Electronic Supplementary Material (ESI) for Organic & Biomolecular Chemistry.

Rapid and Selective Synthesis of Spiropyrazolines and Pyrazolylphthalides Employing Seyferth-Gilbert Reagent

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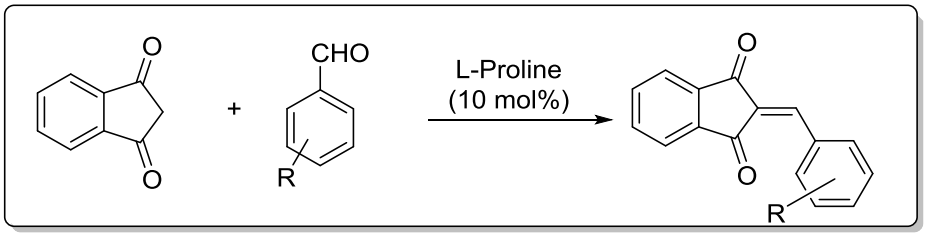
SUPPORTING INFORMATION

Content:

General experimental information:

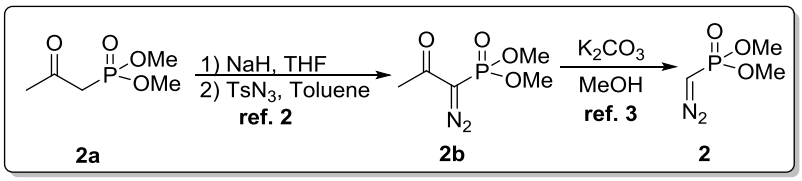
Unless otherwise specified, all reactions were carried out under air atmosphere in oven dried round-bottom flasks. Dimethyl 2-oxopropylphosphonate and 1,3-Indanedione were purchased from commercial sources and were used without further purification. The reactions were monitored by TLC visualized by UV (254 nm) and/or with iodine. Flash chromatography was performed on 100-200 mesh silica gel using the gradient system ethyl acetate-hexane (0-50%)/Acetone-Dichloromethane (0-30%). NMR data were recorded at Bruker AV 400 MHz in DMSO-d_6/$CDCl_3$ using as internal standards the residual DMSO signal for ^1H NMR (δ = 2.50 ppm), $CHCl_3$ (δ = 7.26 ppm) respectively. The corresponding deuterated solvent signal for ^{13}C NMR were assigned as DMSO (δ = 39.51 ppm), $CDCl_3$ (77.16). Coupling constants are given in Hertz (Hz) and the classical abbreviations are used to describe the signal multiplicities. Melting points were measured with a Büchi B-540 apparatus and are uncorrected. High resolution mass spectra were obtained using Q-TOF mass spectrometer. All commercially available reagents were used as received.

General procedure for the synthesis of arylidine indanedione



1,3-Indanedione (1.1 mmol), aldehydes (1 mmol), and L-proline (10 mol%) were mixed in a mortar under solvent-free conditions. The completion of the reaction was monitored by Thin Layer Chromatography (TLC), ethyl acetate was added to the crude reaction mixture. The reaction mixture in ethyl acetate was washed with brine and organic layers were separated and dried over anhydrous Na_2SO_4 and evaporated under vacuum. The crude residues were purified by column chromatography (ethyl acetate/hexane: 1/9). Purified compounds were characterised by comparing with literature data.[1]

Procedure for the preparation of Seyferth-Gilbert reagent (SGR)



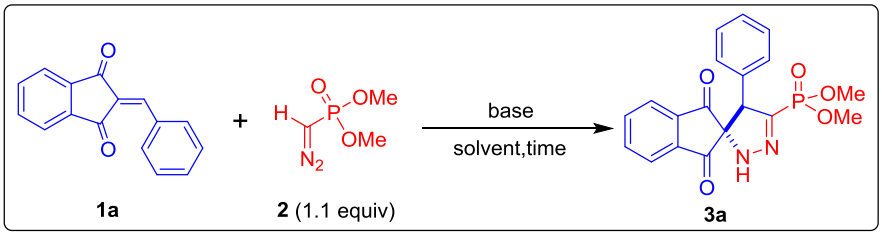
Dimethyl diazomethylphosphonate **2** was prepared by modifying the literature report.[2,3] To a stirred suspension of NaH (576 mg, 24.0 mmol) in dry THF (30 mL) at 0 °C, added a solution of 2-oxopropyl phosphonate **2a** (2.0 g, 12.0 mmol) in dry THF (10 mL) dropwise. The reaction mixture was stirred at 0 °C for 30 minutes and then solution of TsN_3 in toluene was added dropwise, the reaction mixture was further stirred at 25 °C for 3 h. After the completion of reaction, as indicated by TLC, the precipitate was filtered off. Solvent was removed on rotary evaporator and the residue was purified by column chromatography on silica gel (Acetone/Dichloromethane = 1/9). Compound **2b** was obtained as yellow liquid (1.96 g, 83% yield).

A solution of **2b** (1.92 g, 10 mmol) in 10 mL of MeOH was stirred with potassium carbonate (276 mg, 2 mmol) at room temperature for 15 min (monitored by TLC analysis). The precipitate was filtered off. Solvent was removed on rotary evaporator and the residue was purified by column chromatography on silica gel (Acetone/Dichloromethane = 1/9). Compound **2** was obtained as yellow liquid (1.3 g, 87 % yield).

References

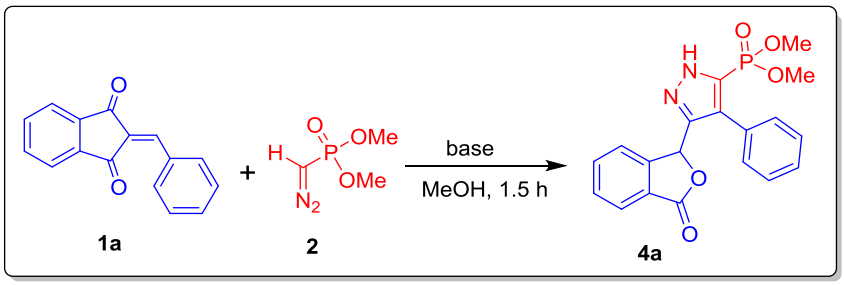
1. P. Goswami and B. Das, *Tetrahedron Lett.,* 2009, **50**, 897.
2. P. Callant, L. Dhaenens and M. Vandewalle, *Synth. Commun.,* 1984, **14**, 155.
3. S. Ohira, *Synth. Commun.,* 1989, **19**, 561.

Optimization studies by varying base, solvent and stoichiometry: Synthesis of spiropyrazolines



S.No	Solvent	Base	Equiv	Yield
1	Et_2O	NaOH	2.0	<5%
2	CH_3CN	*t*-BuOK	2.0	<5%
3	CH_3CN	NaOEt	2.0	<5%
4	CH_3CN	Et_3N	2.0	82%
5	CH_3CN	DBU	2.0	75%
6	CH_3CN	CsF	0.1	72%
7	CH_2Cl_2	CsF	0.1	75%
8	THF	CsF	0.1	81%
9	CH_3COCH_3	CsF	0.1	87%
10	Toluene	CsF	0.1	56%
11	$CH_3COOC_2H_5$	CsF	0.1	79%
12	Et_2O	CsF	0.1	21%

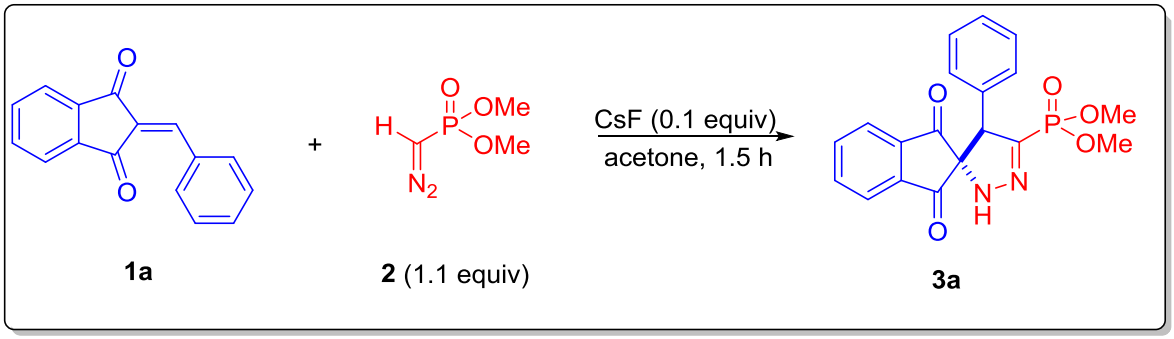
Optimization studies by varying base and stoichiometry of SGR: Synthesis of pyrazolylphthalides



S.No	Base	SGR (equiv)	Yield
1	NaOMe	1.1	75%
2	*t*-BuOK	1.1	70%
3	K_2CO_3	1.1	72%
4	KOH	1.1	77%
5	NaOH	1.1	79%
6	DBU	1.1	75%
7	Na_2CO_3	1.1	32%
8	Et_3N	1.1	<5%
9	NaOH	1.5	86%

Procedures

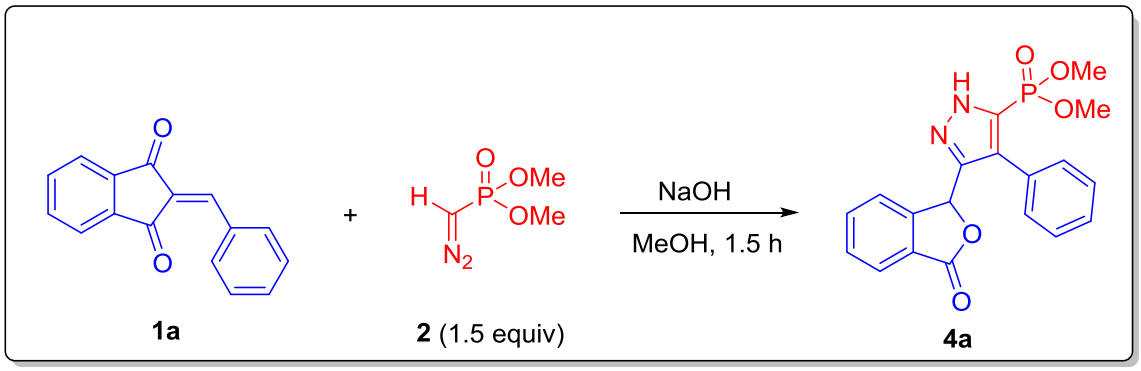
Synthesis of spiropyrazolines 3a-3q



***General procedure for the synthesis of spiropyrazolines* 3a-3q**

To an oven-dried round bottom flask was added 2-(benzylidene)-1H-indene-1,3(2H)-dione **1a** (50 mg, 0.21 mmol) and dissolved in 3 mL of acetone. Subsequently, a solution of Seyferth Gilbert reagent (33 mg, 0.24 mmol) in 2 mL of acetone was added to the reaction mixture and kept stirring. After addition of cesium fluoride (3.0 mg, 0.021 mmol), the reaction mixture was stirred at 25 °C for 1.5 h. After the completion of reaction, as indicated by TLC, solvent was evaporated off and extracted using ethyl acetate. The organic layer was dried over Na_2SO_4 and evaporated under reduced pressure. The residue was purified using column chromatography (100-200 mesh silica gel) using acetone/dichloromethane as the eluent to afford the corresponding spiropyrazoline **3a** as a white solid (70 mg) in 87% yield.

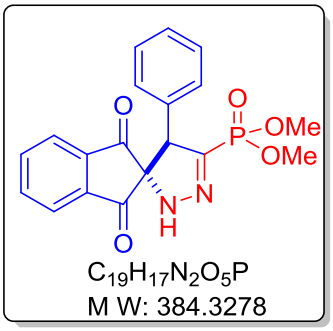
Synthesis of pyrazolylphthalides of 4a – 4w



***General procedure for the synthesis of pyrazolylphthalides* 4a-4w**

To an oven-dried round bottom flask was added 2-(benzylidene)-1H-indene-1,3(2H)-dione **1a** (50 mg, 0.21 mmol) and dissolved in 3 mL of MeOH. Subsequently, Seyferth Gilbert reagent (48 mg, 0.33 mmol) in 2 mL of MeOH was added to the reaction mixture and kept stirring. After addition of Sodium hydroxide (21.0 mg, 0.53 mmol), the reaction mixture was stirred at 25 °C for 1.5 h. After the completion of reaction, as indicated by TLC, solvent was evaporated off and extracted using ethyl acetate. The organic layer was dried over Na_2SO_4 and evaporated under reduced pressure. The residue was purified using column chromatography (100-200 mesh silica gel) using acetone/dichloromethane as the eluent to afford pyrazolylphthalides **4a** as a white solid (69 mg) in 86 % yield.

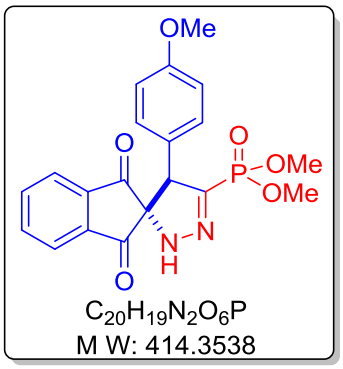
Dimethyl (1,3-dioxo-4'-phenyl-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3a)



$C_{19}H_{17}N_2O_5P$
M W: 384.3278

Following the general procedure, treatment of 2-(benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.21 mmol) with SGR (36 mg, 0.24 mmol) in the presence of CsF (3.0 mg, 0.021 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3a** as a yellow solid (70 mg, 87%). $\mathbf{R_f}$ (Acetone/Dichloromethane: 1/9) = 0.29. **Mp** 178-180 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.1 (C), 195.4 (C), 142.6 (d, J_{C-P} = 227.0 Hz, C), 141.6 (C), 140.1 (C), 136.6 (CH), 136.4 (CH), 131.8 (C), 129.5 (CH), 129.5 (CH), 128.7 (CH), 128.5 (CH), 128.5 (CH), 124.1 (CH), 123.5 (CH), 79.1 (d, J_{C-P} = 23.8 Hz, C), 65.3 (d, J_{C-P} = 23.8 Hz, CH), 53.6 (d, J_{C-P} = 5.5 Hz, CH_3), 53.4 (d, J_{C-P} = 5.9 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.98 (d, J = 7.6 Hz, 1H), 7.82 (t, J = 7.6 Hz, 1H), 7.73 (t, J = 7.4 Hz, 1H), 7.54 (d, J = 7.6 Hz, 1H), 7.16-7.15 (m, 3H), 6.93-6.91 (m, 3H), 4.72 (s, 1H), 3.62 (d, J = 11.6 Hz, 3H), 3.58 (d, J = 11.2 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.95. **HRMS** for $C_{19}H_{18}N_2O_5P^+$: calcd. $[M+H]^+$: 385.0948, found: 385.0949.

Dimethyl (4'-(4-methoxyphenyl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3b)

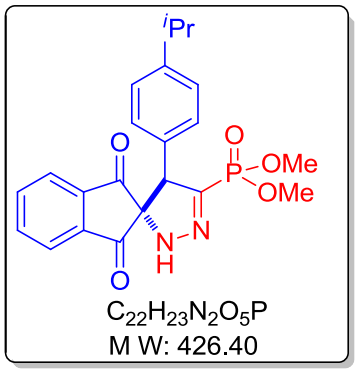


$C_{20}H_{19}N_2O_6P$
M W: 414.3538

Following the general procedure, treatment of 2-(4-methoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (32 mg, 0.21 mmol) in the presence of CsF (3.0 mg, 0.019 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3b** as a yellow solid (59 mg, 75%). $\mathbf{R_f}$ (Acetone/Dichloromethane: 1/9) = 0.31. **Mp** 160-162 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.2 (C), 195.5 (C), 159.6 (C), 142.4 (d, J_{C-P} = 182.0 Hz, C), 141.5 (C), 140.1 (C), 136.5 (CH), 136.2 (CH), 130.7 (CH), 130.7 (CH), 123.9 (C), 123.7 (CH), 123.4 (CH), 113.8 (CH), 113.8 (CH), 79.0 (d, J_{C-P} = 3.6 Hz, C), 64.6 (d, J_{C-P} = 19.1 Hz, CH), 55.1 (CH_3), 53.4 (d, J_{C-P} = 4.5 Hz, CH_3), 53.3 (d, J_{C-P} = 4.9 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.96 (d, J = 7.6 Hz, 1H), 7.81 (t, J = 7.4 Hz, 1H), 7.73 (t, J = 7.4 Hz, 1H), 7.56 (d, J = 7.6 Hz, 1H), 6.92 (s, 1H), 6.83 (d, J = 8.4 Hz, 2H), 6.66 (d, J = 8.4 Hz,

2H), 4.68 (s, 1H), 3.69 (s, 3H), 3.60 (d, *J* = 11.2 Hz, 3H), 3.58 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.14. **HRMS** for $C_{20}H_{20}N_2O_6P^+$: calcd. $[M+H]^+$: 415.1053, found: 415.1053.

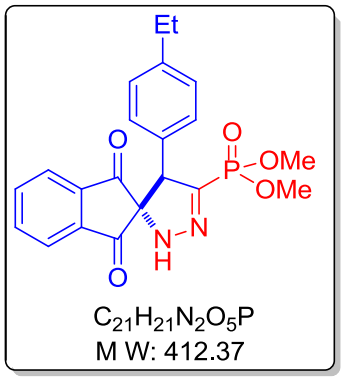
Dimethyl (4'-(4-isopropylphenyl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3c)



$C_{22}H_{23}N_2O_5P$
M W: 426.40

Following the general procedure, treatment of 2-(4-isopropylbenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.18 mmol) with SGR (30 mg, 0.20 mmol) in the presence of CsF (3.0 mg, 0.018 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3c** as a yellow solid (71 mg, 92%). $\mathbf{R}_f$ (Acetone/Dichloromethane: 1/9) = 0.53. **Mp** 178-180 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.1 (C), 195.6 (C), 149.4 (C), 143.1 (d, $J_{C\text{-}P}$ = 226.7 Hz, C), 141.6 (C), 140.3 (C), 136.5 (CH), 136.2 (CH), 129.5 (CH), 129.5 (CH), 129.0 (C), 126.6 (CH), 126.6 (CH), 124.0 (CH), 123.5 (CH), 79.6 (d, $J_{C\text{-}P}$ = 4.1 Hz, C), 65.4 (d, $J_{C\text{-}P}$ = 24.0 Hz, CH), 53.6 (d, $J_{C\text{-}P}$ = 5.6 Hz, CH_3), 53.4 (d, $J_{C\text{-}P}$ = 5.8 Hz, CH_3), 33.8 (CH), 23.9 (CH_3), 23.8 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 8.03 (d, *J* = 7.6 Hz, 1H), 7.86 (t, *J* = 7.4 Hz, 1H), 7.76 (t, *J* = 7.4 Hz, 1H), 7.58 (d, *J* = 7.6 Hz, 1H), 7.02 (d, *J* = 6.8 Hz, 2H), 6.87 (d, *J* = 6.4 Hz, 2H), 6.76-6.71 (m, 1H), 4.78 (s, 1H), 3.69 (d, *J* = 11.2 Hz, 3H), 3.64 (d, *J* = 13.6 Hz, 3H) 2.84-2.77 (m, 1H), 1.16 (d, *J* = 6.8 Hz, 6H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.00. **HRMS** for $C_{22}H_{24}N_2O_5P^+$: calcd. $[M+H]^+$: 427.1417, found: 427.1414.

Dimethyl (4'-(4-ethylphenyl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3d)

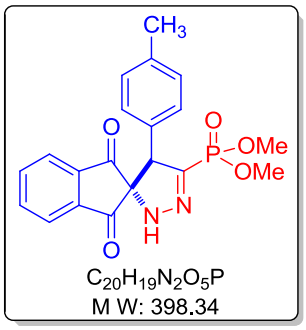


$C_{21}H_{21}N_2O_5P$
M W: 412.37

Following the general procedure, treatment of 2-(4-ethylbenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (32 mg, 0.21 mmol) in the presence of CsF (3.0 mg, 0.019 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3d** as a yellow solid (69 mg, 88%). $\mathbf{R}_f$ (Acetone/Dichloromethane: 1/9) = 0.58. **Mp** 170-172 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.1 (C), 195.5 (C), 144.8 (C), 143.0 (d, $J_{C\text{-}P}$ = 226.7 Hz, C), 141.6 (C), 140.2 (C), 136.5 (CH), 136.3 (CH),

129.5 (CH), 129.5 (CH), 128.9 (C), 128.0 (CH), 128.0 (CH), 124.0 (CH), 123.5 (CH), 79.3 (d, $J_{C\text{-}P}$ = 4.3 Hz, C), 65.2 (d, $J_{C\text{-}P}$ = 23.9 Hz, CH), 53.6 (d, $J_{C\text{-}P}$ = 5.4 Hz, CH_3), 53.3 (d, $J_{C\text{-}P}$ = 5.8 Hz, CH_3), 28.5 (CH_2), 15.3 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.99-7.96 (m, 1H), 7.84-7.79 (m, 1H), 7.73-7.72 (m, 1H), 7.56-7.53 (m, 1H), 6.97 (d, *J* = 4.8 Hz, 2H), 6.84-6.81 (m, 3H), 4.71 (s, 1H), 3.65-3.57 (m, 6H), 2.53-2.50 (m, 2H), 1.13-1.09 (m, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.98. **HRMS** for $C_{21}H_{22}N_2O_5P^+$: calcd. $[M+H]^+$: 413.1261, found: 413.1251.

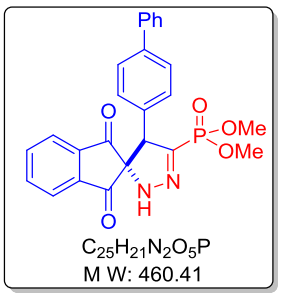
Dimethyl (1,3-dioxo-4'-(*p*-tolyl)-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3e)



$C_{20}H_{19}N_2O_5P$
M W: 398.34

Following the general procedure, treatment of 2-(4-methylbenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.20 mmol) with SGR (33 mg, 0.22 mmol) in the presence of CsF (3.0 mg, 0.02 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3e** as a yellow solid (70 mg, 88%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.41. **Mp** 192-194 °C. **^{13}C NMR** (100 MHz, ppm/ DMSO-d_6): 197.4 (C), 195.0 (C), 141.1 (C), 139.6 (C), 139.0 (d, $J_{C\text{-}P}$ = 182.0 Hz, C), 137.1 (C), 136.8 (CH), 136.6 (CH), 129.8 (C), 129.0 (CH), 129.0 (CH), 128.6 (CH), 128.6 (CH), 123.8 (CH), 122.8 (CH), 77.5 (d, $J_{C\text{-}P}$ = 3.5 Hz, C), 62.2 (d, $J_{C\text{-}P}$ = 19 Hz, CH), 52.8 (d, $J_{C\text{-}P}$ = 4.8 Hz, CH_3), 52.8 (d, $J_{C\text{-}P}$ = 4.8 Hz, CH_3), 20.7 (CH_3). **^{1}H NMR** (400 MHz, ppm/DMSO-d_6): 8.59 (s, 1H), 8.03 (d, *J* = 7.6 Hz, 1H), 7.98 (t, *J* = 7.2 Hz, 1H), 7.92 (t, *J* = 7.4 Hz, 1H), 7.66 (d, *J* = 7.6 Hz, 1H), 6.99 (d, *J* = 8.0 Hz, 2H), 6.82 (d, *J* = 6.4 Hz, 2H), 4.71 (s, 1H), 3.55 (d, *J* = 11.2 Hz, 3H), 3.51 (d, *J* = 11.2 Hz, 3H), 2.21 (s, 3H). **^{31} PNMR** (161.9 MHz, DMSO-d_6): 10.14. **HRMS** for $C_{20}H_{20}N_2O_5P^+$: calcd. $[M+H]^+$: 399.1104, found: 399.1092.

Dimethyl (4'-([1,1'-biphenyl]-4-yl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3f)

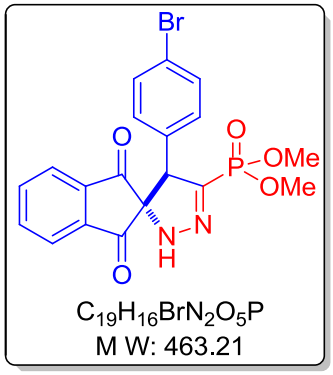


$C_{25}H_{21}N_2O_5P$
M W: 460.41

Following the general procedure, treatment of 2-([1,1'-biphenyl]-4-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.16 mmol) with SGR (27 mg, 0.18 mmol) in the presence of CsF (2.0 mg, 0.016 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3f** as a yellow solid (66 mg,

90%). **R_f** (Acetone/Dichloromethane : 1/9) = 0.41. **Mp** 163-165 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.0 (C), 195.4 (C), 142.5 (d, J_{C-P} = 227.4 Hz, C), 141.6 (C), 141.3 (C), 140.2 (C), 136.7 (CH), 136.4 (CH), 130.8 (C), 130.0 (CH), 130.0 (CH), 128.9 (CH), 128.9 (CH), 127.7 (C), 127.1 (CH), 127.1 (CH), 127.0 (CH), 127.0 (CH), 124.1 (CH), 123.6 (CH), 79.2 (d, J_{C-P} = 4.1 Hz, C), 65.0 (d, J_{C-P} = 23.8 Hz, CH), 53.6 (d, J_{C-P} = 5.5 Hz, CH_3), 53.4 (d, J_{C-P} = 5.8 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 8.02 (d, *J* = 7.6 Hz, 1H), 7.84 (t, *J* = 7.6 Hz, 1H), 7.72 (t, *J* = 7.4 Hz, 1H), 7.59 (d, *J* = 7.6 Hz, 1H), 7.52-7.50 (m, 2H), 7.43-7.38 (m, 4H), 7.34-7.32 (m, 1H), 7.02 (d, *J* = 8.4 Hz, 2H), 6.78 (s, 1H), 4.82 (s, 1H), 3.69 (d, *J* = 11.6 Hz, 3H), 3.65 (d, *J* = 11.2 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.78. **HRMS** for $C_{25}H_{22}N_2O_5P^+$: calcd. $[M+H]^+$: 461.1261, found: 461.1259.

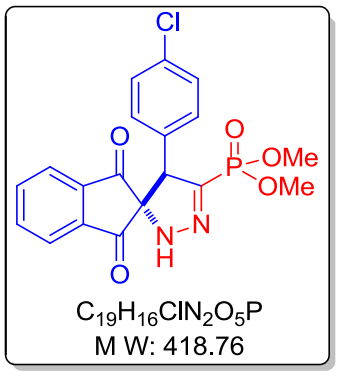
Dimethyl (4'-(4-bromophenyl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3g)



$C_{19}H_{16}BrN_2O_5P$
M W: 463.21

Following the general procedure, treatment of 2-(4-bromobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.16 mmol) with SGR (27 mg, 0.18 mmol) in the presence of CsF (2 mg, 0.016 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3g** as a yellow solid (63 mg, 85%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.44. **Mp** 204-206 °C. **^{13}C NMR** (100 MHz, δ ppm/$CDCl_3$): 196.2 (C), 194.1 (C), 140.5 (C), 138.9 (C), 137.8 (d, J_{C-P} = 200.4 Hz, C), 135.6 (CH), 135.4 (CH), 130.9 (C), 130.3 (CH), 130.3 (CH), 130.1 (CH), 130.1 (CH), 122.9 (CH), 122.2 (CH), 121.3 (C), 61.7 (d, J_{C-P} = 23.5 Hz, CH), 52.2 (d, J_{C-P} = 5.6 Hz, CH CH_3), 52.1 (d, J_{C-P} = 5.6 Hz, CH CH_3). **^{1}H NMR** (400 MHz, δ ppm/$CDCl_3$): 8.03 (d, *J* = 7.6 Hz, 1H), 7.88 (t, *J* = 7.0 Hz, 1H), 7.81 (t, *J* = 7.0 Hz, 1H), 7.66 (d, *J* = 7.6 Hz, 1H), 7.33 (d, *J* = 8.4 Hz, 2H), 6.86 (d, *J* = 8.4 Hz, 2H), 6.70 (s, 1H), 4.74 (s, 1H), 3.71 (d, *J* = 11.2 Hz, 3H), 3.67 (d, *J* = 11.2 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 14.53. **HRMS** for $C_{19}H_{17}N_2O_5PBr^+$: calcd. $[M+H]^+$: 463.0053, found: 463.0052.

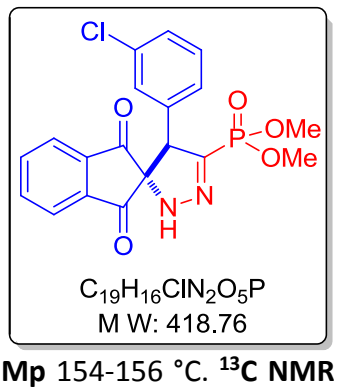
Dimethyl(4'-(4-chlorophenyl)-2-oxo-2',4'-dihydrospiro[indoline-3,3'-pyrazol]-5'-yl)phosphonate (3h)



$C_{19}H_{16}ClN_2O_5P$
M W: 418.76

Following the general procedure, treatment of 2-(4-chlorobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (32 mg, 0.21 mmol) in the presence of CsF (3.0 mg, 0.019 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3h** as a yellow solid (61 mg, 77%). $\mathbf{R_f}$ (Acetone/Dichloromethane: 1/9) = 0.49. **Mp** 180-182 °C. **^{13}C NMR** (100 MHz, δ ppm/DMSO-d_6): 197.0 (C), 194.9 (C), 141.1 (C), 139.5 (C), 138.7 (d, $J_{C\text{-}P}$ = 227.9 Hz, C), 136.8 (CH), 136.7 (CH), 132.7 (C), 132.2 (C), 131.0 (CH), 131.0 (CH), 128.0 (CH), 128.0 (CH), 124.0 (CH), 122.9 (CH), 76.9 (d, $J_{C\text{-}P}$ = 3.7 Hz, C), 60.9 (d, $J_{C\text{-}P}$ = 23.6 Hz, CH), 52.9 (d, $J_{C\text{-}P}$ = 6.2 Hz, CH_3), 52.8 (d, $J_{C\text{-}P}$ = 5.8 Hz, CH_3). **^{1}H NMR** (400 MHz, δ ppm/DMSO-d_6): 8.67 (s, 1H), 8.05-7.93 (m, 3H), 7.71 (s, 1H), 7.29 (d, J = 3.2 Hz, 2H), 7.00 (d, J = 4.0 Hz, 2H), 4.84 (s, 1H), 3.59 (d, J = 9.2 Hz, 3H), 3.55 (d, J = 6.0 Hz, 3H) **^{31}P NMR** (161.9 MHz, DMSO-d_6): 9.90. **HRMS** for $C_{19}H_{17}N_2O_5PCl^+$: calcd. $[M+H]^+$: 419.0558, found: 419.0505.

Dimethyl (4'-(3-chlorophenyl)-2-oxo-2',4'-dihydrospiro[indoline-3,3'-pyrazol]-5'-yl)phosphonate (3i)

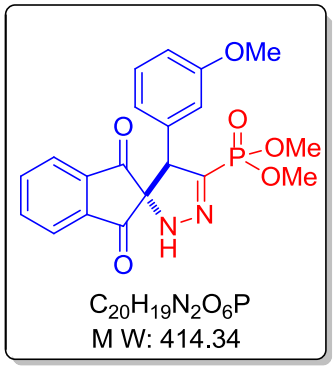


$C_{19}H_{16}ClN_2O_5P$
M W: 418.76

Following the general procedure, treatment of 2-(3-chlorobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (32 mg, 0.21 mmol) in the presence of CsF (3.0 mg, 0.019 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3i** as a yellow solid (74 mg, 93%). $\mathbf{R_f}$ (Acetone/Dichloromethane: 1/9) = 0.44. **Mp** 154-156 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 196.7 (C), 195.0 (C), 142.2 (d, $J_{C\text{-}P}$ = 228.5 Hz, C), 141.6 (C), 140.1 (C), 136.9 (CH), 136.7 (CH), 134.5 (C), 133.9 (C), 129.9 (CH), 129.6 (CH), 129.0 (CH), 127.8 (CH), 124.3 (CH), 123.7 (CH), 78.6 (d, $J_{C\text{-}P}$ = 3.8 Hz, C), 64.4 (d, $J_{C\text{-}P}$ = 23.7 Hz, CH), 53.7 (d, $J_{C\text{-}P}$ = 5.9 Hz, CH_3), 53.4 (d, $J_{C\text{-}P}$ = 5.8 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 8.01 (d, J = 7.6 Hz, 1H), 7.87 (t, J = 7.4 Hz, 1H), 7.78 (t, J = 7.6 Hz, 1H), 7.62 (d, J

= 7.6 Hz, 1H), 7.18-7.11 (m, 2H), 6.93-6.86 (m, 3H), 4.67 (s, 1H), 3.67 (d, *J* = 11.6 Hz, 3H), 3.62 (d, *J* = 11.2 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.55. **HRMS** for $C_{19}H_{17}N_2O_5PCl^+$: calcd. $[M+H]^+$: 419.0558, found: 419.0572.

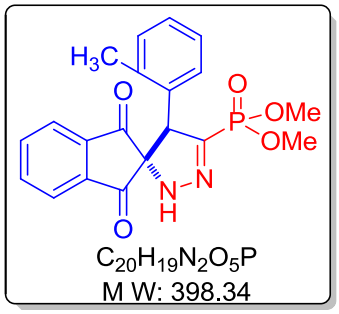
Dimethyl (4'-(3-methoxyphenyl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3j)



$C_{20}H_{19}N_2O_6P$
M W: 414.34

Following the general procedure, treatment of 2-(3-methoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (32 mg, 0.21 mmol) in the presence of CsF (3.0 mg, 0.019mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3j** as a yellow solid (67 mg, 86%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.44. **Mp** 134-136 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.1 (C), 195.3 (C), 159.5 (C), 142.3 (d, J_{C-P} = 227.6 Hz, C), 141.6 (C), 140.2 (C), 136.6 (CH), 136.3 (CH), 133.1 (C), 129.5 (CH), 124.0 (CH), 123.5 (CH), 121.8 (CH), 114.8 (CH), 114.6 (CH), 79.2 (d, J_{C-P} = 4.4 Hz, C), 65.2 (d, J_{C-P} = 23.8 Hz, CH), 55.2 (CH_3), 53.5 (d, J_{C-P} = 5.7 Hz, CH_3), 53.3 (d, J_{C-P} = 6.1 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.95(d, *J* = 7.6 Hz, 1H), 7.80 (t, *J* = 7.2 Hz, 1H), 7.72 (t, *J* = 7.4 Hz, 1H), 7.54 (d, *J* = 7.6 Hz, 1H), 7.02 (t, *J* = 7.8 Hz, 1H), 6.96 (s, 1H), 6.8 (d, *J* = 8.4 Hz, 1H), 6.46 (d, *J* = 10.4 Hz, 2H), 4.67 (s, 1H), 3.64 (s, 3H), 3.61 (d, *J* = 5.6 Hz, 3H), 3.61 (d, *J* = 6.0 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.08. **HRMS** for $C_{20}H_{20}N_2O_6P^+$: calcd. $[M+H]^+$: 415.1053, found: 415.1053.

Dimethyl (1,3-dioxo-4'-(o-tolyl)-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3k)

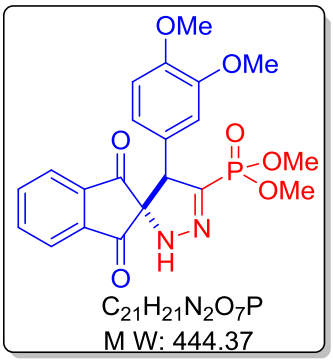


$C_{20}H_{19}N_2O_5P$
M W: 398.34

Following the general procedure, treatment of 2-(2-methylbenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.20 mmol) with SGR (33 mg, 0.22 mmol) in the presence of CsF (3.0 mg, 0.02 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3k** as a yellow solid (70 mg, 88%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.44. **Mp** 160-162 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.3 (C), 195.3 (C), 142.8 (d, J_{C-P} = 227.6 Hz, C), 141.3 (C), 140.1 (C), 136.7 (CH), 136.3 (CH), 135.9 (C), 130.9 (CH), 130.3 (CH), 129.9

(C), 128.5 (CH), 126.3 (CH), 123.8 (CH), 123.3 (CH), 78.7 (d, J_{C-P} = 4.5 Hz, C), 60.7 (d, J_{C-P} = 23.9 Hz, CH), 53.5 (d, J_{C-P} = 5.7 Hz, CH_3), 53.2 (d, J_{C-P} = 6.1 Hz, CH_3), 19.4 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.99 (d, *J* = 7.6 Hz, 1H), 7.83 (t, *J* = 7.4 Hz, 1H), 7.74 (d, *J* = 7.6 Hz, 1H), 7.56 (d, *J* = 7.6 Hz, 1H), 7.17 (d, *J* = 7.4 Hz, 1H), 7.12-7.03 (m, 2H), 6.86 (d, *J* = 7.6 Hz, 1H), 6.84 (s, 1H), 5.01 (s, 1H), 3.62 (d, *J* = 11.2 Hz, 3H), 3.56 (d, *J* = 11.2 Hz, 3H) 1.76 (s, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.02. **HRMS** for $C_{20}H_{20}N_2O_5P^+$: calcd. $[M+H]^+$: 399.1104, found: 399.1103. **CCDC No.** 1547228.

Dimethyl (4'-(3,4-dimethoxyphenyl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3l)

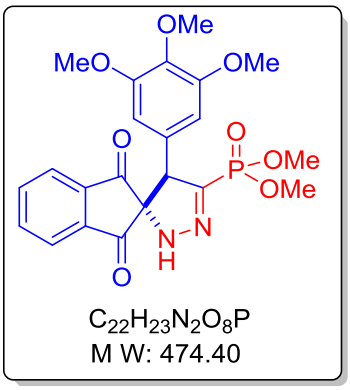


$C_{21}H_{21}N_2O_7P$
M W: 444.37

Following the general procedure, treatment of 2-(3,4-dimethoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.17 mmol) with SGR (29 mg, 0.19 mmol) in the presence of CsF (3.0 mg, 0.017 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3l** as a yellow solid (49 mg, 65%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.42. **Mp** 184-186 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.3 (C), 195.6 (C), 149.1 (C), 148.7 (C), 142.6 (d, J_{C-P} = 227.6 Hz, C), 141.5 (C), 140.4 (C), 136.6 (CH), 136.2 (CH), 124.0 (C), 123.9 (CH), 123.5 (CH), 122.1 (CH), 112.7 (CH), 110.7 (CH), 79.4 (d, J_{C-P} = 4.3 Hz, C), 65.3 (d, J_{C-P} = 24.0 Hz, CH), 55.9 (CH_3), 55.7 (CH_3), 53.6 (d, J_{C-P} = 5.7 Hz, CH_3), 53.4 (d, J_{C-P} = 6.0 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.99 (d, *J* = 7.6 Hz, 1H), 7.83 (t, *J* = 7.2 Hz, 1H), 7.75 (t, *J* = 7.4 Hz, 1H), 7.59 (d, *J* = 7.6 Hz, 1H), 6.75 (s, 1H), 6.61 (d, *J* = 8.8 Hz, 1H), 6.48-6.46 (m, 2H), 4.75 (s, 1H), 3.77 (s, 3H), 3.72 (s, 3H), 3.67 (d, *J* = 6.0 Hz, 3H), 3.64 (d, *J* = 6.0 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.13. **HRMS** for $C_{21}H_{22}N_2O_7P^+$: calcd. $[M+H]^+$: 445.1159, found: 445.1152.

Dimethyl (1,3-dioxo-4'-(3,4,5-trimethoxyphenyl)-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3m)

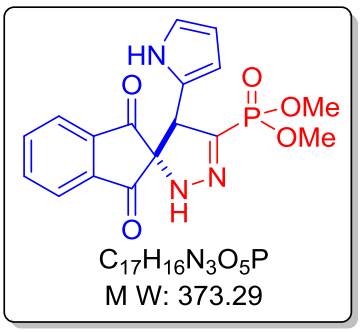
Following the general procedure, treatment of 2-(3,4,5 trimethoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.15 mmol) with SGR (26 mg, 0.17 mmol) in the presence of CsF (2.0



$C_{22}H_{23}N_2O_8P$
M W: 474.40

mg, 0.015 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3m** as a yellow solid (57 mg, 80%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.48. **Mp** 180-182 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.3 (C), 195.6 (C), 152.9 (C), 152.9 (C), 142.0 (d, J_{C-P} = 228.0 Hz, C), 141.5 (C), 140.6 (C), 137.9 (C), 136.7 (CH), 136.1 (CH), 127.0 (C), 123.7 (CH), 123.5 (CH), 106.9 (CH), 106.9 (CH), 79.8 (d, J_{C-P} = 4.0 Hz, C), 65.6 (d, J_{C-P} = 24.0 Hz, CH), 60.8 (CH_3), 56.6 (CH_3), 56.6 (CH_3), 53.6 (d, J_{C-P} = 5.6 Hz, CH_3), 53.3 (d, J_{C-P} = 6.1 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.95 (d, *J* = 7.6 Hz, 1H), 7.80 (t, *J* = 7.4 Hz, 1H), 7.72 (t, *J* = 7.4 Hz, 1H), 7.55 (d, *J* = 7.6 Hz, 1H), 6.95 (s, 1H), 6.09 (s, 2H), 4.70 (s, 1H), 3.68 (s, 3H), 3.66 (d, *J* = 5.2 Hz, 3H), 3.63 (d, *J* = 5.2 Hz, 3H), 3.61 (s, 6H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.10. **HRMS** for $C_{22}H_{24}N_2O_8P^+$: calcd. $[M+H]^+$: 475.1265, found: 475.1277.

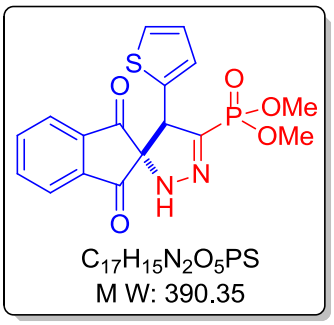
Dimethyl (1,3-dioxo-4'-(1H-pyrrol-2-yl)-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3n)



$C_{17}H_{16}N_3O_5P$
M W: 373.29

Following the general procedure, treatment of 2-(pyrrole-2-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.22 mmol) with SGR (38 mg, 0.25 mmol) in the presence of CsF (3.0 mg, 0.022 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3n** as a yellow solid (66 mg, 80%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.3. **Mp** 176-178 °C. **^{13}C NMR** (100 MHz, ppm/ DMSO-d_6): 198.8 (C), 151.9 (C), 136.9 (C), 134.3 (d, J_{C-P} = 232.0 Hz, C), 132.5 (C), 131.6 (C), 131.0 (CH), 124.9 (CH), 123.6 (CH), 114.6 (CH), 113.1 (CH), 100.7 (CH), 92.3 (CH), 87.0 (d, J_{C-P} = 4.2 Hz, C), 56.0 (d, J_{C-P} = 24.2 Hz, CH), 52.9 (d, J_{C-P} = 5.7 Hz, CH_3), 52.8 (d, J_{C-P} = 5.8 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/DMSO-d_6): 9.17 (s, 1H), 8.10 (d, *J* = 7.6 Hz, 1H), 7.87 (t, *J* = 7 Hz, 1H), 7.67 (d, *J* = 7.6 Hz, 1H), 7.68-7.62 (m, 2H), 7.24 (s, 1H), 6.20 (s, 1H), 5.74 (s, 1H), 4.73 (s, 1H), 3.68 (d, *J* = 11.2 Hz, 3H), 3.65 (d, *J* = 11.2 Hz, 3H). **^{31}P NMR** (161.9 MHz, DMSO-d_6): 11.10. **HRMS** for $C_{17}H_{17}N_3O_5P^+$: calcd. $[M+H]^+$: 374.0900, found: 374.0889.

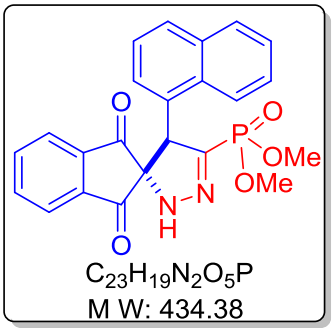
Dimethyl (1,3-dioxo-4'-(thiophen-2-yl)-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3o)



$C_{17}H_{15}N_2O_5PS$
M W: 390.35

Following the general procedure, treatment of 2-(thiophen-2-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.21 mmol) with SGR (35 mg, 0.23 mmol) in the presence of CsF (3.0 mg, 0.021 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3o** as a yellow solid (71 mg, 87%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.41. **Mp** 172-174 °C. **^{13}C NMR** (100 MHz, ppm/ DMSO-d_6): 197.2 (C), 194.7 (C), 141.0 (C), 140.0 (C), 139.1 (C), 136.9 (CH), 136.7 (CH), 134.3 (C), 128.6 (CH), 126.9 (CH), 126.8 (CH), 123.8 (CH), 123.0 (CH), 77.6 (d, $J_{C\text{-}P}$ = 3.7 Hz, C), 56.7 (d, $J_{C\text{-}P}$ = 23.6 Hz, CH), 53.0 (d, $J_{C\text{-}P}$ = 6.1 Hz, CH_3), 52.9 (d, $J_{C\text{-}P}$ = 5.8 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/DMSO-d_6): 8.68 (s, 1H), 8.06-7.92 (m, 3H), 7.76 (d, J = 7.6 Hz, 1H), 7.37 (d, J = 5.2 Hz, 1H), 6.90-6.88 (m, 1H), 6.77 (d, J = 3.2 Hz, 1H), 5.14 (s, 1H), 3.61 (d, J = 11.2 Hz, 3H), 3.57 (d, J = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, DMSO-d_6): 14.35. **HRMS** for $C_{17}H_{16}N_2O_5PS^+$: calcd. $[M+H]^+$: 391.0512, found: 391.0501.

Dimethyl (4'-(naphthalen-1-yl)-1,3-dioxo-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3p)

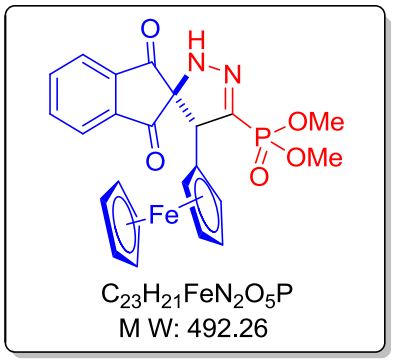


$C_{23}H_{19}N_2O_5P$
M W: 434.38

Following the general procedure, treatment of 2-(naphthalen-1-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.18 mmol) with SGR (30 mg, 0.20 mmol) in the presence of CsF (3.0 mg, 0.02 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3p** as a yellow solid (69 mg, 89%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.44. **Mp** 208-210 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 197.7 (C), 195.5 (C), 143.0 (d, $J_{C\text{-}P}$ = 228.0 Hz, C), 141.0 (C), 140.6 (C), 136.5 (CH), 136.0 (CH), 133.6 (C), 131.1 (C), 129.6 (CH), 129.3 (CH), 129.0 (CH), 127.4 (C), 126.3 (CH), 125.5 (CH), 125.3 (CH), 123.6 (CH), 123.1 (CH), 122.1 (C), 79.0 (d, $J_{C\text{-}P}$ = 3.8 Hz, C), 59.6 (d, $J_{C\text{-}P}$ = 23.5 Hz, CH), 53.5 (d, $J_{C\text{-}P}$ = 5.7 Hz, CH_3), 53.4 (d, $J_{C\text{-}P}$ = 5.7 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.91 (d, J = 7.6 Hz, 1H), 7.71-7.64 (m, 3H), 7.53-7.46 (m, 2H), 7.42 (d, J = 6.8 Hz, 1H), 7.35 (d, J = 8.4 Hz, 1H), 7.12-7.03 (m, 2H), 7.11-7.07 (m, 1H), 6.83 (s, 1H), 5.73 (s, 1H), 3.69 (d, J = 11.6 Hz, 3H), 3.61 (d, J =

11.2 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.98. **HRMS** for $C_{23}H_{20}N_2O_5P^+$: calcd. $[M+H]^+$: 435.1104, found: 435.1089.

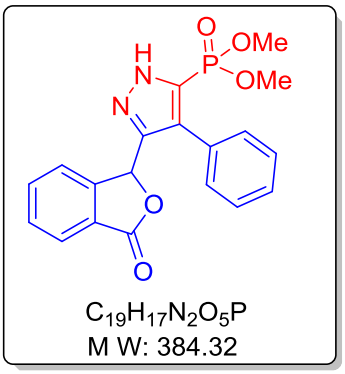
Dimethyl (1,3-dioxo-4'-(ferrocenyl)-1,2',3,4'-tetrahydrospiro[indene-2,3'-pyrazol]-5'-yl)phosphonate (3q)



$C_{23}H_{21}FeN_2O_5P$
M W: 492.26

Following the general procedure, treatment of 2-(ferrocenylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.15 mmol) with SGR (25 mg, 0.17 mmol) in the presence of CsF (2.0 mg, 0.02 mmol) in acetone (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **3q** as a yellow solid (64 mg, 87%). **R_f** (Acetone/Dichloromethane : 1/9) = 0.44. **Mp** 220-222 °C. **^{13}C NMR** (100 MHz, ppm/ DMSO-d_6): 197.9 (C), 195.7 (C), 141.2 (C), 140.2 (C), 137.2 (d, J_{C-P} = 226.1 Hz, C), 136.8 (CH), 136.3 (CH), 123.3 (CH), 122.7 (CH), 80.3 (CH), 77.0 (d, J_{C-P} = 4.1 Hz, C), 69.2 (CH), 68.3 (CH), 68.3 (CH), 68.3 (CH), 68.3 (CH), 68.3 (CH), 67.1 (CH), 66.7 (CH), 66.1 (CH), 57.2 (d, J_{C-P} = 24.0 Hz, CH), 52.7 (d, J_{C-P} = 6.4 Hz, CH_3), 52.4 (d, J_{C-P} = 5.7 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/DMSO-d_6): 8.66 (s, 1H), 8.11 (s, 1H), 8.06 (s, 2H), 7.92 (s, 1H), 4.74 (s, 1H), 4.19 (s, 1H), 4.12 (s, 1H), 3.94 (s, 6H), 3.71 (d, J = 11.2 Hz, 3H), 3.65 (d, J = 11.2 Hz, 3H) 3.45 (s, 1H). **^{31}P NMR** (161.9 MHz, DMSO-d_6): 11.14. **HRMS** for $C_{23}H_{22}N_2O_5PFe^+$: calcd. $[M+H]^+$: 493.0610, found: 493.0598.

Dimethyl (4-phenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4a)

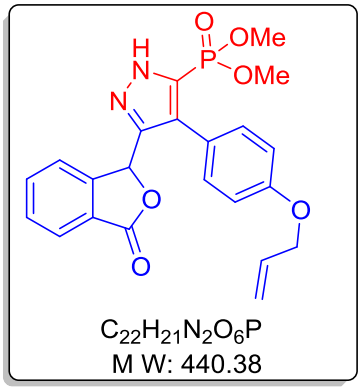


$C_{19}H_{17}N_2O_5P$
M W: 384.32

Following the general procedure, treatment of 2-(benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.21 mmol) with SGR (48 mg, 0.32 mmol) in the presence of NaOH (21.0 mg, 0.53 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4a** as a yellow solid (69 mg, 86%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.42. **Mp** 162-164 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 148.1 (C), 146.0 (C), 134.1 (CH), 130.0 (CH), 130.0 (CH), 129.9 (C), 129.3 (CH), 128.5 (C), 128.3 (C), 128.2 (CH), 128.2 (CH), 128.2 (CH), 126.3 (C), 125.4 (CH), 123.1 (CH), 76.0 (CH), 53.4 (CH_3),

53.4 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.70 (s, 1H), 7.76 (d, *J* = 7.6 Hz, 1H), 7.61 (t, *J* = 7.6 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.33-7.27 (m, 6H), 6.53 (s, 1H), 3.67 (d, *J* = 11.6 Hz, 3H), 3.58 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.52. **HRMS** for $C_{19}H_{18}N_2O_5P^+$: calcd. $[M+H]^+$: 385.0948, found: 385.0944.

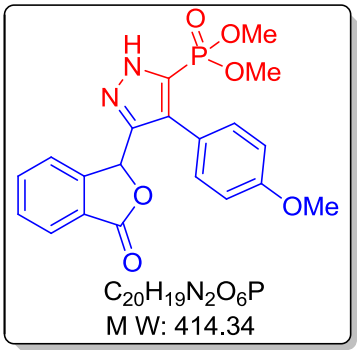
Dimethyl (4-(4-(allyloxy)phenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4b)



$C_{22}H_{21}N_2O_6P$
M W: 440.38

Following the general procedure, treatment of 2-(4-(allyloxy)benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.17 mmol) with SGR (39.0 mg, 0.26 mmol) in the presence of NaOH (17.0 mg, 0.43 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4b** as a yellow solid (55 mg, 73%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.31. **Mp** 142-144 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 158.5 (C), 148.1 (C), 134.1 (CH), 133.2 (CH), 131.2 (CH), 131.2 (CH), 129.3 (CH), 128.3 (C), 128.1 (C), 126.3 (C), 125.4 (CH), 123.1 (CH), 122.1 (C), 117.9 (CH_2), 114.5 (CH), 114.5 (CH), 76.0 (CH), 68.8 (CH_2), 53.4 (CH_3), 53.4 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.83 (s, 1H), 7.75 (d, *J* = 7.6 Hz, 1H), 7.56 (t, *J* = 7.4 Hz, 1H), 7.44 (t, *J* = 7.4 Hz, 1H), 7.28 (d, *J* = 7.6 Hz, 1H), 7.17 (d, *J* = 8.4 Hz, 2H), 6.83 (d, *J* = 8.4 Hz, 2H), 6.48 (s, 1H), 6.07-6.01 (m, 1H), 5.36 (dd, *J* = 48.0, 11.2 Hz, 2H), 4.52 (d, *J* = 4.8 Hz, 2H), 3.64 (d, *J* = 11.6 Hz, 3H), 3.56 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.88. **HRMS** for $C_{22}H_{22}N_2O_6P^+$: calcd. $[M+H]^+$: 441.1210, found: 441.1211.

Dimethyl (4-(4-methoxyphenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4c)

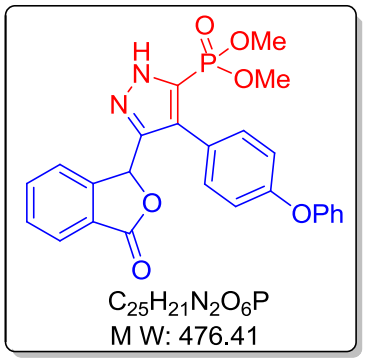


$C_{20}H_{19}N_2O_6P$
M W: 414.34

Following the general procedure, treatment of 2-(4-methoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (44.0 mg, 0.29 mmol) in the presence of NaOH (19.0 mg, 0.48 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4c** as a yellow solid (46 mg, 58%). **R_f** (Acetone/Dichloromethane:

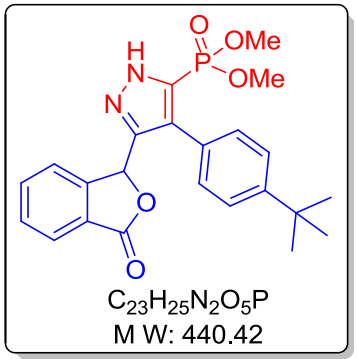
2/8) = 0.44. **Mp** 170-172 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 159.5 (C), 148.1 (C), 145.9 (C), 134.0 (CH), 131.2 (CH), 131.2 (CH), 129.3 (CH), 128.3 (C), 128.2 (C), 126.3 (C), 125.4 (CH), 123.1 (CH), 121.9 (C), 113.7 (CH), 113.7 (CH), 75.9 (CH), 55.3 (CH_3), 53.3 (CH_3), 53.3 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.76 (d, *J* = 7.6 Hz, 1H), 7.59 (t, *J* = 7.4 Hz, 1H), 7.46 (t, *J* = 7.4 Hz, 1H), 7.31 (d, *J* = 7.6 Hz, 1H), 7.20 (d, *J* = 8.4 Hz, 2H), 6.84 (d, *J* = 8.8 Hz, 2H), 6.51 (s, 1H), 3.82 (s, 3H), 3.67 (d, *J* = 11.6 Hz, 3H), 3.58 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.01. **HRMS** for $C_{20}H_{20}N_2O_6P^+$: calcd. $[M+H]^+$: 415.1053, found: 415.1046.

Dimethyl (3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-4-(4-phenoxyphenyl)-1H-pyrazol-5-yl)phosphonate (4d)



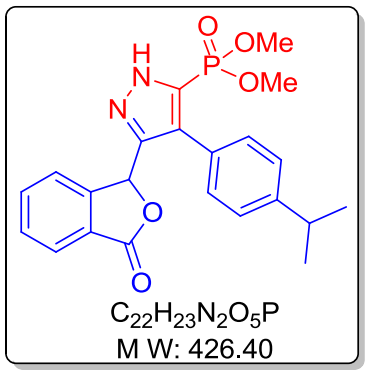
Following the general procedure, treatment of 2-(4-phenoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.15 mmol) with SGR (35 mg, 0.23 mmol) in the presence of NaOH (15.0 mg, 0.38 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4d** as a yellow solid (56 mg, 79%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.38. **Mp** 167-169 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.2 (C), 157.1 (C), 156.9 (C), 148.0 (C), 134.1 (CH), 131.6 (C), 129.9 (CH), 129.9 (CH), 129.6 (CH), 129.4 (CH), 127.6 (C), 127.4 (C), 126.3 (C), 125.4 (CH), 124.9 (CH), 123.6 (CH), 123.1 (CH), 120.1 (CH), 119.1 (CH), 119.1 (CH), 118.5 (CH), 76.1 (CH), 53.4 (d, $J_{C\text{-}P}$ = 4.9 Hz, CH_3), 53.4 (d, $J_{C\text{-}P}$ = 4.9 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.95 (s, 1H), 7.80 (d, *J* = 7.6 Hz, 1H), 7.63 (t, *J* = 7.2 Hz, 1H), 7.51 (t, *J* = 7.6 Hz, 1H), 7.39-7.28 (m, 4H), 7.15 (t, *J* = 7.4 Hz, 1H), 7.02-6.96 (m, 4H), 6.82 (s, 1H), 6.56 (s, 1H), 3.63 (d, *J* = 11.6 Hz, 3H), 3.59 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.43. **HRMS** for $C_{25}H_{22}N_2O_6P^+$: calcd. $[M+H]^+$: 477.1210, found: 477.1234.

Dimethyl (4-(4-(tert-butyl)phenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4e)



Following the general procedure, treatment of 2-(4-(tert-butyl)benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.17 mmol) with SGR (39 mg, 0.26 mmol) in the presence of NaOH (17.0 mg, 0.43 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4e** as a yellow solid (58 mg, 77%). $\mathbf{R_f}$(Acetone/Dichloromethane : 2/8) = 0.41. **Mp** 190-192 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 151.1 (C), 148.1 (C), 134.0 (CH), 129.7 (CH), 129.7 (CH), 129.3 (C), 128.7 (C), 128.5 (C), 126.7 (C), 126.5 (CH), 125.4 (CH), 125.1 (CH), 125.1 (CH), 123.2 (CH), 76.0 (CH), 53.4 (CH_3), 53.4 (CH_3), 34.7 (C), 31.4 (CH_3), 31.4 (CH_3), 31.4 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.64 (s, 1H), 7.73 (d, *J* = 7.6 Hz, 1H), 7.57 (t, *J* = 7.4 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 1H), 7.29 (t, *J* = 7.8 Hz, 3H), 7.20 (d, *J* = 8.0 Hz, 2H), 6.50 (s, 1H), 3.65 (d, *J* = 11.6 Hz, 3H), 3.57 (d, *J* = 11.6 Hz, 3H), 1.31 (s, 9 H).**^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.88. **HRMS** for $C_{23}H_{26}N_2O_5P^+$: calcd. $[M+H]^+$: 441.1574, found: 441.1570.

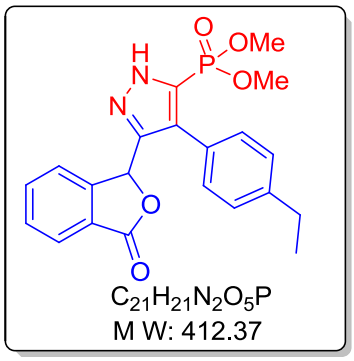
Dimethyl (4-(4-(isopropyl)phenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4f)



Following the general procedure, treatment of 2-(4-(isopropyl)benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.18 mmol) with SGR (41 mg, 0.27 mmol) in the presence of NaOH (18.0 mg, 0.45 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4f** as a yellow solid (61 mg, 80%). $\mathbf{R_f}$(Acetone/Dichloromethane: 2/8) = 0.44. **Mp** 178-180 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 148.8 (C), 148.1 (C), 134.0 (CH), 129.9 (CH), 129.9 (CH), 129.2 (CH), 128.7 (C), 128.5 (C), 127.1 (C), 126.3 (C), 126.3 (CH), 126.3 (CH), 125.3 (CH), 123.1 (CH), 76.0 (CH), 53.4 (d, $J_{C\text{-}P}$ = 5.1 Hz, CH_3), 53.3 (d, $J_{C\text{-}P}$ = 18.0 Hz, CH_3), 33.9 (CH), 24.0 (CH_3), 24.0 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.72 (s, 1H), 7.72 (d, *J* = 7.6 Hz, 1H), 7.56 (t, *J* = 7.6 Hz, 1H), 7.42 (t, *J* = 7.4 Hz, 1H), 7.27 (s, 1H), 7.17 (d, *J* = 8.4 Hz, 2H), 7.12 (d, *J* = 8.4 Hz, 2H), 6.48 (s, 1H), 3.64

(d, *J* = 11.6 Hz, 3H), 3.55 (d, *J* = 11.6 Hz, 3H), 2.92-2.85 (m, 1H), 1.23 (d, *J* = 7.2 Hz, 6H).**^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.85. **HRMS** for $C_{22}H_{24}N_2O_5P^+$: calcd. $[M+H]^+$: 427.1417, found: 427.1426.

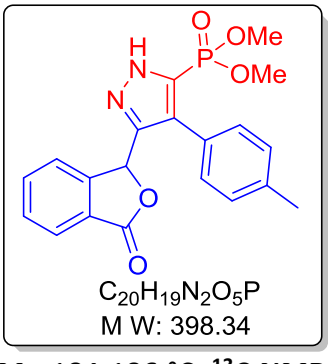
Dimethyl (4-(4-(ethyl)phenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4g)



$C_{21}H_{21}N_2O_5P$
M W: 412.37

Following the general procedure, treatment of 2-(4-(ethyl)benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (44 mg, 0.29 mmol) in the presence of NaOH (19.0 mg, 0.48 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4g** as a yellow solid (75 mg, 96%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.35. **Mp** 158-160 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 148.1 (C), 144.2 (C), 134.0 (CH), 130.0 (CH), 130.0 (CH), 129.3 (CH), 128.7 (C), 128.5 (C), 127.7 (CH), 127.7 (CH), 127.0 (C), 126.4 (C), 125.4 (CH), 123.1 (CH), 75.9 (CH), 53.4 (CH_3), 53.4 (CH_3), 28.7 (CH_2), 15.6 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.85 (s, 1H), 7.73 (d, *J* = 7.6 Hz, 1H), 7.56 (t, *J* = 7.4 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 1H), 7.27 (t, *J* = 8.4 Hz, 1H), 7.17 (d, *J* = 8.0 Hz, 2H), 7.11 (d, *J* = 8.0 Hz, 2H), 6.49 (s, 1H), 3.64 (d, *J* = 11.6 Hz, 3H), 3.55 (d, *J* = 11.6 Hz, 3H), 2.61 (q, *J* = 7.6 Hz, 2H), 1.22 (t, *J* = 7.4 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.04. **HRMS** for $C_{21}H_{22}N_2O_5^+$: calcd. $[M+H]^+$: 413.1261, found: 413.1269.

Dimethyl (3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-4-(p-tolyl)-1H-pyrazol-5-yl)phosphonate (4h)

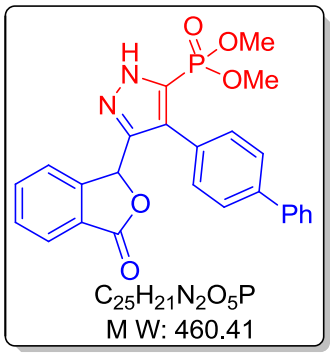


$C_{20}H_{19}N_2O_5P$
M W: 398.34

Following the general procedure, treatment of 2-(4-(methyl)benzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.20 mmol) with SGR (45 mg, 0.30 mmol) in the presence of NaOH (20.0 mg, 0.50 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4h** as a yellow solid (68 mg, 86%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.38. **Mp** 184-186 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 148.1 (C), 138.0 (C), 134.1 (CH), 129.9 (CH), 129.9 (CH), 129.3 (C), 129.0 (CH), 129.0 (CH), 128.7 (d, J_{C-P} = 18.0 Hz, C), 126.8 (C), 126.4 (C), 125.5 (CH), 123.2 (CH), 76.0 (CH), 53.4 (CH_3), 53.4 (CH_3), 21.4 (CH_3). **^{1}H NMR**

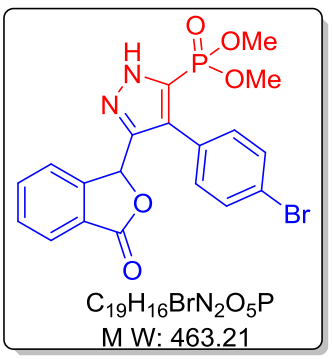
(400 MHz, ppm/$CDCl_3$): 7.76 (d, *J* = 7.2 Hz, 1H), 7.59 (t, *J* = 7.4 Hz, 1H), 7.46 (t, *J*= 7.4 Hz, 1H), 7.29 (d, *J* = 7.6 Hz, 1H), 7.18 (d, *J* = 8.0 Hz, 2H), 7.11 (d, *J* = 7.6 Hz, 2H), 6.47 (s, 1H), 3.66 (d, *J* = 11.6 Hz, 3H), 3.56 (d, *J* = 11.6 Hz, 3H), 2.34 (s, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.82. **HRMS** for $C_{20}H_{20}N_2O_5P^+$: calcd. $[M+H]^+$: 399.1104, found: 399.1104.

Dimethyl (4-([1,1'-biphenyl]-4-yl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4i)



Following the general procedure, treatment of 2-([1,1'-biphenyl]-4-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.16 mmol) with SGR (36.0 mg, 0.24 mmol) in the presence of NaOH (16.0 mg, 0.40 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4i** as a yellow solid (70 mg, 95%). **R_f** (Acetone/Dichloromethane: 2/8 = 0.38). **Mp** 138-140 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 148.0 (C), 146.2 (C), 140.8 (C), 140.5 (C), 134.1 (CH), 130.4 (CH), 130.4 (CH), 129.3 (CH), 128.9 (CH), 128.9 (CH), 128.9 (C), 128.2 (C), 128.0 (C), 127.6 (CH), 127.1 (CH), 127.1 (CH), 126.9 (CH), 126.9 (CH), 126.4 (C), 125.4 (CH), 123.2 (CH), 76.0 (CH), 53.5 (CH_3), 53.5 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.84 (s, 1H), 7.74 (d, *J* = 7.6 Hz, 1H), 7.58 (t, *J* = 7.8 Hz, 3H), 7.53 (d, *J* = 8.0 Hz, 2H), 7.46 (t, *J* = 7.4 Hz, 3H), 7.38-7.30 (m, 4H), 6.54 (s, 1H), 3.69 (d, *J* = 11.6 Hz, 3H), 3.61 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.76. **HRMS** for $C_{25}H_{22}N_2O_5P^+$: calcd. $[M+H]^+$: 461.1261, found: 461.1214.

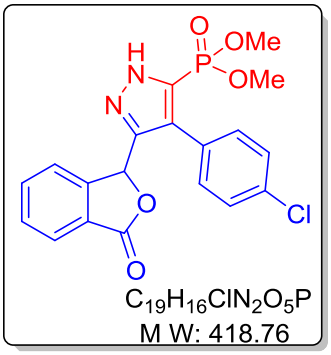
Dimethyl (4-(4-bromophenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4j)



Following the general procedure, treatment of 2-(4-bromobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.16 mmol) with SGR (36 mg, 0.24 mmol) in the presence of NaOH (16.0 mg, 0.40 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4j** as a yellow solid (55 mg, 74%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.45. **Mp** 158-160 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.2 (C), 147.8 (C), 134.2 (CH), 131.6 (CH), 131.6 (CH), 131.5 (CH), 131.5 (CH), 129.5 (CH), 128.8 (C), 127.2 (C), 127.1 (C), 126.3 (C),

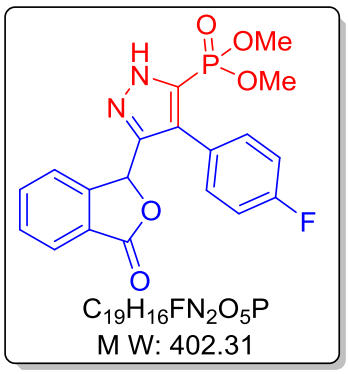
125.5 (CH), 123.2 (CH), 122.6 (C), 75.8 (CH), 53.5 (CH_3), 53.5 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 13.04 (s, 1H), 7.75 (d, *J* = 7.6 Hz, 1H), 7.58 (t, *J* = 7.4 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.40 (d, *J* = 8.4 Hz, 2H), 7.28 (d, *J* = 7.6 Hz, 1H), 7.10 (d, *J* = 8.4 Hz, 2H), 6.47 (s, 1H), 3.66 (d, *J* = 11.6 Hz, 3H), 3.58 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.26. **HRMS** for $C_{19}H_{17}BrN_2O_5P^+$: calcd. $[M+H]^+$: 463.0053, found: 463.0053.

Dimethyl (4-(4-chlorophenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4k)



Following the general procedure, treatment of 2-(4-chlorobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (42 mg, 0.28 mmol) in the presence of NaOH (19.0 mg, 0.48 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4k** as a yellow solid (48 mg, 60%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.40. **Mp** 208-210 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.1 (C), 147.8 (C), 134.5 (C), 134.2 (CH), 131.3 (CH), 131.3 (CH), 129.6 (CH), 128.6 (CH), 128.6 (CH), 128.3 (C), 127.3 (C), 127.1 (C), 126.3 (C), 125.6 (CH), 123.2 (CH), 75.9 (CH), 53.5 (CH_3), 53.5 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.78 (d, *J* = 7.6 Hz, 1H), 7.60 (t, *J* = 7.4 Hz, 1H), 7.49 (t, *J* = 7.6 Hz, 1H), 7.31 (d, *J* = 7.6 Hz, 1H), 7.28 (d, *J* = 4.8 Hz, 2H), 7.19 (d, *J* = 8.8 Hz, 2H), 6.49 (s, 1H), 3.68 (d, *J* = 11.6 Hz, 3H), 3.59 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.20. **HRMS** for $C_{19}H_{17}ClN_2O_5P^+$: calcd. $[M+H]^+$: 419.0558, found: 419.0507.

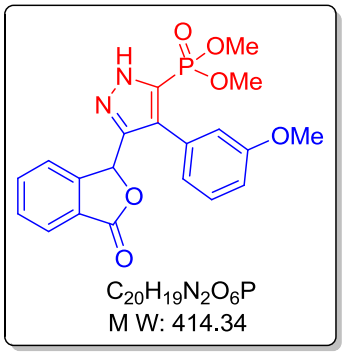
Dimethyl (4-(4-fluorophenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4l)



Following the general procedure, treatment of 2-(4-fluoroobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.20 mmol) with SGR (45 mg, 0.30 mmol) in the presence of NaOH (20.0 mg, 0.50 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4l** as a yellow solid (50 mg, 62%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.27. **Mp** 168-170 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.1 (C), 162.7 (d, $J_{C\text{-}F}$ = 246.6 Hz, C), 147.8 (C), 146.1 (C), 134.2 (CH), 131.8 (d, $J_{C\text{-}F}$ = 8.1 Hz, CH), 131.7

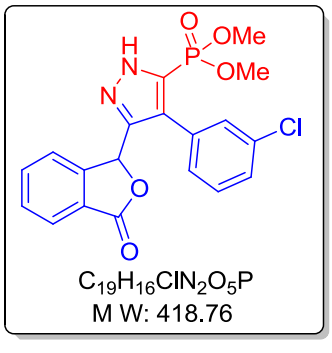
(d, $J_{C\text{-}F}$ = 8.1 Hz, CH), 129.5 (CH), 127.4 (C), 127.2 (C), 126.3 (C), 125.7 (C), 125.5 (CH), 123.1 (CH), 115.4 (d, $J_{C\text{-}F}$ = 21.5 Hz CH), 115.3 (d, $J_{C\text{-}F}$ = 21.5 Hz, CH), 76.0 (CH), 53.4 (CH_3), 53.4 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 13.04 (s, 1H), 7.75 (d, *J* = 7.6 Hz, 1H), 7.58 (t, *J* = 7.4 Hz, 1H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.40 (d, *J* = 8.4 Hz, 2H), 7.28 (d, *J* = 7.6 Hz, 1H), 7.10 (d, *J* = 8.0 Hz, 2H), 6.47 (s, 1H), 3.66 (d, *J* = 11.6 Hz, 3H), 3.58 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.34. **HRMS** for $C_{19}H_{17}FN_2O_5P^+$: calcd. $[M+H]^+$: 403.0854, found: 403.0853.

Dimethyl (4-(3-methoxyphenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4m)



Following the general procedure, treatment of 2-(3-methoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (44.0 mg, 0.29 mmol) in the presence of NaOH (19.0 mg, 0.48 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4m** as a yellow solid (61 mg, 78%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.46. **Mp** 162-164 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 159.2 (C), 148.0 (C), 134.0 (CH), 131.0 (CH), 129.3 (C), 129.3 (CH), 128.3 (C), 128.3 (C), 126.3 (C), 125.4 (CH), 123.1 (CH), 122.4 (C), 115.2 (CH), 114.2 (CH), 76.0 (CH), 55.3 (CH_3), 53.3 (CH_3), 53.3 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.78 (s, 1H), 7.75 (d, *J* = 8.0 Hz, 1H), 7.58 (d, *J* = 7.4 Hz, 1H), 7.45 (t, *J* = 7.6 Hz, 1H), 7.29 (d, *J* = 7.6 Hz, 1H), 7.19 (d, *J* = 8.0 Hz, 1H), 6.85-6.80 (m, 3H), 6.52 (s, 1H), 3.77 (s, 3H), 3.66 (d, *J* = 11.6 Hz, 3H), 3.58 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.67. **HRMS** for $C_{20}H_{20}N_2O_6P^+$: calcd. $[M+H]^+$: 415.1053, found: 415.1049.

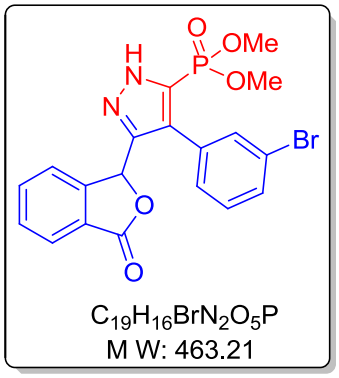
Dimethyl (4-(3-chlorophenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4n)



Following the general procedure, treatment of 2-(3-chlorobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (42 mg, 0.28 mmol) in the presence of NaOH (19.0 mg, 0.48 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4n** as a yellow solid (55 mg, 69%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.35. **Mp** 134-136 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.1 (C), 147.8 (C), 146.0 (C), 134.2 (CH), 133.9 (C),

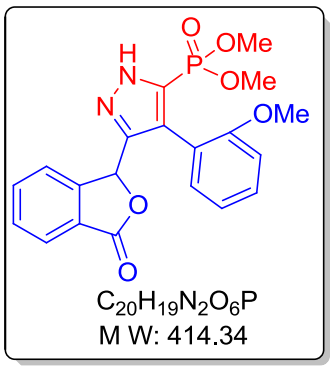
131.8 (C), 129.9 (CH), 129.5 (CH), 129.5 (CH), 128.2 (CH), 126.7 (C), 126.5 (C), 126.2 (C), 125.4 (CH), 123.0 (CH), 76.0 (CH), 53.5 (d, $J_{C\text{-}P}$ = 2.8 Hz, CH_3), 53.4 (d, $J_{C\text{-}P}$ = 2.9 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.72 (d, *J* = 7.6 Hz, 1H), 7.59 (t, *J* = 7.4 Hz, 1H), 7.46 (t, *J* = 7.4 Hz, 1H), 7.29 (d, *J* = 7.6 Hz, 1H), 7.22 (d, *J* = 7.6 Hz, 2H), 7.12 (d, *J* = 6.8 Hz, 1H), 7.04 (s, 1H), 6.51 (s, 1H), 3.67 (d, *J* = 11.6 Hz, 3H), 3.59 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 7.74. **HRMS** for $C_{19}H_{17}ClN_2O_5P^+$: calcd. $[M+H]^+$: 419.0558, found: 419.0551.

Dimethyl (4-(3-bromophenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4o)



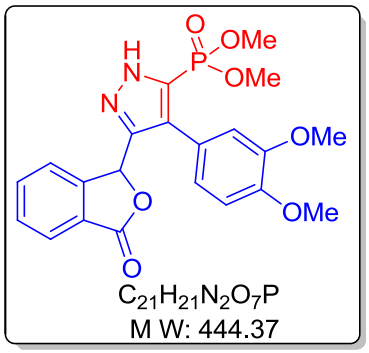
Following the general procedure, treatment of 2-(3-bromobenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.16 mmol) with SGR (36 mg, 0.24 mmol) in the presence of NaOH (16.0 mg, 0.40 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4o** as a yellow solid (51 mg, 69%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.38. **Mp** 160-162 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.1 (C), 147.8 (C), 146.2 (C), 134.2 (CH), 132.8 (CH), 132.0 (CH), 131.2 (C), 129.7 (CH), 129.6 (CH), 128.7 (CH), 126.6 (C), 126.4 (C), 126.2 (C), 125.5 (CH), 123.0 (CH), 122.0 (C), 76.1 (CH), 53.5 (d, $J_{C\text{-}P}$ = 2.4 Hz, CH_3), 53.5 (d, $J_{C\text{-}P}$ = 2.7 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.73 (d, *J* = 7.6 Hz, 1H), 7.60 (t, *J* = 7.2 Hz, 1H), 7.46 (t, *J* = 7.4 Hz, 1H), 7.40-7.37 (m, 1H), 7.29 (d, *J* = 7.6 Hz, 1H), 7.18-7.13 (m, 3H), 6.51 (s, 1H), 3.67 (d, *J* = 11.6 Hz, 3H), 3.59 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.88. **HRMS** for $C_{19}H_{17}BrN_2O_5P^+$: calcd. $[M+H]^+$: 463.0053, found: 463.0053.

Dimethyl (4-(2-methoxyphenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4p)



Following the general procedure, treatment of 2-(2-methoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.19 mmol) with SGR (44.0 mg, 0.29 mmol) in the presence of NaOH (16.0 mg, 0.48 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4p** as a white solid (59 mg, 75%). $\mathbf{R_f}$ (Acetone/Dichloromethane: 2/8) = 0.46. **Mp** 170-172 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 156.8 (C), 148.0 (C), 133.6 (CH), 132.1 (CH), 130.0 (CH), 129.1 (CH), 126.1 (C), 125.0 (CH), 123.2 (CH), 120.2 (CH), 118.7 (CH), 110.2 (C), 75.9 (CH), 55.1 (CH_3), 53.4 (CH_3), 53.3 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.77 (s,1H), 7.69 (s, 1H), 7.55 (t, *J* = 7.0 Hz, 1H), 7.42 (t, *J* = 7.4 Hz, 1H), 7.33 (d, *J* = 7.6 Hz, 1H), 7.29-7.22 (m, 2H), 6.86 (s, 1H), 6.52 (s, 1H), 3.64 (s, 3H), 3.59 (d, J = 11.6 Hz, 6H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.53. **HRMS** for $C_{20}H_{20}N_2O_6P^+$: calcd. $[M+H]^+$: 415.1053, found: 415.1030.

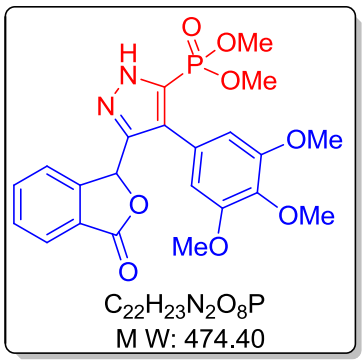
Dimethyl (4-(3,4-dimethoxyphenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4q)



Following the general procedure, treatment of 2-(3,4-dimethoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.17 mmol) with SGR (39 mg, 0.26 mmol) in the presence of NaOH (17.0 mg, 0.43 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4q** as a yellow solid (50 mg, 67%). $\mathbf{R_f}$ (Acetone/Dichloromethane: 2/8) = 0.31. **Mp** 182-184 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 148.9 (C), 148.5 (C), 147.9 (C), 145.7 (C), 134.0 (CH), 129.3 (C), 128.5 (C), 128.2 (C), 126.4 (CH), 125.4 (CH), 123.1 (CH), 122.5 (CH), 122.2 (C), 113.1 (CH), 110.8 (CH), 76.0 (CH), 55.9 (CH_3), 55.9 (CH_3), 53.4 (CH_3), 53.4 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.91 (s, 1 H), 7.74 (d, *J* = 7.6 Hz, 1H), 7.56 (t, *J* = 7.4 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 1H), 7.28 (d, *J* = 7.6 Hz, 1H), 6.86-6.77 (m, 3H), 6.51 (s, 1H), 3.86 (s, 3H), 3.81 (s, 3H), 3.65 (d, *J* = 11.6 Hz, 3H), 3.57 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR**

(161.9 MHz, $CDCl_3$): 9.07. **HRMS** for $C_{21}H_{22}N_2O_7P^+$: 445.1159, calcd. $[M+H]^+$: found: 445.1155. **CCDC No**. 1547535.

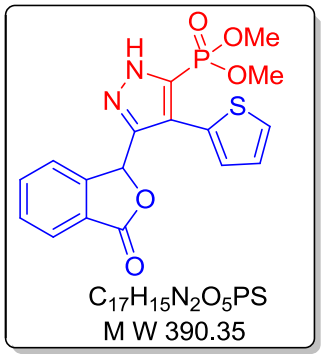
Dimethyl (4-(3,4,5-triimethoxyphenyl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4r)



$C_{22}H_{23}N_2O_8P$
M W: 474.40

Following the general procedure, treatment of 2-(3,4,5-trimethoxybenzylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.15 mmol) with SGR (35.0 mg, 0.23 mmol) in the presence of NaOH (15.0 mg, 0.38 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4r** as a yellow solid (48 mg, 67%). $\mathbf{R_f}$(Acetone/Dichloromethane: 2/8) = 0.30. **Mp** 184-186 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.2 (C), 152.8 (C), 152.8 (C), 152.8 (C), 147.8 (C), 137.8 (C), 134.0 (CH), 129.4 (C), 128.5 (C), 128.4 (C), 126.5 (CH), 125.4 (CH), 125.2 (C), 123.2 (CH), 107.3 (CH), 107.3 (CH), 76.1 (CH), 61.0 (CH_3), 56.2 (CH_3), 56.2 (CH_3), 53.5 (CH_3), 53.5 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.13 (s, 1H), 7.77 (d, *J* = 8.0 Hz, 1H), 7.59 (t, *J* = 7.6 Hz, 1H), 7.47 (t, *J* = 7.4 Hz, 1H), 7.30 (d, *J* = 7.6 Hz, 1H), 6.56 (s, 1H), 6.51 (s, 2H), 3.84 (s, 3H), 3.81 (s, 6H) 3.68 (d, *J* = 11.6 Hz, 3H), 3.63 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.07. **HRMS** for $C_{22}H_{24}N_2O_8P^+$: calcd. $[M+H]^+$: 475.1265, found: 475.1268.

Dimethyl (3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-4-(thiophen-2-yl)-1H-pyrazol-5-yl)phosphonate (4s)

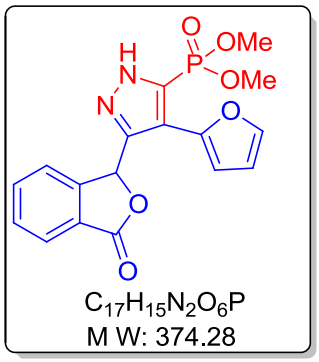


$C_{17}H_{15}N_2O_5PS$
M W 390.35

Following the general procedure, treatment of (2-(thiophen-2-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.21 mmol) with SGR (48.0 mg, 0.32 mmol) in the presence of NaOH (21.0 mg, 0.53 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4s** as a yellow solid (48 mg, 58%). $\mathbf{R_f}$(Acetone/Dichloromethane: 1/9) = 0.40. **Mp** 124-126 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.3 (C), 147.8 (C), 134.2 (CH), 129.5 (CH), 129.5 (CH), 129.4 (C), 127.3 (CH), 127.3 (CH), 126.3 (C), 125.5 (CH), 123.2 (CH), 120.7 (C), 120.5 (C), 75.8 (CH), 53.6 (CH_3), 53.6 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 12.92 (s, 1H), 7.79 (d, *J* = 7.6 Hz, 1H), 7.61 (t, *J* = 7.4 Hz, 1H), 7.48 (t, *J* = 7.6 Hz, 1H), 7.33 (t, *J* = 7.0

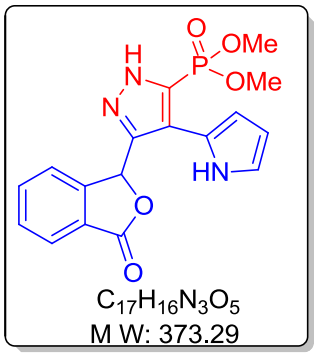
Hz, 2H), 7.01 (s, 1H), 6.99 (d, *J* = 6.4 Hz, 1H), 6.59 (s, 1H), 3.70 (d, *J* = 11.6 Hz, 3H), 3.64 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 7.76. **HRMS** for $C_{17}H_{16}N_2O_5PS^+$: calcd. $[M+H]^+$: 391.0512, found: 391.0502.

Dimethyl (4-(furan-2-yl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4t)



Following the general procedure, treatment of (2-(furan-2-ylmethylene)-1H-indene-1,3(2H)-dione (50 mg, 0.22 mmol) with SGR (50 mg, 0.33 mmol) in the presence of NaOH (22.0 mg, 0.55 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4t** as a yellow solid (68 mg, 83%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.38. **Mp** 178-180 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.6 (C), 148.0 (C), 144.8 (C), 144.3 (C), 142.7 (CH), 134.2 (CH), 129.5 (CH), 126.2 (C), 125.5 (CH), 123.3 (CH), 117.5 (C), 117.3 (C), 111.7(CH), 110.2 (CH), 76.1 (CH), 53.6 (CH_3), 53.6 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.92 (d, *J* = 7.2 Hz, 1H), 7.62 (d, *J* = 7.2 Hz, 1H), 7.54 (d, *J* = 7.2 Hz, 1H), 7.38 (t, *J* = 7.2 Hz, 2H), 6.88 (s, 1H), 6.71 (s, 1H), 6.43 (s, 1H), 3.75 (t, *J* = 11.6 Hz, 6H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.58. **HRMS** for $C_{17}H_{16}N_2O_6P^+$: calcd. $[M+H]^+$: 375.0734, found: 375.0740.

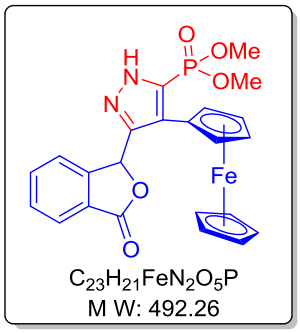
Dimethyl (3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-4-(1H-pyrrol-2-yl)-1H-pyrazol-5-yl)phosphonate (4u)



Following the general procedure, treatment of (2-(1H-pyrrol-2-yl)methylene)-1H-indene-1,3(2H)-dione (50 mg, 0.22 mmol) with SGR (50 mg, 0.33 mmol) in the presence of NaOH (22.0 mg, 0.55mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4u** as a yellow solid (73 mg, 89%). **R_f** (Acetone/Dichloromethane: 1/9) = 0.36. **Mp** 160-162 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.2 (C), 147.3 (C), 134.5 (CH), 129.9 (CH), 126.3 (C), 125.9 (CH), 123.8 (CH), 121.8 (C), 120.4 (C), 120.0 (CH), 109.9 (CH), 109.7 (CH), 75.2 (CH), 53.8 (d, $J_{C\text{-}P}$ = 5.4 Hz, CH_3), 53.8 (d, $J_{C\text{-}P}$ = 5.4 Hz, CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 11.52 (s, 1H), 10.25 (s, 1H), 7.93 (d, *J* = 7.6 Hz, 1H), 7.68 (t, *J* = 7.0 Hz, 1H), 7.57 (t, *J* = 7.4 Hz, 1H), 7.42 (d, *J* = 7.6 Hz, 1H), 6.94 (s, 1H), 6.71 (s, 1H), 6.62 (s, 1H), 6.29 (d, *J*

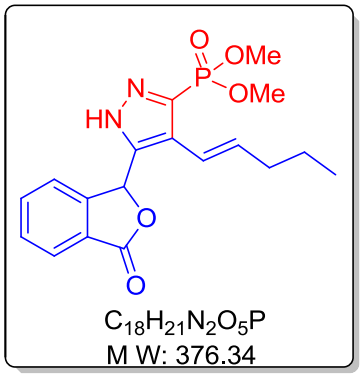
= 4.0 Hz, 1H), 3.76 (d, *J* = 11.2 Hz, 3H), 3.62 (d, *J* = 12.0 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 9.48. **HRMS** for $C_{17}H_{17}N_3O_5P^+$: calcd. $[M+H]^+$: 374.0900, found: 374.0896.

(E)-Dimethyl (4-(ferrocene-2-yl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4v)



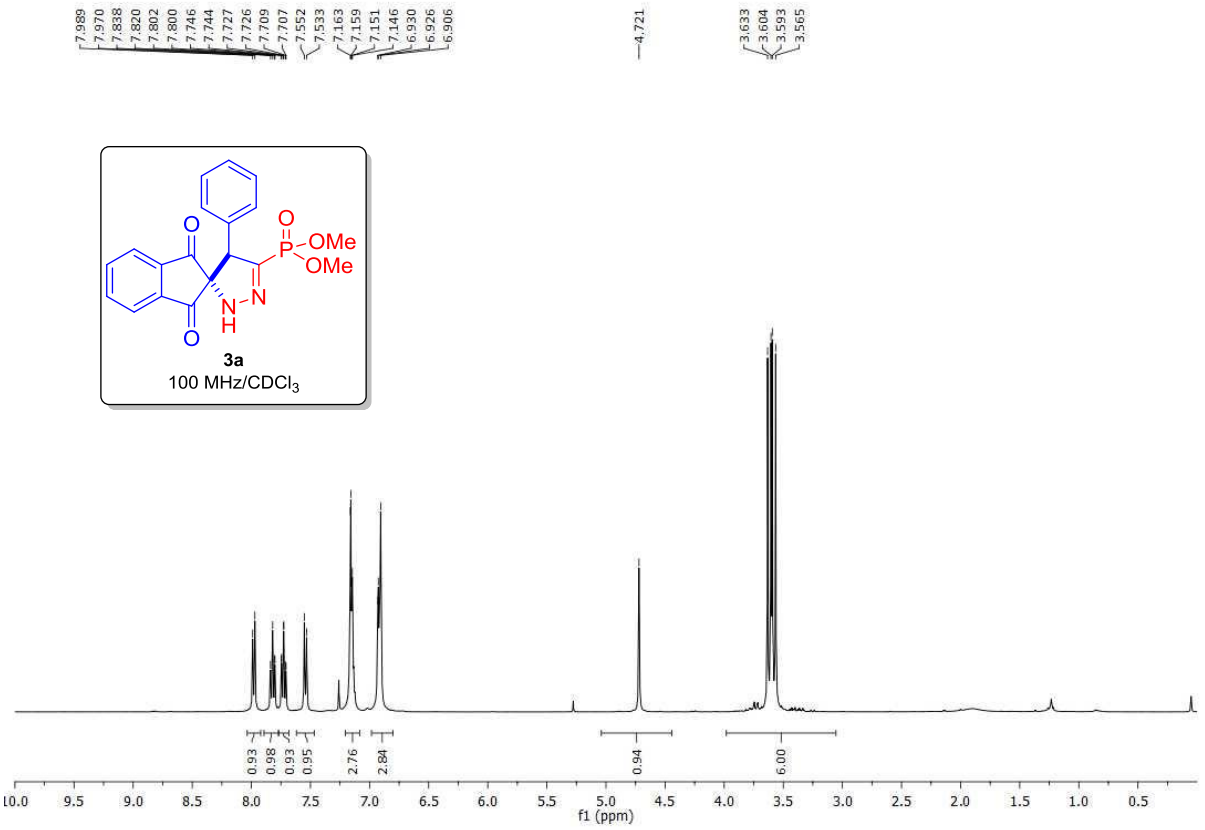
Following the general procedure, treatment of (E)-2-(ferrocenyl)-1H-indene-1,3(2H)-dione (50 mg, 0.15 mmol) with SGR (35 mg, 0.23mmol) in the presence of NaOH (15.0 mg, 0.38 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4v** as a yellow solid (37 mg, 50%). **R_f** (Acetone/Dichloromethane: 2/8) = 0.44. **Mp** 218-220°C. **^{13}C NMR** (100 MHz, ppm/ DMSO-d_6): 170.0 (C), 148.9 (C), 145.7 (C), 134.2 (CH), 129.3 (CH), 127.7 (C), 125.8 (C), 125.6 (C), 125.3 (C), 124.7 (CH), 124.0 (CH), 75.4 (CH), 74.7 (C), 69.4 (CH), 69.4 (CH), 69.4 (CH), 69.4 (CH), 69.4 (CH), 68.7 (CH), 68.4 (CH), 68.3 (C), 53.1 (CH_3), 53.0 (CH_3). **^{1}H NMR** (400 MHz, ppm/ $CDCl_3$): 11.59 (S, 1H), 7.97 (d, *J* = 7.6 Hz, 1H), 7.73 (t, *J* = 7.4 Hz, 1H), 7.61 (t, *J* = 7.4 Hz, 1H), 7.48 (d, *J* = 7.6 Hz, 1H), 7.35 (s, 1H), 4.92 (s, 1H), 4.83 (s, 1H), 4.39 (s, 2H), 4.15 (s, 5H), 3.77 (d, *J* = 11.6 Hz, 3H), 3.64 (d, *J* = 11.6 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.85. **HRMS** for $C_{23}H_{22}FeN_2O_5P^+$: calcd. $[M+Na]^+$: 492.0430, found: 515.0433.

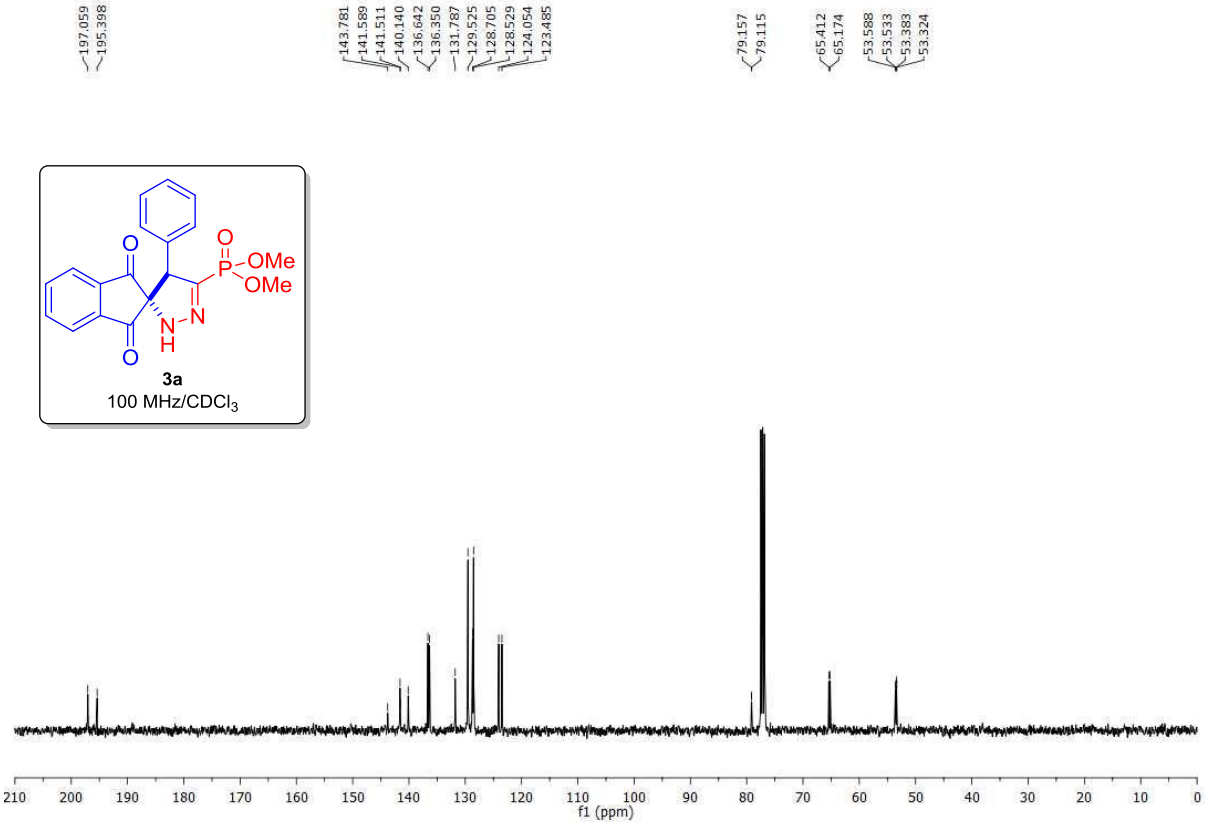
(E)-Dimethyl (4-(but-1-en-1-yl)-3-(3-oxo-1,3-dihydroisobenzofuran-1-yl)-1H-pyrazol-5-yl)phosphonate (4w)

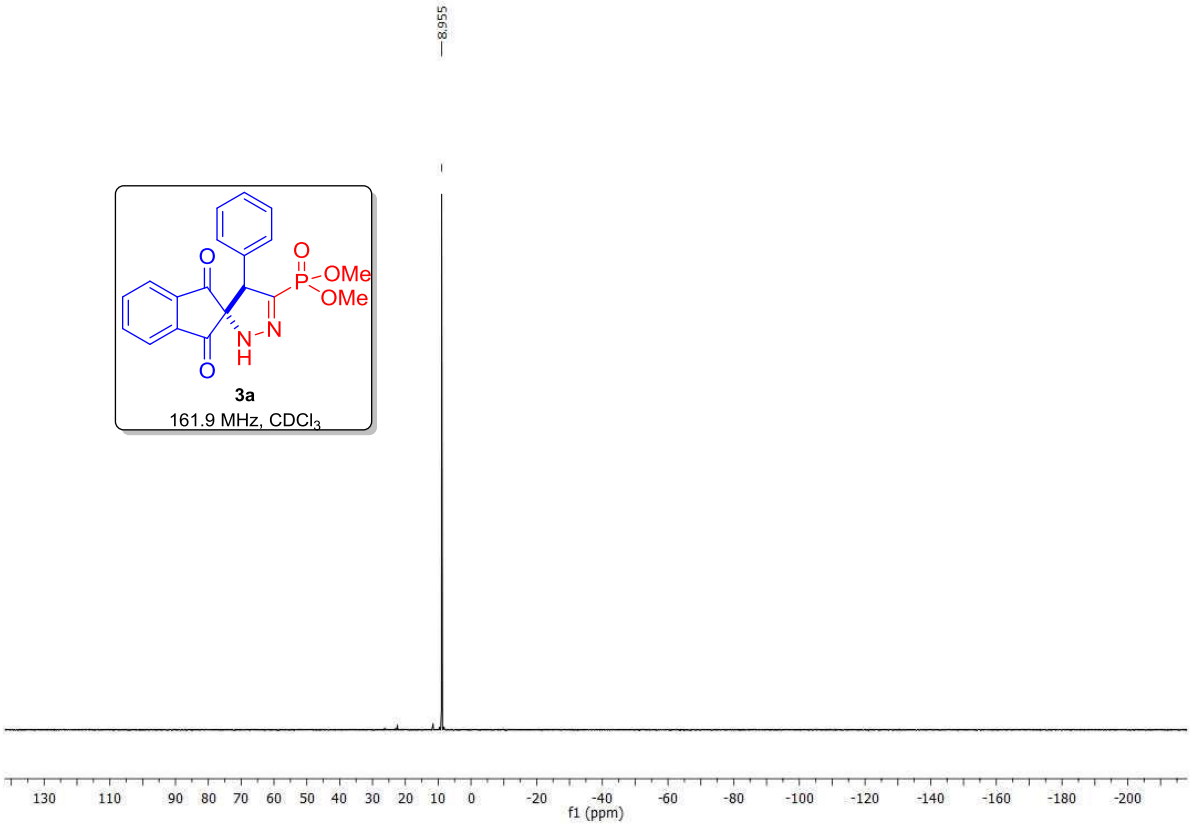


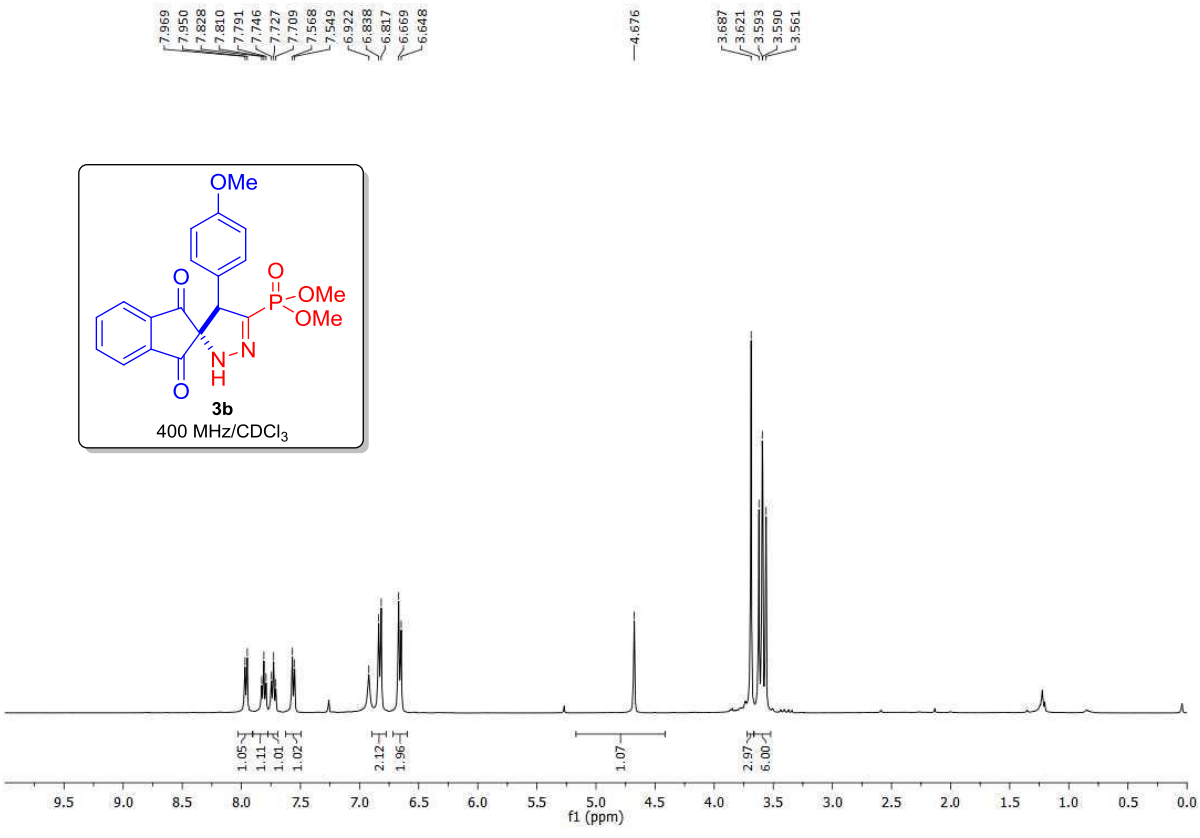
Following the general procedure, treatment of (E)-2-(hex-2-en-1-ylidene)-1H-indene-1,3(2H)-dione (50 mg, 0.22 mmol) with SGR (36 mg, 0.24 mmol) in the presence of NaOH (22.0 mg, 0.55 mmol) in MeOH (3 mL) at 25 °C for 1.5 h followed by column chromatography afforded the product **4w** as a yellow solid (41 mg, 50%). **R_f** (Acetone/Dichloromethane : 1/9) = 0.27. **Mp** 178-180 °C. **^{13}C NMR** (100 MHz, ppm/$CDCl_3$): 170.4 (C), 147.9 (C), 145.0 (C), 137.9 (CH), 134.3 (CH), 129.5 (CH), 127.2 (C), 126.5 (C), 125.8 (C), 125.6 (CH), 123.6 (CH), 117.0 (CH), 76.1 (CH), 53.4 (CH_3), 53.4 (CH_3), 35.6 (CH_2), 22.3 (CH_2), 15.5 (CH_3). **^{1}H NMR** (400 MHz, ppm/$CDCl_3$): 7.91 (d, *J* = 7.6 Hz, 1H), 7.65 (t, *J* = 7.4 Hz, 1H), 7.54 (t, *J* = 7.6 Hz, 1H), 7.39 (d, *J* = 7.6 Hz, 1H), 6.59 (s, 1H), 6.20-6.05 (m, 2H), 3.75 (d, *J* = 11.6 Hz,

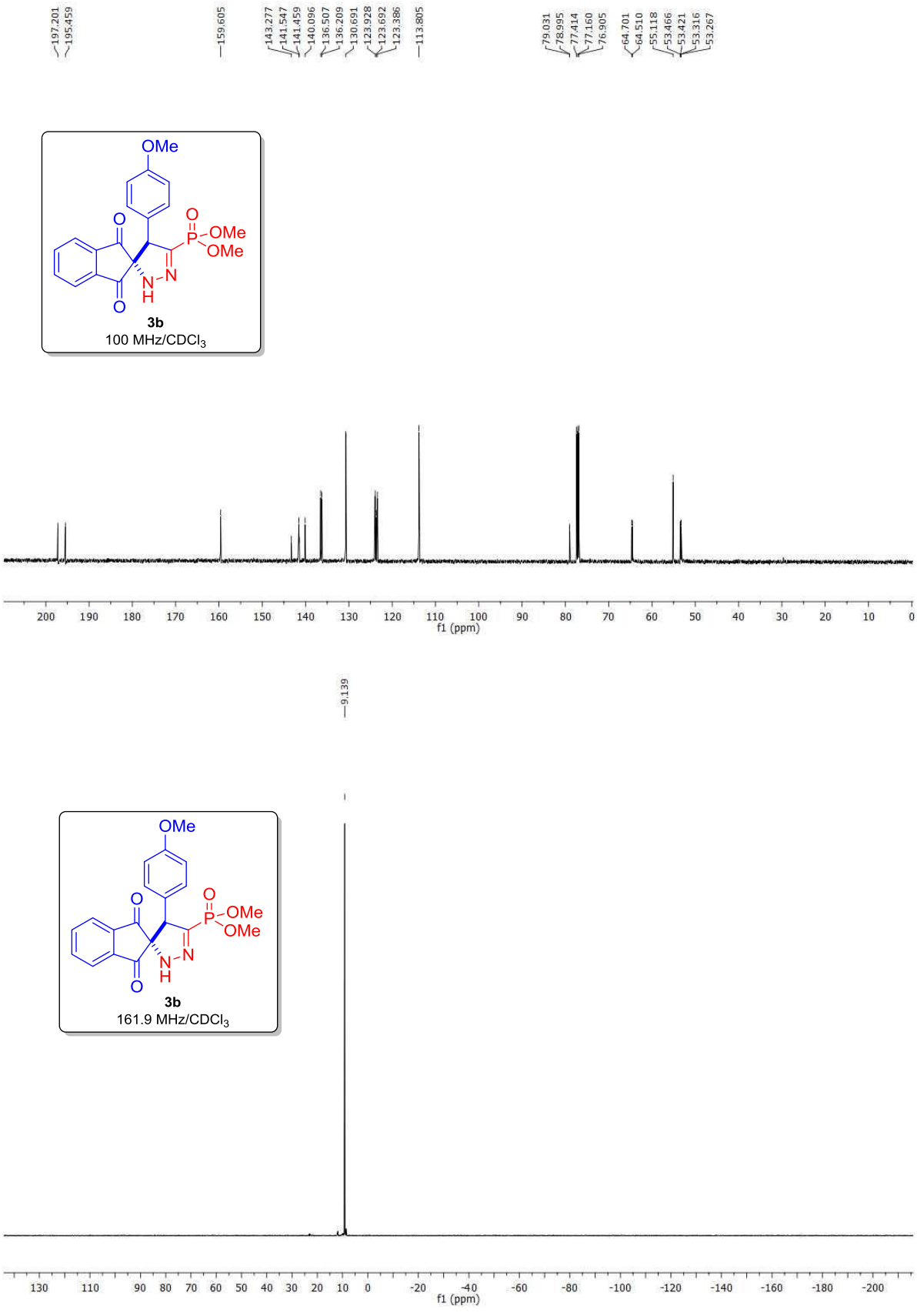
3H), 3.72 (d, *J* = 12.0 Hz, 3H), 2.08 (q, *J* = 7.2 Hz, 2H), 1.39 (q, *J* = 7.2 Hz, 2H), 0.88 (t, *J* = 7.4 Hz, 3H). **^{31}P NMR** (161.9 MHz, $CDCl_3$): 8.85. **HRMS** for $C_{18}H_{22}N_2O_5P^+$: calcd. $[M+H]^+$: 377.1261, found: 377.1256.

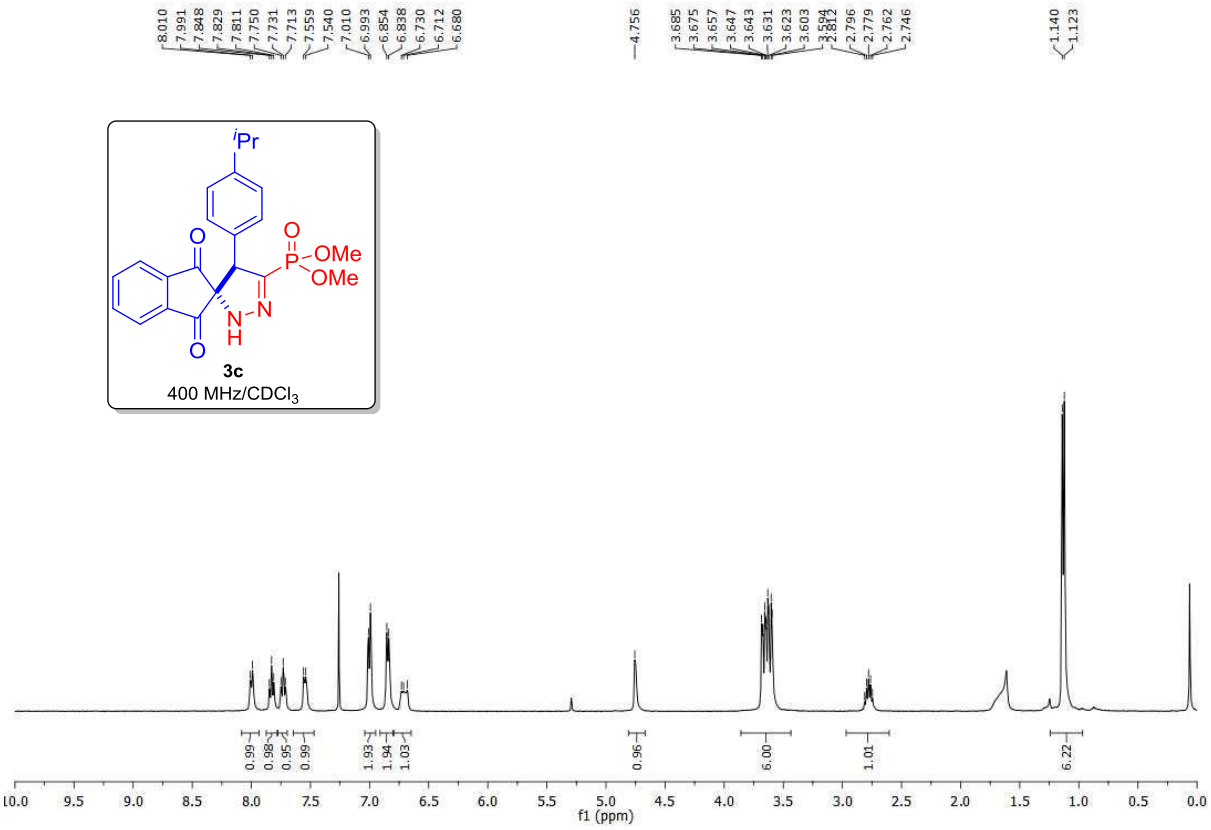

7.989
7.970
7.838
7.820
7.802
7.800
7.746
7.744
7.727
7.726
7.709
7.707
7.552
7.533
7.163
7.159
7.151
7.146
6.930
6.926
6.906
4.721
3.633
3.604
3.593
3.565
O
O
P
OMe
OMe
N
N
H
O
3a
100 MHz/CDCl3
0.93
0.98
0.93
0.95
2.76
2.84
0.94
6.00
f1 (ppm)

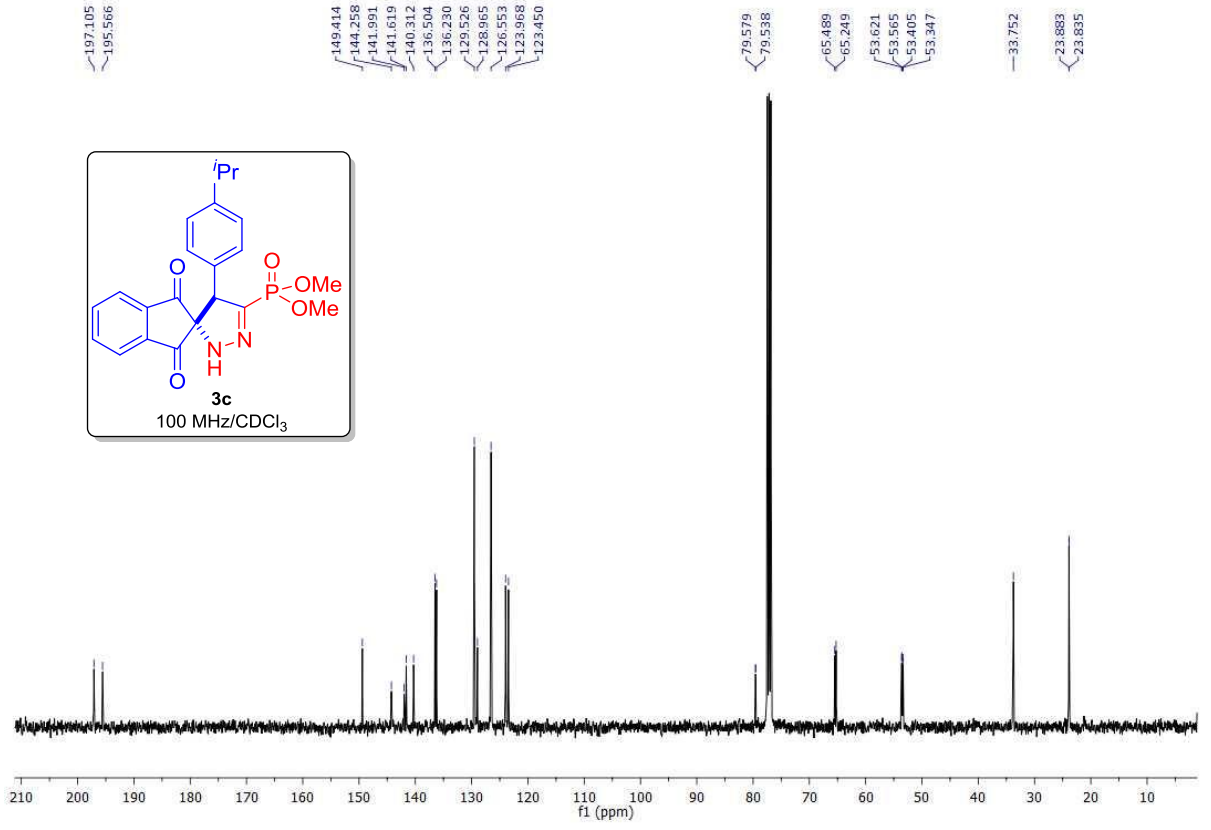

197.059
195.398
143.781
141.589
141.511
140.140
136.642
136.350
131.787
129.525
128.705
128.529
124.054
123.485
79.157
79.115
65.412
65.174
53.588
53.533
53.383
53.324
O
O
P
OMe
OMe
N
N
H
O
3a
100 MHz/CDCl3
f1 (ppm)

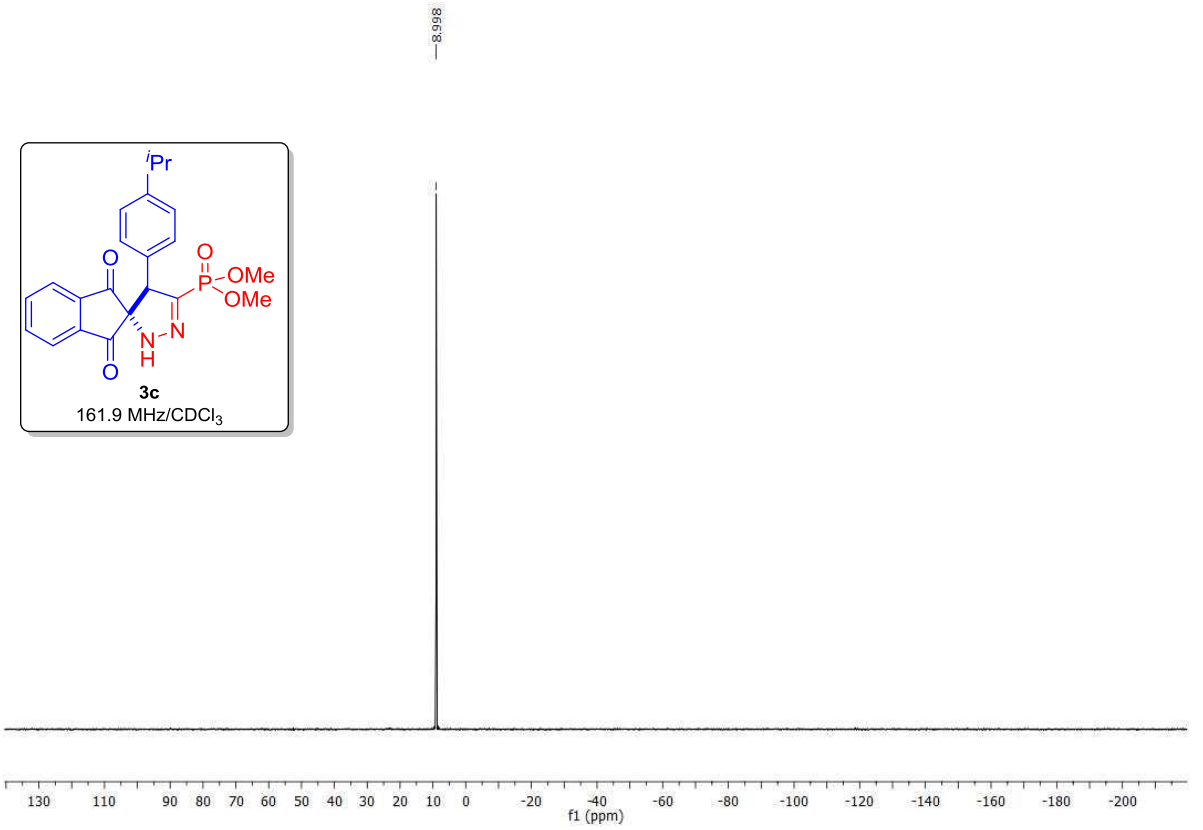

8.955
3a
161.9 MHz, CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)


7.969
7.950
7.828
7.810
7.791
7.746
7.727
7.709
7.568
7.549
6.922
6.838
6.817
6.669
6.648
4.676
3.687
3.621
3.593
3.590
3.561
3b
400 MHz/CDCl3
1.05
1.11
1.01
1.02
2.12
1.96
1.07
2.97
6.00
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

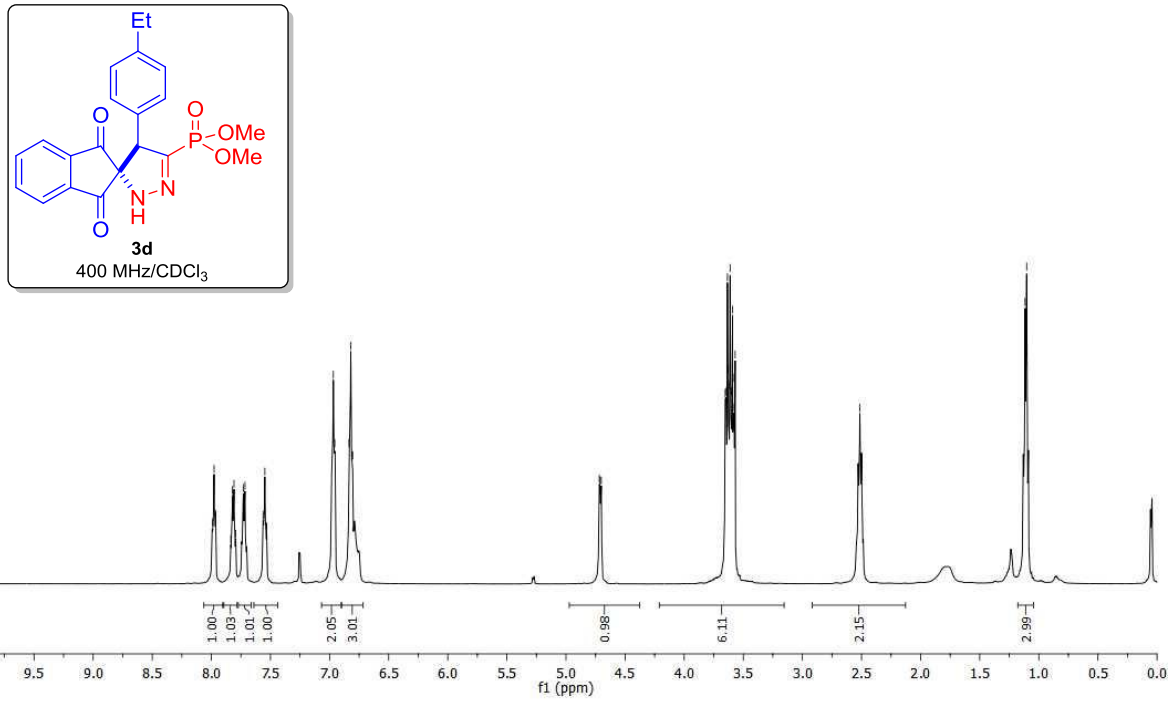

197.201
195.459
159.605
143.277
141.547
141.459
140.096
136.507
136.209
130.691
123.928
123.692
123.386
113.805
79.031
78.995
77.414
77.160
76.905
64.701
64.510
55.118
53.466
53.421
53.316
53.267
OMe
O
P
OMe
OMe
N
N
H
O
O
3b
100 MHz/CDCl3
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)
9.139
OMe
O
P
OMe
OMe
N
N
H
O
O
3b
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

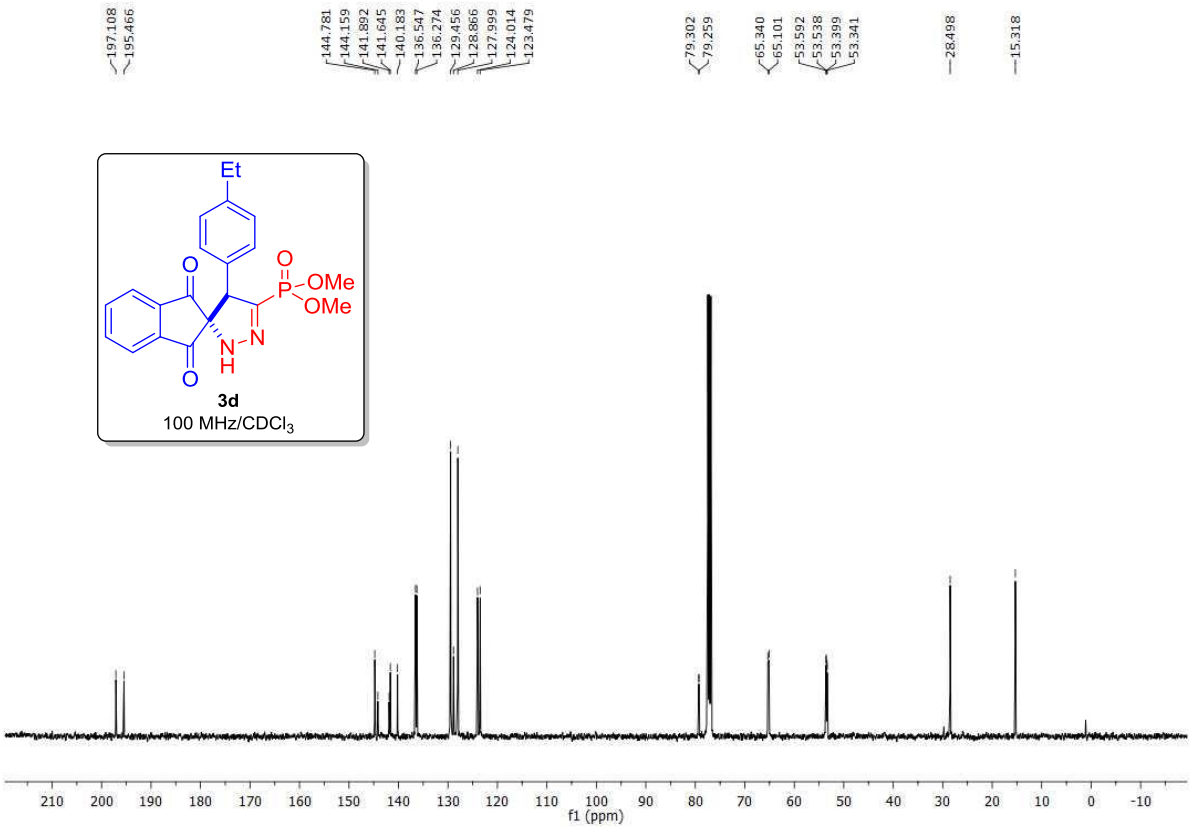

8.010
7.991
7.848
7.829
7.811
7.750
7.731
7.713
7.559
7.540
7.010
6.993
6.854
6.838
6.730
6.712
6.680
4.756
3.685
3.675
3.657
3.647
3.643
3.631
3.623
3.603
2.812
2.796
2.779
2.762
2.746
1.140
1.123
iPr
O
O
P
OMe
OMe
N
N
H
O
3c
400 MHz/CDCl3
0.99
0.98
0.95
0.99
1.93
1.94
1.03
0.96
6.00
1.01
6.22
f1 (ppm)

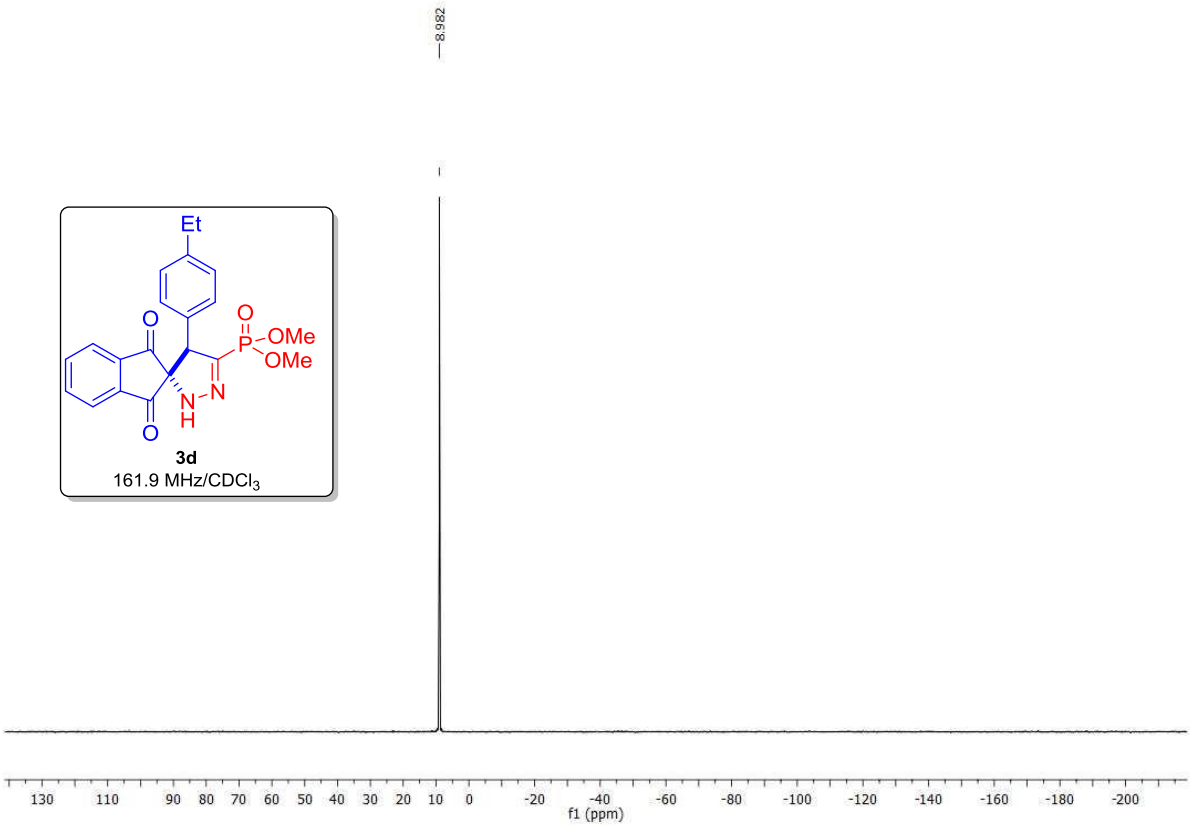

197.105
195.566
149.414
144.258
141.991
141.619
140.312
136.504
136.230
129.526
128.965
126.553
123.968
123.450
79.579
79.538
65.489
65.249
53.621
53.565
53.405
53.347
33.752
23.883
23.835
iPr
O
O
P
OMe
OMe
N
N
H
O
3c
100 MHz/CDCl3
f1 (ppm)

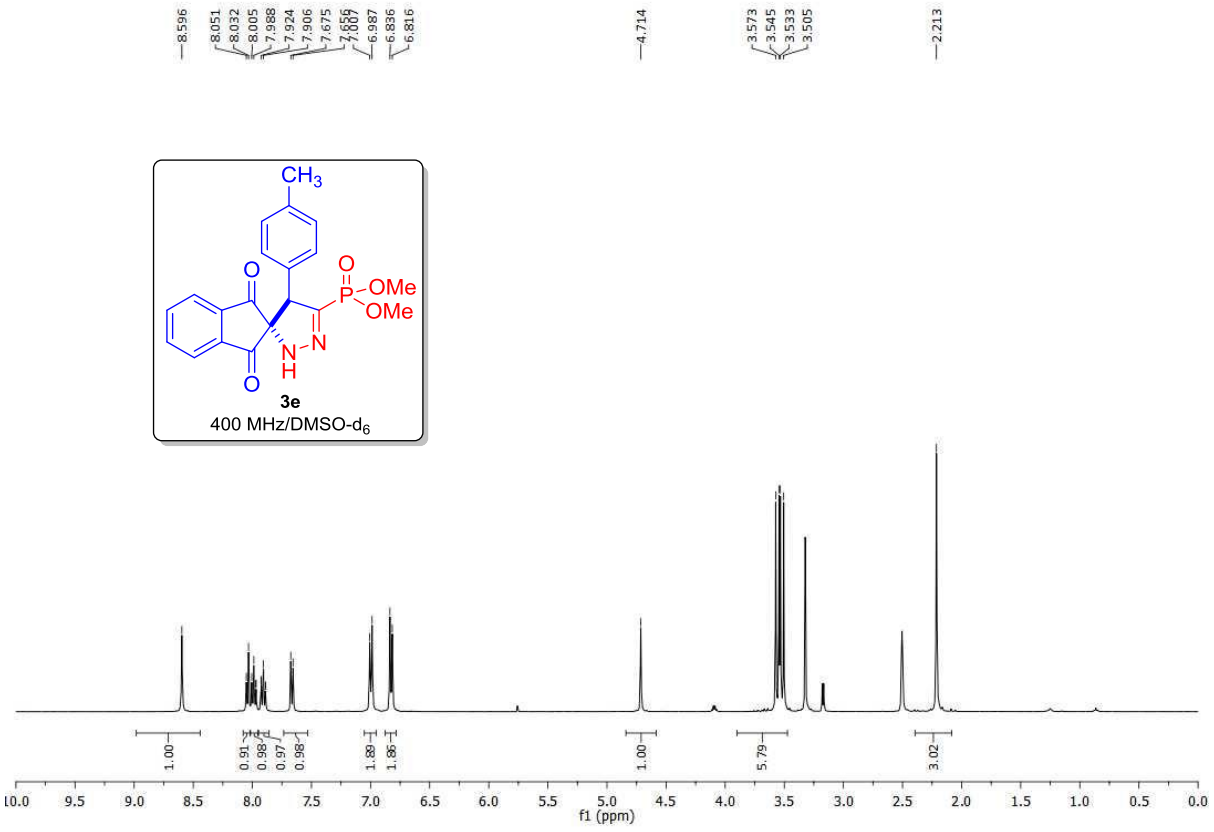

8.998
iPr
O
O
P
OMe
OMe
N
N
H
O
3c
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

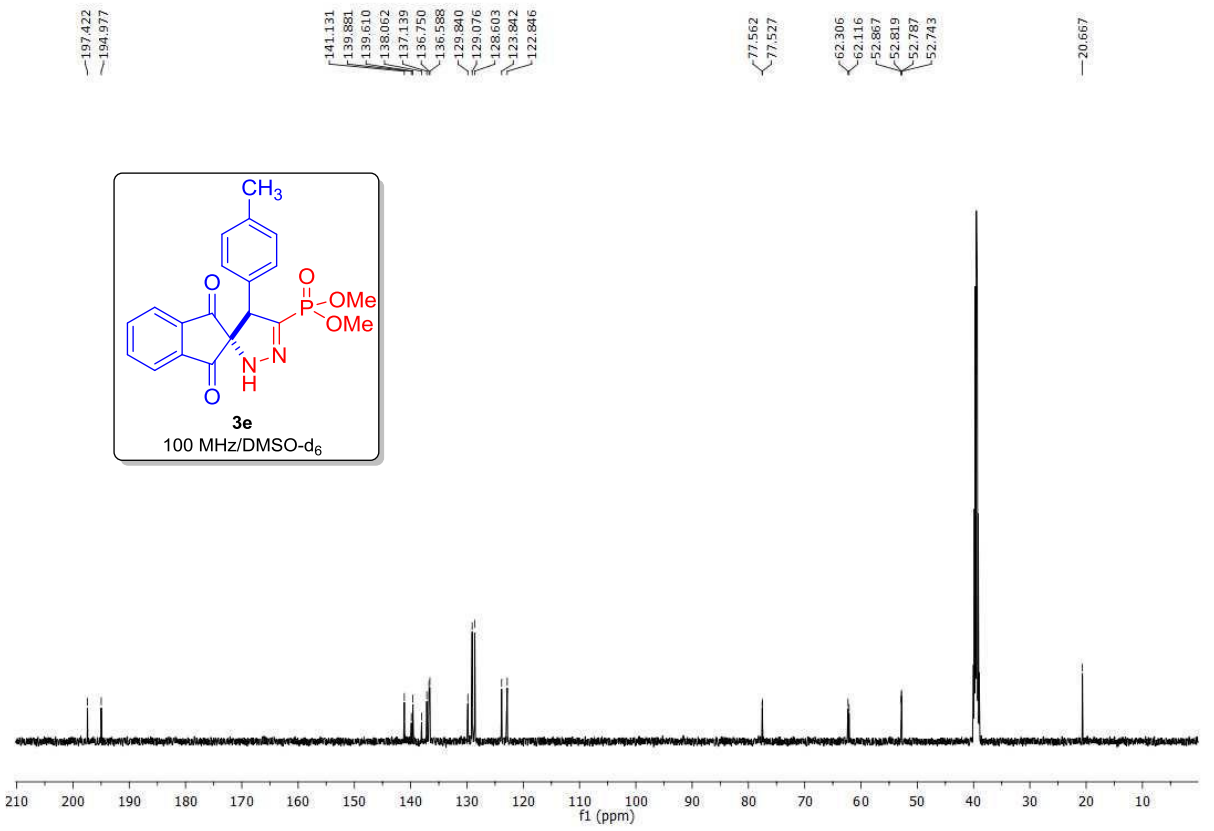

7.992
7.977
7.963
7.962
7.836
7.822
7.809
7.794
7.729
7.716
7.562
7.547
7.532
7.531
6.968
6.956
6.835
6.821
6.809
6.805
4.718
4.711
4.701
3.653
3.645
3.634
3.631
3.622
3.612
3.609
3.600
3.591
3.586
3.577
3.569
2.514
1.116
1.103

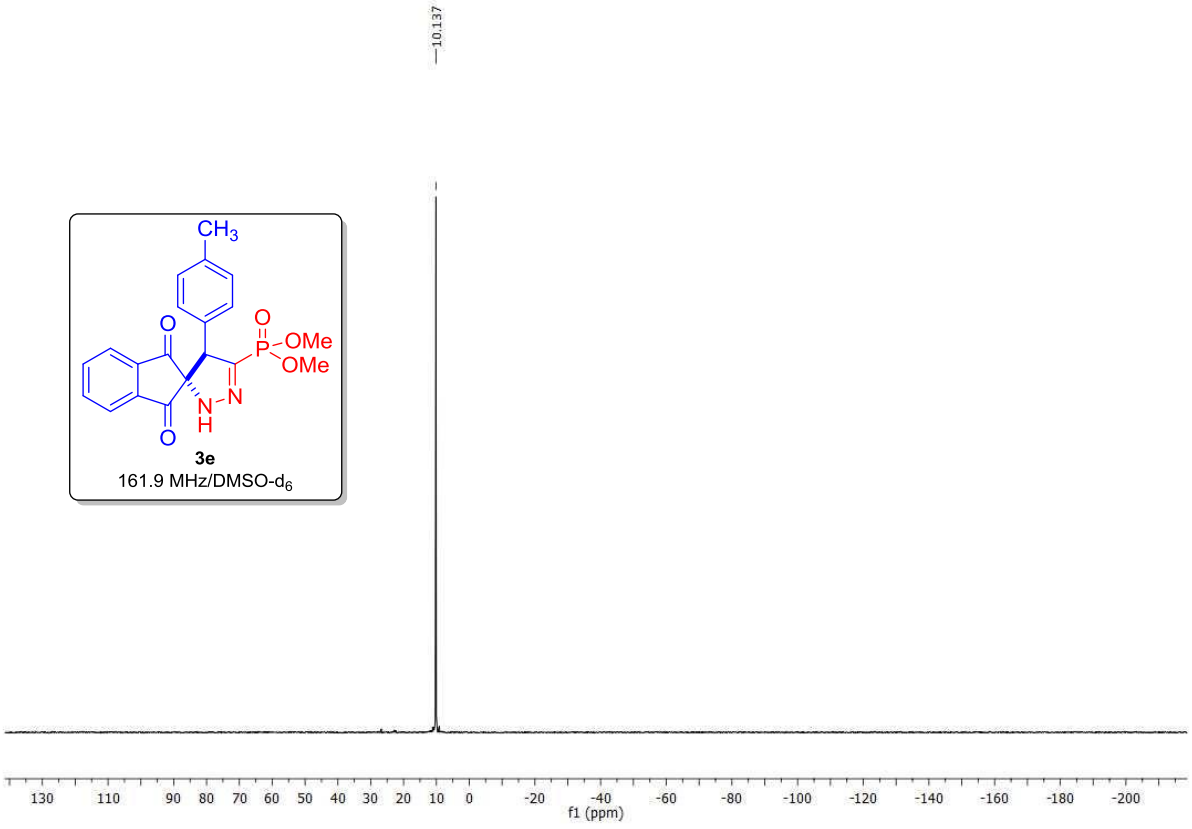

Et
O
O
P
OMe
OMe
N
N
H
O
3d
400 MHz/CDCl3
1.00
1.03
1.01
1.00
2.05
3.01
0.98
6.11
2.15
2.99
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

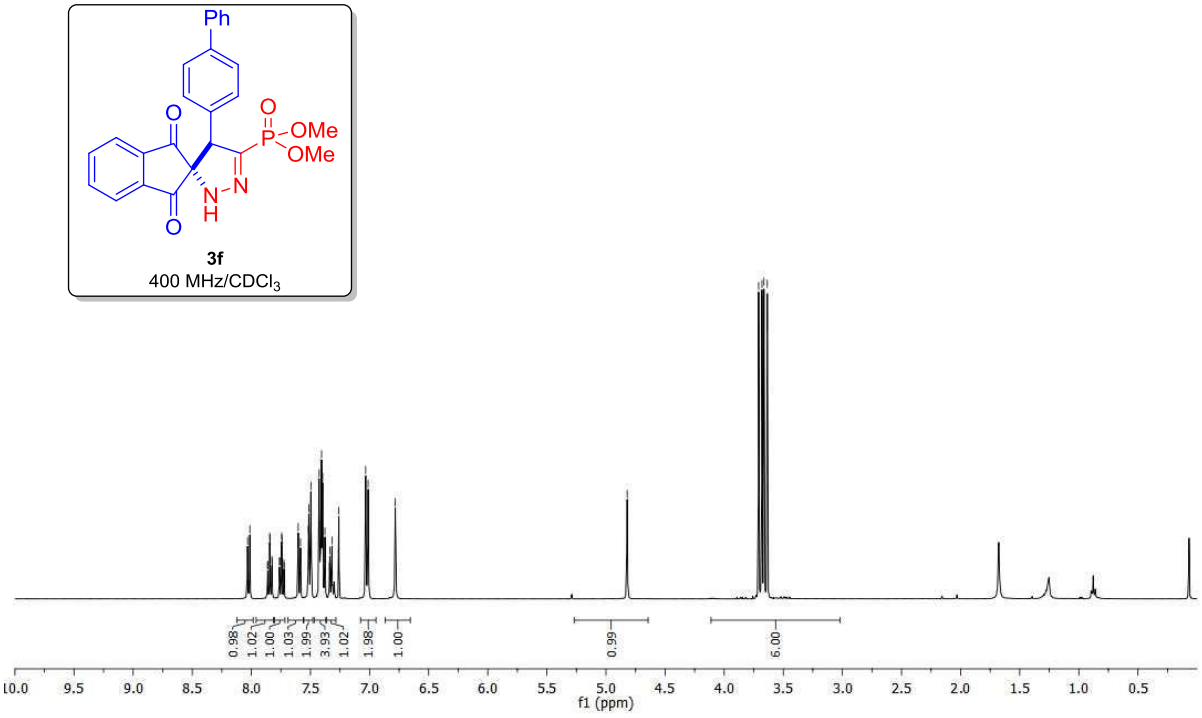

197.108
195.466
144.781
144.159
141.892
141.645
140.183
136.547
136.274
129.456
128.866
127.999
124.014
123.479
79.302
79.259
65.340
65.101
53.592
53.538
53.399
53.341
28.498
15.318
Et
O
O
P
OMe
OMe
N
N
H
O
3d
100 MHz/CDCl3
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

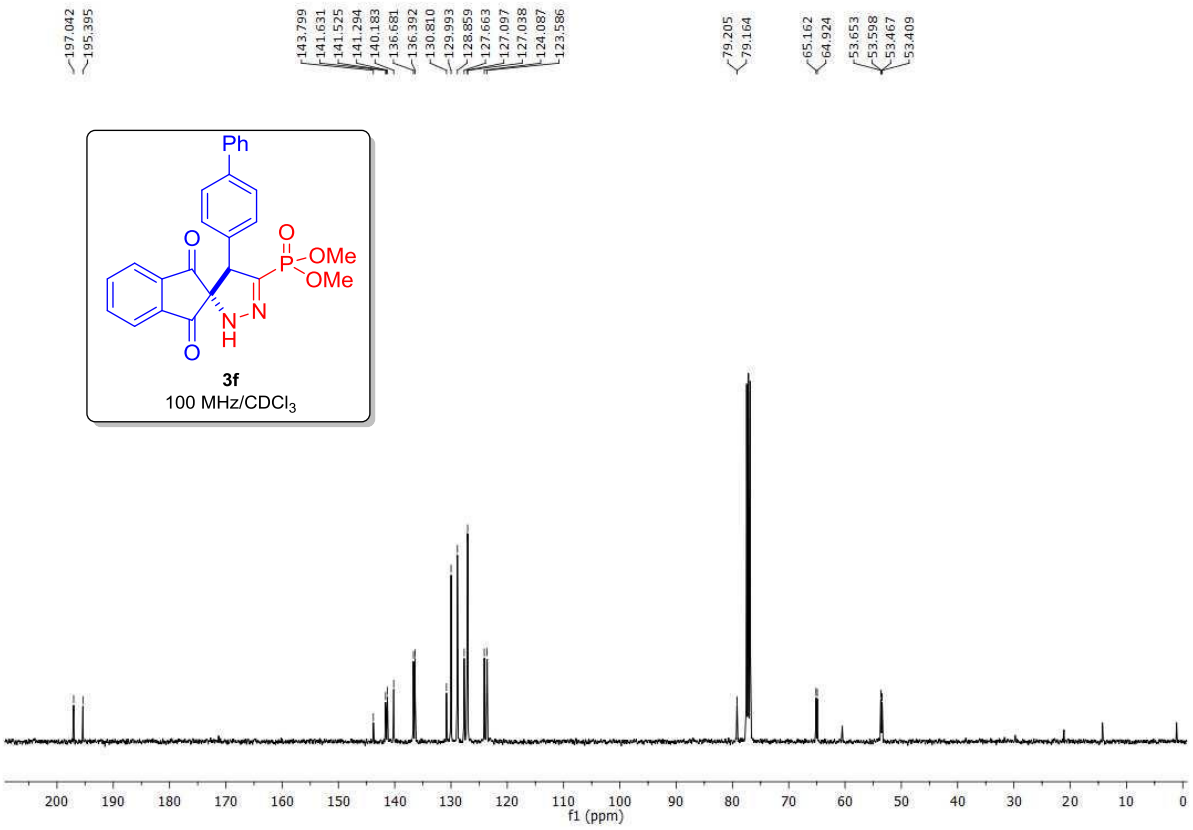

8.982
Et
O
O
P
OMe
OMe
N
N
H
O
3d
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)


8.596
8.051
8.032
8.005
7.988
7.924
7.906
7.675
7.656
7.007
6.987
6.836
6.816
4.714
3.573
3.545
3.533
3.505
2.213
CH3
O
O
P
OMe
OMe
N
N
H
O
3e
400 MHz/DMSO-d6
1.00
0.91
0.98
0.97
0.98
1.89
1.86
1.00
5.79
3.02
f1 (ppm)


197.422
194.977
141.131
139.881
139.610
138.062
137.139
136.750
136.588
129.840
129.076
128.603
123.842
122.846
77.562
77.527
62.306
62.116
52.867
52.819
52.787
52.743
20.667
CH3
O
O
P
OMe
OMe
N
N
H
O
3e
100 MHz/DMSO-d6
f1 (ppm)


10.137
CH3
O
O
P
OMe
OMe
N
N
H
O
3e
161.9 MHz/DMSO-d6
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)
8.032 8.013 7.863 7.861 7.845 7.843 7.826 7.823 7.762 7.760 7.743 7.741 7.725 7.722 7.603 7.584 7.518 7.514 7.510 7.496 7.427 7.415 7.406 7.397 7.378 7.339 7.336 7.318 7.260 7.033 7.012 6.782 4.821
3.709 3.680 3.663 3.635


Ph
O
O
P
OMe
OMe
N
N
H
O
3f
400 MHz/CDCl3
0.98 1.02 1.00 1.03 1.99 3.93 1.02 1.98 1.00 0.99 6.00
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
f1 (ppm)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141.525
141.294
140.183
136.681
136.392
130.810
129.993
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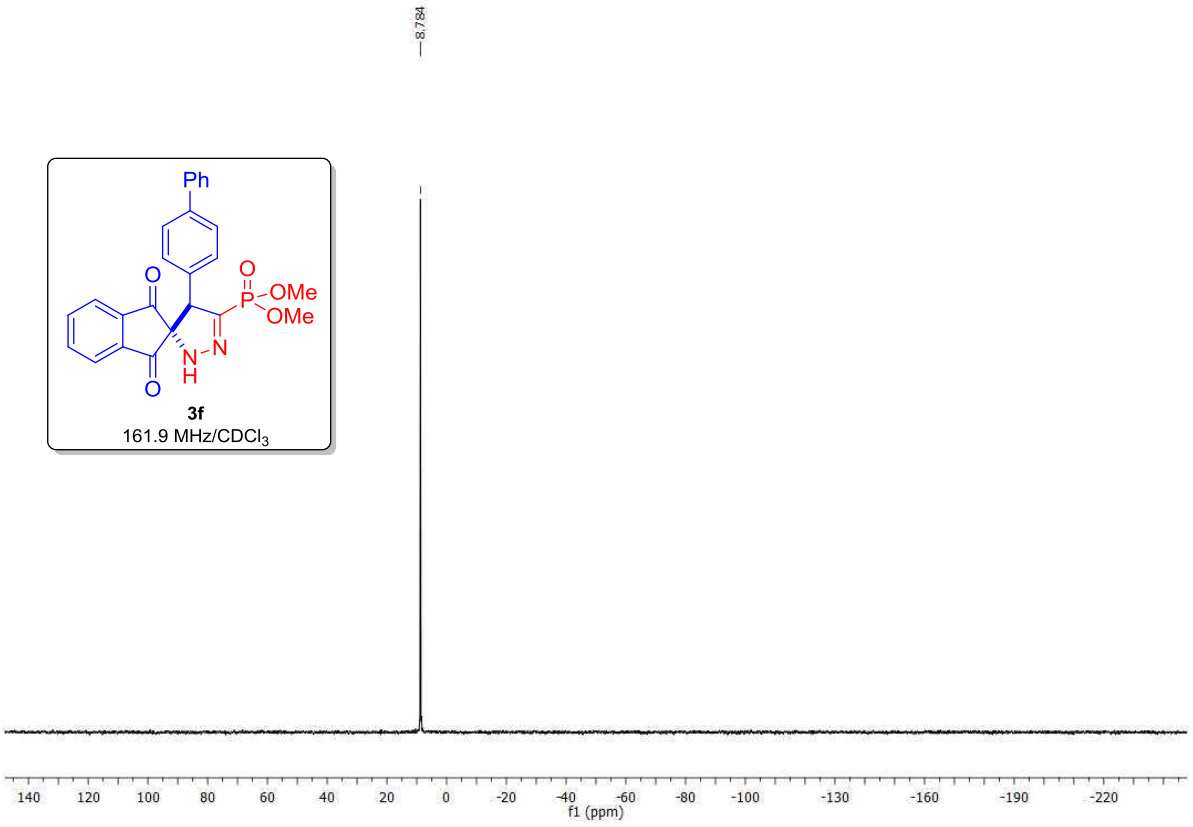
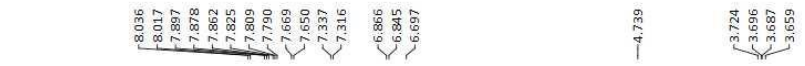
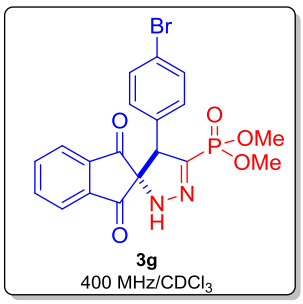
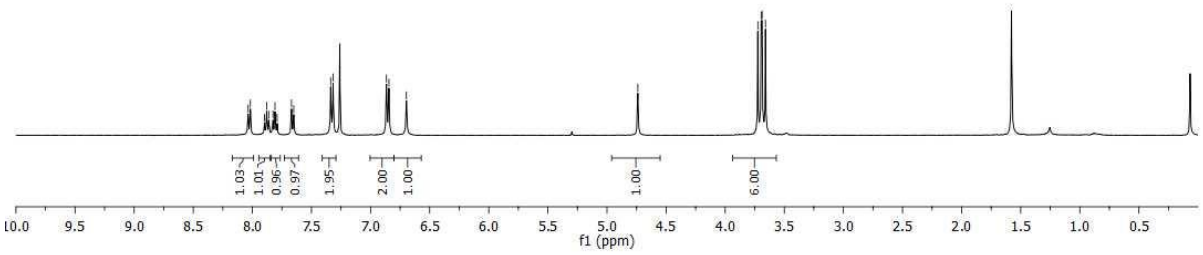
127.663
127.097
127.038
124.087
123.586
79.205
79.164
65.162
64.924
53.653
53.598
53.467
53.409
Ph
O
O
P
OMe
OMe
N
N
H
O
3f
100 MHz/CDCl3
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

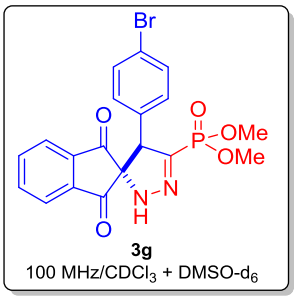

8.784
Ph
O
O
P
OMe
OMe
N
N
H
O
3f
161.9 MHz/CDCl3
140 120 100 80 60 40 20 0 -20 -40 -60 -80 -100 -130 -160 -190 -220
f1 (ppm)



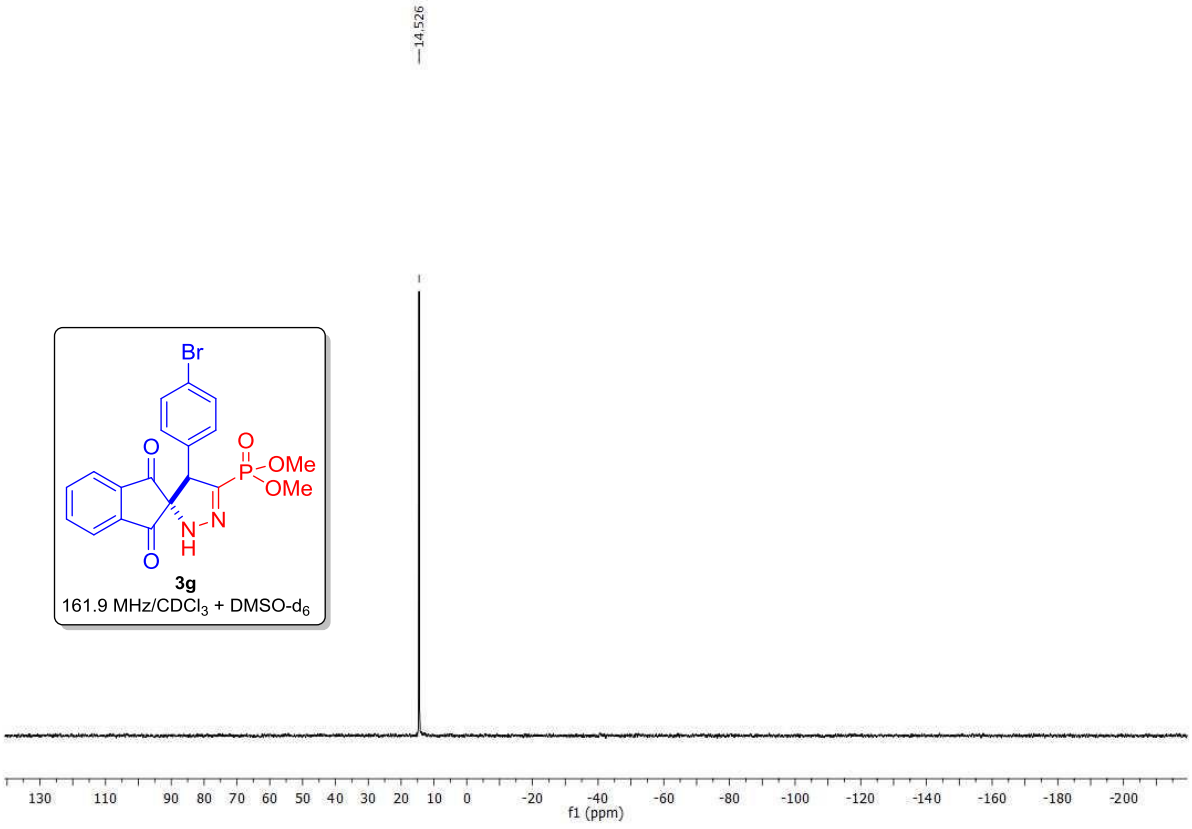


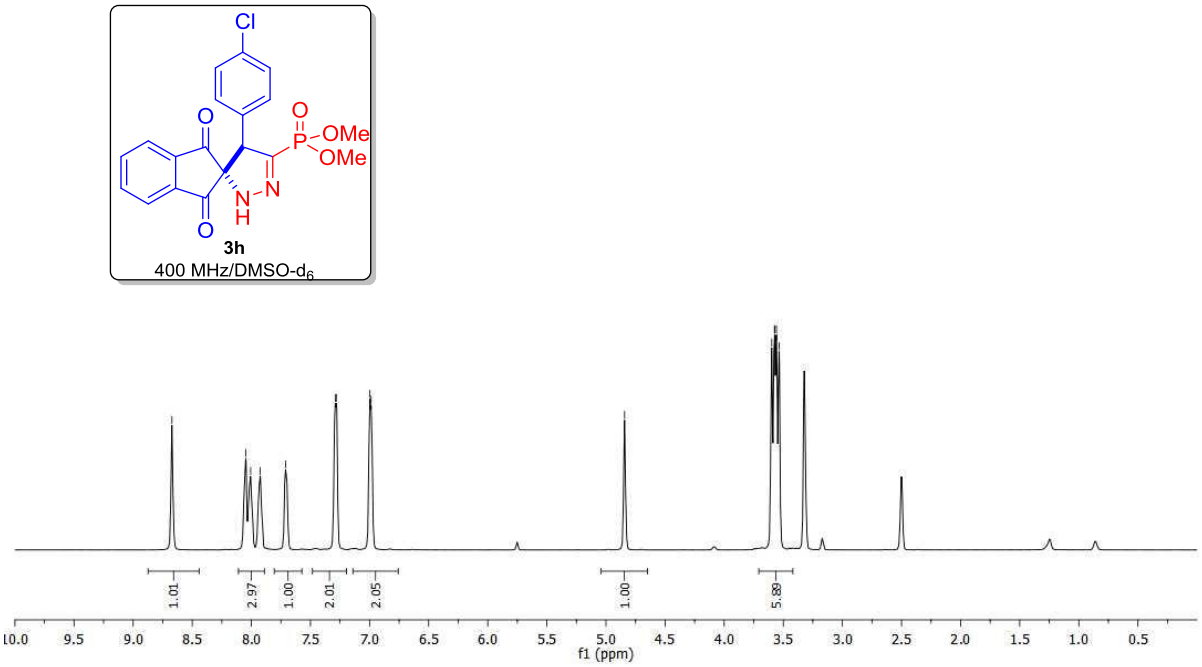
3g
400 MHz/$CDCl_3$

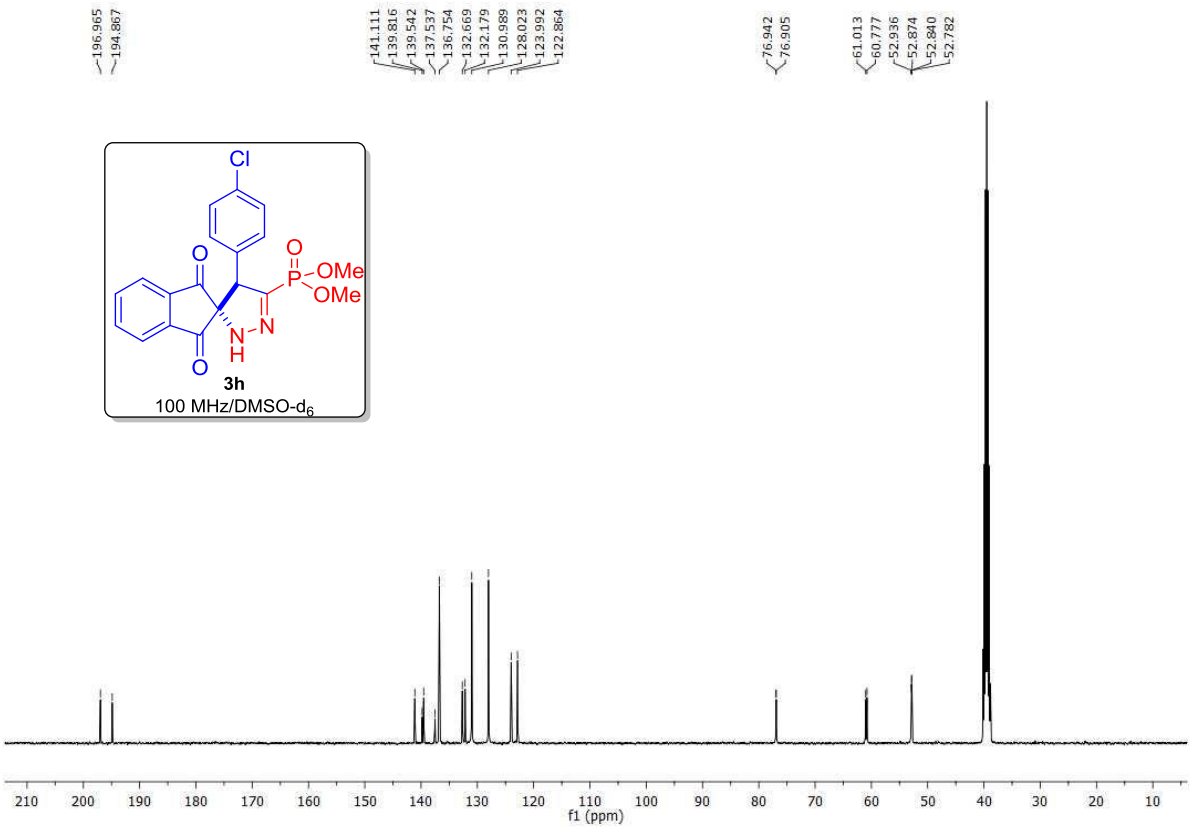


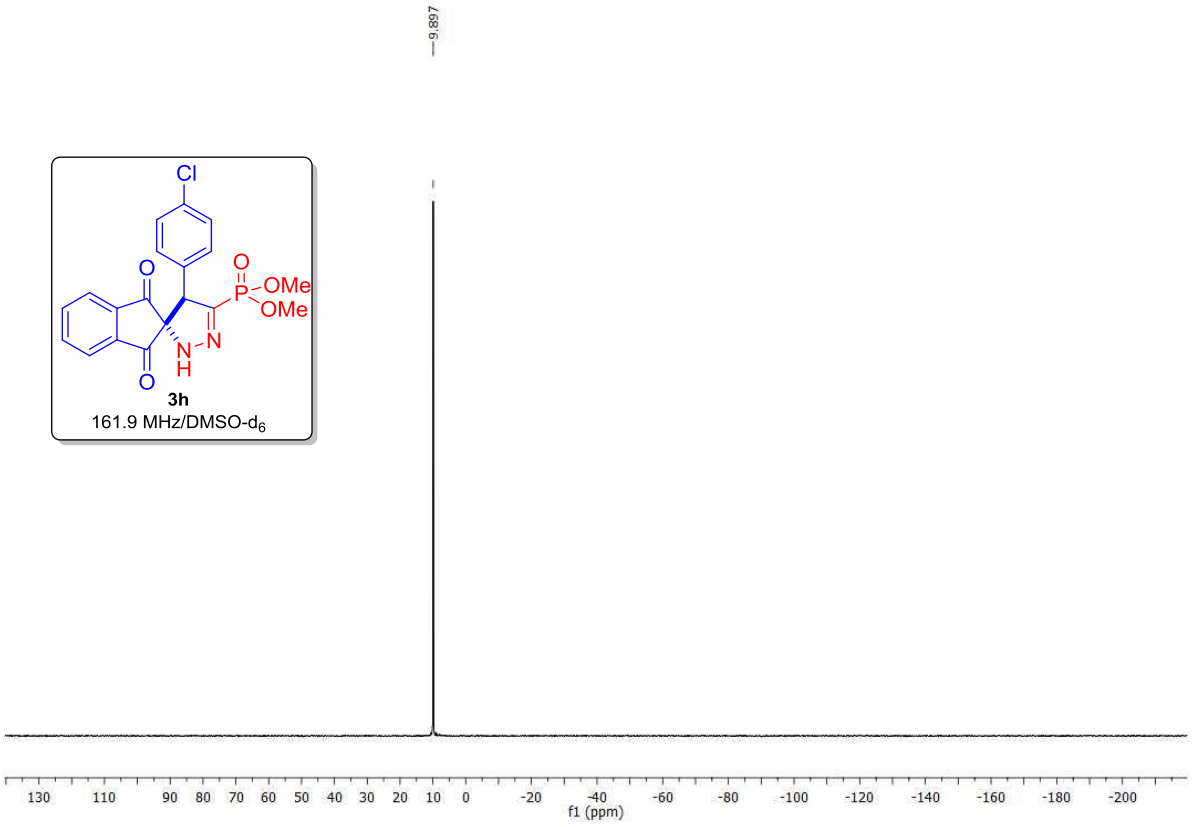


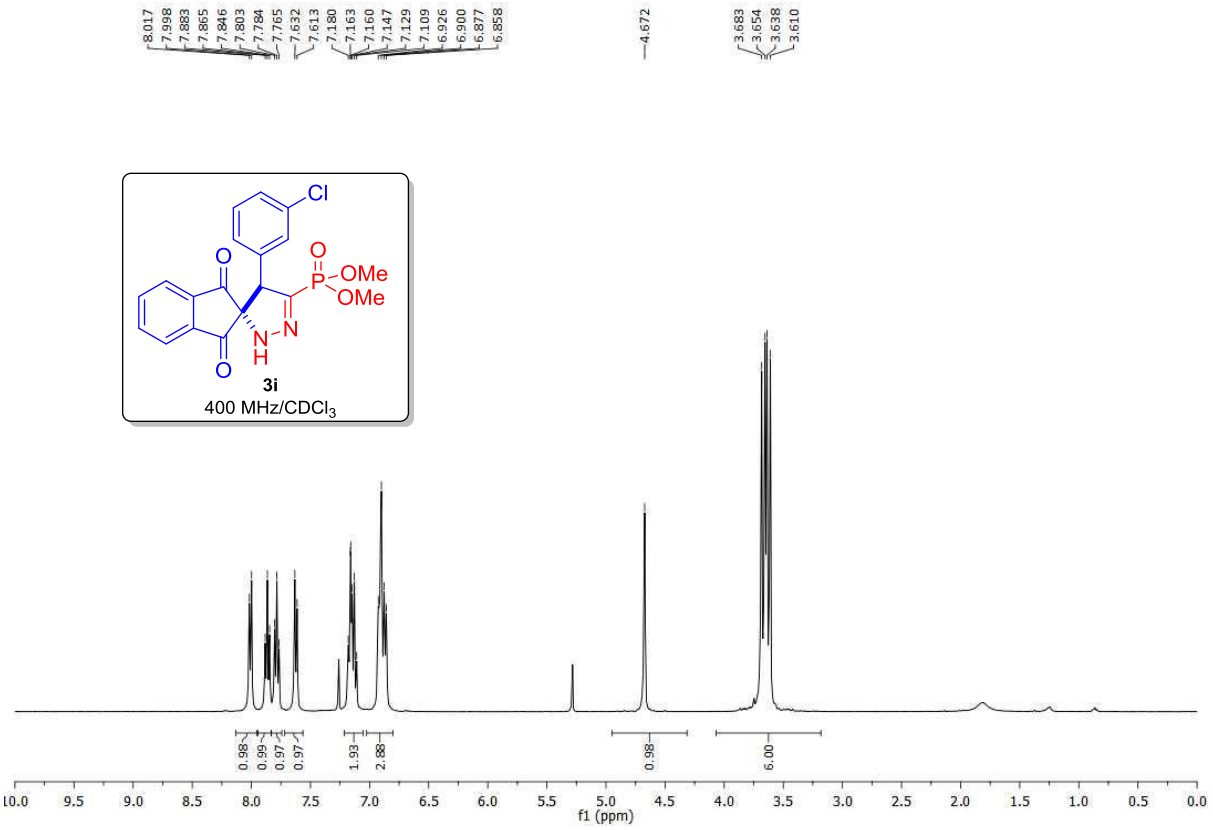
3g
100 MHz/$CDCl_3$ + DMSO-d_6

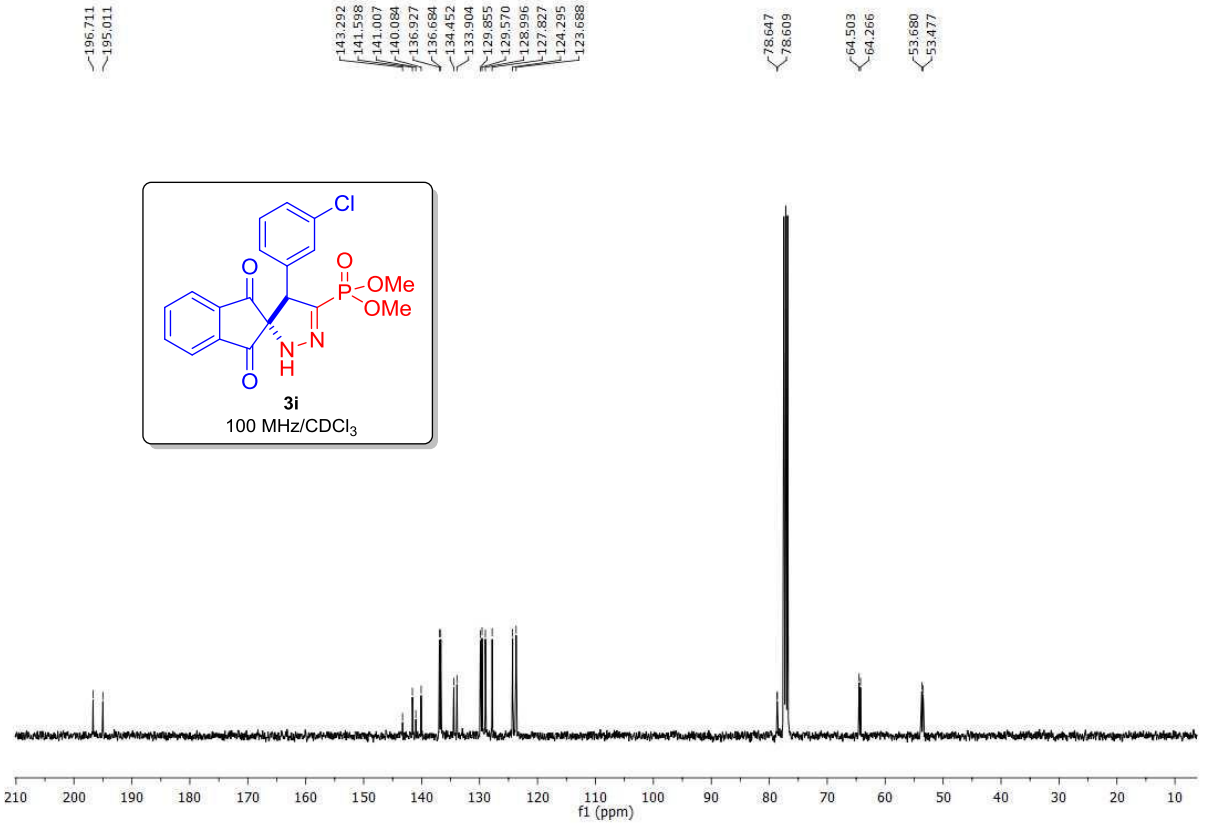

14.526
Br
O
O
P
OMe
OMe
N
N
H
O
3g
161.9 MHz/CDCl3 + DMSO-d6
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)
8.672
8.047
8.006
7.925
7.711
7.290
7.282
6.997
6.987
4.843
3.598
3.575
3.573
3.558
3.536

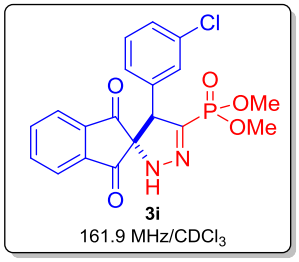

Cl
O
O
P
OMe
OMe
N
N
H
O
3h
400 MHz/DMSO-d6
1.01
2.97
1.00
2.01
2.05
1.00
5.88
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
f1 (ppm)


196.965
194.867
141.111
139.816
139.542
137.537
136.754
132.669
132.179
130.989
128.023
123.992
122.864
76.942
76.905
61.013
60.777
52.936
52.874
52.840
52.782
Cl
O
O
P
OMe
OMe
N
N
H
O
3h
100 MHz/DMSO-d6
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


9.897
Cl
O
O
P
OMe
OMe
N
N
H
O
3h
161.9 MHz/DMSO-d6
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

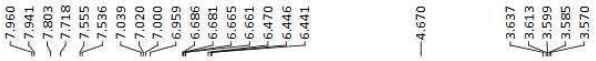

8.017
7.998
7.883
7.865
7.846
7.803
7.784
7.765
7.632
7.613
7.180
7.163
7.160
7.147
7.129
7.109
6.926
6.900
6.877
6.858
4.672
3.683
3.654
3.638
3.610
Cl
O
O
P
OMe
OMe
N
N
H
O
3i
400 MHz/CDCl3
0.98
0.99
0.97
0.97
1.93
2.88
0.98
6.00
f1 (ppm)

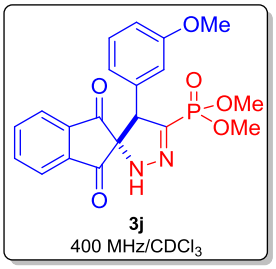

196.711
195.011
143.292
141.598
141.007
140.084
136.927
136.684
134.452
133.904
129.855
129.570
128.996
127.827
124.295
123.688
78.647
78.609
64.503
64.266
53.680
53.477
Cl
O
O
P
OMe
OMe
N
N
H
O
3i
100 MHz/CDCl3
f1 (ppm)



3i

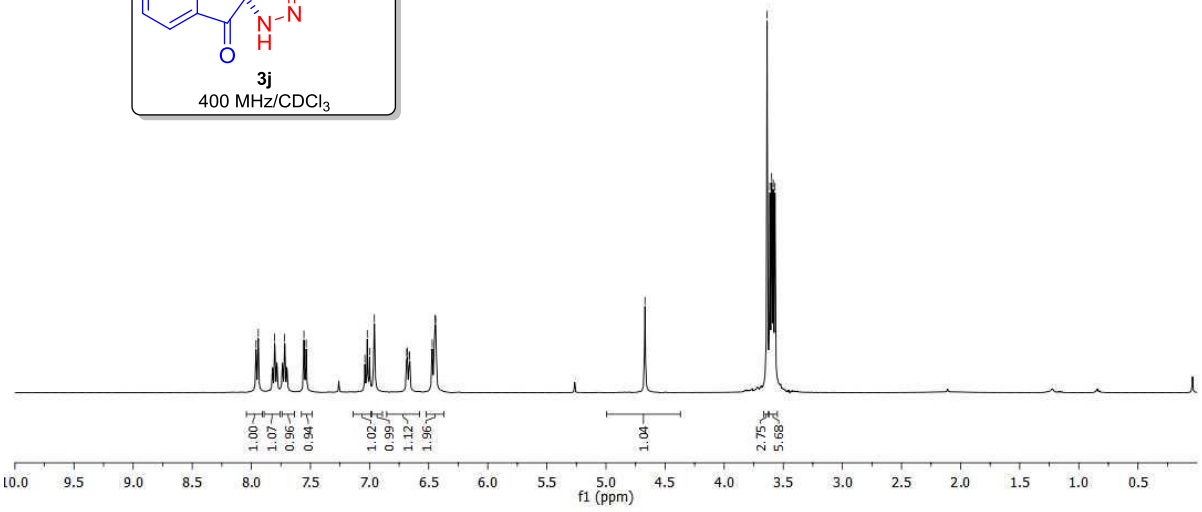
161.9 MHz/$CDCl_3$

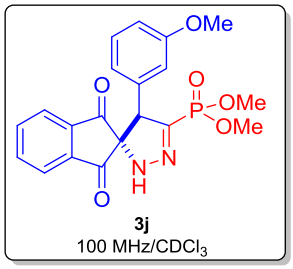




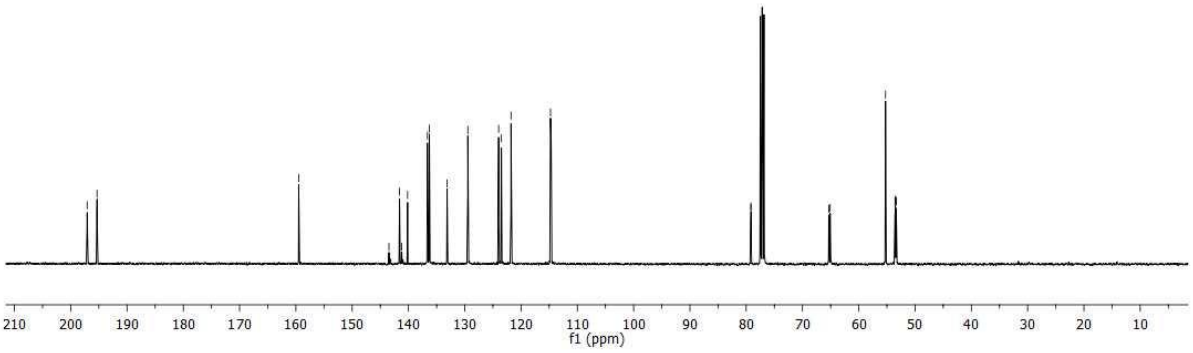
3j

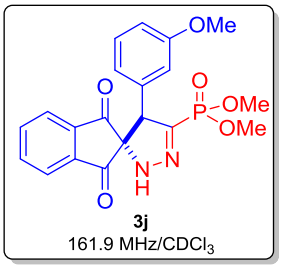
400 MHz/$CDCl_3$



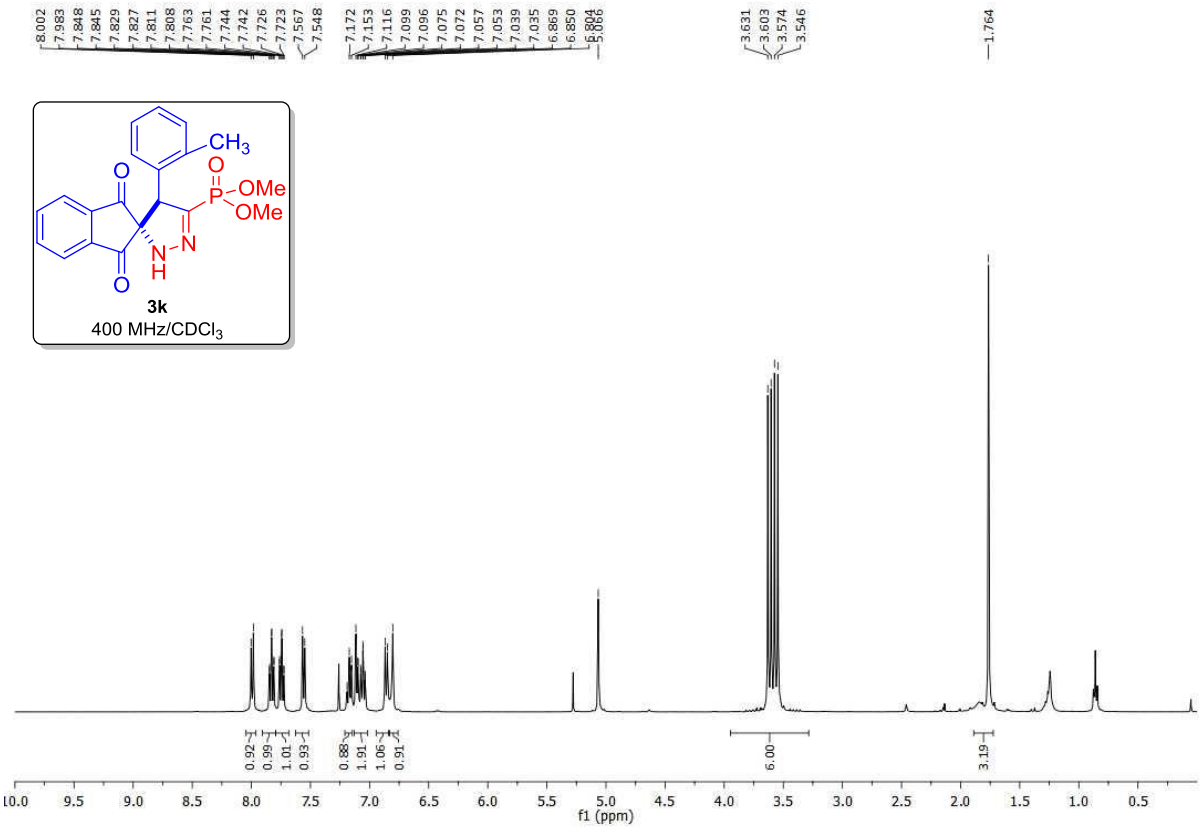


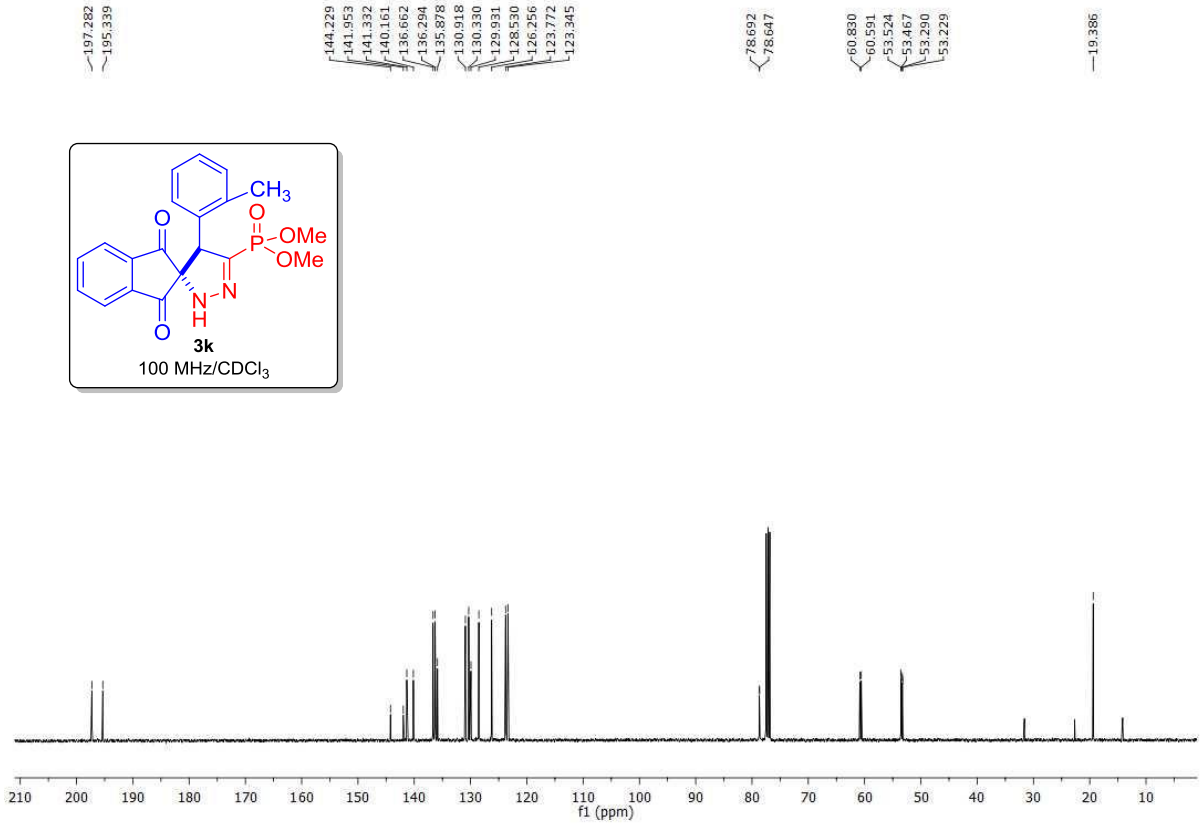
3j
100 MHz/$CDCl_3$

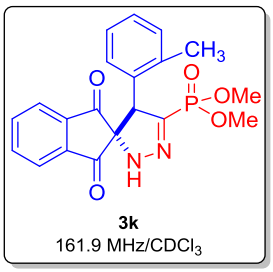




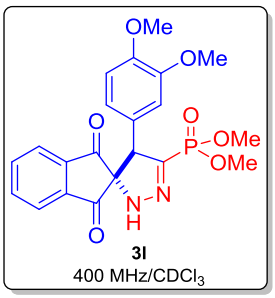
3j
161.9 MHz/$CDCl_3$


8.002
7.983
7.848
7.845
7.829
7.827
7.811
7.808
7.763
7.761
7.744
7.742
7.726
7.723
7.567
7.548
7.172
7.153
7.116
7.099
7.096
7.075
7.072
7.057
7.053
7.039
7.035
6.869
6.850
6.804
5.066
3.631
3.603
3.574
3.546
1.764
CH3
O
P
OMe
OMe
N
H
O
O
3k
400 MHz/CDCl3
0.92
0.99
1.01
0.93
0.88
1.91
1.06
0.91
6.00
3.19
f1 (ppm)

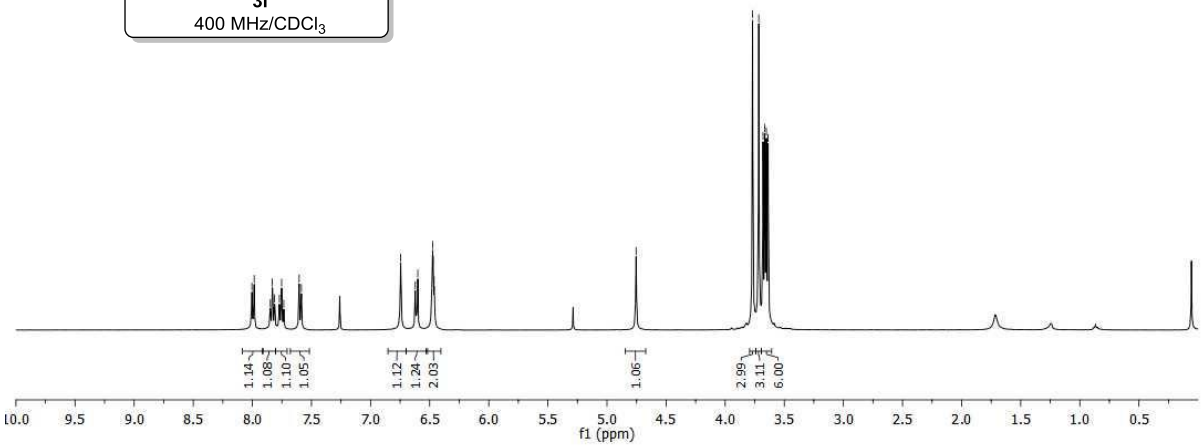

197.282
195.339
144.229
141.953
141.332
140.161
136.662
136.294
135.878
130.918
130.330
129.931
128.530
126.256
123.772
123.345
78.692
78.647
60.830
60.591
53.524
53.467
53.290
53.229
19.386
CH3
O
P
OMe
OMe
N
H
O
O
3k
100 MHz/CDCl3
f1 (ppm)

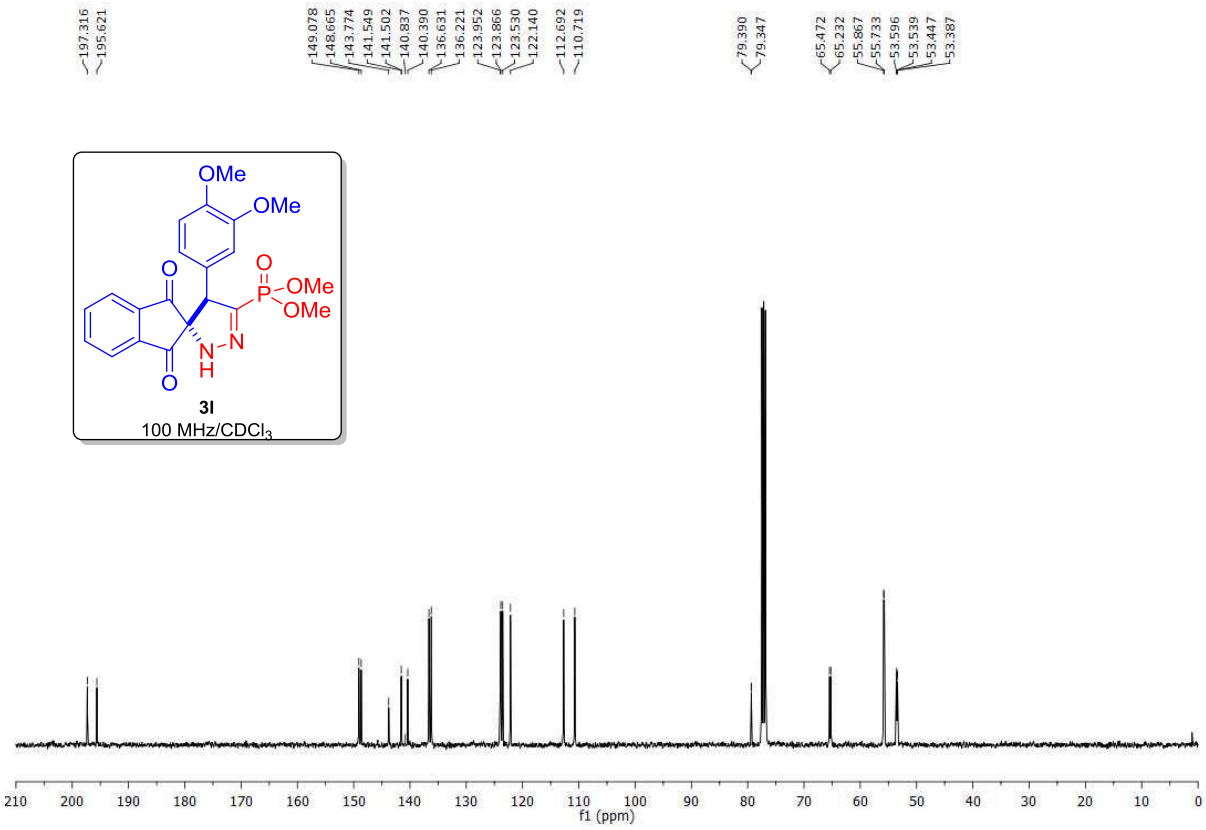


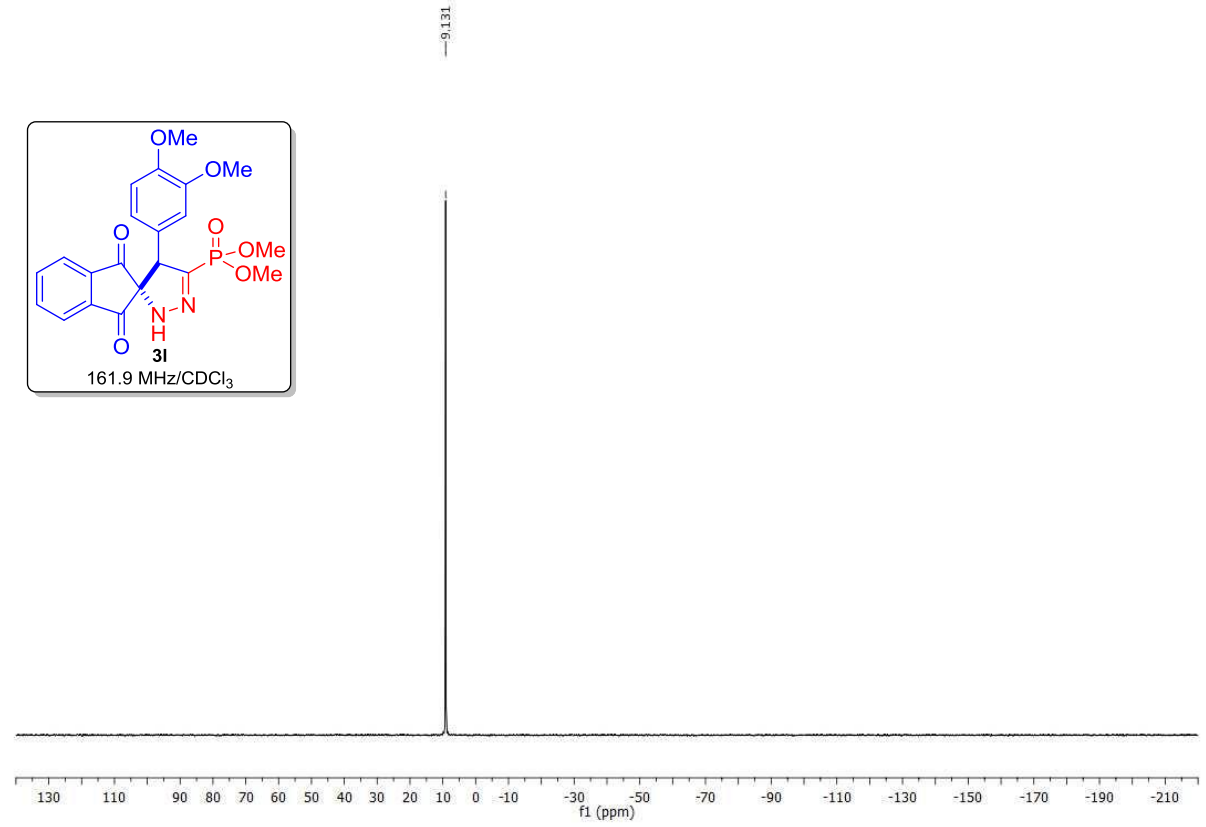
3k
161.9 MHz/$CDCl_3$

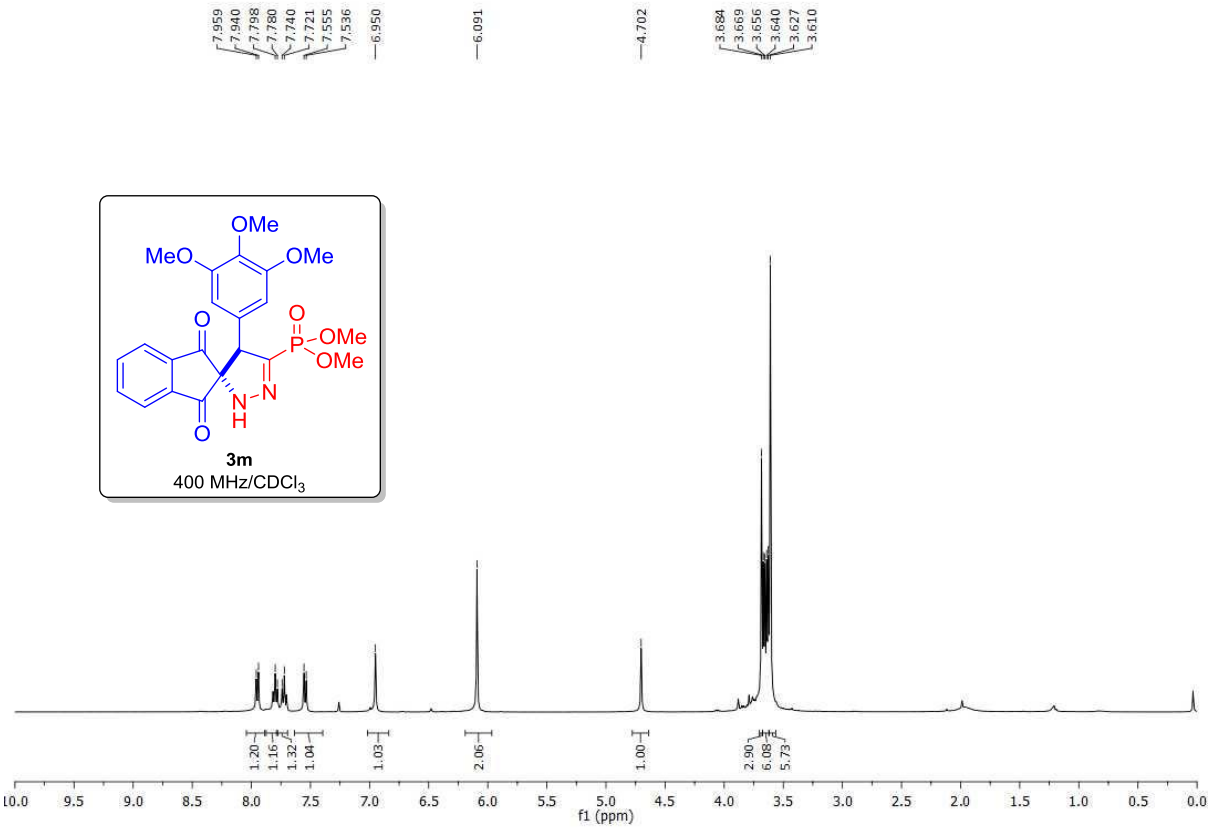


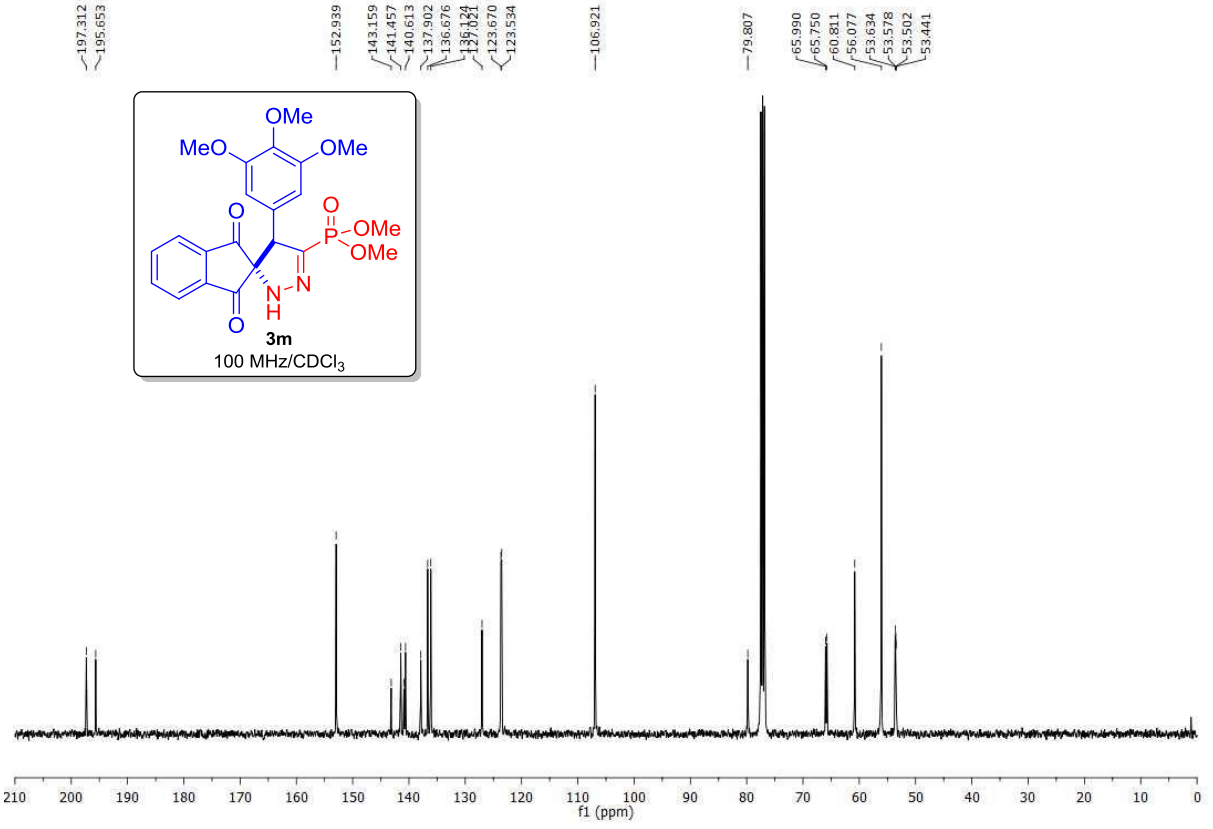
3l
400 MHz/$CDCl_3$

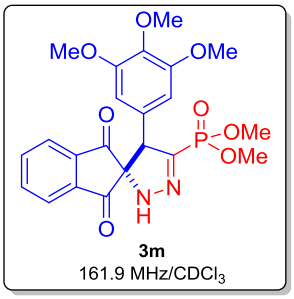



197.316
195.621
149.078
148.665
143.774
141.549
141.502
140.837
140.390
136.631
136.221
123.952
123.866
123.530
122.140
112.692
110.719
79.390
79.347
65.472
65.232
55.867
55.733
53.596
53.539
53.447
53.387
OMe
OMe
O
P
OMe
OMe
N
N
H
O
O
3l
100 MHz/CDCl3
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


9.131
OMe
OMe
O
P
OMe
OMe
N
N
H
O
O
3l
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -10 -30 -50 -70 -90 -110 -130 -150 -170 -190 -210
f1 (ppm)

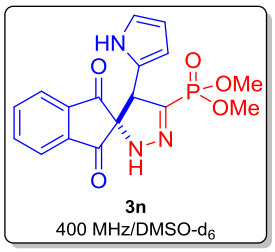

7.959
7.940
7.798
7.780
7.740
7.721
7.555
7.536
6.950
6.091
4.702
3.684
3.669
3.656
3.640
3.627
3.610
OMe
MeO
OMe
O
P
OMe
OMe
N
N
H
O
O
3m
400 MHz/CDCl3
1.20
1.16
1.32
1.04
1.03
2.06
1.00
2.90
6.08
5.73
f1 (ppm)


197.312
195.653
152.939
143.159
141.457
140.613
137.902
136.676
136.124
127.021
123.670
123.534
106.921
79.807
65.990
65.750
60.811
56.077
53.634
53.578
53.502
53.441
OMe
MeO
OMe
O
P
OMe
OMe
N
N
H
O
O
3m
100 MHz/CDCl3
f1 (ppm)



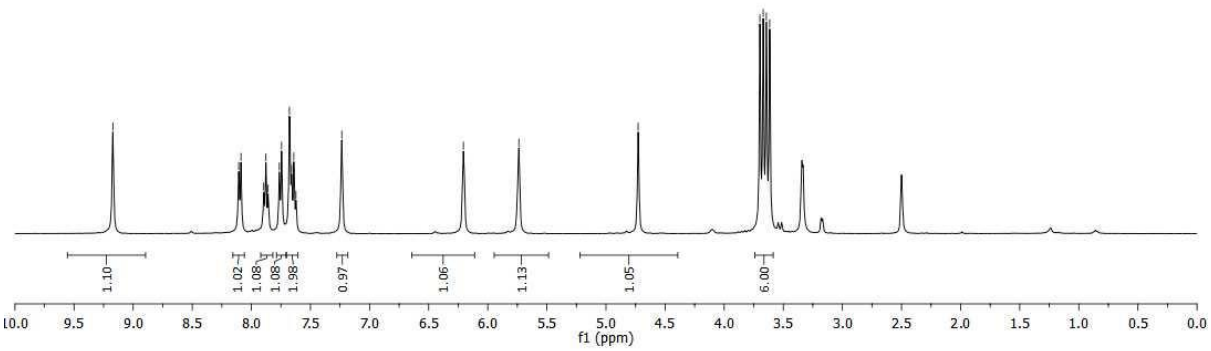
3m

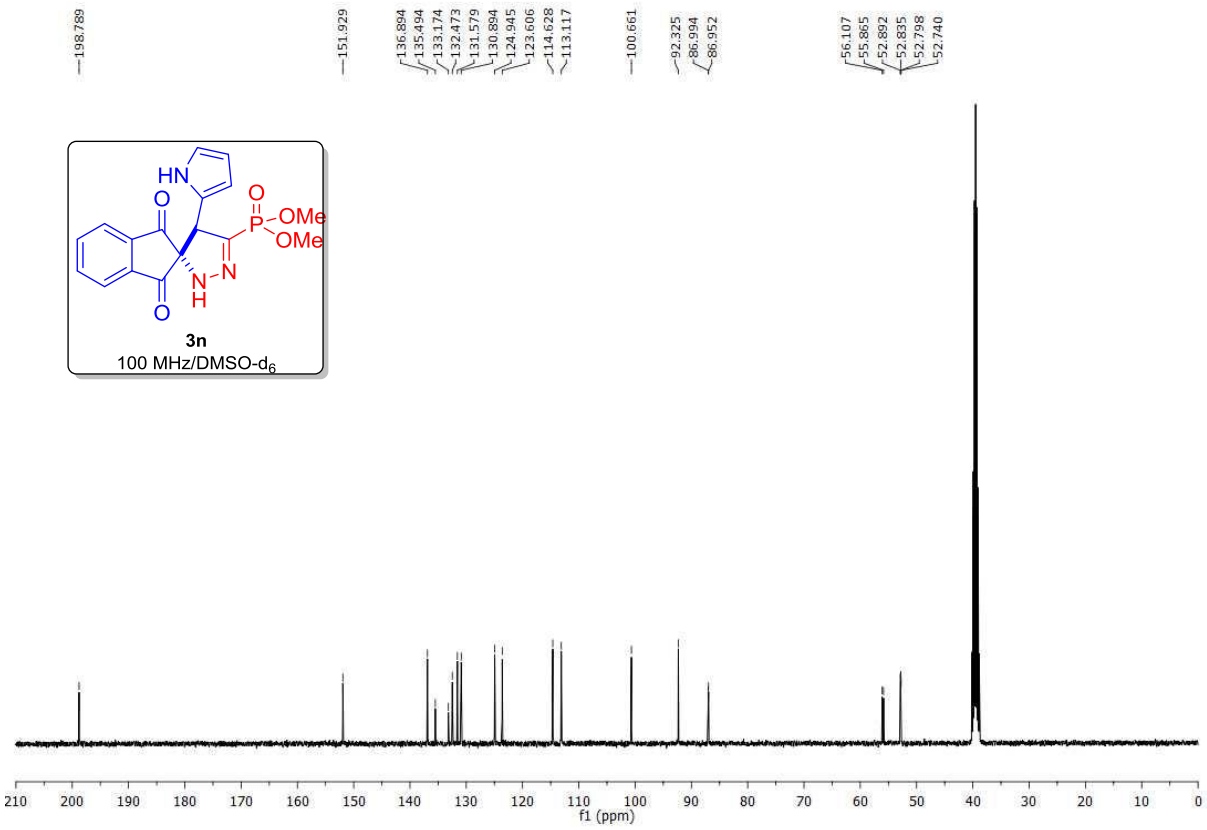
161.9 MHz/$CDCl_3$

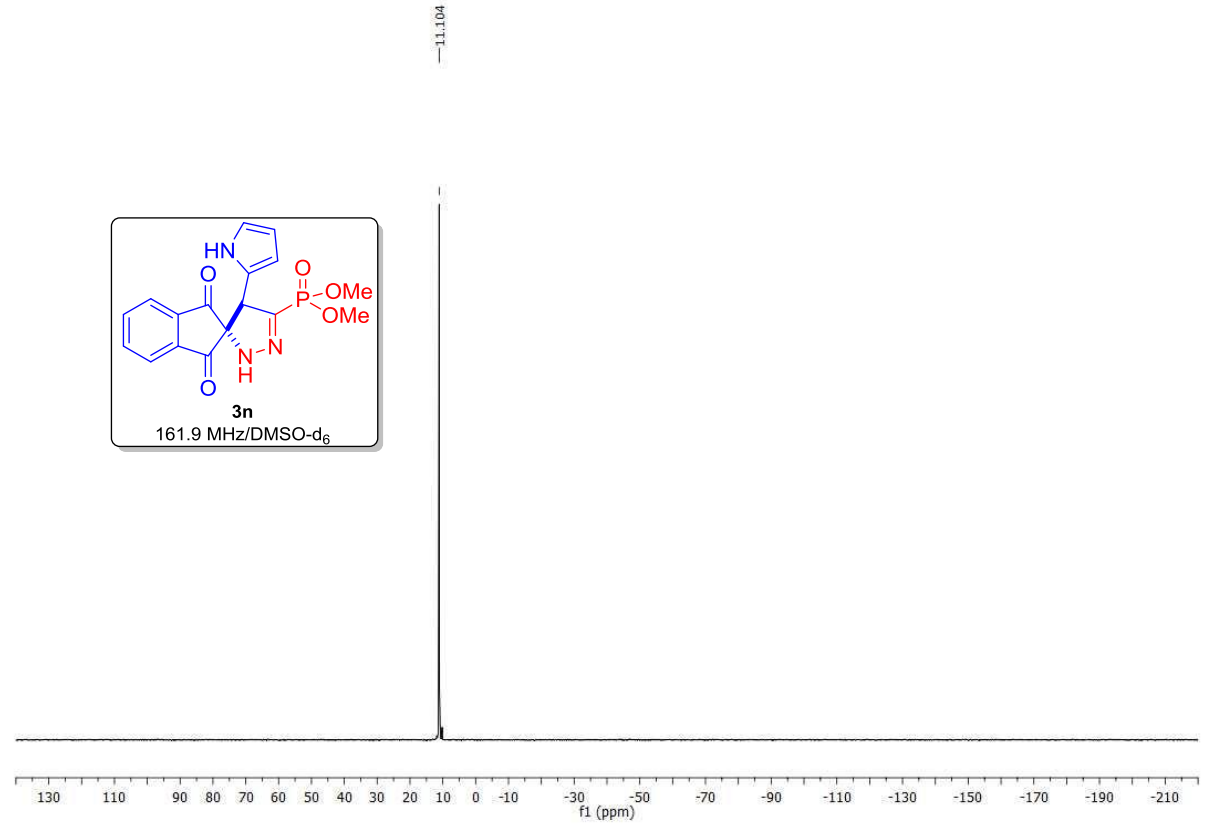


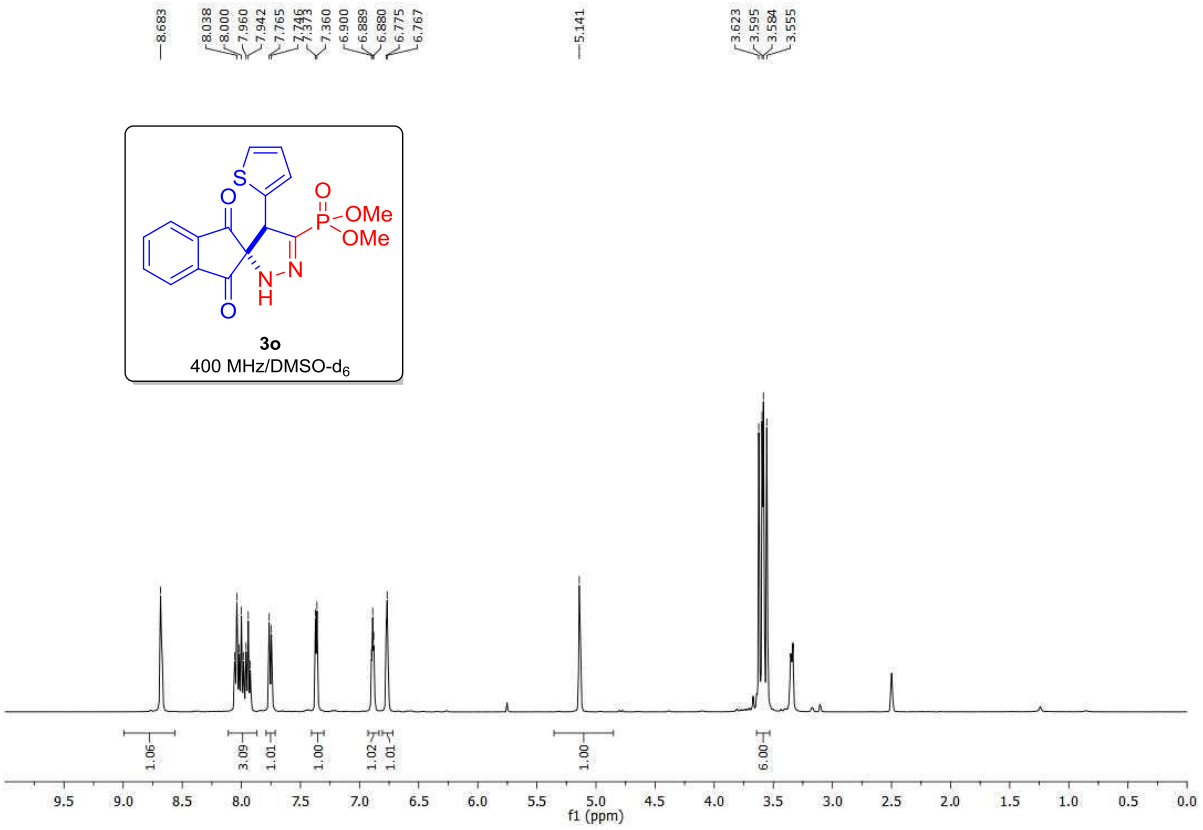
3n

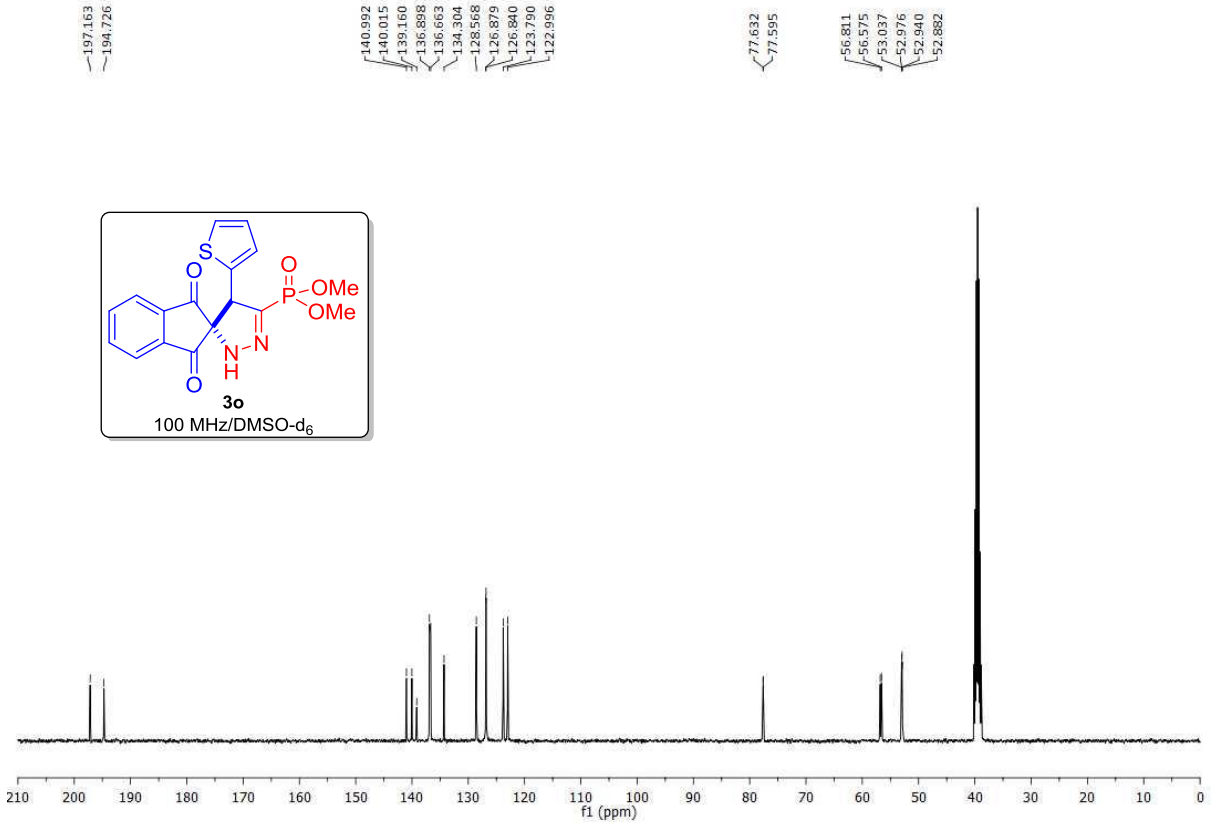
400 MHz/DMSO-d_6

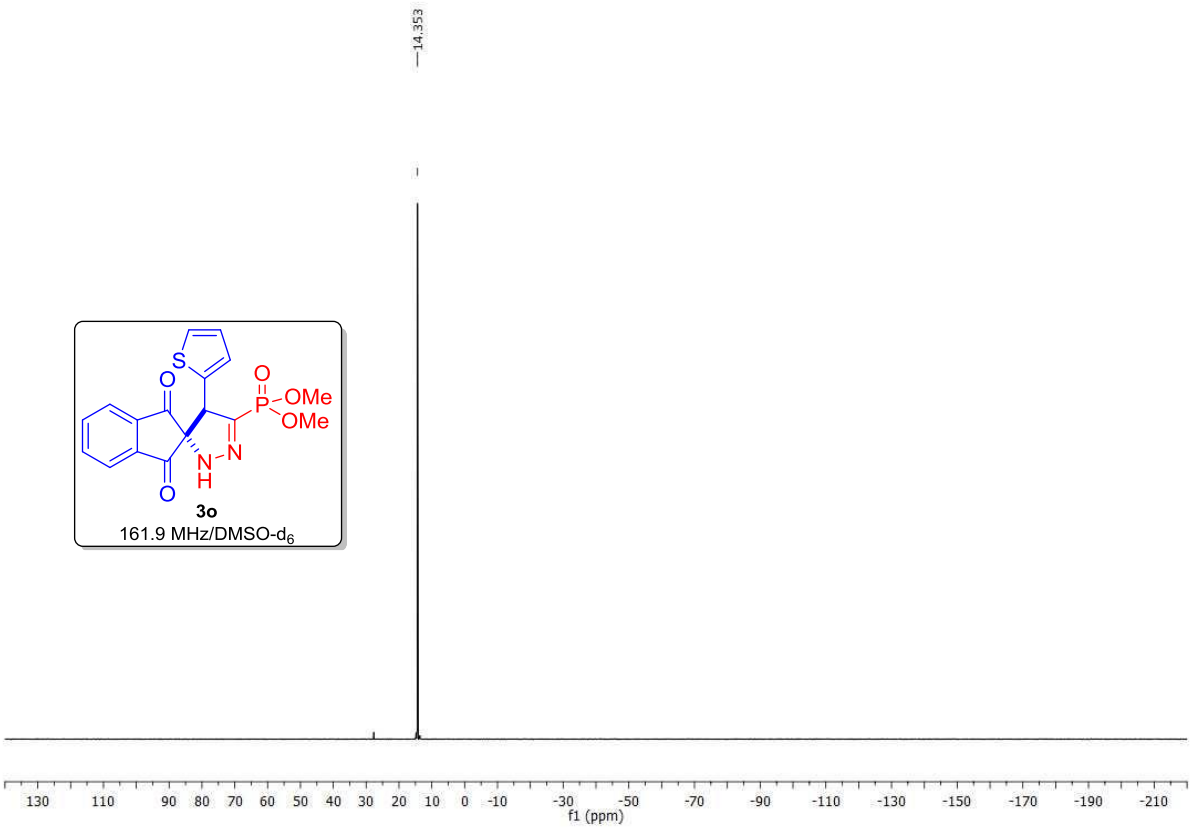



198.789
151.929
136.894
135.494
133.174
132.473
131.579
130.894
124.945
123.606
114.628
113.117
100.661
92.325
86.994
86.952
56.107
55.865
52.892
52.835
52.798
52.740
HN
O
O
P
OMe
OMe
N
N
H
O
3n
100 MHz/DMSO-d6
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

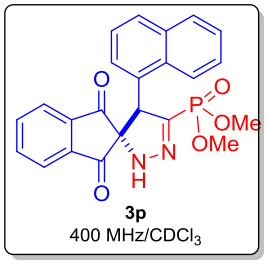

11.104
HN
O
O
P
OMe
OMe
N
N
H
O
3n
161.9 MHz/DMSO-d6
130 110 90 80 70 60 50 40 30 20 10 0 -10 -30 -50 -70 -90 -110 -130 -150 -170 -190 -210
f1 (ppm)


8.683
8.038
8.000
7.960
7.942
7.765
7.746
7.373
7.360
6.900
6.889
6.880
6.775
6.767
5.141
3.623
3.595
3.584
3.555
3o
400 MHz/DMSO-d6
1.06
3.09
1.01
1.00
1.02
1.01
1.00
6.00
f1 (ppm)

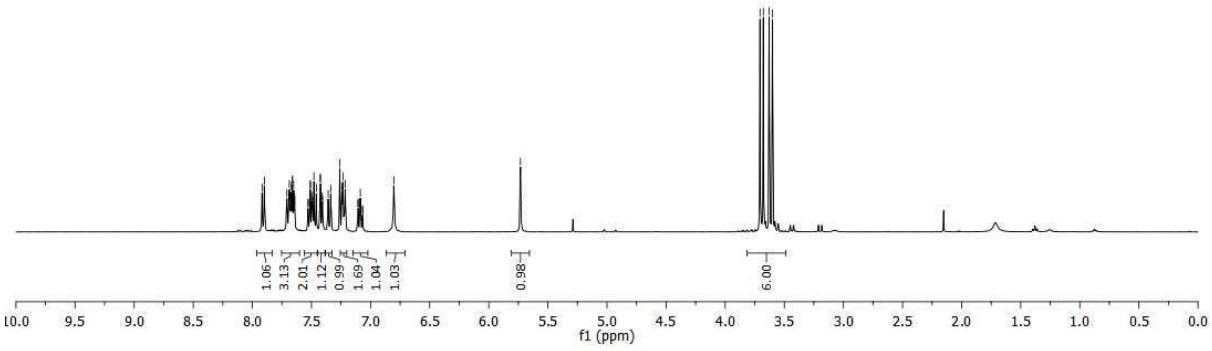

197.163
194.726
140.992
140.015
139.160
136.898
136.663
134.304
128.568
126.879
126.840
123.790
122.996
77.632
77.595
56.811
56.575
53.037
52.976
52.940
52.882
3o
100 MHz/DMSO-d6
f1 (ppm)

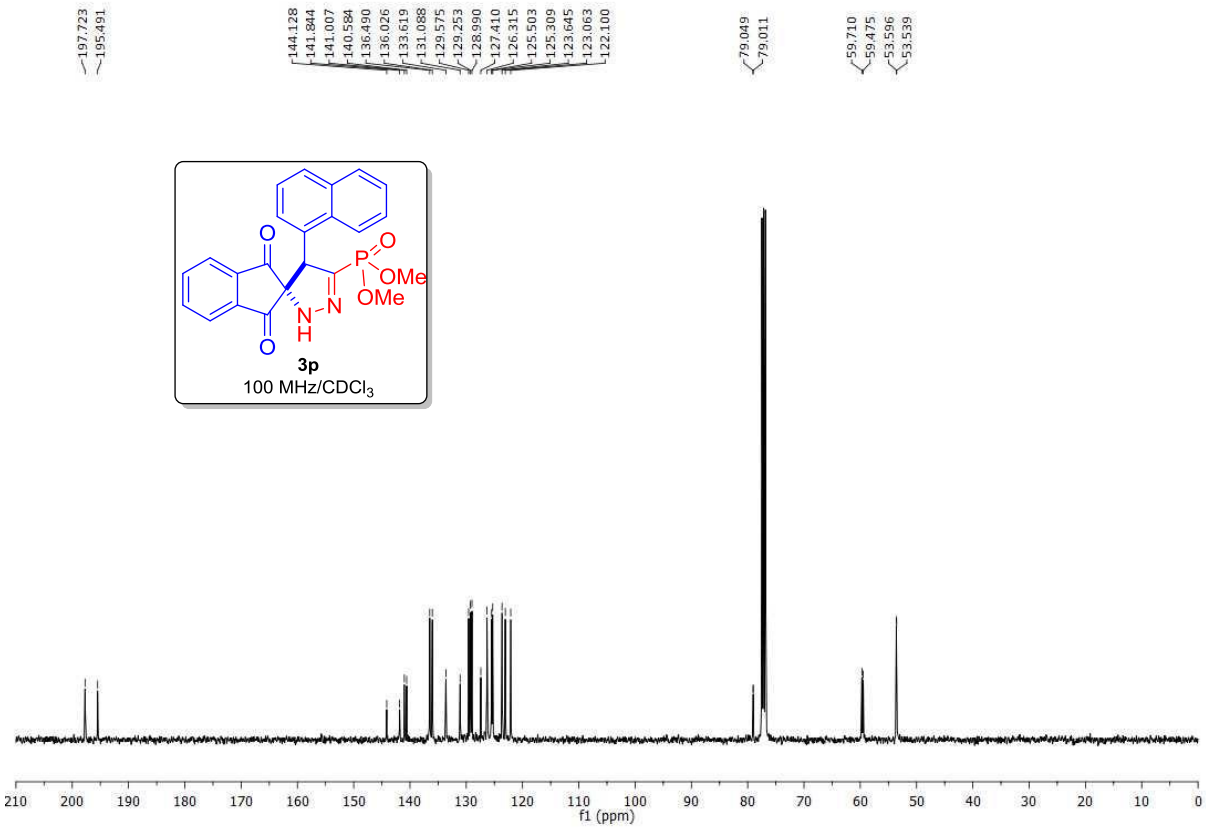


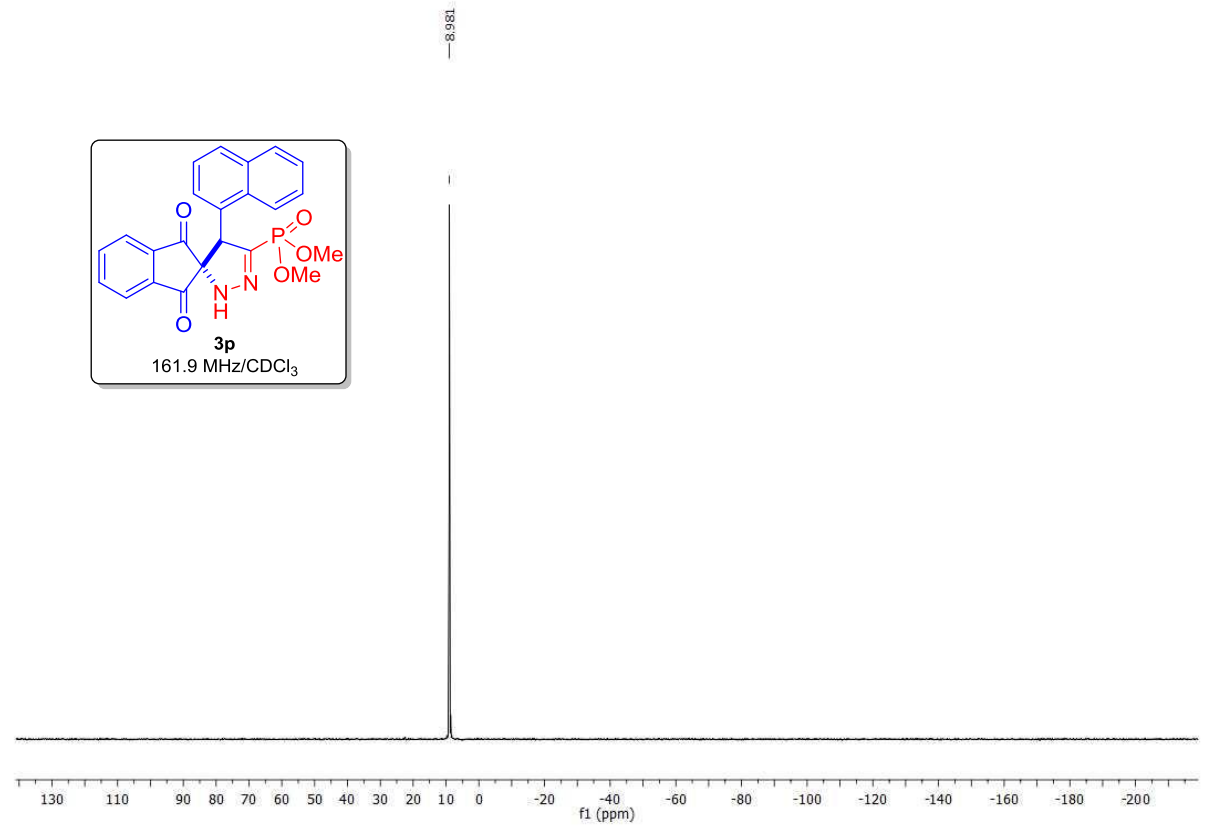
3o
161.9 MHz/DMSO-d_6

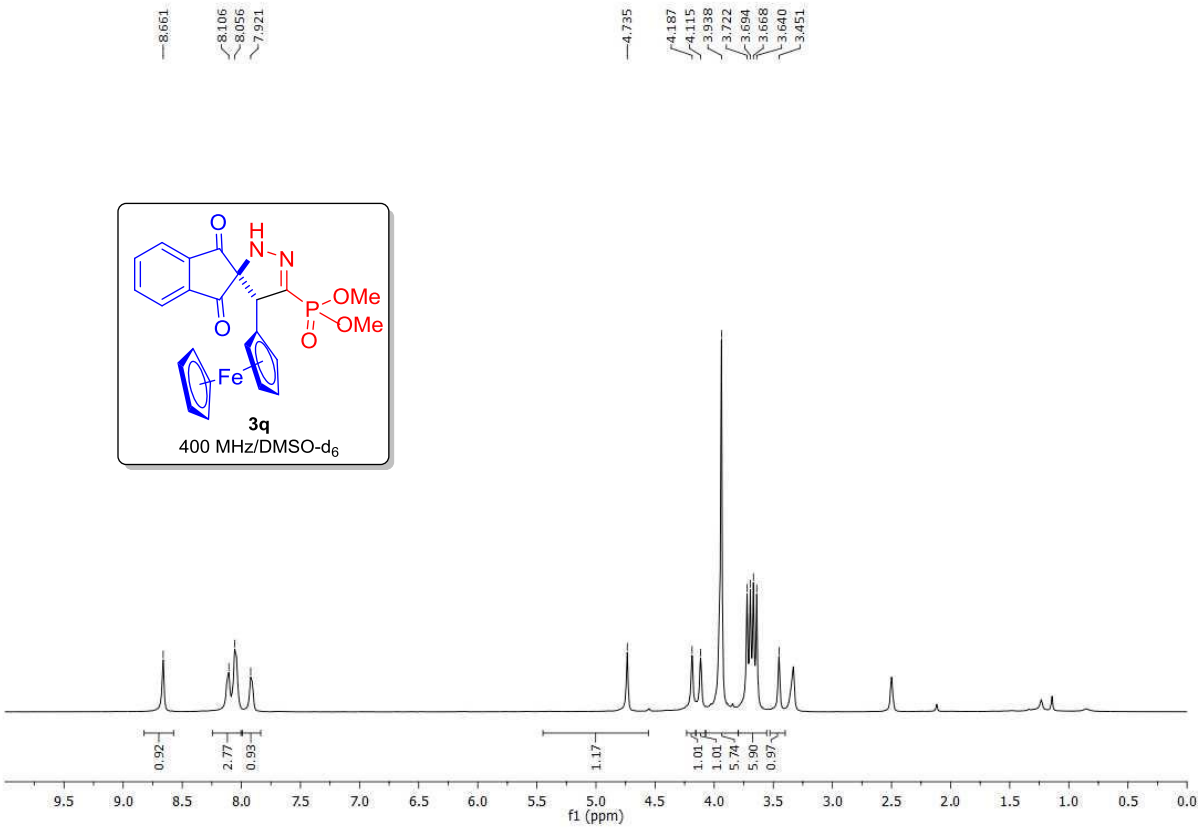


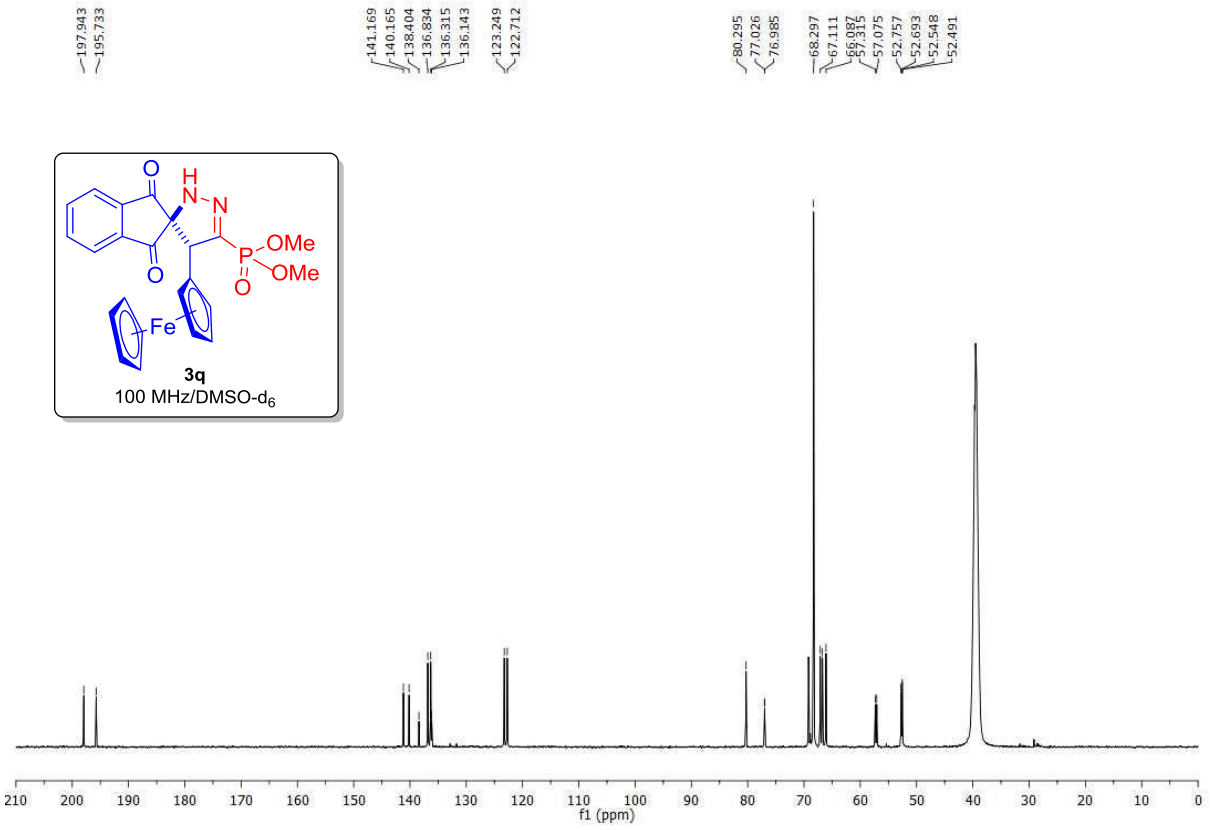
3p
400 MHz/$CDCl_3$

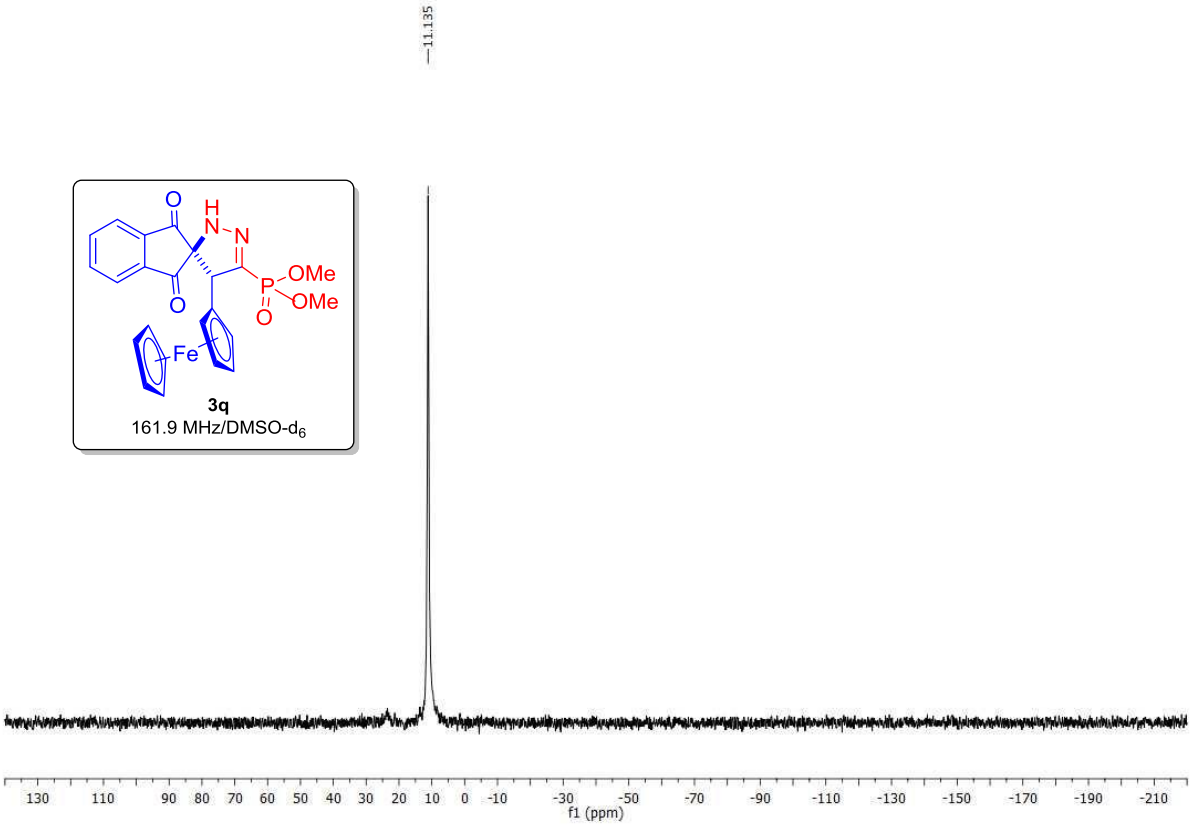


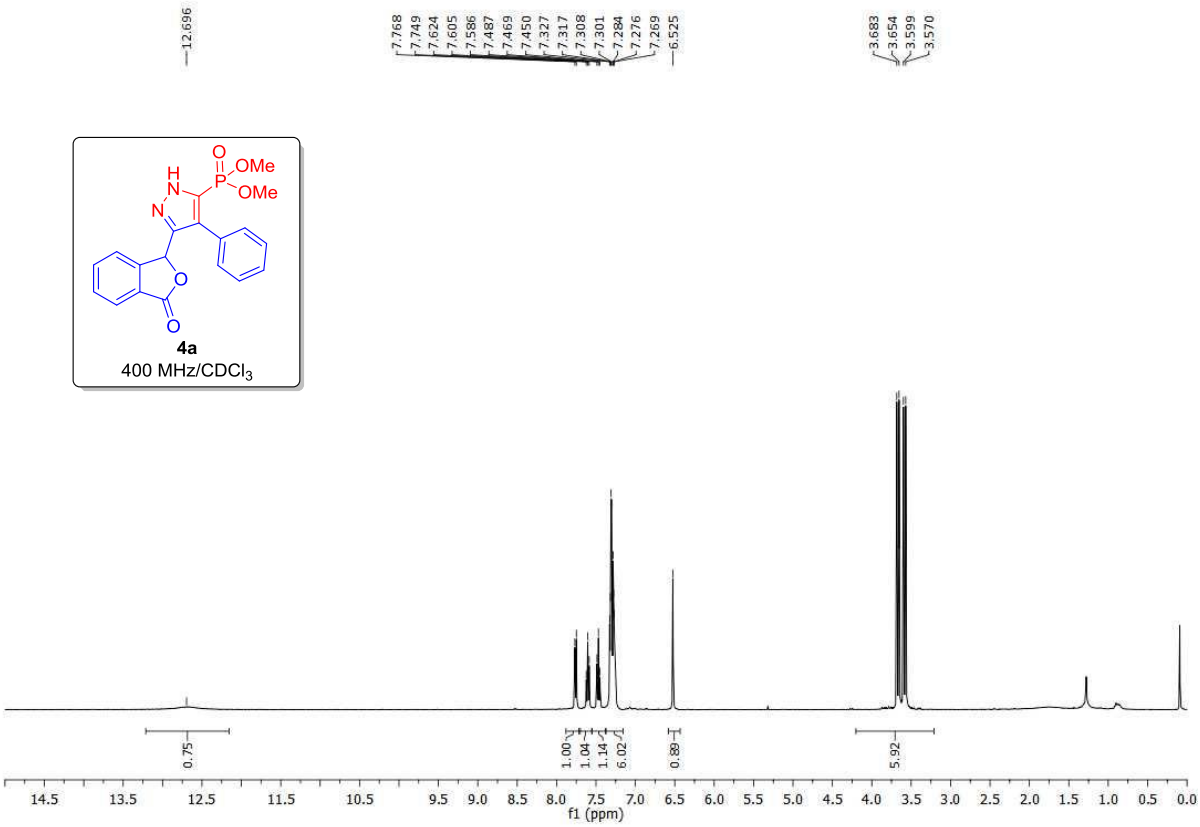

197.723
195.491
144.128
141.844
141.007
140.584
136.490
136.026
133.619
131.088
129.575
129.253
128.990
127.410
126.315
125.503
125.309
123.645
123.063
122.100
79.049
79.011
59.710
59.475
53.596
53.539
O
O
P
OMe
OMe
N
N
H
O
3p
100 MHz/CDCl3
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

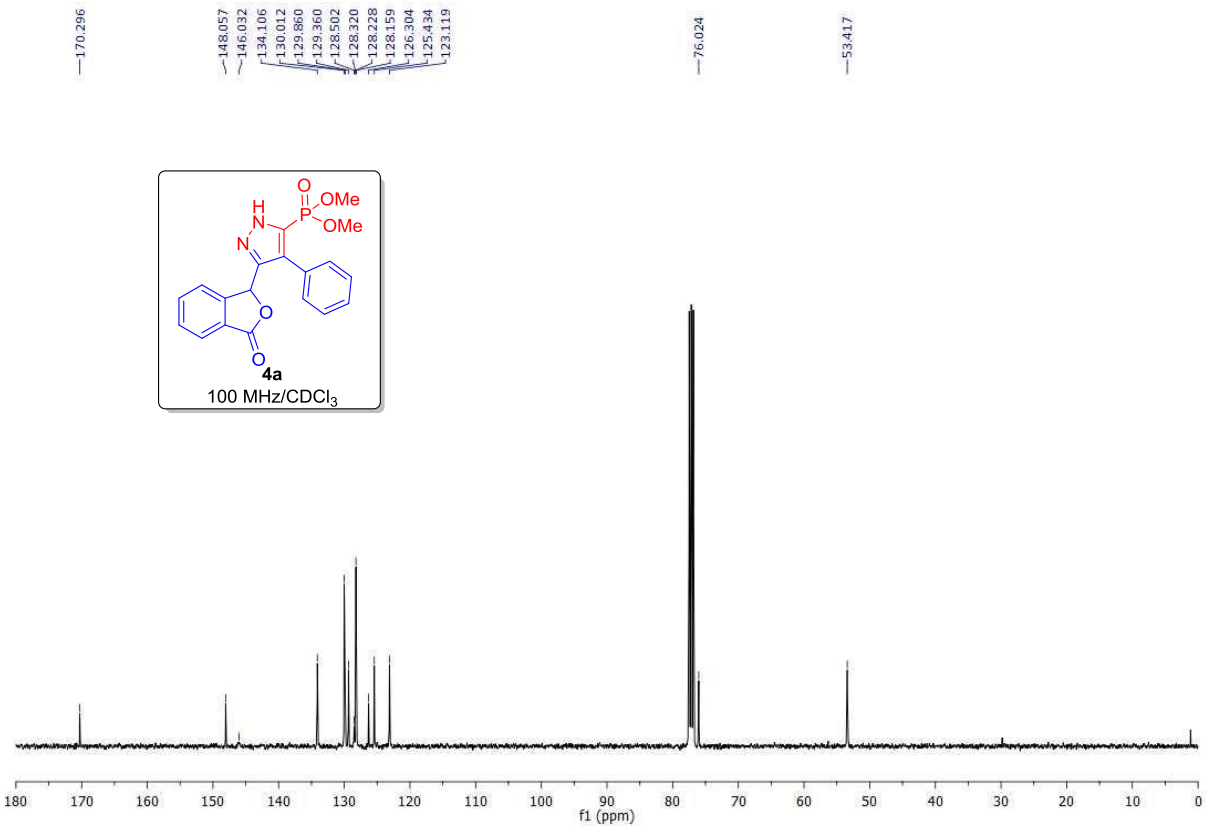

8.981
O
O
P
OMe
OMe
N
N
H
O
3p
161.9 MHz/CDCl3
130
110
90
80
70
60
50
40
30
20
10
0
-20
-40
-60
-80
-100
-120
-140
-160
-180
-200
f1 (ppm)

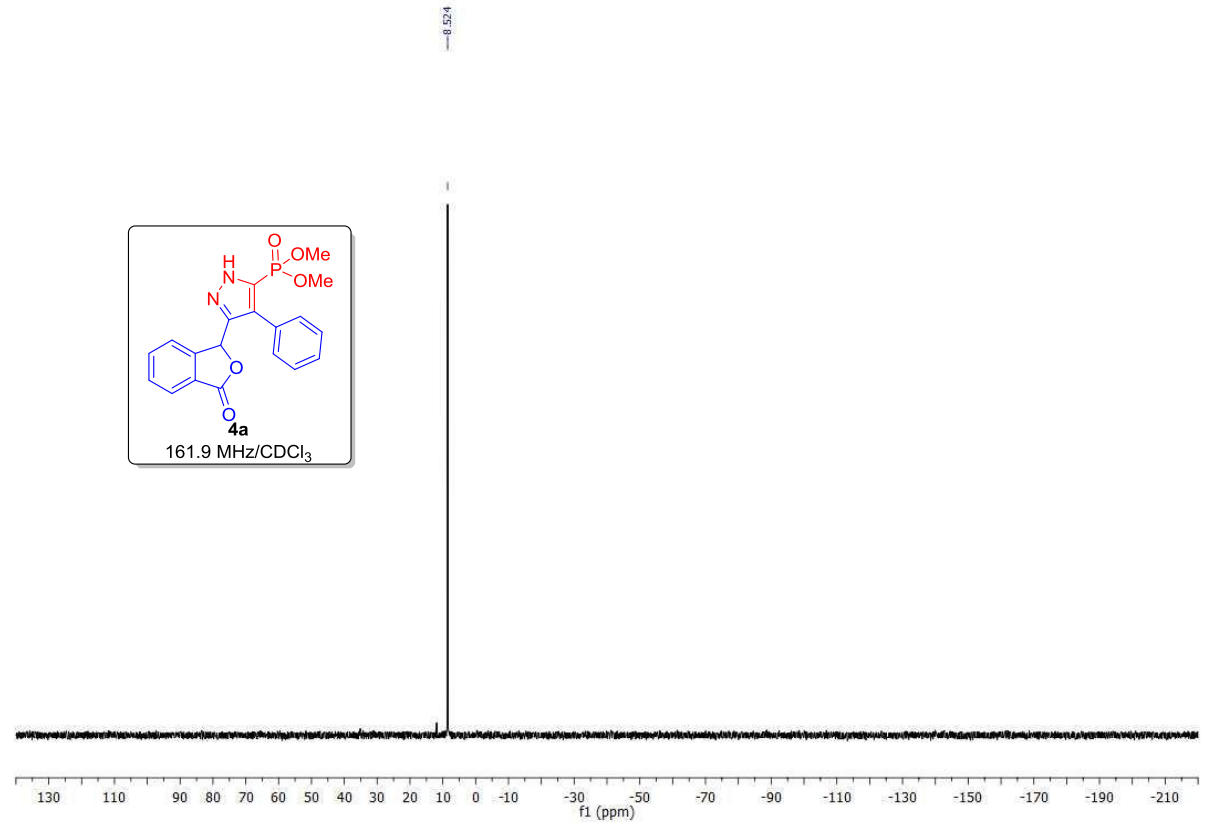

8.661
8.106
8.056
7.921
4.735
4.187
4.115
3.938
3.722
3.694
3.668
3.640
3.451
3q
400 MHz/DMSO-d6
0.92
2.77
0.93
1.17
1.01
1.01
5.74
5.90
0.97
f1 (ppm)

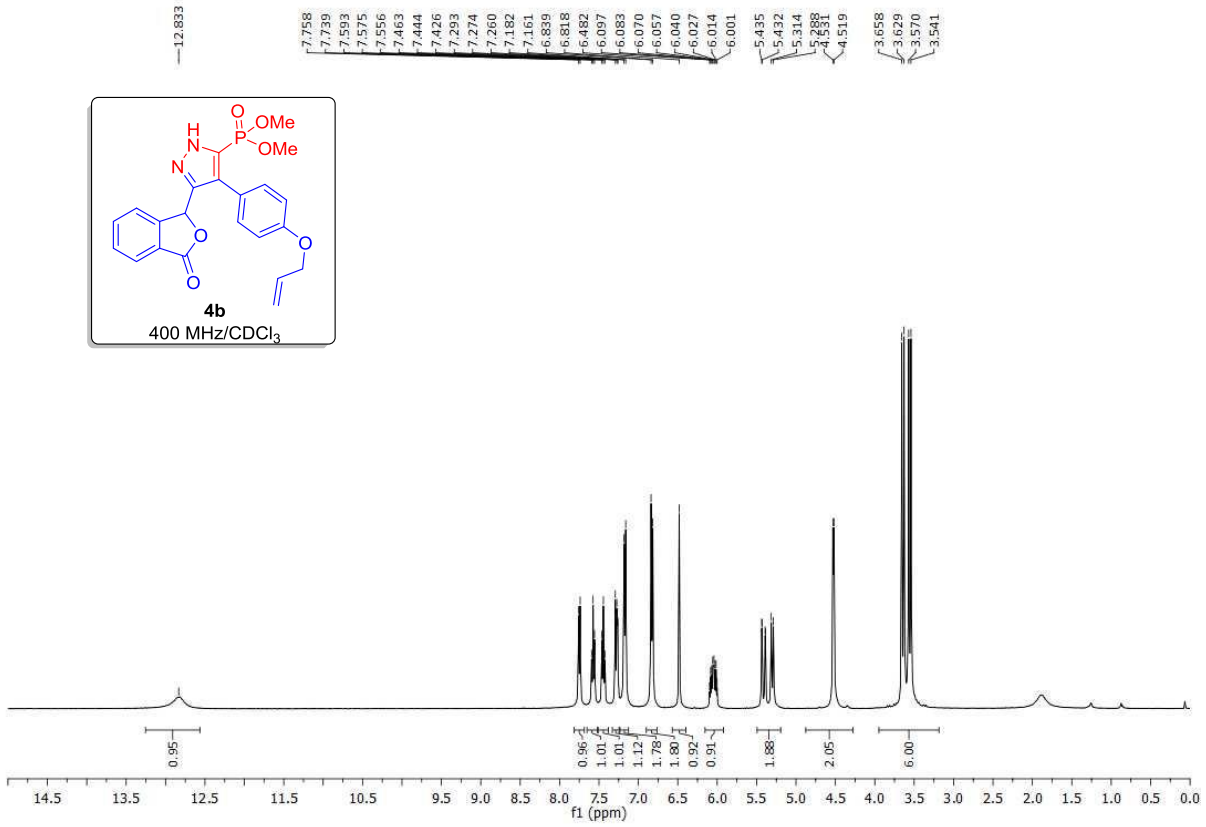

197.943
195.733
141.169
140.165
138.404
136.834
136.315
136.143
123.249
122.712
80.295
77.026
76.985
68.297
67.111
57.075
52.757
52.693
52.548
52.491
3q
100 MHz/DMSO-d6
f1 (ppm)

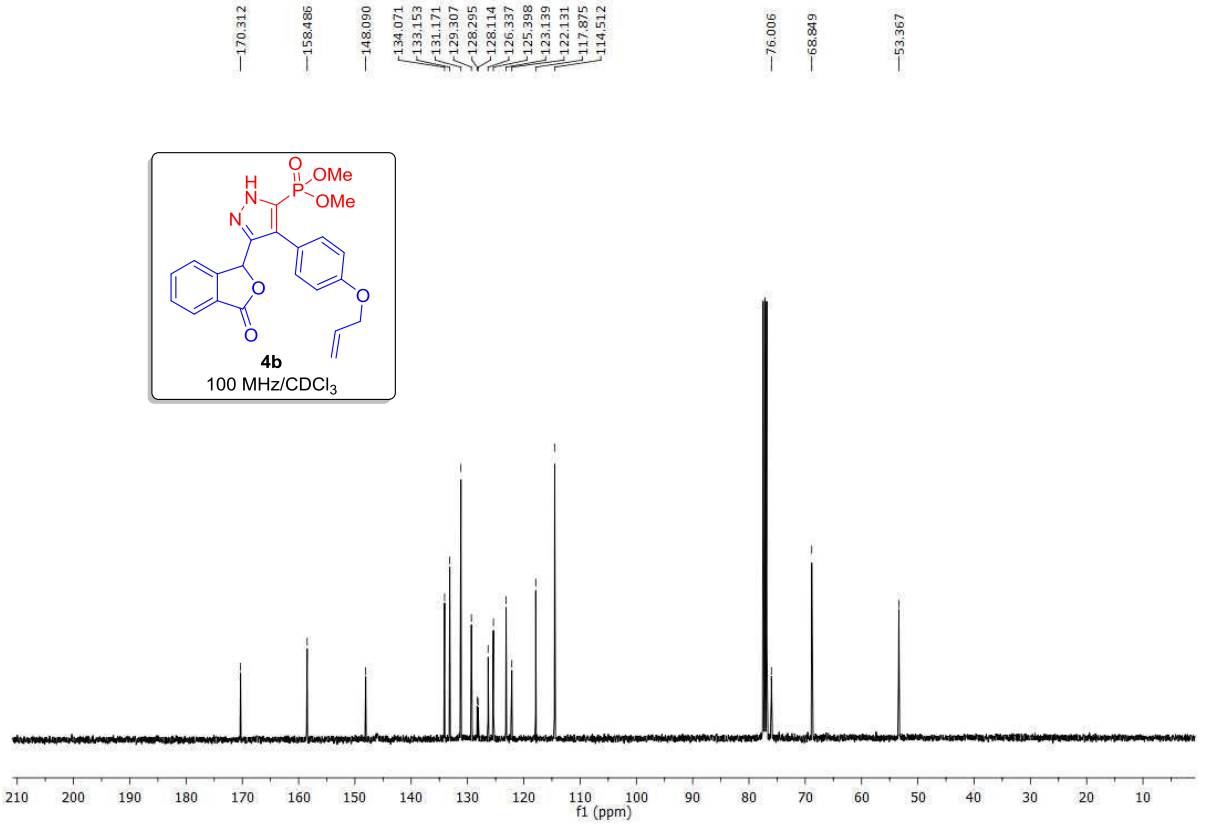

11.135
3q
161.9 MHz/DMSO-d6
f1 (ppm)

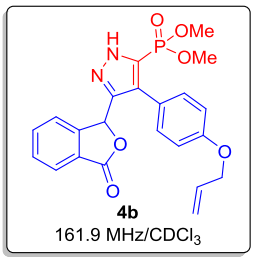

12.696
7.768
7.749
7.624
7.605
7.586
7.487
7.469
7.450
7.327
7.317
7.308
7.301
7.284
7.276
7.269
6.525
3.683
3.654
3.599
3.570
4a
400 MHz/CDCl3
0.75
1.00
1.04
1.14
6.02
0.89
5.92
f1 (ppm)


170.296
148.057
146.032
134.106
130.012
129.860
129.360
128.502
128.320
128.228
128.159
126.304
125.434
123.119
76.024
53.417
4a
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


8.524
4a
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -10 -30 -50 -70 -90 -110 -130 -150 -170 -190 -210
f1 (ppm)

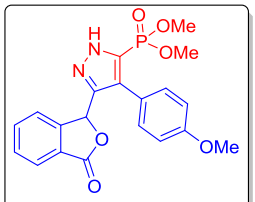

12.833
7.758
7.739
7.593
7.575
7.556
7.463
7.444
7.426
7.293
7.274
7.260
7.182
7.161
6.839
6.818
6.482
6.097
6.083
6.070
6.057
6.040
6.027
6.014
6.001
5.435
5.432
5.314
5.288
5.281
4.519
3.658
3.629
3.570
3.541
OMe
OMe
4b
400 MHz/CDCl3
0.95
0.96
1.01
1.01
1.12
1.78
1.80
0.92
0.91
1.88
2.05
6.00
f1 (ppm)


170.312
158.486
148.090
134.071
133.153
131.171
129.307
128.295
128.114
126.337
125.398
123.139
122.131
117.875
114.512
76.006
68.849
53.367
OMe
OMe
4b
100 MHz/CDCl3
f1 (ppm)



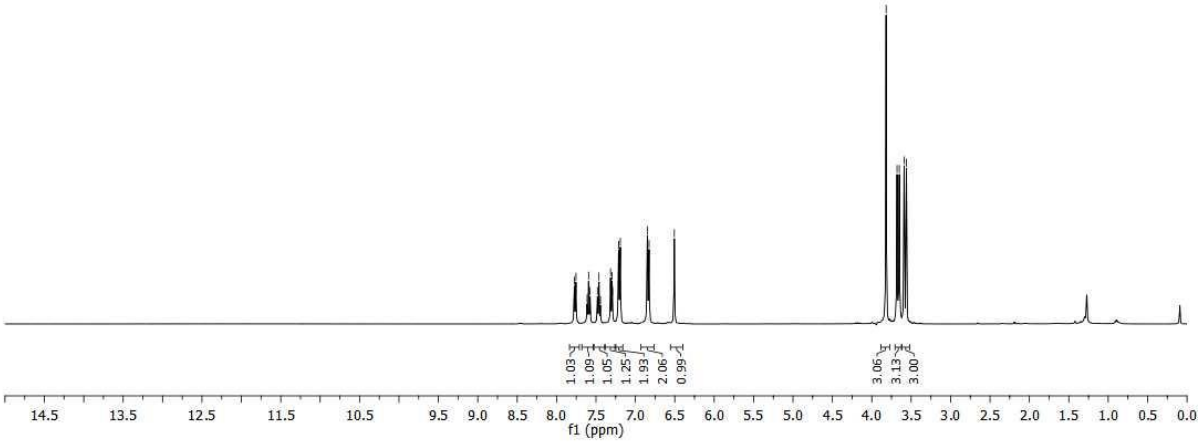
4b

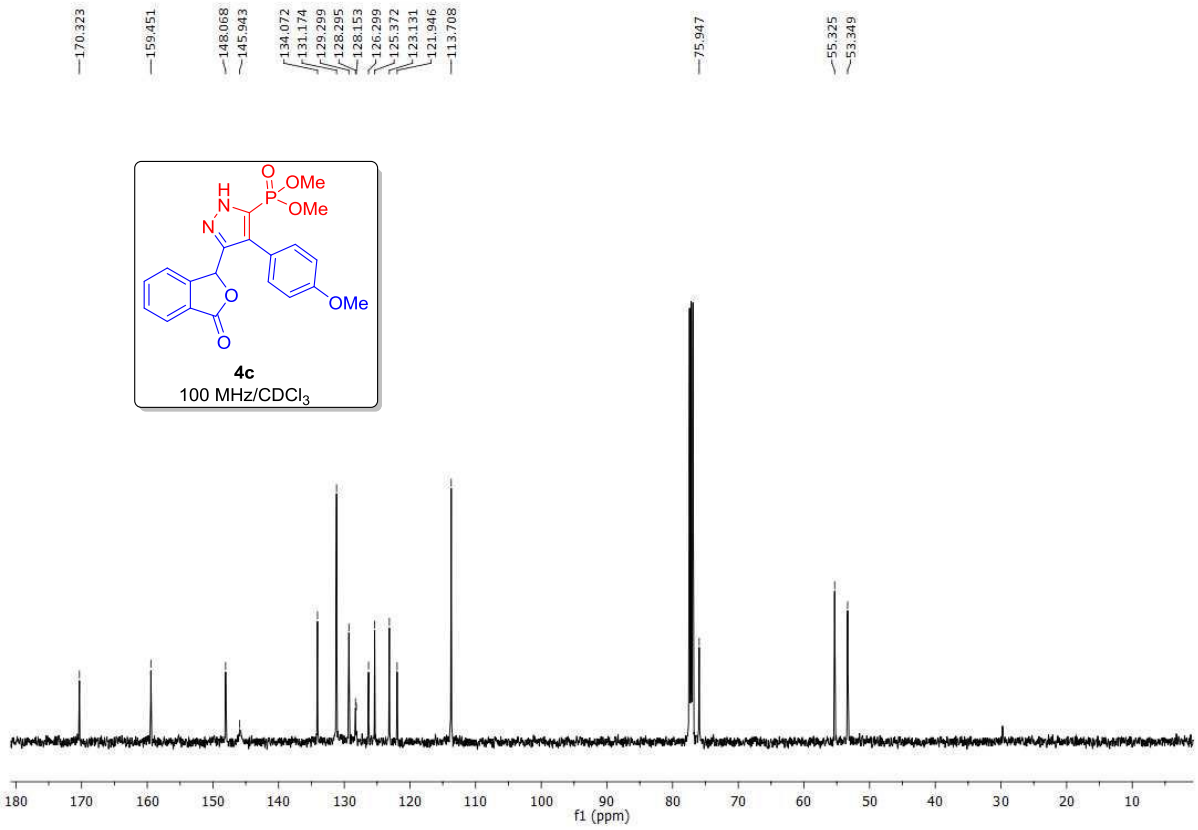
161.9 MHz/$CDCl_3$

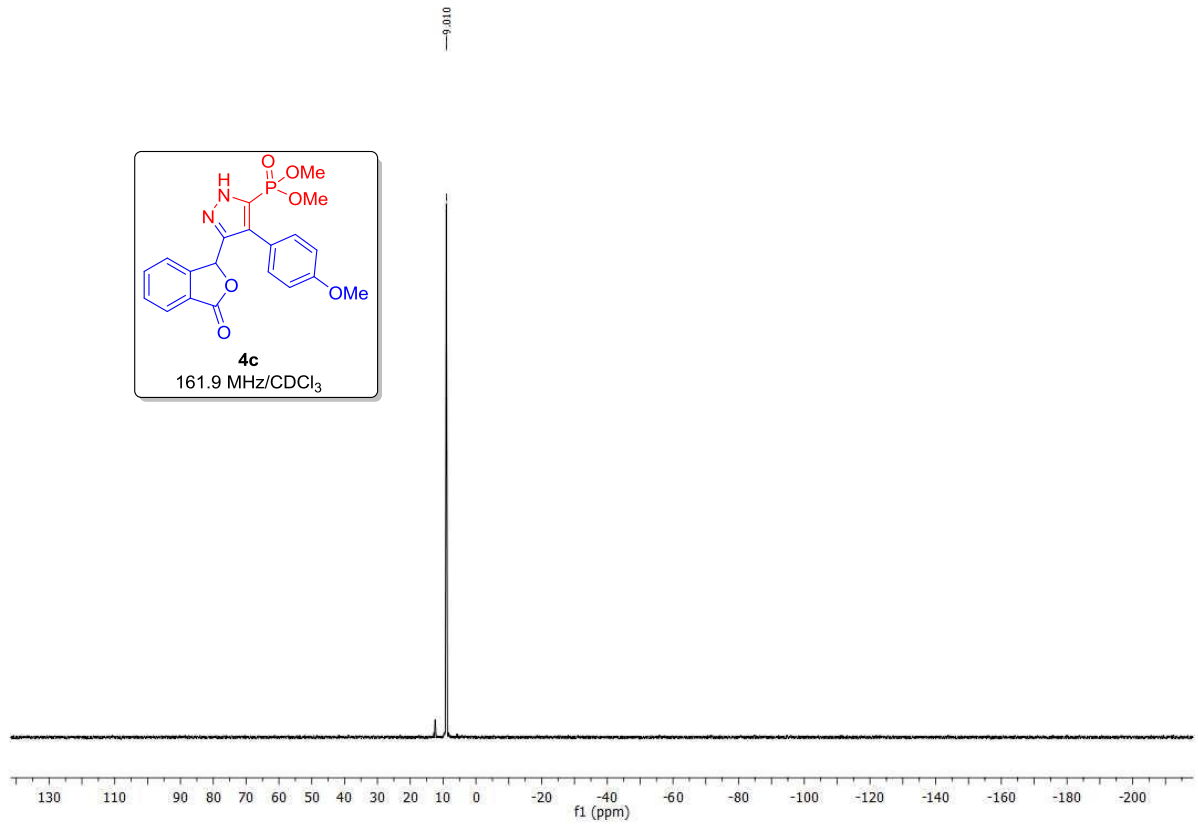


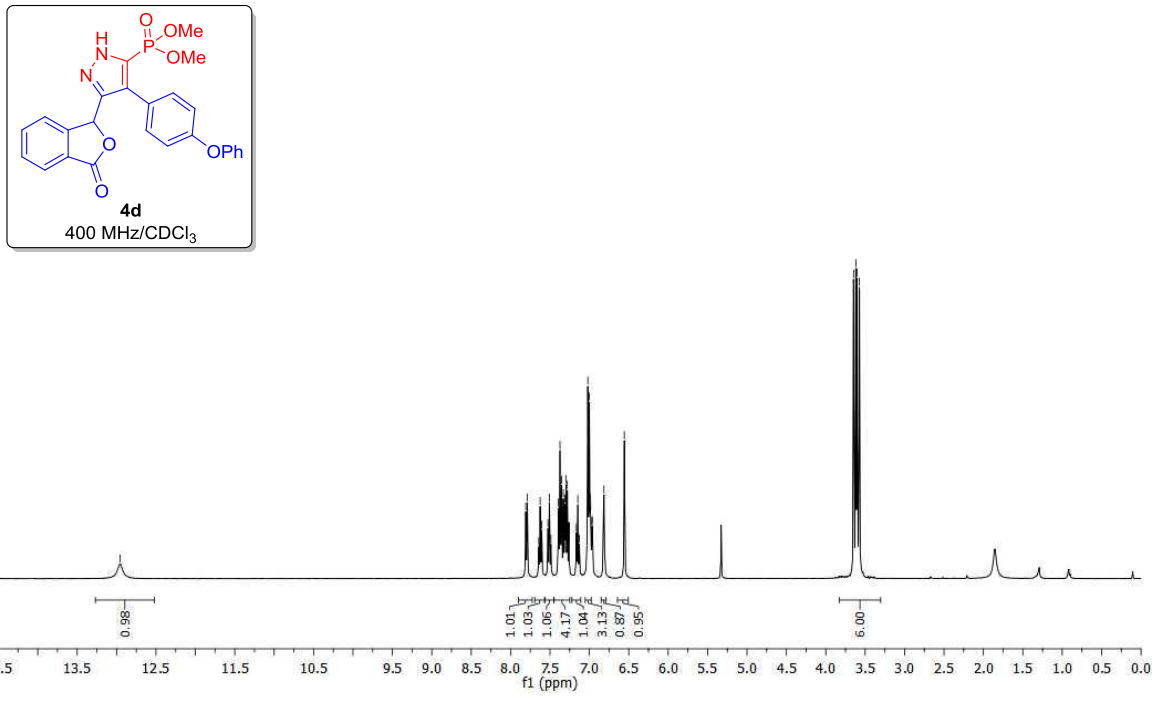
4c

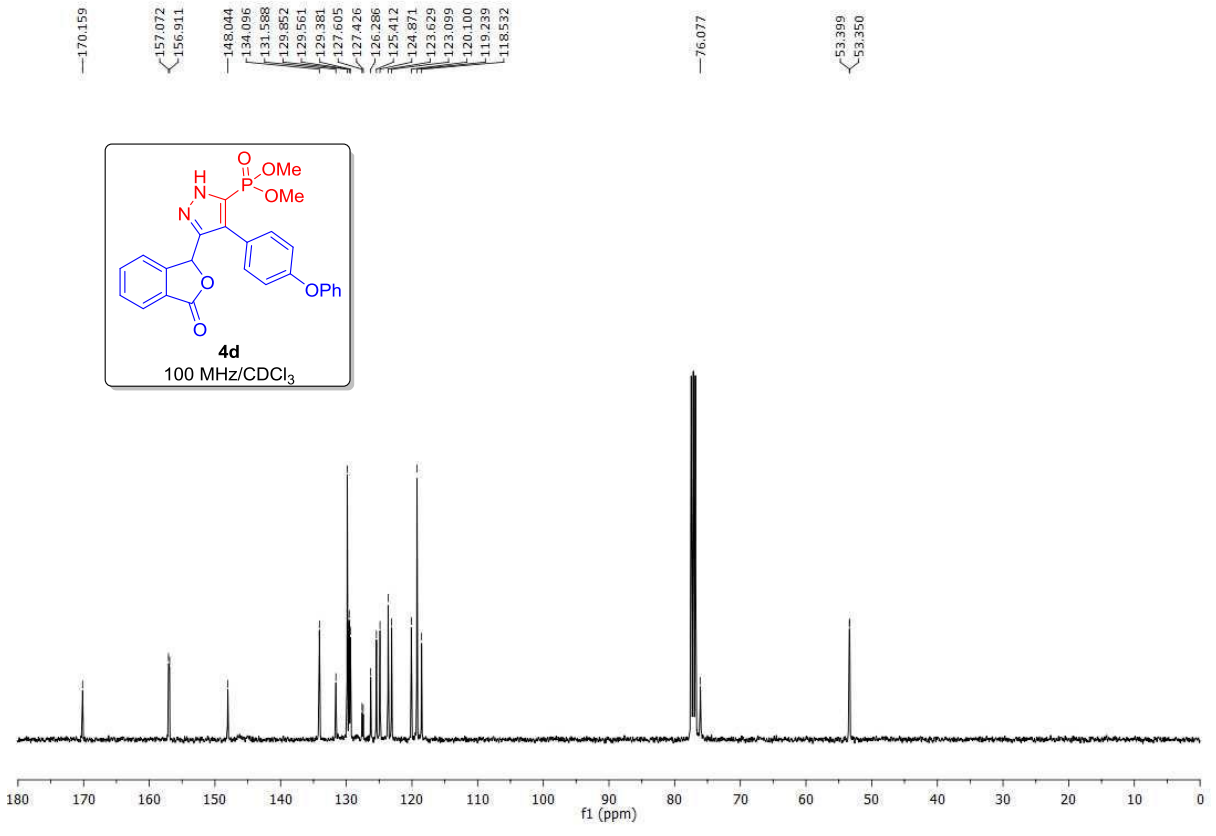
400 MHz/$CDCl_3$

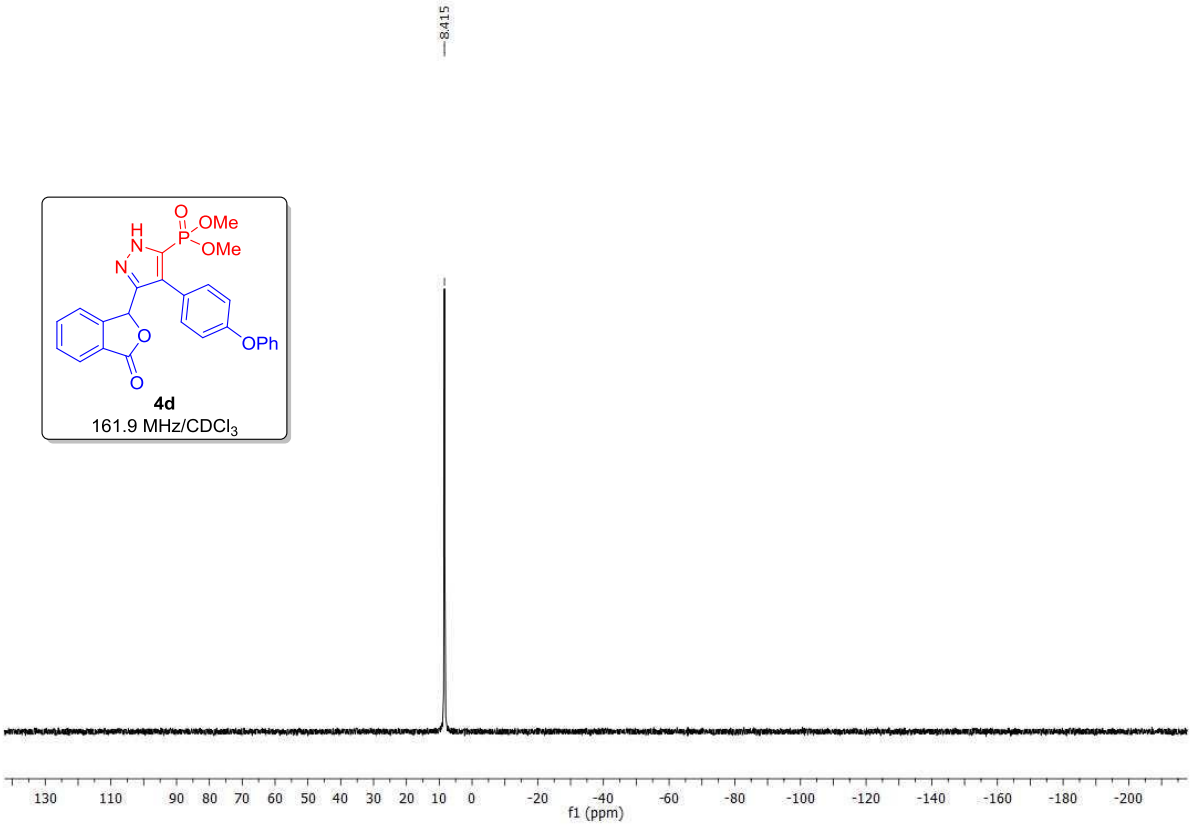


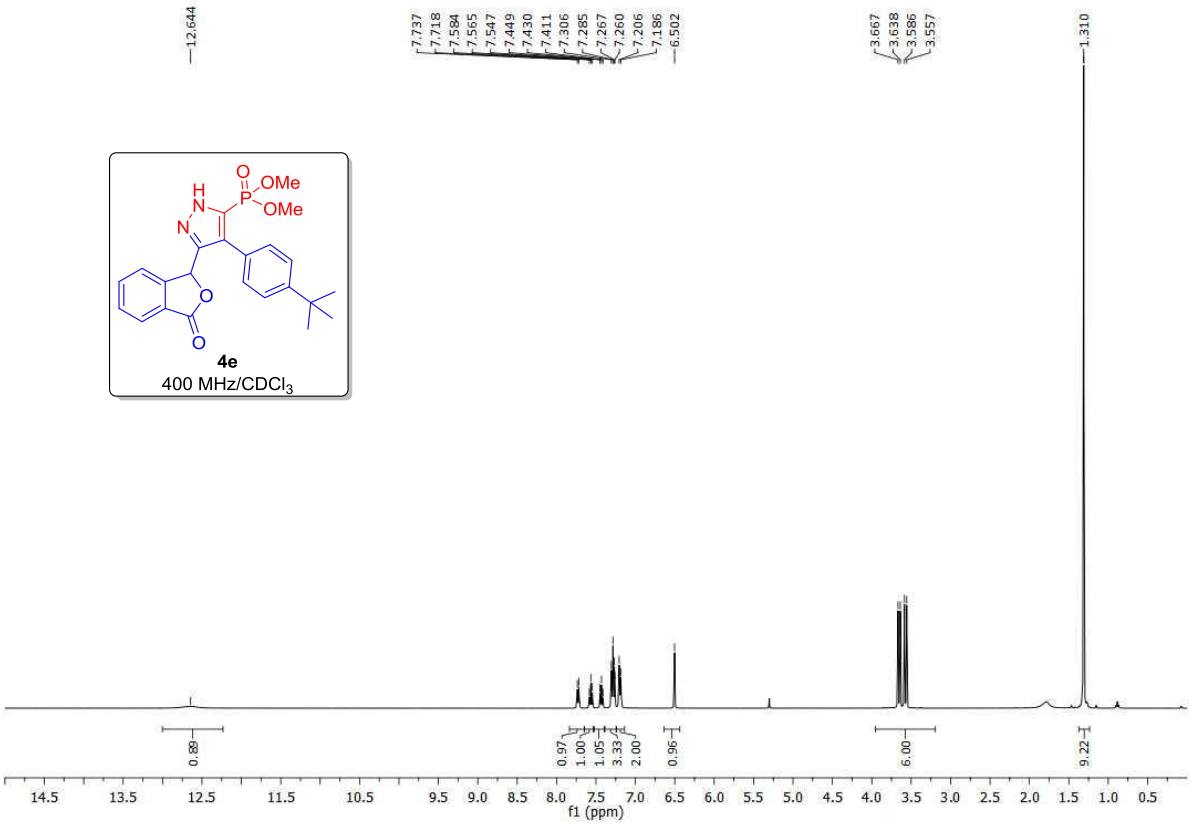

170.323
159.451
148.068
145.943
134.072
131.174
129.299
128.295
128.153
126.299
125.372
123.131
121.946
113.708
75.947
55.325
53.349
OMe
OMe
OMe
4c
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

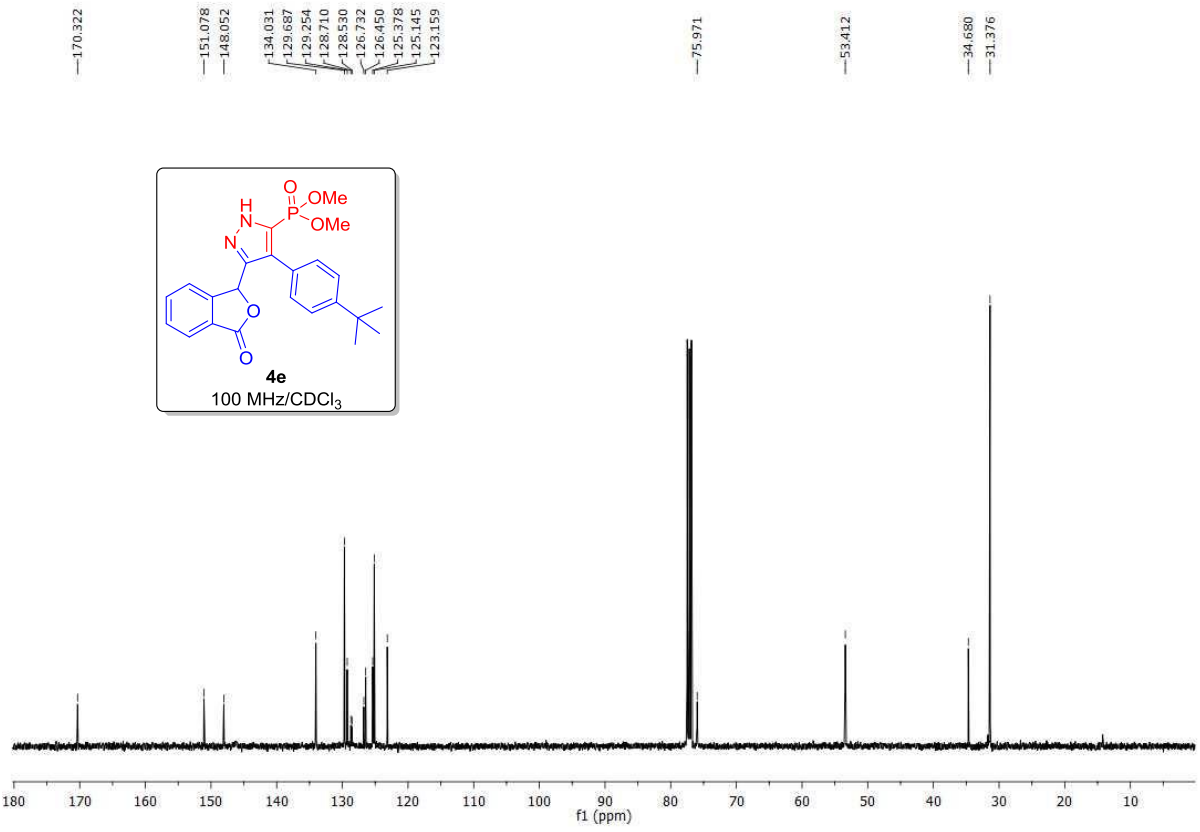

9.010
OMe
OMe
OMe
4c
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

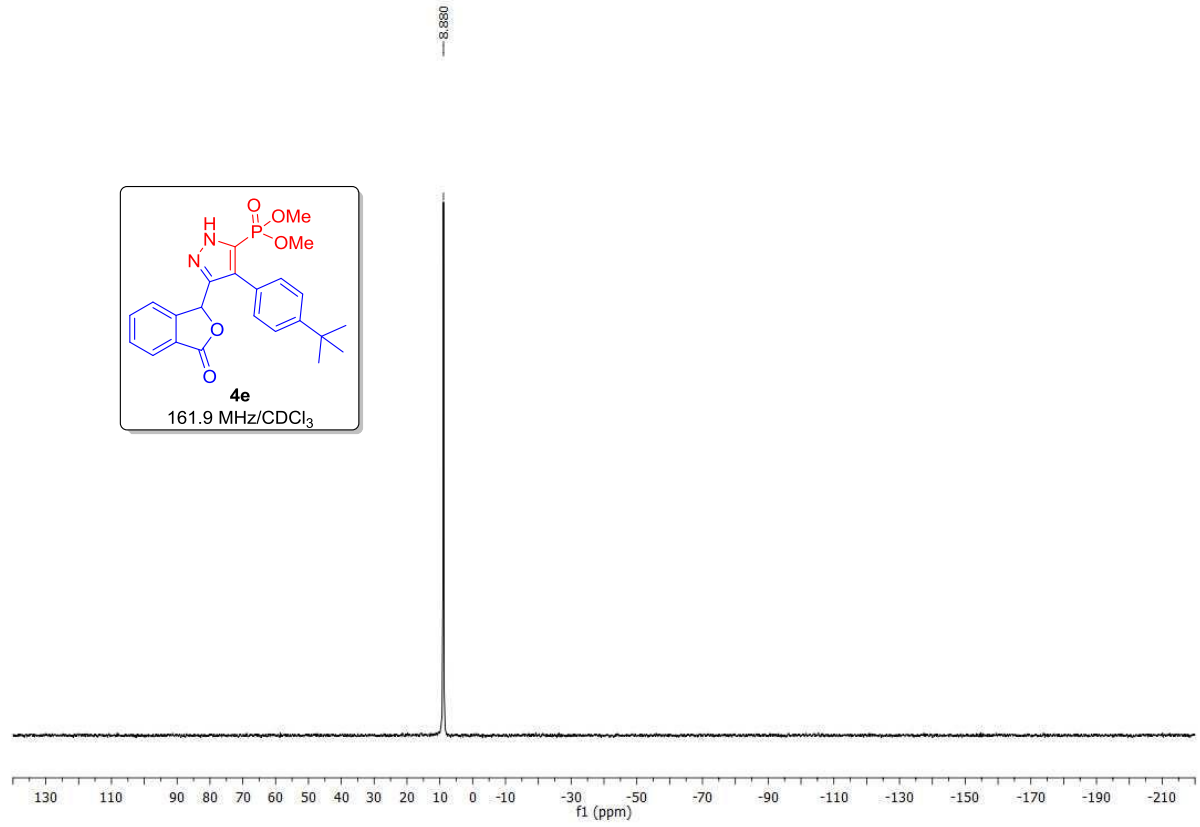

12.954
7.807
7.788
7.642
7.625
7.606
7.526
7.507
7.488
7.393
7.374
7.353
7.330
7.311
7.296
7.280
7.165
7.147
7.128
7.020
7.000
6.990
6.984
6.969
6.963
6.816
6.556
3.648
3.619
3.603
3.574
OMe
OMe
OPh
4d
400 MHz/CDCl3
0.98
1.01
1.03
1.06
4.17
1.04
3.13
0.87
0.95
6.00
f1 (ppm)

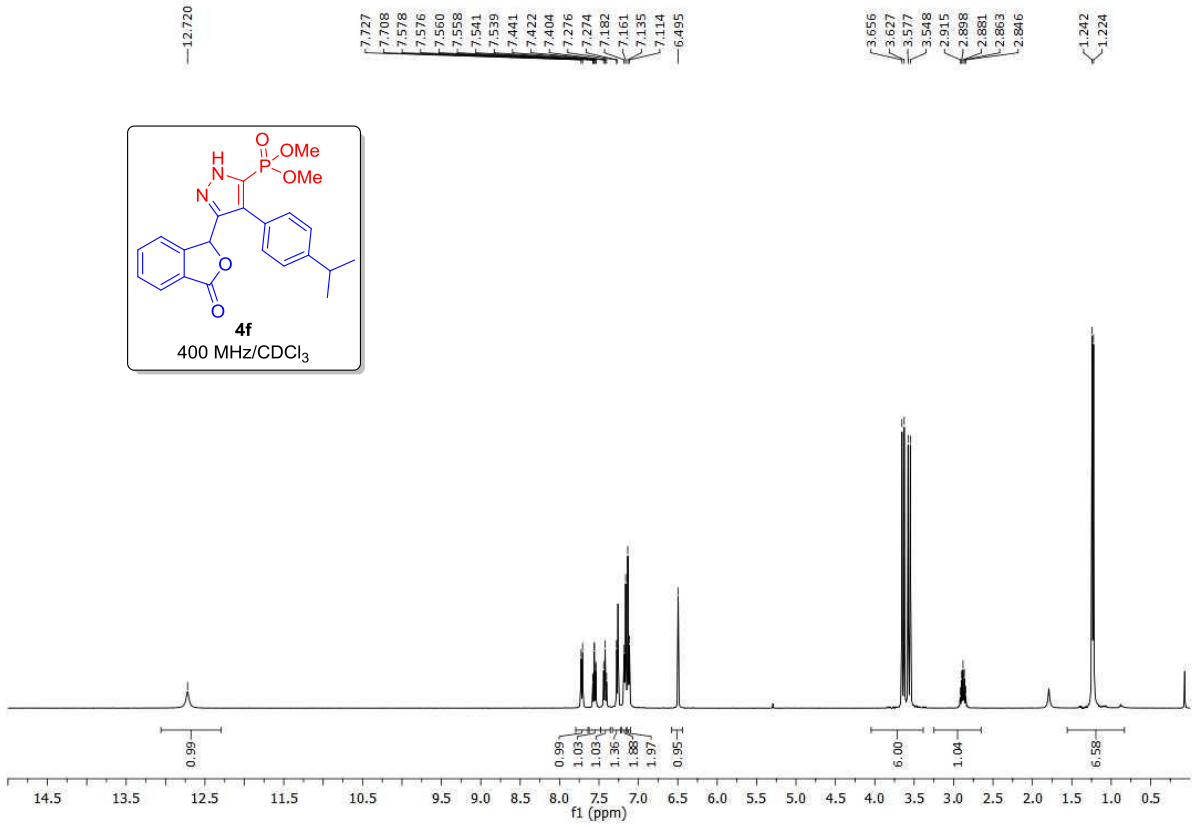

170.159
157.072
156.911
148.044
134.096
131.588
129.852
129.561
129.381
127.605
127.426
126.286
125.412
124.871
123.629
123.099
120.100
119.239
118.532
76.077
53.399
53.350
OMe
OMe
OPh
4d
100 MHz/CDCl3
f1 (ppm)

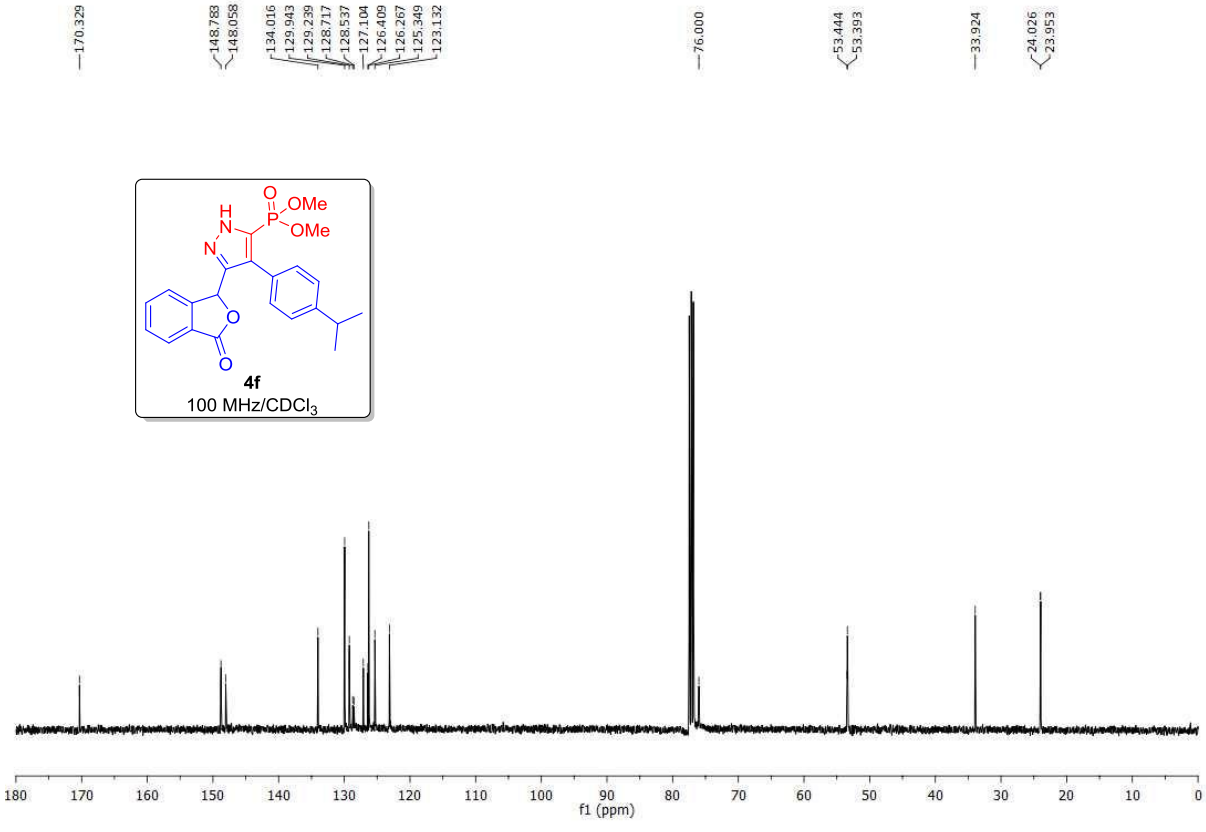

8.415
OMe
OMe
OPh
4d
161.9 MHz/CDCl3
f1 (ppm)

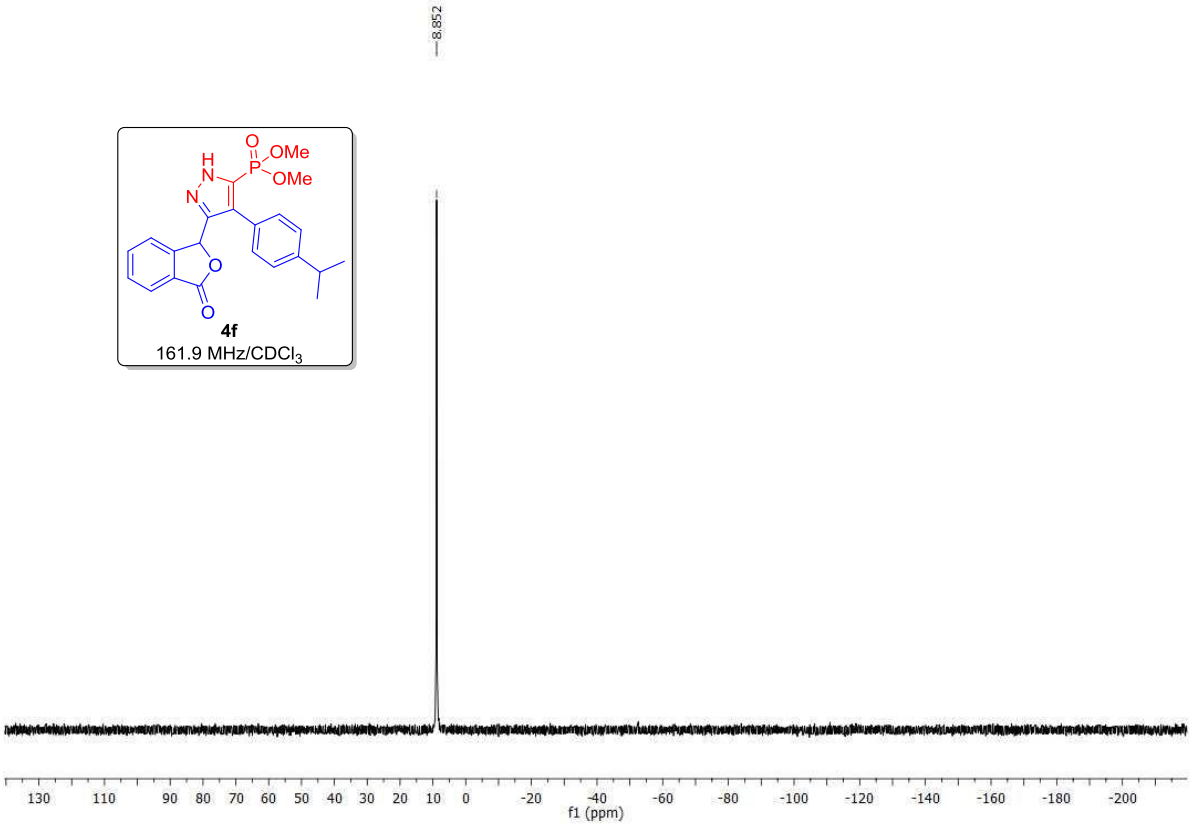

12.644
7.737
7.718
7.584
7.565
7.547
7.449
7.430
7.411
7.306
7.285
7.267
7.260
7.206
7.186
6.502
3.667
3.638
3.586
3.557
1.310
OMe
OMe
4e
400 MHz/CDCl3
0.88
0.97
1.00
1.05
3.33
2.00
0.96
6.00
9.22
f1 (ppm)

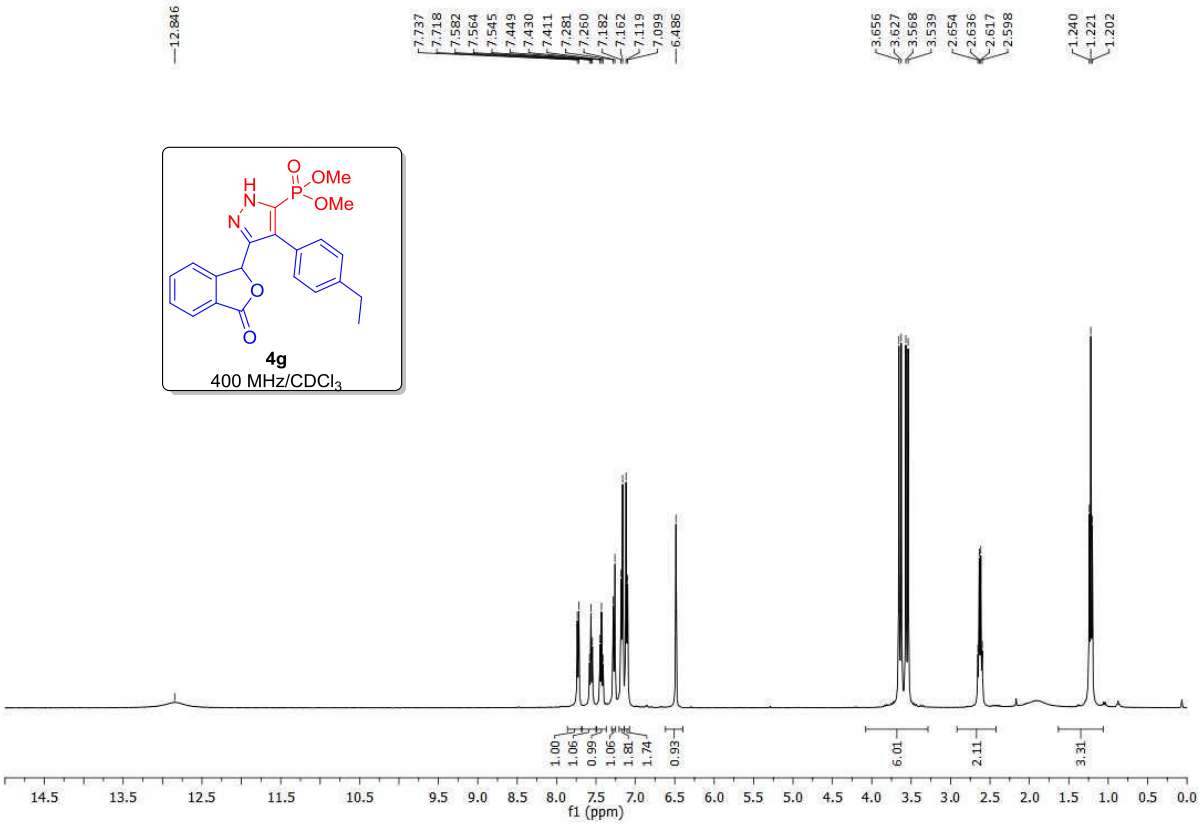

170.322
151.078
148.052
134.031
129.687
129.254
128.710
128.530
126.732
126.450
125.378
125.145
123.159
75.971
53.412
34.680
31.376
OMe
OMe
4e
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

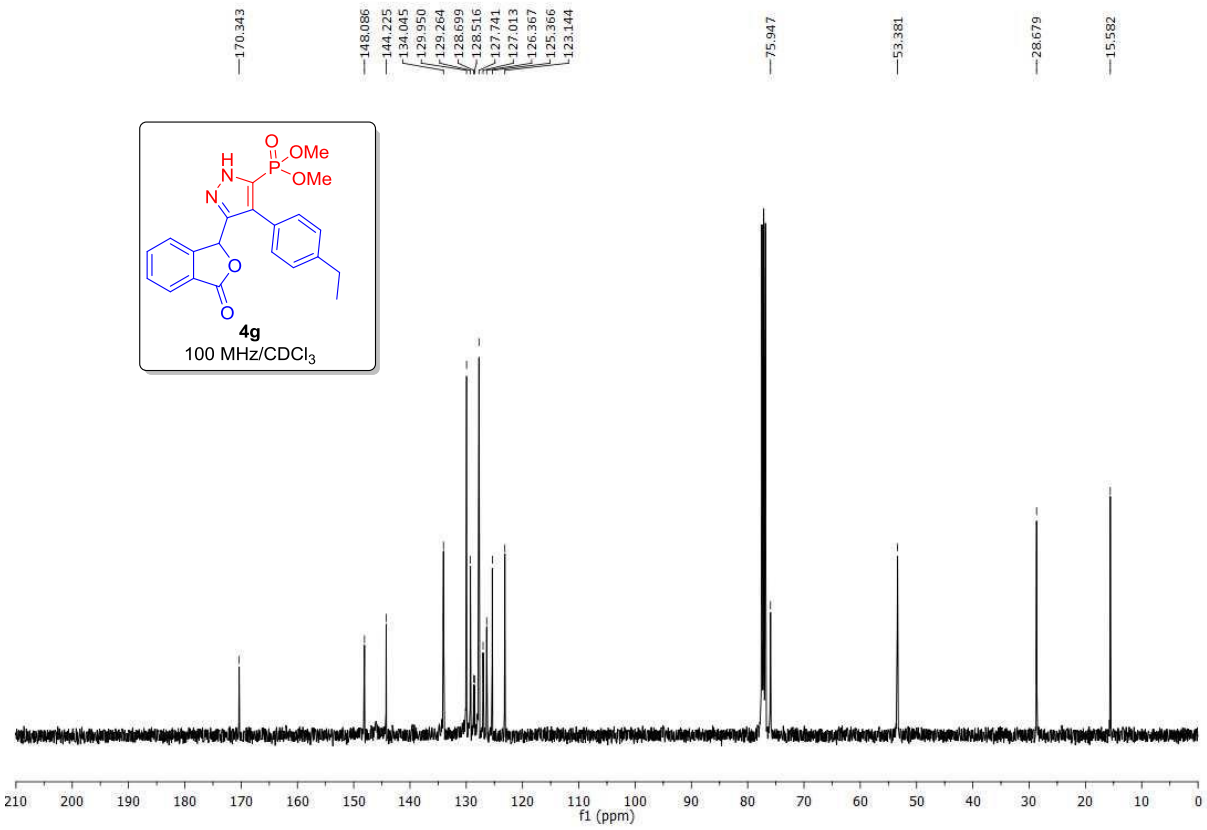

8.880
OMe
OMe
4e
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -10 -30 -50 -70 -90 -110 -130 -150 -170 -190 -210
f1 (ppm)

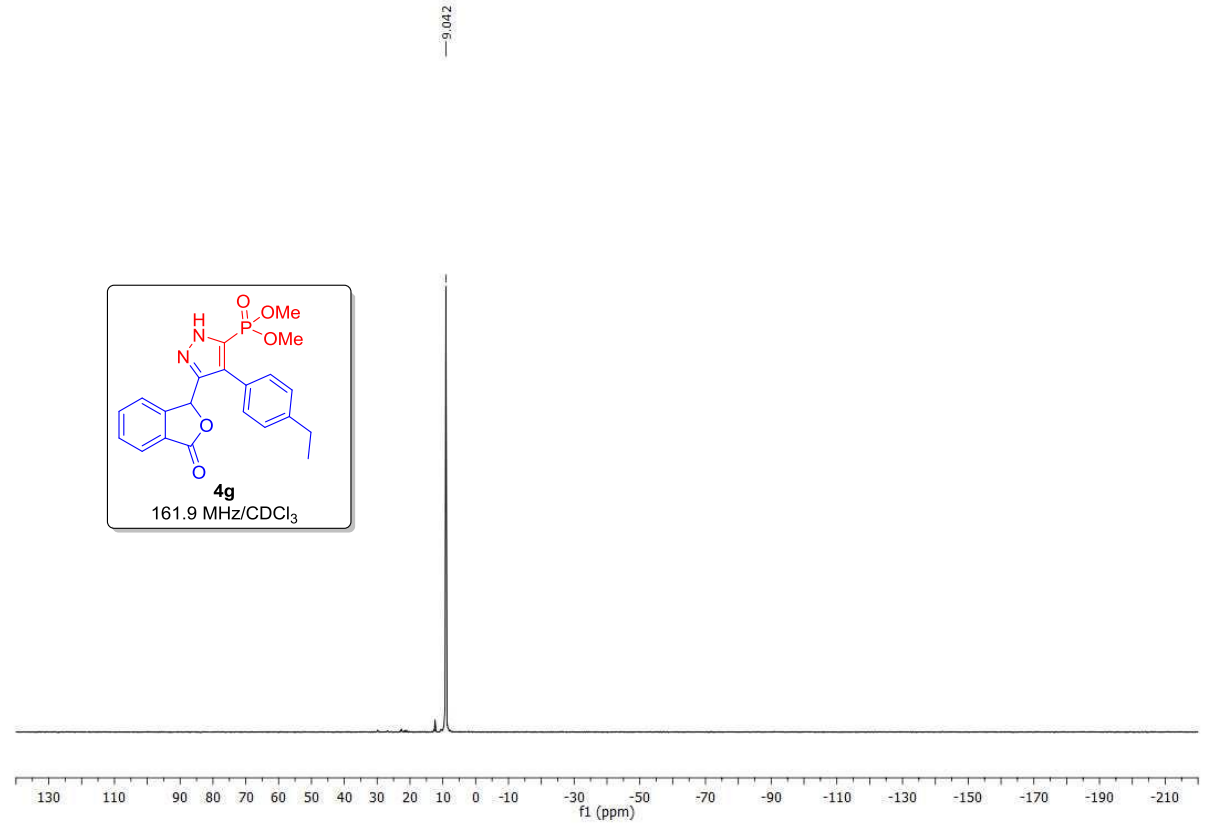

12.720
7.727
7.708
7.578
7.576
7.560
7.558
7.541
7.539
7.441
7.422
7.404
7.276
7.274
7.182
7.161
7.135
7.114
6.495
3.656
3.627
3.577
3.548
2.915
2.898
2.881
2.863
2.846
1.242
1.224
4f
400 MHz/CDCl3
0.99
0.99
1.03
1.03
1.36
1.88
1.97
0.95
6.00
1.04
6.58
f1 (ppm)

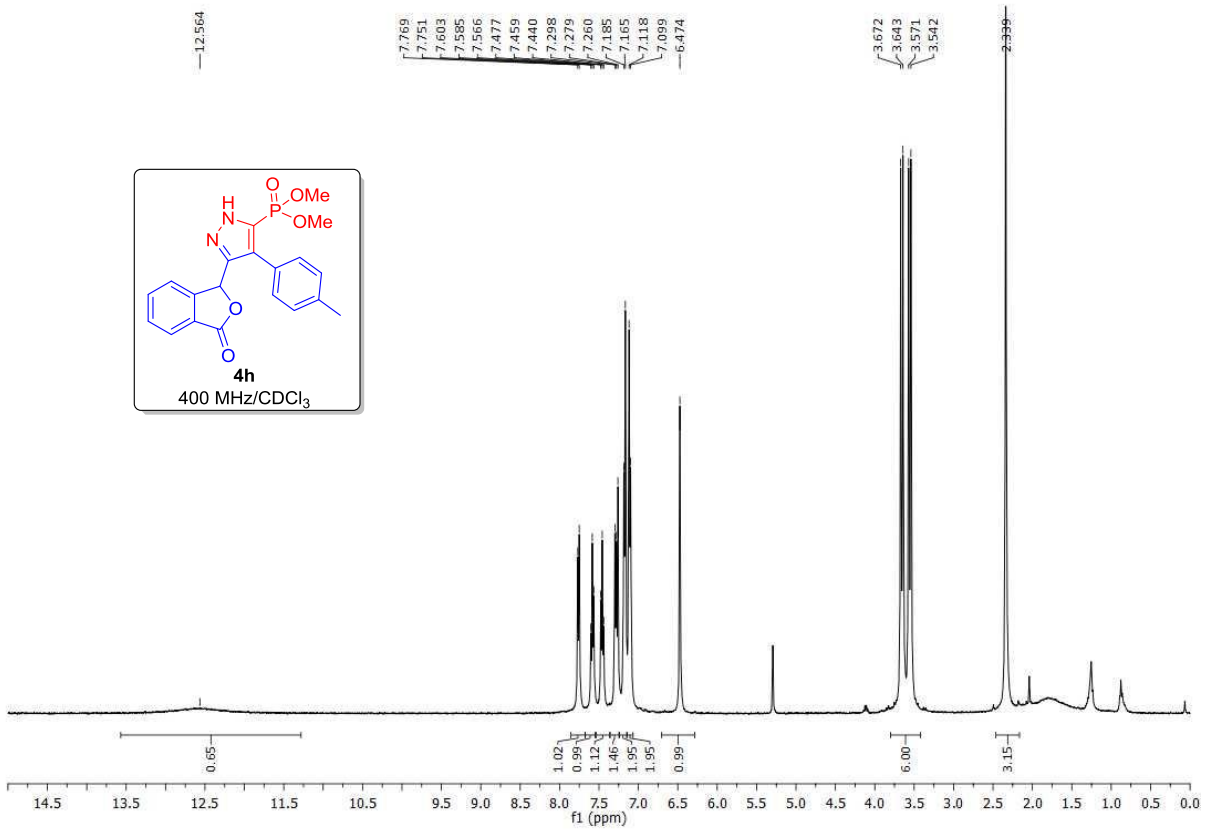

170.329
148.783
148.058
134.016
129.943
129.239
128.717
128.537
127.104
126.409
126.267
125.349
123.132
76.000
53.444
53.393
33.924
24.026
23.953
4f
100 MHz/CDCl3
f1 (ppm)

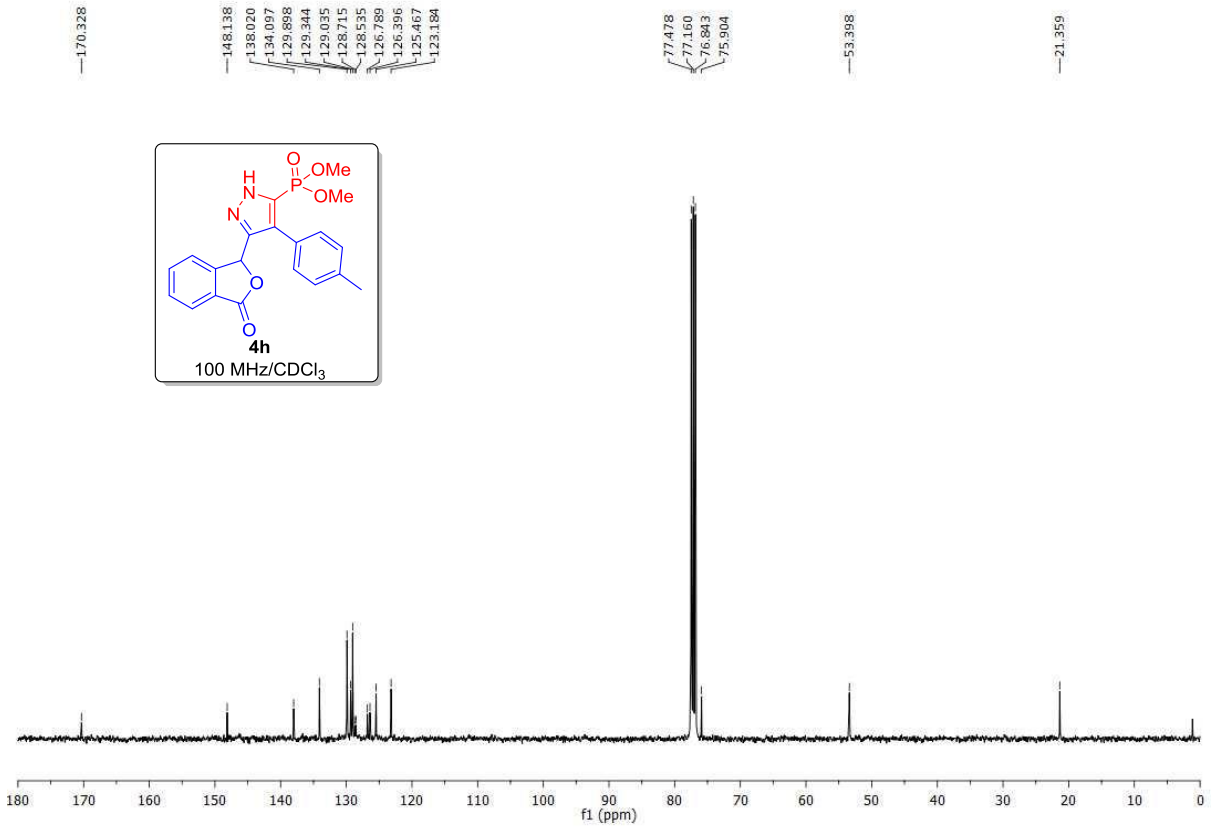

8.852
4f
161.9 MHz/CDCl3
f1 (ppm)

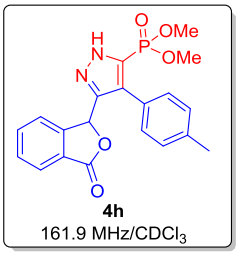

12.846
7.737
7.718
7.582
7.564
7.545
7.449
7.430
7.411
7.281
7.260
7.182
7.162
7.119
7.099
6.486
3.656
3.627
3.568
3.539
2.654
2.636
2.617
2.598
1.240
1.221
1.202
4g
400 MHz/CDCl3
1.00
1.06
0.99
1.06
1.81
1.74
0.93
6.01
2.11
3.31
f1 (ppm)


170.343
148.086
144.225
134.045
129.950
129.264
128.699
128.516
127.741
127.013
126.367
125.366
123.144
75.947
53.381
28.679
15.582
OMe
OMe
4g
100 MHz/CDCl3
f1 (ppm)

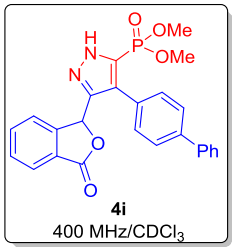

9.042
OMe
OMe
4g
161.9 MHz/CDCl3
f1 (ppm)


12.564
7.769
7.751
7.603
7.585
7.566
7.477
7.459
7.440
7.298
7.279
7.260
7.185
7.165
7.118
7.099
6.474
3.672
3.643
3.571
3.542
2.339
OMe
OMe
4h
400 MHz/CDCl3
0.65
1.02
0.99
1.12
1.46
1.95
1.95
0.99
6.00
3.15
f1 (ppm)

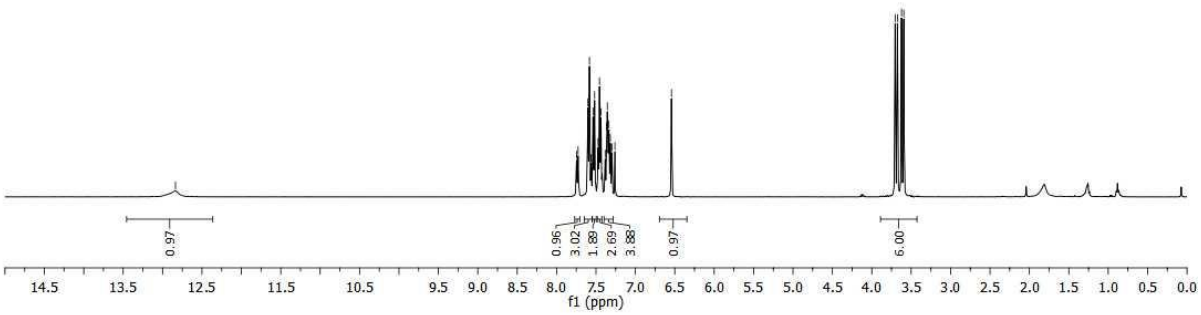

170.328
148.138
138.020
134.097
129.898
129.344
129.035
128.715
128.535
126.789
126.396
125.467
123.184
77.478
77.160
76.843
75.904
53.398
21.359
OMe
OMe
4h
100 MHz/CDCl3
f1 (ppm)

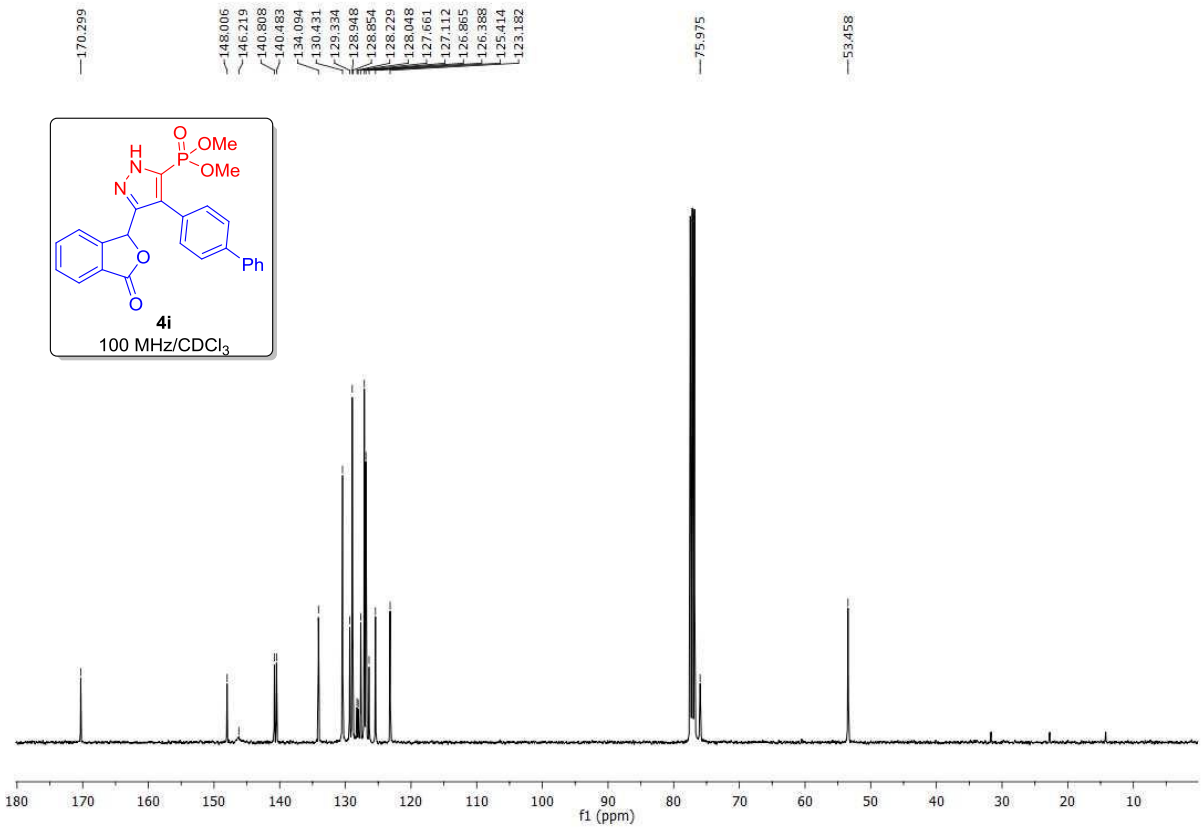


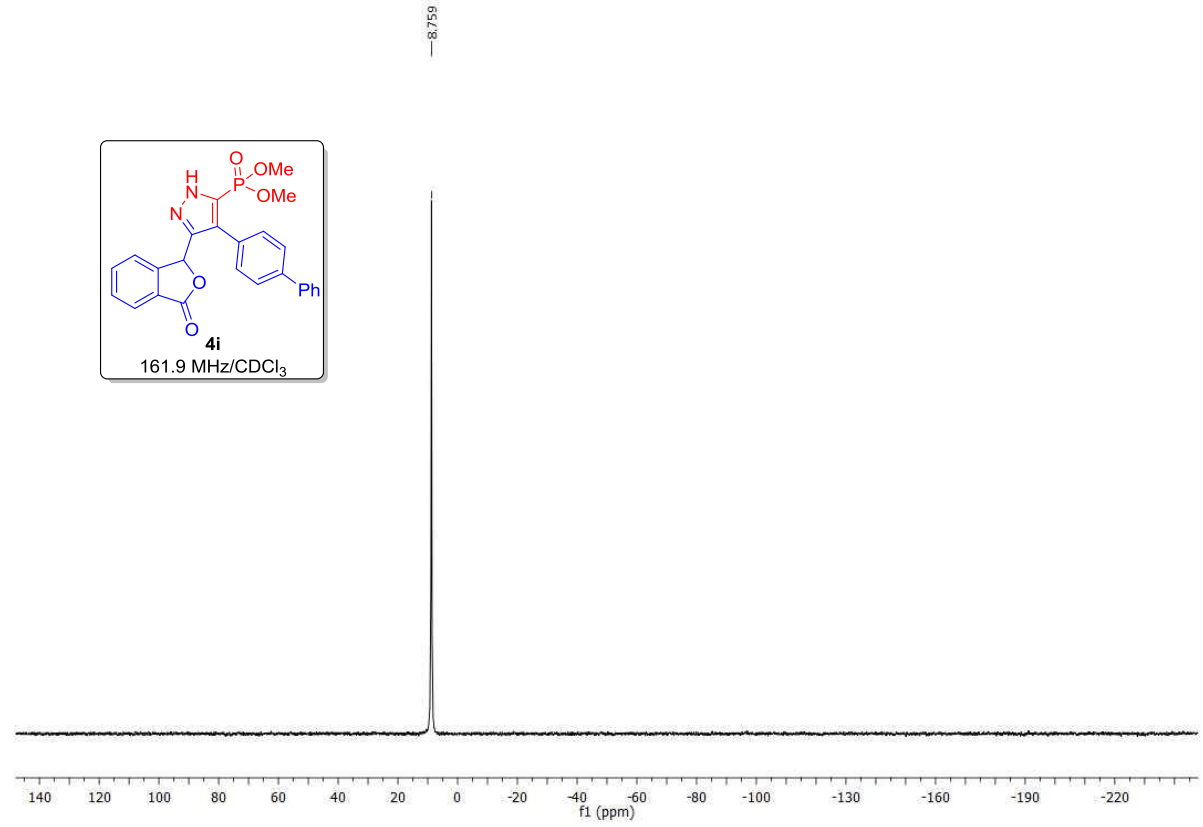
4h
161.9 MHz/$CDCl_3$

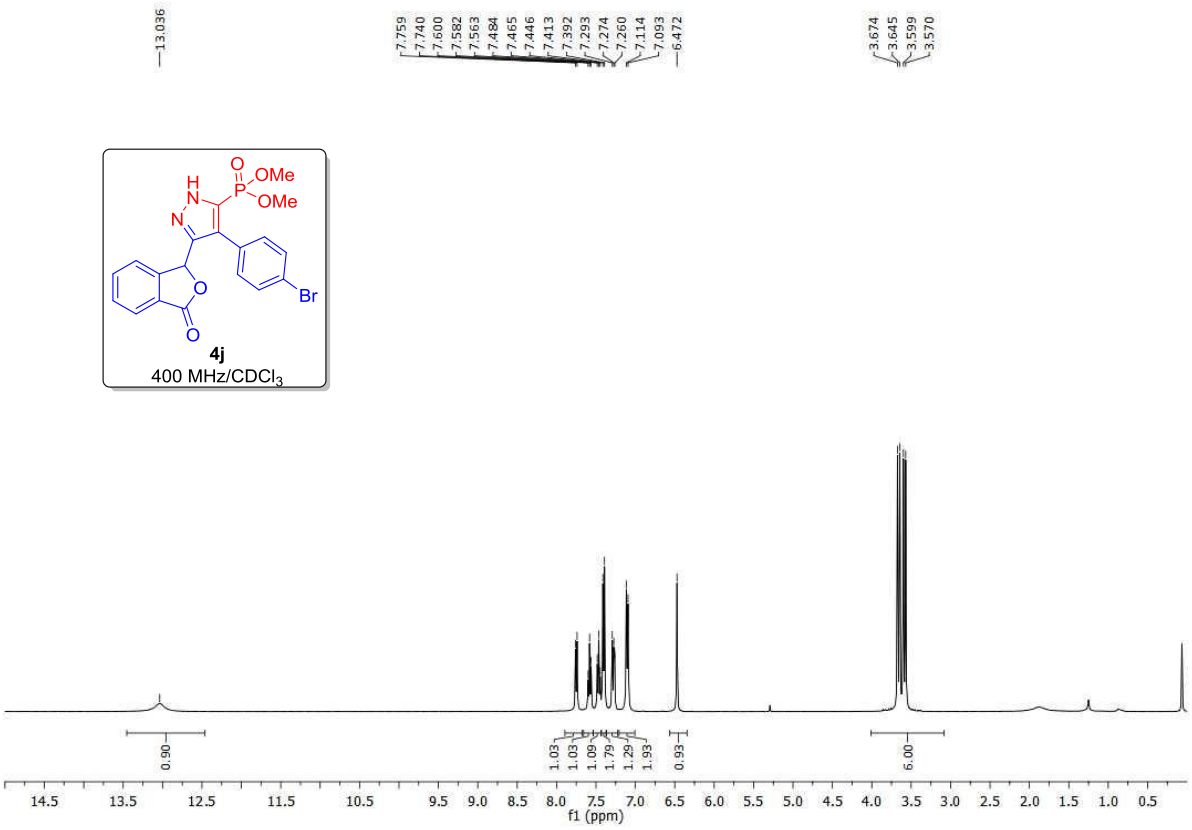


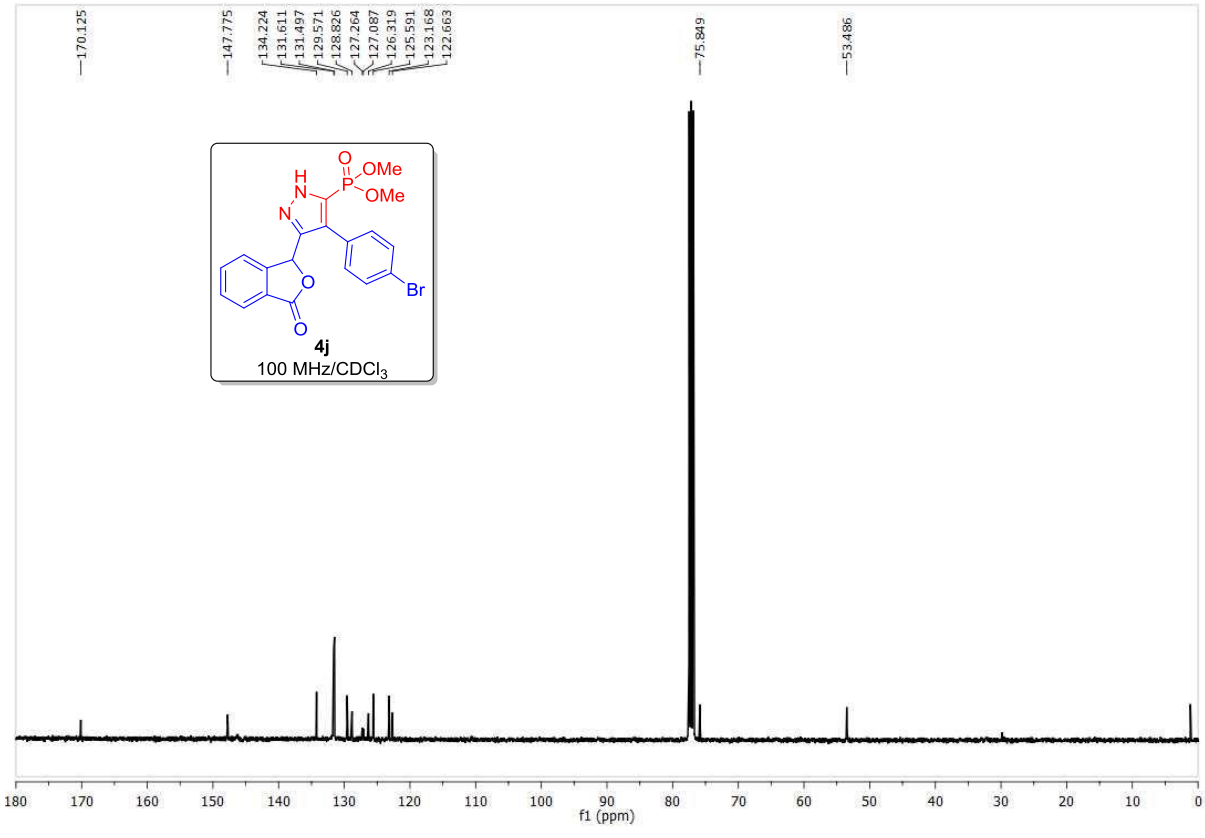
4i
400 MHz/$CDCl_3$

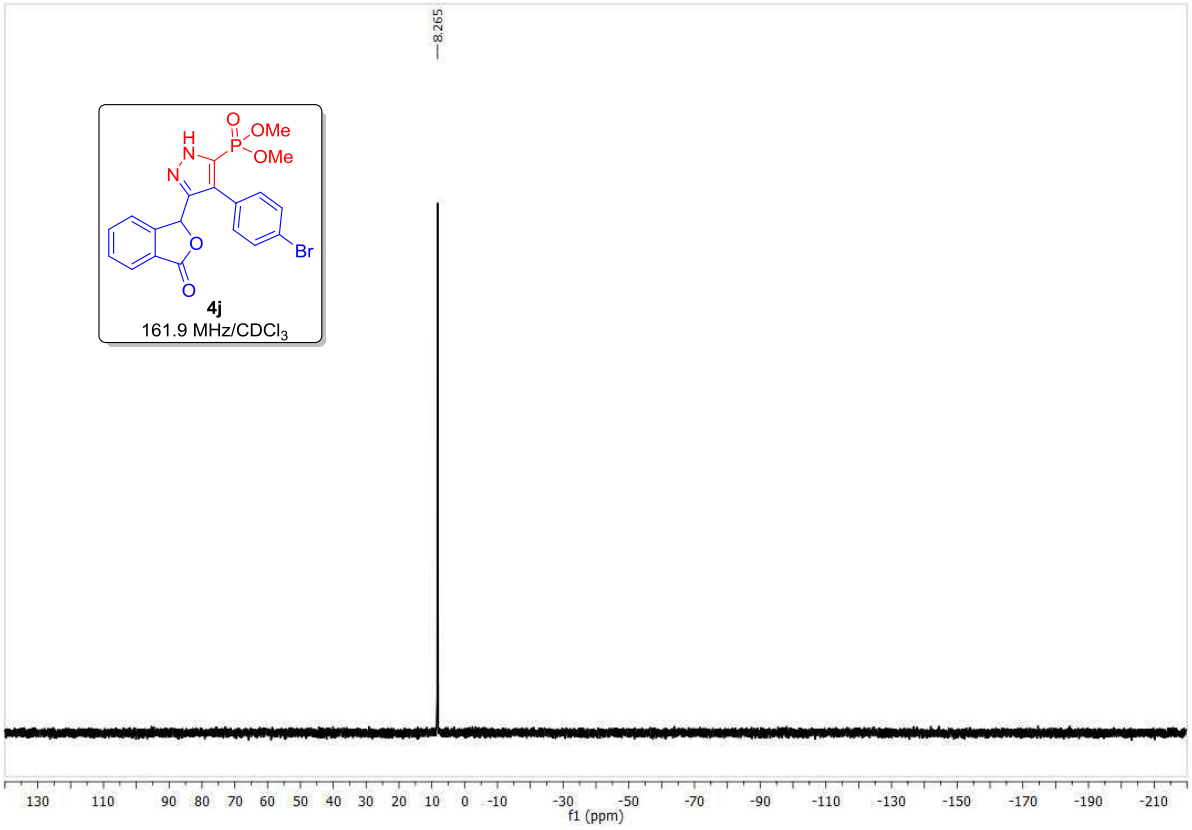


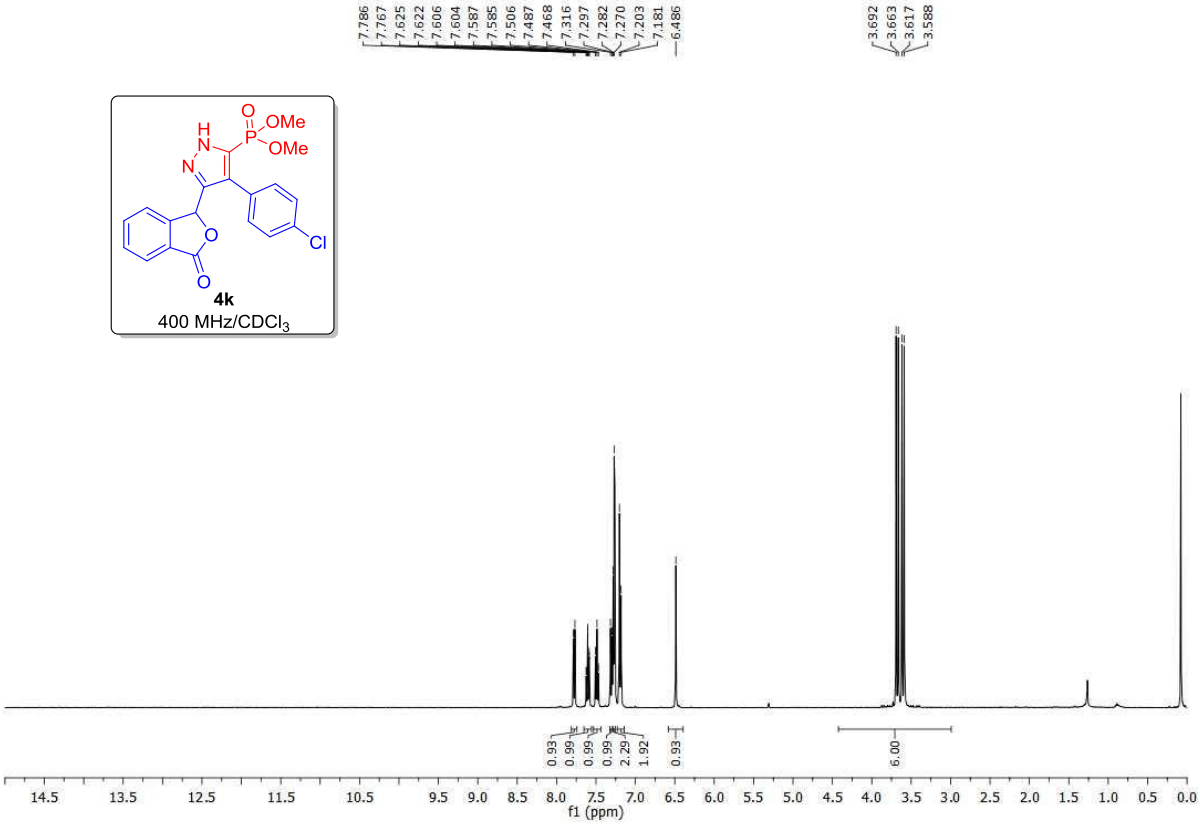

170.299
148.006
146.219
140.808
140.483
134.094
130.431
129.334
128.948
128.854
128.229
128.048
127.661
127.112
126.865
126.388
125.414
123.182
75.975
53.458
OMe
OMe
Ph
4i
100 MHz/CDCl3
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)

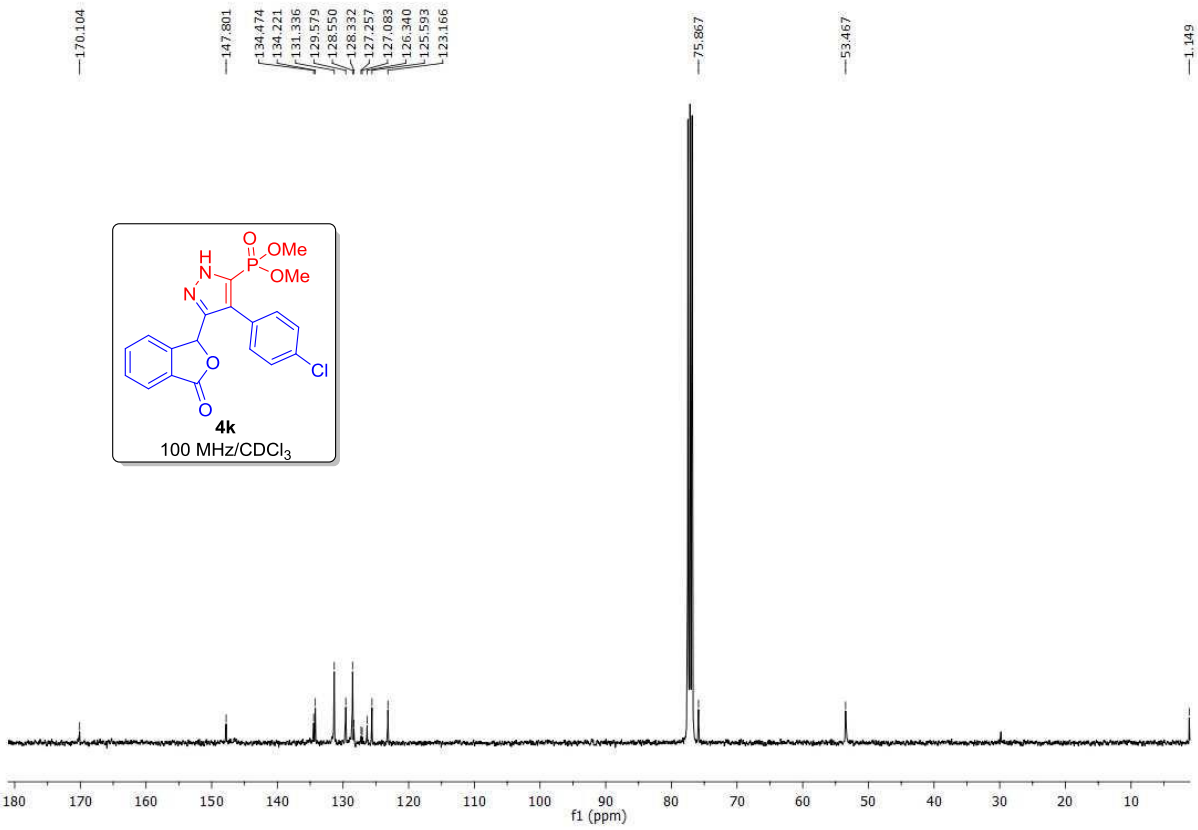

8.759
OMe
OMe
Ph
4i
161.9 MHz/CDCl3
140
120
100
80
60
40
20
0
-20
-40
-60
-80
-100
-130
-160
-190
-220
f1 (ppm)

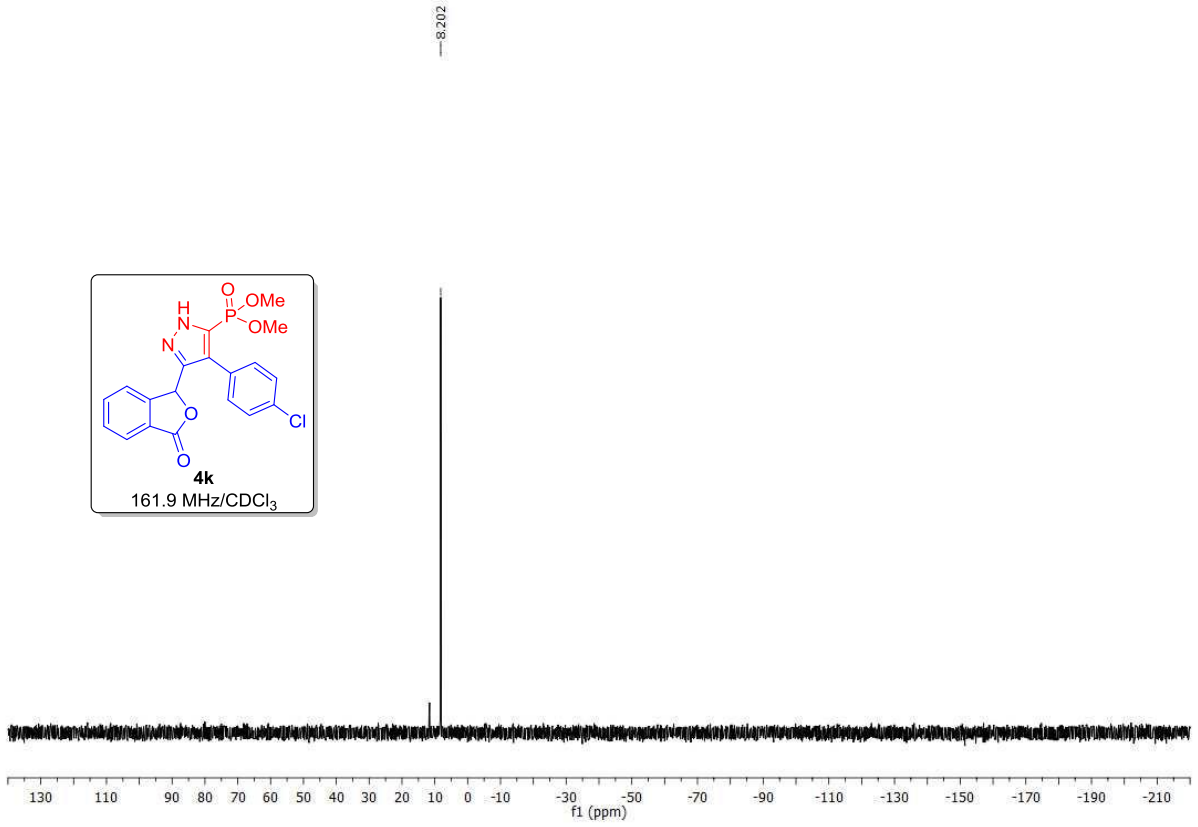

13.036
7.759
7.740
7.600
7.582
7.563
7.484
7.465
7.446
7.413
7.392
7.293
7.274
7.260
7.114
7.093
6.472
3.674
3.645
3.599
3.570
OMe
OMe
Br
4j
400 MHz/CDCl3
0.90
1.03
1.03
1.09
1.79
1.29
1.93
0.93
6.00
f1 (ppm)

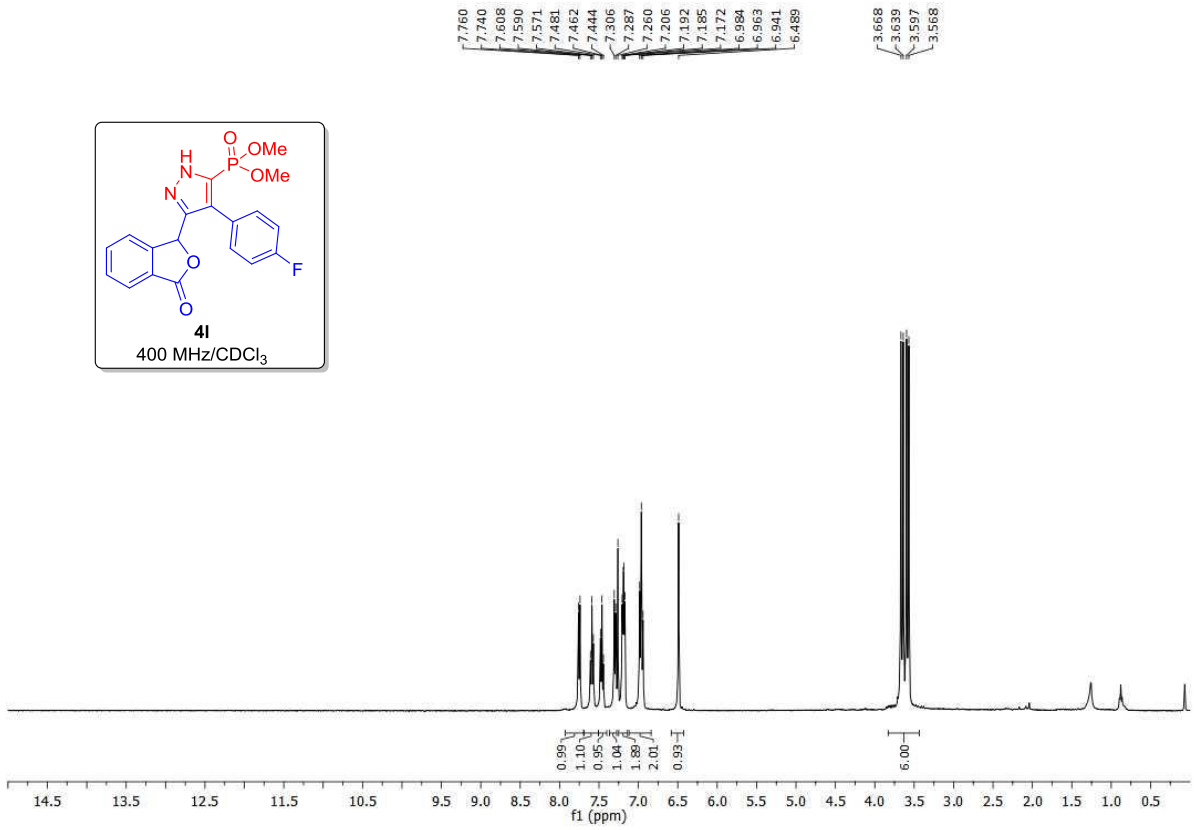

170.125
147.775
134.224
131.611
131.497
129.571
128.826
127.264
127.087
126.319
125.591
123.168
122.663
75.849
53.486
OMe
OMe
Br
4j
100 MHz/CDCl3
f1 (ppm)

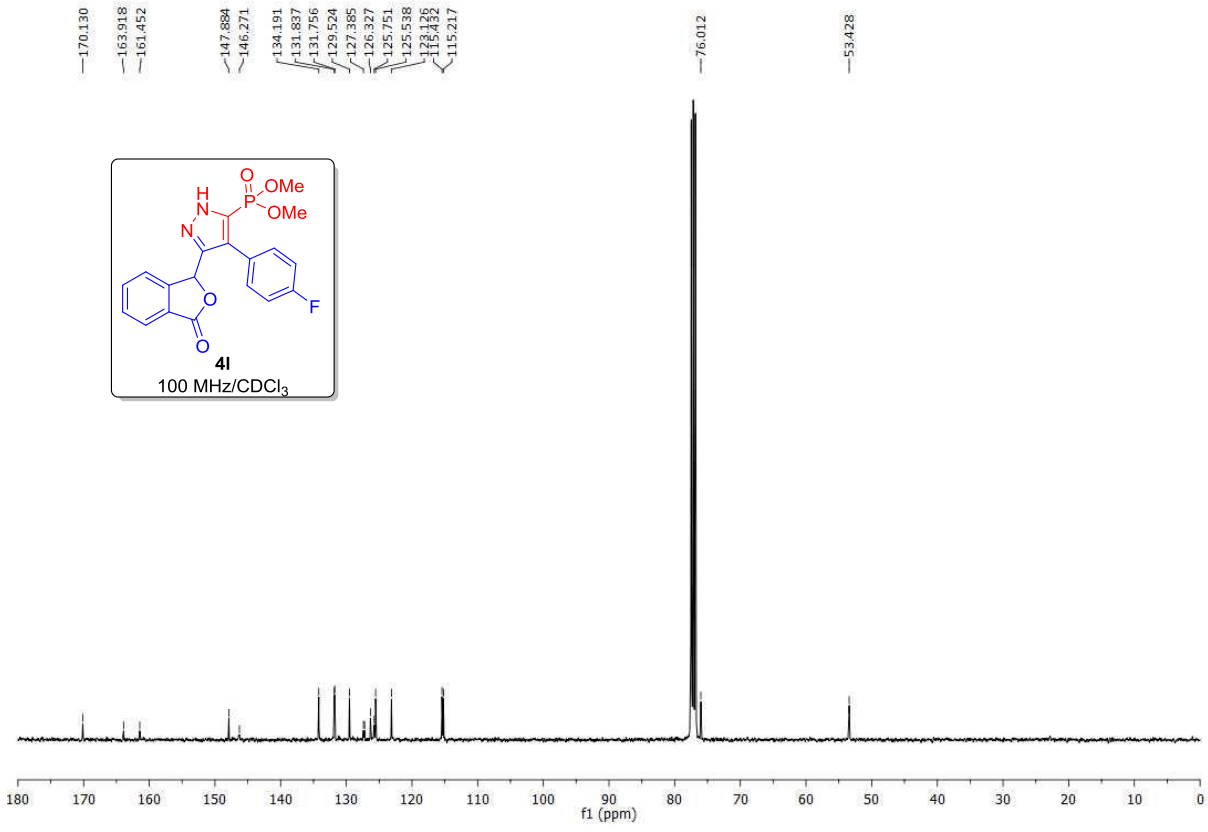

8.265
OMe
OMe
Br
4j
161.9 MHz/CDCl3
f1 (ppm)

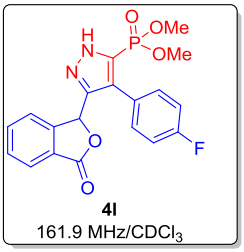

7.786
7.767
7.625
7.622
7.606
7.604
7.587
7.585
7.506
7.487
7.468
7.316
7.297
7.282
7.270
7.203
7.181
6.486
3.692
3.663
3.617
3.588
OMe
OMe
Cl
4k
400 MHz/CDCl3
0.93
0.99
0.99
0.99
2.29
1.92
0.93
6.00
f1 (ppm)


170.104
147.801
134.474
134.221
131.336
129.579
128.550
128.332
127.257
127.083
126.340
125.593
123.166
75.867
53.467
1.149
OMe
OMe
Cl
4k
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


8.202
OMe
OMe
Cl
4k
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -10 -30 -50 -70 -90 -110 -130 -150 -170 -190 -210
f1 (ppm)

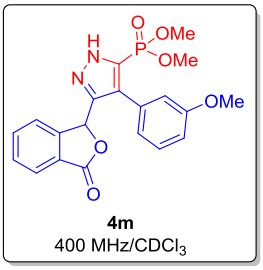

7.760
7.740
7.608
7.590
7.571
7.481
7.462
7.444
7.306
7.287
7.260
7.206
7.192
7.185
7.172
6.984
6.963
6.941
6.489
3.668
3.639
3.597
3.568
OMe
OMe
H
N
N
P
O
O
O
F
4l
400 MHz/CDCl3
0.99
1.10
0.95
1.04
1.88
2.01
0.93
6.00
f1 (ppm)


170.130
163.918
161.452
147.884
146.271
134.191
131.837
131.756
129.524
127.385
126.327
125.751
125.538
123.126
115.432
115.217
76.012
53.428
OMe
OMe
H
N
N
P
O
O
O
F
4l
100 MHz/CDCl3
f1 (ppm)



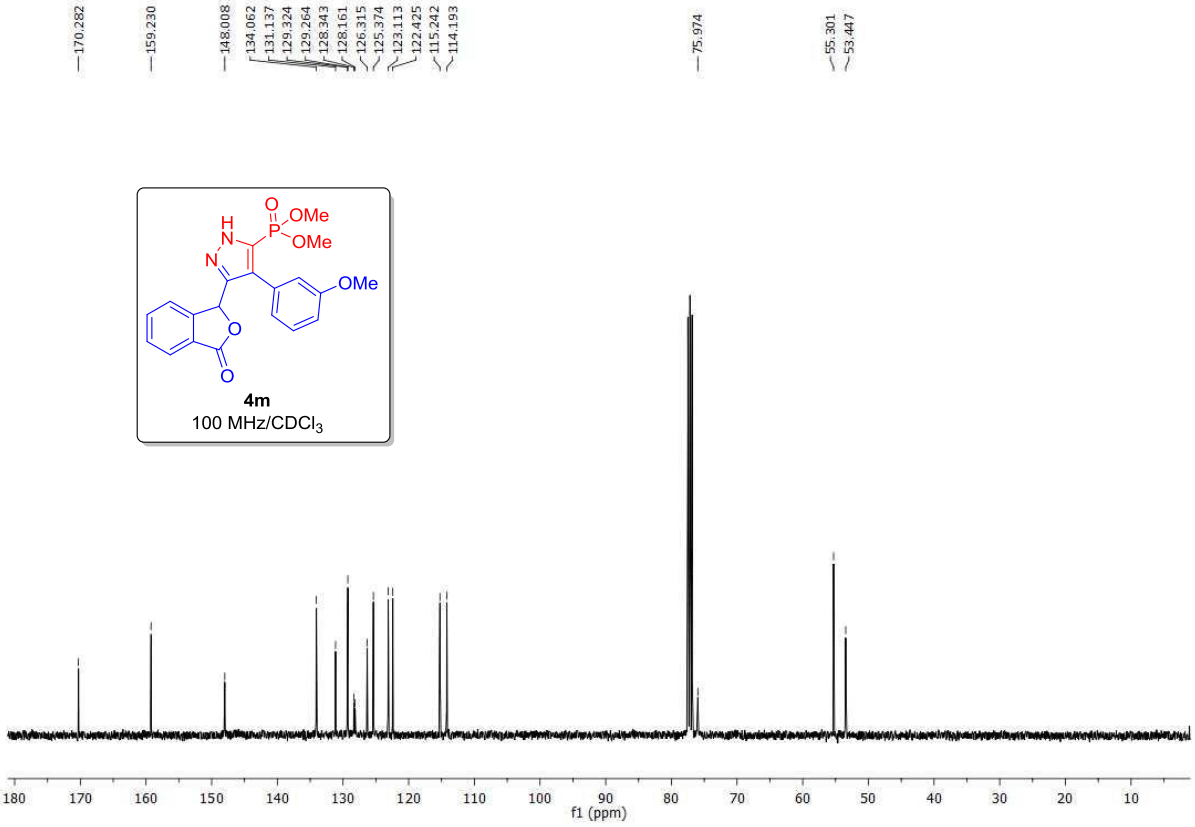
4l

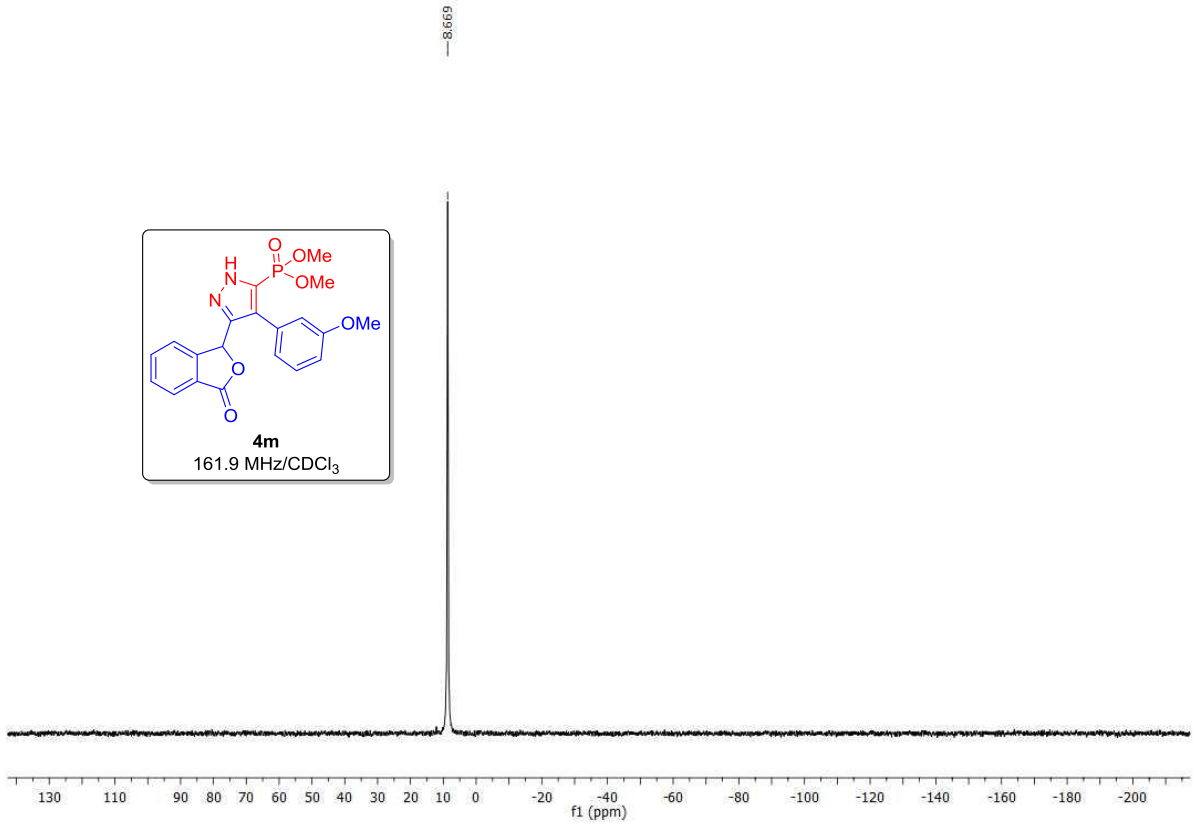
161.9 MHz/$CDCl_3$

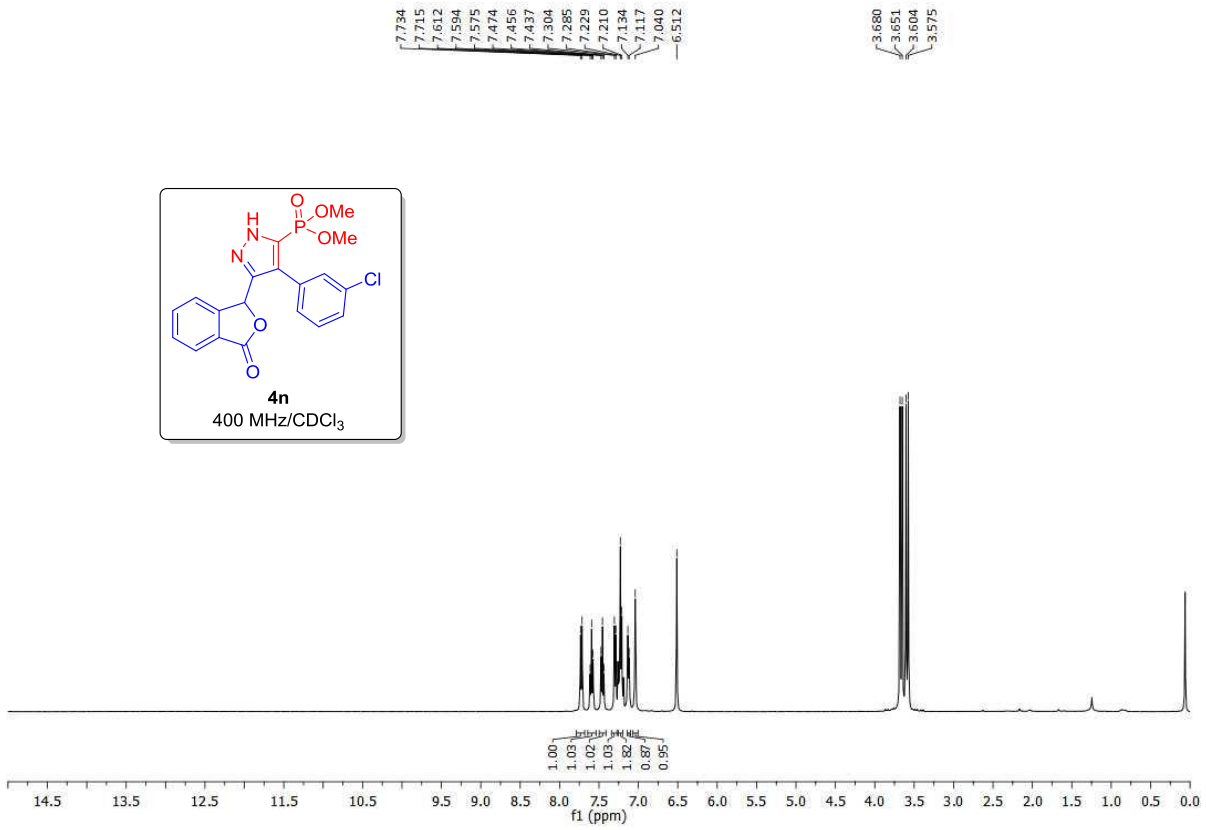


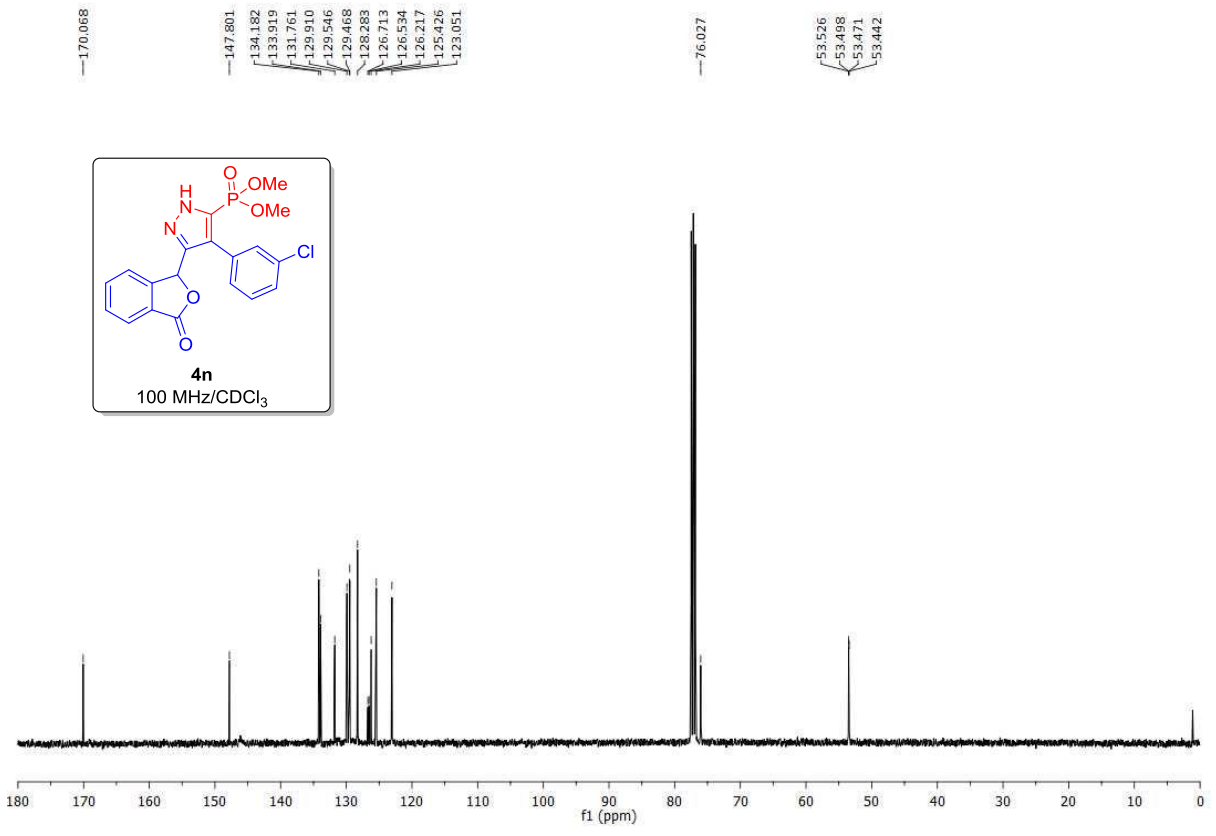
4m

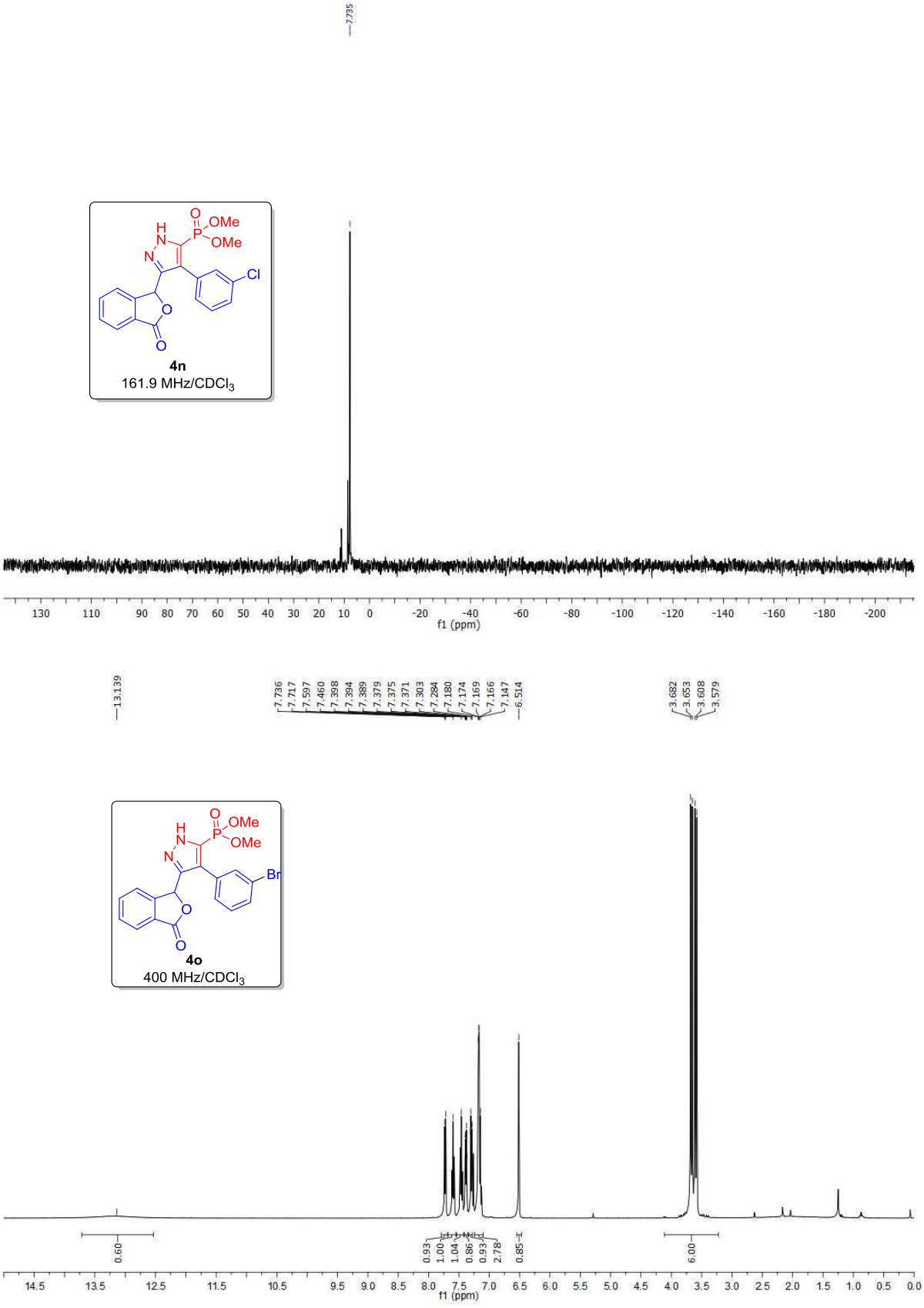
400 MHz/$CDCl_3$

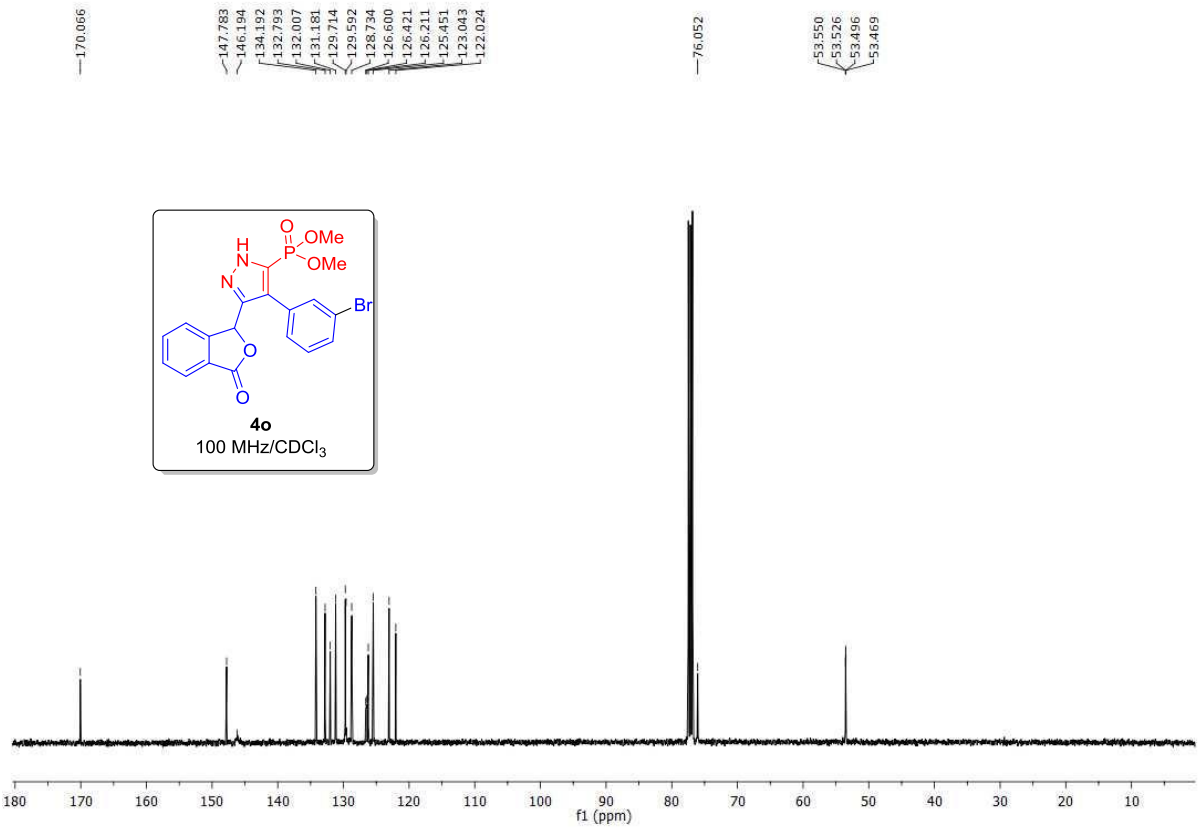

170.282
159.230
148.008
134.062
131.137
129.324
129.264
128.343
128.161
126.315
125.374
123.113
122.425
115.242
114.193
75.974
55.301
53.447
OMe
OMe
OMe
4m
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

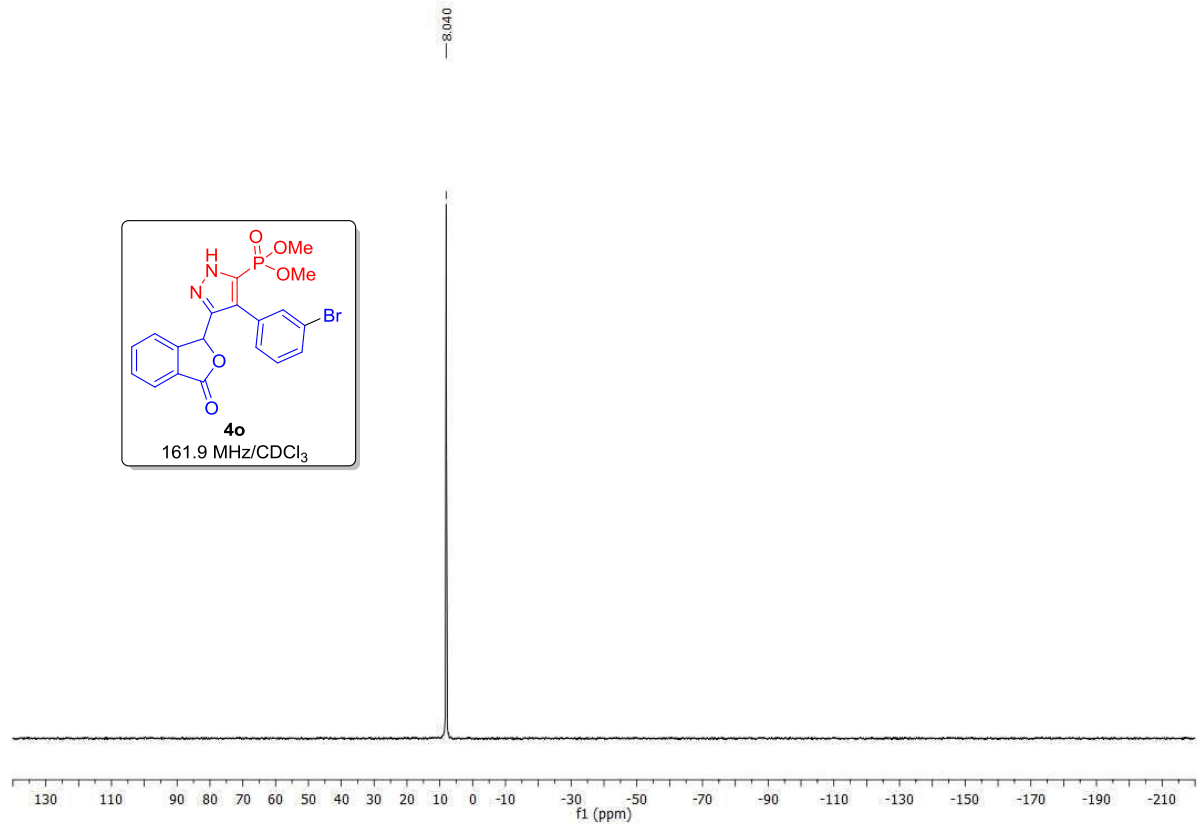

8.669
OMe
OMe
OMe
4m
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

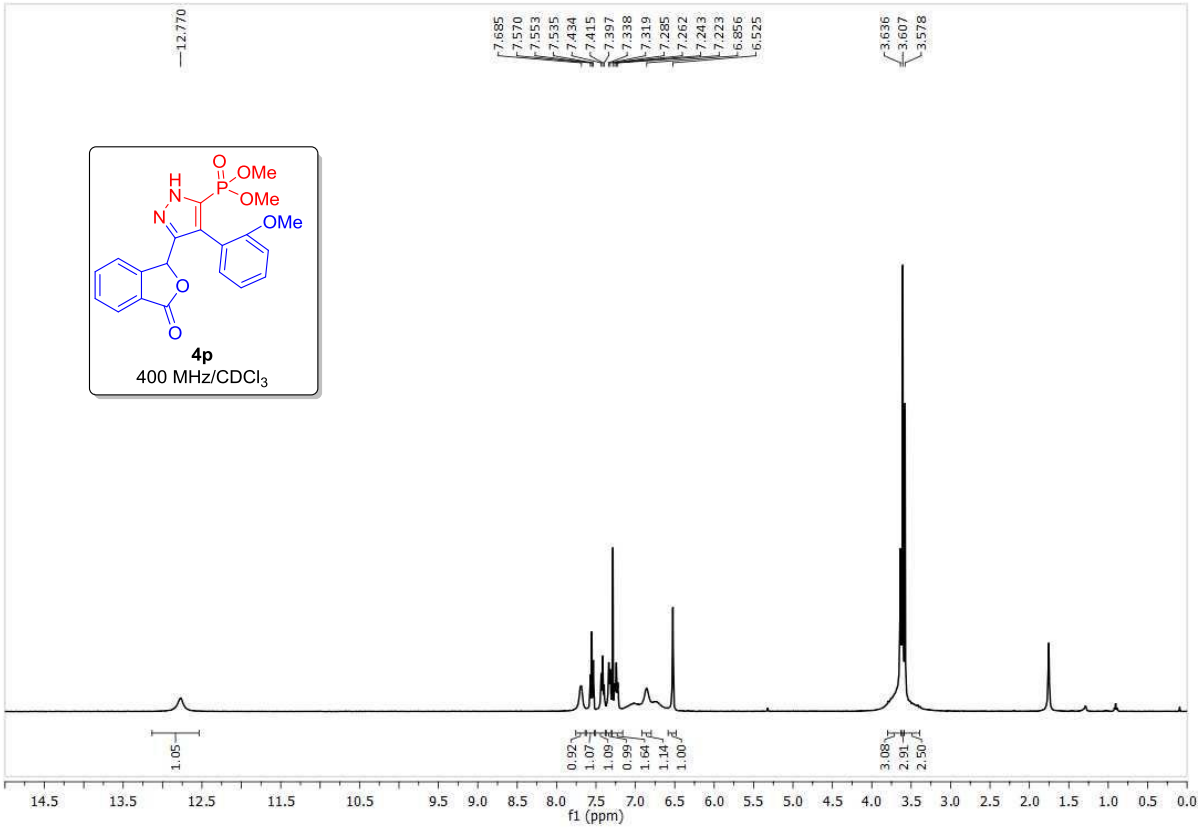

7.734
7.715
7.612
7.594
7.575
7.474
7.456
7.437
7.304
7.285
7.229
7.210
7.134
7.117
7.040
6.512
3.680
3.651
3.604
3.575
O
OMe
H
N
N
P
OMe
Cl
O
O
4n
400 MHz/CDCl3
1.00
1.03
1.02
1.03
1.82
0.87
0.95
14.5
13.5
12.5
11.5
10.5
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

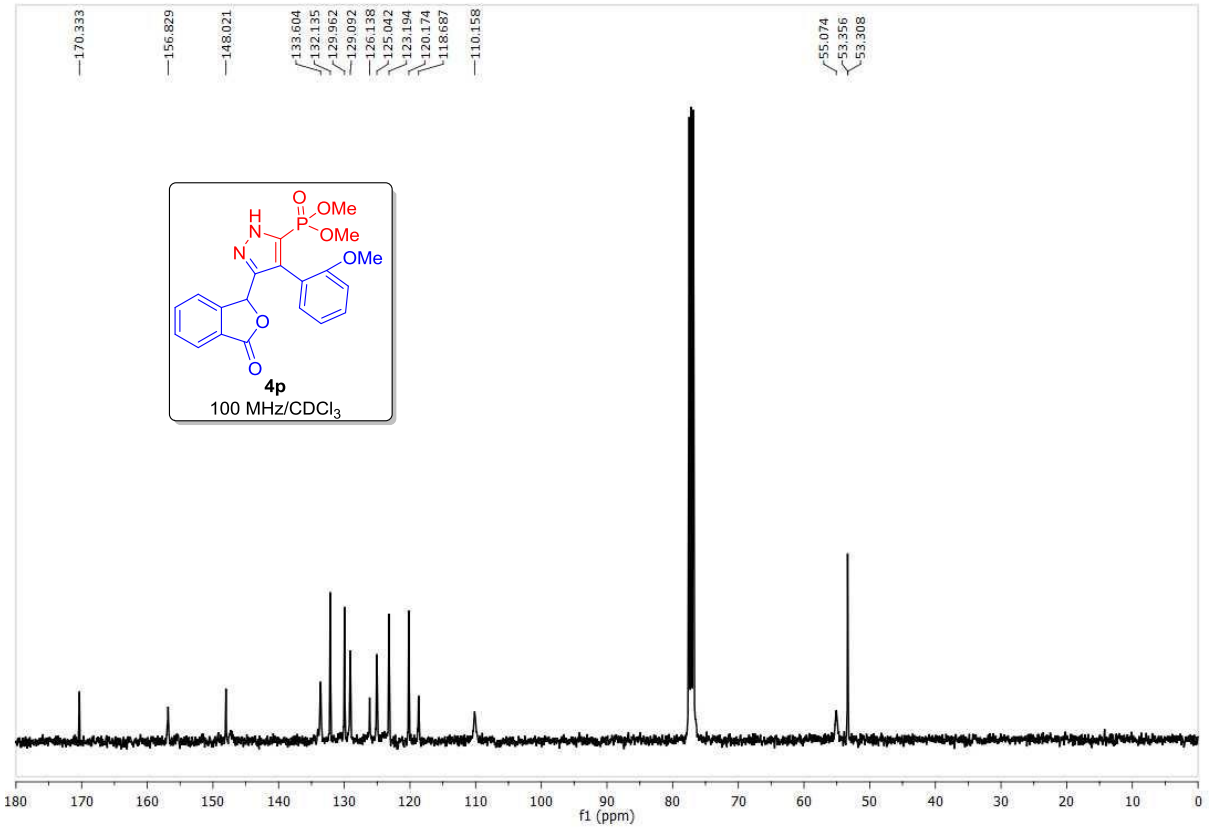

170.068
147.801
134.182
133.919
131.761
129.910
129.546
129.468
128.283
126.713
126.534
126.217
125.426
123.051
76.027
53.526
53.498
53.471
53.442
O
OMe
H
N
N
P
OMe
Cl
O
O
4n
100 MHz/CDCl3
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

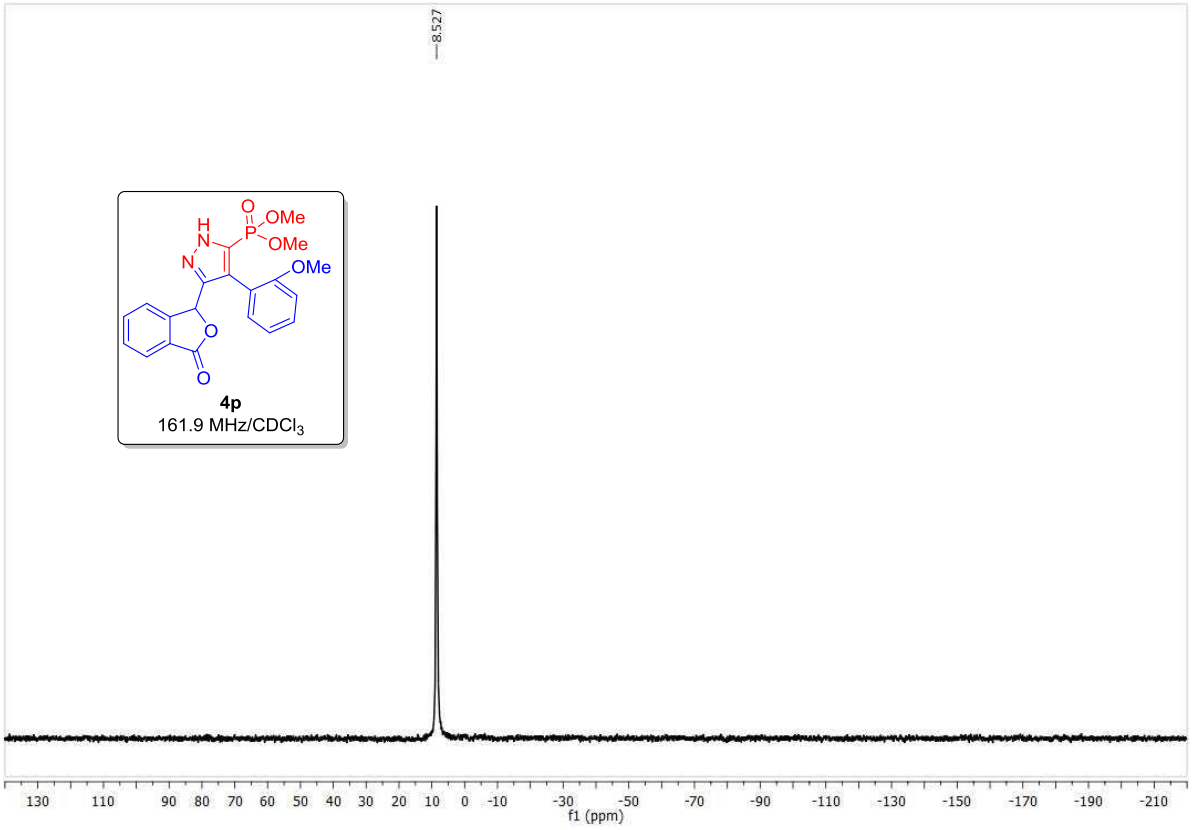

7.735
4n
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)
13.139
7.736 7.717 7.597 7.460 7.398 7.394 7.389 7.379 7.375 7.371 7.303 7.284 7.180 7.174 7.169 7.166 7.147 6.514
3.682 3.653 3.608 3.579
4o
400 MHz/CDCl3
0.60 0.93 1.00 1.04 0.86 0.93 2.78 0.85 6.00
14.5 13.5 12.5 11.5 10.5 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

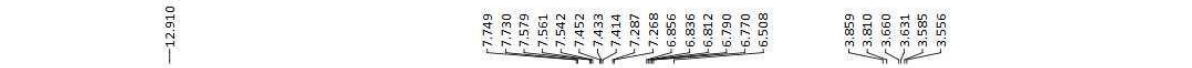

170.066
147.783
146.194
134.192
132.793
132.007
131.181
129.714
129.592
128.734
126.600
126.421
126.211
125.451
123.043
122.024
76.052
53.550
53.526
53.496
53.469
4o
100 MHz/CDCl3
f1 (ppm)

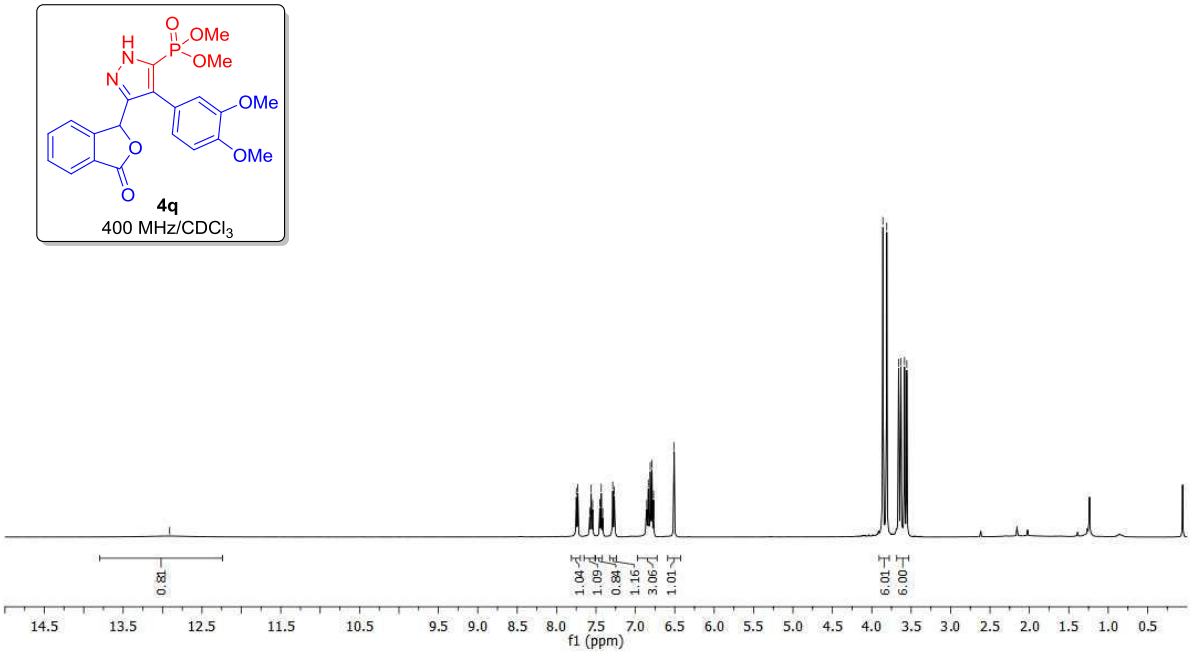

8.040
4o
161.9 MHz/CDCl3
f1 (ppm)

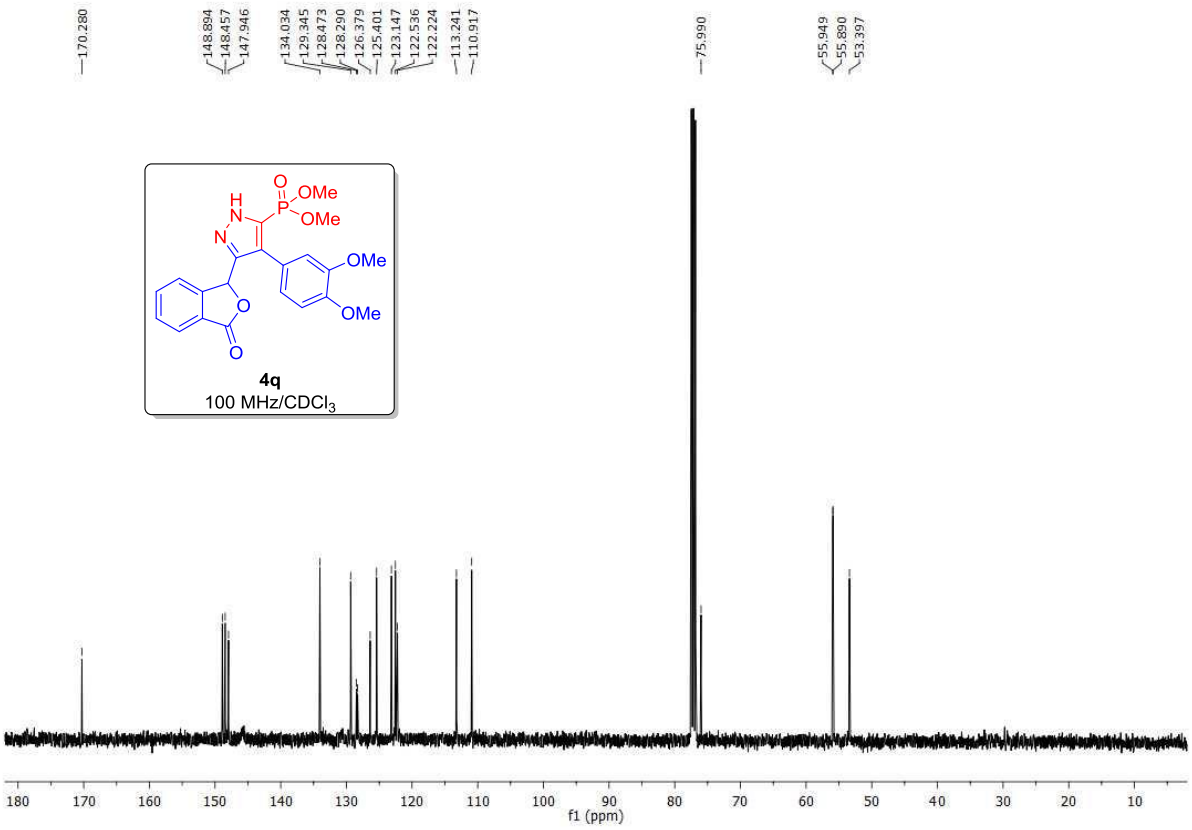

12.770
7.685
7.570
7.553
7.535
7.434
7.415
7.397
7.338
7.319
7.285
7.262
7.243
7.223
6.856
6.525
3.636
3.607
3.578
OMe
OMe
OMe
4p
400 MHz/CDCl3
1.05
0.92
1.07
1.09
0.99
1.64
1.14
1.00
3.08
2.91
2.50
f1 (ppm)

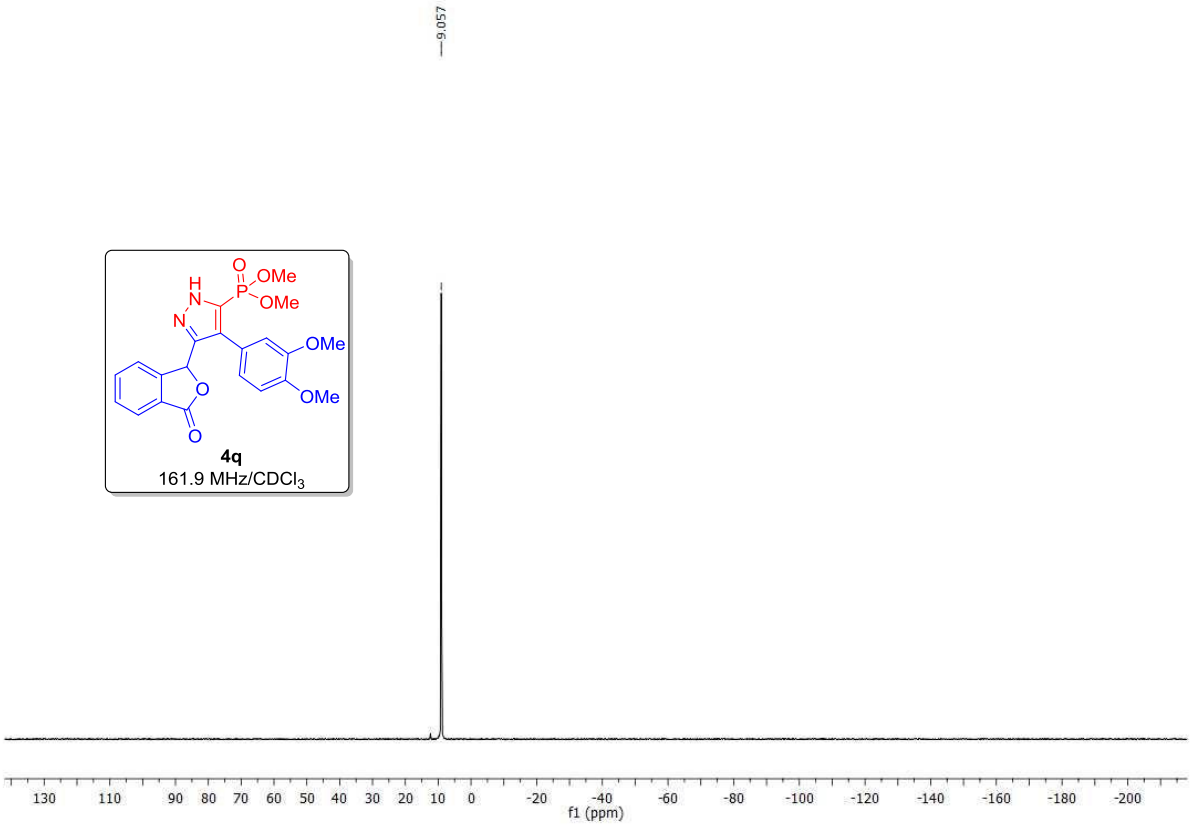

170.333
156.829
148.021
133.604
132.135
129.962
129.092
126.138
125.042
123.194
120.174
118.687
110.158
55.074
53.356
53.308
OMe
OMe
OMe
4p
100 MHz/CDCl3
f1 (ppm)

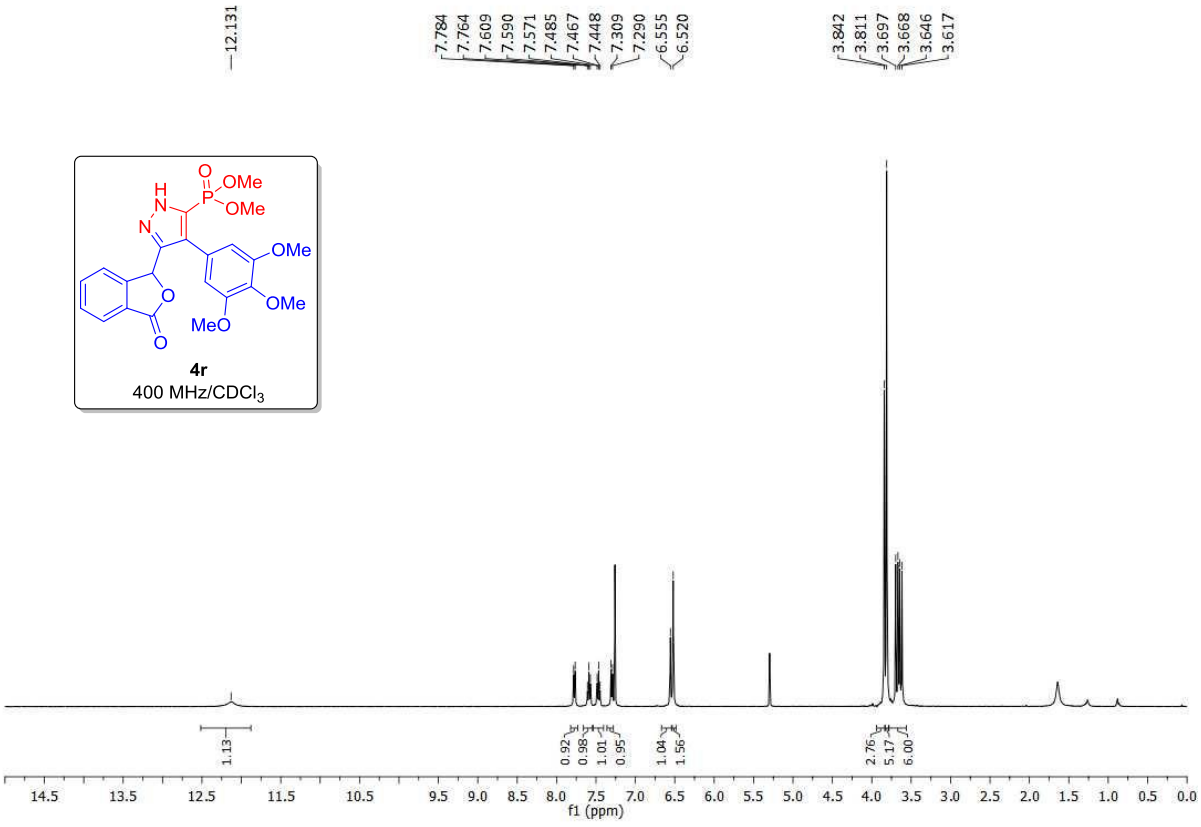

8.527
OMe
OMe
OMe
4p
161.9 MHz/CDCl3
f1 (ppm)

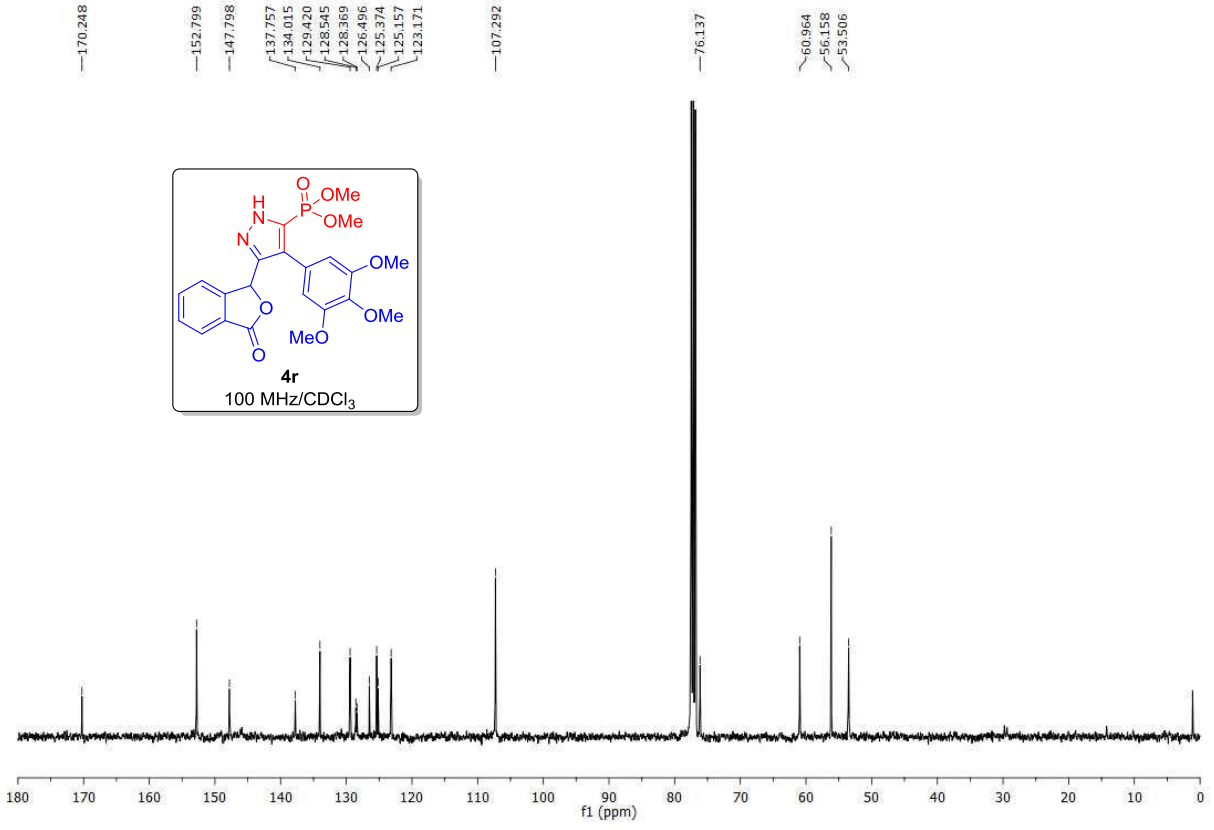

12.910
7.749
7.730
7.579
7.561
7.542
7.452
7.433
7.414
7.287
7.268
6.856
6.836
6.812
6.790
6.770
6.508
3.859
3.810
3.660
3.631
3.585
3.556

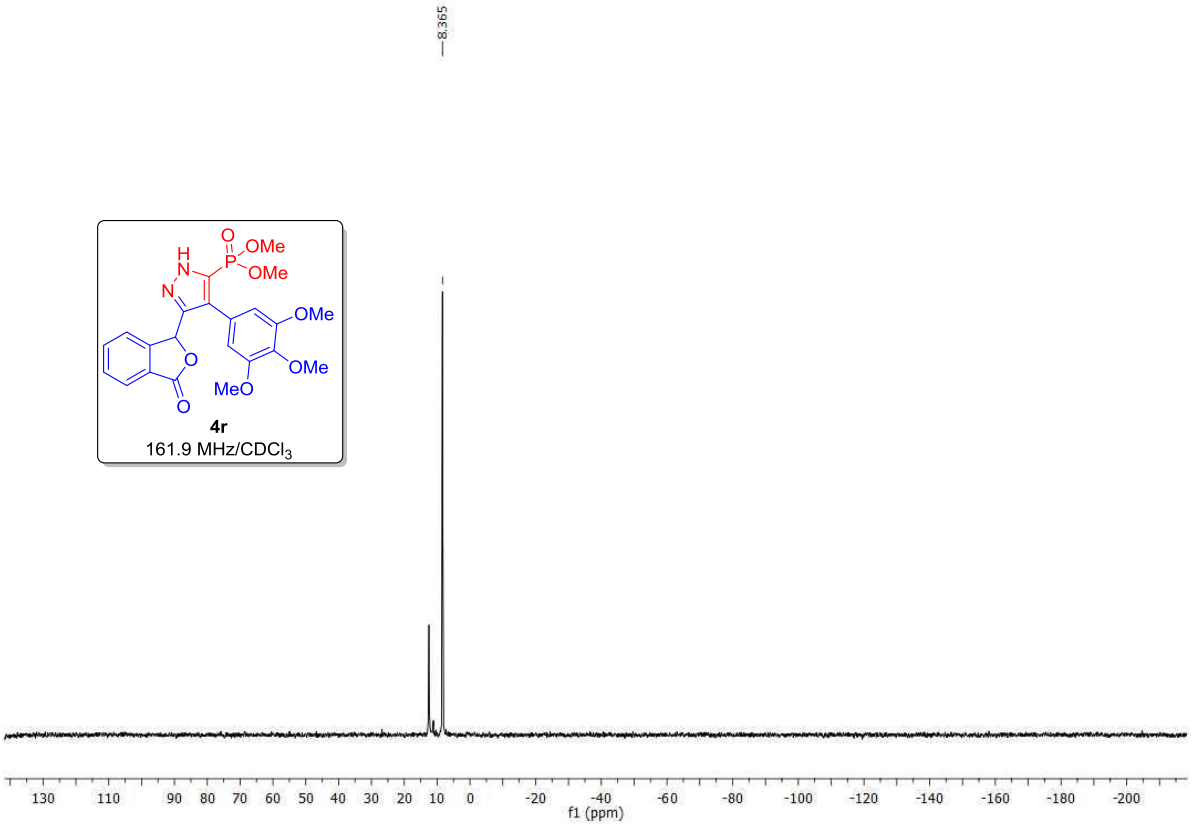

OMe
OMe
OMe
OMe
4q
400 MHz/CDCl3
0.81
1.04
1.09
0.84
1.16
3.06
1.01
6.01
6.00
f1 (ppm)

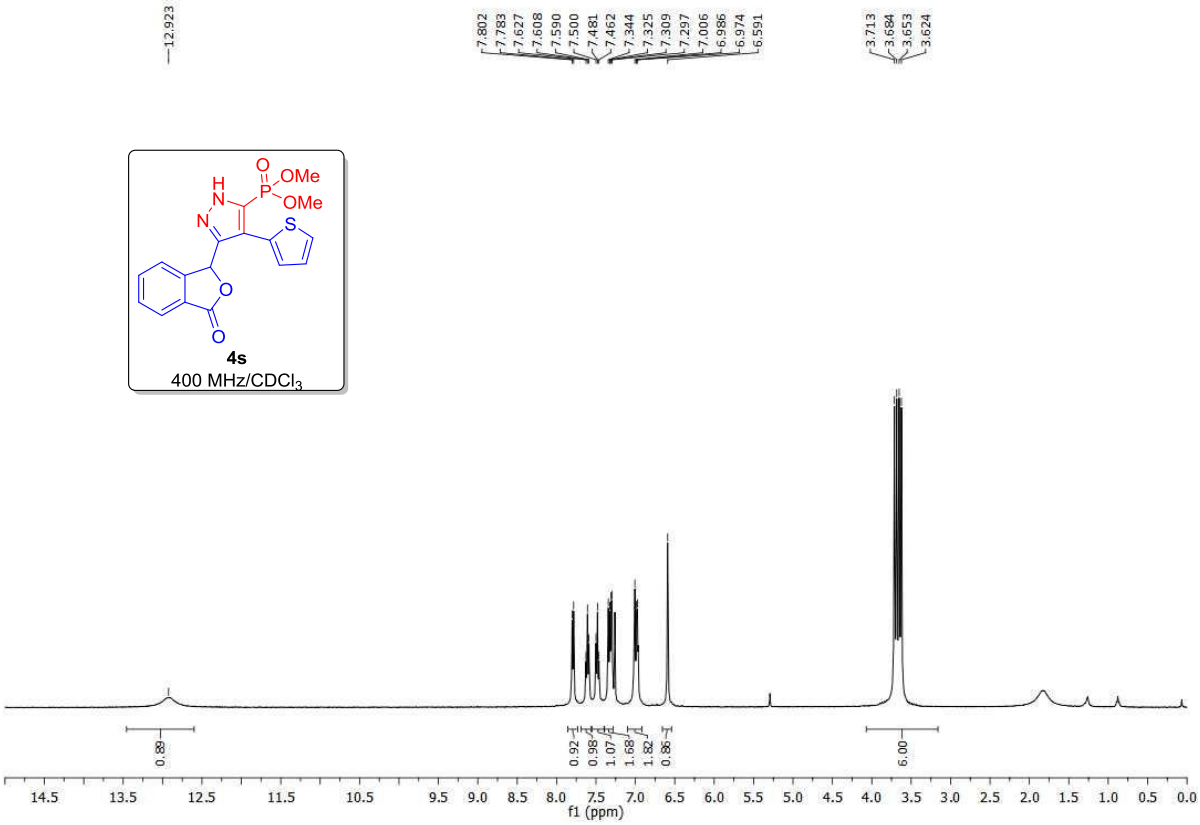

170.280
148.894
148.457
147.946
134.034
129.345
128.473
128.290
126.379
125.401
123.147
122.536
122.224
113.241
110.917
75.990
55.949
55.890
53.397
OMe
OMe
OMe
OMe
4q
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)

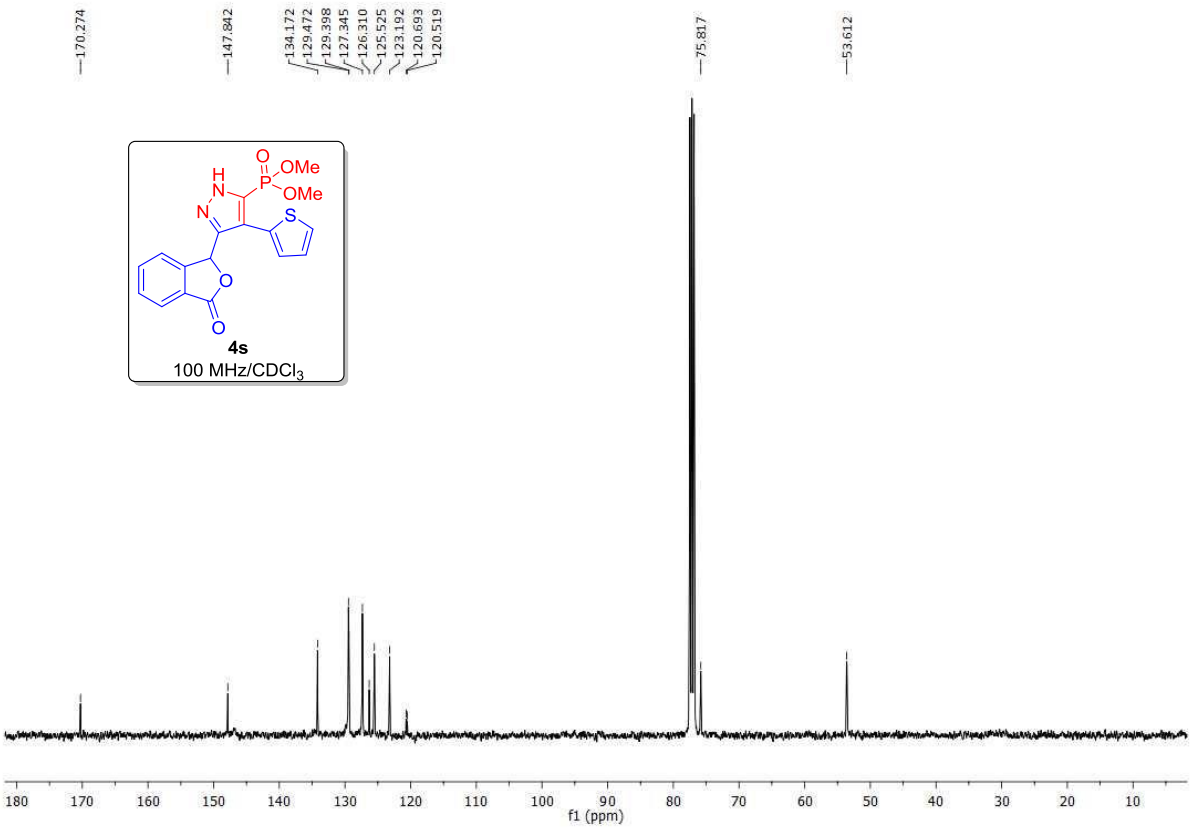

9.057
OMe
OMe
OMe
OMe
4q
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

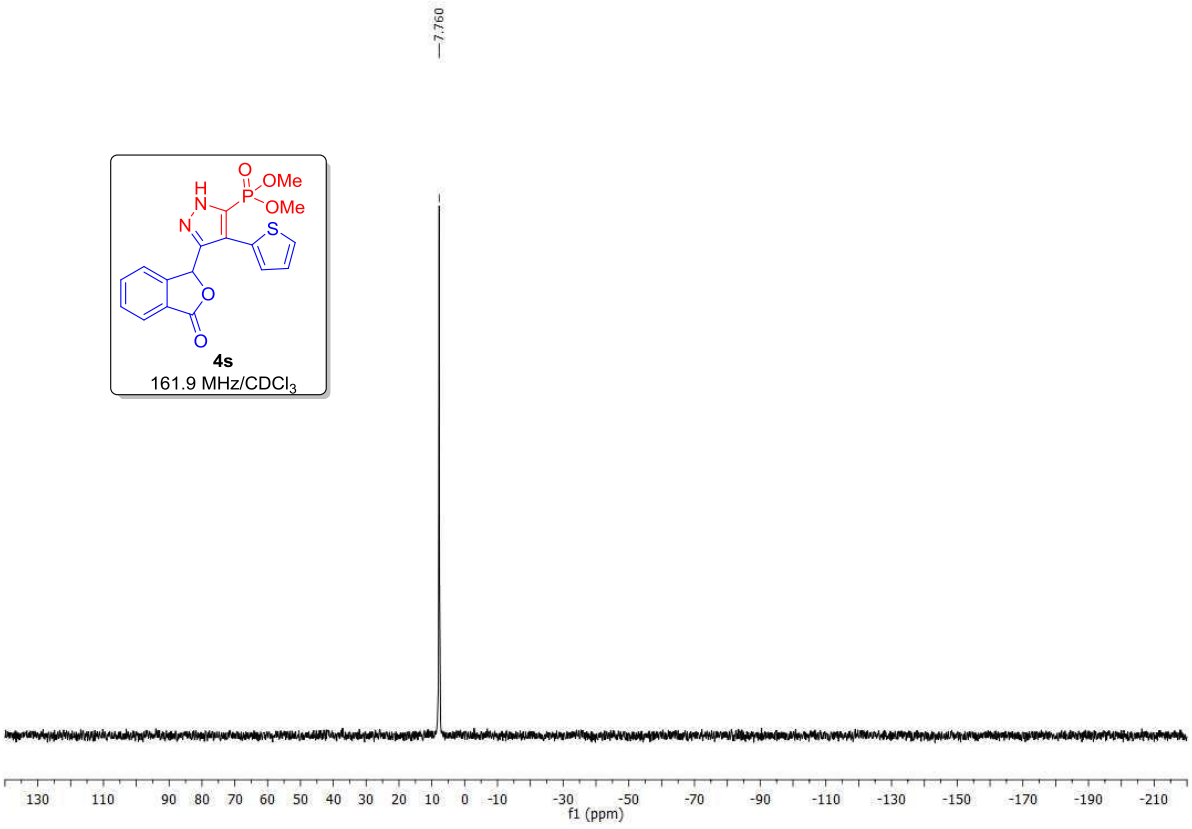

12.131
7.784
7.764
7.609
7.590
7.571
7.485
7.467
7.448
7.309
7.290
6.555
6.520
3.842
3.811
3.697
3.668
3.646
3.617
4r
400 MHz/CDCl3
1.13
0.92
0.98
1.01
0.95
1.04
1.56
2.76
5.17
6.00
f1 (ppm)

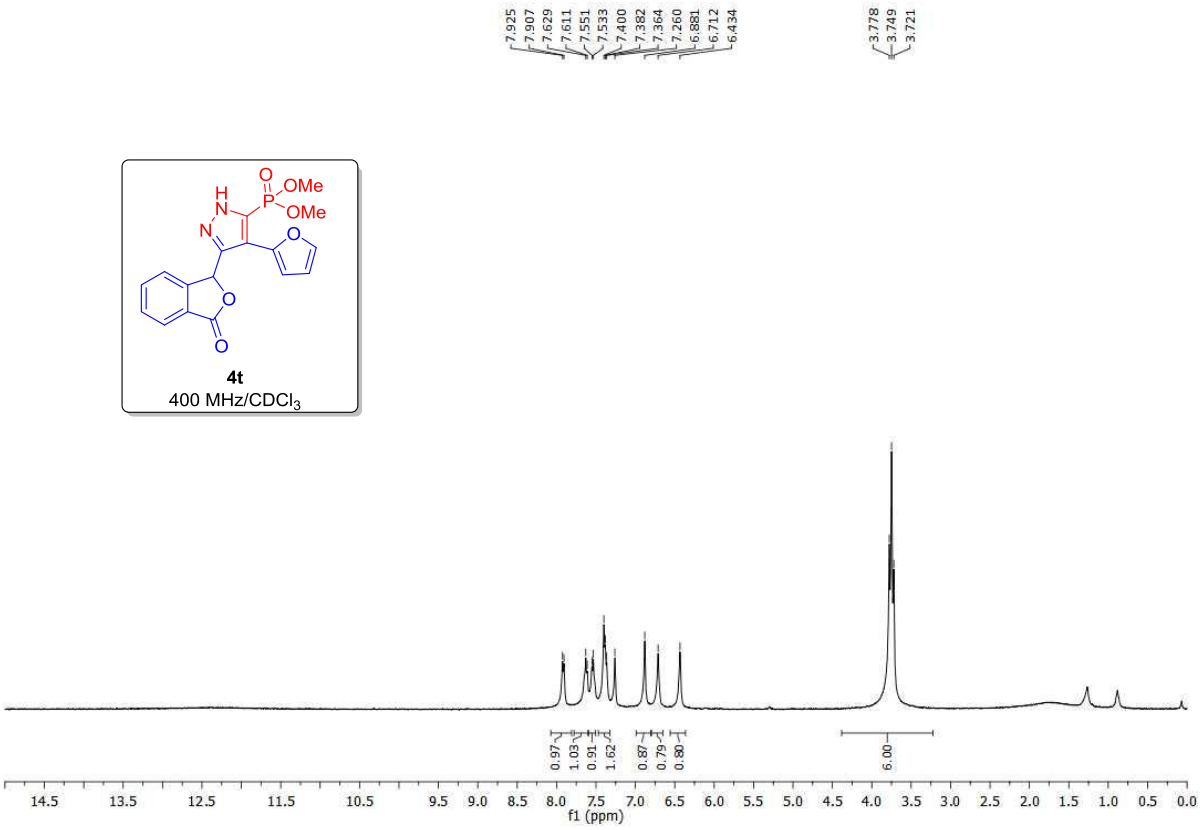

170.248
152.799
147.798
137.757
134.015
129.420
128.545
128.369
126.496
125.374
125.157
123.171
107.292
76.137
60.964
56.158
53.506
4r
100 MHz/CDCl3
f1 (ppm)

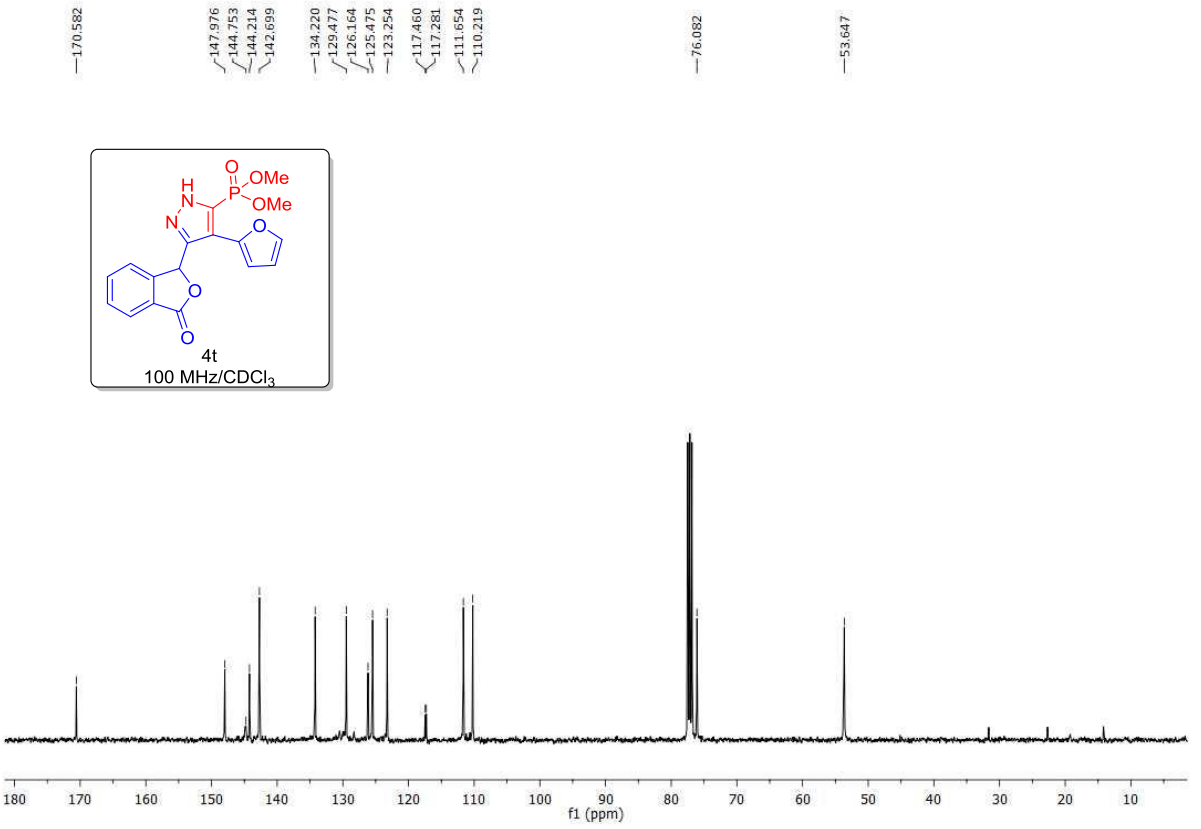

8.365
4r
161.9 MHz/CDCl3
f1 (ppm)

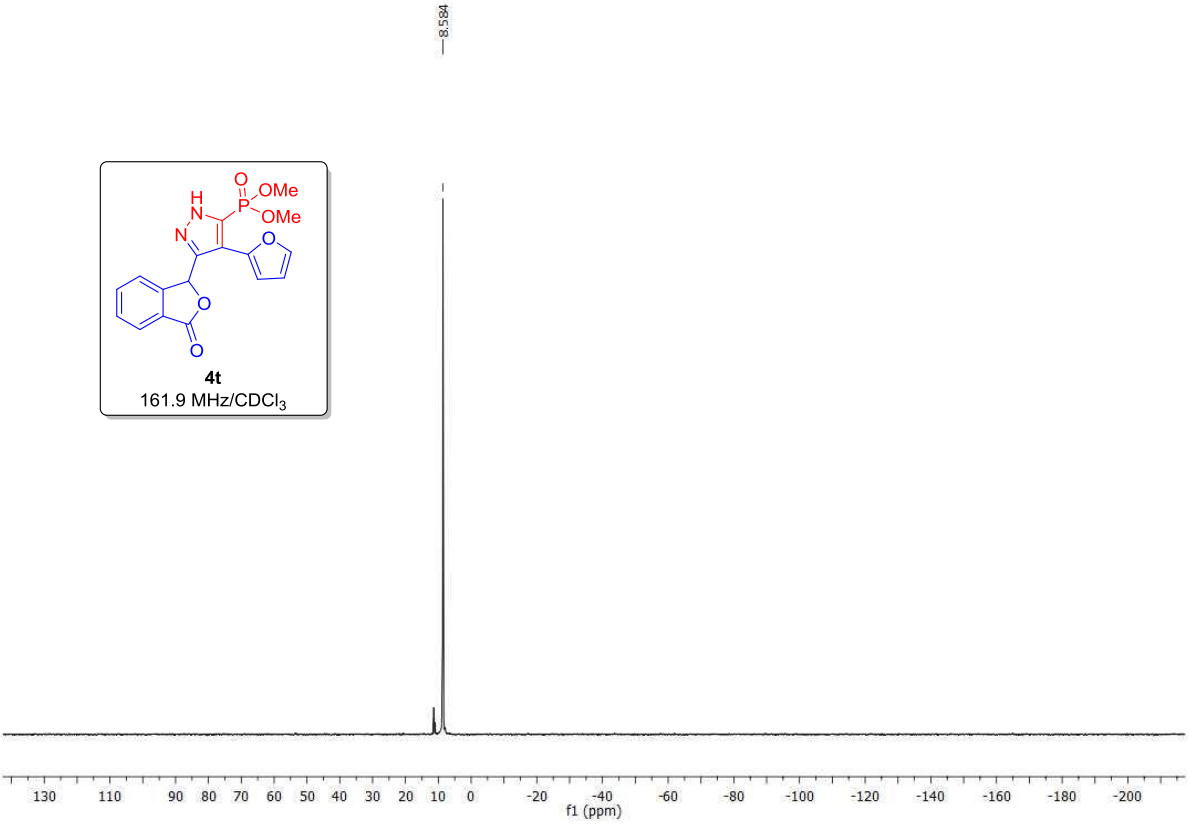

12.923
7.802
7.783
7.627
7.608
7.590
7.500
7.481
7.462
7.344
7.325
7.309
7.297
7.006
6.986
6.974
6.591
3.713
3.684
3.653
3.624
4s
400 MHz/CDCl3
0.89
0.92
0.98
1.07
1.68
1.82
0.86
6.00
f1 (ppm)

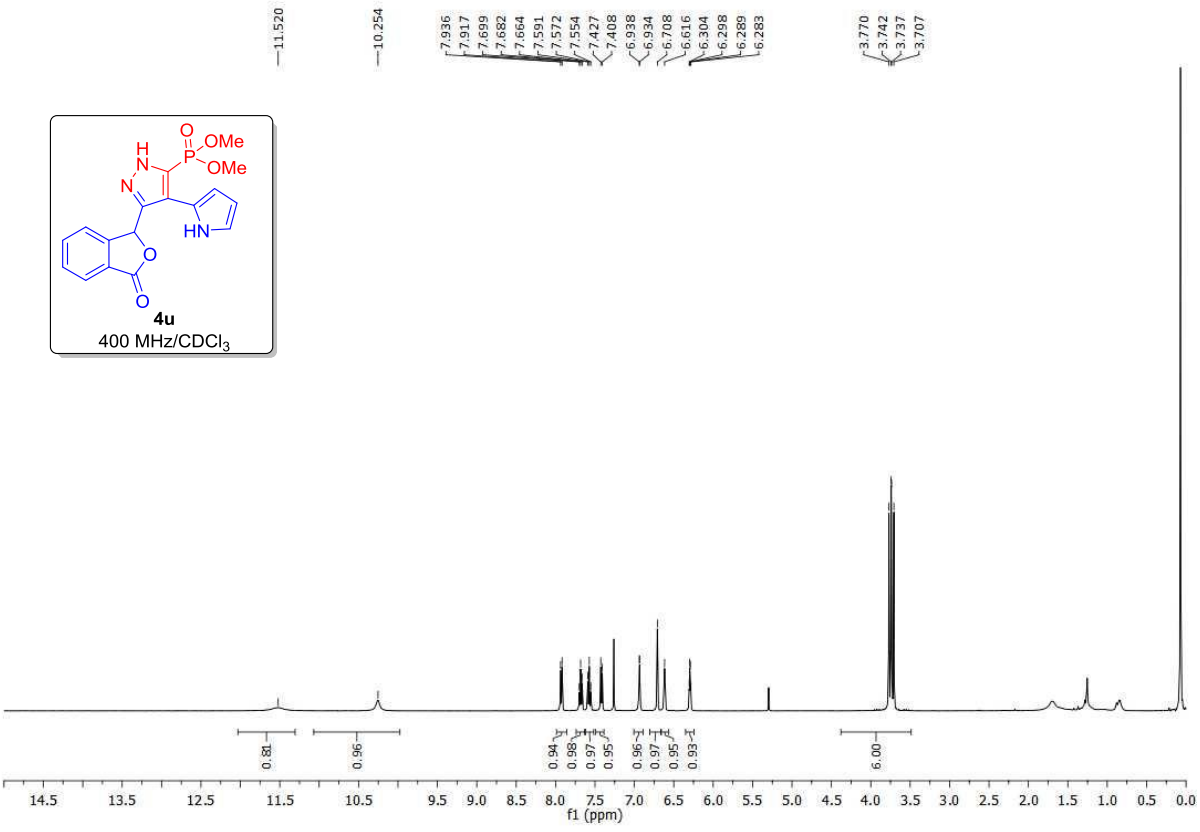

170.274
147.842
134.172
129.472
129.398
127.345
126.310
125.525
123.192
120.693
120.519
75.817
53.612
4s
100 MHz/CDCl3
f1 (ppm)

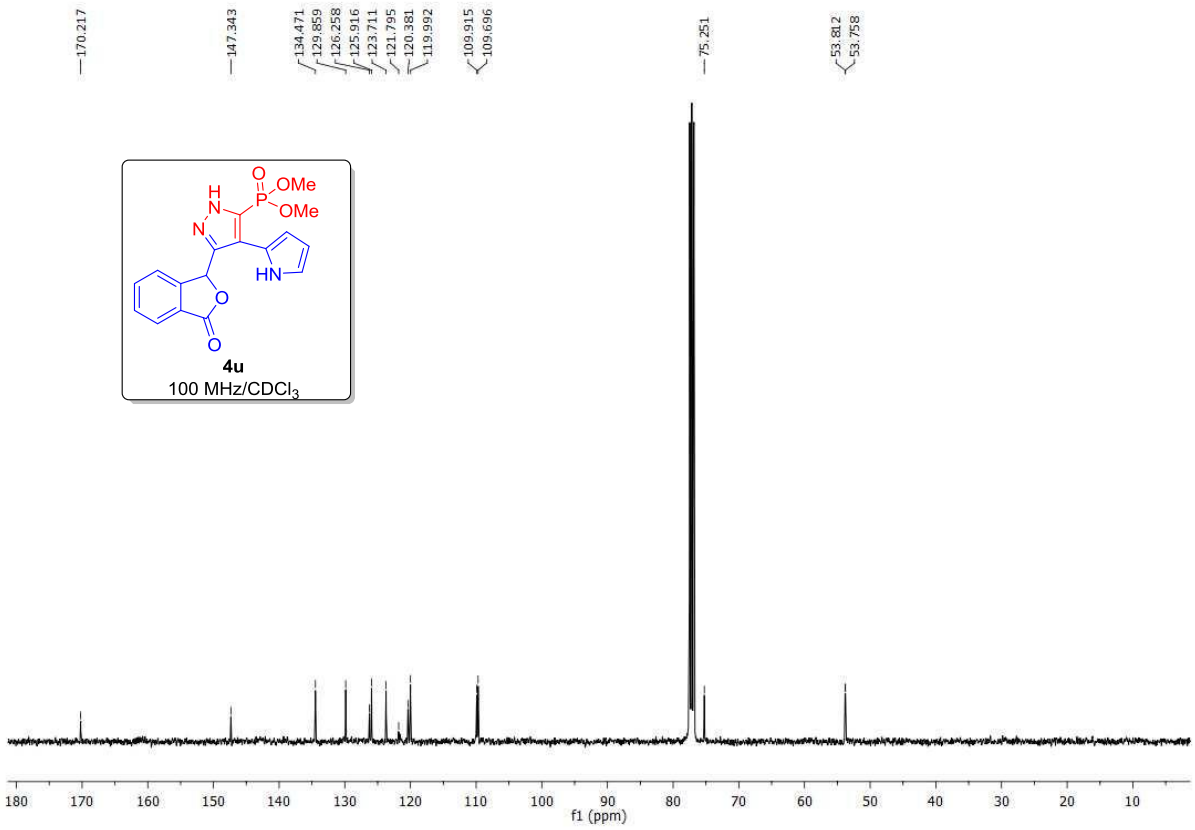

7.760
4s
161.9 MHz/CDCl3
f1 (ppm)

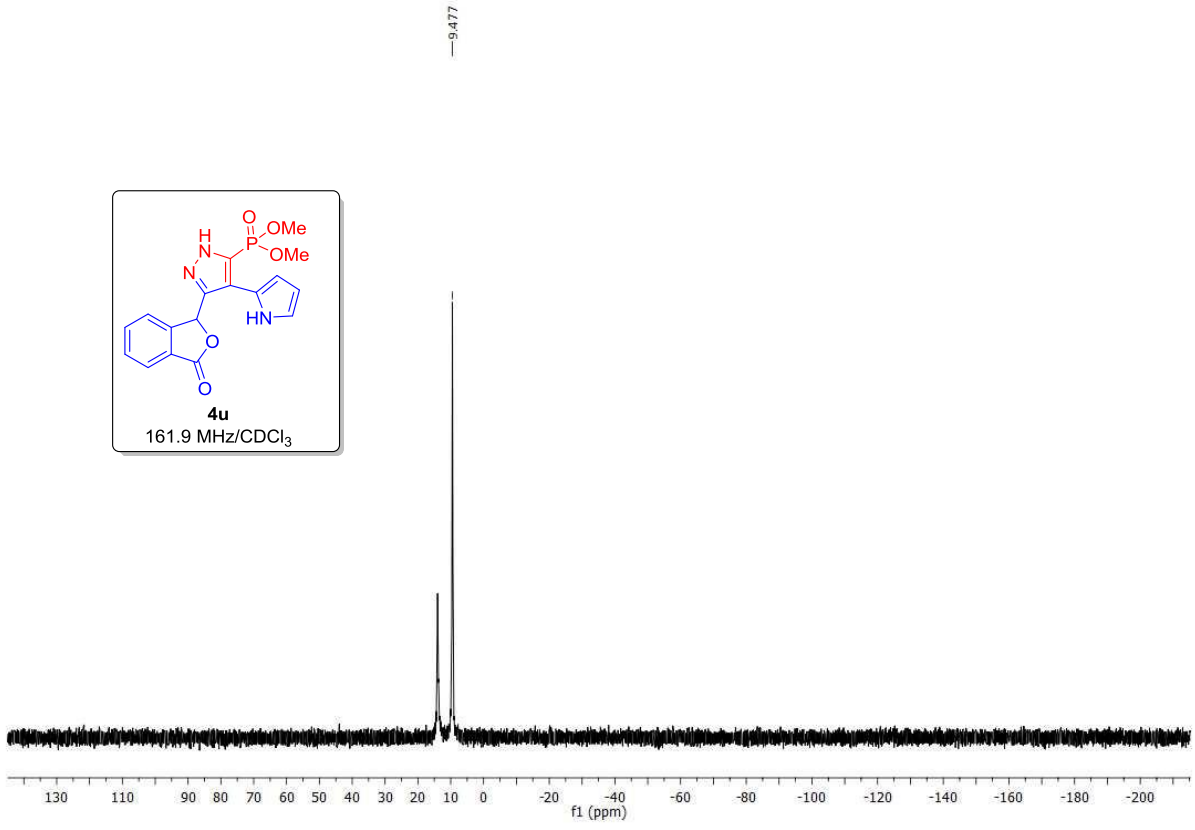

7.925
7.907
7.629
7.611
7.551
7.533
7.400
7.382
7.364
7.260
6.881
6.712
6.434
3.778
3.749
3.721
4t
400 MHz/CDCl3
0.97
1.03
0.91
1.62
0.87
0.79
0.80
6.00
f1 (ppm)

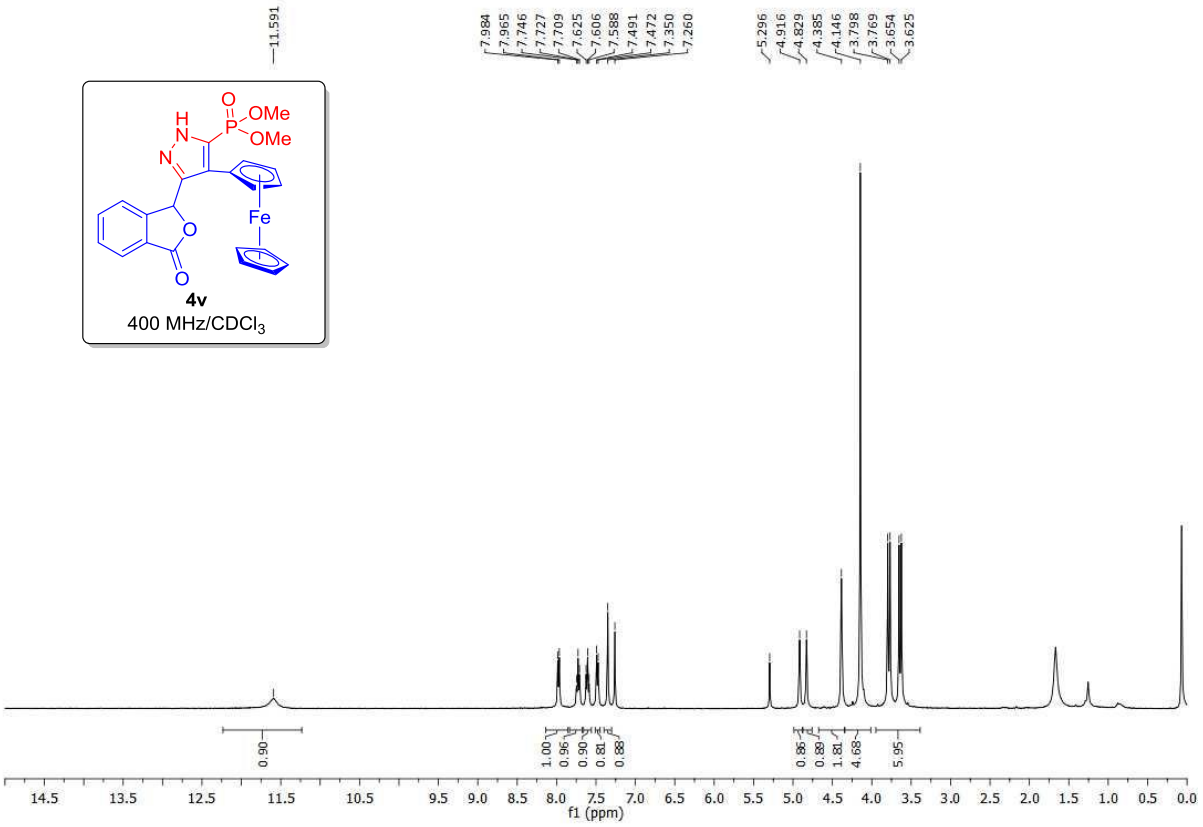

170.582
147.976
144.753
144.214
142.699
134.220
129.477
126.164
125.475
123.254
117.460
117.281
111.654
110.219
76.082
53.647
4t
100 MHz/CDCl3
f1 (ppm)

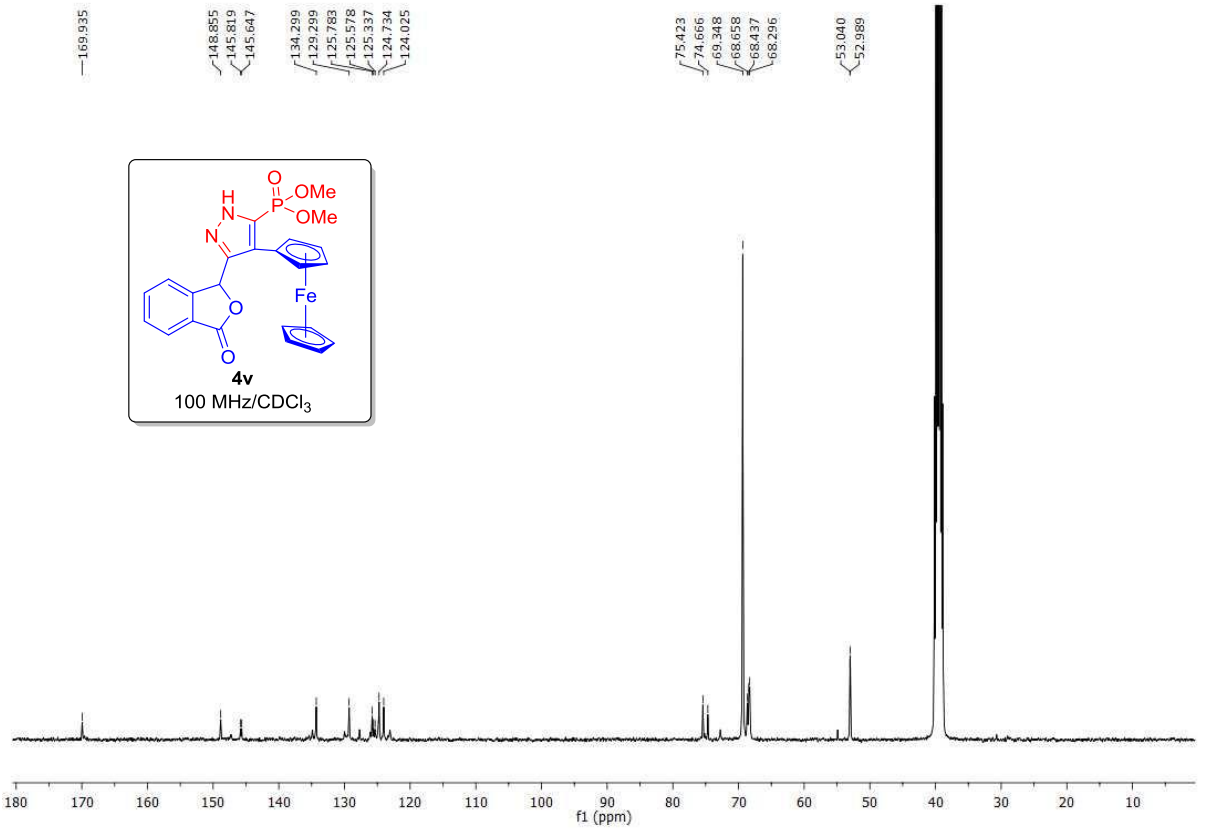

8.584
4t
161.9 MHz/CDCl3
f1 (ppm)

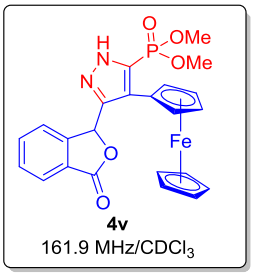

11.520
10.254
7.936
7.917
7.699
7.682
7.664
7.591
7.572
7.554
7.427
7.408
6.938
6.934
6.708
6.616
6.304
6.298
6.289
6.283
3.770
3.742
3.737
3.707
4u
400 MHz/CDCl3
0.81
0.96
0.94
0.98
0.97
0.95
0.96
0.97
0.95
0.93
6.00
f1 (ppm)


170.217
147.343
134.471
129.859
126.258
125.916
123.711
121.795
120.381
119.992
109.915
109.696
75.251
53.812
53.758
4u
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10
f1 (ppm)


9.477
4u
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -20 -40 -60 -80 -100 -120 -140 -160 -180 -200
f1 (ppm)

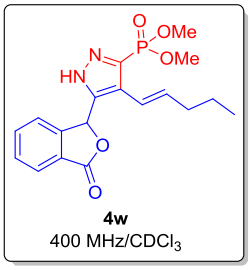

11.591
7.984
7.965
7.746
7.727
7.709
7.625
7.606
7.588
7.491
7.472
7.350
7.260
5.296
4.916
4.829
4.385
4.146
3.798
3.769
3.654
3.625
4v
400 MHz/CDCl3
0.90
1.00
0.96
0.90
0.81
0.88
0.86
0.89
1.81
4.68
5.95
f1 (ppm)


169.935
148.855
145.819
145.647
134.299
129.299
125.783
125.578
125.337
124.734
124.025
75.423
74.666
69.348
68.658
68.437
68.296
53.040
52.989
4v
100 MHz/CDCl3
f1 (ppm)



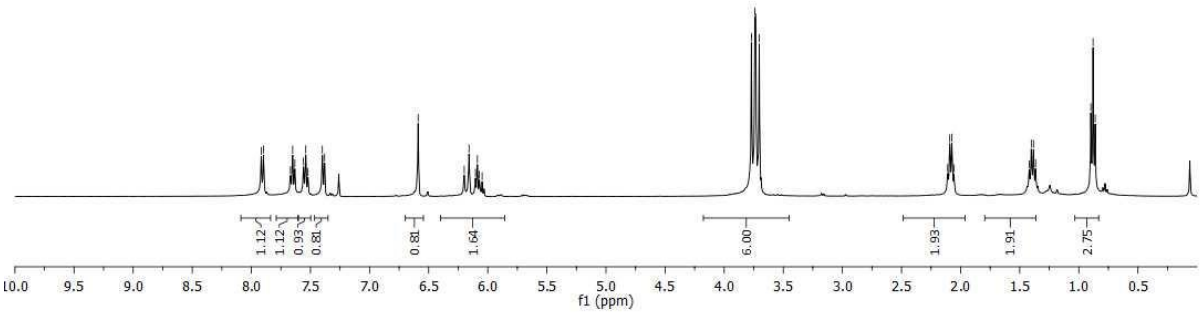
4v

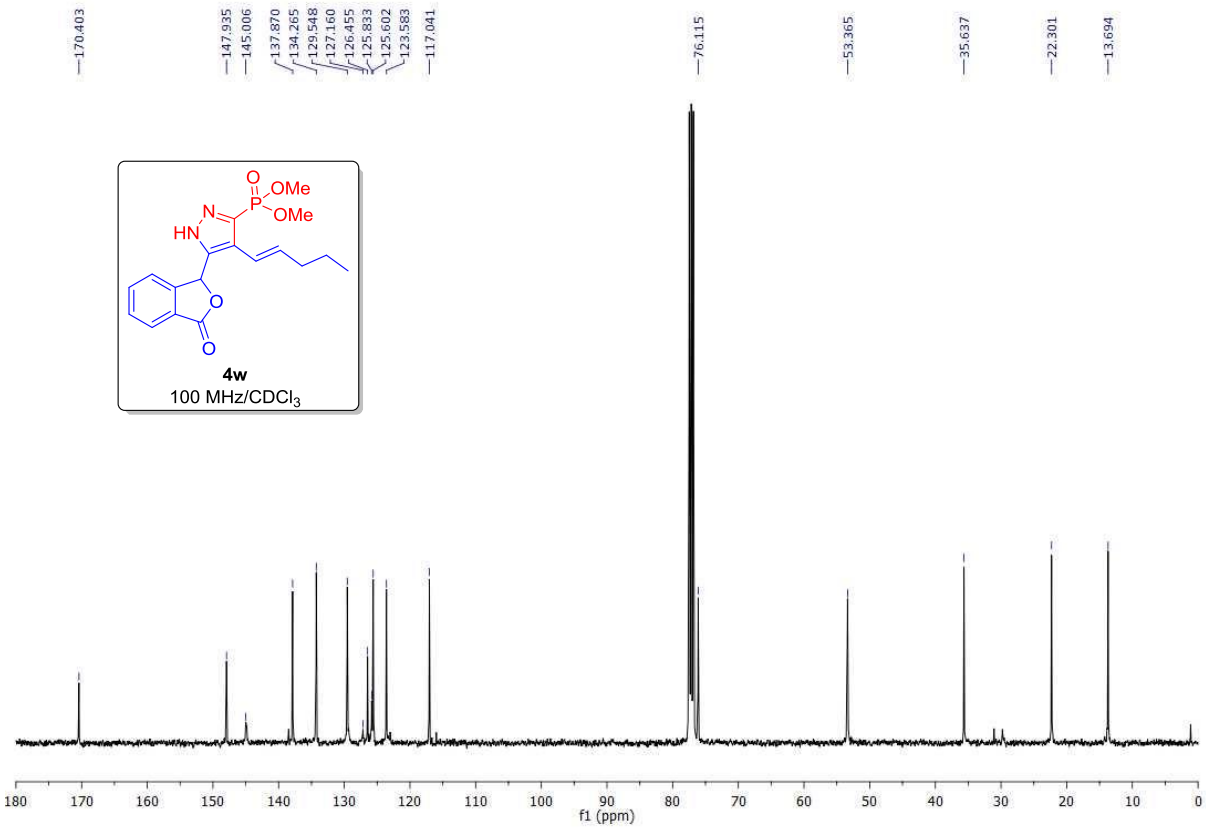
161.9 MHz/$CDCl_3$

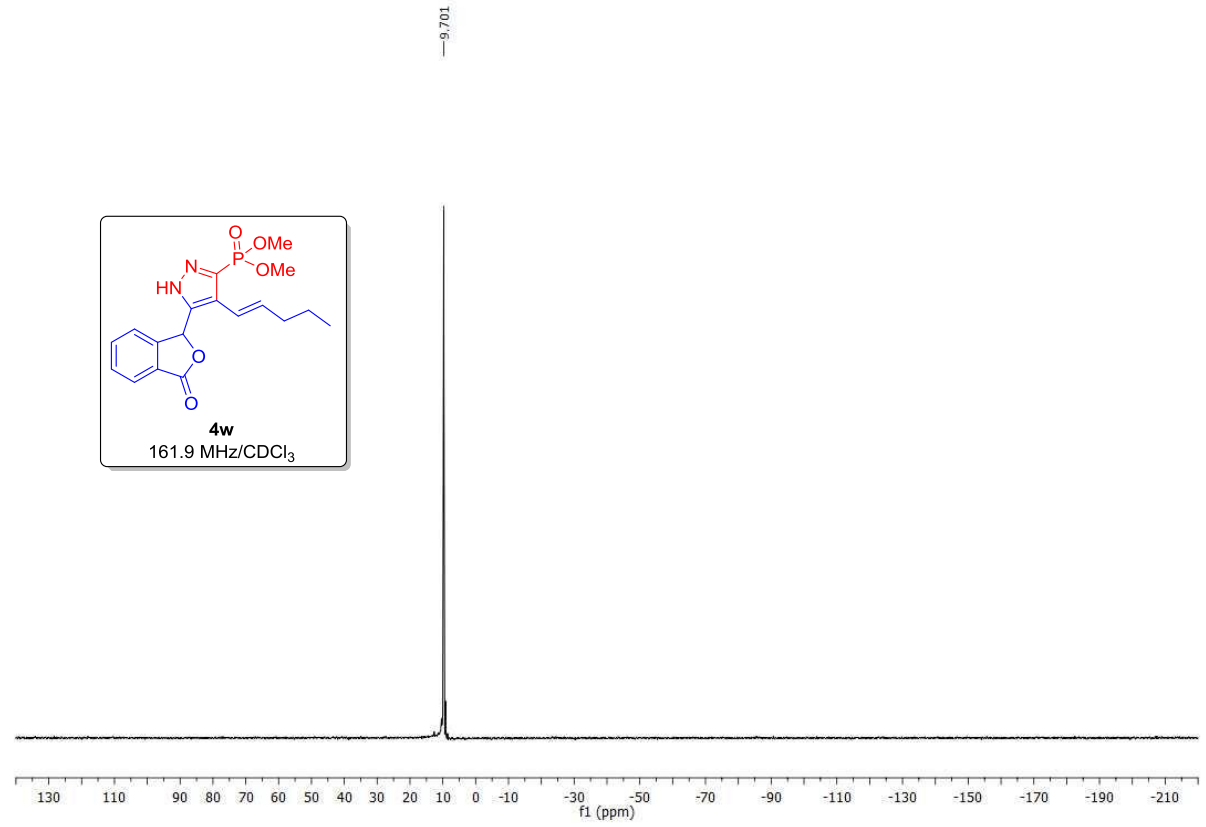


4w

400 MHz/$CDCl_3$




170.403
147.935
145.006
137.870
134.265
129.548
127.160
126.455
125.833
125.602
123.583
117.041
76.115
53.365
35.637
22.301
13.694
OMe
OMe
HN
N
P
O
O
O
4w
100 MHz/CDCl3
180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


9.701
OMe
OMe
HN
N
P
O
O
O
4w
161.9 MHz/CDCl3
130 110 90 80 70 60 50 40 30 20 10 0 -10 -30 -50 -70 -90 -110 -130 -150 -170 -190 -210
f1 (ppm)