

Supplementary Information for

Late-stage Isotopic Exchange of Primary Amines

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1. General Information

Commercial reagents and anhydrous solvents were purchased from Sigma-Aldrich and Fisher Scientific. All catalytic reactions were carried out under N₂ in Fisherbrand™ Disposable Borosilicate Glass Tubes with Threaded End (catalog #14-959-35C) fitted with Teflon caps under irradiation from PR160-456nm Kessil 34W LED lamps. Photocatalyst [Ir(dF-CF₃-ppy)₂(dtbbpy)]PF₆ was synthesized according to the reported procedures and Ru(bpy)₃(PF₆)₂ was purchased from Sigma Aldrich.

¹H NMR spectra were recorded on Bruker 400 or 500 MHz spectrometers at ambient temperature. Chemical shift is reported in parts per million (ppm) from CDCl₃ (7.26 ppm) with multiplicity (s = singlet, bs = broad singlet, d = doublet, t = triplet, q = quartet, and m = multiplet) and coupling constants (Hz). ¹³C NMR was recorded on Bruker 500 or 400 MHz spectrometers (126 MHz) at ambient temperature unless otherwise stated. ¹⁵N NMR was recorded on Bruker 500 spectrometers (51 MHz). Chemical shifts are reported in ppm from CDCl₃ (77.2 ppm). Mass spectra (LRMS) were recorded on an Agilent 7890B GC System 5977B MSD GCMS with an EI ionization method. High-resolution mass spectra (HRMS) were obtained on a Xevo G2 XS Q-ToF mass spectrometer in the positive electrospray ionization mode located at Columbia University chemistry department Mass Spectrometry Core Facility. Infrared spectra were recorded on a Perkin Elmer Spectrum Two FT-IR spectrometer. All cyclic voltammetry studies were performed on a CH Instruments Model 1232B potentiostat using an EDAQ 1-mm disk glassy carbon working electrode in conjunction with an EDAQ Ag/AgCl reference electrode and a platinum wire from VWR as a counter electrode. All CV experiments were performed in acetonitrile (MeCN).

Notes on product purification:

Thin layer chromatography was performed on SiliCycle® 250 μm, 60 Å plates, and preparatory TLC was performed on SiliCycle® 1000 μm, 60 Å plates. All silica plates were basified by soaking in 5% Et₃N/hexanes solution for ~3 minutes, then dried for two hours at room temperature. Visualization was accomplished with 254 nm UV light. Flash chromatography on SiliCycle® Silica Flash® 40-63 μm, 60 Å using basified silica (pre-treated with 5% Et₃N/hexanes), and 1 drop of Et₃N was added per 100 mL of eluent.

The leftover ^{15}N -benzophenone imine can be recovered from the reaction using basified silica mentioned above, or via distillation if on large enough scale using the conditions mentioned in the starting material synthesis.

2. Extended Optimization and Control Experiments

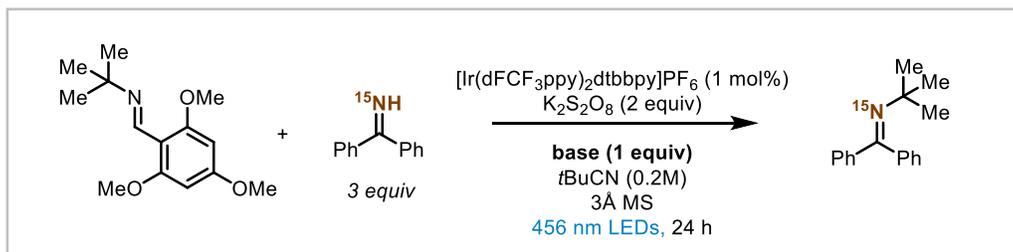
Radical Polar Crossover Optimization

All reactions were run at 0.1 mmol and analyzed via GCMS with mesitylene (0.1 mmol) as an internal standard using a calibration curve.

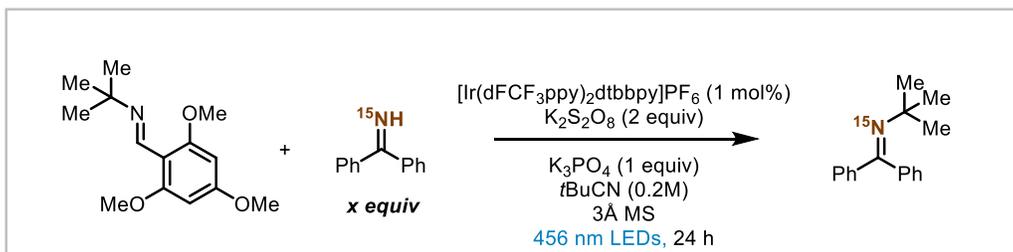
Table S1. Effect of solvent.

<i>solvent</i>	<i>yield</i>	<i>trimethoxybenzointrile</i>	<i>aldehyde (hydrolysis)</i>
DCM	12%	17%	4%
DCE	37%	47%	5%
DCE (no MS)	34%	42%	17%
CHCl_3	4%	4%	8%
benzene	29%	23%	3%
PhCF_3	46%	46%	--%
<i>t</i> BuCN	55%	65%	--%

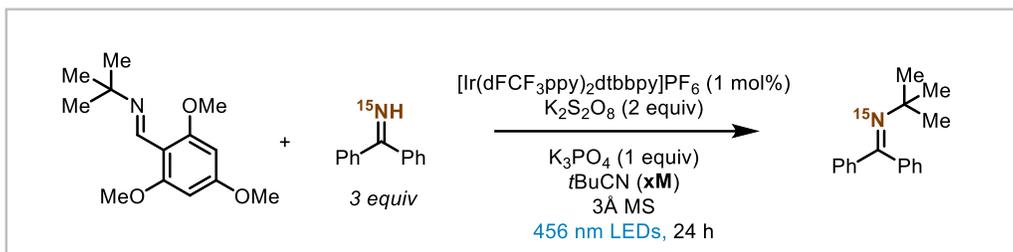
When molecular sieves were omitted from the reaction, significantly more trimethoxybenzaldehyde was observed due to hydrolysis, as well as the formation of the hydrated adamantanol. This supports the intermediacy of a carbocation, and that water can outcompete benzophenone imine as a nucleophile. Interestingly, with pivalonitrile, no amide (Ritter-type byproduct) was observed even with the omission of molecular sieves, but is observed when using acetonitrile as a solvent.

Table S2. Effect of base.

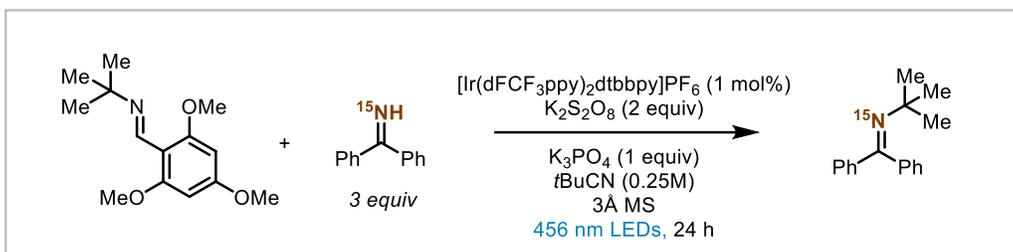
base	yield	trimethoxybenzonitrile
K_3PO_4	37%	51%
TMG	6%	10%
2,4,6-collidine	34%	65%
2,6-lutidine	31%	66%
none	32%	58%
2 equiv K_3PO_4	27%	74%
3 equiv K_3PO_4	24%	61%

Table S3. Nucleophile equivalents.

Nu equiv	yield	trimethoxybenzonitrile
1.2 equiv	29%	54%
2 equiv	34%	54%
5 equiv	37%	48%

Table S4. Concentration.

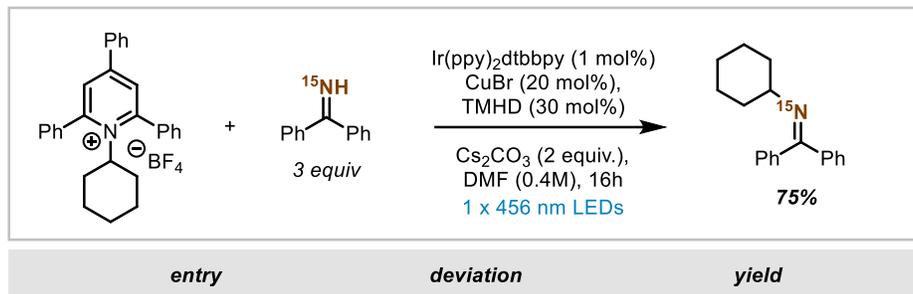
concentration	yield	trimethoxybenzimidazole
0.1M	23%	51%
0.25M	51%	72%
0.4M	46%	73%

Table S5. Controls.

deviation	yield	trimethoxybenzimidazole
no PC	n.d.	n.d.
no light	n.d.	n.d.
no K ₂ S ₂ O ₈	25%	26%
0.5 mol% Ir PC	38%	38%
1.1 equiv K ₂ S ₂ O ₈	51%	65%

Copper catalysis optimization: All reactions were run at 0.1 mmol and analyzed via GCMS with benzyl benzoate (0.1 mmol) as an internal standard using a calibration curve.

Table S6. Initial hit and controls.



Running the reaction without photocatalyst still results in 63% yield, which led us to investigate if the mechanism is going through an electron-donor-acceptor complex. Indeed, running UV-Vis experiments indicate a clear formation of a new complex that we can ascribe to an electron-donor acceptor complex upon mixing of the Cu(I) catalyst with the Katritzky salt and base. Alternatively, the Cu(I)-imido species could also form a UV-active EDA complex with the Katritzky salt.

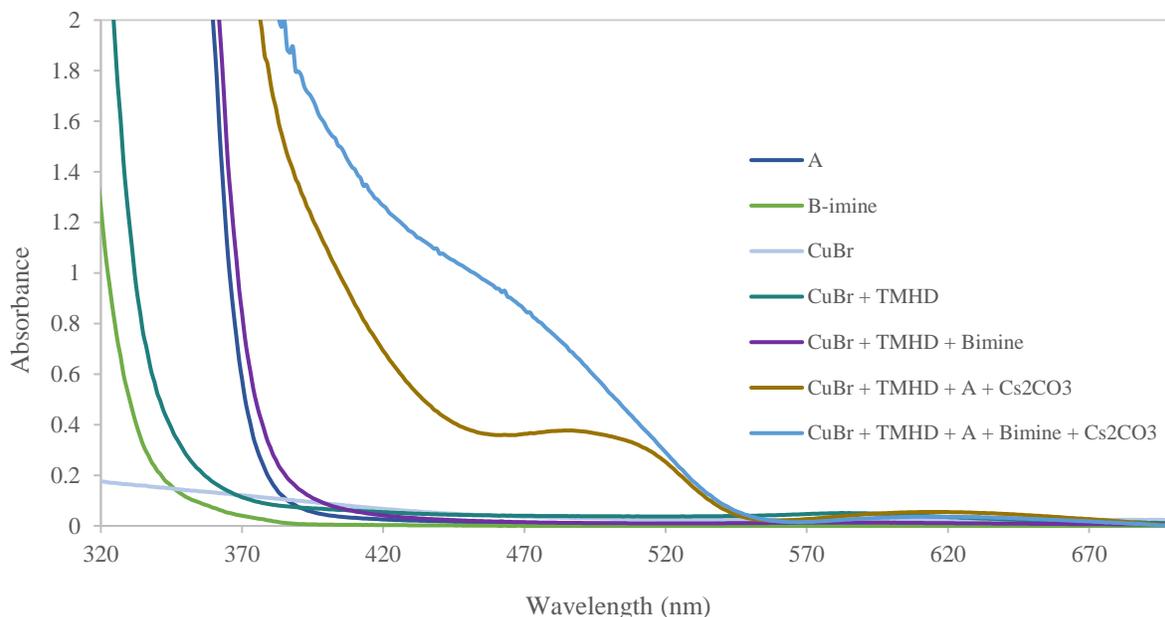


Figure S1. UV-Visible absorption spectra of the reaction system. Each sample contained 0.03 mmol of substrate in 3 mL of degassed and dry DMF. The spectra were taken after ten minutes of stirring and sealed to prevent contact with air.

In addition, a Job plot (method of continuous variation (MCV)) was performed to determine the stoichiometry of the EDA complex. Different solutions of Katritzky salt **A** and Cu(I)(TMHD) in DMF were prepared, keeping the sum of both concentrations at a constant value of $C_{\text{tot}} = 0.1 \text{ mol L}^{-1}$. The UV/Vis absorption at $\lambda = 495 \text{ nm}$ was then recorded as a function of the molar fraction of Cu(I)(TMHD) χ . The curve peaks at 0.5, indicating a 1:1 stoichiometry of the Katritzky salt and the Cu(I)(TMHD) species.

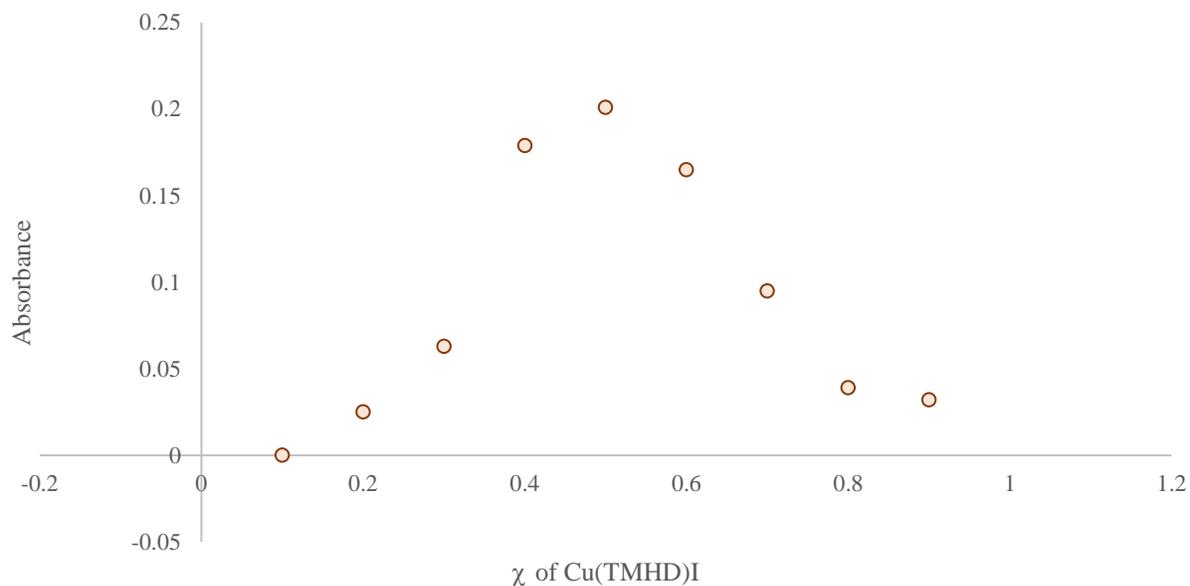


Figure S2. Job plot of a mixture of Katritzky salt **A** and Cu(I)(TMHD).

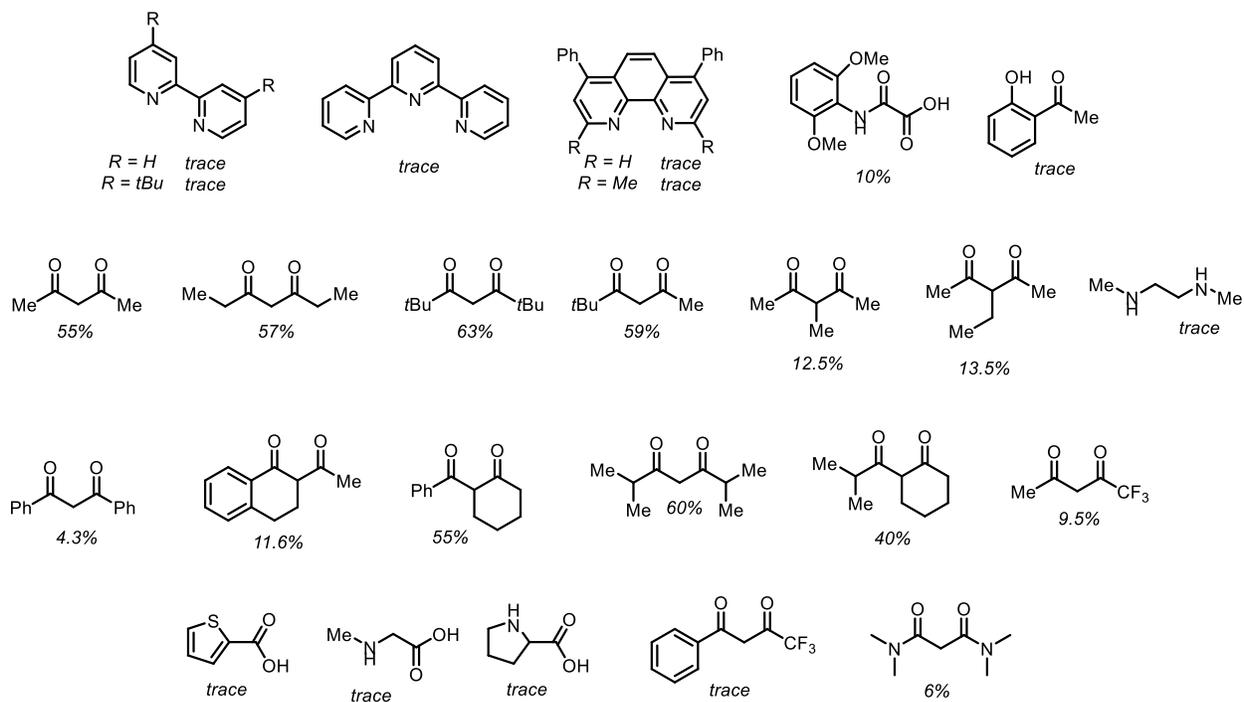
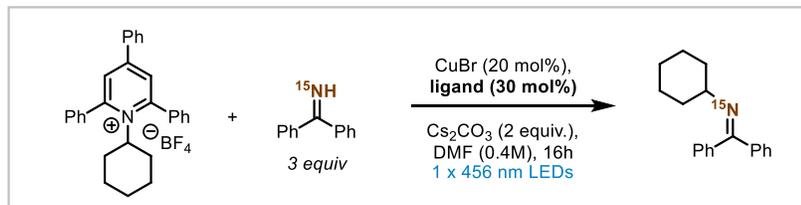
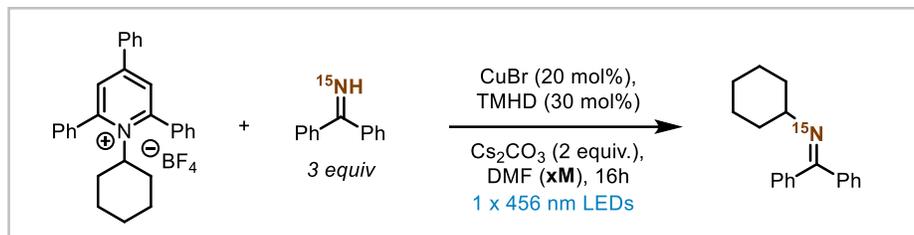


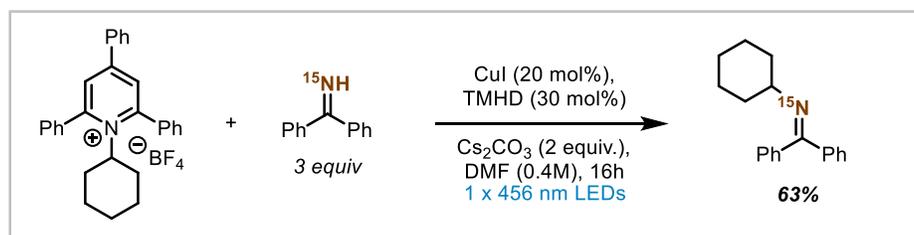
Figure S3. Effect of ligand.

Table S7. Effect of concentration.



entry	concentration	yield
1	0.2M	25%
2	0.4M	63%
3	0.6M	64%
4	1.0M	65%
4	0.4M, 20% CuI	65%

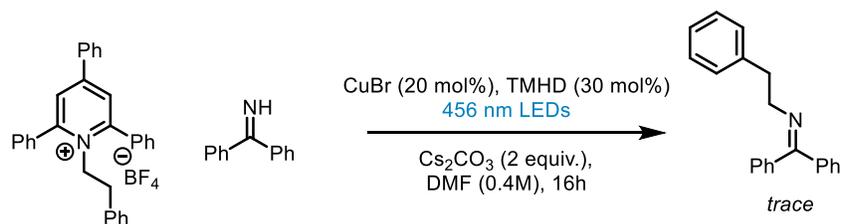
Table S8. Temperature and light source.



entry	deviation	yield
1	no fan (~70C)	60%
2	2 x 456 nm lights (3 cm away)	72%
3	2 x 456 nm lights (1-2 cm away)	87%

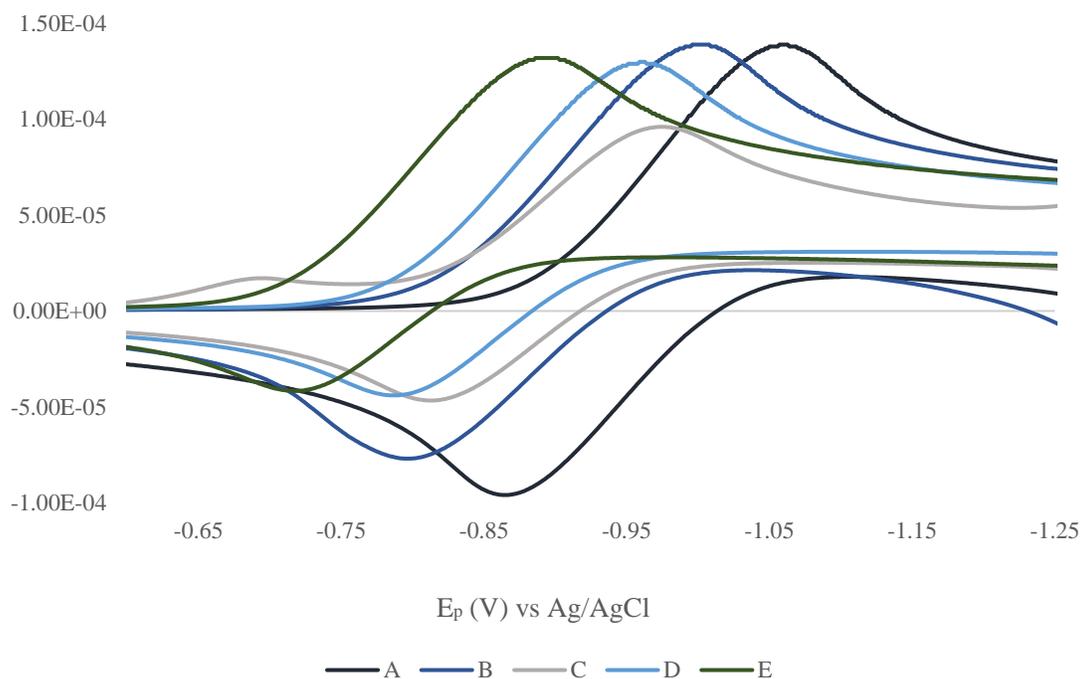
Copper catalysis with α -primary amines:

Initial limitation:



After receiving trace yield of product when using an α -primary amine as the substrate, we wondered if the lack of reactivity was due to a disruption of the EDA complex. Indeed, UV-Vis absorption spectra indicated the EDA complex is lost when utilizing the primary Katritzky salt. Thus, we questioned if it was due to electronic differences between the amine classes, or if a steric contribution was the key contributor.

The reduction potential (E_p) of the cyclohexyl-substituted Katritzky salt **B** is -1.002 V vs Ag/AgCl (see below), while the reduction potential of Katritzky salt **A** is -1.060V, making **A** 58 mV harder to reduce. Therefore, we synthesized **C**, which has a more positive reduction potential. However, the yield only increased to 17%. Thus, we suspected that sterics and rigidity of the Katritzky salt had an influence, leading us to synthesize **D**. Katritzky salts **C** and **D** both have similar reduction potentials, but **D** is rigidified by ethylene bridges between the phenyl substitution at the pyridinium core. The yield was then restored to 63%. Hardly any effect to yield was observed when making this constricted Katritzky salt more electron deficient and therefore easier to reduce (**E**), leading us to believe that steric contributions are more prevalent in reactivity.



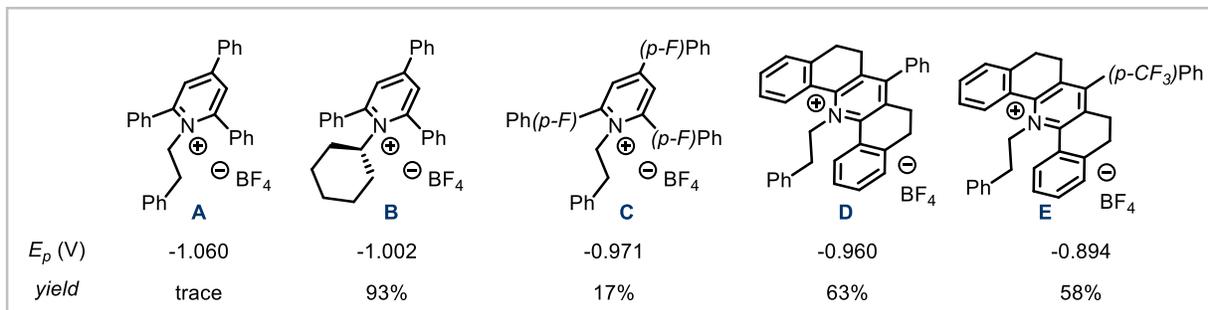


Figure S4. Cyclic voltammetry spectra of the first reduction wave of Katritzky salts A-E. Listed are the reduction potentials and corresponding yields of reaction.

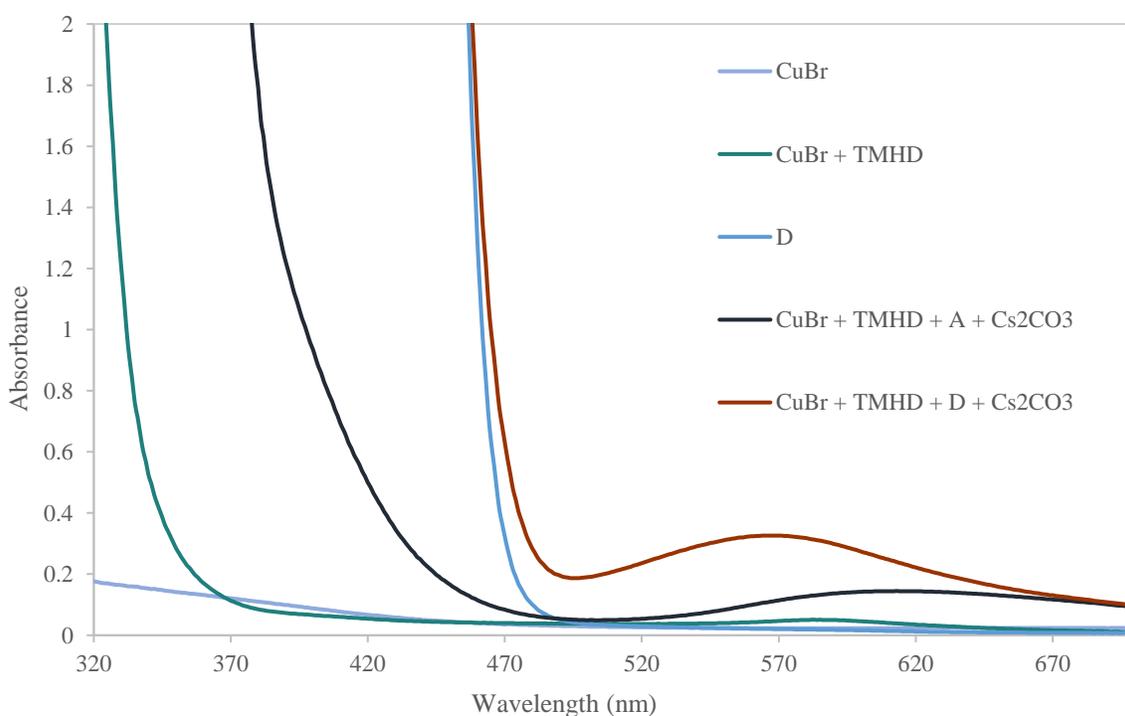


Figure S5. UV-Vis absorption spectra of α -primary Katritzky salts. Each sample contained 0.03 mmol of substrate in 3 mL of degassed and dry DMF. The spectra were taken after thirty minutes of stirring and sealed to prevent contact with air.

Alternatively, $^{15}\text{NH}_4\text{Cl}$ can be used as the nucleophile (**Figure S6**). This bypasses the need for the ^{15}N -benzophenone imine synthesis and gives synthetically useful yields of **S-1**. However, we found that the reaction was rather irreproducible (yields ranged from 50-80%), and the scope of

the reaction was not as general to all classes of amines. The reaction is low yielding for α -2° amines, and no product is observed when moving to α -1° amines.

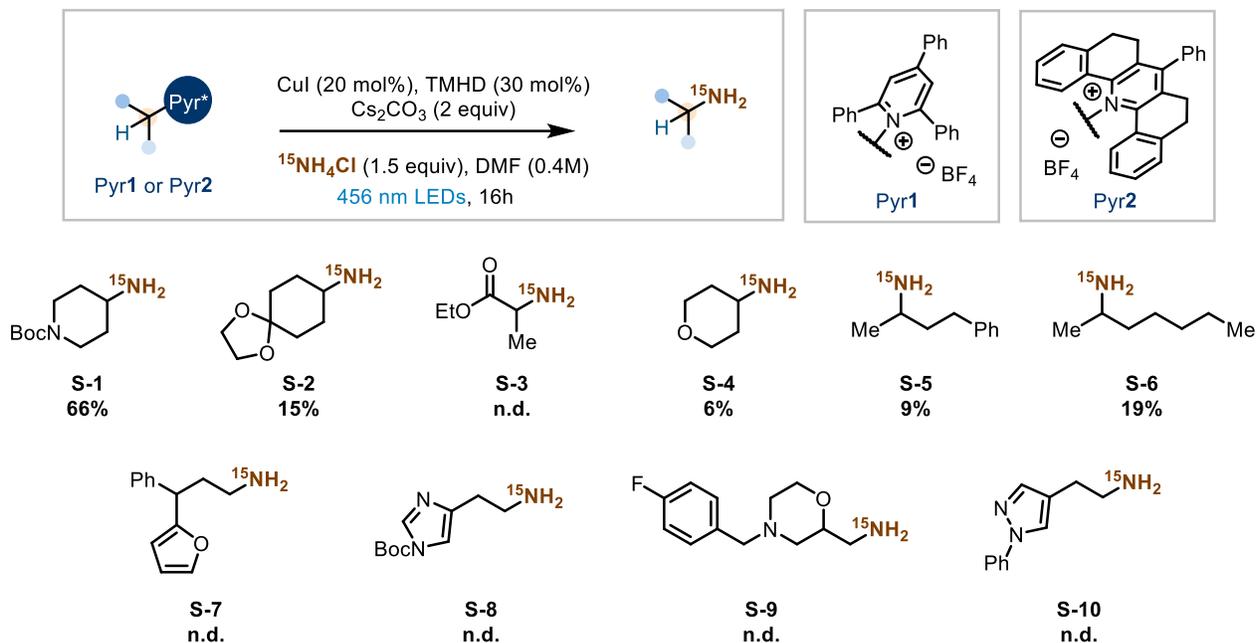


Figure S5. Scope using $^{15}\text{NH}_4\text{Cl}$ as a nucleophile. Reactions were run on 0.1 mmol scale. All yields are corrected GCMS yields with benzyl benzoate as an internal standard.

Unsuccessful Substrates:

In the RPC conditions, substrates S11-S13 provided no desired product. The pyridyl substrates afforded the homodimerization of the tertiary radical, indicating a preference for dimerization over oxidation to form the carbocation. In the case of S-13, no trimethoxybenzonitrile was formed, likely due to competitive oxidation of the tertiary amine.

In the Cu system, drug-like compounds Amlodipine and Primaquine afforded the cyclized product resulting from intramolecular C-N cross-coupling. Substrates S-16 and S-17 underwent elimination to liberate triphenyl pyridine and the corresponding alkene.

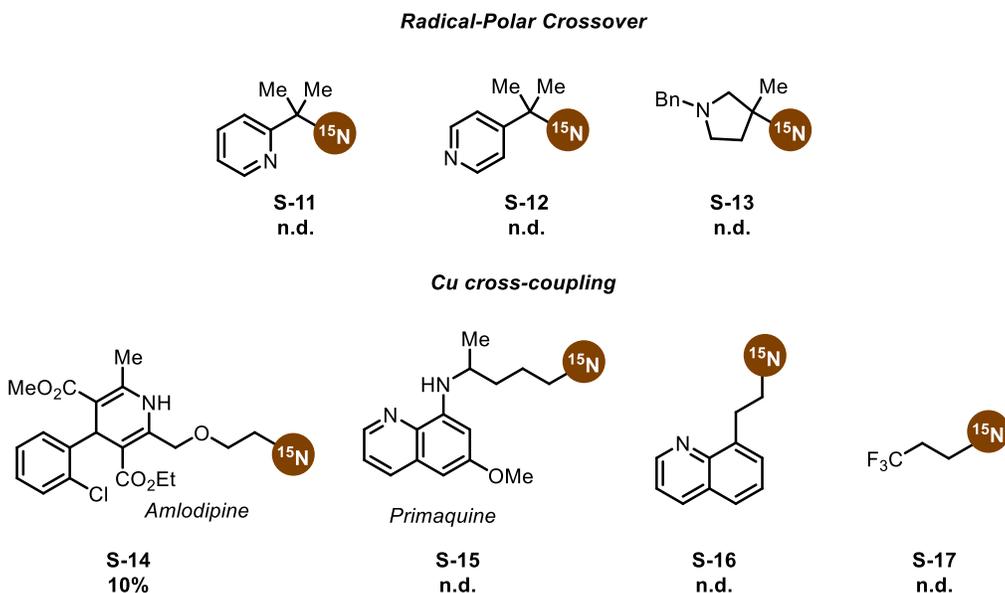
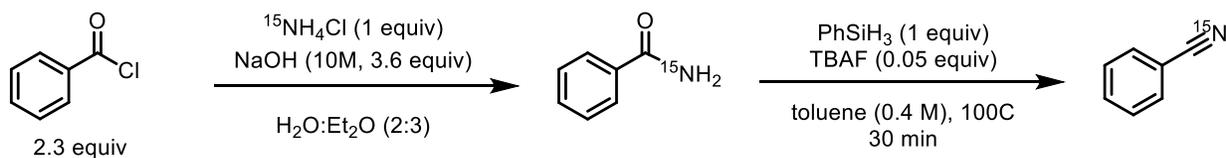


Figure S6. Unsuccessful substrates in the isotopic exchange.

3. Starting Material Synthesis

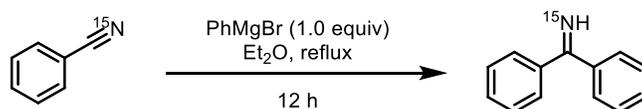
Synthesis of ^{15}N -benzophenone imine:



Synthesis of ^{15}N -benzamide: *Note, no precautions to air were taken*. To a 150 mL round bottom flask was added ^{15}N -ammonium chloride (2.0 g, 1 equiv), H_2O (8 mL), Et_2O (12 mL), and benzoyl chloride (9.80 mL, 2.3 equiv). The solution was cooled to 0°C , to which a 10M NaOH solution (also pre-cooled to 0°C , 13.2 mL, 3.6 equiv) was added dropwise via addition funnel. The solution was stirred for 5 minutes at 0°C and 5 minutes at room temperature, then filtered. The solid was washed with Et_2O and dried to give 4.196 g of a white solid (94%). Spectra matches that in the literature.¹

Synthesis of ^{15}N -benzonitrile: To a flame-dried 100 mL round bottom three-neck flask was added ^{15}N -benzamide (3.0 g, 1 equiv). The flask was evacuated and backfilled with Ar (3x). Phenylsilane (3.028 mL, 1 equiv) and toluene (40 mL) were added to the flask. An outlet was inserted into a

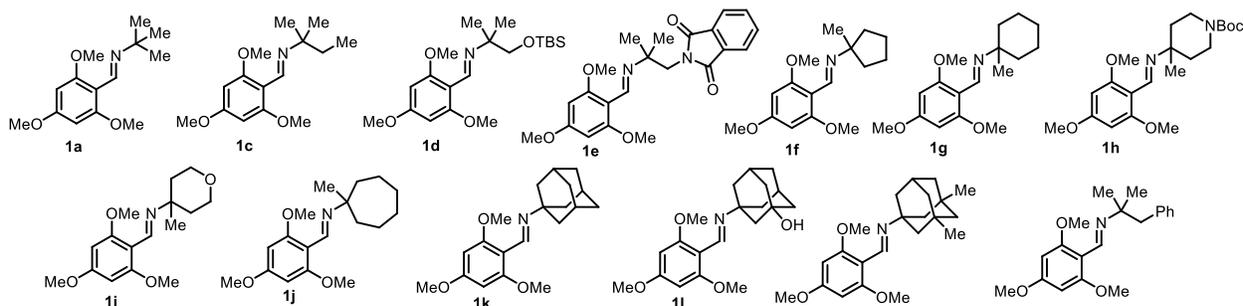
septum of three neck flask under positive Ar pressure, followed by dropwise addition of TBAF (1.228 mL, 1.0 M in THF). A large gas formation was observed. Once gas extrusion had slowed, the mixture was heated to 100°C for 30 minutes. The mixture was concentrated *in vacuo* (rotovap bath temp = 30°C) and then purified by silica gel chromatography to yield 2.032 g (79%) of a light yellow oil. Spectra matches that in the literature.²



To a flame-dried 100 mL round bottom flask under Ar was added Et₂O (9.5 mL) and PhMgBr (4.786 mL, 3.0 M in Et₂O, 1 equiv). To this mixture was added a solution of ¹⁵N-benzonitrile (1.495 g, 1 equiv) in 7 mL of Et₂O dropwise. The resulting mixture was heated to reflux overnight. The heating bath was removed, and methanol (6 equiv) was added slowly, while an intensive heat formation was observed. The slurry was stirred for another two hours, then filtered. The filter cake was washed with Et₂O, and the filtrate was evaporated to give a light orange oil. The oil was purified via distillation under vacuum (b.p. 116-125 @ 0.5 torr, oil bath temp ~170 when it first starts distilling) or purification on basic silica (pre-treated with Et₃N) to yield 1.590 g (61%) of a light yellow oil. Spectra match the literature.³

Synthesis of redox-active imines:

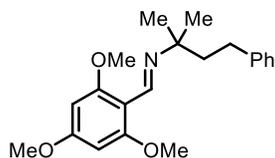
The following redox-active imines were synthesized according to previous literature procedures.^{4,5}



Imines were synthesized according to a modified literature procedure.⁵ A mixture of 2,4,6-trimethoxybenzaldehyde (1.0 equiv.) and primary amine (1.1 equiv. or 2.0 equiv. if volatile) in benzene (0.1M) was heated in a Dean-Stark apparatus to reflux overnight. The reaction was then cooled, dried with Na₂SO₄, filtered, and concentrated *in vacuo*. Volatile amines were pumped off

and/or able to be washed away with hexanes, in which the imine would crash out (additional cooling sometimes required). Imines carried forward without further purification (95-100% purity).

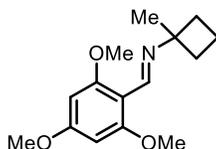
If using an ammonium salt, the following modified procedure can be used: The ammonium salt (1.2 equiv) and crushed potassium hydroxide (1.2 equiv) were added to a flask, followed by benzene (0.1M). Three drops of water were added to the flask and stirred for five minutes. Then, 2,4,6-trimethoxybenzaldehyde (1.0 equiv.) was added and heated in a Dean-Stark apparatus to reflux overnight. After cooling to room temperature, hexanes is added to the mixture and is triturated to remove the ammonium hydroxide. The benzene/hexanes solution is concentrated in vacuo to yield pure imine. If excess amine is remaining, hexanes is added and the solution is cooled to either 0°C or -78°C and triturated to yield the solid imine.



(1c) *N*-(2-methyl-4-phenylbutan-2-yl)-1-(2,4,6-trimethoxyphenyl)methanimine. White solid (84%).

¹H NMR (300 MHz, CDCl₃) δ 8.37 (s, 1H), 7.33 – 7.10 (m, 5H), 6.14 (s, 2H), 3.83 (s, 3H), 3.81 (s, 6H), 2.73 – 2.62 (m, 2H), 1.97 – 1.85 (m, 2H), 1.33 (s, 6H).

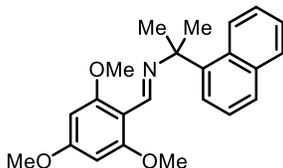
¹³C NMR (126 MHz, CDCl₃) δ 161.87, 160.11, 151.28, 143.68, 128.42, 128.23, 125.38, 108.94, 90.83, 60.01, 55.92, 55.28, 45.99, 30.76, 27.30.



(1m) *N*-(1-methylcyclobutyl)-1-(2,4,6-trimethoxyphenyl)methanimine. Isolated as an off white solid (40%).

¹H NMR (500 MHz, CDCl₃) δ 8.23 (s, 1H), 6.11 (s, 2H), 3.81 (s, 3H), 3.80 (s, 6H), 2.38 – 2.23 (m, 3H), 2.12 – 1.99 (m, 3H), 1.92 – 1.78 (m, 1H), 1.72 (dtt, *J* = 11.0, 9.6, 2.6 Hz, 1H), 1.40 (s, 3H).

^{13}C NMR (126 MHz, CDCl_3) δ 162.11, 160.47, 150.97, 108.36, 90.89, 64.58, 56.12, 55.40, 35.16, 27.87, 13.99.

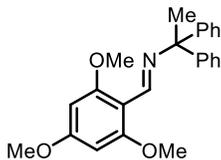


(1n) *N*-(2-phenylpropan-2-yl)-1-(2,4,6-trimethoxyphenyl)methanimine

Basified first with a NaOH/DCM wash and isolated free amine to subject to condensation. White solid (99%).

^1H NMR (500 MHz, CDCl_3) δ 8.40 (s, 1H), 7.57 – 7.53 (m, 2H), 7.33 (dd, $J = 8.4, 7.2$ Hz, 2H), 7.22 – 7.18 (m, 1H), 6.13 (s, 2H), 3.81 (s, 9H), 1.65 (s, 6H).

^{13}C NMR (126 MHz, CDCl_3) δ 162.09, 160.33, 152.95, 149.37, 127.89, 126.38, 125.91, 108.82, 90.87, 63.37, 56.00, 55.32, 29.96.



(1o) *N*-(1,1-diphenylethyl)-1-(2,4,6-trimethoxyphenyl)methanimine

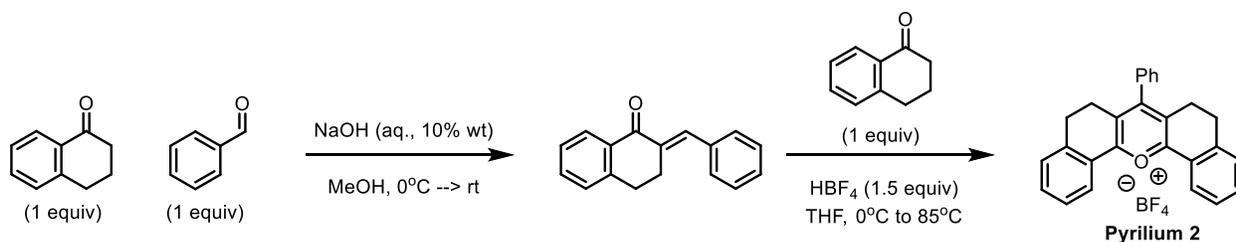
White solid (90%).

^1H NMR (500 MHz, CDCl_3) δ 8.04 (s, 1H), 7.47 – 7.42 (m, 4H), 7.30 (dd, $J = 8.4, 7.1$ Hz, 4H), 7.24 – 7.17 (m, 2H), 6.12 (s, 2H), 3.82 (s, 3H), 3.81 (s, 6H), 1.98 (s, 3H).

^{13}C NMR (126 MHz, CDCl_3) δ 162.32, 160.58, 154.84, 148.35, 128.28, 127.92, 126.24, 108.97, 91.10, 70.47, 56.20, 55.45, 29.67.

Synthesis of Perylium Salts –

The perylium salts were synthesized according to Cornella's procedure.⁶

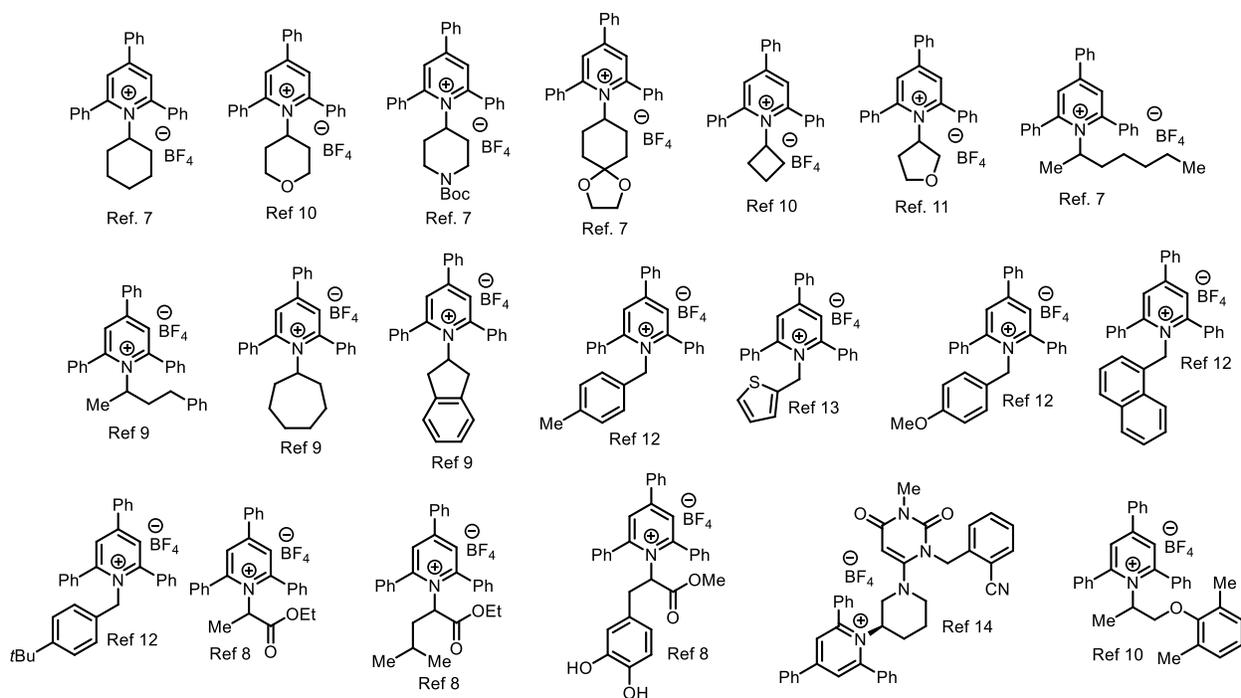


To a round-bottom flask was added α -tetralone (1.0 equiv, 120 mmol), benzaldehyde (1.0 equiv, 120 mmol) and MeOH (0.5 mL/mmol, 60 mL). The reaction system was cooled with an ice bath, to which NaOH aqueous solution (10 wt%, 5 equiv, 600 mL) was added. The ice bath was removed after addition and the mixture was stirred at room temperature until complete conversion was judged by TLC. If the corresponding chalcone crashed out during reaction, the crude product was collected by filtration and washed twice with cool MeOH and the product was dried under high vacuum. If the product didn't precipitate, MeOH was evaporated and EtOAc was used to extract the product. The crude product was taken to the next step without further purification.

To a 100 mL Schlenk flask the corresponding chalcone was charged, then the Schlenk flask was evacuated and refilled with argon three times. The α -tetralone (1.0 equiv, 120 mmol) and anhydrous THF (0.5 mL/mmol, 60 mL) were added successively under argon. The solution was cooled to 0°C and HBF_4 (1.5 equiv., corresponding Et_2O complex, 180 mmol, 28.116 mL) was added dropwise through an addition funnel under stirring, during which the reaction mixture became dark. The reaction mixture was heated to reflux (temperature of oil bath: 85°C) for 12 h, during which the pyrylium salt crashed out. After cooling down to room temperature, the pyrylium salt was collected by filtration, washed with Et_2O three times and dried under high vacuum to yield an orange solid (12.9022g, 24%).

Synthesis of Pyridinium Salts –

The following pyridinium salts have been previously reported.^{7–14}



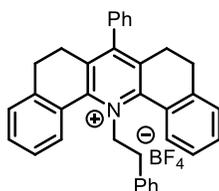
General Procedure A:

Primary amine (1.2 equiv) was added to a suspension of 2,4,6-triphenylpyrylium tetrafluoroborate (1.0 equiv) and EtOH (1.0 M) in a round-bottomed flask. The flask was fitted with a reflux condenser. The mixture was stirred and heated at reflux in an oil bath at 80–85°C for 4 h. The mixture was then allowed to cool to room temperature. If product precipitation occurred during reflux, the solid was filtered, washed with Et₂O, and dried under high vacuum. If product precipitation did not occur during reflux, the solution was diluted with Et₂O (2–3x volume of EtOH used), sonicated then vigorously stirred for 1 h to induce trituration. The resulting solid pyridinium salt was filtered and washed with Et₂O. If the salt did not precipitate, it was subjected to silica gel flash chromatography with acetone/DCM.

The corresponding amine hydrochloride salts can also be used using the following modified procedure: Et₃N (1.2 equiv) was added to a mixture of the corresponding alkyl ammonium hydrochloride salt (1.2 equiv) and EtOH (1.0 M). After stirring the mixture for 30 min at room temperature, 2,4,6-triphenylpyrylium tetrafluoroborate (1 equiv) was added. From this point forward, the same procedure was followed as for alkyl amines described above; however prior to washing the solid product with Et₂O, the mixture was washed with water to remove Et₃N·HCl.

General Procedure B:

The α -primary pyridinium salts were synthesized in accordance to a modified literature procedure.¹⁵ The alkyl amine (1.0 equiv.) was added to a suspension of Perylium-2 (1.0 equiv.), powdered activated 4Å molecular sieves (~500 mg/mmol), and DCM (1 M) in a round-bottomed flask equipped with a stir bar. Acetic acid (1 drop) was added and the mixture was allowed to stir for 4 h at room temperature. The mixture was filtered and subjected to silica gel flash chromatography (20% acetone/DCM). Most of the products are yellow and fluorescent and can be monitored via TLC.



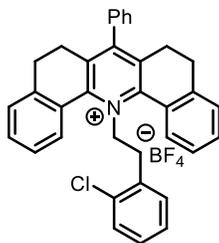
14-phenyl ethyl-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate:

Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (400 mg, 0.89 mmol) and commercially available phenylethylamine (0.112 mL, 0.89 mmol). Purified via automated flash chromatography to yield a yellow solid (416 mg, 85%).

¹H NMR (500 MHz, CDCl₃) δ 8.44 (dd, $J = 7.9, 1.2$ Hz, 2H), 7.70 – 7.58 (m, 3H), 7.55 – 7.45 (m, 4H), 7.38 (dd, $J = 7.6, 1.3$ Hz, 2H), 7.25 – 7.17 (m, 1H), 7.12 (t, $J = 7.6$ Hz, 2H), 7.00 (dd, $J = 12.3, 7.1$ Hz, 2H), 6.60 – 6.55 (m, 2H), 5.84 – 5.78 (m, 2H), 2.72 (dt, $J = 11.4, 3.2$ Hz, 2H), 2.63 – 2.51 (m, 6H), 2.30 – 2.14 (m, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 154.85, 153.36, 140.85, 137.09, 135.17, 134.97, 133.01, 130.10, 129.72, 129.62, 129.57, 128.92, 128.77, 128.56, 128.42, 128.21, 127.65, 127.27, 127.16, 65.17, 36.31, 27.73, 26.26.

¹⁹F NMR (376 MHz, CDCl₃) δ -153.11 (¹¹BF₄, minor), -153.16 (¹⁰BF₄, major).



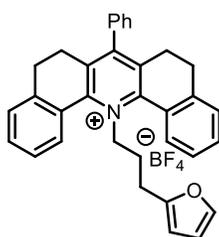
14-(chlorophenethyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium

tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (300 mg, 0.67 mmol) and 2-chlorophenethylamine (0.0942 mL, 0.67 mmol). Purified via automated flash chromatography to yield a yellow solid (320 mg, 82%).

¹H NMR (500 MHz, CDCl₃) δ 8.40 (dd, *J* = 7.9, 1.1 Hz, 2H), 7.69 – 7.59 (m, 3H), 7.55 – 7.44 (m, 4H), 7.36 (dd, *J* = 7.5, 1.2 Hz, 2H), 7.24 (d, *J* = 7.6 Hz, 1H), 7.20 – 7.12 (m, 2H), 7.08 – 6.99 (m, 2H), 6.65 – 6.60 (m, 1H), 5.91 (dd, *J* = 7.0, 5.5 Hz, 2H), 2.73 – 2.67 (m, 4H), 2.70 – 2.60 (m, 2H), 2.55 (td, *J* = 15.2, 4.8 Hz, 2H), 2.25 (td, *J* = 15.8, 5.1 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 155.35, 153.28, 141.27, 137.58, 135.05, 134.40, 133.07, 132.91, 131.04, 130.07, 129.85, 129.70, 129.60, 129.41, 128.72, 128.53, 128.11, 127.37, 127.23, 62.48, 34.94, 27.82, 26.47.

¹⁹F NMR (470 MHz, CDCl₃) δ -153.03 (¹¹BF₄, minor), -153.09 (d, *J* = 2.3 Hz) (¹⁰BF₄, major).



14-(3-(furan-2-yl)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium

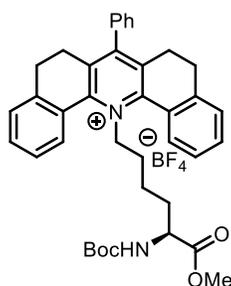
tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (372 mg, 0.83 mmol) and commercially available 3-(furan-2-yl)propan-1-amine (0.10 mL, 0.83 mmol). Purified via automated flash chromatography (gradient 0--> 10% --> 20% DCM/acetone) to yield a yellow solid (190.9 mg, 41%).

¹H NMR (500 MHz, CDCl₃) δ 8.31 (d, *J* = 6.8 Hz, 2H), 7.65 – 7.57 (m, 3H), 7.57 – 7.46 (m, 4H), 7.38 (dd, *J* = 7.6, 1.3 Hz, 2H), 7.12 (br s, 1H), 7.06 – 7.02 (m, 1H), 6.10 (dd, *J* = 3.2, 1.8 Hz, 1H), 5.73 – 5.68 (m, 1H), 5.41 (t, *J* = 6.9 Hz, 2H), 2.85 (m, 6H), 2.38 (m, 2H), 2.17 (t, *J* = 6.9 Hz, 2H), 1.70 (p, *J* = 6.9 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 155.44, 153.43, 152.95, 141.18, 141.09, 137.76, 135.24, 132.91, 129.77, 129.64, 129.57, 128.67, 128.48, 128.40, 127.70, 110.42, 106.00, 63.46, 28.56, 28.13, 26.71, 24.44.

¹⁹F NMR (470 MHz, CDCl₃) δ -153.13 (¹¹BF₄, minor), -153.18 (¹⁰BF₄, major).

IR (CDCl₃): 2920.29, 2850.14, 1503.62, 1468.84, 1293.92, 1188.94, 1056.40, 728.97 cm⁻¹.



14-(5-((*tert*-butoxycarbonyl)amino)-6-methoxy-6-oxohexyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate.

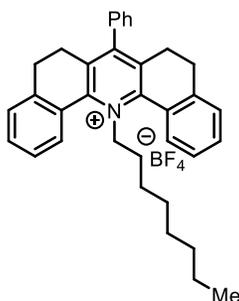
Synthesized according to General Procedure A from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (233 mg, 0.52 mmol) and commercially available BocLysOMe acetate salt (0.20g, 0.624 mmol). Purified via flash chromatography using DCM/acetone as a gradient to yield a yellow solid (248.9 mg, 69%).

¹H NMR (500 MHz, CDCl₃) δ 8.32 (dd, *J* = 8.0, 1.2 Hz, 2H), 7.64 (td, *J* = 7.4, 1.3 Hz, 3H), 7.53 (tt, *J* = 7.4, 2.0 Hz, 5H), 7.40 (dd, *J* = 7.6, 1.3 Hz, 2H), 7.11 (br s, 1H), 5.41 – 5.31 (m, 2H), 4.94 (m, 1H), 3.98 – 3.93 (m, 1H), 3.63 (s, 3H), 2.92-2.82 (m, 6H), 2.37 (m, 2H), 1.61 (m, 2H), 1.38 (s, 9H), 1.31 (m, 1H), 0.83 (m, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 172.74, 155.61, 153.40, 141.15, 137.79, 135.22, 132.98, 129.76, 129.63, 129.56, 128.70, 128.61, 128.42, 80.05, 64.42, 53.20, 52.47, 31.81, 31.06, 29.71, 28.42, 28.11, 26.69, 22.22.

¹⁹F NMR (470 MHz, CDCl₃) δ -152.97 (¹¹BF₄, minor), -153.02 (¹⁰BF₄, major).

IR (CDCl₃): 2954.29, 1740.20, 1707.39, 1537.39, 1366.15, 1162.59, 1056.20, 913.99, 772.52, 729.13, 596.80 cm⁻¹.



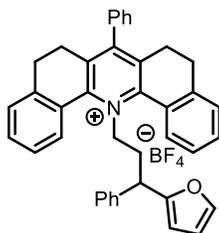
14-(*n*-octyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate:

Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (300 mg, 0.67 mmol) and commercially available octylamine (0.104 mL, 0.67 mmol). Purified via flash chromatography using DCM/acetone as a gradient to yield a yellow solid (328 mg, 88%).

¹H NMR (500 MHz, CDCl₃) δ 8.40 (dd, *J* = 8.0, 1.1 Hz, 2H), 7.64 (td, *J* = 7.7, 1.3 Hz, 3H), 7.57 – 7.47 (m, 4H), 7.39 (dd, *J* = 7.6, 1.3 Hz, 3H), 7.13 (s, 1H), 5.38 (t, *J* = 7.0 Hz, 2H), 2.84 (d, *J* = 5.3 Hz, 5H), 2.38 (s, 2H), 1.31 (p, *J* = 7.2 Hz, 2H), 1.20 – 1.08 (m, 2H), 1.07 – 0.84 (m, 6H), 0.79 (t, *J* = 7.3 Hz, 3H), 0.71 (ddd, *J* = 14.7, 8.4, 6.3 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 155.21, 153.48, 140.87, 137.58, 135.10, 132.91, 129.98, 129.63, 128.75, 128.46, 127.59, 64.57, 31.56, 30.07, 28.84, 28.44, 28.18, 26.64, 25.79, 22.56, 14.10.

¹⁹F NMR (470 MHz, CDCl₃) δ -153.29 (¹¹BF₄, minor), -153.35 (¹⁰BF₄, major).



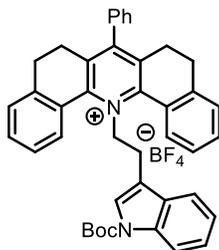
14-(3-(furan-2-yl)3-phenylpropyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (500 mg, 1.11 mmol) and commercially

available 3-(furan-2-yl)-3-phenylpropan-1-amine (223 mg, 1.11 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (387 mg, 55%).

¹H NMR (500 MHz, CDCl₃) δ 8.27 (s, 1H), 8.21 (s, 1H), 7.66 (s, 1H), 7.58 – 7.50 (m, 4H), 7.47 (t, *J* = 7.5 Hz, 3H), 7.35 (d, *J* = 7.4 Hz, 2H), 7.15 – 7.04 (m, 5H), 6.84 – 6.77 (m, 2H), 6.07 (dd, *J* = 3.3, 1.8 Hz, 1H), 5.75 (d, *J* = 3.2 Hz, 1H), 5.48 – 5.32 (m, 2H), 3.28 (t, *J* = 7.6 Hz, 1H), 2.86 – 2.77 (m, 5H), 2.35 (s, 2H), 2.19 – 2.06 (m, 1H), 1.91 (dq, *J* = 14.5, 7.3 Hz, 1H).

¹³C NMR (126 MHz, CDCl₃) δ 155.46, 154.86, 153.57, 153.11, 141.63, 141.02, 140.21, 137.79, 135.15, 132.89, 129.71, 129.63, 128.87, 128.59, 128.45, 128.15, 127.65, 127.30, 127.21, 110.40, 106.38, 62.25, 42.17, 35.01, 28.06, 26.66.

¹⁹F NMR (376 MHz, CDCl₃) δ -152.95 (¹¹BF₄, minor), -153.01 (¹⁰BF₄, major).



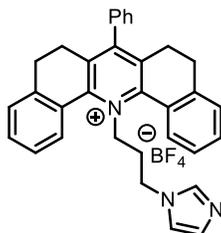
14-(2-(1-(*tert*-butoxycarbonyl)-1*H*-indol-3-yl)ethyl)-7-phenyl-5,6,8,9-

tetrahydrodibenzo[*c,h*]acridin-14-ium: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (500 mg, 1.11 mmol) and commercially available tryptamine (179 mg, 1.11 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid. This solid (300 mg, 0.51 mmol) was dissolved in MeCN (1 mL), and DMAP (0.62 mmol, 1.2 equiv) and Boc₂O (1.01 mmol, 2 equiv) were added. The reaction was stirred for two hours, then purified via flash chromatography to yield an orange solid (272 mg, 77%).

¹H NMR (500 MHz, CDCl₃) δ 8.61 (dd, *J* = 8.0, 1.1 Hz, 2H), 8.19 – 8.14 (m, 3H), 7.97 (s, 1H), 7.72 (td, *J* = 7.7, 1.3 Hz, 2H), 7.57 – 7.46 (m, 6H), 7.28 – 7.17 (m, 1H), 7.04 – 6.99 (m, 1H), 6.96 (s, 1H), 6.89 (s, 1H), 6.78 – 6.72 (m, 1H), 6.61 – 6.56 (m, 2H), 5.98 (t, *J* = 6.1 Hz, 2H), 2.74 – 2.61 (m, 2H), 2.54 (ddd, *J* = 21.6, 15.5, 4.0 Hz, 4H), 2.20 (td, *J* = 15.7, 4.5 Hz, 2H), 2.11 – 2.01 (m, 2H), 1.68 (s, 9H).

^{13}C NMR (126 MHz, CDCl_3) δ 154.54, 153.25, 149.45, 140.87, 137.05, 135.07, 132.98, 130.04, 129.79, 129.54, 129.47, 128.95, 128.61, 128.40, 127.97, 127.42, 126.97, 125.10, 123.94, 123.21, 118.52, 115.24, 114.07, 84.69, 63.38, 28.35, 27.47, 26.47, 26.27.

^{19}F NMR (376 MHz, CDCl_3) δ -152.93 ($^{11}\text{BF}_4$, minor), -152.98 ($^{10}\text{BF}_4$, major).

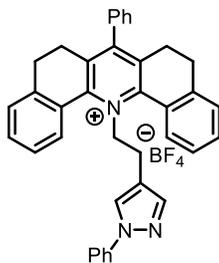


14-(3-(1*H*-imidazol-1-yl)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (250 mg, 0.56 mmol) and 3-(1*H*-imidazol-1-yl)propan-1-amine (0.0665 mL, 0.56 mmol). Purified via flash chromatography using DCM/acetone as a gradient to yield a yellow solid (162.8 mg, 35%).

^1H NMR (500 MHz, CDCl_3) δ 8.08 – 8.00 (m, 2H), 7.76 – 7.68 (m, 1H), 7.62 (s, 2H), 7.50 (qt, J = 6.9, 3.5 Hz, 6H), 7.42 – 7.35 (m, 2H), 7.01 (d, J = 7.1 Hz, 1H), 6.88 – 6.78 (m, 2H), 5.30 – 5.19 (m, 2H), 4.13 – 4.00 (m, 2H), 3.82 (t, J = 6.5 Hz, 2H), 3.11 (t, J = 13.6 Hz, 2H), 2.85 (d, J = 15.1 Hz, 2H), 2.74 (d, J = 13.2 Hz, 2H), 2.30 (t, J = 14.0 Hz, 2H), 2.00 – 1.90 (m, 2H).

^{13}C NMR (126 MHz, CDCl_3) δ 156.44, 152.86, 141.93, 138.30, 136.32, 135.33, 132.94, 129.88, 129.44, 129.18, 128.86, 128.51, 128.25, 128.17, 126.93, 126.30, 119.87, 61.16, 44.83, 31.06, 30.77, 29.41, 27.77, 26.72.

^{19}F NMR (470 MHz, CDCl_3) δ -151.46 ($^{11}\text{BF}_4$, minor), -151.51 ($^{10}\text{BF}_4$, major).

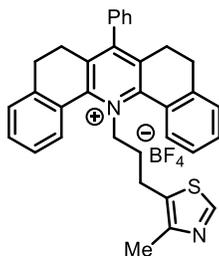


7-phenyl-14-(2-(1-phenyl-1*H*-pyrazol-4-yl)ethyl)-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (500 mg, 1.11 mmol) and commercially available 2-(1-phenyl-1*H*-pyrazol-4-yl)ethan-1-amine (207 mg, 1.11 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (518 mg, 85%).

¹H NMR (500 MHz, CDCl₃) δ 8.41 (d, *J* = 7.9 Hz, 2H), 7.70 (td, *J* = 7.7, 1.4 Hz, 2H), 7.57 – 7.50 (m, 4H), 7.50 – 7.40 (m, 6H), 7.36 (d, *J* = 7.5 Hz, 2H), 7.30 (t, *J* = 7.5 Hz, 1H), 7.03 (d, *J* = 6.5 Hz, 2H), 6.94 (s, 1H), 5.74 (t, *J* = 6.3 Hz, 2H), 2.77 – 2.63 (m, 6H), 2.57 (t, *J* = 6.2 Hz, 2H), 2.30 (d, *J* = 17.6 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 155.15, 153.35, 141.03, 140.34, 139.55, 137.34, 134.86, 133.00, 129.79, 129.73, 129.53, 129.45, 128.75, 128.67, 128.44, 128.21, 127.31, 127.13, 126.79, 126.26, 118.64, 116.94, 65.43, 27.73, 26.26, 25.65.

¹⁹F NMR (471 MHz, CDCl₃) δ -151.94 (¹¹BF₄, minor), -151.99 (¹⁰BF₄, major).



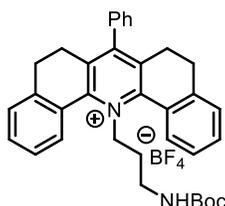
14-(3-(4-methylthiazol-5-yl)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (400 mg, 0.89 mmol) and commercially available 3-(4-methylthiazol-5-yl)propan-1-amine (139 mg, 0.89 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (323 mg, 62%).

¹H NMR (500 MHz, CDCl₃) δ 8.29 (d, *J* = 7.9 Hz, 2H), 7.68 – 7.56 (m, 4H), 7.56 – 7.46 (m, 5H), 7.37 (d, *J* = 7.4 Hz, 2H), 7.11 (s, 1H), 6.58 (d, *J* = 1.3 Hz, 1H), 5.53 (t, *J* = 7.2 Hz, 2H), 2.93 (d, *J* = 9.6 Hz, 2H), 2.80 (s, 4H), 2.35 (d, *J* = 15.3 Hz, 2H), 2.21 (s, 3H), 1.84 (p, *J* = 7.0 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 175.35, 167.84, 155.72, 153.32, 151.33, 150.05, 147.83, 141.26, 137.96, 137.94, 137.88, 135.30, 135.22, 135.09, 132.97, 129.61, 129.59, 129.51, 129.01, 128.88,

128.74, 128.62, 128.56, 128.47, 128.26, 127.93, 127.77, 127.69, 127.59, 127.30, 127.12, 125.40, 121.77, 113.88, 62.82, 51.88, 30.64, 29.69, 28.78, 28.36, 28.22, 27.99, 26.60, 26.44, 26.12, 25.88, 24.60, 16.44, 13.03.

^{19}F NMR (471 MHz, CDCl_3) δ -152.15 ($^{11}\text{BF}_4$, minor), -152.20 ($^{10}\text{BF}_4$, major).

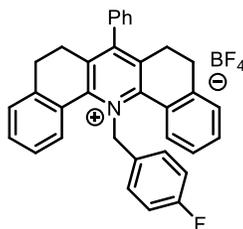


14 **-(3-((*tert*-butoxycarbonyl)amino)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate:** Synthesized according to General Procedure A from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (500 mg, 1.11 mmol) and commercially available *tert*-butyl (3-aminopropyl)carbamate (0.194 mL, 1.11 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (553 mg, 83%).

^1H NMR (500 MHz, CDCl_3) δ 8.17 (dd, $J = 7.9, 1.3$ Hz, 2H), 7.67 (s, 2H), 7.61 (td, $J = 7.8, 1.5$ Hz, 3H), 7.55 (td, $J = 7.5, 1.2$ Hz, 3H), 7.45 – 7.42 (m, 2H), 7.07 (s, 1H), 5.38 (t, $J = 7.3$ Hz, 2H), 4.95 (s, 1H), 3.10 (dd, $J = 14.6, 6.6$ Hz, 3H), 2.97 – 2.76 (m, 3H), 2.72 (q, $J = 6.4$ Hz, 2H), 2.36 (s, 2H), 1.59 (p, $J = 6.7$ Hz, 2H), 1.29 (s, 9H).

^{13}C NMR (126 MHz, CDCl_3) δ 156.30, 156.00, 153.33, 141.50, 137.91, 135.25, 132.89, 129.47, 129.25, 128.74, 128.42, 128.36, 127.14, 79.01, 62.59, 37.10, 30.78, 28.41, 27.88, 26.61.

^{19}F NMR (470 MHz, CDCl_3) δ -152.60 ($^{11}\text{BF}_4$, minor), -152.66 ($^{10}\text{BF}_4$, major).



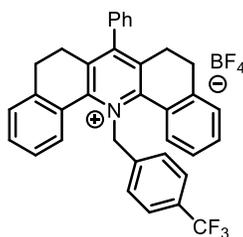
14-(4-fluorobenzyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-

tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (400 mg, 0.89 mmol) and commercially available 4-fluorobenzylamine (0.102 mL, 0.89 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (420 mg, 85%).

¹H NMR (500 MHz, CDCl₃) δ 8.46 (dd, *J* = 7.9, 1.2 Hz, 2H), 7.63 (td, *J* = 7.7, 1.3 Hz, 3H), 7.53 (d, *J* = 7.9 Hz, 3H), 7.42 (td, *J* = 7.5, 1.1 Hz, 2H), 7.23 (dd, *J* = 7.6, 1.3 Hz, 2H), 7.09 (s, 1H), 6.68 – 6.60 (m, 2H), 6.49 – 6.41 (m, 2H), 6.30 (s, 2H), 2.78 (d, *J* = 15.7 Hz, 2H), 2.72 – 2.55 (m, 4H), 2.31 (d, *J* = 17.4 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 163.25, 161.27, 156.12, 153.98, 141.13, 137.66, 135.28, 134.93, 133.01, 131.03, 131.00, 130.33, 129.63, 129.57, 128.66, 128.26, 128.21, 127.36, 126.09, 115.50 (d, *J* = 21.6 Hz), 67.18, 27.92, 26.40.

¹⁹F NMR (282 MHz, CDCl₃) δ -110.97 (dq, *J* = 8.7, 4.3 Hz), -151.84 (¹¹BF₄, minor), -151.88 (¹⁰BF₄, major).



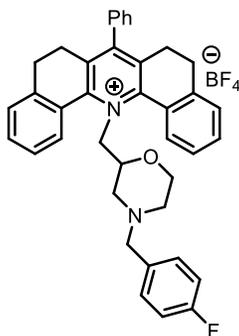
7-phenyl-14-(4-(trifluoromethyl)benzyl)-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium

tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (400 mg, 0.89 mmol) and commercially available 4-trifluoromethylbenzylamine (0.126 mL, 0.89 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (426 mg, 79%).

¹H NMR (500 MHz, CDCl₃) δ 8.51 (dd, *J* = 8.0, 1.1 Hz, 2H), 7.65 (td, *J* = 7.7, 1.3 Hz, 3H), 7.54 (t, *J* = 6.8 Hz, 2H), 7.43 (td, *J* = 7.5, 1.1 Hz, 2H), 7.25 – 7.19 (m, 5H), 7.11 (s, 1H), 6.64 (d, *J* = 8.1 Hz, 2H), 6.45 (s, 2H), 2.80 (s, 2H), 2.69 – 2.55 (m, 4H), 2.33 (s, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 156.33, 154.06, 141.17, 138.98, 137.79, 134.90, 133.19, 130.37, 129.71, 129.66, 128.79, 128.21, 128.09, 128.00, 127.44, 125.40 (q, *J* = 3.9 Hz), 66.91, 27.90, 26.51.

^{19}F NMR (282 MHz, CDCl_3) δ -62.08, -151.40 ($^{11}\text{BF}_4$, minor), -151.45 ($^{10}\text{BF}_4$, major).



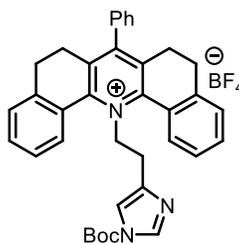
14-((4-(4-fluorobenzyl)morpholin-2-yl)methyl)-7-phenyl-5,6,8,9-

tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (400 mg, 0.89 mmol) and commercially available (4-(4-fluorobenzyl)morpholin-2-yl)methanamine (0.175 mL, 0.89 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (422 mg, 72%).

^1H NMR (500 MHz, CDCl_3) δ 8.52 (d, $J = 7.9$ Hz, 1H), 8.36 (d, $J = 7.9$ Hz, 1H), 7.67 (ddd, $J = 25.5, 12.2, 5.2$ Hz, 3H), 7.58 – 7.46 (m, 4H), 7.38 (t, $J = 8.5$ Hz, 2H), 7.28 (d, $J = 7.9$ Hz, 1H), 7.13 (t, $J = 4.2$ Hz, 1H), 7.09 – 7.01 (m, 2H), 6.95 – 6.87 (m, 2H), 5.91 (dd, $J = 14.8, 3.1$ Hz, 1H), 5.11 (dd, $J = 14.8, 9.9$ Hz, 1H), 3.39 (dt, $J = 11.3, 3.0$ Hz, 1H), 3.32 (d, $J = 13.1$ Hz, 1H), 3.28 – 3.18 (m, 2H), 3.17 – 3.09 (m, 2H), 2.80 (td, $J = 15.1, 10.8$ Hz, 6H), 2.45 – 2.26 (m, 4H), 1.87 (ddd, $J = 12.6, 10.4, 3.2$ Hz, 1H), 1.68 (dd, $J = 11.5, 8.6$ Hz, 1H).

^{13}C NMR (126 MHz, CDCl_3) δ 163.19, 161.24, 155.62, 155.35, 153.10, 140.97, 140.24, 137.28, 136.41, 135.19, 133.05, 132.92, 132.55, 131.36, 130.78, 130.72, 129.89, 129.68, 129.51, 129.34, 129.29, 129.01, 128.47, 128.41, 128.29, 127.60, 127.45, 115.23 (d, $J = 21.2$ Hz), 73.79, 66.24, 65.75, 61.84, 55.21, 51.66, 29.82, 28.38, 28.09, 26.62, 26.47.

^{19}F NMR (471 MHz, CDCl_3) δ -114.48, -152.03 ($^{11}\text{BF}_4$, minor), -152.08 ($^{10}\text{BF}_4$, major).



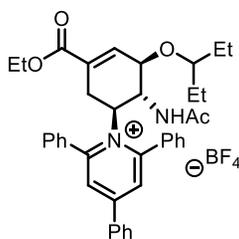
14-(2-(1-(*tert*-butoxycarbonyl)-1*H*-imidazol-4-yl)ethyl)-7-phenyl-5,6,8,9-

tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate: Synthesized according to General Procedure B from 7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]xanthen-14-ium tetrafluoroborate (500 mg, 1.11 mmol) and commercially available histamine (123 mg, 1.11 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (450 mg, 75%). Then the Katritzky salt (250 mg, 0.46 mmol) was dissolved in MeCN (1 mL), followed by the addition of DMAP (0.55 mmol, 1.2 equiv) and Boc₂O (0.92 mmol, 2 equiv). The reaction was stirred until starting material was consumed and then purified via flash chromatography (DCM/acetone) to yield 226 mg (76%).

¹H NMR (500 MHz, CDCl₃) δ 8.36 (dd, *J* = 7.9, 1.2 Hz, 2H), 7.76 (d, *J* = 1.3 Hz, 1H), 7.63 (td, *J* = 7.7, 1.3 Hz, 2H), 7.52 – 7.48 (m, 5H), 7.38 (dd, *J* = 7.6, 1.3 Hz, 3H), 7.17 – 7.06 (m, 2H), 6.80 (d, *J* = 1.3 Hz, 1H), 5.69 (t, *J* = 6.4 Hz, 2H), 2.80 – 2.67 (m, 6H), 2.57 (t, *J* = 6.3 Hz, 2H), 2.37 – 2.24 (m, 2H), 1.59 (s, 9H).

¹³C NMR (126 MHz, CDCl₃) δ 155.00, 153.60, 146.74, 141.54, 140.89, 138.10, 137.21, 136.47, 135.27, 132.97, 129.83, 129.57, 128.80, 128.56, 128.46, 114.90, 86.48, 63.78, 28.17, 27.96, 27.84, 26.41.

¹⁹F NMR (376 MHz, CDCl₃) δ -152.53 (¹¹BF₄, minor), -152.58 (¹⁰BF₄, major).



1-((1*S*,5*R*,6*R*)-6-acetamido-3-(ethoxycarbonyl)-5-(pentan-3-yloxy)cyclohex-3-en-1-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate: Synthesized according to General Procedure A from 2,4,6-triphenylpyrylium tetrafluoroborate (396 mg, 1.0 mmol) and commercially available Tamiflu phosphate (492 mg, 1.2 mmol). Purified via automated flash chromatography (DCM/acetone) to yield a yellow solid (152 mg, 22%).

¹H NMR (500 MHz, CDCl₃) δ 7.94 (d, *J* = 2.4 Hz, 1H), 7.91 – 7.77 (m, 5H), 7.68 (d, *J* = 6.9 Hz, 2H), 7.63 – 7.38 (m, 8H), 6.46 (q, *J* = 1.9 Hz, 1H), 5.43 (ddd, *J* = 11.3, 9.2, 7.3 Hz, 1H), 4.41 –

4.34 (m, 1H), 4.15 (q, $J = 7.2$ Hz, 2H), 3.95 (dt, $J = 11.3, 7.9$ Hz, 1H), 3.27 (p, $J = 5.8$ Hz, 1H), 3.09 – 2.95 (m, 2H), 1.90 (s, 3H), 1.42 – 1.29 (m, 4H), 1.26 (t, $J = 7.1$ Hz, 4H), 0.99 – 0.83 (m, 2H), 0.78 (dt, $J = 14.4, 7.4$ Hz, 6H).

^{13}C NMR (126 MHz, CDCl_3) δ 172.77, 165.23, 160.98, 159.56, 155.90, 139.90, 133.45, 133.42, 133.31, 132.95, 132.16, 131.61, 130.53, 130.08, 129.90, 129.79, 128.49, 126.20, 125.86, 83.19, 73.35, 68.89, 61.25, 56.05, 32.31, 26.41, 26.21, 25.82, 23.04, 14.28, 9.60, 9.28, 8.68.

^{19}F NMR (470 MHz, CDCl_3) δ -150.62 ($^{11}\text{BF}_4$, minor), -150.67 ($^{10}\text{BF}_4$, major).

4. Product synthesis and characterization:

General Procedure C:

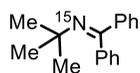
To an oven-dried silicate tube containing a stir bar was added the redox-active imine (1 equiv), $[\text{Ir}(\text{dFCF}_3\text{ppy})_2\text{dtbbpy}]\text{PF}_6$ (1 mol%), potassium persulfate (1.1 equiv), and 3Å molecular sieves (~25mg, 3 or 4 balls). The vial was taken to nitrogen glovebox, where potassium phosphate tribasic (1 equiv) was added. Then, the vial was brought into a nitrogen wetbox and pivalonitrile (0.25M) was added, followed by ^{15}N -benzophenone imine (3 equiv). The reaction was sealed with Teflon and taken out of the glovebox, placed in front of one 456 nm light (~3 cm away) and irradiated for 24 hours. After the reaction, the color should be a light orange. This suspension was filtered over a pad of celite and concentrated *in vacuo*, then purified by flash chromatography or preparatory TLC (see notes on chromatography in General Information).

General Procedure D:

To an oven-dried silicate tube containing a stir bar was added the benzylic Katritzky salt (1 equiv), $\text{Ru}(\text{bpy})_3(\text{PF}_6)_2$ (1 mol%), and 3Å molecular sieves (~25mg, 3 or 4 balls). The vial was taken to a nitrogen glovebox, where DCE (0.1M) and ^{15}N -benzophenone imine (3 equiv) were added. The vial was sealed and wrapped with Teflon, then irradiated with one 456 nm Kessil lamp (~3 cm away) for 6 hours. After completion, the red liquid was concentrated *in vacuo* and subjected to flash chromatography or preparatory TLC (see notes on chromatography in General Information).

General Procedure E:

To an oven-dried silicate tube containing a stir bar was added Pyr1 or Pyr2 (1 equiv), CuI (20 mol%), and 3Å molecular sieves (~25mg, 3 or 4 balls). The vial was taken to a nitrogen dry-box, where Cs₂CO₃ (2 equiv) was added. Then, the vial was taken to a nitrogen wet-box and DMF (0.4M) was added. To this solution TMHD (30 mol%) was added and the reaction was stirred for 30 seconds. During this time, the reaction media should turn dark red (for reactions using Pyr1) or dark yellow/red (for reactions using Pyr2). Then, ¹⁵N-benzophenone imine (3 equiv) was added, and the vial was sealed and wrapped in Teflon. The reaction vial was taken out of the glovebox and irradiated by two 456 nm Kessil lamps (~1 cm away from both lamps) and irradiated for 16 hours. After irradiation, the solids were filtered through celite and rinsed with toluene (4-5x reaction volume). This solution was concentrated *in vacuo* (rotovap bath temperature: 53°C), and more toluene was added and concentrated again to remove mostly all DMF. The residue was then subjected to flash chromatography or preparatory TLC (see notes on chromatography in General Information).



(3a) *N-tert-butyl-1,1-diphenylmethanimine-¹⁵N*: Synthesized according to General Procedure C from *N-tert-butyl-1-(2,4,6-trimethoxyphenyl)methanimine* (62.8 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 31.2 mg (53%) of a clear oil.

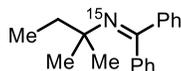
R_f: 0.64 (4% EtOAc/hexanes, 1 drop Et₃N).

¹H NMR (400 MHz, CDCl₃) δ 7.42 (dd, *J* = 8.1, 1.7 Hz, 2H), 7.29 – 7.25 (m, 3H), 7.21 – 7.13 (m, 3H), 7.10 – 7.04 (m, 2H), 1.06 (d, *J* = 2.1 Hz, 9H).

¹³C NMR (101 MHz, CDCl₃) δ 163.70, 142.16, 140.06, 132.55, 130.21, 129.44, 128.59, 128.43, 128.20, 128.00, 127.93, 127.59, 127.32, 57.12, 31.73.

¹⁵N NMR (51 MHz, CDCl₃) δ 354.51.

HRMS: ASAP (positive) M = C₁₇H₁₉N¹⁵: calculated (M+H)⁺ m/z 239.1566; found (M+H)⁺ m/z 239.1577.



(3b) *N*-tert-pentyl-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-tert-pentyl-1-(2,4,6-trimethoxyphenyl)methanimine (66.3 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 35.6 mg (57%) of a clear oil.

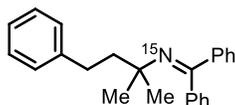
R_f: 0.62 (3% EtOAc/hexanes, 1 drop Et₃N).

¹H NMR (500 MHz, CDCl₃) δ 7.57 – 7.52 (m, 2H), 7.40 – 7.35 (m, 3H), 7.33 – 7.27 (m, 3H), 7.21 – 7.18 (m, 2H), 1.60 (qd, *J* = 7.4, 2.6 Hz, 2H), 1.04 (d, *J* = 1.8 Hz, 6H), 0.97 (t, *J* = 7.4 Hz, 3H).

¹³C NMR (101 MHz, CDCl₃) δ 163.60 (d, *J* = 6.8 Hz), 142.36 (d, *J* = 9.0 Hz), 140.31 (d, *J* = 2.6 Hz), 132.55, 130.21, 129.38, 128.51, 128.42, 128.18, 128.15, 127.98, 127.88, 127.56, 126.58, 59.45 (d, *J* = 2.0 Hz), 38.66 (d, *J* = 4.0 Hz), 28.49, 9.23 (d, *J* = 1.6 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 353.58.

HRMS: ASAP (positive) M = C₁₈H₂₁N¹⁵: calculated (M+H)⁺ m/z 254.1756; found (M+H)⁺ m/z 254.1766.



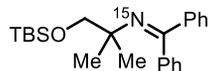
(3c) *N*-(2-methyl-4-phenylbutan-2-yl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(2-methyl-4-phenylbutan-2-yl)-1-(2,4,6-trimethoxyphenyl)methanimine (82.3 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 43.8 mg (53%) of a clear oil.

¹H NMR (500 MHz, CDCl₃) δ 7.60 – 7.57 (m, 2H), 7.41 – 7.38 (m, 3H), 7.35 – 7.27 (m, 5H), 7.25 – 7.20 (m, 4H), 7.20 – 7.16 (m, 1H), 2.84 – 2.77 (m, 2H), 1.95 – 1.87 (m, 2H), 1.11 (d, *J* = 1.7 Hz, 6H).

¹³C NMR (101 MHz, CDCl₃) δ 163.93 (d, *J* = 6.7 Hz), 143.69, 142.12, 140.11 (d, *J* = 2.8 Hz), 132.55, 130.21, 129.55, 128.58, 128.52, 128.44, 128.23, 128.20, 128.04, 128.03, 127.97, 125.62, 59.16 (d, *J* = 2.0 Hz), 48.74 (d, *J* = 4.3 Hz), 31.40 (d, *J* = 1.7 Hz), 29.85, 28.87.

¹⁵N NMR (51 MHz, CDCl₃) δ 352.04.

HRMS: ASAP (positive) M = C₂₄H₂₅N¹⁵: calculated (M+H)⁺ m/z 330.2069; found (M+H)⁺ m/z 330.2069.



(3d) *N*-(1-((*tert*-butyldimethylsilyloxy)-2-methylpropan-2-yl)-1,1-diphenylmethanimine-¹⁵N): Synthesized according to General Procedure C from *N*-(1-((*tert*-butyldimethylsilyloxy)-2-methylpropan-2-yl)-1-(2,4,6-trimethoxyphenyl)methanimine (95.2 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 71.7 mg (78%) of a clear oil.

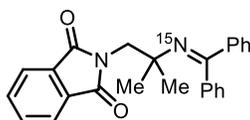
R_f: 0.51 (3% EtOAc/hexanes)

¹H NMR (400 MHz, CDCl₃) δ 7.43 – 7.39 (m, 2H), 7.28 – 7.24 (m, 3H), 7.19 – 7.12 (m, 3H), 7.11 – 7.07 (m, 2H), 3.46 (d, *J* = 1.4 Hz, 2H), 0.88 (d, *J* = 1.7 Hz, 6H), 0.77 (s, 9H), -0.09 (d, *J* = 8.2 Hz, 6H).

¹³C NMR (101 MHz, CDCl₃) δ 164.93 (d, *J* = 6.9 Hz), 142.06 (d, *J* = 8.8 Hz), 140.23 (d, *J* = 2.7 Hz), 130.21, 129.55, 128.90, 128.56, 128.39 (d, *J* = 7.6 Hz), 128.24 (d, *J* = 3.1 Hz), 127.98, 127.86, 127.65 (d, *J* = 1.4 Hz), 127.40, 127.32, 126.60, 73.20 (d, *J* = 8.5 Hz), 60.94, 26.09, 24.99, 18.43 (d, *J* = 6.7 Hz), -5.21.

¹⁵N NMR (51 MHz, CDCl₃) δ 347.28.

HRMS: ASAP (positive) M = C₂₃H₃₃N¹⁵OSi: calculated (M+H)⁺ m/z 369.2380; found (M+H)⁺ m/z 369.2384.



(3e) **2-(2-((diphenylmethylene)amino-¹⁵N)-2-methylpropyl)isoindoline-1,3-dione**: Synthesized according to General Procedure C from 2-(2-methyl-2-((2,4,6-trimethoxybenzylidene)amino)propyl)isoindoline-1,3-dione (79.2 mg, 0.20 mmol). Purified via preparatory TLC (30% EtOAc/hexanes) to afford 39.1 mg (51%) of a clear oil.

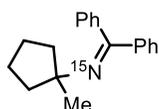
R_f: 0.50 (30% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.91 – 7.83 (m, 2H), 7.76 – 7.68 (m, 2H), 7.56 – 7.50 (m, 2H), 7.43 – 7.27 (m, 7H), 7.25 – 7.18 (m, 2H), 3.88 (d, *J* = 2.5 Hz, 2H), 1.09 (d, *J* = 1.5 Hz, 6H).

^{13}C NMR (126 MHz, CDCl_3) δ 169.09, 165.30, 148.14, 141.65, 139.76, 133.96, 132.48, 129.70, 128.50, 128.47, 128.45, 128.32, 128.15, 128.02, 127.97, 127.11, 125.97, 123.34, 61.27, 50.97 (d, $J = 6.4$ Hz), 27.07.

^{15}N NMR (51 MHz, CDCl_3) δ 344.28.

HRMS: ASAP (positive) $M = \text{C}_{25}\text{H}_{22}\text{N}^{15}\text{NO}_2$: calculated (M+H) $^+$ m/z 385.1763; found (M+H) $^+$ m/z 385.1769.



(3f) *N*-(1-methylcycloheptyl)-1,1-diphenylmethanimine- ^{15}N : Synthesized according to General Procedure C from *N*-(1-methylcycloheptyl)-1-(2,4,6-trimethoxyphenyl)methanimine (69.2 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 39.4 mg (60%) of a clear oil.

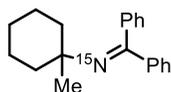
R_f: 0.7 (5% EtOAc/hexanes)

^1H NMR (500 MHz, CDCl_3) δ 7.55 – 7.52 (m, 2H), 7.42 – 7.36 (m, 3H), 7.34 – 7.30 (m, 1H), 7.30 – 7.27 (m, 2H), 7.22 – 7.19 (m, 2H), 1.82 – 1.74 (m, 2H), 1.72 – 1.60 (m, 2H), 1.59 – 1.43 (m, 4H), 1.12 (d, $J = 1.6$ Hz, 3H).

^{13}C NMR (101 MHz, CDCl_3) δ 164.40, 142.04 (d, $J = 8.5$ Hz), 140.22, 130.21, 129.47, 128.67, 128.28, 128.25, 128.01, 128.00, 127.87, 68.06 (d, $J = 1.7$ Hz), 41.76 (d, $J = 3.1$ Hz), 29.85, 26.78, 23.17.

^{15}N NMR (51 MHz, CDCl_3) δ 353.59.

HRMS: ASAP (positive) $M = \text{C}_{19}\text{H}_{21}\text{N}^{15}$: calculated (M+H) $^+$ m/z 266.1757; found (M+H) $^+$ m/z 266.1750.



(3g) *N*-(1-methylcyclohexyl)-1,1-diphenylmethanimine- ^{15}N : Synthesized according to General Procedure C from *N*-(1-methylcyclohexyl)-1-(2,4,6-trimethoxyphenyl)methanimine (82.3 mg,

0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 48.4 mg (70%) of a clear oil.

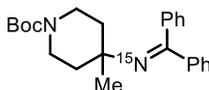
R_f: 0.61 (3% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.59 – 7.55 (m, 2H), 7.40 – 7.37 (m, 3H), 7.34 – 7.26 (m, 3H), 7.22 – 7.19 (m, 2H), 1.76 – 1.71 (m, 2H), 1.68 – 1.60 (m, 2H), 1.53 (td, *J* = 6.8, 2.8 Hz, 1H), 1.43 (dtd, *J* = 10.8, 6.3, 3.8 Hz, 2H), 1.32 – 1.25 (m, 3H), 1.05 (d, *J* = 1.4 Hz, 3H).

¹³C NMR (101 MHz, CDCl₃) δ 163.75 (d, *J* = 6.8 Hz), 142.38, 140.46 (d, *J* = 2.7 Hz), 132.55, 130.21, 129.42, 128.41 (d, *J* = 2.5 Hz), 128.22, 128.18, 128.14, 128.00, 127.93, 127.90, 58.75 (d, *J* = 2.1 Hz), 40.54 (d, *J* = 2.8 Hz), 29.85, 28.44, 26.20, 23.03.

¹⁵N NMR (51 MHz, CDCl₃) δ 353.83.

HRMS: ASAP (positive) *M* = C₂₀H₂₃N¹⁵: calculated (M+H)⁺ *m/z* 280.1913; found (M+H)⁺ *m/z* 280.1919.



(3h) tert-butyl 4-((diphenylmethylene)amino-¹⁵N)-4-methylpiperidine-1-carboxylate:

Synthesized according to General Procedure C from *tert*-butyl-4-methyl-4-((2,4,6-trimethoxybenzylidene)amino)piperidine-1-carboxylate (98 mg, 0.25 mmol). Purified via preparatory TLC (20% EtOAc/hexanes) to afford 50.9 mg (54%) of a clear oil.

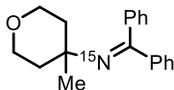
R_f: 0.7 (20% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.59 – 7.53 (m, 2H), 7.41 (dp, *J* = 4.9, 1.9 Hz, 3H), 7.36 – 7.32 (m, 1H), 7.29 (dd, *J* = 8.2, 6.5 Hz, 2H), 7.22 – 7.14 (m, 2H), 3.70 (s, 2H), 3.28 (s, 2H), 1.79 – 1.71 (m, 2H), 1.45 (s, 9H), 1.43 – 1.37 (m, 2H), 1.07 (d, *J* = 1.3 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 165.06 (d, *J* = 6.8 Hz), 155.22, 141.77 (d, *J* = 8.8 Hz), 139.95 (d, *J* = 2.7 Hz), 129.83, 128.27, 128.19, 128.18, 128.09, 128.08, 79.27, 56.89 (d, *J* = 1.9 Hz), 41.23, 39.72, 28.64, 28.08.

¹⁵N NMR (51 MHz, CDCl₃) δ 347.36.

HRMS: ASAP (positive) $M = C_{24}H_{30}N^{15}NO_2$: calculated $(M+H)^+$ m/z 380.2356; found $(M+H)^+$ m/z 380.2366.



(3i) *N*-(4-methyltetrahydro-2*H*-pyran-4-yl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(4-methyltetrahydro-2*H*-pyran-4-yl)-1-(2,4,6-trimethoxyphenyl)methanimine (58.6 mg, 0.20 mmol). Purified via preparatory TLC (85% pentane, 12% toluene, 3% EtOAc) to afford 32.3 mg (58%) of a clear oil.

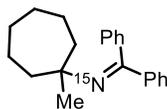
R_f: 0.30 (85% pentane, 12% toluene, 3% EtOAc)

¹H NMR (500 MHz, CDCl₃) δ 7.60 – 7.55 (m, 2H), 7.40 (ddd, $J = 4.4, 2.6, 1.4$ Hz, 3H), 7.37 – 7.33 (m, 1H), 7.32 – 7.27 (m, 2H), 7.21 – 7.14 (m, 2H), 3.82 (ddd, $J = 11.4, 9.8, 2.6$ Hz, 2H), 3.68 (dt, $J = 11.5, 4.2$ Hz, 2H), 1.79 – 1.72 (m, 2H), 1.61 – 1.51 (m, 2H), 1.12 (d, $J = 1.4$ Hz, 3H).

¹³C NMR (not all aryl resonances shown due to overlap) (126 MHz, CDCl₃) δ 164.80 (d, $J = 6.7$ Hz), 141.87 (d, $J = 8.9$ Hz), 140.08, 129.81, 128.22, 128.17, 128.09, 64.91, 56.34, 40.75 (d, $J = 2.7$ Hz), 29.85, 28.10.

¹⁵N NMR (51 MHz, CDCl₃) δ 349.18.

HRMS: ASAP (positive) $M = C_{19}H_{21}N^{15}O$: calculated $(M+H)^+$ m/z 281.1672; found $(M+H)^+$ m/z 281.1679.



(3j) *N*-(1-methylcycloheptyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(1-methylcycloheptyl)-1-(2,4,6-trimethoxyphenyl)methanimine (76.25 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 32.4 mg (45%) of a clear oil.

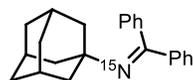
R_f: 0.68 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.58 – 7.54 (m, 2H), 7.39 – 7.36 (m, 3H), 7.32 – 7.27 (m, 3H), 7.20 – 7.18 (m, 2H), 1.90 – 1.84 (m, 2H), 1.74 – 1.65 (m, 2H), 1.62 – 1.57 (m, 3H), 1.54 – 1.40 (m, 5H), 0.96 (d, *J* = 1.3 Hz, 3H).

¹³C NMR (101 MHz, CDCl₃) δ 162.81 (d, *J* = 6.6 Hz), 142.42, 140.60 (d, *J* = 2.6 Hz), 129.35, 128.49, 128.41, 128.20, 128.17, 127.97, 127.83, 127.63, 62.36 (d, *J* = 2.1 Hz), 44.01 (d, *J* = 3.2 Hz), 29.87, 29.60, 23.48.

¹⁵N NMR (51 MHz, CDCl₃) δ 356.93.

HRMS: ASAP (positive) M = C₂₁H₂₅N¹⁵: calculated (M+H)⁺ m/z 294.2069; found (M+H)⁺ m/z 294.2066.



(3k) *N*-(adamantan-1-yl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(adamantan-1-yl)-1-(2,4,6-trimethoxyphenyl)methanimine (82.3 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 50.1 mg (64%) of a clear oil.

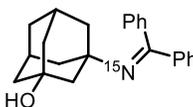
R_f: 0.63 (3% EtOAc/hexanes, 1 drop Et₃N).

¹H NMR (500 MHz, CDCl₃) δ 7.57 – 7.51 (m, 2H), 7.42 – 7.34 (m, 3H), 7.34 – 7.23 (m, 3H), 7.21 – 7.17 (m, 2H), 1.97 (p, *J* = 3.0 Hz, 3H), 1.74 (d, *J* = 2.9 Hz, 6H), 1.63 – 1.50 (m, 7H).

¹³C NMR (101 MHz, CDCl₃) δ 163.01 (d, *J* = 6.6 Hz), 142.38 (d, *J* = 8.7 Hz), 140.57 (d, *J* = 2.6 Hz), 129.38, 128.49, 128.21, 128.18, 127.98, 127.90, 127.82, 58.16, 44.36 (d, *J* = 1.7 Hz), 36.60, 29.92 (d, *J* = 1.5 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 356.00.

HRMS: ASAP (positive) M = C₂₃H₂₄N¹⁵: calculated (M+H)⁺ m/z 319.2103; found (M+H)⁺ m/z 319.2102.



(3l) 3-((diphenylmethylene)amino)adamantan-1-ol-¹⁵N: Synthesized according to General Procedure C from *N*-2,4,6-trimethoxybenzylidene)amino)adamantan-1-ol (69.0 mg, 0.2 mmol).

Purified via preparatory TLC (50:35:15 hexanes:EtOAc:toluene) to afford 31.9 mg (48%) of a clear oil.

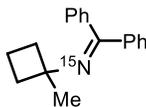
R_f: 0.53 (50:35:15 hexanes:EtOAc:toluene).

¹H NMR (500 MHz, CDCl₃) δ 7.55 – 7.52 (m, 2H), 7.38 (dd, *J* = 5.0, 2.0 Hz, 3H), 7.35 – 7.27 (m, 3H), 7.20 – 7.17 (m, 2H), 2.19 – 2.14 (m, 2H), 1.72 (s, 2H), 1.65 (d, *J* = 3.0 Hz, 4H), 1.62 – 1.54 (m, 6H), 1.42 (dd, *J* = 3.0, 1.4 Hz, 1H).

¹³C NMR (126 MHz, CDCl₃) δ 163.80 (d, *J* = 6.5 Hz), 142.02 (d, *J* = 8.8 Hz), 140.12, 129.63, 128.62, 128.48, 128.36, 128.23, 128.16, 128.02, 127.99, 127.58, 69.76, 61.00, 52.04, 44.29, 42.62, 35.04, 31.16.

¹⁵N NMR (51 MHz, CDCl₃) δ 350.91.

HRMS: ASAP (positive) M = C₂₃H₂₅N¹⁵O: calculated (M+H)⁺ m/z 333.1985; found (M+H)⁺ m/z 333.1992.



(3m) N-(1-methylcyclobutyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(1-methylcyclobutyl)-1-(2,4,6-trimethoxyphenyl)methanimine (52.6 mg, 0.20 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 21.9 mg (44%) of a clear oil.

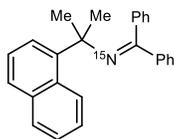
R_f: 0.67 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.56 – 7.51 (m, 2H), 7.37 (tt, *J* = 3.8, 2.7 Hz, 3H), 7.35 – 7.27 (m, 3H), 7.23 – 7.16 (m, 2H), 2.10 – 2.01 (m, 2H), 1.68 – 1.62 (m, 2H), 1.56 (dd, *J* = 10.6, 7.8 Hz, 2H), 1.48 (d, *J* = 2.1 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 163.56, 141.36, 138.94, 131.05, 129.62, 128.78, 128.43, 128.27, 128.07, 127.89, 62.19 (d, *J* = 2.4 Hz), 38.08, 29.85, 26.89, 14.27.

¹⁵N NMR (51 MHz, CDCl₃) δ 350.85.

HRMS: ASAP (positive) $M = C_{18}H_{19}N^{15}$: calculated $(M+H)^+$ m/z 252.1600; found $(M+H)^+$ m/z 252.1608.



(3n) *N*-(1,1-dimethylnaphthyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(1,1-dimethylnaphthyl)-1-(2,4,6-trimethoxyphenyl)methanimine (36.3 mg, 0.1 mmol). Purified via preparatory TLC (5% EtOAc/hexanes) to afford 27.6 mg (79%) of a clear oil.

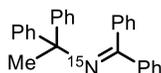
R_f: 0.7 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 8.38 – 8.33 (m, 1H), 7.78 (dd, $J = 7.8, 1.8$ Hz, 1H), 7.66 – 7.57 (m, 2H), 7.55 (d, $J = 8.2$ Hz, 1H), 7.40 (ddt, $J = 7.9, 6.8, 5.1$ Hz, 3H), 7.34 – 7.28 (m, 2H), 7.02 – 6.93 (m, 2H), 6.85 (dd, $J = 7.4, 1.2$ Hz, 1H), 6.77 (td, $J = 7.4, 1.2$ Hz, 2H), 6.06 (d, $J = 7.5$ Hz, 2H), 1.87 (d, $J = 2.5$ Hz, 6H).

¹³C NMR (126 MHz, CDCl₃) δ 166.08, 145.53, 141.46, 138.00, 134.81, 131.12 (d, $J = 16.6$ Hz), 129.66, 129.16, 129.01, 128.16, 128.06, 127.31, 127.01, 126.65, 125.12, 124.88, 122.87, 62.42, 32.33.

¹⁵N NMR (51 MHz, CDCl₃) δ 350.31.

HRMS: ASAP (positive) $M = C_{26}H_{23}N^{15}$: calculated $(M+H)^+$ m/z 351.1879; found $(M+H)^+$ m/z 351.1885.



(3o) *N*-(1,1-diphenylethyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(1,1-diphenylethyl)-1-(2,4,6-trimethoxyphenyl)methanimine (93.8 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 80.4 mg (90%) of a clear oil.

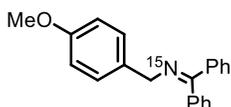
R_f: 0.65 (3% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.73 – 7.67 (m, 2H), 7.41 – 7.26 (m, 7H), 7.22 – 7.14 (m, 7H), 7.06 (dd, *J* = 8.4, 7.0 Hz, 2H), 6.56 – 6.51 (m, 2H), 1.68 (d, *J* = 1.4 Hz, 3H).

¹³C NMR (101 MHz, CDCl₃) δ 166.41, 150.96 (d, *J* = 2.2 Hz), 141.99 (d, *J* = 9.6 Hz), 138.72, 129.77, 128.31, 128.28, 127.93, 127.76, 127.71, 127.51, 127.20, 126.98, 125.83, 29.70, 29.11.

¹⁵N NMR (51 MHz, CDCl₃) δ 350.66.

HRMS: ASAP (positive) M = C₂₇H₂₄N¹⁵: calculated (M+H)⁺ *m/z* 363.1879; found (M+H)⁺ *m/z* 363.1870.



(4a) *N*-(4-methoxybenzyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure D from 1-(4-methoxybenzyl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (51.5 mg, 0.10 mmol). Purified via preparatory TLC (5% EtOAc/hexanes) to afford 26.6 mg (88%) of a clear oil.

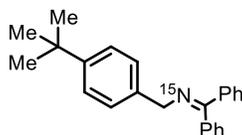
R_f: 0.35 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.71 – 7.65 (m, 2H), 7.53 – 7.41 (m, 3H), 7.40 – 7.37 (m, 1H), 7.35 – 7.30 (m, 2H), 7.26 – 7.19 (m, 4H), 6.90 – 6.84 (m, 2H), 4.55 (d, *J* = 0.9 Hz, 2H), 3.80 (s, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 168.58 (d, *J* = 5.5 Hz), 158.48, 139.97 (d, *J* = 8.3 Hz), 136.92, 132.98, 130.13, 128.88, 128.69, 128.67, 128.61, 128.42, 128.17, 127.96, 113.92, 57.01, 55.44.

¹⁵N NMR (51 MHz, CDCl₃) δ 322.38.

HRMS: ASAP (positive) M = C₂₁H₁₉N¹⁵O: calculated (M+H)⁺ *m/z* 303.1515; found (M+H)⁺ *m/z* 303.1520.



(4b) *N*-(4-*tert*butylbenzyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure D from 1-(4-*tert*butylbenzyl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (108.2

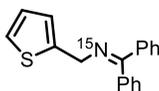
mg, 0.20 mmol). Purified via preparatory TLC (5% EtOAc/hexanes) to afford 24.2 mg (37%) of a clear oil.

R_f: 0.65 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.72 – 7.66 (m, 2H), 7.50 – 7.43 (m, 3H), 7.40 – 7.31 (m, 5H), 7.28 – 7.26 (m, 2H), 7.21 (dd, *J* = 8.0, 1.6 Hz, 2H), 4.58 (s, 2H), 1.32 (s, 9H).

¹³C NMR (126 MHz, CDCl₃) δ 168.64, 149.50, 139.99, 137.77, 136.93, 130.17 (d, *J* = 12.1 Hz), 128.70, 128.60, 128.17, 127.99, 127.51, 125.39, 57.31, 31.57.

¹⁵N NMR (51 MHz, CDCl₃) δ 322.08.



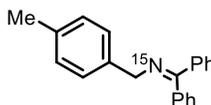
(4c) 1,1-diphenyl-*N*-(thiophen-2-ylmethyl)methanimine-¹⁵N: Synthesized according to General Procedure D from 2,4,6-triphenyl-1-(thiophen-2-ylmethyl)pyridin-1-ium tetrafluoroborate (122.8 mg, 0.25 mmol). Purified via flash chromatography with a gradient of 95:5:0 pentane/toluene/Et₂O → 90:3:2 pentane/toluene/Et₂O to afford 47.2 mg (68%).

¹H NMR (400 MHz, CDCl₃) δ 7.71 – 7.66 (m, 2H), 7.54 – 7.44 (m, 4H), 7.34 (dd, *J* = 8.2, 6.3 Hz, 2H), 7.24 – 7.20 (m, 3H), 6.96 (dd, *J* = 5.1, 3.5 Hz, 1H), 6.91 – 6.85 (m, 1H), 4.75 (s, 2H).

¹³C NMR (101 MHz, CDCl₃) δ 169.26 (d, *J* = 5.6 Hz), 157.68, 144.45, 139.66 (d, *J* = 8.3 Hz), 136.51 (d, *J* = 2.5 Hz), 132.55, 130.37, 130.21, 129.24 (d, *J* = 8.3 Hz), 128.86, 128.79, 128.43, 128.23, 127.32 (d, *J* = 5.8 Hz), 126.77, 124.11, 123.62, 117.28, 52.98.

¹⁵N NMR (51 MHz, CDCl₃) δ 319.48.

HRMS: ASAP (positive) M = C₁₈H₁₅N¹⁵S: calculated (M+H)⁺ m/z 280.1006; found (M+H)⁺ m/z 280.1012.



(4d) *N*-(4-methylbenzyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure D from 1-(4-methylbenzyl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (49.9 mg,

0.10 mmol). Purified via preparatory TLC (5% EtOAc/hexanes) to afford 8.6 mg (30%) of a clear oil.

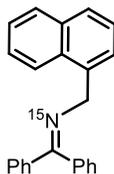
R_f: 0.51 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.73 – 7.67 (m, 2H), 7.53 – 7.45 (m, 3H), 7.43 – 7.38 (m, 1H), 7.37 – 7.33 (m, 2H), 7.25 – 7.22 (m, 4H), 7.15 (d, *J* = 7.7 Hz, 2H), 4.60 (s, 2H), 2.36 (s, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 168.71, 139.98, 137.76, 136.91, 136.16, 132.56, 130.22, 130.13, 129.15, 128.70, 128.61, 128.43, 128.18, 127.97, 127.73, 57.38, 29.85.

¹⁵N NMR (51 MHz, CDCl₃) δ 300.01.

LRMS: (EI) [C₂₁H₁₉N¹⁵]: *m/z* calculated 286.15; found 286.1.



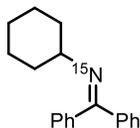
(4e) *N*-(naphthalen-1-ylmethyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure D from 1-(naphthalen-1-ylmethyl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (53.5 mg, 0.1 mmol). Purified via preparatory TLC (3% EtOAc/hexanes) to afford 16.7 mg (52%) of a clear oil.

¹H NMR (500 MHz, CDCl₃) δ 8.03 – 8.00 (m, 1H), 7.88 – 7.85 (m, 1H), 7.76 (d, *J* = 7.9 Hz, 1H), 7.72 – 7.69 (m, 2H), 7.52 – 7.45 (m, 7H), 7.42 – 7.37 (m, 1H), 7.36 – 7.33 (m, 2H), 7.28 – 7.26 (m, 2H), 5.05 (s, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 169.18, 140.00, 136.82, 136.56, 133.87, 131.77, 130.22, 128.78, 128.75, 128.73, 128.42, 128.22, 127.92, 127.49, 127.39, 125.89, 125.75, 125.66, 125.60, 125.36, 125.23, 124.00, 55.40, 29.85.

¹⁵N NMR (51 MHz, CDCl₃) δ 320.53.

LRMS: (EI) [C₂₂H₁₄N¹⁵]: *m/z* calculated 322.15; found 322.1.



(5a) *N*-cyclohexyl-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 1-cyclohexyl-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (119.25 mg, 0.25 mmol). Purified using flash chromatography to yield 52.8 mg of a clear oil (80%).

R_f: 0.8 (5% acetone/pentane, 1 drop Et₃N).

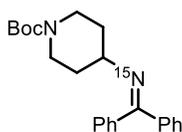
¹H NMR (500 MHz, CDCl₃) δ 7.58 (d, *J* = 6.6 Hz, 2H), 7.44 (d, *J* = 7.4 Hz, 3H), 7.31 (d, *J* = 7.4 Hz, 3H), 7.16 (dd, *J* = 7.8, 1.7 Hz, 2H), 3.22 (m, 1H), 1.74 (m, 2H), 1.61 (s, 6H), 1.14 (m, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 165.69 (d, *J* = 5.9 Hz), 140.52, 137.60 (d, *J* = 2.7 Hz), 129.70, 128.51 (d, *J* = 2.3 Hz), 128.19, 128.11, 127.83, 61.53, 34.10 (d, *J* = 2.7 Hz), 25.83, 24.56.

¹⁵N NMR (51 MHz, CDCl₃) δ 339.22.

IR (CDCl₃): 3058.82, 3023.84, 2925.91, 2852.36, 1594.22, 1572.19, 1445.74, 1285.66, 1177.45, 964.90, 771.17, 695.23 cm⁻¹.

HRMS: ASAP (positive) *M* = C₁₉H₂₁N¹⁵: calculated (*M*+*H*)⁺ *m/z* 266.1755; found (*M*+*H*)⁺ *m/z* 266.1774.



(5b) *tert*-butyl 4-((diphenylmethylene)amino-¹⁵N)piperidine-1-carboxylate: Synthesized according to General Procedure E from 1-(1-(*tert*-butoxycarbonyl)piperidin-4-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (144.5 mg, 0.25 mmol). Purified using flash chromatography to yield 79.1 mg of a yellow oil (87%).

R_f: 0.3 (5% acetone/pentane, 1 drop Et₃N).

¹H NMR (500 MHz, CDCl₃) δ 7.59 (dt, *J* = 7.0, 1.4 Hz, 2H), 7.45 (tdd, *J* = 7.4, 5.7, 2.0 Hz, 3H), 7.39 – 7.34 (m, 1H), 7.31 (dd, *J* = 8.1, 6.4 Hz, 2H), 7.19 – 7.12 (m, 2H), 3.99 (s, 2H), 3.42 (tq, *J*

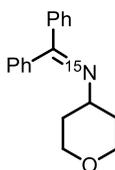
= 7.7, 3.9 Hz, 1H), 2.86 (t, $J = 11.4$ Hz, 2H), 1.73 (dtd, $J = 13.6, 9.6, 3.9$ Hz, 2H), 1.58 (dd, $J = 13.3, 4.3$ Hz, 2H), 1.46 (s, 9H).

^{13}C NMR (126 MHz, CDCl_3) δ 166.74 (d, $J = 5.5$ Hz), 155.08, 140.08 (d, $J = 8.0$ Hz), 137.19 (d, $J = 3.0$ Hz), 130.05, 128.69, 128.54 (d, $J = 3.0$ Hz), 128.46, 128.19, 127.67, 79.44, 58.73, 41.58, 32.98, 28.62.

^{15}N NMR (51 MHz, CDCl_3) δ 334.07.

IR (CDCl_3): 2974.20, 2927.82, 1858.65, 1688.62, 1608.87, 1420.91, 1364.58, 1233.91, 1169.64, 1133.01, 772.06, 696.54 cm^{-1} .

HRMS: ASAP (positive) $\text{M} = \text{C}_{23}\text{H}_{28}\text{O}_2\text{N}(\text{N}^{15})$: calculated (M+H) $^+$ m/z 367.2231; found (M+H) $^+$ m/z 367.2229.



(5c) 1,1-diphenyl- N -(tetrahydro-2H-pyran-4-yl)methanimine- ^{15}N : Synthesized according to General Procedure E from 2,4,6-triphenyl-1-(tetrahydro-2H-pyran-4-yl)pyridin-1-ium tetrafluoroborate (120 mg, 0.25 mmol). Purified via flash chromatography (2% EtOAc/pentane \rightarrow 10% EtOAc/pentane) to afford 51.1 mg (77%) of a white solid.

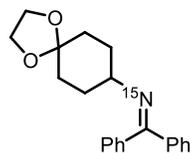
R_f: 0.17 (5% EtOAc/pentane, 1 drop Et_3N)

^1H NMR (400 MHz, CDCl_3) δ 7.66 – 7.60 (m, 2H), 7.50 – 7.43 (m, 3H), 7.34 (ddd, $J = 12.7, 7.9, 6.1$ Hz, 3H), 7.20 – 7.13 (m, 2H), 4.01 (dt, $J = 11.5, 3.9$ Hz, 2H), 3.50 (dtt, $J = 9.6, 4.4, 2.9$ Hz, 1H), 3.37 (td, $J = 11.2, 2.4$ Hz, 2H), 2.00 – 1.86 (m, 2H), 1.58 (dd, $J = 13.2, 3.6$ Hz, 2H).

^{13}C NMR (101 MHz, CDCl_3) δ 166.82, 139.99 (d, $J = 8.1$ Hz), 137.13, 132.51, 130.16, 130.07, 128.65, 128.57 (d, $J = 2.9$ Hz), 128.47, 128.38, 128.17, 127.66, 66.06 (d, $J = 1.8$ Hz), 58.02, 33.81 (d, $J = 2.6$ Hz).

^{15}N NMR (51 MHz, CDCl_3) δ 333.99.

HRMS: ASAP (positive) M = C₁₈H₁₉N¹⁵O: calculated (M+H)⁺ m/z 268.1549; found (M+H)⁺ m/z 268.1551.



(5d) 1,1-diphenyl-N-(1,4-dioxaspiro[4.5]decan-8-yl)methanimine-¹⁵N: Synthesized according to General Procedure E from 2,4,6-triphenyl-1-(1,4-dioxaspiro[4.5]decan-8-yl)pyridin-1-ium tetrafluoroborate (133.75 mg, 0.25 mmol). Purified using flash chromatography to yield a clear oil (57.7 mg, 72%).

R_f: 0.35 (5% EtOAc/pentane, 1 drop Et₃N).

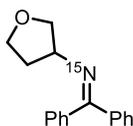
¹H NMR (500 MHz, CDCl₃) δ 7.63 – 7.58 (m, 2H), 7.49 – 7.39 (m, 3H), 7.38 – 7.24 (m, 3H), 7.18 – 7.13 (m, 2H), 4.00 – 3.90 (m, 4H), 3.35 (tq, *J* = 8.5, 3.7 Hz, 1H), 1.97 – 1.81 (m, 4H), 1.70 – 1.59 (m, 2H), 1.49 (m, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 166.10 (d, *J* = 6.0 Hz), 140.25 (d, *J* = 8.0 Hz), 137.32, 129.83, 128.59, 128.54, 128.29, 128.10, 127.72, 108.76, 64.37, 58.96, 32.40, 31.23.

¹⁵N NMR (51 MHz, CDCl₃) δ 336.25.

IR (CDCl₃): 2931.65, 2870.38, 1609.15, 1444.11, 1285.01, 1144.79, 1099.73, 948.62, 696.41 cm⁻¹.

HRMS: ASAP (positive) M = C₂₁H₂₃O₂N¹⁵: calculated (M+H)⁺ m/z 324.1810; found (M+H)⁺ m/z 324.1816.



(5e) 1,1-diphenyl-N-(tetrahydrofuran-3-yl)methanimine-¹⁵N: Synthesized according to General Procedure E from 2,4,6-triphenyl-1-(tetrahydrofuran-3-yl)pyridin-1-ium tetrafluoroborate (116.25 mg, 0.25 mmol). Purified using flash chromatography to yield a clear oil (47.5 mg, 75%).

R_f: 0.28 (5% EtOAc/pentane, 1 drop Et₃N).

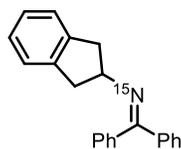
¹H NMR (500 MHz, CDCl₃) δ 7.62 (dt, *J* = 7.1, 1.6 Hz, 2H), 7.52 – 7.41 (m, 3H), 7.40 – 7.35 (m, 1H), 7.32 (dd, *J* = 8.3, 6.4 Hz, 2H), 7.17 – 7.11 (m, 2H), 4.10 (dt, *J* = 8.5, 7.1 Hz, 1H), 4.02 (td, *J* = 6.2, 4.9 Hz, 1H), 3.85 (dt, *J* = 8.4, 6.7 Hz, 2H), 3.81 – 3.70 (m, 1H), 2.08 – 2.00 (m, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 167.75 (d, *J* = 6.0 Hz), 139.93 – 139.66 (m), 137.16, 130.16, 128.68, 128.63, 128.58, 128.19, 127.96, 74.49 (d, *J* = 4.5 Hz), 68.28, 62.11, 35.31.

¹⁵N NMR (51 MHz, CDCl₃) δ 329.33.

IR (CDCl₃): 2970.43, 2940.26, 2860.66, 1610.08, 1571.56, 1444.80, 1285.91, 1074.41, 912.63, 778.80, 696.53 cm⁻¹.

HRMS: ASAP (positive) *M* = C₁₇H₁₇ON¹⁵: calculated (M+H)⁺ *m/z* 254.1391; found (M+H)⁺ *m/z* 254.1398.



(5f) *N*-(2,3-dihydro-1H-inden-2-yl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 1-(2,3-dihydro-1*H*-inden-2-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (51.1 mg, 0.1 mmol). Purified using flash chromatography to yield a clear oil (25.4 mg, 85%).

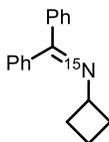
Rf: (5% EtOAc/pentane, 1 drop Et₃N).

¹H NMR (500 MHz, CDCl₃) δ 7.68 – 7.63 (m, 2H), 7.50 – 7.46 (m, 2H), 7.45 – 7.43 (m, 1H), 7.40 – 7.37 (m, 1H), 7.35 (t, *J* = 7.0 Hz, 2H), 7.22 (ddd, *J* = 16.9, 6.6, 2.6 Hz, 4H), 7.15 (dd, *J* = 5.6, 3.2 Hz, 2H), 4.37 – 4.26 (m, 1H), 3.21 (dd, *J* = 15.5, 7.3 Hz, 2H), 3.03 (dd, *J* = 15.5, 7.6 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 167.62, 142.46, 140.04, 137.48, 130.03, 128.63, 128.46, 128.20, 127.88, 126.42, 124.52, 63.48, 41.41.

¹⁵N NMR (51 MHz, CDCl₃) δ 334.29.

HRMS: ASAP (positive) *M* = C₂₂H₁₉N¹⁵: calculated (M+H)⁺ *m/z* 300.1600; found (M+H)⁺ *m/z* 300.1606.



(5g) *N*-cyclobutyl-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-cyclobutyl-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (125 mg, 0.25 mmol). Purified via preparatory TLC (3% EtOAc/pentane) to afford 41.5 mg (70%) of a yellow oil.

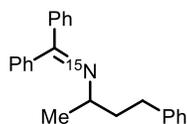
R_f: 0.72 (3% EtOAc/pentane, 1 drop Et₃N).

¹H NMR (400 MHz, CDCl₃) δ 7.66 – 7.58 (m, 2H), 7.44 (qd, *J* = 4.7, 1.6 Hz, 3H), 7.34 (ddd, *J* = 12.7, 7.8, 6.0 Hz, 3H), 7.19 – 7.08 (m, 2H), 4.08 – 3.95 (m, 1H), 2.30 (dddq, *J* = 14.2, 9.8, 4.9, 2.4 Hz, 2H), 2.16 – 2.03 (m, 2H), 1.86 (tdd, *J* = 12.2, 9.4, 2.9 Hz, 1H), 1.67 (qt, *J* = 10.4, 8.3 Hz, 1H).

¹³C NMR (101 MHz, CDCl₃) δ 166.82 (d, *J* = 5.6 Hz), 140.02 (d, *J* = 7.7 Hz), 137.57 (d, *J* = 2.5 Hz), 129.96, 128.60 (d, *J* = 2.9 Hz), 128.48, 128.38, 128.14, 127.99, 57.27 (d, *J* = 2.1 Hz), 31.54 (d, *J* = 2.1 Hz), 16.16 (d, *J* = 2.9 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 320.10.

HRMS: ASAP (positive) *M* = C₁₇H₁₇N¹⁵: calculated (*M*+H)⁺ *m/z* 238.1443; found (*M*+H)⁺ *m/z* 238.1452.



(5h) *N*-(2-methyl-4-phenylbutyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 2,4,6-triphenyl-1-(4-phenylbutan-2-yl)pyridin-1-ium tetrafluoroborate (131.8 mg). Purified via preparatory TLC (3% EtOAc/pentane) to afford 57.8 mg (70%) of a yellow oil.

R_f: 0.72 (3% EtOAc/pentane, 1 drop Et₃N).

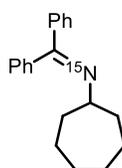
¹H NMR (500 MHz, CDCl₃) δ 7.71 – 7.66 (m, 2H), 7.51 – 7.33 (m, 6H), 7.29 (t, *J* = 7.4 Hz, 2H), 7.22 – 7.15 (m, 5H), 3.53 (dddd, *J* = 8.3, 6.3, 4.4, 1.9 Hz, 1H), 2.66 (ddd, *J* = 13.6, 11.0, 5.4 Hz,

1H), 2.50 (ddd, $J = 13.6, 10.9, 5.6$ Hz, 1H), 2.08 – 1.95 (m, 1H), 1.92 – 1.81 (m, 1H), 1.26 (dd, $J = 6.3, 2.6$ Hz, 3H).

^{13}C NMR (126 MHz, CDCl_3) δ 166.59, 142.68, 140.34 (d, $J = 7.7$ Hz), 137.52 (d, $J = 2.4$ Hz), 129.84, 128.52, 128.50, 128.42, 128.35, 128.21, 128.16, 127.82, 125.68, 57.29, 40.17 (d, $J = 3.5$ Hz), 33.23, 22.34 (d, $J = 2.3$ Hz).

^{15}N NMR (51 MHz, CDCl_3) δ 338.12.

HRMS: ASAP (positive) $M = \text{C}_{23}\text{H}_{23}\text{N}^{15}$: calculated (M+H)⁺ m/z 316.1913; found (M+H)⁺ m/z 316.1851.



(5i) *N*-cycloheptyl-1,1-diphenylmethanimine- ^{15}N : Synthesized according to General Procedure E from 1-cycloheptyl-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (122.7 mg, 0.25 mmol). Purified using flash chromatography to yield a light yellow oil (28.5 mg, 41%).

R_f: 0.76 (5% EtOAc/pentane, 1 drop Et_3N).

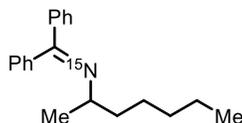
^1H NMR (500 MHz, CDCl_3) δ 7.60 – 7.55 (m, 2H), 7.43 (m, 3H), 7.37 – 7.28 (m, 3H), 7.17 – 7.11 (m, 2H), 3.39 (dtd, $J = 8.7, 4.3, 2.2$ Hz, 1H), 1.73 (m, 4H), 1.68 – 1.45 (m, 6H), 1.41 – 1.29 (m, 2H).

^{13}C NMR (126 MHz, CDCl_3) δ 164.36, 140.57, 137.64, 129.63, 128.51, 128.46, 128.17, 128.11, 127.85, 63.26, 35.98, 28.78, 24.69.

^{15}N NMR (51 MHz, CDCl_3) δ 341.02.

IR (CDCl_3): 2923.06, 2854.68, 1608.46, 1444.32, 1313.79, 1108.21, 776.86, 695.76 cm^{-1} .

HRMS: ASAP (positive) $M = \text{C}_{20}\text{H}_{23}\text{N}^{15}$: calculated (M+H)⁺ m/z 281.1947; found (M+H)⁺ m/z 281.1944.



(5j) *N*-(heptan-2-yl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 1-(heptan-2-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (123.35 mg, 0.25 mmol). Purified using flash chromatography to yield a light yellow oil (44.9 mg, 69%).

R_f: 0.8 (5% EtOAc/pentane, 1 drop Et₃N).

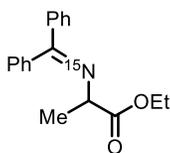
¹H NMR (500 MHz, CDCl₃) δ 7.60 (d, *J* = 6.6 Hz, 2H), 7.51 – 7.38 (m, 3H), 7.37 – 7.28 (m, 3H), 7.18 – 7.12 (m, 2H), 3.38 (m, 1H), 1.62 (m, 1H), 1.46 (m, 1H), 1.38 – 1.06 (m, 9H), 0.85 (t, *J* = 7.3 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 165.97 (d, *J* = 5.5 Hz), 140.54, 140.48, 137.79, 137.77, 129.71, 128.51, 128.48, 128.46, 128.15, 127.90, 57.60, 38.47, 32.02, 26.50, 22.78, 22.46, 14.21.

¹⁵N NMR (51 MHz, CDCl₃) δ 339.65.

IR (CDCl₃): 3059.43, 2959.02, 2925.72, 2856.96, 1608.20, 1573.23, 1284.70, 1137.19, 775.74, 695.04, 640.26 cm⁻¹.

HRMS: ASAP (positive) *M* = C₂₀H₂₅N¹⁵: calculated (*M*+H)⁺ *m/z* 282.2069; found (*M*+H)⁺ *m/z* 282.2075.



(5k) ethyl 2-((diphenylmethylene)amino-¹⁵N)propanoate: Synthesized according to General Procedure E from 1-(1-ethoxy-1-oxopropan-2-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (123.75 mg, 0.25 mmol). Isolated via flash chromatography (2% EtOAc/pentane until all triphenyl pyridine elutes → 5% EtOAc/pentane) to yield a light yellow oil (44.2 mg, 64%).

R_f: 0.34 (5% EtOAc/pentane, 1 drop Et₃N).

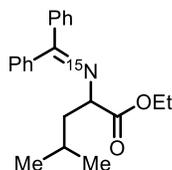
¹H NMR (500 MHz, CDCl₃) δ 7.64 (d, *J* = 7.6 Hz, 2H), 7.50 – 7.41 (m, 3H), 7.39 (t, *J* = 7.6 Hz, 1H), 7.33 (t, *J* = 7.6 Hz, 2H), 7.23 – 7.15 (m, 2H), 4.17 (m, 3H), 1.43 (dd, *J* = 6.8, 2.7 Hz, 3H), 1.26 (t, *J* = 7.1 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 173.04 (d, *J* = 2.3 Hz), 169.76 (d, *J* = 5.4 Hz), 139.70 (d, *J* = 8.2 Hz), 136.47 (d, *J* = 2.3 Hz), 130.40, 128.92, 128.89, 128.75, 128.70, 128.17, 127.84, 60.97, 60.79, 19.30 (d, *J* = 2.7 Hz), 14.33.z

¹⁵N NMR (51 MHz, CDCl₃) δ 321.17.

IR (CDCl₃): 3059.51, 2933.89, 1734.22, 1608.92, 1444.96, 1285.21, 1191.77, 1115.24, 779.93, 695.76 cm⁻¹.

HRMS: ASAP (positive) M = C₁₈H₁₉O₂N¹⁵: calculated (M+H)⁺ m/z 284.1498; found (M+H)⁺ m/z 284.1507.



(5l) ethyl 2-((diphenylmethylene)amino-¹⁵N)-4-methylpentanoate: Synthesized according to General Procedure E from 1-(1-ethoxy-4-methyl-1-oxopentan-2-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (134.2 mg, 0.25 mmol). Isolated via flash chromatography (2% EtOAc/pentane until all triphenyl pyridine elutes → 5% EtOAc/pentane) to yield a light yellow oil (53.2 mg, 68%).

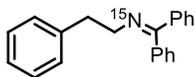
R_f: 0.3 (5% EtOAc/pentane, 1 drop Et₃N)

¹H NMR (500 MHz, CDCl₃) δ 7.65 (d, *J* = 7.2 Hz, 2H), 7.45 (dd, *J* = 5.2, 2.0 Hz, 3H), 7.42 – 7.37 (m, 1H), 7.33 (dd, *J* = 8.2, 6.6 Hz, 2H), 7.23 – 7.16 (m, 2H), 4.25 – 4.13 (m, 2H), 4.10 (dd, *J* = 9.0, 4.9 Hz, 1H), 1.88 (br s, 1H), 1.78 (ddt, *J* = 13.3, 8.7, 4.5 Hz, 1H), 1.63 – 1.51 (sep, 1H), 1.27 (t, *J* = 7.1 Hz, 3H), 0.85 (d, *J* = 6.6 Hz, 3H), 0.67 (d, *J* = 6.6 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 172.88, 157.64, 139.72, 132.55, 130.19, 129.05, 128.84, 128.58, 128.41, 128.18, 128.07, 64.05, 60.98, 42.86, 24.84, 23.32, 21.81, 14.36.

¹⁵N NMR (51 MHz, CDCl₃) δ 320.03.

HRMS: ASAP (positive) $M = C_{21}H_{25}N^{15}O_2$: calculated $(M+H)^+$ m/z 326.1968; found $(M+H)^+$ m/z 326.1972.



(6a) *N*-phenethyl-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-phenethyl-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (137.8 mg, 0.25 mmol). Purified via flash chromatography (1% EtOAc/pentane → 2% EtOAc/pentane, let all triphenylpyridine elute, followed by product) to yield 49.3 mg (69%) of a clear oil.

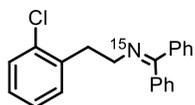
R_f: 0.37 (3% EtOAc/pentane, 1 drop Et₃N).

¹H NMR (400 MHz, CDCl₃) δ 7.62 – 7.55 (m, 2H), 7.41 – 7.29 (m, 6H), 7.24 – 7.21 (m, 2H), 7.19 – 7.09 (m, 3H), 7.00 – 6.91 (m, 2H), 3.64 (t, $J = 7.4$ Hz, 2H), 3.01 (td, $J = 7.4, 2.3$ Hz, 2H).

¹³C NMR (101 MHz, CDCl₃) δ 168.61 (d, $J = 6.1$ Hz), 140.54 (d, $J = 1.9$ Hz), 140.08, 140.00, 136.96 (d, $J = 2.5$ Hz), 130.00, 129.18, 128.50, 128.49, 128.45, 128.36, 128.18, 127.84, 126.06, 55.71, 37.84 (d, $J = 3.2$ Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 323.60.

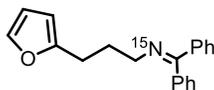
HRMS: ASAP (positive) $M = C_{21}H_{19}N^{15}$: calculated $(M+H)^+$ m/z 288.1600; found $(M+H)^+$ m/z 288.1598.



(6b) *N*-(2-chlorophenethyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-(2-chlorophenethyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (116.8 mg, 0.20 mmol). Purified via preparatory TLC (3% EtOAc/pentane) to afford 45.0mg (70%) of a yellow oil.

¹H NMR (300 MHz, CDCl₃) δ 7.65 (d, $J = 7.5$ Hz, 2H), 7.54 – 7.35 (m, 7H), 7.12 (td, $J = 6.7, 3.7$ Hz, 3H), 6.95 (dd, $J = 6.4, 2.9$ Hz, 2H), 3.75 (t, 2H), 3.19 (t, 2H).

^{13}C NMR (101 MHz, CDCl_3) δ 169.01, 137.90 (d, $J = 23.1$ Hz), 134.43, 132.55, 131.51, 130.21, 130.08, 129.49, 128.54, 128.42, 128.19, 127.83, 127.61, 126.68, 53.47, 35.38 (d, $J = 3.3$ Hz).



(6c) *N*-(3-(furan-3-yl)propyl)-1,1-diphenylmethanimine- ^{15}N : Synthesized according to General Procedure E from 14-(3-(furan-2-yl)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (111 mg, 0.2 mmol). Purified via flash chromatography with a gradient of 9:1 pentane:toluene \rightarrow 9:0.9:0.1 pentane:toluene: Et_2O \rightarrow 9:0.7:0.3 pentane:toluene: Et_2O to afford 47.4 mg (82%).

Rf: 0.4 (90:0.5:0.5 pentane:toluene: Et_2O , 1 drop Et_3N).

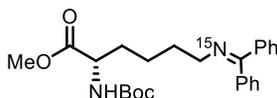
^1H NMR (500 MHz, CDCl_3) δ 7.61 (d, $J = 7.6$ Hz, 2H), 7.43 (m, 7H), 7.15 (d, $J = 7.1$ Hz, 2H), 6.24 (t, $J = 2.4$ Hz, 1H), 5.94 (d, $J = 2.4$ Hz, 1H), 3.42 (t, $J = 6.8$ Hz, 2H), 2.71 (t, $J = 7.0$ Hz, 2H), 2.03 (pd, $J = 7.2, 2.5$ Hz, 2H).

^{13}C NMR (126 MHz, CDCl_3) δ 168.39 (d, $J = 6.0$ Hz), 156.24, 140.82, 140.07 (d, $J = 8.1$ Hz), 137.10, 129.99, 128.59, 128.46 (d, $J = 3.1$ Hz), 128.42, 128.18, 127.94, 110.17, 104.85, 53.13, 29.63 (d, $J = 3.4$ Hz), 26.00.

^{15}N NMR (51 MHz, CDCl_3) δ 324.66.

IR (CDCl_3): 3058.81, 2926.14, 1594.54, 1444.80, 1288.34, 1006.02, 779.50, 696.69 cm^{-1} .

HRMS: ASAP (positive) $\text{M} = \text{C}_{20}\text{H}_{18}\text{ON}^{15}$: calculated $(\text{M}+\text{H})^+ m/z$ 292.1549; found $(\text{M}+\text{H})^+ m/z$ 292.1544.



(6d) Boc- ^{15}N -Lys-OMe: Synthesized according to General Procedure E from (*S*)-14-(5-((tert-butoxycarbonyl)amino)-6-methoxy-6-oxohexyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (69.0 mg, 0.1 mmol). Purified by flash chromatography (5 \rightarrow 10 \rightarrow 15 \rightarrow 20 \rightarrow 100 EtOAc /pentane, comes out right after leftover benzophenone imine) to yield 22.4 mg (53%) of a light yellow oil.

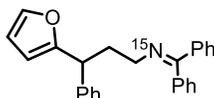
Rf: 0.2 (20% EtOAc/pentane, 1 drop Et₃N).

¹H NMR (500 MHz, CDCl₃) δ 7.61 – 7.55 (m, 2H), 7.44 (ddd, *J* = 12.1, 7.8, 6.1 Hz, 3H), 7.38 – 7.29 (m, 3H), 7.17 – 7.12 (m, 2H), 4.98 (d, *J* = 8.7 Hz, 1H), 4.27 (d, *J* = 6.7 Hz, 1H), 3.70 (s, 3H), 3.38 – 3.32 (m, 2H), 1.77 (d, *J* = 9.8 Hz, 1H), 1.73 – 1.57 (m, 4H), 1.42 (s, 9H), 1.39 (m, 1H).

¹³C NMR (101 MHz, CDCl₃) δ 173.59, 168.18 (d, *J* = 6.1 Hz), 155.50, 140.07 (d, *J* = 8.1 Hz), 137.13 (d, *J* = 2.5 Hz), 129.96, 128.62, 128.45, 128.44, 128.18, 127.93, 79.94, 53.57 (d, *J* = 6.5 Hz), 52.30, 32.76, 30.90 (d, *J* = 3.2 Hz), 28.46, 23.45 (d, *J* = 1.9 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 325.12.

HRMS: ASAP (positive) *M* = C₂₅H₃₃N¹⁵NO₄: calculated (*M*+*H*)⁺ *m/z* 427.2444; found (*M*+*H*)⁺ *m/z* 427.2449.



(6e) *N*-(3-(furan-2-yl)-3-phenylpropyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-(3-(furan-2-yl)-3-phenylpropyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (157.8 mg, 0.25 mmol). Purified by flash chromatography to yield 65.3 mg (65%) of a yellow oil.

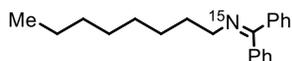
Rf: 0.36 (85% hexanes, 12% toluene, 3% EtOAc)

¹H NMR (400 MHz, CDCl₃) δ 7.68 – 7.61 (m, 2H), 7.45 – 7.31 (m, 6H), 7.30 – 7.25 (m, 3H), 7.24 – 7.18 (m, 3H), 7.09 – 7.04 (m, 2H), 6.27 (dd, *J* = 3.2, 1.8 Hz, 1H), 6.04 (d, *J* = 3.2 Hz, 1H), 4.22 (t, *J* = 7.7 Hz, 1H), 3.42 – 3.26 (m, 2H), 2.49 (dq, *J* = 14.3, 7.2, 2.3 Hz, 1H), 2.28 (dddd, *J* = 15.7, 13.4, 6.7, 2.7 Hz, 1H).

¹³C NMR (101 MHz, CDCl₃) δ 168.42 (d, *J* = 6.0 Hz), 157.96, 142.63, 141.38, 140.08, 140.00, 136.97, 132.54, 130.20, 129.99, 128.54, 128.48, 128.47, 128.44, 128.32, 128.17, 128.07, 127.80, 126.59, 110.05, 105.46, 51.48, 42.89 (d, *J* = 1.8 Hz), 36.14 (d, *J* = 3.5 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 324.13.

HRMS: ASAP (positive) *M* = C₂₆H₂₃N¹⁵O: calculated (*M*+*H*)⁺ *m/z* 368.1862; found (*M*+*H*)⁺ *m/z* 368.1858.



(6f) *N*-octyl-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-octyl-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (139.5 mg, 0.25 mmol). Purified via preparatory TLC (5% EtOAc/pentane) to afford 57.4 mg (78%) of a yellow oil.

R_f: 0.8 (5% EtOAc/pentane, 1 drop Et₃N).

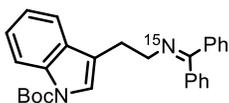
¹H NMR (500 MHz, CDCl₃) δ 7.62 – 7.59 (m, 2H), 7.48 – 7.41 (m, 3H), 7.34 (ddd, *J* = 14.5, 7.9, 6.1 Hz, 3H), 7.18 – 7.15 (m, 2H), 3.38 (t, *J* = 7.1 Hz, 2H), 1.69 (pd, *J* = 7.1, 2.3 Hz, 2H), 1.36 – 1.24 (m, 10H), 0.88 (t, *J* = 6.9 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 167.77 (d, *J* = 6.1 Hz), 140.23 (d, *J* = 7.9 Hz), 137.25 (d, *J* = 2.6 Hz), 129.83, 128.52, 128.42 (d, *J* = 3.0 Hz), 128.32, 128.14, 127.99, 54.07, 31.99, 31.38 (d, *J* = 3.1 Hz), 29.56, 29.39, 27.64, 22.79, 14.24.

¹⁵N NMR (51 MHz, CDCl₃) δ 326.81.

IR (CDCl₃): 2924.03, 2853.19, 1608.52, 1444.63, 1286.78, 1249.55, 1155.22, 694.81 cm⁻¹.

HRMS: ASAP (positive) *M* = C₂₁H₂₇N¹⁵: calculated (*M*+*H*)⁺ *m/z* 296.2226; found (*M*+*H*)⁺ *m/z* 296.2209.



(6g) *tert*-butyl 3-(2-((diphenylmethylene)amino-¹⁵N)ethyl)-1*H*-indole-1-carboxylate: Synthesized according to General Procedure E from 14-(2-(1-(*tert*-butoxycarbonyl)-1*H*-indol-3-yl)ethyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (172.2 mg, 0.25 mmol). Purified by flash chromatography (2% EtOAc/hexanes until triphenylpyridine elutes, then ramp to 5→10% EtOAc/hexanes) to yield 55.4 mg (52%) of a yellow oil.

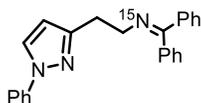
R_f: 0.47 (10% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 8.11, (br s, 1H), 7.63 (d, *J* = 7.0 Hz, 2H), 7.42 – 7.37 (m, 6H), 7.34 (dd, *J* = 8.2, 6.5 Hz, 2H), 7.29 (ddd, *J* = 8.4, 7.1, 1.2 Hz, 1H), 7.19 – 7.15 (m, 1H), 7.06 (dd, *J* = 6.5, 2.9 Hz, 2H), 3.77 – 3.70 (m, 2H), 3.09 (tdd, *J* = 7.1, 2.3, 1.1 Hz, 2H), 1.65 (s, 9H).

¹³C NMR (126 MHz, CDCl₃) δ 168.77 (d, *J* = 6.0 Hz), 149.93, 139.97 (d, *J* = 8.1 Hz), 136.85 (d, *J* = 2.6 Hz), 135.52, 132.53, 130.94, 130.19, 130.04, 128.57, 128.50, 128.48, 128.42, 128.17, 127.81, 124.25, 123.16, 122.34, 119.32 – 119.11 (m), 115.25, 83.36, 53.76, 28.34, 26.90 (d, *J* = 3.6 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 323.86.

HRMS: ASAP (positive) M = C₂₈H₂₈N¹⁵NO₂: calculated (M+H)⁺ m/z 427.2232; found (M+H)⁺ m/z 427.2233.



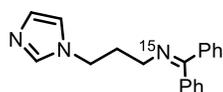
(6h) 1,1-diphenyl-N-(2-(1-phenyl-1H-pyrazol-3-yl)ethyl)methanimine-¹⁵N: Synthesized according to General Procedure E from 7-phenyl-14-(2-(1-phenyl-1H-pyrazol-4-yl)ethyl)-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (154.3 mg, 0.25 mmol). Purified by flash chromatography to yield 70.3 mg (80%) of a clear oil.

¹H NMR (500 MHz, CDCl₃) δ 7.70 (s, 1H), 7.62 (ddd, *J* = 6.7, 5.0, 2.4 Hz, 4H), 7.54 (s, 1H), 7.43 – 7.37 (m, 6H), 7.36 – 7.31 (m, 2H), 7.26 – 7.21 (m, 1H), 7.09 – 7.00 (m, 2H), 3.63 (td, *J* = 6.9, 1.1 Hz, 2H), 2.94 (td, *J* = 6.9, 2.7 Hz, 2H).

¹³C NMR (101 MHz, CDCl₃) δ 168.74 (d, *J* = 6.1 Hz), 141.56, 140.40, 139.94, 139.86, 136.93 (d, *J* = 2.5 Hz), 132.51, 130.17, 130.08, 129.45, 128.56, 128.46, 128.44, 128.39, 128.21, 127.82, 126.11, 125.48, 121.87 (d, *J* = 1.7 Hz), 118.88, 54.82 (d, *J* = 1.4 Hz), 26.25 (d, *J* = 3.5 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 323.54.

LRMS: (EI) [C₂₄H₂₁N₂N¹⁵]: m/z calculated 352.17; found 352.1.



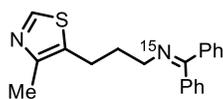
(6i) *N*-(3-(1H-imidazol-1-yl)propyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-(3-(1*H*-imidazol-1-yl)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (139 mg, 0.25 mmol). Purified by preparatory TLC (50% EtOAc/hexanes) to yield 59.4 mg (82%) of a clear oil.

R_f: 0.09 (50% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.65 – 7.59 (m, 2H), 7.52 – 7.39 (m, 5H), 7.34 (dd, *J* = 8.2, 6.5 Hz, 2H), 7.14 – 7.08 (m, 2H), 7.03 (s, 1H), 6.90 (s, 1H), 4.17 – 4.08 (m, 2H), 3.32 (td, *J* = 6.4, 1.4 Hz, 2H), 2.12 (pd, *J* = 6.8, 2.9 Hz, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 169.19 (d, *J* = 6.0 Hz), 139.69, 139.62, 137.38, 136.74, 130.29, 129.54, 128.77, 128.68, 128.44, 128.41, 128.27, 127.66, 119.01, 49.98, 44.93, 32.54 (d, *J* = 3.9 Hz).

¹⁵N NMR (51 MHz, CDCl₃) δ 320.60.



(6j) *N*-(3-(4-methylthiazol-5-yl)propyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-(3-(4-methylthiazol-5-yl)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (146.25 mg, 0.25 mmol). Purified by flash chromatography (2% EtOAc/hexanes until triphenylpyridine elutes, then ramp to 5% EtOAc/hexanes) to yield 54.5 mg (68%) of a yellow oil.

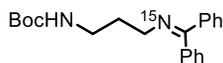
R_f: 0.44 (5% EtOAc/hexanes, 1 drop Et₃N).

¹H NMR (500 MHz, CDCl₃) δ 7.61 (d, *J* = 7.6 Hz, 2H), 7.42 (dd, *J* = 9.2, 6.9 Hz, 3H), 7.38 (t, *J* = 7.3 Hz, 1H), 7.32 (t, *J* = 7.4 Hz, 2H), 7.17 – 7.12 (m, 2H), 6.69 (d, *J* = 1.4 Hz, 1H), 3.47 (t, *J* = 6.8 Hz, 2H), 3.07 (t, *J* = 7.8 Hz, 2H), 2.39 (s, 3H), 2.16 (t, *J* = 7.2 Hz, 2H).

¹³C NMR (101 MHz, CDCl₃) δ 170.71, 152.24, 136.99, 132.55, 130.21, 130.10, 128.63, 128.54, 128.51, 128.42, 128.19, 127.90, 112.44, 52.91, 31.62 (d, *J* = 3.5 Hz), 31.56 (d, *J* = 1.9 Hz), 17.18.

¹⁵N NMR (51 MHz, CDCl₃) δ 323.81.

HRMS: ASAP (positive) M = C₂₀H₂₀N¹⁵NS: calculated (M+H)⁺ m/z 323.1427; found (M+H)⁺ m/z 323.1428.



(6k) tert-butyl (3-((diphenylmethylene)amino-¹⁵N)propyl)carbamate: Synthesized according to General Procedure E from 14-(3-((*tert*-butoxycarbonyl)amino)propyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (151 mg, 0.25 mmol). Purified by flash chromatography to yield 56.7 mg (67%) of a white solid.

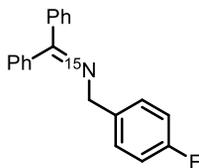
R_f: 0.54 (30% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.62 – 7.57 (m, 2H), 7.45 (dt, *J* = 12.2, 6.7 Hz, 3H), 7.38 (t, *J* = 7.2 Hz, 1H), 7.32 (t, *J* = 7.5 Hz, 2H), 7.18 – 7.12 (m, 2H), 5.41 (s, 1H), 3.43 (t, *J* = 6.4 Hz, 2H), 3.28 (d, *J* = 6.5 Hz, 2H), 1.88 – 1.78 (m, 2H), 1.44 (s, 9H).

¹³C NMR (126 MHz, CDCl₃) δ 168.59, 156.16, 139.75 (d, *J* = 8.0 Hz), 136.80, 130.15, 128.72, 128.58, 128.46, 128.44, 128.23, 127.81, 78.86, 52.50, 40.08, 30.93, 28.60.

¹⁵N NMR (51 MHz, CDCl₃) δ 321.87.

HRMS: ASAP (positive) M = C₂₁H₂₆N¹⁵NO₂: calculated (M+H)⁺ m/z 341.2076; found (M+H)⁺ m/z 341.2081.



(6l) N-(4-fluorobenzyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-(4-fluorobenzyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (111 mg, 0.2 mmol). Isolated via preparatory TLC (90% pentane, 9% toluene, 1% EtOAc) to yield 37.1 mg (64%) of a white solid.

R_f: 0.21 (90% pentane, 9% toluene, 1% EtOAc)

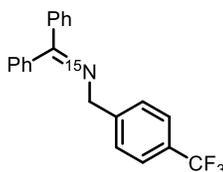
¹H NMR (500 MHz, CDCl₃) δ 7.70 – 7.65 (m, 2H), 7.52 – 7.45 (m, 3H), 7.42 – 7.38 (m, 1H), 7.34 (dd, *J* = 8.2, 6.6 Hz, 2H), 7.29 (dd, *J* = 8.4, 5.6 Hz, 2H), 7.22 – 7.16 (m, 2H), 7.05 – 6.97 (m, 2H), 4.56 (s, 2H).

¹³C NMR (126 MHz, CDCl₃) δ 169.11 (d, *J* = 5.6 Hz), 162.83, 160.89, 139.79 (d, *J* = 8.4 Hz), 136.79 (d, *J* = 2.6 Hz), 136.50, 130.30, 129.26, 129.20, 128.77, 128.74, 128.69, 128.67, 128.24, 127.87, 115.22 (d, *J* = 21.3 Hz), 56.78.

¹⁵N NMR (51 MHz, CDCl₃) δ 320.41.

¹⁹F NMR (471 MHz, CDCl₃) δ -115.91.

HRMS: ASAP (positive) M = C₂₀H₁₆N¹⁵F: calculated (M+H)⁺ m/z 292.1349; found (M+H)⁺ m/z 292.1342.



(6m) *N*-(4-trifluoromethylbenzyl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure E from 14-(4-trifluoromethylbenzyl)-7-phenyl-5,6,8,9-tetrahydridibenzo[*c,h*]acridin-14-ium tetrafluoroborate (121 mg, 0.2 mmol). Isolated via preparatory TLC (90% pentane, 9% toluene, 1% EtOAc) to yield 23.0 mg (34%) of a clear oil.

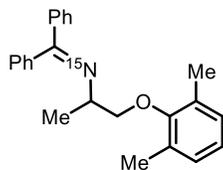
R_f: 0.20 (90% pentane, 9% toluene, 1% EtOAc)

¹H NMR (500 MHz, CDCl₃) δ 7.67 – 7.61 (m, 2H), 7.46 – 7.40 (m, 3H), 7.33 (dd, *J* = 7.8, 1.6 Hz, 5H), 7.07 (d, *J* = 8.0 Hz, 2H), 7.01 – 6.95 (m, 2H), 4.92 (s, 2H).

¹³C NMR (101 MHz, CDCl₃) δ 145.64, 136.83, 130.99, 130.28, 128.88, 128.63, 128.40, 128.20 (d, *J* = 7.6 Hz), 124.86, 73.55.

¹⁵N NMR (51 MHz, CDCl₃) δ 326.96.

LRMS: (EI) [C₂₁H₁₆F₃N¹⁵]: m/z calculated 340.12; found 340.2.

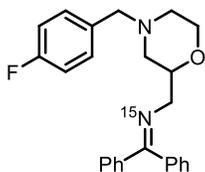


(7a) ¹⁵N-Mexilitene: Synthesized according to General Procedure E from 1-(1-(2,6-dimethylphenoxy)propan-2-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (55.7 mg, 0.1 mmol). Isolated via preparatory TLC (5% EtOAc/hexanes) to yield a light yellow oil (26.6 mg, 77%).

R_f: 0.45 (5% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.74 (ddt, *J* = 8.4, 6.9, 1.2 Hz, 3H), 7.45 – 7.37 (m, 3H), 7.33 – 7.27 (m, 1H), 7.22 – 7.14 (m, 3H), 6.94 (d, *J* = 7.4 Hz, 2H), 6.87 (dd, *J* = 8.2, 6.6 Hz, 1H), 4.17 – 4.06 (m, 1H), 3.87 – 3.72 (m, 2H), 2.17 (s, 6H), 1.29 (d, *J* = 6.4 Hz, 3H).

¹³C NMR (126 MHz, CDCl₃) δ 159.43, 155.98 (d, *J* = 11.0 Hz), 154.42, 142.70, 138.43, 132.02, 131.10, 130.44, 129.42, 128.96, 128.78, 128.68, 128.41, 128.15, 127.54 (d, *J* = 6.0 Hz), 127.29, 124.20, 123.67, 105.77, 76.67, 59.77, 18.54, 16.38.



(7b) ¹⁵N-Mosapride interemediate: Synthesized according to General Procedure E from 14-((4-(4-fluorobenzyl)morpholin-2-yl)methyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (130.8 mg, 0.2 mmol). Isolated via preparatory TLC (25% EtOAc/hexanes) to yield 38.2 mg (50%) of a clear oil.

R_f: 0.18 (25% EtOAc/hexanes)

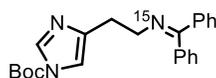
¹H NMR (500 MHz, CDCl₃) δ 7.61 – 7.54 (m, 2H), 7.47 – 7.40 (m, 3H), 7.39 – 7.35 (m, 1H), 7.34 – 7.26 (m, 4H), 7.17 – 7.11 (m, 2H), 7.06 – 6.96 (m, 2H), 3.93 – 3.86 (m, 1H), 3.84 (ddd, *J* = 11.3, 3.3, 1.7 Hz, 1H), 3.69 (td, *J* = 11.3, 2.4 Hz, 1H), 3.56 – 3.47 (m, 1H), 3.44 (d, *J* = 13.0 Hz, 1H), 3.33 (ddd, *J* = 14.2, 6.9, 1.5 Hz, 1H), 2.95 (dt, *J* = 11.2, 2.1 Hz, 1H), 2.62 (dq, *J* = 11.4, 2.0 Hz, 1H), 2.14 (td, *J* = 11.3, 3.3 Hz, 1H), 1.95 (dd, *J* = 11.3, 9.9 Hz, 1H).

¹³C NMR (126 MHz, CDCl₃) δ 169.50 (d, *J* = 6.3 Hz), 163.15, 161.21, 139.86 (d, *J* = 8.3 Hz), 136.82 (d, *J* = 2.6 Hz), 133.74 (d, *J* = 3.3 Hz), 130.76 (d, *J* = 7.9 Hz), 130.13, 128.67, 128.57, 128.13, 127.93, 115.24, 115.07, 76.32 (d, *J* = 4.8 Hz), 66.98, 62.63, 57.34, 56.89, 53.05.

¹⁵N NMR (51 MHz, CDCl₃) δ 316.14.

¹⁹F NMR (470 MHz, CDCl₃) δ -115.88 (m).

HRMS: ASAP (positive) M = C₂₅H₂₅N¹⁵NOF: calculated (M+H)⁺ m/z 390.2000; found (M+H)⁺ m/z 390.1993.



(7c) ¹⁵N-Boc-Histamine: Synthesized according to General Procedure E from 14-(2-(1-(tert-butoxycarbonyl)-1*H*-imidazol-4-yl)ethyl)-7-phenyl-5,6,8,9-tetrahydrodibenzo[*c,h*]acridin-14-ium tetrafluoroborate (64.1 mg, 0.1 mmol). Isolated via preparatory TLC (20% EtOAc/hexanes) to yield 30.1 mg (80%) of a clear oil.

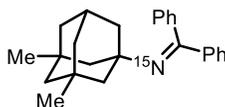
R_f: 0.2 (20% EtOAc/hexanes).

¹H NMR (500 MHz, CDCl₃) δ 7.95 (d, *J* = 1.3 Hz, 1H), 7.61 – 7.57 (m, 2H), 7.46 – 7.35 (m, 4H), 7.33 – 7.30 (m, 2H), 7.14 – 7.09 (m, 3H), 3.66 (td, *J* = 7.2, 1.2 Hz, 2H), 2.96 (tdd, *J* = 7.2, 2.5, 1.0 Hz, 2H), 1.59 (s, 9H).

¹³C NMR (126 MHz, CDCl₃) δ 168.63 (d, *J* = 6.0 Hz), 147.32, 142.42, 140.09 (d, *J* = 8.2 Hz), 136.95, 136.52, 130.01, 128.63, 128.51, 128.17, 127.90, 113.59, 85.24, 53.14, 30.39 (d, *J* = 4.0 Hz), 27.57.

¹⁵N NMR (51 MHz, CDCl₃) δ 323.97.

HRMS: ASAP (positive) M = C₁₇H₁₇N¹⁵: calculated (M+H)⁺ m/z 238.1443; found (M+H)⁺ m/z 238.1452.



(7d) *N*-(3,5-dimethyladamantan-1-yl)-1,1-diphenylmethanimine-¹⁵N: Synthesized according to General Procedure C from *N*-(3,5-dimethyladamantan-1-yl)-1-(2,4,6-trimethoxyphenyl)methanimine (71.4 mg, 0.2 mmol). Isolated via preparatory TLC (3% EtOAc/hexanes) to yield a clear oil (38.5 mg, 56%).

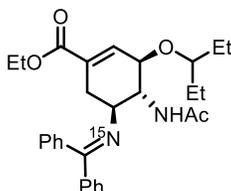
R_f: 0.60 (5% EtOAc/hexanes)

¹H NMR (500 MHz, CDCl₃) δ 7.54 (dt, *J* = 6.9, 1.6 Hz, 2H), 7.41 – 7.32 (m, 3H), 7.31 – 7.26 (m, 3H), 7.21 – 7.15 (m, 2H), 2.01 (p, *J* = 3.2 Hz, 1H), 1.51 (d, *J* = 3.2 Hz, 2H), 1.38 (s, 4H), 1.19 (d, *J* = 3.3 Hz, 4H), 1.10 – 0.99 (m, 2H), 0.75 (s, 6H).

¹³C NMR (126 MHz, CDCl₃) δ 163.13 (d, *J* = 6.7 Hz), 142.31 (d, *J* = 8.9 Hz), 140.47, 130.22, 129.39, 128.90, 128.47, 128.38, 128.18 (d, *J* = 3.2 Hz), 127.98, 127.91, 127.77, 127.64, 127.40, 127.32, 126.55, 60.01, 50.74, 50.68, 42.86, 42.48, 32.50, 30.68, 30.57, 29.85.

¹⁵N NMR (51 MHz, CDCl₃) δ 354.16.

HRMS: ASAP (positive) *M* = C₂₅H₂₉N¹⁵: calculated (*M*+*H*)⁺ *m/z* 346.2383; found (*M*+*H*)⁺ *m/z* 346.2389.



(7e) ¹⁵N-Tamiflu: Synthesized according to General Procedure E from 1-((1*S*,5*R*,6*R*)-6-acetamido-3-(ethoxycarbonyl)-5-(pentan-3-yloxy)cyclohex-3-en-1-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (69 mg, 0.1 mmol). Isolated via preparatory TLC (60% hexanes, 30% EtOAc, 10% toluene) to yield a clear oil (33.9 mg, 71%). NMR showed rotameric protons that could be resolved through variable temperature NMR (see spectra below). Reported shifts are for one rotamer.

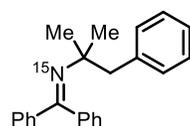
¹H NMR (500 MHz, CDCl₃) δ 7.62 – 7.58 (m, 2H), 7.49 – 7.43 (m, 3H), 7.34 (dd, *J* = 8.1, 6.5 Hz, 2H), 7.14 (ddd, *J* = 6.9, 3.7, 1.6 Hz, 3H), 6.76 (t, *J* = 2.2 Hz, 1H), 5.46 (d, *J* = 7.9 Hz, 1H), 4.45 (ddt, *J* = 9.0, 3.6, 1.9 Hz, 1H), 4.26 – 4.16 (m, 3H), 4.01 (ddd, *J* = 10.7, 9.8, 6.0 Hz, 1H), 3.88 – 3.77 (m, 1H), 3.32 (dp, *J* = 17.2, 5.7 Hz, 1H), 2.62 – 2.52 (ddt, *J* = 25, 10, 5 Hz 1H), 2.51 – 2.40

(m, 2H), 1.93 (s, 3H), 1.59 – 1.42 (m 4H), 1.29 (td, $J = 7.1, 5.6$ Hz, 3H), 0.90 (dt, $J = 11.3, 7.4$ Hz, 6H).

^{13}C NMR (126 MHz, CDCl_3) δ 174.00, 169.79, 166.54, 140.08, 138.59, 137.33, 136.51, 130.21, 129.32, 129.03, 128.68, 128.57, 128.23, 127.96, 127.52, 81.94, 76.53, 73.41, 60.84, 58.93, 31.80, 26.49, 25.84, 23.94, 14.32, 9.72, 9.51.

^{15}N NMR (51 MHz, CDCl_3) δ 322.92.

HRMS: ASAP (positive) $\text{M} = \text{C}_{29}\text{H}_{36}\text{N}^{15}\text{NO}_4$: calculated $(\text{M}+\text{H})^+$ m/z 478.2724; found $(\text{M}+\text{H})^+$ m/z 478.2725.

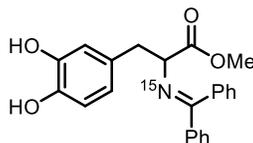


(7f) N -(2-methyl-1-phenylpropan-2-yl)-1,1-diphenylmethanimine- ^{15}N : Synthesized according to General Procedure C from N -(2-methyl-1-phenylpropan-2-yl)-1-(2,4,6-trimethoxyphenyl)methanimine (65.4 mg, 0.2 mmol). Isolated via preparatory TLC (3% EtOAc/hexanes). Clear oil (31.2 mg, 50%).

^1H NMR (500 MHz, CDCl_3) δ 7.58 – 7.55 (m, 2H), 7.31 (dddd, $J = 8.9, 8.2, 5.9, 3.9$ Hz, 6H), 7.25 – 7.20 (m, 5H), 6.96 – 6.89 (m, 2H), 2.95 (d, $J = 2.9$ Hz, 2H), 1.04 (d, $J = 1.8$ Hz, 6H).

^{13}C NMR (101 MHz, CDCl_3) δ 163.22 (d, $J = 6.8$ Hz), 142.07 (d, $J = 9.0$ Hz), 140.01 (d, $J = 2.6$ Hz), 139.43, 132.55, 131.06, 130.21, 129.43, 128.48, 128.42, 128.40, 128.25, 128.22, 127.99, 127.79, 127.75, 127.40, 126.64, 126.06, 60.73 (d, $J = 1.9$ Hz), 52.04 (d, $J = 4.0$ Hz), 28.97.

^{15}N NMR (51 MHz, CDCl_3) δ 351.15.



(7g) ^{15}N -DOPA: Synthesized according to General Procedure E from 1-(3-(3,4-dihydroxyphenyl)-1-methoxy-1-oxopropan-2-yl)-2,4,6-triphenylpyridin-1-ium tetrafluoroborate (50.0 mg, 0.08 mmol). Isolated via preparatory TLC (10% toluene, 90% EtOAc) to yield a light yellow oil (17.2 mg, 53%).

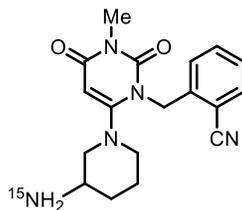
Rf: 0.1 (10% toluene, 90% EtOAc).

¹H NMR (500 MHz, CDCl₃) δ 7.62 – 7.56 (m, 2H), 7.41 – 7.29 (m, 6H), 6.74 (d, *J* = 7.1 Hz, 2H), 6.70 (d, *J* = 8.0 Hz, 1H), 6.53 (d, *J* = 2.0 Hz, 1H), 6.50 – 6.47 (m, 1H), 4.27 (dd, *J* = 9.2, 4.4 Hz, 1H), 3.73 (d, *J* = 2.2 Hz, 3H), 3.17 (dd, *J* = 13.4, 4.5 Hz, 1H), 3.10 – 3.01 (m, 1H).

¹³C NMR (126 MHz, CDCl₃) δ 172.63, 171.25, 143.39, 142.48, 139.51, 136.19, 130.68, 130.49, 128.96, 128.62, 128.35, 128.18, 127.85, 122.41, 116.89, 115.21, 67.46, 52.38, 39.30.

¹⁵N NMR (51 MHz, CDCl₃) δ 314.44.

HRMS: ASAP (positive) M = C₂₃H₂₁N¹⁵O₄: calculated (M+H)⁺ m/z 377.1519; found (M+H)⁺ m/z 377.1510.



(7h) ¹⁵N-Agoliptin: ¹⁵N-labeled benzophenone imine protected Agoliptin was synthesized according to General Procedure E from the corresponding Katritzky pyridinium salt (143.4 mg, 0.2 mmol). The imine was isolated via preparatory TLC (30% EtOAc/hexanes) to yield a clear oil. This imine was dissolved in 1:1 THF: HCl (1M), total concentration 0.05 M and stirred for four hours, checking that starting material was consumed via TLC. The aqueous solution was extracted with EtOAc three times, then basified with NaOH to pH ~10. This aqueous solution was again extracted with EtOAc (3x), organics were collected and dried with Na₂SO₄, then concentrated *in vacuo* to yield a clear oil (35.3 mg, 52%).

¹H NMR (500 MHz, CDCl₃) δ 7.67 (dt, *J* = 7.8, 0.9 Hz, 1H), 7.56 (td, *J* = 7.7, 1.4 Hz, 1H), 7.38 (td, *J* = 7.7, 1.2 Hz, 1H), 7.15 – 7.11 (m, 1H), 5.37 (s, 1H), 5.34 – 5.24 (m, 2H), 3.31 (s, 3H), 3.02 (dd, *J* = 11.6, 3.8 Hz, 1H), 2.97 – 2.86 (m, 2H), 2.60 (t, *J* = 11.0 Hz, 1H), 2.39 (d, *J* = 8.4 Hz, 1H), 1.98 – 1.89 (m, 1H), 1.76 (tdd, *J* = 8.0, 6.0, 3.9 Hz, 1H), 1.66 – 1.54 (m, 1H), 1.21 (d, *J* = 10.9 Hz, 1H).

¹³C NMR (126 MHz, CDCl₃) δ 163.23, 159.88, 152.87, 140.97, 133.39, 133.25, 128.05, 126.76, 117.21, 110.92, 90.68, 59.70, 52.02, 47.47, 46.44, 33.53, 29.83, 28.12, 23.33.

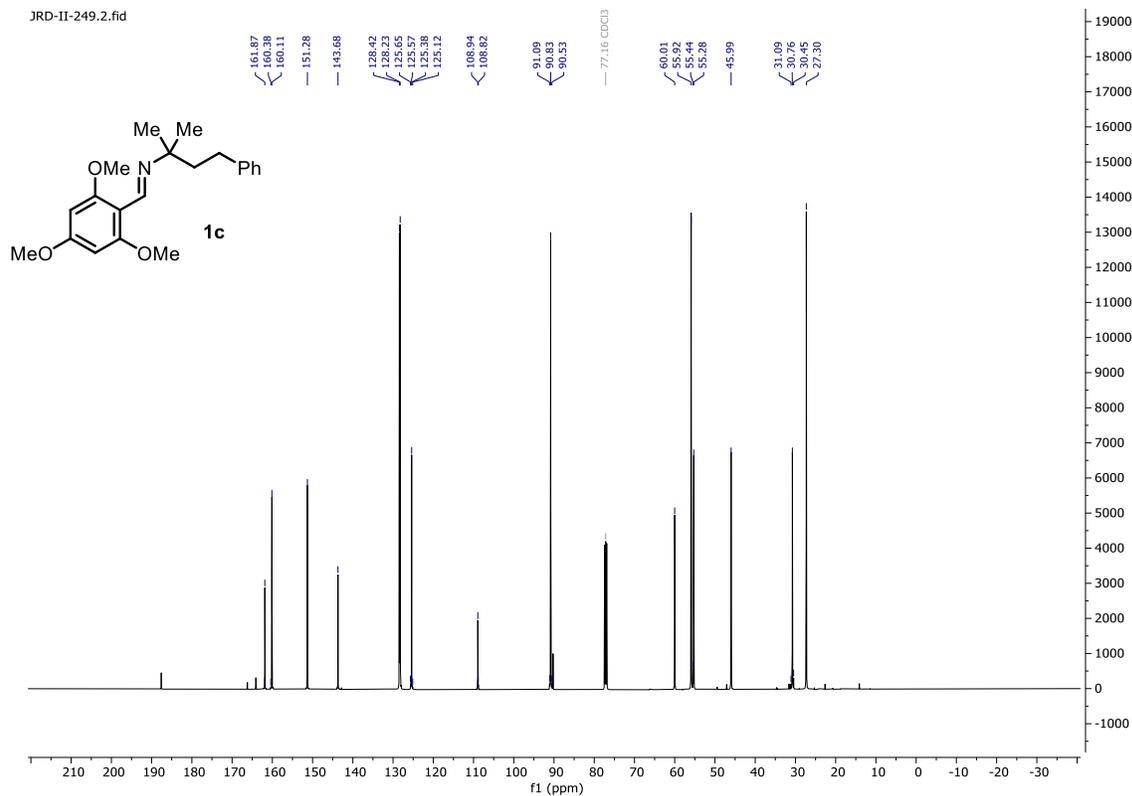
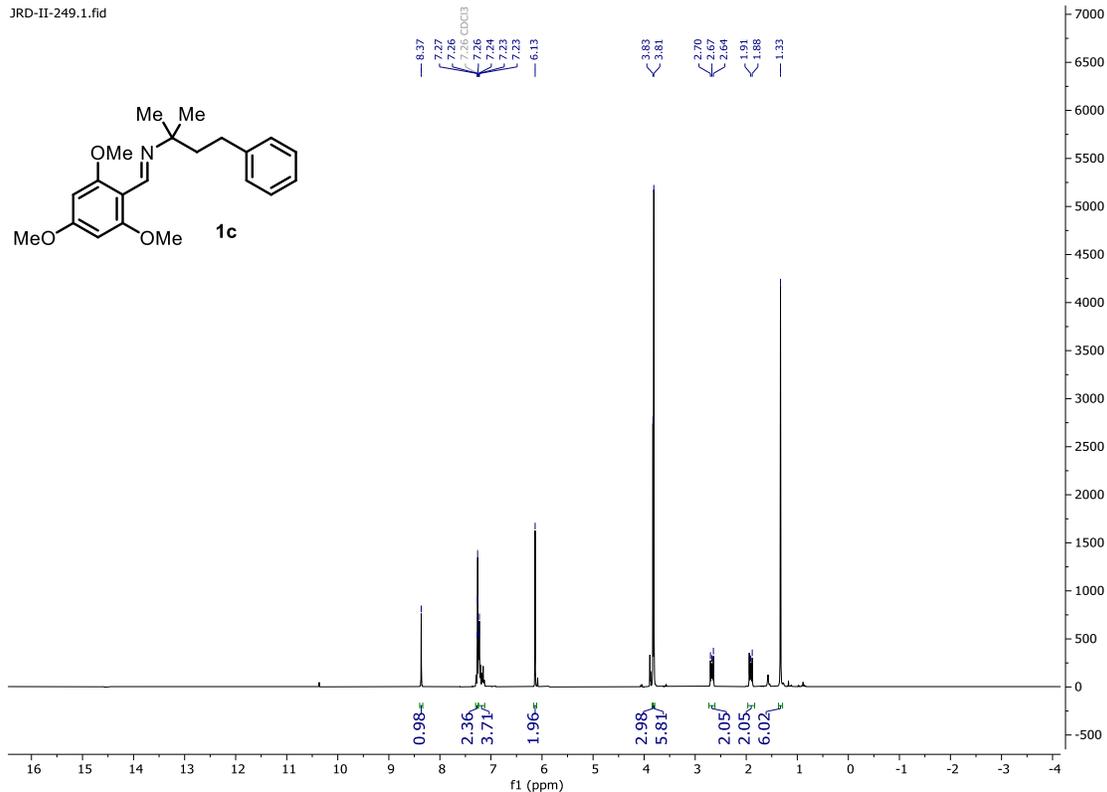
¹⁵N NMR (51 MHz, CDCl₃) δ 33.95.

5. References

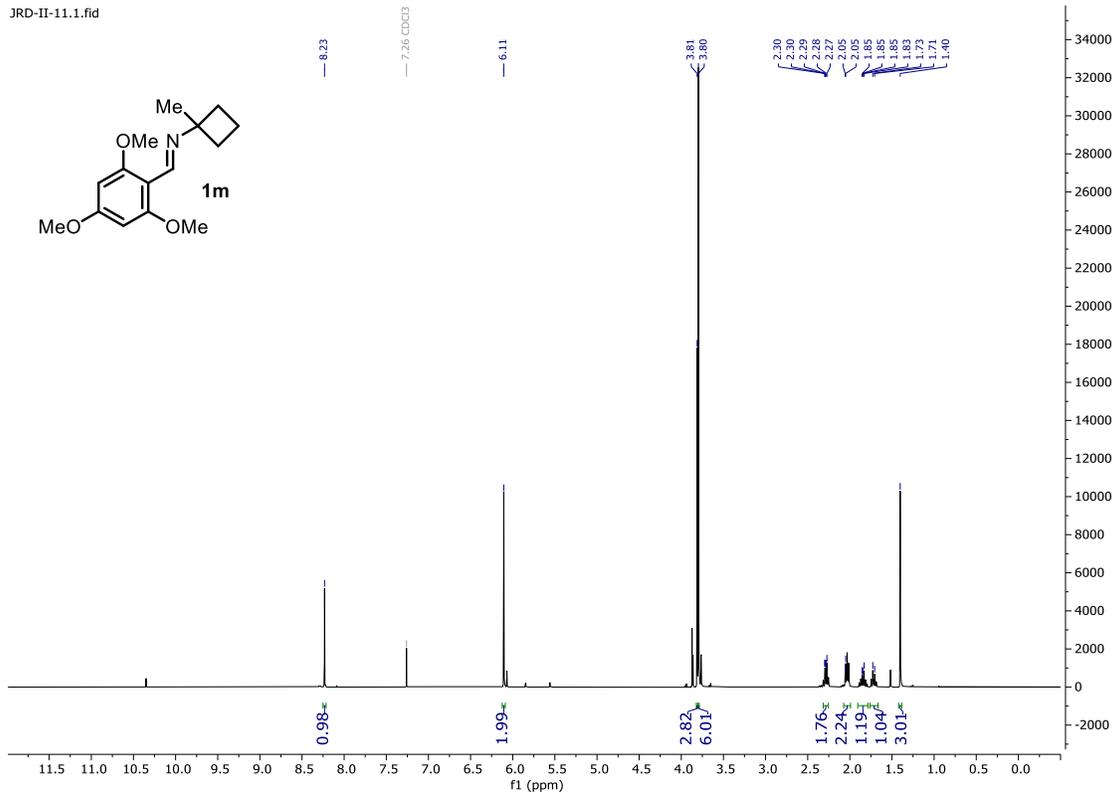
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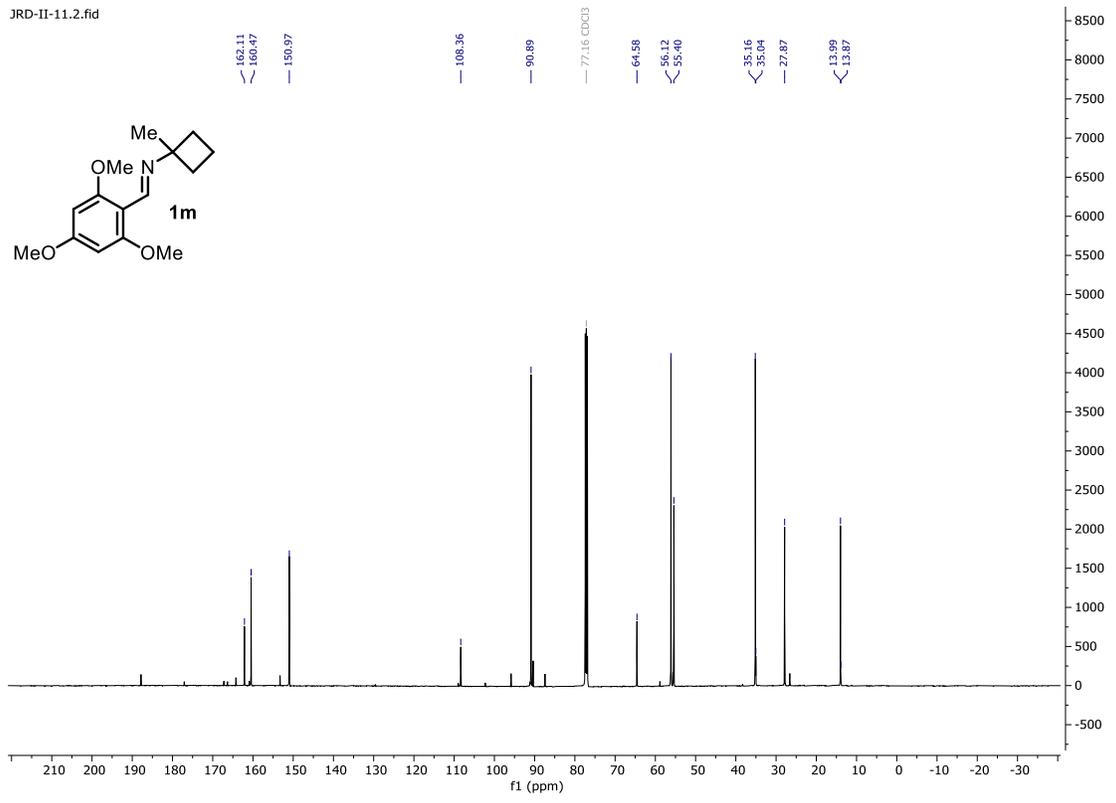
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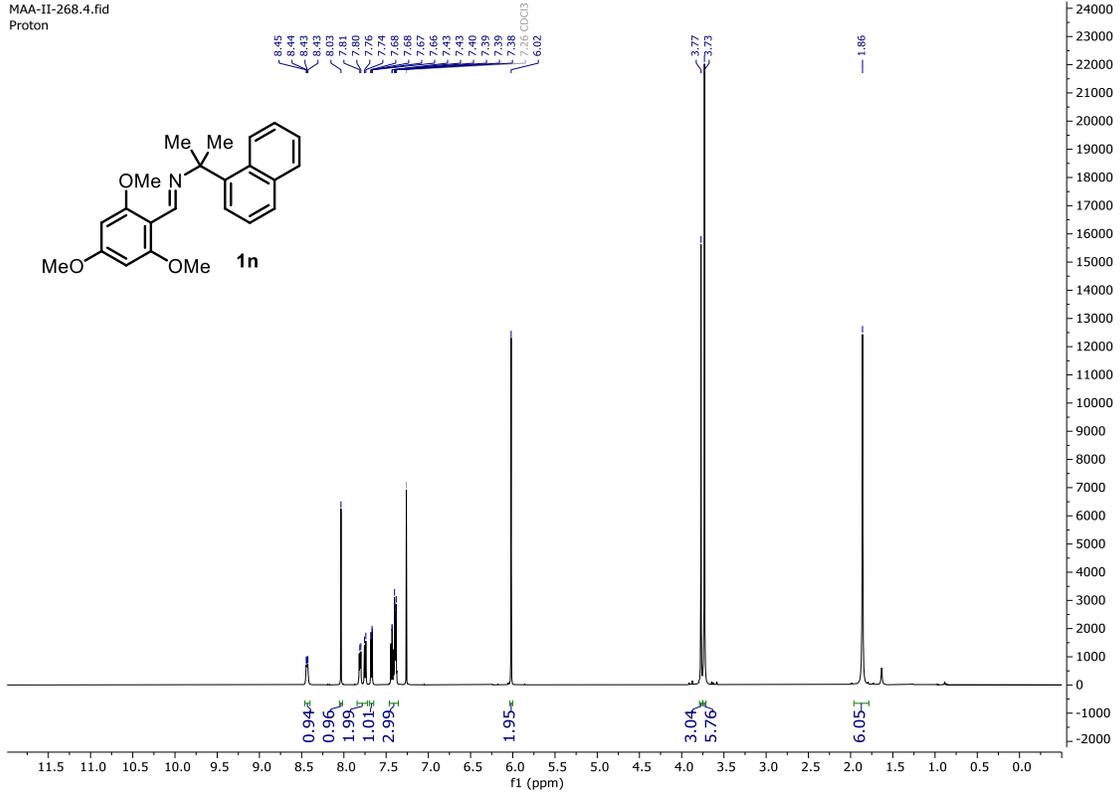
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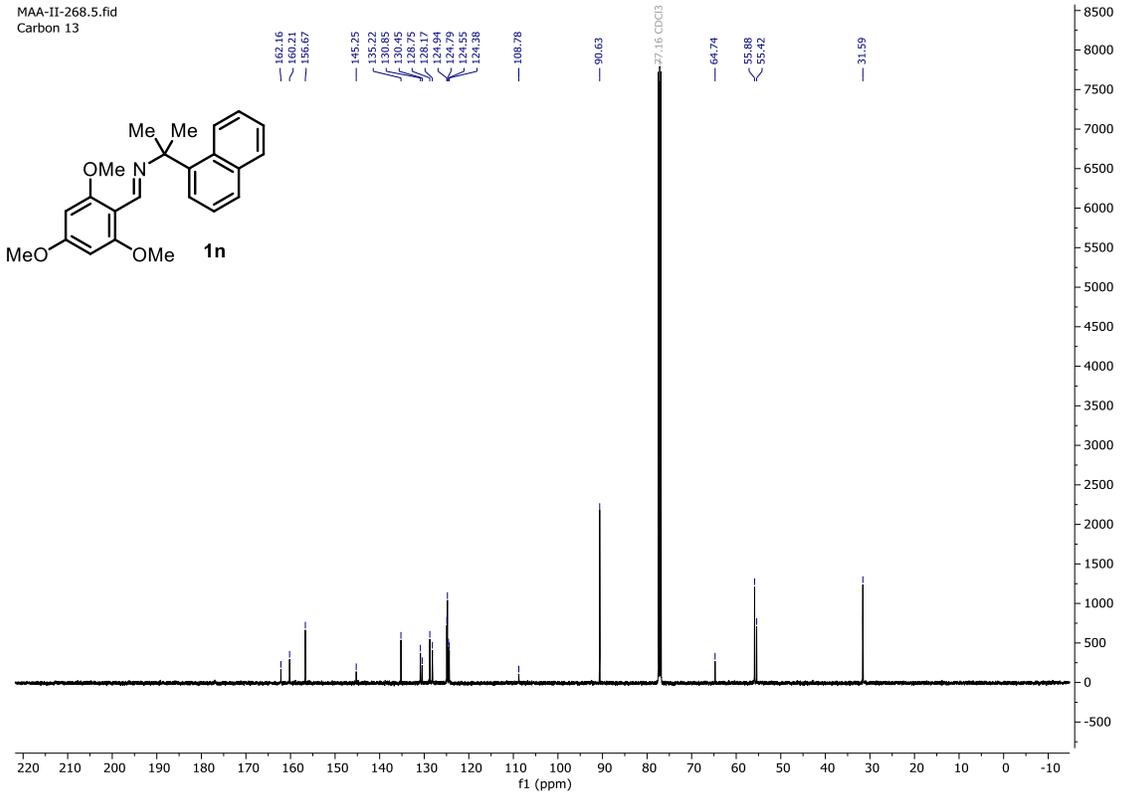
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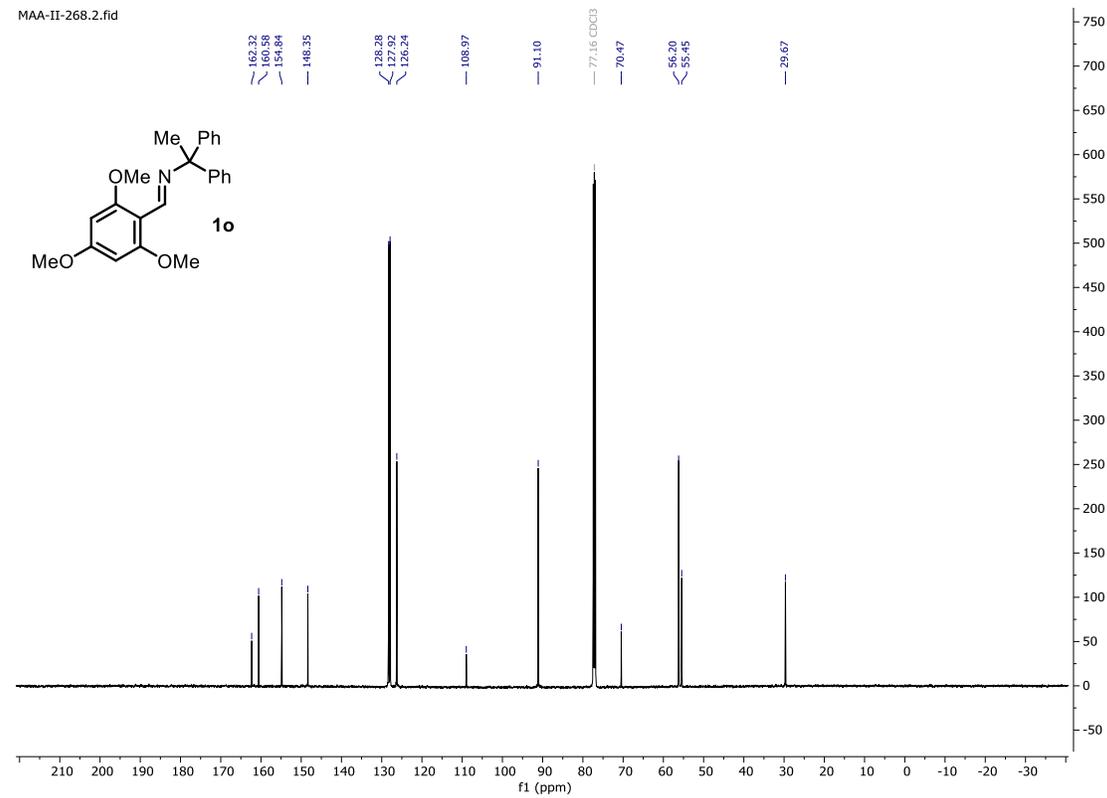
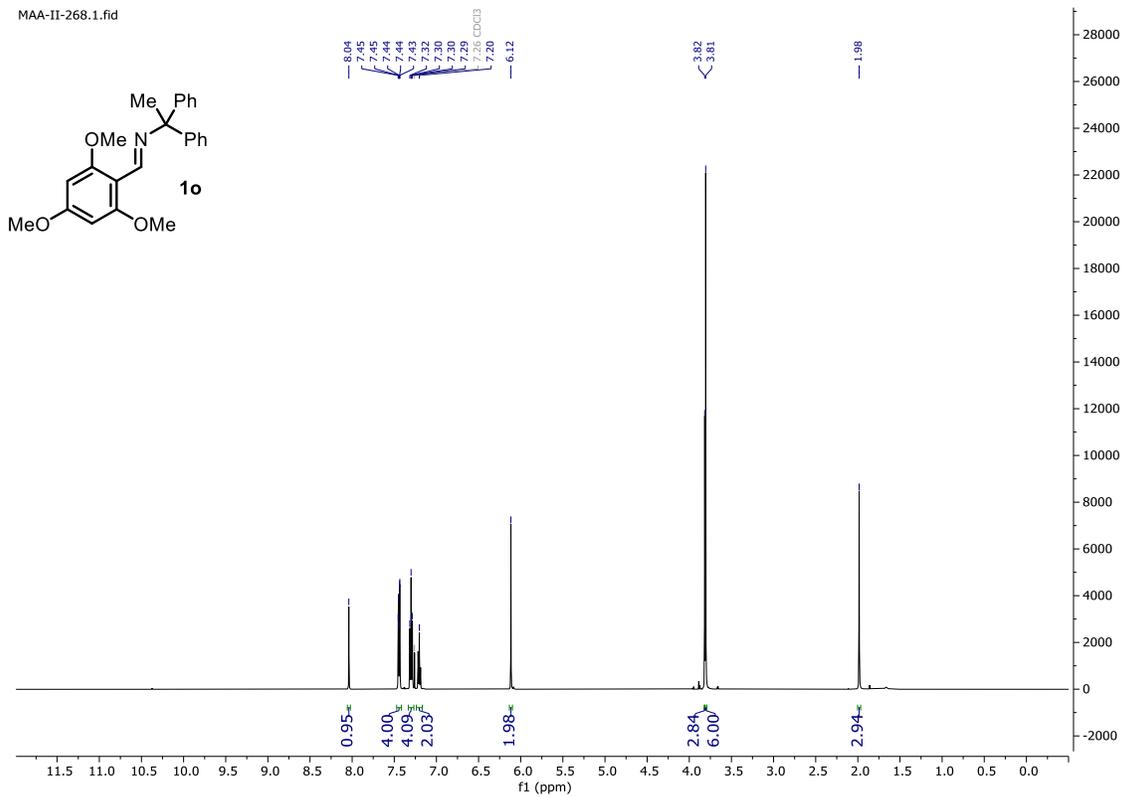


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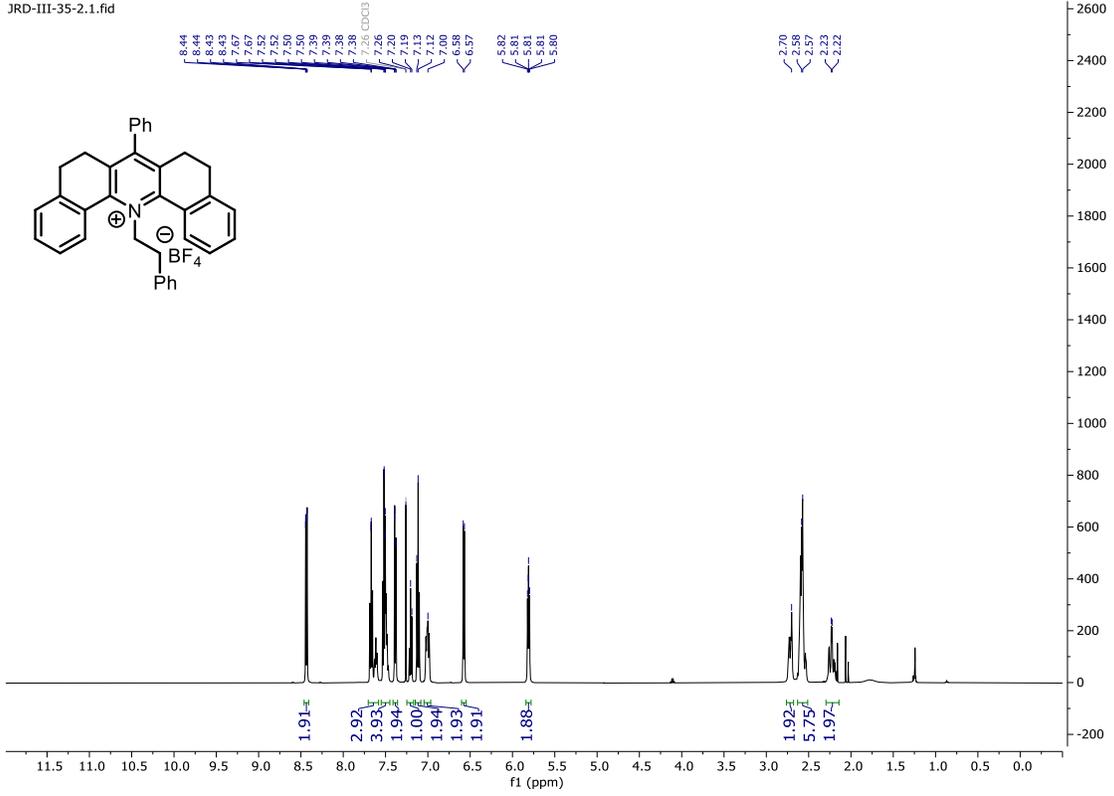


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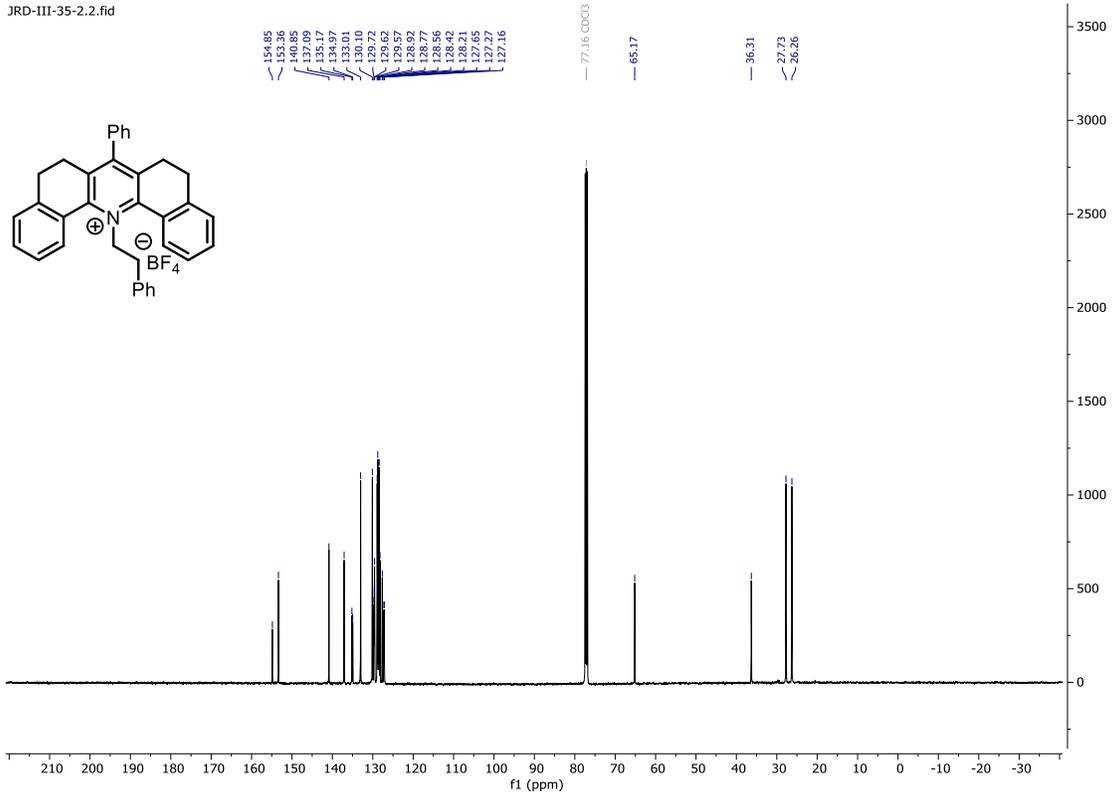




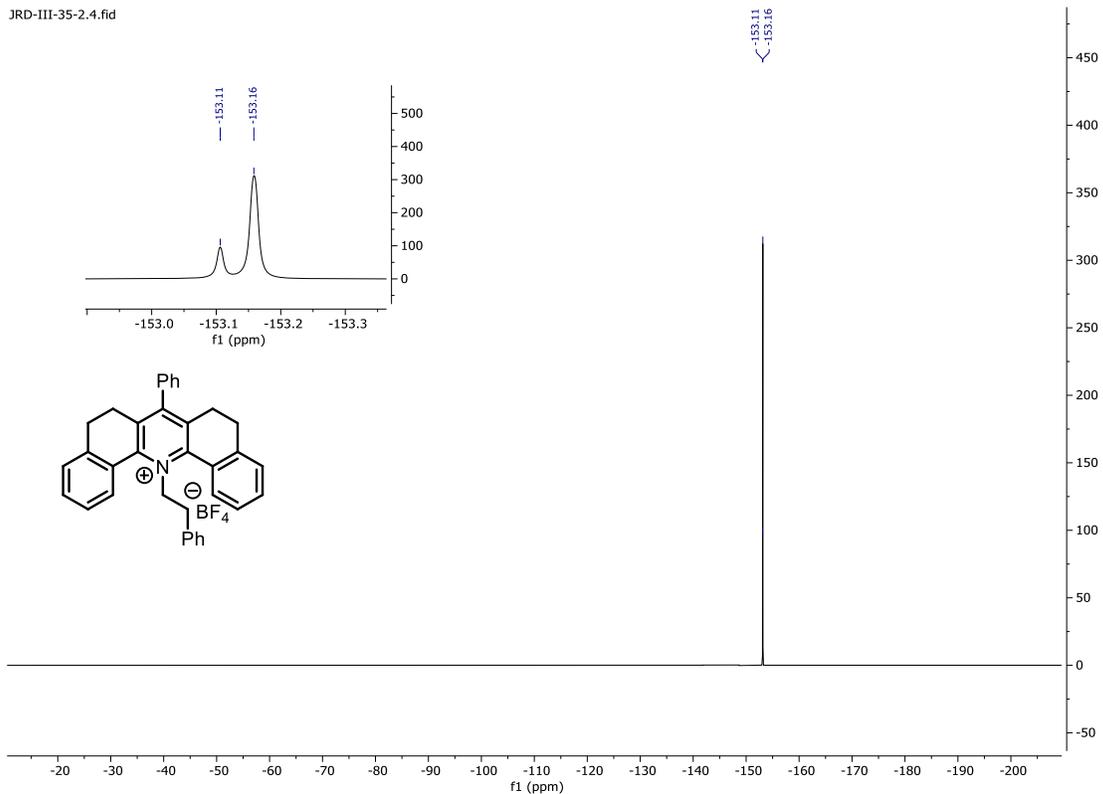
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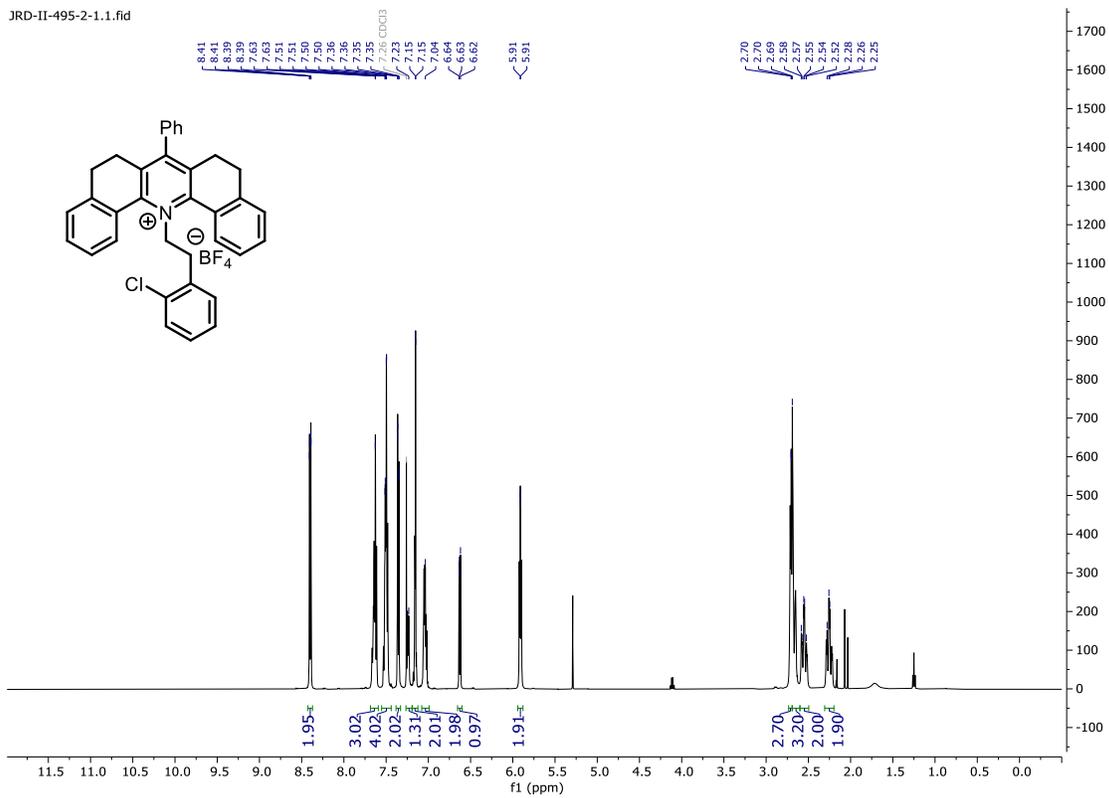
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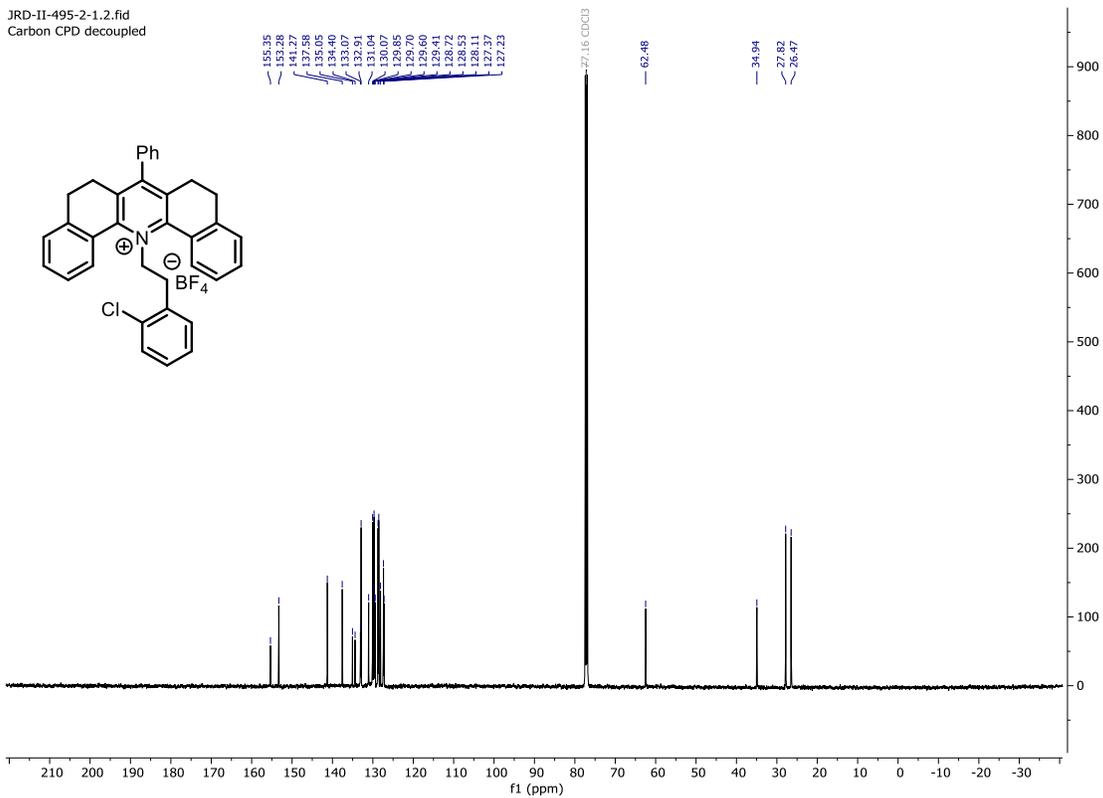
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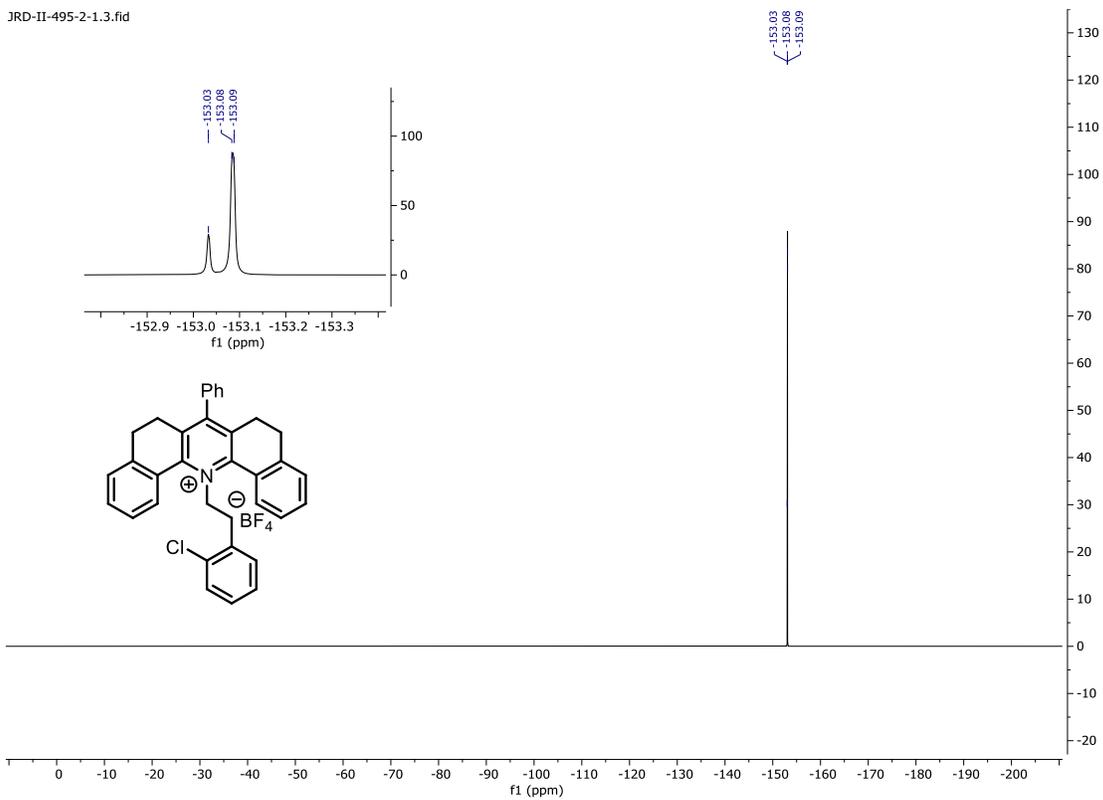
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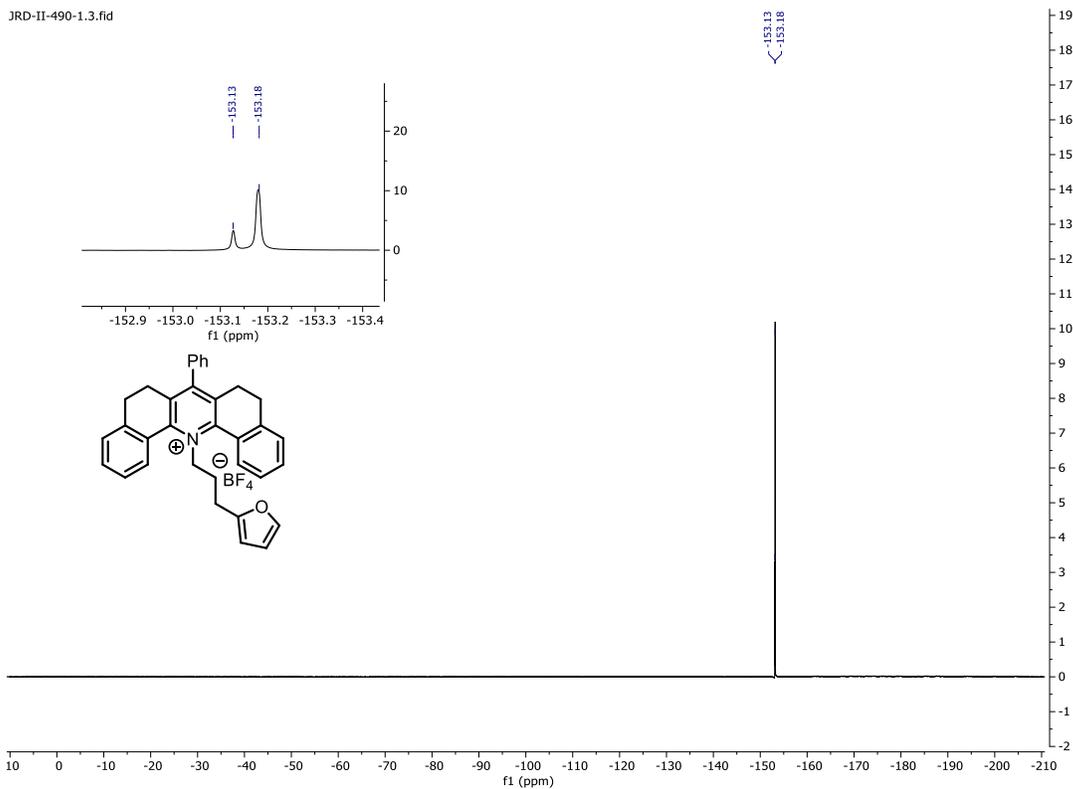
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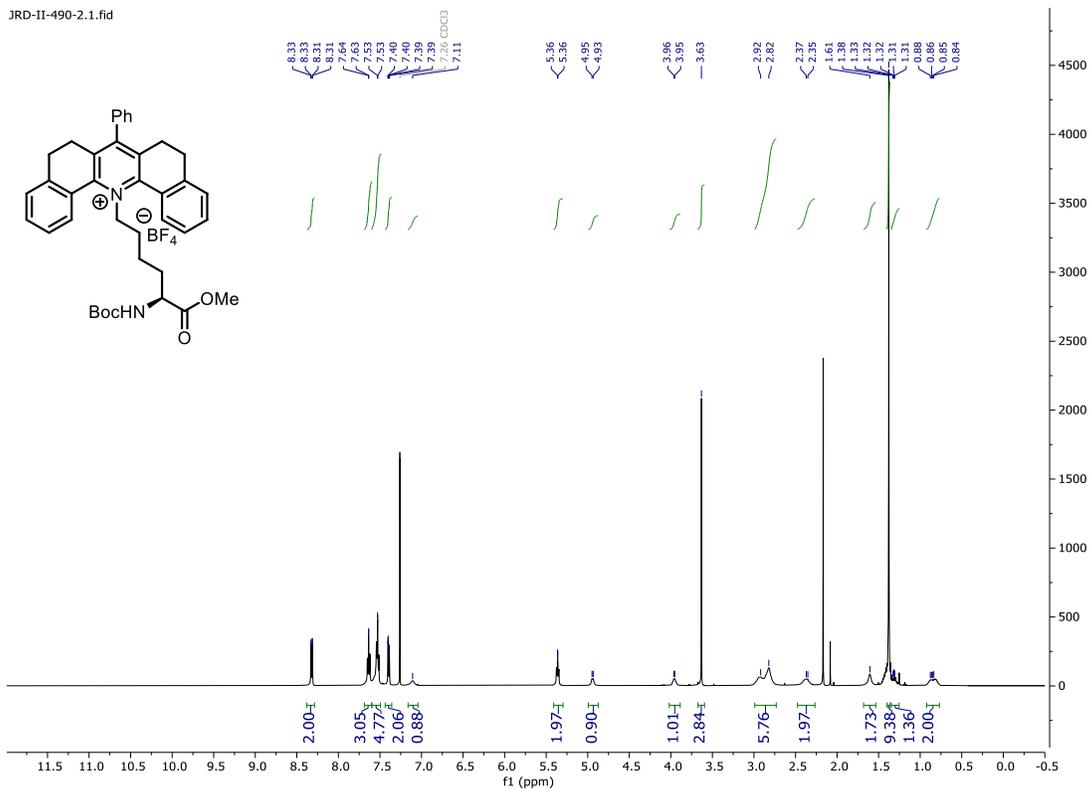
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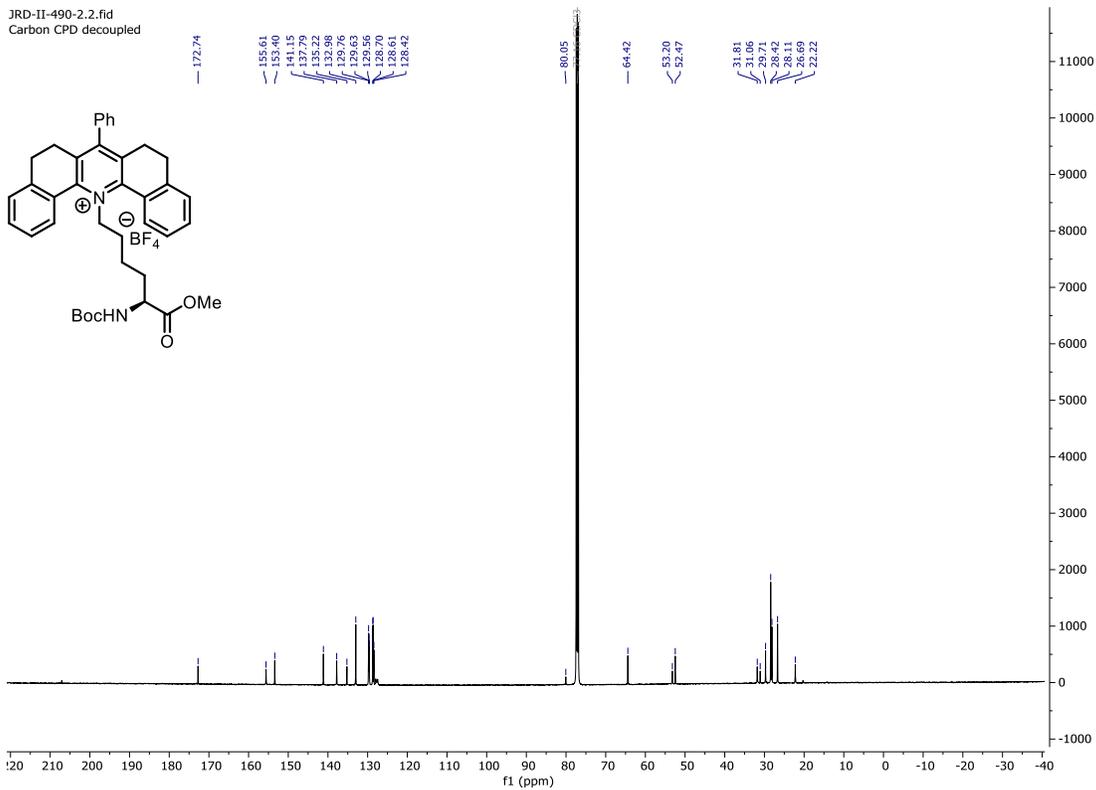
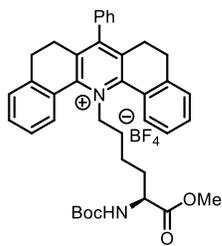
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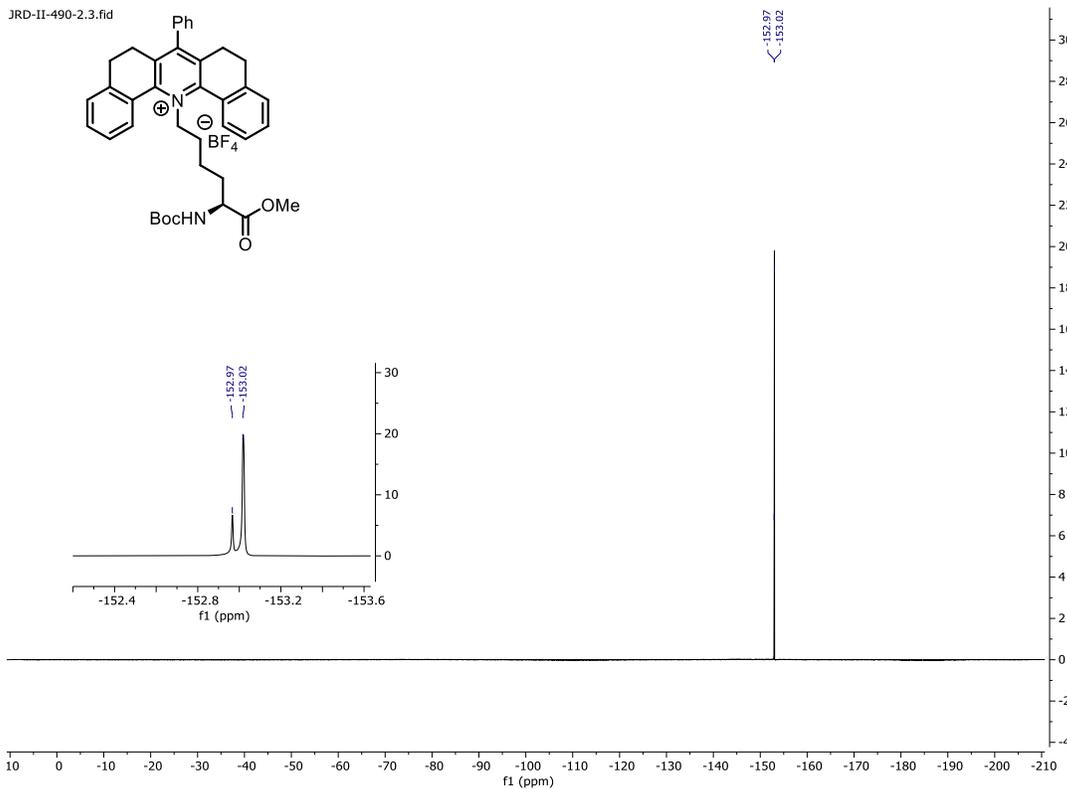
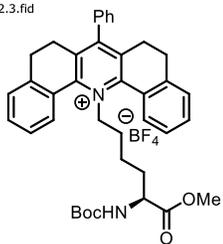
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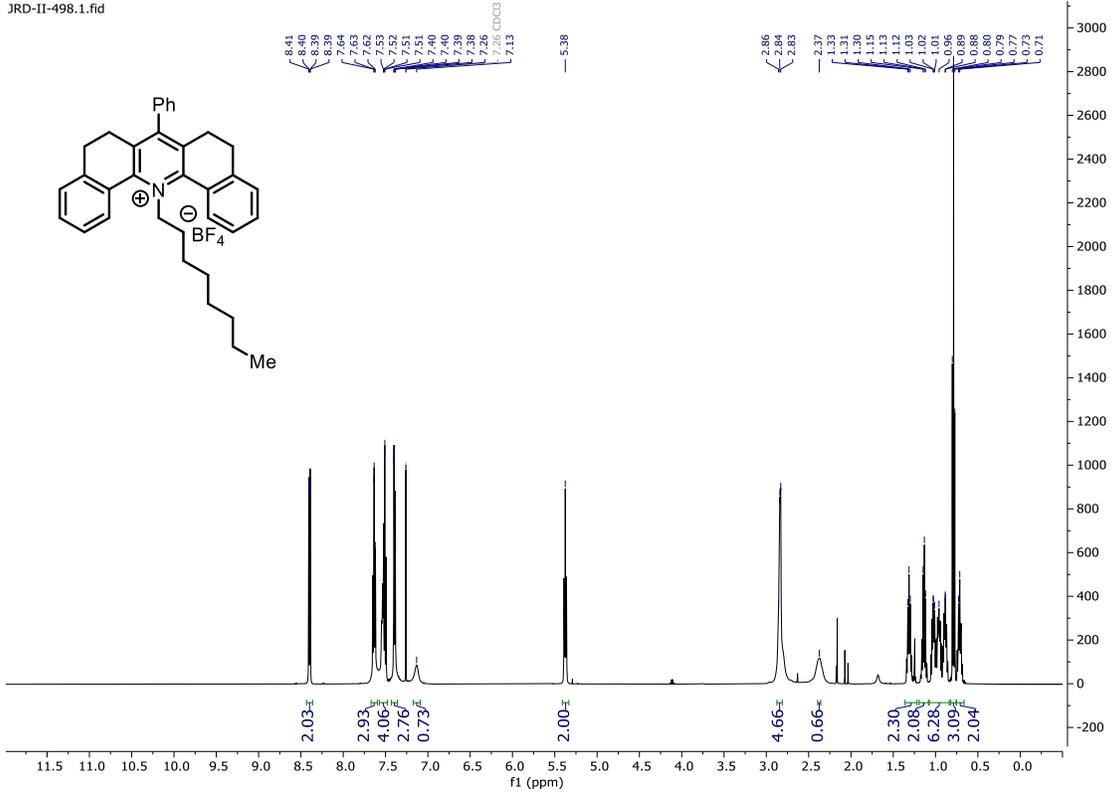
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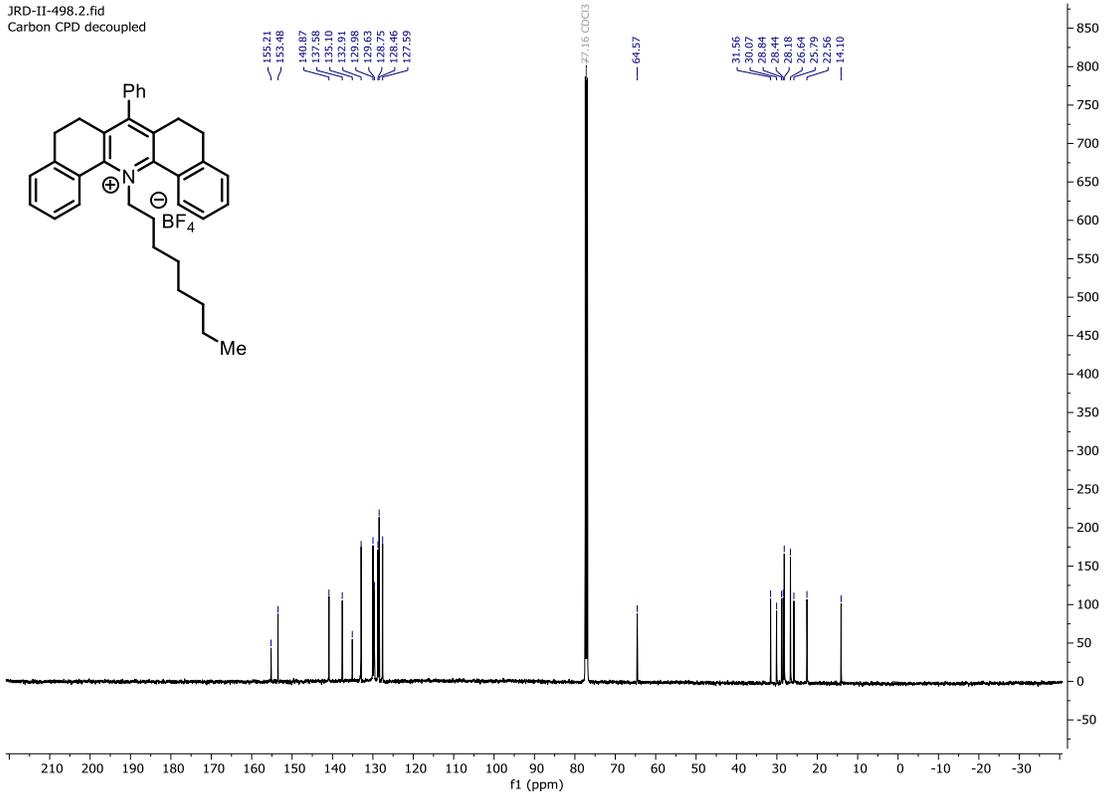
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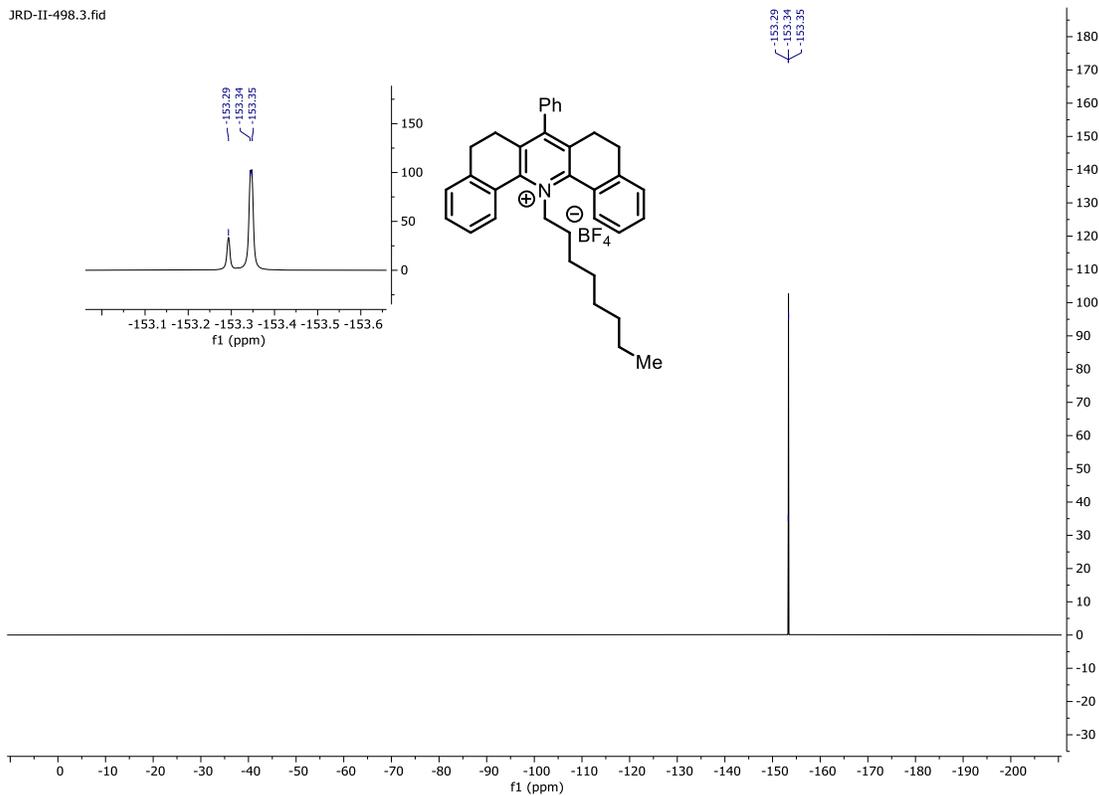
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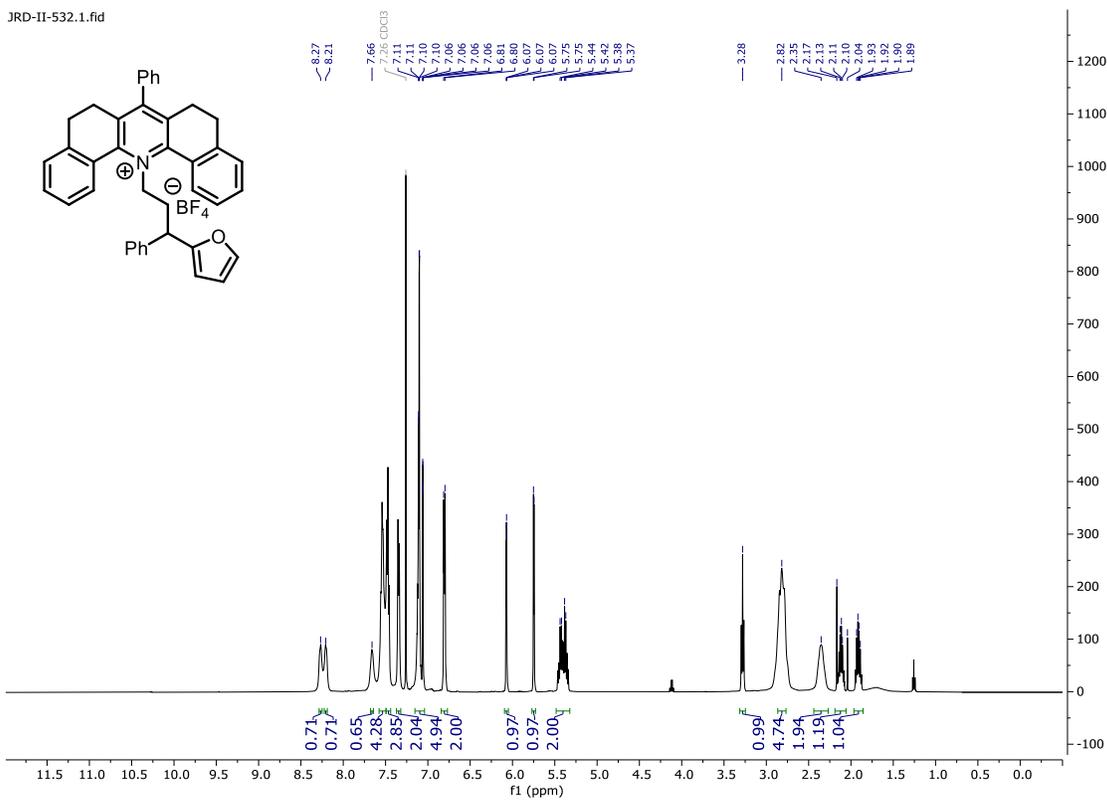
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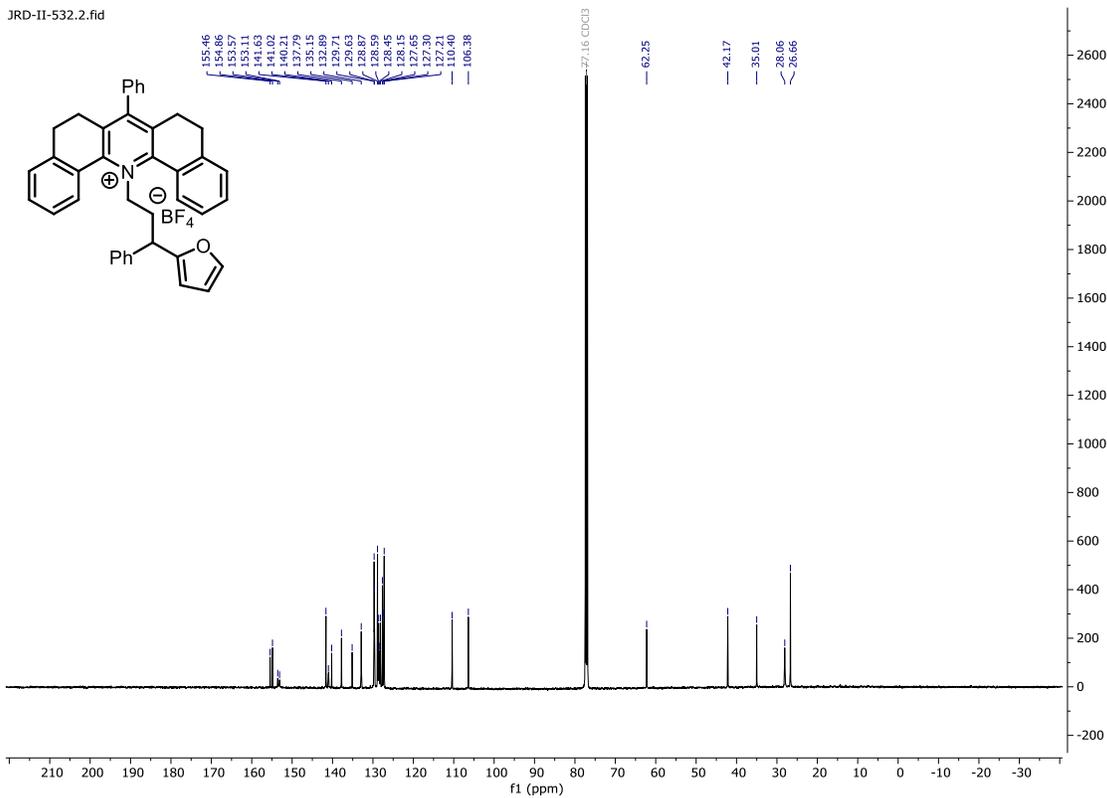
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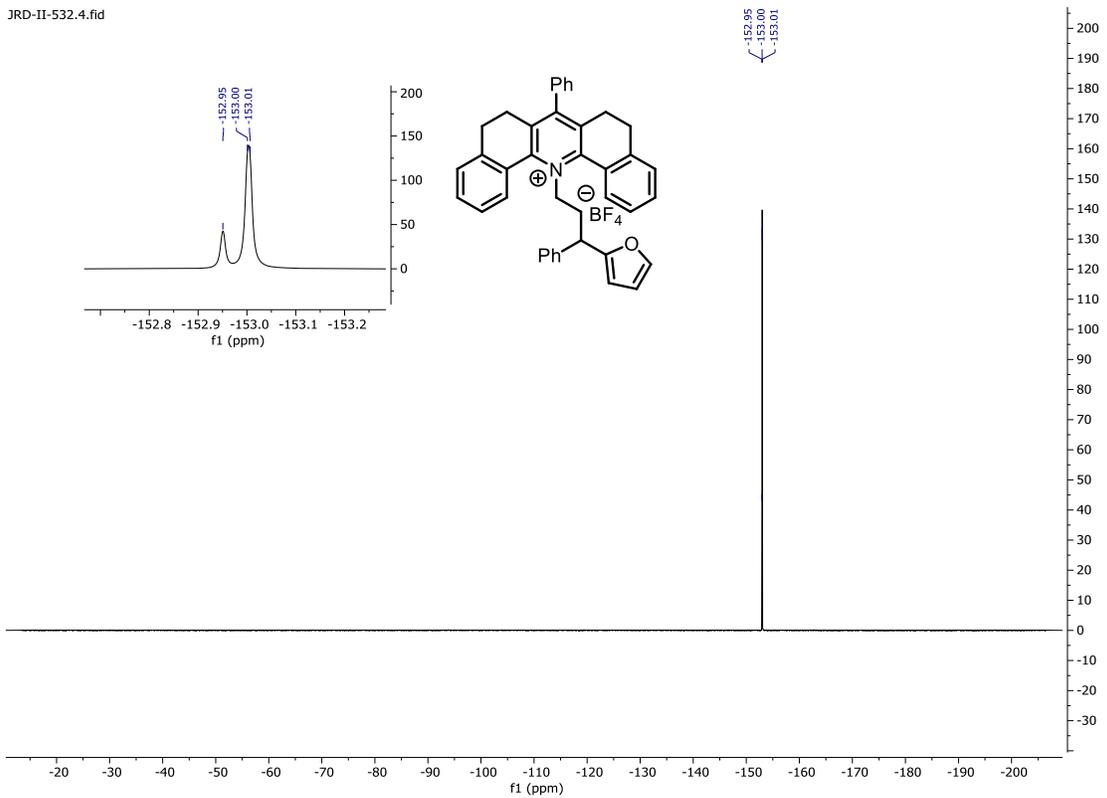
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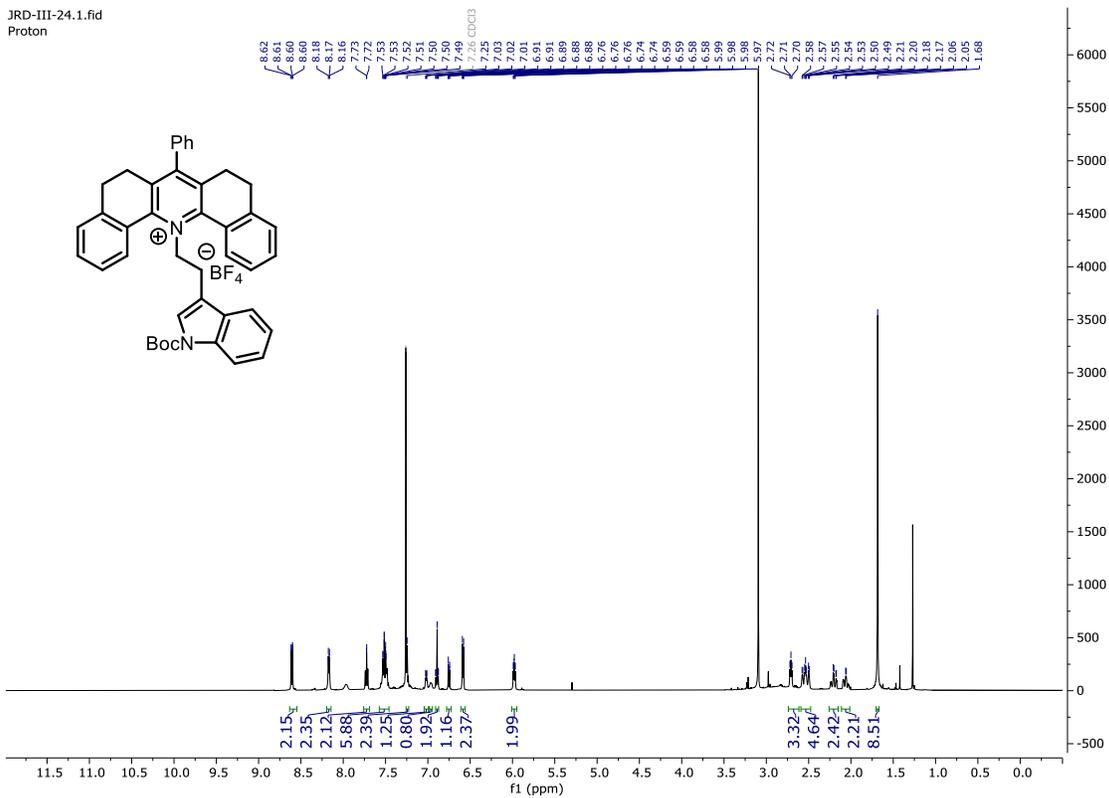
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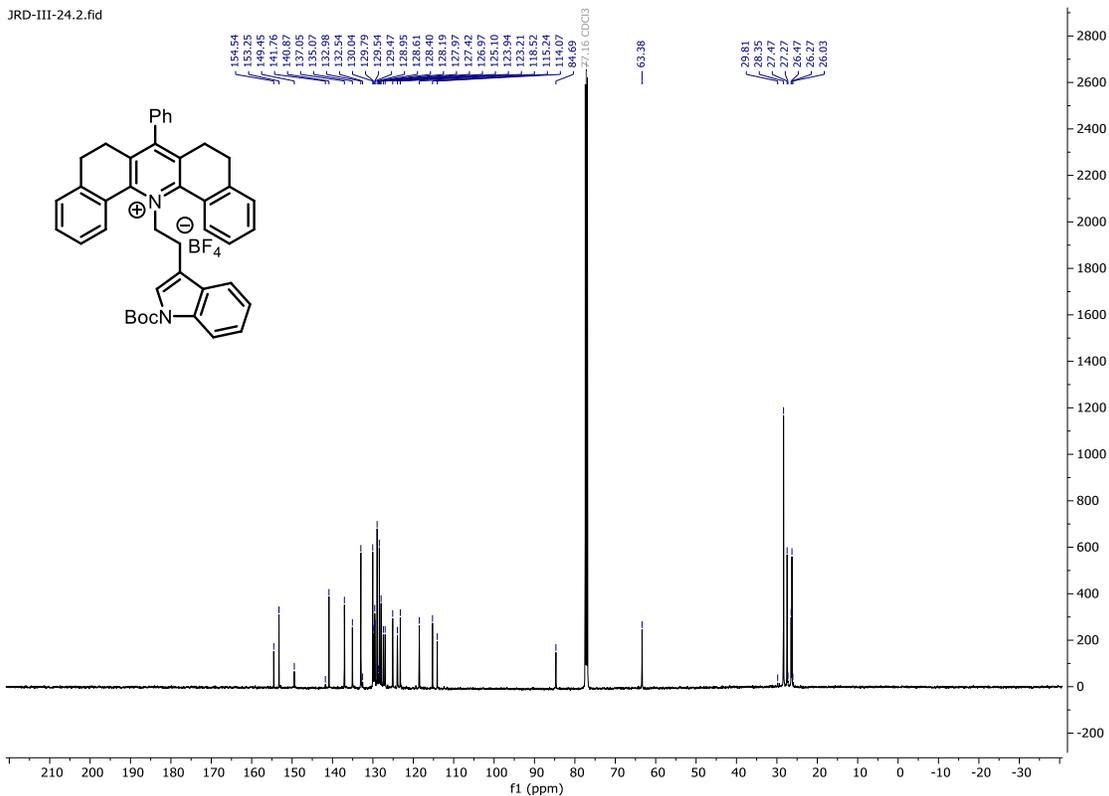
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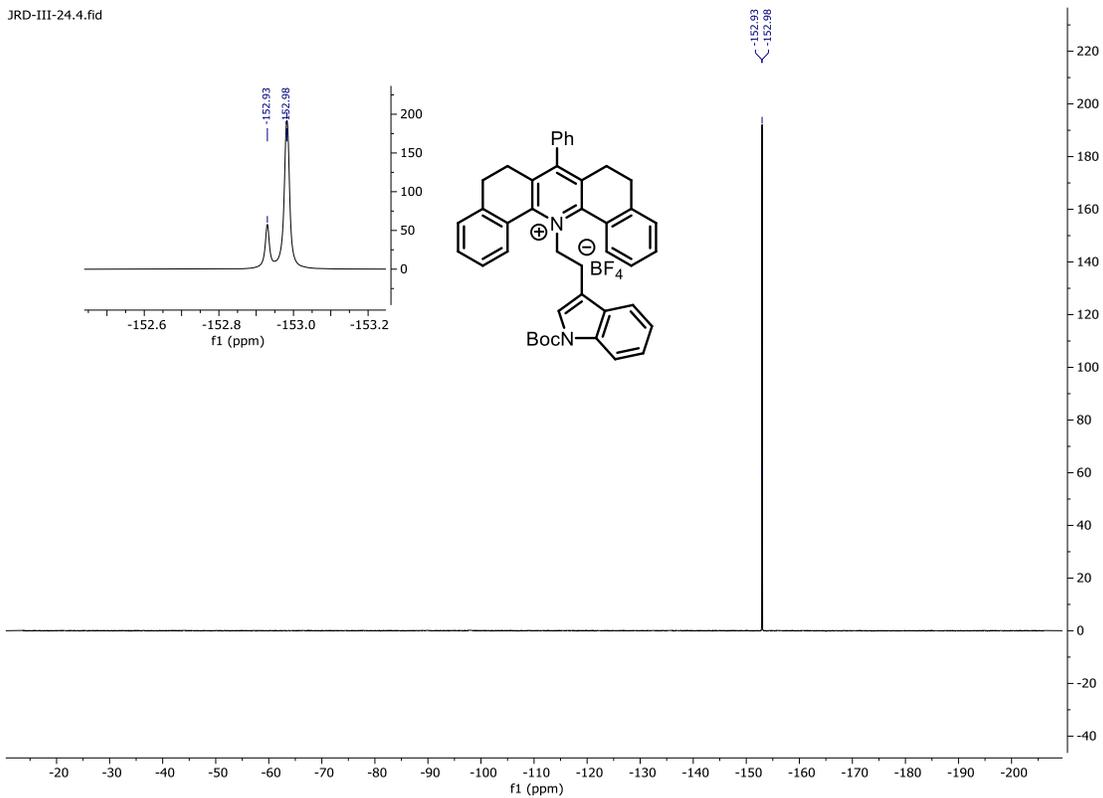
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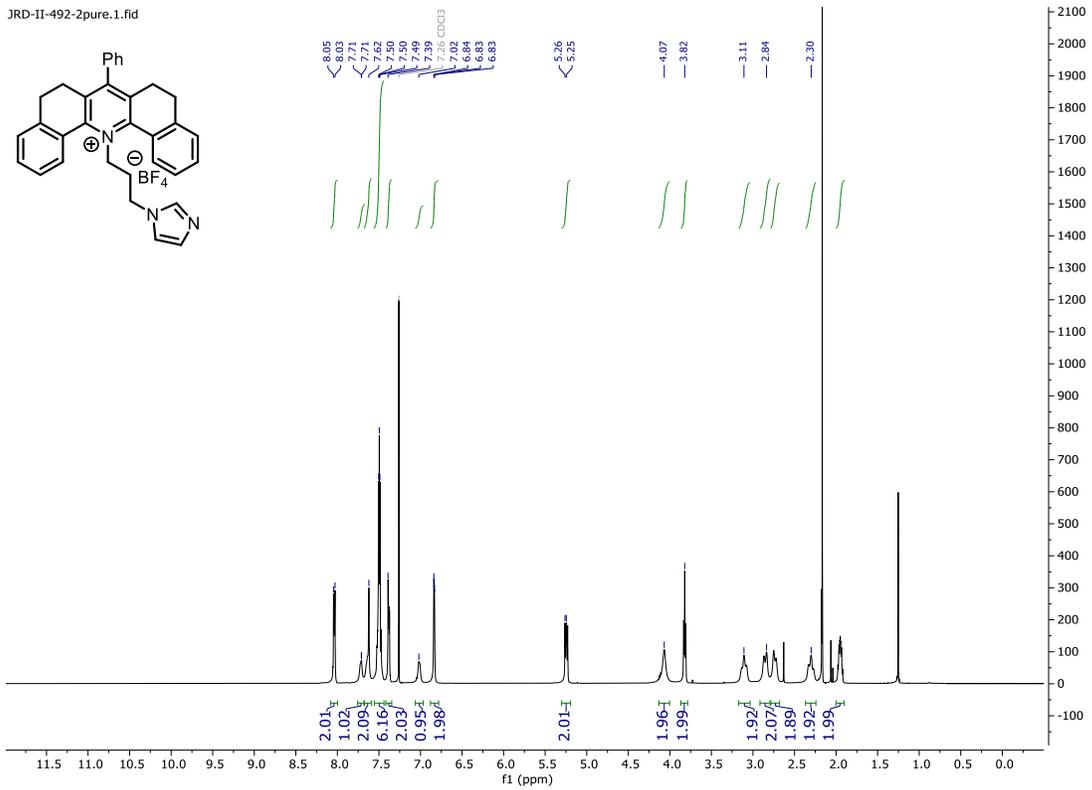
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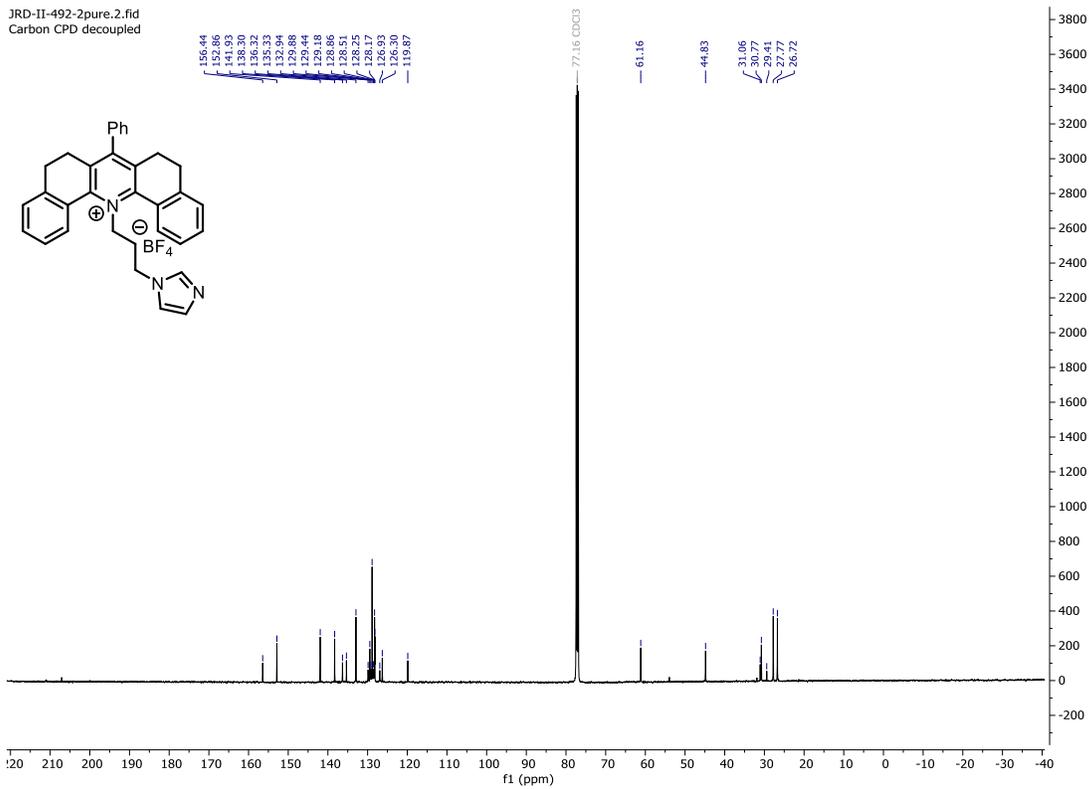
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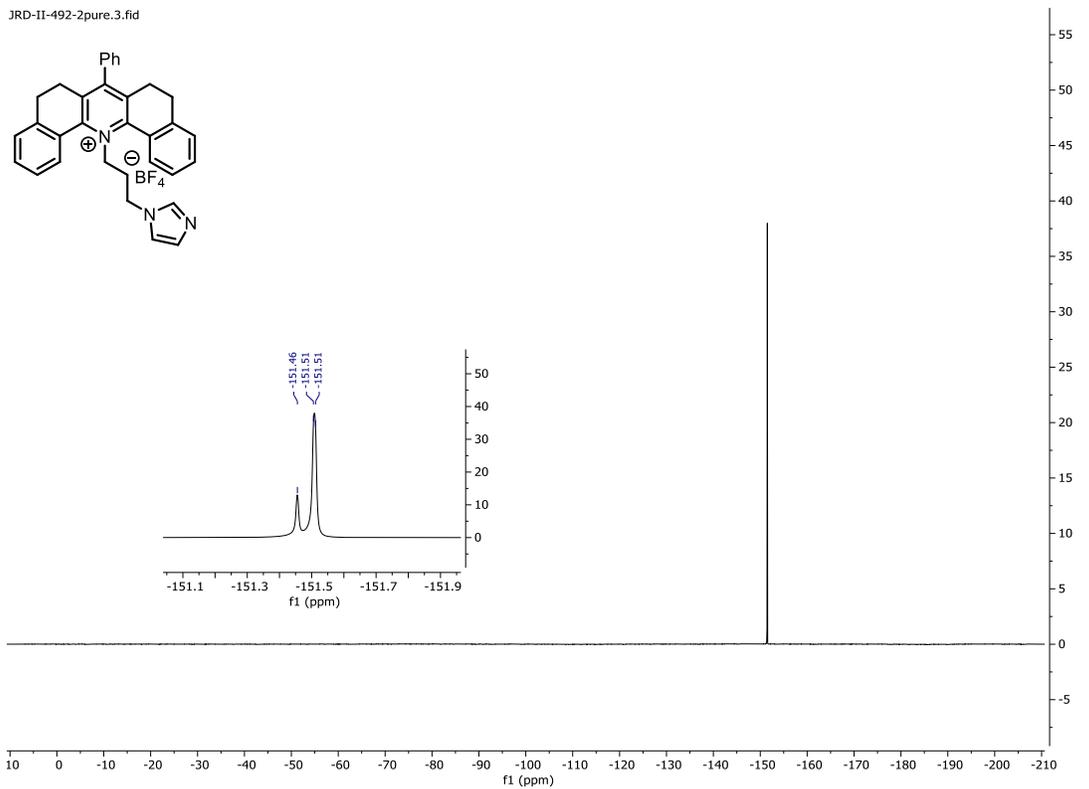
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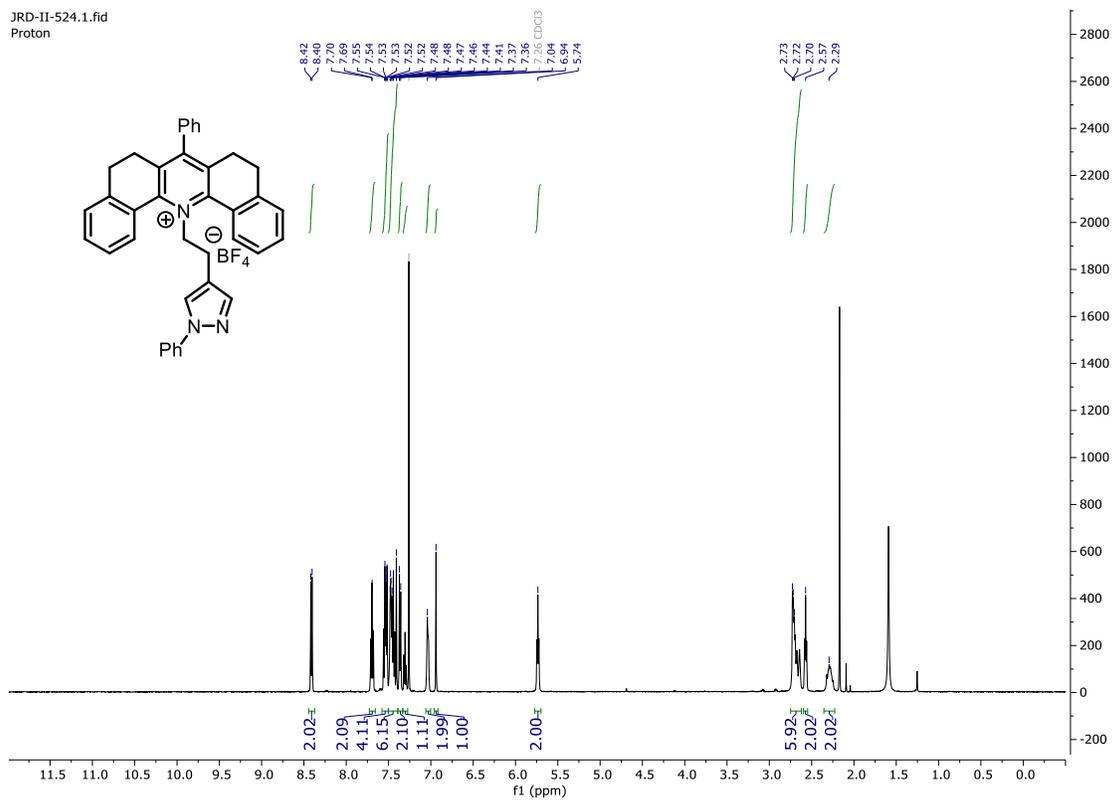
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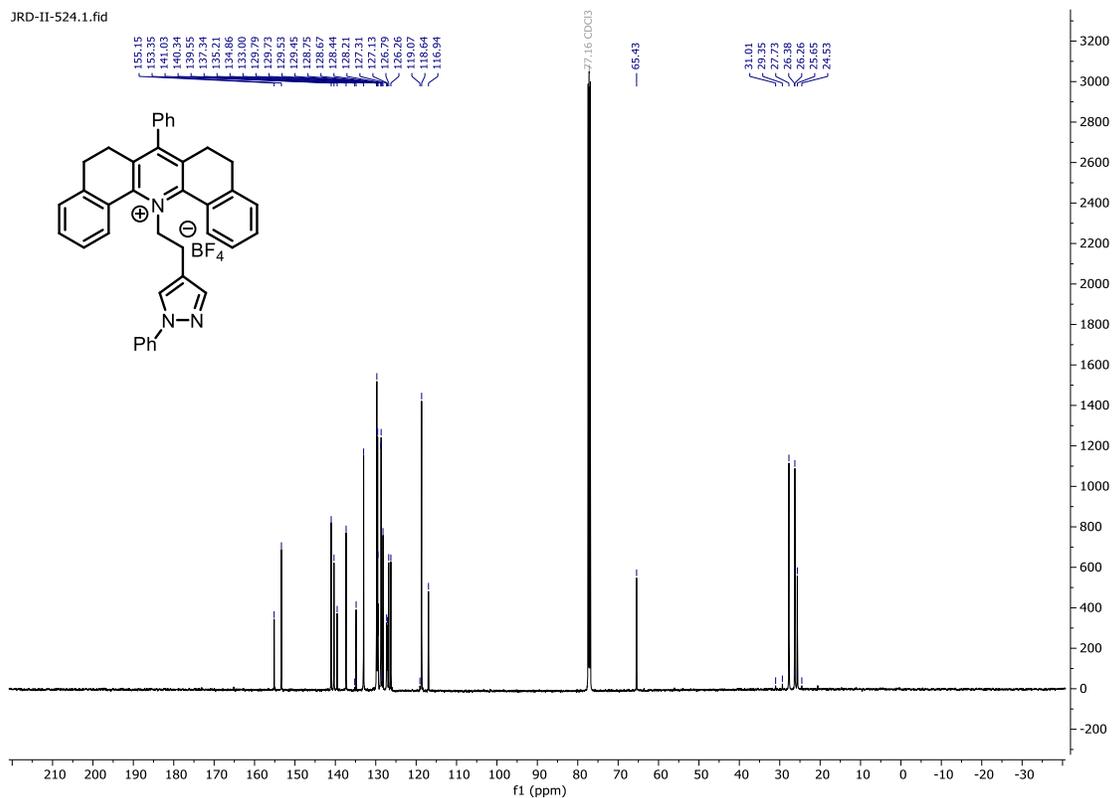
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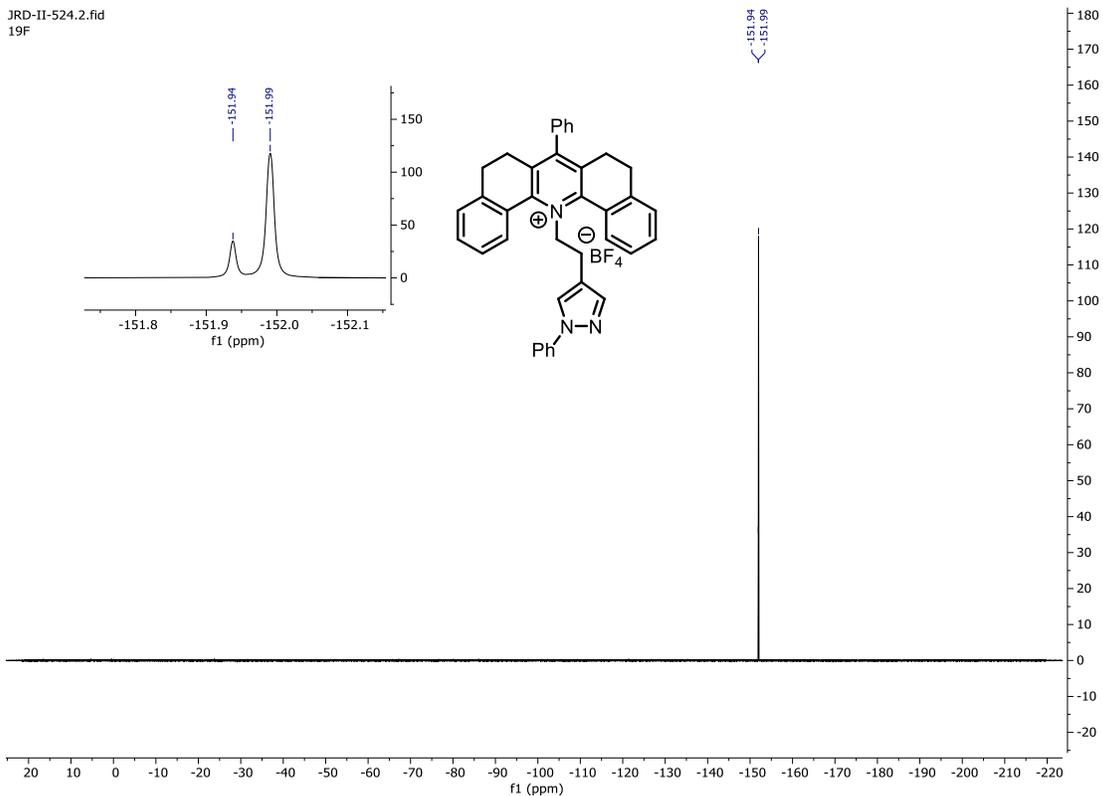
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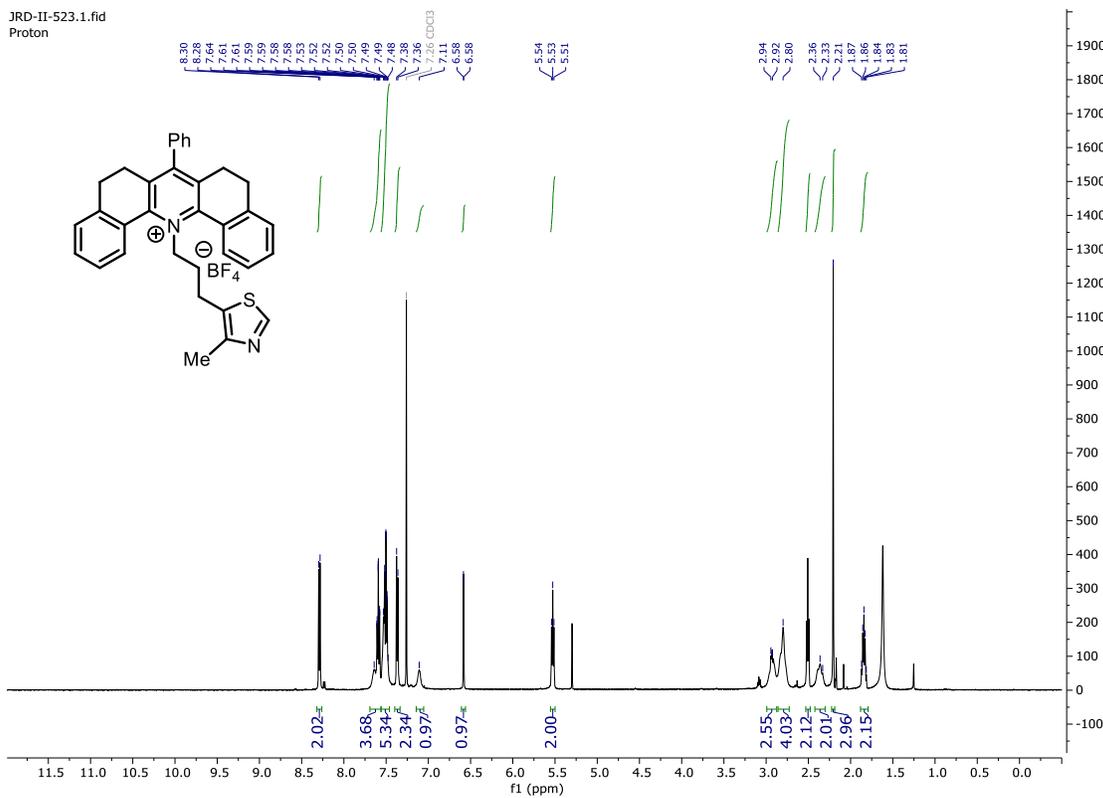
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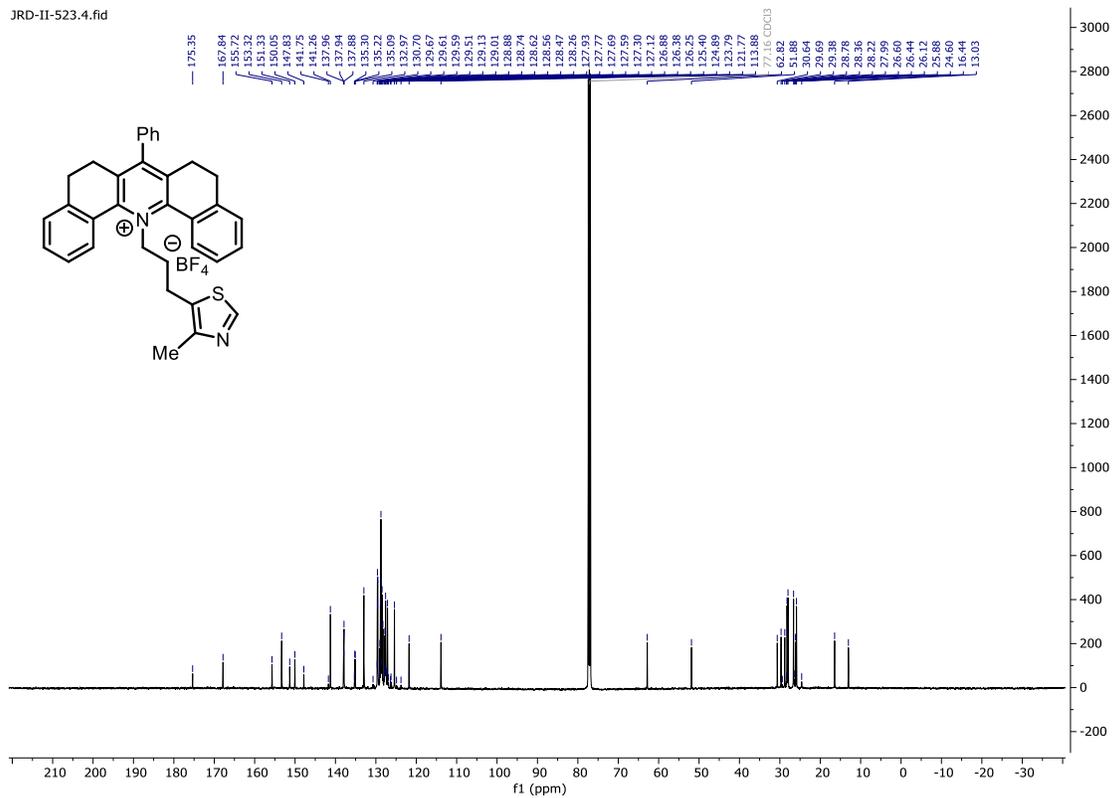
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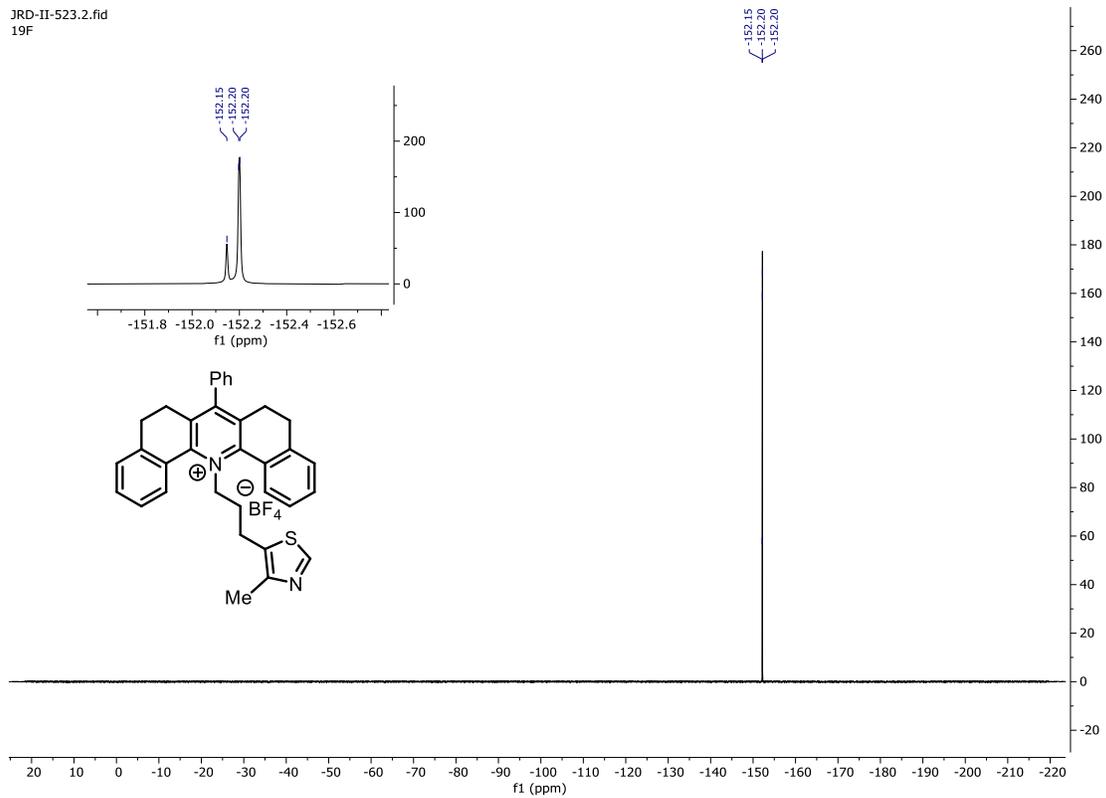


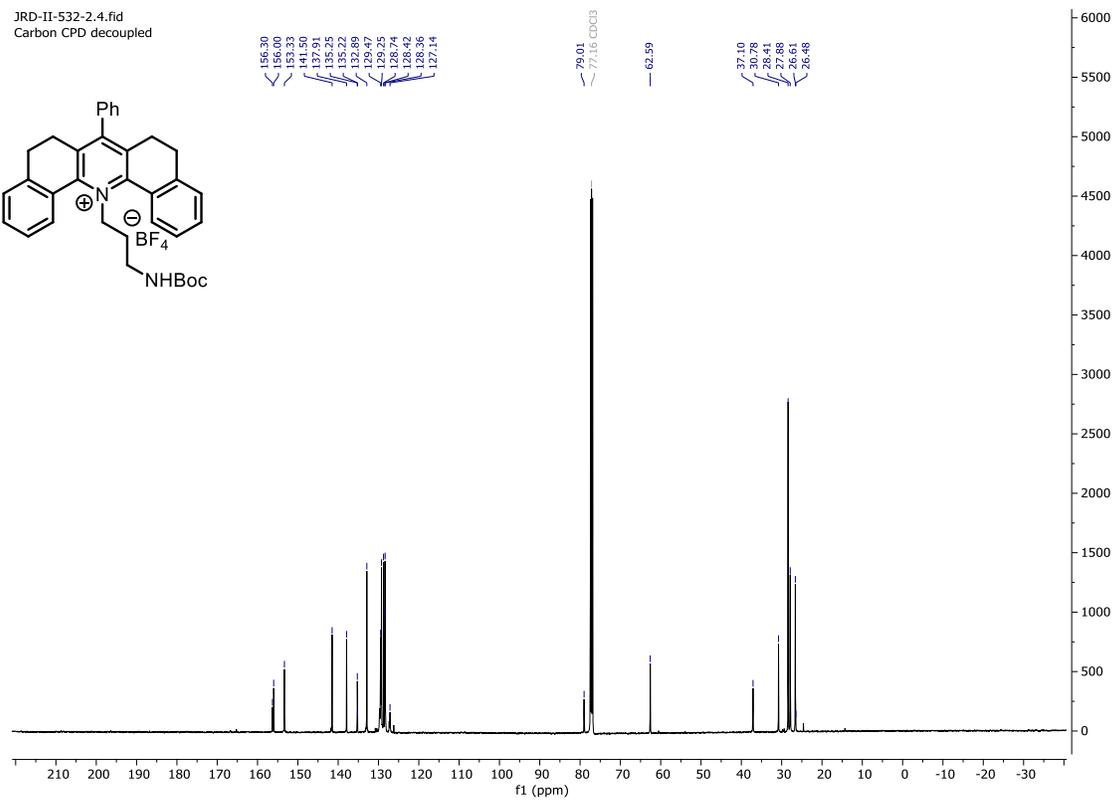
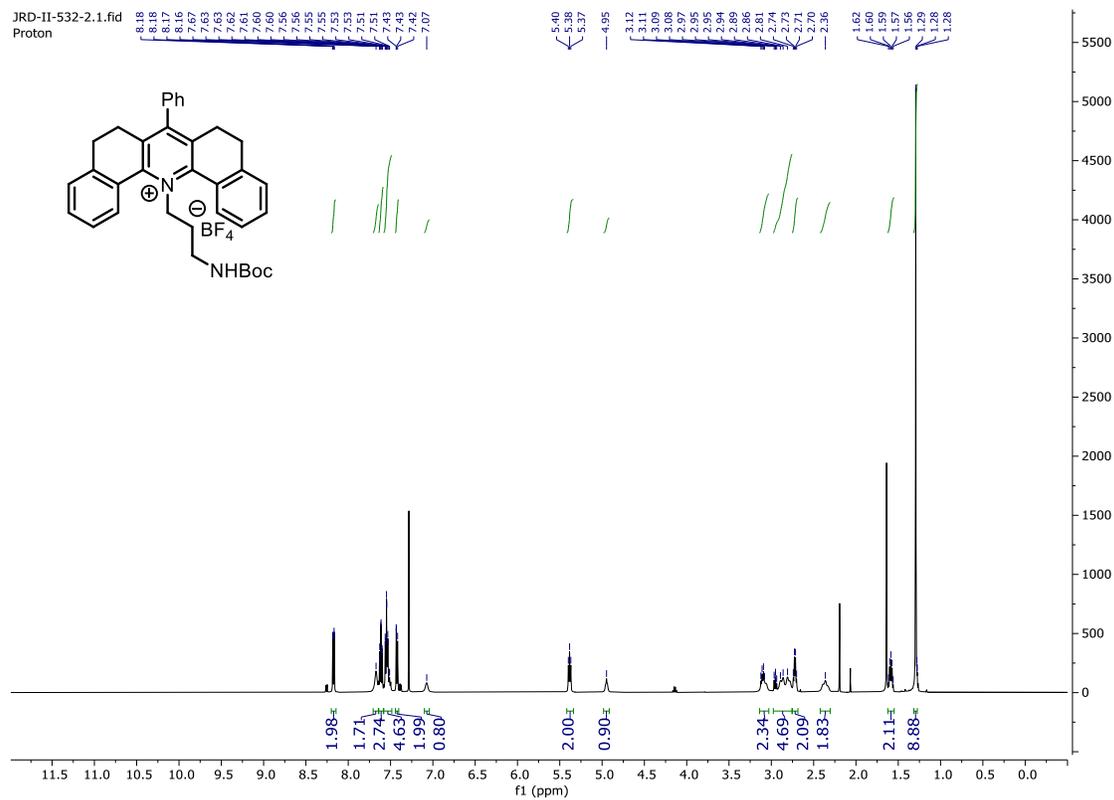
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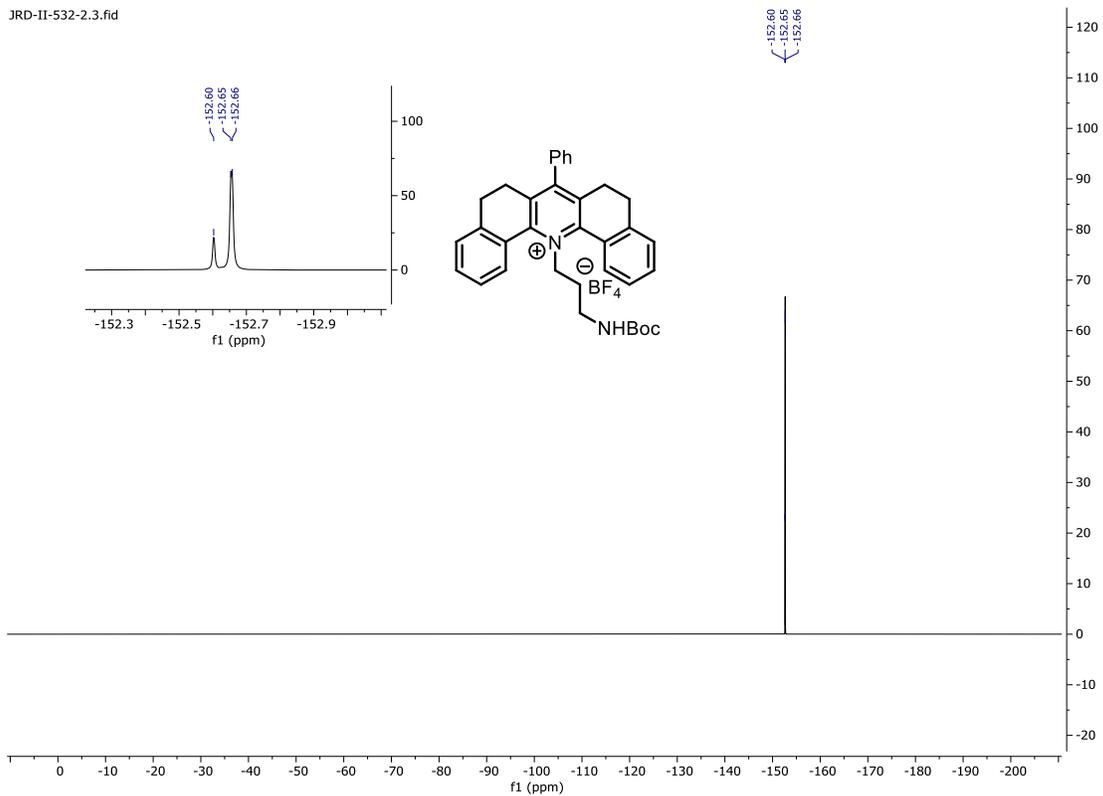
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19F

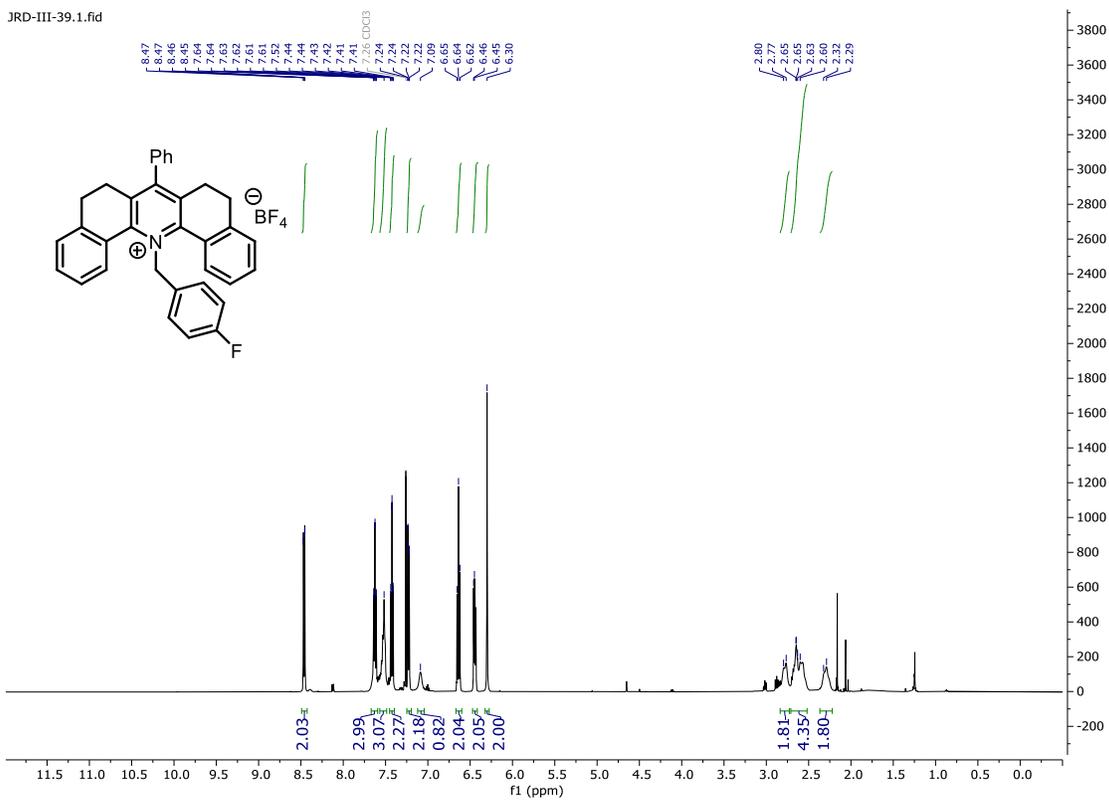


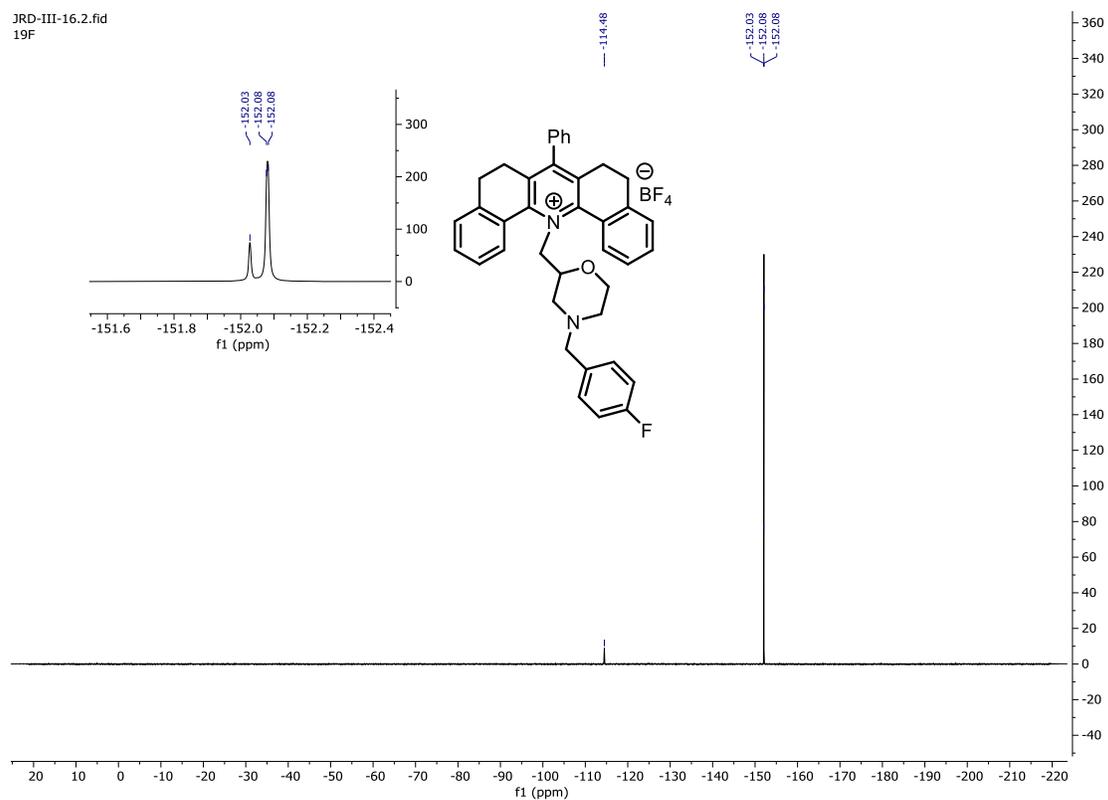
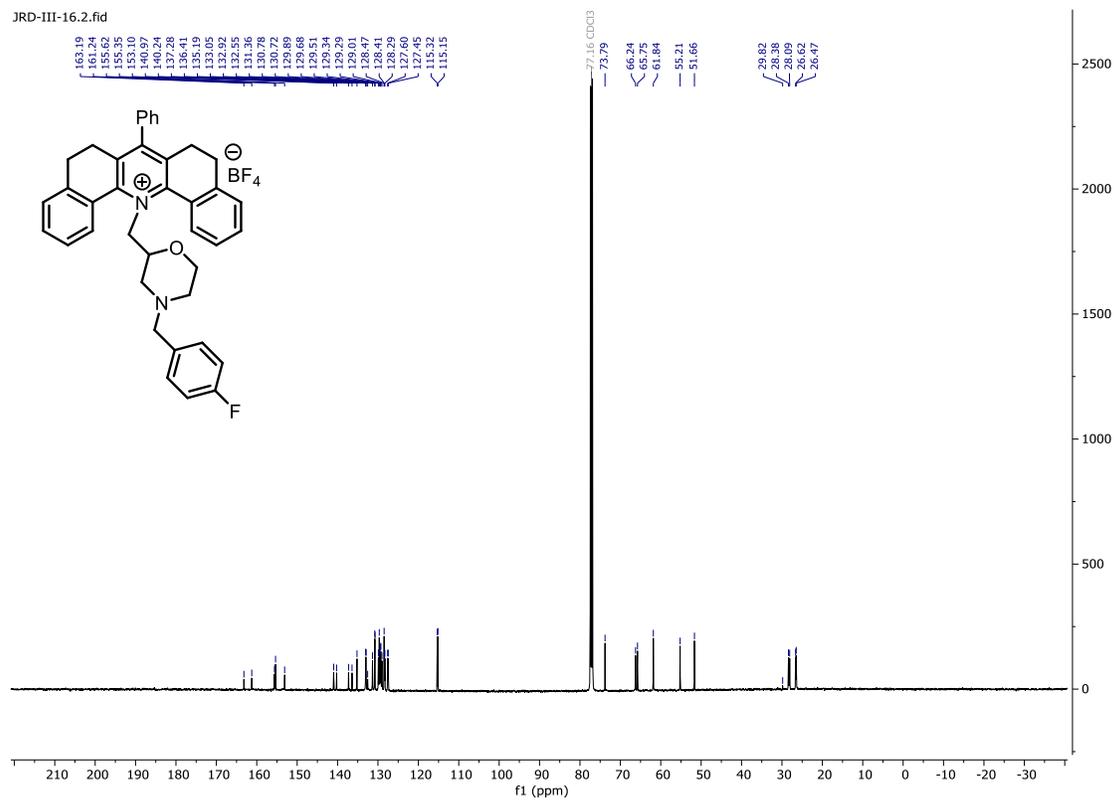


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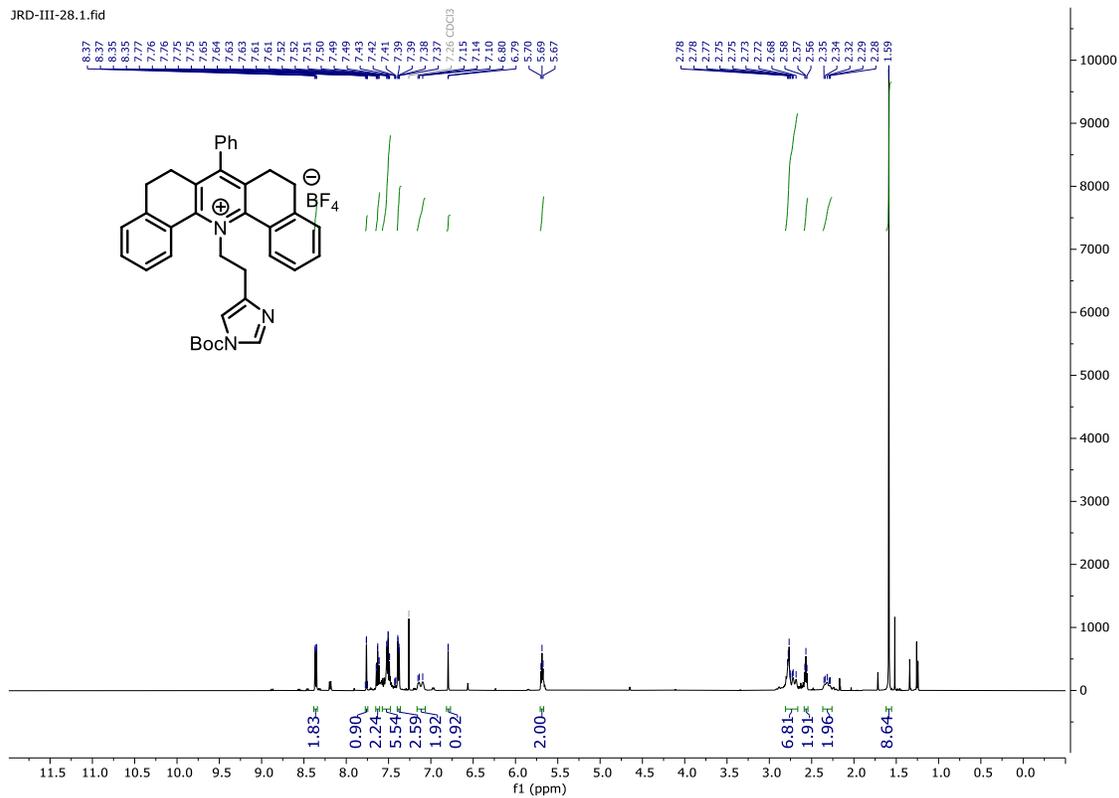


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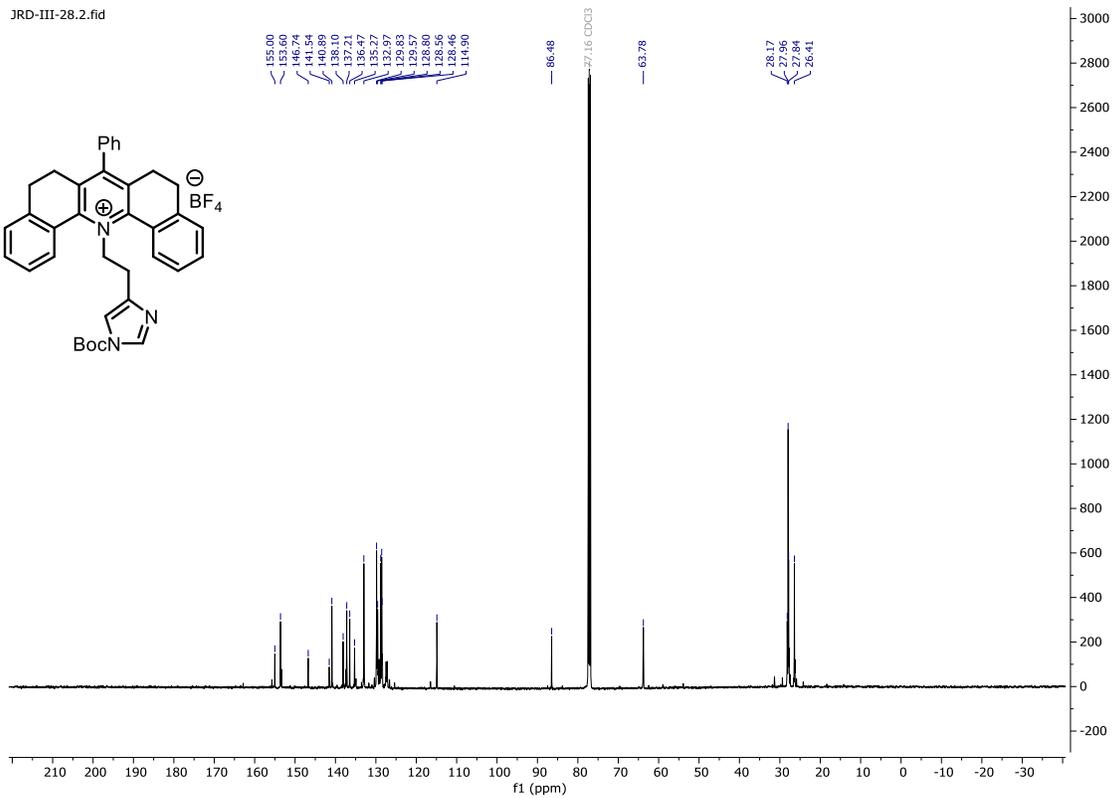




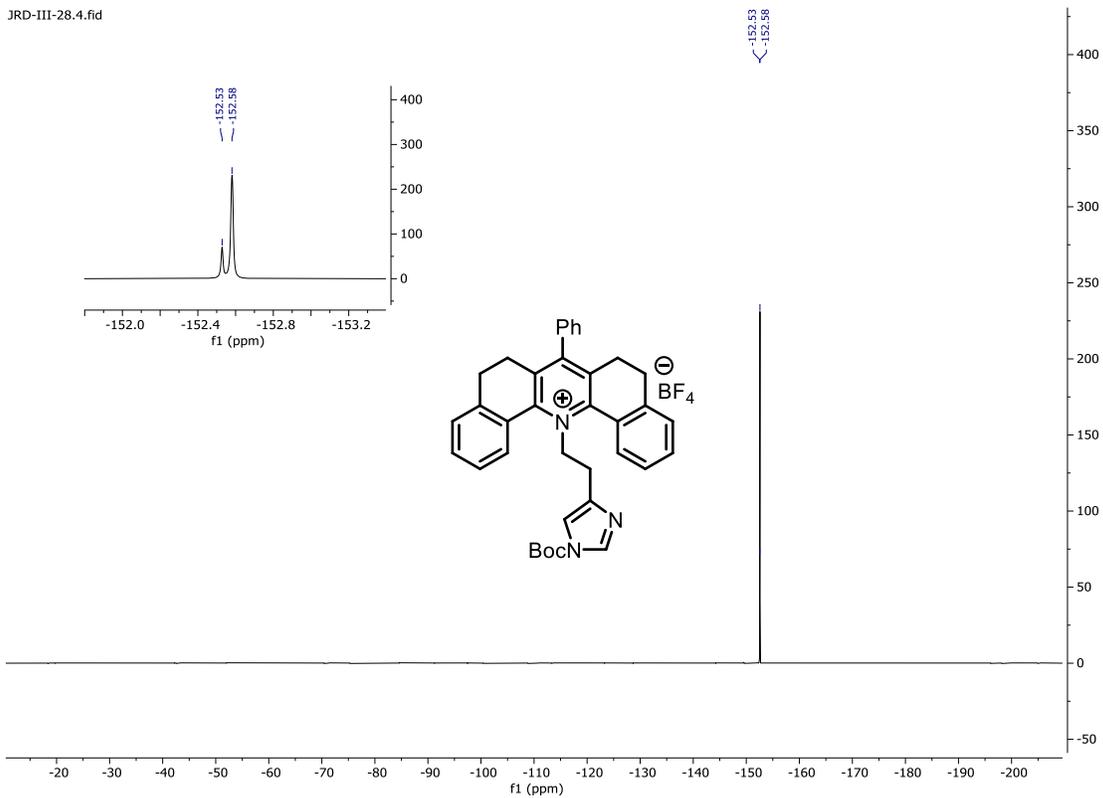
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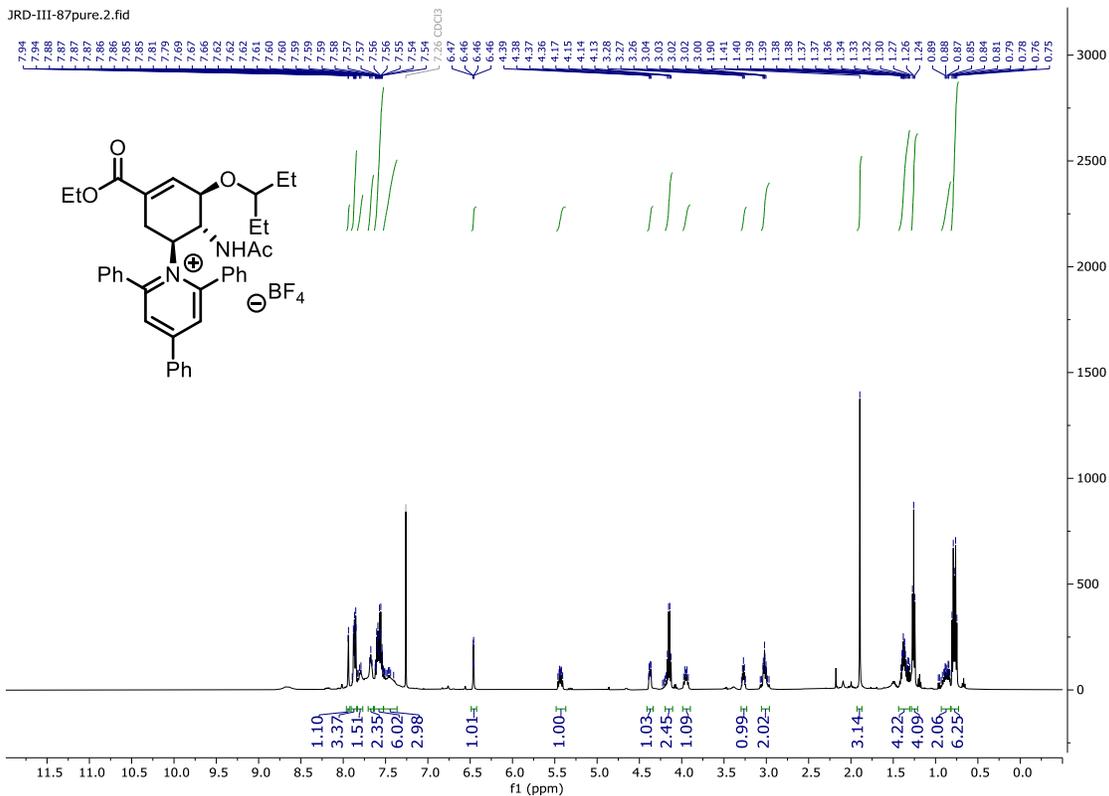
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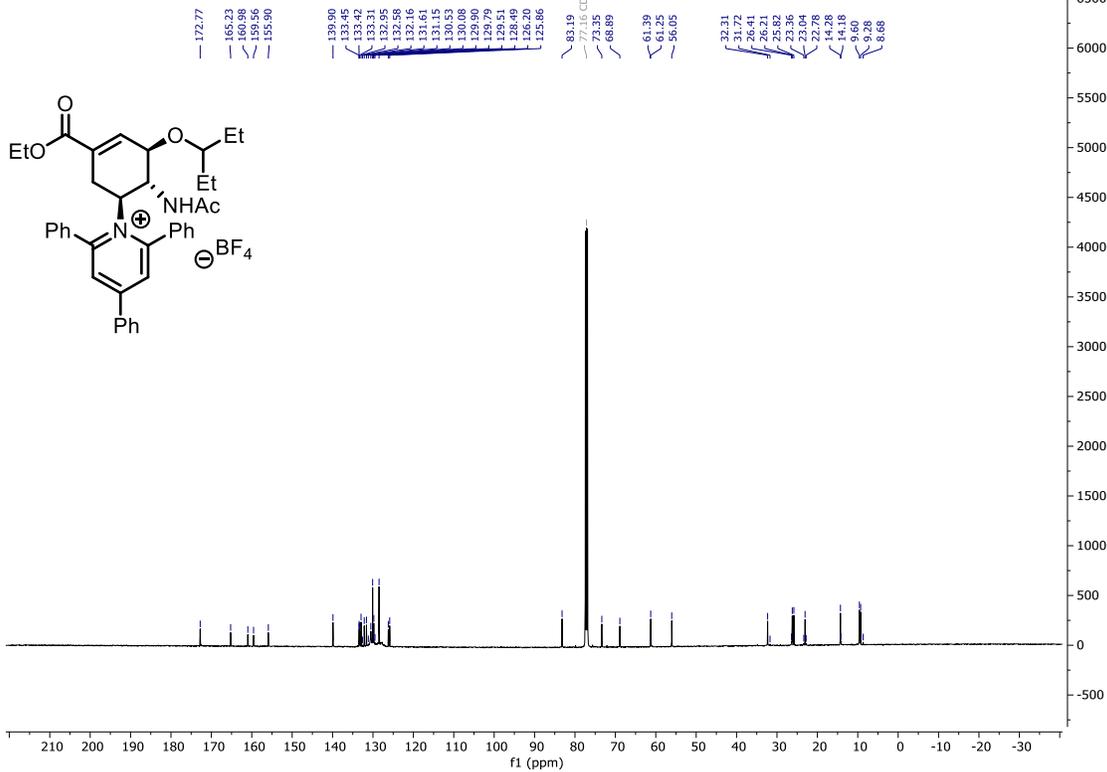
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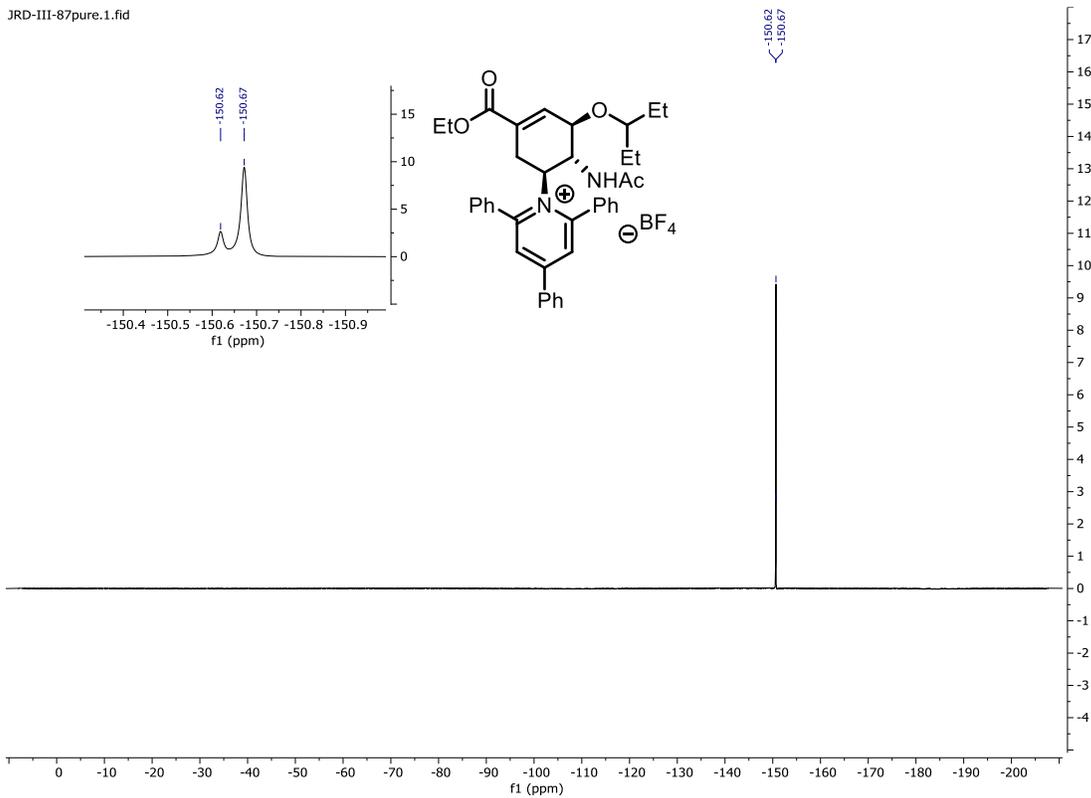
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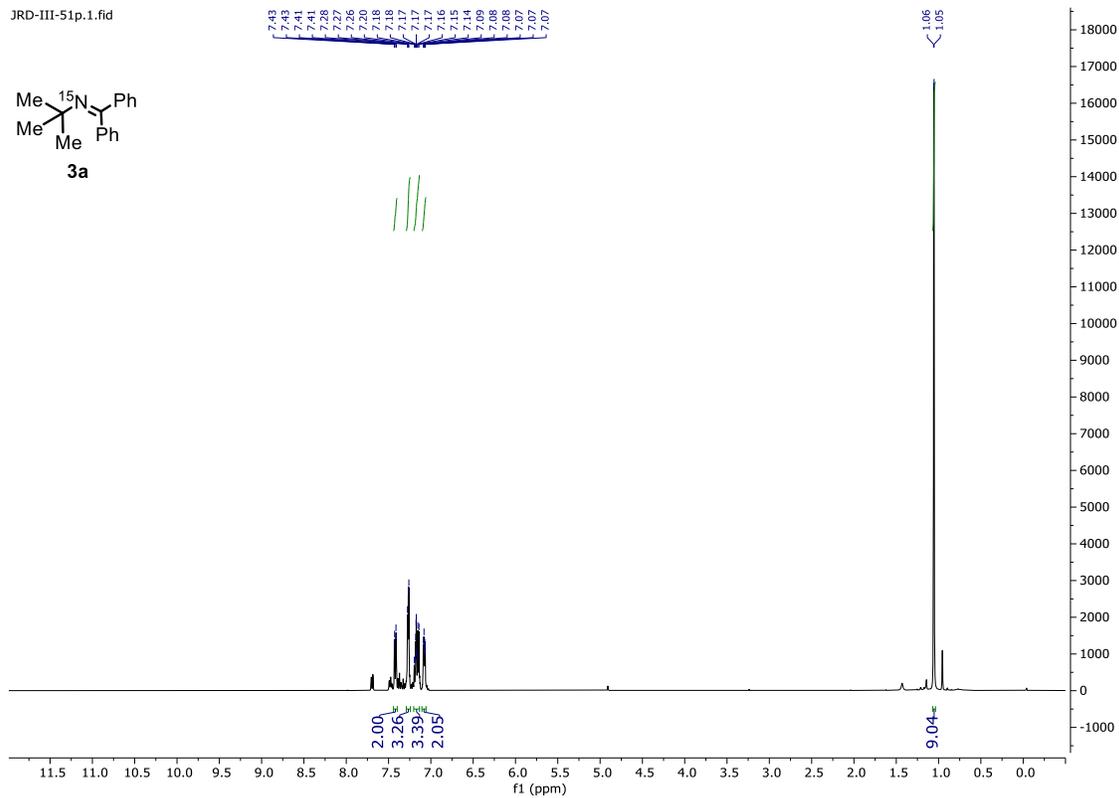
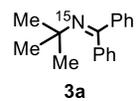
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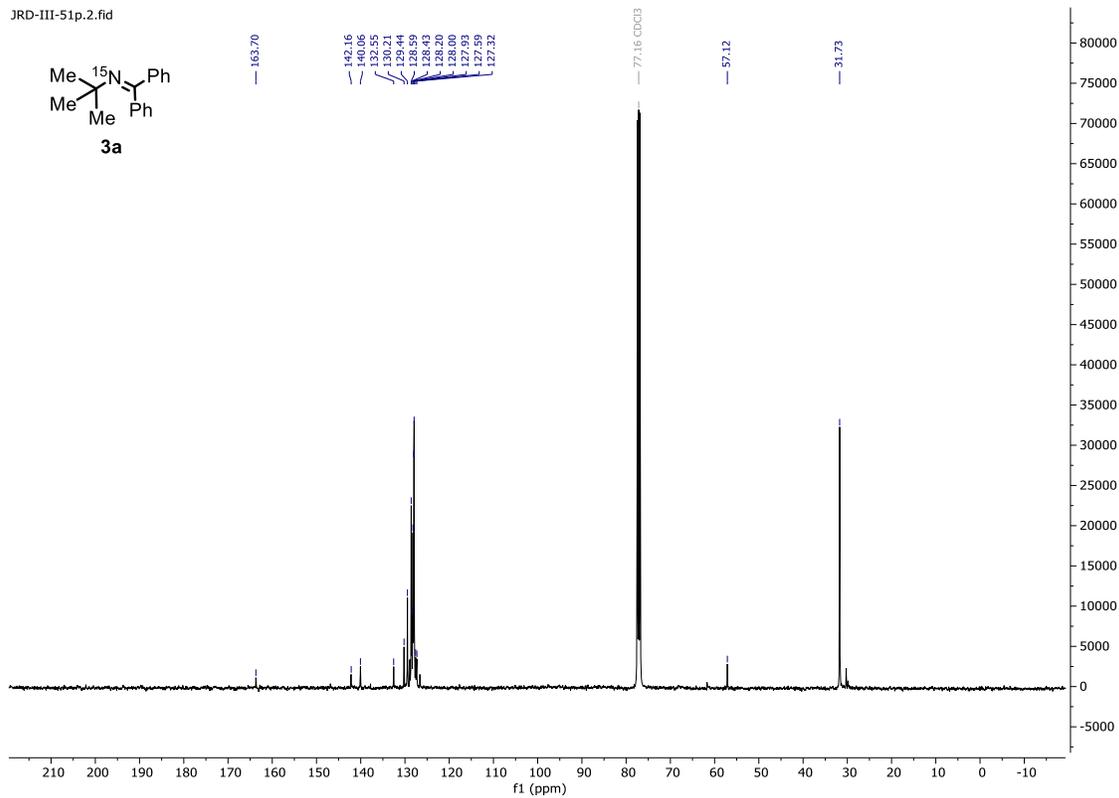
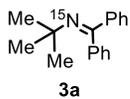
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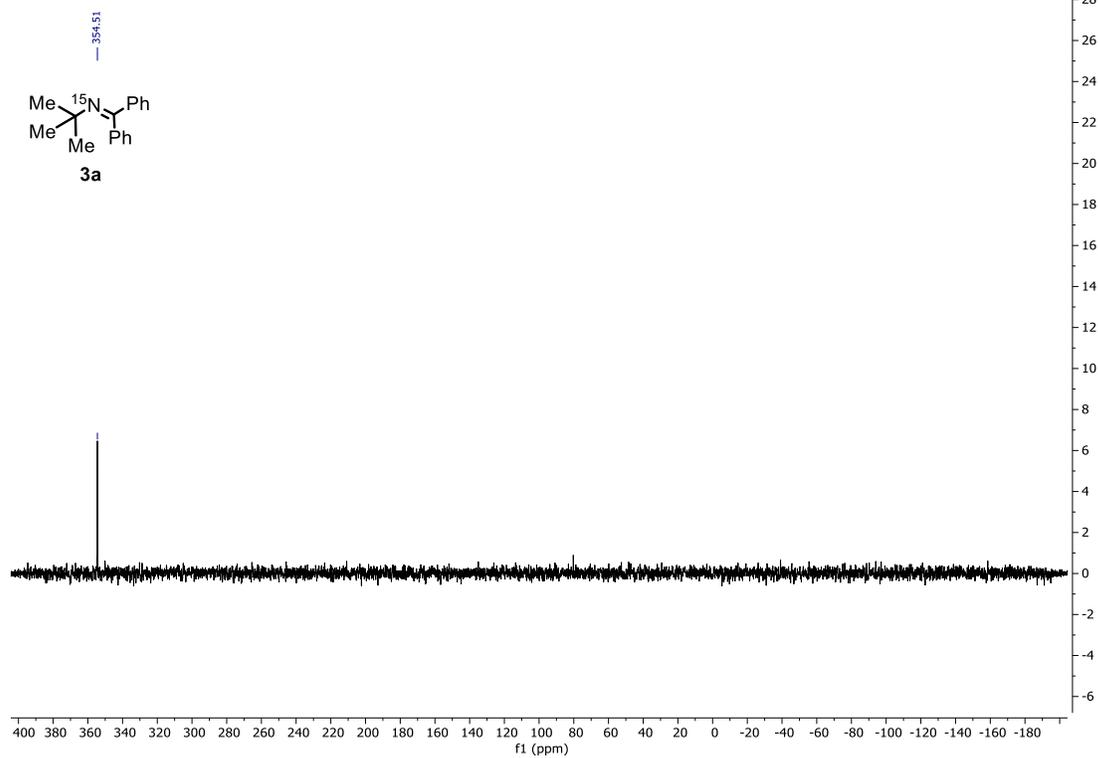
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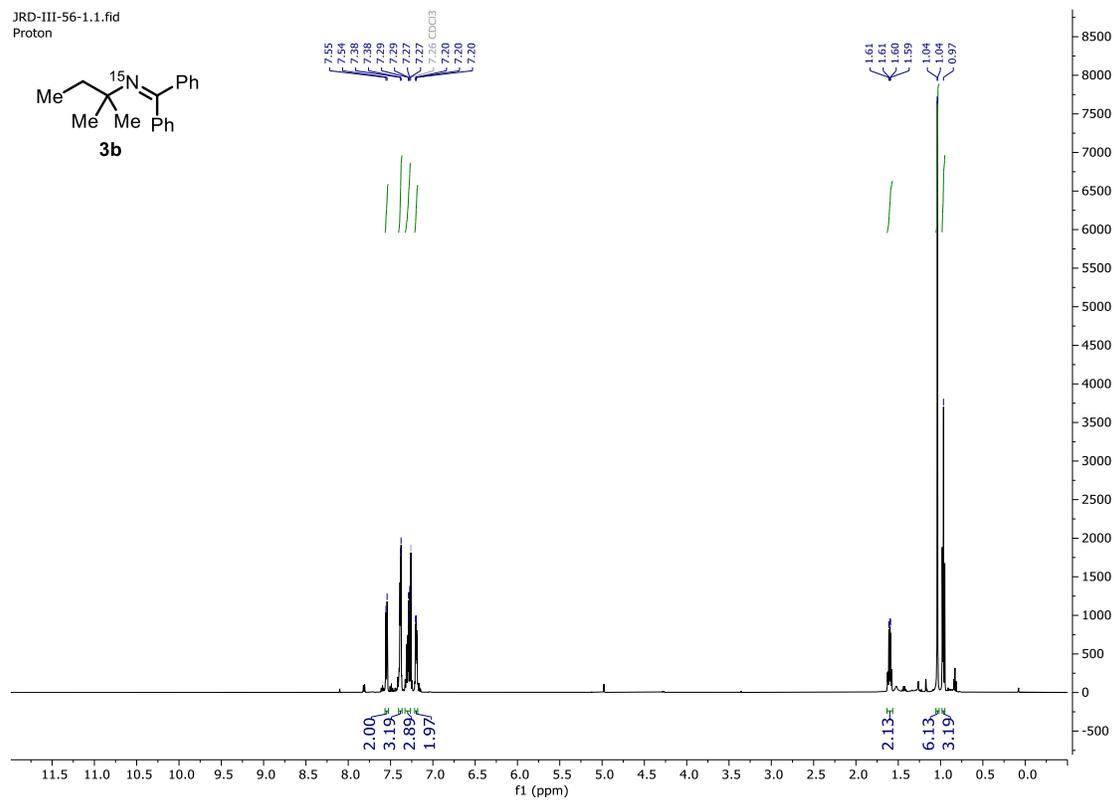
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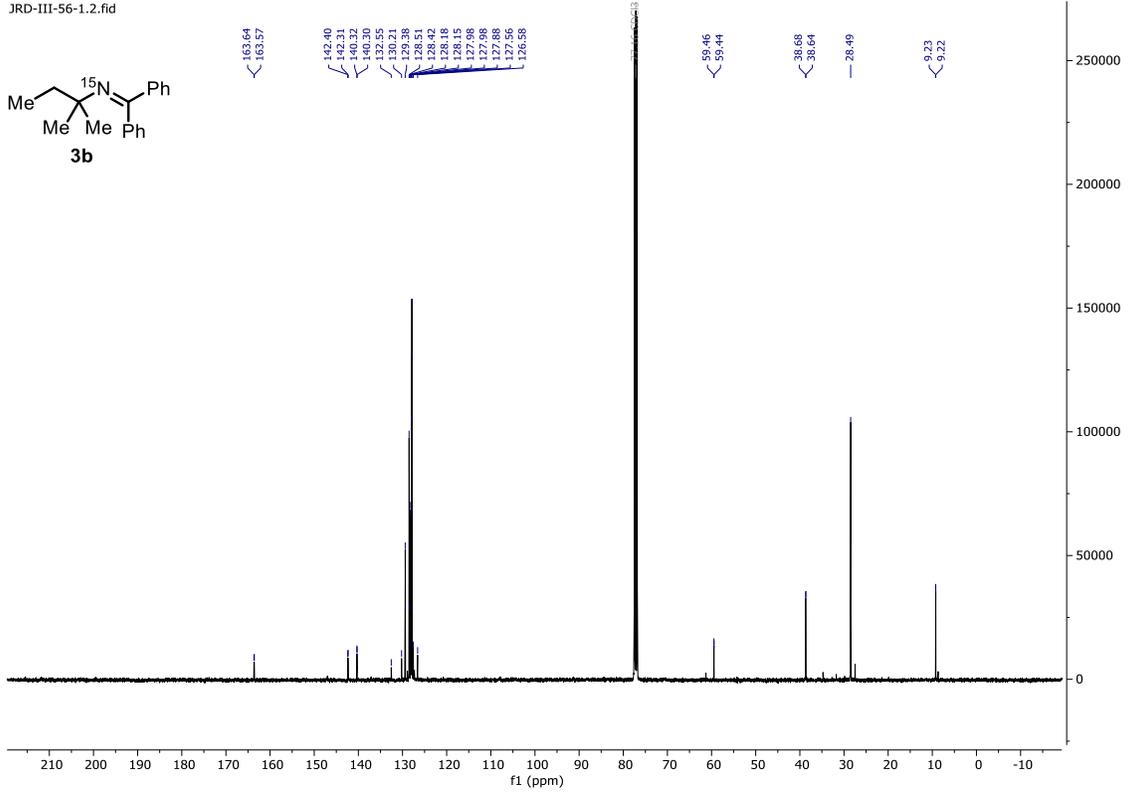
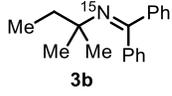
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Nitrogen 15



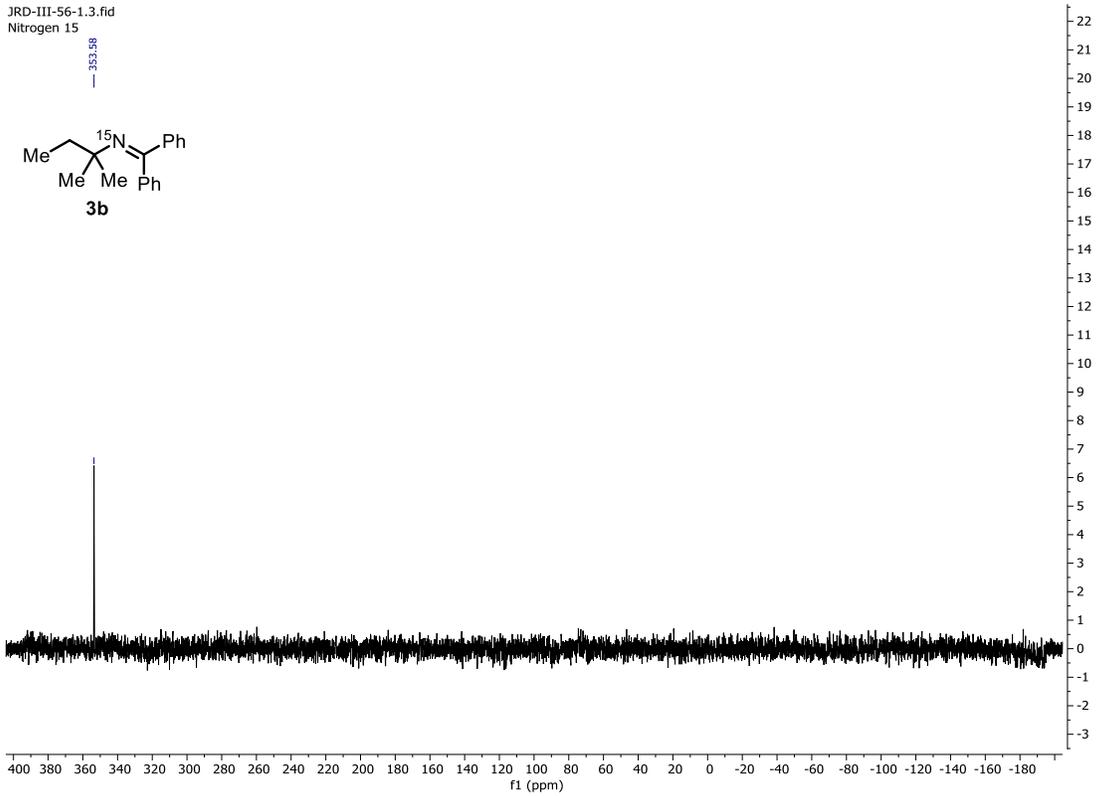
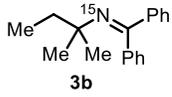
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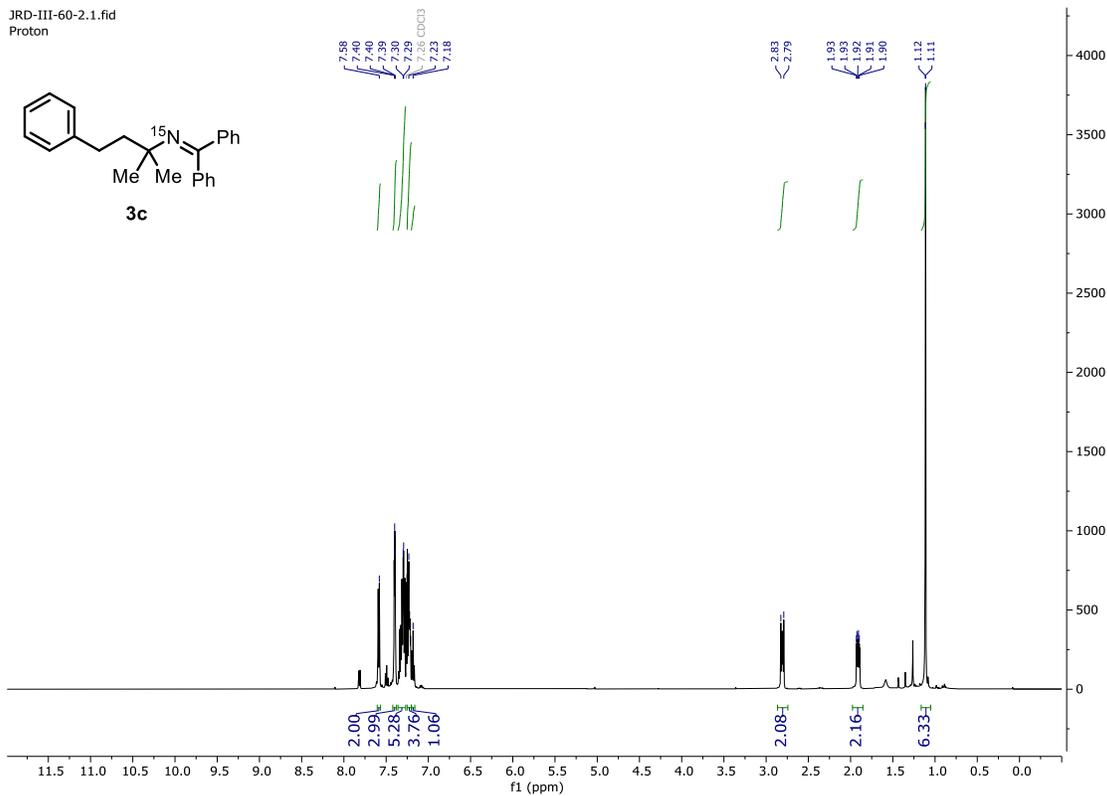
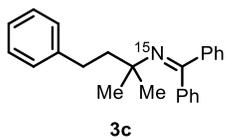
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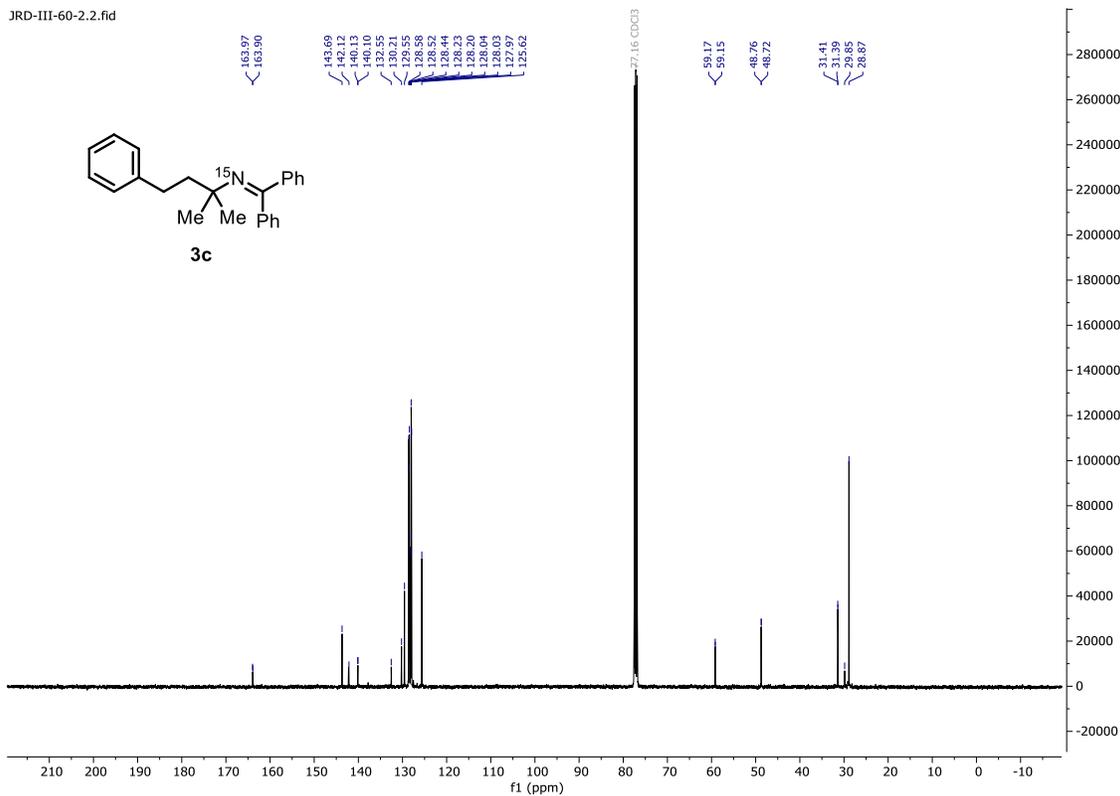
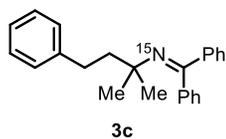
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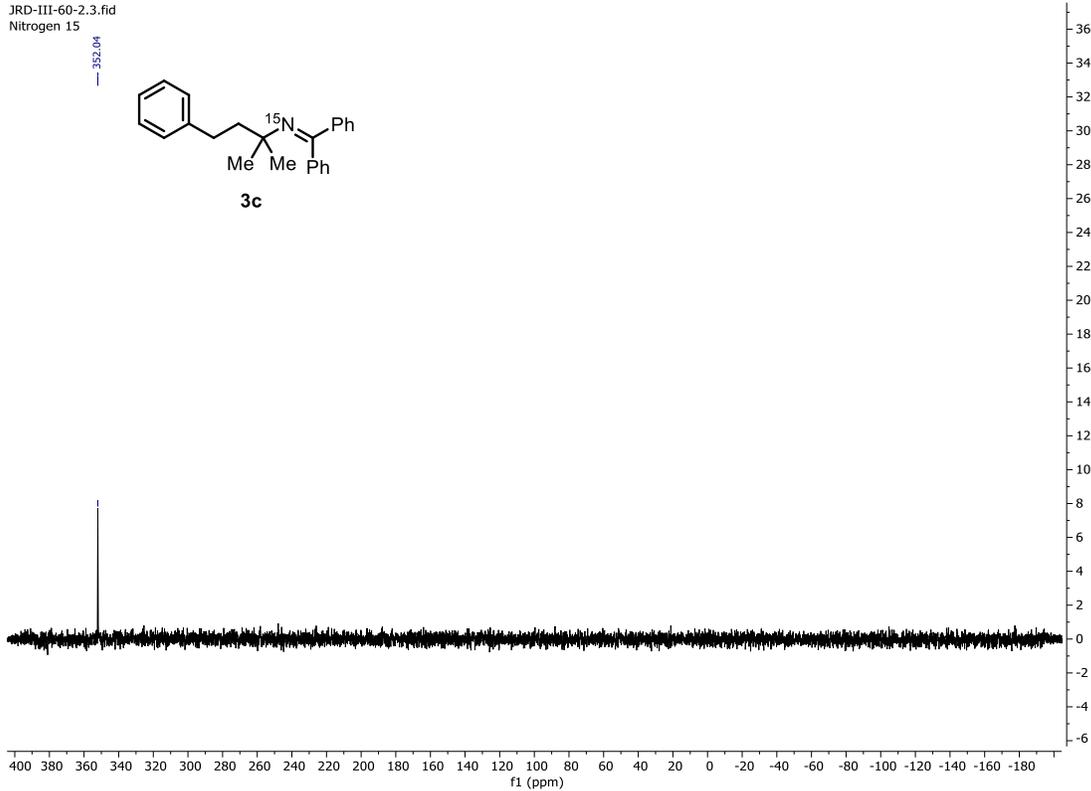
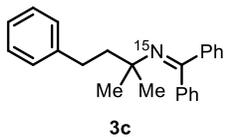
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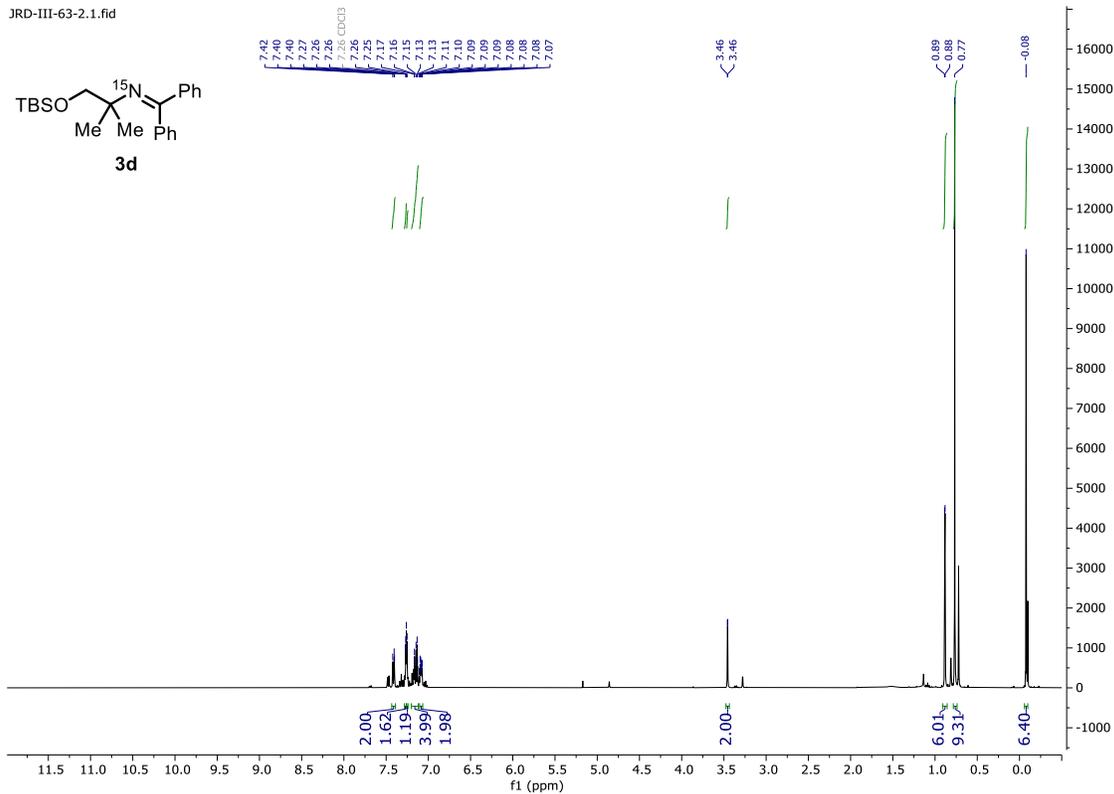
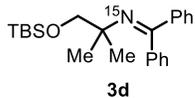
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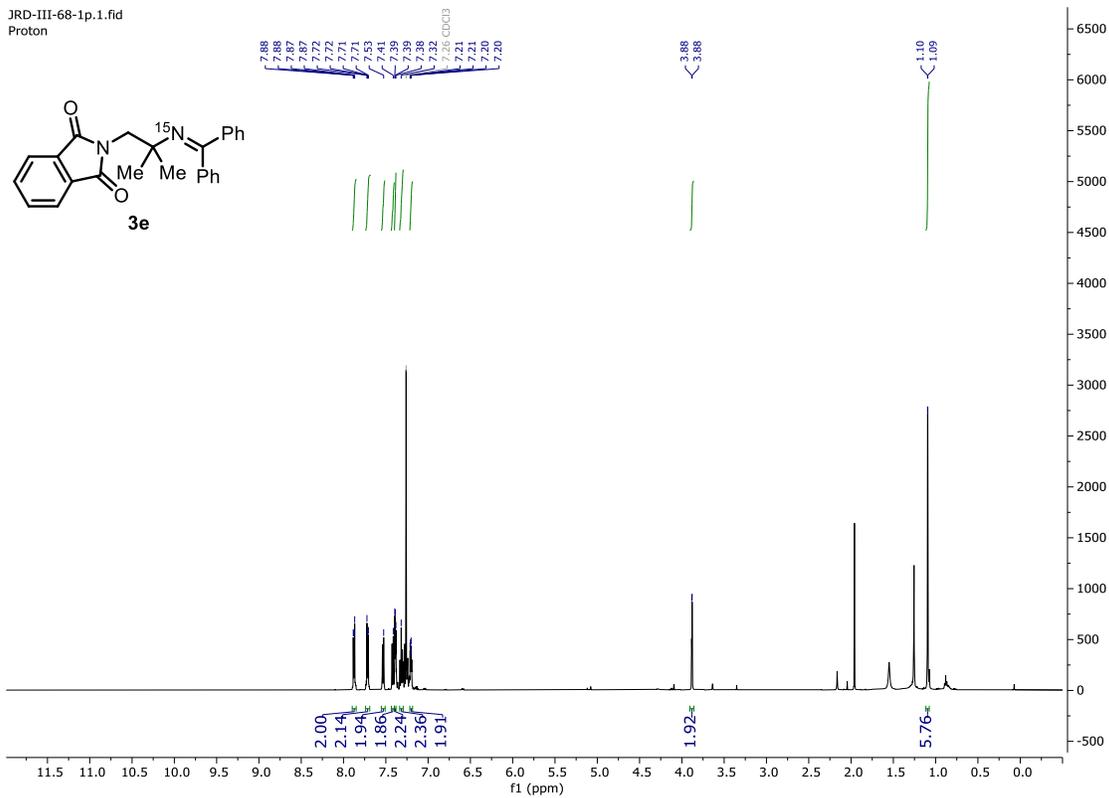
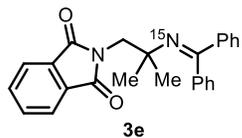
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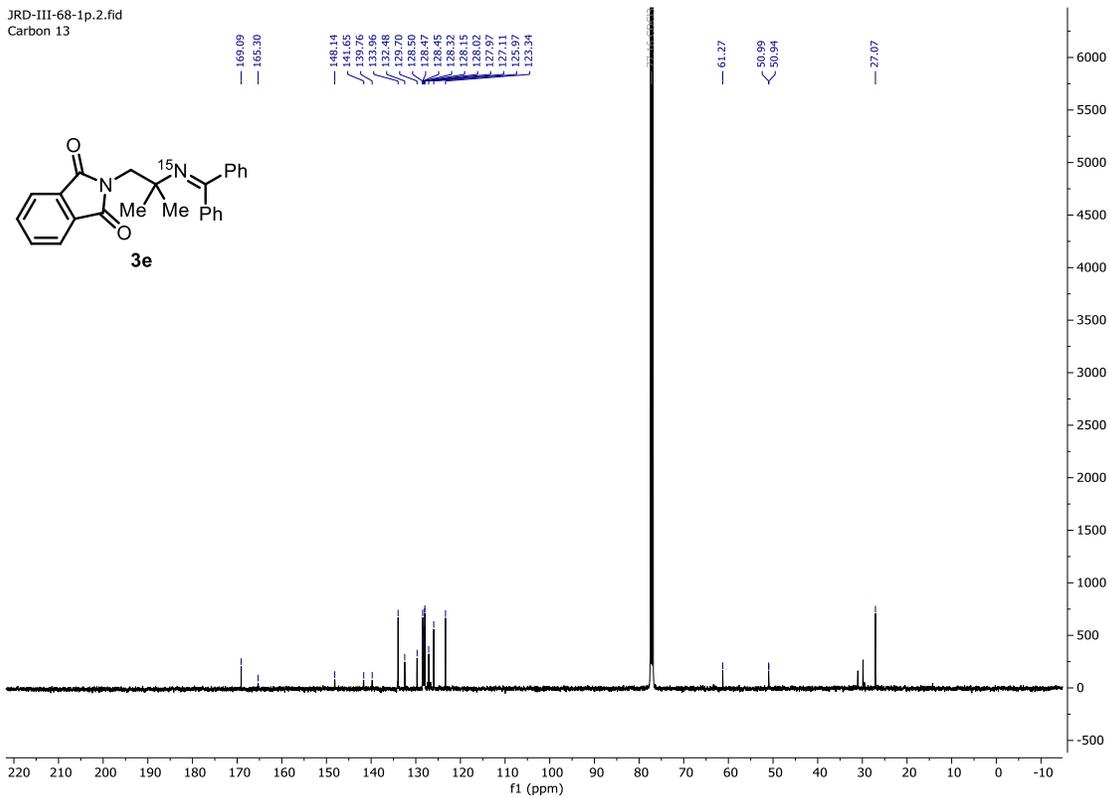
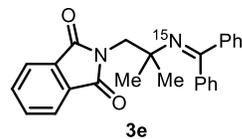
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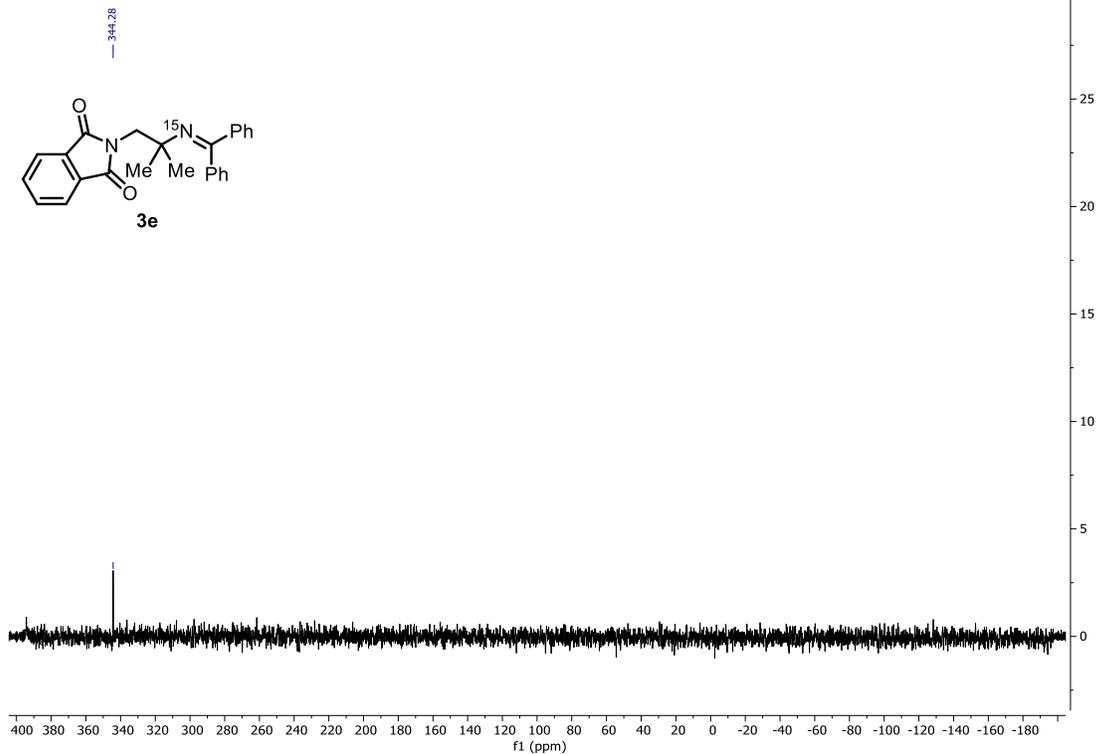
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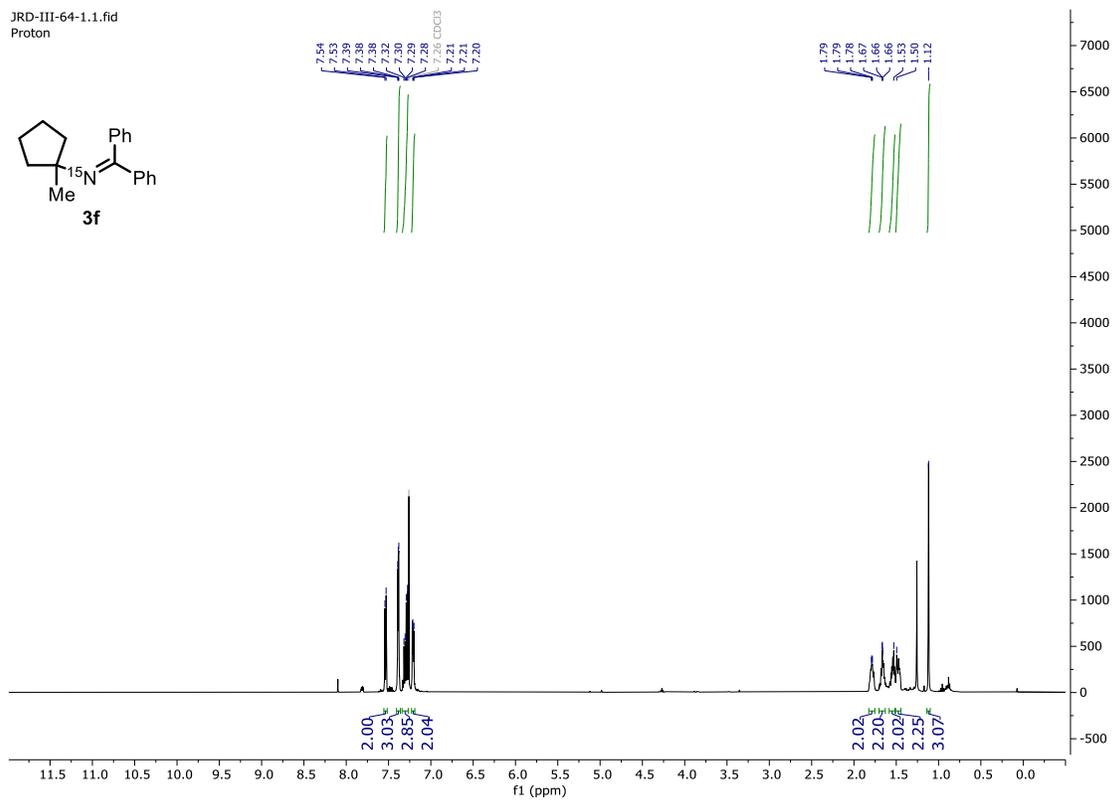
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Carbon 13



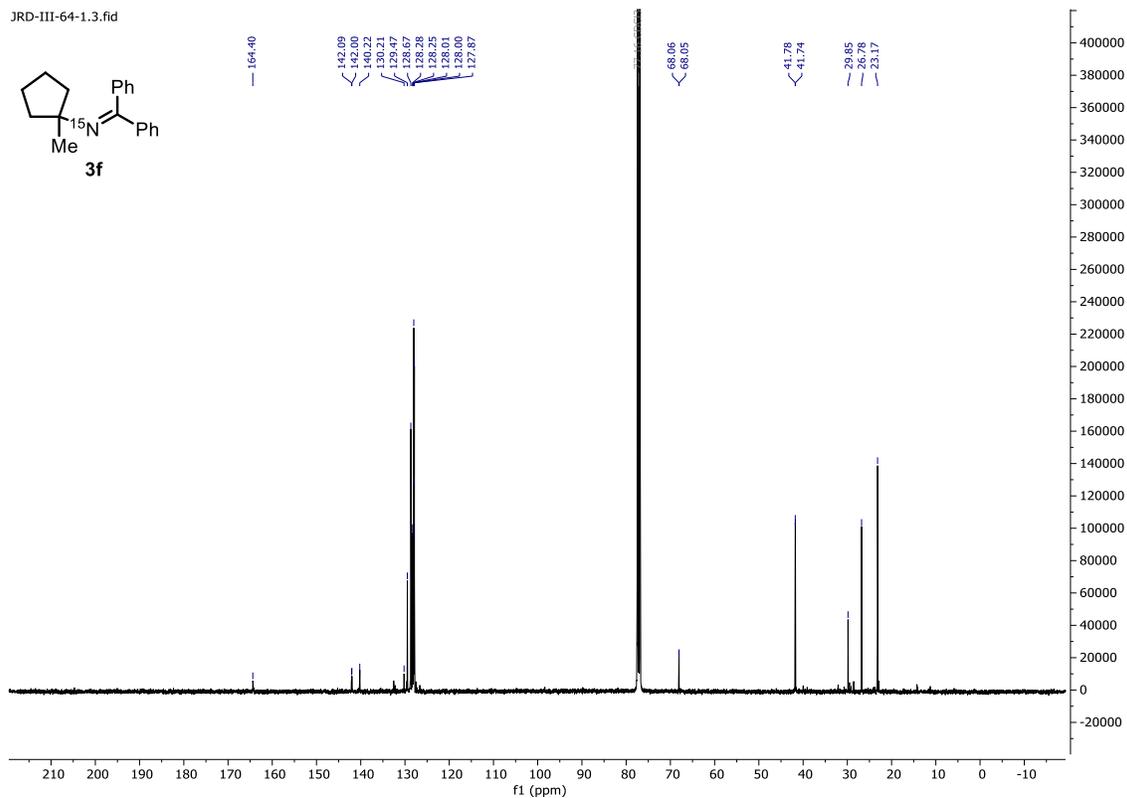
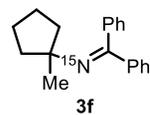
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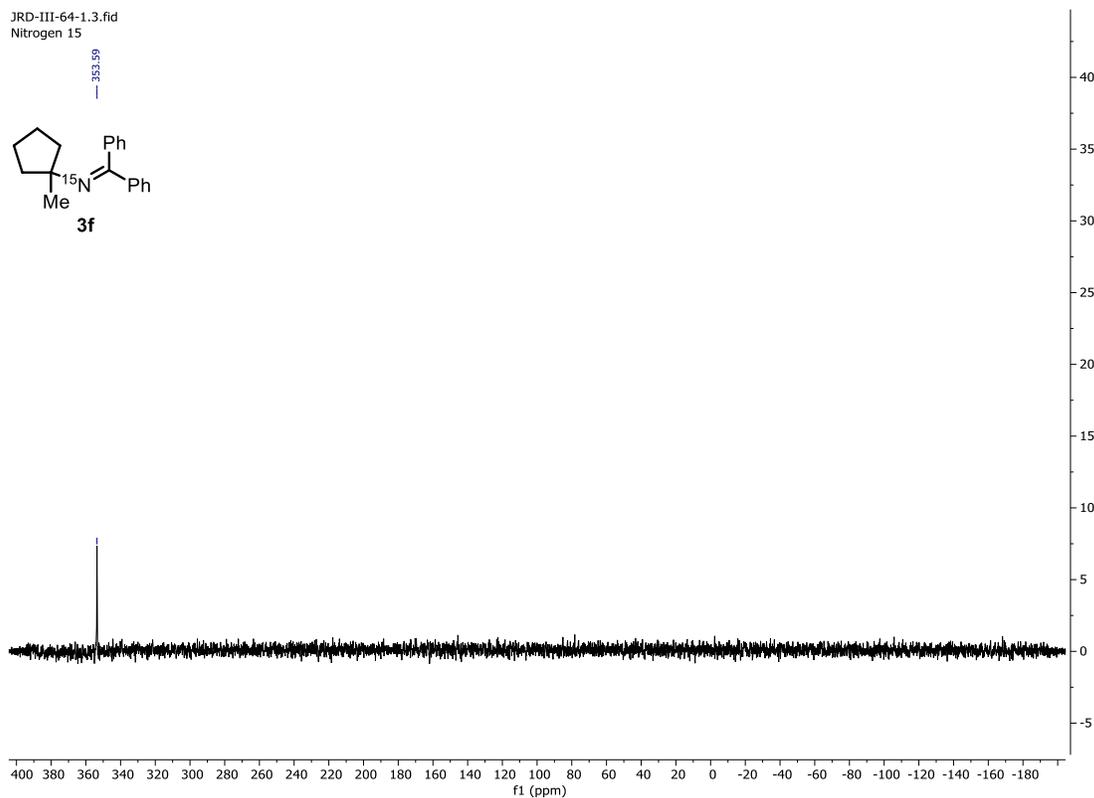
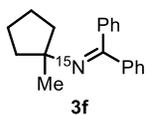
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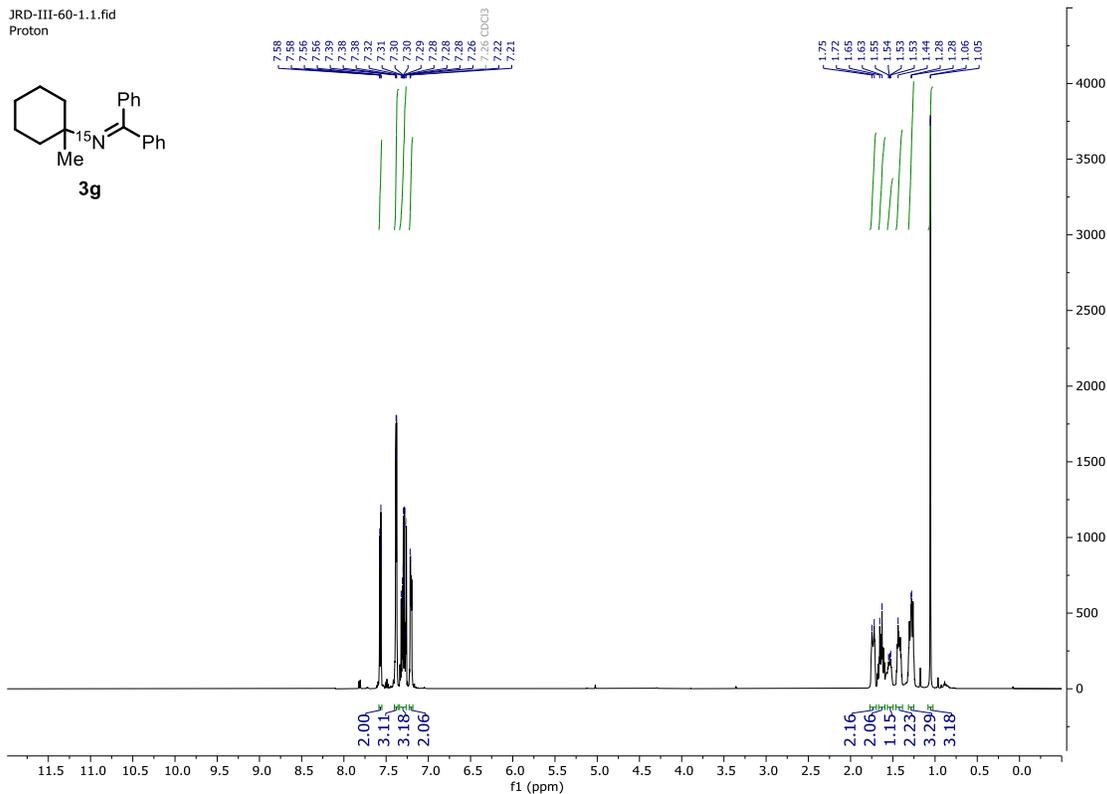
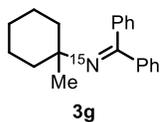
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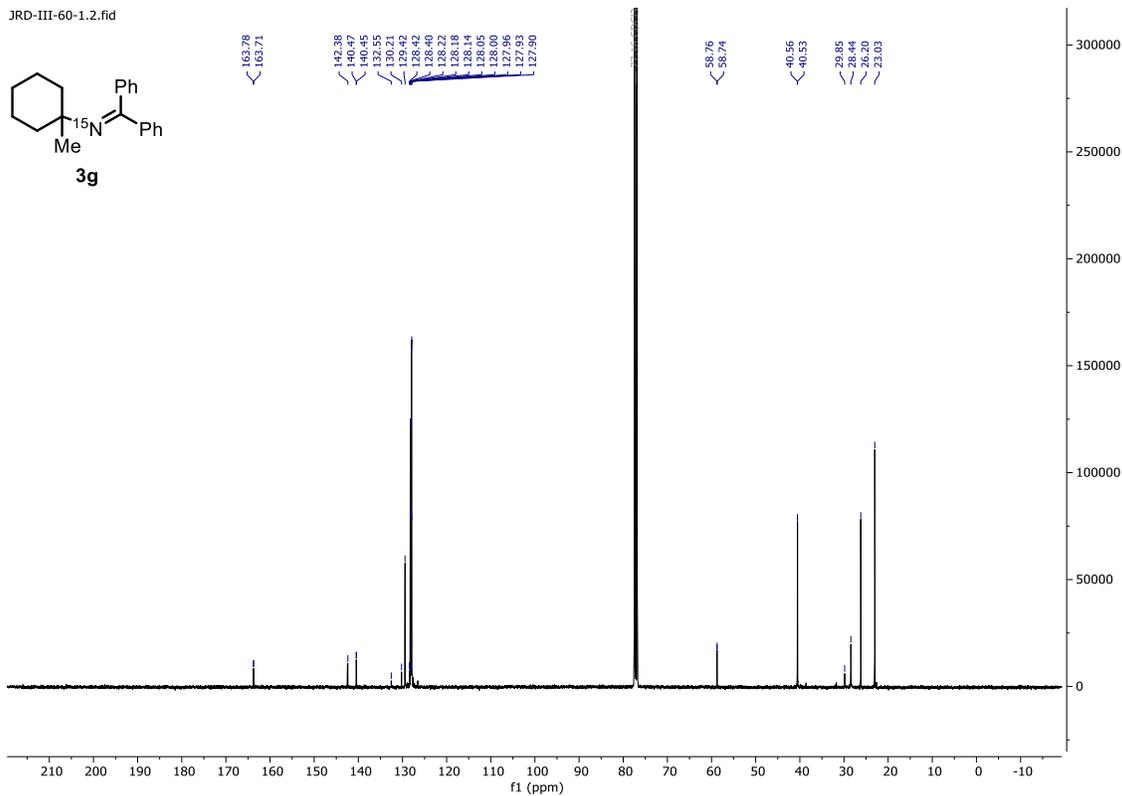
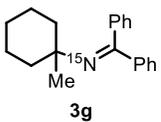
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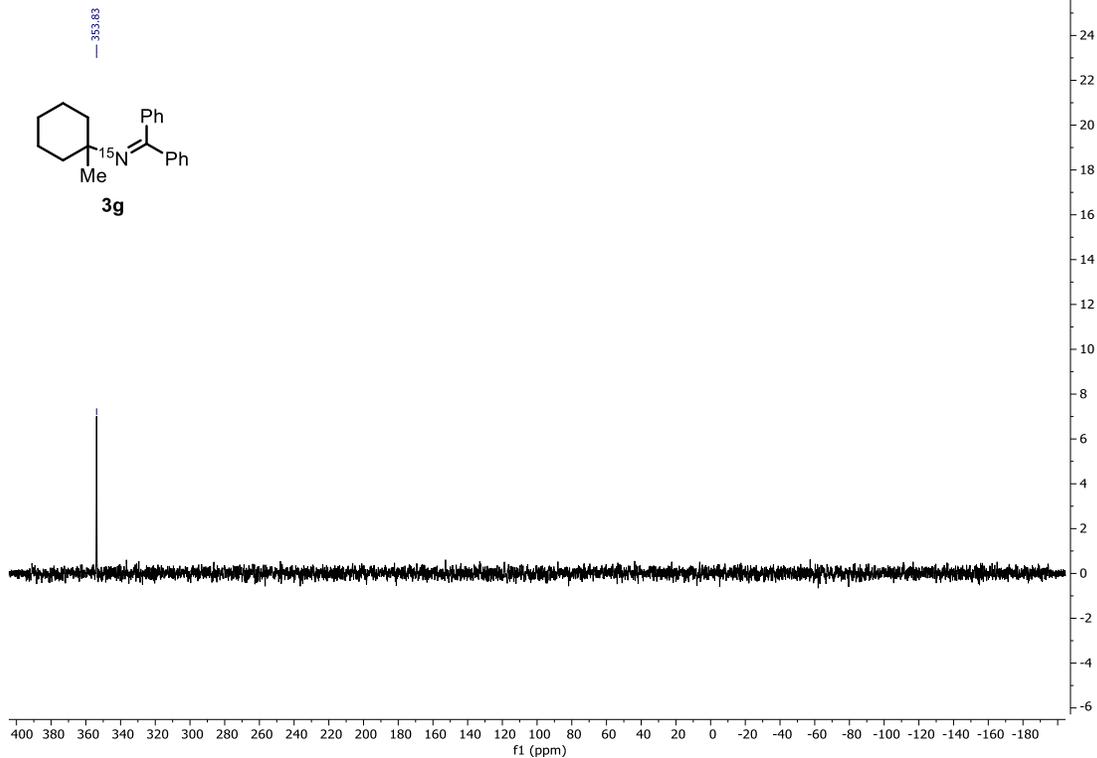
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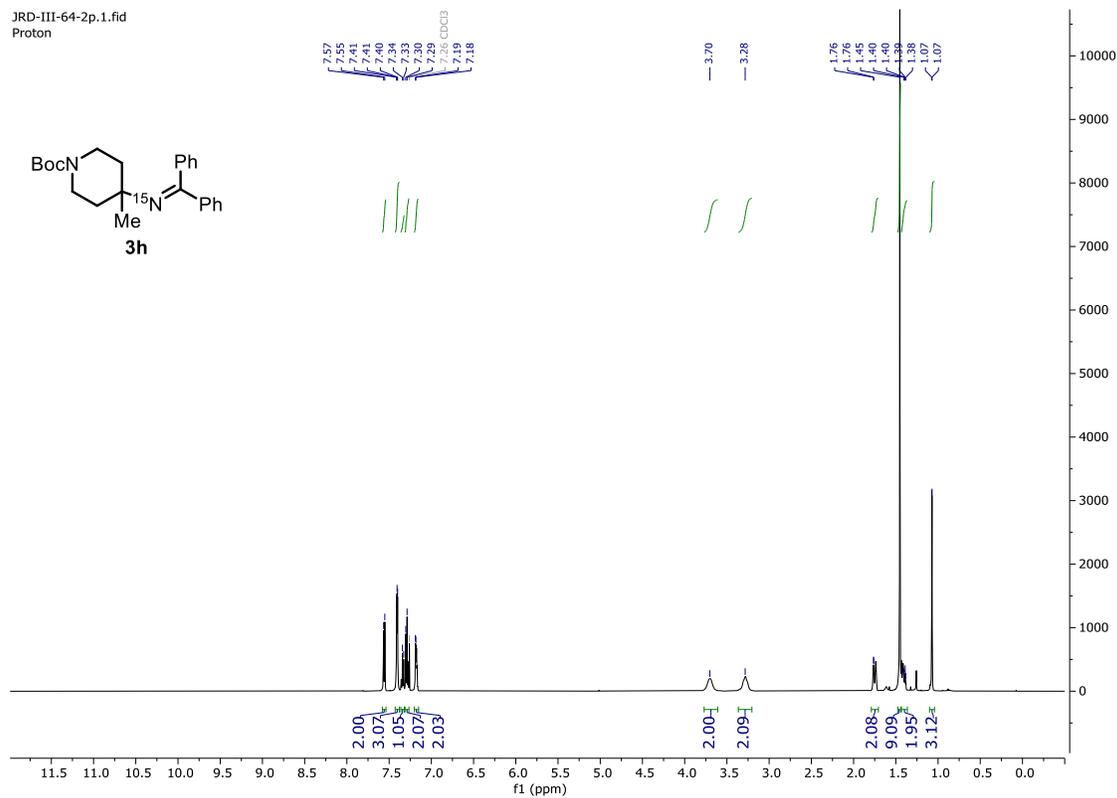
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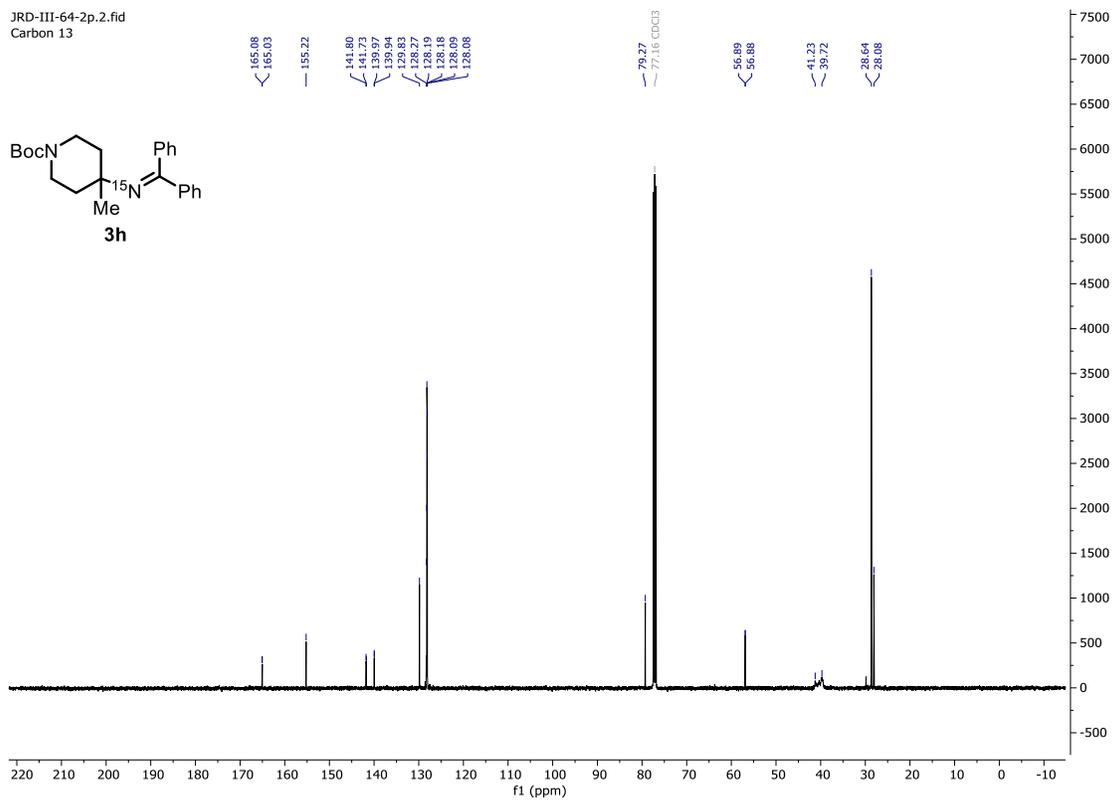
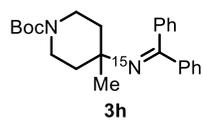
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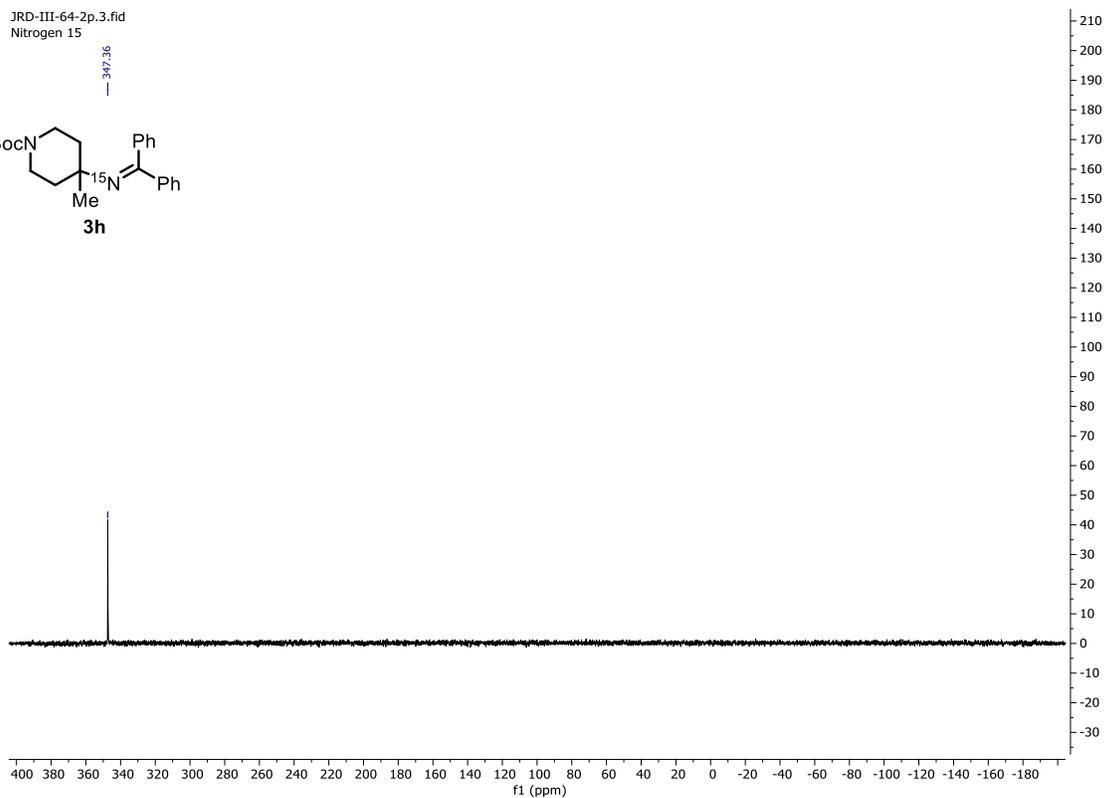
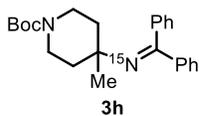
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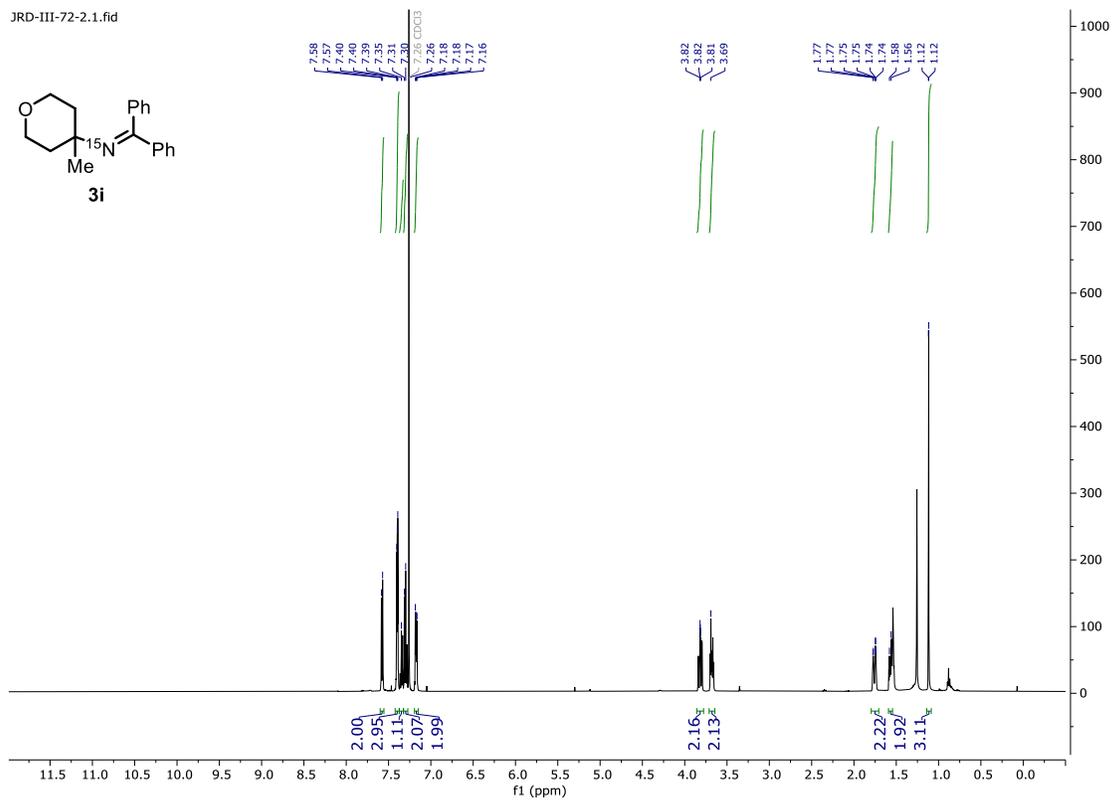
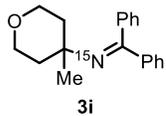
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Carbon 13



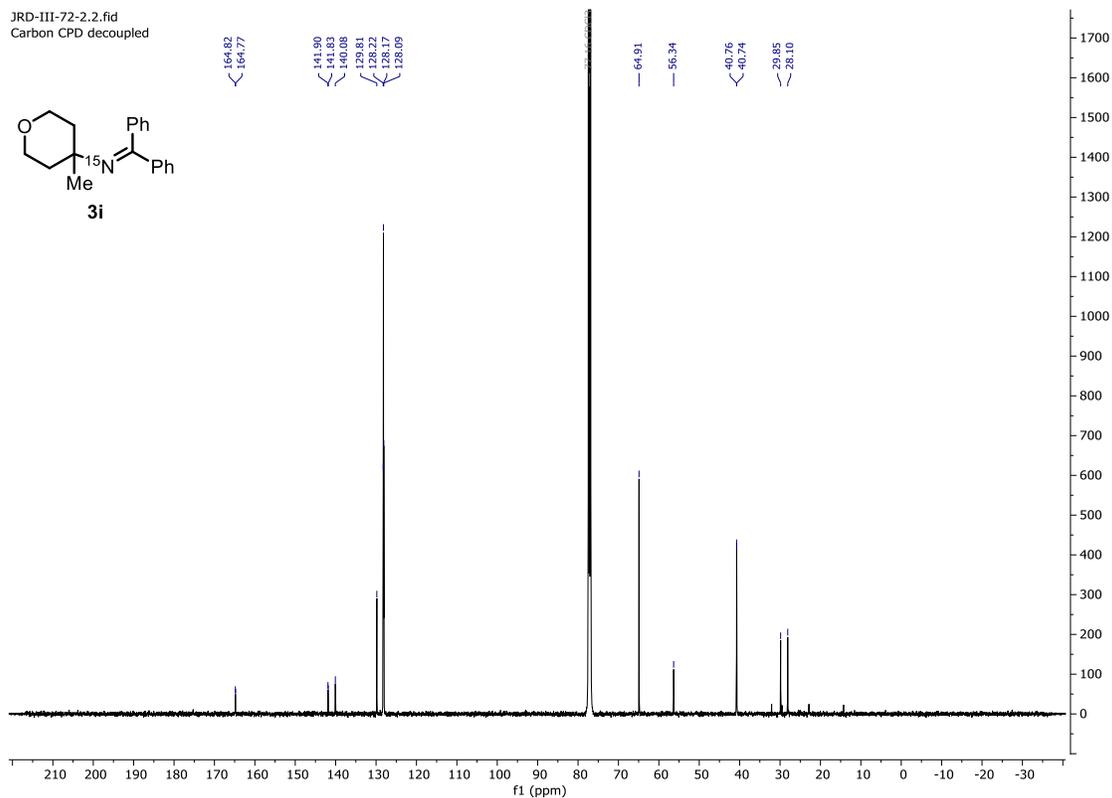
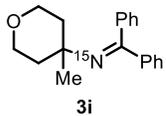
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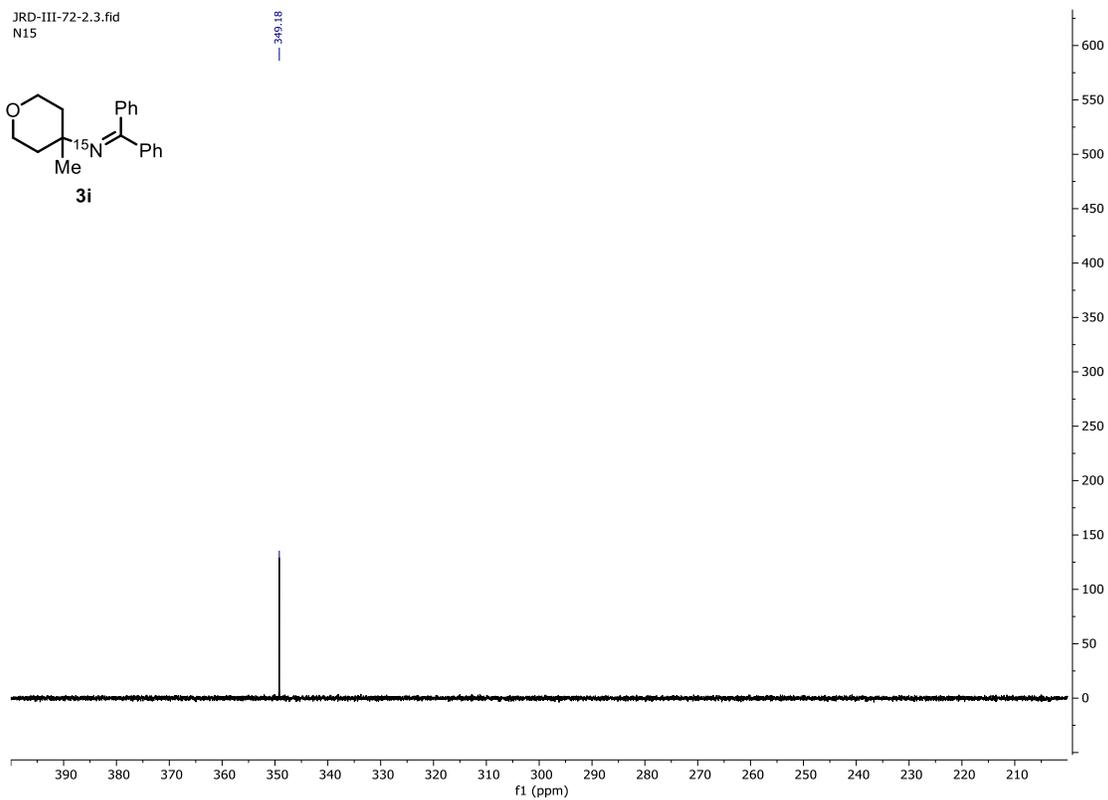
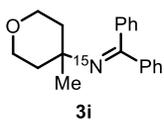
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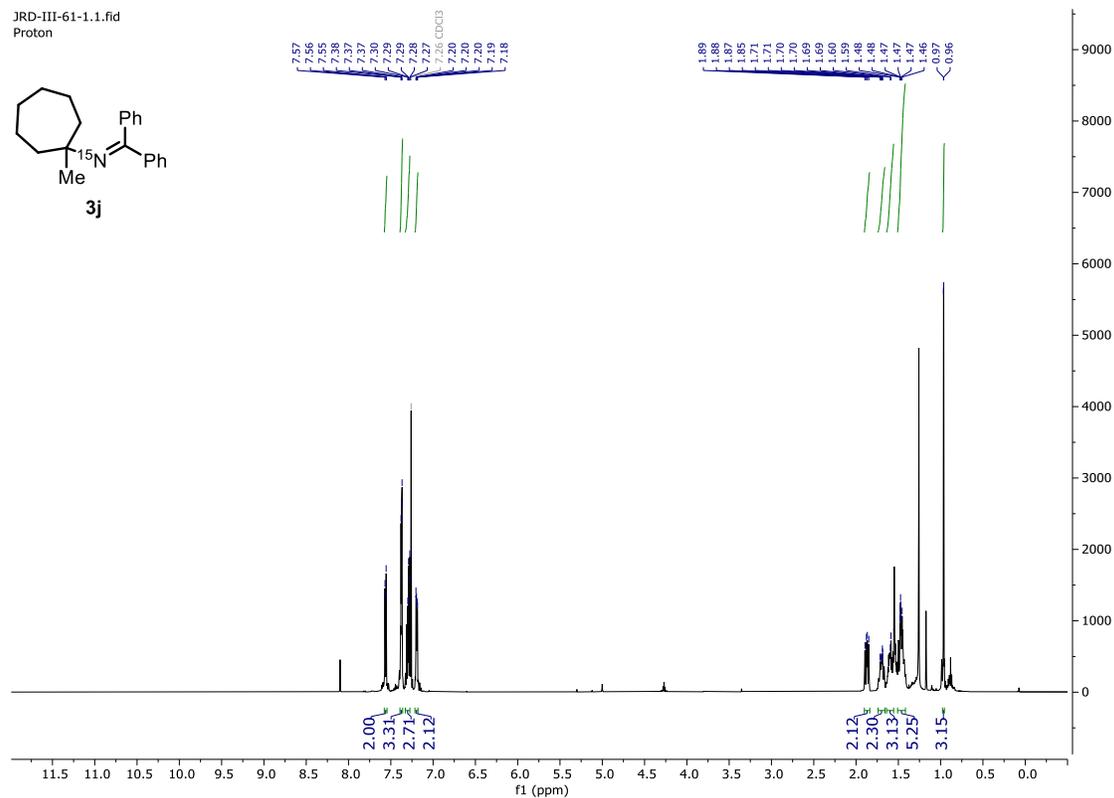
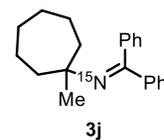
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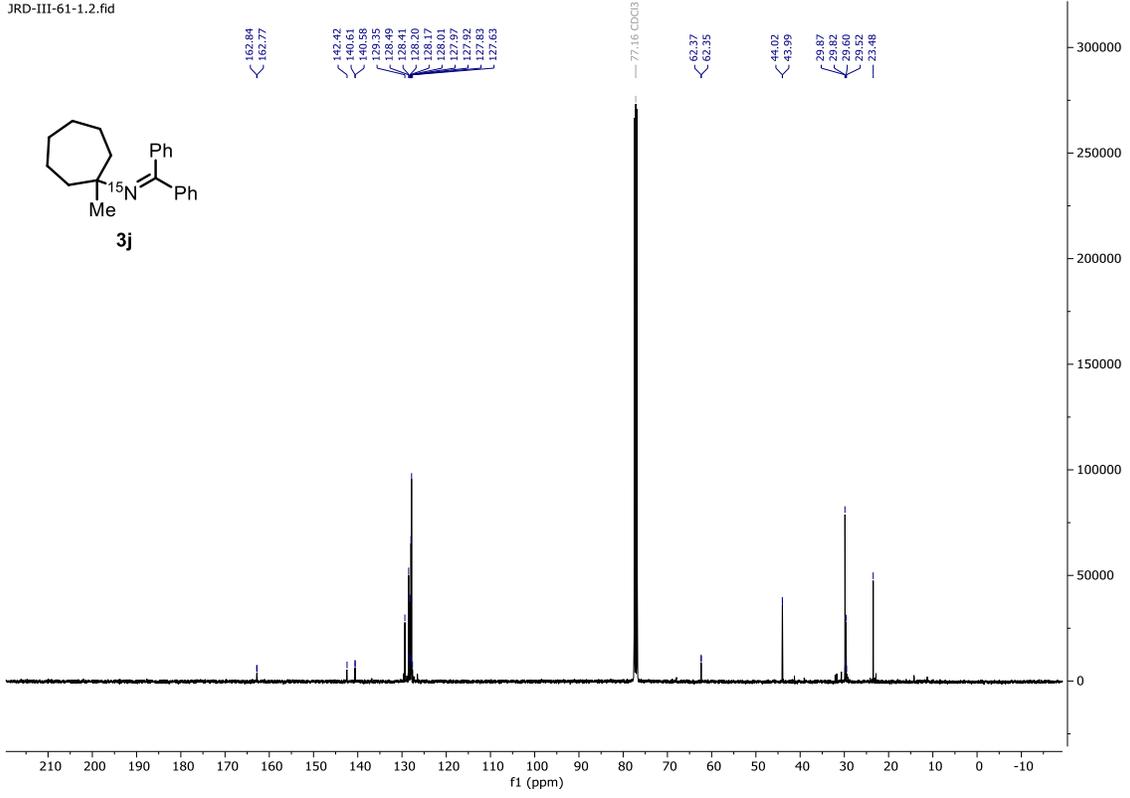
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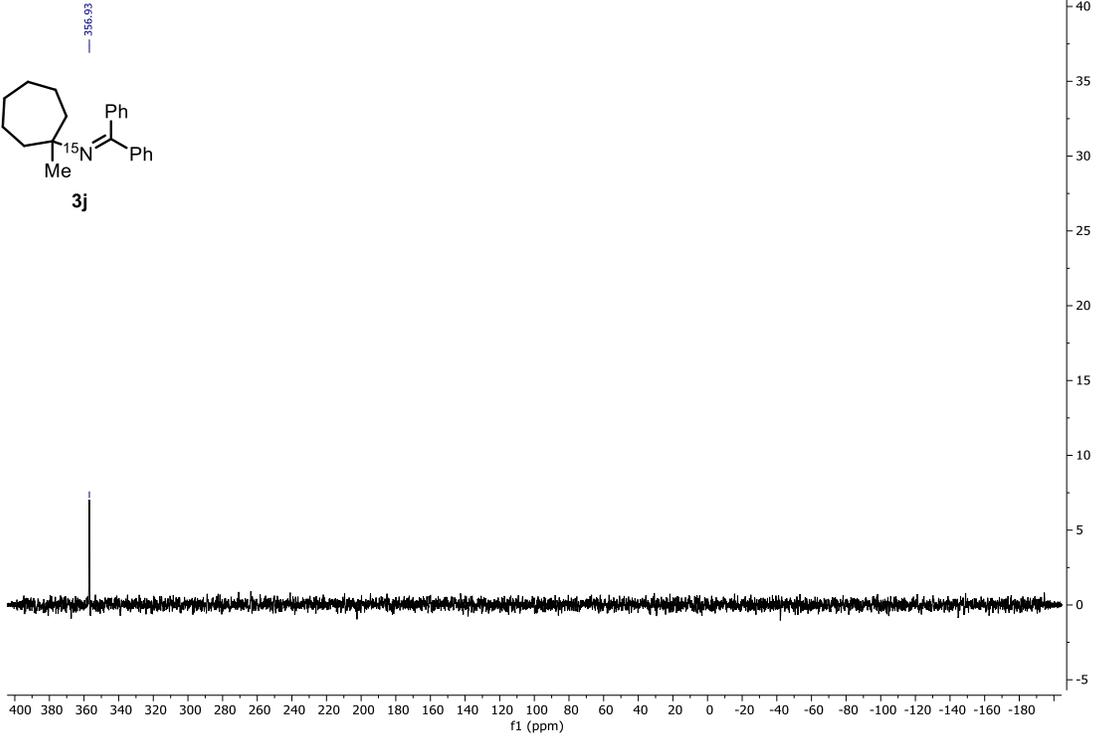
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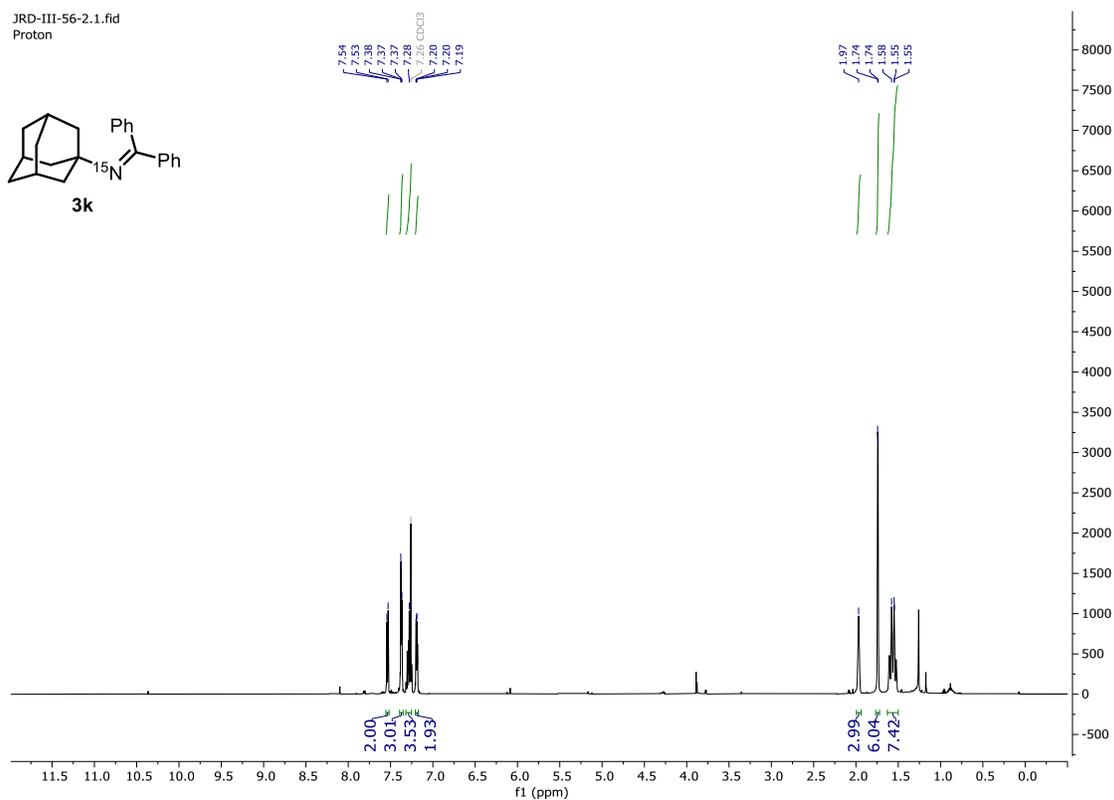
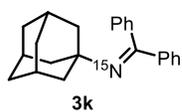
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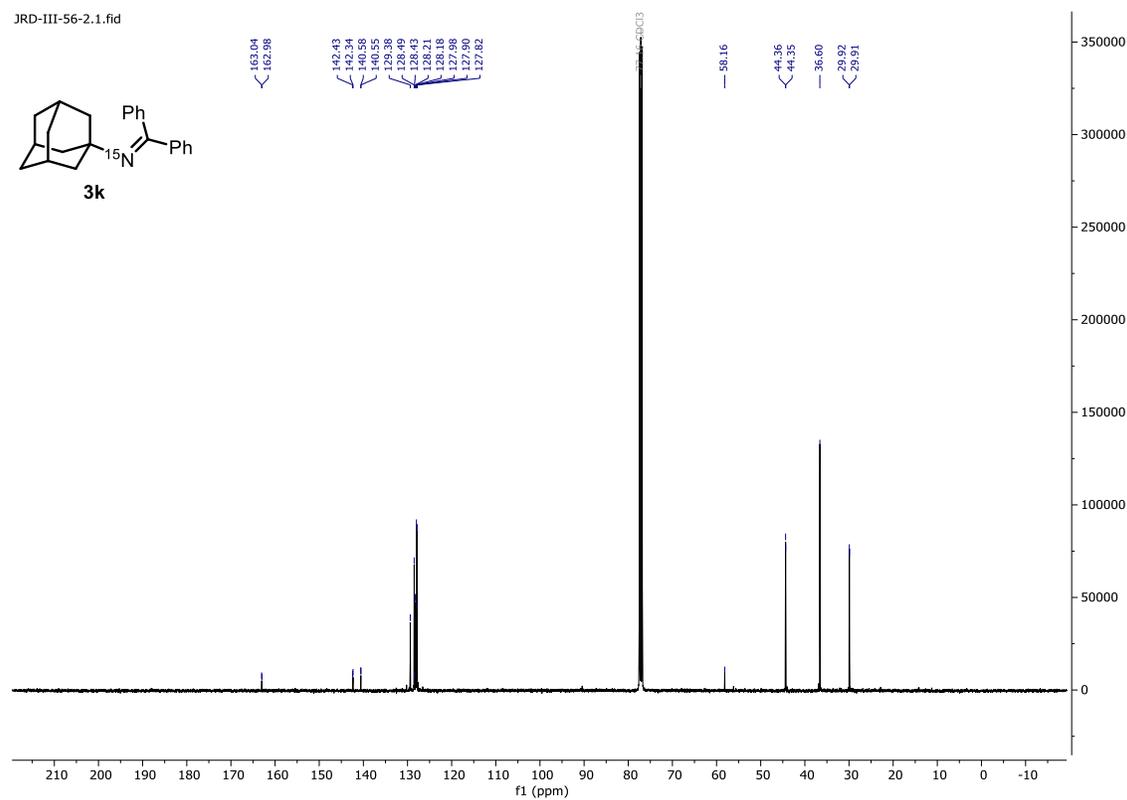
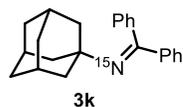
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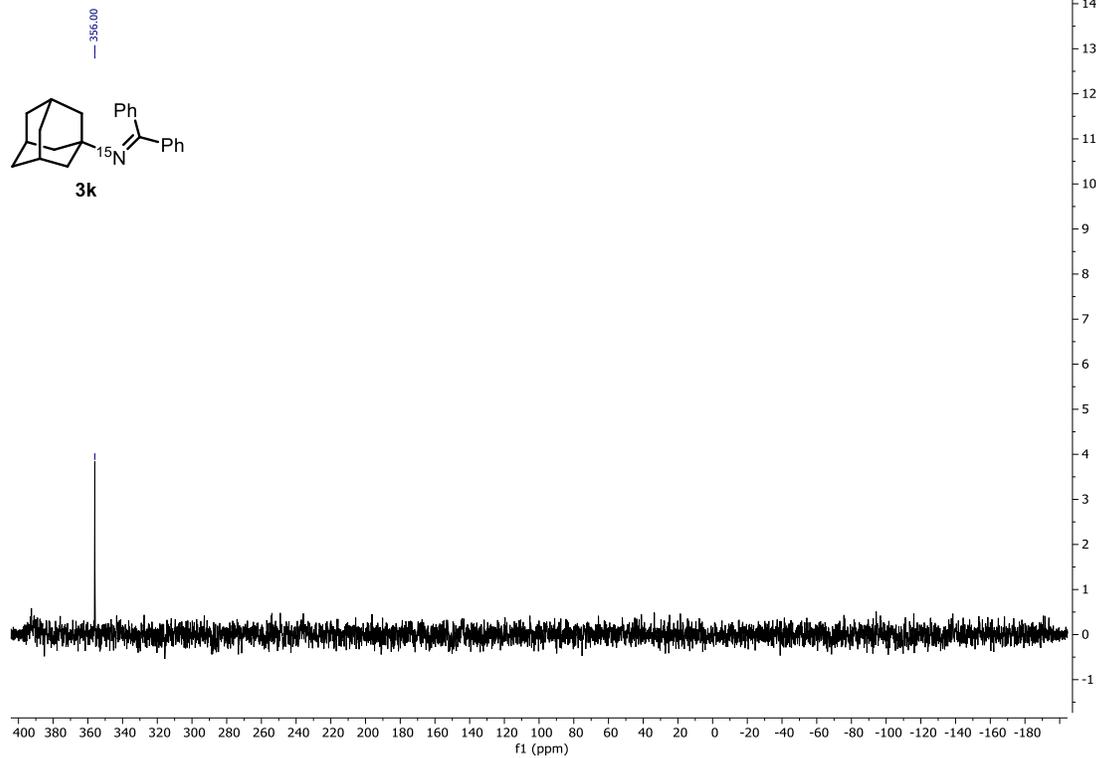
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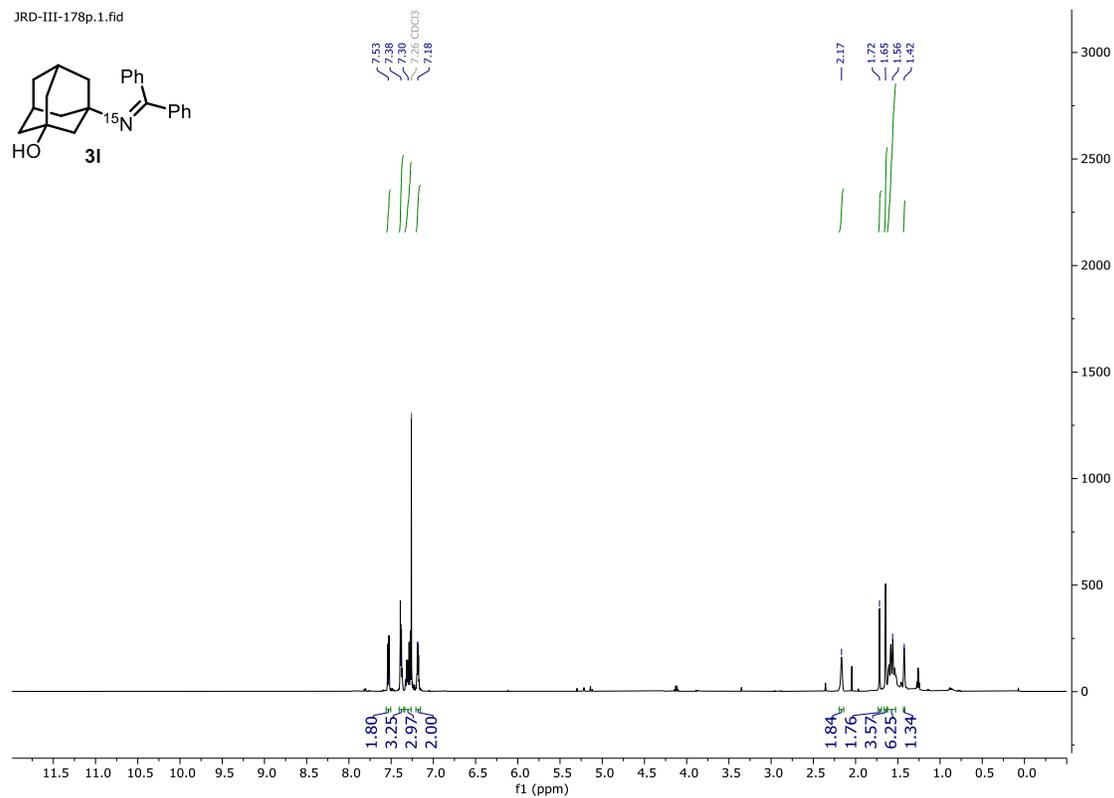
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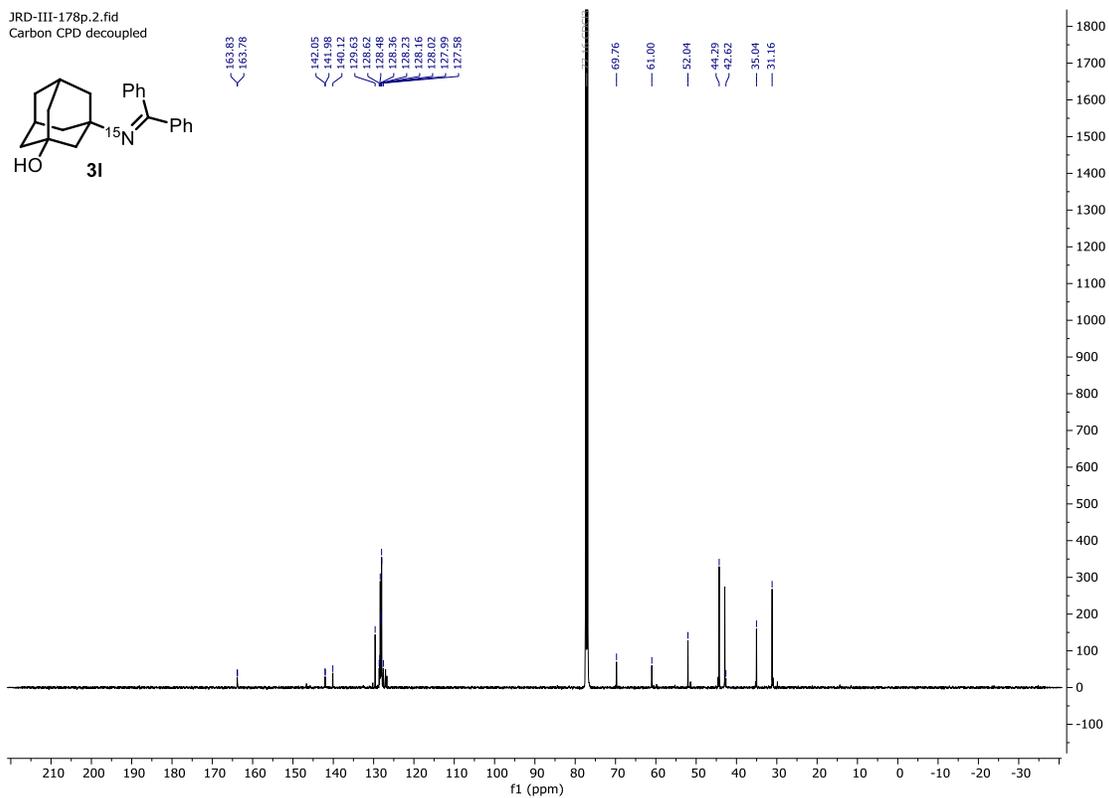
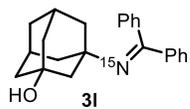
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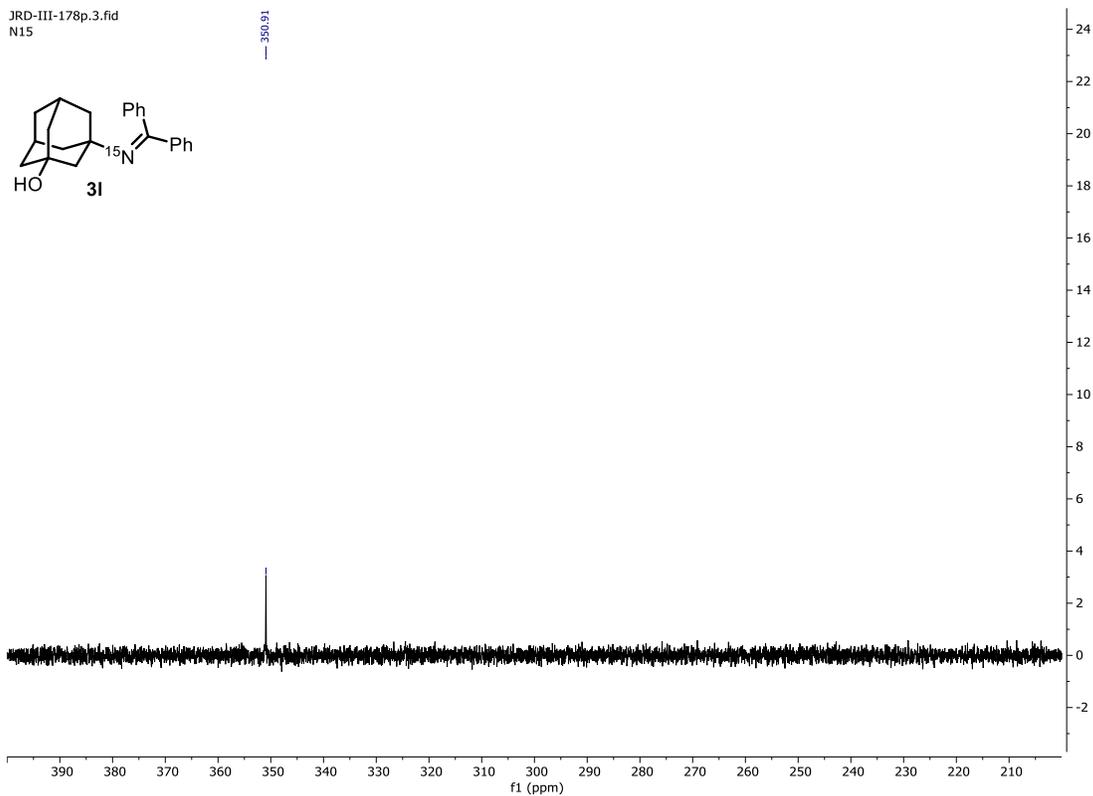
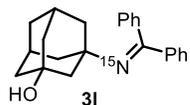
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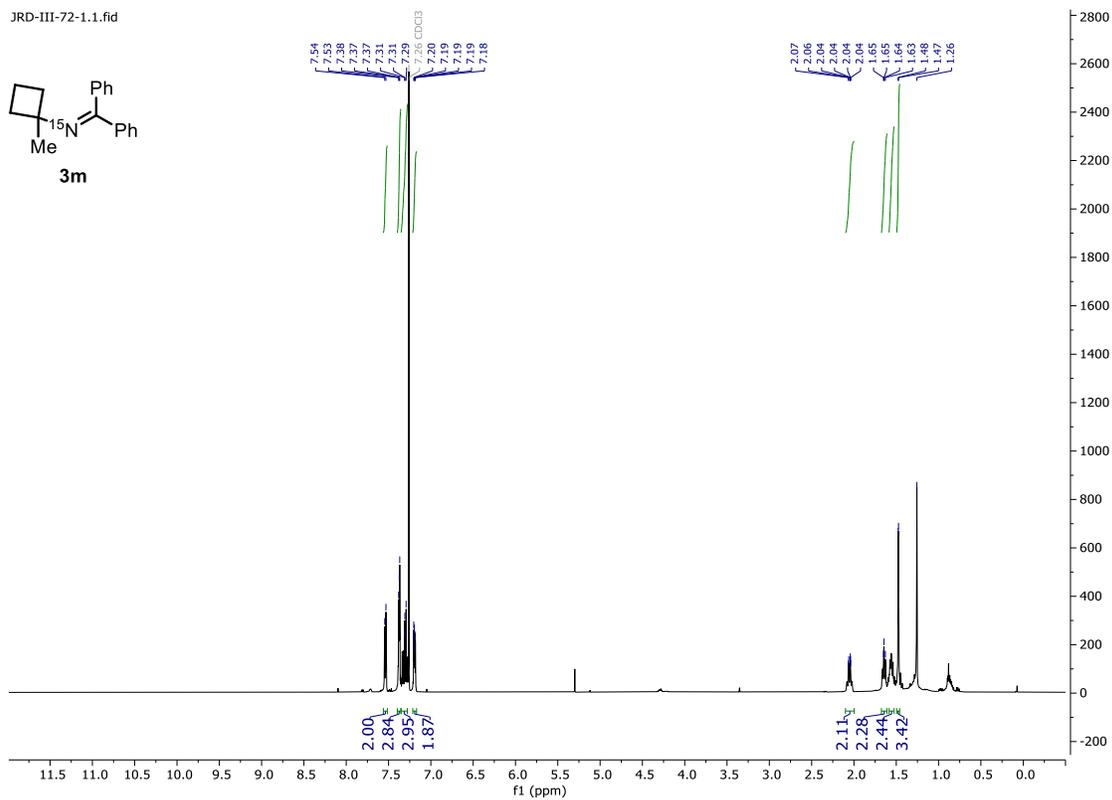
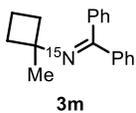
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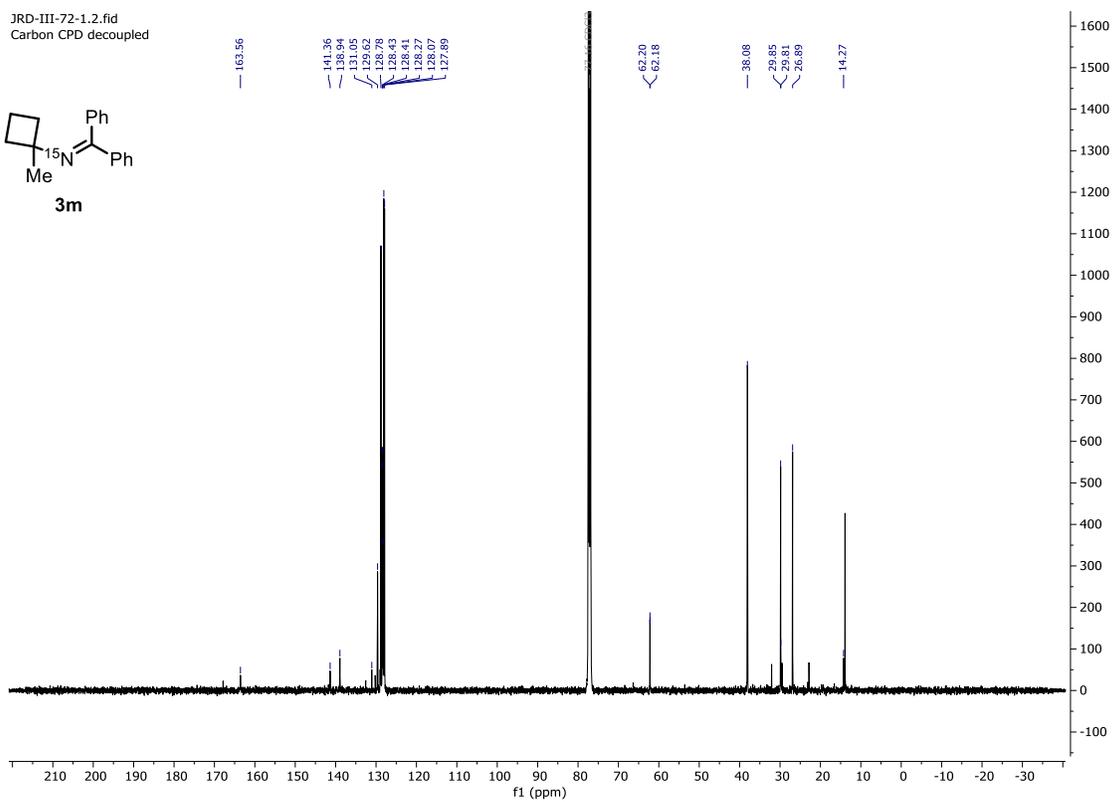
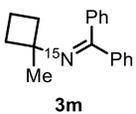
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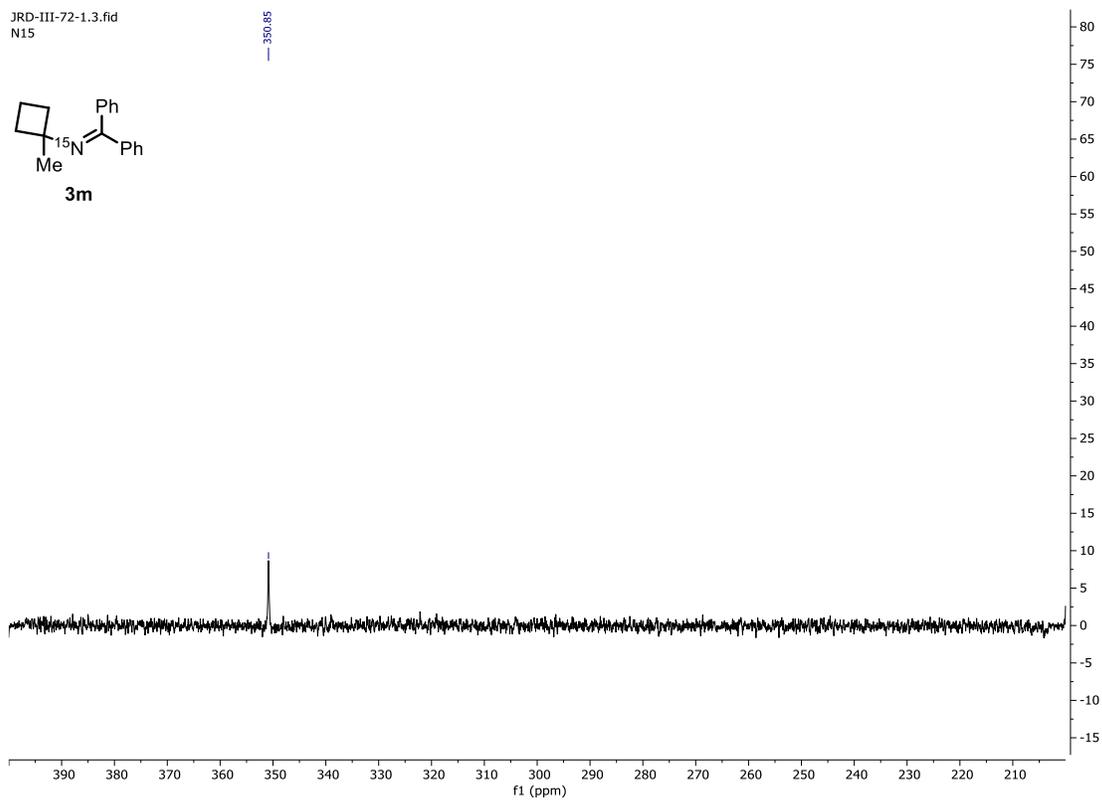
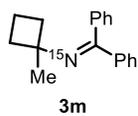
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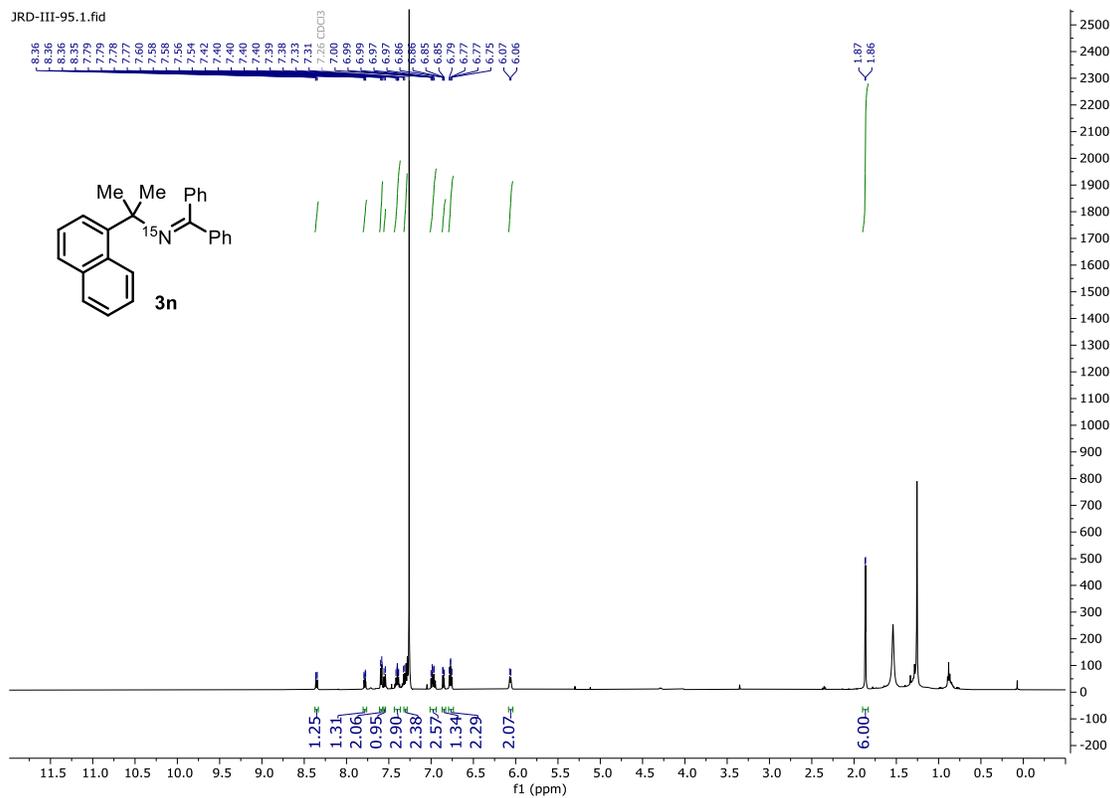
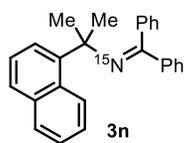
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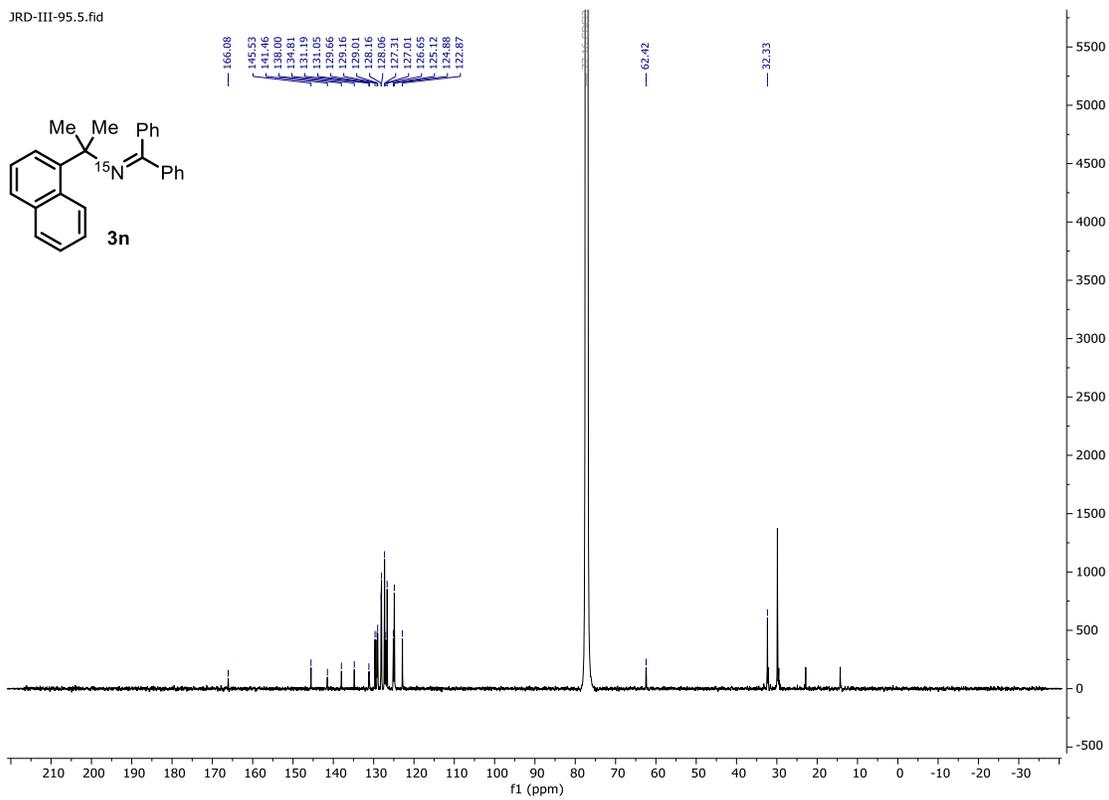
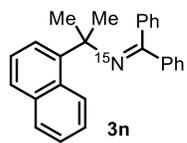
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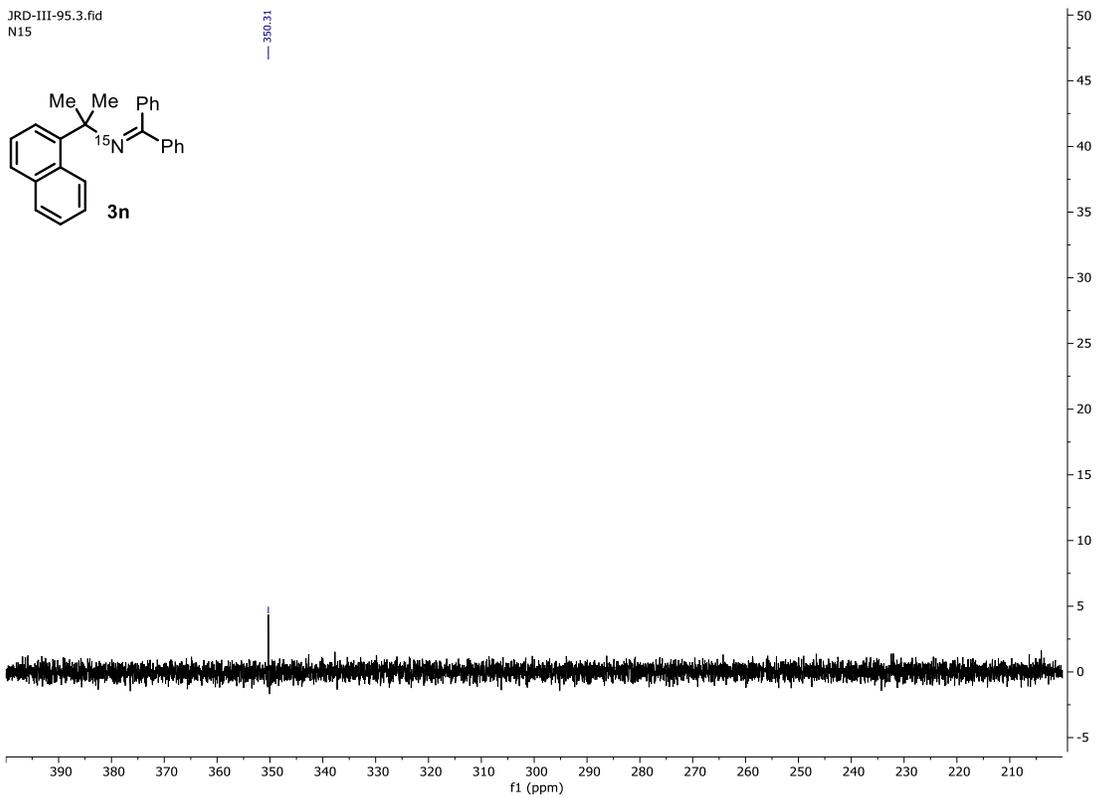
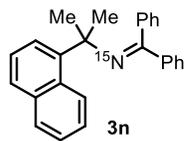
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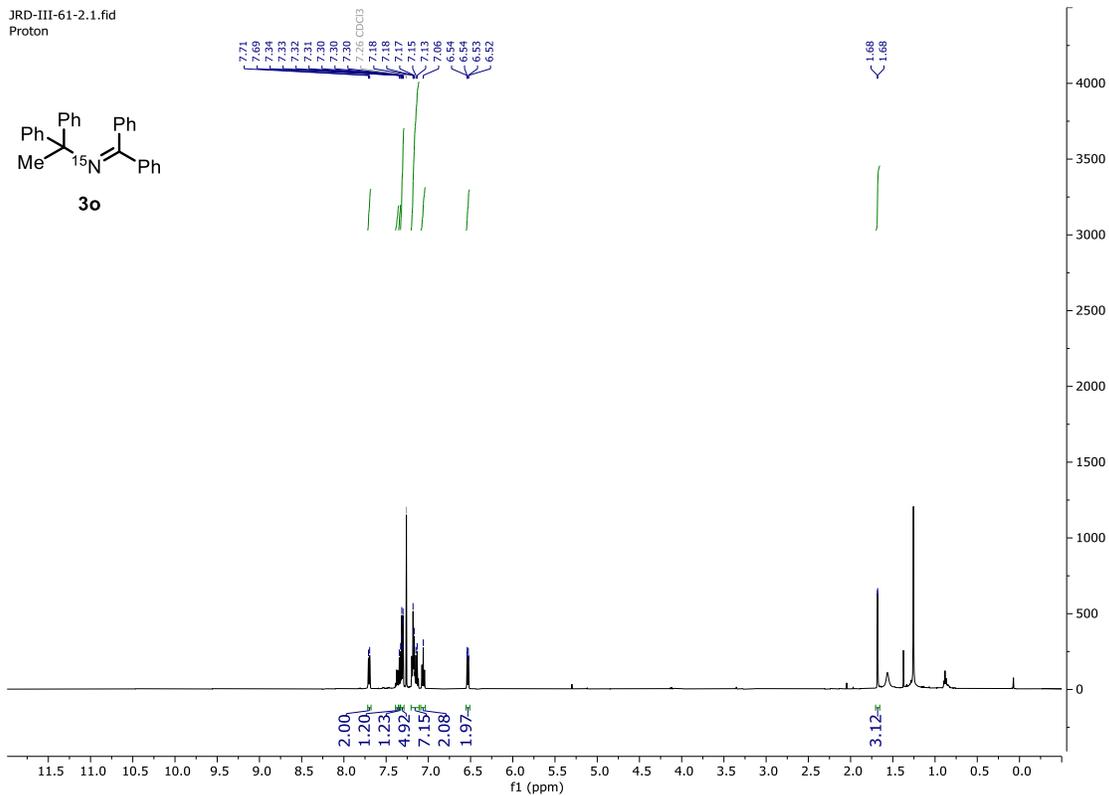
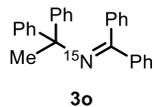
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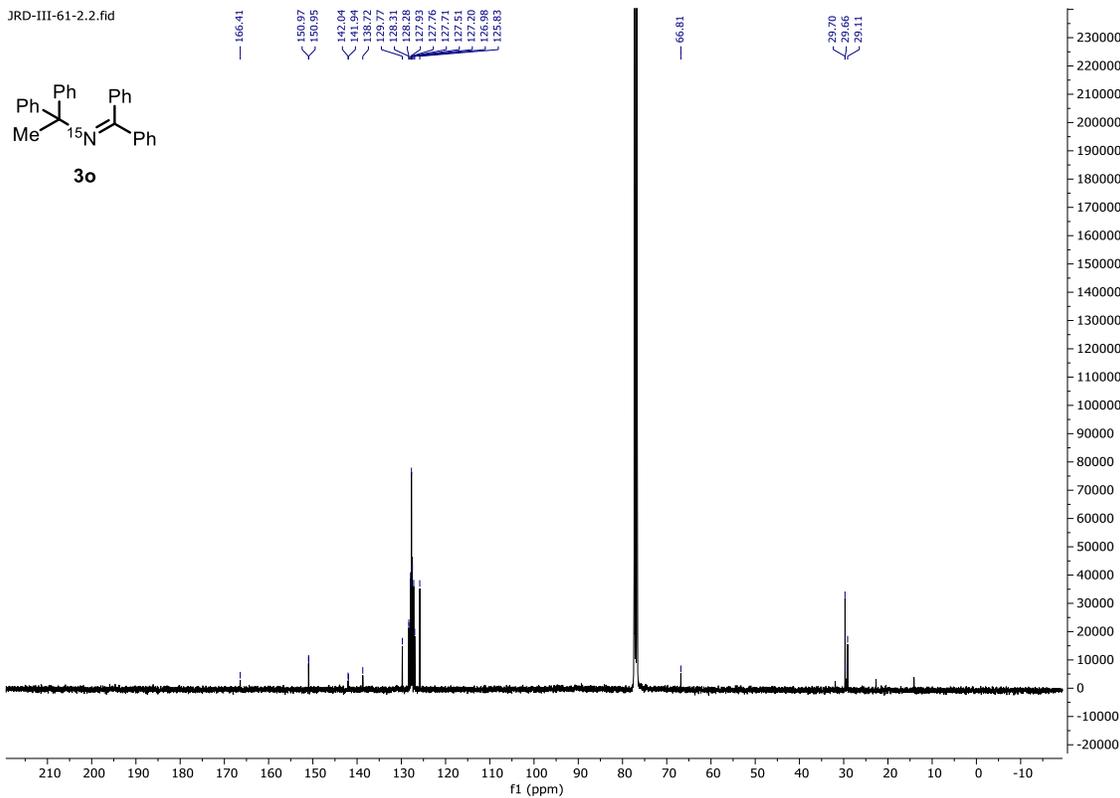
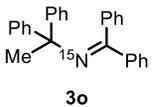
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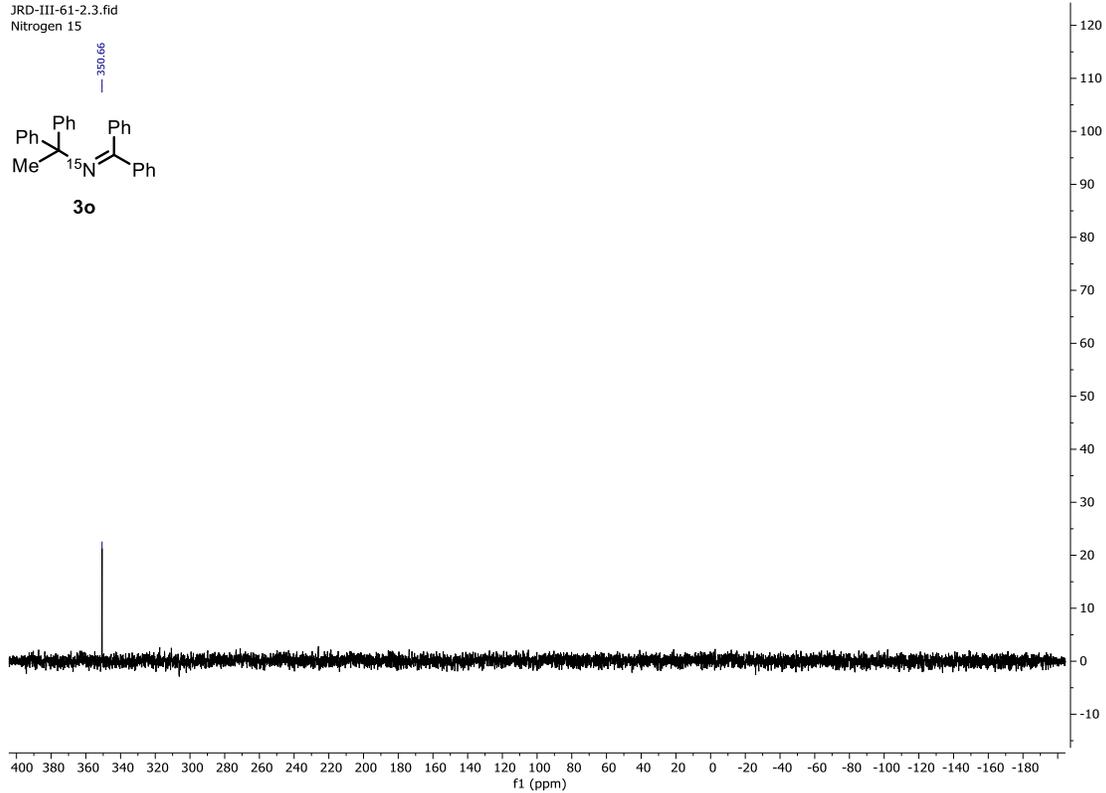
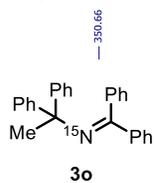
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Proton



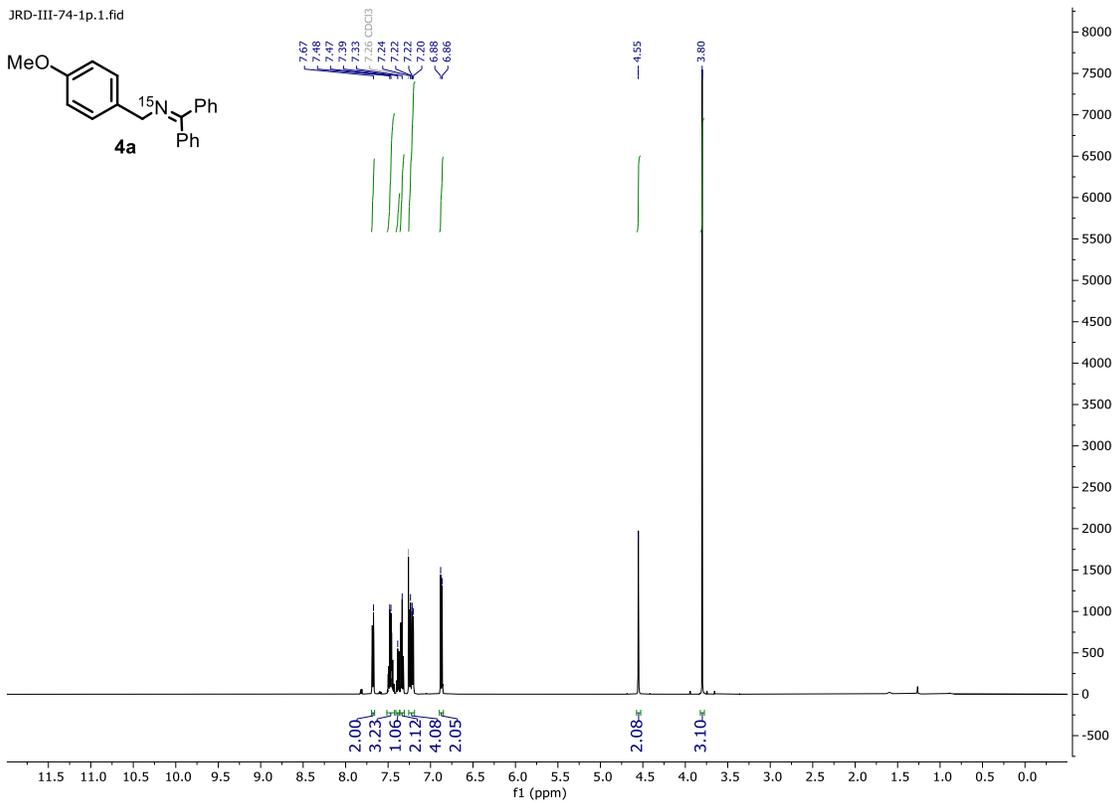
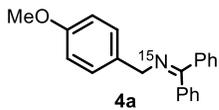
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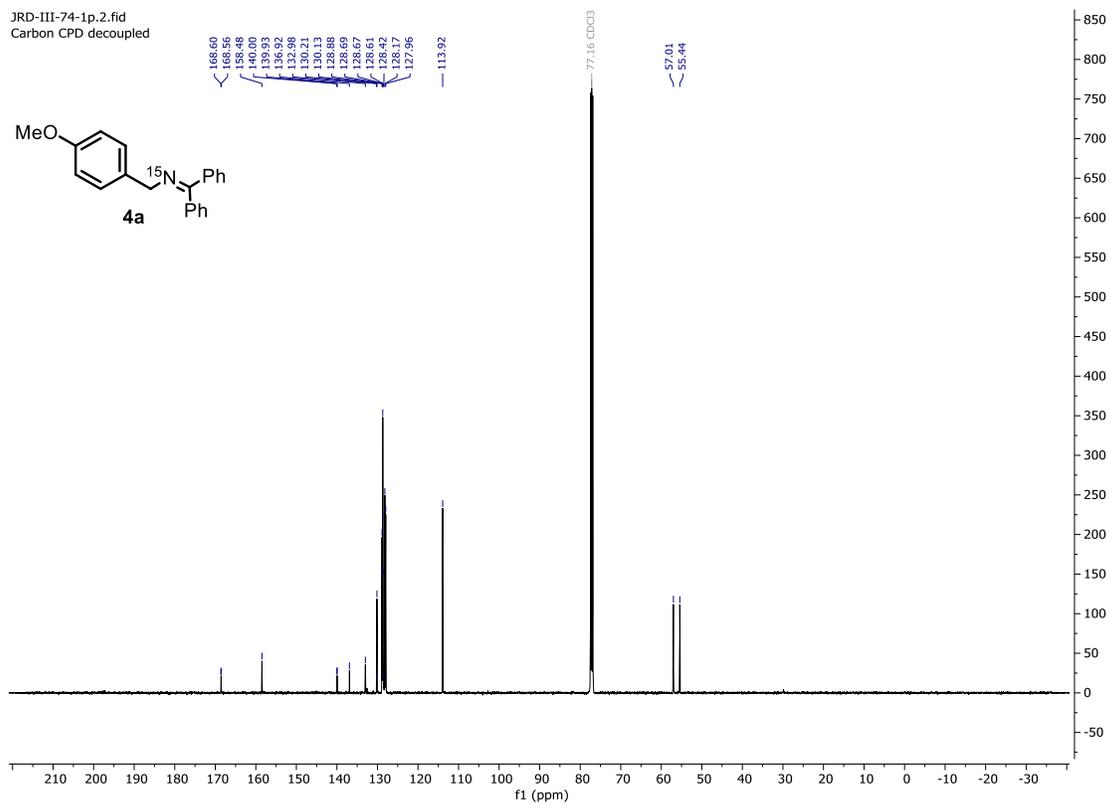
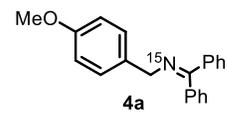
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Nitrogen 15



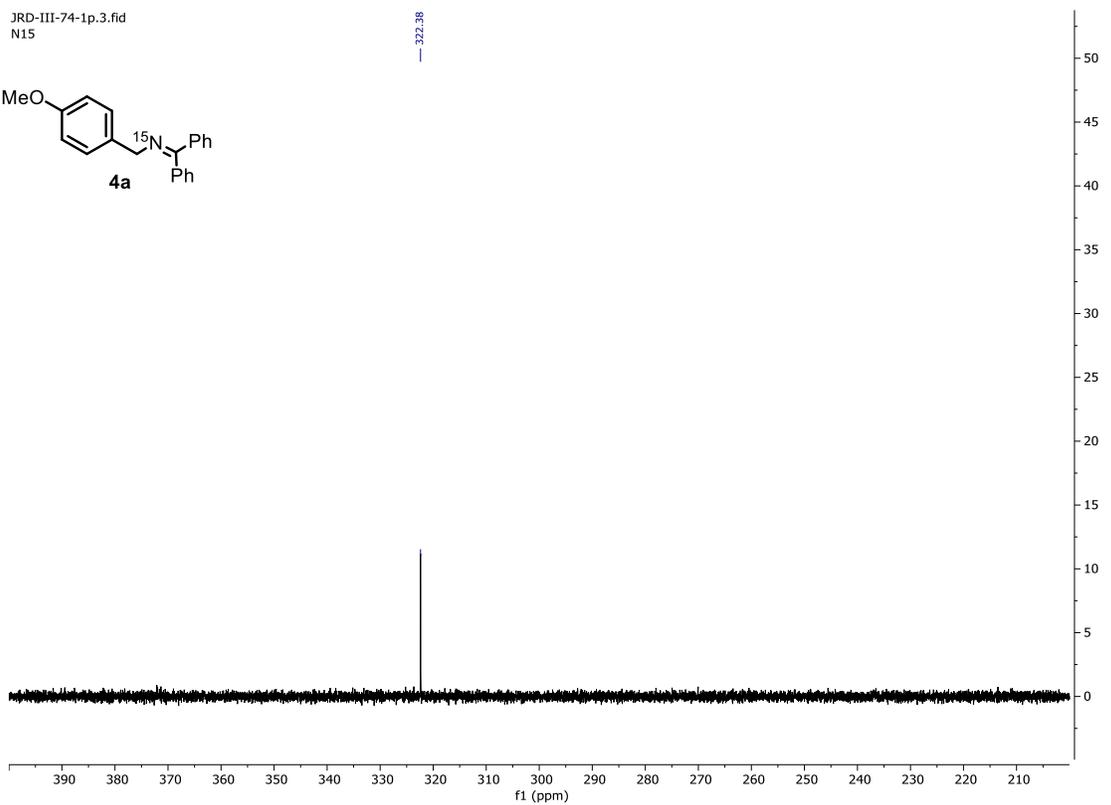
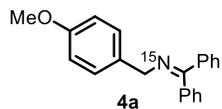
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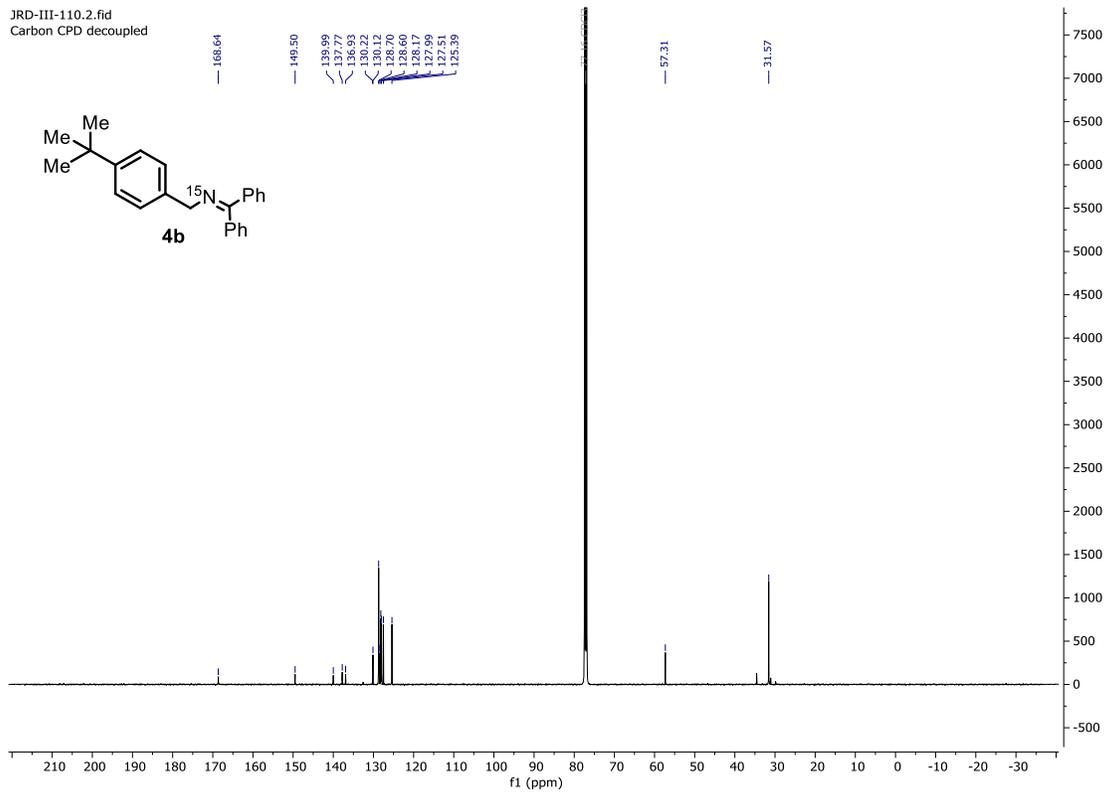
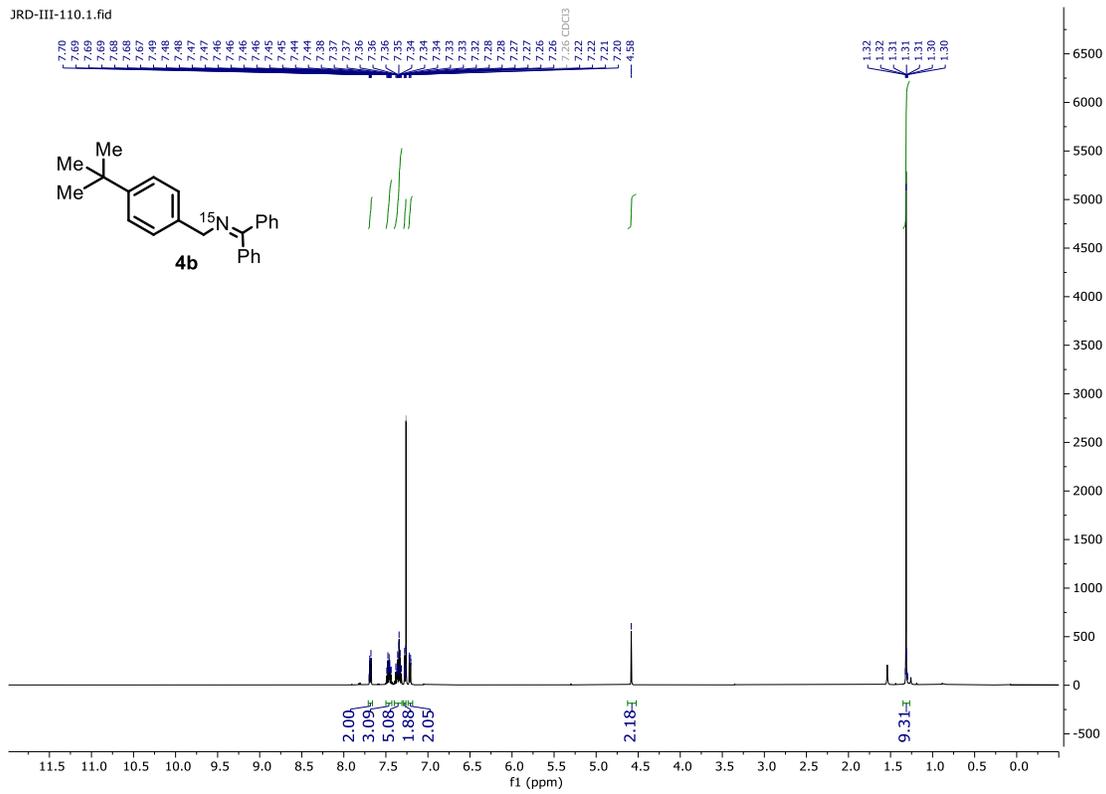


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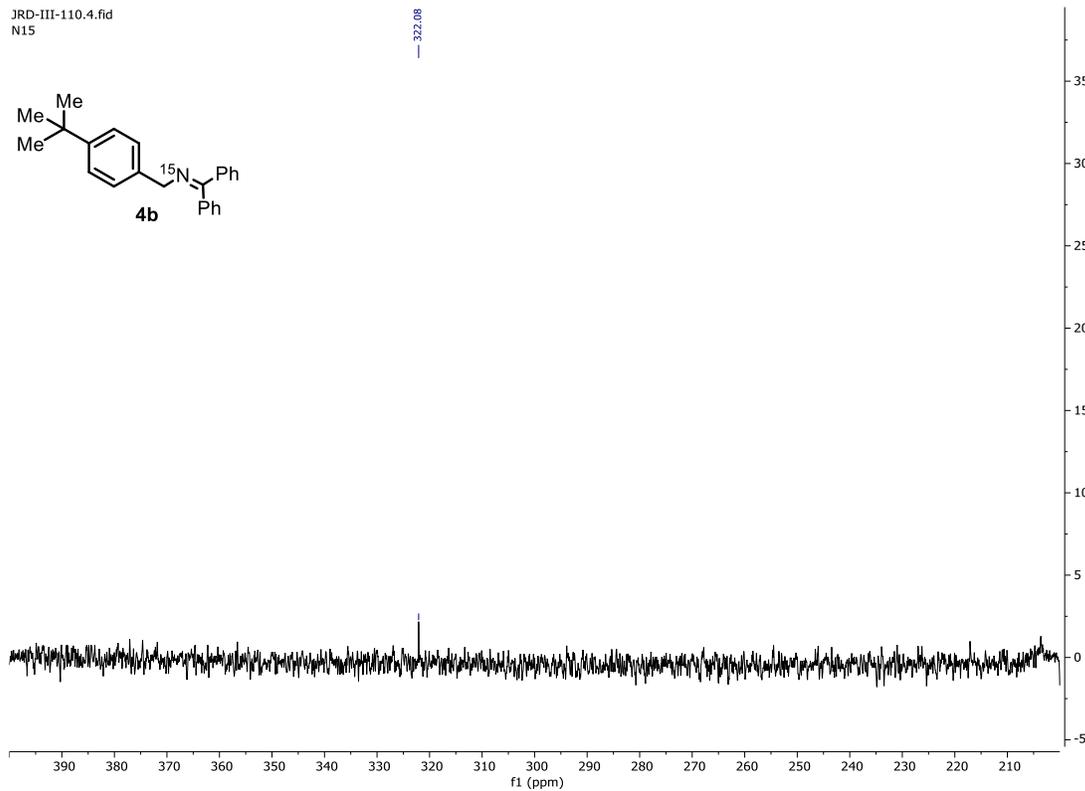
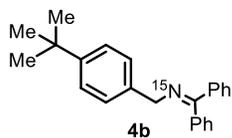


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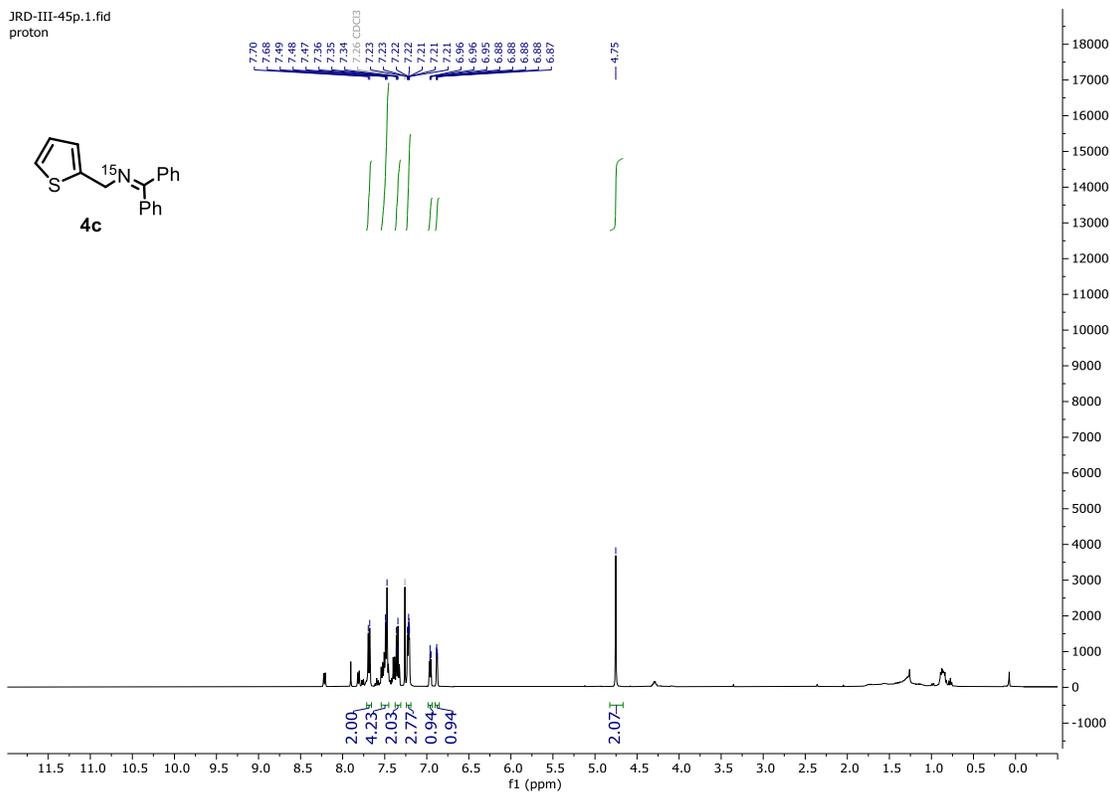
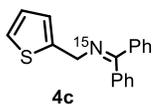




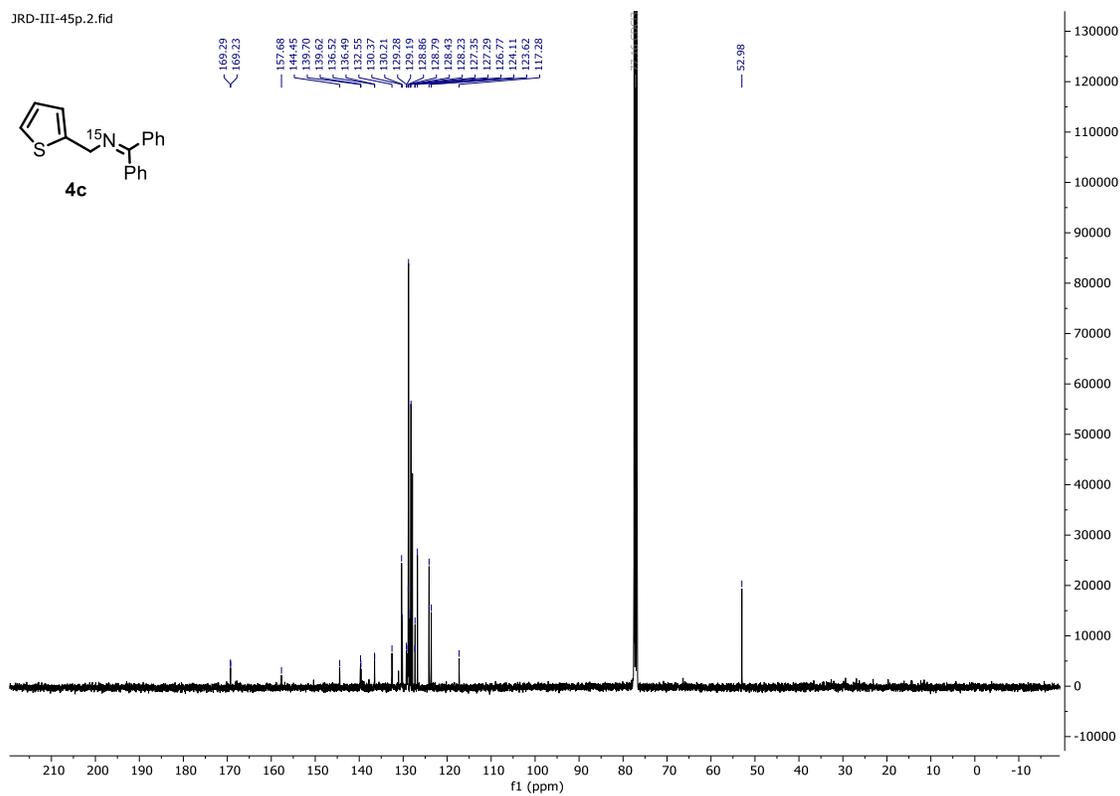
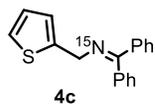
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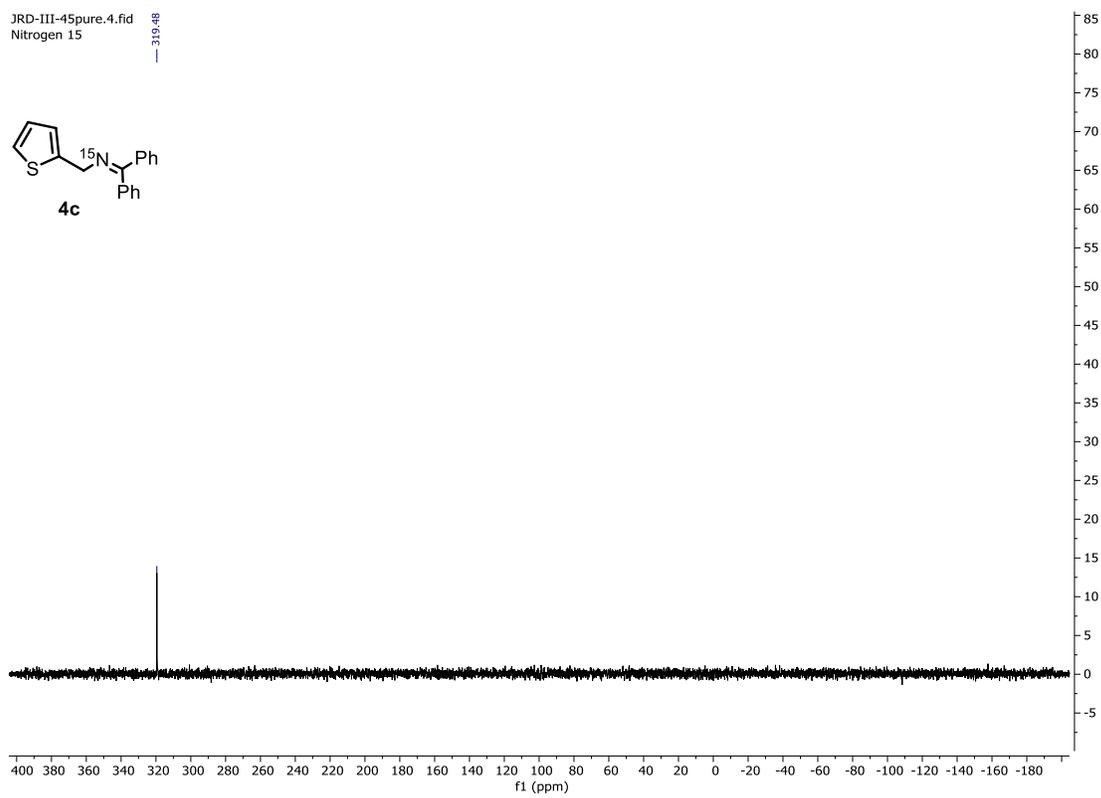
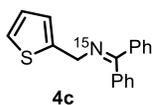
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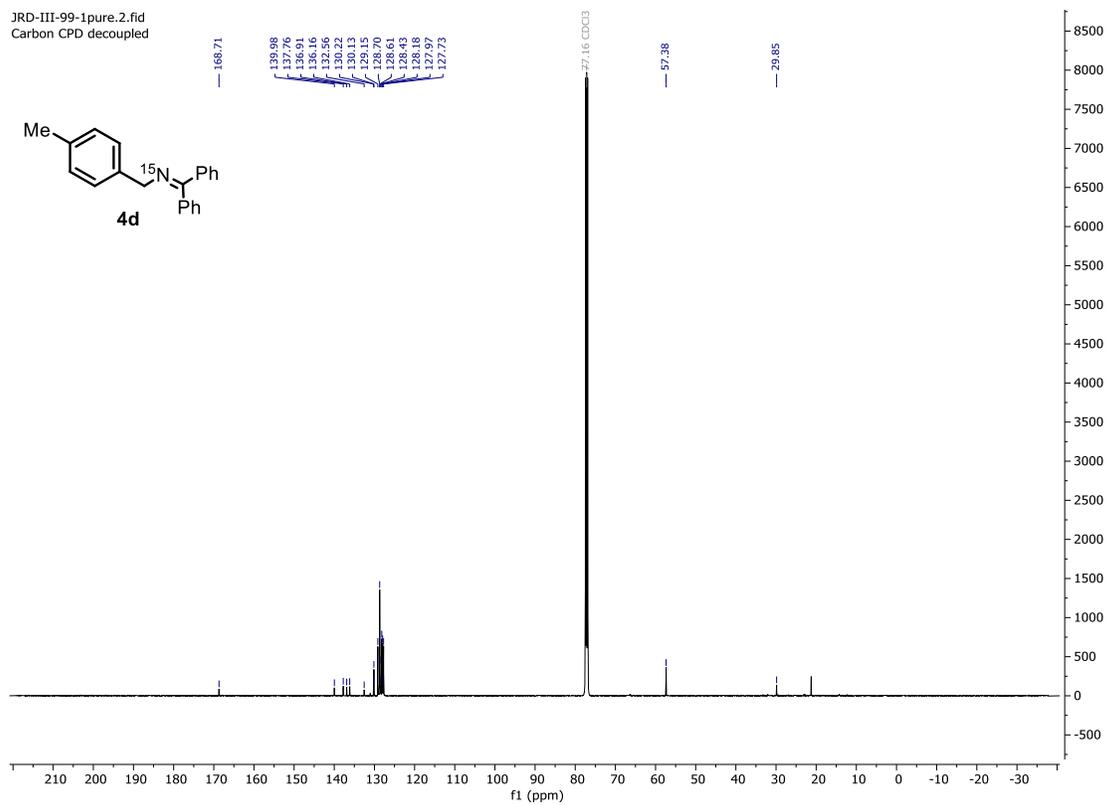
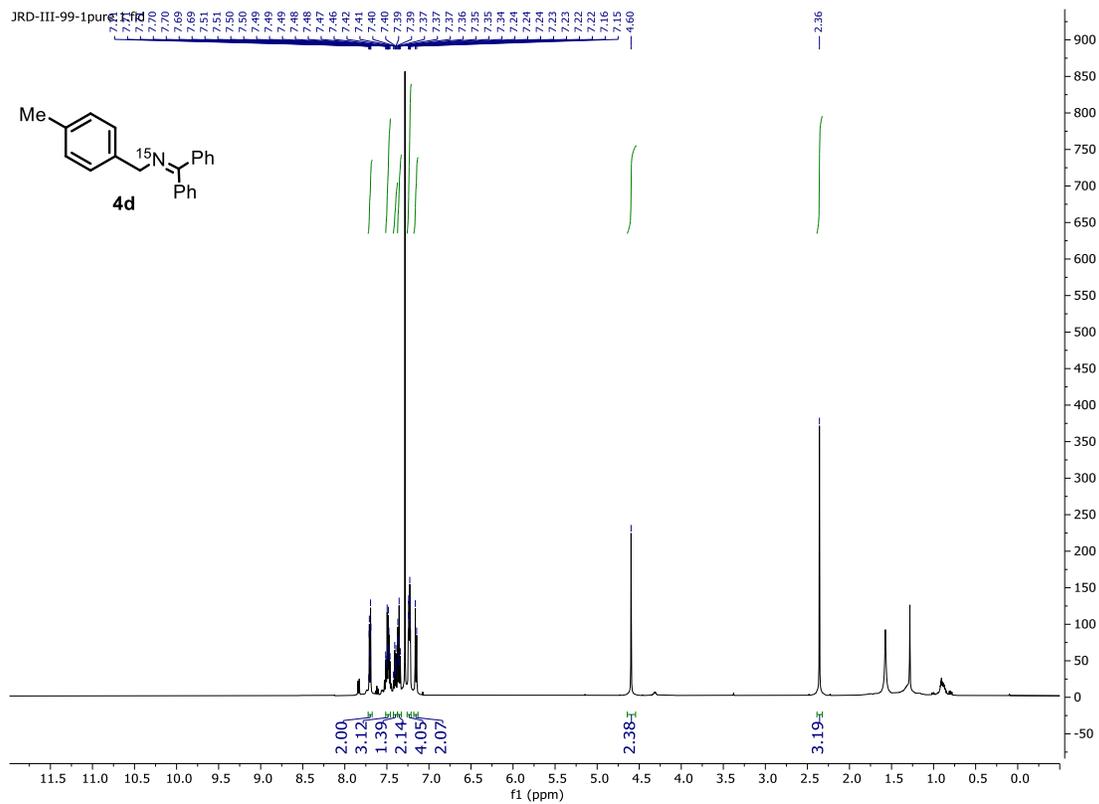


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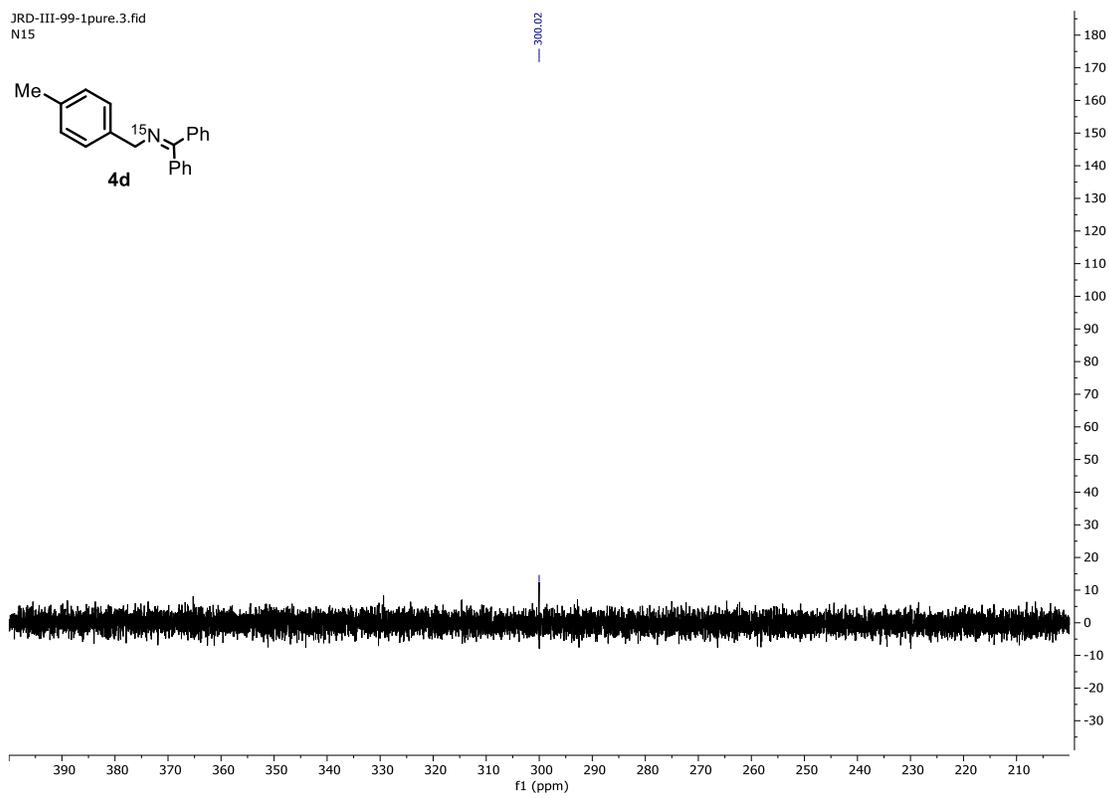
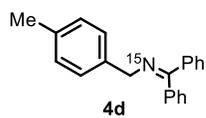


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Nitrogen 15

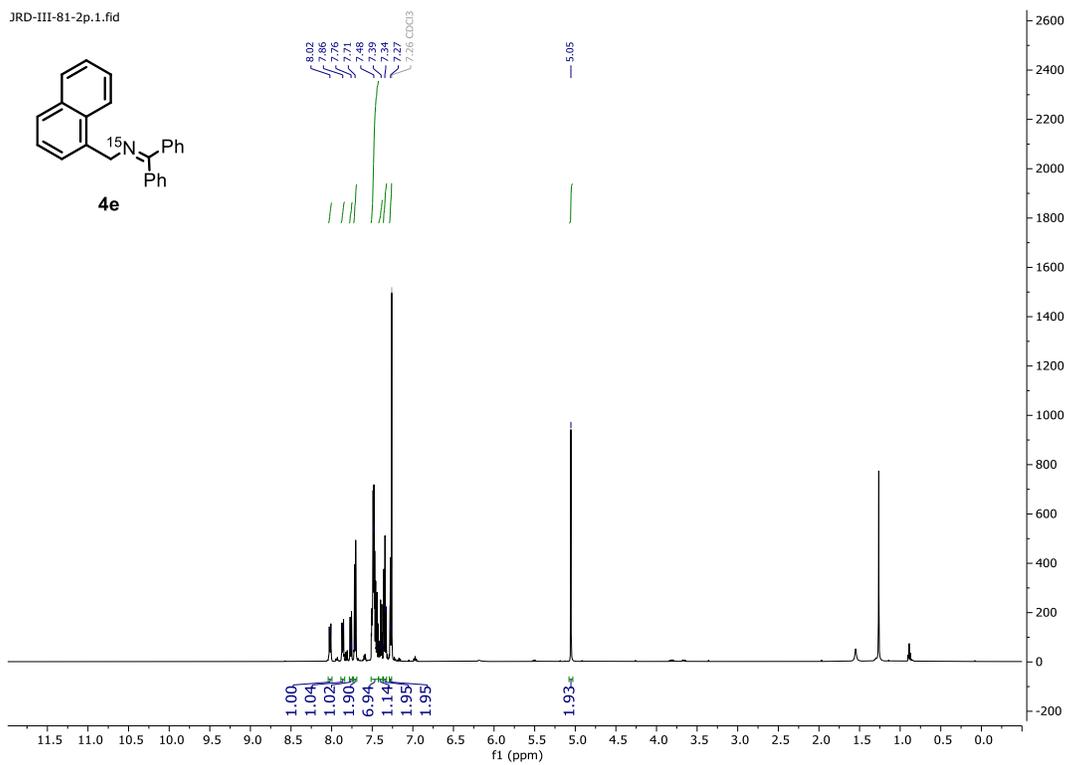
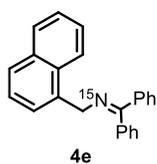




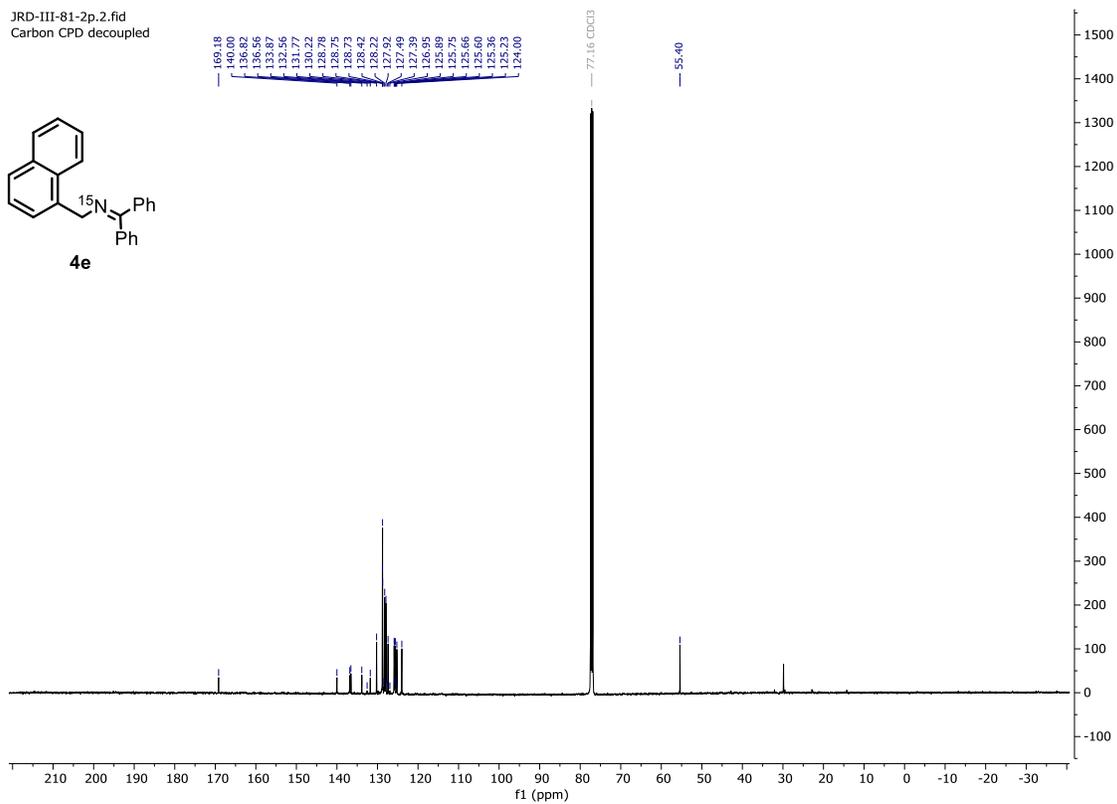
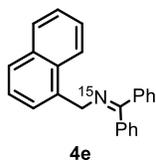
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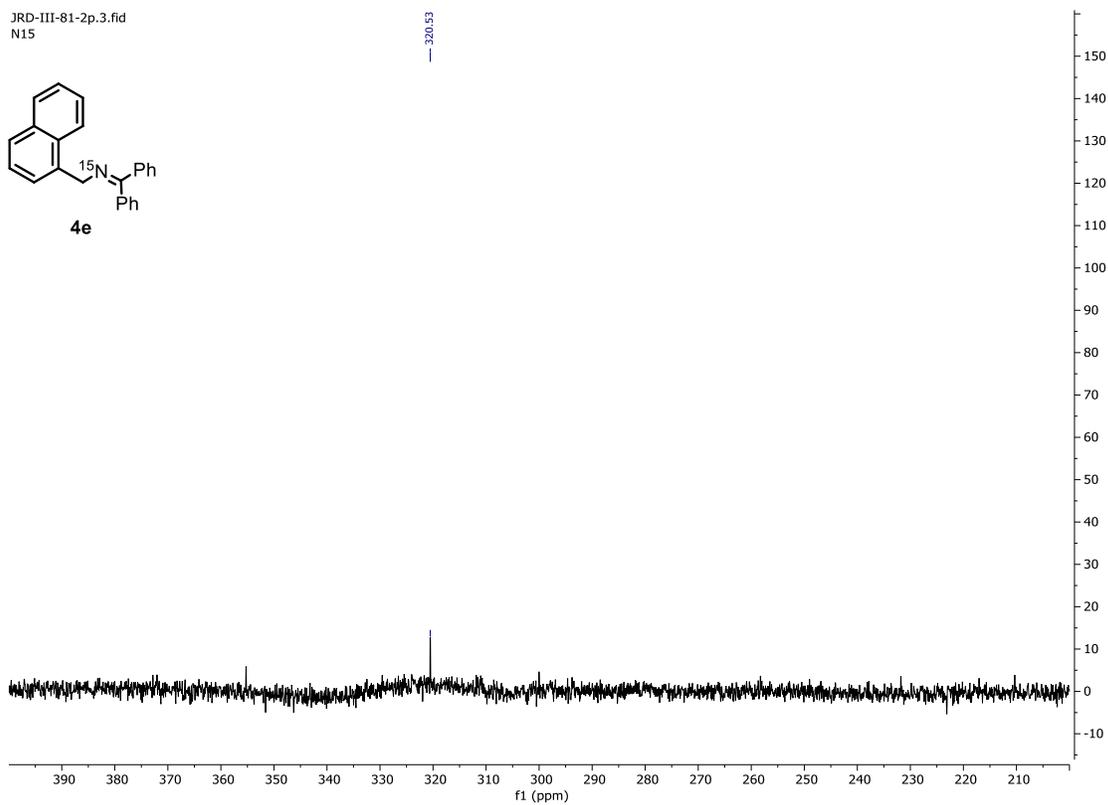
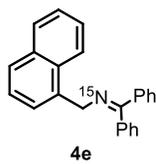
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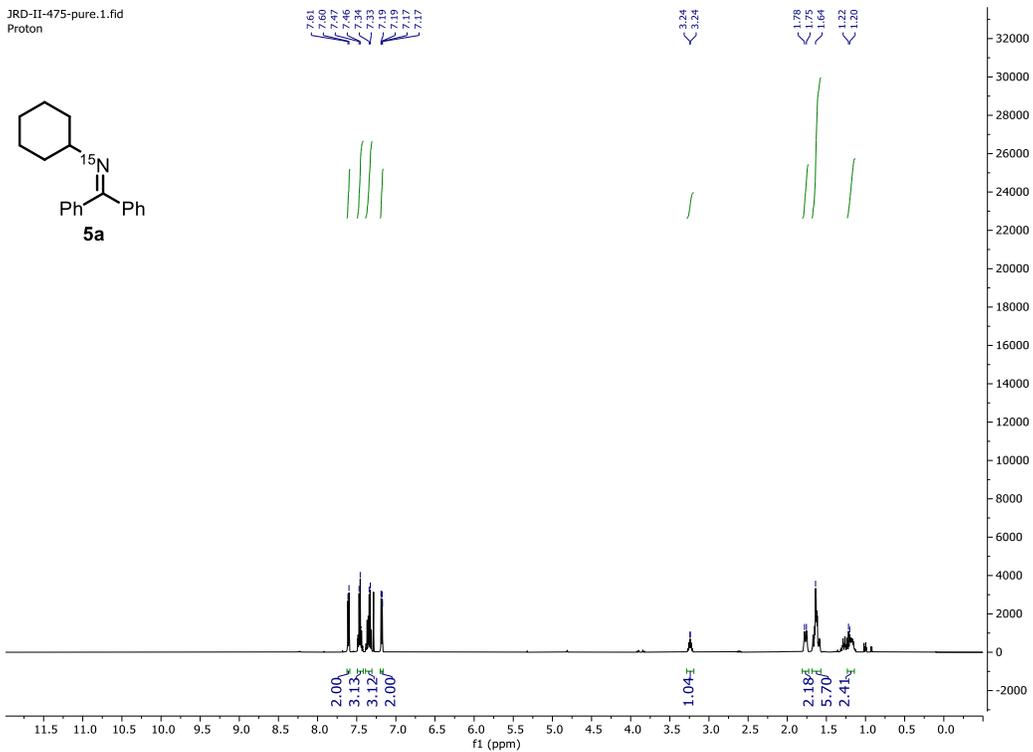
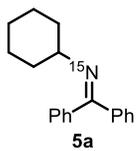
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Carbon CPD decoupled



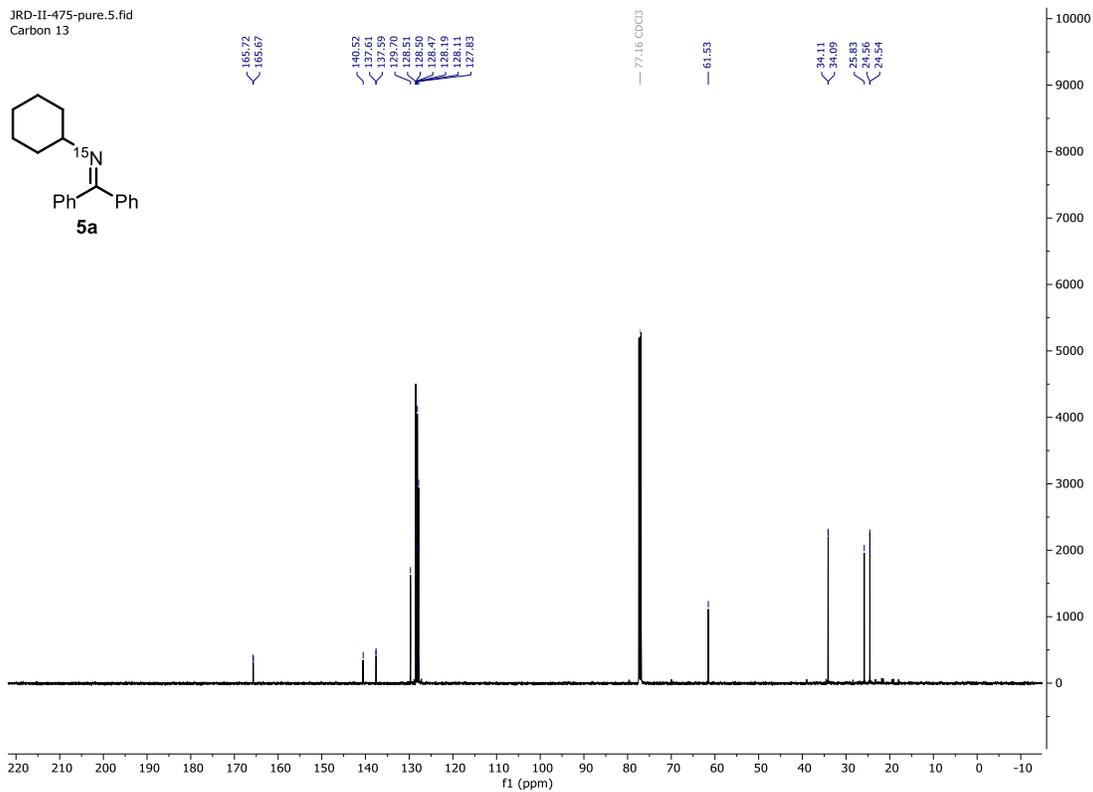
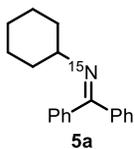
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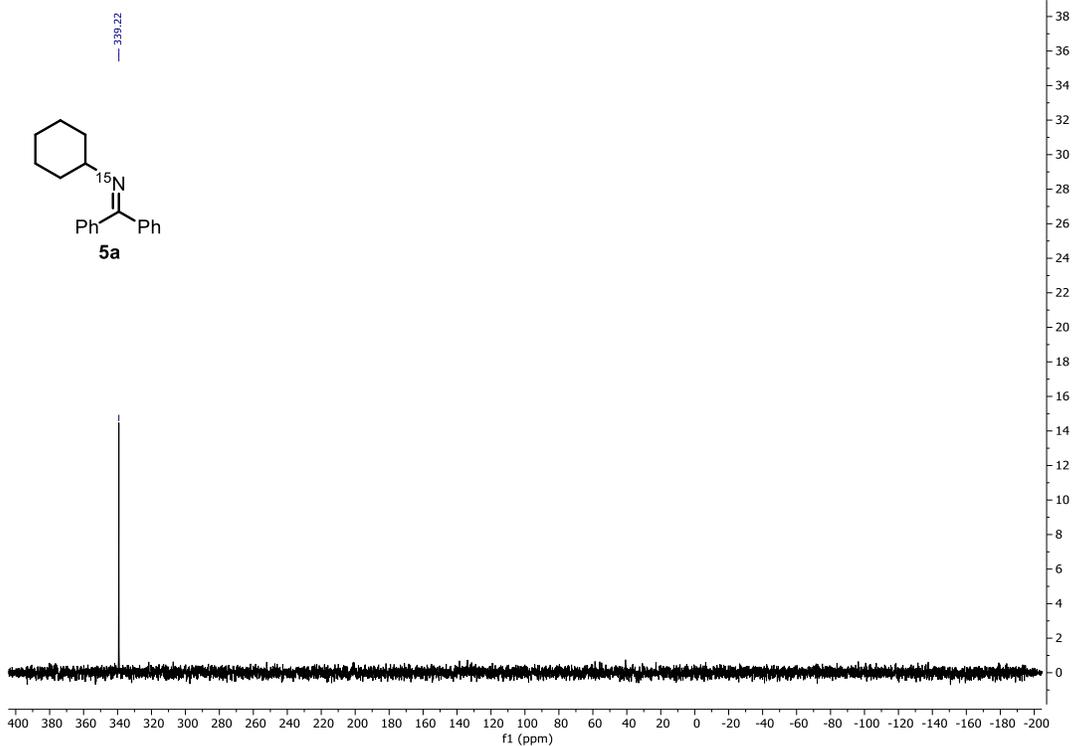
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Proton



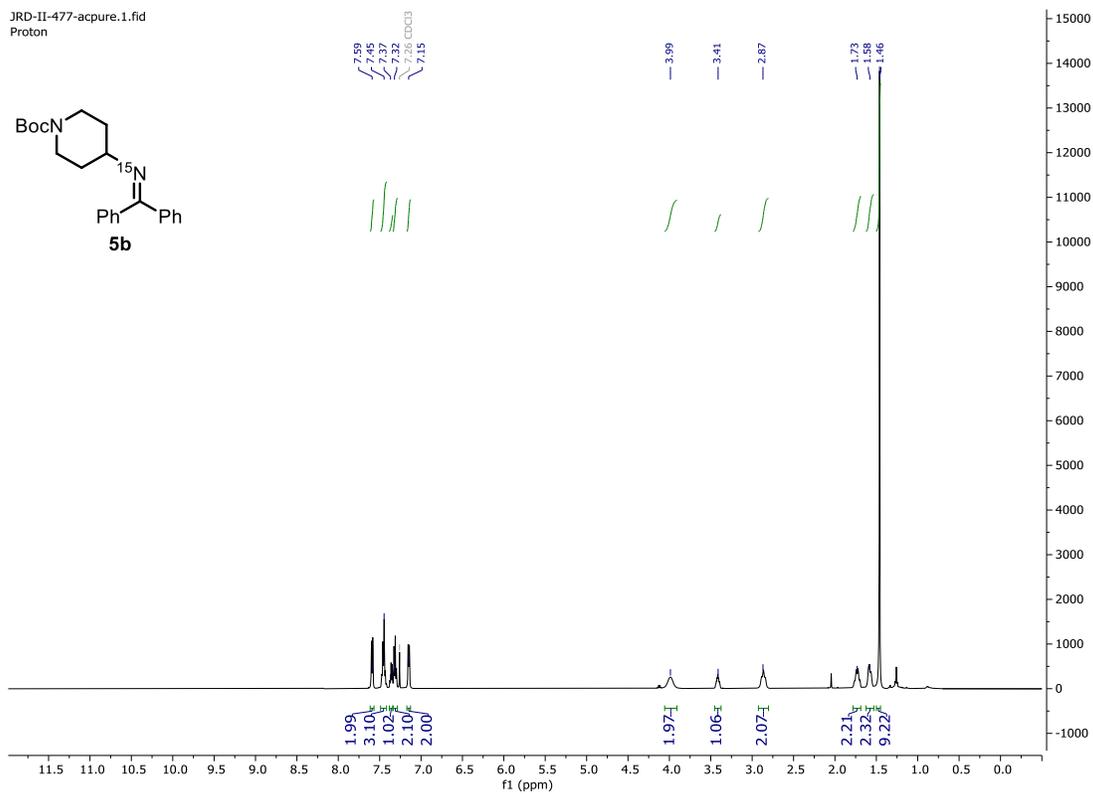
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Carbon 13



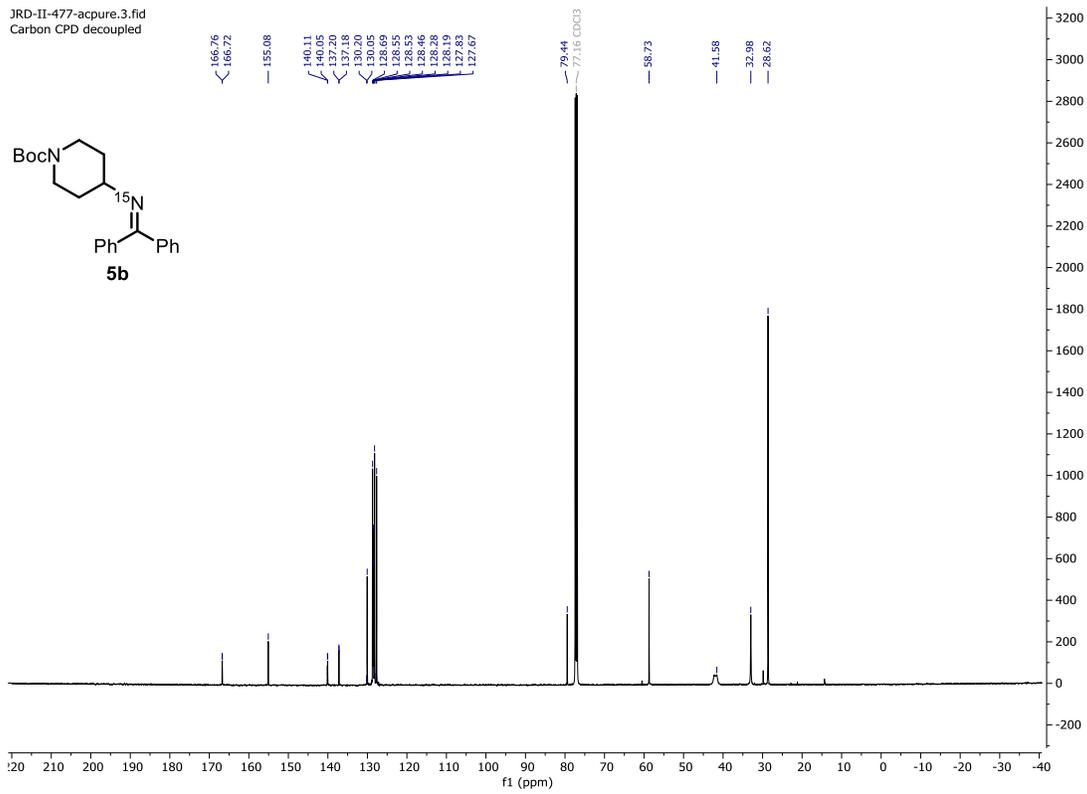
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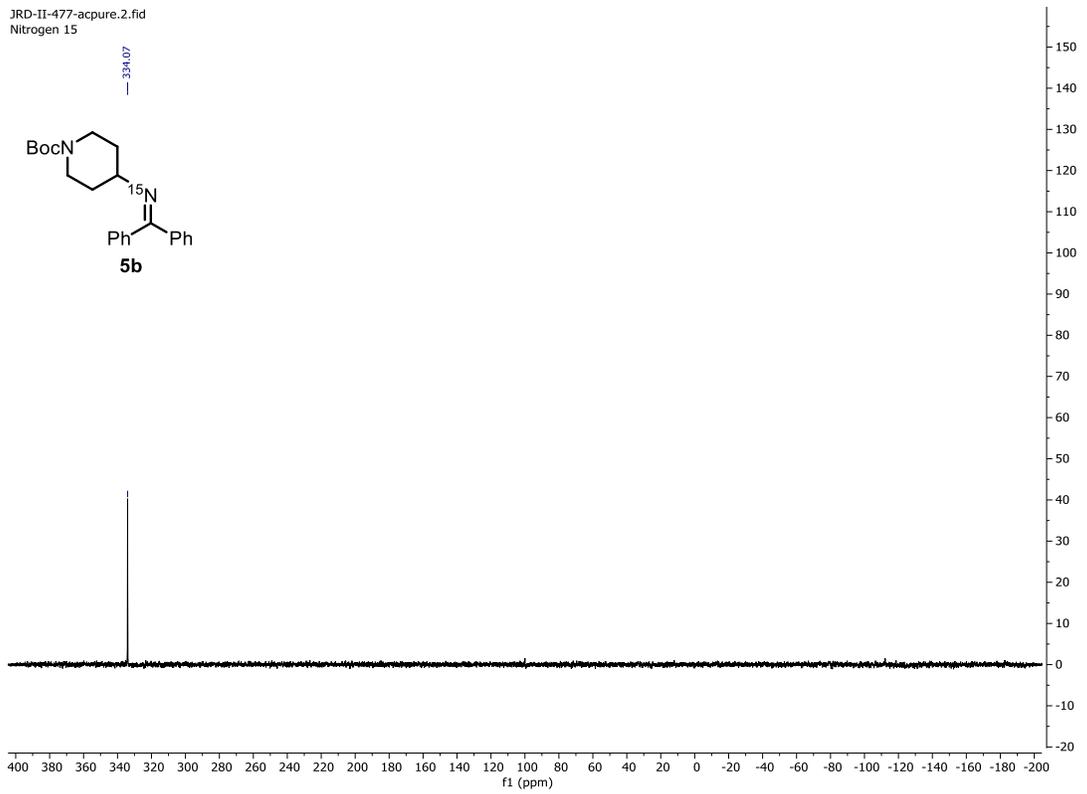
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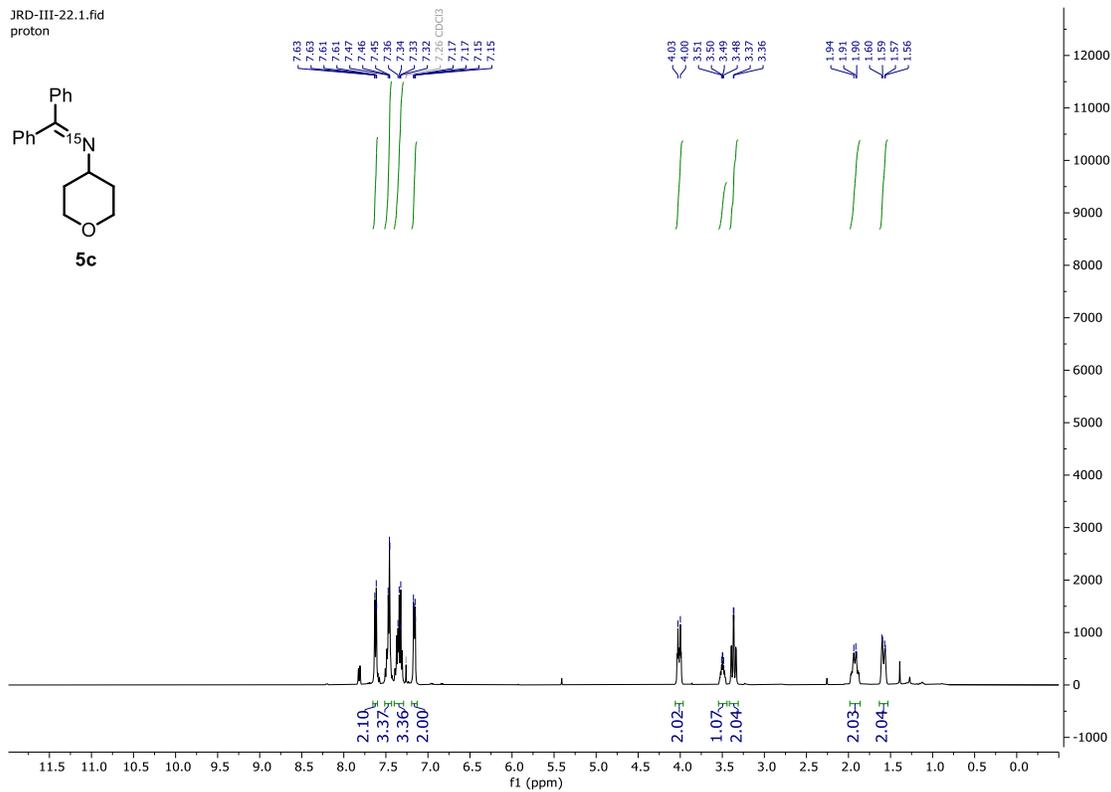
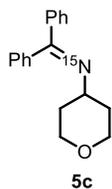
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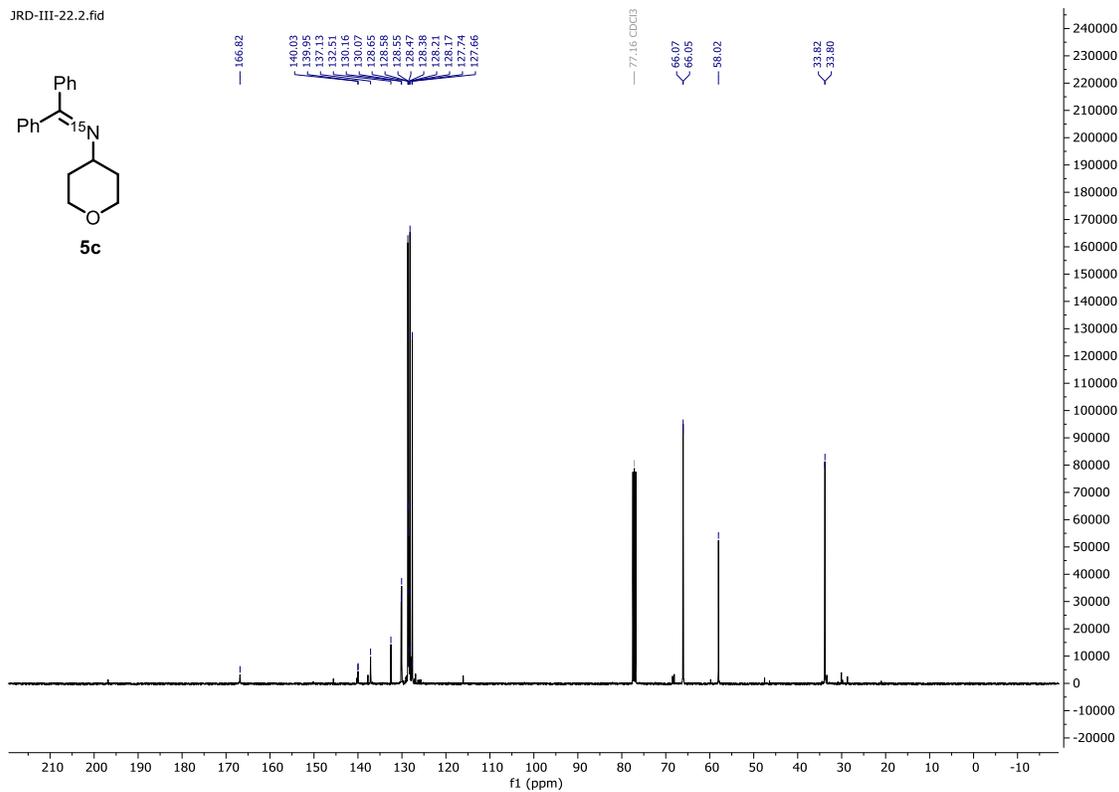
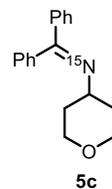
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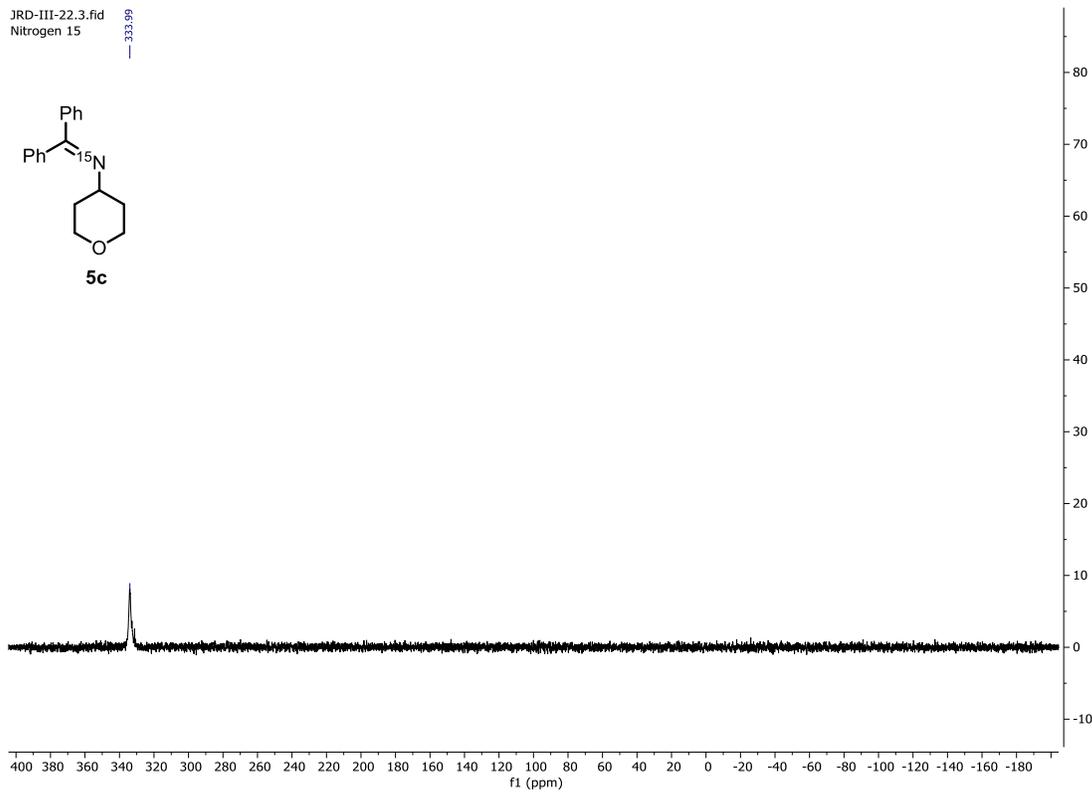
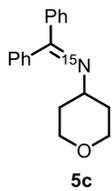
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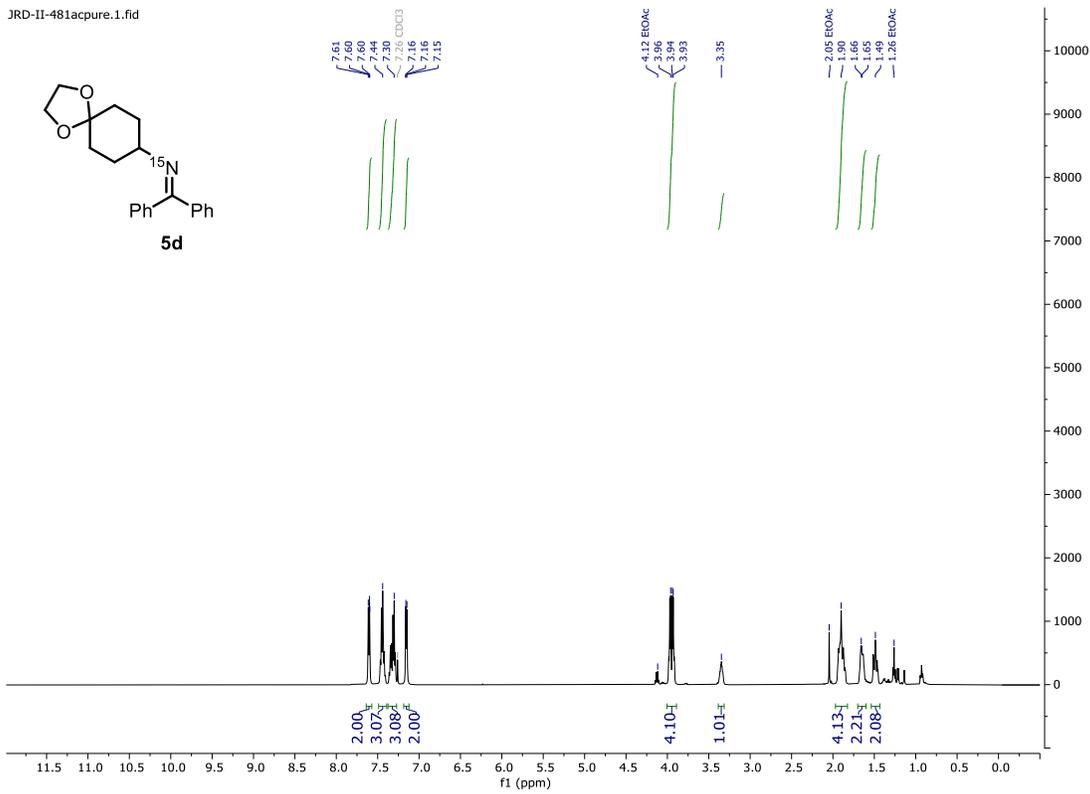
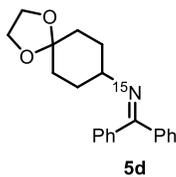
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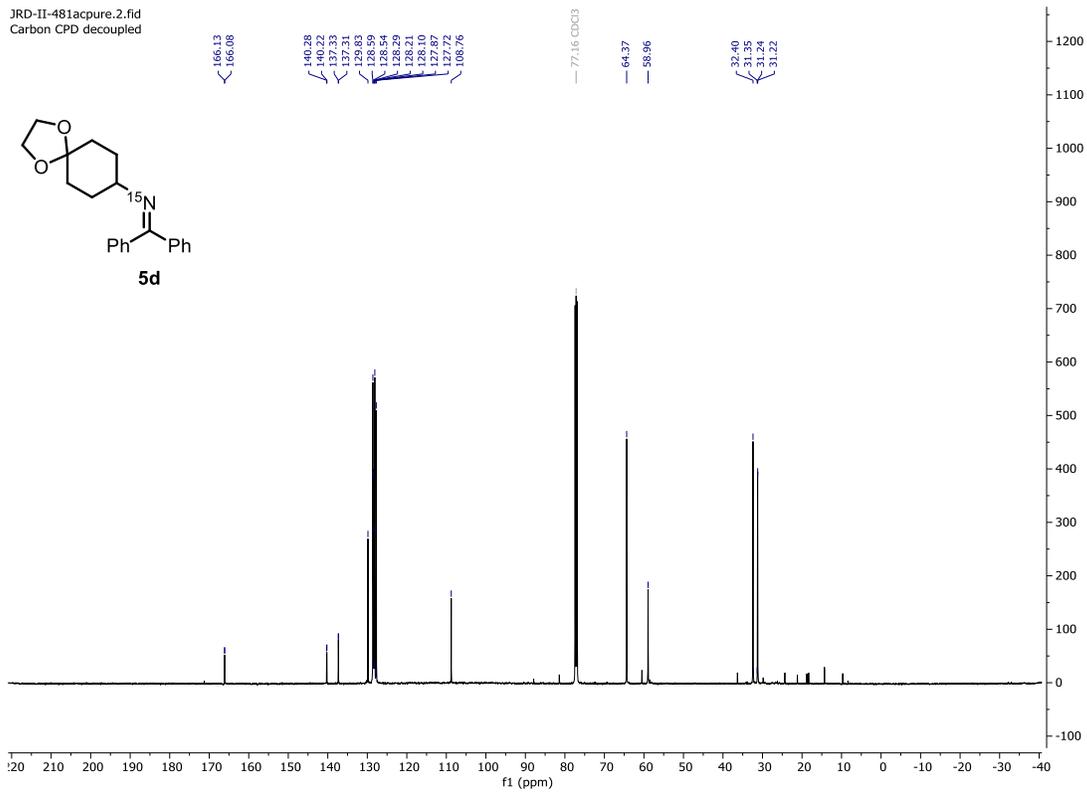
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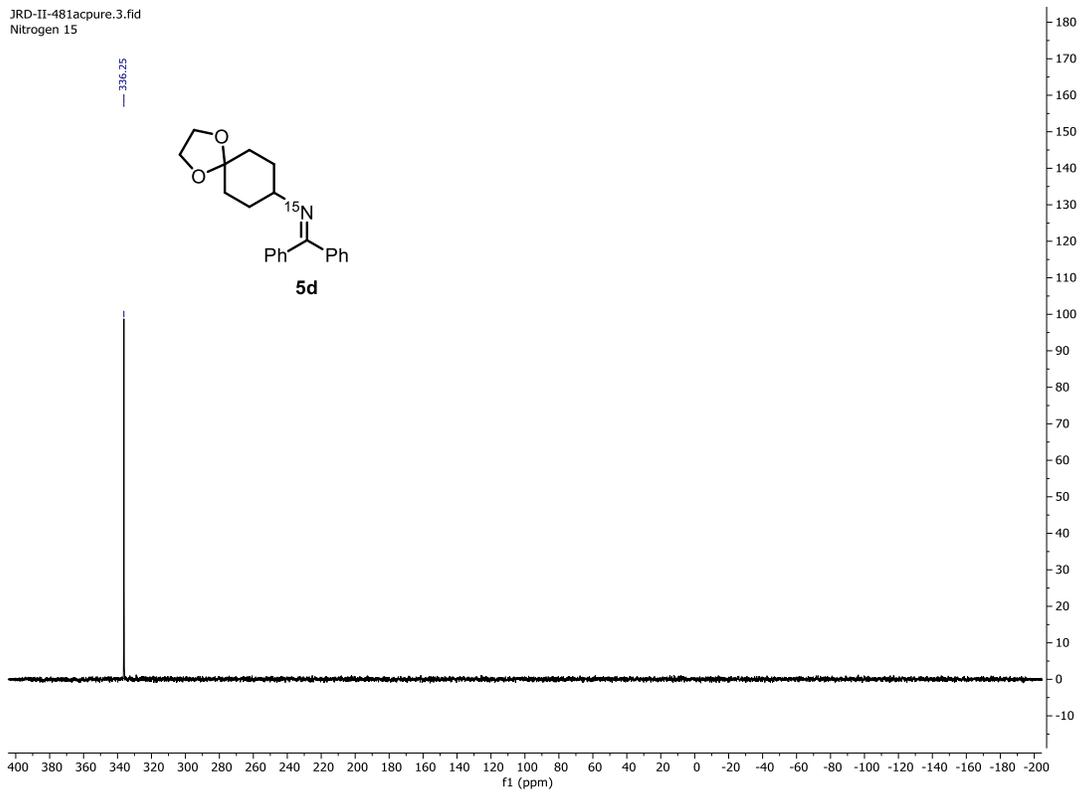
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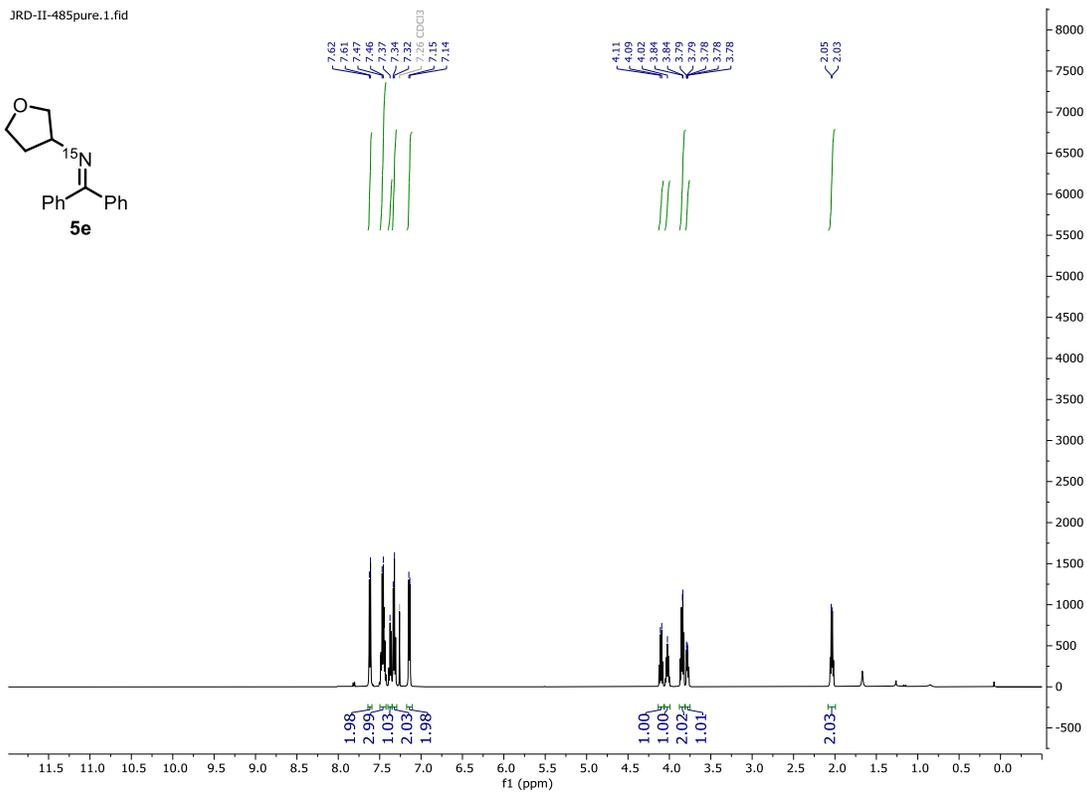
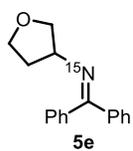
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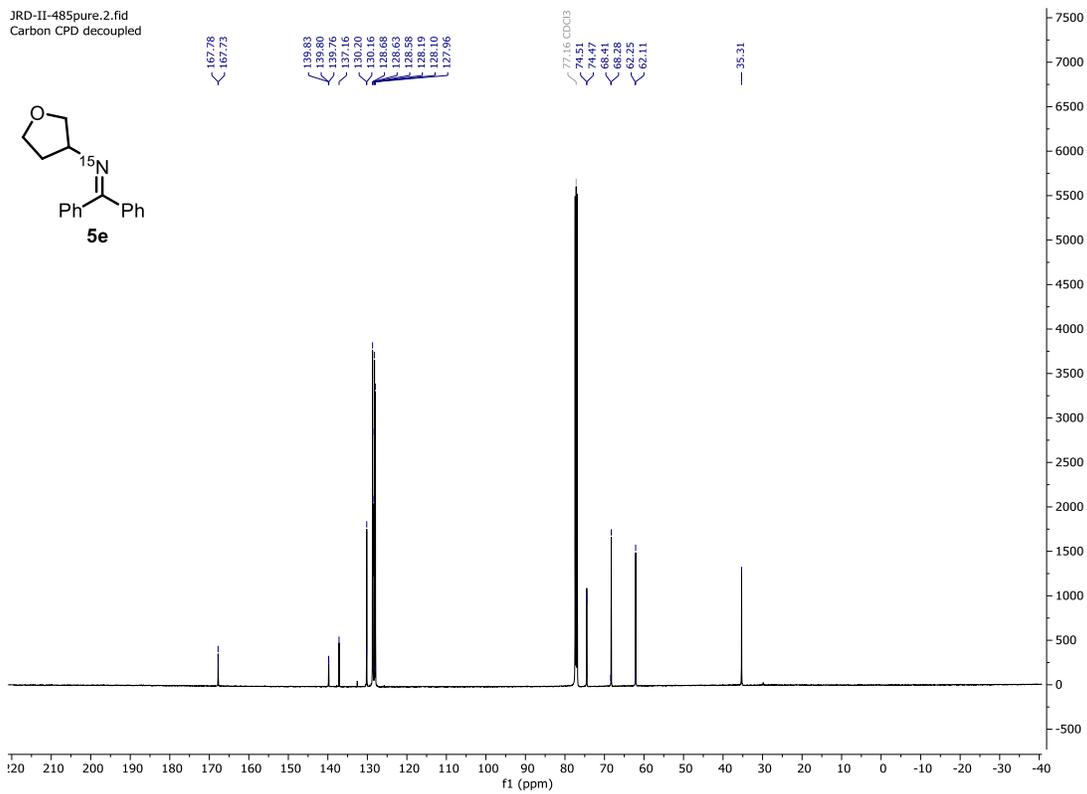
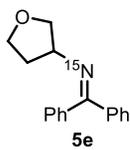
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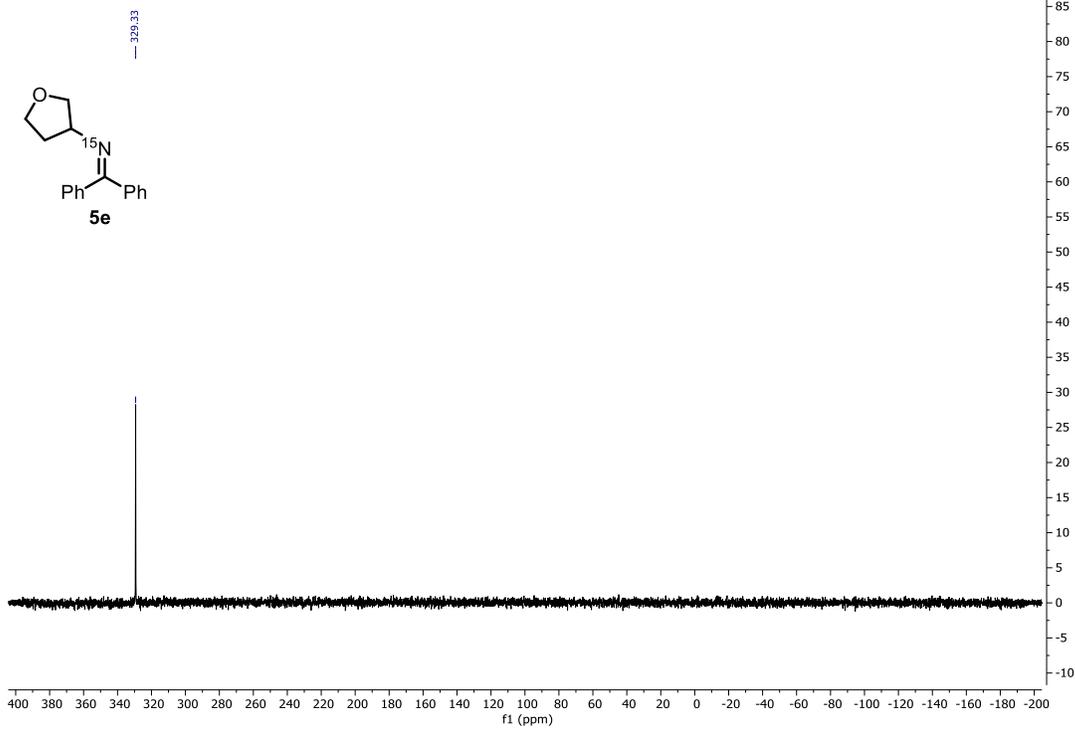
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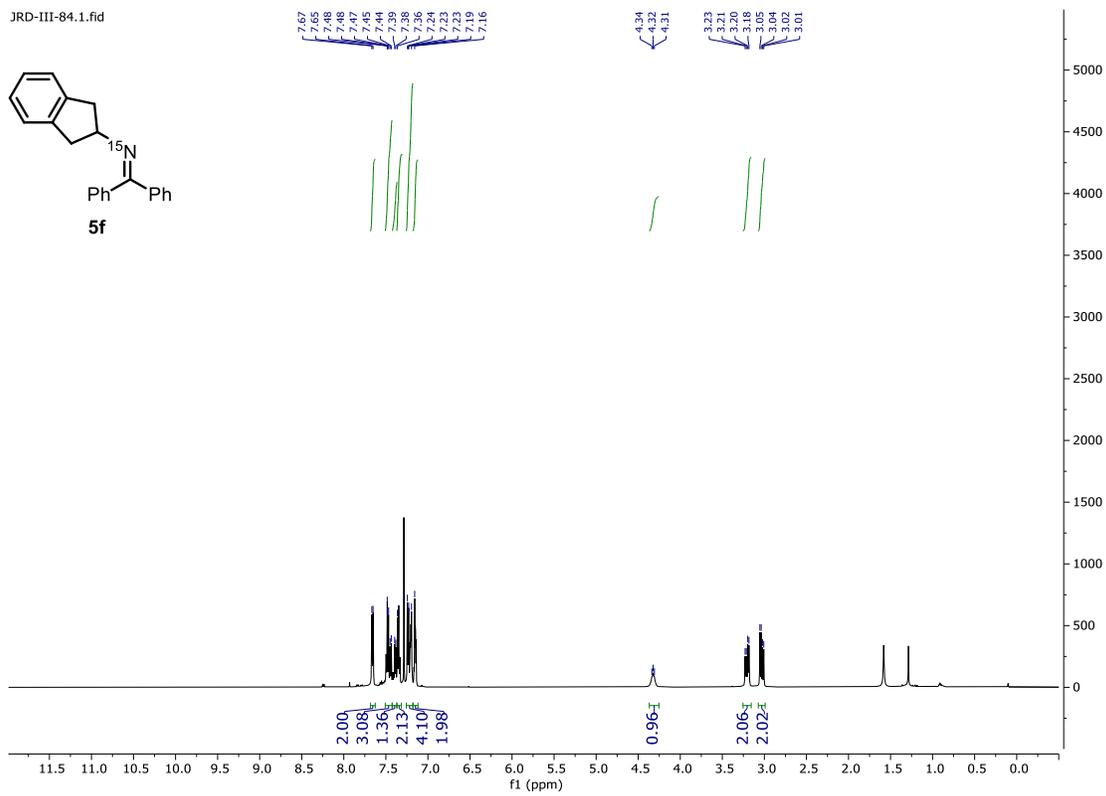
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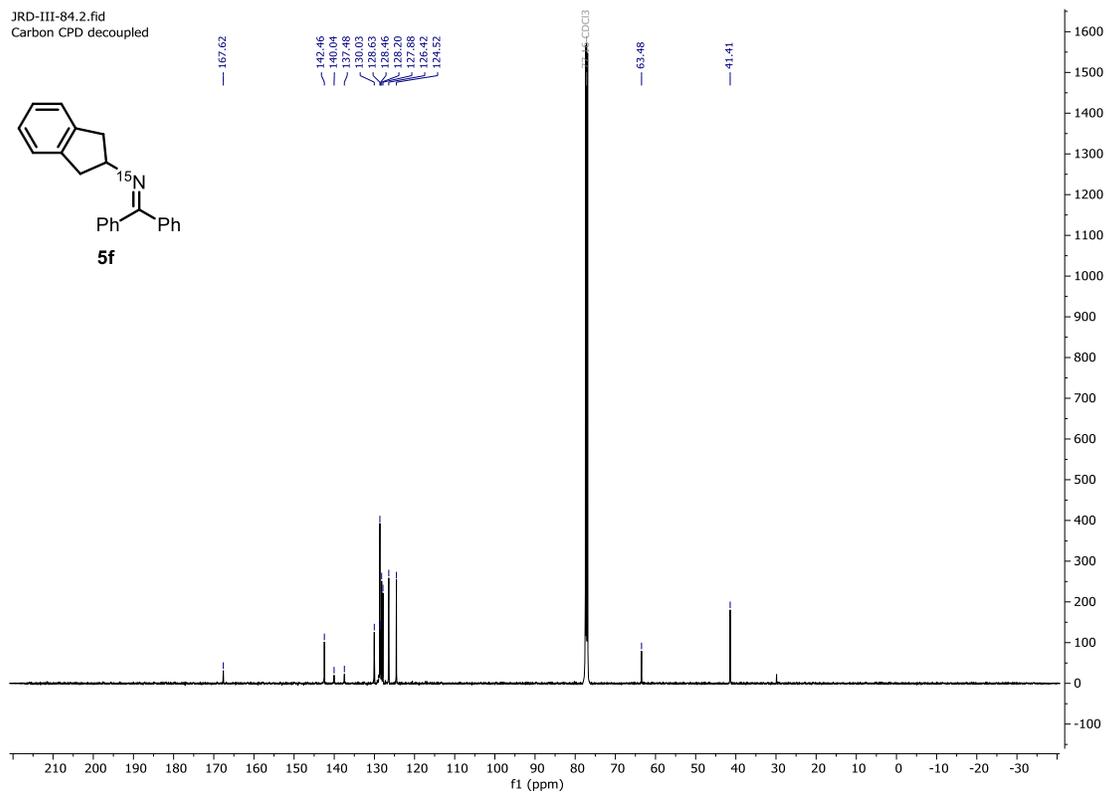
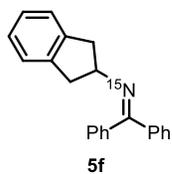
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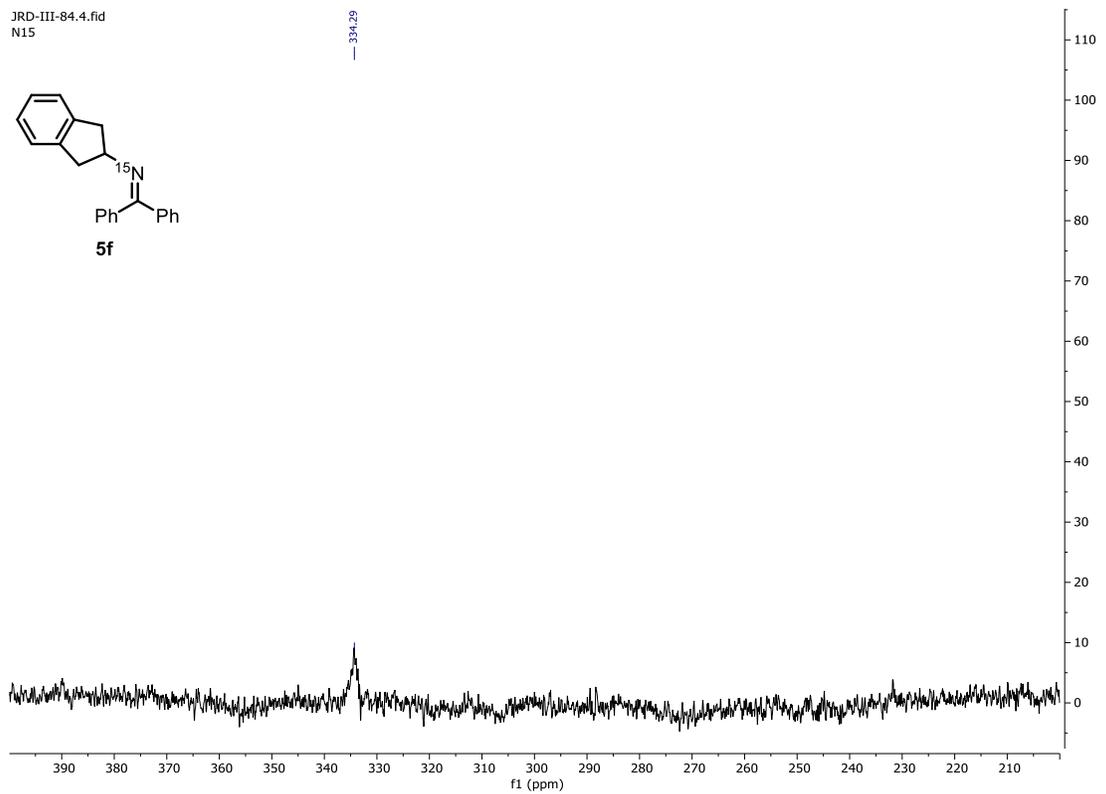
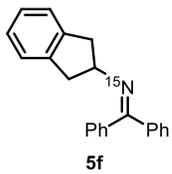
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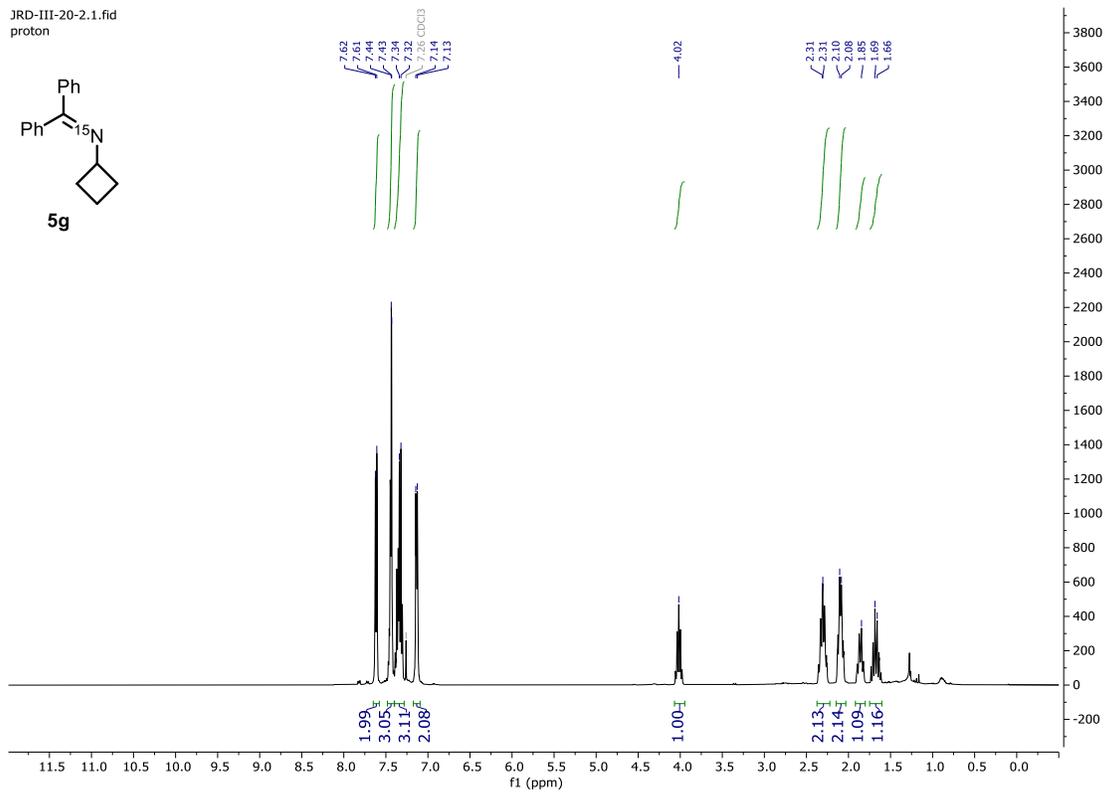
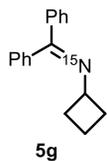
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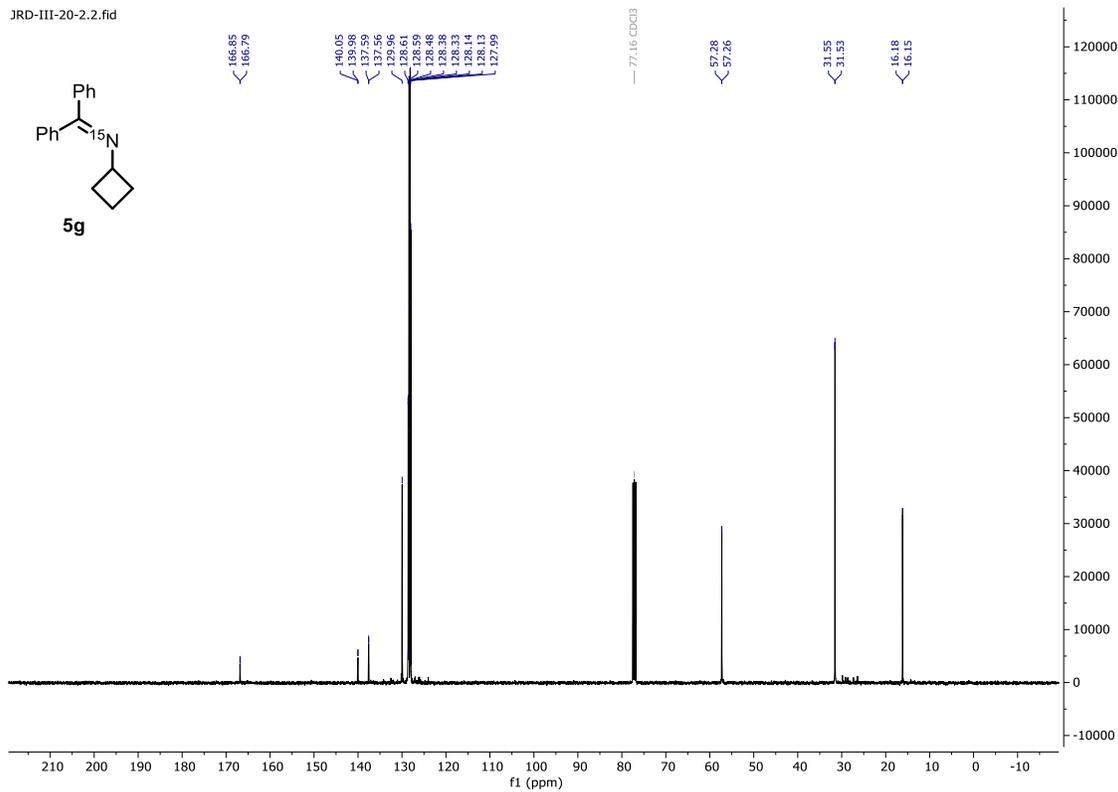
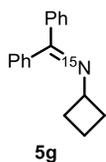
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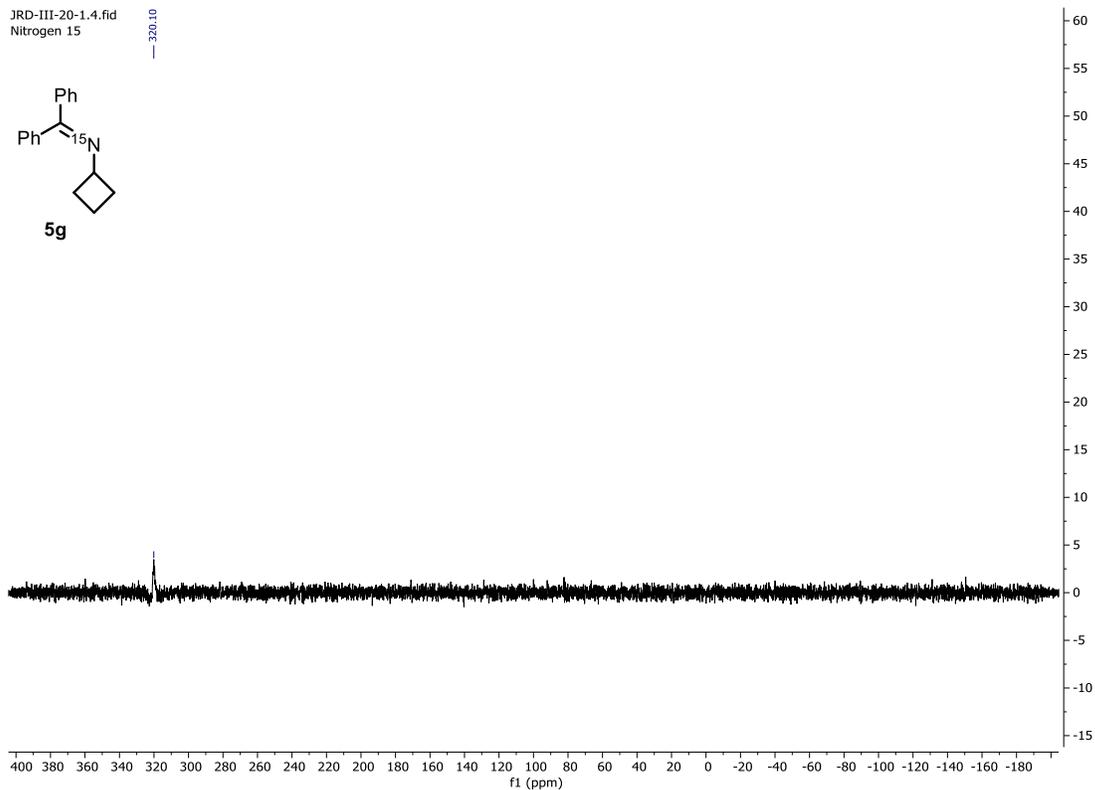
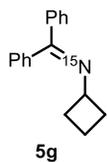
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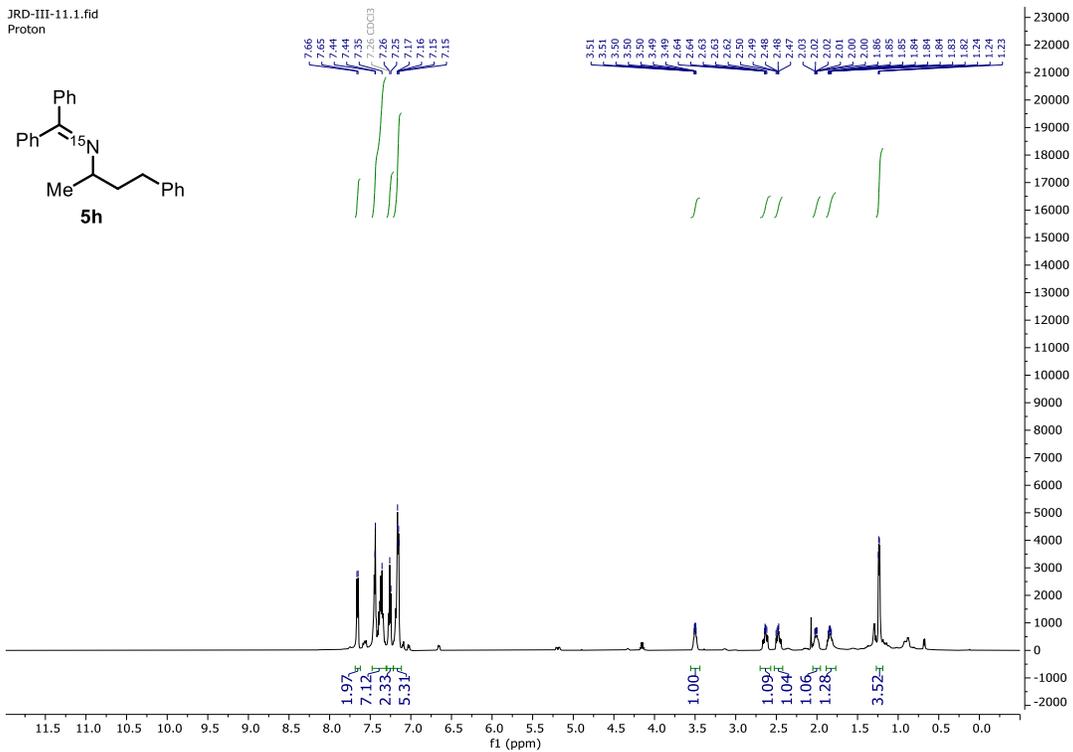
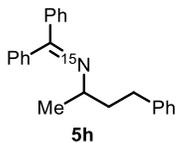
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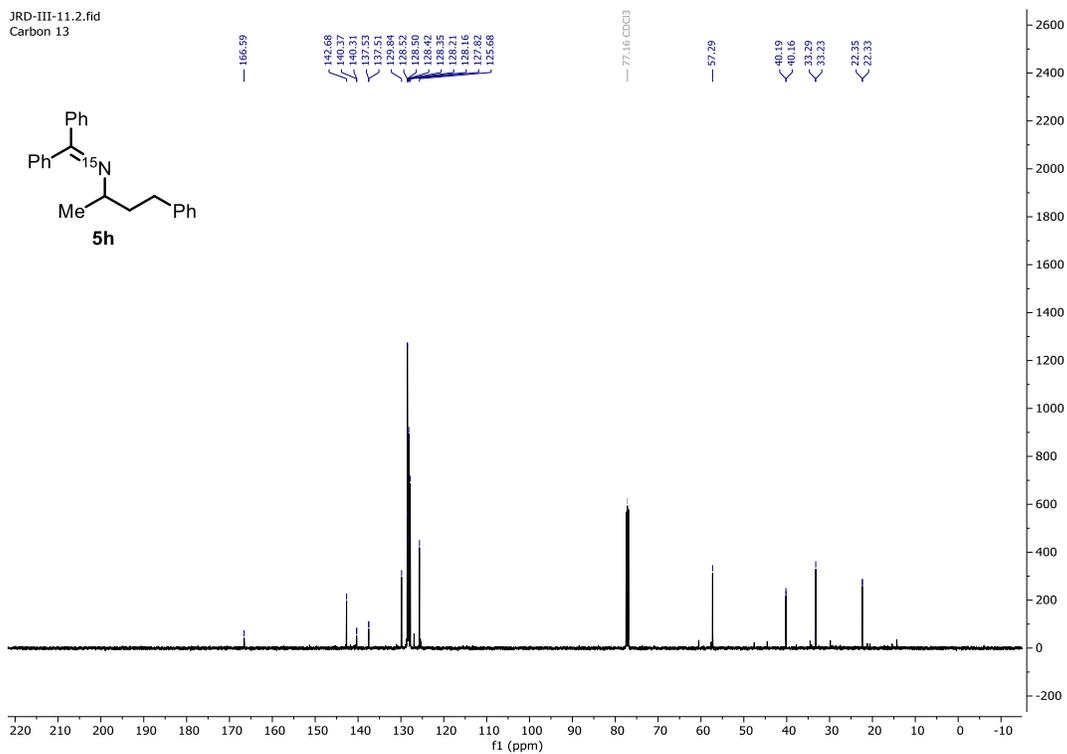
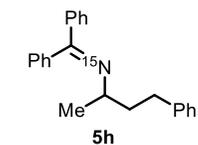
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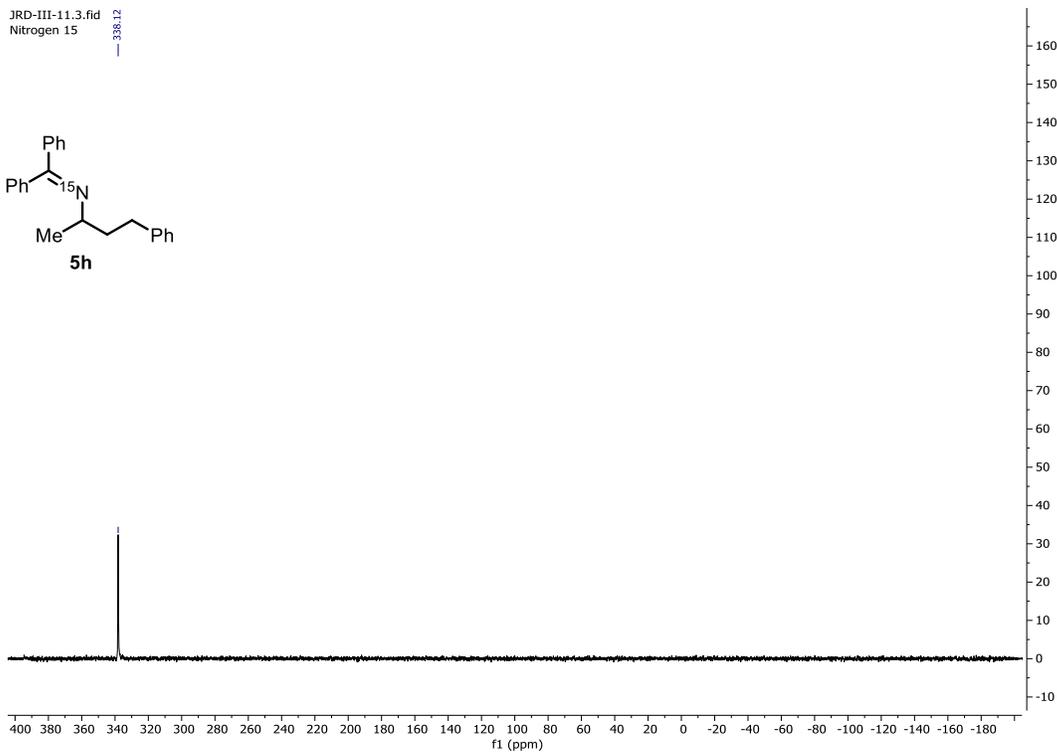
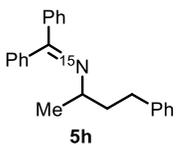
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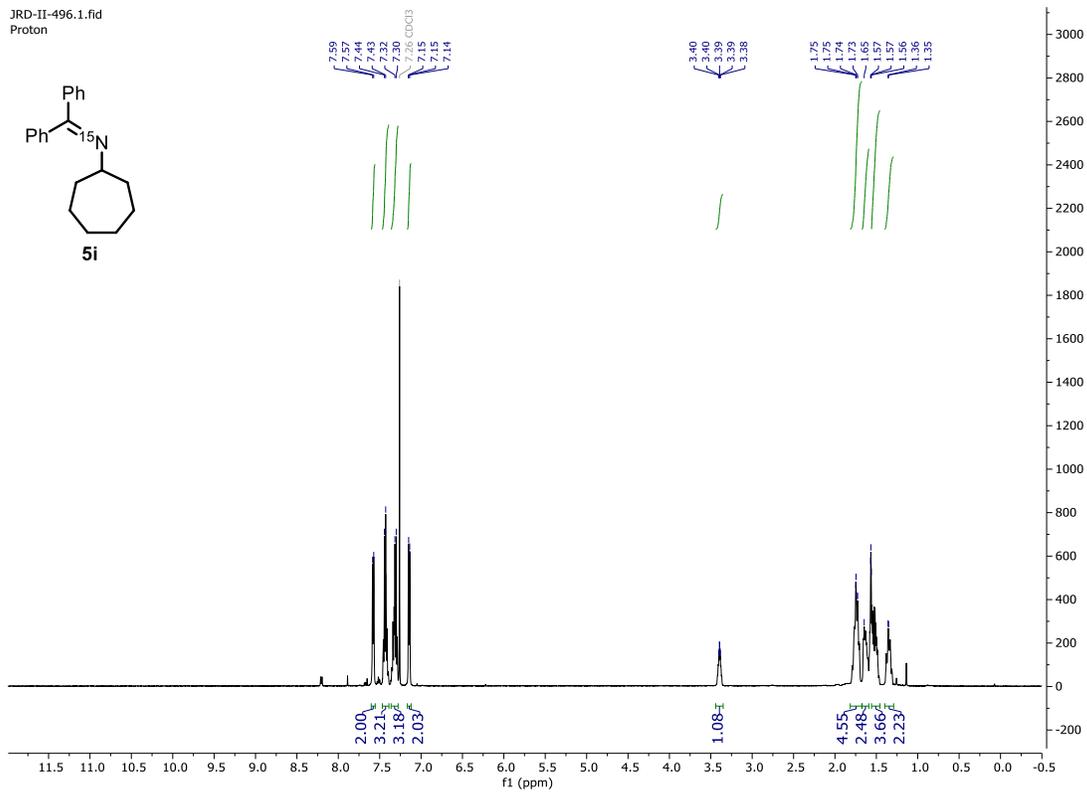
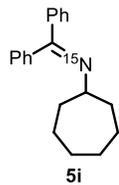
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Carbon 13



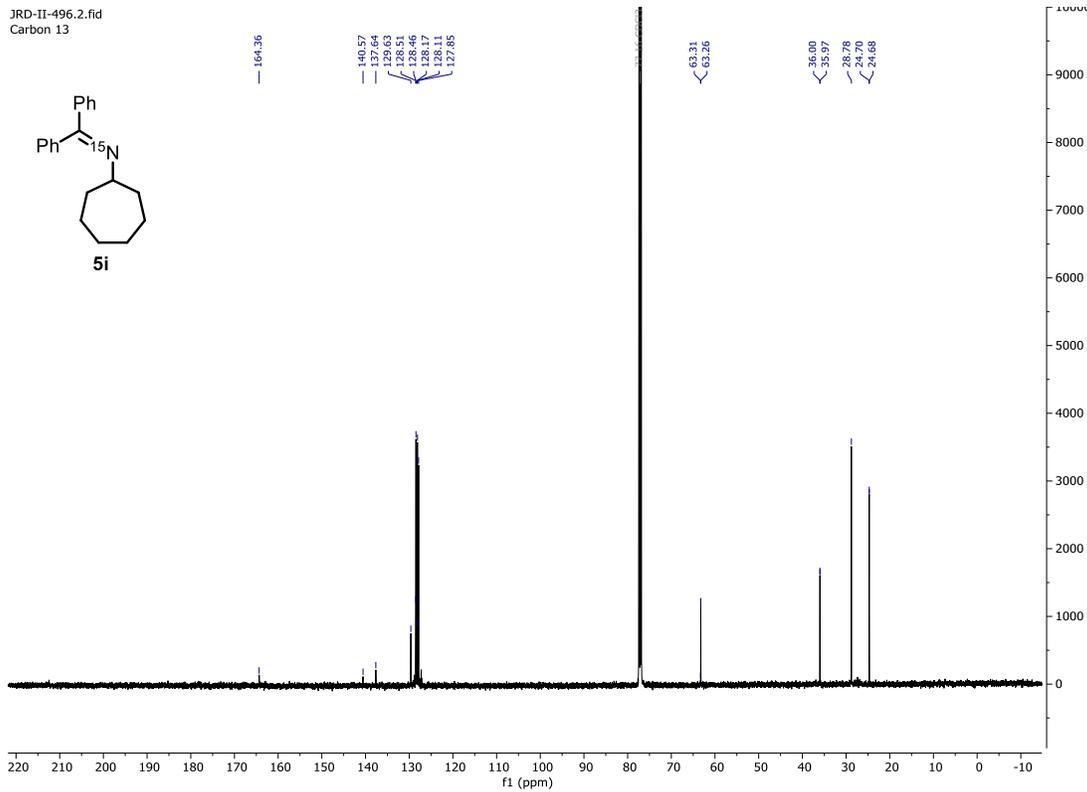
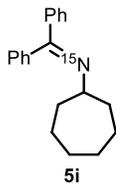
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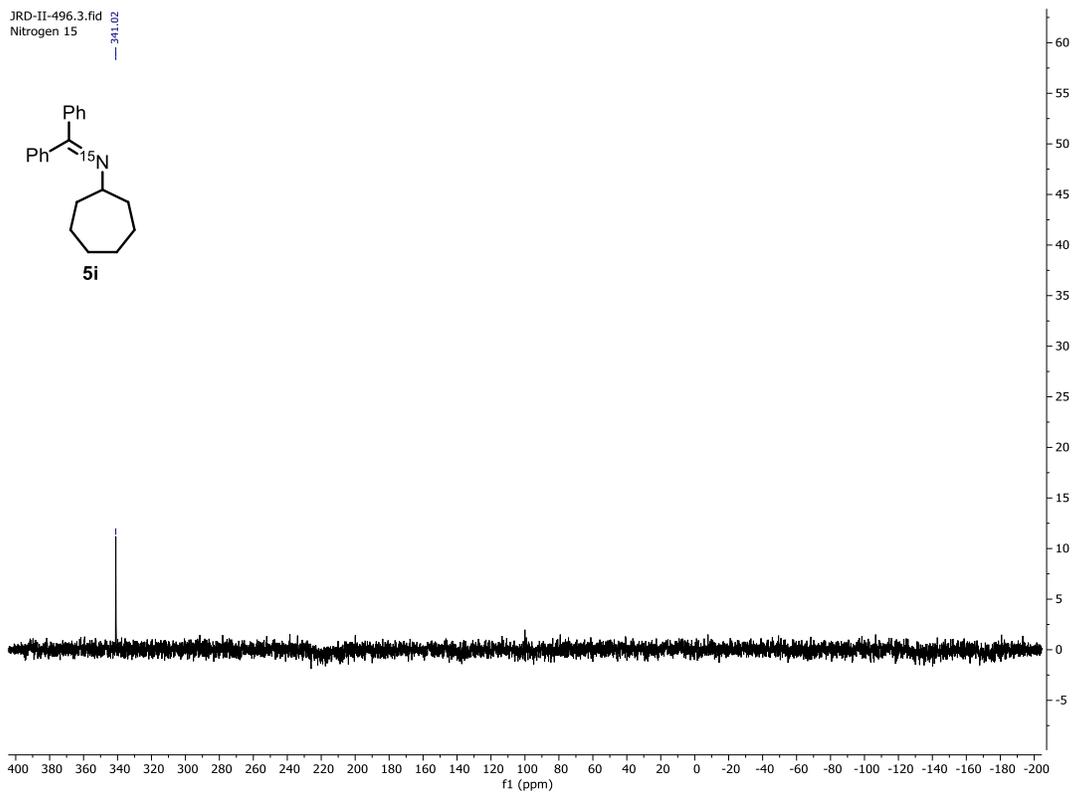
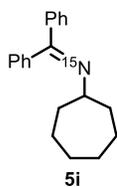
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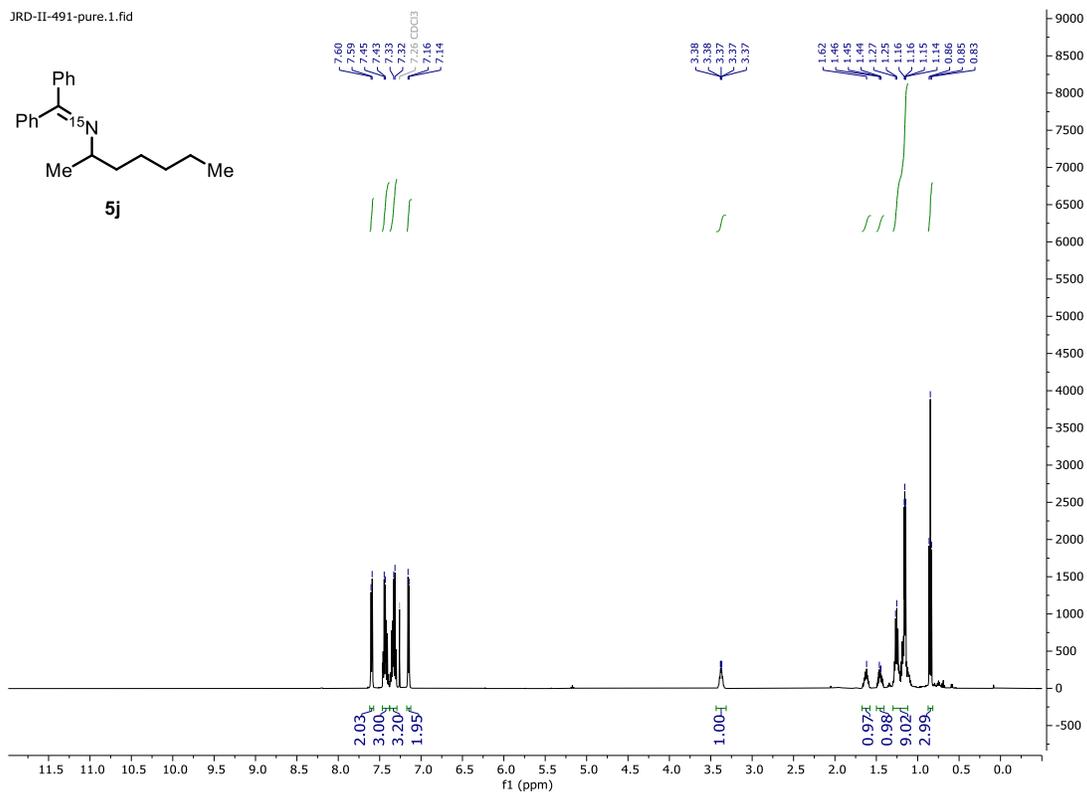
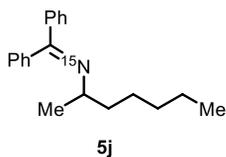
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Carbon 13



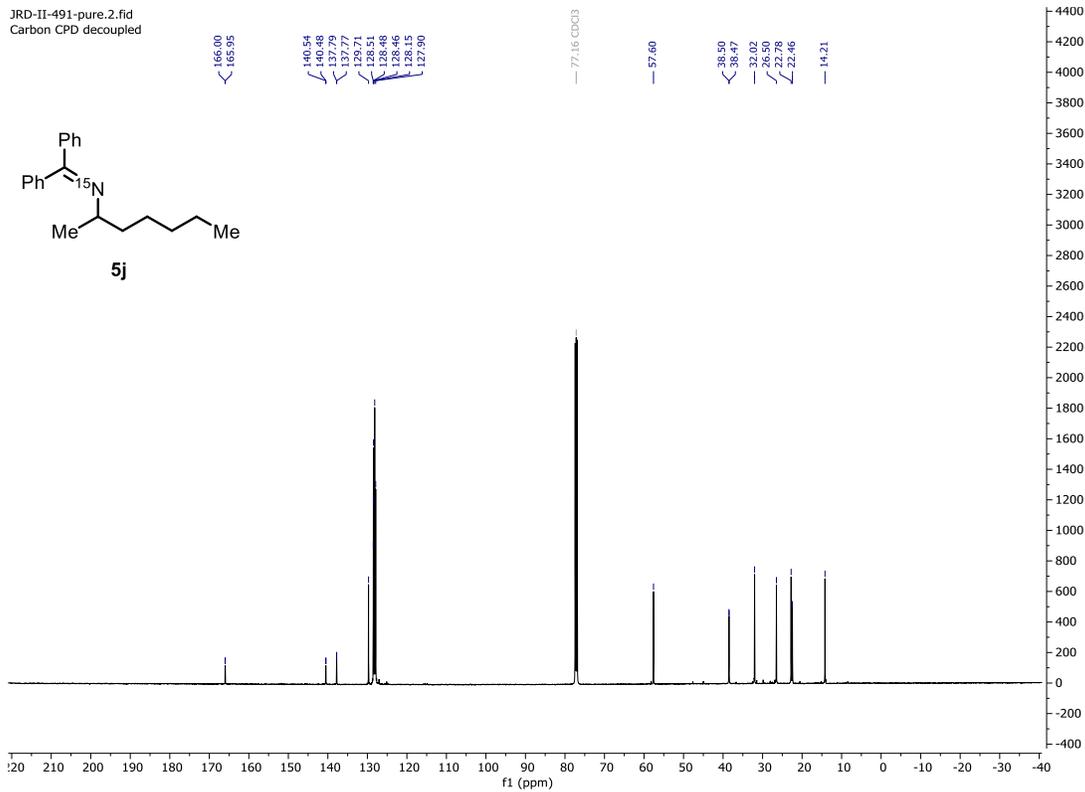
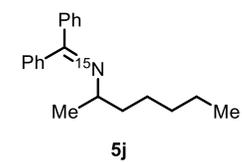
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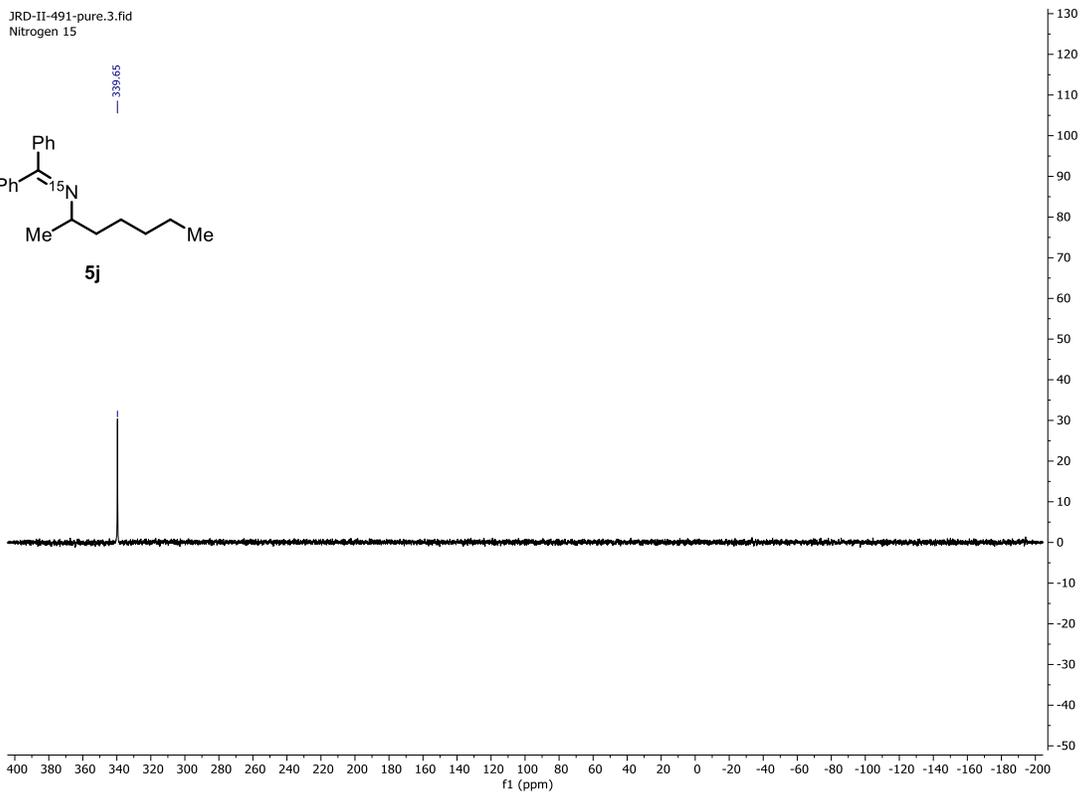
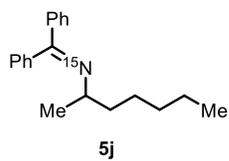
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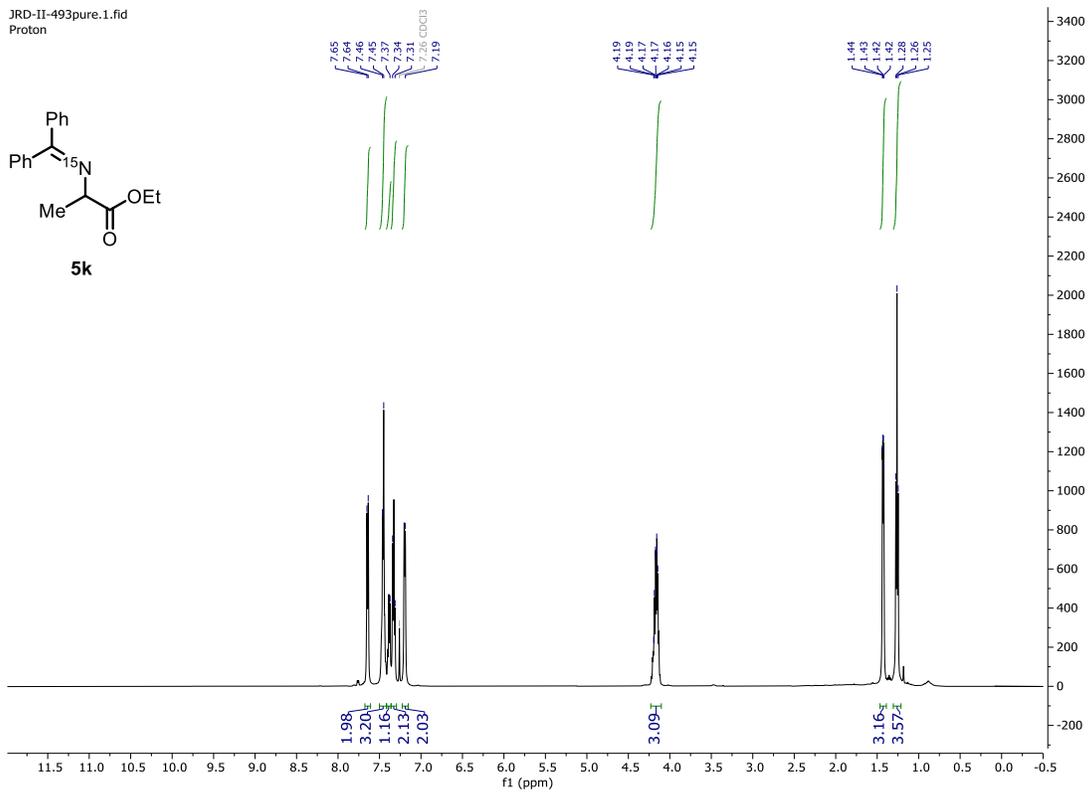
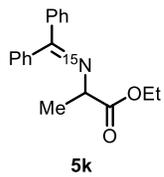
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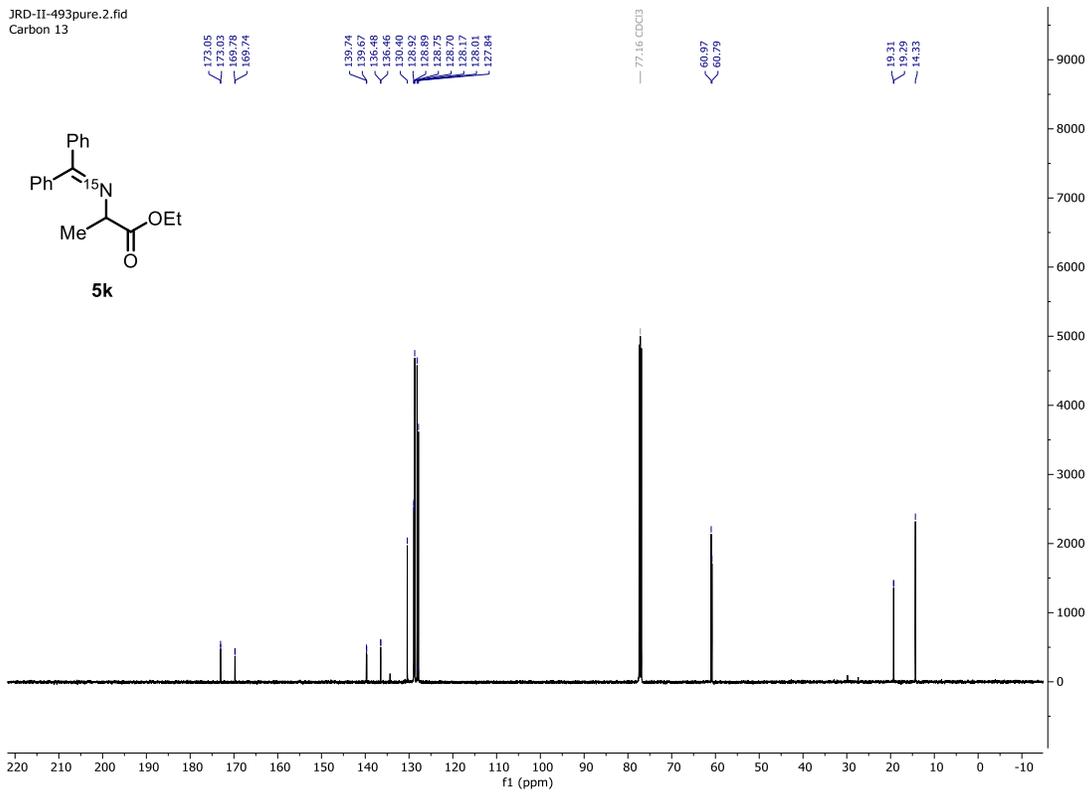
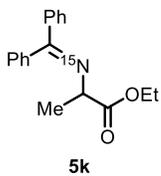
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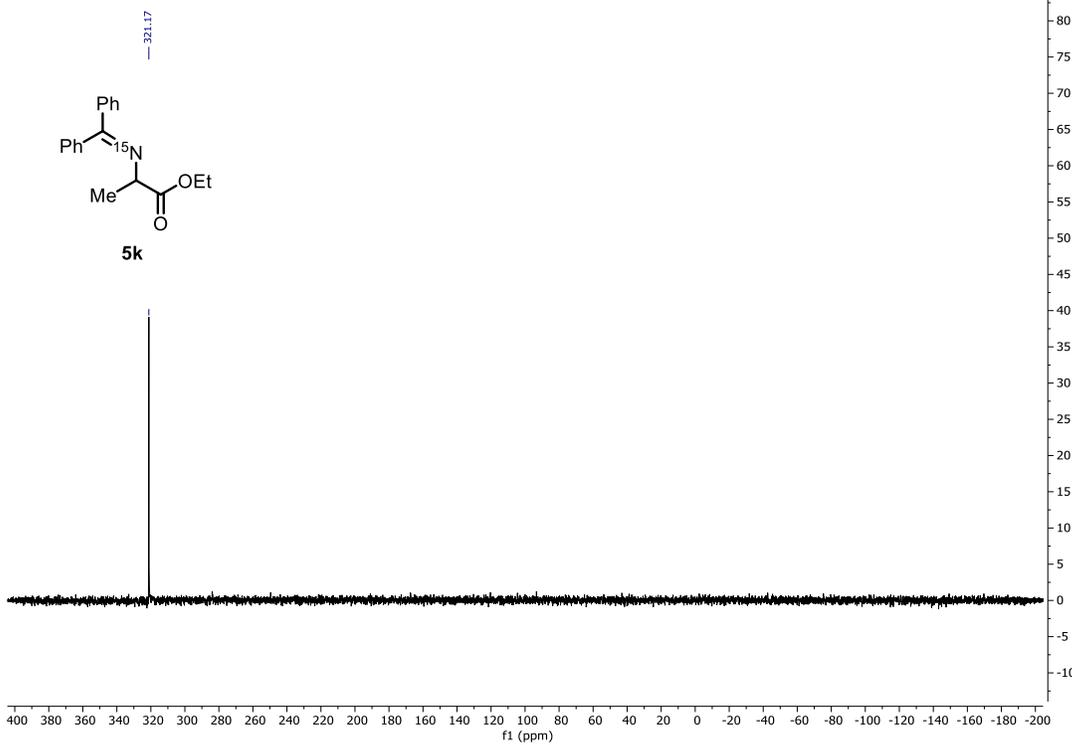
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Proton



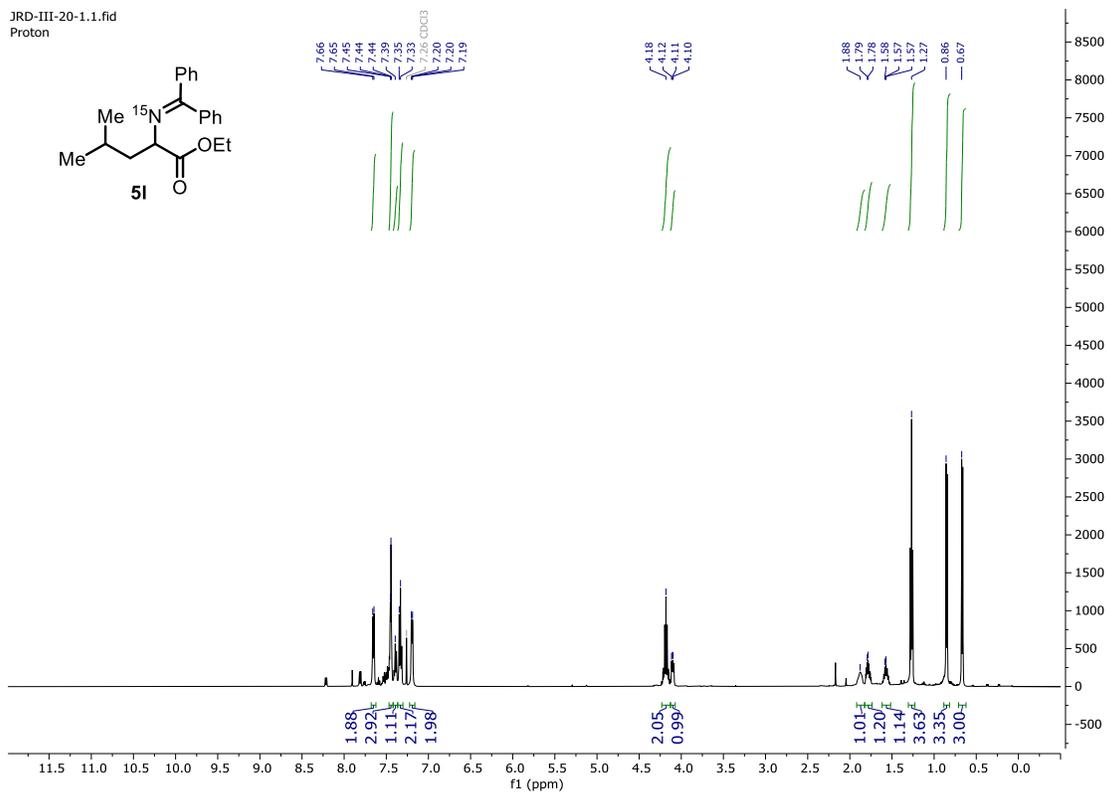
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Carbon 13



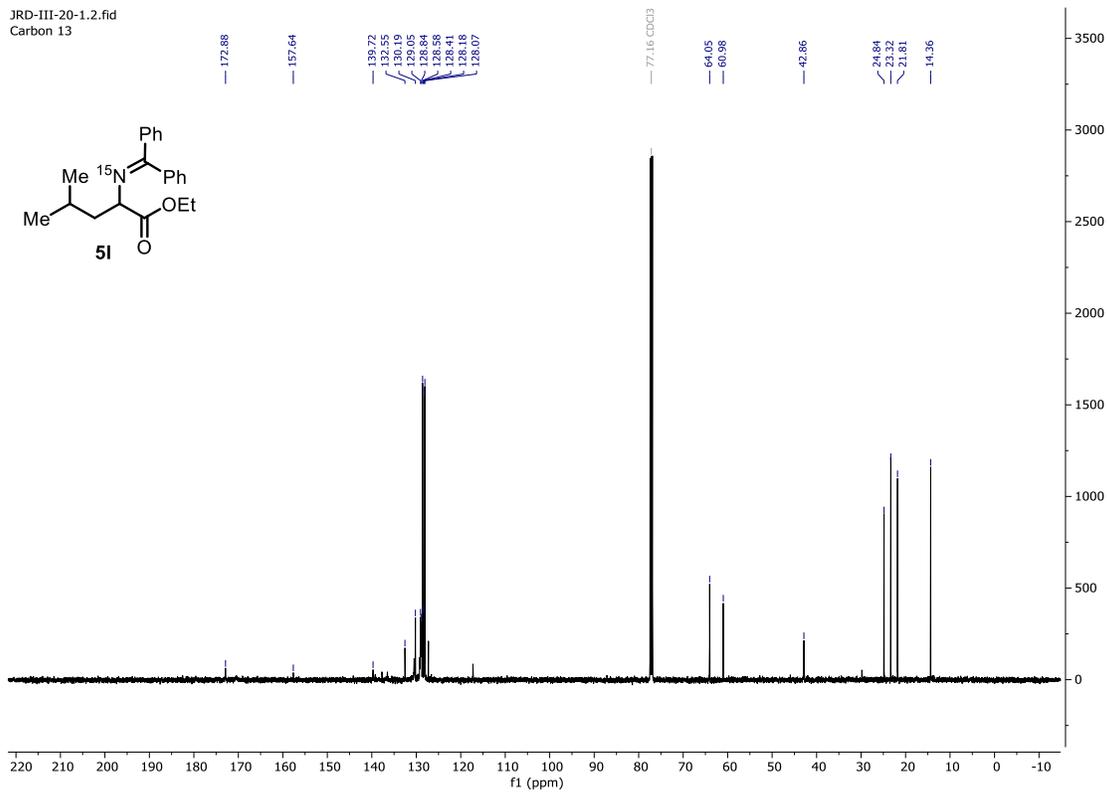
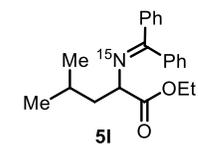
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Nitrogen 15



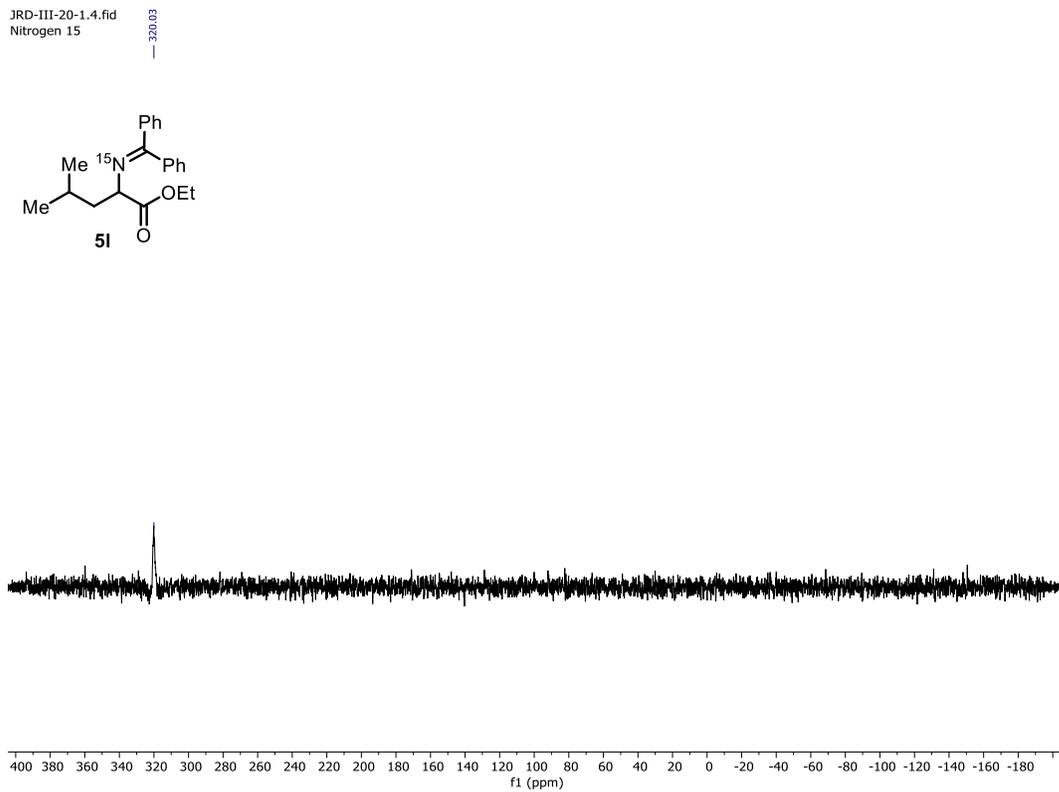
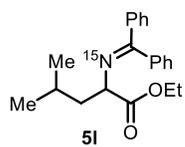
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Proton

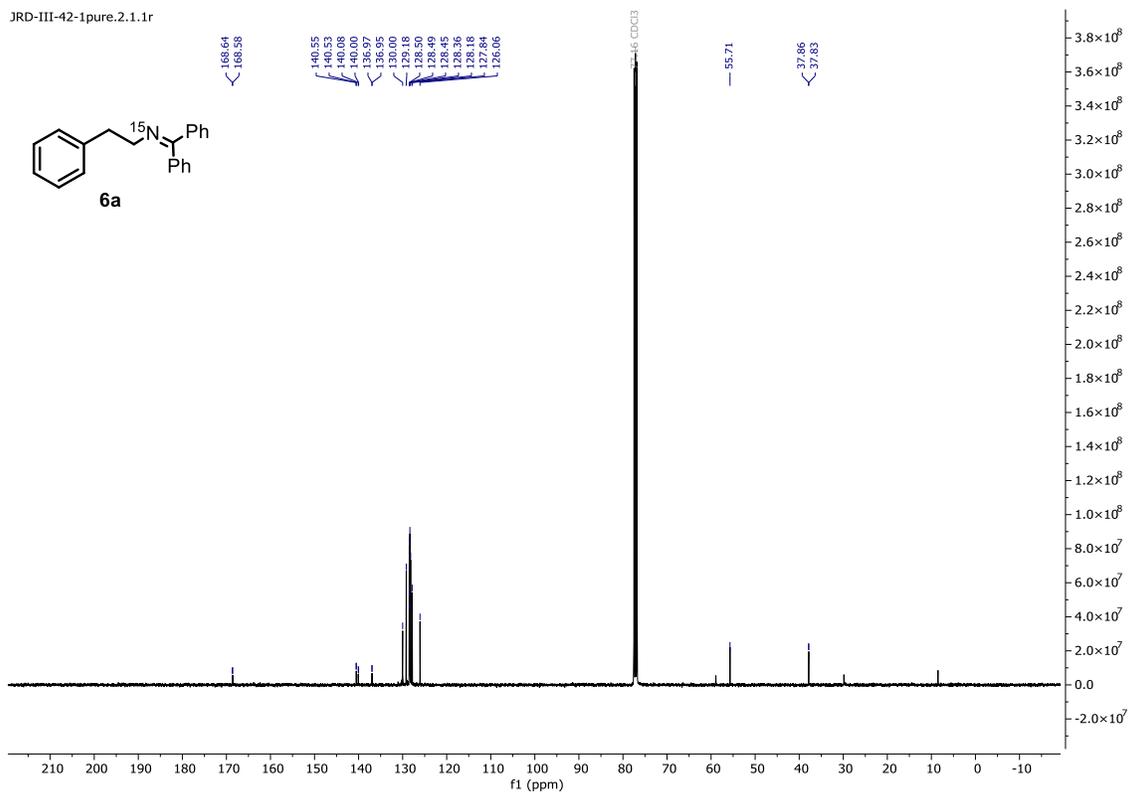
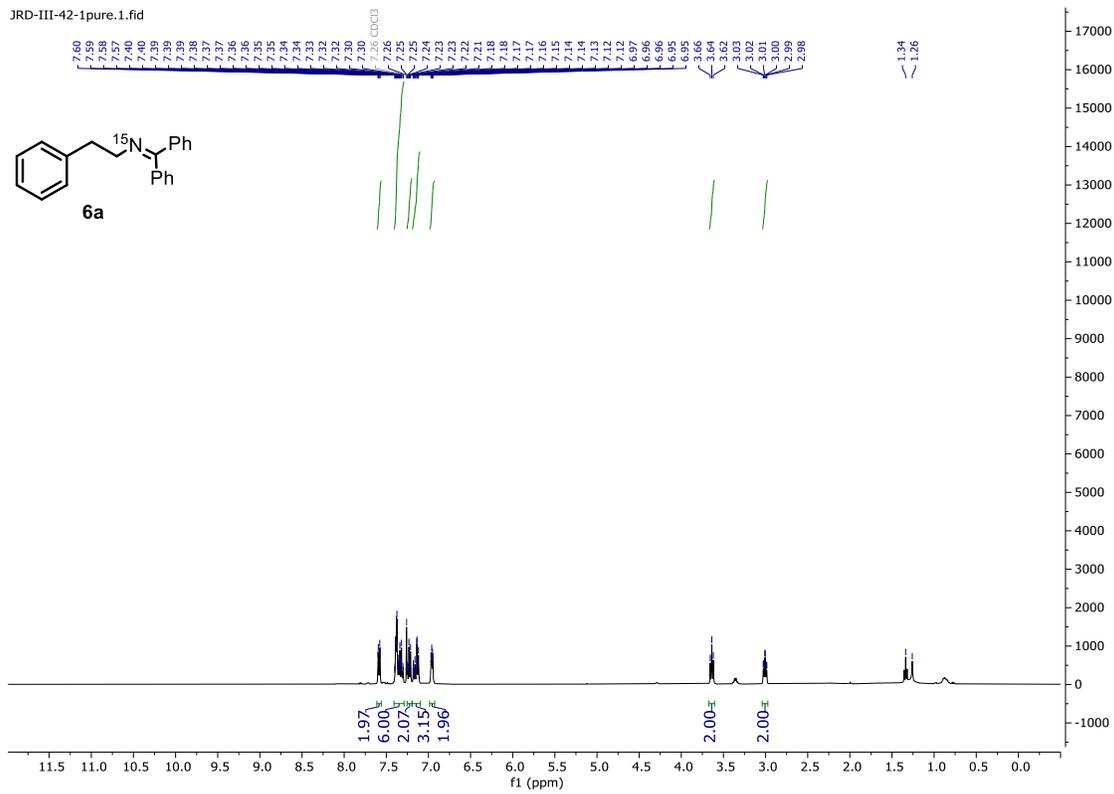


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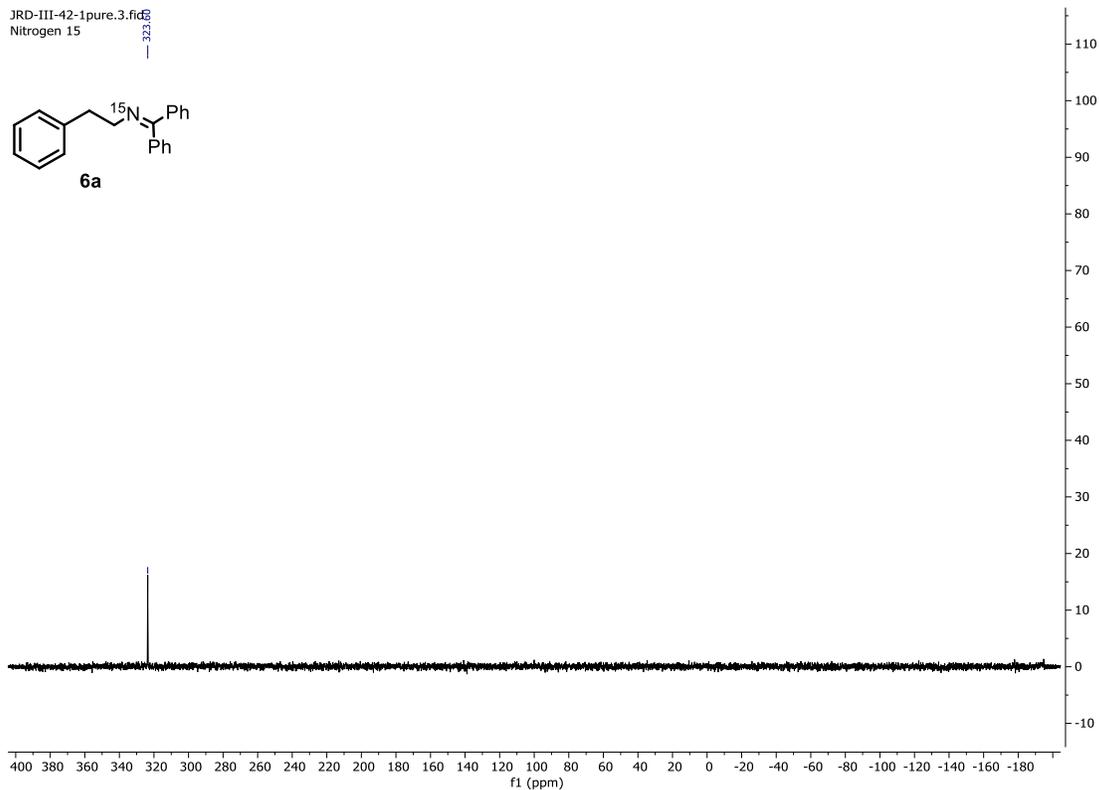
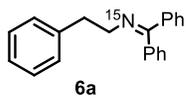


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Nitrogen 15

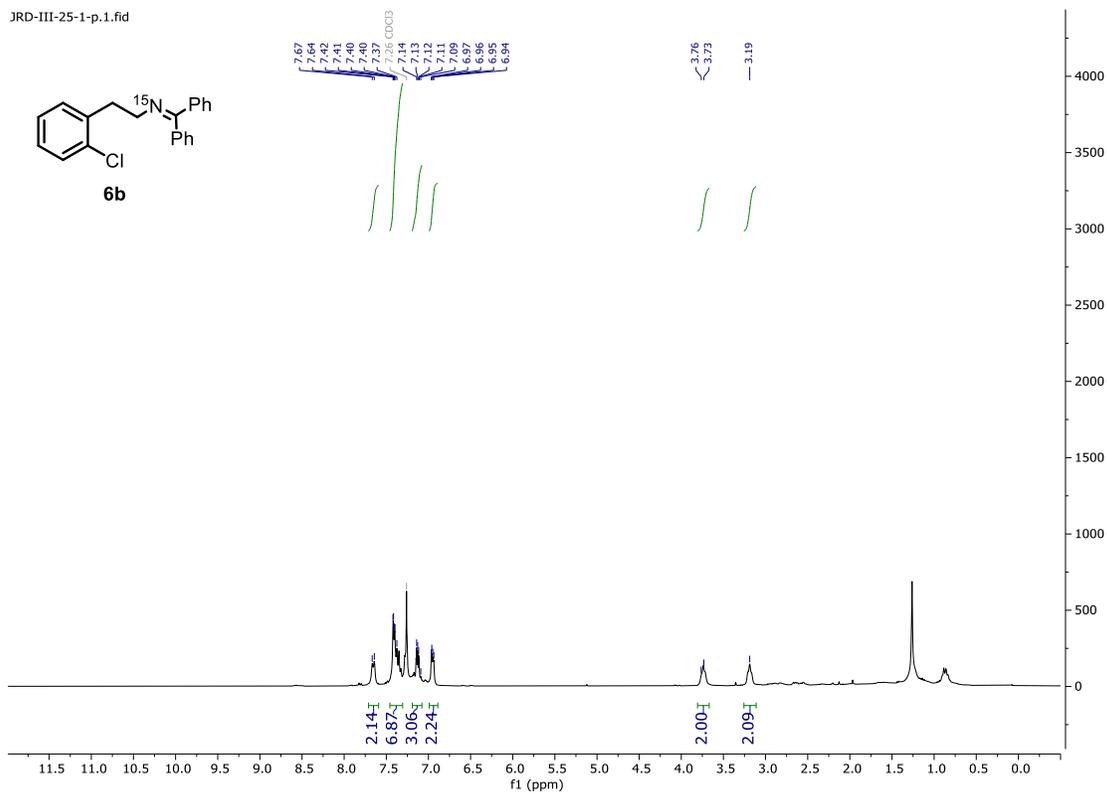
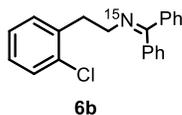




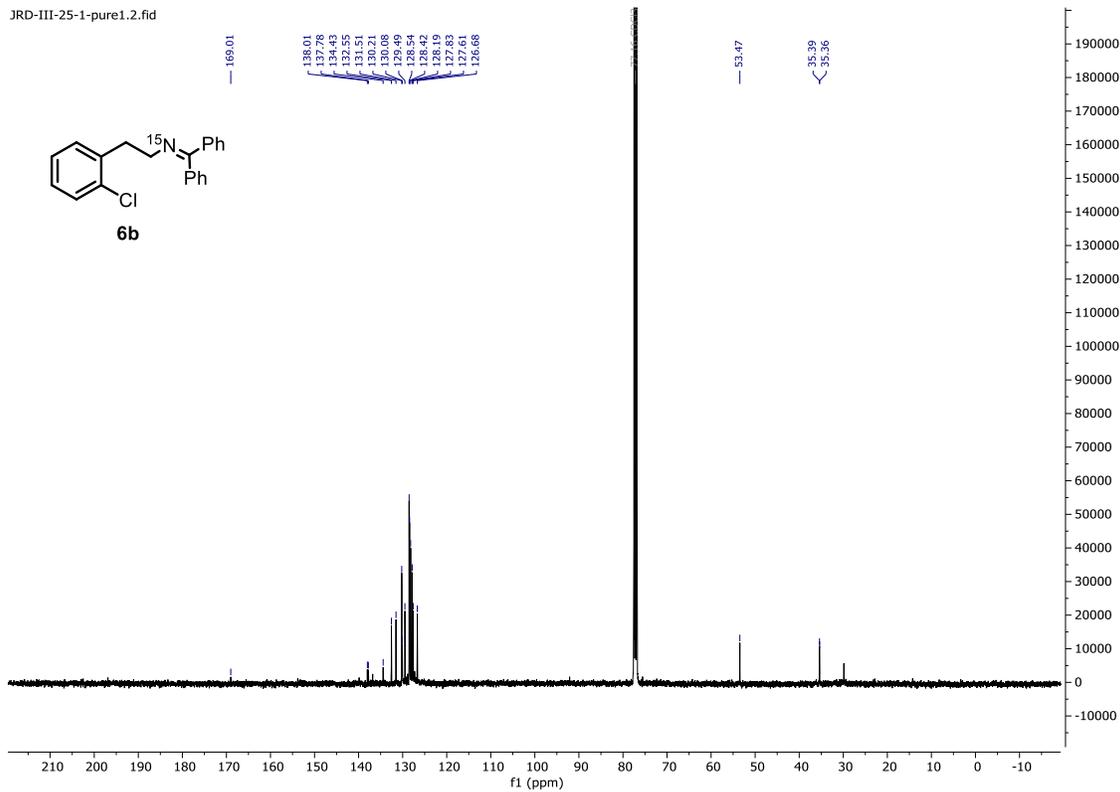
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Nitrogen 15



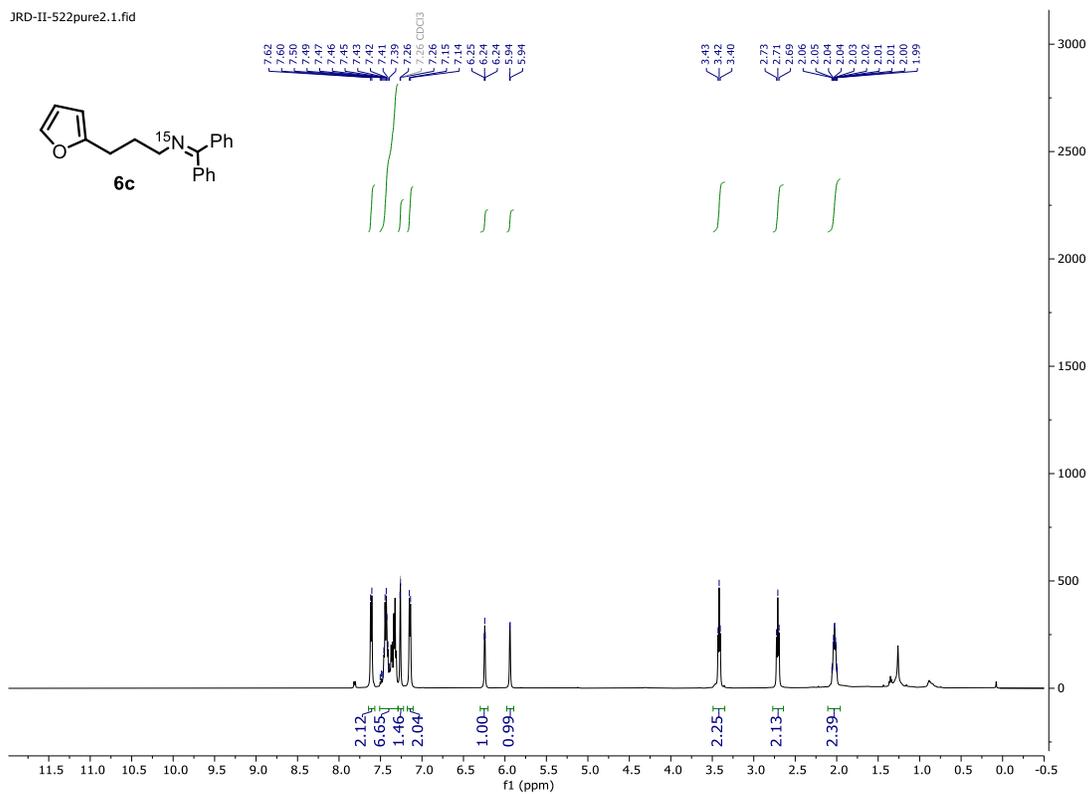
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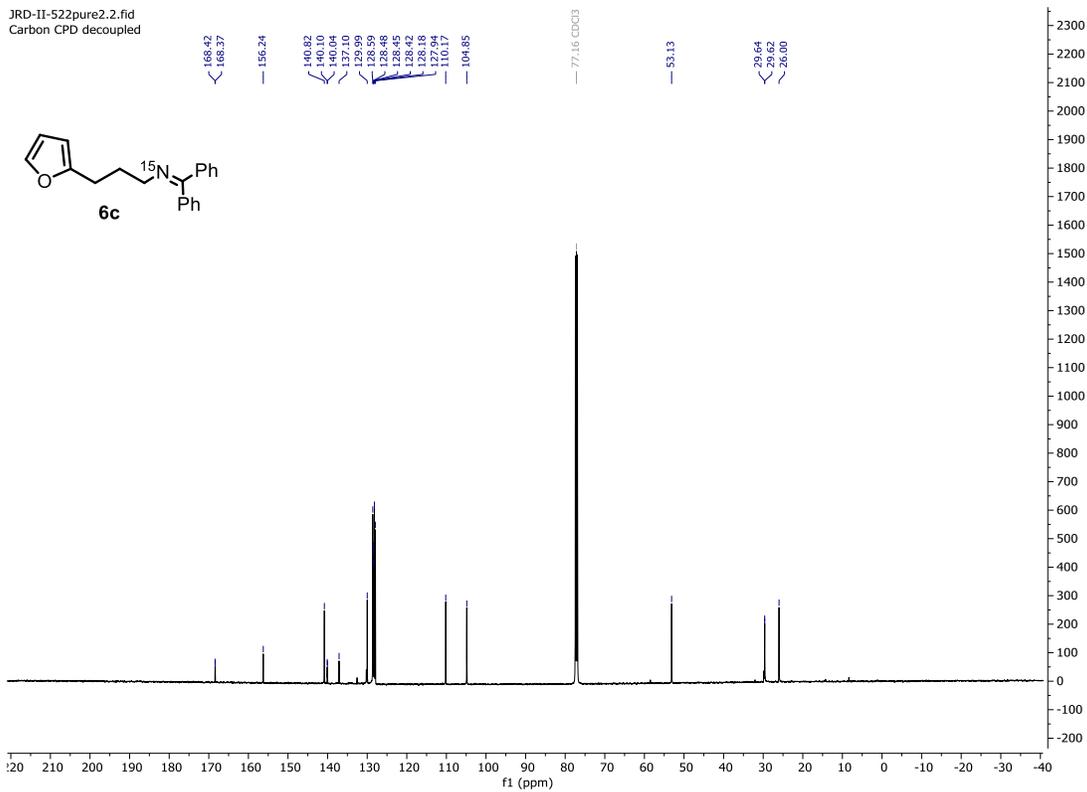
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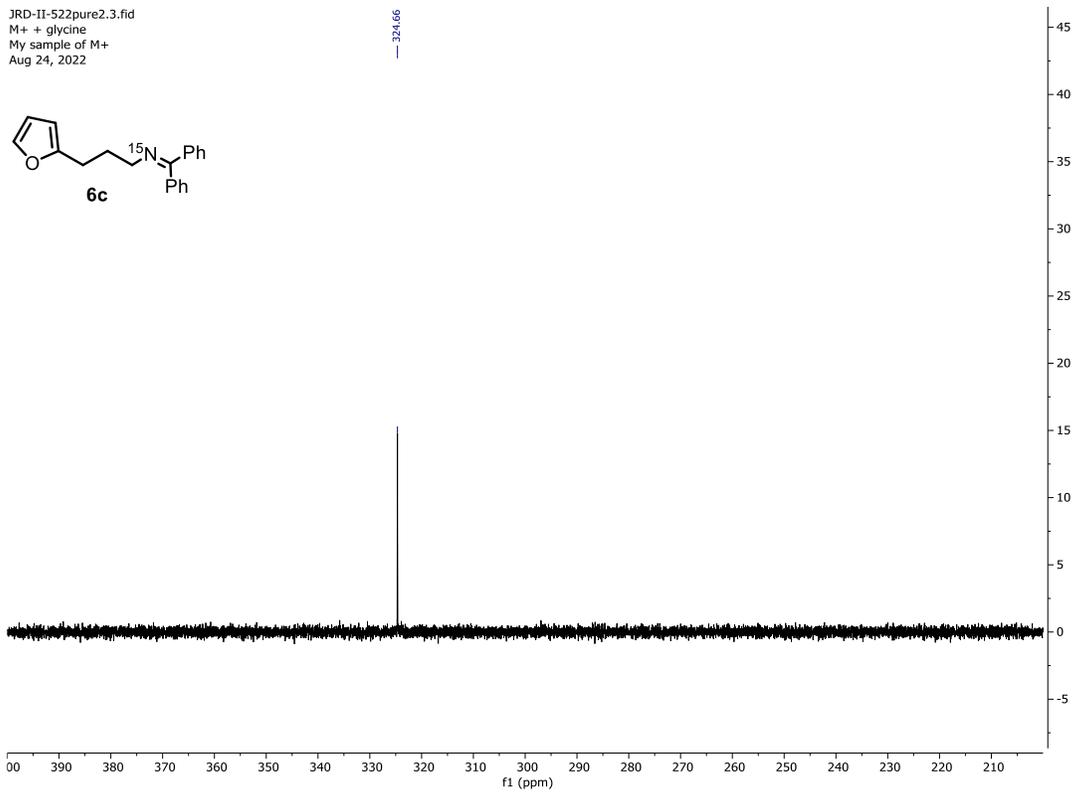
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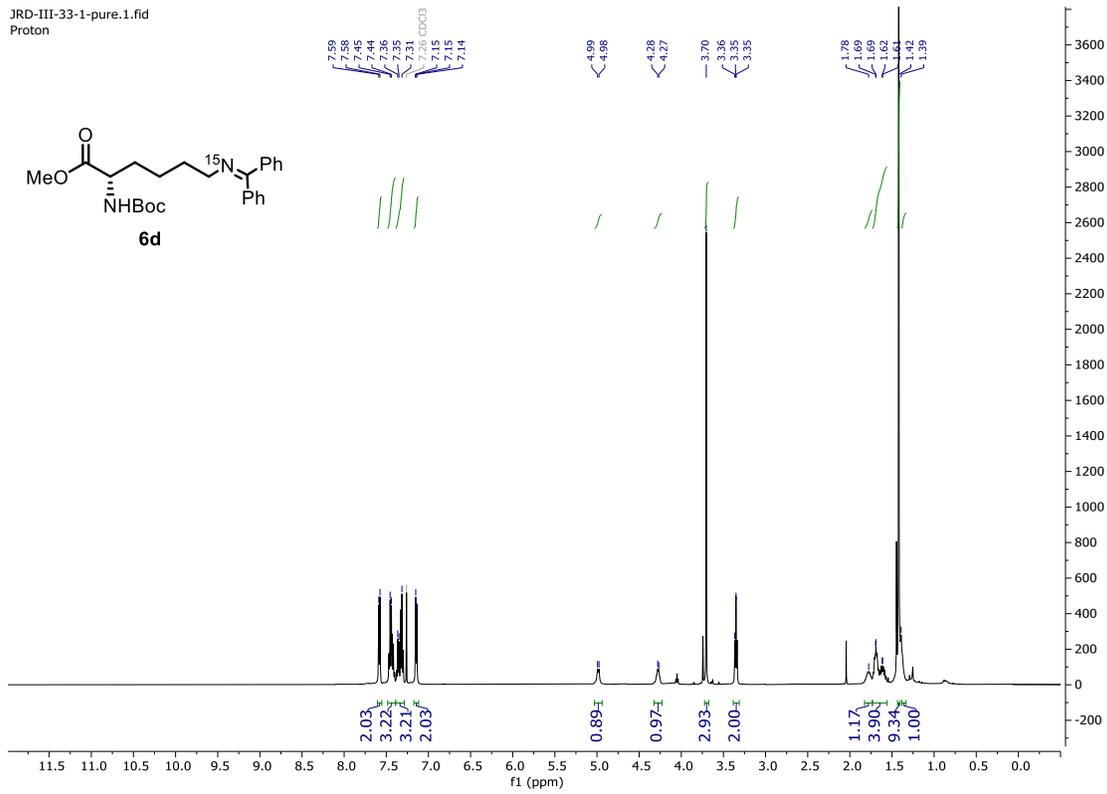
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Carbon CPD decoupled



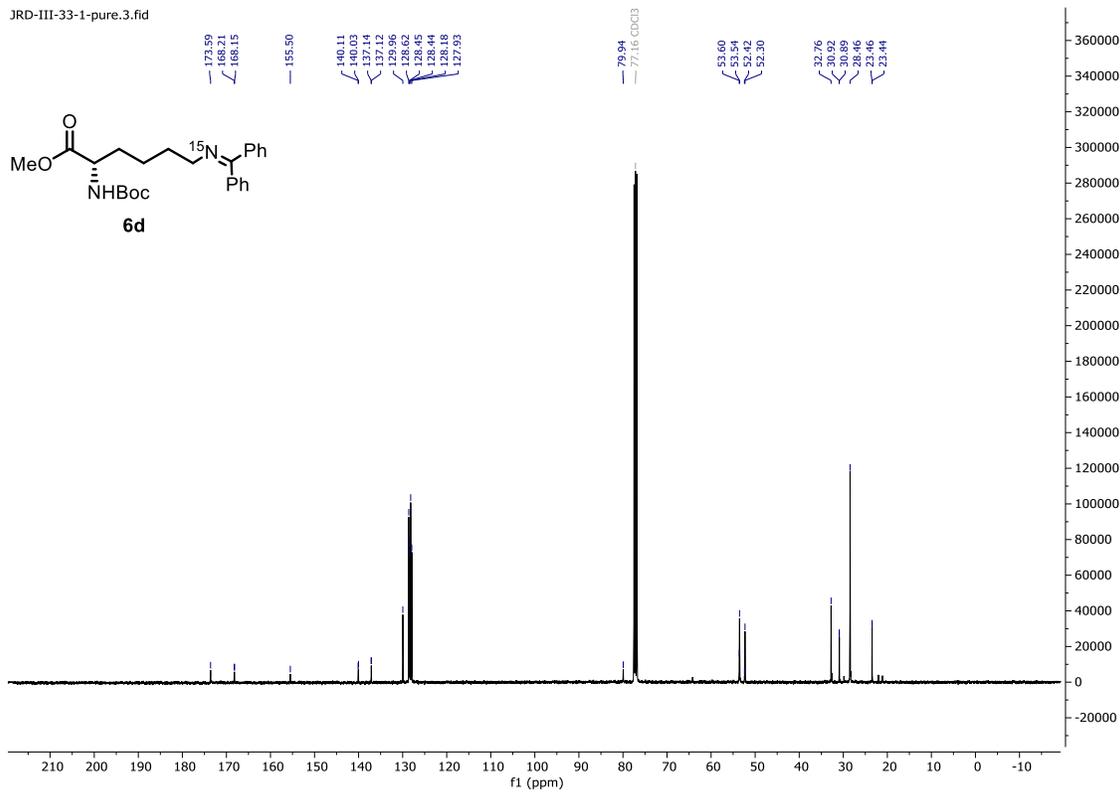
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M+ + glycine
My sample of M+
Aug 24, 2022



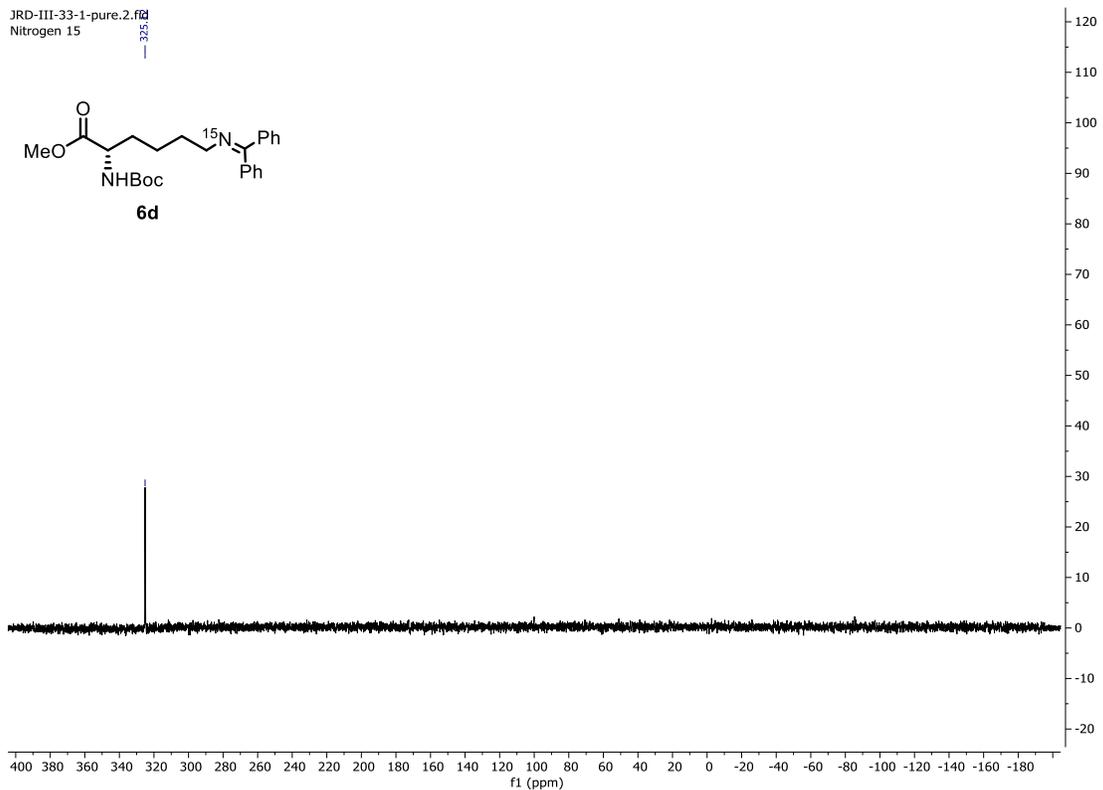
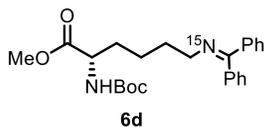
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Proton



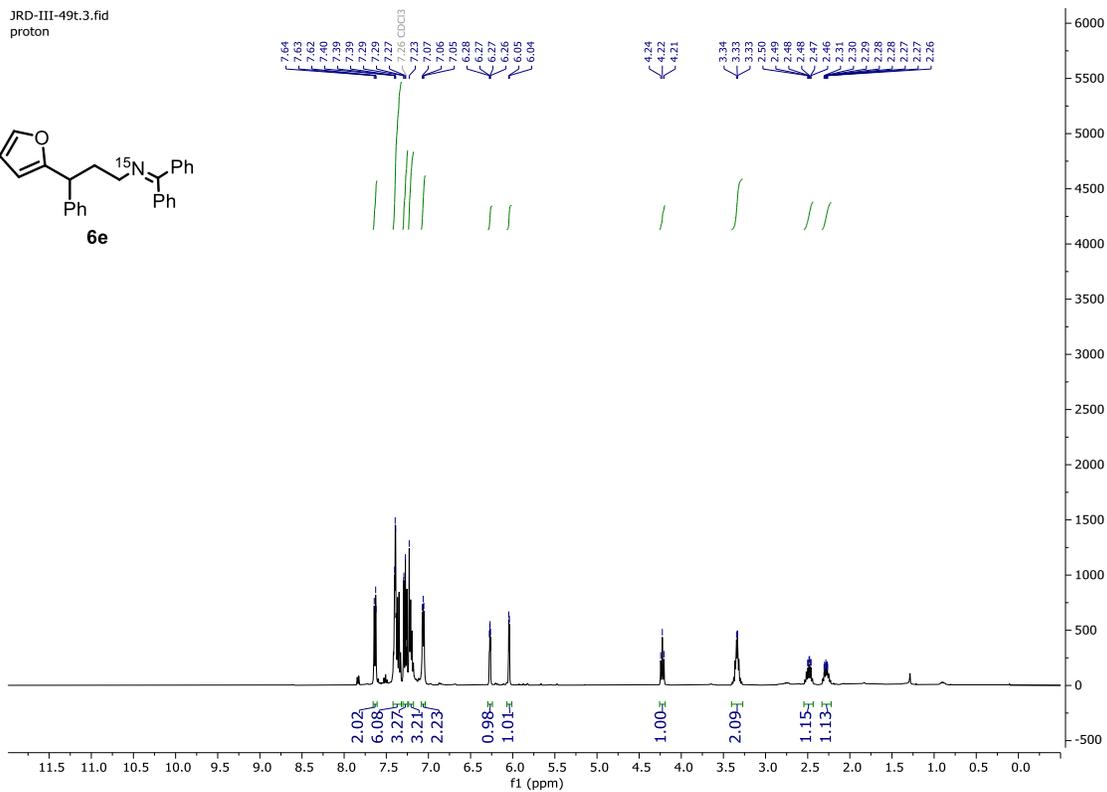
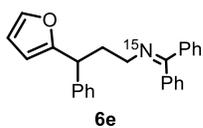
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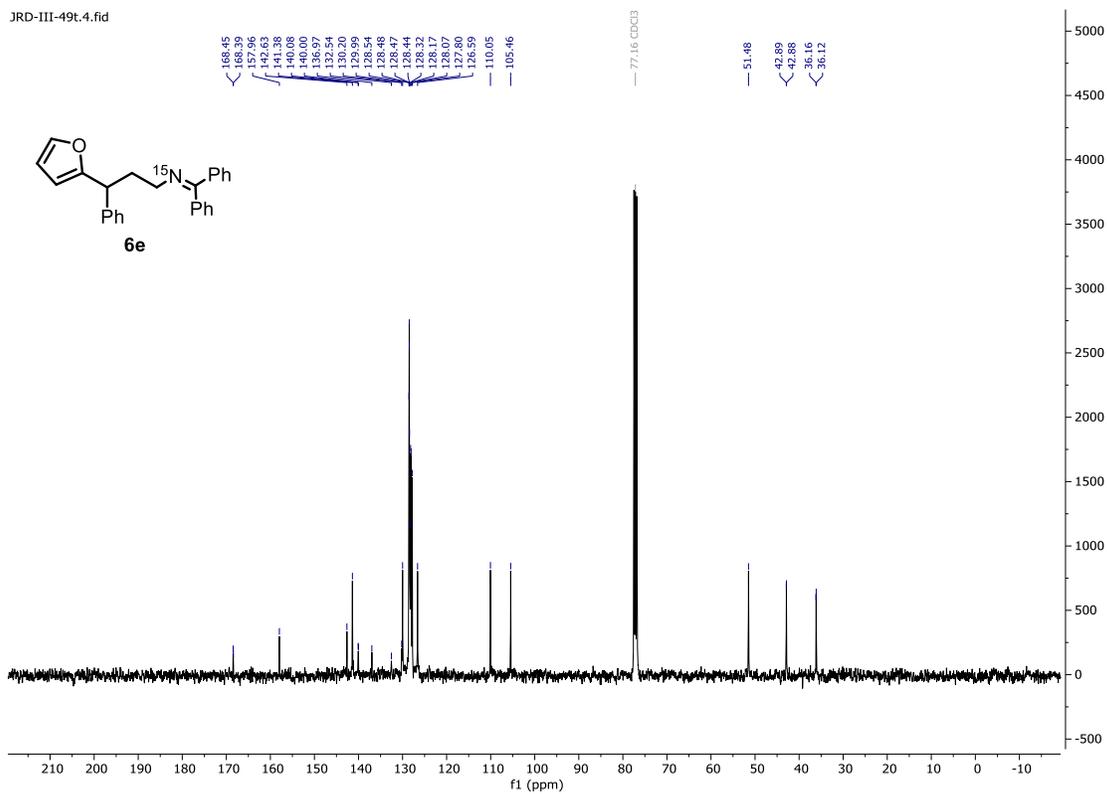
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Nitrogen 15



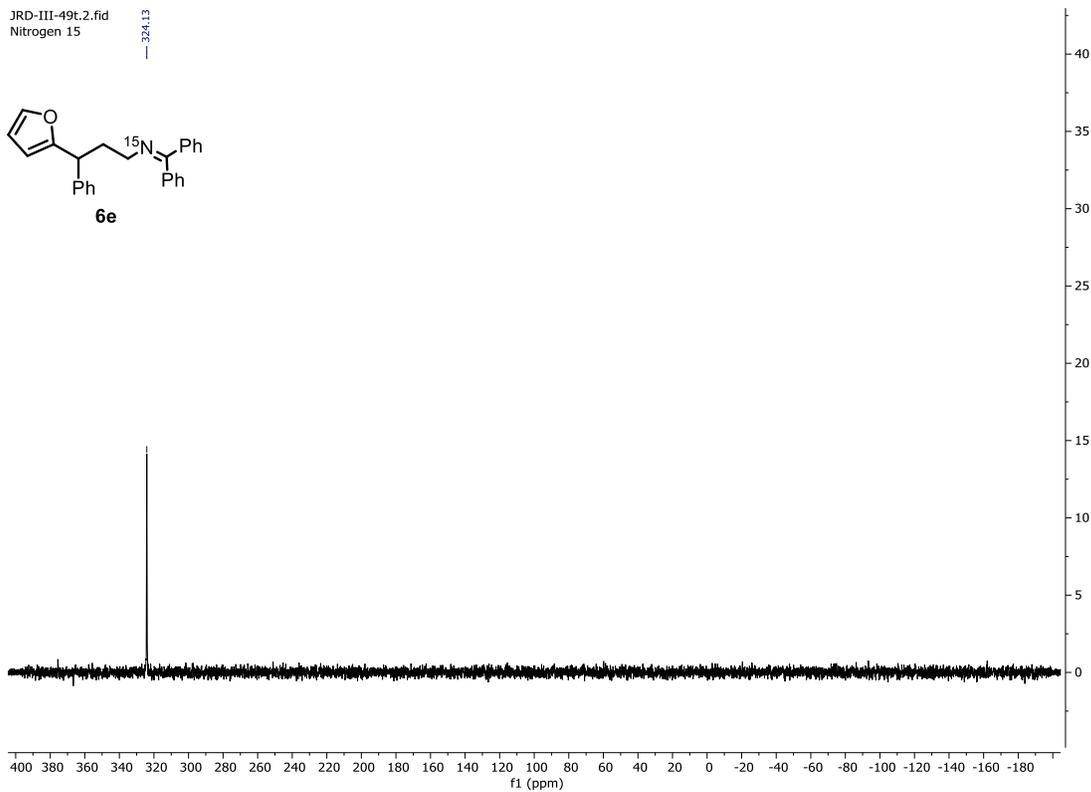
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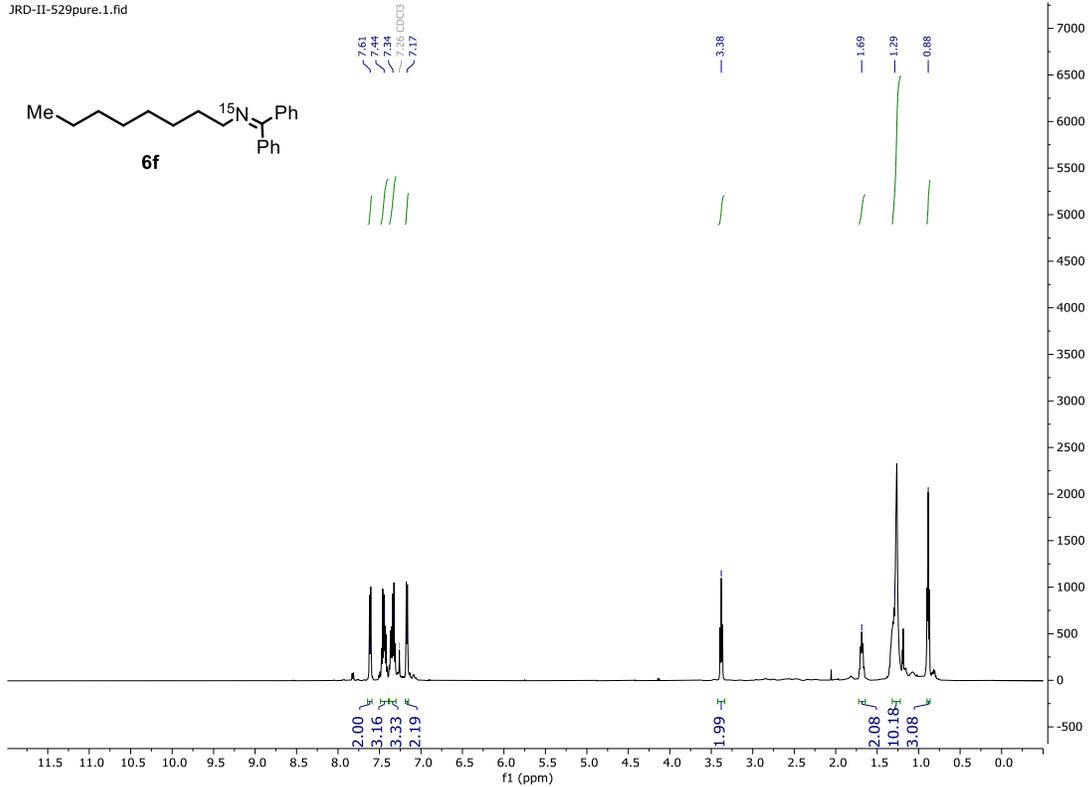
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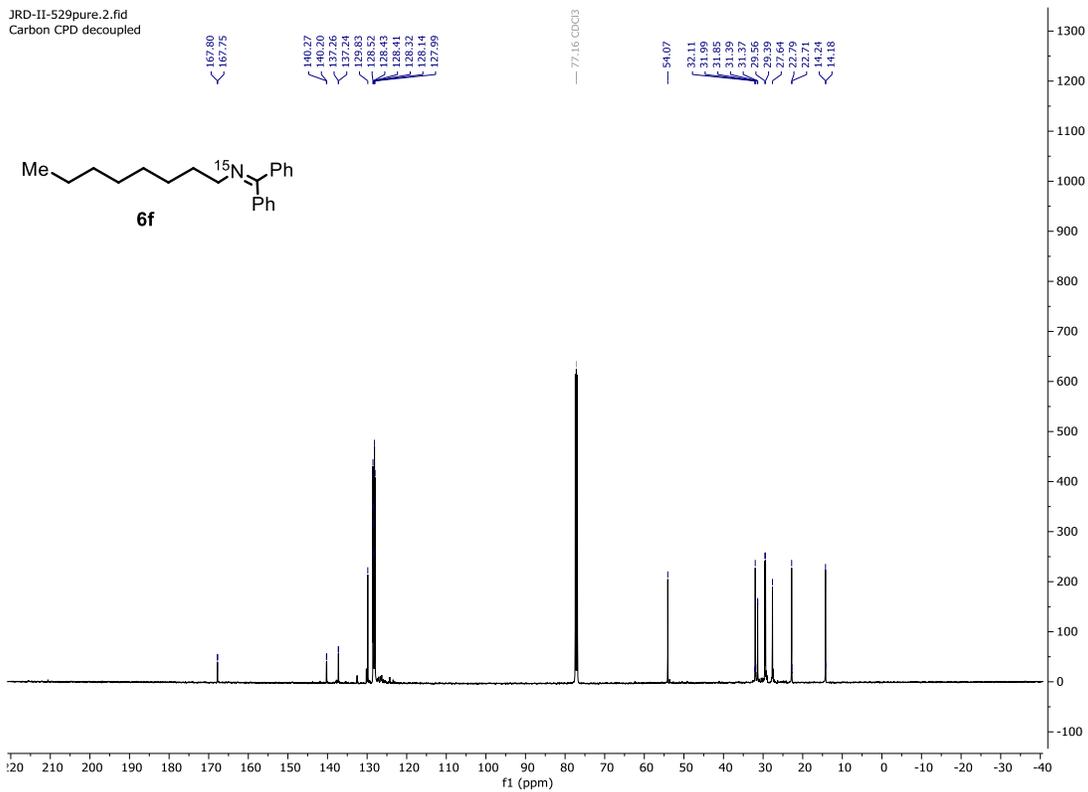
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Nitrogen 15

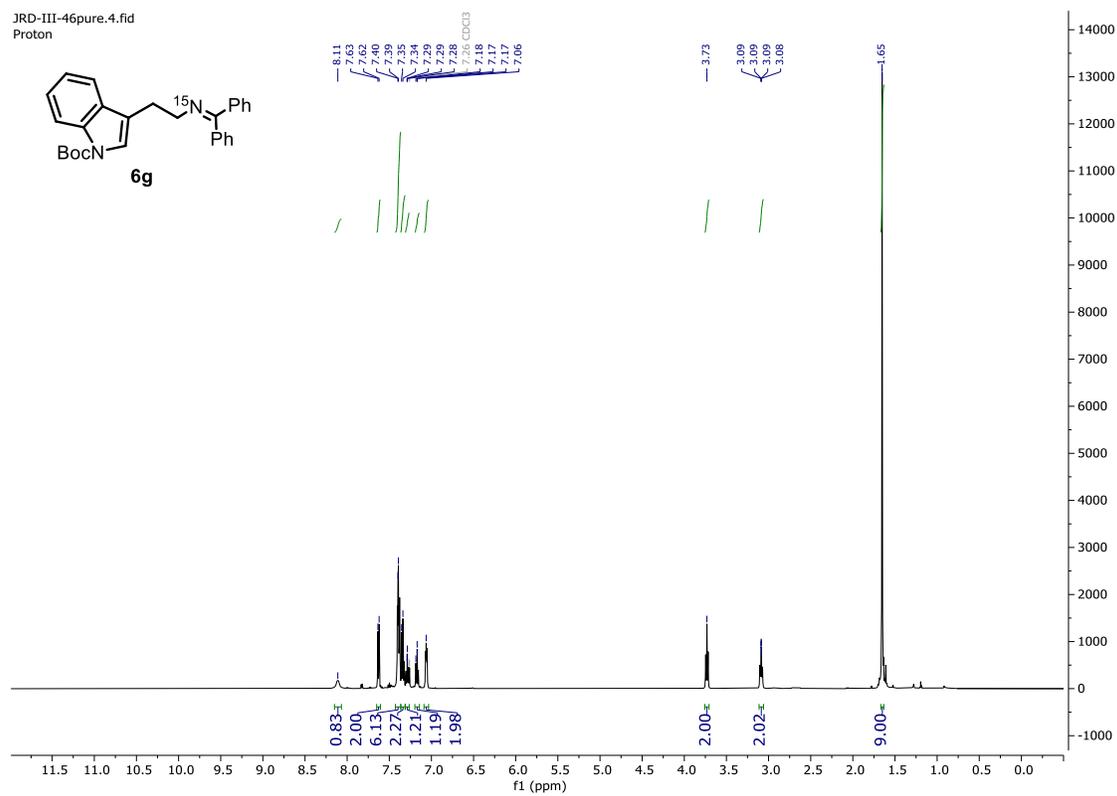
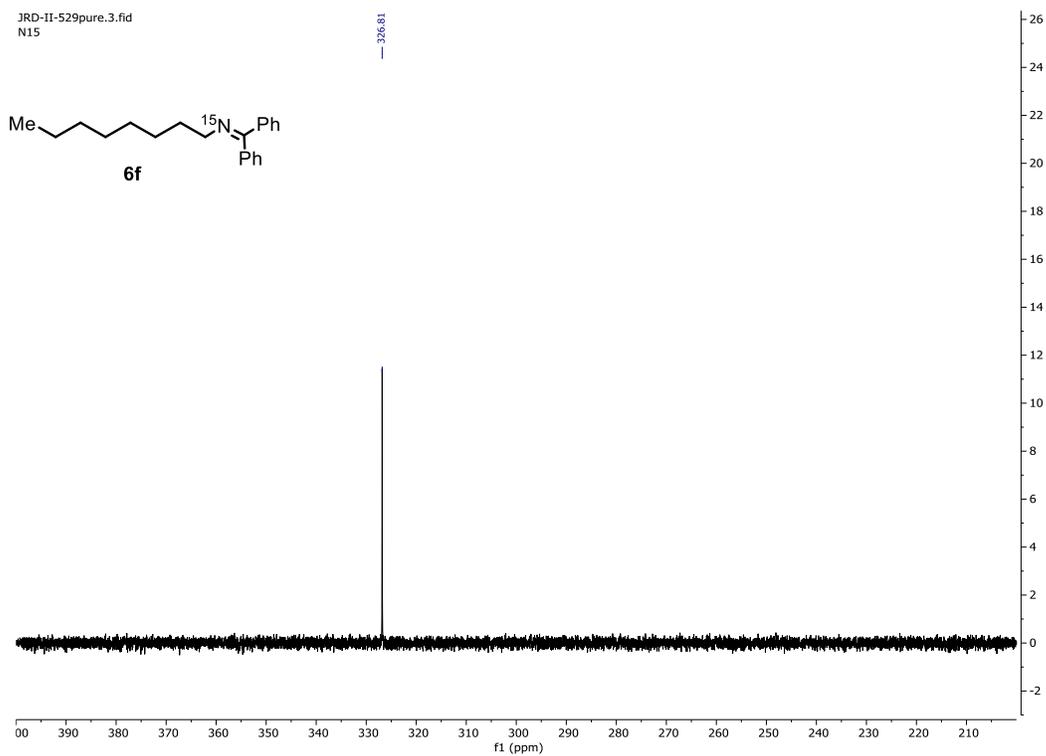


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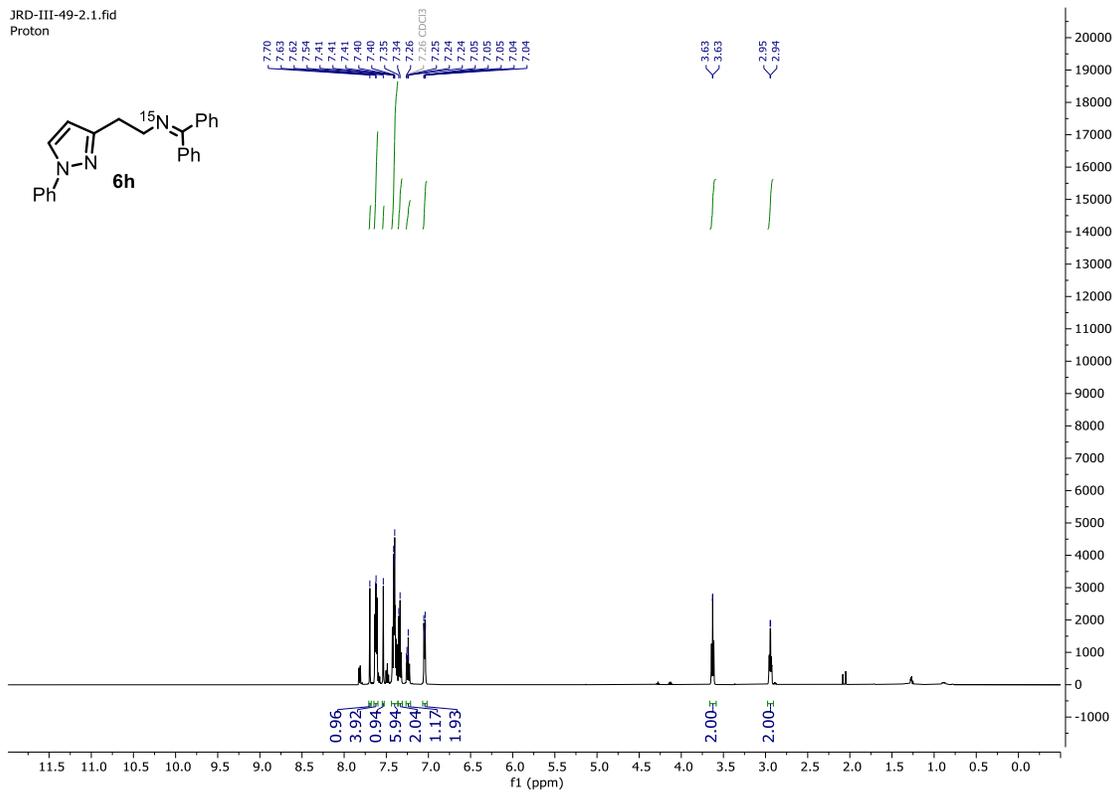
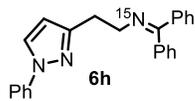


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Carbon CPD decoupled

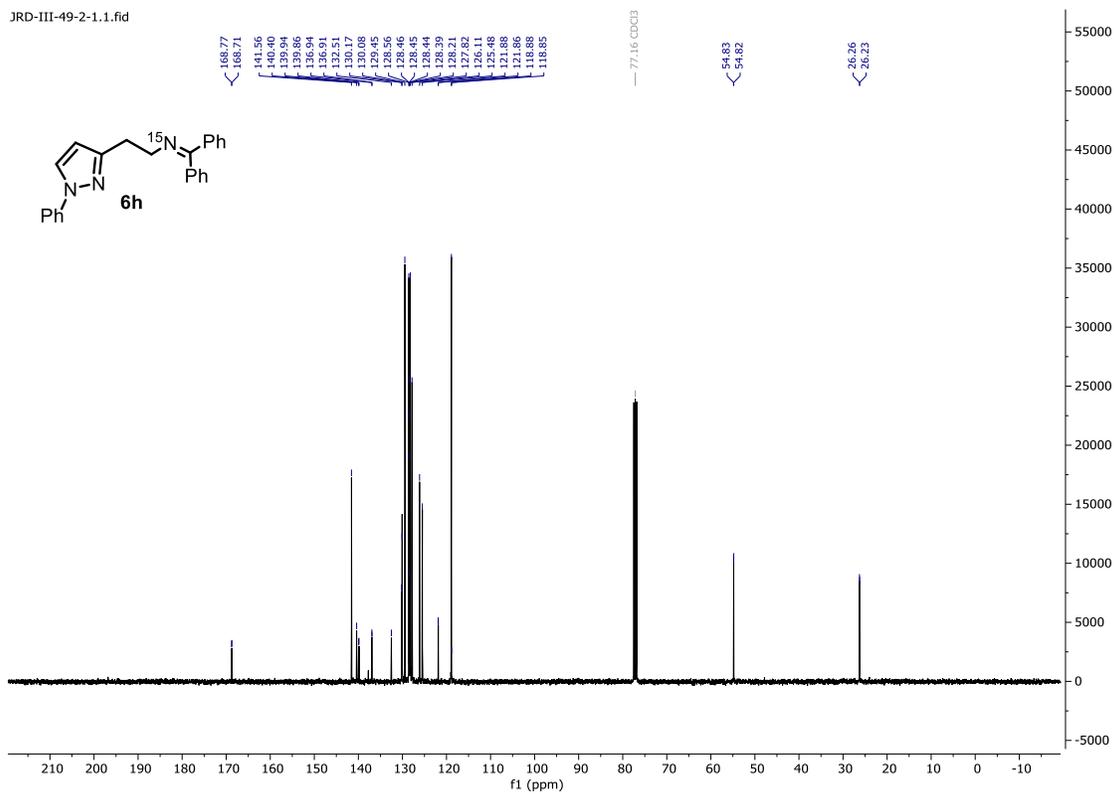
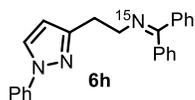




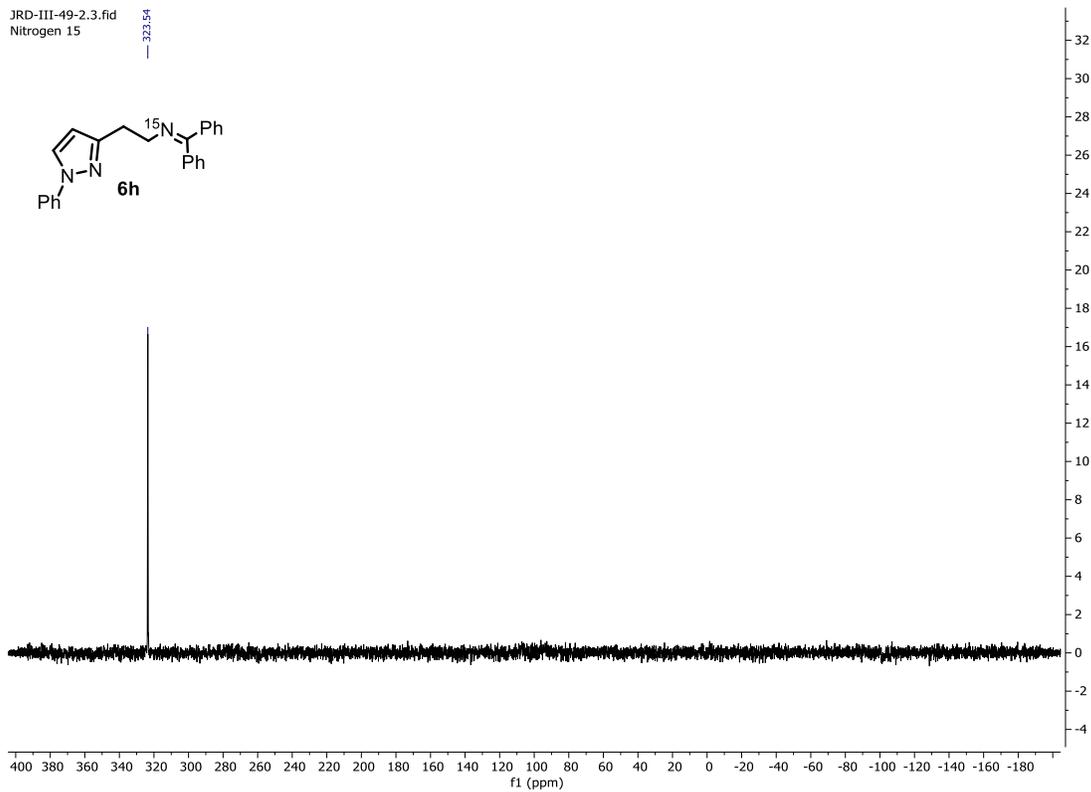
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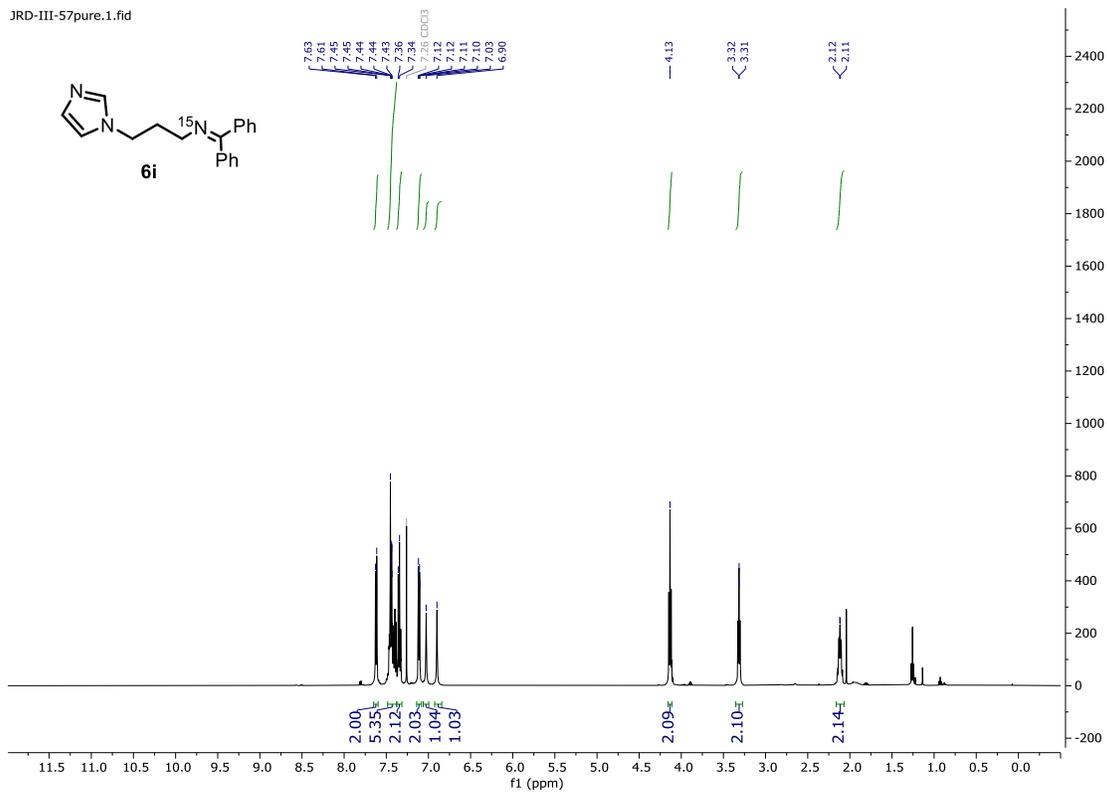
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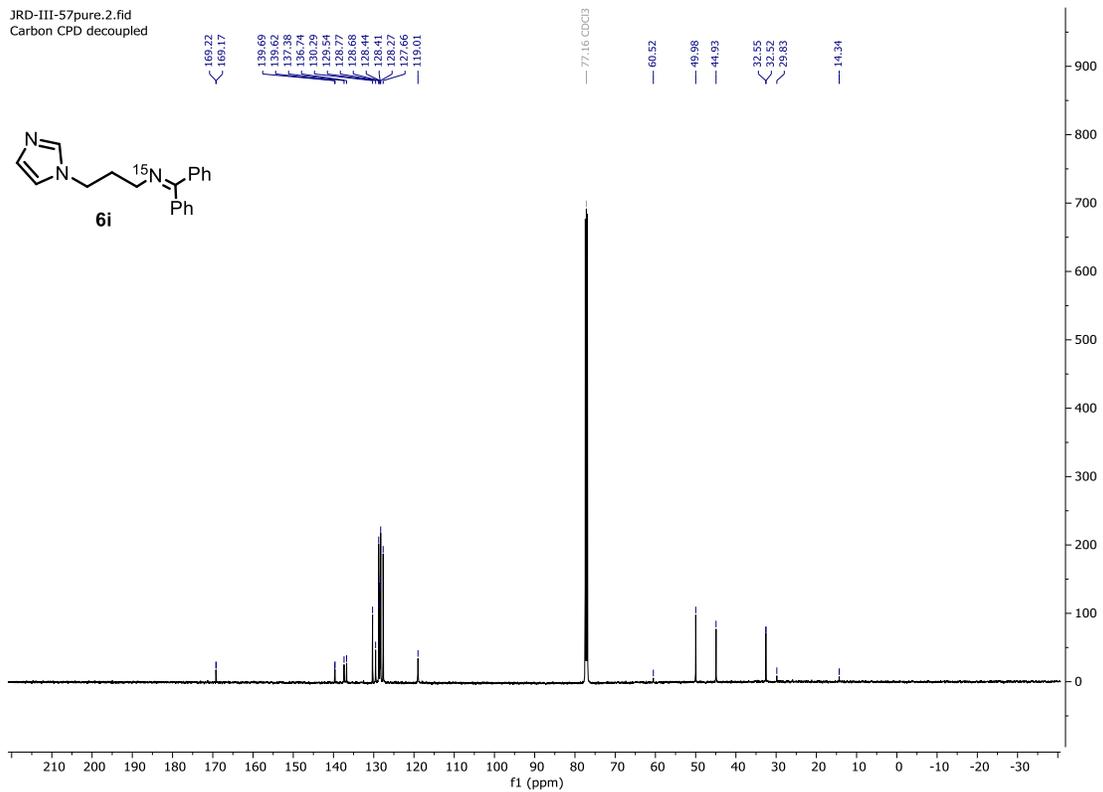
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Nitrogen 15



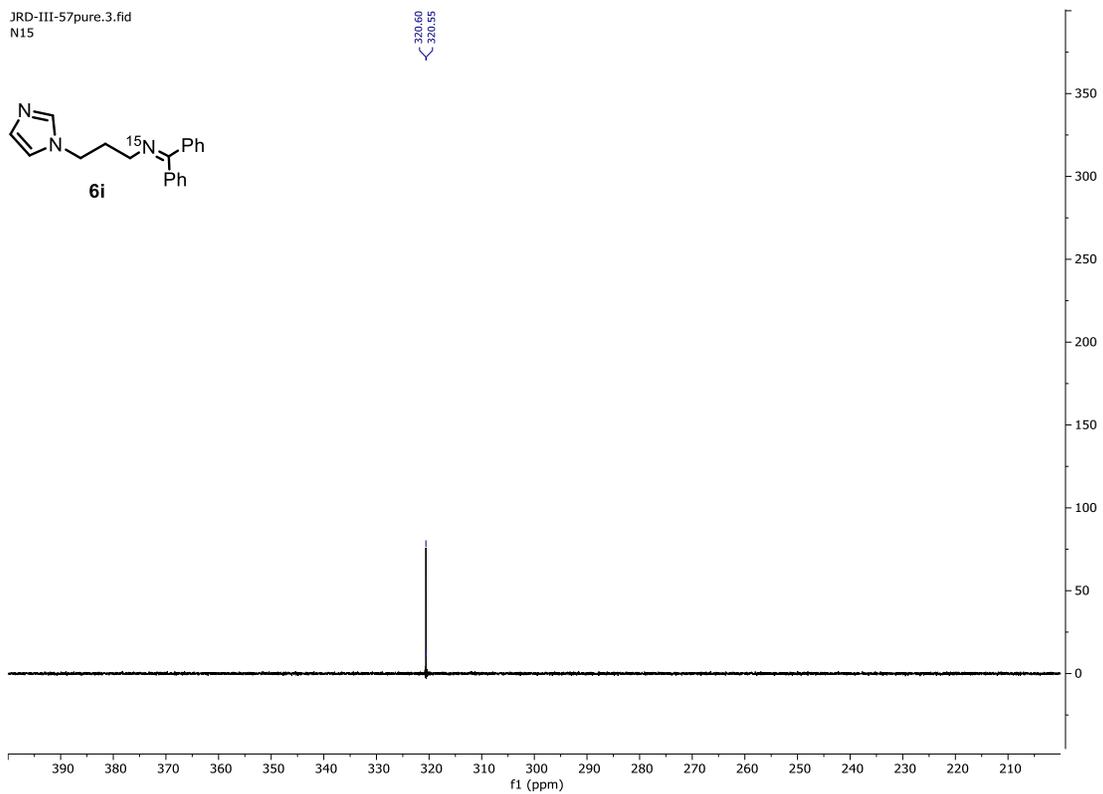
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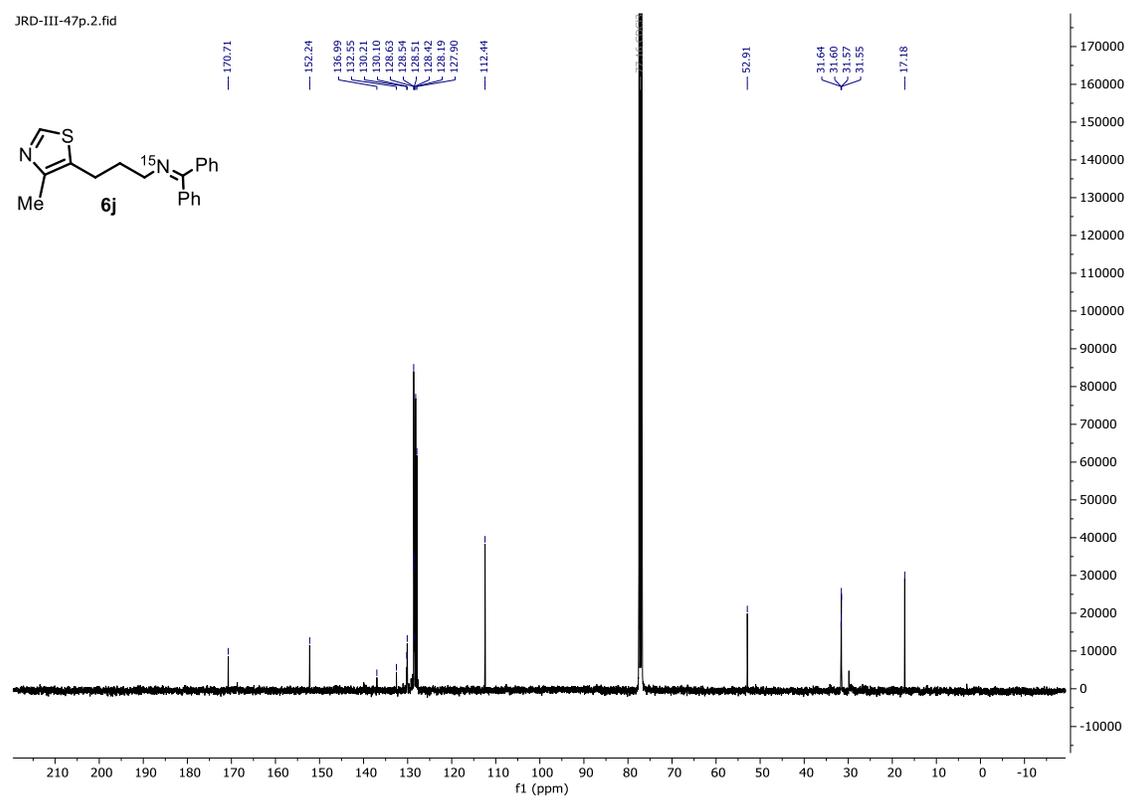
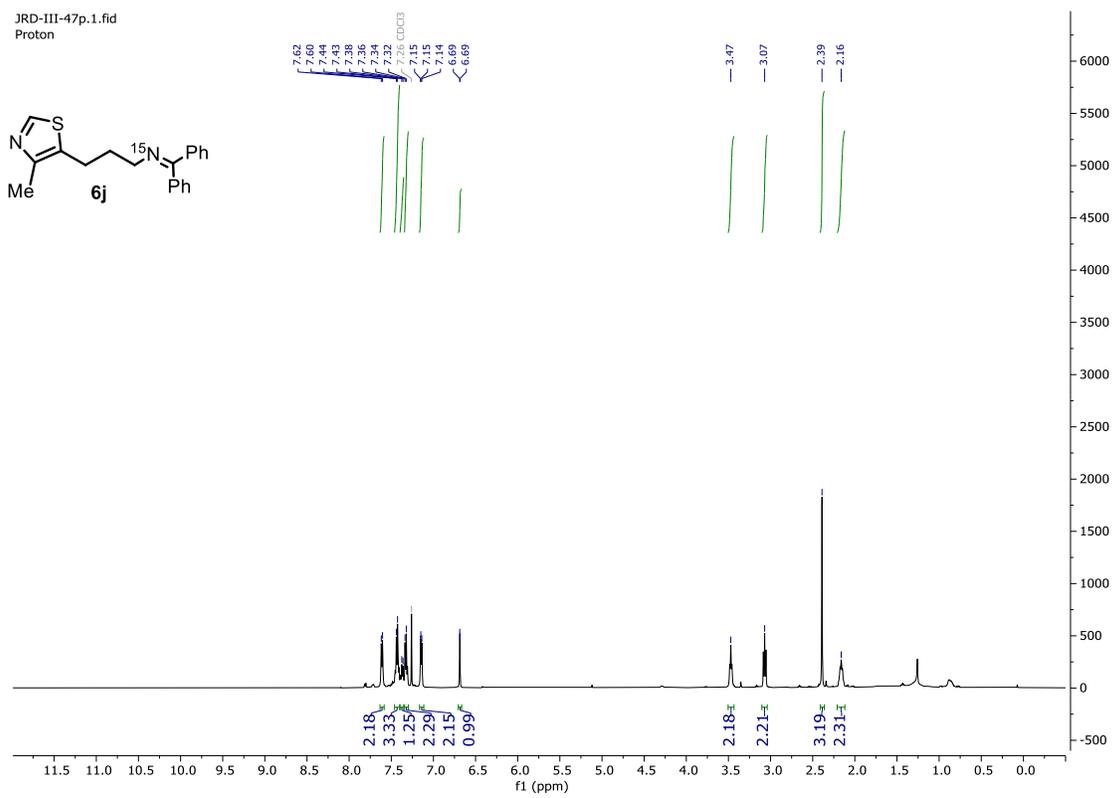


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Carbon CPD decoupled



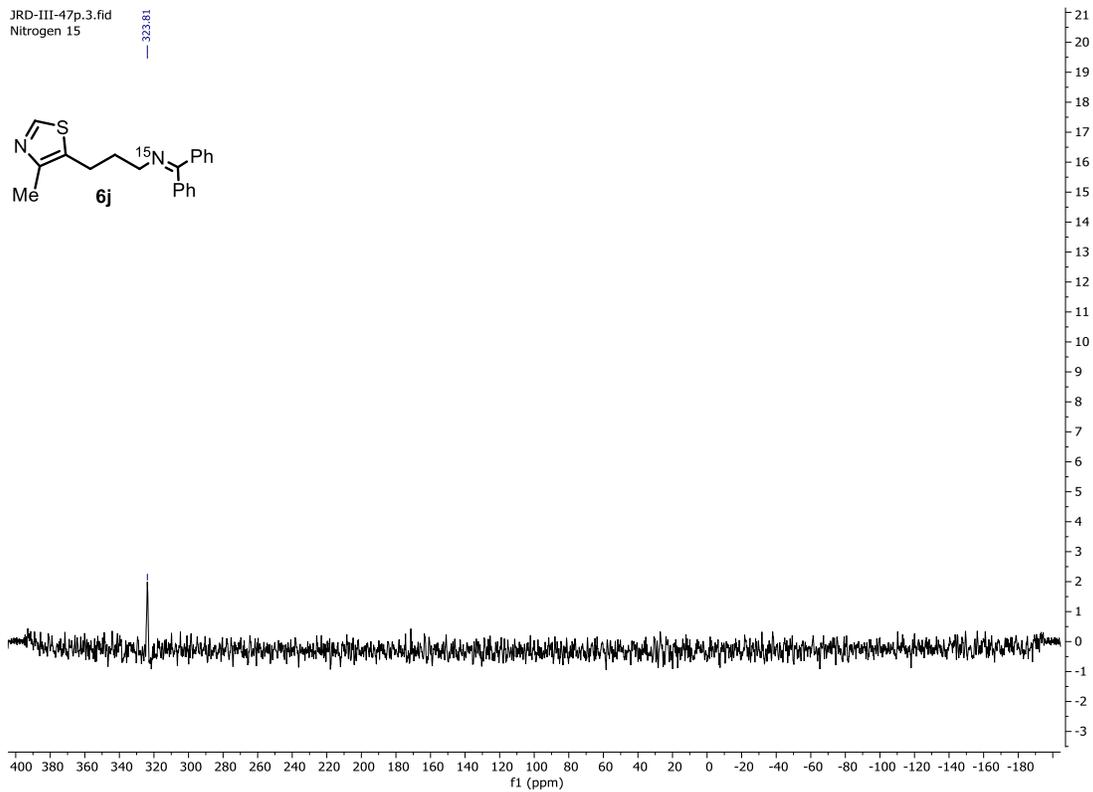
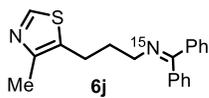
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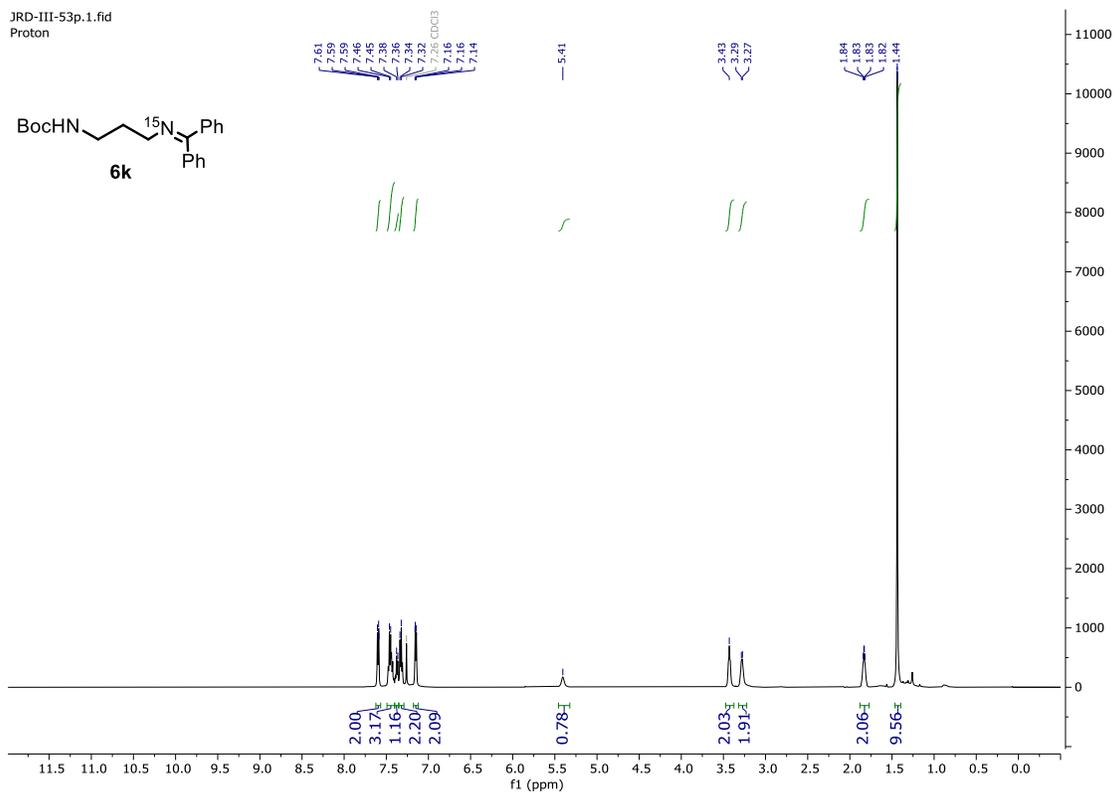
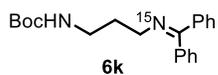


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Nitrogen 15

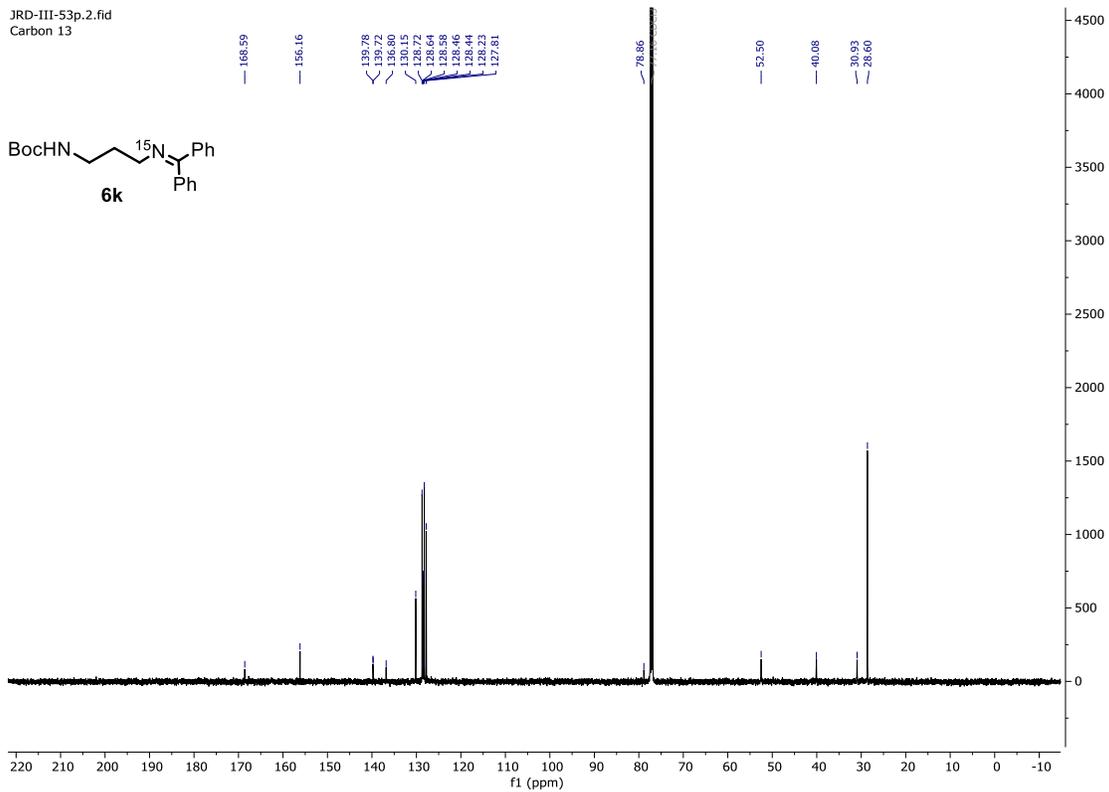
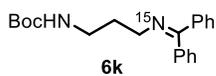
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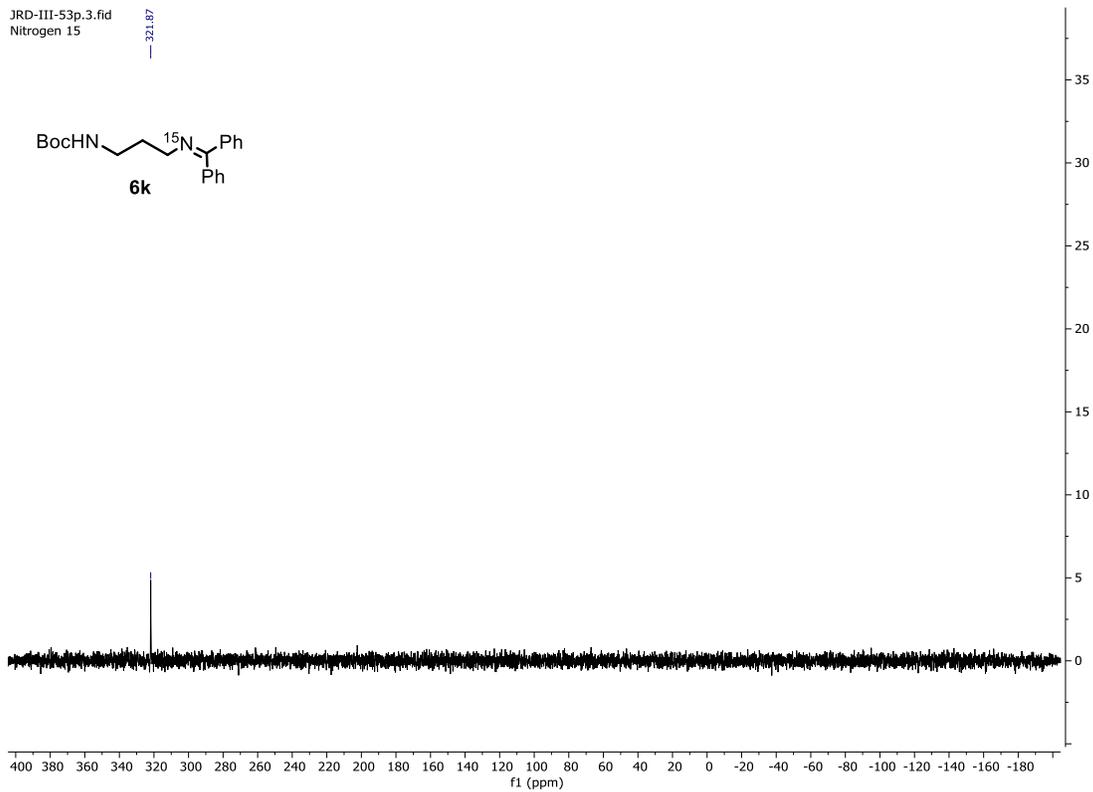
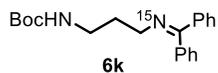
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Proton



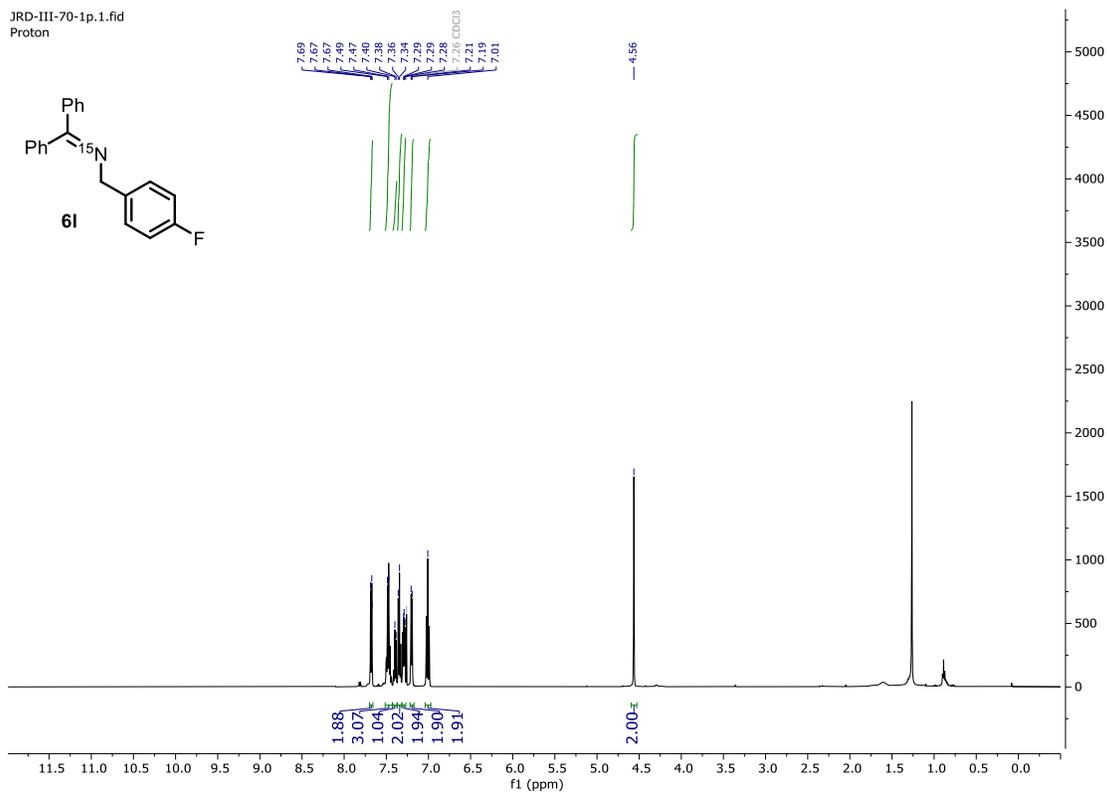
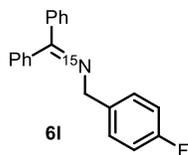
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Carbon 13



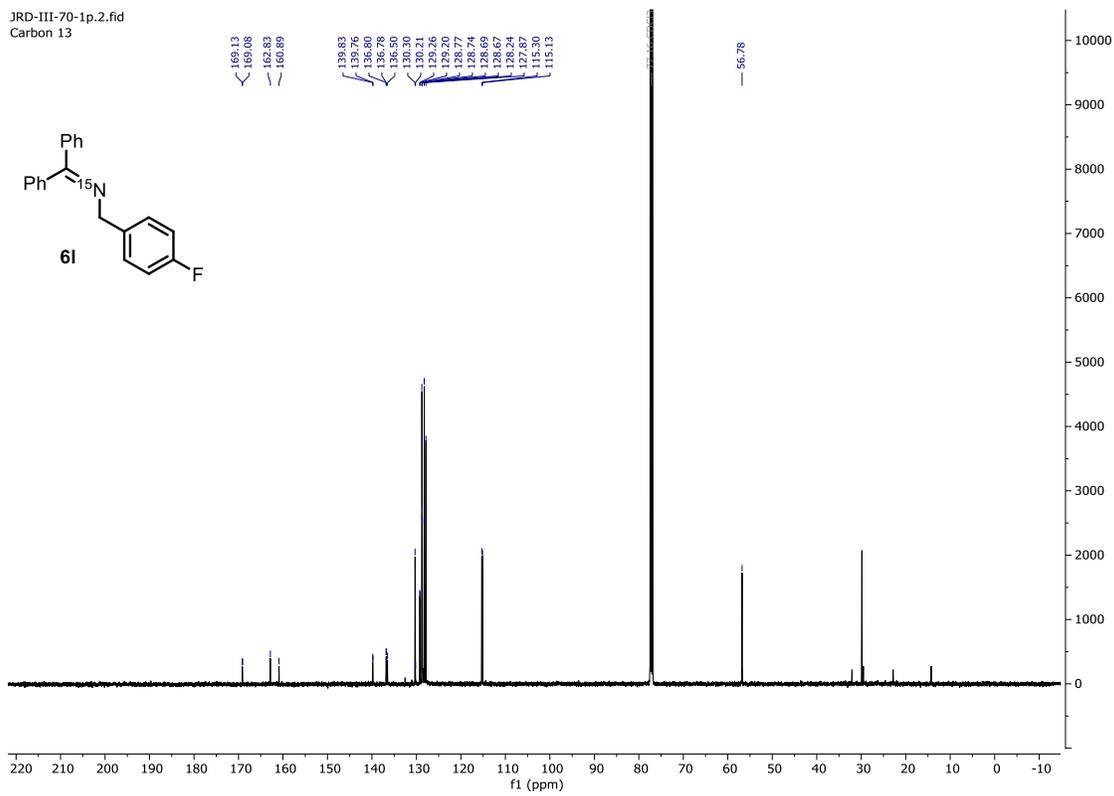
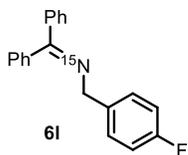
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Nitrogen 15



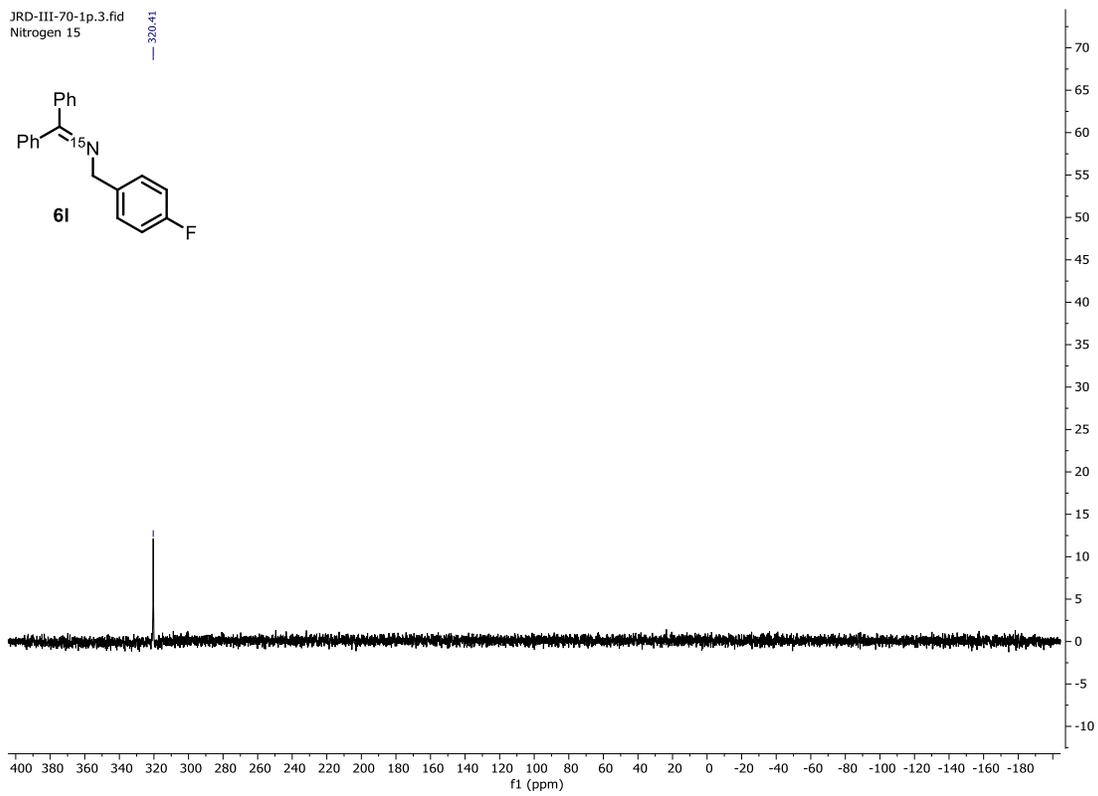
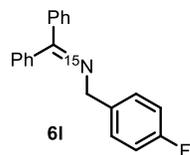
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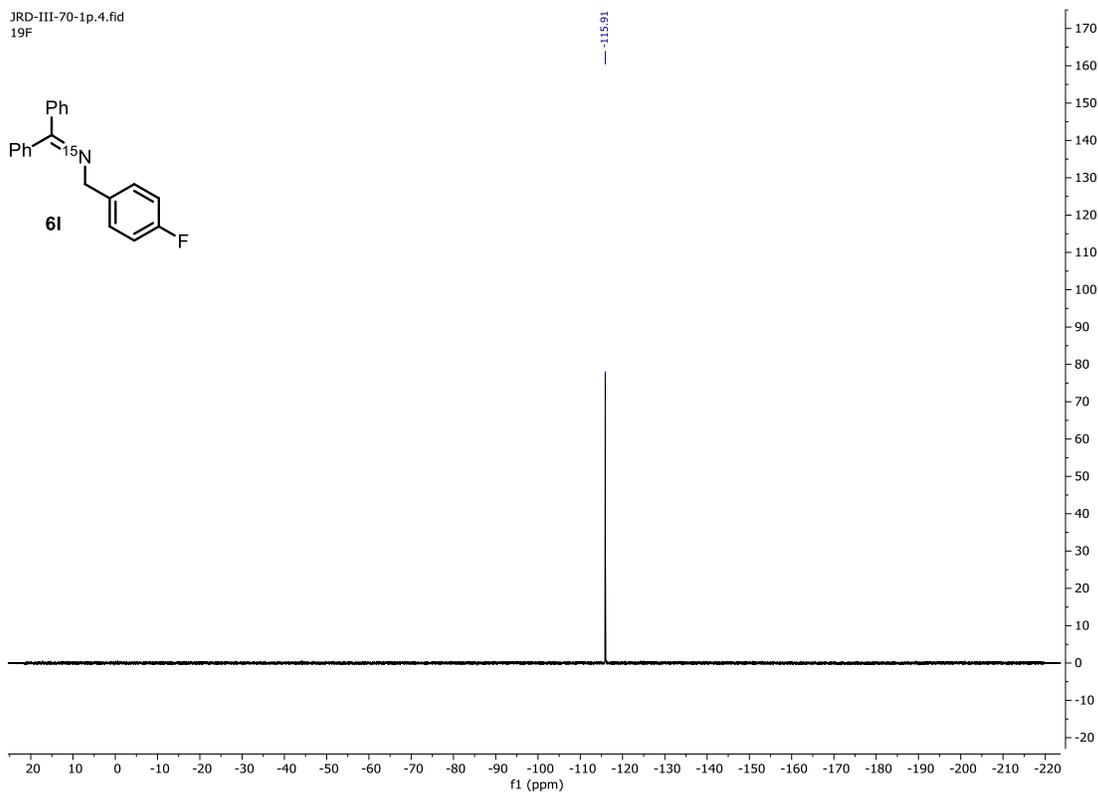
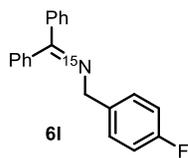
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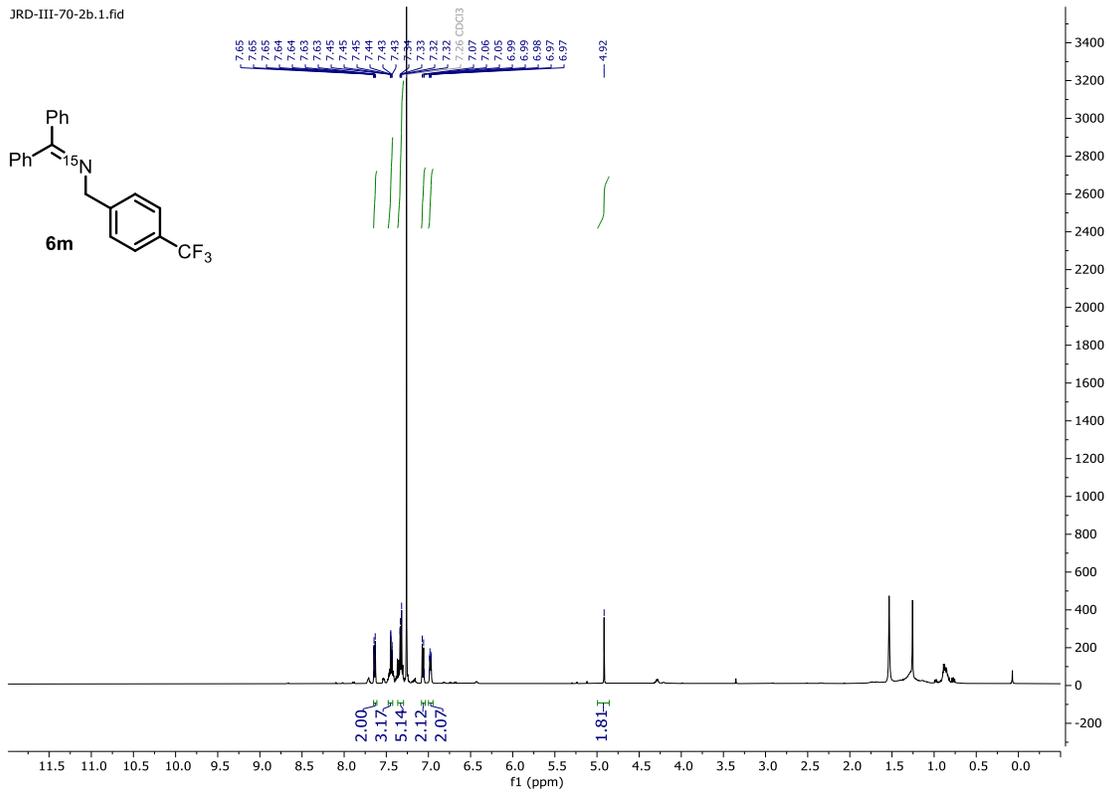
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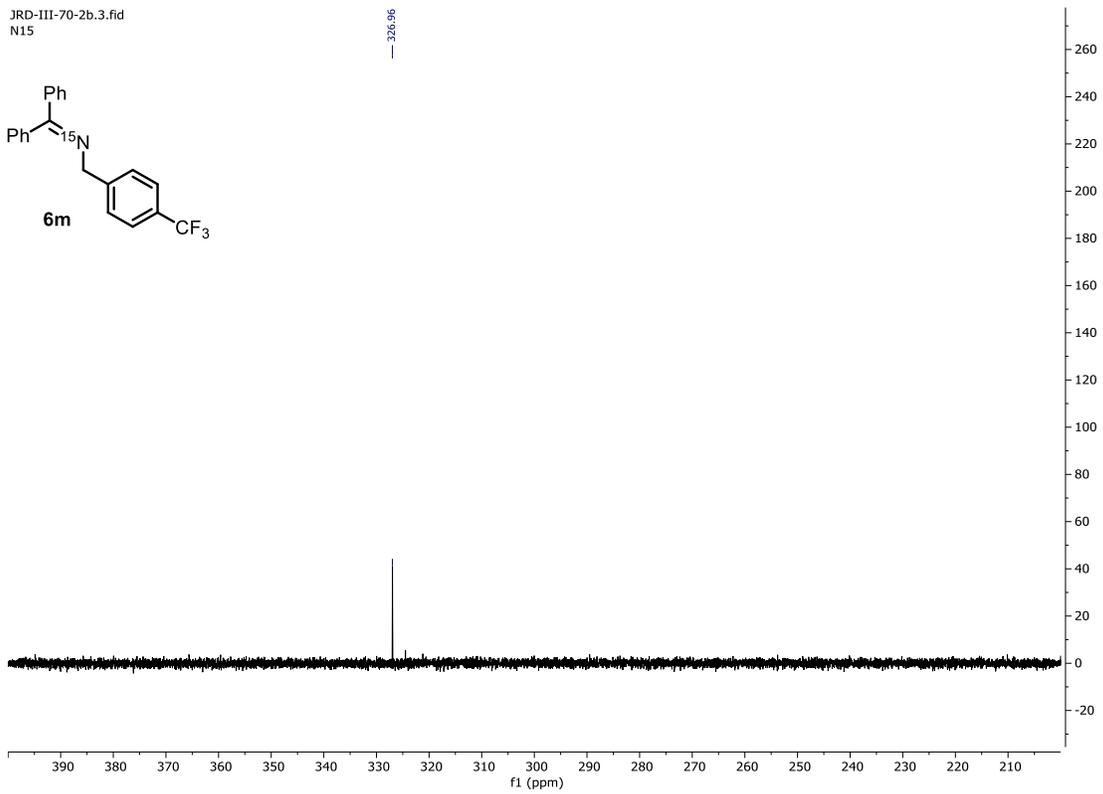
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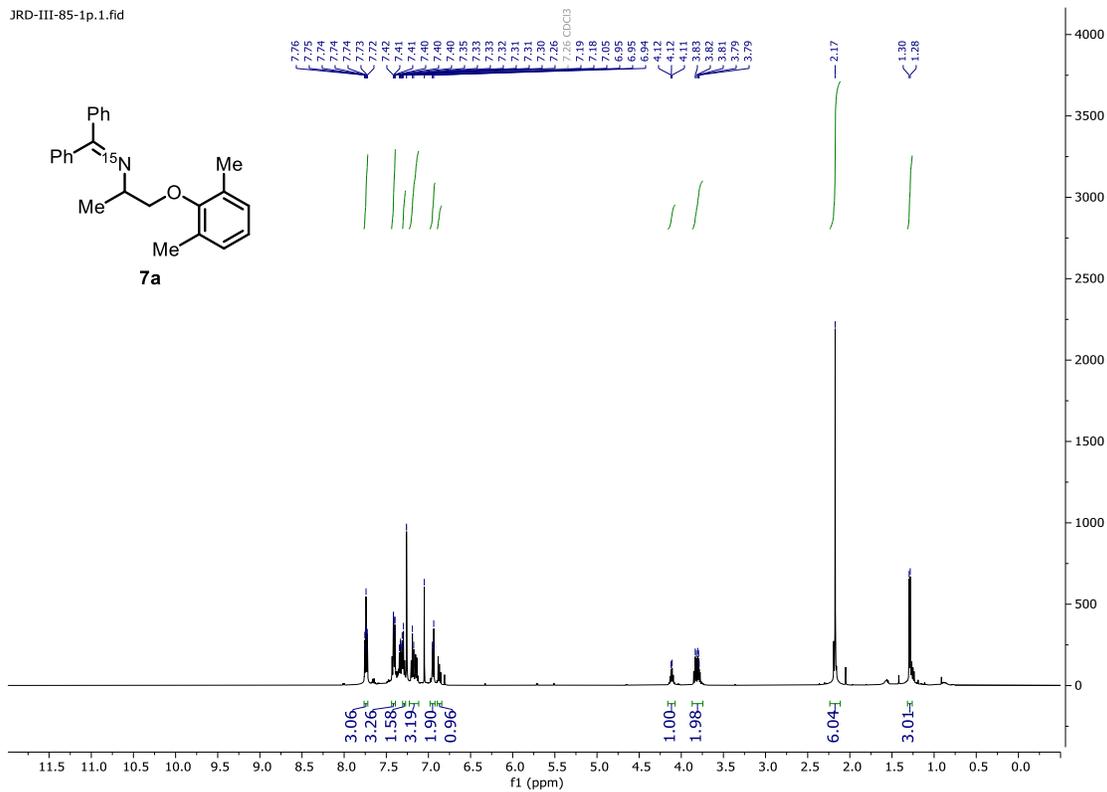
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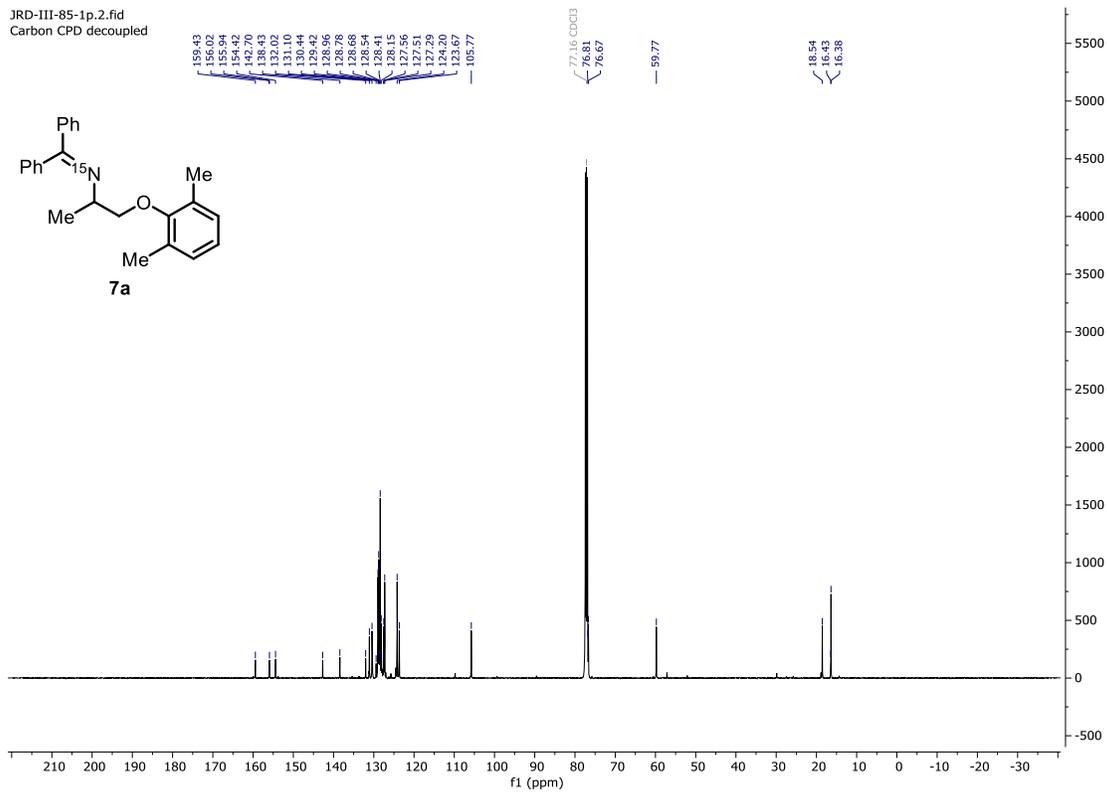
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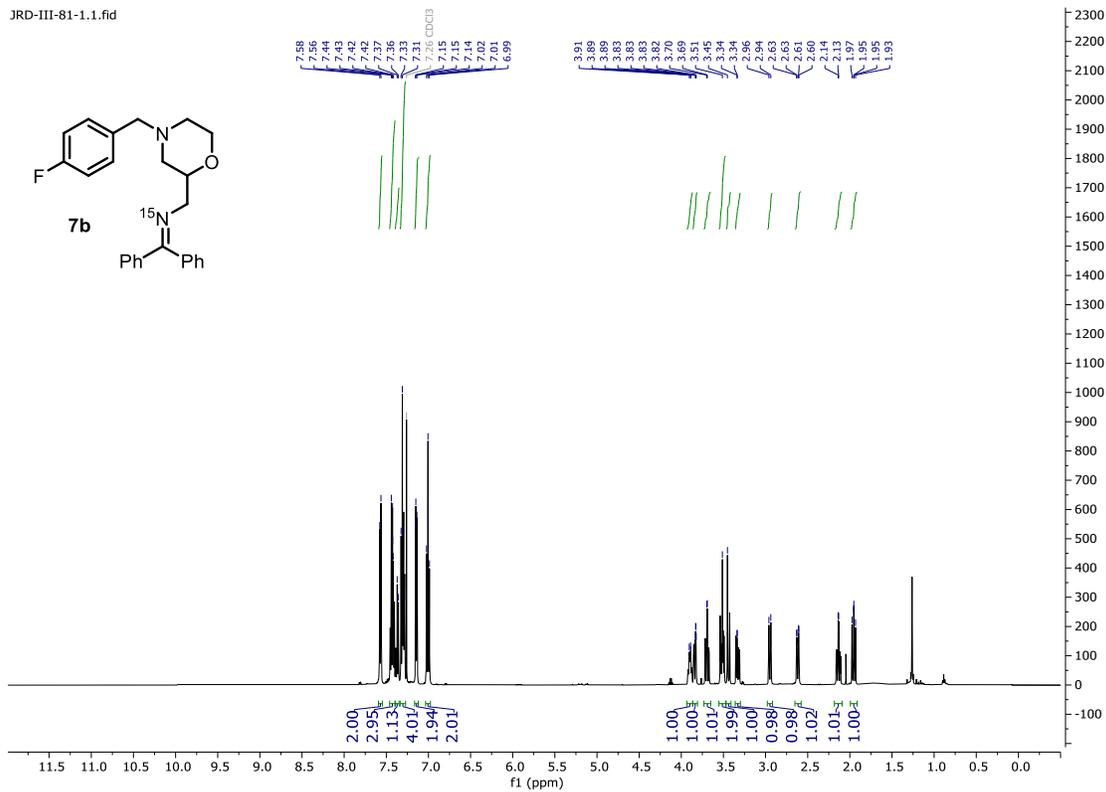
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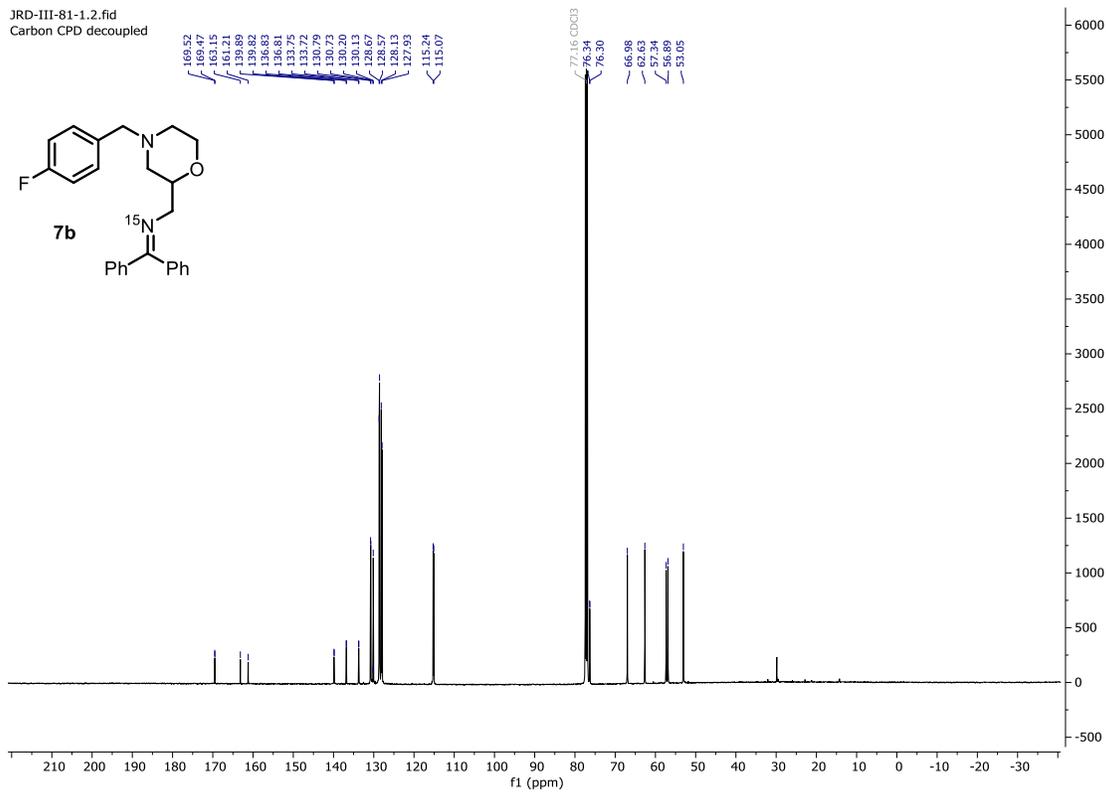
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Carbon CPD decoupled



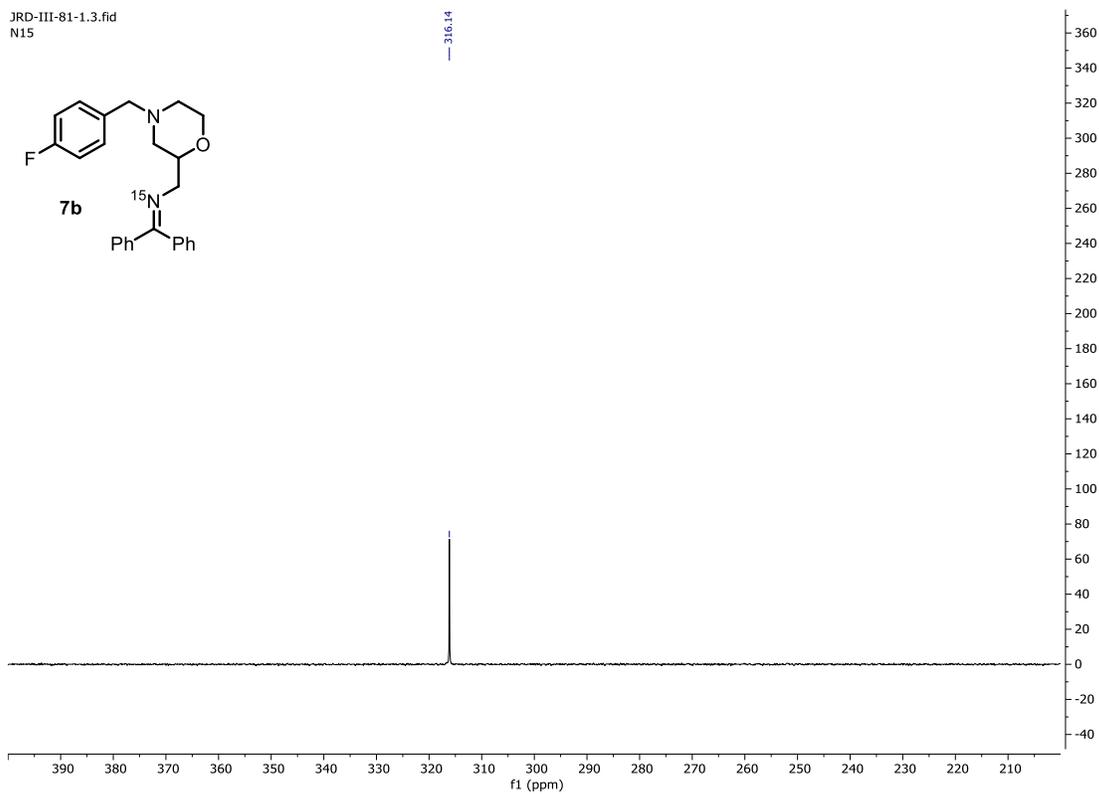
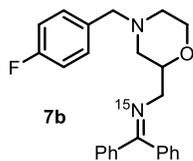
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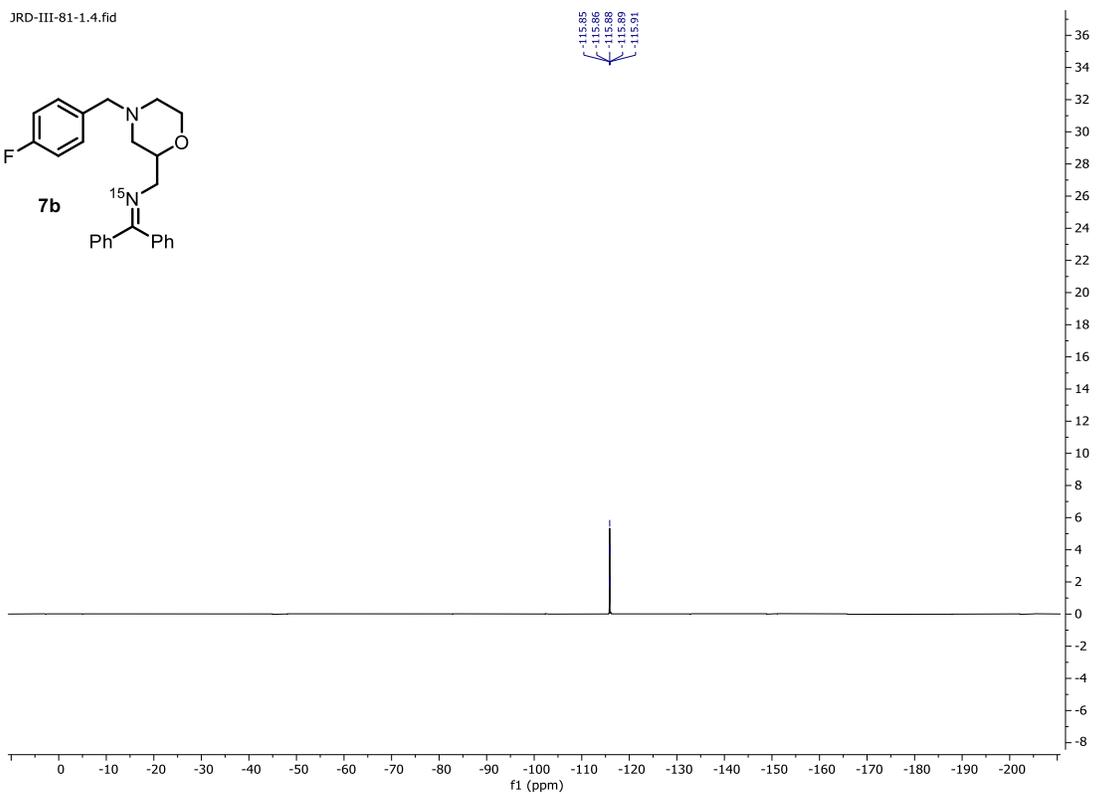
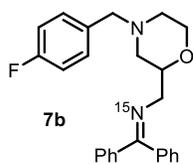
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Carbon CPD decoupled



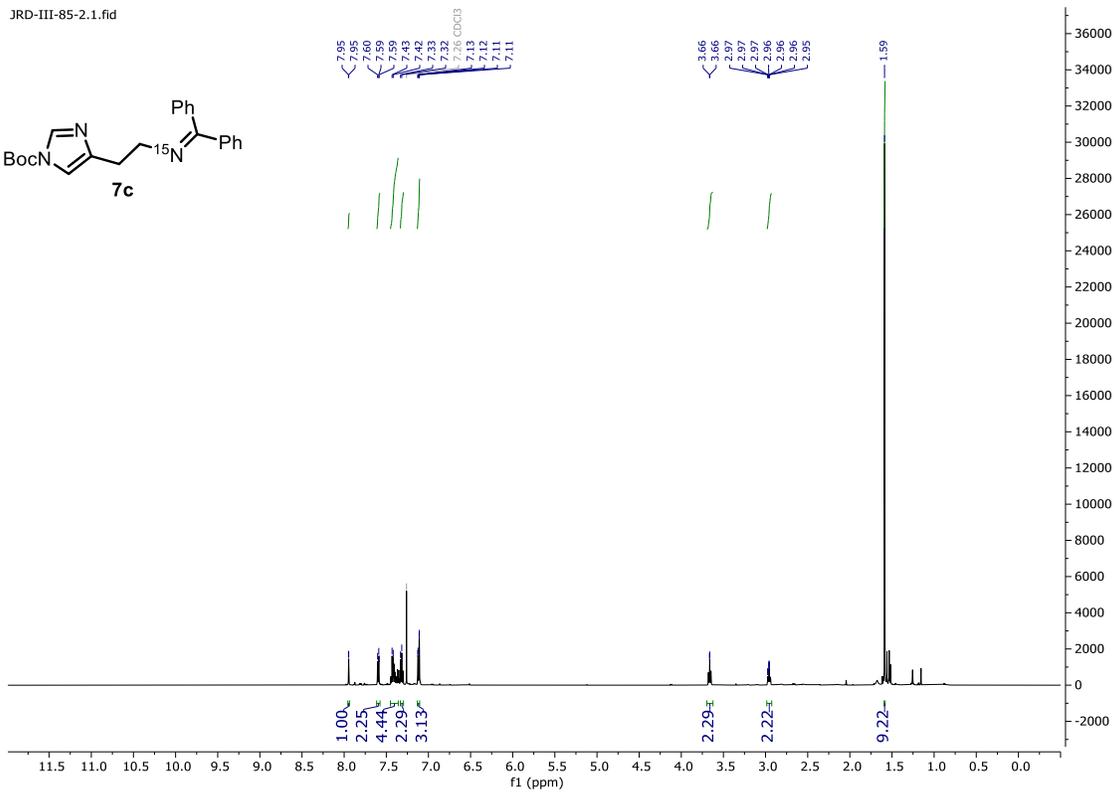
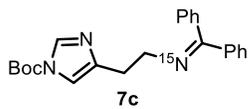
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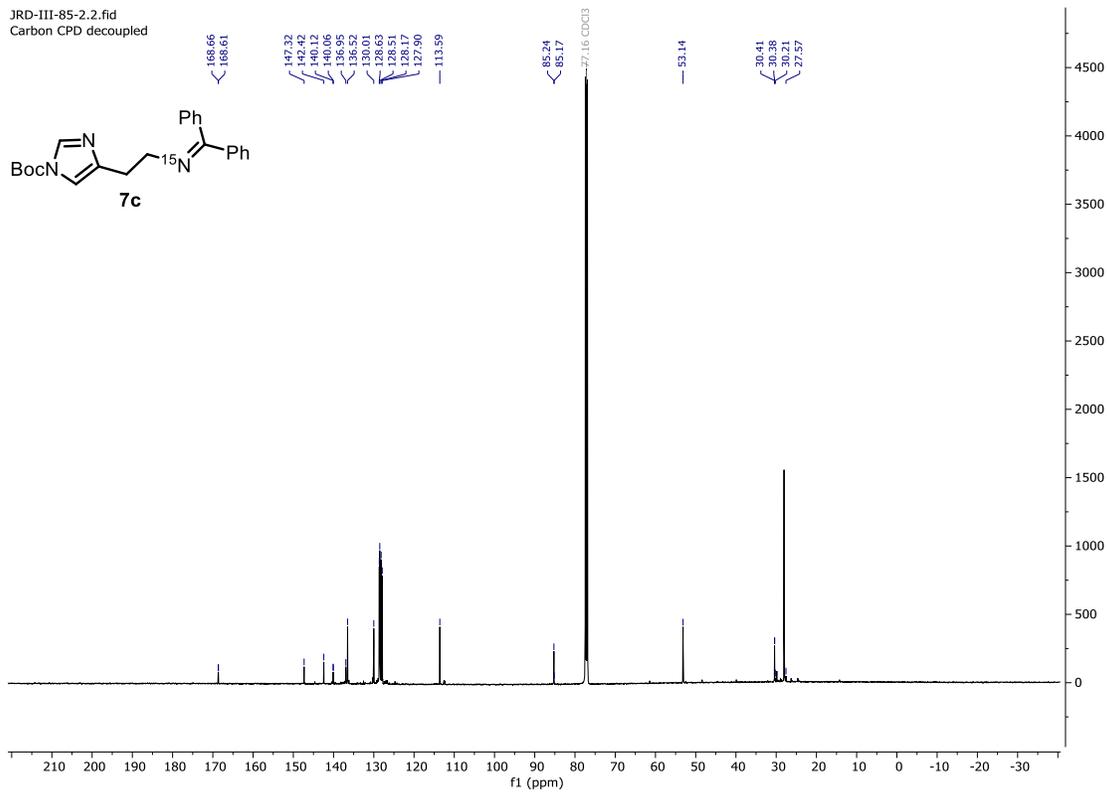
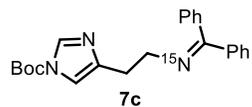
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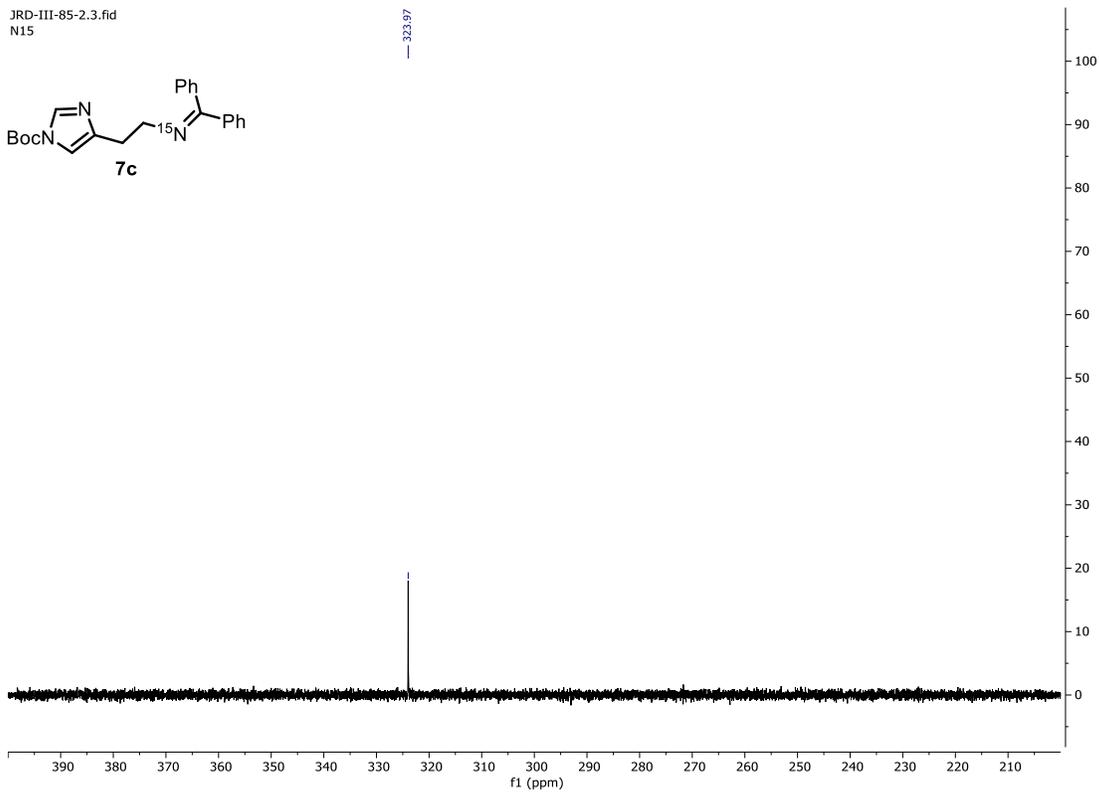
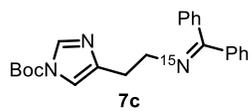
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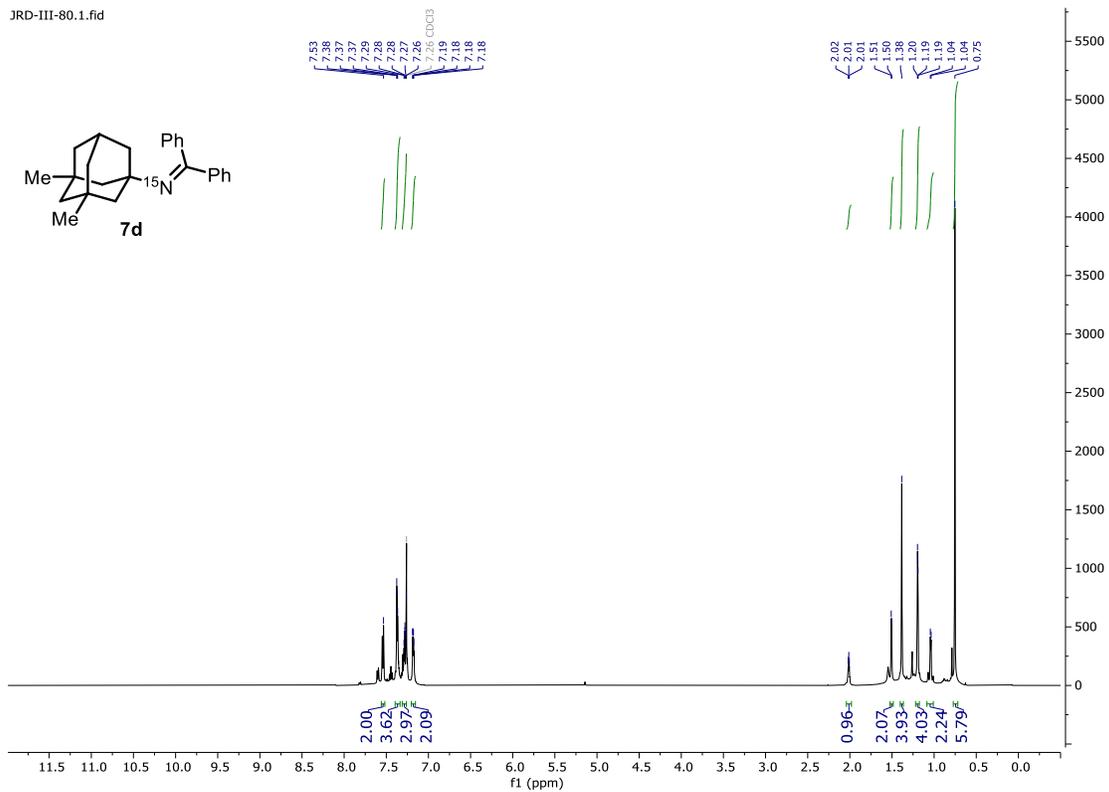
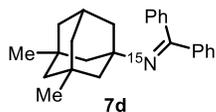
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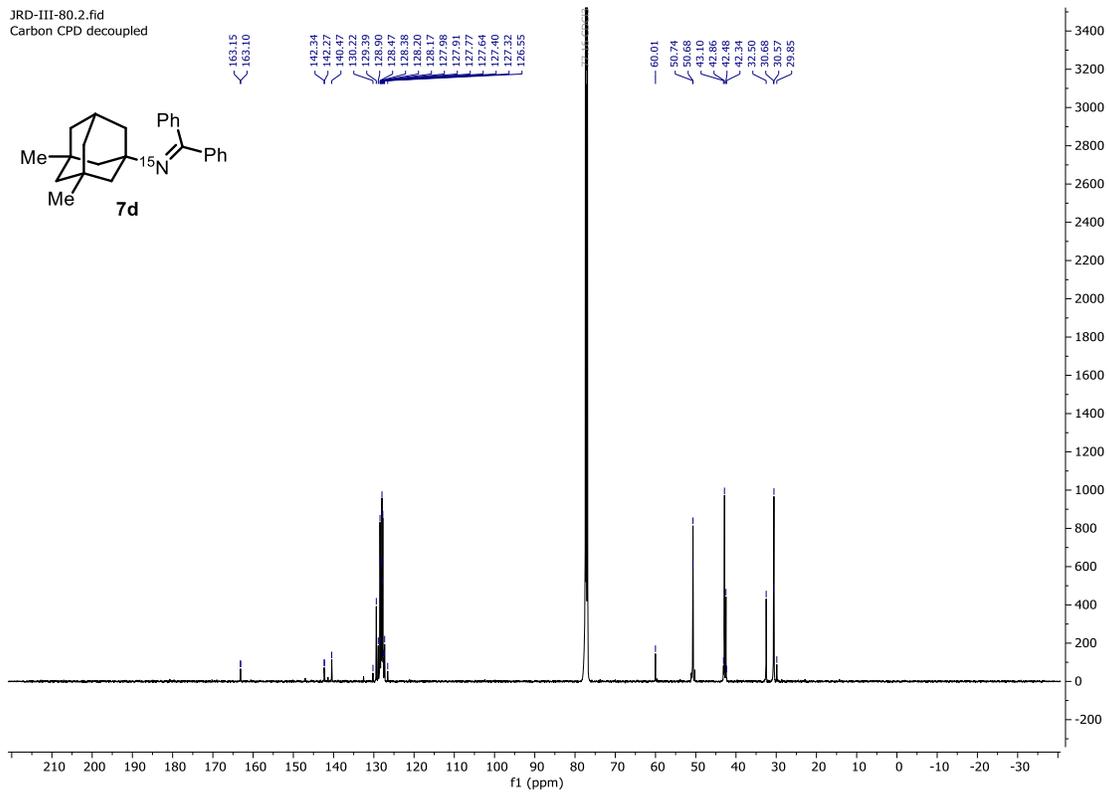
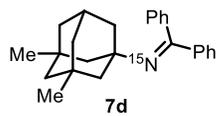
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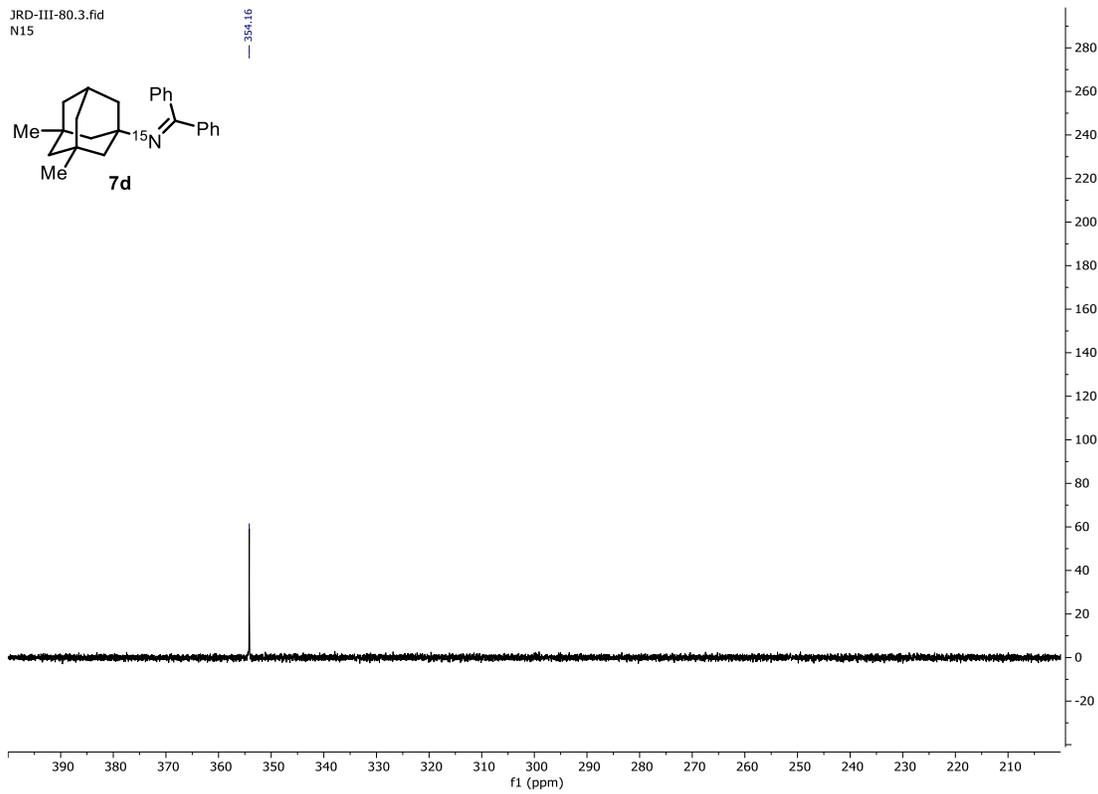
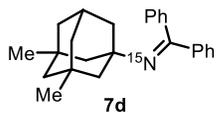
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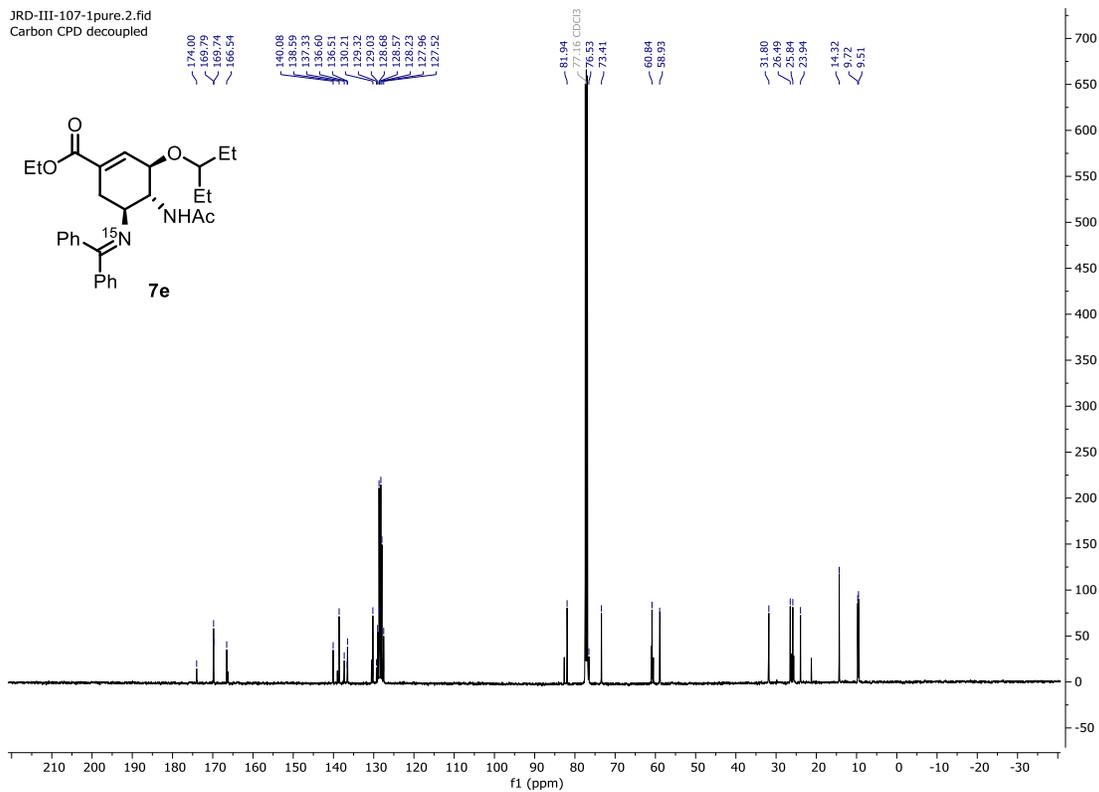
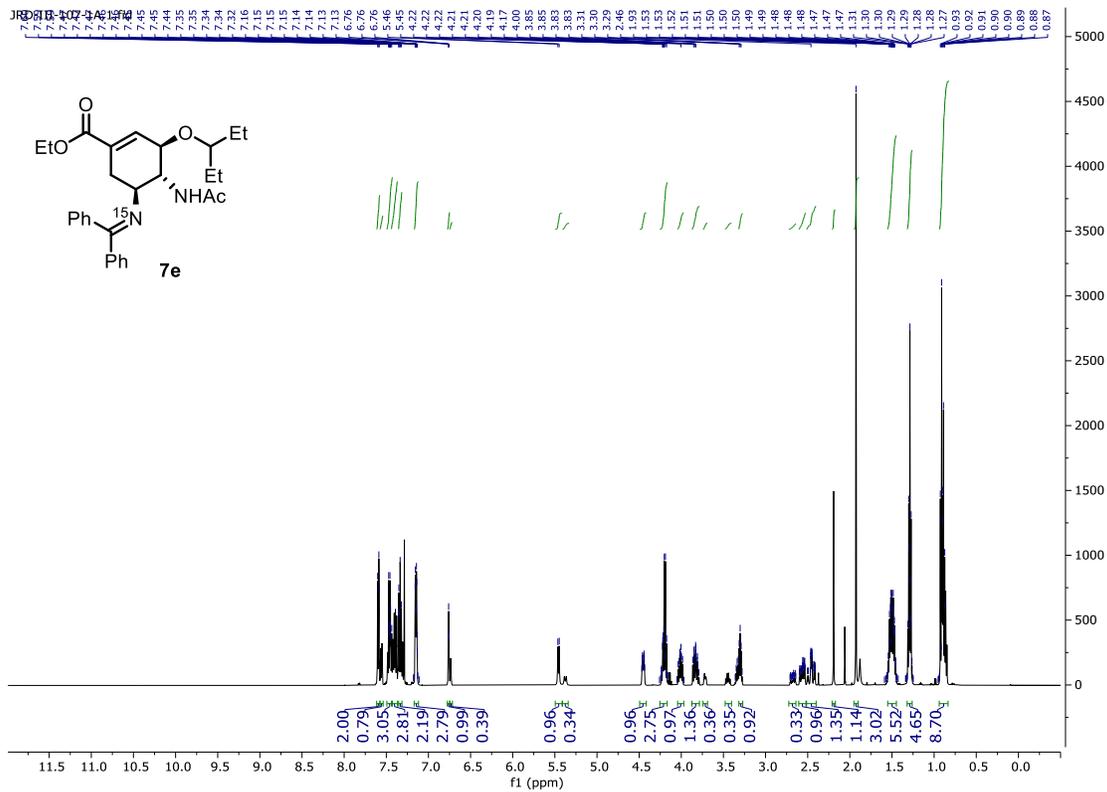


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Carbon CPD decoupled

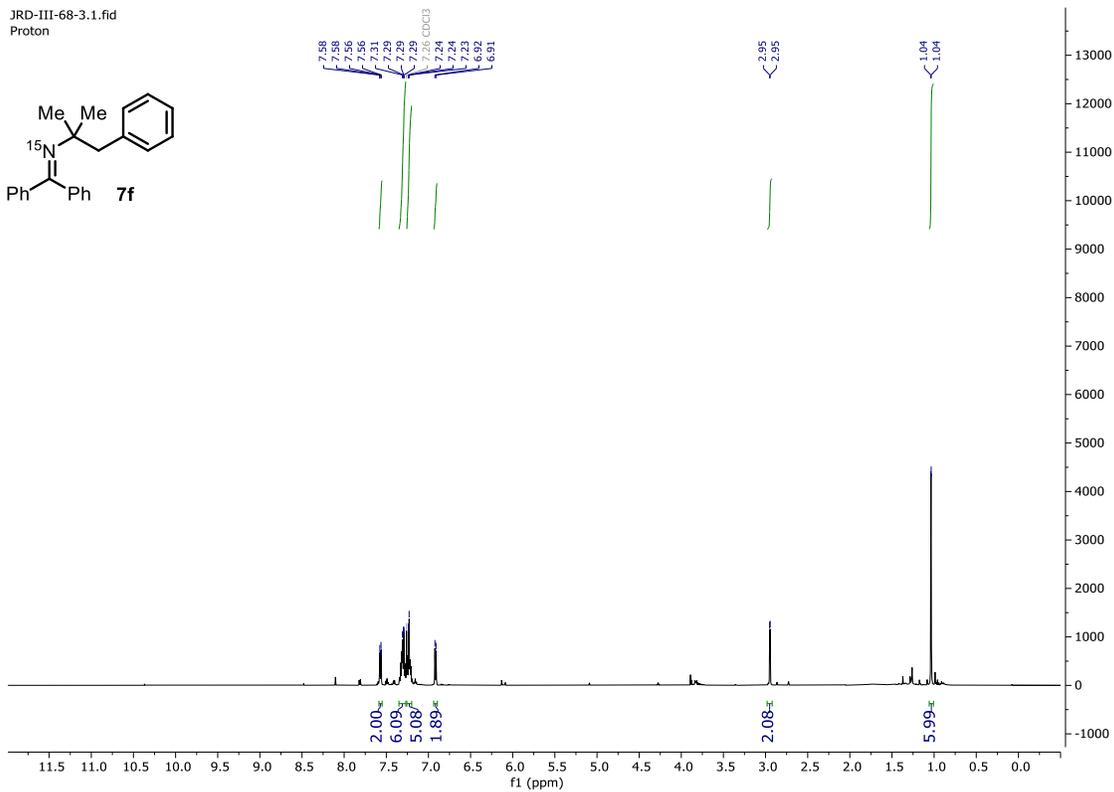
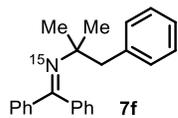


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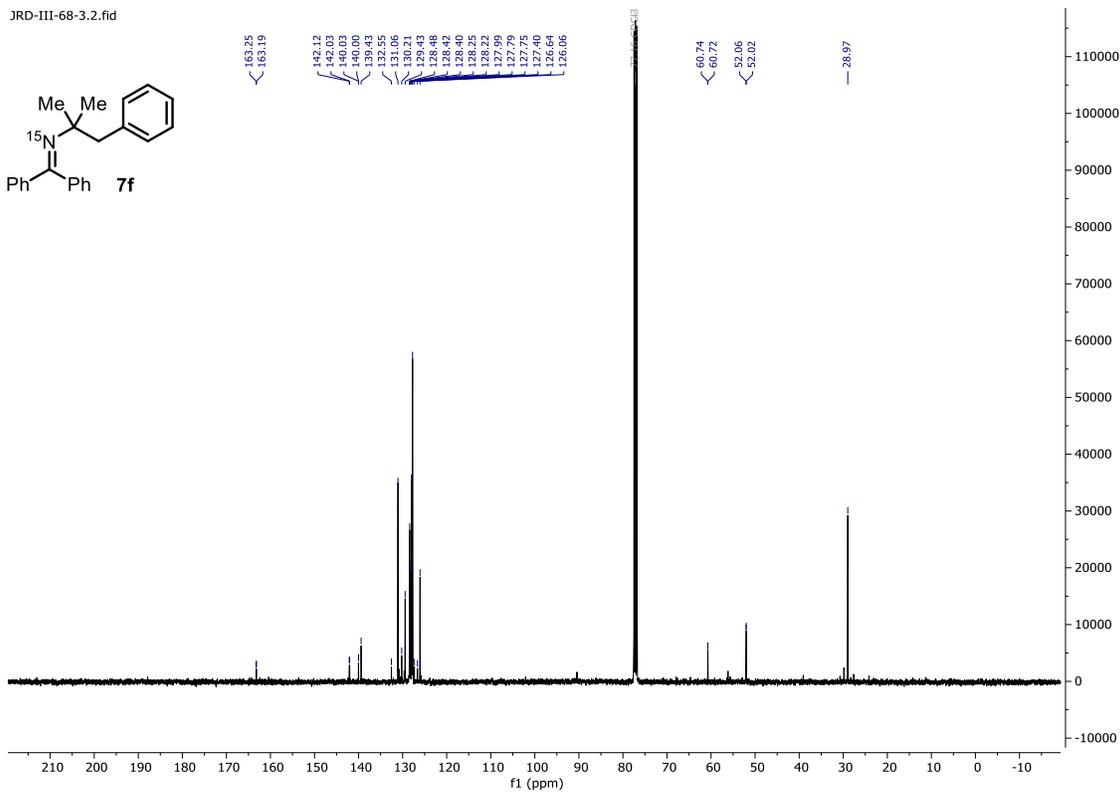
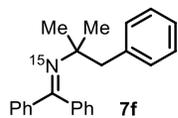




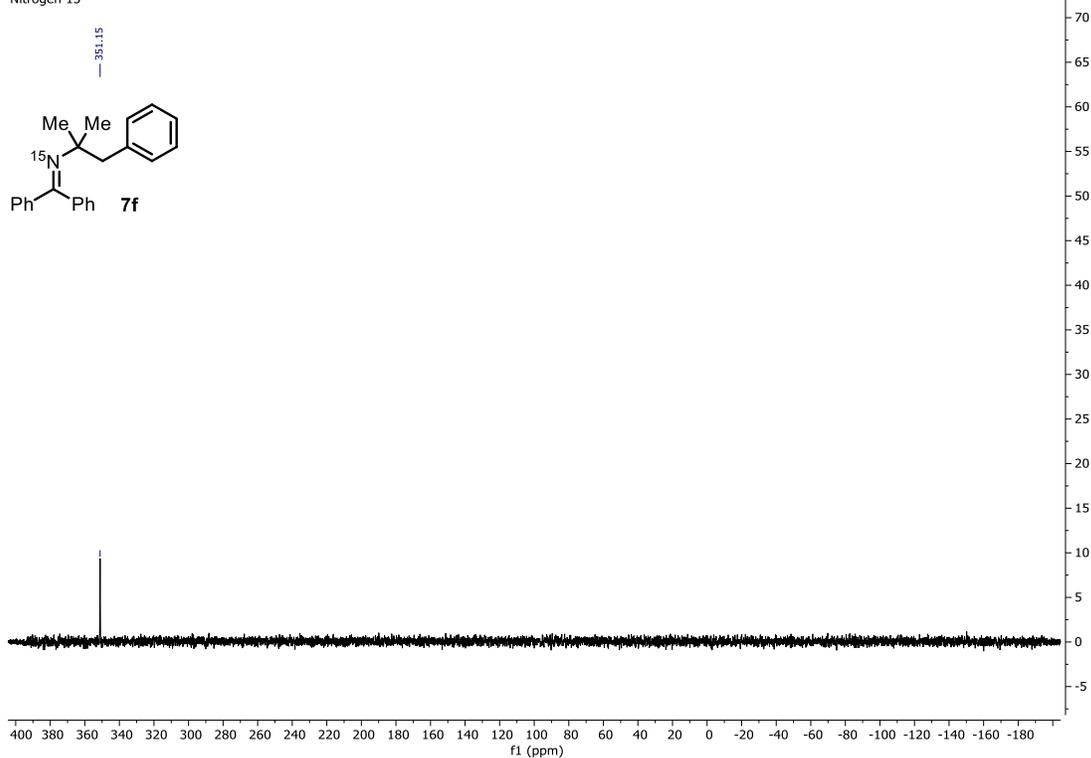
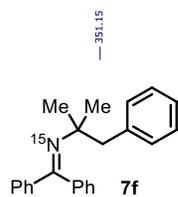
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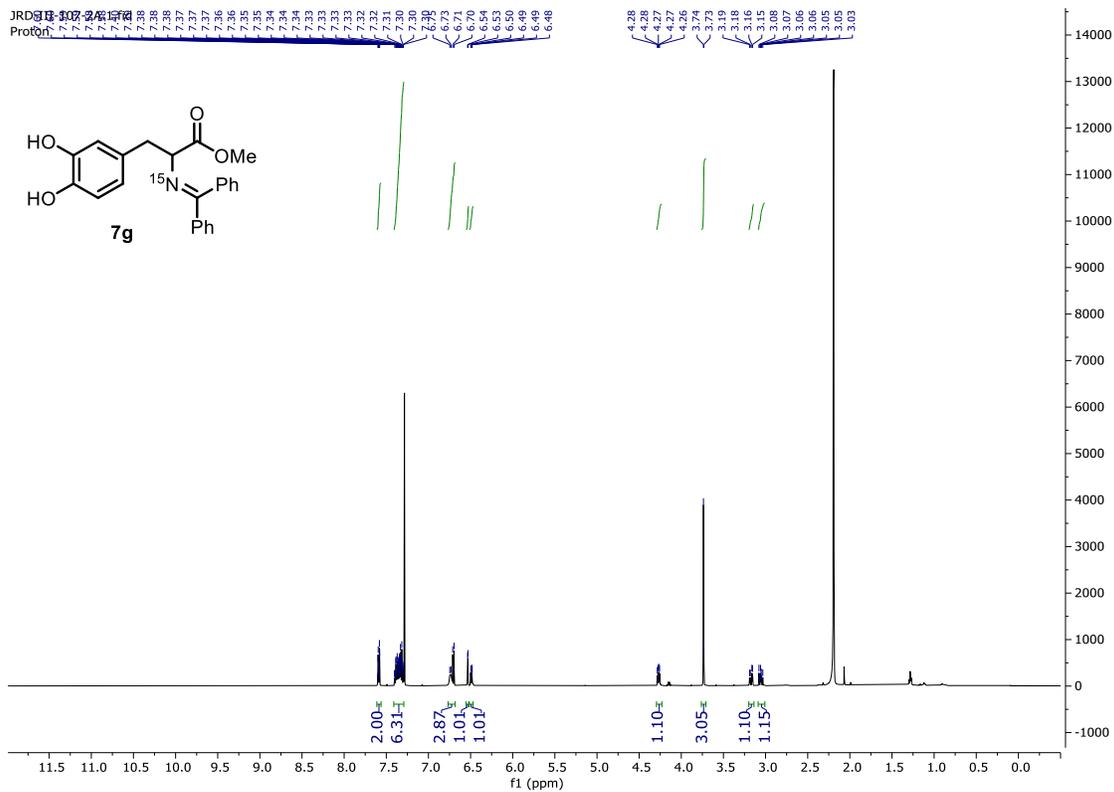
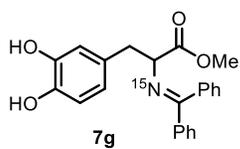
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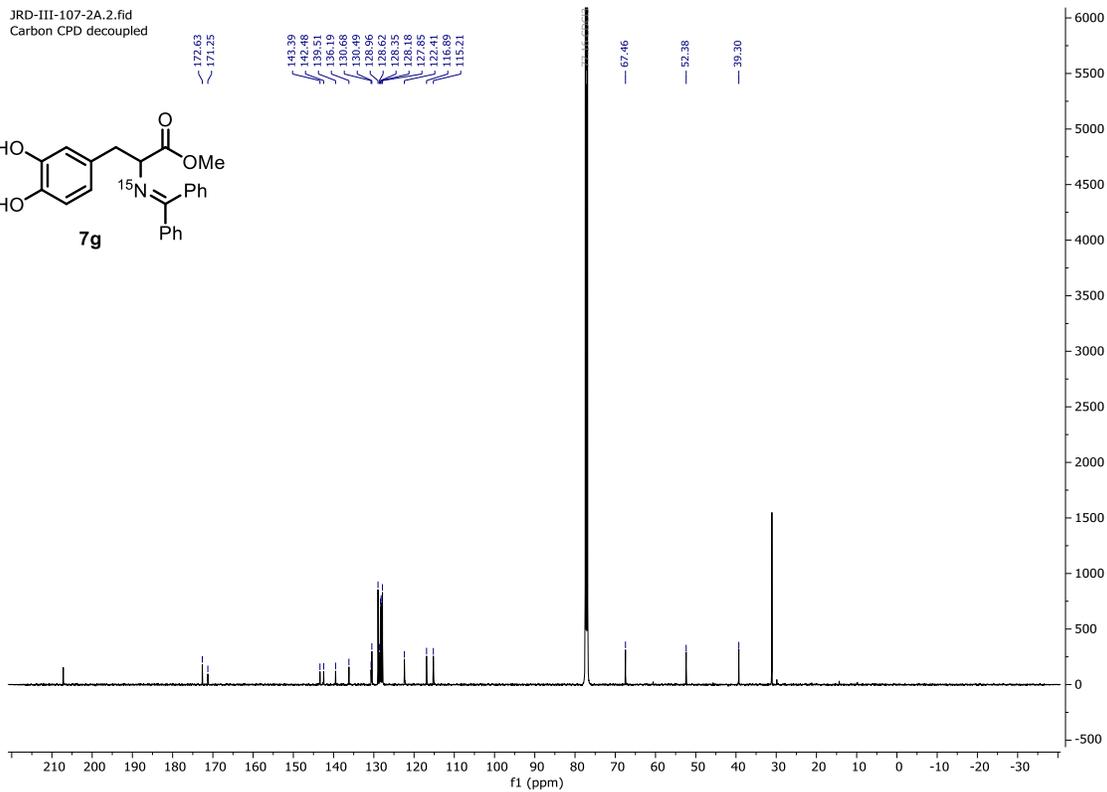
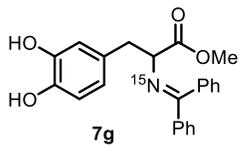
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Nitrogen 15



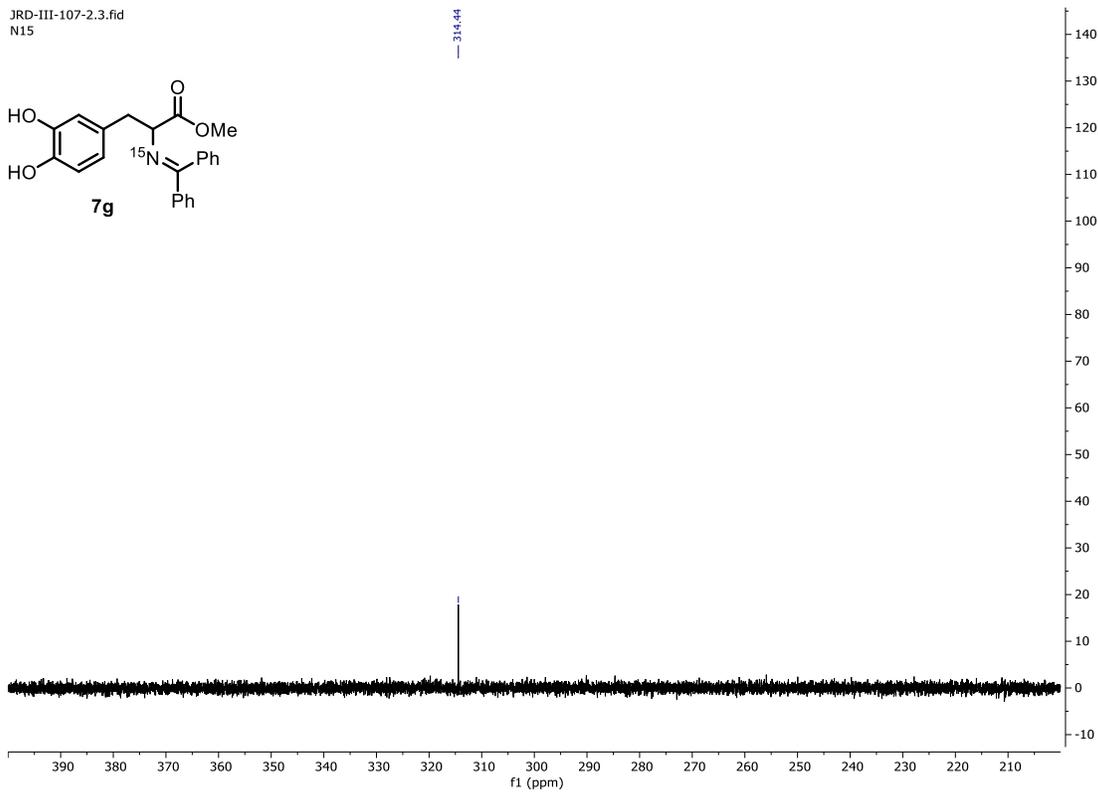
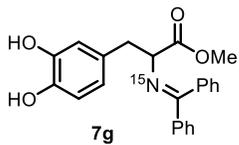
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Proton



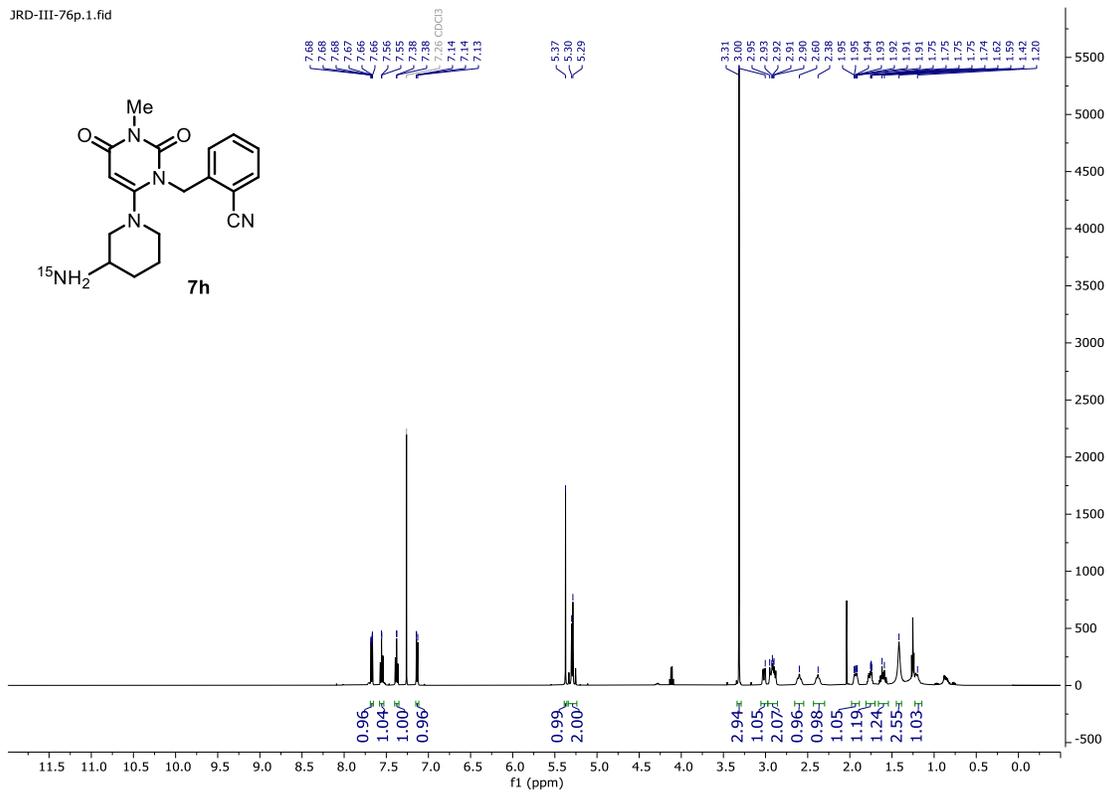
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Carbon CPD decoupled



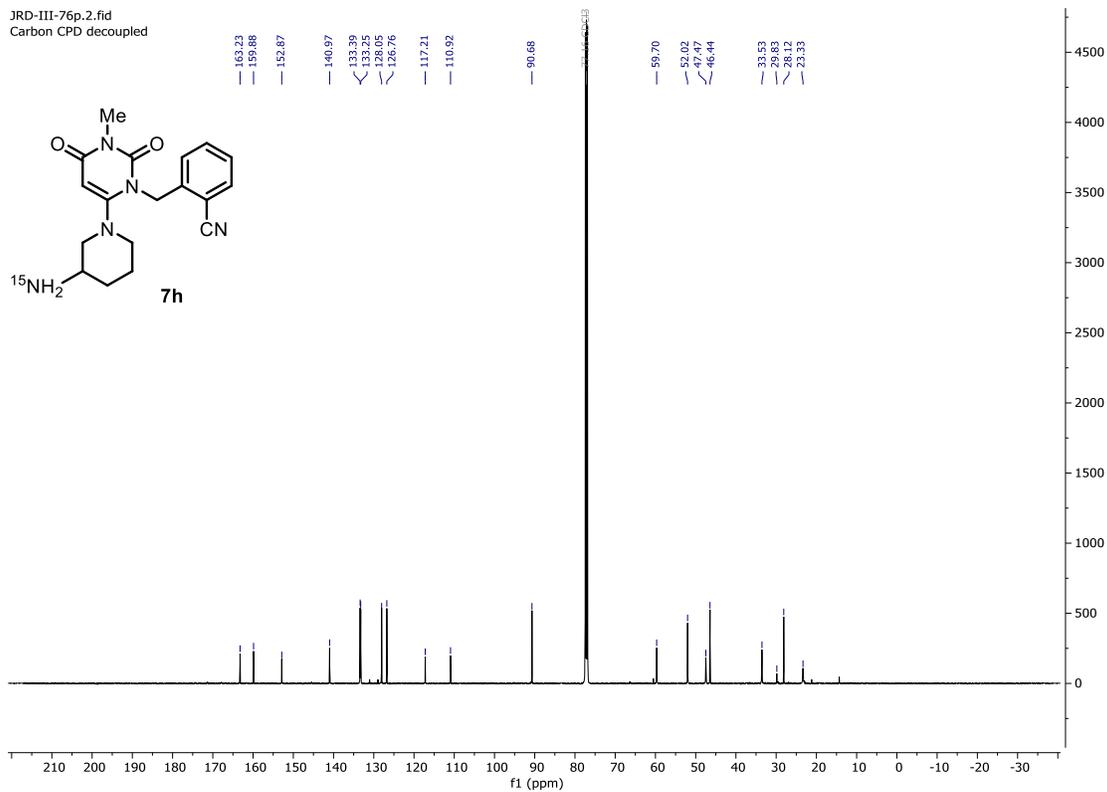
JRD-III-107-2.3.fid
N15



JRD-III-76p.1.fid



JRD-III-76p.2.fid
Carbon CPD decoupled



JRD-III-76p.3.fid
N15

— 33.95

