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*Supporting Information*

# Cobalt-Catalyzed Coupling Reactions of 2-Halobenzamides with Alkynes: Investigation of the Ligand-Controlled Dual Pathways

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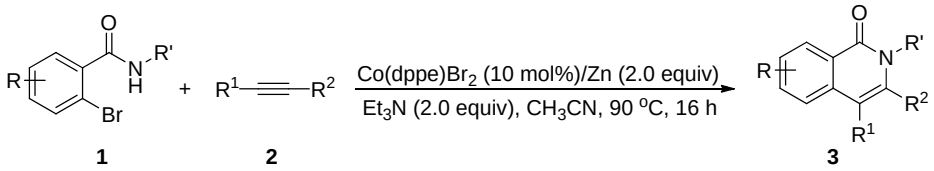
**General information:**

All reagents were purchased from Sigma-Aldrich, Alfa-Aesar, TCI and Fisher-Acros, which were used without further purification unless otherwise noted. All manipulations of oxygen- and moisture-sensitive materials were conducted with a standard Schlenk technique or in the glove box. Flash column chromatography was performed using silica gel (230–400 mesh). Analytical thin layer chromatography (TLC) was performed on 60 $F_{254}$ (0.25 mm) plates and visualization was accomplished with UV light (254 and 354 nm) and/or an aqueous alkaline $KMnO_4$ solution followed by heating. Proton and carbon nuclear magnetic resonance spectra ($^1$H NMR and $^{13}$C NMR) were recorded on Bruker 300, 400 or 600 spectrometer with $Me_4Si$ or solvent resonance as the internal standard ($^1$H NMR, $Me_4Si$ at 0 ppm, $CDCl_3$ at 7.26 ppm, $d_6$-DMSO at 2.49 ppm; $^{13}$C NMR, $Me_4Si$ at 0 ppm, $CDCl_3$ at 77.0 ppm, $d_6$-DMSO at 39.7 ppm). $^1$H NMR data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, quint = quintet, sext = sextet, sept = septet, br = broad, m = multiplet), coupling constants (Hz), and integration. IR spectral data were recorded on a Bruker TENSOR 37 spectrometer. Melting points (mp) were determined using SRS OptiMelt MPA100 or Buchi B-540. GC-MS data were obtained from the HP 5890 Series II GC/HP 5972 GC MASS Spectrometer System. High Resolution Mass spectral data were obtained from MAT-95XL HRMS by using EI method. X-ray data was obtained from Bruker APEX DUO.

**General procedure for the Co-catalyzed cyclization reaction:**

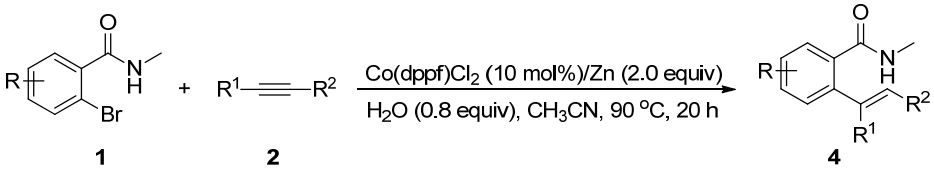



**1** **2** **3**

Addition of all reagents was conducted in a glove box. A screw-capped vial (10-mL) was added Co(dppe)$Br_2$ (dppe = 1,2-bis(diphenylphosphino)ethane) (31 mg, 0.05 mmol), Zn (1.0 mmol), $NEt_3$ (1.0 mmol), substrate **1** (0.5 mmol) and alkyne **2** (0.75 mmol) in dry $CH_3CN$ (1.5 mL). The vial was then removed from the glove box, and allowed to stir at 90 °C for 16 h. The mixture was filtered through a celite pad and washed with $CH_2Cl_2$. The filtrate was concentrated and the residue was purified through a column chromatography by using hexane and ethyl acetate as eluent to afford the desired products **3**.

All structures were characterized by the HRMS, $^1$H NMR and $^{13}$C NMR spectra; products **3a** and **3w** were verified by single crystal X-ray diffraction. Spectral data, melting point, IR data, HRMS data and the copies of $^1$H NMR and $^{13}$C NMR spectra for all compounds are listed below.

**General procedure for the Co-catalyzed reductive coupling reaction:**

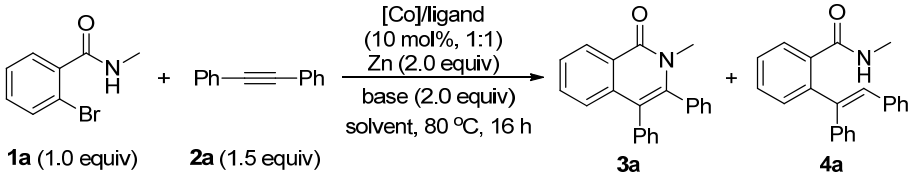



**1** **2** **4**

Addition of all reagents was conducted in a glove box. A screw-capped vial (10-mL) was added Co(dppf)$Cl_2$ (dppf = 1,2-bis(diphenylphosphino)ferrocene) (33 mg, 0.05 mmol), Zn (1.0 mmol), $H_2O$ (0.4 mmol), substrate **1** (0.5 mmol) and alkyne **2** (0.6 mmol) in dry $CH_3CN$ (2.0 mL). The vial was then removed from the glove box, and allowed to stir at 90 °C for 20 h. The mixture was filtered through a celite pad and washed with $CH_2Cl_2$. The filtrate was concentrated and the residue was purified through a column chromatography by using hexane and ethyl acetate as eluent to afford the desired products **4**.

All structures were characterized by the HRMS, $^1$H NMR and $^{13}$C NMR spectra; products **4a** and **4v'** were verified by single crystal X-ray diffraction. Spectral data, melting point, IR data, HRMS data and the copies of $^1$H NMR and $^{13}$C NMR spectra for all compounds are listed below.

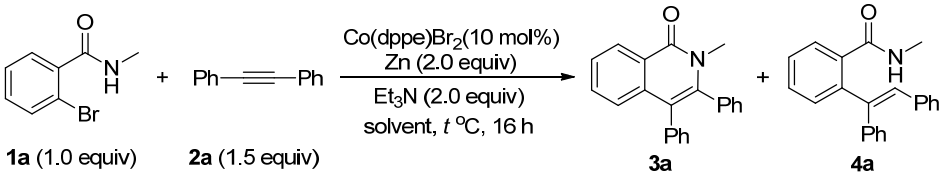
**Table S1. Optimization study of the Co-catalyzed cyclization**[a, b]




| entry | [Co]/ligand | base | solvent | yield (%) 3a | yield (%) 4a | entry | [Co]/ligand | base | solvent | yield (%) 3a | yield (%) 4a |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | $CoI_2/PPh_3$ | $Et_3N$ | $CH_3CN$ | 9 | 23 | 13 | $Co(dppe)Br_2$ | pyrolidine | $CH_3CN$ | 10 | 16 |
| 2 | $CoI_2/PCy_3$ | $Et_3N$ | $CH_3CN$ | 18 | 20 | 14 | $Co(dppe)Br_2$ | morpholine | $CH_3CN$ | trace | 13 |
| 3 | $CoI_2$/dppe | $Et_3N$ | $CH_3CN$ | 58 | trace | 15 | $Co(dppe)Br_2$ | DIPEA | $CH_3CN$ | trace | 36 |
| 4 | $CoI_2$/dppp | $Et_3N$ | $CH_3CN$ | 46 | 8 | 16 | $Co(dppe)Br_2$ | DBU | $CH_3CN$ | 21 | 17 |
| 5 | $CoI_2$/dppb | $Et_3N$ | $CH_3CN$ | 41 | 27 | 17 | $Co(dppe)Br_2$ | $NaHCO_3$ | $CH_3CN$ | trace | 13 |
| 6 | $CoI_2$/dppm | $Et_3N$ | $CH_3CN$ | 14 | 17 | 18 | $Co(dppe)Br_2$ | $K_2CO_3$ | $CH_3CN$ | trace | 22 |
| 7 | $CoI_2$/dppf | $Et_3N$ | $CH_3CN$ | 0 | 31 | 19 | $Co(dppe)Br_2$ | $Et_3N$ | DMSO | 19 | 24 |
| 8 | $Co(acac)_2$/dppe | $Et_3N$ | $CH_3CN$ | 41 | 8 | 20 | $Co(dppe)Br_2$ | $Et_3N$ | DMF | 22 | 15 |
| 9 | $Co(dppe)I_2$ | $Et_3N$ | $CH_3CN$ | 68 | 17 | 21 | $Co(dppe)Br_2$ | $Et_3N$ | THF | 46 | 10 |
| 10 | $Co(dppe)Br_2$ | $Et_3N$ | $CH_3CN$ | 63 | 10 | 22 | $Co(dppe)Br_2$ | $Et_3N$ | 1,4-dioxane | 53 | 11 |
| 11 | $Co(dppf)Br_2$ | $Et_3N$ | $CH_3CN$ | 0 | 45 | 23 | $Co(dppe)Br_2$ | $Et_3N$ | DCM | trace | 20 |
| 12 | $Co(dppe)Br_2$ | pyridine | $CH_3CN$ | 18 | 13 | 24 | $Co(dppe)Br_2$ | $Et_3N$ | toluene | 18 | 0 |

[a]Reaction conditions: **1a (**0.2 mmol, 1.0 equiv), **2a (**0.3 mmol, 1.5 equiv), cobalt source (0.02 mmol, 10 mol%), bidentate ligand (0.02 mol, 10 mol%; for $PPh_3$, 20 mol% was used), base (0.4 mmol, 2 equiv) in 0.6 mL solvent at 80 °C for 16 h. [b]Yields were measured from the crude products by $^1$H NMR integration method using mesitylene as an internal standard.

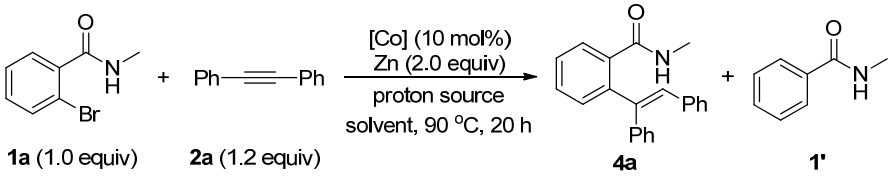
**Table S2. Optimization study of the Co-catalyzed cyclization**[a, b]




| entry | solvent | *t* (°C) | yield (%) 3a | yield (%) 4a | entry | solvent | *t* (°C) | yield (%) 3a | yield (%) 4a |
|---|---|---|---|---|---|---|---|---|---|
| 1 | $CH_3CN$ | 80 | 63 | 10 | 7 | 1,4-dioxane | 90 | 67 | 21 |
| 2 | $CH_3CN$ | 90 | 73 | 6 | 8 | toluene | 80 | 18 | 0 |
| 3 | $CH_3CN$ | 100 | 58 | 13 | 9 | toluene | 90 | 26 | 0 |
| 4 | THF | 80 | 46 | 10 | 10 | toluene | 100 | 41 | 0 |
| 5 | THF | 90 | 57 | 12 | 11 | toluene | 110 | 38 | 0 |
| 6 | 1,4-dioxane | 80 | 53 | 11 | 12 | toluene | 120 | 19 | 0 |

[a]Reaction conditions: **1a (**0.2 mmol, 1.0 equiv), **2a (**0.3 mmol, 1.5 equiv), $Co(dppe)Br_2$ (0.02 mmol, 10 mol%), $Et_3N$ (0.4 mmol, 2 equiv) in 0.6 mL solvent at *t* °C for 16 h. [b]Yields were measured from the crude products by $^1$H NMR integration method using mesitylene as an internal standard.

**Table S3. Optimization study of the Co-catalyzed reductive coupling reaction**[a, b]

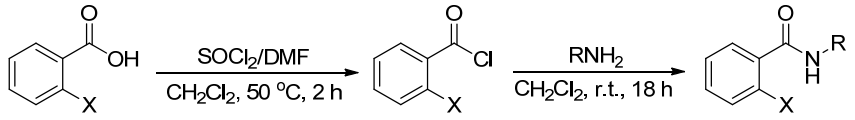



**1a** (1.0 equiv) **2a** (1.2 equiv) **4a** **1'**

| entry | [Co] | proton source (x equiv) | solvent | yield (%) | |
|---|---|---|---|---|---|
| | | | | **4a** | **1'** |
| 1 | $CoI_2$/dppf | $H_2O$ (1.0 equiv) | $CH_3CN$ | 61 | 14 |
| 2 | Co(dppf)$I_2$ | $H_2O$ (1.0 equiv) | $CH_3CN$ | 67 | 20 |
| 3 | Co(dppf)$Br_2$ | $H_2O$ (1.0 equiv) | $CH_3CN$ | 71 | 18 |
| 4 | Co(dppf)$Cl_2$ | $H_2O$ (1.0 equiv) | $CH_3CN$ | 74 | 12 |
| 5 | Co(dppf)$Cl_2$ | $H_2O$ (1.2 equiv) | $CH_3CN$ | 54 | 33 |
| 6 | Co(dppf)$Cl_2$ | $H_2O$ (0.8 equiv) | $CH_3CN$ | 79 | 11 |
| 7 | Co(dppf)$Cl_2$ | $H_2O$ (0.6 equiv) | $CH_3CN$ | 71 | 6 |
| 8 | Co(dppf)$Cl_2$ | $H_2O$ (0.4 equiv) | $CH_3CN$ | 57 | 7 |
| 8 | Co(dppf)$Cl_2$ | MeOH (1.0 equiv) | $CH_3CN$ | 43 | 45 |
| 9 | Co(dppf)$Cl_2$ | EtOH (1.0 equiv) | $CH_3CN$ | 73 | 10 |
| 10 | Co(dppf)$Cl_2$ | IPA (1.0 equiv) | $CH_3CN$ | 79 | 11 |
| 11 | Co(dppf)$Cl_2$ | TFA (1.0 equiv) | $CH_3CN$ | 38 | 45 |
| 12 | Co(dppf)$Cl_2$ | 2,6-DTBP (1.0 equiv) | $CH_3CN$ | 74 | 5 |
| 13 | Co(dppf)$Cl_2$ | HOAc (1.0 equiv) | $CH_3CN$ | 27 | 61 |
| 14 | Co(dppf)$Cl_2$ | $H_2O$ (1.0 equiv) | THF | 21 | 64 |
| 15 | Co(dppf)$Cl_2$ | $H_2O$ (1.0 equiv) | DMF | 16 | 57 |

[a]Reaction conditions: **1a (**0.2 mmol, 1.0 equiv), **2a (**0.24 mmol, 1.2 equiv), Co(dppf)$X_2$ (0.02 mmol, 10 mol%), proton source (x equiv) in 0.8 mL dry solvent at 90 °C for 20 h. [b]Yields were measured from the crude products by $^1$H NMR integration method using mesitylene as an internal standard. IPA = isopropanol; 2,6-DTBP = 2,6-di-*tert*-butylphenol.

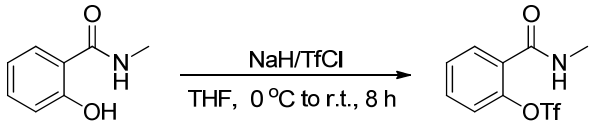
**Procedure for the synthesis of 2-halo-*N*-substitutedbenzamide (1):**[1]




X = Br, Cl, I and OH **1**

2-Halo-benzoic acid (20 mmol, 1.0 equiv) stirring in $SOCl_2$ (12.5 mL) was added DMF (0.1 mL) and kept stirring at 50 °C for 2 h. The resulted mixture was concentrated in *vacuo* and then dissolved in $CH_2Cl_2$ (100 mL). Amine (40 mmol, 2.0 equiv) was then added to the residue/$CH_2Cl_2$ solution at 0 °C and kept stirring at room temperature for 18 h. Upon completion of the reaction as observed by TLC, the mixture was diluted with 10% HCl solution at 0 °C and then extracted with EtOAc (4 x 150 mL). The organic layer was collected, dried over anhydrous $Na_2SO_4$ and concentrated *in vacuo*. The residue was purified by flash column chromatography to give compound **1**. The spectral data and the copies of NMR spectra, please see reference 1 for the detail.

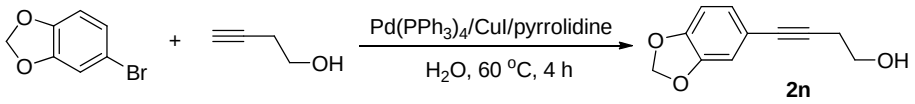
**Procedure for the synthesis of 2-(methylcarbamoyl)phenyltrifluoromethanesulfonate (1a-f):**




**1a-f**

2-Hydroxy-N-methylbenzamide (554 mg, 4 mmol, 1.0 equiv) was kept stirring with NaH (336 mg, 14 mmol, 3.5 equiv) in THF (20 mL) at 0 °C for 2 h. The suspension solution was then moved from ice bath and allowed to increase the temperature slowly from 0 °C to room temperature. TfCl (0.47 mL) was added into the solution mixture and kept stirring at room temperature for 6 h. Upon completion of the reaction as observed by TLC, the mixture was extracted by brine and EtOAc (3 x 50 mL), and the organic layer was collected, dried over anhydrous $MgSO_4$ and concentrated *in vacuo*. The residue was purified through a flash column chromatography by using hexane and ethyl acetate as eluent to provide the desired compound as white powder (962 mg, 85%); mp: 86 $^oC$; IR (KBr): 3287, 1638, 1207, 901, 729, 626, 519, 467 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$) δ 7.80 (dd, $J$ = 7.5, 1.8 Hz, 1H), 7.54 (td, $J$ = 7.5, 1.8 Hz, 1H), 7.46 (td, $J$ = 7.5, 0.9 Hz, 1H), 7.33 (d, $J$ = 8.1 Hz, 1H), 3.02 (d, $J$ = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$) δ 164.6, 146.0, 132.2, 131.0, 129.6, 128.7, 122.2, 118.6 (q, $J_{C-F}$ = 318 Hz), 26.8; HRMS [(ESI), $(M+H)^+$]: 284.0204 (cal. for $C_9H_9NO_4F_3S$ 284.0204); New compound.

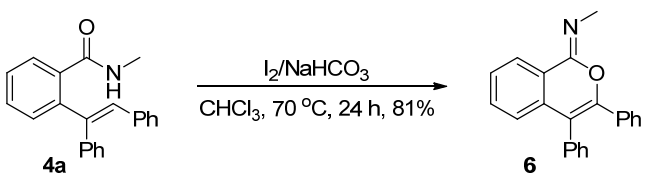
**Procedure for the synthesis of 4-(benzo[*d*][1,3]dioxol-5-yl)but-3-yn-1-ol (2n):**




**2n**

To a solution of 1-bromo-3,4-(methylenedioxy)benzene (2.05 mL, 17 mmol, 1.0 equiv) in ultra pure water (35 mL) was added but-3-yn-1-ol (1.54 mL, 20.4 mmol, 1.2 equiv), pyrrolidine (1.40 mL, 17 mmol, 1.0 equiv), $Pd(PPh_3)_4$ (982 mg, 0.85 mmol, 5 mol%) and CuI (324 mg, 1.7 mmol, 10 mol%) under nitrogen atmosphere. The reaction mixture was kept stirring at 60 $^oC$ for 4 h and cooled to the room temperature. The aqueous layer was extracted with EtOAc (100 mL×2), and the organic layer was washed with $H_2O$ and brine, dried over $MgSO_4$ and concentrated in *vacuo*. The residue was purified by flash column chromatography [$R_f$ = 0.4 (25 % ethyl acetate in hexanes)] to give compound **2n** as brownish oil (2.59 g, 80%); IR (KBr): 3005, 1643, 1275, 1260, 1038 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$) δ 6.92 (dd, $J$ = 8.4, 1.2 Hz, 1H), 6.85 (d, $J$ = 1.2 Hz, 1H), 6.71 (d, $J$ = 7.8 Hz, 1H), 5.94 (s, 2H), 3.78 (t, $J$ = 6.0 Hz, 2H), 2.64 (t, $J$ = 6.0 Hz, 2H); $^{13}C$ NMR (150 MHz, $CDCl_3$) δ 147.5, 147.3, 126.0, 116.5, 111.6, 108.3, 101.2, 84.6, 82.1, 61.1, 23.7; HRMS [(EI), $(M^+)$]: 190.0627 (cal. for $C_{11}H_{10}O_3$ 190.0630); Registry Number: [912649-12-8].

**Procedure for the synthesis of (Z)-*N*-(3,4-diphenyl-1*H*-isochromen-1-ylidene)methanamine (6):**

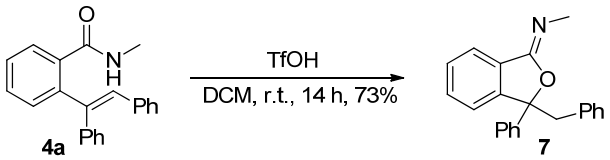



**4a** **6**

Compound **4a** (125 mg, 0.4 mmol, 1.0 equiv), $I_2$ (340 mg, 1.2 mmol, 3.0 equiv) and $NaHCO_3$ (100 mg, 1.2 mmol, 3.0 equiv) in $CHCl_3$ (5 mL) was kept stirring at 70 °C. Upon completion of the reaction as observed by TLC, the mixture was diluted with $CH_2Cl_2$, filtered through a celite pad, washed with $CH_2Cl_2$, and then concentrated *in vacuo*. The residue was purified by flash column chromatography to

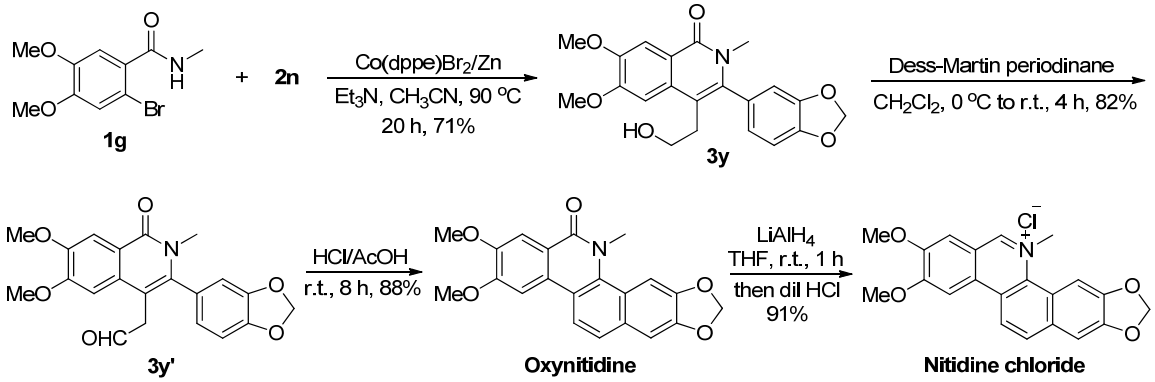
afford compound **6** as yellow solid (100 mg, 81%); mp: 130 °C; IR(KBr): 3450, 1664, 1383, 756, 698 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.28 (d, *J* = 7.2 Hz, 1H), 7.40–7.35 (m, 5H), 7.31–7.30 (m, 2H), 7.25–7.18 (m, 4H), 6.99 (d, *J* = 7.8 Hz, 1H), 6.85–6.83 (m, 1H), 3.29 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 151.9, 148.9, 135.1, 134.6, 133.9, 131.5, 131.4, 128.9, 128.7, 128.5, 127.9, 127.8, 127.7, 126.6, 126.2, 123.7, 115.4, 33.6; HRMS [(ESI), $(M+H)^+$]: 312.1388 (cal. for $C_{22}H_{18}NO$ 312.1391); New compound.

**Procedure for the synthesis of (*Z*)-*N*-(3-benzyl-3-phenylisobenzofuran-1(3*H*)-ylidene)methanamine (7):**




Compound **4a** (250 mg, 0.8 mmol) in $CH_2Cl_2$ (10 mL) was slowly added TfOH (48 mg, 0.32 mmol, 40 mol%), and kept stirring at room temperature. Upon completion of the reaction as observed by TLC, the mixture was diluted with $CH_2Cl_2$, filtered through a celite pad and washed with $CH_2Cl_2$. The filtrate was then extracted by $CH_2Cl_2$ and brine solution. The combined organic layer was collected, dried over anhydrous $Na_2SO_4$ and concentrated *in vacuo*. The residue was purified by flash column chromatography using ethyl acetate and hexane as eluent to afford compound **7** as yellow solid (183 mg, 73%); mp: 148 °C; IR(KBr): 3415, 1713, 1244, 638, 516 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.22 (d, *J* = 7.8 Hz, 1H), 7.86 (t, *J* = 7.2 Hz, 1H), 7.63 (d, *J* = 7.8 Hz, 1H), 7.57 (t, *J* = 7.2 Hz, 1H), 7.48–7.46 (m, 5H), 7.16 (t, *J* = 7.2 Hz, 1H), 7.08 (t, *J* = 7.2 Hz, 2H), 6.76 (d, *J* = 7.2 Hz, 2H), 3.90 (d, *J* = 14.4 Hz, 1H), 3.77 (d, *J* = 14.4 Hz, 1H), 3.33 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 171.2, 149.5, 136.5, 136.1, 131.7, 130.9, 130.0, 129.3, 128.3, 128.0, 126.1, 125.5, 123.4, 123.3, 102.4, 45.5, 30.2; HRMS [(ESI), $(M+H)^+$]: 314.1543 (cal. for $C_{22}H_{20}NO$ 314.1545); New compound.

**Synthetic pathway to approach oxynitidine and nitidine chloride:**




**Procedures and spectral data:**

**Procedure for the synthesis compound 3y:**

In an nitrogen-filled glove box, a 4-mL vial equipped with a magnetic stirrer bar was charged sequentially with **1g** (136 mg, 0.5 mmol, 1.0 equiv), **2n** (143 mg, 0.75 mmol, 1.5 equiv), Co(dppe)$Br_2$ (62 mg, 0.1 mmol), Zn (65 mg, 1.0 mmol), and $Et_3N$ (0.14 mL, 1.0 mmol, 2.0 equiv), followed by the addition of $CH_3CN$ (1.5 mL). The vial was closed and removed from the glove box, and the mixture was kept stirring at 90 °C for 20 h. Upon cooling to room temperature, the reaction mixture was diluted with $CH_2Cl_2$ (10 mL) and filtered through a celite pad with additional $CH_2Cl_2$ (10 mL) as an eluent. The organic solution was concentrated under reduced pressure, and the residue was purified through flash column chromatography [$R_f$ = 0.2 (75 % ethyl acetate in hexanes)] to give the desired compound **3y** as white solid (136 mg, 71%); mp: 237 °C; IR (KBr): 2875, 1636, 1240, 1034 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$) δ 7.90 (s, 1H), 7.11 (s, 1H), 6.93 (d, *J* = 8.4 Hz, 1H), 6.74–6.73 (m, 2H),

6.07 (s, 2H), 4.03 (s, 3H), 4.01 (s, 3H), 3.72 (t, *J* = 7.2 Hz, 2H), 3.26 (s, 3H), 2.82–2.73 (m, 2H); $^{13}$C NMR (150 MHz, $CDCl_3$) δ 161.7, 153.4, 149.1, 148.2, 148.0, 140.2, 131.7, 128.9, 123.1, 119.6, 110.9, 109.7, 108.8, 108.2, 103.7, 101.5, 62.5, 56.2, 56.1, 34.0, 31.9; HRMS [(EI), ($M^+$)]: 383.1363 (cal. for $C_{21}H_{21}NO_6$ 383.1369); Registry Number: [1207666-57-6].

**Procedure for the synthesis compound 3y':**

Compound **3y** (115 mg, 0.3 mmol, 1.0 equiv) was dissolved in dry $CH_2Cl_2$ (10 mL) and stirred at 0 °C in an ice bath. The solution was added Dess-Martin periodinane (191 mg, 0.45 mmol, 1.5 equiv) in one portion and the reaction was kept stirring at room temperature for 4 h. The reaction was quenched at 0°C by stirring with a solution of $Na_2S_2O_3$ (0.2 g in 5 mL water) and $NaHCO_{3(aq)}$ (saturated, 5 mL) for 10 min to quench the unreacted Dess-Martin reagent. The reaction mixture was diluted with $CH_2Cl_2$, and extracted by aqueous $NaHCO_3$. The combined organic layer was collected, dried over the $MgSO_4$ and concentrated *in vacuo*. The residue was purified through flash column chromatography [$R_f$ = 0.5 (40% ethyl acetate in hexanes)] to give the compound **3y'** as pale yellow solid (94 mg, 82%); mp: 223 $^o$C; IR (KBr): 3005, 1716, 1638, 1514, 1274, 1035 cm$^{-1}$; $^1$H NMR (600 MHz, $CDCl_3$) δ 9.56 (s, 1H), 7.87 (s, 1H), 6.91 (d, *J* = 8.4 Hz, 1H), 6.75 (s, 1H), 6.71–6.70 (m, 2H), 6.05 (s, 2H), 3.99 (s, 3 H), 3.93 (s, 3H), 3.51–3.50 (m, 2H), 3.30 (s, 3H); $^{13}$C NMR (150 MHz, $CDCl_3$) δ 199.6, 161.8, 153.5, 149.2, 148.4, 148.3, 141.6, 131.4, 128.4, 122.9, 119.4, 109.3, 109.0, 108.3, 106.0, 103.3, 101.6, 56.2, 56.0, 44.4, 34.2; HRMS [(EI), ($M^+$)]: 381.1216 (cal. for $C_{21}H_{19}NO_6$ 381.1212); Registry Number: [1207666-60-1].

**Procedure for the synthesis of oxynitidine:**

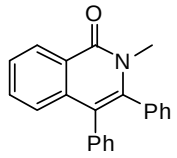
To a solution of compound **3y'** (114 mg, 0.3 mmol, 1.0 equiv) in acetic acid (4 mL) was added 10% hydrochloric acid (0.2 mL) at room temperature. After stirring the reaction for 8 h, acetic acid was removed in *vacuo*. The resulted solid was then dissolved in $CH_2Cl_2$ and extracted by aqueous $NaHCO_3$. The combined organic layer was collected, dried over the $MgSO_4$ and concentrated *in vacuo*. The residue was purified through flash column chromatography [$R_f$ = 0.59 (70% ethyl acetate in hexanes)] to give oxynitidine as white solid (96 mg, 88%); mp: 279 $^o$C; IR (KBr): 3005, 1637, 1274, 1041 cm$^{-1}$; $^1$H NMR (600 MHz, $CDCl_3$) δ 7.99 (d, *J* = 9.0 Hz, 1H), 7.93 (s, 1H), 7.64 (s, 1H), 7.59 (s, 1H), 7.56 (d, *J* = 9.0 Hz, 1H), 7.18 (s, 1H), 6.10 (s, 2H), 4.10 (s, 3H), 4.06 (s, 3H), 3.98 (s, 3H); $^{13}$C NMR (150 MHz, $CDCl_3$) δ 164.3, 153.5, 149.7, 147.5, 147.0, 135.9, 131.8, 128.9, 123.2, 121.0, 119.1, 118.3, 116.7, 108.6, 104.8, 102.8, 102.6, 101.5, 56.3, 56.1, 41.2; HRMS [(EI), ($M^+$)]: 363.1110 (cal. for $C_{21}H_{17}NO_5$ 363.1107); Registry Number: [548-31-2].

**Procedure for the synthesis of nitidine chloride:**

$LiAlH_4$ (11 mg, 0.3 mmol, 1.0 equiv) was added to a solution of oxynitidine (109 mg, 0.3 mmol, 1.0 equiv) in dry THF (5 mL) and kept stirring at room temperature for 60 min. EtOAc was then added to quench the excess hydride. Filter and concentrated, the reaction residue was then treated with 10% HCl (5 mL) at room temperature. the resulting precipitates were collected by filtration to afford nitidine chloride as yellow solid (95 mg, 91%); mp: 280 $^o$C; IR (KBr): 1260, 1275, 1636 cm$^{-1}$; $^1$H NMR (600 MHz, DMSO-$d_6$) δ 9.87 (s, 1H), 8.89 (d, *J* = 9.0 Hz, 1H), 8.35 (s, 1H), 8.30 (s, 1H), 8.27 (d, *J* = 9.0 Hz, 1H), 7.91 (s, 1H), 7.76 (s, 1H), 6.33 (s, 2H), 4.89 (s, 3H), 4.22 (s, 3H), 4.03 (s, 3H); $^{13}$C NMR (150 MHz, $CDCl_3$) δ 164.3, 153.5, 149.7, 147.5, 147.0, 135.9, 131.9, 128.9, 123.2, 121.0, 119.2, 118.4, 116.7, 108.7, 104.8, 102.8, 102.7, 101.5, 56.3, 56.1, 41.2; HRMS [(FAB), ($M^+$)]: 348.1236 (cal. for $C_{21}H_{18}NO_4^+$ 348.1236); Registry Number: [13063-04-2].

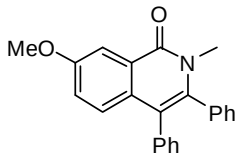
## Spectral data for all products:

**2-Methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3a):**



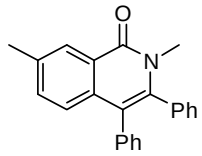
Work-up and purification by column chromatography, white solid (117 mg, 75%), mp: 246–248 $^{o}$C; IR (KBr): 1646, 1604, 1552, 1489, 1414, 1176, 1074, 1025, 924, 781 cm$^{-1}$; $^{1}$H NMR (600 MHz, $CDCl_3$): δ 8.57 (d, $J$ = 7.6 Hz, 1H), 7.53–7.47 (m, 2H), 7.26–7.12 (m, 9H), 7.07–7.05 (m, 2H), 3.36 (s, 3H); $^{13}$C NMR (150 MHz, $CDCl_3$): δ 162.7, 141.2, 137.1, 136.4, 135.0, 132.0, 131.5, 129.9, 128.1, 127.9, 127.7, 126.7, 126.5, 125.3, 124.9, 118.8, 34.3; HRMS [(ESI), $(M+H)^+$]: 312.1374 (cal. for $C_{22}H_{18}NO$ 312.1388); Registry Number: [148564-77-6].

**7-Methoxy-2-methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3b):**



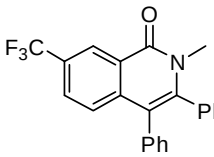
Work-up and purification by column chromatography, white solid (111 mg, 65%), mp: 213–214 $^{o}$C; IR (KBr): 2946, 1641, 1606, 1589, 1497, 1352, 1253, 1146, 1052, 949, 833, 727 cm$^{-1}$; $^{1}$H NMR (600 MHz, $CDCl_3$): δ 7.96 (s, 1H), 7.26–7.05 (m, 12H), 3.96 (s, 3H), 3.37 (s, 3H); $^{13}$C NMR (150 MHz, $CDCl_3$): δ 162.4, 158.6, 138.9, 136.6, 135.1, 131.5, 131.3, 130.2, 128.1, 128.1, 127.9, 127.1, 126.7, 126.1, 122.5, 118.9, 107.5, 55.7, 34.5; HRMS [(EI), $(M^+)$]: 341.1416 (cal. for $C_{23}H_{19}NO_2$ 341.1416); Registry Number: [1235478-95-1].

**2,7-Dimethyl-3,4-diphenylisoquinolin-1(2*H*)-one (3c):**



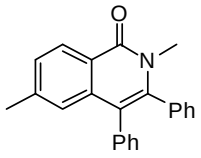
Work-up and purification by column chromatography, colorless solid (104 mg, 64%), mp: 224–225 $^{o}$C; IR (KBr): 1645, 1499, 1340, 1145, 948, 830, 769, 730 cm$^{-1}$; $^{1}$H NMR (400 MHz, $CDCl_3$): δ 8.36 (s, 1H), 7.35 (dd, $J$ = 8.2, 1.8 Hz, 1H), 7.24–7.04 (m, 11H), 3.35 (s, 3H), 2.50 (s, 3H); $^{13}$C NMR (100 MHz, $CDCl_3$): δ 162.7, 140.3, 136.7, 136.3, 135.2, 134.9, 133.5, 131.5, 130.0, 128.2, 128.1, 127.8, 127.4, 126.7, 125.3, 124.8, 118.8, 34.3, 21.4; HRMS [(ESI), $(M+Na)^+$]: 348.1361 (cal. for $C_{23}H_{19}NONa$ 348.1364); Registry Number: [1315257-16-9].

**2-Methyl-3,4-diphenyl-7-(trifluoromethyl)isoquinolin-1(2*H*)-one (3d):**



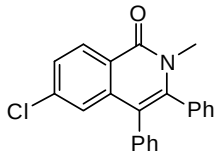
Work-up and purification by column chromatography, colorless solid (144 mg, 76%), mp: 199–200 $^{o}$C; IR (KBr): 3054, 2924, 1727, 1652, 1553, 1496, 1409, 1313, 1117, 1070, 1008, 839, 703 cm$^{-1}$; $^{1}$H NMR (400 MHz, $CDCl_3$): δ 8.85 (s, 1H), 7.70 (dd, $J$ = 8.6, 2.0 Hz, 1H), 7.29–7.03 (m, 10H), 6.84 (s, 1H), 3.37 (s, 3H); $^{13}$C NMR (100 MHz, $CDCl_3$): δ 162.1, 143.6, 139.5, 135.1 (q, $J_{C–F}$ = 90 Hz), 131.4, 129.6, 128.7, 128.5, 128.3, 128.1, 128.0 (q, $J_{C–F}$ = 3 Hz), 127.2, 126.3, 125.8, 125.6 (q, $J_{C–F}$ = 4 Hz), 124.6, 123.4 (q, $J_{C–F}$ = 272 Hz), 118.3, 34.5; HRMS [(ESI), $(M+Na)^+$]: 402.1076 (cal. for $C_{23}H_{16}NOF_3Na$ 402.1082); Registry Number: [1315257-17-0].

**2,6-Dimethyl-3,4-diphenylisoquinolin-1(2*H*)-one (3e):**



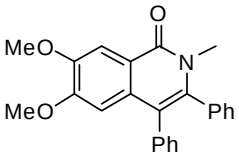
Work-up and purification by column chromatography, white solid (99 mg, 61%), mp: 230–231 $^{o}$C; IR (KBr): 2980, 1665, 1278, 1107, 1039, 816, 721 cm$^{-1}$; $^{1}$H NMR (600 MHz, $CDCl_3$): δ 8.45 (d, $J$ = 8.1 Hz, 1H), 7.32 (d, $J$ = 8.1 Hz, 1H), 7.26–7.14 (m, 6H), 7.12 (d, $J$ = 7.2 Hz, 2H), 7.06 (d, $J$ = 7.2 Hz, 2H), 6.93 (s, 1H), 3.34 (s, 3H), 2.34 (s, 3H); $^{13}$C NMR (150 MHz, $CDCl_3$): δ 162.8, 142.5, 141.3, 137.2, 136.6, 135.2, 131.6, 129.9, 128.2, 128.1, 128.1, 127.9, 126.7, 125.0, 122.8, 118.7, 34.2, 21.0; HRMS [(ESI), $(M+Na)^+$]: 348.1358 (cal. for $C_{23}H_{19}NONa$ 348.1364); Registry Number: [1989524-19-7].

**6-Chloro-2-methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3f):**



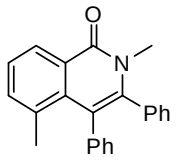
Work-up and purification by column chromatography, colorless solid (119 mg, 69%), mp: 266–267 °C; IR (KBr): 1649, 1597, 1443, 1416, 1360, 1190, 1070, 936, 869, 834, 784 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.48 (d, *J* = 8.4 Hz, 1H), 7.41 (d, *J* = 8.4 Hz, 1H), 7.28–7.00 (m, 10H), 6.91 (s, 1H), 3.37 (s, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 152.1, 142.7, 138.7, 138.5, 135.7, 131.4, 129.8, 128.4, 128.3, 128.2, 128.1, 127.2, 127.1, 124.7, 123.3, 118.0, 34.4; HRMS [(EI), ($M^+$)]: 345.0926 (cal. for $C_{22}H_{16}ClNO$ 345.0920); Registry Number: [1315257-11-4].

**6,7-Dimethoxy-2-methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3g):**



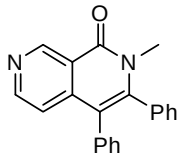
Work-up and purification by column chromatography, white solid (113 mg, 61%), mp: 240–242 °C; IR (KBr): 2954, 1645, 1604, 1483, 1415, 1230, 1143, 1072, 1001, 856, 781 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 7.93 (s, 1H), 7.26–7.04 (m, 10H), 6.50 (s, 1H), 4.03 (s, 3H), 3.68 (s, 3H), 3.35 (s, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.0, 153.1, 149.1, 140.0, 136.4, 135.2, 132.6, 131.1, 130.1, 128.2, 128.1, 128.0, 126.8, 119.0, 118.6, 107.7, 105.7, 56.3, 55.7, 34.5; HRMS [(EI), ($M^+$)]: 371.1520 (cal. for $C_{24}H_{21}NO_3$ 371.1521); Registry Number: [2101507-74-6].

**2,5-Dimethyl-3,4-diphenylisoquinolin-1(2*H*)-one (3h):**



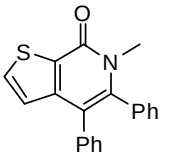
Work-up and purification by column chromatography, white solid (63 mg, 39%), mp: 200–201 °C; IR (KBr): 1644, 1495, 1337, 1142, 949, 834, 769, 733 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.55 (d, *J* = 7.8 Hz, 1H), 7.39 (t, *J* = 7.5 Hz, 1H), 7.36 (d, *J* = 7.2 Hz, 1H), 7.21–7.15 (m, 3H), 7.08–7.03 (m, 7H), 3.29 (s, 3H), 1.74 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 162.9, 141.9, 139.9, 136.3, 135.5, 135.0, 134.9, 132.0, 130.0, 128.0, 127.8, 127.3, 126.7, 126.5, 126.5, 126.3, 118.6, 34.3, 23.7; HRMS [(ESI), ($M+Na)^+$]: 348.1361 (cal. for $C_{23}H_{19}NONa$ 348.1364); New compound.

**2-Methyl-3,4-diphenyl-2,7-naphthyridin-1(2*H*)-one (3i):**



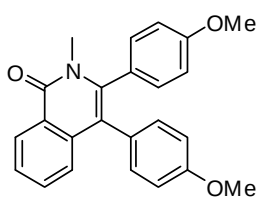
Work-up and purification by column chromatography, brown solid (83 mg, 53%), mp: 185–186 °C; IR (KBr): 1667, 1495, 1337, 1142, 1010, 940, 834, 769, 743 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.71–8.65 (m, 2H), 8.34 (d, *J* = 5.1 Hz, 1H), 7.29–7.21 (m, 6H), 7.16–7.09 (m, 4H), 3.39 (s, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 161.5, 148.9, 145.6, 143.3, 134.5, 134.2, 131.6, 131.3, 129.8, 129.6, 128.7, 128.4, 128.2, 127.4, 120.0, 117.2, 34.7; HRMS [(ESI), ($M+H)^+$]: 313.1335 (cal. for $C_{21}H_{17}N_2O$ 313.1341); New compound.

**6-Methyl-4,5-diphenylthieno[2,3-*c*]pyridin-7(6*H*)-one (3j):**



Work-up and purification by column chromatography, colorless oil (95 mg, 60%); IR (KBr): 3058, 1640, 1567, 1488, 1441, 789, 713 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 7.59 (d, *J* = 5.2 Hz, 1H), 7.27–7.05 (m, 10H), 6.92 (d, *J* = 5.2 Hz, 1H), 3.39 (s, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 158.8, 145.6, 142.4, 136.8, 134.6, 132.7, 130.6, 130.1, 128.8, 128.4, 128.3, 127.9, 126.8, 124.7, 117.7, 34.2; HRMS [(ESI), ($M+Na)^+$]: 340.0766 (cal. for $C_{20}H_{15}NOSNa$ 340.0772); Registry Number: [1235479-02-3].

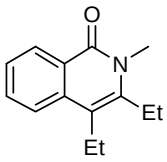
**3,4-Bis(4-methoxyphenyl)-2-methylisoquinolin-1(2*H*)-one (3k):**



Work-up and purification by column chromatography, white solid (102 mg, 55%); mp: 224–225 °C; IR (KBr): 3502, 1698, 1329, 782 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.54 (d, *J* = 7.2 Hz, 1H), 7.52 (td, *J* = 7.2, 0.6 Hz, 1H), 7.47 (t, *J* = 7.8 Hz, 1H), 7.18 (d, *J* = 7.8 Hz, 1H), 7.03 (d, *J* = 8.7 Hz, 2H), 6.97 (d, *J* = 8.4 Hz, 2H), 6.77 (d, *J* = 7.8 Hz, 2H), 6.75 (d, *J* = 7.8 Hz, 2H), 3.76 (s, 6H), 3.35 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 162.8, 159.1, 158.2, 141.3, 137.5, 132.5, 131.9, 131.1, 128.9, 127.8, 127.6, 126.4, 125.3, 124.9, 118.7, 113.6, 113.4,

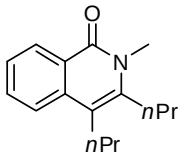
55.1, 55.0, 34.3; HRMS [(ESI), $(M+Na)^+$]: 394.1414 (cal. for $C_{24}H_{21}NO_3Na$ 394.1419); Registry Number: [161730-00-3].

**3,4-Diethyl-2-methylisoquinolin-1(2*H*)-one (3l):**



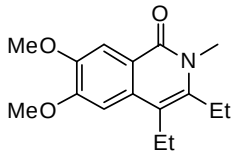
Work-up and purification by column chromatography, yellow oil (57 mg, 53%); IR (KBr): 1646, 1587, 1557, 1457, 1372, 1173, 1057, 898, 740 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.45 (dd, *J* = 7.6, 1.2 Hz, 1H), 7.63–7.61 (m, 2H), 7.41 (t, *J* = 7.6 Hz, 1H), 3.65 (s, 3H), 2.80–2.72 (m, 4H), 1.25–1.17 (m, 6H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.9, 140.7, 136.2, 132.0, 128.3, 125.6, 124.8, 122.4, 114.9, 31.1, 22.7, 20.5, 14.8, 13.5; HRMS [(ESI), $(M+Na)^+$]: 238.1204 (cal. for $C_{14}H_{17}NONa$ 238.1208); New compound.

**2-Methyl-3,4-dipropylisoquinolin-1(2*H*)-one (3m):**



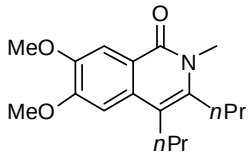
Work-up and purification by column chromatography, colorless solid (66 mg, 54%), mp: 72 °C; IR (KBr): 3274, 1644, 1587, 1591, 1555, 1448, 1333, 1173, 1057, 892, 776, 703 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.45 (dd, *J* = 8.0, 1.0 Hz, 1H), 7.62–7.61 (m, 2H), 7.42–7.38 (m, 1H), 3.65 (s, 3H), 2.73–2.67 (m, 4H), 1.66–1.54 (m, 4H), 1.06 (q, *J* = 6.8 Hz, 6H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.9, 139.8, 136.5, 131.9, 128.3, 125.5, 124.8, 122.6, 113.8, 31.8, 31.3, 29.8, 23.6, 22.6, 14.4, 14.2; HRMS [(ESI), $(M+Na)^+$]: 266.1515 (cal. for $C_{16}H_{21}NONa$ 266.1521); Registry Number: [1315257-19-2].

**3,4-Diethyl-6,7-dimethoxy-2-methylisoquinolin-1(2*H*)-one (3n):**



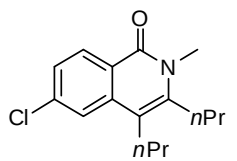
Work-up and purification by column chromatography, yellow oil (66 mg, 48%); IR (KBr): 2994, 1635, 1611, 1483, 1425, 1229, 1155, 1069, 1011, 876, 787 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 7.80 (s, 1H), 6.98 (s, 1H), 3.98 (s, 6H), 3.66 (s, 3H), 2.77 (q, *J* = 7.2 Hz, 4H), 1.26–1.20 (m, 6H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.1, 153.1, 148.3, 139.4, 131.7, 118.8, 114.5, 108.2, 102.9, 56.1, 56.0, 31.2, 22.7, 20.9, 14.7, 13.6; HRMS [(EI), $(M^+)$]: 275.1521 (cal. for $C_{16}H_{21}NO_3$ 275.1521); New compound.

**6,7-Dimethoxy-2-methyl-3,4-dipropylisoquinolin-1(2*H*)-one (3o):**



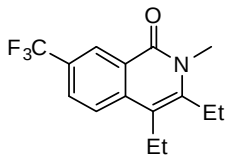
Work-up and purification by column chromatography, white solid (65 mg, 43%), mp: 93 °C; IR (KBr): 2974, 1645, 1617, 1411, 1217, 1143, 1072, 1001, 967, 856, 781 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 7.82 (s, 1H), 6.94 (s, 1H), 3.97 (s, 6H), 3.63 (s, 3H), 2.70–2.63 (m, 4H), 1.59 (sext, *J* = 7.2 Hz, 4H), 1.07–1.02 (m, 6H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.1, 153.1, 148.3, 138.6, 132.0, 118.8, 113.4, 108.1, 103.2, 56.1, 55.8, 31.8, 31.4, 30.1, 23.6, 22.7, 14.5, 14.3; HRMS [(EI), $(M^+)$]: 303.1829 (cal. for $C_{18}H_{25}NO_3$ 303.1834); New compound.

**6-Chloro-2-methyl-3,4-dipropylisoquinolin-1(2*H*)-one (3p):**



Work-up and purification by column chromatography, white solid (82 mg, 59%), mp: 88 °C; IR (KBr): 3283, 1653, 1547, 1451, 1257, 1340, 1197, 1075, 939, 868 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.37 (d, *J* = 8.6 Hz, 1H), 7.57 (d, *J* = 1.9 Hz, 1H), 7.35 (dd, *J* = 8.6, 1.8 Hz, 1H), 3.64 (s, 3H), 2.73–2.62 (m, 4H), 1.67–1.53 (m, 4H), 1.08 (q, *J* = 7.0 Hz, 6H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.4, 141.5, 138.6, 137.9, 130.2, 126.1, 123.1, 122.2, 113.1, 31.9, 31.4, 29.8, 23.6, 22.5, 14.4, 14.3; HRMS [(ESI), $(M+Na)^+$]: 300.1132 (cal. for $C_{16}H_{20}ClNONa$ 300.1131); New compound.

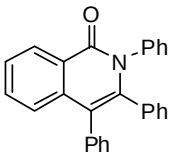
**3,4-Diethyl-2-methyl-7-(trifluoromethyl)isoquinolin-1(2*H*)-one (3q):**



Work-up and purification by column chromatography, white solid (89 mg, 63%), mp: 82–83 °C; IR (KBr): 2953, 1648, 1604, 1483, 1315, 1230, 1110, 1072, 1001, 855, 777 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.73 (s, 1H), 7.81–7.73 (m, 2H), 3.68 (s, 3H), 2.85–2.75 (m, 4H), 1.29–1.20 (m, 6H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.3, 143.5, 138.7, 127.9 (q, $J_{C-F}$ = 3 Hz), 127.4 (q, $J_{C-F}$ = 33 Hz),

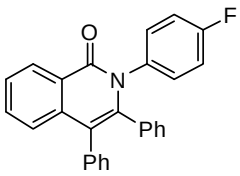
126.1 (q, $J_{C-F}$ = 4 Hz), 124.5, 124.1 (q, $J_{C-F}$ = 270 Hz), 123.4, 114.5, 31.3, 22.9, 20.6, 14.7, 13.4; HRMS [(ESI), $(M+H)^+$]: 284.1264 (cal. for $C_{15}H_{17}F_3NO$ 284.1262); New compound.

**2,3,4-Triphenylisoquinolin-1(2*H*)-one (3r):**



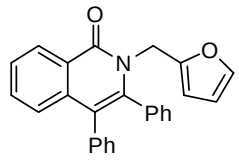
Work-up and purification by column chromatography, white solid (86 mg, 46%), mp: 202–203 °C; IR (KBr): 3013, 1951, 1652, 1613, 1588, 1489, 1422, 1327, 1257, 1120, 1029, 922, 803, 767 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.58 (d, *J* = 7.5 Hz, 1H), 7.60 (t, *J* = 7.2 Hz, 1H), 7.54 (t, *J* = 7.2 Hz, 1H), 7.28–7.11 (m, 11H), 6.90 (br, 5H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.7, 141.1, 139.5, 137.7, 136.4, 134.8, 132.6, 131.6, 131.1, 129.5, 128.6, 128.3, 128.0, 127.6, 127.3, 127.1, 126.9, 126.8, 125.6, 125.5, 118.9; HRMS [(ESI), $(M+Na)^+$]: 396.1358 (cal. for $C_{27}H_{19}NONa$ 396.1364); Registry Number: [14959-72-9].

**2-(4-Fluorophenyl)-3,4-diphenylisoquinolin-1(2*H*)-one (3s):**



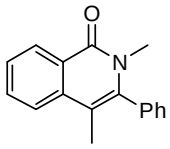
Work-up and purification by column chromatography, white solid (123 mg, 63%), mp: 238–239 °C; IR (KBr): 1899, 1661, 1587, 1554, 1442, 1328, 1227, 1090, 1012, 923, 854, 784, 747 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.58 (d, *J* = 10.6 Hz, 1H), 7.62–7.53 (m, 2H), 7.30–7.08 (m, 8H), 6.95–6.90 (m, 7H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.7, 139.3 (d, $J_{C-F}$ = 234 Hz), 137.6, 136.2, 134.6, 132.6, 131.5, 131.2, 131.1, 131.0, 130.9, 128.2, 128.0, 127.4, 127.3, 127.0, 126.9, 125.7, 125.4, 119.0, 115.6 (d, $J_{C-F}$ = 30 Hz); HRMS [(ESI), $(M+Na)^+$]: 414.1263 (cal. for $C_{27}H_{18}FNONa$ 414.1270); Registry Number: [1253388-48-5].

**2-(Furan-2-ylmethyl)-3,4-diphenylisoquinolin-1(2*H*)-one (3t):**



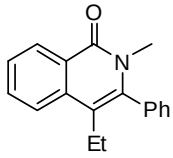
Work-up and purification by column chromatography, brown solid (108 mg, 57%), mp: 152–153 °C; IR (KBr): 1950, 1656, 1606, 1557, 1486, 1422, 1324, 1008, 748 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.59 (s, 1H), 7.56–7.48 (m, 2H), 7.25–7.06 (m, 12H), 6.25–6.23 (m, 1H), 6.06 (dd, *J* = 3.2, 0.6 Hz, 1H), 5.13 (s, 2H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.3, 150.5, 141.5, 140.9, 137.3, 136.4, 134.2, 132.3, 131.5, 130.5, 128.3, 128.1, 127.9, 127.8, 126.8, 126.7, 125.4, 125.1, 119.4, 110.3, 108.2, 42.6; HRMS [(EI), $(M^+)$]: 377.1412 (cal. for $C_{26}H_{19}NO_2$ 377.1416); New compound.

**2,4-Dimethyl-3-phenylisoquinolin-1(2*H*)-one (3u):**



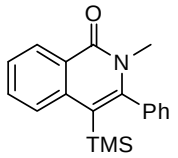
Work-up and purification by column chromatography, yellow solid (54 mg, 43%), mp: 104–105 °C; IR (KBr): 2978, 1644, 1641, 1283, 1085, 762, 709 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.54 (d, *J* = 8.4 Hz, 1H), 7.70–7.69 (m, 2H), 7.53–7.46 (m, 4H) 7.28–7.26 (m, 2H), 3.26 (s, 3H), 2.02 (s, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.6, 140.2, 137.1, 135.8, 132.1, 129.4, 129.0, 128.7, 128.1, 126.4, 125.2, 123.2, 110.4, 34.2, 14.8; HRMS [(EI), $(M^+)$]: 249.1142 (cal. for $C_{17}H_{15}NO$ 249.1154); Registry Number: [51089-64-6]. For this stereoisomer, spectral data matches the reported literature.[2] NOE data was not collected.

**4-Ethyl-2-methyl-3-phenylisoquinolin-1(2*H*)-one (3v):**



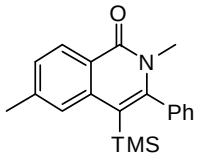
Work-up and purification by column chromatography, pale yellow solid (53 mg, 40%), mp: 144 °C; IR (KBr): 2968, 1648, 1613, 1487, 1333, 1055, 762, 703 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 8.56 (d, *J* = 8.0 Hz, 1H), 7.72–7.70 (m, 2H), 7.53–7.48 (m, 4H), 7.30–7.26 (m, 2H), 3.23 (s, 3H), 2.44 (q, *J* = 7.2 Hz, 2H), 1.05 (t, *J* = 7.2 Hz, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 162.4, 140.1, 136.0, 135.6, 132.0, 129.1, 129.0, 128.7, 128.4, 126.3, 125.7, 123.1, 116.6, 34.0, 21.6, 14.8; HRMS [(ESI), $(M+Na)^+$]: 286.1206 (cal. for $C_{18}H_{17}NONa$ 286.1208); Registry Number: [1235479-12-5]. For this stereoisomer, spectral data matches the reported literature.[2] NOE data was not collected.

**2-Methyl-3-phenyl-4-(trimethylsilyl)isoquinolin-1(2*H*)-one (3w):**



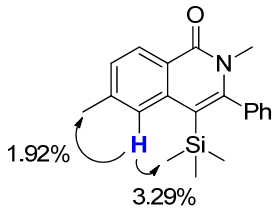

Work-up and purification by column chromatography, white solid (80 mg, 52%), mp: 174–175 °C; IR (KBr): 3275, 1645, 1328, 928, 524 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.53 (d, *J* = 8.1 Hz, 1H), 7.88 (d, *J* = 8.1 Hz, 1H), 7.63 (t, *J* = 7.8 Hz, 1H), 7.49–7.47 (m, 4H), 7.30–7.30 (m, 2H), 3.19 (s, 3H), -0.04 (s, 9H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 163.1, 148.7, 139.6, 137.7, 131.1, 130.2, 129.2, 128.6, 128.2, 127.2, 125.9, 125.6, 111.7, 34.0, 2.0; HRMS [(ESI), $(M+H)^+$]: 308.1473 (cal. for $C_{19}H_{22}NOSi$ 308.1471); New compound.

**2,6-Dimethyl-3-phenyl-4-(trimethylsilyl)isoquinolin-1(2*H*)-one (3x):**

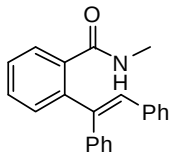



Work-up and purification by column chromatography, white solid (74 mg, 46%), mp: 134–135 °C; IR (KBr): 3726, 2958, 1650, 885 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 8.41 (d, *J* = 8.1 Hz, 1H), 7.64 (s, 1H), 7.47–7.46 (m, 3H), 7.31–7.28 (m, 3H), 3.17 (s, 3H), 2.50 (s, 3H), -0.04 (s, 9H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 163.1, 148.8, 141.4, 139.7, 137.8, 130.2, 129.1, 128.6, 128.2, 127.5, 127.2, 123.3, 111.4, 33.9, 22.1, 2.1; HRMS [(ESI), $(M+H)^+$]: 322.1631 (cal. for $C_{20}H_{24}NOSi$ 322.1627); New compound.

**NOE data:**

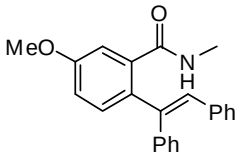



**(*E*)-2-(1,2-Diphenylvinyl)-*N*-methylbenzamide (4a):**



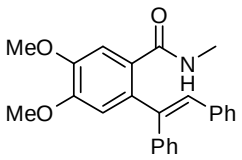

Work-up and purification by column chromatography, yellow solid (118 mg, 75%), mp: 132–133 °C; IR (KBr): 3449, 1625, 1312, 764, 517 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.51 (d, *J* = 7.8 Hz, 1H), 7.44–7.39 (m, 2H), 7.36 (t, *J* = 7.2 Hz, 1H), 7.22–7.10 (m, 10H), 6.79 (s, 1H), 2.61 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.0, 142.2, 141.9, 139.4, 137.1, 136.5, 130.9, 130.6, 130.4, 129.9, 129.4, 128.3, 128.1, 128.0, 127.8, 127.6, 127.1, 26.6; HRMS [(EI), $(M^+)$]: 313.1462 (cal. for $C_{22}H_{19}NO$ 313.1467); Registry Number: [1315257-10-3].

**(*E*)-2-(1,2-Diphenylvinyl)-5-methoxy-*N*-methylbenzamide (4b):**



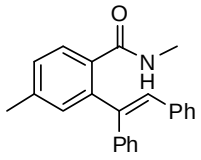

Work-up and purification by column chromatography, white solid (113 mg, 66%), mp: 214–215 °C; IR (KBr): 3280, 1645, 1612, 1589, 1526, 1350, 1261, 1134, 1052, 961, 866, 728 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.31 (d, *J* = 8.4 Hz, 1H), 7.21–7.08 (m, 11H), 6.95 (dd, *J* = 8.4, 2.7 Hz, 1H), 6.76 (s, 1H), 3.83 (s, 3H), 2.6 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.6, 159.2, 141.5, 139.6, 137.5, 137.2, 134.5, 132.3, 130.3, 130.0, 129.4, 128.1, 128.0, 127.6, 127.0, 116.0, 113.2, 55.5, 26.6; HRMS [(ESI), $(M+H)^+$]: 344.1650 (cal. for $C_{23}H_{22}NO_2$ 344.1651); New compound.

**(*E*)-2-(1,2-Diphenylvinyl)-4,5-dimethoxy-*N*-methylbenzamide (4c):**



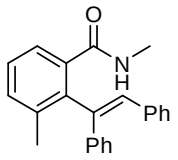

Work-up and purification by column chromatography, yellow solid (121 mg, 65%), mp: 228–230 °C; IR (KBr): 3262, 1633, 1316, 761, 565 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.21–7.09 (m, 11H), 6.85 (s, 1H), 6.79 (s, 1H), 3.92 (s, 3H), 3.90 (s, 3H), 2.62 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.3, 149.9, 148.4, 142.0, 139.2, 136.9, 135.2, 130.3, 130.1, 129.3, 128.5, 128.1, 127.8, 127.2, 113.8, 111.8, 56.1, 56.0, 26.7; HRMS [(ESI), $(M+H)^+$]: 374.1757 (cal. for $C_{24}H_{24}NO_3$ 374.1756); New compound.

**(*E*)-2-(1,2-Diphenylvinyl)-*N*,4-dimethylbenzamide (4d):**



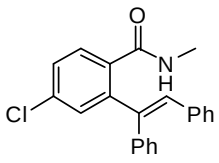
Work-up and purification by column chromatography, white solid (105 mg, 64%), mp: 154–155 °C; IR (KBr): 3283, 1622, 1309, 831, 696 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.44 (d, *J* = 7.8 Hz, 1H), 7.21–7.12 (m, 12H), 6.78 (s, 1H), 2.61 (d, *J* = 4.8 Hz, 3H), 2.39 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.9, 142.1, 142.1, 139.9, 139.4, 137.1, 133.6, 131.5, 130.3, 129.4, 128.5, 128.4, 128.1, 127.5, 127.1, 26.6, 21.3; HRMS [(ESI), $(M+H)^+$]: 328.1703 (cal. for $C_{23}H_{22}NO$ 328.1701); Registry Number: [1427042-11-2].

**(*E*)-2-(1,2-Diphenylvinyl)-*N*,3-dimethylbenzamide (4e):**



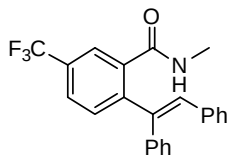
Work-up and purification by column chromatography, white solid (93 mg, 57%), mp: 157–158 °C; IR (KBr): 1702, 1516, 1328, 1122, 936, 836, 762, 730 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.43 (d, *J* = 7.2 Hz, 1H), 7.31–7.21 (m, 2H), 7.21 (br, 5H), 7.18 (br, 5H), 6.63 (s, 1H), 2.76 (d, *J* = 4.8 Hz, 3H), 2.27 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.4, 140.9, 140.1, 138.8, 137.3, 137.1, 131.9, 130.8, 130.0, 129.2, 128.2, 128.1, 127.4, 127.3, 127.1, 125.5, 26.7, 20.5; HRMS [(ESI), $(M+H)^+$]: 328.1702 (cal. for $C_{23}H_{22}NO$ 328.1701); New compound.

**(*E*)-4-Chloro-2-(1,2-diphenylvinyl)-*N*-methylbenzamide (4f):**



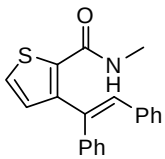
Work-up and purification by column chromatography, pale yellow solid (117 mg, 67%), mp: 177–178 °C; IR (KBr): 3302, 1626, 1307, 952, 712, 564 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.46 (d, *J* = 8.1 Hz, 1H), 7.40 (s, 1H), 7.33 (d, *J* = 8.1 Hz, 1H), 7.23–7.10 (m, 10H), 6.79 (s, 1H), 2.60 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.0, 144.0, 140.7, 138.7, 136.6, 135.7, 134.9, 131.4, 130.7, 130.3, 129.8, 129.4, 128.2, 128.1, 127.9, 127.8, 127.4, 26.6; HRMS [(ESI), $(M+H)^+$]: 348.1158 (cal. for $C_{22}H_{19}NOCl$ 348.1155); New compound.

**(*E*)-2-(1,2-diphenylvinyl)-*N*-methyl-5-(trifluoromethyl)benzamide (4g):**



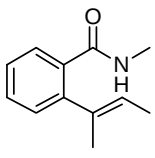
Work-up and purification by column chromatography, white solid (130 mg, 68%), mp: 158–159 °C; IR (KBr): 3279, 1648, 1341, 774 $cm^{-1}$; $^1H$ NMR (600 MHz, $C_6D_6$): δ 7 .57 (s, 1H), 7.29 (d, *J* = 8.4 Hz, 1H), 7.18–7.10 (m, 4H), 7.04 (d, *J* = 7.8 Hz, 1H), 6.99–6.92 (m, 6H), 6.64 (s, 1H), 4.74 (br, 1H), 2.20 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (75 MHz, $CDCl_3$): δ 168.5, 140.5, 145.7, 138.7, 137.1, 136.4, 131.7, 131.3, 131.3, 130.3, 129.8 (q, $J_{C-F}$ = 33 Hz), 129.4, 128.2, 128.1, 127.9, 127.5, 126.4 (q, $J_{C-F}$ = 4 Hz), 125.3 (q, $J_{C-F}$ = 4 Hz), 120 (q, $J_{C-F}$ = 271 Hz), 26.6; HRMS [(ESI), $(M+Na)^+$]: 404.1238 (cal. for $C_{23}H_{18}F_3NONa$ 404.1238); New compound.

**(*E*)-3-(1,2-Diphenylvinyl)-*N*-methylthiophene-2-carboxamide (4h):**



Work-up and purification by column chromatography, white solid (107 mg, 67%), mp: 170–171 °C; IR (KBr): 3445, 1698, 1339, 749 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.35 (dd, *J* = 4.8, 1.8 Hz, 1H), 7.27–7.26 (m, 4H), 7.20–7.19 (m, 4H), 7.16–7.15 (m, 2H), 6.88–6.87 (m, 2H), 6.44 (s, 1H), 2.76 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 162.9, 143.7, 138.6, 136.5, 136.0, 135.6, 132.2, 130.8, 129.5, 129.3, 128.6, 128.3, 128.1, 128.0, 127.7, 26.5; HRMS [(ESI), $(M+Na)^+$]: 342.0925 (cal. for $C_{20}H_{17}NOSNa$ 342.0928); Registry Number: [1315257-18-1].

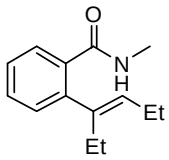
**(*E*)-2-(But-2-en-2-yl)-*N*-methylbenzamide (4i):**



Work-up and purification by column chromatography, colorless liquid, (47 mg, 50%); IR (KBr): 3446, 1698, 1540, 740 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.61 (d, *J* = 7.8 Hz, 1H), 7.33 (t, *J* = 7.8 Hz, 1H), 7.26 (t, *J* = 7.8 Hz, 1H), 7.13 (d, *J* = 7.8 Hz, 1H), 6.11 (s, 1H), 5.60 (q, *J* = 6.6 Hz, 1H), 2.92 (d, *J* = 4.8 Hz, 3H), 1.91 (s, 3H), 1.77 (d, *J* = 6.6 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.1, 143.5, 137.3, 134.1, 130.1,

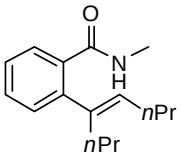
129.2, 128.5, 126.9, 125.1, 26.8, 17.7, 14.1; HRMS [(ESI), $(M+Na)^+$]: 212.1052 (cal. for $C_{12}H_{15}NONa$ 212.1051); New compound.

**(*E*)-2-(Hex-3-en-3-yl)-*N*-methylbenzamide (4j):**



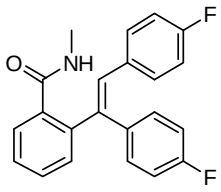
Work-up and purification by column chromatography, colorless liquid, (59 mg, 54%); IR (KBr): 3421, 1646, 1540, 750 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.70 (dd, *J* = 7.8, 1.8 Hz, 1H), 7.34 (td, *J* = 7.8, 1.8 Hz, 1H), 7.29 (t, *J* = 7.8, 1.2 Hz, 1H), 7.12 (d, *J* = 7.2 Hz, 1H), 6.24 (s, 1H), 5.47 (t, *J* = 7.8 Hz, 1H), 2.91 (d, *J* = 5.4 Hz, 3H), 2.34 (q, *J* = 7.2 Hz, 2H), 2.20 (quint, *J* = 7.2 Hz, 2H), 1.05 (t, *J* = 7.8 Hz, 3H), 0.84 (t, *J* = 7.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.8, 142.8, 141.8, 134.0, 132.1, 130.1, 130.0, 128.8, 127.1, 26.6, 25.0, 21.4, 14.4, 13.0; HRMS [(ESI), $(M+Na)^+$]: 240.1373 (cal. for $C_{14}H_{19}NONa$ 240.1364); New compound.

**(*E*)-*N*-Methyl-2-(oct-4-en-4-yl)benzamide (4k):**



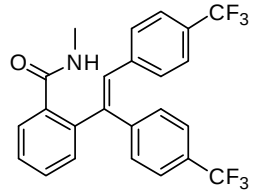
Work-up and purification by column chromatography, colorless liquid, (65 mg, 53%); IR (KBr): 3420, 1646, 1540, 750 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.69 (dd, *J* = 7.8, 1.2 Hz, 1H), 7.33 (td, *J* = 7.2, 1.2 Hz, 1H), 7.28 (td, *J* = 7.8, 1.2 Hz, 1H), 7.11 (d, *J* = 7.8 Hz, 1H), 6.27 (s, 1H), 5.51 (t, *J* = 7.2 Hz, 1H), 2.90 (d, *J* = 1.2 Hz, 3H), 2.29 (t, *J* = 7.8 Hz, 2H), 2.18 (q, *J* = 7.2 Hz, 2H), 1.46 (sext, *J* = 7.2 Hz, 2H), 1.22 (sext, *J* = 7.8 Hz, 2H), 0.97 (t, *J* = 7.8 Hz, 3H), 0.81 (t, *J* = 7.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.7, 142.3, 142.1, 133.9, 131.3, 130.1, 130.0, 128.8, 127.0, 34.0, 30.3, 26.6, 22.9, 21.5, 13.9, 13.8; HRMS [(ESI), $(M+Na)^+$]: 268.1678 (cal. for $C_{16}H_{23}NONa$ 268.1677); Registry Number: [1427042-15-6].

**(*E*)-2-(1,2-Bis(4-fluorophenyl)vinyl)-*N*-methylbenzamide (4l):**



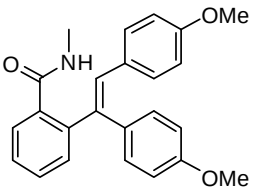
Work-up and purification by column chromatography, white solid (112 mg, 64%), mp: 139–140 °C; IR (KBr): 3446, 1683, 1457, 1262, 746 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.46 (d, *J* = 7.8 Hz, 1H), 7.43 (t, *J* = 7.8 Hz, 1H), 7.39–7.34 (m, 2H), 7.09–7.05 (m, 4H), 6.91 (t, *J* = 8.4 Hz, 2H), 6.87 (t, *J* = 8.4 Hz, 2H), 6.73 (s, 1H), 5.59 (s, 1H), 2.63 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.9, 162.7 (d, $J_{C–F}$ = 247 Hz), 161.8 (d, $J_{C–F}$ = 246 Hz), 141.9, 140.8, 136.6, 135.2 (d, $J_{C–F}$ = 3 Hz), 133.0 (d, $J_{C–F}$ = 3 Hz), 132.2 (d, $J_{C–F}$ = 8 Hz), 131 (d, $J_{C–F}$ = 8 Hz), 130.7, 129.9, 129.2, 128 (d, $J_{C–F}$ = 9 Hz), 115.1 ($J_{C–F}$ = 21 Hz), 115.1 (d, $J_{C–F}$ = 21 Hz), 26.5; HRMS [(ESI), $(M+Na)^+$]: 372.1172 (cal. for $C_{22}H_{17}F_2NONa$ 372.1175); New compound.

**(*E*)-2-(1,2-Bis(4-(trifluoromethyl)phenyl)vinyl)-*N*-methylbenzamide (4m):**



Work-up and purification by column chromatography, colorless liquid, (153 mg, 68%); IR (KBr): 3445, 1698, 1324, 749 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.47–7.36 (m, 8H), 7.26–7.24 (m, 2H), 7.17 (d, *J* = 8.4 Hz, 2H), 6.86 (s, 1H), 5.55 (s, 1H), 2.58 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.7, 142.9, 142.7, 141.3, 140.1, 136.8, 130.9, 130.8, 130.1, 129.8, 129.6, 129.1 (q, $J_{C–F}$ = 32 Hz), 128.3, 127.6, 125.2, 125.1, 125.0, 125.0, 124.0 (q, $J_{C–F}$ = 270 Hz), 123.9 (q, $J_{C–F}$ = 270 Hz), 26.4; HRMS [(ESI), $(M+Na)^+$]: 472.1110 (cal. for $C_{24}H_{17}F_6NONa$ 472.1112); New compound.

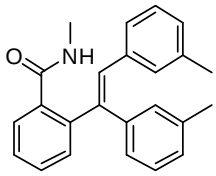
**(*E*)-2-(1,2-Bis(4-methoxyphenyl)vinyl)-*N*-methylbenzamide (4n):**



Work-up and purification by column chromatography, white solid (117 mg, 63%), mp: 231–232 °C; IR (KBr): 3310, 1645, 1540, 757 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.52 (d, *J* = 7.8 Hz, 1H), 7.41–7.31 (m, 3H), 7.08 (d, *J* = 8.4 Hz, 2H), 7.04 (d, *J* = 8.4 Hz, 2H), 6.74 (d, *J* = 8.4 Hz, 2H), 6.71 (d, *J* = 8.4 Hz, 2H), 6.65 (s, 1H), 5.82 (s, 1H), 3.76 (s, 6H), 2.62 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.9, 158.8, 158.5, 142.6, 139.7, 136.3, 131.9, 131.5, 130.8, 130.5, 129.8, 129.7, 129.3, 128.4, 127.5, 113.5, 113.4, 55.1, 55.0, 26.6; HRMS

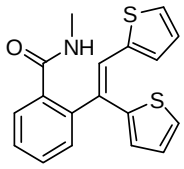
[(ESI), $(M+Na)^+$]: 396.1568 (cal. for $C_{24}H_{23}NO_3Na$ 396.1575); Registry Number: [1427042-13-4].

**(*E*)-2-(1,2-Di-*m*-tolylvinyl)-*N*-methylbenzamide (4o):**



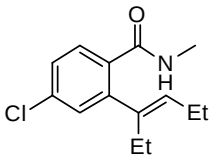
Work-up and purification by column chromatography, yellow solid (107 mg, 63%), mp: 118–119 °C; IR (KBr): 3445, 1646, 1540, 744 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.55 (d, *J* = 7.8Hz, 1H), 7.43–7.40 (m, 1H), 7.37–7.34 (m, 2H), 7.10 (t, *J* = 7.8 Hz, 1H), 7.04 (t, *J* = 7.8 Hz, 2H), 6.97 (d, *J* = 7.2 Hz, 2H), 6.93–6.89 (m, 3H), 6.72 (s, 1H), 5.74 (s, 1H), 2.65 (d, *J* = 5.4 Hz, 3H), 2.22 (d, *J* = 3.6 Hz, 6H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.9, 142.3, 141.7, 139.4, 137.6, 137.5, 136.9, 136.3, 130.8, 130.7, 130.6, 130.3, 129.8, 128.4, 128.3, 127.9, 127.9, 127.8, 127.7, 127.4, 126.3, 26.6, 21.3; HRMS [(ESI), $(M+Na)^+$]: 364.1671 (cal. for $C_{24}H_{23}NONa$ 364.1677); New compound.

**((*Z*)-2-(1,2-Di(thiophen-2-yl)vinyl)-*N*-methylbenzamide (4p):**



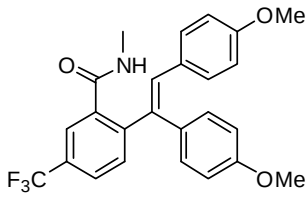
Work-up and purification by column chromatography, white solid (96 mg, 59%), mp: 227–228 °C; IR (KBr): 3556, 1652, 1540, 744 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.51 (d, *J* = 7.8 Hz, 1H), 7.41–7.37 (m, 3H), 7.35–7.33 (m, 1H), 7.17 (d, *J* = 4.8 Hz, 1H), 7.09 (d, *J* = 3.6 Hz, 1H), 7.05 (d, *J* = 3.6 Hz, 1H), 7.04–7.03 (m, 1H), 6.95 (s, 1H), 6.93 (t, *J* = 4.8 Hz, 1H), 5.77 (s, 1H), 2.78 (d, *J* = 5.4 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.1, 140.9, 140.3, 139.6, 136.3, 132.2, 129.9, 129.8, 129.3, 128.3, 127.9, 127.5, 127.1, 126.6, 126.5, 126.1, 26.8; HRMS [(ESI), $(M+Na)^+$]: 348.0491 (cal. for $C_{18}H_{15}NOS_2Na$ 348.0492); New compound.

**(*E*)-4-Chloro-2-(hex-3-en-3-yl)-*N*-methylbenzamide (4q):**



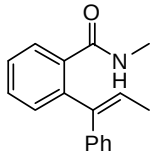
Work-up and purification by column chromatography, white solid (70 mg, 56%), mp: 181–182 °C; IR (KBr): 3283, 1653, 1547, 1451, 1257, 1340, 1197, 1075, 939, 868 $cm^{-1}$; $^1H$ NMR (400 MHz, $CDCl_3$): δ 7.65 (d, *J* = 8.4 Hz, 1H), 7.36 (dd, *J* = 8.4, 2.1 Hz, 1H), 7.12 (d, *J* = 2.1 Hz, 1H), 5.50 (t, *J* = 7.5 Hz, 1H), 2.90 (d, *J* = 4.8 Hz, 3H), 2.32 (q, *J* = 7.5 Hz, 2H), 2.20 (quint, *J* = 7.5 Hz, 2H), 1.05 (t, *J* = 7.5 Hz, 3H), 0.85 (t, *J* = 7.5 Hz, 3H); $^{13}C$ NMR (100 MHz, $CDCl_3$): δ 168.7, 143.5, 141.8, 135.8, 133.0, 132.3, 130.4, 130.0, 127.2, 26.7, 24.8, 21.4, 14.3, 13.0; HRMS [(EI), $(M^+)$]: 251.1075 (cal. for $C_{14}H_{18}NOCl$ 251.1077); New compound.

**(*E*)-2-(1,2-Bis(4-methoxyphenyl)vinyl)-*N*-methyl-5-(trifluoromethyl)benzamide (4r):**



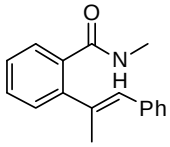
Work-up and purification by column chromatography, brown liquid (128 mg, 58%); IR (KBr): 3446, 1652, 1508, 744 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.79 (s, 1H), 7.65 (d, *J* = 7.2 Hz, 1H), 7.50 (d, *J* = 8.4 Hz, 1H), 7.09 (d, *J* = 8.4 Hz, 2H), 7.03 (d, *J* = 8.4 Hz, 2H), 6.77 (d, *J* = 9 Hz, 2H), 6.73 (d, *J* = 8.4 Hz, 2H), 6.68 (s, 1H), 5.74 (s, 1H), 3.79 (s, 3H), 3.78 (s, 3H), 2.65 (d, *J* = 4.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 168.6, 159.1, 158.9, 146.3, 138.3, 136.9, 131.6, 131.3, 130.7, 130.6, 130.4, 129.7 (q, $J_{C-F}$ = 33 Hz), 129.3, 126.4, 125.6, 123.7 (q, $J_{C-F}$ = 270 Hz), 114 (q, $J_{C-F}$ = 14 Hz), 113.7, 113.6, 55.1, 26.7; HRMS [(ESI), $(M+Na)^+$]: 464.1453 (cal. for $C_{25}H_{22}F_3NO_3Na$ 464.1449); New compound.

**(*E*)-*N*-Methyl-2-(1-phenylprop-1-en-1-yl)benzamide (4s):**



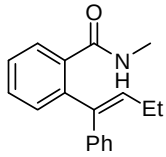
Work-up and purification by column chromatography, colorless viscous oil (42 mg, 34%); IR (KBr): 3445, 1635, 1507, 741 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.48 (d, *J* = 7.8 Hz, 1H), 7.38 (t, *J* = 7.8 Hz, 1H), 7.32–7.28 (m, 4H), 7.22 (t, *J* = 7.2 Hz, 1H), 7.12 (d, *J* = 7.2 Hz, 2H), 5.98 (q, *J* = 7.2 Hz, 1H), 5.63 (s, 1H), 2.63 (d, *J* = 4.8 Hz, 3H), 1.92 (d, *J* = 6.6 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.1, 141.9, 141.7, 139.1, 136.1, 130.8, 129.8, 129.7, 128.2, 127.8, 127.3, 127.2, 127.0, 26.5, 15.8; HRMS [(ESI), $(M+Na)^+$]: 274.1205 (cal. for $C_{17}H_{17}NONa$ 274.1207); New compound.

**(*E*)-*N*-Methyl-2-(1-phenylprop-1-en-2-yl)benzamide (4s'):**



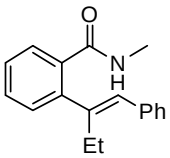
Work-up and purification by column chromatography, white solid (19 mg, 15%), mp: 149–150 °C; IR (KBr): 3501, 1635, 1456, 748 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.68 (d, *J* = 7.8 Hz, 1H), 7.44–7.26 (m, 8H), 6.59 (s, 1H), 6.10 (s, 1H), 2.95 (d, *J* = 4.8 Hz, 3H), 2.21 (d, *J* = 1.2 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.9, 143.6, 139.0, 137.4, 134.4, 130.3, 130.1, 128.9, 128.9, 128.7, 128.4, 127.4, 126.9, 26.9, 20.0; HRMS [(ESI), $(M+Na)^+$]: 274.1208 (cal. for $C_{17}H_{17}NONa$ 274.1207); New compound.

**(*E*)-*N*-Methyl-2-(1-phenylbut-1-en-1-yl)benzamide (4t):**



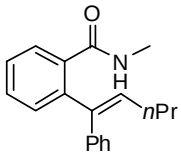
Work-up and purification by column chromatography, white solid (38 mg, 28%), mp: 128–129 °C; IR (KBr): 3520, 1646, 1540, 759 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.50 (d, *J* = 7.8 Hz, 1H), 7.39–7.36 (m, 1H), 7.31–7.28 (m, 4H), 7.22 (t, *J* = 7.2 Hz, 1H), 7.12 (d, *J* = 7.2 Hz, 2H), 5.85 (t, *J* = 7.2 Hz, 1H), 5.68 (s, 1H), 2.66 (d, *J* = 5.4 Hz, 3H), 2.32 (quint, *J* = 7.8 Hz, 2H), 1.07 (t, *J* = 7.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.0, 141.8, 140.2, 139.4, 136.1, 134.9, 130.8, 129.7, 129.6, 128.2, 127.8, 127.3, 127.1, 26.5, 23.0, 14.5; HRMS [(ESI), $(M+Na)^+$]: 288.1364 (cal. for $C_{18}H_{19}N_1O_1Na$ 288.1363); New compound.

**(*E*)-*N*-Methyl-2-(1-phenylbut-1-en-2-yl)benzamide (4t'):**



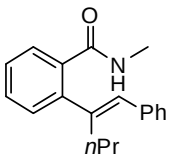
Work-up and purification by column chromatography, white solid (24 mg, 18%), mp: 93–94 °C; IR (KBr): 3419, 1646, 1521, 760 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.73 (d, *J* = 7.2 Hz, 1H), 7.44–7.35 (m, 4H), 7.33 (d, *J* = 7.2 Hz, 2H), 7.28 (t, *J* = 7.2 Hz, 2H), 6.54 (s, 1H), 6.19 (s, 1H), 2.93 (d, *J* = 4.8 Hz, 3H), 2.62 (q, *J* = 7.8 Hz, 2H), 0.93 (t, *J* = 7.8 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.8, 145.7, 141.5, 137.2, 134.5, 130.1, 129.9, 129.6, 128.9, 128.6, 128.4, 127.5, 126.9, 26.9, 25.6, 12.9; HRMS [(ESI), $(M+Na)^+$]: 288.1364 (cal. for $C_{18}H_{19}N_1O_1Na$ 288.1363); Registry Number: [1427042-14-5].

**(*E*)-*N*-Methyl-2-(1-phenylpent-1-en-1-yl)benzamide (4u):**



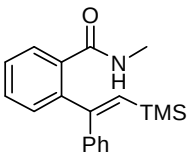
Work-up and purification by column chromatography, white solid (43 mg, 31%), mp: 78–79 °C; IR (KBr): 3565, 1698, 1540, 1264, 748 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.49 (d, *J* = 7.2 Hz, 1H), 7.38 (t, *J* = 7.8 Hz, 1H), 7.31–7.28 (m, 4H), 7.22 (t, *J* = 7.2 Hz, 1H), 7.12 (d, *J* = 7.2 Hz, 2H), 5.87 (t, *J* = 7.2 Hz, 1H), 5.70 (s, 1H), 2.66 (d, *J* = 4.8 Hz, 3H), 2.29 (q, *J* = 7.8 Hz, 2H), 1.49 (sext, *J* = 7.2 Hz, 2H), 0.94 (t, *J* = 7.2 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.0, 141.9, 140.8, 139.5, 136.1, 133.3, 130.8, 129.7, 129.7, 128.3, 127.8, 127.2, 127.0, 31.7, 26.5, 23.2, 13.8; HRMS [(ESI), $(M+Na)^+$]: 302.1512 (cal. for $C_{19}H_{21}NONa$ 302.1520); New compound.

**(*E*)-*N*-Methyl-2-(1-phenylpent-1-en-2-yl)benzamide (4u'):**



Work-up and purification by column chromatography, white solid (28 mg, 20%), mp: 92–93 °C; IR (KBr): 3446, 1683, 1558, 1265, 743 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.73 (d, *J* = 7.2 Hz, 1H), 7.44–7.35 (m, 4H), 7.33–7.27 (m, 4H), 6.57 (s, 1H), 6.19 (s, 1H), 2.95 (d, *J* = 1.8 Hz, 3H), 2.56–2.53 (m, 2H), 1.31 (sext, *J* = 7.8 Hz, 2H), 0.82 (t, *J* = 7.2 Hz, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.8, 144.6, 141.9, 137.2, 134.3, 130.1, 130.1, 129.7, 128.8, 128.6, 128.4, 127.5, 126.9, 34.5, 26.9, 21.7, 14.0; HRMS [(ESI), $(M+Na)^+$]: 302.1518 (cal. for $C_{19}H_{21}NONa$ 302.1520); New compound.

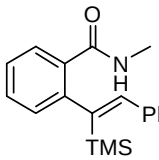
**(*E*)-*N*-Methyl-2-(1-phenyl-2-(trimethylsilyl)vinyl)benzamide (4v):**



Work-up and purification by column chromatography, white solid (12 mg, 8%), mp: 95–96 °C; IR (KBr): 3445, 1698, 1558, 749 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.47 (dd, *J* = 7.2, 1.2 Hz, 1H), 7.36 (td, *J* = 7.5, 1.2 Hz, 1H), 7.31–7.26 (m, 5H), 7.18–7.17 (m, 2H), 6.01 (s, 1H), 5.68 (s, 1H), 2.70 (d, *J* = 5.4 Hz, 3H), -0.03 (s, 9H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 170.0, 157.5, 143.5, 142.1, 135.7,

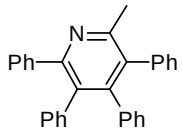
134.3, 130.2, 129.6, 129.3, 128.0, 127.8, 127.7, 127.6, 26.5, 0.1; HRMS [(ESI), $(M+Na)^+$]: 332.1444 (cal. for $C_{19}H_{23}NOSiNa$ 332.1446); New compound.

**(Z)-*N*-Methyl-2-(2-phenyl-1-(trimethylsilyl)vinyl)benzamide (4v'):**




Work-up and purification by column chromatography, white solid (85 mg, 55%), mp: 101–102 °C; IR (KBr): 3419, 1683, 1540, 749 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.84 (dd, *J* = 7.8, 1.2 Hz, 1H), 7.42–7.37 (m, 3H), 7.34–7.30 (m, 5H), 7.09 (dd, *J* = 7.2, 0.6 Hz, 1H), 6.40 (s, 1H), 2.98 (d, *J* = 4.8 Hz, 3H), -0.10 (s, 9H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 169.3, 148.9, 145.1, 144.6, 139.0, 133.2, 130.3, 129.0, 128.9, 128.3, 128.2, 127.8, 126.4, 26.7, 0.3; HRMS [(ESI), $(M+Na)^+$]: 332.1444 (cal. for $C_{19}H_{23}NOSiNa$ 332.1446); New compound.

**2-Methyl-3,4,5,6-tetraphenylpyridine (5):**




Work-up and purification by column chromatography, white solid (119 mg, 60%), mp: 157–158 °C; IR (KBr): 2957, 1537, 1398, 1028, 698 $cm^{-1}$; $^1H$ NMR (600 MHz, $CDCl_3$): δ 7.34–7.33 (m, 2H), 7.21–7.18 (m, 5H), 7.16–7.15 (m, 1H), 7.06–7.05 (m, 2H), 6.97–6.95 (m, 3H), 6.90–6.88 (m, 3H), 6.85–6.83 (m, 2H), 6.74–6.72 (m, 2H), 2.48 (s, 3H); $^{13}C$ NMR (150 MHz, $CDCl_3$): δ 156.1, 155.3, 149.3, 140.9, 138.8, 138.4, 138.1, 134.7, 132.6 131.4, 130.2, 130.0, 129.9, 127.8, 127.6, 127.3, 127.2, 126.9, 126.6, 126.1, 24.3; HRMS [(ESI), $(M+H)^+$]: 398.1906 (cal. for $C_{30}H_{24}N$ 398.1909); Registry Number: [41728-97-6].

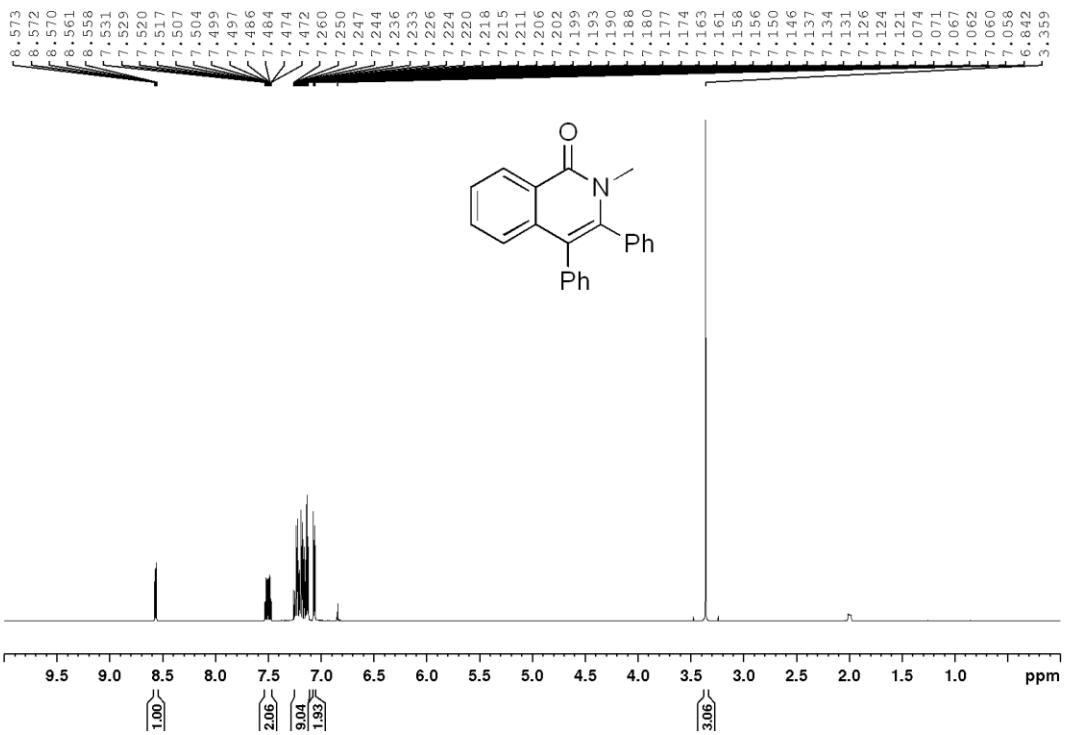
## References

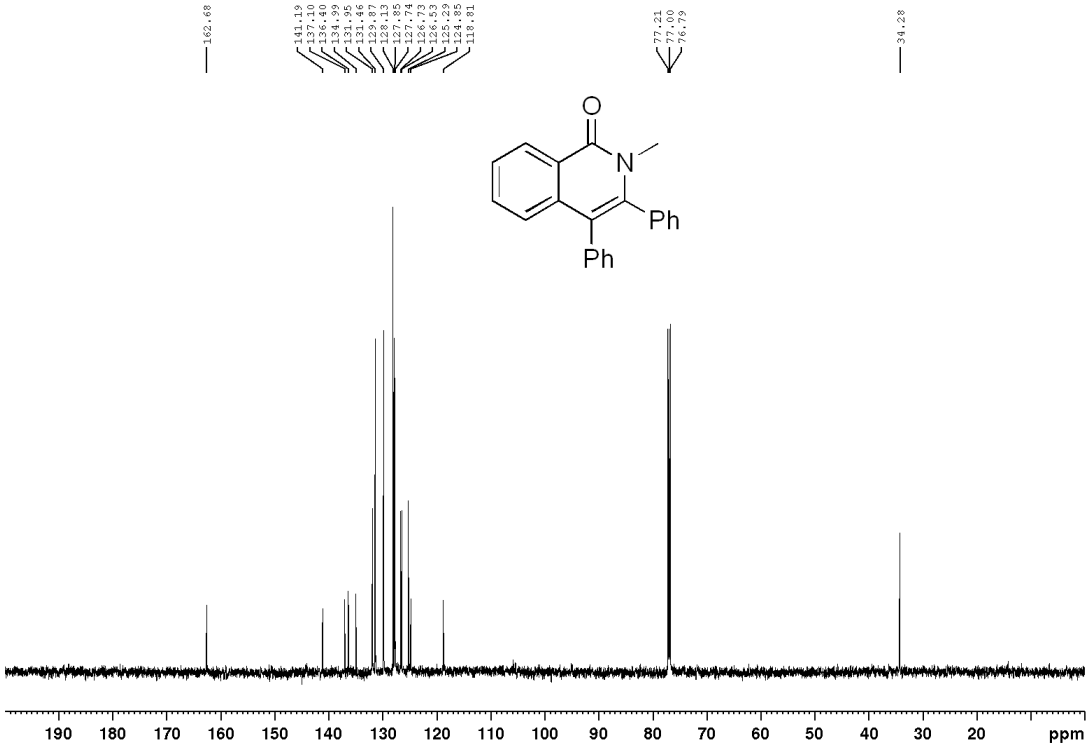
(1) Abe T.; Takahashi Y.; Matsubara Y.; Yamada K. *Org. Chem. Front.* **2017**, *4*, 2124.

(2) Shu, Z.; Guo, Y.; Li, W.; Wang, B. *Catal. Today* **2017**, *297*, 292.

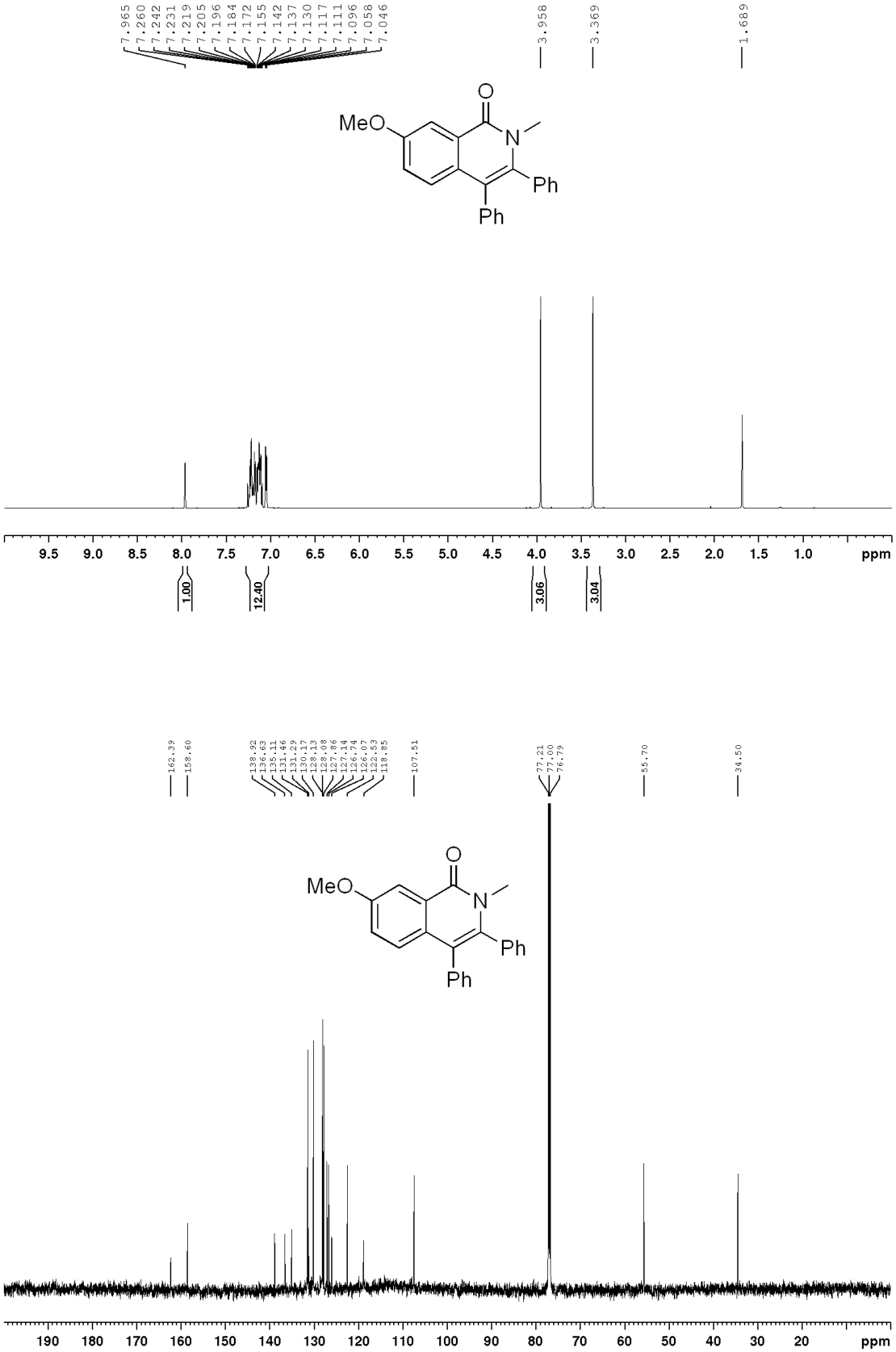
# $^{1}H$ and $^{13}C$ NMR Spectra for Products ($CDCl_3$)

**2-Methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3a) (600 MHz)**

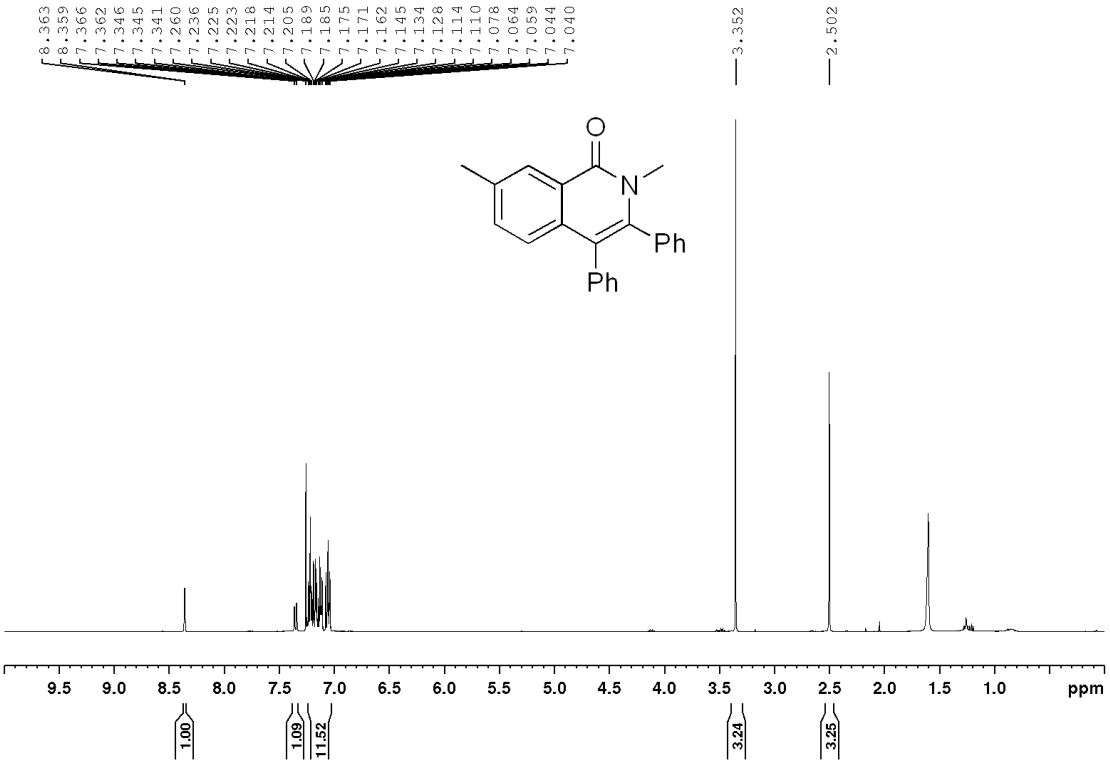


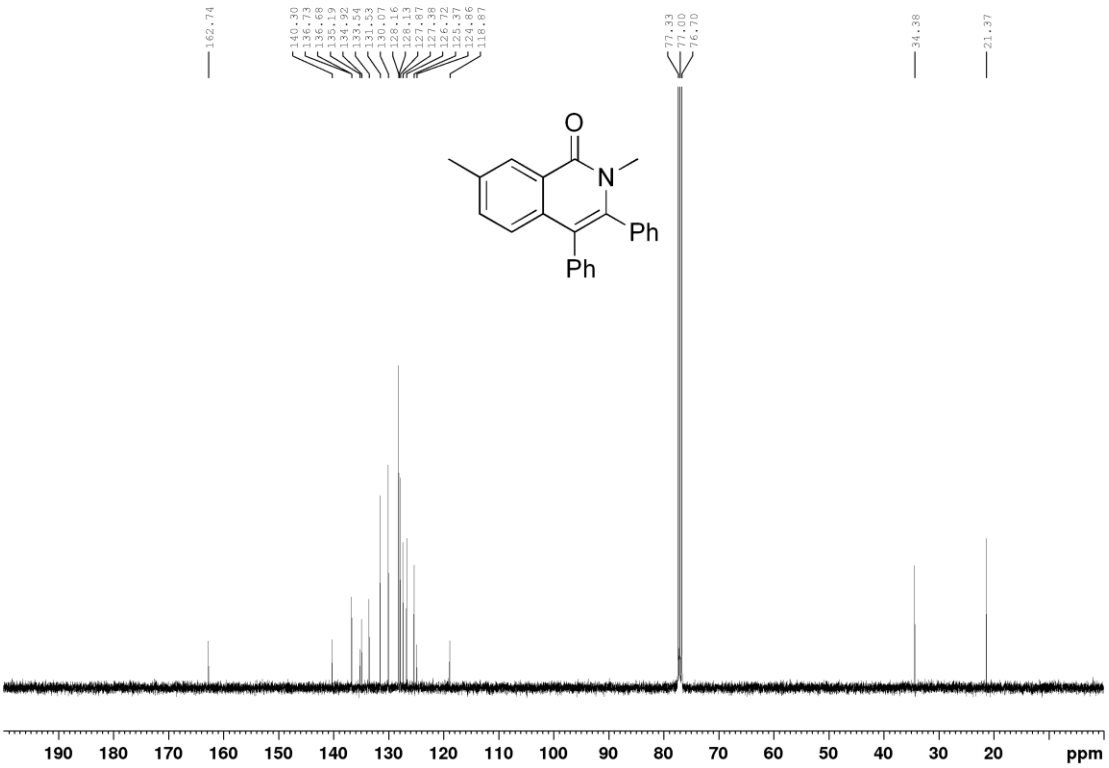



**7-Methoxy-2-methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3b) (600 MHz)**

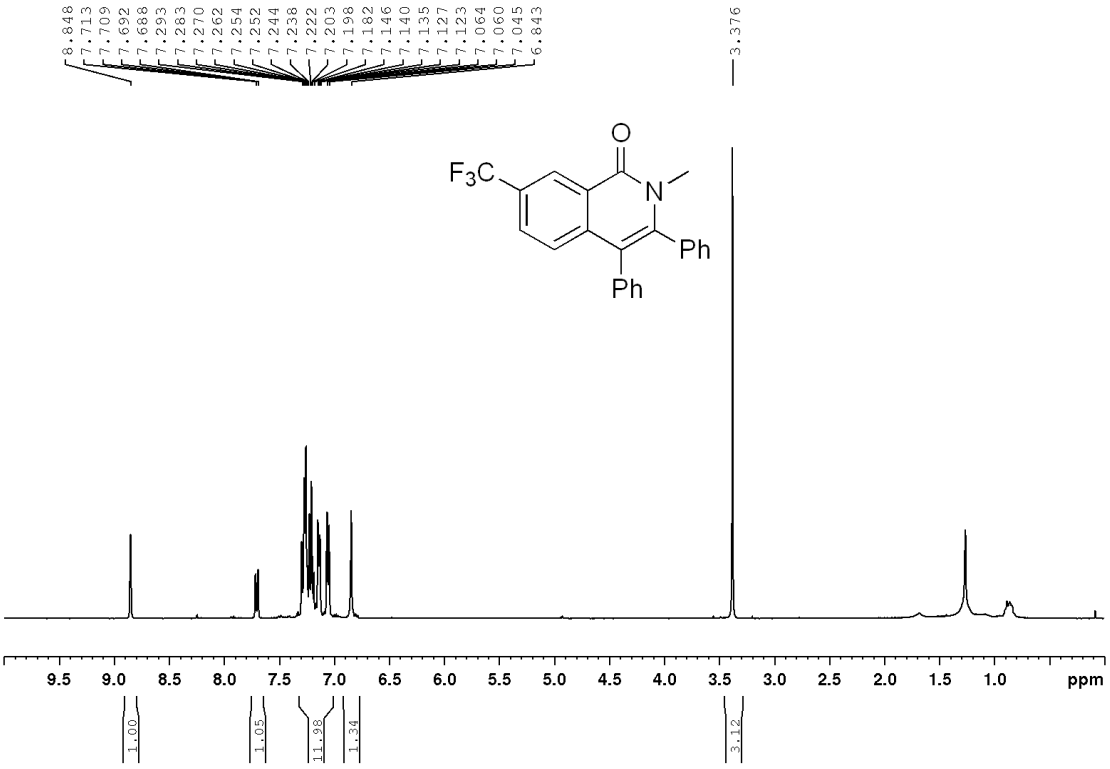


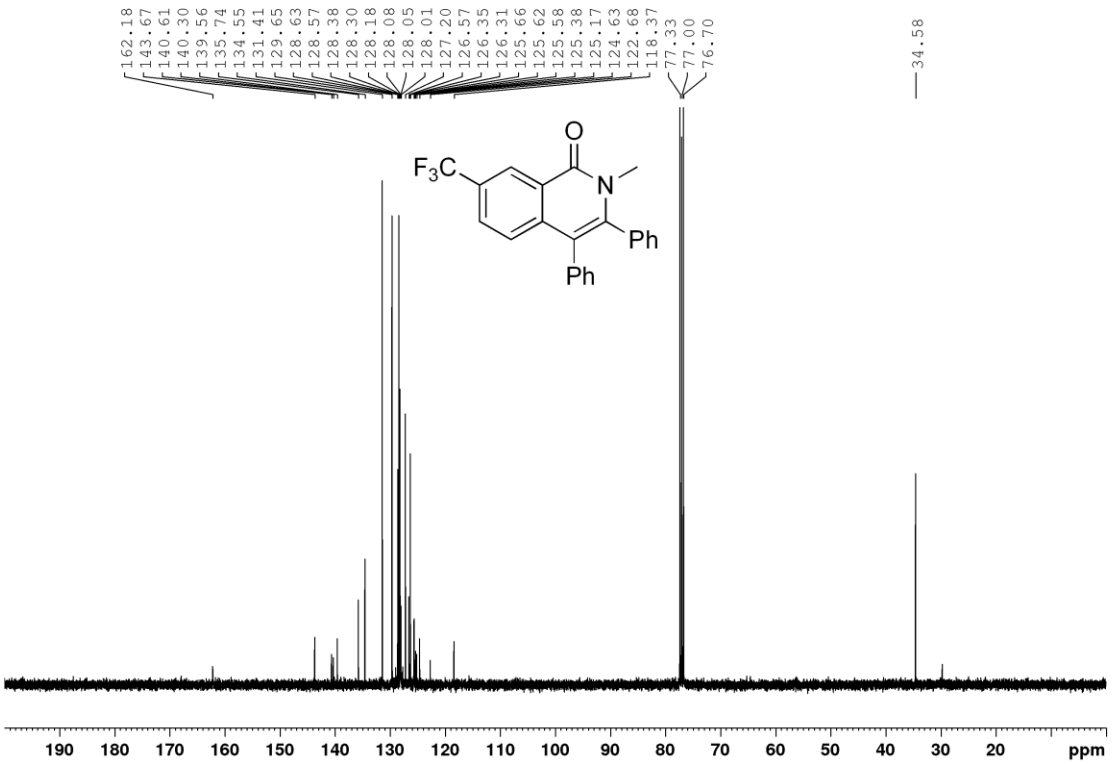
**2,7-Dimethyl-3,4-diphenylisoquinolin-1(2*H*)-one (3c) (400 MHz)**



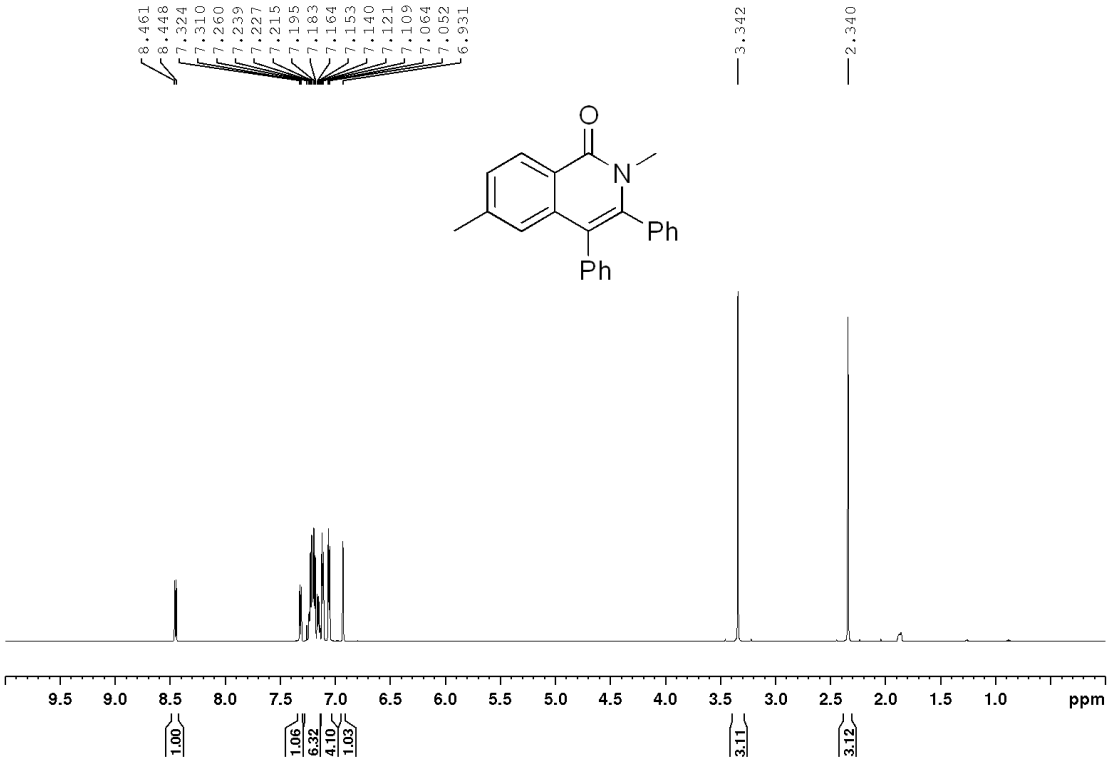



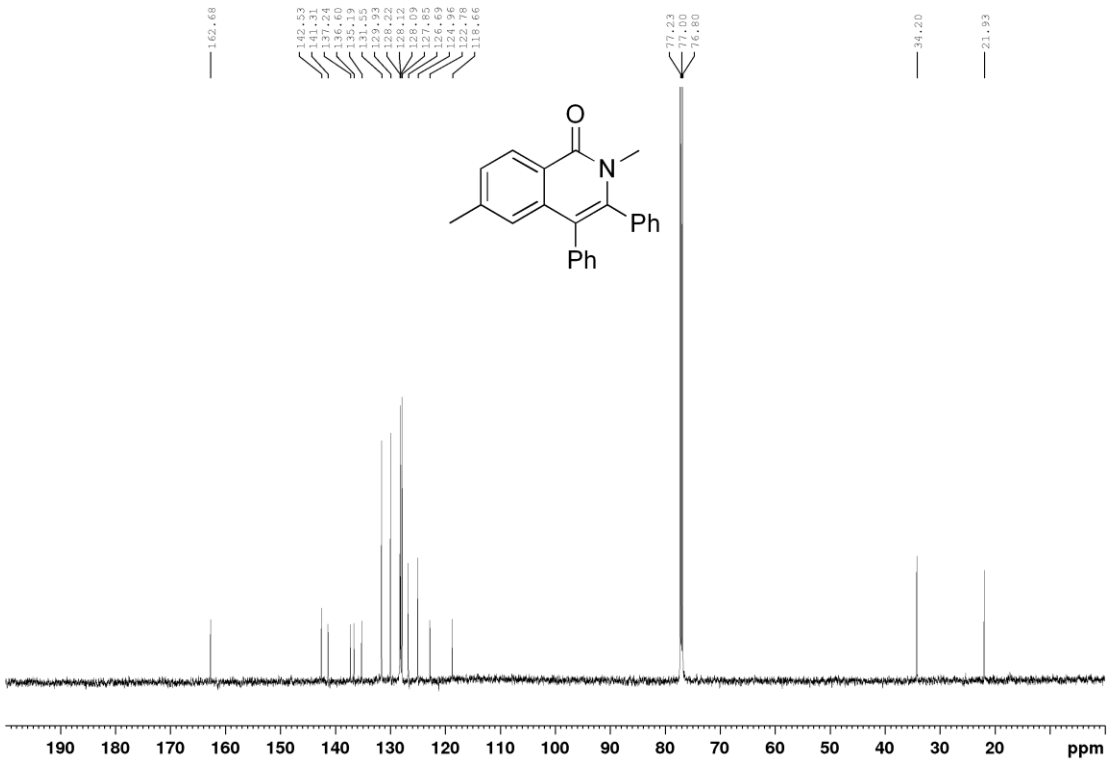
**2-Methyl-3,4-diphenyl-7-(trifluoromethyl)isoquinolin-1(2*H*)-one (3d) (400 MHz)**



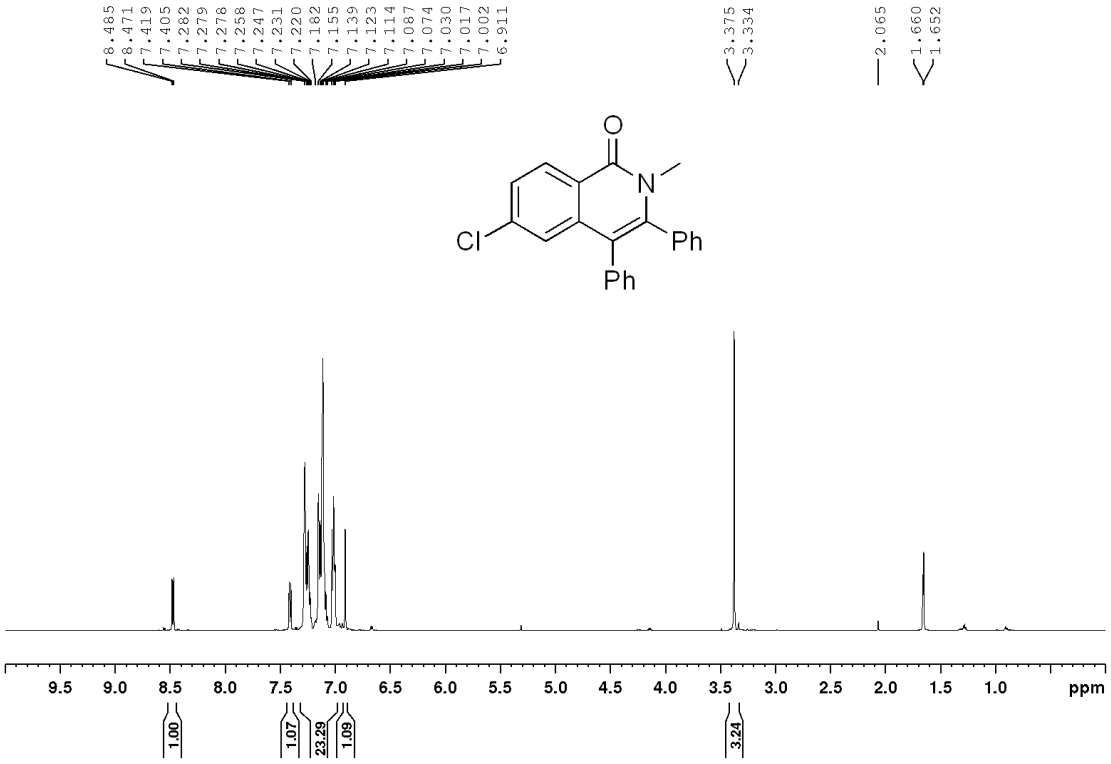



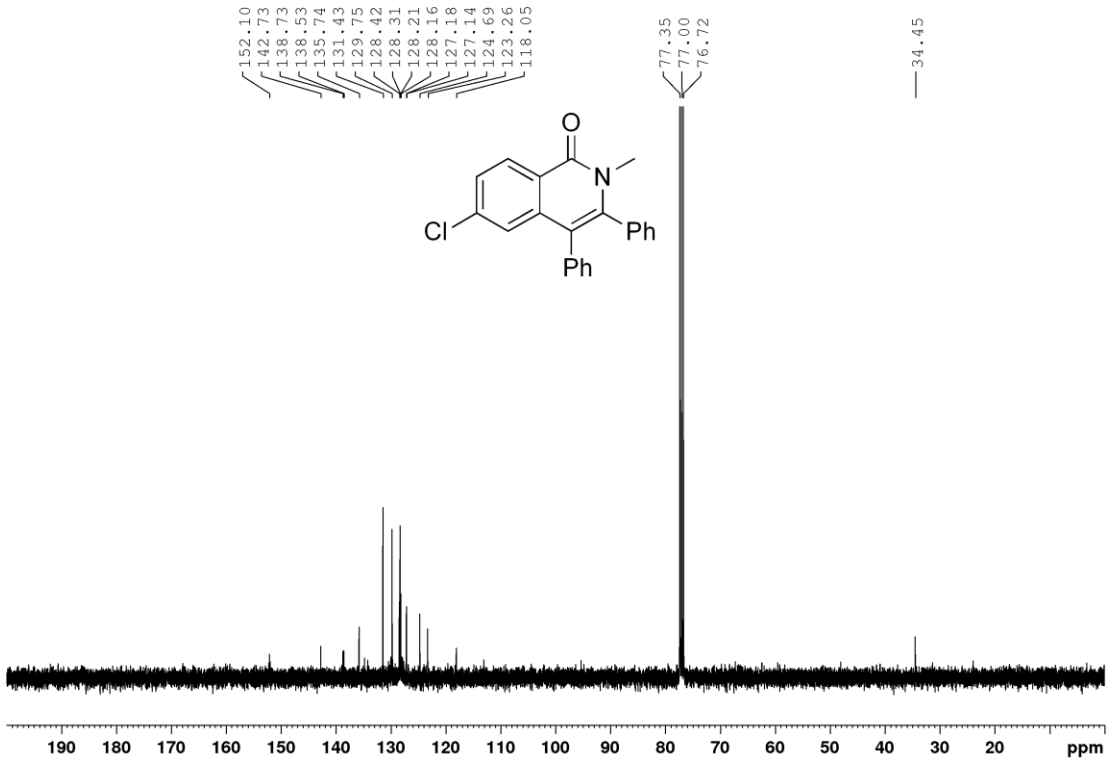
**2,6-Dimethyl-3,4-diphenylisoquinolin-1(2*H*)-one (3e) (600 MHz)**



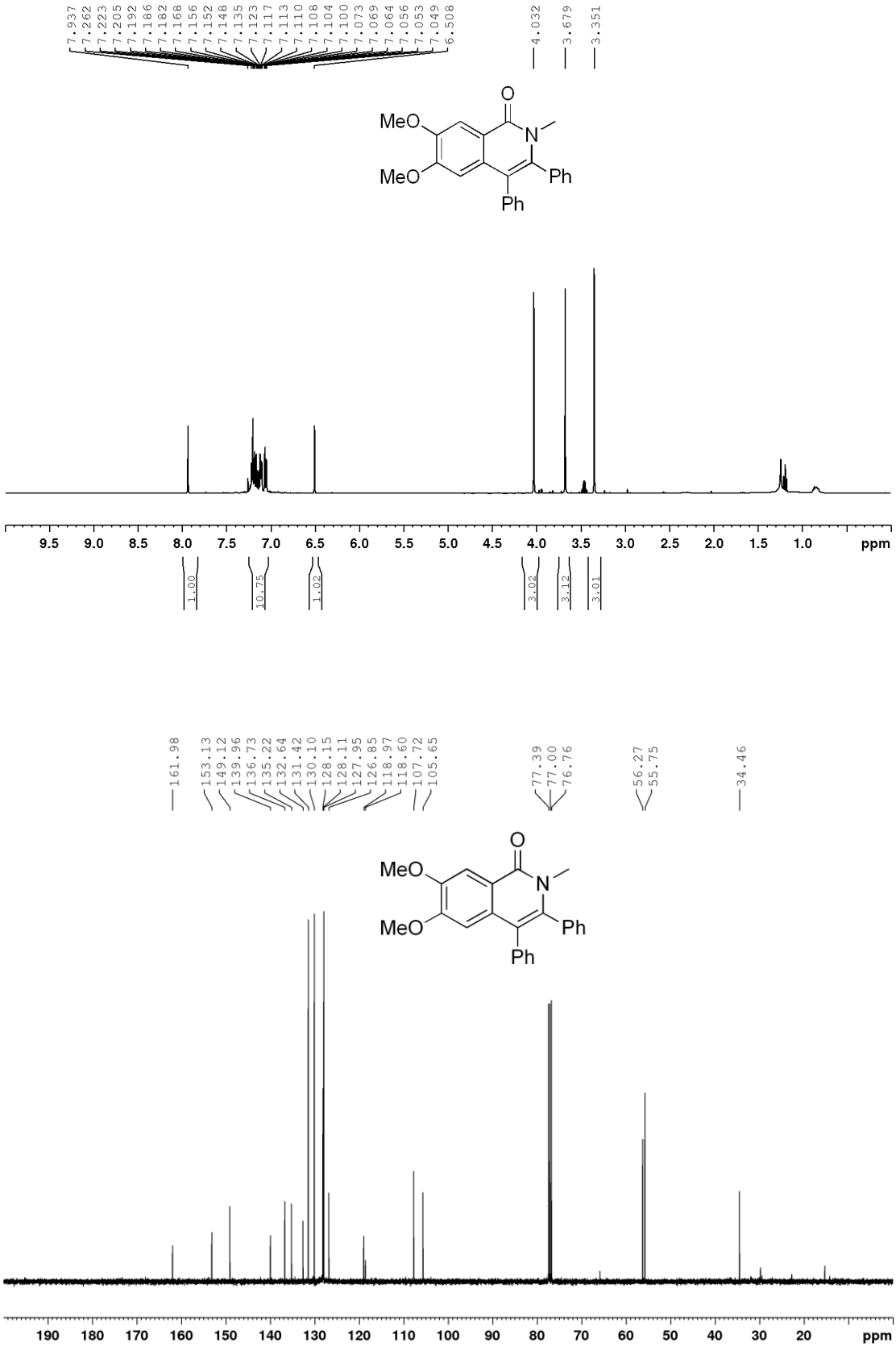



**6-Chloro-2-methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3f) (600 MHz for $^{1}$H, 100 MHz $^{13}$C)**

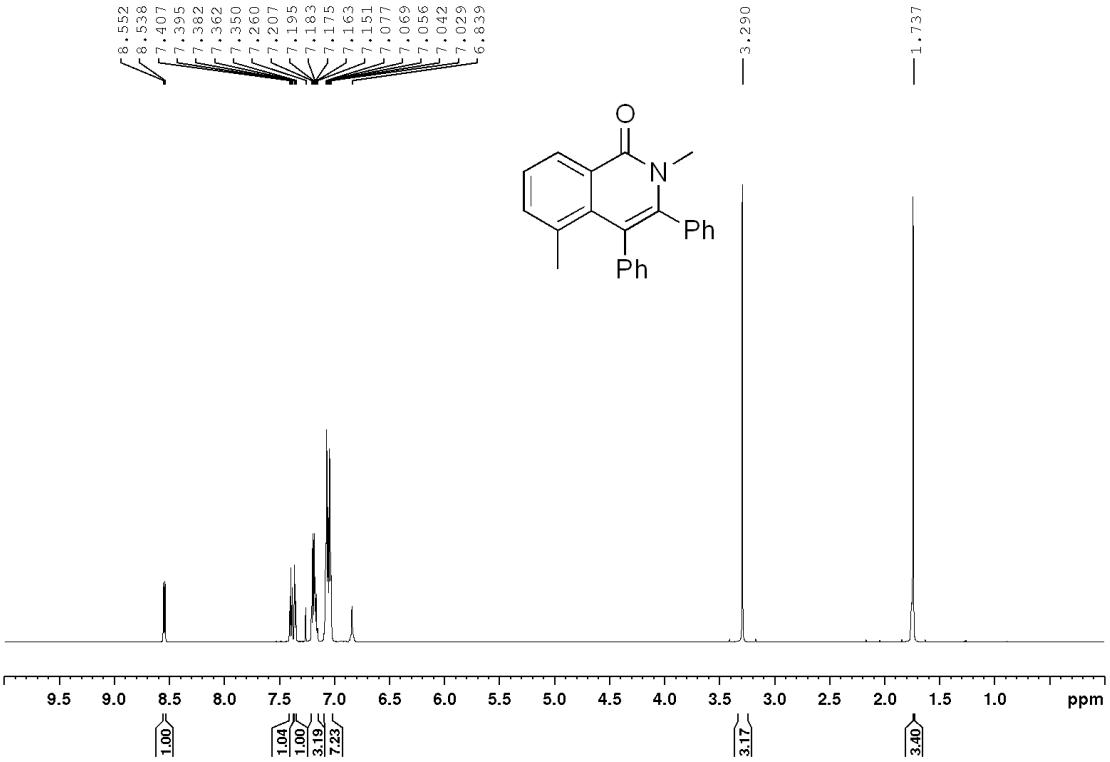


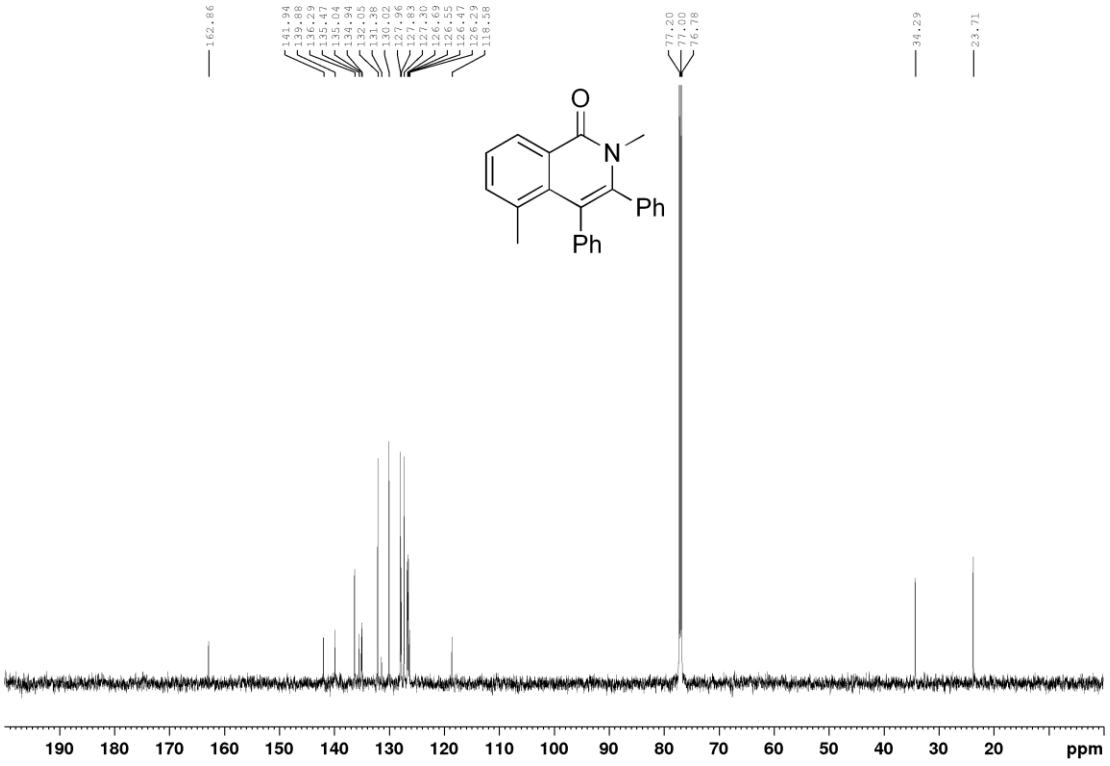



**6,7-Dimethoxy-2-methyl-3,4-diphenylisoquinolin-1(2*H*)-one (3g) (400 MHz)**

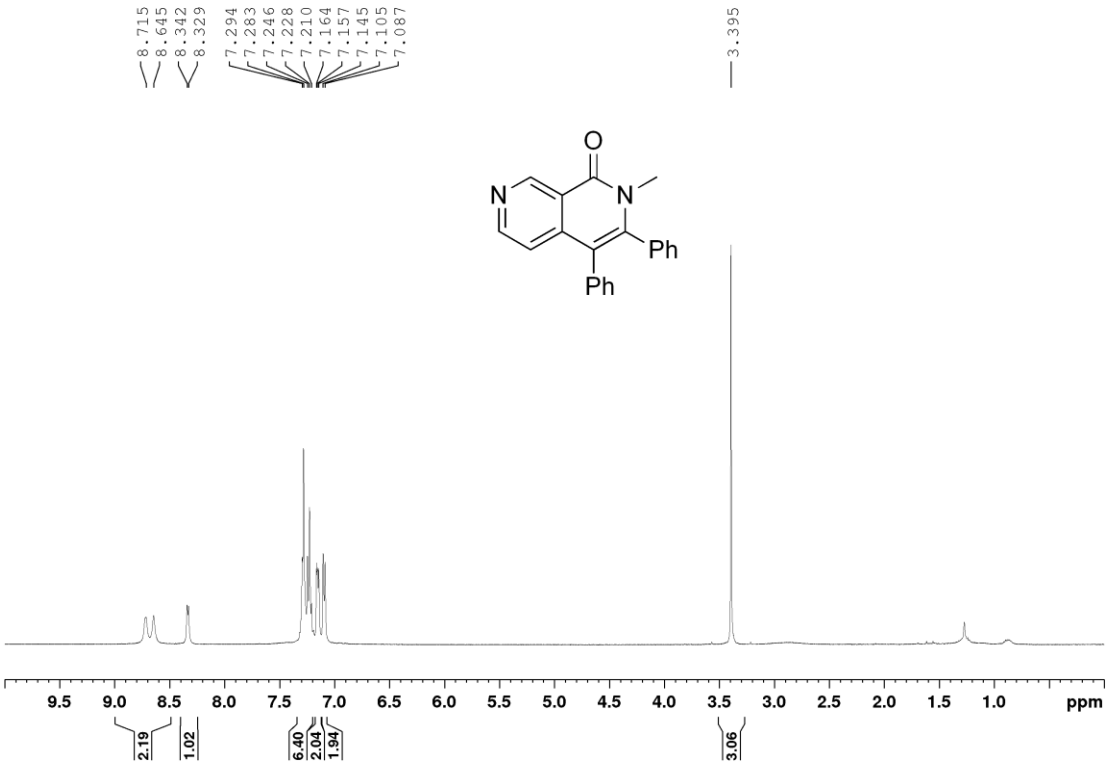


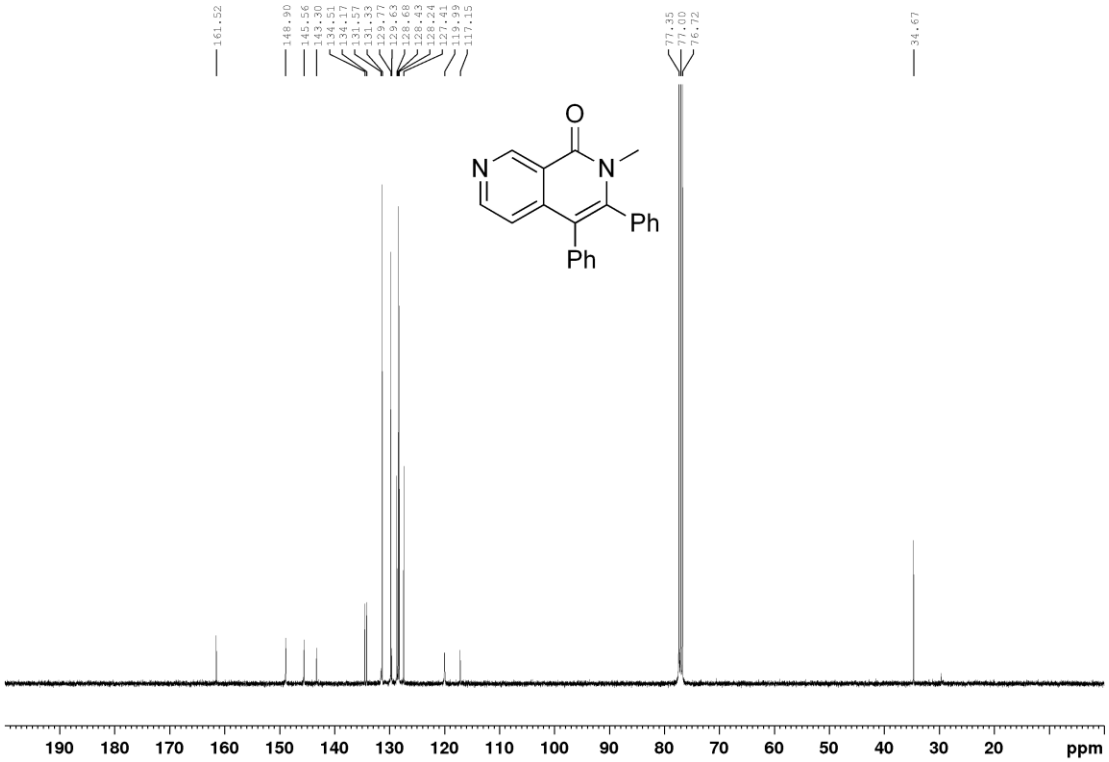
**2,5-Dimethyl-3,4-diphenylisoquinolin-1(2*H*)-one (3h) (600 MHz)**



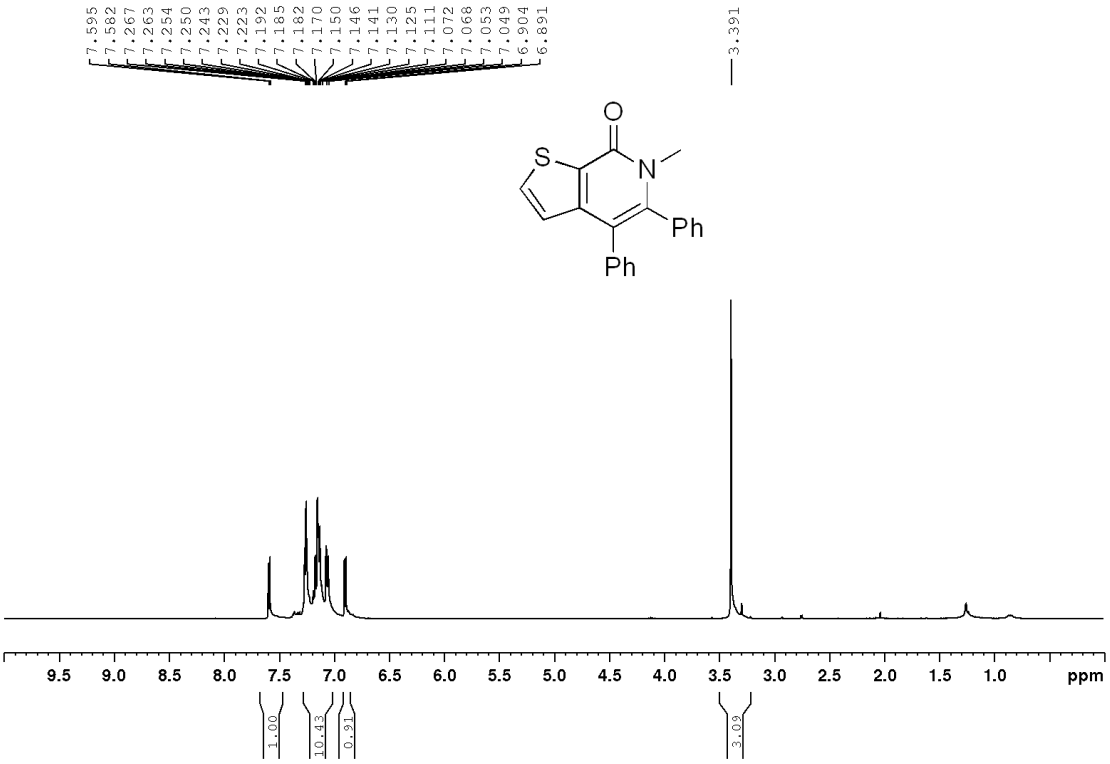



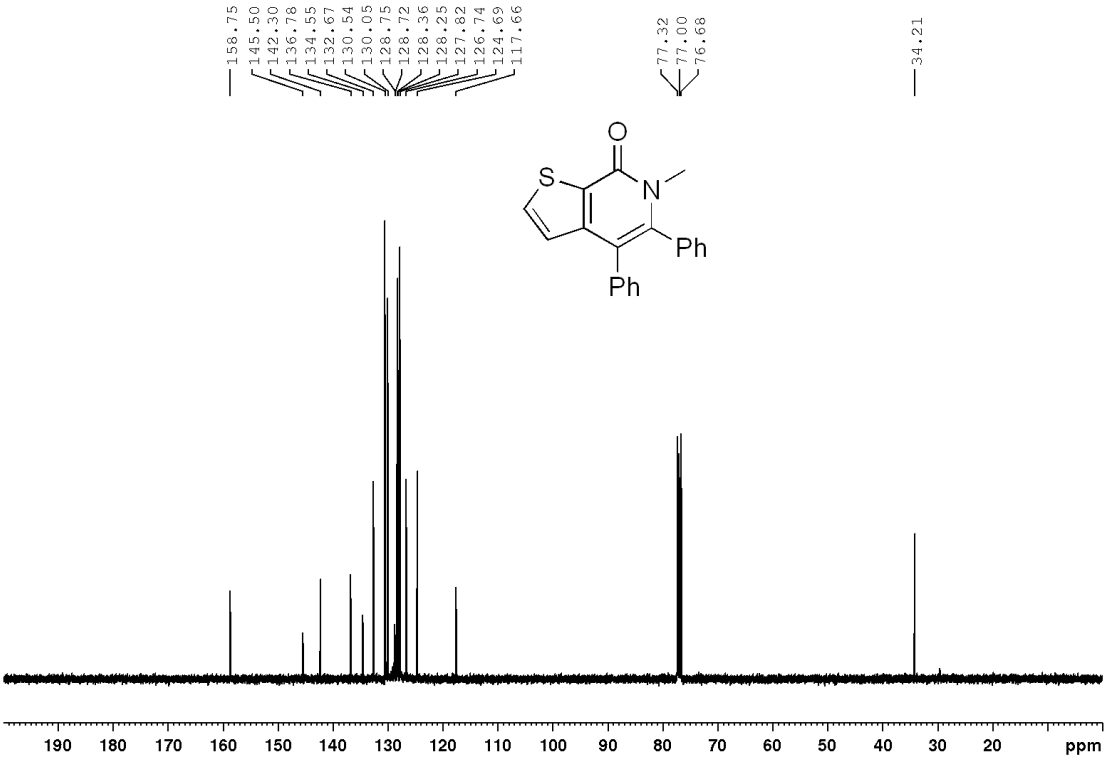
**2-Methyl-3,4-diphenyl-2,7-naphthyridin-1(2*H*)-one (3i) (400 MHz)**



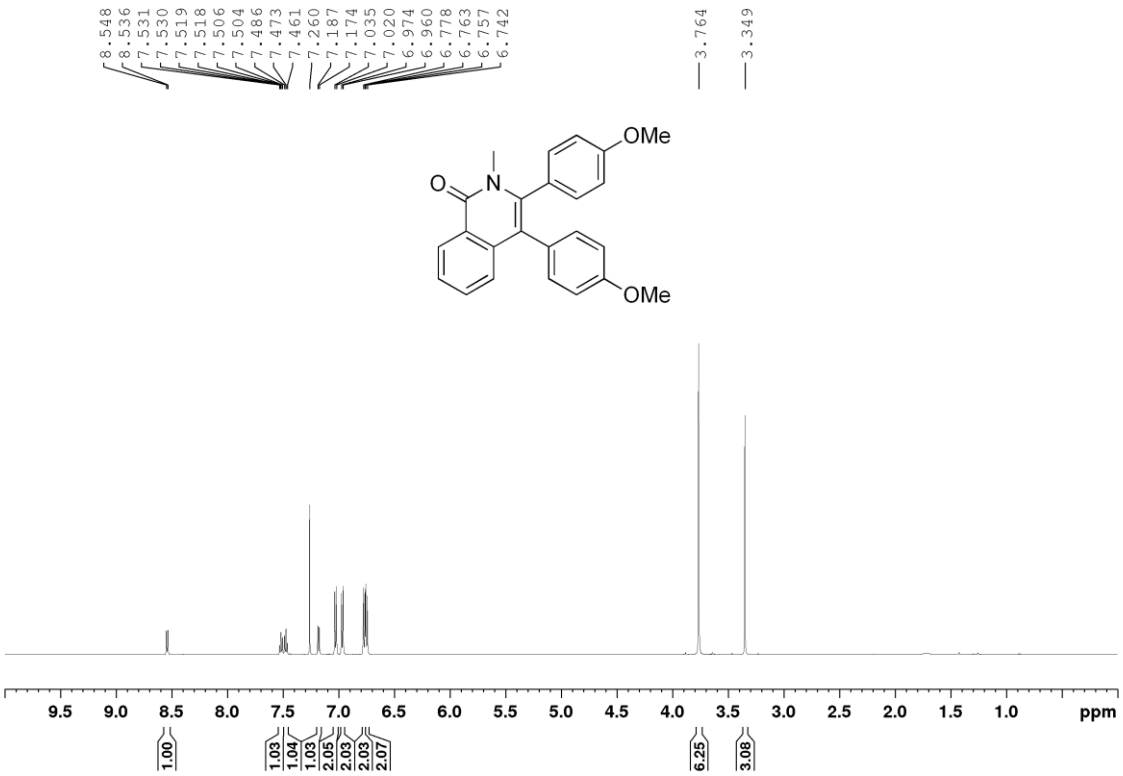



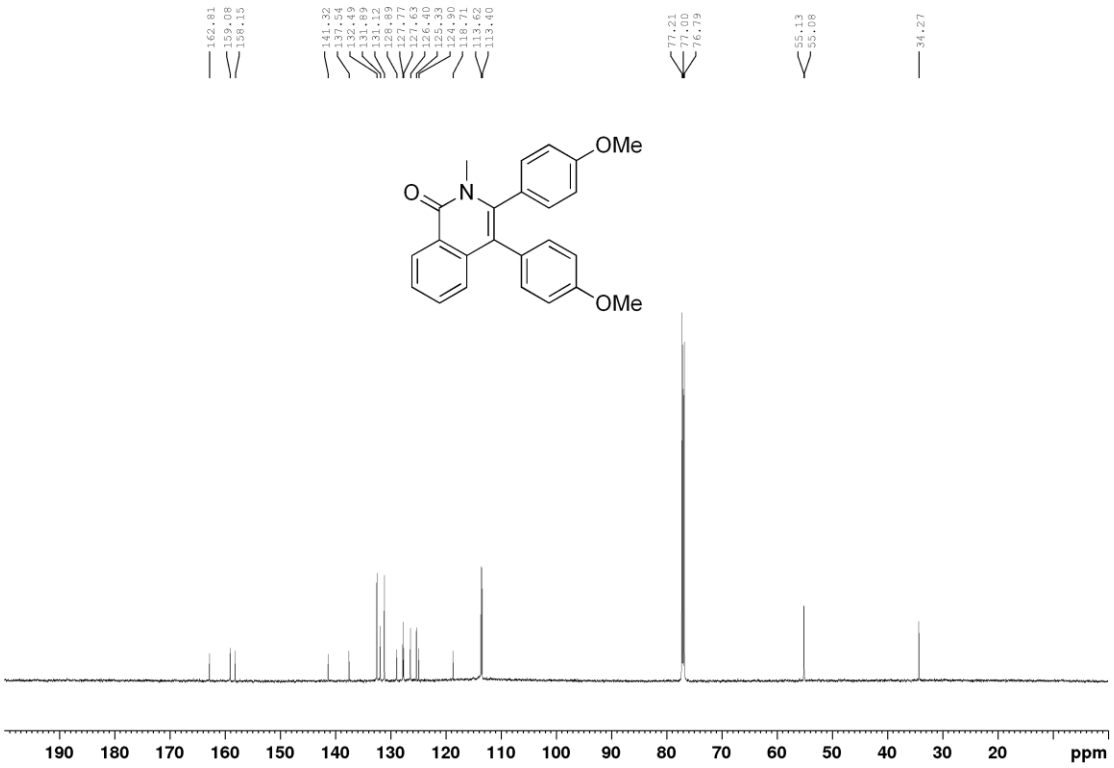
**6-Methyl-4,5-diphenylthieno[2,3-*c*]pyridin-7(6*H*)-one (3j) (400 MHz)**



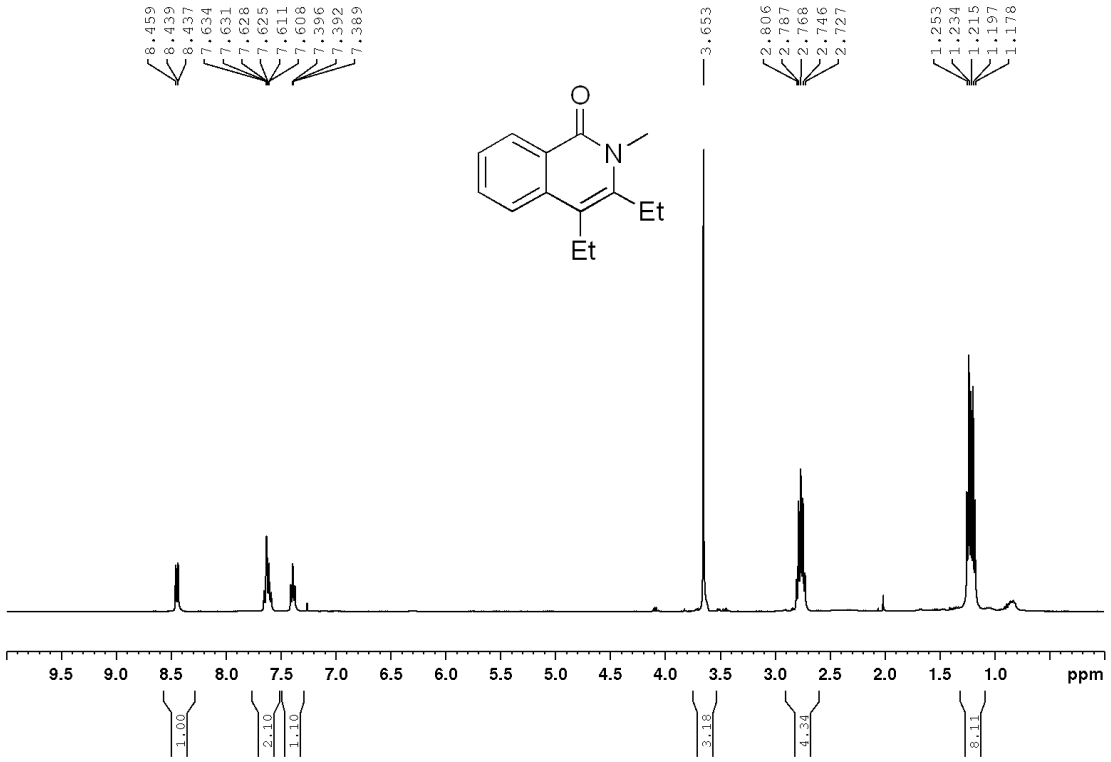



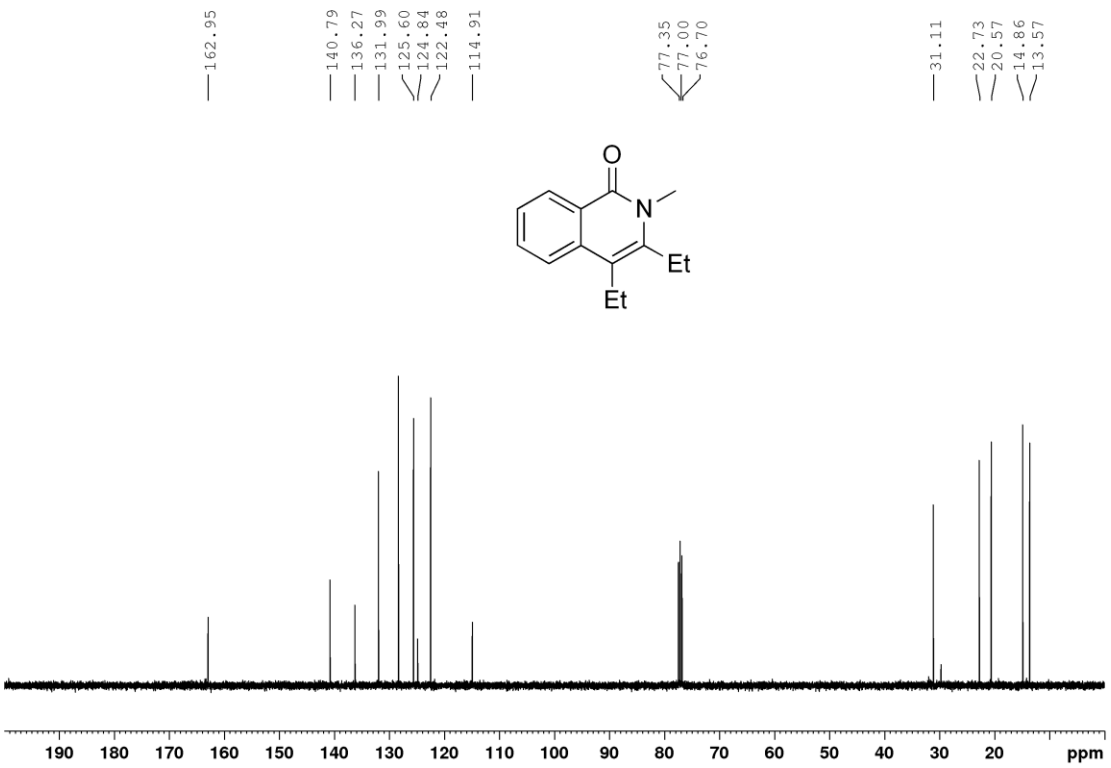
**3,4-Bis(4-methoxyphenyl)-2-methylisoquinolin-1(2*H*)-one (3k) (600 MHz)**



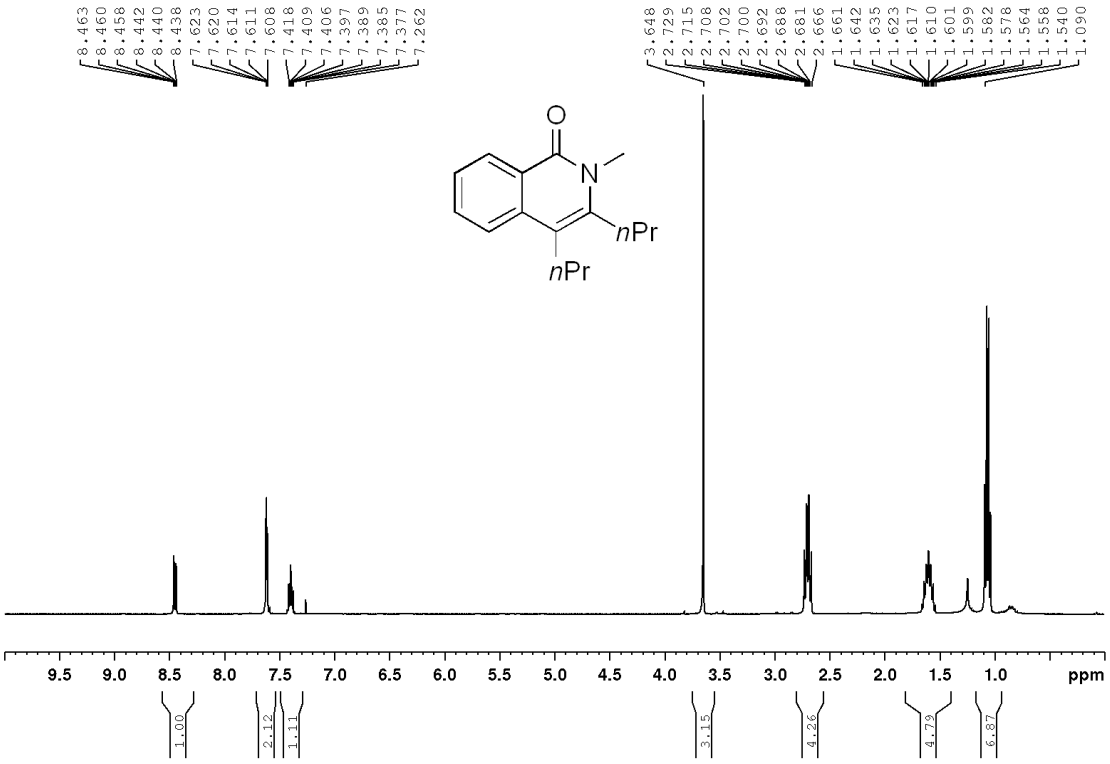



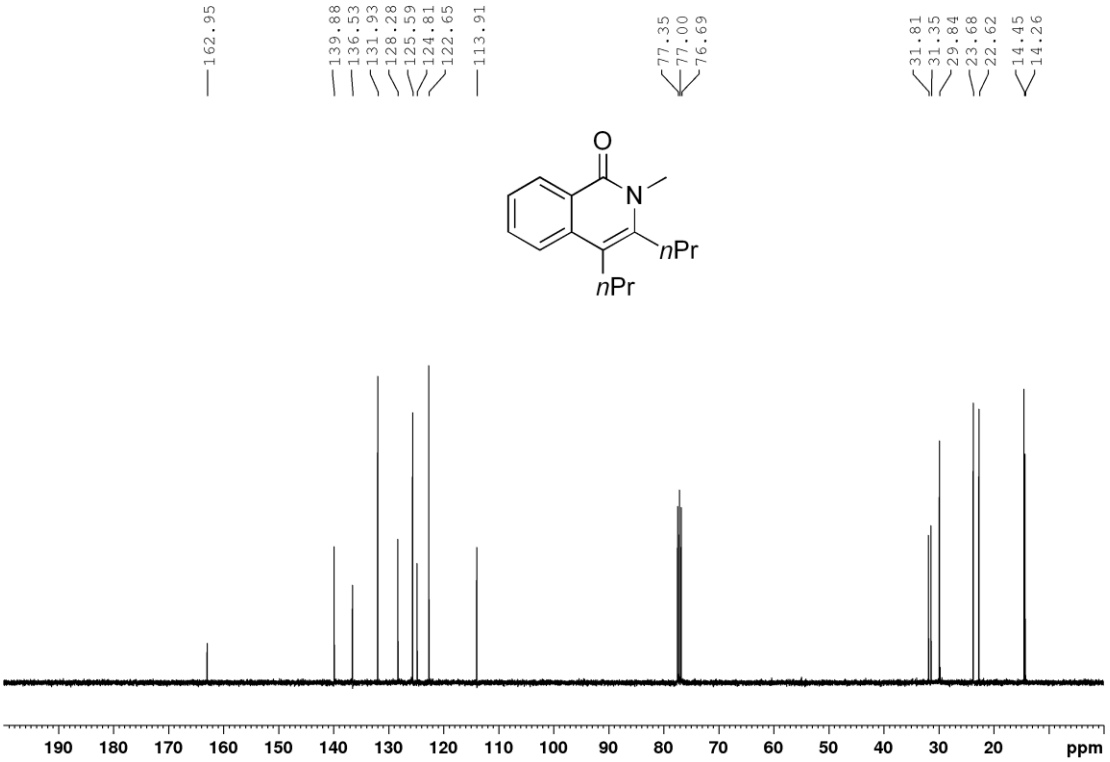
**3,4-Diethyl-2-methylisoquinolin-1(2*H*)-one (3l) (400 MHz)**



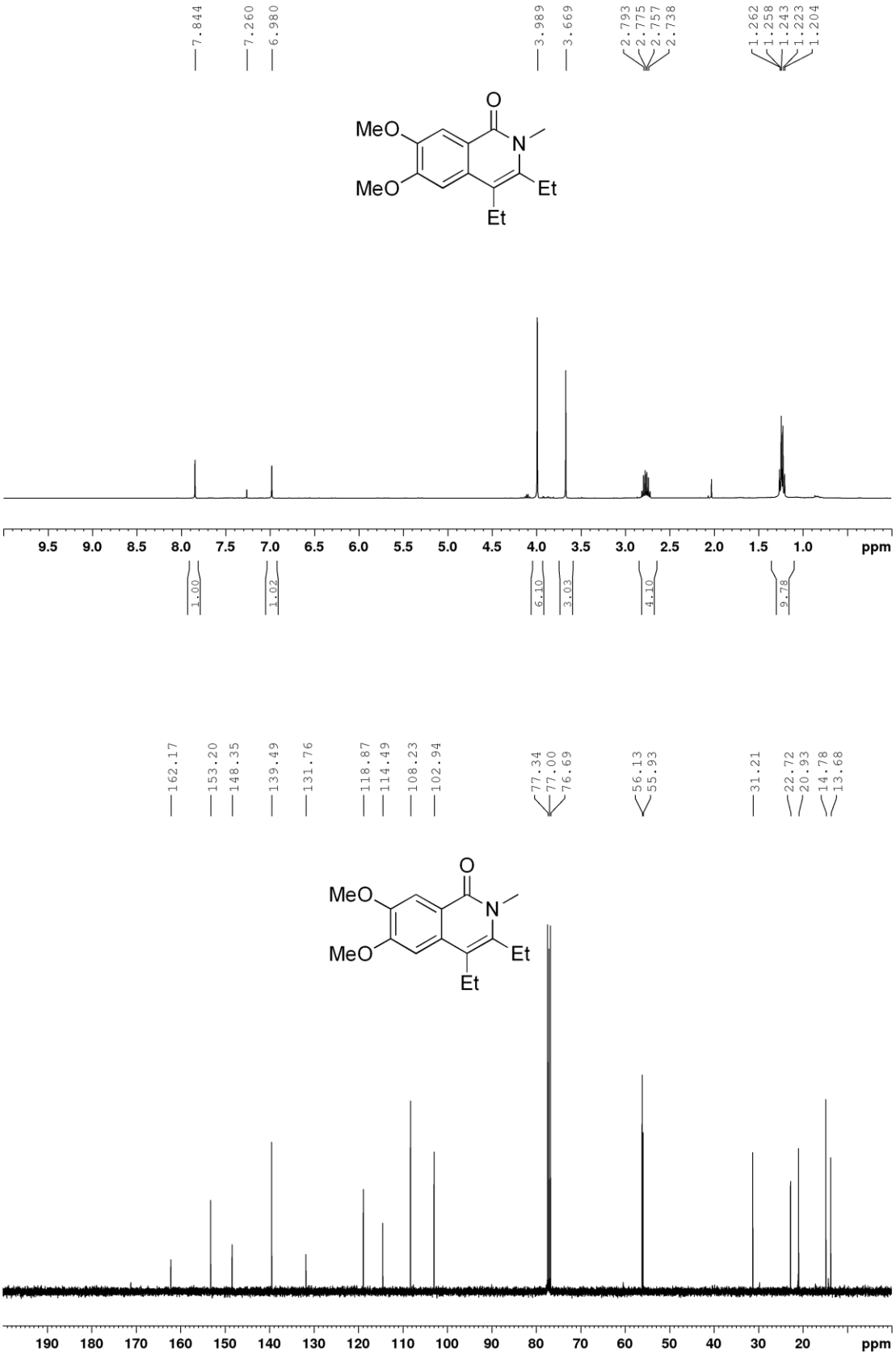



**2-Methyl-3,4-dipropylisoquinolin-1(2*H*)-one (3m) (400 MHz)**

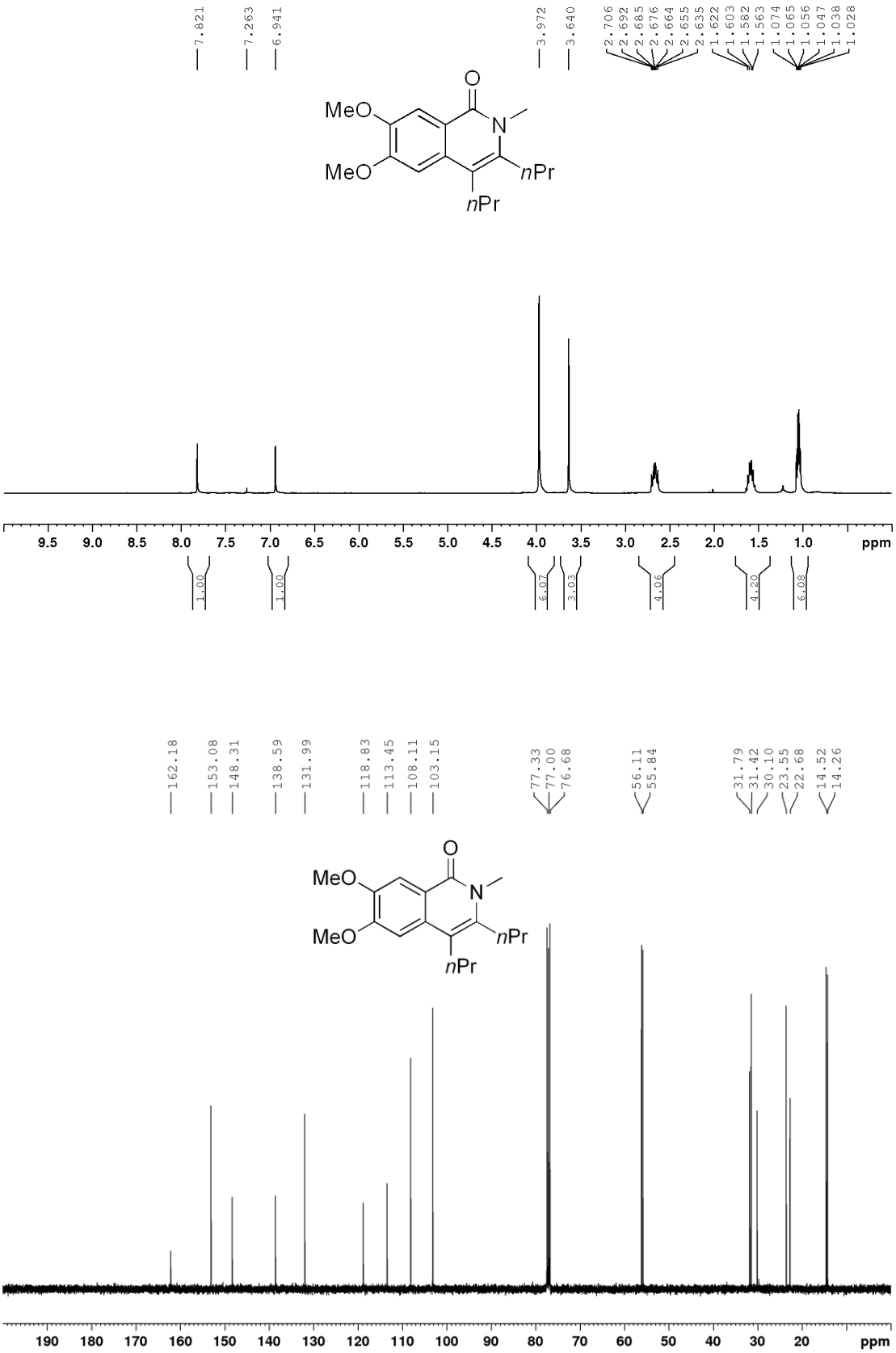


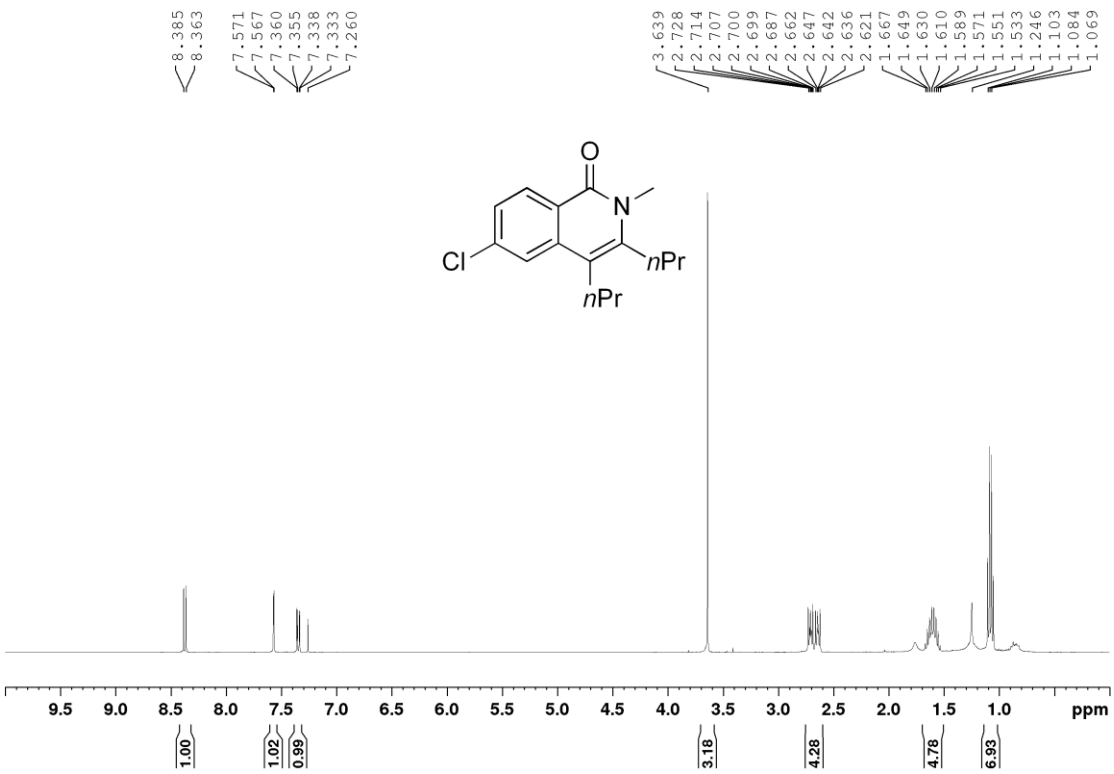
**3,4-Diethyl-6,7-dimethoxy-2-methylisoquinolin-1(2*H*)-one (3n) (400 MHz)**

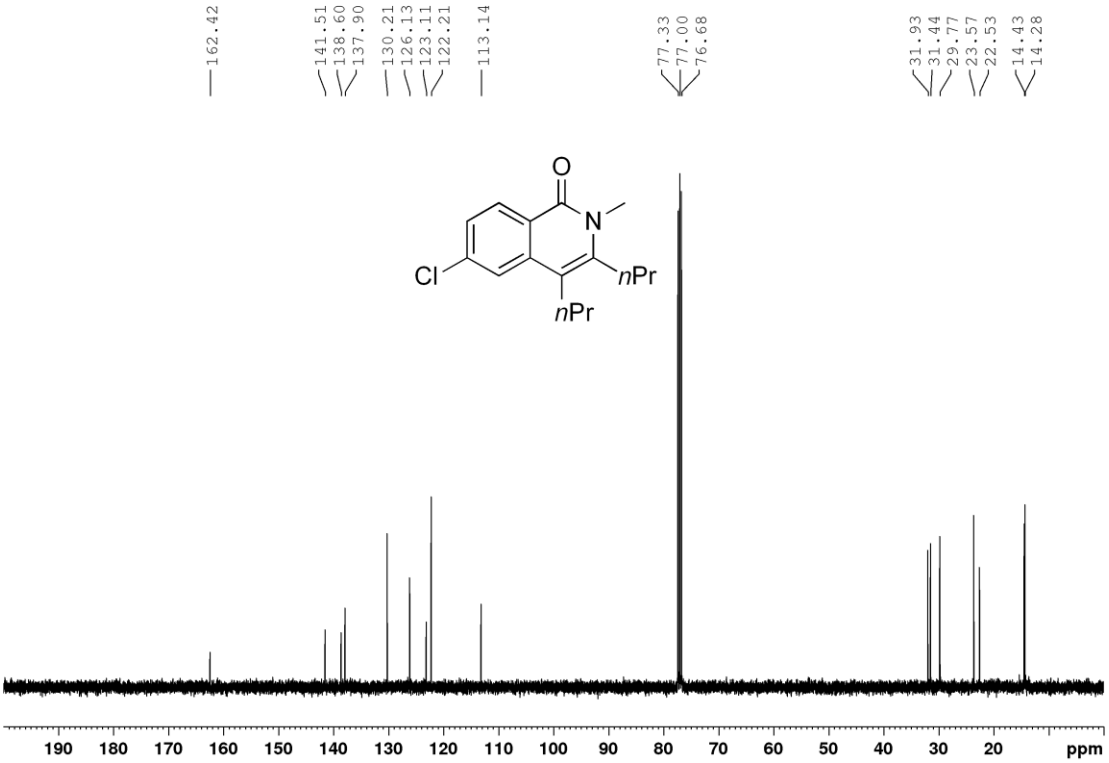


**6,7-Dimethoxy-2-methyl-3,4-dipropylisoquinolin-1(2*H*)-one (3o) (400 MHz)**

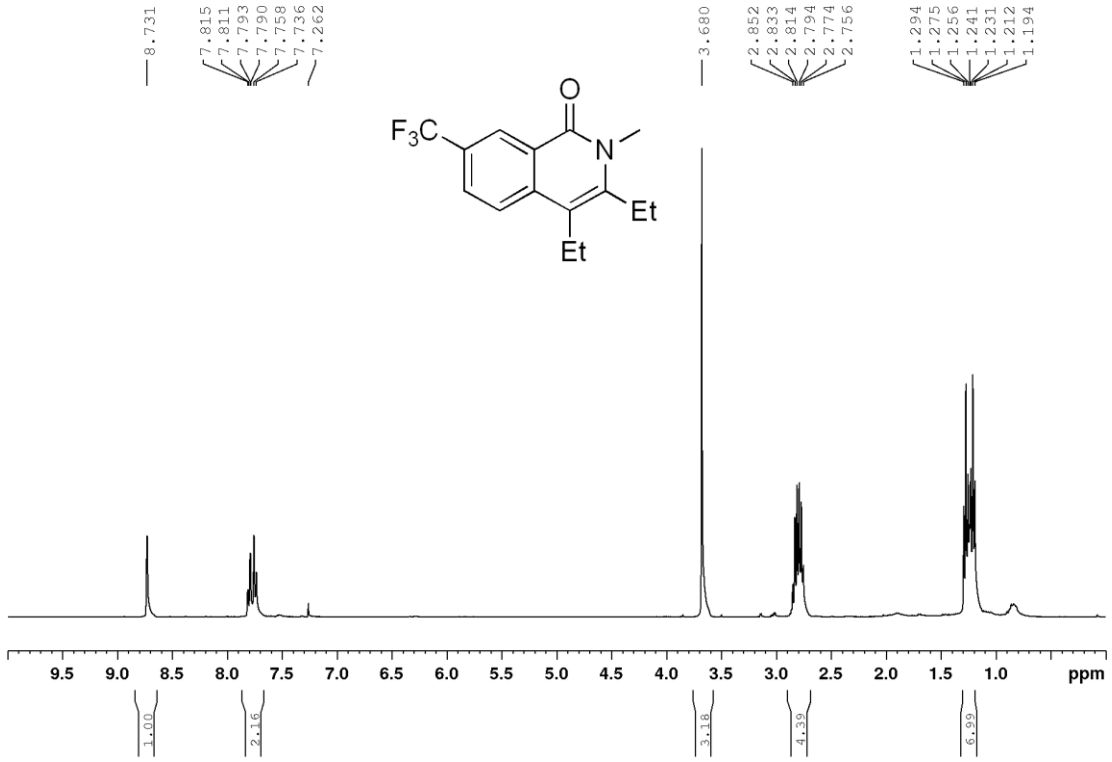


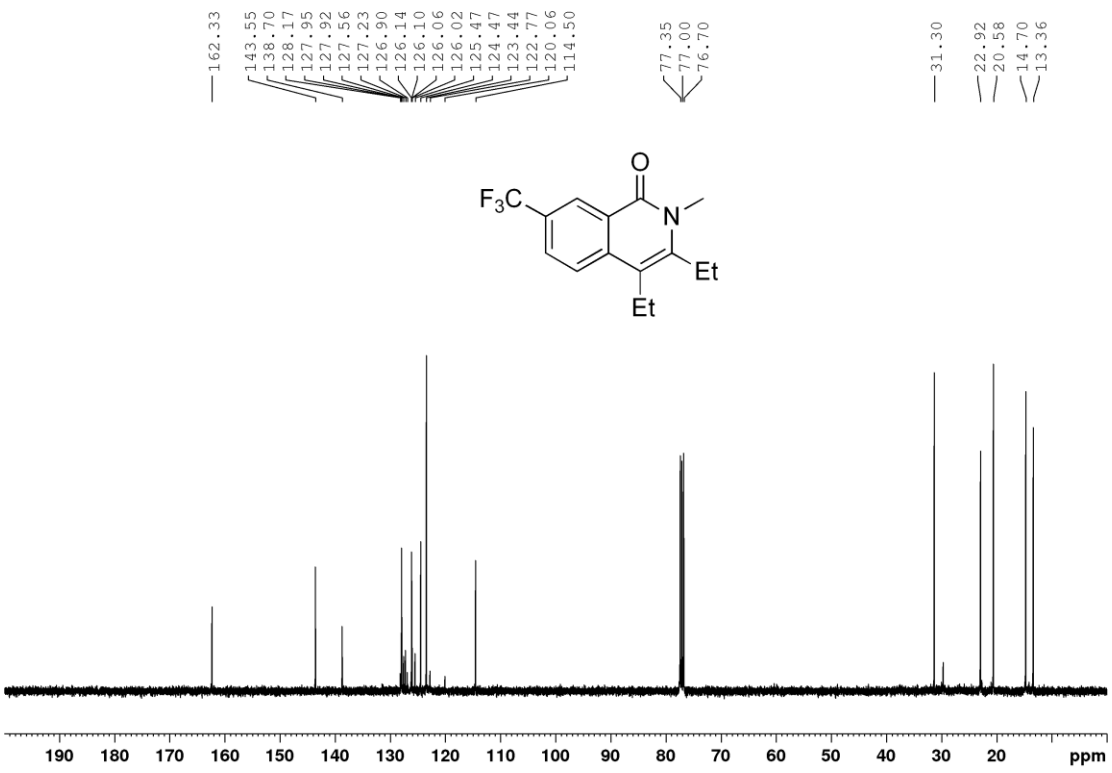
**6-Chloro-2-methyl-3,4-dipropylisoquinolin-1(2*H*)-one (3p) (400 MHz)**



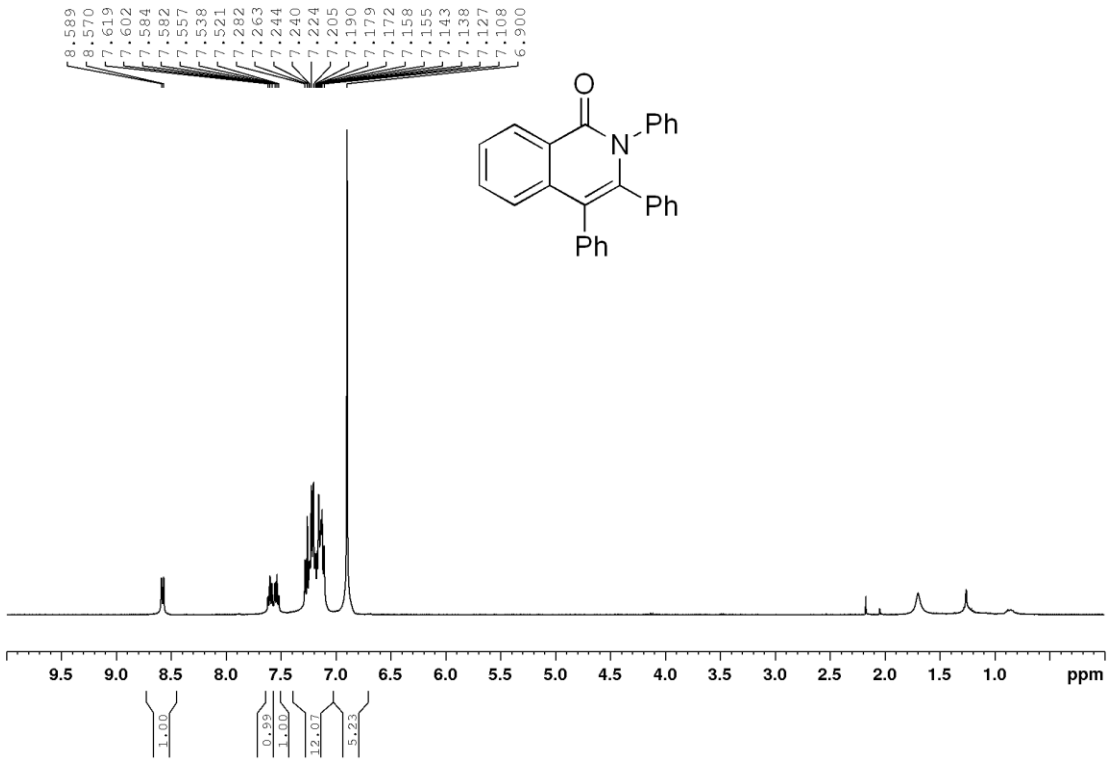



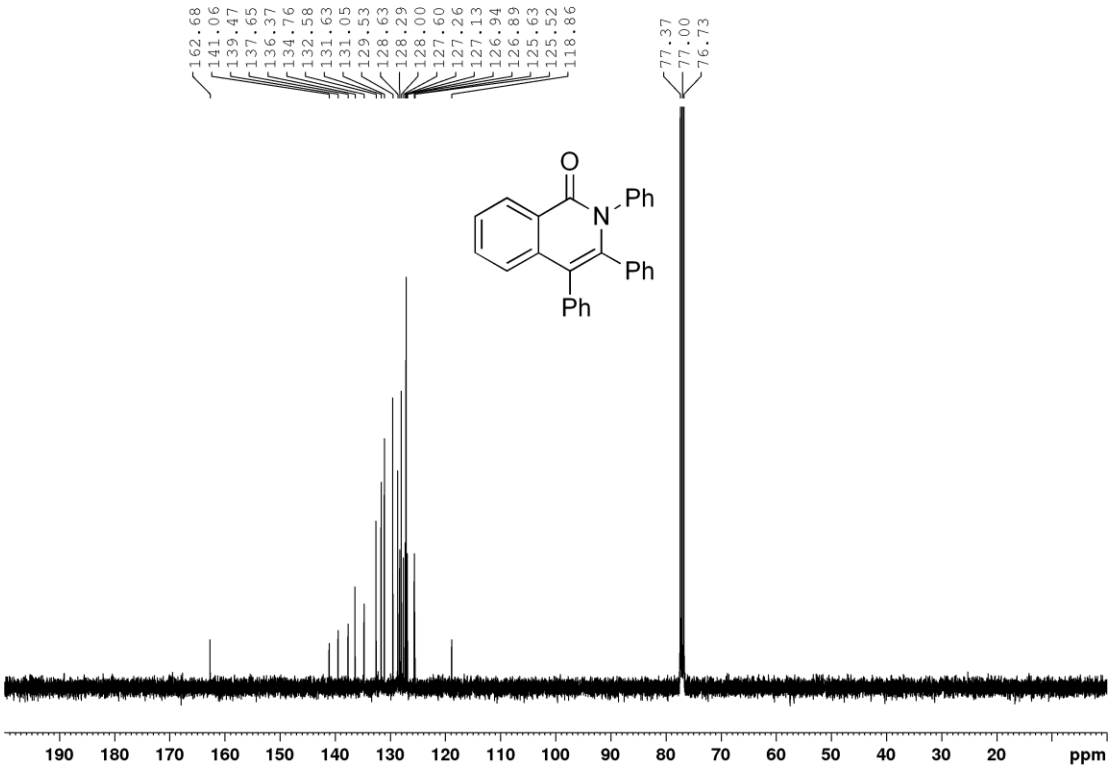
**3,4-Diethyl-2-methyl-7-(trifluoromethyl)isoquinolin-1(2*H*)-one (3q) (400 MHz)**



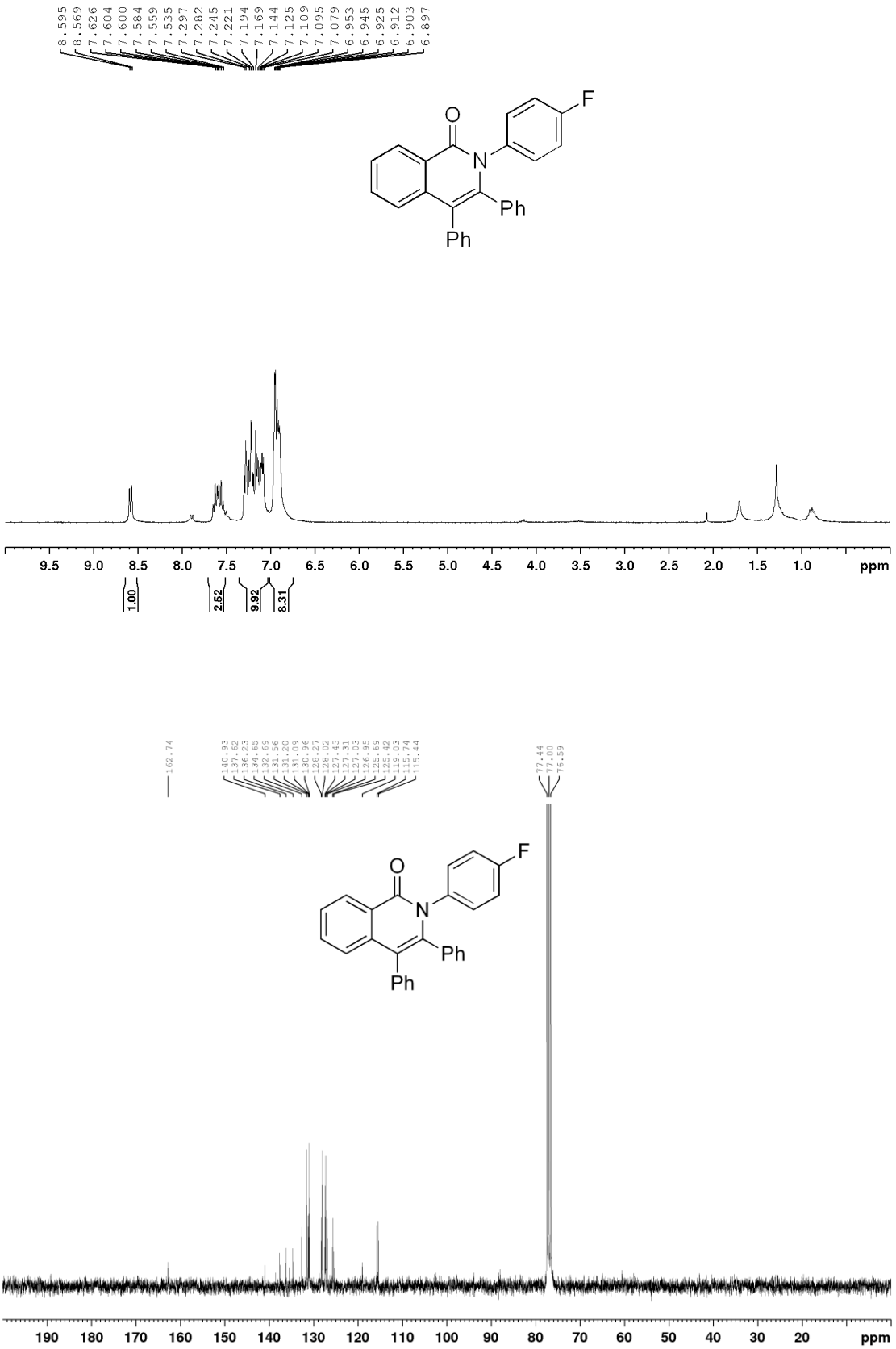



**2,3,4-Triphenylisoquinolin-1(2*H*)-one (3r) (400 MHz)**

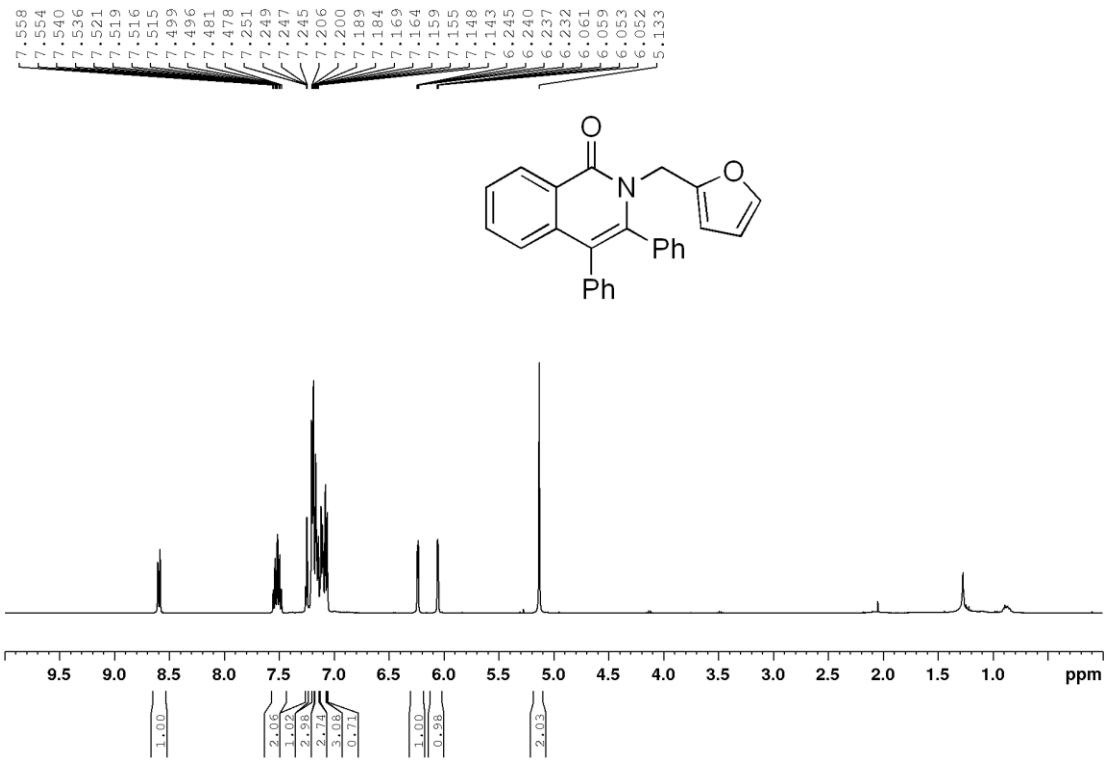


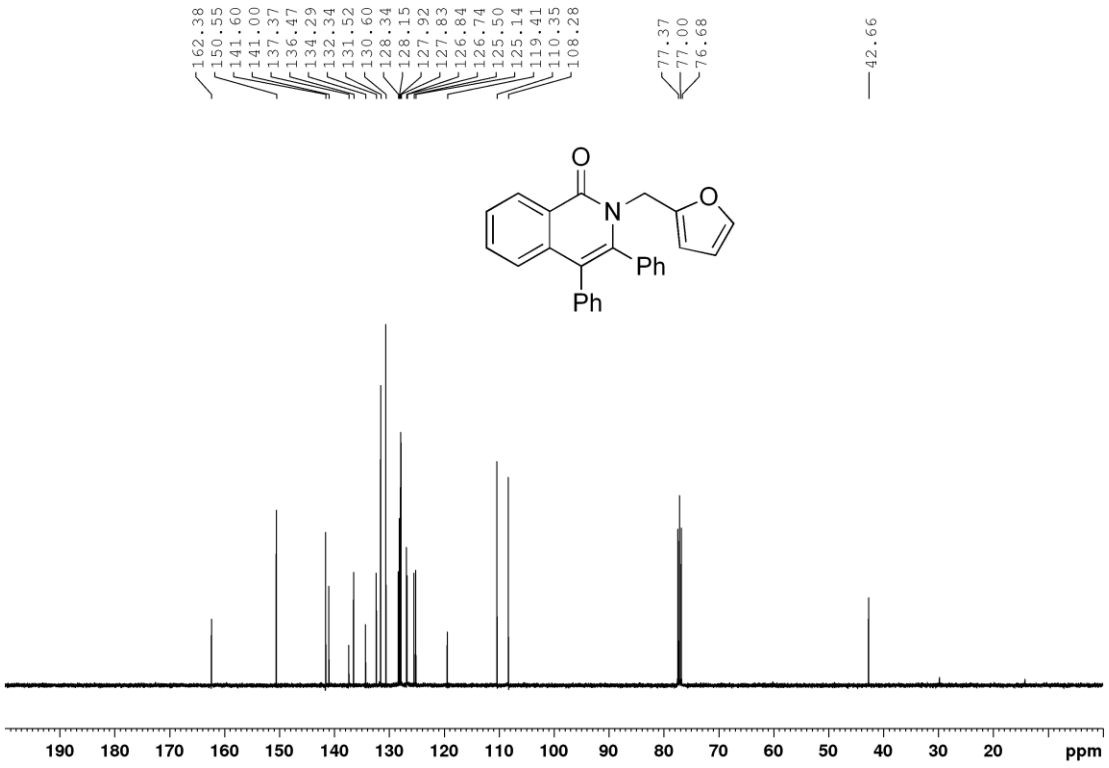



**2-(4-Fluorophenyl)-3,4-diphenylisoquinolin-1(2*H*)-one (3s) (400 MHz)**

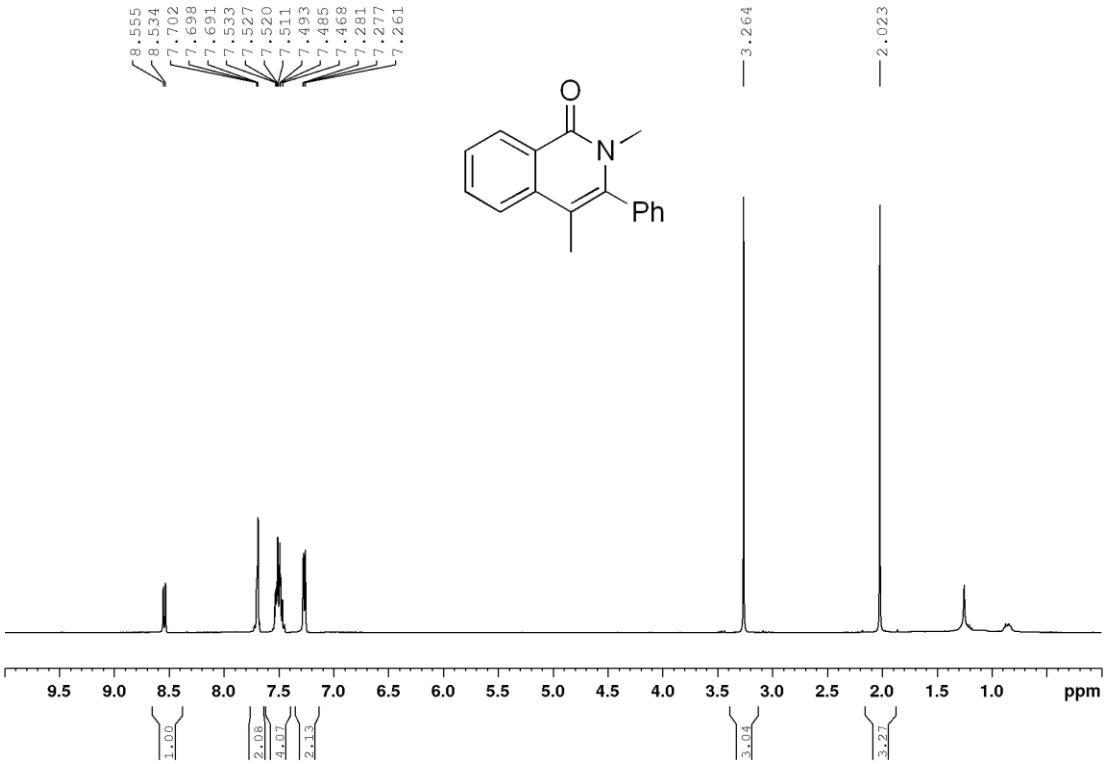


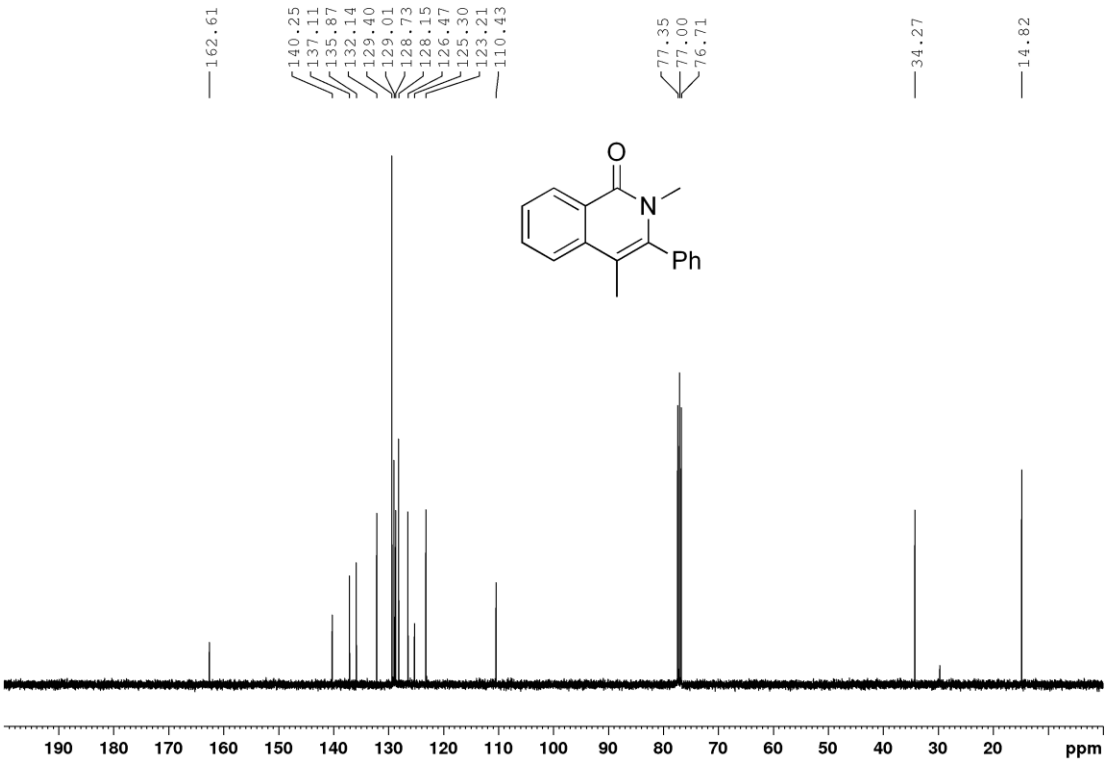
**2-(Furan-2-ylmethyl)-3,4-diphenylisoquinolin-1(2*H*)-one (3t) (400 MHz)**



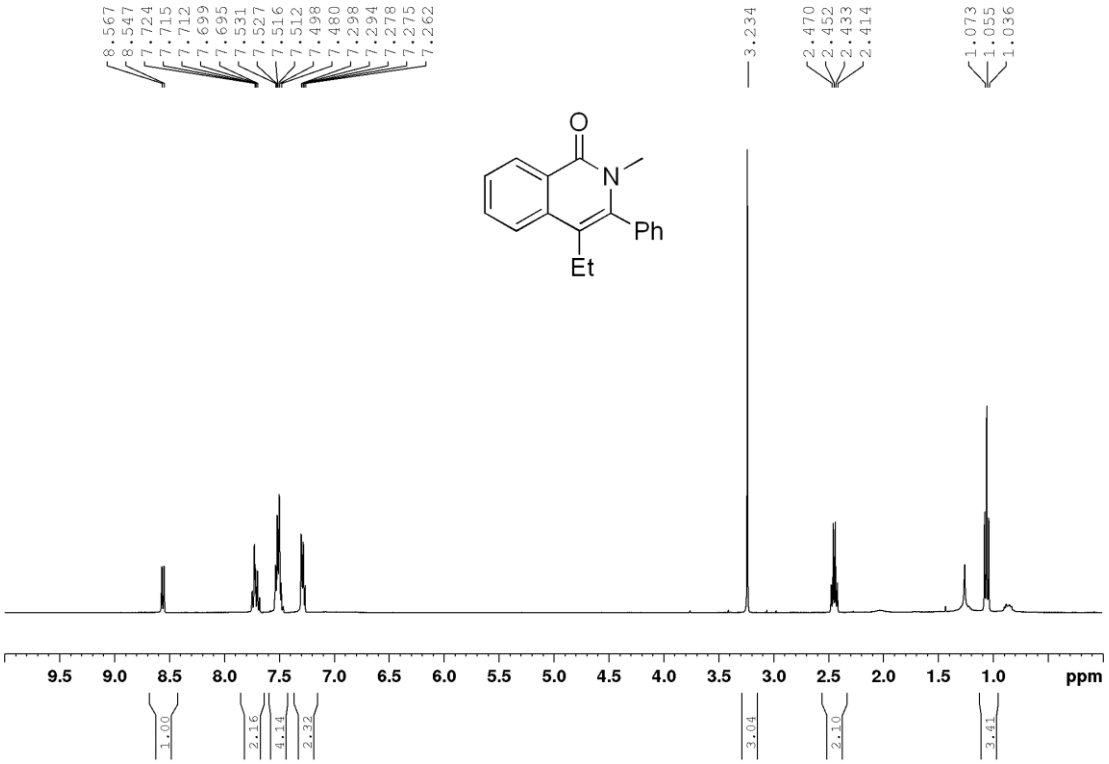



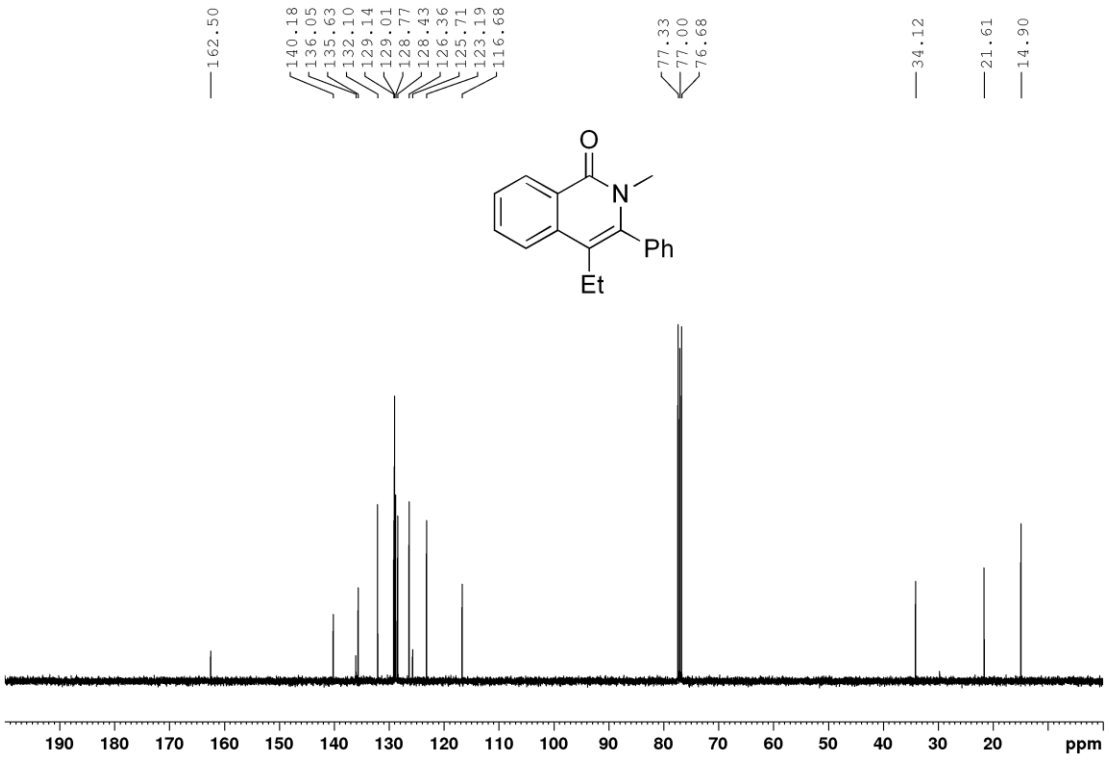
**2,4-Dimethyl-3-phenylisoquinolin-1(2*H*)-one (3u) (400 MHz)**



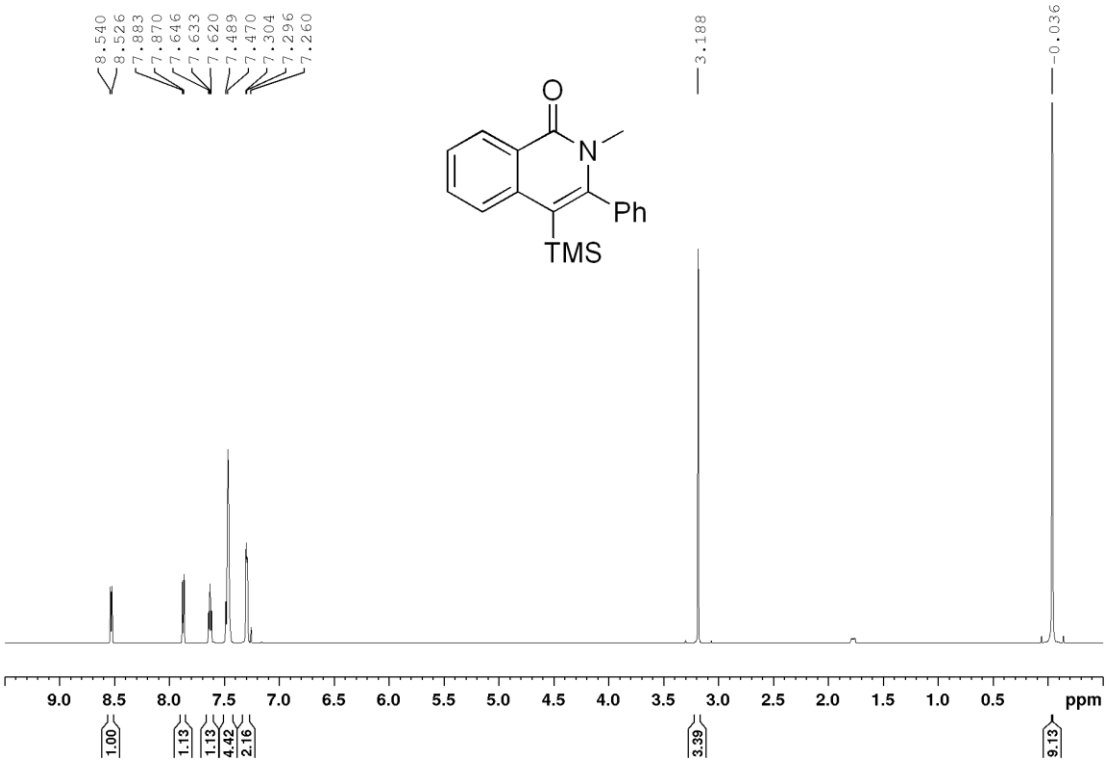



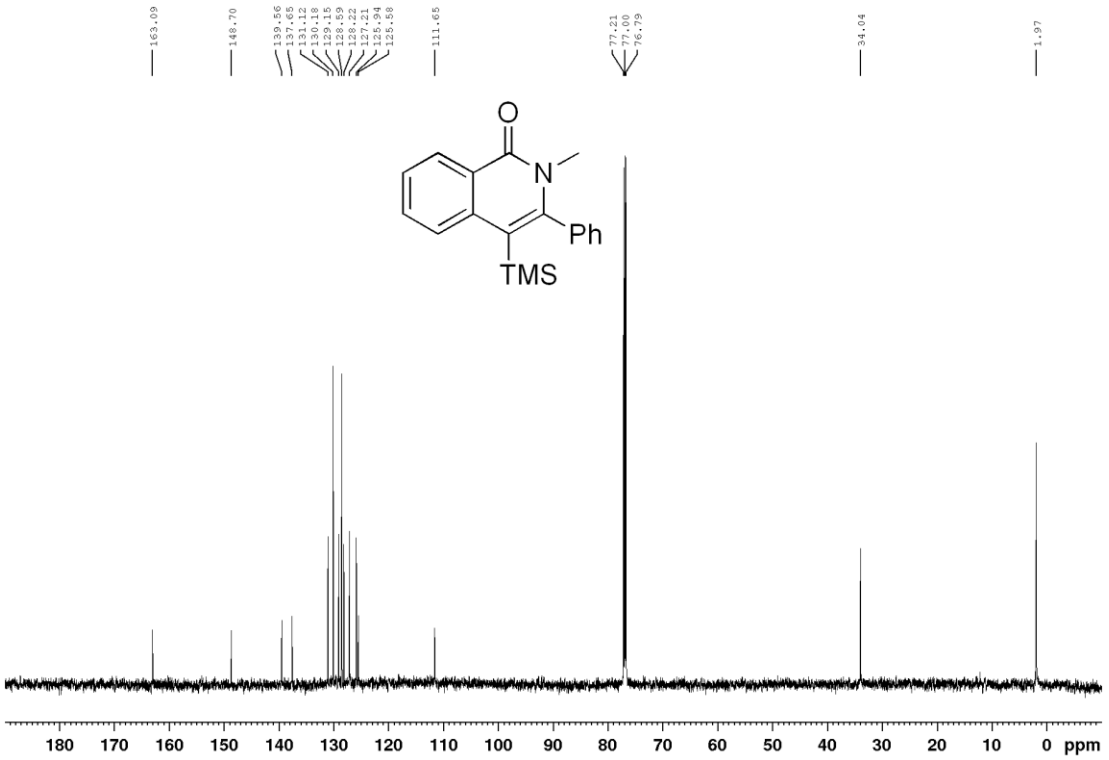
**4-Ethyl-2-methyl-3-phenylisoquinolin-1(2*H*)-one (3v) (400 MHz)**



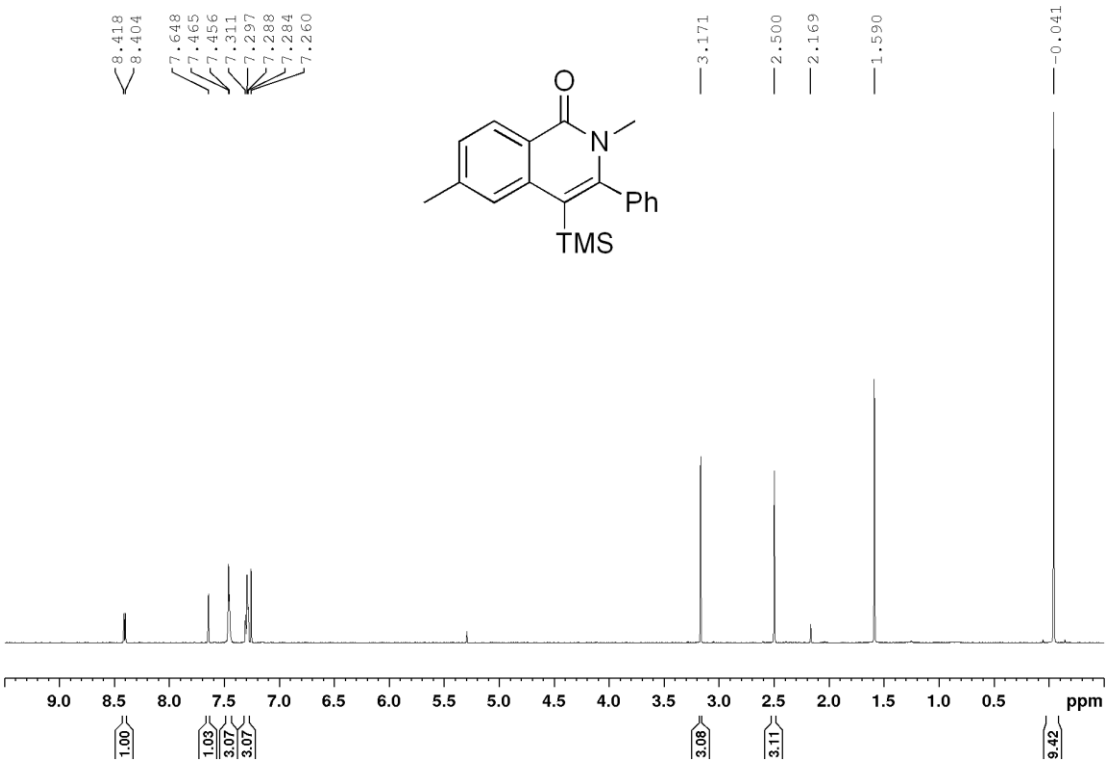



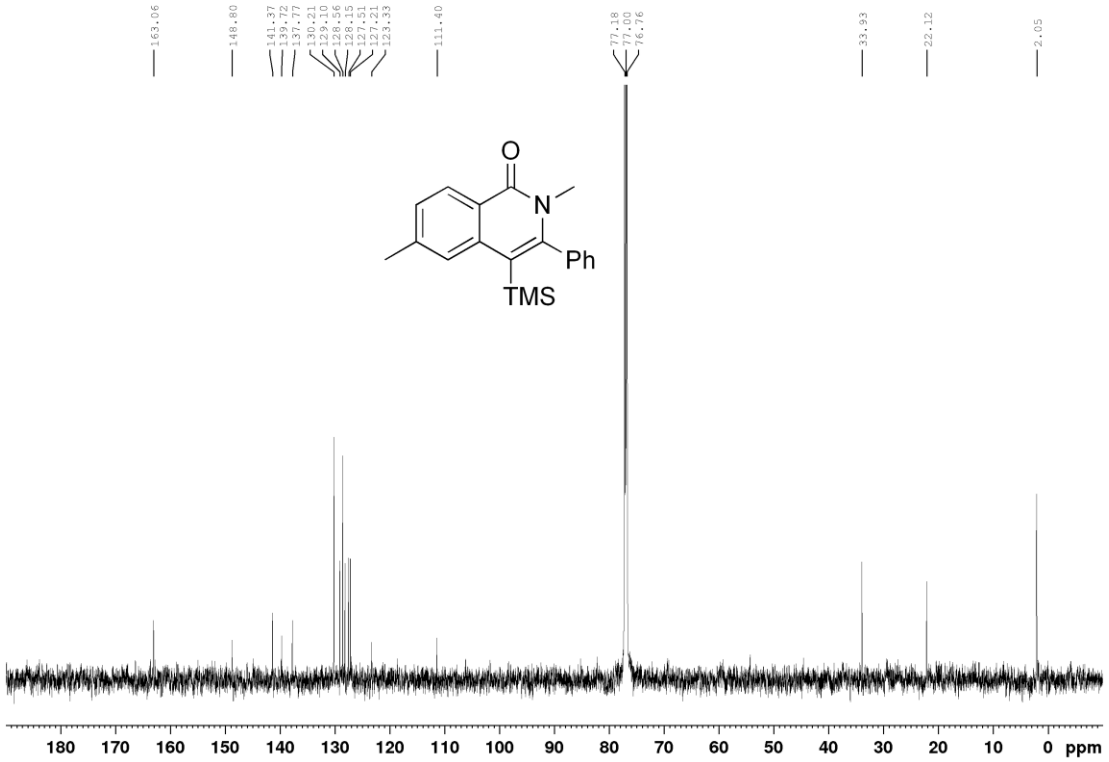
**2-Methyl-3-phenyl-4-(trimethylsilyl)isoquinolin-1(2*H*)-one (3w) (600 MHz)**



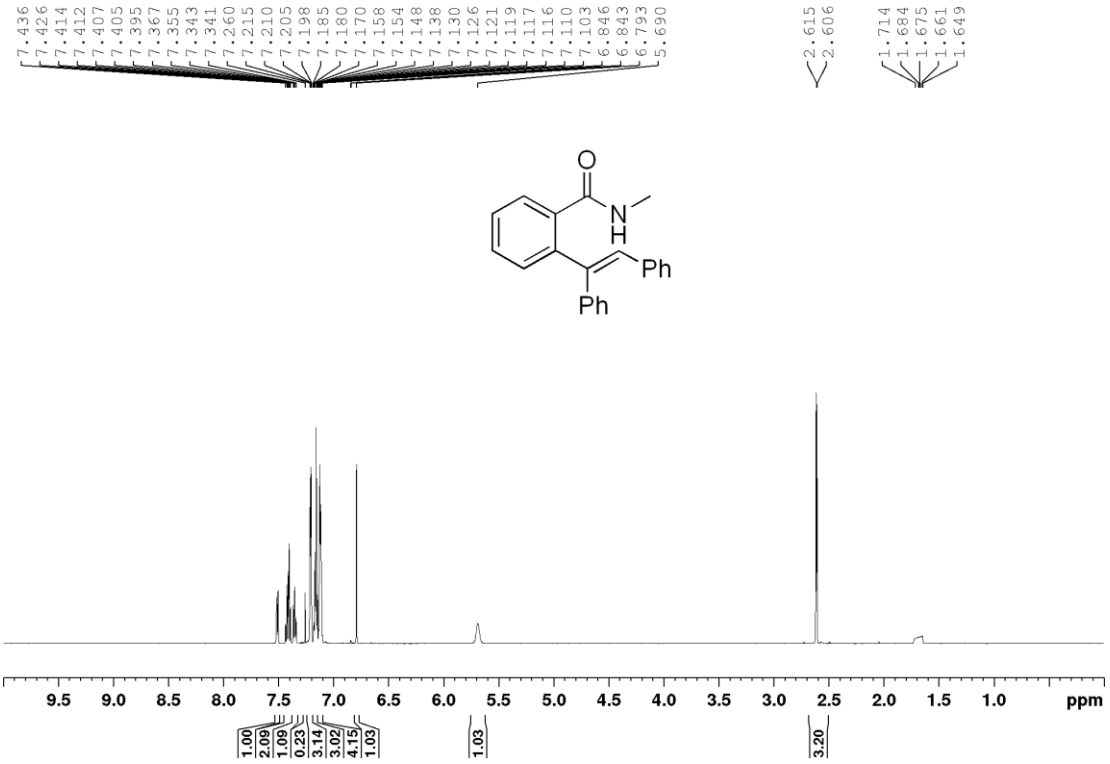



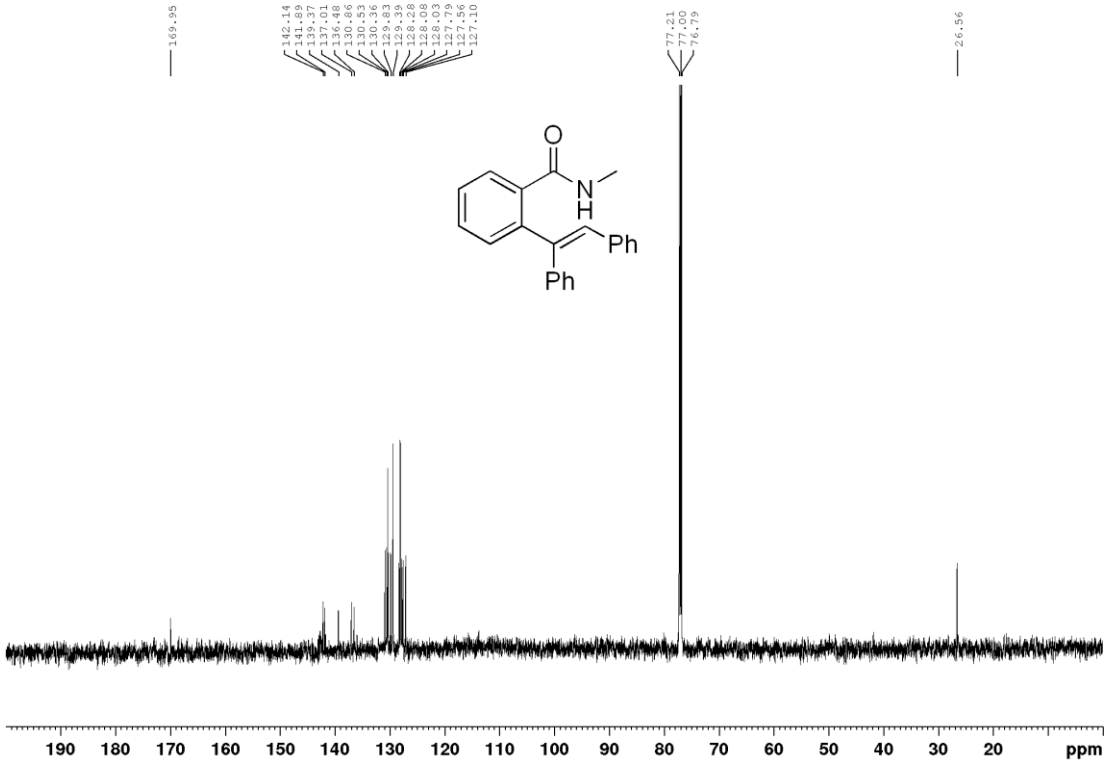
**2,6-Dimethyl-3-phenyl-4-(trimethylsilyl)isoquinolin-1(2*H*)-one (3x) (600 MHz)**



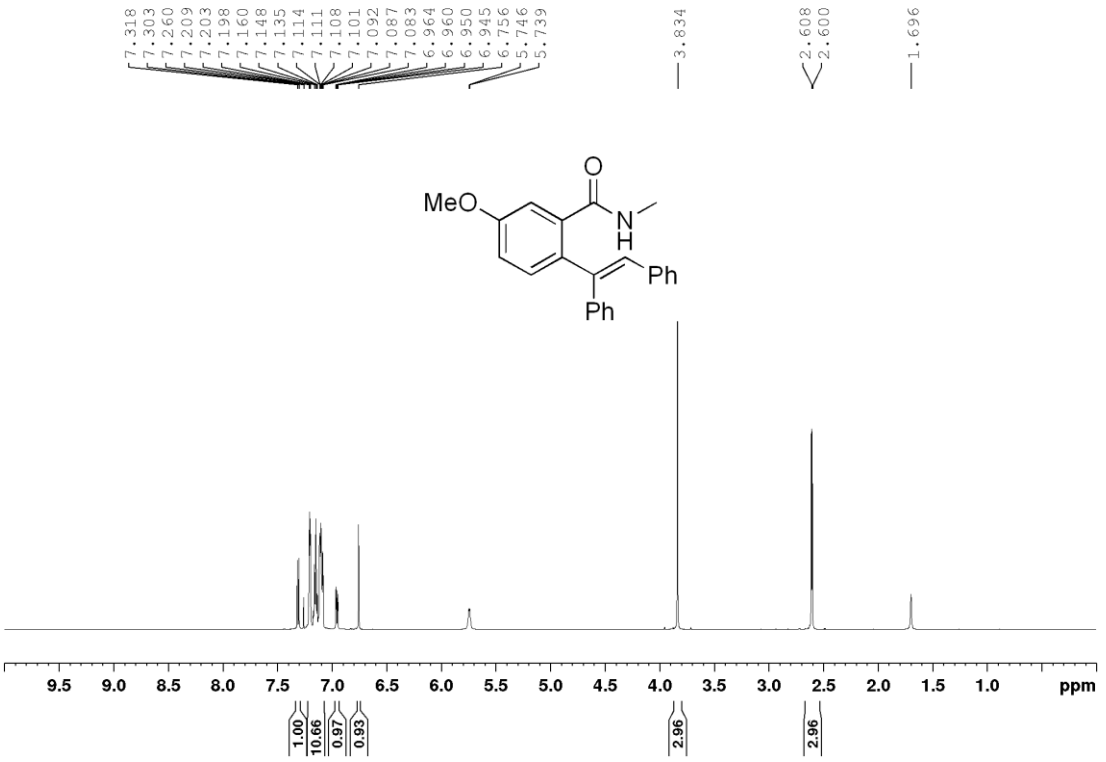



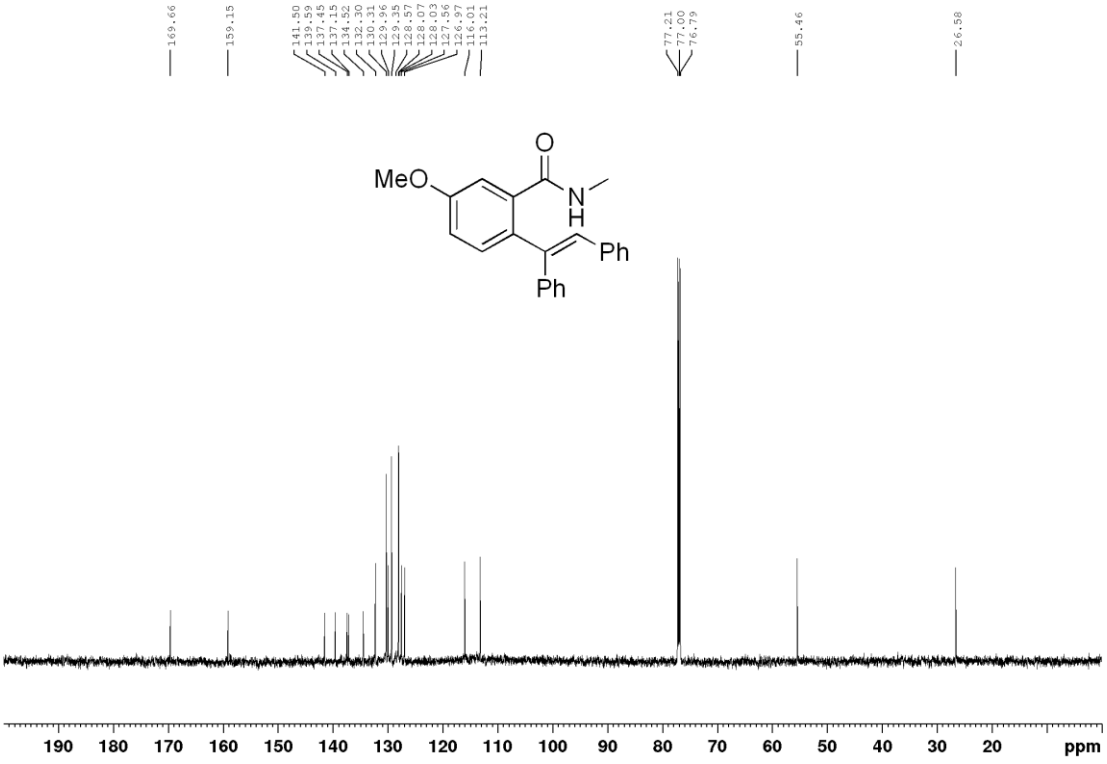
**(*E*)-2-(1,2-Diphenylvinyl)-*N*-methylbenzamide (4a) (600 MHz)**



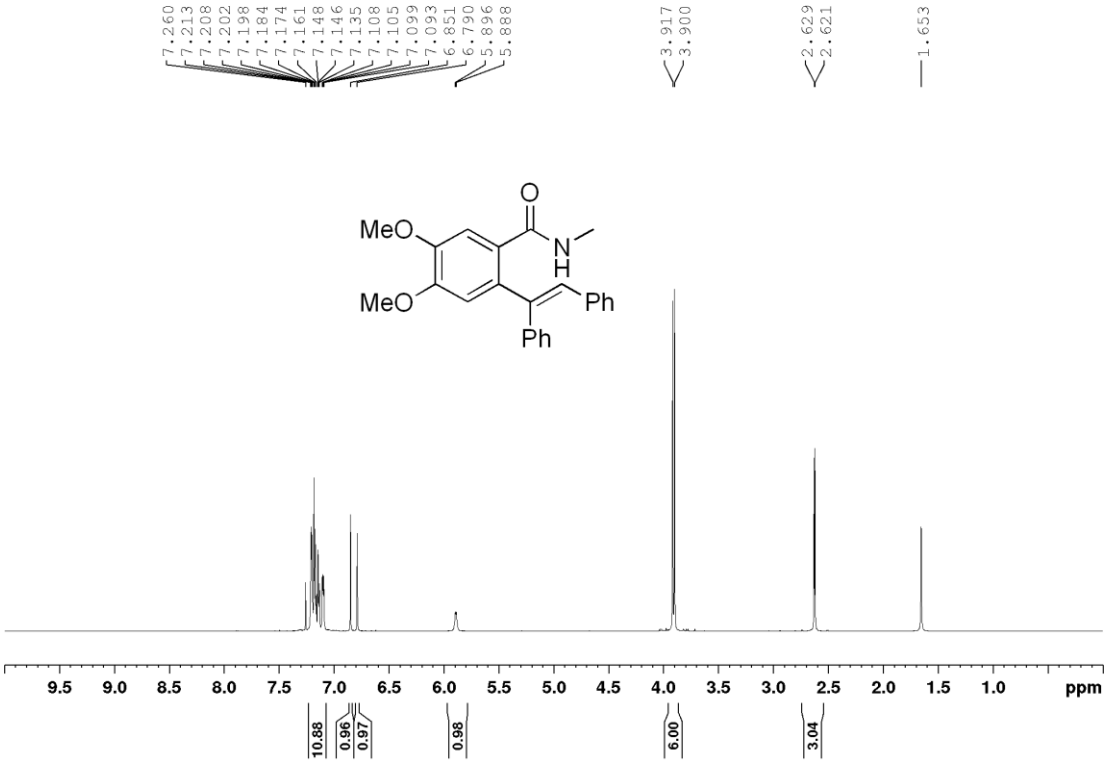



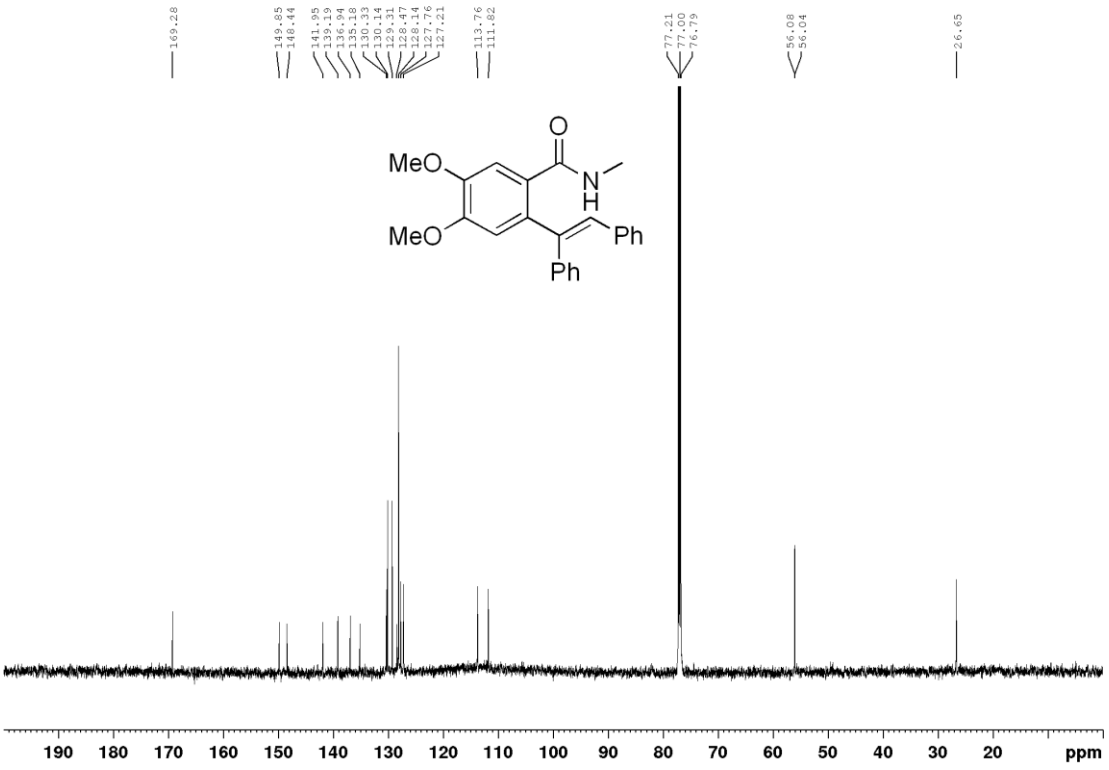
**(*E*)-2-(1,2-Diphenylvinyl)-5-methoxy-*N*-methylbenzamide (4b) (600 MHz)**



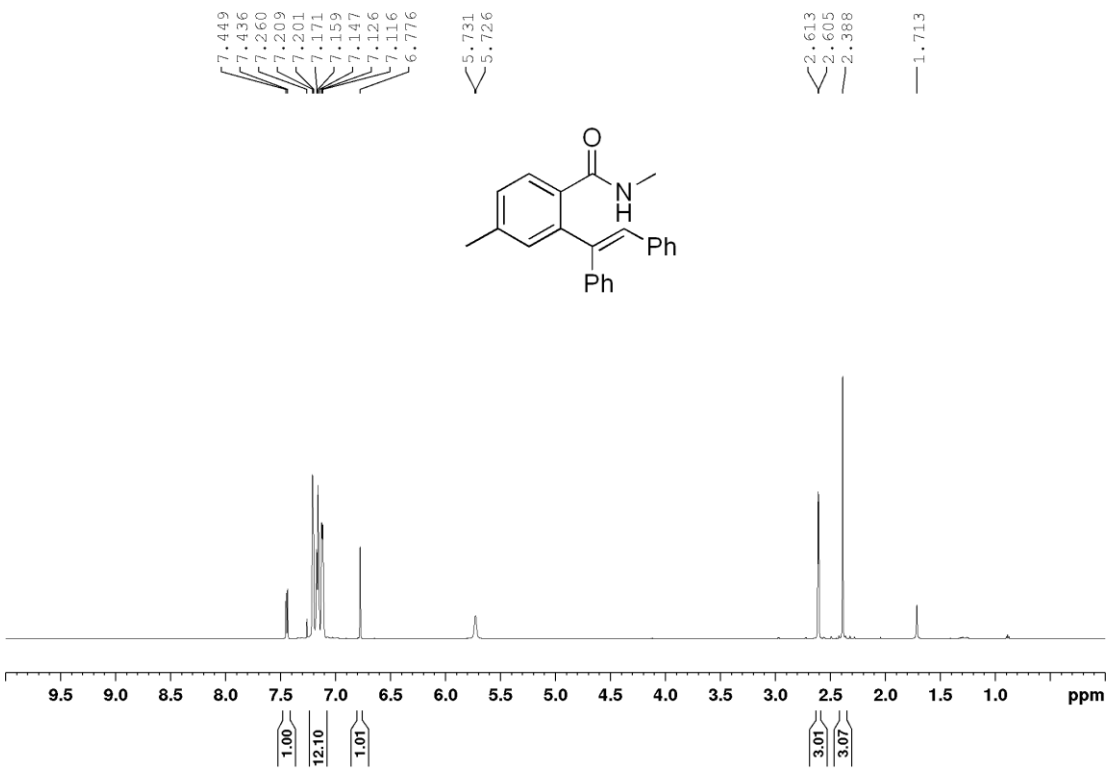



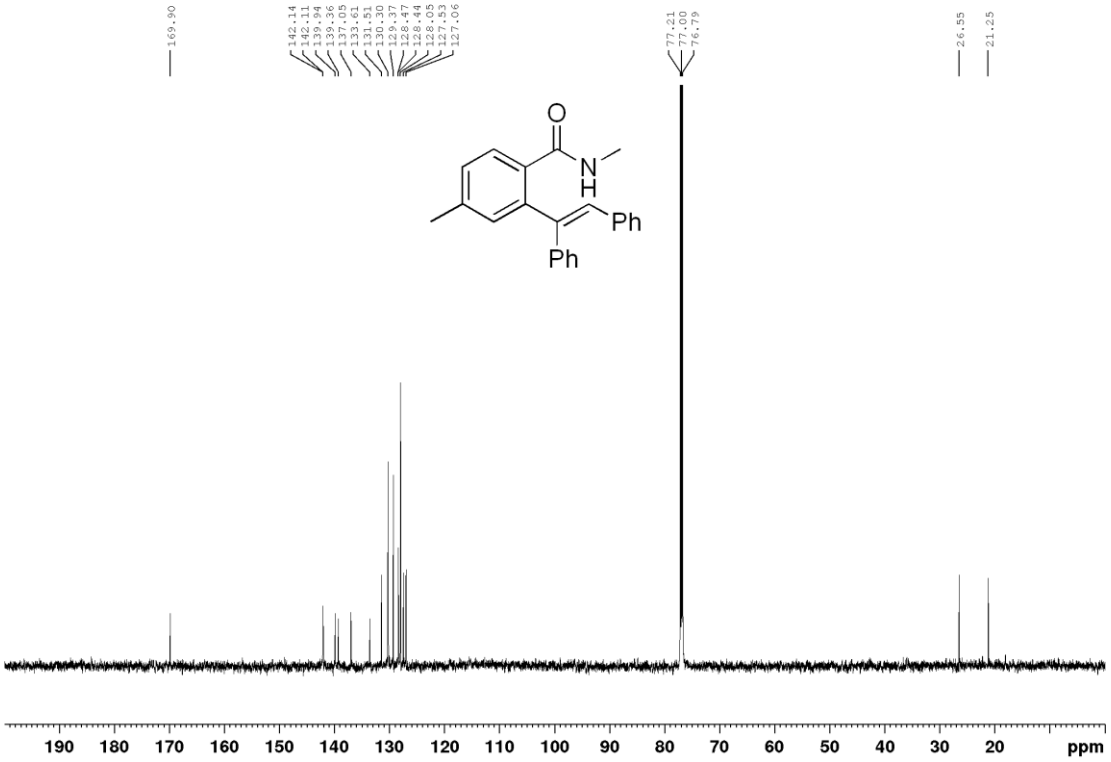
**(*E*)-2-(1,2-Diphenylvinyl)-4,5-dimethoxy-*N*-methylbenzamide (4c) (600 MHz)**



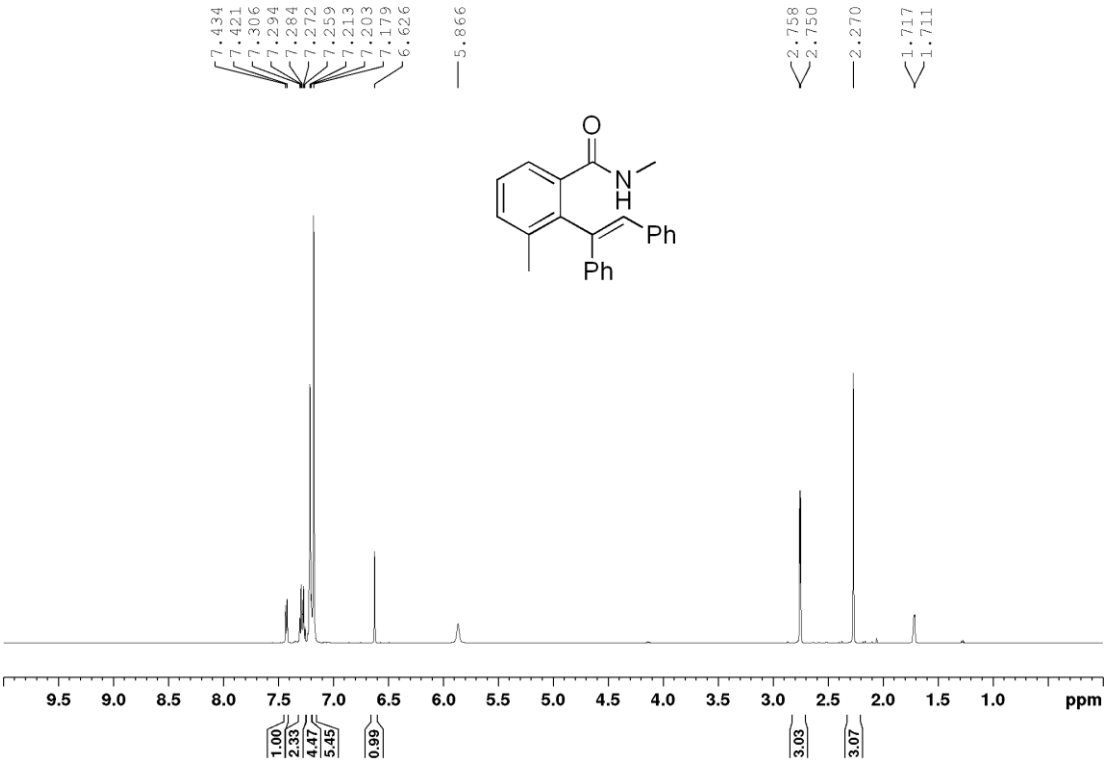



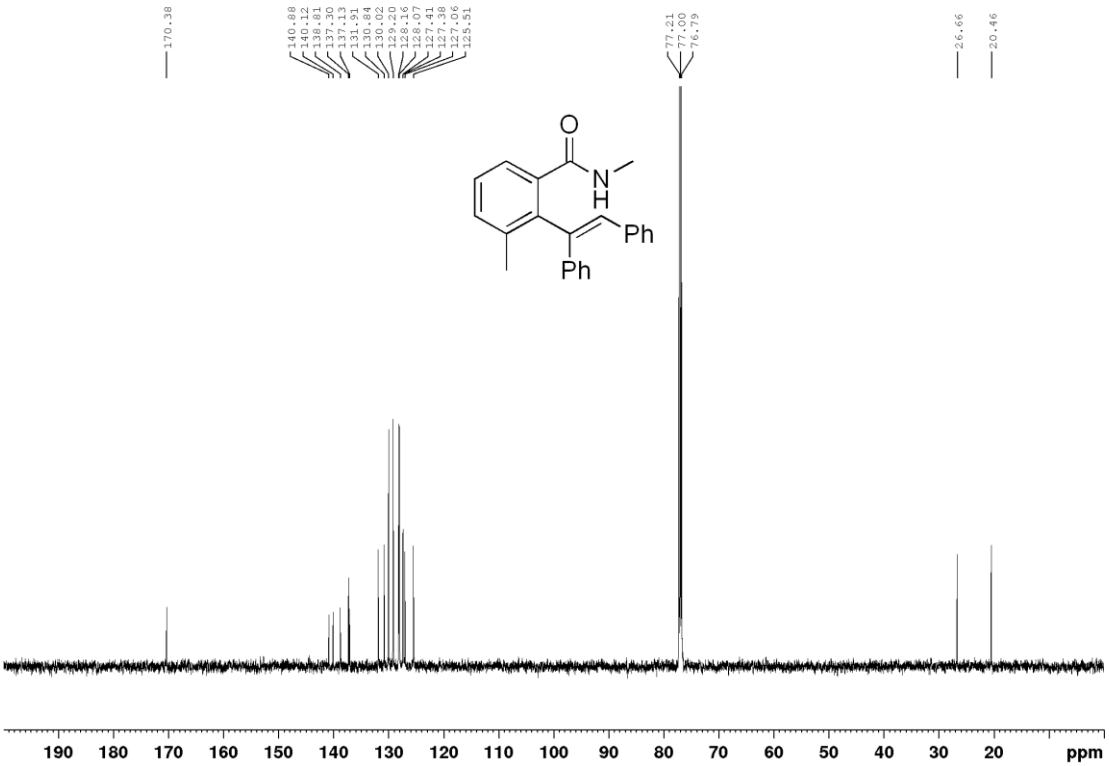
**(*E*)-2-(1,2-Diphenylvinyl)-*N*,4-dimethylbenzamide (4d) (600 MHz)**



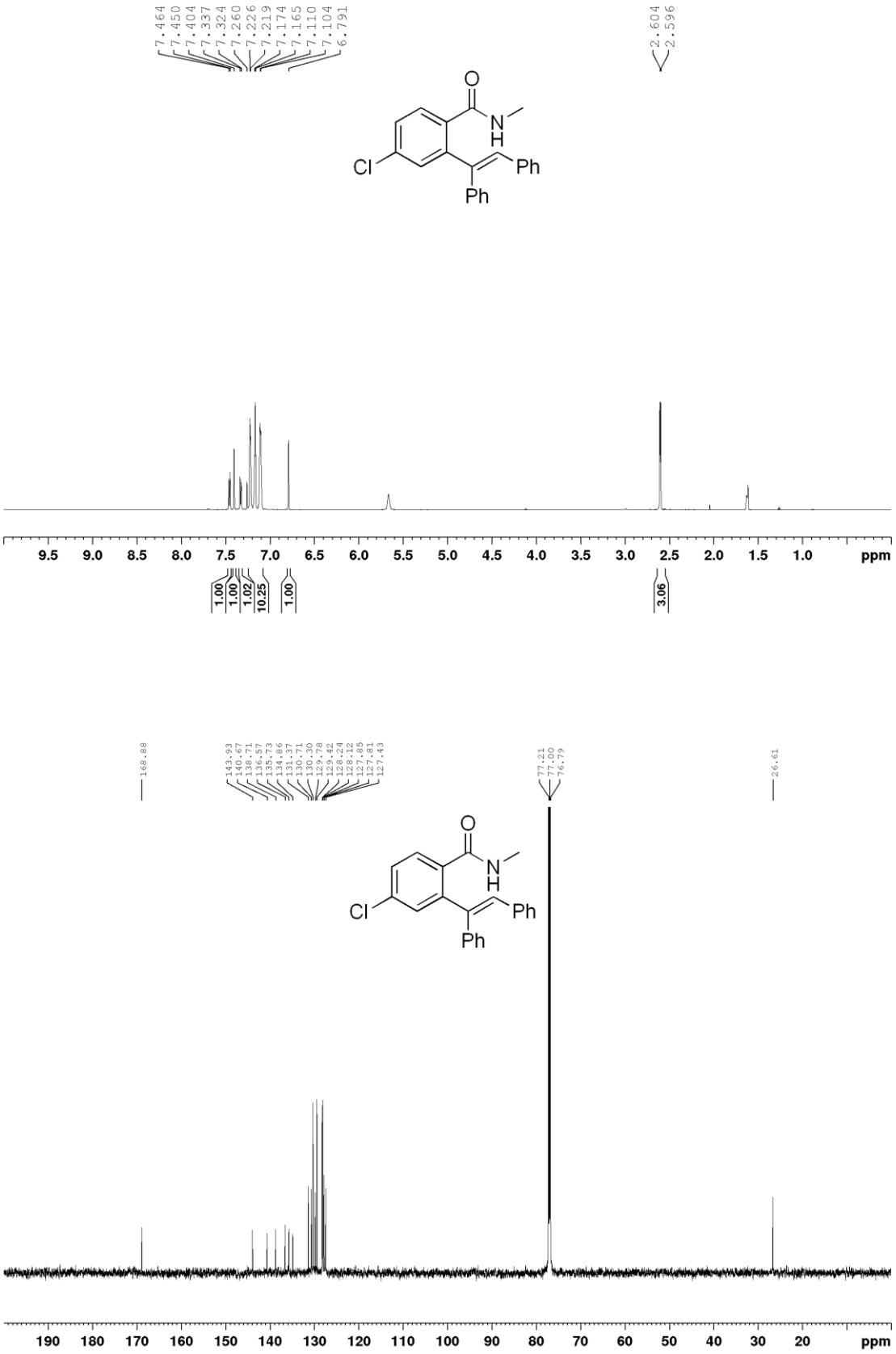



**(*E*)-2-(1,2-Diphenylvinyl)-*N*,3-dimethylbenzamide (4e) (600 MHz)**

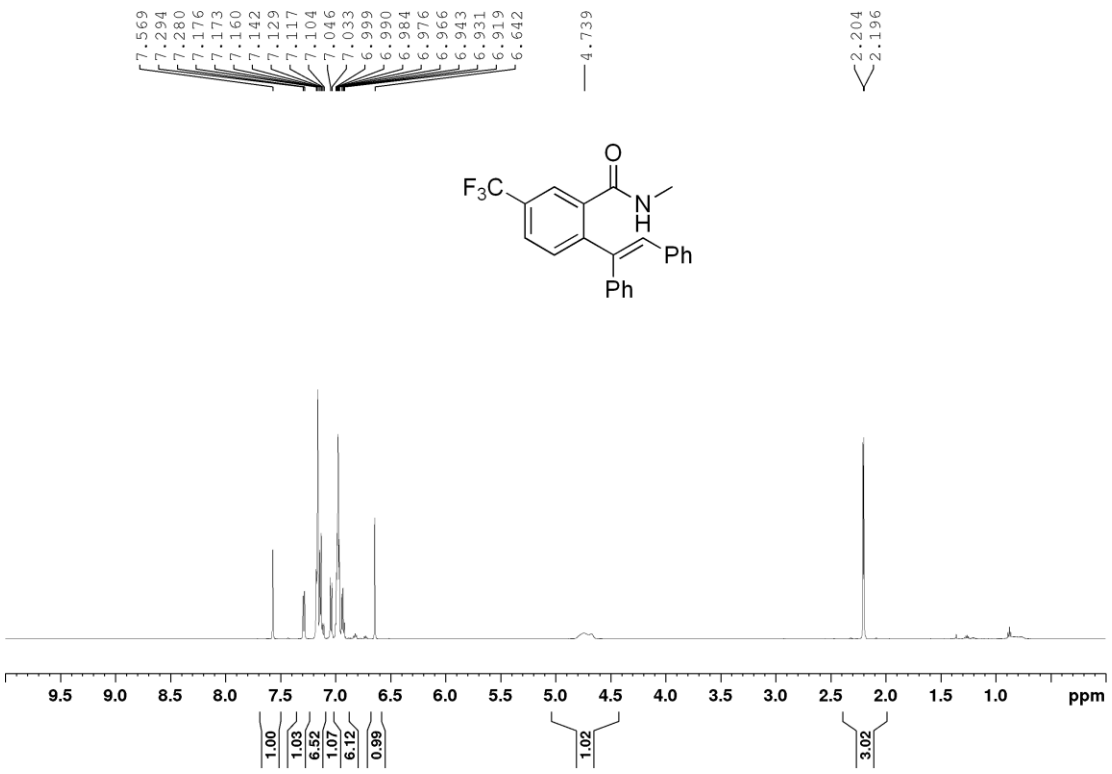


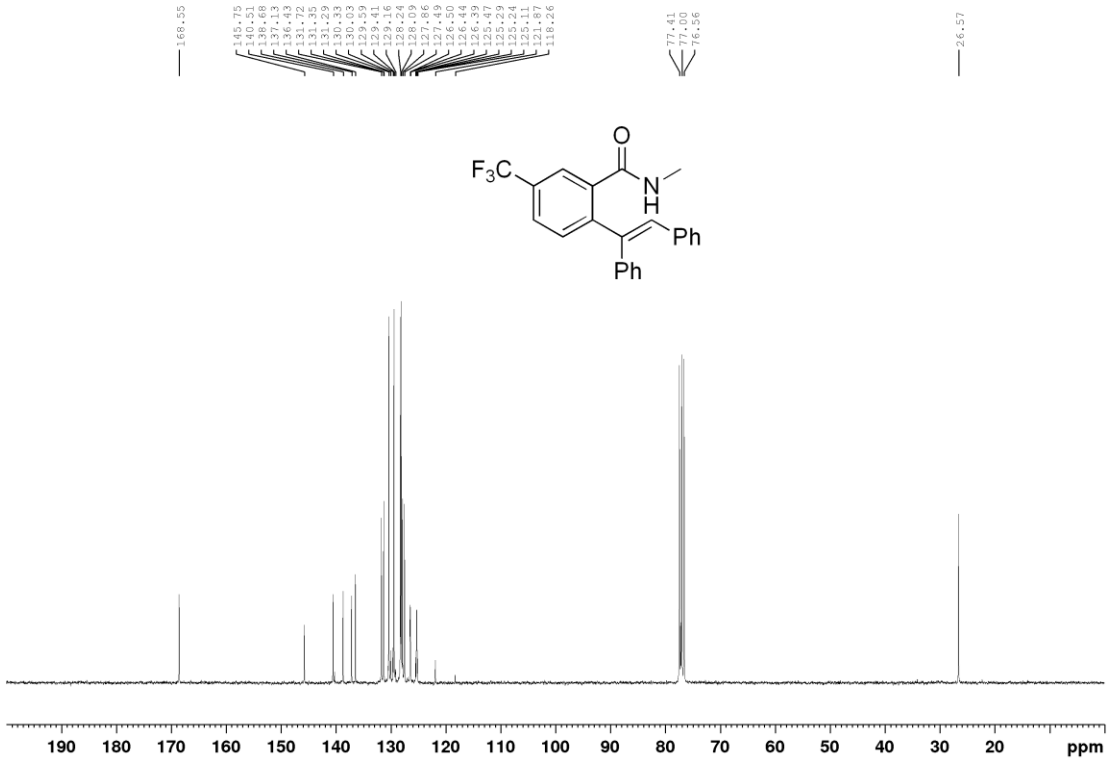



**(*E*)-4-Chloro-2-(1,2-diphenylvinyl)-*N*-methylbenzamide (4f) (600 MHz)**

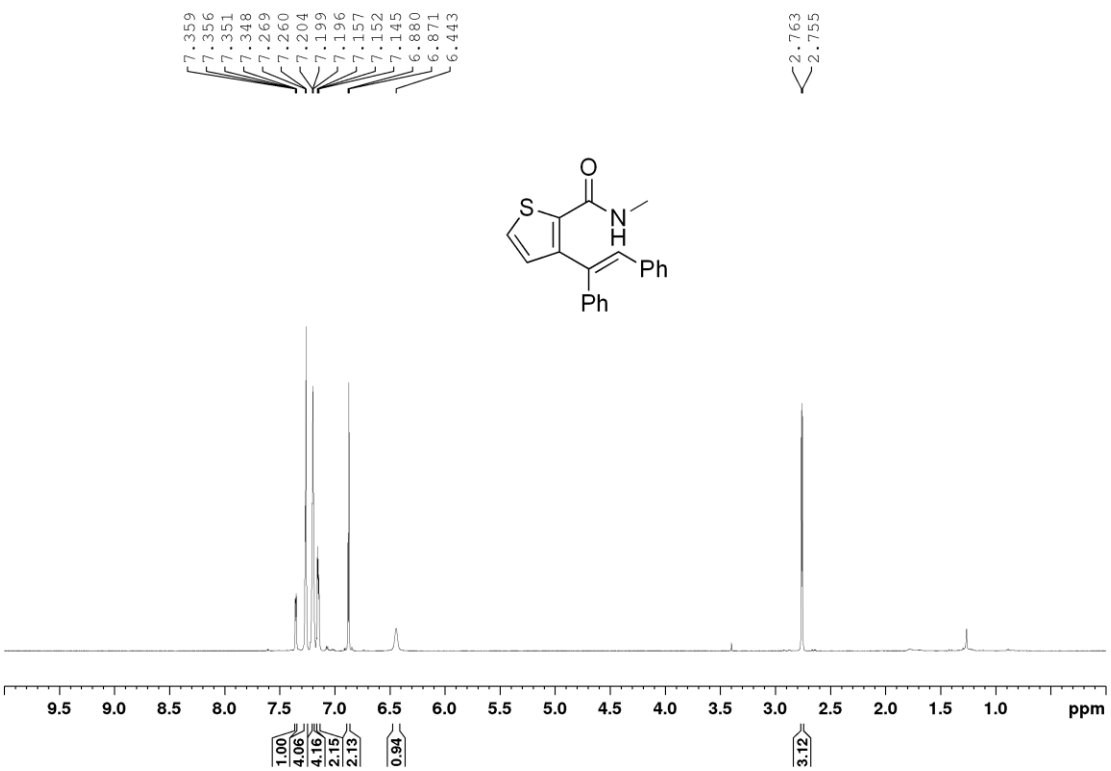


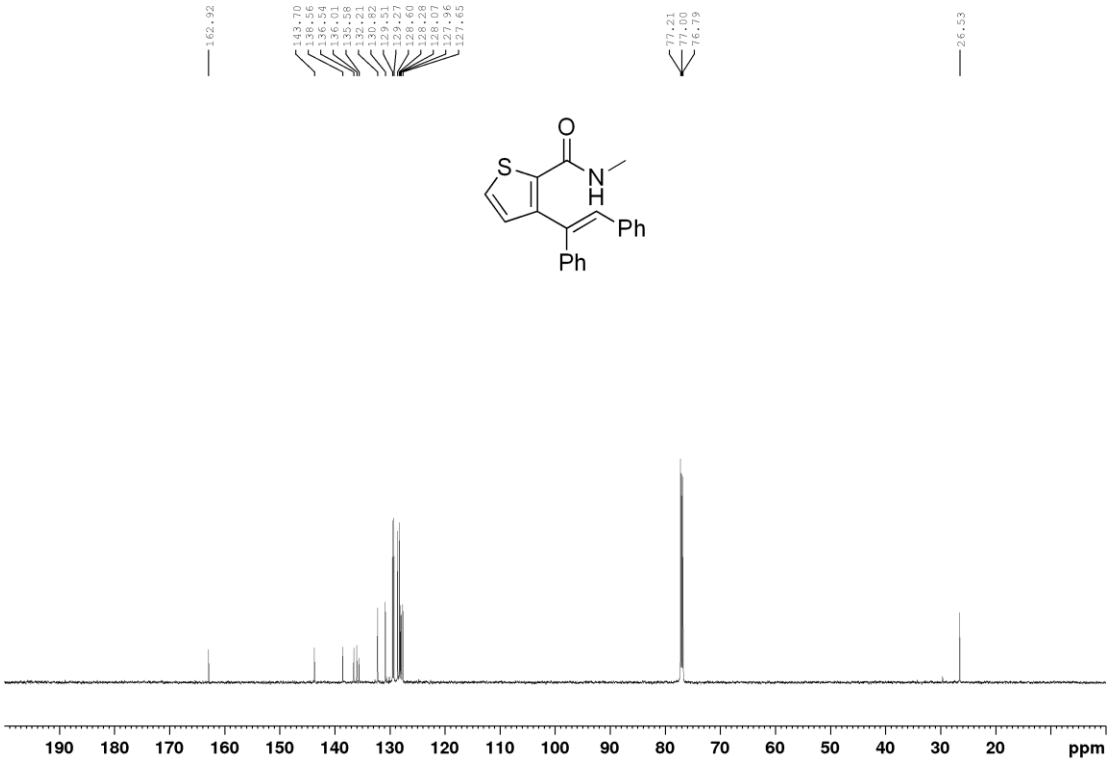
**(*E*)-2-(1,2-diphenylvinyl)-*N*-methyl-5-(trifluoromethyl)benzamide (4g) [$^{1}$H NMR (600 MHz, $C_6D_6$); $^{13}$C NMR (300 MHz, $CDCl_3$)]**



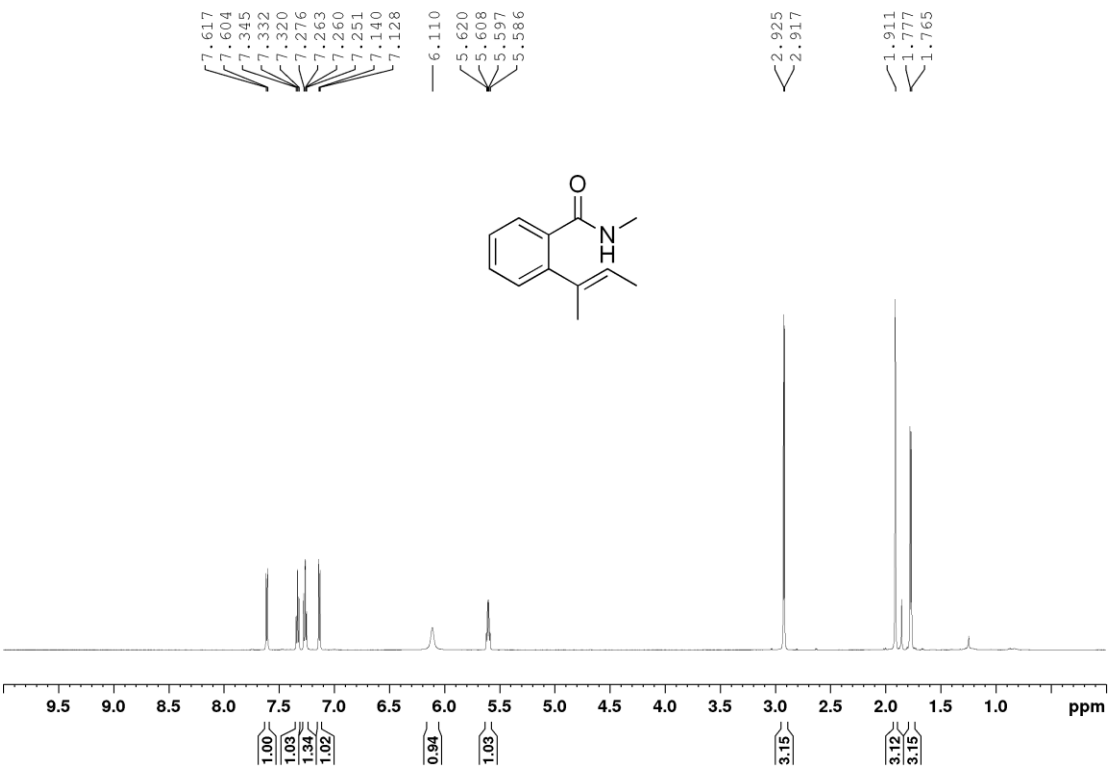



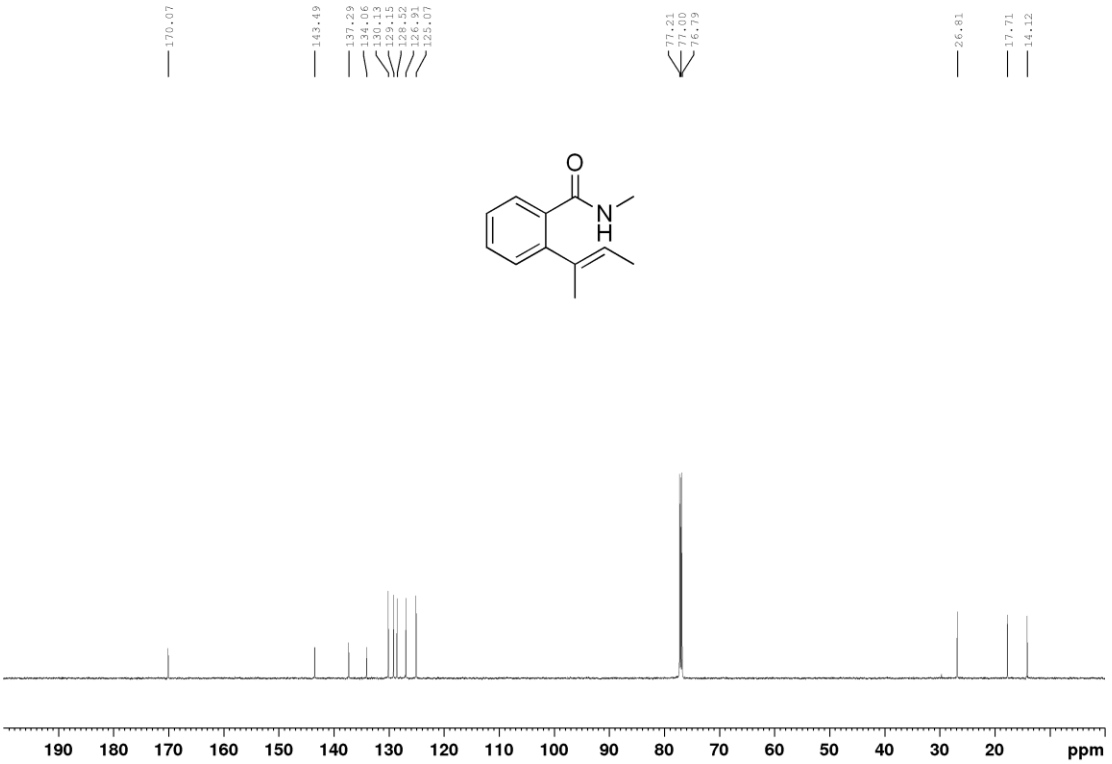
**(*E*)-3-(1,2-Diphenylvinyl)-*N*-methylthiophene-2-carboxamide (4h) (600 MHz)**



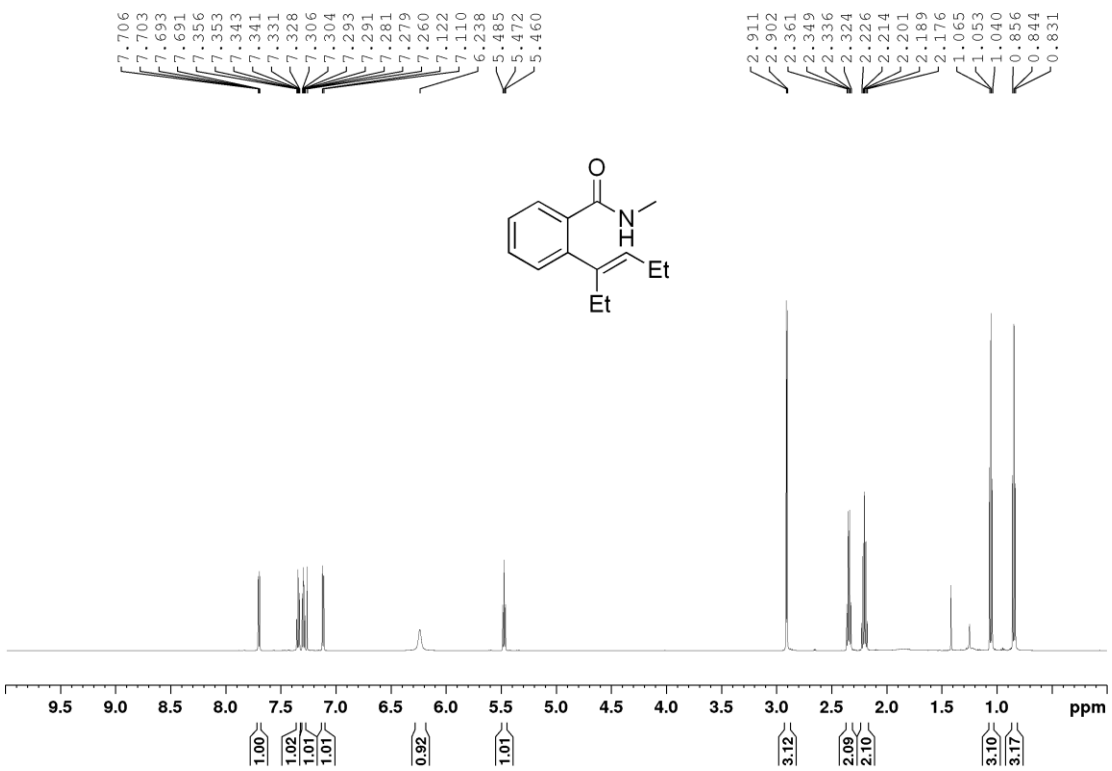



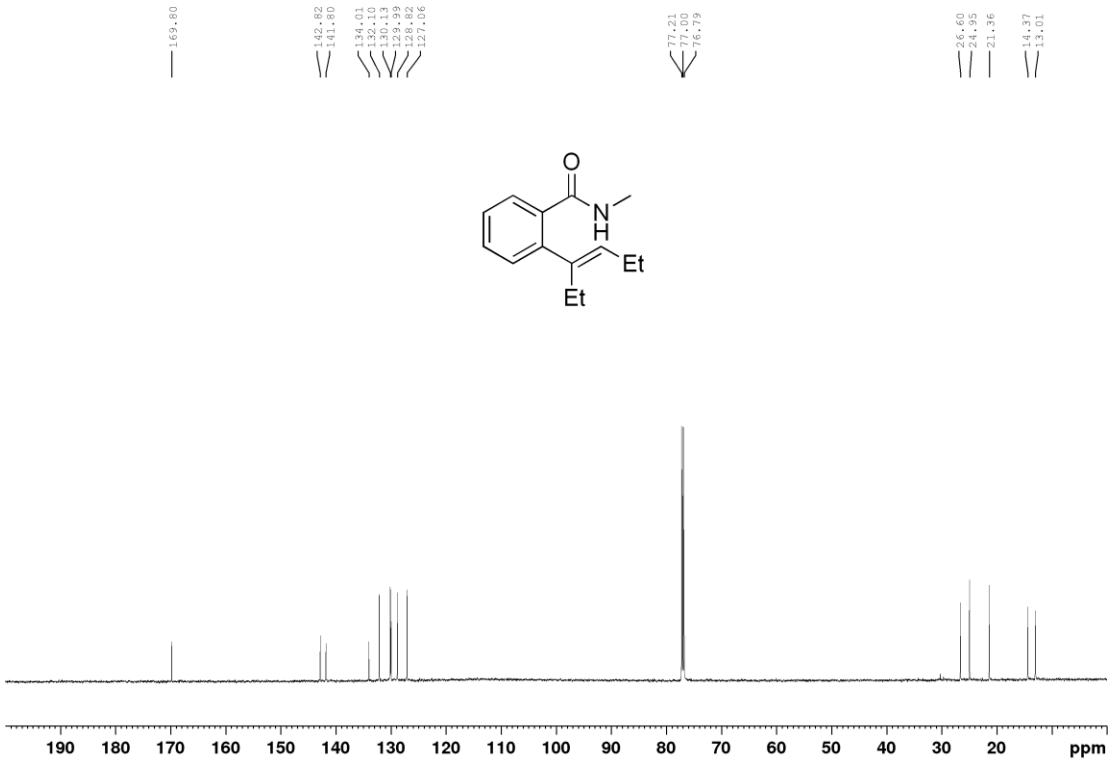
**(*E*)-2-(But-2-en-2-yl)-*N*-methylbenzamide (4i) (600 MHz)**



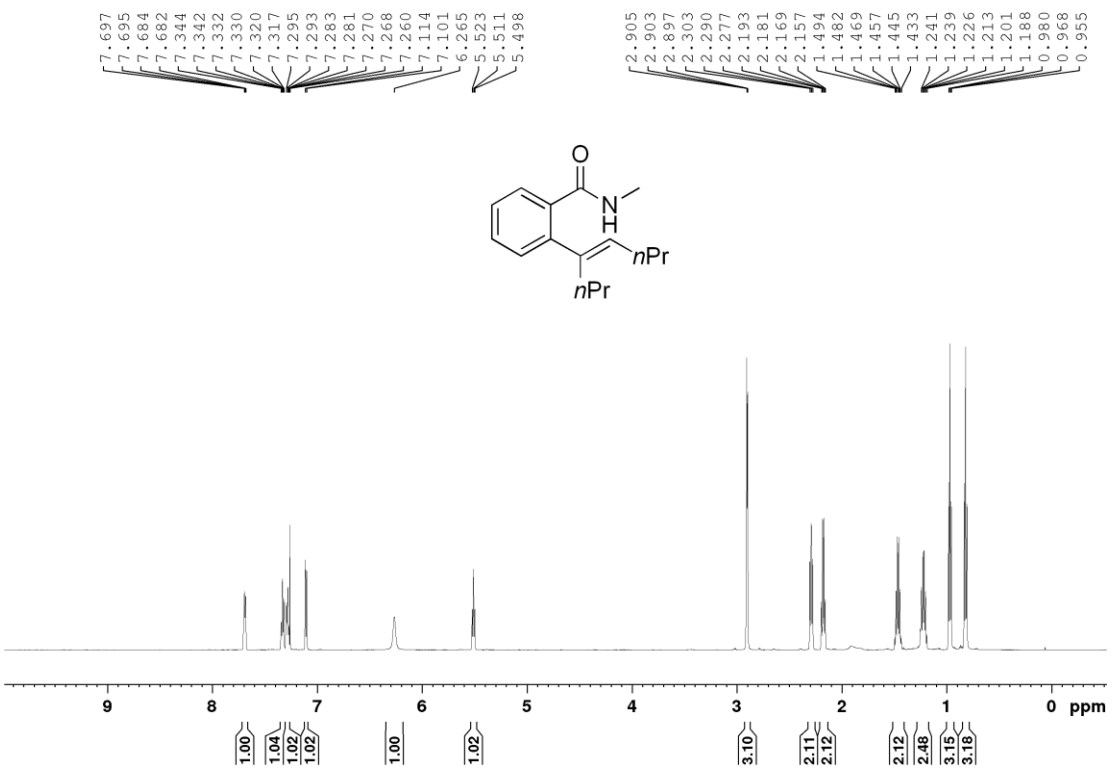



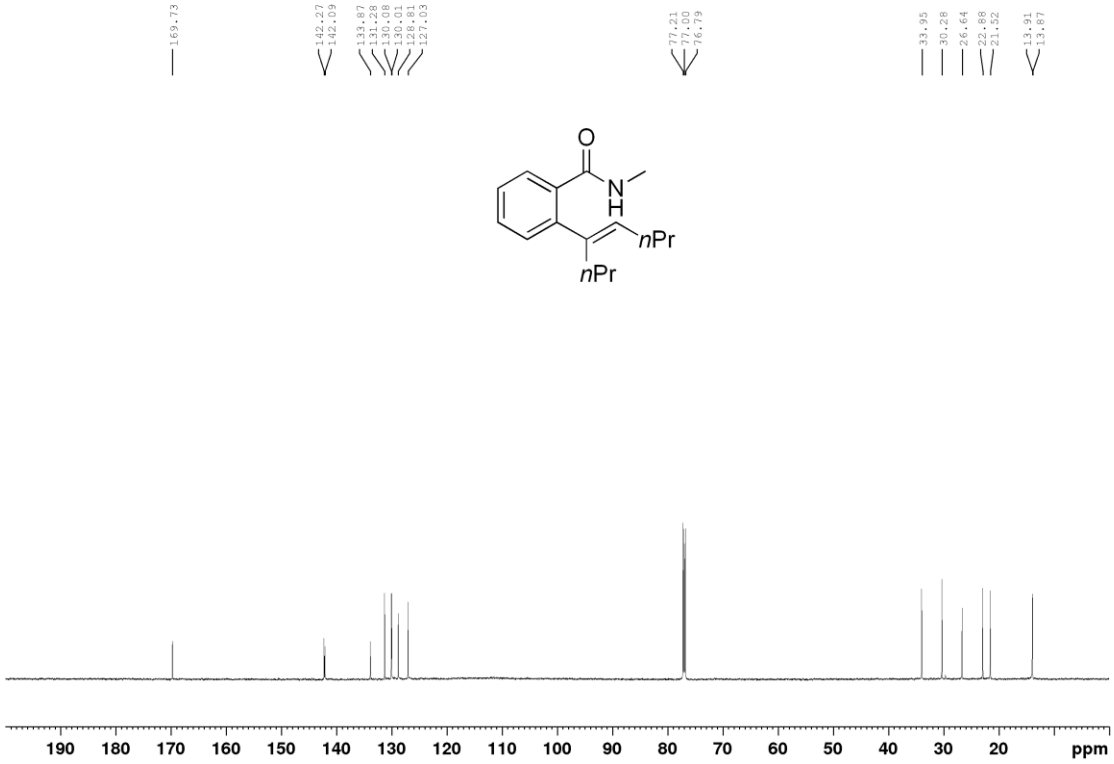
**(*E*)-2-(Hex-3-en-3-yl)-*N*-methylbenzamide (4j) (600 MHz)**



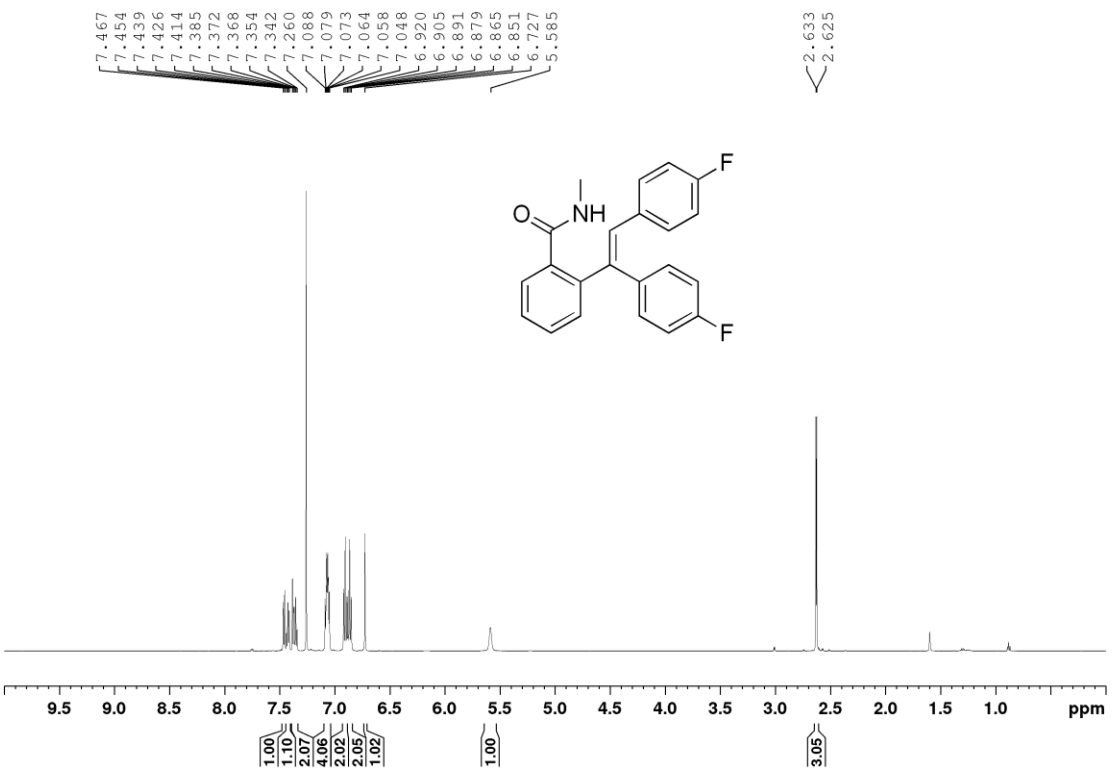



**(*E*)-*N*-Methyl-2-(oct-4-en-4-yl)benzamide (4k) (600 MHz)**

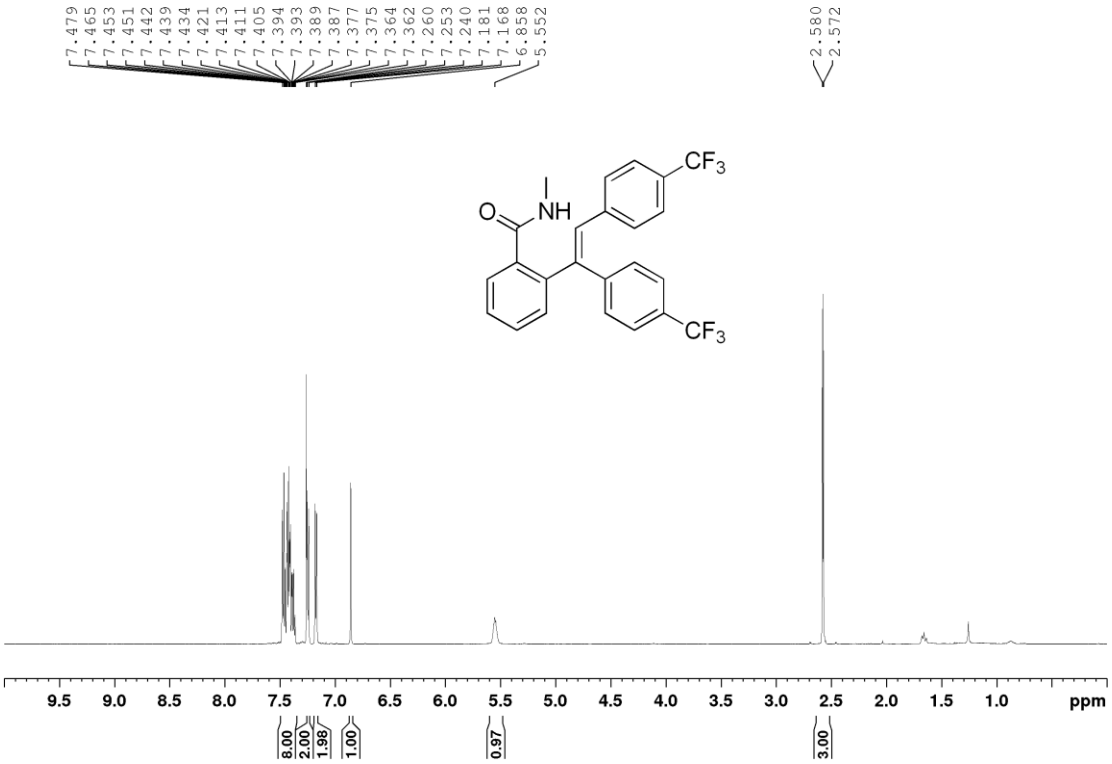


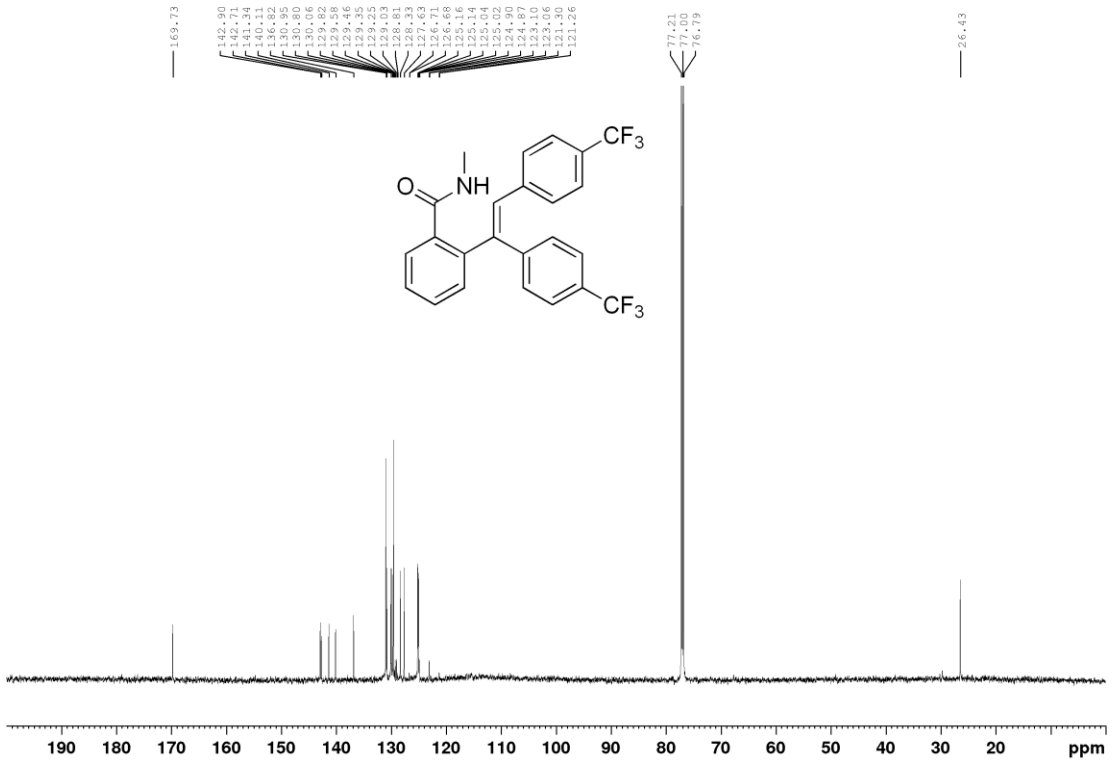



**(*E*)-2-(1,2-Bis(4-fluorophenyl)vinyl)-*N*-methylbenzamide (4l) (600 MHz)**

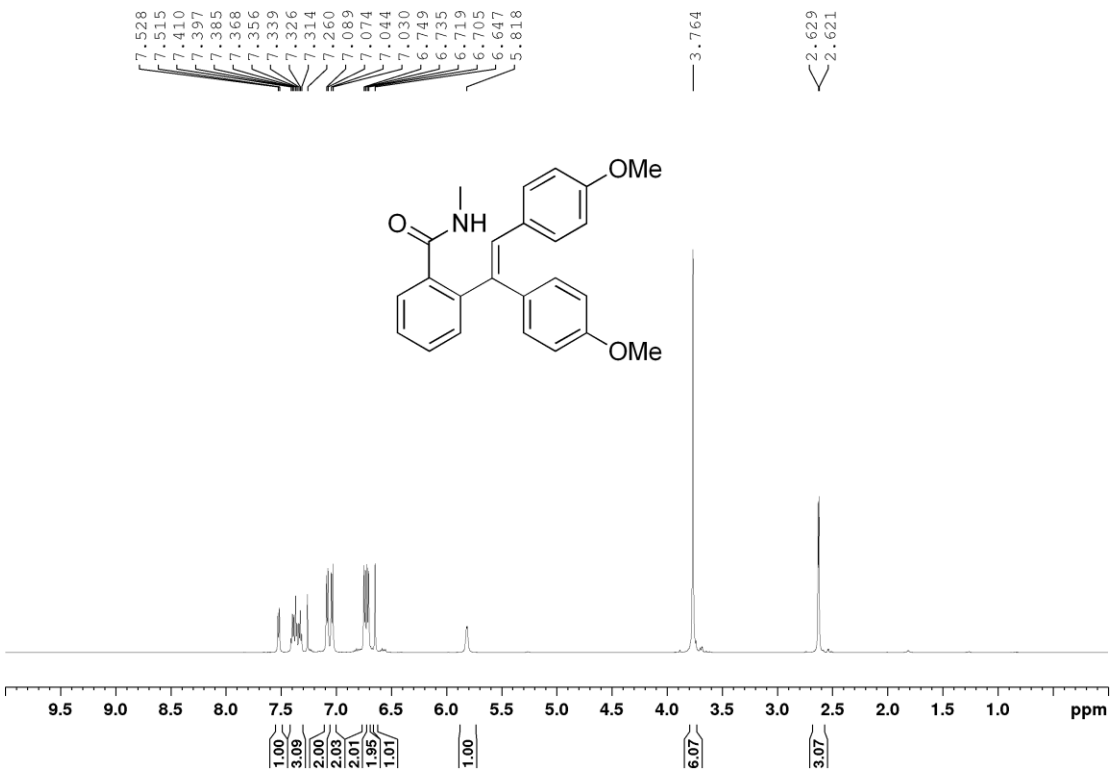


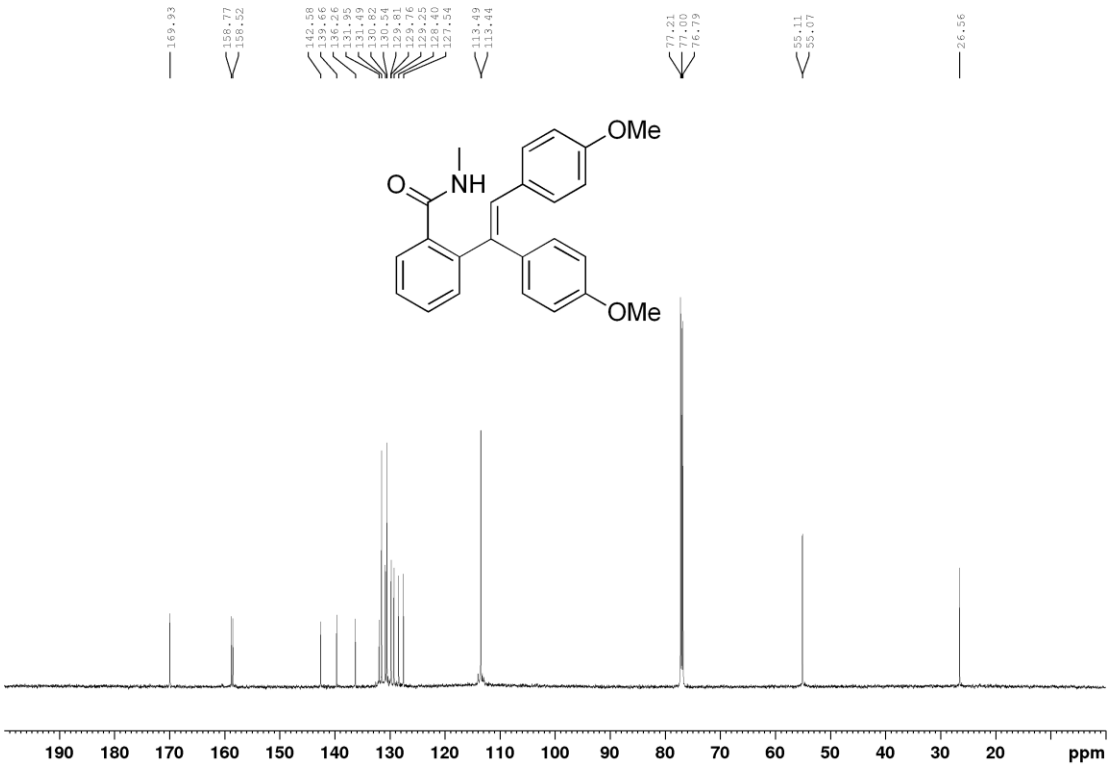

**(*E*)-2-(1,2-Bis(4-(trifluoromethyl)phenyl)vinyl)-*N*-methylbenzamide (4m) (600 MHz)**



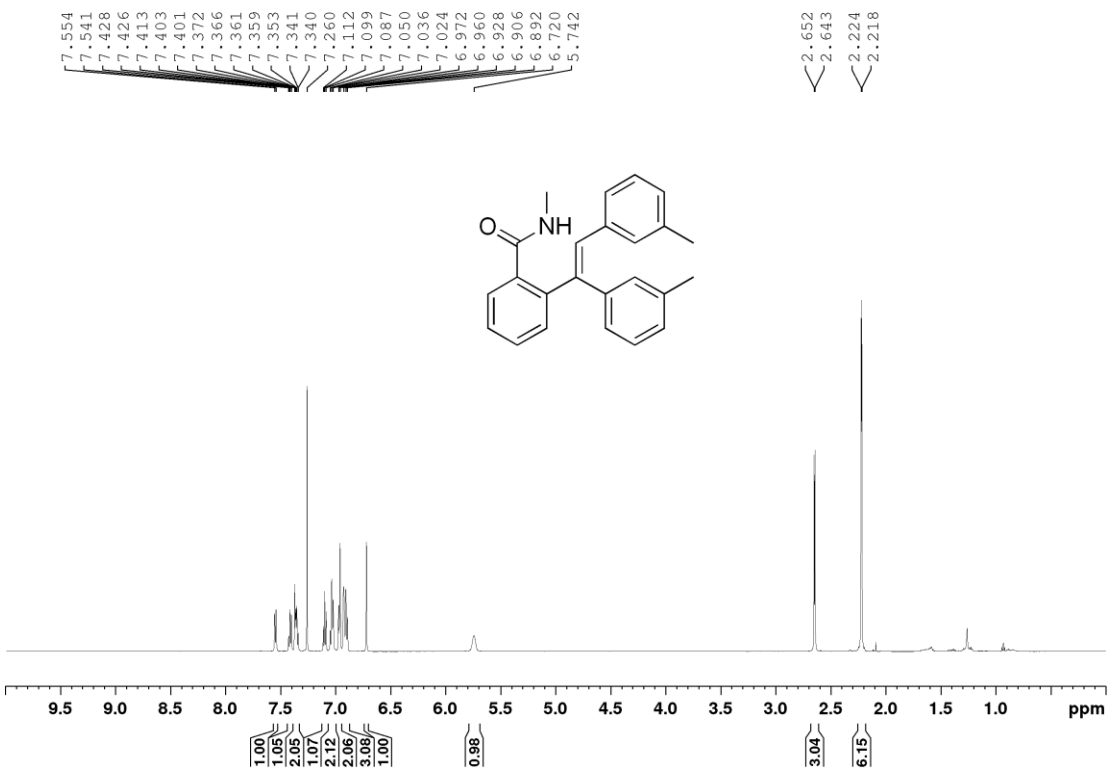



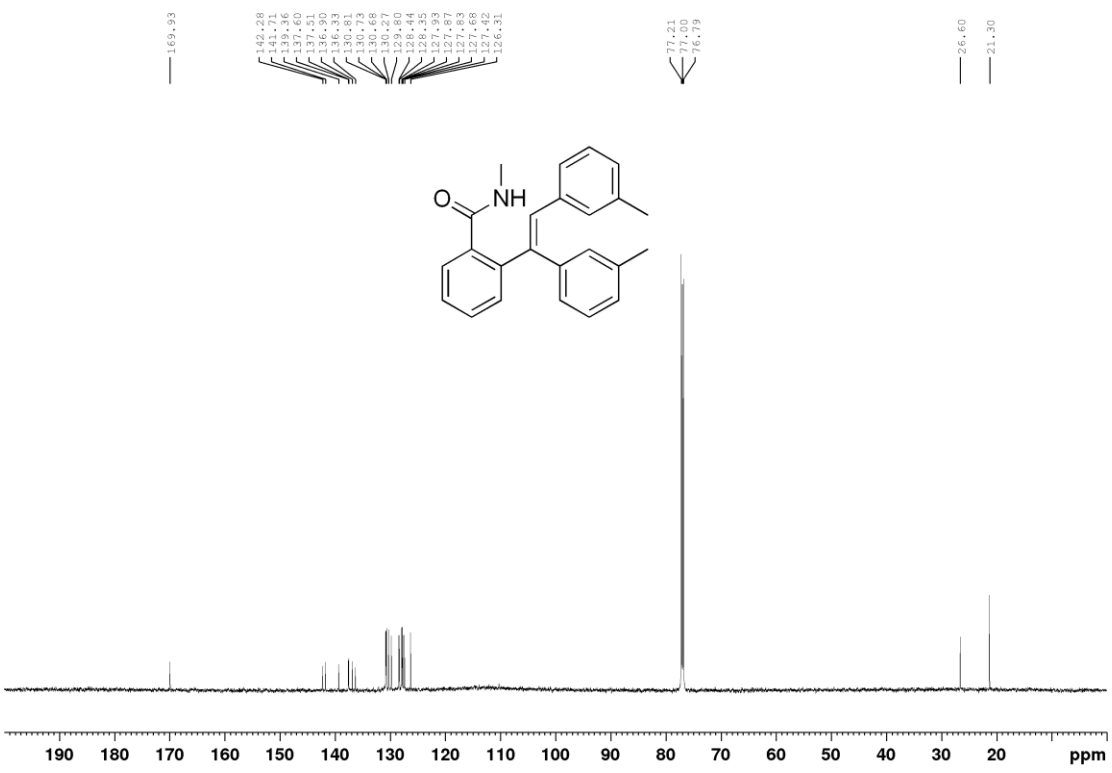
**(*E*)-2-(1,2-Bis(4-methoxyphenyl)vinyl)-*N*-methylbenzamide (4n) (600 MHz)**



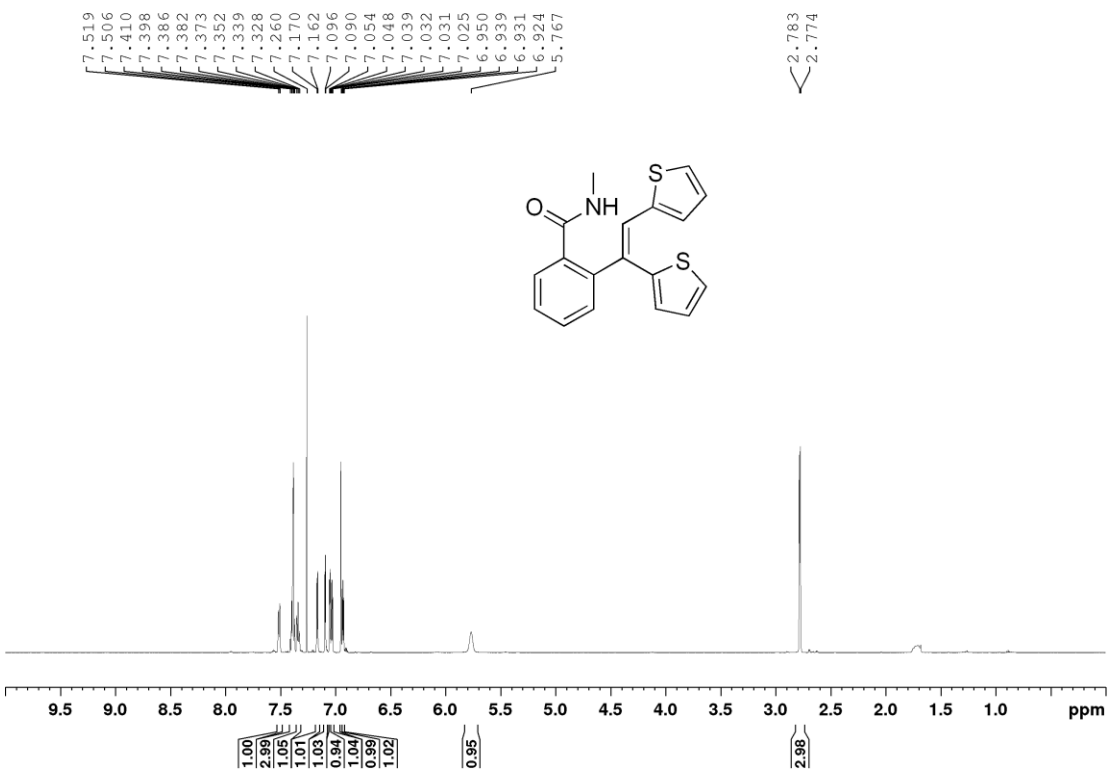



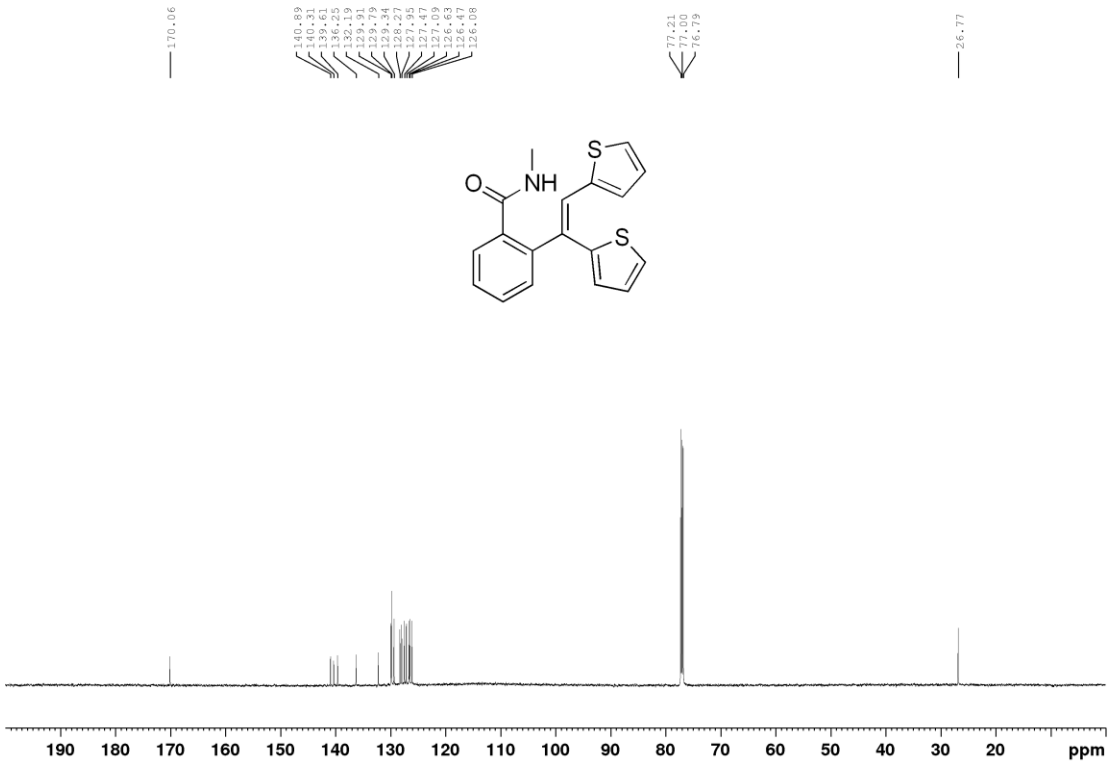
**(*E*)-2-(1,2-Di-*m*-tolylvinyl)-*N*-methylbenzamide (4o) (600 MHz)**



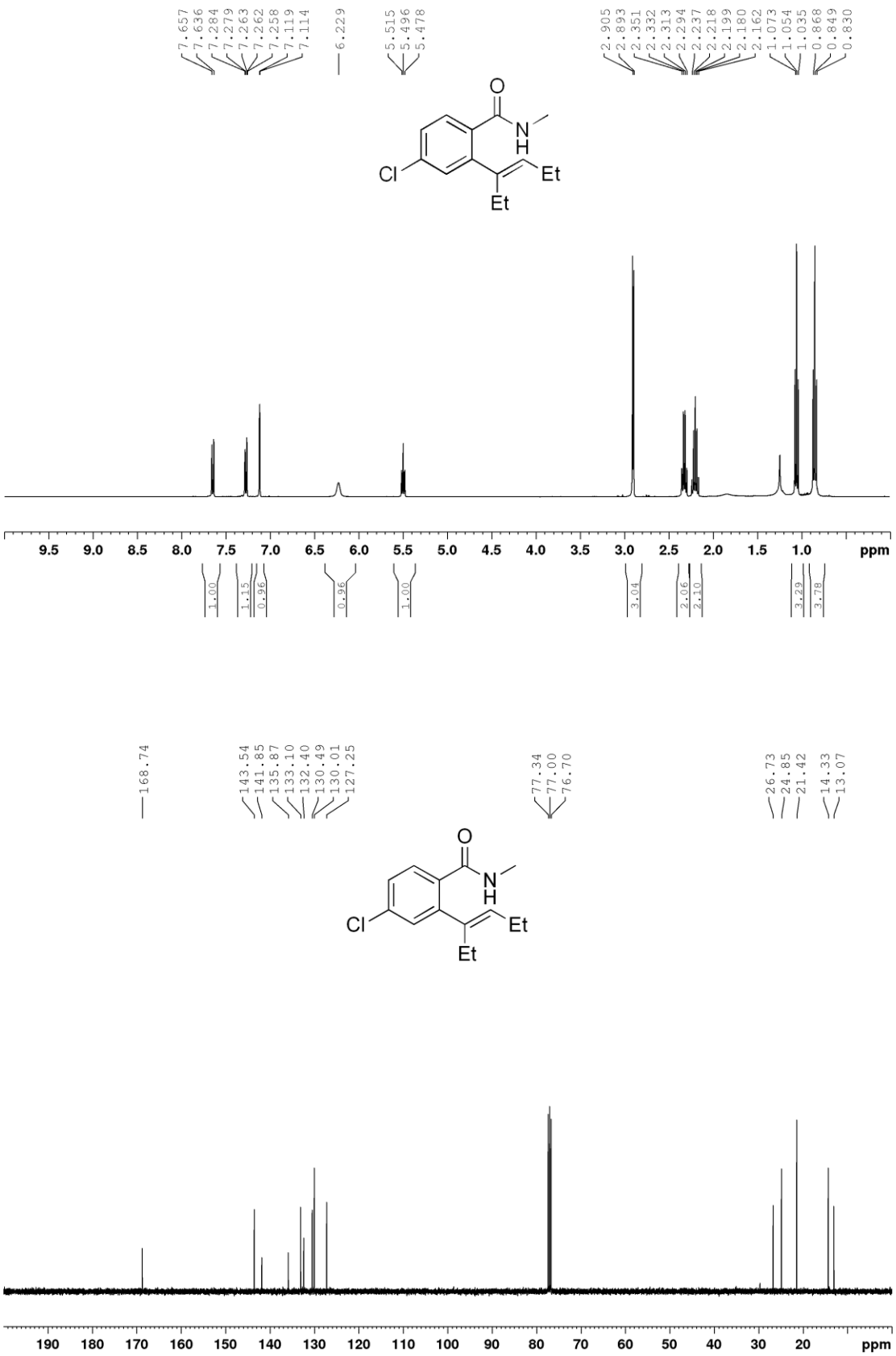



**((*Z*)-2-(1,2-Di(thiophen-2-yl)vinyl)-*N*-methylbenzamide (4p) (600 MHz)**

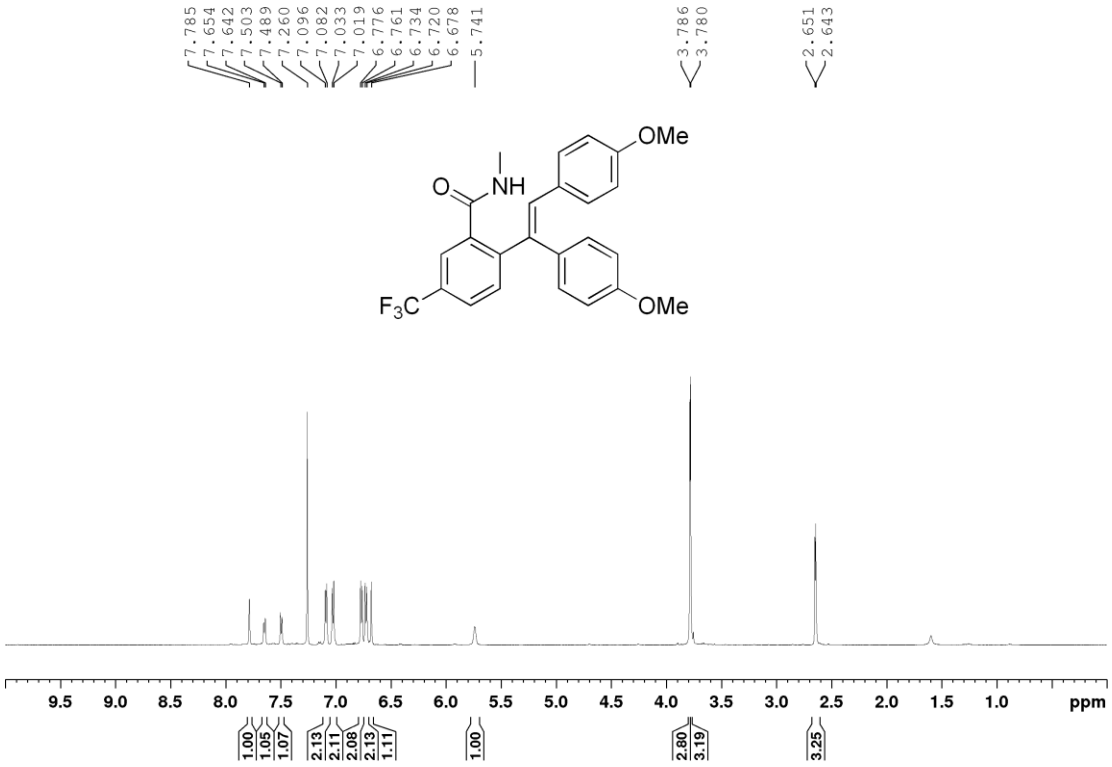


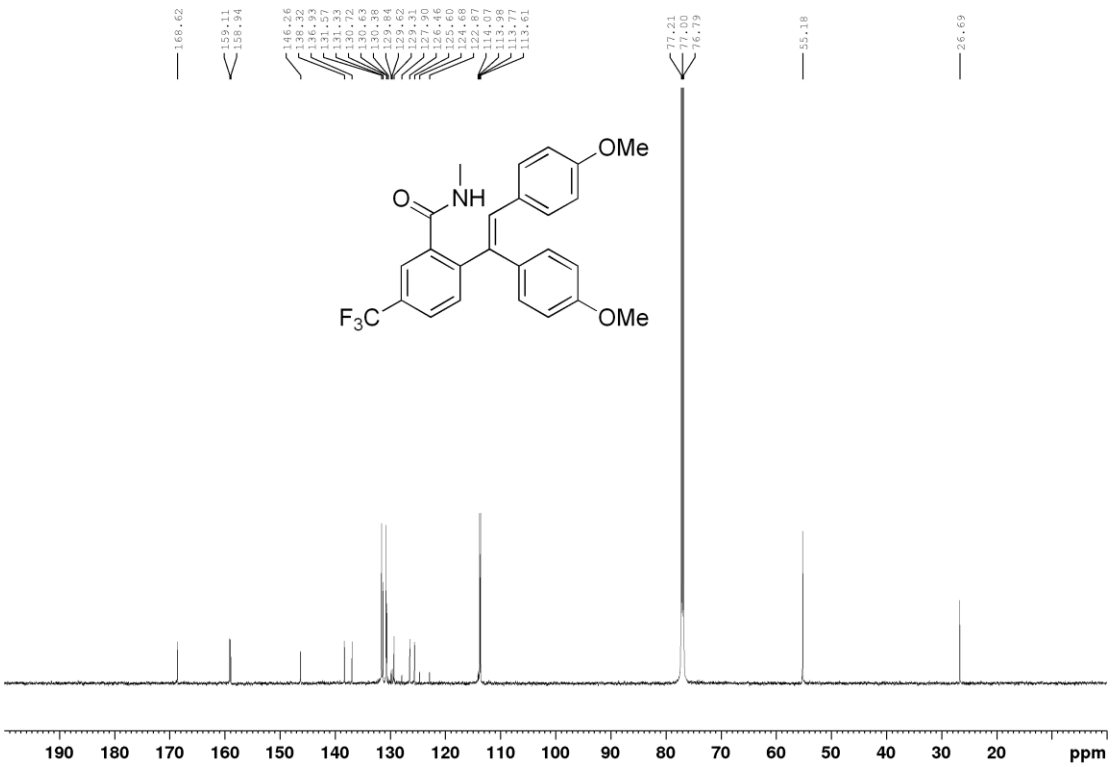



**(*E*)-4-Chloro-2-(hex-3-en-3-yl)-*N*-methylbenzamide (4q) (400 MHz)**

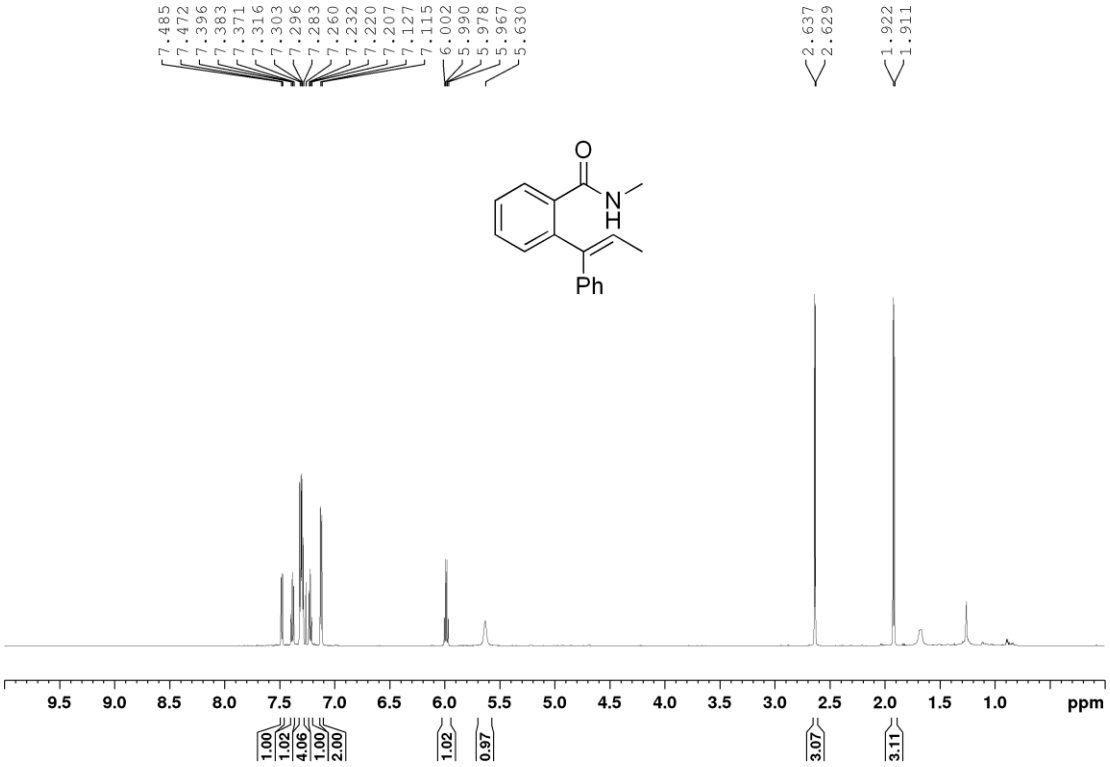


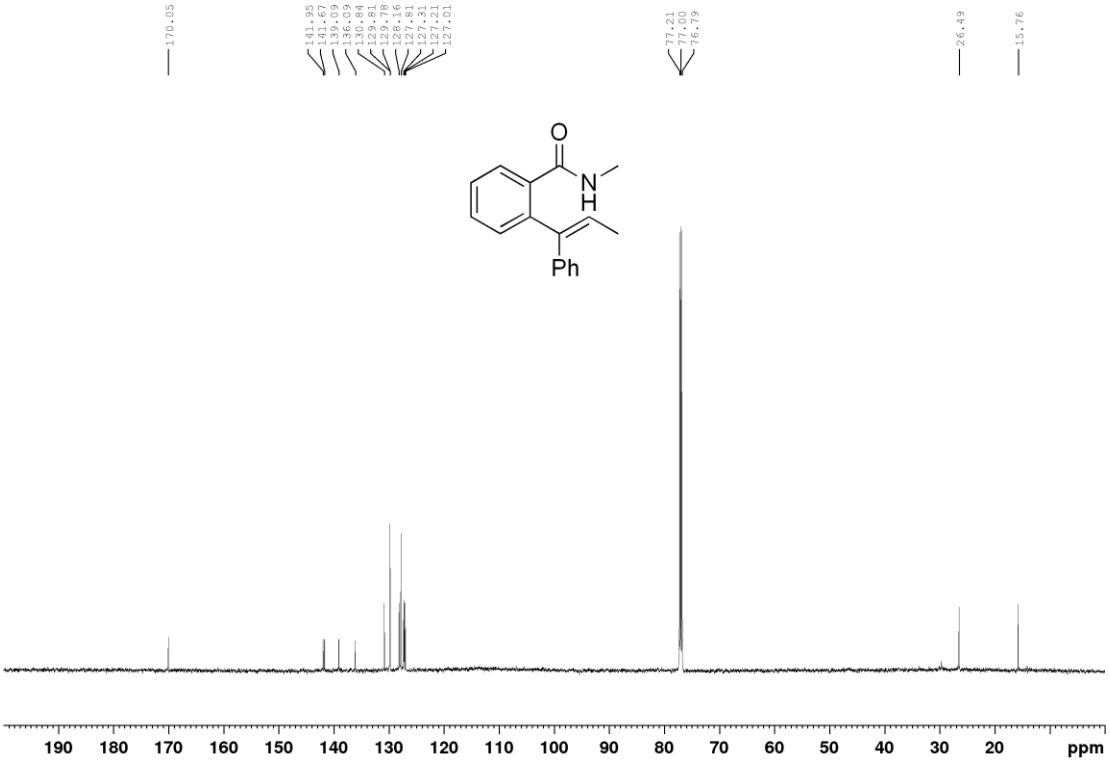
**(*E*)-2-(1,2-Bis(4-methoxyphenyl)vinyl)-*N*-methyl-5-(trifluoromethyl)benzamide (4r) (600 MHz)**



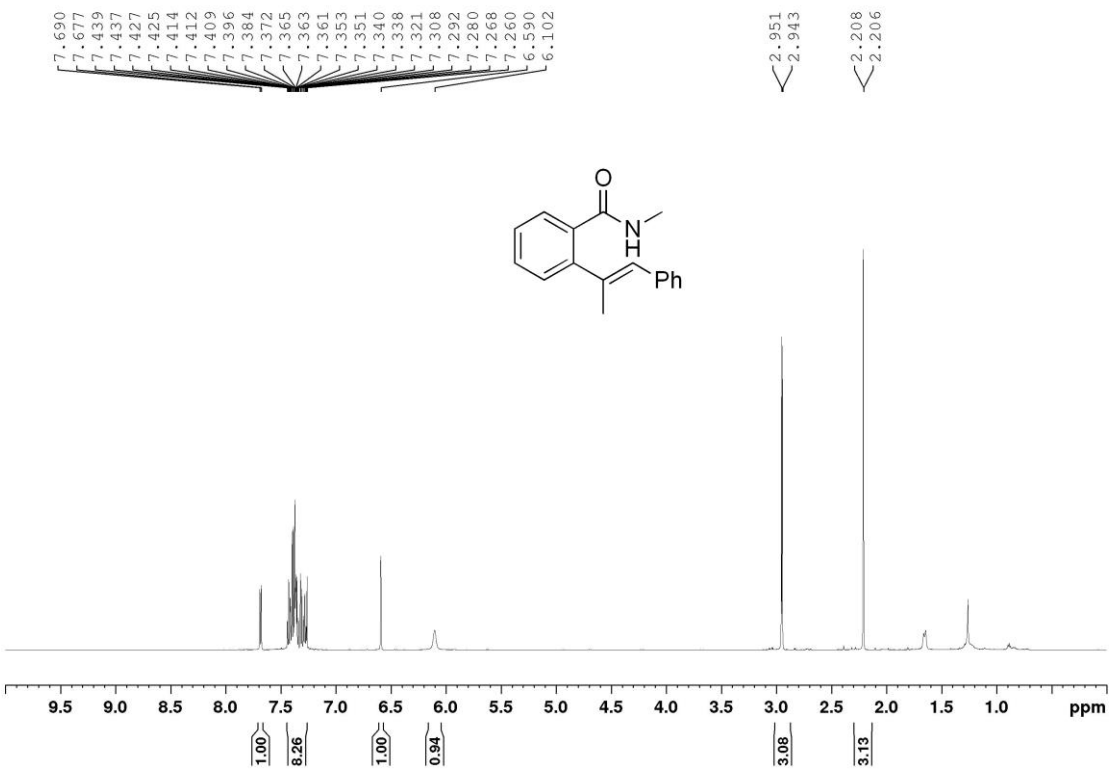



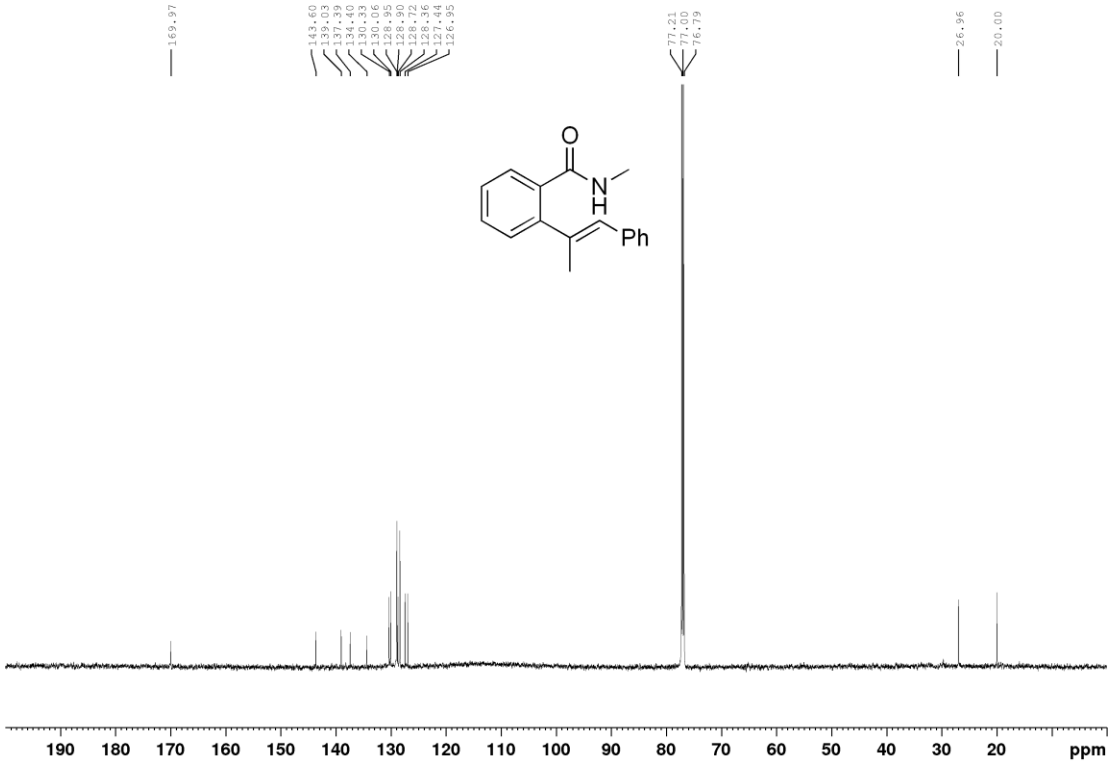
**(*E*)-*N*-Methyl-2-(1-phenylprop-1-en-1-yl)benzamide (4s) (600 MHz)**



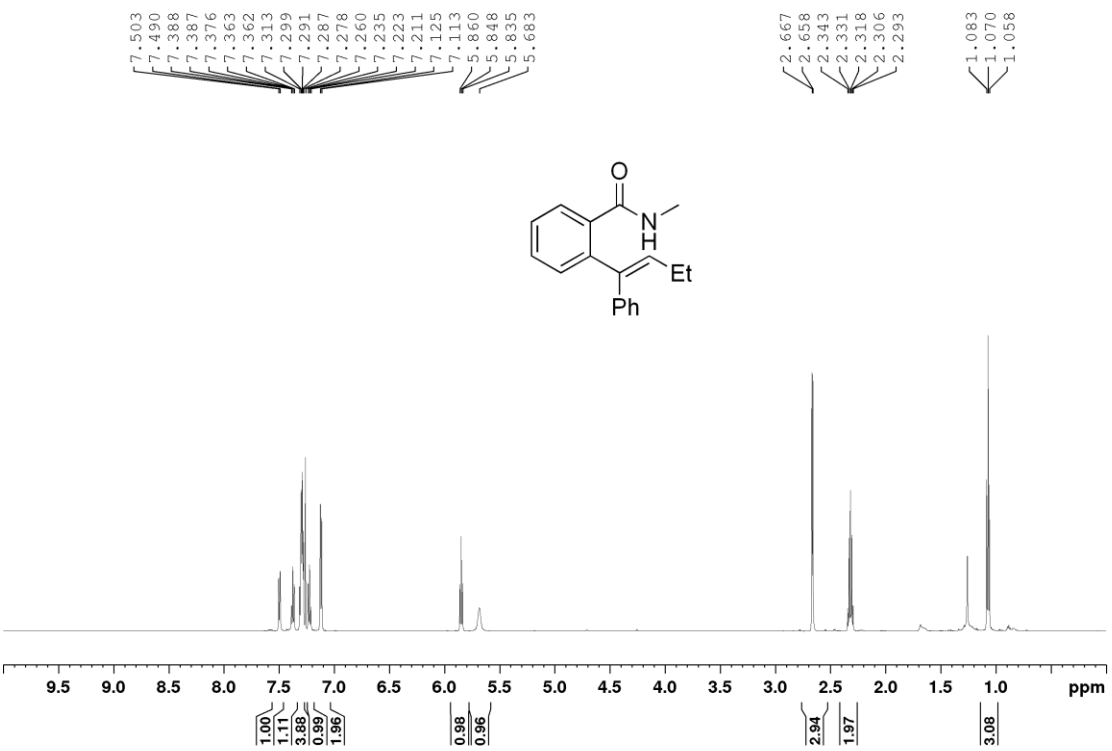



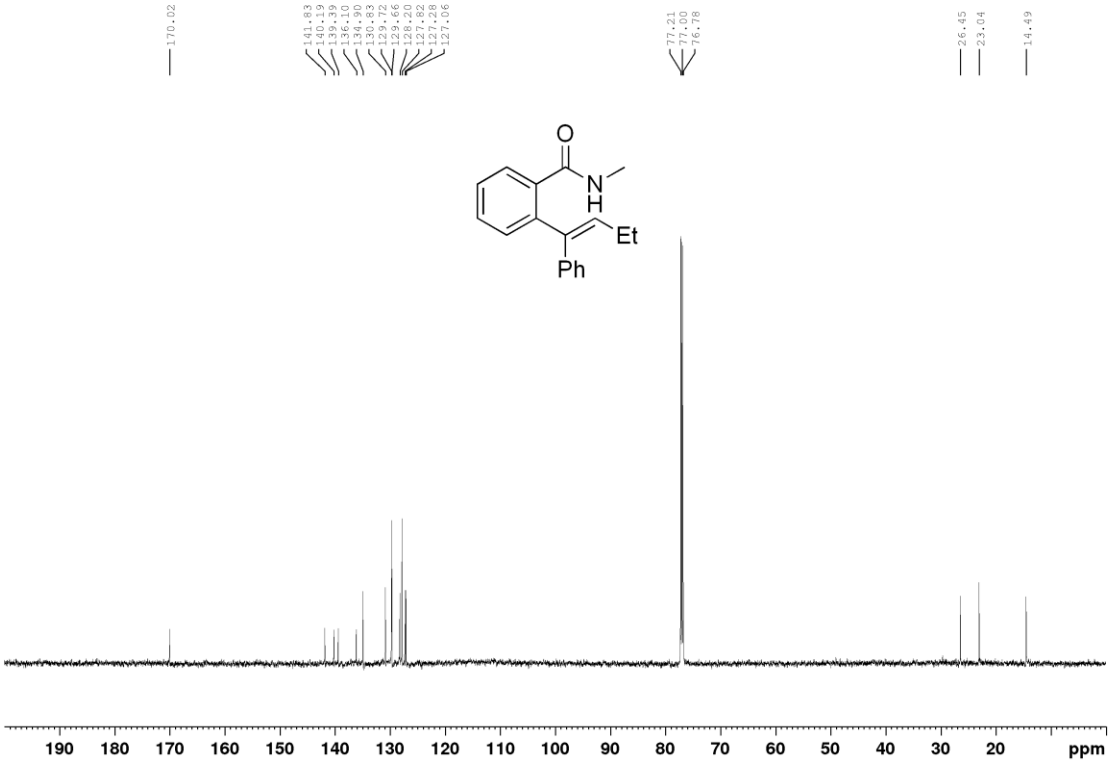
**(*E*)-*N*-Methyl-2-(1-phenylprop-1-en-2-yl)benzamide (4s') (600 MHz)**



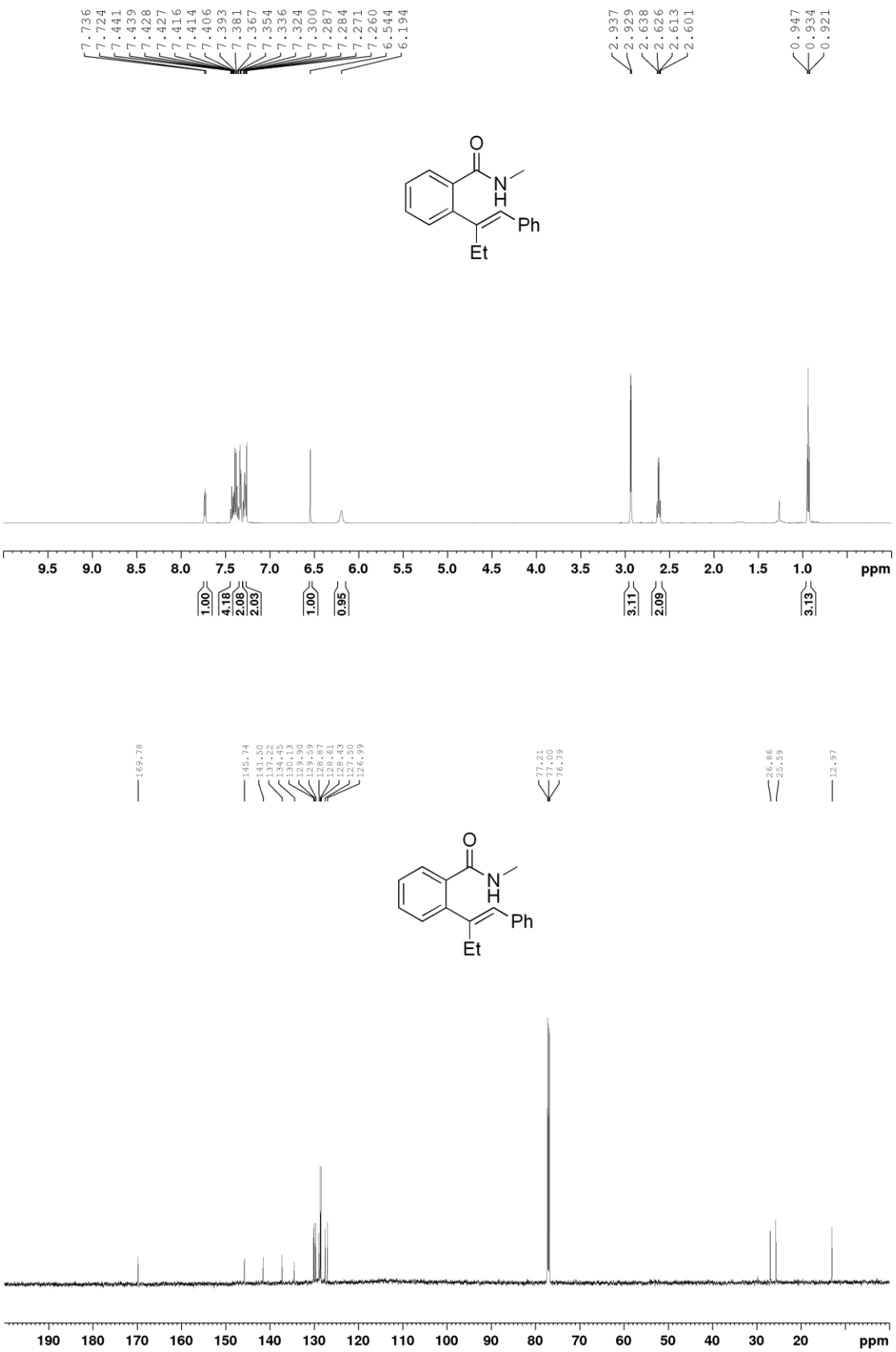



**(*E*)-*N*-Methyl-2-(1-phenylbut-1-en-1-yl)benzamide (4t) (600 MHz)**

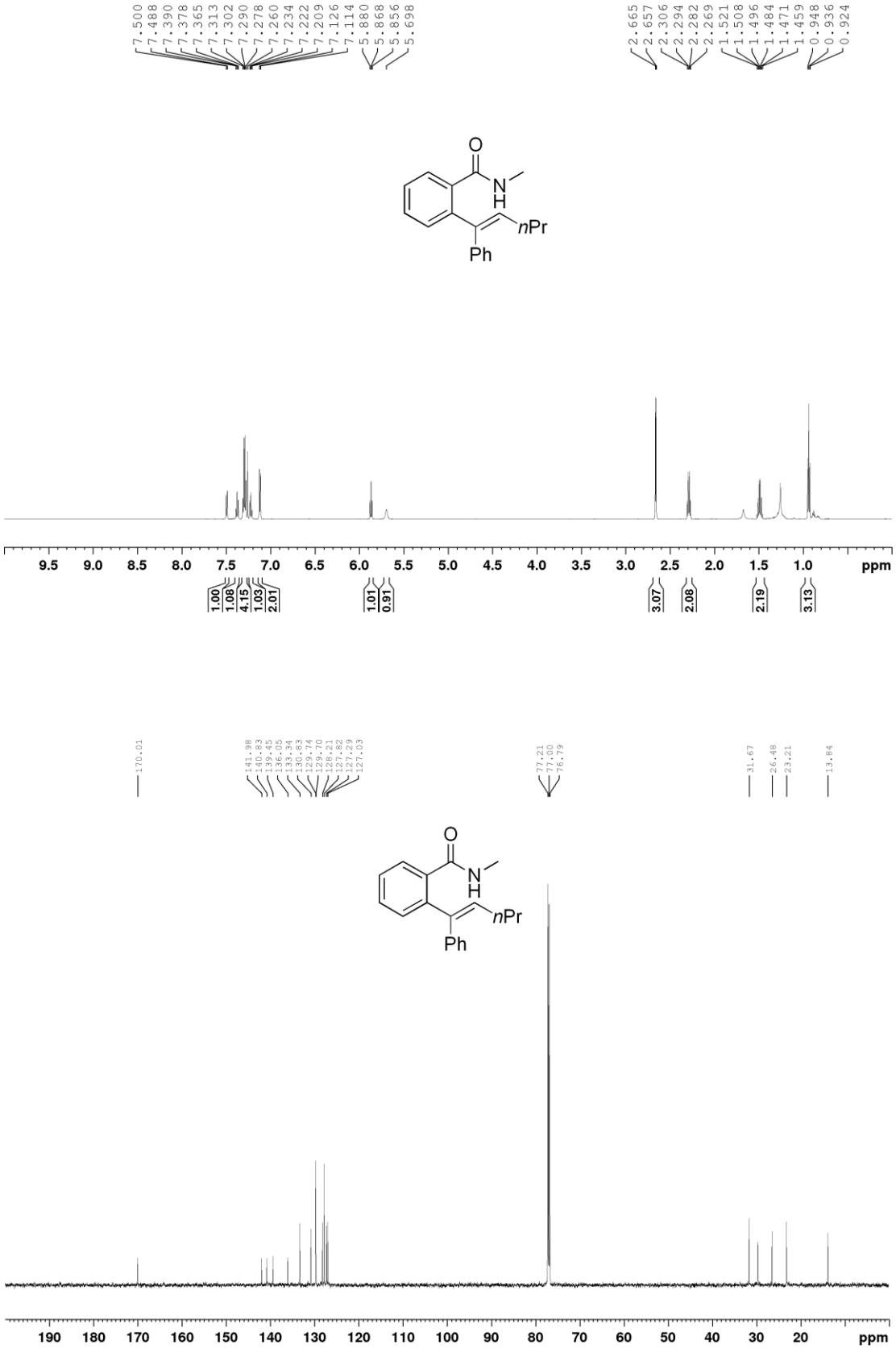


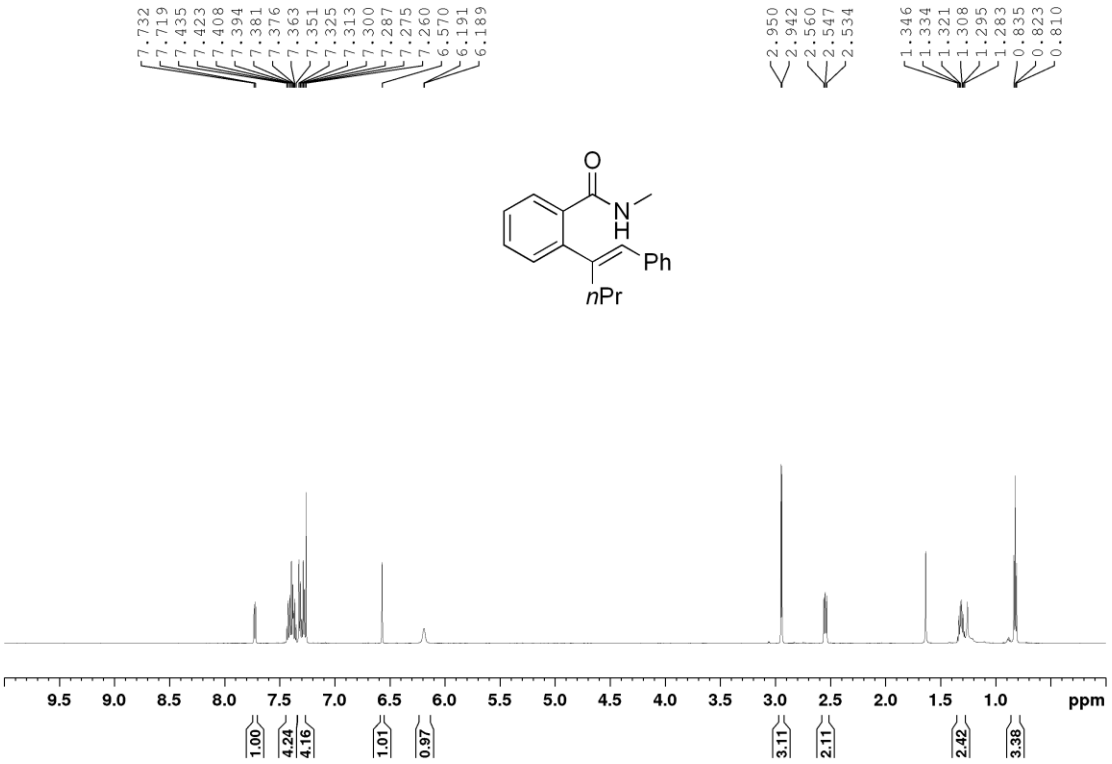
**(*E*)-*N*-Methyl-2-(1-phenylbut-1-en-2-yl)benzamide (4t') (600 MHz)**

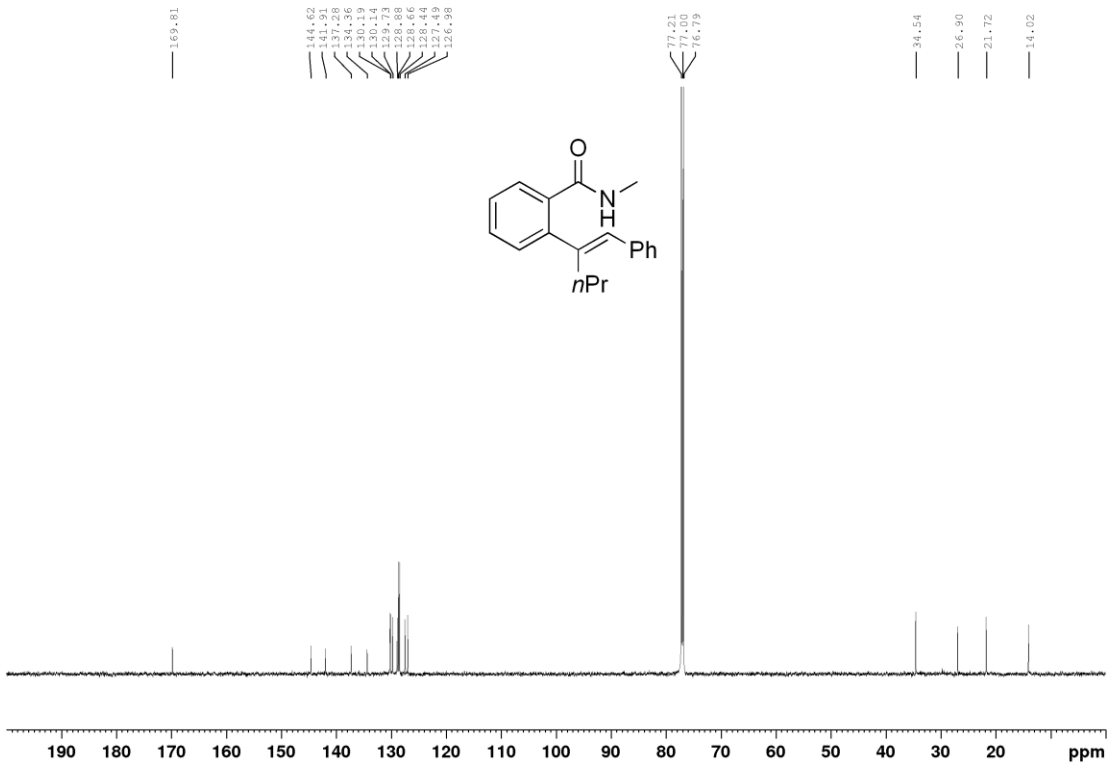


**(*E*)-*N*-Methyl-2-(1-phenylpent-1-en-1-yl)benzamide (4u) (600 MHz)**

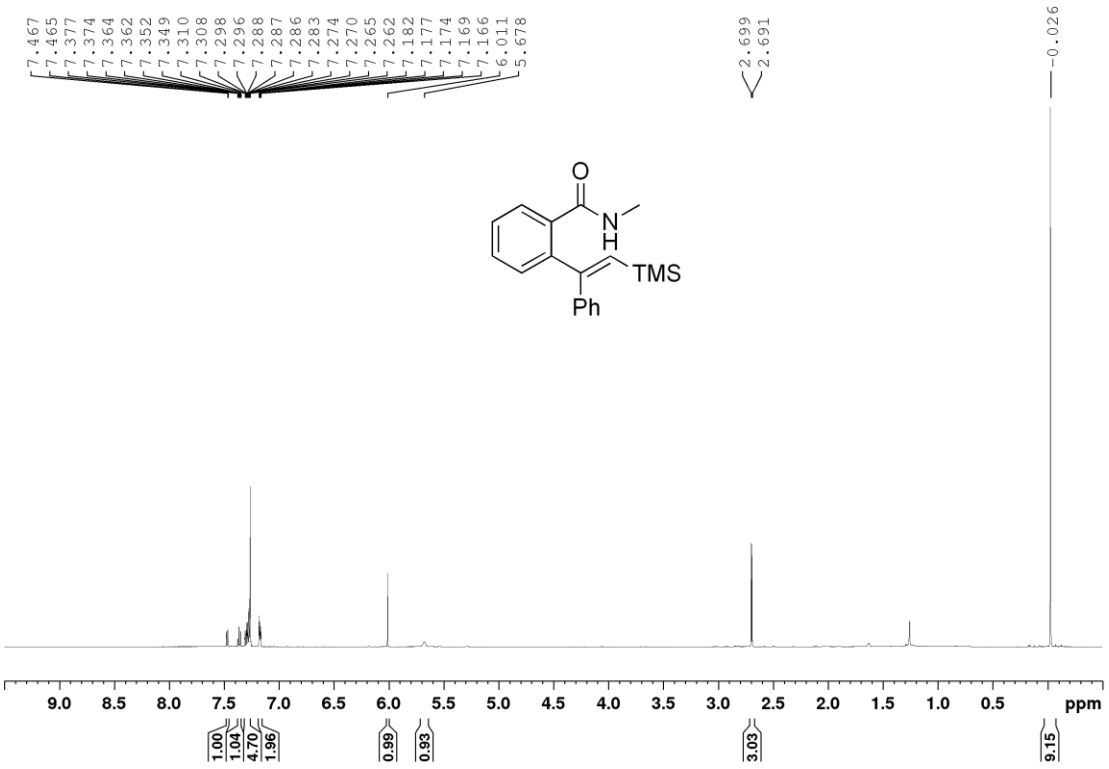


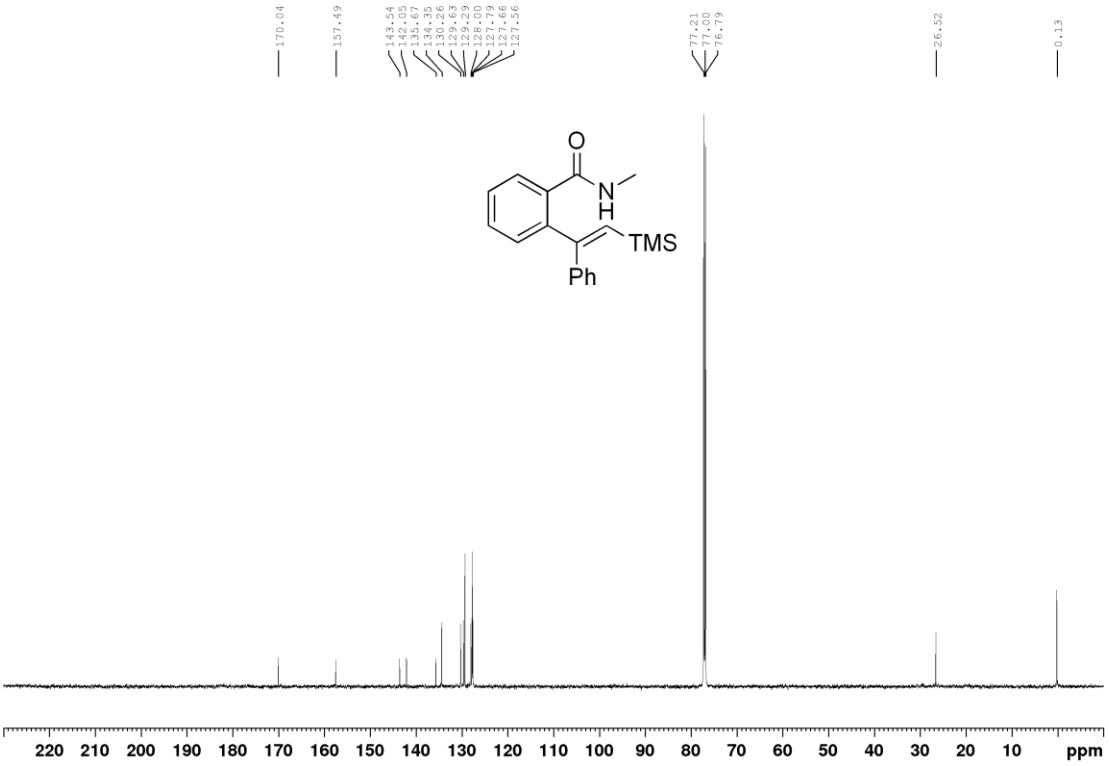
**(*E*)-*N*-Methyl-2-(1-phenylpent-1-en-2-yl)benzamide (4u') (600 MHz)**



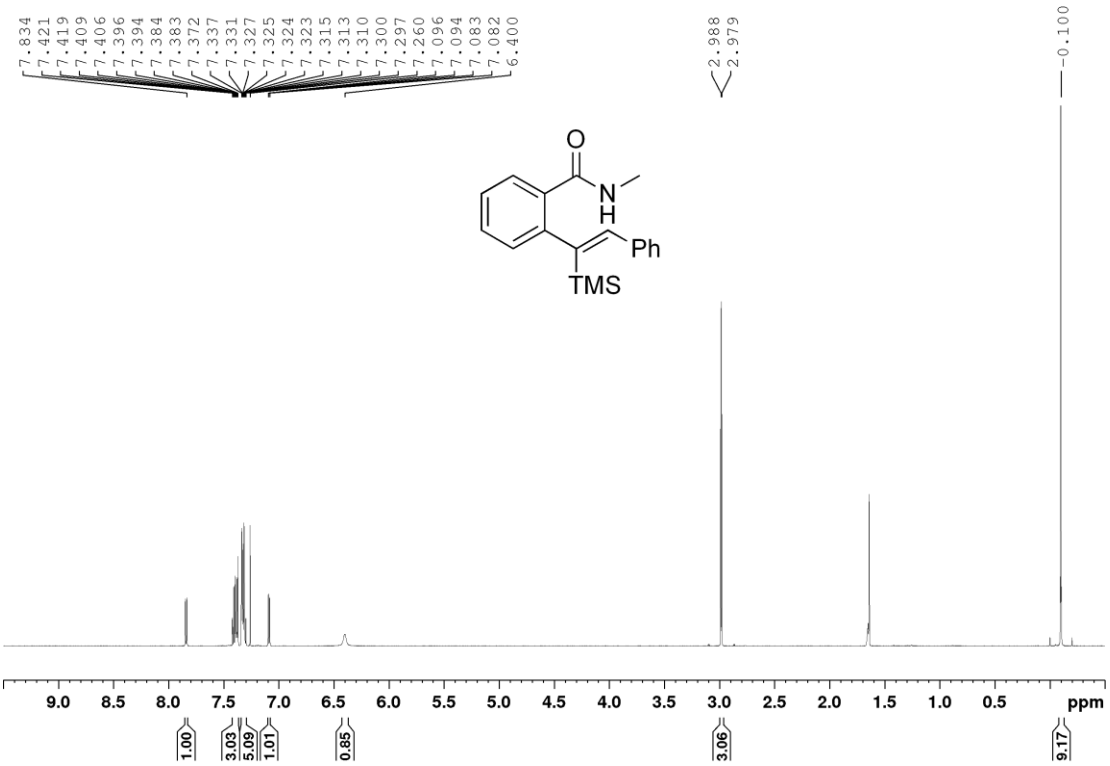



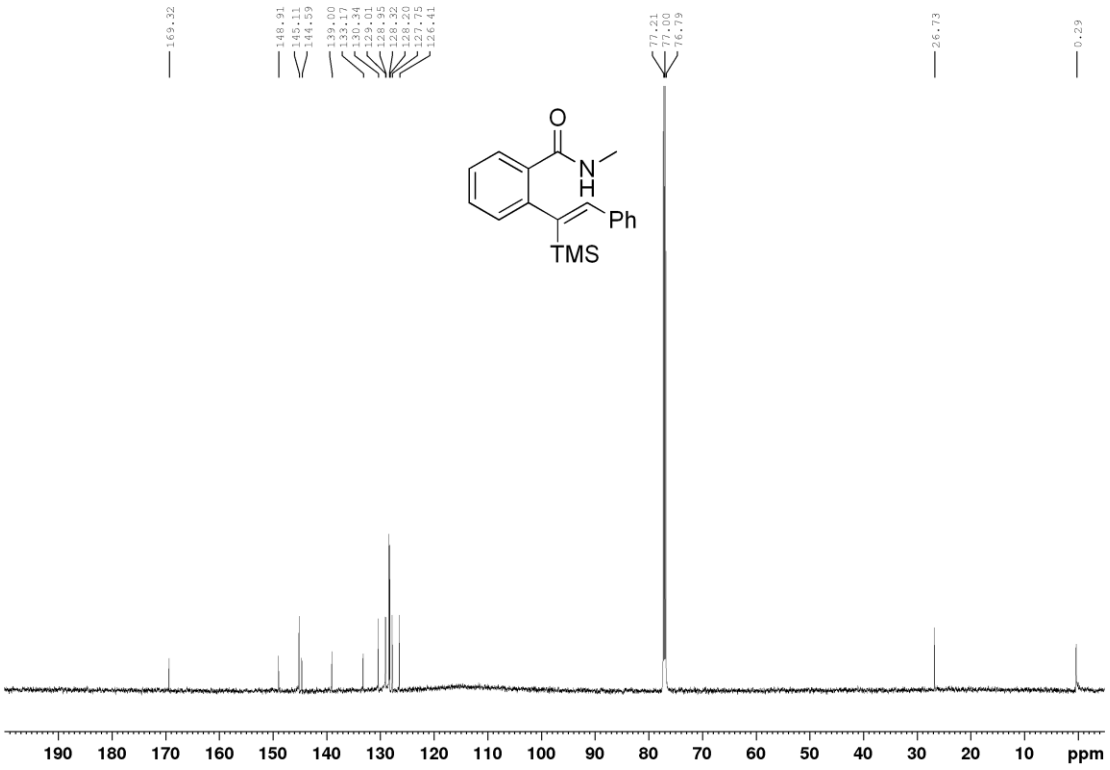
**(*E*)-*N*-Methyl-2-(1-phenyl-2-(trimethylsilyl)vinyl)benzamide (4v) (600 MHz)**



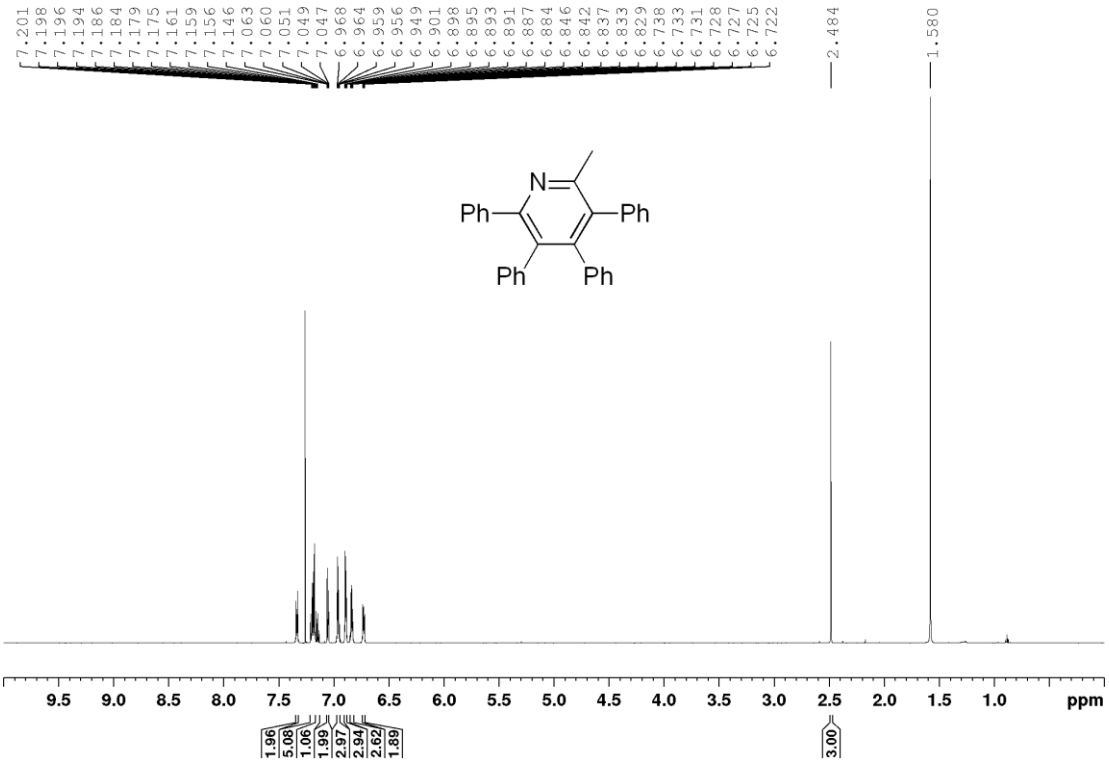



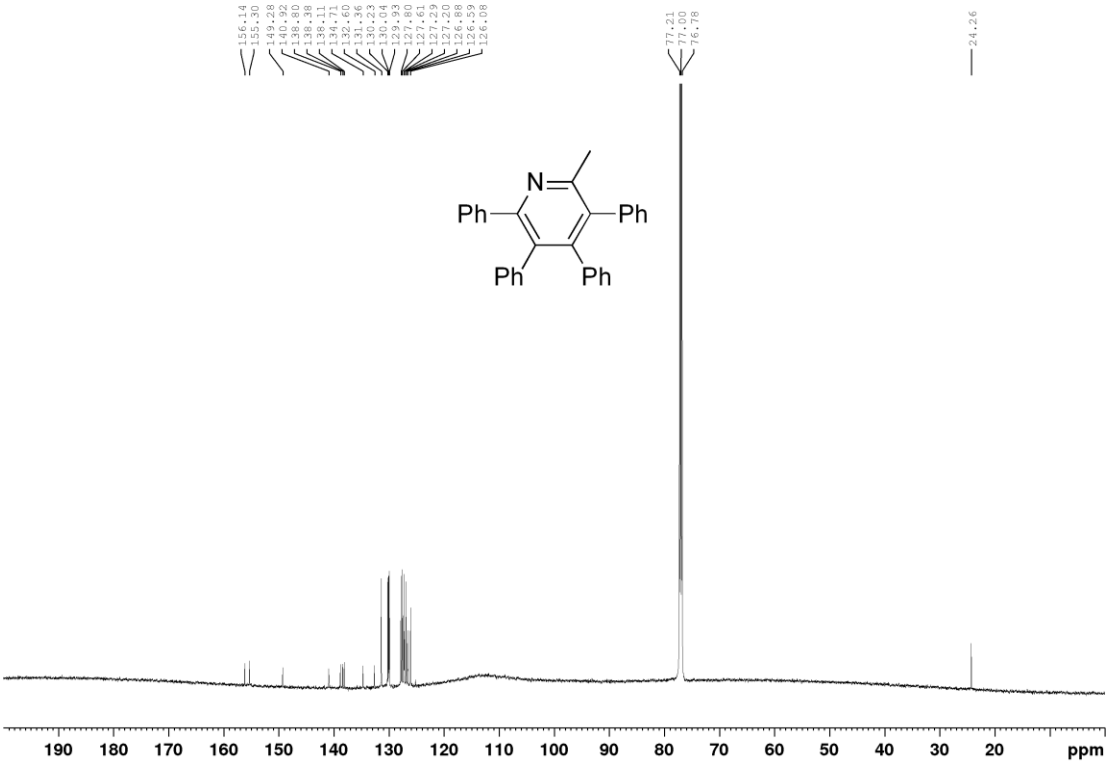
**(Z)-*N*-Methyl-2-(2-phenyl-1-(trimethylsilyl)vinyl)benzamide (4v') (600 MHz)**



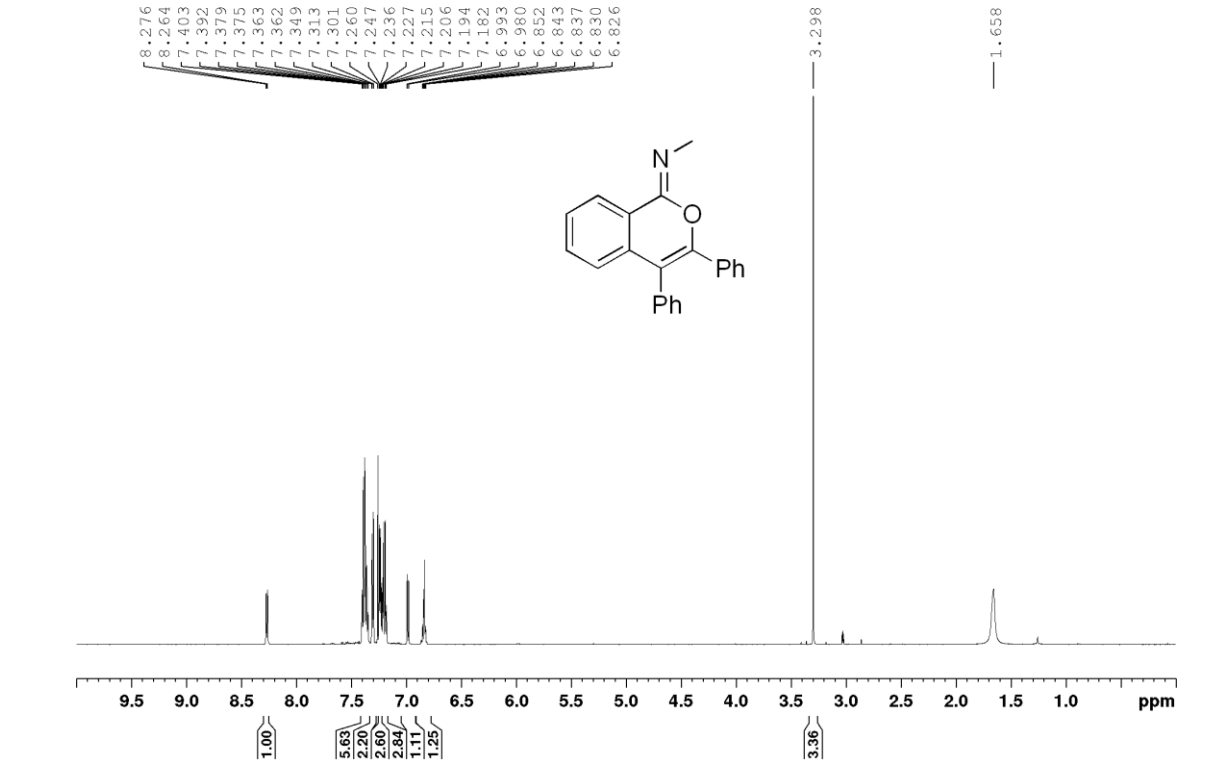



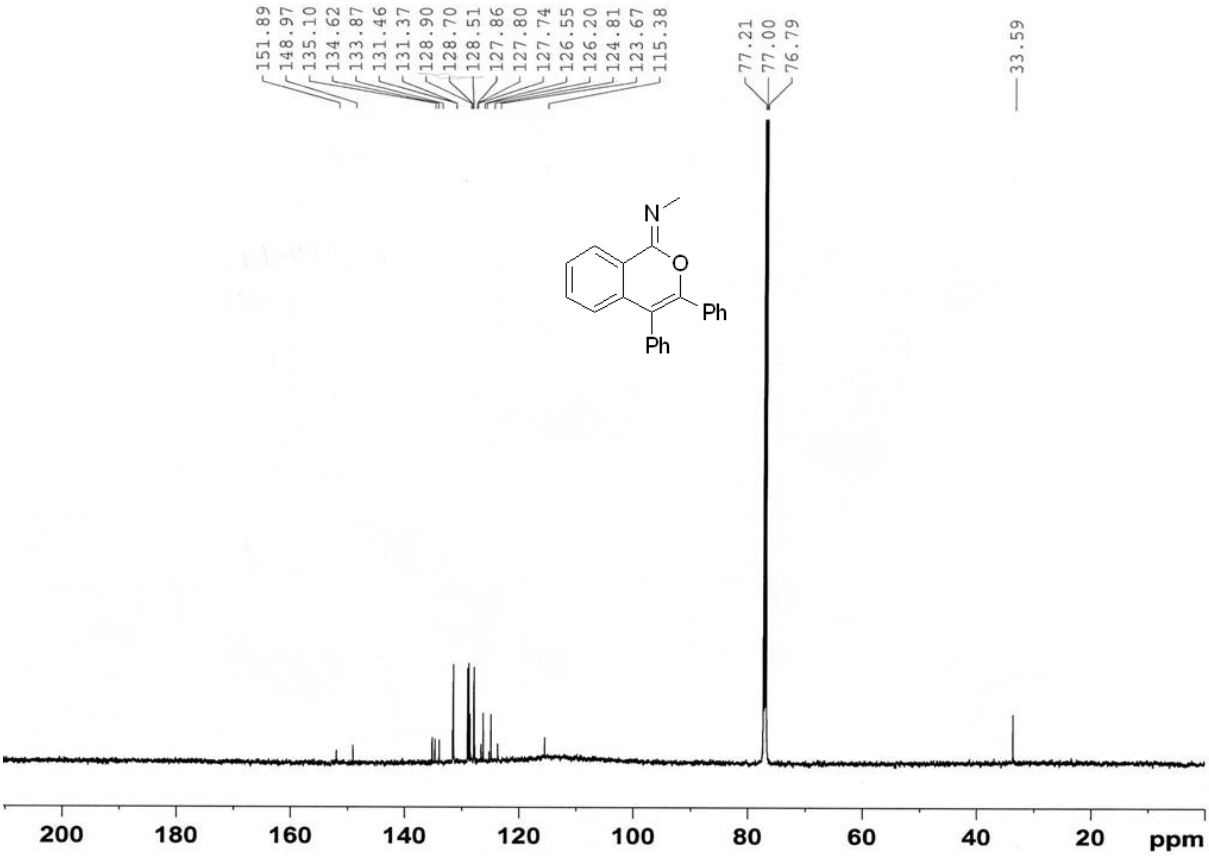
**2-Methyl-3,4,5,6-tetraphenylpyridine (5) (600 MHz)**



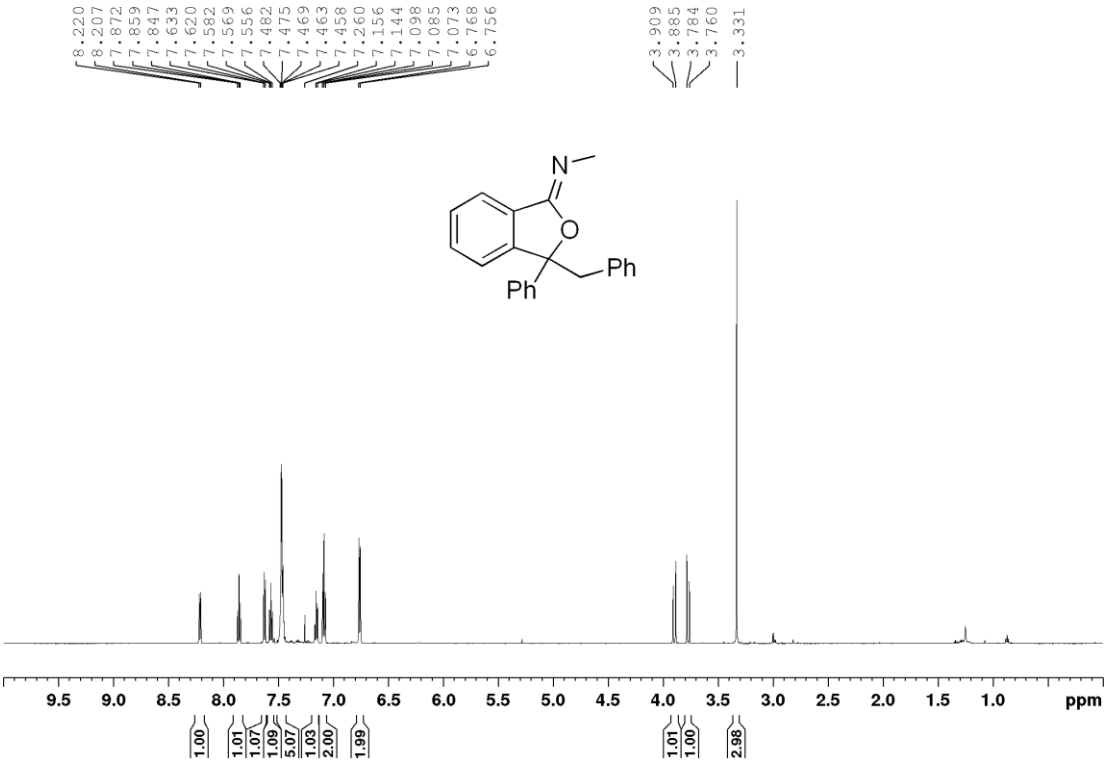



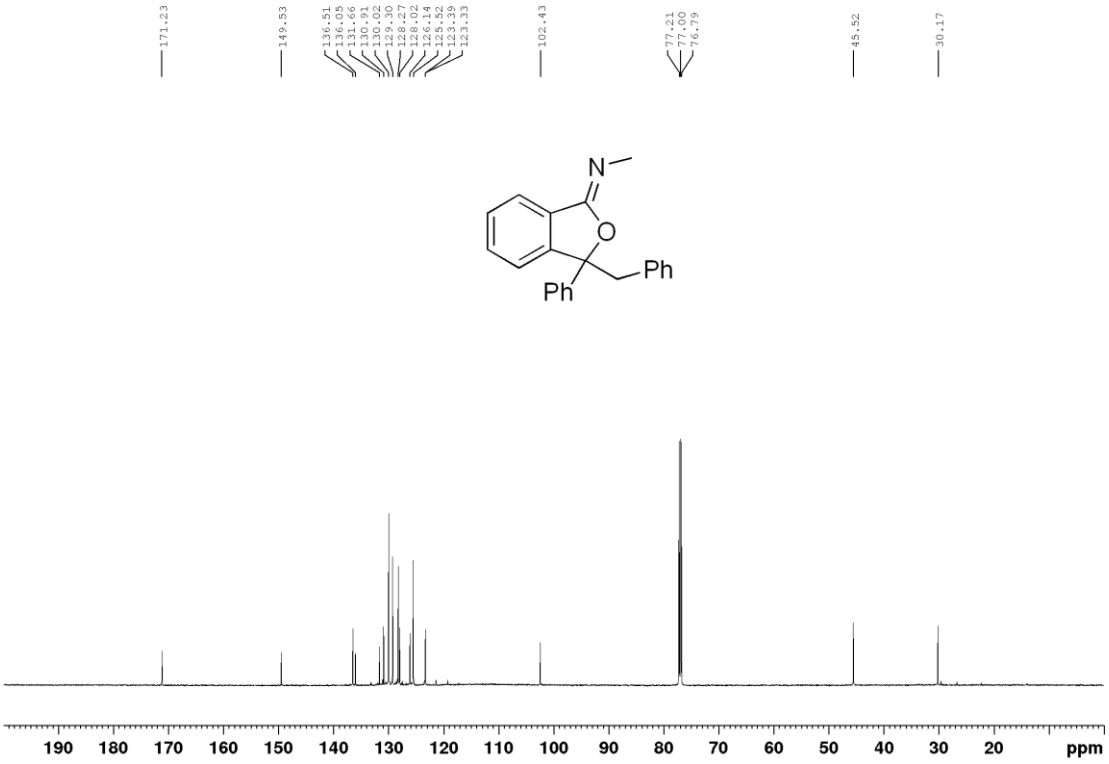
**(Z)-*N*-(3,4-Diphenyl-1*H*-isochromen-1-ylidene)methanamine (6) (600 MHz)**



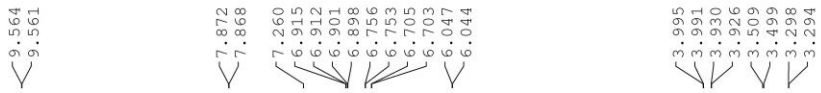
**(*Z*)-*N*-(3-Benzyl-3-phenylisobenzofuran-1(3*H*)-ylidene)methanamine (7) (600 MHz)**

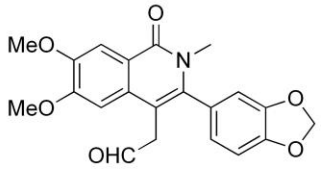


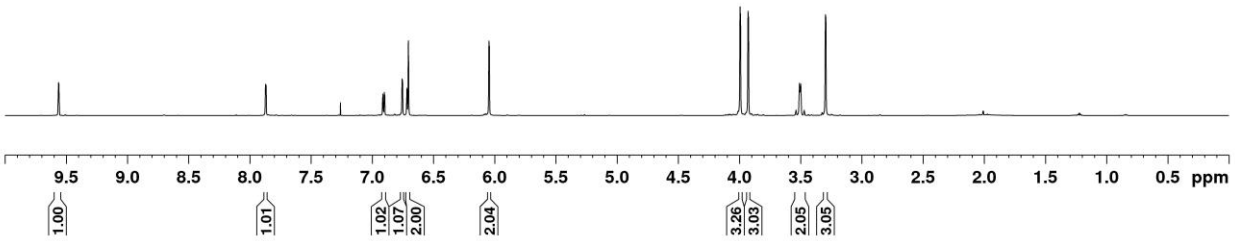



**3-(Benzo[*d*][1,3]dioxol-5-yl)-4-(2-hydroxyethyl)-6,7-dimethoxy-2-methylisoquinolin-1(2*H*)-one (3y) (600 MHz)**

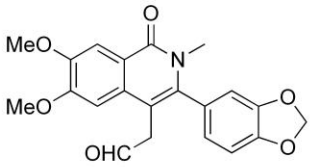
**2-(3-(Benzo[*d*][1,3]dioxol-5-yl)-6,7-dimethoxy-2-methyl-1-oxo-1,2-dihydroisoquinolin-4-yl)acetaldehyde (3y') (600 MHz)**

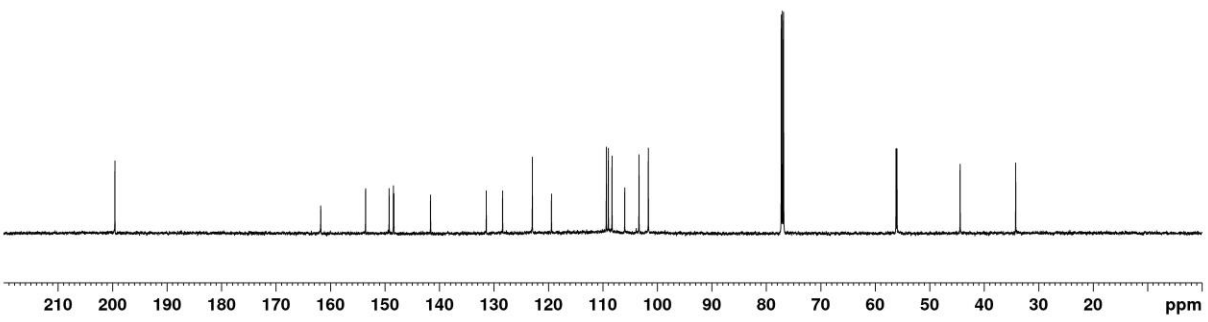


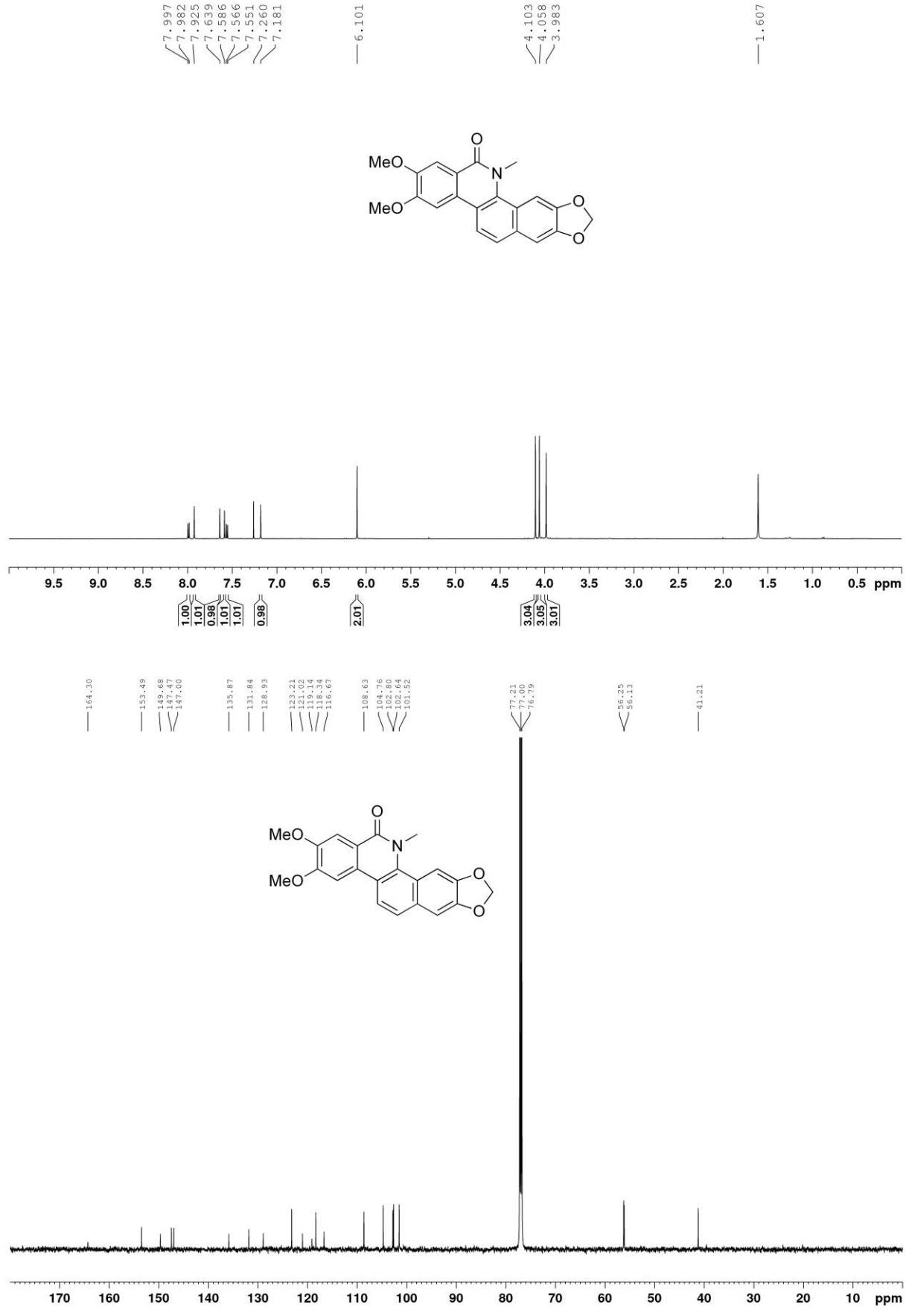





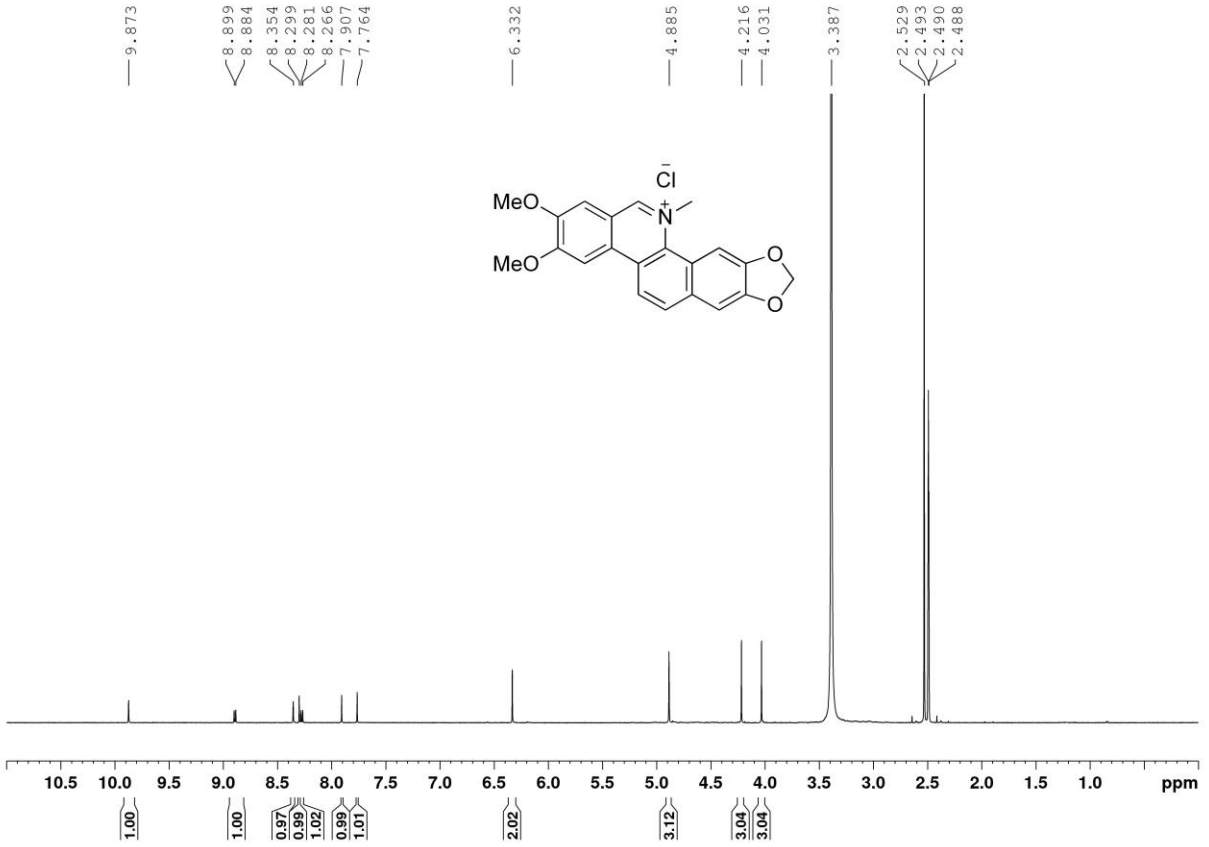


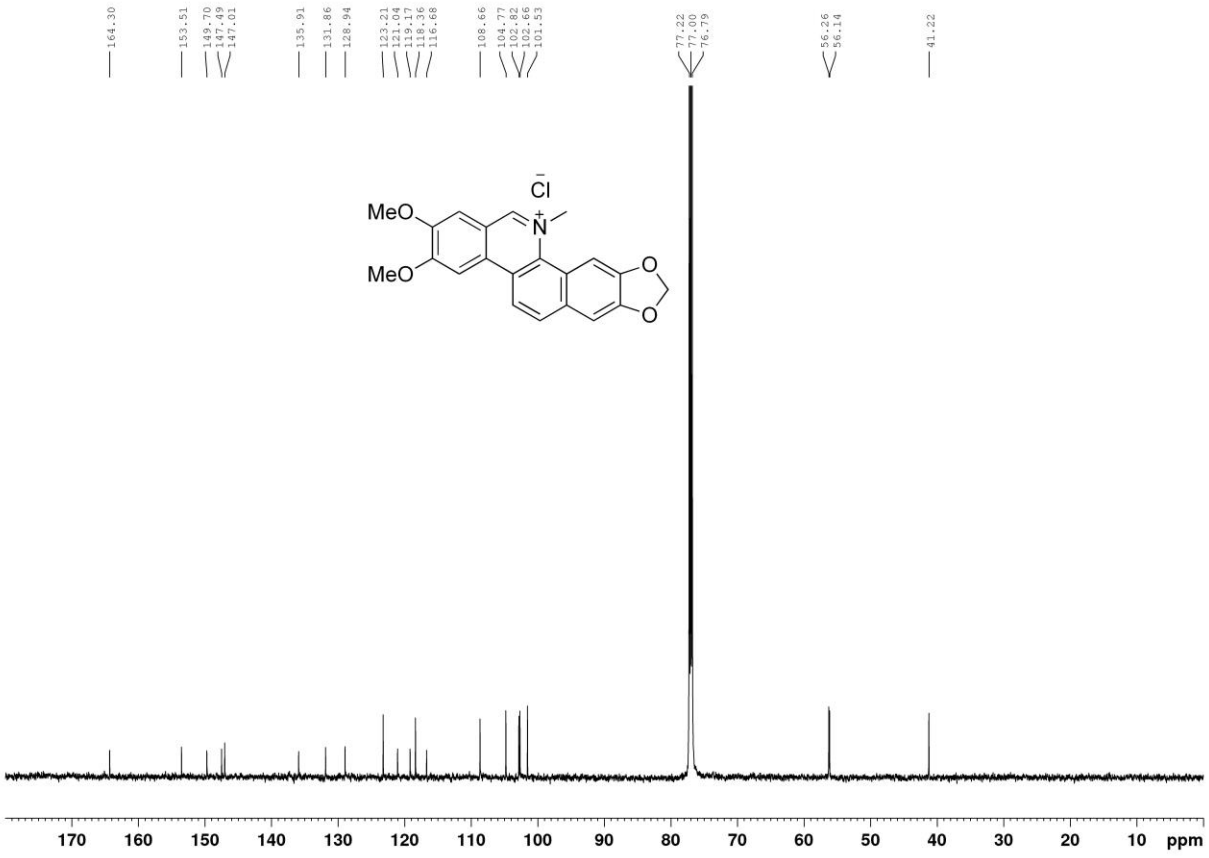



**Oxynitidine (600 MHz)**

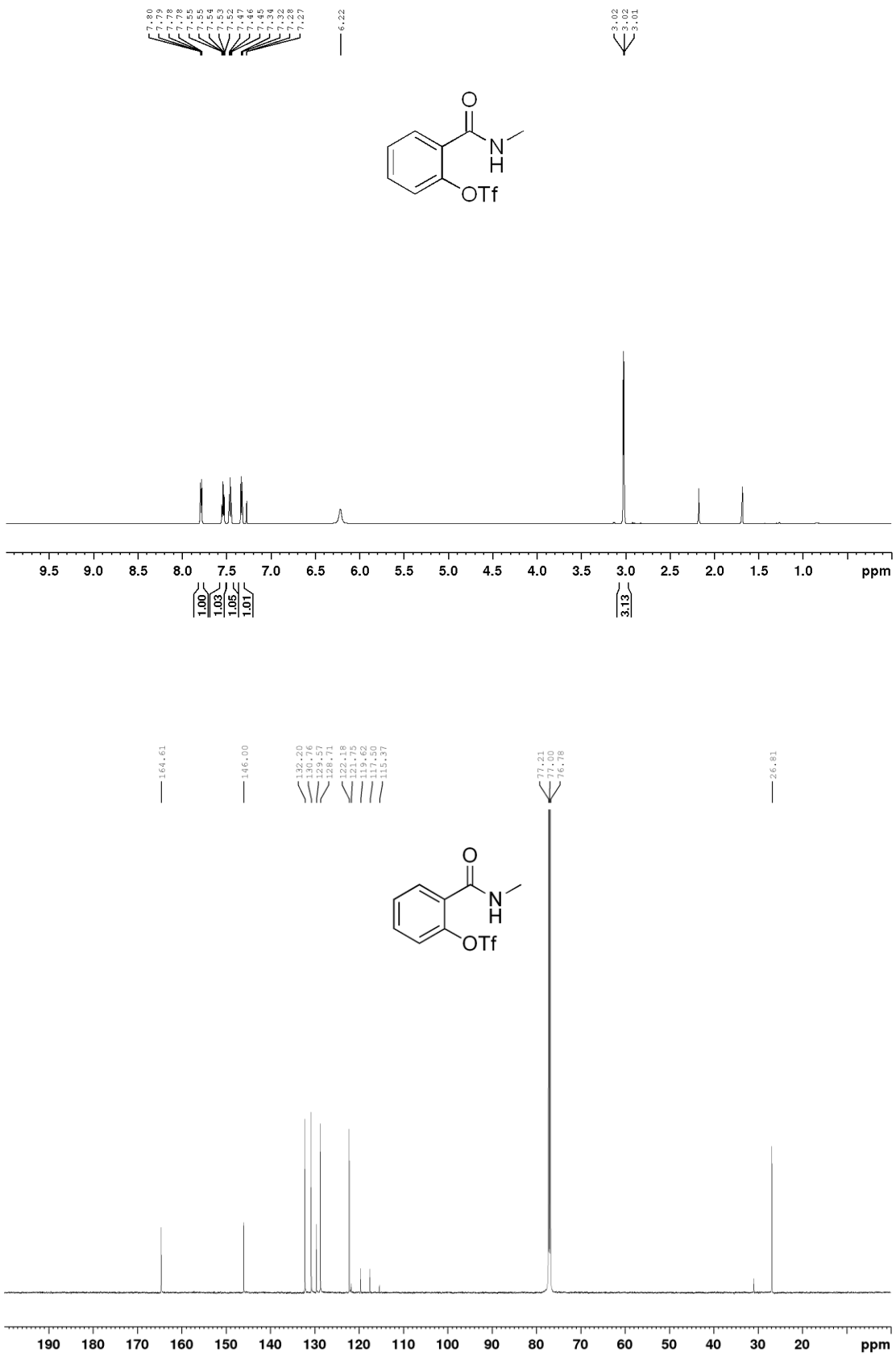


**Nitidine Chloride (DMSO-$d_6$ *for* $^{1}$H NMR and $CDCl_3$ for $^{13}$C NMR, 600 MHz)**

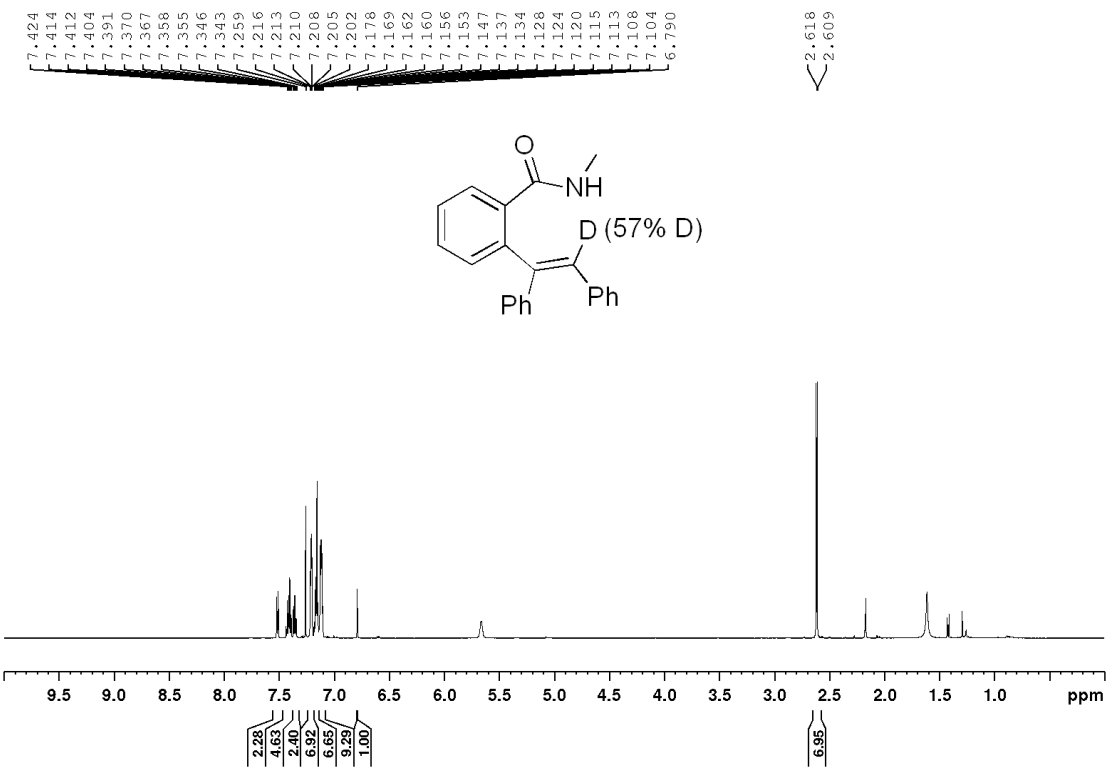


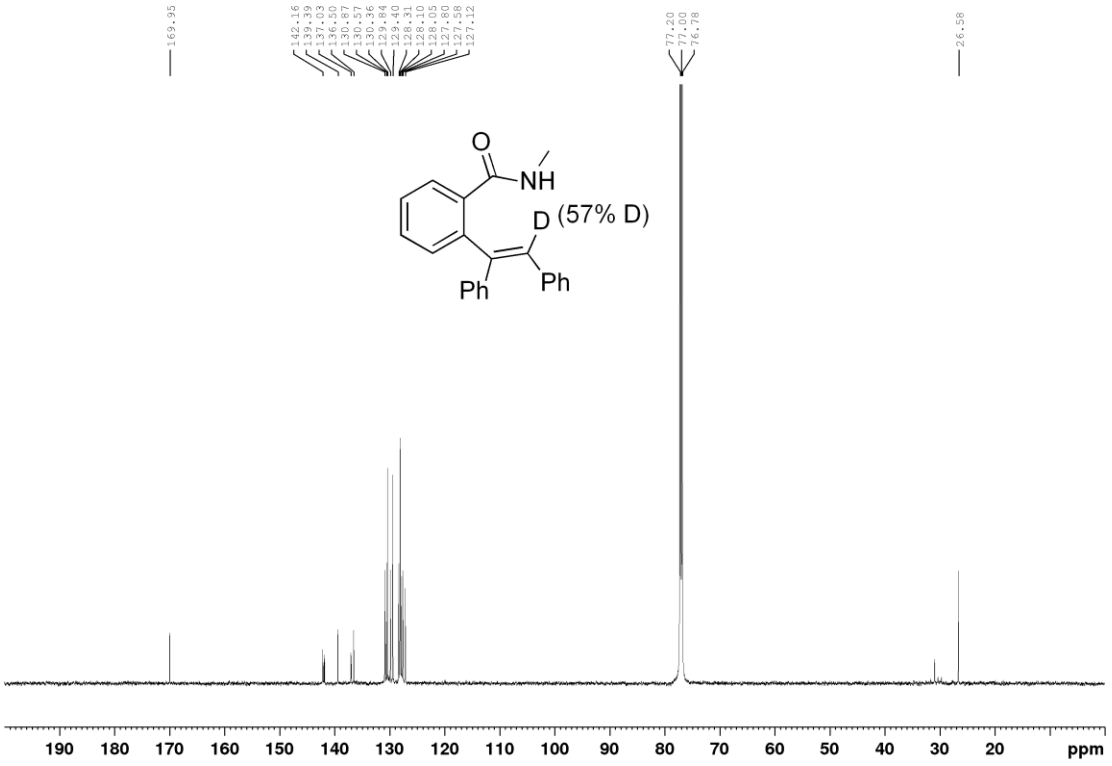



**2-(Methylcarbamoyl)phenyltrifluoromethanesulfonate (1a-f) (600 MHz)**

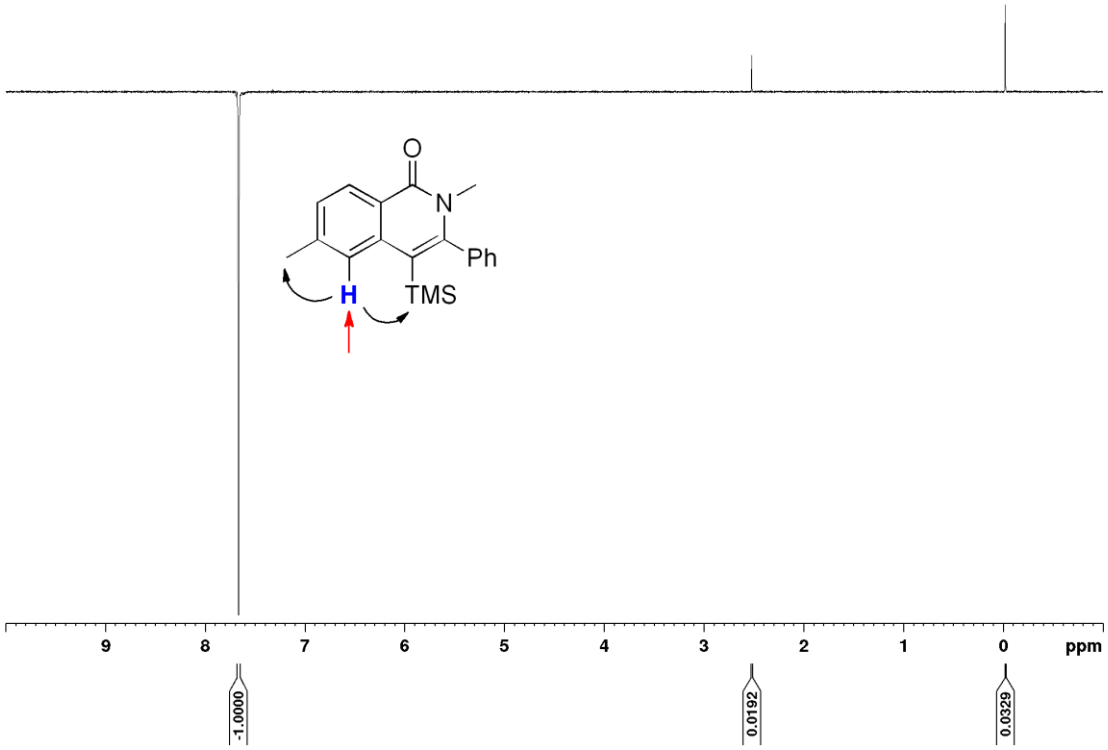


**Deuterated product 4a-D (600 MHz)**



**NOE spectrum of 3x (600 MHz)**



# Single-Crystal X-Ray Diffraction Analysis:

### X-Ray Structure of Compound 3a:

(CCDC 2087800 (**3a**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

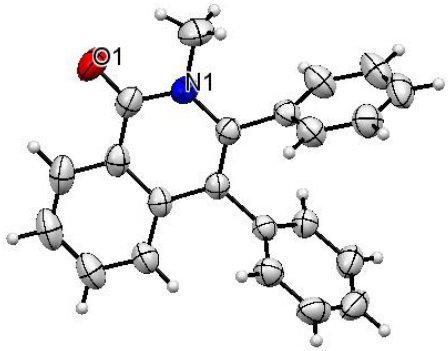



**Figure S1**. X-ray crystal structure of **3a**. Ellipsoids are drawn at the 50% probability level.

**Table S4.** Crystal data and structure refinement for **3a**.

| | | |
|---|---|---|
| Identification code | 1_a | |
| Empirical formula | C22H17NO | |
| Formula weight | 311.36 | |
| Temperature | 296(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Triclinic | |
| Space group | P-1 | |
| Unit cell dimensions | a = 9.3912(2) Å | a = 69.3430(10)°. |
| | b = 9.6810(3) Å | b = 66.3600(10)°. |
| | c = 10.9201(3) Å | g = 67.4890(10)°. |
| Volume | 816.58(4) $Å^3$ | |
| Z | 2 | |
| Density (calculated) | 1.266 $Mg/m^3$ | |
| Absorption coefficient | 0.077 $mm^{-1}$ | |
| F(000) | 328 | |
| Crystal size | ? x ? x ? $mm^3$ | |
| Theta range for data collection | 2.343 to 28.052°. | |
| Index ranges | -12<=h<=12, -12<=k<=12, -14<=l<=14 | |
| Reflections collected | 32840 | |
| Independent reflections | 3957 [R(int) = 0.0656] | |
| Completeness to theta = 25.242° | 99.9 % | |
| Refinement method | Full-matrix least-squares on $F^2$ | |
| Data/restraints/parameters | 3957/0/218 | |
| Goodness-of-fit on $F^2$ | 1.051 | |
| Final R indices [I>2sigma(I)] | R1 = 0.0565, wR2 = 0.1232 | |
| R indices (all data) | R1 = 0.1061, wR2 = 0.1513 | |
| Extinction coefficient | n/a | |
| Largest diff. peak and hole | 0.193 and -0.209 $e.Å^{-3}$ | |

**X-Ray Structure of Compound 4a:**

(CCDC 2087801 (**4a**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

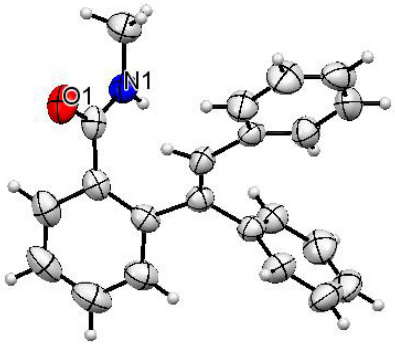



**Figure S2**. X-ray crystal structure of **4a**. Ellipsoids are drawn at the 50% probability level.

**Table S5.** Crystal data and structure refinement for **4a**.

| | | |
|---|---|---|
| Identification code | GP_75P | |
| Empirical formula | C22H19NO | |
| Formula weight | 313.38 | |
| Temperature | 296(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Monoclinic | |
| Space group | $P2_1/n$ | |
| Unit cell dimensions | a = 13.0738(5) Å | = 90°. |
| | b = 8.9358(4) Å | = 92.019(2)°. |
| | c = 14.7988(7) Å | = 90°. |
| Volume | 1727.79(13) $Å^3$ | |
| Z | 4 | |
| Density (calculated) | 1.205 $Mg/m^3$ | |
| Absorption coefficient | 0.073 $mm^{-1}$ | |
| F(000) | 664 | |
| Crystal size | ? x ? x ? $mm^3$ | |
| Theta range for data collection | 2.663 to 28.318°. | |
| Index ranges | -17<=h<=16, -11<=k<=11, -19<=l<=19 | |
| Reflections collected | 40164 | |
| Independent reflections | 4266 [R(int) = 0.1175] | |
| Completeness to theta = 25.242° | 99.6 % | |
| Refinement method | Full-matrix least-squares on $F^2$ | |
| Data/restraints/parameters | 4266/0/218 | |
| Goodness-of-fit on $F^2$ | 1.099 | |
| Final R indices [I>2sigma(I)] | R1 = 0.0828, wR2 = 0.1294 | |
| R indices (all data) | R1 = 0.1618, wR2 = 0.1556 | |
| Extinction coefficient | n/a | |
| Largest diff. peak and hole | 0.140 and -0.171 $e.Å^{-3}$ | |

**X-Ray Structure of Compound 3w:**

(CCDC 2087803 (**3w**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

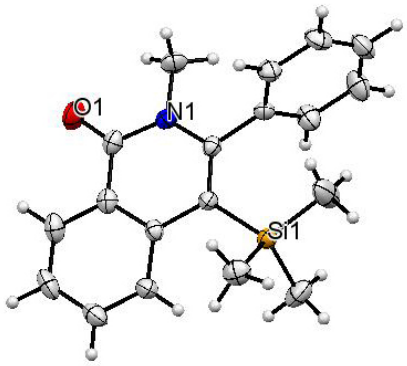



**Figure S3**. X-ray crystal structure of **3w**. Ellipsoids are drawn at the 50% probability level.

**Table S6.** Crystal data and structure refinement for **3w**.

| | | |
|---|---|---|
| Identification code | d23248 | |
| Empirical formula | C19H21NOSi | |
| Formula weight | 307.46 | |
| Temperature | 200(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Triclinic | |
| Space group | P -1 | |
| Unit cell dimensions | a = 8.9558(3) Å | = 102.940(2)°. |
| | b = 9.7312(4) Å | = 96.5450(10)°. |
| | c = 10.0870(4) Å | = 103.5060(10)°. |
| Volume | 820.15(5) $Å^3$ | |
| Z | 2 | |
| Density (calculated) | 1.245 $Mg/m^3$ | |
| Absorption coefficient | 0.145 $mm^{-1}$ | |
| F(000) | 328 | |
| Crystal size | 0.68 x 0.47 x 0.40 $mm^3$ | |
| Theta range for data collection | 2.23 to 25.09°. | |
| Index ranges | -10<=h<=10, -11<=k<=11, -12<=l<=12 | |
| Reflections collected | 11110 | |
| Independent reflections | 2884 [R(int) = 0.0364] | |
| Completeness to theta = 25.09° | 99.0 % | |
| Absorption correction | multi-scan | |
| Max. and min. transmission | 0.9444 and 0.9080 | |
| Refinement method | Full-matrix least-squares on $F^2$ | |
| Data/restraints/parameters | 2884/0/203 | |
| Goodness-of-fit on $F^2$ | 1.047 | |
| Final R indices [I>2sigma(I)] | R1 = 0.0450, wR2 = 0.1275 | |
| R indices (all data) | R1 = 0.0493, wR2 = 0.1306 | |
| Largest diff. peak and hole | 0.320 and -0.285 $e.Å^{-3}$ | |

**X-Ray Structure of Compound 4v':**

(CCDC 2109230 (**4v'**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

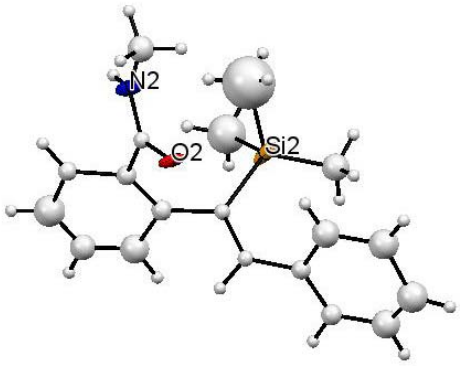



**Figure S4**. X-ray crystal structure of **4v'**. Ellipsoids are drawn at the 50% probability level.

**Table S7.** Crystal data and structure refinement for **4v'**.

| | | |
|---|---|---|
| Identification code | d23470 | |
| Empirical formula | C19 H23NOSi | |
| Formula weight | 309.47 | |
| Temperature | 200(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Triclinic | |
| Space group | P -1 | |
| Unit cell dimensions | a = 10.156(3) Å | = 101.448(7)°. |
| | b = 13.973(3) Å | = 95.499(8)°. |
| | c = 21.035(5) Å | = 110.186(6)°. |
| Volume | 2702.1(11) $Å^3$ | |
| Z | 4 | |
| Density (calculated) | 0.761 $Mg/m^3$ | |
| Absorption coefficient | 0.088 $mm^{-1}$ | |
| F(000) | 664 | |
| Crystal size | 0.79 x 0.03 x 0.01 $mm^3$ | |
| Theta range for data collection | 2.01 to 25.11°. | |
| Index ranges | -12<=h<=12, -16<=k<=16, -25<=l<=25 | |
| Reflections collected | 56021 | |
| Independent reflections | 9592 [R(int) = 0.2132] | |
| Completeness to theta = 25.11° | 99.6 % Absorption correction | multi-scan |
| Max. and min. transmission | 0.9991 and 0.9337 | |
| Refinement method | Full-matrix least-squares on $F^2$ | |
| Data / restraints / parameters | 9592/12/201 | |
| Goodness-of-fit on $F^2$ | 1.686 | |
| Final R indices [I>2sigma(I)] | R1 = 0.2415, wR2 = 0.5306 | |
| R indices (all data) | R1 = 0.3061, wR2 = 0.5518 | |
| Largest diff. peak and hole | 0.640 and -1.702 $e.Å^{-3}$ | |

## X-Ray Structure of Compound 5:

(CCDC 2087805 (**5**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

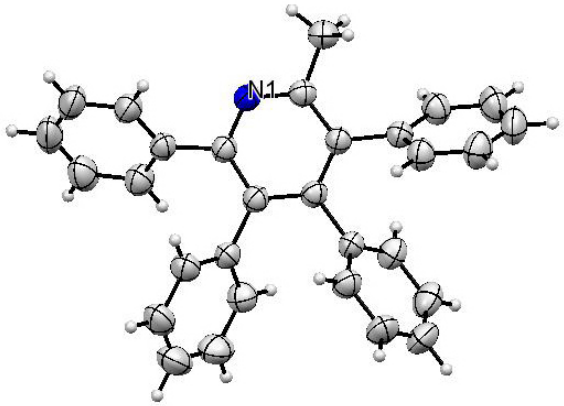



**Figure S5**. X-ray crystal structure of **5**. Ellipsoids are drawn at the 50% probability level.

**Table S8.** Crystal data and structure refinement for **5**.

| | | |
|---|---|---|
| Identification code | 19AP04_1 | |
| Empirical formula | C30H23N | |
| Formula weight | 397.2 | |
| Temperature | 296(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Triclinic | |
| Space group | P-1 | |
| Unit cell dimensions | a = 6.5913(10) Å | = 69.396(4)°. |
| | b = 11.309(2) Å | = 84.651(7)°. |
| | c = 16.066(4) Å | = 88.169(4)°. |
| Volume | 1116.1(4) $Å^3$ | |
| Z | 4 | |
| Density (calculated) | 1.183 $Mg/m^3$ | |
| Absorption coefficient | 0.068 $mm^{-1}$ | |
| F(000) | 420 | |
| Crystal size | 0.391 x 0.069 x 0.060 $mm^3$ | |
| Theta range for data collection | 2.718 to 28.355°. | |
| Index ranges | -8<=h<=8, -15<=k<=15, -21<=l<=21 | |
| Reflections collected | 91333 | |
| Independent reflections | 5569 [R(int) = 0.0462] | |
| Completeness to theta = 25.242° | 99.8 % | |
| Absorption correction | Numerical Mu Calculated | |
| Max. and min. transmission | 0.7457 and 0.7106 | |
| Refinement method | Full-matrix least-squares on $F^2$ | |
| Data/restraints/parameters | 5569/0/281 | |
| Goodness-of-fit on $F^2$ | 1.030 | |
| Final R indices [I>2sigma(I)] | R1 = 0.0453, wR2 = 0.1143 | |
| R indices (all data) | R1 = 0.0651, wR2 = 0.1292 | |
| Extinction coefficient | n/a | |
| Largest diff. peak and hole | 0.178 and -0.146 $e.Å^{-3}$ | |

## X-Ray Structure of Compound 6:

(CCDC 2089397 (**6**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

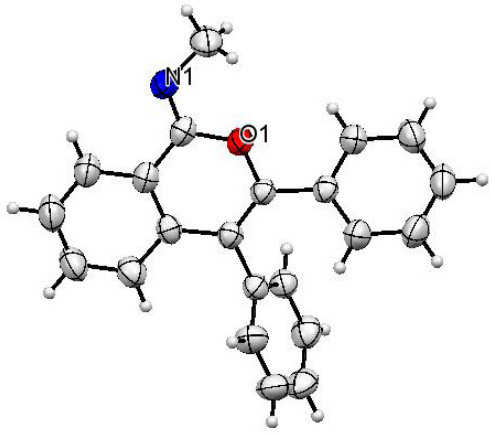



**Figure S6**. X-ray crystal structure of **6**. Ellipsoids are drawn at the 50% probability level.

**Table S9.** Crystal data and structure refinement for **6**.

| | | |
|---|---|---|
| Identification code | 02_a | |
| Empirical formula | C22H17NO | |
| Formula weight | 311.36 | |
| Temperature | 296(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Monoclinic | |
| Space group | $P2_1/n$ | |
| Unit cell dimensions | a = 8.8118(4) Å | = 90°. |
| | b = 18.3413(8) Å | = 102.612(2)°. |
| | c = 10.4793(5) Å | = 90°. |
| Volume | 1652.80(13) Å$^3$ | |
| Z | 4 | |
| Density (calculated) | 1.251 Mg/m$^3$ | |
| Absorption coefficient | 0.076 mm$^{-1}$ | |
| F(000) | 656 | |
| Crystal size | 0.468 x 0.272 x 0.214 mm$^3$ | |
| Theta range for data collection | 2.221 to 28.343°. | |
| Index ranges | -11<=h<=11, -24<=k<=24, -13<=l<=13 | |
| Reflections collected | 25049 | |
| Independent reflections | 4104 [R(int) = 0.0452] | |
| Completeness to theta = 25.242° | 99.4 % | |
| Absorption correction | Numerical Mu Calculated | |
| Max. and min. transmission | 0.7379 and 0.7199 | |
| Refinement method | Full-matrix least-squares on F$^2$ | |
| Data / restraints / parameters | 4104 / 0 / 218 | |
| Goodness-of-fit on F$^2$ | 1.068 | |
| Final R indices [I>2sigma(I)] | $R_1$ = 0.0570, $wR_2$ = 0.1543 | |
| R indices (all data) | $R_1$ = 0.0745, $wR_2$ = 0.1695 | |
| Extinction coefficient | n/a | |
| Largest diff. peak and hole | 0.354 and -0.193 e.Å$^{-3}$ | |

## X-Ray Structure of Compound 7:

(CCDC 2087808 (**7**) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.)

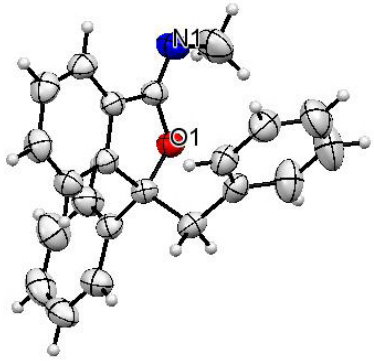



**Figure S7**. X-ray crystal structure of **7**. Ellipsoids are drawn at the 50% probability level.

**Table S10.** Crystal data and structure refinement for **7**.

| | | |
|---|---|---|
| Identification code | 19JUN01 | |
| Empirical formula | C22H19NO | |
| Formula weight | 313.38 | |
| Temperature | 296(2) K | |
| Wavelength | 0.71073 Å | |
| Crystal system | Monoclinic | |
| Space group | $P2_1/c$ | |
| Unit cell dimensions | a = 12.0230(3) Å | = 90°. |
| | b = 7.74160(10) Å | = 103.8500(10)°. |
| | c = 19.2617(4) Å | = 90°. |
| Volume | 1740.70(6) $Å^3$ | |
| Z | 4 | |
| Density (calculated) | 1.196 $Mg/m^3$ | |
| Absorption coefficient | 0.073 $mm^{-1}$ | |
| F(000) | 664 | |
| Crystal size | 0.510 x 0.270 x 0.152 $mm^3$ | |
| Theta range for data collection | 2.848 to 28.316°. | |
| Index ranges | -16<=h<=15, -10<=k<=10, -25<=l<=25 | |
| Reflections collected | 28519 | |
| Independent reflections | 4302 [R(int) = 0.0433] | |
| Completeness to theta = 25.242° | 99.6 % | |
| Absorption correction | Numerical Mu Calculated | |
| Max. and min. transmission | 0.7457 and 0.7231 | |
| Refinement method | Full-matrix least-squares on $F^2$ | |
| Data/restraints/parameters | 4302/0/218 | |
| Goodness-of-fit on $F^2$ | 1.045 | |
| Final R indices [I>2sigma(I)] | R1 = 0.0488, wR2 = 0.1079 | |
| R indices (all data) | R1 = 0.0787, wR2 = 0.1268 | |
| Extinction coefficient | n/a | |
| Largest diff. peak and hole | 0.159 and -0.155 $e.Å^{-3}$ | |