Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information

Photocatalyzed intermolecular amination for the synthesis of hydrazonamides

Wei Guo,*† Liuhuan Cai,† Zhen Xie, Weijie Mei, Gongping Liu, Ling Deng, Xiaoya Zhuo, Yumei Zhong, Xiaoying Zou, Lvyin Zheng*, Xiaolin Fan*

Key Laboratory of Organo-pharmaceutical Chemistry of Jiangxi Province, Gannan Normal University, Ganzhou 341000, China

†These authors contributed equally: Wei Guo, Liuhuan Cai.

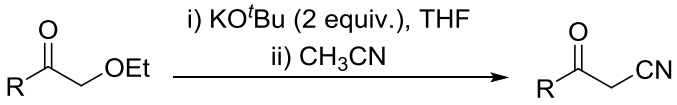
Correspondence and requests for materials should be addressed to W.G. (email: guoweigw@126.com) /or to L.Z (zhenglvyin@126.com) /or to X.F (fanxl2008@gnnu.edu.cn)

Table of Contents **Pages**

A. General Methods

Melting points were investigated using a melting point instrument and are uncorrected. IR spectra were obtained with an infrared spectrometer on either potassium bromide pellets or liquid films between two potassium bromide pellets. ^{1}H and ^{13}C NMR spectra were carried out on a 400 MHz NMR spectrometer. GC−MS data were obtained using electron ionization. HRMS data were collected from a high-resolution mass spectrometer (LCMS-IT-TOF). The reaction was performed on the photoreaction instrument (WP-TEC-1020L, WATTCAS, China) using a condenser system. The distance from the light source to the irradiation vessel is 5 mm. TLC was used on commercially available 100−400 mesh silica gel plates (GF254). Starting materials including **1-34**, **1-35**, **1-36**, **1-37**, **2-39**, **2-40**, **2-42**, **2-43**, **2-45**, **2-48**, **2-50**, **2-52**, **2-54**, **2-59**, **2-60**, **76**, **78** and **84** were synthesized in our laboratory, and other materials were purchased commercially. Unless otherwise noted, all purchased chemicals were used without further purification.

B. General Procedure for the Synthesis of Benzoylacetonitrile Derivatives (1-35, 1-36, 1-37, 1-38, 76)[1]

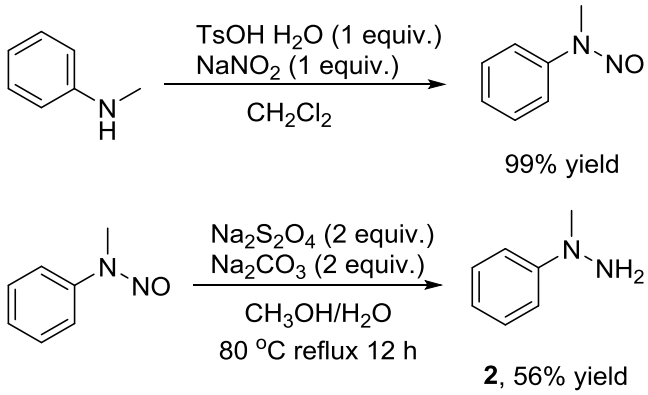


Ethyl Esters

To a flame-dried 50 mL round-bottom flask was charged with ethyl esters (3 mmol, 1 equiv.), potassium tertbutoxide (0.67 g, 6.00 mmol) and THF (15 mL) with stirring (about 230 rpm) at ambient temperature for 5 min. Then CH_3CN (3 mmol, 1 equiv) was added to the reaction system. The resulting mixture was stirred at ambient temperature for 1 h. The reaction was quenched with water (30 mL). After adding ethyl acetate (40 mL) and then HCl solution (1 mL, 12 M), the combined organic phase was dried with Na_2SO_4, filtered, and then concentrated in vacuo. The residue was purified by flash chromatography on silica gel to afford the corresponding products (using the mixture of petroleum ether and ethyl acetate (v/v = 10:1) as an

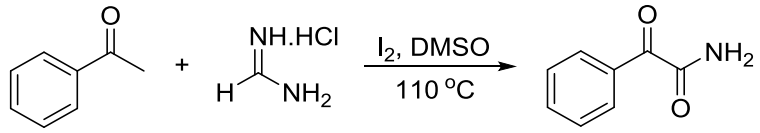
eluent).

C. General Procedure for the Synthesis of *N*,*N*-Disubstituted hydrazines (2, 2-39, 2-40, 2-42, 2-43, 2-45, 2-48, 2-50, 2-52, 2-54, 2-59, 2-60, 78)[2]



Step 1: To a flame-dried 50 mL round-bottom flask was charged with *N*-methylaniline (0.749 g, 7.0 mmol), *p*-toluenesulfonic acid (1.33 g, 7.0 mmol), $NaNO_2$ (0.483 g, 7.0 mmol) and CH_2Cl_2 (10 mL) with stirring (about 230 rpm) at room temperature for 5~10 min. The completion of the reaction was monitored by TLC. After completing the reaction, the insoluble solid was removed by filtration, and the filtrate was concentrated in vacuo to afford *N*-methyl-*N*-phenylnitrous amide (0.96 g, 99%) as a pale yellow powder; **Step 2**: To a flame-dried 100 ml round bottom flask was charged with *N*-methyl-*N*-phenylnitrous amide (0.96 g, 7.0 mmol), $Na_2S_2O_4$ (2.436 g, 14.0 mmol), Na_2CO_3 (1.484 g, 14.0 mmol), methanol (15 mL) and H_2O (15 mL) at 80 ^oC 12 h. After completing the reaction, 50 mL water was added to the reaction system. Then the mixture was extracted with EtOAc (3 × 50 mL), dried over anhydrous Na_2SO_4 and concentrated in vacuo. The crude product was purified by column chromatography on silica gel (eluent: petroleum ether/ethyl acetate = 5:1) to afford the product 1-methyl-1-phenylhydrazine **2** (0.48 g, 56% yield).

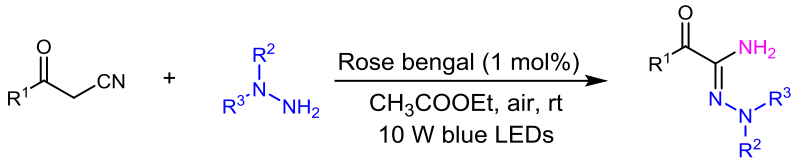
D. Synthesis of 84[3]



84, 85%

In a flame-dried test tube with a stir bar, acetophenone (0.12 g, 1.0 mmol), formamidine hydrochloride (0.08 g, 1.0 mmol), I_2 (0.20 g, 0.8 mmol) were added to DMSO (2 mL) at 110 ^{o}C for 12 h. After evaporating the solvent, the crude residue was purified by chromatography on a silica gel for separation with petroleum ether/ethyl acetate (5:1, v/v) as an eluent to produce the desired product **84** (0.126 g, 85% yield).

E. General Procedure for the Synthesis of Hydrazonamides



In a flame-dried test tube with a stir bar, β-ketonitriles (0.1 mmol), *N*,*N*-disubstituted hydrazines (0.3 mmol), Rose bengal (1 mg, 1 mol%) were added to $CH_3COOC_2H_5$ (1 mL, 0.1 M). The reaction system was performed on the photoreaction apparatus (WP-TEC-1020L, WATTCAS, China) with a condenser system. The distance from the light source to the irradiation vessel is 5 mm. The mixture was stirred at room temperature under the irradiation of 10 W (465 nm) blue LEDs for 12 h. After evaporating the solvent, the crude residue was purified by chromatography on a silica gel for separation using petroleum ether/ethyl acetate (20:1, v/v) as an eluent to produce the desired product.

F. Gram-Scale Reaction Synthesis of 3 under the Irradiation of Sunlight

Figure S1. Gram-scale synthesis of **3** under sunlight irradiation

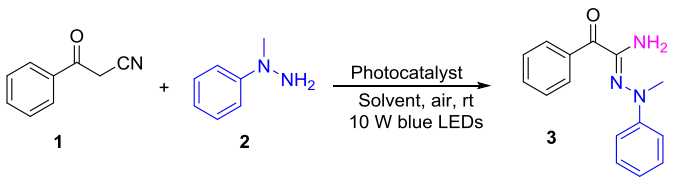
In a 100 mL round-bottomed flask with a stir bar, benzoylacetonitrile **1** (1.015 g, 7 mmol), 1-methyl-1-phenylhydrazine **2** (2.562 g, 21 mmol), rose bengal (0.05 g, 1 mol%) were added in $CH_3COOC_2H_5$ (60 mL) under air atmosphere, and then the mixture was stirred under sunlight for 10 h (7:00~17:00). After completing the reaction, solvents were removed under reduced pressure and the crude product was purified by column chromatography on silica gel column using petroleum ether/ethyl acetate (20:1, v/v) as an eluent to afford the desired orange solid product **3** (0.903 g, 51% yield).

G. Optimization of Reaction Conditions

In order to confirm our working hypothesis, the photocatalyzed reaction of 3-oxo-3-phenylpropanenitrile **1** with 1-methyl-1-phenylhydrazine **2** was investigated using $Ru(bpy)_3Cl_2$ (1 mol%) as photocatalyst, in the presence of CH_3CN solvent under a 10 W blue LED irradiation at room temperature (Table S1). Gratifyingly, the desired product **3** was obtained in 53% isolated yield (Table S1, entry 1). The structural of **3** was further unambiguously confirmed by X-ray crystal diffraction measurement (see Figure S12, Supporting Information). Then, screening of other photocatalysts, such as *fac*-$Ir(ppy)_3$, Esoin B, Eosin Y, and Rose bengal indicated that Rose bengal was the best, providing the product **3** in 68% yield (Table S1, entries 2-5). Conducting the reaction with THF or CH_3COOEt as solvents afforded satisfactory

results (Table S1, entries 6-7). Other solvents including CH_2Cl_2, DMSO, DMA, DMF, CH_3CH_2OH, 1,4-dioxane and H_2O resulted into the inferior yields (Table S1, entries 8-14). Control experiments showed that Rose bengal and light sources are dispensable for the reaction (Table S1, entries 15-16). Even if the reaction was performed at 80 ^oC in the dark, only a trace amount of product **3** was observed (Table S1, entry 17). Upon treatment of the conversation under N_2 atmosphere, the yield decreased obviously (Table S1, entry 18). Further experiment results revealed that O_2 in air should be crucial to the transformation (Table S1, entry 19). Other light sources were also screened, giving the moderate to good yields (Table S1, entries 20-22). Especially, gram-scale reaction worked well under the irradiation of sunlight, delivering the desired product **3** in 51% yield (see Figure S2). After investigating a series of variables, including photocatalysts, solvents, light sources, the amount of substrates and reaction time (see Tables S2-S5), we established the optimal reaction conditions [Rose bengal (1 mol%) in 1.0 mL CH_3COOEt under a 10 W Blue LED (465 nm) at room temperature for 12 h] (Table S1, entry 7).

Table S1. Optimization of Reaction Conditions[a]

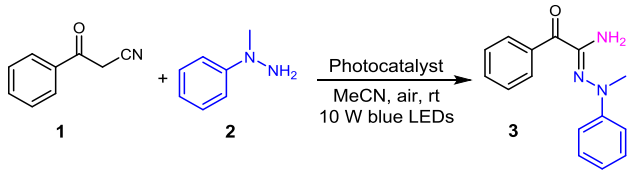


Entry	Catalyst	Solvent	Light (nm)	Yield (%)[b]
1	$Ru(bpy)_3Cl_2$	MeCN	465	53
2	*fac*-$Ir(ppy)_3$	MeCN	465	41
3	Eosin B	MeCN	465	38
4	Eosin Y	MeCN	465	28
5	Rose bengal	MeCN	465	68
6	Rose bengal	THF	465	83
7	Rose bengal	CH_3COOEt	465	80
8	Rose bengal	CH_2Cl_2	465	60
9	Rose bengal	DMSO	465	51
10	Rose bengal	DMA	465	68
11	Rose bengal	DMF	465	62
12	Rose bengal	CH_3CH_2OH	465	27

13	Rose bengal	1,4-dioxane	465	64
14	Rose bengal	H_2O	465	trace
15[c]	-	CH_3COOEt	465	trace
16[d]	Rose bengal	CH_3COOEt	-	0
17[e]	Rose bengal	CH_3COOEt	-	trace
18[f]	Rose bengal	CH_3COOEt	465	34
19[g]	Rose bengal	CH_3COOEt	465	78
20	Rose bengal	CH_3COOEt	365	62
21	Rose bengal	CH_3COOEt	520	79
22	Rose bengal	CH_3COOEt	Sunlight	79

[a]Reaction conditions: **1** (0.1 mmol), **2** (0.3 mmol) and solvent (1 mL), under a 10 W Blue LED at room temperature for 12 h. [b]isolated yield. [c]no photocatalyst.[d]in the dark. [e]in the dark at 80 °C. [f]Reaction under N_2 atmosphere. [g]Reaction under O_2 atmosphere.

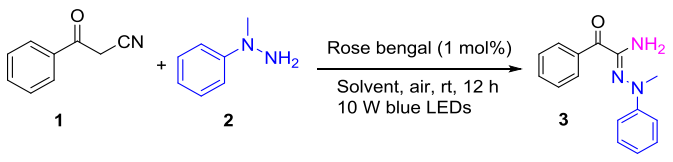
Table S2. Optimization of Photocatalysts.[a]



Entry	Catalyst	Solvent	Light source (nm)	Yield(%)[b]
1	$Ru(bpy)_3Cl_2$	MeCN	465	53
2	*fac*-$Ir(ppy)_3$	MeCN	465	41
3	Eosin B	MeCN	465	38
4	Eosin Y	MeCN	465	28
5	Rose bengal	MeCN	465	68
6	/	MeCN	465	trace

[a]Reaction conditions: Unless otherwise noted, all reactions were carried out with **1** (0.1mmol), **2** (0.3 mmol) and photocatalyst (1 mol%) in MeCN (1 mL), irradiation under a 10 W blue LEDs at room temperature for 12 h in the open air. [b]Isolated yield.

Table S3. Optimization of Solvents.[a]

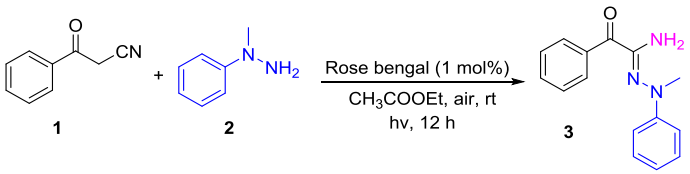


Entry	Catalyst	Solvent	Light source (nm)	Yield (%)[b]
1	Rose bengal	MeCN	465	68
2	Rose bengal	CH_2Cl_2	465	60
3	Rose bengal	CH_3CH_2OH	465	27
4	Rose bengal	THF	465	83

5	Rose bengal	DMSO	465	51
6	Rose bengal	DMA	465	68
7	Rose bengal	DMF	465	62
8	Rose bengal	CH_3COOEt	465	80
9	Rose bengal	1,4-dioxane	465	64
10	Rose bengal	H_2O	465	trace

[a]Reaction conditions: Unless otherwise noted, all reactions were carried out with **1** (0.1mmol), **2** (0.3 mmol) and Rose bengal (1 mol%) in solvent (1 mL), irradiation under a 10 W blue LEDs at room temperature for 12 h in the open air. [b]Isolated yield.

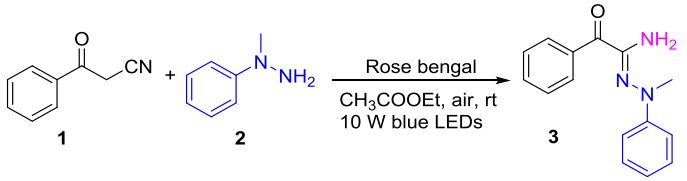
Table S4. Optimization of Light Sources.[a]



Entry	catalyst	solvent	Light source (nm)	Yield(%)[b]
1	Rose bengal	CH_3COOEt	365	62
2	Rose bengal	CH_3COOEt	435	75
3	Rose bengal	CH_3COOEt	465	80
4	Rose bengal	CH_3COOEt	520	79
5	Rose bengal	CH_3COOEt	Sunlight	79
6	Rose bengal	CH_3COOEt	/	n.d.

[a]Reaction conditions: Unless otherwise noted, all reactions were carried out with **1** (0.1 mmol), **2** (0.3 mmol) and Rose bengal (1 mol%) in CH_3COOEt (1 mL), irradiation under different light sources at room temperature for 12 h in the open air. [b]Isolated yield.

Table S5. Optimization of the Amount of Substrates and Reaction Time.[a]

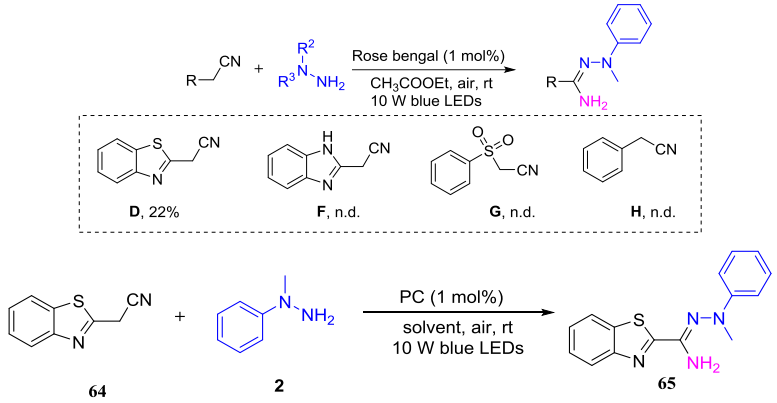


Entry	Mole Ratio of **1** and **2**	Time (h)	Yield(%)[b]
1	1:0.5	12	20%
2	1:1	12	33%
3	1:1.5	12	49%
4	1:2	12	64%
5	1:2.5	12	77%
5	1:3	12	80%
6	1:4	12	77%

7	1:3	6	70%
8	1:3	18	81%
9	1:3	24	81%

[a]Reaction conditions: Unless otherwise noted, all reactions were carried out with **1** (0.1 mmol), **2** (0.05-0.4 mol) and Rose bengal (1 mol%) in CH_3COOEt (1 mL), irradiation under a 10 W LEDs at room temperature in the open air. [b]Isolated yield.

Table S6. Screening of Reaction Conditions for Synthetic Applications[a]



Entry	Catalyst	Solvent	Yield (%)[b]
1	Rose bengal	CH_3COOEt	22
2	Eosin Y	CH_3COOEt	10
3	Eosin B	CH_3COOEt	10
4	*fac*-Ir(ppy)$_3$	CH_3COOEt	14
5	Ru(bpy)$_3$ Cl_2	CH_3COOEt	11
6	Rose bengal	MeCN	18
7	Rose bengal	THF	11
8	Rose bengal	CH_2Cl_2	31
9	Rose bengal	DMSO	30
10	Rose bengal	DMF	35
11	Rose bengal	CH_3CH_2OH	50
12	Rose bengal	CH_3COOEt/DMSO (1/1)	61
13	Rose bengal	CH_3COOEt/CH_3CH_2OH (1/1)	43
14	Rose bengal	CH_3CH_2OH/DMSO (1/1)	72
15[c]	/	CH_3CH_2OH/DMSO (1/1)	Trace
16[d]	Rose bengal	CH_3CH_2OH/DMSO (1/1)	n.d.

[a]Reaction conditions: **64** (0.1 mmol), **2** (0.3 mmol) and solvent (1 mL), under a 10 W Blue LED at room temperature. [b]isolated yield. [c]no photocatalyst. [d]in the dark.

To further expand the synthetic application research of the reaction, we investigated other substituted acetonitriles including 2-(benzo[*d*]thiazol-2-yl)acetonitrile (**D**), 2-(1*H*-benzo[*d*]imidazol-2-yl)acetonitrile (**F**), 2-(phenylsulfonyl)acetonitrile (**G**) and 2-phenylacetonitrile (**H**) under the standard conditions (Table S6). The preliminary experimental results showed that 2-(benzo[*d*]thiazol-2-yl)acetonitrile (**D**) was feasible.

Then, the optimal reaction conditions were discovered by screening photocatalysts and solvents (Table S6). The reaction using Rose bengal as photocatalyst, CH_3CH_2OH/DMSO (*v*/*v* = 1/1) as mixture solvent afforded the best results (Table S6, entry 14).

H. HRMS of 25

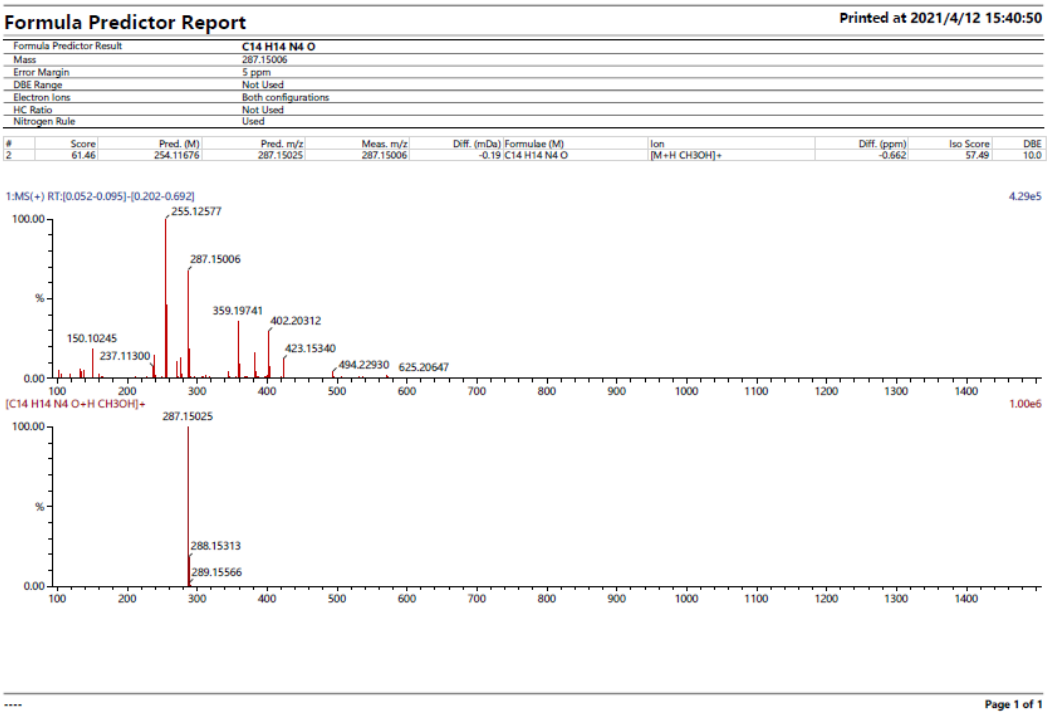


Figure S2. HRMS of 25

HRMS (ESI) calcd for $C_{14}H_{14}N_4O$ [M + H CH_3OH]$^+$ *m/z* 287.1503; found *m/z* 287.1501.

I. Data of Isotope Labeling 76-79

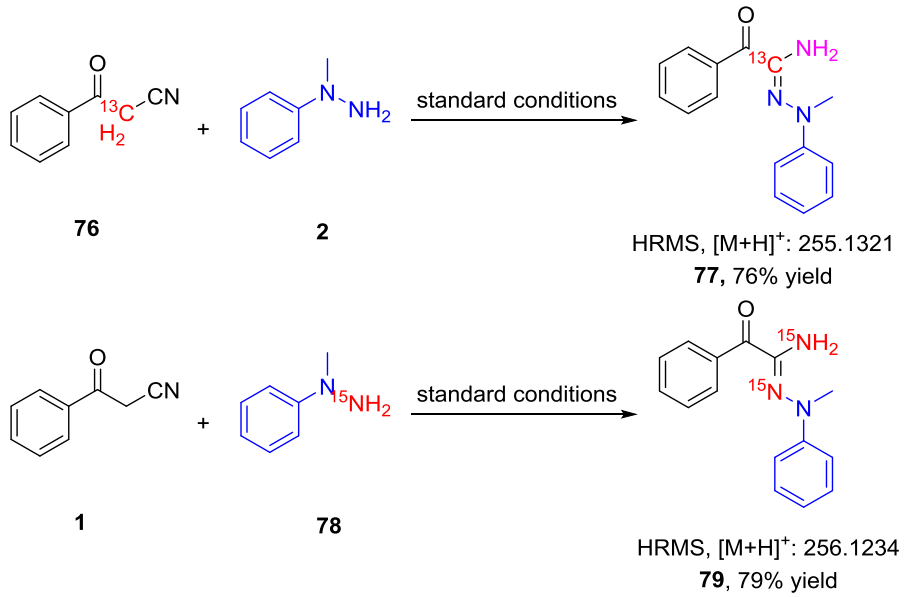
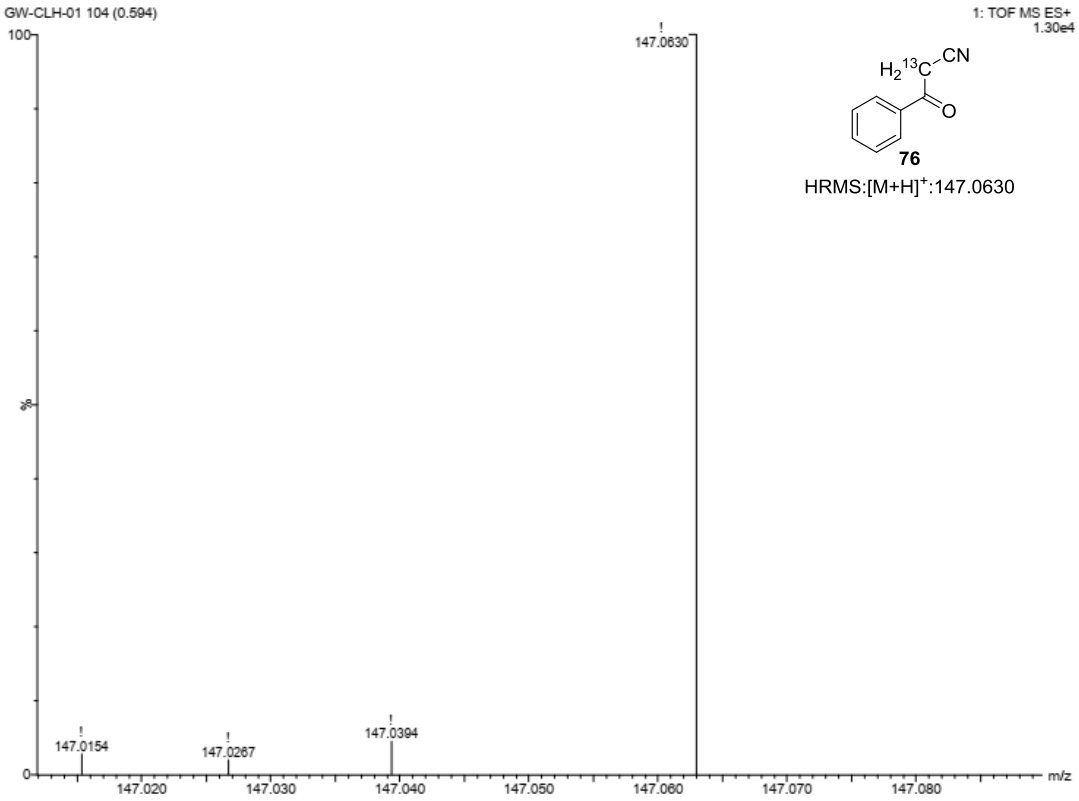
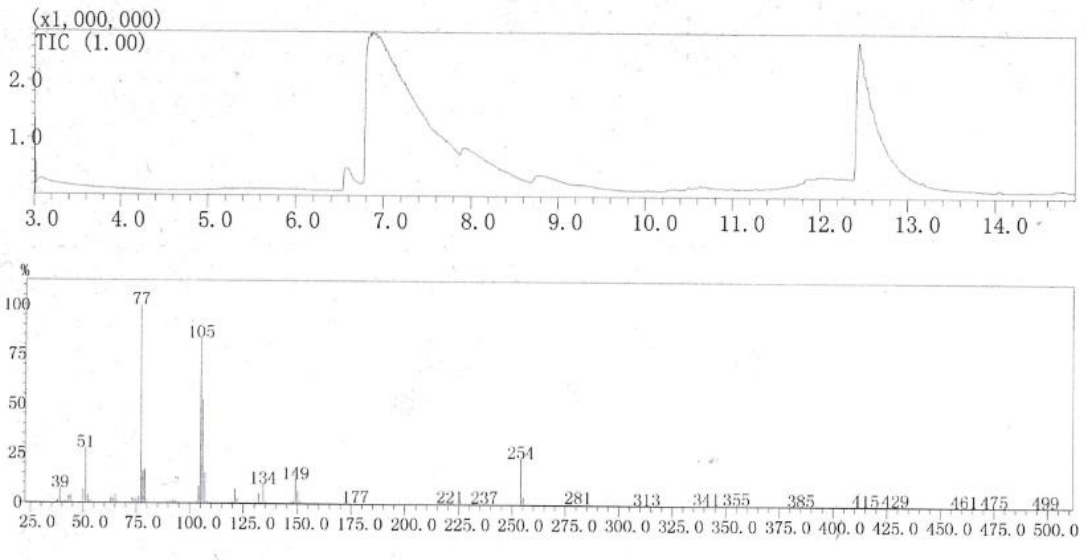


Figure S3. HRMS of ^{13}C-76[a]



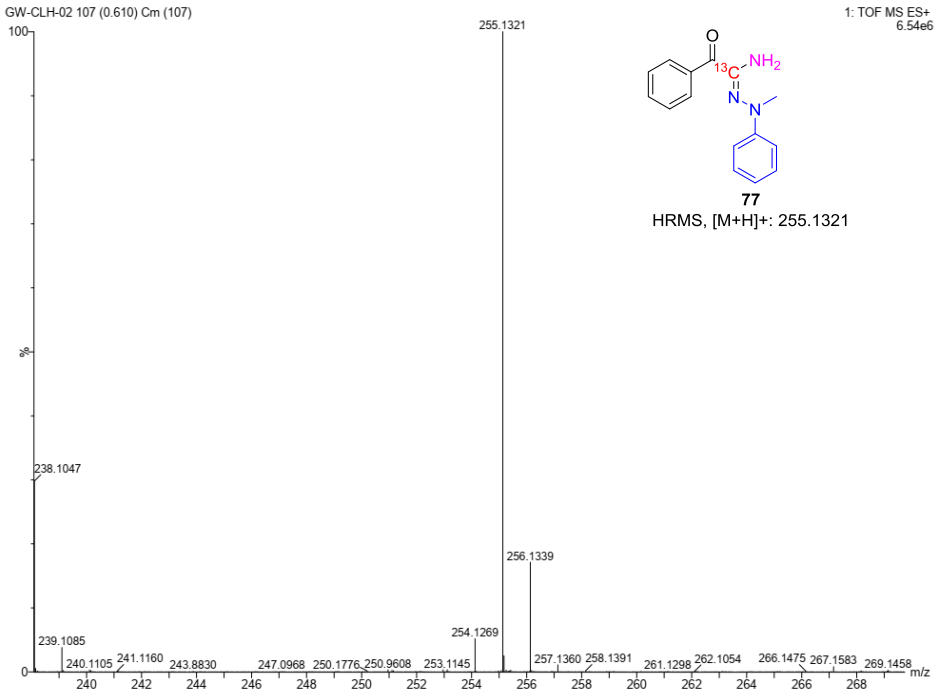
[a]**^{13}C-76**: HRMS (ESI) calc. $C_8{}^{13}CH_8NO$ $[M + H]^+$: 147.0639, found: 147.0630.

Figure S4. GC-MS of ^{13}C-77[a]



[a]RT = 12.45 min, **^{13}C-77**: MS (EI, 70 eV) m/z: 254.

Figure S5. HR-MS of ^{13}C-77[a]



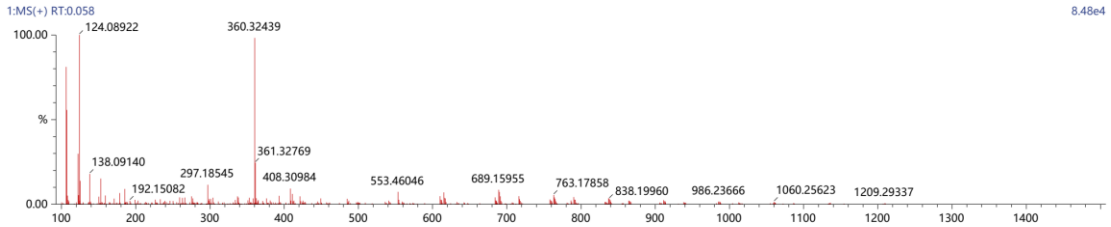
[a]**^{13}C-77**: HRMS (ESI) calc. $C_{14}{}^{13}CH_{16}N_3O$ $[M + H]^+$: 254.1327, found: 254.1321.

Figure S6. HR-MS of ^{15}N-78[a]

Formula Predictor Report Printed at 2021/1/26 13:40:38

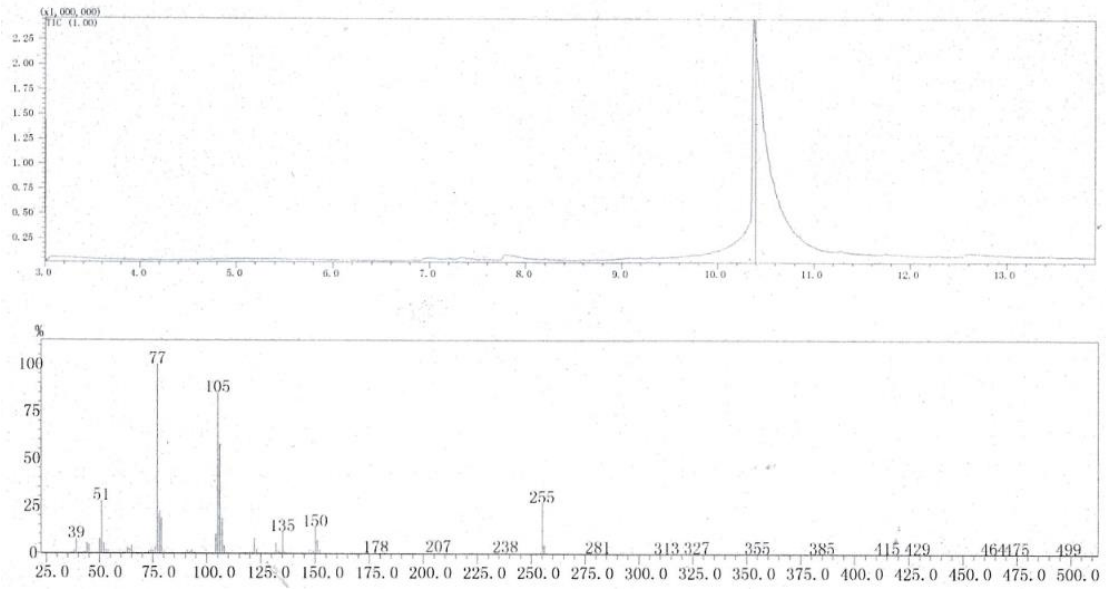
Formula Predictor Result	**C7 H10 N 15N**
Mass	124.08922
Error Margin	5 ppm
DBE Range	Not Used
Electron Ions	Both configurations
HC Ratio	Not Used
Nitrogen Rule	Used

#	Score	Pred. (M)	Pred. m/z	Meas. m/z	Diff. (mDa)	Formulae (M)	Ion	Diff. (ppm)	Iso Score	DBE
1	68.53	123.08143	124.08871	124.08922	0.51	C7 H10 N 15N	[M+H]+	4.110	69.06	3.5



[a]**^{15}N-78**: HRMS (ESI) calc. $C_7H_{11}N^{15}N$ $[M + H]^+$: 124.0887, found: 124.0892.

Figure S7. GC-MS of ^{15}N-79[a]



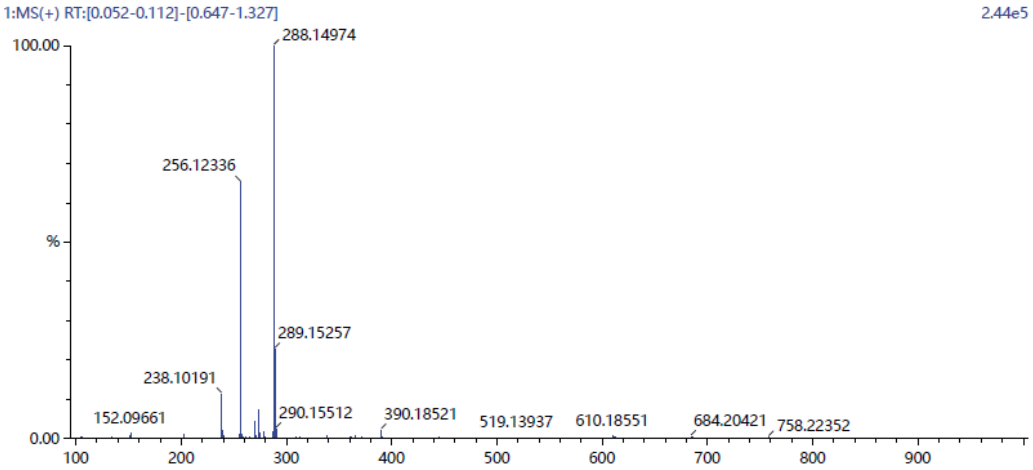
[a]RT = 10.45 min, **^{15}N-79**: MS (EI, 70 eV) m/z: 255

Figure S8. HR-MS of ^{15}N- 79[a]

Formula Predictor Report Printed at 2020/11/18 15:44:32

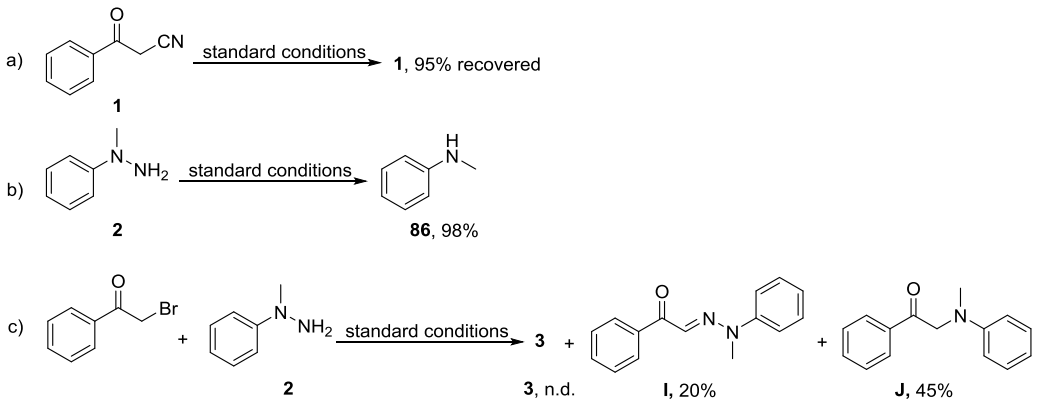
Formula Predictor Result	C15 H15 N O 15N2
Mass	256.12336
Error Margin	1000 ppm
DBE Range	Not Used
Electron Ions	Both configurations
HC Ratio	Not Used
Nitrogen Rule	Used

#	Score	Pred. (M)	Pred. m/z	Meas. m/z	Diff. (mDa)	Formulae (M)	Ion	Diff. (ppm)	Iso Score	DBE
1	94.50	255.11558	256.12286	256.12336	0.50	C15 H15 N O 15N2	[M+H]+	1.952	95.05	9.0



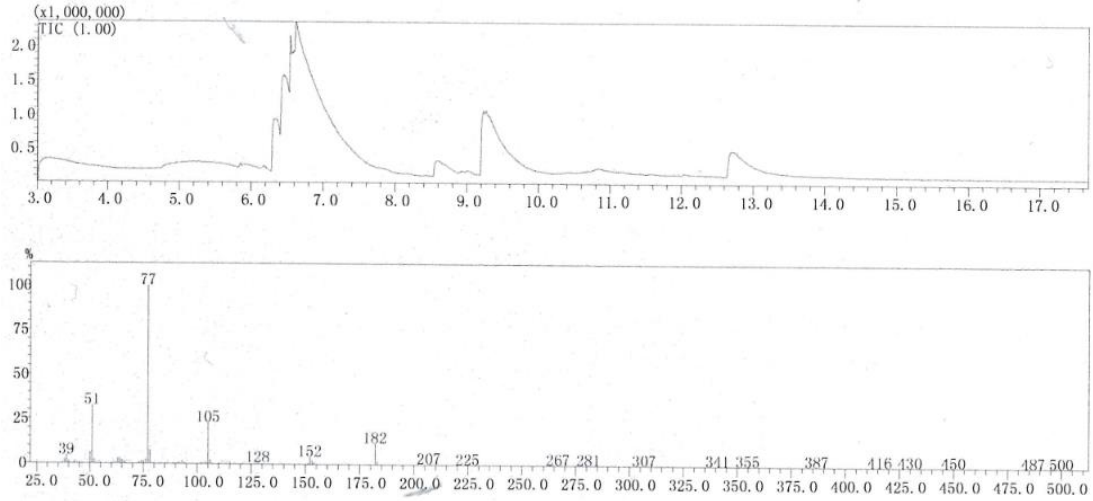
[a]**^{15}N-79**: HRMS (ESI) calc. $C_{15}H_{16}N^{15}N_2O$ $[M + H]^+$: 256.1229, found: 256.1234.

J. Control experiments



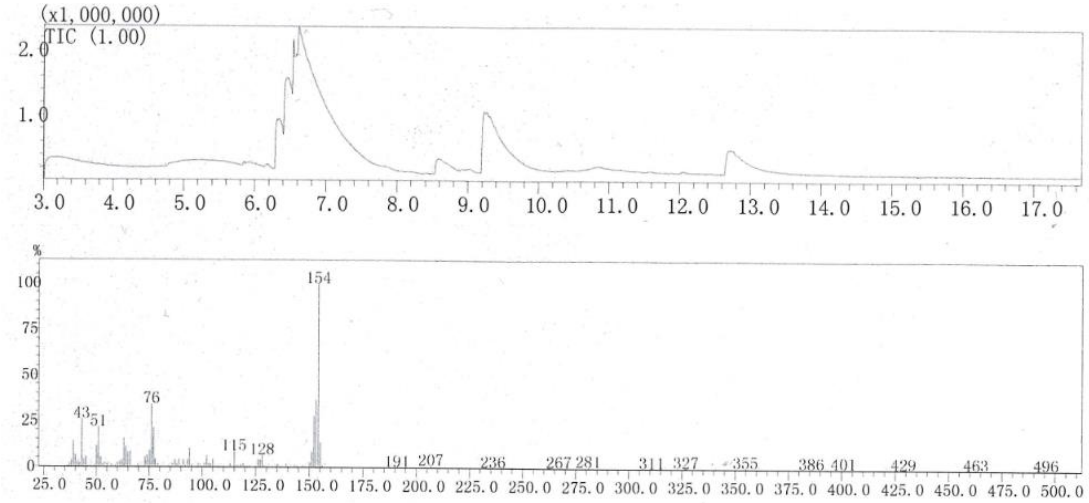
Scheme S1. Control experiments

Figure S9. GC-MS of 82[a]



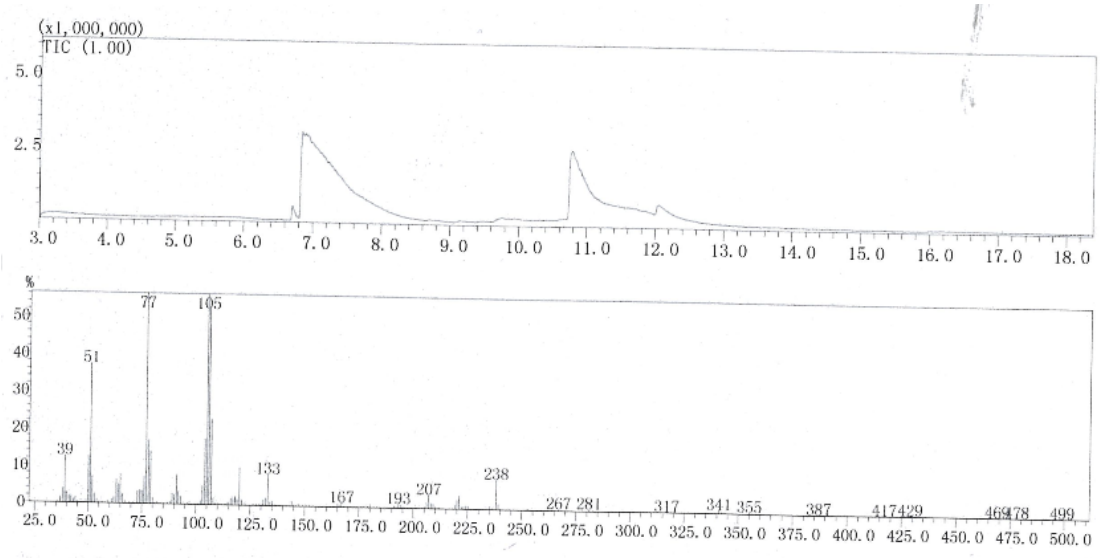
[a]RT = 12.8 min: **82**; MS (EI, 70 eV) m/z: 182.

Figure S10. GC-MS of 83[a]



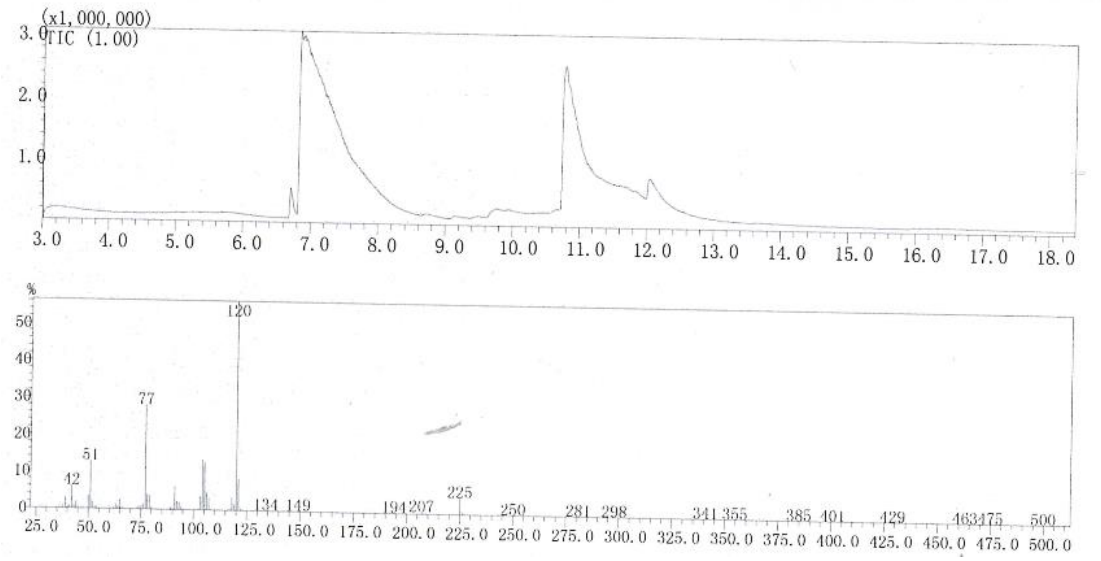
[a]RT = 8.6 min: compound **83**; MS (EI, 70 eV) m/z: 154.

Figure S11. GC-MS of I[a]



[a]RT = 12.0 min: compound **I**; MS (EI, 70 eV) m/z: 238.

Figure S12. GC-MS of J[a]



[a]RT = 11.0 min: compound **J**; MS (EI, 70 eV) m/z: 225.

K. The Crystal Structure of 3, 22, 34, 67

(1) The Crystal Structure of 3

The crystal growth procedure: Compound **3** (30 mg) was dissolved in 1 mL of EtOH. The mixed solution was evaporated slowly at room temperature to afford the crystals **3**.

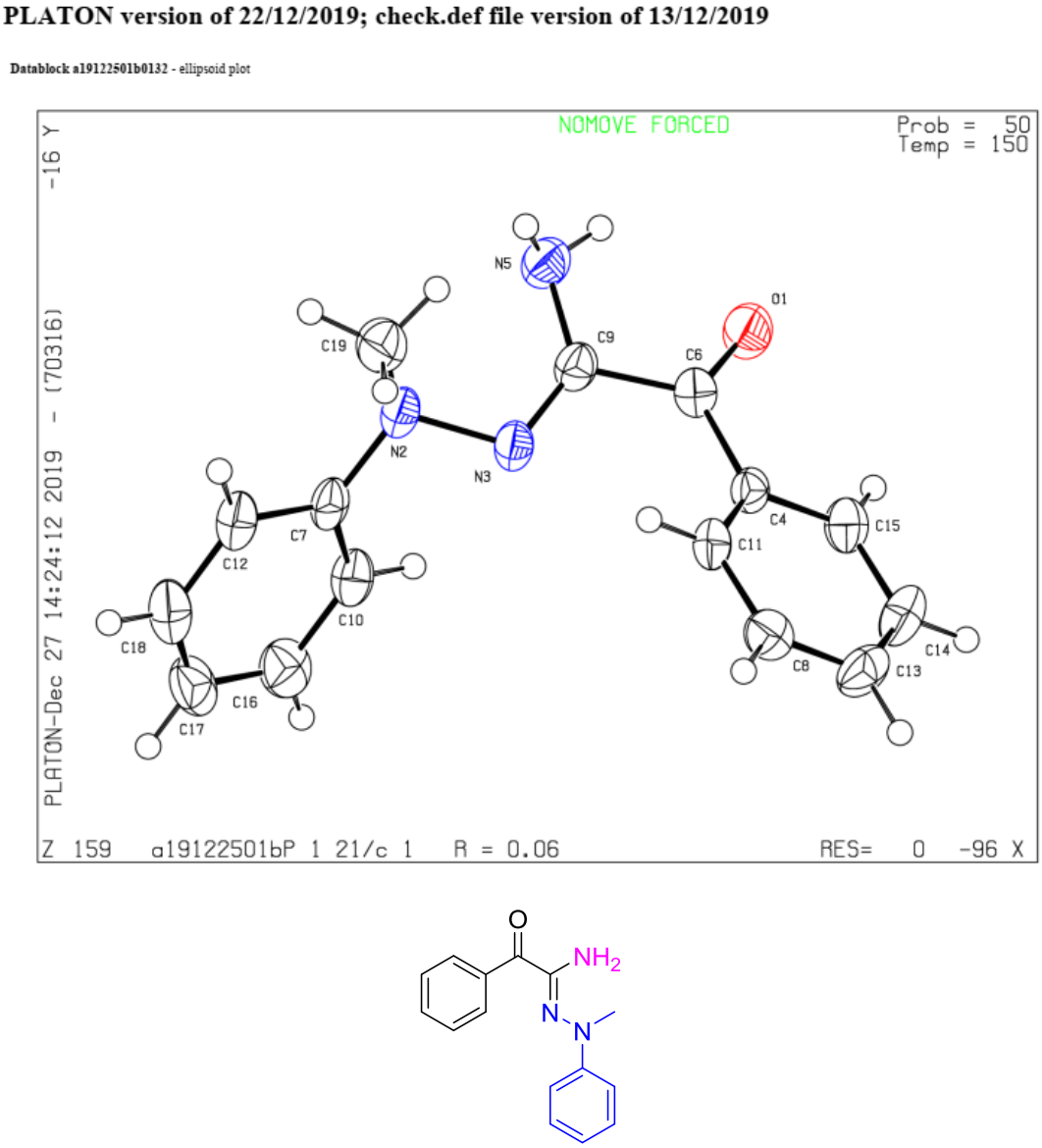


Figure S13. The Crystal Structure of 3

The CCDC number of **3** is 2049062, the detail information please see 3.cif document. Deposition Number: 2049062
(L. Cai, CCDC 2049062 (compound **3**). *CSD Commun.*, 2020. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre.)

(2) The Crystal Structure of 22

The crystal growth procedure: Compound **22** (25 mg) was dissolved in 1 mL of $CDCl_3$. The mixed solution was evaporated slowly at 8 ^{o}C to afford crystals **22**.

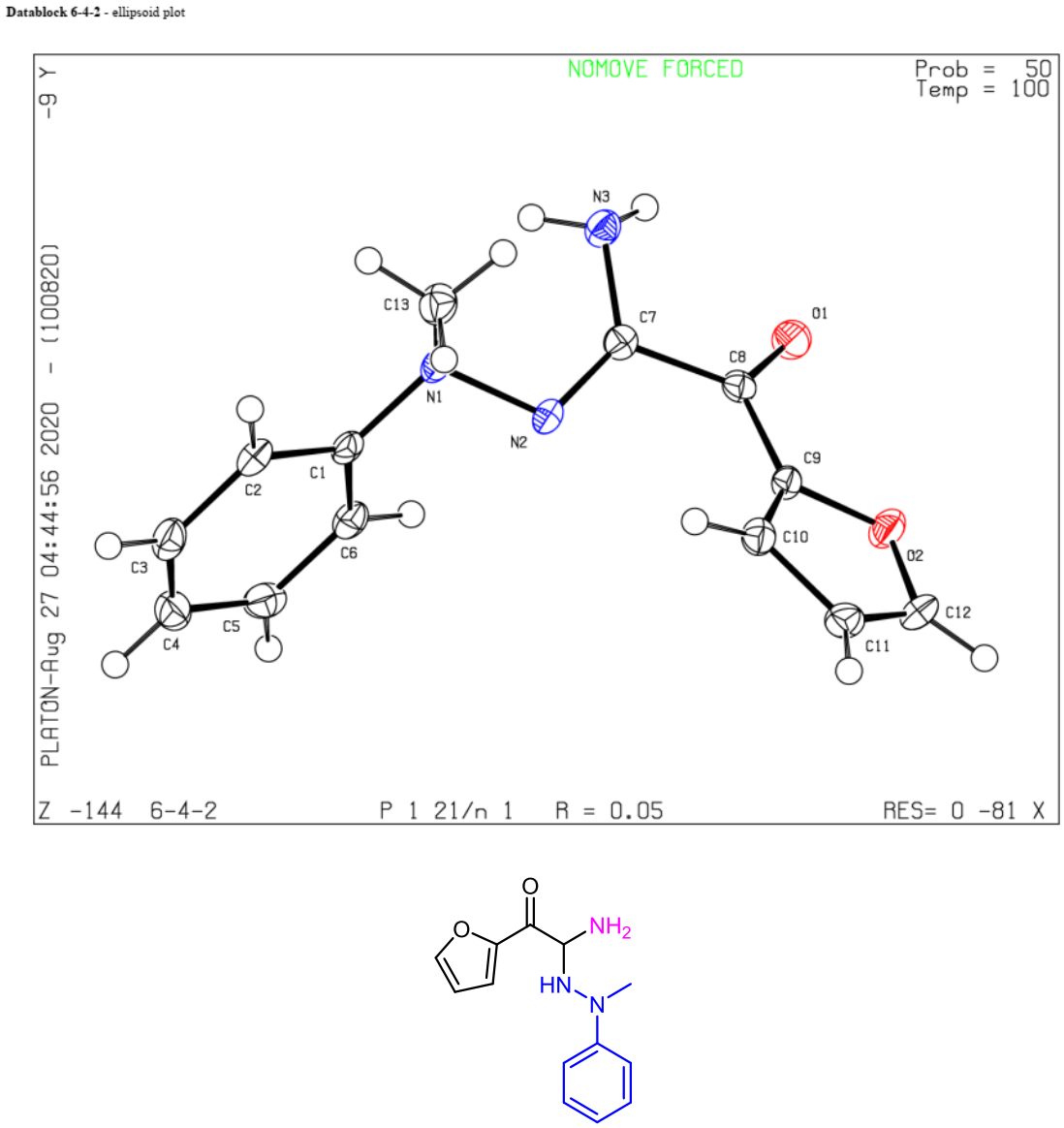


Figure S14. The Crystal Structure of 22

The CCDC number of **22** is 2049077, the detail information please see 22.cif document. Deposition Number: 2049077

(L. Cai, CCDC 2049077 (compound **22**). *CSD Commun.*, 2020. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre.)

(3) The Crystal Structure of 34

The crystal growth procedure: Compound **34** (25 mg) was dissolved in 1 mL of $CDCl_3$. The mixed solution was evaporated slowly at 8 ^oC to afford crystals **34**.

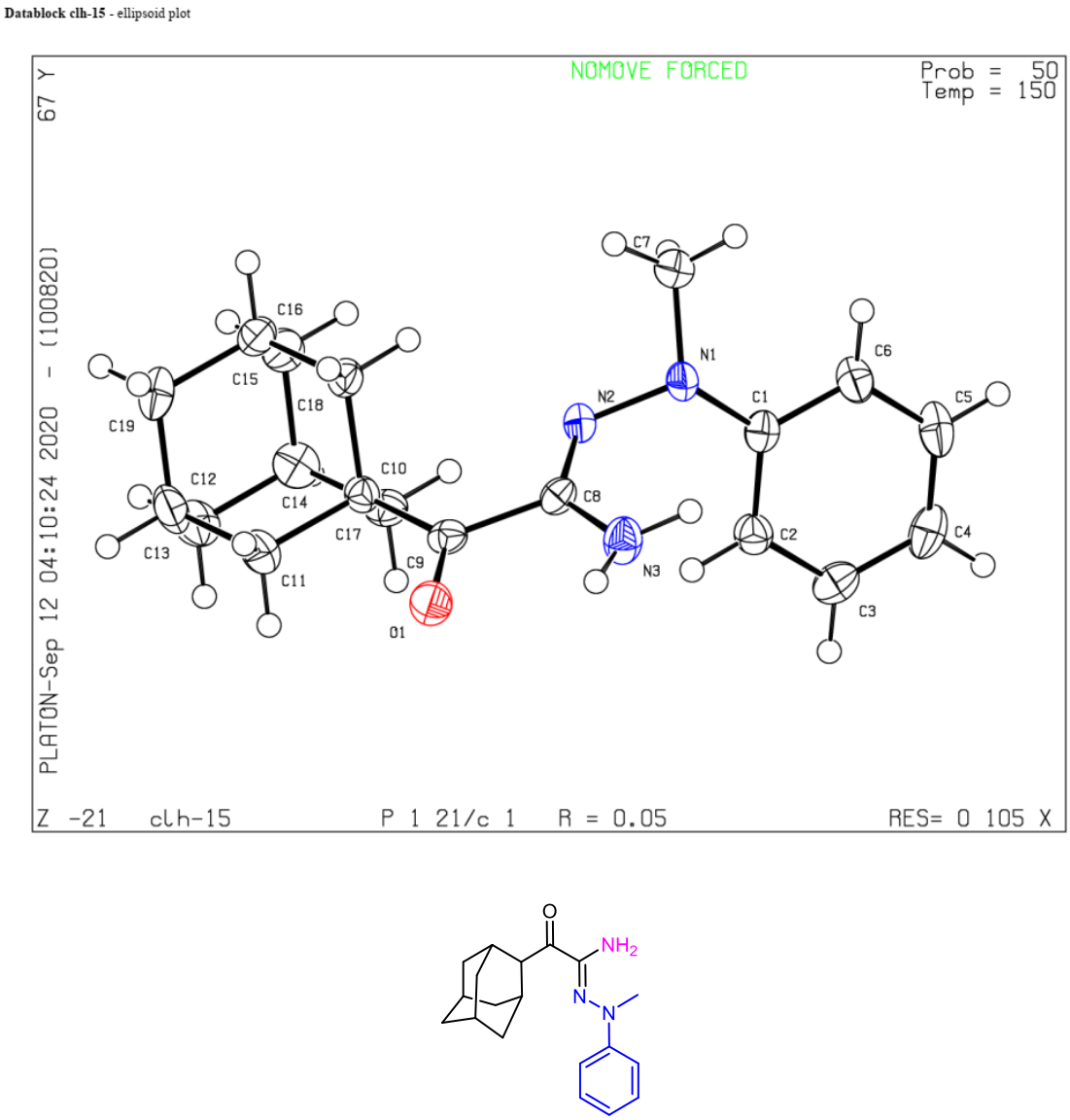


Figure S15. The Crystal Structure of 34

The CCDC number of **34** is 2049054, the detail information please see 34.cif document. Deposition Number: 2049054

(L. Cai, CCDC 2049054 (compound **34**). *CSD Commun.*, 2020. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre.)

(4) The Crystal Structure of 67

The crystal growth procedure: Compound **66** (25 mg) was dissolved in 1 mL of $CDCl_3$. The mixed solution was evaporated slowly at 8 ^{o}C to afford crystals **67**.

PLATON version of 18/09/2020; check.def file version of 20/08/2020

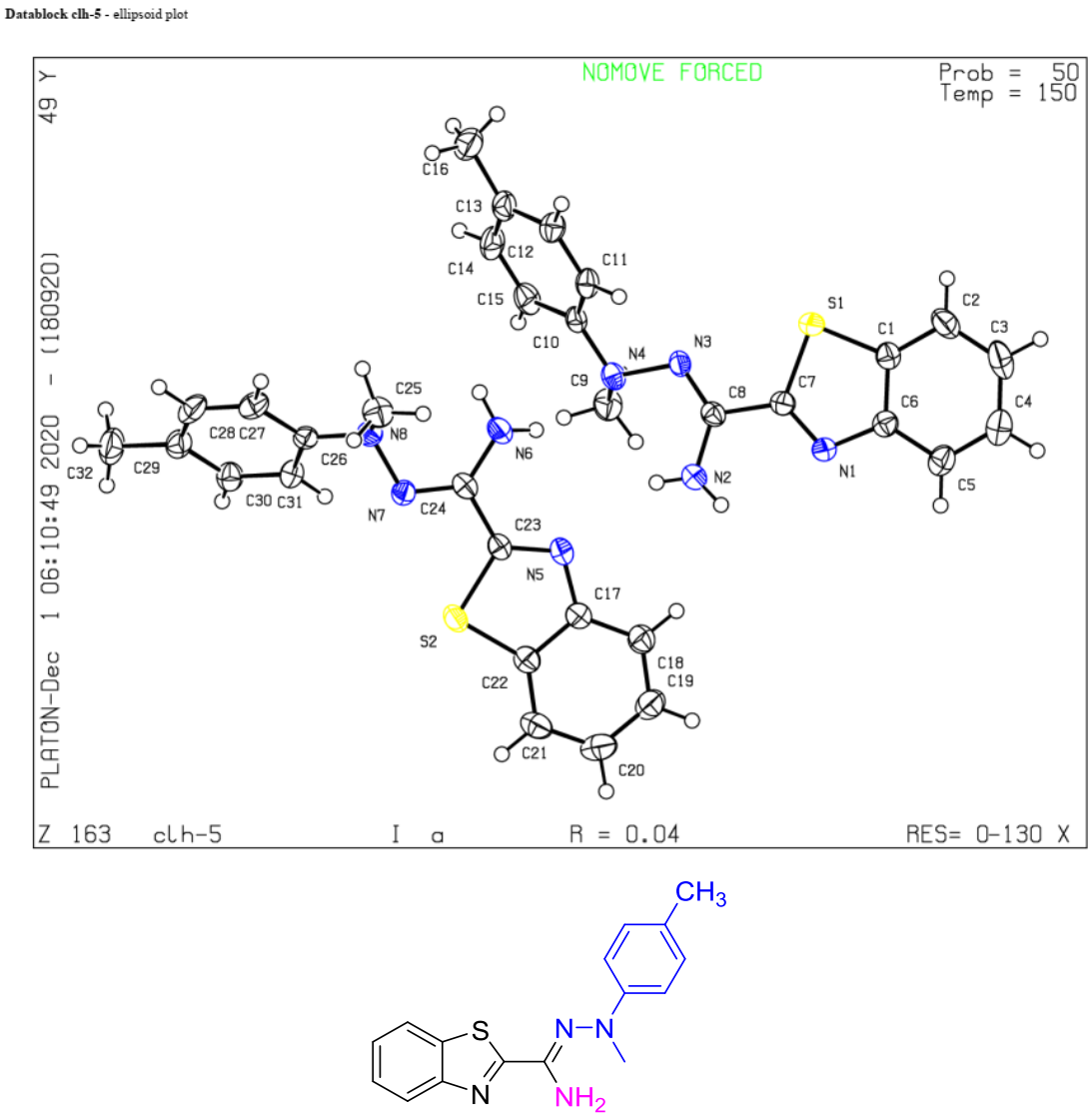


Figure S16. The Crystal Structure of 67

The CCDC number of **67** is 2049056, the detail information please see 66.cif document.
Deposition Number: 2049056
(L. Cai, CCDC 2049056 (compound **67**). *CSD Commun.*, 2020. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre.)

L. "CN^-" and HCN Detection

Figure S17. "CN^-" Detection

Methods: (a) Rinse the measuring vessel with the solution and fill it to the 5 ml mark; (b) Add 1 spoon Cyanide-1 (buffer solution) to the sample; (c) Add 5 drops Cyanide-2 to the sample; (d) Shake carefully; (e) Immediately dip the test strip into the sample; f: Wait 45 seconds; (f) Shake off excess liquid; (g) Compare within 10 seconds with the color scale.

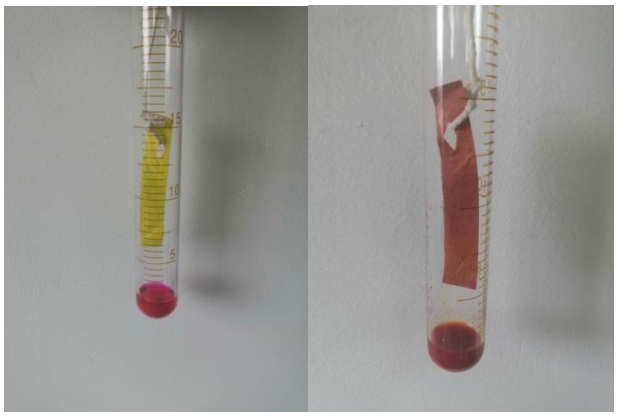
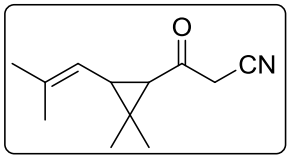


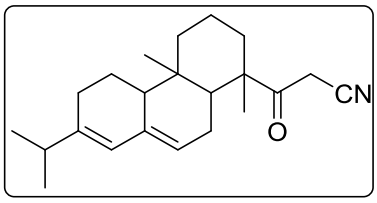
Figure S18. HCN detection

The release of HCN was detected using yellow picric acid (trinitrophenol) test strips as indictors. The color changes from yellow to red-brown, indicating the generation of HCN from the reaction system.

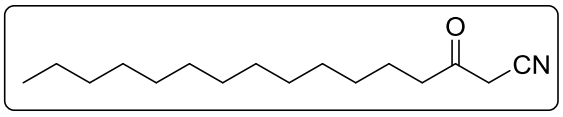
M. Characterization Data



3-(2,2-dimethyl-3-(2-methylprop-1-en-1-yl)cyclopropyl)-3-oxopropanenitrile **(1-35):** Yellow oil liquid: 80% yield (0.46 g); IR (KBr, cm^{-1}) 2919, 2256, 1704, 1379, 1232, 1104, 1051, 953, 845, 725; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 4.90 (d, J = 8 Hz, 1H), 3.56 (d, J = 4 Hz, 2H), 2.35 (m, 1H), 1.83 (d, J = 8 Hz, 1H), 1.70 (s, 3H), 1.66 (s, 3H), 1.22 (s, 3H), 1.19 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 194.7, 136.5, 120.1, 114.3, 42.5, 36.5, 35.4, 33.7, 25.6, 22.0, 19.7, 18.5; MS (EI, 70 eV) *m/z* 191, 175, 149, 136, 109; HRMS (ESI) calcd for $C_{12}H_{18}NO$ $[M + H]^+$ *m/z* 192.1388; found *m/z* 192.1388.

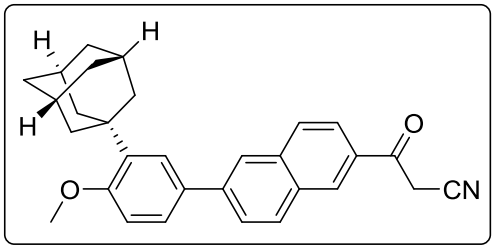


3-(7-isopropyl-1,4a-dimethyl-1,2,3,4,4a,4b,5,6,10,10a-decahydrophenanthren-1-yl)-3-oxopropanenitrile **(1-36):** Yellow oil liquid: 75% yield (0.73 g); IR (KBr, cm^{-1}) 2931,2259, 1712, 1625, 1461, 1305, 1217, 1035, 913, 829, 732, 647; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 5.74 (s, 1H), 5.31-5.30 (m, 1H), 3.66 (m, 2H), 2.24-2.17 (m, 1H), 2.08-2.02 (m, 2H), 1.96-1.76 (m, 5H), 1.65-1.52 (m, 7H), 1.25 (s, 3H), 0.99 (q, J = 4 Hz, 6H), 0.82 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 203.4, 145.6, 135.7, 122.2, 119.8, 114.6, 52.2, 51.0, 44.0, 38.0, 36.4, 34.9, 34.5, 27.9, 27.4, 25.5, 22.5, 21.4, 20.8, 17.8, 16.6, 14.1; MS (EI, 70 eV) *m/z* 325, 310, 255, 187, 131; HRMS (ESI) calcd for $C_{22}H_{32}NO$ $[M + H]^+$ *m/z* 326.2484; found *m/z* 326.2485.

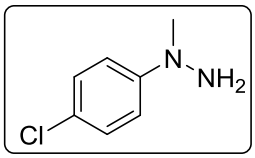


3-oxohexadecanenitrile **(1-37):** White solid: 85% yield (0.64 g); mp 70-73 ^{o}C. IR

(KBr, cm^{-1}) 2825, 2259, 1717, 1468, 1311, 1224, 1128, 1068, 930, 720; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 3.45 (s, 2H), 2.60 (t, J = 8 Hz, 2H), 1.65-1.57 (s, 2H), 1.24 (s, 20H), 0.87 (t, J = 8 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 197.7, 113.9, 42.2, 32.0, 32.0, 29.7, 29.7, 29.6, 29.6, 29.4, 29.4, 29.3, 28.9, 23.4, 22.7, 14.2; HRMS (ESI) calcd for $C_{16}H_{30}NO$ $[M + H]^+$ *m/z* 252.2327; found *m/z* 252.2325.

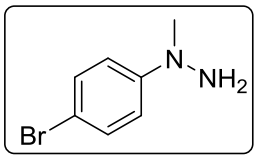


3-(6-(3-((3r,5r,7r)-Adamantan-1-yl)-4-methoxyphenyl)naphthalen-2-yl)-3-oxopropanenitrile **(1-38):** Yellow solid: 78% yield (1.02 g); mp 174-177 ^oC. IR (KBr, cm^{-1}) 3030, 2897, 2249, 1697, 1627, 1596, 1474, 1319, 1249, 1173, 1019, 913, 823, 731, 644; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.42 (s, 1H), 8.05-7.99 (m, 4H), 7.87 (dd, J = 8 Hz, 1H), 7.64 (d, J = 4 Hz, 1H), 4.24 (s, 2H), 3.94 (s, 3H), 2.22-2.21 (m, 6H), 2.14 (s, 3H), 1.84-1.82 (m, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.8, 159.2, 142.7, 139.1, 136.7, 132.0, 131.1, 131.0, 130.6, 130.1, 129.2, 127.2, 126.0, 125.8, 124.7, 123.8, 114.0, 112.1, 55.2, 40.6, 37.2, 37.1, 29.4, 29.1; HRMS (ESI) calcd for $C_{30}H_{30}NO_2$ $[M + H]^+$ *m/z* 436.2277; found *m/z* 436.2271.



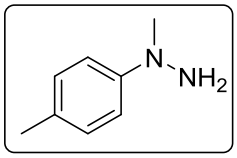
1-(4-Chlorophenyl)-1-methylhydrazine **(2-39)**[2]

Yellow liquid: 60% yield (0.66 g); IR (KBr, cm^{-1}) 3425, 3340, 2868, 1591, 1321, 1187, 1112, 1003, 883, 822, 612, 520; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.21-7.17 (m, 2H)，6.94-6.90 (m，2 H), 3.59 (s, 2H), 3.06 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 151.2, 128.7, 123.3, 114.8, 44.6; MS (EI, 70 eV) *m/z* 156, 141, 111, 105, 77; HRMS (ESI) calcd for $C_7H_{10}ClN_2$ $[M + H]^+$ *m/z* 157.0533; found *m/z* 157.0537.



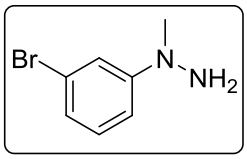
***1-(4-Bromophenyl)-1-methylhydrazine* (2-40)**[2]

Yellow liquid: 56% yield (0.79 g); IR (KBr, cm^{-1}) 3337, 2956, 2867, 2801, 1590, 1486, 1318, 1186, 1110, 1075, 996, 820, 750, 697, 585; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.31-7.29 (m, 2H), 6.85-6.83 (m, 2H), 3.64 (s, 2H), 3.03 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 151.6, 131.6, 115.2, 110.3, 44.4; MS (EI, 70 eV) *m/z* 156, 141, 111, 105, 77; MS (EI, 70 eV) *m/z* 200, 185, 155, 105, 77; HRMS (ESI) calcd for $C_7H_{10}BrN_2$ $[M + H]^+$ *m/z* 201.0027; found *m/z* 201.0022.



***1-Methyl-1-(p-tolyl)hydrazine* (2-42)**[2]

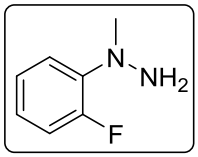
Yellow liquid: 60% yield (0.57 g); IR (KBr, cm^{-1}) 3411, 2920, 2862, 1617, 1512, 1453, 1316, 1261, 1185, 1107, 808, 682; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.15-7.11 (m, 2H), 6.99-6.95 (m, 2H), 3.61 (s, 2H), 3.10 (s, 3H), 2.34 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 150.9, 129.6, 128.2, 114.1, 45.1, 20.4; MS (EI, 70 eV) *m/z* 136, 121, 91, 77, 65; HRMS (ESI) calcd for $C_8H_{13}N_2$ $[M + H]^+$ *m/z* 137.1079; found *m/z* 137.1077.



***1-(3-Bromophenyl)-1-methylhydrazine* (2-43)**[2]

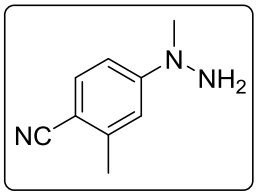
Yellow liquid: 58% yield (0.82 g); IR (KBr, cm^{-1}) 3338, 2919, 2853, 1587, 1477, 1332, 1176, 1093, 983, 866, 760, 682, 496; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.19-7.18 (m, 1H), 7.10 (t, J = 8 Hz, 1H), 6.92-6.85 (m, 2H), 3.65 (s, 2H), 3.05 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 153.7, 130.3, 123.2, 120.8, 116.1, 111.8, 44.1; MS

(EI, 70 eV) *m/z* 200, 185, 155, 105, 77; HRMS (ESI) calcd for $C_7H_{10}BrN_2$ $[M+H]^+$ *m/z* 201.0027; found *m/z* 201.0030.



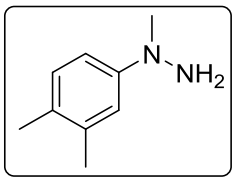
***1-(2-Fluorophenyl)-1-methylhydrazine* (2-45)**[2]

Yellow liquid: 45% yield (0.44 g); IR (KBr, cm^{-1}) 3342, 2957, 2852, 2796, 1618, 1496, 1455, 1212, 1097, 895, 752, 475; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.20-7.15 (m, 1H), 7.07-7.00 (m, 2H), 6.95-6.90 (m, 1H), 3.72 (s, 2H), 3.02 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 154.5 (d, J = 244 Hz), 141.8 (d, J = 8 Hz), 124.3 (d, J = 3 Hz), 122.8 (d, J = 8 Hz), 118.3 (d, J = 2 Hz), 116.2 (d, J = 20 Hz), 46.9 (d, J = 4 Hz); MS (EI, 70 eV) *m/z* 140, 125, 111, 95, 77; HRMS (ESI) calcd for $C_7H_{10}FN_2$ $[M + H]^+$ *m/z* 141.0828; found *m/z* 141.0830.



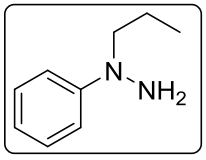
***2-Methyl-4-(1-methylhydrazinyl)benzonitrile* (2-48)**

Yellow solid: 50% yield (0.57 g); mp 92-95 ^oC. IR (KBr, cm^{-1}) 3434, 3323, 2205, 1629, 1504, 1361, 1213, 1056, 941, 863, 660; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.36 (d, J = 8 Hz, 1H), 6.81-6.75 (m, 2H), 3.81 (s, 2H), 3.19 (s, 3H), 2.43 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 154.5, 142.9, 133.6, 120.0, 113.0, 109.8, 99.4, 43.1, 20.9; MS (EI, 70 eV) *m/z* 161, 146, 116, 89, 63; HRMS (ESI) calcd for $C_9H_{12}N_3$ $[M + H]^+$ *m/z* 162.1031; found *m/z* 162.1031.



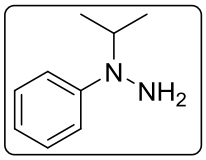
***1-(3,4-Dimethylphenyl)-1-methylhydrazine* (2-50)**

Yellow liquid: 58% yield (0.61 g); IR (KBr, cm^{-1}) 3423, 2923, 2862, 1628, 1507, 1384, 1275, 1106, 927; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 6.99 (d, J = 8 Hz, 1H), 6.81 (d, J = 4 Hz, 1H); 6.72-6.69 (m, 1H), 3.59 (s, 2H), 2.99 (s, 3H), 2.21 (s, 3H), 2.16 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 151.4, 137.1, 130.2, 127.0, 115.7, 111.5, 45.2, 20.4, 18.9; MS (EI, 70 eV) *m/z* 150, 135, 105, 91, 77; HRMS (ESI) calcd for $C_9H_{15}N_2$ $[M + H]^+$ *m/z* 151.1235; found *m/z* 151.1231.



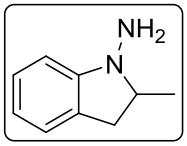
***1-Phenyl-1-propylhydrazine* (2-52)**[2]

Yellow liquid: 62% yield (0.65 g); IR (KBr, cm^{-1}) 3339, 2961, 2872, 1597, 1492, 1380, 1236, 1132, 1032, 991, 877, 749, 692; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.44-7.40 (m, 2H), 7.13-7.10 (m, 2H), 6.95 (t, J = 8 Hz, J = 4 Hz, 1H), 3.62 (s, 2H), 3.48-3.44 (m, 2H), 1.85-1.76 (m, 2H), 1.13 (t, J = 8 Hz, J = 4 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 152.1, 129.2, 118.0, 113.3, 57.5, 19.3, 11.7; MS (EI, 70 eV) *m/z* 150, 134, 121, 104, 77; HRMS (ESI) calcd for $C_9H_{15}N_2$ $[M + H]^+$ *m/z* 151.1235; found *m/z* 151.1239.



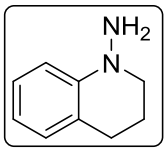
***1-Isopropyl-1-phenylhydrazine* (2-54)**[2]

Yellow liquid: 61% yield (0.64 g); IR (KBr, cm^{-1}) 3352, 2969, 2870, 1597, 1493, 1382, 1286, 1155, 1034, 878, 838, 751, 692; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.33-7.27 (m, 2H), 7.08-7.05 (m, 2H), 6.84-6.80 (m, 1H), 4.14-4.06 (m, 1H), 3.25 (s, 2H), 1.20 (d, J = 4 Hz, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 151.6, 129.1, 118.2, 113.9, 50.7, 17.7; MS (EI, 70 eV) *m/z* 150, 135, 118, 107, 77; HRMS (ESI) calcd for $C_9H_{15}N_2$ $[M + H]^+$ *m/z* 151.1235; found *m/z* 151.1239.



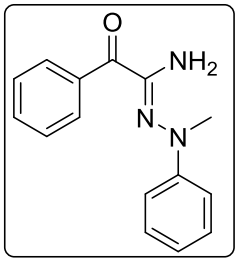
1-Methylindolin-1-amine **(2-59)**[2]

Yellow liquid: 58% yield (0.60 g); IR (KBr, cm^{-1}) 3301, 2929, 2843, 1618, 1488, 1307, 1187, 1031, 942, 878, 751; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.15-7.10 (m, 1H), 7.06-7.03 (m, 1H), 6.79-6.75 (m, 2H), 3.37 (s, 2H), 3.25-3.19 (m, 1H), 3.05-2.99 (m, 1H), 2.56-2.49 (m, 1H), 1.38 (d, J = 8 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 154.7, 127.9, 127.5, 124.3, 120.1, 109.7, 68.1, 36.2, 18.2; MS (EI, 70 eV) *m/z* 148, 133, 116, 91, 77; HRMS (ESI) calcd for $C_9H_{13}N_2$ $[M + H]^+$ *m/z* 149.1079; found *m/z* 149.1078.



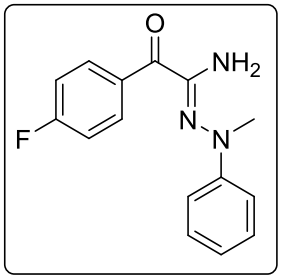
3,4-Dihydroquinolin-1(2H)-amine **(2-60)**[2]

Yellow liquid: 60% yield (0.62 g); IR (KBr, cm^{-1}) 3435, 2964, 1608, 1475, 1382, 1244, 1115, 857, 750; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.07-7.02 (m, 2H), 690-6.87 (m, 1H), 6.66-6.62 (m, 1H), 3.46 (s, 2H), 3.17 (t, J = 8 Hz, J = 4 Hz, 2H), 2.67 (t, J = 4 Hz, J = 8 Hz, 2 H), 1.98-1.92 (m, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 149.2, 129.0, 127.1, 123.4, 118.4, 113.1, 55.2, 27.4, 22.9; MS (EI, 70 eV) *m/z* 148, 130, 119, 91, 77; HRMS (ESI) calcd for $C_9H_{13}N_2$ $[M + H]^+$ *m/z* 149.1079; found *m/z* 149.1077.

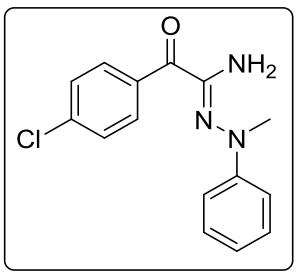


(Z)-N'-Methyl-2-oxo-N',2-diphenylacetohydrazonamide (**3**): Orange solid: 80%

yield (20.2 mg); mp 82-84 ^{o}C. IR (KBr, cm^{-1}) 3475, 3365, 2923, 1623, 1543, 1492, 1376, 1227, 1180, 1088, 999, 821, 756; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.32 (d, J = 8 Hz, 2H), 7.57 (t, J = 8 Hz, 1H), 7.45 (t, J = 8 Hz, 2H), 7.28 (t, J = 8 Hz, 2H), 7.00 (d, J = 8 Hz, 2H), 6.93 (t, J = 8 Hz, 1H), 5.48 (s, 2H), 3.17 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 188.1, 153.9, 150.5, 135.4, 133.2, 131.5, 128.9, 128.0, 121.0, 116.4, 41.57; MS (EI, 70 eV) *m/z* 253, 207, 148, 133, 105; HRMS (ESI) calcd for $C_{15}H_{16}N_3O$ $[M + H]^+$ *m/z* 254.1293; found *m/z* 254.1293.

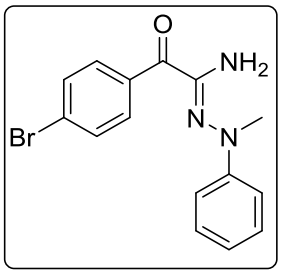


(Z)-2-(4-Fluorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide (**4**): Yellow liquid: 75% yield (20.3 mg); IR (KBr, cm^{-1}) 3474, 3362, 3067, 1664, 1597, 1495, 1377, 1232, 1157, 992, 851, 755, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.46-8.40 (m, 2H), 7.31-7.27 (m, 2H), 7.15-7.10 (m, 2H), 7.00-6.93 (m, 3H), 5.48 (s, 2H), 3.16 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 186.3, 153.9, 150.4, 134.3 (d, J = 10 Hz), 131.6 (d, J = 3 Hz), 129.0, 121.1, 116.4, 115.3, 115.1, 41.5; ^{19}F NMR (376 MHz, $CDCl_3$, ppm) δ -104.64; MS (EI, 70 eV) *m/z* 271, 228, 210, 176, 148; HRMS (ESI) calcd for $C_{15}H_{15}FN_3O$ $[M + H]^+$ *m/z* 272.1199; found *m/z* 272.1198.

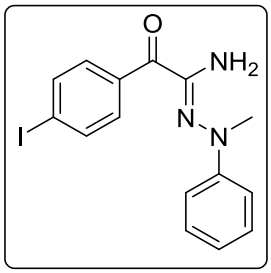


(Z)-2-(4-Chlorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide (**5**): Yellow

liquid: 70% yield (20.2 mg); IR (KBr, cm^{-1}) 3475, 3365, 2924, 1659, 1597, 1492, 1376, 1224, 1175, 1091, 991, 846, 755; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.32 (d, J = 8 Hz, 2H), 7.43 (d, J = 8 Hz, 2H), 7.30 (t, J = 8 Hz, 2H), 7.01-6.94 (m, 3H), 5.47 (s, 2H), 3.17 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.7, 153.6, 150.4, 139.8, 133.6, 133.0, 129.0, 128.4, 121.2, 116.4, 41.6; MS (EI, 70 eV) *m/z* 287, 270, 148, 139, 106; HRMS (ESI) calcd for $C_{15}H_{15}ClN_3O$ $[M + H]^+$ *m/z* 288.0904; found *m/z* 288.0905.

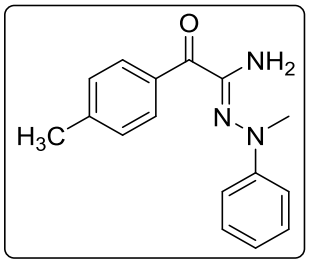


(Z)-2-(4-Bromophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(6):** Yellow liquid: 86% yield (28.6 mg); IR (KBr, cm^{-1}) 3474, 3365, 2922, 1656, 1581, 1492, 1398, 1222, 1110, 1070, 990, 846, 752; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.25-8.22 (m, 2H), 7.61-7.58 (m, 2H), 7.32-7.28 (m, 2H), 7.01-6.95 (m, 3H), 5.46 (s, 2H), 3.17 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.9, 153.5, 150.4, 134.1, 133.1, 131.3, 129.0, 128.7, 121.2, 116.5, 41.7; MS (EI, 70 eV) *m/z*; 331, 316, 281, 229, 183; HRMS (ESI) calcd for $C_{15}H_{15}BrN_3O$ $[M + H]^+$ *m/z* 332.0399; found *m/z* 332.0396.

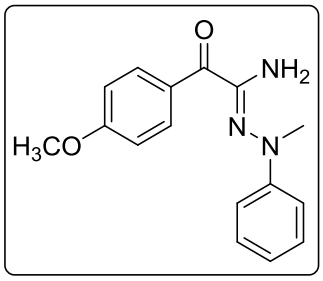


(Z)-2-(4-Iodophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(7):** Yellow liquid: 85% yield (32.3 mg); IR (KBr, cm^{-1}) 3474, 3363, 2932, 1623, 1542, 1492, 1394, 1282, 1179, 1087, 989, 843, 754; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 2H), 7.81 (d, J = 12 Hz, 2H), 7.29 (t, J = 4 Hz, 2H), 6.99-6.93 (m, 3H), 5.42

(s, 2H), 3.15 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.2, 153.5, 150.3, 137.3, 134.6, 132.9, 129.0, 121.2, 116.5, 101.6, 41.7; MS (EI, 70 eV) *m/z* 379, 275, 203, 148, 133; HRMS (ESI) calcd for $C_{15}H_{15}IN_3O$ $[M + H]^+$ *m/z* 380.0260; found *m/z* 380.0259.

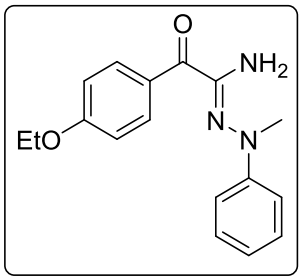


***(Z)-N'-Methyl-2-oxo-N'-phenyl-2-(p-tolyl)acetohydrazonamide* (8):** Yellow solid: 68% yield (18.2 mg); mp 88-91 ^{o}C. IR (KBr, cm^{-1}) 3478, 3356, 2926, 1620, 1560, 1493, 1375, 1232, 1180, 1086, 994, 838, 769; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.25 (d, *J* = 8 Hz, 2H), 7.30-7.24 (m, 4H), 7.00 (d, *J* = 8 Hz, 2H), 6.93 (t, *J* = 8 Hz, 1H), 5.48 (s, 2H), 3.15 (s, 3H), 2.41 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.7, 154.4, 150.6, 144.3, 132.7, 131.7, 128.9, 128.8, 120.8, 116.3, 41.4, 21.8; MS (EI, 70 eV) *m/z* 267, 224, 163, 148, 133; HRMS (ESI) calcd for $C_{16}H_{18}N_3O$ $[M + H]^+$ *m/z* 268.1450; found *m/z* 268.1447.



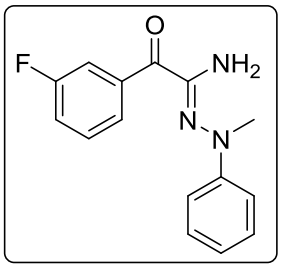
***(Z)-2-(4-Methoxyphenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide* (9):** Yellow solid: 76% yield (21.6 mg); mp 74-77 ^{o}C IR (KBr, cm^{-1}) 3474, 3359, 2928, 1653, 1508, 1492, 1376, 1260, 1169, 1029, 987, 846, 756; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.44-8.41 (m, 2H), 7.31-7.26 (m, 2H), 7.01-6.91 (m, 5H), 5.52 (s, 2H), 3.86 (s, 3H), 3.14 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.2, 163.9, 155.0, 150.6, 134.1, 128.9, 128.0, 120.7, 116.2, 113.5, 55.5, 41.3; MS (EI, 70 eV) *m/z*

283, 179, 148, 135, 106; HRMS (ESI) calcd for $C_{16}H_{18}N_3O_2$ $[M + H]^+$ *m/z* 284.1399; found *m/z* 284.1396.



.

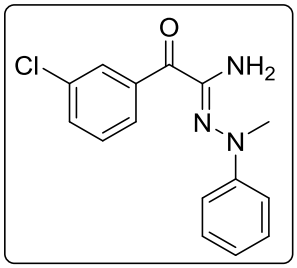
(Z)-2-(4-Ethoxyphenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(10):** Yellow liquid: 56% yield (16.7 mg); IR (KBr, cm^{-1}) 3473, 3359, 2927, 1652, 1598, 1493, 1378, 1237, 1170.6, 1044, 989, 848, 756; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.43-8.40 (m, 2H), 7.30-7.25 (m, 2H), 7.01-6.99 (m, 2H), 6.94-6.89 (m, 3H), 5.50 (s, 2H), 4.10 (q, *J* = 8 Hz, 2H), 3.14 (s, 3H), 1.43 (t, *J* = 8 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.1, 163.4, 155.0, 150.7, 134.1, 128.9, 127.8, 120.7, 116.2, 113.9, 63.8, 41.3, 14.7; MS (EI, 70 eV) *m/z* 297, 236, 193, 149, 133, 121, 106; HRMS (ESI) calcd for $C_{17}H_{20}N_3O_2$ $[M + H]^+$ *m/z* 298.1556; found *m/z* 298.1553.



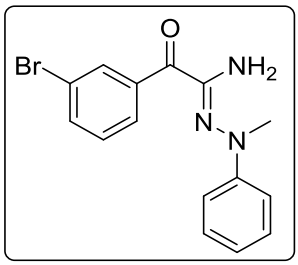
(Z)-2-(3-Fluorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(11):**

Yellow liquid: 79% yield (21.0 mg); IR (KBr, cm^{-1}) 3475, 3363, 2924, 1672, 1598, 1492, 1377, 1252, 1171, 1028, 991, 888, 756, 695; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.16-8.14 (m, 1H), 8.09-8.05 (m, 1H), 7.46-7.40 (m, 1H), 7.33-7.27 (m, 3H), 7.02-6.95 (m, 3H), 5.46 (s, 2H), 3.19 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.6 (d, *J* = 3 Hz), 162.1 (d, *J* = 245 Hz), 153.3, 150.3, 137.2 (d, *J* = 7 Hz), 129.6

(d, *J* = 7 Hz), 129.0, 127.3 (d, *J* = 3 Hz), 121.2, 120.1 (d, *J* = 22 Hz), 118.4 (d, *J* = 23 Hz), 116.5, 41.8; ^{19}F NMR (376 MHz, $CDCl_3$, ppm) δ -112.50; MS (EI, 70 eV) *m/z* 271, 228, 210, 176, 148; HRMS (ESI) calcd for $C_{15}H_{15}FN_3O$ $[M + H]^+$ *m/z* 272.1199; found *m/z* 272.1194.

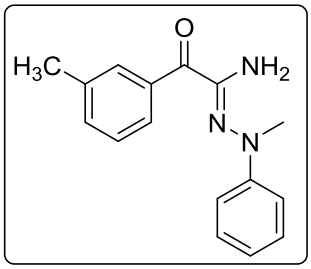


(Z)-2-(3-Chlorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(12):** Yellow liquid: 61% yield (17.5 mg); IR (KBr, cm^{-1}) 3476, 3365, 2926, 1665.1, 1598, 1492, 1381, 1219, 1088, 994, 888, 757, 692; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.29-8.28 (m, 1H), 8.23-8.21 (m, 1H), 7.55-7.52 (m, 1H), 7.38 (t, *J* = 8 Hz, 1H), 7.31-7.27 (m, 2H), 7.01-6.94 (m, 3H), 5.42 (s, 2H), 3.18 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 186.7, 153.0, 150.3, 136.9, 134.1, 133.0, 131.4, 129.7, 129.3, 129.0, 121.3, 116.6, 41.8; MS (EI, 70 eV) *m/z* 287, 183, 148, 139, 106; HRMS (ESI) calcd for $C_{15}H_{15}ClN_3O$ $[M + H]^+$ *m/z* 288.0904; found *m/z* 288.0901.

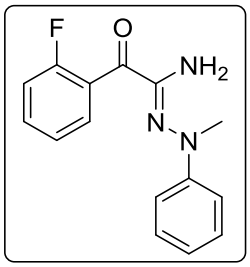


(Z)-2-(3-Bromophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(13):** Yellow solid: 76% yield (25.2 mg); mp 52-55 ^{o}C. IR (KBr, cm^{-1}) 3473, 3365, 2923, 1664, 1625, 1544, 1382, 1217, 1109, 1087, 995, 880, 755; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.43 (t, *J* = 8 Hz, 1H), 8.27-8.24 (m, 1H), 7.69-7.66 (m, 1H), 7.33-7.26 (m, 3H), 7.01-6.92 (m, 3H), 5.40 (s, 2H), 3.17 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz,

$CDCl_3$, ppm) δ 186.6, 152.9, 150.3, 137.1, 135.8, 134.2, 130.1, 129.5, 129.6, 122.1, 121.3, 116.6, 41.9; MS (EI, 70 eV) *m/z* 331, 314, 2644, 183, 133; HRMS (ESI) calcd for $C_{15}H_{15}BrN_3O$ $[M + H]^+$ *m/z* 332.0399; found *m/z* 332.0390.

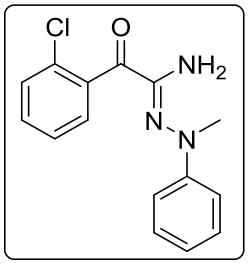


(Z)-N'-Methyl-2-oxo-N'-phenyl-2-(m-tolyl)acetohydrazonamide **(14):** Yellow liquid: 74% yield (19.8 mg); IR (KBr, cm^{-1}) 3474.1, 3364, 2925, 1671, 1598, 1492, 1376, 1230, 1111, 1087, 992, 882, 753; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.75-7.72 (m, 1H), 7.39-7.35 (m, 1H), 7.27-7.23 (m, 4H), 6.95-6.88 (m, 3H), 5.38 (s, 2H), 3.12 (s, 3H), 2.45 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.5, 154.0, 150.6, 137.8, 135.3, 134.1, 131.7, 128.9, 128.9, 127.9, 120.9, 116.3, 41.5, 21.4; MS (EI, 70 eV) *m/z* 267, 224, 163, 148, 133; HRMS (ESI) calcd for $C_{16}H_{18}N_3O$ $[M + H]^+$ *m/z* 268.1450; found *m/z* 268.1451.

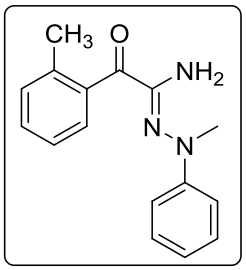


(Z)-2-(2-Fluorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(15):** Yellow liquid: 63% yield (17.1 mg); IR (KBr, cm^{-1}) 3468, 3368, 2926, 1675, 1600, 1452, 1379, 1232, 1105, 1030, 994, 834, 755; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.67-7.63 (m, 1H), 7.44-7.38 (m, 1H), 7.20-7.18 (m, 3H), 7.08-7.03 (m, 1H), 6.90-6.83 (m, 3H), 5.22 (s, 2H), 3.05 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm)

δ 188.3, 160.8 (d, J = 252 Hz), 151.4 (d, J = 244 Hz), 133.4 (d, J = 9 Hz), 131.4 (d, J = 2 Hz), 129.6, 128.9, 123.7 (d, J = 4 Hz), 121.1, 116.4, 116.2 (d, J = 21 Hz), 113.2, 41.4; ^{19}F NMR (376 MHz, $CDCl_3$, ppm) δ -111.03; MS (EI, 70 eV) *m/z* 271, 235, 176, 149, 134; HRMS (ESI) calcd for $C_{15}H_{15}FN_3O$ $[M + H]^+$ *m/z* 272.1199; found *m/z* 272.1202.

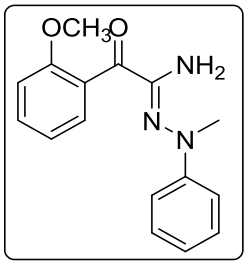


(Z)-2-(2-Chlorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(16):** Yellow solid: 74% yield (21.2 mg); mp 60-63 ^{o}C. IR (KBr, cm^{-1}) 3475, 3368, 1675, 1597, 1546, 1492, 1379, 1272, 1088, 993, 882, 750; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.52-7.50 (m, 1H), 7.42-7.29 (m, 3H), 7.25-7.20 (m, 2H), 6.94-6.88 (m, 3H), 5.25 (s, 2H), 3.11 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 190.6, 152.0, 150.2, 137.0, 131.9, 131.3, 129.8, 129.8, 128.8, 126.1, 121.1, 116.4, 41.3; MS (EI, 70 eV) *m/z* 287, 183, 148, 139, 106; HRMS (ESI) calcd for $C_{15}H_{15}ClN_3O$ $[M + H]^+$ *m/z* 288.0904; found *m/z* 288.0902.

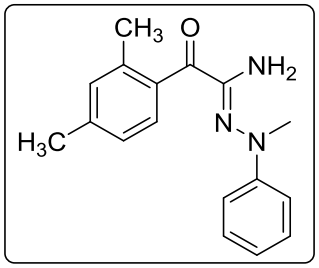


(Z)-N'-Methyl-2-oxo-N'-phenyl-2-(o-tolyl)acetohydrazonamide **(17):** Yellow liquid: 81% yield (21.6 mg); IR (KBr, cm^{-1}) 3474, 3363, 2922, 1623, 1581, 1493, 1376, 1254, 1175, 1088.1, 933, 882, 790, 757, 692; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.74-7.72 (m, 1H), 7.39-7.35 (m, 1H), 7.26-7.22 (m, 4H), 6.94-6.88 (m, 3H), 5.37 (s, 2H), 3.11

(s, 3H), 2.45 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.5, 154.0, 150.6, 137.8, 135.3, 134.1, 131.7, 128.9, 128.9, 127.9, 120.9, 116.3, 41.5, 21.4; MS (EI, 70 eV) *m/z* 267, 224, 210, 194, 133; HRMS (ESI) calcd for $C_{16}H_{18}N_3O$ $[M + H]^+$ *m/z* 268.1450; found *m/z* 268.1447.

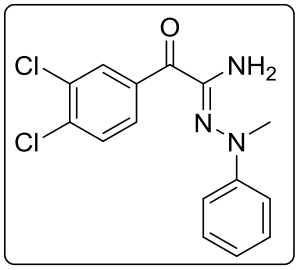


(Z)-2-(2-Methoxyphenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(18):** Yellow liquid: 80% yield (22.6 mg); IR (KBr, cm^{-1}) 3452, 1599, 1491, 1384, 1251, 1118, 1025, 995, 821, 754, 697; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.55 (dd, *J* = 8 Hz, 1H), 7.46-7.42 (m, 1H), 7.25-7.21 (m, 2H), 7.01 (t, *J* = 4 Hz, *J* = 8 Hz, 1H), 6.96-6.92 (m, 3H), 6.88 (t, *J* = 8 Hz, 1H), 5.34 (s, 2H), 3.86 (s, 3H), 3.07 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 191.5, 158.2, 154.5, 150.5, 132.8, 130.6, 128.7, 126.8, 120.5, 120.1, 115.9, 111.3, 55.6, 40.8; MS (EI, 70 eV) *m/z* 283, 267, 235, 179, 148; HRMS (ESI) calcd for $C_{16}H_{18}N_3O_2$ $[M + H]^+$ *m/z* 284.1399; found *m/z* 284.1395.

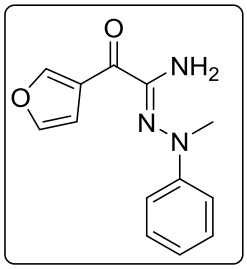


(Z)-2-(2,4-Dimethylphenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(19):** Brown solid: 75% yield (21.1 mg); mp 78-80 ^{o}C. IR (KBr, cm^{-1}) 3474, 3363, 2923, 1664, 1543, 1491, 1379, 1286, 1185, 1088, 989, 759, 697; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.72 (d, *J* = 8 Hz, 1H), 7.26-7.21 (m, 2H), 7.05-7.04 (m, 2H), 6.94-6.87 (m, 3H), 5.39 (s, 2H), 3.10 (s, 3H), 2.44 (s, 3H), 2.34 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 191.7, 154.2, 150.5, 141.8, 138.6, 132.9, 132.0,

131.4, 128.8, 125.5, 120.8, 116.3, 41.4, 21.4, 20.5; MS (EI, 70 eV) *m/z* 281, 175, 148, 133, 105; HRMS (ESI) calcd for $C_{17}H_{20}N_3O$ $[M + H]^+$ *m/z* 282.1606; found *m/z* 282.1604.

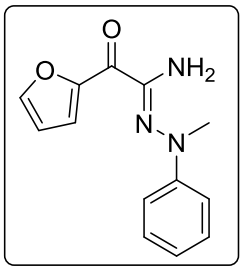


(Z)-2-(3,4-Dichlorophenyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(20):** Yellow liquid: 75% yield (24.1 mg); IR (KBr, cm^{-1}) 3476, 3365, 2922, 1663, 1598, 1492, 1388, 1217, 1112, 1031, 939, 838, 756; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.46 (d, J = 4 Hz, 1H), 8.22 (d, J = 8 Hz, 1H), 7.53 (d, J = 12 Hz, 1H), 7.33-7.28 (m, 2H), 7.02-6.95 (m, 3H), 5.42 (s, 2H), 3.19 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 185.5, 152.8, 150.2, 137.8, 134.9, 133.4, 132.5, 130.7, 130.1, 129.0, 121.4, 116.6, 41.9; MS (EI, 70 eV) *m/z* 321, 281, 173, 148, 106; HRMS (ESI) calcd for $C_{15}H_{14}Cl_2N_3O$ $[M + H]^+$ *m/z* 322.0514; found *m/z* 322.0507.

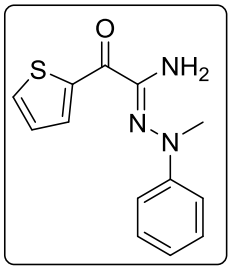


(Z)-2-(Furan-3-yl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(21):** Yellow solid: 68% yield (16.6 mg); mp 62-64 ^oC. IR (KBr, cm^{-1}) 3475, 3364, 3166, 2922, 1628, 1598, 1454, 1400, 1281, 1154, 1088, 914, 871, 755, 694; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.83 (m, 1H), 7.42 (t, J = 4 Hz, 1H); 7.31-7.27 (m, 2H), 7.01-6.98 (m, 3H), 6.94 (t, J = 8 Hz, 1H), 5.41 (s, 2H), 3.13 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 181.4, 153.5, 152.2, 150.7, 143.0, 128.9, 123.6, 120.9, 116.2, 110.1, 41.0; MS (EI, 70 eV) *m/z* 243, 148, 133, 105, 95; HRMS (ESI) calcd for

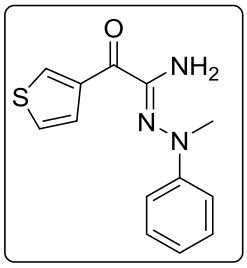
$C_{13}H_{14}N_3O_2$ [M + H]$^+$ *m/z* 244.1086; found *m/z* 244.1086.



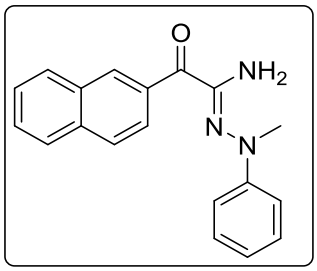
(Z)-2-(Furan-2-yl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(22):** Yellow solid: 88% yield (21.5 mg); mp 147-150 ^{o}C. IR (KBr, cm^{-1}) 3434, 3339, 1614, 1555, 1461, 1376, 1270, 1113, 1039, 990, 902, 882, 779; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.12-8.11 (m, 1H), 7.74-7.73 (m, 1H), 7.33-7.28 (m, 2H), 7.03-7.00 (m, 2H), 6.98-6.93 (m, 1H), 6.57-6.55 (d, *J* = 4 Hz, 1H), 5.47 (s, 2H), 3.15 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 174.2, 153.1, 150.9, 150.0, 148.3, 128.9, 125.8, 120.9, 116.2, 112.7, 40.9; MS (EI, 70 eV) *m/z* 243, 210, 182, 148, 106; HRMS (ESI) calcd for $C_{13}H_{14}N_3O_2$ [M + H]$^+$ *m/z* 244.1086; found *m/z* 244.1089.



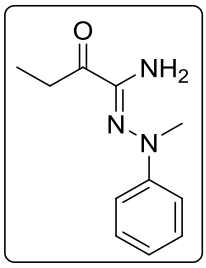
(Z)-N'-Methyl-2-oxo-N'-phenyl-2-(thiophen-2-yl)acetohydrazonamide **(23):** Orange solid: 73% yield (19.0 mg); mp 121-124 ^{o}C. IR (KBr, cm^{-1}) 3435, 3340, 3100, 2879, 1619, 1555, 1494, 1344, 1281, 1086, 913, 857, 734, 690; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.38 (d, *J* = 4 Hz, 1H), 7.73 (d, *J* = 8 Hz, 1H), 7.32 (t, *J* = 8 Hz, 2H), 7.15 (t, *J* = 4 Hz, 1H), 7.10-7.08 (m, 2H), 6.99-6.95 (m, 1H), 5.44 (s, 2H), 3.19 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 178.7, 152.6, 150.6, 137.8, 136.9, 128.9, 127.4, 121.0, 116.5, 41.3; MS (EI, 70 eV) *m/z* 259, 242, 173, 148, 133; HRMS (ESI) calcd for $C_{13}H_{14}N_3OS$ [M + H]$^+$ *m/z* 260.0858; found *m/z* 260.0858.



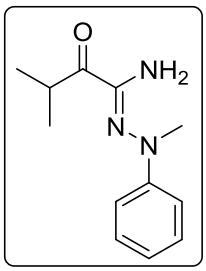
(Z)-N'-Methyl-2-oxo-N'-phenyl-2-(thiophen-3-yl)acetohydrazonamide **(24):** Yellow solid: 66% yield (17.1 mg); mp 100-103 ^{o}C. IR (KBr, cm^{-1}) 3474, 3362, 3125, 2879, 1623, 1509, 1493, 1284, 1158, 1084, 1031, 868, 757, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 9.10 (d, J = 4 Hz, 1H), 7.92 (d, J = 4 Hz, 1H), 7.35-7.31 (m, 3H), 7.06 (m, 2H), 6.99 (t, J = 8 Hz, 1H), 5.53 (s, 2H), 3.19 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 180.4, 154.4, 150.7, 138.2, 129.4, 129.0, 125.1, 120.9, 116.2, 41.1; MS (EI, 70 eV) *m/z* 259, 207, 173, 148, 133; HRMS (ESI) calcd for $C_{13}H_{14}N_3OS$ [M + H]$^{+}$ *m/z* 260.0858; found *m/z* 260.0859.



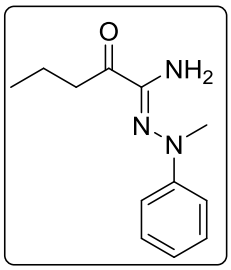
(Z)-N'-Methyl-2-(naphthalen-2-yl)-2-oxo-N'-phenylacetohydrazonamide **(26):** Pale brown solid: 74% yield (22.4 mg); mp 160-163 ^{o}C. IR (KBr, cm^{-1}) 3475, 3352, 2923, 2581, 1762, 1627, 1579, 1497, 1370, 1230, 1173, 1086, 969, 835, 751; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.68 (d, J = 8 Hz, 1H), 8.12 (d, J = 8 Hz, 1H), 7.81 (d, J = 8 Hz, 1H), 7.70 (t, J = 8 Hz, 1H), 7.46 (t, J = 8 Hz, 1H), 7.28 (t, J = 8 Hz, 2H), 7.19 (d, J = 8 Hz, 1H), 6.97-6.89 (m, 3H), 3.47 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 179.9, 157.9, 153.5, 147.3, 141.1, 131.5, 130.7, 129.6, 129.2, 129.2, 125.9, 123.7, 121.1, 113.2, 110.4, 109.3, 39.1; MS (EI, 70 eV) *m/z* 303, 281, 263, 196, 126; HRMS (ESI) calcd for $C_{19}H_{18}N_3O$ [M + H]$^{+}$ *m/z* 304.1450; found *m/z* 304.1441.



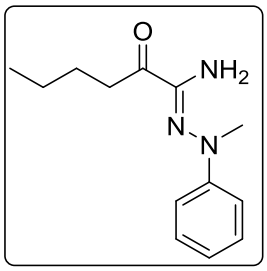
(Z)-N'-Methyl-2-oxo-N'-phenylbutanehydrazonamide **(27):** Yellow liquid: 46% yield (9.5 mg); IR (KBr, cm^{-1}) 3473, 3365, 2924, 2876, 1701, 1659, 1493, 1381, 1287, 1114, 1031, 995, 887, 754, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.20 (t, J = 8 Hz, 2H), 6.89 (d, J = 8 Hz, 2H), 6.84 (t, J = 8 Hz, 1H), 5.18 (s, 2H), 3.00-2.94 (m, 5H), 1.10 (t, J = 4 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 198.4, 152.9, 150.6, 128.9, 120.8, 116.0, 40.7, 30.1, 8.0; MS (EI, 70 eV) *m/z* 205, 176, 148, 133, 106; HRMS (ESI) calcd for $C_{11}H_{16}N_3O$ $[M + H]^+$ *m/z* 206.1293; found *m/z* 206.1295.



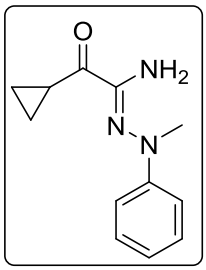
(Z)-N',3-Dimethyl-2-oxo-N'-phenylbutanehydrazonamide **(28):** Yellow liquid: 50% yield (11 mg); IR (KBr, cm^{-1}) 3477, 3368, 2971, 2874, 1697, 1598, 1493, 1282, 1110, 1044, 930, 882, 754, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.30-7.25 (m, 2H), 6.99-6.91 (m, 3H), 5.25 (s, 2H), 3.90-3.83 (m, 1H), 3.08 (s, 3H), 1.21-1.19 (m, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 201.6, 152.2, 150.7, 128.9, 120.8, 116.0, 40.7, 33.6, 18.9; MS (EI, 70 eV) *m/z* 219, 176, 148, 133,107; HRMS (ESI) calcd for $C_{12}H_{18}N_3O$ $[M + H]^+$ *m/z* 220.1450; found *m/z* 220.1449.



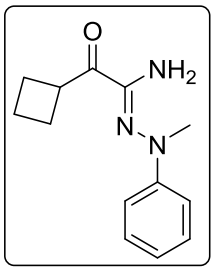
(Z)-N'-Methyl-2-oxo-N'-phenylpentanehydrazonamide **(29):** Yellow liquid: 50% yield (11 mg); IR (KBr, cm^{-1}) 3477, 3365, 2926, 2874, 1698, 1599, 1493, 1385, 1285, 1123, 1031, 994, 885, 754; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.30-7.28 (m, 2H), 6.98-6.91 (m, 3H), 5.25 (s, 2H), 3.08 (s, 3H), 3.00 (t, J = 8 Hz, J = 4 Hz, 2H), 1.75-1.69 (m, 2H), 0.99 (t, J = 8 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 197.9, 153.1, 150.6, 128.9, 120.8, 116.0, 40.7, 38.4, 17.7, 13.8; MS (EI, 70 eV) *m/z* 219, 176, 148, 133,107; HRMS (ESI) calcd for $C_{12}H_{18}N_3O$ $[M + H]^+$ *m/z* 220.1450; found *m/z* 220.1449.



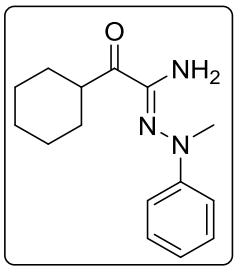
(Z)-N'-Methyl-2-oxo-N'-phenylhexanehydrazonamide **(30):** Yellow liquid: 45% yield (10.5 mg); IR (KBr, cm^{-1}) 3473, 3365, 2959, 2872, 1698, 1599, 1493, 1285, 1115, 1062, 995, 755, 693, 529; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.27 (t, J = 8 Hz, 2H), 6.98-6.91 (m, 3H), 5.24 (s, 2H), 3.07 (s, 3H), 3.02 (t, J = 8 Hz, 2H), 1.67-1.63 (m, 2H), 1.42-1.34 (m, 2H), 0.94 (t, J = 8 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 198.0, 153.1, 150.6, 128.9, 120.8, 116.0, 40.7, 36.3, 26.4, 22.4, 14.0; MS (EI, 70 eV) *m/z* 233, 191, 148, 127, 106; HRMS (ESI) calcd for $C_{13}H_{20}N_3O$ $[M + H]^+$ *m/z* 234.1606; found *m/z* 234.1608.



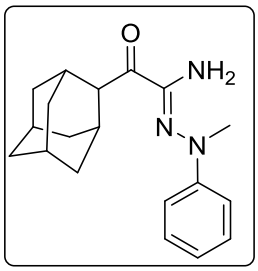
(Z)-2-Cyclopropyl-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(31):** Yellow liquid: 47% yield (10.2 mg); IR (KBr, cm^{-1}) 3474, 3361, 3008, 2875, 1683, 1630, 1599, 1493, 1283, 1185, 1076, 993, 881, 755; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.29 (t, J = 8 Hz, 2H), 7.02 (d, J = 8 Hz, 2H), 6.94 (t, J = 8 Hz, 1H), 5.29 (s, 2H), 3.30-3.25 (m, 1H), 3.11 (s, 3H), 1.23-1.19 (m, 2H), 1.11-1.07 (m, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 197.1, 153.3, 150.7, 128.9, 120.8, 116.1, 40.8, 15.3, 13.2; MS (EI, 70 eV) *m/z* 217, 176, 148, 133, 106; HRMS (ESI) calcd for $C_{12}H_{16}N_3O$ $[M + H]^+$ *m/z* 218.1294; found *m/z* 218.1293.



(Z)-2-Cyclobutyl-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(32):** Yellow liquid: 40% yield (9 mg); IR (KBr, cm^{-1}) 3475, 3363, 2942, 2867, 1693, 1599, 1493, 1386, 1285, 1120, 1088, 993, 881; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.22-7.18 (m, 2H), 6.89-6.83 (m, 3H), 5.17 (s, 2H), 4.24-4.16 (m, 1H), 2.98 (s, 3H), 2.32-2.18 (m, 4H), 2.03-1.91 (m, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 198.1, 152.2, 150.6, 128.8, 120.7, 115.9, 40.6, 40.0, 25.1, 18.1; MS (EI, 70 eV) *m/z* 231, 203, 148, 133, 107; HRMS (ESI) calcd for $C_{13}H_{18}N_3O$ $[M + H]^+$ *m/z* 232.1450; found *m/z* 232.1449.

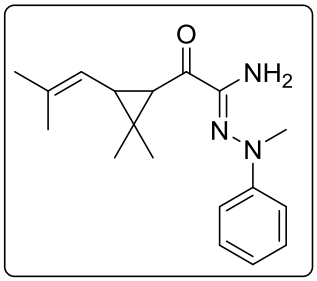


(Z)-2-Cyclohexyl-N'-methyl-2-oxo-N'-phenylacetohydrazonamide **(33):** Yellow liquid: 57% yield (14.8 mg); IR (KBr, cm^{-1}) 3473, 3373, 2924, 2856, 1693, 1597, 1494, 1384, 1283, 1149, 1090, 927, 868; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.28 (t, J = 8 Hz, 2H), 6.97-6.90 (m, 3H), 5.23 (s, 2H), 3.65-3.60 (m, 1H), 3.08 (s, 3H), 1.94-1.79 (m, 4H), 1.72-1.69 (m, 1H), 1.47-1.33 (m, 4H), 1.28-1.24 (m, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 200.7, 152.3, 150.6, 128.9, 120.7, 115.9, 43.3, 40.8, 29.1, 25.9, 25.5; MS (EI, 70 eV) *m/z* 259, 242, 153, 133, 107; HRMS (ESI) calcd for $C_{15}H_{22}N_3O$ $[M + H]^+$ *m/z* 260.1763; found *m/z* 260.1765.

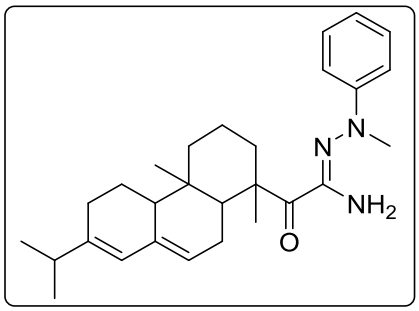


.

(Z)-2-((1r,3r,5r,7r)-Adamantan-2-yl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamid e **(34):** Yellow solid: 57% yield (17.8 mg); mp 115-118 ^{o}C. IR (KBr, cm^{-1}) 3483, 3366, 2905, 2851, 1674, 1598, 1453, 1361, 1285, 1165, 1011, 976, 877, 754; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.29-7.25 (m, 2H), 6.98-6.95 (m, 2H), 6.91 (t, J = 8 Hz, 1H), 5.23 (s, 2H), 3.07 (s, 3H), 2.25-2.24 (m, 6H), 2.05-2.03 (m, 3H), 1.78-1.74 (m, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 201.3, 153.1, 150.9, 128.9, 120.5, 116.1, 46.9, 41.3, 38.9, 36.7, 28.2; MS (EI, 70 eV) *m/z* 311, 190, 176, 148, 107; HRMS (ESI) calcd for $C_{19}H_{26}N_3O$ $[M + H]^+$ *m/z* 312.2076; found *m/z* 312.2074.

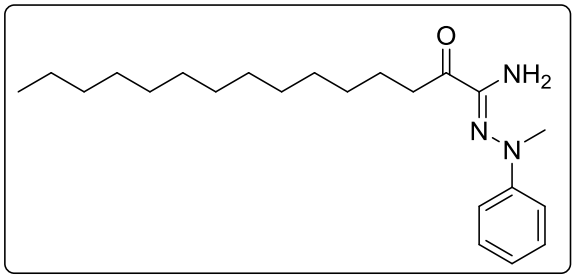


***(Z)-2-(2,2-Dimethyl-3-(2-methylprop-1-en-1-yl)cyclopropyl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide* (35):** Yellow liquid: 60% yield (18.0 mg); IR (KBr, cm^{-1}) 3479, 3363, 2923, 2873, 1677, 1540, 1493, 1377, 1281, 1168, 1031, 975, 851, 755; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.20 (t, J = 8 Hz, 2H), 6.91 (d, J = 8 Hz, 2H), 6.84 (t, J = 8 Hz, 1H), 5.21 (s, 2H), 4.95 (d, J = 8 Hz, 1H), 3.01 (s, 3H), 2.31 (t, J = 8 Hz, J = 4 Hz, 1H), 1.64 (d, J = 8 Hz, 6H), 1.19 (d, J = 4 Hz, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 194.3, 154.2, 150.8, 135.4, 128.8, 121.5, 120.5, 115.8, 40.6, 37.7, 36.6, 34.9, 25.6, 22.4, 19.9, 18.6; MS (EI, 70 eV) *m/z* 299, 193, 176, 149, 107 ; HRMS (ESI) calcd for $C_{18}H_{26}N_3O$ $[M + H]^+$ *m/z* 300.2076; found *m/z* 300.2075.

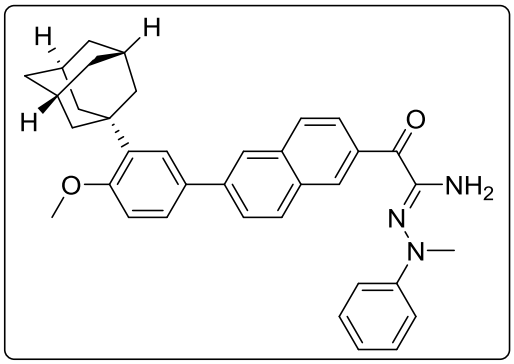


***(Z)-2-(7-Isopropyl-1,4a-dimethyl-1,2,3,4,4a,4b,5,6,10,10a-decahydrophenanthren-1-yl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide* (36):** Yellow solid: 56% yield (24.2 mg); mp 119-122 ^{o}C. IR (KBr, cm^{-1}) 3463, 3338, 2925, 2870, 1673, 1598, 1492, 1381, 1282, 1168, 1091, 993, 893, 758; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.14 (t, J = 8 Hz, 2H), 6.86 (d, J = 8 Hz, 2H), 6.80 (t, J = 8 Hz, 1H), 5.74 (s, 1H), 5.31 (d, J = 4 Hz, 1H), 5.22 (s, 2H), 3.32 (dd, J = 4 Hz, 1H), 2.94 (s, 3H), 2.59-2.52 (m, 1H), 2.17 (p, J = 4 Hz, 1H), 2.03-1.95 (m, 4H), 1.81-1.70 (m, 3H), 1.66-1.57 (m, 3H), 1.29 (s, 3H), 1.18 (d, J = 4 Hz, 2H), 0.96 (q, J = 4 Hz, 6H), 0.80 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 202.4, 153.8, 151.3, 145.1, 135.9, 128.8, 122.6, 120.6, 120.4, 116.0, 52.4, 51.2, 43.0, 40.3, 38.0, 36.3, 35.0, 34.5, 27.5, 26.1, 22.6, 21.5, 20.9, 18.5,

17.6, 14.3; MS (EI, 70 eV) *m/z* 433, 231, 176, 149, 107; HRMS (ESI) calcd for $C_{28}H_{40}N_3O$ $[M + H]^+$ *m/z* 434.3171; found *m/z* 434.3169.

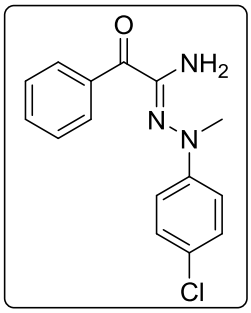


***(Z)-N'-Methyl-2-oxo-N'-phenylpentadecanehydrazonamide* (37):** Yellow solid: 43% yield (15.5 mg); mp 40-42 ^oC. IR (KBr, cm^{-1}) 3480, 3367, 2924, 2853, 1699, 1599, 1493, 1387, 1285, 1188, 1028, 995, 877, 754; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 7.20 (t, *J* = 8 Hz, 2H), 6.90-6.83 (m, 3H), 5.17 (s, 2H), 3.00 (s, 3H), 2.94 (t, *J* = 8 Hz, 2H), 1.64-1.57 (m, 2H), 1.18 (s, 20H), 0.81 (t, *J* = 4 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 198.1, 153.1, 150.6, 128.9, 120.8, 116.0, 40.7, 36.6, 31.9, 29.7, 29.7, 29.7, 29.7, 29.5, 29.5, 29.4, 29.2, 24.3, 22.7, 14.1; MS (EI, 70 eV) *m/z* 359, 316, 253, 191, 148; HRMS (ESI) calcd for $C_{22}H_{38}N_3O$ $[M + H]^+$ *m/z* 360.3015; found *m/z* 360.3013.

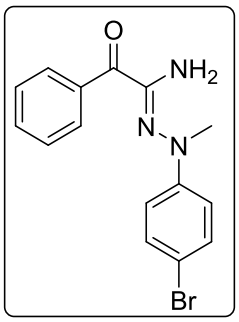


***(Z)-2-(6-(3-((3r,5r,7r)-Adamantan-1-yl)-4-methoxyphenyl)naphthalen-2-yl)-N'-methyl-2-oxo-N'-phenylacetohydrazonamide* (38):** Orange solid: 30% yield (16.3 mg); mp 165-167 ^oC. IR (KBr, cm^{-1}) 3484, 3369, 2903, 2849, 1655, 1621, 1599, 1492, 1382, 1272, 1238, 1173, 1031, 995, 885; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 9.07 (s, 1H), 8.29 (d, *J* = 8 Hz, 1H), 8.00-7.98 (m, 2H), 7.91 (d, *J* =8 Hz, 1H), 7.78 (d, *J* = 8 Hz, 1H), 7.61 (d, *J* = 8 Hz, 1H), 7.54 (d, *J* = 8 Hz, 1H), 7.33-7.29 (m, 2H), 7.07 (d, *J*

= 8 Hz, 2H), 7.01-6.93 (m, 2H), 5.54 (s, 2H), 3.89 (s, 3H), 3.21 (s, 3H), 2.19-2.18 (m, 6H), 2.11-2.10 (m, 3H), 1.81-1.79 (m, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.6, 159.0, 154.4, 150.7, 141.8, 139.0, 136.1, 134.3, 132.5, 132.1, 131.0, 130.5, 129.0, 127.9, 126.8, 126.4, 126.0, 125.8, 124.6, 120.9, 116.4, 112.1, 55.2, 41.5, 40.6, 37.2, 37.2, 29.1; HRMS (ESI) calcd for $C_{36}H_{38}N_3O_2$ $[M + H]^+$ *m/z* 544.2964; found *m/z* 544.2962.

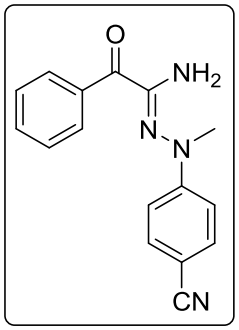


(Z)-N'-(4-Chlorophenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(39):** Orange solid: 79% yield (22.7 mg); mp 94-97 °C. IR (KBr, cm^{-1}) 3475, 3366, 1658, 1623, 1595, 1491, 1379, 1228, 1179, 1097, 989, 820, 733; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.29 (d, *J* = 8 Hz, 2H), 7.58 (t, *J* = 8 Hz, 1H), 7.45-7.43 (t, *J* = 8 Hz, 2H), 7.23-7.20 (d, *J* = 8 Hz, 2H), 6.91 (d, *J* = 8 Hz, 2H), 5.50 (s, 2H), 3.12 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 154.3, 149.3, 135.2, 133.3, 131.4, 128.7, 128.1, 125.8, 117.4, 41.1; MS (EI, 70 eV) *m/z* 287, 182, 149, 140, 105; HRMS (ESI) calcd for $C_{15}H_{15}ClN_3O$ $[M + H]^+$ *m/z* 288.0904; found *m/z* 288.0906.

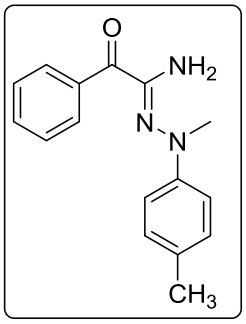


(Z)-N'-(4-Bromophenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(40):** Yellow liquid: 77% yield (23.9 mg); IR (KBr, cm^{-1}) 3474, 3365, 2929, 1623, 1595, 1487, 1377, 1228, 1104, 1001, 988, 816, 733; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ

8.29 (d, *J* = 8 Hz, 2H), 7.57 (t, *J* = 8 Hz, 1H), 7.44 (t, *J* = 8 Hz, 2H), 7.35 (d, *J* = 8 Hz, 2H), 6.87-6.84 (d, *J* = 12 Hz, 2H), 5.50 (s, 2H), 3.12 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 154.3, 149.7, 135.2, 133.3, 131.7, 131.4, 128.1, 117.8, 113.2, 41.0; MS (EI, 70 eV) *m/z* 331, 226, 185, 147, 105; HRMS (ESI) calcd for $C_{15}H_{15}BrN_3O$ $[M + H]^+$ *m/z* 332.0399; found *m/z* 332.0395.

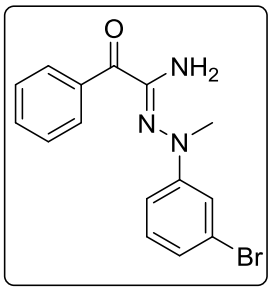


(Z)-N'-(4-Cyanophenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(41):** Yellow solid: 81% yield (22.5 mg); mp 126-129 ^{o}C. IR (KBr, cm^{-1}) 3483, 3366, 2906, 2851, 1674, 1538, 1493, 1361, 1285, 1165, 1011, 937, 877; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.28 (d, *J* = 8 Hz, 2H), 7.60 (t, *J* = 8 Hz, 1H), 7.50-7.45 (m, 4H), 6.93 (d, *J* = 8 Hz, 2H), 5.63 (s, 2H), 3.21 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.8, 155.4, 153.2, 134.9, 133.7, 133.2, 131.4, 128.2, 120.0, 114.7, 101.8, 39.3; MS (EI, 70 eV) *m/z* 278, 173, 149, 131, 105; HRMS (ESI) calcd for $C_{16}H_{15}N_4O$ $[M + H]^+$ *m/z* 279.1246; found *m/z* 279.1254.

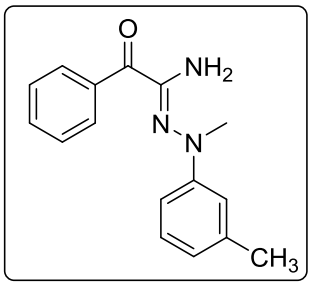


(Z)-N'-Methyl-2-oxo-2-phenyl-N'-(p-tolyl)acetohydrazonamide **(42):** Yellow liquid: 77% yield (20.6 mg); IR (KBr, cm^{-1}) 3474, 3362, 3125, 2879, 1623, 1598, 1492, 1284, 1158, 1083, 932, 868, 757, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.31 (d, *J* = 8 Hz, 2H), 7.55 (t, *J* = 8 Hz, 1H), 7.43 (t, *J* = 8 Hz, 2H), 7.09 (d, *J* = 12 Hz, 2H), 6.92 (d, *J*

= 12 Hz, 2H), 5.40 (s, 2H), 3.14 (s, 3H), 2.29 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 153.3, 148.3, 135.5, 133.1, 131.5, 130.8, 129.5, 128.0, 117.0, 42.6, 20.6; MS (EI, 70 eV) *m/z* 267, 234, 162, 147, 105; HRMS (ESI) calcd for $C_{16}H_{18}N_3O$ $[M + H]^+$ *m/z* 268.1450; found *m/z* 268.1452.

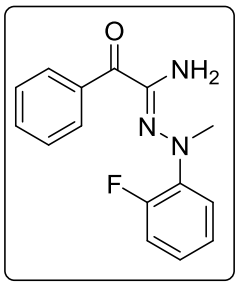


(Z)-N'-(3-Bromophenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(43):** Brown liquid: 69% yield (22.8 mg); IR (KBr, cm^{-1}) 3473, 3365, 3075, 2918, 1664, 1591, 1477, 1306, 1228, 1177, 1084, 987, 821; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.31 (d, *J* = 8 Hz, 2H), 7.59 (t, *J* = 8 Hz, 1H), 7.47 (t, *J* = 8 Hz, 2H), 7.16-7.14 (m, 1H), 7.11 (d, *J* = 8 Hz, 1H), 7.05-7.03 (d, *J* = 8 Hz, 1H), 6.90 (d, *J* = 8 Hz, 1H), 5.54 (s, 2H), 3.13 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 154.5, 151.9, 135.1, 133.3, 131.3, 130.0, 128.0, 123.3, 122.8, 118.8, 114.3, 40.5; MS (EI, 70 eV) *m/z* 331, 228, 185, 147, 105; HRMS (ESI) calcd for $C_{15}H_{15}BrN_3O$ $[M + H]^+$ *m/z* 332.0399; found *m/z* 332.0396.



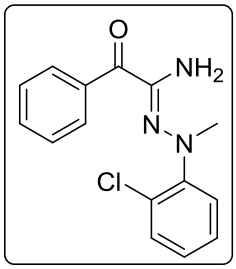
(Z)-N'-Methyl-2-oxo-2-phenyl-N'-(m-tolyl)acetohydrazonamide **(44):** Yellow liquid: 81% yield (21.6 mg); IR (KBr, cm^{-1}) 3470, 3363, 2924, 1657, 1544, 1491, 1378, 1228, 1115, 989, 863, 773, 691; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.32 (d, *J* = 8 Hz, 2H), 7.56 (t, *J* = 8 Hz, 1H), 7.44 (t, *J* = 8 Hz, 2H), 7.16 (t, *J* = 8 Hz, 1H), 6.82-6.72 (m, 3H), 5.45 (s, 2H), 3.15 (s, 3H), 2.32 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ

188.2, 153.8, 150.5, 138.7, 135.4, 133.2, 131.5, 128.8, 128.0, 122.0, 117.3, 113.6, 41.8, 21.8; MS (EI, 70 eV) *m/z* 267, 231, 162, 147, 105; HRMS (ESI) calcd for $C_{16}H_{18}N_3O$ $[M + H]^+$ *m/z* 268.1450; found *m/z* 268.1448.



(Z)-N'-(2-Fluorophenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(45):**

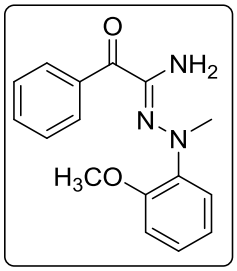
Yellow liquid: 47% yield (12.8 mg); IR (KBr, cm^{-1}) 3483, 3373, 3069, 2924, 1656, 1579, 1492, 1379, 1225, 1180, 1094, 989, 829, 753; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.27 (d, *J* = 8 Hz, 2H), 7.58 (t, *J* = 4 Hz, *J* =8 Hz, 1H), 7.46 (t, *J* = 8 Hz, 2H), 7.29-7.24 (m, 1H), 7.10-7.00 (m, 3H), 5.42 (s, 2H), 3.13 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 154.9 (d, *J* = 244 Hz), 152.1, 139.0 (d, *J* = 9 Hz), 135.3, 133.0, 131.3, 127.9, 124.3 (d, *J* = 4 Hz), 124.0 (d, *J* = 8 Hz), 120.7 (d, *J* = 2 Hz), 116.2 (d, *J* = 20 Hz), 44.3 (d, *J* = 2 Hz); ^{19}F NMR (376 MHz, $CDCl_3$, ppm) δ -112.65; MS (EI, 70 eV) *m/z* 271, 251, 166, 149, 124, 105; HRMS (ESI) calcd for $C_{15}H_{15}FN_3O$ $[M + H]^+$ *m/z* 272.1199; found *m/z* 272.1203.



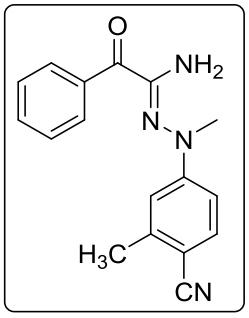
(Z)-N'-(2-Chlorophenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(46):**

Yellow solid: 40% yield (11.5 mg); mp 58-60 ^oC. IR (KBr, cm^{-1}) 3474, 3362, 2725,

1623, 1509, 1422, 1236, 1158, 1083, 933, 868, 757; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.26 (d, J = 8 Hz, 2H), 7.57 (t, J = 8 Hz, 1H), 7.42-7.39 (m, 4H), 7.24-7.20 (m, 1H), 7.07 (t, J = 8 Hz, 1H), 5.35 (s, 2H), 3.08 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 152.2, 148.9, 135.5, 133.0, 131.4, 130.4, 128.5, 127.9, 127.8, 125.7, 122.9, 45.1; MS (EI, 70 eV) *m/z* 287, 252, 207, 182, 105; HRMS (ESI) calcd for $C_{15}H_{15}ClN_3O$ $[M + H]^+$ *m/z* 288.0904; found *m/z* 288.0903.

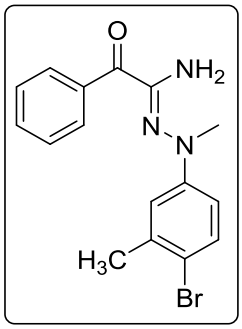


(Z)-N'-(2-Methoxyphenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(47):** Yellow liquid: 49% yield (13.9 mg); IR (KBr, cm^{-1}) 3476, 3363, 3064, 2922, 1656, 1544, 1493, 1376, 1225, 1107, 1026, 989, 812, 748; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.26 (d, J = 8 Hz, 2 H), 7.55 (t, J = 8 Hz, 1H), 7.44 (t, J = 8 Hz, 2H), 7.21 (d, J = 8 Hz, 1H), 7.10 (t, J = 8 Hz, 1H), 6.95-6.89 (m, 2H), 5.16 (s, 2H), 3.91 (s, 3H), 3.15 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.9, 152.3, 149.8, 139.4, 135.8, 132.8, 131.4, 127.8, 125.4, 121.9, 121.2, 111.7, 55.6, 46.2; MS (EI, 70 eV) *m/z* 283, 251, 207, 178, 135, 105; HRMS (ESI) calcd for $C_{16}H_{18}N_3O_2$ $[M + H]^+$ *m/z* 284.1399; found *m/z* 284.1397.

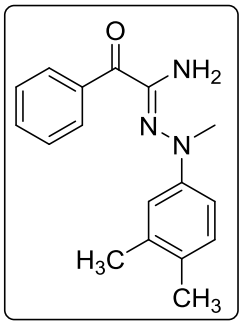


(Z)-N'-(4-Cyano-3-methylphenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide **(48):** Fuchsia solid, 63% yield (18.4 mg); mp 100-112 ^{o}C. IR (KBr, cm^{-1}) 3465, 3356, 2223, 1625, 1502, 1449, 1308, 1228, 1179, 1077, 992, 824, 708; ^{1}H NMR (400 MHz,

$CDCl_3$, ppm) δ 8.27 (d, J = 8 Hz, 2H), 7.60 (t, J = 8 Hz, 1H), 7.46 (t, J = 8 Hz, 2H), 7.19-7.17 (m, 2H), 7.10-7.07 (m, 1H), 5.57 (s, 2H), 3.12 (s, 3H), 2.45 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 154.8, 149.1, 135.1, 133.5, 133.5, 131.4, 130.7, 128.1, 120.5, 119.2, 118.6, 112.8, 40.4, 19.5; MS (EI, 70 eV) *m/z* 292, 187, 172, 149, 105; HRMS (ESI) calcd for $C_{17}H_{17}N_4O$ $[M + H]^+$ *m/z* 293.1402; found *m/z* 293.1401.

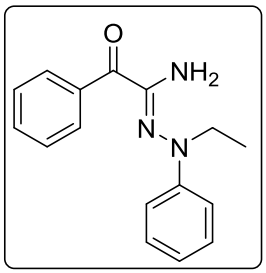


***(Z)-N'-(4-Bromo-3-methylphenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide* (49):** Yellow solid: 58% yield (20.1 mg); mp 93-96 ^oC. IR (KBr, cm^{-1}) 3474, 3366, 2922, 2876, 1626, 1594, 1477, 1378, 1227, 1110, 1624, 988, 855, 731; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.31 (d, J = 8 Hz, 2H), 7.59 (t, J = 8 Hz, 1H), 7.46 (t, J = 8 Hz, 2H), 7.39 (d, J = 8 Hz, 1H), 6.88-6.87 (m, 1H), 6.71-6.09 (m, 1H), 5.49 (s, 2H), 3.13 (s, 3H), 2.36 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 154.1, 149.9, 138.0, 135.2, 133.3, 132.4, 131.5, 128.1, 118.6, 116.0, 115.4, 41.2, 23.3; MS (EI, 70 eV) *m/z* 345, 281, 218, 148, 106; HRMS (ESI) calcd for $C_{16}H_{17}BrN_3O$ $[M + H]^+$ *m/z* 346.0555; found *m/z* 346.0552.

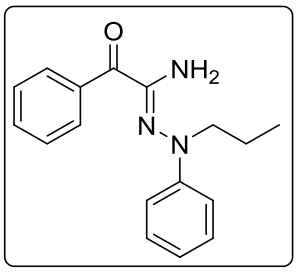


***(Z)-N'-(3,4-Dimethylphenyl)-N'-methyl-2-oxo-2-phenylacetohydrazonamide* (50):** Orange solid: 55% yield (15.5 mg); mp 82-85 ^oC. IR (KBr, cm^{-1}) 3476, 3363, 2921,

1658, 1502, 1449, 1347, 1227, 1179, 1002, 955, 810, 734; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.25 (d, J = 8 Hz, 2H), 7.49 (t, J = 8 Hz, 1H), 7.37 (d, J = 8 Hz, 2H), 6.96 (d, J = 8 Hz, 1H), 6.75 (s, 1H), 6.69 (d, J = 8 Hz, 1H), 5.31 (s, 2H), 3.07 (s, 3H), 2.16 (s, 3H), 2.13 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 188.1, 153.0, 148.6, 137.1, 135.5, 133.1, 131.6, 130.0, 129.7, 128.0, 118.7, 114.6, 42.9, 20.2, 18.9; MS (EI, 70 eV) *m/z* 281, 264, 207, 176, 134; HRMS (ESI) calcd for $C_{17}H_{20}N_3O$ [M + H]$^{+}$ *m/z* 282.1606; found *m/z* 282.1604.

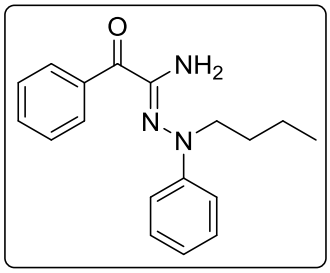


(Z)-N'-Ethyl-2-oxo-N',2-diphenylacetohydrazonamide **(51):** Yellow liquid: 79% yield (21.2 mg); IR (KBr, cm^{-1}) 3484, 3371, 3069, 2977, 1625, 1597, 1491, 1378, 1226, 1179, 1098, 992, 754; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.32 (d, J = 8 Hz, 2H), 7.57 (t, J = 8 Hz, 1H), 7.45 (t, J = 8 Hz, 2H), 7.27 (d, J = 8 Hz, 2H), 6.96 (d, J = 8 Hz, 3H), 5.29 (s, 2H), 3.61-3.56 (m, 2H), 1.27-1.19 (m, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 187.9, 152.6, 148.1, 135.6, 133.1, 131.5, 129.0, 128.0, 121.7, 118.5, 52.0, 12.7; MS (EI, 70 eV) *m/z* 267, 235, 162, 134, 105; HRMS (ESI) calcd for $C_{16}H_{18}N_3O$ [M + H]$^{+}$ *m/z* 268.1450; found *m/z* 268.1448.

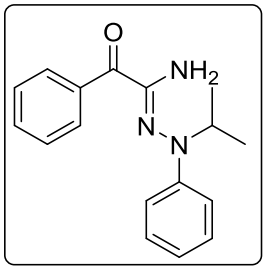


(Z)-2-Oxo-N',2-diphenyl-N'-propylacetohydrazonamide **(52):** Yellow liquid: 85% yield (24.0 mg); IR (KBr, cm^{-1}) 3485, 3375, 3061, 2963, 2873, 1624, 1597, 1490, 1380, 1226, 1179, 985, 807, 753; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.31 (d, J = 8

Hz, 2H), 7.57 (d, *J* = 8 Hz, 1H), 7.45 (t, *J* = 8 Hz, 2H), 7.26 (t, *J* = 8 Hz, 2H), 6.95-6.92 (m, 3H), 5.25 (s, 2H), 3.49 (t, *J* = 8, 4 Hz, 2H), 1,75-1.66 (m, 2H), 0.96 (t, *J* = 8 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.8, 152.3, 148.4, 135.6, 133.0, 131.5, 129.0, 128.0, 121.6, 118.3, 60.2, 21.3, 11.7; MS (EI, 70 eV) *m/z* 281, 235, 176, 149, 134; HRMS (ESI) calcd for $C_{17}H_{20}N_3O$ $[M + H]^+$ *m/z* 282.1606; found *m/z* 282.1606.

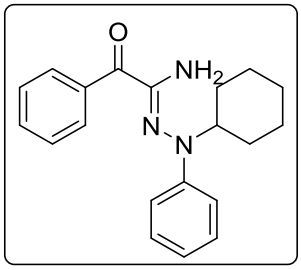


(Z)-N'-Butyl-2-oxo-N',2-diphenylacetohydrazonamide **(53):** Yellow liquid: 83% yield (24.6 mg); IR (KBr, cm^{-1}) 3487, 3372, 3058, 2958, 2871, 1624, 1542, 1490, 1377, 1226, 1179, 984, 811, 753; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.31 (d, *J* = 8 Hz, 2H), 7.56 (t, *J* = 8 Hz, 1H), 7.44 (t, *J* = 8 Hz, 2H), 7.25 (t, *J* = 8 Hz, 2H), 6.95-6.91 (m, 3H), 5.25 (s, 2H), 3.52 (t, *J* = 8 Hz, 2H), 1.70-1.63 (m, 2H), 1.43-1.34 (m, 2H), 0.94 (t, *J* = 4 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.8, 152.4, 148.5, 135.6, 133.0, 131.5, 129.0, 127.9, 121.6, 118.3, 58.0, 30.1, 20.3, 14.0; MS (EI, 70 eV) *m/z* 295, 235, 190, 149, 105; HRMS (ESI) calcd for $C_{18}H_{22}N_3O$ $[M + H]^+$ *m/z* 296.1763; found *m/z* 296.1762.

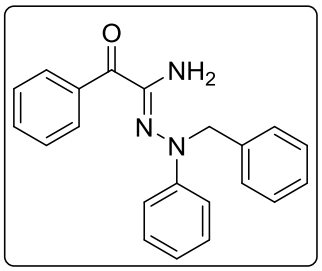


(Z)-N'-Isopropyl-2-oxo-N',2-diphenylacetohydrazonamide **(54):** Yellow liquid: 72% yield (20.2 mg); IR (KBr, cm^{-1}) 3490, 3378, 2974, 2929, 1656, 1543, 1488, 1449, 1377, 1224, 1179, 1037, 973, 815, 739; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.32 (d, *J* = 8 Hz, 2H), 7.56 (t, *J* = 8 Hz, 1H), 7.45 (t, *J* = 8 Hz, 2H), 7.27 (t, *J* = 4 Hz, *J* = 8 Hz,

2H), 7.00-6.93 (m, 3H), 5.06 (s, 2H), 3.91-3.85 (m, 1H), 1.20 (d, *J* = 4 Hz, 6H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.5, 150.8, 147.2, 136.0, 132.6, 131.4, 128.9, 127.7, 122.7, 120.9, 58.8, 19.7; MS (EI, 70 eV) *m/z* 281, 266, 176, 134, 105; HRMS (ESI) calcd for $C_{17}H_{20}N_3O$ $[M + H]^+$ *m/z* 282.1606; found *m/z* 282.1608.

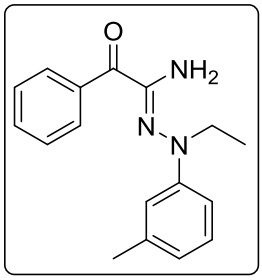


(Z)-N'-Cyclohexyl-2-oxo-N',2-diphenylacetohydrazonamide **(55):** Brown yellow solid: 78% yield (25.1 mg); mp 83-86 ^{o}C. IR (KBr, cm^{-1}) 3487, 3384, 2931, 2854, 1625, 1590, 1487, 1378, 1224, 1123, 1071, 975, 899, 734, 692; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.24 (d, *J* = 8 Hz, 2H), 7.50 (t, *J* = 8 Hz, 1H), 7.39 (t, *J* = 8 Hz, 2H), 7.19 (t, *J* = 8 Hz, 2H), 6.93-6.87 (m, 3H), 4.97 (s, 2H), 3.38-3.32 (m, 1H), 1.79-1.69 (m, 4H), 1.61-1.54 (m, 4H), 1.12-0.99 (m, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.5, 150.4, 147.1, 135.9, 132.6, 131.4, 128.9, 127.7, 122.7, 121.1, 67.3, 30.3, 26.1, 26.0; MS (EI, 70 eV) *m/z* 321, 148, 134, 107, 77; HRMS (ESI) calcd for $C_{20}H_{24}N_3O$ $[M + H]^+$ *m/z* 322.1919; found *m/z* 322.1919.

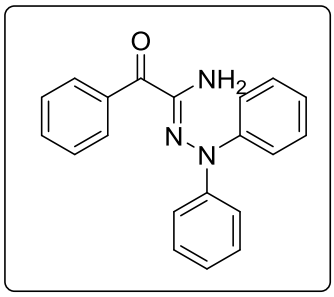


(Z)-N'-Benzyl-2-oxo-N',2-diphenylacetohydrazonamide **(56):** Yellow solid: 80% yield (26.4 mg); mp 67-70 ^{o}C. IR (KBr, cm^{-1}) 3485, 3375, 3061, 3029, 1626, 1544, 1490, 1380, 1226, 1124, 1028, 987, 807, 730; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 7.97 (d, *J* = 8 Hz, 2H), 7.50 (t, *J* = 8 Hz, 1H), 7.39-7.31 (m, 6H), 7.28-7.24 (m, 3H), 6.98-6.94 (m, 3H), 5.20 (s, 2H), 4.72 (s, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 151.7, 148.3, 138.9, 135.4, 133.0, 131.4, 129.1, 128.5, 128.2, 127.9, 127.2,

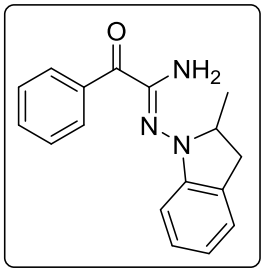
122.1, 118.2, 62.7; MS (EI, 70 eV) *m/z* 329, 252, 148, 133, 107; HRMS (ESI) calcd for $C_{21}H_{20}N_3O$ $[M + H]^+$ *m/z* 330.1606; found *m/z* 330.1601.



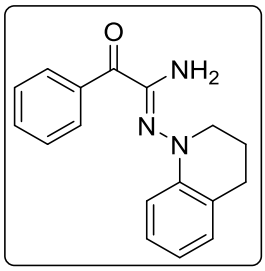
(Z)-N'-Ethyl-2-oxo-2-phenyl-N'-(p-tolyl)acetohydrazonamide **(57):** Yellow liquid: 74% yield (20.8 mg); IR (KBr, cm^{-1}) 3484, 3370, 2971, 1624, 1543, 1488, 1376, 1227, 1120, 1098, 989, 860, 773; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.25 (d, J = 8 Hz, 2H), 7.50 (t, J = 8 Hz, 1H), 7.38 (t, J =8 Hz, 2H), 7.08 (t, J = 8 Hz, 1H), 6.71-6.69 (m, 3H), 5.17 (s, 2H), 3.49 (q, J = 8 Hz, 2H), 2.24 (s, 3H), 1.15 (t, J = 4 Hz, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.8, 152.2, 148.1, 138.8, 135.6, 133.0, 131.5, 128.8, 127.9, 122.8, 119.4, 115.8, 52.4, 21.7, 12.8; MS (EI, 70 eV) *m/z* 281, 263, 207, 176, 148; HRMS (ESI) calcd for $C_{17}H_{20}N_3O$ $[M + H]^+$ *m/z* 282.1606; found *m/z* 282.1603.



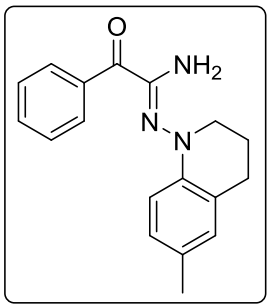
(Z)-2-Oxo-N',N',2-triphenylacetohydrazonamide **(58):** Yellow liquid: 56% yield (17.7 mg); IR (KBr, cm^{-1}) 3477, 2923, 2856, 2351, 1645, 1555, 1485, 1383, 722, 600, 539; ^1H NMR (400 MHz, $CDCl_3$, ppm) δ 8.35-8.33 (d, J = 8 Hz, 2H), 5.58 (t, J = 8 Hz, 1H), 7.47 (t, J = 8 Hz, 2H), 7.38-7.33 (m, 4H), 7.20-7.18 (m, 4H), 7.13 (t, J = 8 Hz, 2H), 5.20 (s, 2H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.0, 150.3, 147.0, 135.8, 132.9, 131.4, 129.3, 128.0, 124.0, 121.7; MS (EI, 70 eV) *m/z* 315, 148, 134, 107, 77; HRMS (ESI) calcd for $C_{20}H_{18}N_3O$ $[M + H]^+$ *m/z* 316.1450; found *m/z* 316.1447.



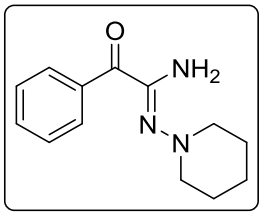
(Z)-N'-(2-Methylindolin-1-yl)-2-oxo-2-phenylacetimidamide **(59):** Yellow liquid: 73% yield (20.4 mg); IR (KBr, cm^{-1}) 3483, 3366, 3070, 2926, 1664, 1543, 1459, 1380, 1227, 1178, 1020, 982, 824, 750, 689; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.30 (d, J = 8 Hz, 2H), 7.55 (t, J = 8 Hz, 1H), 7.42 (t, J = 8 Hz, 2H), 7.15-7.07 (m, 2H), 6.83 (t, J = 8 Hz, 1H), 6.50 (d, J = 8 Hz, 1H), 5.56 (s, 2H), 4.07-3.98 (m, 1H), 3.17 (dd, J = 8 Hz, 1H), 2.69 (dd, J = 8 Hz, 1H), 1.31 (d, J = 4 Hz, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.2, 155.4, 150.0, 135.3, 133.3, 131.6, 129.7, 128.0, 127.1, 124.5, 120.6, 110.8, 65.0, 36.3, 19.2; MS (EI, 70 eV) *m/z* 279, 264, 247, 132, 105; HRMS (ESI) calcd for $C_{17}H_{18}N_3O$ $[M + H]^+$ *m/z* 280.1450; found *m/z* 280.1449.



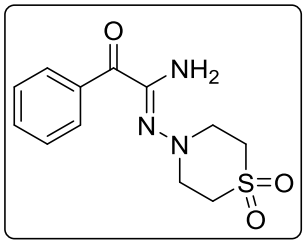
(Z)-N'-(3,4-Dihydroquinolin-1(2H)-yl)-2-oxo-2-phenylacetimidamide **(60):** Yellow solid: 76% yield (21.2 mg); mp 82-85 ^{o}C. IR (KBr, cm^{-1}) 3472, 3361, 3069, 2928, 2846, 1657, 1543, 1450, 1385, 1227, 1127, 1097, 981, 815, 752, 689; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.34 (d, J = 8 Hz, 2H), 7.55 (d, J = 8 Hz, 1H), 7.43 (t, J = 8 Hz, 2H), 7.10-7.02 (m, 2H), 6.82-6.79 (m, 2H), 5.54 (s, 2H), 3.34 (t, J = 4 Hz, 2H), 2.87 (t, J = 8 Hz, 2H), 2.18-2.12 (m, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 188.2, 155.7, 146.5, 135.3, 133.3, 131.6, 128.9, 128.0, 126.7, 124.6, 120.1, 116.1, 48.6, 27.1, 22.4; MS (EI, 70 eV) *m/z* 279, 218, 174, 149, 132; HRMS (ESI) calcd for $C_{17}H_{18}N_3O$ $[M + H]^+$ *m/z* 280.1450; found *m/z* 280.1449.



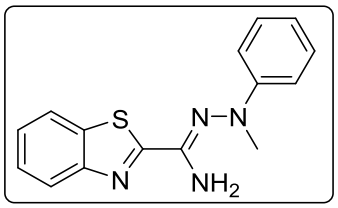
(Z)-N'-(6-Methyl-3,4-dihydroquinolin-1(2H)-yl)-2-oxo-2-phenylacetimidamide **(61):** Yellow solid: 75% yield (22.1 mg); mp 90-93 ^{o}C. IR (KBr, cm^{-1}) 3473, 3359, 2926, 2854, 1602, 1543, 1499, 1362, 1288, 1180, 1095, 980, 810, 723; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.37 (d, J = 8 Hz, 2H), 7.57 (t, J = 8 Hz, 1H), 7.44 (t, J = 8 Hz, 2H), 6.90 (d, J = 8 Hz, 2H), 6.74 (d, J = 8 Hz, 1H), 5.55 (s, 2H), 3.33 (t, J = 4 Hz, 2H), 2.86 (t, J = 4 Hz, 2H), 2.28 (s, 3H), 2.18-2.12 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 188.2, 155.5, 144.3, 135.4, 133.2, 131.6, 129.5, 129.5, 128.0, 127.3, 124.7, 116.4, 48.7, 27.0, 22.5, 20.5; MS (EI, 70 eV) *m/z* 293, 227, 148, 133, 107; HRMS (ESI) calcd for $C_{18}H_{20}N_3O$ $[M + H]^+$ *m/z* 294.1606; found *m/z* 294.1608.



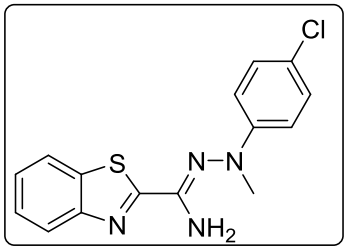
(Z)-2-Oxo-2-phenyl-N'-(piperidin-1-yl)acetimidamide **(62):** Yellow solid: 35% yield (8.1 mg); mp 91-93 ^{o}C. IR (KBr, cm^{-1}) 3393, 2932, 2875, 1648, 1594, 1445, 1361, 1226, 1032, 954, 862, 736, 689; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.21 (d, J = 8 Hz, 2H), 7.54 (t, J = 8 Hz, 1H), 7.41 (d, J = 8 Hz, 2H), 5.45 (s, 2H), 2.81 (t, J = 4 Hz, 4H), 1.73-1.67 (m, 4H), 1.53-1.47 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 188.7, 152.8, 135.4, 133.1, 131.6, 127.9, 54.8, 25.5, 24.1; MS (EI, 70 eV) *m/z* 231, 188, 149, 126, 105; HRMS (ESI) calcd for $C_{13}H_{18}N_3O$ $[M + H]^+$ *m/z* 232.1450; found *m/z* 232.1447.



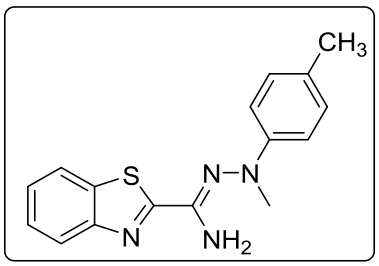
(Z)-N'-(1,1-Dioxidothiomorpholino)-2-oxo-2-phenylacetimidamide **(63):** Yellow solid: 33% yield (9.3 mg); mp 158-161 ^{o}C. IR (KBr, cm^{-1}) 3459, 3346, 2935, 2842, 1666, 1564, 1457, 1387, 1265, 1117, 1034, 989, 861, 726; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.12 (d, J = 8 Hz, 2H), 7.58 (t, J = 8 Hz, 1H), 7.43 (t, J = 8 Hz, 2H), 5.66 (s, 2H), 3.50-3.48 (m, 4H), 3.20-3.17 (m, 4H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 187.8, 154.1, 134.9, 133.5, 131.1, 128.1, 51.1, 48.6; MS (EI, 70 eV) *m/z* 281, 267, 176, 149, 134; HRMS (ESI) calcd for $C_{12}H_{16}N_3O_3S$ $[M + H]^+$ *m/z* 282.0912; found *m/z* 282.0917.



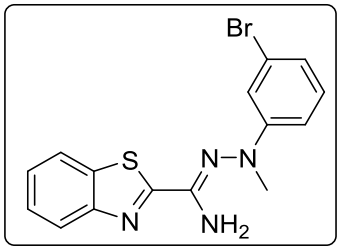
(Z)-N'-Methyl-N'-phenylbenzo[d]thiazole-2-carbohydrazonamide **(65):** Yellow liquid: 72% yield (20.3 mg); IR (KBr, cm^{-1}) 3478, 3363, 2923, 2873, 1629, 1598, 1493, 1376, 1281, 1108, 975, 851, 755, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.06 (d, J = 8 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7.54-7.44 (m, 2H), 7.29 (t, J = 8 Hz, 2H), 7.03 (d, J = 8 Hz, 2H), 6.92 (t, J = 8 Hz, 1H), 5.83 (s, 2H), 3.16 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 163.1, 153.6, 152.8, 150.9, 135.9, 128.9, 126.4, 126.3, 123.6, 122.0, 120.4, 115.6, 41.0; MS (EI, 70 eV) *m/z* 282, 254, 237, 212, 178; HRMS (ESI) calcd for $C_{15}H_{15}N_4S$ $[M + H]^+$ *m/z* 283.1017; found *m/z* 283.1020.



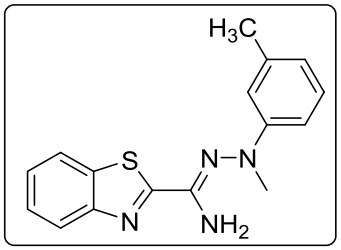
(Z)-N'-(4-Chlorophenyl)-N'-methylbenzo[d]thiazole-2-carbohydrazonamide **(66):** Yellow solid: 30% yield (9.5 mg); mp 104-107 ^{o}C. IR (KBr, cm^{-1}) 3330, 2921, 1714, 1616, 1555, 1491, 1386, 1297, 1098, 942, 824, 756; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7,52-7.44 (m, 2H), 7.23 (t, J = 8 Hz, 2H), 6.93 (d, J = 8 Hz, 2H), 5.85 (s, 2H), 3.13 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 162.7, 153.8, 152.7, 149.6, 135.8, 129.5, 128.7, 126.5, 125.3, 123.6, 122.0, 116.7, 40.7; MS (EI, 70 eV) *m/z* 316, 271, 178, 161, 135; HRMS (ESI) calcd for $C_{15}H_{14}ClN_4S$ $[M + H]^+$ *m/z* 317.0628; found *m/z* 317.0638.



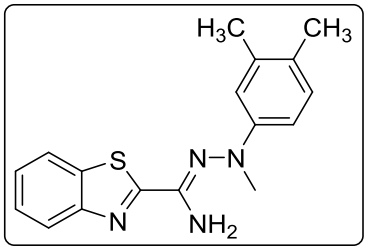
(Z)-N'-Methyl-N'-(p-tolyl)benzo[d]thiazole-2-carbohydrazonamide **(67):** Yellow solid: 64% yield (18.9 mg); mp 110-113 ^{o}C. IR (KBr, cm^{-1}) 3477, 3361, 2920, 1625, 1510, 1455, 1312, 1280, 1111, 1056, 927, 809, 760; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 1H), 7.92 (d, J = 8 Hz, 1H), 7.54-7.43 (m, 2H), 7.10 (d, J = 8 Hz, 2H), 6.94 (d, J = 8 Hz, 2H), 5.80 (s, 2H), 3.13 (s, 3H), 2.30 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 163.3, 153.2, 152.8, 148.7, 135.8, 129.9, 129.4, 126.3, 126.3, 123.6, 122.0, 116.0, 41.6, 20.6; MS (EI, 70 eV) *m/z* 296, 251, 178, 161, 135; HRMS (ESI) calcd for $C_{16}H_{17}N_4S$ $[M + H]^+$ *m/z* 297.1174; found *m/z* 297.1176.



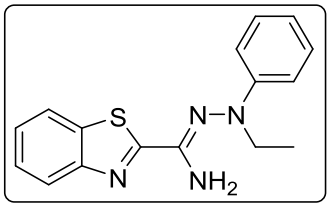
(Z)-N'-(3-Bromophenyl)-N'-methylbenzo[d]thiazole-2-carbohydrazonamide **(68):** Yellow liquid: 61% yield (22.0 mg); IR (KBr, cm^{-1}) 3364, 2922, 1627, 1588, 1477, 1313, 1180, 1055, 986, 947, 820, 760; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 1H), 7.94 (d, J = 8 Hz, 1H), 7.53-7.45 (m, 2H), 7.15-7.10 (m, 2H), 7.03-6.99 (m, 1H), 6.93-6.90 (m, 1H), 5.86 (s, 2H), 3.13 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 162.8, 154.2, 152.7, 152.2, 135.8, 130.1, 126.5, 126.5, 123.7, 123.0, 123.0, 122.0, 118.3, 113.8, 40.0; MS (EI, 70 eV) *m/z* 360, 281, 178, 161, 135; HRMS (ESI) calcd for $C_{15}H_{14}BrN_4S$ $[M + H]^+$ *m/z* 361.0123; found *m/z* 361.0121.



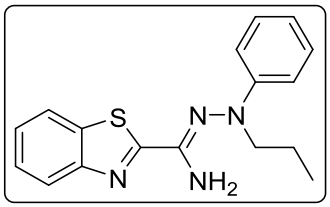
(Z)-N'-Methyl-N'-(m-tolyl)benzo[d]thiazole-2-carbohydrazonamide **(69):** Yellow liquid: 62% yield (18.1 mg); IR (KBr, cm^{-1}) 3477, 3363, 2918, 2870, 1601, 1555, 1488, 1312, 1243, 1110, 996, 870, 761; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7.54-7.44 (m, 2H), 7.17 (t, J = 8 Hz, 1H), 6.84-6.82 (m, 2H), 6.75 (d, J = 8 Hz, 1H), 5.82 (s, 2H), 3.16 (s, 3H), 2.33 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 163.2, 153.5, 152.8, 150.9, 138.6, 135.8, 128.7, 126.4, 126.3, 123.6, 122.0, 121.3, 116.4, 112.7, 41.1, 21.8; MS (EI, 70 eV) *m/z* 296, 251, 178, 161, 135; HRMS (ESI) calcd for $C_{16}H_{17}N_4S$ $[M + H]^+$ *m/z* 297.1174; found *m/z* 297.1175.



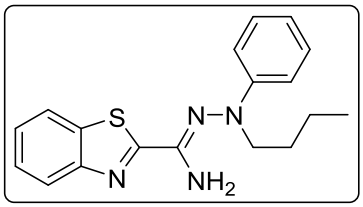
(Z)-N'-(3,4-Dimethylphenyl)-N'-methylbenzo[d]thiazole-2-carbohydrazonamide **(70):** Yellow solid: 47% yield (14.6 mg); mp 110-113 ^{o}C. IR (KBr, cm^{-1}) 3473, 3361, 2920, 2868, 1626, 1502, 1455, 1313, 1274, 1097, 960, 851, 761; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7.54-7.43 (m, 2H), 7.05 (d, J = 8 Hz, 1H), 6.89-6.79 (m, 2H), 5.80 (s, 2H), 3.14 (s, 3H), 2.25 (s, 3H), 2.22 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 163.4, 153.0, 152.9, 149.1, 137.0, 135.9, 130.0, 128.8, 126.3, 126.2, 123.6, 122.0, 117.7, 113.5, 41.9, 20.2, 18.9; MS (EI, 70 eV) *m/z* 310, 265, 178, 161, 133; HRMS (ESI) calcd for $C_{17}H_{19}N_4S$ $[M + H]^+$ *m/z* 311.1330; found *m/z* 311.1335.



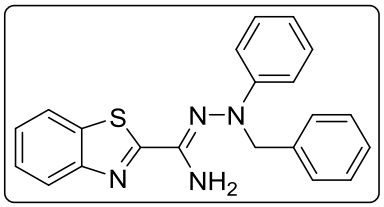
(Z)-N'-Ethyl-N'-phenylbenzo[d]thiazole-2-carbohydrazonamide **(71):** Yellow liquid: 51% yield (15.1 mg); IR (KBr, cm^{-1}) ; 3485, 3371. 2970, 1630, 1554, 1490, 1380, 1244, 1119, 901, 755, 693; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.03 (d, J = 8 Hz, 1H), 7.92 (d, J = 8 Hz, 1H), 7.50-7.42 (m, 2H), 7.28-7.24 (m, 2H), 7.00 (d, J = 8 Hz, 2H), 6.89 (t, J = 8 Hz, 1H), 5.73 (s, 2H), 3.58 (q, J = 8 Hz, 2H), 1.23 (t, J = 8 Hz, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 163.3, 153.2, 152.9, 149.2, 135.9, 129.0, 126.3, 126.3, 123.6, 121.9, 120.6, 116.9, 50.4, 12.4; MS (EI, 70 eV) *m/z* 296, 252, 177, 161, 135; HRMS (ESI) calcd for $C_{16}H_{17}N_4S$ $[M + H]^+$ *m/z* 297.1174; found *m/z* 297.1183.



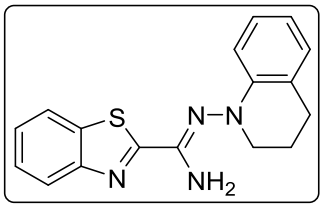
(Z)-N'-Phenyl-N'-propylbenzo[d]thiazole-2-carbohydrazonamide **(72):** Yellow solid: 31% yield (9.6 mg); mp 79-82 ^{o}C.IR (KBr, cm^{-1}) 3485, 3373, 2961, 2872, 1630, 1554, 1488, 1313, 1229, 1031, 943, 877, 756; ^{1}H NMR (400 MHz, $CDCl_{3,}$ ppm) δ 8.03 (d, *J* = 8 Hz, 1H), 7.92 (d, *J* = 8 Hz, 1H), 7.52-7.42 (m ,2H), 7.25 (t, *J* = 8 Hz, 2H), 6.98 (d, *J* = 8 Hz, 2H), 6.89 (t, *J* = 8 Hz, 1H), 5.89 (s, 2H), 3.48 (t, *J* = 8 Hz, 2H), 1.76-1.67 (m, 2H), 0.97 (t, *J* = 8 Hz, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 163.2, 152.9, 152.8, 149.4, 135.9, 129.0, 126.3, 126.2, 123.6, 121.9, 120.6, 116.7, 58.7, 20.9, 11.8; MS (EI, 70 eV) *m/z* 310, 281, 264, 177, 161; HRMS (ESI) calcd for $C_{17}H_{19}N_4S$ $[M + H]^+$ *m/z* 311.1330; found *m/z* 311.1339.



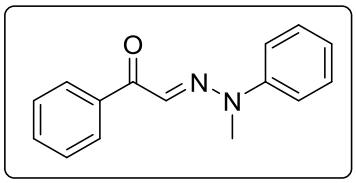
(Z)-N'-Butyl-N'-phenylbenzo[d]thiazole-2-carbohydrazonamide **(73):** Yellow liquid: 63% yield (20.4 mg); IR (KBr, cm^{-1}) 3483, 3373, 3067, 2957, 1629, 1596, 1489, 1313, 1211, 1116, 1033, 996, 879, 758; ^{1}H NMR (400 MHz, $CDCl_{3,}$ ppm) δ 8.02 (d, *J* = 8 Hz, 1H), 7.90 (d, *J* = 8 Hz, 1H), 7.51-7.40 (m, 2H), 7.25 (t, *J* = 8 Hz, 2H), 6.98 (d, *J* = 8 Hz, 2H), 6.88 (t, *J* = 8 Hz, 1H), 5.70 (s, 2H), 3.51 (t, *J* = 8 Hz, 2H), 1.71-1.63 (m, 2H), 1.45-1.35 (m, 2H), 0.94 (t, *J* = 8 Hz, 3H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$, ppm) δ 163.3, 152.9, 152.9, 149.4, 135.9, 129.0, 126.3, 126.3, 123.6, 121.9, 120.6, 116.7, 56.5, 29.8, 20.5, 14.1; MS (EI, 70 eV) *m/z* 324, 281, 264, 177, 135; HRMS (ESI) calcd for $C_{18}H_{21}N_4S$ $[M + H]^+$ *m/z* 325.1487; found *m/z* 325.1496.



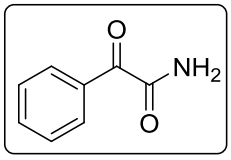
(Z)-N'-Benzyl-N'-phenylbenzo[d]thiazole-2-carbohydrazonamide **(74):** Yellow liquid: 41% yield (14.7 mg); IR (KBr, cm^{-1}) 3481, 3374, 3060, 2923, 1631, 1596, 1495, 1314, 1230, 1118, 1034, 936, 868, 757, 696; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.04 (d, J = 8 Hz, 1H), 7.94 (d, J = 8 Hz, 1H), 7.54-7.45 (m, 4H), 7.37-7.27 (m, 5H), 7.08 (d, J = 8 Hz, 2H), 6.96 (t, J = 8 Hz, 1H), 5.66 (s, 2H), 4.76 (s, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 163.1, 152.8, 152.4, 149.4, 138.8, 136.0, 129.0, 128.4, 128.1, 127.1, 126.3, 126.3, 123.6, 121.9, 120.9, 116.6, 61.2; MS (EI, 70 eV) *m/z* 358, 281, 251, 161, 109; HRMS (ESI) calcd for $C_{21}H_{19}N_4S$ $[M + H]^+$ *m/z* 359.1330; found *m/z* 359.1334.



(Z)-N'-(3,4-Dihydroquinolin-1(2H)-yl)benzo[d]thiazole-2-carboximidamide **(75):** Green solid: 46% yield (14.2 mg); mp 134-137 ^{o}C. IR (KBr, cm^{-1}) 3309, 2927, 2843, 1623, 1598, 1488, 1384, 1276, 1125, 945, 810, 749; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.05 (d, J = 8 Hz, 1H), 7.92 (d, J = 8 Hz, 1H), 7.53-7.43 (m, 2H), 7.10-7.01 (m, 2H), 6.85-6.78 (m, 2H), 5.85 (s, 2H), 3.34 (t, J = 4 Hz, 2H), 2.88 (t, J = 4 Hz, 2H), 2.21-2.15 (m, 2H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 163.1, 154.7, 152.8, 146.7, 135.8, 128.9, 126.8, 126.4, 126.4, 124.4, 123.6, 122.0, 119.9, 115.8, 49.0, 27.0, 22.5; MS (EI, 70 eV) *m/z* 308, 281, 207, 177, 130; HRMS (ESI) calcd for $C_{17}H_{17}N_4S$ $[M + H]^+$ *m/z* 309.1174; found *m/z* 309.1183.



(E)-2-(2-Methyl-2-phenylhydrazono)-1-phenylethan-1-one **(H)**:[4] ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.06 (d, J = 8.0 Hz, 2H), 7.54 (t, J = 8.0 Hz, 1H), 7.45 (t, J = 8 Hz, 2H), 7.36-7.32 (m, 5H), 7.09-7.05 (m, 1H), 3.47 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 190.9, 146.6, 137.8, 131.8, 130.3, 130.1, 129.3, 127.9, 123.4, 116.9, 33.9; MS (EI, 70 eV) *m*/*z* 238, 207, 167, 133, 105.



2-Oxo-2-phenylacetamide **(84):**[3] Yield 85% (0.126 g); Yellow solid; mp 70-73 ^{o}C; IR (KBr): 3393, 3209, 1663, 1594, 1577, 1234, 1097, 722; ^{1}H NMR (400 MHz, $CDCl_3$, ppm) δ 8.28 (d, J = 8 Hz, 2H), 7.63 (t, J = 4, 8 Hz, 1H), 7.48 (t, J = 8 Hz, 2H), 7.10 (s, 1H), 6.59 (s, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$, ppm) δ 187.6, 164.4, 134.6, 133.0, 131.1, 128.6; MS (EI, 70 eV) *m*/*z* 149, 123, 105, 77, 51; HRMS (ESI) calcd for $C_8H_8NO_2$ $[M + H]^+$ *m/z* 150.0555; found *m/z* 150.0553.

References

1. Kim, B.-R.; Lee, H.-G.; Kang, S.-B.; Jung, K.-J.; Sung, G.-H.; Kim, J.-J.; Lee, S.-G.; Yoon, Y.-G. Synthesis of β-Ketonitriles, α,β-Alkynones and Biscabinols from Esters Using *tert*-Butoxide-Assisted C(O)- (i.e., acyl–C) Coupling under Ambient Conditions. *Tetrahedron* **2013**, *69*, 10331-10336.

2. (a) Borikar, S. P.; Paul, V. *N*-Nitrosation of Secondary Amines Using *p*-TSA-$NaNO_2$ as a Novel Nitrosating Agent under Mild Conditions. *Synth. Commun.* **2010**, *40*, 654-660. (b) Giuseppone, N.; Schmitt, J.-L.; Schwartz, E.; Lehn, J.-M. Scandium(III) Catalysis of Transimination Reactions. Independent and Constitutionally Coupled Reversible Processes. *J. Am. Chem. Soc.* **2005**, *127*, 5528–5539. (c) Gonzâez, A. Simple Preparation of *N*-Alkyl-*N*-Arylhydrazines from

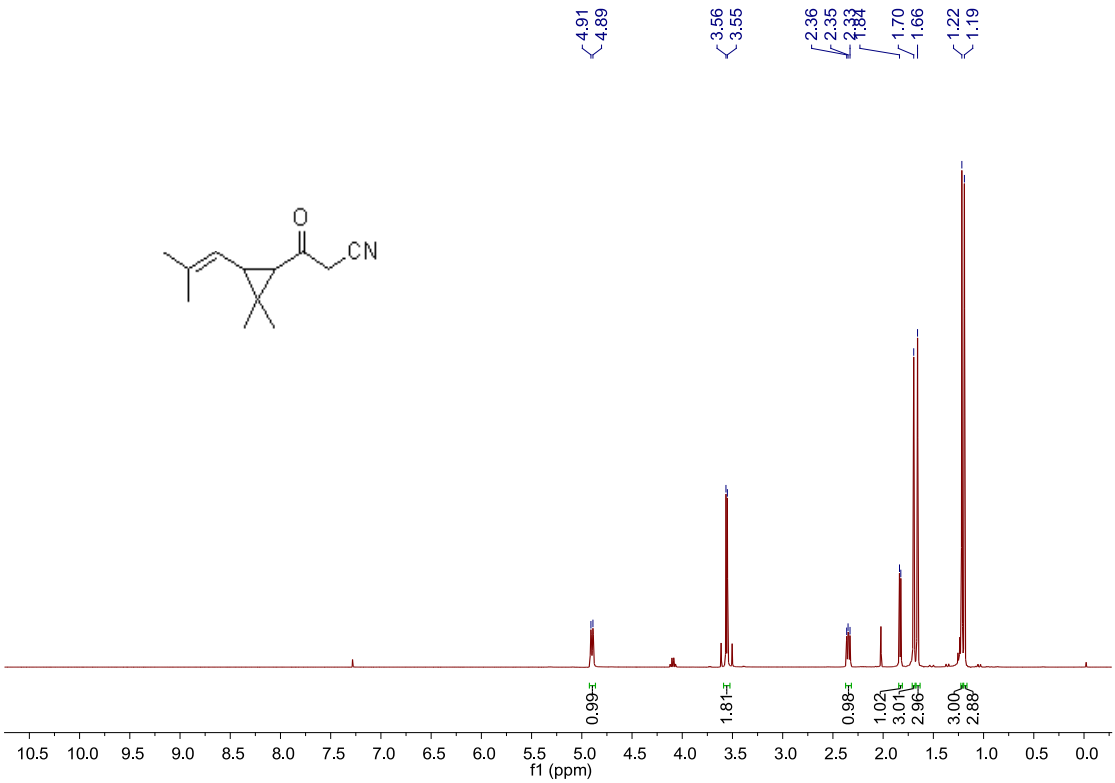
Diazotable *N*-Arylamines. *Synth. Commun*. **1988**, *18*, 1225–1229. (d) Entwistle, I. D.; Johnstone, R. A. W.; Wilby, A. H. Rapid Reduction of *N*-Nitrosamines to *N*,*N*-Disubstituted Hydrazines; the Utility of Some Low-Valent Titanium Reagents. *Tetrahedron* **1982**, *38*, 419–423. (e) Smith, R. F.; Dennis, L. A.; Ryan, W. J.; Rodriguez, G.; Brophy, K. A. Amidrazones. 14. The Formation of 1,1–Disubstituted Hydrazines from the Base-Promoted Hydrolysis of 1,l-Disubstituted-3-Amino-4,5-Dihydro-1*H*-Pyrazolium Halides: Mechanistic Considerations. *J. Heterocycl. Chem.* **1992**, *29*, 181–183. (f) Zhou, S.; Wang, J.; Zhang, F.; Song, C.; Zhu, J. A Versatile, Traceless C-H Activation-Based Approach for the Synthesis of Heterocycles. *Org. Lett.* **2016**, *18*, 2527–2430. (g) Saczewski, F.; Wasllewska, A.; Hudson, A. L.; Ferdousi, M.; Rybczynska, A.; Boblewski, K.; Lehmann, A. 1-[Imidazolin-2-yl]amino]indoline and 1-[(Imidazolin-2-yl)amino]1,2,3,4-tetrahydroquinoline Derivatives: New Insights into Their Circulatory Activities. *Acta Pol. Pharm.* **2015**, *72*, 277-287.

3. Liu, S.; Gao, Q.-H.; Wu, X.; Zhang, J.-J.; Ding, K.; Wu, A.-X. Formamidine Hydrochloride as an Amino Surrogate: I_2-Catalyzed Oxidative Amidation of Aryl Methyl Ketones Leading to Free (N–H) α-Ketoamides. *Org. Biomol. Chem.* **2015**, *13***,** 2239-2242.

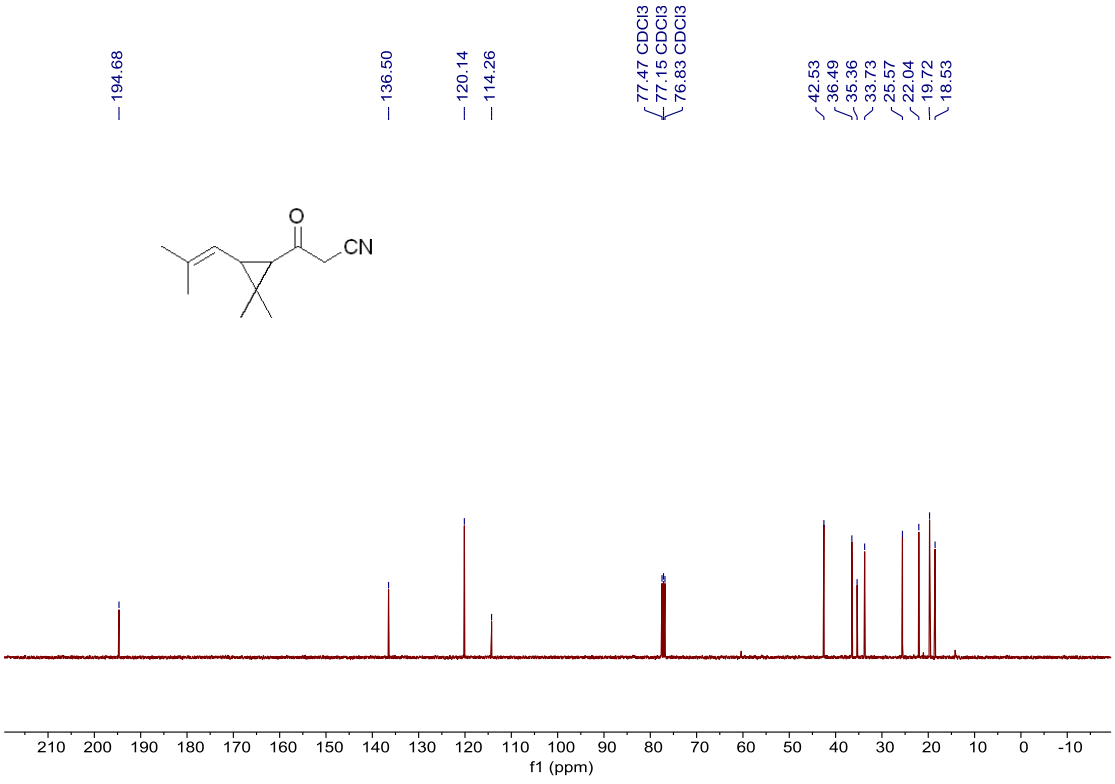
4. MöHrle, H.; Keller, G. α-Dicarbonylmonophenylhydrazone als Nucleophile und Nachbargruppen/α-Dicarbonylmonophenylhydrazones as Nucleophiles and Neighbouring Groups. *Zeitschrift Für Naturforschung* B, **2001**, *56*, 533-546.

N. ^{1}H and ^{13}C NMR Spectra of Compounds

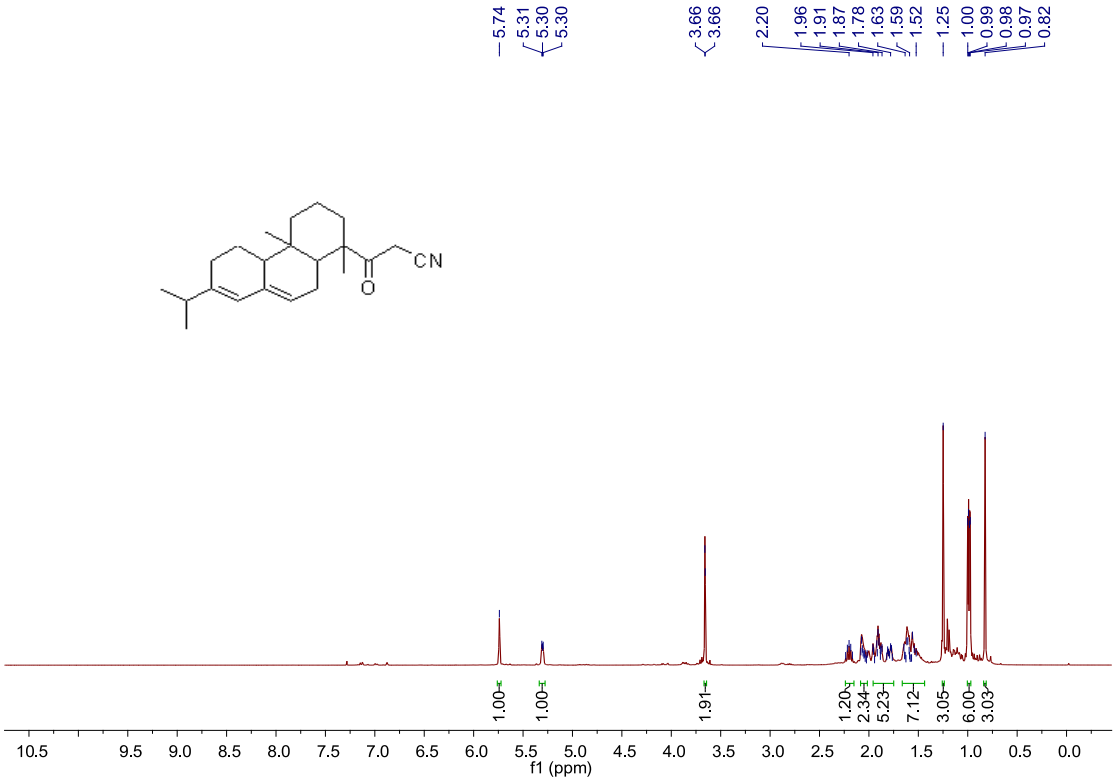
^{1}H NMR of product 1-35 in $CDCl_3$ (400 MHz)



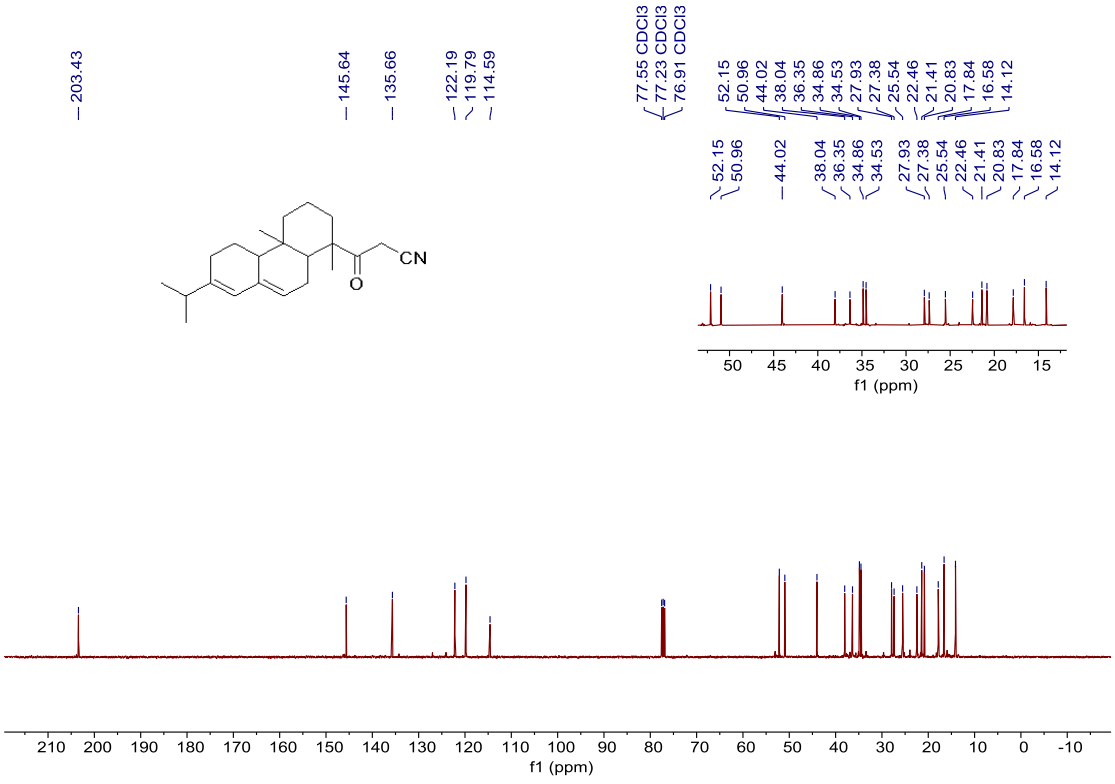
^{13}C NMR of product 1-35 in $CDCl_3$ (100 MHz)



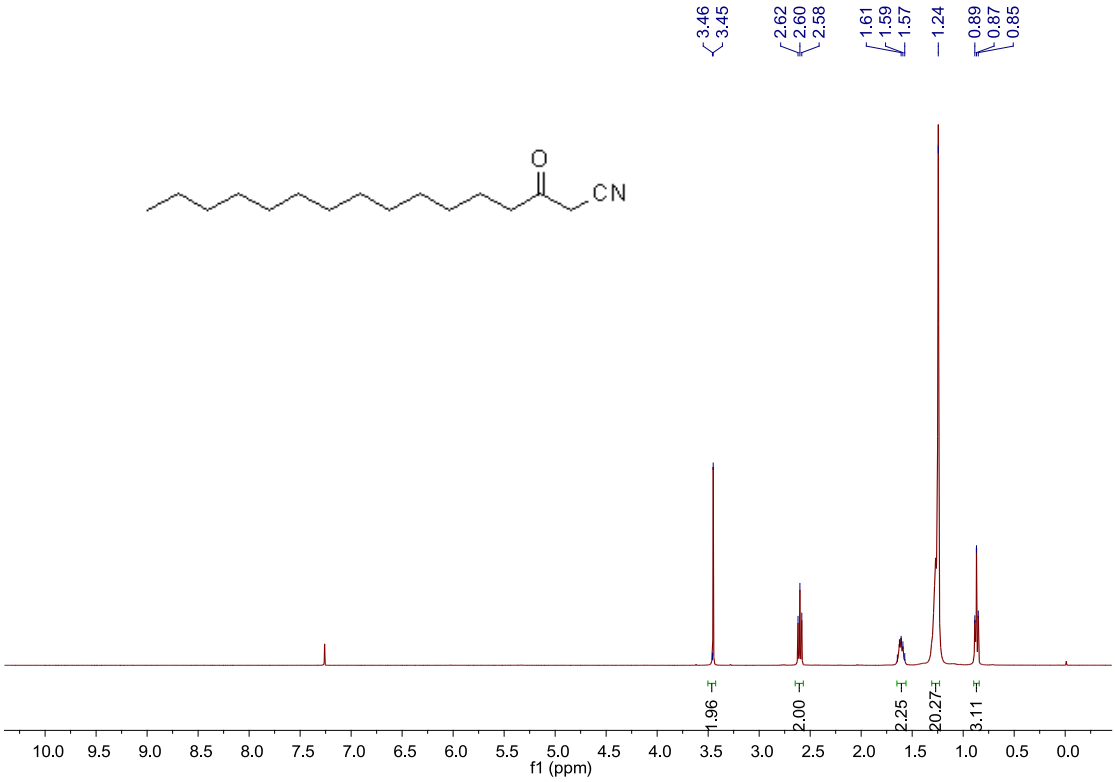
^{1}H NMR of product 1-36 in $CDCl_3$ (400 MHz)



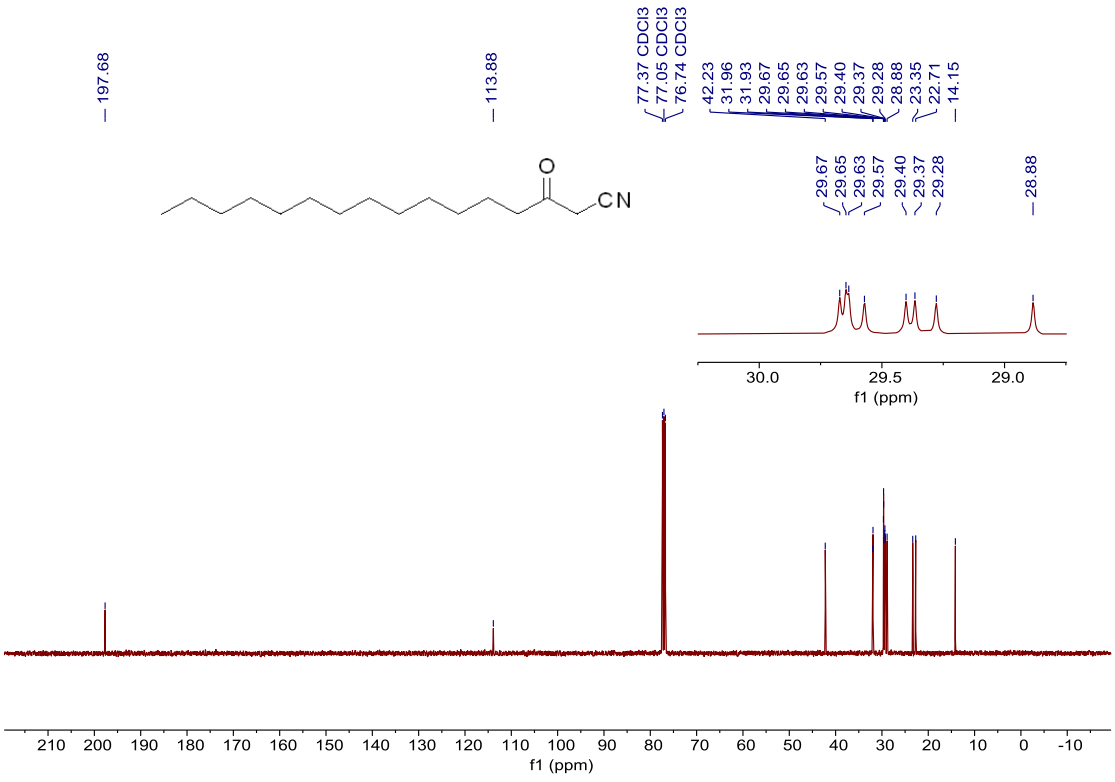
^{13}C NMR of product 1-36 in $CDCl_3$ (100 MHz)



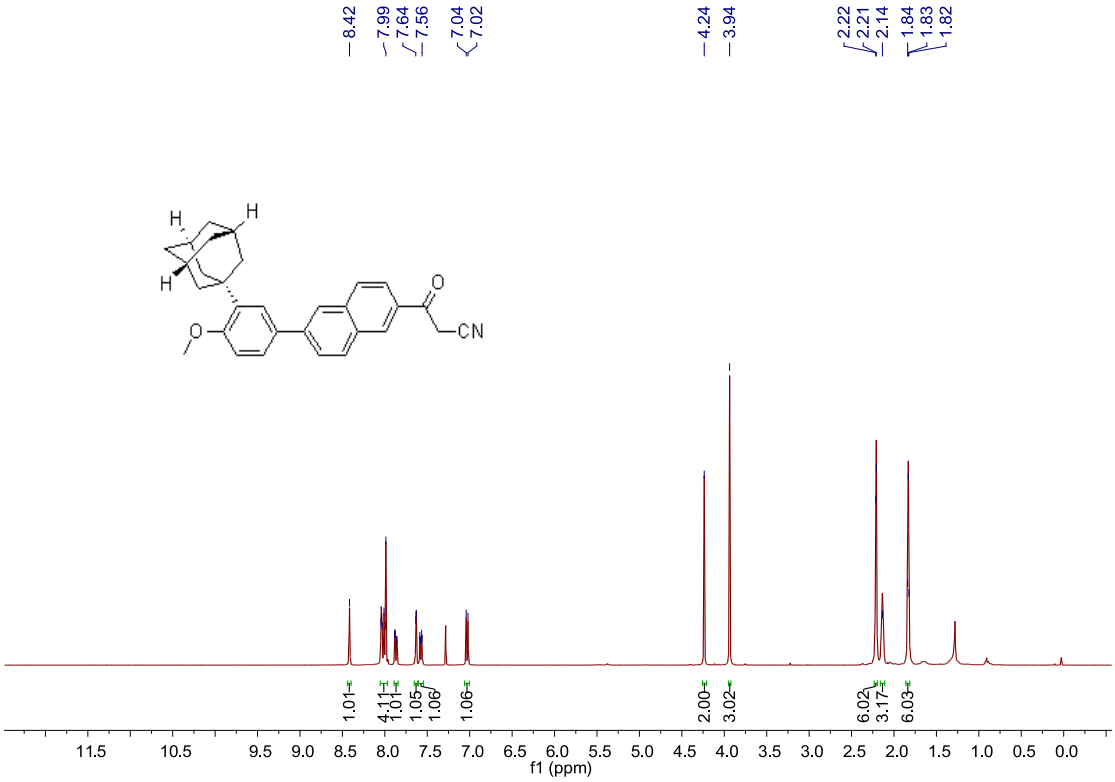
^{1}H NMR of product 1-37 in $CDCl_3$ (400 MHz)



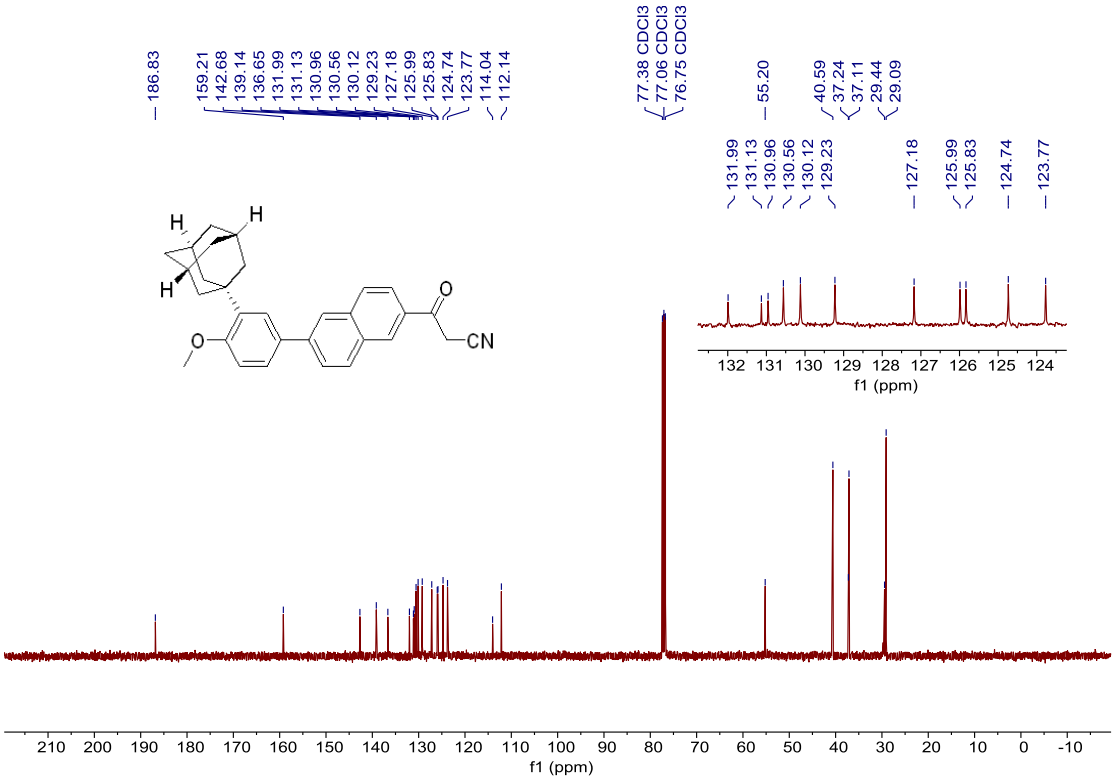
^{13}C NMR of product 1-37 in $CDCl_3$ (100 MHz)



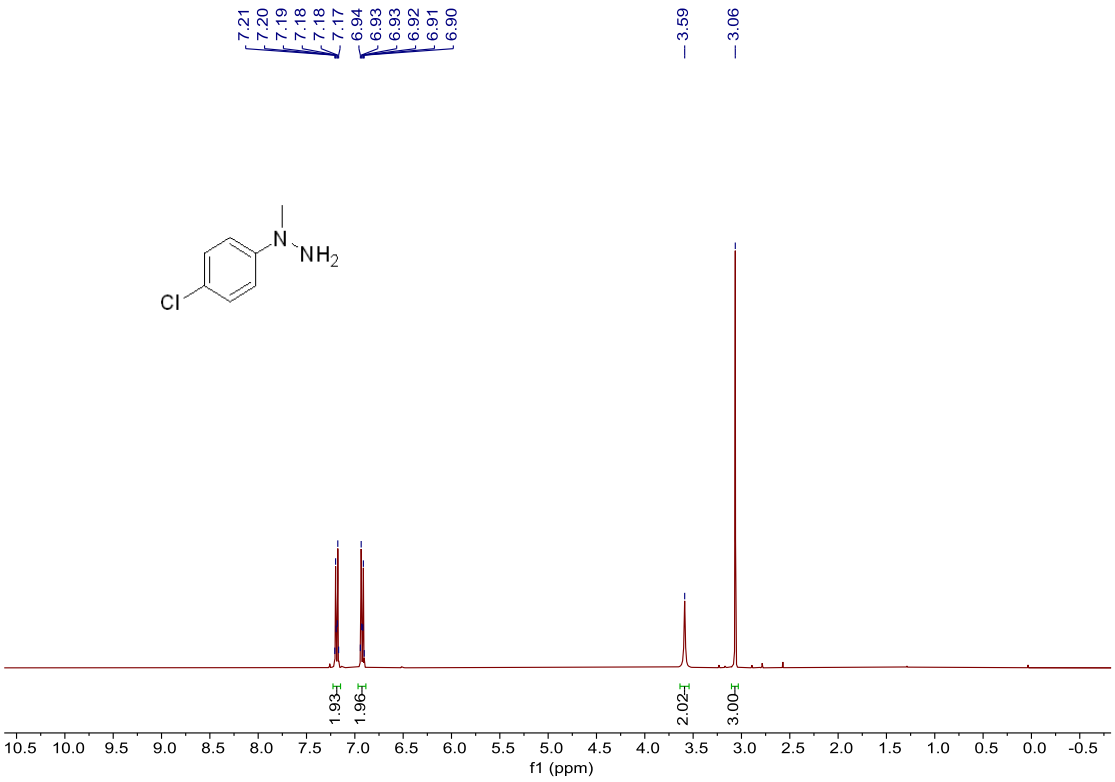
^{1}H NMR of product 1-38 in $CDCl_3$ (400 MHz)



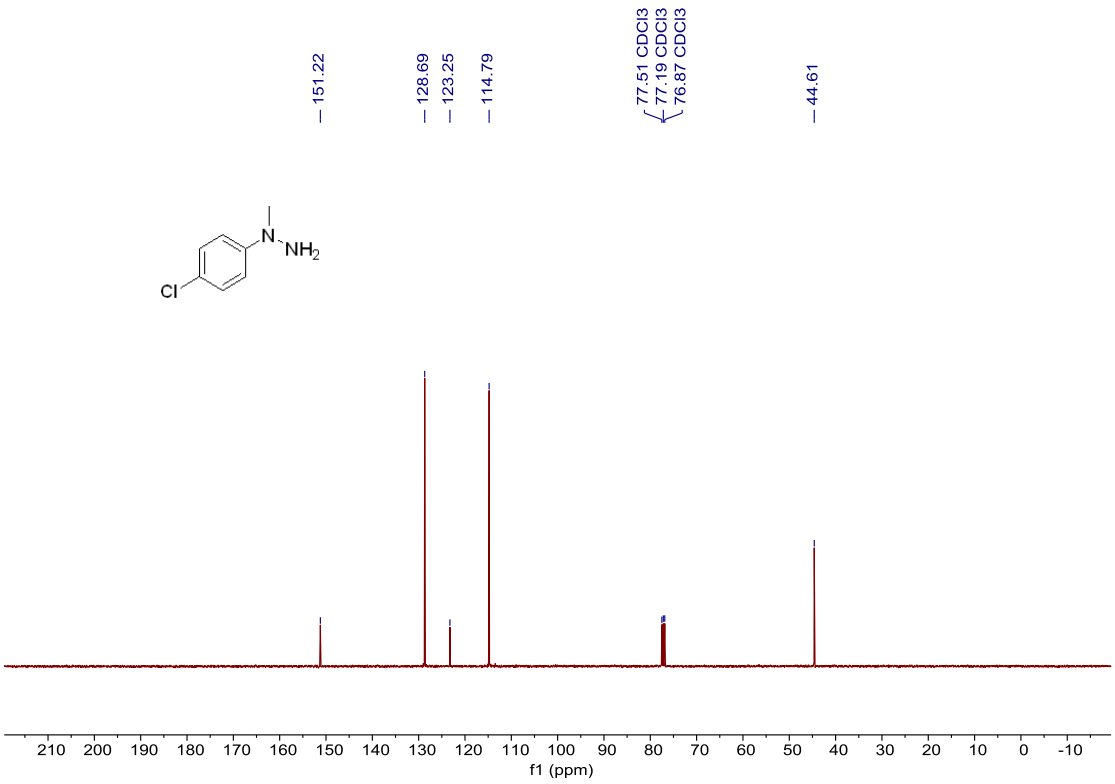
^{13}C NMR of product 1-38 in $CDCl_3$ (100 MHz)



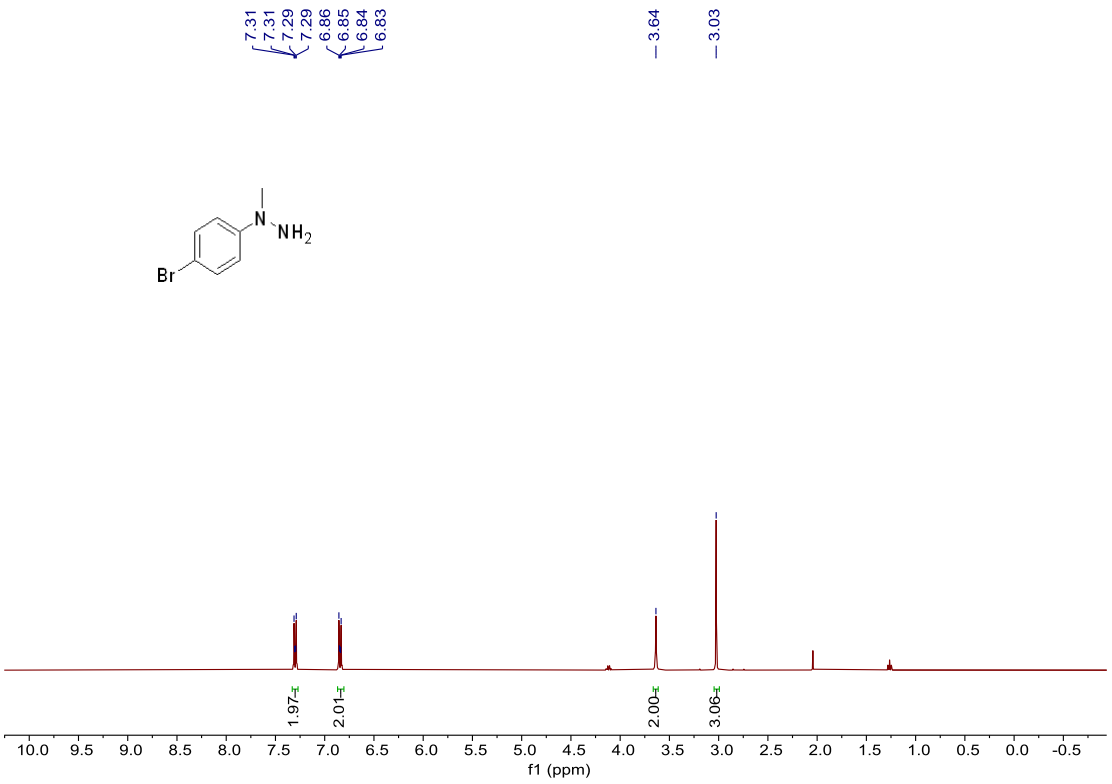
^{1}H NMR of product 2-39 in $CDCl_3$ (400 MHz)



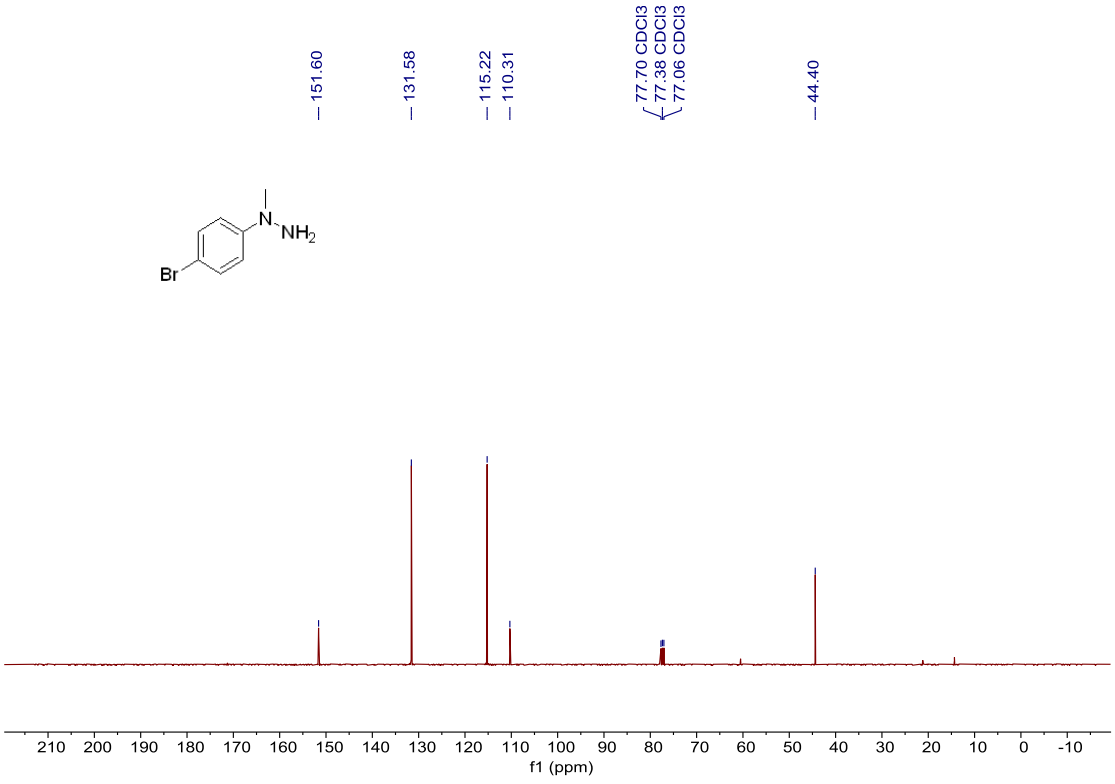
^{13}C NMR of product 2-39 in $CDCl_3$ (100 MHz)



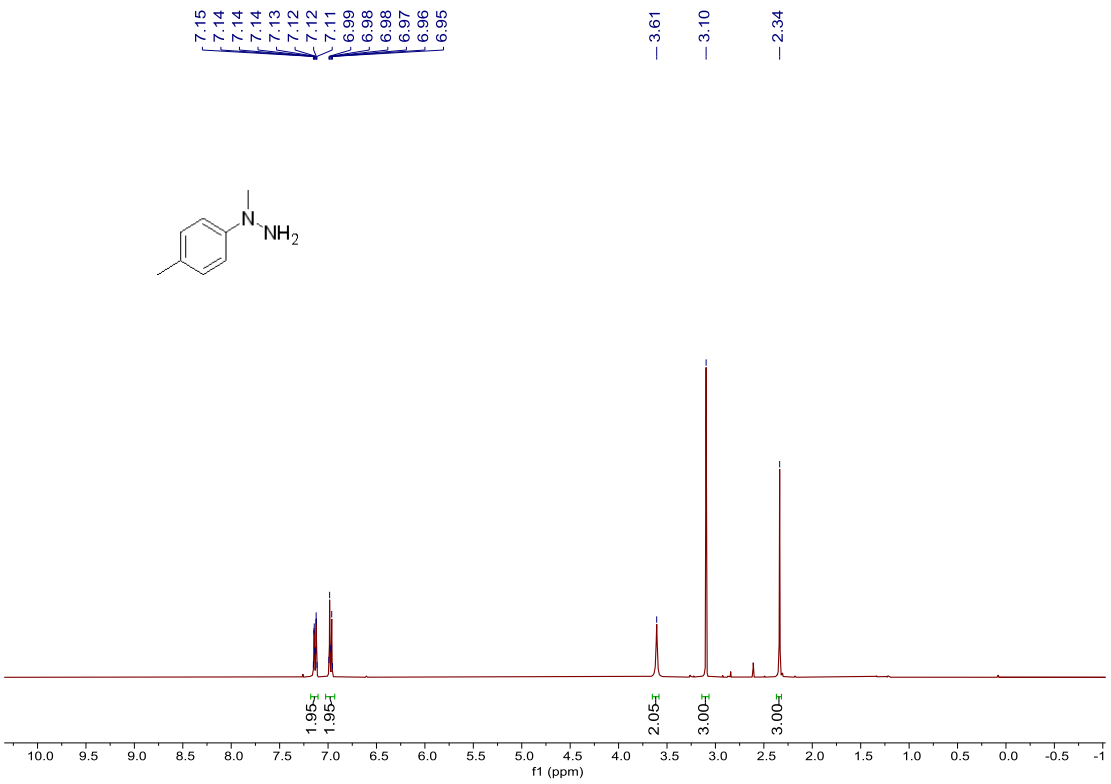
^{1}H NMR of product 2-40 in $CDCl_3$ (400 MHz)



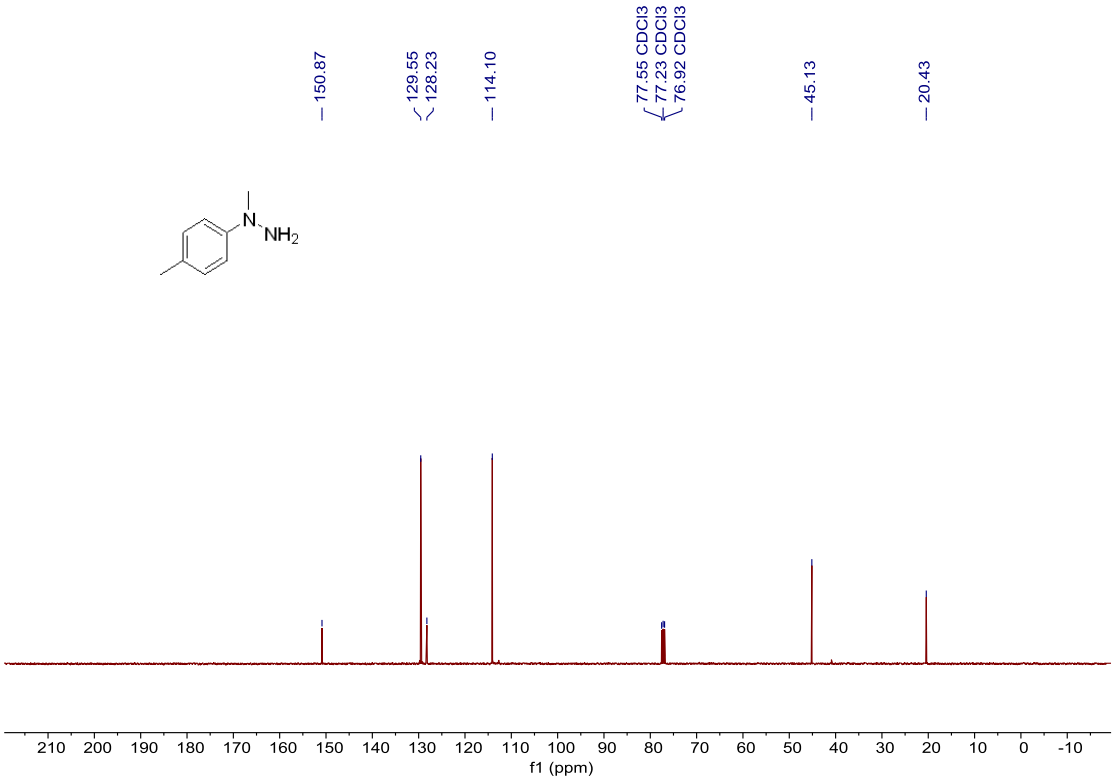
^{13}C NMR of product 2-40 in $CDCl_3$ (100 MHz)



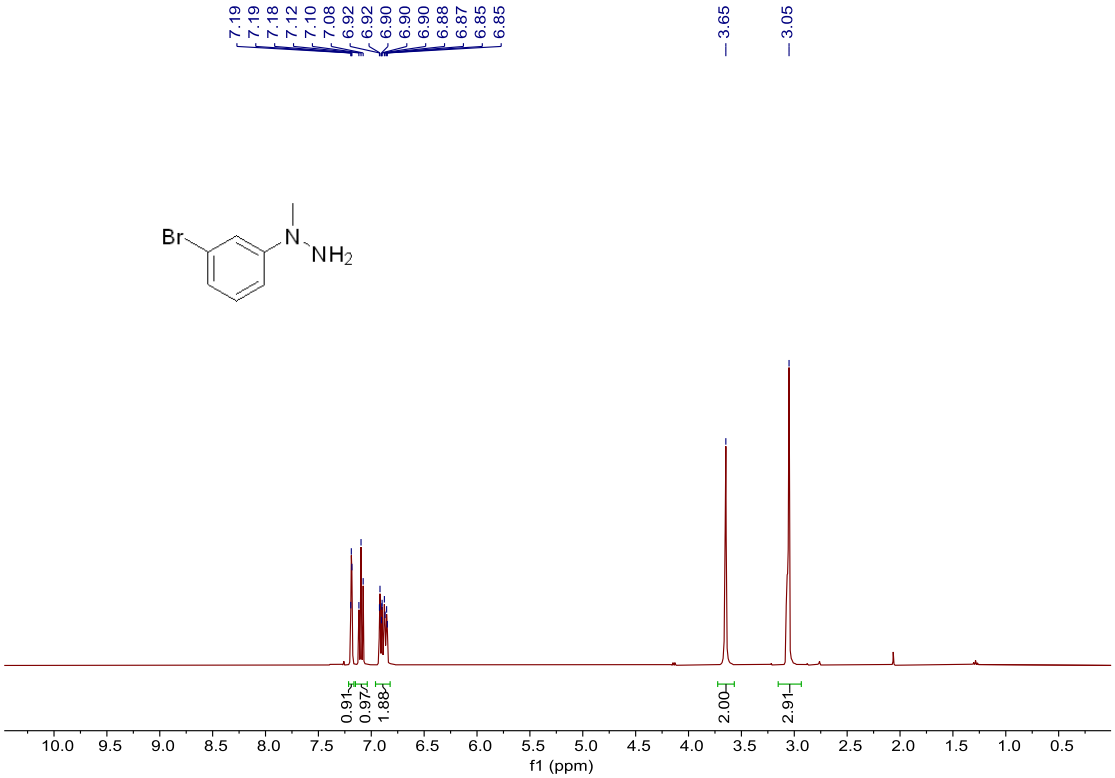
^{1}H NMR of product 2-42 in $CDCl_3$ (400 MHz)



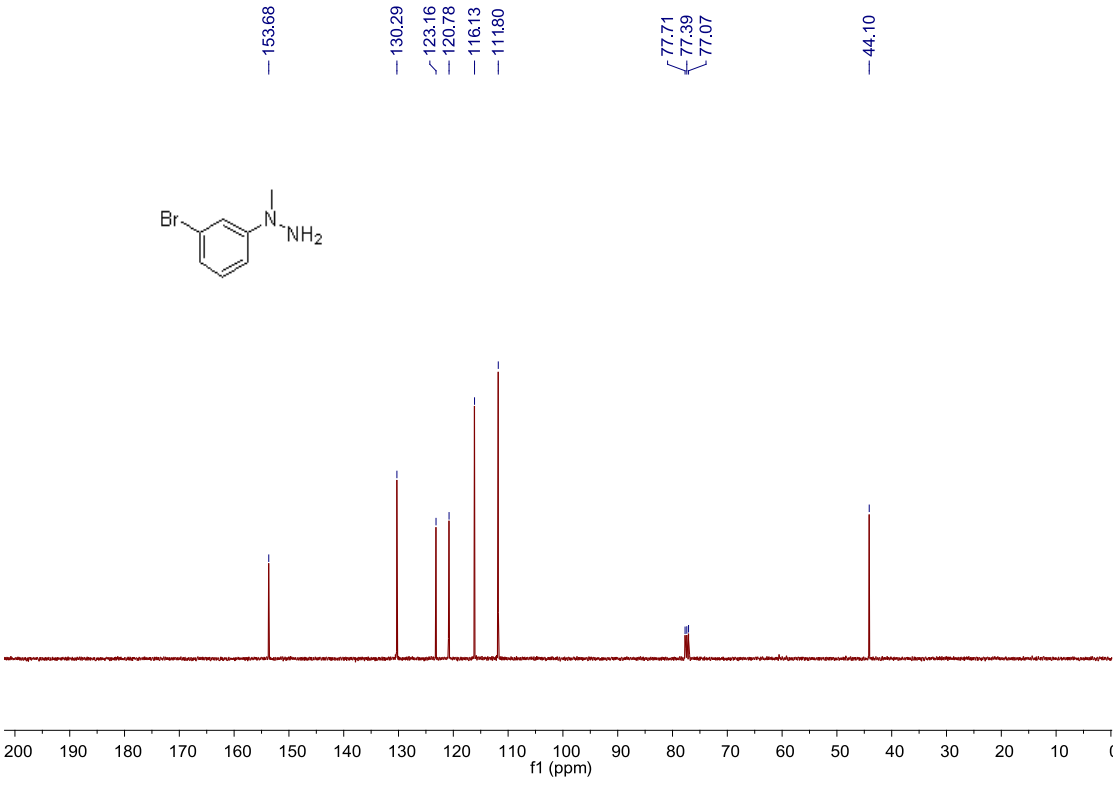
^{13}C NMR of product 2-42 in $CDCl_3$ (100 MHz)



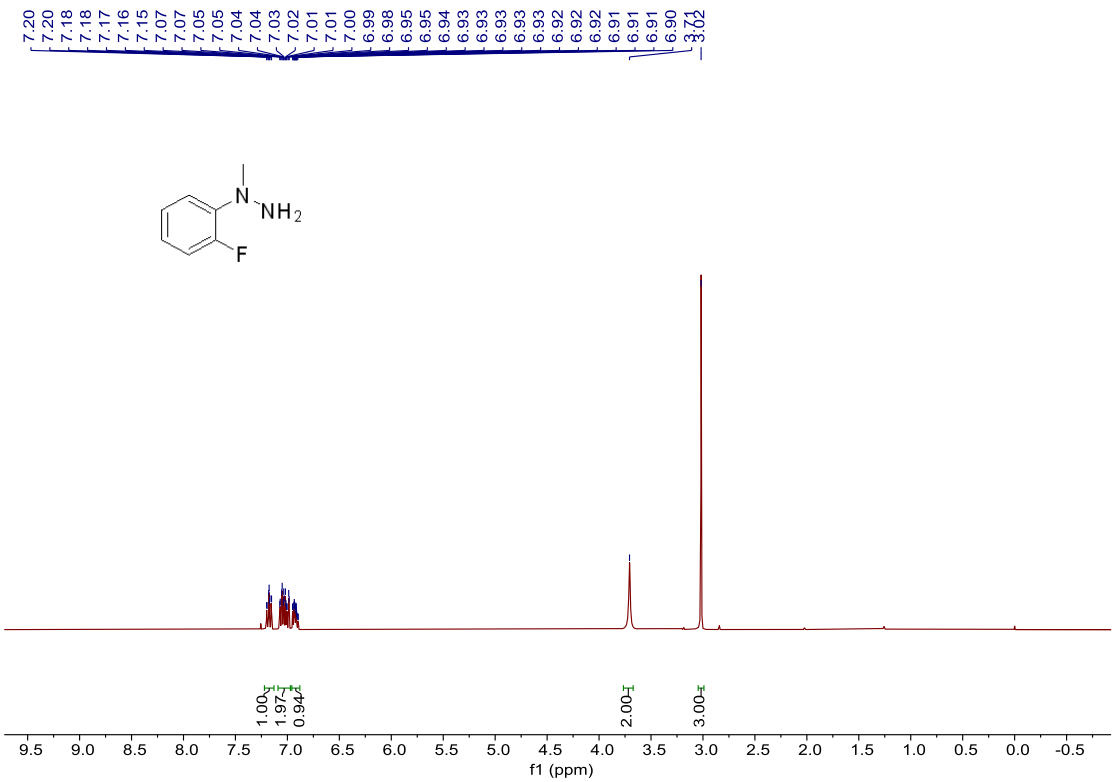
^{1}H NMR of product 2-43 in $CDCl_3$ (400 MHz)



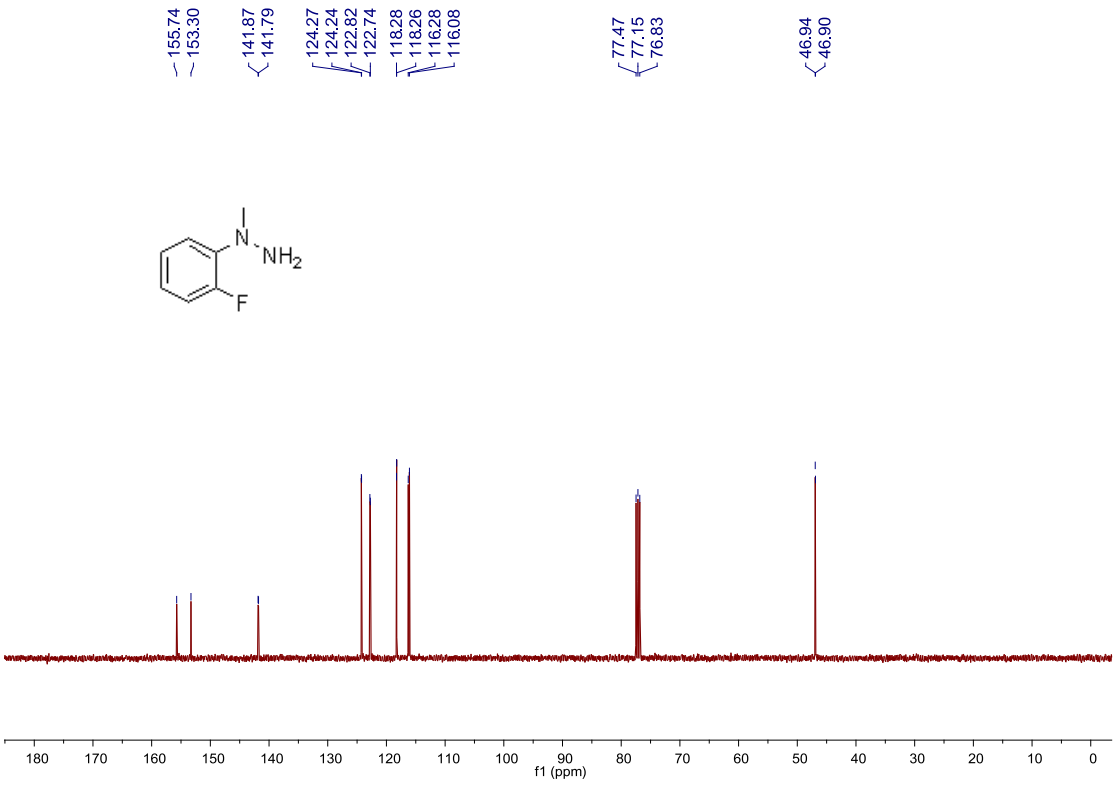
^{13}C NMR of product 2-43 in $CDCl_3$ (100 MHz)



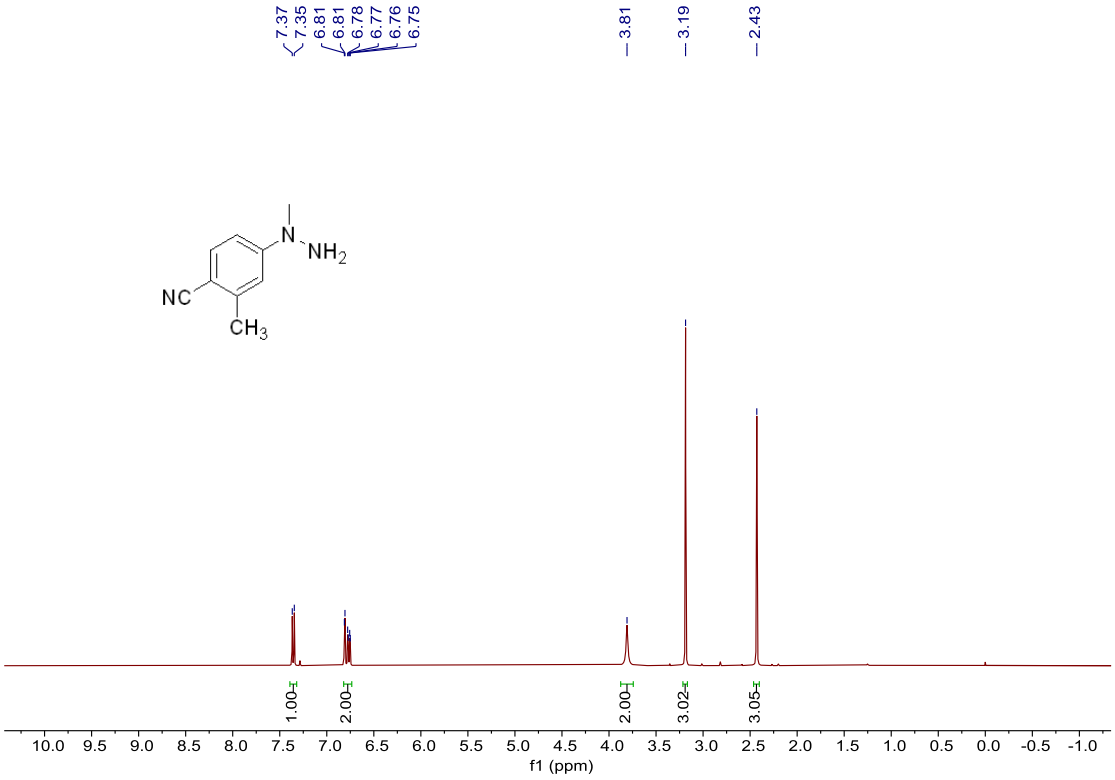
^{1}H NMR of product 2-45 in $CDCl_3$ (400 MHz)



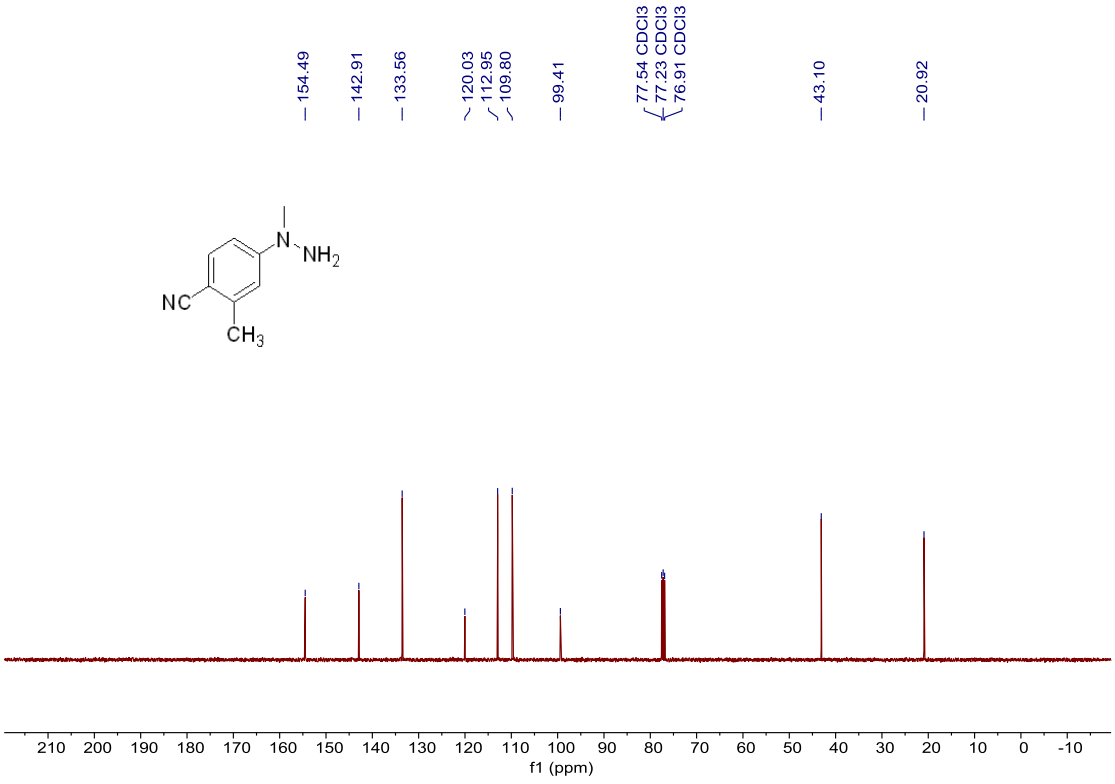
^{13}C NMR of product 2-45 in $CDCl_3$ (100 MHz)



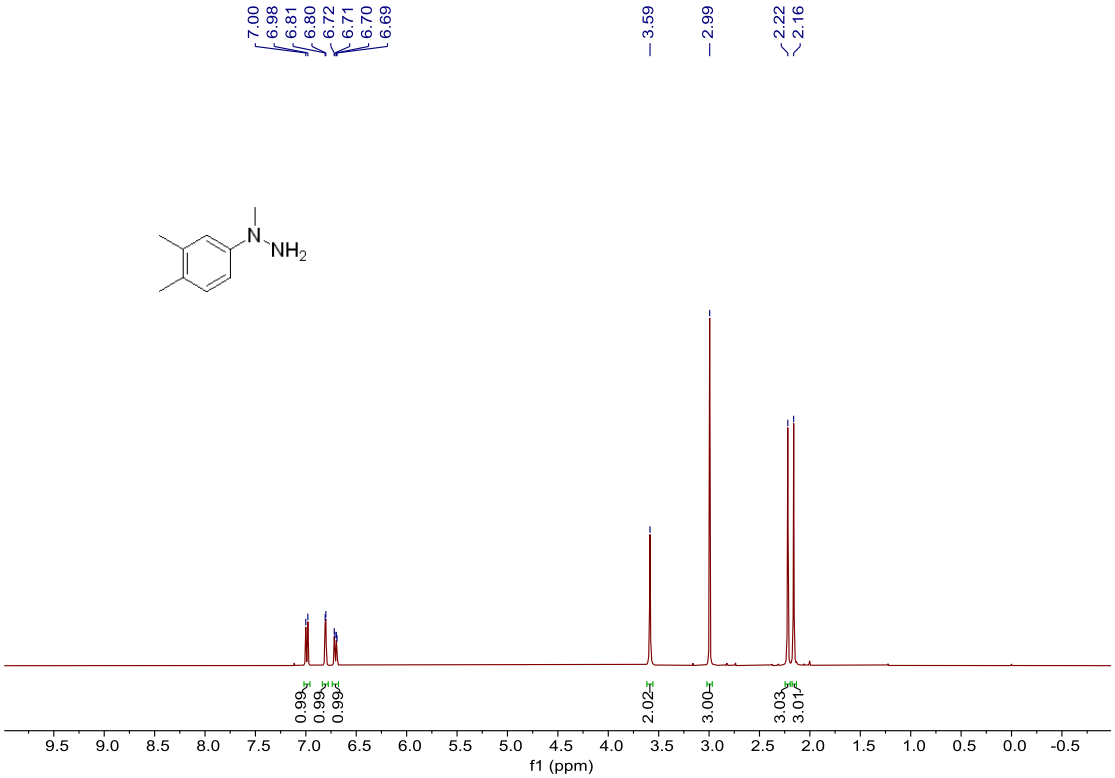
^{1}H NMR of product 2-48 in $CDCl_3$ (400 MHz)



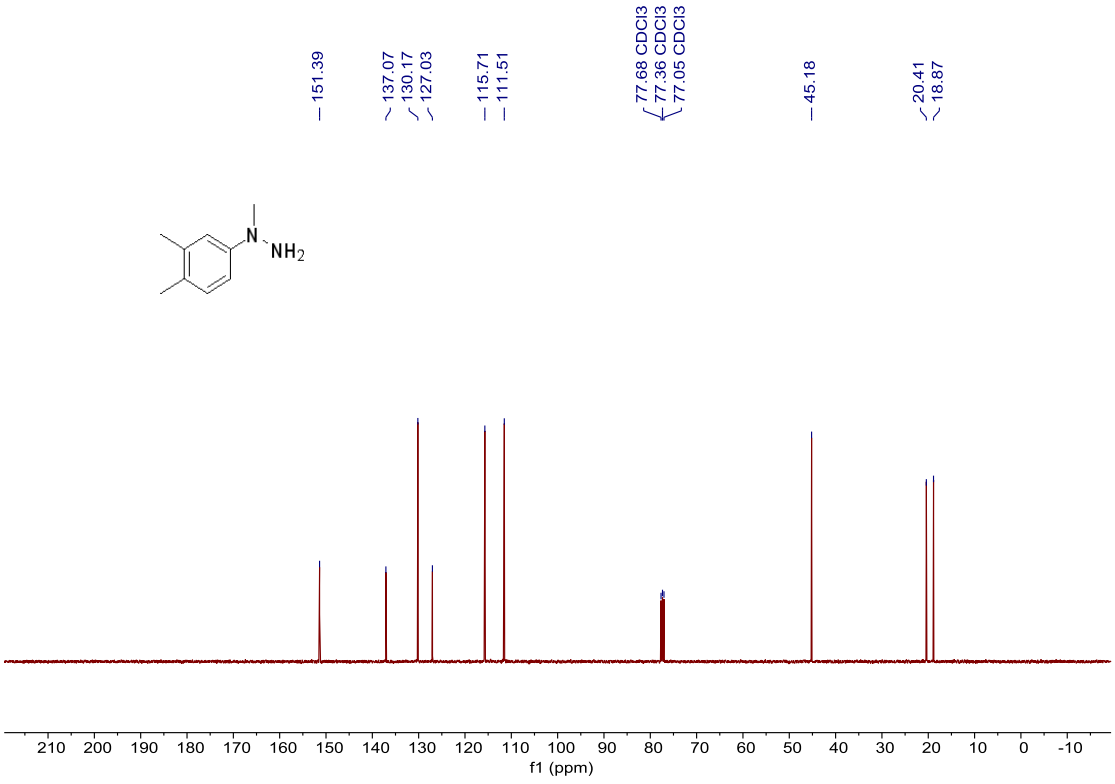
^{13}C NMR of product 2-48 in $CDCl_3$ (100 MHz)



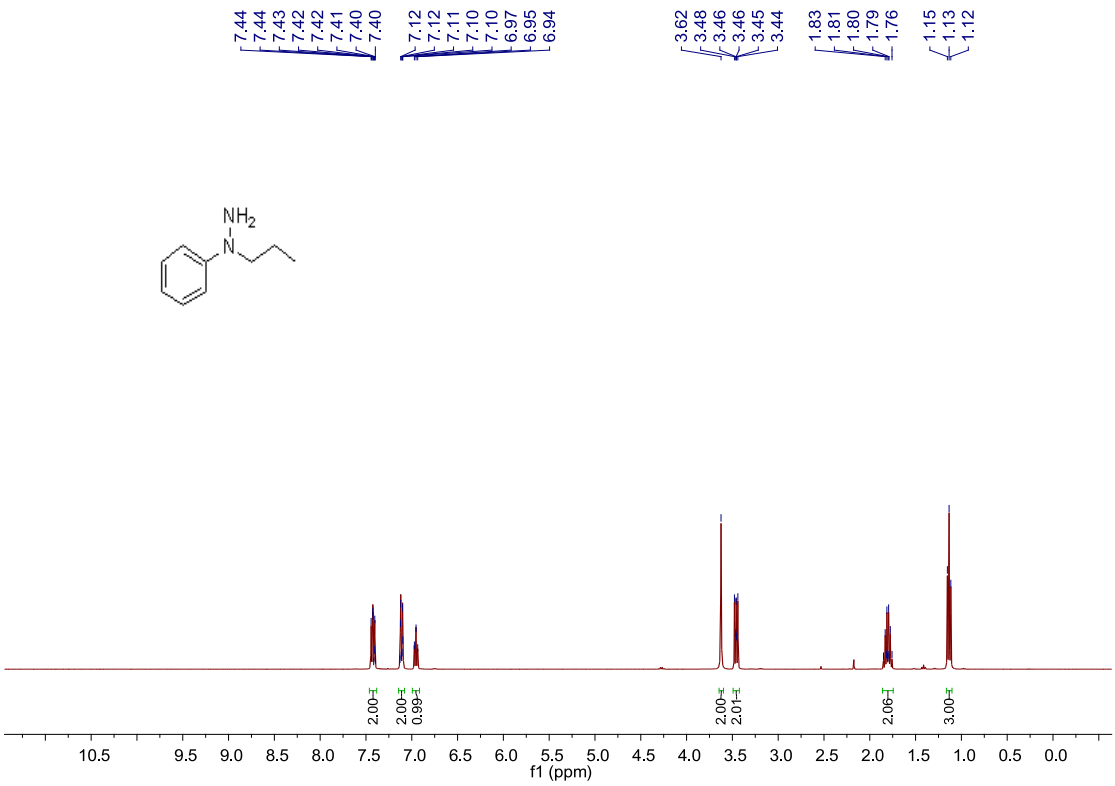
^{1}H NMR of product 2-50 in $CDCl_3$ (400 MHz)



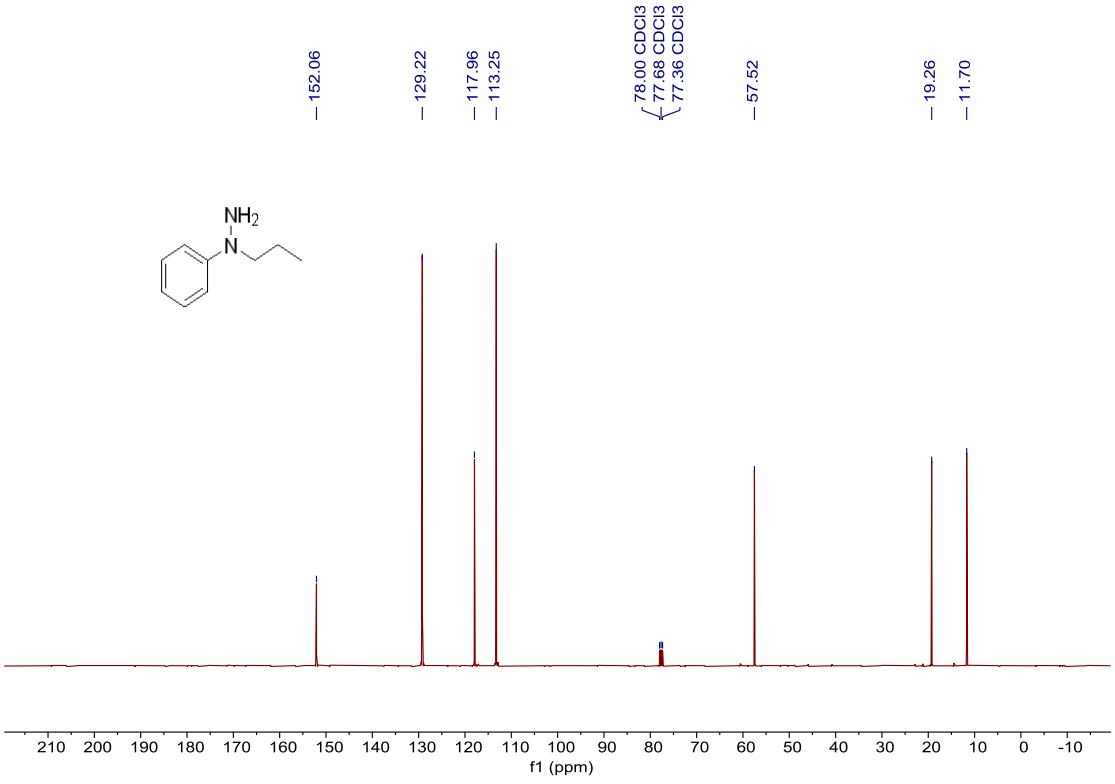
^{13}C NMR of product 2-50 in $CDCl_3$ (100 MHz)



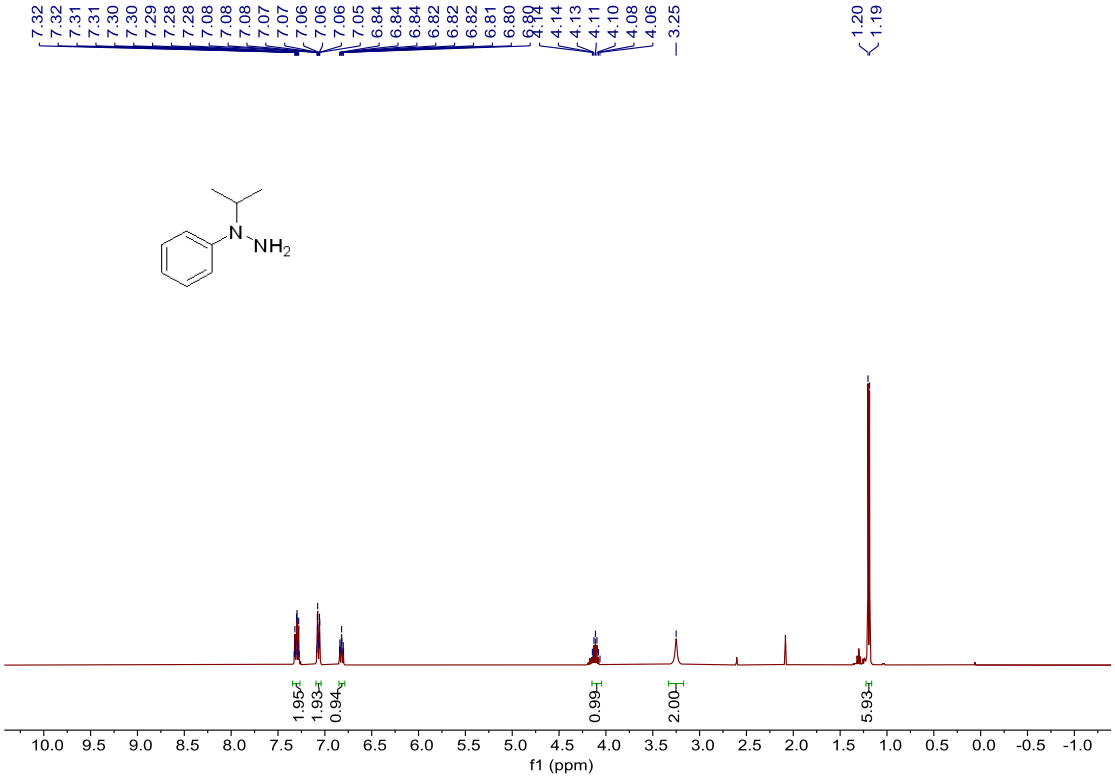
^{1}H NMR of product 2-52 in $CDCl_3$ (400 MHz)



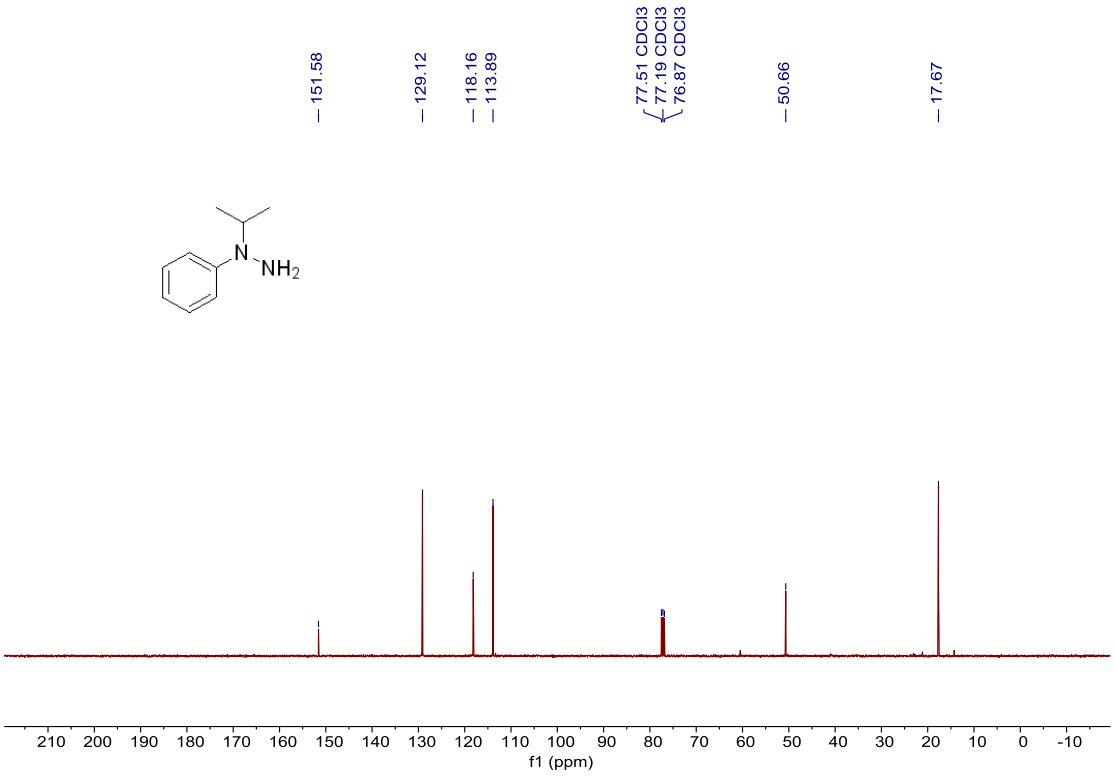
^{13}C NMR of product 2-52 in $CDCl_3$ (100 MHz)



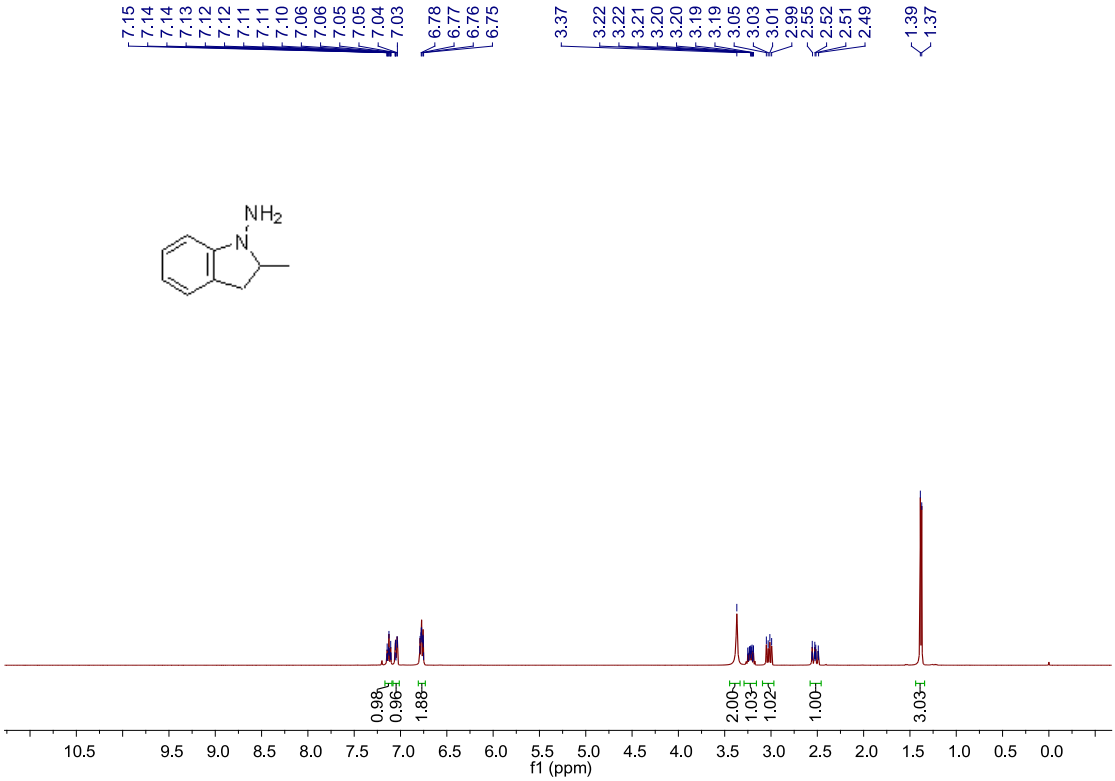
^{1}H NMR of product 2-54 in $CDCl_3$ (400 MHz)



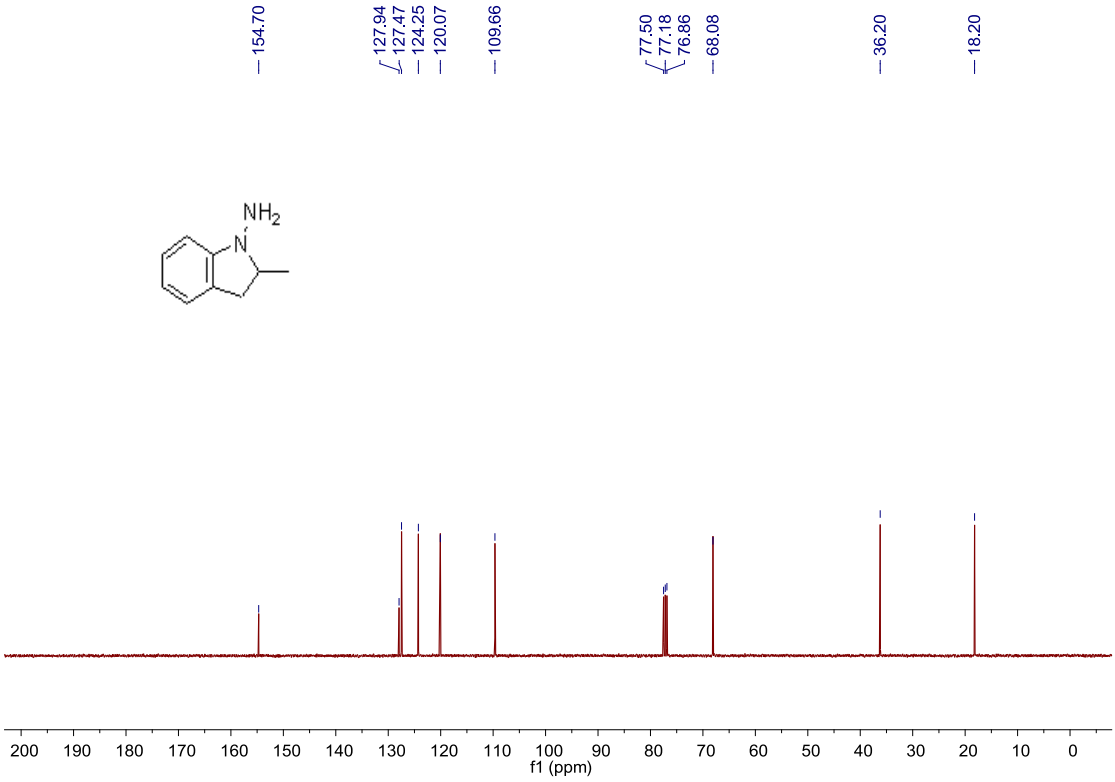
^{13}C NMR of product 2-54 in $CDCl_3$ (100 MHz)



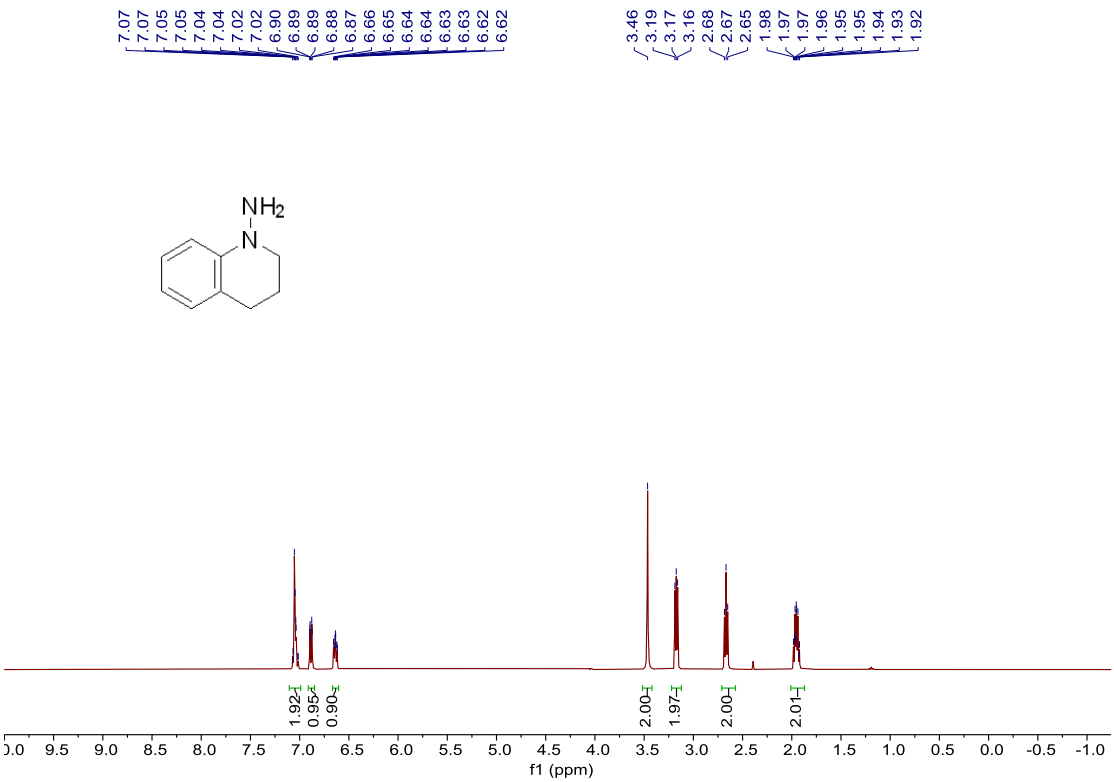
^{1}H NMR of product 2-59 in $CDCl_3$ (400 MHz)



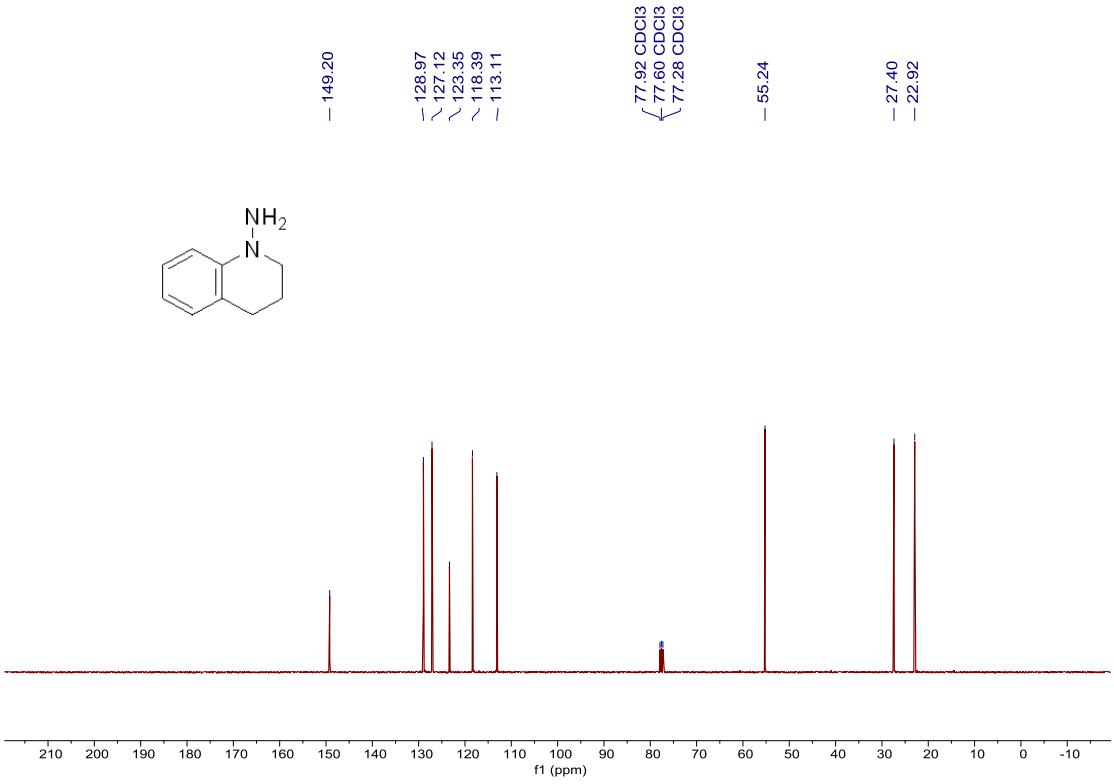
^{13}C NMR of product 2-59 in $CDCl_3$ (100 MHz)



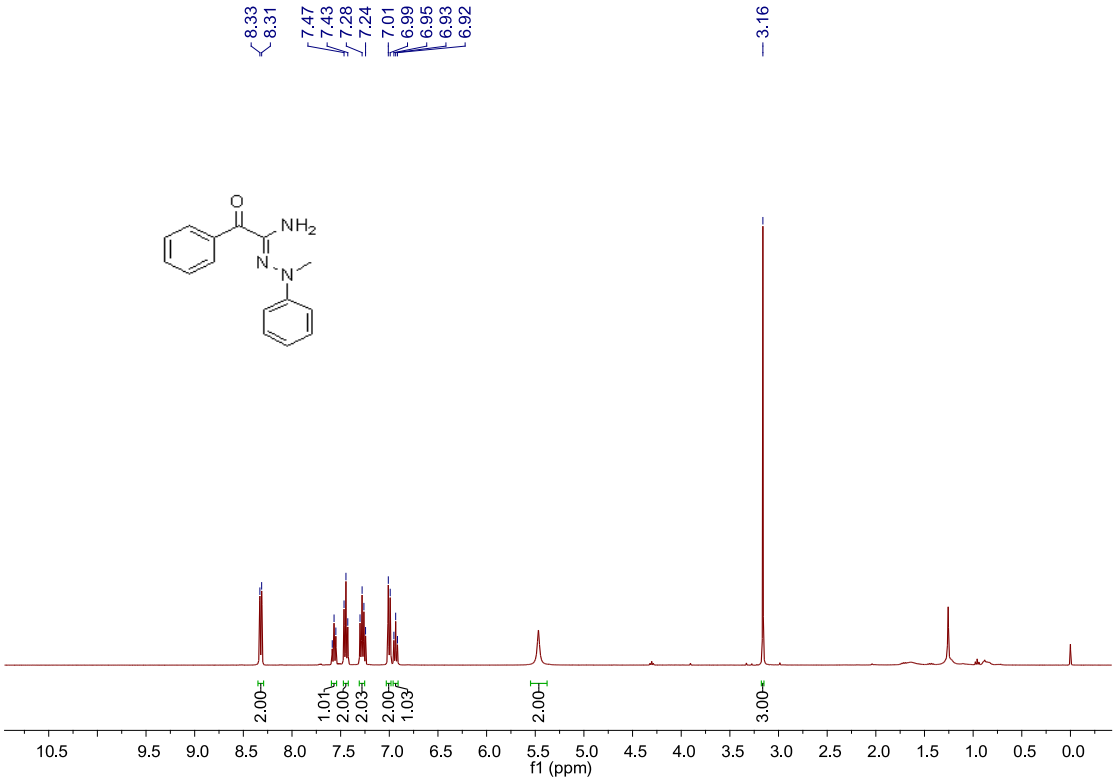
^{1}H NMR of product 2-60 in $CDCl_3$ (400 MHz)



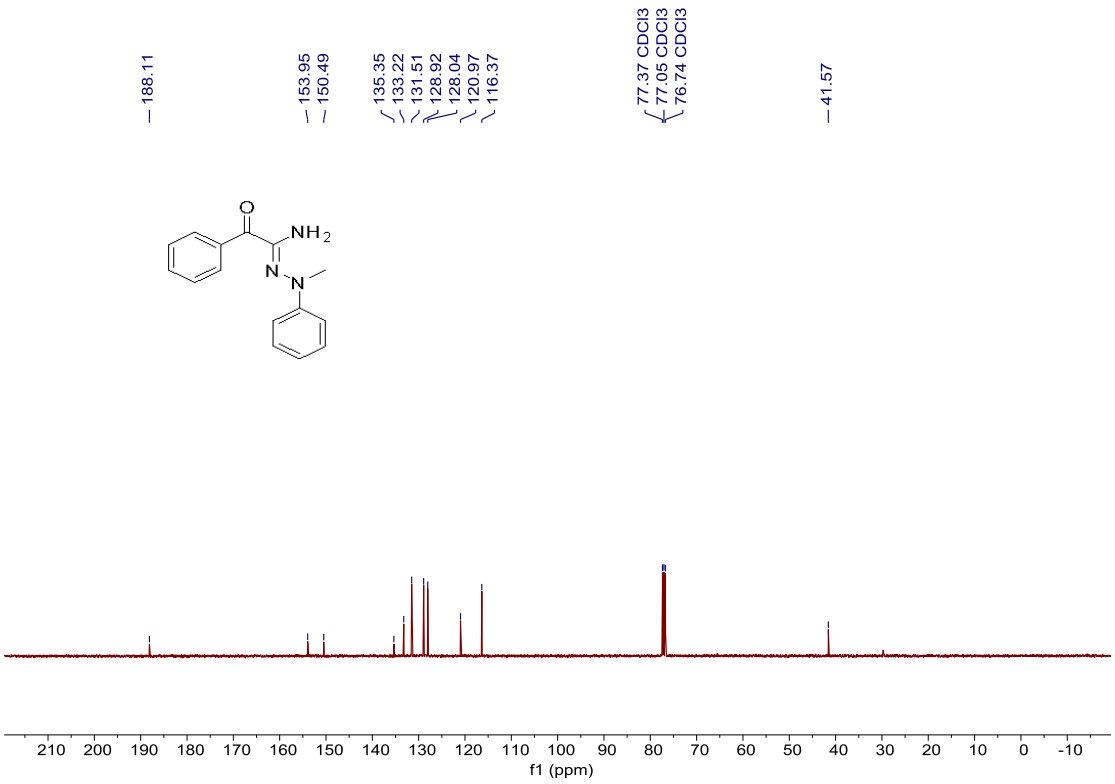
^{13}C NMR of product 2-60 in $CDCl_3$ (100 MHz)



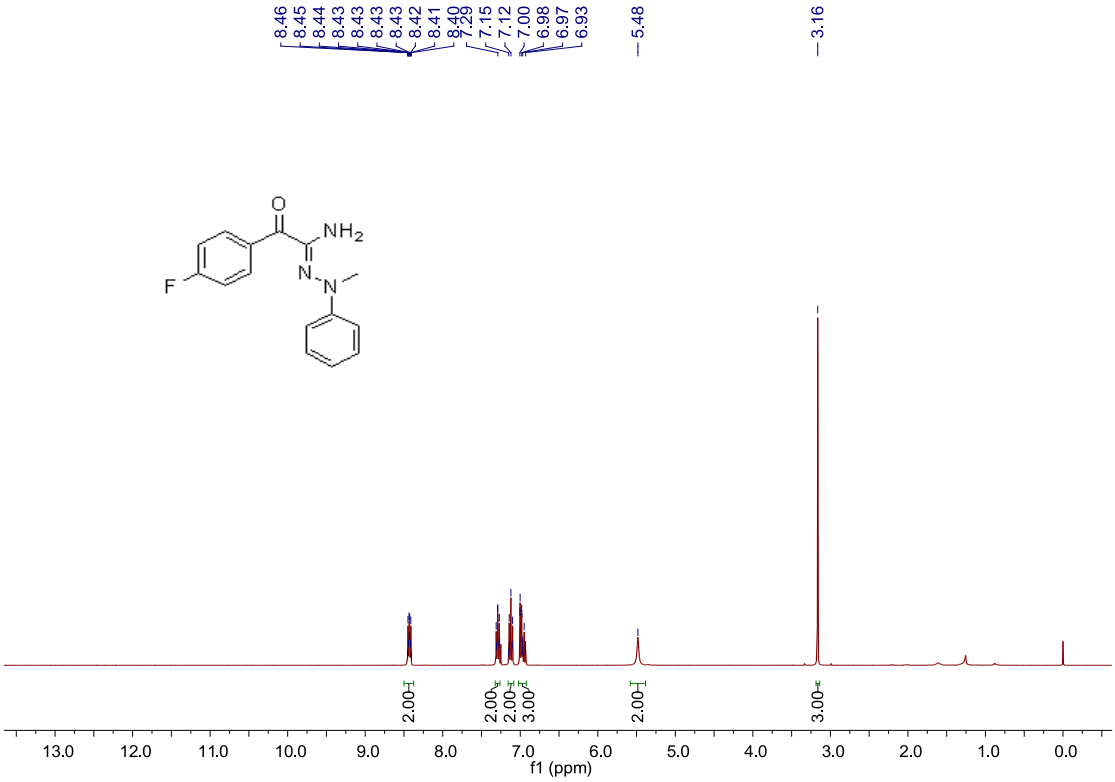
^{1}H NMR of product 3 in $CDCl_3$ (400 MHz)



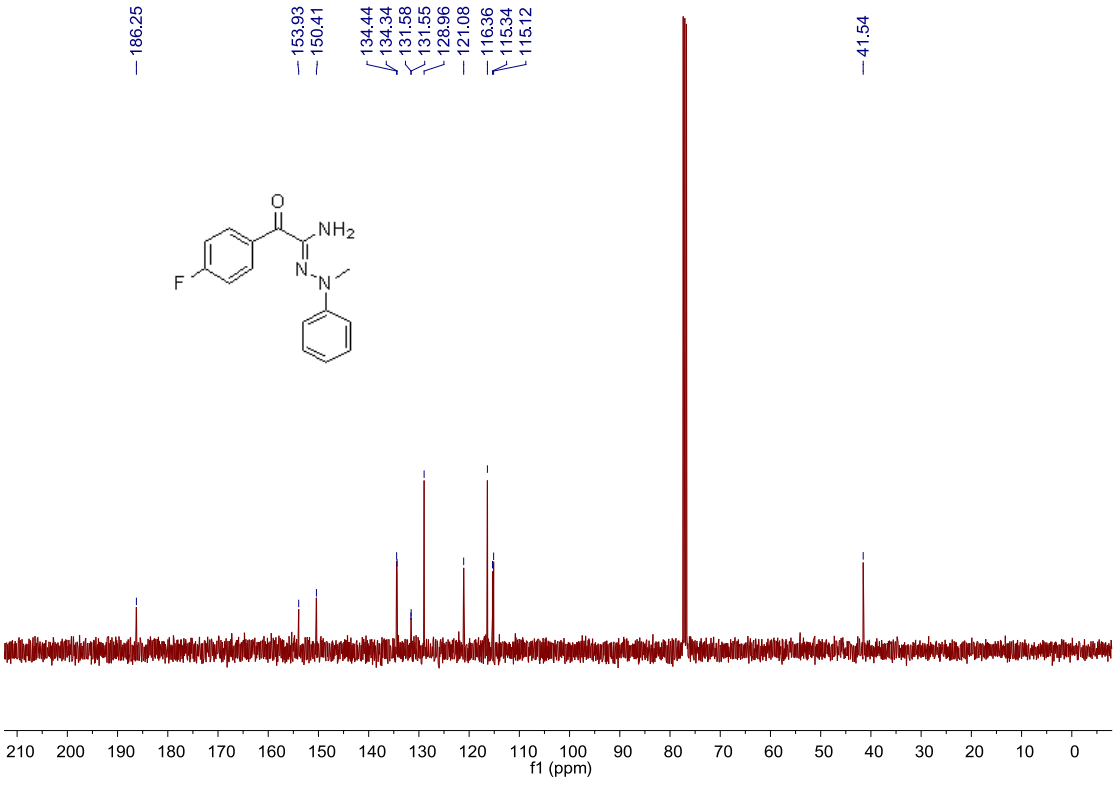
^{13}C NMR of product 3 in $CDCl_3$ (100 MHz)



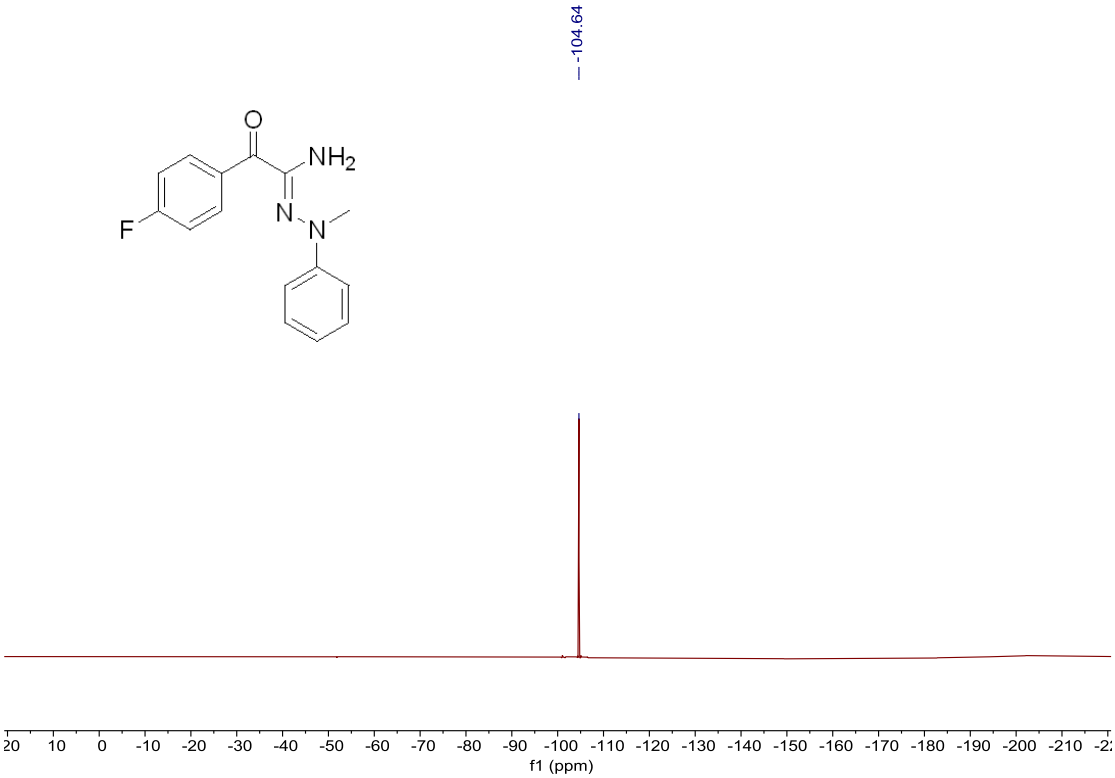
^{1}H NMR of product 4 in $CDCl_3$ (400 MHz)



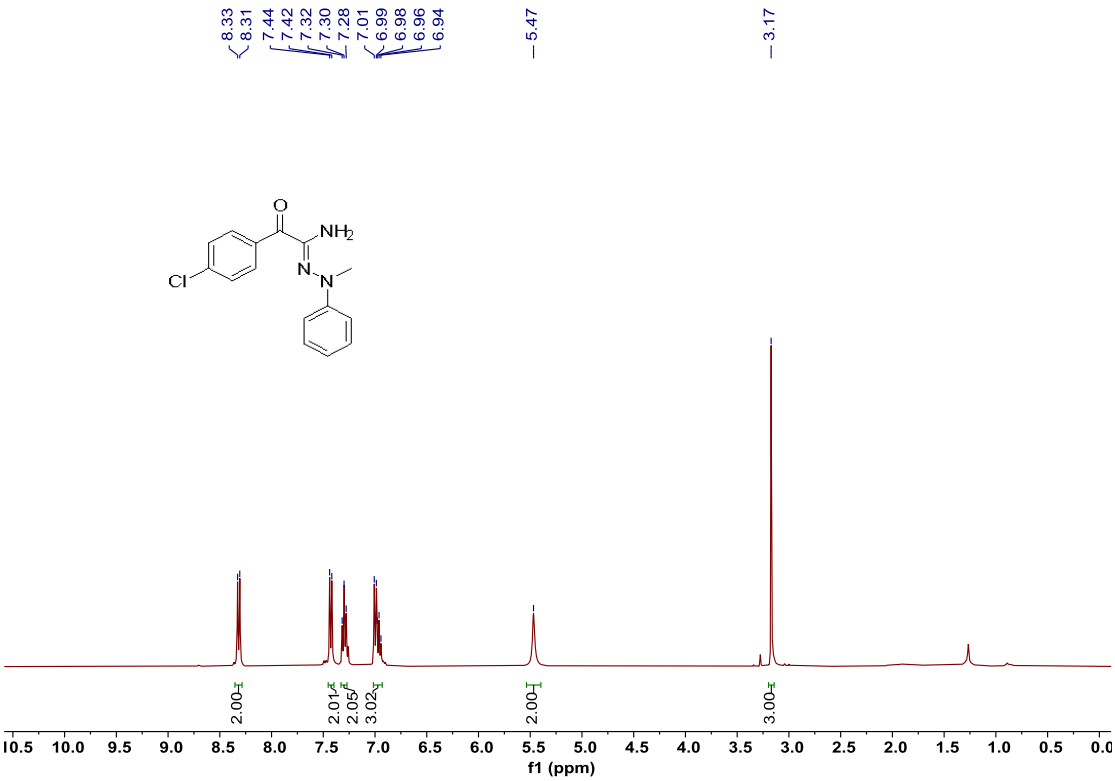
^{13}C NMR of product 4 in $CDCl_3$ (100 MHz)



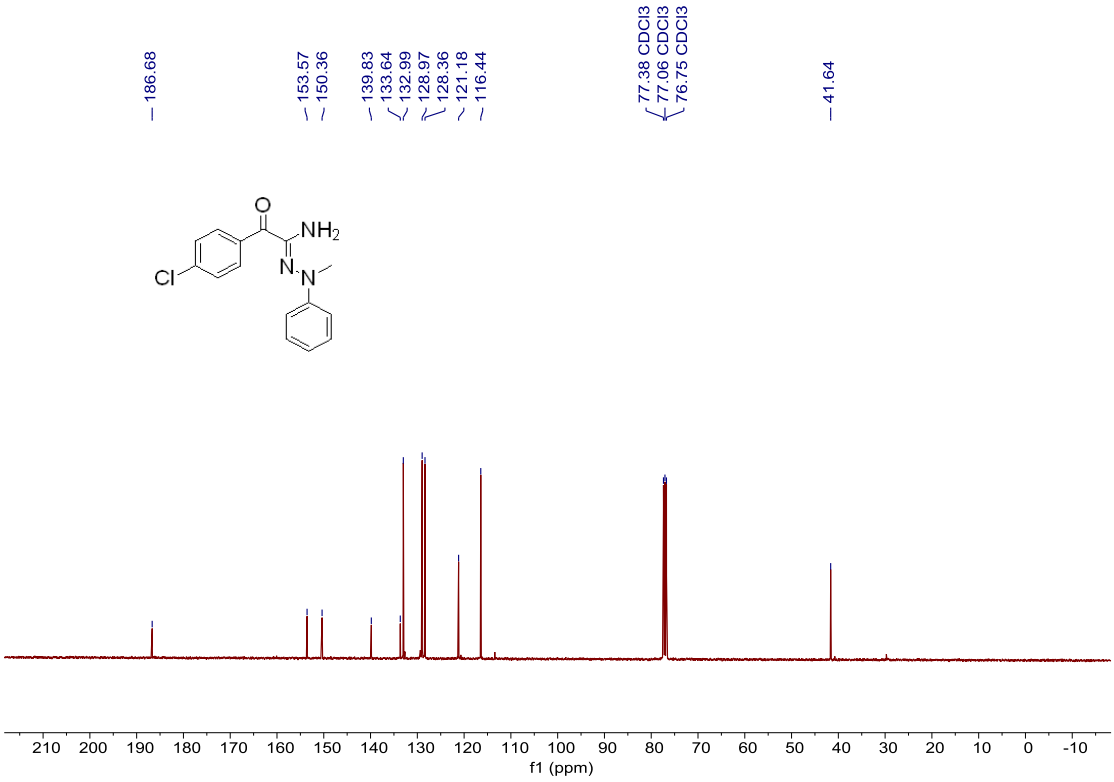
^{19}F NMR of product 4 in $CDCl_3$ (100 MHz)



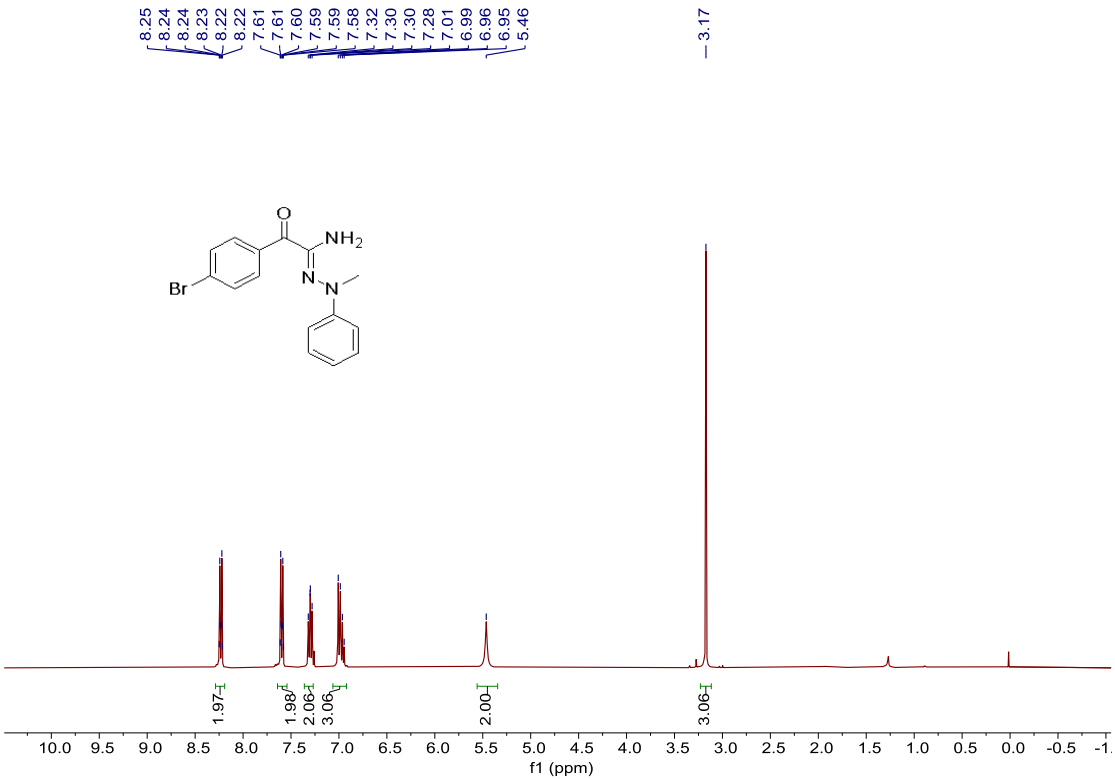
^{1}H NMR of product 5 in $CDCl_3$ (400 MHz)



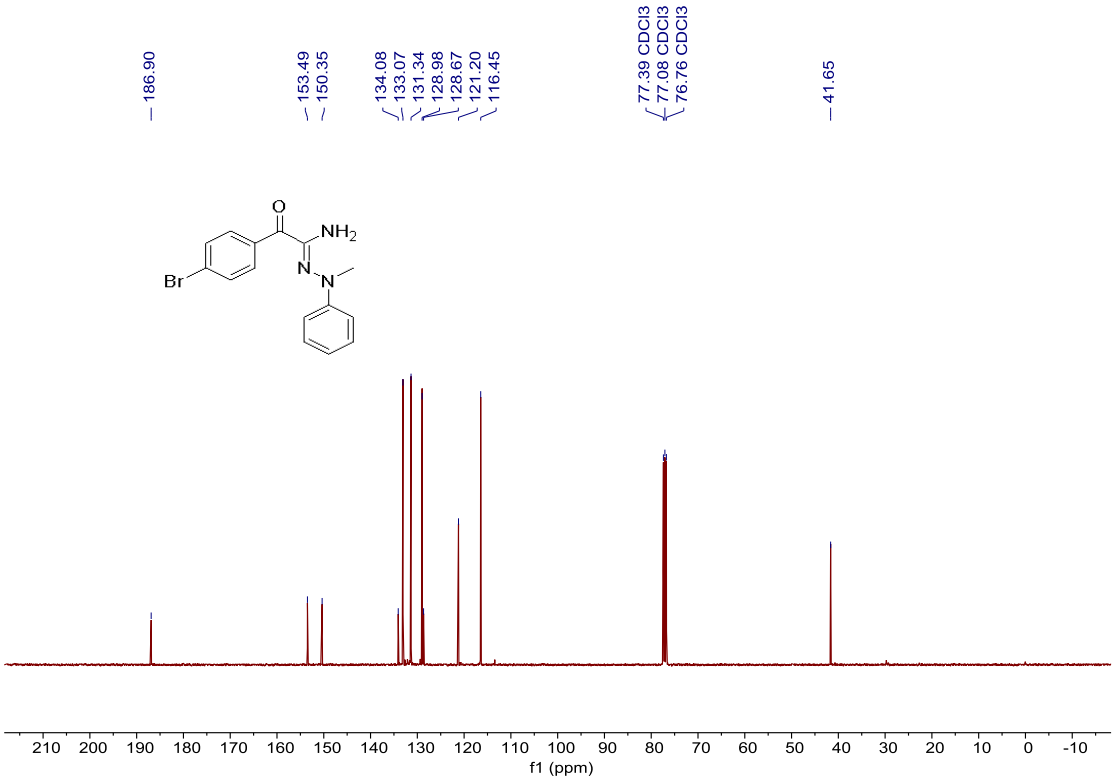
^{13}C NMR of product 5 in $CDCl_3$ (100 MHz)



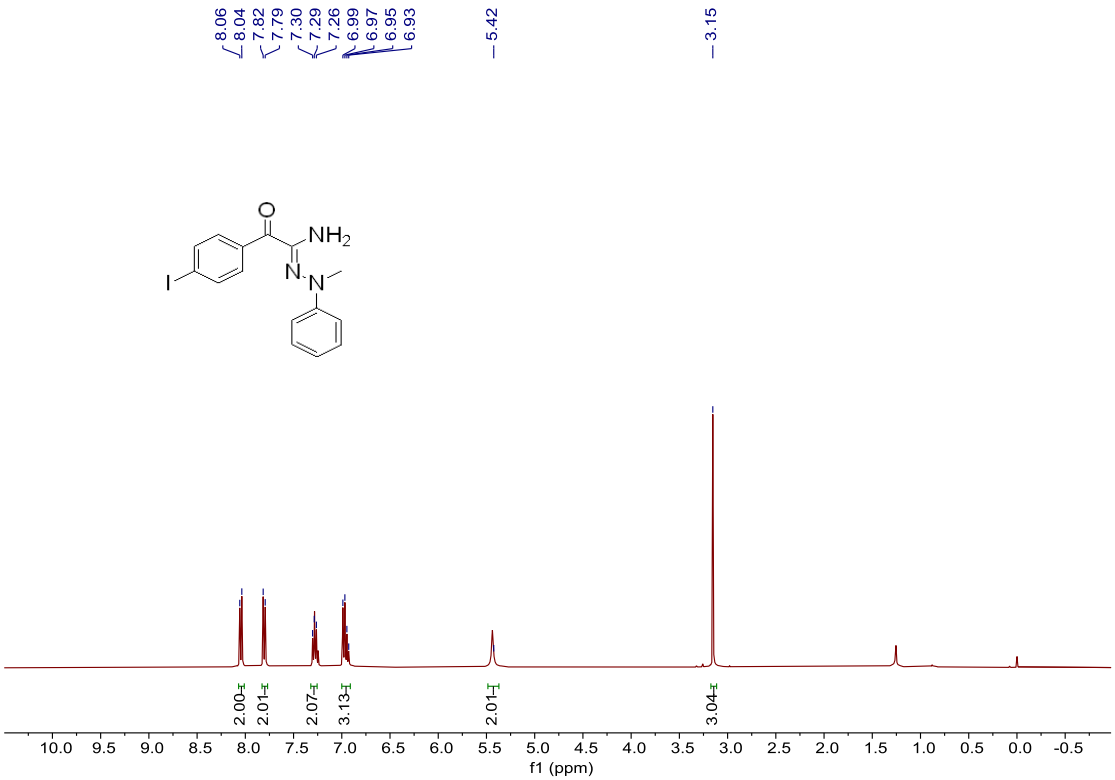
^{1}H NMR of product 6 in $CDCl_3$ (400 MHz)



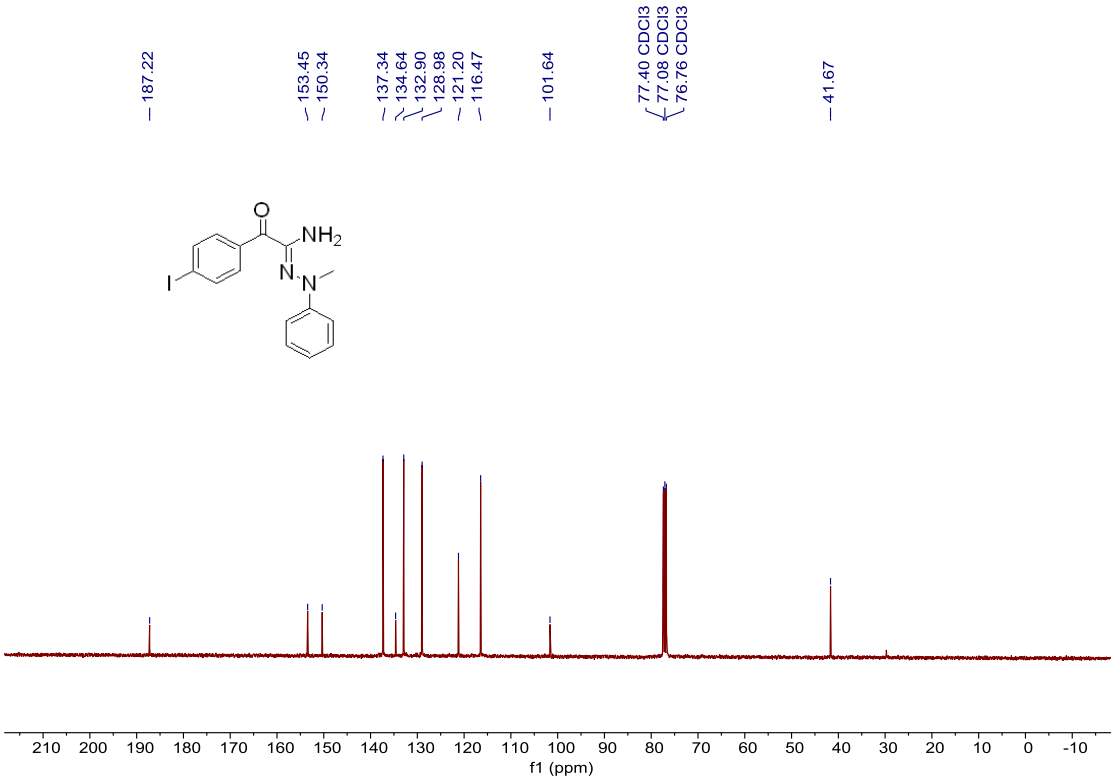
^{13}C NMR of product 6 in $CDCl_3$ (100 MHz)



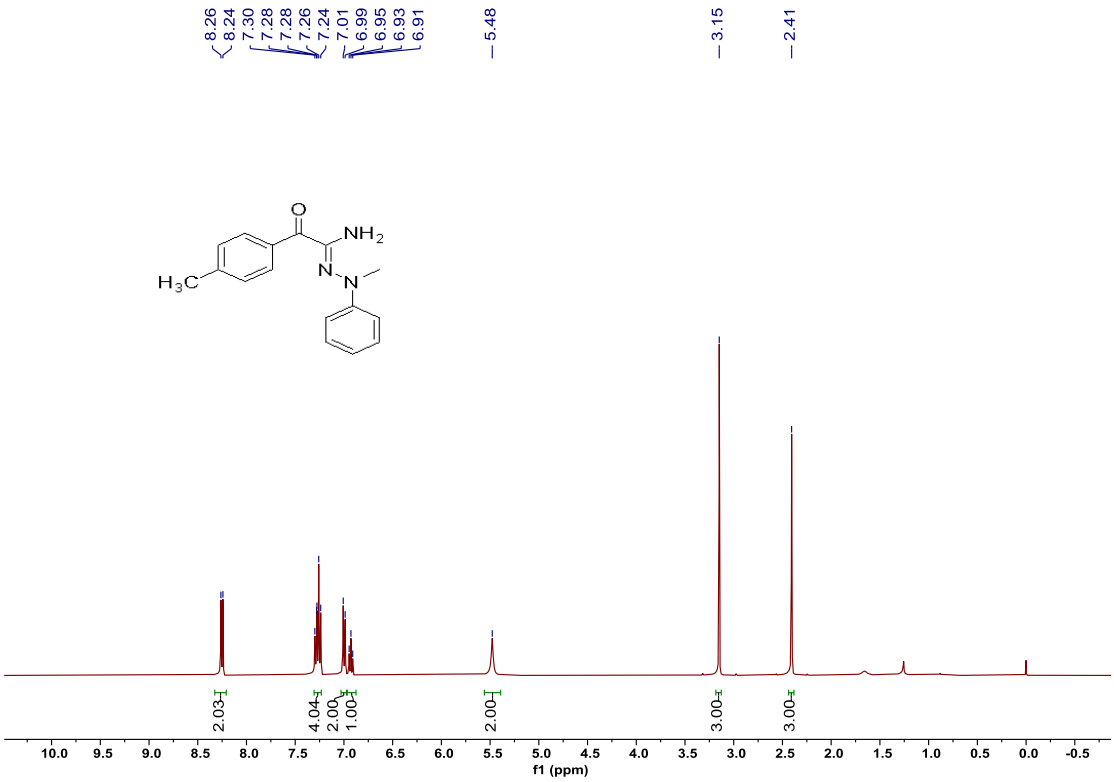
^{1}H NMR of product 7 in $CDCl_3$ (400 MHz)



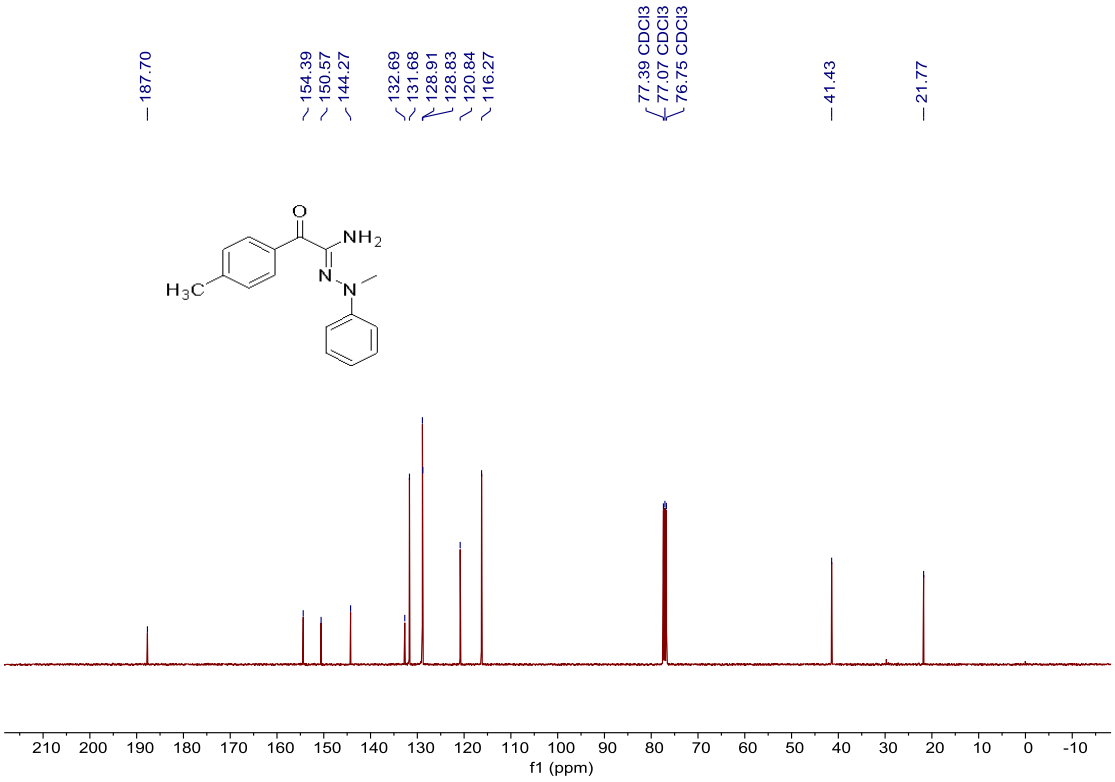
^{13}C NMR of product 7 in $CDCl_3$ (100 MHz)



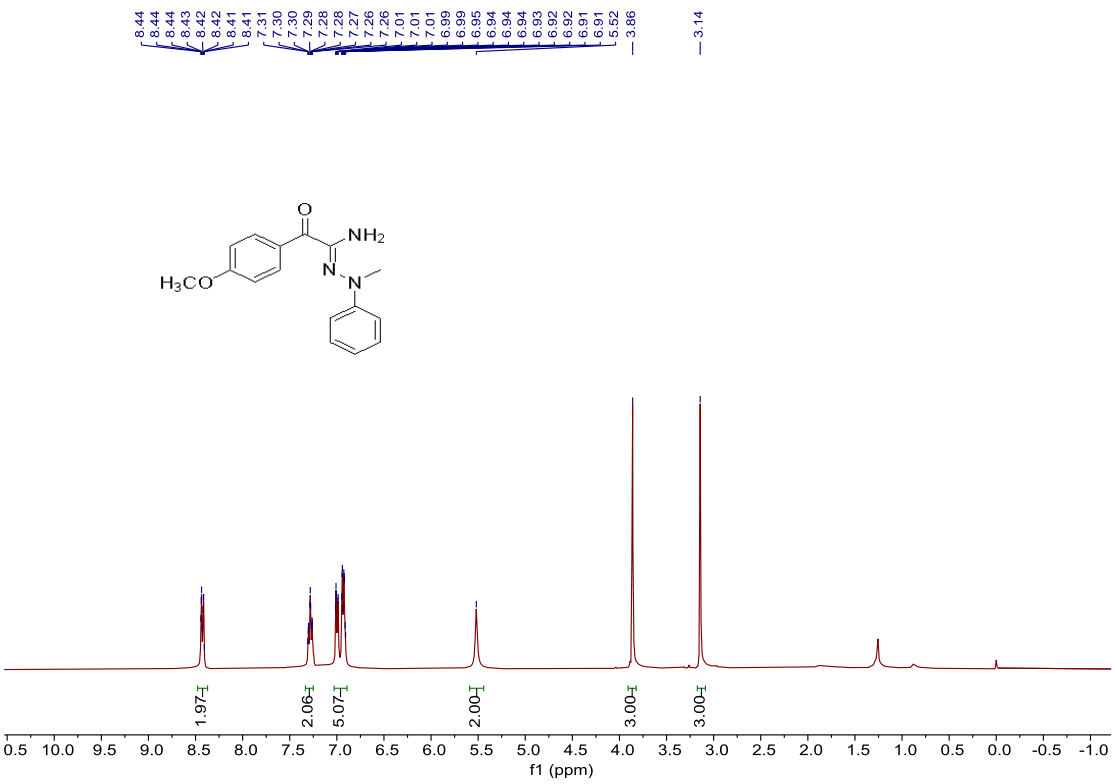
^{1}H NMR of product 8 in $CDCl_3$ (400 MHz)



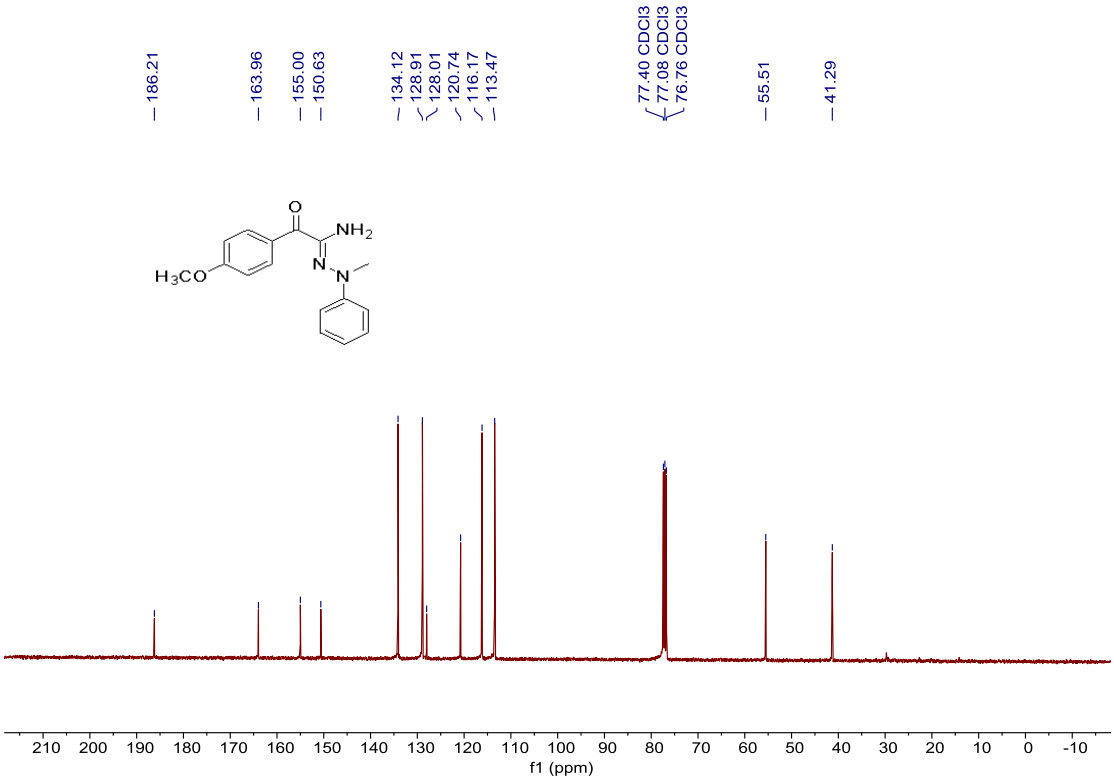
^{13}C NMR of product 8 in $CDCl_3$ (100 MHz)



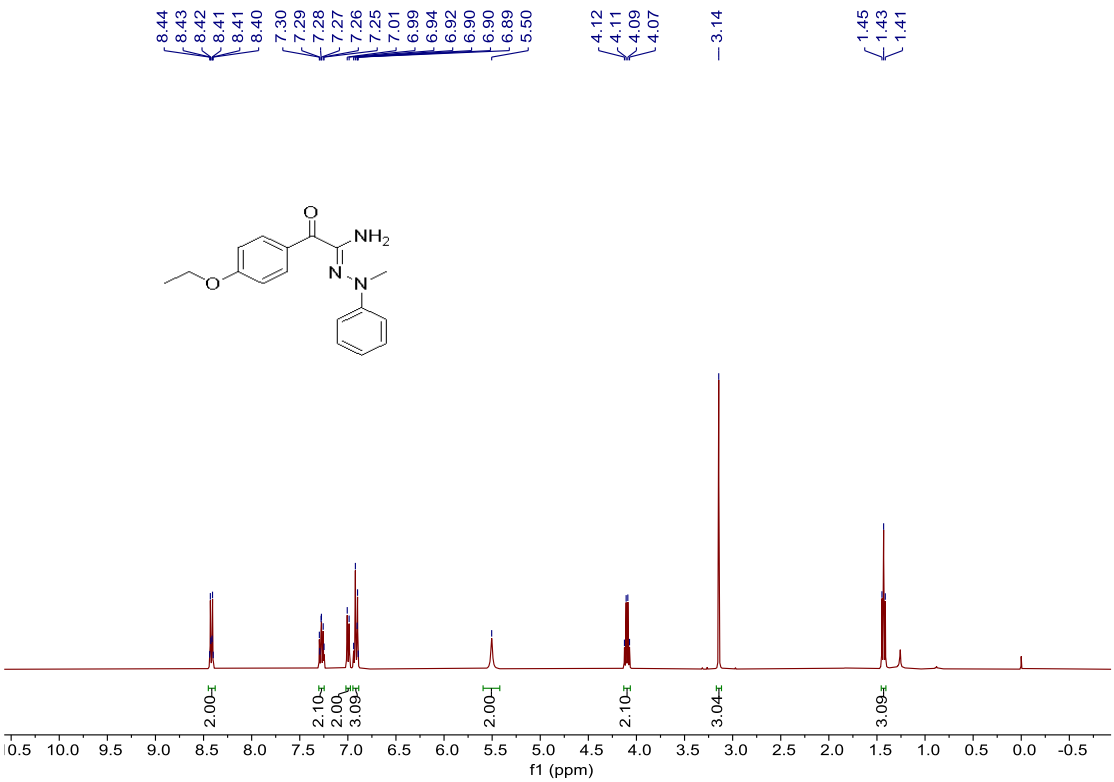
^{1}H NMR of product 9 in $CDCl_3$ (400 MHz)



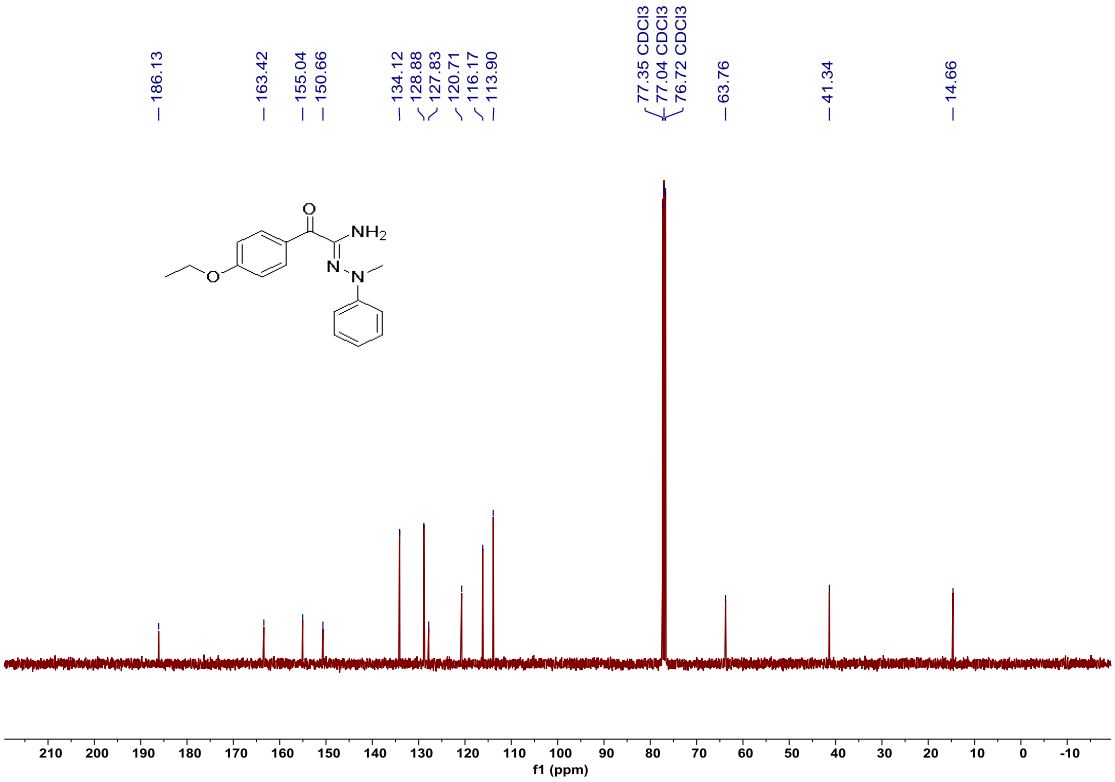
^{13}C NMR of product 9 in $CDCl_3$ (100 MHz)



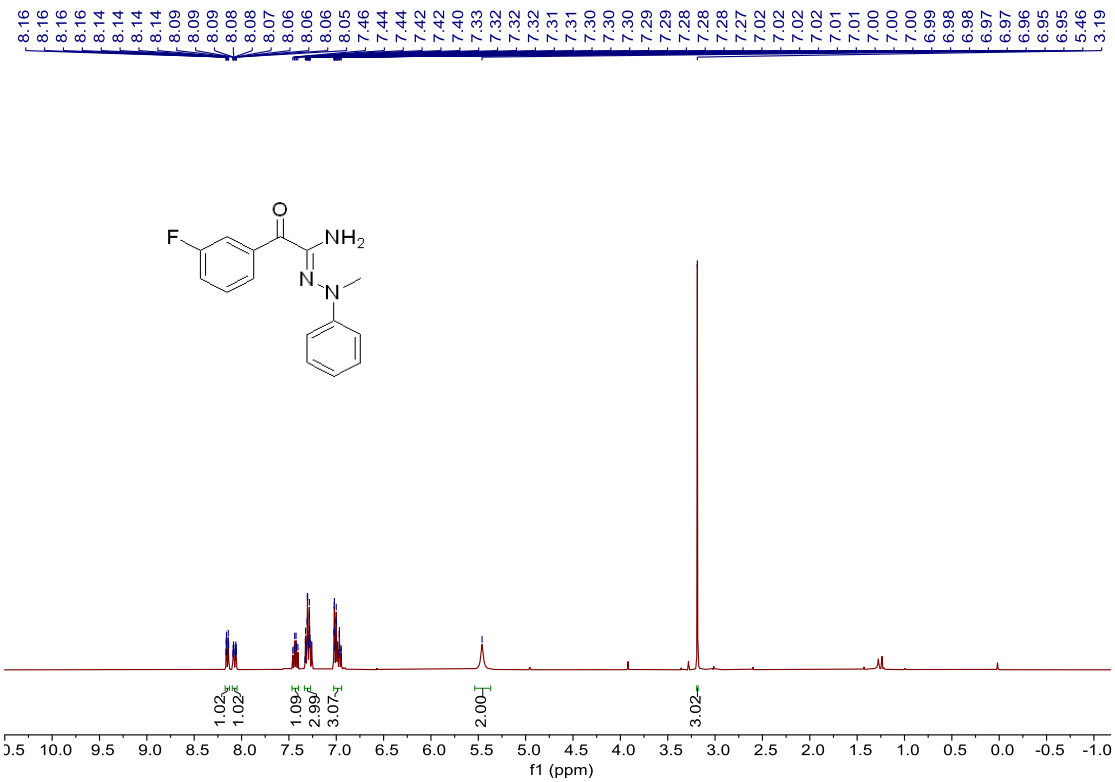
^{1}H NMR of product 10 in $CDCl_3$ (400 MHz)



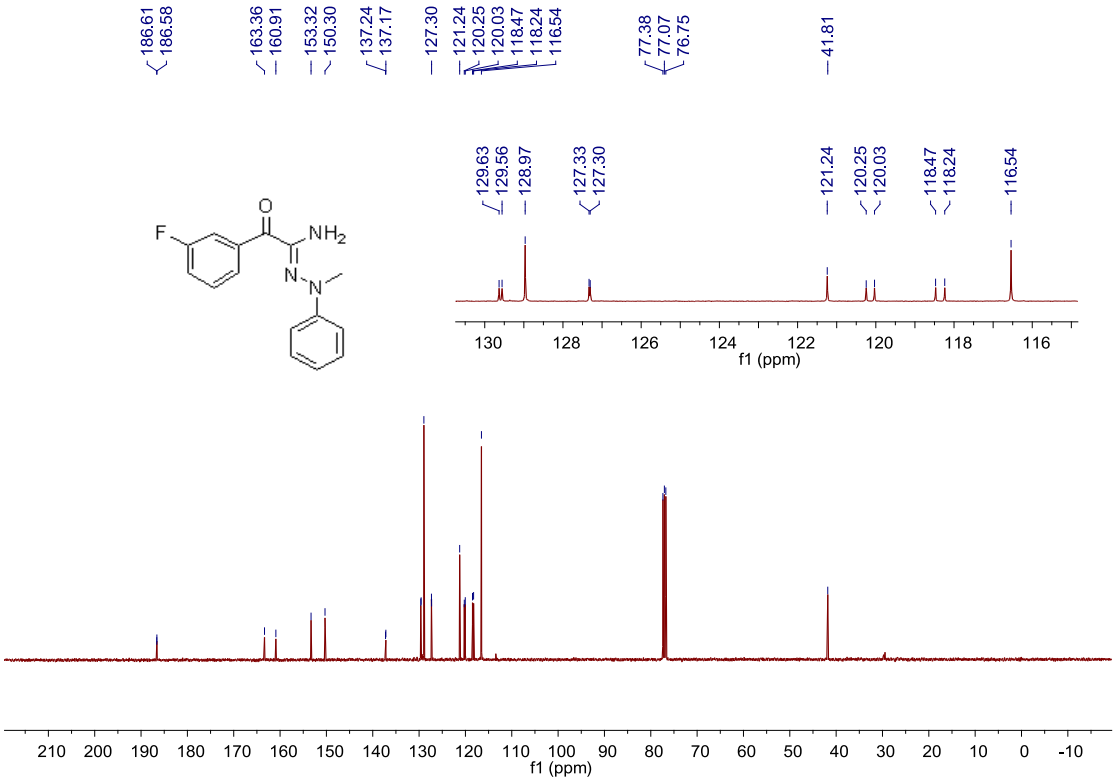
^{13}C NMR of product 10 in $CDCl_3$ (100 MHz)



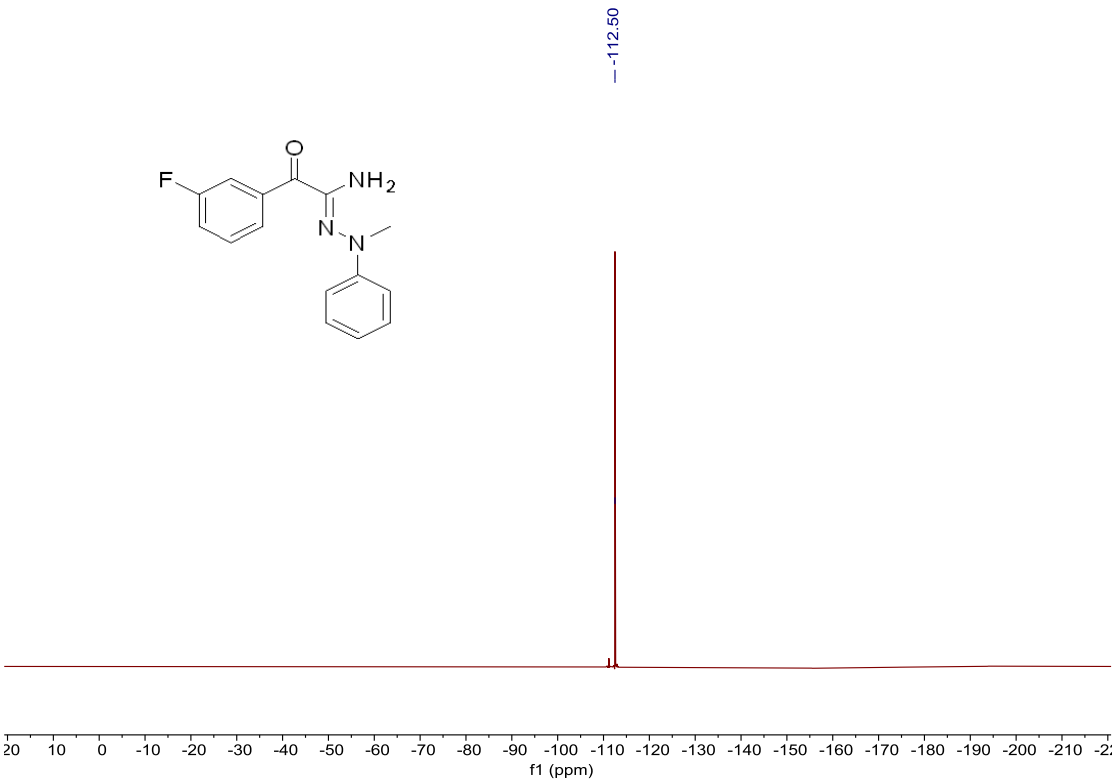
^{1}H NMR of product 11 in $CDCl_3$ (400 MHz)



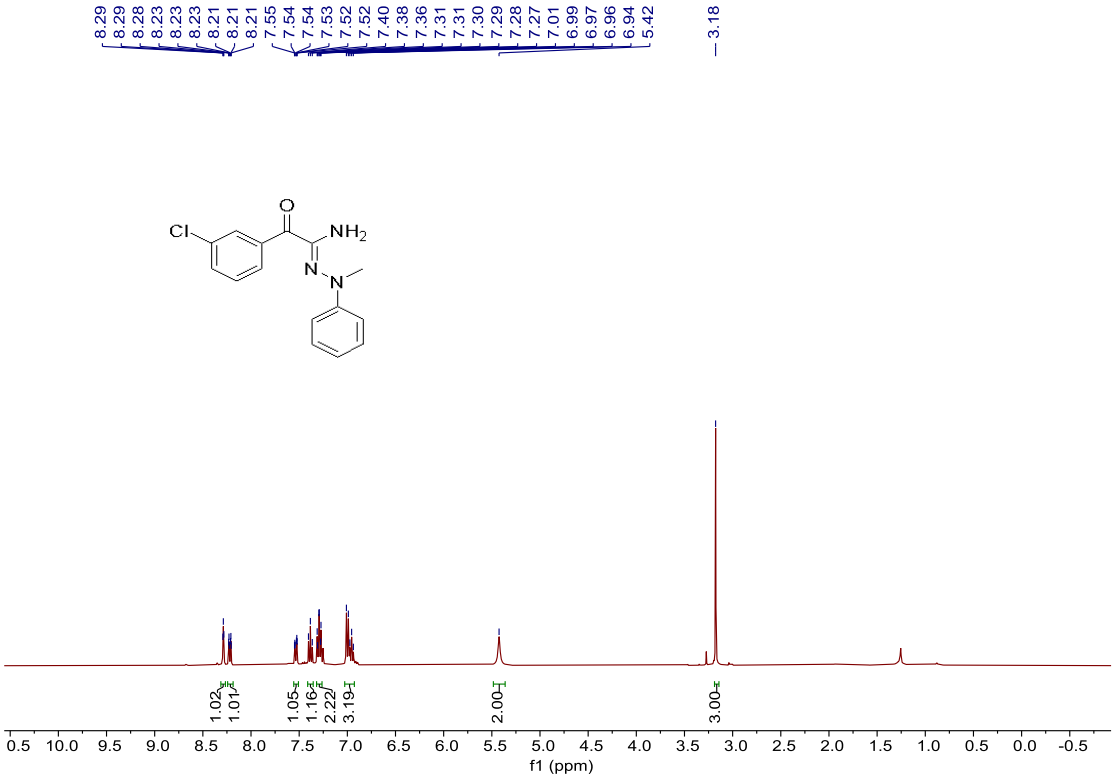
^{13}C NMR of product 11 in $CDCl_3$ (100 MHz)



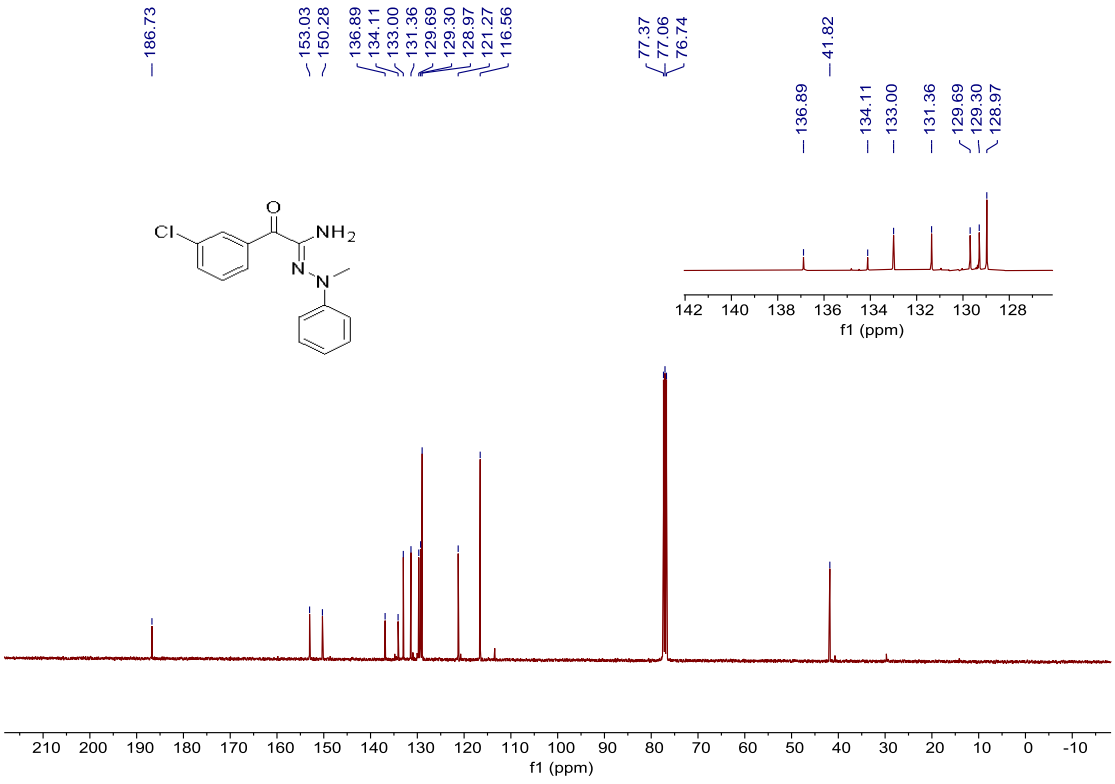
^{19}F NMR of product 11 in $CDCl_3$ (100 MHz)



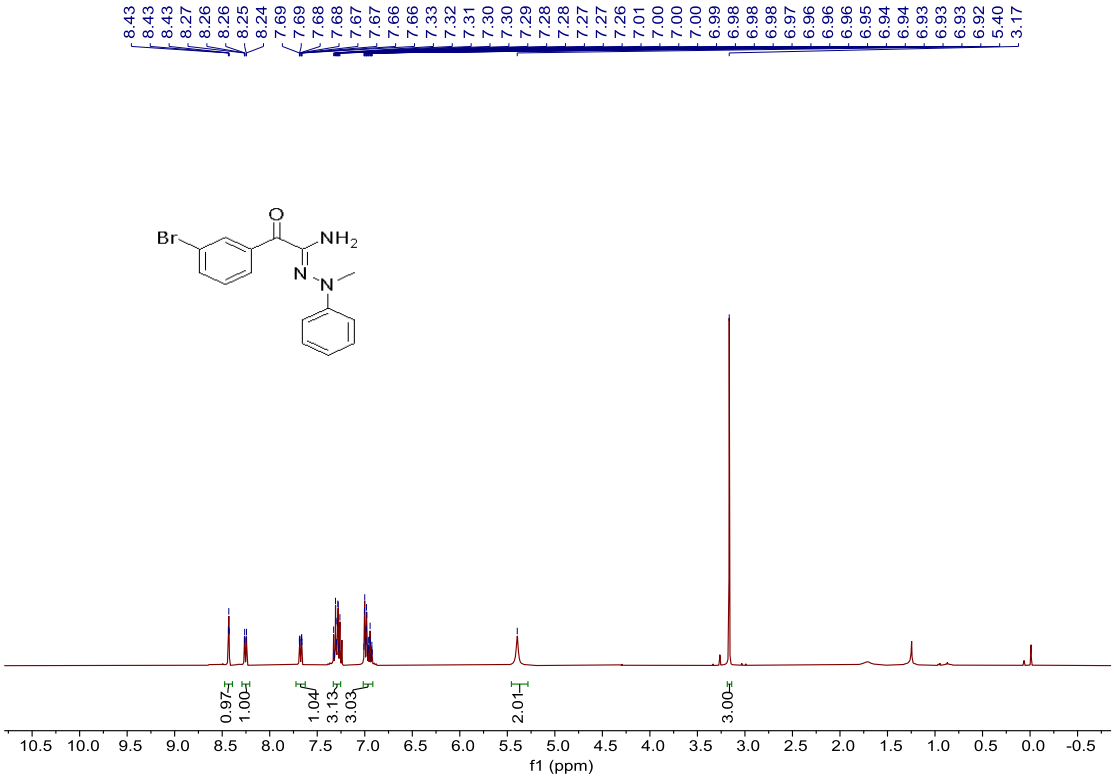
^{1}H NMR of product 12 in $CDCl_3$ (400 MHz)



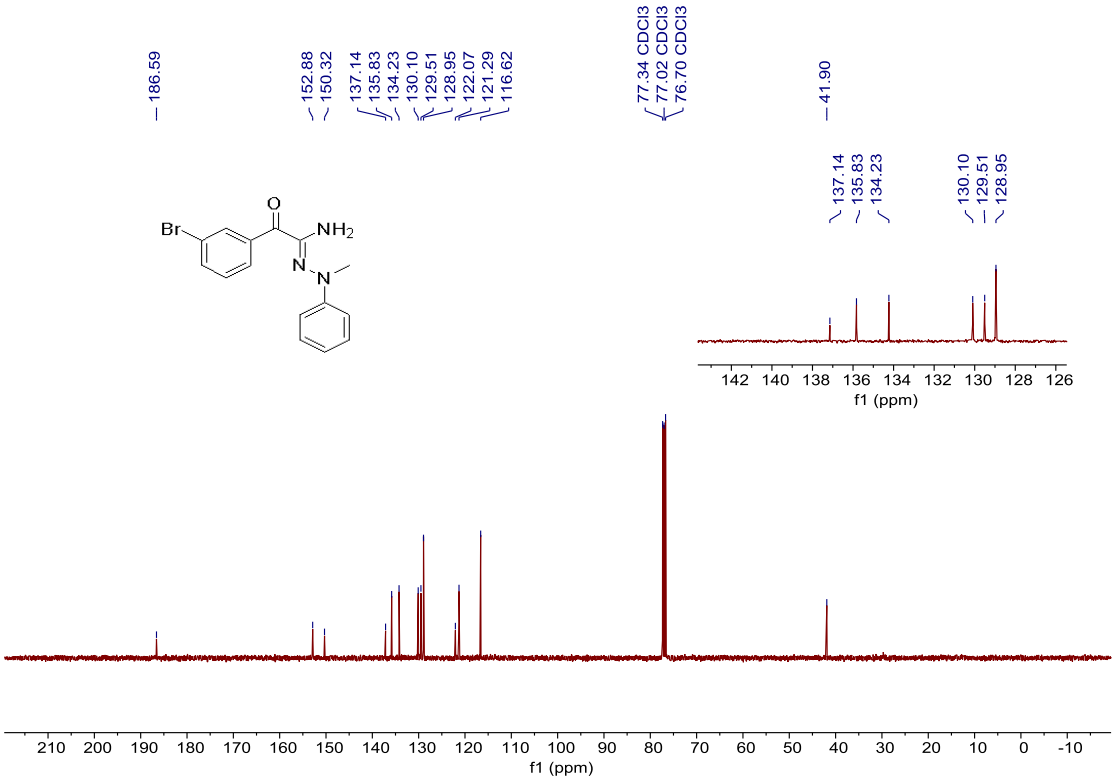
^{13}C NMR of product 12 in $CDCl_3$ (100 MHz)



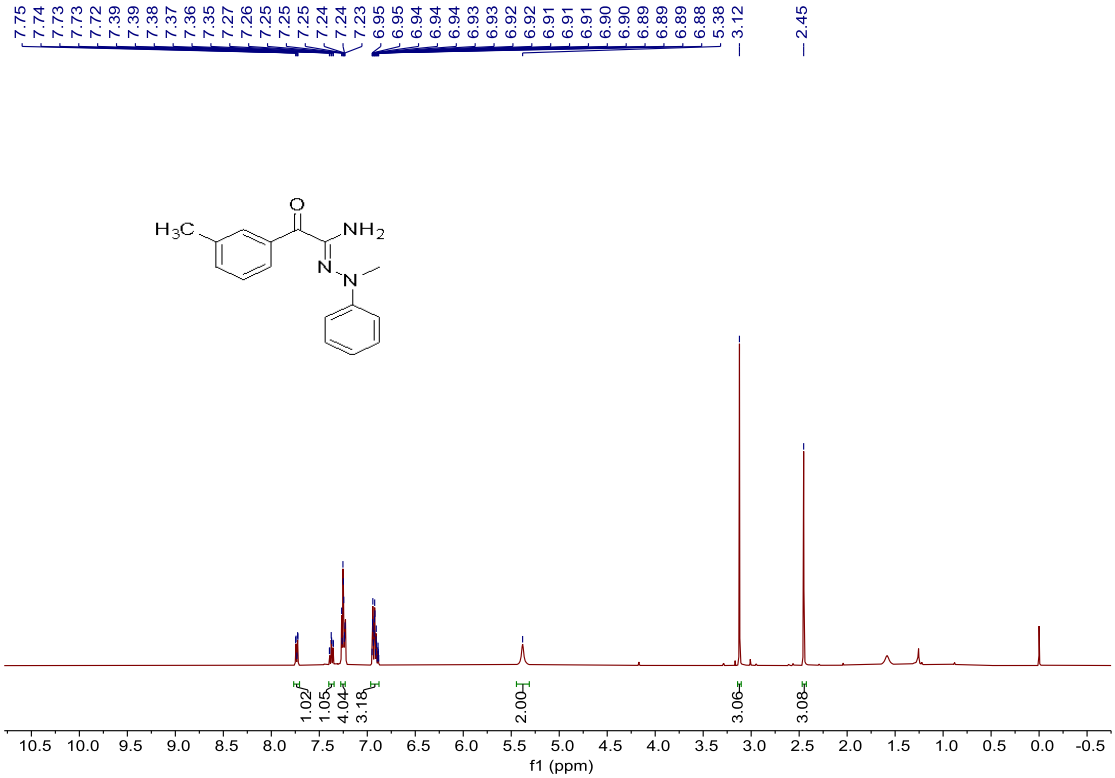
^{1}H NMR of product 13 in $CDCl_3$ (400 MHz)



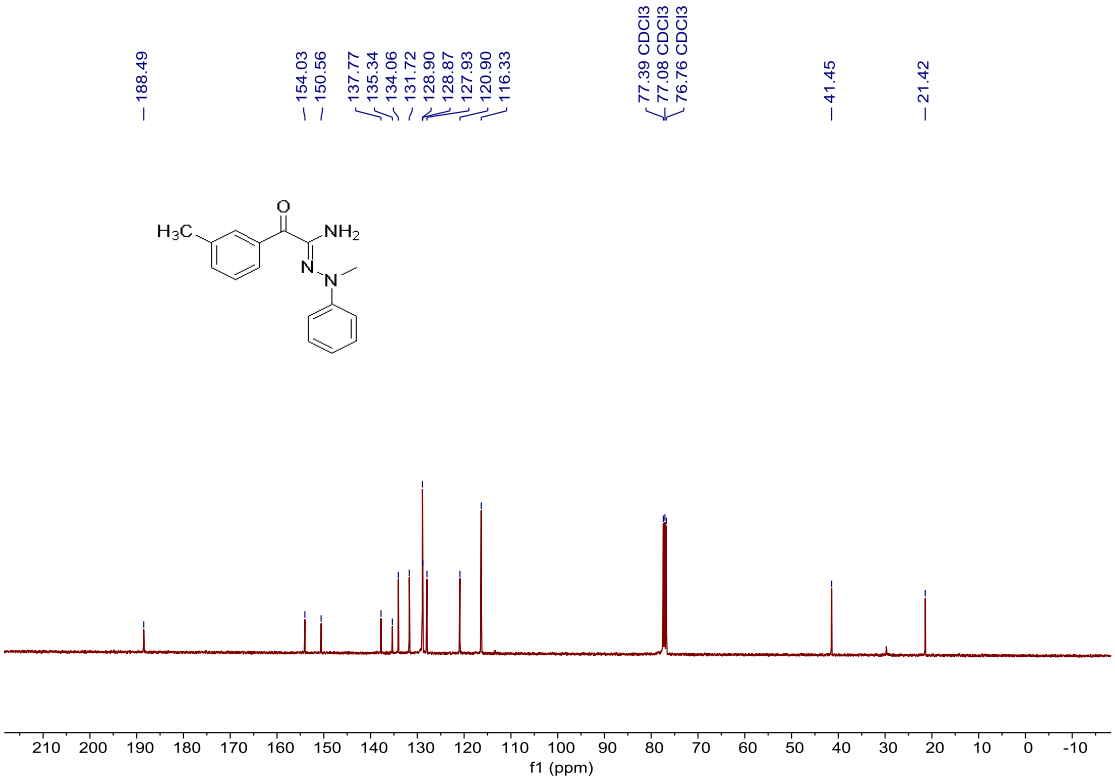
^{13}C NMR of product 13 in $CDCl_3$ (100 MHz)



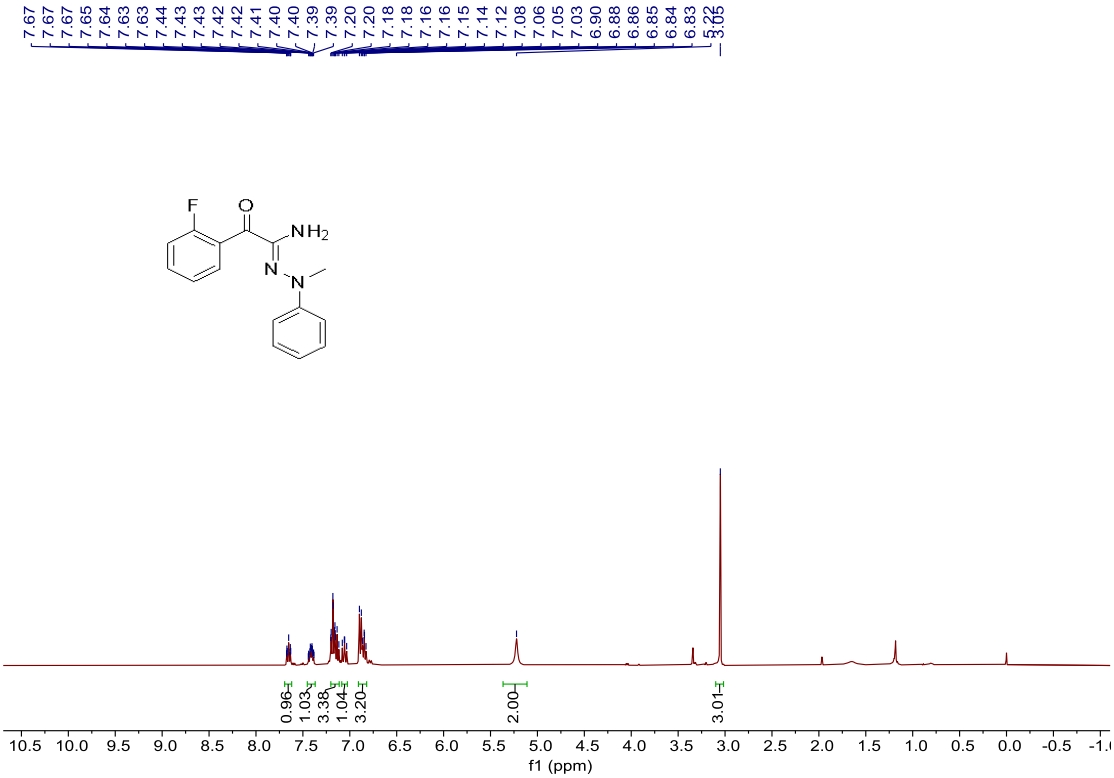
^{1}H NMR of product 14 in $CDCl_3$ (400 MHz)



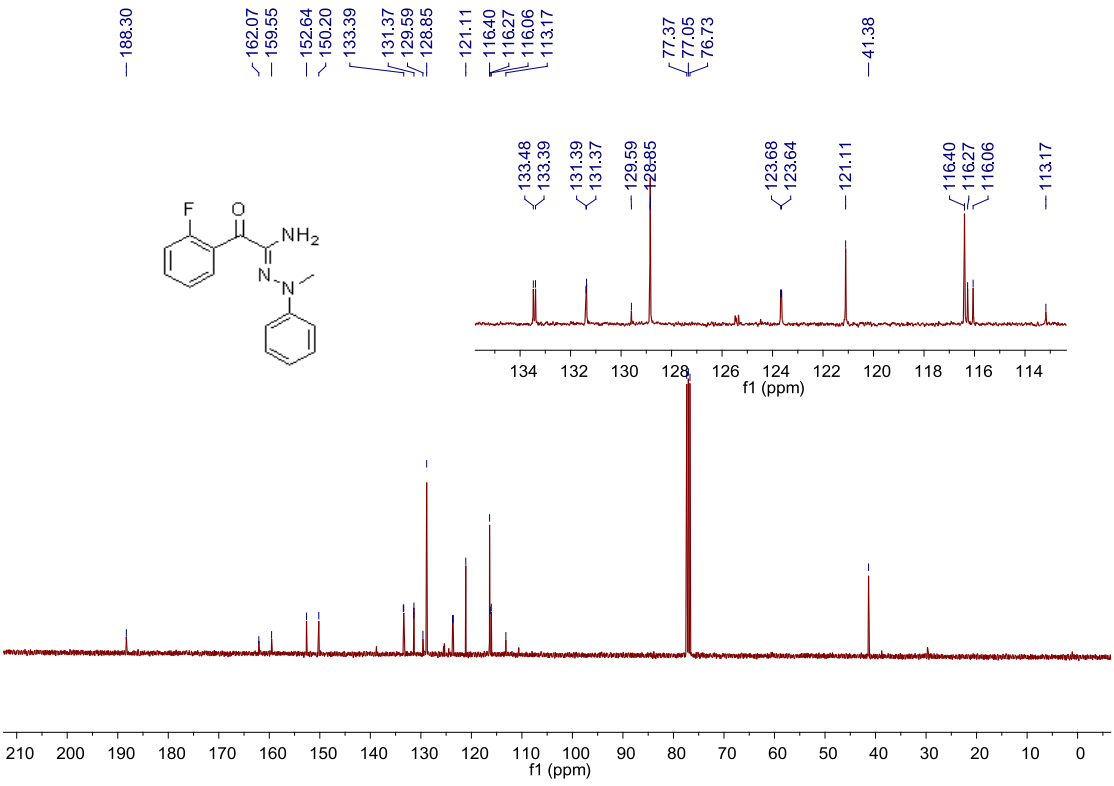
^{13}C NMR of product 14 in $CDCl_3$ (100 MHz)



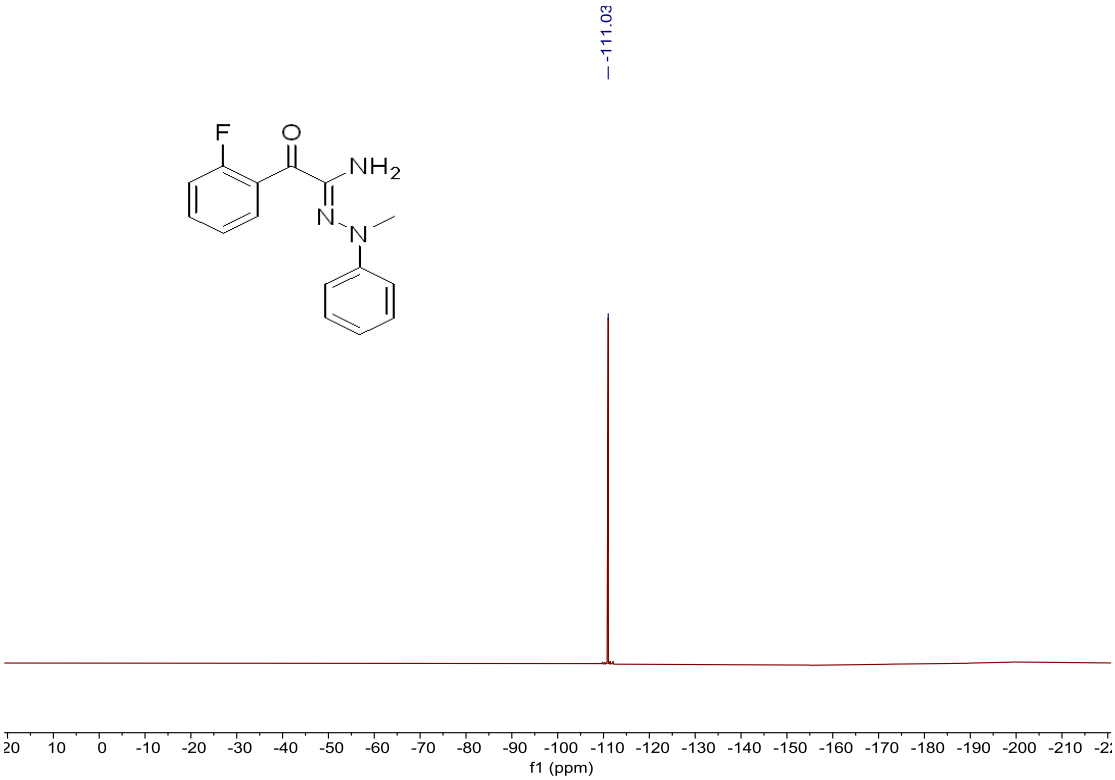
^{1}H NMR of product 15 in $CDCl_3$ (400 MHz)



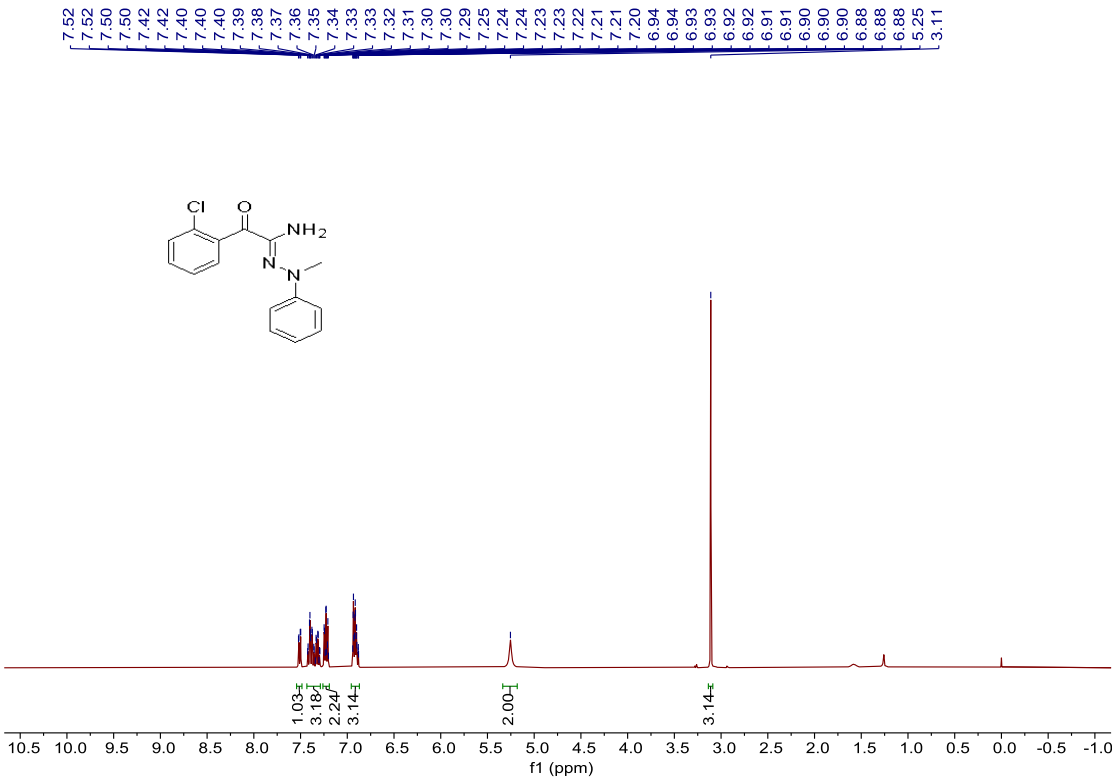
^{13}C NMR of product 15 in $CDCl_3$ (100 MHz)



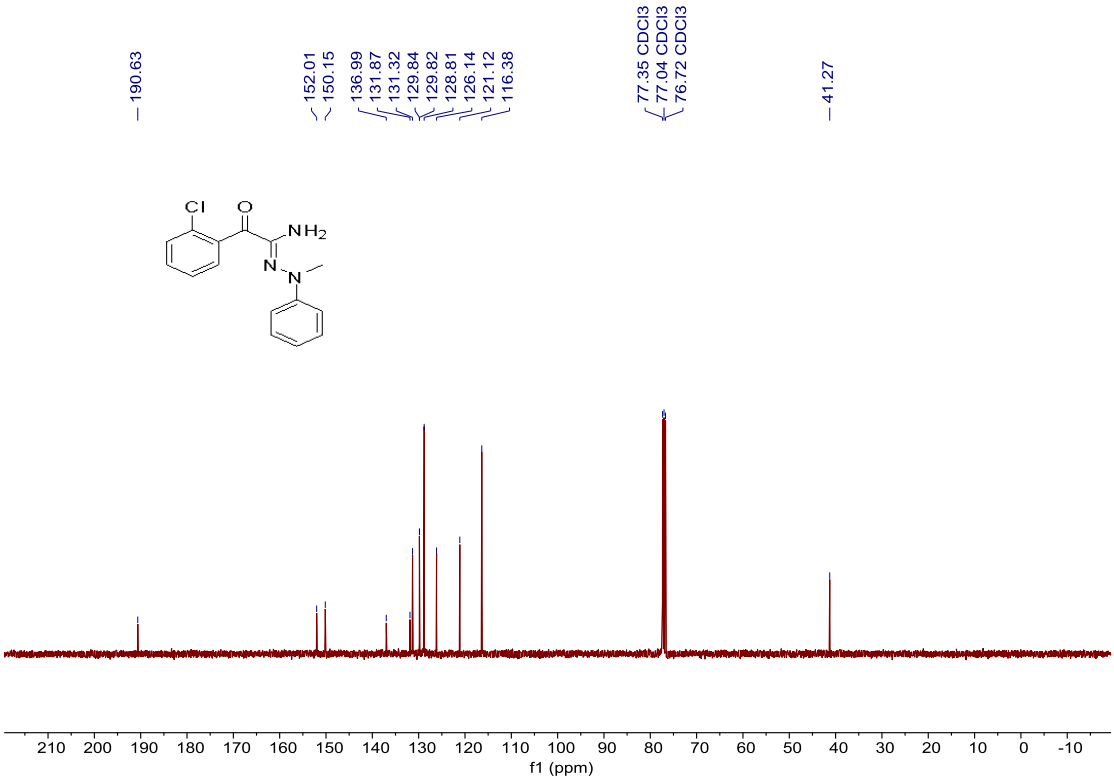
^{19}F NMR of product 15 in $CDCl_3$ (100 MHz)



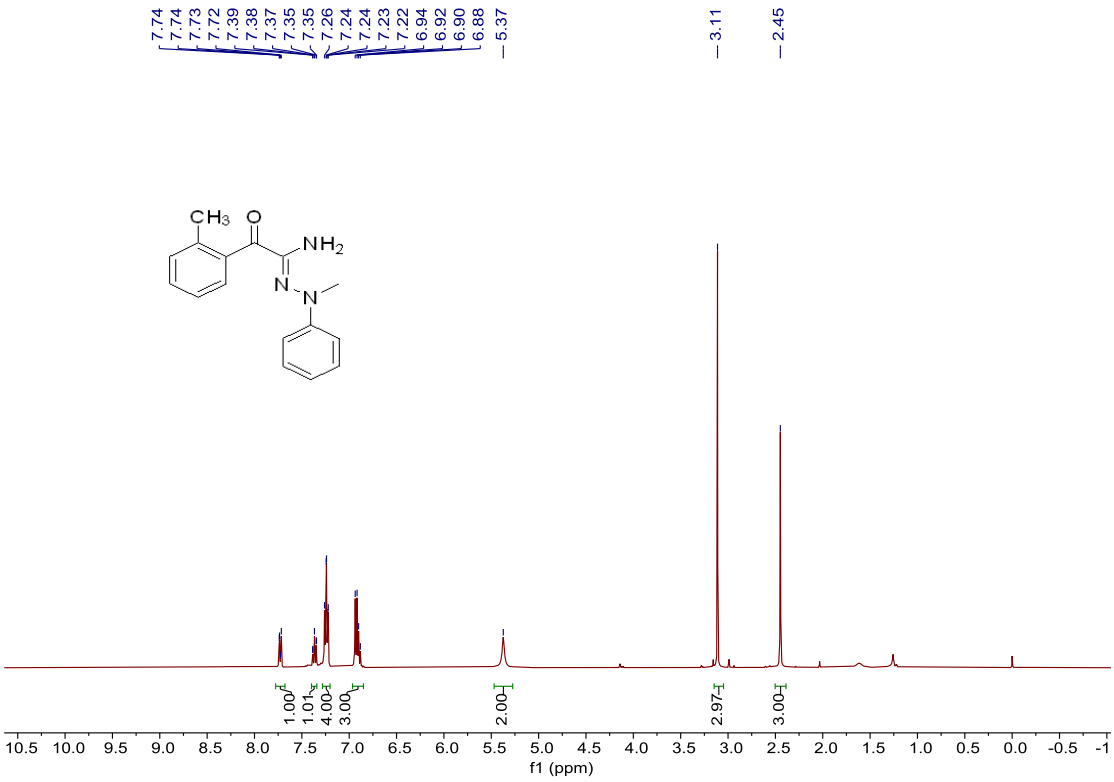
^{1}H NMR of product 16 in $CDCl_3$ (400 MHz)



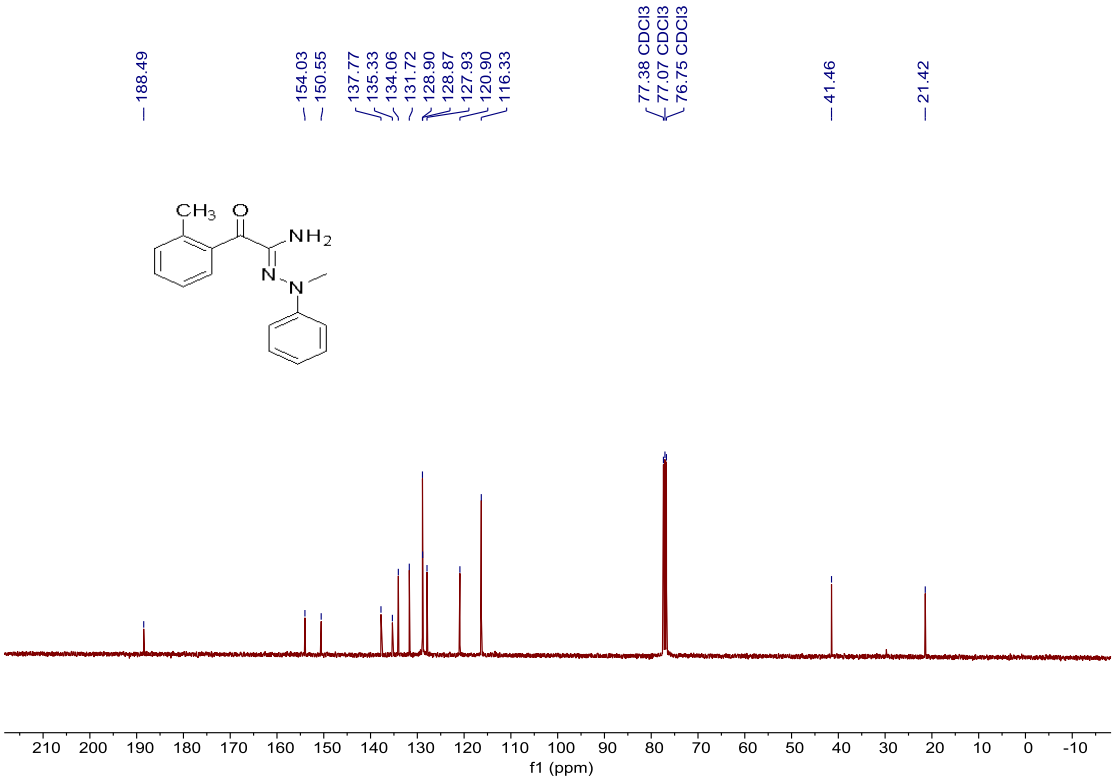
^{13}C NMR of product 16 in $CDCl_3$ (100 MHz)



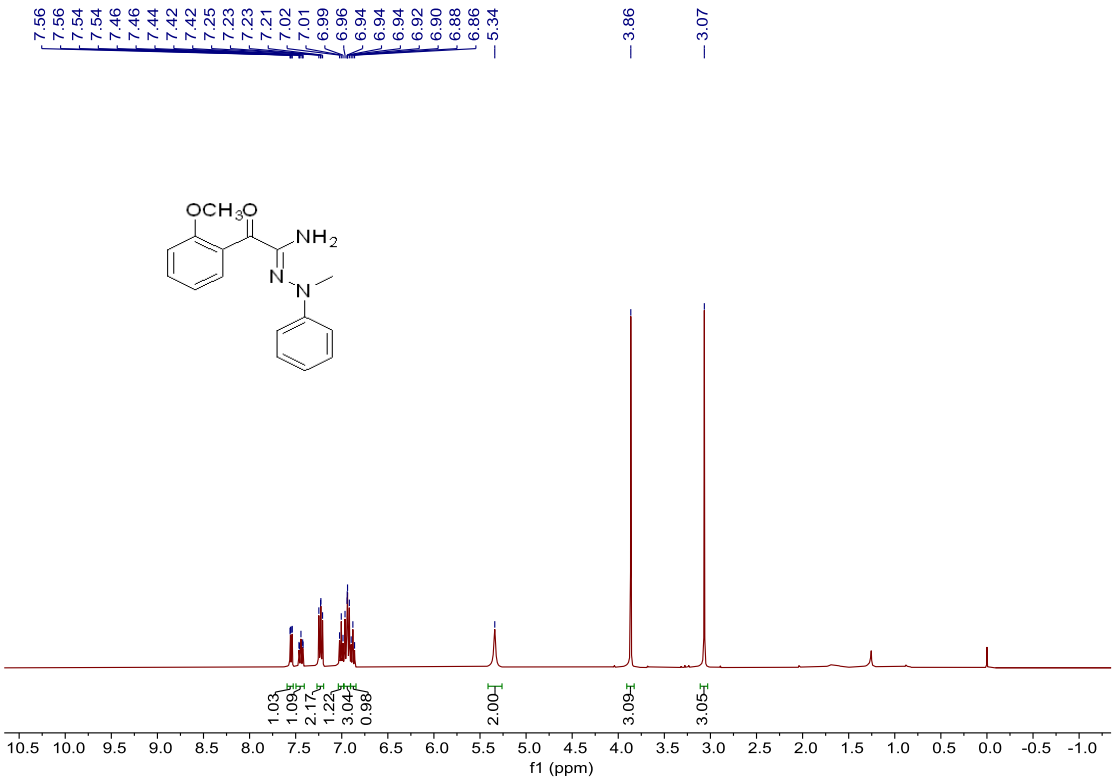
^{1}H NMR of product 17 in $CDCl_3$ (400 MHz)



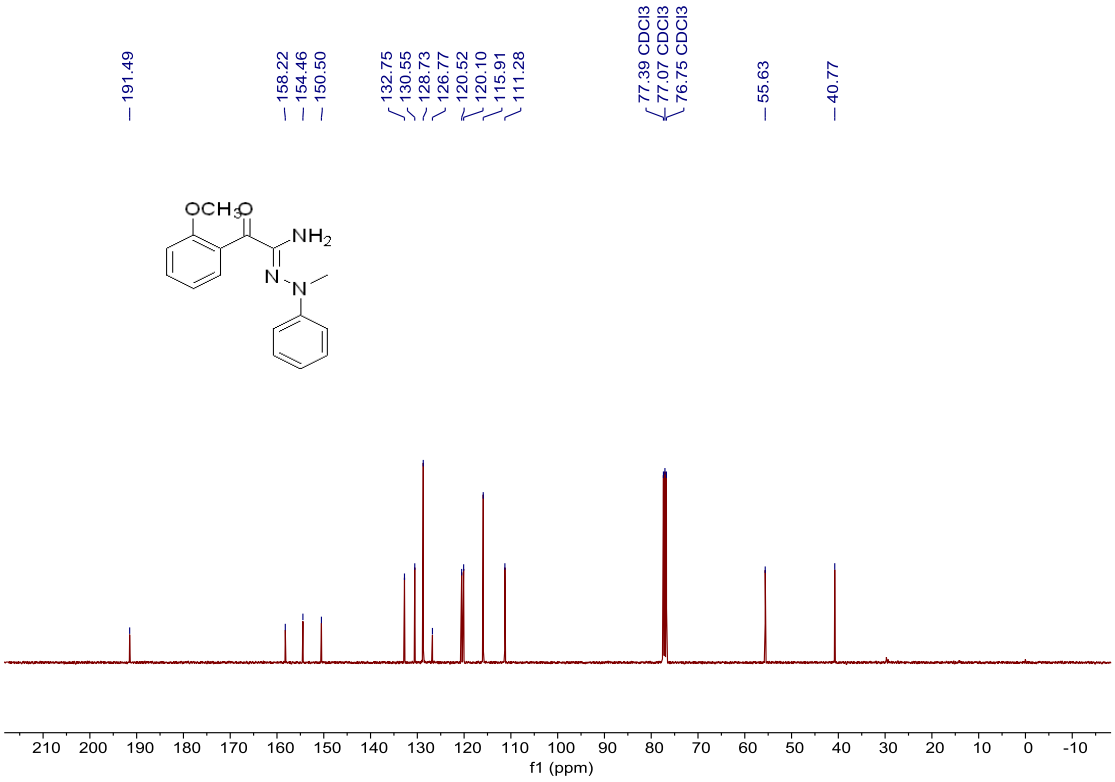
^{13}C NMR of product 17 in $CDCl_3$ (100 MHz)



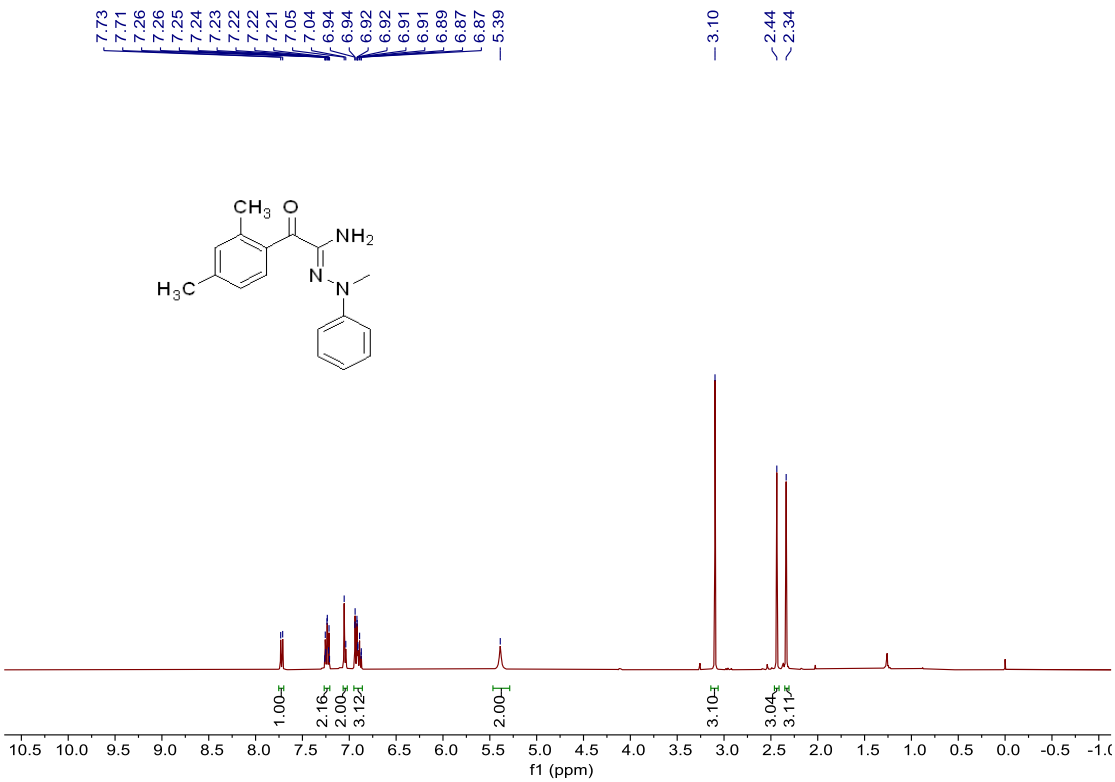
^{1}H NMR of product 18 in $CDCl_3$ (400 MHz)



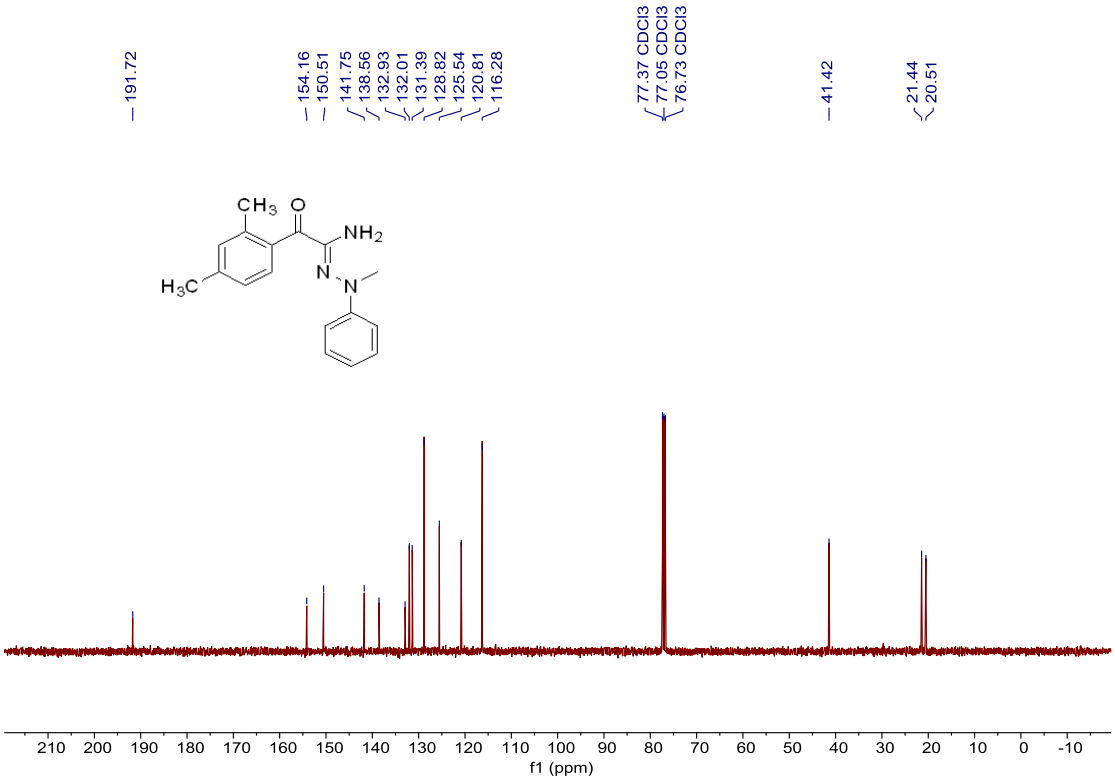
^{13}C NMR of product 18 in $CDCl_3$ (100 MHz)



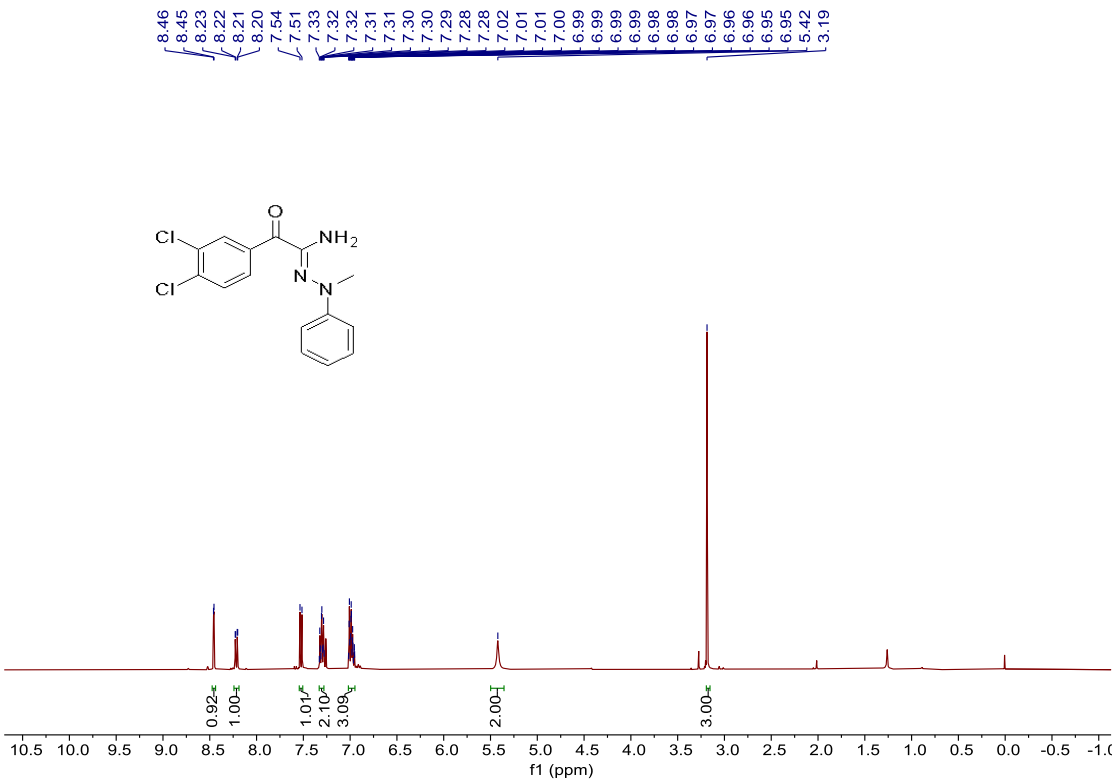
^{1}H NMR of product 19 in $CDCl_3$ (400 MHz)



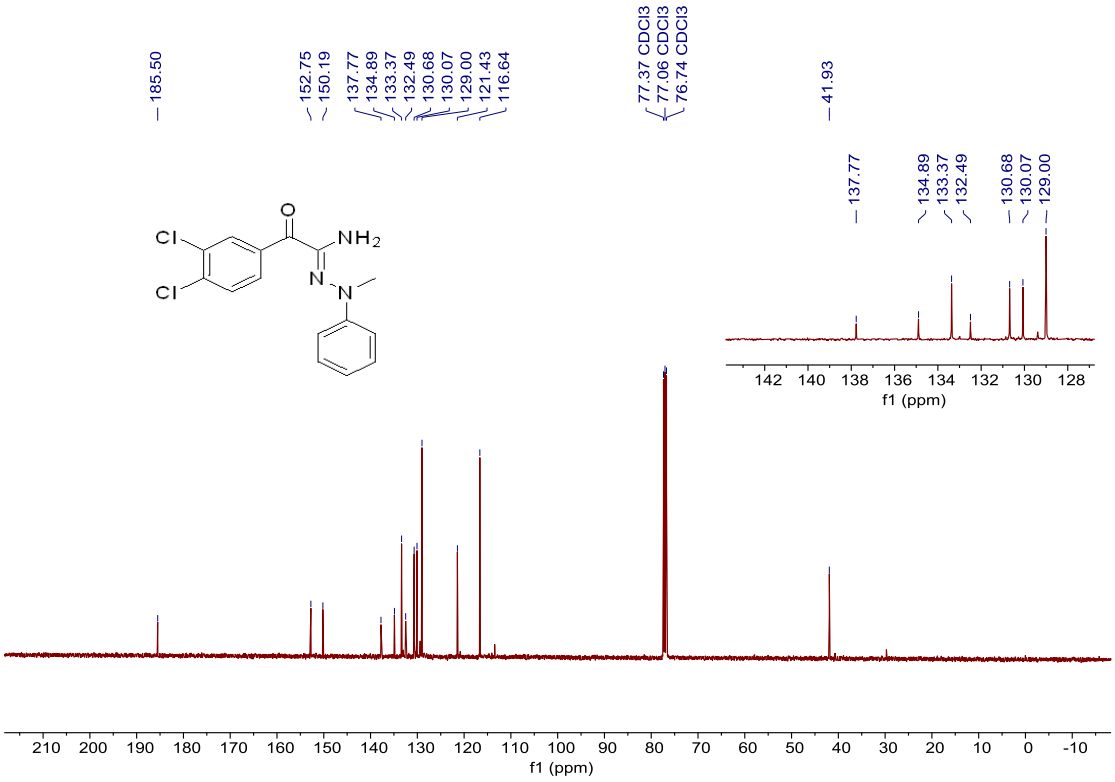
^{13}C NMR of product 19 in $CDCl_3$ (100 MHz)



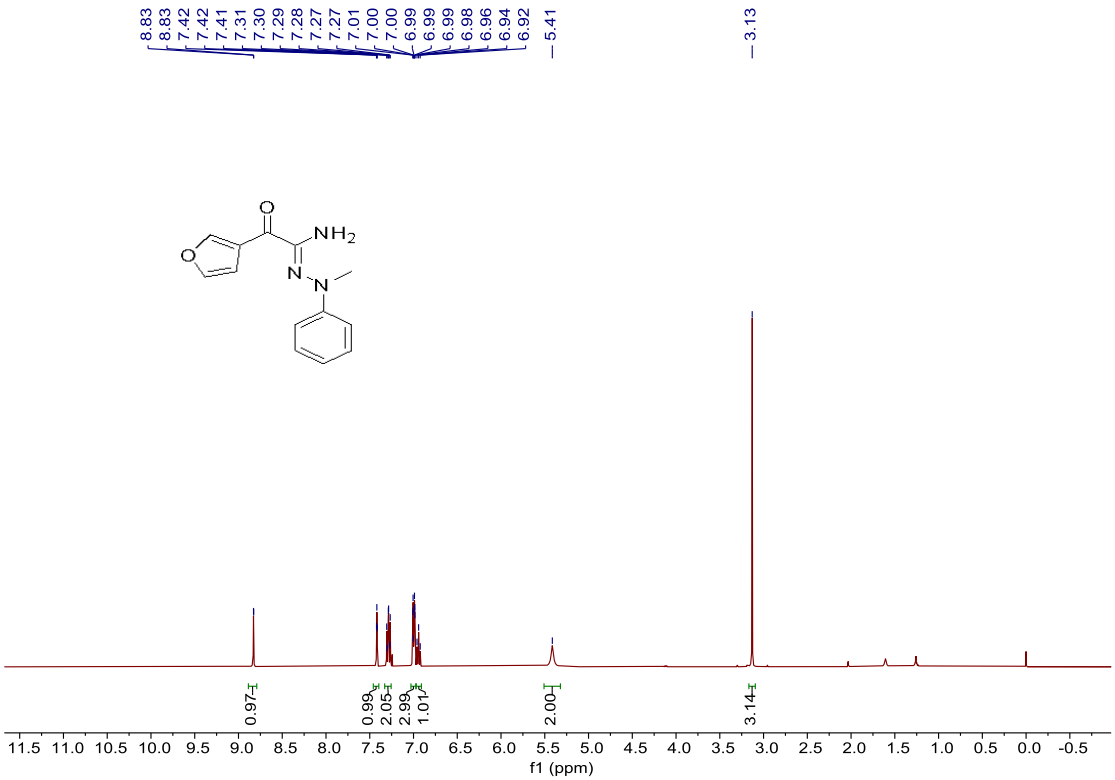
^{1}H NMR of product 20 in $CDCl_3$ (400 MHz)



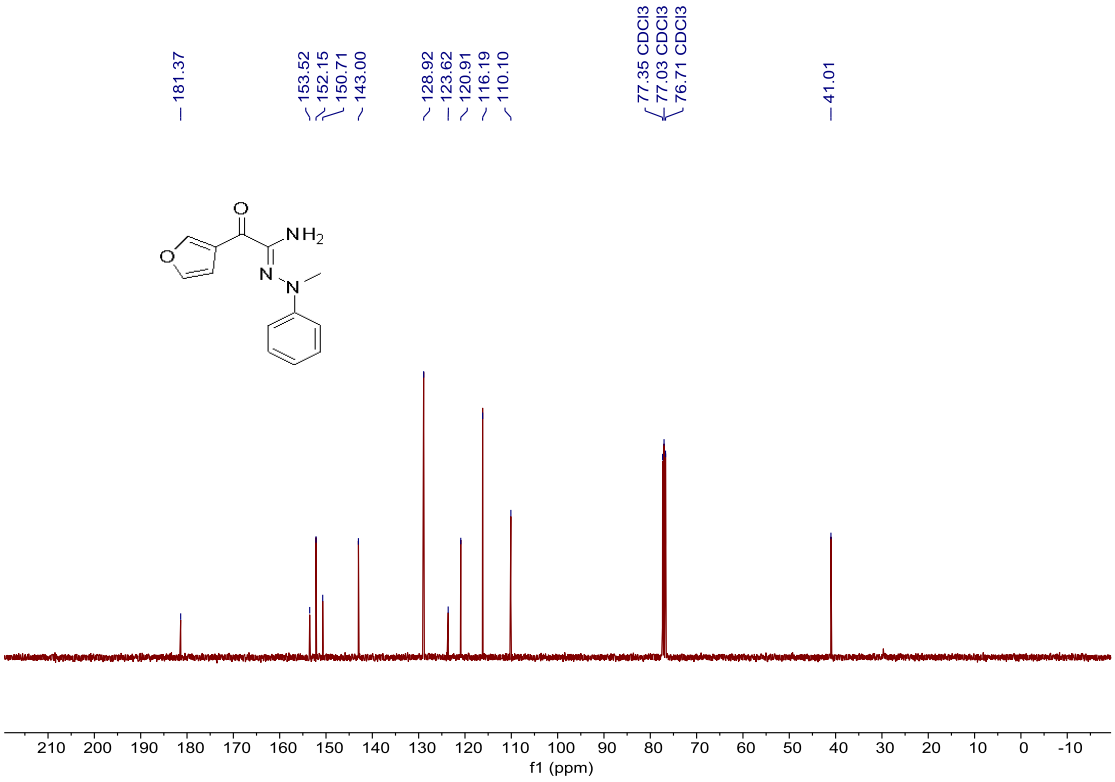
^{13}C NMR of product 20 in $CDCl_3$ (100 MHz)



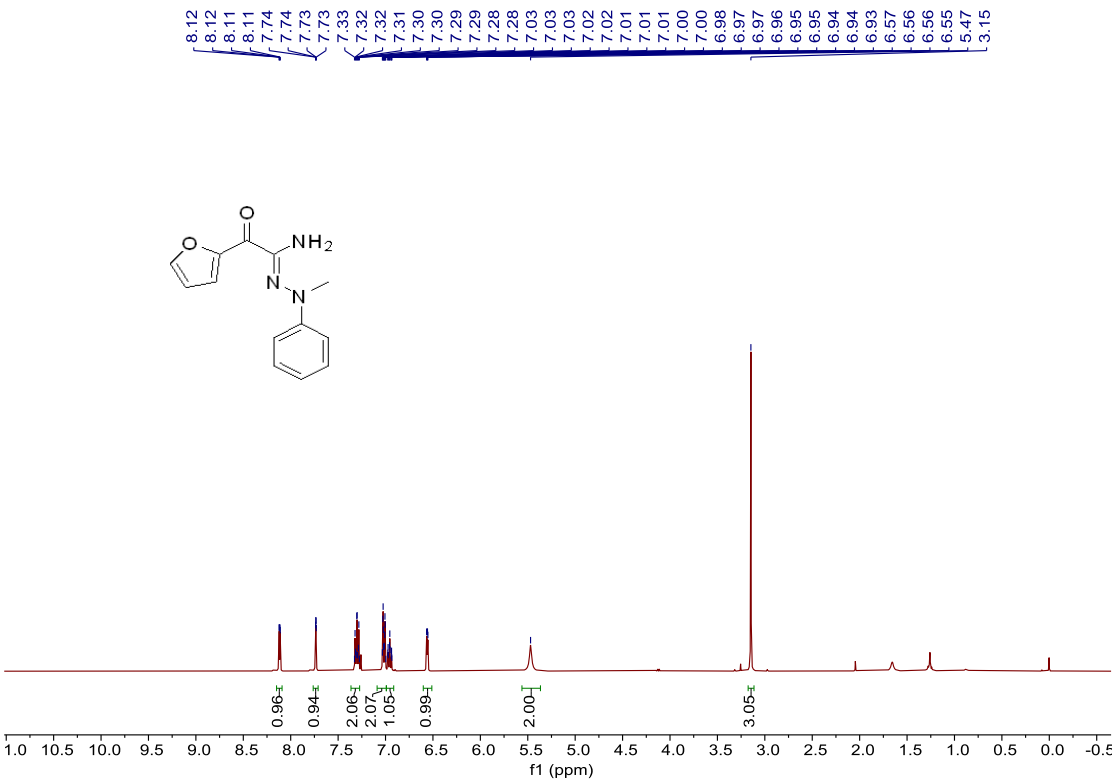
^{1}H NMR of product 21 in $CDCl_3$ (400 MHz)



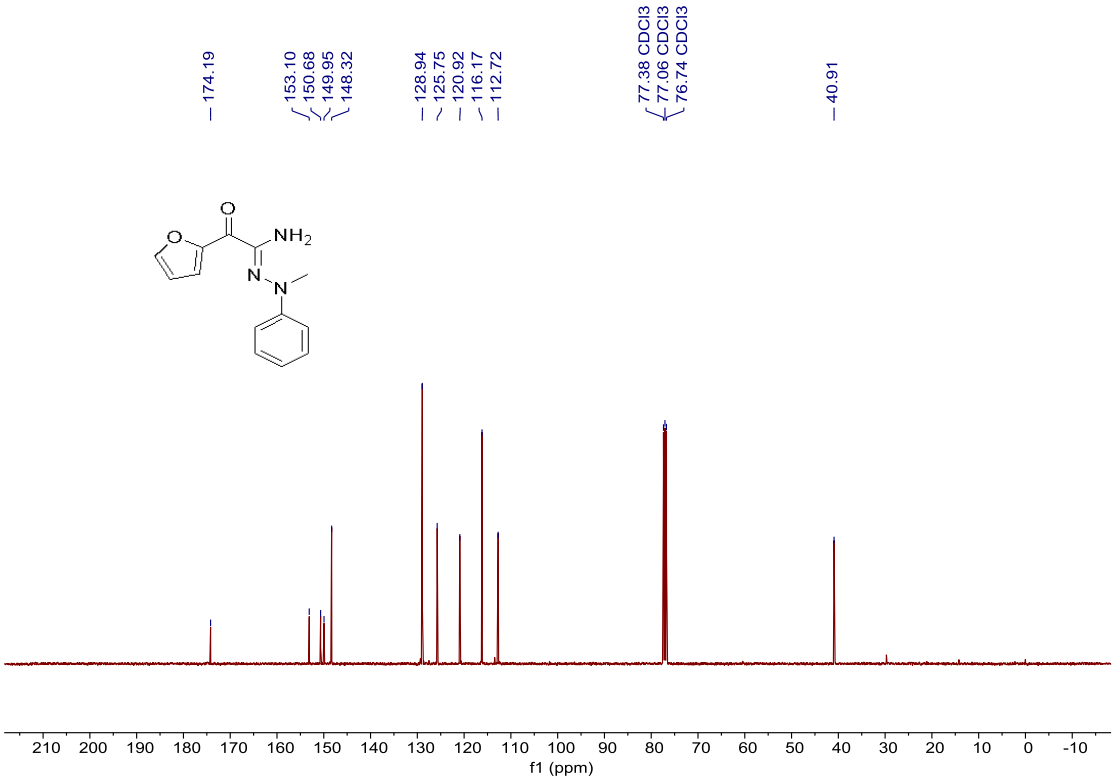
^{13}C NMR of product 21 in $CDCl_3$ (100 MHz)



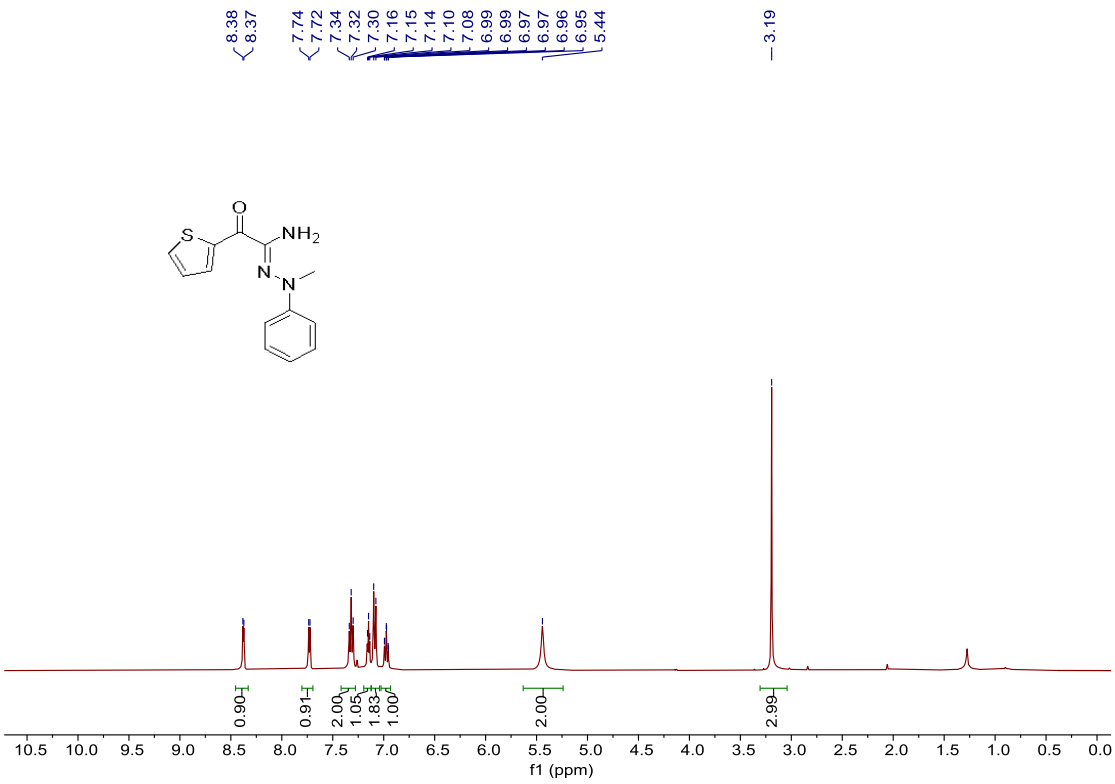
^{1}H NMR of product 22 in $CDCl_3$ (400 MHz)



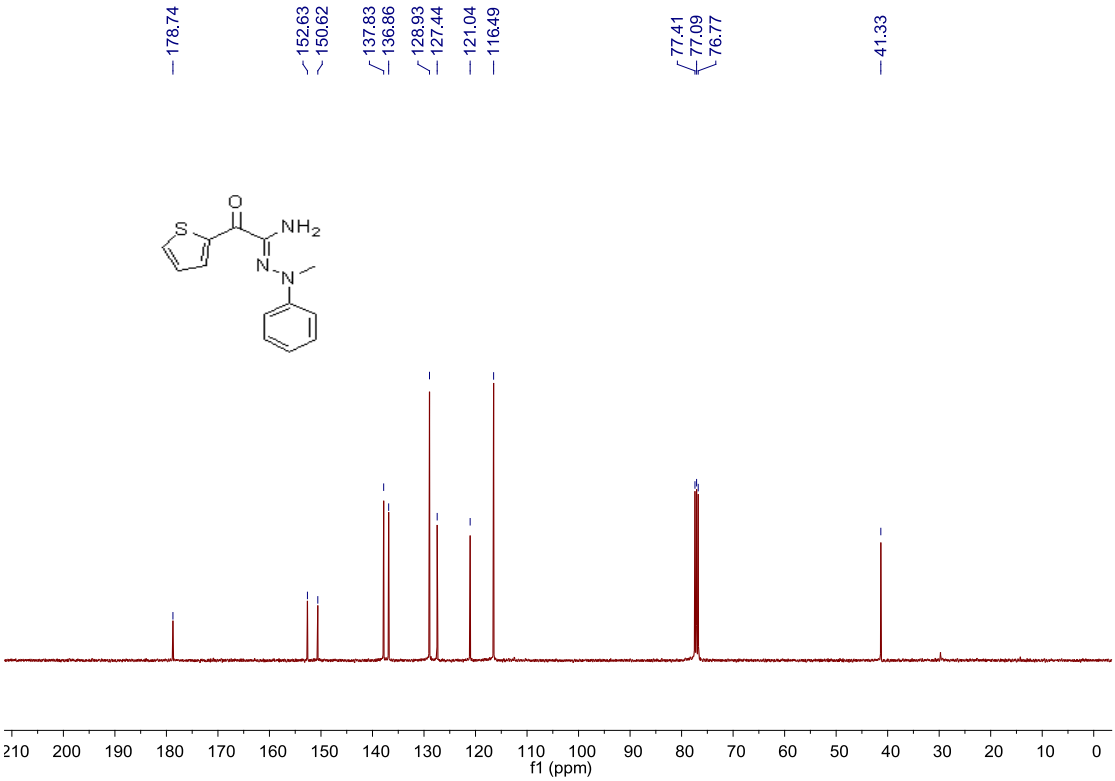
^{13}C NMR of product 22 in $CDCl_3$ (100 MHz)



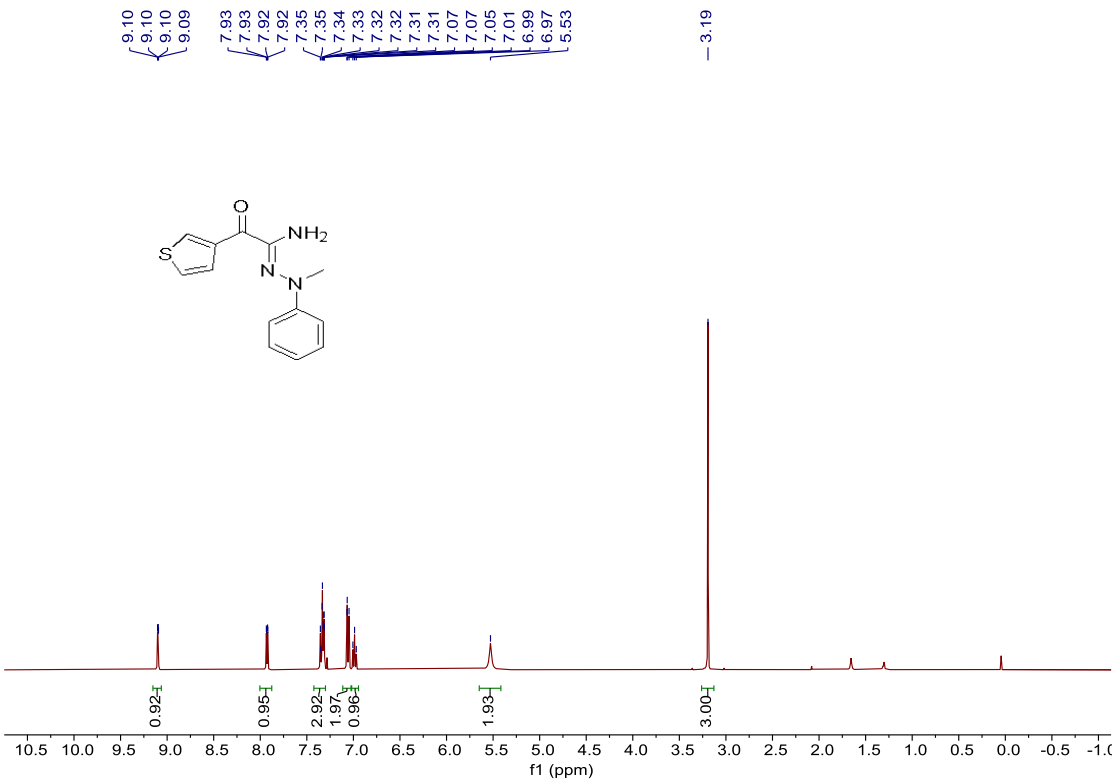
^{1}H NMR of product 23 in $CDCl_3$ (400 MHz)



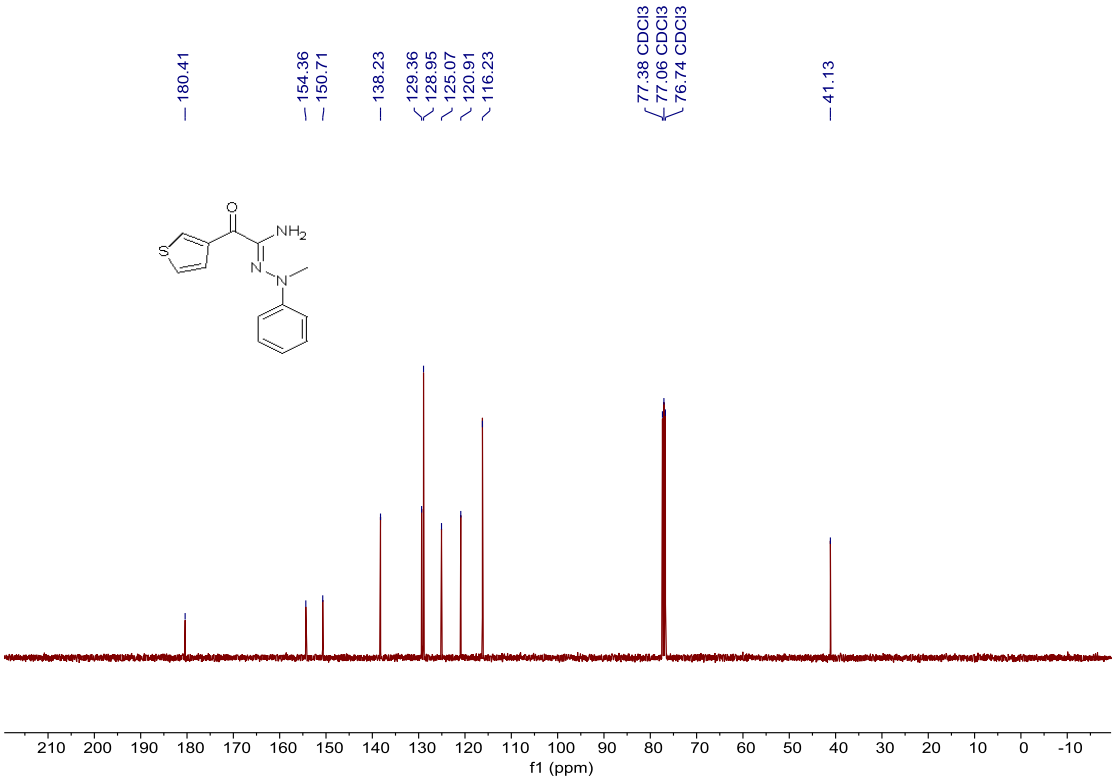
^{13}C NMR of product 23 in $CDCl_3$ (100 MHz)



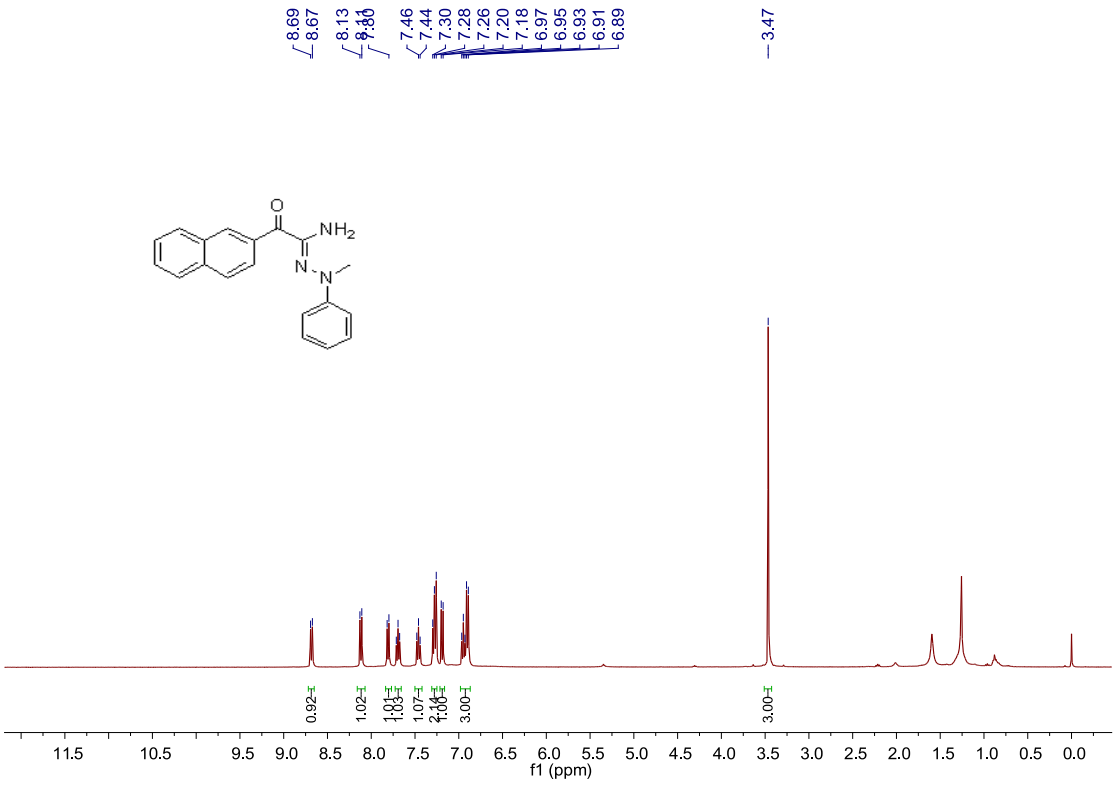
^{1}H NMR of product 24 in $CDCl_3$ (400 MHz)



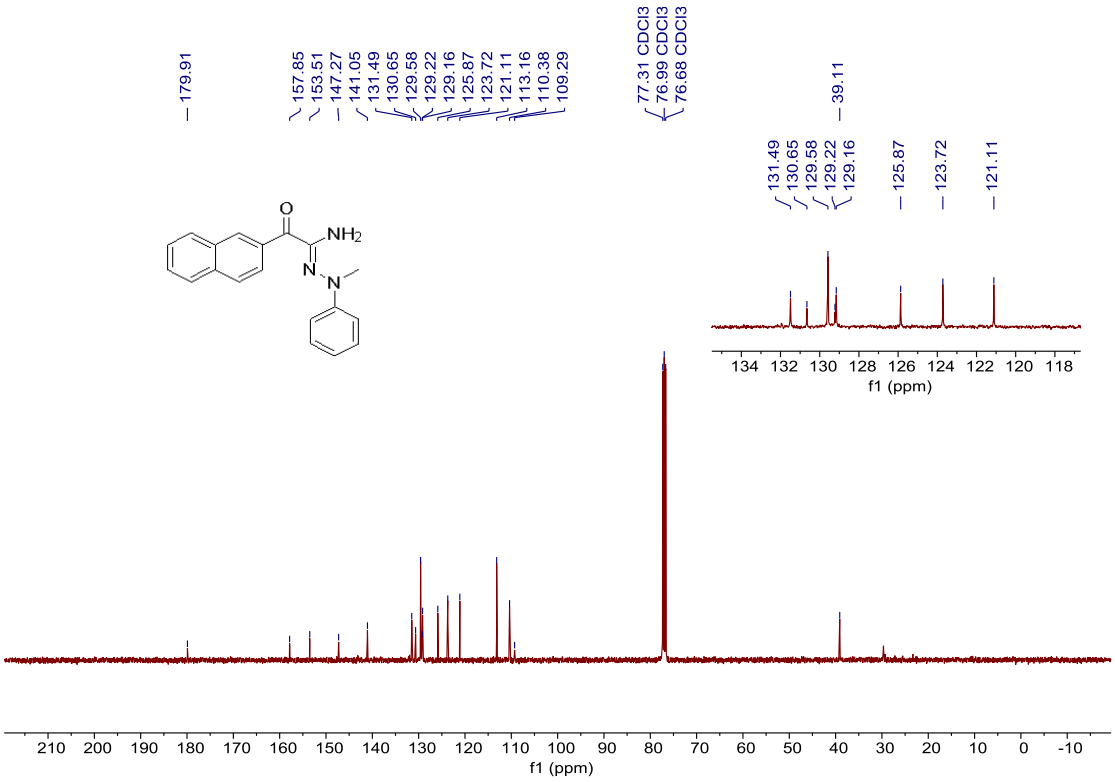
^{13}C NMR of product 24 in $CDCl_3$ (100 MHz)



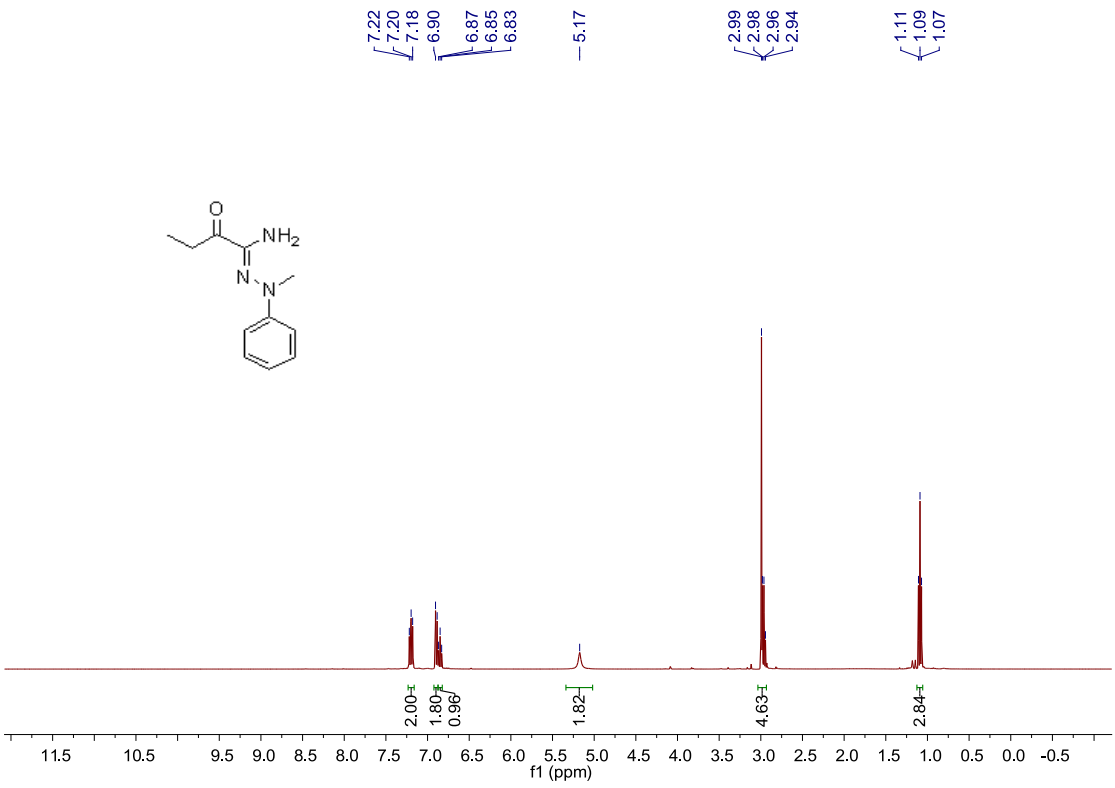
^{1}H NMR of product 26 in $CDCl_3$ (400 MHz)



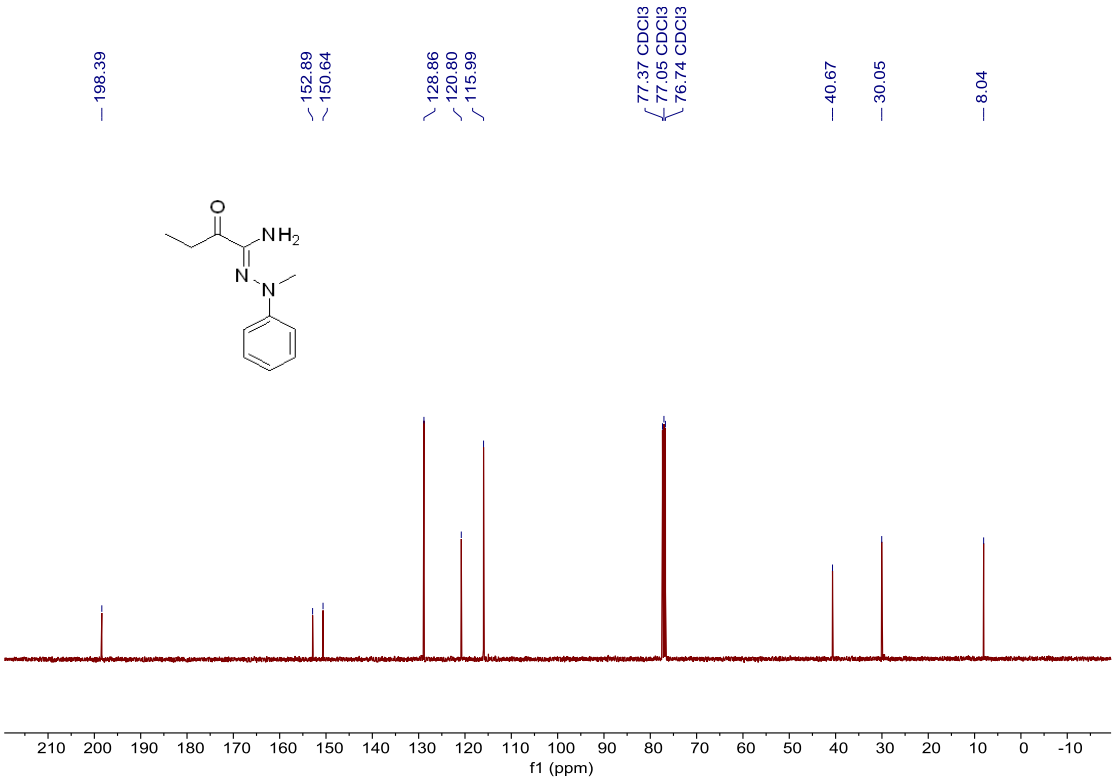
^{13}C NMR of product 26 in $CDCl_3$ (100 MHz)



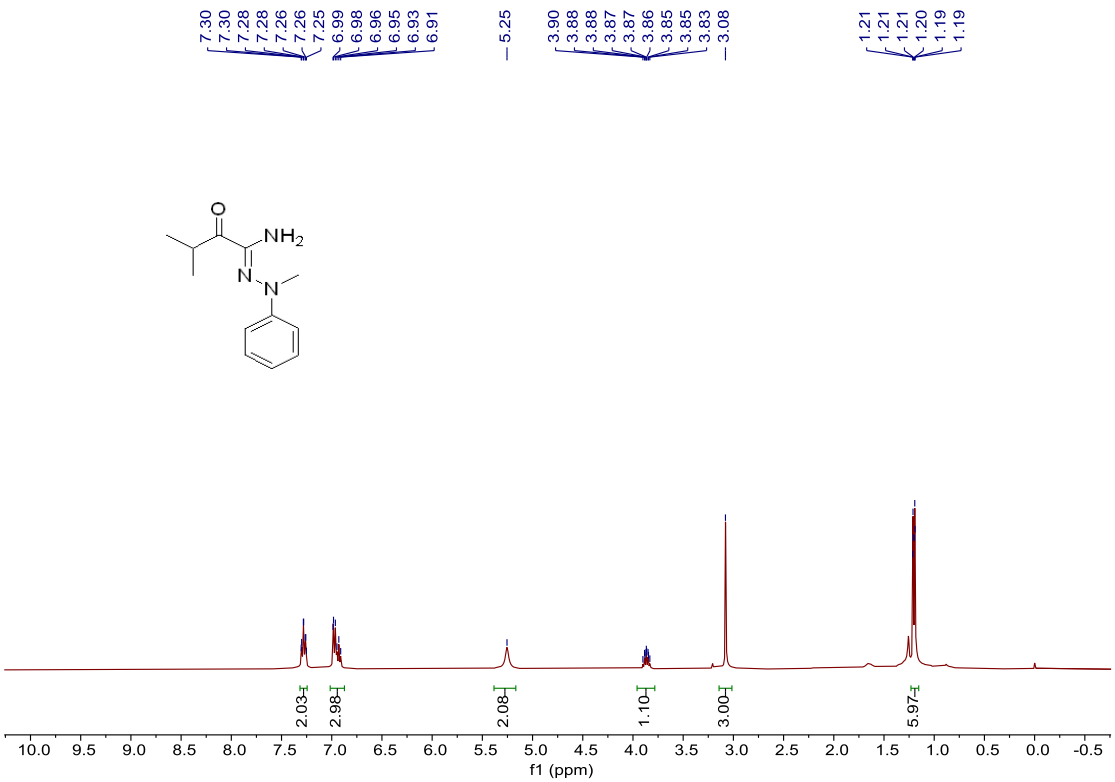
^{1}H NMR of product 27 in $CDCl_3$ (400 MHz)



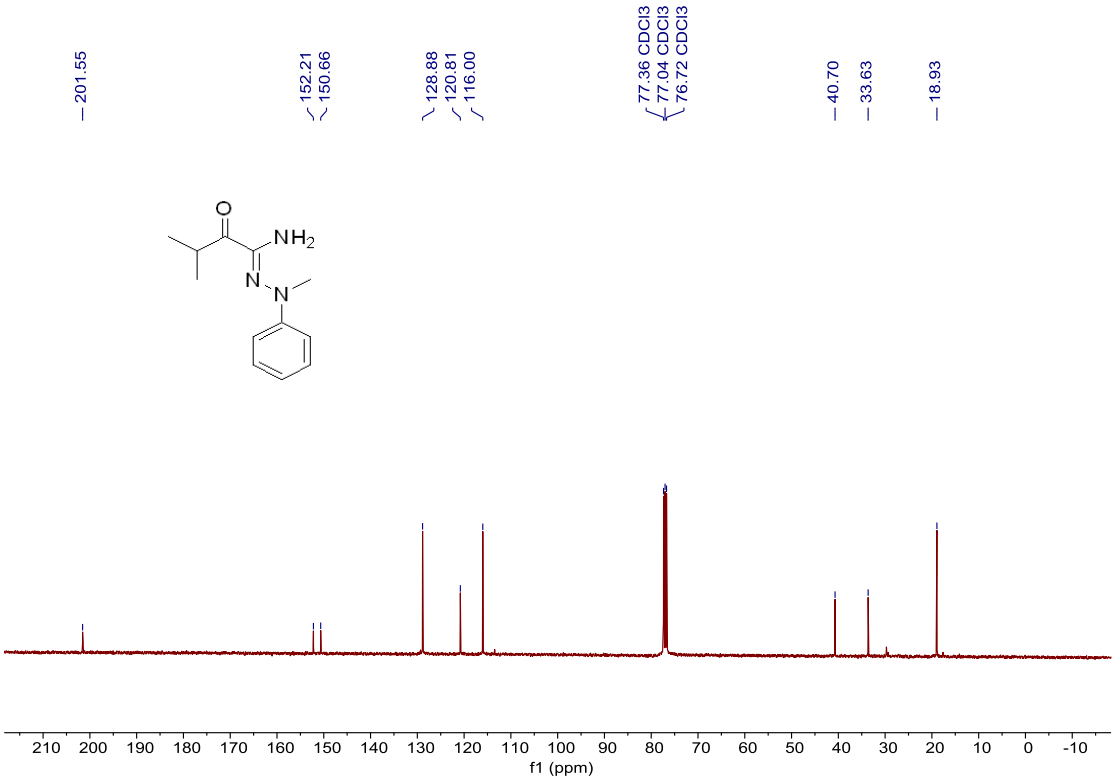
^{13}C NMR of product 27 in $CDCl_3$ (100 MHz)



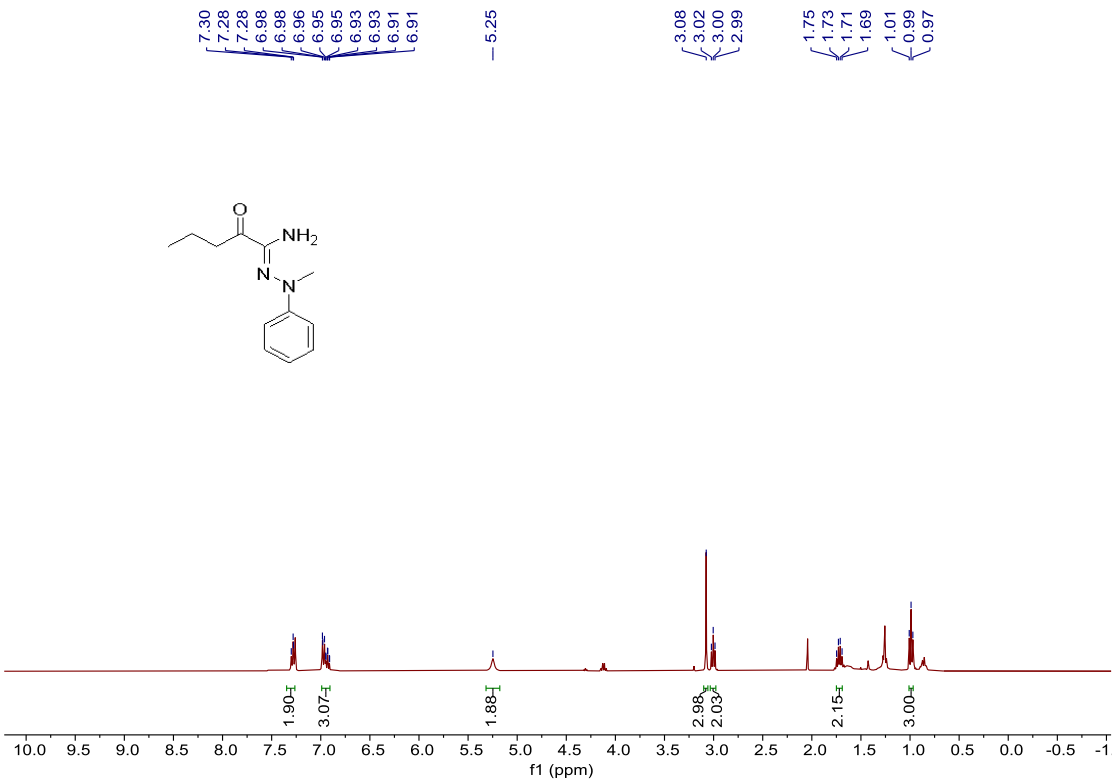
^{1}H NMR of product 28 in $CDCl_3$ (400 MHz)



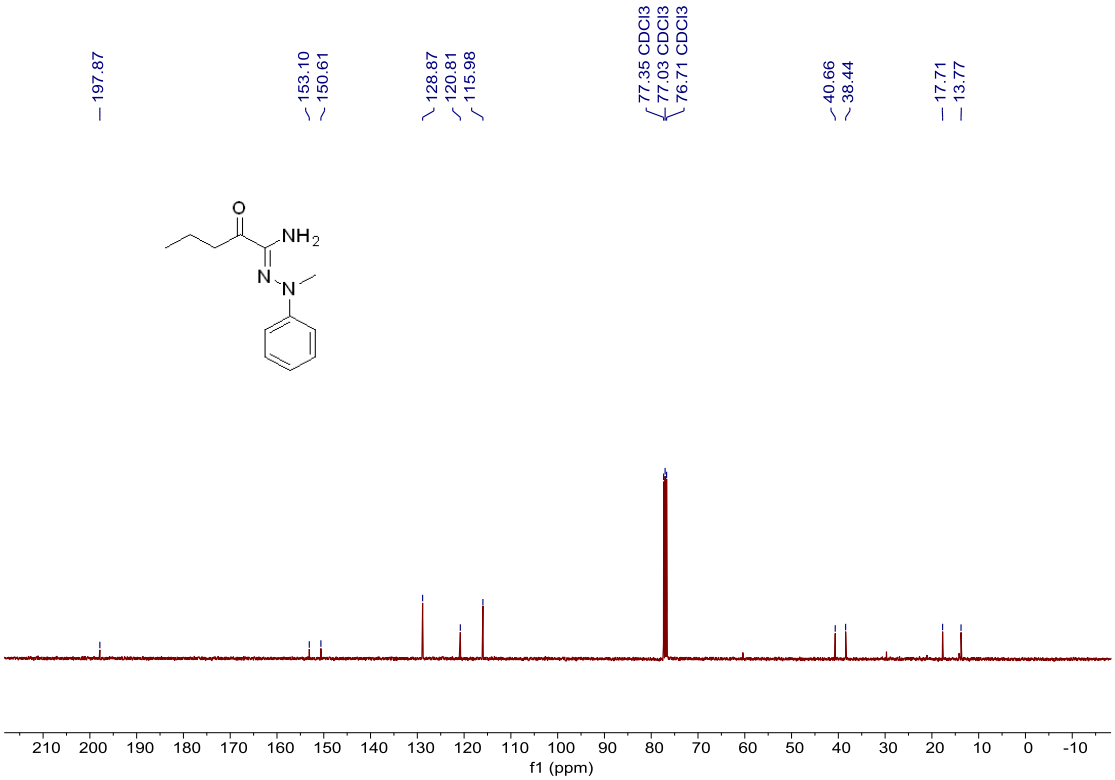
^{13}C NMR of product 28 in $CDCl_3$ (100 MHz)



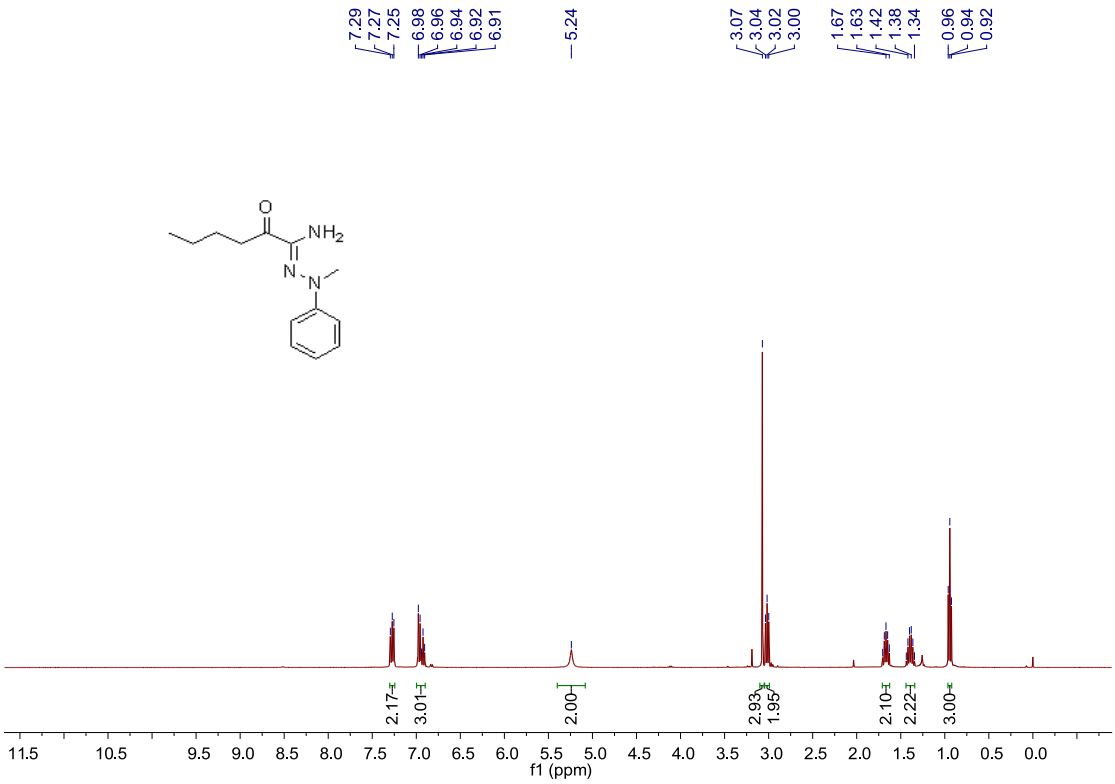
^{1}H NMR of product 29 in $CDCl_3$ (400 MHz)



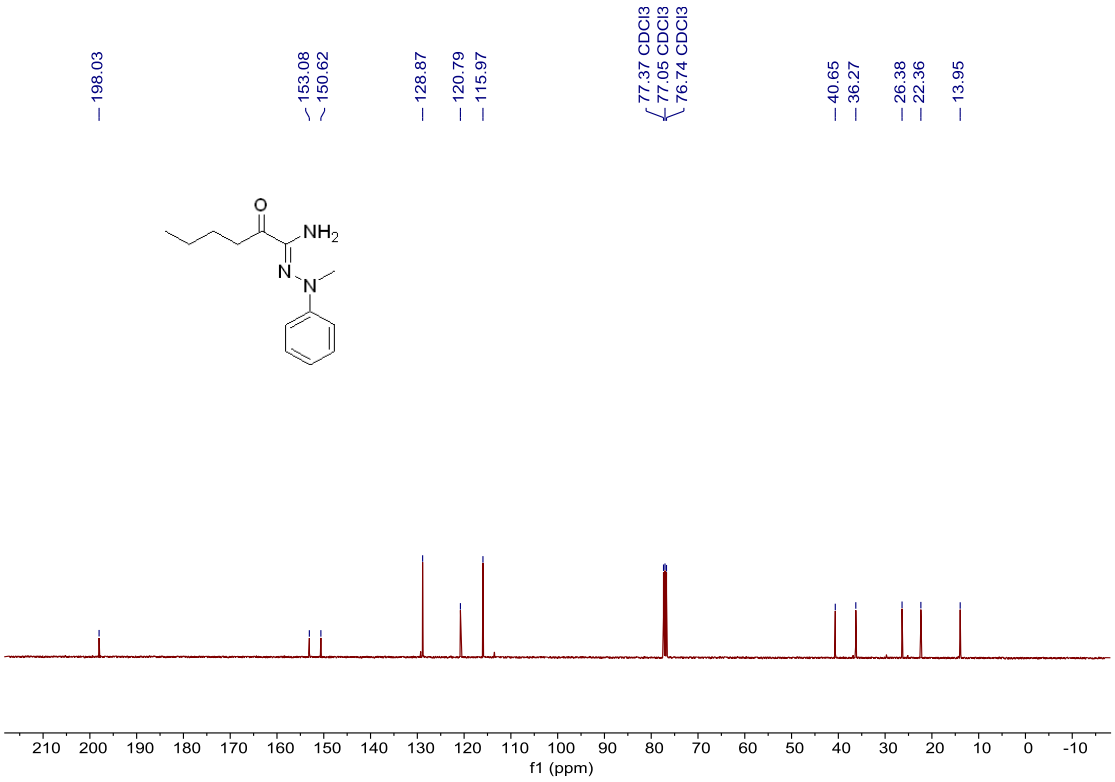
^{13}C NMR of product 29 in $CDCl_3$ (100 MHz)



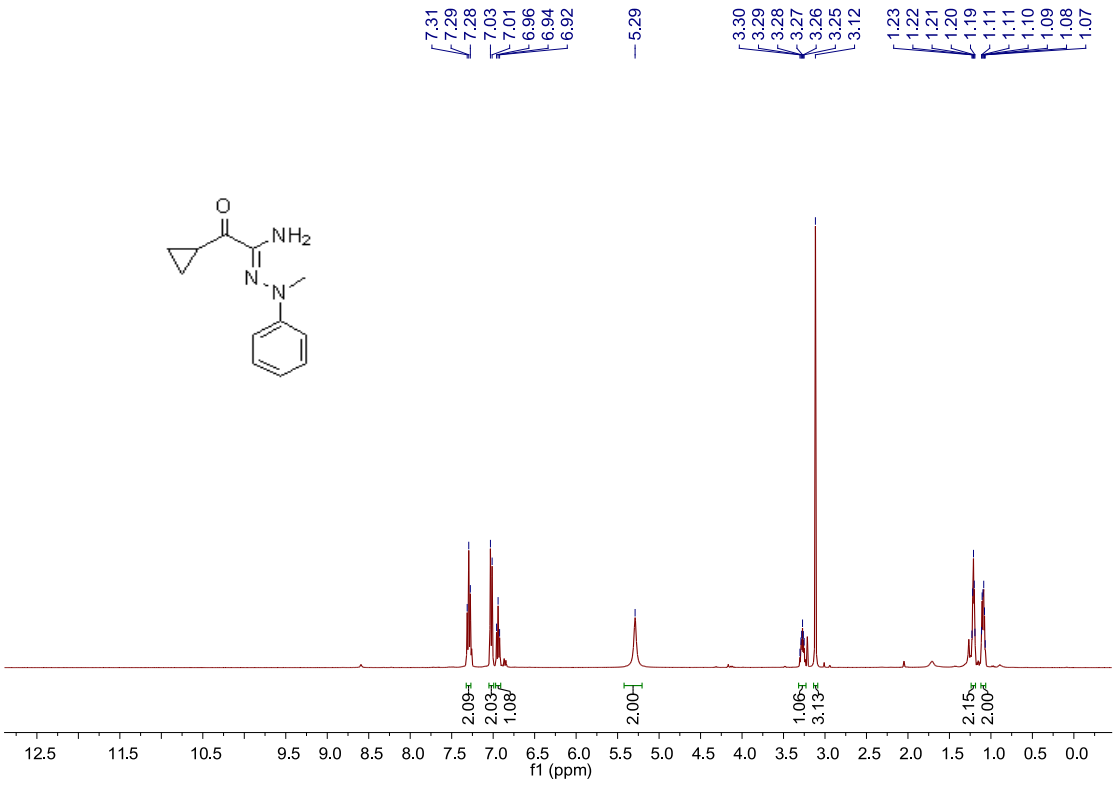
^{1}H NMR of product 30 in $CDCl_3$ (400 MHz)



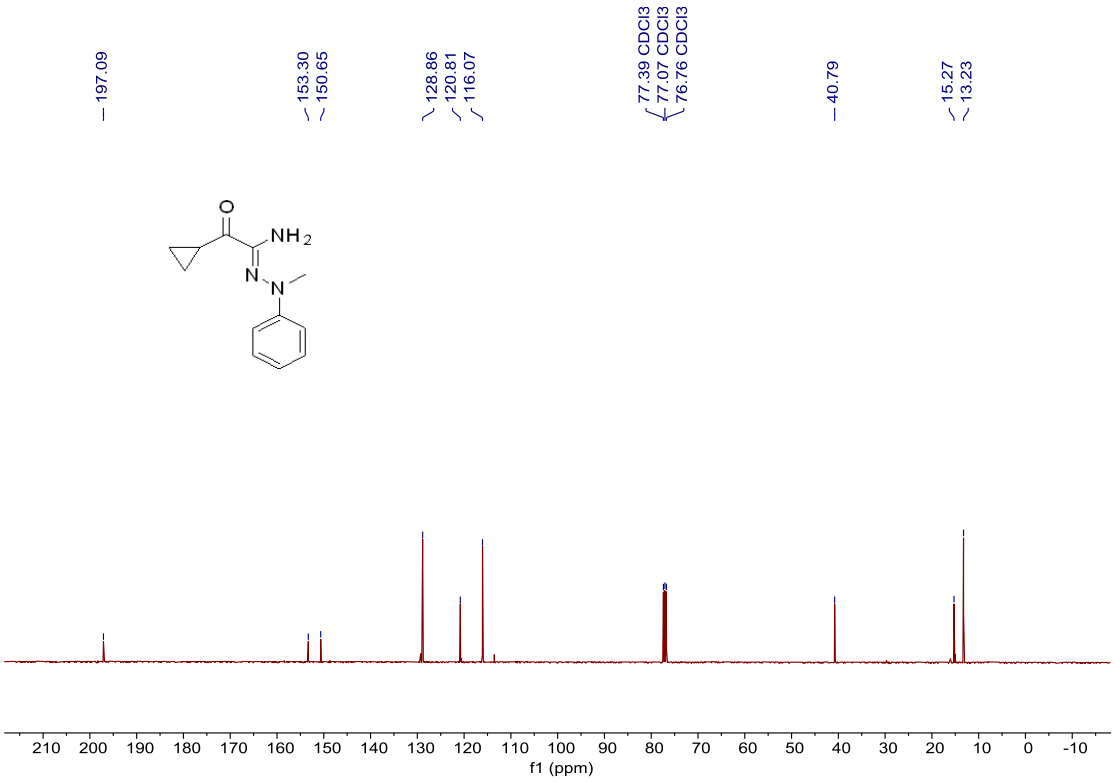
^{13}C NMR of product 30 in $CDCl_3$ (100 MHz)



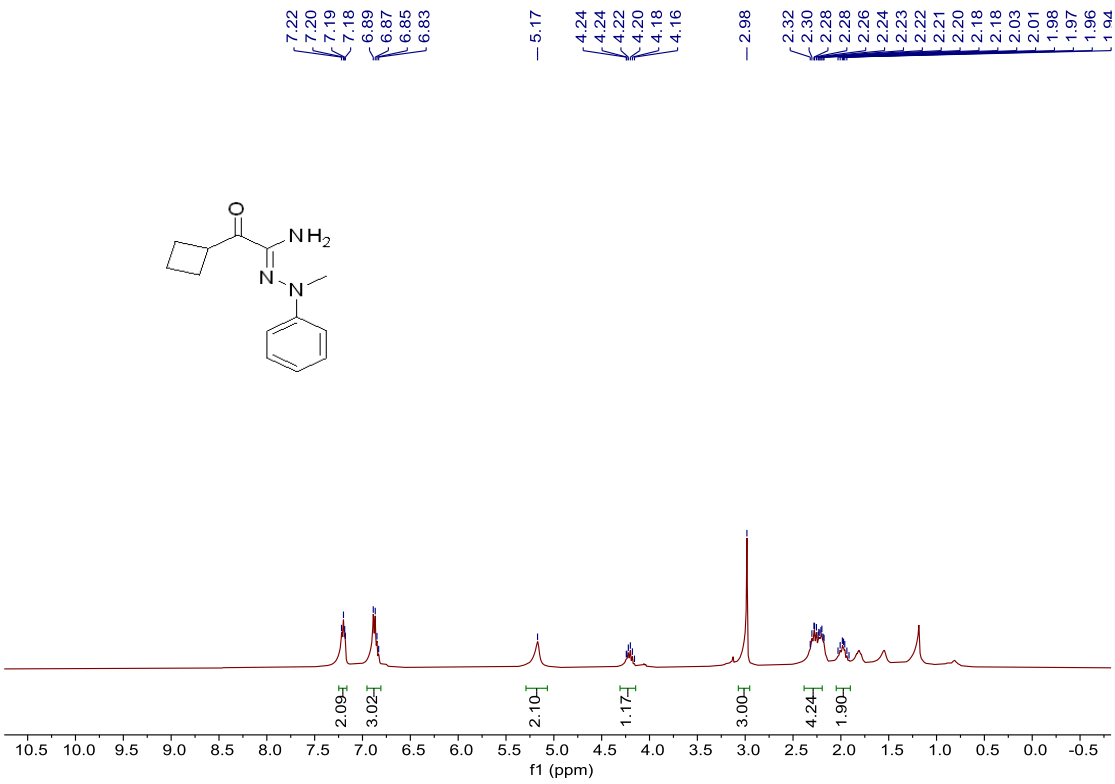
^{1}H NMR of product 31 in $CDCl_3$ (400 MHz)



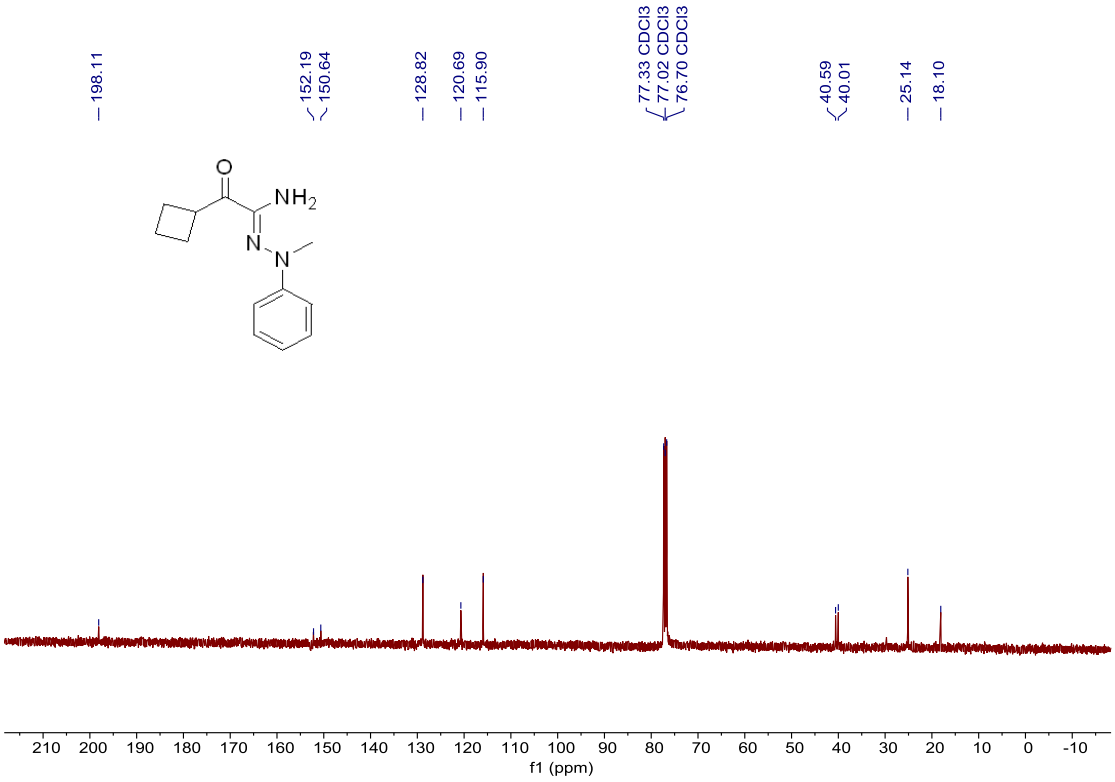
^{13}C NMR of product 31 in $CDCl_3$ (100 MHz)



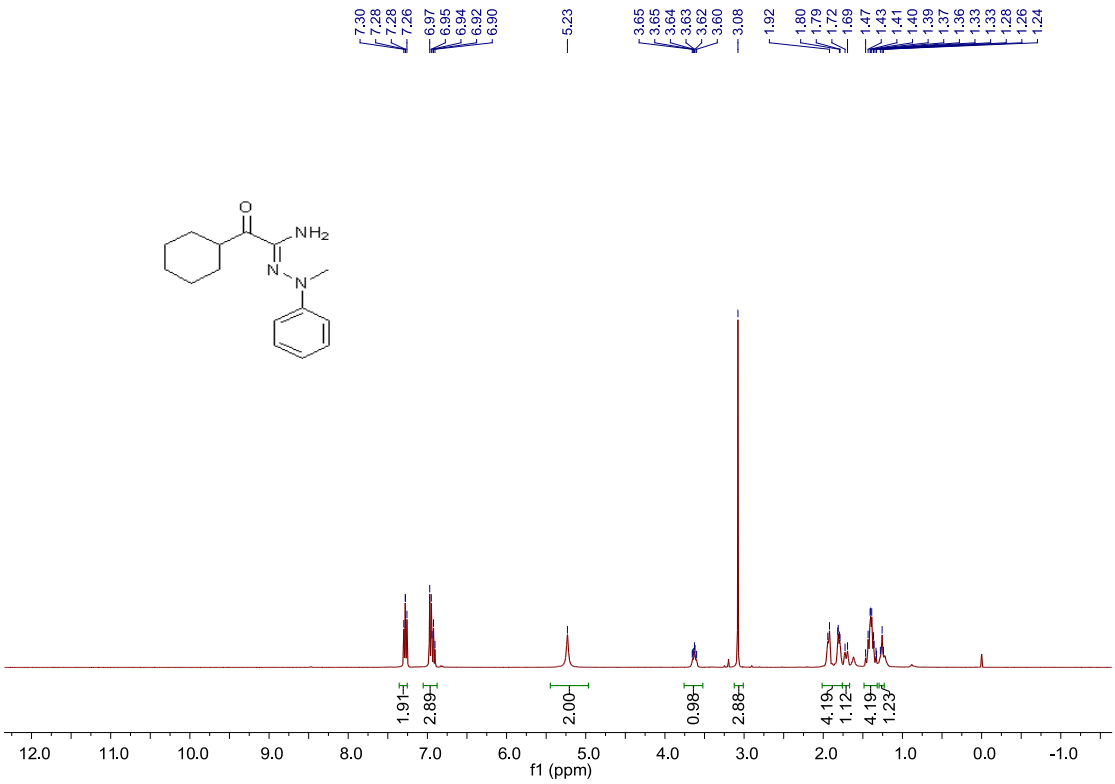
^{1}H NMR of product 32 in $CDCl_3$ (400 MHz)



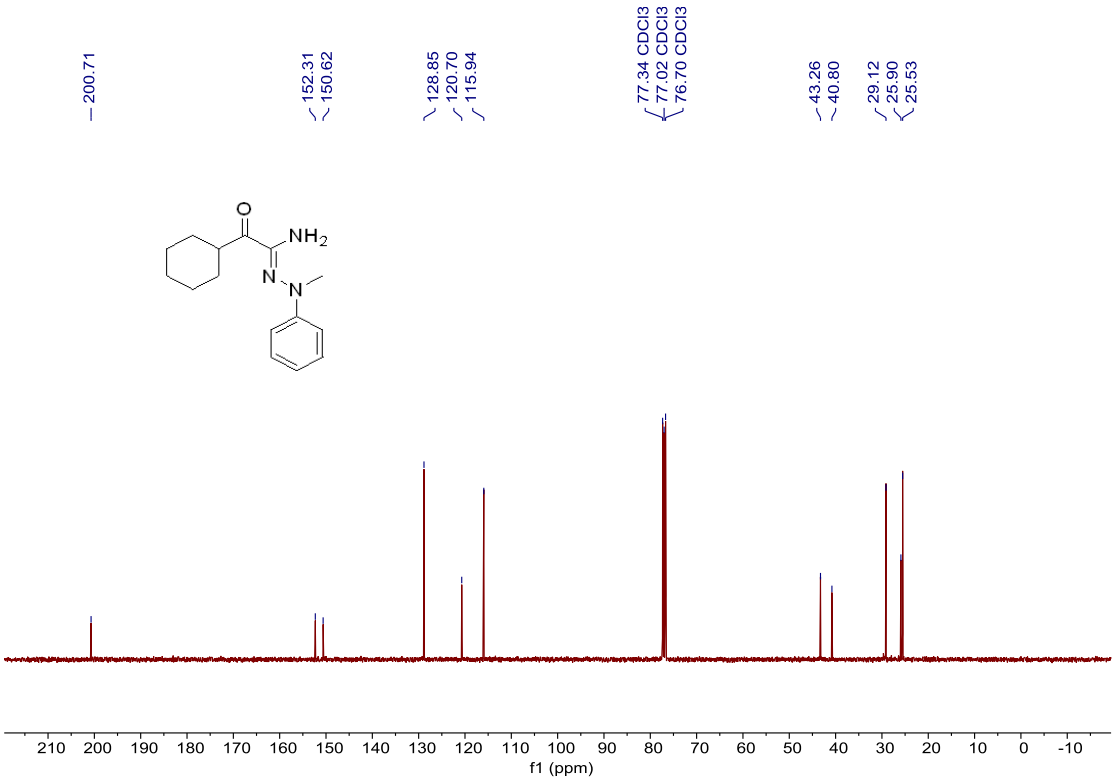
^{13}C NMR of product 32 in $CDCl_3$ (100 MHz)



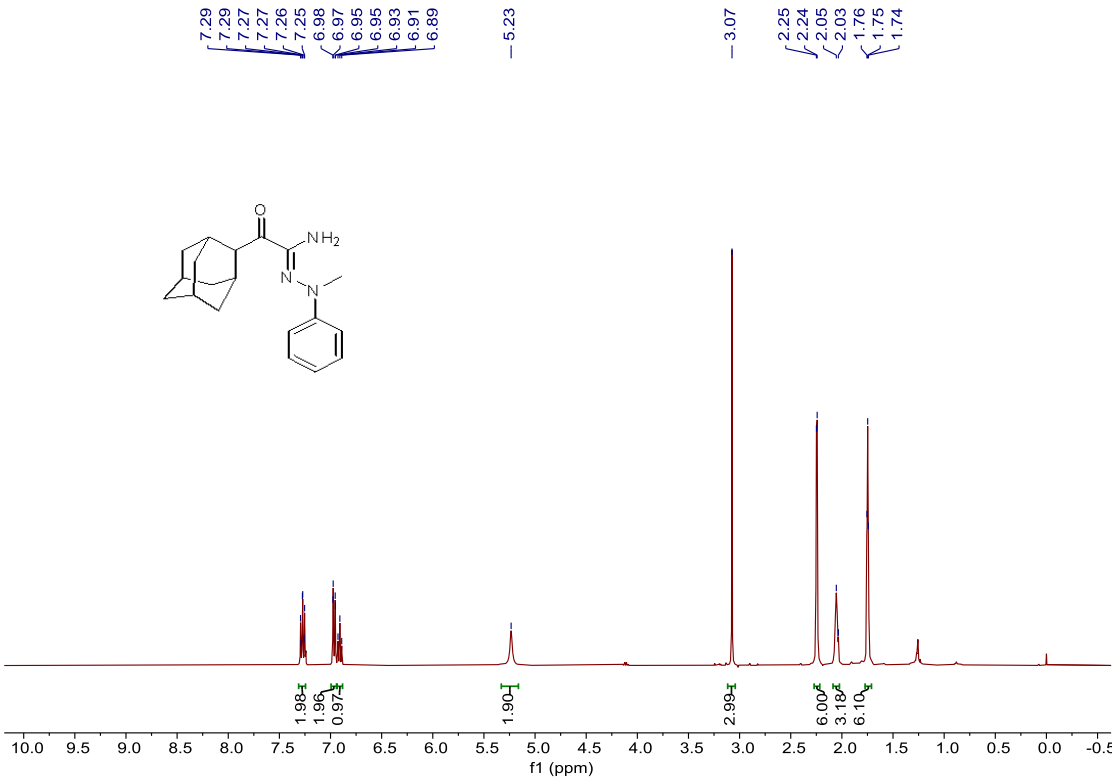
^{1}H NMR of product 33 in $CDCl_3$ (400 MHz)



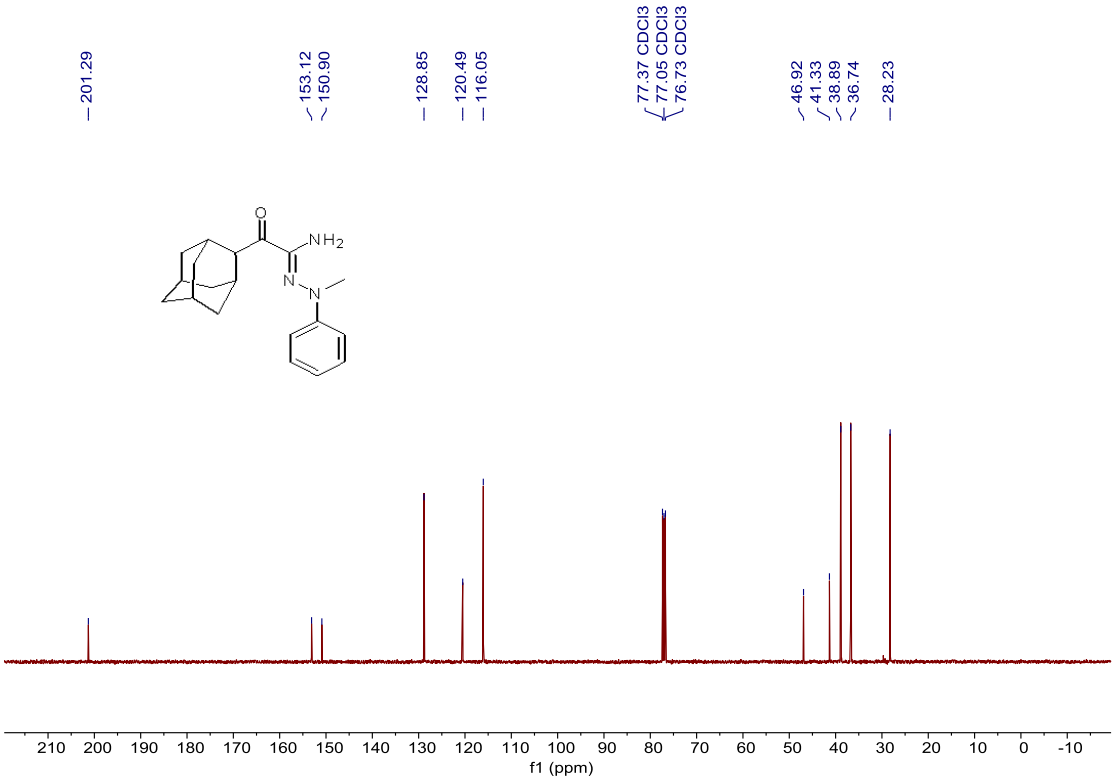
^{13}C NMR of product 33 in $CDCl_3$ (100 MHz)



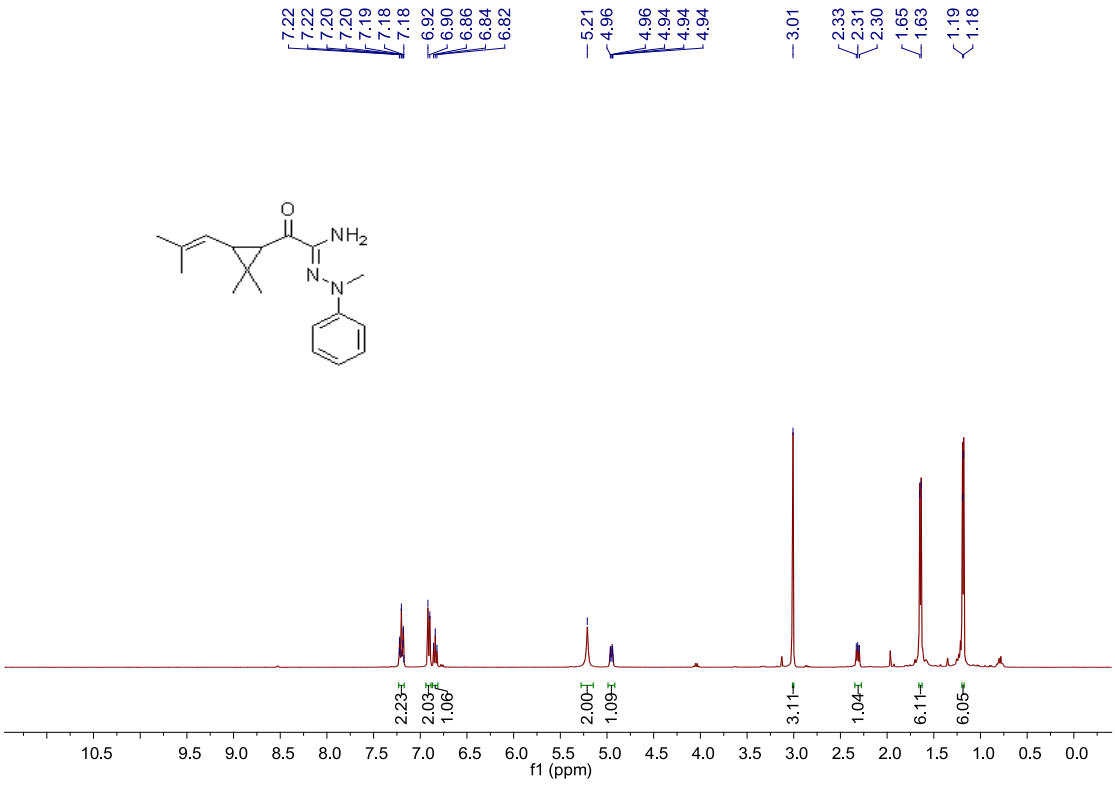
^{1}H NMR of product 34 in $CDCl_3$ (400 MHz)



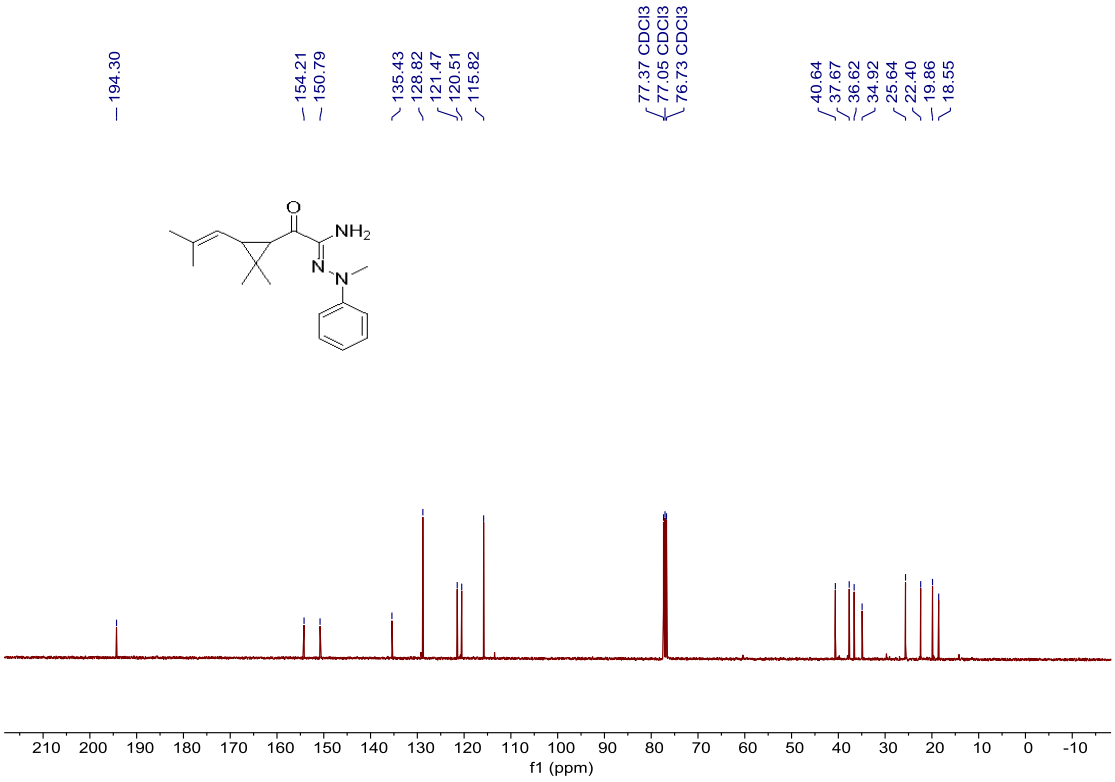
^{13}C NMR of product 34 in $CDCl_3$ (100 MHz)



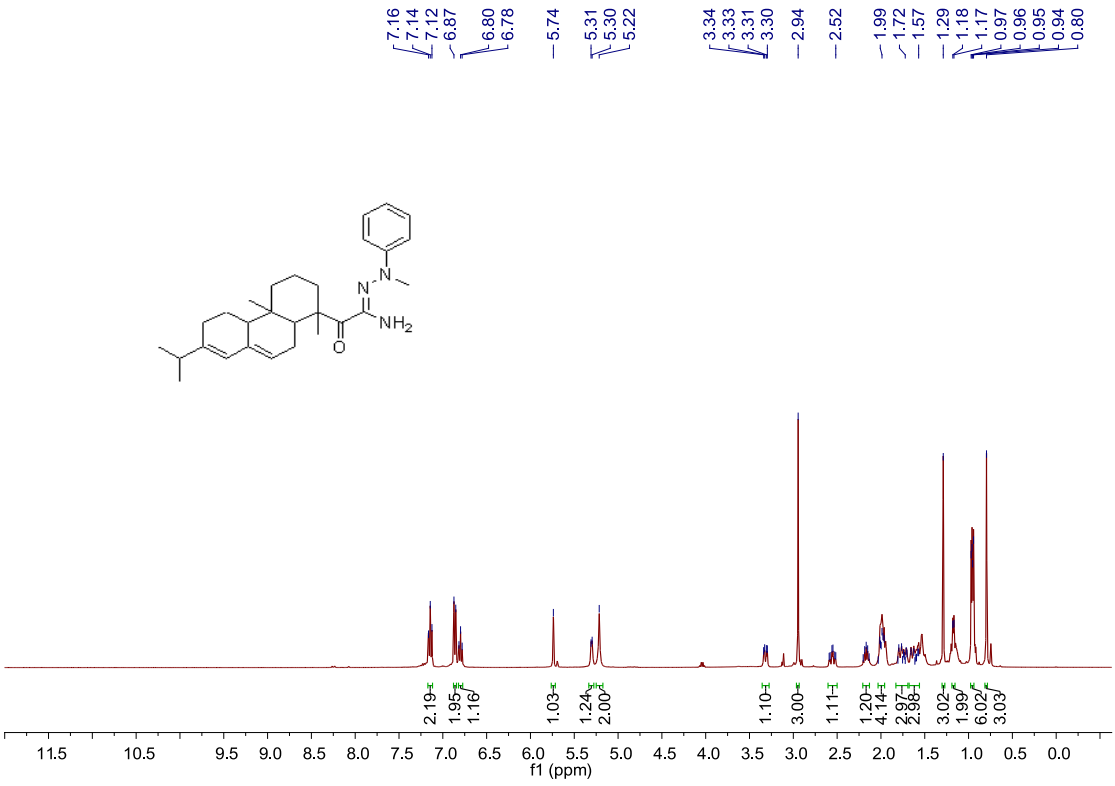
^{1}H NMR of product 35 in $CDCl_3$ (400 MHz)



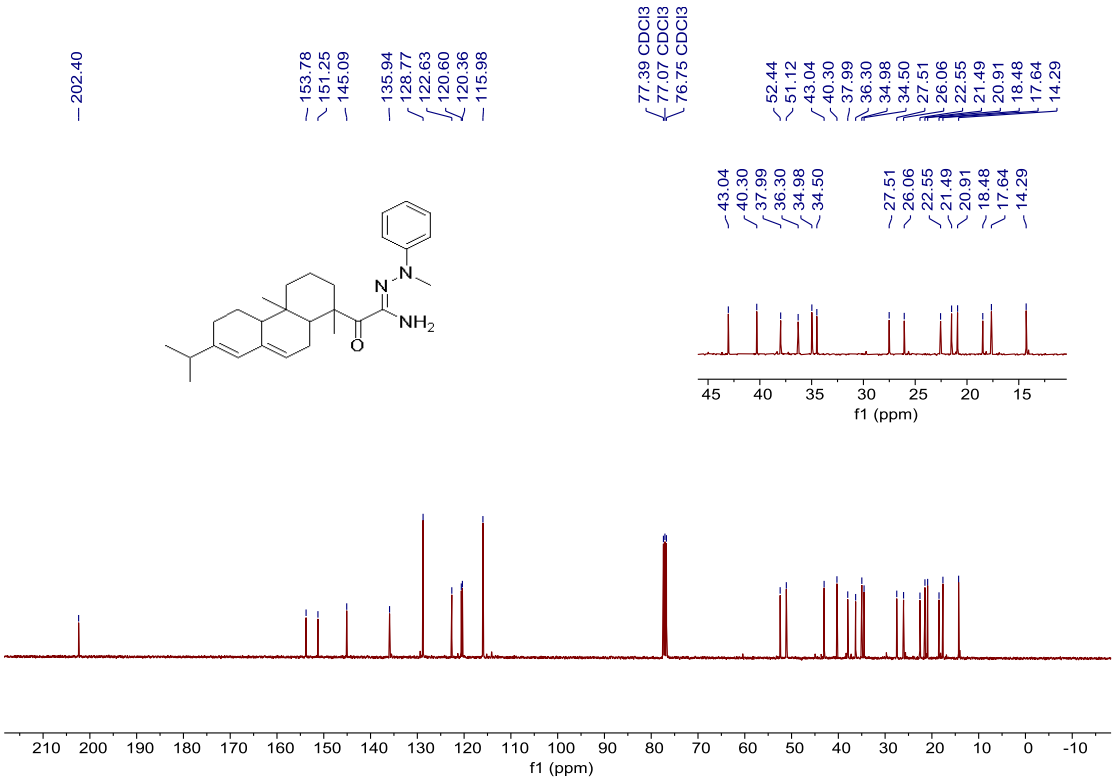
^{13}C NMR of product 35 in $CDCl_3$ (100 MHz)



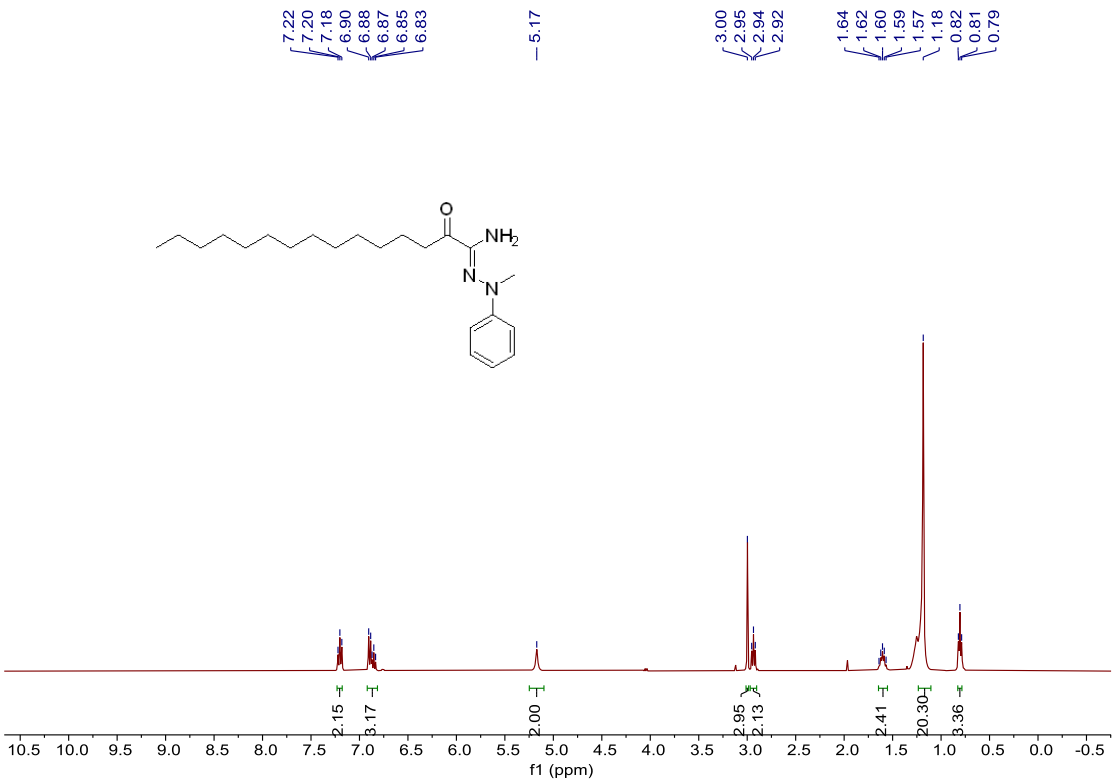
^{1}H NMR of product 36 in $CDCl_3$ (400 MHz)



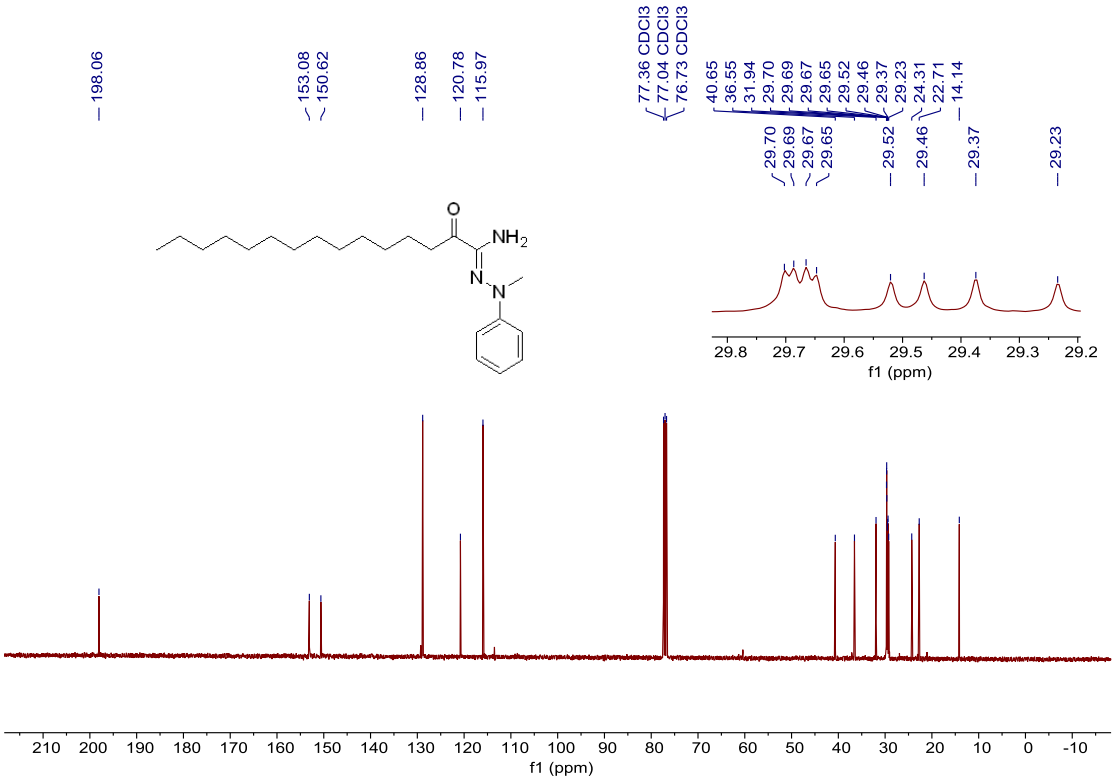
^{13}C NMR of product 36 in $CDCl_3$ (100 MHz)



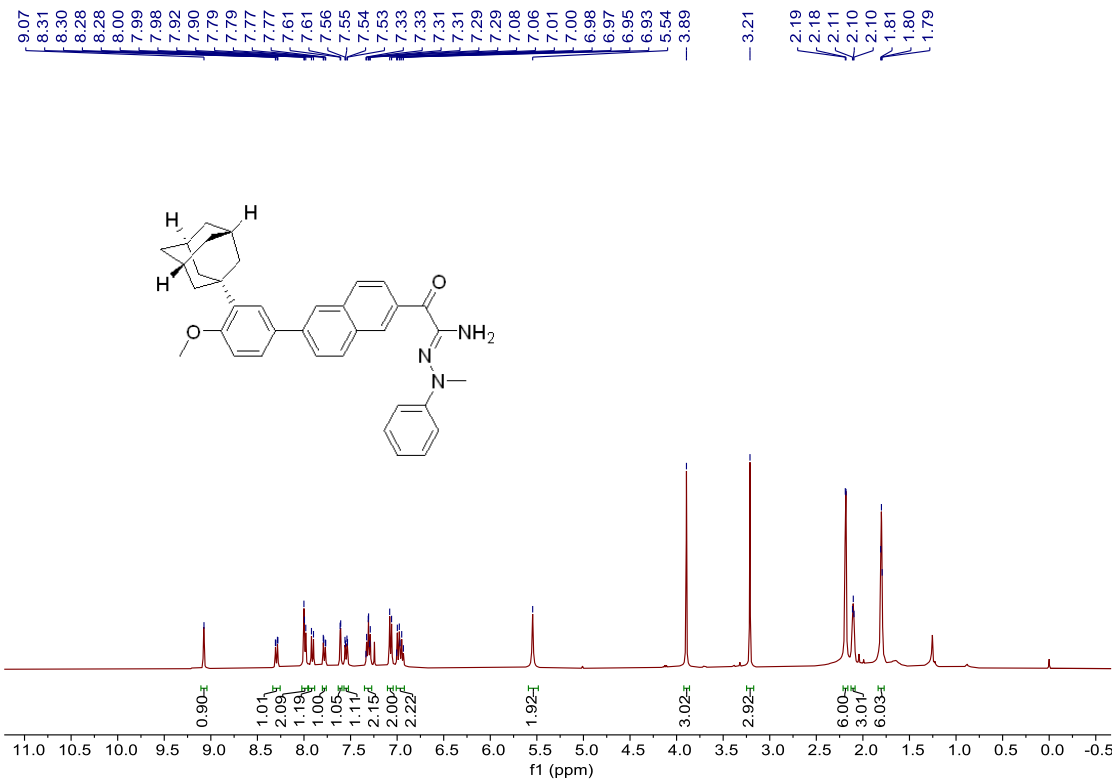
^{1}H NMR of product 37 in $CDCl_3$ (400 MHz)



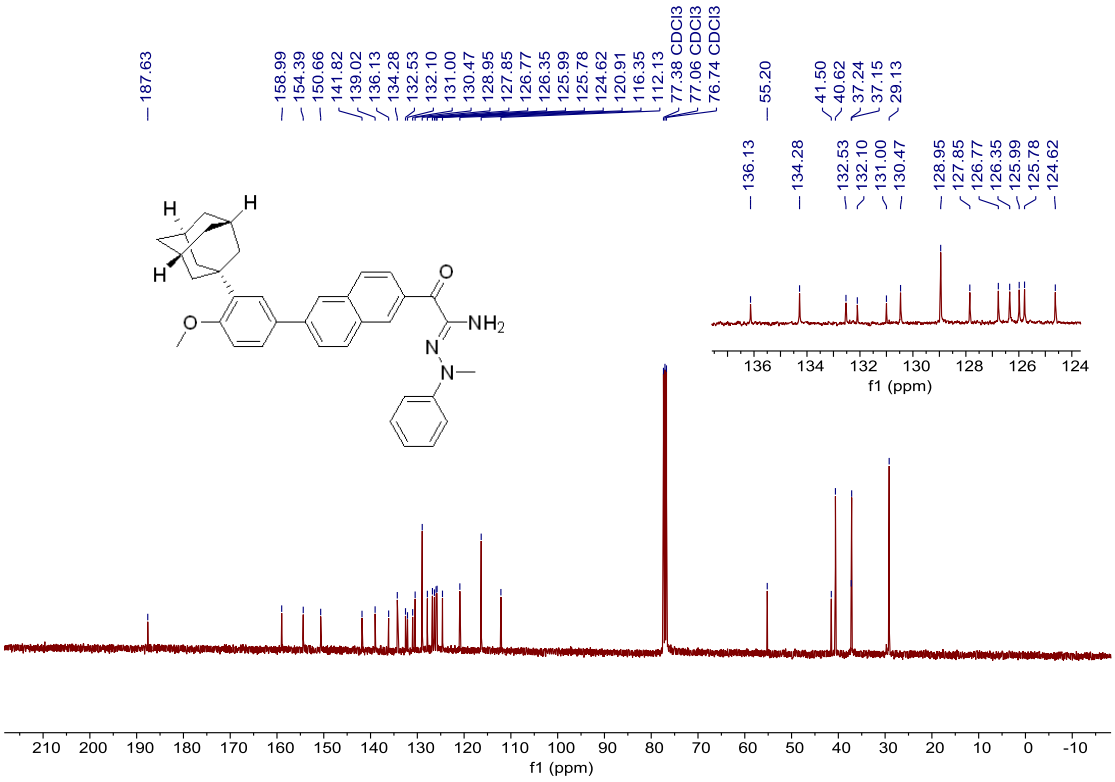
^{13}C NMR of product 37 in $CDCl_3$ (100 MHz)



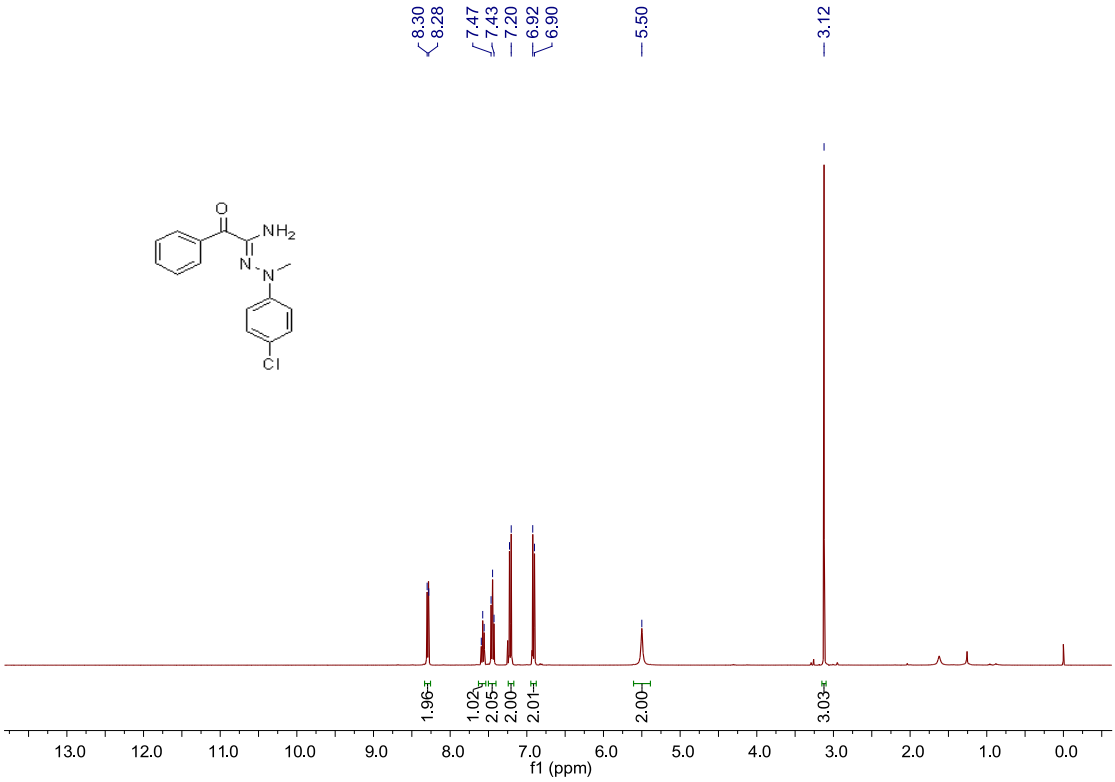
^{1}H NMR of product 38 in $CDCl_3$ (400 MHz)



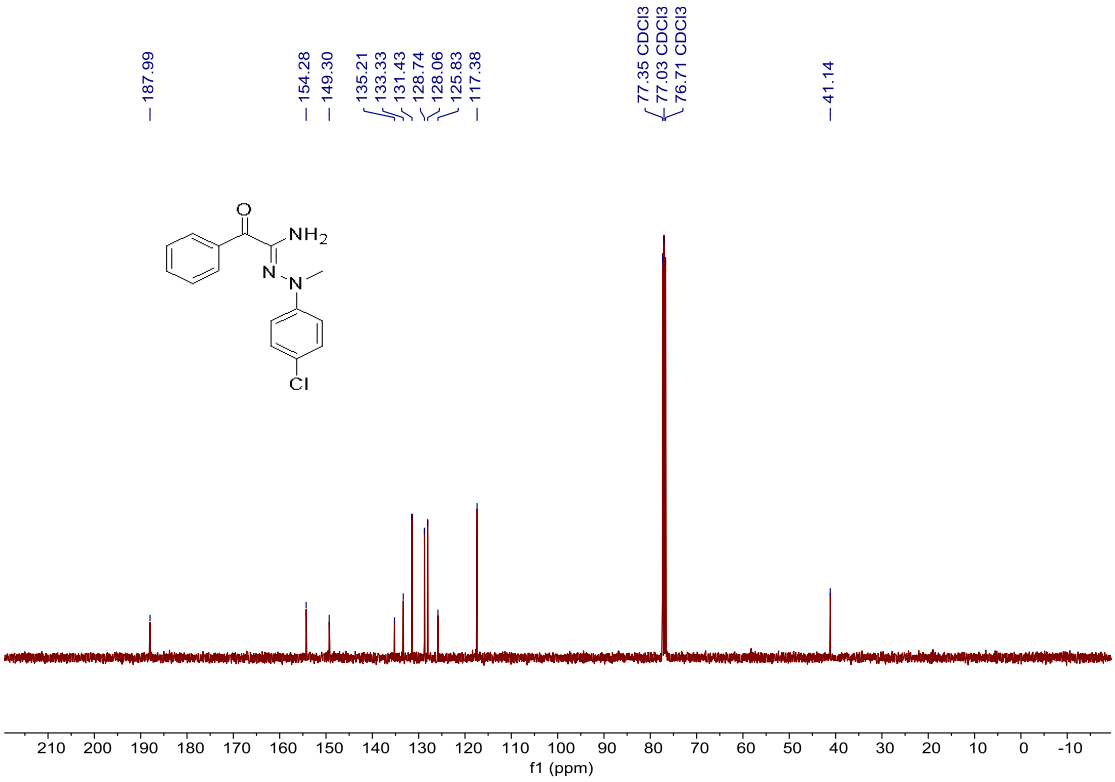
^{13}C NMR of product 38 in $CDCl_3$ (100 MHz)



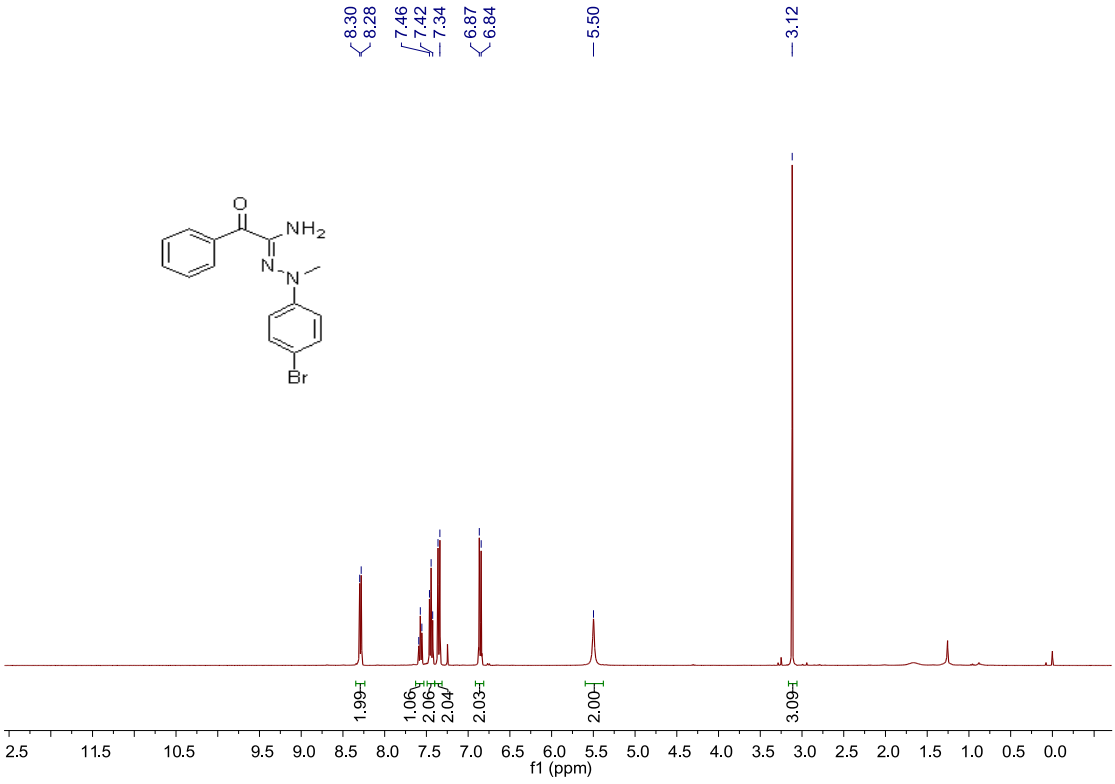
^{1}H NMR of product 39 in $CDCl_3$ (400 MHz)



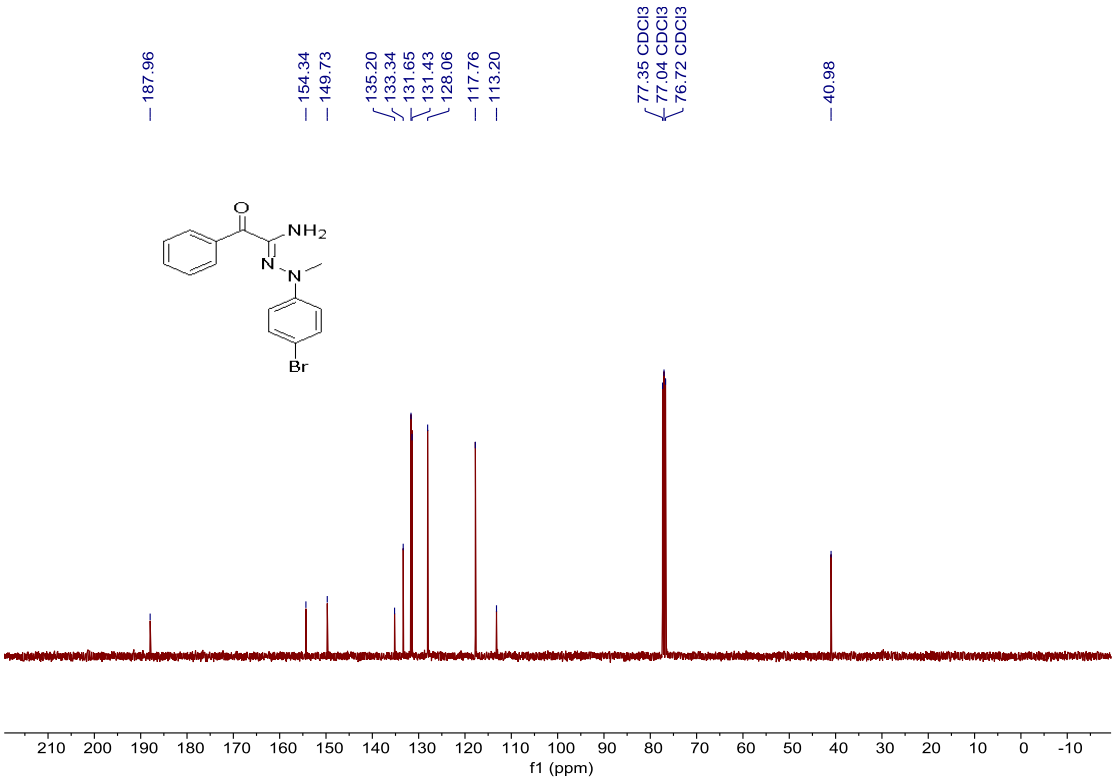
^{13}C NMR of product 39 in $CDCl_3$ (100 MHz)



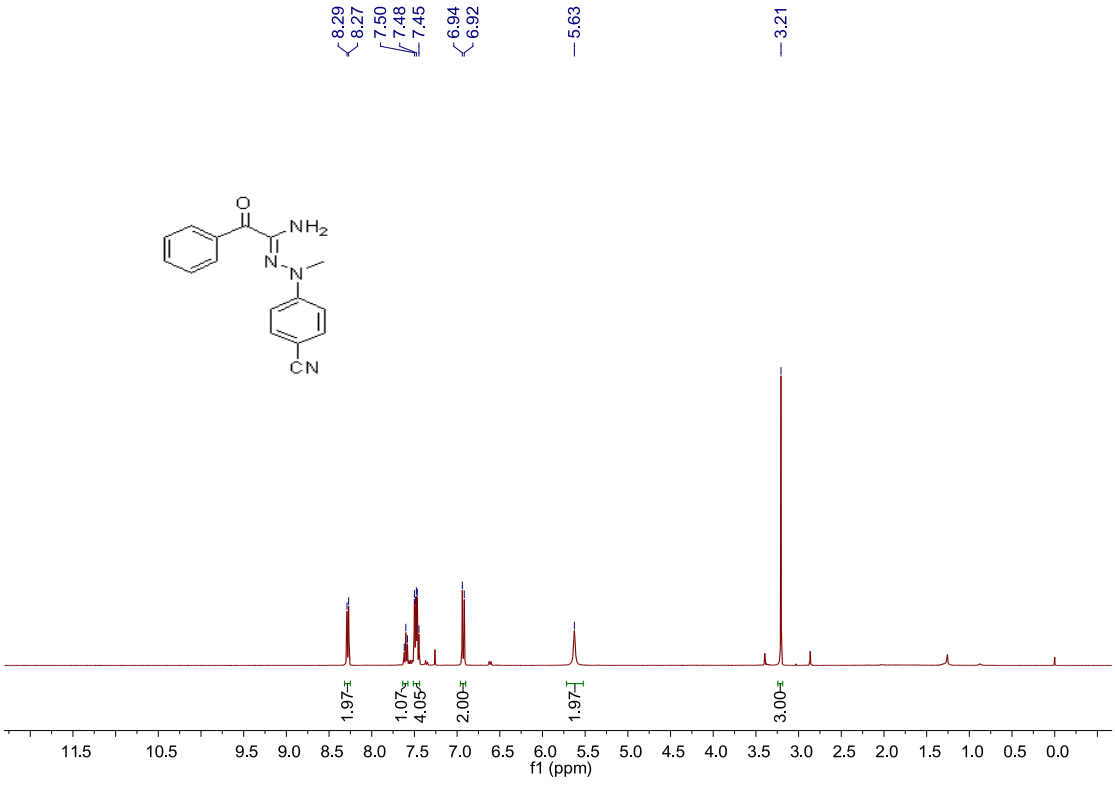
^{1}H NMR of product 40 in $CDCl_3$ (400 MHz)



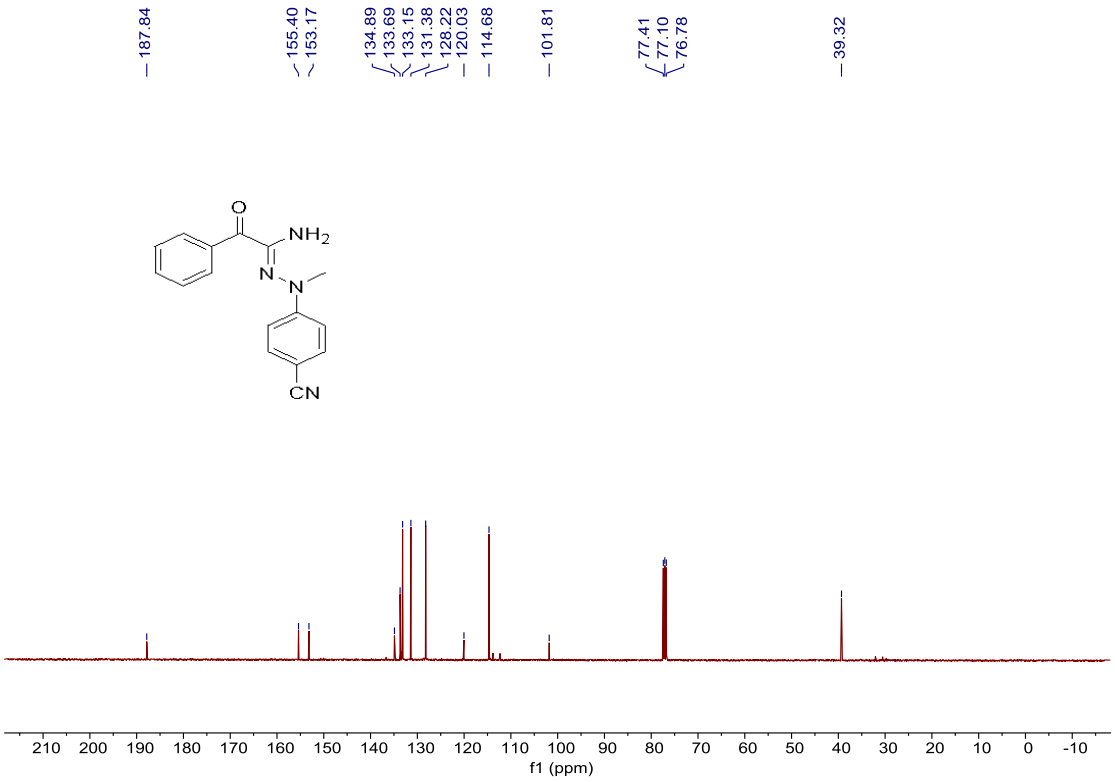
^{13}C NMR of product 40 in $CDCl_3$ (100 MHz)



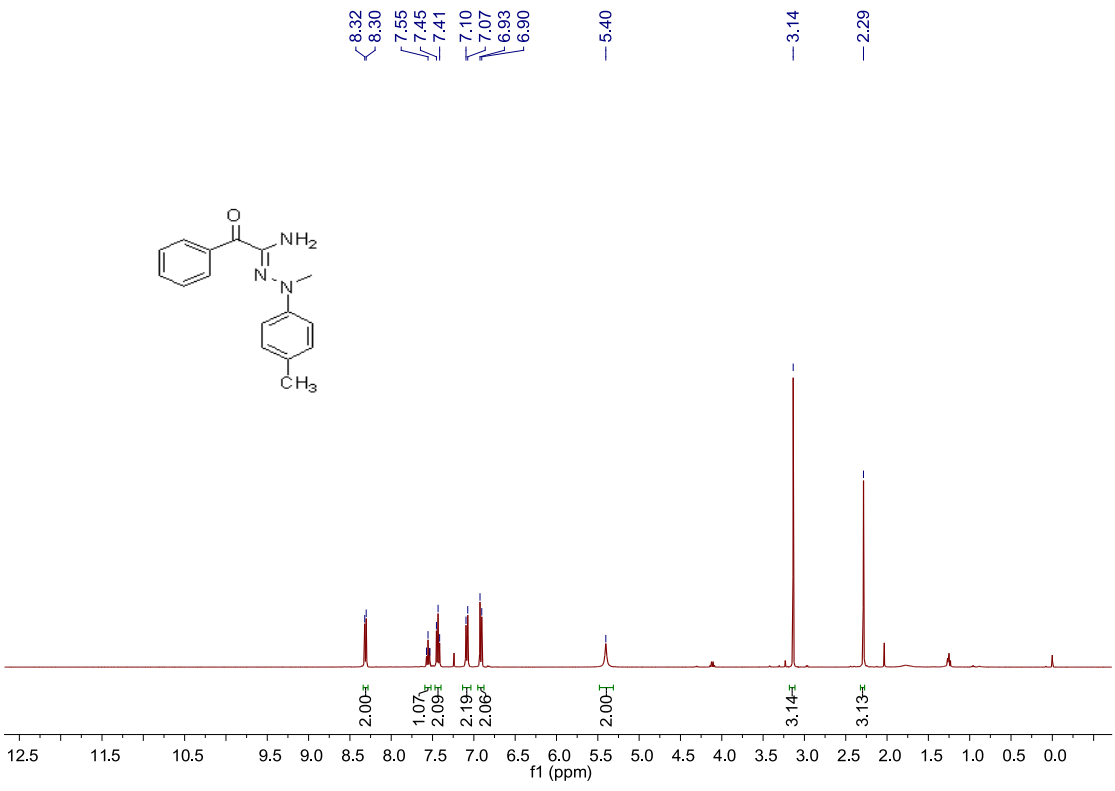
^{1}H NMR of product 41 in $CDCl_3$ (400 MHz)



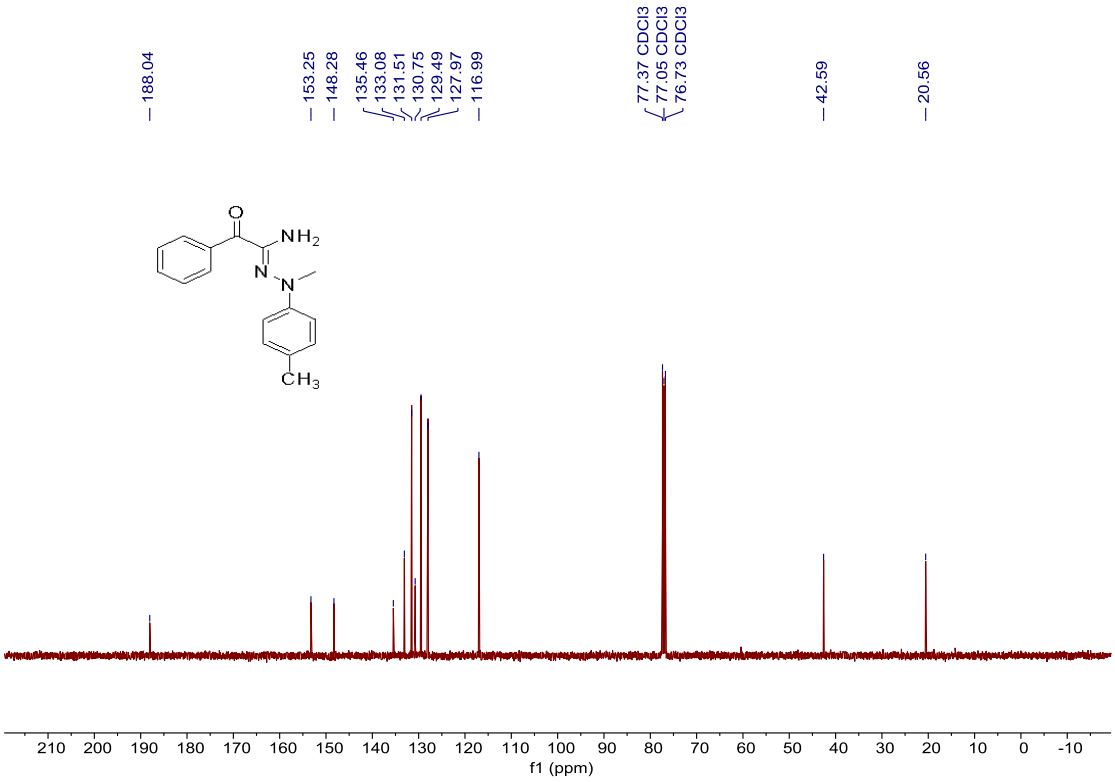
^{13}C NMR of product 41 in $CDCl_3$ (100 MHz)



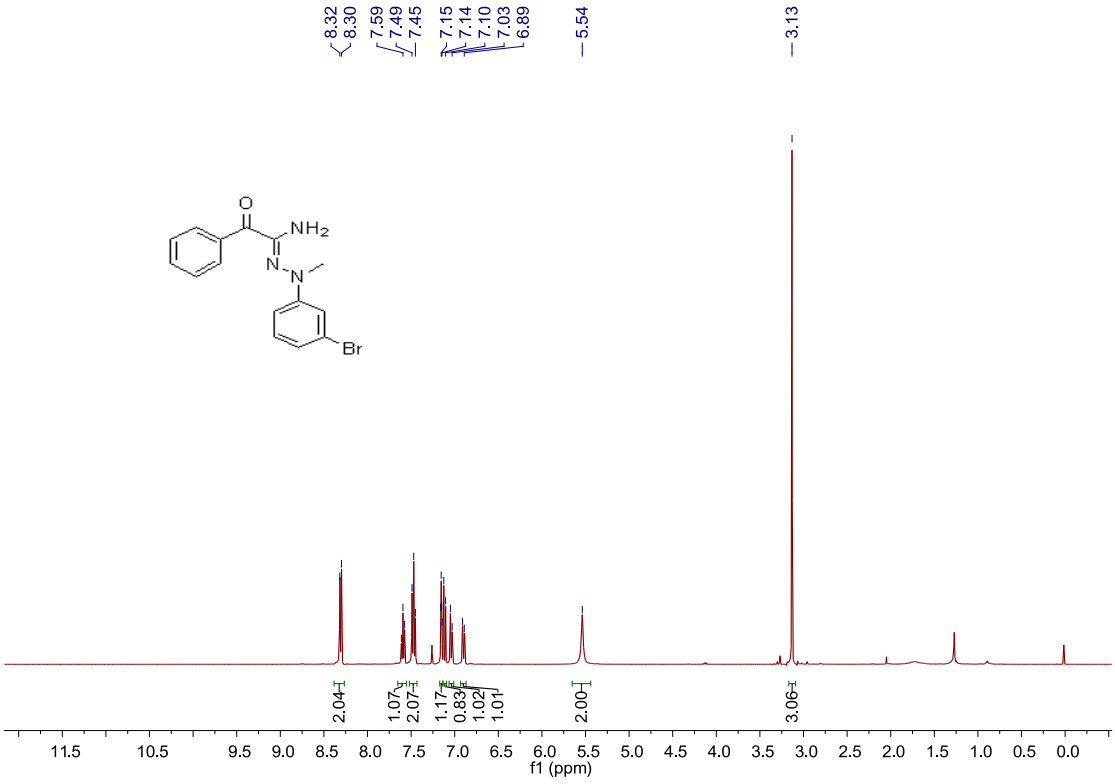
^{1}H NMR of product 42 in $CDCl_3$ (400 MHz)



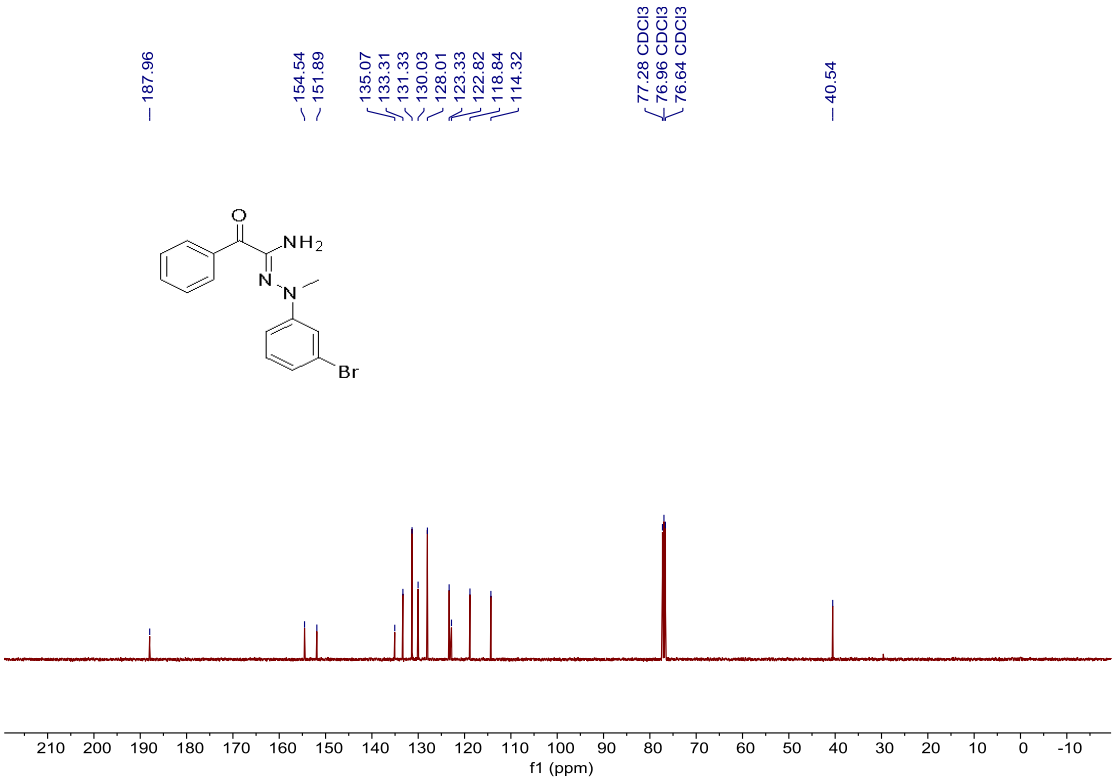
^{13}C NMR of product 42 in $CDCl_3$ (100 MHz)



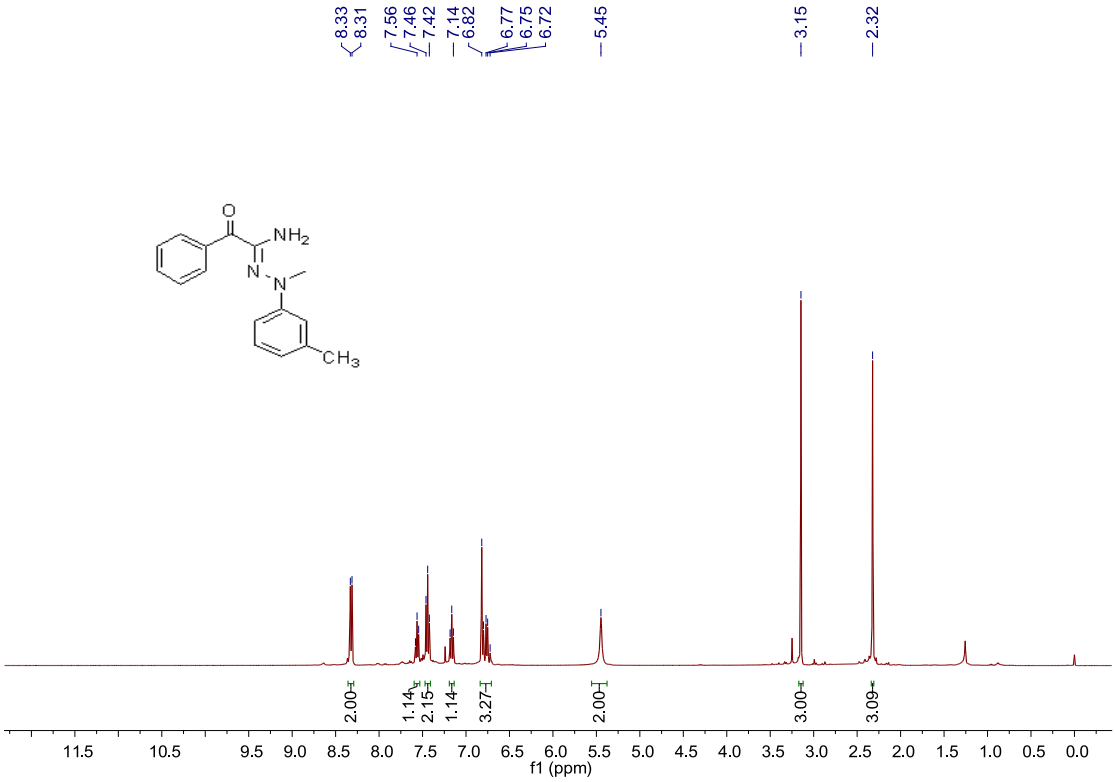
^{1}H NMR of product 43 in $CDCl_3$ (400 MHz)



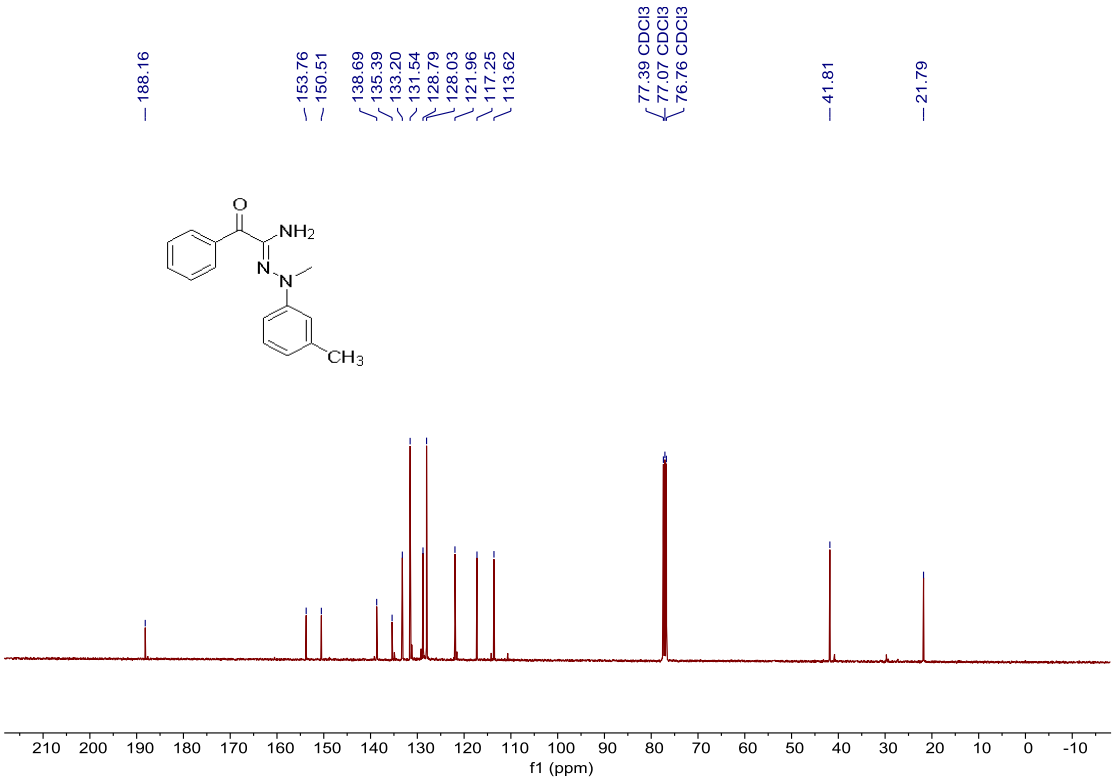
^{13}C NMR of product 43 in $CDCl_3$ (100 MHz)



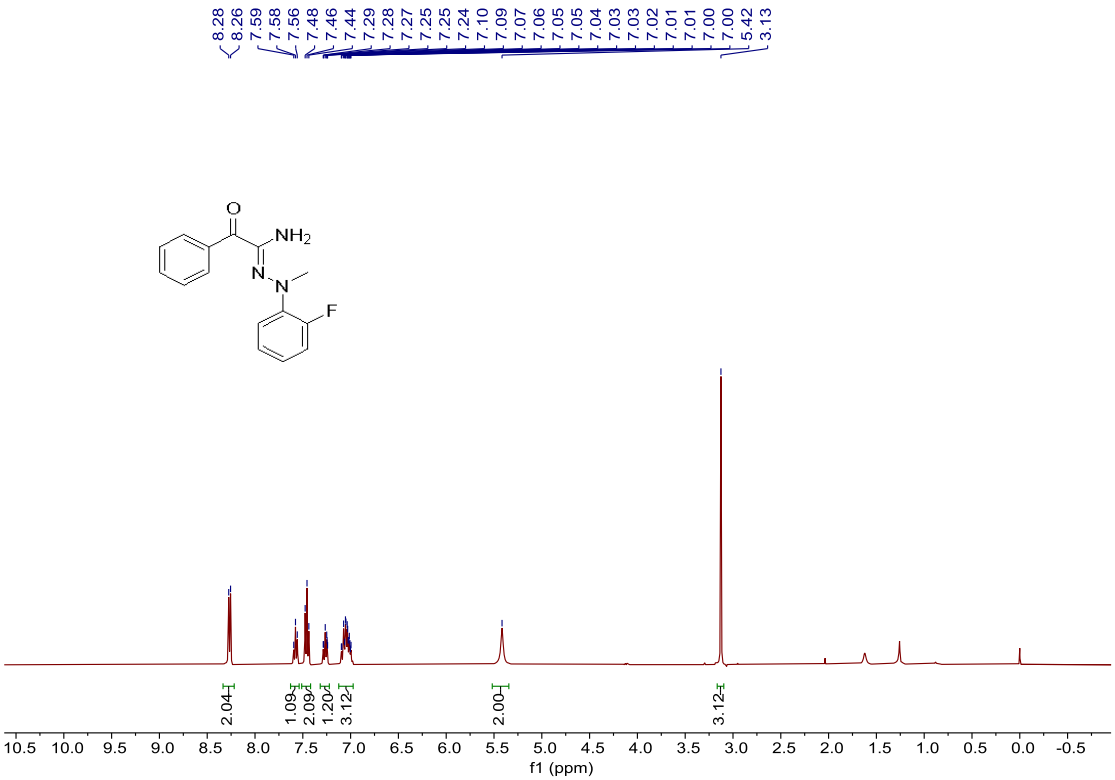
^{1}H NMR of product 44 in $CDCl_3$ (400 MHz)



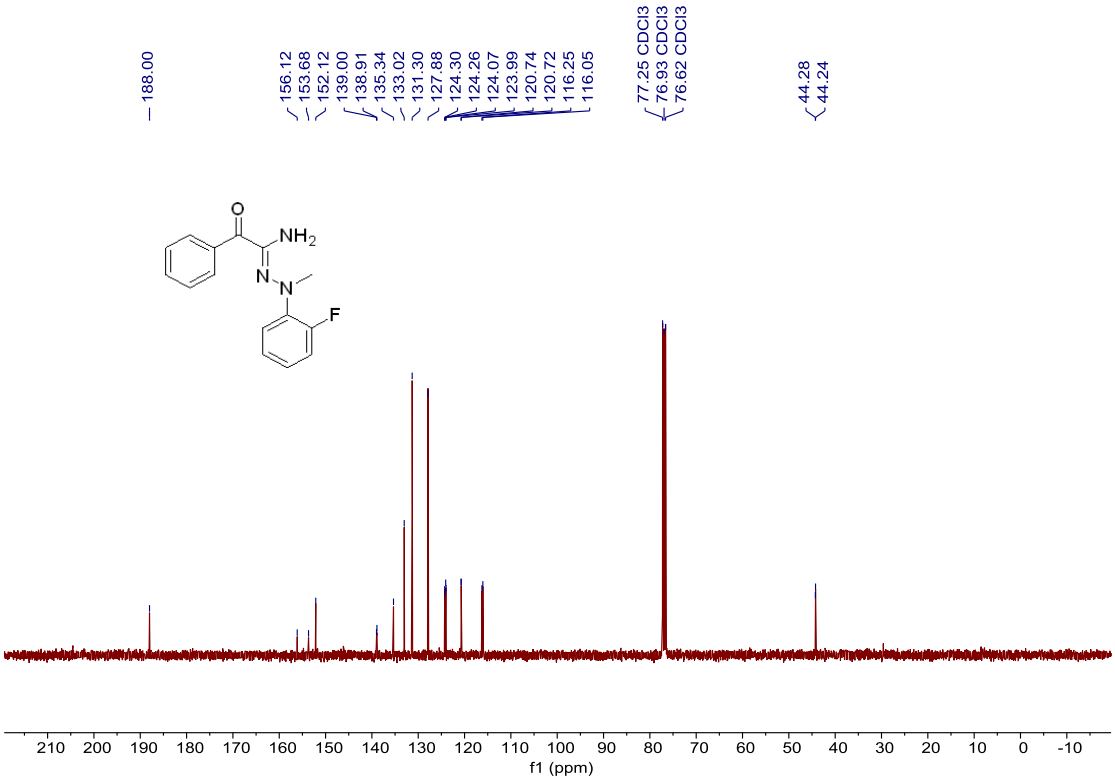
^{13}C NMR of product 44 in $CDCl_3$ (100 MHz)



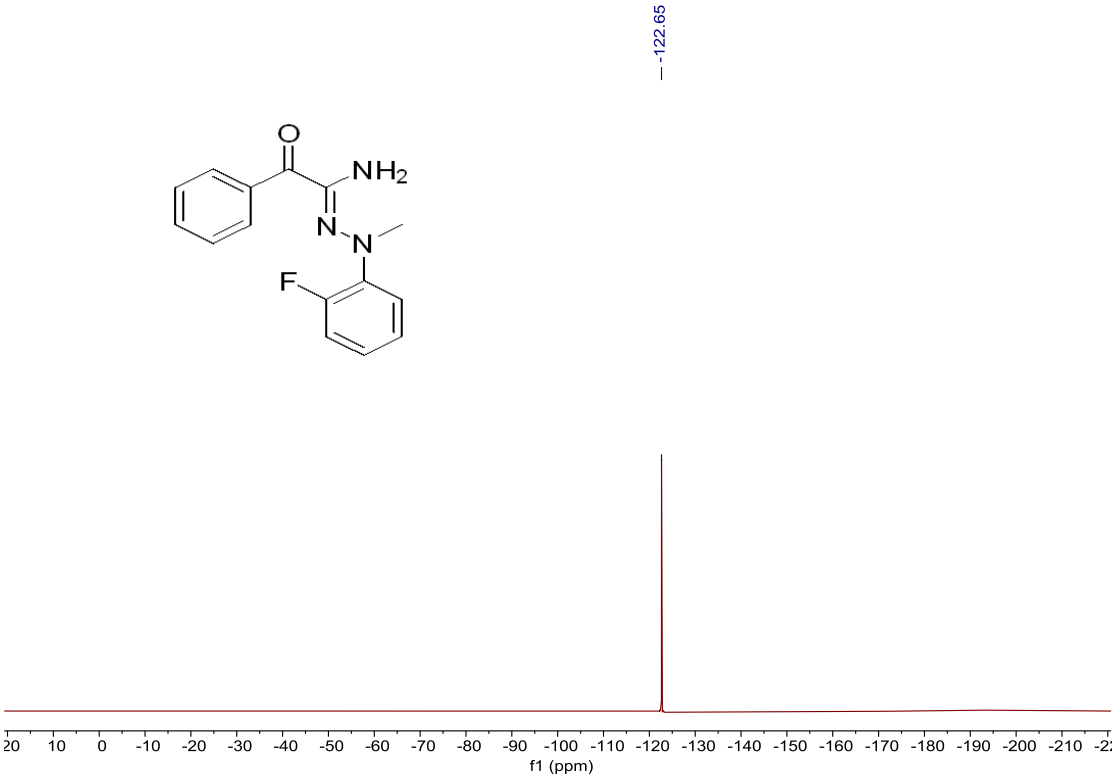
^{1}H NMR of product 45 in $CDCl_3$ (400 MHz)



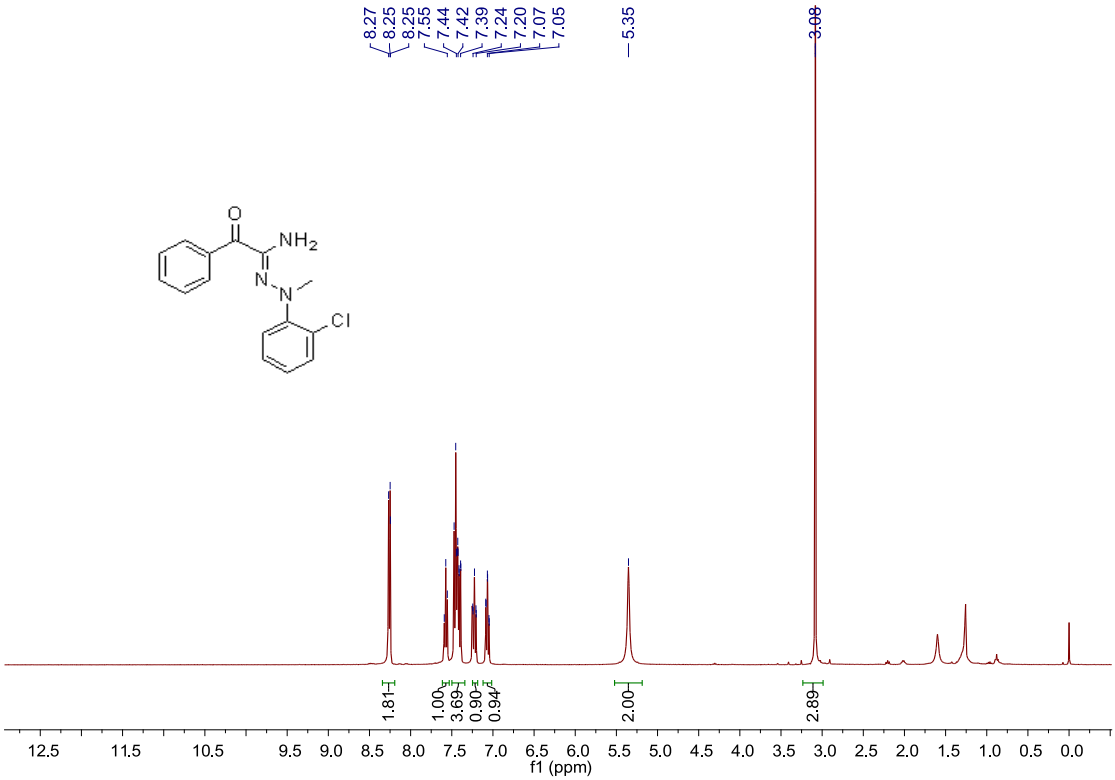
^{13}C NMR of product 45 in $CDCl_3$ (100 MHz)



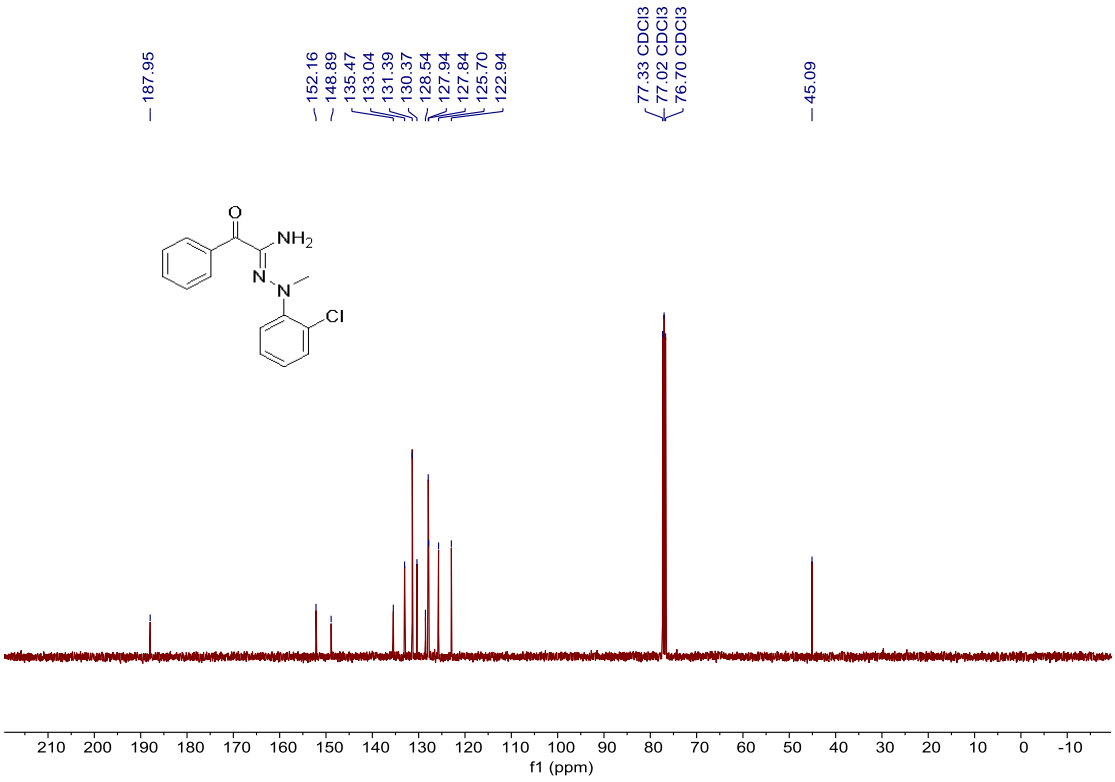
^{19}F NMR of product 45 in $CDCl_3$ (100 MHz)



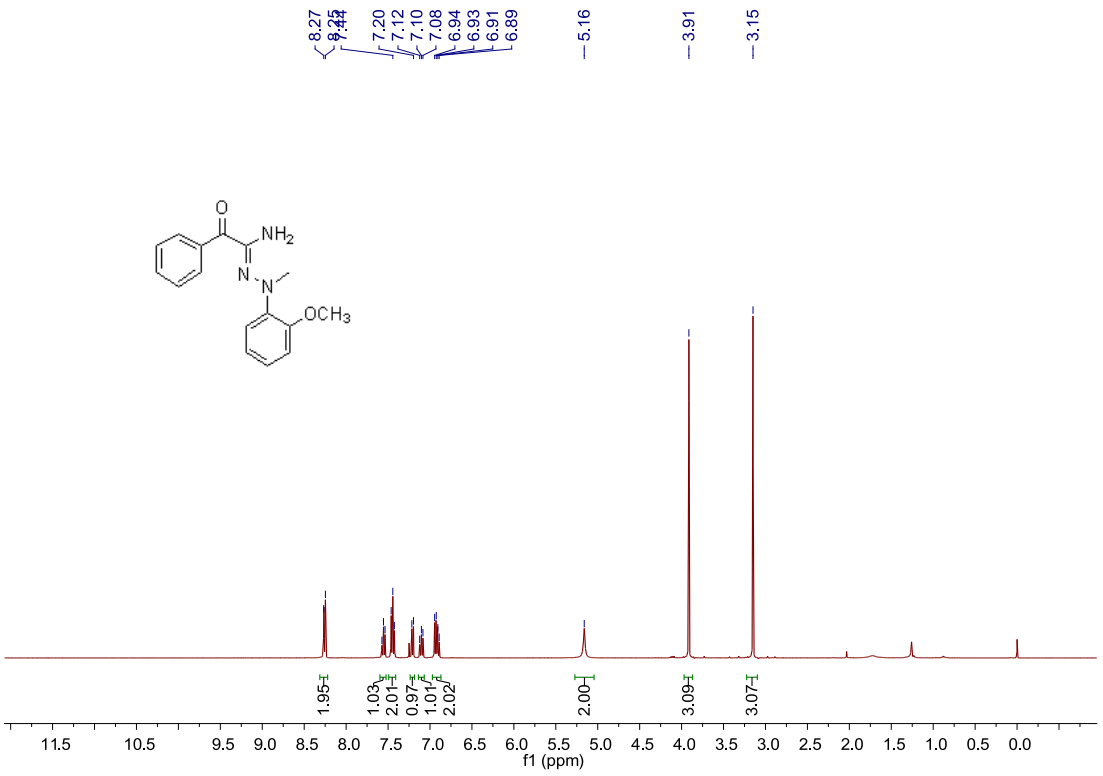
^{1}H NMR of product 46 in $CDCl_3$ (400 MHz)



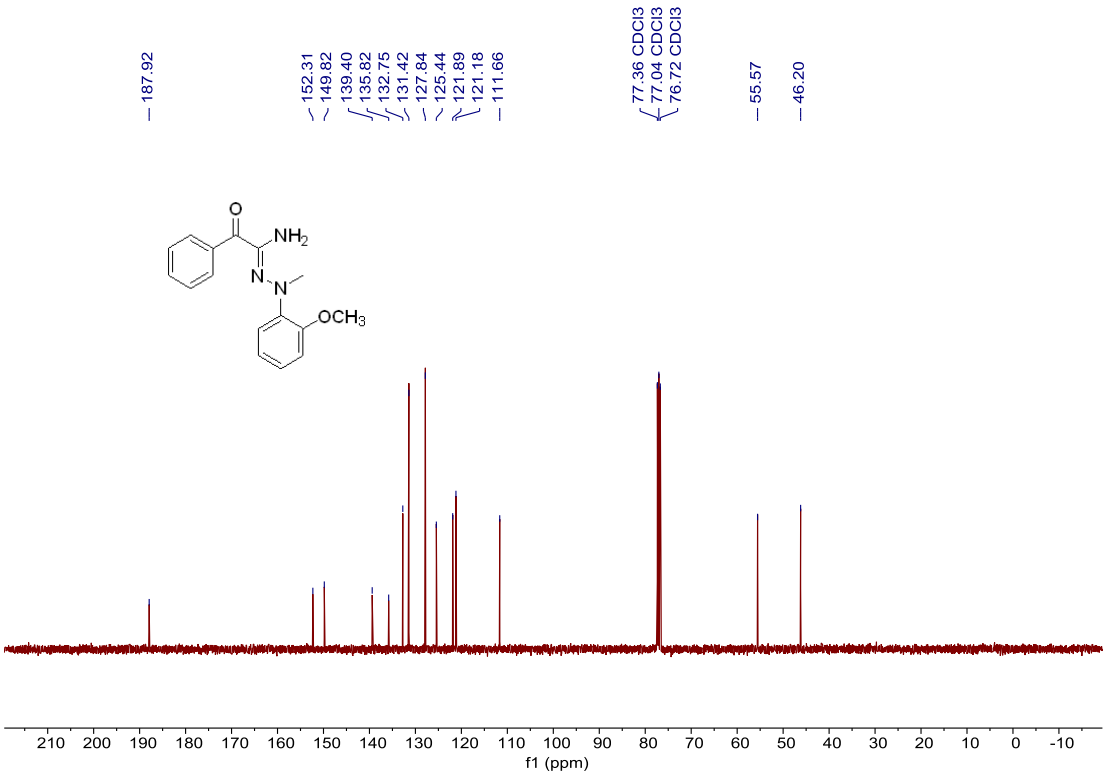
^{13}C NMR of product 46 in $CDCl_3$ (100 MHz)



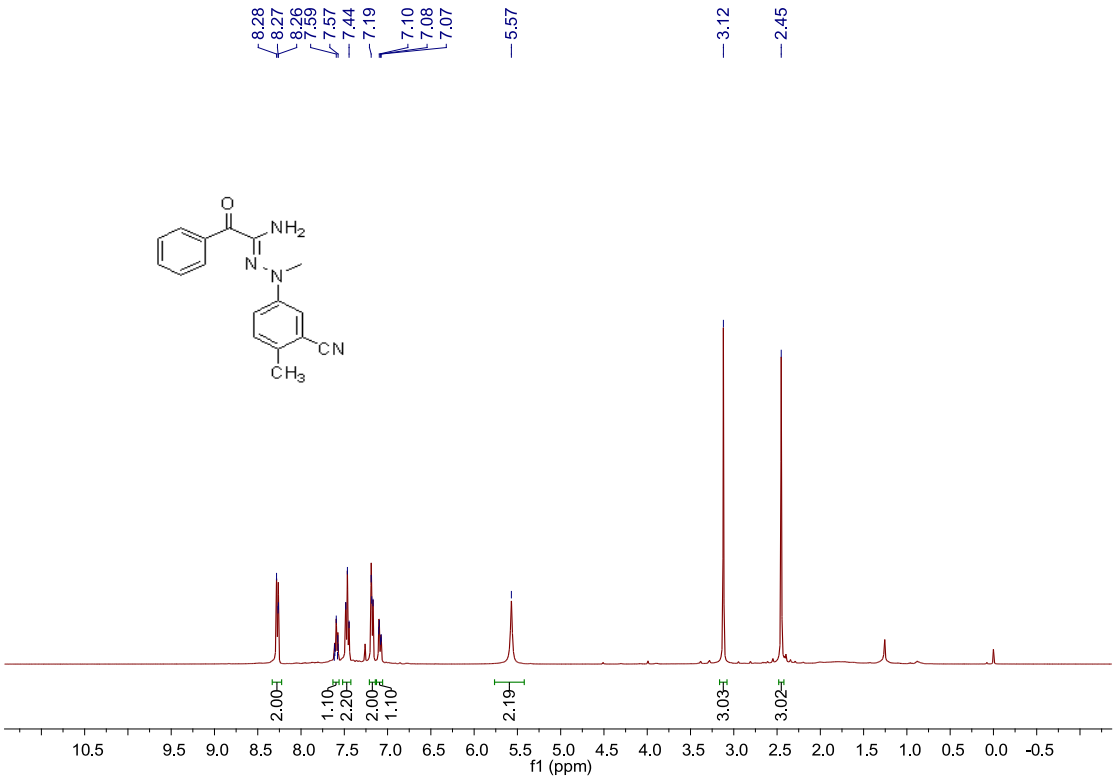
^{1}H NMR of product 47 in $CDCl_3$ (400 MHz)



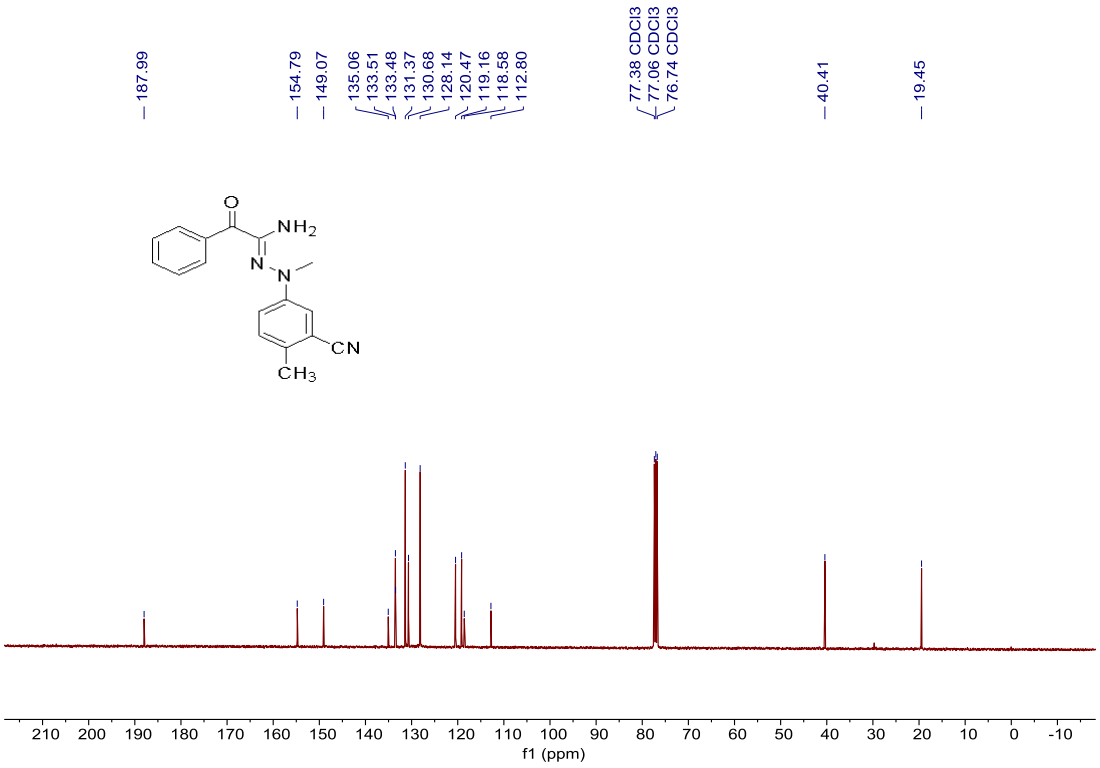
^{13}C NMR of product 47 in $CDCl_3$ (100 MHz)



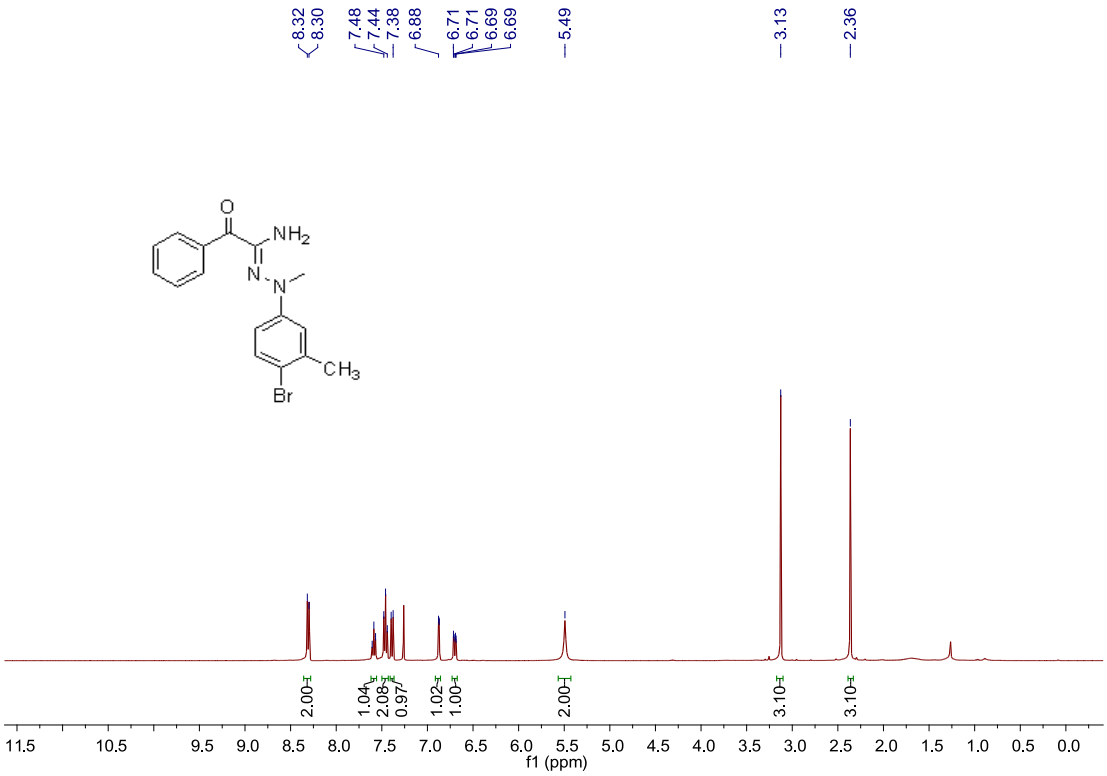
^{1}H NMR of product 48 in $CDCl_3$ (400 MHz)



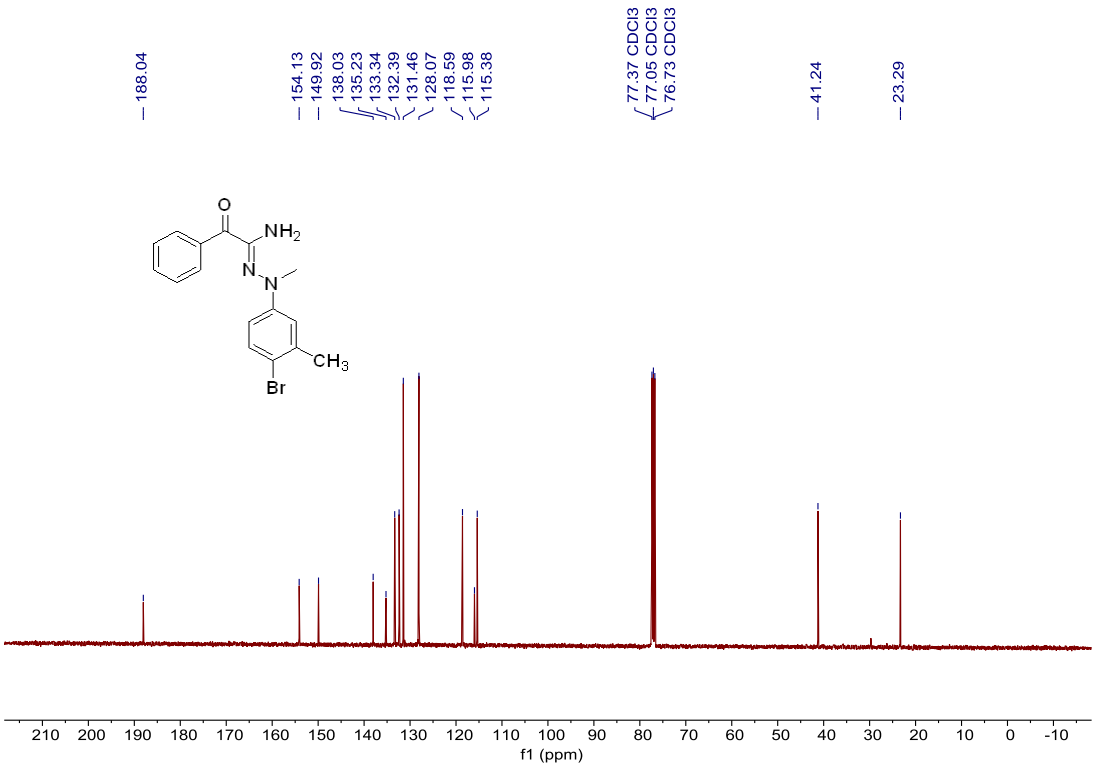
^{13}C NMR of product 48 in $CDCl_3$ (100 MHz)



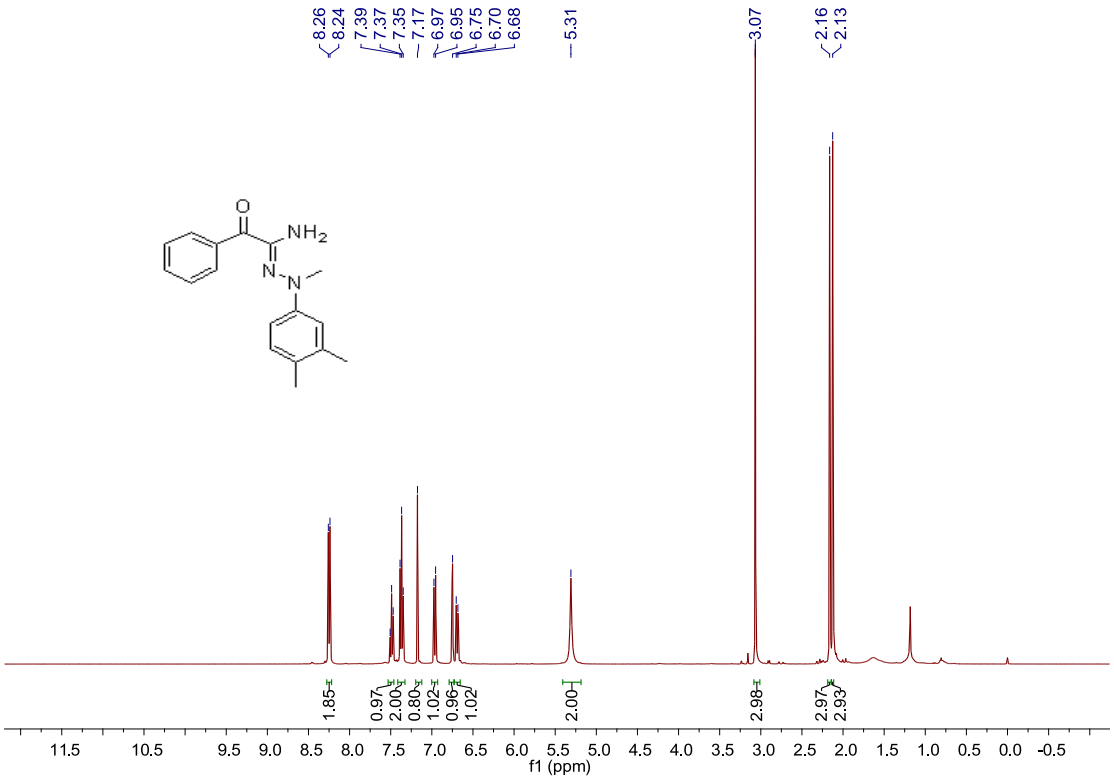
^{1}H NMR of product 49 in $CDCl_3$ (400 MHz)



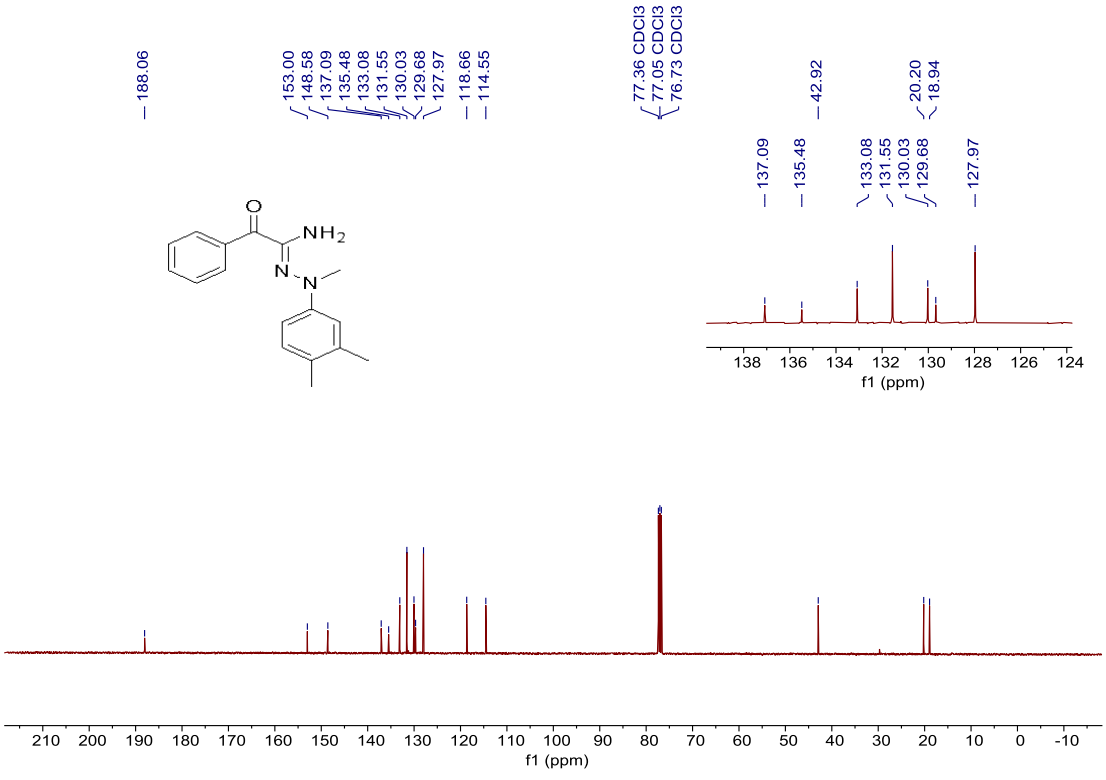
^{13}C NMR of product 49 in $CDCl_3$ (100 MHz)



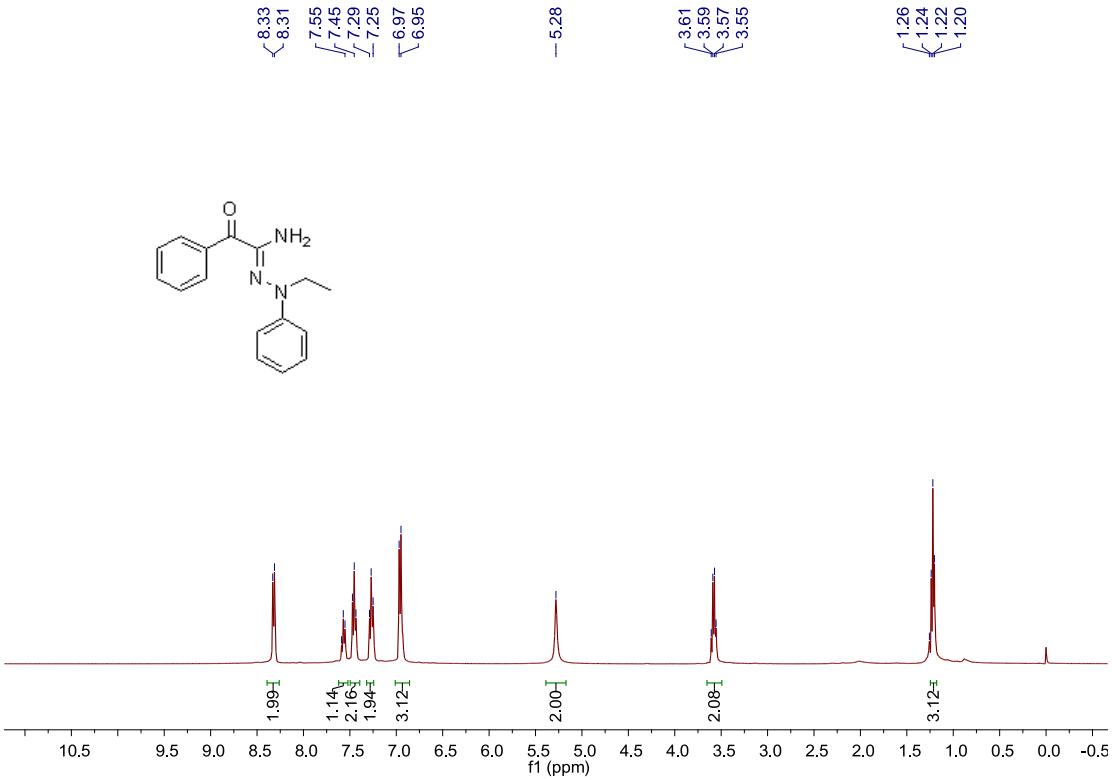
^{1}H NMR of product 50 in $CDCl_3$ (400 MHz)



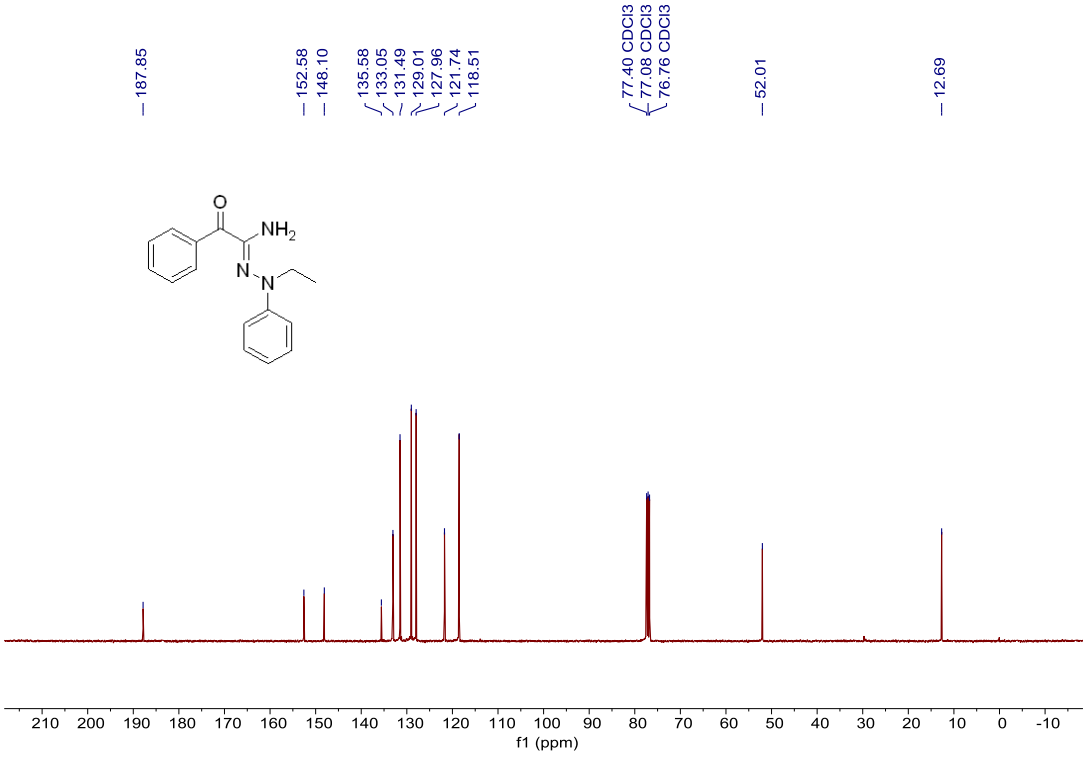
^{13}C NMR of product 50 in $CDCl_3$ (100 MHz)



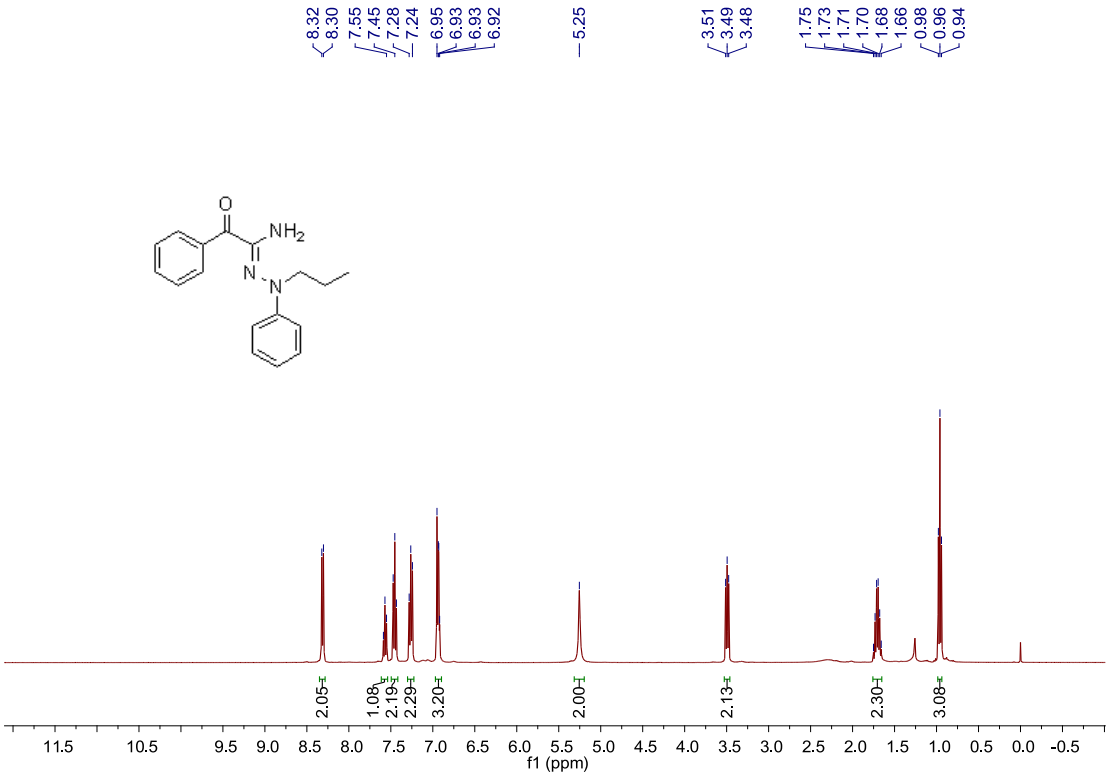
^{1}H NMR of product 51 in $CDCl_3$ (400 MHz)



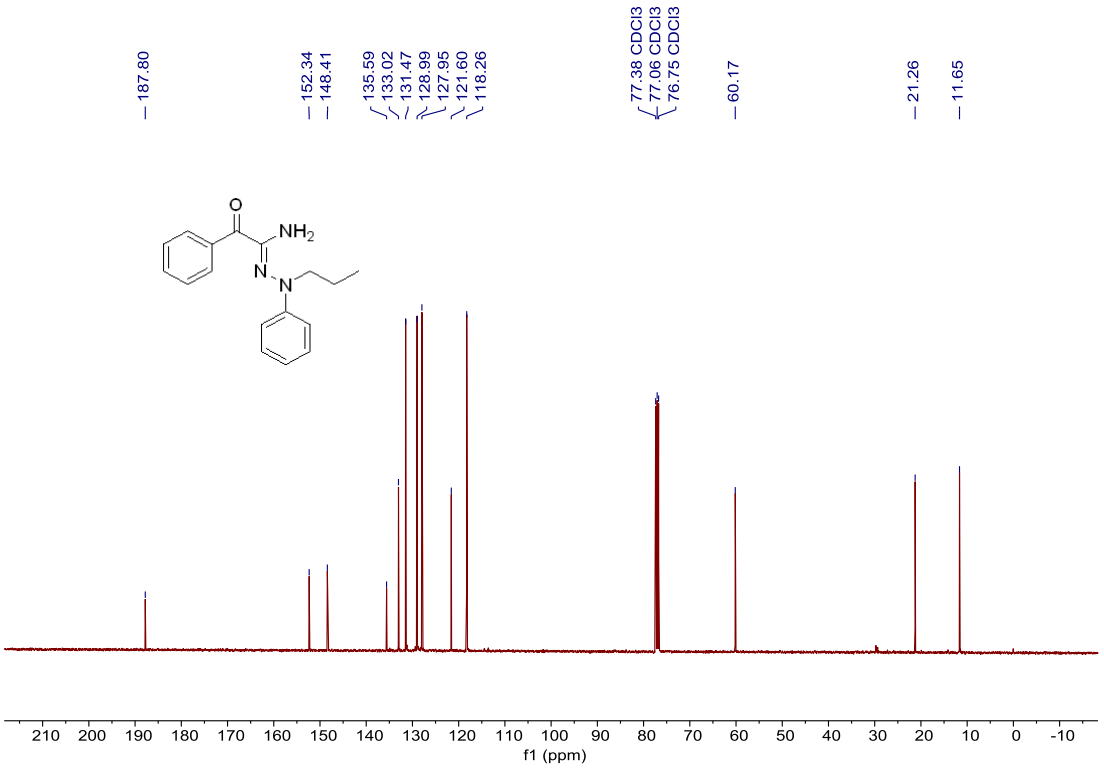
^{13}C NMR of product 51 in $CDCl_3$ (100 MHz)



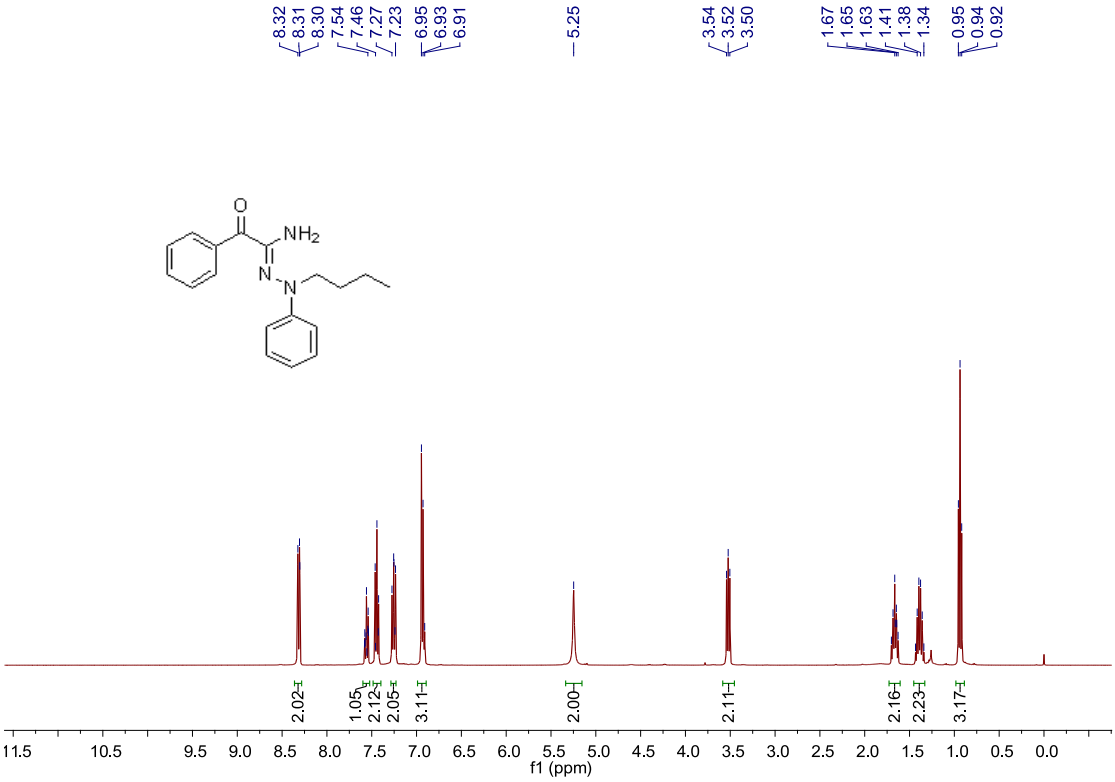
^{1}H NMR of product 52 in $CDCl_3$ (400 MHz)



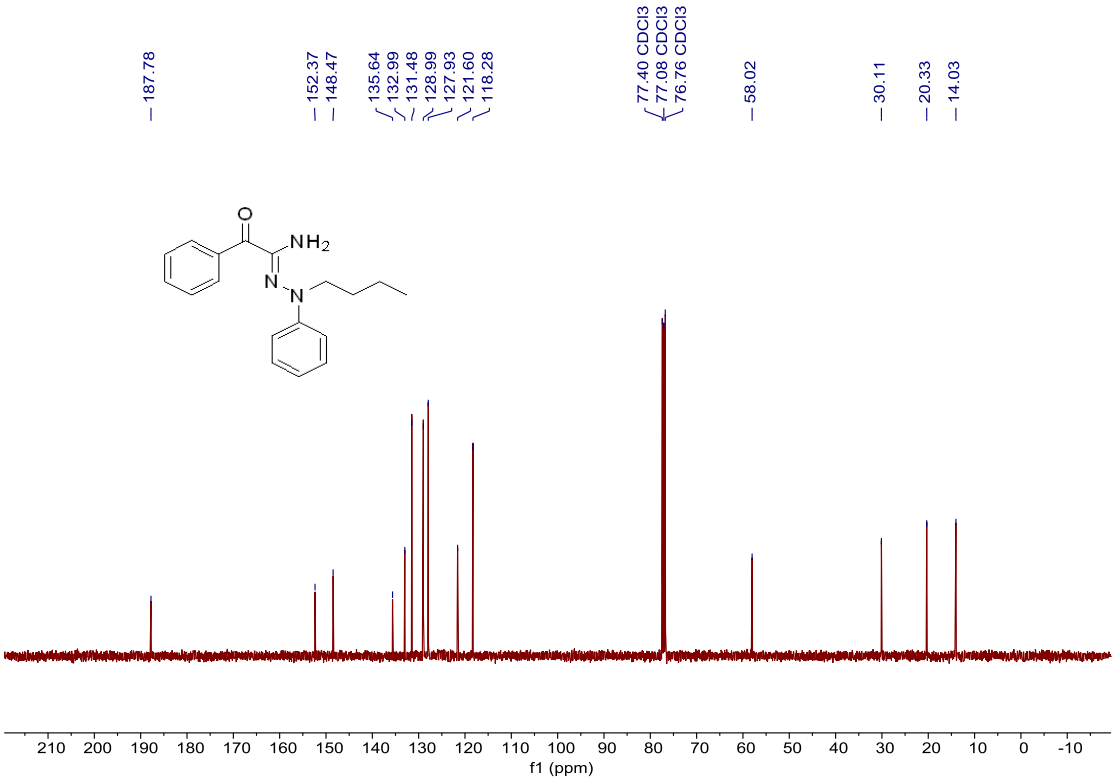
^{13}C NMR of product 52 in $CDCl_3$ (100 MHz)



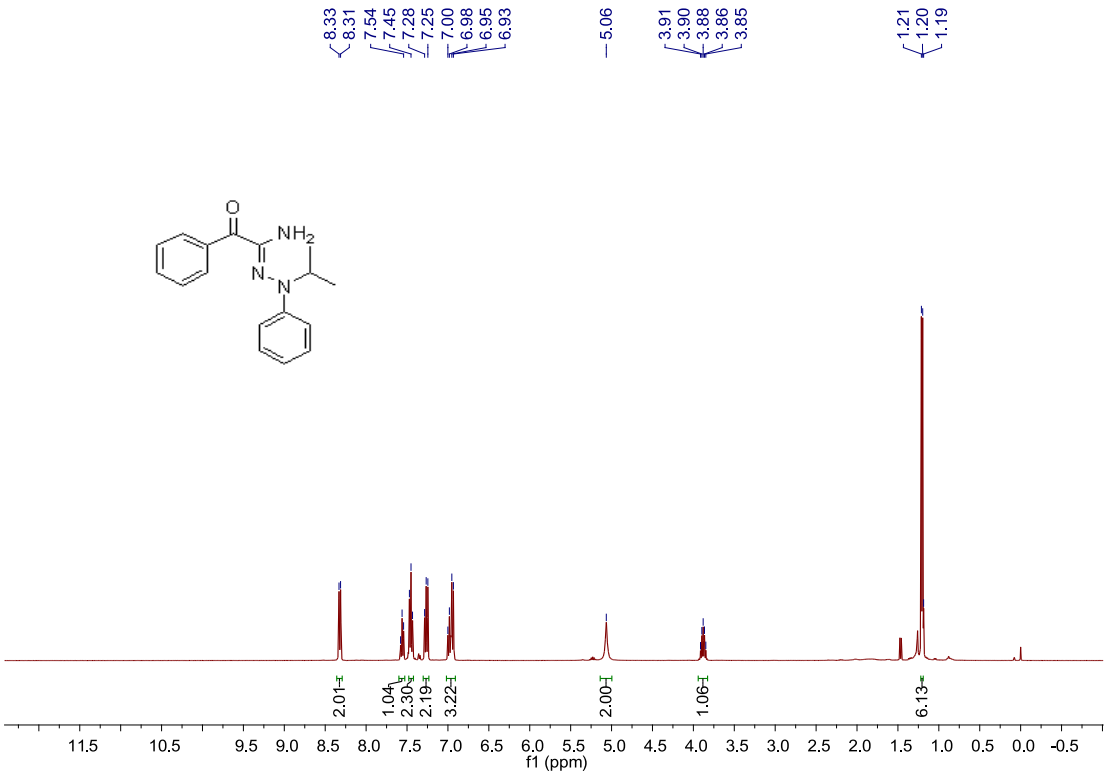
^{1}H NMR of product 53 in $CDCl_3$ (400 MHz)



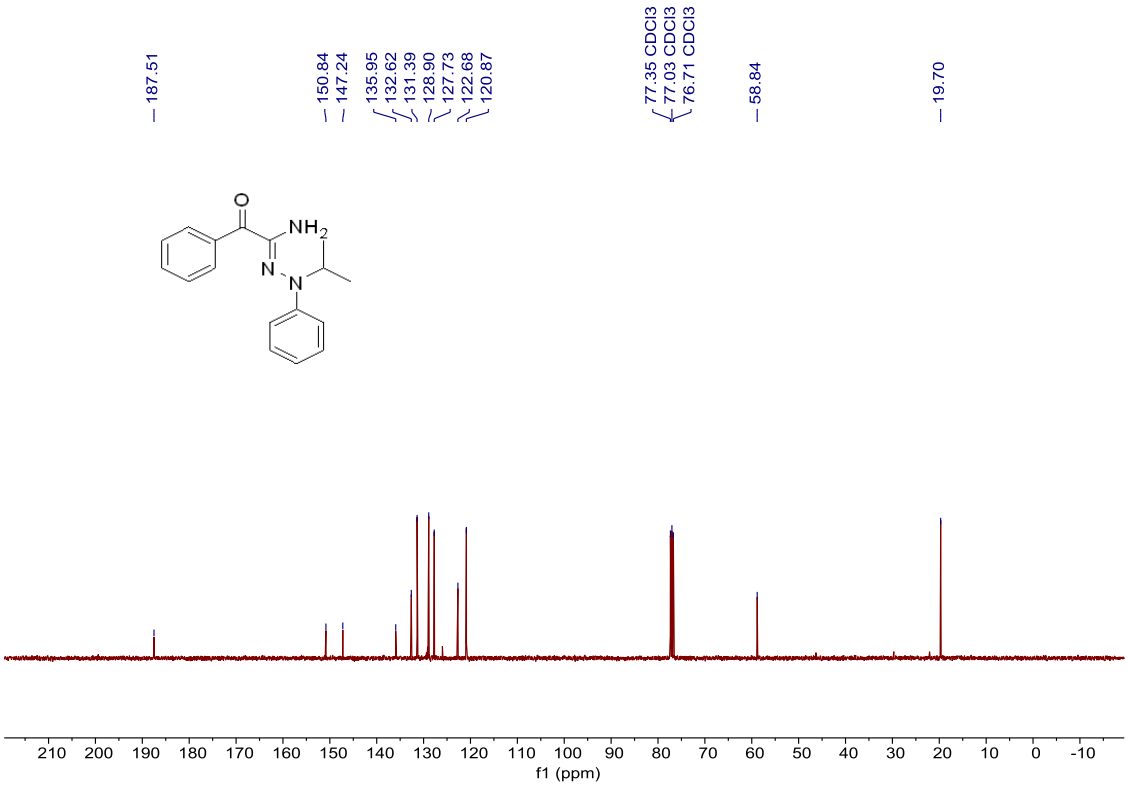
^{13}C NMR of product 53 in $CDCl_3$ (100 MHz)



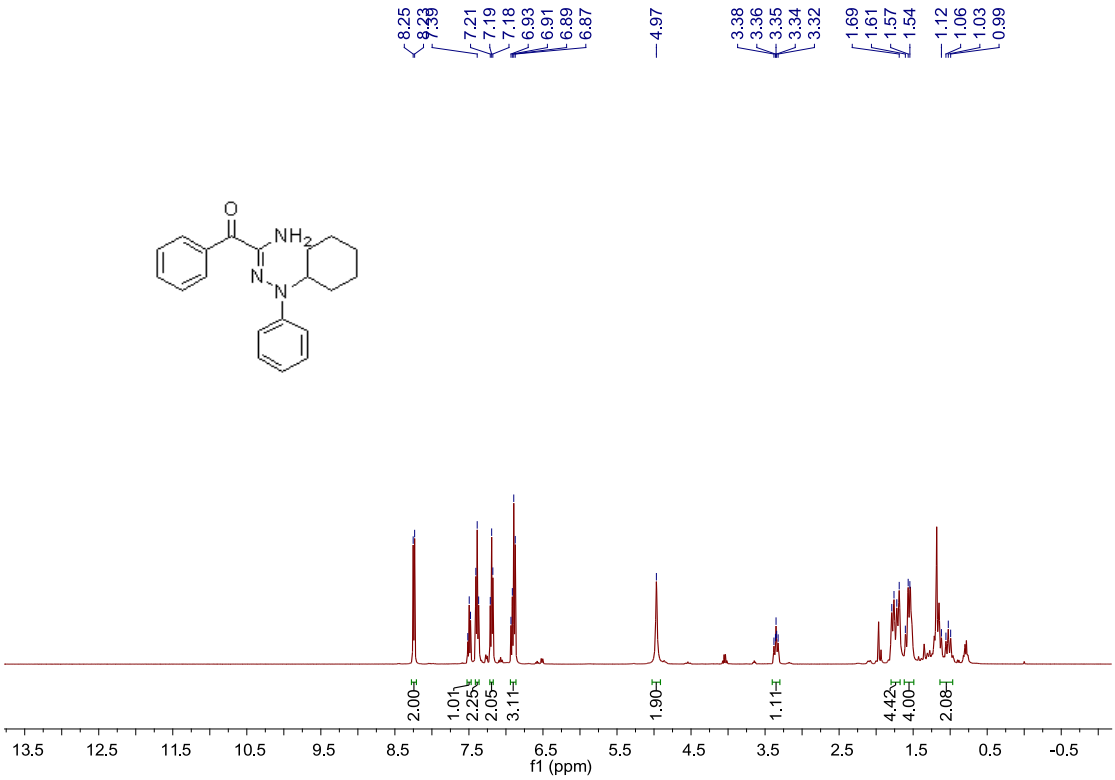
^{1}H NMR of product 54 in $CDCl_3$ (400 MHz)



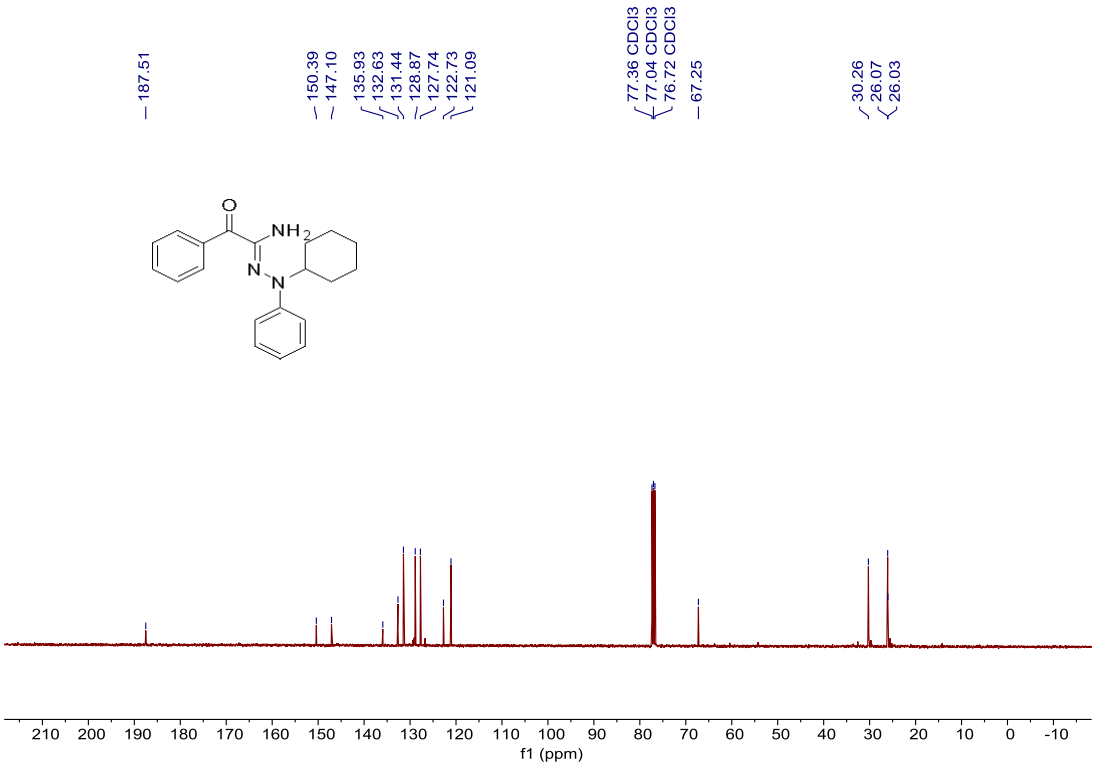
^{13}C NMR of product 54 in $CDCl_3$ (100 MHz)



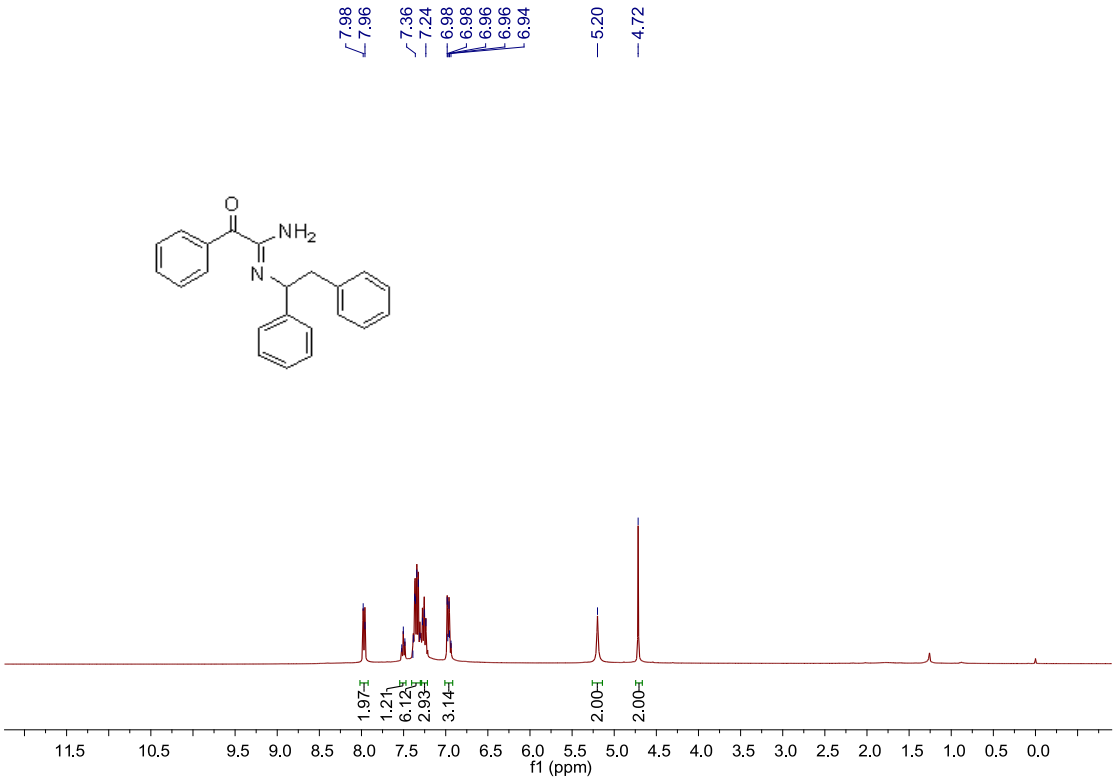
^{1}H NMR of product 55 in $CDCl_3$ (400 MHz)



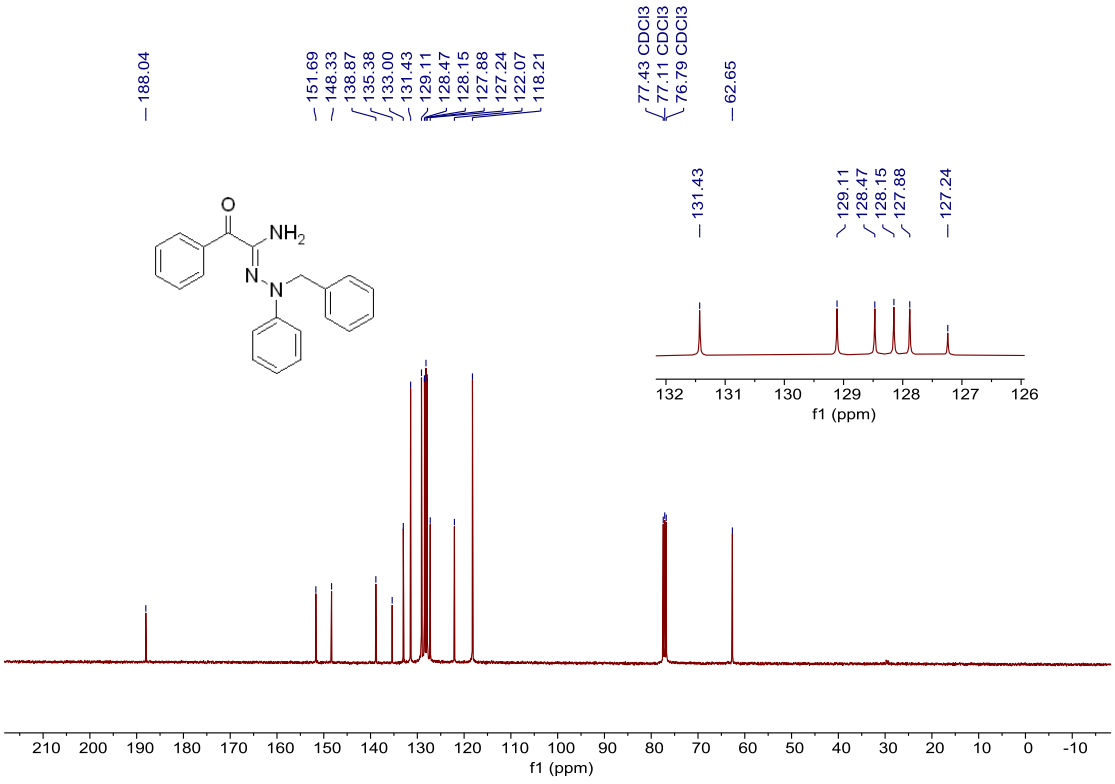
^{13}C NMR of product 55 in $CDCl_3$ (100 MHz)



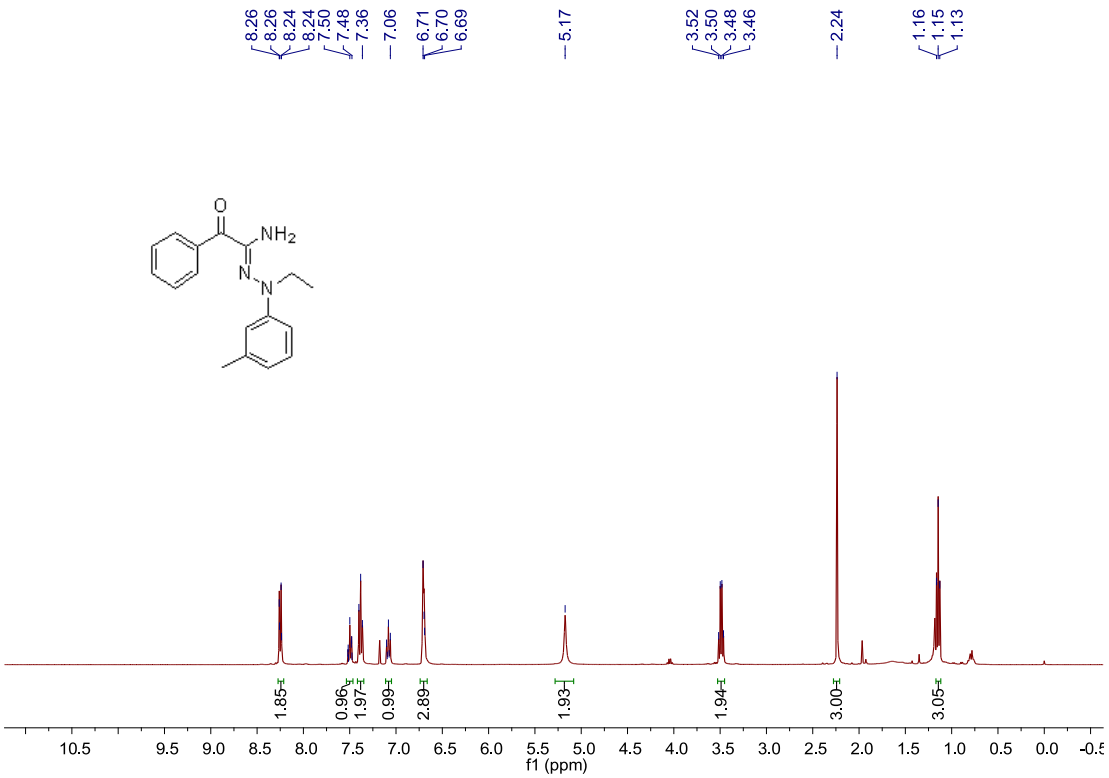
^{1}H NMR of product 56 in $CDCl_3$ (400 MHz)



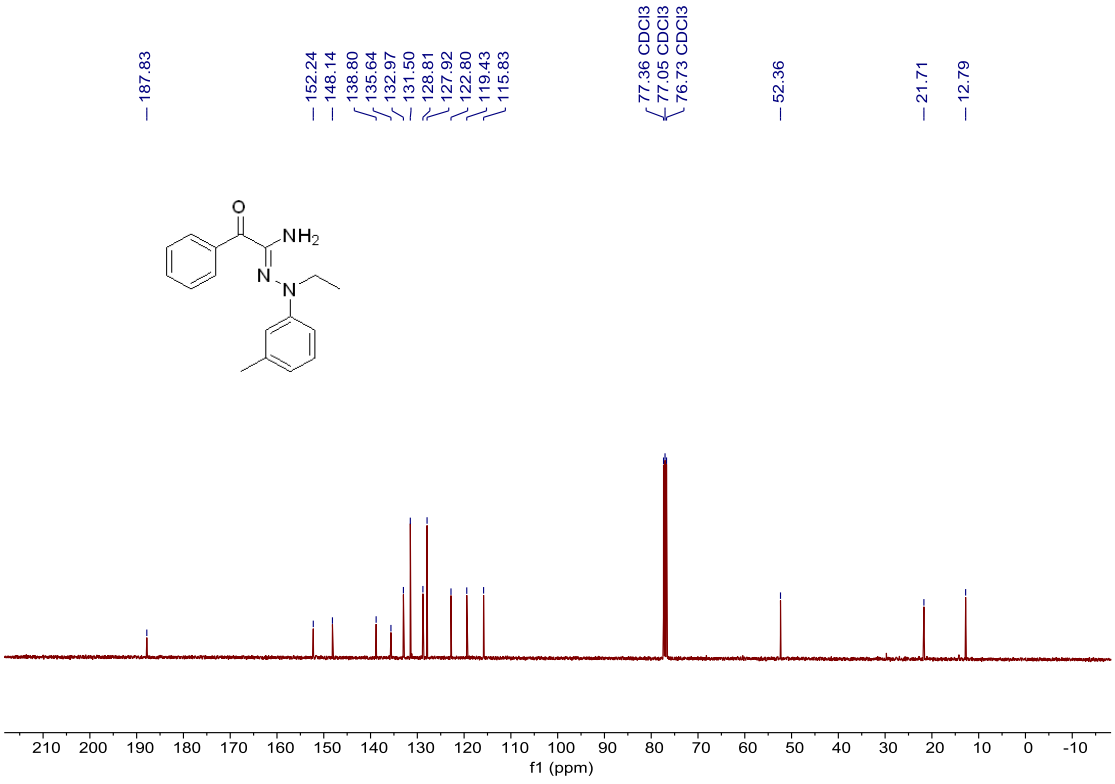
^{13}C NMR of product 56 in $CDCl_3$ (100 MHz)



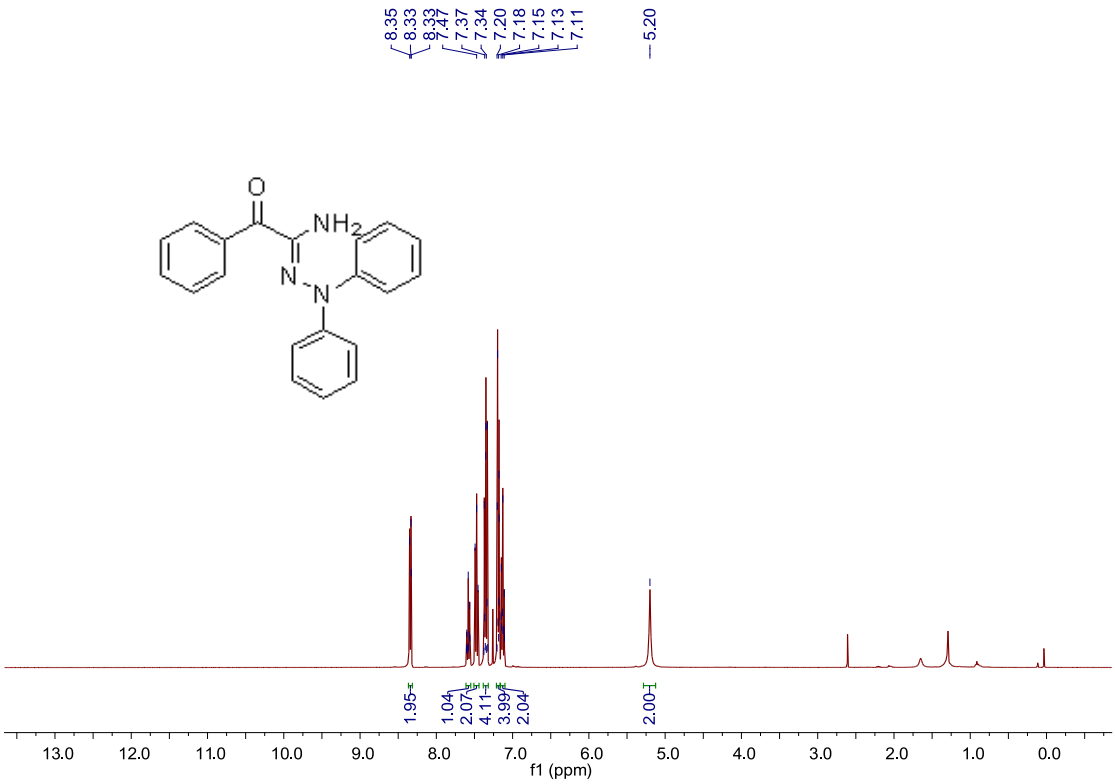
^{1}H NMR of product 57 in $CDCl_3$ (400 MHz)



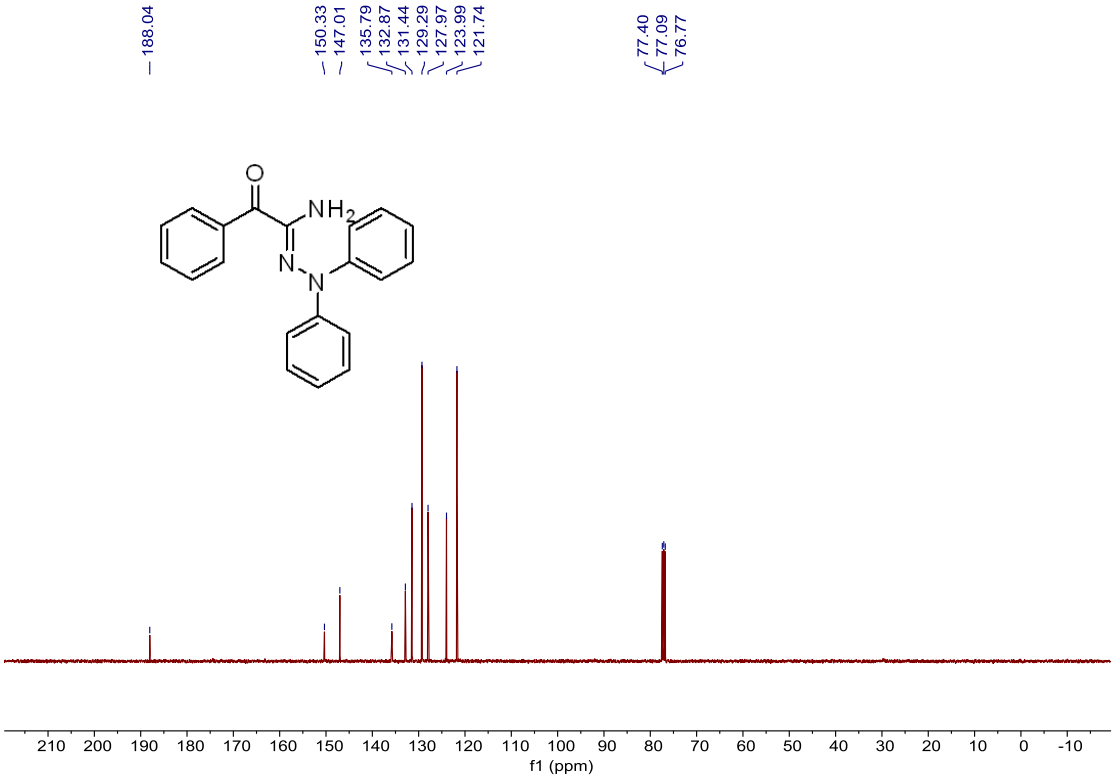
^{13}C NMR of product 57 in $CDCl_3$ (100 MHz)



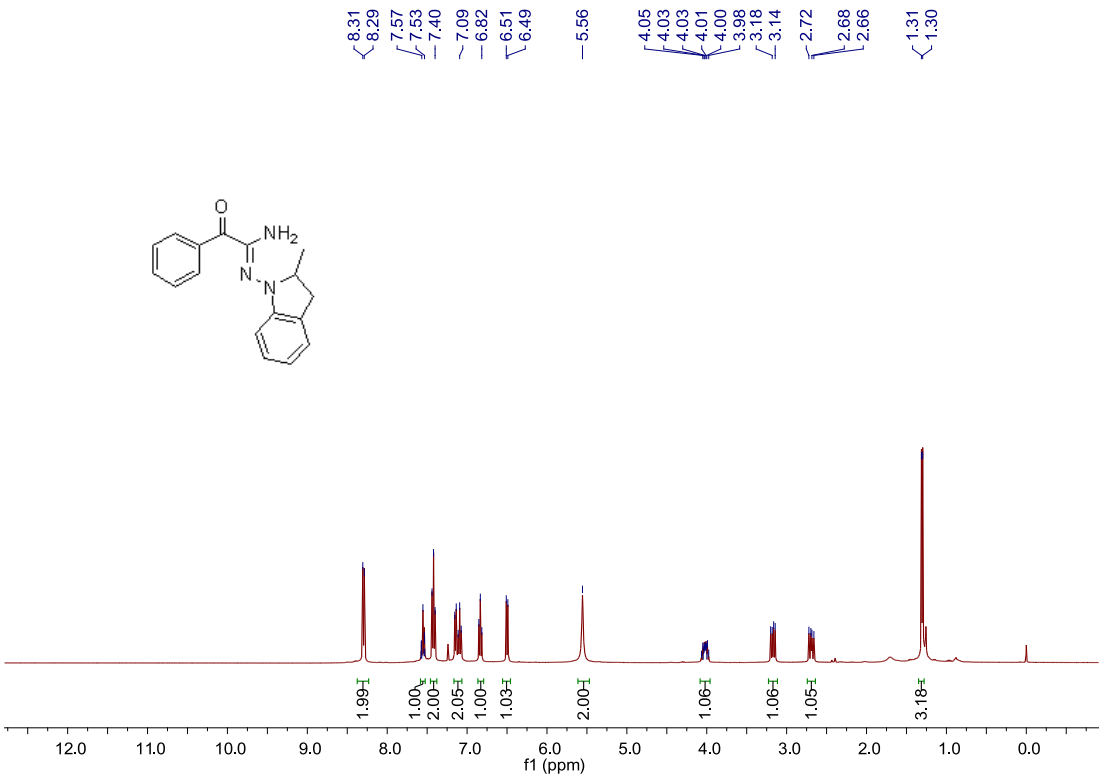
^{1}H NMR of product 58 in $CDCl_3$ (400 MHz)



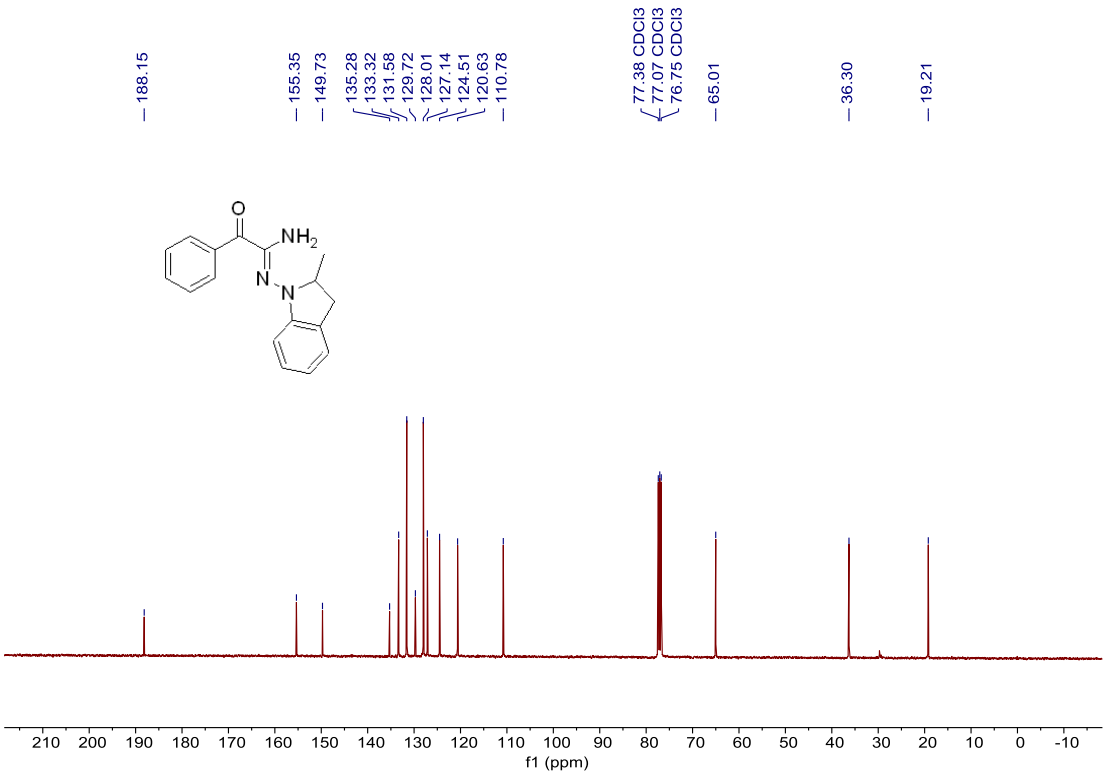
^{13}C NMR of product 58 in $CDCl_3$ (100 MHz)



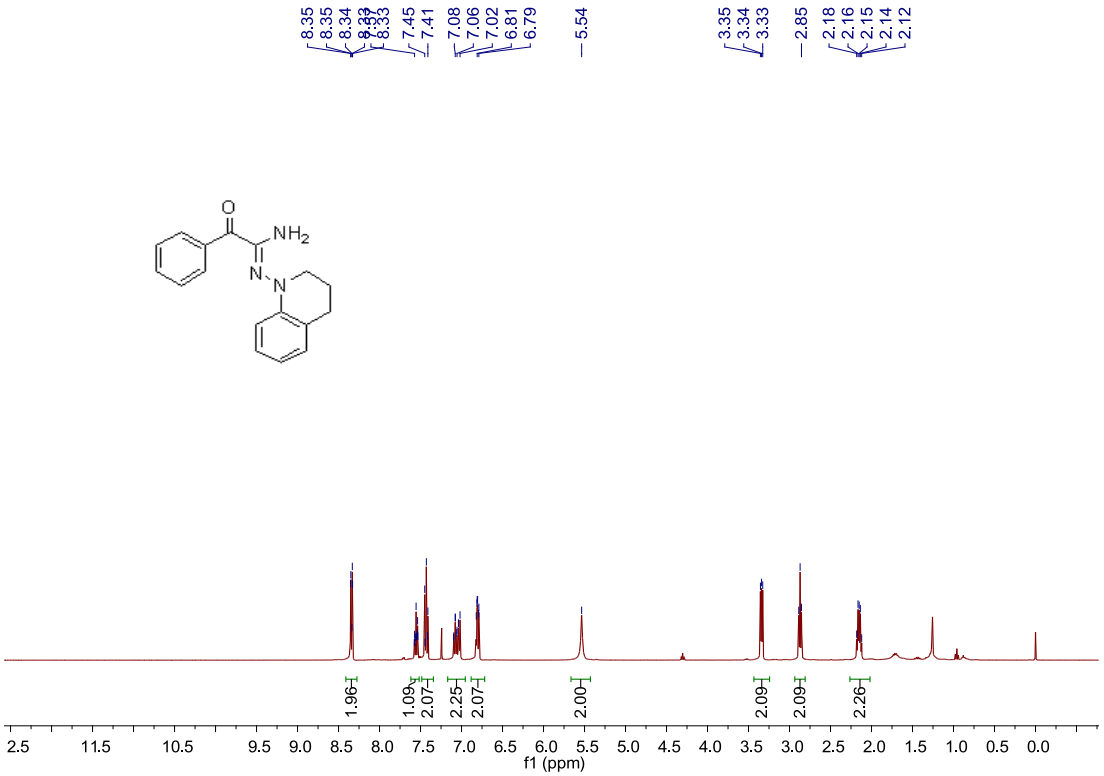
^{1}H NMR of product 59 in $CDCl_3$ (400 MHz)



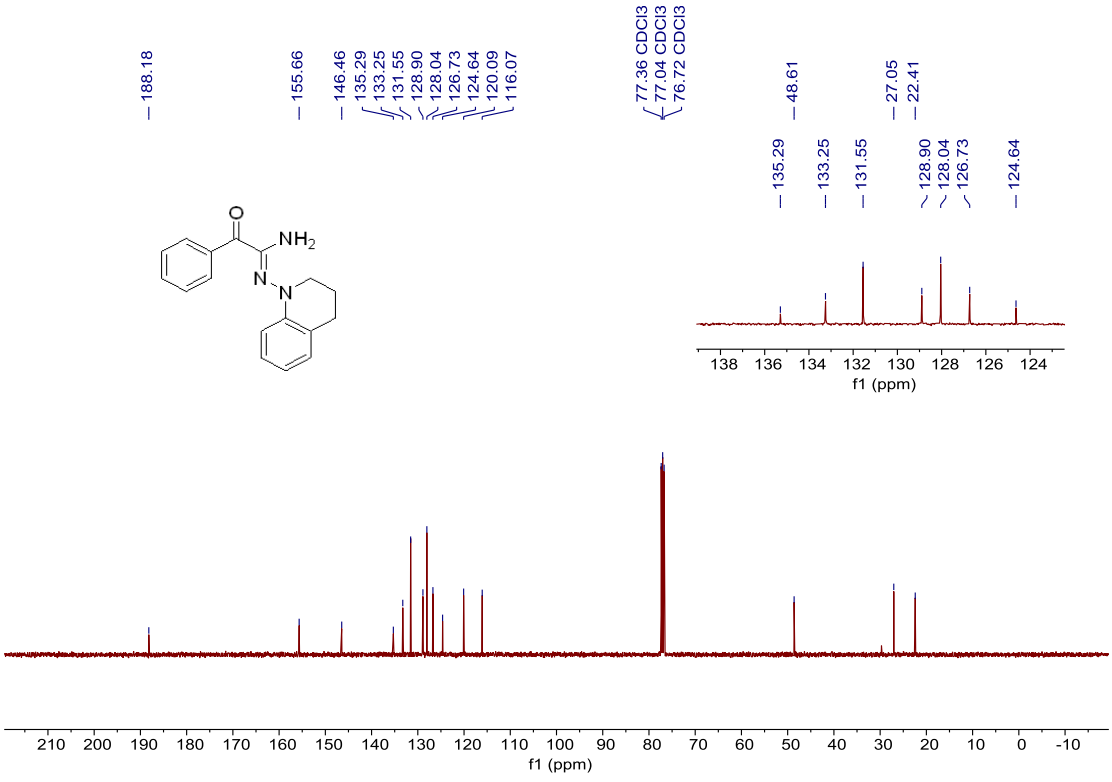
^{13}C NMR of product 59 in $CDCl_3$ (100 MHz)



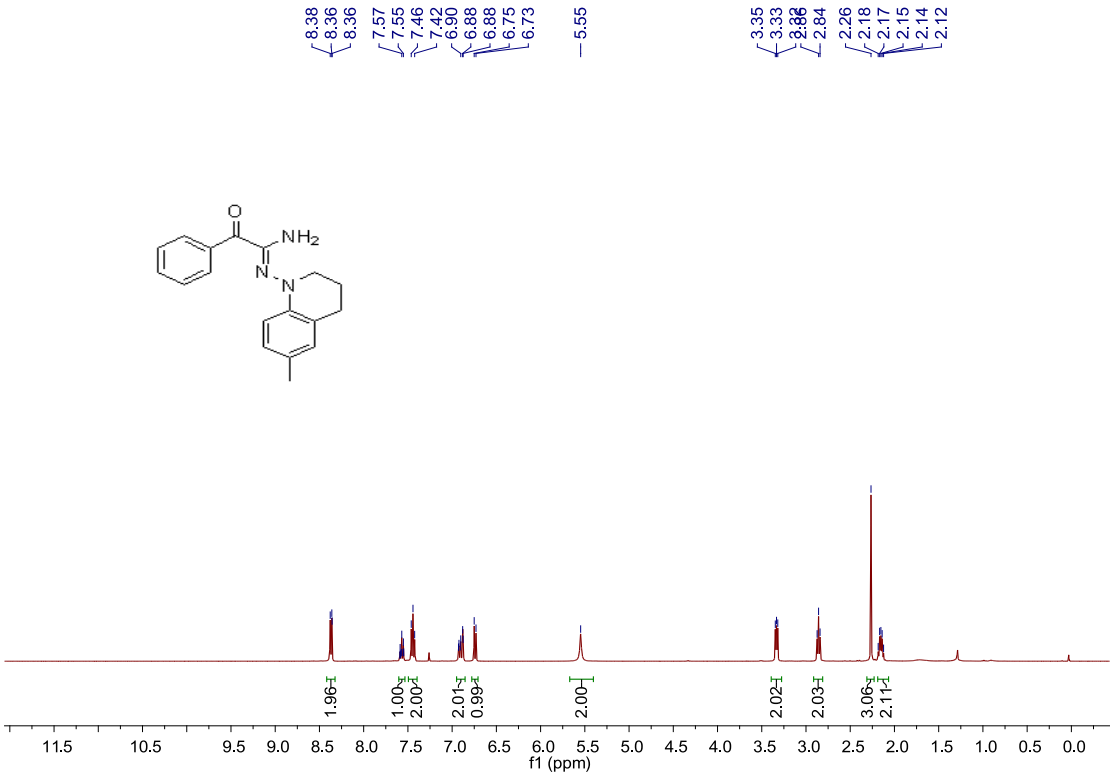
^{1}H NMR of product 60 in $CDCl_3$ (400 MHz)



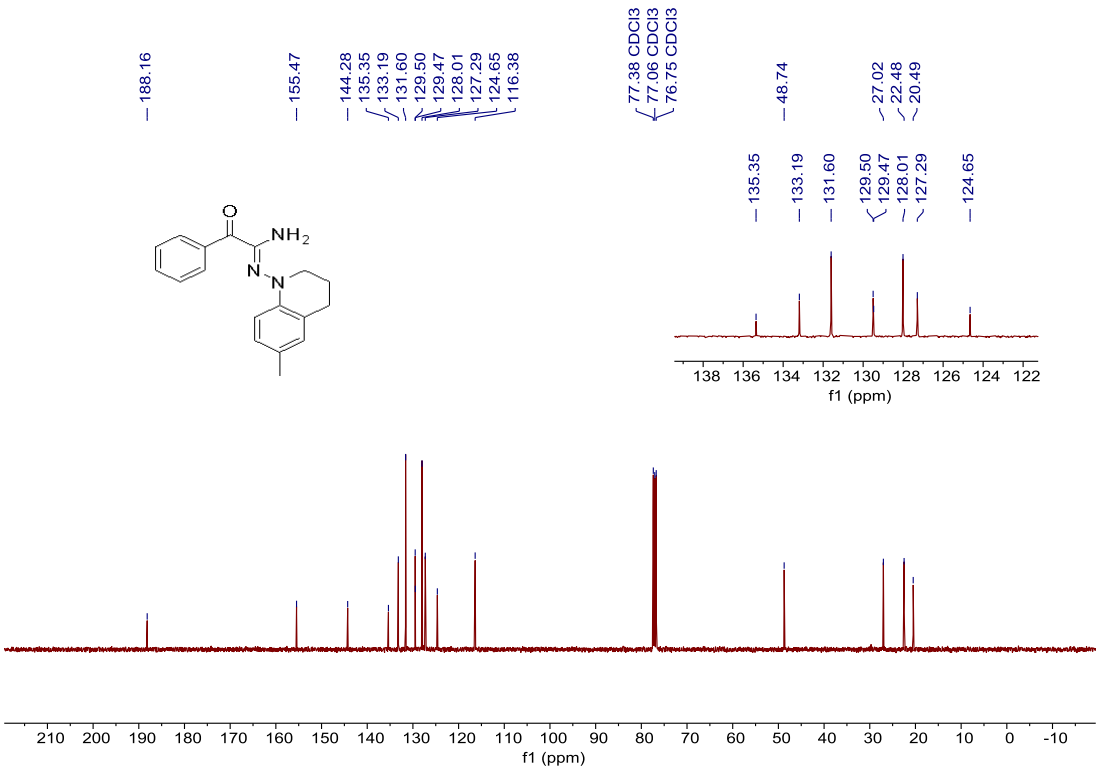
^{13}C NMR of product 60 in $CDCl_3$ (100 MHz)



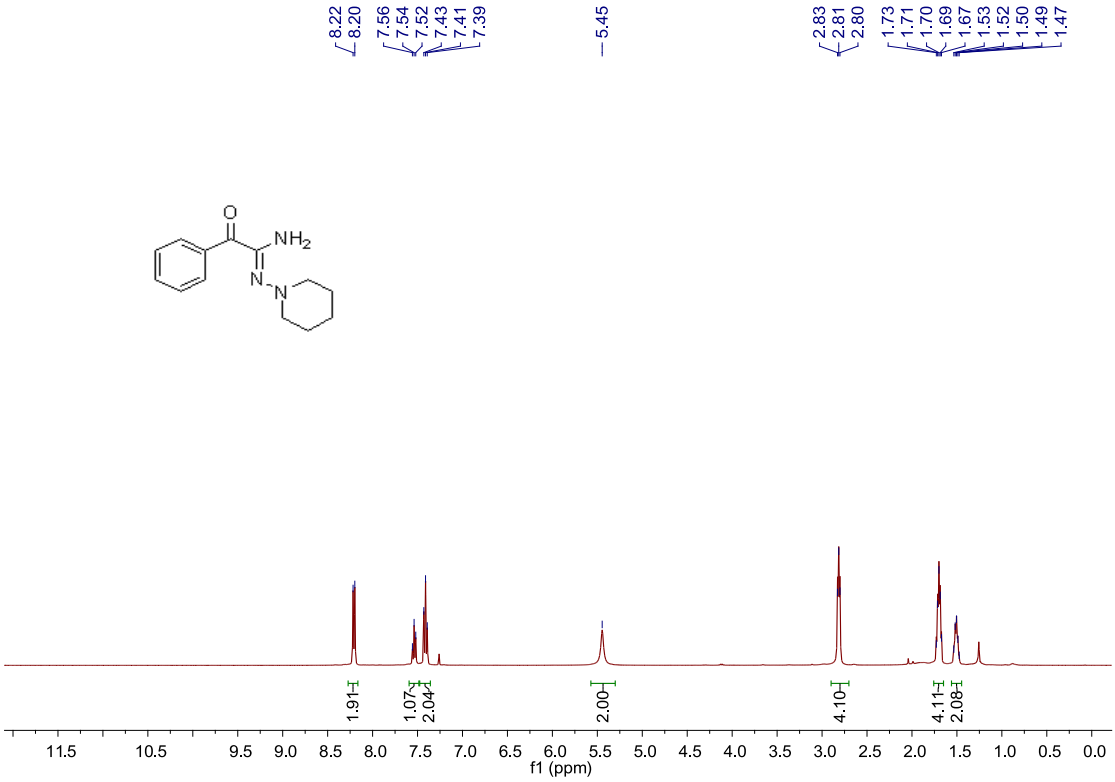
^{1}H NMR of product 61 in $CDCl_3$ (400 MHz)



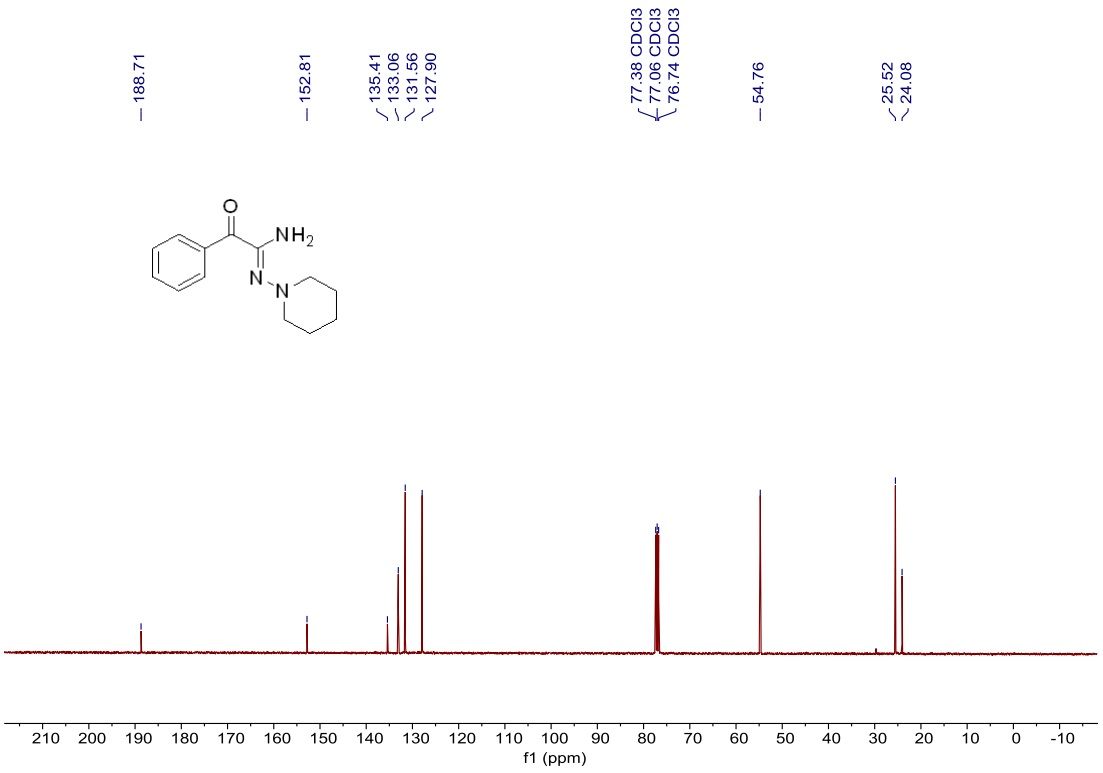
^{13}C NMR of product 61 in $CDCl_3$ (100 MHz)



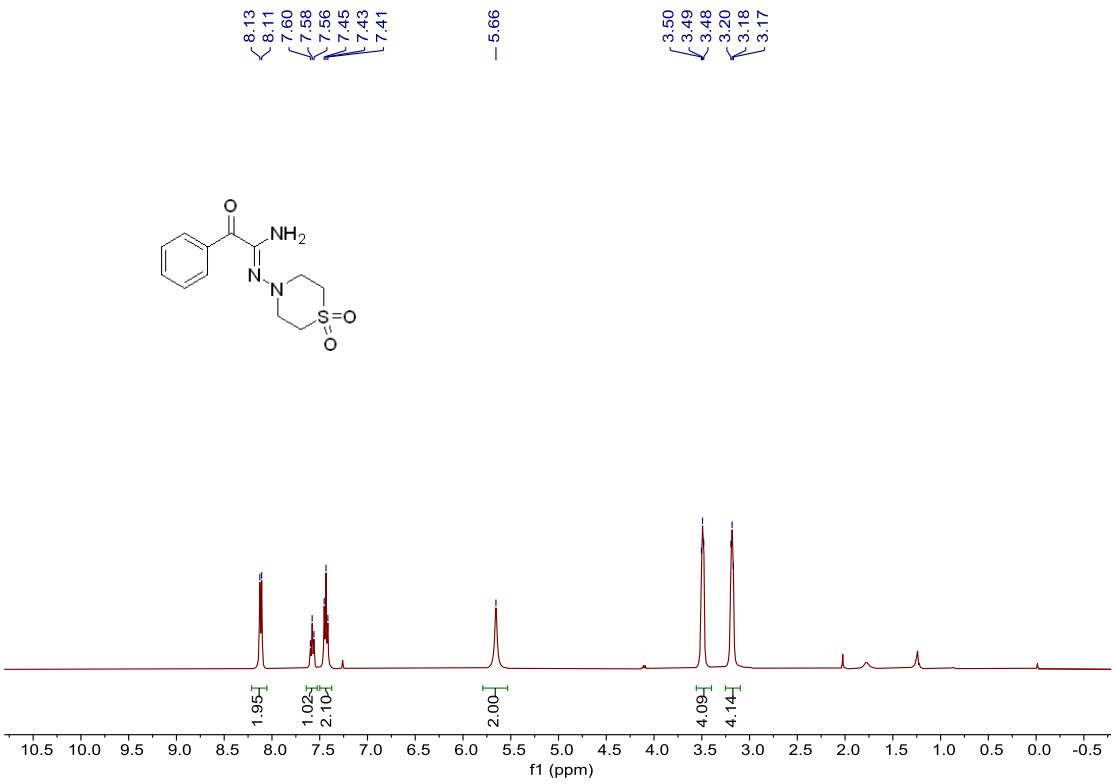
^{1}H NMR of product 62 in $CDCl_3$ (400 MHz)



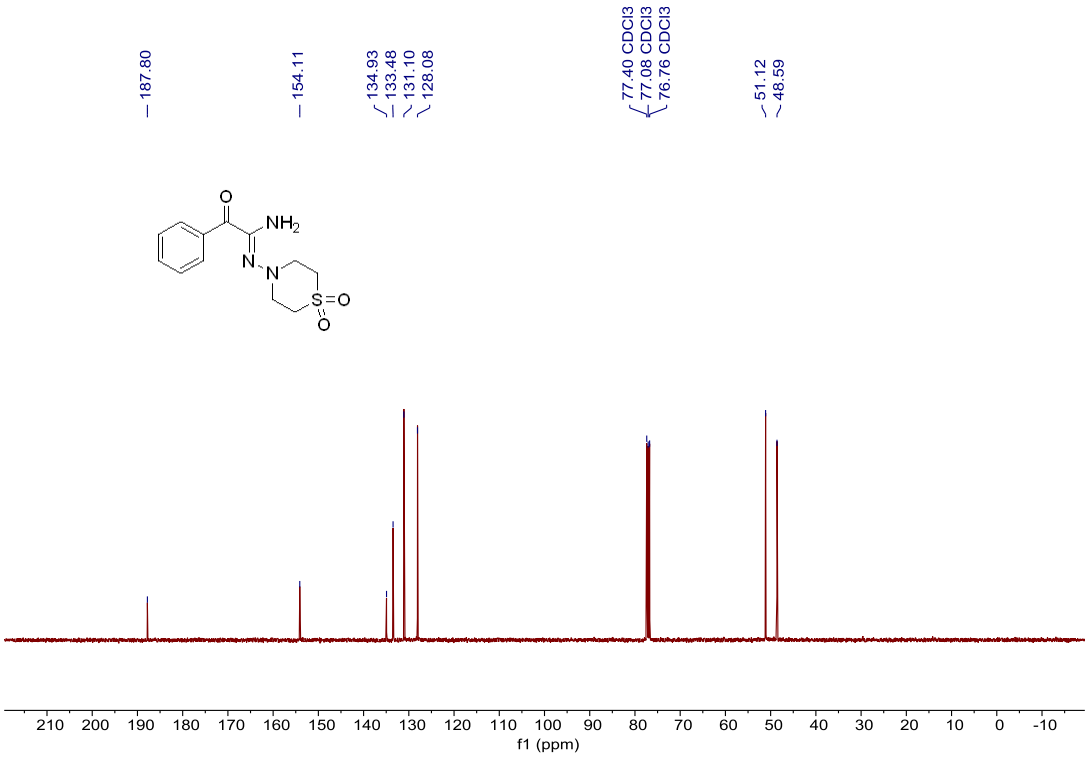
^{13}C NMR of product 62 in $CDCl_3$ (100 MHz)



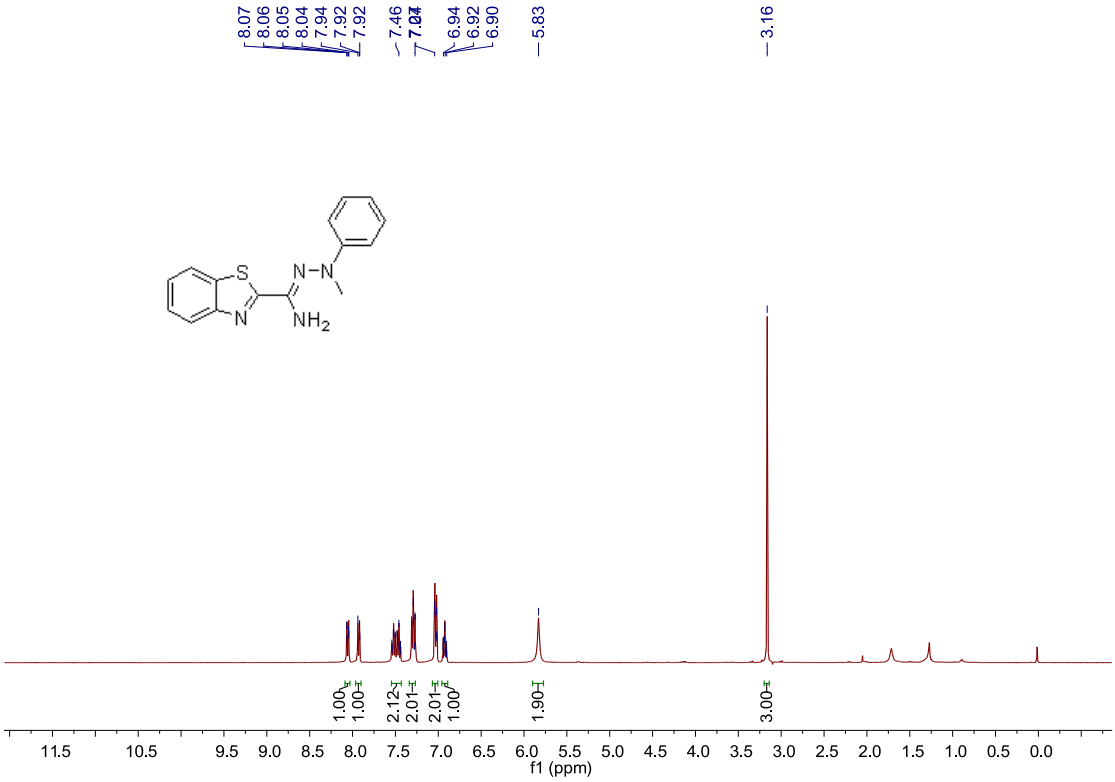
^{1}H NMR of product 63 in $CDCl_3$ (400 MHz)



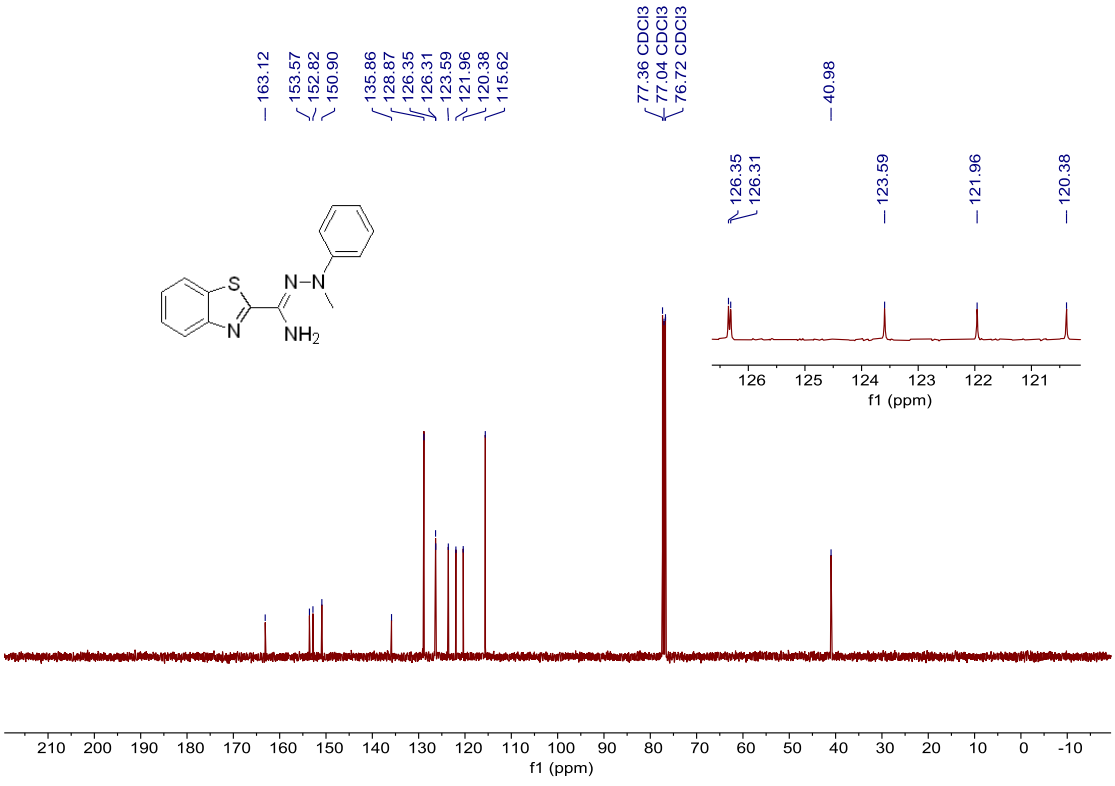
^{13}C NMR of product 63 in $CDCl_3$ (100 MHz)



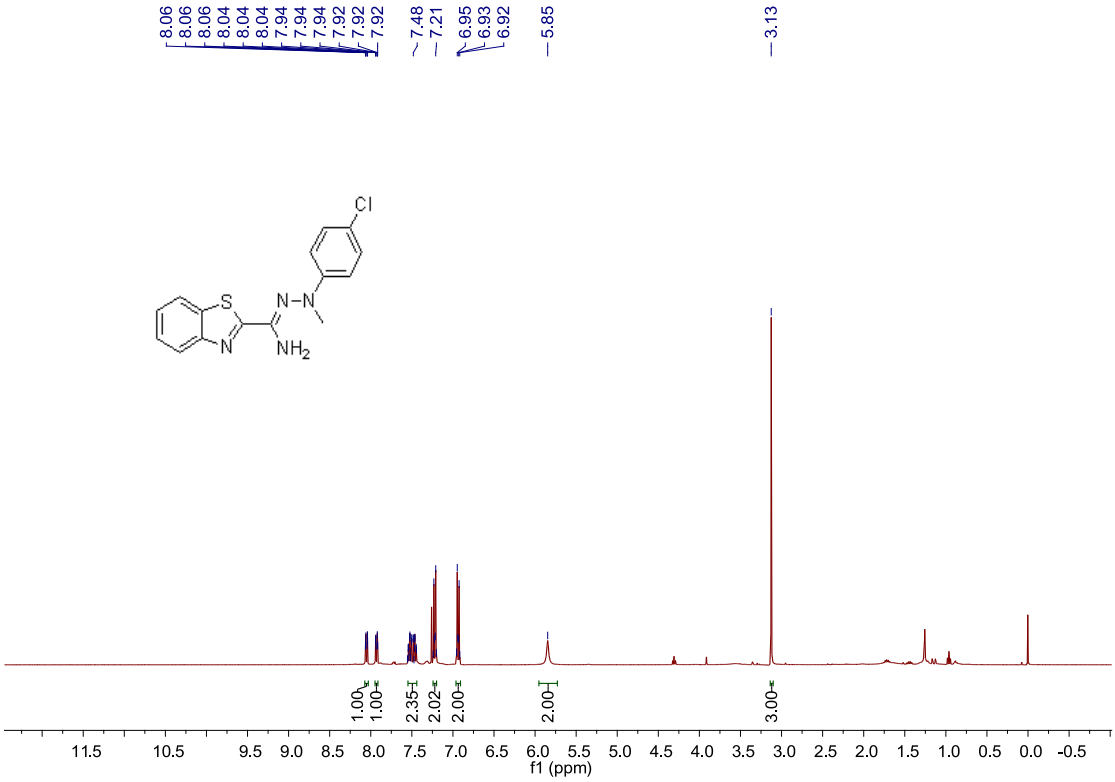
^{1}H NMR of product 65 in $CDCl_3$ (400 MHz)



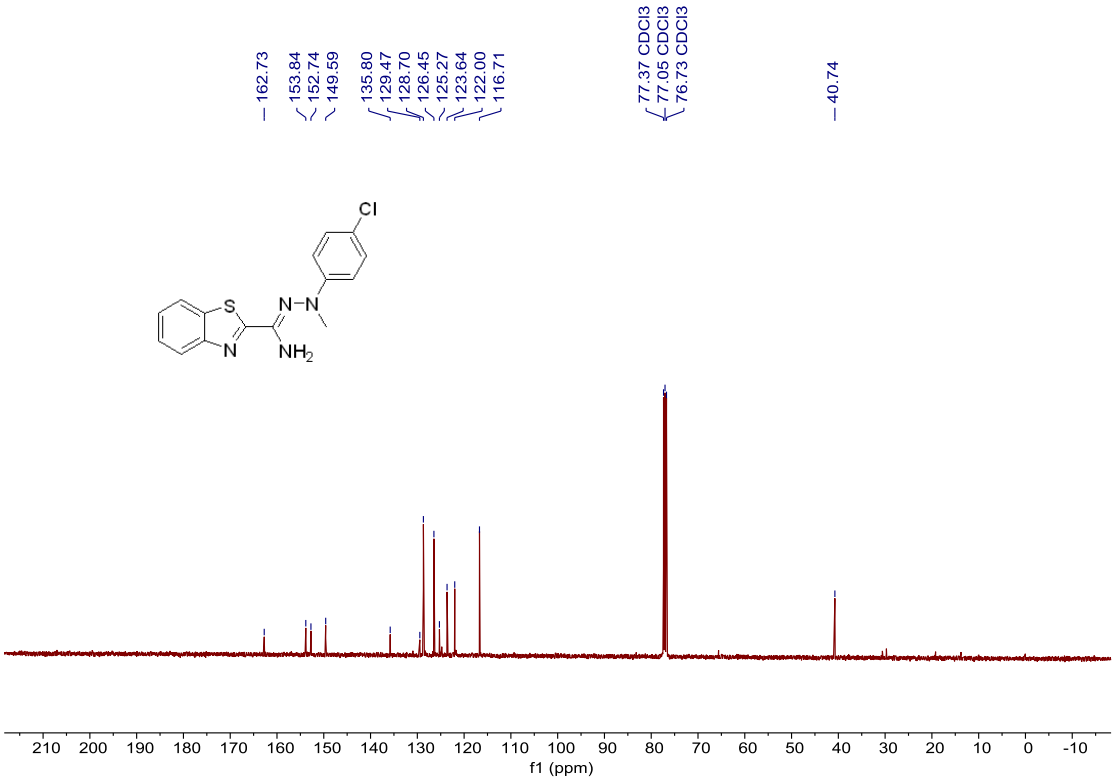
^{13}C NMR of product 65 in $CDCl_3$ (100 MHz)



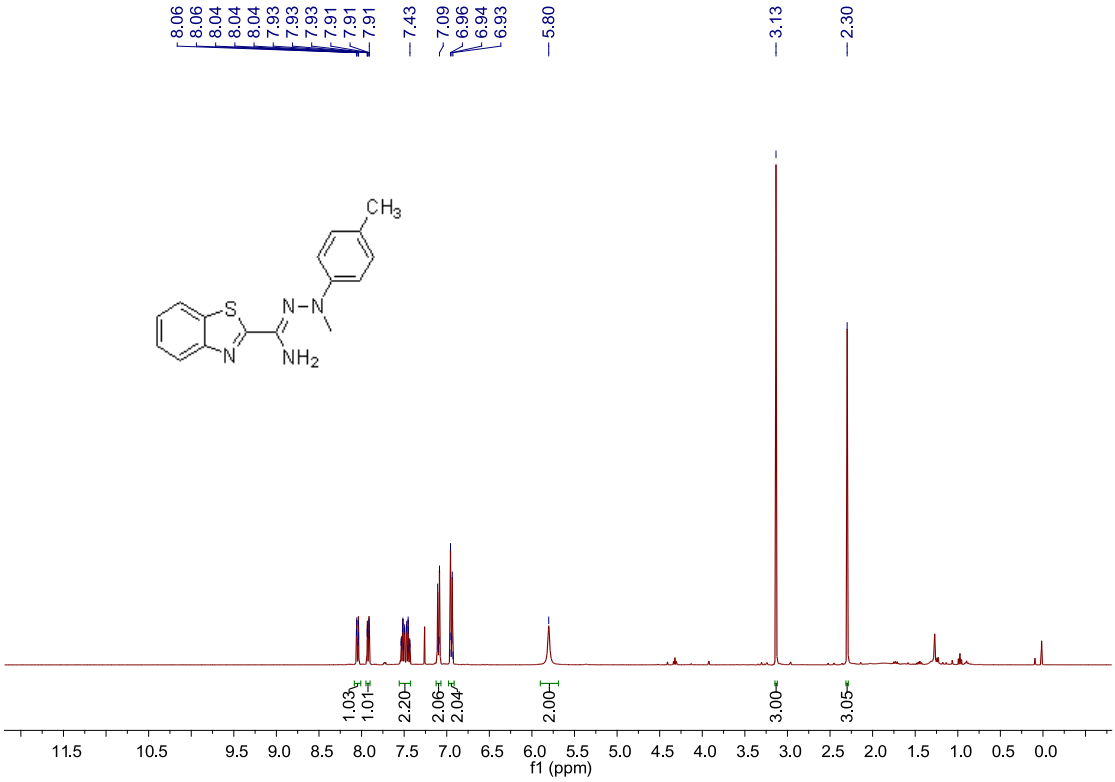
^{1}H NMR of product 66 in $CDCl_3$ (400 MHz)



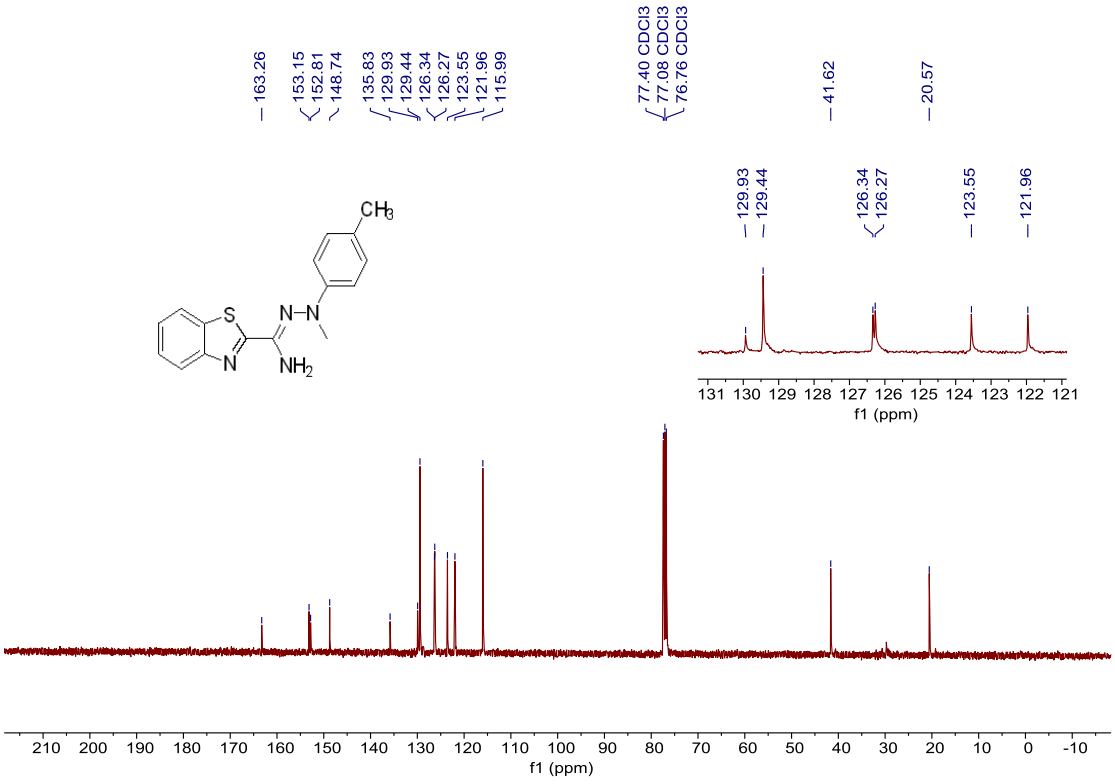
^{13}C NMR of product 66 in $CDCl_3$ (100 MHz)



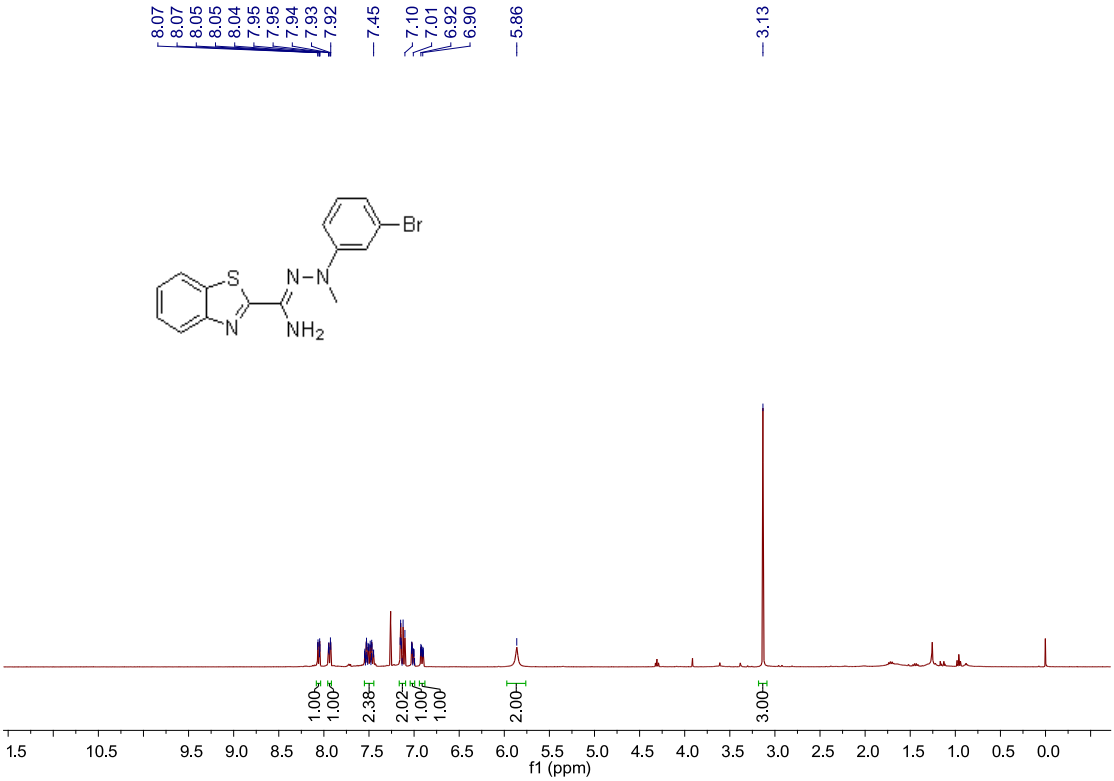
^{1}H NMR of product 67 in $CDCl_3$ (400 MHz)



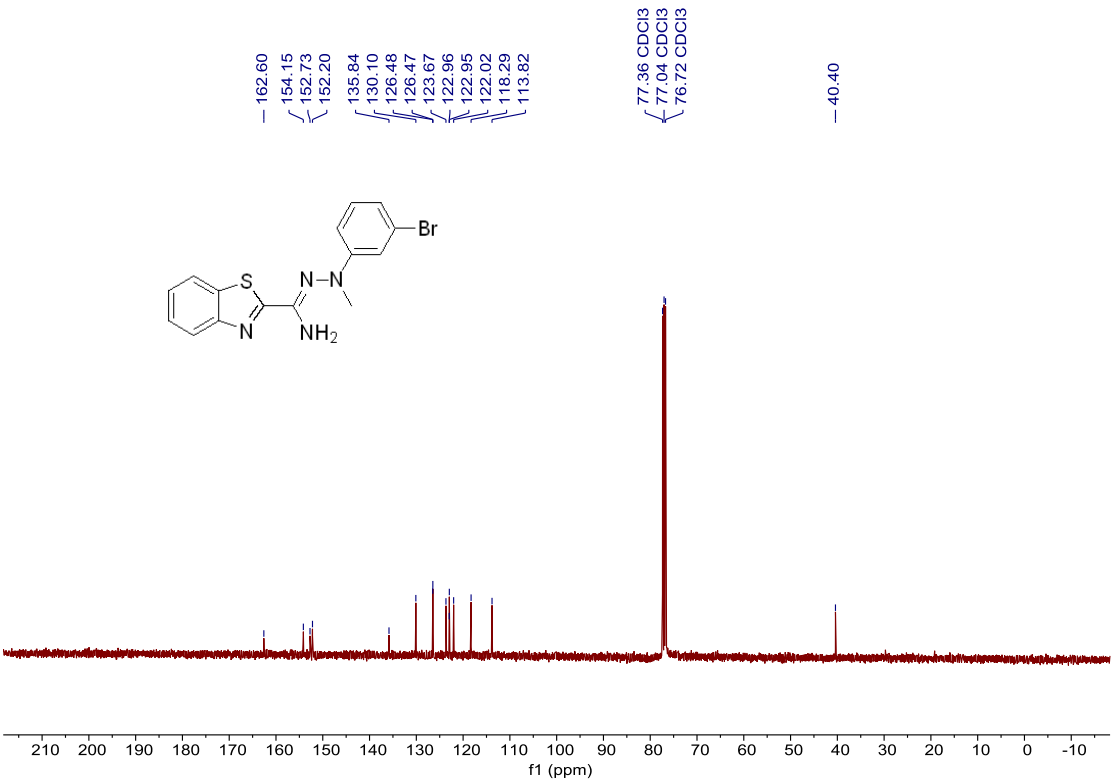
^{13}C NMR of product 67 in $CDCl_3$ (100 MHz)



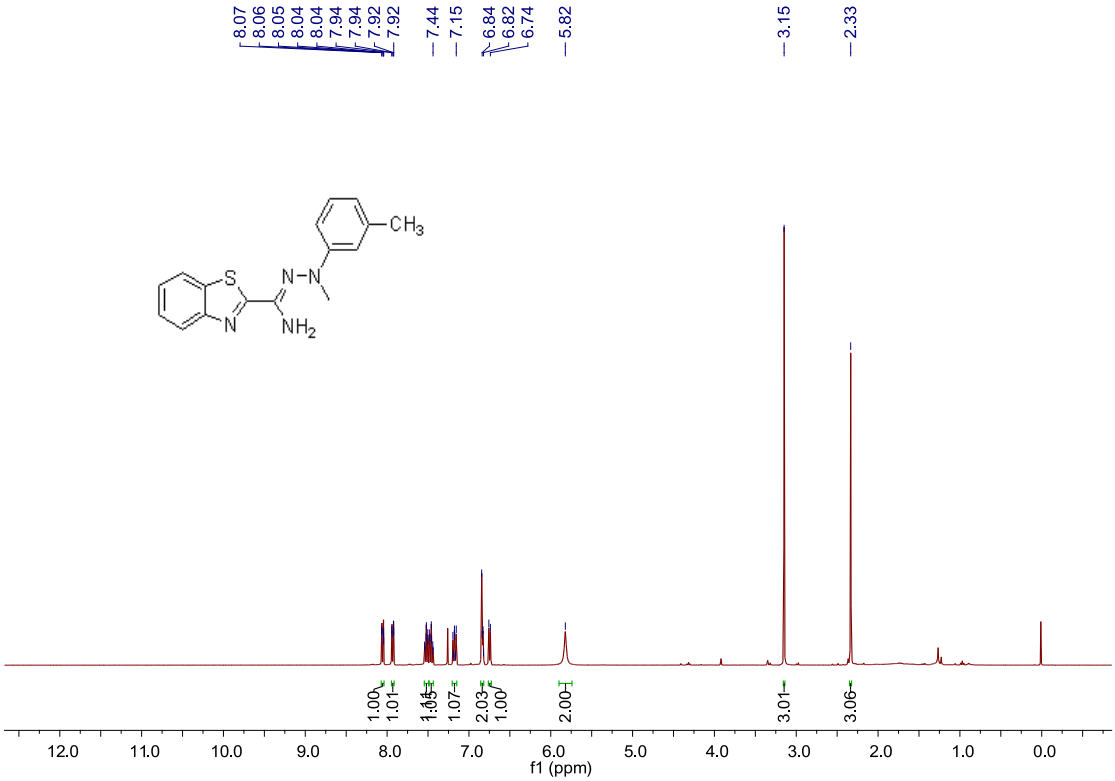
^{1}H NMR of product 68 in $CDCl_3$ (400 MHz)



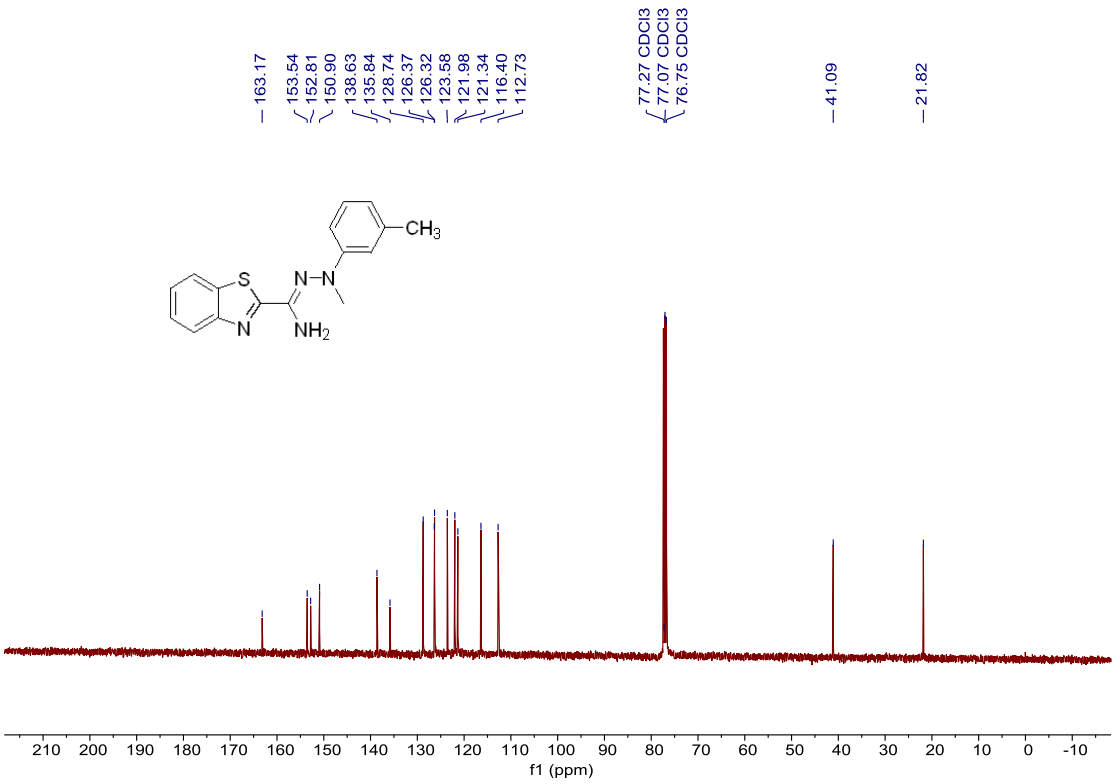
^{13}C NMR of product 68 in $CDCl_3$ (100 MHz)



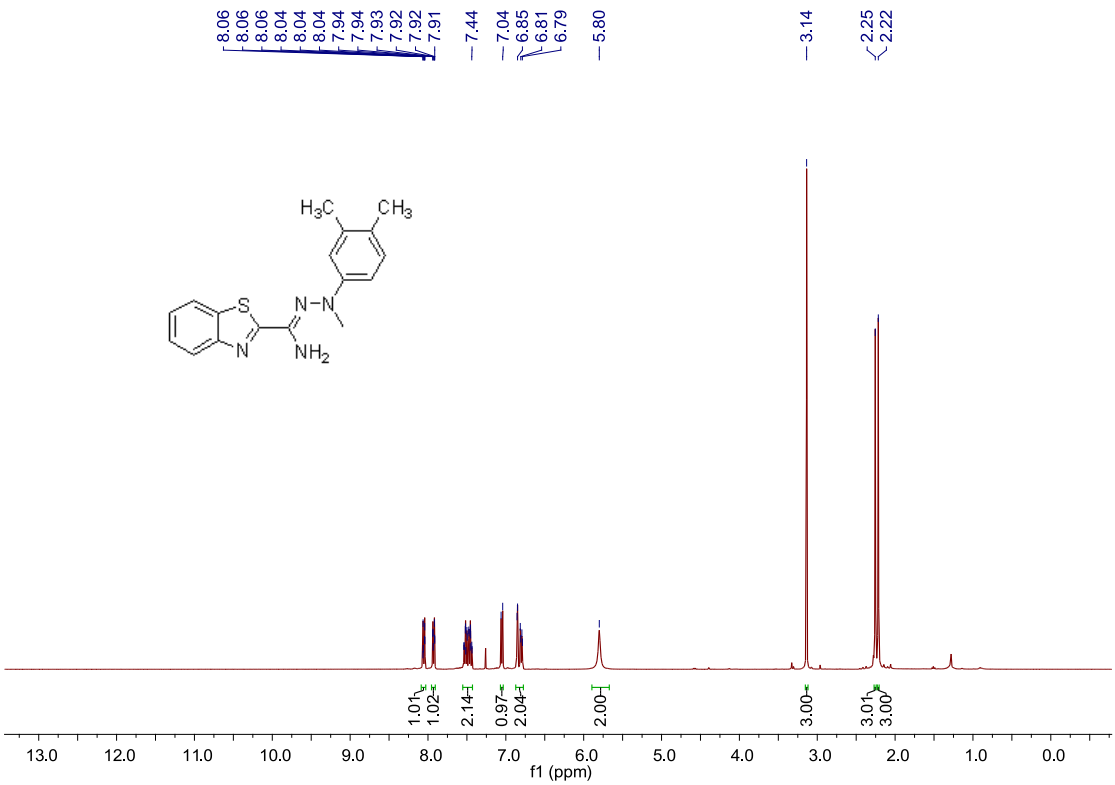
^{1}H NMR of product 69 in $CDCl_3$ (400 MHz)



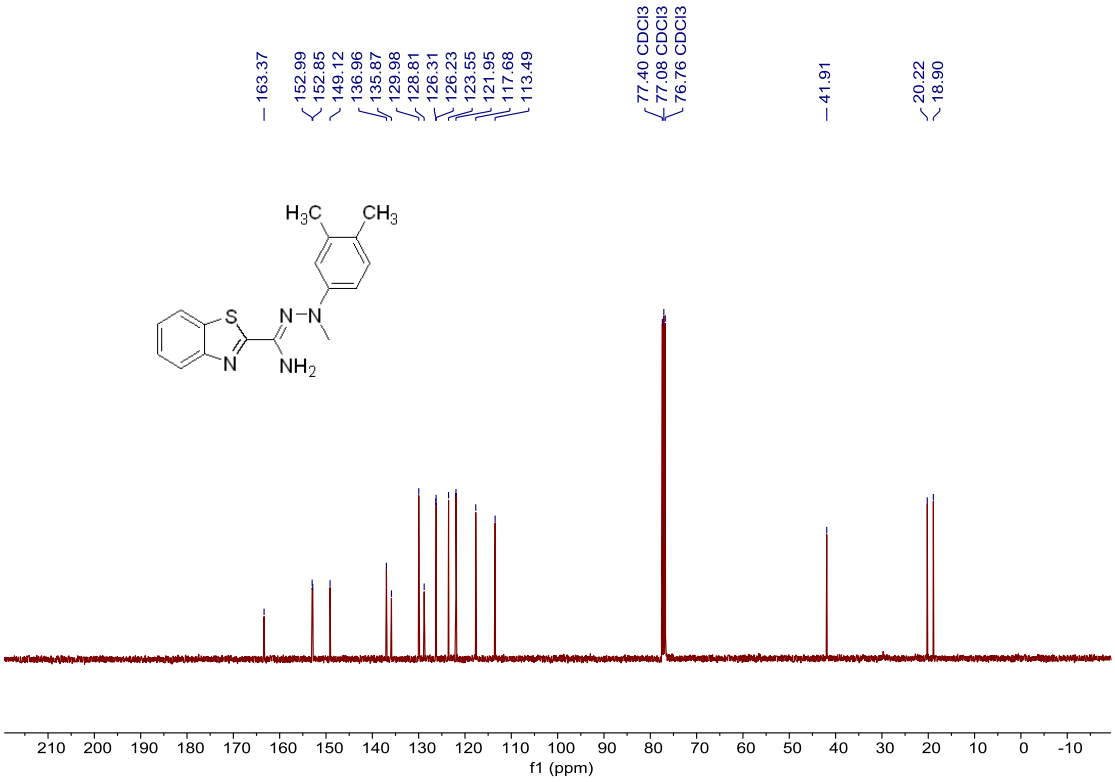
^{13}C NMR of product 69 in $CDCl_3$ (100 MHz)



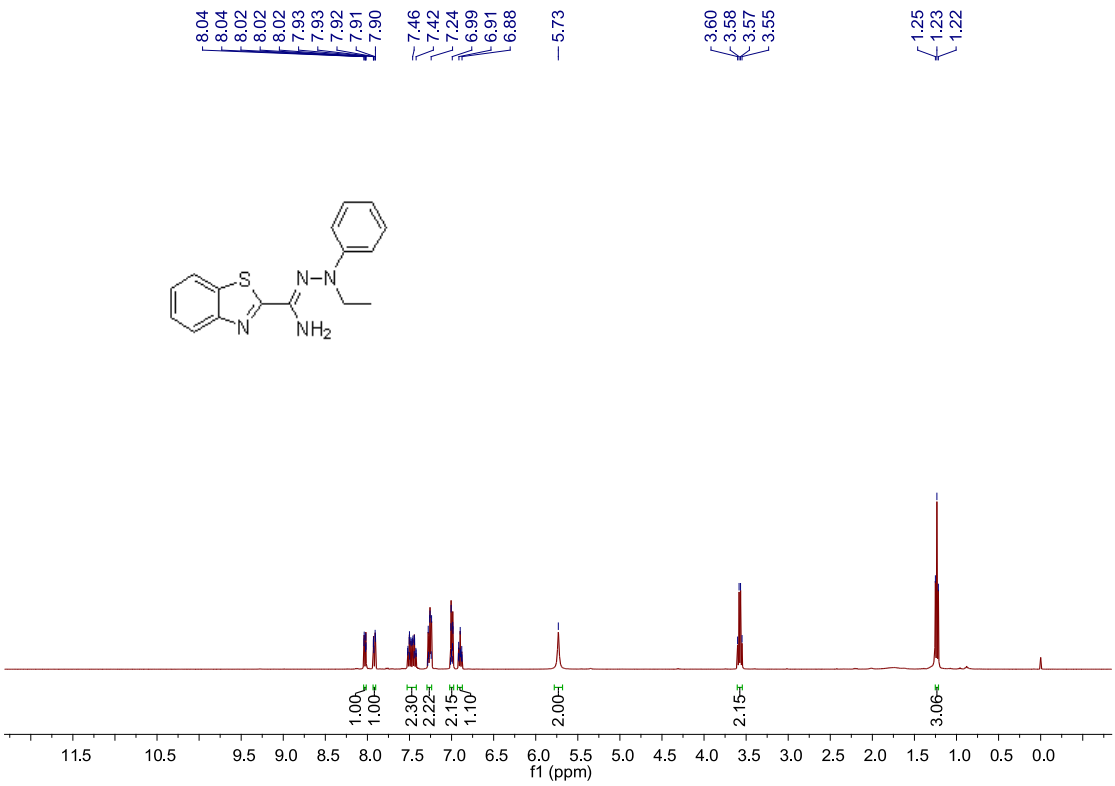
^{1}H NMR of product 70 in $CDCl_3$ (400 MHz)



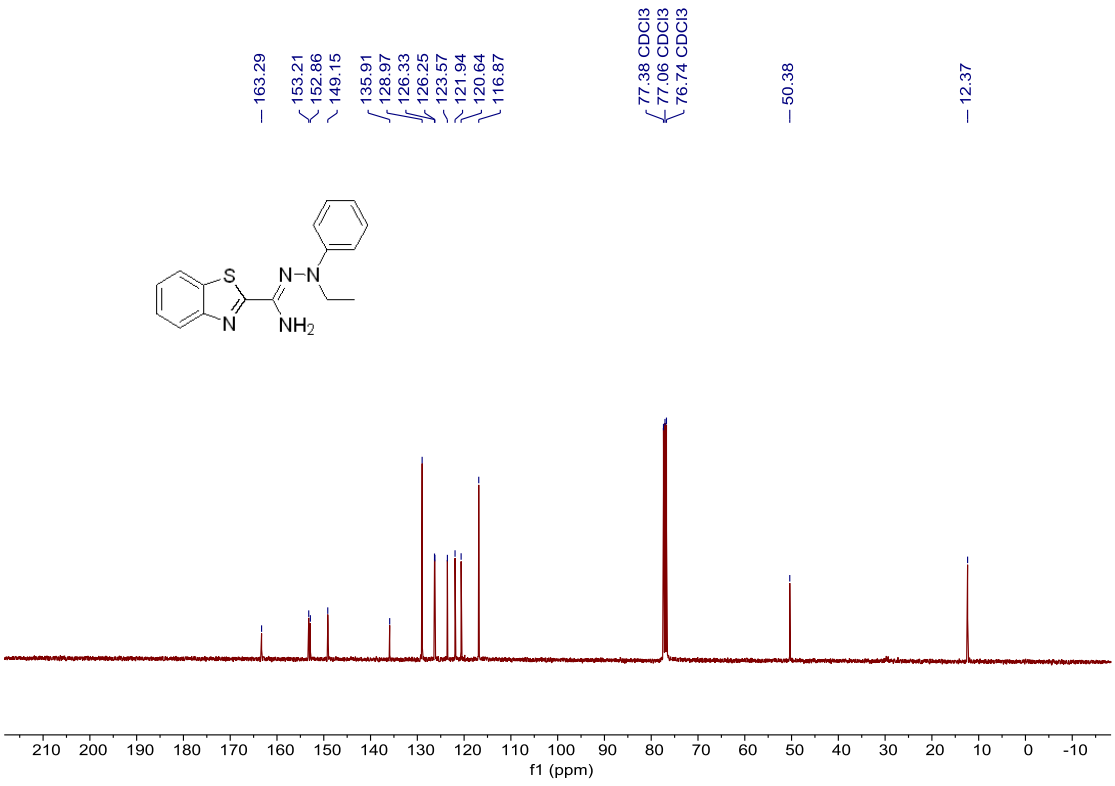
^{13}C NMR of product 70 in $CDCl_3$ (100 MHz)



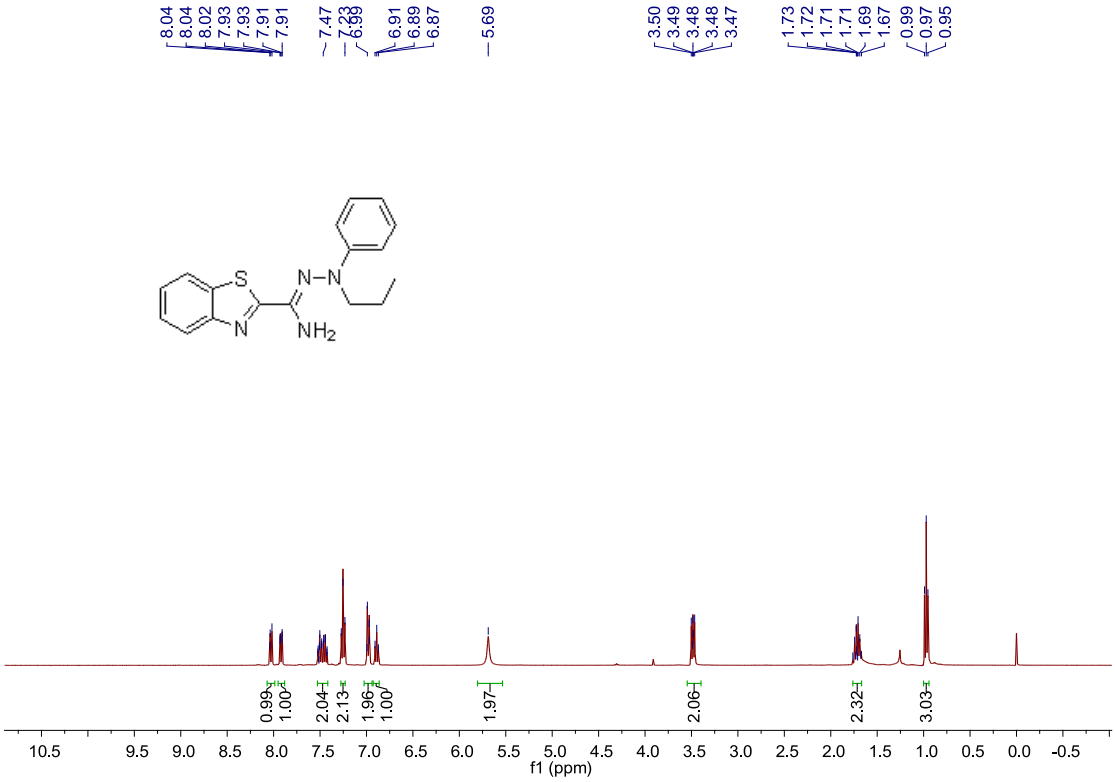
^{1}H NMR of product 71 in $CDCl_3$ (400 MHz)



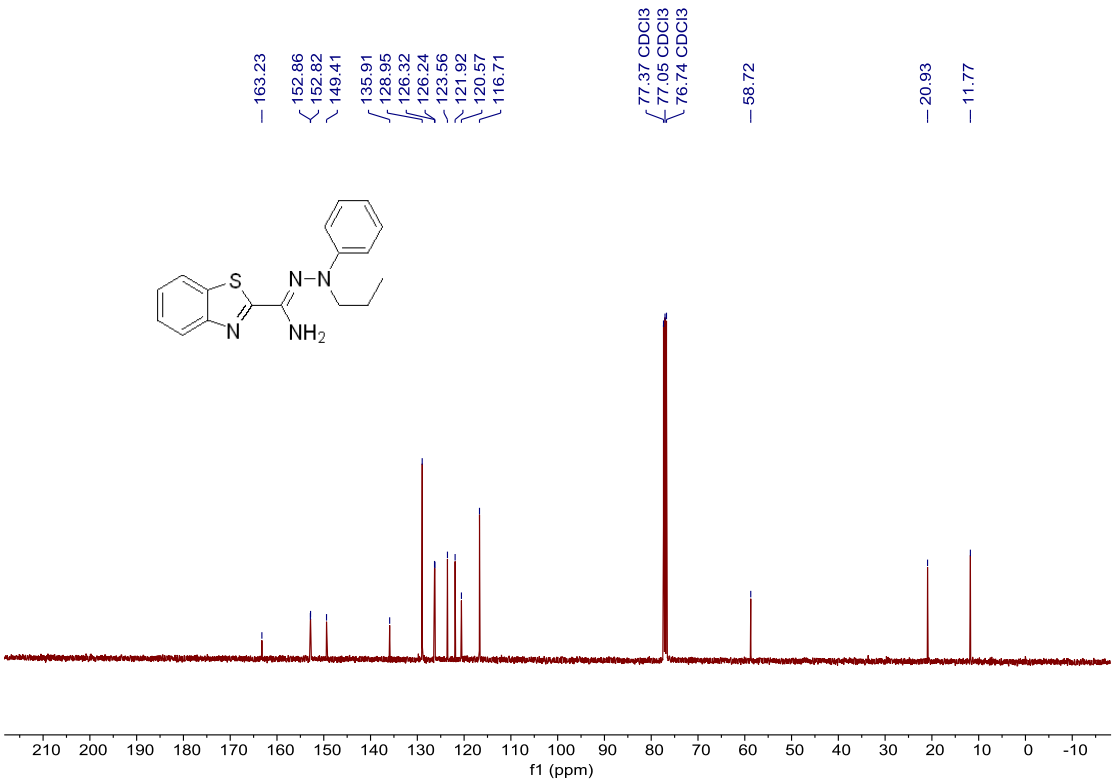
^{13}C NMR of product 71 in $CDCl_3$ (100 MHz)



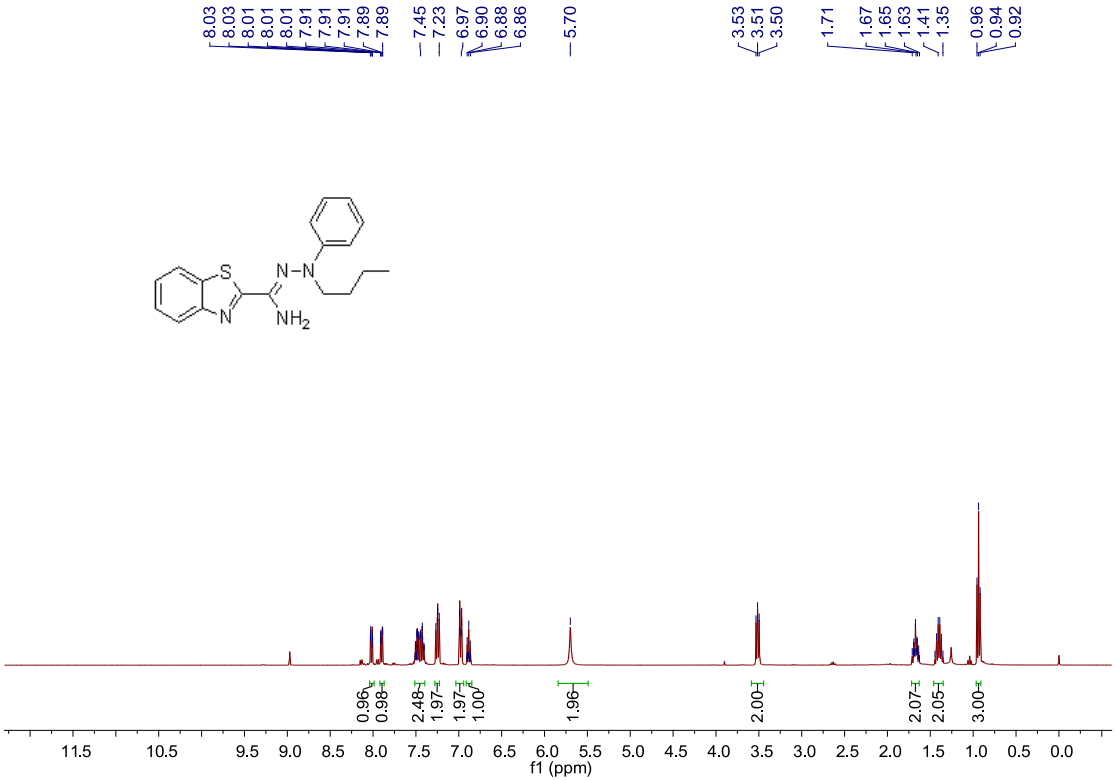
^{1}H NMR of product 72 in $CDCl_3$ (400 MHz)



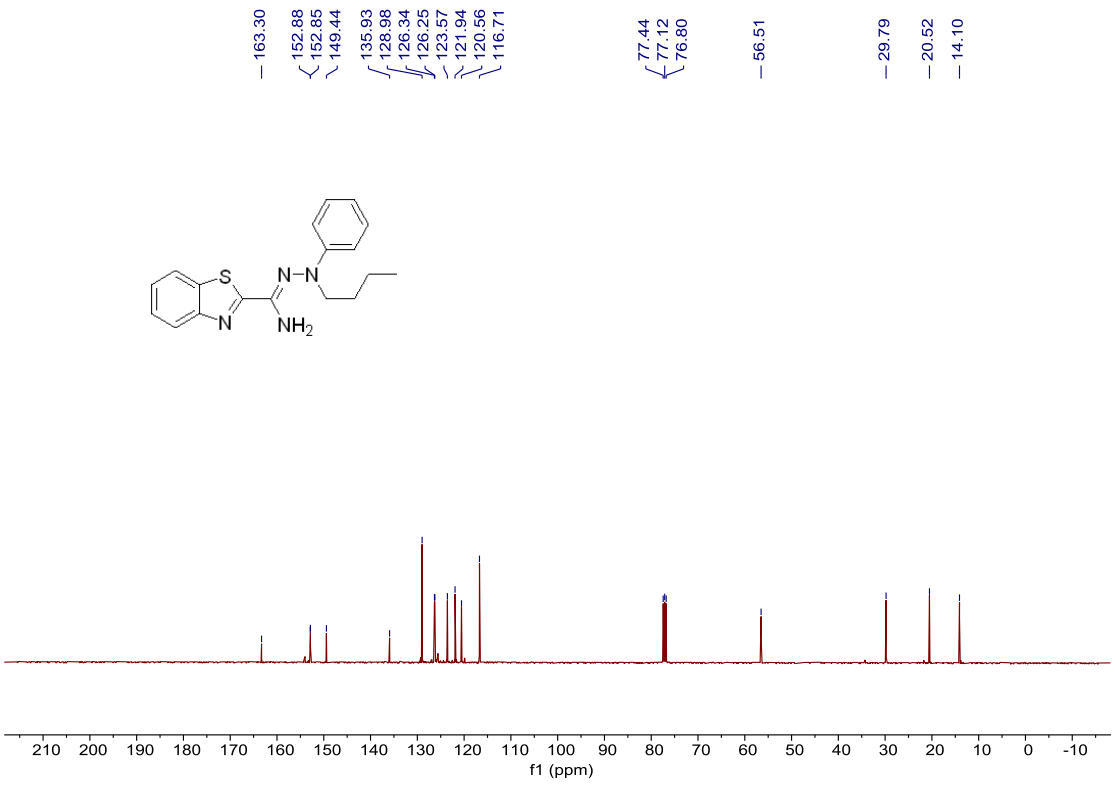
^{13}C NMR of product 72 in $CDCl_3$ (100 MHz)



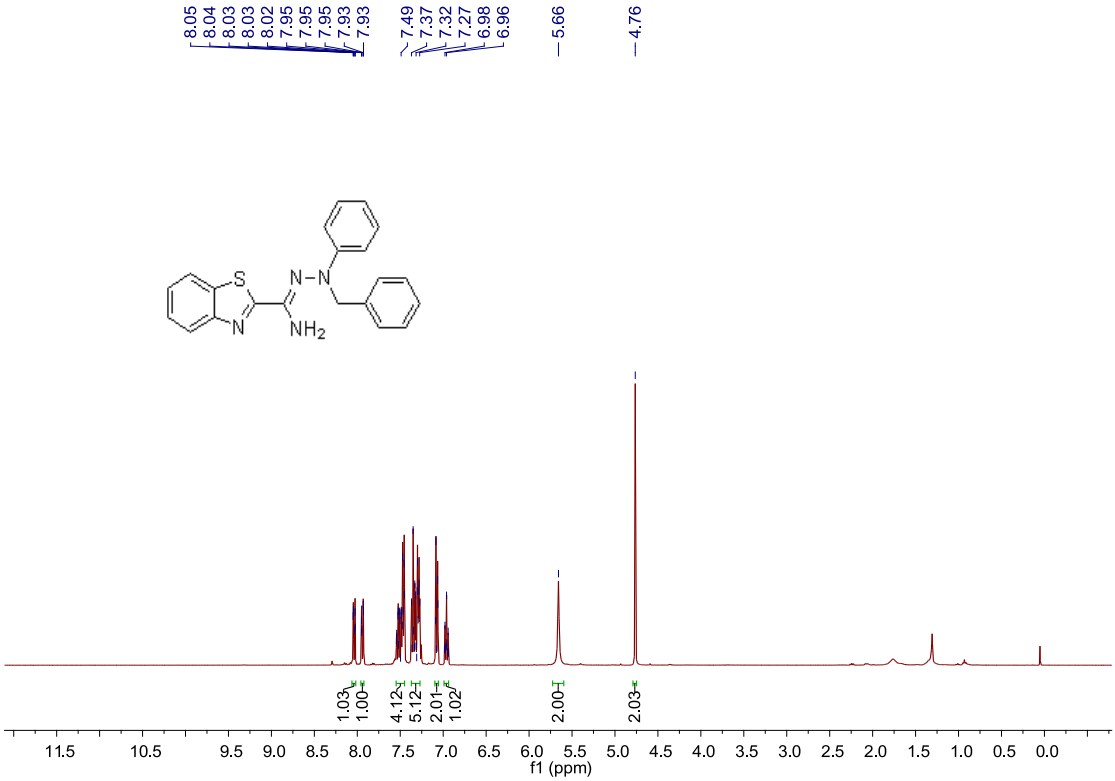
^{1}H NMR of product 73 in $CDCl_3$ (400 MHz)



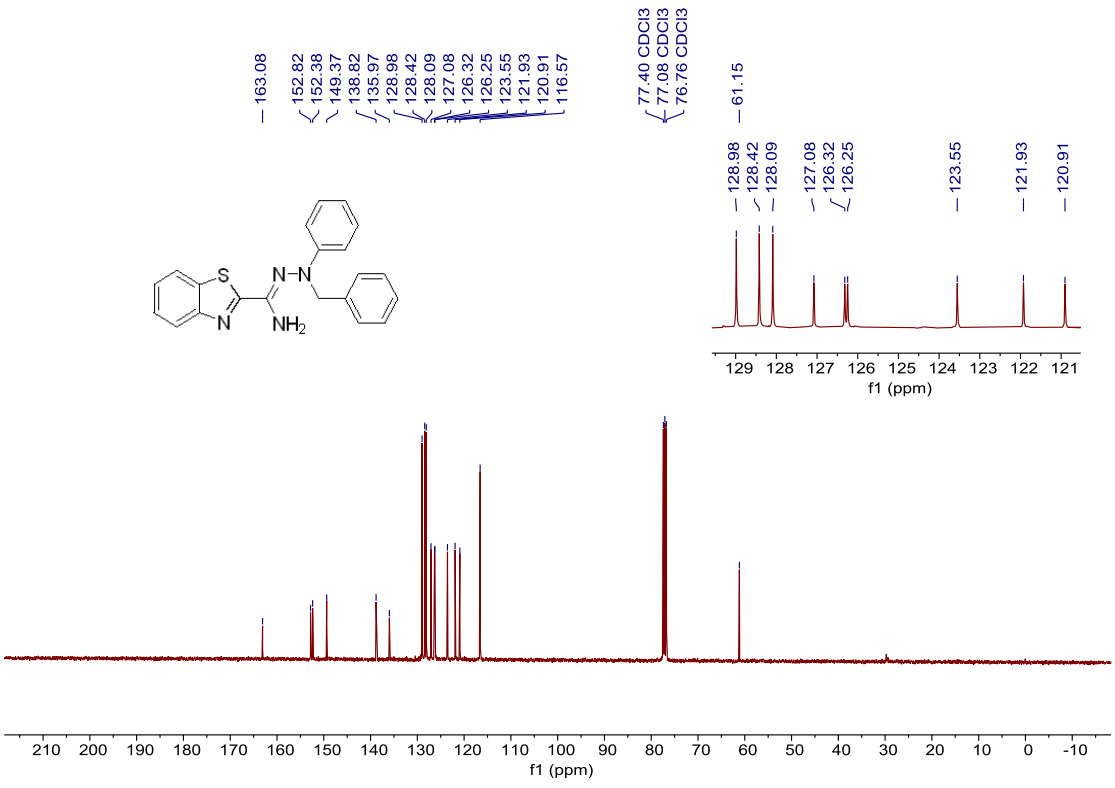
^{13}C NMR of product 73 in $CDCl_3$ (100 MHz)



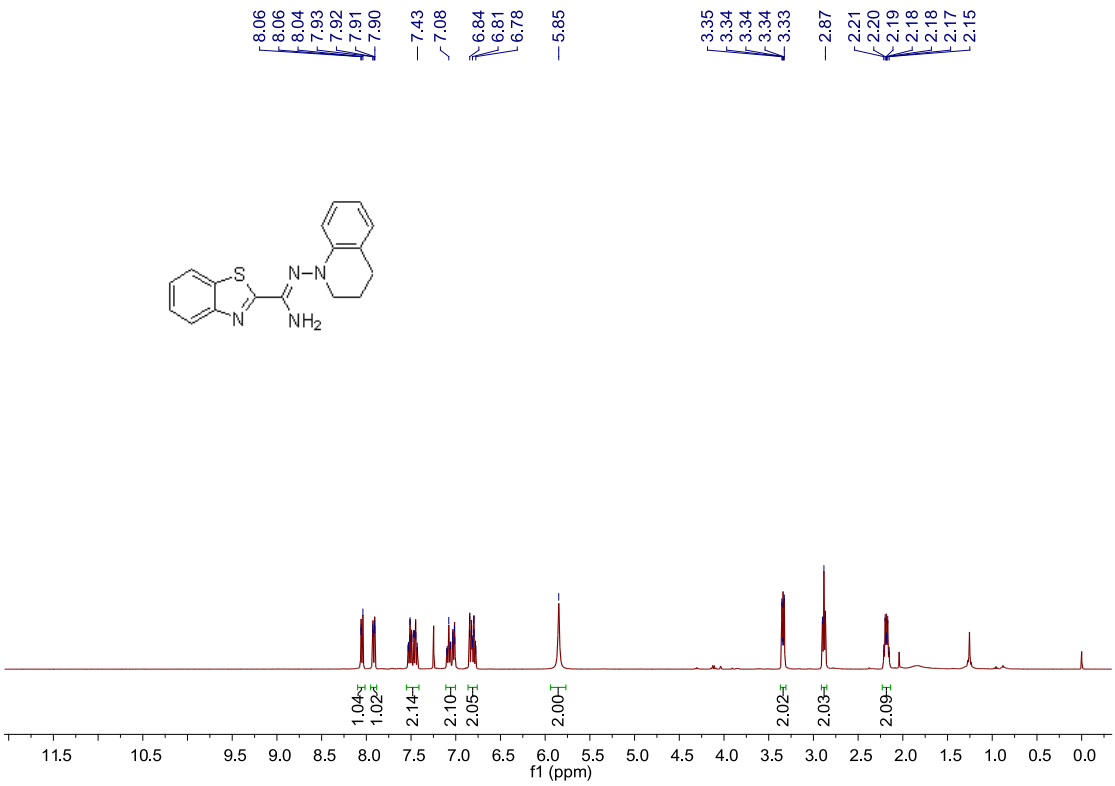
^{1}H NMR of product 74 in $CDCl_3$ (400 MHz)



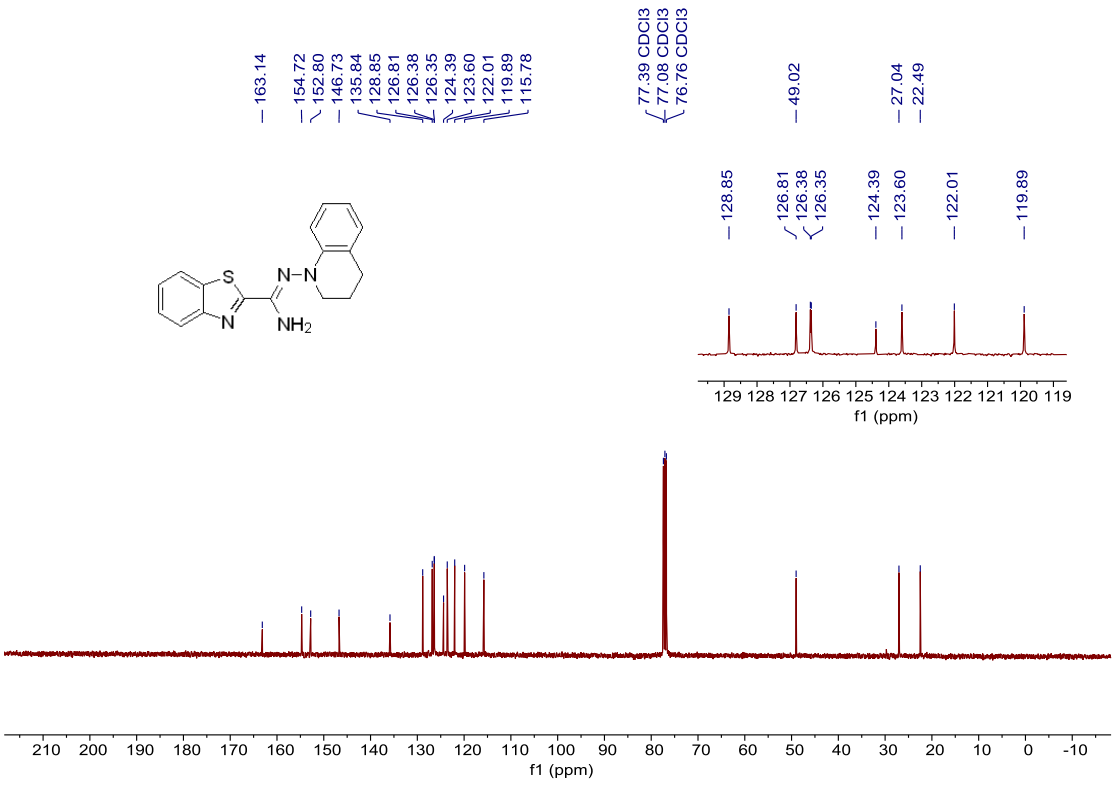
^{13}C NMR of product 74 in $CDCl_3$ (100 MHz)



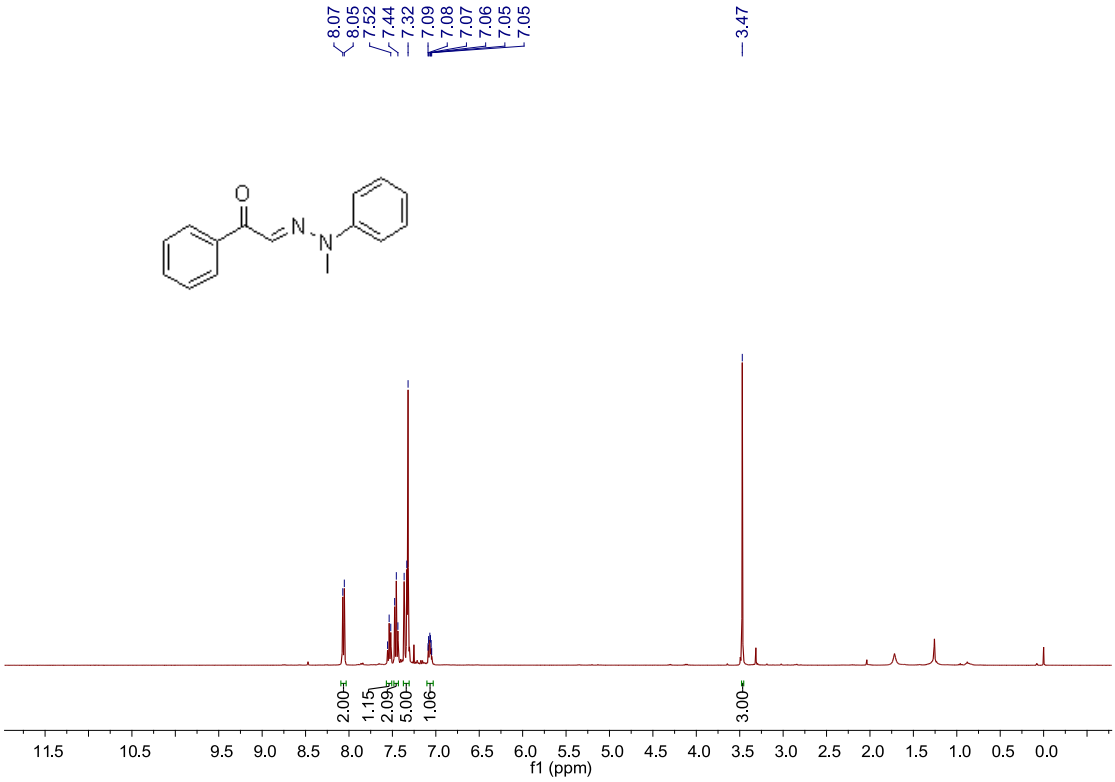
^{1}H NMR of product 75 in $CDCl_3$ (400 MHz)



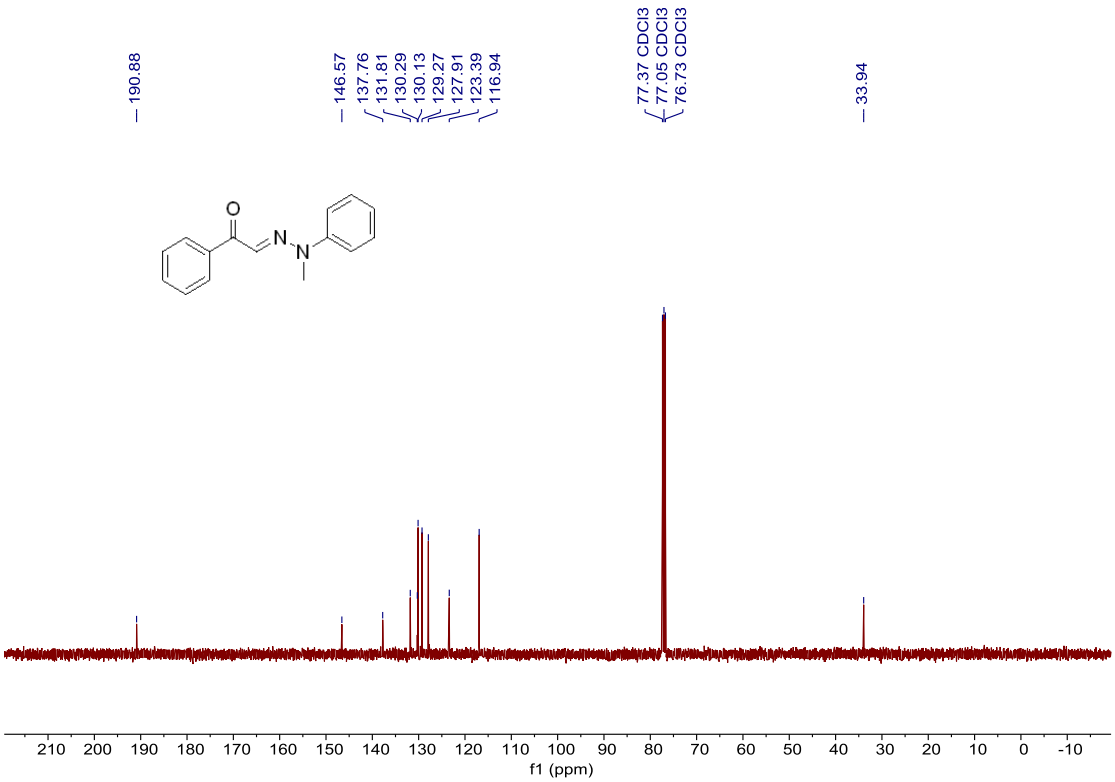
^{13}C NMR of product 75 in $CDCl_3$ (100 MHz)



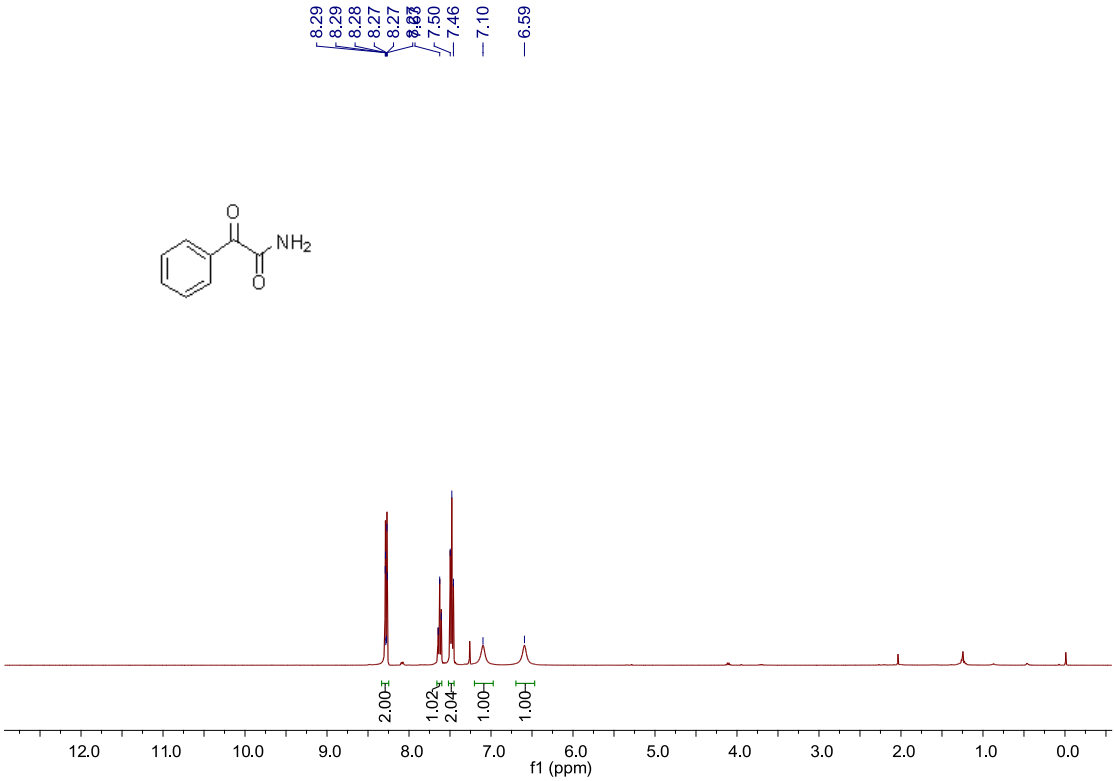
^{1}H NMR of H in $CDCl_3$ (400 MHz)



^{13}C NMR of H in $CDCl_3$ (100 MHz)



^{1}H NMR of product 84 in $CDCl_3$ (400 MHz)



^{13}C NMR of product 84 in $CDCl_3$ (100 MHz)