Supporting Information

Tandem Reaction of Tertiary Enamides as a Synthetic Strategy to Construct the Fused *N*-Pentacyclic Skeleton of Erythrina Alkaloid Derivatives

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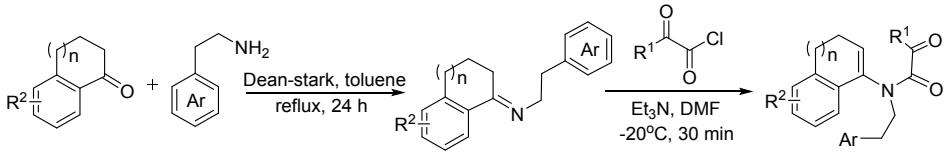
1. General information

All chemicals were dried or purified according to standard procedures prior to use. Flash column chromatography was performed on silica gel (100-200). Reactions were monitored using pre-coated, glass-backed silica gel plates and visualized by means of UV irridation (254 nm) or $KMnO_4$, phosphomolybdic acid, ninhydrine, pancaldi, and *p*-anisaldehyde vanillin. ^1H NMR and ^{13}C NMR spectra were recorded using 500 MHz spectrometers at ambient temperature. Chemical shifts are reported in ppm with either tetramethylsilane or the residual solvent resonance used as an internal standard. Abbreviations are used in the description of NMR data as follows: chemical shift (δ, ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet), coupling constant (*J*, Hz). Mass spectra was measured on Bruker APEX-2 (HRMS) using GCT-MS spectrometer. All yields reported were isolated yields.

2. Preparation of Tertiary Enamides

2.1 General Procedure for the Synthesis of Tertiary Enamides

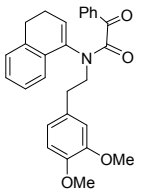


Step 1. The toluene solution of amine (0.5 M) and ketone (0.5 M) was vigorously stirred with Dean-stark at reflux under heating in oil bath until the conversion of amine to imine was completed. The mixture was concentrated in vacuo to give a crude imine product which was used immediately without further purification.

Step 2. Under argon atmosphere, imine (5 mmol) was dissolved in DMF (10 mL), and then Et_3N (6 mmol) was added. After cooling to -20℃, acyl chloride (6 mmol) was added dropwise during 20 min. The resulting mixture was kept stirring at -20℃ for another 30 min. A saturated aqueous $NaHCO_3$ solution (20 mL) was added to quench the reaction. The mixture was extracted with ethyl acetate (3 × 50 mL), and washed with brine (2 × 50 mL). The organic layer was dried over anhydrous Na_2SO_4, and concentrated under vacuum. The residue was chromatographed on a silica gel column eluted with a mixture of petroleum ether and ethyl acetate (5:1) to give a pure

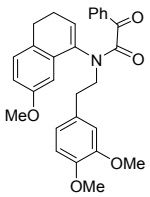
enamide product **1**. The structure of tertiary enamides was fully characterized and the characterization data are listed below.

2.2 Characterization of Tertiary Enamides



N-(3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-oxo-2-phenylacetamide (1a)

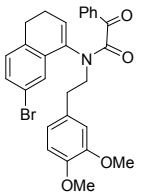
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1a** was obtained as a white solid (1.50 g, 68% yield, 2 steps). m.p. 140-141 ^{o}C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.75-7.69 (m, 2H), 7.57-7.50 (m, 1H), 7.38 (t, *J* = 7.8 Hz, 2H), 7.24 (d, *J* = 4.1 Hz, 2H), 7.22-7.18 (m, 1H), 7.11 (d, *J* = 7.3 Hz, 1H), 6.87-6.79 (m, 3H), 5.58 (dd, *J* = 6.3, 3.1 Hz, 1H), 4.68-4.62 (m, 1H), 3.88 (s, 3H), 3.87 (s, 3H), 3.30-3.25 (m, 1H), 3.00-2.91 (m, 2H), 2.81-2.69 (m, 1H), 2.65-2.60 (m, 1H), 2.15-1.98 (m, 2H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 190.4, 167.4, 149.0, 147.9, 136.9, 136.3, 134.2, 133.8, 130.7, 130.2, 129.7, 129.4, 128.8, 128.6, 127.9, 126.9, 123.4, 121.3, 112.4, 111.3, 56.1, 56.0, 45.7, 33.5, 27.1, 22.8; HRMS (ESI) m/z: $[M+Na]^+$ Calcd. for $C_{28}H_{27}NO_4Na$ 464.1832; Found 464.1829.



N-(3,4-dimethoxyphenethyl)-N-(7-methoxy-3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenylacetamide (1b)

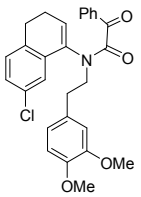
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1b** was obtained as a white solid (1.65 g, 70% yield, 2 steps). m.p. 140-140 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.71-7.59 (m, 3H), 7.48 (t, *J* = 5.0 Hz, 2H), 7.09 (d, *J* = 8.2 Hz, 1H), 6.92-6.91 (m, 2H), 6.82-6.80 (m, 2H), 6.66 (d, *J* = 2.6 Hz, 1H), 5.59 (dd, *J* = 6.4, 2.9

Hz, 1H), 4.57-4.51 (m, 1H), 3.76 (s, 3H), 3.75 (s, 3H), 3.73 (s, 3H), 3.27-3.22 (m, 1H), 2.94-2.82 (m, 2H), 2.65-2.52 (m, 2H), 2.10-2.04 (m, 1H), 1.85-1.76 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 167.1, 158.5, 149.1, 148.0, 135.6, 135.1, 133.5, 131.3, 131.2, 131.0, 129.4, 129.4, 129.1, 129.0, 121.5, 113.5, 113.4, 112.2, 109.6, 56.1, 55.9, 55.7, 45.6, 33.0, 25.7, 22.9; HRMS (ESI) m/z: $[M+Na]^+$ Calcd. for $C_{29}H_{29}NO_5Na$ 494.1938; Found 494.1932.



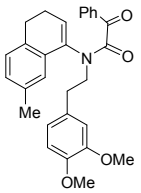
N-(7-bromo-3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-oxo-2-phenylacetamide (1c)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1c** was obtained as a white solid (1.74 g, 67% yield, 2 steps). m.p. 147-148 ^{o}C; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.71-7.65 (m, 1H), 7.64-7.60 (m, 2H), 7.51-7.45 (m, 2H), 7.41 (dd, J = 8.0, 2.1 Hz, 1H), 7.22 (d, J = 2.1 Hz, 1H), 7.14 (dd, J = 8.0, 0.9 Hz, 1H), 6.95-6.88 (m, 2H), 6.81 (dd, J = 8.1, 2.0 Hz, 1H), 5.64 (dd, J = 6.3, 3.0 Hz, 1H), 4.52 (dt, J = 13.5, 7.5 Hz, 1H), 3.75 (s, 3H), 3.72 (s, 3H), 3.24 (ddd, J = 13.4, 7.4, 5.6 Hz, 1H), 2.97-2.77 (m, 2H), 2.68-2.53 (m, 2H), 2.15-2.07 (m, 1H), 1.88-1.78 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.5, 166.5, 148.7, 147.5, 135.9, 134.7, 134.2, 132.9, 132.2, 131.9, 130.8, 130.4, 129.8, 129.0, 128.9, 125.3, 121.0, 119.4, 113.0, 111.8, 55.6, 55.5, 45.2, 32.5, 25.4, 21.9; HRMS (ESI) m/z $[M+Na]^+$ Calcd. for $C_{28}H_{26}NO_4BrNa$ 542.0937, 544.0917; Found 542.0931, 544.0911.



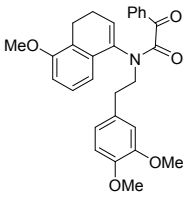
N-(7-chloro-3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-oxo-2-phenylacetamide (1d)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1d** was obtained as a white solid (1.54 g, 65% yield, 2 steps). m.p. 140-141 ^{o}C; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.72-7.65 (m, 1H), 7.65-7.57 (m, 2H), 7.52-7.43 (m, 2H), 7.31-7.24 (m, 1H), 7.19 (d, J = 7.9 Hz, 1H), 7.10 (d, J = 2.2 Hz, 1H), 6.98-6.88 (m, 2H), 6.81 (dd, J = 8.1, 2.0 Hz, 1H), 5.65 (dd, J = 6.3, 3.0 Hz, 1H), 4.52 (dt, J = 13.5, 7.5 Hz, 1H), 3.74 (d, J = 12.7 Hz, 6H), 3.24 (ddd, J = 13.4, 7.5, 5.7 Hz, 1H), 2.97-2.77 (m, 2H), 2.71-2.52 (m, 2H), 2.15-2.07 (m, 1H), 1.90-1.76 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.5, 166.5, 148.7, 147.5, 135.5, 134.7, 134.3, 132.9, 131.92, 131.89, 131.2, 130.4, 129.5, 128.96, 128.95, 127.9, 122.5, 121.1, 113.0, 111.8, 55.6, 55.5, 45.2, 32.5, 25.4, 21.9; HRMS (ESI) m/z [M+Na]$^{+}$ Calcd. for $C_{28}H_{26}NO_4ClNa$ 498.1443; Found 498.1433.



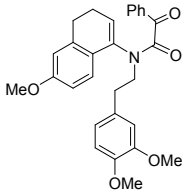
N-(3,4-dimethoxyphenethyl)-N-(7-methyl-3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenylacetamide (1e)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1e** was obtained as a white solid (1.60 g, 70% yield, 2 steps). m.p. 141-141 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.68-7.62 (m, 3H), 7.47 (t, J = 7.8 Hz, 2H), 7.05-7.01 (m, 2H), 6.95 (s, 1H), 6.92 (dd, J = 5.1, 3.1 Hz, 2H), 6.81 (dd, J = 8.2, 2.0 Hz, 1H), 5.55 (dd, J = 6.4, 3.0 Hz, 1H), 4.56-4.51 (m, 1H), 3.76 (s, 3H), 3.72 (s, 3H), 3.27-3.22 (m, 1H), 2.93-2.82 (m, 2H), 2.66-2.58 (m, 1H), 2.54-2.51 (m, 1H), 2.28 (s, 3H), 2.10-2.03 (m, 1H), 1.85-1.77 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 167.0, 149.1, 148.0, 136.0, 135.8, 135.0, 134.1, 133.5, 131.0, 130.3, 130.1, 129.4, 129.2, 128.1, 124.0, 121.5, 113.4, 112.3, 56.1, 55.9, 45.7, 33.0, 26.3, 22.7, 21.3; HRMS (ESI) m/z [M+Na]$^{+}$ Calcd. for $C_{29}H_{29}NO_4Na$ 478.1989; Found 478.1986.



N-(3,4-dimethoxyphenethyl)-N-(5-methoxy-3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenylacetamide (1f)

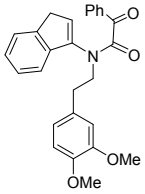
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1f** was obtained as a white solid (1.67 g, 71% yield, 2 steps). m.p. 130-131 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.68-7.66 (m, 1H), 7.65-7.60 (m, 2H), 7.47 (t, J = 7.8 Hz, 2H), 7.23 (t, J = 8.0 Hz, 1H), 6.96 (d, J = 8.3 Hz, 1H), 6.91 (dd, J = 5.1, 3.1 Hz, 2H), 6.80 (d, J = 8.0 Hz, 2H), 5.55 (dd, J = 6.4, 2.9 Hz, 1H), 4.56-4.50 (m, 1H), 3.77 (s, 3H), 3.76 (s, 3H), 3.73 (s, 3H), 3.21-3.16 (m, 1H), 2.92-2.77 (m, 3H), 2.40-2.32 (m, 1H), 2.12-2.06 (m, 1H), 1.81-1.73 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 167.0, 156.2, 149.1, 148.0, 135.5, 135.0, 133.6, 131.2, 131.0, 130.5, 129.4, 129.3, 127.3, 124.4, 121.5, 116.1, 113.4, 112.2, 111.8, 56.0, 56.0, 55.9, 45.7, 33.0, 22.0, 19.0; HRMS (ESI) m/z $[M+Na]^+$ Calcd. for $C_{29}H_{29}NO_5Na$ 494.1938; Found 494.1932.



N-(3,4-dimethoxyphenethyl)-N-(6-methoxy-3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenylacetamide (1g)

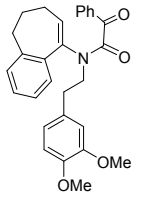
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1g** was obtained as a white solid (1.65 g, 70% yield, 2 steps). m.p. 131-132 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.68-7.65 (m, 1H), 7.62-7.60 (m, 2H), 7.47 (t, J = 7.8 Hz, 2H), 7.09 (d, J = 8.4 Hz, 1H), 6.92-6.91 (m, 2H), 6.82-6.77 (m, 3H), 5.40 (dd, J = 6.4, 2.9 Hz, 1H), 4.55-4.49 (m, 1H), 3.76 (d, 6H), 3.73 (s, 3H), 3.24-3.18 (m, 1H), 2.92-2.81 (m, 2H), 2.70-2.63 (m, 1H), 2.57-2.52 (m, 1H), 2.09-2.02 (m, 1H), 1.83-1.77 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 167.0, 159.7, 149.1, 148.0, 139.1, 135.5, 135.0,

133.6, 131.0, 129.4, 129.3, 127.6, 124.9, 123.1, 121.5, 114.3, 113.4, 112.2, 111.9, 56.1, 55.9, 55.6, 45.4, 33.0, 27.1, 22.5; HRMS (ESI) m/z $[M+Na]^+$ Calcd. for $C_{29}H_{29}NO_5Na$ 494.1938; Found, 494.1934.



N-(3,4-dimethoxyphenethyl)-N-(1H-inden-3-yl)-2-oxo-2-phenylacetamide (1h)

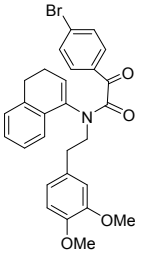
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1h** was obtained as a white solid (0.90 g, 42% yield, 2 steps). m.p. 200-201 ^oC; ^1H NMR (500 MHz, DMSO-d_6) δ 7.65 (t, J = 7.5 Hz, 1H), 7.58 (d, J = 7.7 Hz, 2H), 7.50-7.38 (m, 4H), 7.31 (t, J = 7.5 Hz, 1H), 7.25 (t, J = 7.3 Hz, 1H), 6.96-6.87 (m, 2H), 6.79 (dd, J = 8.1, 2.0 Hz, 1H), 6.04 (d, J = 2.2 Hz, 1H), 4.13 (s, 2H), 3.76 (s, 3H), 3.71 (s, 3H), 3.23 (s, 2H), 2.86 (t, J = 7.1 Hz, 2H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 166.8, 149.1, 148.0, 143.1, 140.24, 140.15, 135.2, 133.7, 133.2, 130.8, 129.45, 129.40, 126.8, 126.3, 124.8, 121.5, 119.7, 113.4, 112.2, 56.0, 55.9, 45.8, 36.4, 33.1; HRMS (ESI) m/z $[M+Na]^+$ Calcd. for $C_{27}H_{25}NO_4Na$ 450.1676; Found 450.1670.



N-(6,7-dihydro-5H-benzo[7]annulen-9-yl)-N-(3,4-dimethoxyphenethyl)-2-oxo-2-phenylacetamide (1i)

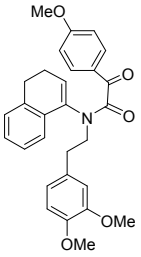
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1i** was obtained as a white solid (1.16 g, 51% yield, 2 steps). m.p. 137-138 ^oC; ^1H NMR (500 MHz, DMSO-d_6) δ 7.70-7.67 (m, 1H), 7.65-7.64 (m, 2H), 7.50 (t, J = 7.8 Hz, 2H), 7.37 (d, J = 7.1 Hz, 1H), 7.33-7.26 (m, 3H), 6.93-6.88 (m, 2H), 6.77 (dd, J = 8.2, 2.0 Hz, 1H), 5.65 (t, J = 7.0 Hz, 1H), 4.32 (b, 1H), 3.77 (s, 3H), 3.73 (s, 3H), 3.07 (b, 1H), 2.82 (s, 2H), 2.53 (s, 2H), 1.94 (s, 2H), 1.46 (b, 2H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 191.0,

167.1, 149.2, 148.1, 142.9, 138.0, 135.0, 134.2, 133.9, 131.1, 130.5, 129.8, 129.4, 129.2, 127.5, 126.9, 121.5, 113.4, 112.3, 56.1, 55.9, 45.6, 34.4, 33.2, 32.4, 24.2; HRMS (ESI) m/z $[M+Na]^+$ Calcd. For $C_{29}H_{29}NO_4Na$ 478.1989; Found 478.1986.



2-(4-bromophenyl)-N-(3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-oxoacetamide (1j)

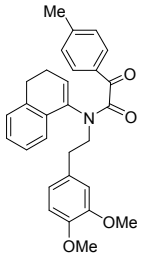
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1j** was obtained as a white solid (1.82 g, 70% yield, 2 steps). m.p. 132-132 ^oC; ^1H NMR (500 MHz, DMSO-d_6) δ 7.70-7.65 (m, 2H), 7.57-7.54 (m, 2H), 7.27-7.7.21 (m, 2H), 7.17-7.14 (m, 2H), 6.93-6.91 (m, 2H), 6.80 (dd, J = 8.1, 2.0 Hz, 1H), 5.57 (dd, J = 6.4, 3.0 Hz, 1H), 4.53-4.47 (m, 1H), 3.76 (s, 3H), 3.72 (s, 3H), 3.26-3.20 (m, 1H), 2.93-2.83 (m, 2H), 2.73-2.65 (m, 1H), 2.61-2.55 (m, 1H), 2.15-2.08 (m, 1H), 1.91-1.82 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 189.9, 166.6, 149.1, 148.0, 137.2, 135.7, 132.6, 132.5, 131.2, 131.0, 130.6, 130.2, 129.3, 128.8, 128.2, 127.1, 123.4, 121.5, 113.4, 112.3, 56.0, 55.9, 45.8, 33.0, 26.6, 22.5; HRMS (ESI) m/z $[M+Na]^+$ Calcd. For $C_{28}H_{26}NO_4BrNa$ 542.0937, 544.0917; Found 542.0935, 544.0912.



N-(3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-(4-methoxyphenyl)-2-oxoacetamide (1k)

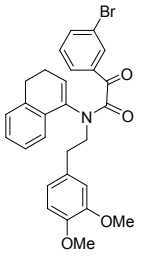
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1k** was obtained

as a white solid (1.60 g, 68% yield, 2 steps). m.p. 130-131 ^{o}C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.72-7.66 (m, 2H), 7.26-7.23 (m, 2H), 7.22-7.16 (m, 1H), 7.11 (d, *J* = 7.3 Hz, 1H), 6.88-6.78 (m, 5H), 5.59 (dd, *J* = 6.3, 3.1 Hz, 1H), 4.64 (dt, *J* = 13.4, 8.0 Hz, 1H), 3.88 (s, 1H), 3.84 (s, 1H), 3.82-3.73 (m, 1H), 3.25 (ddd, *J* = 13.6, 8.3, 5.4 Hz, 1H), 2.98-2.88 (m, 2H), 2.76 (td, *J* = 15.1, 7.1 Hz, 1H), 2.63 (ddd, *J* = 15.6, 6.6, 3.6 Hz, 1H), 2.17-1.98 (m, 2H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 189.0, 167.7, 164.3, 148.9, 147.7, 136.8, 136.3, 131.7, 130.7, 130.2, 129.4, 128.4, 127.7, 126.9, 126.8, 123.4, 121.2, 114.0, 112.3, 111.1, 56.0, 55.5, 45.6, 33.4, 27.1, 22.7; HRMS (ESI) m/z $[M+H]^+$ Calcd. For $C_{29}H_{30}NO_5$ 472.2118; Found 472.2118.



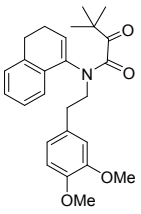
N-(3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-oxo-2-(p-tolyl)acetamide (1l)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1l** was obtained as a white solid (1.46 g, 65% yield, 2 steps). m.p. 120-120 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.53 (d, *J* = 8.0 Hz, 2H), 7.29-7.20 (m, 4H), 7.17-7.15 (m, 2H), 6.92-6.89 (m, 2H), 6.80 (dd, *J* = 8.1, 2.0 Hz, 1H), 5.55 (dd, *J* = 6.5, 2.9 Hz, 1H), 4.56-4.50 (m, 1H), 3.76 (s, 3H), 3.73 (s, 3H), 3.23-3.17 (m, 1H), 2.92-2.82 (m, 2H), 2.72-2.65 (m, 1H), 2.60-2.55 (m, 1H), 2.37 (s, 3H), 2.12-2.05 (m, 1H), 1.87-1.70 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.5, 167.2, 149.1, 148.0, 145.7, 137.2, 135.7, 131.2, 131.0, 130.3, 130.0, 129.5, 128.7, 128.2, 127.0, 123.5, 121.5, 113.4, 112.2, 56.0, 55.9, 45.5, 33.0, 26.6, 22.5, 21.8; HRMS (ESI) $[M+H]^+$ Calcd. For $C_{29}H_{30}NO_4$ 456.2169; Found 456.2169.



2-(3-bromophenyl)-N-(3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-2-oxoacetamide (1m)

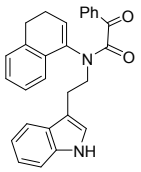
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1m** was obtained as a white solid (1.84 g, 71% yield, 2 steps). m.p. 152-153 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.89-7.83 (m, 2H), 7.60 (dt, J = 7.8, 1.3 Hz, 1H), 7.43 (t, J = 7.9 Hz, 1H), 7.27-7.20 (m, 2H), 7.16 (dd, J = 7.3, 1.7 Hz, 2H), 6.92-6.88 (m, 2H), 6.80 (dd, J = 8.2, 2.1 Hz, 1H), 5.57 (dd, J = 6.3, 3.0 Hz, 1H), 4.52-4.46 (m, 1H), 3.75 (s, 3H), 3.72 (s, 3H), 3.28-3.23 (m, 1H), 2.93-2.83 (m, 2H), 2.72-2.65 (m, 1H), 2.60-2.55 (m, 1H), 2.15-2.08 (m, 1H), 1.92-1.83 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 189.5, 166.3, 149.2, 148.0, 137.8, 137.1, 135.7, 135.3, 131.7, 131.2, 130.9, 130.7, 130.1, 128.8, 128.7, 128.2, 127.0, 123.5, 122.8, 121.3, 113.3, 112.3, 56.0, 55.9, 45.9, 33.0, 26.6, 22.5; HRMS (ESI) m/z $[M+H]^+$ Calcd. For $C_{28}H_{27}NO_4Br$ 520.1118, 522.1098; Found 520.1118, 522.1096.



N-(3,4-dihydronaphthalen-1-yl)-N-(3,4-dimethoxyphenethyl)-3,3-dimethyl-2-oxo butanamide (1n)

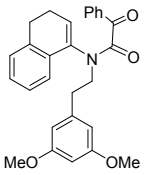
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1n** was obtained as a white solid (1.13 g, 54% yield, 2 steps). m.p. 165-166 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.24-7.19 (m, 3H), 7.06-7.04 (m, 1H), 6.85-6.81 (m, 2H), 6.71 (dd, J = 8.2, 1.9 Hz, 1H), 5.80 (dd, J = 6.0, 3.3 Hz, 1H), 4.33-4.27 (m, 1H), 3.71 (s, 6H), 3.14-3.08 (m, 1H), 2.84-2.76 (m, 2H), 2.75-2.62 (m, 2H), 2.32-2.25 (m, 1H),

2.19-2.11 (m, 1H), 1.08 (s, 9H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 207.4, 167.4, 149.1, 147.9, 137.1, 136.4, 131.0, 130.9 , 129.3, 128.6, 128.2, 127.0, 123.6, 121.2, 113.1, 112.3, 56.0, 55.9, 46.3, 42.2, 33.1, 27.1, 26.7, 22.5; HRMS (ESI) m/z $[M+H]^+$ Calcd. For $C_{26}H_{32}NO_4$ 422.2326; Found: 422.2324.



N-(2-(1H-indol-3-yl)ethyl)-N-(3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenylaceta mide (1o)

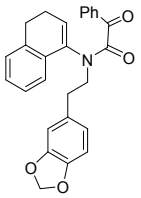
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1o** was obtained as a white solid (1.62 g, 77% yield, 2 steps). m.p. 148-149 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 10.92 (d, J = 2.5 Hz, 1H), 7.71-7.64 (m, 3H), 7.50 (q, J = 8.0 Hz, 3H), 7.37 (d, J = 8.1 Hz, 1H), 7.38-7.14 (m, 5H), 7.09-7.05 (m, 1H), 6.96 (t, J = 7.5 Hz, 1H), 5.56 (dd, J = 6.4, 3.0 Hz, 1H), 4.57-4.51 (m, 1H), 3.33-3.22 (m, 1H), 3.15-3.02 (m, 2H), 2.63-2.53 (m, 2H), 2.05-1.99 (m, 1H), 1.86-1.77 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 191.1, 167.0, 137.1, 136.8, 135.7, 135.0, 133.5, 130.4, 130.3, 129.5, 129.4, 128.7, 128.2, 127.7, 127.0, 123.8, 123.5, 121.5, 118.8, 118.7, 111.9, 111.0, 45.4, 26.6, 23.5, 22.5; HRMS (ESI) m/z $[M+H]^+$ Calcd. For $C_{28}H_{25}N_2O_2$ 421.1911; Found 421.1910.



N-(3,4-dihydronaphthalen-1-yl)-N-(3,5-dimethoxyphenethyl)-2-oxo-2-phenylacet amide (1p)

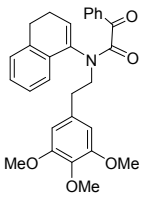
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1p** was obtained as a white solid (1.32 g, 60% yield, 2 steps). m.p. 168-169 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.68-7.66 (m, 3H), 7.49 (t, J = 7.7 Hz, 2H), 7.27-7.20 (m, 2H), 7.16 (d, J

= 7.0 Hz, 2H), 6.48 (d, *J* = 2.2 Hz, 2H), 6.42 (d, *J* = 2.2 Hz, 1H), 5.54 (dd, *J* = 6.4, 2.9 Hz, 1H), 4.56-4.50 (m, 1H), 3.73 (s, 6H), 3.26-3.21 (m, 1H), 2.94-2.83 (m, 2H), 2.71-2.64 (m, 1H), 2.59-2.54 (m, 1H), 2.11-2.04 (m, 1H), 1.87-1.79 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 167.0, 160.9, 141.0, 137.2, 135.7, 135.1, 130.5, 130.1, 129.5, 129.4, 128.7, 128.2, 127.0, 123.4, 107.5, 98.8, 55.6, 45.3, 33.7, 26.6, 22.5; HRMS (ESI) m/z $[M+Na]^+$ Calcd. For $C_{28}H_{27}NO_4Na$ 464.1832; Found 464.1833.



N-(2-(benzo[d][1,3]dioxol-5-yl)ethyl)-N-(3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenylacetamide (1q)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1q** was obtained as a white solid (1.42 g, 67% yield, 2 steps). m.p. 151-151 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.72-7.66 (m, 3H), 7.52-7.49 (m, 2H), 7.27-7.20 (m, 2H), 7.19-7.12 (m, 2H), 6.91 (d, *J* = 1.6 Hz, 1H), 6.88 (d, *J* = 7.8 Hz, 1H), 6.75 (dd, *J* = 7.9, 1.7 Hz, 1H), 6.00 (s, 2H), 5.57 (dd, *J* = 6.4, 3.0 Hz, 1H), 4.50-4.45 (m, 1H), 3.21-3.15 (m, 1H), 2.91-2.81 (m, 2H), 2.73-2.65 (m, 1H), 2.60-2.55 (m, 1H), 2.14-2.07 (m, 1H), 1.88-1.80 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.9, 167.0, 147.8, 146.3, 137.2, 135.7, 135.1, 133.5, 132.5, 130.5, 130.2, 129.5, 129.4, 128.7, 128.2, 127.0, 123.4, 122.6, 109.9, 108.5, 101.3, 45.9, 33.2, 26.6, 22.5; HRMS (ESI) m/z $[M+H]^+$ Calcd. For $C_{27}H_{24}NO_4$ 426.1700; Found 426.1700.

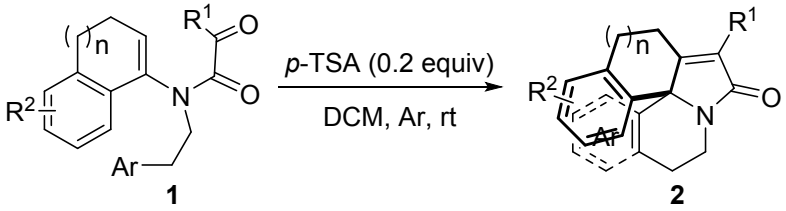


N-(3,4-dihydronaphthalen-1-yl)-2-oxo-2-phenyl-N-(3,4,5-trimethoxyphenethyl)acetamide (1r)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 5 : 1). **1r** was obtained as a white solid (1.46 g, 62% yield, 2 steps). m.p. 132-133 ^{o}C; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.70-7.64 (m, 3H), 7.51-7.45 (m, 2H), 7.27-7.19 (m, 2H), 7.15 (dd, J = 7.2, 1.7 Hz, 2H), 6.61 (s, 2H), 5.51 (dd, J = 6.4, 3.0 Hz, 1H), 4.62-4.48 (m, 1H), 3.74 (s, 6H), 3.65 (s, 3H), 3.27 (ddd, J = 13.4, 7.2, 6.0 Hz, 1H), 2.98-2.81 (m, 2H), 2.70-2.52 (m, 2H), 2.12-2.01 (m 1H), 1.87-1.77 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 190.4, 166.6, 152.8, 136.6, 136.1, 135.3, 134.6, 133.9, 133.1, 129.9, 129.8, 129.0, 128.9, 128.2, 127.7, 126.6, 123.0, 106.3, 60.0, 55.8, 45.0, 33.3, 26.1, 22.0, 21.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. For $C_{29}H_{30}NO_5$ 472.2118; Found 472.2119.

3. Scope of the Reactions

3.1 General Procedure for the Synthesis of 2



To a flask (10 mL) equipped with a magnetic stirrer was added **1** (1.0 mmol), *p*-TSA catalyst (20 mol%) and dry CH_2Cl_2 (10 mL) under argon protection. The reaction mixture was stirred at room temperature for 0.5 h and then saturated $NaHCO_3$ aqueous solution (20 mL) was added to quench the reaction. The mixture was extracted with DCM (3 × 20 mL), and combined organic layer was washed with brine (2 × 40 mL) and dried over anhydrous Na_2SO_4. The solvents were removed in *vacuo* and the residue was purified by flash column chromatography (PE : EA = 3 : 1) to afford products **2**.

3.2. Characterization of 2

13,14-dimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2a)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2a** was obtained as a white solid (419.0 mg, 99% yield). m.p. 182-183 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.47-7.41 (m, 3H), 7.39 (m, 2H), 7.34-7.29 (m, 1H), 7.22-7.14 (m, 3H), 6.67 (s, 1H), 6.63 (s, 1H), 4.76 (ddd, *J* = 13.3, 5.9, 1.9 Hz, 1H), 3.84 (s, 3H), 3.77 (ddd, *J* = 13.3, 11.5, 3.6 Hz, 1H), 3.71 (s, 3H), 3.40-3.26 (m, 2H), 3.20-3.06 (m, 2H), 3.06-2.97 (m, 1H), 2.75 (ddd, *J* = 15.9, 3.7, 2.0 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.3, 159.1, 148.1, 147.5, 141.7, 134.9, 131.1, 130.5, 130.0, 129.4, 128.7, 128.3, 128.0, 127.74, 127.71, 127.3, 127.0, 112.0, 111.8, 67.9, 56.2, 55.9, 40.4, 32.1, 29.6, 23.0; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{26}NO_3$ 424.1907; Found 424.1900.

10,13,14-trimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2b)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2b** was obtained as a white solid (444.2 mg, 98% yield). m.p. 195-196 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.44-7.38 (m, 4H), 7.34-7.31 (m, 1H), 7.11 (d, *J* = 8.5 Hz, 1H), 6.94 (d, *J* = 2.6 Hz, 1H), 6.80 (dd, *J* = 8.5, 2.7 Hz, 1H), 6.66 (s, 2H), 4.78 (ddd, *J* = 13.4, 5.8, 1.9 Hz, 1H), 3.86 (s, 3H), 3.77-3.70 (m, 7H), 3.33 (ddd, *J* = 13.4, 7.1, 2.0 Hz, 1H), 3.25 (ddd, *J* = 16.0, 7.1, 1.9 Hz, 1H), 3.16-3.09 (m, 1H), 3.06-2.98 (m, 2H), 2.74 (ddd, *J* = 15.8, 3.6, 1.9 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.4, 159.2, 158.4, 148.1, 147.5, 142.7, 131.1, 130.4, 129.8, 129.6, 129.4, 128.3, 128.0, 127.2, 127.1, 113.7, 113.0, 111.9, 111.8, 67.9, 56.2, 55.9, 55.3, 40.5, 31.2, 29.6, 23.0; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_4$ 454.2013; Found 454.1997.

10-bromo-13,14-dimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2c)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2c** was obtained as a white solid (486.0 mg, 97% yield). m.p. 240-240 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.55 (d, *J* = 2.1 Hz, 1H), 7.43-7.38 (m, 4H), 7.35-7.31 (m, 2H), 7.07

(d, *J* = 8.3 Hz, 1H), 6.68 (s, 1H), 6.59 (s, 1H), 4.78 (ddd, *J* = 13.4, 5.8, 1.9 Hz, 1H), 3.86 (s, 3H), 3.75-3.68 (m, 4H), 3.35 (ddd, *J* = 13.6, 7.3, 1.9 Hz, 1H), 3.26 (ddd, *J* = 16.7, 7.4, 2.0 Hz, 1H), 3.16-3.10 (m, 1H), 3.05-2.96 (m, 2H), 2.75 (ddd, *J* = 16.0, 3.6, 1.9 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.3, 158.2, 148.3, 147.6, 143.7, 133.9, 130.9, 130.8, 130.5, 130.3, 129.4, 129.1, 128.4, 128.2, 127.4, 120.6, 111.9, 111.8, 67.4, 56.3, 55.9, 40.6, 31.4, 29.5, 22.5; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{25}NO_3Br$ 502.1012, 504.0992; Found 502.0998, 504.0978.

10-chloro-13,14-dimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a, 1-a]isoquinolin-4-one (2d)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2d** was obtained as a white solid (443.5 mg, 97% yield). m.p. 215-216 ^{o}C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.43-7.38 (m, 5H), 7.35-7.32 (m, 1H), 7.19 (dd, *J* = 8.3, 2.1 Hz, 1H), 7.13 (d, *J* = 8.4 Hz, 1H), 6.68 (s, 1H), 6.59 (s, 1H), 4.78 (ddd, *J* = 13.4, 5.8, 1.9 Hz, 1H), 3.87 (s, 3H), 3.73-3.68 (m, 4H), 3.38-3.33 (m, 1H), 3.30-3.25 (m, 1H), 3.16-3.10 (m, 1H), 3.07-2.97 (m, 2H), 2.77-2.73 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.3, 158.3, 148.3, 147.6, 143.4, 133.4, 132.6, 130.8, 130.0, 129.4, 129.1, 128.4, 128.2, 128.0, 127.6, 127.4, 111.9, 111.8, 67.5, 56.3, 55.9, 40.5, 31.4, 29.5, 22.6; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{25}NO_3Cl$ 458.1517; Found 458.1510.

13,14-dimethoxy-10-methyl-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a, 1-a]isoquinolin-4-one (2e)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2e** was obtained as a white solid (433.0 mg, 99% yield). m.p. 210-211 ^{o}C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.44-7.38 (m, 4H), 7.34-7.31 (m, 1H), 7.23 (s, 1H), 7.08 (d, *J* = 7.9 Hz, 1H), 7.04-7.02 (m, 1H), 6.68 (s, 1H), 6.65 (s, 1H), 4.79 (ddd, *J* = 13.3, 5.7, 1.7 Hz, 1H), 3.86 (s, 3H), 3.84-3.75 (m, 1H), 3.73 (s, 3H), 3.36-3.32 (m, 1H), 3.30-3.25 (m, 1H), 3.16-2.98 (m, 3H), 2.75 (ddd, *J* = 15.8, 3.5, 1.8 Hz, 1H), 2.27 (s, 3H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.4, 159.3, 148.0, 147.5, 141.5, 136.6, 131.8, 131.1, 130.4, 130.0, 129.4, 128.8, 128.6, 128.3, 128.0, 127.9, 127.3, 112.0, 111.7, 67.9, 56.2,

55.9, 40.6, 31.6, 29.7, 22.8, 21.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_3$ 438.2064; Found 438.2058.

8,13,14-trimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2f)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2f** was obtained as a white solid (439.5 mg, 97% yield). m.p. 223-224 ^oC; ^1H NMR (500 MHz, Chloroform-*d*) δ 7.43-7.37 (m, 4H), 7.34-7.30 (m, 1H), 7.17 (t, *J* = 8.1 Hz, 1H), 7.06 (d, *J* = 8.0 Hz, 1H), 6.76 (d, *J* = 8.0 Hz, 1H), 6.66 (d, *J* = 2.5 Hz, 2H), 4.79 (ddd, *J* = 13.4, 5.8, 1.6 Hz, 1H), 3.86 (s, 3H), 3.84 (s, 3H), 3.77-3.71 (m, 4H), 3.39-3.32 (m, 2H), 3.10-3.06 (m, 1H), 3.04-2.96 (m, 1H), 2.93-2.85 (m, 1H), 2.73-2.69 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.5, 159.3, 157.0, 148.1, 147.5, 143.1, 131.1, 130.1, 129.7, 129.4, 128.3, 127.9, 127.6, 127.4, 124.3, 119.3, 111.9, 111.7, 108.4, 67.6, 56.3, 55.9, 55.4, 40.6, 29.8, 26.1, 21.8; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_4$ 454.2013; Found 454.2009.

9,13,14-trimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2g)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2g** was obtained as a white solid (444.0 mg, 98% yield). m.p. 203-204 ^oC; ^1H NMR (500 MHz, Chloroform-*d*) δ 7.44-7.38 (m, 4H), 7.35-7.31 (m, 2H), 6.75 (dd, *J* = 8.8, 2.8 Hz, 1H), 6.68 (d, *J* = 2.7 Hz, 1H), 6.66 (s, 1H), 6.63 (s, 1H), 4.75 (ddd, *J* = 13.4, 5.8, 1.8 Hz, 1H), 3.86 (s, 3H), 3.78 (s, 3H), 3.75-3.69 (m, 4H), 3.37-3.32 (m, 1H), 3.27-3.22 (m, 1H), 3.16-2.98 (m, 3H), 2.72 (ddd, *J* = 15.9, 3.6, 1.8 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.2, 159.2, 158.7, 148.0, 147.5, 136.4, 134.2, 131.1, 130.5, 130.3, 129.4, 129.0, 128.3, 128.0, 127.2, 113.5, 112.8, 112.0, 111.7, 67.6, 56.2, 55.9, 55.3, 40.2, 32.4, 29.6, 23.0; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_4$ 454.2013; Found 454.2009.

10,11-dimethoxy-4-phenyl-2,3,7,8-tetrahydrobenzo[6',7']cyclohepta[1',2':2,3]pyrrolo[2,1-a]isoquinolin-5(1H)-one (2i)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2i** was obtained as a white solid (432.7 mg, 99% yield). m.p. 195-196 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.50-7.47 (m, 2H), 7.42-7.39 (m, 2H), 7.35-7.32 (m, 1H), 7.24-7.21 (m, 1H), 7.14-7.11 (m, 2H), 6.93 (dd, *J* = 7.8, 1.4 Hz, 1H), 6.77 (s, 1H), 6.68 (s, 1H), 4.36-4.31 (m, 1H), 3.89 (s, 3H), 3.75 (s, 3H), 3.41-3.36 (m, 1H), 3.26-3.15 (m, 2H), 2.96-2.84 (m, 2H), 2.71-2.63 (m, 1H), 2.48 (dt, *J* = 14.0, 3.6 Hz, 1H), 1.85-1.79 (m, 2H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 172.0, 161.1, 148.3, 147.4, 140.2, 138.0, 132.6, 132.2, 131.8, 130.6, 128.7, 128.6, 128.5, 128.0, 127.2, 126.6, 111.8, 110.4, 73.3, 56.1, 55.8, 35.8, 31.5, 28.4, 25.1, 24.12; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_3$ 438.2064; Found 438.2068.

5-(4-bromophenyl)-13,14-dimethoxy-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2j)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2j** was obtained as a white solid (481.0 mg, 96% yield). m.p. 226-227 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.53 (d, *J* = 8.3 Hz, 2H), 7.46-7.40 (m, 1H), 7.32 (d, *J* = 8.4 Hz, 2H), 7.24-7.15 (m, 3H), 6.66 (s, 1H), 6.61 (s, 1H), 4.75 (ddd, *J* = 13.4, 5.8, 1.9 Hz, 1H), 3.85 (s, 3H), 3.76 (ddd, *J* = 13.2, 11.4, 3.7 Hz, 1H), 3.71 (s, 3H), 3.36-3.26 (m, 2H), 3.19-3.06 (m, 2H), 3.05-2.97 (m, 1H), 2.75 (ddd, *J* = 15.8, 3.6, 1.9 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 172.9, 159.6, 148.3, 147.6, 141.6, 134.8, 131.7, 131.1, 130.1, 129.8, 129.6, 128.8, 127.9, 127.8, 127.3, 127.2, 122.4, 112.0, 111.9, 77.4, 68.1, 56.3, 56.0, 40.5, 32.1, 29.7, 23.1; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{25}NO_3Br$ 502.1012, 504.0992; Found 502.1004, 504.0984.

13,14-dimethoxy-5-(4-methoxyphenyl)-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2k)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2k** was obtained as a white solid (448.6 mg, 99% yield). m.p. 200-201 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.44-7.42 (m, 1H), 7.41-7.38 (m, 2H), 7.23-7.16 (m, 3H), 6.95-6.92

(m, 2H), 6.66 (s, 1H), 6.63 (s, 1H), 4.75 (ddd, *J* = 13.3, 5.9, 2.1 Hz, 1H), 3.85 (s, 3H), 3.82 (s, 3H), 3.79-3.72 (m, 4H), 3.39-3.27 (m, 2H), 3.17-3.06 (m, 2H), 3.04-2.98 (m, 1H), 2.77-2.72 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.6, 159.4, 157.6, 148.0, 147.5, 141.7, 135.0, 130.7, 130.2, 130.0, 128.7, 127.7, 127.6, 127.0, 123.5, 113.9, 112.0, 111.7, 67.8, 56.2, 55.9, 55.3, 40.4, 32.0, 29.6, 23.0; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_4$ 454.2013; Found: 454.1999.

13,14-dimethoxy-5-(p-tolyl)-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2l)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2l** was obtained as a white solid (428.4 mg, 98% yield). m.p. 198-198 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.47-7.40 (m, 1H), 7.36-7.32 (m, 2H), 7.24-7.15 (m, 5H), 6.66 (s, 1H), 6.62 (s, 1H), 4.76 (ddd, *J* = 13.2, 5.9, 2.0 Hz, 1H), 3.85 (s, 3H), 3.81-3.73 (m, 1H), 3.71 (s, 3H), 3.38-3.33 (m, 1H), 3.33-3.26 (m, 1H), 3.17-3.06 (m, 2H), 3.06-2.98 (m, 1H), 2.78-2.70 (m, 1H), 2.36 (s, 3H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.6, 158.5, 158.4, 148.2, 147.6, 141.9, 138.0, 135.1, 130.6, 130.2, 129.4, 129.2, 128.8, 128.2, 127.9, 127.8, 127.4, 127.1, 112.1, 111.9, 67.9, 56.3, 56.0, 40.5, 32.1, 29.7, 23.1, 21.5; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_3$ 438.2064; Found 438.2052.

5-(3-bromophenyl)-13,14-dimethoxy-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2m)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2m** was obtained as a white solid (486.0 mg, 97% yield). m.p. 184-185 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.57 (t, *J* = 1.8 Hz, 1H), 7.49-7.41 (m, 2H), 7.41-7.35 (m, 1H), 7.28 (d, *J* = 7.9 Hz, 1H), 7.25-7.15 (m, 3H), 6.67 (s, 1H), 6.61 (s, 1H), 4.75 (ddd, *J* = 13.3, 5.8, 2.0 Hz, 1H), 3.86 (s, 3H), 3.77 (ddd, *J* = 13.2, 11.5, 3.7 Hz, 1H), 3.72 (s, 3H), 3.39-3.28 (m, 2H), 3.22-3.06 (m, 2H), 3.01 (ddd, *J* = 16.6, 11.4, 5.8 Hz, 1H), 2.75 (ddd, *J* = 15.9, 3.7, 2.0 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 172.8, 160.3, 148.3, 147.7, 141.6, 134.8, 133.3, 132.3, 131.2, 130.1, 129.8, 129.4, 128.8, 128.2, 127.9, 127.8, 127.4, 127.2, 122.5, 112.0, 111.9, 68.1, 56.3, 56.0, 40.5, 32.2,

29.7, 23.2; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{25}NO_3Br$ 502.1012, 504.0992; Found 502.1000, 504.0980.

5-(tert-butyl)-13,14-dimethoxy-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2n)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2n** was obtained as a white solid (387.0 mg, 96% yield). m.p. 172-173 ^oC; ^1H NMR (500 MHz, Chloroform-*d*) δ 7.35 (d, *J* = 7.7 Hz, 1H), 7.21-7.13 (m, 3H), 6.64 (s, 1H), 6.58 (s, 1H), 4.57 (ddd, *J* = 13.0, 5.6, 3.4 Hz, 1H), 3.84 (s, 3H), 3.70 (s, 3H), 3.67-3.62 (m, 1H), 3.53-3.49 (m, 1H), 3.35-3.29 (m, 1H), 3.14-3.06 (m, 2H), 2.92-2.86 (m, 1H), 2.77-2.73 (m, 1H), 1.36 (s, 9H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 174.5, 155.0, 148.0, 147.1, 142.3, 136.3, 135.1, 130.4, 128.3, 128.1, 127.4, 127.2, 126.8, 112.5, 111.6, 68.2, 56.3, 55.8, 40.9, 34.2, 32.4, 29.8, 29.7, 23.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{26}H_{30}NO_3$ 404.2220; Found 404.2213.

12,14-dimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2p)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2p** was obtained as a white solid (419.0 mg, 99% yield). m.p. 212-213 ^oC; ^1H NMR (500 MHz, Chloroform-*d*) δ 7.45-7.37 (m, 5H), 7.34-7.30 (m, 1H), 7.18-7.09 (m, 3H), 6.37 (d, *J* = 2.5 Hz, 1H), 6.29 (d, *J* = 2.5 Hz, 1H), 4.66-4.62 (m, 1H), 3.78 (s, 3H), 3.70-3.64 (m, 1H), 3.49-3.43 (m, 1H), 3.28 (s, 3H), 3.21-3.16 (m, 1H), 3.14-3.07 (m, 2H),2.99-2.93 (m, 1H), 2.76-2.72 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 172.5, 162.3, 159.4, 159.2, 141.6, 138.2, 136.8, 131.7, 131.4, 129.6, 128.3, 128.2, 127.8, 126.9, 126.8, 125.7, 120.4, 106.4, 99.6, 67.5, 55.5, 55.3, 38.6, 33.6, 31.1, 24.8; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{26}NO_3$ 424.1907; Found 424.1906.

3-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a][1,3]dioxolo[4,5-g]isoquinolin-4-one (2q)

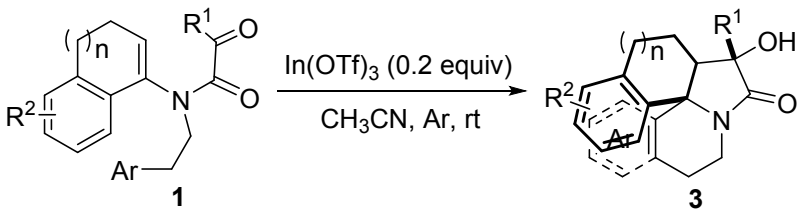
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2q** was obtained

as a white solid (387.0 mg, 95% yield). m.p. 196-197 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.45-7.39 (m, 5H), 7.35-7.31 (m, 1H), 7.24-7.17 (m, 3H), 6.65 (s, 1H), 6.61 (s, 1H), 5.90-5.89 (m, 2H), 4.75-4.70 (m, 1H), 3.78-3.72 (m, 1H), 3.36-3.25 (m, 2H), 3.16-3.00 (m, 2H), 2.99-2.95 (m, 1H), 2.74 (dt, *J* = 15.8, 2.9 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 173.3, 159.0, 146.5, 146.4, 141.7, 135.1, 131.1, 131.0, 130.7, 129.4, 128.8, 128.4, 128.0, 127.7, 127.6, 127.0, 109.0, 108.5, 101.2, 68.1, 40.4, 32.0, 30.2, 22.7; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{27}H_{22}NO_3$ 408.1594; Found 408.1591.

12,13,14-trimethoxy-5-phenyl-1,2,6,7-tetrahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (2r)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 3 : 1). **2r** was obtained as a white solid (448.5 mg, 99% yield). m.p. 153-154 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.48 (dd, *J* = 7.7, 1.5 Hz, 1H), 7.46-7.42 (m, 2H), 7.41-7.37 (m, 2H), 7.34-7.30 (m, 1H), 7.22-7.12 (m, 3H), 6.50 (s, 1H), 4.67 (ddd, *J* = 13.4, 5.9, 1.4 Hz, 1H), 3.85 (s, 3H), 3.82 (s, 3H), 3.71-3.65 (m, 1H), 3.63-3.57 (m, 1H), 3.19-3.11 (m, 2H), 3.11-2.93 (m, 2H), 2.77 (s, 3H), 2.70 (ddd, *J* = 15.9, 3.4, 1.4 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 171.6, 161.3, 152.0, 151.6, 140.9, 140.3, 135.8, 130.4, 130.3, 128.5, 127.8, 127.2, 126.8, 126.2, 126.1, 124.8, 123.2, 106.9 66.5, 59.5, 57.7, 54.8, 38.0, 32.6, 29.8, 23.1; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{28}NO_4$ 454.2013; Found 454.2015.

3.3. General Procedure for the Synthesis of 3



To a flask (10 mL) equipped with a magnetic stirrer was added **1** (1.0 mmol), $In(OTf)_3$ catalyst (20 mol%) and dry CH_3CN (10 mL) under argon protection. The reaction mixture was stirred at room temperature for 0.5 h and then saturated $NaHCO_3$

aqueous solution (20 mL) was added to quench the reaction. The mixture was extracted with DCM (3 × 20 mL), and combined organic layer was washed with brine (2 × 40 mL) and dried over anhydrous Na_2SO_4. The solvents were removed in *vacuo* and the residue was purified by flash column chromatography (PE : EA = 2 : 1) to afford products **3**.

3.4. Characterization of 3

5-hydroxy-13,14-dimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3a)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3a** was obtained as a white solid (410.5 mg, 93% yield). m.p. 251-252 ^oC; ^1H NMR (500 MHz, DMSO-d_6) δ 7.27-7.21 (m, 5H), 7.11-7.05 (m, 2H), 6.94-6.88 (m, 2H), 6.78 (s, 1H), 6.38 (s, 1H), 5.97 (s, 1H), 4.26 (dd, J = 13.1, 6.2 Hz, 1H), 3.76 (s, 3H), 3.55 (s, 3H), 3.28-3.23 (m, 1H), 2.97-2.84 (m, 2H), 2.77 (dd, J = 16.0, 4.0 Hz, 1H), 2.40-2.32 (m, 1H), 1.93-1.78 (m, 2H), 1.74-1.65 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 174.8, 147.5, 147.4, 140.8, 139.5, 137.9, 133.7, 129.2, 128.2, 127.4, 127.4, 127.0, 127.0, 125.8, 125.5, 111.8, 110.5, 80.9, 62.0, 55.6, 55.4, 53.6, 36.3, 27.2, 24.8, 22.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{28}NO_4$ 442.2013; Found 442.2012.

5-hydroxy-10,13,14-trimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3b)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3b** was obtained as a white solid (367.6 mg, 78% yield). m.p. 228-229 ^oC; ^1H NMR (500 MHz, DMSO-d_6) δ 7.30-7.20 (m, 5H), 6.83 (d, J = 8.3 Hz, 1H), 6.79 (s, 1H), 6.73 (dd, J = 8.3, 2.6 Hz, 1H), 6.41-6.37 (m, 2H), 5.96 (d, J = 2.3 Hz, 1H), 4.27 (dd, J = 13.0, 6.0 Hz, 1H), 3.77 (s, 3H), 3.61 (s, 3H), 3.57 (s, 3H), 3.28-3.23 (m, 1H), 2.98-2.76 (m, 3H), 2.33-2.25 (m, 1H), 1.92-1.82 (m, 1H), 1.79-1.64 (m, 2H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 175.3, 157.5, 148.0, 147.8, 141.2, 141.0, 133.9, 130.5, 129.6, 127.9, 127.5, 125.9, 115.6, 112.6, 112.3, 110.9, 81.3, 62.5, 56.1, 55.9, 55.4, 53.9, 36.9, 27.6,

24.4, 23.0; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{30}NO_5$ 472.2118; Found 472.2110.

10-bromo-5-hydroxy-13,14-dimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3c)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3c** was obtained as a white solid (462.0 mg, 89% yield). m.p. 230-231 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.35-7.27 (m, 2H), 7.25-7.18 (m, 4H), 7.13 (d, *J* = 2.0 Hz, 1H), 6.72 (d, *J* = 8.1 Hz, 1H), 6.66 (s, 1H), 6.31 (s, 1H), 4.55 (dd, *J* = 13.2, 6.0 Hz, 1H), 3.90 (s, 3H), 3.70 (s, 3H), 3.41-3.35 (m, 1H), 3.17-3.10 (m, 1H), 2.96 (dd, *J* = 5.9, 2.5 Hz, 1H), 2.78 (dd, *J* = 16.1, 3.8 Hz, 1H), 2.35-2.29 (m, 1H), 2.04-1.94 (m, 2H), 1.79-1.72 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.7, 148.2, 148.0, 141.4, 139.5, 136.9, 132.5, 131.9, 130.5, 130.1, 128.2, 128.1, 126.9, 125.5, 119.6, 111.6, 110.2, 81.6, 62.7, 56.0, 55.9, 53.4, 37.6, 28.0, 24.5, 21.8; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{27}NO_4Br$ 520.1118, 522.1098; Found 520.1108, 522.1086.

10-chloro-5-hydroxy-13,14-dimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3d)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3d** was obtained as a white solid (408.5 mg, 86% yield). m.p. 218-219 °C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.34-7.21 (m, 5H), 7.18 (dd, *J* = 8.2, 2.3 Hz, 1H), 6.94 (d, *J* = 8.2 Hz, 1H), 6.89 (d, *J* = 2.2 Hz, 1H), 6.82 (s, 1H), 6.37 (s, 1H), 6.03 (br, 1H), 4.29 (dd, *J* = 13.2, 6.0 Hz, 1H), 3.78 (s, 3H), 3.58 (s, 3H), 3.27-3.21 (m, 1H), 2.97-2.75 (m, 3H), 2.45-2.34 (m, 1H), 1.96-1.84 (m, 1H), 1.77-1.69 (m, 2H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 175.5, 148.1, 147.9, 142.3, 141.0, 137.5, 133.2, 130.64, 130.59, 128.8, 128.0, 127.6, 127.4, 126.0, 112.4, 110.9, 81.2, 62.2, 56.1, 55.9, 53.9, 37.0, 27.6, 24.7, 22.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{27}NO_4Cl$ 476.1623; Found 476.1613.

5-hydroxy-13,14-dimethoxy-10-methyl-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3e)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3e** was obtained as a white solid (318.6 mg, 70% yield). m.p. 223-224 ^{o}C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.32 (d, *J* = 7.3 Hz, 2H), 7.27-7.20 (m, 3H), 6.90 (dd, *J* = 7.7, 1.7 Hz, 1H), 6.77 (d, *J* = 1.7 Hz, 1H), 6.74 (d, *J* = 7.7 Hz, 1H), 6.65 (s, 1H), 6.38 (s, 1H), 4.50 (ddd, *J* = 13.1, 6.3, 1.1 Hz, 1H), 3.89 (s, 3H), 3.70 (s, 3H), 3.42 (td, *J* = 12.7, 4.1 Hz, 1H), 3.18-3.12 (m, 1H), 3.00 (s, 1H), 2.98 (dd, *J* = 6.5, 2.1 Hz, 1H), 2.76 (dd, *J* = 16.1, 3.9 Hz, 1H), 2.34-2.29 (m, 1H), 2.20 (s, 3H), 2.03-1.88 (m, 2H), 1.85-1.78 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.6, 147.9, 147.8, 139.9, 138.9, 135.4, 134.9, 133.6, 129.6, 128.31, 128.27, 128.04, 127.97, 127.0, 125.5, 111.3, 110.3, 81.9, 63.0, 55.9, 55.8, 53.6, 37.4, 28.0, 24.6, 22.5, 21.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{30}NO_4$ 456.2169; Found 456.2159.

5-hydroxy-8,13,14-trimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3f)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3f** was obtained as a white solid (340.0 mg, 72% yield). m.p. 210-212 ^{o}C; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.26-7.18 (m, 3H), 7.18-7.10 (m, 2H), 7.04 (t, *J* = 8.0 Hz, 1H), 6.72 (dd, *J* = 8.0, 1.0 Hz, 1H), 6.67-6.60 (m, 2H), 6.37 (s, 1H), 4.60 (ddd, *J* = 13.2, 6.3, 1.3 Hz, 1H), 3.87 (s, 3H), 3.68 (s, 3H), 3.66 (s, 3H), 3.51 (td, *J* = 12.7, 4.0 Hz, 1H), 3.12 (ddd, *J* = 16.0, 12.2, 6.1 Hz, 1H), 2.92-2.86 (m, 1H), 2.82-2.72 (m, 1H), 2.47 (ddd, *J* = 18.1, 5.6, 3.1 Hz, 1H), 2.12-1.98 (m, 2H), 1.46-1.32 (m, 1H); ^{13}C NMR NMR (101 MHz, Chloroform-*d*) δ 175.9, 156.4, 148.0, 147.9, 140.3, 139.8, 133.6, 128.0, 127.8, 127.0, 126.9, 126.3, 125.4, 120.9, 111.5, 110.6, 108.5, 81.7, 62.6, 56.1, 55.9, 55.5, 53.7, 37.9, 28.5, 20.3, 18.2; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{30}NO_5$ 472.2118; Found 472.2109.

5-hydroxy-12,13-dimethoxy-5-phenyl-1,2,5a,6-tetrahydroindeno[1',2':2,3]pyrrolo[2,1-a]isoquinolin-4(5H)-one (3h)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3h** was obtained

as a white solid (397.2 mg, 93% yield). m.p. 271-272 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.25-7.20 (m, 5H), 7.18-7.12 (m, 2H), 7.07 (dd, *J* = 7.0, 1.8 Hz, 1H), 6.93 (dd, *J* = 7.3, 1.7 Hz, 1H), 6.64 (s, 1H), 6.29 (s, 1H), 4.48 (ddd, *J* = 13.1, 6.4, 1.1 Hz, 1H), 3.87 (s, 3H), 3.67 (s, 3H), 3.59-3.53 (m, 1H), 3.46 (dd, *J* = 9.3, 4.8 Hz, 1H), 3.23-3.16 (m, 1H), 3.06 (dd, *J* = 17.2, 9.3 Hz, 1H), 2.79 (dd, *J* = 16.2, 4.1 Hz, 1H), 2.60 (dd, *J* = 17.2, 4.8 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 174.3, 148.19, 148.17, 146.1, 141.9, 140.0, 131.6, 128.8, 127.93, 127.90, 127.3, 126.7, 125.2, 124.7, 124.6, 111.3, 109.0, 82.6, 73.4, 58.4, 56.0, 55.9, 36.8, 33.8, 27.6; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{27}H_{26}NO_4$ 428.1856; Found 428.1852.

5-(4-bromophenyl)-5-hydroxy-13,14-dimethoxy-1,2,5,5a,6,7-hexahydro-4H-benzo [6,7]indolo[7a,1-a]isoquinolin-4-one (3j)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3j** was obtained as a white solid (472.4 mg, 91% yield). m.p. 262-263 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.37 (d, *J* = 8.2 Hz, 2H), 7.22 (d, *J* = 8.1 Hz, 2H), 7.14-7.11 (m, 1H), 7.08-7.05 (m, 1H), 6.96-6.91 (m, 2H), 6.64 (s, 1H), 6.36 (s, 1H), 4.43 (dd, *J* = 13.1, 6.0 Hz, 1H), 3.89 (s, 3H), 3.70 (s, 3H), 3.38-3.32 (m, 1H), 3.15-3.09 (m, 1H), 3.03-3.01 (m, 1H), 2.73 (dd, *J* = 16.2, 3.9 Hz, 1H), 2.45-2.37 (m, 1H), 2.02-1.85 (m, 3H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.1, 148.0, 147.9, 139.1, 138.9, 138.1, 133.2, 131.1, 129.4, 128.9, 128.5, 127.5, 126.1, 125.6, 122.1, 111.3, 110.1, 81.6, 63.3, 55.93, 55.88, 53.4, 37.2, 27.7, 25.4, 23.0; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{27}NO_4Br$ 520.1118; Found 520.1110.

5-hydroxy-13,14-dimethoxy-5-(p-tolyl)-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]ind olo[7a,1-a]isoquinolin-4-one (3l)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3l** was obtained as a white solid (400.0 mg, 88% yield). m.p. 248-249 °C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.23 (d, *J* = 7.7 Hz, 2H), 7.14-7.02 (m, 4H), 6.94 (dd, *J* = 7.8, 1.4 Hz, 1H), 6.90 (dd, *J* = 7.5, 1.4 Hz, 1H), 6.64 (s, 1H), 6.38 (s, 1H), 4.44 (dd, *J* = 13.1, 5.8 Hz, 1H), 3.89 (s, 3H), 3.70 (s, 3H), 3.34 (t, *J* = 11.9 Hz, 1H), 3.24-3.13 (m, 1H),

3.09-3.01 (m, 1H), 2.73 (dd, *J* = 16.5, 3.5 Hz, 1H), 2.38-2.33 (s, 4H), 2.03-1.90 (m, 3H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.7, 148.0, 147.9, 139.1, 138.4, 137.8, 136.9, 133.6, 129.5, 128.7, 128.4, 127.3, 126.9, 126.0, 125.6, 111.3, 110.1, 82.0, 63.2, 55.94, 55.85, 53.3, 37.0, 27.8, 25.4, 23.3, 21.2; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{30}NO_4$ 456.2169; Found 456.2159.

5-(3-bromophenyl)-5-hydroxy-13,14-dimethoxy-1,2,5,5a,6,7-hexahydro-4H-benzo [6,7]indolo[7a,1-a]isoquinolin-4-one (3m)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3m** was obtained as a white solid (477.0 mg, 92% yield). m.p. 257-258 ^{o}C; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.49 (s, 1H), 7.39 (dd, *J* = 8.0, 1.8 Hz, 1H), 7.26-7.18 (m, 1H), 7.16-7.04 (m, 3H), 6.99 (dd, *J* = 7.8, 1.4 Hz, 1H), 6.90 (dd, *J* = 7.5, 1.5 Hz, 1H), 6.65 (s, 1H), 6.36 (s, 1H), 4.48 (dd, *J* = 13.1, 6.0 Hz, 1H), 3.89 (s, 3H), 3.69 (s, 3H), 3.42-3.36 (m, 1H), 3.17-3.10 (m, 1H), 3.04-2.96 (m, 1H), 2.75 (dd, *J* = 16.1, 3.9 Hz, 1H), 2.50-2.39 (m, 1H), 2.07-1.89 (m, 3H), 1.63 (br, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.0, 148.0, 147.9, 142.2, 138.8, 137.8, 133.1, 131.2, 130.3, 129.4, 129.2, 128.4, 127.6, 126.3, 125.7, 125.5, 122.3, 111.4, 110.3, 81.4, 63.1, 56.0, 55.9, 53.6, 37.5, 28.0, 25.1, 22.3; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{27}NO_4Br$ 520.1118, 522.1098; Found 520.1110, 522.1087.

8-hydroxy-8-phenyl-4,5,8,8a,9,10-hexahydro-1H,7H-benzo[g]indolo[2',3':3,4]pyri do[2,1-i]indol-7-one (3o)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3o** was obtained as a white solid (418.0 mg, 99% yield). m.p. >300 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 10.72 (s, 1H), 7.49 (d, *J* = 7.7 Hz, 1H), 7.29-7.19 (m, 6H), 7.14-7.04 (m, 4H), 7.01 (t, *J* = 7.4 Hz, 1H), 6.91 (d, *J* = 7.4 Hz, 1H), 6.03 (s, 1H), 4.47 (dd, *J* = 13.1, 5.7 Hz, 1H), 3.51-3.45 (m, 1H), 2.99-2.87 (m, 3H), 2.35-2.30 (m, 1H), 2.08-2.00 (m, 1H), 1.80-1.75 (m, 1H), 1.70-1.63 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 175.1, 140.9, 137.95, 137.89, 137.0, 136.3, 128.8, 128.5, 127.5, 127.4, 126.8, 126.1, 125.9,

121.2, 118.6, 118.1, 111.2, 105.7, 80.9, 60.1, 51.4, 36.3, 24.8, 21.5, 20.1; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{27}N_2O_2$ 423.2067; Found 421.1971.

5-hydroxy-12,14-dimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3p)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3p** was obtained as a white solid (423.6 mg, 96% yield). m.p. 241-242 ^oC; ^1H NMR (500 MHz, Chloroform-*d*) δ 7.54 (d, *J* = 7.4 Hz, 2H), 7.41-7.32 (m, 3H), 7.15 (t, *J* = 7.5 Hz, 1H), 7.10-6.93 (m, 2H), 6.72 (d, *J* = 7.8 Hz, 1H), 6.35 (d, *J* = 8.9 Hz, 2H), 4.07 (dd, *J* = 13.1, 6.4 Hz, 1H), 3.83 (s, 3H), 3.56 (s, 3H), 3.35 (d, *J* = 7.3 Hz, 1H), 3.30-3.17 (m, 1H), 2.96 (dt, *J* = 13.2, 6.6 Hz, 1H), 2.68-2.59 (m, 1H), 2.52 (t, *J* = 14.5 Hz, 1H), 2.30 (d, *J* = 15.1 Hz, 1H), 1.73-1.66 (m, 1H), 1.48 (d, *J* = 13.8 Hz, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.3, 159.5, 157.2, 140.6, 139.8, 138.8, 136.6, 128.2, 128.14, 128.08, 128.0, 127.5, 127.2, 125.4, 122.0, 104.8, 98.1, 82.6, 63.5, 55.30, 55.26, 51.3, 35.7, 27.8, 26.8, 26.6; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{28}H_{28}NO_4$ 442.2013; Found 442.2005.

3-hydroxy-3-phenyl-1,2,2a,3,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a][1,3]dioxolo[4,5-g]isoquinolin-4-one (3q)

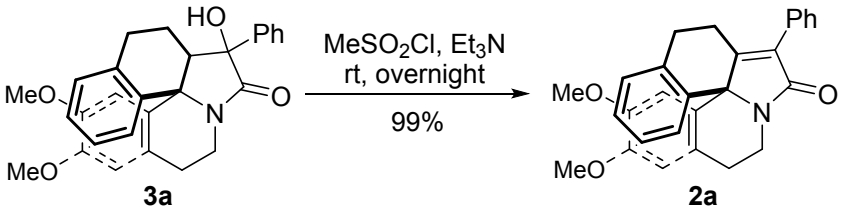
The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3q** was obtained as a white solid (161.5 mg, 38% yield). m.p. 280-281 ^oC; ^1H NMR (500 MHz, Chloroform-*d*) δ 7.30 (d, *J* = 7.4 Hz, 2H), 7.25-7.16 (m, 3H), 7.13-7.01 (m, 3H), 6.83 (dd, *J* = 6.4, 2.4 Hz, 1H), 6.62 (s, 1H), 6.36 (s, 1H), 5.89 (s, 2H), 4.56-4.44 (m, 1H), 3.40 (td, *J* = 12.7, 4.0 Hz, 1H), 3.16-3.04 (m, 2H), 2.95 (dd, *J* = 6.5, 2.4 Hz, 1H), 2.73 (dd, *J* = 16.2, 3.8 Hz, 1H), 2.38-2.32 (m, 1H), 2.06-1.89 (m, 2H), 1.86-1.79 (m, 1H); ^{13}C NMR (126 MHz, Chloroform-*d*) δ 175.7, 146.6, 146.5, 139.7, 139.0, 137.9, 134.7, 129.2, 128.5, 128.1, 128.0, 127.3, 127.0, 126.6, 126.0, 108.6, 107.4, 101.1, 81.8, 63.2, 53.6, 37.3, 28.6, 25.0, 21.9; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{27}H_{24}NO_4$ 426.1700; Found 426.1690.

5-hydroxy-12,13,14-trimethoxy-5-phenyl-1,2,5,5a,6,7-hexahydro-4H-benzo[6,7]indolo[7a,1-a]isoquinolin-4-one (3r)

The title compound was synthesized following the general procedure and the residue was purified by flash chromatography on silica gel (PE : EA = 2 : 1). **3r** was obtained as a white solid (321.2 mg, 77% yield). m.p. 243-245 ^{o}C; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.53 (d, *J* = 7.2 Hz, 2H), 7.42-7.31 (m, 3H), 7.15 (td, *J* = 7.3, 1.3 Hz, 1H), 7.05 (ddd, *J* = 11.2, 6.9, 2.0 Hz, 2H), 6.79 (dd, *J* = 7.8, 1.3 Hz, 1H), 6.47 (s, 1H), 4.19-4.09 (m, 1H), 3.88 (s, 3H), 3.83 (s, 3H), 3.35 (dd, *J* = 7.3, 3.0 Hz, 1H), 3.25 (s, 3H), 3.22-3.04 (m, 2H), 2.78 (s, 1H), 2.67-2.58 (m, 1H), 2.48 (ddd, *J* = 15.8, 12.4, 3.5 Hz, 1H), 2.34 (dt, *J* = 15.4, 4.2 Hz, 1H), 1.82-1.68 (m, 1H), 1.58-1.51 (M, 1H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 175.5, 152.9, 150.6, 141.0, 140.5, 139.6, 139.1, 129.8, 129.0, 128.4, 128.2, 128.1, 127.8, 127.7, 127.5, 125.4, 107.0, 82.6, 63.4, 60.7, 59.4, 55.9, 52.2, 35.8, 27.4, 26.9, 26.4; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{30}NO_5$ 472.2118; Found 472.2109.

4. Mechanistic Studies

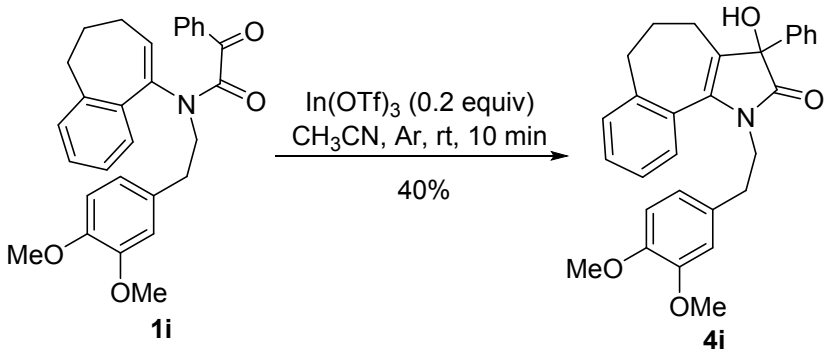
4.1. The *p*-TSA-Catalyzed Reaction Procedure of 3



3a **2a**

To a solution of **3a** (132.4 mg, 0.3 mmol) in 5 ml of CH_2Cl_2, methanesulfonyl chloride (27 μl, 0.36 mmol) and triethylamine (57 μl, 0.42 mmol) were added at 0℃. The solution was stirred at 0℃ for 1 h, and then allowed to warm to rt overnight. Water was added and the mixture was extracted with CH_2Cl_2. The combined organic phases were dried over anhydrous Na_2SO_4 and concentrated in vacuo. Flash chromatography on silica gel (PE : EA = 3 : 1) afforded **2a** as solid (126.0 mg, 99%).

4.2. The Synthesis and Characterization of Compound 4i

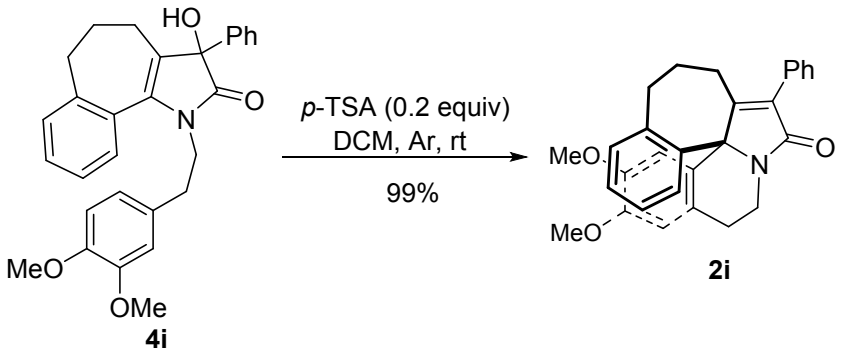


To a flask (10 mL) equipped with a magnetic stirrer was added **1i** (0.5 mmol), $In(OTf)_3$ catalyst (20 mol%) and dry CH_3CN (5 mL) under argon protection. The reaction mixture was stirred at room temperature for 10 min and then saturated $NaHCO_3$ aqueous solution (10 mL) was added to quench the reaction. The mixture was extracted with DCM (3 × 10 mL), and combined organic layer was washed with brine (2 × 20 mL) and dried over anhydrous Na_2SO_4. The solvents were removed in *vacuo* and the residue was purified by flash column chromatography (PE : EA = 2 : 1) to afford products **4i**.

1-(3,4-dimethoxyphenethyl)-3-hydroxy-3-phenyl-3,4,5,6-tetrahydrobenzo[6,7]cyclohepta[1,2-b]pyrrol-2(1H)-one (4i)

White solid (91.0 mg, 40% yield). m.p. 194-195 ^{o}C; ^{1}H NMR (500 MHz, DMSO-d_6) δ 7.45 (d, J = 7.6 Hz, 1H), 7.41-7.35 (m, 1H), 7.35-7.31 (m, 2H), 7.30-7.22 (m, 3H), 7.22-7.17 (m, 2H), 6.76 (d, J = 8.2 Hz, 1H), 6.47 (d, J = 2.1 Hz, 1H), 6.41 (dd, J = 8.1, 2.0 Hz, 1H), 6.30 (s, 1H), 3.94 (dt, J = 14.7, 7.6 Hz, 1H), 3.78 (ddd, J = 13.9, 8.0, 5.6 Hz, 1H), 3.70 (s, 3H), 3.58 (s, 3H), 2.67-2.54 (m, 2H), 2.49-2.34 (m, 2H), 2.04-1.91 (m, 1H), 1.85 (dt, J = 15.7, 7.9 Hz, 1H), 1.78-1.71 (m, 1H), 1.65-1.57 (m, 1H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 179.7, 148.6, 147.4, 142.1, 140.3, 137.0, 130.3, 130.1, 129.6, 128.3, 128.0, 127.2, 126.6, 126.1, 125.1, 124.8, 120.5, 112.1, 111.8, 79.9, 55.5, 55.2, 41.2, 33.47, 33.44, 32.0, 20.2; HRMS (ESI) m/z $[M+H]^+$ Calcd. for $C_{29}H_{29}NO_4$ 456.2169; Found 456.2160.

4.3. The *p*-TSA-Catalyzed Reaction Procedure of 4i



To a flask (10 mL) equipped with a magnetic stirrer was added **4i** (0.5 mmol), *p*-TSA catalyst (20 mol%) and dry CH_2Cl_2 (5 mL) under argon protection. The reaction mixture was stirred at room temperature for 0.5 h and then saturated $NaHCO_3$ aqueous solution (10 mL) was added to quench the reaction. The mixture was extracted with DCM (3 × 10 mL), and combined organic layer was washed with brine (2 × 20 mL) and dried over anhydrous Na_2SO_4. The solvents were removed in *vacuo* and the residue was purified by flash column chromatography (PE : EA = 3 : 1) to afford products **2i**.

5. Crystallographic Data

High quality single crystals of **3a** were cultivated from the evaporation of a solution of **3a** in the mixture of EtOAc and *n*-hexane. As depicted below, the molecular structures were determined by X-ray diffraction analysis.

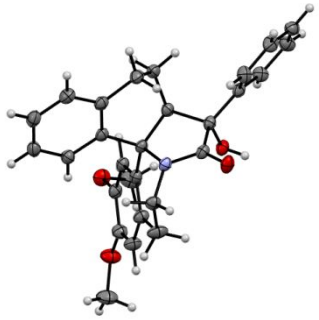


Figure S1. X-ray molecular structure of **3a**. The molecular structure is depicted in an ellipsoid style at 50% probability level.

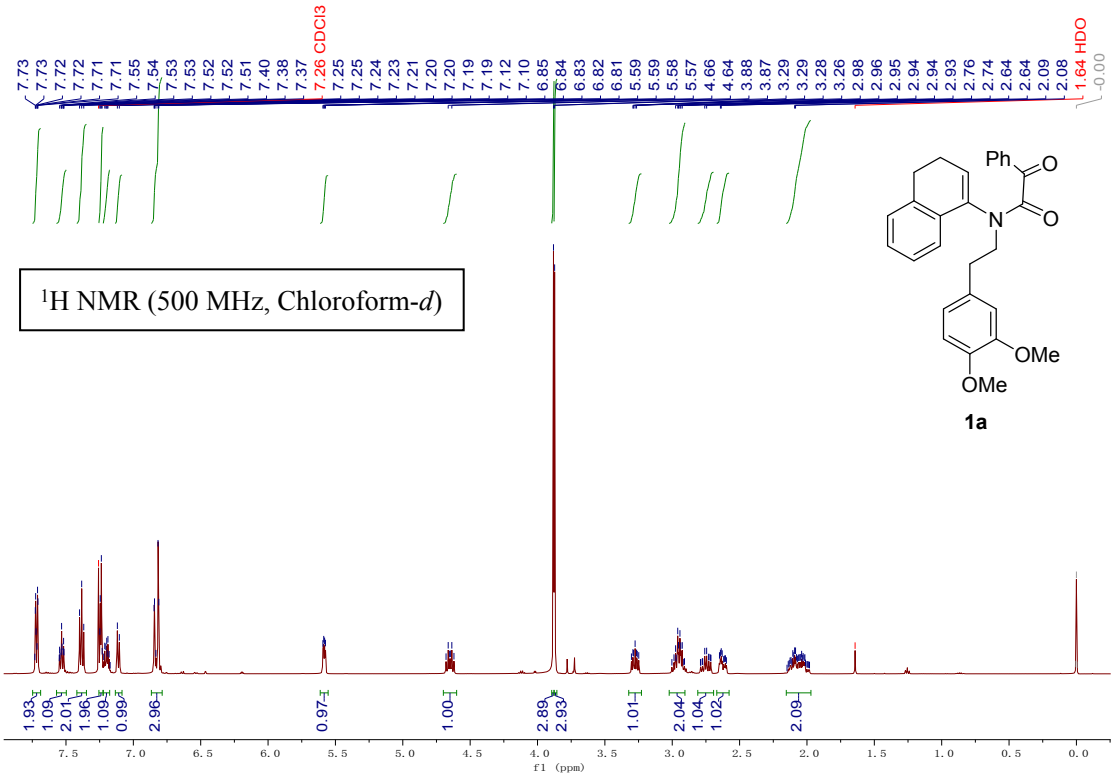
5.1 Crystallographic data and structure refinement of 3a

CCDC Number	2032742
Empirical formula	$C_{28}H_{27}NO_4$
Formula weight	441.19
Temperature	293.00(2) K
Wavelength	1.54184 Å
Crystal system	Triclinic
Space group	P -1
a	7.1879 (10) Å
b	11.8388 (10) Å
c	c = 13.4990 (10) Å
α	105.357 (10)º
β	93.267 (10)°
γ	91.181 (10)º
Volumn	1105.14 (2) $Å^3$
Z	2
Density (calculated)	1.327
Absorption coefficient	0.710 mm^{-1}
F(000)	468.0
Radiation type	CuK\a
Goodness-of-fit on F^2	1.046
Completeness to theta = 74.759°	97.2%
R (reflections)	0.0499 (4230)
wR2 (reflections)	0.1361 (4400)

6. References

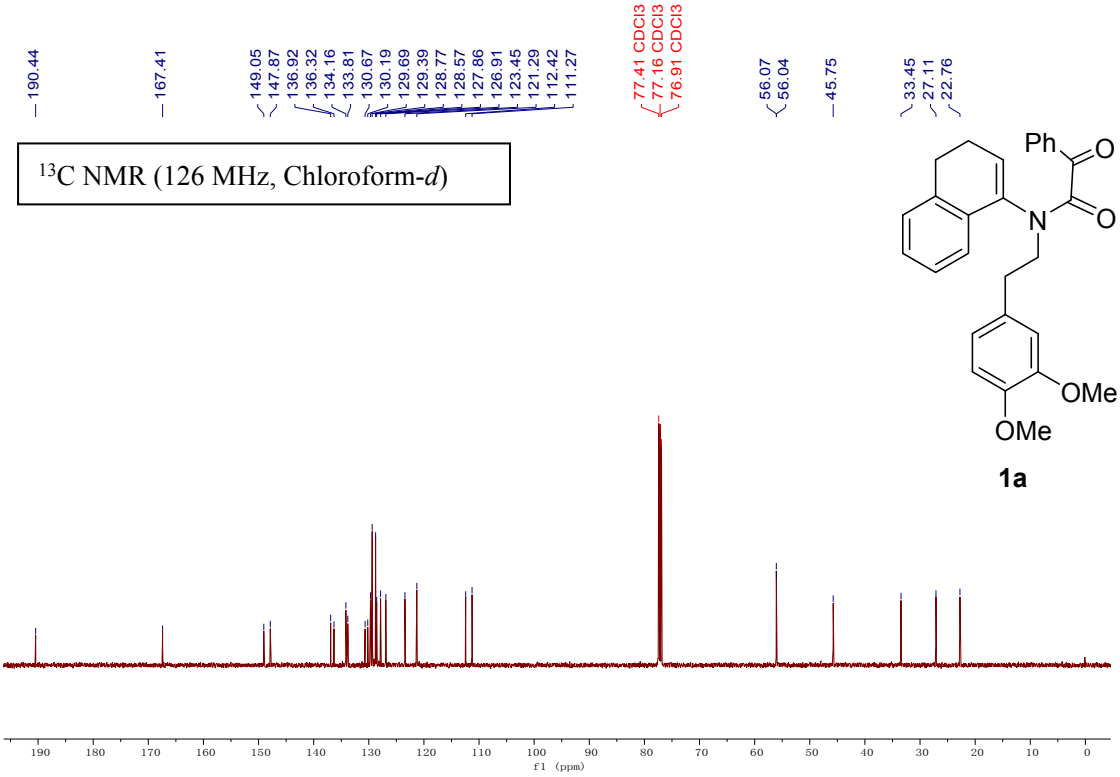
[1] (a) L. Yang, D.-X. Wang, Z.-T. Huang and M.-X. Wang, *J. Am. Chem. Soc.* **2009**, ***131***, 10390. (b) S. Tong, D.-X. Wang, L. Zhao, J. Zhu, M.-X. Wang, *Angew. Chem. Int. Ed.* **2012**, *51*, 4417; *Angew. Chem.* **2012**, *124*, 4493.
[2] (a) L. He, L. Zhao, D.-X. Wang, M.-X. Wang, *Org. Lett.* **2014**, *16*, 5972.(ba) S. Tong, X. Yang, D.-X. Wang, L. Zhao, J. Zhu, M.-X. Wang, *Tetrahedron* **2012**, *68*, 6492. (c) L. Yang, G. Deng, D.-X. Wang, Z.-T. Huang, J. Zhu, M.-X. Wang, *Org. Lett.* **2007**, *9*, 1387.

7. Copies of ^{1}H NMR and ^{13}C NMR Spectra of Products



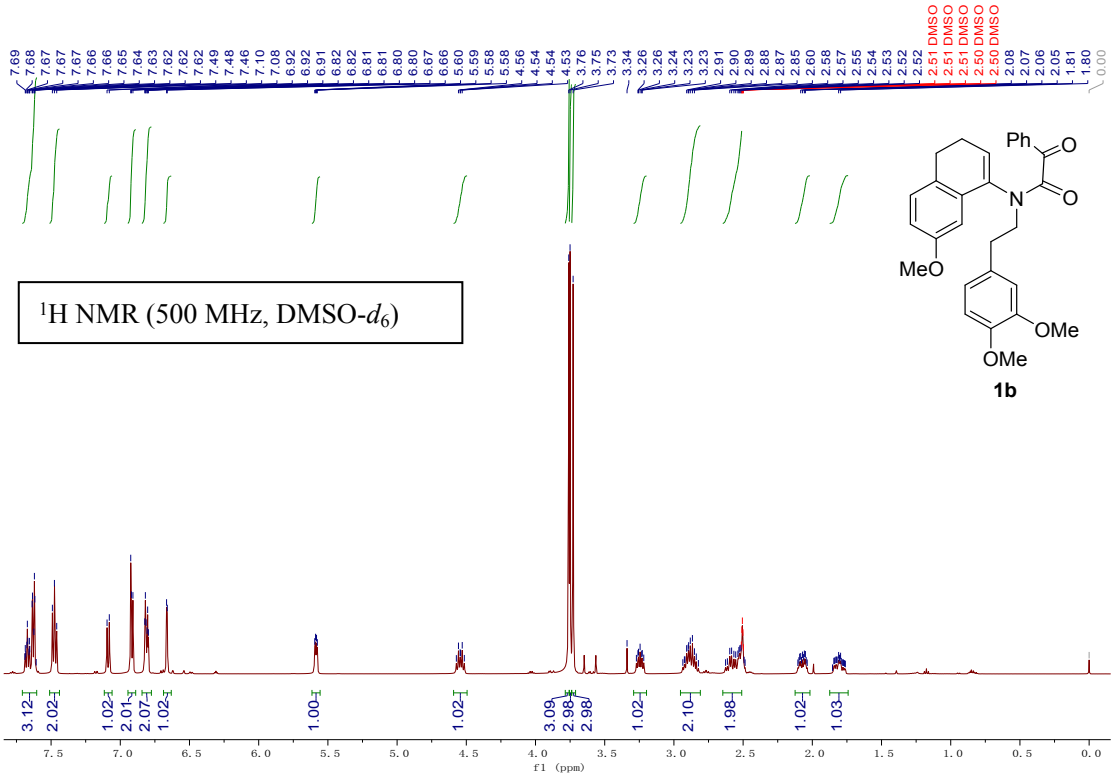
^{1}H NMR (500 MHz, Chloroform-*d*)

1a

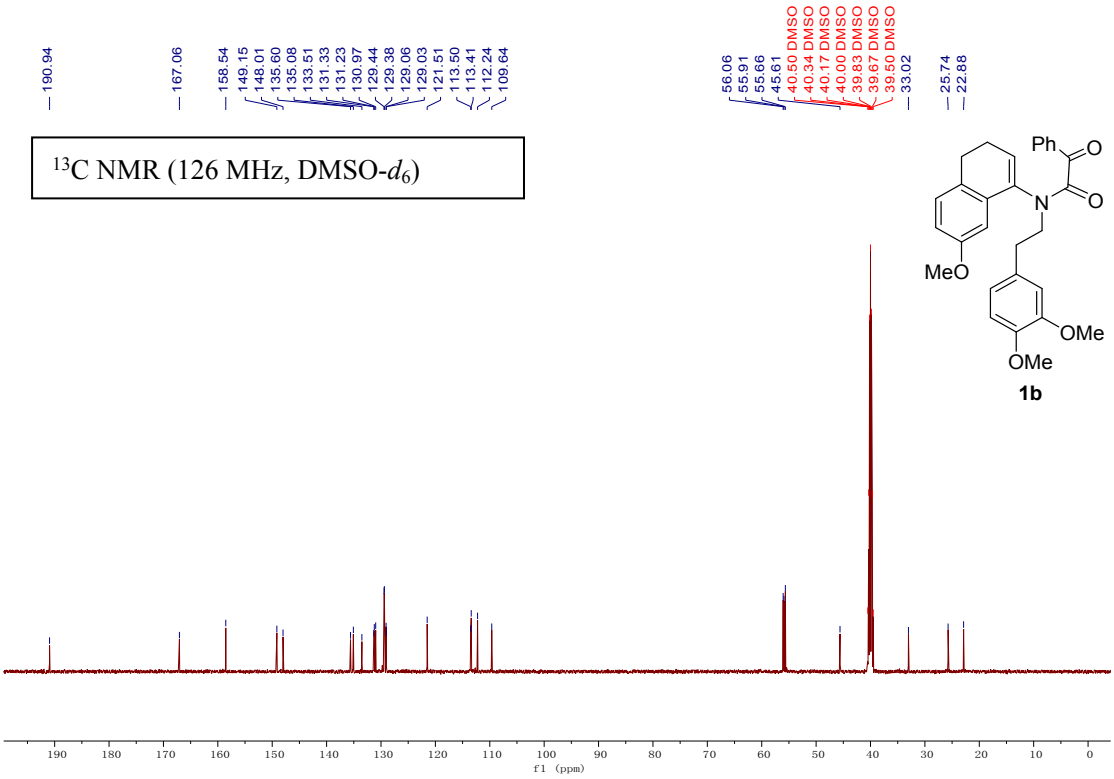


^{13}C NMR (126 MHz, Chloroform-*d*)

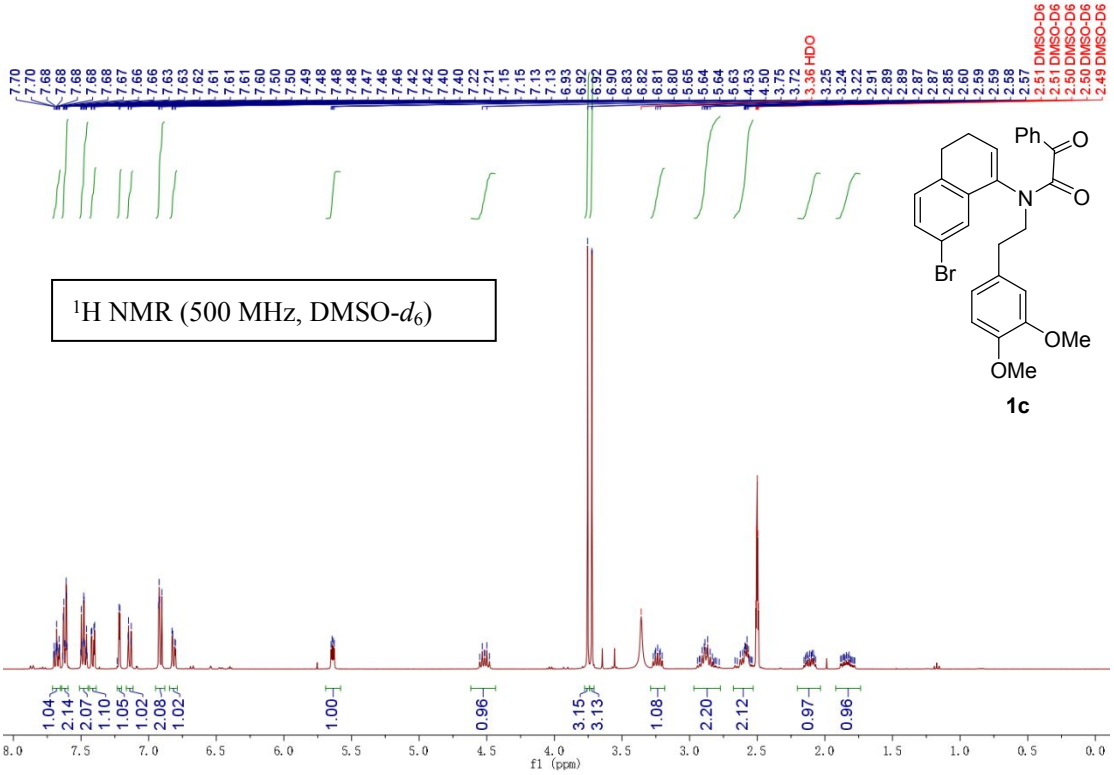
1a



^{1}H NMR (500 MHz, DMSO-d_6)

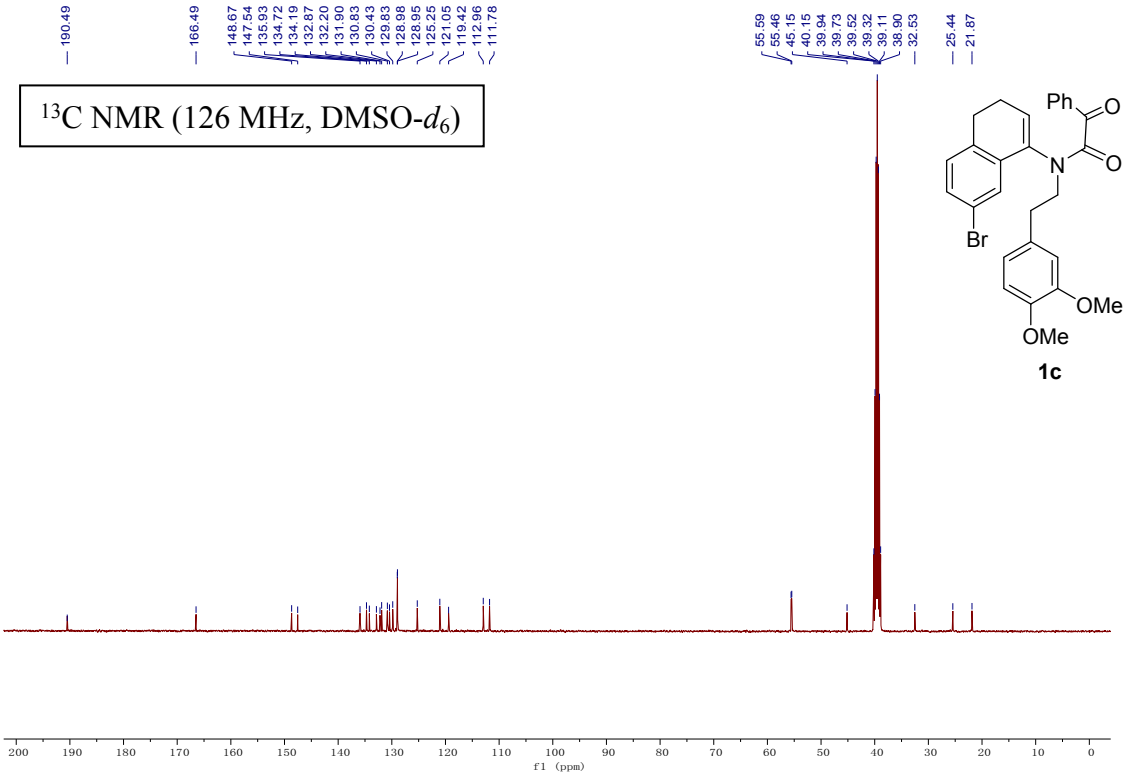


^{13}C NMR (126 MHz, DMSO-d_6)



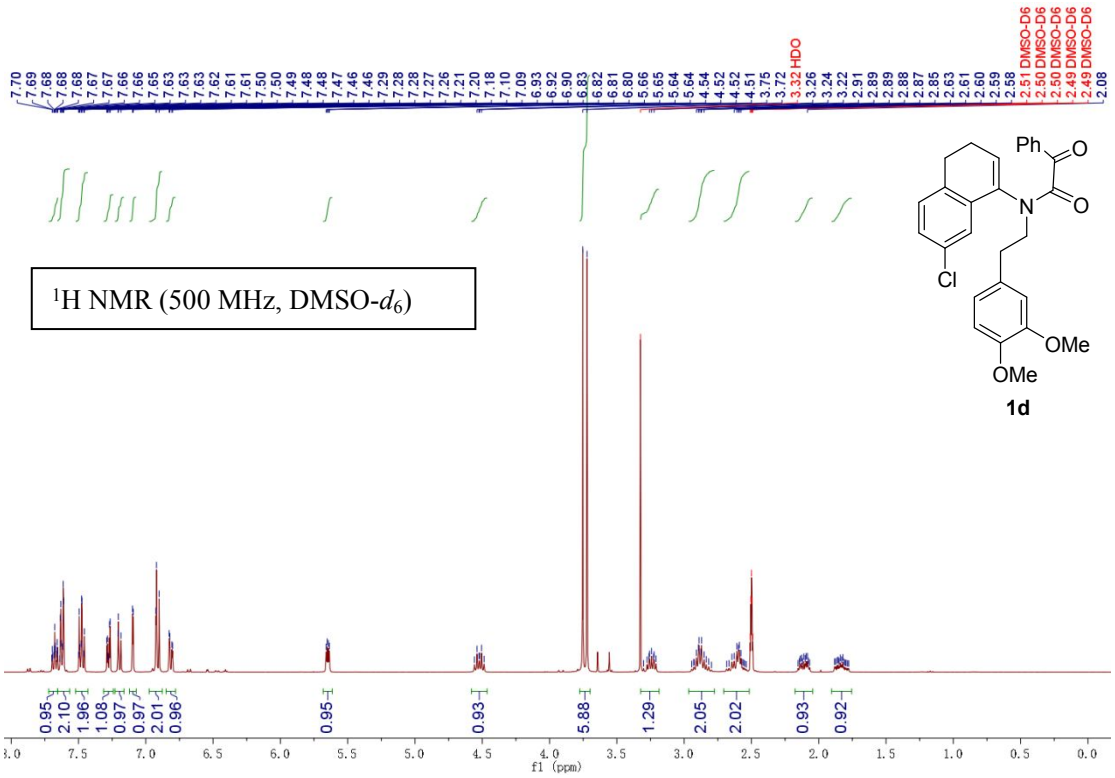
^{1}H NMR (500 MHz, DMSO-d_6)

1c

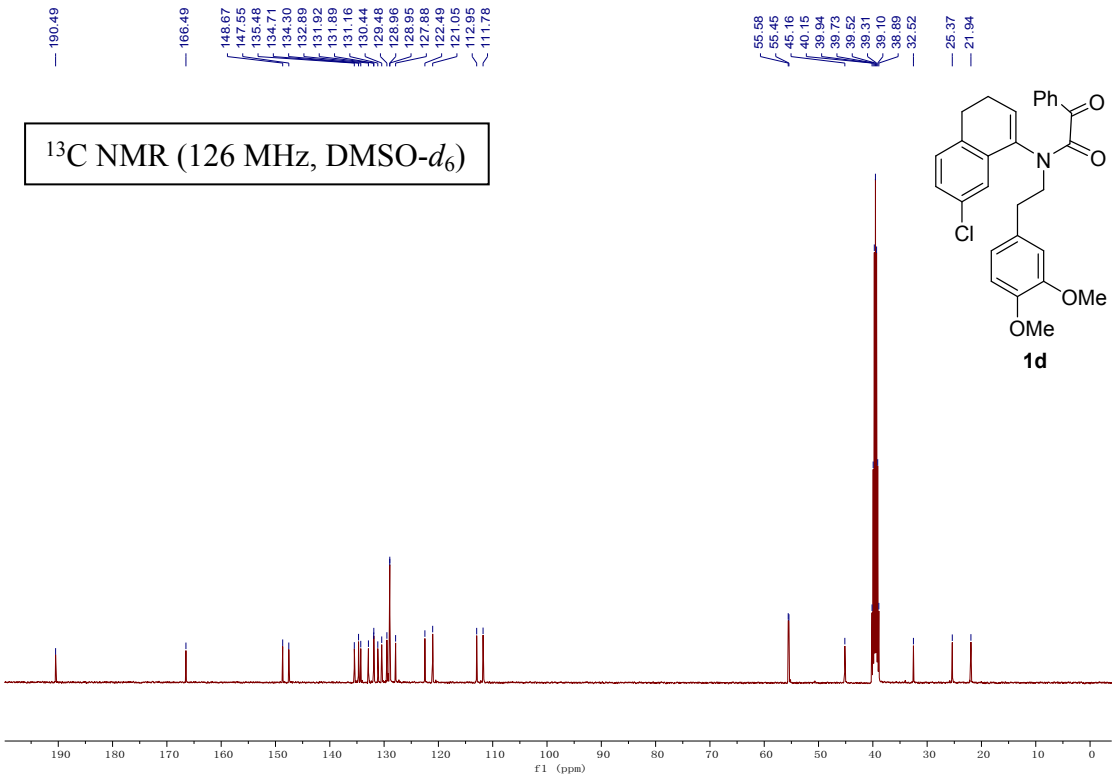


^{13}C NMR (126 MHz, DMSO-d_6)

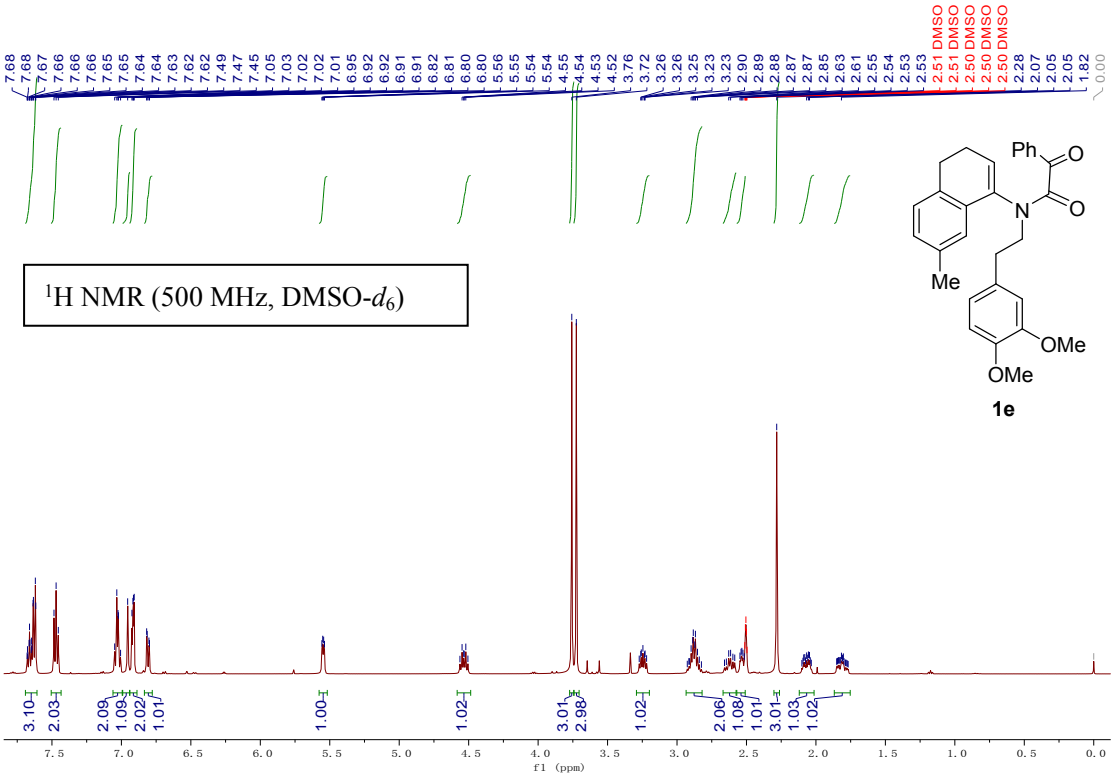
1c



^{1}H NMR (500 MHz, DMSO-d_6)

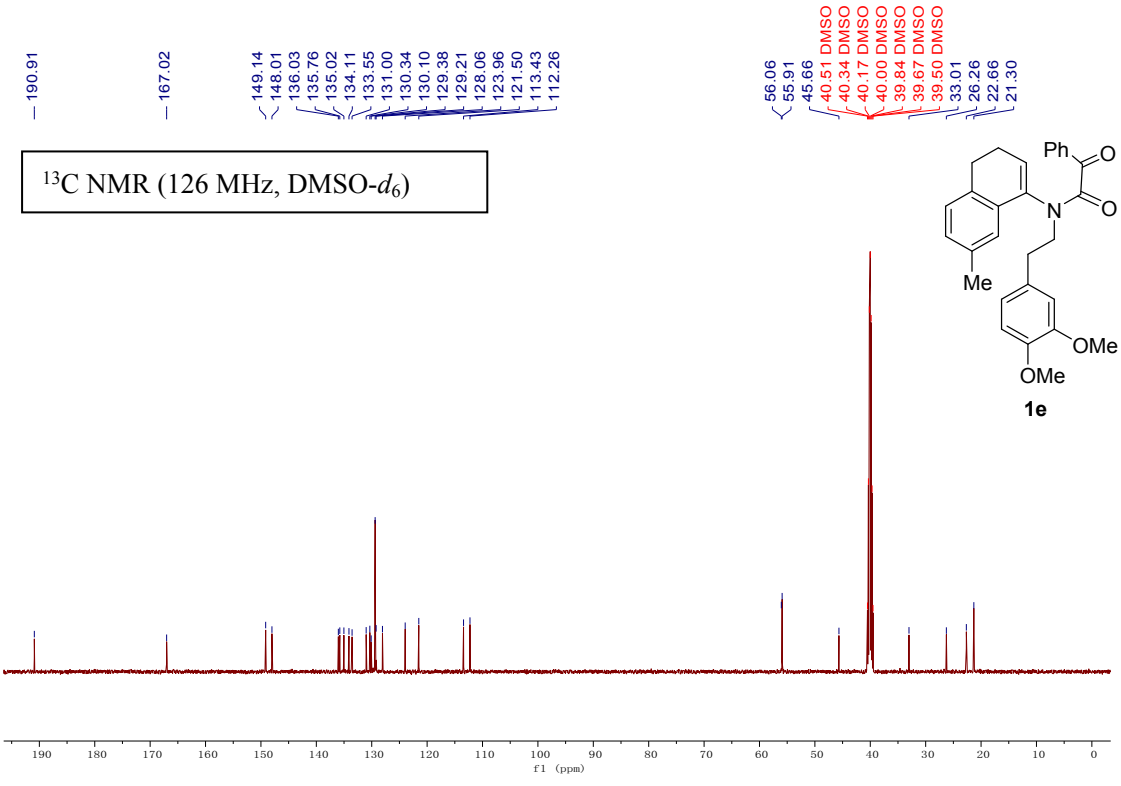


^{13}C NMR (126 MHz, DMSO-d_6)



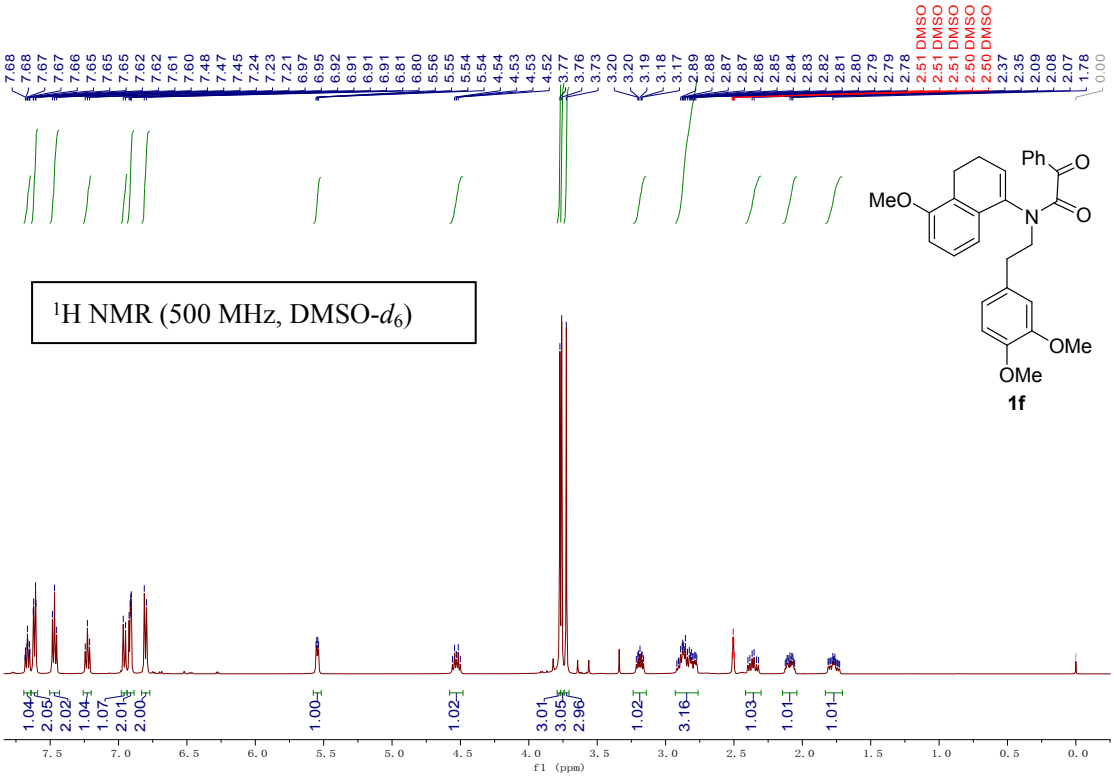
^{1}H NMR (500 MHz, DMSO-d_6)

1e



^{13}C NMR (126 MHz, DMSO-d_6)

1e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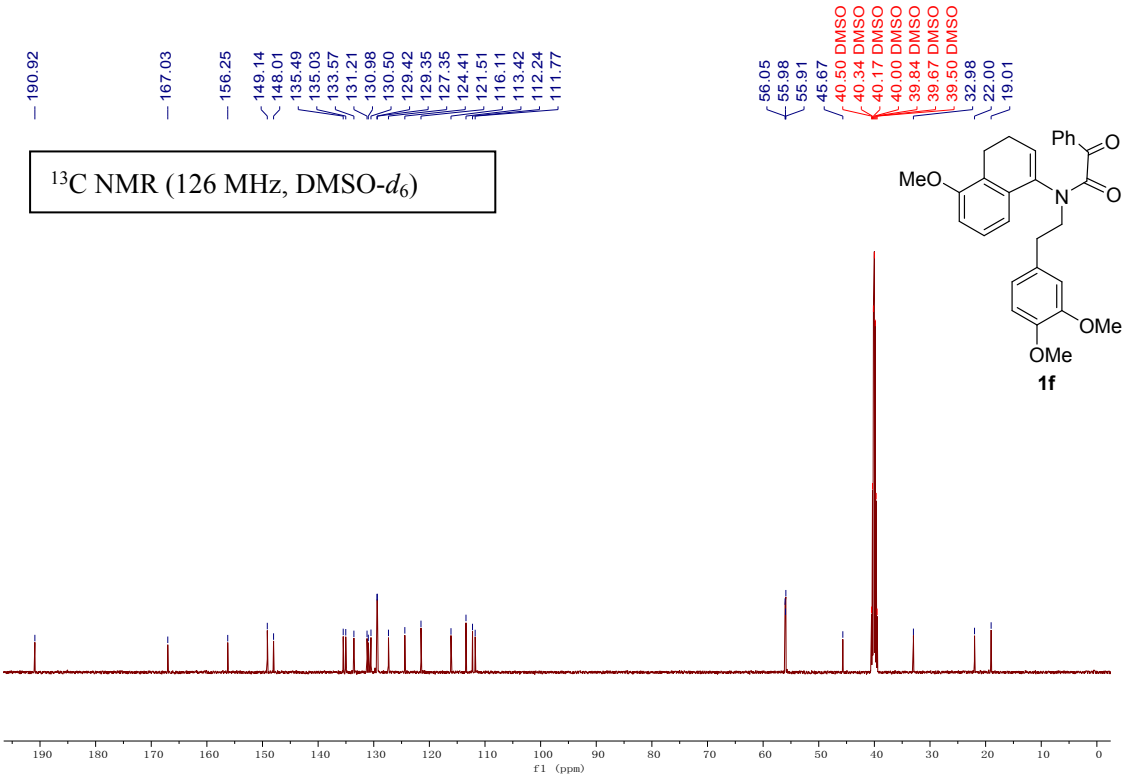
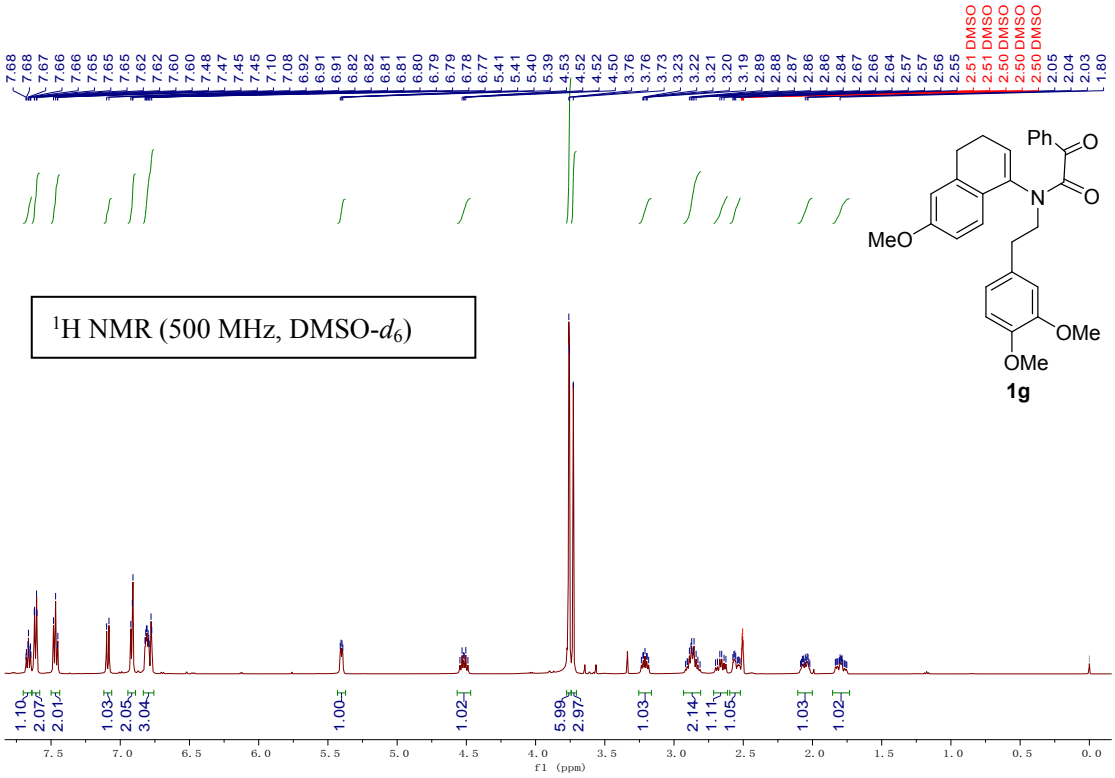
Ph
O
O
MeO
N
OMe
OMe
1f
1H NMR (500 MHz, DMSO-d6)
1.04 2.05 2.02 1.04 1.07 2.01 2.00 1.00 1.02 3.01 3.05 2.96 1.02 3.16 1.03 1.01 1.01
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


190.92 167.03 156.25 149.14 148.01 135.49 135.03 133.57 131.21 130.98 130.50 129.42 129.35 127.35 124.41 121.51 116.11 113.42 112.24 111.77
56.05 55.98 55.91 45.67 40.50 DMSO 40.34 DMSO 40.17 DMSO 40.00 DMSO 39.84 DMSO 39.67 DMSO 39.50 DMSO 32.98 22.00 19.01
Ph
O
O
MeO
N
OMe
OMe
1f
13C NMR (126 MHz, DMSO-d6)
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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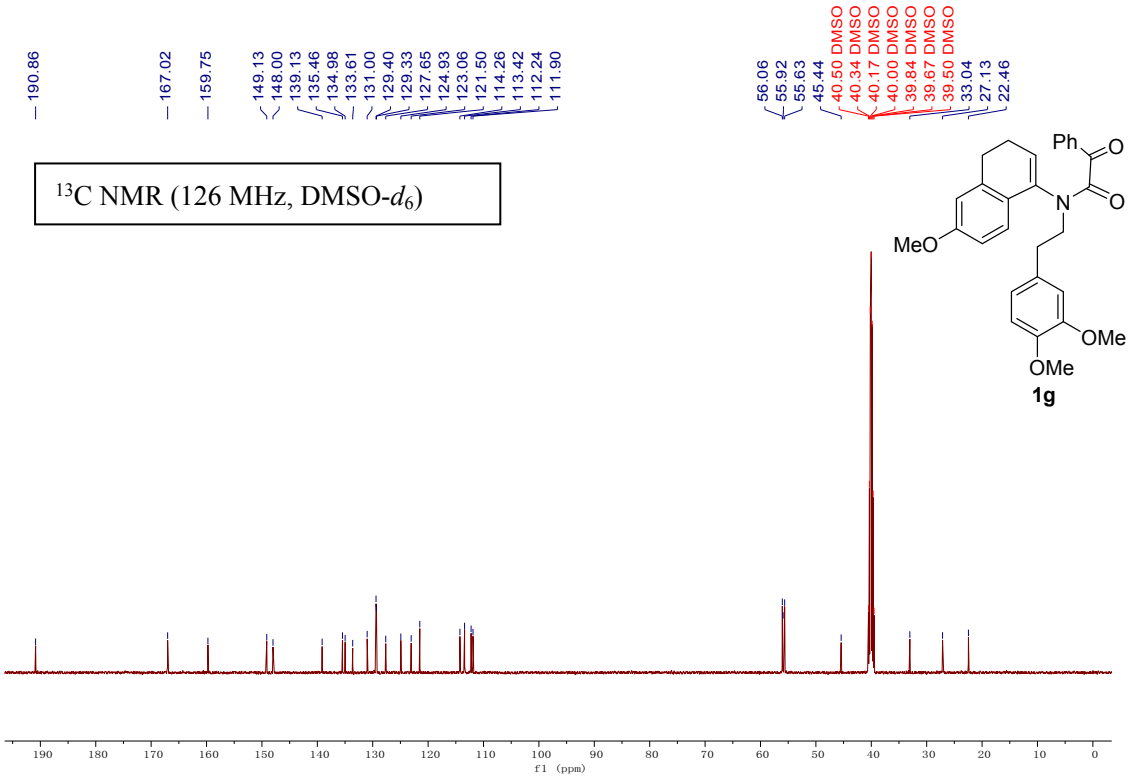
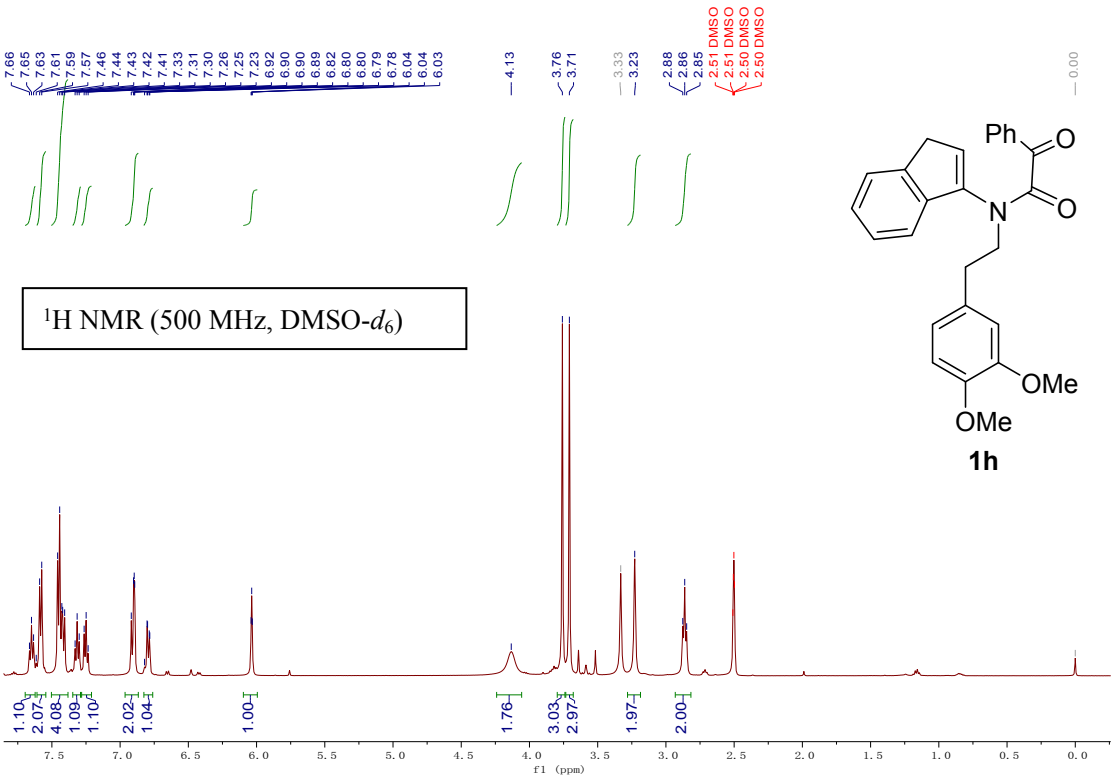
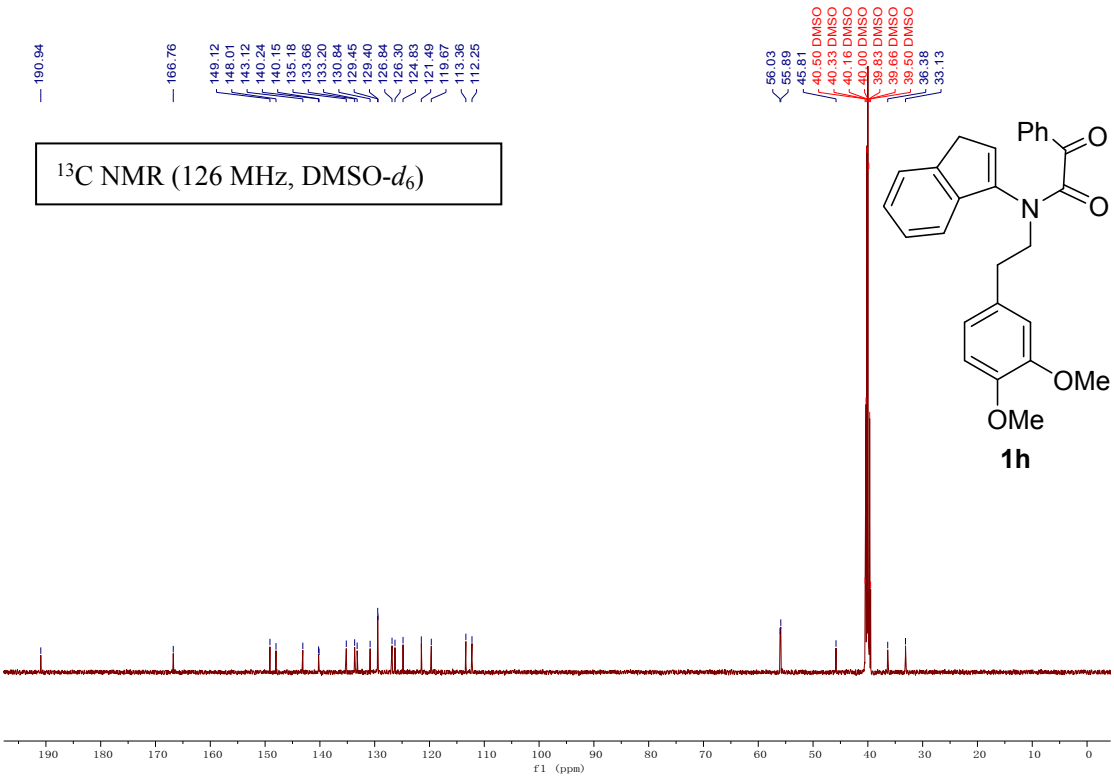
O
MeO
OMe
OMe
1g
^{1}H NMR (500 MHz, DMSO-d_6)

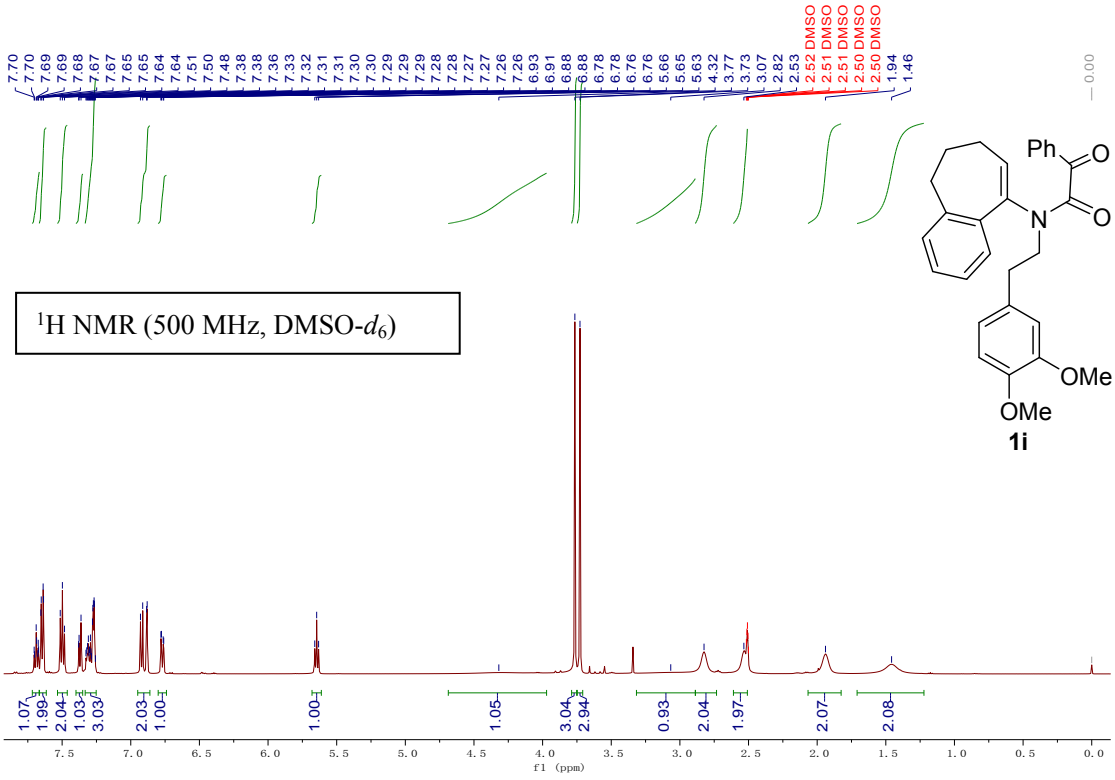

Ph
O
N
O
MeO
OMe
OMe
1g
^{13}C NMR (126 MHz, DMSO-d_6)

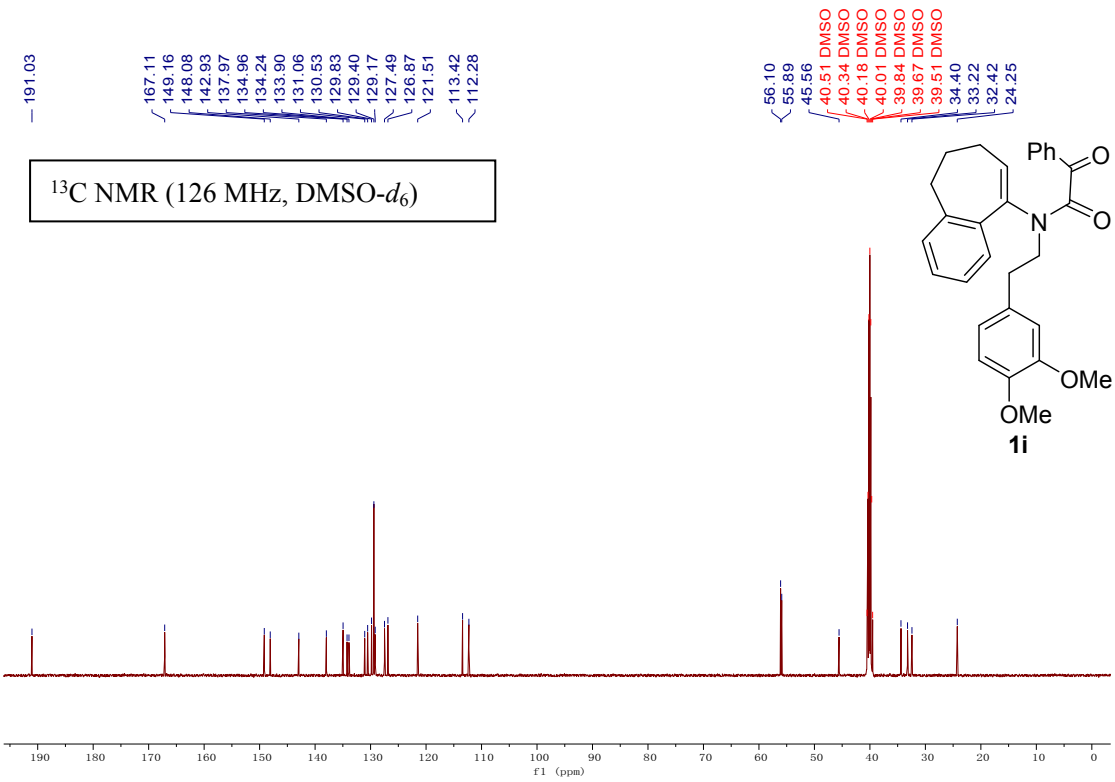


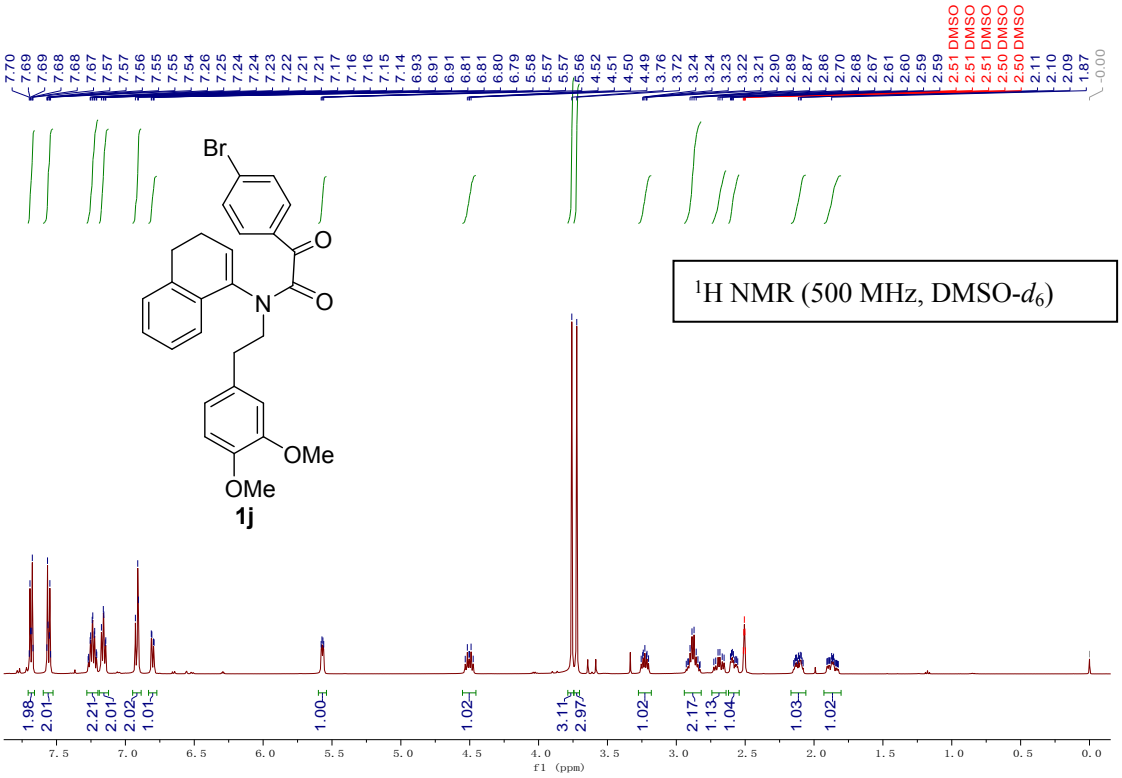
^{1}H NMR (500 MHz, DMSO-d_6)

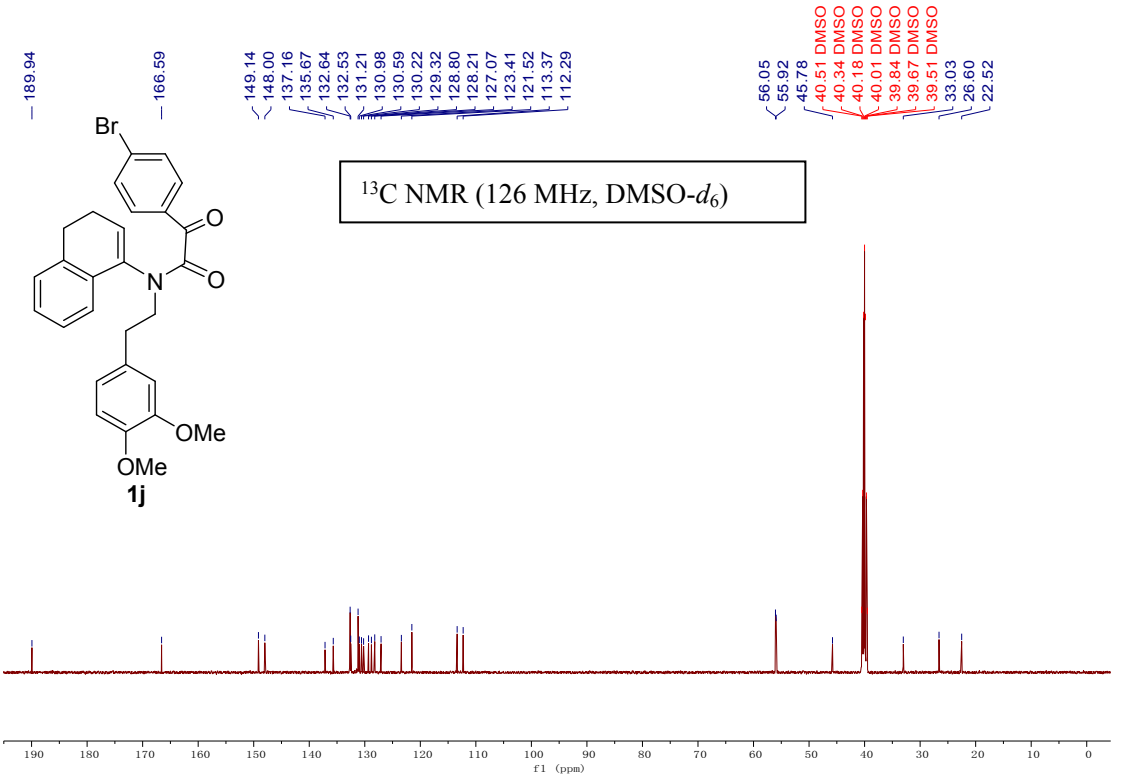


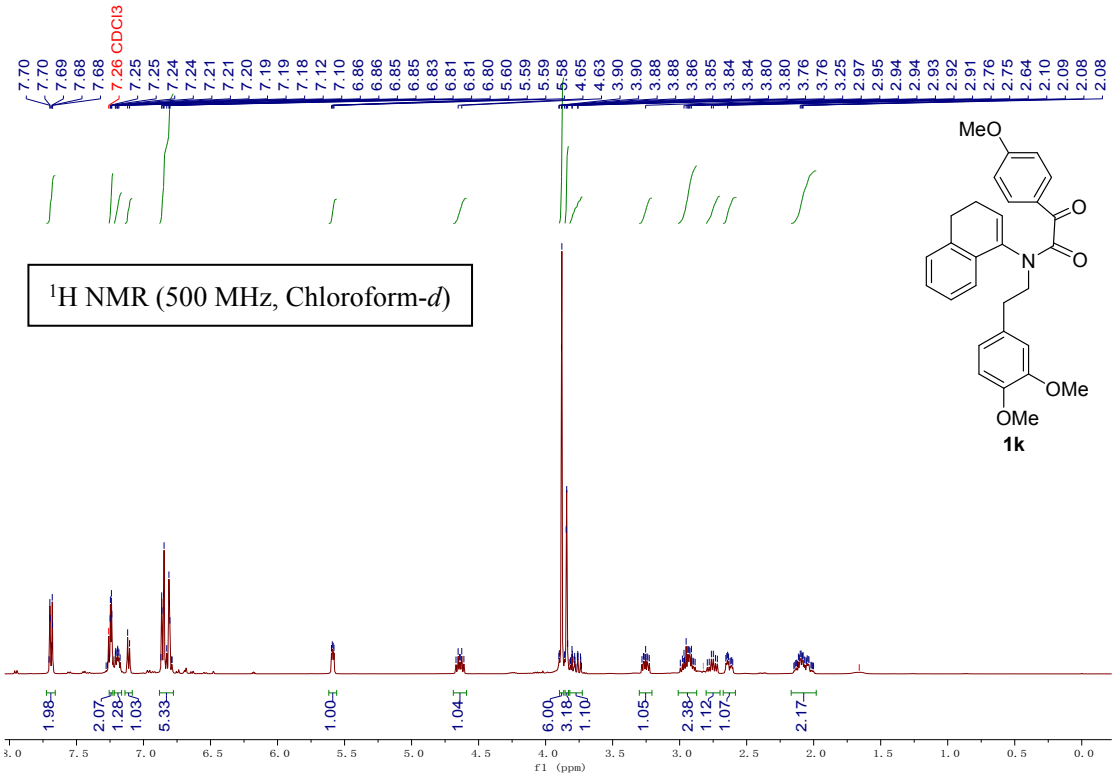
^{13}C NMR (126 MHz, DMSO-d_6)


Ph
O
O
N
OMe
OMe
1i
[1]H NMR (500 MHz, DMSO-d_6)


Ph
O
O
N
OMe
OMe
1i
[13]C NMR (126 MHz, DMSO-d_6)

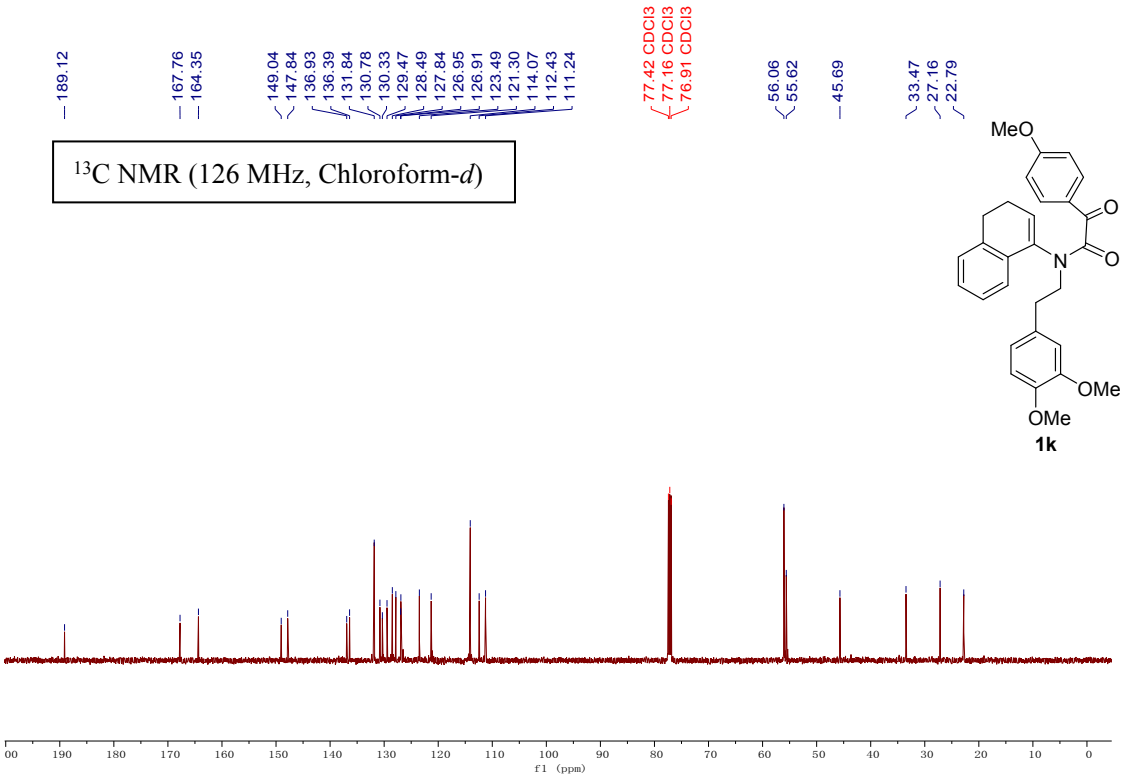

Br
O
O
N
OMe
OMe
1j
^{1}H NMR (500 MHz, DMSO-d_6)


Br
O
O
N
OMe
OMe
1j
^{13}C NMR (126 MHz, DMSO-d_6)



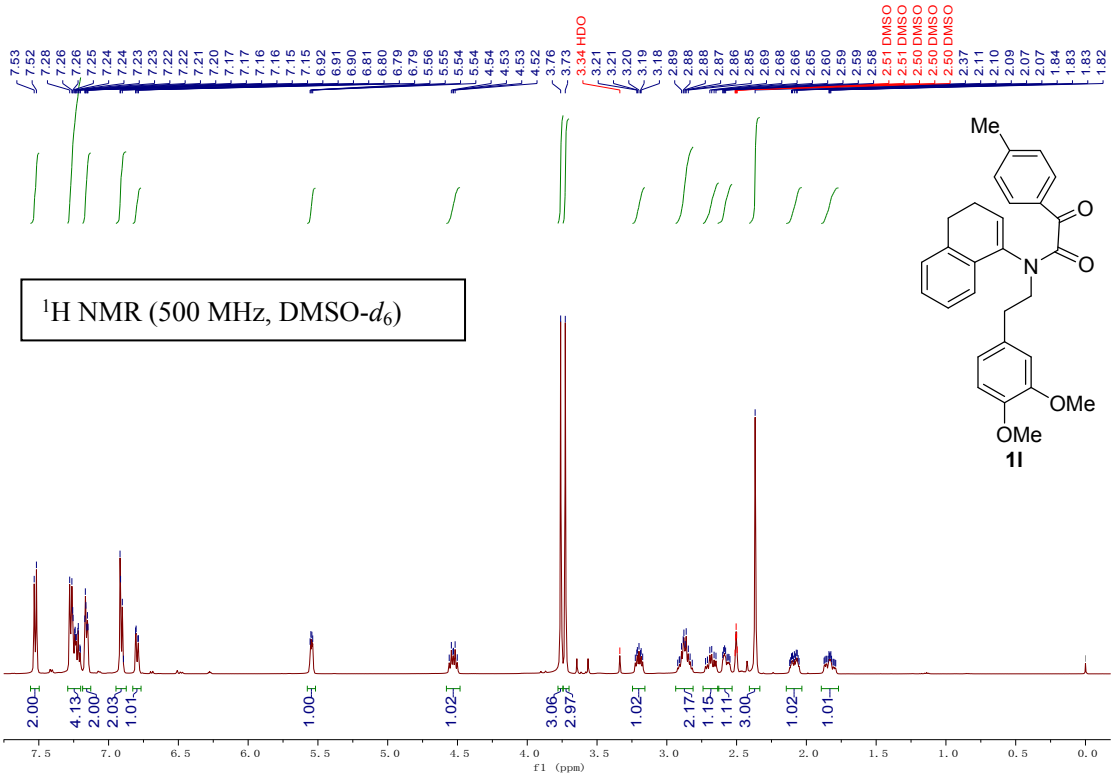
^{1}H NMR (500 MHz, Chloroform-*d*)

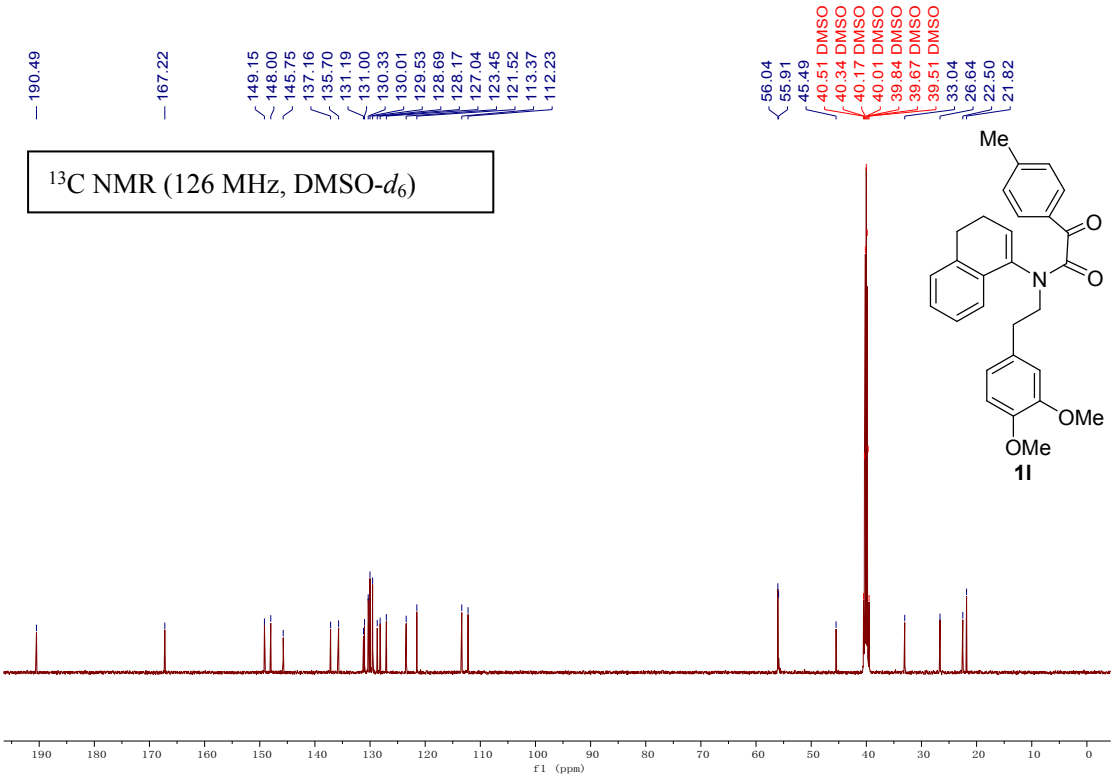
1k

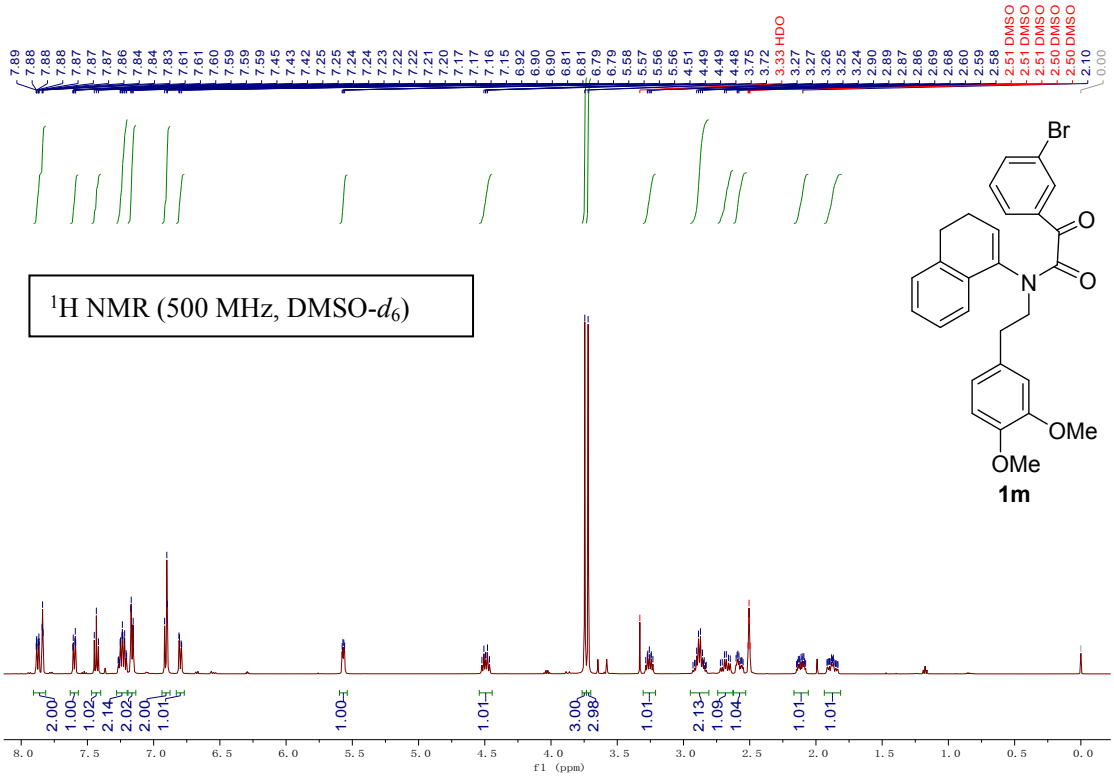


^{13}C NMR (126 MHz, Chloroform-*d*)

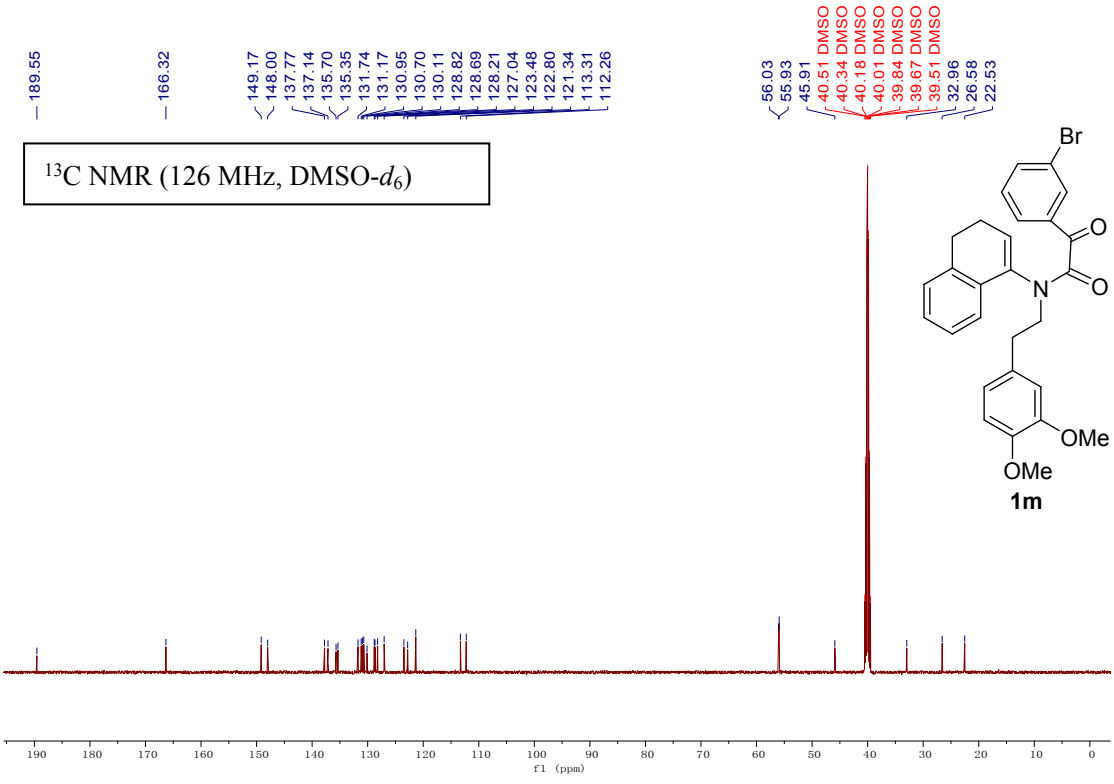
1k


7.53 7.52 7.28 7.26 7.26 7.25 7.24 7.24 7.23 7.23 7.22 7.22 7.21 7.20 7.17 7.17 7.16 7.16 7.15 7.15 6.92 6.91 6.90 6.81 6.80 6.79 6.79 5.56 5.55 5.54 5.54 4.54 4.53 4.52 3.76 3.73 3.34 HDO 3.21 3.21 3.20 3.19 3.18 2.89 2.88 2.88 2.87 2.86 2.85 2.69 2.68 2.66 2.65 2.60 2.59 2.59 2.58 2.51 DMSO 2.51 DMSO 2.50 DMSO 2.50 DMSO 2.50 DMSO 2.37 2.11 2.10 2.09 2.07 2.07 1.84 1.83 1.83 1.82
Me
O
O
N
OMe
OMe
1l
^{1}H NMR (500 MHz, DMSO-d_6)
2.00 4.13 2.00 2.03 1.01 1.00 1.02 3.06 2.97 1.02 2.17 1.15 1.11 3.00 1.02 1.01
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

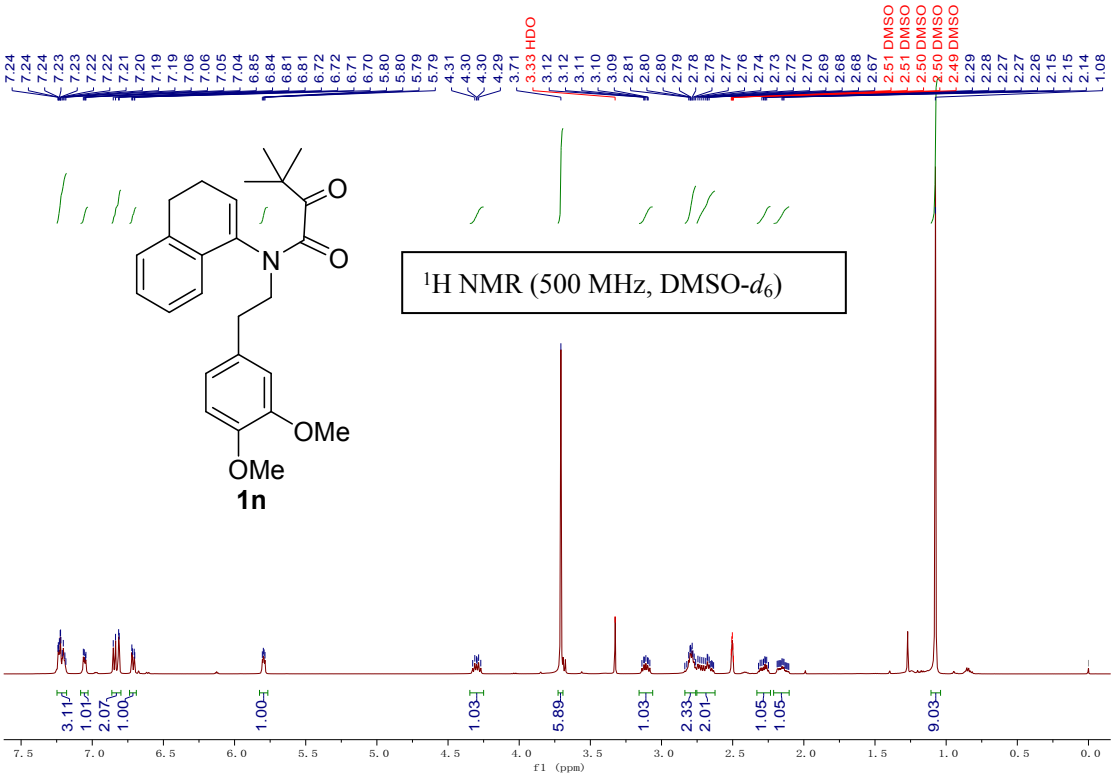

190.49 167.22 149.15 148.00 145.75 137.16 135.70 131.19 131.00 130.33 130.01 129.53 128.69 128.17 127.04 123.45 121.52 113.37 112.23 56.04 55.91 45.49 40.51 DMSO 40.34 DMSO 40.17 DMSO 40.01 DMSO 39.84 DMSO 39.67 DMSO 39.51 DMSO 33.04 26.64 22.50 21.82
Me
O
O
N
OMe
OMe
1l
^{13}C NMR (126 MHz, DMSO-d_6)
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

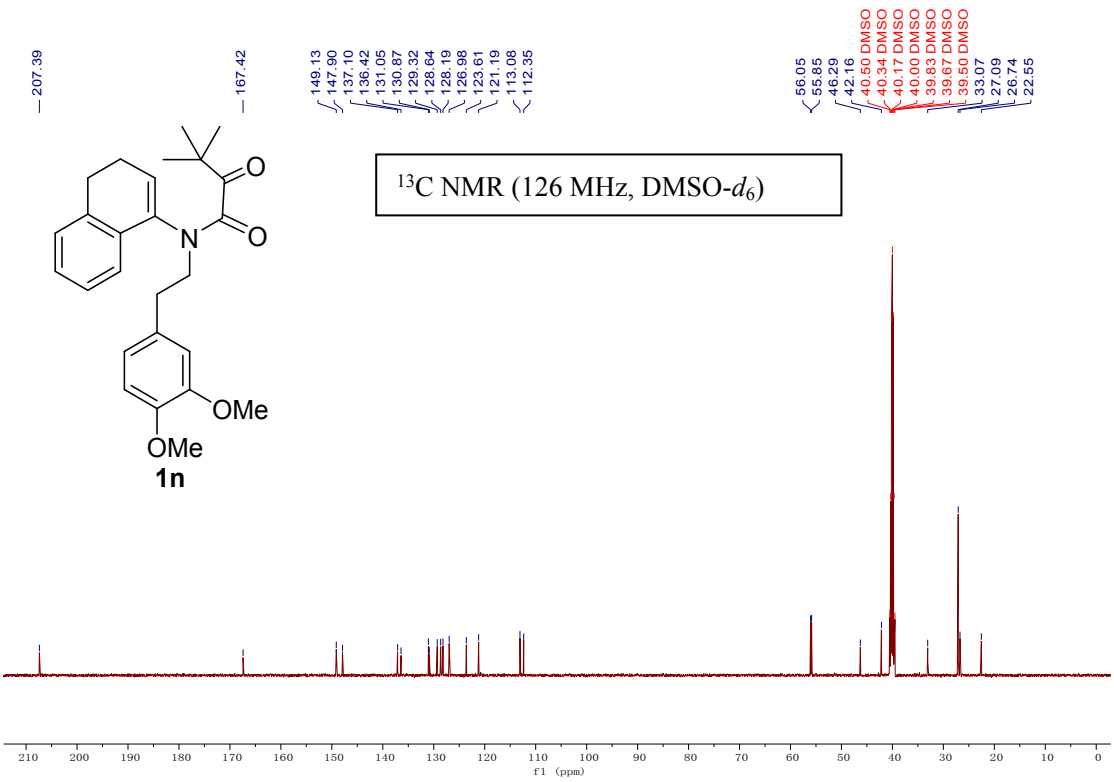


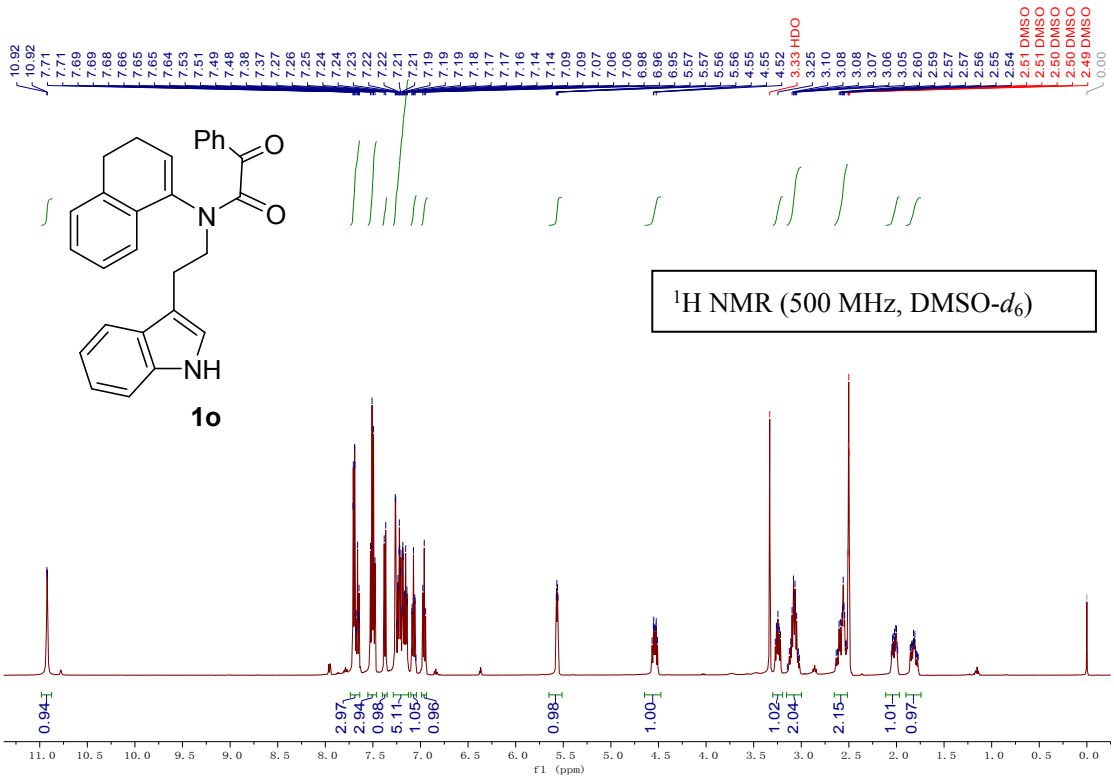
^{1}H NMR (500 MHz, DMSO-d_6)



^{13}C NMR (126 MHz, DMSO-d_6)


7.24 7.24 7.24 7.23 7.23 7.22 7.22 7.21 7.20 7.19 7.19 7.06 7.06 7.05 7.04 6.85 6.84 6.81 6.81 6.72 6.72 6.71 6.70 5.80 5.80 5.79 5.79 4.31 4.30 4.30 4.29 3.71 3.33 HDO 3.12 3.12 3.11 3.10 3.09 2.81 2.80 2.80 2.79 2.78 2.78 2.77 2.76 2.74 2.73 2.72 2.70 2.69 2.68 2.68 2.67 2.51 DMSO 2.51 DMSO 2.50 DMSO 2.50 DMSO 2.49 DMSO 2.29 2.28 2.27 2.27 2.26 2.15 2.15 2.14 1.08
O
O
N
OMe
OMe
1n
^{1}H NMR (500 MHz, DMSO-d_6)
3.11 1.01 2.07 1.00 1.00 1.03 5.89 1.03 2.33 2.01 1.05 1.05 9.03
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


207.39 167.42 149.13 147.90 137.10 136.42 131.05 130.87 129.32 128.64 128.19 126.98 123.61 121.19 113.08 112.35 56.05 55.85 46.29 42.16 40.50 DMSO 40.34 DMSO 40.17 DMSO 40.00 DMSO 39.83 DMSO 39.67 DMSO 39.50 DMSO 33.07 27.09 26.74 22.55
O
O
N
OMe
OMe
1n
^{13}C NMR (126 MHz, DMSO-d_6)
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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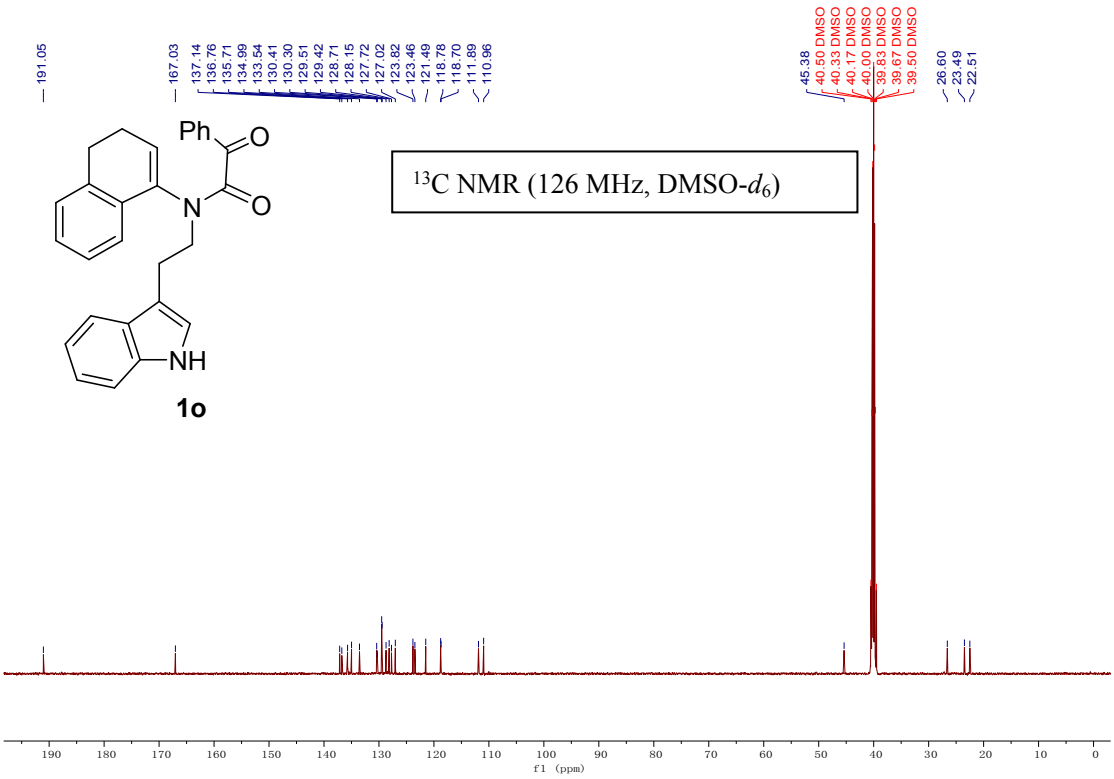
NH
1o
^{1}H NMR (500 MHz, DMSO-d_6)
f1 (ppm)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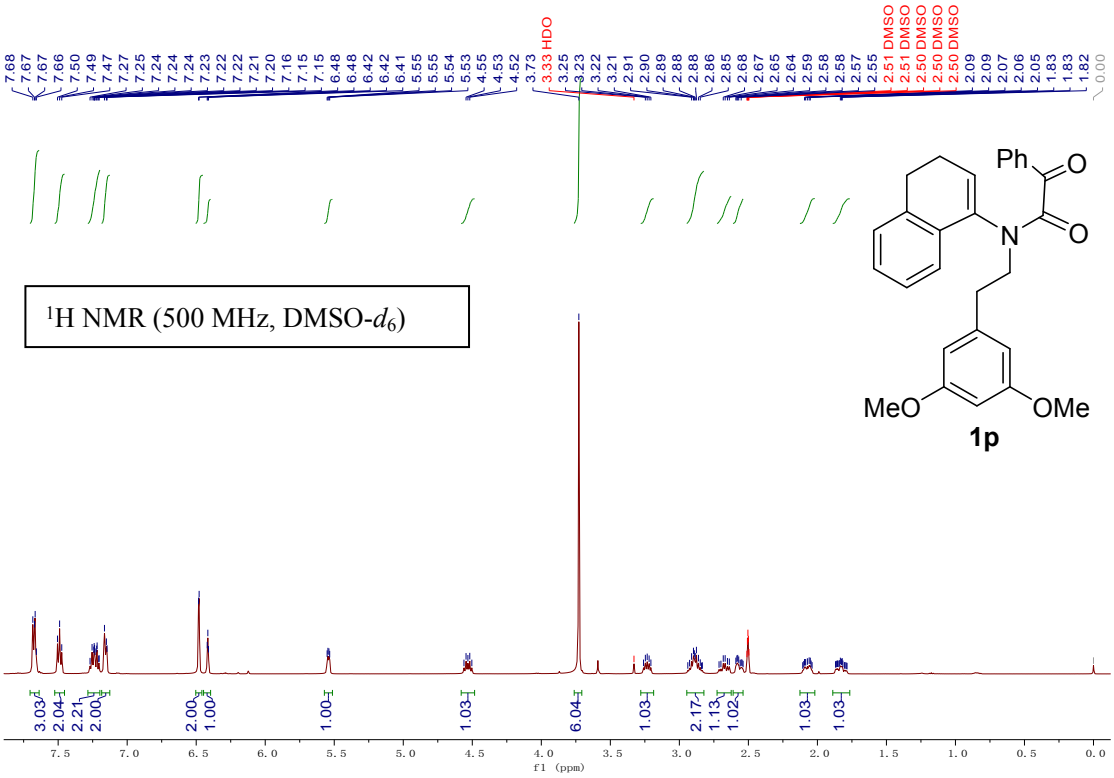
NH
1o
^{13}C NMR (126 MHz, DMSO-d_6)
f1 (ppm)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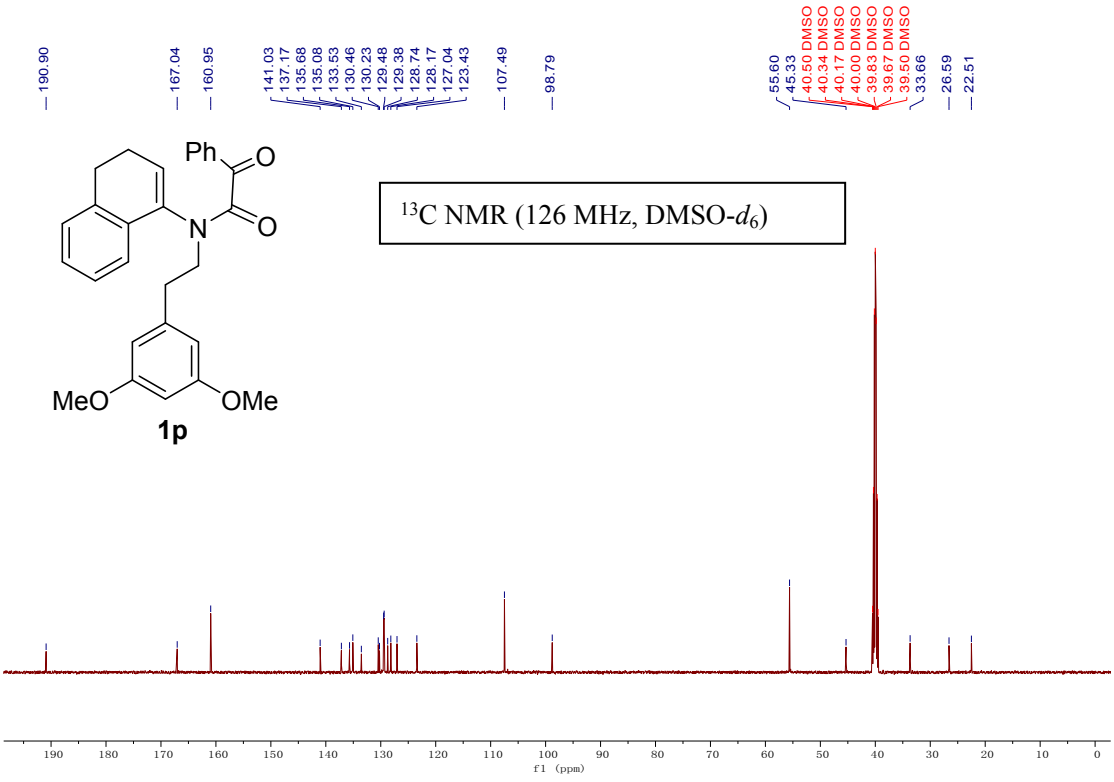
O
O
N
MeO
OMe
1p
[1]H NMR (500 MHz, DMSO-d_6)


Ph
O
O
N
MeO
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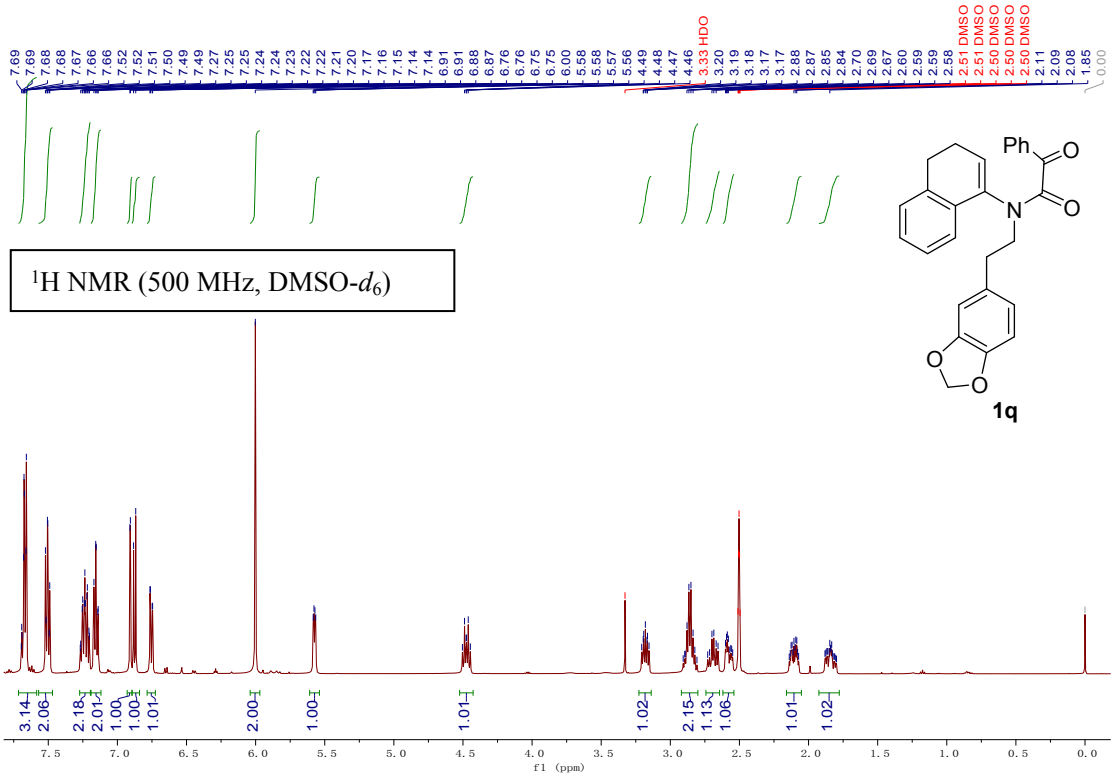
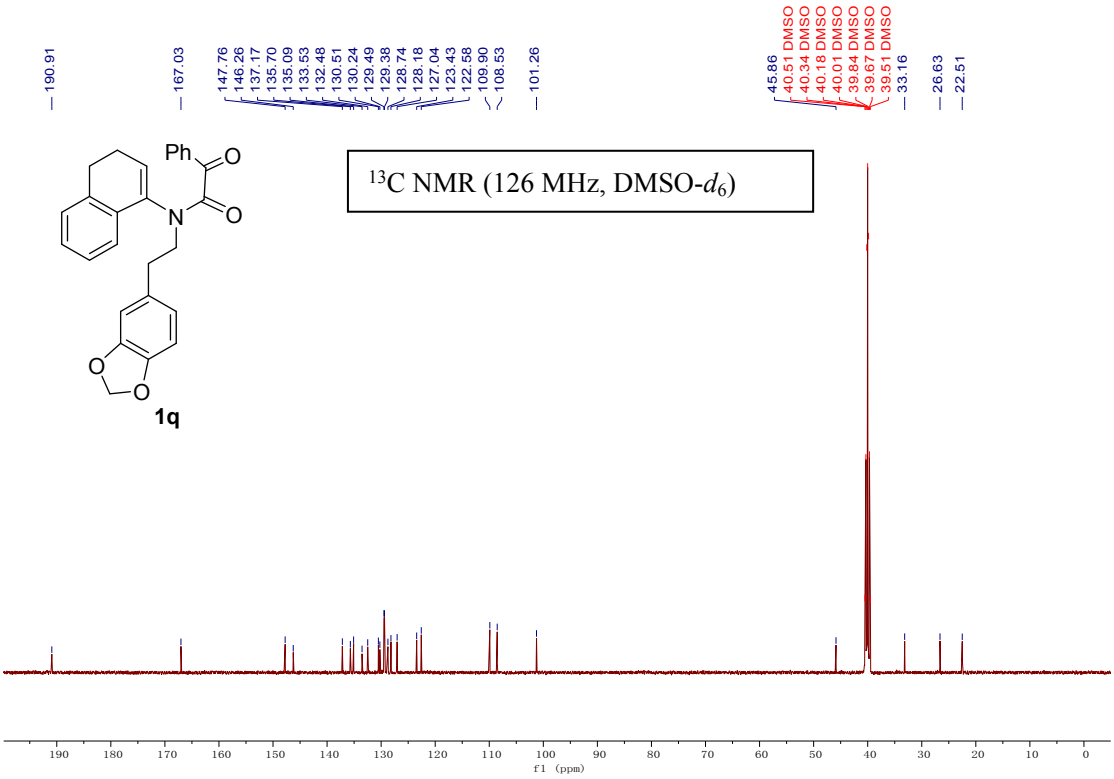
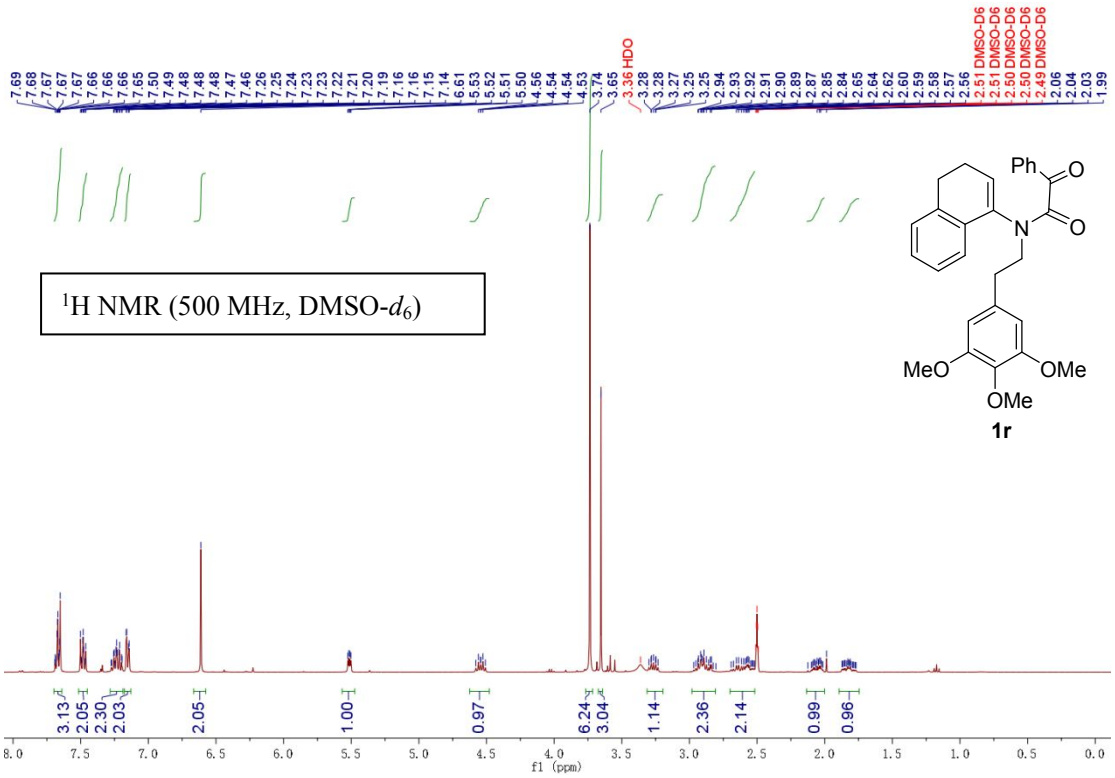
1p
[13]C NMR (126 MHz, DMSO-d_6)


7.69 7.69 7.68 7.68 7.67 7.66 7.66 7.52 7.51 7.50 7.49 7.49 7.27 7.25 7.24 7.24 7.23 7.22 7.22 7.21 7.20 7.17 7.16 7.15 7.14 7.14 6.91 6.91 6.88 6.87 6.76 6.76 6.75 6.75 6.00 5.58 5.58 5.57 5.56 4.49 4.48 4.47 4.46 3.33 HDO 3.20 3.19 3.18 3.17 3.17 2.88 2.87 2.85 2.84 2.70 2.69 2.67 2.60 2.59 2.59 2.58 2.51 DMSO 2.51 DMSO 2.50 DMSO 2.50 DMSO 2.50 DMSO 2.11 2.09 2.08 1.85 0.00
[1]H NMR (500 MHz, DMSO-d6)
Ph
O
O
N
O
O
1q
3.14 2.06 2.18 2.01 1.00 1.00 1.01 2.00 1.00 1.01 1.02 2.15 1.13 1.06 1.01 1.02
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


190.91 167.03 147.76 146.26 137.17 135.70 135.09 133.53 132.48 130.51 130.24 129.49 129.38 128.74 128.18 127.04 123.43 122.58 109.90 108.53 101.26 45.86 40.51 DMSO 40.34 DMSO 40.18 DMSO 40.01 DMSO 39.84 DMSO 39.67 DMSO 39.51 DMSO 33.16 26.63 22.51
Ph
O
O
N
O
O
1q
[13]C NMR (126 MHz, DMSO-d6)
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Ph
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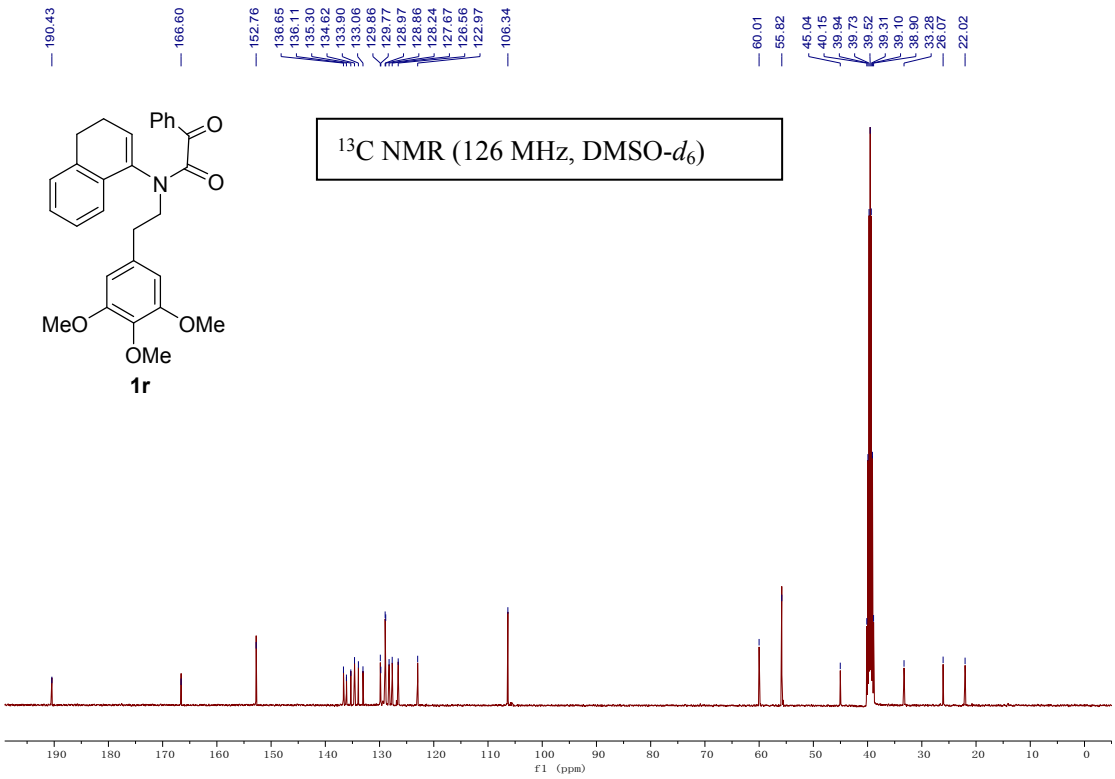
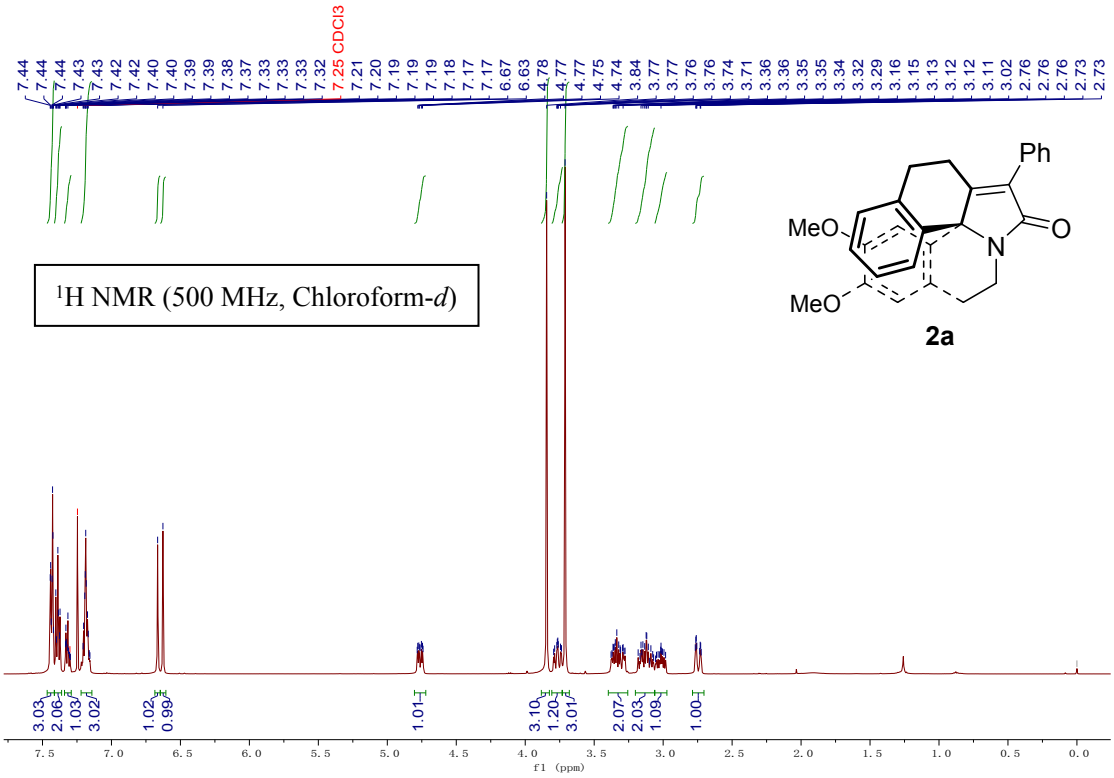
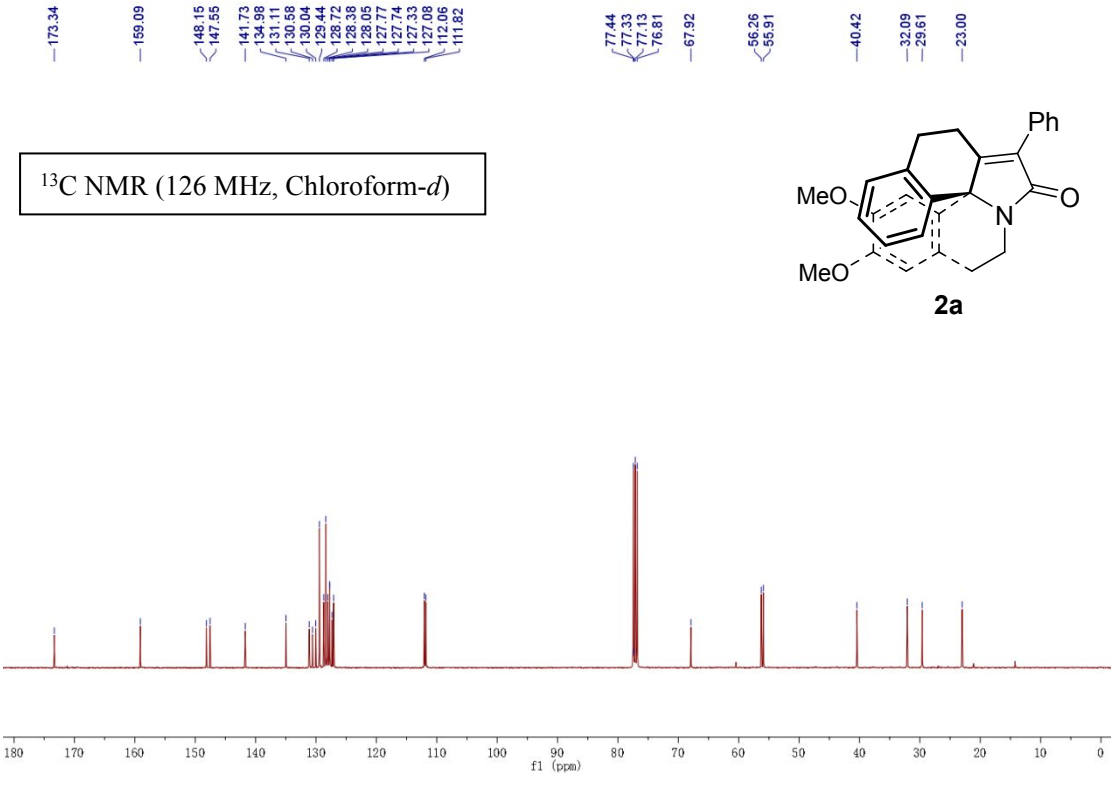
O
N
MeO
OMe
OMe
1r
^{1}H NMR (500 MHz, DMSO-d_6)


Ph
O
O
N
MeO
OMe
OMe
1r
^{13}C NMR (126 MHz, DMSO-d_6)



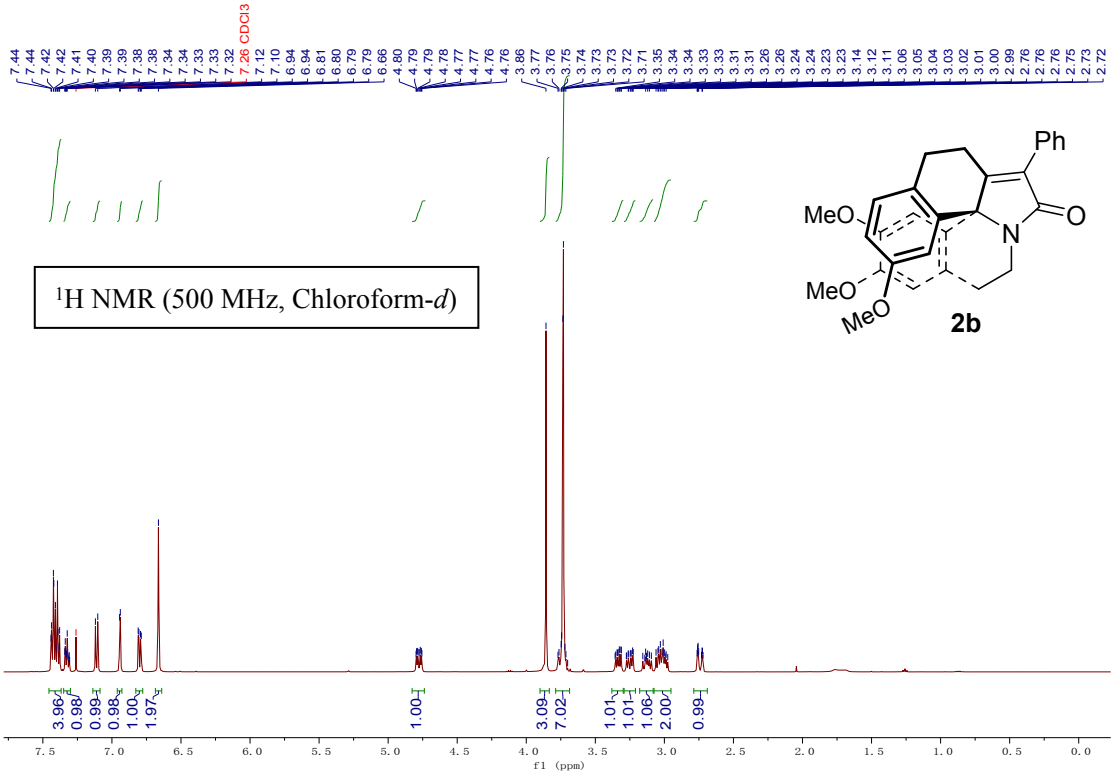
[1]H NMR (500 MHz, Chloroform-*d*)

2a



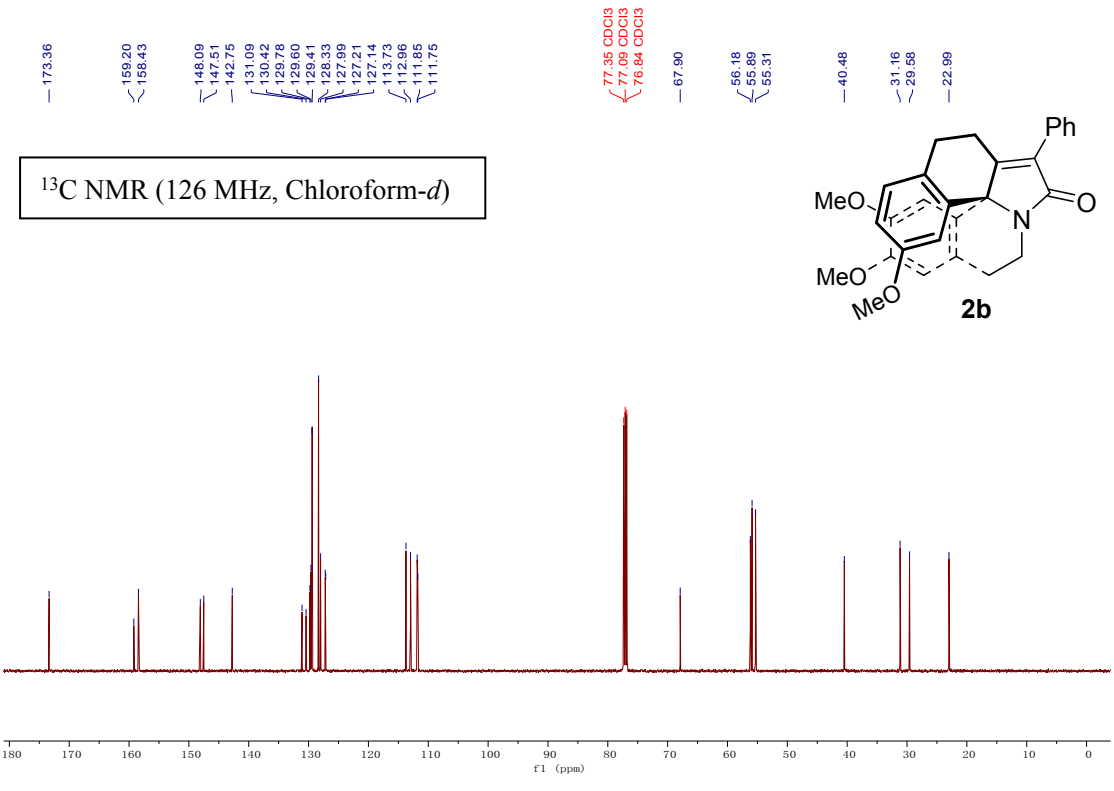
[13]C NMR (126 MHz, Chloroform-*d*)

2a



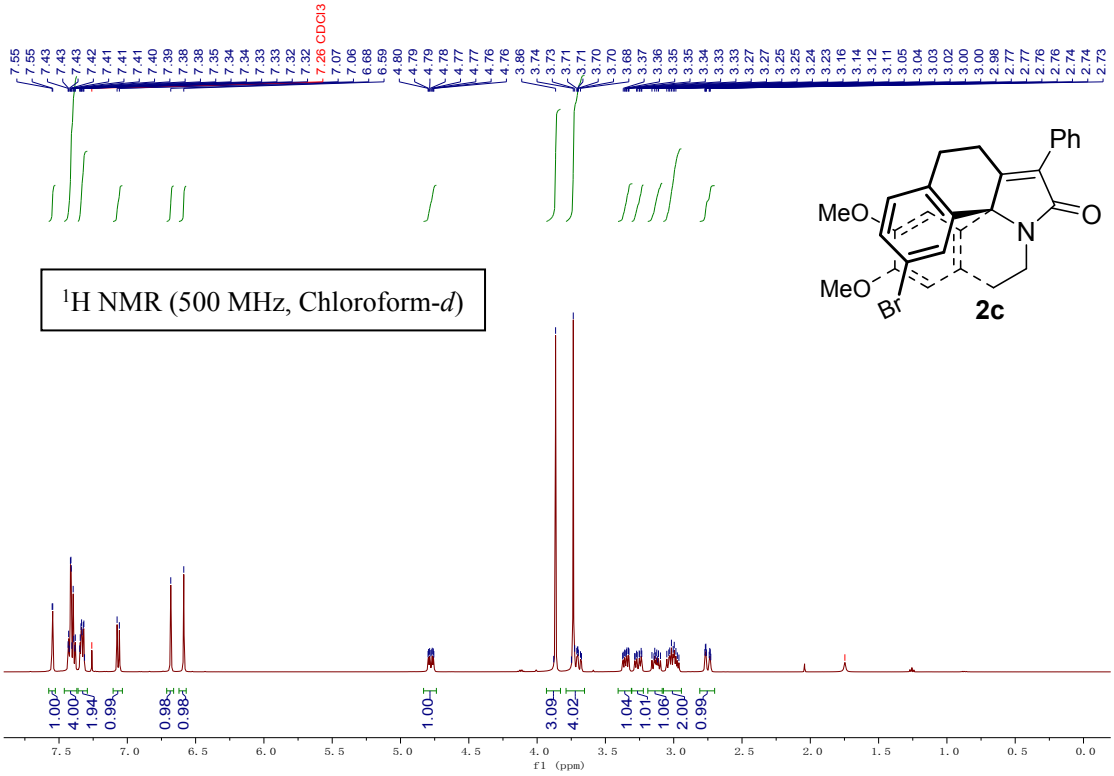
^{1}H NMR (500 MHz, Chloroform-*d*)

2b

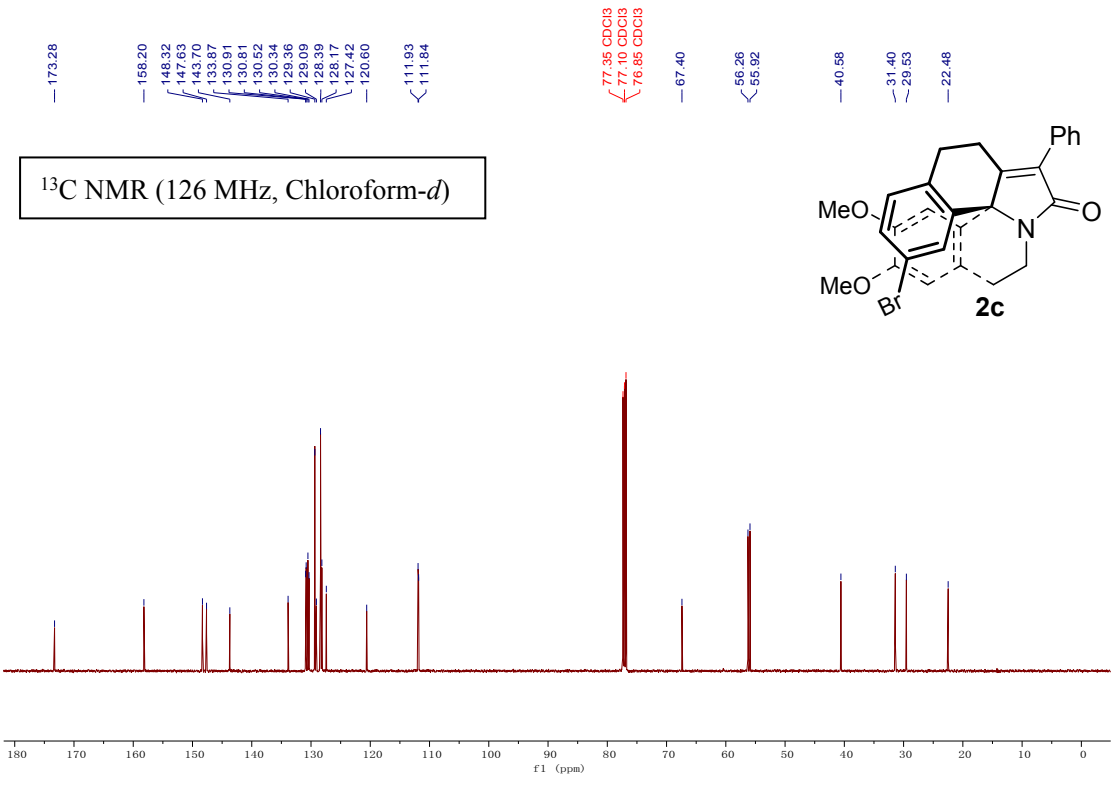


^{13}C NMR (126 MHz, Chloroform-*d*)

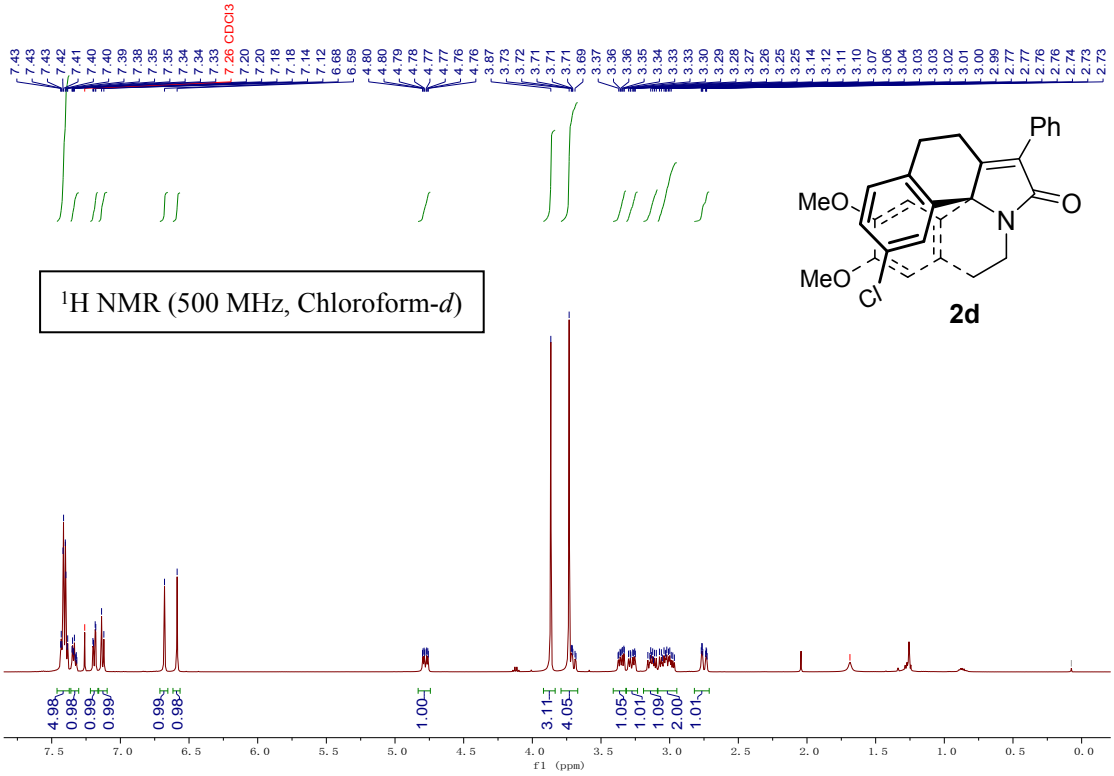
2b

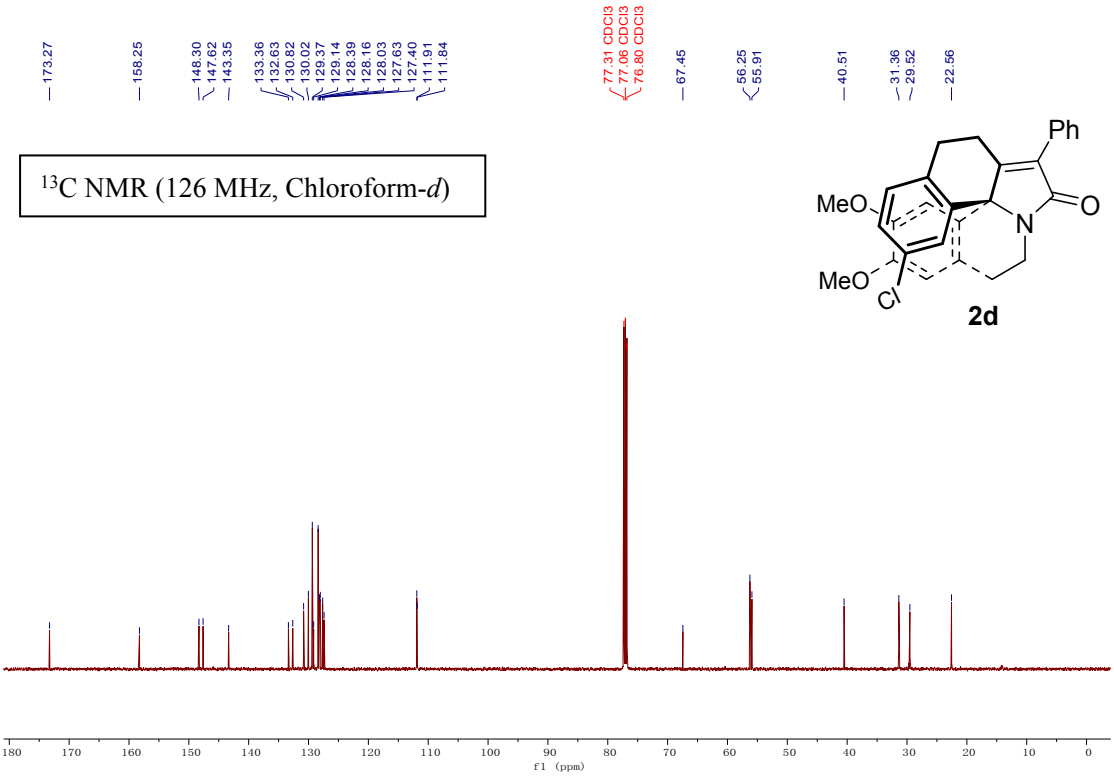


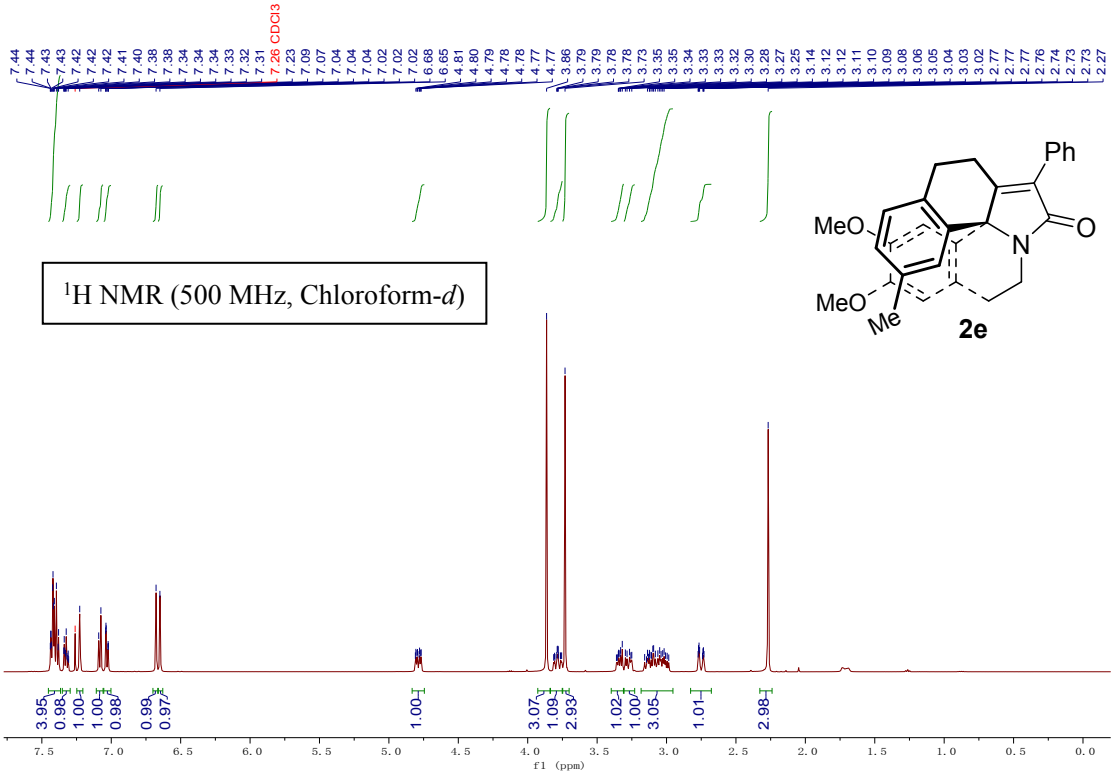
^{1}H NMR (500 MHz, Chloroform-*d*)



^{13}C NMR (126 MHz, Chloroform-*d*)

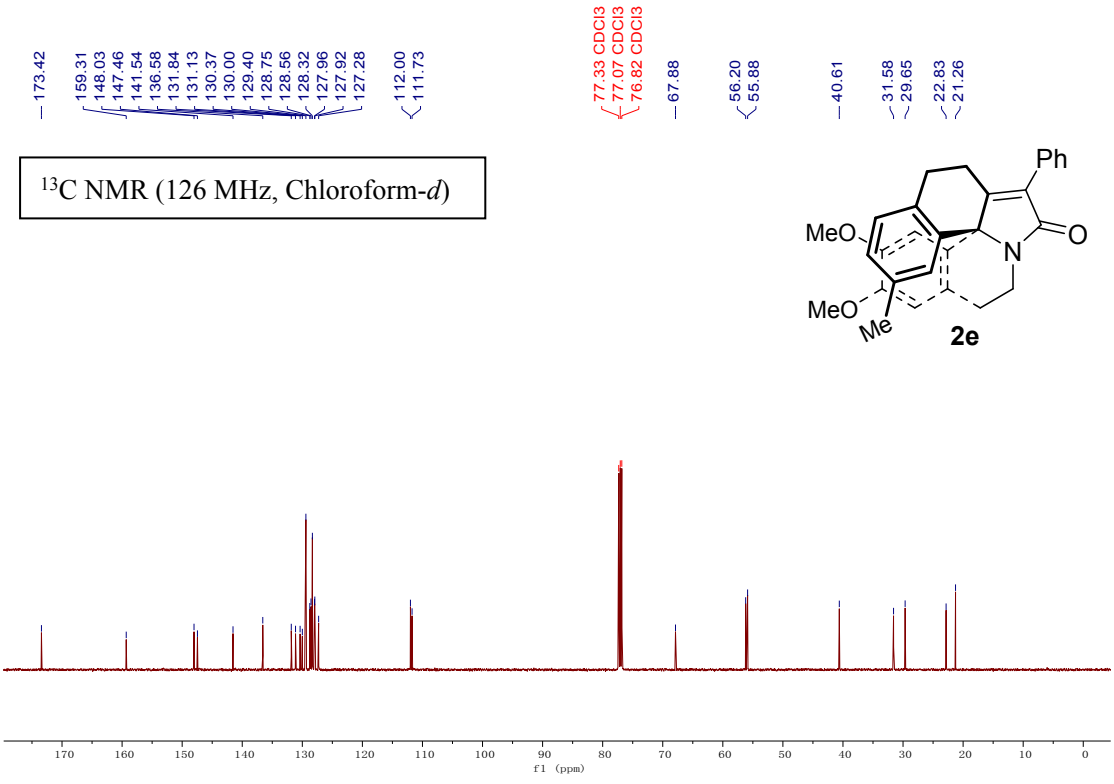

7.43 7.43 7.43 7.42 7.41 7.40 7.40 7.39 7.38 7.35 7.35 7.34 7.34 7.33 7.26 CDCl3 7.20 7.20 7.18 7.18 7.14 7.12 6.68 6.59 4.80 4.80 4.79 4.78 4.77 4.77 4.76 4.76 3.87 3.73 3.72 3.71 3.71 3.71 3.69 3.37 3.36 3.36 3.35 3.34 3.33 3.33 3.30 3.29 3.28 3.27 3.26 3.25 3.25 3.14 3.12 3.11 3.10 3.07 3.06 3.04 3.03 3.03 3.02 3.01 3.00 2.99 2.77 2.77 2.76 2.76 2.74 2.73 2.73
Ph
MeO
N
O
MeO
Cl
2d
[1]H NMR (500 MHz, Chloroform-d)
4.98 0.98 0.99 0.99 0.99 0.98 1.00 3.11 4.05 1.05 1.01 1.09 2.00 1.01
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


173.27 158.25 148.30 147.62 143.35 133.36 132.63 130.82 130.02 129.37 129.14 128.39 128.16 128.03 127.63 127.40 111.91 111.84
77.31 CDCl3 77.06 CDCl3 76.80 CDCl3 67.45 56.25 55.91 40.51 31.36 29.52 22.56
Ph
MeO
N
O
MeO
Cl
2d
[13]C NMR (126 MHz, Chloroform-d)
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)



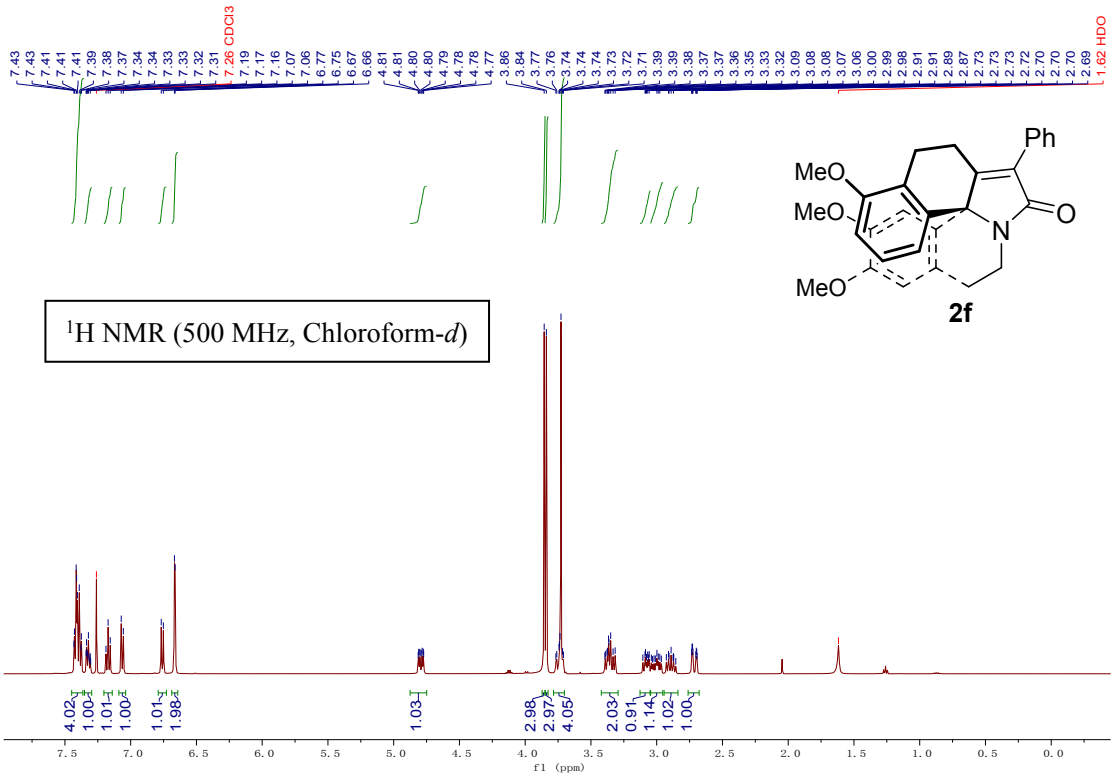
^{1}H NMR (500 MHz, Chloroform-*d*)

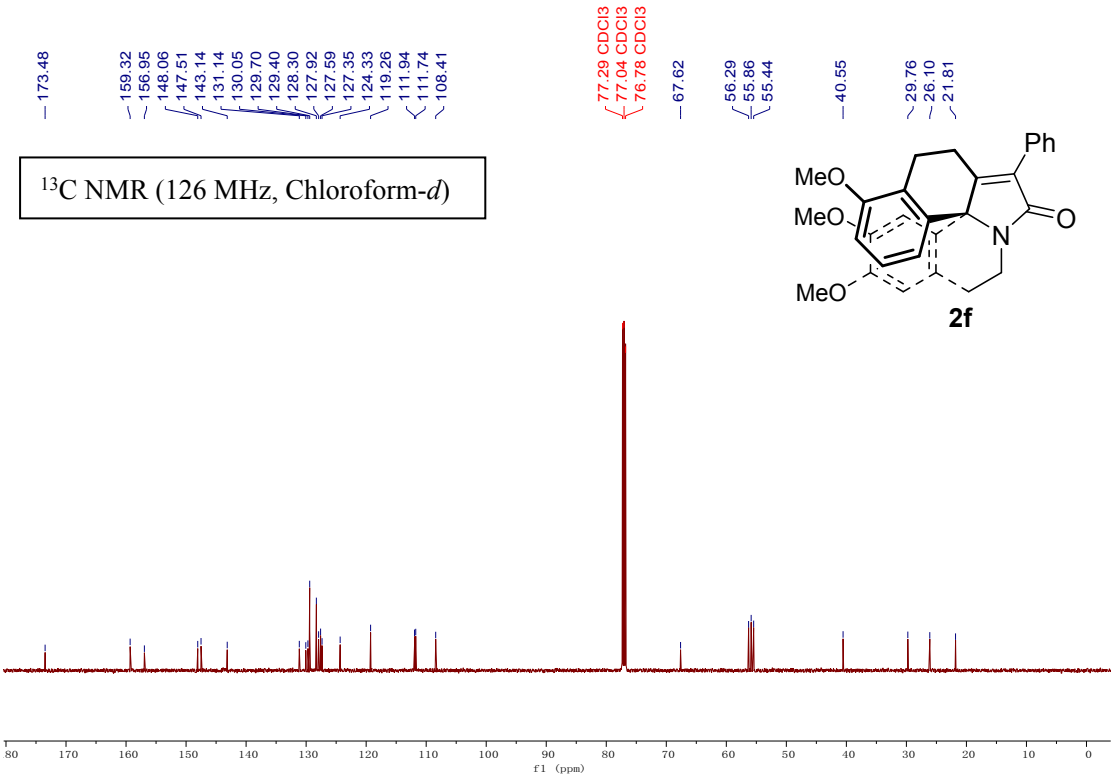
2e

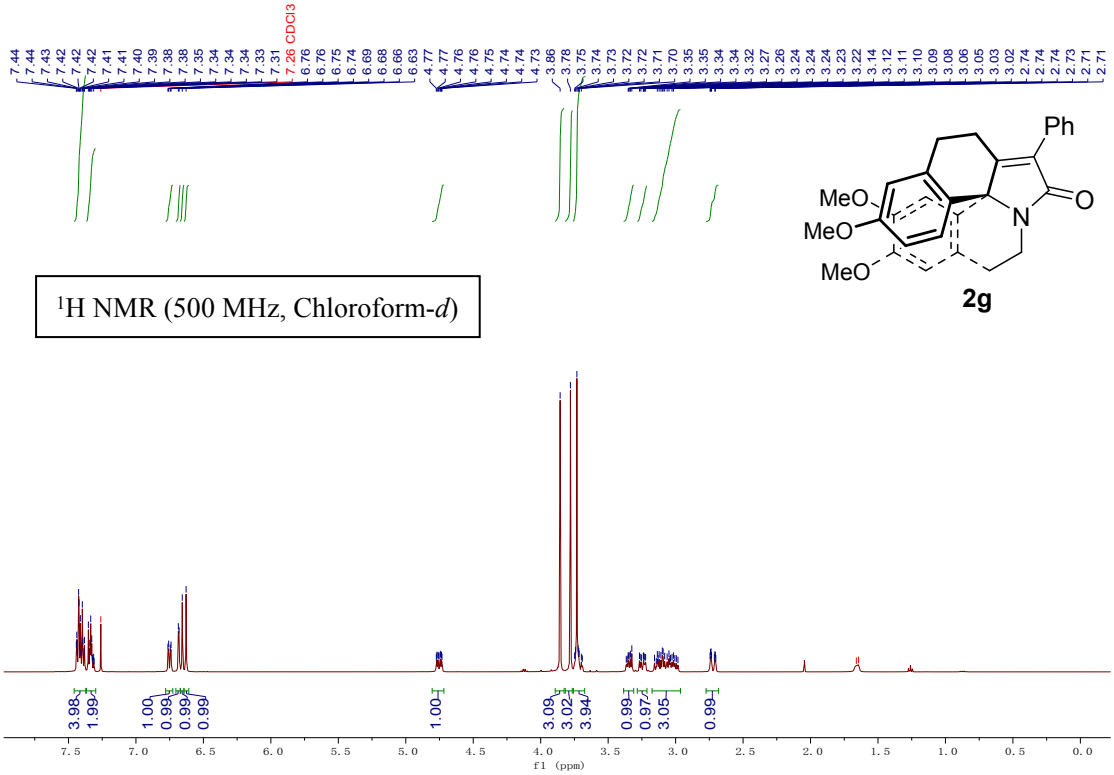


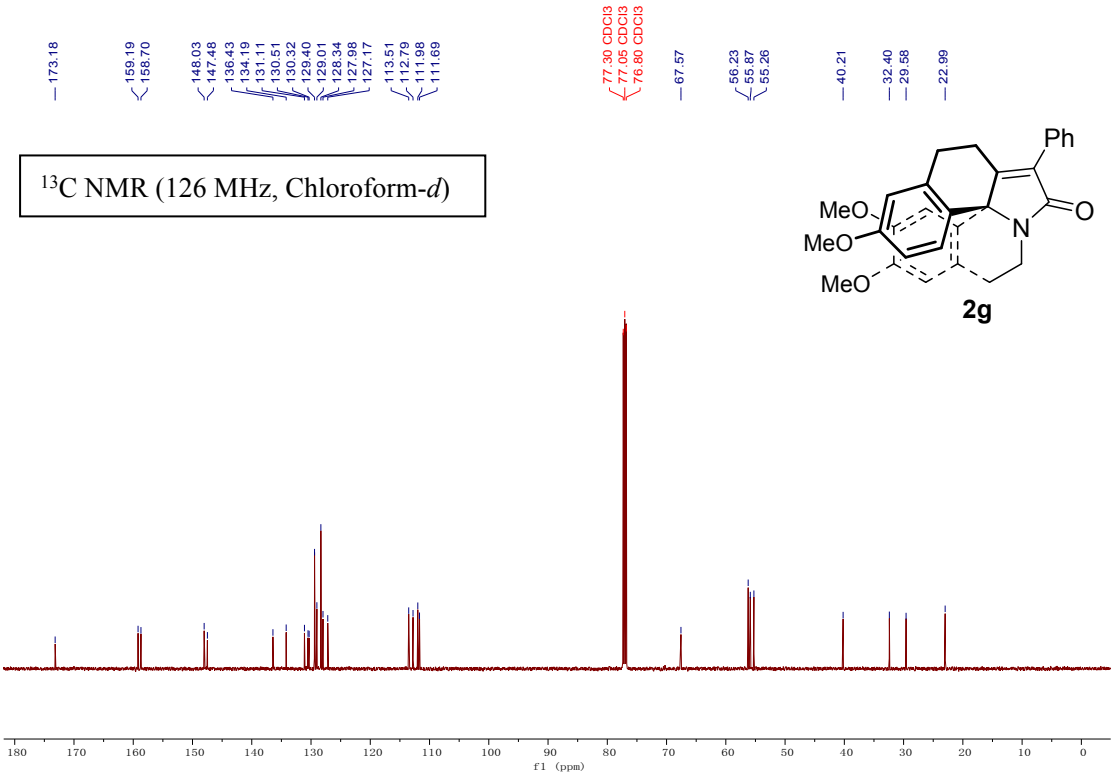
^{13}C NMR (126 MHz, Chloroform-*d*)

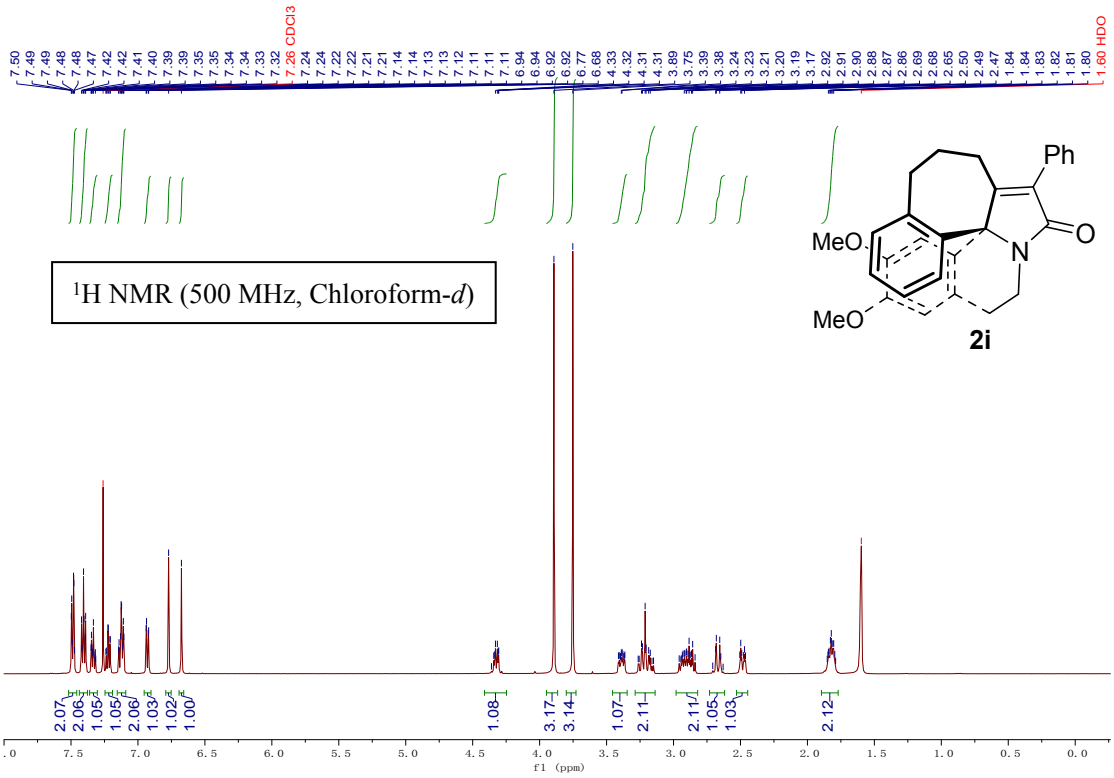
2e

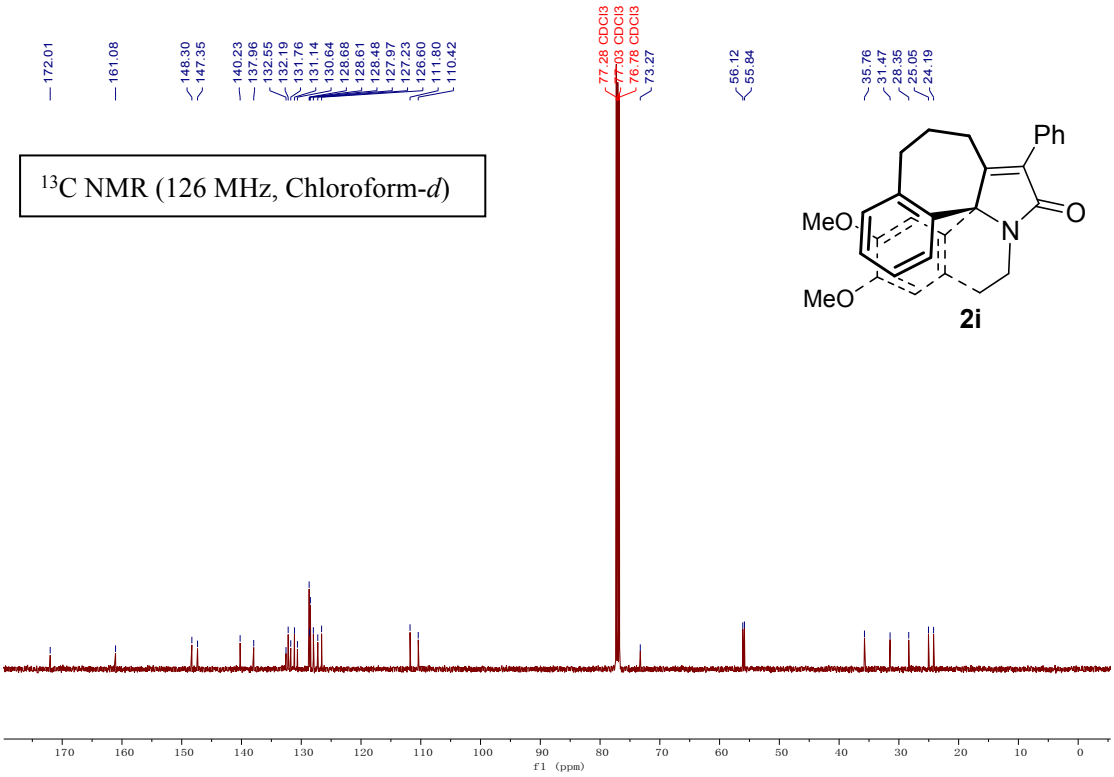

7.43 7.43 7.41 7.41 7.41 7.39 7.38 7.37 7.34 7.34 7.33 7.32 7.31 7.26 CDCl3 7.19 7.17 7.16 7.07 7.06 6.77 6.75 6.67 6.66 4.81 4.81 4.80 4.80 4.79 4.78 4.78 4.77 3.86 3.84 3.77 3.76 3.74 3.74 3.74 3.73 3.72 3.71 3.39 3.39 3.38 3.37 3.36 3.35 3.33 3.32 3.09 3.08 3.08 3.07 3.06 3.00 2.99 2.98 2.91 2.91 2.89 2.87 2.73 2.73 2.73 2.72 2.70 2.70 2.69 1.62 HDO
Ph
MeO
MeO
MeO
O
N
2f
1H NMR (500 MHz, Chloroform-d)
4.02 1.00 1.01 1.00 1.01 1.98 1.03 2.98 2.97 4.05 2.03 0.91 1.14 1.02 1.00
f1 (ppm)

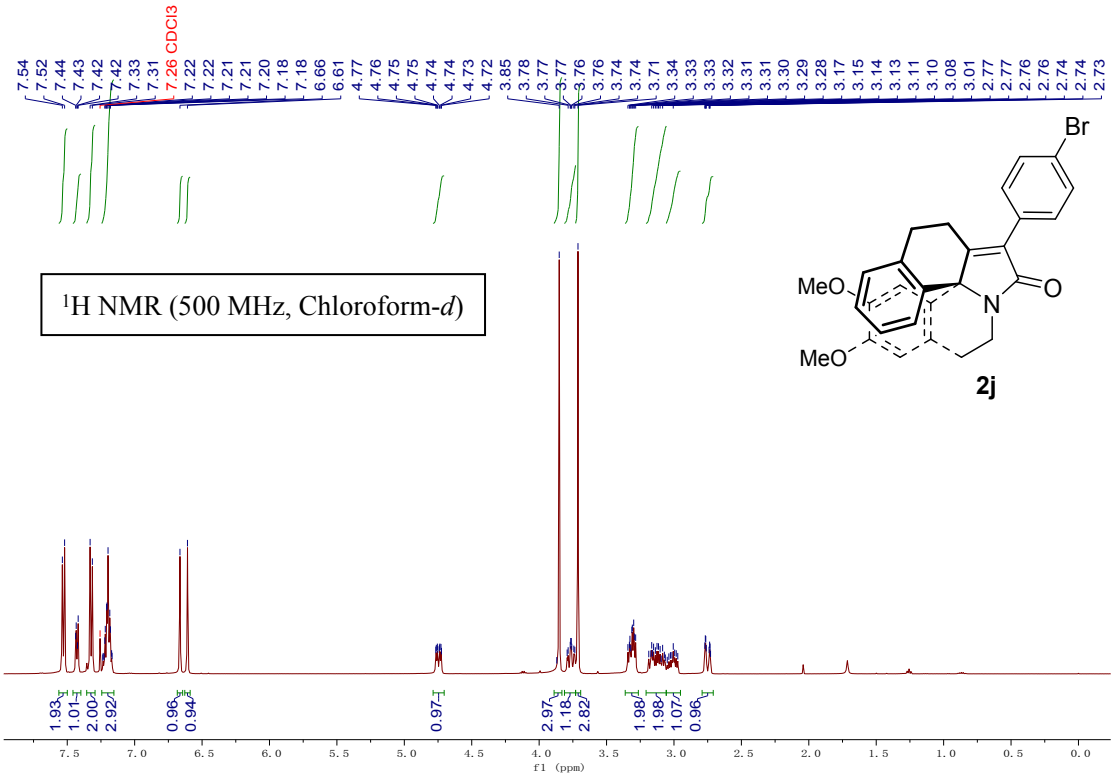

173.48 159.32 156.95 148.06 147.51 143.14 131.14 130.05 129.70 129.40 128.30 127.92 127.59 127.35 124.33 119.26 111.94 111.74 108.41
77.29 CDCl3 77.04 CDCl3 76.78 CDCl3 67.62 56.29 55.86 55.44 40.55 29.76 26.10 21.81
Ph
MeO
MeO
MeO
O
N
2f
13C NMR (126 MHz, Chloroform-d)
f1 (ppm)


7.44 7.44 7.43 7.42 7.42 7.42 7.41 7.41 7.40 7.39 7.38 7.38 7.35 7.34 7.34 7.34 7.33 7.31 7.26 CDCl3 6.76 6.76 6.75 6.74 6.69 6.68 6.66 6.63 4.77 4.76 4.76 4.75 4.74 4.74 4.73 3.86 3.78 3.75 3.74 3.73 3.72 3.71 3.70 3.35 3.35 3.34 3.34 3.32 3.27 3.26 3.24 3.24 3.24 3.23 3.22 3.14 3.12 3.11 3.10 3.09 3.08 3.06 3.05 3.03 3.02 2.74 2.74 2.74 2.73 2.71 2.71
Ph
MeO
MeO
MeO
N
O
2g
[1]H NMR (500 MHz, Chloroform-d)
3.98 1.99 1.00 0.99 0.99 0.99 1.00 3.09 3.02 3.94 0.99 0.97 3.05 0.99
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


173.18
159.19 158.70
148.03 147.48 136.43 134.19 131.11 130.51 130.32 129.40 129.01 128.34 127.98 127.17 113.51 112.79 111.98 111.69
77.30 CDCl3 77.05 CDCl3 76.80 CDCl3
67.57
56.23 55.87 55.26
40.21
32.40 29.58
22.99
Ph
MeO
MeO
MeO
N
O
2g
[13]C NMR (126 MHz, Chloroform-d)
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

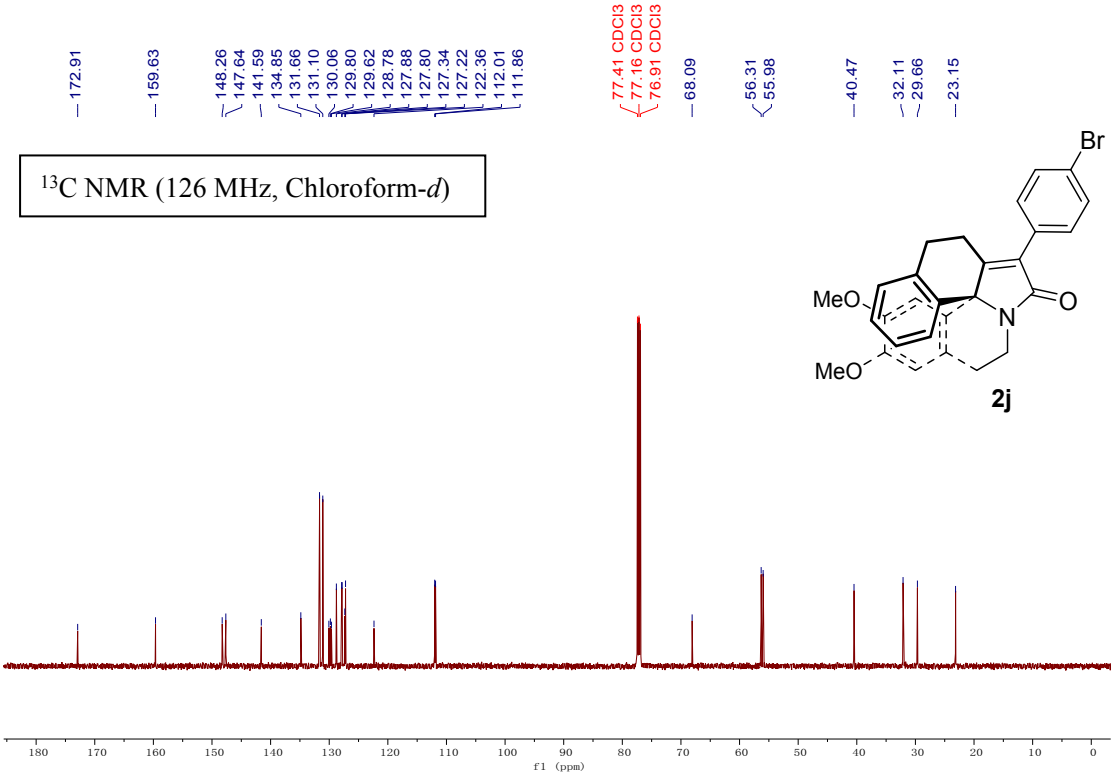

7.50 7.49 7.49 7.48 7.48 7.47 7.42 7.42 7.41 7.40 7.39 7.39 7.35 7.35 7.34 7.34 7.33 7.32 7.26 CDCl3 7.24 7.24 7.22 7.21 7.21 7.14 7.13 7.13 7.12 7.11 7.11 7.11 6.94 6.94 6.92 6.92 6.77 6.68 4.33 4.32 4.31 4.31 3.89 3.75 3.39 3.38 3.24 3.23 3.21 3.20 3.19 3.17 2.92 2.91 2.90 2.88 2.87 2.86 2.69 2.68 2.65 2.50 2.49 2.47 1.84 1.84 1.83 1.82 1.81 1.80 1.60 HDO
[1]H NMR (500 MHz, Chloroform-d)
Ph
O
N
MeO
MeO
2i
2.07 2.06 1.05 1.05 2.06 1.03 1.02 1.00 1.08 3.17 3.14 1.07 2.11 2.11 1.05 1.03 2.12
f1 (ppm)


172.01 161.08 148.30 147.35 140.23 137.96 132.55 132.19 131.76 131.14 130.64 128.68 128.61 128.48 127.97 127.23 126.60 111.80 110.42 77.28 CDCl3 77.03 CDCl3 76.78 CDCl3 73.27 56.12 55.84 35.76 31.47 28.35 25.05 24.19
[13]C NMR (126 MHz, Chloroform-d)
Ph
O
N
MeO
MeO
2i
f1 (ppm)



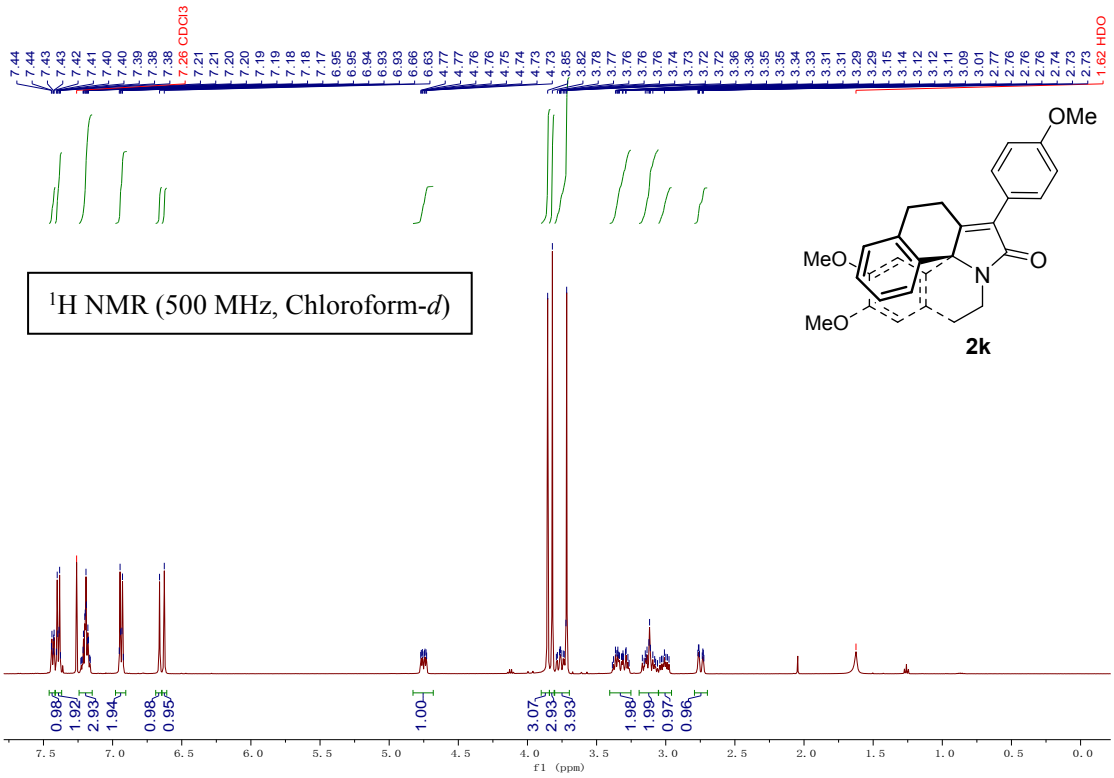
^{1}H NMR (500 MHz, Chloroform-*d*)

2j



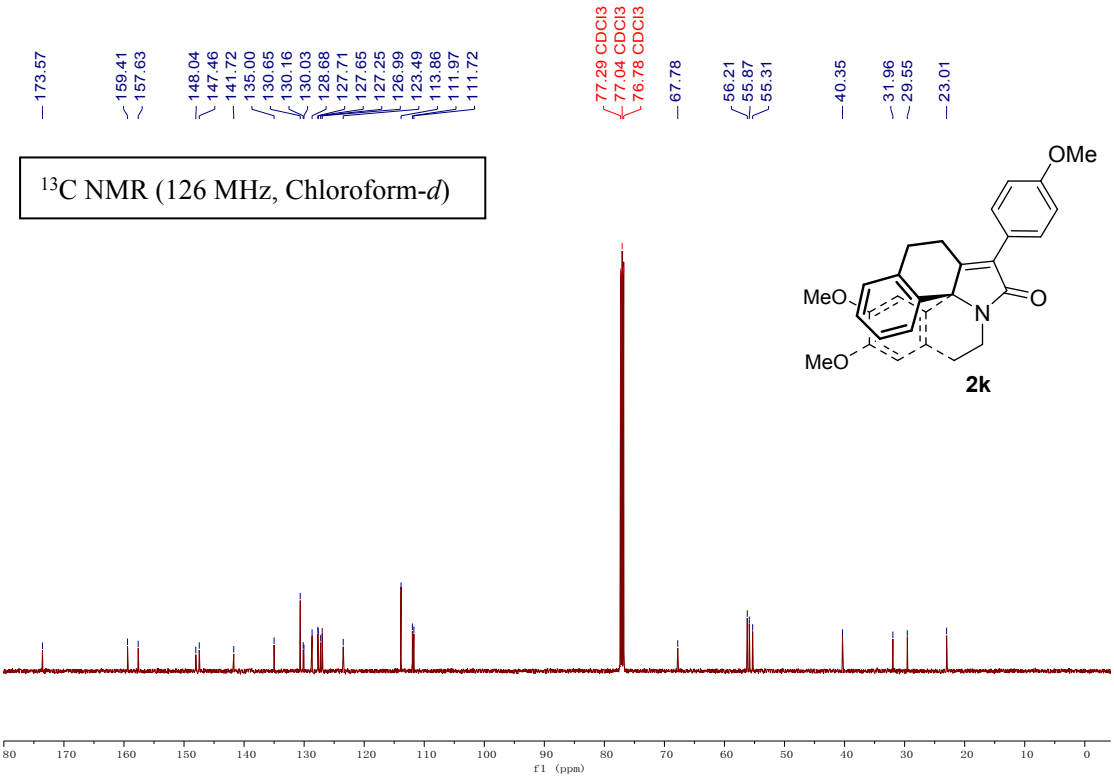
^{13}C NMR (126 MHz, Chloroform-*d*)

2j



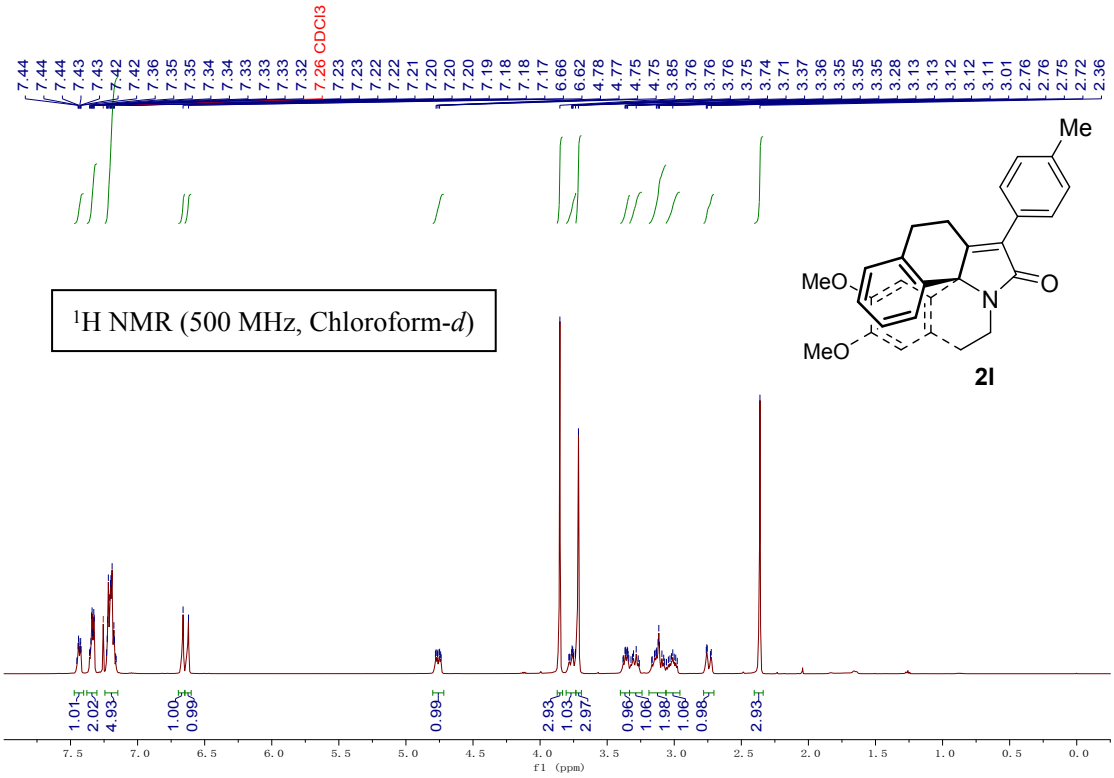
^{1}H NMR (500 MHz, Chloroform-*d*)

2k

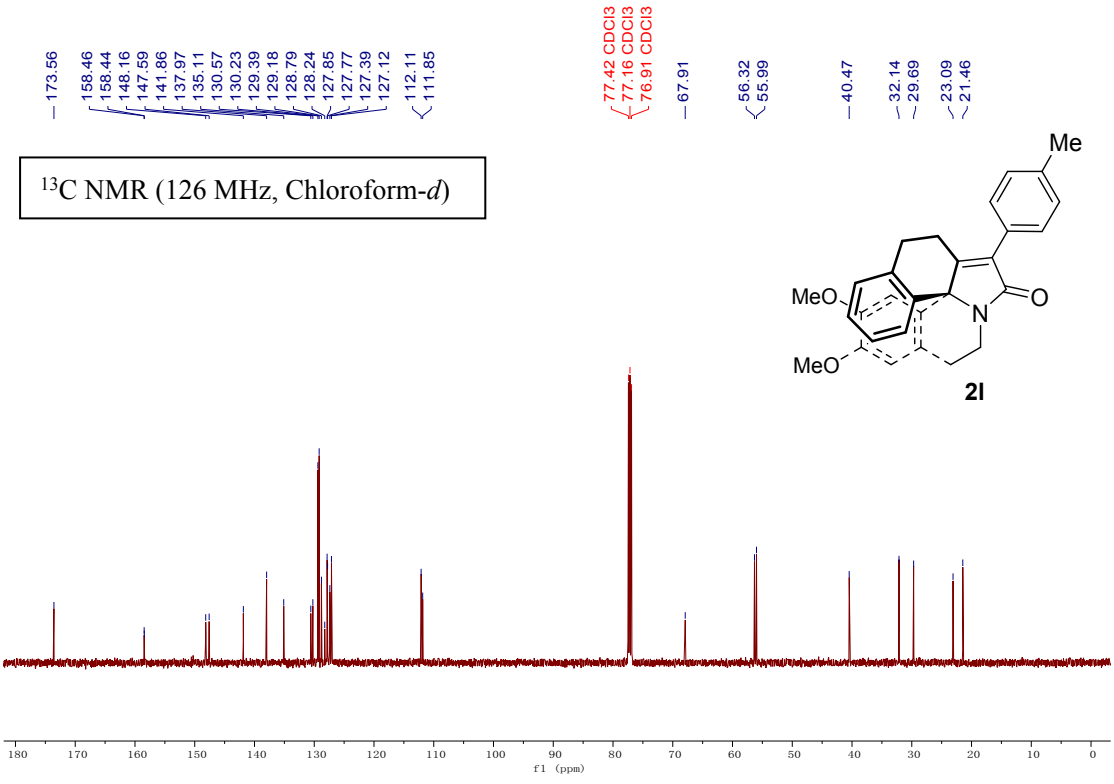


^{13}C NMR (126 MHz, Chloroform-*d*)

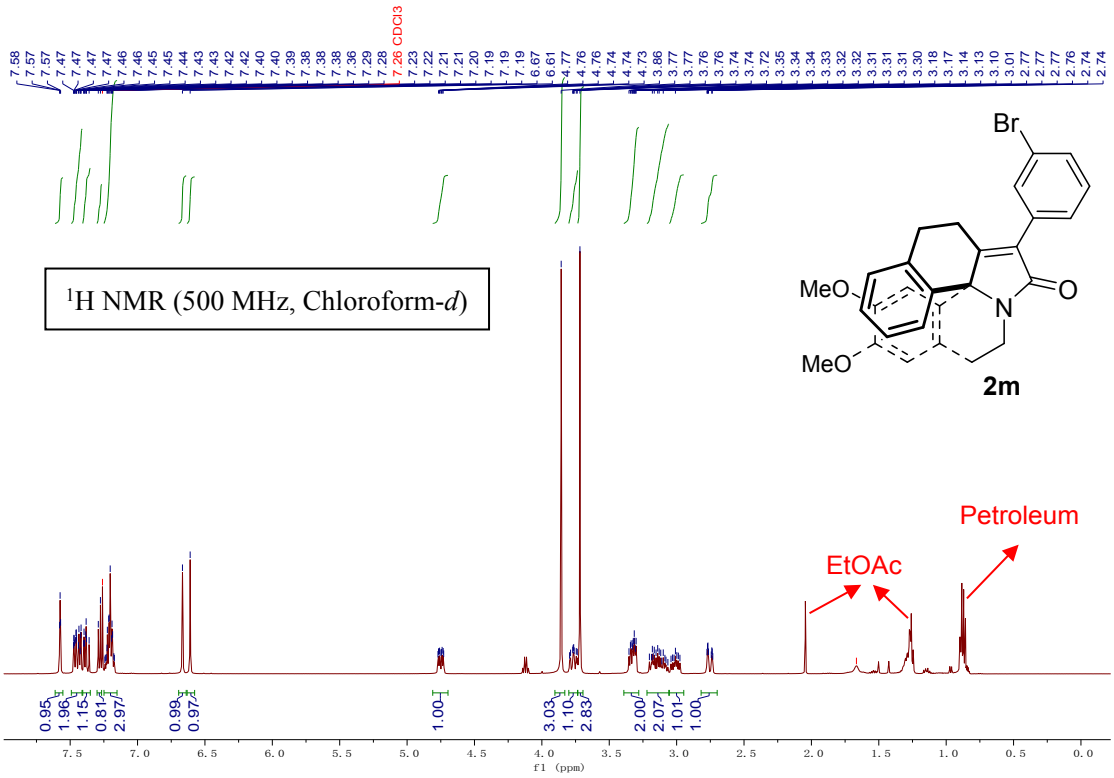
2k



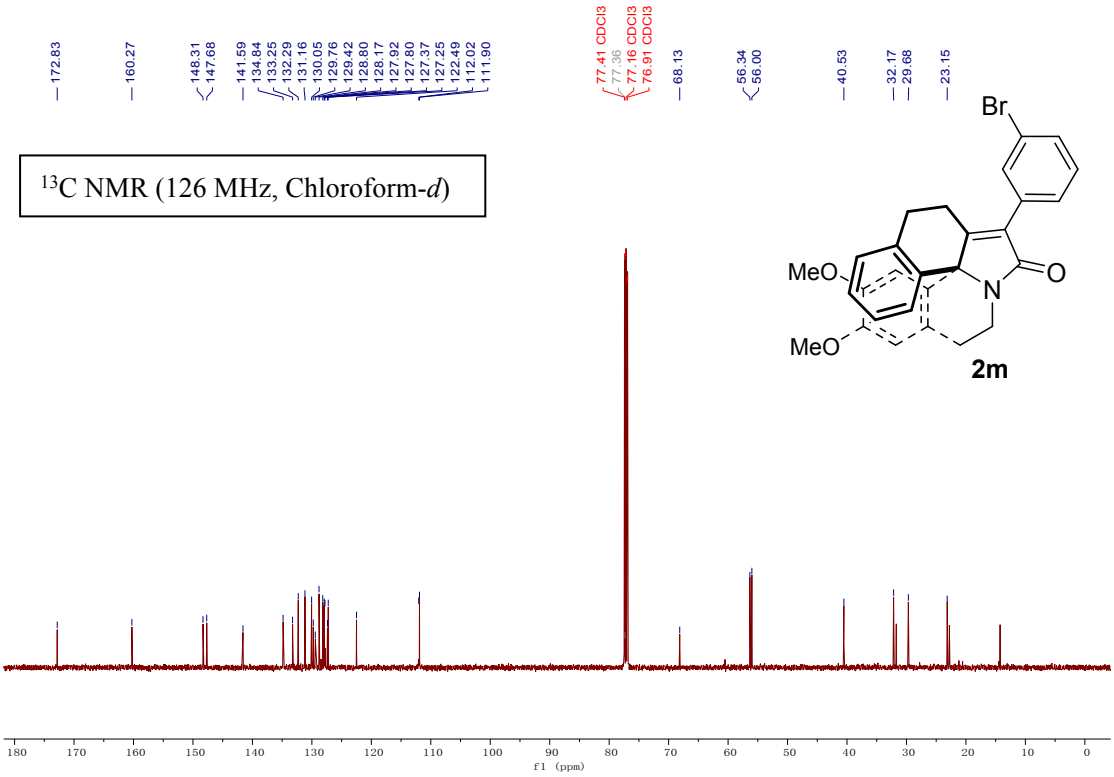
^{1}H NMR (500 MHz, Chloroform-*d*)



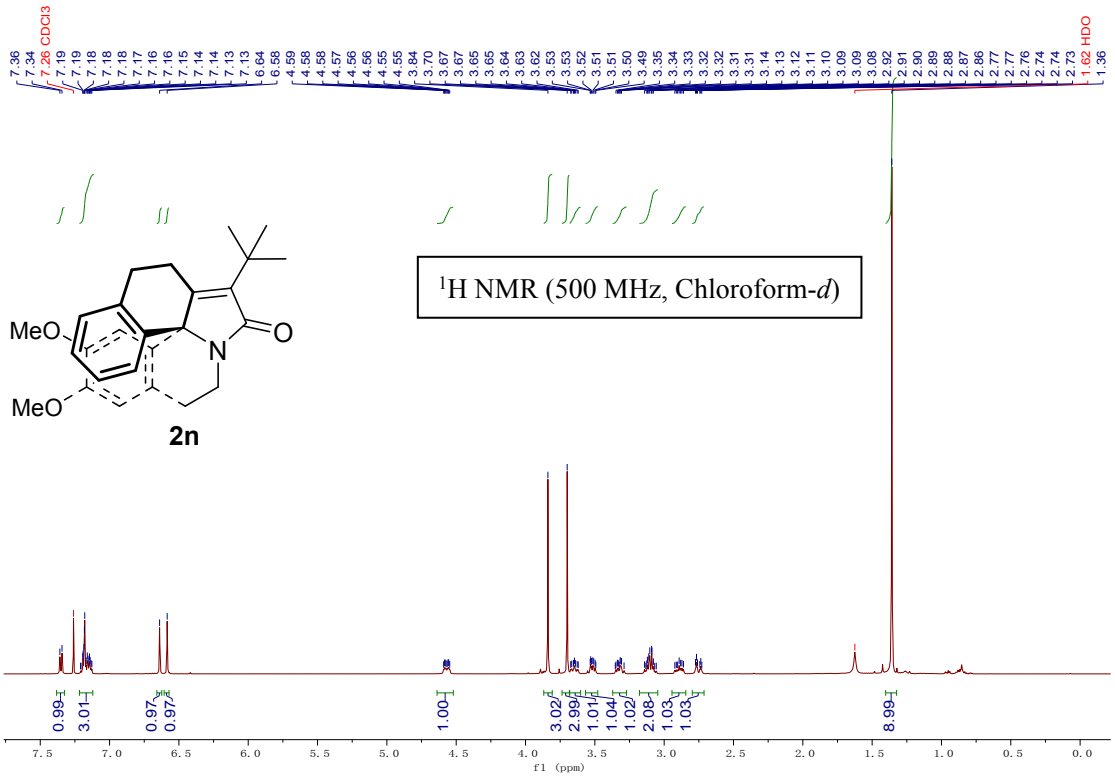
^{13}C NMR (126 MHz, Chloroform-*d*)

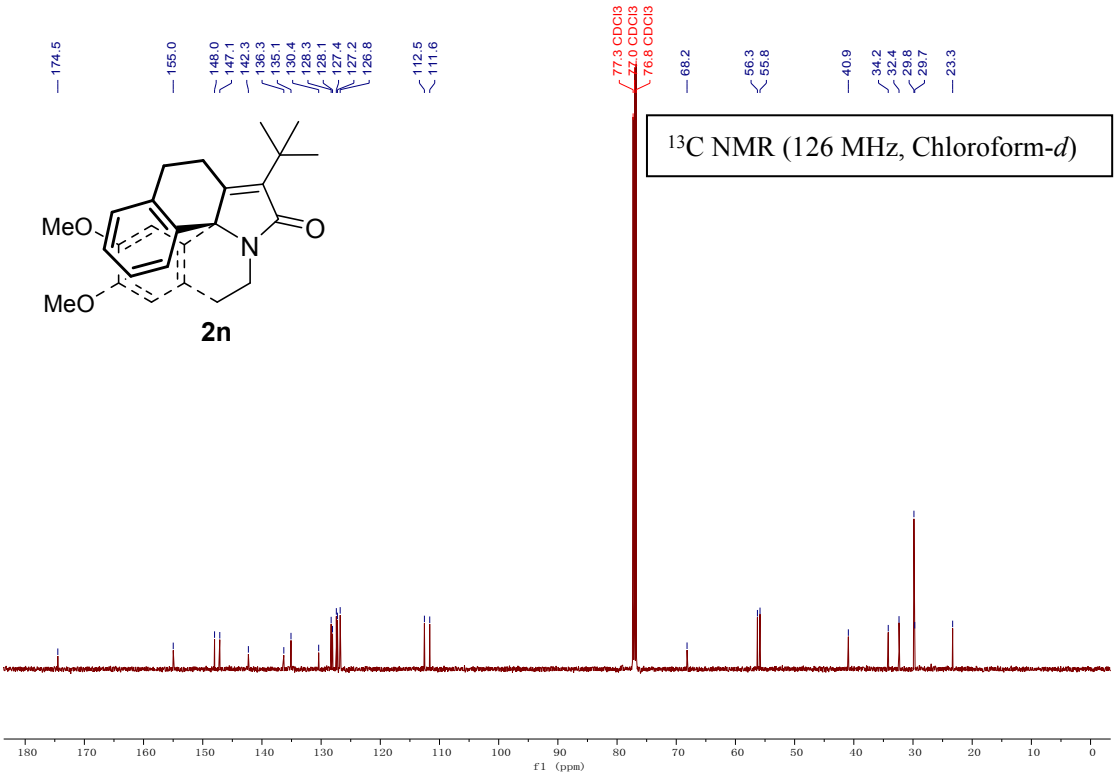


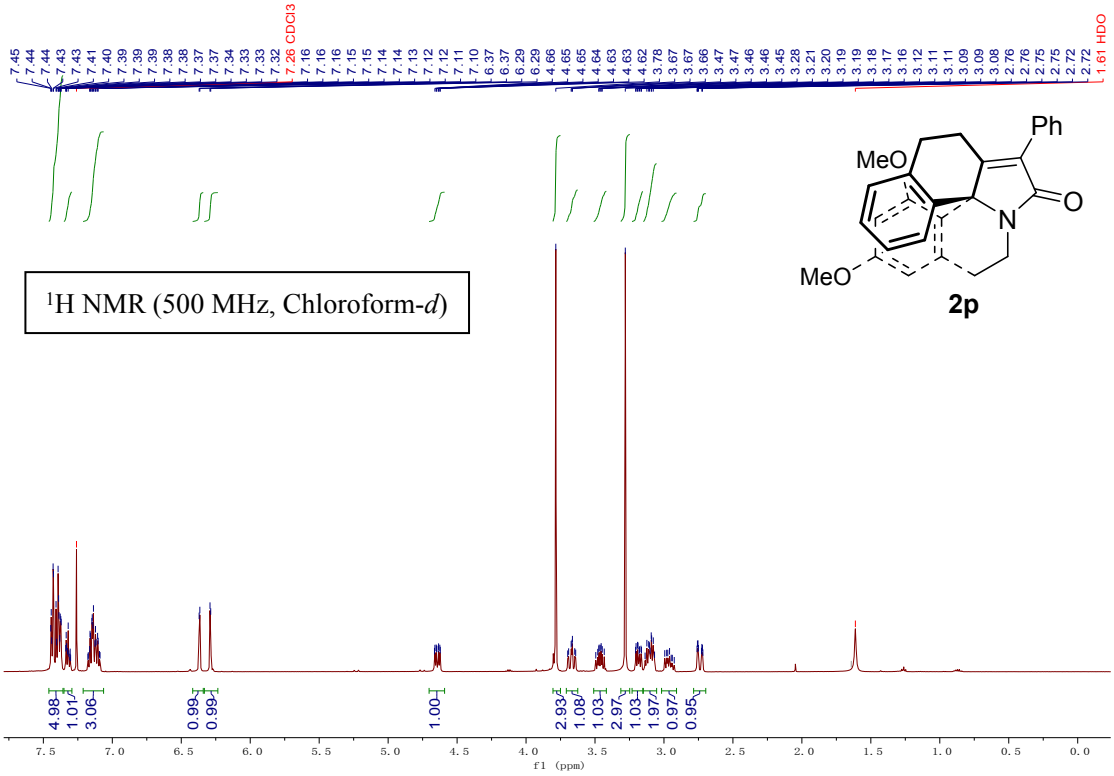
^{1}H NMR (500 MHz, Chloroform-*d*)

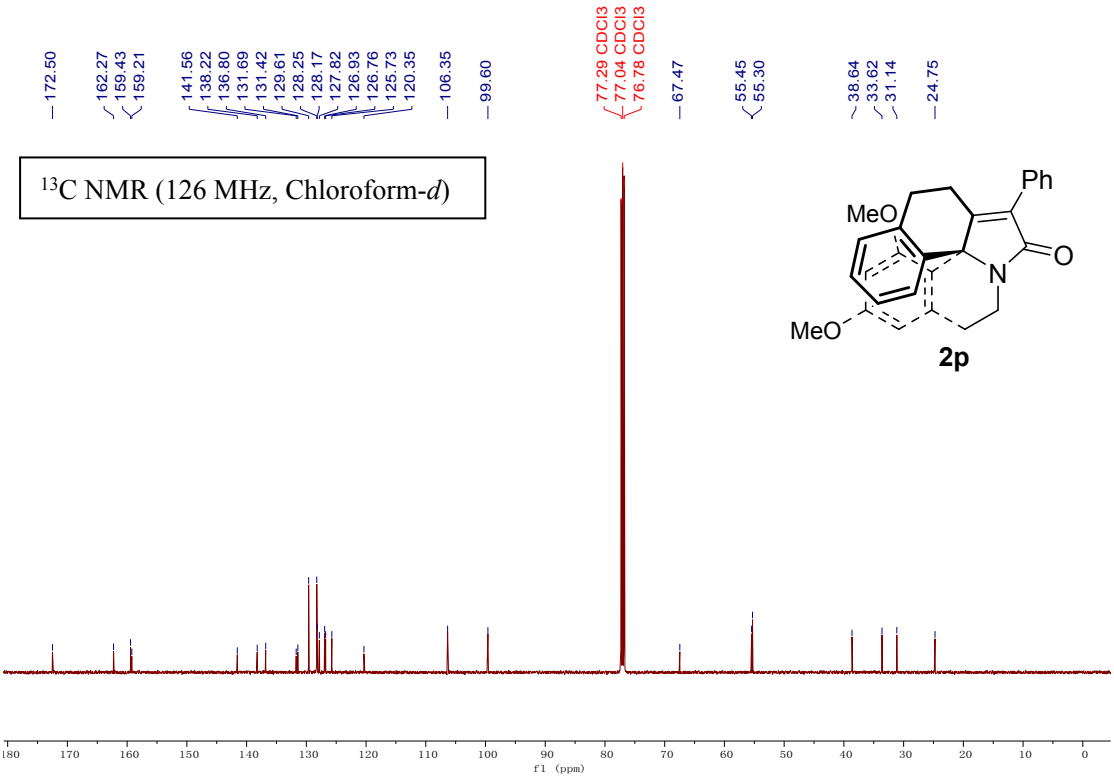


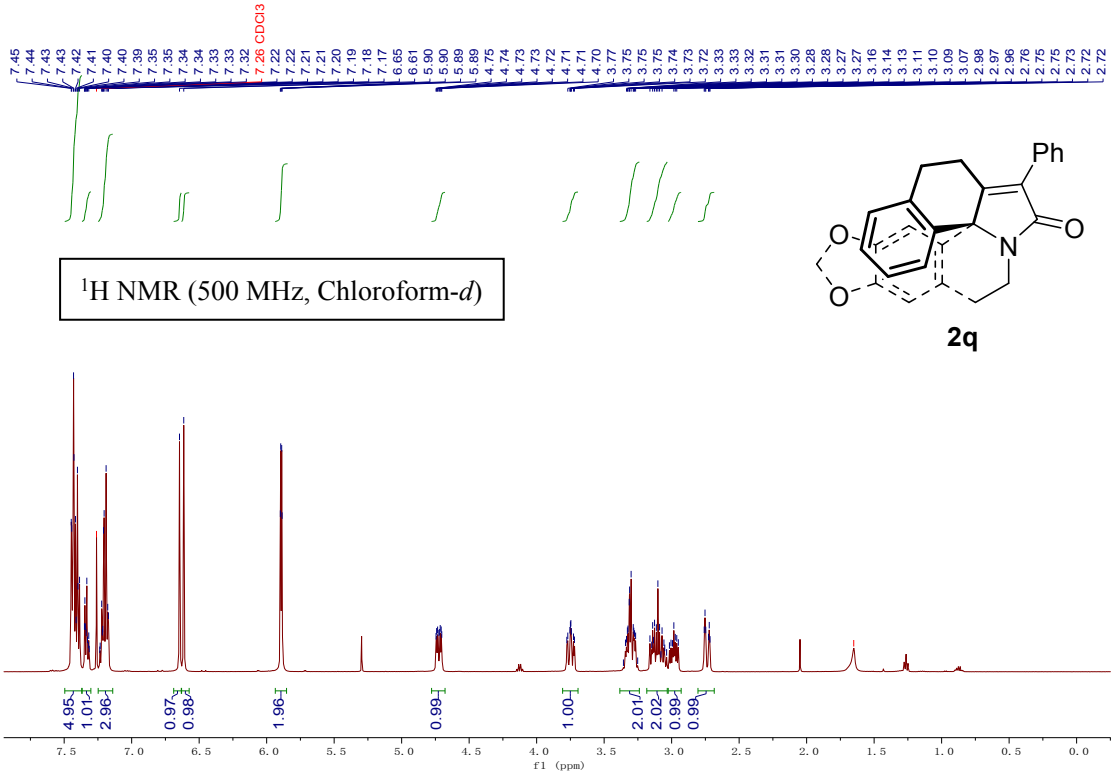
^{13}C NMR (126 MHz, Chloroform-*d*)


MeO
MeO
N
O
2n
^{1}H NMR (500 MHz, Chloroform-*d*)
f1 (ppm)


MeO
MeO
N
O
2n
^{13}C NMR (126 MHz, Chloroform-*d*)
f1 (ppm)

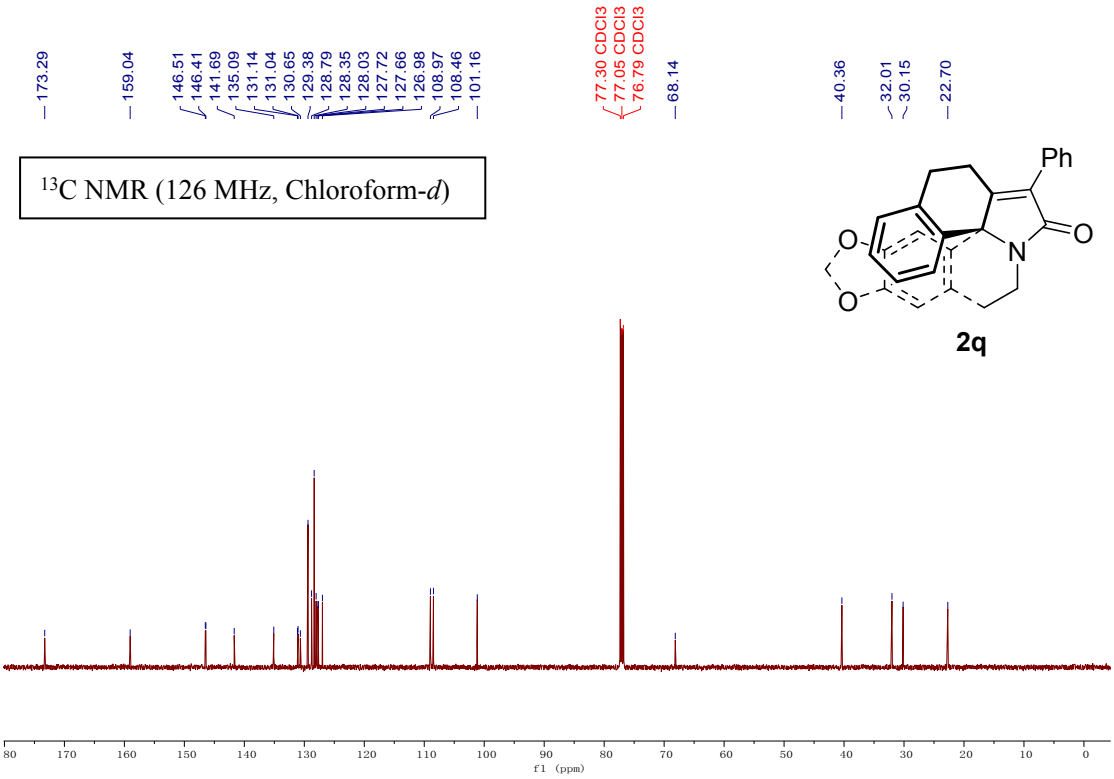

7.45 7.44 7.44 7.43 7.43 7.41 7.40 7.39 7.39 7.38 7.38 7.37 7.34 7.33 7.32 7.26 CDCl3 7.16 7.16 7.16 7.15 7.15 7.14 7.13 7.12 7.11 7.10 6.37 6.37 6.29 6.29 4.66 4.65 4.64 4.63 4.63 4.62 3.78 3.67 3.67 3.66 3.47 3.47 3.46 3.46 3.45 3.28 3.21 3.20 3.19 3.19 3.18 3.17 3.16 3.12 3.11 3.11 3.09 3.09 3.08 2.76 2.76 2.75 2.75 2.72 2.72 1.61 HDO
Ph
MeO
O
N
MeO
2p
1H NMR (500 MHz, Chloroform-d)
4.98 1.01 3.06 0.99 0.99 1.00 2.93 1.08 1.03 2.97 1.03 1.97 0.97 0.95
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


172.50 162.27 159.43 159.21 141.56 138.22 136.80 131.69 131.42 129.61 128.25 128.17 127.82 126.93 126.76 125.73 120.35 106.35 99.60 77.29 CDCl3 77.04 CDCl3 76.78 CDCl3 67.47 55.45 55.30 38.64 33.62 31.14 24.75
13C NMR (126 MHz, Chloroform-d)
Ph
MeO
O
N
MeO
2p
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)



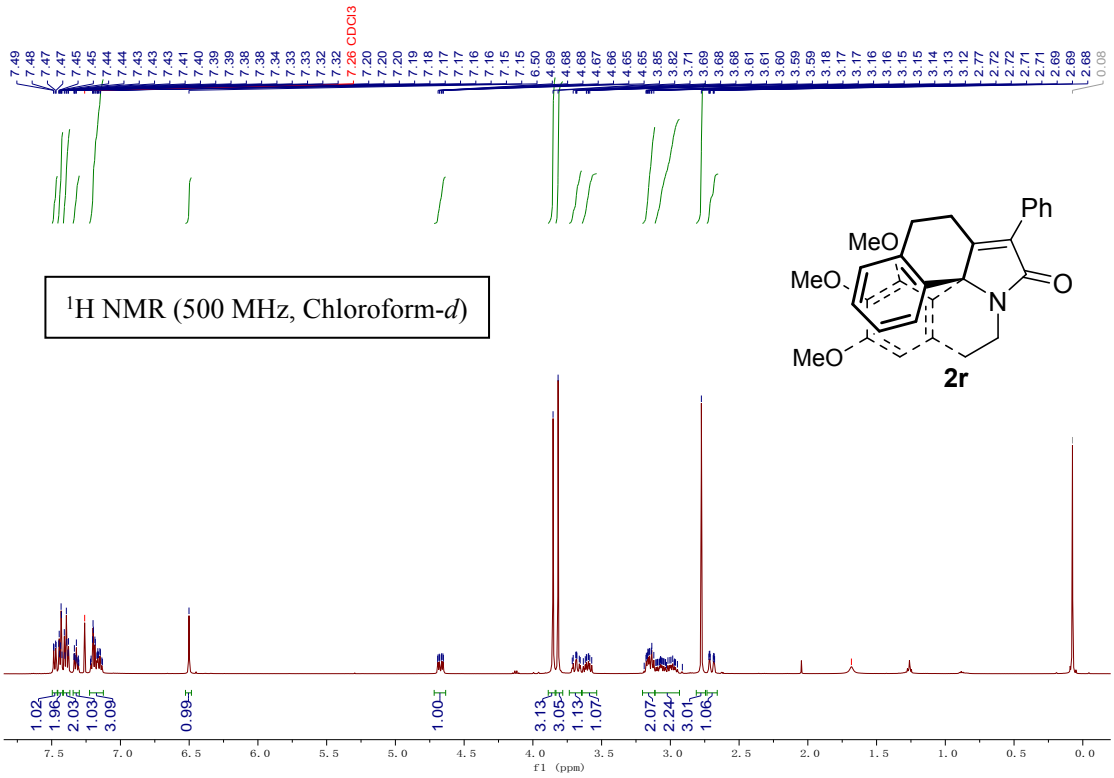
^{1}H NMR (500 MHz, Chloroform-*d*)

2q



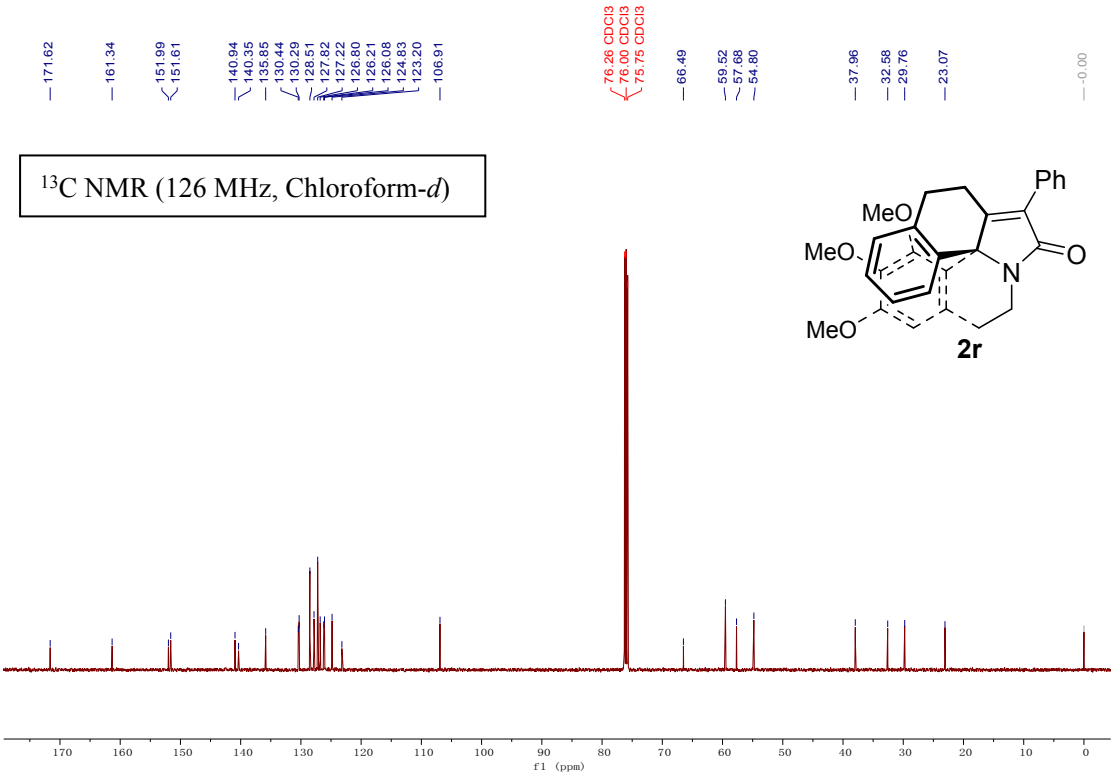
^{13}C NMR (126 MHz, Chloroform-*d*)

2q



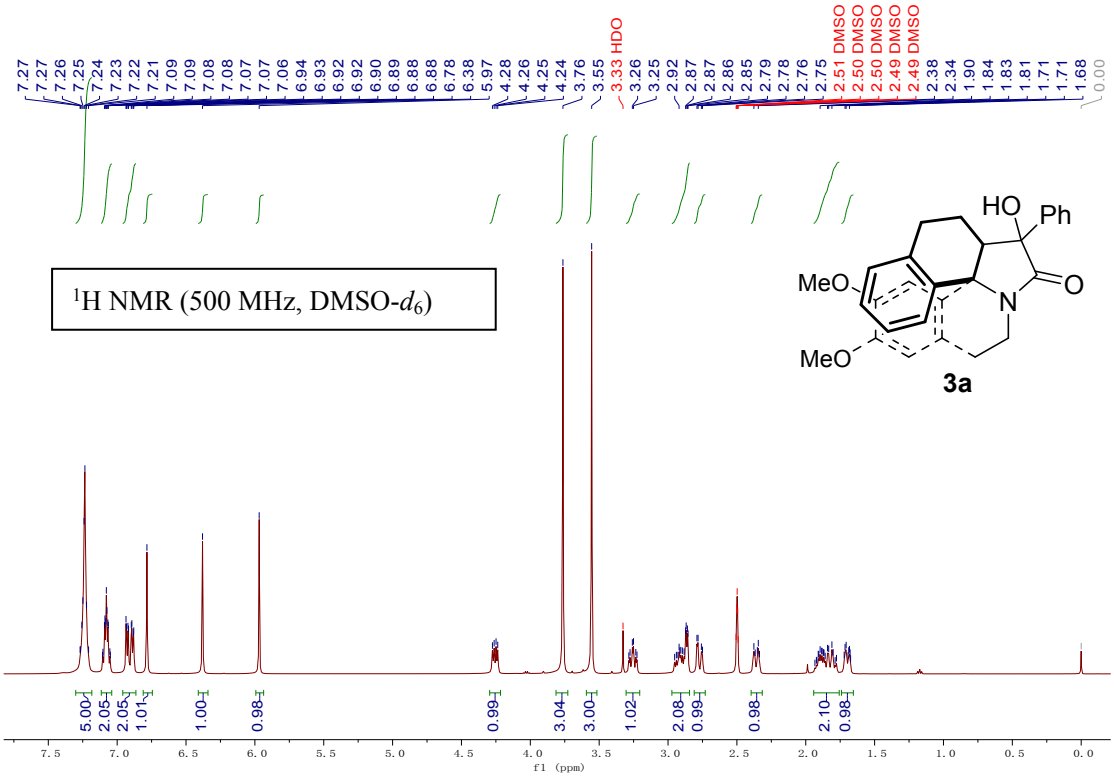
^{1}H NMR (500 MHz, Chloroform-*d*)

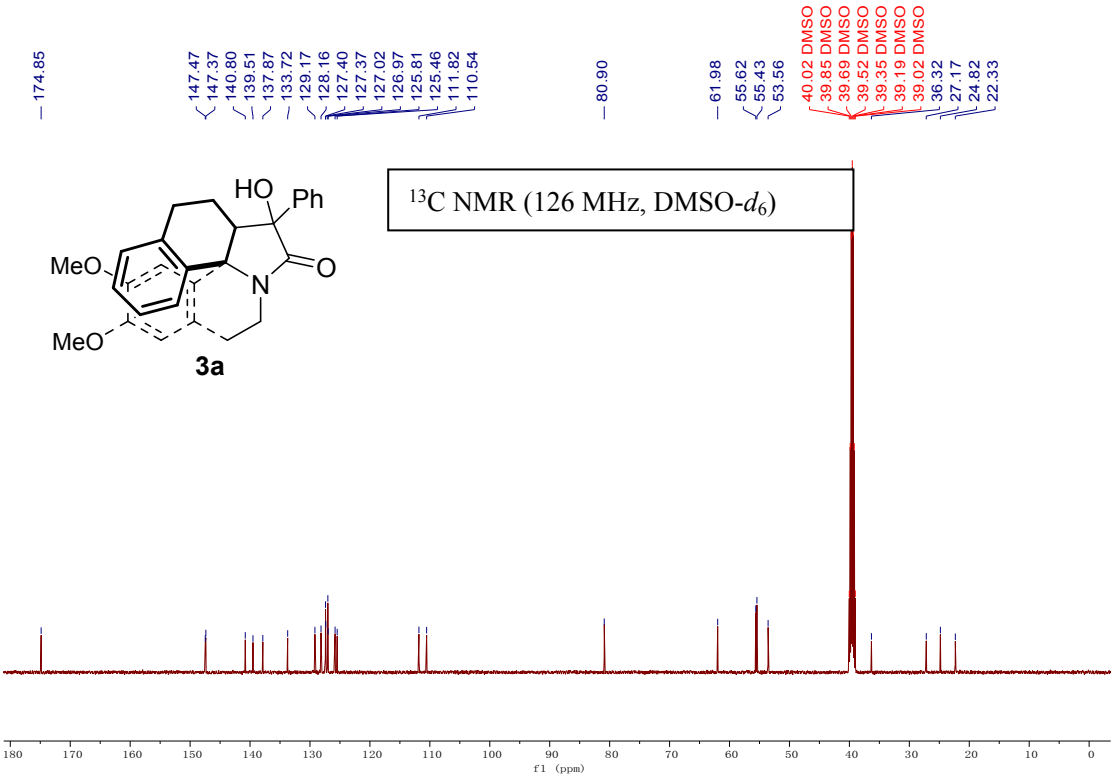
2r

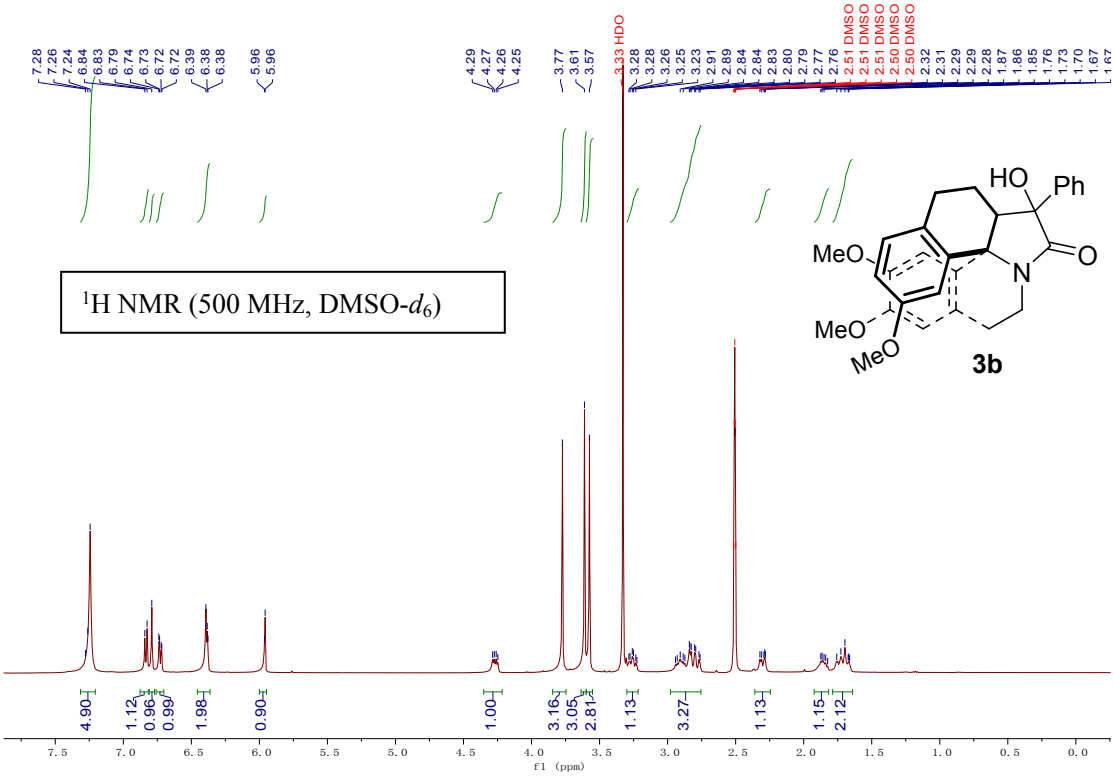


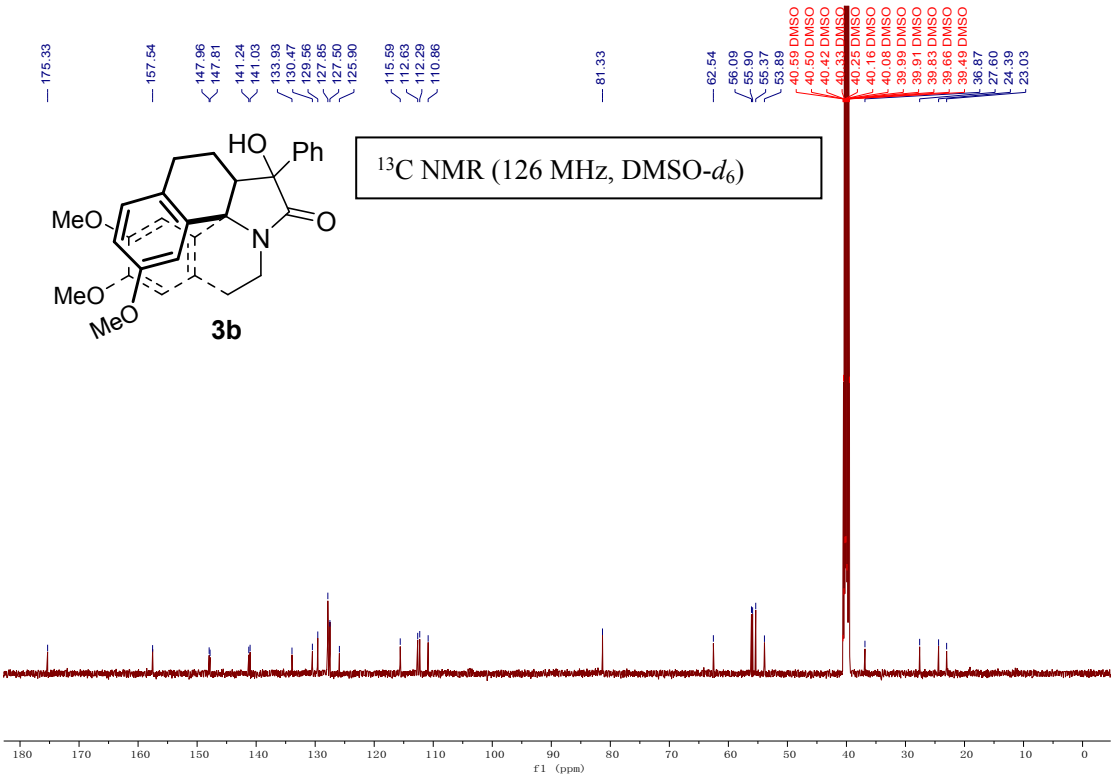
^{13}C NMR (126 MHz, Chloroform-*d*)

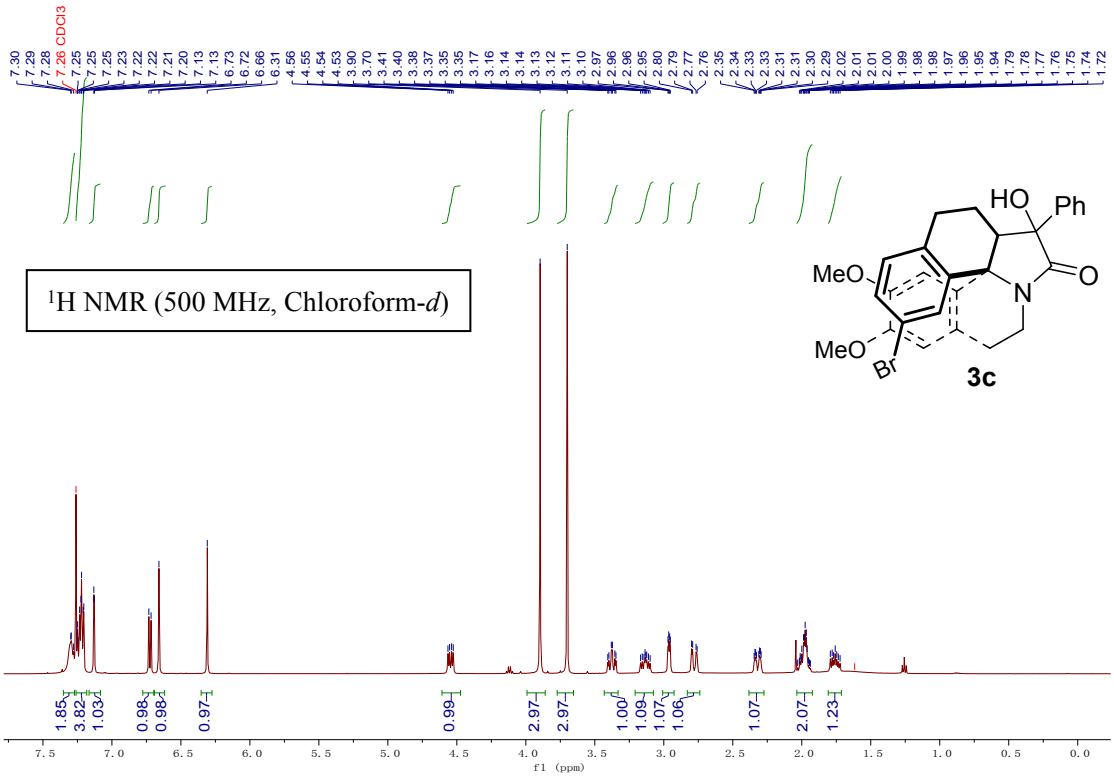
2r

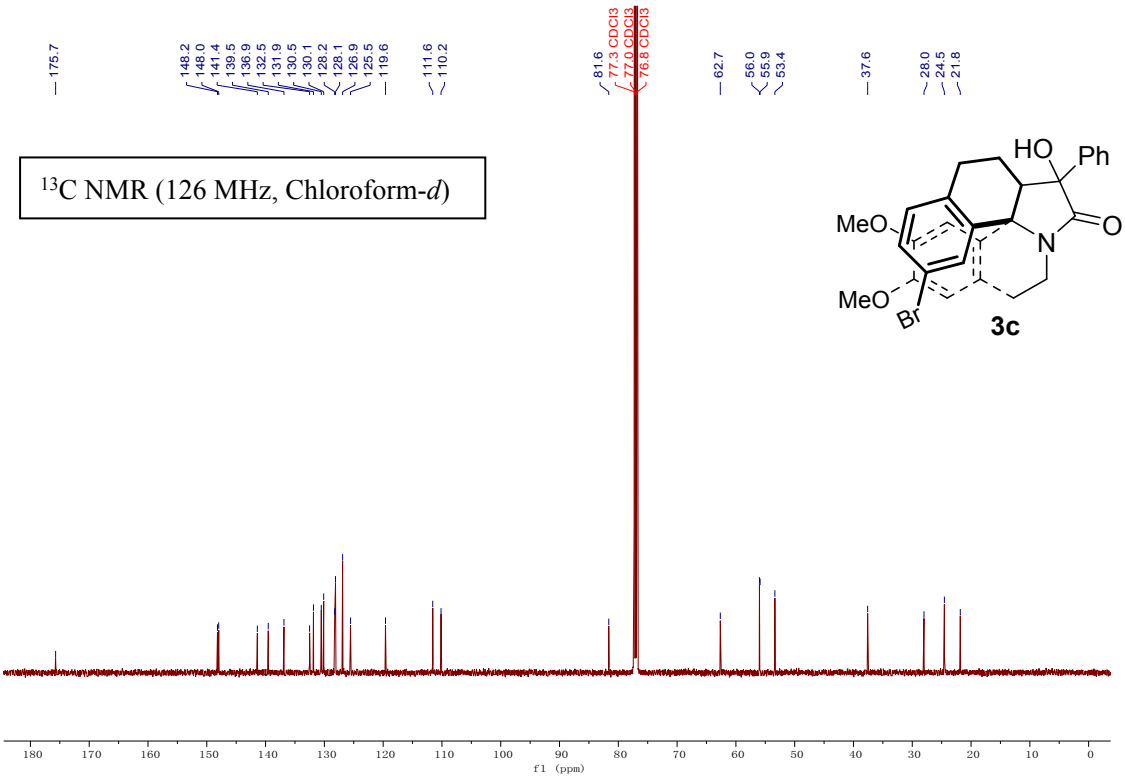

^{1}H NMR (500 MHz, DMSO-d_6)
HO
Ph
MeO
MeO
N
O
3a

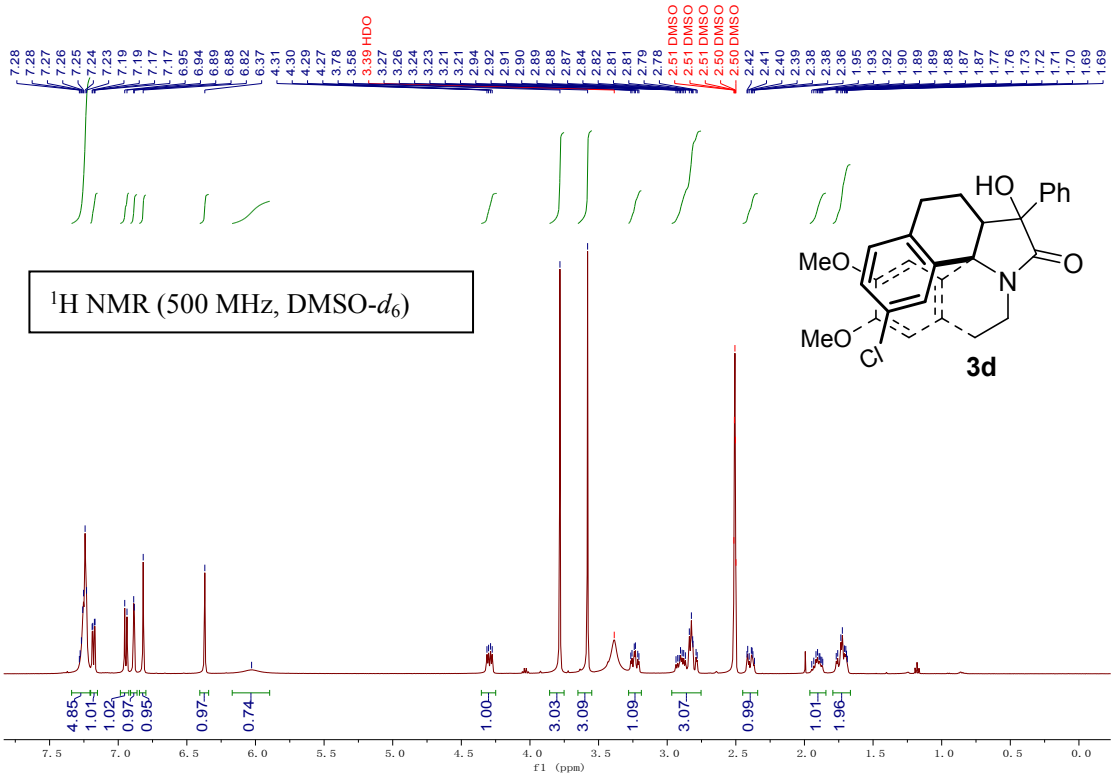

^{13}C NMR (126 MHz, DMSO-d_6)
HO
Ph
MeO
MeO
N
O
3a

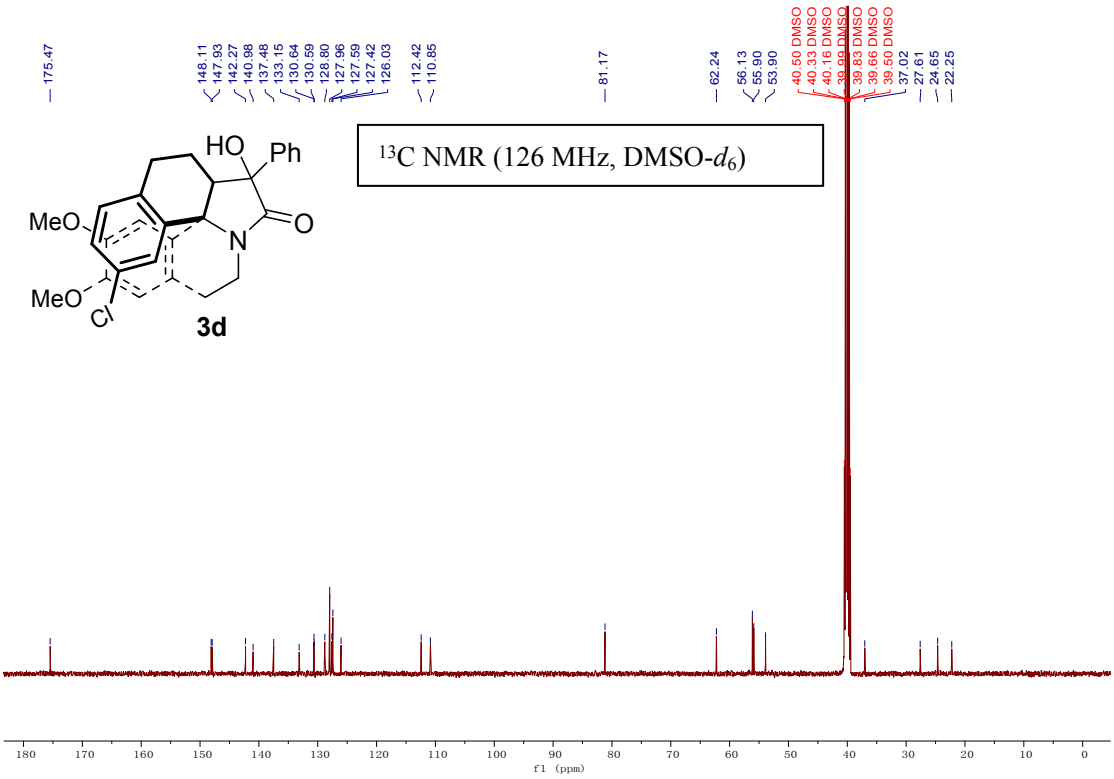

^{1}H NMR (500 MHz, DMSO-d_6)
HO
Ph
MeO
MeO
MeO
N
O
3b

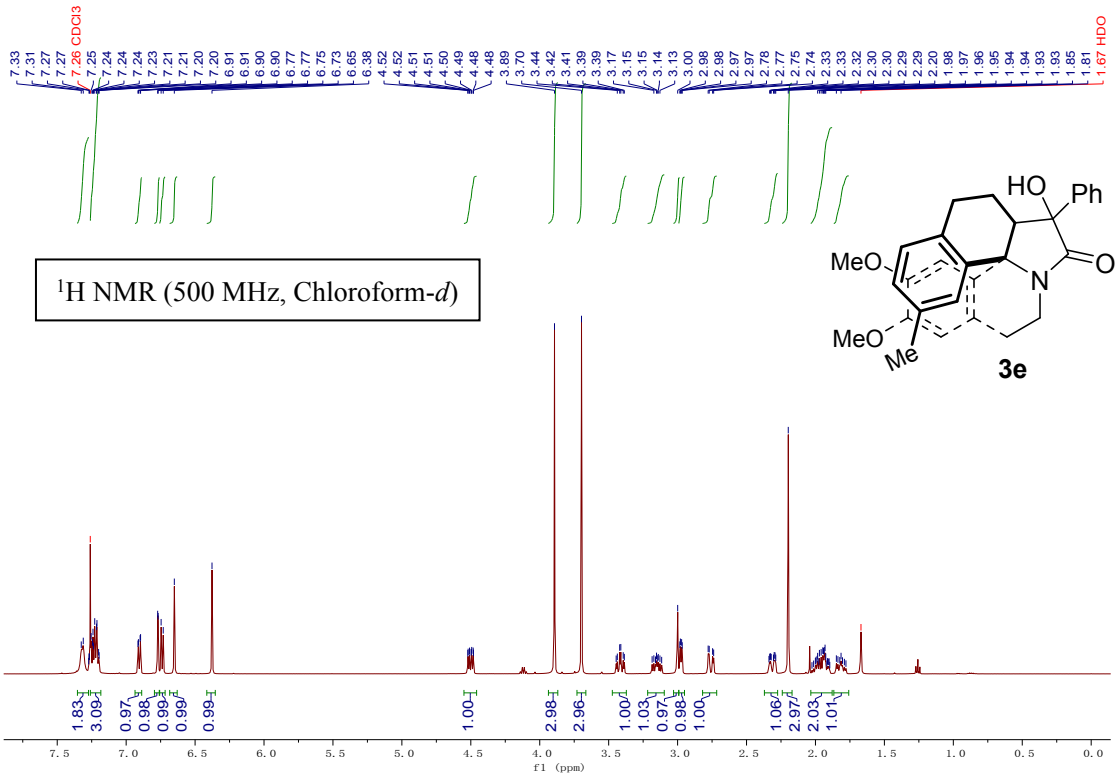

^{13}C NMR (126 MHz, DMSO-d_6)
HO
Ph
MeO
MeO
MeO
N
O
3b

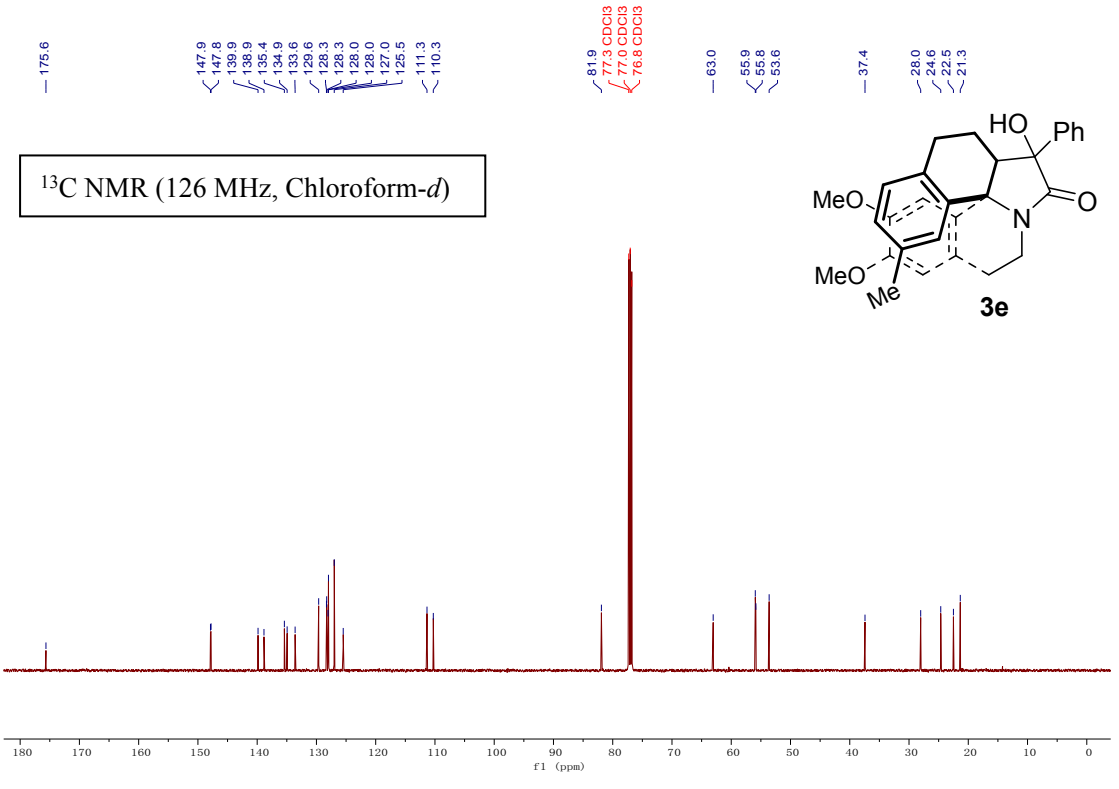

7.30 7.29 7.28 7.26 CDCl3 7.25 7.25 7.25 7.23 7.22 7.22 7.21 7.20 7.13 7.13 6.73 6.72 6.66 6.31
4.56 4.55 4.54 4.53 3.90 3.70 3.41 3.40 3.38 3.37 3.35 3.35 3.17 3.16 3.14 3.14 3.13 3.12 3.11 3.10 2.97 2.96 2.96 2.95 2.80 2.79 2.77 2.76 2.35 2.34 2.33 2.33 2.31 2.31 2.30 2.29 2.02 2.01 2.01 2.00 1.99 1.98 1.98 1.97 1.96 1.95 1.94 1.79 1.78 1.77 1.76 1.75 1.74 1.72
1H NMR (500 MHz, Chloroform-d)
HO
Ph
MeO
O
N
MeO
Br
3c
1.85 3.82 1.03 0.98 0.98 0.97 0.99 2.97 2.97 1.00 1.09 1.07 1.06 1.07 2.07 1.23
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

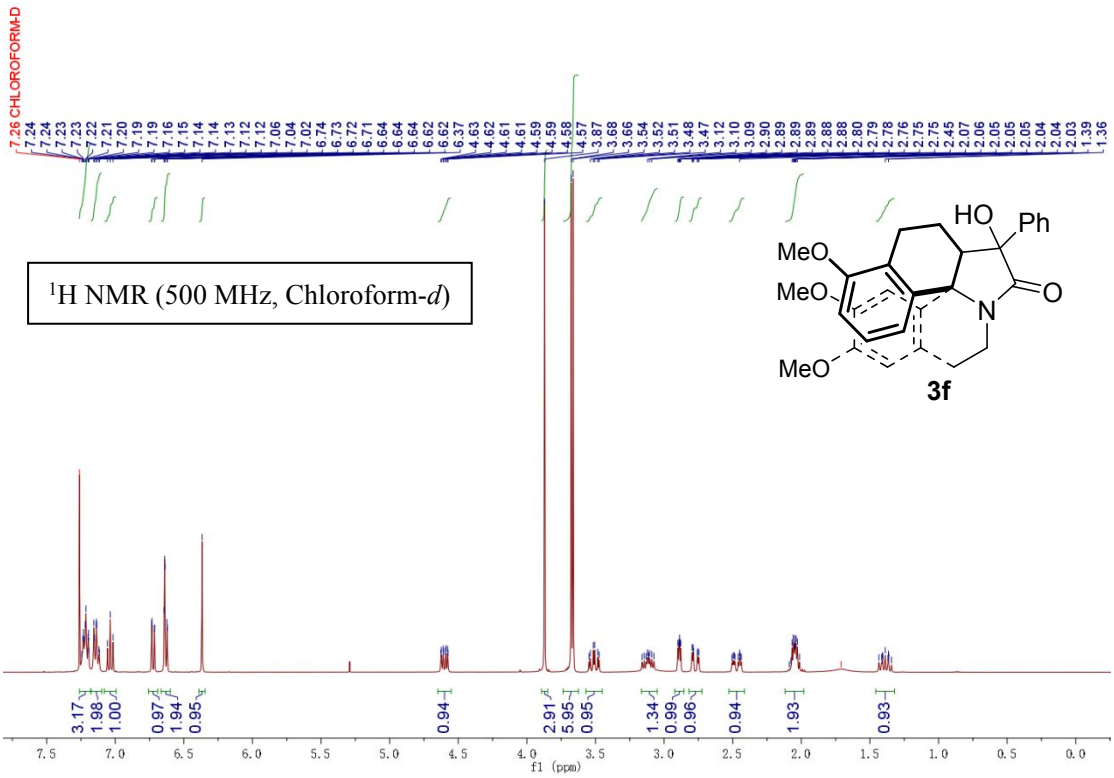

175.7
148.2 148.0 141.4 139.5 136.9 132.5 131.9 130.5 130.1 128.2 128.1 126.9 125.5 119.6
111.6 110.2
81.6 77.3 CDCl3 77.0 CDCl3 76.8 CDCl3
62.7
56.0 55.9 53.4
37.6
28.0 24.5 21.8
13C NMR (126 MHz, Chloroform-d)
HO
Ph
MeO
O
N
MeO
Br
3c
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


^{1}H NMR (500 MHz, DMSO-d_6)
HO
Ph
MeO
MeO
Cl
N
O
3d

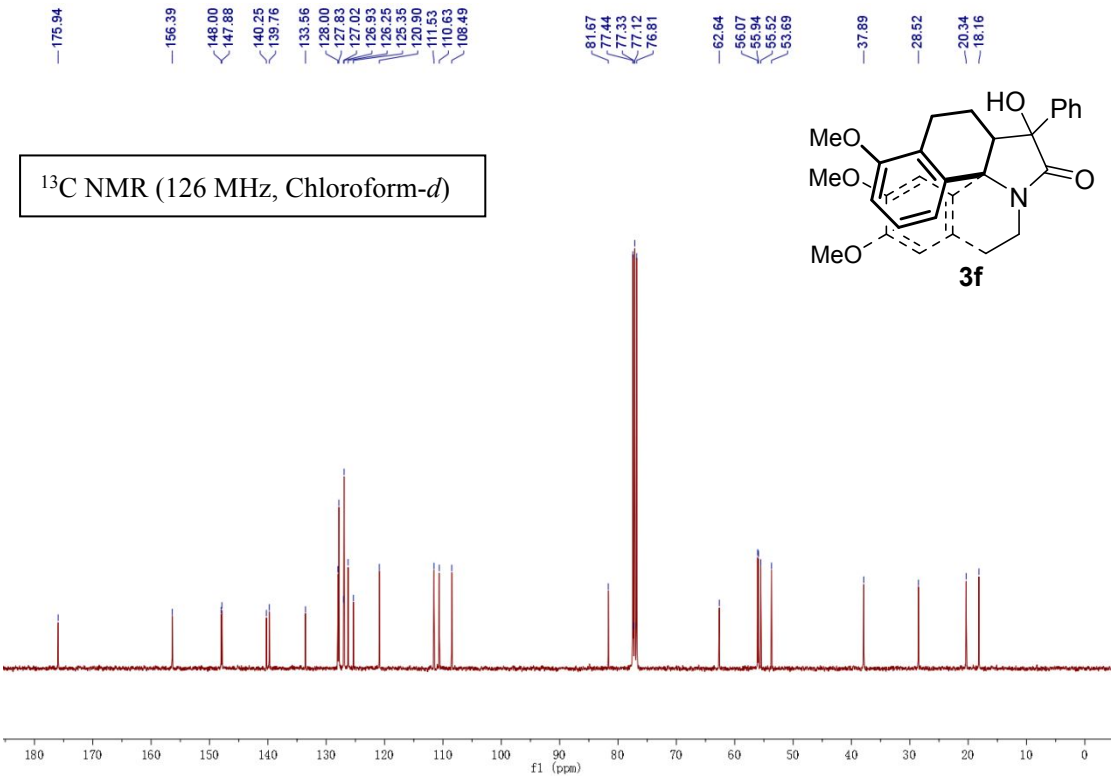

^{13}C NMR (126 MHz, DMSO-d_6)
HO
Ph
MeO
MeO
Cl
N
O
3d


7.33 7.31 7.27 7.27 7.26 CDCl3 7.25 7.24 7.24 7.24 7.23 7.21 7.21 7.20 7.20 6.91 6.91 6.90 6.90 6.77 6.77 6.75 6.73 6.65 6.38 4.52 4.52 4.51 4.51 4.50 4.49 4.48 4.48 3.89 3.70 3.44 3.42 3.41 3.39 3.39 3.17 3.15 3.15 3.14 3.13 3.00 2.98 2.98 2.97 2.97 2.78 2.77 2.75 2.74 2.33 2.33 2.32 2.30 2.30 2.29 2.29 2.20 1.98 1.97 1.96 1.95 1.94 1.94 1.93 1.85 1.81 1.67 HDO
1H NMR (500 MHz, Chloroform-d)
HO
Ph
MeO
O
N
MeO
Me
3e
1.83 3.09 0.97 0.98 0.99 0.99 0.99 1.00 2.98 2.96 1.00 1.03 0.97 0.98 1.00 1.06 2.97 2.03 1.01
7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

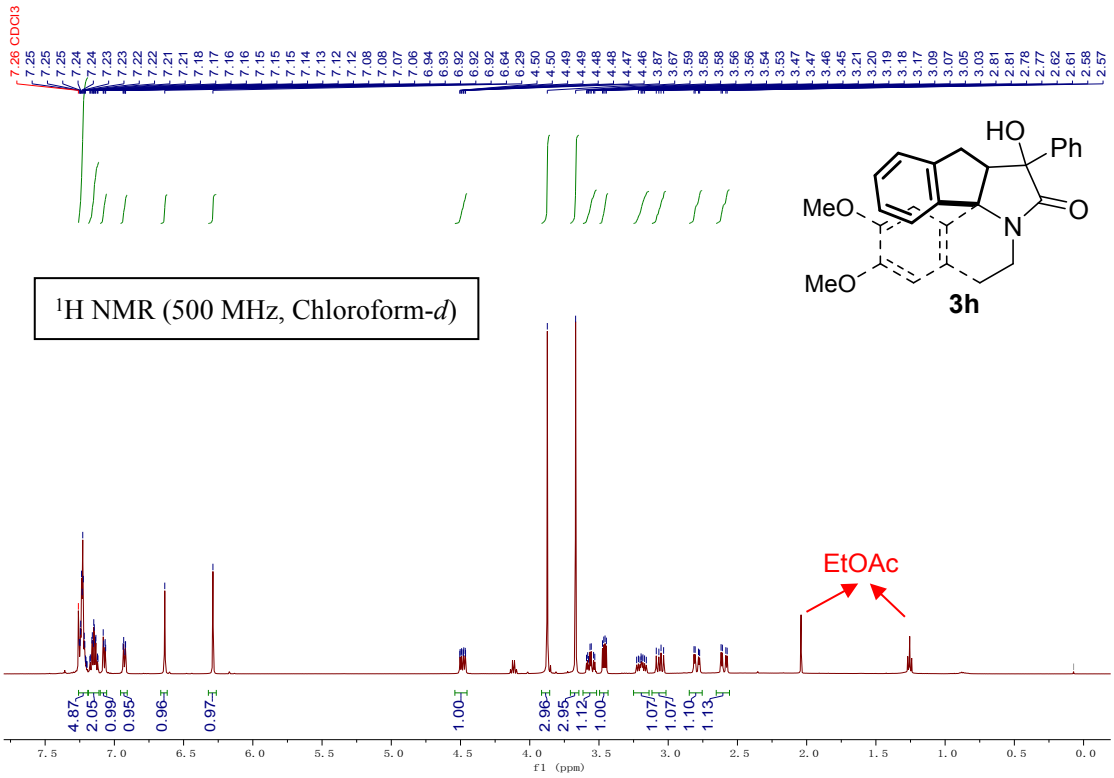

175.6
147.9 147.8 139.9 138.9 135.4 134.9 133.6 129.6 128.3 128.3 128.0 128.0 127.0 125.5 111.3 110.3
81.9 77.3 CDCl3 77.0 CDCl3 76.8 CDCl3
63.0 55.9 55.8 53.6 37.4 28.0 24.6 22.5 21.3
13C NMR (126 MHz, Chloroform-d)
HO
Ph
MeO
O
N
MeO
Me
3e
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)



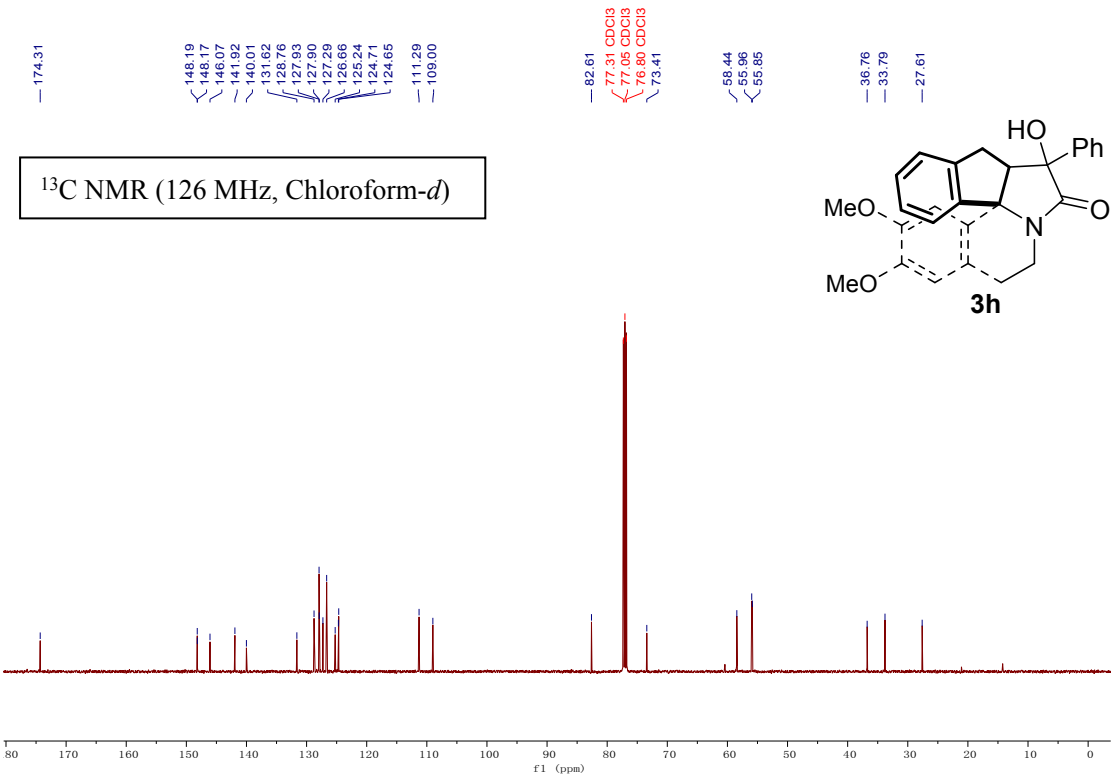
^{1}H NMR (500 MHz, Chloroform-*d*)



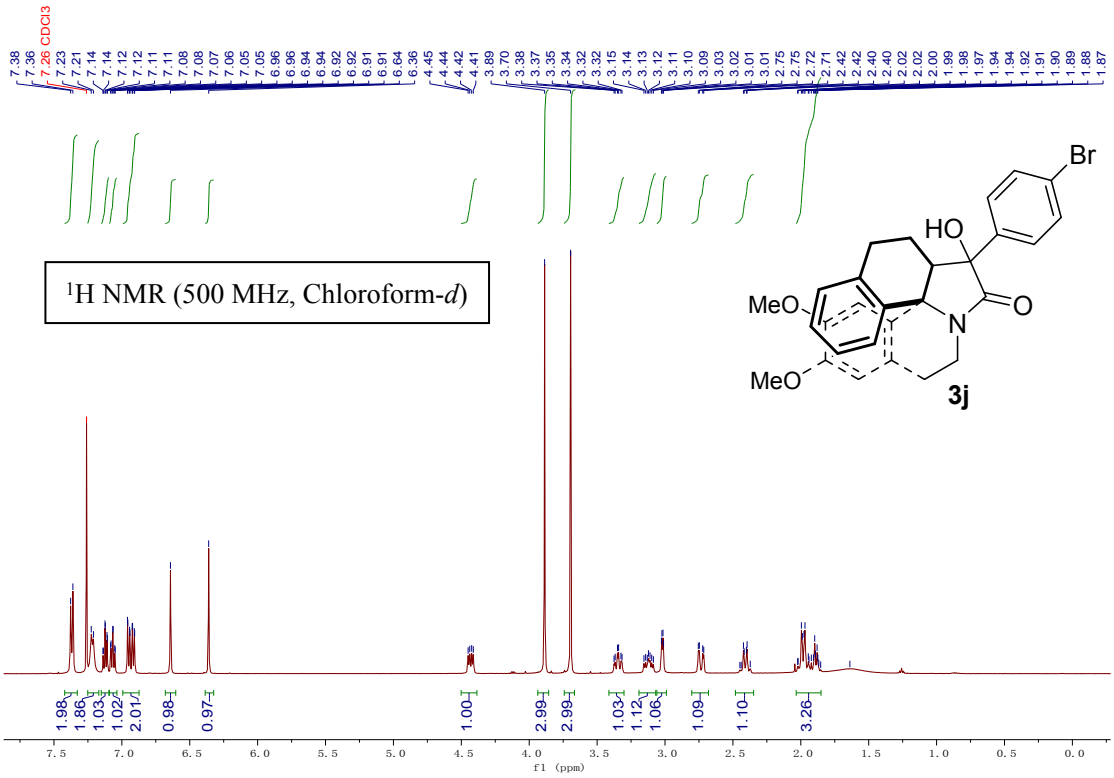
^{13}C NMR (126 MHz, Chloroform-*d*)



^{1}H NMR (500 MHz, Chloroform-*d*)

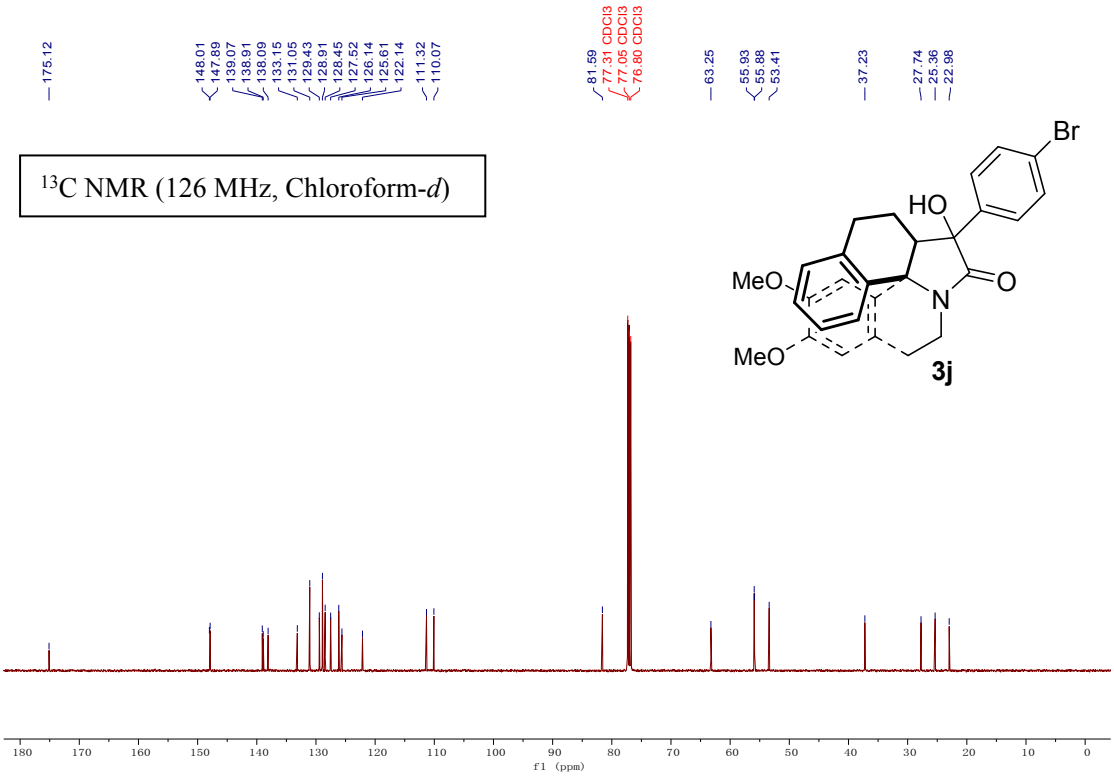


^{13}C NMR (126 MHz, Chloroform-*d*)



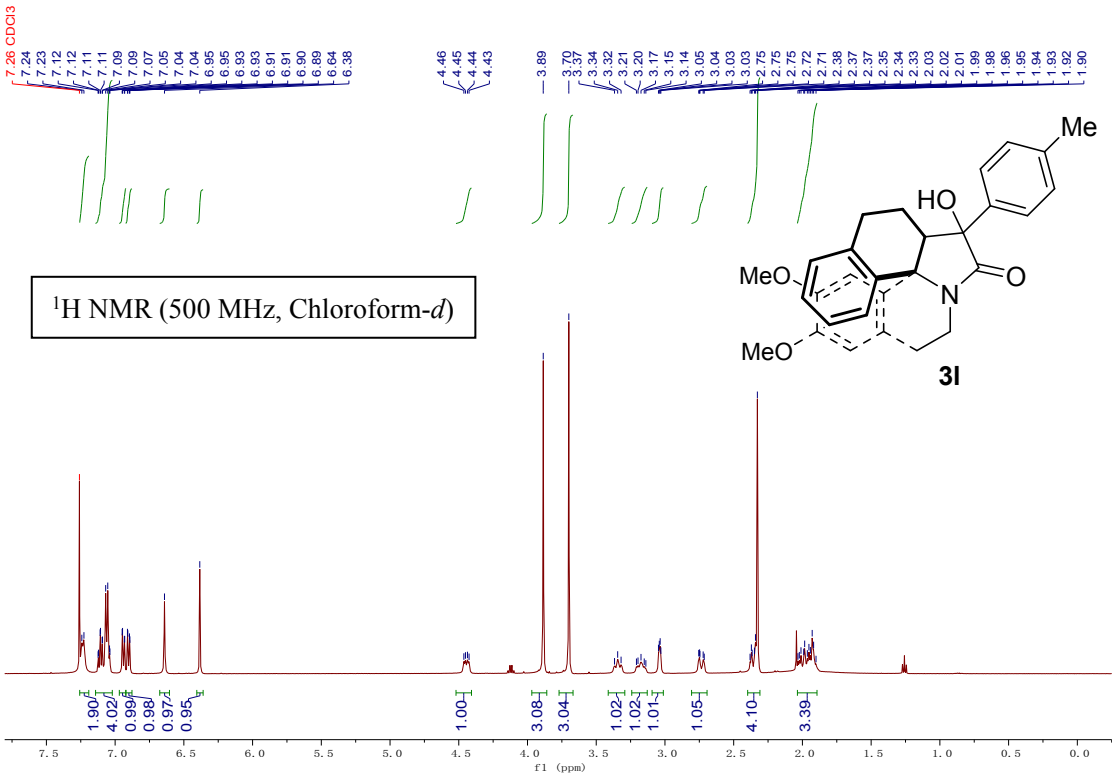
^{1}H NMR (500 MHz, Chloroform-*d*)

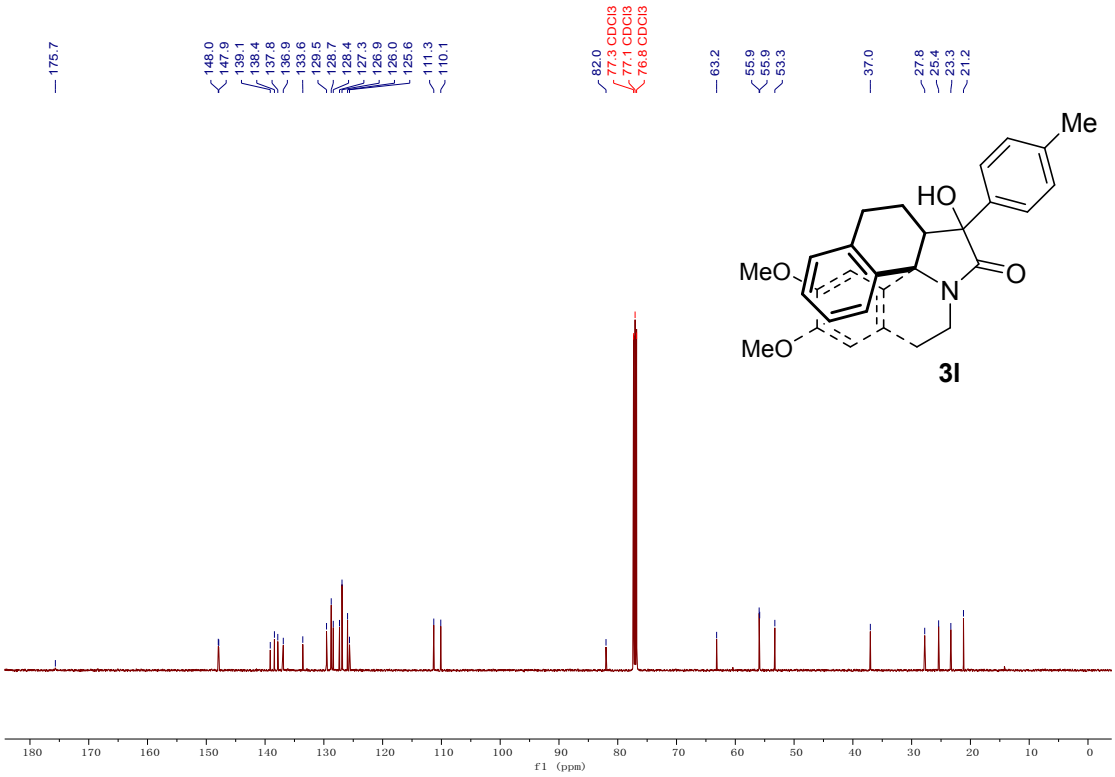
3j

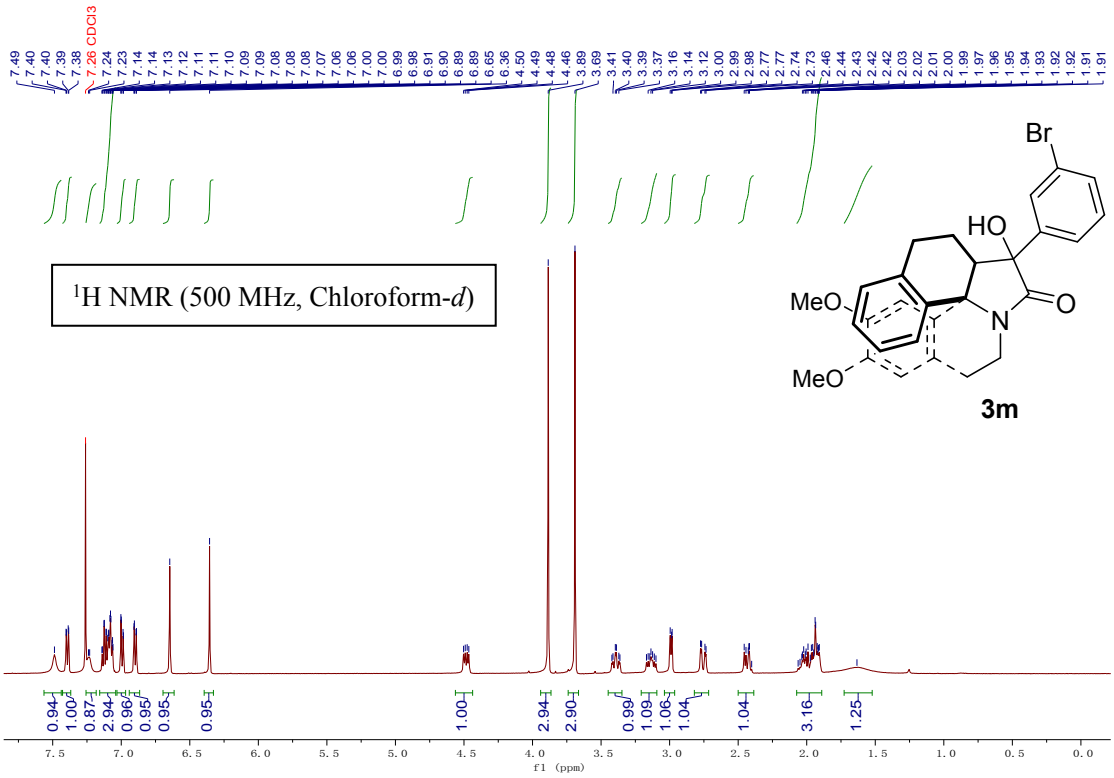


^{13}C NMR (126 MHz, Chloroform-*d*)

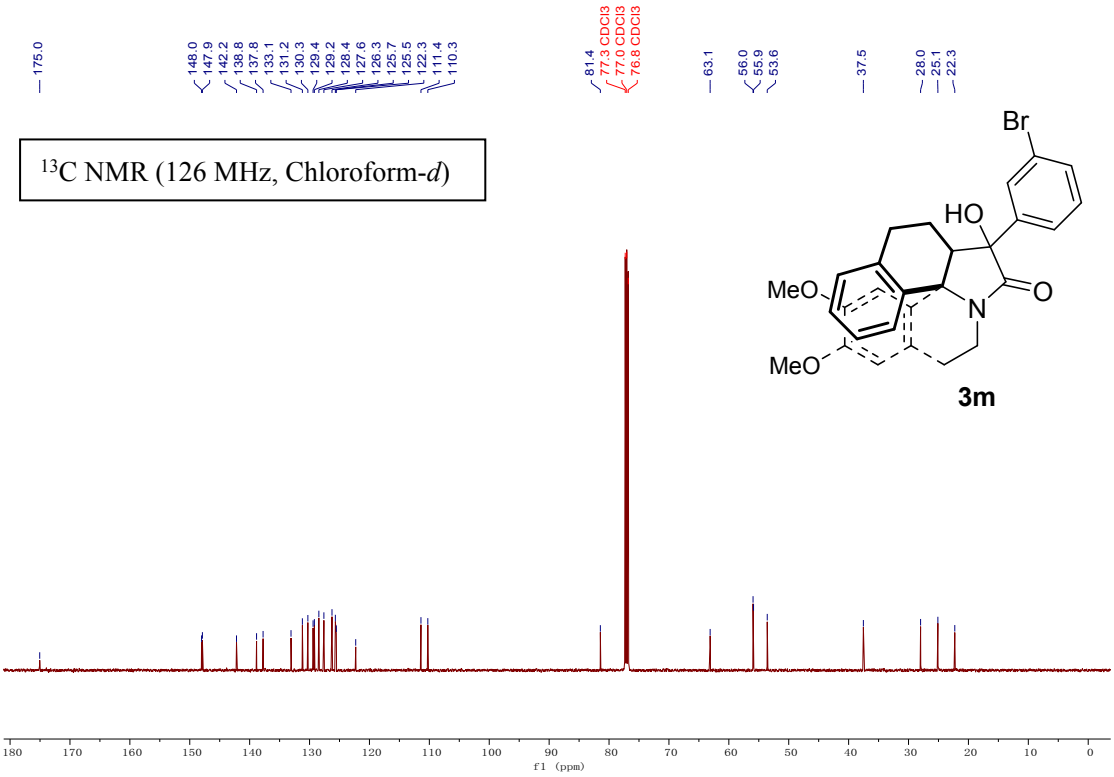
3j


7.26 CDCl3
7.24
7.23
7.12
7.12
7.11
7.11
7.09
7.09
7.07
7.05
7.04
7.04
6.95
6.95
6.93
6.93
6.91
6.91
6.90
6.89
6.64
6.38
4.46
4.45
4.44
4.43
3.89
3.70
3.37
3.34
3.32
3.21
3.20
3.17
3.15
3.14
3.05
3.04
3.03
3.03
2.75
2.75
2.72
2.71
2.38
2.37
2.37
2.35
2.34
2.33
2.03
2.02
2.01
1.99
1.98
1.96
1.95
1.94
1.93
1.92
1.90
Me
HO
MeO
N
O
MeO
3l
1H NMR (500 MHz, Chloroform-d)
1.90
4.02
0.99
0.98
0.97
0.95
1.00
3.08
3.04
1.02
1.02
1.01
1.05
4.10
3.39
f1 (ppm)

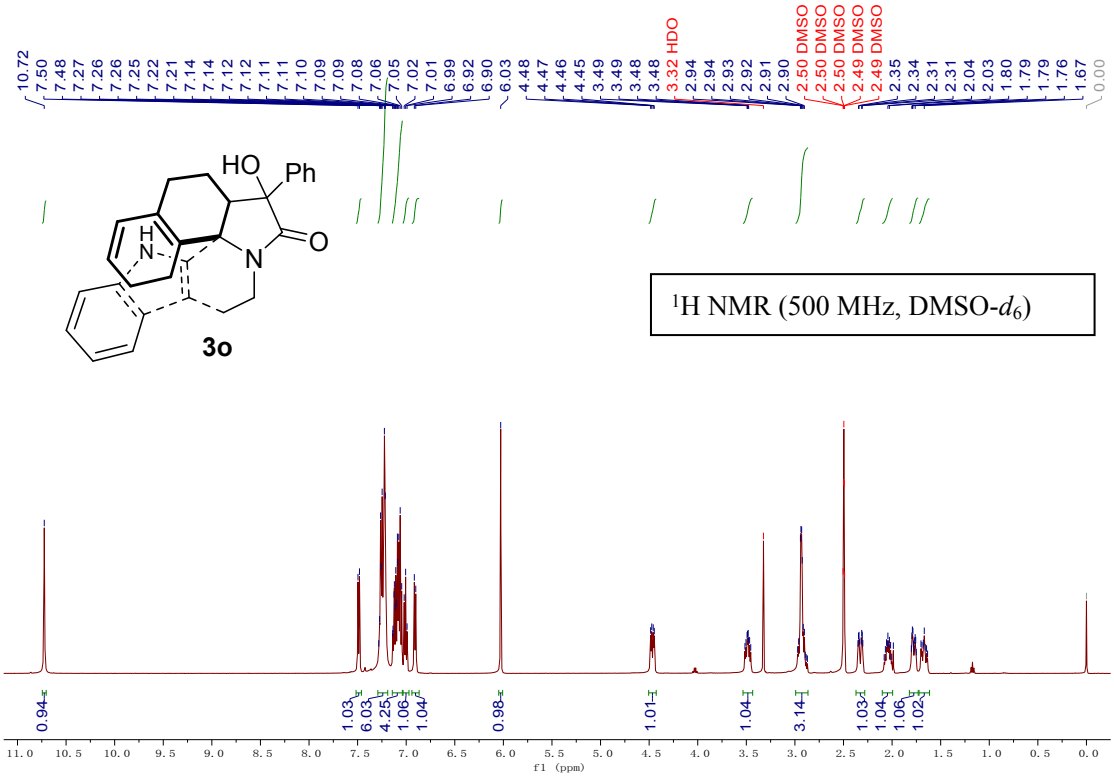

175.7
148.0
147.9
139.1
138.4
137.8
136.9
133.6
129.5
128.7
128.4
127.3
126.9
126.0
125.6
111.3
110.1
82.0
77.3 CDCl3
77.1 CDCl3
76.8 CDCl3
63.2
55.9
55.9
53.3
37.0
27.8
25.4
23.3
21.2
Me
HO
MeO
N
O
MeO
3l
f1 (ppm)

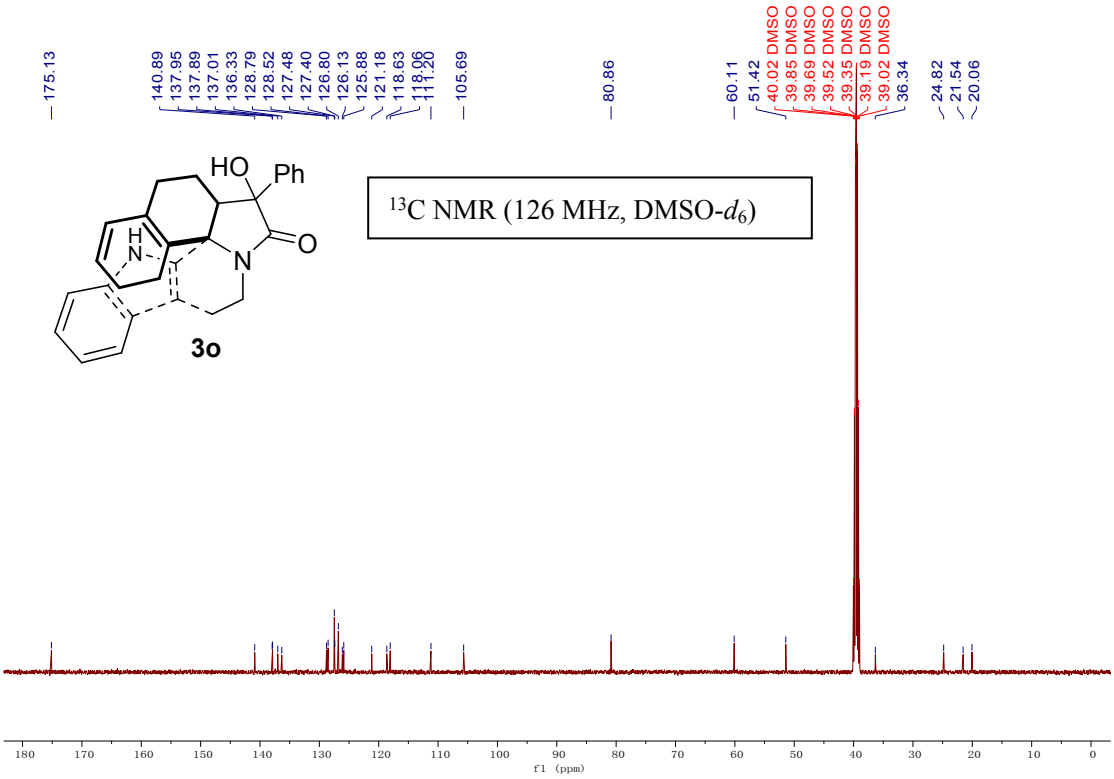


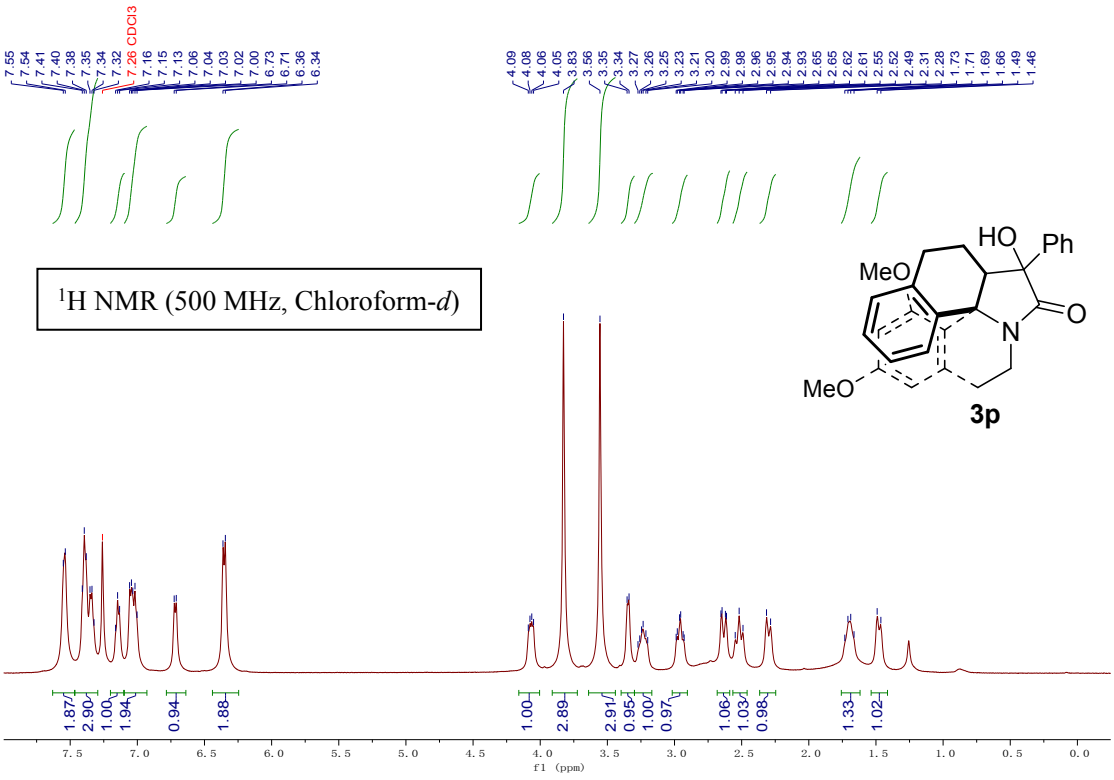
^{1}H NMR (500 MHz, Chloroform-*d*)

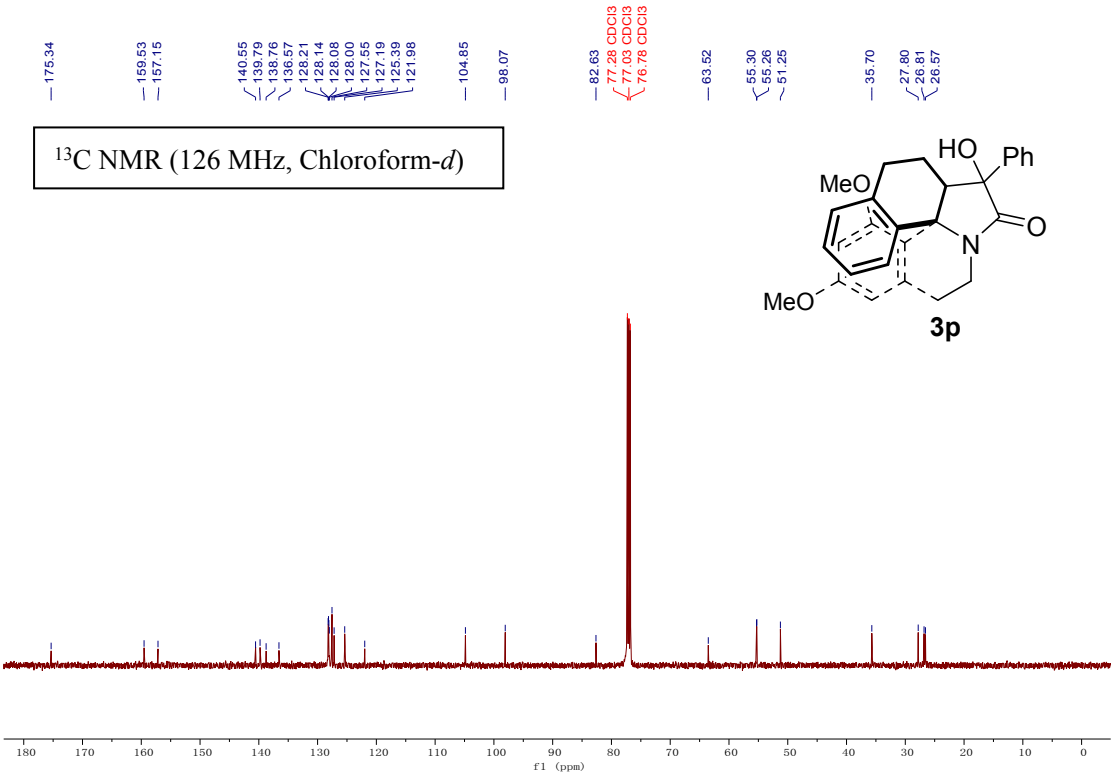


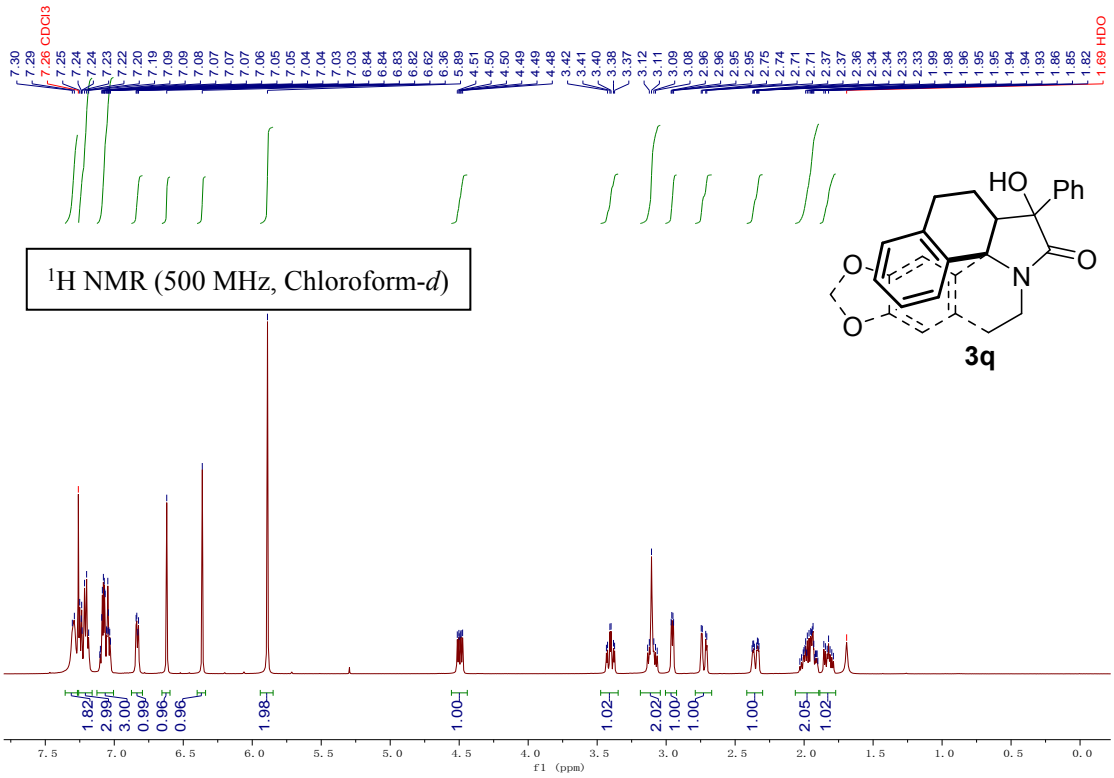
^{13}C NMR (126 MHz, Chloroform-*d*)


HO
Ph
H
N
N
O
3o
[1]H NMR (500 MHz, DMSO-d_6)


HO
Ph
H
N
N
O
3o
[13]C NMR (126 MHz, DMSO-d_6)

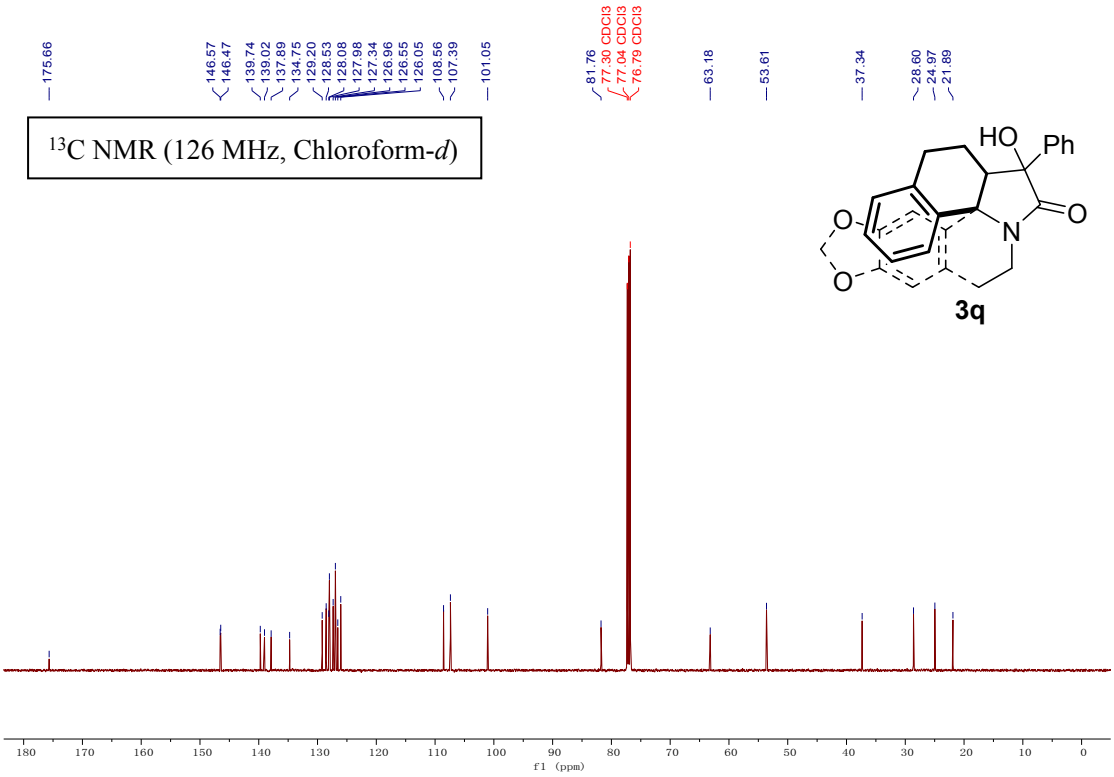

7.55
7.54
7.41
7.40
7.38
7.35
7.34
7.32
7.26 CDCl3
7.16
7.15
7.13
7.06
7.04
7.03
7.02
7.00
6.73
6.71
6.36
6.34
4.09
4.08
4.06
4.05
3.83
3.56
3.35
3.34
3.27
3.26
3.25
3.23
3.21
3.20
2.99
2.98
2.96
2.95
2.94
2.93
2.65
2.62
2.61
2.55
2.52
2.49
2.31
2.28
1.73
1.71
1.69
1.66
1.49
1.46
1H NMR (500 MHz, Chloroform-d)
HO
Ph
MeO
O
N
MeO
3p
1.87
2.90
1.00
1.94
0.94
1.88
1.00
2.89
2.91
0.95
1.00
0.97
1.06
1.03
0.98
1.33
1.02
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


175.34
159.53
157.15
140.55
139.79
138.76
136.57
128.21
128.14
128.08
128.00
127.55
127.19
125.39
121.98
104.85
98.07
82.63
77.28 CDCl3
77.03 CDCl3
76.78 CDCl3
63.52
55.30
55.26
51.25
35.70
27.80
26.81
26.57
13C NMR (126 MHz, Chloroform-d)
HO
Ph
MeO
O
N
MeO
3p
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)



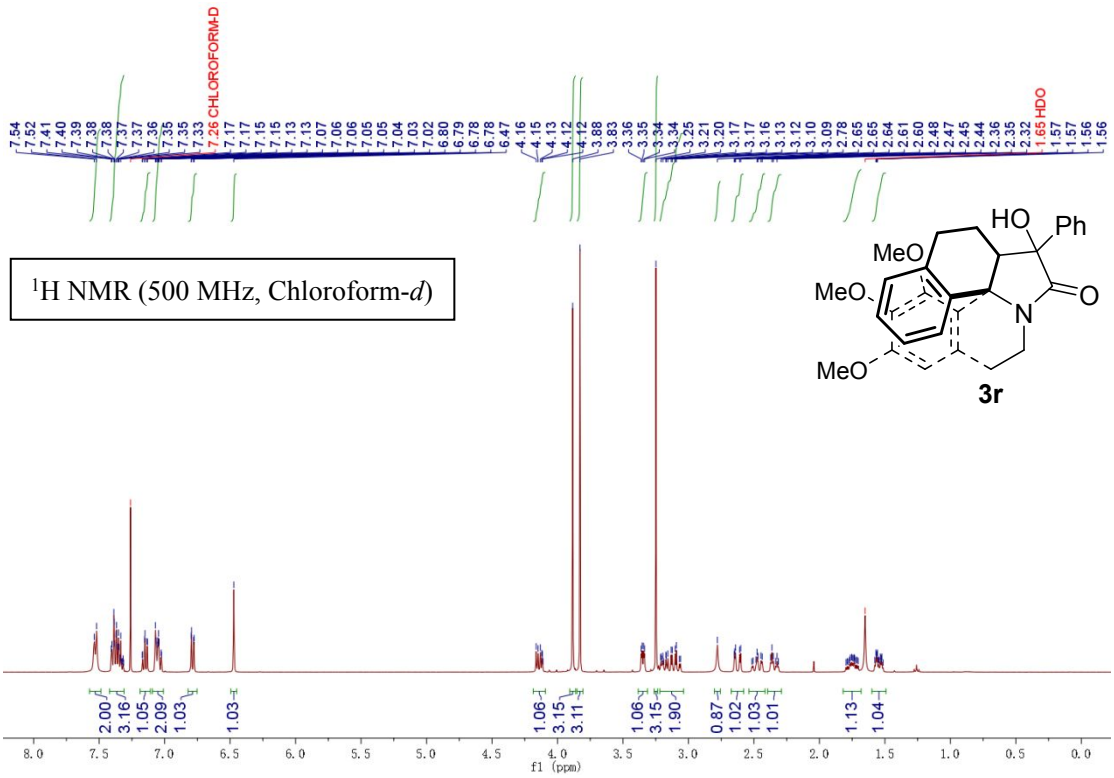
^{1}H NMR (500 MHz, Chloroform-*d*)

3q

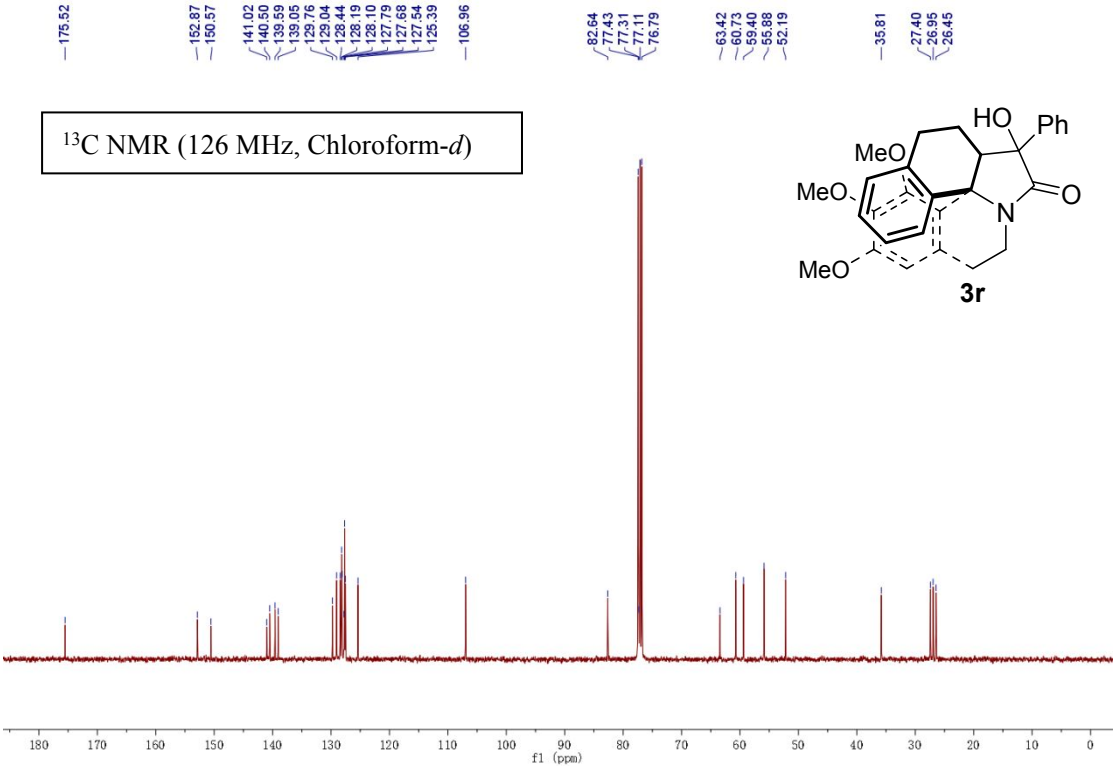


^{13}C NMR (126 MHz, Chloroform-*d*)

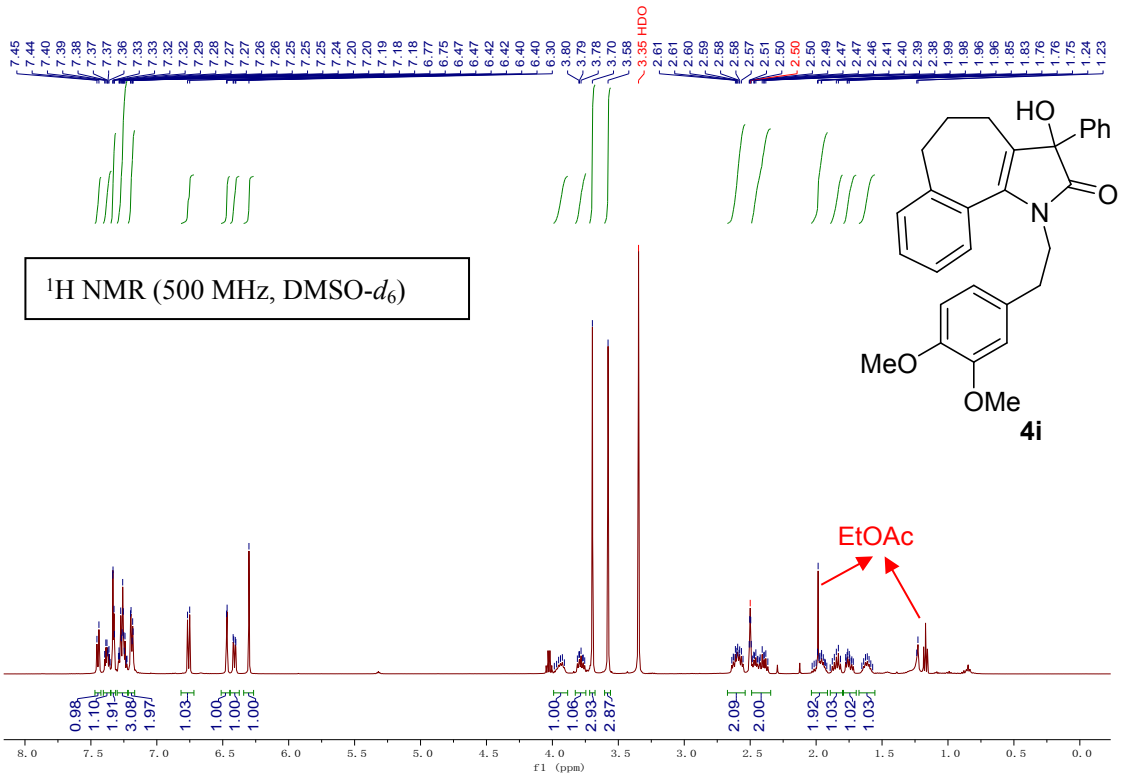
3q



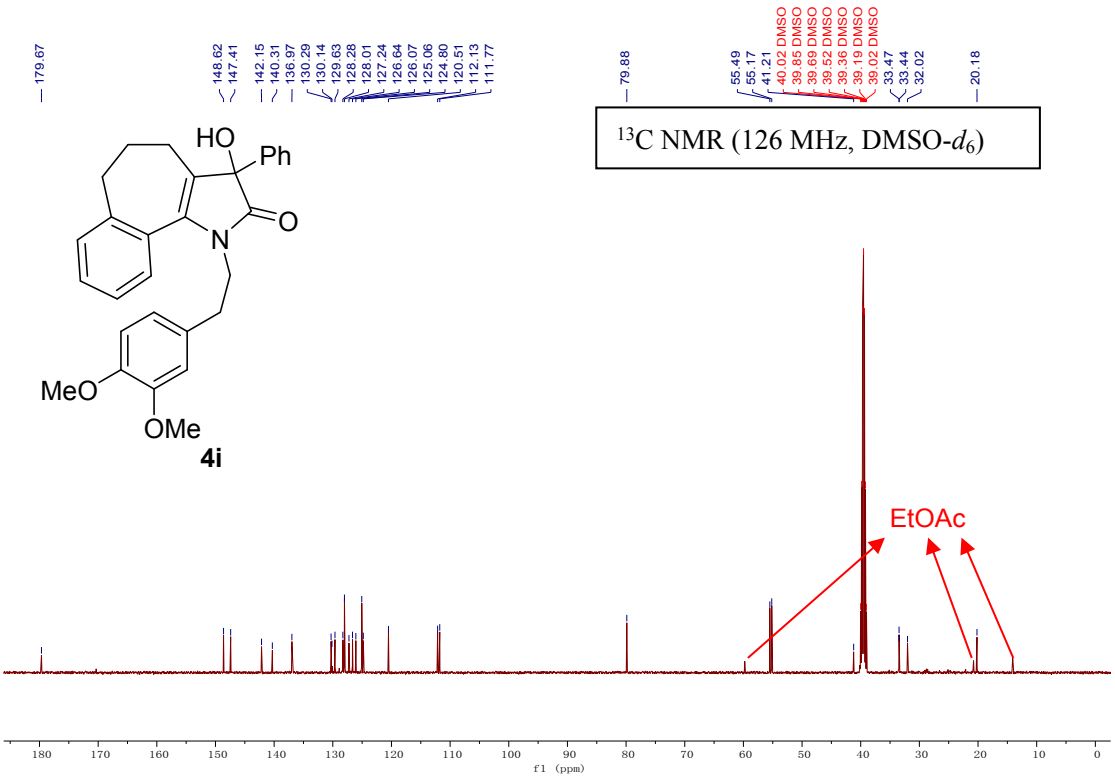
^{1}H NMR (500 MHz, Chloroform-*d*)



^{13}C NMR (126 MHz, Chloroform-*d*)



^{1}H NMR (500 MHz, DMSO-d_6)



^{13}C NMR (126 MHz, DMSO-d_6)