78 (d, J_{C-F} = 2.9 Hz), 121.67, 116.95 (d, J_{C-F} = 21.3 Hz), 114.0, 113.80, 111.94, 56.24 (2C). **¹⁹F NMR (376 MHz, CDCl₃):** δ -115.00. **HRMS (ESI):** m/z calcd for $C_{26}H_{21}FNO_3$ ($M+H$)⁺ 414.1505 found 414.1508.

(E)-1-(4,5-Dimethoxy-2-(quinolin-4-yl)phenyl)-3-(4-methoxyphenyl)prop-2-en-1-one (7h).

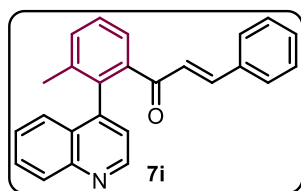
This compound was isolated as pale-yellow solid by following the general procedure-2. 200



mg of **AF** afforded 221 mg of **7h** (80% yield), M.P = 153-156 °C. R_f = 0.4 (5:5 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2921, 1651, 1595, 1570, 1510, 1463, 1250, 1100, 991, 828, 806. **¹H NMR (400 MHz, CDCl₃):** δ 8.90 (dd, J = 4.2, 1.7 Hz, 1H), 8.12-8.06 (m, 2H), 7.71 (dd, J = 8.5, 7.0 Hz, 1H), 7.50 (dd, J = 7.1, 1.2 Hz, 1H), 7.42 (s, 1H), 7.38 (dd, J = 8.6, 4.2 Hz, 1H), 7.18 (d, J = 15.7 Hz, 1H), 6.89 (s, 1H), 6.81 (d, J = 8.8 Hz, 2H), 6.66 (d, J = 8.8 Hz, 2H), 6.19 (d, J = 15.7 Hz, 1H), 4.02 (s, 3H), 3.92 (s, 3H), 3.73 (s, 3H). **¹³C NMR (100 MHz, CDCl₃):** δ 193.05, 161.34, 150.93, 150.34, 148.08, 142.62, 139.18, 134.34, 133.65, 132.13, 132.03, 131.64, 129.62 (2C), 128.58, 128.46, 128.19, 127.45, 127.02, 123.24, 121.54, 114.09 (2C), 111.90, 56.19 (2C), 55.29. **HRMS (ESI):** m/z calcd for $C_{27}H_{24}NO_4$ ($M+H$)⁺ 426.1705 found 426.1693.

(E)-1-(3-Methyl-2-(quinolin-4-yl)phenyl)-3-phenylprop-2-en-1-one (7i).

This compound was isolated as pale-yellow solid by following the general procedure-2. 150

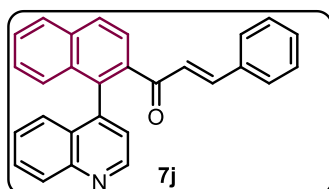


mg of **AF** afforded 157 mg of **7i** (78% yield), M.P = 86-89 °C. R_f = 0.4 (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 3060, 2922, 1668, 1643, 1601, 1574, 1505, 1448, 1330, 1266, 1055, 979, 845, 828, 761, 734, 701. **¹H NMR (400 MHz, CDCl₃):** δ 8.88 (d, J = 4.4 Hz, 1H), 8.14 (d, J = 8.4 Hz, 1H), 7.70 (ddd, J = 8.4, 6.8, 1.5 Hz, 1H), 7.62-7.56 (m, 2H), 7.53-7.47 (m, 3H), 7.26 (dd, J =

8.3, 6.0 Hz, 2H), 7.20 (dd, $J = 7.4, 5.4$ Hz, 3H), 7.04-7.00 (m, 2H), 6.56 (d, $J = 15.9$ Hz, 1H), 2.01 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 194.92, 150.05, 148.23, 146.38, 144.67, 140.84, 137.35, 135.50, 134.14, 132.52, 130.47, 130.13, 129.41, 128.73(2C), 128.44, 128.15(2C), 127.31, 127.18, 126.06, 125.81, 125.67, 122.24, 20.21. HRMS (ESI): m/z calcd for $\text{C}_{25}\text{H}_{20}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 350.1545 found 350.1551.

(E)-3-phenyl-1-(1-(quinolin-4-yl)naphthalen-2-yl)prop-2-en-1-one (7j).

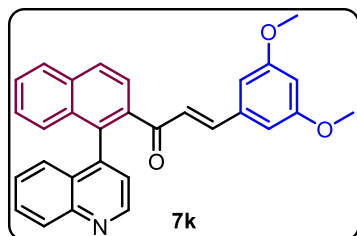
This compound was isolated as reddish-brown sticky oil by following the general procedure-



2. 200 mg of **AF** afforded 220 mg of **7j** (90% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2953, 2922, 2852, 1659, 1638, 1581, 1506, 1420, 1388, 1367, 1207, 1011, 971, 854, 762, 708. ^1H NMR (400 MHz, CDCl_3): δ 8.96 (d, $J = 4.3$ Hz, 1H), 8.24-8.15 (m, 1H),

8.13-8.06 (m, 1H), 8.05-7.96 (m, 1H), 7.85 (d, $J = 8.5$ Hz, 1H), 7.72 (ddd, $J = 8.4, 6.6, 1.7$ Hz, 1H), 7.59 (ddd, $J = 8.2, 6.7, 1.3$ Hz, 1H), 7.50 – 7.40 (m, 2H), 7.40-7.34 (m, 2H), 7.32-7.17 (m, 5H), 6.99-6.92 (m, 2H), 6.59 (d, $J = 16.0$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 195.24, 150.00, 148.16, 145.24, 144.73, 138.06, 134.16, 134.07, 131.83, 130.55, 130.04, 129.59, 129.20, 128.75 (2C), 128.34, 128.18 (2C), 127.61 (2C), 127.37 (2C), 127.28, 127.06, 126.33, 125.84, 124.93, 123.56. HRMS (ESI): m/z calcd for $\text{C}_{28}\text{H}_{20}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 386.1545 found 386.1539.

(E)-3-(3,5-dimethoxyphenyl)-1-(1-(quinolin-4-yl)naphthalen-2-yl)prop-2-en-1-one (7k).

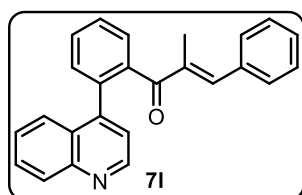


This compound was isolated as pale-yellow oil by following the general procedure-2. 150 mg of **AF** afforded 202 mg of **7k** (90% yield), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2921, 1654, 1593, 1572, 1508, 1462, 1247, 1108, 992, 829, 808. ^1H NMR (400 MHz, CDCl_3): δ 8.97 (d, $J = 4.3$ Hz, 1H), 8.20 (d, $J = 8.5$ Hz, 1H), 8.11 (d, $J = 8.5$ Hz, 1H), 8.02 (d, $J = 8.2$ Hz, 1H),

7.84 (d, $J = 8.5$ Hz, 1H), 7.72 (ddd, $J = 8.5, 6.4, 1.9$ Hz, 1H), 7.60 (ddd, $J = 8.2, 6.7, 1.2$ Hz, 1H), 7.50-7.35 (m, 4H), 7.27 (d, $J = 8.6$ Hz, 1H), 7.22 (d, $J = 15.9$ Hz, 1H), 6.52 (d, $J = 15.9$ Hz, 1H), 6.41 (t, $J = 2.3$ Hz, 1H), 6.18 (d, $J = 2.3$ Hz, 2H), 3.70 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3): δ 195.25, 160.78 (2C), 150.02, 148.13, 145.10, 144.94, 137.96, 135.91, 134.17, 134.03, 131.77, 130.05, 129.65, 129.21, 128.32, 128.11, 127.60, 127.35, 127.17, 127.06, 126.36, 126.07, 124.96, 123.60, 105.90 (2C), 103.16, 55.40, 55.37. HRMS (ESI): m/z calcd for $\text{C}_{30}\text{H}_{24}\text{NO}_3$ ($\text{M}+\text{H}$) $^+$ 446.1756 found 446.1757.

(E)-2-methyl-3-phenyl-1-(2-(quinolin-4-yl)phenyl)prop-2-en-1-one (7l).

This compound was isolated as a reddish-brown solid by following the general procedure-4.

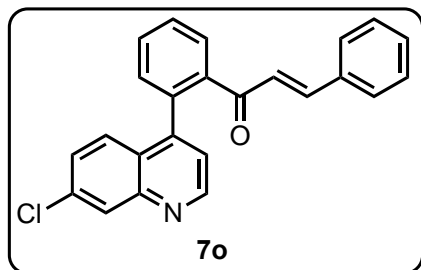


200 mg of **AL** afforded 187 mg of **7l** (70% yield), M.P = 132.8-134 °C. $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2923, 1649, 1583, 1508, 1444, 1419, 1357, 1240, 1011, 764. ^1H NMR (400 MHz, CDCl_3): δ 8.81

(d, $J = 4.4$ Hz, 1H), 8.12 (dd, $J = 8.5, 1.2$ Hz, 1H), 7.76 (dd, $J = 8.4, 1.4$ Hz, 1H), 7.74-7.69 (m, 1H), 7.65-7.60 (m, 3H), 7.50-7.46 (m, 1H), 7.39 (ddd, $J = 8.4, 6.9, 1.4$ Hz, 1H), 7.20-7.15 (m, 4H), 6.87 (d, $J = 1.6$ Hz, 1H), 6.77-6.72 (m, 2H), 1.60 (d, $J = 1.4$ Hz, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 200.5, 149.5, 148.3, 147.2, 142.9, 140.7, 137.9, 136.3, 135.0, 130.4, 129.9, 129.7, 129.5, 129.1 (2C), 129.0, 128.6, 128.4, 128.1 (2C), 126.9, 126.7, 126.0, 121.9, 12.9. HRMS (ESI): m/z calcd for $\text{C}_{25}\text{H}_{20}\text{NO}$ ($\text{M}+\text{H}^+$) 350.1545 found 350.1548.

(E)-1-(2-(7-Chloroquinolin-4-yl)phenyl)-3-phenylprop-2-en-1-one (7o).

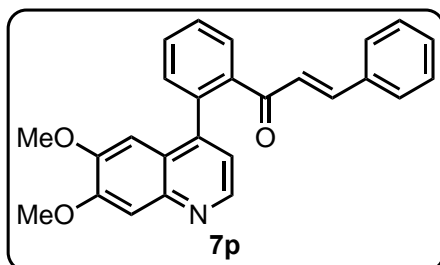
This compound was isolated as pale-yellow solid by following the general procedure-2. 150 mg of AF afforded 166 mg of **7o** (84% yield), $R_f = 0.3$



(3:7 EtOAc: Hexanes, visualized by 254 nm UV light). M.P = 123-125 °C. IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3058, 2924, 2854, 1667, 1643, 1580, 1497, 1447, 1416, 1331, 1288, 1209, 1070, 1018, 881, 828, 760. ^1H NMR (400 MHz, CDCl_3): δ 8.85 (d, $J = 4.4$ Hz, 1H), 8.09 (d, $J = 2.0$ Hz, 1H), 7.82 (dd, $J = 7.0, 1.8$ Hz, 1H), 7.65-7.60 (m, 3H), 7.43-7.40 (m, 2H), 7.30-7.23 (m, 5H), 7.13 (d, $J = 7.2$ Hz, 2H), 6.66 (d, $J = 15.9$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 194.08, 150.79, 148.73, 147.57, 145.23, 140.11, 136.23, 135.39, 134.02, 130.95 (2C), 130.76, 129.04, 128.91, 128.87 (2C), 128.81, 128.21 (2C), 127.93, 127.13, 125.59, 125.27, 121.96. HRMS (ESI): m/z calcd for $\text{C}_{24}\text{H}_{17}\text{ClNO}$ ($\text{M}+\text{H}^+$) 370.0999 found 370.1002.

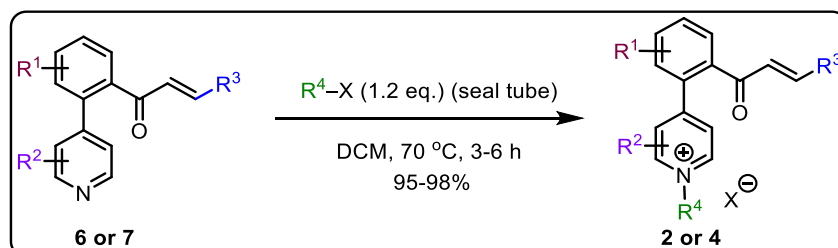
(E)-1-(2-(6,7-Dimethoxyquinolin-4-yl)phenyl)-3-phenylprop-2-en-1-one (7p).

This compound was isolated as pale-yellow sticky oil by following the general procedure-2.



150 mg of AF afforded 173 mg of **7p** (89% yield), $R_f = 0.5$ (1:1 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3058, 2960, 2833, 1666, 1643, 1602, 1493, 1350, 1244, 1214, 1109, 1005, 861, 760. ^1H NMR (400 MHz, CDCl_3): δ 8.62 (d, $J = 4.5$ Hz, 1H), 7.76 (dd, $J = 7.4, 1.2$ Hz, 1H), 7.61-7.52 (m, 2H), 7.42 (dd, $J = 7.5, 1.2$ Hz, 1H), 7.34 (s, 1H), 7.21-7.11 (m, 5H), 6.98 (d, $J = 7.2$ Hz, 2H), 6.85 (s, 1H), 6.48 (d, $J = 15.9$ Hz, 1H), 3.93 (s, 3H), 3.76 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 194.49, 152.25, 150.04, 147.52, 145.58, 145.38, 144.34, 140.38, 137.01, 134.10, 131.06, 130.71, 130.48, 129.06, 128.74 (2C), 128.69, 128.06 (2C), 125.34, 122.35, 120.31, 108.33, 103.11, 56.08, 55.90. HRMS (ESI): m/z calcd for $\text{C}_{26}\text{H}_{22}\text{NO}_3$ ($\text{M}+\text{H}^+$) 396.1600 found 396.1601.

General procedure-5: Synthesis of enone-tethered pyridinium (2a-2z)- and quinolinium salts (4a-4m)



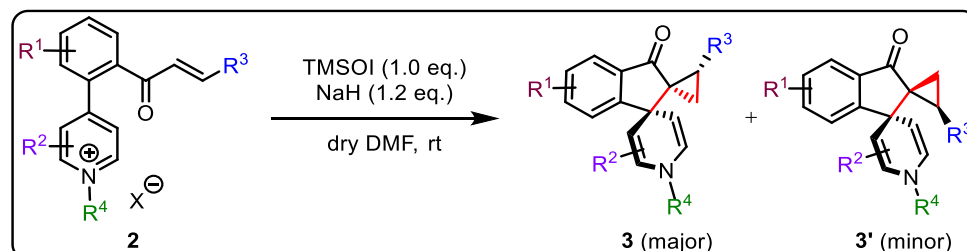
Scheme S5: Synthesis of enone-tethered pyridinium **2** and quinolinium salts **4**

In a sealed tube, biaryl enone **6** or **7** (1.0 eq.) was dissolved in DCM (2 mL). Alkyl bromide or iodide (1.2 eq.) was added in one portion, and the reaction mixture was stirred at 70 °C for 3-6 h and monitored the reaction was on TLC. After the completion of starting material, the solvent was evaporated, and the crude product was washed with ethyl acetate 4-5 times. The product was dried over a vacuum to get a pale-yellow or reddish-brown solid and transferred to the final step without further purification.

4-(2-Cinnamoylphenyl)-1-methylpyridin-1-ium (Iodide) (**2a**).

This compound was isolated as an orangish-brown solid by following the general procedure-**5**. 500 mg of **6a** afforded 718 mg of **2a** (98% yield), M.P = 136.8-138.5 °C. $R_f = 0.2$ (10:1 DCM: MeOH, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2928, 1706, 1638, 1595, 1572, 1448, 1361, 1284, 1214, 1015, 982, 764, 727. **^1H NMR (400 MHz, CDCl_3):** δ 9.26 (d, $J = 6.1$ Hz, 2H), 7.88 (d, $J = 6.1$ Hz, 2H), 7.81 (dd, $J = 7.1, 1.8$ Hz, 1H), 7.66 (td, $J = 6.9, 1.6$ Hz, 2H), 7.60-7.47 (m, 4H), 7.40 (t, $J = 3.7$ Hz, 3H), 7.17 (d, $J = 16.0$ Hz, 1H), 4.60 (s, 3H). **^{13}C NMR (100 MHz, CDCl_3):** δ 193.0, 158.5, 147.8, 144.9 (2C), 138.5, 135.5, 133.8, 132.1, 131.4, 131.0, 130.7, 129.6, 129.1 (2C), 128.8 (2C), 127.8 (2C), 124.6, 48.7. **HRMS (ESI):** m/z calcd for $\text{C}_{21}\text{H}_{18}\text{NO}$ ($\text{M}-\text{I}$) $^+$ 300.1383 found. 300.1403.

General procedure-6: Synthesis of vicinal bis-spirocyclic indanone (**3**)



Scheme S6: Synthesis of vicinal bis-spirocyclic indanones **3**

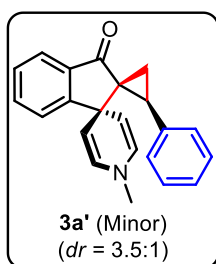
A mixture of sodium hydride (60% in oil, 12 mg, 0.28 mmol) and trimethylsulfoxonium iodide (TMSOI) (51 mg, 0.23 mmol) were placed in an oven-dried flask, and dry DMF (4.0 mL) was added to the mixture. After hydrogen evolution ceased, the milky solution turned clear, and the reaction mixture was stirred for 15 min. The compound

2a (100 mg, 0.23 mmol) was dissolved in dry DMF (2.0 mL) and was added to the clear solution drop wise over a period of 5-10 min and stirred at room temperature until the reactant **2a** disappeared as monitored by TLC. The reaction mixture was quenched using ice water and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate.

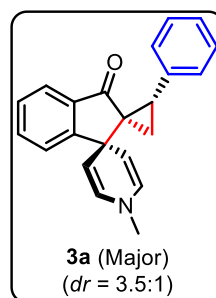
Spectral data of all vicinal bis-spirocyclic indanones reported in this study

1''-Methyl-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (**3a'**).

This compound was isolated as pale-yellow solid by following the general procedure-6. 100 mg of **2a** afforded 18 mg of **3a'** (25% yield), $R_f = 0.6$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light), M.P = 122.6-125.3 °C. **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3055, 1697, 1674, 1600, 1497, 1408, 1290, 1113, 1007, 932, 756, 725. **3a' (Minor):** $^1\text{H NMR}$ (500 MHz, CDCl_3): δ 7.72-7.69 (m, 1H), 7.64 (ddd, $J = 7.8, 7.1, 1.2$ Hz, 1H), 7.57 (dt, $J = 7.8, 1.0$ Hz, 1H), 7.35 (ddd, $J = 7.7, 7.1, 1.1$ Hz, 1H), 7.21-7.10 (m, 5H), 5.96 (dd, $J = 7.8, 1.8$ Hz, 1H), 4.90 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.12 (dd, $J = 7.8, 3.0$ Hz, 1H), 3.52 (dd, $J = 7.8, 3.0$ Hz, 1H), 3.20 (dd, $J = 9.3, 7.9$ Hz, 1H), 2.71 (s, 3H), 2.02 (dd, $J = 7.9, 4.2$ Hz, 1H), 1.57 (dd, $J = 9.3, 4.1$ Hz, 1H). $^{13}\text{C NMR}$ (125 MHz, CDCl_3): δ 204.98, 164.03, 137.56, 134.80, 132.64, 130.54, 129.37 (2C), 127.65, 127.55, 127.41, 127.09 (2C), 125.68, 121.52, 104.21, 100.29, 53.20, 45.23, 40.14, 32.66, 18.85.

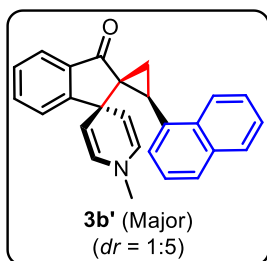


3a (Major): This compound was isolated as pale-yellow solid following general procedure-6. 100 mg of **2a** afforded 48 mg of **3a** (61% yield), $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light), M.P = 102.4-105.6 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.69-7.65 (m, 2H), 7.57 (dd, $J = 7.6, 1.1$ Hz, 1H), 7.34 (ddd, $J = 8.0, 4.9, 3.4$ Hz, 1H), 7.29 (d, $J = 4.4$ Hz, 4H), 7.25-7.18 (m, 1H), 6.20 (dd, $J = 7.8, 1.7$ Hz, 1H), 6.09 (dd, $J = 7.8, 1.8$ Hz, 1H), 4.32 (dd, $J = 7.8, 2.9$ Hz, 1H), 4.09 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.02 (s, 3H), 2.99 (t, $J = 8.6$ Hz, 1H), 2.10 (dd, $J = 8.2, 4.3$ Hz, 1H), 1.71 (dd, $J = 9.1, 4.3$ Hz, 1H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3): δ 202.95, 162.65, 136.43, 134.65, 134.46, 130.49, 129.91, 129.28 (2C), 127.74 (2C), 127.71, 127.48, 126.52, 121.56, 101.94, 101.88, 52.93, 46.14, 40.51, 33.67, 18.70. **HRMS (ESI):** m/z calcd for $\text{C}_{22}\text{H}_{18}\text{NO}$ (M-H) $^+$ 312.1388 found: 312.1395.



1''-Methyl-2-(naphthalen-1-yl)-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (**3b'**).

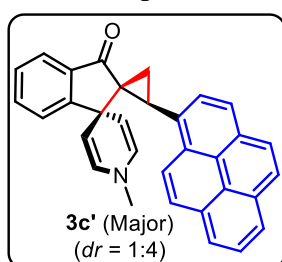
This compound was isolated as pale-yellow oil by following the general procedure-6. 80 mg of **2b** afforded 46 mg of **3b** (76% yield combined), $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3051, 2953, 2907, 2834, 1695, 1673, 1599, 1462, 1293, 1083, 797. $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 8.05-7.97 (m, 1H), 7.85-7.78 (m, 2H), 7.72 (d, $J = 8.1$ Hz, 1H), 7.64 (td, $J = 7.4, 1.2$ Hz, 1H), 7.53 (d, $J = 7.7$ Hz, 1H),



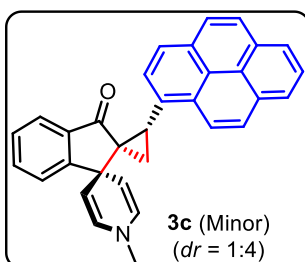
7.42 (ddd, $J = 8.2, 5.0, 1.4$ Hz, 3H), 7.39-7.33 (m, 1H), 7.31-7.26 (m, 1H), 5.90 (dd, $J = 7.8, 1.7$ Hz, 1H), 4.32 (dd, $J = 7.8, 1.7$ Hz, 1H), 4.08 (dd, $J = 7.8, 3.0$ Hz, 1H), 3.76 (t, $J = 8.6$ Hz, 1H), 3.16 (dd, $J = 7.8, 2.9$ Hz, 1H), 2.55 (s, 3H), 2.30 (dd, $J = 8.0, 4.0$ Hz, 1H), 1.74 (dd, $J = 9.2, 4.0$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 205.29, 163.90, 134.81, 134.76, 134.16, 133.01, 132.78, 130.26, 128.27, 127.67, 127.43, 127.08, 126.66, 125.93, 125.34, 124.59, 124.36, 124.33, 121.61, 103.19, 99.98, 53.35, 45.37, 39.92, 30.14, 18.51. HRMS (ESI): m/z calcd for $\text{C}_{26}\text{H}_{22}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 364.1701 found 364.1691.

1''-Methyl-2-(pyren-1-yl)-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3c/3c').

This compound was isolated as yellowish-brown sticky oil by following the general procedure-6. 120 mg of 2c afforded 57 mg of 3c' (60% yield) $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2929, 1700, 1674, 1601, 1463, 1384, 1360, 1206, 1104, 1006, 845, 726. 3c' (Major): ^1H NMR (500 MHz, CDCl_3): δ 8.22 (d, $J = 9.3$ Hz, 1H), 8.14-8.08 (m, 2H), 8.03-7.96 (m, 4H), 7.94 (t, $J = 7.6$ Hz, 1H), 7.83-7.76 (m, 2H), 7.58 (td, $J = 7.4, 1.3$ Hz, 1H), 7.45 (dt, $J = 7.7, 0.9$ Hz, 1H), 7.37 (td, $J = 7.5, 1.0$ Hz, 1H), 5.83 (dd, $J = 7.8, 1.7$ Hz, 1H), 4.07 (dd, $J = 7.8, 3.0$ Hz, 1H), 4.02 (t, $J = 8.6$ Hz, 1H), 3.85 (dd, $J = 7.8, 1.7$ Hz, 1H), 3.14 (dd, $J = 7.8, 3.0$ Hz, 1H), 2.44 (dd, $J = 8.0, 4.1$ Hz, 1H), 2.31 (s, 3H), 1.83 (dd, $J = 9.2, 4.1$ Hz, 1H). ^{13}C NMR (125 MHz, CDCl_3): δ 205.03, 163.92, 134.82, 133.00, 131.85, 131.79, 131.37, 130.92, 130.43, 129.98, 127.63, 127.46, 127.37 (2C), 127.18, 126.79, 125.80, 125.19, 125.06, 124.86, 124.71, 124.12, 124.10, 123.57, 121.68, 102.89, 99.81, 53.48, 45.42, 39.66, 30.64, 18.88.

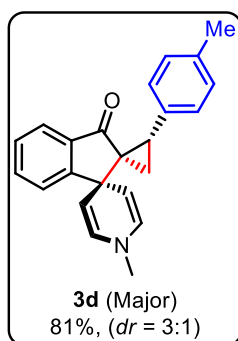


3c (Minor): This compound was isolated as pale-yellow sticky oil by following the general procedure-6. 100 mg of 2c afforded 19 mg of 3c (20% yield) $R_f = 0.4$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). ^1H NMR (500 MHz, CDCl_3): δ 8.15 (dd, $J = 8.6, 1.7$ Hz, 2H), 8.12 (dd, $J = 7.5, 1.2$ Hz, 1H), 8.06-8.01 (m, 3H), 7.99 (d, $J = 8.9$ Hz, 1H), 7.92 (t, $J = 7.6$ Hz, 1H), 7.84 (d, $J = 9.2$ Hz, 1H), 7.76 (dt, $J = 7.8, 1.0$ Hz, 1H), 7.68 (td, $J = 7.5, 1.2$ Hz, 1H), 7.38 (dt, $J = 7.7, 1.0$ Hz, 1H), 7.28 (td, $J = 7.3, 1.1$ Hz, 1H), 6.34 (dd, $J = 7.9, 1.8$ Hz, 1H), 6.12 (dd, $J = 7.8, 1.8$ Hz, 1H), 4.73 (dd, $J = 7.9, 2.9$ Hz, 1H), 4.17 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.54 (t, $J = 8.4$ Hz, 1H), 3.05 (s, 3H), 2.35 (dd, $J = 7.8, 4.3$ Hz, 1H), 1.97 (dd, $J = 9.0, 4.3$ Hz, 1H). ^{13}C NMR (125 MHz, CDCl_3): δ 202.56, 162.45, 134.68, 134.51, 131.67, 131.39, 131.15, 130.75, 130.36, 129.63, 128.17, 127.68, 127.60, 127.56 (2C), 126.96, 126.75, 125.59, 124.90, 124.88, 124.77, 124.70, 124.38, 123.56, 121.69, 103.24, 100.21, 52.93, 46.63, 40.55, 31.50, 19.44. HRMS (ESI): m/z calcd for $\text{C}_{32}\text{H}_{24}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 438.1858 found: 438.1859.



1''-Methyl-2-(p-tolyl)-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3d).

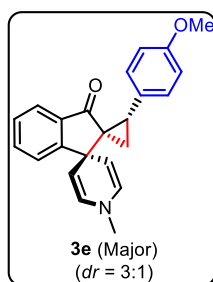
This compound was isolated as pale-yellow semi-solid by following the general procedure-6.



70 mg of **2d** afforded 42 mg of **3d** (81% yield, combined) $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2924, 2965, 2854, 1701, 1671, 1600, 1517, 1462, 1378, 1208, 1022, 1008, 821, 765, 709. **^1H NMR (500 MHz, CDCl_3):** δ 7.65-7.60 (m, 2H), 7.54 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.30 (ddd, $J = 7.6, 5.2, 3.1$ Hz, 1H), 7.14 (d, $J = 8.0$ Hz, 2H), 7.06 (d, $J = 7.7$ Hz, 2H), 6.16 (dd, $J = 7.8, 1.8$ Hz, 1H), 6.05 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.28 (dd, $J = 7.9, 2.9$ Hz, 1H), 4.06 (dd, $J = 7.8, 3.0$ Hz, 1H), 2.98 (s, 3H), 2.93 (t, $J = 8.6$ Hz, 1H), 2.29 (s, 3H), 2.04 (dd, $J = 8.2, 4.2$ Hz, 1H), 1.65 (dd, $J = 9.1, 4.3$ Hz, 1H). **^{13}C (125 MHz, CDCl_3):** δ 202.97, 162.66, 135.92, 134.72, 134.36, 133.28, 130.44, 129.86, 129.10 (2C), 128.50 (2C), 127.66, 127.42, 121.54, 102.05, 101.95, 52.89, 46.13, 40.48, 33.46, 21.18, 18.73. **HRMS (ESI):** m/z calcd for $\text{C}_{23}\text{H}_{21}\text{NNaO}$ ($\text{M}+\text{Na}$) $^+$ 350.1521 found. 350.1546.

2-(4-Methoxyphenyl)-1''-methyl-1''*H*,3''*H*-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (**3e**).

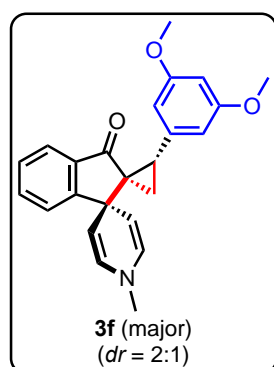
This compound was isolated as pale-yellow solid by following the general procedure-6. 100



mg of **2e** afforded 56 mg of **3e** (74% yield, combined), M.P = 135-142 °C. $R_f = 0.4$ (1:4 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2929, 2834, 1698, 1671, 1599, 1513, 1462, 1320, 1209, 1175, 1007, 973, 800, 726, 708. **^1H NMR (400 MHz, CDCl_3):** δ 7.70 -7.64 (m, 2H), 7.57 (d, $J = 7.6$ Hz, 1H), 7.34 (ddd, $J = 8.0, 4.8, 3.3$ Hz, 1H), 7.23-7.18 (m, 2H), 6.85-6.81 (m, 2H), 6.19 (dd, $J = 7.9, 1.7$ Hz, 1H), 6.08 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.29 (dd, $J = 7.8, 2.9$ Hz, 1H), 4.08 (dd, $J = 7.8, 2.9$ Hz, 1H), 3.79 (s, 3H), 3.01 (s, 3H), 2.95 (t, $J = 8.6$ Hz, 1H), 2.06 (dd, $J = 8.1, 4.2$ Hz, 1H), 1.69 (dd, $J = 9.0, 4.3$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 203.09, 162.71, 158.14, 134.70, 134.40, 130.49, 130.20 (2C), 129.88, 128.34, 127.69, 127.45, 121.51, 113.19 (2C), 102.01, 101.87, 55.15, 52.95, 46.12, 40.50, 33.22, 18.90. **HRMS (ESI):** m/z calcd for $\text{C}_{23}\text{H}_{22}\text{NO}_2$ ($\text{M}+\text{H}$) $^+$ 344.1651, found: 344.1663.

2-(3,5-Dimethoxyphenyl)-1''-methyl-1''*H*,3''*H*-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (**3f**).

This compound was isolated as yellowish-brown semi-solid by following the general

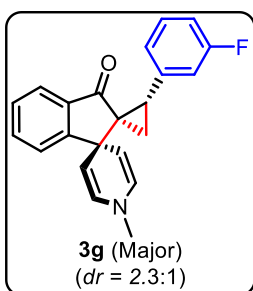


procedure-6. 120 mg of **2f** afforded 70 mg of **3f** (77% yield, combined), $R_f = 0.4$ (3:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2927, 1688, 1671, 1595, 1495, 1358, 1277, 1118, 1011, 866, 725. **^1H NMR (400 MHz, CDCl_3):** δ 7.71-7.63 (m, 2H), 7.58 (d, $J = 7.6$ Hz, 1H), 7.39-7.30 (m, 1H), 6.44 (d, $J = 2.3$ Hz, 2H), 6.33 (t, $J = 2.3$ Hz, 1H), 6.18 (dd, $J = 7.8, 1.8$ Hz, 1H), 6.08 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.29 (dd, $J = 7.8, 3.0$ Hz, 1H), 4.07 (dd, $J = 7.9, 3.0$ Hz, 1H), 3.78 (s, 6H), 3.01 (s, 3H), 2.92 (t, $J = 8.6$ Hz, 1H), 2.05 (dd, $J = 8.2, 4.3$ Hz, 1H), 1.66 (dd, $J = 9.0, 4.3$ Hz,

1H). ^{13}C NMR (100 MHz, CDCl_3): δ 202.86, 162.57, 160.13 (2C), 139.01, 134.63, 134.49, 130.50, 129.91, 127.71, 127.51, 121.57, 107.59 (2C), 101.86, 101.77, 98.45, 55.24, 55.21, 52.93, 46.11, 40.50, 33.81, 18.80. HRMS (ESI): m/z calcd for $\text{C}_{24}\text{H}_{24}\text{NO}_3$ ($\text{M}+\text{H}^+$) 374.1756 found 374.1768.

2-(3-Fluorophenyl)-1''-methyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3g).

This compound was isolated as a pale-yellow liquid by following the general procedure-6. 80



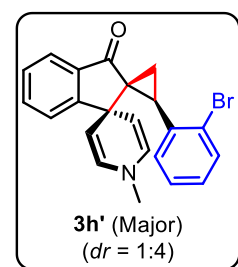
mg of **2g** afforded 48 mg of **3g** (81% yield, combined), R_f = 0.5 (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat):

$\nu_{\text{max}}/\text{cm}^{-1}$ 2929, 1700, 1671, 1614, 1600, 1587, 1446, 1382, 1222, 1206, 1011, 942, 729, 680. ^1H NMR (400 MHz, CDCl_3): δ 7.71-7.64 (m, 2H), 7.58 (dt, J = 7.7, 1.1 Hz, 1H), 7.35 (ddd, J = 8.0, 4.6, 3.7 Hz, 1H), 7.30-7.20 (m, 1H), 7.06 (d, J = 7.4 Hz, 1H), 7.00 (dt, J = 10.1, 2.1 Hz, 1H), 6.90 (td, J = 8.5, 2.6 Hz, 1H), 6.20 (dd, J = 7.9, 1.8 Hz, 1H), 6.09 (dd, J = 7.9, 1.8 Hz, 1H), 4.29 (dd, J = 7.8, 3.0 Hz, 1H), 4.08 (dd, J =

7.9, 3.0 Hz, 1H), 3.02 (s, 3H), 2.95 (t, J = 8.6 Hz, 1H), 2.06 (dd, J = 8.1, 4.3 Hz, 1H), 1.71 (dd, J = 9.0, 4.3 Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 202.80, 162.61, 162.48 (d, $J_{\text{C-F}}$ = 243.2 Hz), 139.25 (d, $J_{\text{C-F}}$ = 7.7 Hz), 134.66, 134.47, 130.58, 130.02, 129.05 (d, $J_{\text{C-F}}$ = 8.2 Hz), 127.78 (2C), 127.58, 124.95 (d, $J_{\text{C-F}}$ = 2.8 Hz), 121.58, 116.22 (d, $J_{\text{C-F}}$ = 21.4 Hz), 113.45 (d, $J_{\text{C-F}}$ = 20.8 Hz), 101.68, 53.00, 46.12, 40.51, 33.07, 18.74. ^{19}F NMR (376 MHz, CDCl_3): δ -114.45. HRMS (ESI): m/z calcd for $\text{C}_{22}\text{H}_{19}\text{FNO}$ ($\text{M}+\text{H}^+$) 332.1451 found: 332.1458.

2-(2-Bromophenyl)-1''-methyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3h').

This compound was isolated as yellowish-brown semi-solid by following the general procedure-6. 100 mg of **2h** afforded 60 mg of **3h'** (77% yield,

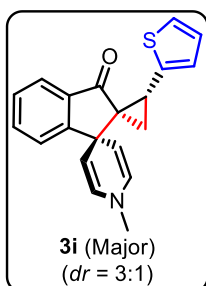


combined), R_f = 0.6 (1:9 EtOAc: Hexanes, visualized by 254nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3054, 2956, 2920, 2836, 1699, 1674, 1600, 1463, 1378, 1292, 1208, 1023, 1007, 765, 748, 719. ^1H

NMR (400 MHz, CDCl_3): δ 7.76 (dt, J = 7.6, 1.0 Hz, 1H), 7.67 (td, J = 7.4, 1.2 Hz, 1H), 7.59 (dt, J = 7.7, 0.9 Hz, 1H), 7.45 (dd, J = 7.9, 1.3 Hz, 1H), 7.39 (td, J = 7.4, 1.1 Hz, 1H), 7.18 (dd, J = 7.3, 1.3 Hz, 1H), 7.13 (dd, J = 7.8, 1.9 Hz, 1H), 7.07 (dd, J = 7.5, 1.9 Hz, 1H), 5.92 (dd, J = 7.8, 1.7 Hz, 1H), 4.87 (dd, J = 7.9, 1.7 Hz, 1H), 4.06 (dd, J = 7.8, 3.0 Hz, 1H), 3.86 (dd, J = 7.9, 3.0 Hz, 1H), 3.31 (t, J = 8.6 Hz, 1H), 2.63 (s, 3H), 2.05 (dd, J = 8.1, 4.2 Hz, 1H), 1.61 (dd, J = 9.1, 4.2 Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 204.45, 163.63, 137.73, 134.81, 132.82, 131.64, 130.32, 128.86, 128.76, 127.61, 127.59, 127.45, 127.28, 126.06, 121.70, 102.28, 100.12, 52.25, 45.25, 39.94, 33.46, 18.44. HRMS (ESI): m/z calcd for $\text{C}_{22}\text{H}_{19}\text{BrNO}$ ($\text{M}+\text{H}^+$) 392.0650 found: 392.0643.

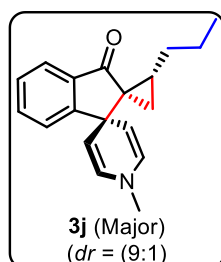
1'-Methyl-2-(thiophen-2-yl)-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3i).

This compound was isolated as reddish-brown semi-solid by following the general procedure-6. 100 mg of **2i** afforded 54 mg of **3i** (73% yield, combined), $R_f = 0.6$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2963, 2911, 1700, 1671, 1599, 1462, 1315, 1222, 1105, 993, 765, 695. **^1H NMR (400 MHz, CDCl_3):** δ 7.72-7.64 (m, 2H), 7.62 (dt, $J = 7.7, 1.1$ Hz, 1H), 7.36 (ddd, $J = 8.0, 5.0, 3.2$ Hz, 1H), 7.14 (dd, $J = 4.4, 1.9$ Hz, 1H), 6.96 (d, $J = 4.5$ Hz, 2H), 6.19 (dd, $J = 7.8, 1.7$ Hz, 1H), 6.08 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.26 (dd, $J = 7.8, 2.9$ Hz, 1H), 4.06 (dd, $J = 7.8, 2.9$ Hz, 1H), 3.01 (m, 4H), 2.08 (dd, $J = 7.8, 4.2$ Hz, 1H), 1.79 (dd, $J = 9.0, 4.2$ Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 202.16, 162.49, 140.39, 134.60, 134.48, 130.69, 130.00, 127.72, 127.58, 126.59, 126.33, 123.98, 121.64, 101.77, 101.44, 53.06, 46.13, 40.51, 27.79, 20.30. **HRMS (ESI):** m/z calcd for $\text{C}_{20}\text{H}_{18}\text{NOS}$ ($\text{M}+\text{H}$) $^+$ 320.1109 found: 320.1120.



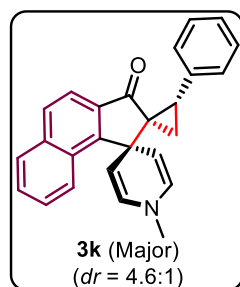
1'-Methyl-2-propyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3j).

This compound was isolated as pale-yellow oil by following the general procedure-6. 120 mg of **2j** afforded 63 mg of **6j** (74% yield, combined) $R_f = 0.6$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3055, 1697, 1674, 1600, 1497, 1405, 1290, 1107, 1010, 930, 760, 719. **^1H NMR (400 MHz, CDCl_3):** δ 7.71-7.59 (m, 3H), 7.37 (ddd, $J = 8.0, 6.8, 1.5$ Hz, 1H), 6.04 (ddd, $J = 28.4, 7.8, 1.8$ Hz, 2H), 4.03 (ddd, $J = 35.0, 7.8, 2.9$ Hz, 2H), 2.96 (s, 3H), 1.78-1.66 (m, 3H), 1.39 (dq, $J = 7.9, 3.8$ Hz, 2H), 1.33-1.19 (m, 2H), 0.89 (t, $J = 7.4$ Hz, 3H). **^{13}C NMR (100 MHz, CDCl_3):** δ 205.76, 162.87, 134.71, 134.30, 130.05, 129.63, 127.68, 127.37, 121.23, 102.15, 102.00, 49.55, 46.04, 40.43, 30.32, 28.02, 23.14, 21.46, 13.91. **HRMS (ESI):** m/z calcd for $\text{C}_{19}\text{H}_{21}\text{NO}$ (M) $^+$ 279.1623 found: 279.1654.



1'-Methyl-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-cyclopenta[a]naphthalene-1',4''-pyridin]-3'-one (3k).

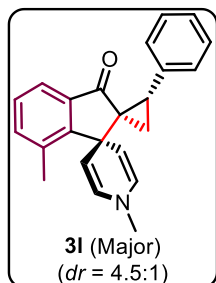
This compound was isolated as yellowish-brown sticky oil following general procedure-6. 80 mg of **2k** afforded 44 mg of **3k** (73% yield, combined), $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2929, 2958, 1711, 1699, 1674, 1594, 1454, 1310, 1221, 1038, 1006, 764, 700. **^1H NMR (500 MHz, CDCl_3):** δ 8.61-8.56 (m, 1H), 7.95-7.89 (m, 1H), 7.81-7.76 (m, 1H), 7.64-7.54 (m, 3H), 7.32-7.22 (m, 4H), 7.21-7.16 (m, 1H), 6.21 (dd, $J = 7.9, 1.8$ Hz, 1H), 6.14 (dd, $J = 7.9, 1.7$ Hz, 1H), 4.41 (dd, $J = 7.9, 3.0$ Hz, 1H), 4.17 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.13 (s, 3H), 3.10 (t, $J = 8.7$ Hz, 1H), 2.11 (dd, $J = 8.3, 4.3$ Hz, 1H), 1.81 (dd, $J = 9.2, 4.3$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 202.47, 159.10, 137.78, 136.50, 133.11, 131.05, 130.51, 130.36, 129.31(3C), 129.23, 127.96, 127.77 (2C), 126.51,



126.36, 126.18, 118.38, 103.04, 102.32, 54.00, 46.51, 40.75, 32.70, 18.12. **HRMS (ESI):** m/z calcd for $C_{26}H_{22}NO$ ($M+H$)⁺ 364.1701 found 364.1708.

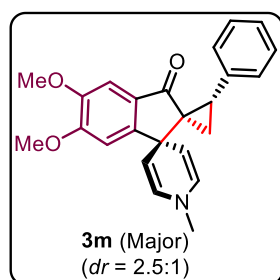
1',7'-Dimethyl-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3l).

This compound was isolated as reddish-brown oil by following the general procedure-6. 100 mg of **2i** afforded 52 mg of **3i** (71% yield, combined) $R_f = 0.6$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2927, 2854, 1697, 1671, 1599, 1478, 1379, 1217, 1199, 1015, 765, 694. **¹H NMR (500 MHz, CDCl₃):** δ 7.44 (ddd, $J = 7.6, 1.3, 0.7$ Hz, 1H), 7.38 (dt, $J = 7.4, 1.0$ Hz, 1H), 7.28-7.22 (m, 5H), 7.20-7.15 (m, 1H), 6.12 (dd, $J = 7.9, 1.7$ Hz, 1H), 6.05 (dd, $J = 7.9, 1.7$ Hz, 1H), 4.21 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.97 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.00 (s, 4H), 2.50 (s, 3H), 2.07 (dd, $J = 8.3, 4.2$ Hz, 1H), 1.72 (dd, $J = 9.1, 4.2$ Hz, 1H). **¹³C (125 MHz, CDCl₃):** δ 203.27, 157.00, 138.21, 136.97, 136.55, 135.33, 130.85, 130.62, 129.29 (2C), 127.73, 127.72 (2C), 126.47, 119.68, 100.74, 100.22, 53.45, 46.01, 40.52, 33.60, 18.78, 17.95. **HRMS (ESI):** m/z calcd for $C_{23}H_{22}NO_2$ ($M+H$)⁺ 328.1701 found: 328.1710.



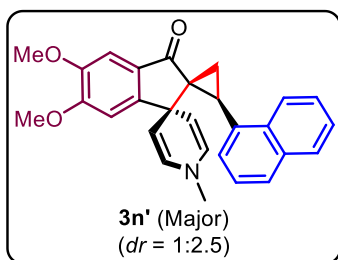
5',6'-Dimethoxy-1''-methyl-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3m).

This compound was isolated as pale-yellow semi-solid by following the general procedure-6. 100 mg of **2m** afforded 59 mg of **3m** (77% yield, combined) $R_f = 0.4$ (1:4 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2929, 1689, 1670, 1593, 1494, 1359, 1279, 1121, 1018, 869, 722. **¹H NMR (400 MHz, CDCl₃):** δ 7.30-7.24 (m, 4H), 7.23-7.16 (m, 1H), 7.05 (s, 1H), 7.02 (s, 1H), 6.19 (dd, $J = 7.8, 1.7$ Hz, 1H), 6.08 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.28 (dd, $J = 7.8, 2.9$ Hz, 1H), 4.07 (dd, $J = 7.9, 2.9$ Hz, 1H), 4.02 (s, 3H), 3.87 (s, 3H), 3.02 (s, 3H), 2.93 (t, $J = 8.6$ Hz, 1H), 2.04 (dd, $J = 8.1, 4.3$ Hz, 1H), 1.66-1.62 (m, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 201.50, 157.81, 155.29, 149.47, 136.71, 130.57, 129.95, 129.27 (2C), 127.70 (3C), 126.39, 108.59, 102.36, 102.16, 101.94, 56.30, 56.11, 53.06, 45.94, 40.52, 32.87, 17.99. **HRMS (ESI):** m/z calcd for $C_{24}H_{24}NO_3$ ($M+H$)⁺ 374.1756, found 374.1742.



5',6'-Dimethoxy-1''-methyl-2-(naphthalen-1-yl)-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3n').

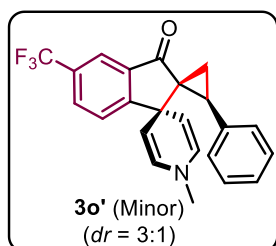
This compound was isolated as reddish-brown oil by following the general procedure-6. 120 mg of **2n** afforded 67 mg of **3n'** (71% yield, combined) $R_f = 0.5$ (1:4 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 3064, 2959, 2871, 1670, 1594, 1278, 1118, 870, 732. **¹H NMR (500 MHz, CDCl₃):** δ 8.03-7.92 (m, 1H), 7.80-7.76 (m, 1H), 7.68 (d, $J = 8.2$ Hz, 1H), 7.42-7.36 (m, 2H), 7.32 (dd, $J = 8.2, 7.2$ Hz, 1H), 7.24 (dt, $J = 7.2, 1.1$ Hz, 1H), 7.22 (s, 1H), 6.88 (s, 1H), 5.87 (dd, $J = 7.8, 1.8$ Hz, 1H), 4.29 (dd, $J =$



7.8, 1.8 Hz, 1H), 4.02 (dd, $J = 7.8, 3.0$ Hz, 1H), 3.95 (s, 3H), 3.90 (s, 3H), 3.66 (t, $J = 8.6$ Hz, 1H), 3.11 (dd, $J = 7.8, 3.0$ Hz, 1H), 2.54 (s, 3H), 2.20 (dd, $J = 8.0, 4.0$ Hz, 1H), 1.67 (dd, $J = 9.2, 4.0$ Hz, 1H). ^{13}C NMR (125 MHz, CDCl_3): δ 203.59, 159.31, 155.65, 149.54, 134.78, 134.52, 132.78, 130.25, 128.19, 127.24, 126.50, 125.90, 125.87, 125.29, 124.53, 124.44, 124.20, 108.56, 103.31, 102.25, 100.22, 56.28, 56.22, 53.49, 45.16, 39.89, 29.61, 17.70. HRMS (ESI): m/z calcd for $\text{C}_{28}\text{H}_{26}\text{NO}_3$ ($\text{M}+\text{H}$) $^+$ 424.1913 found: 424.1919.

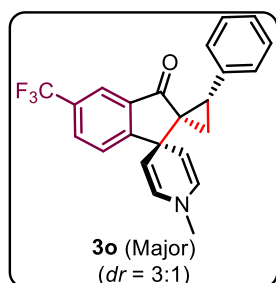
1''-Methyl-2-phenyl-5'-(trifluoromethyl)-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3o').

This compound was isolated as a pale-yellow oil by following the general procedure-6. 100



mg of **2o** afforded 20 mg of **3o'** (26% yield) $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2928, 1688, 1672, 1595, 1492, 1358, 1277, 1118, 1018, 872, 729. **3o'** (Minor):- ^1H NMR (400 MHz, CDCl_3): 7.99 (dt, $J = 1.7, 0.8$ Hz, 1H), 7.90 (dd, $J = 8.2, 1.8$ Hz, 1H), 7.74 (dt, $J = 8.1, 0.8$ Hz, 1H), 7.26 – 7.12 (m, 5H), 6.01 (dd, $J = 7.8, 1.7$ Hz, 1H), 4.95 (dd, $J = 7.9, 1.7$ Hz, 1H), 4.12 (dd, $J = 7.8, 3.0$ Hz, 1H), 3.51 (dd, $J = 7.9, 3.0$ Hz, 1H), 3.27 (dd, $J = 9.4, 8.0$ Hz, 1H), 2.74 (s, 3H), 2.09 (dd, $J = 8.0, 4.2$ Hz, 1H), 1.64 (dd, $J = 9.4, 4.2$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3): δ 203.88, 166.33, 137.00, 132.85, 131.20 (q, $J_{\text{C-F}} = 3.4$ Hz), 130.99, 130.01 (q, $J_{\text{C-F}} = 32.4$ Hz), 129.37 (2C), 128.50, 128.03, 127.19 (2C), 125.93, 123.94 (q, $J_{\text{C-F}} = 270.9$ Hz), 118.95 (q, $J_{\text{C-F}} = 3.90$ Hz), 103.33, 99.37, 53.54, 45.61, 40.15, 33.19, 19.35. ^{19}F NMR (376 MHz, CDCl_3): δ 62.22.

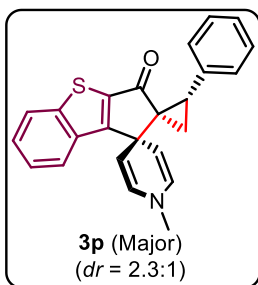
3o (Major): This compound was isolated as a pale-yellow semi-solid following general



procedure-6. 100 mg of **2o** afforded 40 mg of **3o** (52% yield) $R_f = 0.4$ (1:8 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2928, 1688, 1672, 1595, 1492, 1358, 1277, 1118, 1018, 872, 729. ^1H NMR (400 MHz, CDCl_3): δ 7.95–7.73 (m, 3H), 7.32–7.21 (m, 5H), 6.23 (dd, $J = 7.8, 1.8$ Hz, 1H), 6.12 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.29 (dd, $J = 7.8, 3.0$ Hz, 1H), 4.07 (dd, $J = 7.8, 2.9$ Hz, 1H), 3.04 (s, 4H), 2.15 (dd, $J = 8.3, 4.4$ Hz, 1H), 1.75 (dd, $J = 9.1, 4.4$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 201.71, 165.09, 135.89, 134.80, 131.00, 130.89 (q, $J_{\text{C-F}} = 3.5$ Hz), 130.36, 130.06 (q, $J_{\text{C-F}} = 32.5$ Hz), 129.24 (2C), 128.54, 127.84 (2C) 126.74, 123.92 (q, $J_{\text{C-F}} = 270.8$ Hz), 118.93 (q, $J_{\text{C-F}} = 3.95$ Hz), 101.08, 100.91, 53.42, 46.47, 40.52, 34.37, 19.17. ^{19}F NMR (376 MHz, CDCl_3): δ 62.28. HRMS (ESI): m/z calcd for $\text{C}_{23}\text{H}_{19}\text{F}_3\text{NNaO}$ ($\text{M}+\text{H}$) $^+$ 404.1238 found: 404.1228.

1''-Methyl-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-benzo[b]cyclopenta[d]thiophene-1',4''-pyridin]-3'-one (3p).

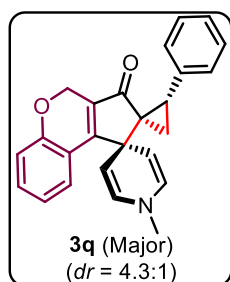
This compound was isolated as reddish-brown sticky oil by following the general procedure-6. 100 mg of **2p** afforded 37 mg of **3p** (49% yield, combined) $R_f = 0.4$ (1:7 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2928, 1689, 1671, 1599, 1568, 1494, 1447, 1348, 1229, 1041, 768, 747. ^1H NMR (500 MHz, CDCl_3): δ 8.05-7.99 (m, 1H), 7.90-7.85 (m, 1H), 7.47-7.40 (m, 2H), 7.31-7.25 (m, 4H), 7.20 (ddt, $J = 8.6, 7.2, 1.9$ Hz,



1H), 6.23 (dd, $J = 7.8, 1.8$ Hz, 1H), 6.16 (dd, $J = 7.8, 1.7$ Hz, 1H), 4.39 (dd, $J = 7.8, 3.0$ Hz, 1H), 4.15 (dd, $J = 7.8, 2.9$ Hz, 1H), 3.10 (s, 3H), 3.00 (t, $J = 8.8$ Hz, 1H), 2.07-2.04 (m, 1H), 1.74 (dd, $J = 9.3, 4.5$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 195.20, 167.72, 148.15, 138.34, 136.21, 134.71, 131.38, 131.20, 129.32 (2C), 127.80 (2C), 127.33, 126.54, 124.64, 124.61, 124.39, 100.94, 100.54, 56.81, 44.85, 40.69, 32.01, 17.31. **HRMS (ESI):** m/z calcd for $\text{C}_{24}\text{H}_{20}\text{NOS}$ ($\text{M}+\text{H}$)⁺ 370.1266 found: 370.1270.

1''-Methyl-2-phenyl-1''*H*,3''*H*,4''*H*-dispiro[cyclopropane-1,2'-cyclopenta[*c*]chromene-1',4''-pyridin]-3'-one (3q).

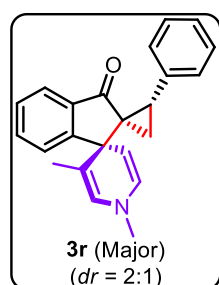
This compound was isolated as pale-yellow oil by following the general procedure-6. 80 mg



of **2q** afforded 48 mg of **3q** (78% yield, combined) $R_f = 0.4$ (1:8 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\text{max}}/\text{cm}^{-1}$ 2929, 2854, 1711, 1684, 1670, 1601, 1566, 1496, 1450, 1345, 1227, 1038, 993, 762, 750, 696. **^1H NMR (500 MHz, CDCl_3):** δ 7.81 (dd, $J = 7.7, 1.7$ Hz, 1H), 7.29-7.22 (m, 5H), 7.21-7.16 (m, 1H), 6.92 (td, $J = 7.6, 1.2$ Hz, 1H), 6.86 (dd, $J = 8.2, 1.2$ Hz, 1H), 6.16 (dd, $J = 7.9, 1.8$ Hz, 1H), 6.09 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.98 (d, $J_{AB} = 14.4$ Hz, 1H), 4.93 (d, $J_{AB} = 14.4$ Hz, 1H) 4.30 (dd, $J = 7.9, 3.0$ Hz, 1H), 4.07 (dd, $J = 7.9, 3.0$ Hz, 1H), 3.05 (s, 3H), 2.89 (t, $J = 8.7$ Hz, 1H), 1.92 (dd, $J = 8.2, 4.4$ Hz, 1H), 1.65 (dd, $J = 9.2, 4.4$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 200.39, 162.06, 156.93, 136.29, 132.05, 131.57, 131.45, 129.18 (2C), 128.42, 127.74 (2C), 126.90, 126.49, 121.01, 120.36, 116.73, 101.42, 100.77, 63.10, 53.81, 46.95, 40.64, 31.78, 17.09.

1'',3''-Dimethyl-2-phenyl-1''*H*,3''*H*-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3r).

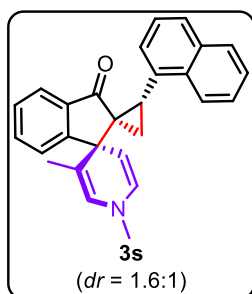
This compound was isolated as pale-yellow oil by following the general procedure-6. 80 mg



of **2r** afforded 39 mg of **3r** (65% yield, combined) $R_f = 0.4$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\text{max}}/\text{cm}^{-1}$ 2954, 2927, 1703, 1682, 1602, 1498, 1464, 1292, 1266, 1105, 1081, 765, 749, 700. **^1H NMR (500 MHz, CDCl_3):** δ 7.71 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.65-7.58 (m, 2H), 7.35 (ddd, $J = 7.6, 6.8, 1.4$ Hz, 1H), 7.20 (dq, $J = 6.9, 2.9, 2.5$ Hz, 2H), 7.18-7.11 (m, 3H), 6.01 (dd, $J = 7.7, 1.7$ Hz, 1H), 4.69 (p, $J = 1.3$ Hz, 1H) 4.07 (d, $J = 7.7$ Hz, 1H), 3.16 (dd, $J = 9.3, 7.9$ Hz, 1H), 2.72 (s, 3H), 1.96 (dd, $J = 7.9, 4.1$ Hz, 1H), 1.54 (dd, $J = 9.3, 4.1$ Hz, 1H), 0.72 (d, $J = 1.3$ Hz, 3H). **^{13}C NMR (125 MHz, CDCl_3):** δ 204.75, 162.47, 136.70, 134.71, 134.29, 130.26, 128.72 (2C), 127.34, 126.83 (3C), 126.08, 125.56, 121.44, 107.13, 99.21, 50.64, 48.62, 39.92, 31.70, 19.89, 16.50. **HRMS (ESI):** m/z calcd for $\text{C}_{23}\text{H}_{22}\text{NO}$ ($\text{M}+\text{H}$)⁺ 328.1701 found: 328.1711.

1'',3''-Dimethyl-2-(naphthalen-1-yl)-1''*H*,3''*H*-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3s).

This compound was isolated as pale-yellow sticky oil by following the general procedure-6.



100 mg of **2s** afforded 53 mg of **3s** (69% yield, combined) $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):**

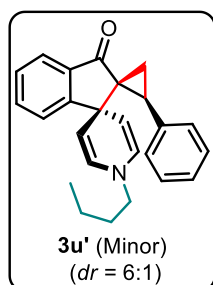
$\nu_{\max}/\text{cm}^{-1}$ 3053, 2920, 1698, 1682, 1600, 1463, 1293, 1264, 1240, 1084, 983, 776, 765. **3s (Major);- $^1\text{H NMR}$ (500 MHz, CDCl_3):** δ 8.23-8.21 (m, 1H), 7.79-7.75 (m, 2H), 7.67 (t, $J = 8.7$ Hz, 1H), 7.60-7.56 (m, 1H), 7.52 (dt, $J = 7.7, 1.0$ Hz, 1H), 7.43-7.22 (m, 3H), 7.16 (d, $J = 7.3$ Hz, 1H), 5.97 (dd, $J = 7.6, 1.7$ Hz, 1H), 4.28 (q, $J = 1.3$ Hz, 1H), 4.05 (d, $J = 7.6$ Hz, 1H), 3.91 (dd, $J = 9.3, 8.0$ Hz, 1H), 2.60 (s, 3H), 2.25 (dd, $J =$

7.9, 4.0 Hz, 1H), 1.62 (dd, $J = 9.3, 4.0$ Hz, 1H), 0.31 (d, $J = 1.3$ Hz, 3H). **$^{13}\text{C NMR}$ (125 MHz, CDCl_3):** δ 205.21, 162.20, 134.60, 134.51, 133.99, 133.45, 132.89, 130.40, 128.15, 127.32, 126.79, 125.97, 125.49, 125.28, 125.14, 124.51, 124.46, 123.73, 121.64, 101.45, 99.0, 52.83, 48.85, 39.95, 27.89, 19.64, 17.03.

3s' (Minor):- 7.89 (ddd, $J = 7.8, 1.9, 0.8$ Hz, 1H), 7.81-7.75 (m, 2H), 7.67 (t, $J = 8.7$ Hz, 1H), 7.60-7.56 (m, 1H), 7.48 (dt, $J = 7.7, 1.0$ Hz, 1H), 7.43-7.22 (m, 4H), 7.16 (d, $J = 7.3$ Hz, 1H), 5.75-5.74 (m, 1H), 4.26 (d, $J = 5$ Hz, 1H), 3.66 (t, $J = 8.7$ Hz, 1H), 2.98 (d, $J = 7.6$ Hz, 1H), 2.55 (s, 3H), 2.00 (dd, $J = 7.9, 4.0$ Hz, 1H), 1.95 (dd, $J = 9.3, 4.0$ Hz, 1H), 1.16 (d, $J = 1.2$ Hz, 3H). **$^{13}\text{C NMR}$ (125 MHz, CDCl_3):** δ 205.21, 162.20, 134.80, 134.63, 134.40, 132.8, 128.25, 128.21, 127.41, 127.32, 127.18, 126.79, 125.97, 125.49, 125.31, 125.28, 124.46, 124.24, 121.91, 107.72, 105.40, 50.68, 48.8, 39.8, 33.0, 19.7, 16.6.

1'-Butyl-2-phenyl-1'H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3u')

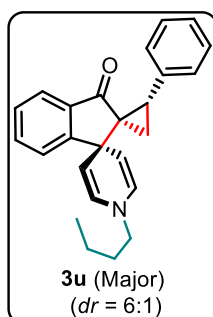
This compound was isolated as a pale-yellow semi-solid by following the general procedure-



6. 100 mg of **2u** afforded 11 mg of **3u'** (13% yield) $R_f = 0.6$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$

2955, 2863, 1699, 1669, 1599, 1497, 1461, 1321, 1029, 998, 766, 696. **3u' (Minor):- $^1\text{H NMR}$ (500 MHz, CDCl_3):** δ 7.70 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.64 (td, $J = 7.4, 1.2$ Hz, 1H), 7.56 (dt, $J = 7.7, 1.0$ Hz, 1H), 7.35 (td, $J = 7.4, 1.1$ Hz, 1H), 7.22-7.14 (m, 4H), 7.13-7.07 (m, 1H), 6.02 (dd, $J = 7.9, 1.8$ Hz, 1H), 5.04 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.11 (dd, $J = 7.9, 3.0$ Hz, 1H),

3.49 (dd, $J = 7.9, 3.0$ Hz, 1H), 3.17 (dd, $J = 9.3, 8.0$ Hz, 1H), 2.97-2.89 (m, 1H), 2.85 (q, $J = 7.1$ Hz, 1H), 1.99 (dd, $J = 8.0, 4.1$ Hz, 1H), 1.63-1.56 (m, 1H), 1.46-1.37 (m, 2H), 1.34-1.24 (m, 2H), 0.95 (t, $J = 7.3$ Hz, 3H). **$^{13}\text{C NMR}$ (125 MHz, CDCl_3):** δ 205.06, 164.24, 137.53, 134.84, 132.41, 129.78, 129.30 (2C), 127.68, 127.39, 127.29 (2C), 126.83, 125.65, 121.45, 103.98, 100.38, 52.93, 52.85, 45.60, 33.21, 32.11, 19.79, 19.07, 13.84.

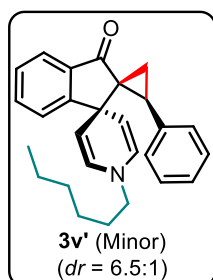


3u (Major):- This compound was isolated as greenish-yellow oil by following the general procedure-6. 100 mg of **2u** afforded 56 mg of **3u** (66% yield) $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). **$^1\text{H NMR}$ (500 MHz, CDCl_3):** δ 7.65-7.60 (m, 2H), 7.53 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.29 (ddd, $J = 7.6, 4.8, 3.4$ Hz, 1H), 7.27-7.23 (m, 4H), 7.17 (dt, $J = 5.2, 1.8$ Hz, 1H), 6.20 (dd, $J = 7.9, 1.8$ Hz, 1H), 6.09 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.27 (dd, $J = 7.8, 3.0$ Hz, 1H), 4.04 (dd, $J = 7.9, 2.9$ Hz,

1H), 3.14 (t, $J = 7.0$ Hz, 2H), 3.01-2.91 (m, 1H), 2.07 (dd, $J = 8.2, 4.3$ Hz, 1H), 1.67 (dd, $J = 9.1, 4.3$ Hz, 1H), 1.61-1.49 (m, 2H), 1.40-1.30 (m, 2H), 0.97 (t, $J = 7.4$ Hz, 3H). ^{13}C NMR (125 MHz, CDCl_3): δ 202.95, 162.76, 136.51, 134.67, 134.45, 129.78, 129.29 (2C), 129.18, 127.75 (2C), 127.74, 127.44, 126.51, 121.53, 101.67, 101.59, 53.25, 53.10, 46.52, 33.68, 32.28, 19.82, 18.70, 13.87. HRMS (ESI): m/z calcd for $\text{C}_{25}\text{H}_{26}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 356.2014 found: 356.2027.

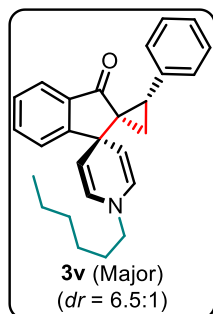
1''-Hexyl-2-phenyl-1''H,3''H-dispiro[cyclopropane-1,2''-indene-1',4''-pyridin]-3'-one (3v').

This compound was isolated as pale-yellow oil by following the general procedure-6. 80 mg



of **2v** afforded 7 mg of **3v'** (12% yield), $R_f = 0.6$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2952, 2926, 2857, 1699, 1671, 1600, 1498, 1461, 1375, 1147, 1114, 1007, 931, 724, 695. ^1H NMR (500 MHz, CDCl_3): δ 7.70 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.66-7.62 (m, 1H), 7.56 (dt, $J = 7.7, 1.0$ Hz, 1H), 7.37-7.32 (m, 1H), 7.27-7.24 (m, 1H), 7.18-7.10 (m, 4H), 6.02 (dd, $J = 7.9, 1.8$ Hz, 1H), 5.03 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.11 (dd, $J = 7.9, 3.0$ Hz, 1H), 3.49 (dd, $J = 7.9, 3.0$ Hz, 1H), 3.17 (dd, $J = 9.3, 8.0$ Hz, 1H), 2.94-2.89 (m, 1H), 2.86-2.80 (m, 1H), 1.99 (dd, $J = 8.0, 4.1$ Hz, 1H), 1.62-1.59 (m, 1H), 1.45-1.40 (m, 2H), 1.36-1.28 (m, 6H), 0.92 (t, $J = 7.0$ Hz, 3H). ^{13}C NMR (125 MHz, CDCl_3): δ 205.06, 164.23, 137.53, 134.82, 132.41, 129.78, 129.29 (2C), 127.68, 127.38, 127.29 (2C), 126.83, 125.65, 121.45, 103.98, 100.38, 53.27, 52.86, 45.61, 33.21, 31.56, 29.97, 26.29, 22.64, 19.07, 14.05.

3v (Major):- This compound was isolated as brownish-yellow oil by following the general

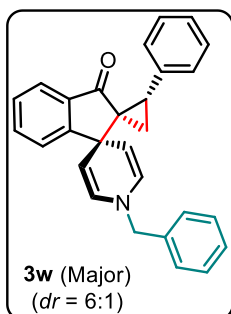


procedure-6. 80 mg of **2v** afforded 41 mg of **3v** (66% yield), $R_f = 0.5$ (1:9 EtOAc: Hexanes, visualized by 254 nm UV light). ^1H NMR (500 MHz, CDCl_3): δ 7.64-7.61 (m, 2H), 7.53 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.32-7.27 (m, 1H), 7.27-7.23 (m, 4H), 7.19-7.15 (m, 1H), 6.20 (dd, $J = 7.8, 1.8$ Hz, 1H), 6.09 (dd, $J = 7.9, 1.7$ Hz, 1H), 4.27 (dd, $J = 7.8, 2.9$ Hz, 1H), 4.04 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.14 (t, $J = 7.0$ Hz, 2H), 3.00-2.93 (m, 1H), 2.07 (dd, $J = 8.2, 4.3$ Hz, 1H), 1.67 (dd, $J = 9.1, 4.3$ Hz, 1H), 1.57 (t, $J = 7.0$ Hz, 2H), 1.33 (qd, $J = 5.3, 3.6, 3.0$ Hz, 6H), 0.94-0.88 (m, 3H). ^{13}C NMR (125

MHz, CDCl_3): δ 202.95, 162.76, 136.51, 134.66, 134.43, 129.76, 129.28 (2C), 129.18, 127.74 (2C), 127.72, 127.43, 126.50, 121.53, 101.65, 101.58, 53.55, 53.06, 46.51, 33.68, 31.52, 30.12, 26.28, 22.64, 18.70, 14.04. HRMS (ESI): m/z calcd for $\text{C}_{27}\text{H}_{30}\text{NO}$ ($\text{M}+\text{H}$) $^+$ 384.2327 found: 384.2339.

1''-Benzyl-2-phenyl-1''H,3''H-dispiro[cyclopropane-1,2''-indene-1',4''-pyridin]-3'-one (3w).

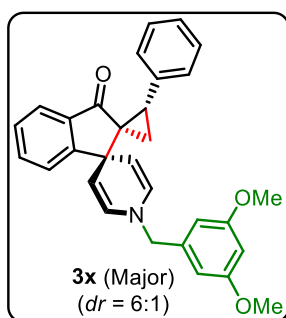
This compound was isolated as reddish-orange oil by following the general procedure-6. 70 mg of **2w** afforded 45 mg of **3w** (75% yield, combined), $R_f = 0.3$ (1:8 EtOAc: Hexanes, visualized by 254 nm UV light). IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 3085, 3062, 3027, 1700, 1670, 1601, 1496, 1403, 1324, 1209, 1021, 766, 733. ^1H NMR (400 MHz, CDCl_3): δ 7.72-7.65 (m, 2H), 7.59 (dt, $J = 7.9, 1.1$ Hz, 1H), 7.44 (dd, $J = 8.1, 6.7$ Hz, 2H), 7.36 (ddd, $J = 7.9,$



5.7, 2.1 Hz, 2H), 7.33-7.28 (m, 6H), 7.23 (ddd, $J = 8.4, 3.9, 2.3$ Hz, 1H), 6.33 (dd, $J = 7.9, 1.8$ Hz, 1H), 6.22 (dd, $J = 7.9, 1.7$ Hz, 1H), 4.40 (d, $J = 8.0$ Hz, 3H), 4.17 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.04 (t, $J = 8.6$ Hz, 1H), 2.14 (dd, $J = 8.2, 4.3$ Hz, 1H), 1.74 (dd, $J = 9.1, 4.3$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 202.89, 162.48, 138.36, 136.36, 134.72, 134.53, 130.10, 129.49 (2C), 129.32, 128.84 (2C), 127.79 (2C), 127.72, 127.67, 127.58, 127.01 (2C), 126.58, 121.64, 102.67, 102.56, 57.09, 52.90, 46.34, 33.70, 18.70. HRMS (ESI): m/z calcd for $\text{C}_{28}\text{H}_{23}\text{NNaO}$ ($\text{M}+\text{Na}^+$) 412.1677 found 412.1660.

1''-(3,5-Dimethoxybenzyl)-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3x).

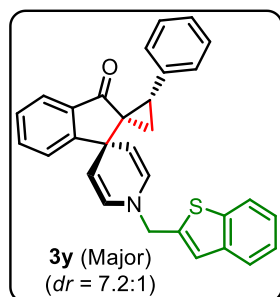
This compound was isolated as pale-yellow solid by following the general procedure-6. 75 mg of **2x** afforded 51 mg of **3x** (78% yield, combined), M.P = 94-96 °C. $R_f = 0.4$ (1:4 EtOAc: Hexanes, visualized by 254 nm UV light).



IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2955, 2923, 2853, 16999, 1671, 1596, 1462, 1429, 1402, 1321, 1204, 1155, 1064, 1021, 1010, 766, 733, 696. ^1H NMR (400 MHz, CDCl_3): δ 7.73-7.65 (m, 2H), 7.59 (dt, $J = 7.6, 1.0$ Hz, 1H), 7.39-7.33 (m, 1H), 7.31 (d, $J = 4.4$ Hz, 4H), 7.26-7.20 (m, 1H), 6.44 (s, 3H), 6.32 (dd, $J = 7.9, 1.8$ Hz, 1H), 6.21 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.40 (dd, $J = 7.8, 2.9$ Hz, 1H), 4.34 (s, 2H), 4.18 (dd, $J = 7.9, 2.9$ Hz, 1H), 3.85 (s, 6H), 3.04 (t, $J = 8.6$ Hz, 1H), 2.14 (dd, $J = 8.2, 4.3$ Hz, 1H), 1.74 (dd, $J = 9.1, 4.3$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 202.79, 162.45, 161.24 (2C), 140.99, 136.32, 134.73, 134.50, 130.13, 129.54 (2C), 129.29 (2C), 127.79, 127.72, 127.59, 126.59, 121.65, 104.79 (2C), 102.56, 102.48, 99.35, 57.08, 55.39, 55.36, 52.83, 46.32, 33.70, 18.72. HRMS (ESI): m/z calcd for $\text{C}_{30}\text{H}_{28}\text{NO}_3$ ($\text{M}+\text{H}^+$) 450.2069 found 450.2086.

1''-(Benzo[*b*]thiophen-2-ylmethyl)-2-phenyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3y).

This compound was isolated as reddish-brown oil by following the general procedure-6. 70 mg of **2y** afforded 42 mg of **3y** (70% yield, combined) $R_f = 0.4$ (1:7 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\text{max}}/\text{cm}^{-1}$ 3052, 2920, 2854, 1698, 1671, 1601, 1461, 1399,

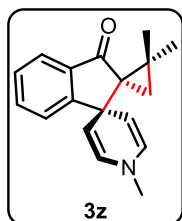


1322, 1206, 1190, 1018, 766, 748. ^1H NMR (400 MHz, CDCl_3): δ 7.84 (dd, $J = 8.0, 1.2$ Hz, 1H), 7.75 (dd, $J = 6.8, 2.0$ Hz, 1H), 7.65-7.63 (m, 2H), 7.55 (d, $J = 7.6$ Hz, 1H), 7.39-7.32 (m, 3H), 7.27 (d, $J = 4.4$ Hz, 4H), 7.21-7.18 (m, 2H), 6.33 (dd, $J = 8.0, 1.6$ Hz, 1H), 6.22 (dd, $J = 8.0, 1.6$ Hz, 1H), 4.59 (s, 2H), 4.42 (dd, $J = 8.0, 2.8$ Hz, 1H), 4.19 (dd, $J = 8.0, 2.8$ Hz, 1H), 3.01 (t, $J = 8.4$ Hz, 1H), 2.10 (dd, $J = 8.4, 4.0$ Hz, 1H), 1.71 (dd, $J = 9.2, 4.4$ Hz, 1H). ^{13}C NMR (125 MHz, CDCl_3): δ 202.73, 162.20, 142.40, 139.86, 139.47, 136.27, 134.75, 134.53, 129.38, 129.30 (2C), 128.77, 127.76 (2C), 127.73, 127.62,

126.58, 124.54, 124.48, 123.49, 122.53, 122.02, 121.67, 103.60, 103.47, 53.08, 52.65, 46.20, 33.70, 18.70. **HRMS (ESI):** m/z calcd for $C_{30}H_{24}NOS$ ($M+H^+$) 446.1579 found 446.1566.

1'',2,2-Trimethyl-1''H,3'H-dispiro[cyclopropane-1,2'-indene-1',4''-pyridin]-3'-one (3z).

This compound was isolated as a pale-yellow semi-solid by following the general procedure-



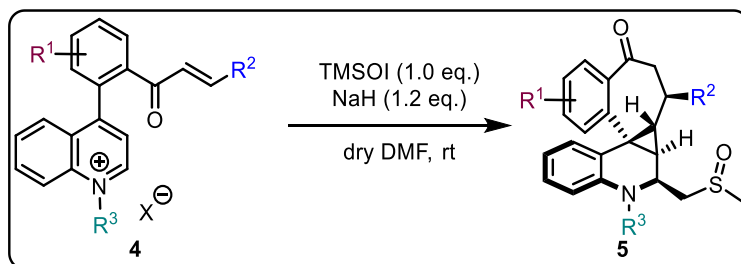
6. 60 mg of **2z** afforded 37 mg of **3z** (88% yield) $R_f = 0.4$ (1:9 EtOAc:

Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 3054, 1699, 1676, 1600, 1498, 1406, 1293, 1111, 1006, 929, 757, 725. **¹H**

NMR (400 MHz, CDCl₃): δ 7.70-7.59 (m, 3H), 7.38-7.31 (m, 1H), 6.05 (dd, $J = 7.9, 1.7$ Hz, 1H), 5.79 (dd, $J = 7.9, 1.8$ Hz, 1H), 4.25 (dd, $J = 7.9, 3.0$ Hz, 1H), 4.09 (dd, $J = 7.9, 2.9$ Hz, 1H), 2.99 (s, 3H), 1.57 (s, 3H), 1.40 (s, 3H),

1.30 (d, $J = 3.3$ Hz, 1H), 1.19 (d, $J = 3.3$ Hz, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 206.26, 163.16, 134.32, 133.50, 130.65, 128.35, 127.40, 127.28, 121.25, 105.80, 100.99, 51.99, 46.21, 40.52, 30.30, 30.06, 25.01, 20.01. **HRMS (ESI):** m/z calcd for $C_{18}H_{20}NO$ ($M+H^+$)⁺ 266.1545 found: 266.1549.

General procedure-7: Synthesis of the polycyclic benzocycloheptanones (5a-5m)



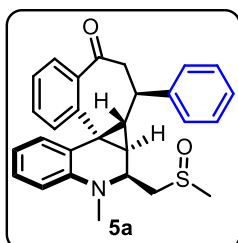
Scheme S7: Synthesis of the polycyclic benzocycloheptanones **5a-5m**

A mixture of sodium hydride (60% in oil, 10 mg, 0.28 mmol) and TMSOI (46 mg, 0.23 mmol) was placed in an oven-dried flask, and dry DMF (3.0 mL) was added to the mixture. After the hydrogen evolution ceased and the milky solution turned clear, the reaction mixture was stirred for 15 min. Then **4a** (100 mg, 0.21 mmol) was dissolved in dry DMF (1.0 mL), added to the clear solution drop wise over 5-10 min, and stirred at room temperature until **4a** disappeared as monitored by TLC. The reaction mixture was quenched using ice water and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate to afford **5a**.

Spectral data of all the polycyclic benzocycloheptanones reported in this study

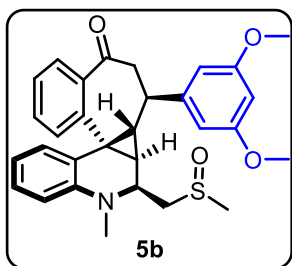
1-Methyl-2-((methylsulfinyl)methyl)-3-phenyl-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (**5a**).

This compound was isolated as pale-yellow solid by following the general procedure-7. 100 mg of **4a** afforded 72 mg of **5a** (78% yield), M.P = 225-227 °C. R_f = 0.3 (9:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3028, 2920, 1679, 1598, 1497, 1282, 1230, 1050, 750. **^1H NMR (400 MHz, CDCl_3):** δ 7.57 (ddd, J = 8.9, 7.1, 1.8 Hz, 2H), 7.51-7.45 (m, 1H), 7.42 (dd, J = 7.8, 1.4 Hz, 1H), 7.33 (dd, J = 8.1, 6.5 Hz, 2H), 7.28-7.23 (m, 1H), 7.24-7.19 (m, 2H), 7.08 (ddd, J = 8.6, 7.3, 1.7 Hz, 1H), 6.71 (dd, J = 8.3, 1.1 Hz, 1H), 6.60 (td, J = 7.4, 1.1 Hz, 1H), 6.48 (dd, J = 7.7, 1.7 Hz, 1H), 3.93 (ddd, J = 10.9, 4.4, 2.7 Hz, 1H), 3.30-3.14 (m, 2H), 2.86 (d, J = 16.3 Hz, 5H), 2.43 (t, J = 4.8 Hz, 1H), 2.15-2.02 (m, 4H), 1.87 (dd, J = 10.5, 5.2 Hz, 1H). **^{13}C NMR (100 MHz, CDCl_3):** δ 206.19, 142.71, 142.69, 140.85, 139.31, 132.75, 132.08, 128.96 (2C), 128.25, 128.01, 127.94, 127.41 (2C), 27.15, 127.01, 126.54, 118.29, 112.48, 61.27, 49.65, 48.91, 44.75, 39.01, 35.93, 35.17, 34.66, 31.06. **HRMS (ESI):** m/z calcd for $\text{C}_{28}\text{H}_{28}\text{NO}_2\text{S}$ ($\text{M}+\text{H}^+$) 442.1841 found: 442.1832



3-(3,5-Dimethoxyphenyl)-1-methyl-2-((methylsulfinyl)methyl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (**5b**).

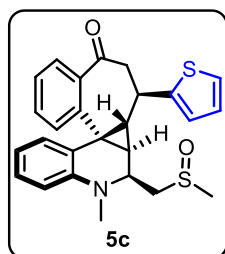
This compound was isolated as an off-white solid by following the general procedure-7. 80 mg of **4b** afforded 55 mg of **5b** (74% yield), M.P = 102-105 °C. R_f = 0.1 (10:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2928, 2871, 2850,



1679, 1595, 1483, 1362, 1204, 1057, 845, 830, 750. **¹H NMR (400 MHz, CDCl₃):** δ 7.59-7.53 (m, 2H), 7.53-7.46 (m, 1H), 7.42 (d, *J* = 7.6 Hz, 1H), 7.11-7.05 (m, 1H), 6.71 (d, *J* = 8.3 Hz, 1H), 6.60 (t, *J* = 7.4 Hz, 1H), 6.46 (dd, *J* = 7.6, 1.7 Hz, 1H), 6.36 (s, 3H), 3.94 (dt, *J* = 11.2, 3.1 Hz, 1H), 3.78 (s, 6H), 3.30-3.18 (m, 2H), 2.89 (s, 3H), 2.80-2.70 (m, 1H), 2.43 (t, *J* = 4.7 Hz, 1H), 2.23 (s, 3H), 2.15 (t, *J* = 11.5 Hz, 1H), 1.84 (dd, *J* = 10.8, 5.2 Hz, 1H), 1.68 (d, *J* = 4.7 Hz, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 205.97, 161.12 (2C), 145.09, 142.75, 140.80, 139.35, 132.77, 132.14, 128.24, 128.08, 127.89, 127.00, 126.56, 118.27, 112.50, 105.24 (2C), 98.69, 61.57, 55.39, 55.37, 49.64, 48.45, 44.90, 38.95, 35.90, 35.20, 34.97, 30.99. **HRMS (ESI):** *m/z* calcd for C₃₀H₃₂NO₄S (M+H⁺) 502.2052 found: 502.2040.

1-Methyl-2-((methylsulfinyl)methyl)-3-(thiophen-2-yl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (5c).

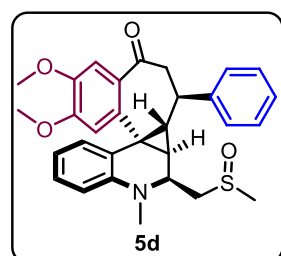
This compound was isolated as a reddish-brown solid by following the general procedure-7.



80 mg of **4c** afforded 55 mg of **5c** (69% yield), M.P = 190-193 °C. *R_f* = 0.3 (4:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** *v*_{max}/cm⁻¹ 2928, 2853, 1674, 1595, 1494, 1361, 1276, 1041, 963, 764, 701. **¹H NMR (400 MHz, CDCl₃):** δ 7.56 (ddd, *J* = 15.2, 7.6, 1.6 Hz, 2H), 7.48 (td, *J* = 7.5, 1.3 Hz, 1H), 7.42 (dd, *J* = 7.7, 1.3 Hz, 1H), 7.20 (dd, *J* = 5.1, 1.2 Hz, 1H), 7.08 (ddd, *J* = 8.6, 7.3, 1.7 Hz, 1H), 6.97 (dd, *J* = 5.1, 3.4 Hz, 1H), 6.87 (dd, *J* = 3.5, 1.2 Hz, 1H), 6.72 (dd, *J* = 8.4, 1.1 Hz, 1H), 6.61 (td, *J* = 7.5, 1.1 Hz, 1H), 6.46 (dd, *J* = 7.6, 1.7 Hz, 1H), 4.01-3.93 (m, 1H), 3.34-3.27 (m, 1H), 3.27-3.13 (m, 2H), 2.97 (d, *J* = 15.6 Hz, 1H), 2.91 (s, 3H), 2.45 (t, *J* = 4.8 Hz, 1H), 2.36 (dd, *J* = 12.3, 10.8 Hz, 1H), 2.30 (s, 3H), 1.92 (dd, *J* = 10.2, 5.1 Hz, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 205.33, 146.03, 142.84, 140.62, 139.15, 132.86, 132.09, 128.33, 128.01, 127.94, 127.08 (2C), 126.45, 124.05, 123.65, 118.42, 112.60, 61.30, 50.30, 49.86, 39.96, 39.36, 35.89, 35.74, 34.89, 30.94. **HRMS (ESI):** *m/z* calcd for C₂₆H₂₆NO₂S₂ (M+H)⁺ 448.1405 found: 448.1394.

7,8-dimethoxy-1-methyl-2-((methylsulfinyl)methyl)-3-phenyl-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (5d).

This compound was isolated as a reddish-brown semi-solid by following the general

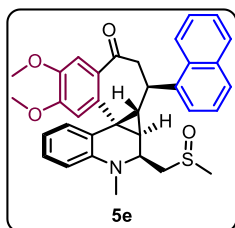


procedure-7. 100 mg of **4d** afforded 71 mg of **5d** (76% yield) *R_f* = 0.3 (10:0 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** *v*_{max}/cm⁻¹ 2930, 2831, 1712, 1674, 1595, 1498, 1445, 1265, 1205, 1171, 1035, 884, 747. **¹H NMR (500 MHz, CDCl₃):** δ 7.31 (dd, *J* = 8.0, 6.7 Hz, 2H), 7.26-7.22 (m, 1H), 7.22-7.18 (m, 2H), 7.13 (s, 1H), 7.07 (ddd, *J* = 8.5, 7.3, 1.7 Hz, 1H), 6.77 (s, 1H), 6.69 (dd, *J* = 8.4, 1.1 Hz, 1H), 6.59 (td, *J* = 7.5, 1.1 Hz, 1H), 6.49 (dd, *J* = 7.7, 1.7 Hz, 1H), 3.96 (s, 3H), 3.94-3.91 (m, 1H), 3.89 (s, 3H), 3.27-3.14 (m, 2H), 2.87 (s, 3H), 2.83 (dt, *J* = 16.2, 2.7 Hz, 2H), 2.40 (dd, *J* = 5.2, 4.4 Hz, 1H), 2.06 (s, 4H), 1.81 (dd, *J* = 10.5, 5.3 Hz, 1H). **¹³C NMR (125 MHz, CDCl₃):** δ 204.49, 152.98, 148.82, 143.02, 142.66,

133.33, 128.93 (2C), 127.80, 127.38 (2C), 127.08, 126.94, 126.88, 118.22, 113.69, 112.38, 110.84, 61.66, 56.24, 56.05, 49.68, 49.08, 44.75, 39.13, 36.06, 36.02, 34.54, 31.02. **HRMS (ESI):** m/z calcd for C₃₀H₃₂NO₄S (M+H)⁺ 502.2052 found: 502.2067.

7,8-dimethoxy-1-methyl-2-((methylsulfinyl)methyl)-3-(naphthalen-1-yl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5b(2bH)-one (5e).

This compound was isolated as greenish-yellow stick oil by following the general procedure-

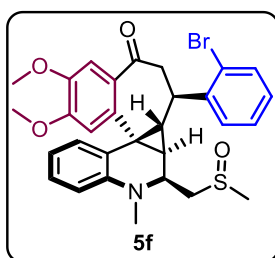


7. 80 mg of **4e** afforded 55 mg of **5e** (73% yield) $R_f = 0.2$ (9:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2925, 2850, 1710, 1664, 1597, 1511, 1363, 1263, 1212, 1038, 750. **¹H NMR (400 MHz, CDCl₃):** δ 7.91-7.88 (m, 2H), 7.75 (dd, $J = 7.3, 2.0$ Hz, 1H), 7.54-7.43 (m, 4H), 7.24 (s, 1H), 7.08 (ddd, $J = 8.3, 7.3, 1.7$ Hz, 1H), 6.83 (s, 1H), 6.70 (dd, $J = 8.4, 1.1$ Hz, 1H), 6.62 (td, $J = 7.4, 1.1$

Hz, 1H), 6.56 – 6.47 (m, 1H), 4.02 (s, 3H), 3.95 (s, 3H), 3.85-3.82 (m, 2H), 3.49-3.45 (m, 1H), 3.06-2.84 (m, 2H), 2.84 (s, 3H), 2.51 (t, $J = 4.7$ Hz, 1H), 1.97 (br s, 1H), 1.81 (t, $J = 11.5$ Hz, 1H), 1.53 (s, 3H). **¹³C NMR (125 MHz, CDCl₃):** δ 204.28, 153.20, 148.90, 142.81, 139.61, 133.92, 133.69, 132.64, 130.80, 128.99, 127.76, 127.22, 126.94 (2C), 126.67, 126.09, 125.82 (2C), 123.79, 122.86, 118.33, 113.94, 112.55, 111.06, 61.92, 56.29, 56.08, 49.68, 49.39, 38.47, 37.30, 36.34, 35.93, 35.78, 31.06. **HRMS (ESI):** m/z calcd for C₃₄H₃₄NO₄S (M+H)⁺ 552.2209 found: 552.2200.

3-(2-bromophenyl)-7,8-dimethoxy-1-methyl-2-((methylsulfinyl)methyl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5b(2bH)-one (5f).

This compound was isolated as reddish-brown solid by following the general procedure-7. 80

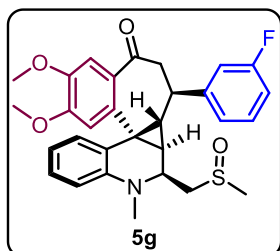


mg of **4f** afforded 58 mg of **5f** (77% yield), M.P = 217-219 °C. $R_f = 0.3$ (4:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2929, 2856, 1710, 1665, 1597, 1511, 1497, 1467, 1361, 1262, 1212, 1171, 1035, 874, 751, 731. **¹H NMR (500 MHz, CDCl₃):** δ 7.57 (dd, $J = 8.0, 1.2$ Hz, 1H), 7.34-7.27 (m, 2H), 7.16 (s, 1H), 7.11 (ddd, $J = 8.0, 6.8, 2.2$ Hz, 1H), 7.07 (ddd, $J = 8.3, 7.3, 1.7$ Hz, 1H), 6.77 (s, 1H), 6.70 (dd, $J = 8.3, 1.1$ Hz, 1H), 6.60 (td,

$J = 7.4, 1.1$ Hz, 1H), 6.49 (dd, $J = 7.6, 1.7$ Hz, 1H), 3.97 (s, 3H), 3.96-3.92 (m, 1H), 3.90 (s, 3H), 3.56 (t, $J = 11.6$ Hz, 1H), 3.21 (dt, $J = 17.8, 7.0$ Hz, 2H), 2.89 (s, 3H), 2.81 (dd, $J = 18.7, 2.5$ Hz, 1H), 2.54 (t, $J = 4.9$ Hz, 1H), 2.16 (s, 3H), 1.70 (s, 2H). **¹³C NMR (125 MHz, CDCl₃):** δ 203.92, 153.05, 148.79, 142.72, 142.32, 133.40, 133.19, 132.51, 128.42, 128.21, 128.07, 127.96, 127.01, 126.97, 124.20 118.33, 113.74, 112.50, 110.88, 61.80, 56.22, 56.02, 50.07, 48.63, 42.29 39.23, 36.15, 34.50, 30.67. **HRMS (ESI):** m/z calcd for C₃₀H₃₂BrNO₄S (M+H)⁺ 580.1157 found: 580.1174.

3-(3-Fluorophenyl)-7,8-dimethoxy-1-methyl-2-((methylsulfinyl)methyl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (5g).

This compound was isolated as an off-white solid by following the general procedure-7. 110

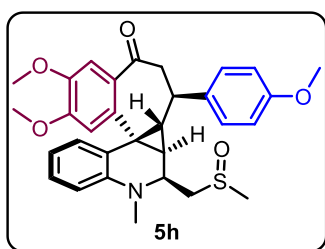


mg of **4g** afforded 72 mg of **5g** (70% yield), M.P = 242-245 °C. R_f = 0.3 (8:2 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3004, 2960, 2932, 2915, 2837, 1667, 1598, 1512, 1463, 1362, 1260, 1212, 1176, 1052, 1035, 750. **^1H NMR (500 MHz, CDCl_3):** δ 7.30 (td, J = 7.9, 5.9 Hz, 1H), 7.13 (s, 1H), 7.07 (ddd, J = 8.3, 7.3, 1.7 Hz, 1H), 7.00 (dt, J = 7.7, 1.3 Hz, 1H), 6.98-6.90 (m, 2H), 6.77 (s, 1H), 6.70 (dd, J = 8.4, 1.1 Hz, 1H), 6.60 (td, J

= 7.4, 1.1 Hz, 1H), 6.48 (dd, J = 7.7, 1.7 Hz, 1H), 3.96 (s, 3H), 3.95-3.92 (m, 1H), 3.89 (s, 3H), 3.28-3.18 (m, 2H), 2.88 (s, 3H), 2.86-2.82 (m, 1H), 2.44 (dd, J = 5.2, 4.3 Hz, 1H), 2.20 (s, 3H), 2.07 (dd, J = 12.3, 10.7 Hz, 1H), 1.82-1.75 (m, 2H). **^{13}C NMR (125 MHz, CDCl_3):** δ 203.87, 162.95 (d, $J_{\text{C-F}}$ = 245.8 Hz), 153.09, 148.88, 145.67 (d, $J_{\text{C-F}}$ = 6.7 Hz), 142.73, 133.26, 132.61, 130.50 (d, $J_{\text{C-F}}$ = 8.2 Hz), 127.81, 127.03, 126.86, 122.91 (d, $J_{\text{C-F}}$ = 2.8 Hz), 118.37, 114.32 (d, $J_{\text{C-F}}$ = 21.1 Hz), 113.78 (d, $J_{\text{C-F}}$ = 20.8 Hz), 113.71, 112.53, 110.87, 61.27, 56.26, 56.06, 49.69, 48.69, 44.37, 39.06, 35.95, 35.75, 34.68, 30.85. **^{19}F NMR (376 MHz, CDCl_3):** δ -111.86. **HRMS (ESI):** m/z calcd for $\text{C}_{30}\text{H}_{31}\text{FNO}_4\text{S}$ ($\text{M}+\text{H}^+$) 520.1958 found: 520.1971.

(7,8-dimethoxy-3-(4-methoxyphenyl)-1-methyl-2-((methylsulfinyl)methyl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (5h).

This compound was isolated as an off-white solid by following the general procedure-7. 100

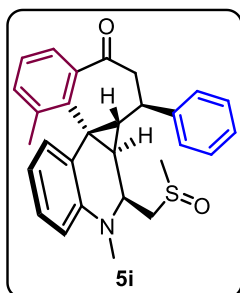


mg of **4h** afforded 72 mg of **5h** (77% yield), M.P = 242-244 °C. R_f = 0.2 (10:0 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2927, 2850, 1664, 1596, 1505, 1448, 1362, 1211, 1146, 1018, 743. **^1H NMR (500 MHz, CDCl_3):** δ 7.11 (dd, J = 7.0, 1.7 Hz, 3H), 7.06 (ddd, J = 8.2, 7.2, 1.7 Hz, 1H), 6.88-6.81 (m, 2H), 6.76 (s, 1H), 6.69 (dd, J = 8.4, 1.1 Hz, 1H), 6.59 (td, J = 7.4, 1.1 Hz, 1H), 6.48 (dd, J = 7.6, 1.7

Hz, 1H), 3.96 (s, 3H), 3.92 (ddd, J = 10.9, 4.4, 2.8 Hz, 1H), 3.89 (s, 3H), 3.78 (s, 3H), 3.26-3.14 (m, 2H), 2.87 (s, 3H), 2.84-2.74 (m, 2H), 2.39 (dd, J = 5.3, 4.4 Hz, 1H), 2.13 (s, 3H), 2.08 (dd, J = 12.2, 10.7 Hz, 1H), 1.77 (dd, J = 10.6, 5.3 Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 204.67, 158.64, 152.95, 148.79, 142.67, 135.04, 133.37, 132.90, 128.28 (2C), 127.77, 126.96, 126.90, 118.21, 114.21 (2C), 113.69, 112.37, 110.82, 61.61, 56.23, 56.04, 55.38, 49.71, 49.36, 43.87, 39.18, 36.22, 36.01, 34.57, 30.99. **HRMS (ESI):** m/z calcd for $\text{C}_{31}\text{H}_{34}\text{NO}_5\text{S}$ ($\text{M}+\text{H}^+$) 532.2158 found: 532.2172.

1,9-Dimethyl-2-((methylsulfinyl)methyl)-3-phenyl-1,2,2a,2b,3,4-hexahydro-5H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5-one (5i).

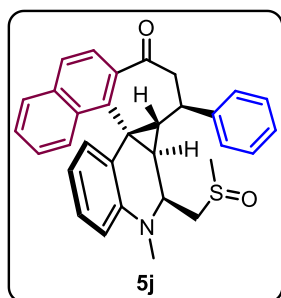
This compound was isolated as pale-yellow oil by following the general procedure-7. 75 mg



of **4i** afforded 49 mg of **5i** (71% yield) $R_f = 0.3$ (4:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2960, 2927, 1676, 1598, 1493, 1270, 1220, 1041, 764. **^1H NMR (500 MHz, CDCl_3):** δ 7.40-7.34 (m, 2H), 7.31 (ddd, $J = 7.6, 6.3, 1.8$ Hz, 3H), 7.25 (d, $J = 7.8$ Hz, 1H), 7.22-7.19 (m, 2H), 7.05 (ddd, $J = 8.3, 7.2, 1.8$ Hz, 1H), 6.67 (dd, $J = 8.3, 1.1$ Hz, 1H), 6.56 (td, $J = 7.4, 1.1$ Hz, 1H), 6.51 (dd, $J = 7.7, 1.8$ Hz, 1H), 3.96 (ddd, $J = 10.9, 4.5, 2.6$ Hz, 1H), 3.22-3.13 (m, 2H), 2.87 (s, 3H), 2.84-2.80 (m, 1H), 2.80-2.76 (m, 1H), 2.22 (dd, $J = 5.4, 4.5$ Hz, 1H), 2.18 (s, 3H), 2.08-2.01 (m, 4H), 1.84 (dd, $J = 10.8, 5.3$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 207.08, 142.77, 142.66, 142.18, 139.27, 136.08, 134.27, 128.96 (2C), 128.28, 127.49 (2C), 127.14, 127.11, 127.03, 125.63, 124.90, 118.27, 112.42, 61.42, 49.53, 48.99, 44.72, 39.17, 36.09, 36.03, 34.61, 29.71, 20.37. **HRMS (ESI):** m/z calcd for $\text{C}_{29}\text{H}_{30}\text{NO}_2\text{S}$ ($\text{M}+\text{H}$)⁺ 456.1997 found: 456.1972.

5-Methyl-6-((methylsulfinyl)methyl)-7-phenyl-6,6a,7,8-tetrahydro-5H naphtho[2'',1'':6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-9(6bH)-one (**5j**).

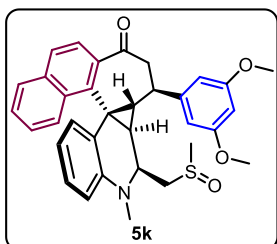
This compound was isolated as a pale-yellow solid by following the general procedure-7.



85mg of **4j** afforded 53 mg of **5j** (68% yield), M.P = 240-243 °C. $R_f = 0.4$ (4:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2927, 2853, 1709, 1676, 1597, 1495, 1453, 1398, 1274, 1044, 1029, 815, 762, 701. **^1H NMR (500 MHz, CDCl_3):** δ 8.07 (dd, $J = 8.5, 1.1$ Hz, 1H), 7.95 (dd, $J = 8.6, 0.8$ Hz, 1H), 7.90- 7.88 (m, 1H), 7.54-7.51 (m, 2H), 7.48 (ddd, $J = 8.3, 6.8, 1.4$ Hz, 1H), 7.32-7.29 (m, 2H), 7.26-7.24 (m, 1H), 7.20-7.18 (m, 2H), 7.03 (ddd, $J = 8.3, 7.3, 1.7$ Hz, 1H), 6.72 (dd, $J = 8.3, 1.2$ Hz, 1H), 6.56 (dd, $J = 7.7, 1.7$ Hz, 1H), 6.48 (td, $J = 7.4, 1.1$ Hz, 1H), 4.26-4.22 (m, 1H), 3.32-3.26 (m, 1H), 3.18 (dd, $J = 12.4, 2.6$ Hz, 1H), 2.99 (s, 3H), 2.94 (dd, $J = 18.5, 3.5$ Hz, 1H), 2.85 (ddd, $J = 13.0, 10.8, 3.5$ Hz, 1H), 2.43 (t, $J = 5.2$ Hz, 1H), 2.15-2.10 (m, 1H), 1.99 (s, 3H), 1.91 (dd, $J = 10.8, 5.5$ Hz, 1H). **^{13}C NMR (125 MHz, CDCl_3):** δ 207.37, 142.46, 142.23, 138.83, 135.59, 134.78, 131.79, 129.22, 129.02 (2C), 128.78, 127.69, 127.52 (2C), 127.43, 127.39, 127.22, 127.19, 126.10, 125.68, 124.42, 118.27, 112.60, 61.26, 50.10, 49.13, 44.78, 39.17, 36.35, 36.20, 34.34, 28.96. **HRMS (ESI):** m/z calcd for $\text{C}_{32}\text{H}_{30}\text{NO}_2\text{S}$ ($\text{M}+\text{H}$)⁺ 492.1997 found: 492.1982.

7-(3,5-Dimethoxyphenyl)-5-methyl-6-((methylsulfinyl)methyl)-5,6,6a,6b,7,8-hexahydro-9H-naphtho[2'',1'':6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-9-one (**5k**).

This compound was isolated as a pale-yellow oil by following the general procedure-7. 80

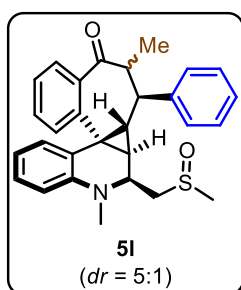


mg of **4k** afforded 49 mg of **5k** (65% yield), M.P = 240-243 °C. $R_f = 0.2$ (9:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 2927, 2854, 1666, 1595, 1504, 1447, 1360, 1218, 1140, 1011, 760. **^1H NMR (400 MHz, CDCl_3):** δ 8.08 (dd, $J = 8.5, 1.2$ Hz, 1H), 7.97 (dd, $J = 8.5, 4.1$ Hz, 1H), 7.94-7.90 (m, 1H),

7.59-7.54 (m, 1H), 7.52-7.47 (m, 2H), 7.06 (ddd, $J = 8.6, 7.2, 1.8$ Hz, 1H), 6.75 (dd, $J = 8.4, 1.1$ Hz, 1H), 6.56 (dd, $J = 7.6, 1.7$ Hz, 1H), 6.53-6.46 (m, 1H), 6.35 (s, 3H), 4.28 (ddd, $J = 11.0, 4.9, 2.5$ Hz, 1H), 3.76 (s, 6H), 3.38-3.20 (m, 2H), 3.02 (s, 3H), 2.97 (dd, $J = 18.5, 3.5$ Hz, 1H), 2.82 (ddd, $J = 13.0, 10.7, 3.5$ Hz, 1H), 2.46 (t, $J = 5.1$ Hz, 1H), 2.25 (dd, $J = 12.4, 10.8$ Hz, 1H), 2.18 (s, 3H), 1.91 (dd, $J = 10.8, 5.5$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3): δ 207.33, 161.18 (2C), 144.79, 142.26, 138.80, 135.58, 134.75, 131.76, 129.24, 128.79, 127.74, 127.43, 127.36, 127.20, 126.10, 125.60, 124.45, 118.23, 112.60, 105.28 (2C), 98.90, 61.46, 55.40, 55.36, 50.00, 48.64, 44.97, 38.97, 36.36, 36.19, 29.72, 28.89. HRMS (ESI): m/z calcd for $\text{C}_{34}\text{H}_{34}\text{NO}_4\text{S}$ ($\text{M}+\text{H}$) $^+$ 552.2209 found: 552.2200.

1,4-Dimethyl-2-((methylsulfinyl)methyl)-3-phenyl-1,2,2a,2b,3,4-hexahydro-5H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5-one (5l).

This compound was isolated as a dark brownish-red semi-solid by following the general procedure-7. 75 mg of **4l** afforded 52 mg of **5l** (75% yield), M.P = 212-214 °C. $R_f = 0.4$ (9:1 EtOAc: Hexanes, visualized by 254 nm UV light).

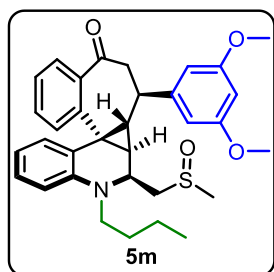


IR (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2955, 2928, 1677, 1594, 1495, 1430, 1202, 1152, 1053, 835, 735. ^1H NMR (400 MHz, CDCl_3): δ 7.56 (td, $J = 7.6, 1.5$ Hz, 1H), 7.47 (td, $J = 7.5, 1.4$ Hz, 1H), 7.40 (dd, $J = 7.7, 1.2$ Hz, 1H), 7.38-7.31 (m, 3H), 7.26 (d, $J = 7.4$ Hz, 1H), 7.20-7.14 (m, 2H), 7.07 (dd, $J = 8.7, 1.7$ Hz, 1H), 6.70 (dd, $J = 8.4, 1.1$ Hz, 1H), 6.62 (td, $J = 7.5, 1.1$ Hz, 1H), 6.51 (dd, $J = 7.7, 1.7$ Hz, 1H), 3.89-3.85 (m, 1H),

3.16 (dd, $J = 12.3, 2.6$ Hz, 1H), 3.08-3.03 (m, 1H), 2.87 (d, $J = 9.2$ Hz, 3H), 2.38 – 2.23 (m, 2H), 1.99 (s, 3H), 1.91-1.84 (m, 1H), 1.78 (s, 1H), 0.94 (d, $J = 7.2$ Hz, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 211.71, 142.74, 141.71, 141.04, 138.60, 131.84, 131.24, 128.96 (2C), 128.20, 128.09, 127.87, 127.16, 127.08, 126.99, 126.96, 118.32, 112.41, 61.56, 52.76, 51.59, 49.54, 39.20, 35.91, 34.48, 34.14, 31.62, 17.90. HRMS (ESI): m/z calcd for $\text{C}_{29}\text{H}_{30}\text{NO}_2\text{S}$ ($\text{M}+\text{H}$) $^+$ 456.1997 found: 456.2000.

1-Butyl-3-(3,5-dimethoxyphenyl)-2-((methylsulfinyl)methyl)-2,2a,3,4-tetrahydro-1H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5(2bH)-one (5m).

This compound was isolated as a yellowish-green solid by following the general procedure-7.



70 mg of **4m** afforded 47 mg of **5m** (72% yield), M.P = 212-214 °C. $R_f = 0.2$ (9:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR** (thin film, neat): $\nu_{\text{max}}/\text{cm}^{-1}$ 2955, 2928, 1677, 1594, 1495, 1430,

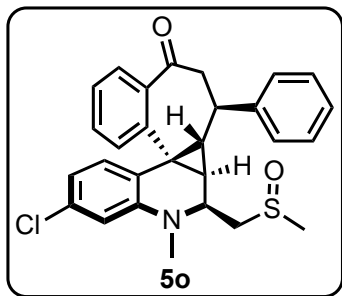
1202, 1152, 1053, 835, 735. ^1H NMR (400 MHz, CDCl_3): δ 7.59-7.51 (m, 2H), 7.47 (ddd, $J = 8.1, 7.2, 1.3$ Hz, 1H), 7.40 (dd, $J = 8.2, 1.2$ Hz, 1H), 7.05 (ddd, $J = 8.6, 7.2, 1.8$ Hz, 1H), 6.66 (dd, $J = 8.5, 1.0$ Hz, 1H), 6.56 (td, $J = 7.4, 1.0$ Hz, 1H), 6.46 (dd, $J = 7.6, 1.7$ Hz, 1H),

6.35 (s, 3H), 4.08 (ddd, $J = 11.0, 4.4, 2.6$ Hz, 1H), 3.77 (d, $J = 8.4$ Hz, 7H), 3.36 (ddd, $J = 15.9, 11.0, 5.0$ Hz, 1H), 3.29-3.12 (m, 3H), 2.98–2.90 (m, 1H), 2.87 (dd, $J = 18.7, 3.0$ Hz, 1H), 2.74 (ddd, $J = 13.3, 10.6, 3.1$ Hz, 1H), 2.42 (t, $J = 4.8$ Hz, 1H), 2.22 (s, 3H), 2.17-2.08 (m, 1H), 1.78 (dd, $J = 10.7, 5.2$ Hz, 1H), 1.39 (q, $J = 7.5$ Hz, 2H), 0.99 (t, $J = 7.3$ Hz, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ 205.94, 161.12 (2C), 145.14, 141.50, 140.76, 139.52,

132.76, 132.12, 128.26, 128.18, 128.08, 126.95, 126.43, 117.74, 112.56, 105.23 (2C), 98.76, 61.65, 55.39, 55.36, 48.50, 48.13, 47.72, 44.95, 39.31, 35.19, 34.83, 30.78, 28.40, 20.24, 13.97. **HRMS (ESI):** m/z calcd for $C_{33}H_{38}NO_4S$ ($M+H$)⁺ 544.2522 found: 544.2527.

12-Chloro-1-methyl-2-((methylsulfinyl)methyl)-3-phenyl-1,2,2a,2b,3,4-hexahydro-5H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5-one (5o).

This compound was isolated as pale-yellow oil by following the general procedure-7. 50 mg

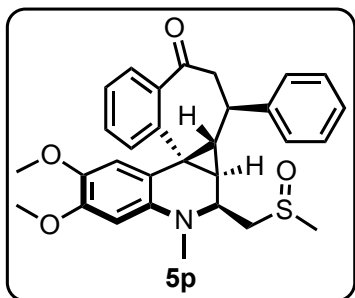


of **7o** afforded 28 mg of **5o** (60% yield), $R_f = 0.2$ (4:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 2924, 2855, 1675, 1594, 1494, 1456, 1401, 1284, 1212, 1109, 1045, 1025, 878, 763. **¹H NMR (400 MHz, CDCl₃):** δ 7.57-7.51 (m, 2H), 7.48-7.44 (m, 1H), 7.36-7.29 (m, 3H), 7.24-7.18 (m, 3H), 6.63 (d, $J = 1.9$ Hz, 1H), 6.53 (dd, $J = 8.2, 2.0$ Hz, 1H), 6.35 (d, $J = 8.2$ Hz, 1H), 3.96-3.91 (m, 1H), 3.25-3.13 (m, 2H), 2.87-2.76 (m, 5H), 2.38 (t, $J = 4.9$ Hz, 1H),

2.04 (s, 3H), 1.99 (d, $J = 11.9$, Hz, 1H), 1.75 (dd, $J = 10.7, 5.2$ Hz, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 206.00, 143.65, 142.47, 140.82, 138.77, 132.79, 132.68, 131.87, 128.99 (2C), 128.89, 128.44, 128.04, 127.38 (2C), 127.23, 124.93, 117.92, 112.50, 61.97, 49.75, 48.89, 44.70, 39.11, 35.99, 35.49, 34.23, 30.71. **HRMS (ESI):** m/z calcd for $C_{28}H_{27}ClNO_2S$ ($M+H$)⁺ 476.1451 found 476.1446.

11,12-Dimethoxy-1-methyl-2-((methylsulfinyl)methyl)-3-phenyl-1,2,2a,2b,3,4-hexahydro-5H-benzo[6',7']cyclohepta[1',2':2,3]cyclopropa[1,2-c]quinolin-5-one (5p).

This compound was isolated as yellowish-brown oil by following the general procedure-7. 50



mg of **7p** afforded 34 mg of **5p** (73% yield), $R_f = 0.1$ (10:1 EtOAc: Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** ν_{max}/cm^{-1} 3056, 2926, 2852, 1679, 1597, 1515, 1455, 1396, 1221, 1026, 765, 735, 701. **¹H NMR (400 MHz, CDCl₃):** δ 7.56-7.49 (m, 2H), 7.46-7.42 (m, 1H), 7.39 (d, $J = 7.7$ Hz, 1H), 7.30 (t, $J = 7.0$ Hz, 2H), 7.23-7.18 (m, 3H), 6.29 (s, 1H), 6.02 (s, 1H), 3.86-3.81 (m, 4H), 3.52 (s, 3H), 3.22 (dd, $J = 18.6, 12.0$ Hz, 1H), 3.15 (dd, $J = 14.6, 11.9$ Hz, 1H), 2.89-

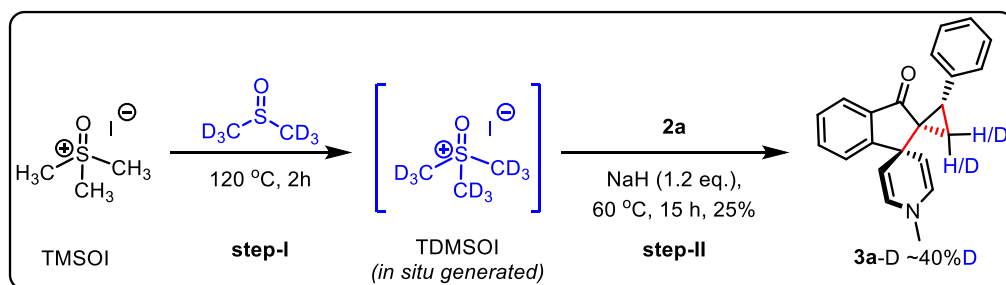
2.82 (m, 4H), 2.74 (dd, $J = 10.9, 3.0$ Hz, 1H), 2.34 (t, $J = 4.8$ Hz, 1H), 2.05 (s, 3H), 2.03-1.96 (m, 1H), 1.75 (dd, $J = 10.0, 5.0$ Hz, 1H). **¹³C NMR (100 MHz, CDCl₃):** δ 206.24, 147.88, 142.67, 141.60, 140.85, 139.30, 137.07, 132.60, 131.95, 128.92 (2C), 128.21, 127.80, 127.38 (2C), 127.11, 118.60, 112.48, 98.73, 61.03, 56.42, 56.13, 50.29, 49.00, 44.70, 39.15, 36.39, 35.39, 34.30, 30.55. **HRMS (ESI):** m/z calcd for $C_{30}H_{32}NO_4S$ ($M+H$)⁺ 502.2052 found 502.2051.

General Procedure-8: One-pot synthesis of pyridinium salts and spirannulation

In DMF: In a sealed tube, biaryl enone **6a** (0.09 g, 0.31 mmol) or **6b** (0.084 g, 0.24 mmol) was dissolved in DMF (2 mL). Then, methyl iodide (1.2 eq.) was added in one portion and the reaction mixture was stirred at 70 °C for 3-6 h, and monitored the reaction on TLC. After complete consumption of the starting material, the reaction mixture was added to a suspension of sodium hydride (60% in oil, 1.2 eq.) and TMSOI (1.1 eq.) in a dry DMF (2.0 mL) under an N₂ atmosphere. The reaction mixture was stirred at room temperature until the salt disappeared (by TLC). Then, quenched the reaction mixture using ice water, and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate to afford **3a** in 28% and **3b** in 57% yield.

In DCM: In a sealed tube, biaryl enone **6a** (0.09 g, 0.31 mmol) or **6b** (0.084 g, 0.24 mmol) or **7f** (0.12 g, 0.85 mmol) or **7m** (0.11 g, 0.27 mmol) was dissolved in DCM (3.0 mL). Then, alkyl iodide was added in one portion; the reaction mixture was stirred at 70 °C for 3-6 h, and monitored the reaction on TLC. After complete consumption of the starting material, the solvent was evaporated and added to a suspension of sodium hydride (60% in oil, 1.2 eq.) and TMSOI (1.1 eq.) in dry DMF (2.0 mL) was added under N₂ atmosphere. The reaction mixture was stirred at room temperature until the salt disappeared (as monitored by TLC). Then, quenched the reaction mixture using ice water, and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate to afford **6** or **7**.

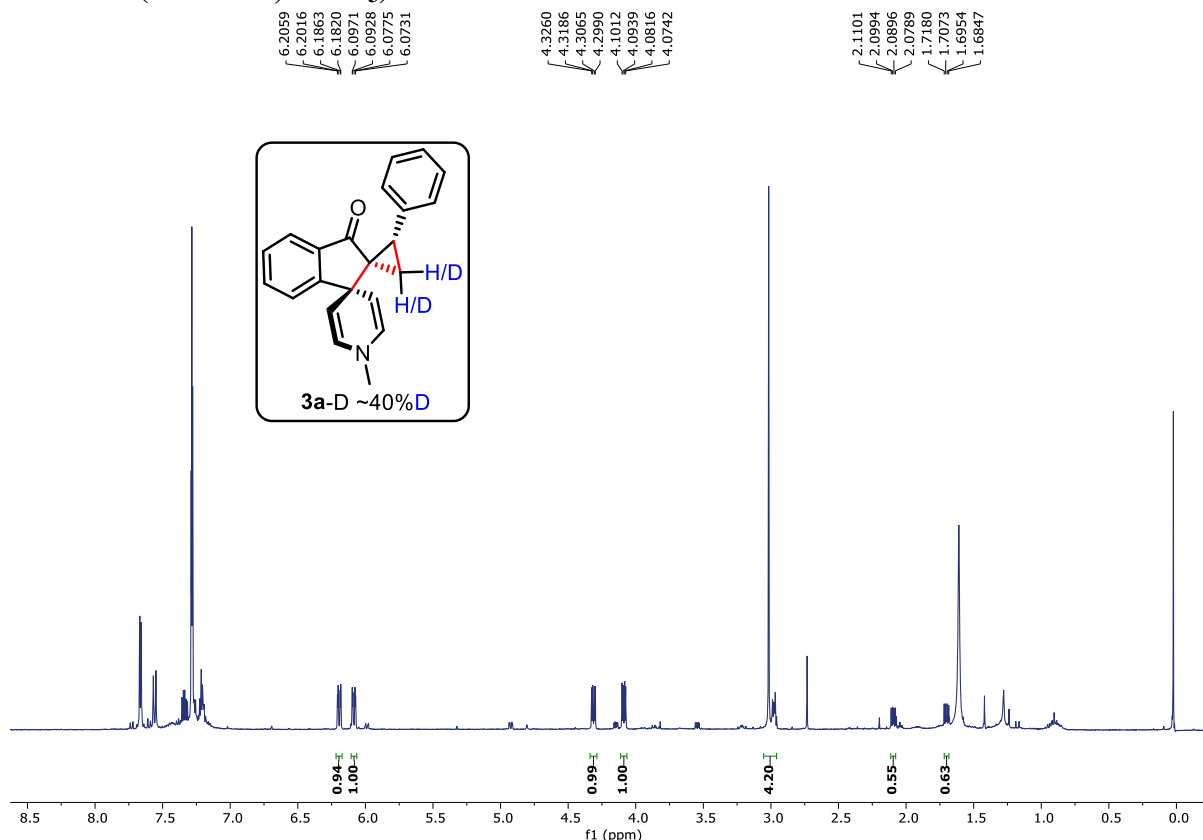
General Procedure-9: Reaction of **2a** with trideuteromethyl sulfoxonium iodide (TMSOI)



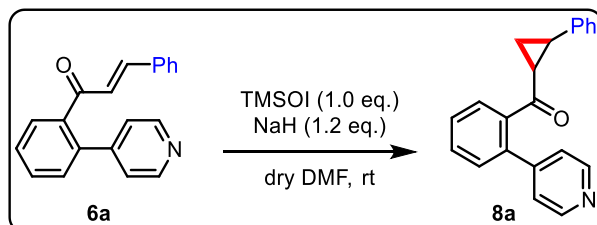
Scheme S8: Reaction of **2a** with TMSOI

To a sealed reaction tube, TMSOI (0.18 g, 0.82 mmol) and DMSO-*d*₆ (40.0 eq.) were added and stirred at 120 °C for 2 h. The reaction mixture was cooled to room temperature and a suspension of sodium hydride (60% in oil, 36 mg, 0.9 mmol) in dry DMF (1.0 mL), **2a** (0.21 g, 0.75 mmol) were added drop wise over a period of 5-10 min and stirred at 60 °C for 15 h. The reaction mixture was quenched using ice water and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate. The level of deuterium incorporation in bis-spiro indanone product **3a-D** was determined by ¹H-NMR spectroscopy. The metathesis reaction between DMSO-*d*₆ and TMSOI yielded the bis-spiro indanone **3a-D** with 40% deuterium incorporation.

¹H NMR (400 MHz, CDCl₃) for 3a-D



General procedure-10: To study the role of the pyridinium portion

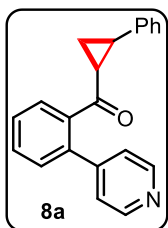


Scheme S9: Reaction of **6a** with TMSOI.

A mixture of sodium hydride (60% in oil, 16 mg, 0.42 mmol) and TMSOI (85 mg, 0.23 mmol) was placed in an oven-dried flask, and dry DMF (4.0 mL) was added to the mixture. After the evolution of hydrogen ceased and the milky solution turned clear, the reaction mixture was stirred for 15 min. The compound **6a** (100 mg, 0.35 mmol) dissolved in dry DMF (1.0 mL) was added over a period of 5-10 min and stirred at room temperature until the reactant **6a** disappeared as monitored by TLC. The reaction mixture was quenched using ice water and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate.

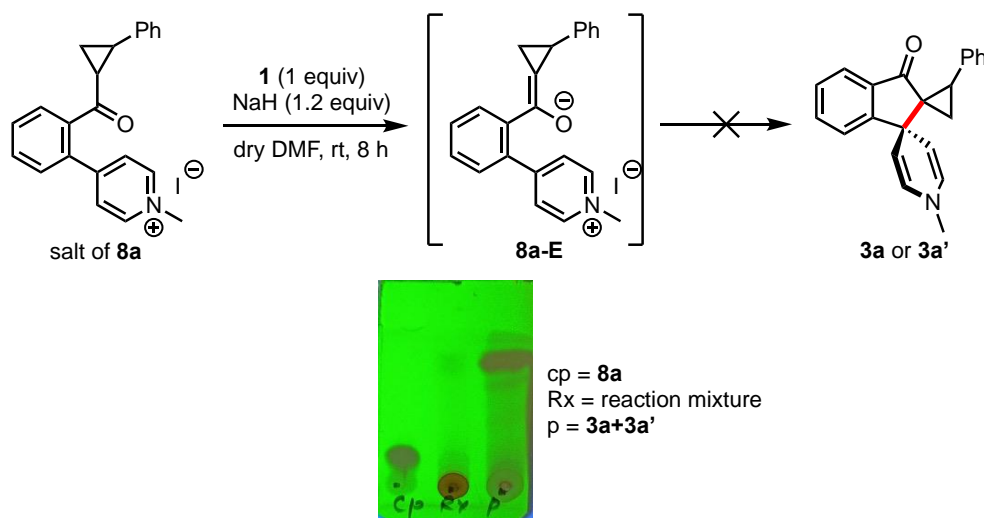
(2-Phenylcyclopropyl)(2-(pyridin-4-yl)phenyl)methanone (**8a**).

This compound was isolated as a reddish-brown sticky oil by following the general procedure-10. 100 mg of **6a** afforded 84 mg of **8a** (80% yield), $R_f = 0.3$ (8:2 EtOAc:



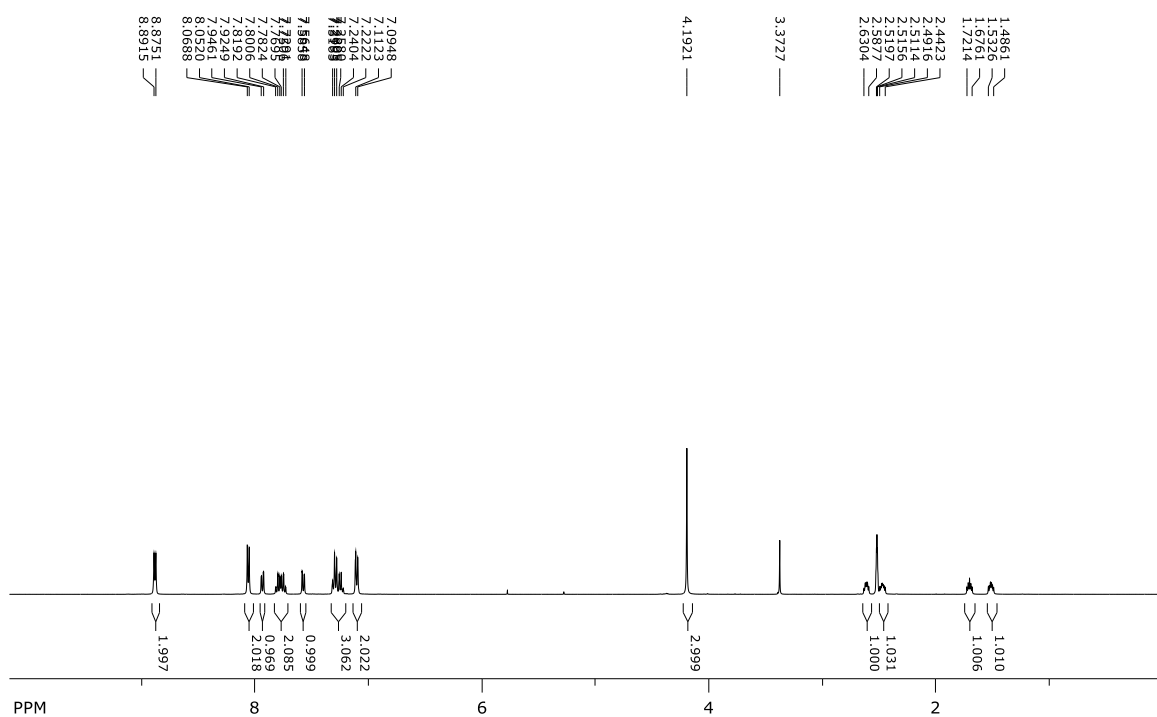
Hexanes, visualized by 254 nm UV light). **IR (thin film, neat):** $\nu_{\max}/\text{cm}^{-1}$ 3052, 2925, 1685, 1598, 1498, 1480, 1288, 1044, 720. **^1H NMR (400 MHz, CDCl_3)** δ 8.55 – 8.51 (m, 2H), 7.70 – 7.65 (m, 1H), 7.58 (td, $J = 7.5, 1.5$ Hz, 1H), 7.52 (td, $J = 7.5, 1.4$ Hz, 1H), 7.39 (dd, $J = 7.4, 1.4$ Hz, 1H), 7.28 – 7.24 (m, 2H), 7.24 – 7.18 (m, 3H), 6.88 – 6.82 (m, 2H), 2.59 (ddd, $J = 9.1, 6.7, 4.0$ Hz, 1H), 2.11 (ddd, $J = 8.1, 5.2, 4.0$ Hz, 1H), 1.79 (ddd, $J = 9.2, 5.3, 4.1$ Hz, 1H), 1.32 – 1.23 (m, 1H). **^{13}C NMR (100 MHz, CDCl_3)** δ 203.75, 149.74 (2C), 148.33, 140.50, 139.65, 138.04, 131.15, 130.08, 128.73, 128.43 (2C), 128.36, 126.69, 125.65 (2C), 123.75 (2C), 34.42, 31.92, 21.72. **HRMS (ESI):** m/z calcd for $\text{C}_{21}\text{H}_{18}\text{NO}$ ($\text{M}+\text{H}$)⁺ 300.1388 found: 300.1424.

To rule out the reaction goes through the initial formation of **8a** and a subsequent base-mediated cyclization on to the pyridinium moiety, a control experiment has been performed with the salt of **8a**, Scheme 10. However, the formation of only a trace of **3a** was observed under the optimized condition. This pathway was not preferred possibly because of the high strain associated with the intermediate enolate **8a-E**. This result further validates the proposed mechanism in Scheme 8.

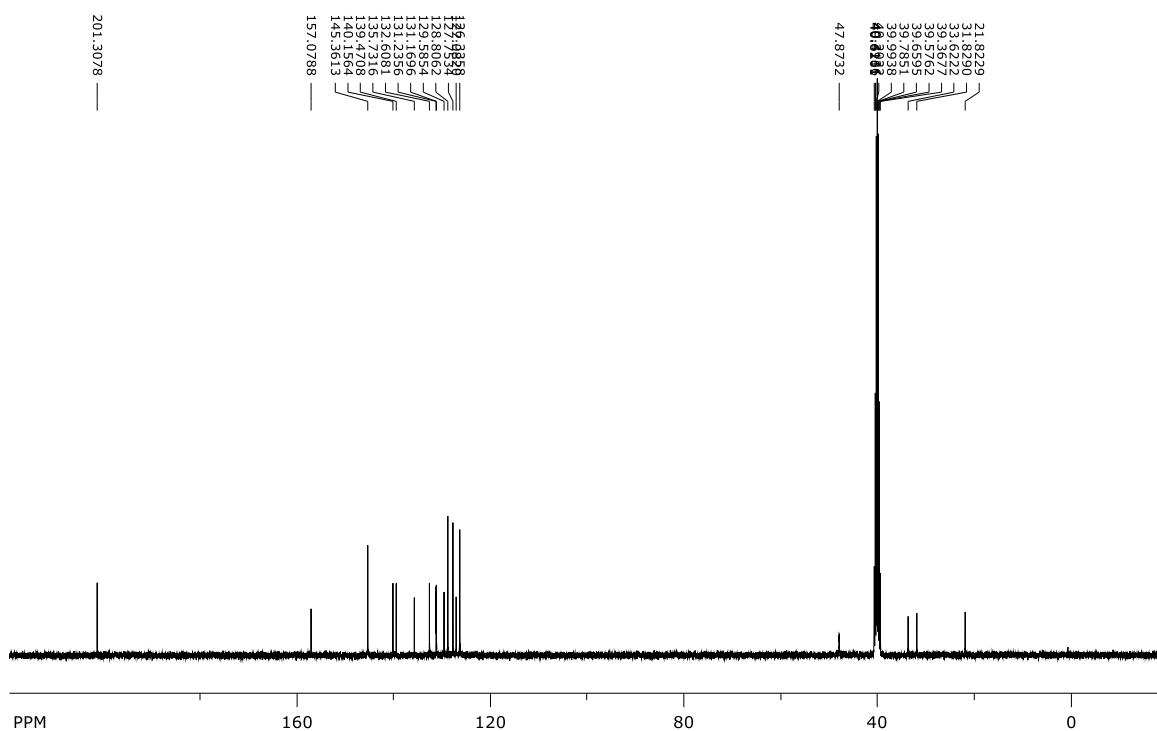


Scheme S10. A control experiment to rule out the intermediacy of **8a-E**

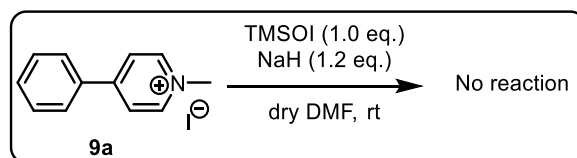
^1H NMR of the salt of 8a (400 MHz, DMSO- d_6)



^{13}C NMR of the salt of 8a (100 MHz, DMSO- d_6)



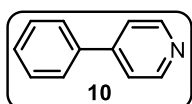
General procedure-11: To study the role of the enone moiety (General procedure-6 was followed)



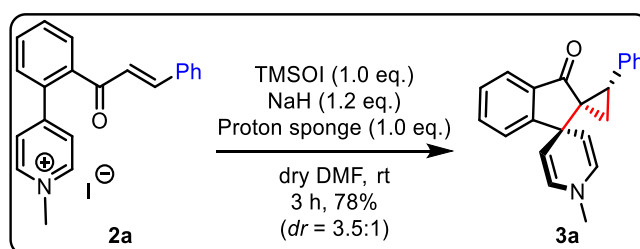
Scheme S11: Reaction of **9a** with trimethylsulfoxonium iodide

4-Phenylpyridine (**10**).

This compound was synthesized according to the reported literature.³ $R_f = 0.4$ (4:1 EtOAc: Hexanes, visualized by 254 nm UV light. $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 8.70 (d, $J = 5.8$ Hz, 2H), 7.71–7.61 (m, 2H), 7.59–7.40 (m, 5H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3): δ 150.11, 148.44, 138.09, 129.15 (3C), 129.12, 127.01 (3C), 121.80.



General procedure 12: Reaction of **2a** in the presence of proton sponge



Scheme S12: reaction of **2a** in the presence of proton sponge.

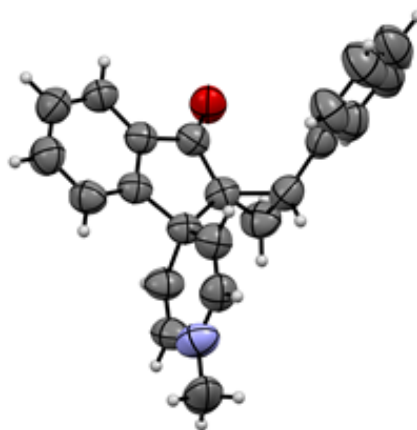
A mixture of sodium hydride (60% in oil, 10 mg, 0.25 mmol) and TMSOI (50 mg, 0.23 mmol) was placed in an oven-dried flask, and dry DMF (4.0 mL) was added to the mixture. After the evolution of hydrogen ceased, the reaction mixture was stirred for 15 min. Then, **2a** (0.90 mg, 0.21 mmol) and proton sponge (44 mg, 0.21 mmol) were dissolved in dry DMF (1.0 mL) and were added drop wise over a period of 5-10 min and stirred at room temperature until **2a** disappeared (as monitored by TLC). The reaction mixture was quenched using ice water and extracted with diethyl ether. The organic extracts were combined, dried over anhydrous sodium sulfate, and concentrated. The crude product was purified by silica gel column chromatography using hexane/ethyl acetate.

³ Feuerstein, M.; Doucet, H.; Santelli, M. Efficient *J. Organomet. Chem.* **2003**, 687, 327–336.

Crystal Structure of 3a (CCDC 2133525): The structure of the **3a** was confirmed by single crystal X-ray diffraction analysis.

Crystal data and structure refinement for 3a (major)

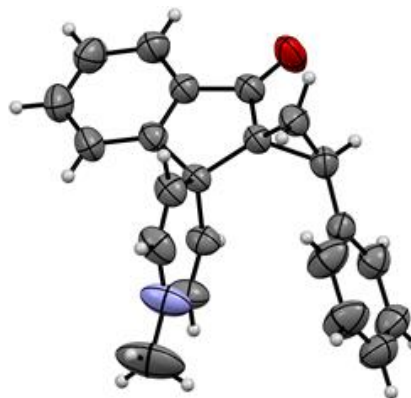
Identification code	3a (Major)
Empirical formula	C ₂₂ H ₁₉ NO
Formula weight	311.407
Temperature/K	289
Crystal system	Monoclinic
Space group	P2 ₁ /n
a/Å	10.4984(6)
b/Å	13.6723(8)
c/Å	12.6001(8)
α/°	90
β/°	108.468(7)
γ/°	90
Volume/Å ³	1715.44(19)
Z	4
ρ _{calc} /cm ³	1.206
μ/mm ⁻¹	0.072
F(000)	660.3
Crystal size/mm ³	0.3 × 0.2 × 0.2
Radiation	Mo Kα (λ = 0.71073)
2θ range for data collection/°	5.06 to 65.54
Index ranges	-14 ≤ h ≤ 15, -9 ≤ k ≤ 18, -19 ≤ l ≤ 17
Reflections collected	10342
Independent reflections	5728 [R _{int} = 0.0279, R _{sigma} = 0.0405]
Data/restraints/parameters	5728/0/218
Goodness-of-fit on F ²	1.143
Final R indexes [I ≥ 2σ (I)]	R ₁ = 0.0623, wR ₂ = 0.1825
Final R indexes [all data]	R ₁ = 0.1075, wR ₂ = 0.2261
Largest diff. peak/hole / e Å ⁻³	0.28/-0.23



Crystal Structure of 3a' (CCDC 2133527): The structure of **3a'** was confirmed by single crystal X-ray diffraction analysis.

Crystal data and structure refinement for 3a' (minor).

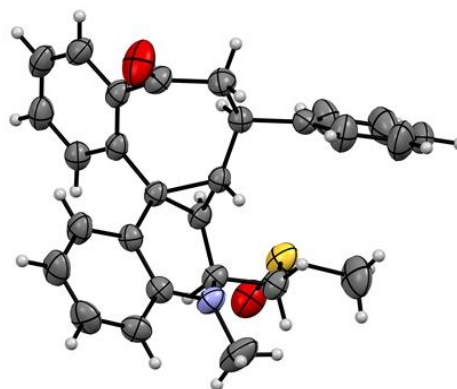
Identification code	3a' (minor)
Empirical formula	C ₂₂ H ₁₉ NO
Formula weight	313.402
Temperature/K	298
Crystal system	monoclinic
Space group	P2 ₁ /c
a/Å	8.2759(4)
b/Å	18.6849(8)
c/Å	11.6613(6)
α/°	90
β/°	106.981(5)
γ/°	90
Volume/Å ³	1724.62(15)
Z	4
ρ _{calc} /cm ³	1.207
μ/mm ⁻¹	0.074
F(000)	664.3
Crystal size/mm ³	0.6 × 0.3 × 0.2
Radiation	Mo Kα (λ = 0.71073)
2θ range for data collection/°	6.74 to 65.34
Index ranges	-7 ≤ h ≤ 11, -28 ≤ k ≤ 23, -17 ≤ l ≤ 11
Reflections collected	8796
Independent reflections	5576 [R _{int} = 0.0188, R _{sigma} = 0.0323]
Data/restraints/parameters	5576/0/218
Goodness-of-fit on F ²	1.211
Final R indexes [I >= 2σ (I)]	R ₁ = 0.0577, wR ₂ = 0.1745
Final R indexes [all data]	R ₁ = 0.0778, wR ₂ = 0.1994
Largest diff. peak/hole / e Å ⁻³	0.27/-0.19



Crystal Structure of 5a (CCDC 2143316): The structure of the **5a** was confirmed by single crystal X-ray diffraction analysis.

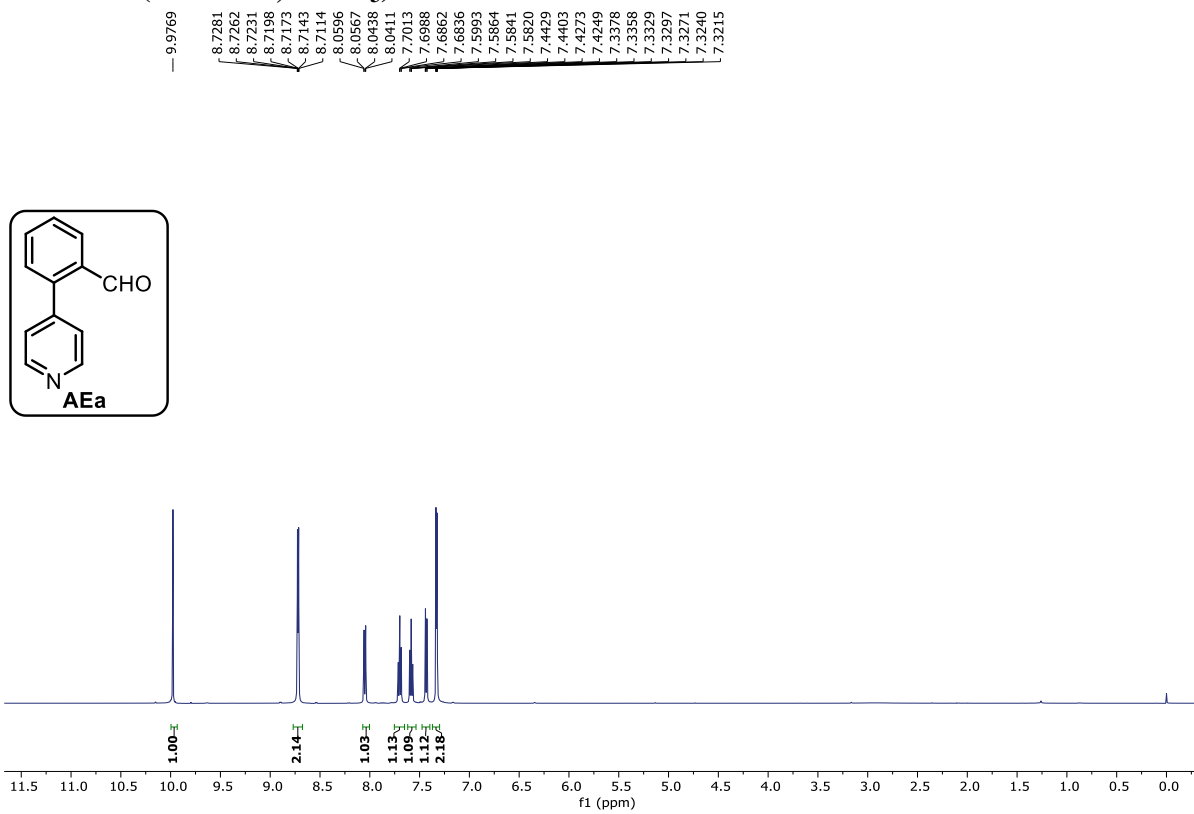
Crystal data and structure refinement for 5a.

Identification code	5a
Empirical formula	C ₂₈ H ₂₇ NO ₂ S
Formula weight	441.56
Temperature/K	293(2)
Crystal system	triclinic
Space group	P-1
a/Å	9.0157(3)
b/Å	11.1293(4)
c/Å	11.8264(3)
α/°	88.918(3)
β/°	87.476(3)
γ/°	74.544(3)
Volume/Å ³	1142.58(6)
Z	2
ρ _{calc} /cm ³	1.283
μ/mm ⁻¹	0.167
F(000)	468.0
Crystal size/mm ³	0.12 × 0.08 × 0.04
Radiation	MoKα (λ = 0.71073)
2θ range for data collection/°	6.354 to 54.952
Index ranges	-11 ≤ h ≤ 10, -14 ≤ k ≤ 14, -14 ≤ l ≤ 15
Reflections collected	16391
Independent reflections	4891 [R _{int} = 0.0484, R _{sigma} = 0.0464]
Data/restraints/parameters	4891/0/291
Goodness-of-fit on F ²	1.120
Final R indexes [I ≥ 2σ (I)]	R ₁ = 0.0502, wR ₂ = 0.1363
Final R indexes [all data]	R ₁ = 0.0743, wR ₂ = 0.1490
Largest diff. peak/hole / e Å ⁻³	0.38/-0.34

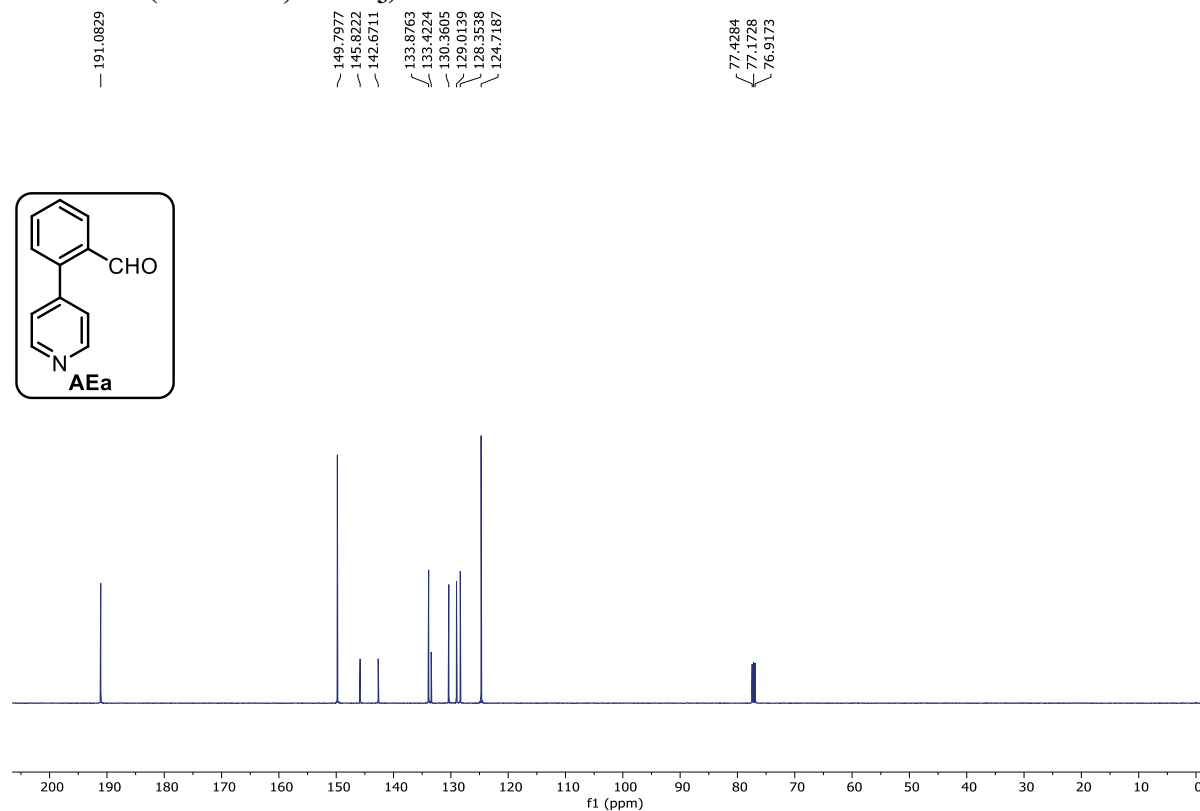


Copies of ^1H , ^{13}C and ^{19}F NMR spectra of all the new compounds reported in this study

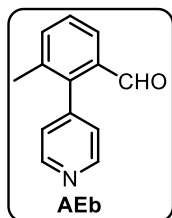
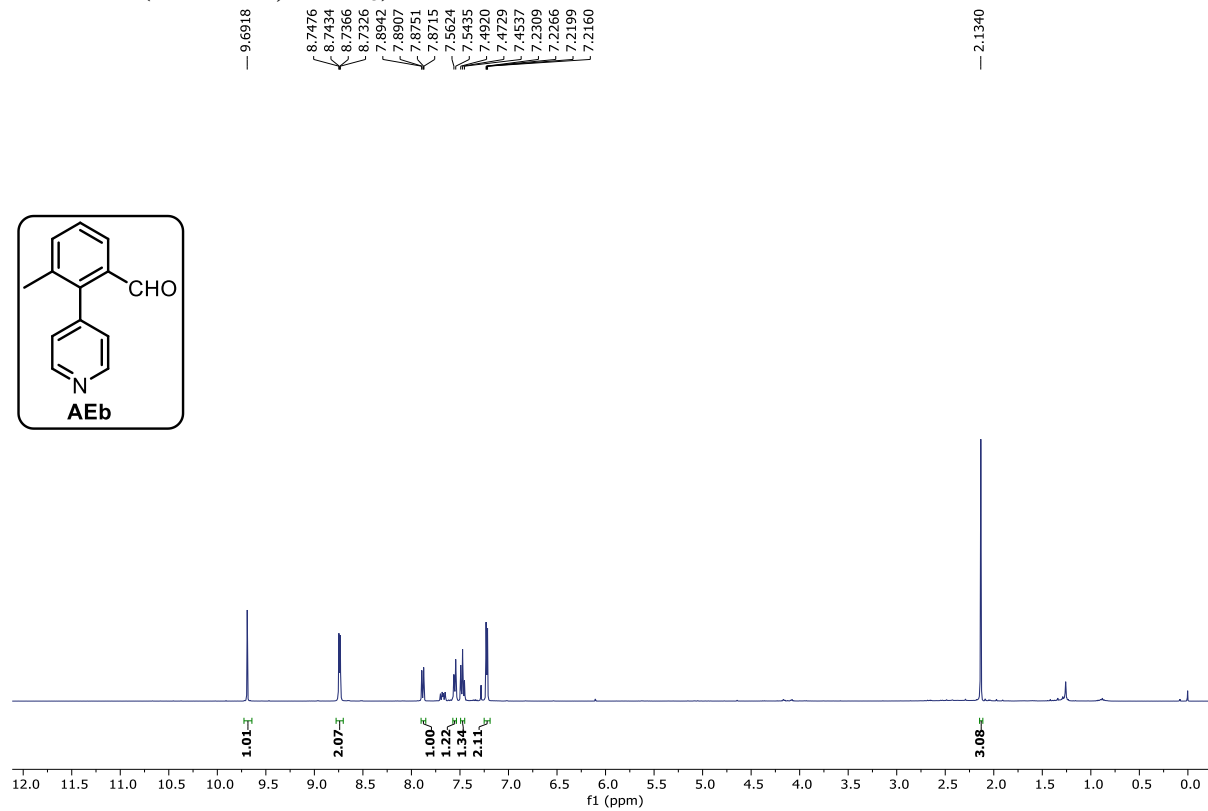
^1H NMR (500 MHz, CDCl_3)



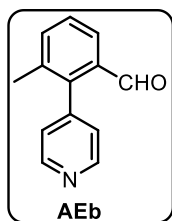
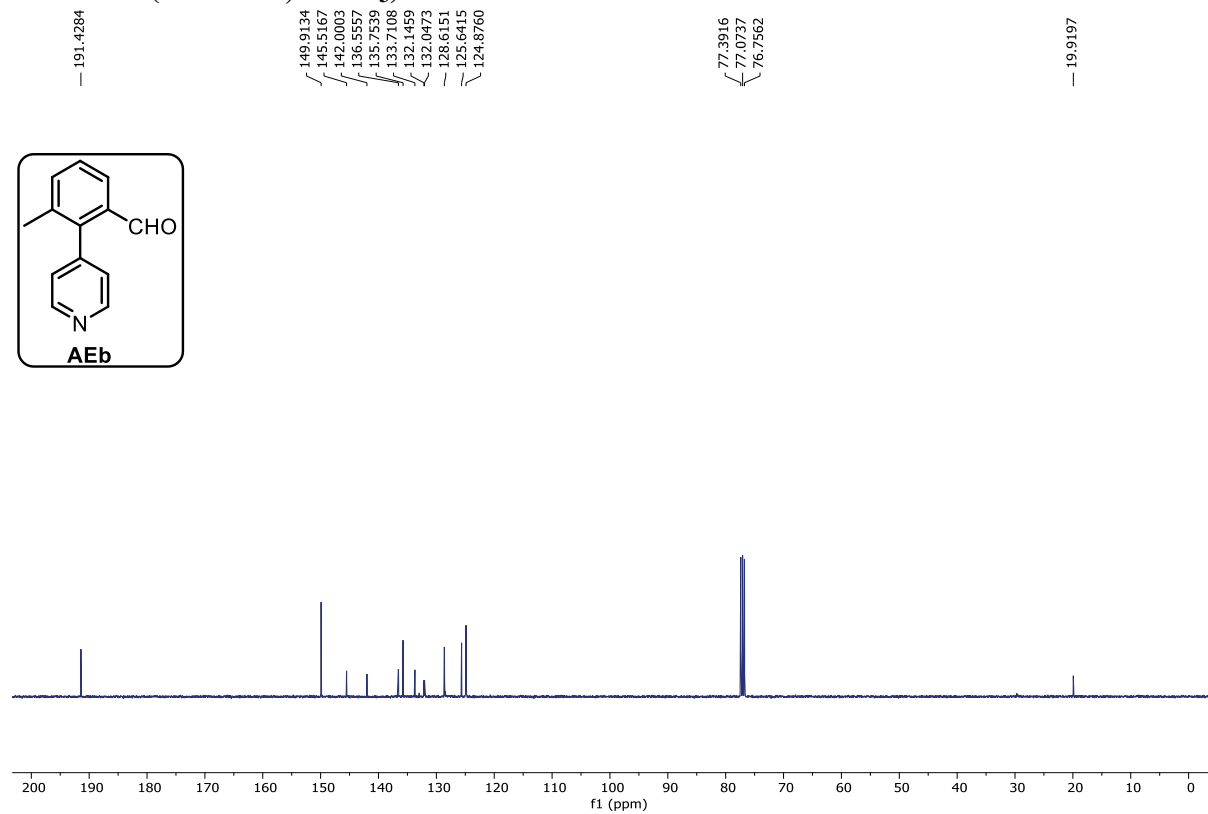
^{13}C NMR (125 MHz, CDCl_3)



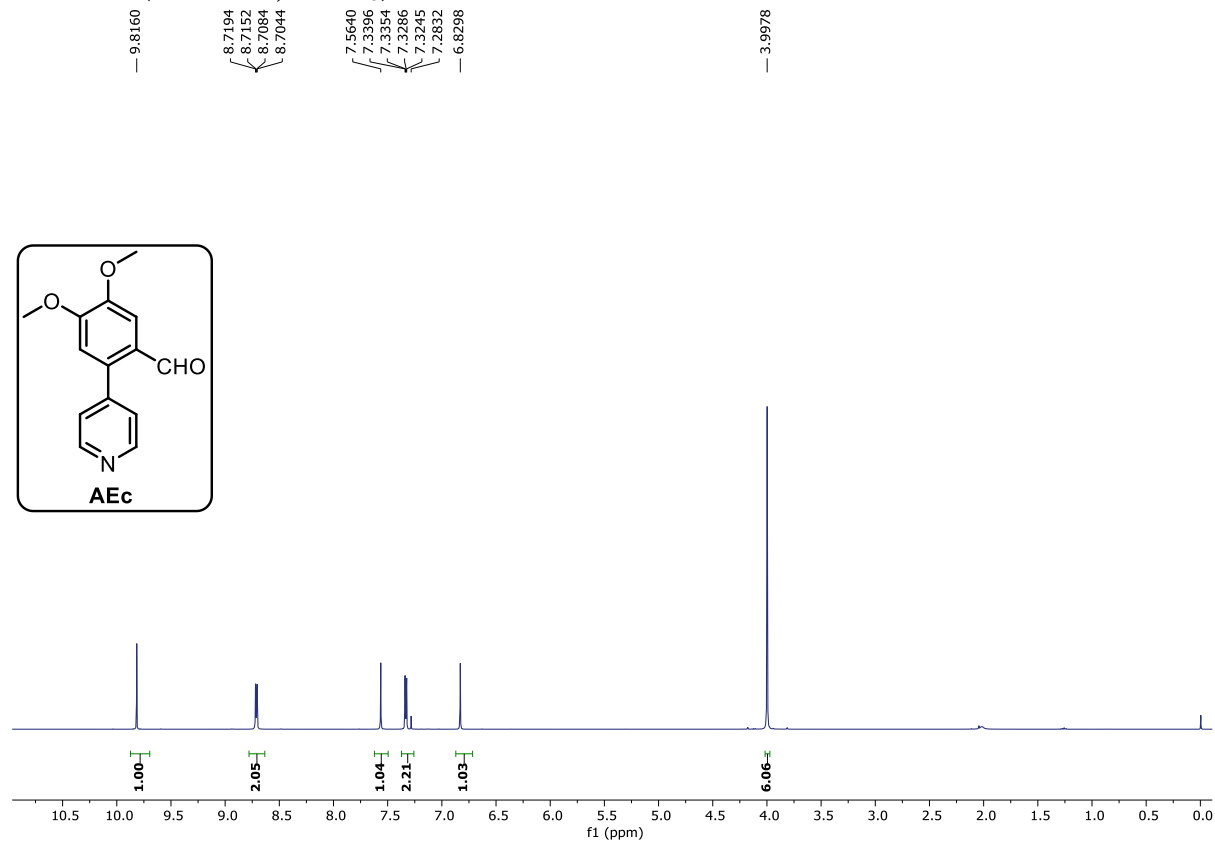
^1H NMR (400 MHz, CDCl_3)



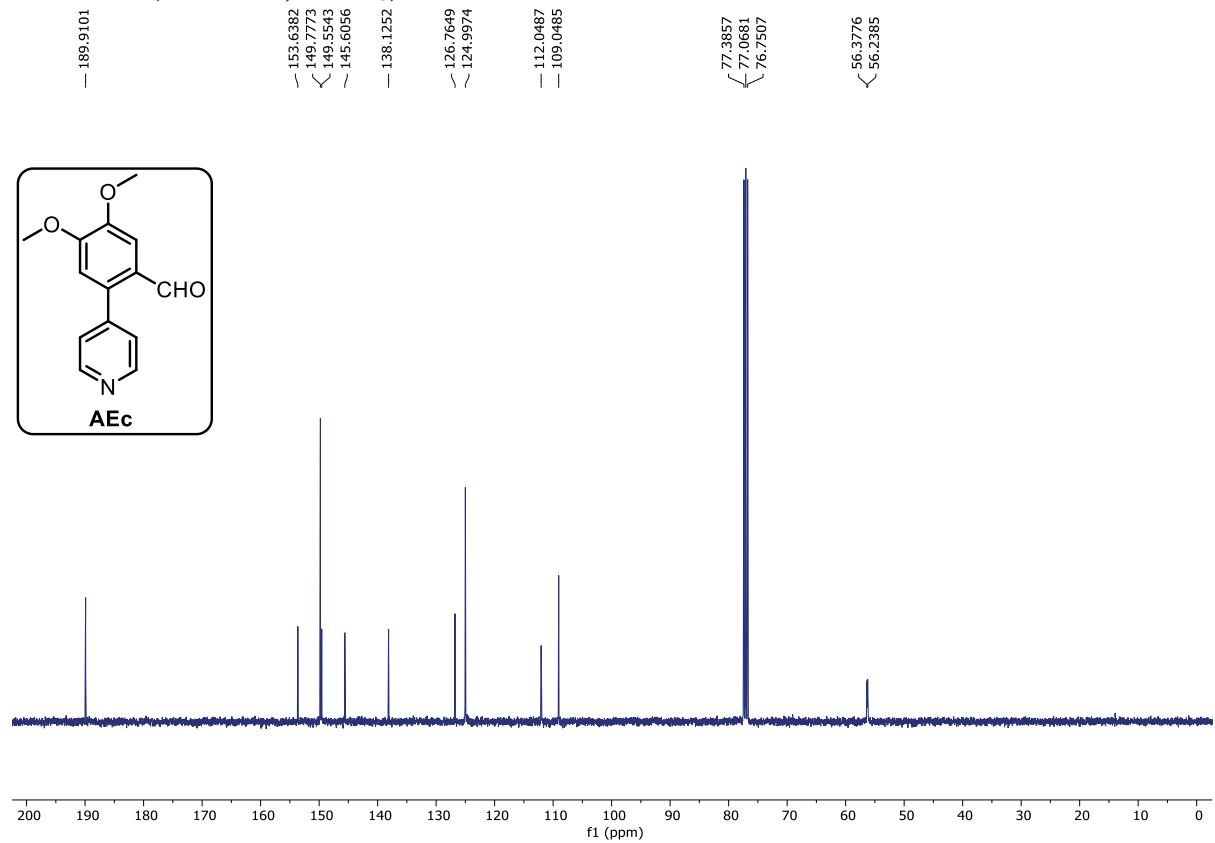
^{13}C NMR (100 MHz, CDCl_3)



¹H NMR (400 MHz, CDCl₃)

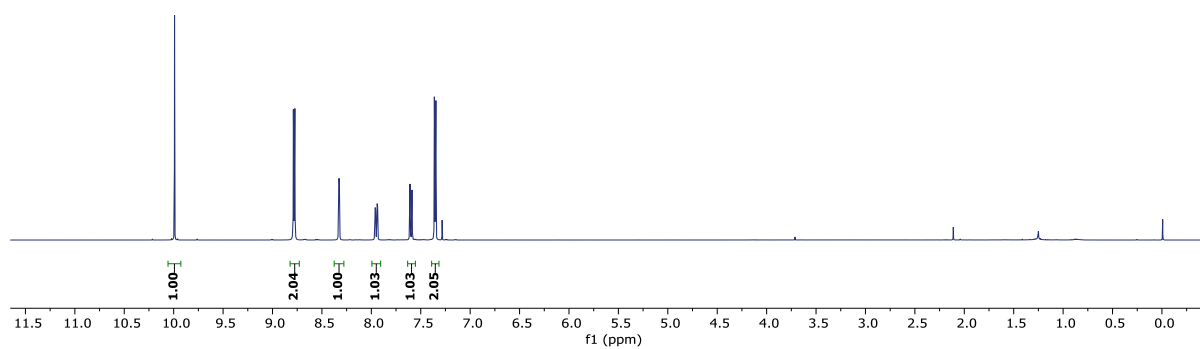
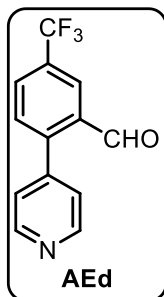


¹³C NMR (100 MHz, CDCl₃)



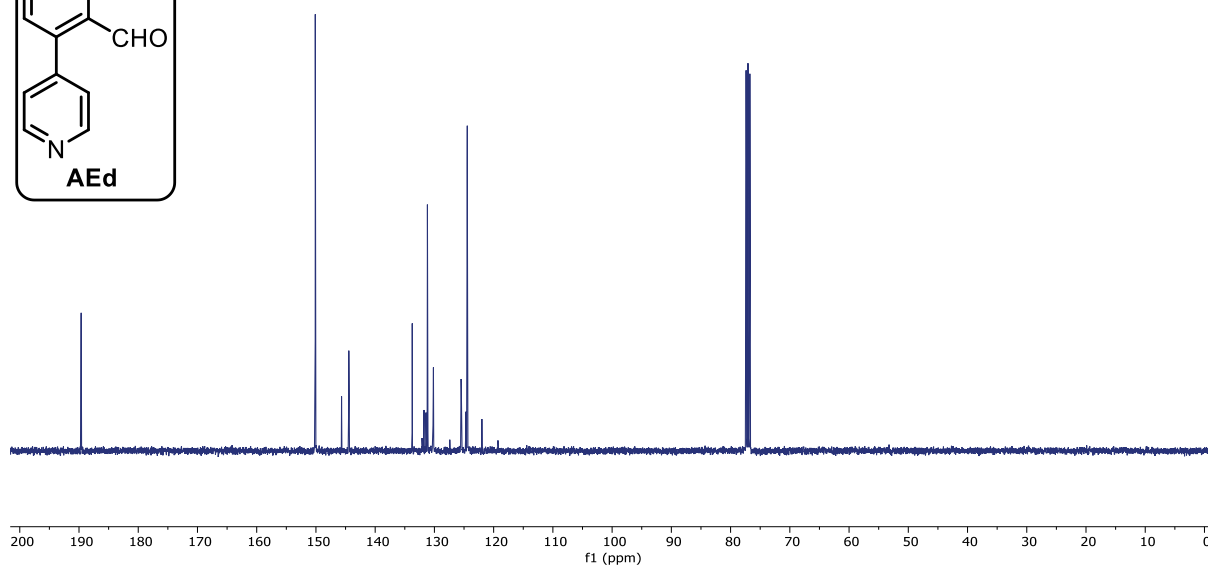
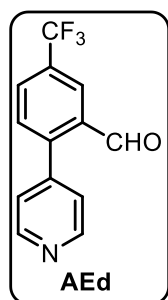
¹H NMR (400 MHz, CDCl₃)

9.9912
8.7888
8.7846
8.7779
8.7738
8.3305
8.3286
8.3270
8.3253
8.3237
8.3219
7.9626
7.9607
7.9578
7.9558
7.9425
7.9404
7.9375
7.9357
7.6106
7.6088
7.6069
7.5904
7.5888
7.5870
7.3621
7.3578
7.3511
7.3469
7.2836

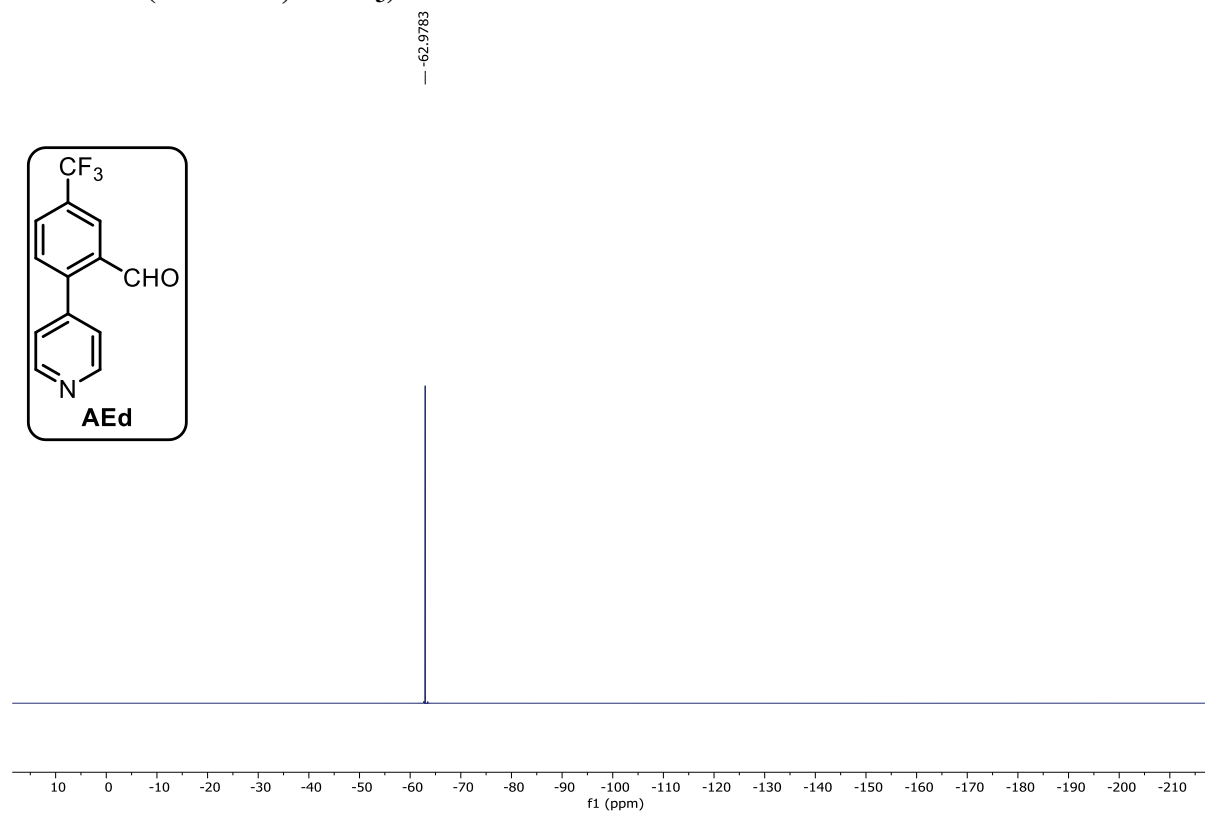


¹³C NMR (100 MHz, CDCl₃)

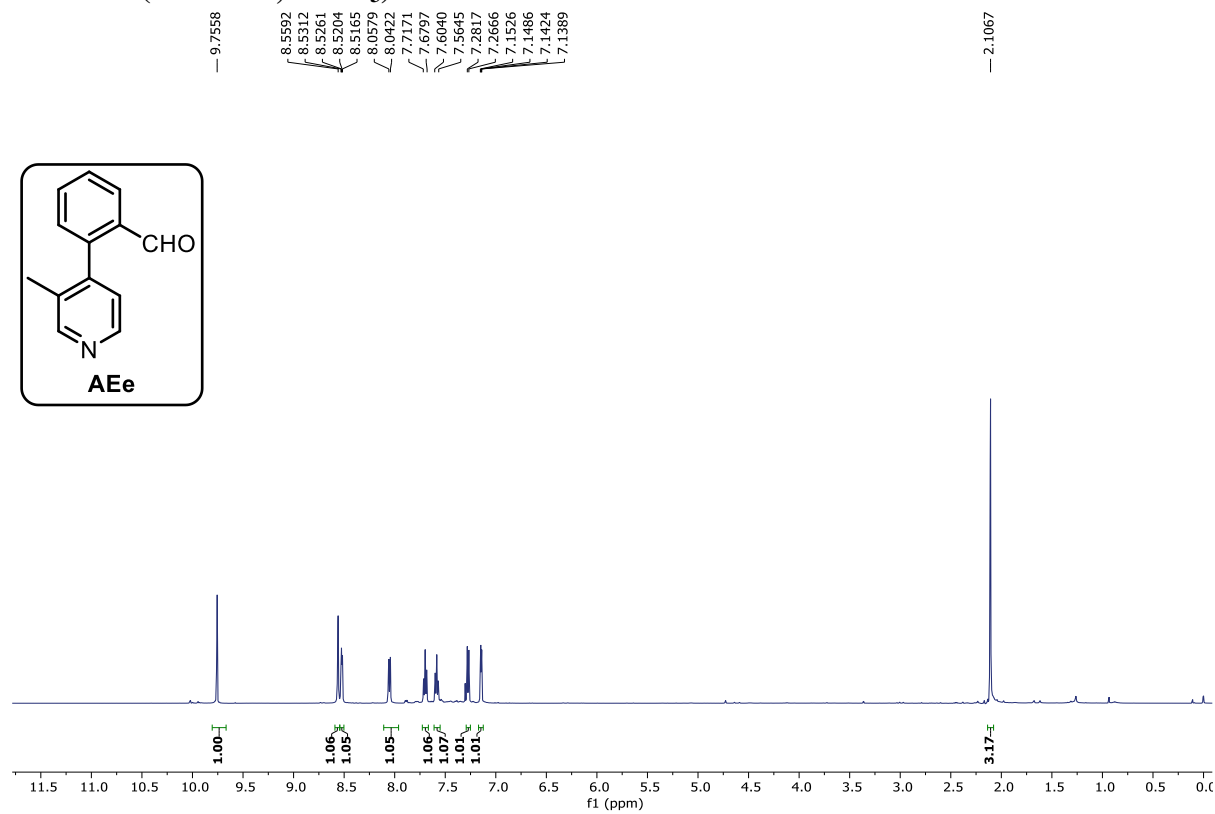
189.6441
150.1109
145.6751
145.6659
144.4501
133.7443
132.0908
131.7570
131.4228
131.1818
131.0889
130.2356
130.2015
130.1669
130.1319
127.4044
125.5315
125.4939
125.4566
125.4192
124.6948
124.4509
121.9850
119.2733
77.3814
77.0642
76.7464



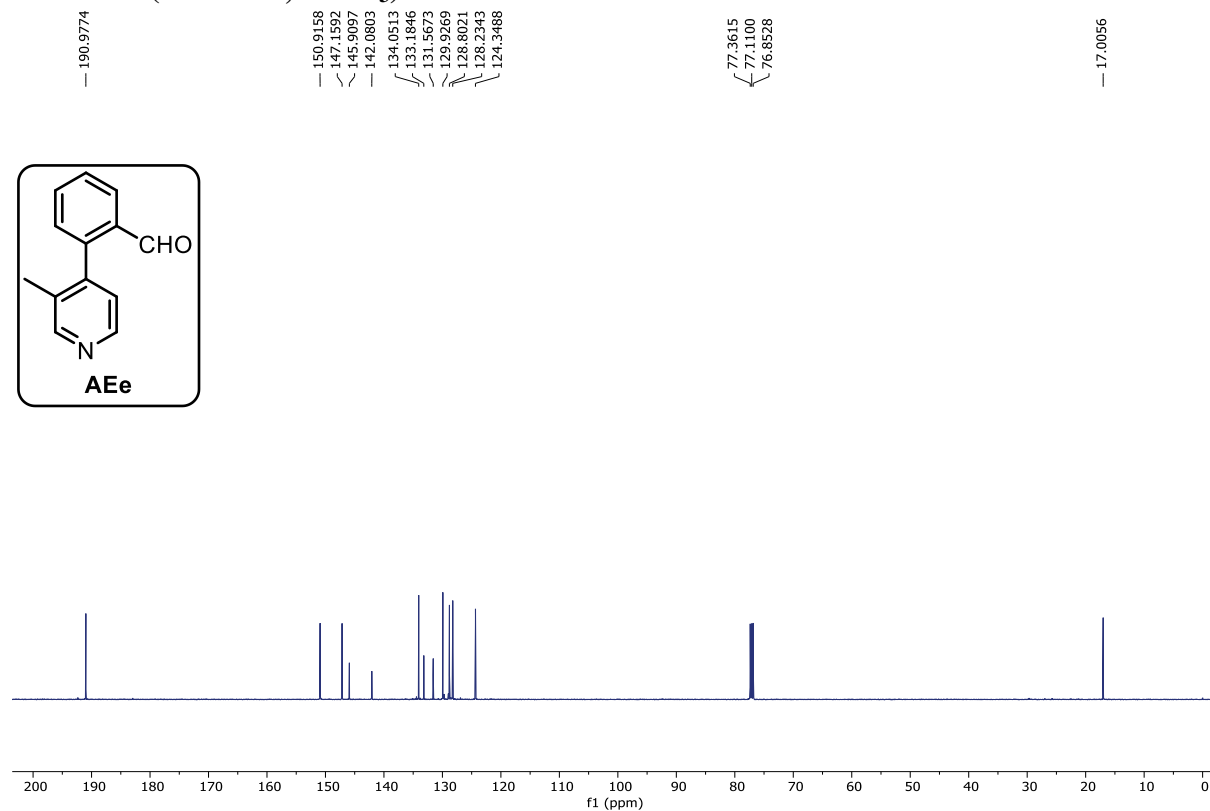
^{19}F NMR (376 MHz, CDCl_3)



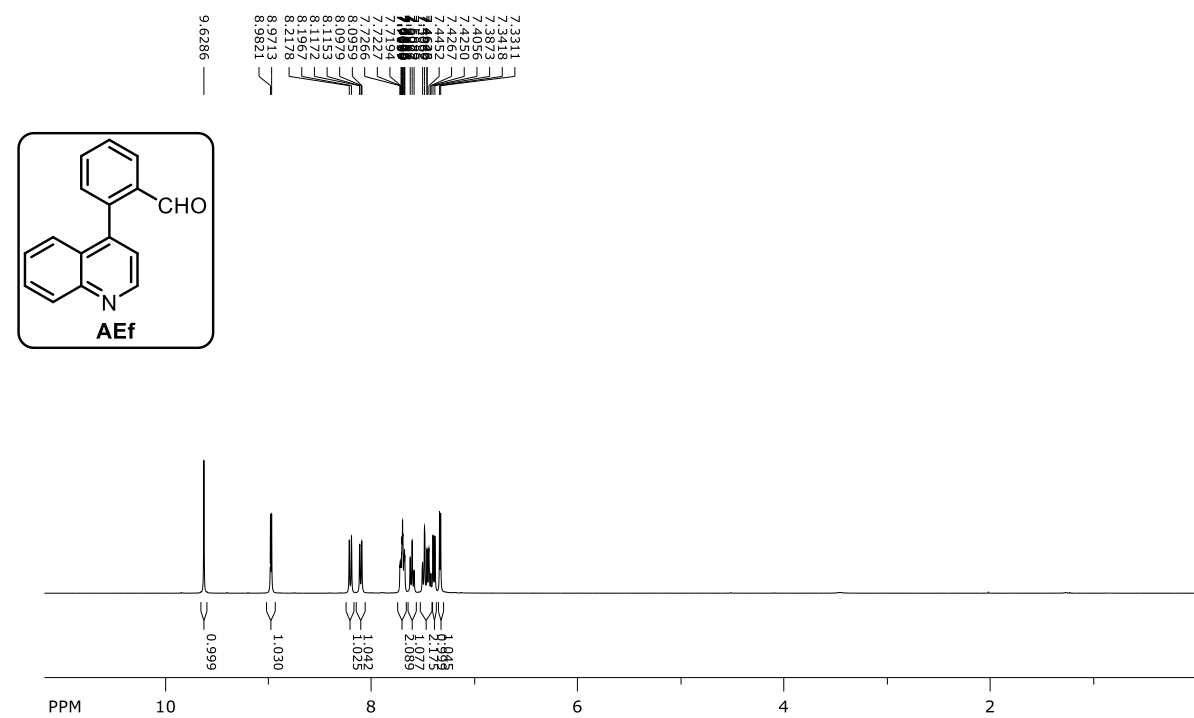
^1H NMR (500 MHz, CDCl_3)



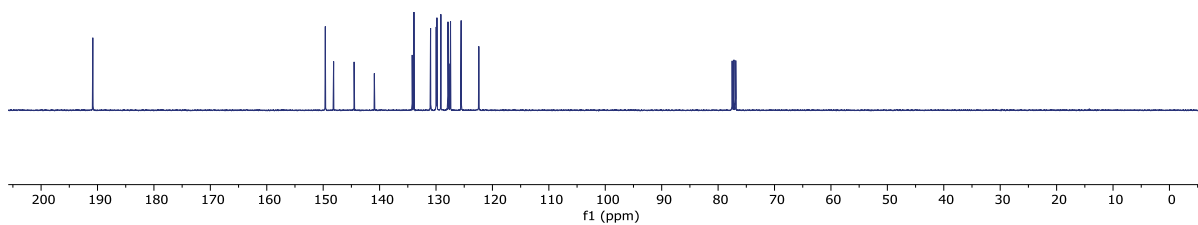
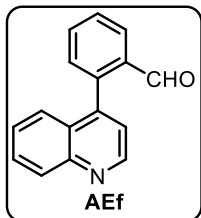
^{13}C NMR (125 MHz, CDCl_3)



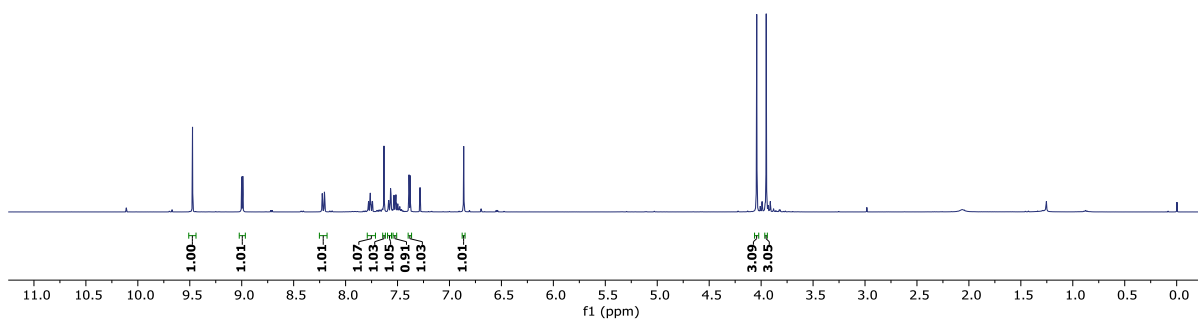
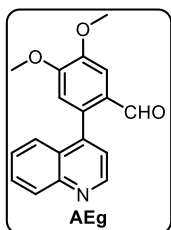
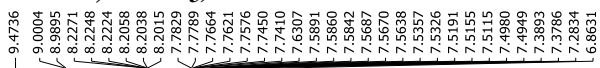
^1H NMR (400 MHz, CDCl_3)



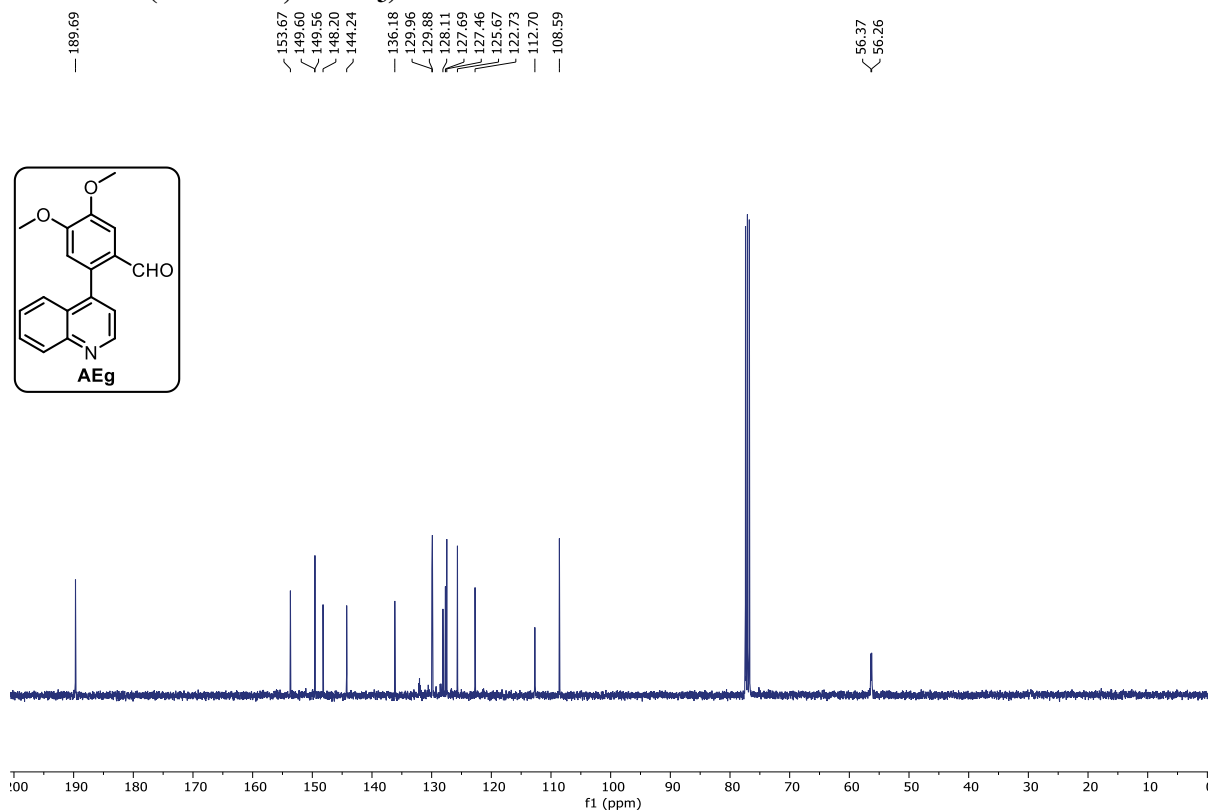
¹³C NMR (100 MHz, CDCl₃)



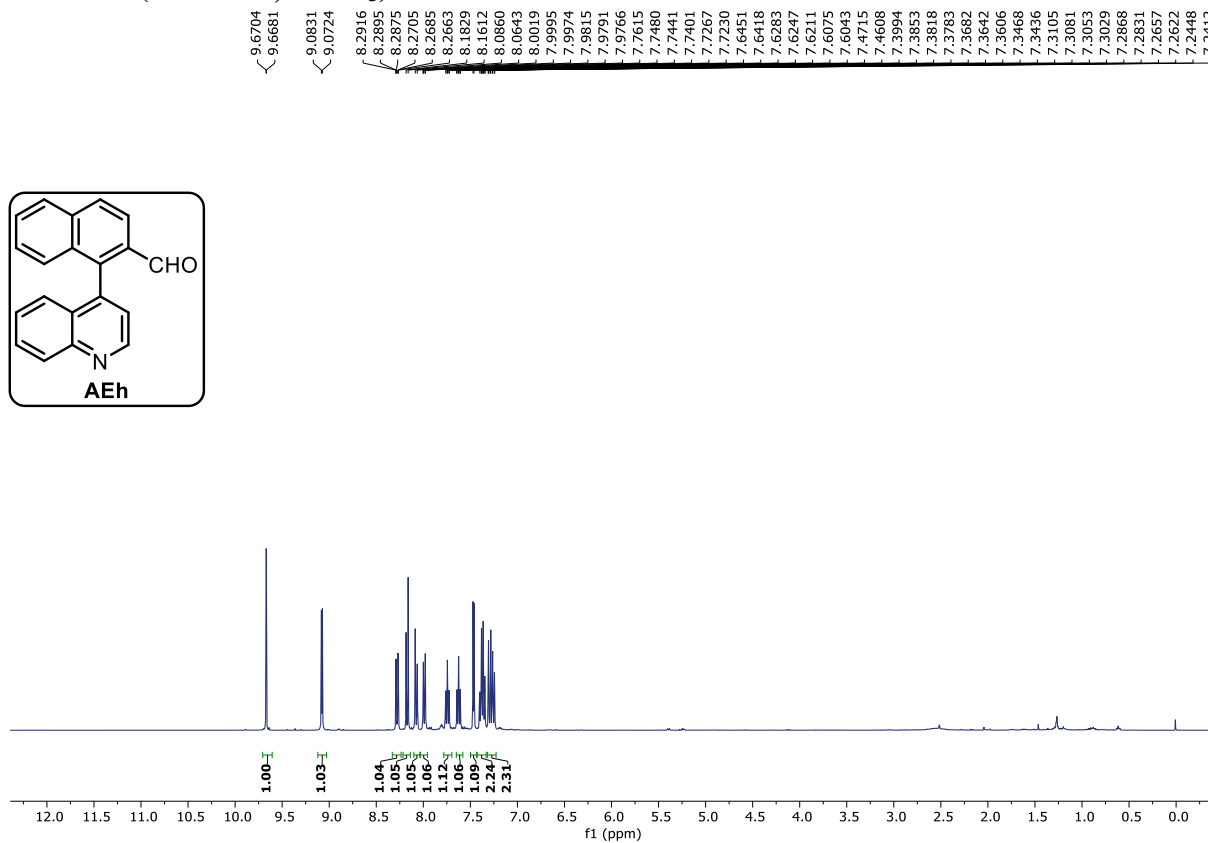
¹H NMR (400 MHz, CDCl₃)



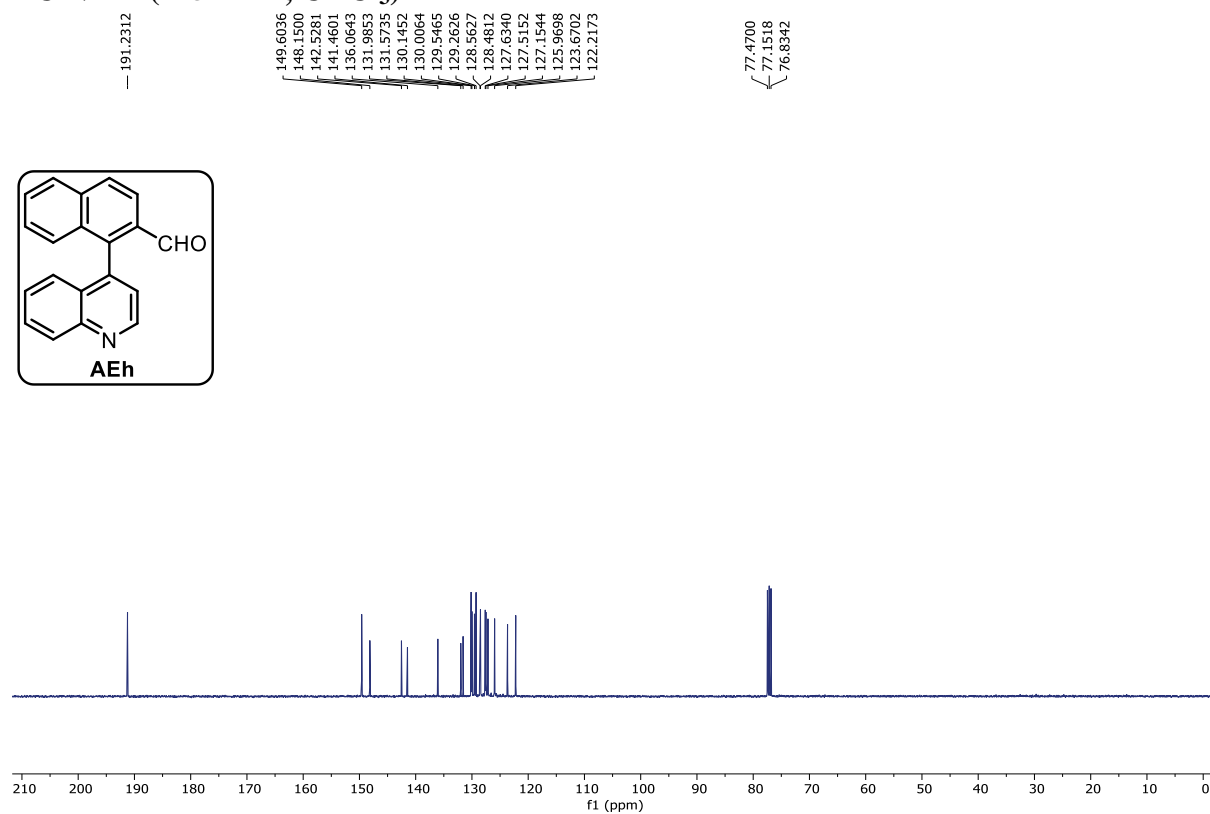
^{13}C NMR (100 MHz, CDCl_3)



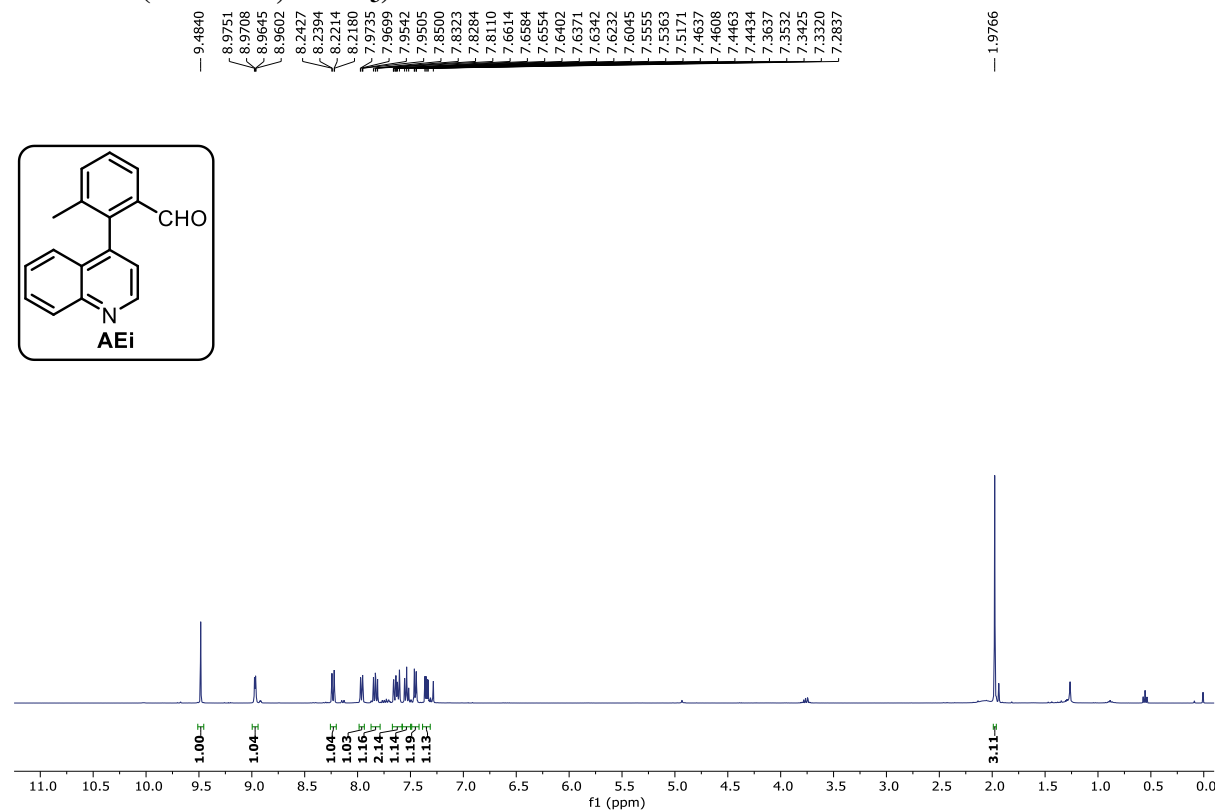
^1H NMR (400 MHz, CDCl_3)



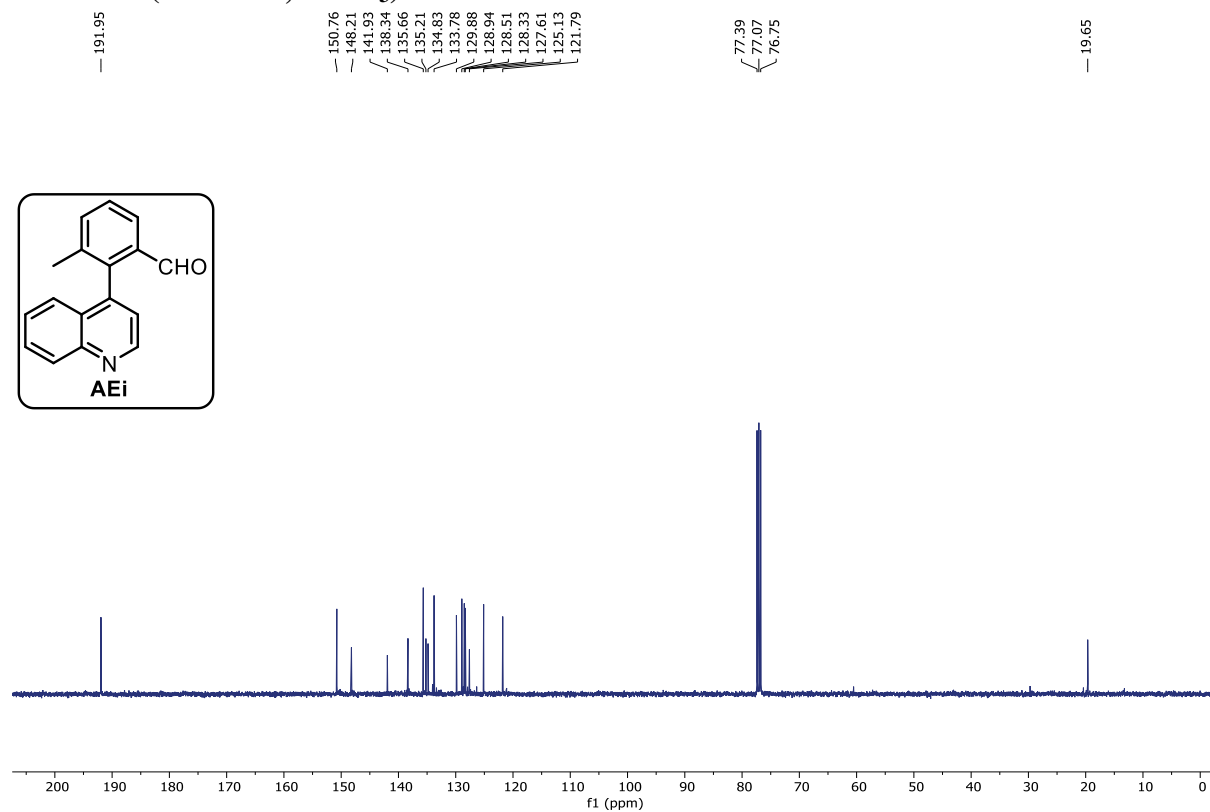
^{13}C NMR (125 MHz, CDCl_3)



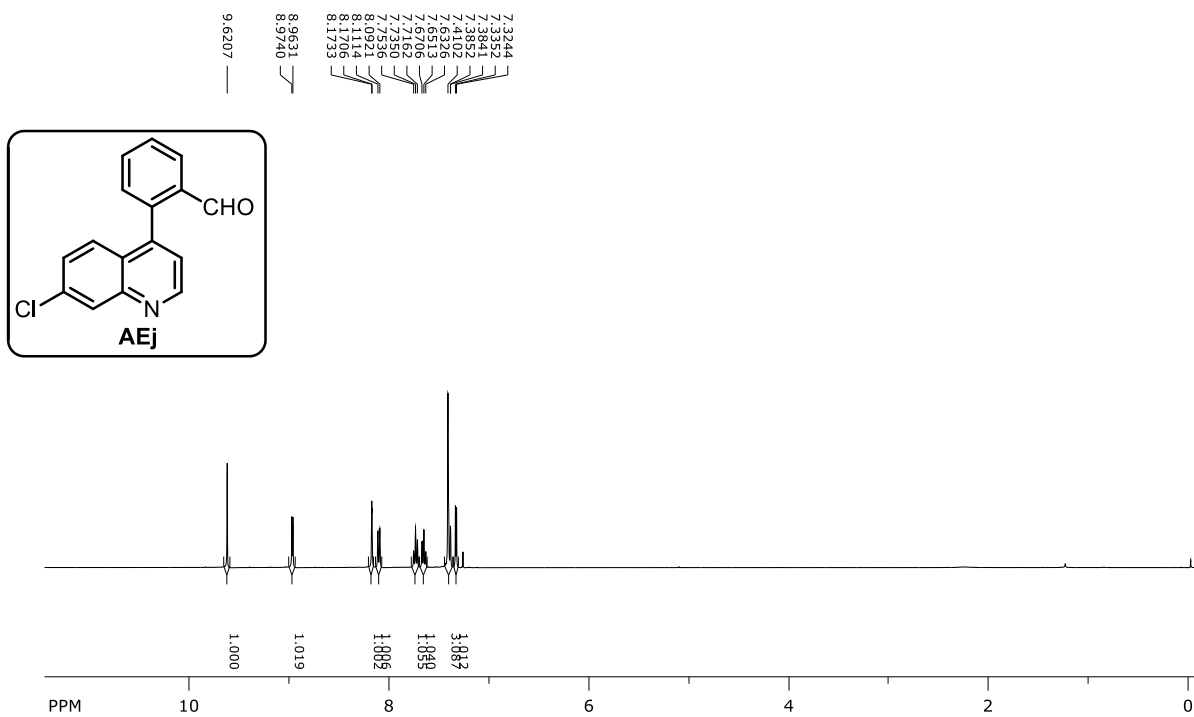
^1H NMR (400 MHz, CDCl_3)



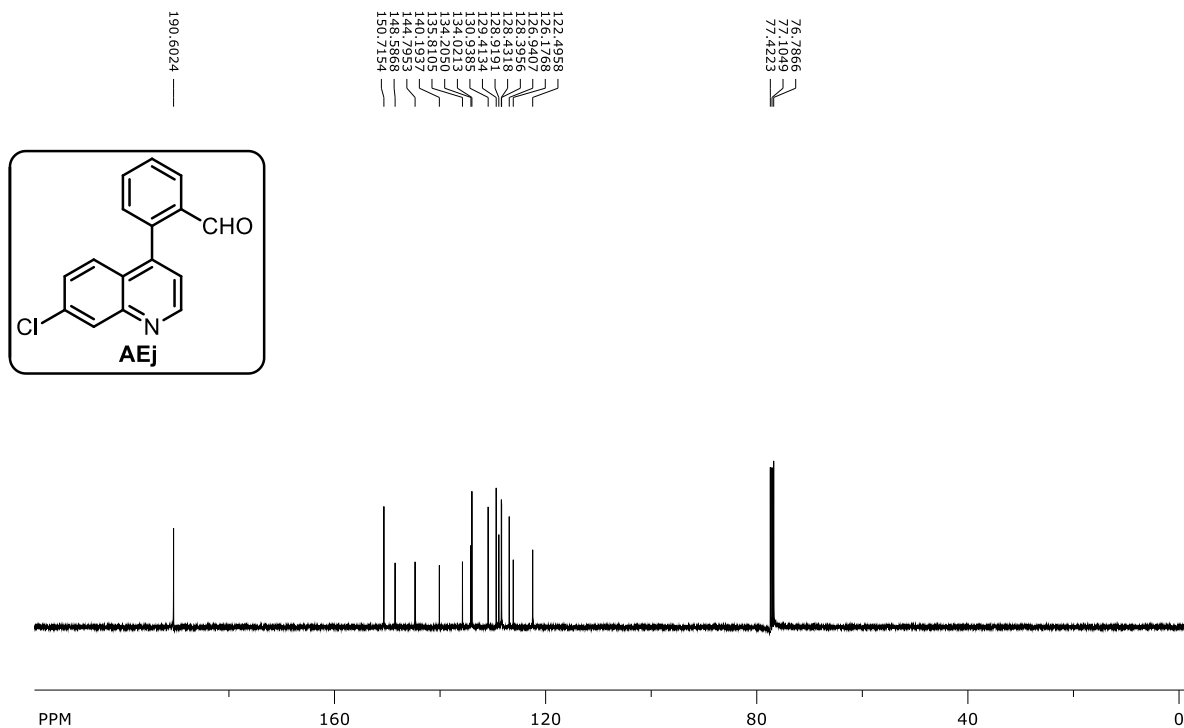
^{13}C NMR (100 MHz, CDCl_3)



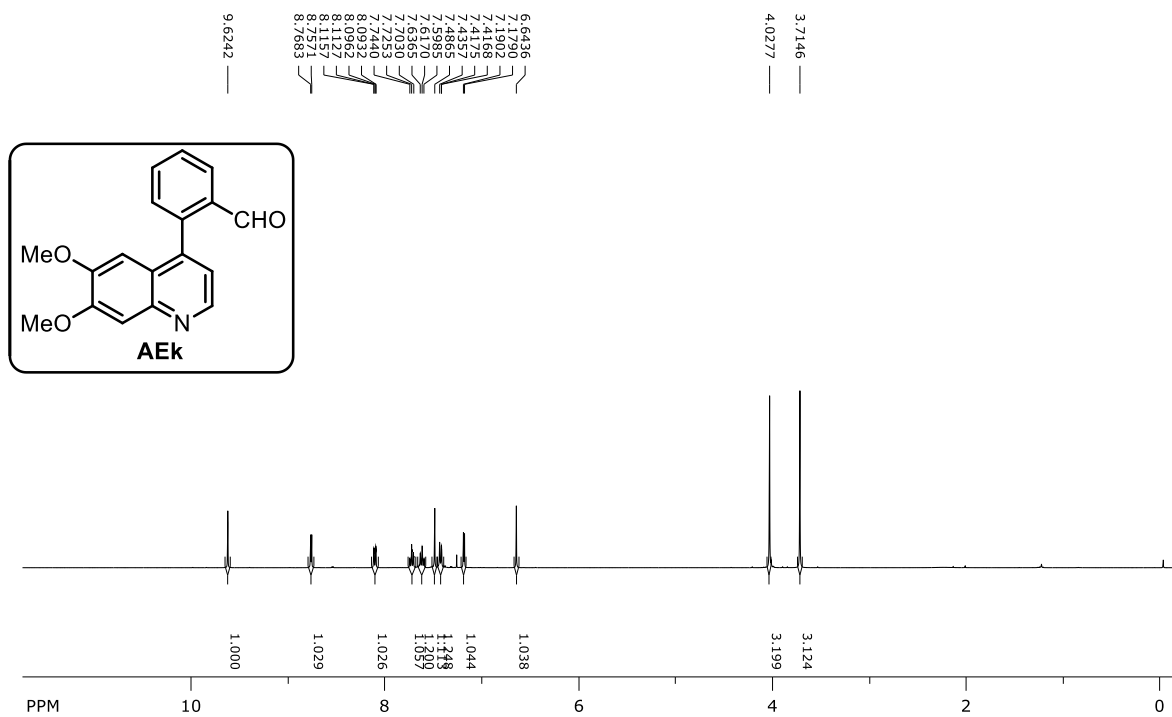
^1H NMR (400 MHz, CDCl_3)



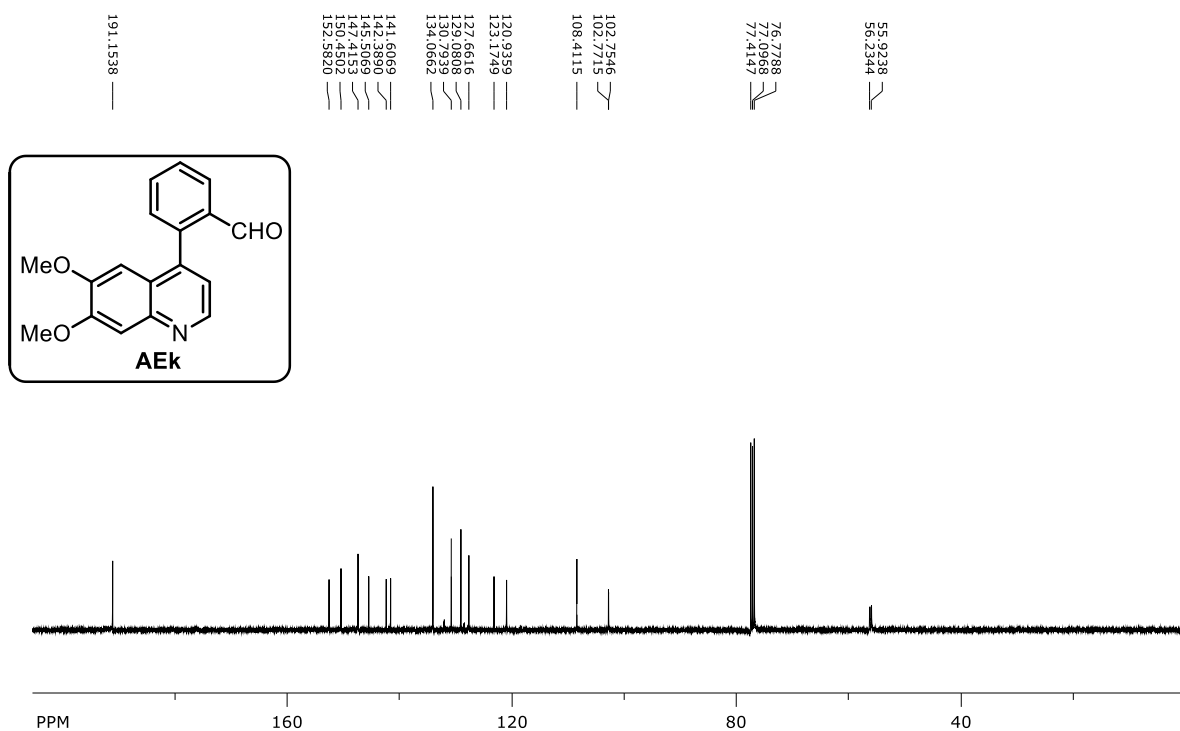
¹³C NMR (100 MHz, CDCl₃)



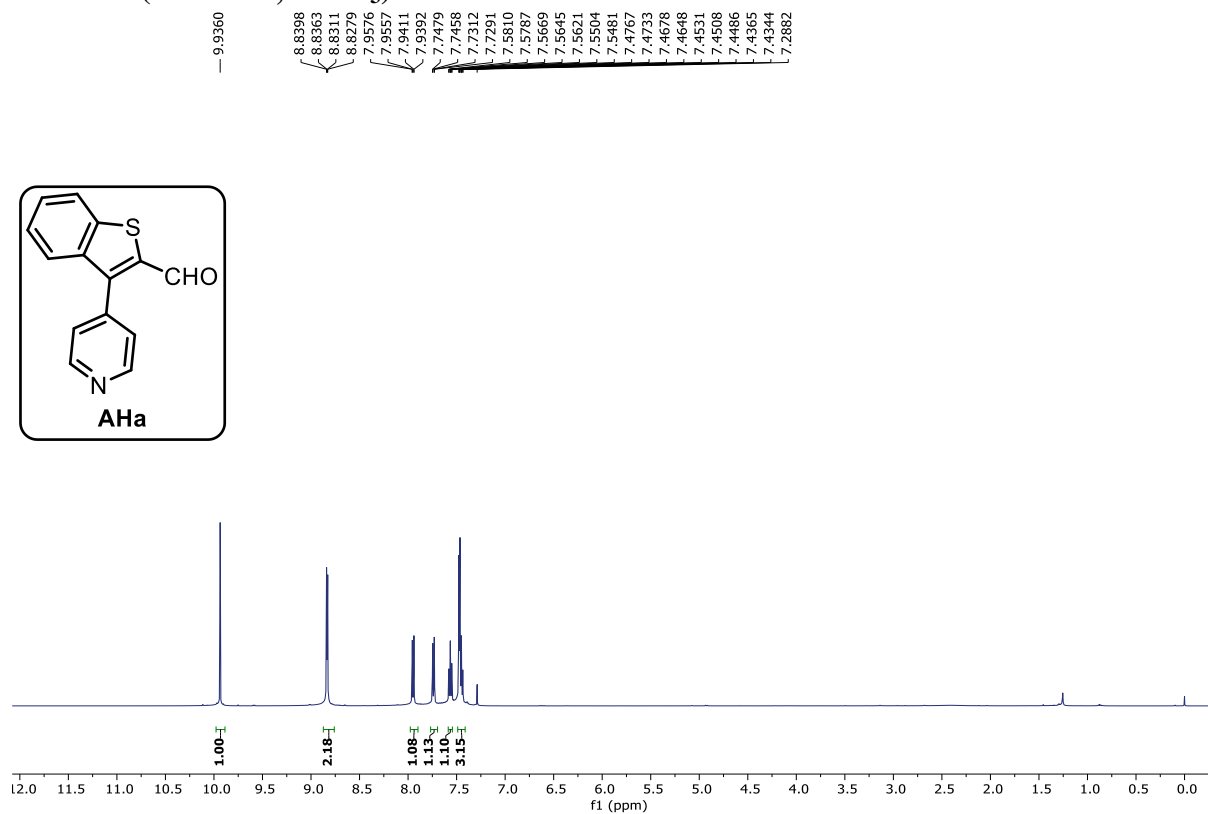
¹H NMR (400 MHz, CDCl₃)



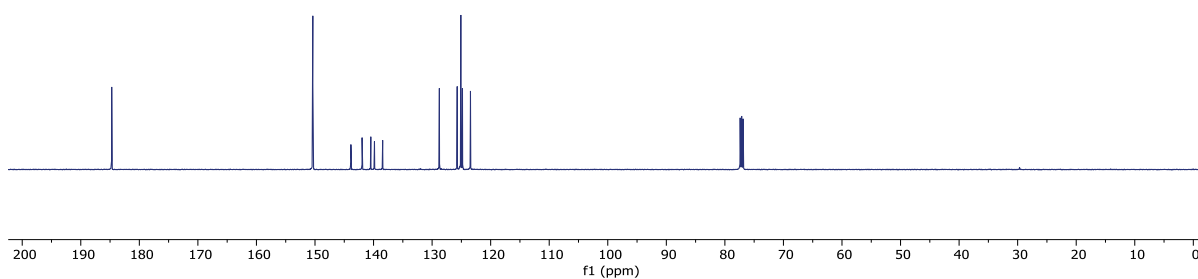
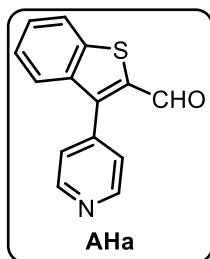
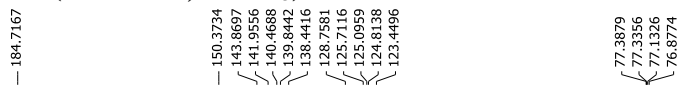
^{13}C NMR (100 MHz, CDCl_3)



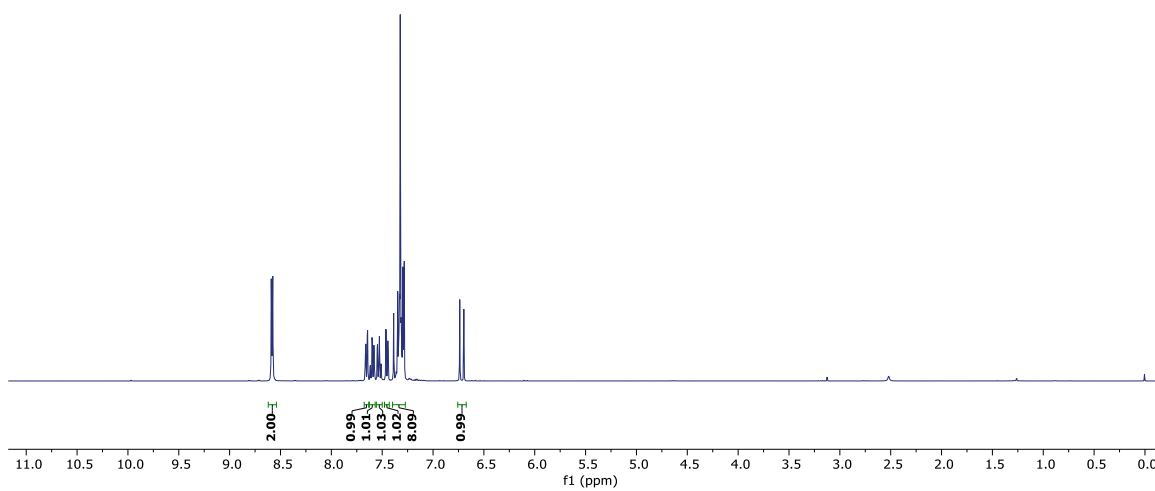
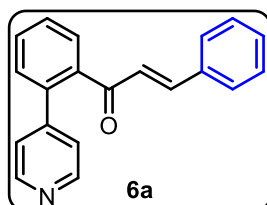
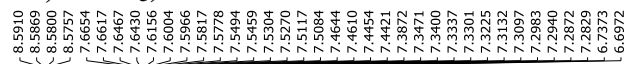
^1H NMR (500 MHz, CDCl_3)



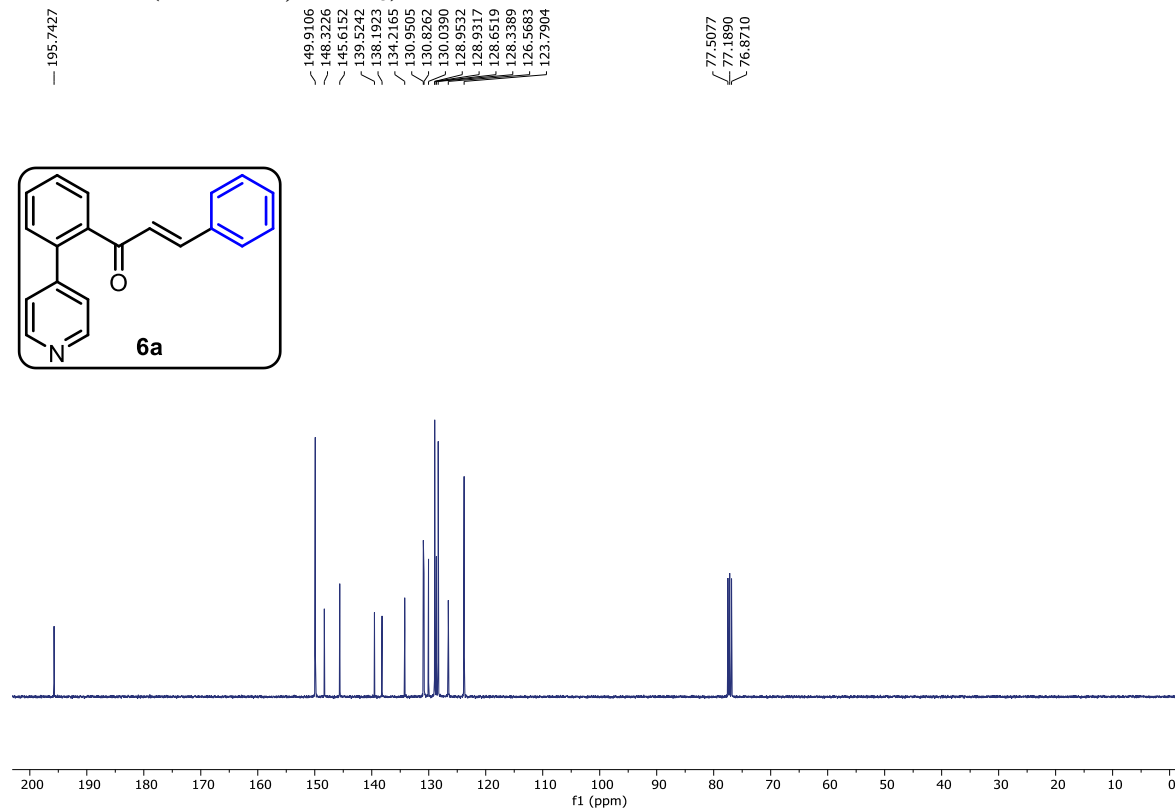
^{13}C NMR (125 MHz, CDCl_3)



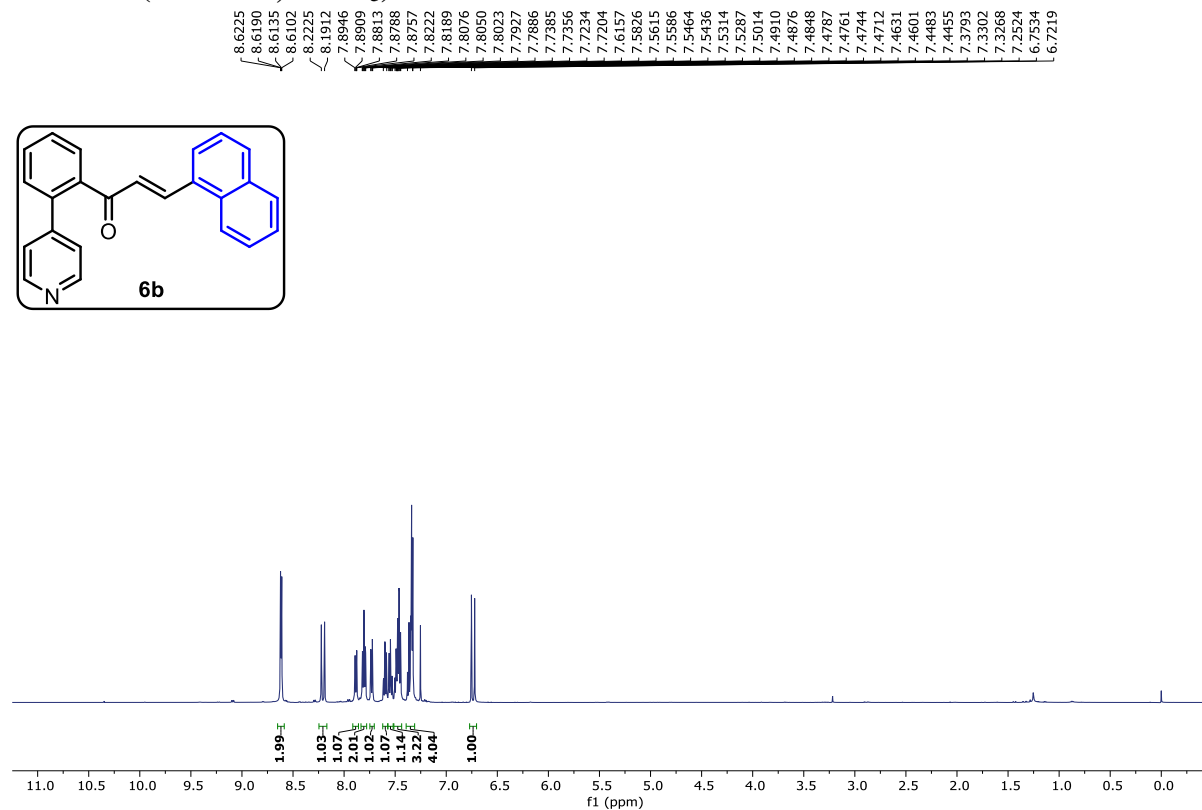
^1H NMR (400 MHz, CDCl_3)



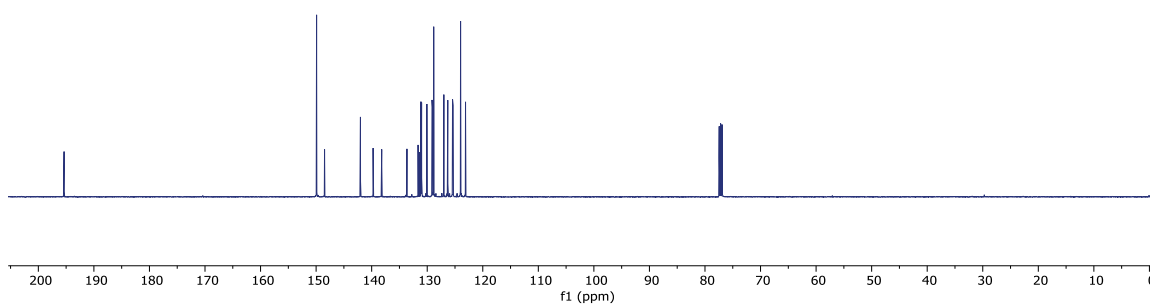
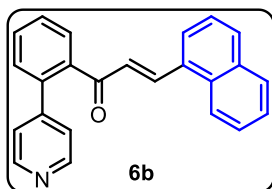
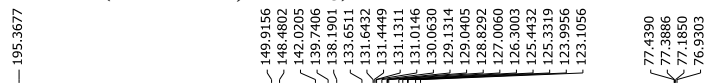
^{13}C NMR (100 MHz, CDCl_3)



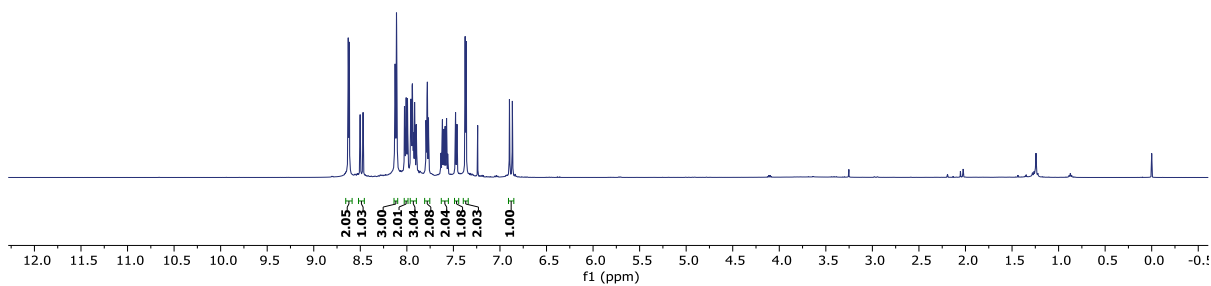
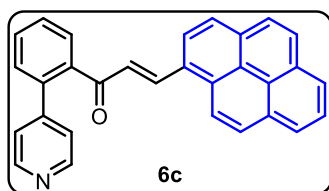
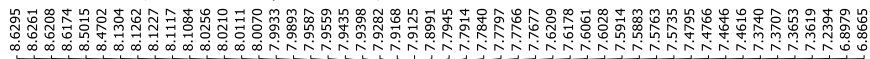
^1H NMR (500 MHz, CDCl_3)



^{13}C NMR (125 MHz, CDCl_3)

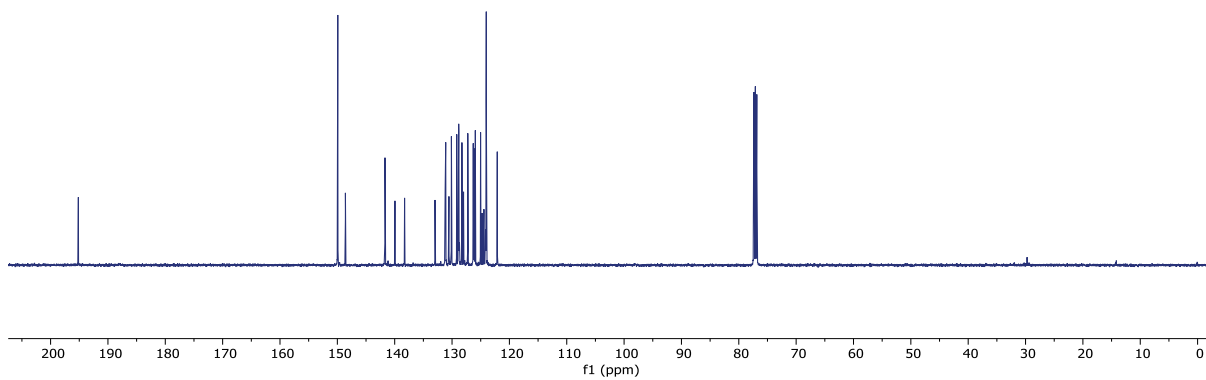
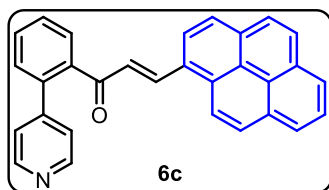


^1H NMR (500 MHz, CDCl_3)



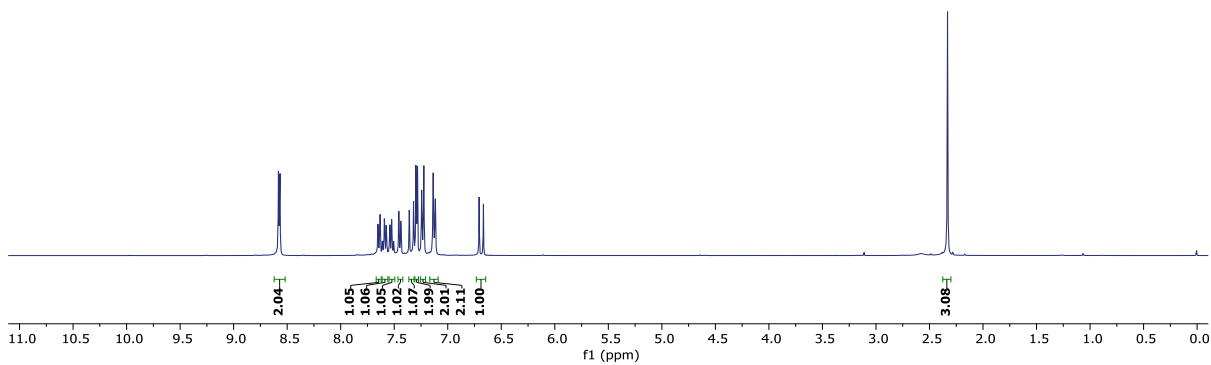
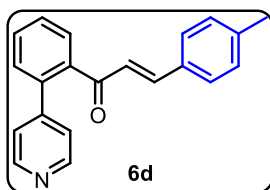
^{13}C NMR (125 MHz, CDCl_3)

195.1591
149.9247
148.5929
141.6910
139.9599
138.2769
132.9703
131.1898
131.1051
130.5421
130.1117
130.0615
129.1764
128.8392
128.8087
128.7236
128.2768
127.9947
127.2530
126.3006
126.1593
125.9410
125.0243
124.7629
124.4409
124.1482
124.0319
122.1113
77.3698
77.1147
76.8598

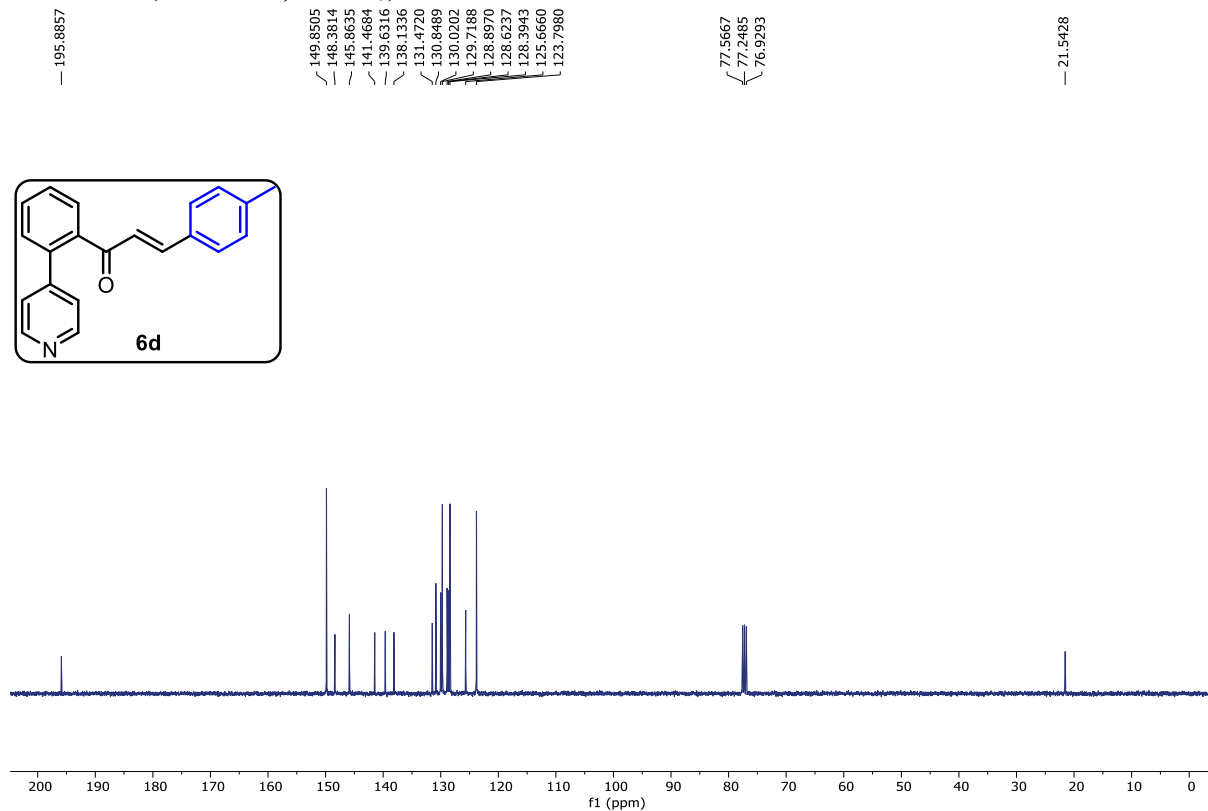


^1H NMR (400 MHz, CDCl_3)

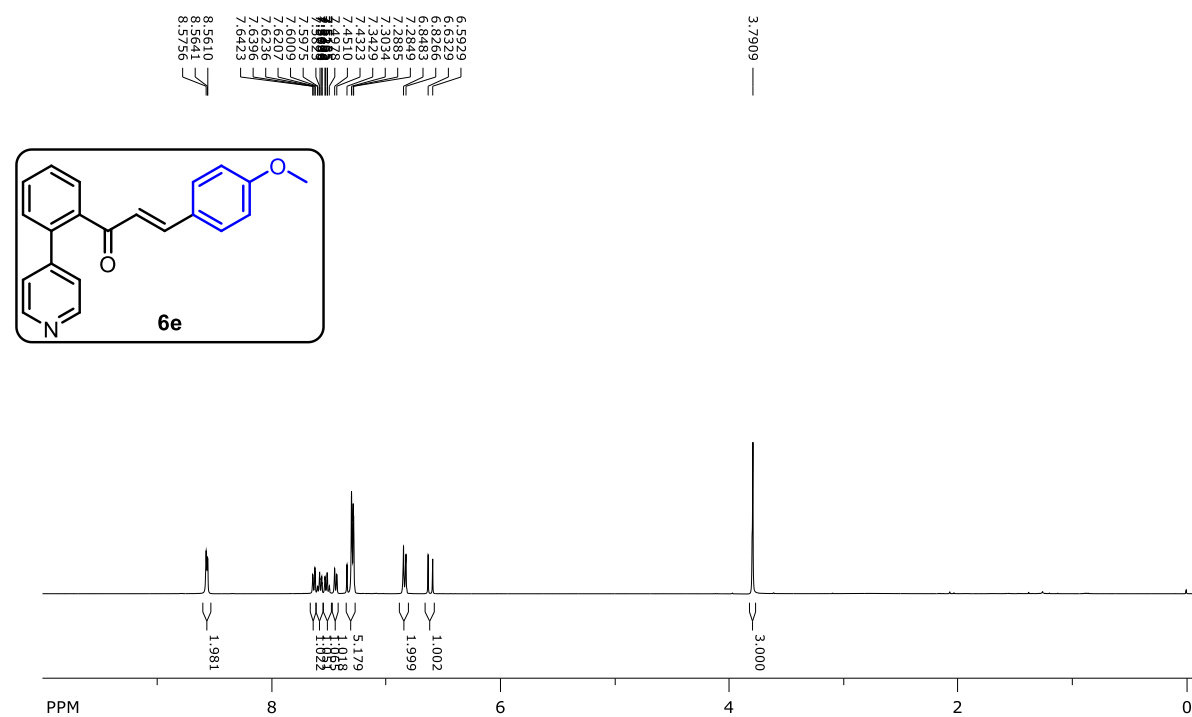
8.5816
8.5774
8.5706
8.5665
7.6536
7.6498
7.6348
7.6311
7.5929
7.5891
7.5741
7.5702
7.5434
7.5399
7.5247
7.5213
7.4584
7.4550
7.4395
7.4361
7.3595
7.3194
7.2976
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7.2236
7.1361
7.1164
6.7088
6.6667
2.3314



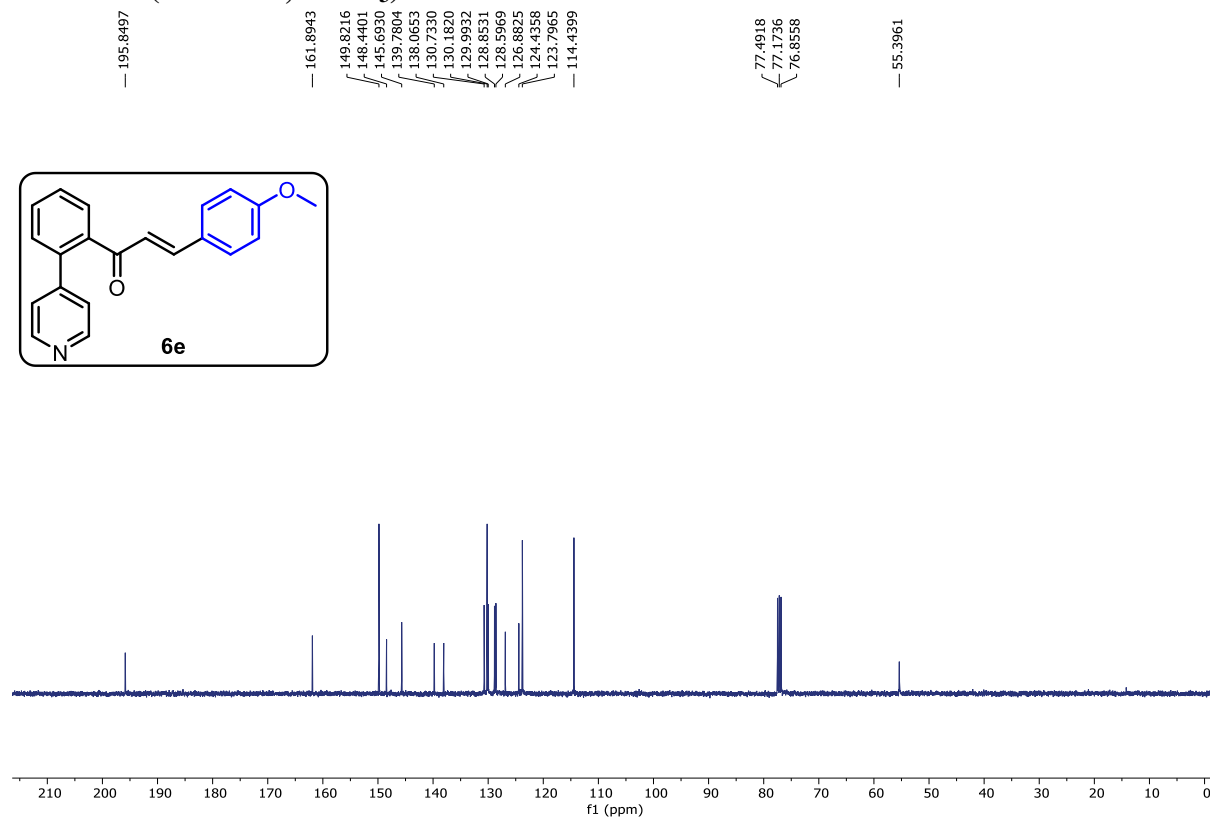
¹³C NMR (100 MHz, CDCl₃)



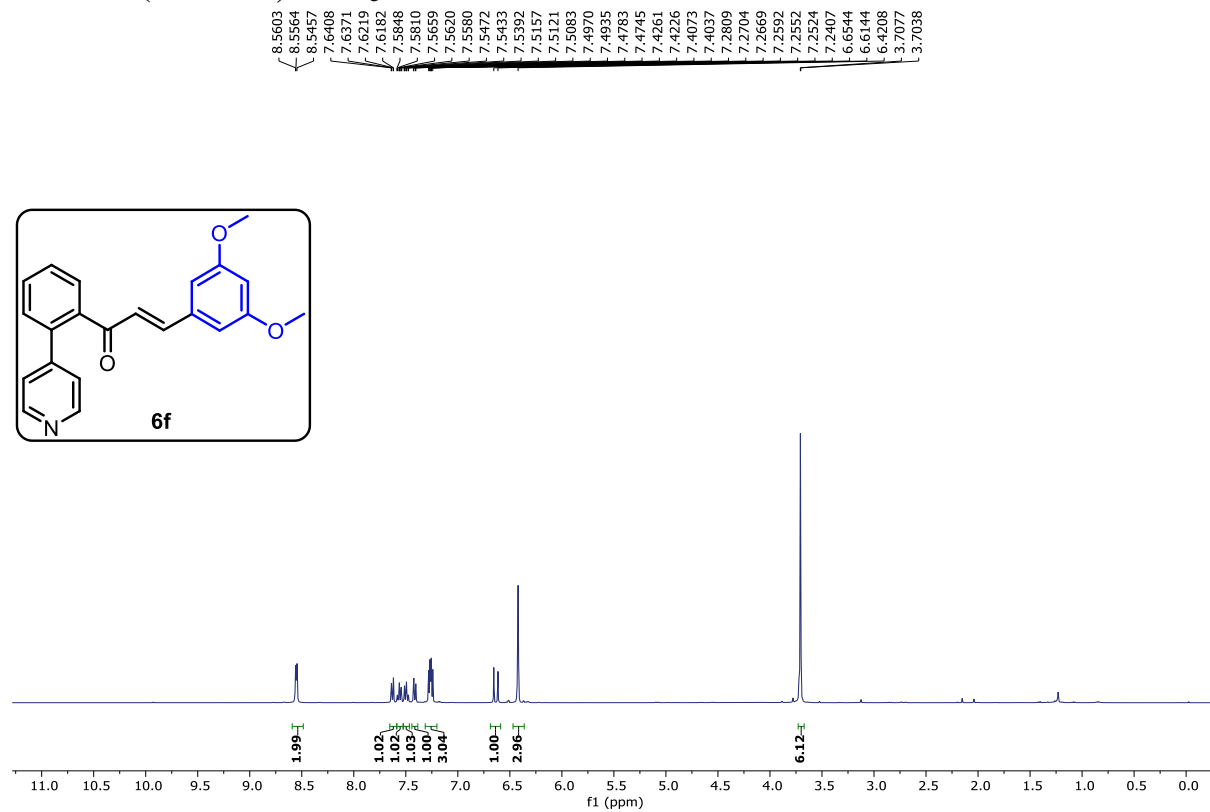
¹H NMR (400 MHz, CDCl₃)



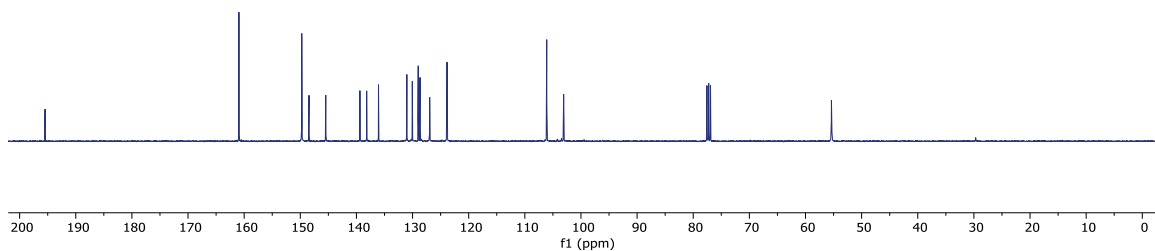
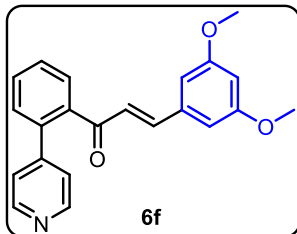
^{13}C NMR (100 MHz, CDCl_3)



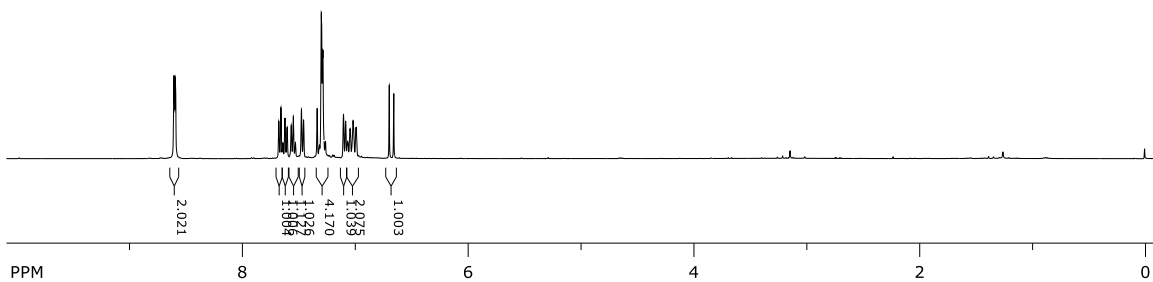
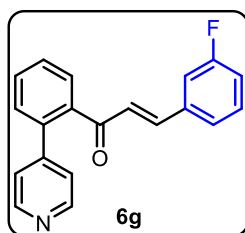
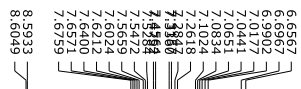
^1H NMR (400 MHz, CDCl_3)



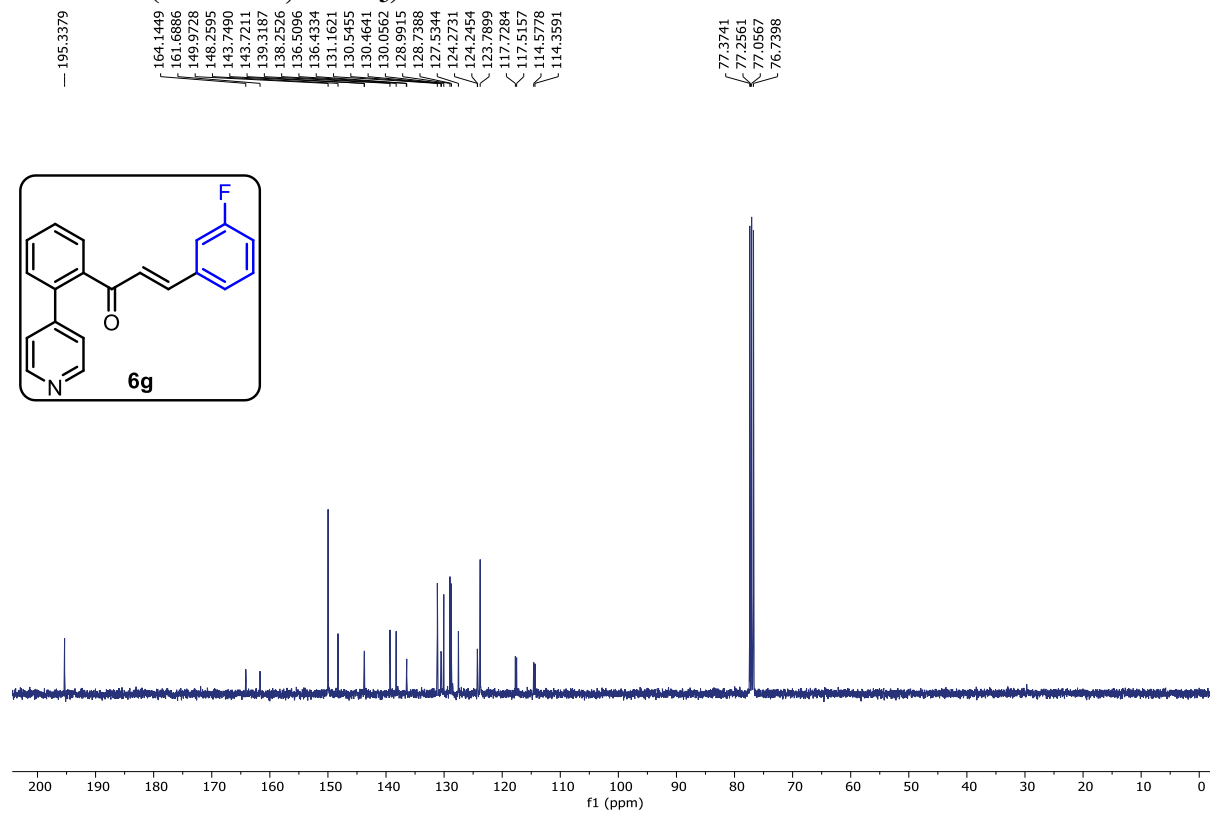
¹³C NMR (100 MHz, CDCl₃)



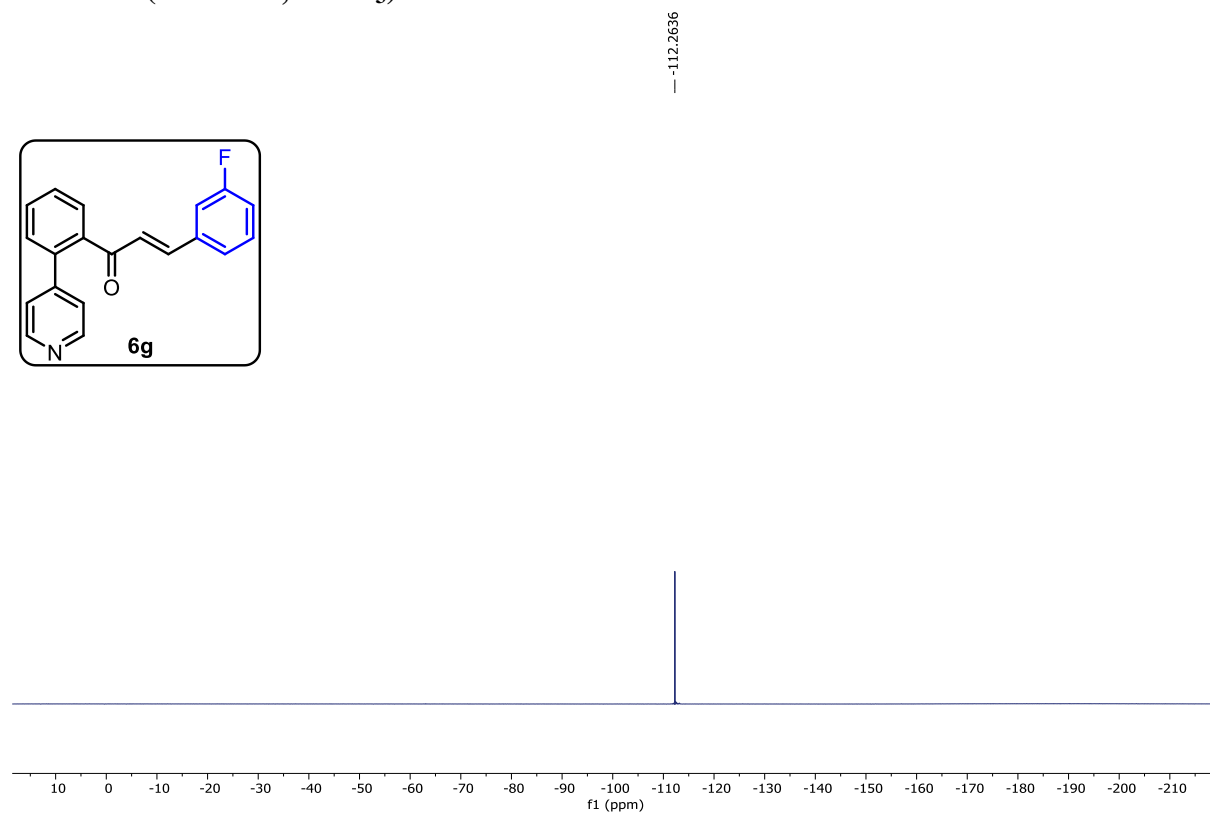
¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)

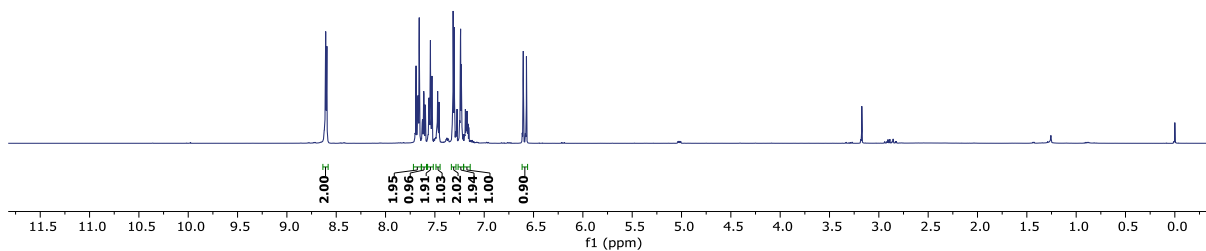
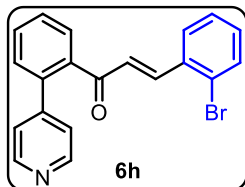


¹⁹F NMR (376 MHz, CDCl₃)



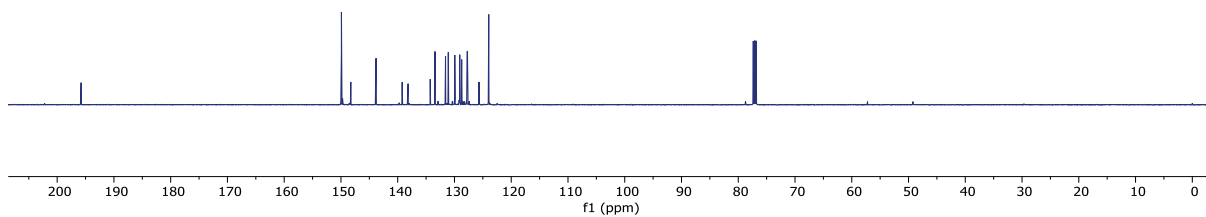
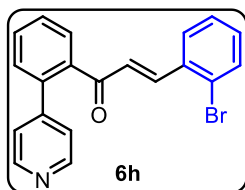
¹H NMR (500 MHz, CDCl₃)

8.6180, 8.6138, 8.6067, 8.6034, 8.5979, 8.5945, 7.6906, 7.6737, 7.6706, 7.6653, 7.6586, 7.6556, 7.6285, 7.6235, 7.6134, 7.6105, 7.5983, 7.5953, 7.5625, 7.5598, 7.5547, 7.5506, 7.5472, 7.5450, 7.5433, 7.5385, 7.5322, 7.5298, 7.5277, 7.4740, 7.4714, 7.4588, 7.4561, 7.3256, 7.3221, 7.3028, 7.2750, 7.2511, 7.2486, 7.2277, 7.1905, 7.1679, 7.1632, 6.6040, 6.5719



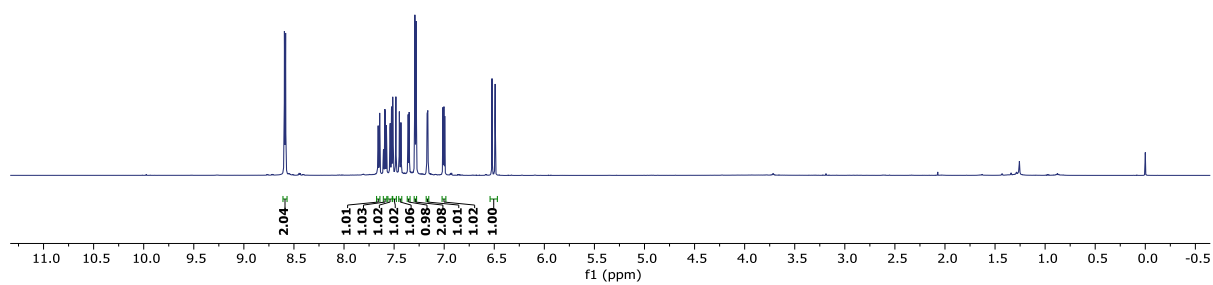
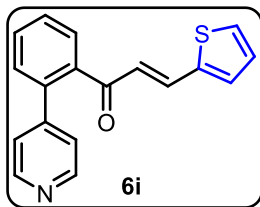
¹³C NMR (125 MHz, CDCl₃)

195.7839, 149.9198, 148.2659, 143.8163, 139.2190, 138.1725, 134.2728, 133.4277, 131.5760, 131.0867, 129.9455, 129.1533, 129.0687, 128.7269, 127.7748, 127.7119, 125.6855, 123.9598, 77.3529, 77.0986, 76.8448



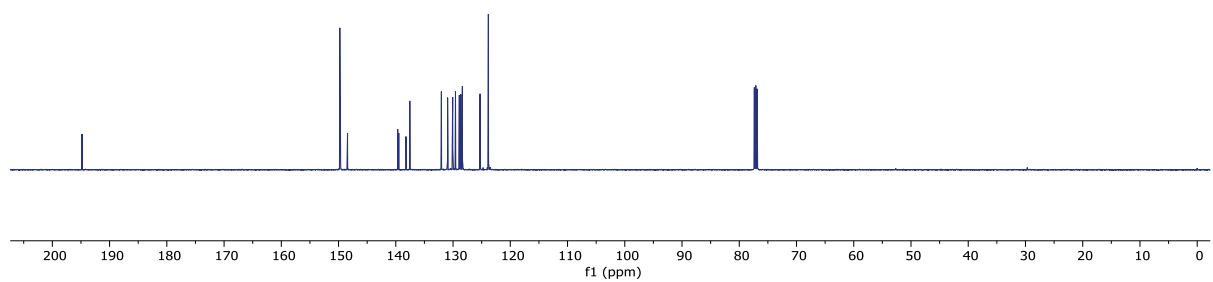
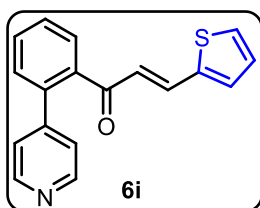
¹H NMR (400 MHz, CDCl₃)

8.5951
8.5828
7.6615
7.6436
7.6068
7.6038
7.5917
7.5888
7.5766
7.5736
7.5412
7.5236
7.5086
7.4838
7.4823
7.4608
7.4480
7.4453
7.4331
7.4319
7.4299
7.3615
7.3596
7.3575
7.3515
7.3495
7.3474
7.2927
7.2806
7.1719
7.1617
7.0118
7.0045
7.0016
6.9944
6.5223
6.4910



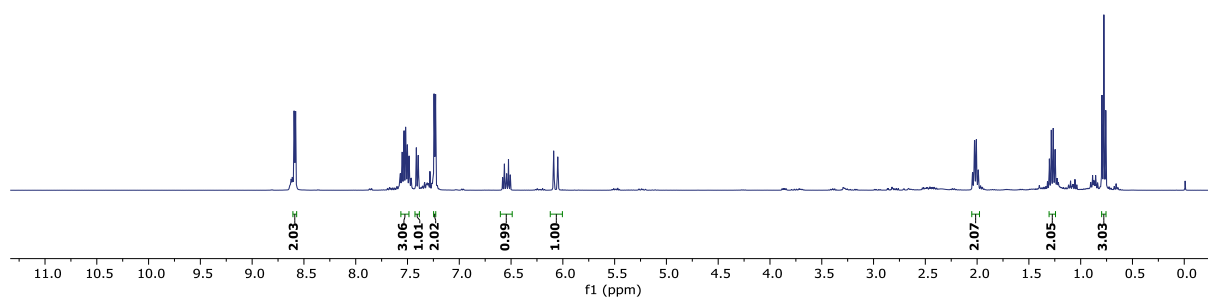
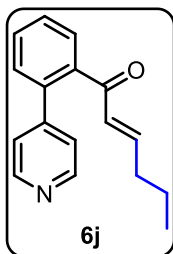
¹³C NMR (125 MHz, CDCl₃)

194.8175
149.7783
148.4309
139.6641
139.4803
138.2190
137.5516
132.0354
130.9483
130.0588
129.5709
128.9033
128.6553
128.3716
125.2816
123.8288
77.3652
77.1103
76.8570



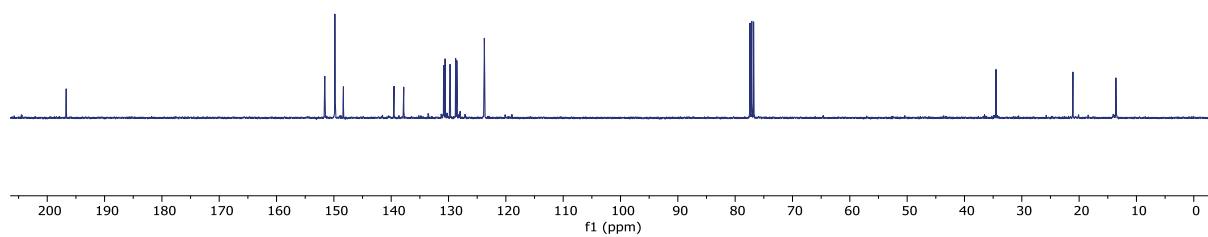
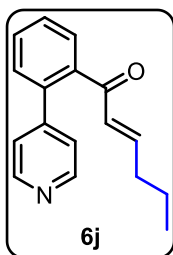
¹H NMR (400 MHz, CDCl₃)

8.6123
8.5952
8.5913
8.5842
8.5799
8.5736
7.5687
7.5554
7.5507
7.5376
7.5327
7.5185
7.5141
7.5042
7.5008
7.4869
7.4840
7.4645
7.4167
7.4134
7.3976
7.3946
7.2833
7.2545
7.2512
7.2437
7.2398
7.2327
7.2284
6.5816
6.5644
6.5471
6.5423
6.5249
6.5076
6.0927
6.0888
6.0850
6.0531
6.0494
6.0457
2.0461
2.0424
2.0283
2.0246
2.0100
2.0063
1.9920
1.9882
1.3204
1.3020
1.2837
1.2652
1.2470
1.2286
0.7945
0.7761
0.7576



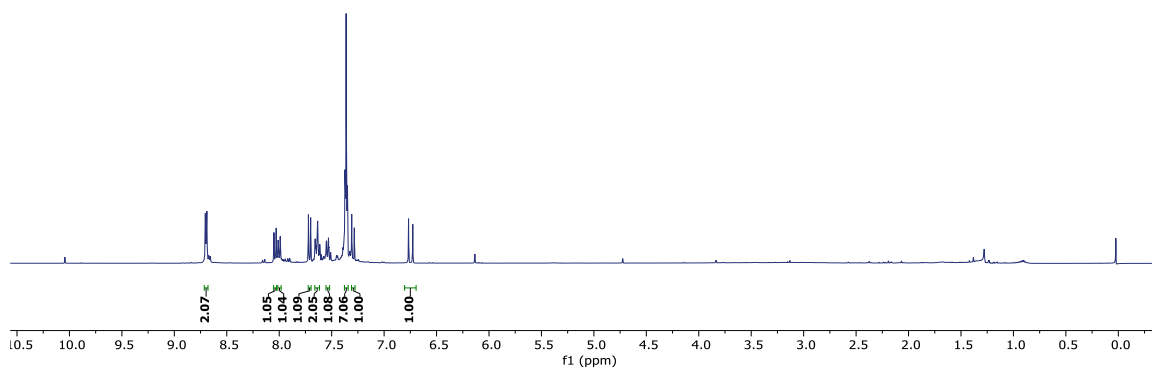
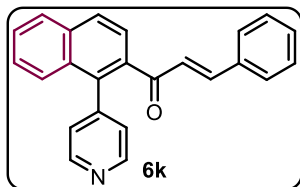
¹³C NMR (100 MHz, CDCl₃)

196.6996
151.5850
149.8287
148.3373
139.5061
137.8111
130.8524
130.6017
129.7359
128.7550
128.5532
123.7968
123.7531
77.4208
77.1028
76.7846
34.5057
21.0828
13.5881



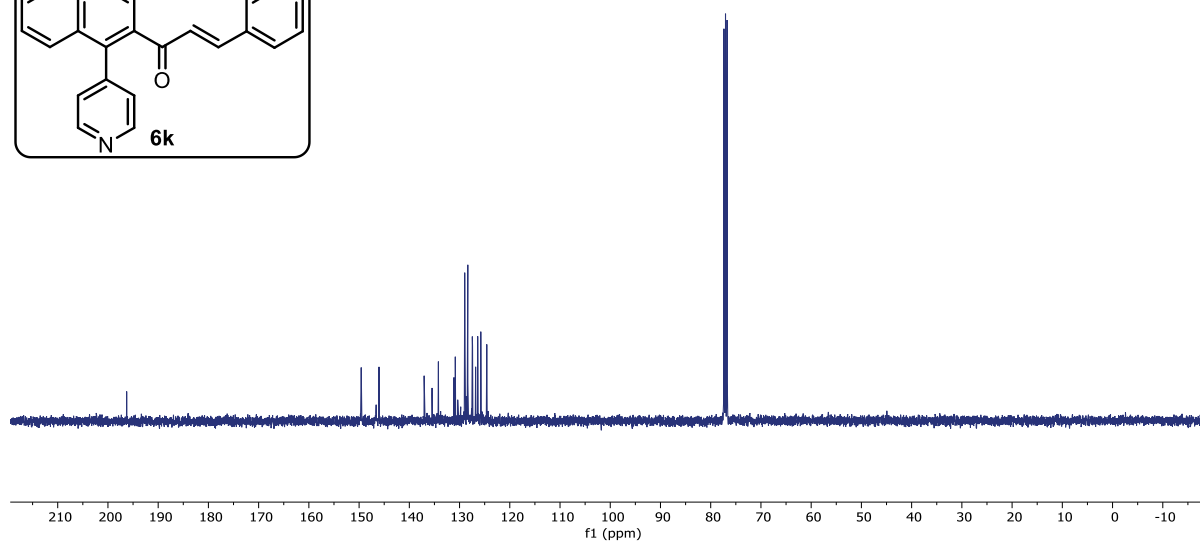
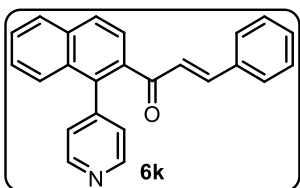
¹H NMR (400 MHz, CDCl₃)

8.7048
8.7007
8.6838
8.6897
8.0501
8.0389
8.0107
8.0070
7.9904
7.9870
7.7212
7.7001
7.6602
7.6568
7.6513
7.6378
7.6336
7.6307
7.6136
7.5510
7.5476
7.5305
7.3729
7.3687
7.3621
7.3577
7.3490
7.3086
7.2839
6.7669
6.7267



¹³C NMR (100 MHz, CDCl₃)

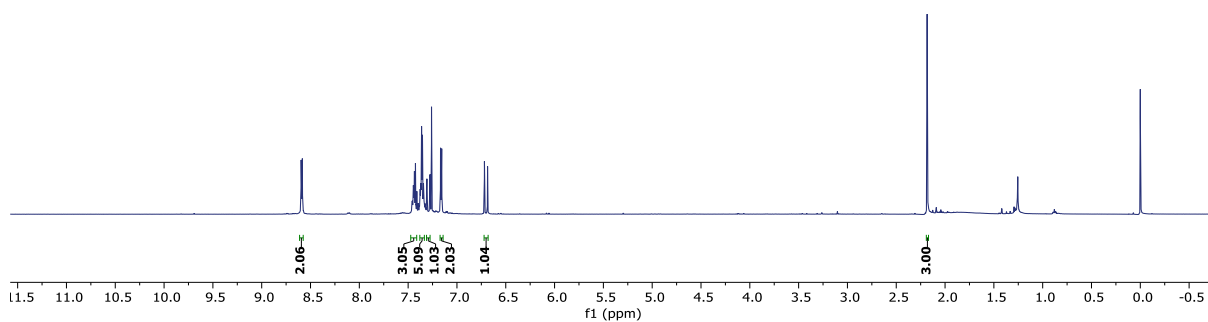
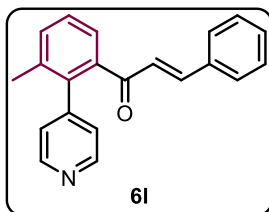
196.2766
149.5678
146.0360
137.0490
135.4674
134.2263
134.2122
131.1252
130.8498
128.9738
128.8912
128.6216
128.3839
128.3883
127.4579
127.4131
126.8038
126.3864
125.7745
124.5830
77.3731
77.0551
76.7361



¹H NMR (500 MHz, CDCl₃)

8.5959
8.5925
8.5871
8.5838
7.4477
7.4463
7.4420
7.4364
7.4352
7.4325
7.4307
7.4249
7.4112
7.4094
7.3784
7.3763
7.3716
7.3691
7.3648
7.3624
7.3592
7.3575
7.3530
7.3430
7.3385
7.3079
7.2757
7.2605
7.1681
7.1648
7.1593
7.1560
6.7183
6.6862

-2.1834



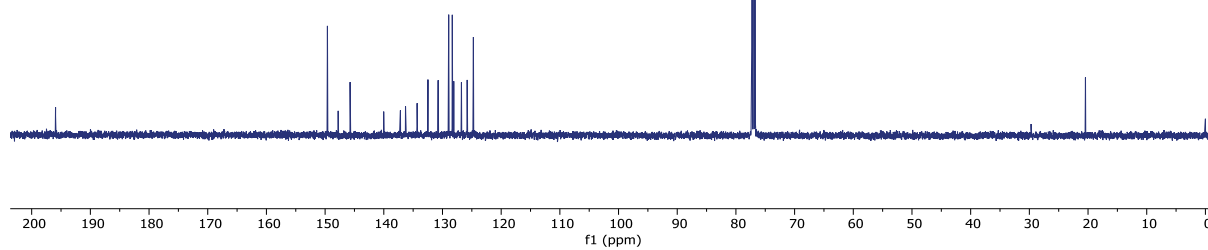
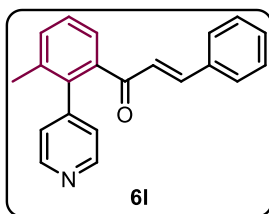
¹³C NMR (125 MHz, CDCl₃)

195.9321

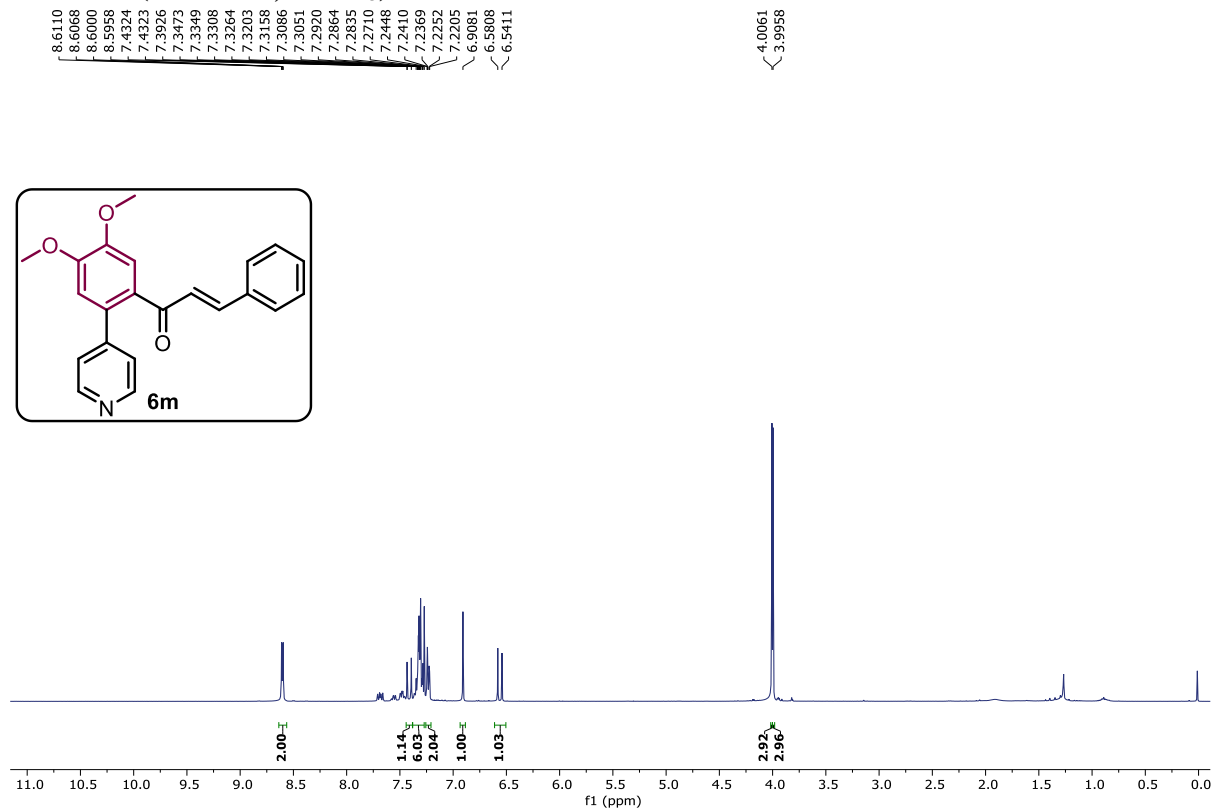
149.6039
147.7723
145.7409
140.0194
137.1657
136.2988
134.3176
132.4788
130.7355
128.9425
128.3267
128.0808
126.7669
125.7860
124.7318

77.2763
77.0202
76.7678

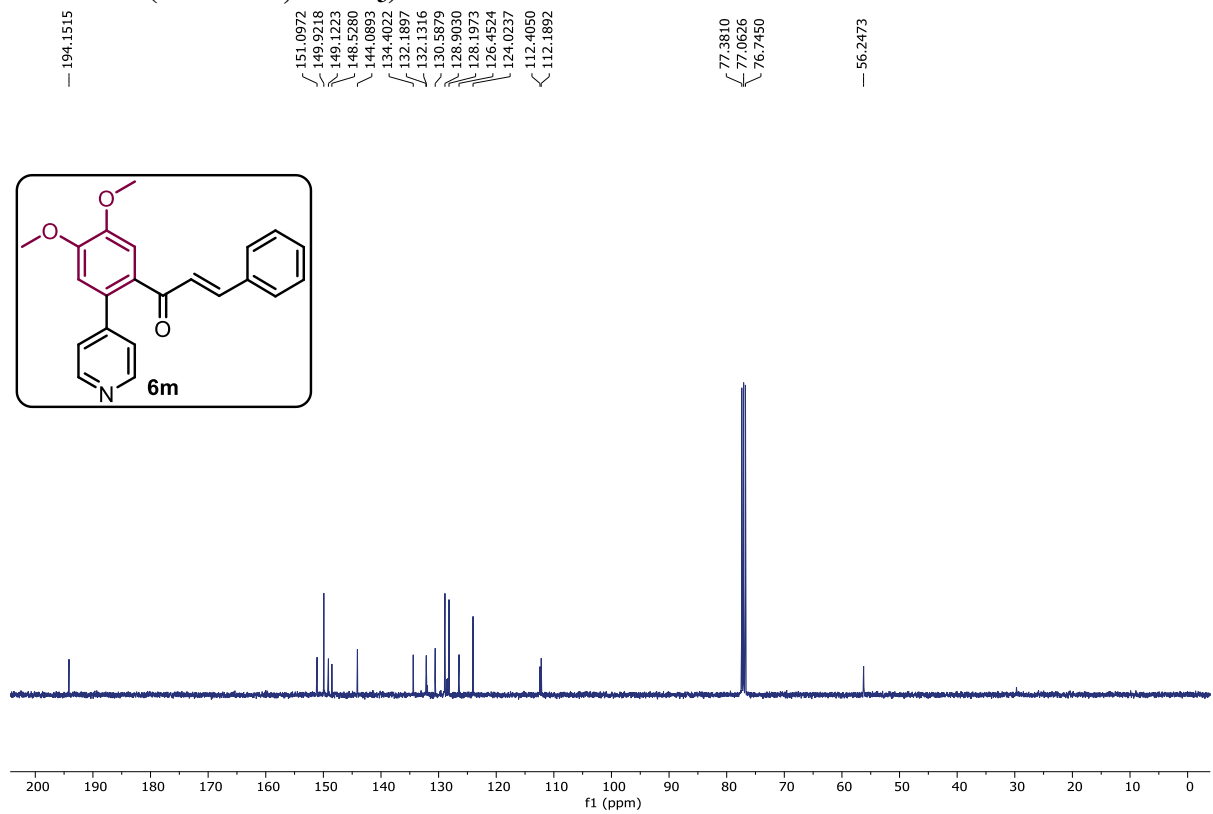
20.4385



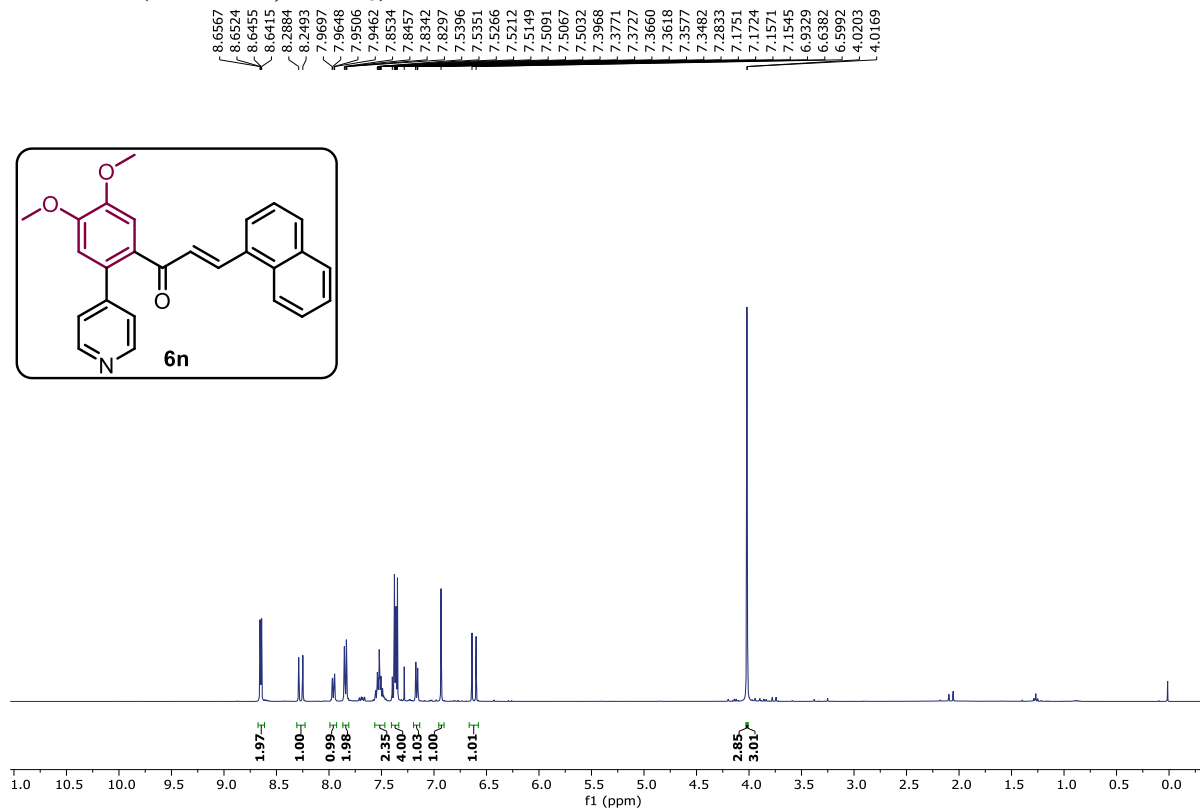
¹H NMR (400 MHz, CDCl₃)



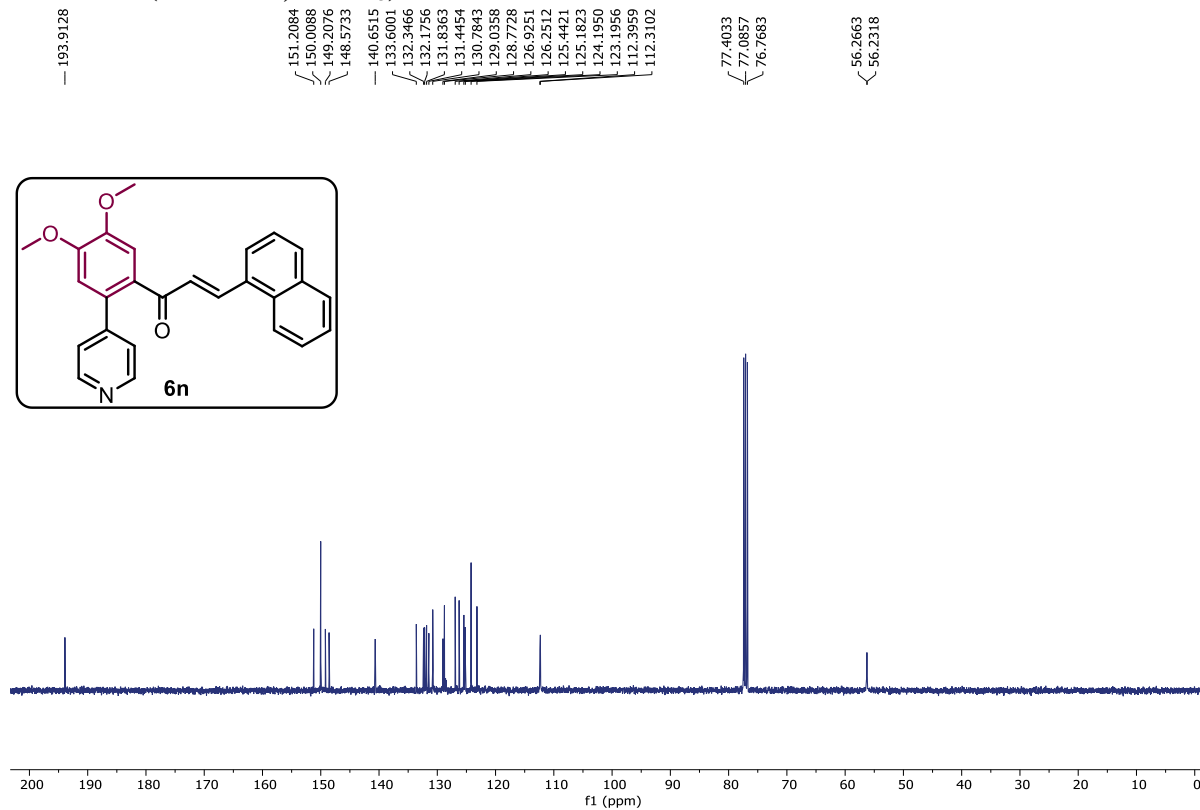
¹³C NMR (100 MHz, CDCl₃)



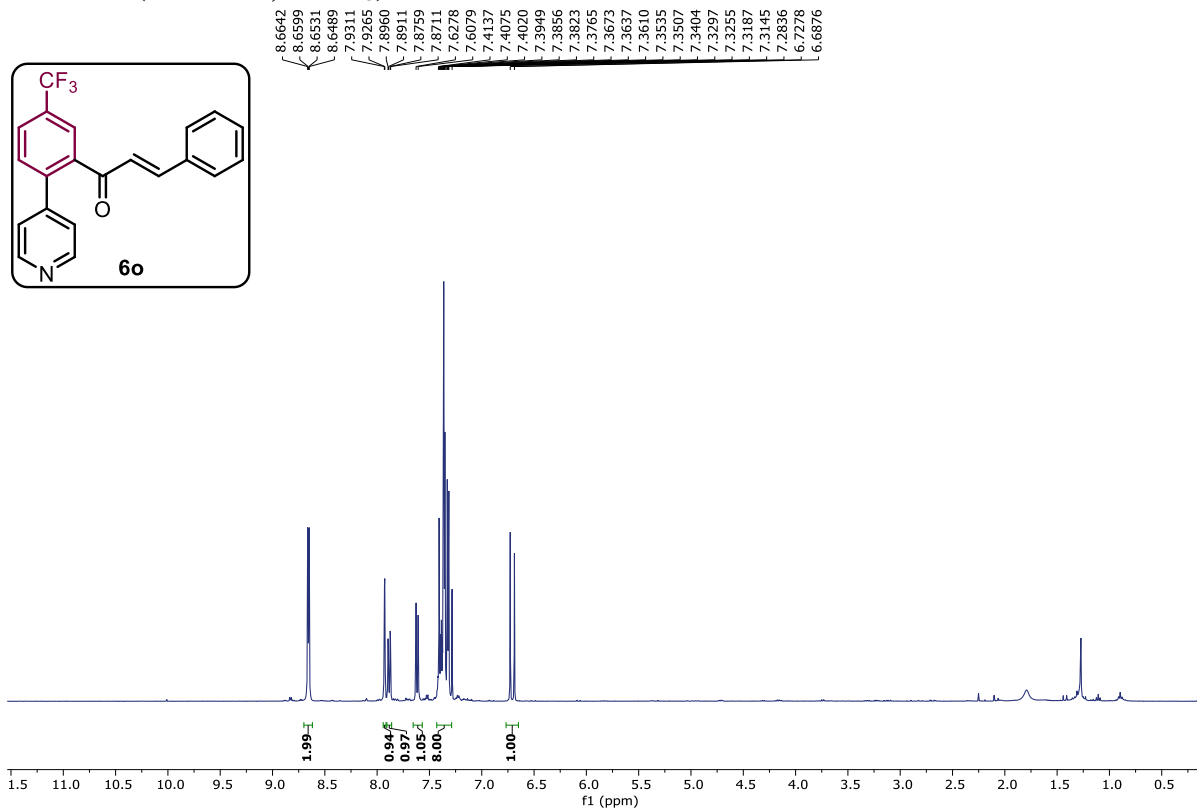
¹H NMR (400 MHz, CDCl₃)



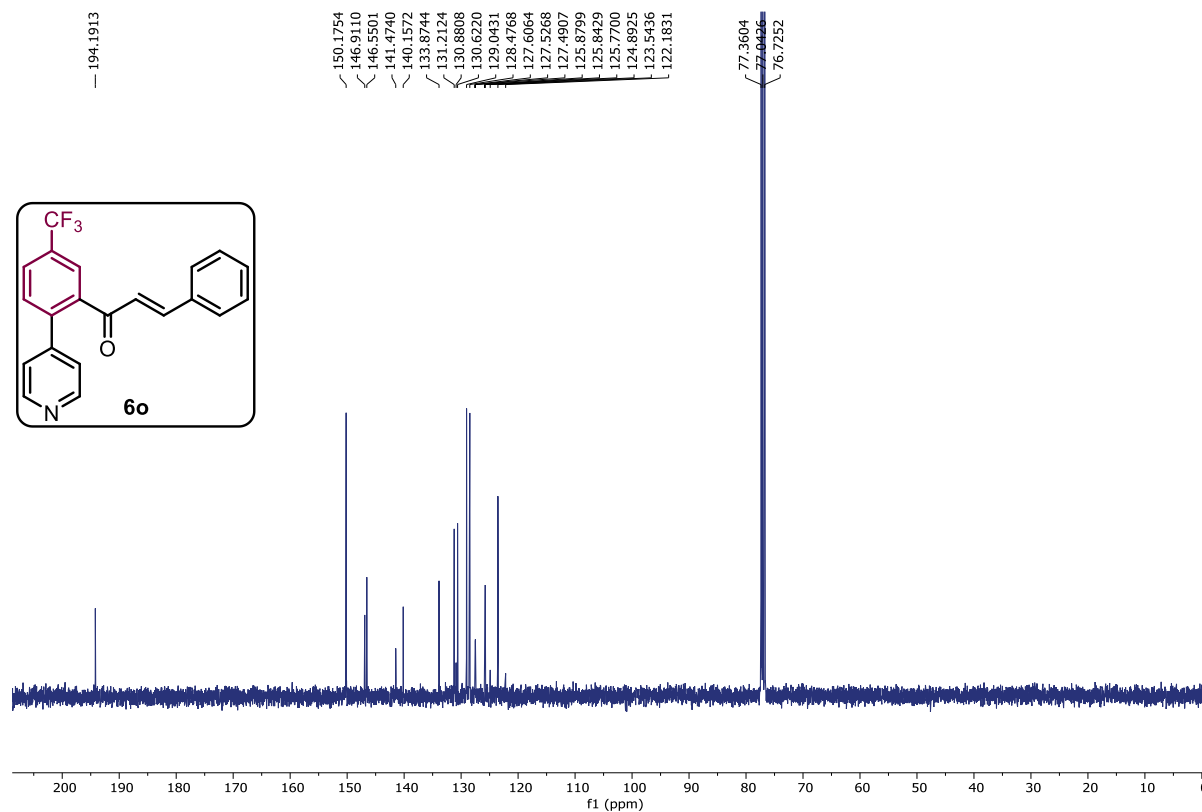
¹³C NMR (100 MHz, CDCl₃)



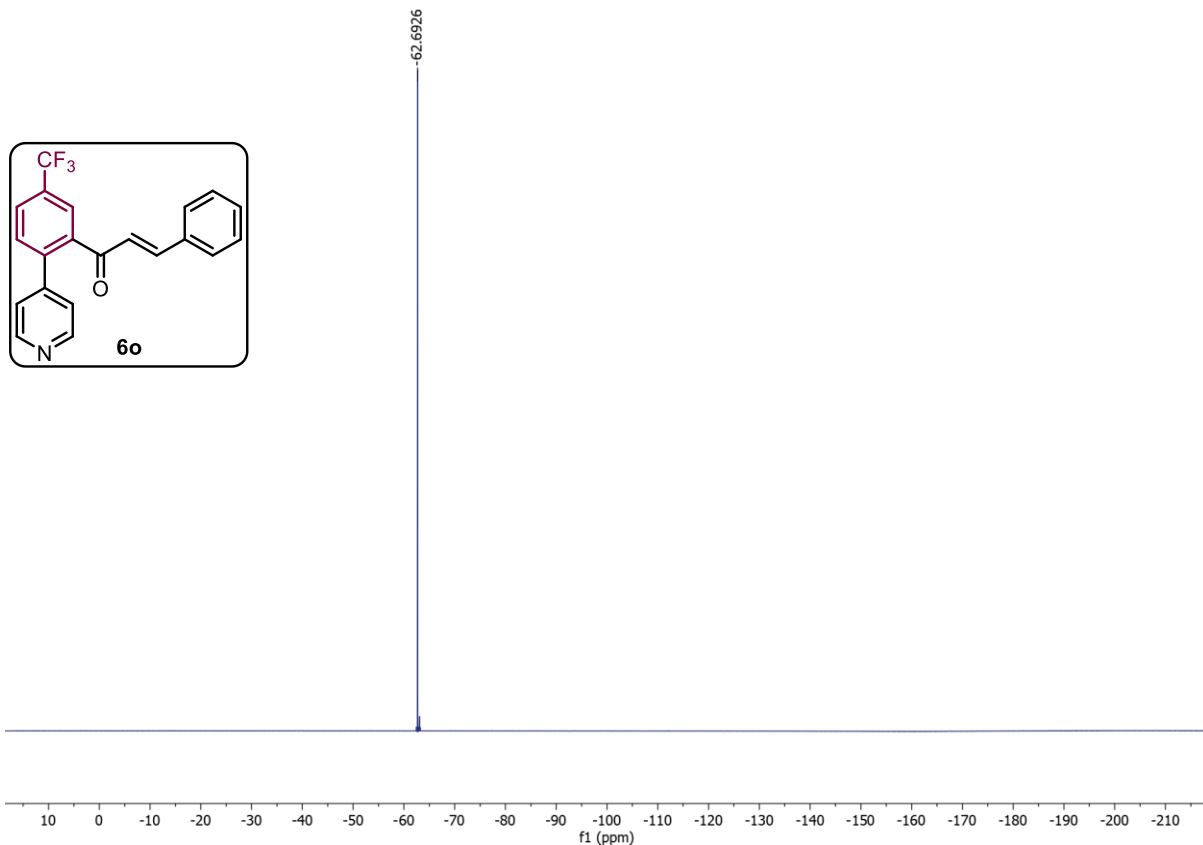
¹H NMR (400 MHz, CDCl₃)



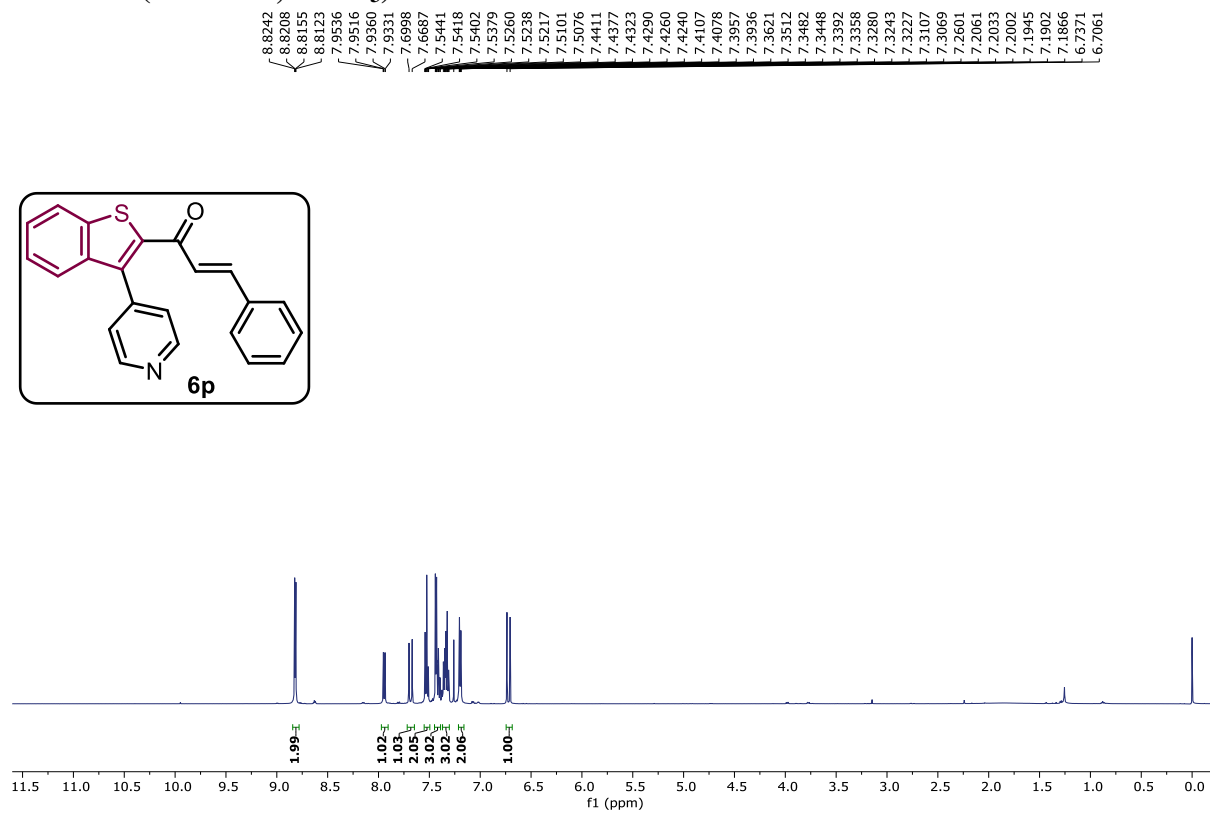
¹³C NMR (100 MHz, CDCl₃)



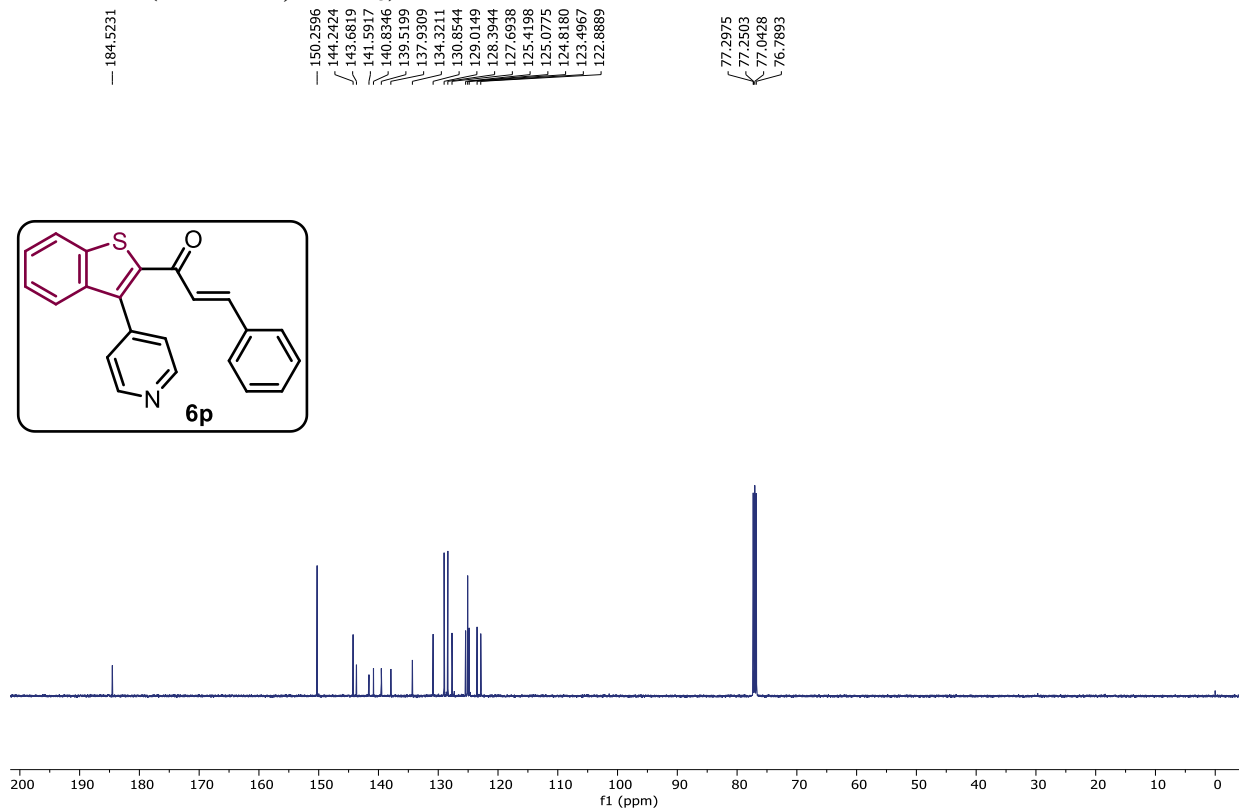
^{19}F NMR (376 MHz, CDCl_3)



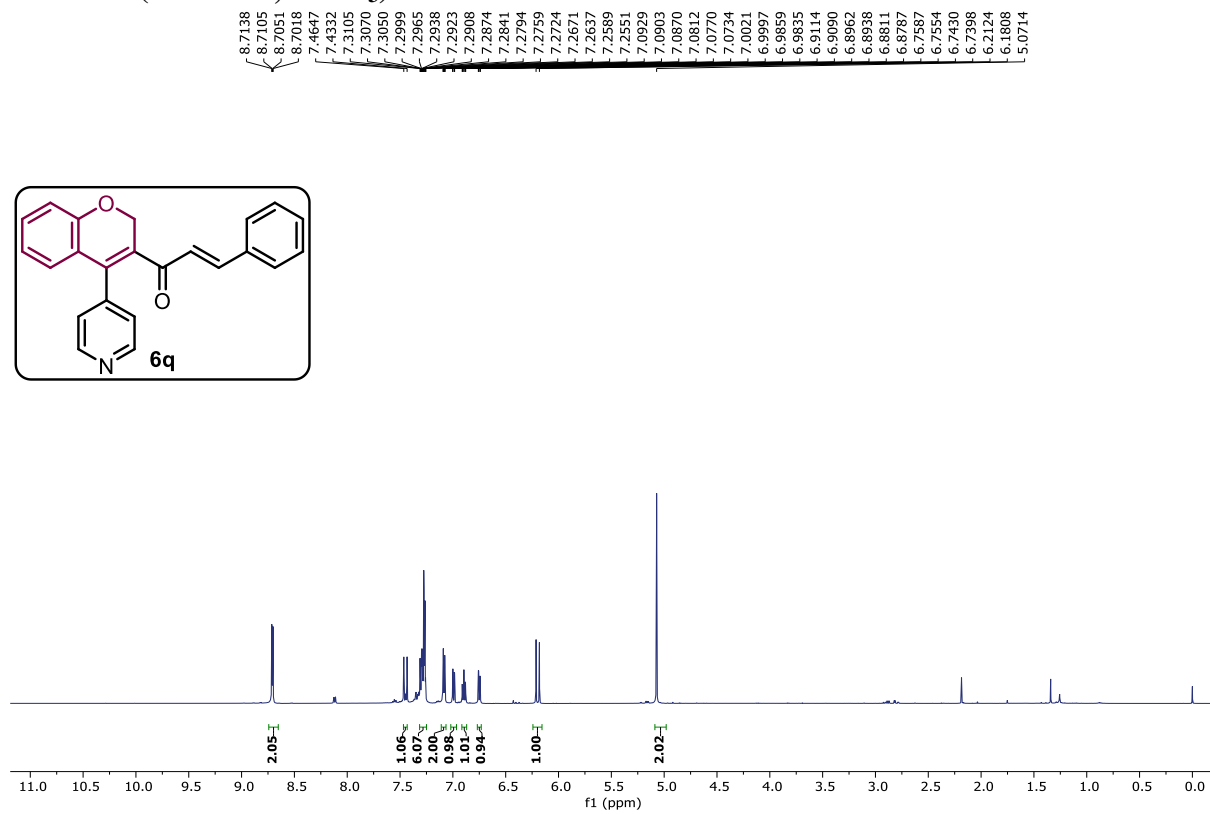
^1H NMR (500 MHz, CDCl_3)



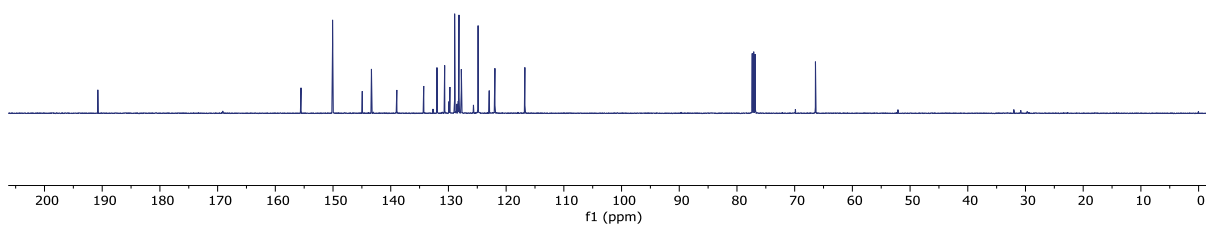
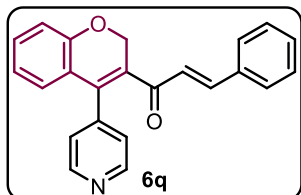
^{13}C NMR (125 MHz, CDCl_3)



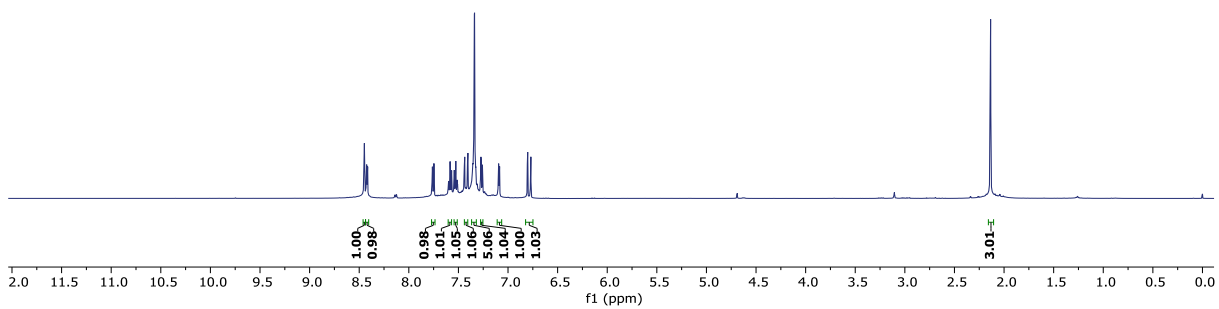
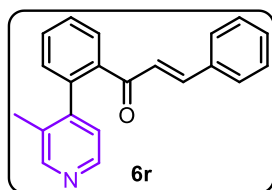
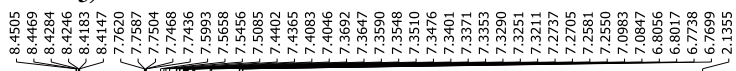
^1H NMR (500 MHz, CDCl_3)



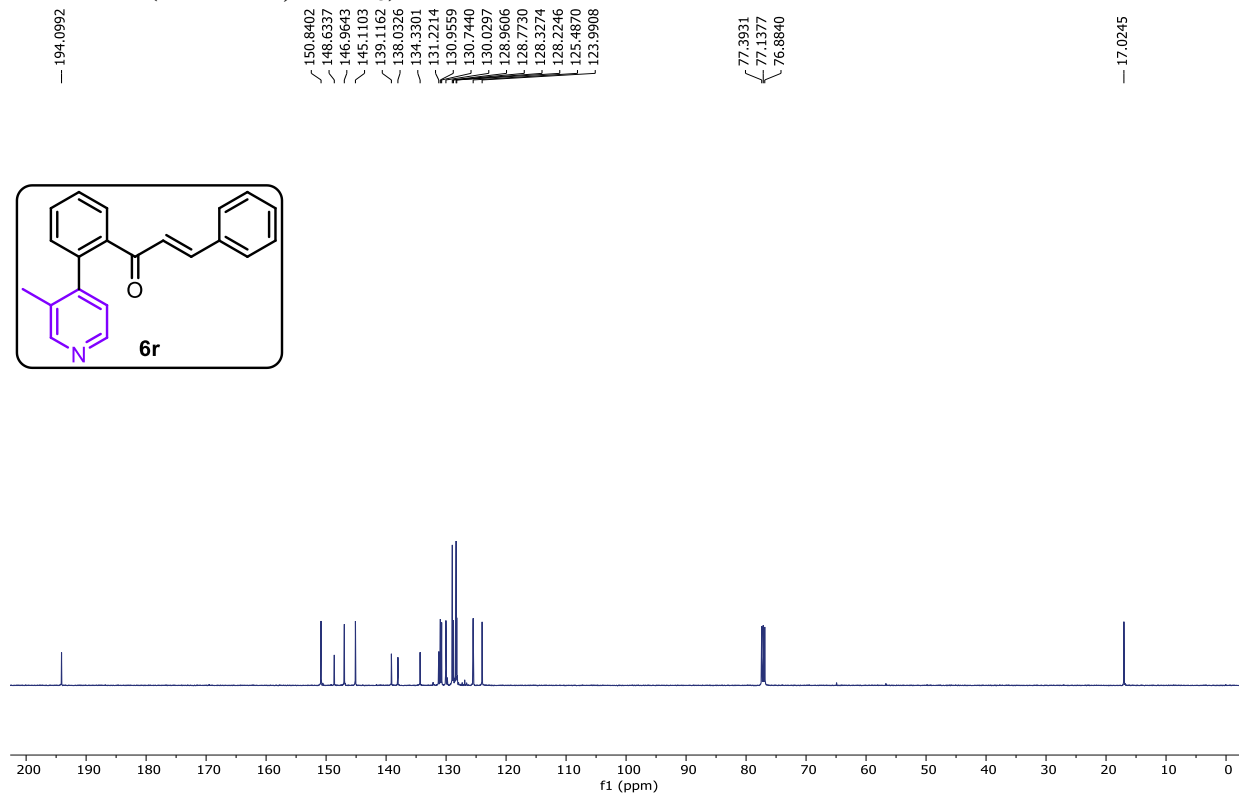
^{13}C NMR (125 MHz, CDCl_3)



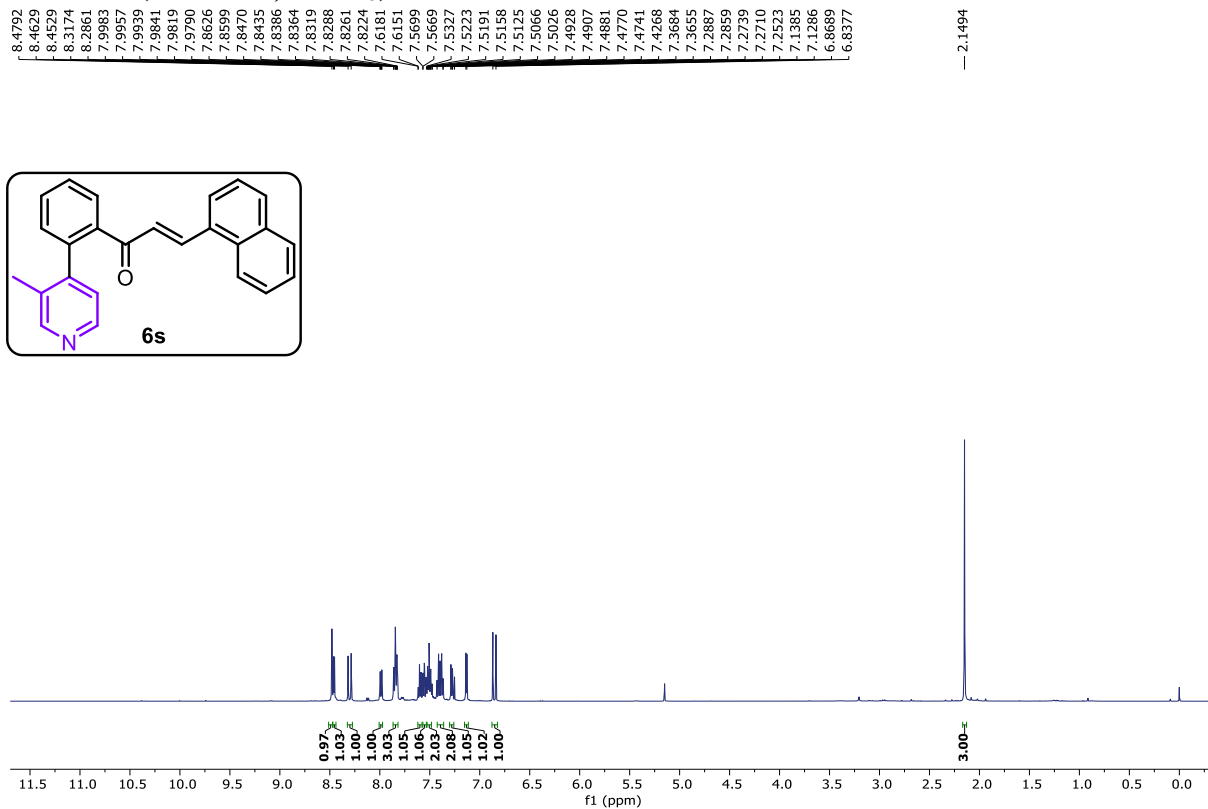
^1H NMR (500 MHz, CDCl_3)



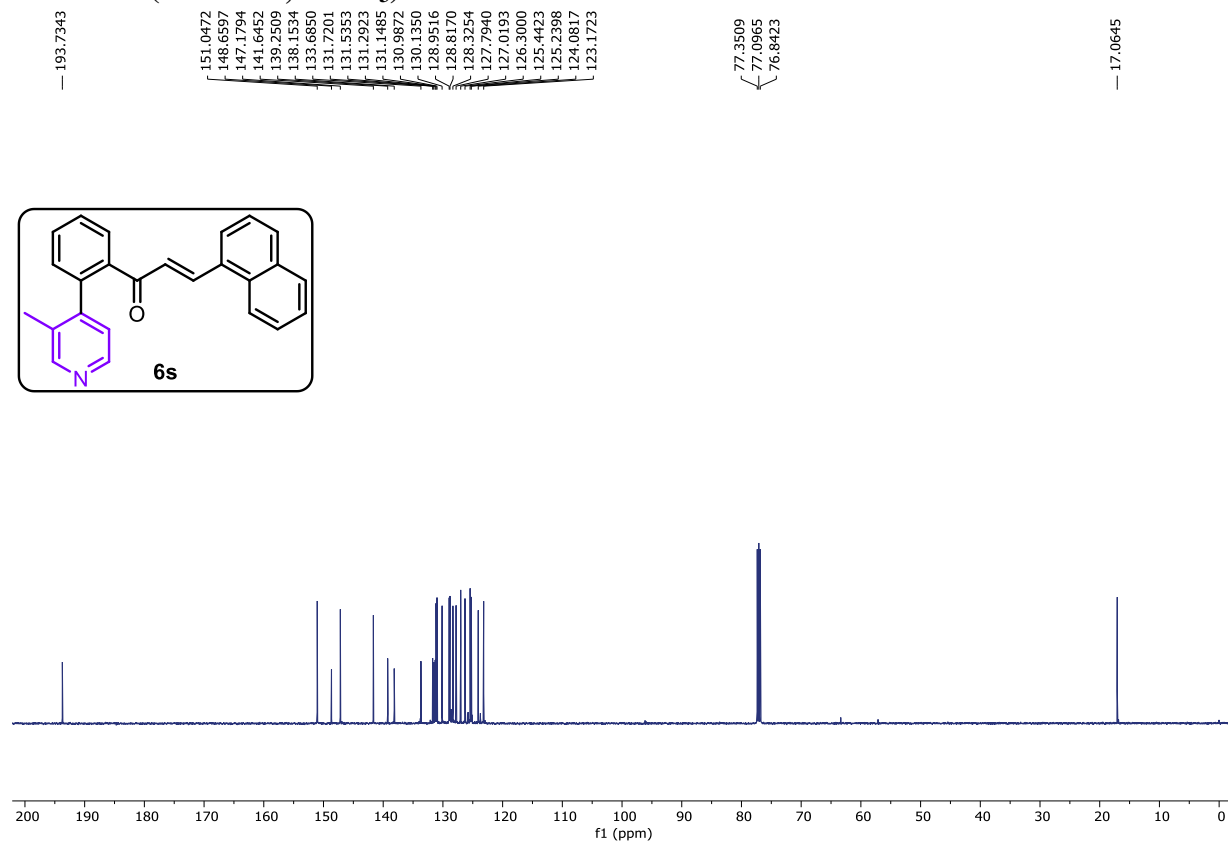
¹³C NMR (125 MHz, CDCl₃)



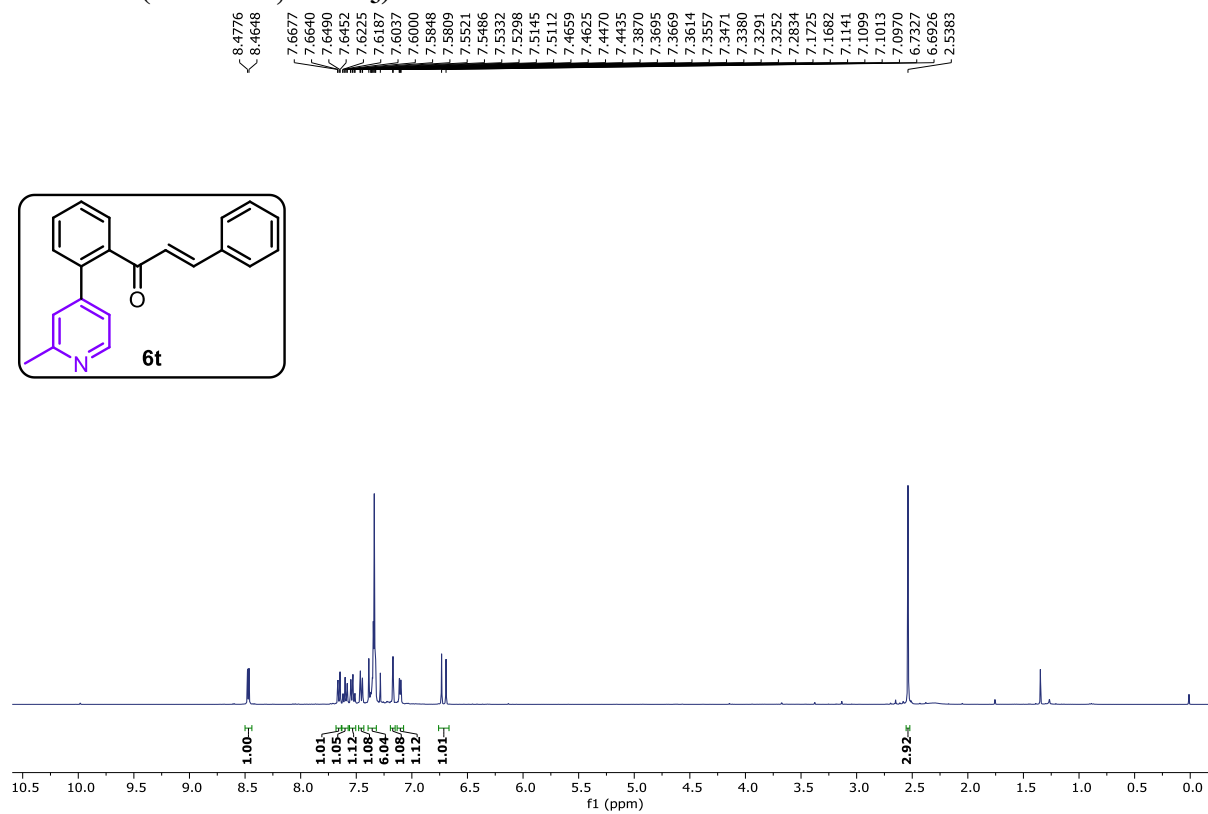
¹H NMR (500 MHz, CDCl₃)



^{13}C NMR (125 MHz, CDCl_3)



^1H NMR (400 MHz, CDCl_3)

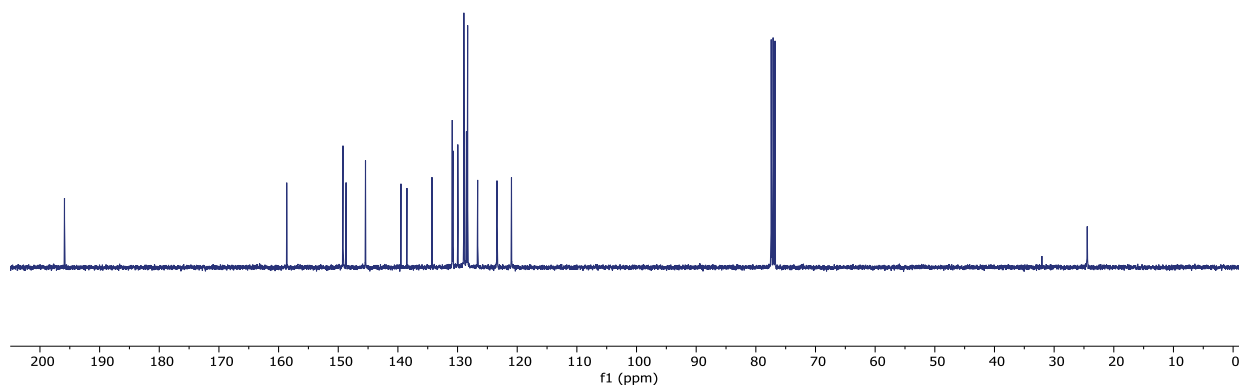
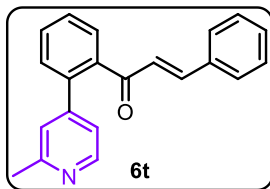


^{13}C NMR (100 MHz, CDCl_3)

195.8861
158.6352
149.2042
148.6863
145.4313
139.4885
138.4859
134.2861
130.8886
130.7675
129.9582
128.9417
128.8988
128.5119
128.3073
126.6303
123.3884
120.9632

77.4251
77.1064
76.7885

24.4504

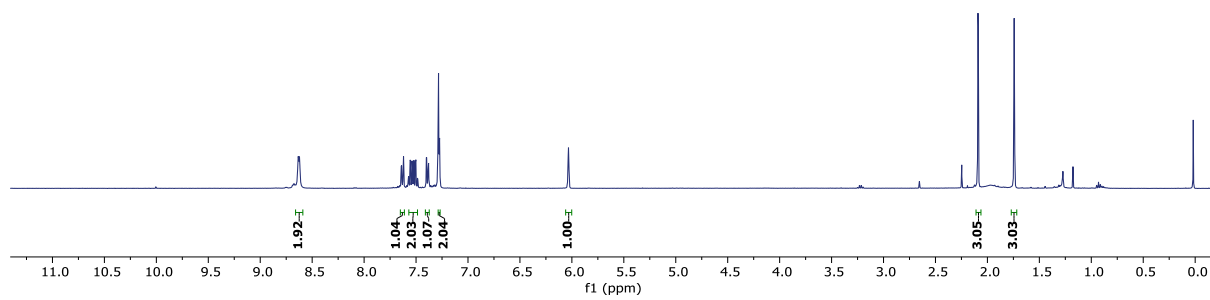
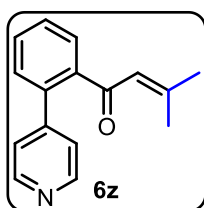


^1H NMR (400 MHz, CDCl_3)

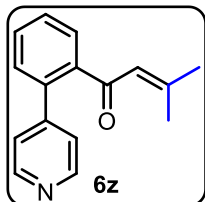
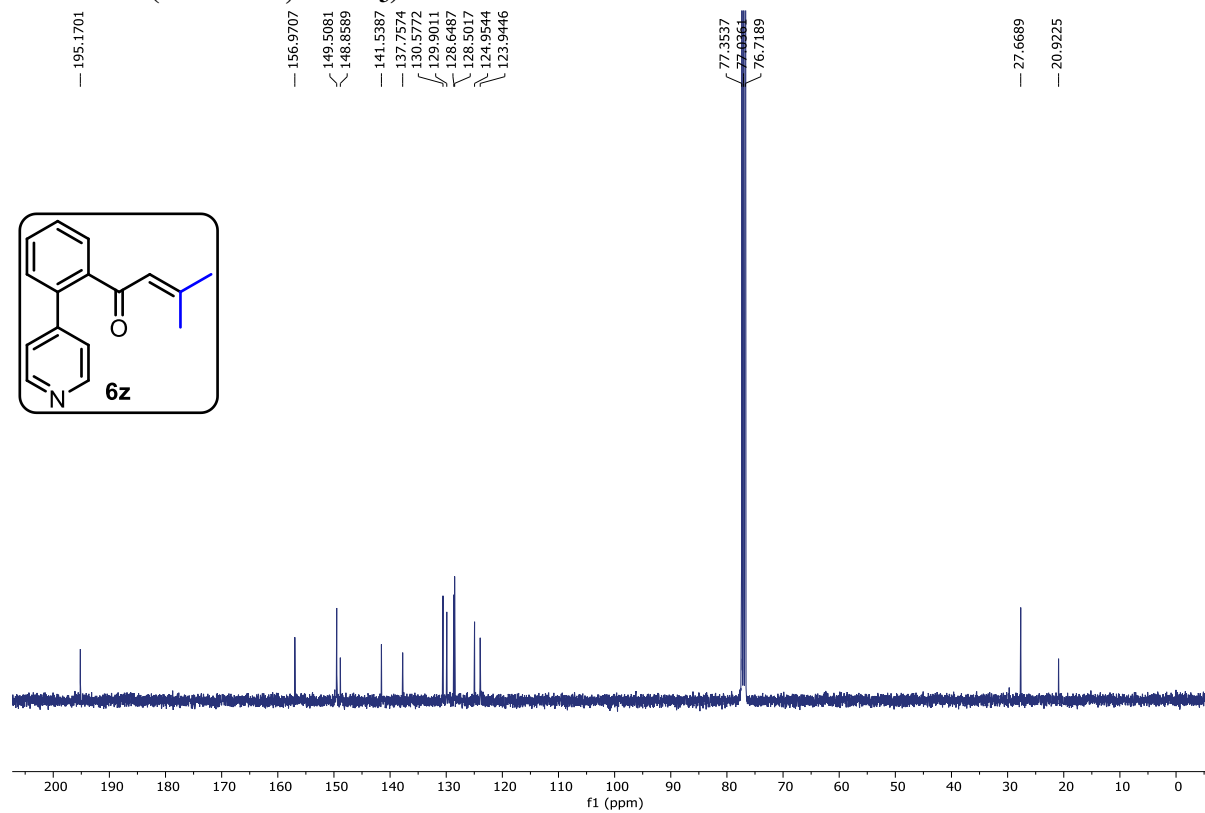
8.6344
8.6217
7.6431
7.6391
7.6245
7.6204
7.5759
7.5719
7.5574
7.5533
7.5391
7.5346
7.5241
7.5202
7.5094
7.5018
7.4868
7.4831
7.4011
7.3974
7.3829
7.3792
7.2885
7.2837
7.2772
7.2732
6.0326

2.0914

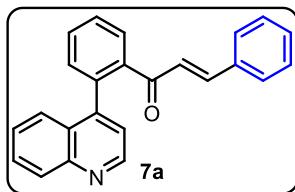
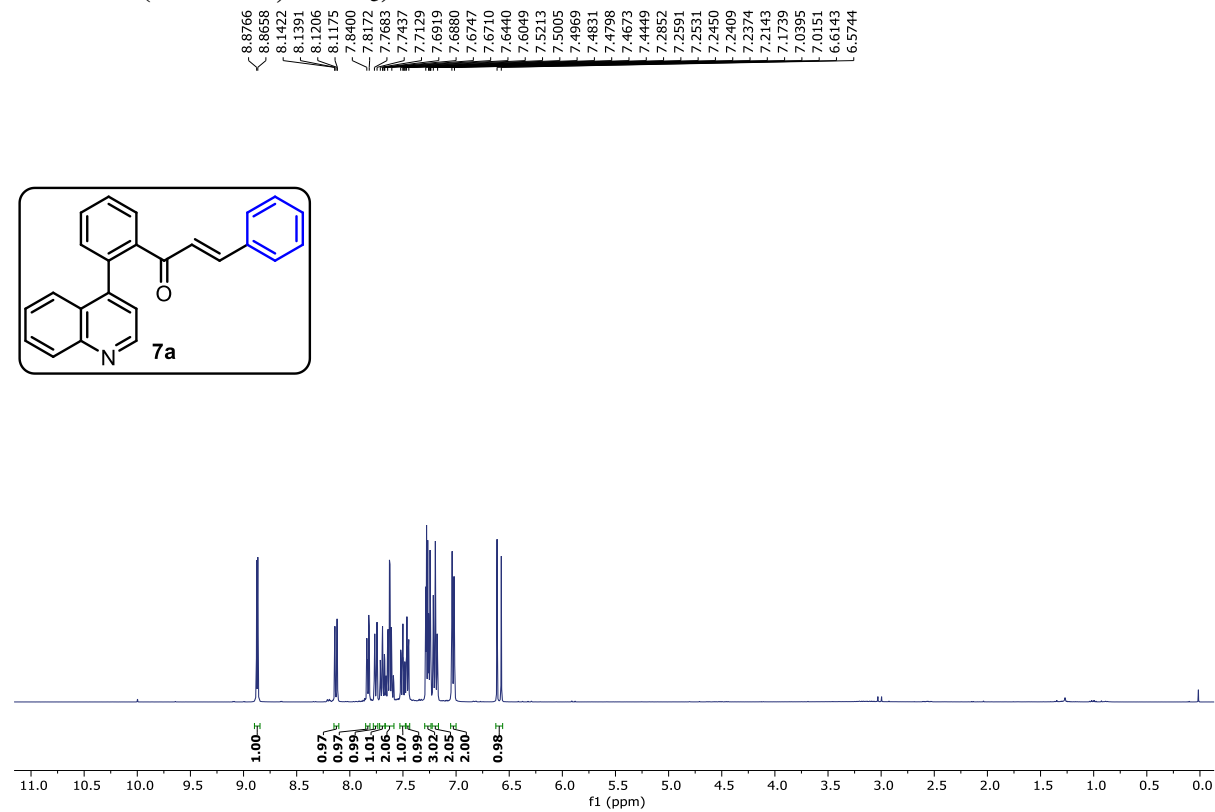
1.7430



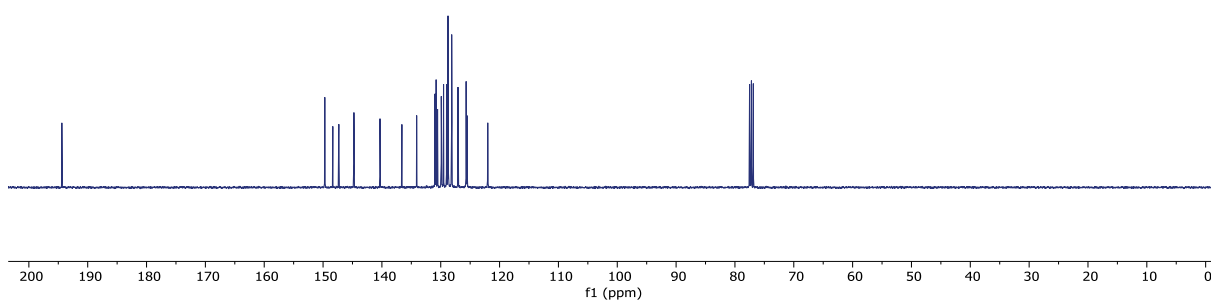
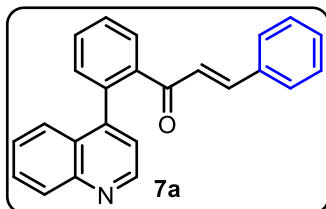
^{13}C NMR (100 MHz, CDCl_3)



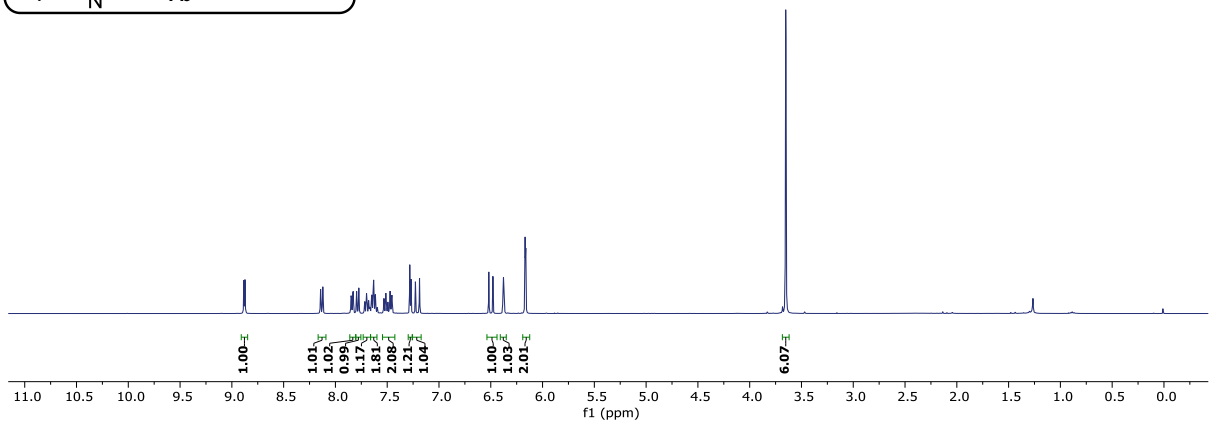
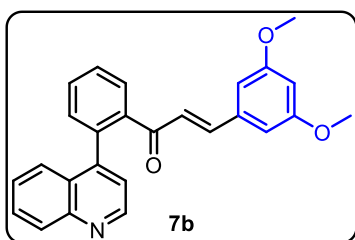
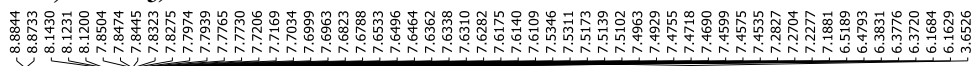
^1H NMR (400 MHz, CDCl_3)



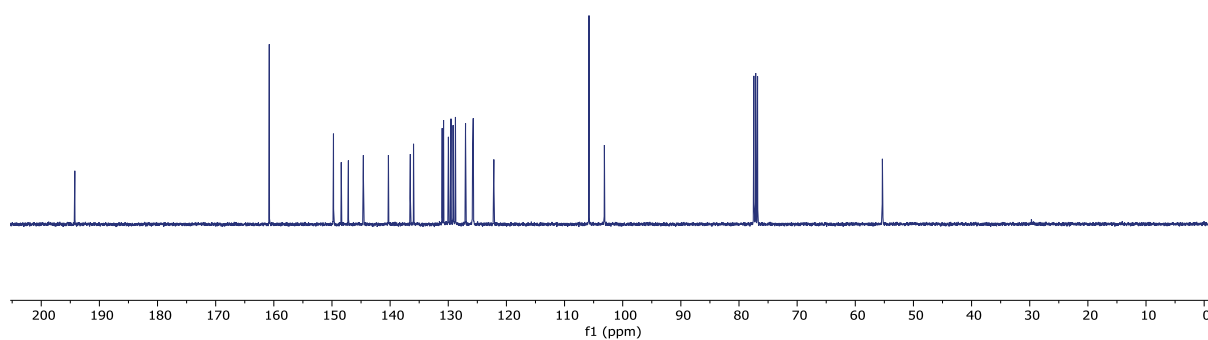
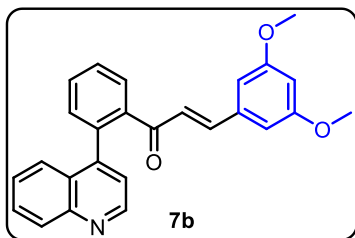
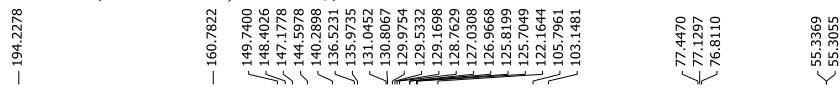
^{13}C NMR (100 MHz, CDCl_3)



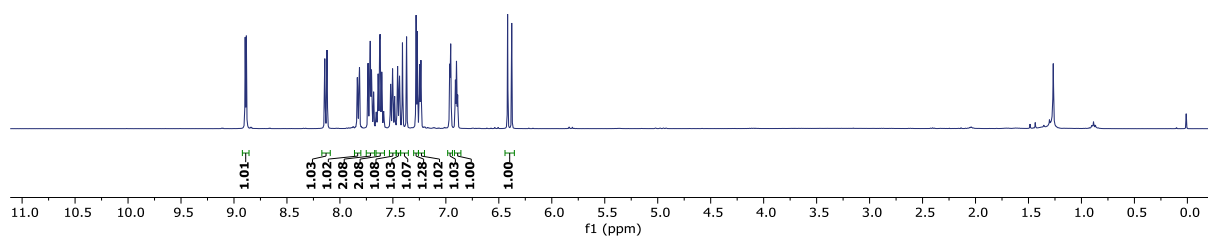
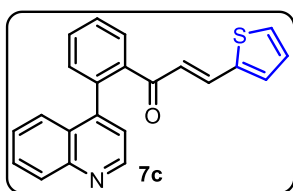
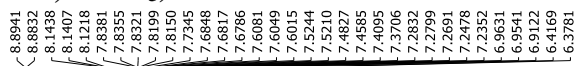
^1H NMR (400 MHz, CDCl_3)



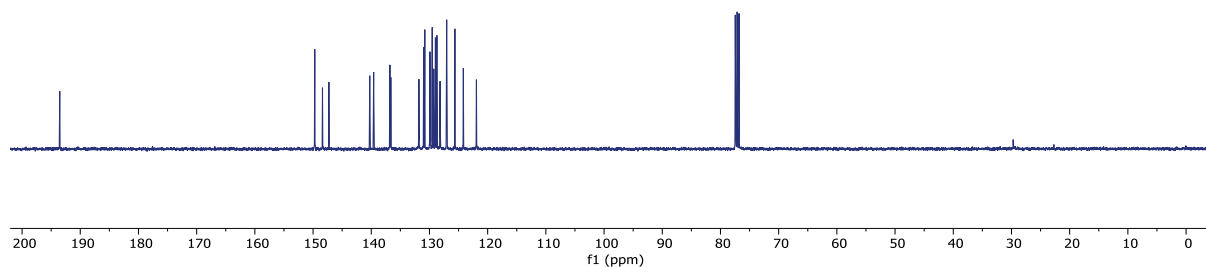
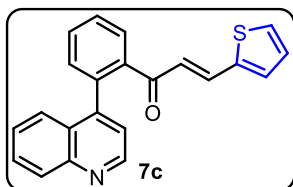
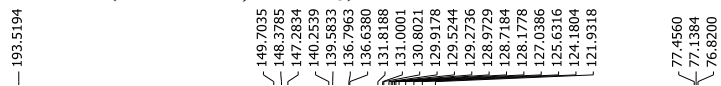
^{13}C NMR (100 MHz, CDCl_3)



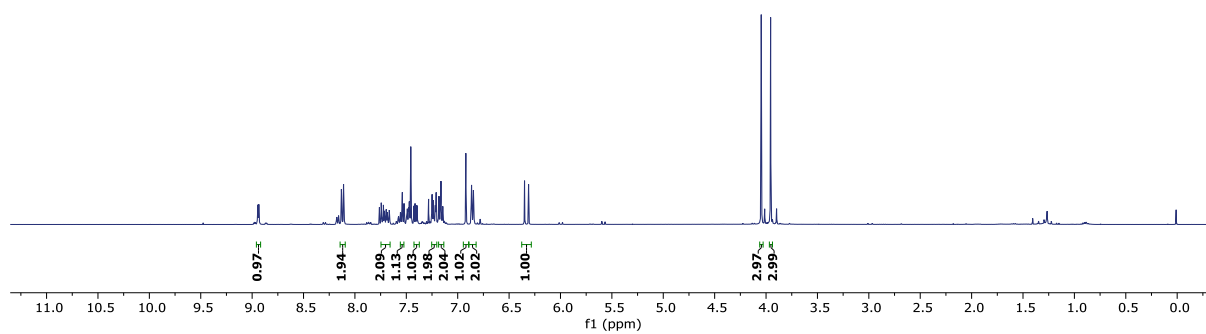
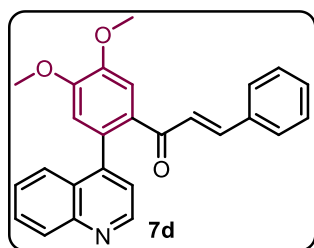
^1H NMR (400 MHz, CDCl_3)



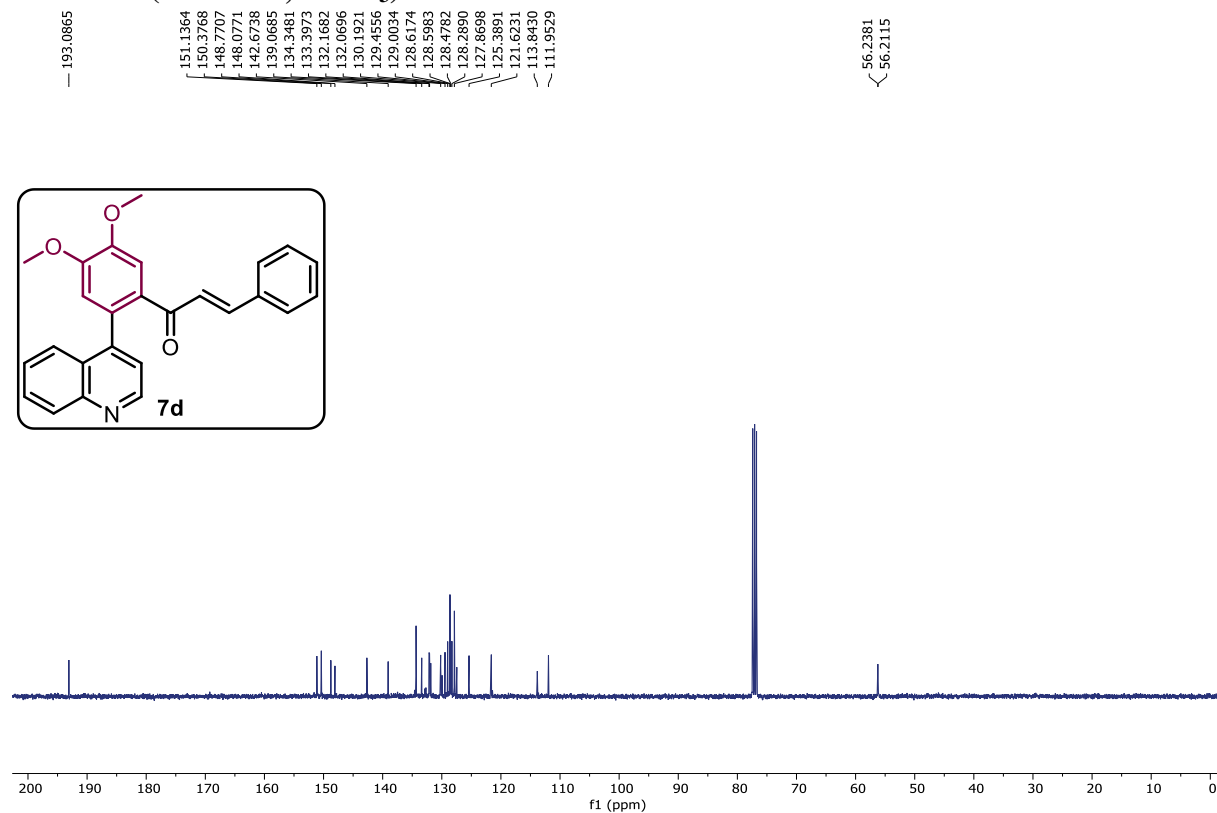
¹³C NMR (100 MHz, CDCl₃)



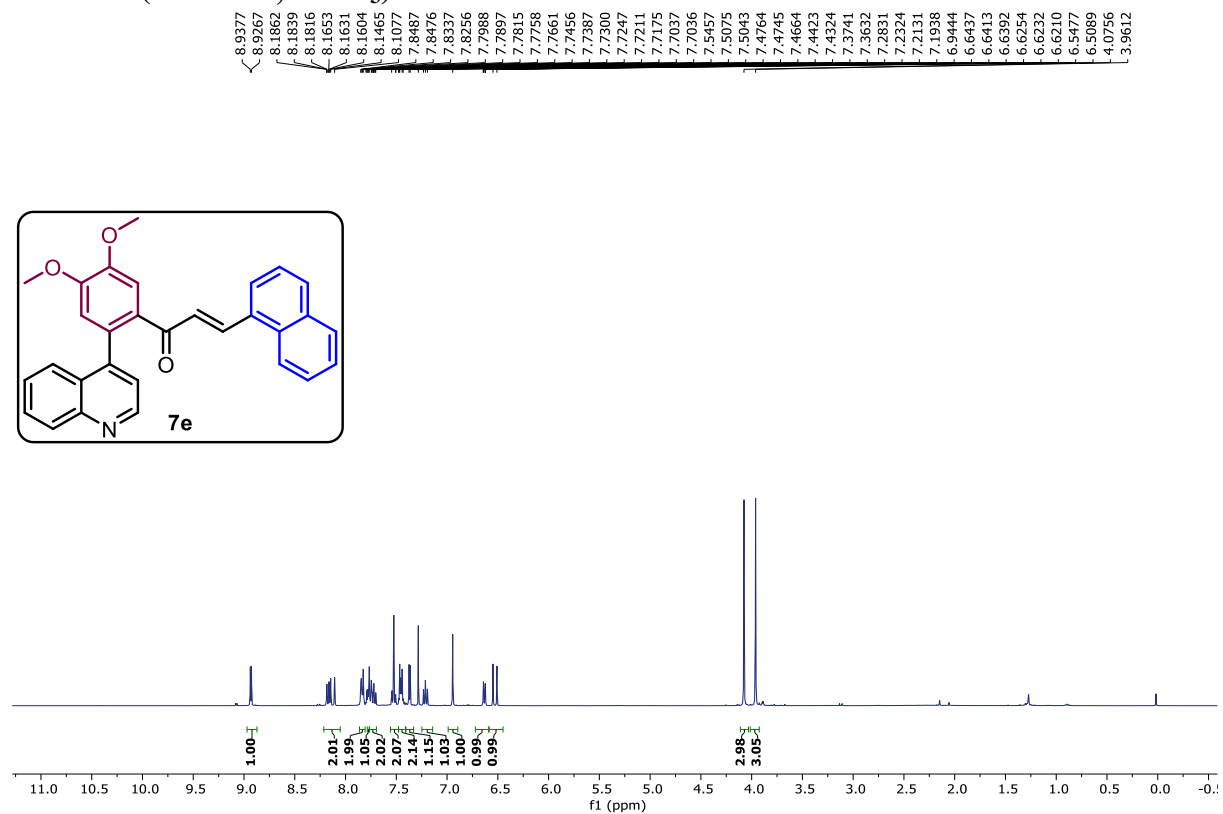
¹H NMR (400 MHz, CDCl₃)



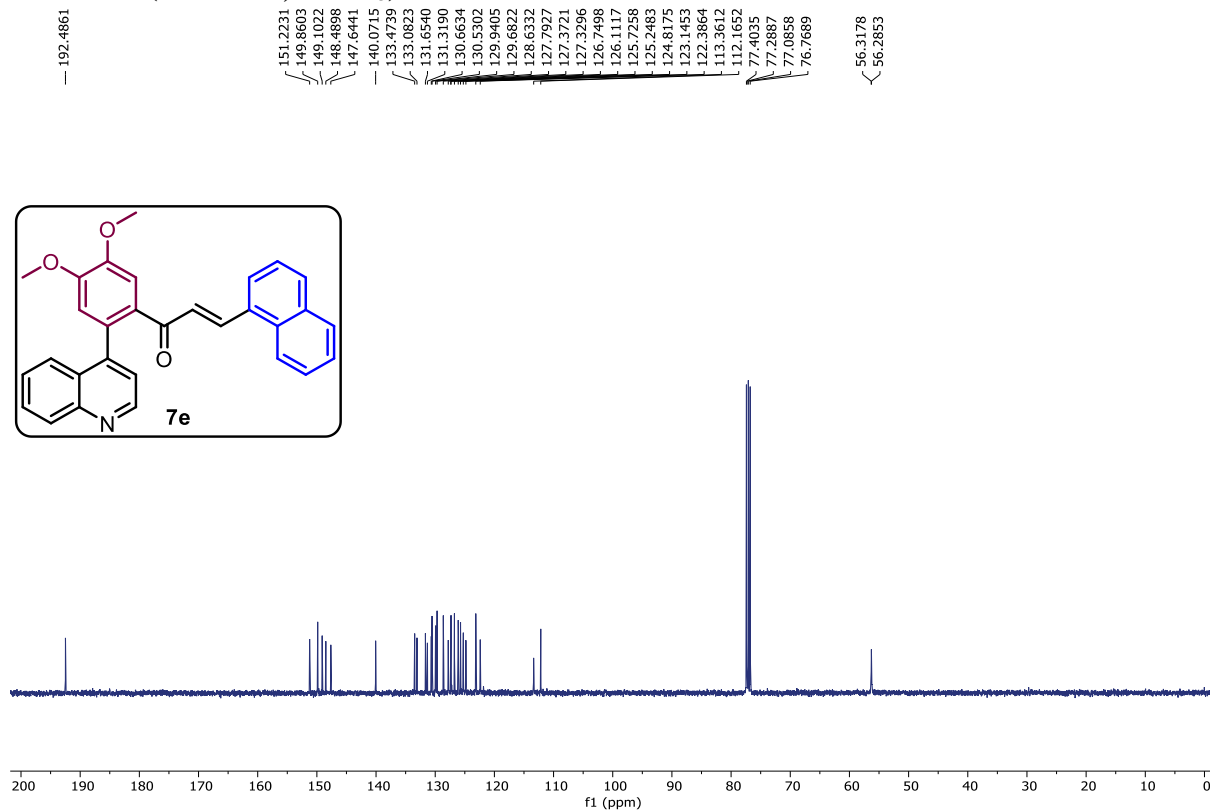
^{13}C NMR (100 MHz, CDCl_3)



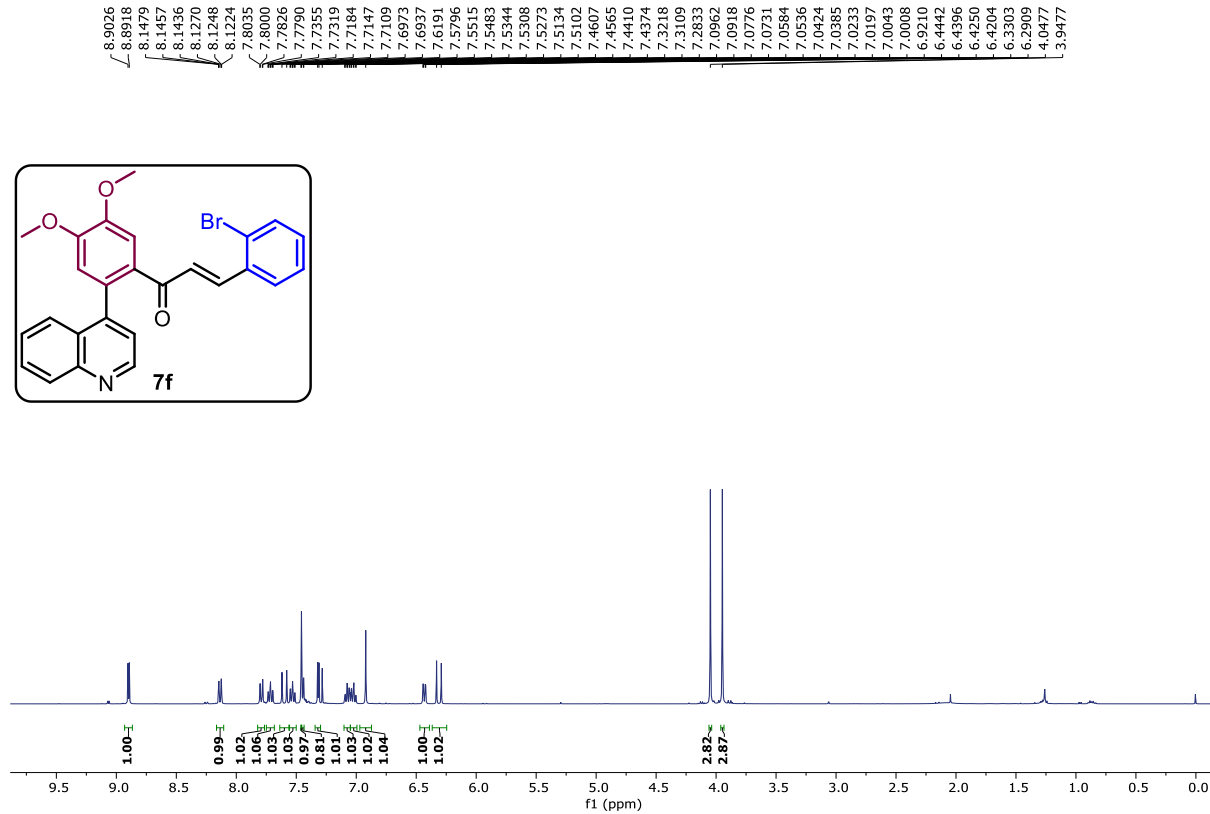
^1H NMR (400 MHz, CDCl_3)



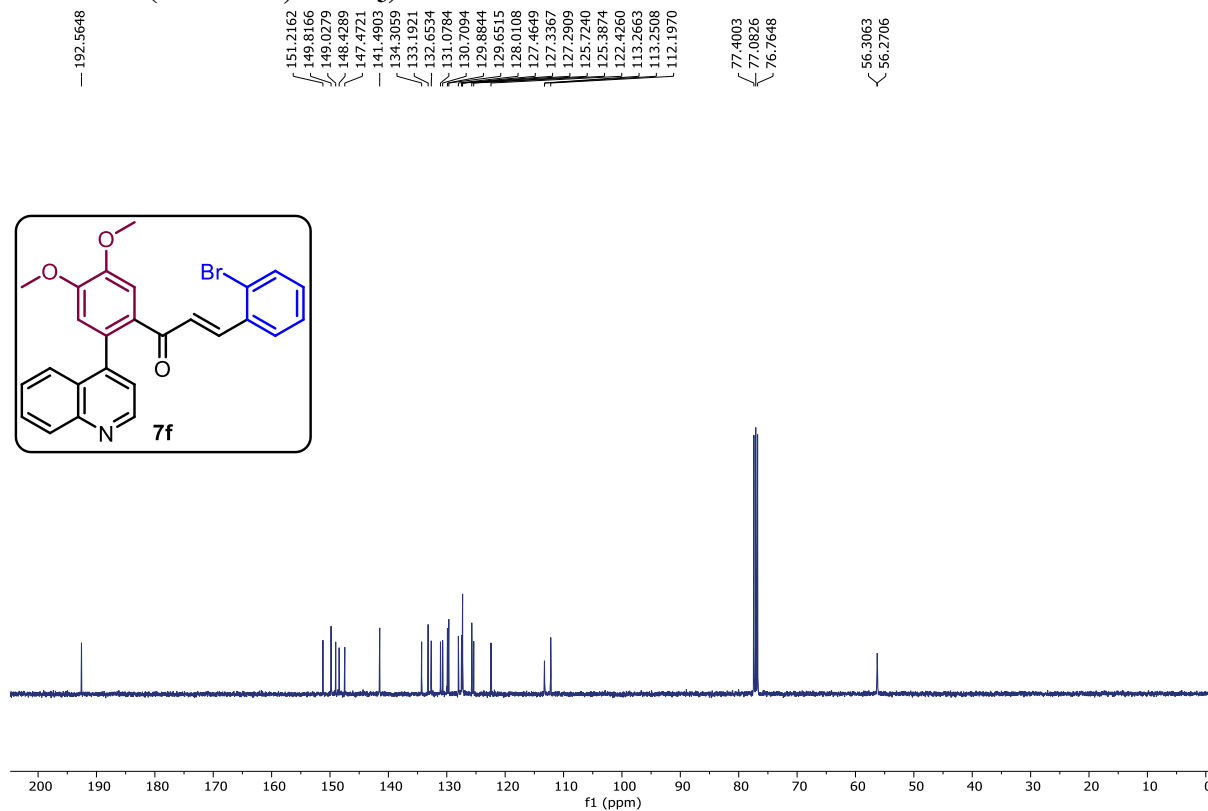
^{13}C NMR (100 MHz, CDCl_3)



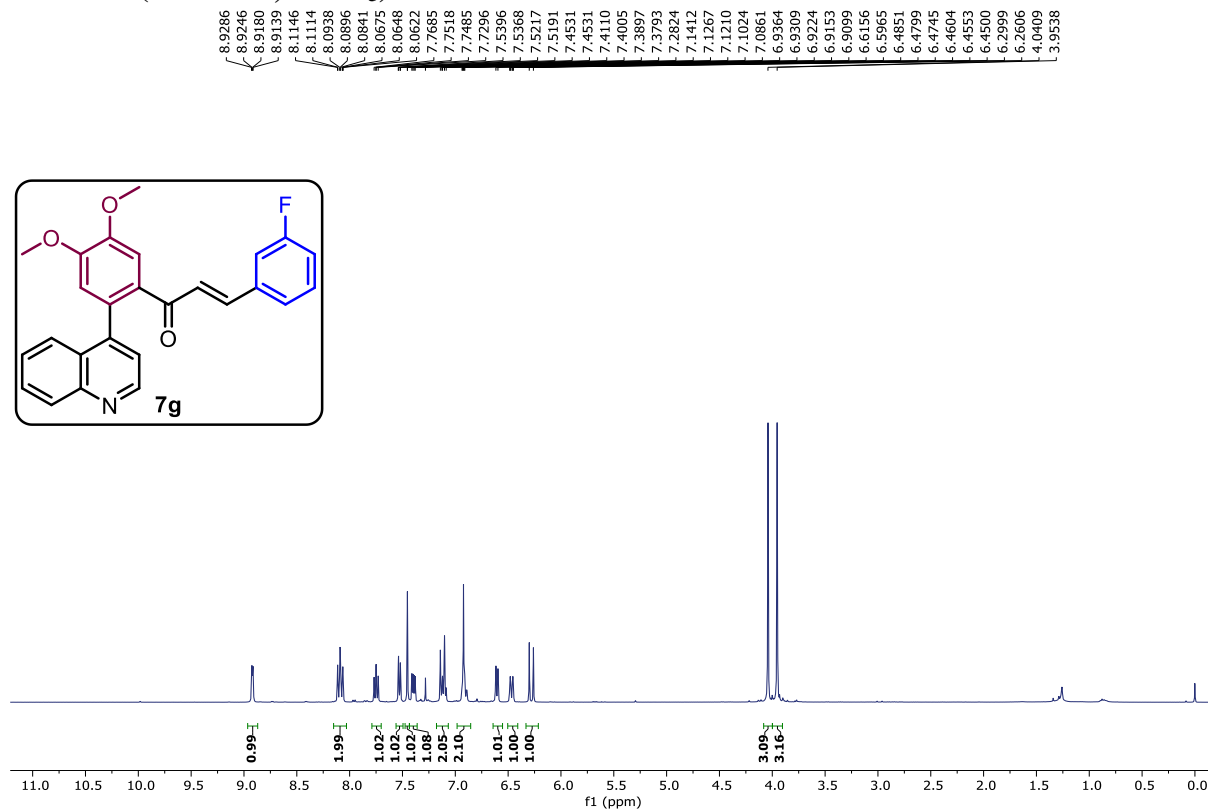
^1H NMR (400 MHz, CDCl_3)



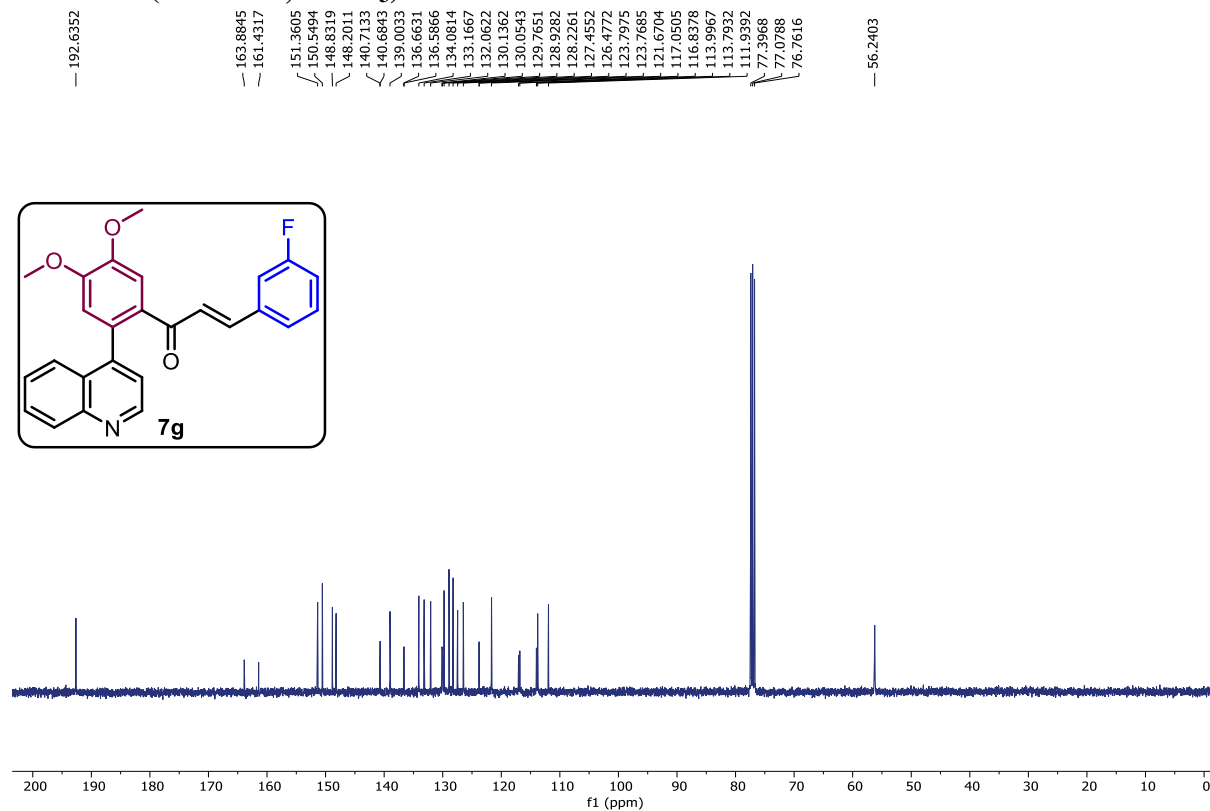
¹³C NMR (100 MHz, CDCl₃)



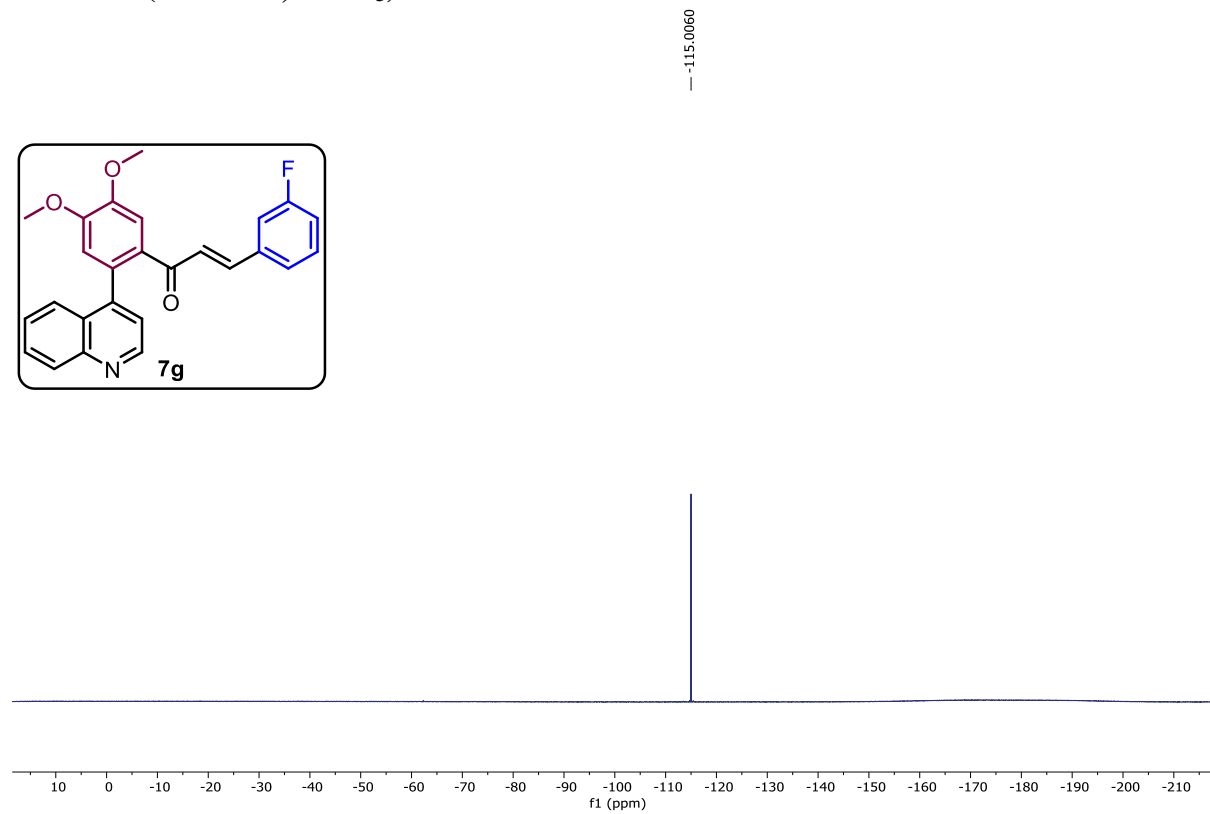
¹H NMR (400 MHz, CDCl₃)



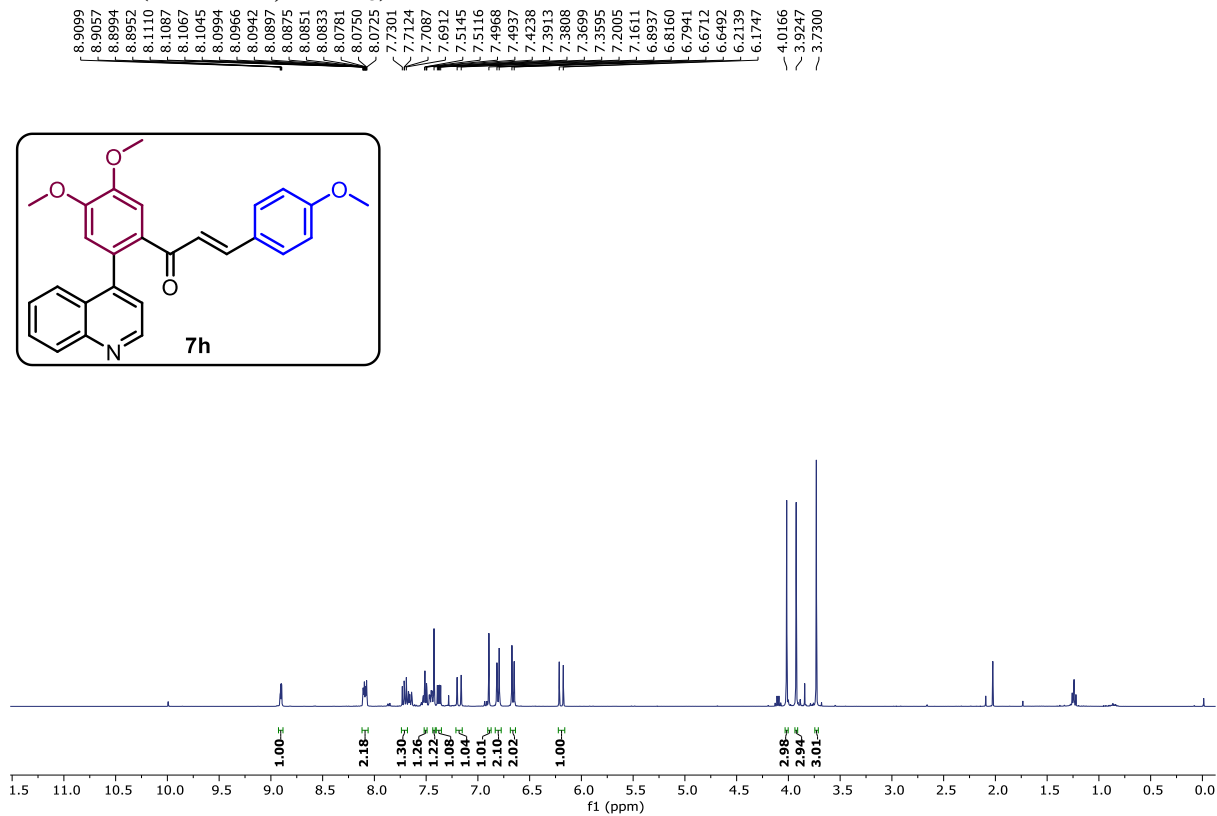
¹³C NMR (100 MHz, CDCl₃)



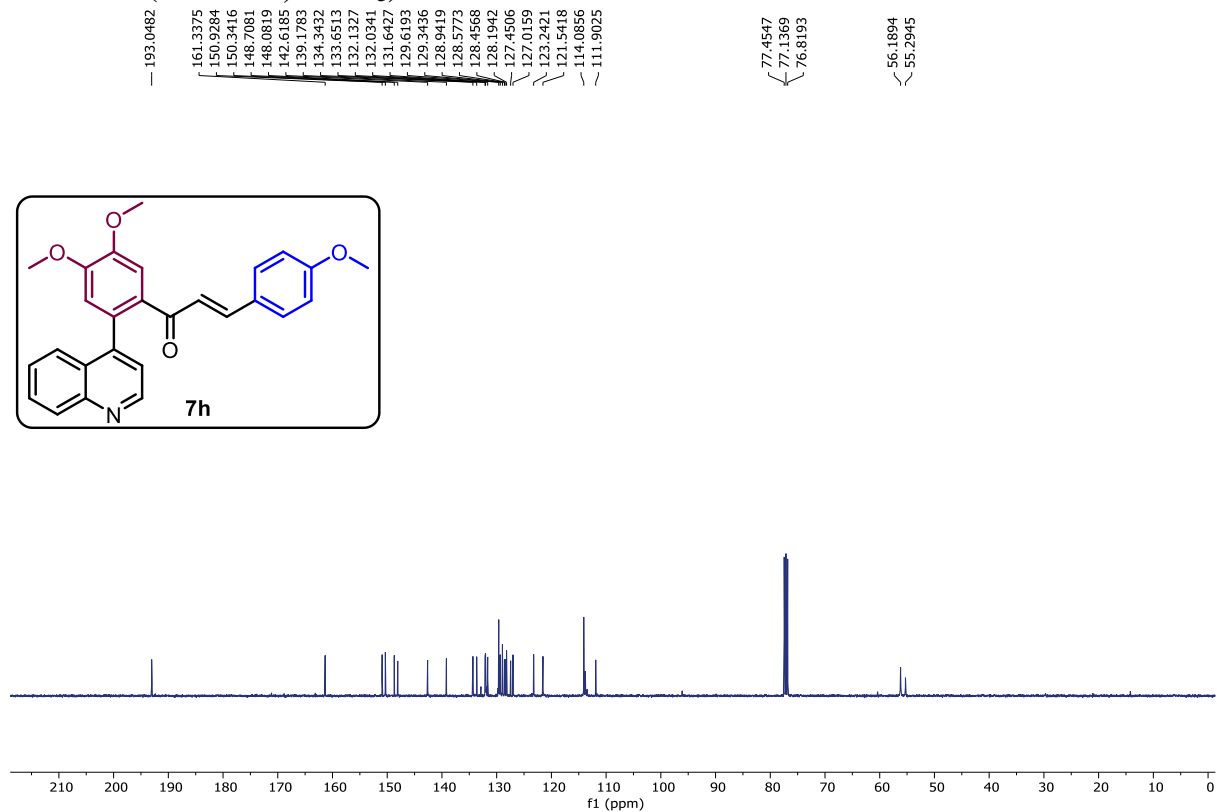
¹⁹F NMR (376 MHz, CDCl₃)



¹H NMR (400 MHz, CDCl₃)

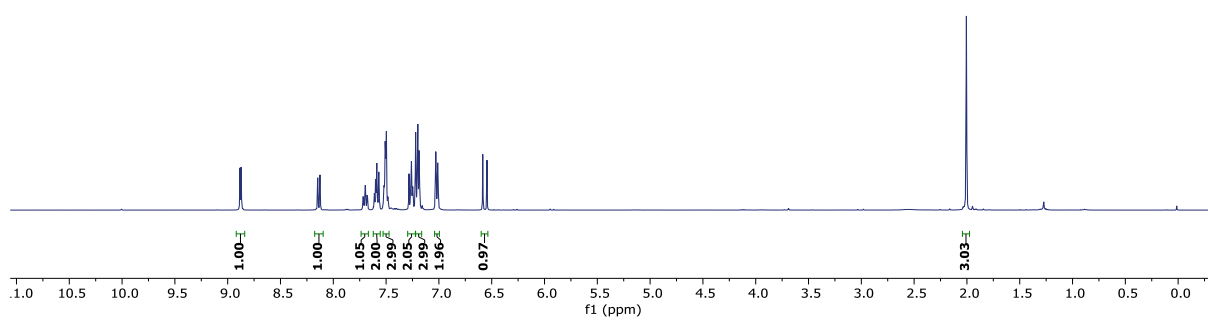
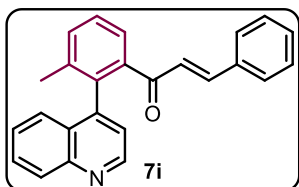


¹³C NMR (100 MHz, CDCl₃)



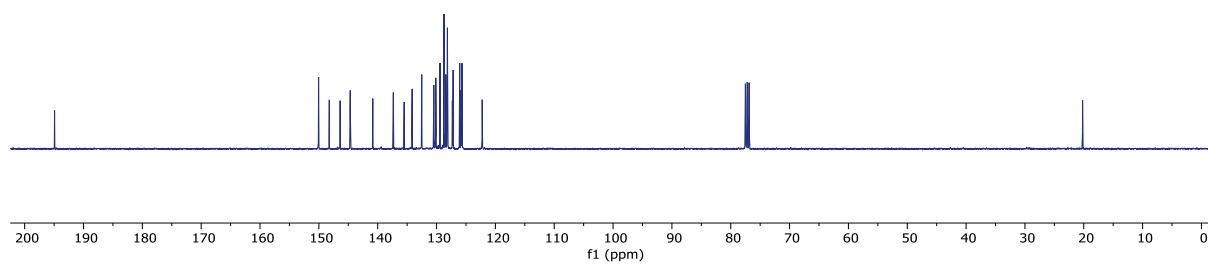
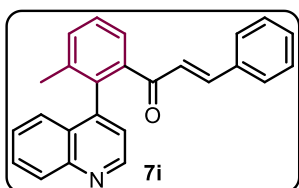
¹H NMR (400 MHz, CDCl₃)

8.8833
8.8724
8.1472
8.1262
7.7179
7.7141
7.7010
7.6970
7.6931
7.6797
7.6759
7.6106
7.5999
7.5882
7.5697
7.5660
7.5211
7.5180
7.5096
7.5062
7.5005
7.4969
7.4836
7.4800
7.2833
7.2690
7.2658
7.2601
7.2511
7.2475
7.2439
7.2201
7.2046
7.1997
7.1880
7.1817
7.0294
7.0115
7.0075
6.5846
6.5448
2.0052



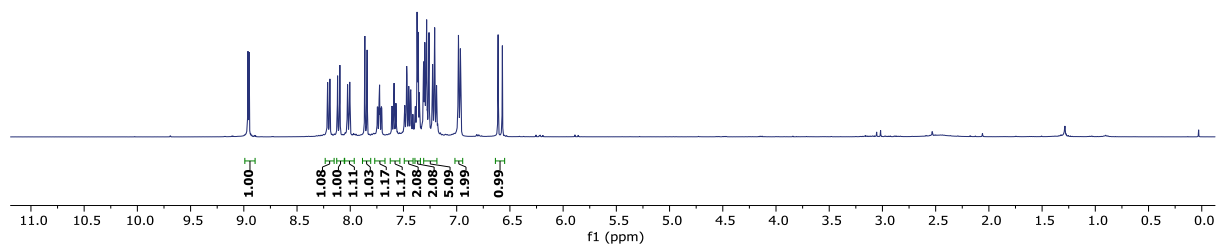
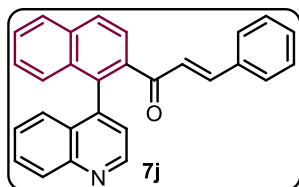
¹³C NMR (100 MHz, CDCl₃)

194.9166
150.0460
148.2275
146.3837
144.6662
140.8392
137.3462
135.5015
134.1409
132.5200
130.4698
130.1261
129.4126
128.7295
128.4429
128.1524
127.3117
127.1764
126.0580
125.8142
125.6678
122.2410
77.5073
77.1901
76.8716
20.2061



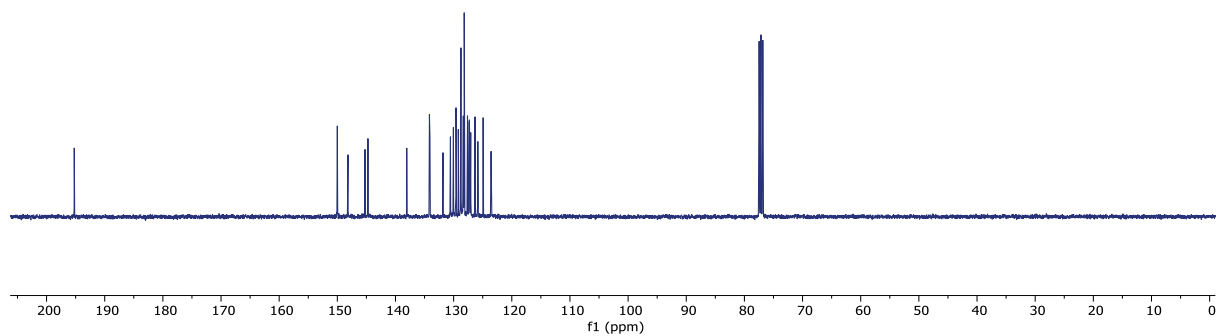
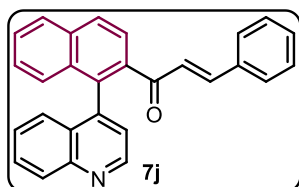
¹H NMR (400 MHz, CDCl₃)

8.9607
8.9500
8.2143
8.2121
8.2098
8.1927
8.1909
8.1888
8.1197
8.1179
8.0978
8.0274
8.0244
8.0066
8.0044
8.0021
8.0004
7.8628
7.8416
7.7456
7.7413
7.7292
7.7246
7.7199
7.7080
7.6084
7.6052
7.5917
7.5882
7.5846
7.5711
7.5678
7.4872
7.4706
7.4663
7.4504
7.4474
7.4342
7.4306
7.3902
7.3869
7.3725
7.3688
7.3655
7.3615
7.3520
7.3487
7.3117
7.3091
7.2997
7.2906
7.2876
7.2822
7.2660
7.2626
7.2596
7.2265
7.2223
7.2108
7.2068
7.1892
7.1859
6.9854
6.9826
6.9787
6.9702
6.9652
6.9611
6.6107
6.5707

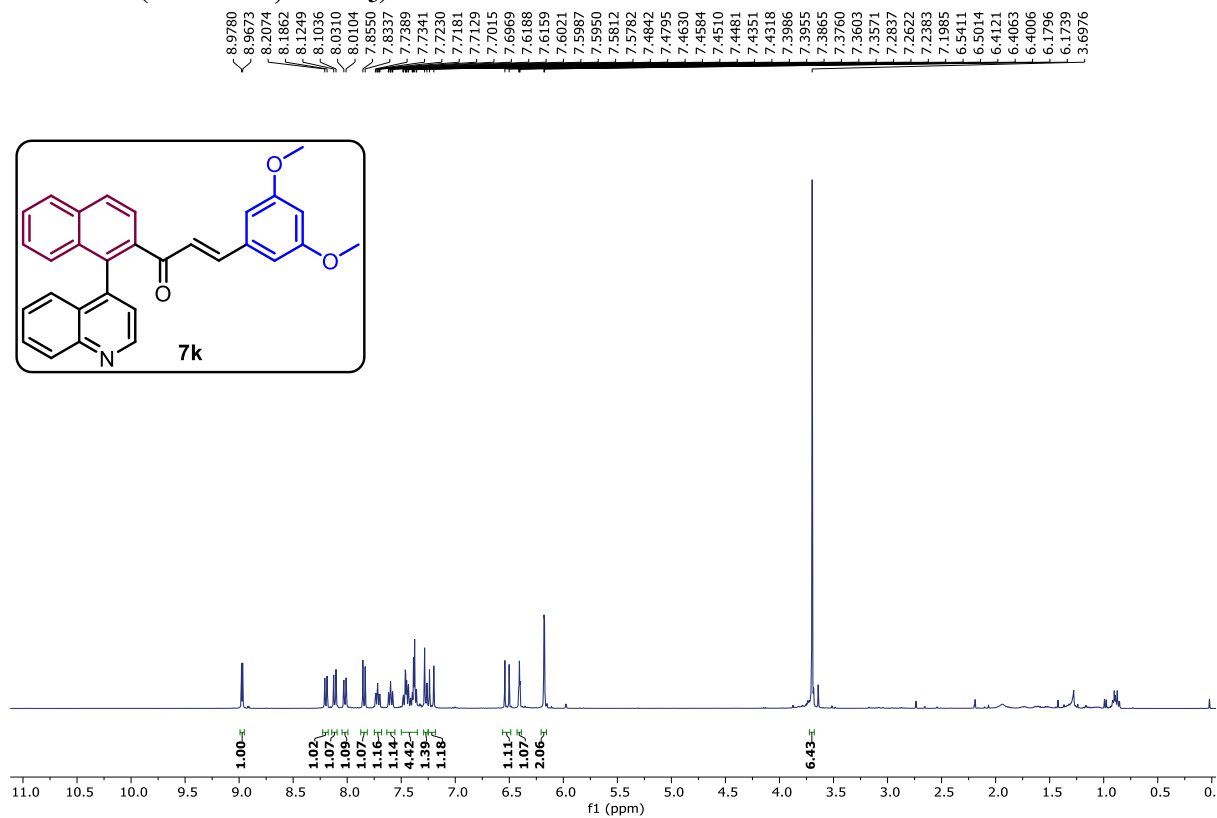


¹³C NMR (100 MHz, CDCl₃)

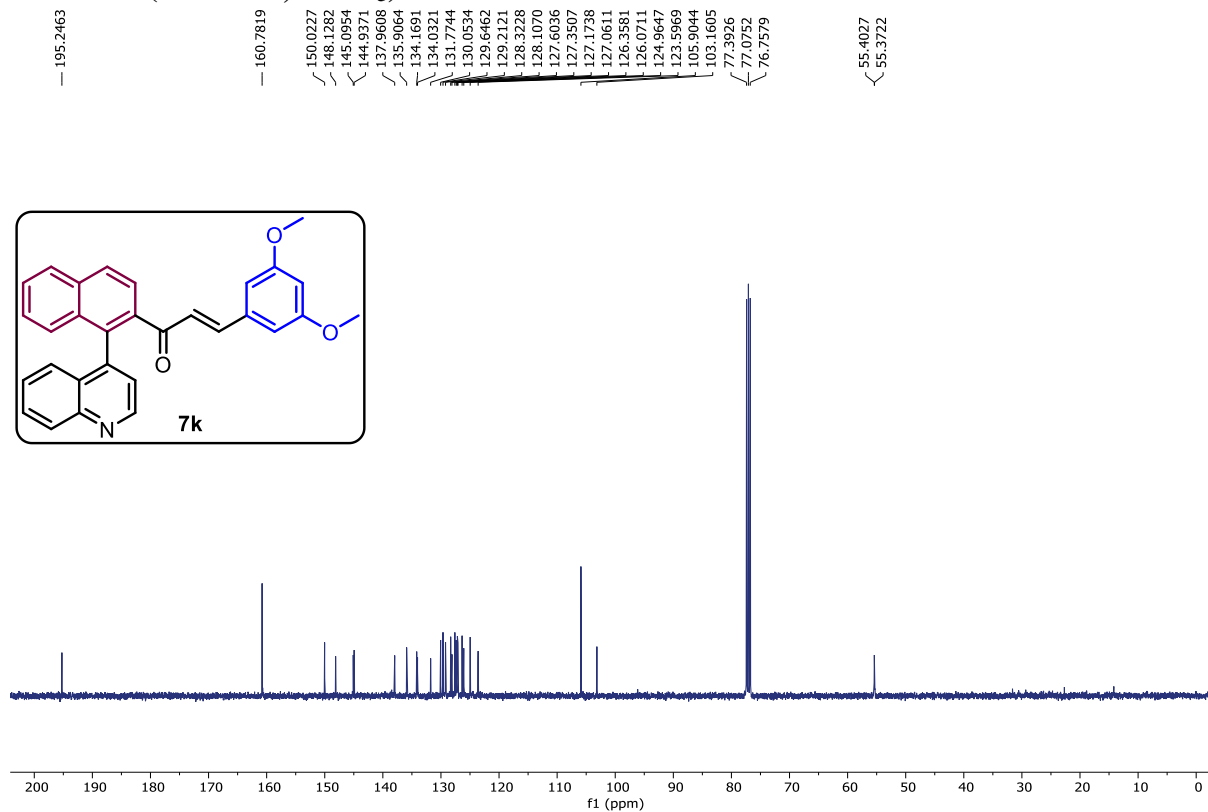
195.2374
150.0015
148.1649
145.2366
144.7345
138.0629
134.1638
134.0746
131.8292
130.5523
130.0419
129.5916
129.1960
128.7460
128.3365
128.1795
127.6141
127.3709
127.2846
127.0627
126.3279
125.8392
124.9284
123.5617
77.4713
77.1527
76.8345



¹H NMR (400 MHz, CDCl₃)

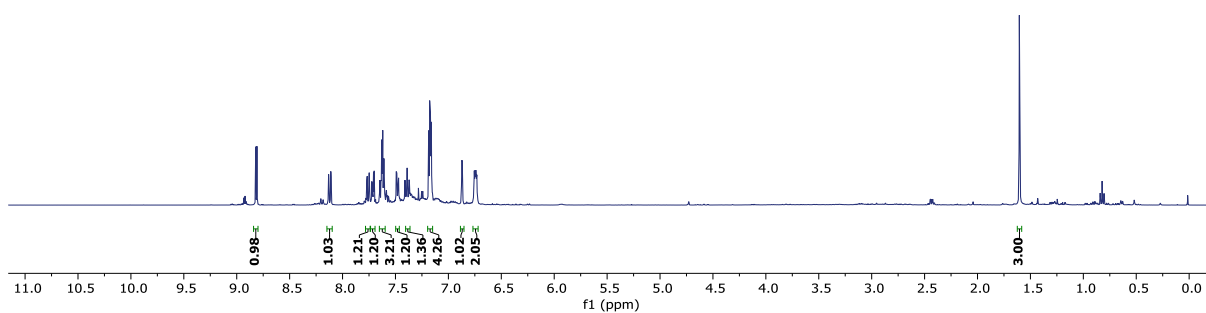
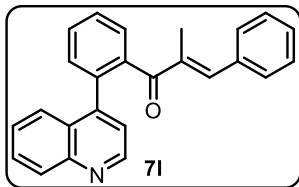


¹³C NMR (100 MHz, CDCl₃)



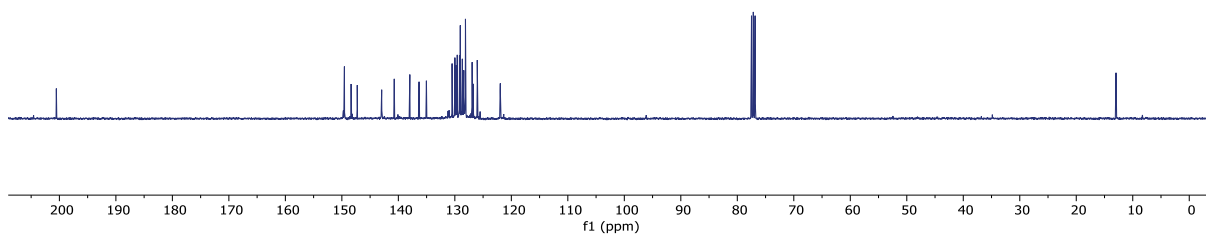
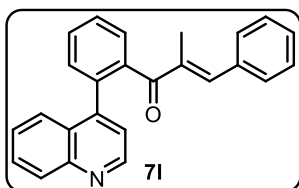
¹H NMR (400 MHz, CDCl₃)

8.8203
8.8093
8.1332
8.1302
8.1120
8.1089
7.7701
7.7665
7.7491
7.7455
7.7238
7.7074
7.7013
7.6488
7.6359
7.6313
7.6272
7.6236
7.6203
7.6133
7.6093
7.6050
7.4929
7.4871
7.4706
7.4104
7.4071
7.3933
7.3896
7.3860
7.3722
7.3687
7.1861
7.1816
7.1772
7.1747
7.1694
7.1659
7.1612
7.1565
6.8726
6.8687
6.7553
6.7492
6.7448
6.7406
6.7368
6.7309
1.6050
1.6014
0.8223

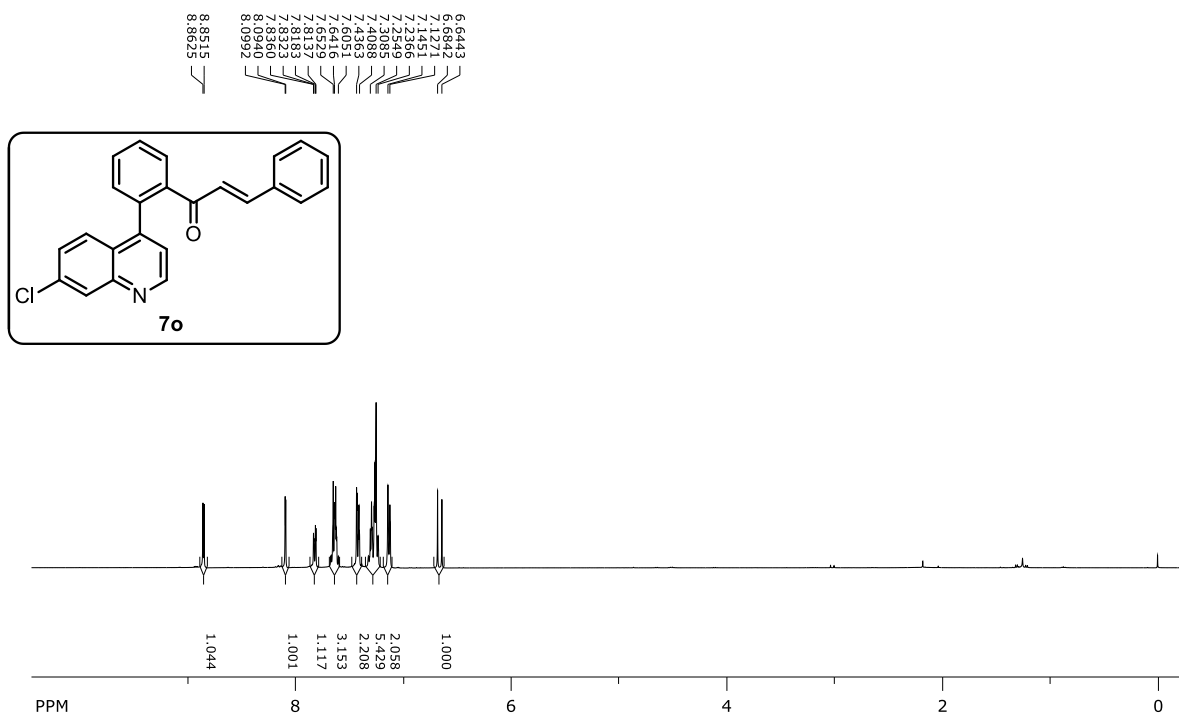


¹³C NMR (100 MHz, CDCl₃)

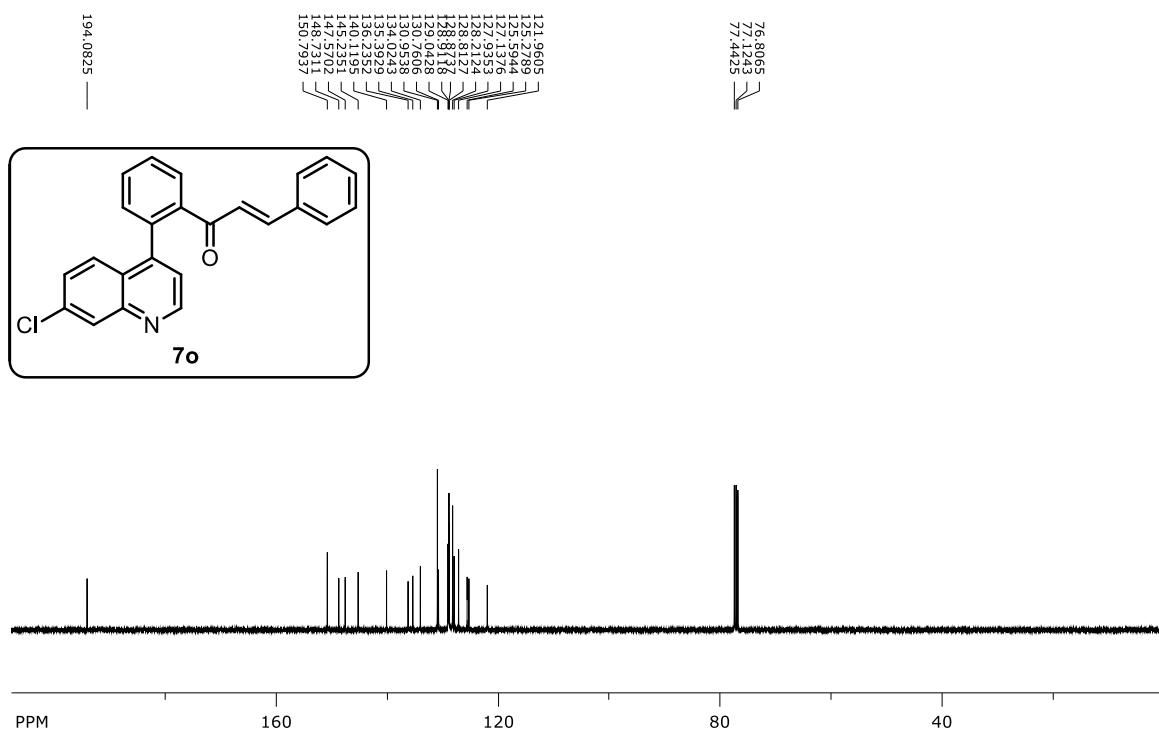
200.5409
149.5468
148.3583
147.2717
142.9535
140.7244
137.9685
136.3308
135.0329
130.4841
130.4841
129.9913
129.7338
129.5722
129.1786
129.0360
128.7200
128.6754
128.4234
128.3767
128.1195
126.9366
126.7138
126.0324
121.9530
77.4841
77.1663
76.8488
12.9622



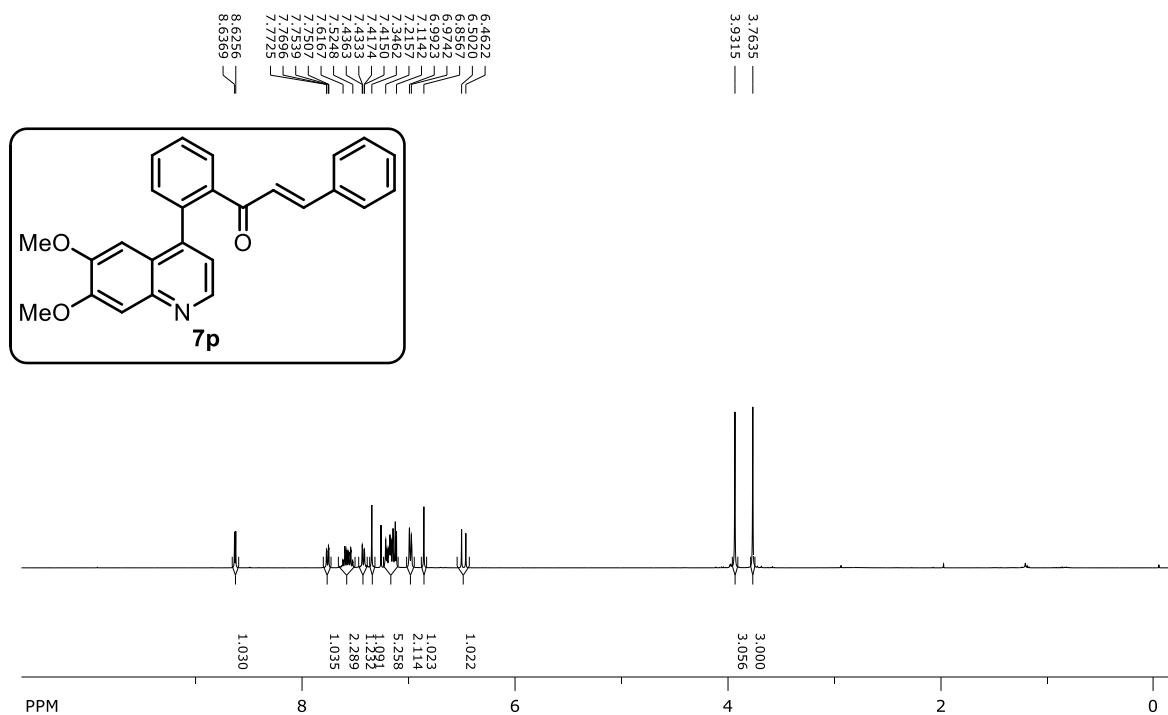
¹H NMR (400 MHz, CDCl₃)



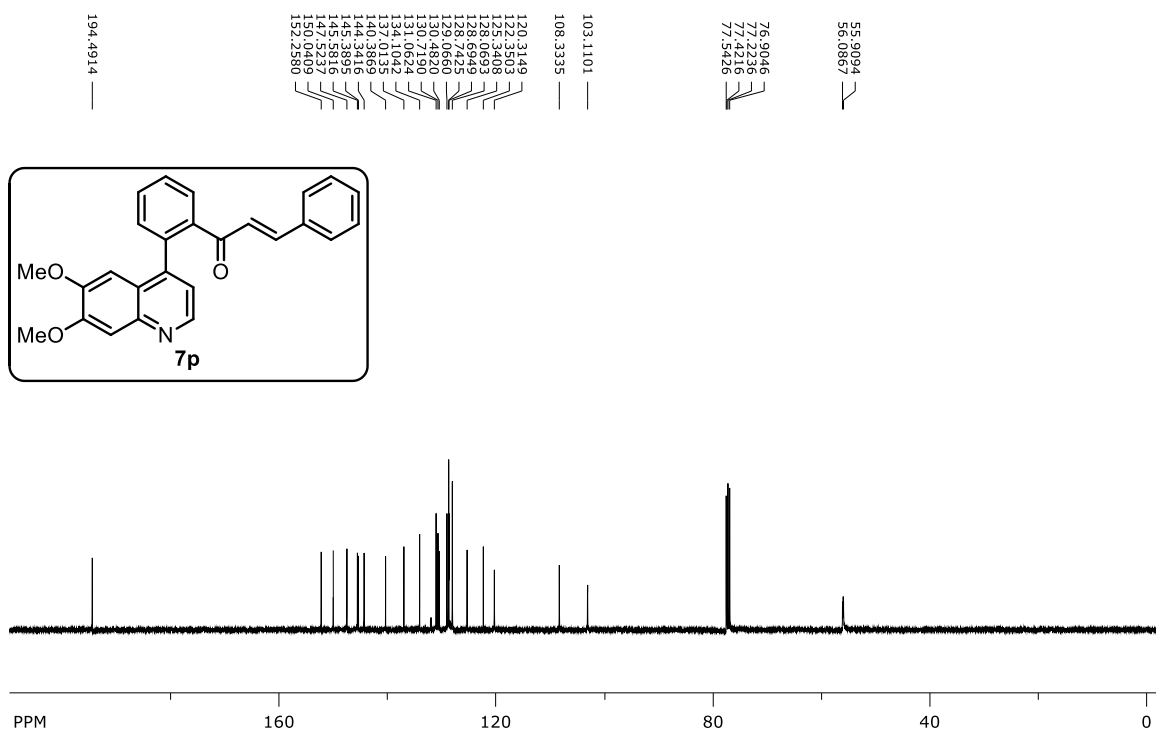
¹³C NMR (100 MHz, CDCl₃)



¹H NMR (400 MHz, CDCl₃)

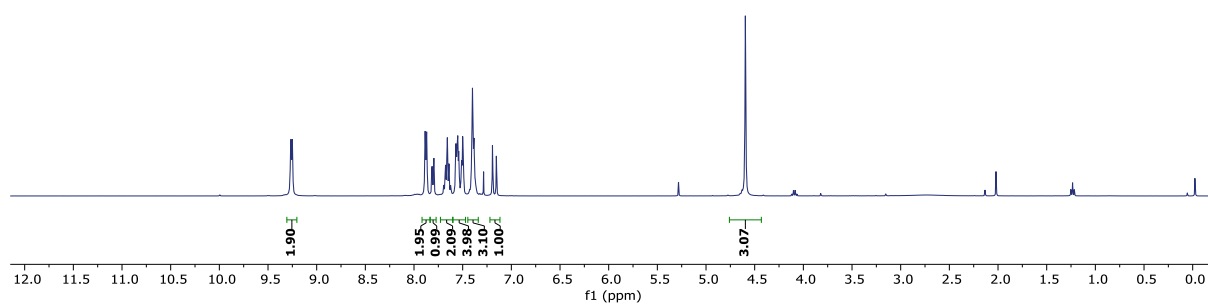
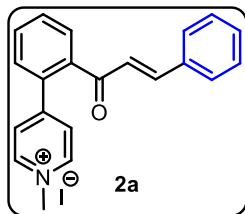


¹³C NMR (100 MHz, CDCl₃)



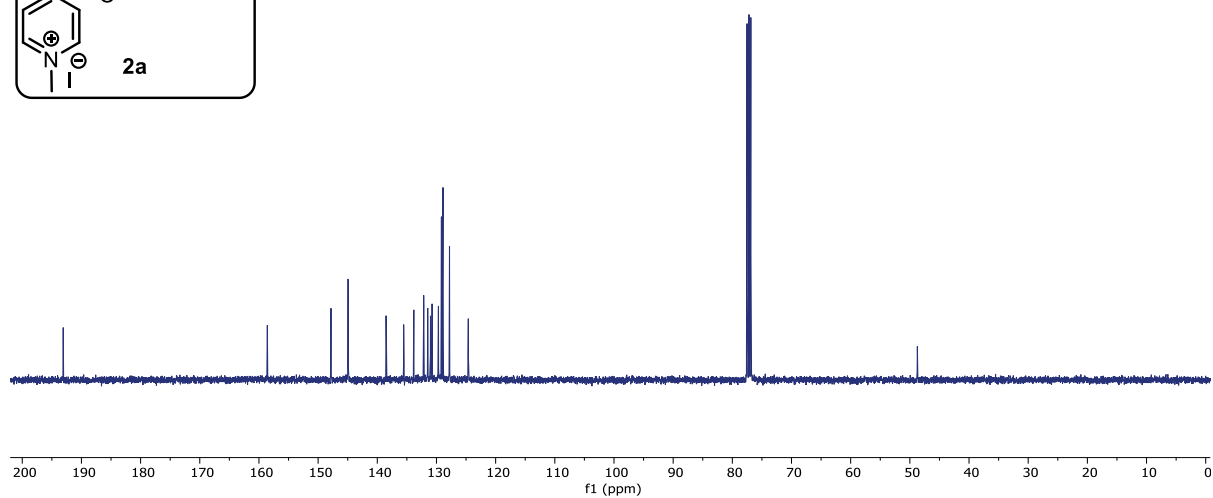
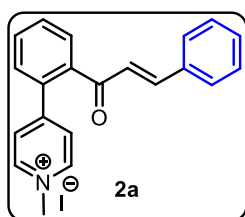
¹H NMR (400 MHz, CDCl₃)

9.2662
9.2509
7.8852
7.8700
7.8179
7.8132
7.8002
7.7957
7.6796
7.6754
7.6623
7.6572
7.6438
7.6399
7.5747
7.5700
7.5568
7.5505
7.5407
7.5354
7.5155
7.5109
7.5005
7.4939
7.4112
7.4028
7.3987
7.3842
7.2853
7.1935
7.1535
4.5957

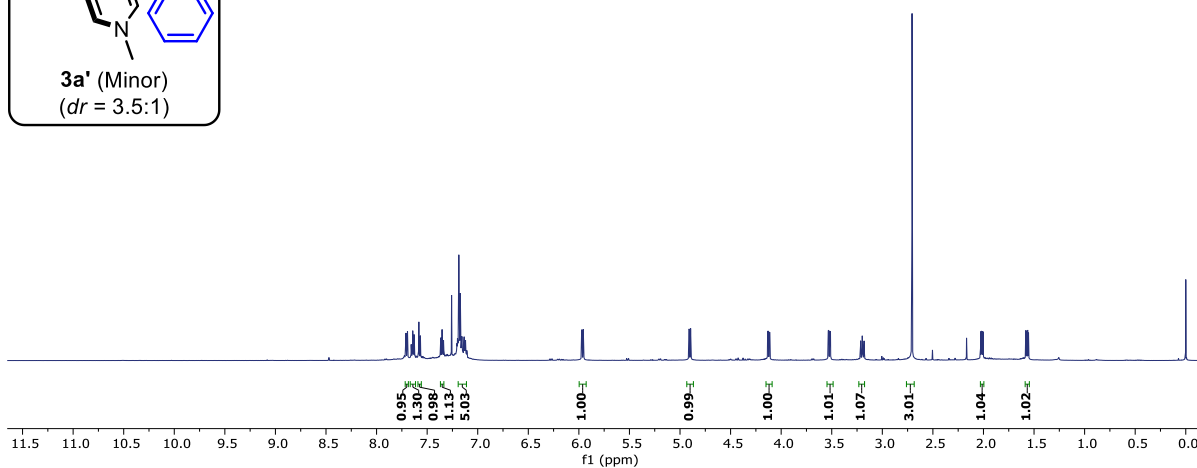
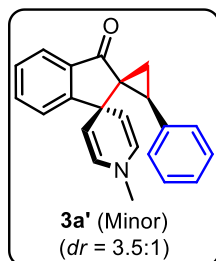


¹³C NMR (100 MHz, CDCl₃)

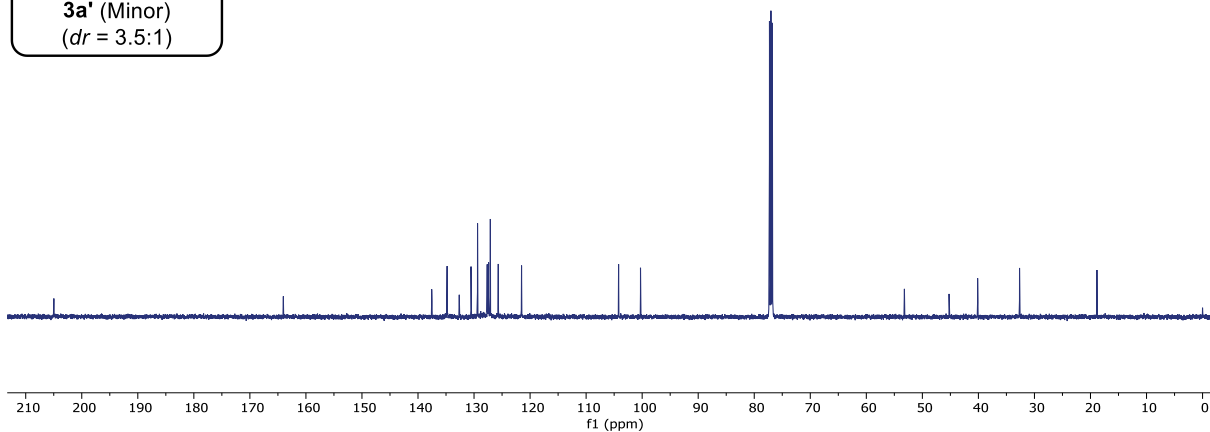
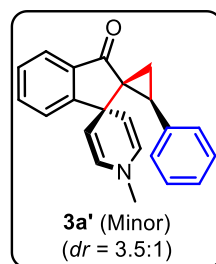
193.08
158.58
147.83
144.93
138.51
135.54
133.83
132.16
131.48
131.01
130.74
129.67
129.16
128.89
127.80
124.62
77.51
77.19
76.88
48.73



¹H NMR (500 MHz, CDCl₃)

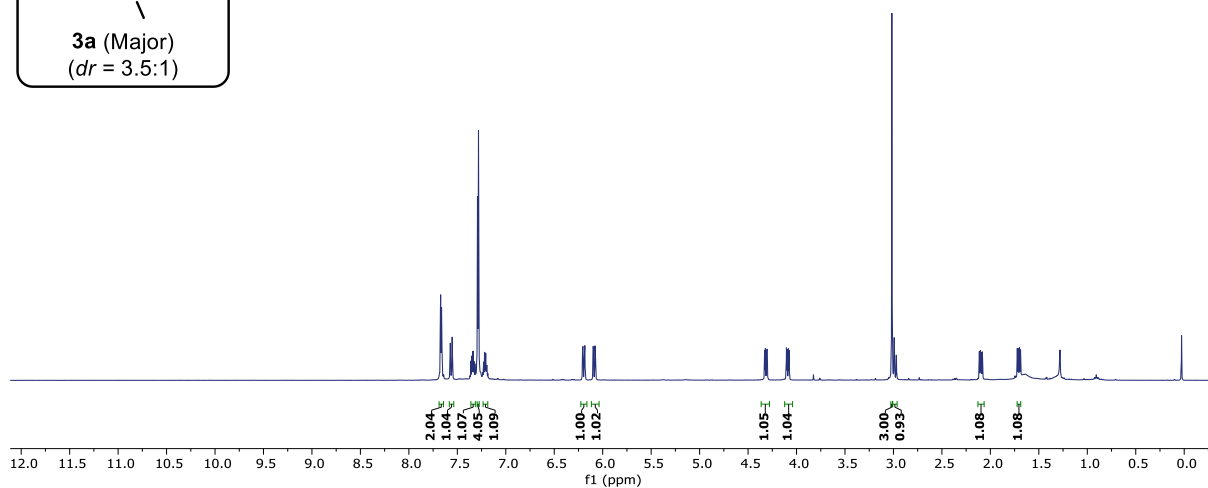
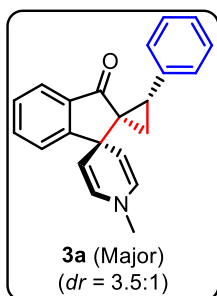


¹³C NMR (125 MHz, CDCl₃)



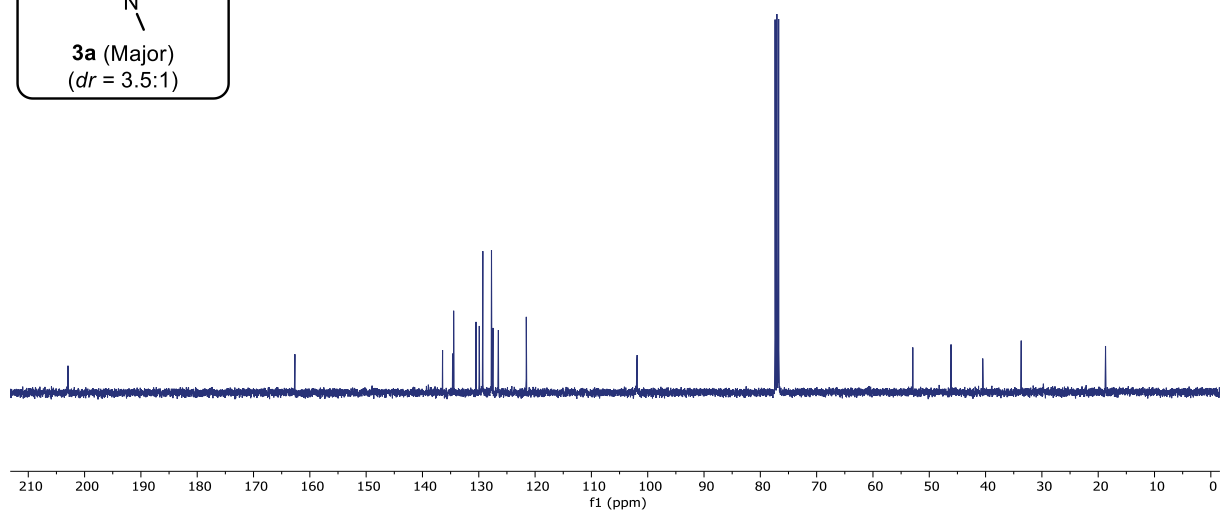
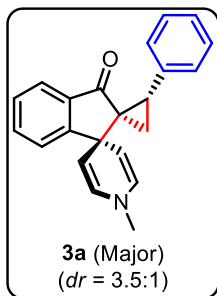
¹H NMR (400 MHz, CDCl₃)

7.6818, 7.6741, 7.6716, 7.6653, 7.6628, 7.6595, 7.5751, 7.5723, 7.5696, 7.5557, 7.5532, 7.5506, 7.3584, 7.3494, 7.3468, 7.3385, 7.3307, 7.3273, 7.3186, 7.2921, 7.2815, 7.2706, 7.2625, 7.2592, 7.2387, 7.2286, 7.2181, 7.2072, 7.1981, 7.1880, 6.2037, 6.1884, 6.1840, 6.0992, 6.0946, 6.0797, 6.0752, 4.3291, 4.3217, 4.3095, 4.3022, 4.1039, 4.0965, 4.0843, 4.0770, 3.0166, 2.9919, 2.9703, 2.1138, 2.1031, 2.0933, 2.0826, 1.7220, 1.7113, 1.6894, 1.6867

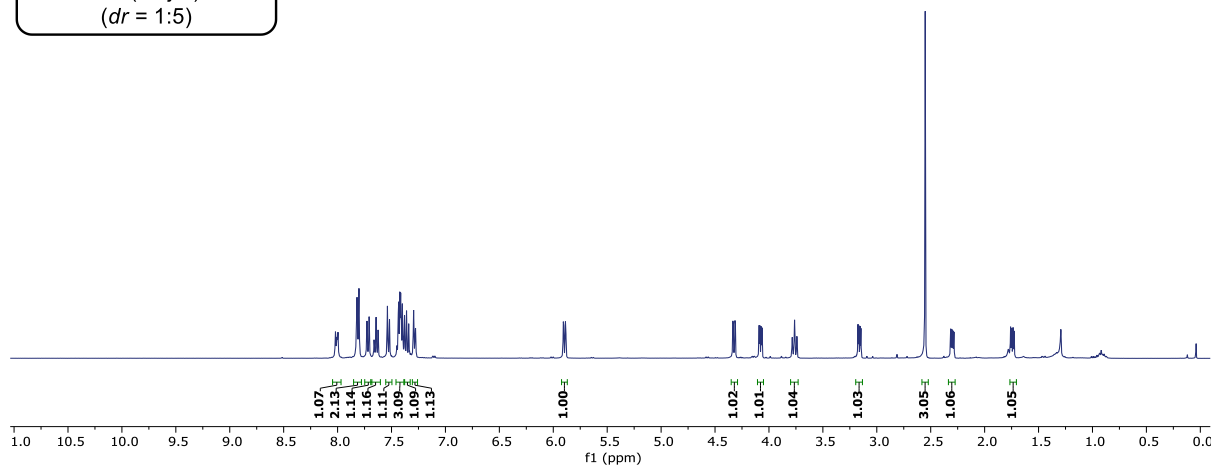
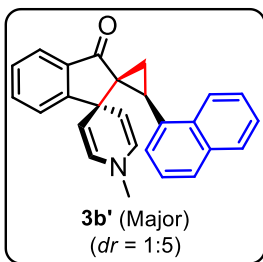
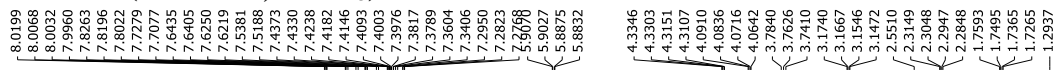


¹³C NMR (100 MHz, CDCl₃)

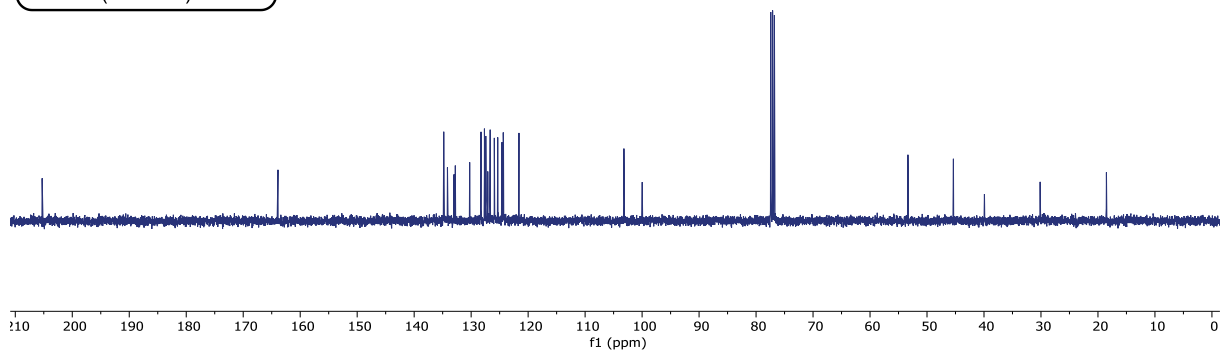
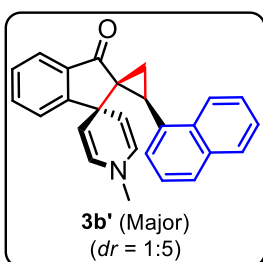
202.9499, 162.6521, 136.4329, 134.6460, 134.4622, 130.4886, 129.9097, 129.2790, 127.7421, 127.7105, 127.4760, 126.5183, 121.5628, 101.9438, 101.8790, 52.9273, 46.1357, 40.5078, 33.6704, 18.7015



¹H NMR (400 MHz, CDCl₃)

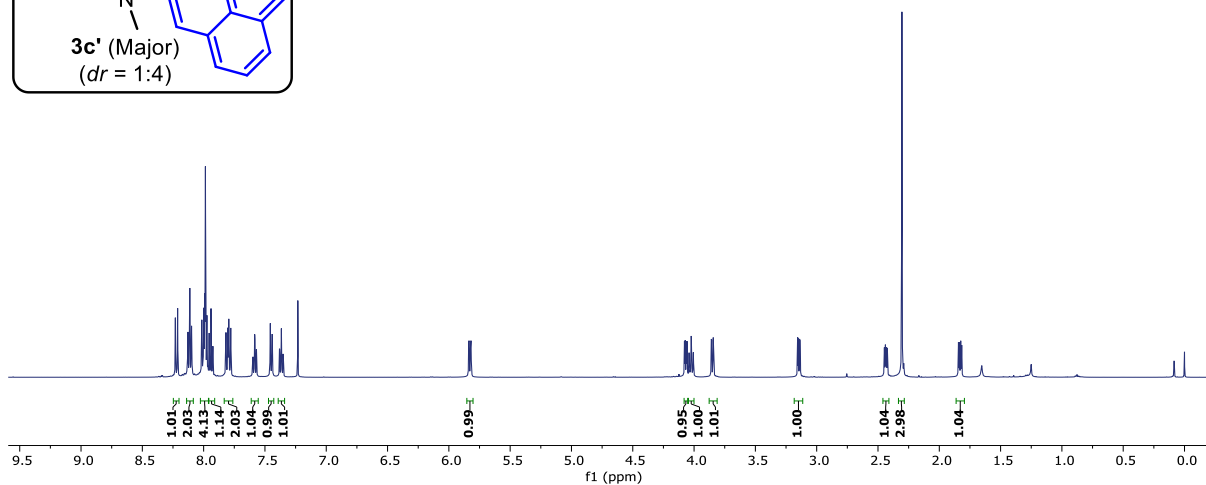
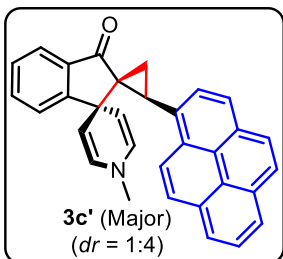


¹³C NMR (100 MHz, CDCl₃)



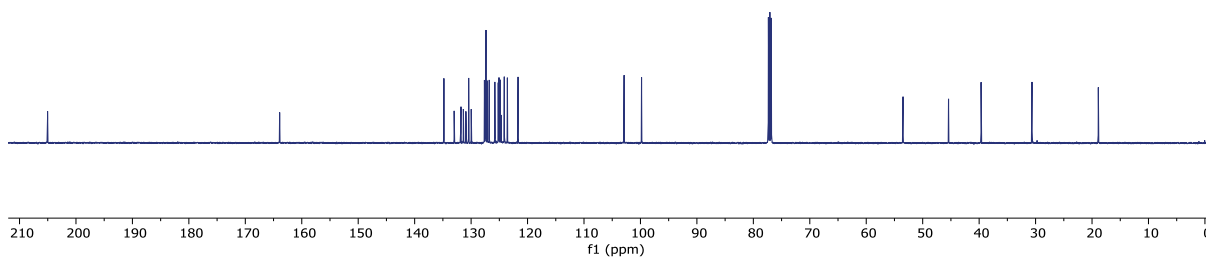
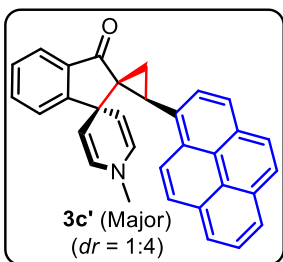
¹H NMR (500 MHz, CDCl₃)

8.2307
8.2122
8.1297
8.1274
8.1147
8.1124
8.0984
8.0148
7.9990
7.9919
7.9856
7.9734
7.9649
7.9398
7.9245
7.8200
7.8180
7.8160
7.8046
7.8027
7.8008
7.7788
7.7768
7.5969
7.5844
7.5819
7.5696
7.5671
7.4582
7.4562
7.4544
7.4427
7.4409
7.4390
7.3819
7.3798
7.3667
7.3646
7.3520
7.3500
7.2320
5.8383
5.8349
5.8228
5.8193
4.0785
4.0725
4.0629
4.0570
4.0405
4.0233
4.0060
3.8596
3.8561
3.8439
3.8404
3.1556
3.1497
3.1401
3.1341
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2.4317
2.4236
2.3051
1.8435
1.8354
1.8252
1.8170



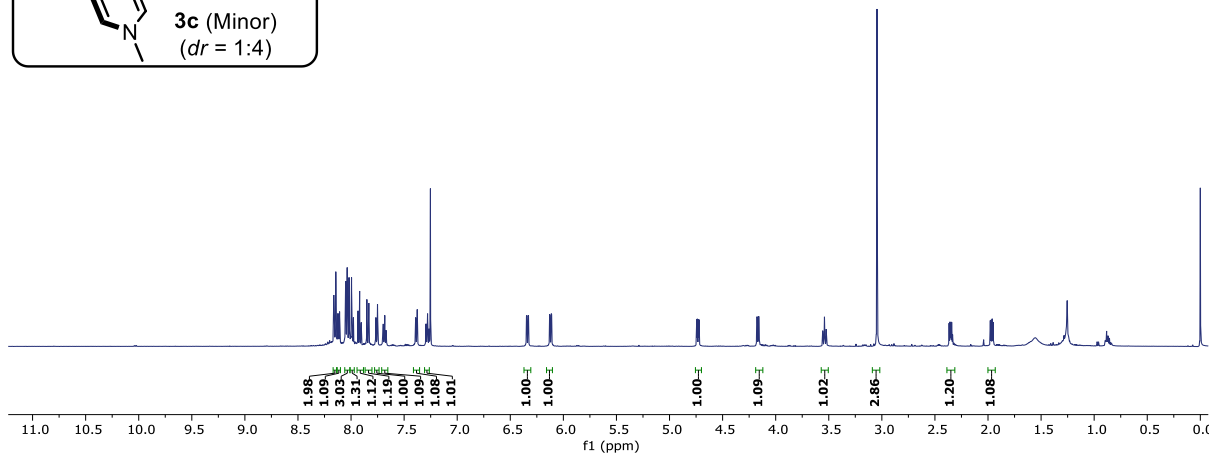
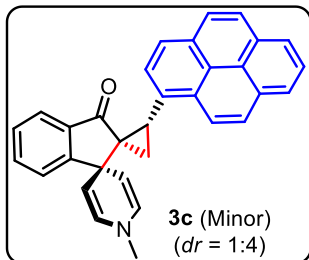
¹³C NMR (125 MHz, CDCl₃)

205.0324
163.9154
134.8203
133.0006
131.8501
131.7915
131.3698
130.9239
130.4312
129.9759
127.6347
127.4589
127.3653
127.1760
126.7861
125.7970
125.1913
125.0615
124.8626
124.7051
124.1211
124.1013
123.5730
121.6770
102.8940
99.8064
53.4788
45.4217
39.6562
30.6356
18.8850



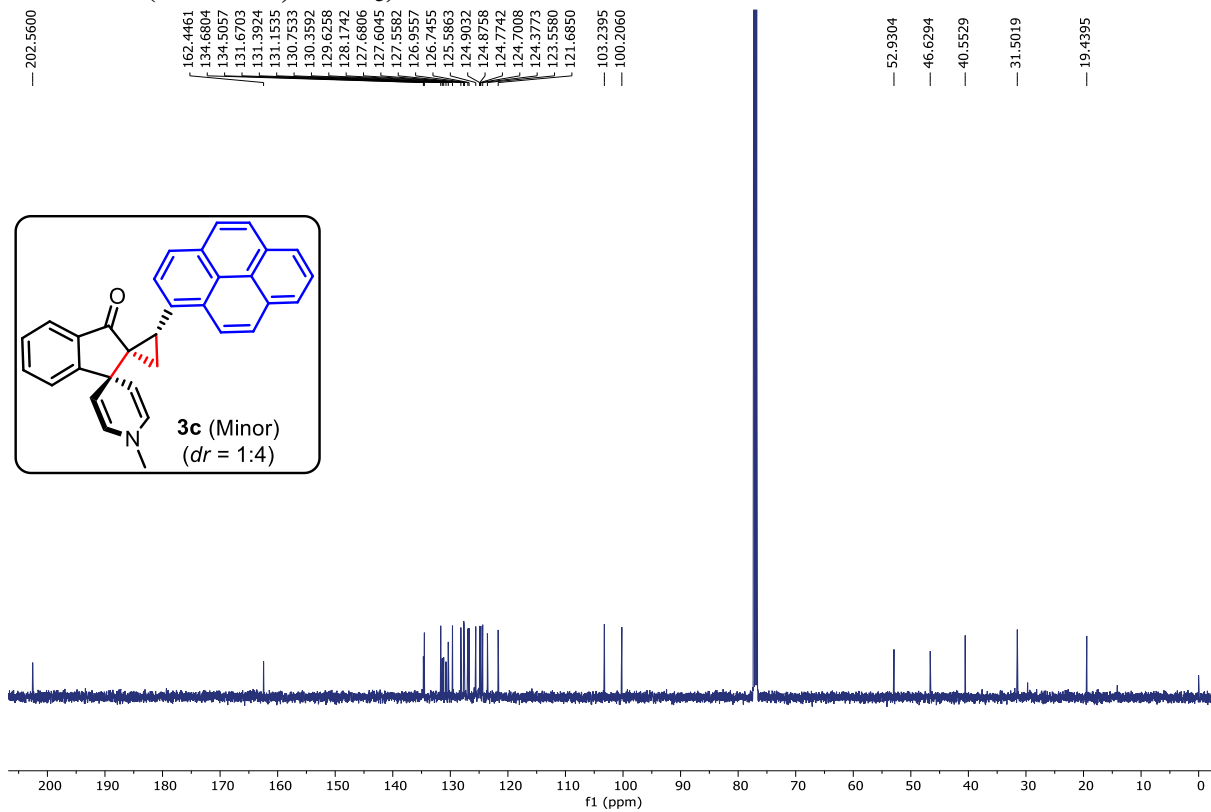
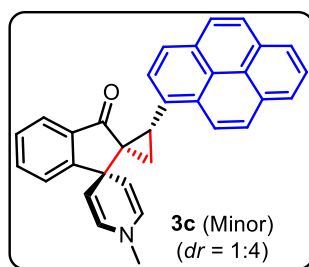
¹H NMR (500 MHz, CDCl₃)

8.1626, 8.1419, 8.1249, 8.1226, 8.1098, 8.1075, 8.0540, 8.0334, 8.0188, 7.9954, 7.9776, 7.9341, 7.9189, 7.9038, 7.8510, 7.8326, 7.7682, 7.7488, 7.6991, 7.6991, 7.3934, 7.3915, 7.3894, 7.3783, 7.3763, 7.3741, 7.2956, 7.2524, 6.3478, 6.3442, 6.3320, 6.3285, 6.1296, 6.1262, 6.1141, 6.1105, 4.7438, 4.7379, 4.7281, 4.7222, 4.1782, 4.1724, 4.1625, 4.1567, 3.5571, 3.5404, 3.5236, 3.0454, 2.3665, 2.3579, 2.3508, 2.3422, 1.9783, 1.9698, 1.9604, 1.9519



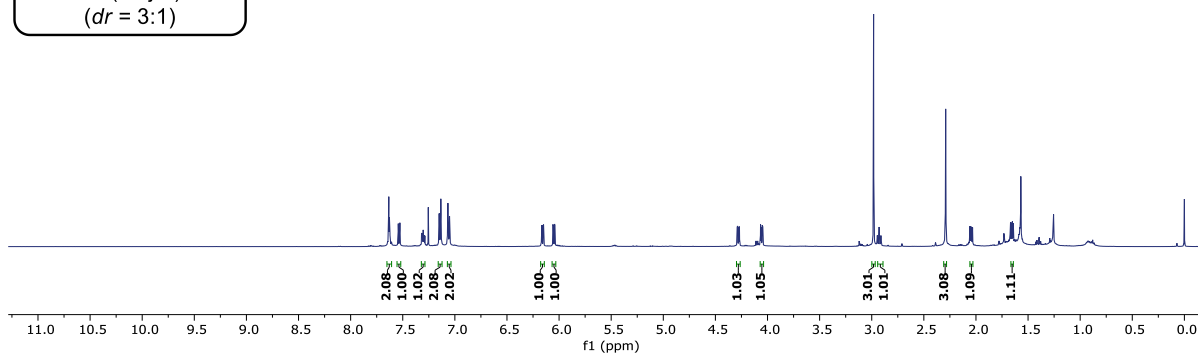
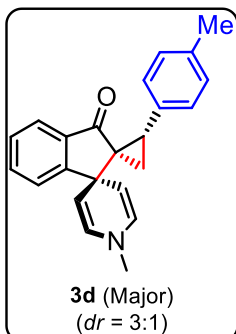
¹³C NMR (125 MHz, CDCl₃)

202.5600, 162.4461, 134.6804, 134.5057, 131.6703, 131.3924, 131.1535, 130.7533, 130.3592, 129.6258, 128.1742, 127.6806, 127.6045, 127.5582, 126.9557, 126.7455, 125.5863, 124.9032, 124.8758, 124.7742, 124.7008, 124.3773, 123.5580, 121.6850, 103.2395, 100.2060, 52.9304, 46.6294, 40.5529, 31.5019, 19.4395



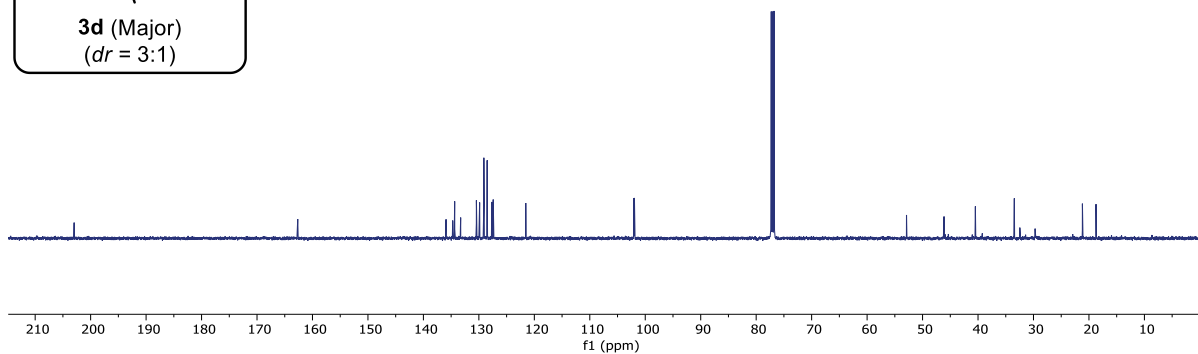
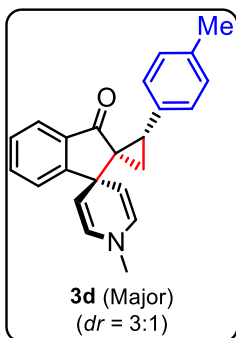
¹H NMR (500 MHz, CDCl₃)

7.6360
7.6340
7.6297
7.6280
7.6258
7.6235
7.5449
7.5429
7.5409
7.5296
7.5276
7.5256
7.3197
7.3134
7.3094
7.3044
7.3032
7.2984
7.2939
7.2879
7.2849
7.1505
7.1466
7.1387
7.1345
7.0668
7.0514
7.0487
6.1646
6.1524
6.1489
6.0590
6.0554
6.0433
6.0397
6.0375
4.2909
4.2851
4.2753
4.2694
4.0703
4.0669
4.0609
4.0511
4.0452
2.9821
2.9462
2.9291
2.9119
2.2905
2.0574
2.0489
2.0411
2.0326
1.6682
1.6597
1.6501
1.6415

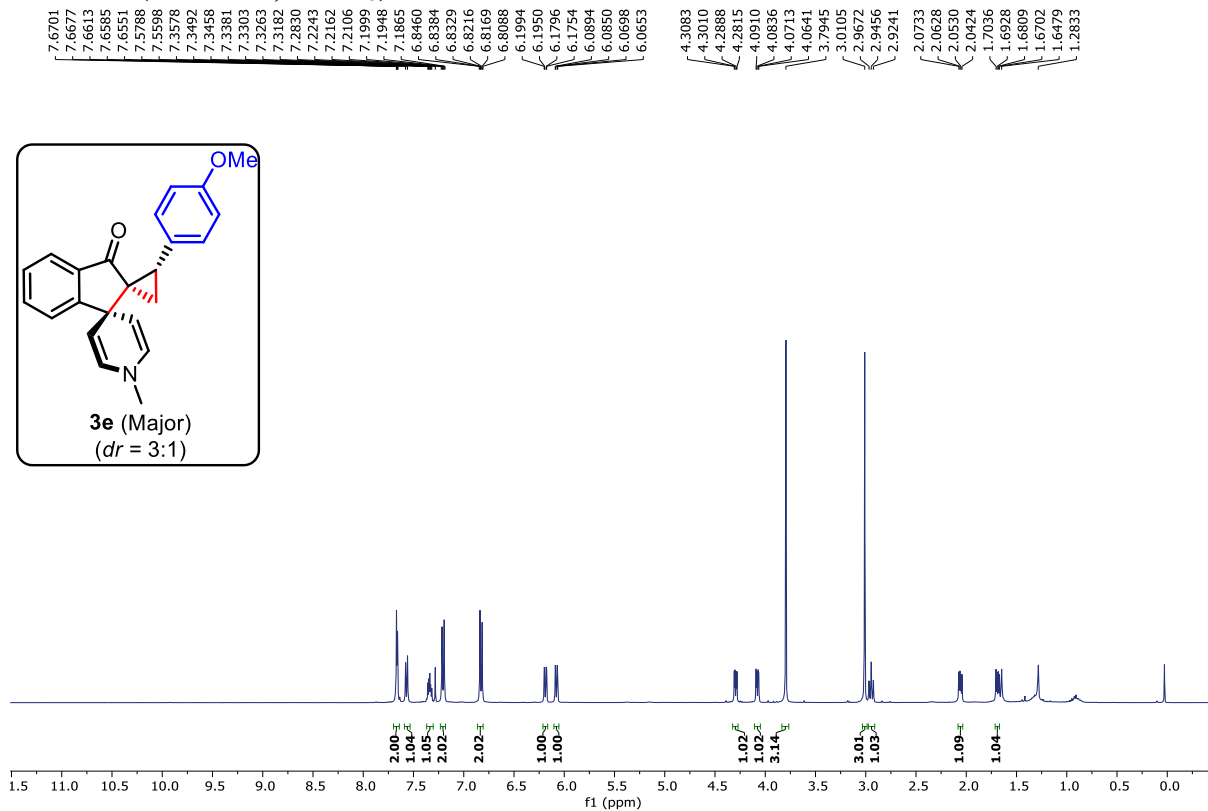


¹³C NMR (125 MHz, CDCl₃)

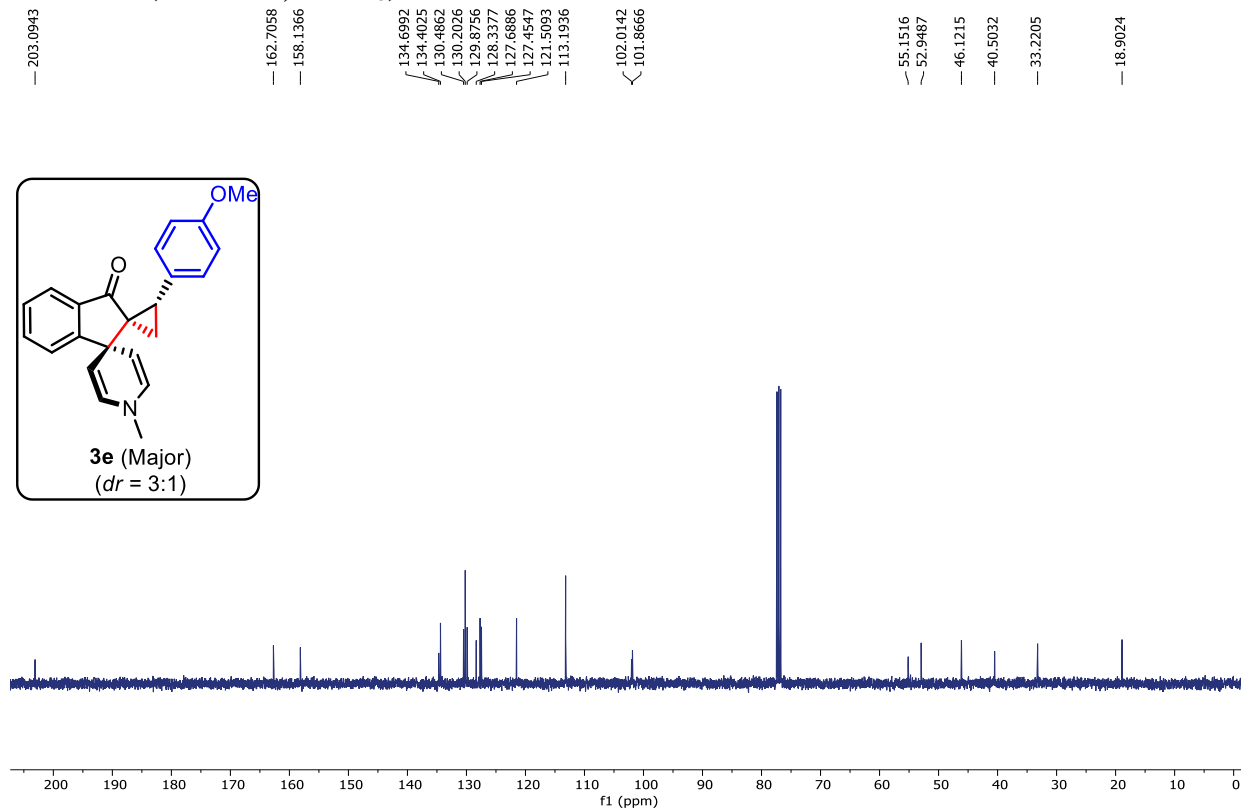
202.9738
162.6578
135.9163
134.7171
134.3617
133.2839
130.4435
129.8591
129.1007
128.5019
127.6616
127.4153
121.5411
102.0490
101.9548
77.2811
77.0280
76.7730
52.8942
46.1267
40.4782
33.4560
21.1783
18.7271



¹H NMR (400 MHz, CDCl₃)

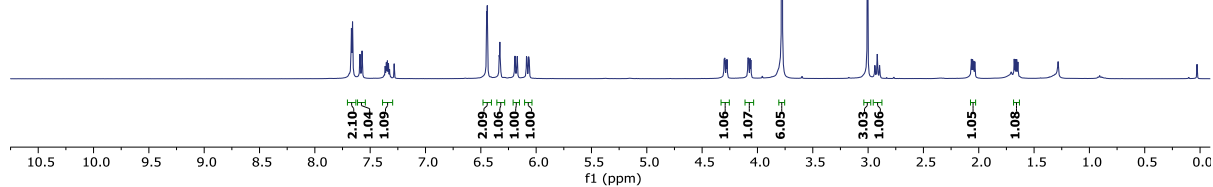
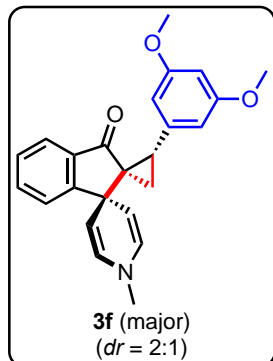


¹³C NMR (100 MHz, CDCl₃)



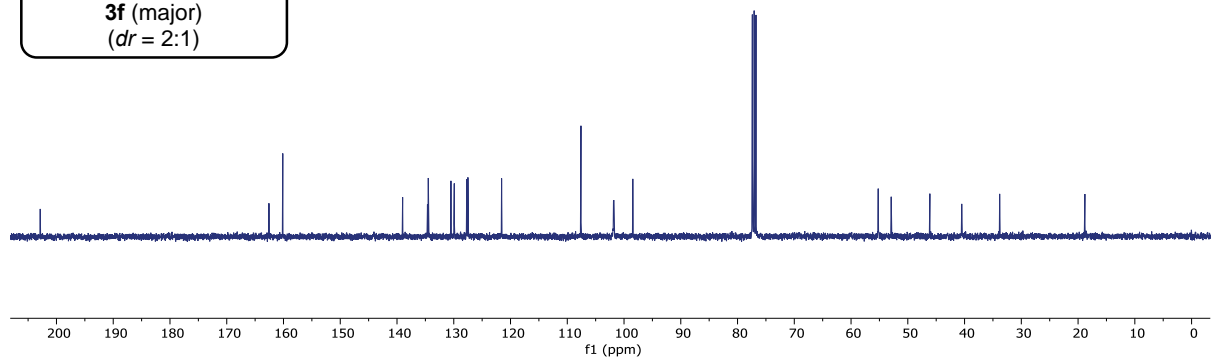
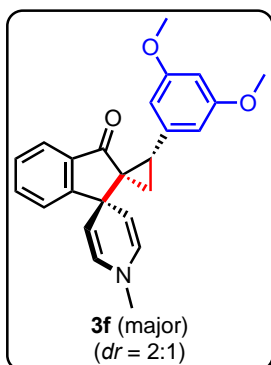
¹H NMR (400 MHz, CDCl₃)

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7.3552
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7.3444
7.3347
7.3245
7.2830
6.4472
6.4414
6.3339
6.3282
6.3225
6.1942
6.1898
6.1747
6.1702
6.0875
6.0830
6.0678
6.0634
4.3015
4.2941
4.2819
4.2745
4.0865
4.0791
4.0668
4.0594
3.8129
3.8097
3.7973
3.7788
3.7629
3.0054
2.9389
2.9174
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1.6569
1.6463
1.2829



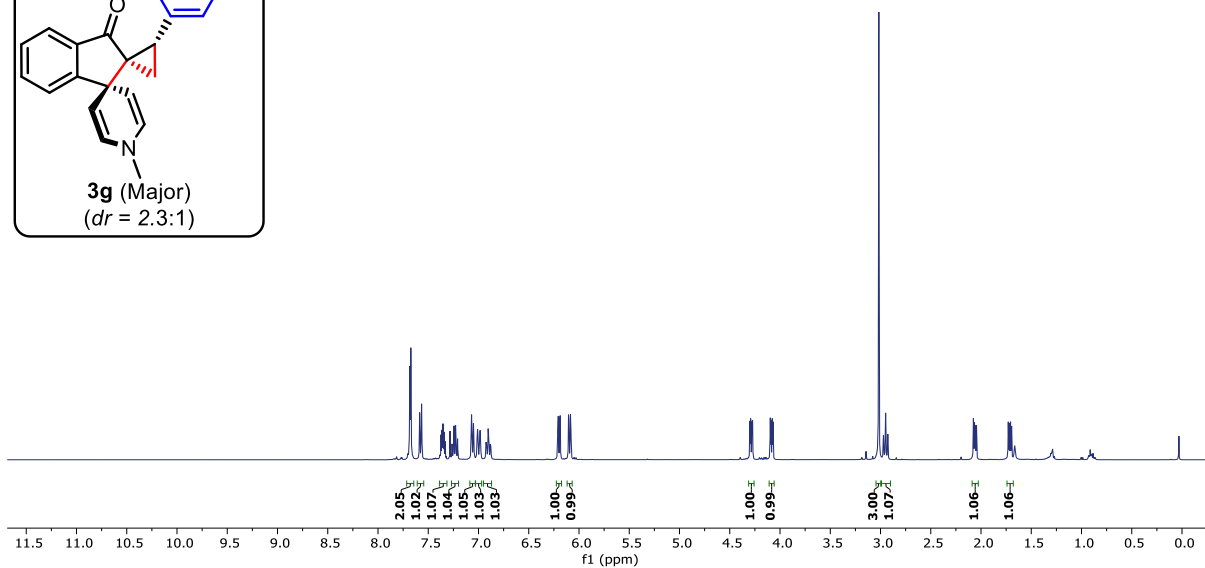
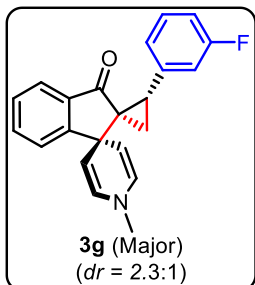
¹³C NMR (100 MHz, CDCl₃)

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134.6296
134.4871
130.4985
129.9081
127.7097
127.5056
121.5712
107.5897
101.8570
101.7727
98.4547
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33.8138
18.8007



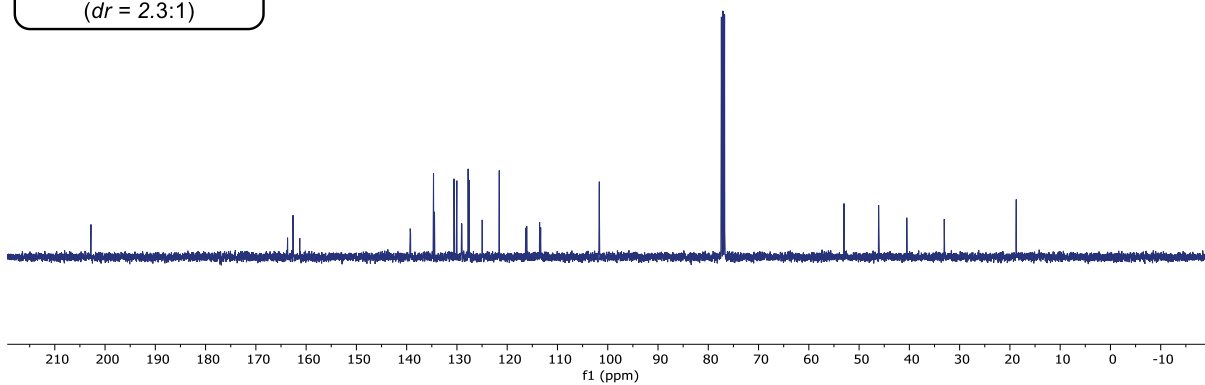
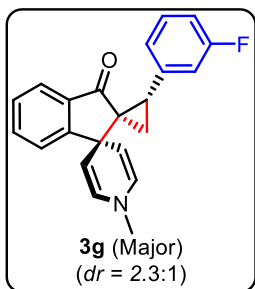
¹H NMR (400 MHz, CDCl₃)

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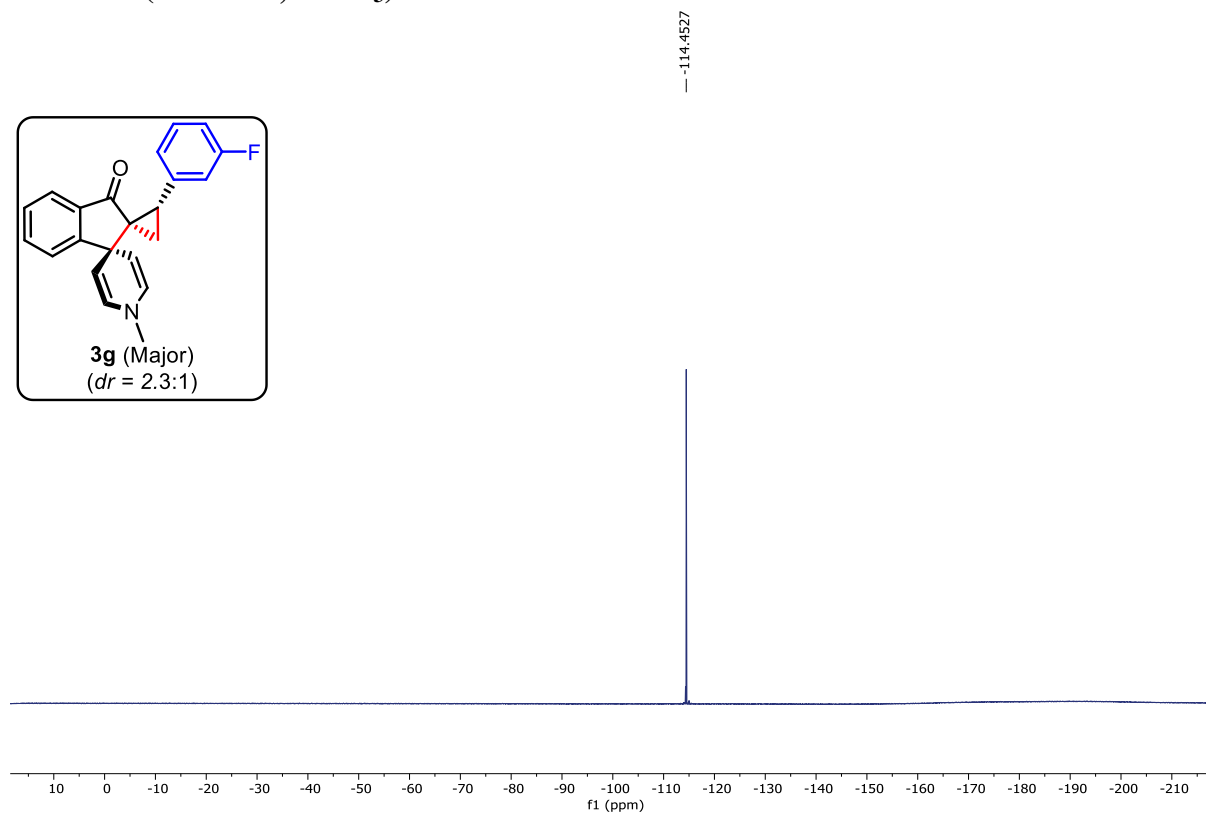
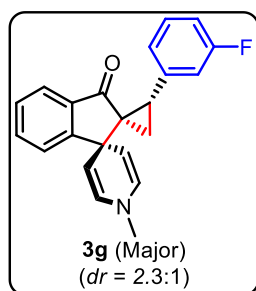


¹³C NMR (100 MHz, CDCl₃)

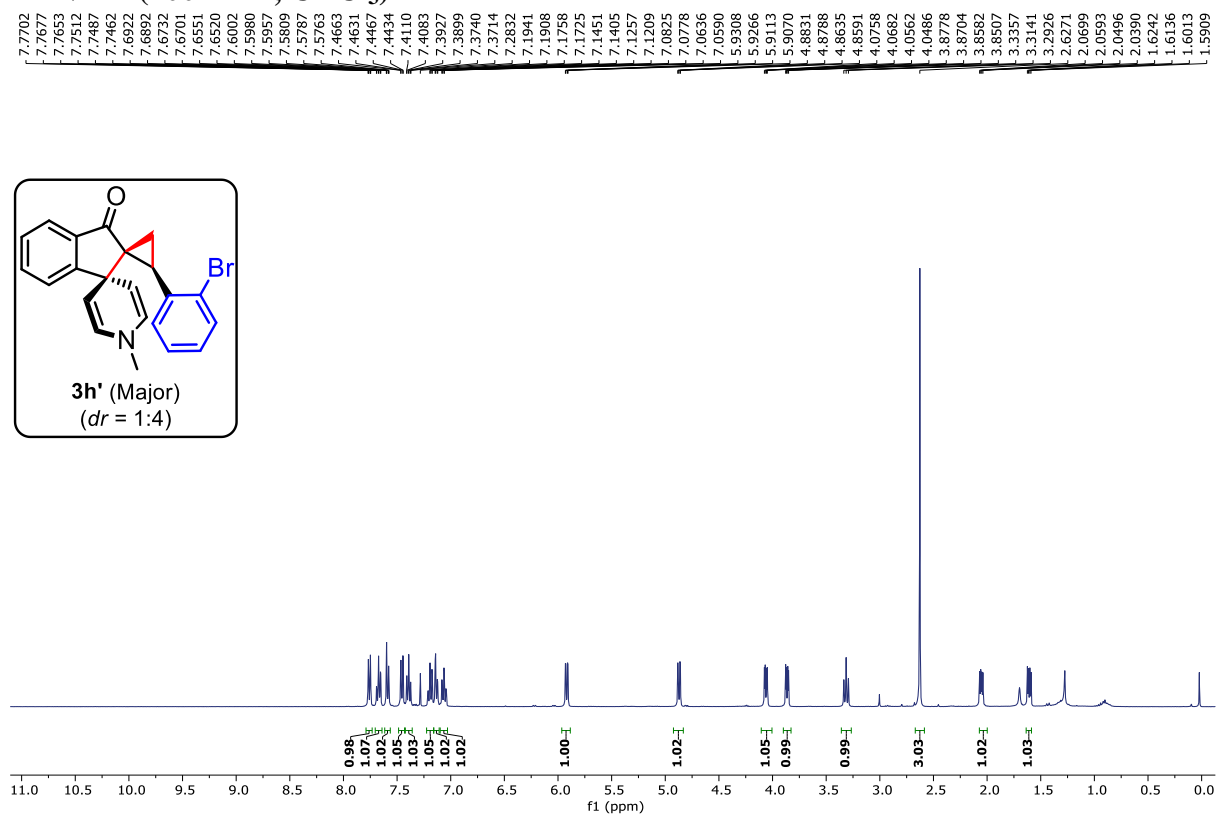
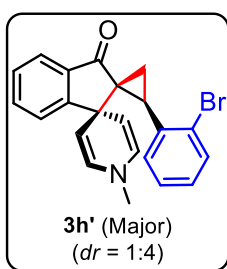
202.80, 163.70, 162.61, 161.26, 139.29, 139.22, 134.66, 134.47, 130.58, 130.02, 130.00, 129.09, 129.00, 127.78, 127.58, 124.97, 124.94, 121.58, 116.33, 116.12, 113.55, 113.34, 101.68, 53.00, 46.12, 40.51, 33.07, 18.74



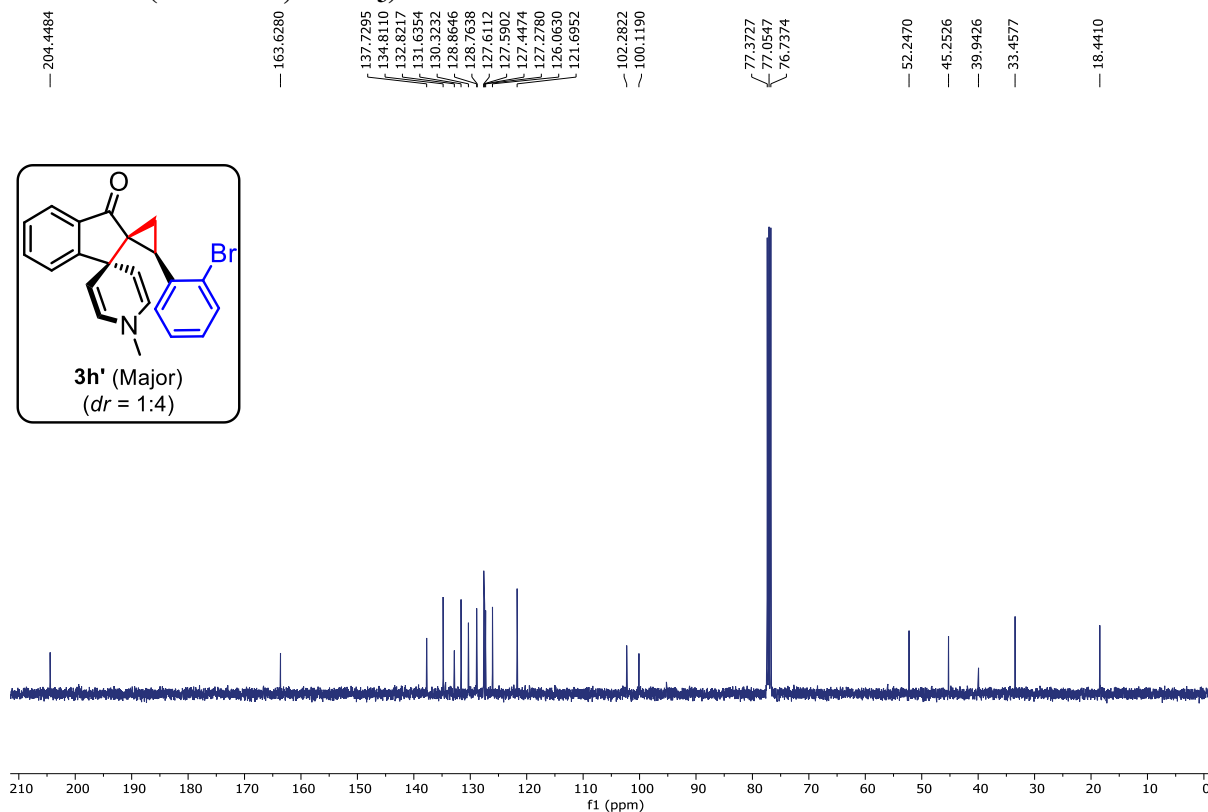
^{19}F NMR (376 MHz, CDCl_3)



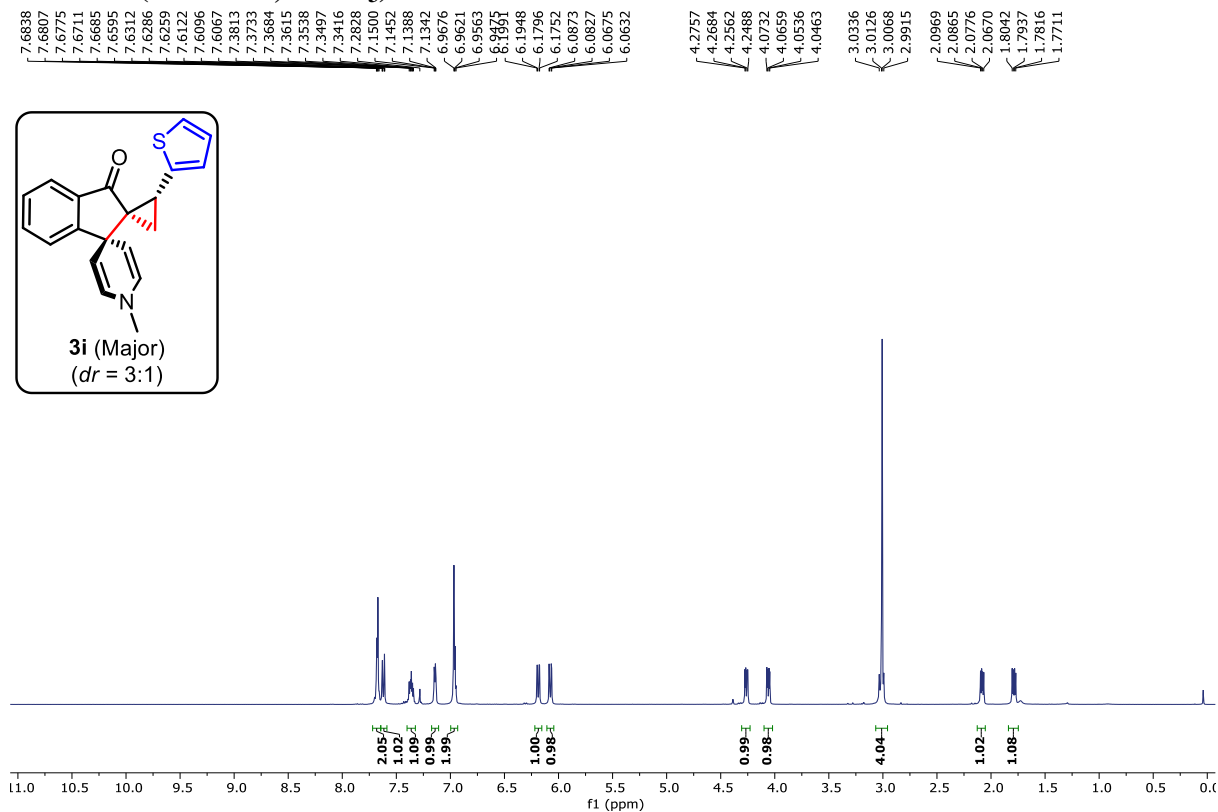
^1H NMR (400 MHz, CDCl_3)



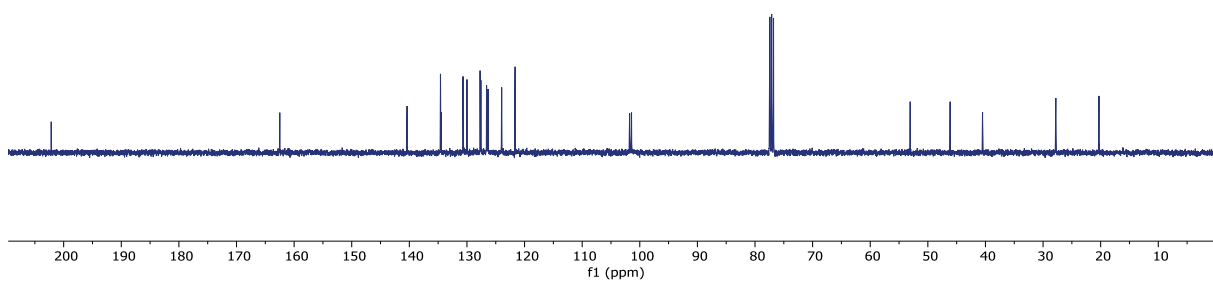
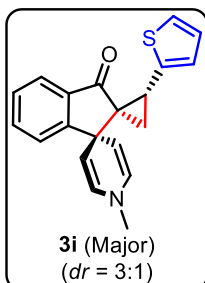
¹³C NMR (100 MHz, CDCl₃)



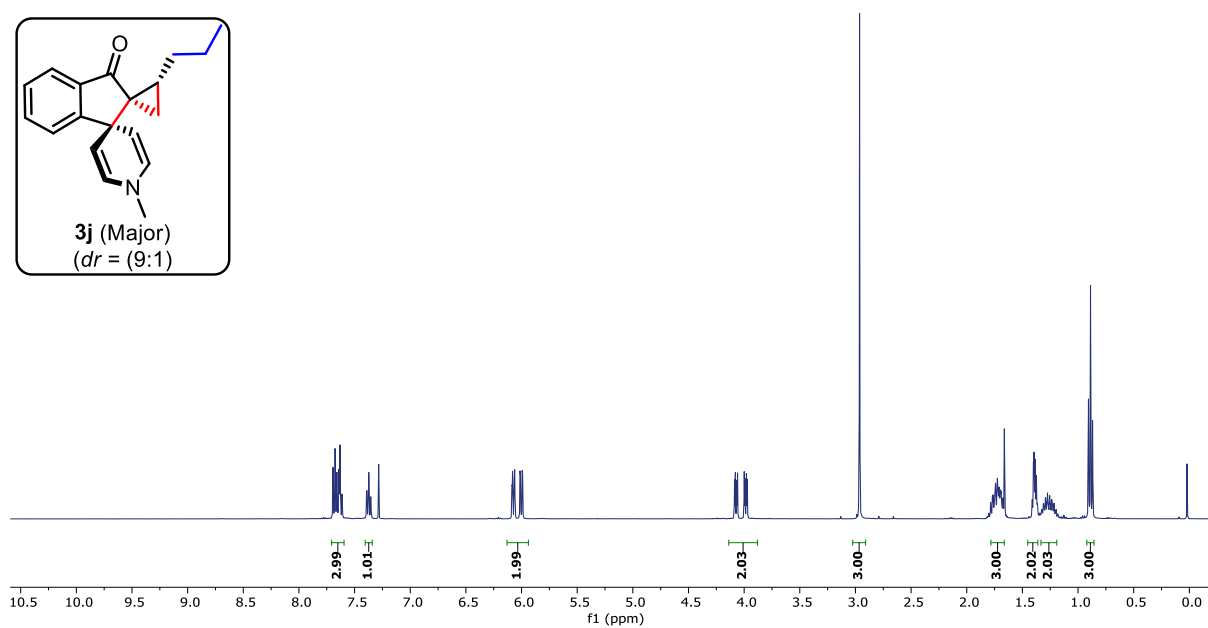
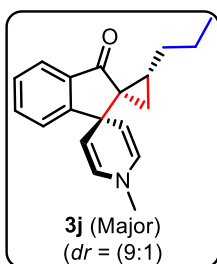
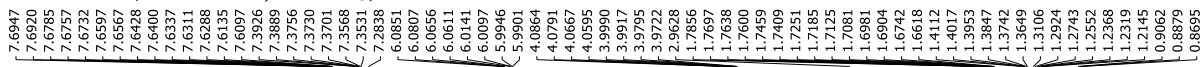
¹H NMR (400 MHz, CDCl₃)



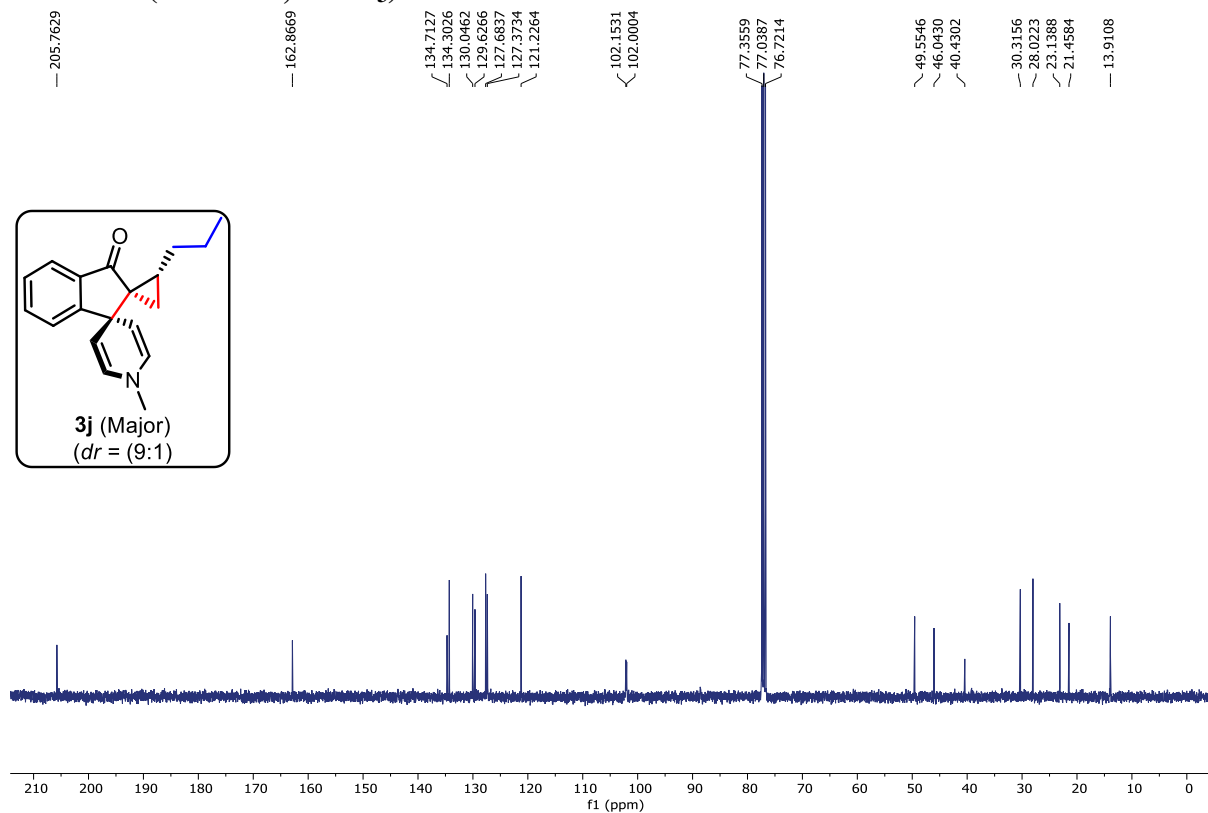
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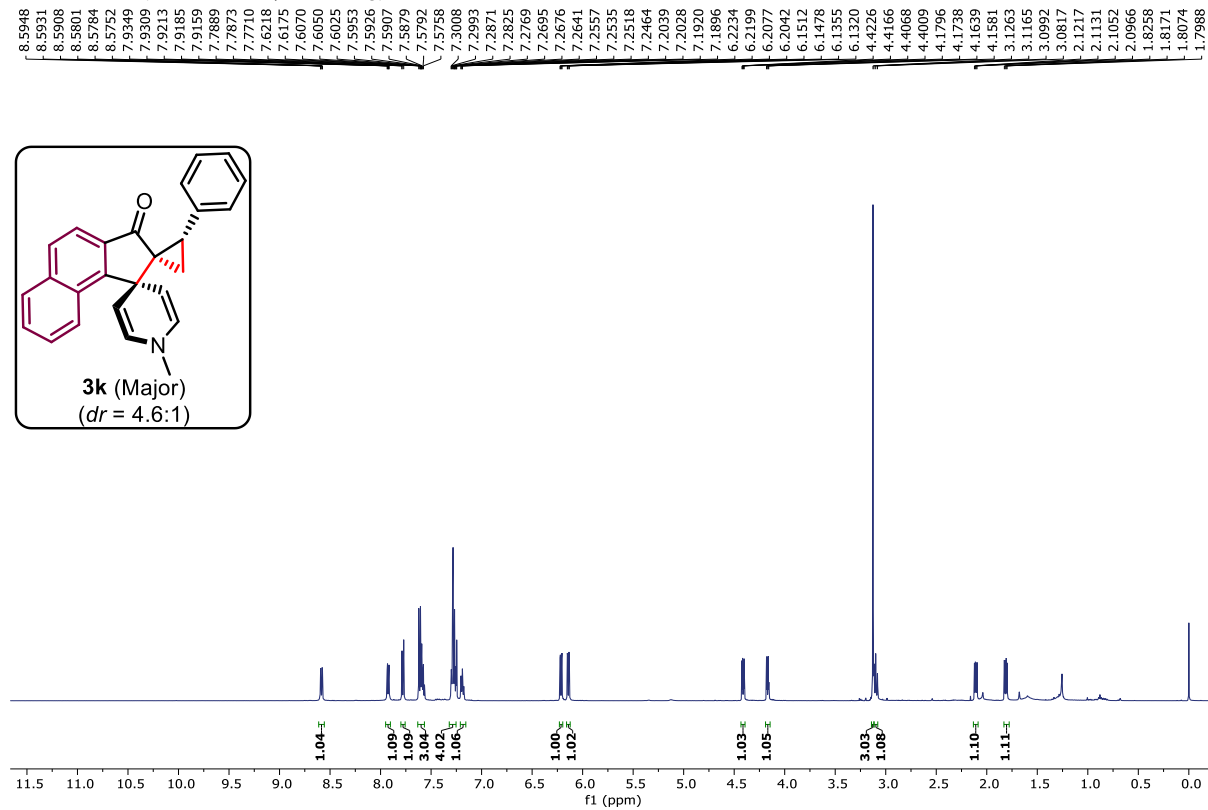
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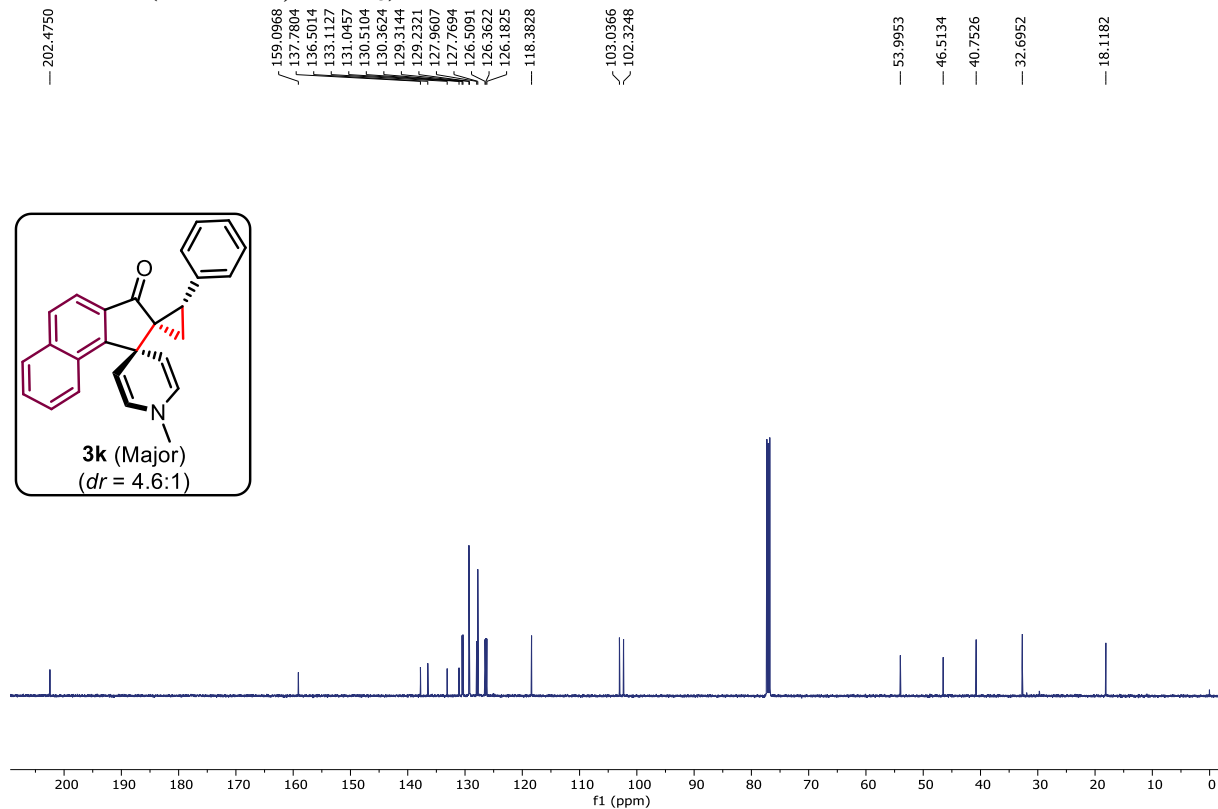
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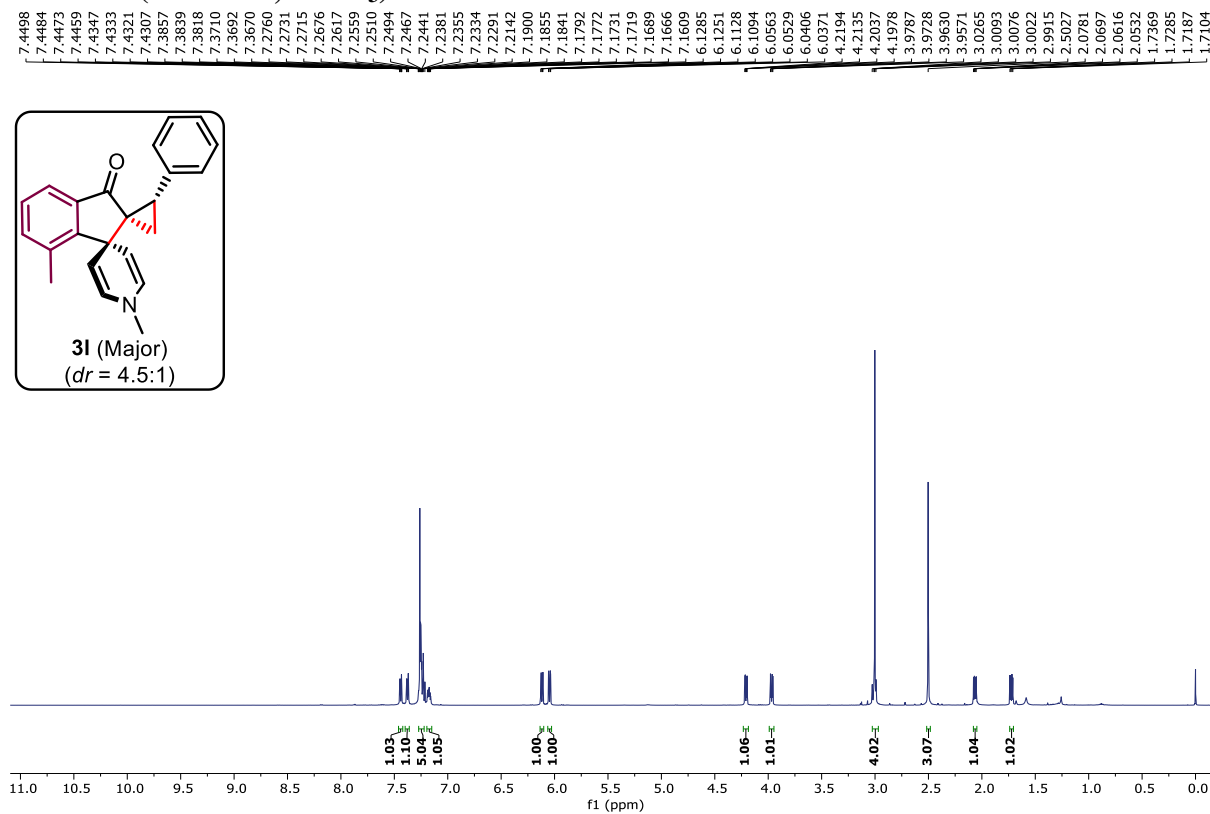
¹H NMR (500 MHz, CDCl₃)



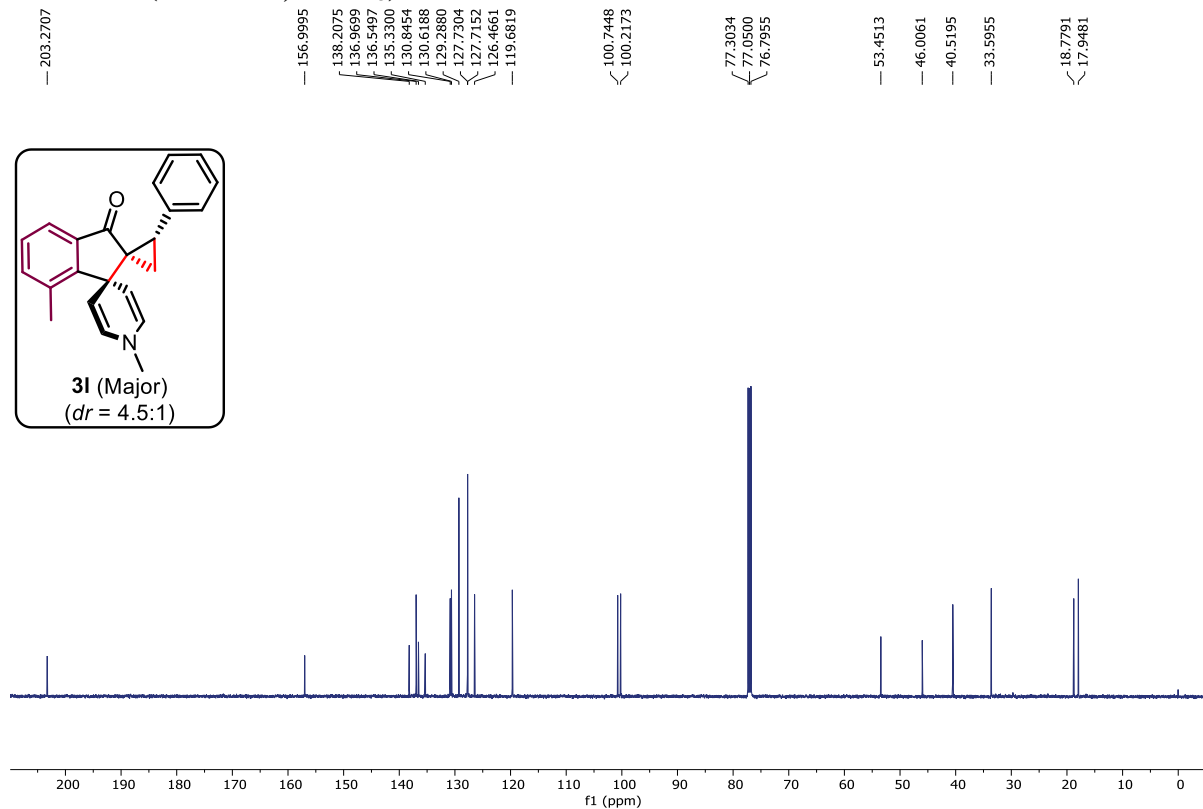
^{13}C NMR (125 MHz, CDCl_3)



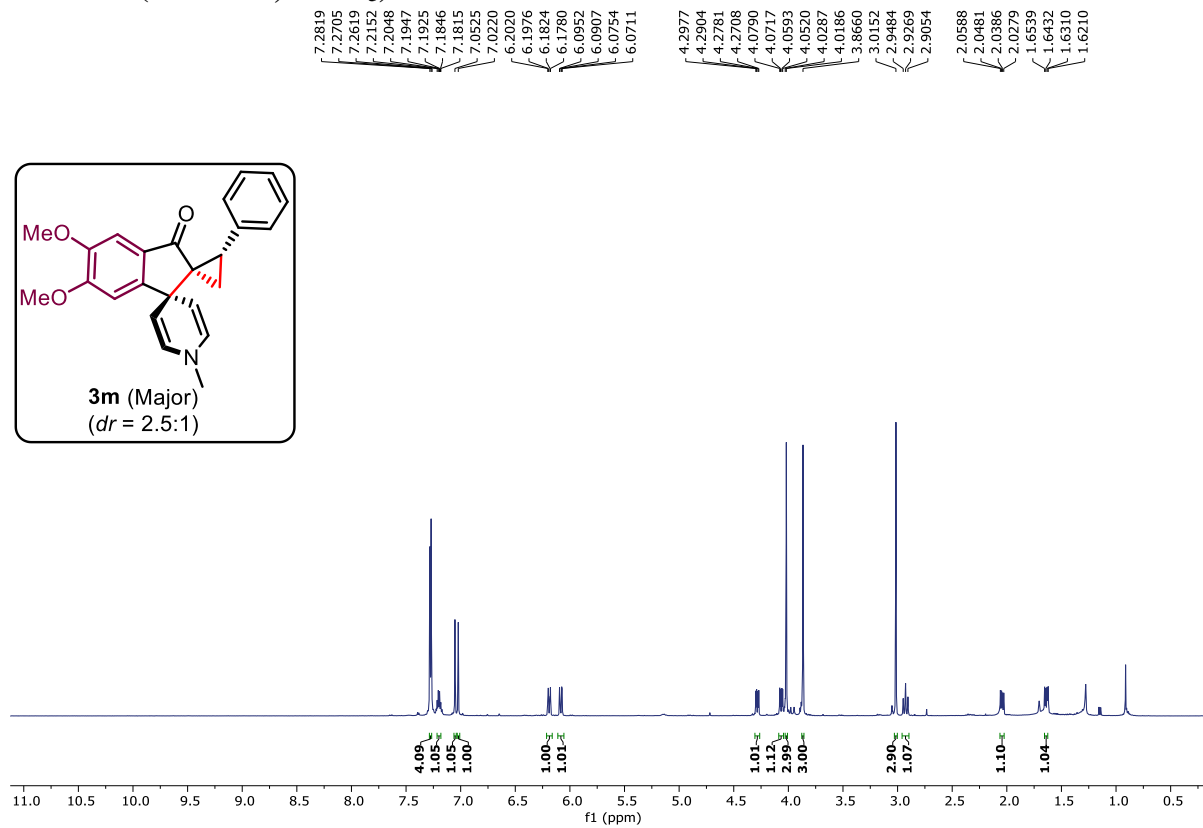
^1H NMR (500 MHz, CDCl_3)



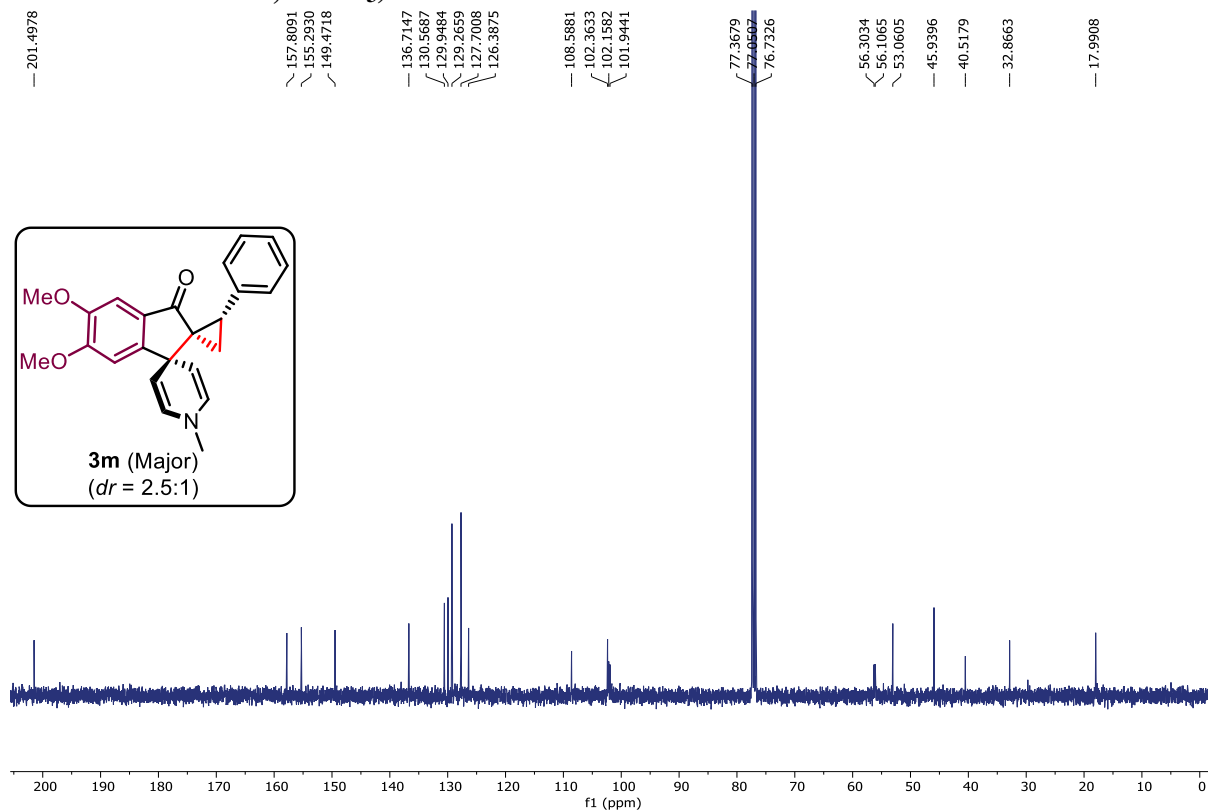
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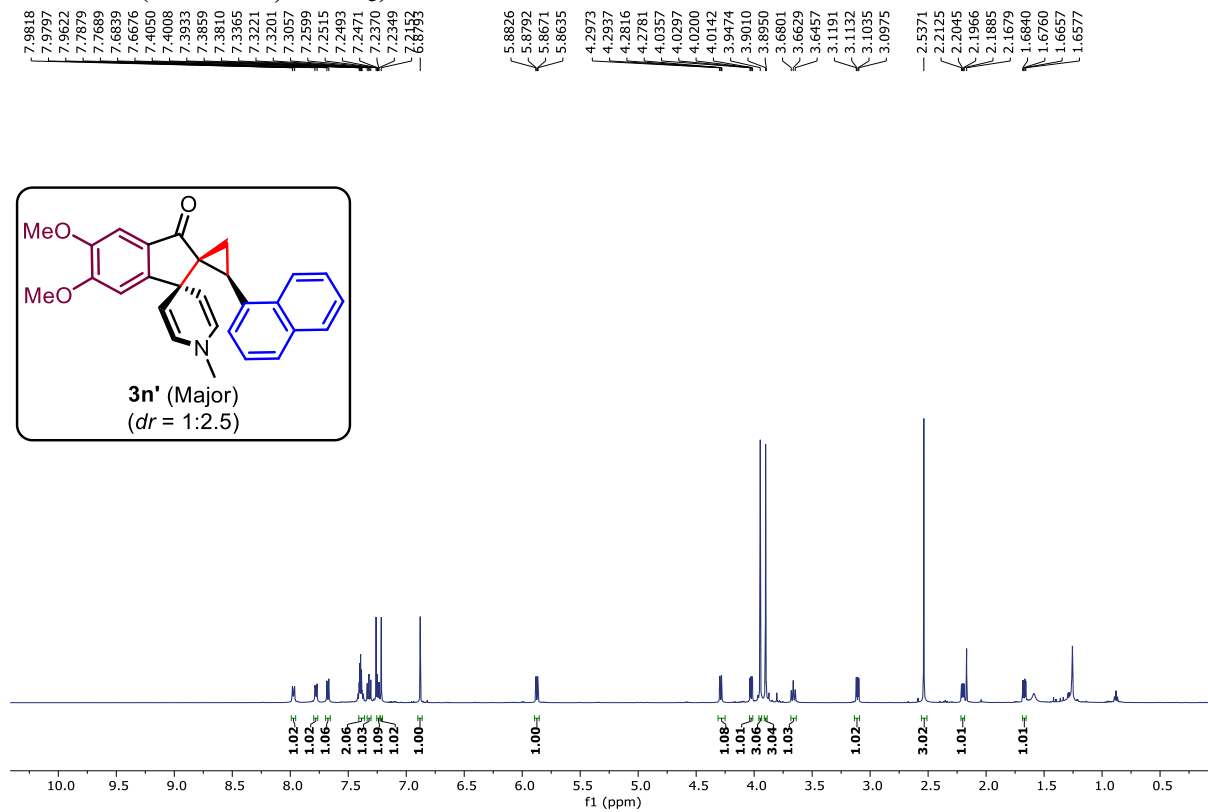
¹H NMR (400 MHz, CDCl₃)



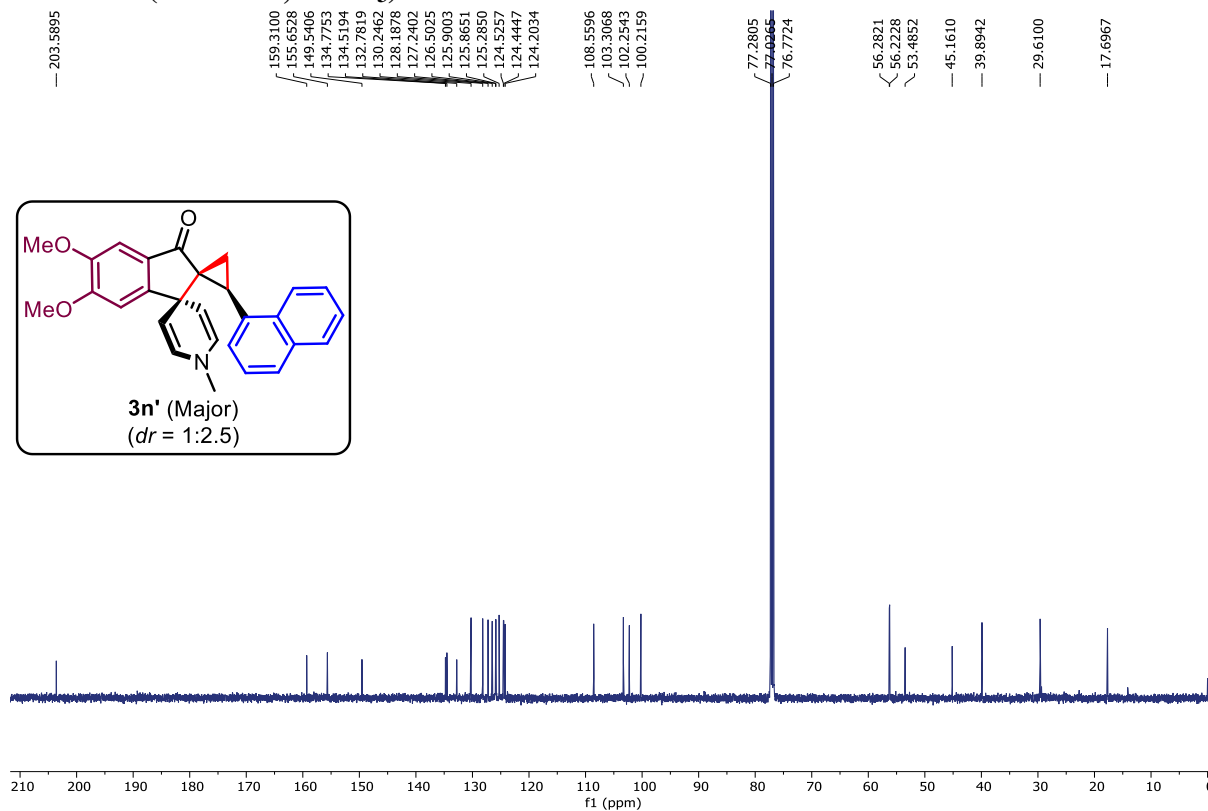
¹³C NMR 100 MHz, CDCl₃



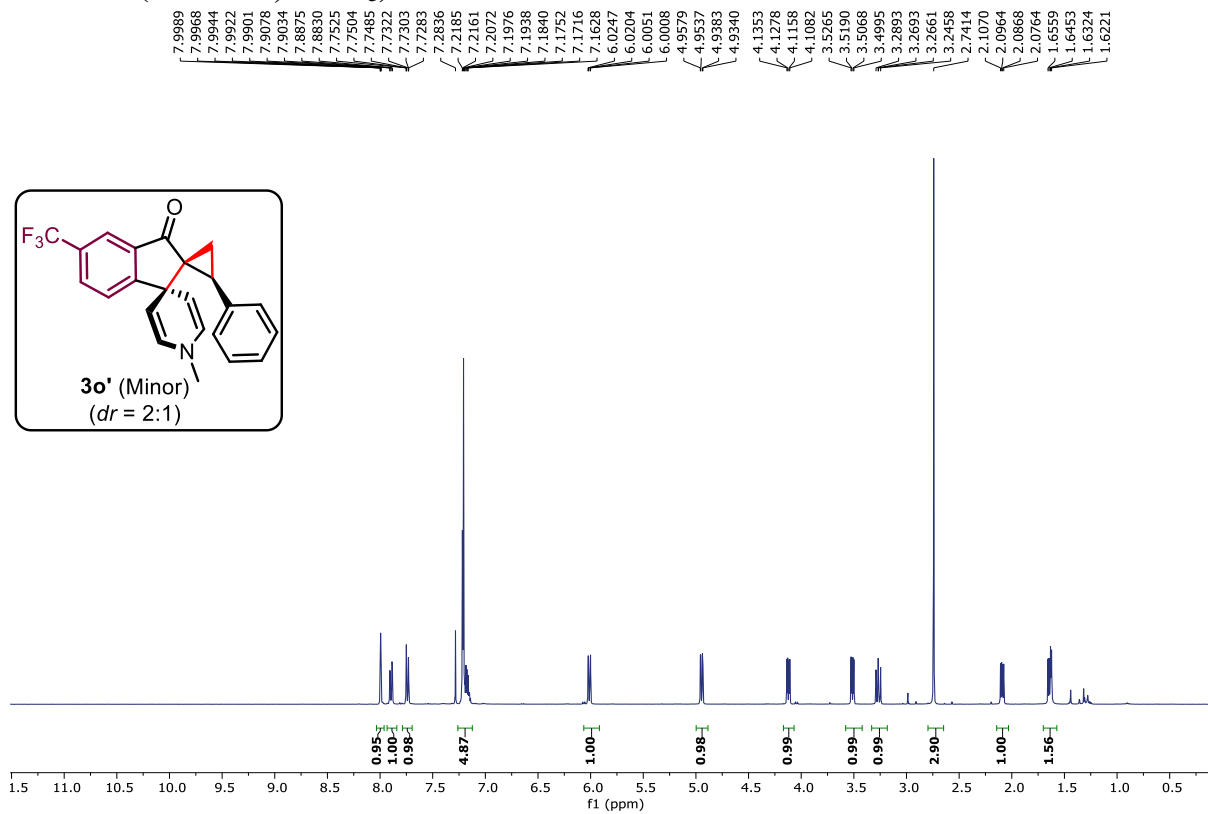
¹H NMR (500 MHz, CDCl₃)



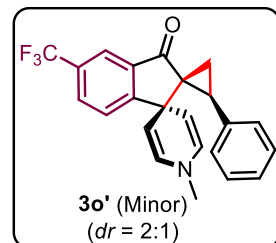
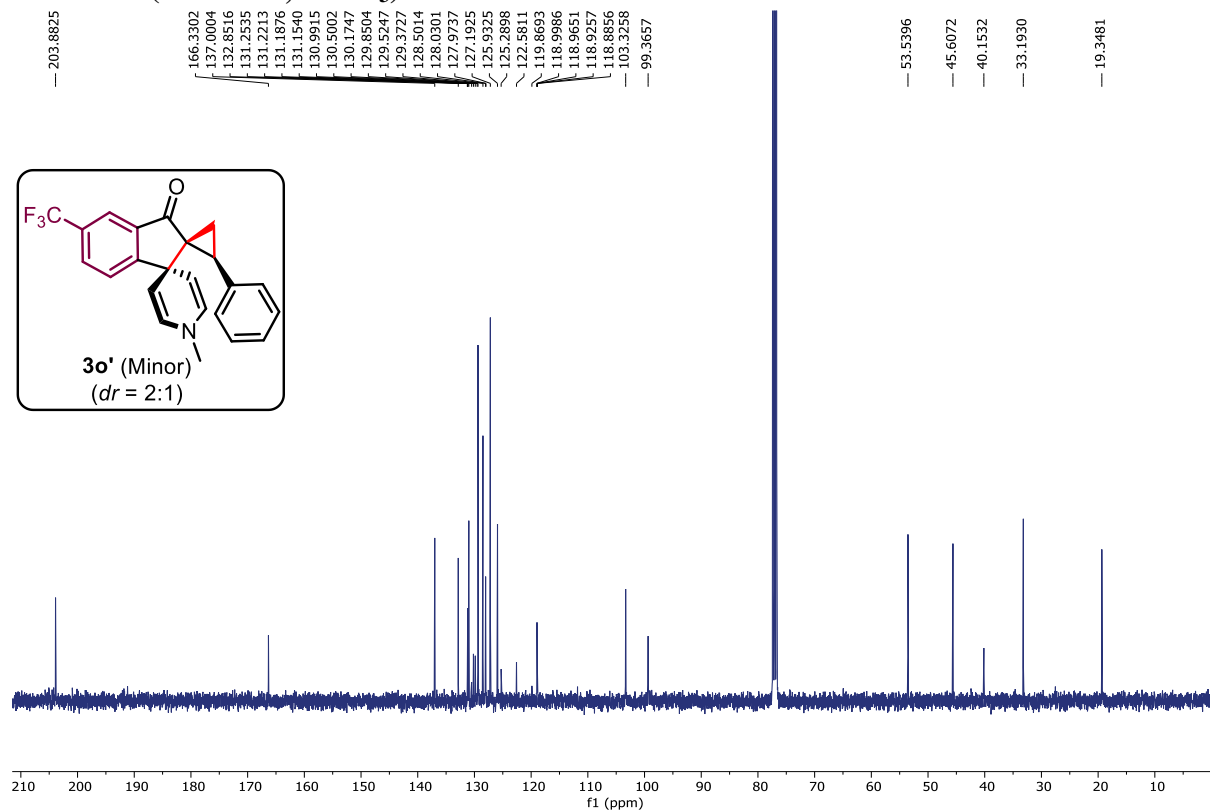
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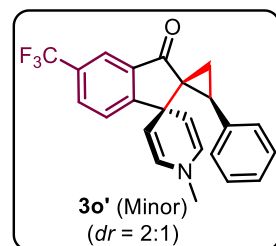
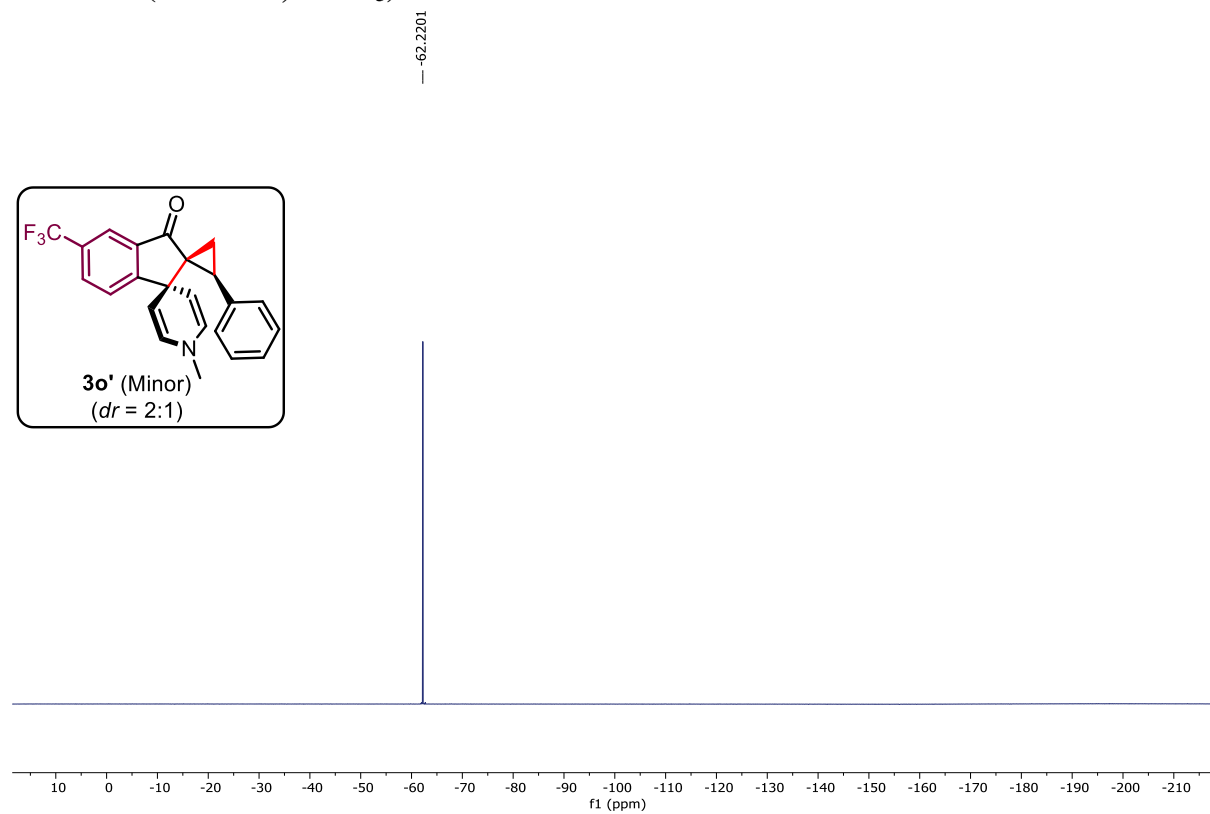
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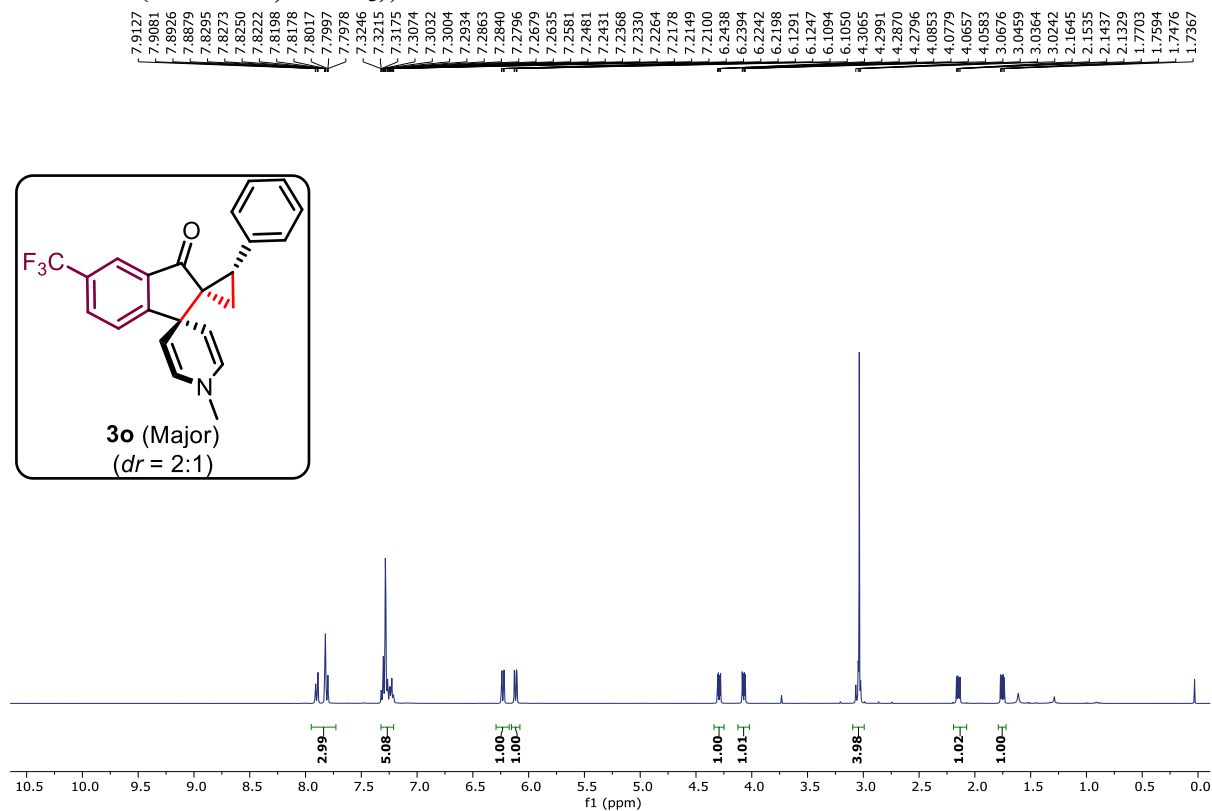
^{13}C NMR (100 MHz, CDCl_3)



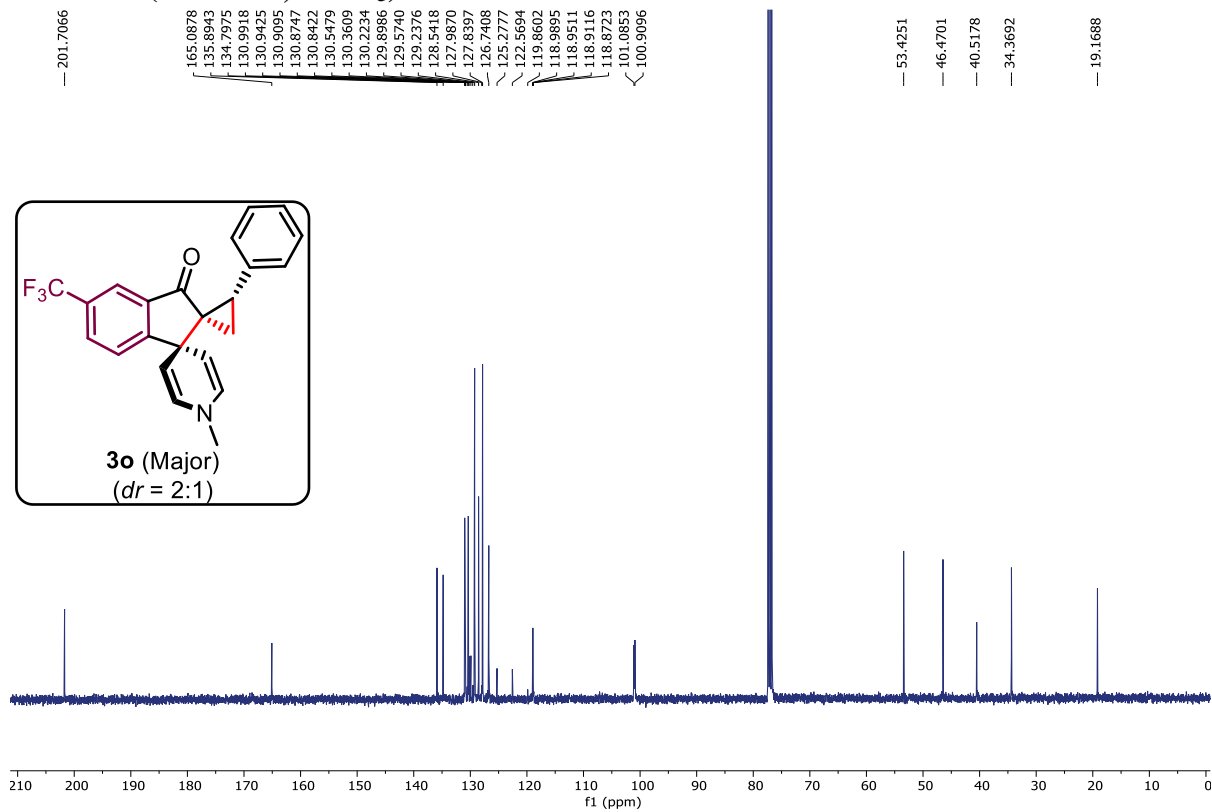
^{19}F NMR (376 MHz, CDCl_3)



¹H NMR (400 MHz, CDCl₃)

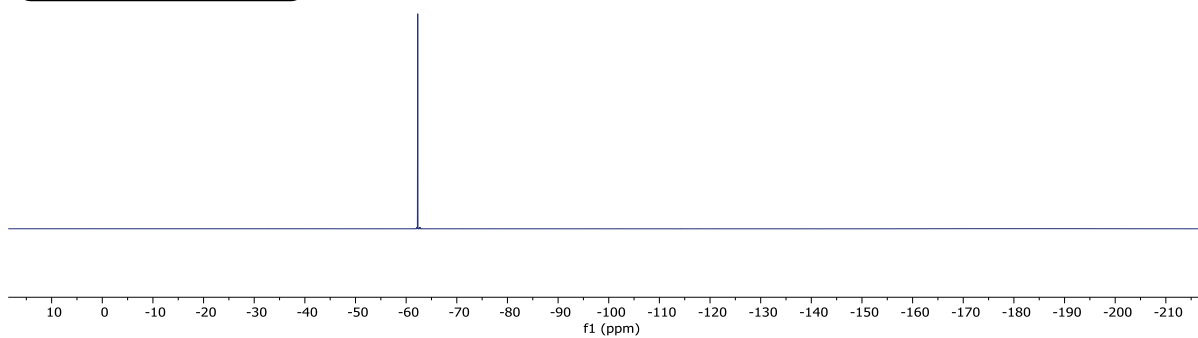
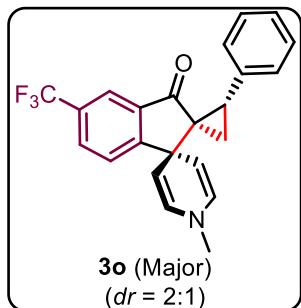


¹³C NMR (100 MHz, CDCl₃)



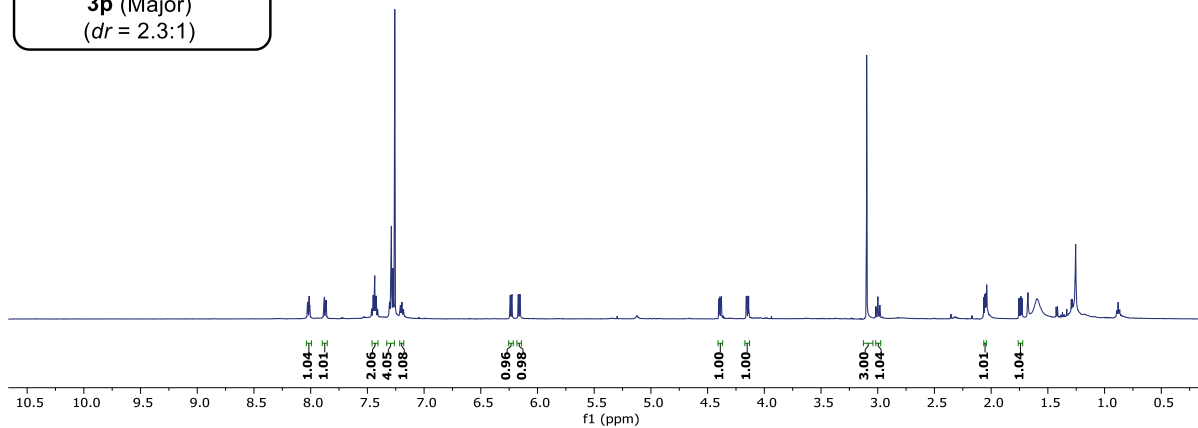
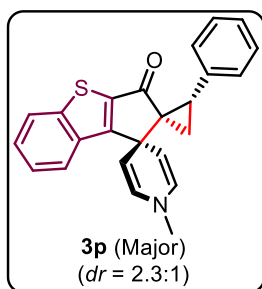
^{19}F NMR (376 MHz, CDCl_3)

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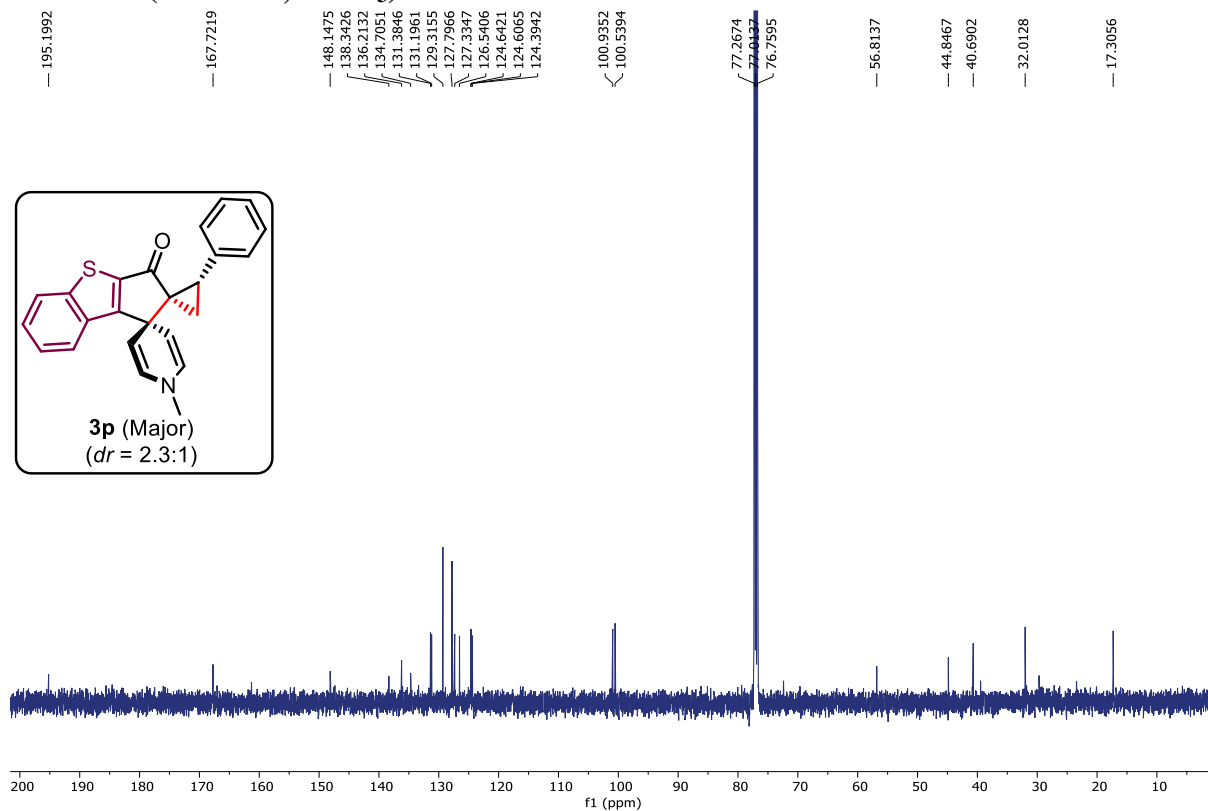


^1H NMR (500 MHz, CDCl_3)

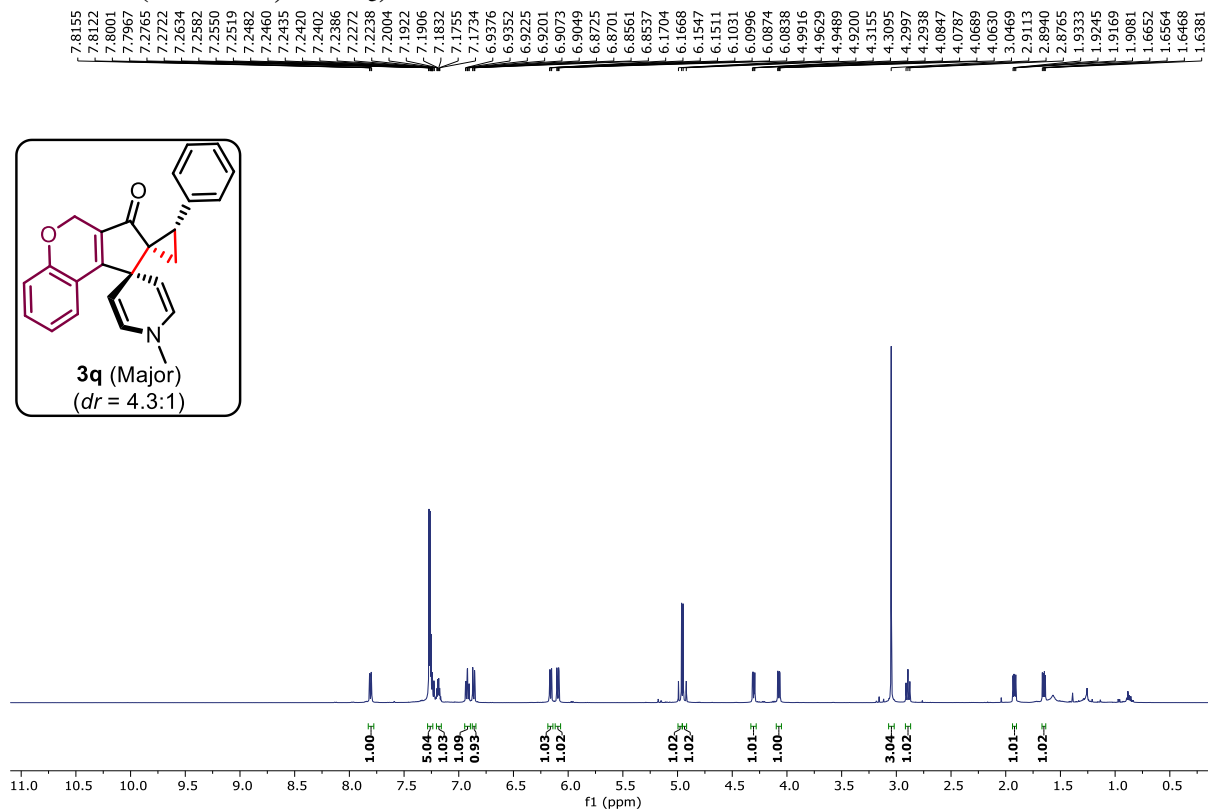
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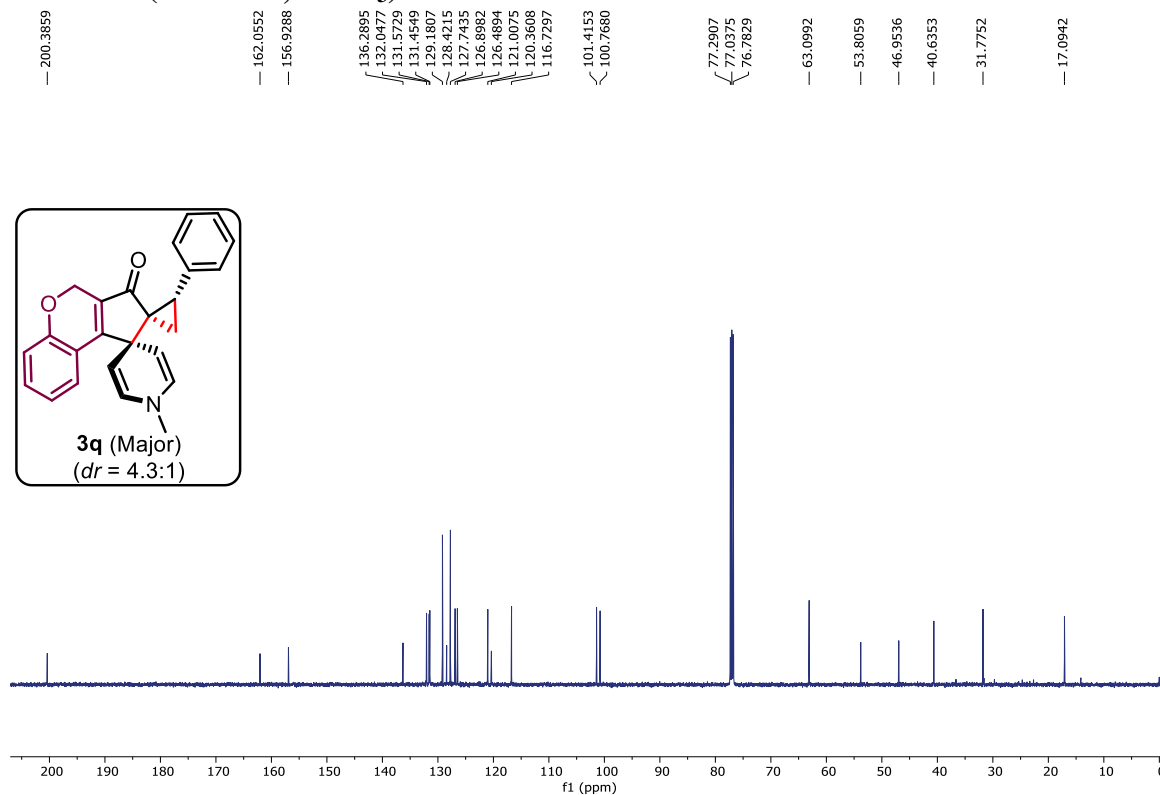
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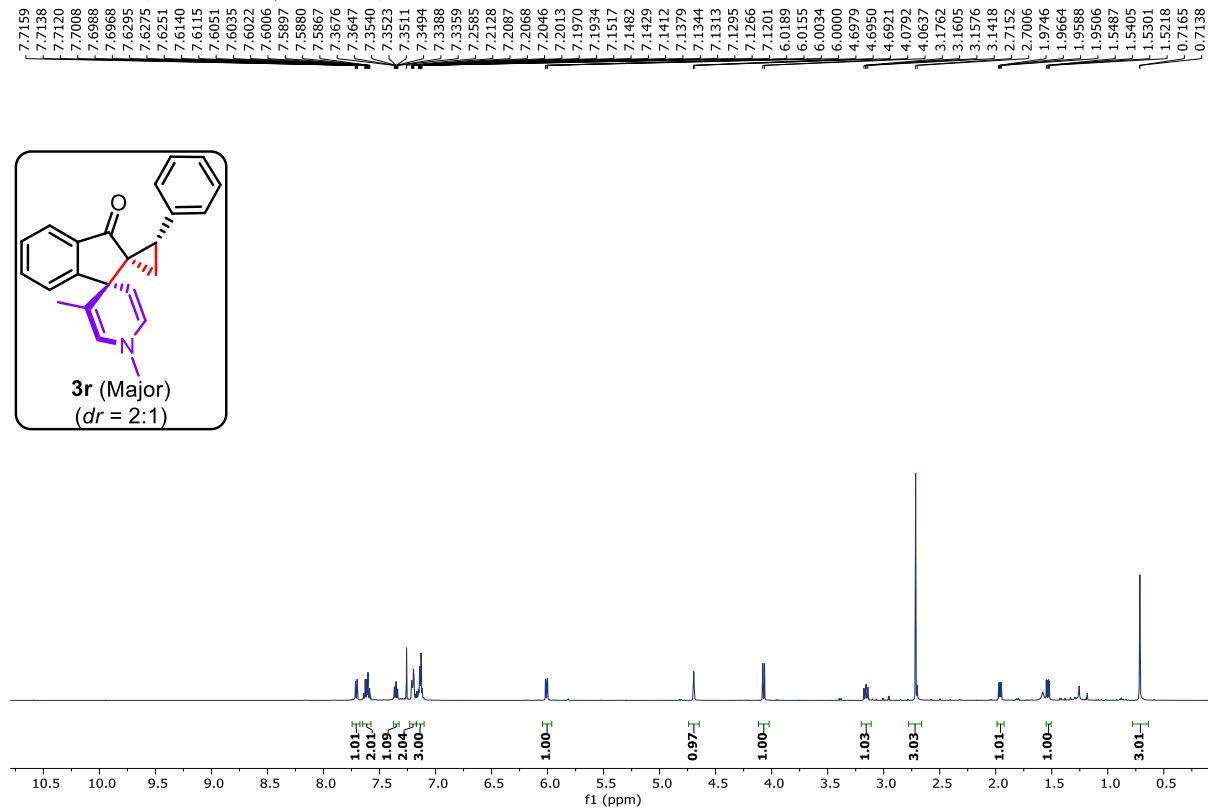
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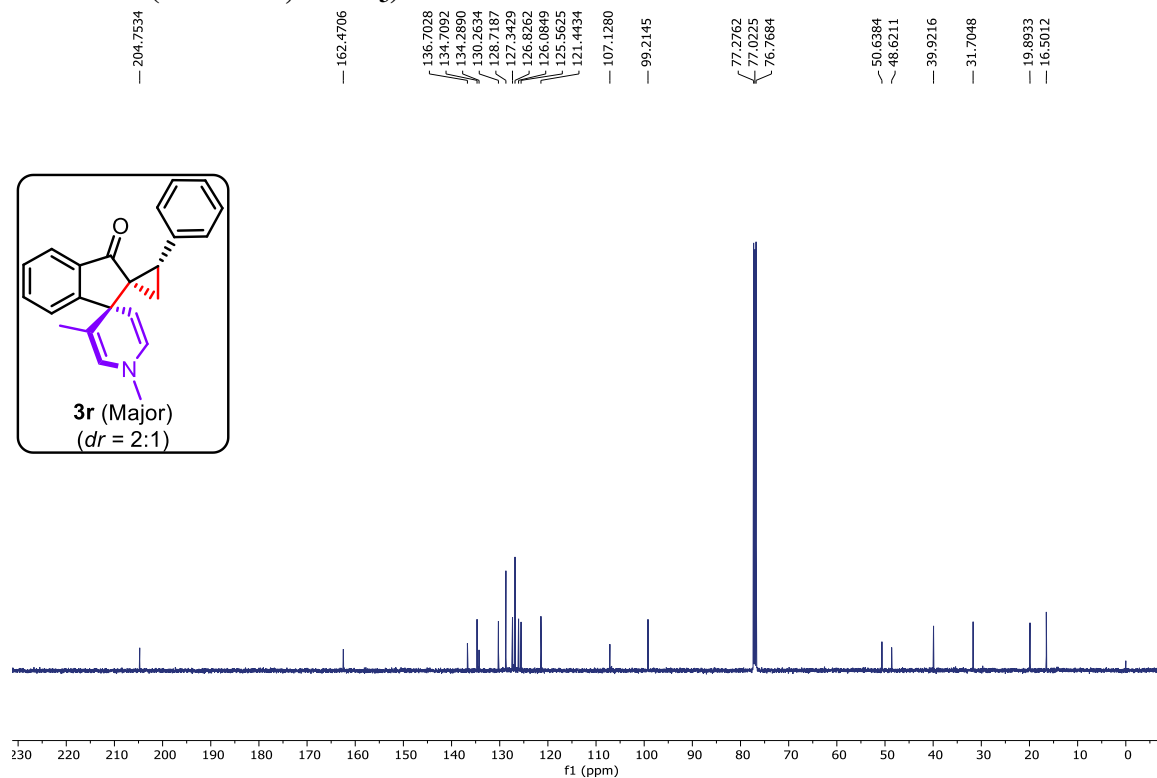
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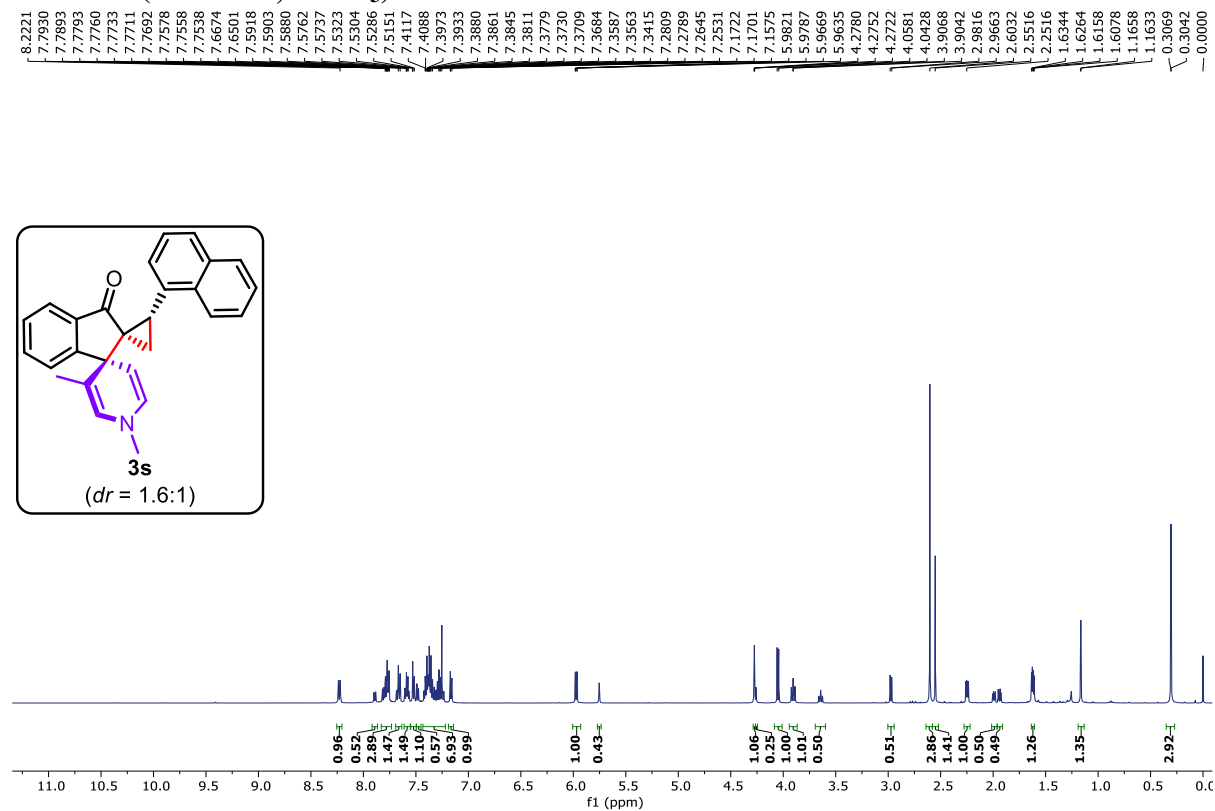
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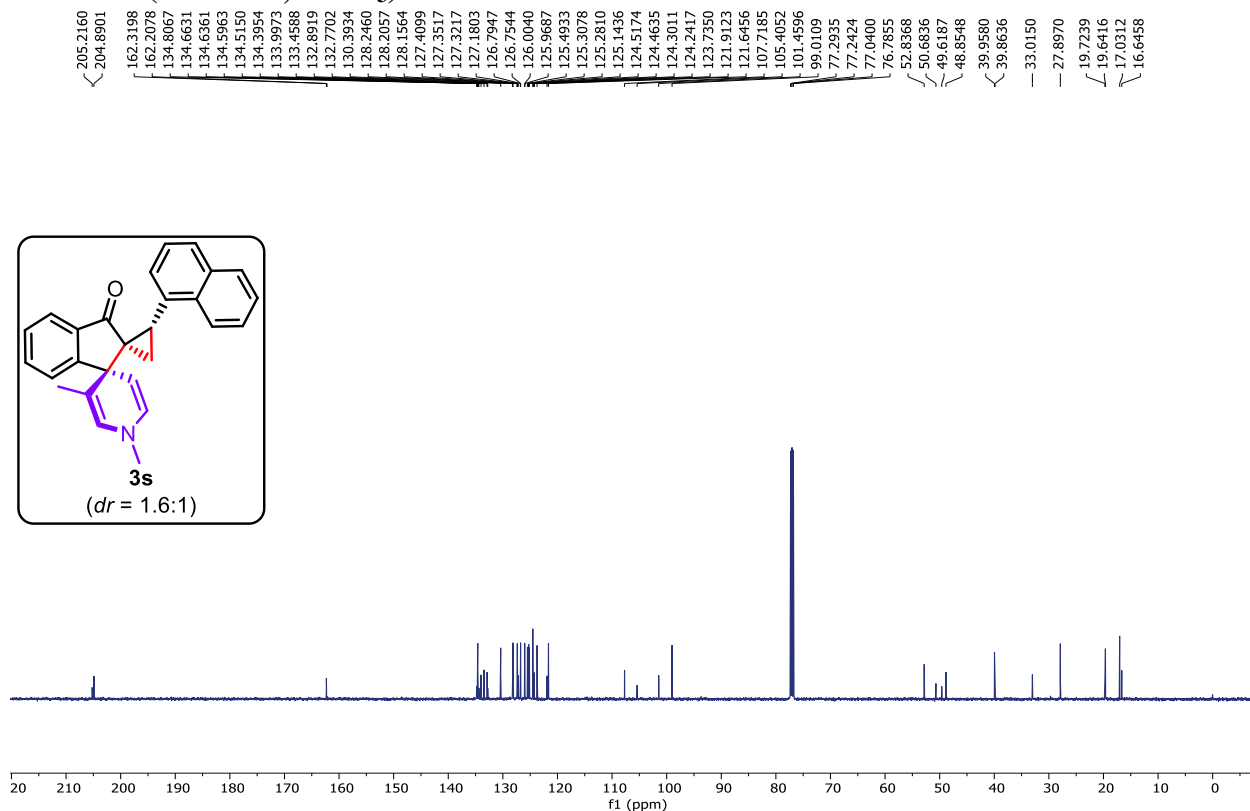
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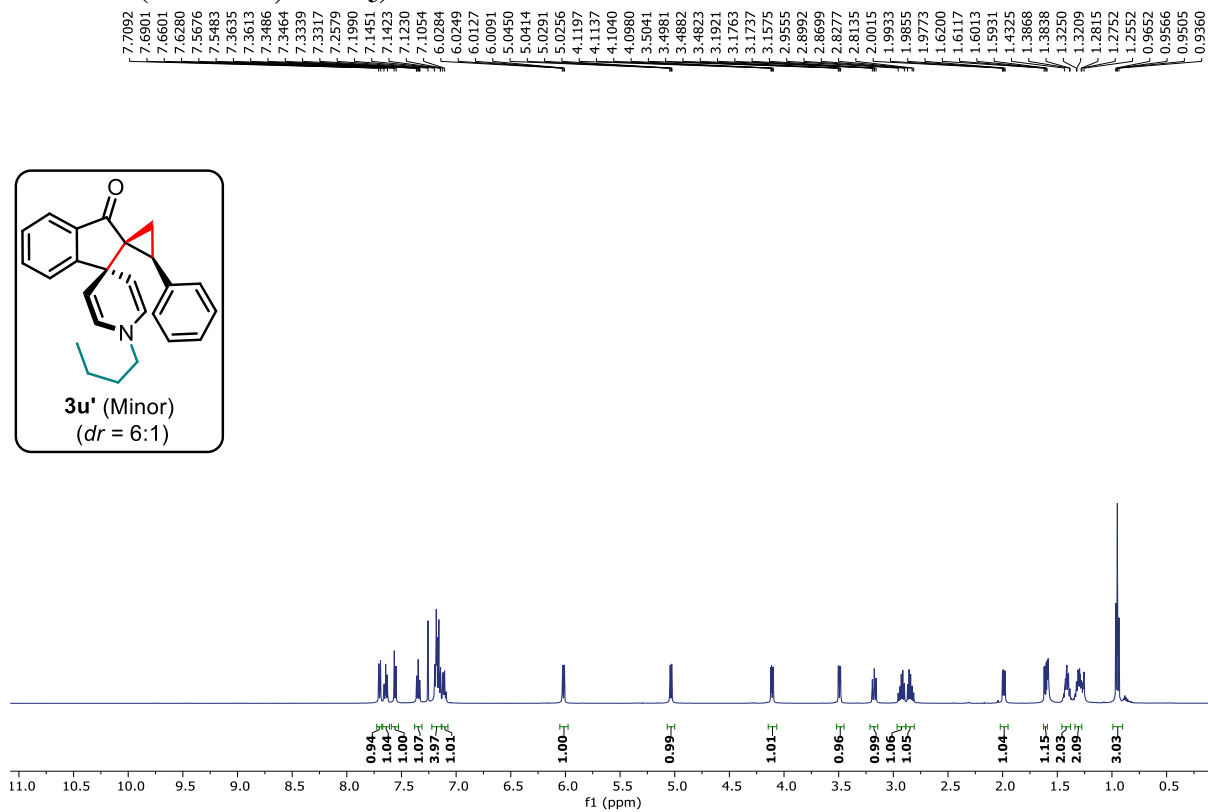
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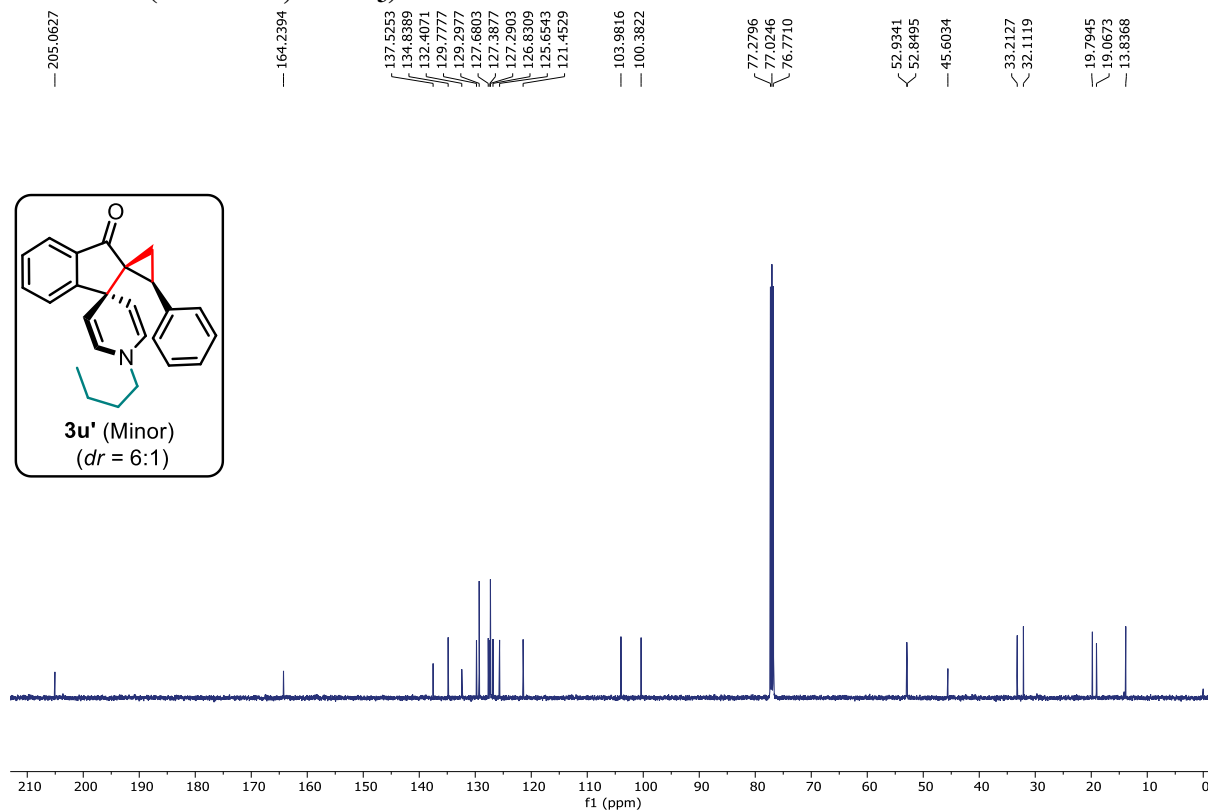
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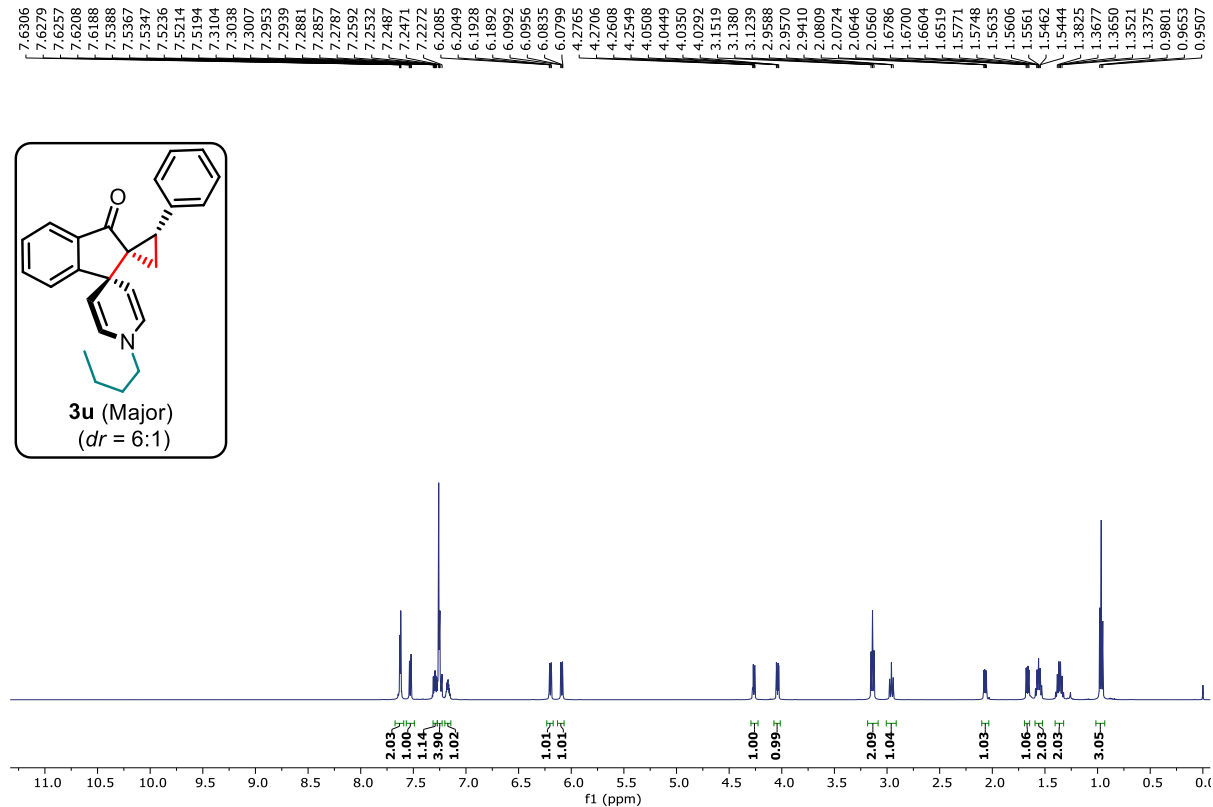
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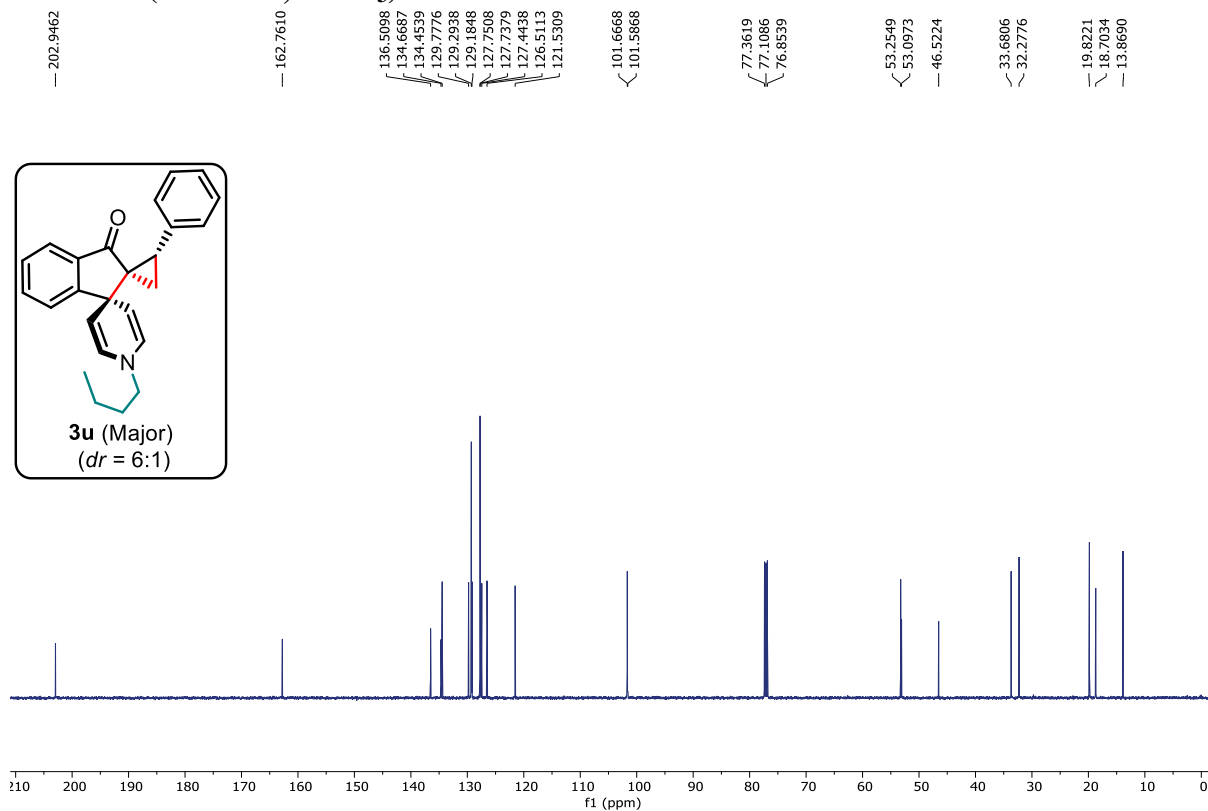
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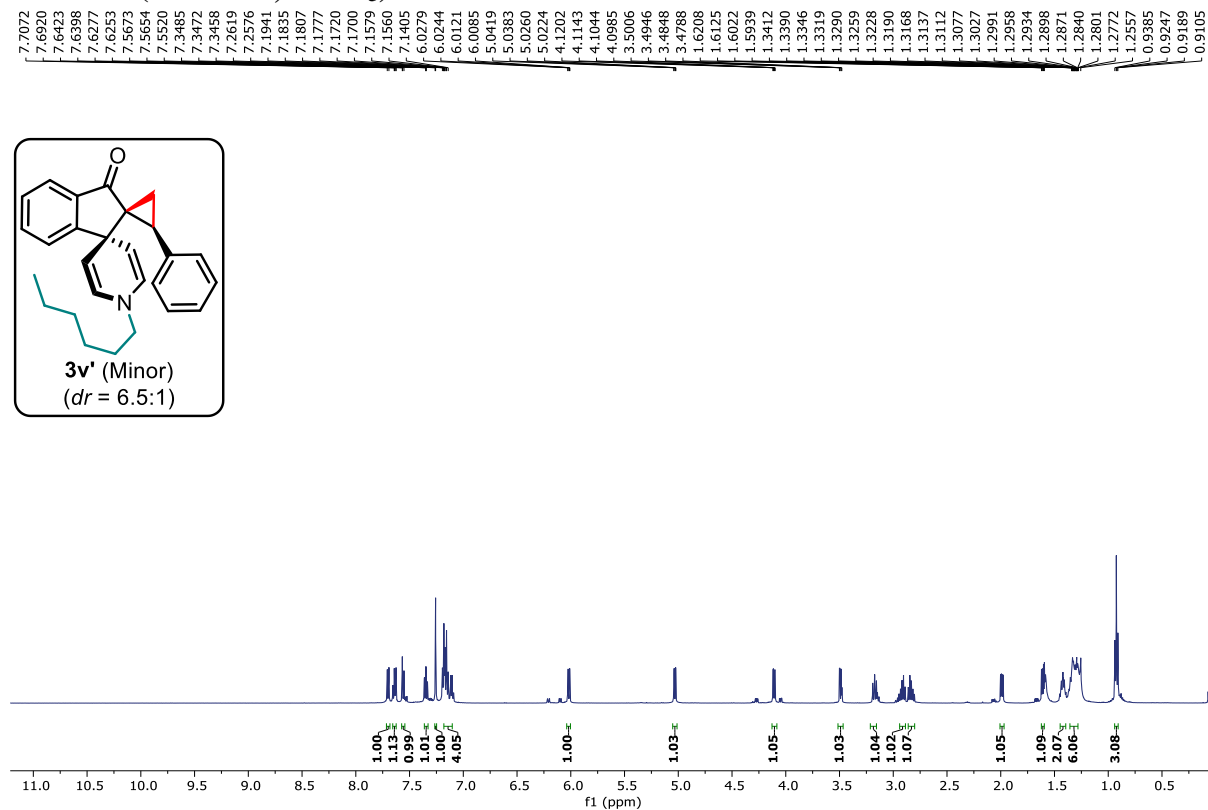
¹H NMR (500 MHz, CDCl₃)



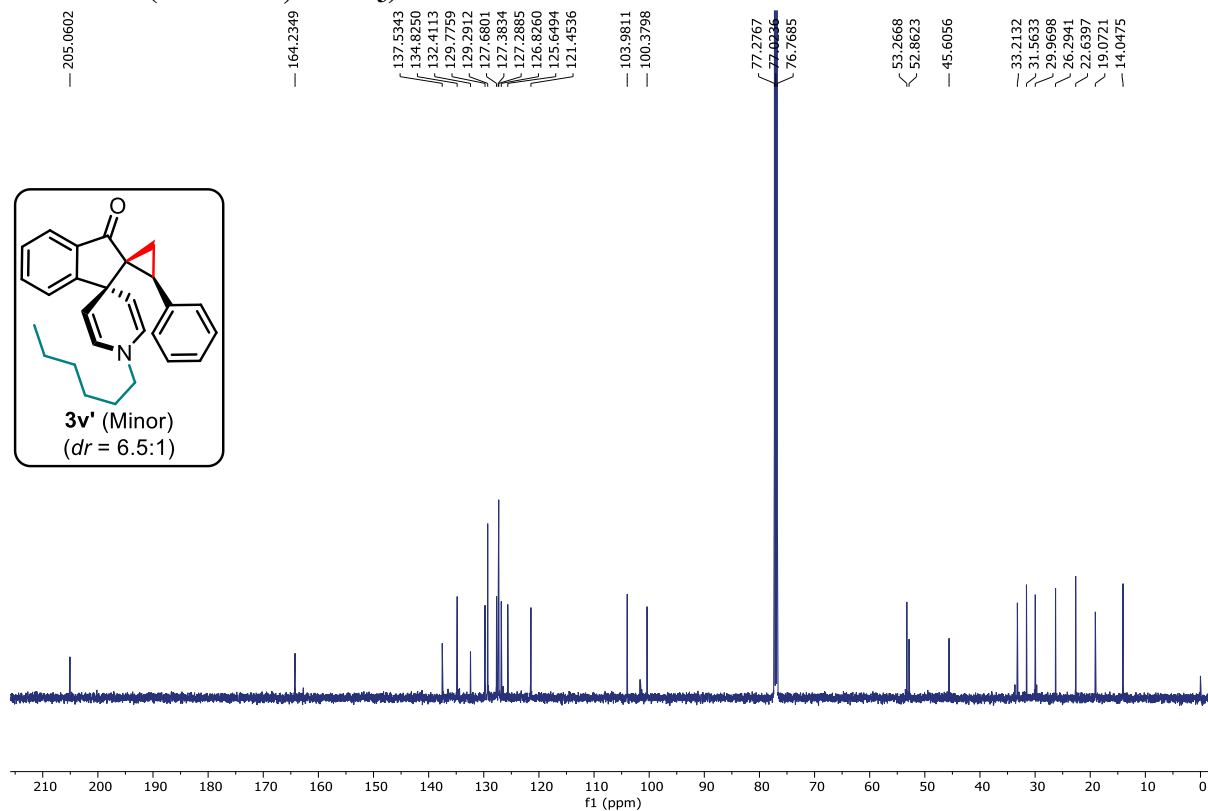
^{13}C NMR (125 MHz, CDCl_3)



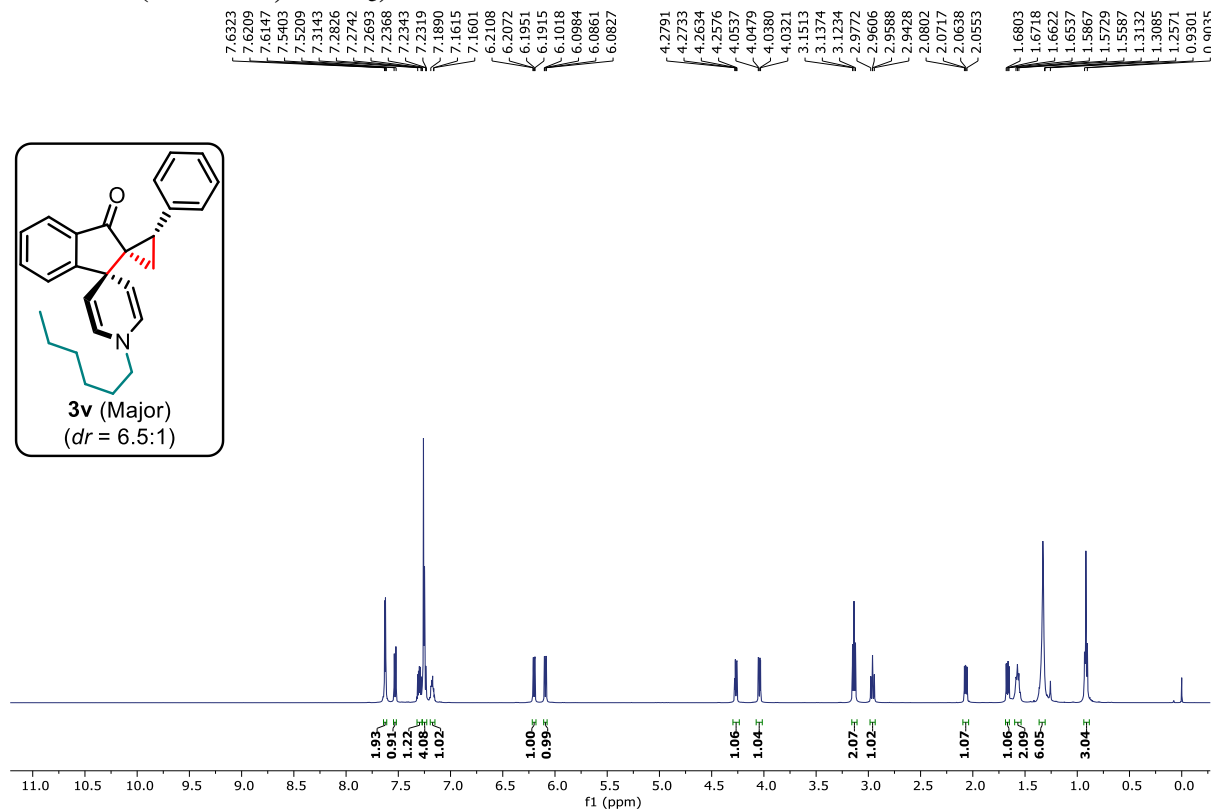
^1H NMR (500 MHz, CDCl_3)



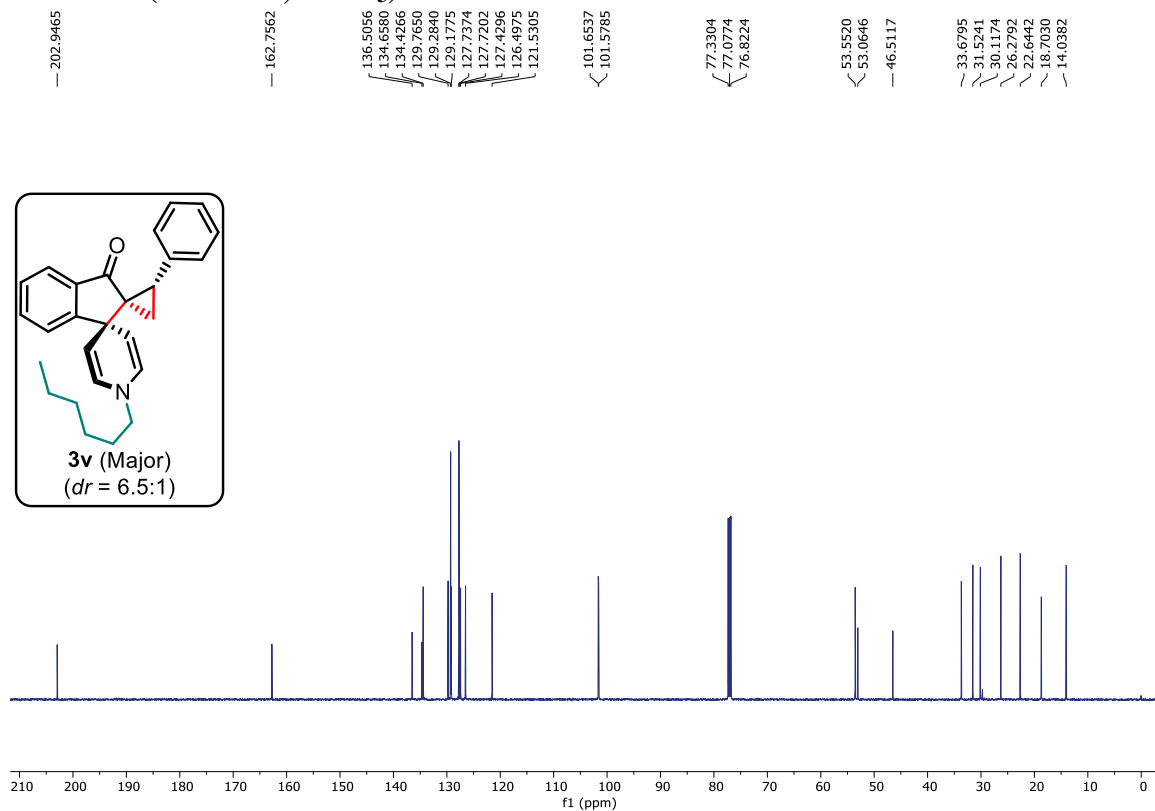
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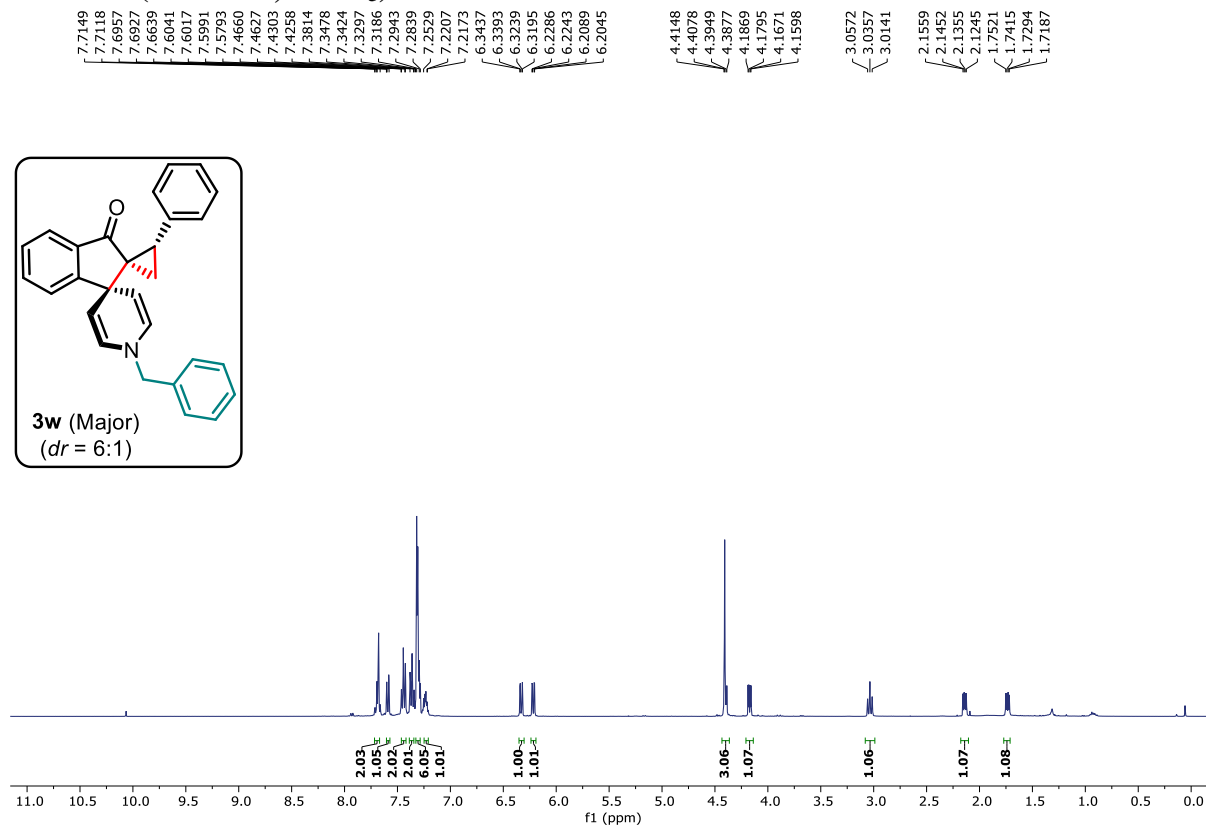
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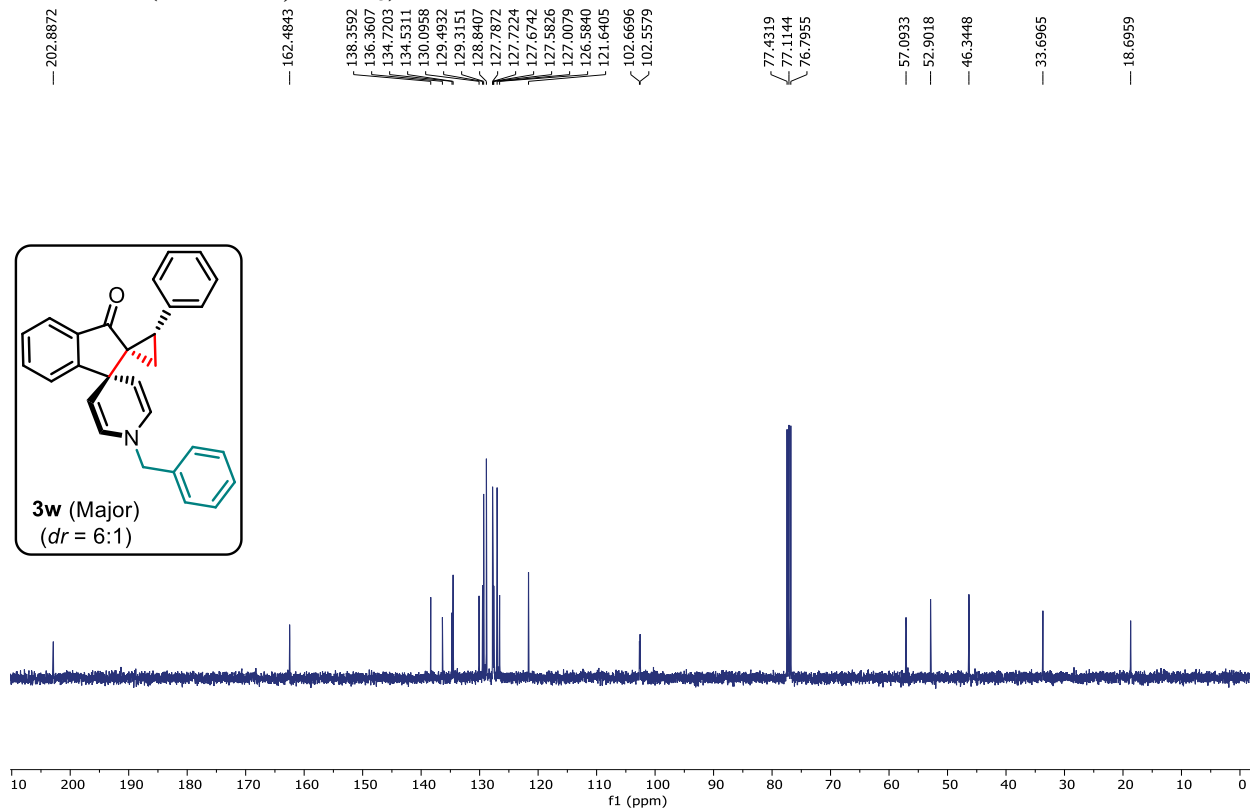
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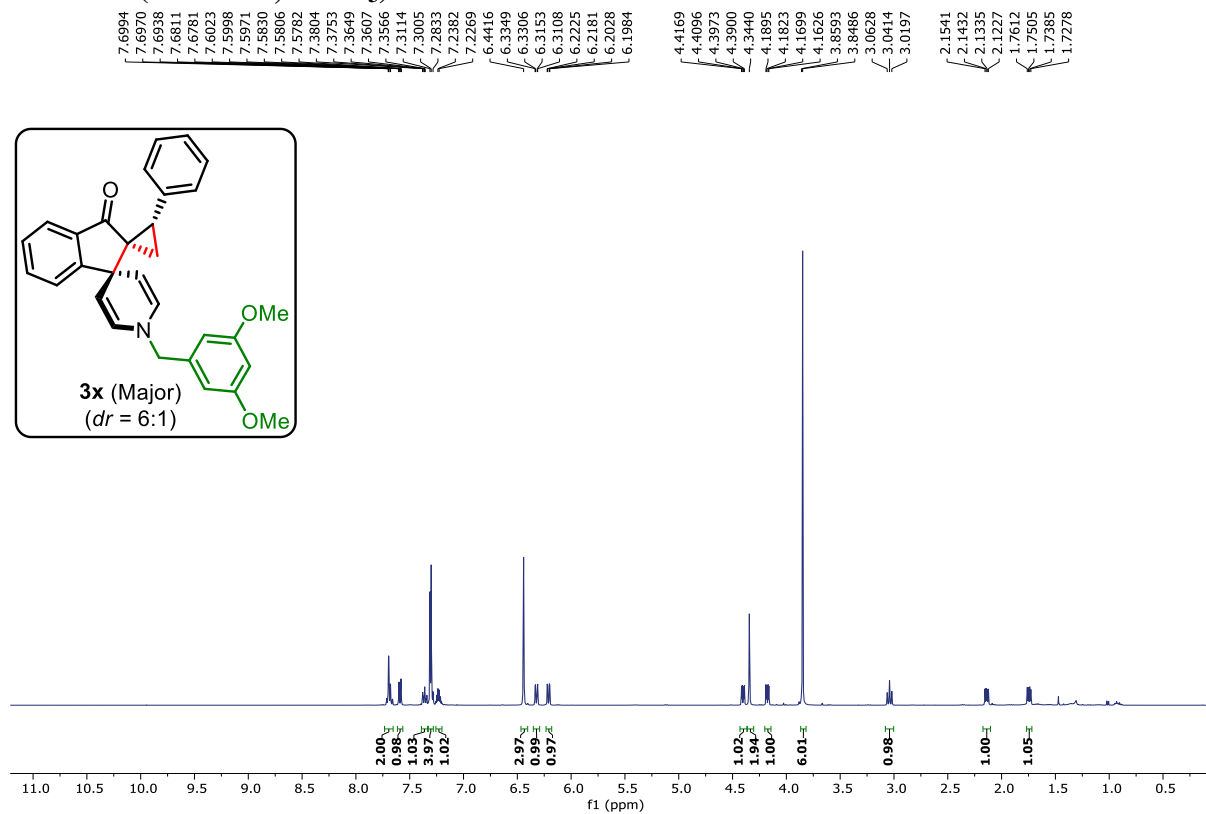
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^{13}C NMR (100 MHz, CDCl_3)

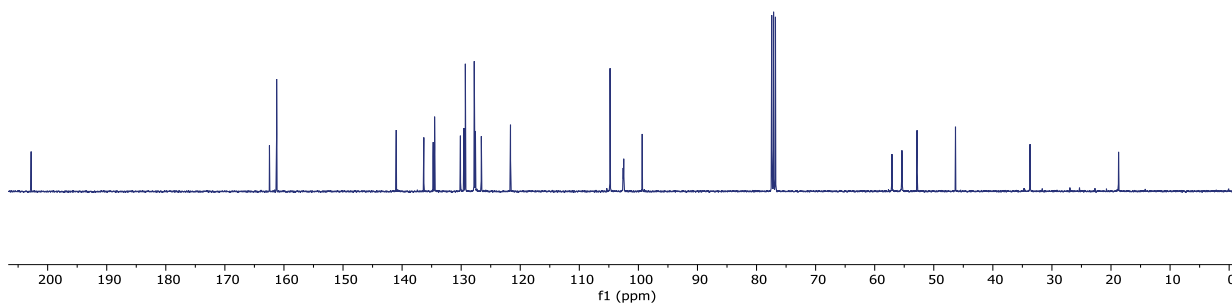
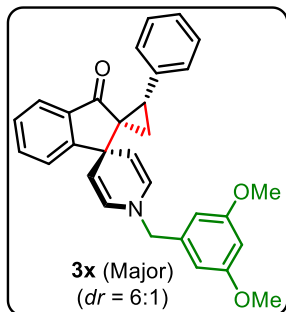


^1H NMR (400 MHz, CDCl_3)



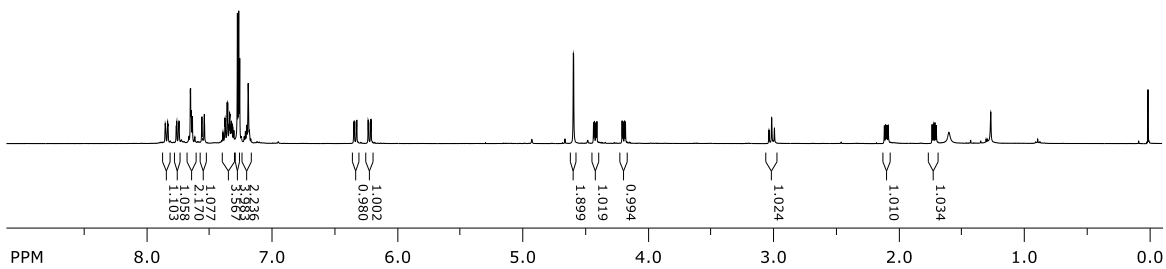
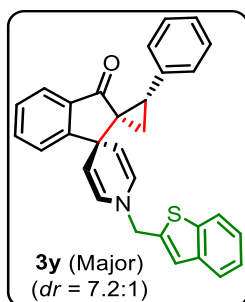
^{13}C NMR (100 MHz, CDCl_3)

202.7943, 162.4532, 161.2365, 140.9920, 136.3214, 134.7320, 134.4988, 130.1273, 129.5370, 129.2927, 127.7872, 127.7236, 127.5865, 126.5879, 121.6507, 104.7931, 102.5638, 102.4845, 99.3546, 77.4335, 77.1163, 76.7980, 57.0761, 55.3886, 55.3622, 52.8287, 46.3234, 33.7040, 18.7175

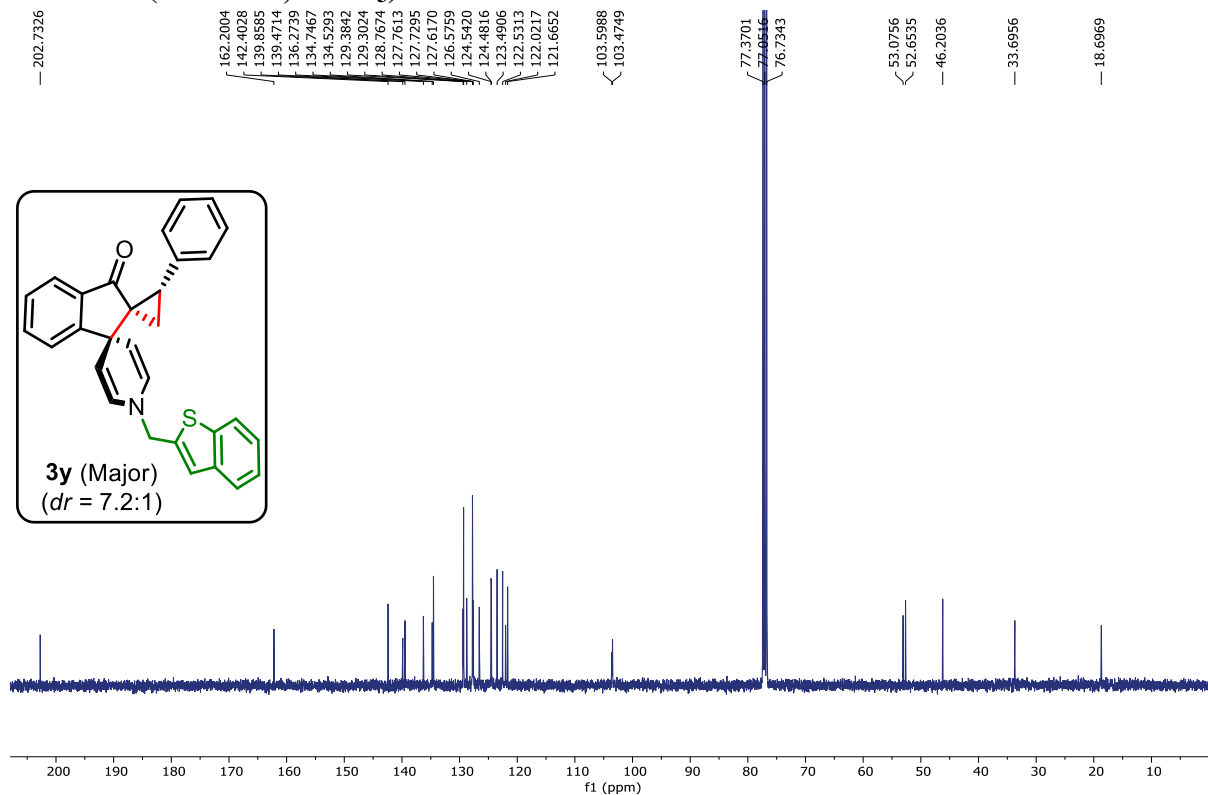


^1H NMR (400 MHz, CDCl_3)

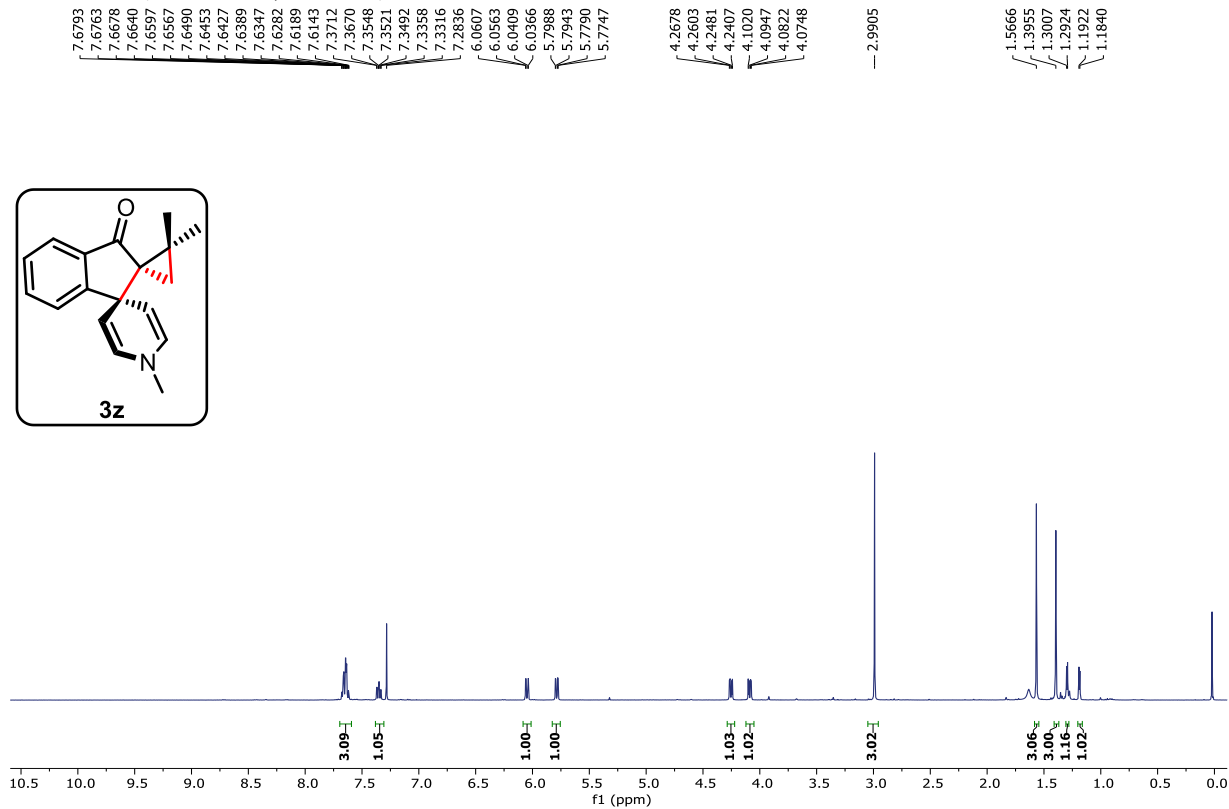
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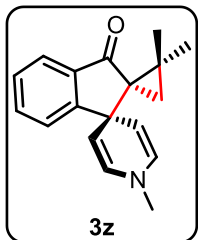
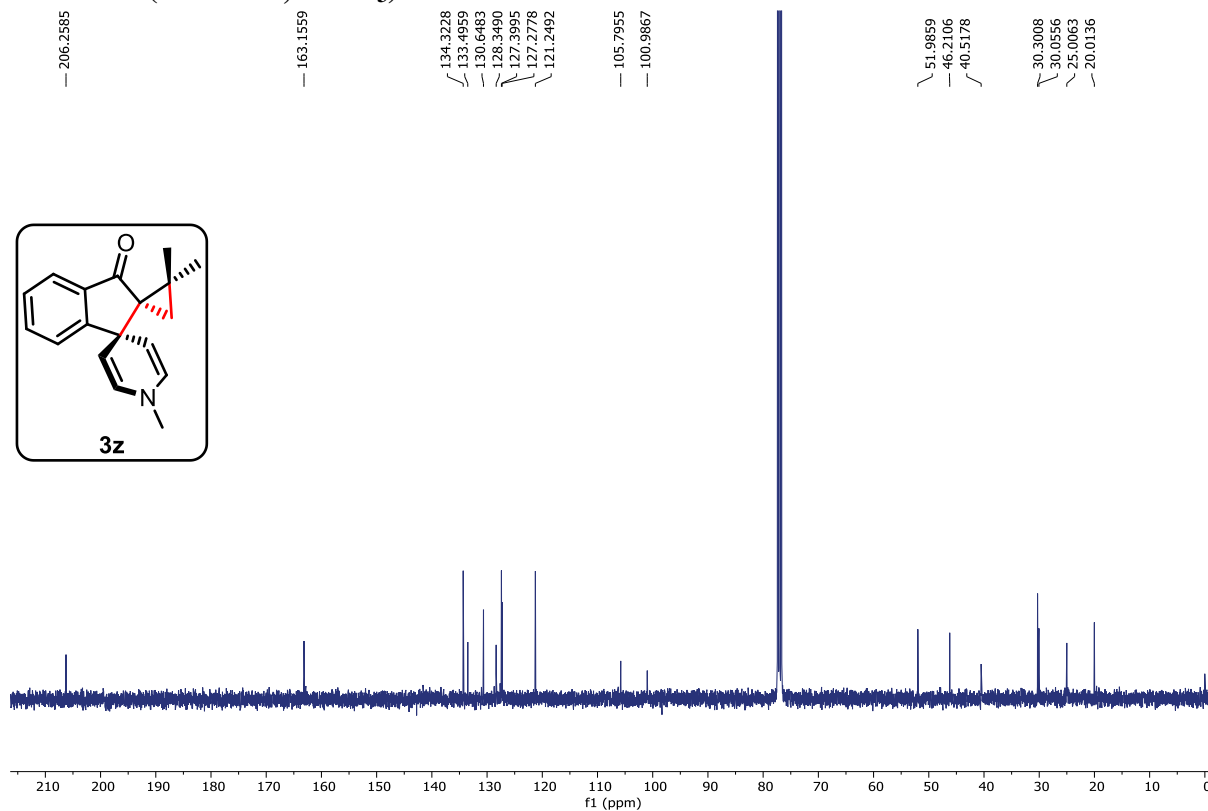
¹³C NMR (100 MHz, CDCl₃)



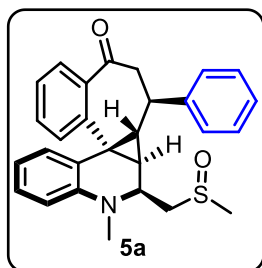
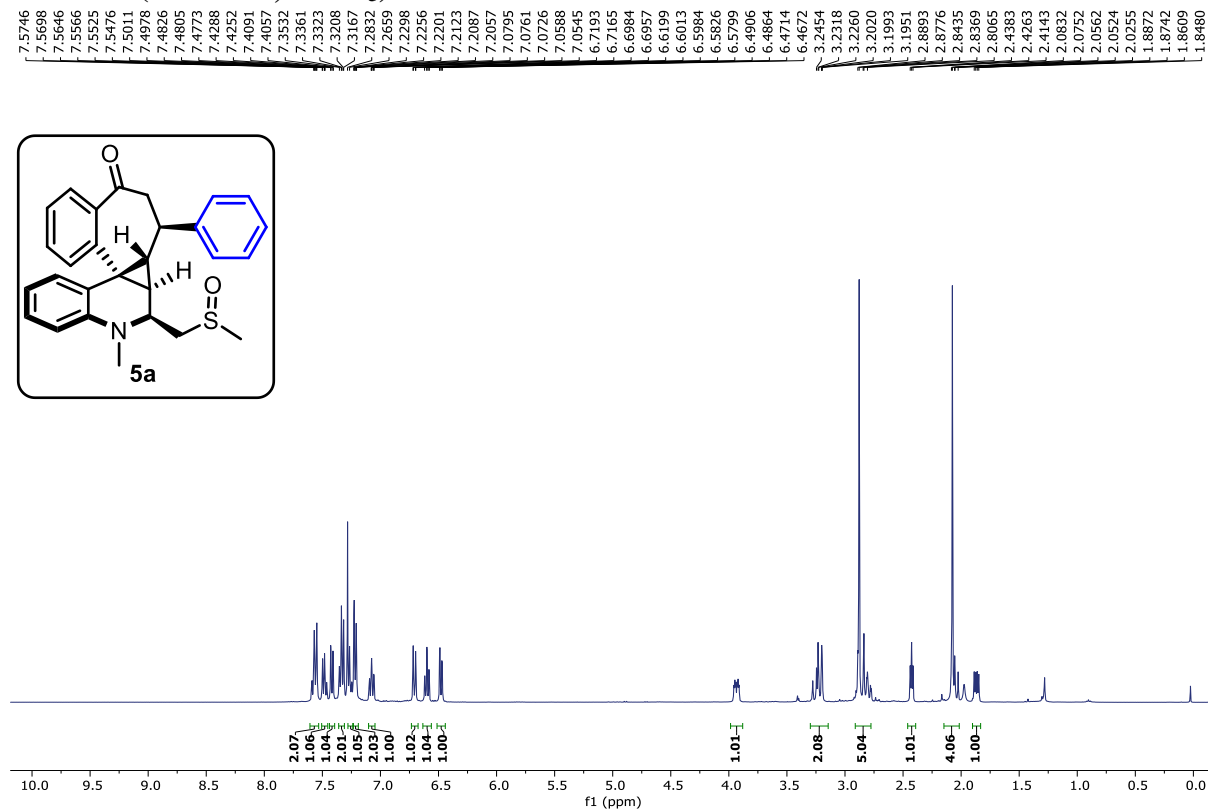
¹H NMR (400 MHz, CDCl₃)



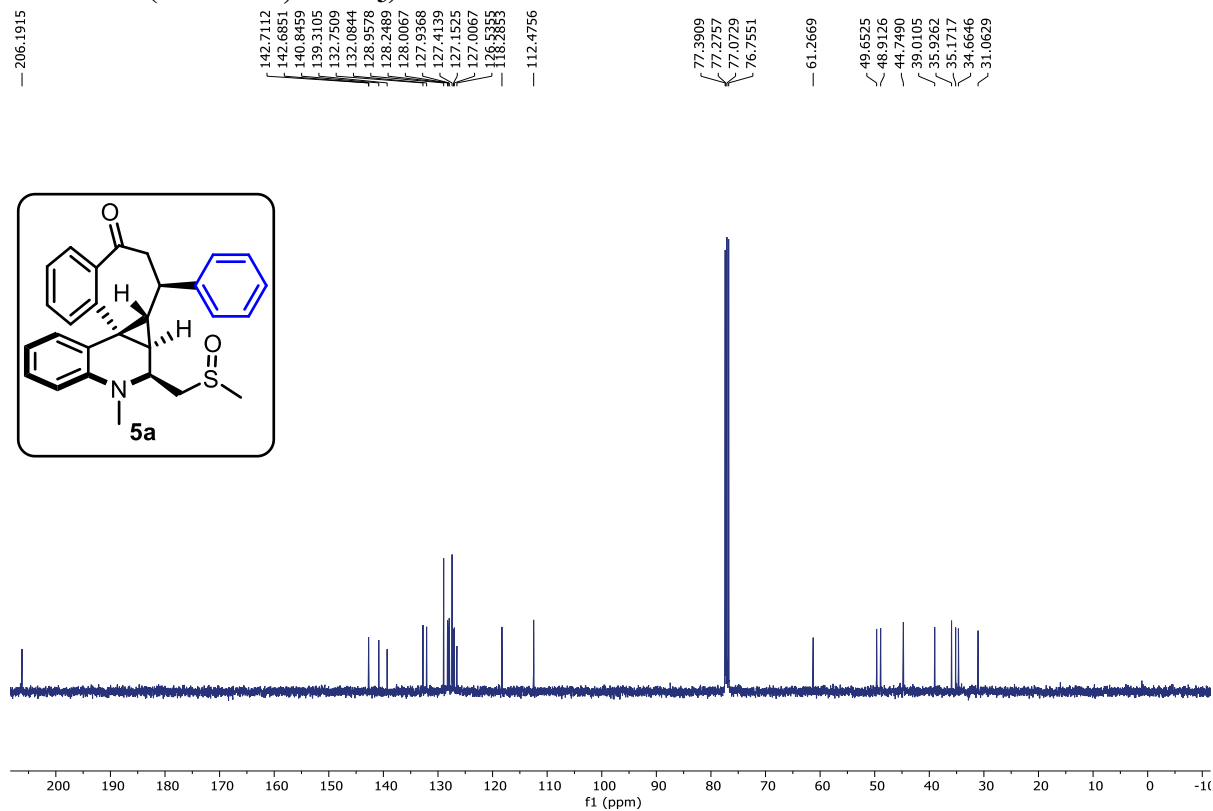
¹³C NMR (100 MHz, CDCl₃)



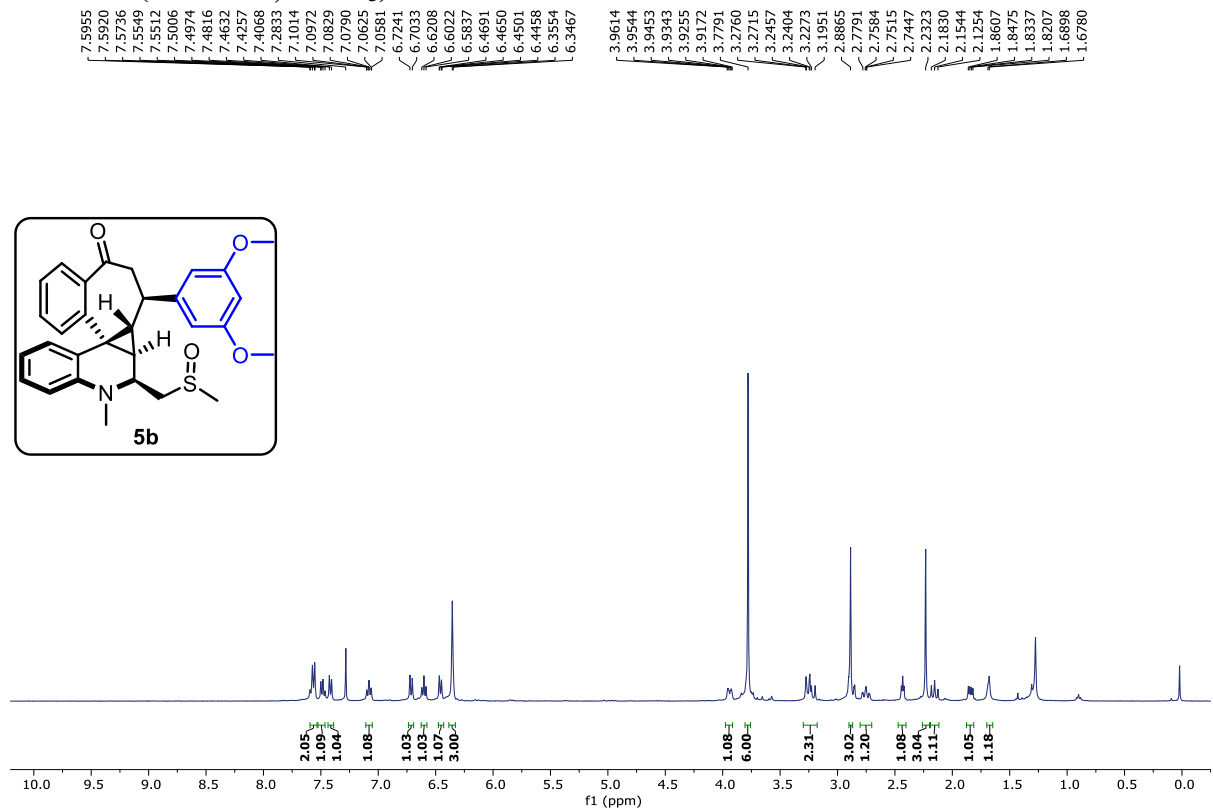
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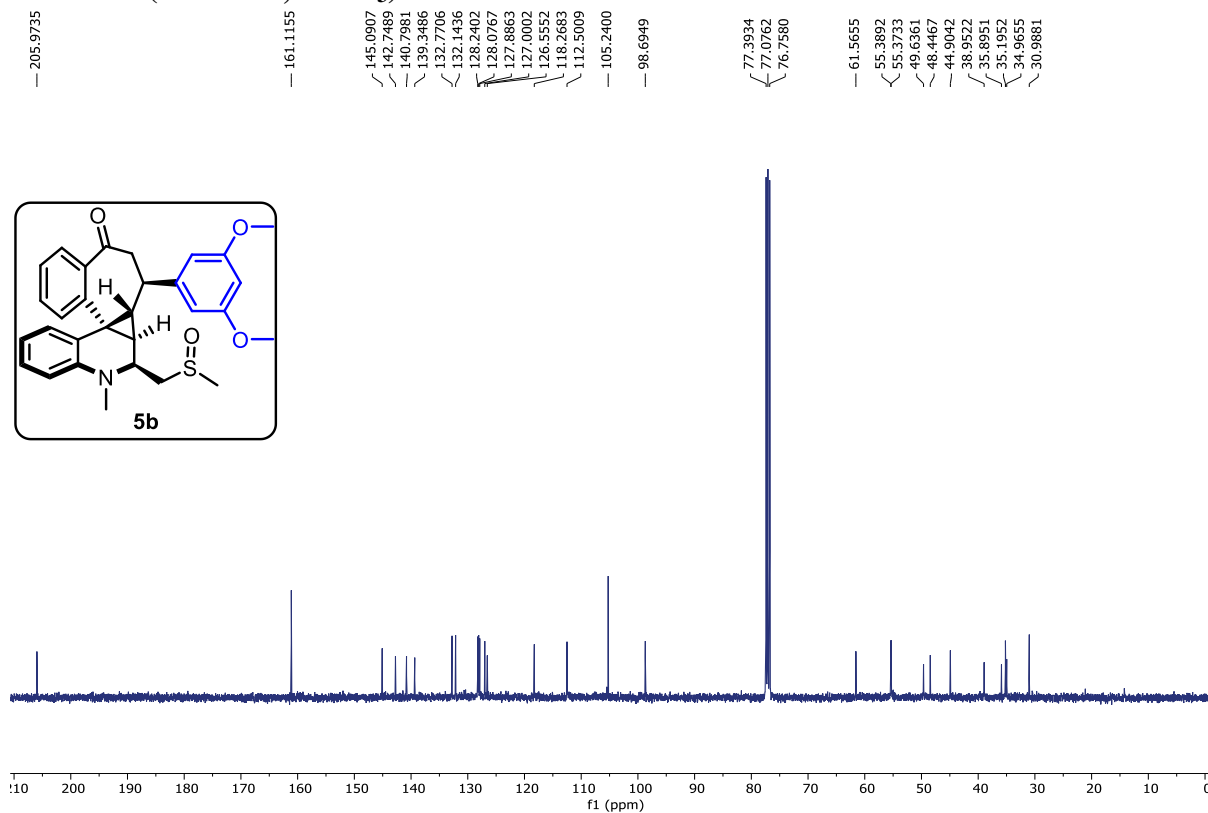
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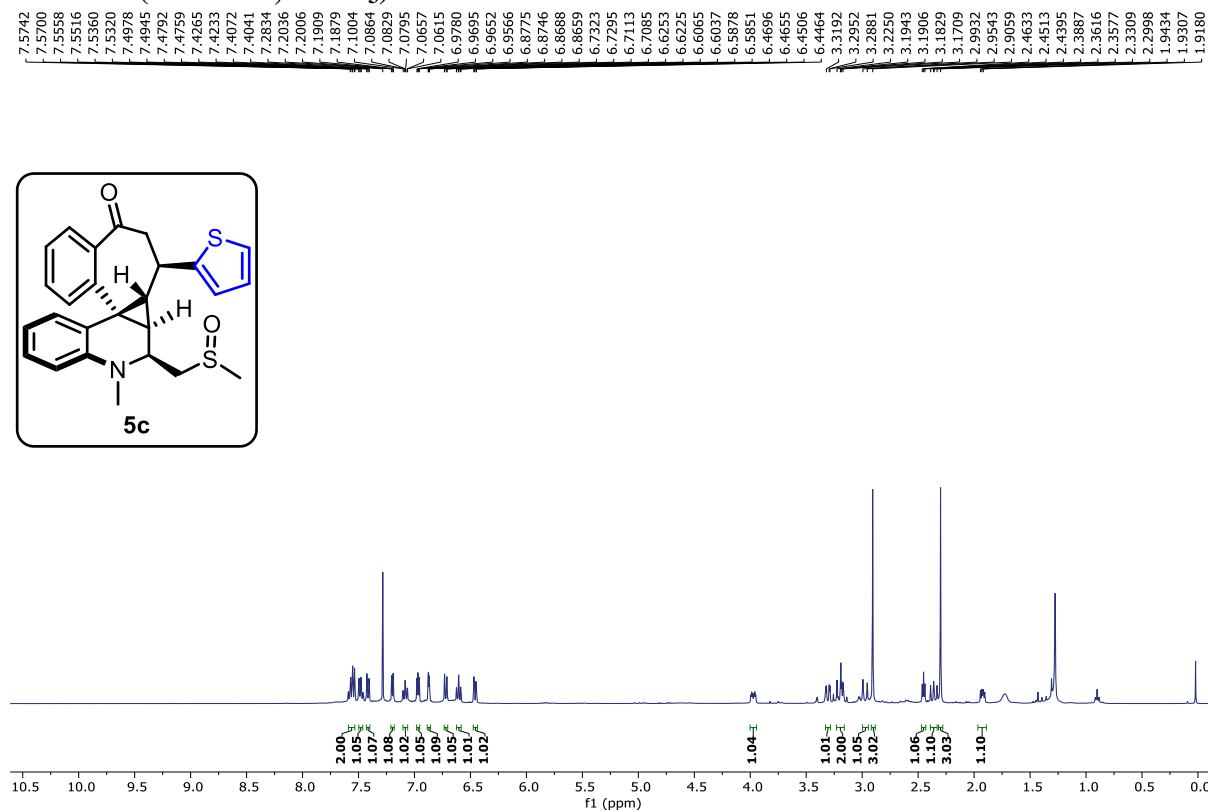
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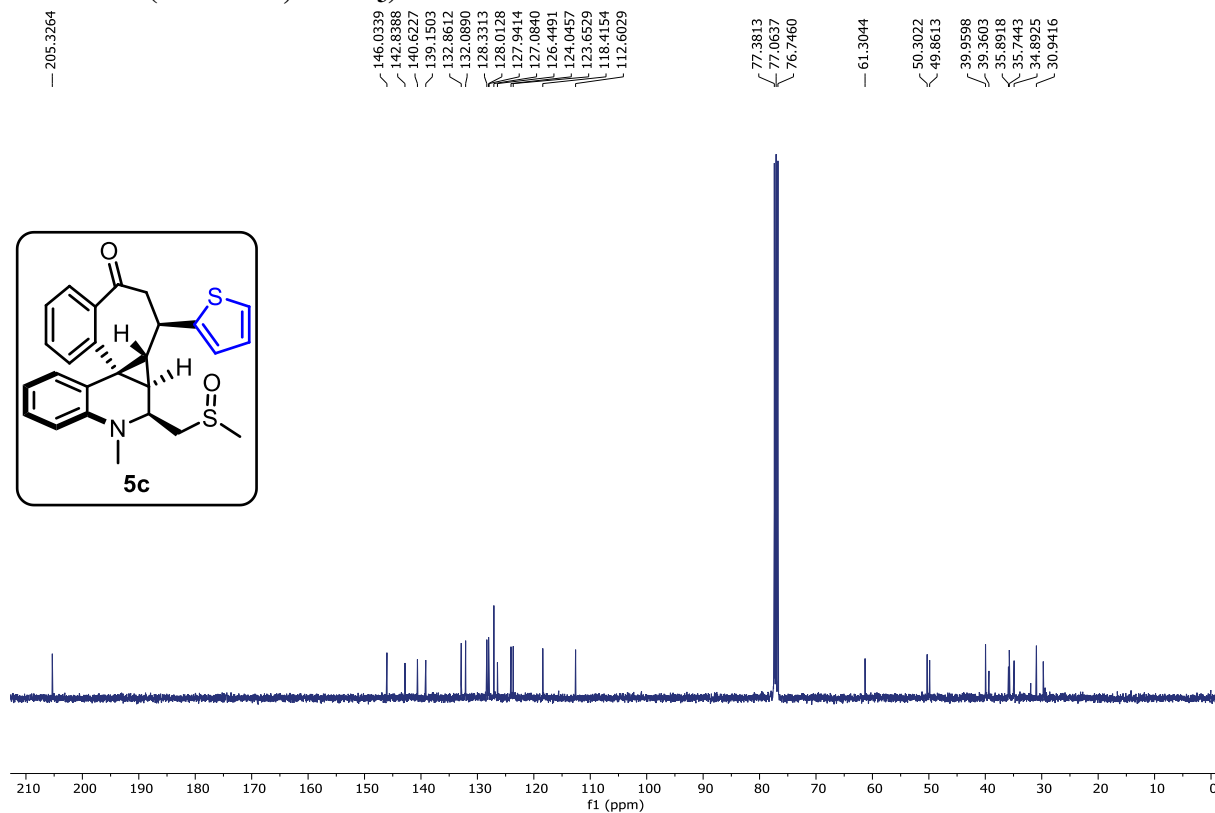
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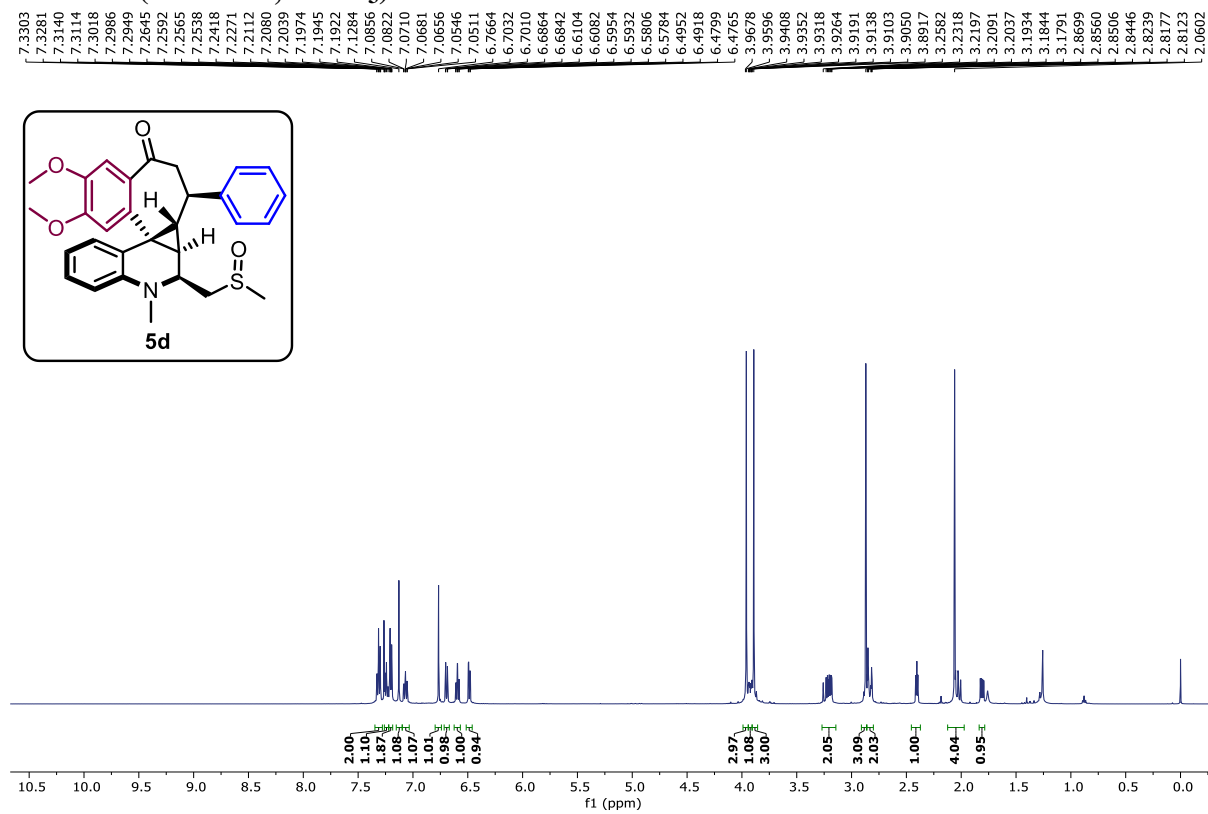
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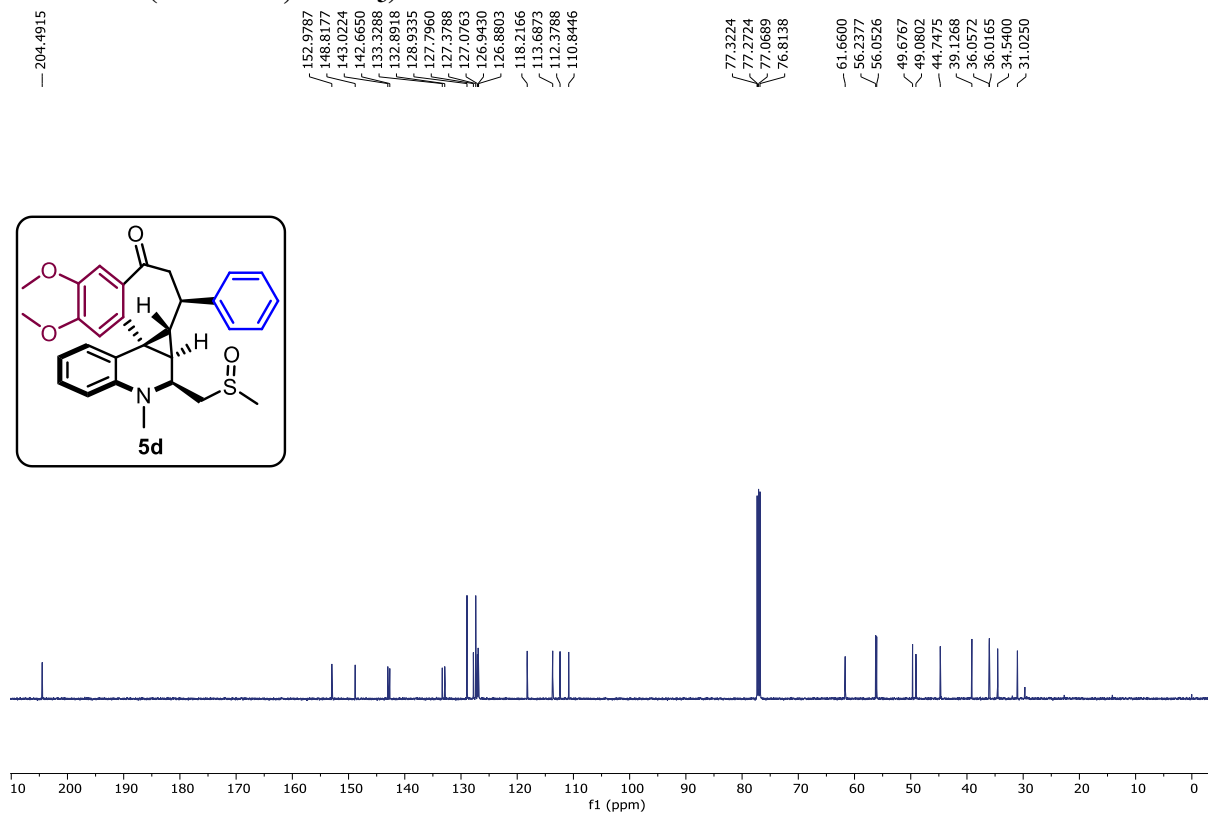
¹³C NMR (100 MHz, CDCl₃)



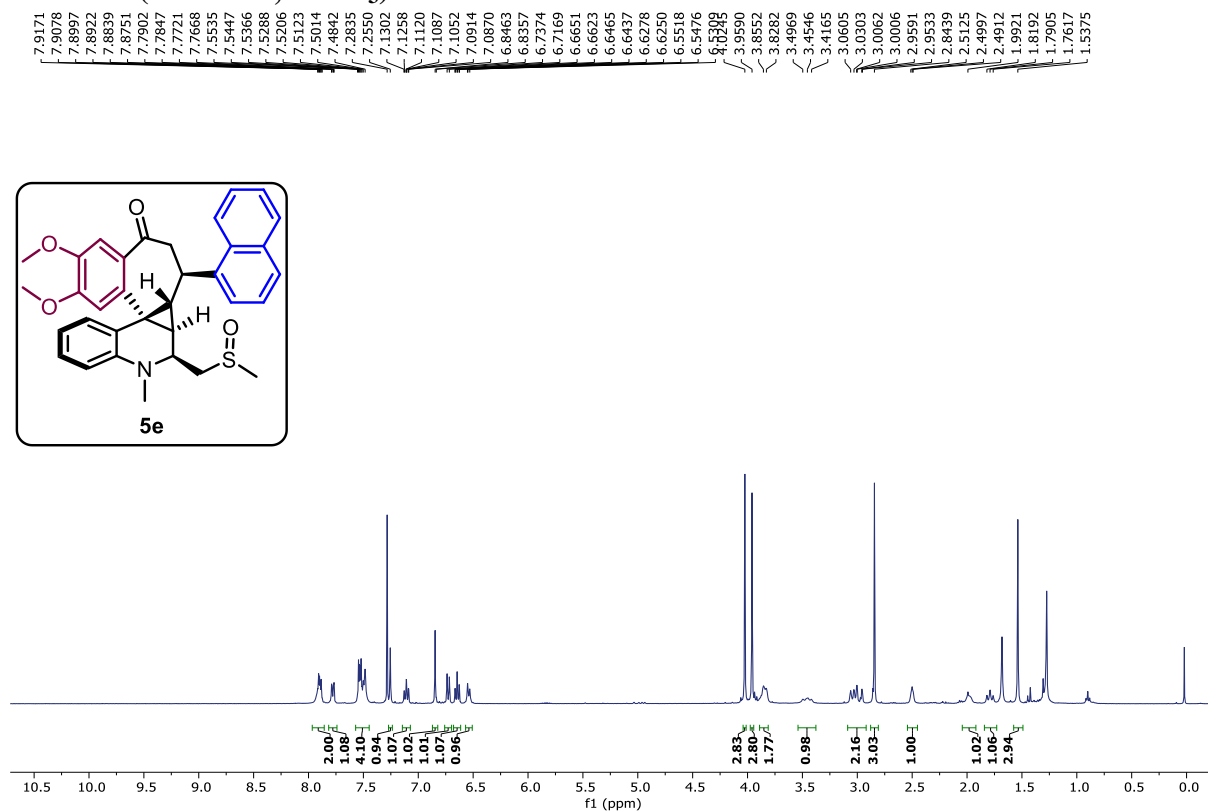
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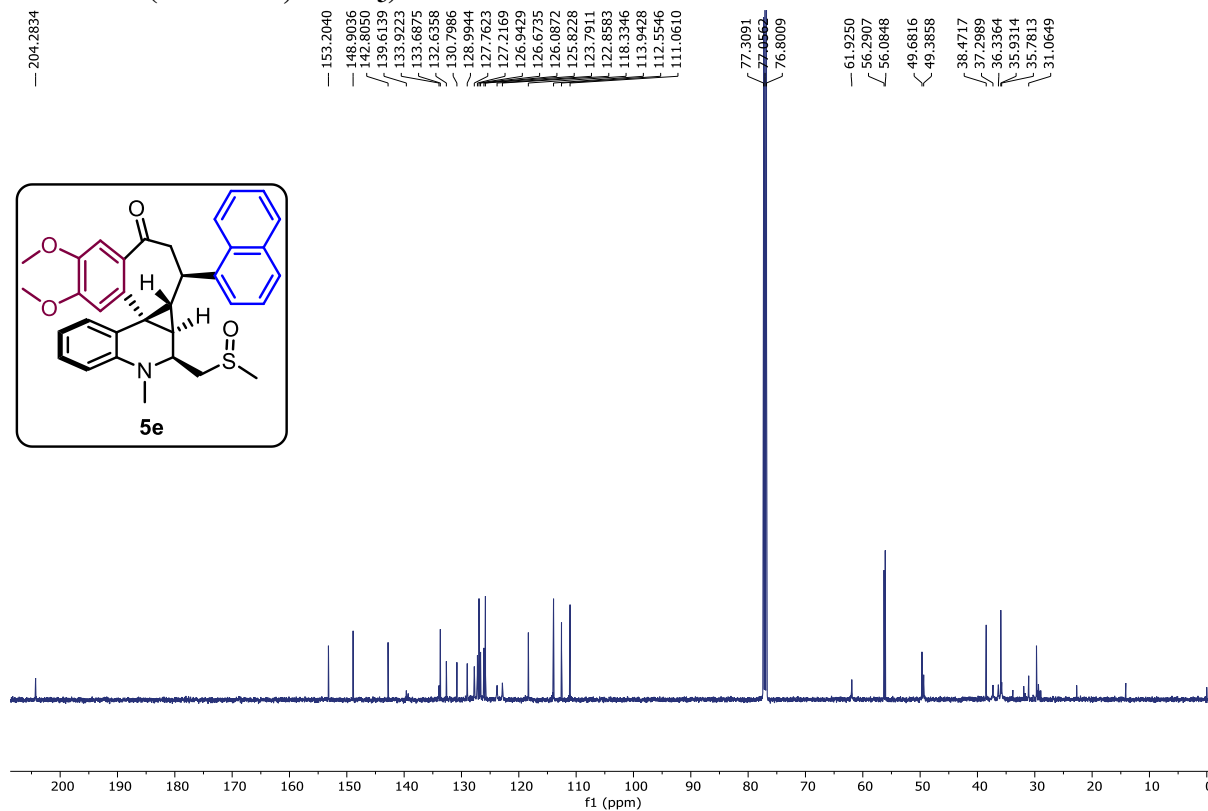
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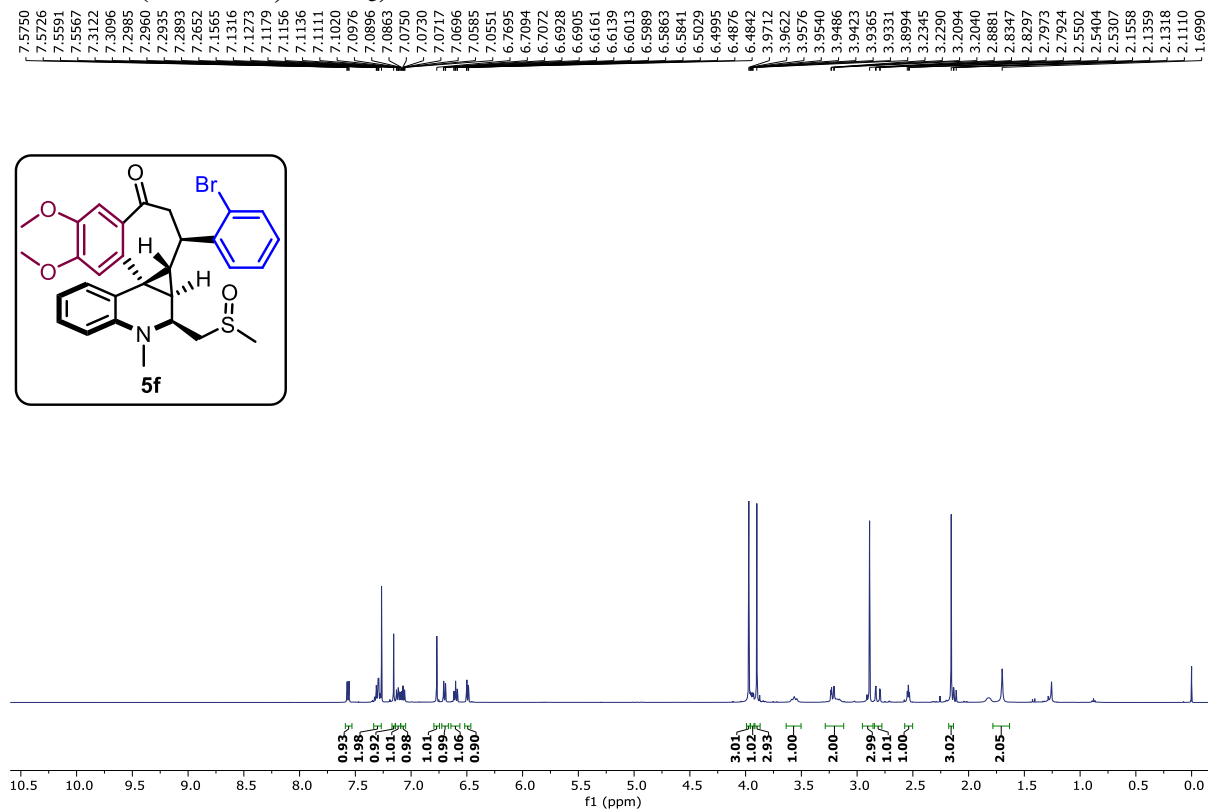
^1H NMR (400 MHz, CDCl_3)



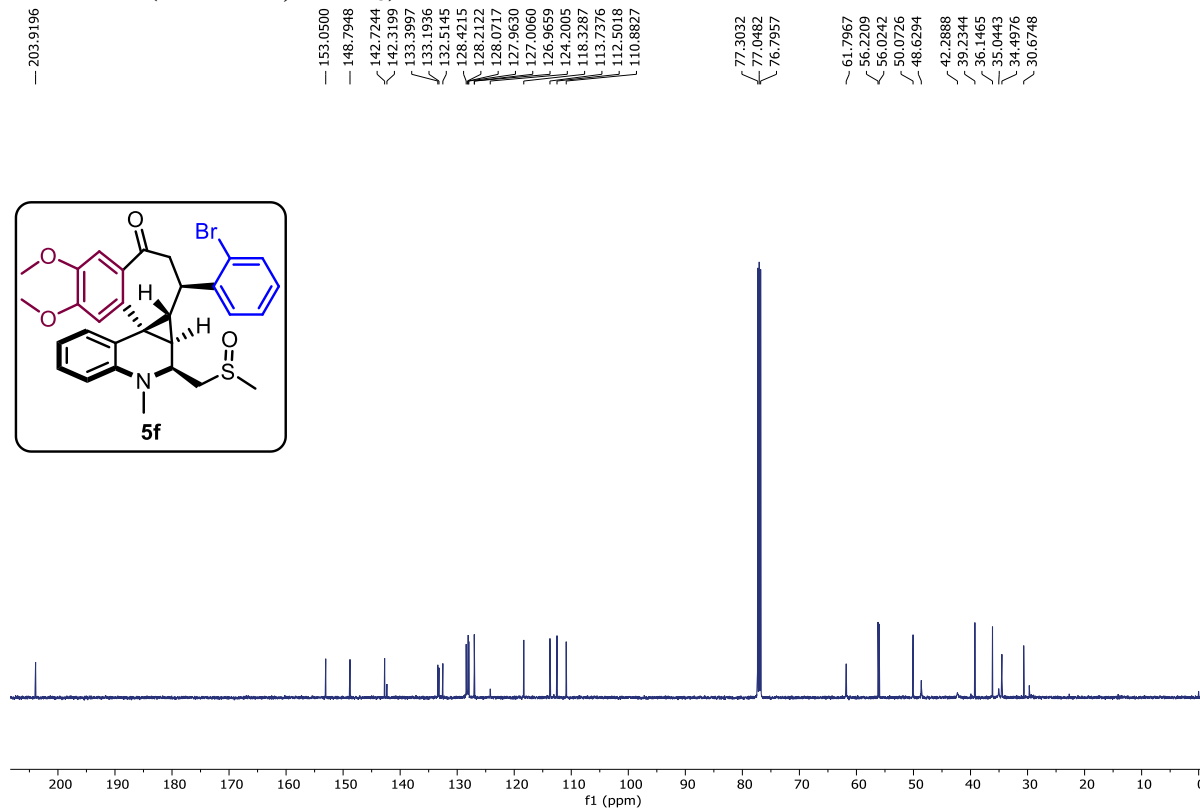
^{13}C NMR (125 MHz, CDCl_3)



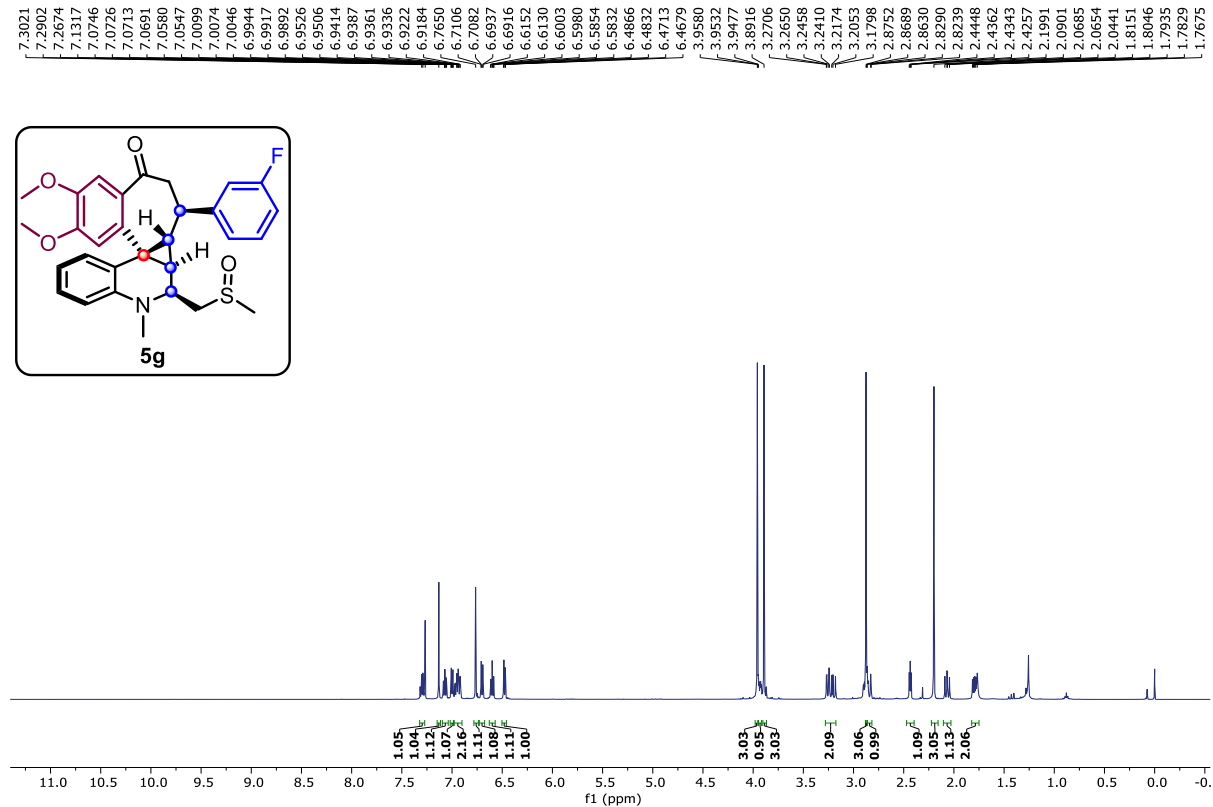
^1H NMR (500 MHz, CDCl_3)



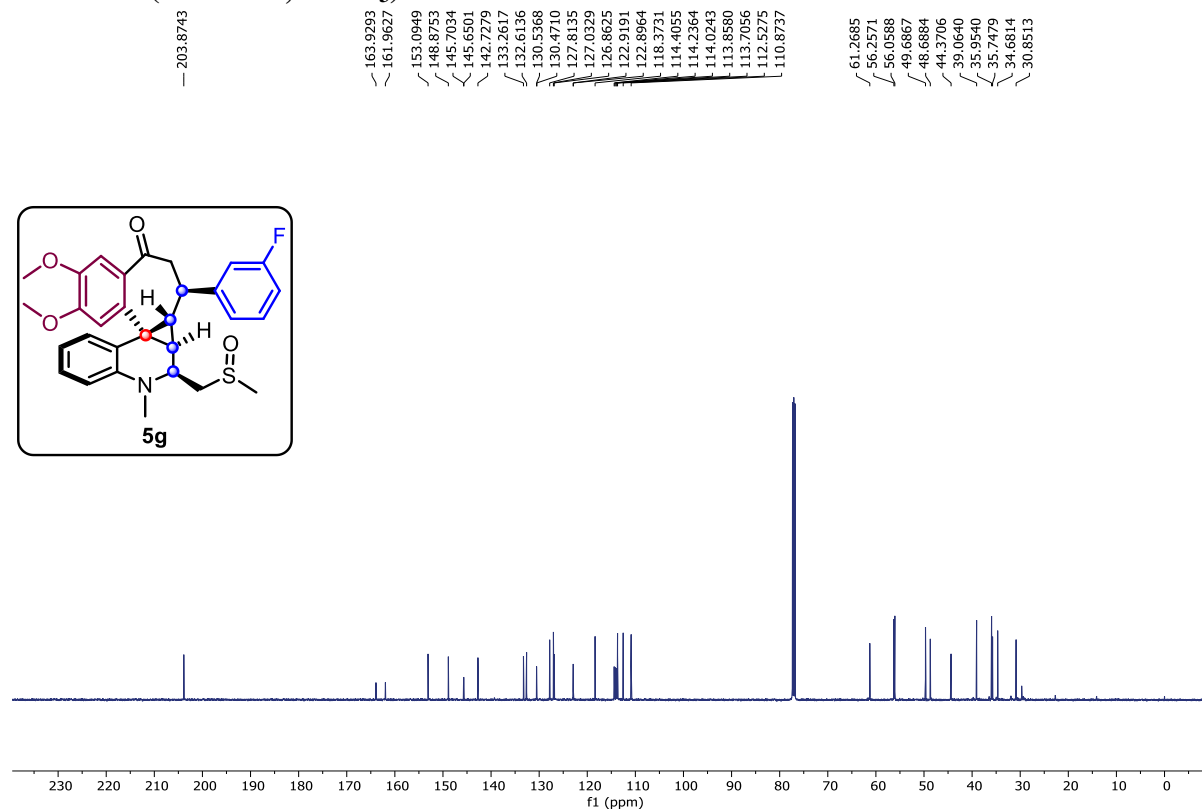
¹³C NMR (125 MHz, CDCl₃)



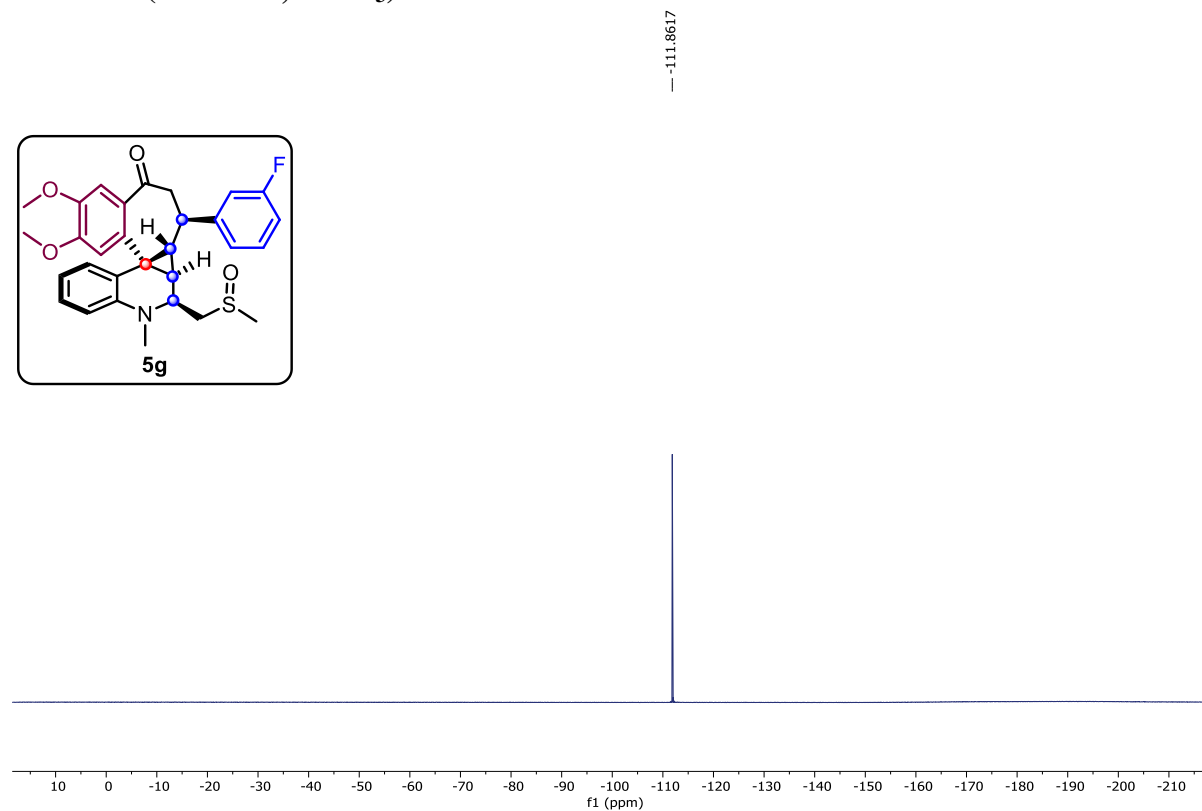
¹H NMR (500 MHz, CDCl₃)



¹³C NMR (125 MHz, CDCl₃)

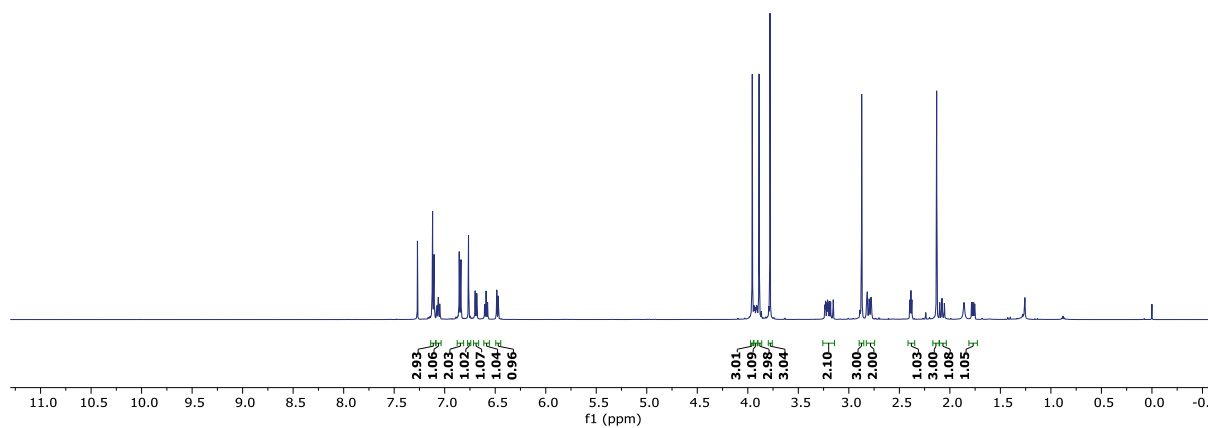
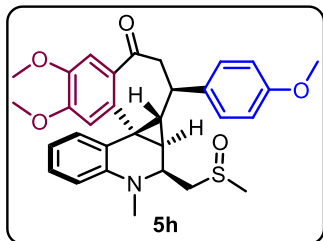


¹⁹F NMR (376 MHz, CDCl₃)



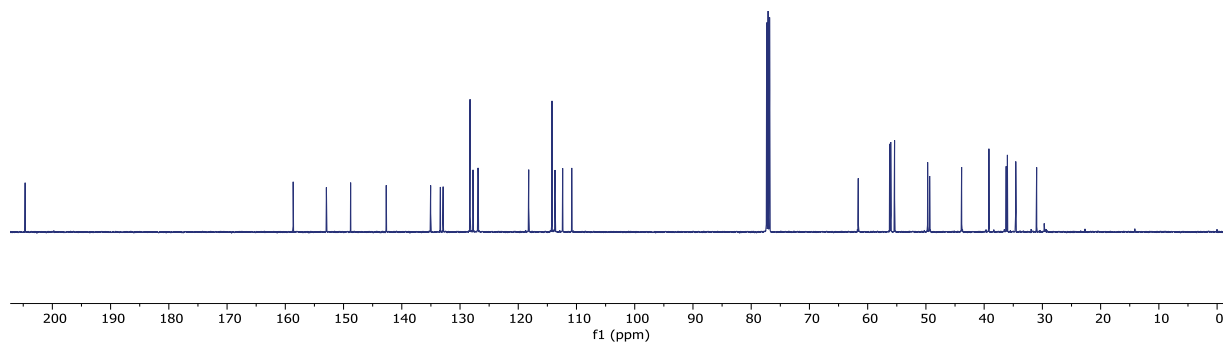
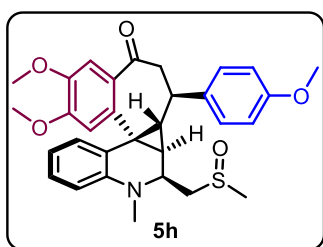
¹H NMR (500 MHz, CDCl₃)

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7.0800
7.0766
7.0654
7.0633
7.0619
7.0600
7.0489
7.0455
6.8564
6.8524
6.8431
6.8390
6.7643
6.7003
6.6980
6.6835
6.6812
6.6054
6.6032
6.5906
6.5882
6.5756
6.5734
6.4873
6.4839
6.4720
6.4686
3.9555
3.9369
3.9210
3.9194
3.9122
3.8880
3.7933
3.7802
3.2357
3.2303
3.2185
3.2111
3.2057
3.1923
3.1802
3.1591
2.8727
2.8217
2.8171
2.7989
2.7840
2.7785
2.3950
2.3862
2.3844
2.3758
2.1307
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1.7752
1.7645
1.7540
1.2579



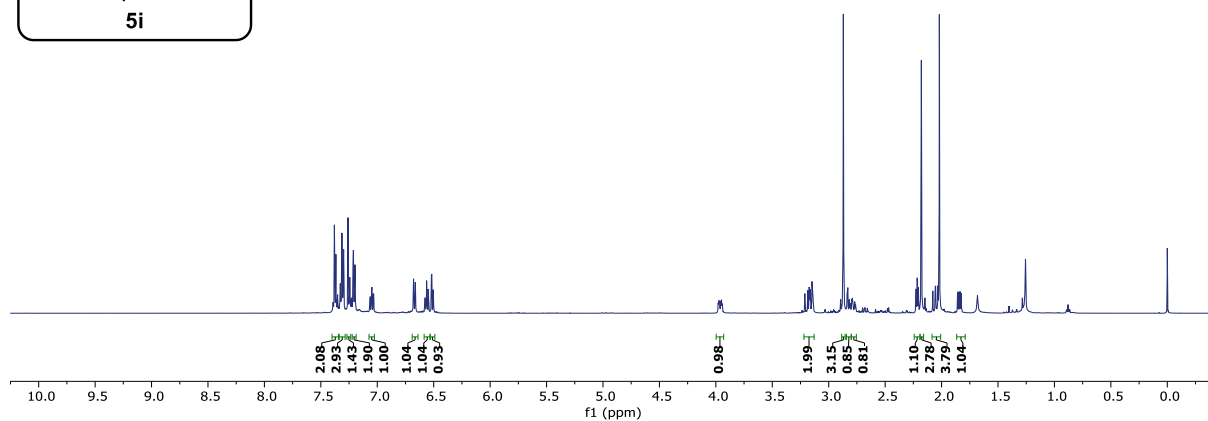
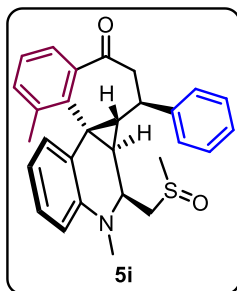
¹³C NMR (125 MHz, CDCl₃)

204.6718
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152.9482
148.7862
142.6658
135.0441
133.3731
132.8978
128.2770
127.7651
126.9606
126.9030
118.2081
114.2056
113.6910
112.3748
110.8157
61.6143
56.2287
56.0443
55.3829
49.7052
49.3599
43.8707
39.1850
36.2195
36.0090
34.5747
30.9879



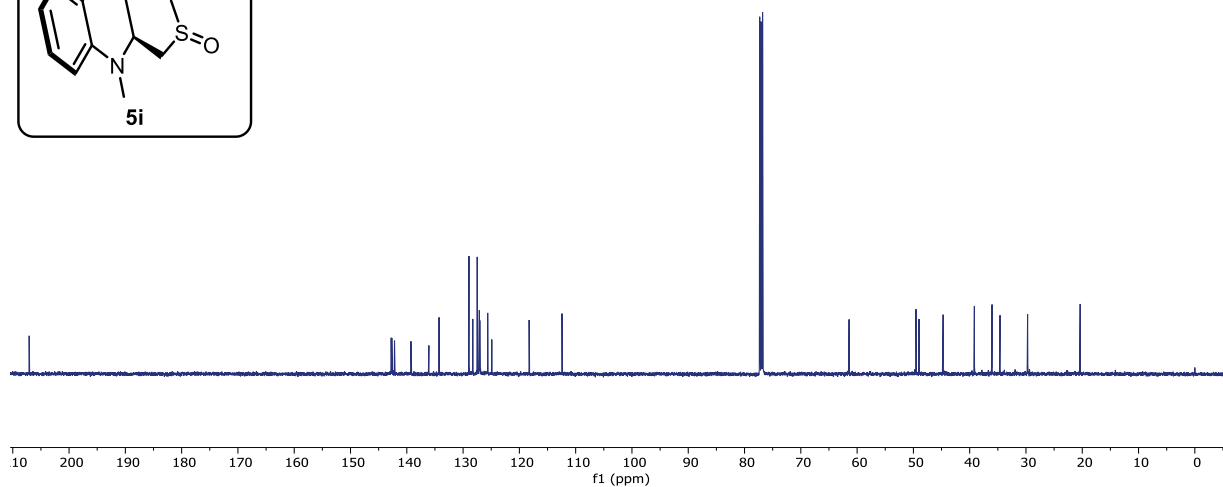
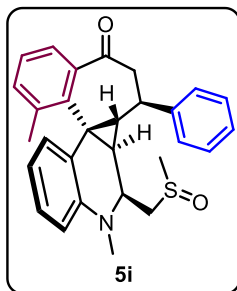
¹H NMR (500 MHz, CDCl₃)

7.3801
7.3752
7.3667
7.3516
7.3294
7.3275
7.3242
7.3169
7.3133
7.3106
7.3032
7.3010
7.2980
7.2591
7.2555
7.2478
7.2435
7.2288
7.2145
7.2114
7.2073
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7.0480
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7.0314
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6.6634
6.6612
6.5629
6.5605
6.5484
6.5462
6.5202
6.5166
6.5048
6.5013
3.2113
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3.1747
3.1692
3.1638
3.1487
3.1445
3.1392
3.1392
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2.8377
2.8304
2.2257
2.2169
2.2151
2.2151
2.2061
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1.8367
1.8260

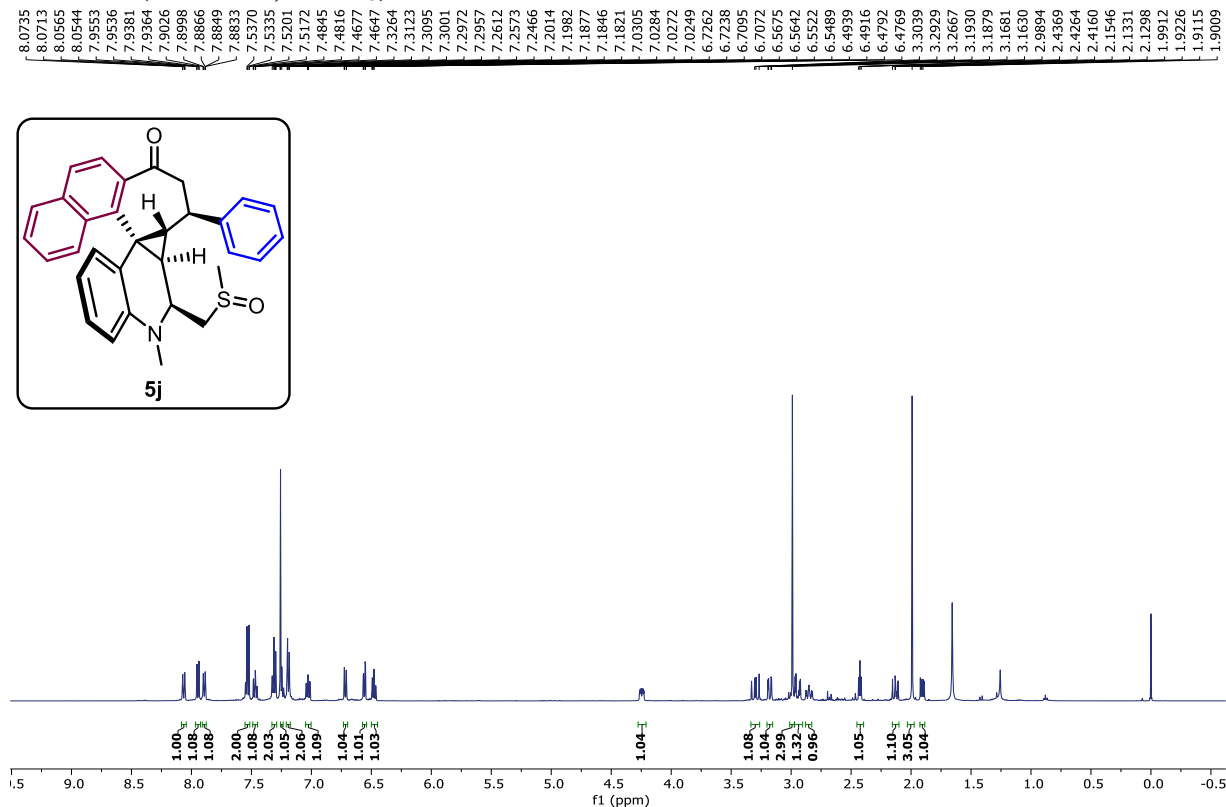


¹³C NMR (125 MHz, CDCl₃)

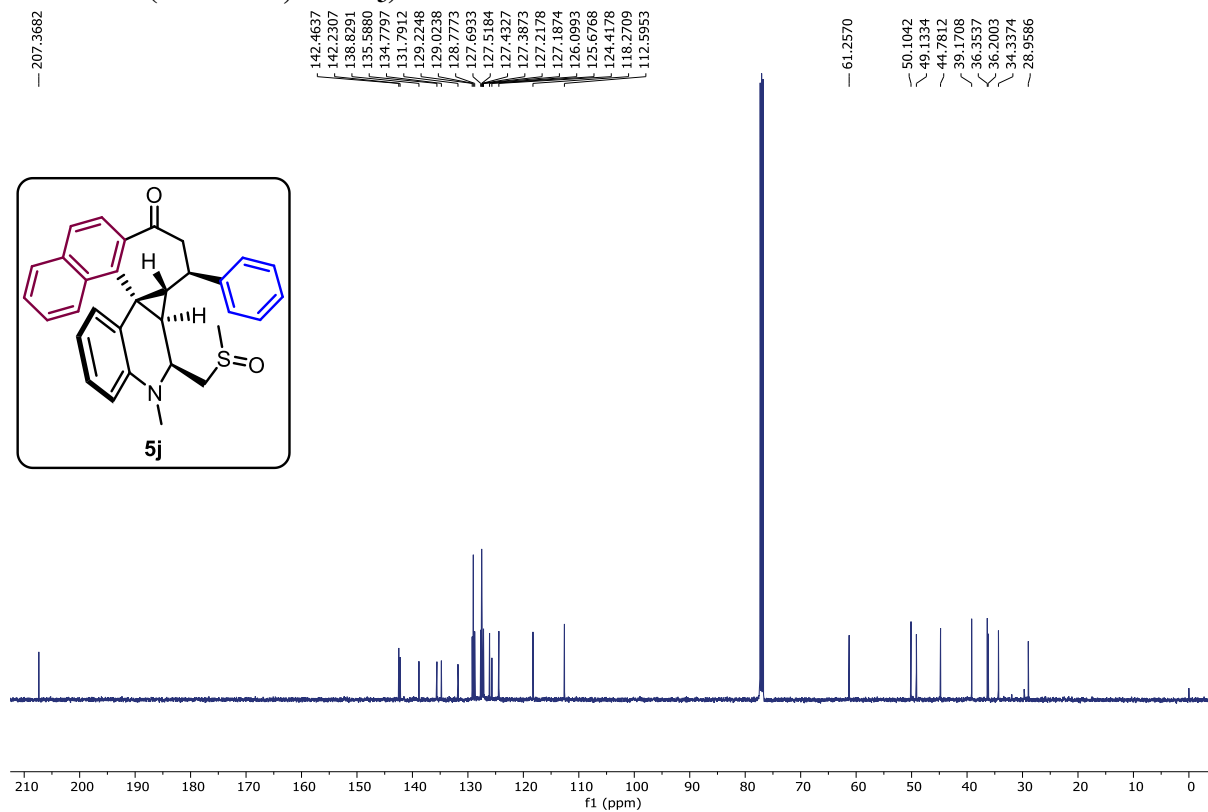
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142.6603
142.1807
139.2661
136.0803
134.2677
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128.2773
127.4921
127.1417
127.1132
127.0274
125.6266
118.8976
118.8976
112.4202
61.4234
49.5332
48.9930
44.7151
39.1668
36.0925
36.0257
34.6123
29.7113
20.3707



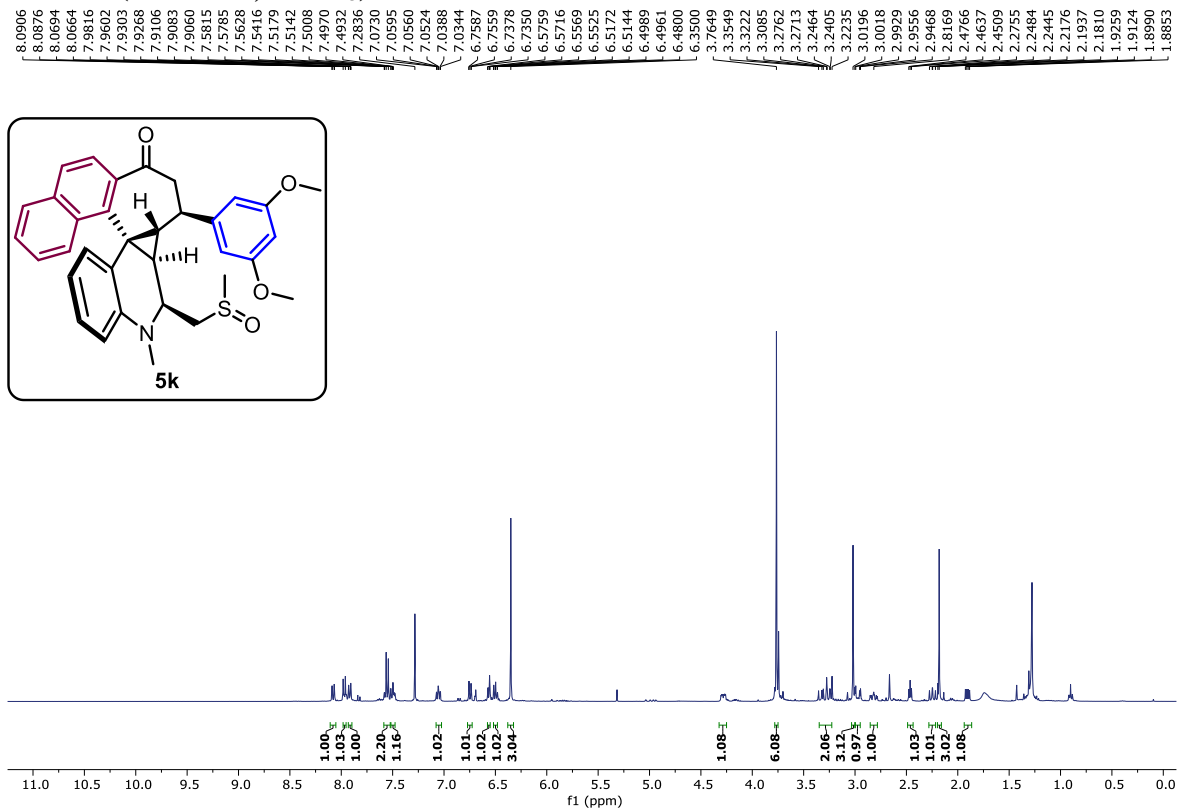
¹H NMR (500 MHz, CDCl₃)



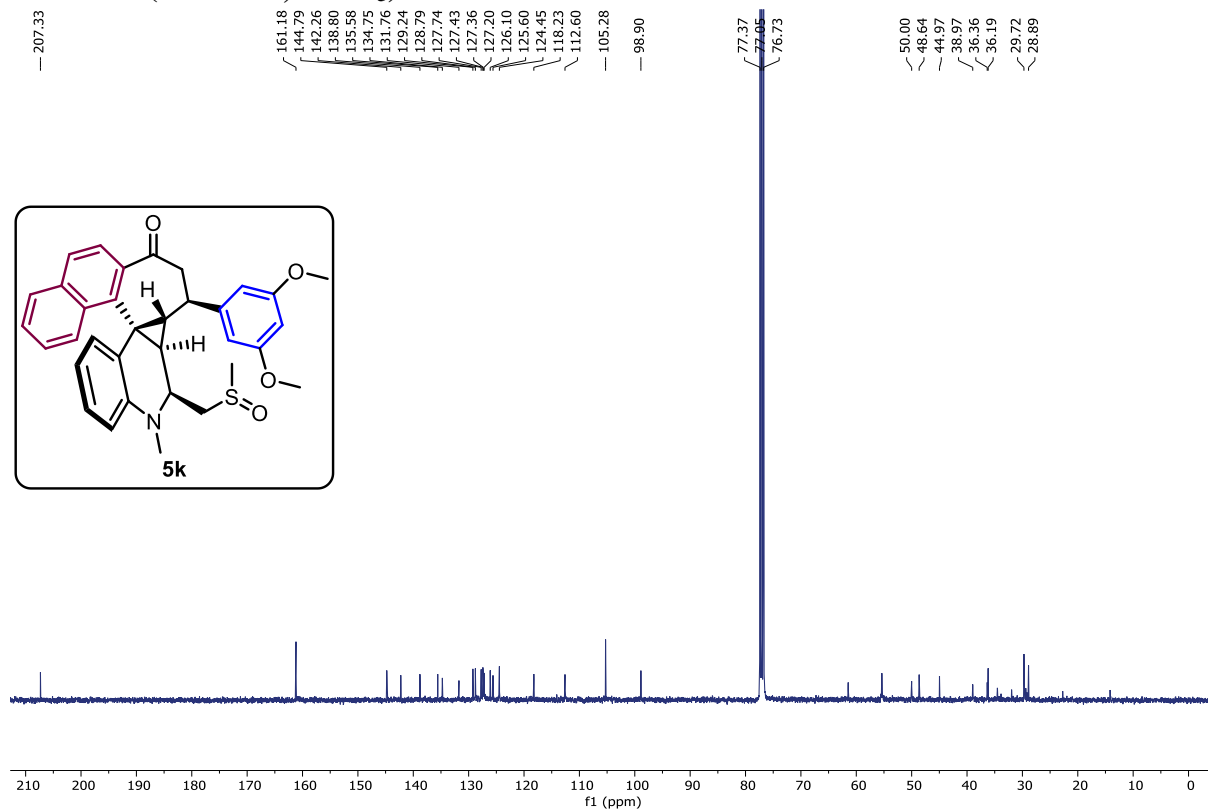
¹³C NMR (125 MHz, CDCl₃)



¹H NMR (400 MHz, CDCl₃)

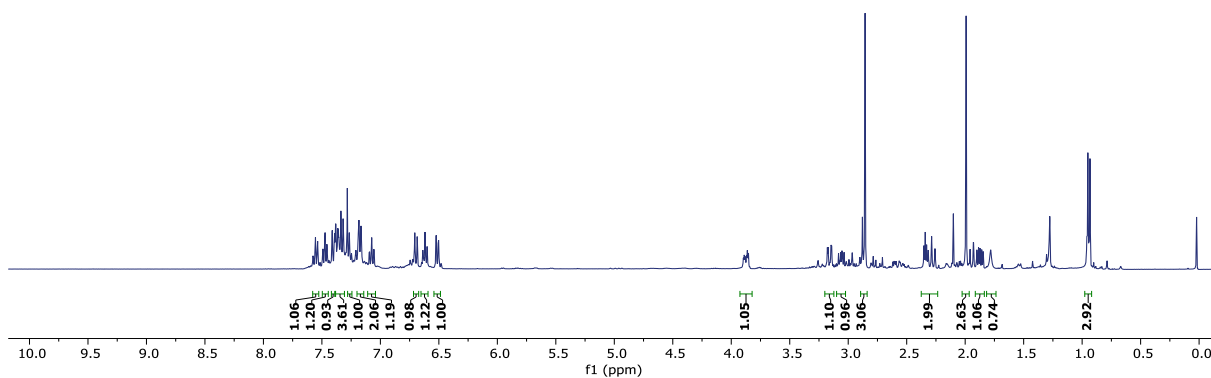
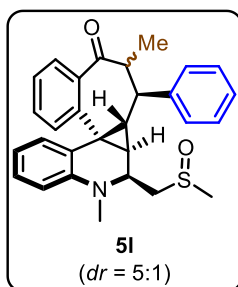


¹³C NMR (100 MHz, CDCl₃)



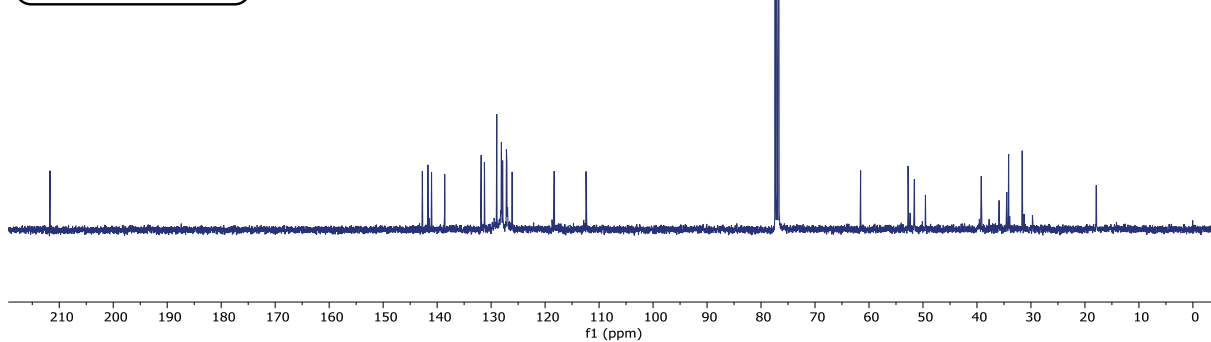
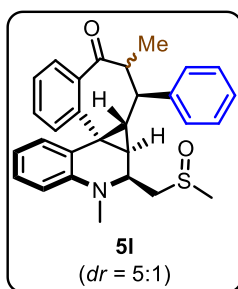
¹H NMR (400 MHz, CDCl₃)

7.5586
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7.4539
7.4127
7.4097
7.3936
7.3905
7.3835
7.3796
7.3647
7.3601
7.3547
7.3473
7.3382
7.3347
7.3187
7.2879
7.2828
7.2656
7.2091
7.1875
7.1828
7.1655
7.0780
7.0746
7.0710
7.0572
7.0529
6.7076
6.7048
6.6866
6.6841
6.6193
6.6164
6.6009
6.5980
6.5251
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6.5017
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3.1719
3.1478
3.1410
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2.3533
2.3415
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2.3153
2.2870
2.2582
1.9917
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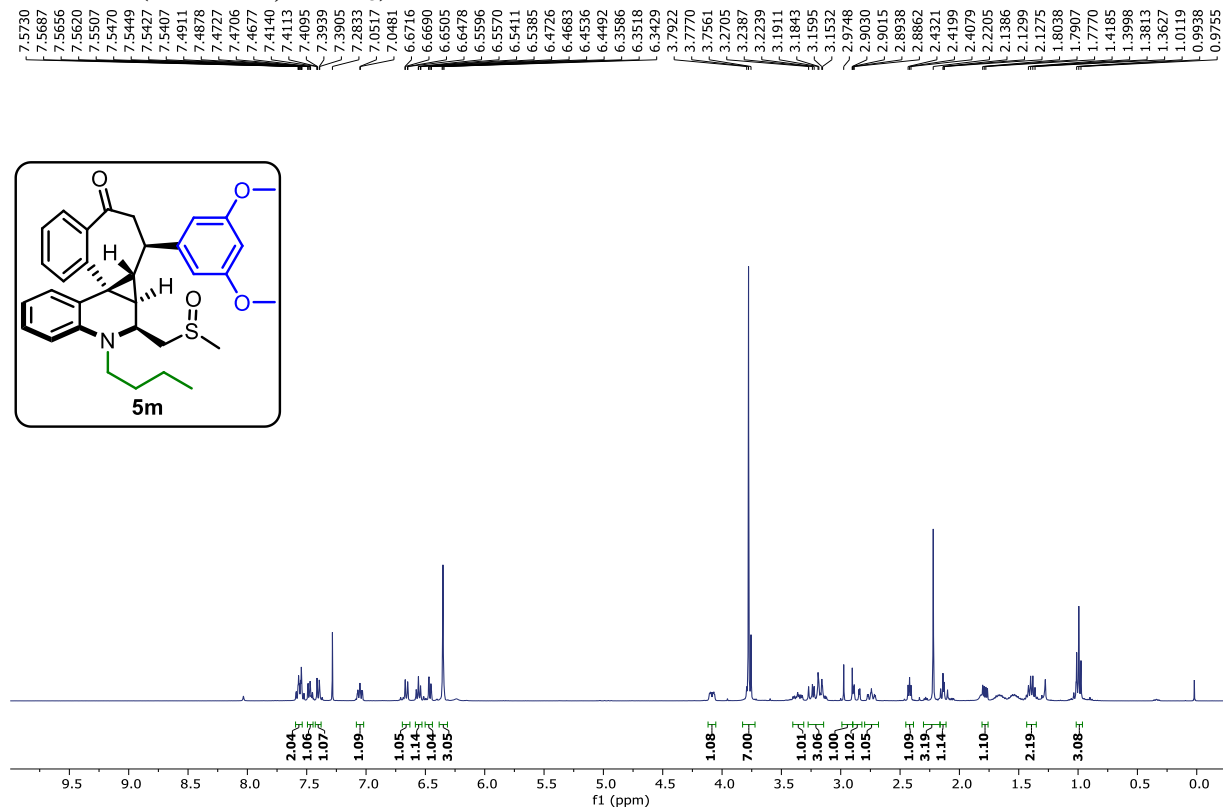


¹³C NMR (100 MHz, CDCl₃)

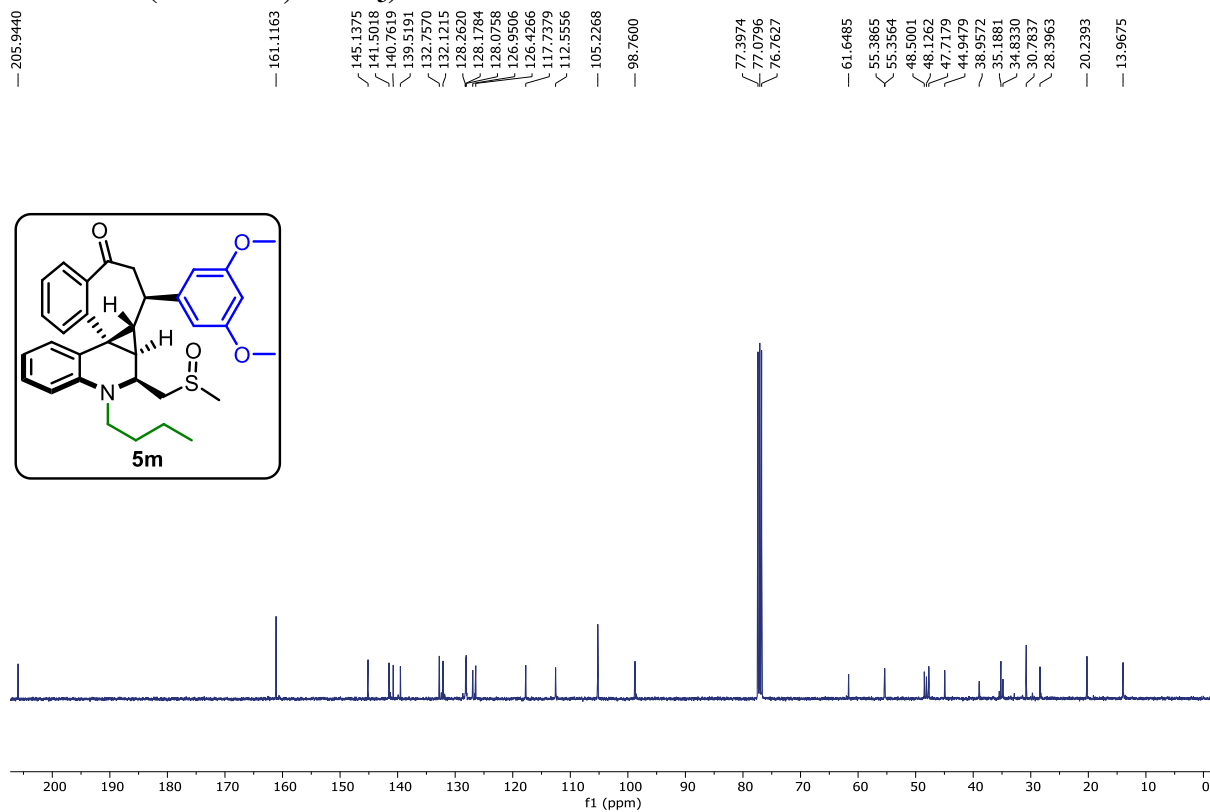
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127.8672
127.8080
127.1637
127.0811
126.9941
126.9562
126.1012
118.3179
112.4093
77.3857
77.0680
76.7504
52.7610
51.5945
49.5377
39.1992
35.9087
34.4832
34.1405
31.6170
29.7177
17.8975



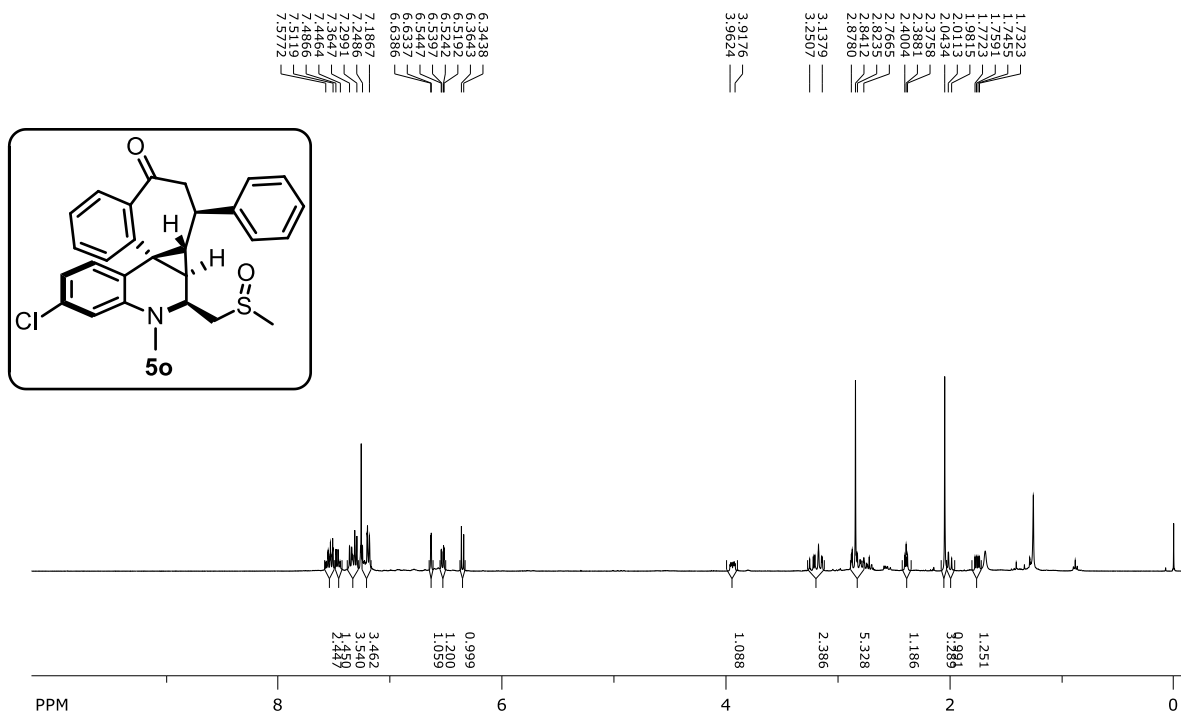
¹H NMR (400 MHz, CDCl₃)



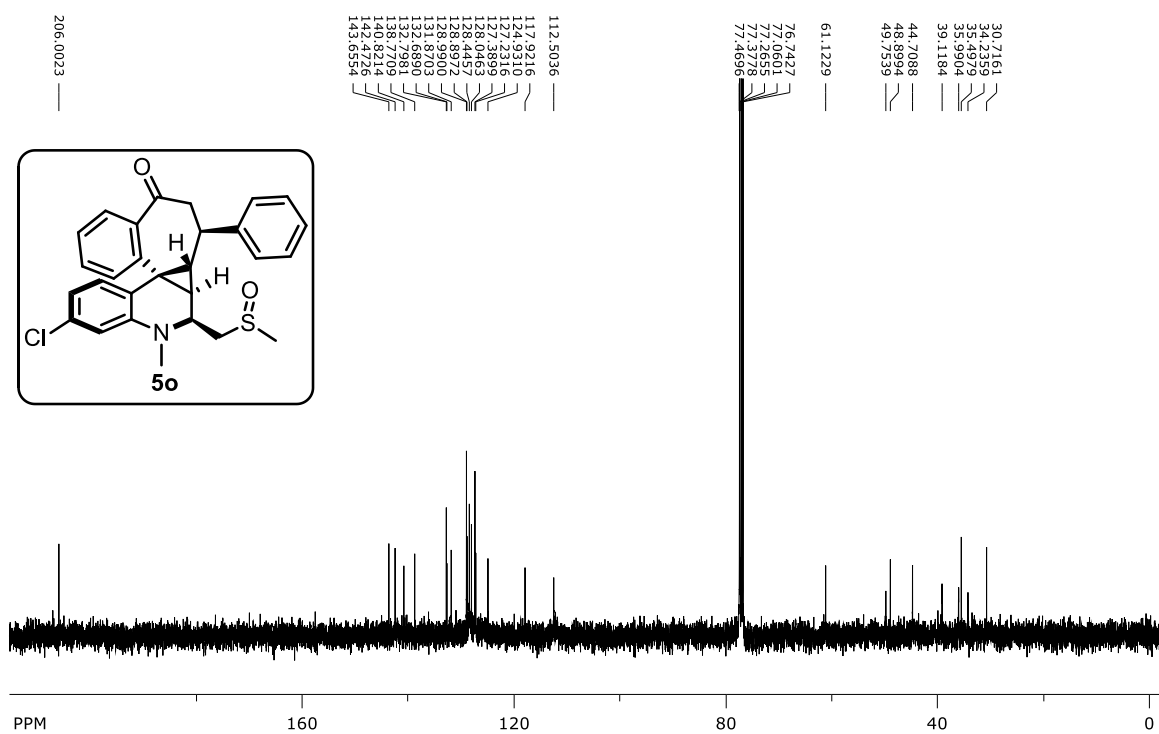
¹³C NMR (100 MHz, CDCl₃)



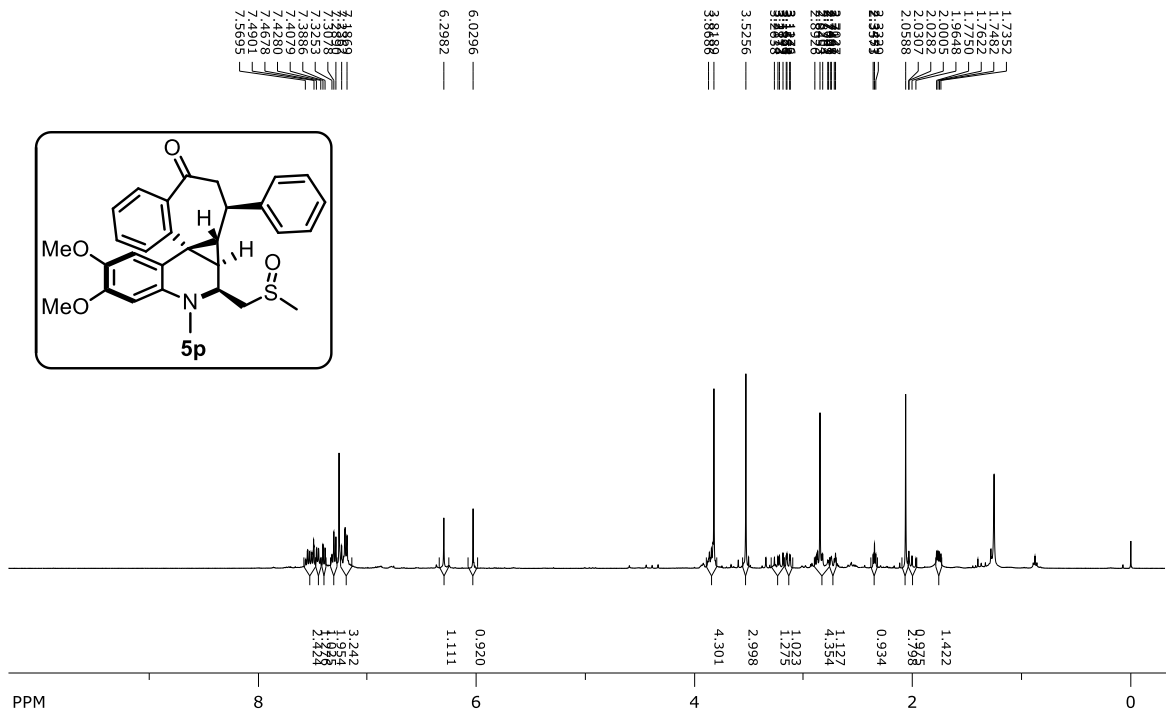
¹H NMR (400 MHz, CDCl₃)



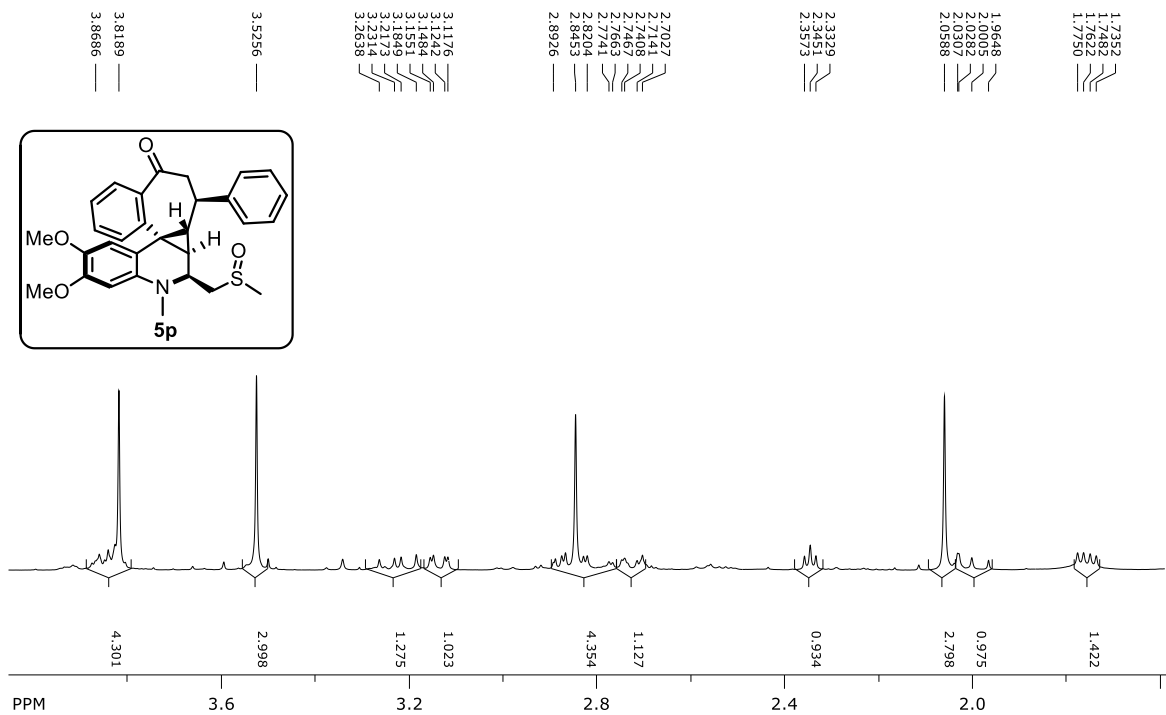
¹³C NMR (100 MHz, CDCl₃)



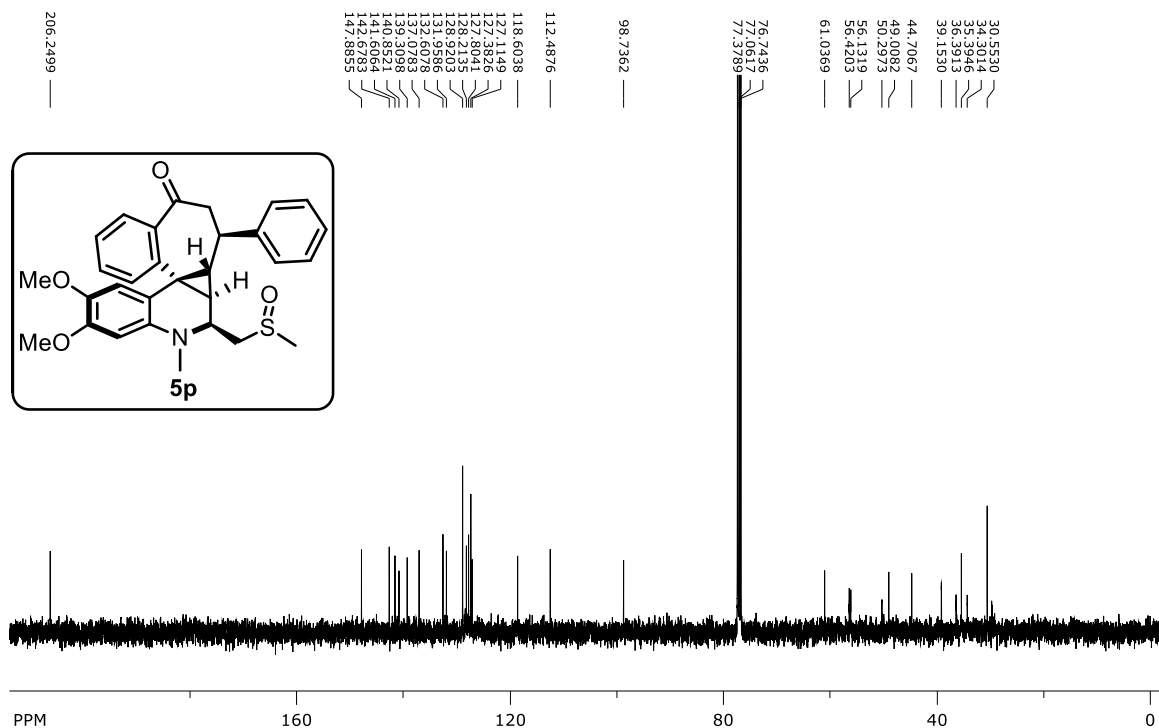
¹H NMR (400 MHz, CDCl₃)



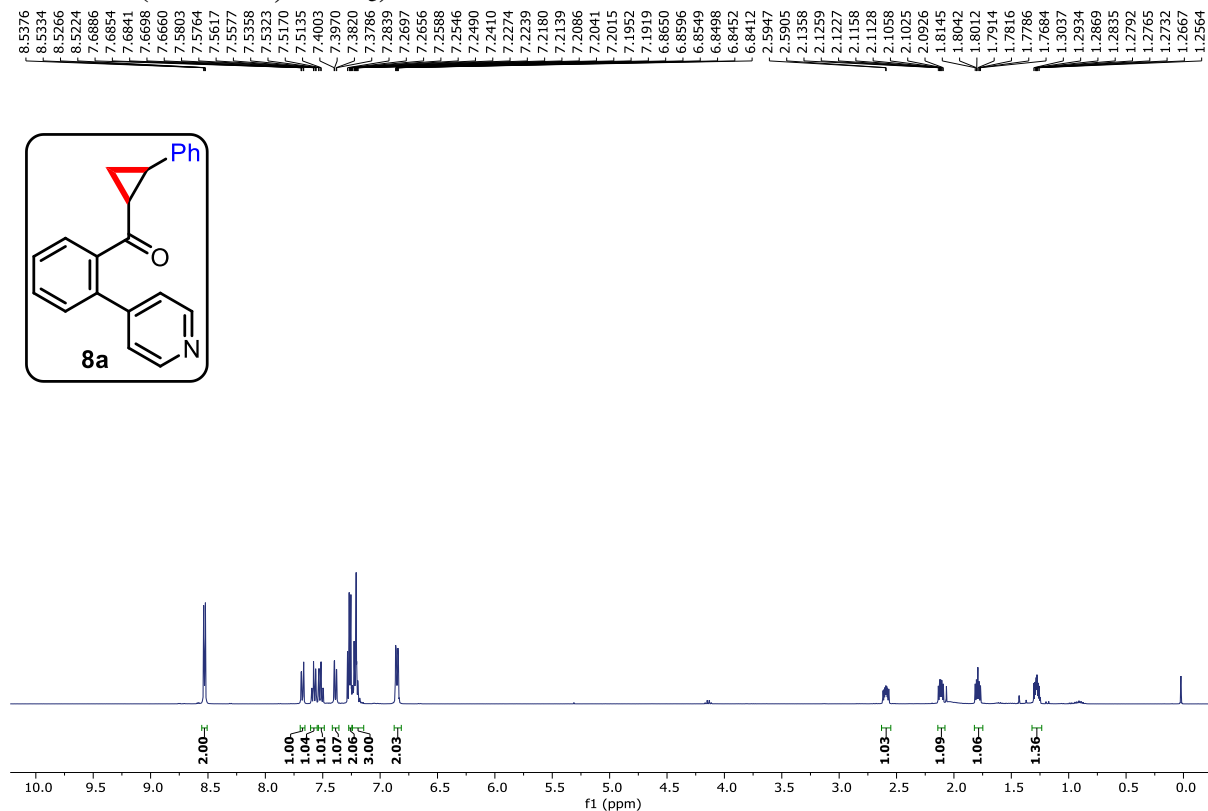
¹H NMR (400 MHz, CDCl₃): expansion of 1.60-4.00 ppm region



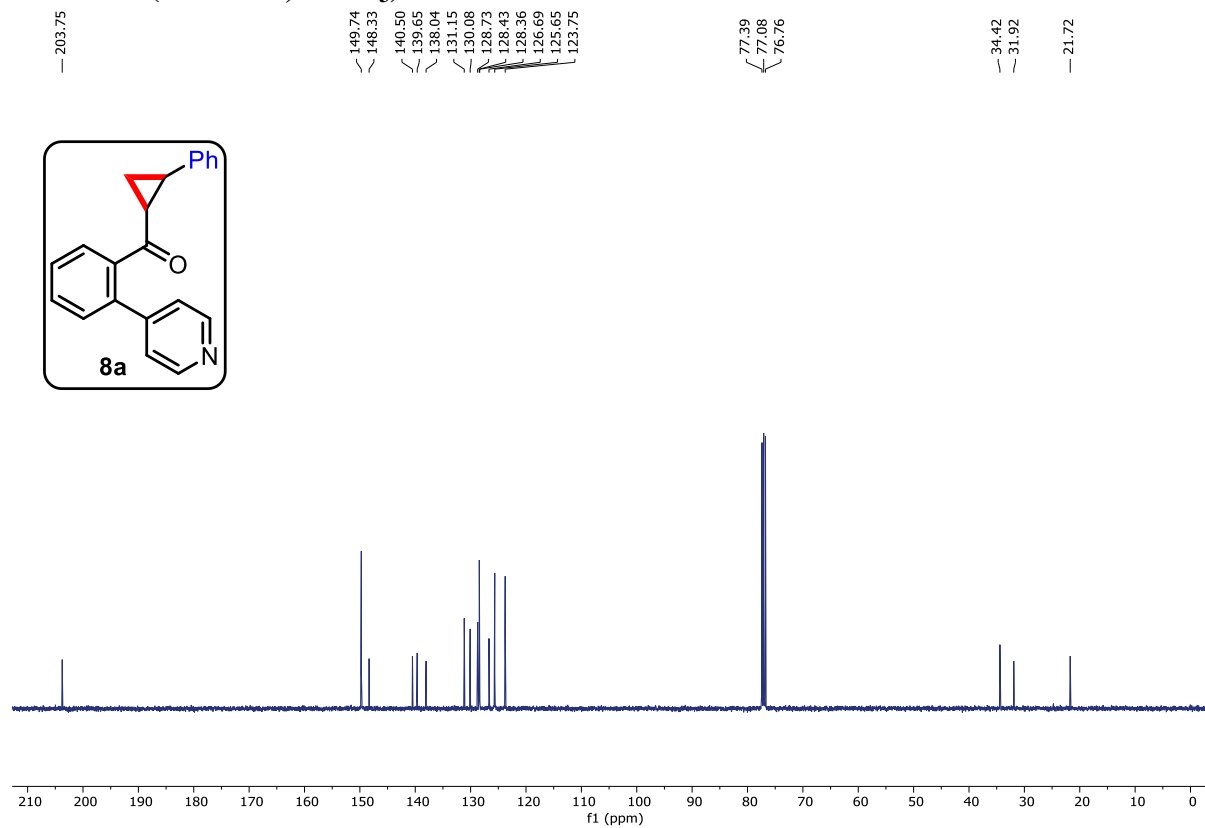
¹³C NMR (100 MHz, CDCl₃)



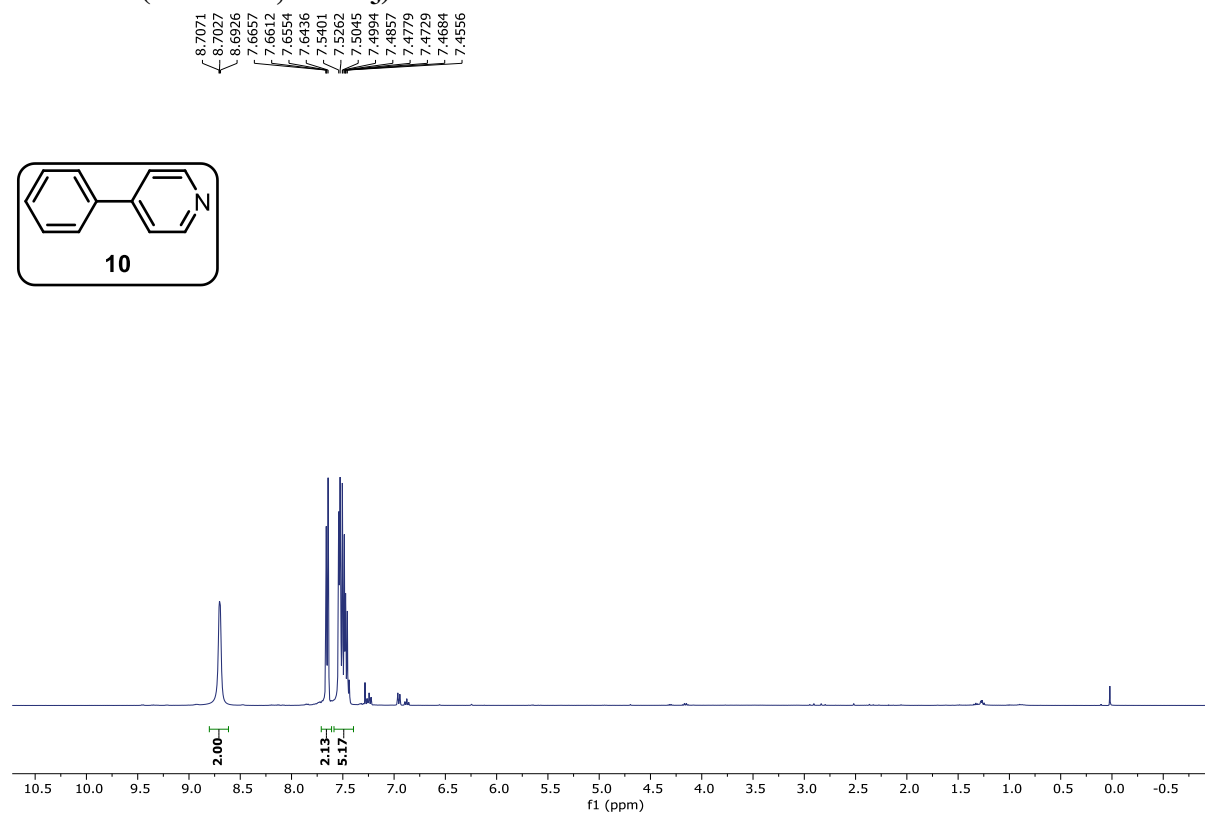
¹H NMR (400 MHz, CDCl₃)



^{13}C NMR (100 MHz, CDCl_3)



^1H NMR (400 MHz, CDCl_3)



^{13}C NMR (100 MHz, CDCl_3)

150.1068
148.4378

138.0857

129.1532
129.1241
127.0132
121.7999

77.4324
77.1147
76.7966

