Supporting Information

Expedient Tandem Dehydrogenative Alkylation and Cyclization Reactions Under Mn(I)-Catalysis

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Contents

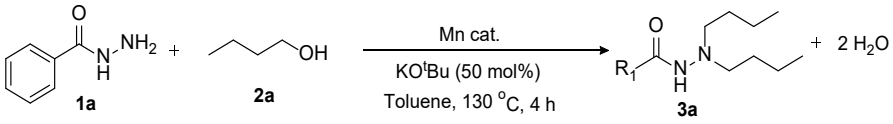
1. General Information

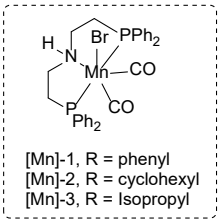
All catalytic experiments were carried out using standard Schlenk techniques. All solvents were reagent grade or better. Deuterated solvents were used as received. Most of the chemicals used in the catalytic reactions were purified according to standard procedure. Thin layer chromatography (TLC) was performed using silica gel 60 F_{254} coated on aluminium sheet purchased from Merck which was visualized with UV light at 254 nm or under iodine. Column chromatography was performed with SiO_2 (Silicycle Silica flash F60 (230-400 mesh). ^1H NMR (400 MHz), ^{13}C NMR (100 MHz) spectra were recorded on the NMR spectrometer. Deuterated chloroform was used as the solvent, and chemical shift values (δ) are reported in parts per million relatives to the residual signals of this solvent [δ 7.27 for ^1H (chloroform-d), δ 77.0 for ^{13}C (chloroform-d). Abbreviations used in the NMR follow-up experiments: br, broad; s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet. High-resolution mass spectra (HRMS) were obtained by fast atom bombardment (FAB) using a double-focusing magnetic sector mass spectrometer and electron impact (EI) ionization technique (magnetic sector-electric sector double-focusing mass analyzer).

2. Experimental Section

2.1. Optimization conditions

Table S1: Screening of manganese pincer catalysts[a]

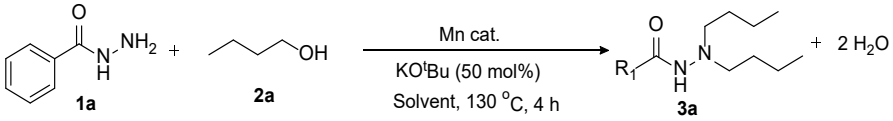




Entry	Catalyst	Yield of 3a (%)
1	**[Mn-1]**	**91 (>99)[b]**
2	[Mn-2]	71
3	[Mn-3]	68
4	-	NR

Reaction conditions: **1a** (0.5 mmol), **2a** (1.1 mmol), Mn-catalyst and KO^{t}Bu (50 mol%) using 1 mL of toluene as solvent at 130 °C (oil-bath temperature). [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR= No reaction.

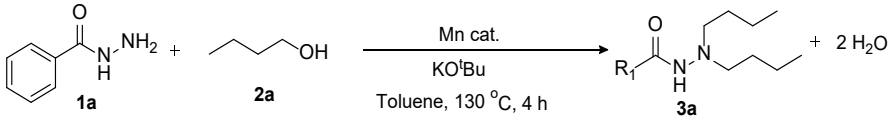
Table S2: Screening of solvent[a]



Entry	Solvent	Yield of 3a (%)
1	**Toluene**	**91 (>99)[b]**
2	DMF	10
3	MeCN	27
4	THF	trace
5	1,4 Dioxane	trace
6	n-octane	43
7	*m*-xylene	NR
8	-	trace

Reaction conditions: **1a** (0.5 mmol), **2a** (1.1 mmol), [Mn-1] (3 mol%) and KO^{t}Bu (50 mol%) using 1 mL of solvent at 130 °C (oil-bath temperature). [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR= No reaction.

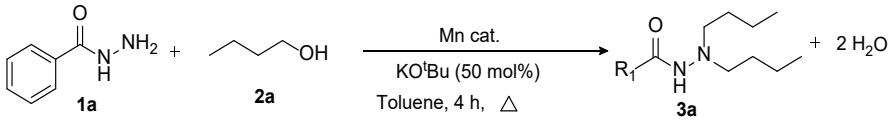
Table S3: Screening of base amount[a]



Entry	KO^{t}Bu(eq)	Yield of 3a (%)[b]
1	30 mol%	68
2	**50 mol%**	**91 (>99)[b]**
3	75 mol%	95
4	1 equiv.	85
5	-	trace

Reaction conditions: **1a** (0.5 mmol), **2a** (1.1 mmol), [Mn-1] (3 mol%), KO^{t}Bu using 1 mL of toluene as solvent at 130 °C (oil-bath temperature). [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR = No reaction.

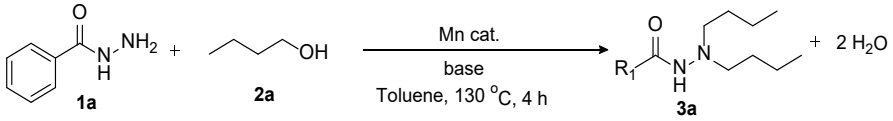
Table S4: Screening of temperature[a]



Entry	Temperature (°C)	Yield of 3a (%)[b]
1	RT	NR
2	40 °C	NR
3	80 °C	13
4	110 °C	59
5	120 °C	71
6	**130 °C**	**91 (>99)[b]**

Reaction conditions: Reaction conditions: **1a** (0.5 mmol), **2a** (1.1 mmol), [Mn-1] (3 mol%), KO^{t}Bu (50 mol%) using 1 mL of toluene as solvent at different (oil-bath) temperatures. [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR = No reaction.

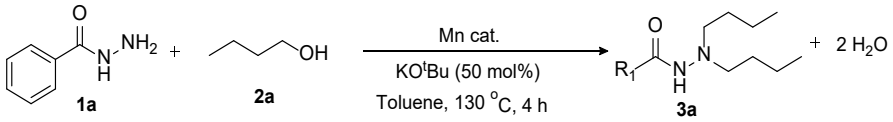
Table S5: Screening of base[a]



Entry	Base	Yield of 3a (%)[b]
1	$NaHCO_3$	trace
2	**KO^{t}Bu**	**91 (>99)[b]**
3	KOH	57
4	NaOH	45
5	K_2CO_3	25
6	NaO^{t}Bu	59
7	KH	NR
8	-	NR

Reaction conditions: Reaction conditions: **1a** (0.5 mmol), **2a** (0.5 mmol), [Mn-1] (3 mol%), base (50 mol%) using 1 mL of toluene as solvent at at 130 ºC (oil-bath temperature). [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR = No reaction.

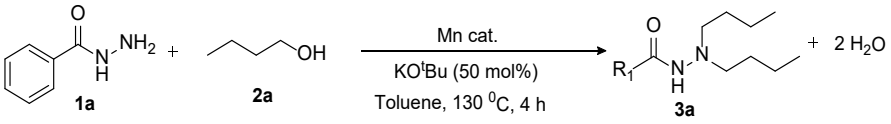
Table S6: Screening of primary alcohol amount[a]



Entry	Alcohol 2a	Yield of 3a (%)[b]
1	**2.2 equiv.**	**91 (>99)[b]**
2	2.5 equiv.	87
3	3 equiv.	79

Reaction conditions: **1a** (0.5 mmol), **2a**, Mn-catalyst (3 mol%) and KO^{t}Bu (50 mol%) using 1 mL of toluene as solvent at 130 ºC (oil-bath temperature). [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR= No reaction.

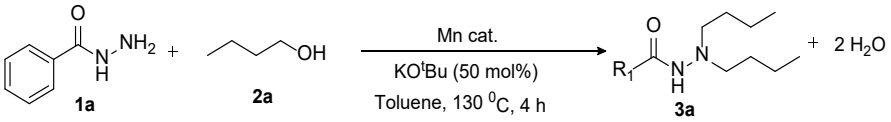
Table S7: Screening of catalyst amount[a]



Entry	Catalyst I (mol%)	Yield of 3a (%)[b]
1	1	62
2	3	**91 (>99)[b]**
3	**5**	**93**
4	10	88

Reaction conditions: **1a** (0.5 mmol), **2a** (1.1 mmol), Mn-catalyst and KO^{t}Bu (50 mol%) using 1 mL of toluene as solvent at 130 °C (oil-bath temperature). [a]Isolated yield. [b]GC conversion of benzohydrazides using mesitylene as an internal standard. NR= No reaction.

2.2. 1 mmol Scale Reaction of Symmetrical *N*,*N*-Dialkylation of Benzohydrazide (1a) Using 1-butanol (2a)



In an oven-dried screw cap reaction tube (15 mL), benzohydrazide 1a (1 mmol), 1-butanol (2.2 mmol), [Mn-1] (3 mol%), KO^{t}Bu (50 mol%), and dry toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 °C (oil-bath temperature) for 4 h. After completion of the reaction, the crude mixture was filtered through a celite filter and washed with ethyl acetate (3×5 mL), followed by the solvent was removed under vacuum, and finally, the residue was purified by silica gel column chromatography (230- 400 mesh size) using petroleum-ether and ethyl acetate as an eluent to give the *N*,*N*-dialkylated product **3a**. The reaction provided product **3a** in 113mg, 91% isolated yield.

2.3. General procedure for the manganese catalyzed one-pot symmetrical *N*,*N*-dialkylation

In an oven-dried screw cap reaction tube (15 mL), acyl hydrazide (0.5 mmol), alcohol (1.1 mmol), [Mn-1] (3 mol%), KO^{t}Bu (50 mol%), and dry toluene (2 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 ^{O}C (oil-bath temperature) for 4 h. After completion of the reaction, the crude mixture was filtered through a celite filter and washed with ethyl acetate (3×5 mL), followed by the solvent was removed under vacuum. Finally, the residue was purified by silica gel column chromatography (230- 400 mesh size) using petroleum ether and ethyl acetate as an eluent to give the *N*,*N*-dialkylated product. Yields were calculated for isolated pure products

2.4 General Procedure for *N*,*N*-Diethylation of Acyl hydrazides Using Ethanol

In an oven-dried screw cap reaction tube (15 mL), acyl hydrazide (0.5 mmol), ethanol (10 equiv.), [Mn-1] (3 mol%), KO^{t}Bu (50 mol%), and dry toluene (2 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 ^{O}C (oil-bath temperature) for 4 h. After completion of the reaction, the crude mixture was filtered through a celite filter and washed with ethyl acetate (3×5 mL), followed by the solvent was removed under vacuum, and finally, the residue was purified by silica gel column chromatography (230- 400 mesh size) using petroleum-ether and ethyl acetate as an eluent to give the *N*,*N*-diethylated product. Yields were calculated for isolated pure products.

2.5 General Procedure for *N*,*N*-Dimethylation of Acyl hydrazides Using Methanol

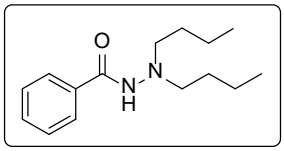
In an oven-dried screw cap reaction tube (15 mL), acyl hydrazide (0.5 mmol), dry methanol (1 mL), [Mn-1] (3 mol%), KO^{t}Bu (50 mol%), and dry toluene (1 mL) were added in a gentle stream of argon. Then the reaction mixture was stirred with a magnetic stirring bar at 130 ^{O}C (oil-bath temperature) for 4 h. After completion of the reaction the crude mixture was filtered through a celite filter and washed with ethyl acetate (3×5 mL), followed by the solvent was removed under vacuum, and finally, the residue was purified by silica gel column chromatography (230- 400 mesh size) using petroleum-ether and ethyl acetate as an eluent to give the *N*,*N*-diethylated product. Yields were calculated for isolated pure products.

2.6. General Procedure for Manganese Catalyzed Unsymmetrical *N*,*N*-Dialkylation of Acylhydrazides Using Alcohols

An oven-dried screw cap reaction tube (15 mL) was equipped with a stir bar, [Mn-1] (3 mol%), KO^{t}Bu (0.5 mmol), acylhydrazide (0.5 mmol, 1 equiv.), alcohol (0.55 mmol, 1.1 equiv.) and dry toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 ^{o}C (oil-bath temperature) for 4 h. After 4 hours, [Mn-1] (3 mol%), KO^{t}Bu (0.5 mmol), another alcohol (0.55 mmol, 1.1 equiv) and toluene (1 mL) were taken in a separate vial, and the solution was added inside the reaction mixture under argon atmosphere. Further, the reaction continued for another 4 hours, and the completion of the reaction was monitored using TLC analysis. After completion of the reaction, the crude mixture was filtered through a celite filter and washed with ethyl acetate (3×5 mL), followed by the solvent was removed under vacuum, and finally, the residue was purified by silica gel column chromatography (230- 400 mesh size) using petroleum-ether and ethyl acetate as an eluent to give the *N*,*N*-diethylated product. Yields were calculated for isolated pure products.

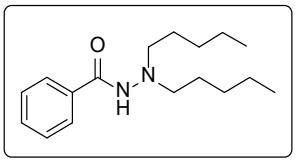
3. Characterization

N',N'-dibutylbenzohydrazide (3a)



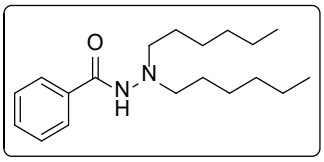
The title compound was prepared according to the general procedure and isolated as a white solid (113 mg, 91% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.74 (d, *J* = 7.3 Hz, 2 H), 7.57 - 7.32 (m, 3 H), 6.69 (br. s, 1 H), 2.83 (t, *J* = 7.4 Hz, 4 H), 1.64 - 1.48 (m, 4 H), 1.43 - 1.32 (m, 4 H), 0.90 (t, *J* = 7.3 Hz, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.5, 128.6, 127.0, 58.2, 29.2, 20.4, 14.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{15}H_{25}N_2O$ 249.1961; Found 249.1959.

N',N'-dipentylbenzohydrazide (3b)



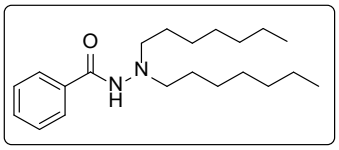
The title compound was prepared according to the general procedure and isolated as a white solid (129 mg, 93% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.29 (br. s, 3 H), 7.00 (d, *J* = 7.1 Hz, 3 H), 4.09 - 3.99 (m, 4 H), 2.99 (br. s, 4 H), 1.90 (br. s, 8 H), 1.41 (t, *J* = 6.9 Hz, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 159.1, 129.5, 118.7, 118.1, 113.1, 63.7, 55.5, 22.3, 14.8. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{17}H_{29}N_2O$ 277.2266; Found 277.2265.

N',N'-dihexylbenzohydrazide (3c)



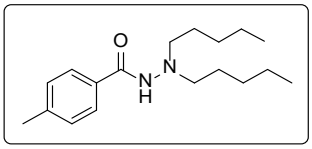
The title compound was prepared according to the general procedure and isolated as a white solid (143 mg, 94% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.73 (d, *J* = 7.1 Hz, 2 H), 7.52 - 7.32 (m, 3 H), 6.81 (br. s, 1 H), 2.82 (t, *J* = 7.5 Hz, 4 H), 1.66 - 1.44 (m, 4 H), 1.28 (br. s, 12 H), 0.99 - 0.77 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.4, 128.5, 127.0, 58.4, 31.7, 27.0, 26.9, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{19}H_{33}N_2O$ 305.2587; Found 305.2583.

N',N'-diheptylbenzohydrazide (3d)



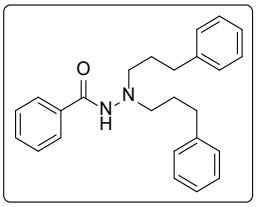
The title compound was prepared according to the general procedure and isolated as a white solid (158 mg, 95% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.73 (d, *J* = 7.3 Hz, 2 H), 7.55 - 7.36 (m, 3 H), 6.67 (s, 1 H), 2.82 (t, *J* = 7.5 Hz, 4 H), 1.70 - 1.43 (m, 4 H), 1.37 - 1.17 (m, 16 H), 0.94 - 0.77 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.5, 128.6, 127.0, 58.5, 31.8, 29.2, 27.2, 27.1, 22.6, 14.1. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{21}H_{37}N_2O$ 333.2900; Found 333.2897.

4-methyl-N',N'-dipentylbenzohydrazide (3e)



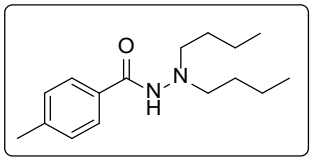
The title compound was prepared according to the general procedure and isolated as a white solid (140 mg, 95% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.63 (d, *J* = 7.6 Hz, 2 H), 7.33 - 7.07 (m, 2 H), 6.54 (br. s, 1 H), 2.81 (br. s, 4 H), 2.39 (s, 3 H), 1.57 (br. s, 4 H), 1.30 (br. s, 8 H), 0.88 (br. s, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 168.8, 141.9, 131.2, 129.3, 126.9, 58.5, 29.4, 26.7, 22.6, 21.4, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{18}H_{31}N_2O$ 291.2431; Found 291.2428.

N',N'-bis(3-phenylpropyl)benzohydrazide (3f)



The title compound was prepared according to the general procedure and isolated as a white solid (164 mg, 88% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.58 (d, *J* = 7.6 Hz, 2 H), 7.45 - 7.31 (m, 1 H), 7.31 - 7.22 (m, 2 H), 7.22 - 7.01 (m, 10 H), 6.68 (br. s, 1 H), 3.53 (t, *J* = 6.5 Hz, 1 H), 2.73 (t, *J* = 7.3 Hz, 3 H), 2.68 - 2.48 (m, 4 H), 1.86 - 1.64 (m, 4 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.9, 142.1, 134.0, 131.6, 128.7, 128.6, 128.4, 127.0, 125.9, 57.7, 33.4, 28.9. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{25}H_{29}N_2O$ 373.2267; Found 373.22.

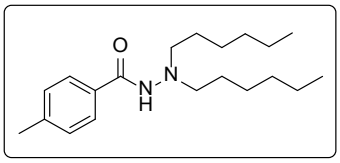
N',N'-dibutyl-4-methylbenzohydrazide (3g)



The title compound was prepared according to the general procedure and isolated as a white solid (122 mg, 93% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.73 - 7.48 (m, *J* = 8.0 Hz, 2 H), 7.28 - 7.11 (m, *J* = 7.9 Hz, 2 H), 6.70 (br. s, 1 H), 2.82 (t, *J* =

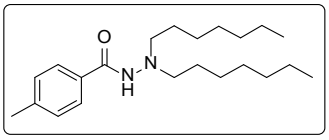
7.5 Hz, 4 H), 2.38 (s, 3 H), 1.55 (t, *J* = 7.3 Hz, 4 H), 1.43 - 1.26 (m, 4 H), 0.90 (t, *J* = 7.3 Hz, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.6, 141.9, 131.2, 129.2, 127.0, 58.2, 29.2, 21.4, 20.4, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{16}H_{27}N_2O$ 263.2114; Found 263.2118.

N',N'-dihexyl-4-methylbenzohydrazide (3h)



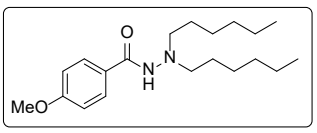
The title compound was prepared according to the general procedure and isolated as a white solid (151 mg, 95% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.73 - 7.48 (m, *J* = 7.8 Hz, 2 H), 7.25 - 7.06 (m, *J* = 7.6 Hz, 2 H), 6.70 (br. s, 1 H), 2.81 (t, *J* = 7.3 Hz, 4 H), 2.38 (s, 3 H), 1.71 - 1.45 (m, 4 H), 1.27 (br. s, 12 H), 0.95 - 0.78 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.6, 141.8, 131.2, 129.2, 127.0, 58.5, 31.7, 27.0, 26.9, 22.6, 21.4, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{20}H_{35}N_2O$ 319.2744; Found 319.2736.

N',N'-diheptylbenzohydrazide (3i)



The title compound was prepared according to the general procedure and isolated as a white solid (168 mg, 97% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.73 - 7.52 (m, *J* = 8.0 Hz, 2 H), 7.34 - 7.08 (m, *J* = 7.9 Hz, 2 H), 6.79 (s, 1 H), 2.81 (t, *J* = 7.6 Hz, 4 H), 2.38 (s, 3 H), 1.56 (br. s, 4 H), 1.36 - 1.16 (m, 16 H), 0.94 - 0.77 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.6, 141.8, 131.2, 129.2, 127.0, 58.4, 31.8, 29.2, 27.2, 27.1, 22.6, 21.4, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{22}H_{39}N_2O$ 347.3057; Found 347.3045.

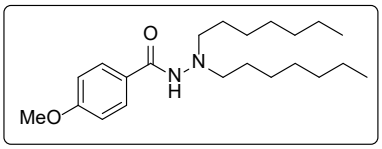
N',N'-dihexyl-4-methoxybenzohydrazide (3j)



The title compound was prepared according to the general procedure and isolated as a white solid (162 mg, 97% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ =

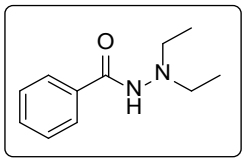
7.71 (d, *J* = 8.6 Hz, 2 H), 6.91 (d, *J* = 8.8 Hz, 2 H), 6.66 (br. s, 1 H), 3.84 (s, 3 H), 2.81 (t, *J* = 7.5 Hz, 4 H), 1.66 - 1.43 (m, 4 H), 1.38 - 1.10 (m, 12 H), 0.94 - 0.69 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 162.2, 128.7, 126.3, 113.8, 58.5, 55.4, 31.7, 27.0, 27.0, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{19}H_{33}N_2O$ 335.2693; Found 335.2690.

N',N'-diheptyl-4-methoxybenzohydrazide (3k)



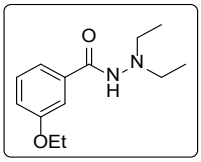
The title compound was prepared according to the general procedure and isolated as a white solid (180 mg, 99% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.84 - 7.58 (m, *J* = 8.6 Hz, 2 H), 7.00 - 6.77 (m, *J* = 8.6 Hz, 2 H), 6.57 (br. s, 1 H), 3.84 (s, 3 H), 2.81 (t, *J* = 7.3 Hz, 4 H), 1.56 (br. s, 4 H), 1.39 - 1.21 (m, 16 H), 1.04 - 0.76 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 162.2, 128.7, 126.3, 113.8, 58.5, 55.4, 31.8, 29.2, 27.2, 27.1, 22.6, 14.1. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{22}H_{39}N_2O_2N_2$ 363.3000; Found 363.3006.

N',N'-diethylbenzohydrazide (3l)



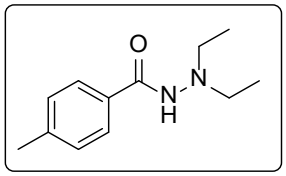
The title compound was prepared according to the general procedure and isolated as a white solid (89 mg, 93% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.76 (br. s, 2 H), 7.61 - 7.46 (m, 1 H), 7.46 - 7.21 (m, 2 H), 6.74 (br. s, 1 H), 3.05 - 2.78 (m, 4 H), 1.31 - 1.07 (m, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 167.1, 134.0, 131.5, 128.6, 127.0, 52.3, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{17}N_2O$ 193.1335; Found 193.1331.

3-ethoxy-N',N'-diethylbenzohydrazide (3m)



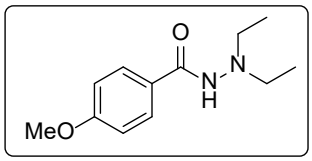
The title compound was prepared according to the general procedure and isolated as a white solid (115 mg, 97% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.40 - 7.10 (m, 3 H), 6.98 - 6.85 (m, 1 H), 6.71 (br. s, 1 H), 3.97 (q, *J* = 6.9 Hz, 2 H), 2.79 (q, *J* = 6.9 Hz, 4 H), 1.32 (t, *J* = 6.9 Hz, 3 H), 1.06 (t, *J* = 7.1 Hz, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.0, 158.1, 134.3, 128.5, 117.7, 117.0, 112.1, 62.6, 51.2, 13.7, 11.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{21}N_2O_2$ 237.1659; Found 237.1657.

N',N'-diethyl-4-methylbenzohydrazide (3n)



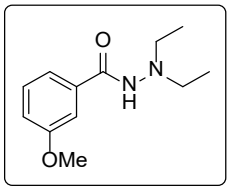
The title compound was prepared according to the general procedure and isolated as a white solid (98 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 7.9 Hz, 2 H), 7.21 (d, *J* = 7.9 Hz, 2 H), 6.65 (br. s, 1 H), 2.87 (q, *J* = 7.0 Hz, 4 H), 2.38 (s, 3 H), 1.15 (t, *J* = 7.1 Hz, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 167.0, 141.9, 131.1, 129.2, 127.0, 52.3, 21.4, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{19}N_2O$ 207.1492; Found 207.1487.

N',N'-diethyl-4-methoxybenzohydrazide (3o)



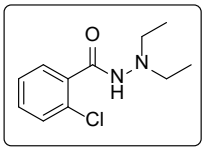
The title compound was prepared according to the general procedure and isolated as a white solid (109 mg, 98% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 8.6 Hz, 2 H), 6.82 (d, *J* = 8.8 Hz, 2 H), 6.60 (br. s, 1 H), 3.75 (s, 3 H), 2.79 (q, *J* = 6.9 Hz, 4 H), 1.07 (t, *J* = 7.1 Hz, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.6, 162.2, 128.8, 126.1, 113.7, 55.4, 52.3, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{19}N_2O_2$ 223.1441; Found 223.1440.

N',N'-diethyl-3-methoxybenzohydrazide (3p)



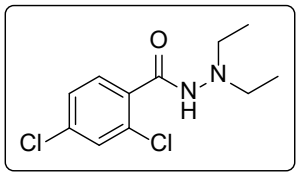
The title compound was prepared according to the general procedure and isolated as a white solid (108 mg, 97% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.51 - 7.22 (m, 3 H), 7.04 (d, *J* = 7.8 Hz, 1 H), 6.69 (br. s, 1 H), 3.94 - 3.75 (m, 3 H), 2.88 (d, *J* = 6.9 Hz, 4 H), 1.25 - 1.10 (m, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.9, 159.8, 135.4, 129.6, 118.6, 117.7, 112.5, 55.5, 52.2, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{19}N_2O_2$ 223.1441; Found 223.1441.

2-chloro-N',N'-diethylbenzohydrazide (3q)



The title compound was prepared according to the general procedure and isolated as a white solid (99 mg, 87% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.75 - 7.54 (m, *J* = 8.4 Hz, 2 H), 7.38 - 7.18 (m, 2 H), 6.80 (br. s, 1 H), 2.81 (q, *J* = 6.9 Hz, 4 H), 1.06 (t, *J* = 7.1 Hz, 6 H) ppm. ^{13}C NMR (100MHz, $CDCl_3$) δ = 166.1, 137.7, 132.3, 128.8, 128.7, 128.6, 128.5, 52.1, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{16}N_2OCl$ 227.0940; Found 227.0946.

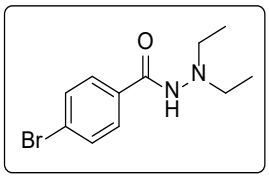
2,4-dichloro-N',N'-diethylbenzohydrazide (3r)



The title compound was prepared according to the general procedure and isolated as a white solid (108 mg, 83% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.54 (d, *J* = 8.3 Hz, 1 H), 7.42 (d, *J* = 1.8 Hz, 1 H), 7.35 - 7.26 (m, 1 H), 6.56 (br. s, 1 H), 2.88 (q, *J* = 6.7 Hz, 4 H), 1.20 (t, *J* = 7.1 Hz, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.0,

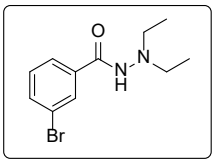
136.8, 133.0, 131.1, 129.9, 127.5, 126.4, 52.1, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{15}N_2OCl_2$ 261.0556; Found 261.0558.

4 -bromo-N',N'-diethylbenzohydrazide (3s)



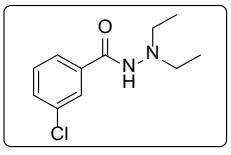
The title compound was prepared according to the general procedure and isolated as a white solid (111 mg, 82% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.83 (br. s, 1 H), 7.71 - 7.45 (m, 2 H), 7.30 - 7.11 (m, 1 H), 6.90 (br. s, 1 H), 2.82 (d, *J* = 6.8 Hz, 4 H), 1.07 (t, *J* = 6.9 Hz, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.7, 134.5, 130.1, 125.6, 122.8, 52.1, 12.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{16}N_2OBr$ 271.0441; Found 271.0441.

3-bromo-N',N'-diethylbenzohydrazide (3t)



The title compound was prepared according to the general procedure and isolated as a white solid (114 mg, 84% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.65 - 7.50 (m, *J* = 8.1 Hz, 2 H), 7.50 - 7.37 (m, *J* = 8.0 Hz, 2 H), 6.78 (br. s, 1 H), 2.81 (d, *J* = 6.8 Hz, 4 H), 1.07 (t, *J* = 6.9 Hz, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 132.7, 131.8, 131.1, 130.8, 128.7, 126.1, 52.1, 12.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{16}N_2OBr$ 271.0441; Found 271.0440.

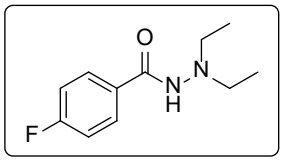
3-chloro-N',N'-diethylbenzohydrazide (3u)



The title compound was prepared according to the general procedure and isolated as a white solid (101 mg, 89% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ =

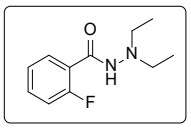
7.75 (br. s, 1 H), 7.64 (d, *J* = 7.3 Hz, 1 H), 7.47 (d, *J* = 7.6 Hz, 1 H), 7.36 (t, *J* = 7.8 Hz, 1 H), 3.02 - 2.78 (m, 4 H), 1.16 (br. s, 6 H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.8, 135.7, 134.8, 131.6, 130.8, 129.9, 127.5, 127.4, 125.1, 77.4, 77.3, 77.1, 76.7, 52.1, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{16}N_2OCl$ 227.0946; Found 227.0944.

N',N'-diethyl-4-fluorobenzohydrazide (3v)



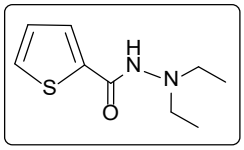
The title compound was prepared according to the general procedure and isolated as a white solid (86 mg, 82% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.85 - 7.57 (m, 2 H), 7.00 (t, *J* = 8.5 Hz, 2 H), 6.82 (br. s, 1 H), 2.80 (q, *J* = 6.4 Hz, 4 H), 1.06 (t, *J* = 7.0 Hz, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.9, 163.4, 130.1, 129.3, 115.7, 52.1, 12.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{16}N_2OF$ 211.1241; Found 211.1241.

N',N'-diethyl-2-fluorobenzohydrazide (3w)



The title compound was prepared according to the general procedure and isolated as a white solid (86 mg, 80% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.79 (s, 1 H), 7.41 (s, 1 H), 7.24 (s, 1 H), 7.07 (s, 1 H), 4.21 (q, *J* = 7.2 Hz, 4 H), 1.45 (t, *J* = 7.2 Hz, 6 H); ^{13}C NMR (100 Hz, $CDCl_3$) δ = 156.7, 141.3, 128.5, 121.3, 119.3, 112.8, 108.8, 43.1, 14.5. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{16}N_2OF$ 211.1241; Found 211.1240.

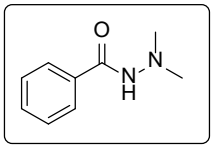
N',N'-diethylthiophene-2-carbohydrazide (3x)



The title compound was prepared according to the general procedure and isolated as a white solid (84 mg, 80% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 8.03 (d, *J* = 2.4 Hz, 1 H), 7.45 (d, *J* = 3.9 Hz, 1 H), 7.00 (dd, *J* = 3.9, 4.8 Hz, 1 H), 6.62 (br. s,

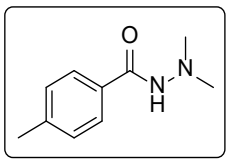
1 H), 2.99 - 2.74 (m, 2 H), 2.60 (dd, *J* = 6.9, 12.4 Hz, 2 H), 1.12 - 0.99 (m, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.8, 135.3, 133.5, 132.6, 126.0, 52.4, 11.2. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_9H_{15}N_2OS$ 199.0900; Found 199.0895.

N',N'-dimethylbenzohydrazide (3y)



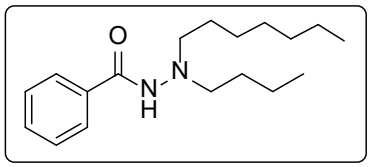
The title compound was prepared according to the general procedure and isolated as a white solid (76 mg, 92% yield). Petroleum ether/EtOAc = 60:40. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.74 (d, *J* = 7.3 Hz, 2 H), 7.57 - 7.34 (m, 3 H), 2.71 (br. s, 6 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.78, 133.7, 131.6, 128.6, 127.1, 77.4, 77.1, 76.8, 47.5 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_9H_{13}N_2O$ 165.1022; Found 165.1023.

N',N',4-trimethylbenzohydrazide (3z)



The title compound was prepared according to the general procedure and isolated as a white solid (95 mg, 85% yield). Petroleum ether/EtOAc = 60:40. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.84 - 7.55 (m, *J* = 7.6 Hz, 2 H), 7.28 - 6.98 (m, 2 H), 6.81 (br. s, 1 H), 2.71 (br. s, 6 H), 2.39 (s, 3 H) ppm; ^{13}C NMR (100 MHz, CHLOROFORM-d) d = 165.7, 142.1, 130.8, 129.2, 127.0, 47.7, 21.5 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{10}H_{15}N_2O$ 179.1175; Found 179.1179.

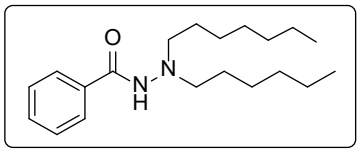
N'-butyl-N'-heptylbenzohydrazide (4a)



The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.70 - 7.58 (m, 2 H), 7.38 (d, *J* = 7.4 Hz, 1 H), 7.34 - 7.26 (m, 2 H), 6.87 (s, 1 H), 2.79 - 2.65 (m, 4 H), 1.52 - 1.36 (m, 4 H), 1.30 - 1.14 (m, 10 H), 0.82 - 0.74 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.4,

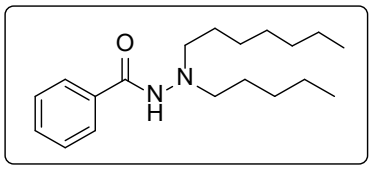
128.5, 127.5, 127.0, 58.5, 58.1, 31.8, 29.2, 27.2, 22.6, 20.4, 14.1. Petroleum ether/EtOAc = 70:30. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{18}H_{31}N_2O$ 291.2431; Found 291.2440.

N'-heptyl-N'-hexylbenzohydrazide (4b)



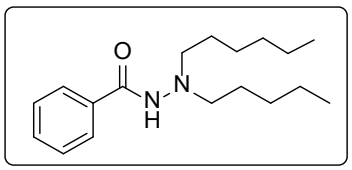
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.68 - 7.58 (m, 2 H), 7.39 (d, *J* = 7.4 Hz, 1 H), 7.36 - 7.25 (m, 2 H), 6.77 (s, 1 H), 2.86 - 2.63 (m, 4 H), 1.48 (t, *J* = 7.4 Hz, 4 H), 1.26 - 1.16 (m, 14 H), 0.81 - 0.75 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.4, 128.5, 127.0, 58.5, 31.8, 29.2, 27.2, 27.1, 26.9, 22.6, 14.1. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{20}H_{35}N_2O$ 319.4556; Found 319.4559.

N'-heptyl-N'-pentylbenzohydrazide (4c)



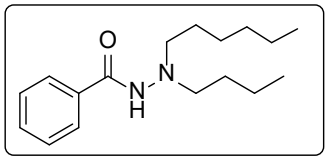
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 7.3 Hz, 2 H), 7.40 (d, *J* = 6.9 Hz, 1 H), 7.37 - 7.25 (m, 2 H), 6.73 (br. s, 1 H), 2.77 - 2.71 (m, 4 H), 1.53 - 1.44 (m, 4 H), 1.26 - 1.15 (m, 12 H), 0.79 (d, *J* = 6.4 Hz, 5 H), 0.82 - 0.75 (m, 1 H); ^{13}C NMR (100 MHz $CDCl_3$) δ = 166.7, 134.1, 131.4, 129.2, 128.5, 127.5, 127.0, 58.4, 58.4, 31.8, 29.4, 29.2, 27.2, 27.1, 26.7, 22.6, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{19}H_{33}N_2O$ 305.2587; Found 305.2598.

N'-hexyl-N'-pentylbenzohydrazide (4d)



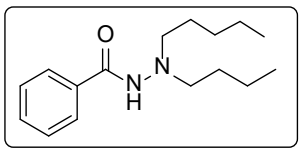
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.70 - 7.60 (m, 2 H), 7.47 - 7.37 (m, 1 H), 7.37 - 7.25 (m, 2 H), 6.72 (br. s, 1 H), 2.81 - 2.66 (m, 4 H), 1.58 - 1.42 (m, 4 H), 1.27 - 1.17 (m, 10 H), 0.83 - 0.75 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.4, 128.6, 127.0, 58.5, 31.7, 29.4, 27.0, 26.9, 26.7, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{18}H_{31}N_2O$ 291.2431; Found 291.2439.

N'-butyl-N'-hexylbenzohydrazide (4e)



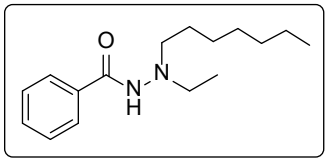
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 7.9 Hz, 2 H), 7.45 - 7.37 (m, 1 H), 7.37 - 7.28 (m, 2 H), 6.73 (br. s, 1 H), 2.87 - 2.65 (m, 4 H), 1.63 - 1.37 (m, 4 H), 1.33 - 1.17 (m, 8 H), 0.85 - 0.75 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.1, 131.5, 128.6, 127.0, 58.5, 31.7, 29.1, 26.9, 22.6, 20.4, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{17}H_{29}N_2O$ 277.2274; Found 277.2278.

N'-butyl-N'-pentylbenzohydrazide (4f)



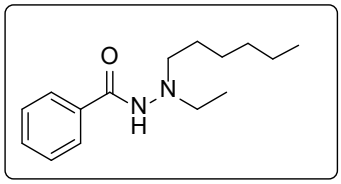
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.71 - 7.60 (m, 2 H), 7.47 - 7.40 (m, 1 H), 7.39 - 7.27 (m, 2 H), 6.57 (br. s, 1 H), 2.84 - 2.67 (m, 4 H), 1.59 - 1.41 (m, 4 H), 1.34 - 1.22 (m, 6 H), 0.86 - 0.77 (m, 6 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.7, 134.0, 131.6, 128.7, 127.0, 58.5, 58.2, 29.4, 26.7, 22.6, 20.4, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{16}H_{27}N_2O$ 263.2118; Found 263.2108.

N'-ethyl-N'-heptylbenzohydrazide (4g)



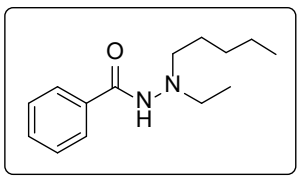
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 8.0 Hz, 2 H), 7.47 - 7.36 (m, 1 H), 7.36 - 7.28 (m, 2 H), 6.76 (br. s, 1 H), 2.80 (q, *J* = 7.0 Hz, 2 H), 2.75 - 2.65 (m, 2 H), 1.56 - 1.41 (m, 2 H), 1.27 - 1.15 (m, 8 H), 1.09 - 1.04 (m, 3 H), 0.80 - 0.74 (m, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.9, 134.0, 131.4, 128.5, 127.0, 58.3, 52.4, 31.7, 29.2, 27.2, 27.0, 22.6, 14.0, 12.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{16}H_{27}N_2O$ 263.2118; Found 263.2109.

N'-ethyl-N'-hexylbenzohydrazide (4h)



The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 7.8 Hz, 2 H), 7.45 - 7.36 (m, 1 H), 7.35 - 7.26 (m, 2 H), 6.78 (br. s, 1 H), 2.80 (q, *J* = 7.0 Hz, 2 H), 2.76 - 2.66 (m, 2 H), 1.55 - 1.44 (m, 2 H), 1.28 - 1.16 (m, 6 H), 1.06 (t, *J* = 7.1 Hz, 3 H), 0.78 (t, *J* = 6.6 Hz, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.9, 134.0, 131.5, 128.6, 127.3, 127.0, 58.3, 52.5, 31.7, 27.0, 22.6, 14.0, 12.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{15}H_{25}N_2O$ 249.1961; Found 249.1953.

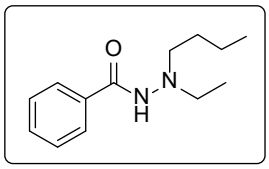
N'-ethyl-N'-pentylbenzohydrazide (4i)



The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.71 - 7.61 (m, 2 H), 7.43 - 7.35 (m, 1 H), 7.35 - 7.26 (m, 2 H), 6.86 (br. s, 1 H), 2.80 (q, *J* = 7.1 Hz, 2 H), 2.75 - 2.68 (m, 2 H), 1.54 - 1.42 (m, 2 H), 1.26 - 1.18 (m, 4 H), 1.05 (t, *J* = 7.1 Hz, 3 H), 0.82 - 0.77 (m, 3 H); ^{13}C

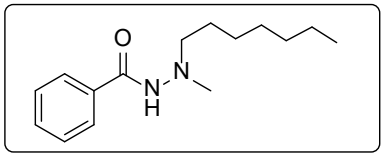
NMR (100 MHz, $CDCl_3$) δ = 166.9, 134.0, 131.4, 128.5, 127.0, 58.3, 52.4, 29.4, 26.7, 22.6, 14.0, 12.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{14}H_{23}N_2O$ 235.1805; Found 235.1796.

N'-butyl-N'-ethylbenzohydrazide (4j)



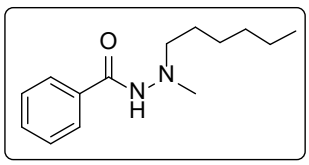
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^1H NMR (400 MHz, $CDCl_3$) δ = 7.67 (d, *J* = 7.8 Hz, 2 H), 7.40 (d, *J* = 7.4 Hz, 1 H), 7.37 - 7.28 (m, 2 H), 6.77 (br. s, 1 H), 2.81 (q, *J* = 7.1 Hz, 2 H), 2.77 - 2.70 (m, 2 H), 1.53 - 1.42 (m, 2 H), 1.33 - 1.21 (m, 2 H), 1.06 (t, *J* = 7.1 Hz, 3 H), 0.82 (t, *J* = 7.3 Hz, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.9, 134.0, 131.4, 128.5, 127.0, 57.9, 52.4, 29.1, 20.4, 14.0, 12.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{21}N_2O$ 221.1654; Found 221.1648.

N'-heptyl-N'-methylbenzohydrazide (4k)



The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^1H NMR (400 MHz, $CDCl_3$) δ = 7.71 - 7.59 (m, 2 H), 7.44 - 7.35 (m, 1 H), 7.35 - 7.26 (m, 2 H), 7.00 (br. s, 1 H), 2.74 - 2.66 (m, 2 H), 2.63 (s, 3 H), 1.48 (quin, *J* = 7.4 Hz, 2 H), 1.29 - 1.12 (m, 8 H), 0.82 - 0.74 (m, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.5, 127.1, 59.8, 46.1, 31.7, 29.2, 27.2, 27.1, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{15}H_{25}N_2O$ 249.3467; Found 249.3464.

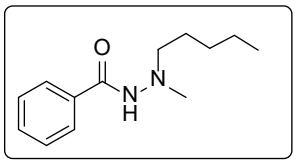
N'-hexyl-N'-methylbenzohydrazide (4l)



The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^1H NMR (400 MHz, $CDCl_3$) δ = 7.75 - 7.61 (m, 2 H), 7.40 (d, *J* =

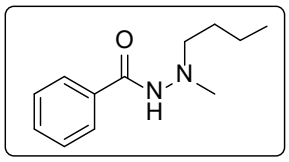
7.4 Hz, 1 H), 7.38 - 7.30 (m, 2 H), 6.86 (br. s, 1 H), 2.77 - 2.67 (m, 2 H), 2.64 (s, 3 H), 1.48 (t, J = 7.6 Hz, 2 H), 1.32 - 1.14 (m, 6 H), 0.79 (t, J = 6.8 Hz, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.5, 127.0, 59.8, 46.2, 31.7, 27.2, 26.9, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{14}H_{22}N_2O$ 235.3478; Found 235.3474.

N'-methyl-N'-pentylbenzohydrazide (4m)



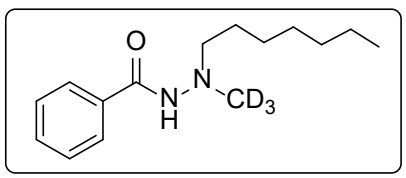
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.71 - 7.63 (m, 2 H), 7.42 - 7.33 (m, 1 H), 7.33 - 7.25 (m, 2 H), 7.19 (br. s, 1 H), 2.73 - 2.63 (m, 2 H), 2.60 (s, 3 H), 1.54 - 1.38 (m, 2 H), 1.25 - 1.17 (m, 4 H), 0.81 - 0.76 (m, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.4, 128.4, 127.1, 59.7, 46.0, 29.3, 26.9, 22.5, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{21}N_2O$ 221.1648; Found 221.1650.

N'-butyl-N'-methylbenzohydrazide (4n)



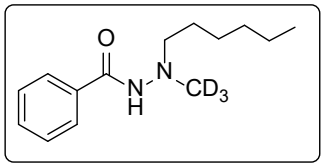
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.73 - 7.61 (m, 2 H), 7.40 (d, J = 7.4 Hz, 1 H), 7.35 - 7.26 (m, 2 H), 6.97 (br. s, 1 H), 2.71 (t, J = 7.5 Hz, 2 H), 2.63 (s, 3 H), 1.52 - 1.38 (m, 2 H), 1.35 - 1.20 (m, 2 H), 0.82 (t, J = 7.4 Hz, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.5, 127.1, 59.5, 46.2, 29.3, 20.3, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{19}N_2O$ 207.1356; Found 207.1354.

N'-heptyl-N'-(methyl-d3)benzohydrazide (4o)



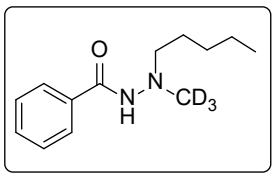
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.66 (d, *J* = 7.8 Hz, 2 H), 7.43 - 7.36 (m, 1 H), 7.35 - 7.27 (m, 2 H), 6.98 (br. s, 1 H), 2.69 (t, *J* = 7.5 Hz, 2 H), 1.47 (quin, *J* = 7.0 Hz, 2 H), 1.30 - 1.10 (m, 8 H), 0.78 (t, *J* = 6.3 Hz, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.6, 127.0, 59.8, 31.8, 29.2, 27.2, 27.1, 22.6, 14.1. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{15}H_{22}{}^{2}H_3N_2O$ 252.3987; Found 252.3983.

N'-hexyl-N'-(methyl-d3)benzohydrazide (4p)



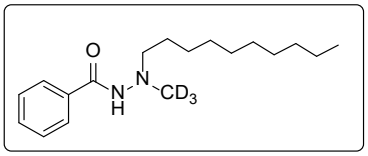
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz $CDCl_3$) δ = 7.71 - 7.62 (m, 2 H), 7.40 (d, *J* = 7.4 Hz, 1 H), 7.36 - 7.27 (m, 2 H), 6.91 (br. s, 1 H), 2.77 - 2.65 (m, 2 H), 1.48 (t, *J* = 7.6 Hz, 2 H), 1.32 - 1.16 (m, 6 H), 0.79 (t, *J* = 6.8 Hz, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.5, 127.0, 59.7, 31.7, 27.2, 26.9, 22.6, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{14}H_{20}{}^{2}H_3N_2O$ 238.1993; Found 238.1996.

N'-(methyl-d3)-N'-pentylbenzohydrazide (4q)



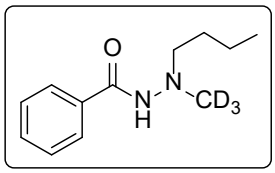
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.71 - 7.63 (m, 2 H), 7.43 - 7.36 (m, 1 H), 7.36 - 7.26 (m, 2 H), 6.90 (br. s, 1 H), 2.76 - 2.65 (m, 2 H), 1.58 - 1.41 (m, 2 H), 1.30 - 1.18 (m, 4 H), 0.84 - 0.75 (m, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.6, 127.0, 59.7, 29.3, 26.8, 22.5, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{18}{}^{2}H_3N_2O$ 224.1873; Found 224.1879.

N'-decyl-N'-(methyl-d3)benzohydrazide (4r)



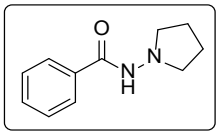
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.70 - 7.63 (m, 2 H), 7.48 - 7.39 (m, 1 H), 7.39 - 7.31 (m, 2 H), 6.61 (br. s, 1 H), 2.77 - 2.67 (m, 2 H), 1.55 - 1.41 (m, 2 H), 1.30 - 1.16 (m, 14 H), 0.85 - 0.75 (m, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.1, 133.6, 131.7, 128.6, 127.1, 59.8, 31.9, 29.7, 29.5, 29.3, 27.1, 27.0, 22.7, 22.67, 14.1. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{18}H_{28}{}^2H_3N_2O$ 294.4773; Found 294.4779.

N'-butyl-N'-(methyl-d3)benzohydrazide (4s)



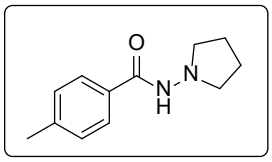
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.71 - 7.63 (m, 2 H), 7.43 - 7.36 (m, 1 H), 7.36 - 7.26 (m, 2 H), 6.90 (br. s, 1 H), 2.76 - 2.65 (m, 2 H), 1.58 - 1.41 (m, 2 H), 1.30 - 1.18 (m, 4 H), 0.84 - 0.75 (m, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 133.9, 131.5, 128.6, 127.0, 59.7, 29.3, 26.8, 22.5, 14.0. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{16}{}^2H_3N_2O$ 210.1690; Found 210.1695.

N-(pyrrolidin-1-yl)benzamide (5a)



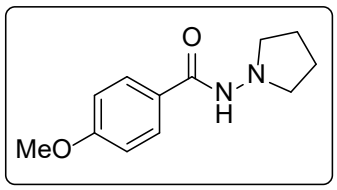
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 95% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.74 (d, *J* = 7.3 Hz, 2 H), 7.54 - 7.39 (m, 3 H), 6.99 (br. s, 1 H), 3.01 (br. s, 4 H), 1.90 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.3, 133.9, 131.5, 128.5, 127.1, 55.5, 22.3 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{15}N_2O$ 191.1179; Found 191.1178.

4-methyl-N-(pyrrolidin-1-yl)benzamide (5b)



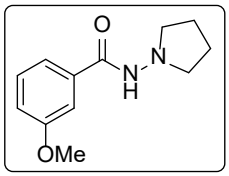
The title compound was prepared according to the general procedure and isolated as a white solid (98 mg, 96% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.64 (d, *J* = 7.5 Hz, 2 H), 7.19 (d, *J* = 7.8 Hz, 2 H), 7.03 (br. s, 1 H), 2.99 (br. s, 4 H), 2.38 (s, 3 H), 1.89 (br. s, 4 H) ppm; ^{13}C NMR (100mMHz, $CDCl_3$) δ = 166.2, 141.8, 131.0, 129.1, 127.0, 55.5, 22.2, 21.4 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{17}N_2O$ 205.1335; Found 205.1332.

4-methoxy-N-(pyrrolidin-1-yl)benzamide (5c)



The title compound was prepared according to the general procedure and isolated as a white solid (108 mg, 98% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.81 - 7.57 (m, 2 H), 6.90 (d, *J* = 8.4 Hz, 2 H), 6.82 (br. s, 1 H), 3.84 (s, 3 H), 2.99 (br. s, 4 H), 1.90 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.8, 162.2, 128.8, 126.1, 113.7, 76.7, 55.6, 55.4, 22.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{17}N_2O_2$ 221.1276; Found 221.1274.

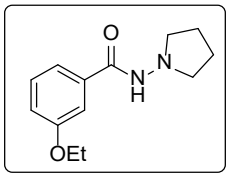
3-methoxy-N-(pyrrolidin-1-yl)benzamide (5d)



The title compound was prepared according to the general procedure and isolated as a white solid (106 mg, 96% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.38 - 7.12 (m, 3 H), 6.95 (d, *J* = 6.6 Hz, 1 H), 3.76 (s, 3 H), 2.93 (br. s, 4 H), 1.83 (br. s, 4 H)

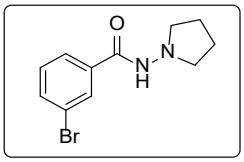
ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.2, 159.8, 135.2, 129.6, 118.8, 117.7, 112.5, 55.5, 30.0, 22.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{17}N_2O_2$ 221.1282; Found 221.1285.

3-ethoxy-N-(pyrrolidin-1-yl)benzamide (5e)



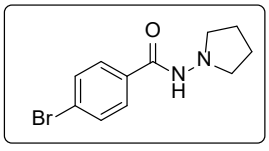
The title compound was prepared according to the general procedure and isolated as a white solid (110 mg, 94% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400MHz, $CDCl_3$) δ = 7.34 - 7.23 (m, 3 H), 7.00 (d, *J* = 7.1 Hz, 1 H), 4.06 (q, *J* = 6.9 Hz, 2 H), 2.99 (br. s, 4 H), 1.90 (br. s, 4 H), 1.41 (t, *J* = 6.9 Hz, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 166.1, 159.1, 135.3, 129.5, 118.7, 118.1, 113.1, 63.7, 55.5, 22.3, 14.8 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{19}N_2O_2$ 235.1441; Found 235.1439.

3-bromo-N-(pyrrolidin-1-yl)benzamide (5f)



The title compound was prepared according to the general procedure and isolated as a white solid (113 mg, 84% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.90 (br. s, 1 H), 7.67 (br. s, 1 H), 7.59 (d, *J* = 7.5 Hz, 1 H), 7.28 (d, *J* = 8.6 Hz, 1 H), 3.00 (br. s, 4 H), 1.89 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.9, 135.8, 134.8, 134.4, 130.4, 130.1, 125.8, 122.7, 55.4, 22.3 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{14}N_2OBr$ 269.0284; Found 269.0285.

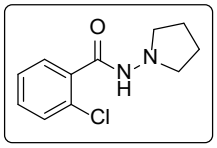
4-bromo-N-(pyrrolidin-1-yl)benzamide (5g)



The title compound was prepared according to the general procedure and isolated as a white solid (116 mg, 86% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ =

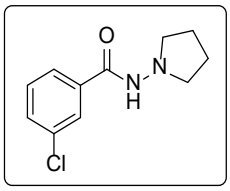
7.55 (d, *J* = 8.1 Hz, 2 H), 7.42 (d, *J* = 8.4 Hz, 2 H), 2.90 (br. s, 4 H), 1.78 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.4, 132.7, 131.6, 128.8, 126.0, 55.2, 22.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{14}N_2OBr$ 269.0284; Found 269.0285.

2-chloro-N-(pyrrolidin-1-yl)benzamide (5h)



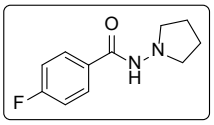
The title compound was prepared according to the general procedure and isolated as a white solid (100 mg, 89% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.67 - 7.41 (m, 1 H), 7.40 - 7.28 (m, 3 H), 6.96 (br. s, 1 H), 3.01 (br. s, 4 H), 1.90 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 171.2, 165.2, 136.2, 134.4, 131.2, 130.0, 130.0, 127.0, 55.3, 22.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{14}N_2OCl$ 225.0787; Found 225.0789.

3-chloro-N-(pyrrolidin-1-yl)benzamide (5i)



The title compound was prepared according to the general procedure and isolated as a white solid (99 mg, 88% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.67 (br. s, 1 H), 7.55 (d, *J* = 7.3 Hz, 1 H), 7.37 (d, *J* = 7.8 Hz, 1 H), 7.25 (t, *J* = 7.8 Hz, 1 H), 2.93 (br. s, 4 H), 1.81 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.1, 134.6, 133.6, 130.4, 128.8, 126.5, 124.3, 54.3, 21.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{14}N_2OCl$ 225.0789; Found 225.0790.

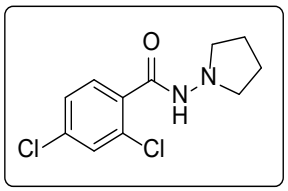
4-fluoro-N-(pyrrolidin-1-yl)benzamide (5j)



The title compound was prepared according to the general procedure and isolated as a white solid (85 mg, 82% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.78 (br. s, 2 H), 7.21 - 6.95 (m, 2 H), 2.99 (br. s, 4 H), 1.87 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz,

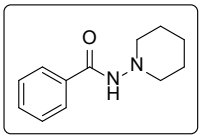
$CDCl_3$) δ = 165.9, 165.3, 163.4, 129.4, 115.4, 55.3, 22.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{14}N_2OF$ 209.1086; Found 209.1085.

2,4-dichloro-N-(pyrrolidin-1-yl)benzamide (5k)



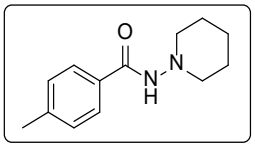
The title compound was prepared according to the general procedure and isolated as a white solid (102 mg, 79% yield). Petroleum ether/EtOAc = 90:10. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.54 (d, *J* = 8.3 Hz, 1 H), 7.40 (s, 1 H), 7.34 - 7.26 (m, 1 H), 6.95 (br. s, 1 H), 3.03 (br. s, 4 H), 1.92 (br. s, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.2, 136.8, 131.7, 131.2, 129.9, 128.3, 127.5, 55.4, 22.2. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{13}N_2OCl_2$ 259.0399; Found 259.0399.

N-(piperidin-1-yl)benzamide (5l)



The title compound was prepared according to the general procedure and isolated as a white solid (92 mg, 90% yield). Petroleum ether/EtOAc. ^{1}H NMR (400MHz, $CDCl_3$) δ = 7.66 (d, *J* = 7.1 Hz, 2 H), 7.51 - 7.29 (m, 3 H), 6.87 (br. s, 1 H), 2.78 (br. s, 4 H), 1.68 (br. s, 4 H), 1.36 (br. s, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.3, 134.1, 131.5, 128.5, 127.1, 57.1, 25.4, 23.3 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{17}N_2O$ 205.1335; Found 205.1332.

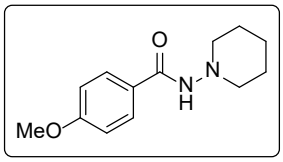
4-methyl-N-(piperidin-1-yl)benzamide (5m)



The title compound was prepared according to the general procedure and isolated as a white solid (100 mg, 92% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.64 (d, *J* = 7.8 Hz, 2 H), 7.18 (d, *J* = 7.8 Hz, 2 H), 7.08 (br. s, 1 H), 2.84 (br. s, 4 H), 2.37 (s, 3 H), 1.73 (br. s, 4 H), 1.42 (br. s, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.3, 141.8,

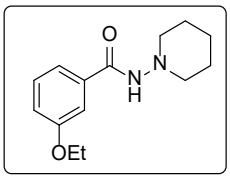
131.2, 129.1, 128.5, 128.4, 127.1, 57.0, 25.4, 23.3, 21.4 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{19}N_2O$ 219.1492; Found 219.1484.

4-methoxy-N-(piperidin-1-yl)benzamide (5n)



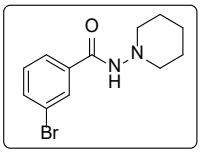
The title compound was prepared according to the general procedure and isolated as a white solid (110 mg, 94% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.71 (d, *J* = 7.4 Hz, 2 H), 6.89 (d, *J* = 8.8 Hz, 2 H and N-H proton, 1 H), 3.83 (s, 3 H), 2.85 (br. s, 4 H), 1.74 (br. s, 4 H), 1.43 (br. s, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.9, 162.1, 128.8, 126.3, 113.7, 57.2, 55.4, 25.4, 23.3 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{13}H_{19}N_2O_2$ 235.1441; Found 235.1439.

3-ethoxy-N-(piperidin-1-yl)benzamide (5o)



The title compound was prepared according to the general procedure and isolated as a white solid (109 mg, 88% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.36 - 7.07 (m, 4 H), 6.90 (br. s, 1 H), 3.95 (d, *J* = 7.0 Hz, 2 H), 2.76 (br. s, 4 H), 1.74 - 1.45 (m, 4 H), 1.32 (d, *J* = 6.9 Hz, 3 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 165.2, 159.0, 135.4, 129.5, 118.9, 117.9, 113.1, 63.6, 56.9, 25.3, 23.2, 14.7 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{14}H_{21}N_2O_2$ 249.1598; Found 249.1595.

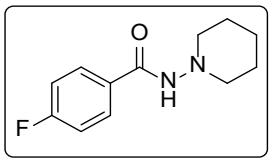
3-bromo-N-(piperidin-1-yl)benzamide (5p)



The title compound was prepared according to the general procedure and isolated as a white solid (118 mg, 83% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ =

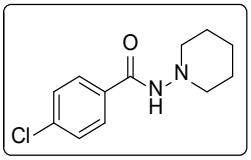
7.82 (br. s, 1 H), 7.60 (d, *J* = 7.5 Hz, 1 H), 7.51 (d, *J* = 7.6 Hz, 1 H), 7.18 (t, *J* = 7.8 Hz, 1 H), 2.78 (br. s, 4 H), 1.71 - 1.58 (m, 4 H), 1.34 (br. s, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.0, 136.0, 134.3, 130.3, 130.1, 125.8, 122.6, 56.9, 25.3, 23.2 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{16}N_2OBr$ 283.0441; Found 283.0442.

4-fluoro-N-(piperidin-1-yl)benzamide (5q)



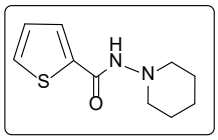
The title compound was prepared according to the general procedure and isolated as a white solid (90 mg, 81% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 8.06 (dt, *J* = 1.6, 7.7 Hz, 1 H), 7.58 - 7.32 (m, 2 H), 7.32 - 7.17 (m, 1 H), 7.10 (dd, *J* = 8.8, 11.3 Hz, 1 H), 2.89 (br. s, 4 H), 1.88 - 1.69 (m, 4 H), 1.46 (br. s, 2 H) ppm;^{13}C NMR (100 MHz, $CDCl_3$) δ = 161.1, 133.2, 132.2, 124.9, 116.0, 57.0, 25.4, 25.3, 23.3 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{16}N_2OF$ 223.1240; Found 223.1241.

4-chloro-N-(piperidin-1-yl)benzamide (5r)



The title compound was prepared according to the general procedure and isolated as a white solid (104 mg, 87% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz $CDCl_3$) δ = 7.62 (d, *J* = 8.1 Hz, 2 H), 7.27 (d, *J* = 8.1 Hz, 2 H), 2.77 (br. s, 4 H), 1.63 (br. s, 4 H), 1.34 (br. s, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.4, 137.6, 132.4, 128.7, 128.6, 56.9, 25.3, 23.2. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{12}H_{16}N_2OCl$ 239.0943; Found 239.0946.

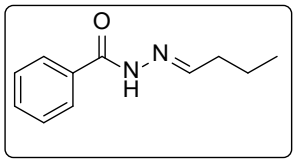
N-(piperidin-1-yl)thiophene-2-carboxamide (5s)



The title compound was prepared according to the general procedure and isolated as a white solid (81 mg, 77% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ =

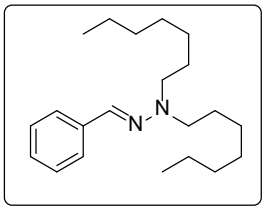
8.20 - 7.90 (m, 1 H), 7.54 (d, *J* = 3.9 Hz, 1 H), 7.28 (br. s, 1 H), 7.17 - 6.91 (m, 1 H), 3.15 (d, *J* = 9.4 Hz, 2 H), 2.46 (t, *J* = 10.6 Hz, 2 H), 1.83 (d, *J* = 12.0 Hz, 2 H), 1.78 - 1.61 (m, 4 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 163.5, 135.0, 133.8, 132.1, 126.2, 57.7, 25.4, 23.0 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{10}H_{15}N_2OS$ 211.0900; Found 211.0900.

(E)-N'-butylidenebenzohydrazide (3a')



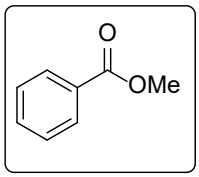
The title compound was prepared according to the general procedure and isolated as a white solid (84 mg, 88% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 10.85 (br. s, 1 H), 7.76 (d, *J* = 7.4 Hz, 2 H), 7.73 - 7.55 (m, 1 H), 7.43 - 7.28 (m, 1 H), 7.25 - 7.06 (m, 2 H), 2.28 - 2.02 (m, 2 H), 1.48 - 1.22 (m, 2 H), 0.79 (t, *J* = 7.3 Hz, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$) δ = 164.7, 153.5, 133.1, 131.7, 128.3, 127.6, 34.4, 19.9, 13.7. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{11}H_{15}N_2O$ 191.1179; Found 191.1178.

(E)-2-benzylidene-1,1-diheptylhydrazine (6a)



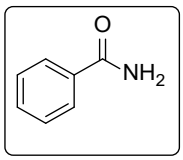
The title compound was prepared according to the general procedure and isolated as a pale-yellow liquid (146 mg, 93% yield). Petroleum ether/EtOAc = 98:2. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 0.75 - 0.86 (m, 6 H), 1.18 - 1.33 (m, 16 H), 1.50 - 1.56 (m, 4 H), 3.07 - 3.31 (m, 4 H), 7.04 - 7.11 (m, 1 H), 7.12 (s, 1 H), 7.17 - 7.26 (m, 2 H), 7.45 (d, *J*=7.38 Hz, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 13.07, 21.61, 25.87 26.18, 28.15, 30.83, 52.48, 124.05, 125.46, 127.35, 127.54, 136.67 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{21}H_{37}N_2$ 316.1239; Found 316.1241.

Methyl benzoate (6b)



The title compound was prepared according to the general procedure and isolated as a colourless liquid (22 mg, 32% yield). Petroleum ether/EtOAc = 99:1. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 3.92 (s, 3 H), 7.34 - 7.51 (m, 2 H), 7.51 - 7.60 (m, 1 H), 7.92 - 8.20 (m, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 52.10, 76.73, 128.36, 129.58, 130.17, 132.91, 167.13 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for C_8H_9O 137.5459; Found 137.5456.

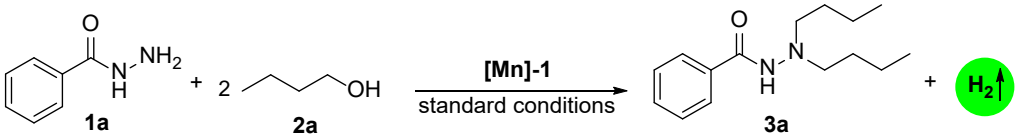
Benzamide (6c)



The title compound was prepared according to the general procedure and isolated as a white solid (43 mg, 71% yield). Petroleum ether/EtOAc = 70:30. ^{1}H NMR (400 MHz, $CDCl_3$) δ = 7.35 - 7.49 (m, 2 H), 7.54 (d, *J*=7.38 Hz, 1 H), 7.73 - 7.88 (m, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ = 76.71, 127.41, 128.68, 132.20, 133.03, 169.63 ppm. HRMS (ESI) m/z: $[M+H]^+$ Calcd for C_7H_8ON 122.0597; Found 122.0600.

4. Mechanistic investigations

4.1. Detection of H_2 gas



In an oven-dried screw cap reaction tube (15 mL), benzohydrazide **1a** (1 mmol), 1-butanol **2a** (2.2 mmol), **[Mn-1]** (3 mol%), KO^{t}Bu (50 mol%), and dry toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 ºC (oil-bath temperature) for 4 h. After completion of the reaction the crude mixture was cooled to room temperature followed by the sample was submitted to GC as well as gasometer for detection of H_2 gas (Figure S1).

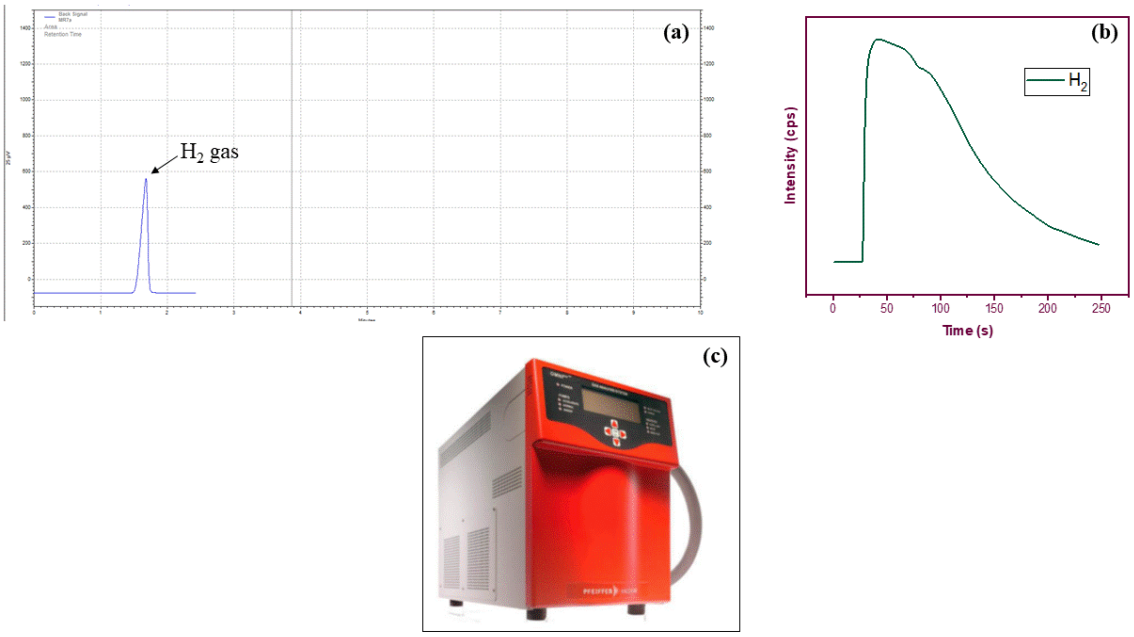


Figure S1. Detection of H_2 gas (a) by GC analysis, (b) Gasometer analysis. (c) Photograph of OmniStarTM Gas Analysis System GSD 320 (Pfeiffer) quadrupole mass spectrometer apparatus used for the analysis of hydrogen gas (Gasometer).

4.2. Deuterium labeling experiments

4.2.1. Synthesis of deuterated heptanol-D_3 (2d-D)

To an oven-dried 10 mL screw-capped vial, Ru-MACHO (3 mol%), 1-heptanol **2d** (0.5 mmol), KOH (0.55 mmol, 1.1 equivalent), and deuterium oxide (1 mL) were added under a gentle stream of argon. The reaction mixture was kept for stirring at 130 ºC (oil-bath temperature) for 18 h. Then, the reaction mixture was diluted with water (4 mL) and extracted with dichloromethane (3 x 5 mL). The resultant organic layer was dried over anhydrous Na_2SO_4 and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica

gel column chromatography (230-400 mesh size) using Petroleum ether/EtOAc = 90:10 as an eluting system to give the deuterated product **2d-D** (92.5% deuterated).

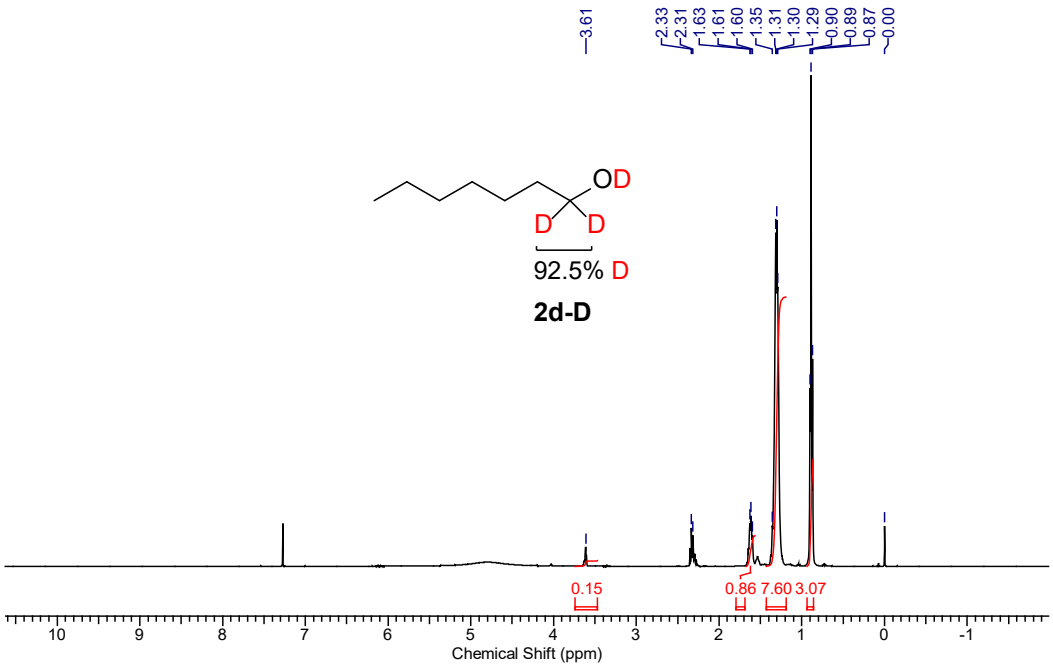


Figure S2. ^{1}H NMR of **2d-D**, 400 MHz, $CDCl_3$

4.2.2. Synthesis of 3y-D

To an oven-dried 15 mL screw-capped vial, CD_3OD (1 mL), benzohydrazide (0.5 mmol), KO^{t}Bu (50 mol%), [Mn-1] (5 mol%), and toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130^o C (oil-bath temperature). After completion of the reaction the crude mixture was cooled to room temperature followed by filtered through a celite pad with several washings (3 x 3 mL ethyl acetate) and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (eluent: Petroleum ether/EtOAc = 70:30) to afford the product **3y-D** in 91% yield.

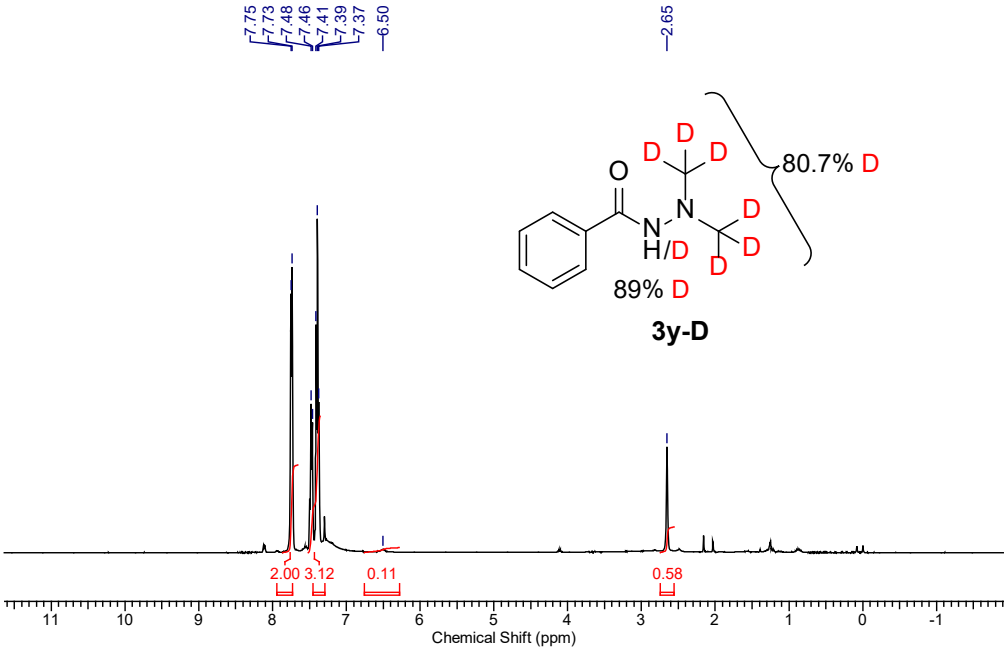


Figure S3. ^{1}H NMR of **3y-D**, 400 MHz, $CDCl_3$

4.2.3. Synthesis of 3d-D

To an oven-dried 15 mL screw-capped vial, heptanol-D_3 (1.1 mmol), benzohydrazide (0.5 mmol), KO^{t}Bu (50 mol%), [Mn-1] (5 mol%), and toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130^{o} C (oil-bath temperature). After completion of the reaction the crude mixture was cooled to room temperature followed by filtered through a celite pad with several washings (3 x 3 mL ethyl acetate) and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (eluent: Petroleum ether/EtOAc = 70:30) to afford the product **3d-D** in 90% yield.

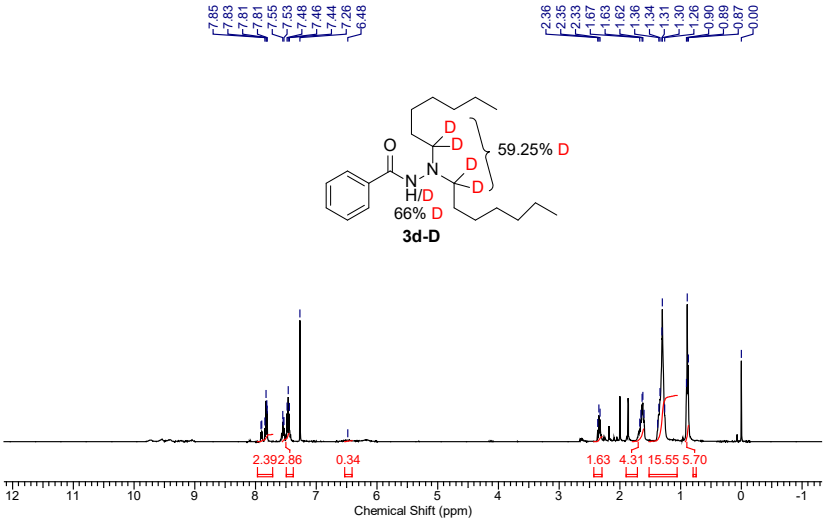


Figure S4. ^{1}H NMR of **3d-D**, 400 MHz, $CDCl_3$

4.2.4. Synthesis of 3I-D

To an oven-dried 15 mL screw-capped vial, C_2D_6O (10 equivalent), benzohydrazide (0.5 mmol), KO^{t}Bu (50 mol%), [Mn-1] (5 mol%), and toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130^o C (oil-bath temperature). After completion of the reaction the crude mixture was cooled to room temperature followed by filtered through a celite pad with several washings (3 x 3 mL ethyl acetate) and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (eluent: Petroleum ether/EtOAc = 70:30) to afford the product **3I-D** in 89% yield.

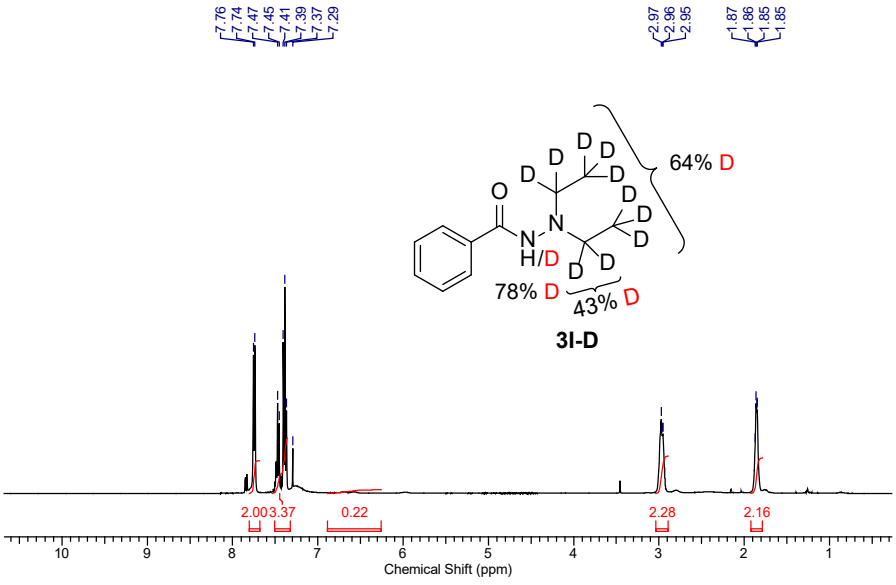


Figure S5. ^{1}H NMR of **3I-D**, 400 MHz, $CDCl_3$

4.3. Synthesis of intermediate hydrazone

To a solution of the benzohydrazide (1 mmol) and butyraldehyde (1.2 mmol) containing EtOH was reflux in a 50 mL round bottom flask at room temperature for 10 h. After completion of the reaction the crude mixture was cooled to room temperature followed by filtered through a celite pad with several washings (3 x 3 mL ethyl acetate) and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (eluent: Petroleum ether/EtOAc = 90:10) to afford the product.

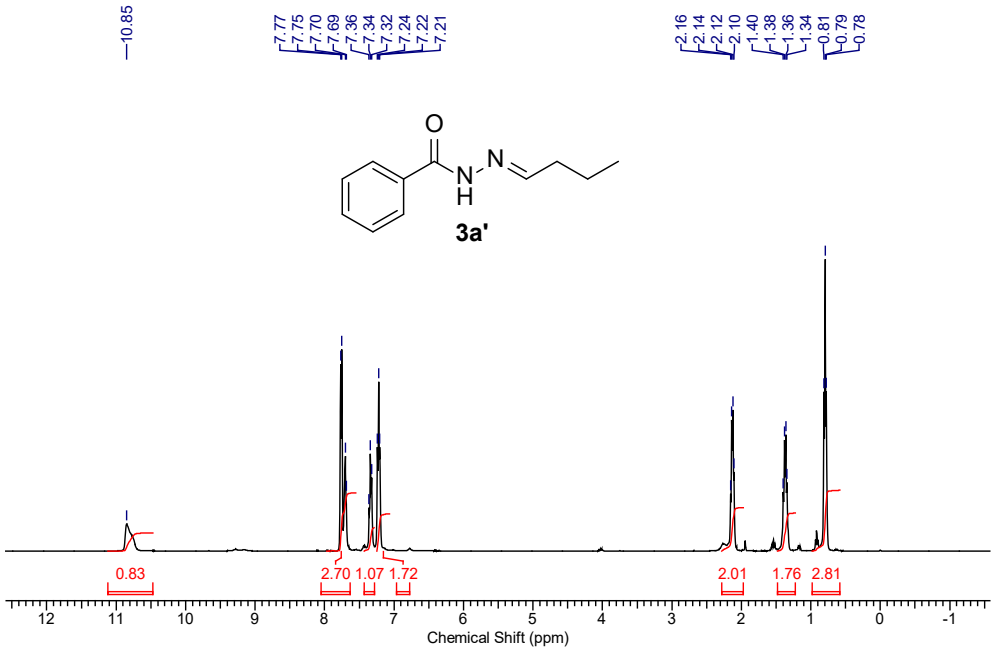


Figure S6. ^{1}H NMR of **3a'**, 400 MHz, $CDCl_3$

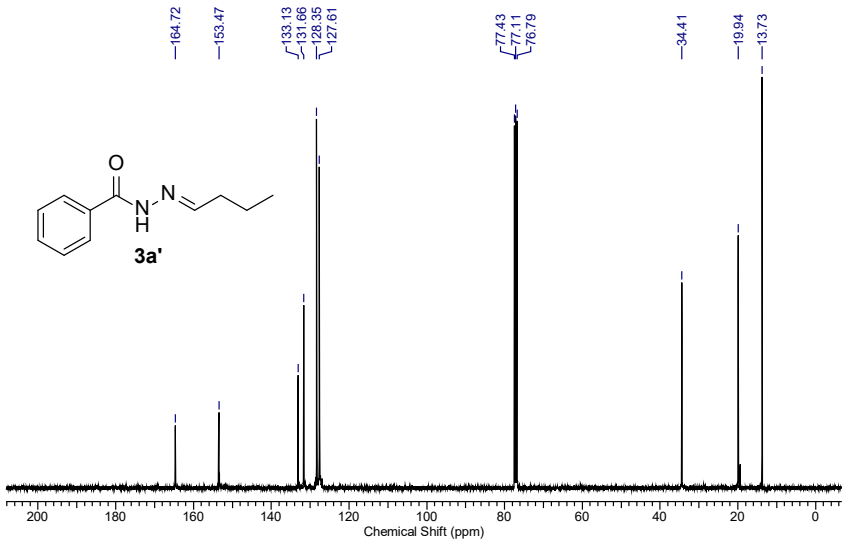
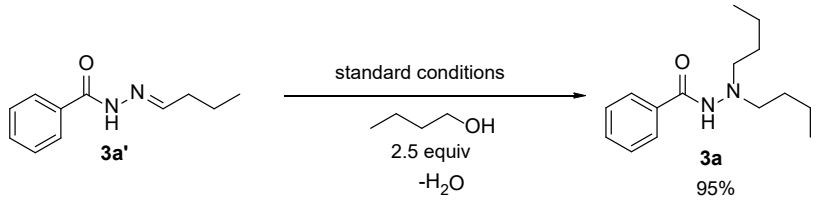


Figure S7. ^{13}C NMR of **3a'**, 100 MHz, $CDCl_3$

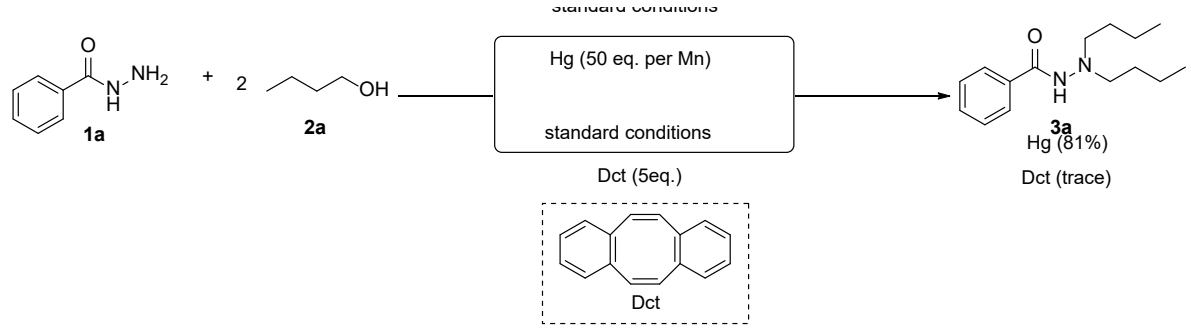
4.4. Alkylation of intermediate hydrazone with 1-butanol



In an oven dried screw cap reaction tube (15 mL), 1-butanol (0.75 mmol), hydrazone (0.5 mmol), KO^{t}Bu (50 mol%), [Mn-1] (5 mol%), and toluene (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 ºC (oil-bath temperature) for 4 h. After completion of the reaction the crude mixture was filtered through celite filter and washed with ethyl acetate (3×5 mL), the residual solvent was removed under vacuum and finally the residue was purified by silica gel column chromatography (230-400 mesh size) using petroleum-ether and ethyl acetate as an eluent. The reaction of hydrazone in the presence of 1-butanol identified *N',N'*-dibutylbenzohydrazide as the major product.

4.5. Reaction in presence of Hg and DCT

In an oven dried screw cap reaction tube (15 mL), 1-butanol (1.1 mmol), benzohydrazide (0.5 mmol), KO^{t}Bu (50 mol%), [Mn-1] (5 mol%), Hg (50 equivalent with respect to catalyst) or DCT (5 equivalent) and toluene (1 mL) were added in a gentle stream of argon. Then the reaction mixture was stirred with a magnetic stirring bar at 130 ºC (oil-bath temperature) for 4 h. After completion of the reaction the crude mixture was filtered through celite filter and washed with ethyl acetate (3×5 mL), the residual solvent was removed under vacuum and finally the residue was purified by silica gel column chromatography (230-400 mesh size) using petroleum-ether and ethyl acetate as an eluent.



5. Monitoring the kinetic profile of the reaction

In an oven dried screw cap pressure tube (15 mL), **1a** (0.5 mmol, 1 eq.), **2d** (1.1 mmol, 2.2 eq.), **[Mn-1]** (3 mol%), KO^{t}Bu (50 mol%), n-octane (0.5 mmol, 1 eq.) as an internal standard and dry toluene (2 mL) were added in a gentle stream of argon. Then the reaction mixture was stirred with a magnetic stirring bar at 130 ^{o}C (oil-bath temperature). At regular intervals (5 min, 10 min, 15 min, 20 min, 25 min, 30 min, 35 min, 40 min, 45 min, 50 min) the reaction mixture was cooled to ambient temperature and an aliquot of mixture was taken in a GC vial. The GC sample was diluted with methanol and subjected to gas chromatographic analysis. The concentration of the products was determined with respect to n-octane as internal standard. The data represented was taken from the average of two independent set of experiments. Next, the reaction kinetic profile for the manganese-catalyzed *N,N*-dialkylation of benzohydrazide **1a** with 1-heptanol **2d** was conducted (Figure S8). The time dependent product formation data clearly convey that the product **3d** formation increases rapidly at ten minutes.

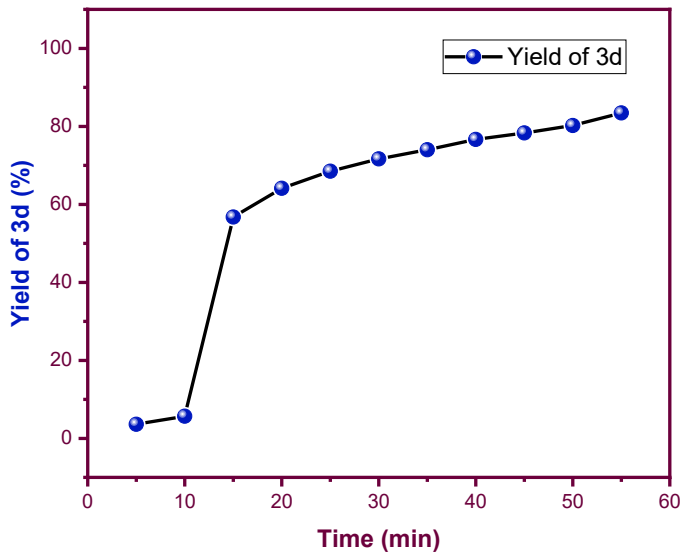
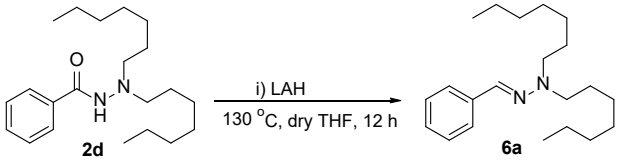


Figure S8. Kinetic plot for the manganese-catalyzed *N,N*-dialkylation of benzohydrazide **(1a)** with 1-heptanol **(2d).**

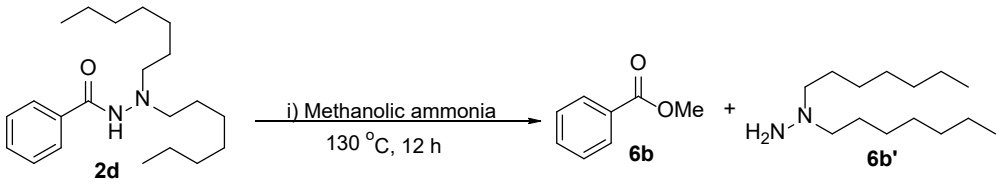
6. Diversification of the N,N-dialkylated product

6a. Lithium aluminium hydride reduction



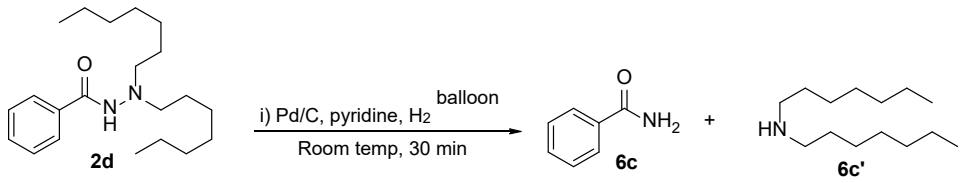
In an oven dried screw cap reaction tube (15 mL) with a magnetic stirring bar was charged with **2d** (0.5 mmol), LAH (0.75 mmol), dry THF (2 mL) followed by pyridine (10 mol %) under nitrogen atmosphere and the reaction tube was stirred under nitrogen atmosphere at 0 °C for 10 minutes. Then, the reaction mixture was transferred with a magnetic stirring bar at 130° C (oil-bath temperature) for 12 h. After completion of the reaction the crude mixture was filtered through celite filter and washed with ethyl acetate (3×5 mL), the residual solvent was removed under vacuum and finally the residue was purified by silica gel column chromatography (230-400 mesh size) using petroleum-ether and ethyl acetate as an eluent.

6b. Reaction with methanolic ammonia



In an oven dried screw cap reaction tube (15 mL), **2d** (0.5 mmol) and methanolic ammonia (1 mL) were added in a gentle stream of argon. Then, the reaction mixture was stirred with a magnetic stirring bar at 130 °C (oil-bath temperature) for 12 h. After completion of the reaction the crude mixture was filtered through celite filter and washed with ethyl acetate (3×5 mL), the residual solvent was removed under vacuum and finally the residue was purified by silica gel column chromatography (230-400 mesh size) using petroleum-ether and ethyl acetate as an eluent.

6c. Hydrogenation with Pd/C



In an oven-dried 10 mL round-bottomed flask with a magnetic stirring bar was charged with **2d** (0.5 mmol), 2.1 mg 5 wt% Pd/C (0.2% of Pd loading), dry methanol (2 mL) followed by pyridine (10 mol %) under nitrogen atmosphere. The round-bottomed flask was fitted to an adapter which is connected to a hydrogen balloon (1 atm.). The N_2 gas from the round-bottomed flask was flushed out by releasing hydrogen gas and then the reaction flask was placed on a magnetic stirrer under hydrogen atmosphere and allowed to stir at room temperature. After 15-30 minutes the color of the reaction mixture disappears which indicates the completion of the reaction (completion of the reaction was confirmed by checking TLC). Then the catalyst was removed by a quick filtration. Then the clear solution was dissolved in dry methanol for HRMS analysis. The solvent of the reaction mixture was removed under reduced pressure, and the residue was washed with cold n-hexane (5 mL×3) to afford desired pure product **6c** as colourless solid.

7. Copy of ^{1}H and ^{13}CNMR

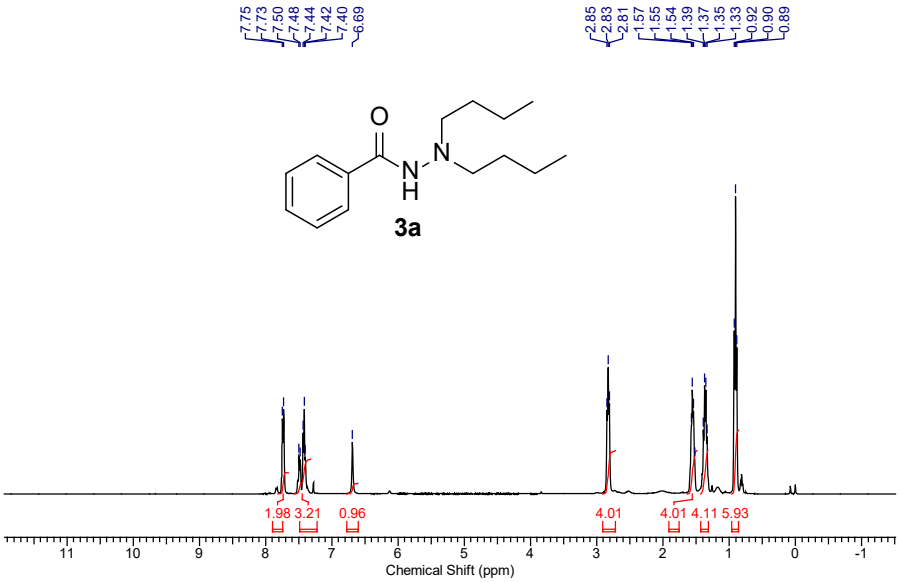


Figure S9. ^{1}H NMR of **3a** (400 MHz, $CDCl_3$)

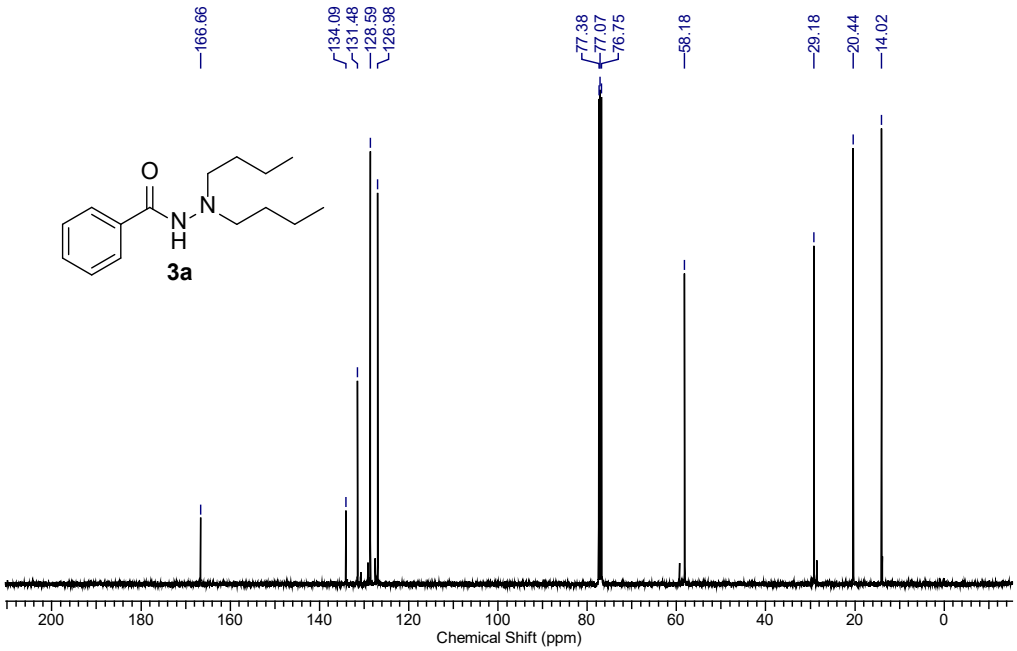


Figure S10. ^{13}C NMR of **3a** (00 MHz, $CDCl_3$)

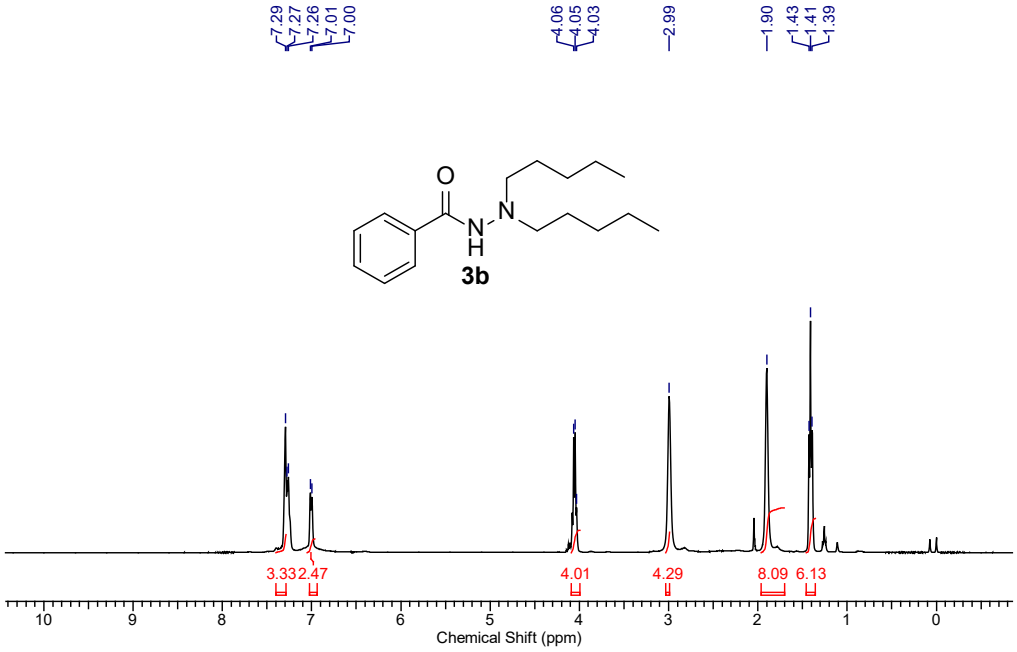


Figure S11. ^{1}H NMR of **3b** (400 MHz, $CDCl_3$)

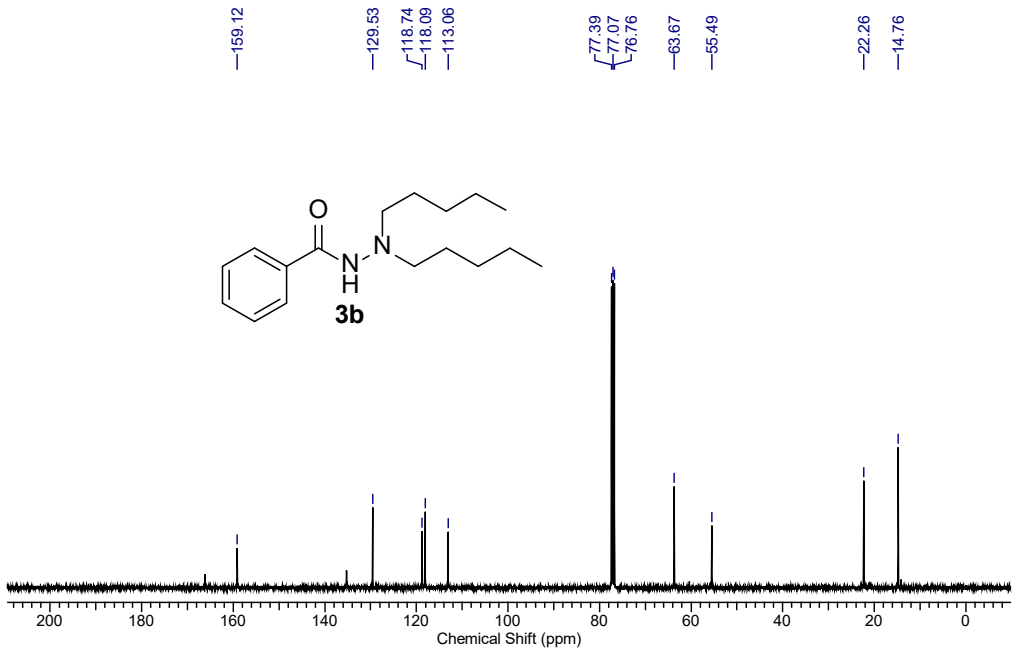


Figure S12. ^{13}C NMR of **3b** (100 MHz, $CDCl_3$)

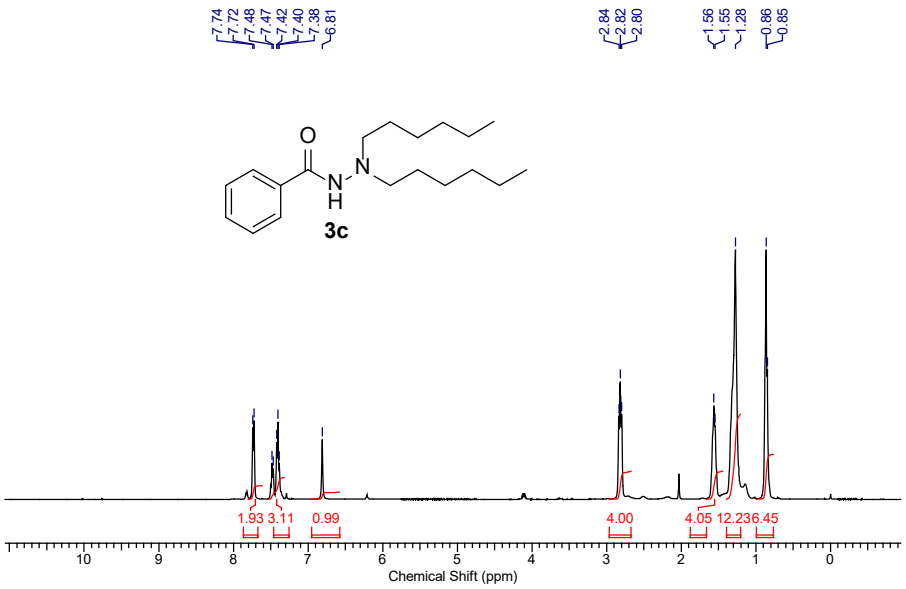


Figure S13. ^{1}H NMR of **3c** (400 MHz, $CDCl_3$)

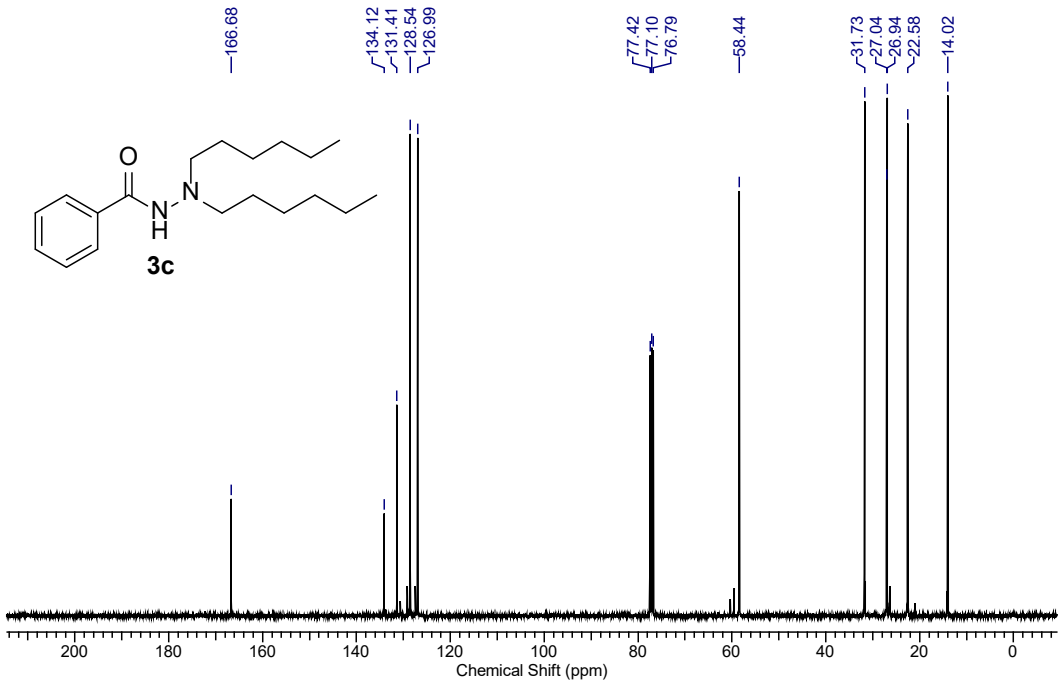


Figure S14. ^{13}C NMR of **3c** (100 MHz, $CDCl_3$)

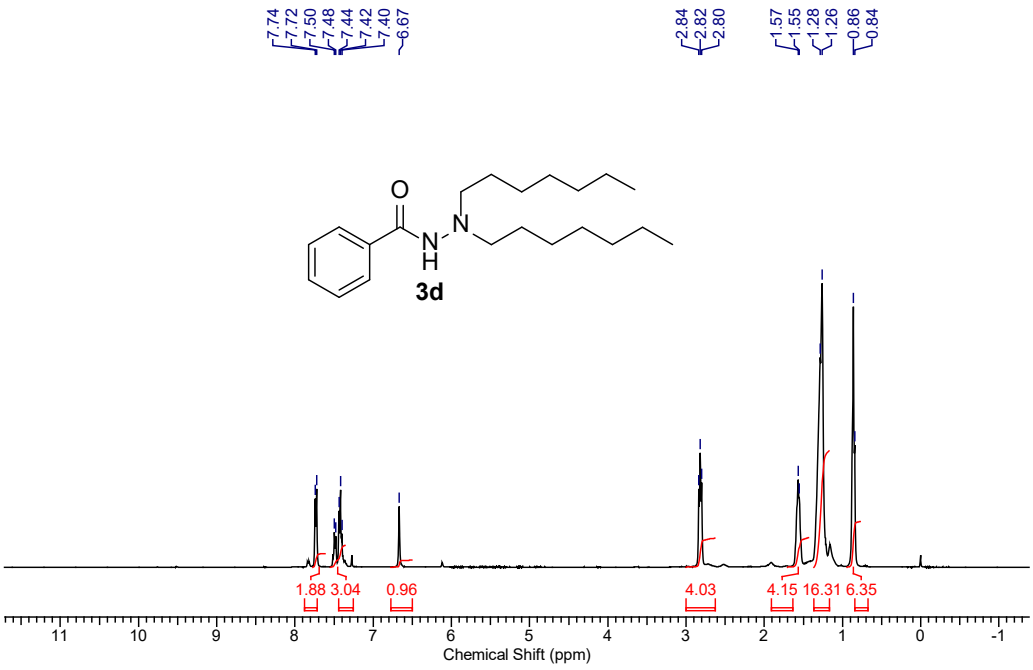


Figure S15. ^{1}H NMR of **3d** (400 MHz, $CDCl_3$)

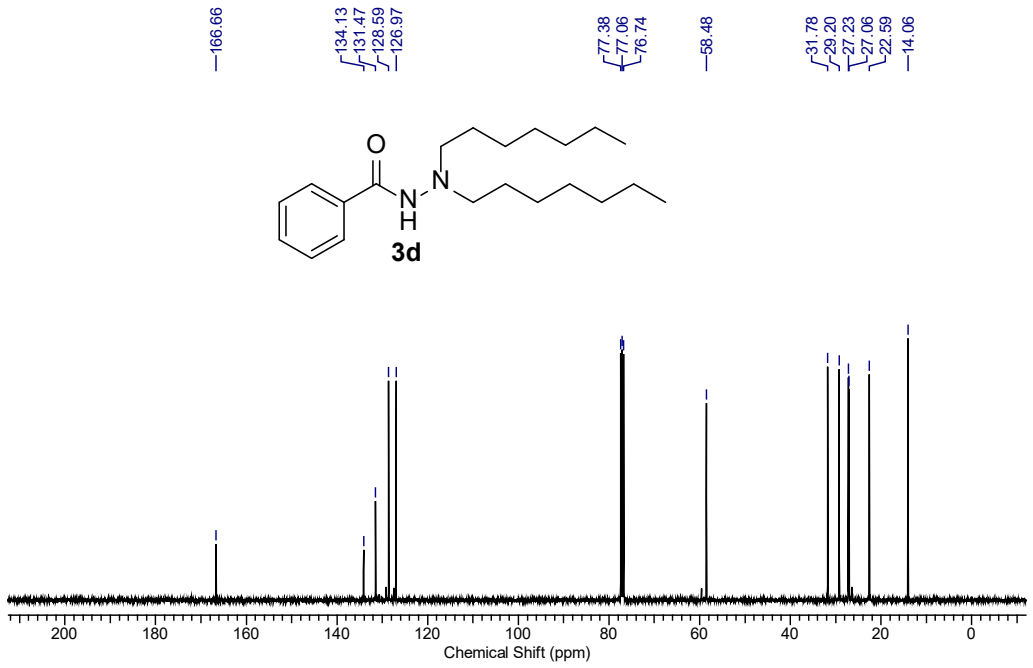


Figure S16. ^{13}C NMR of **3d** (100 MHz, $CDCl_3$)

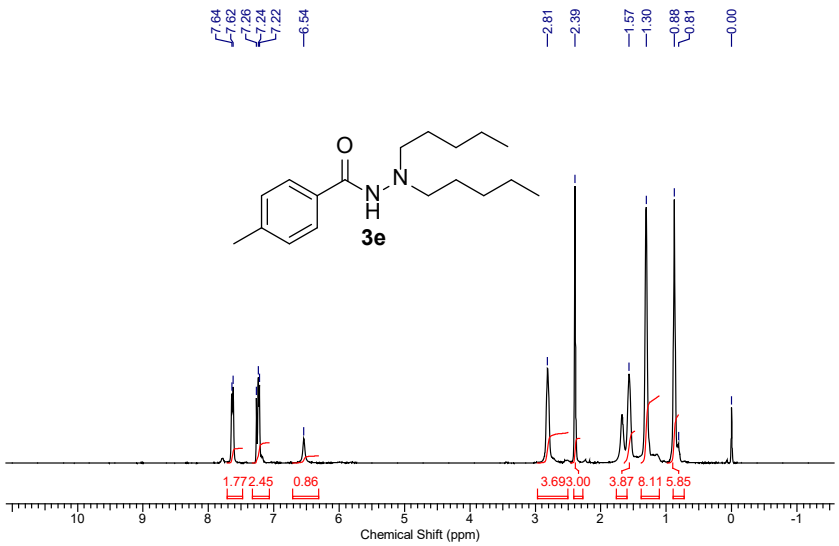


Figure S17. ^{1}H NMR of **3e** (400 MHz, $CDCl_3$)

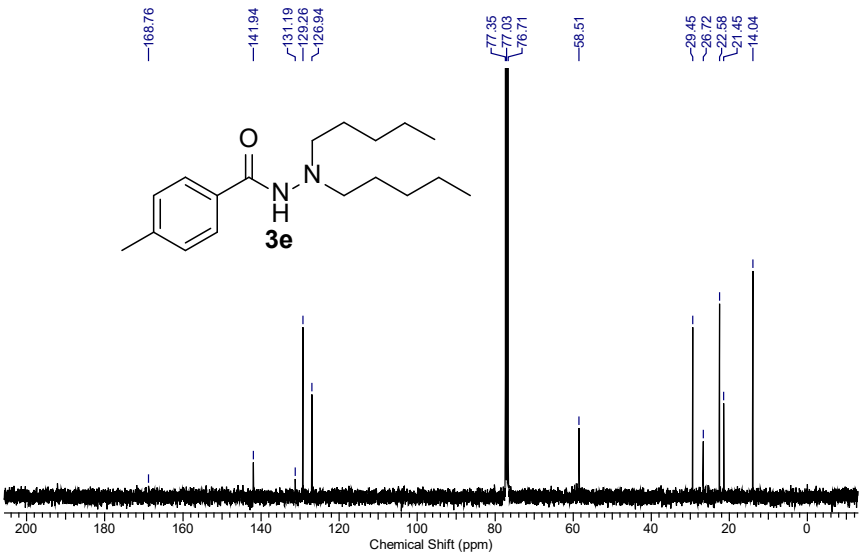


Figure S18. ^{13}C NMR of **3e** (100 MHz, $CDCl_3$)

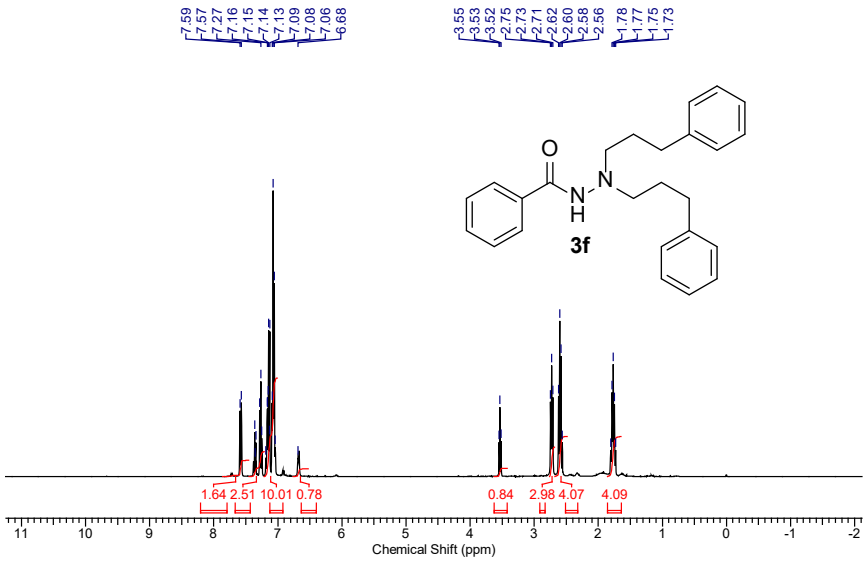


Figure S19. ^{1}H NMR of **3f** (400 MHz, $CDCl_3$)

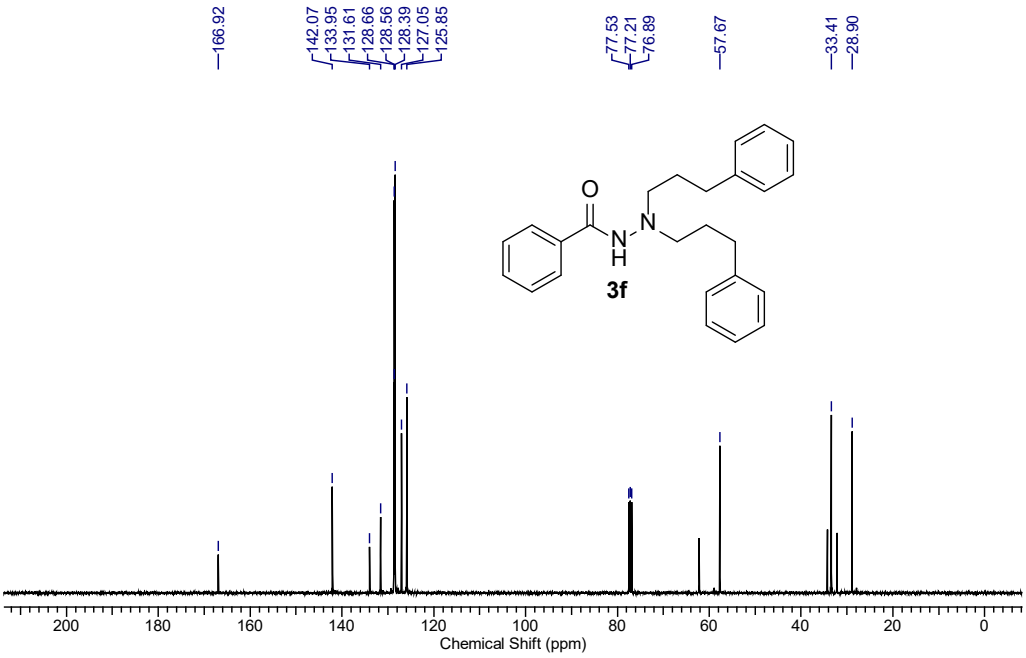


Figure S20. ^{13}C NMR of **3f** (100 MHz, $CDCl_3$)

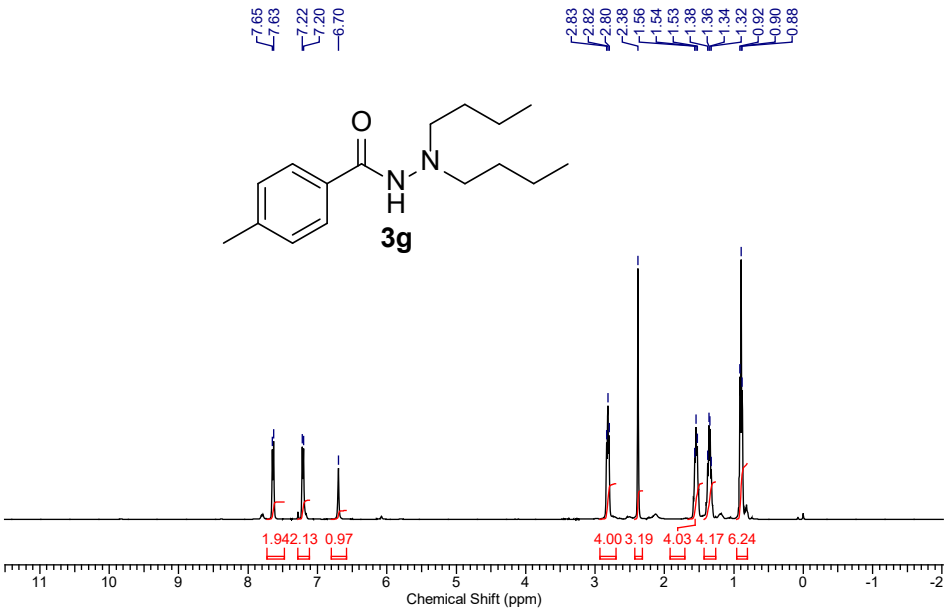


Figure S21. ^{1}H NMR of **3g** (400 MHz, $CDCl_3$)

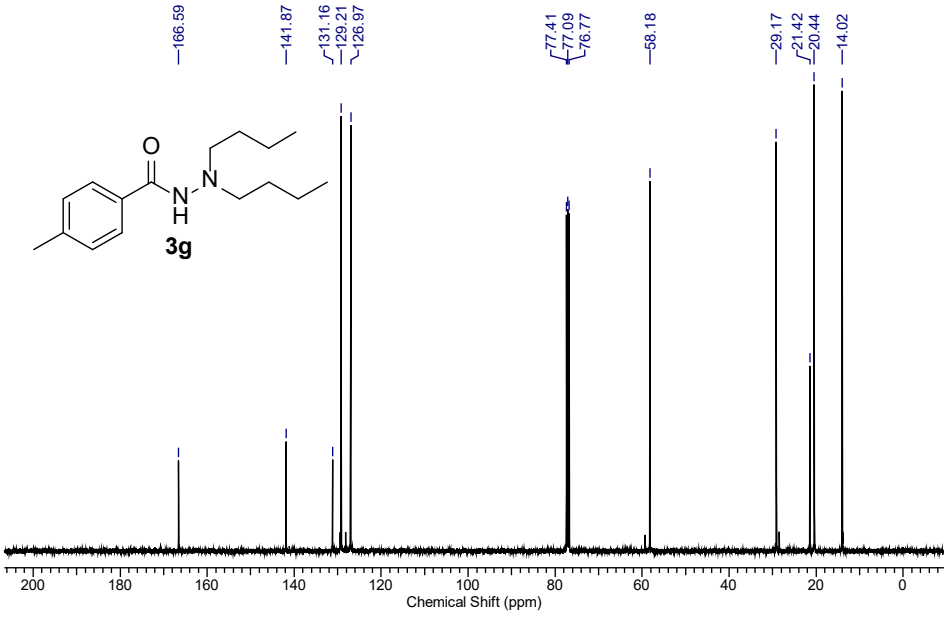


Figure S22. ^{13}C NMR of **3g** (100 MHz, $CDCl_3$)

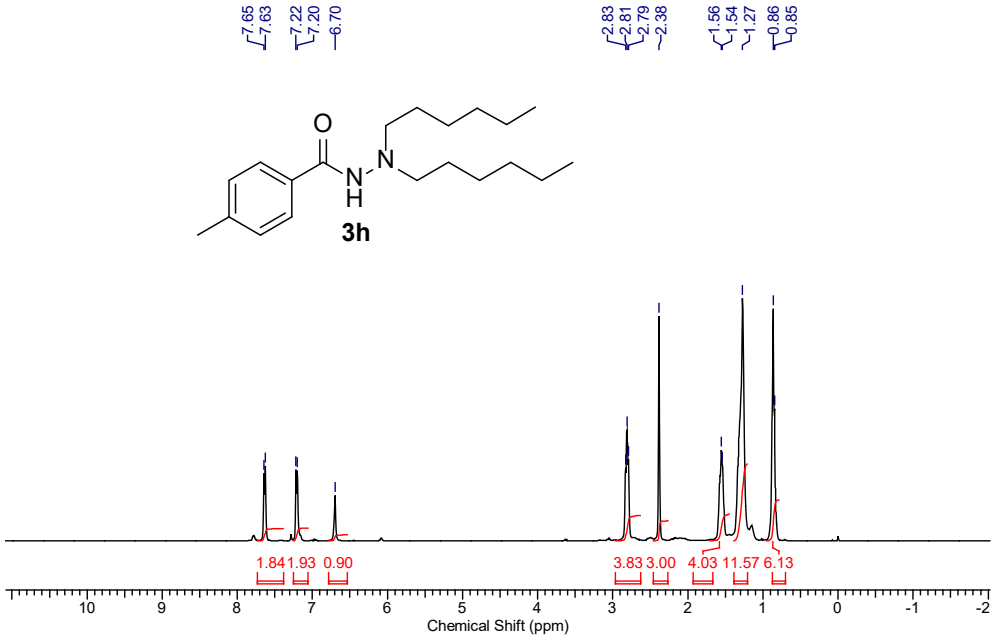


Figure S23. ^{1}H NMR of **3h** (400 MHz, $CDCl_3$)

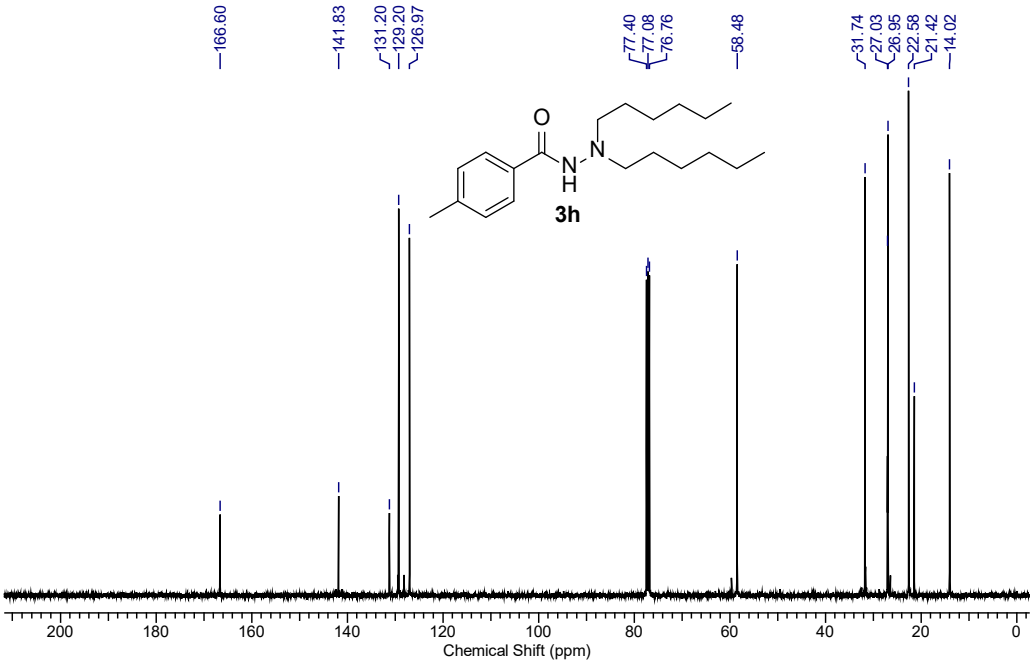


Figure S24. ^{13}C NMR of **3h** (100 MHz, $CDCl_3$)

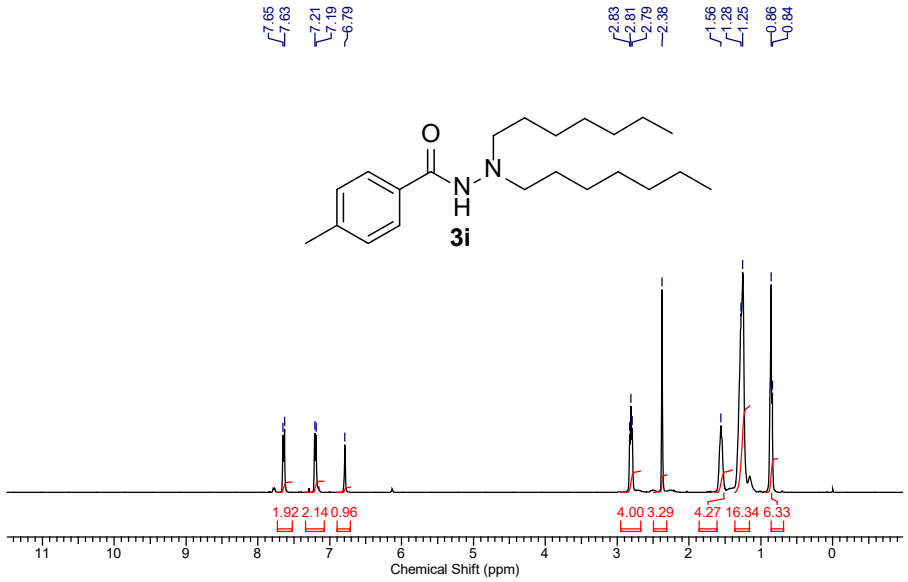


Figure S25. ^{1}H NMR of **3i** (400 MHz, $CDCl_3$)

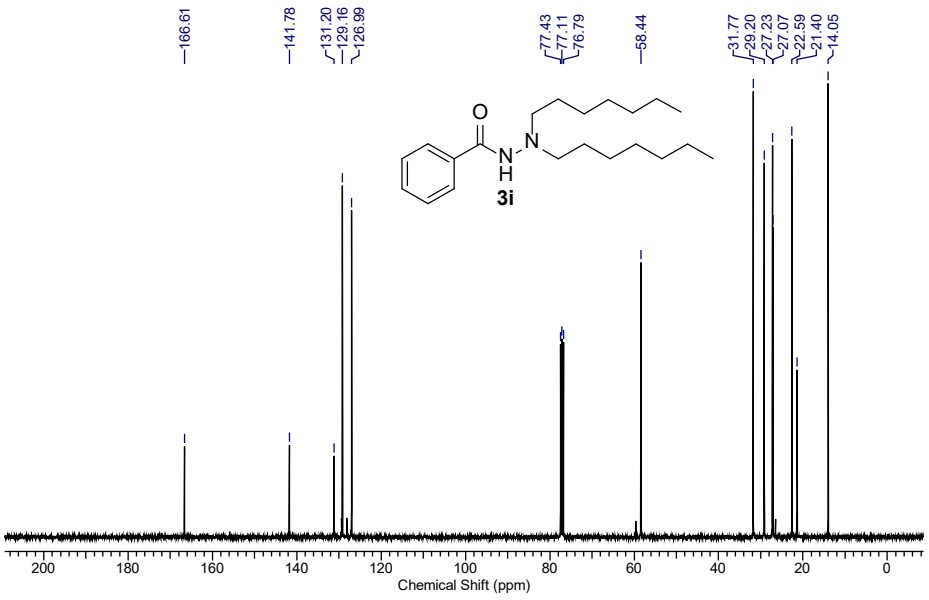


Figure S26. ^{13}C NMR of **3i** (100 MHz, $CDCl_3$)

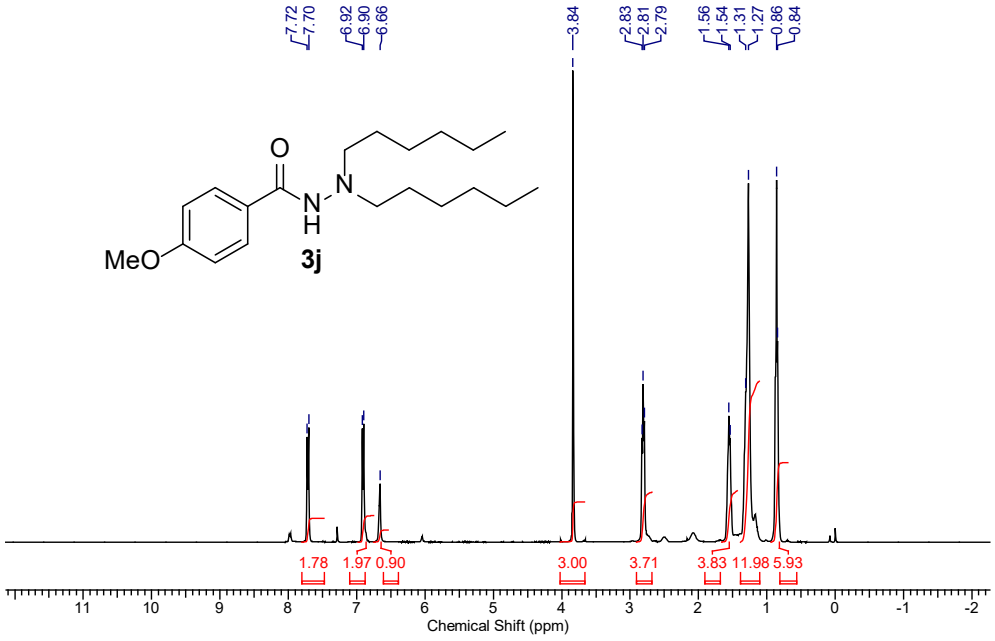


Figure S27. ^{1}H NMR of **3j** (400 MHz, $CDCl_3$)

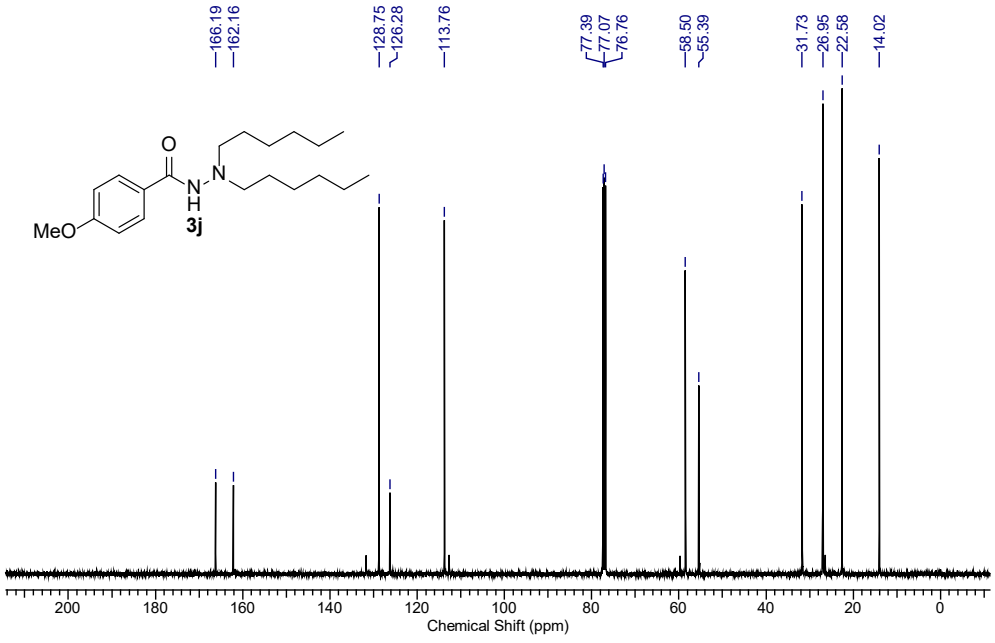


Figure S28. ^{13}C NMR of **3j** (100 MHz, $CDCl_3$)

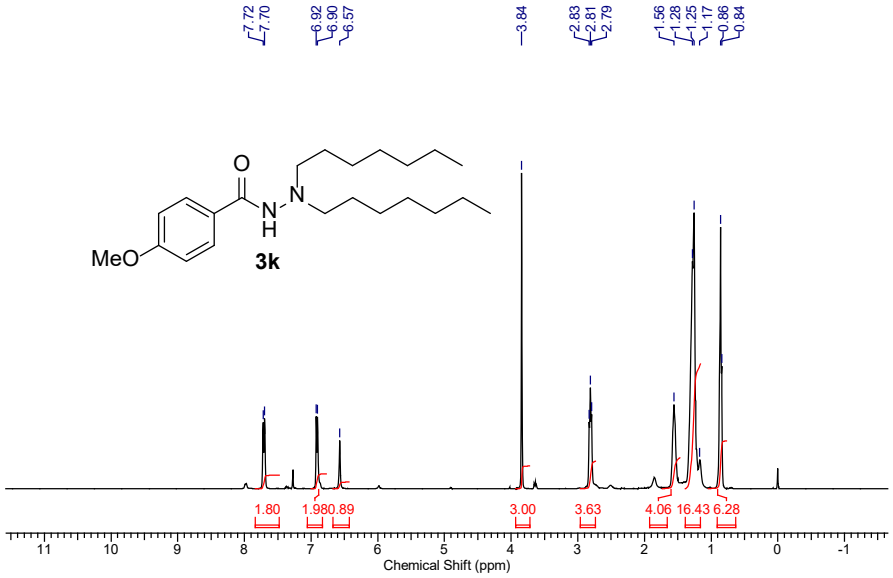


Figure S29. ^{1}H NMR of **3k** (400 MHz, $CDCl_3$)

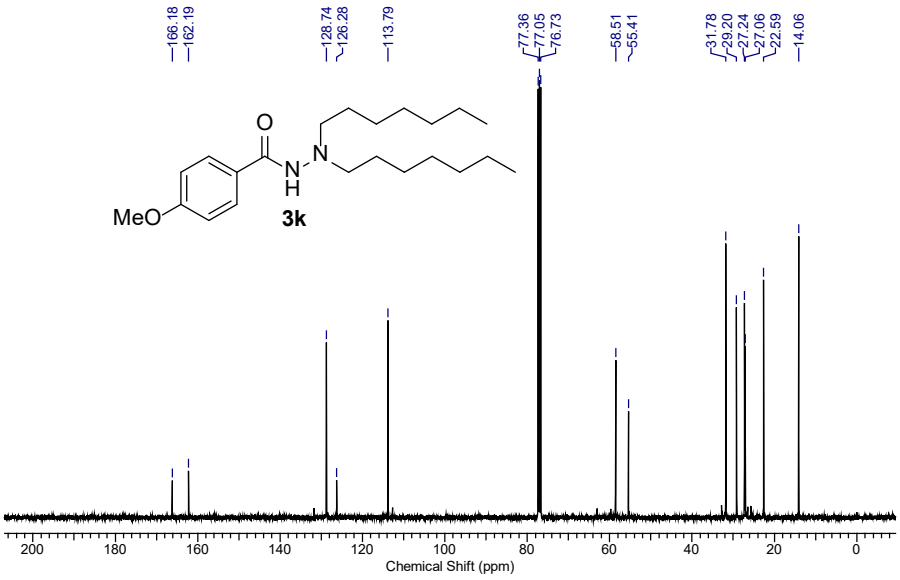


Figure S30. ^{13}C NMR of **3k** (100 MHz, $CDCl_3$)

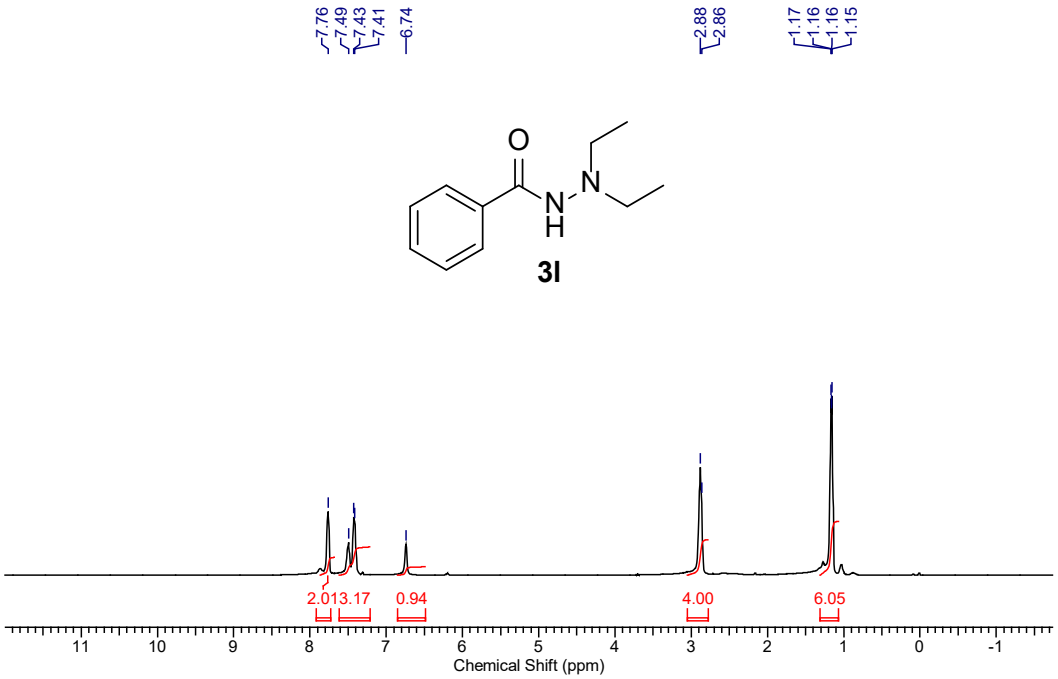


Figure S31. ^{1}H NMR of **3l** (400 MHz, $CDCl_3$)

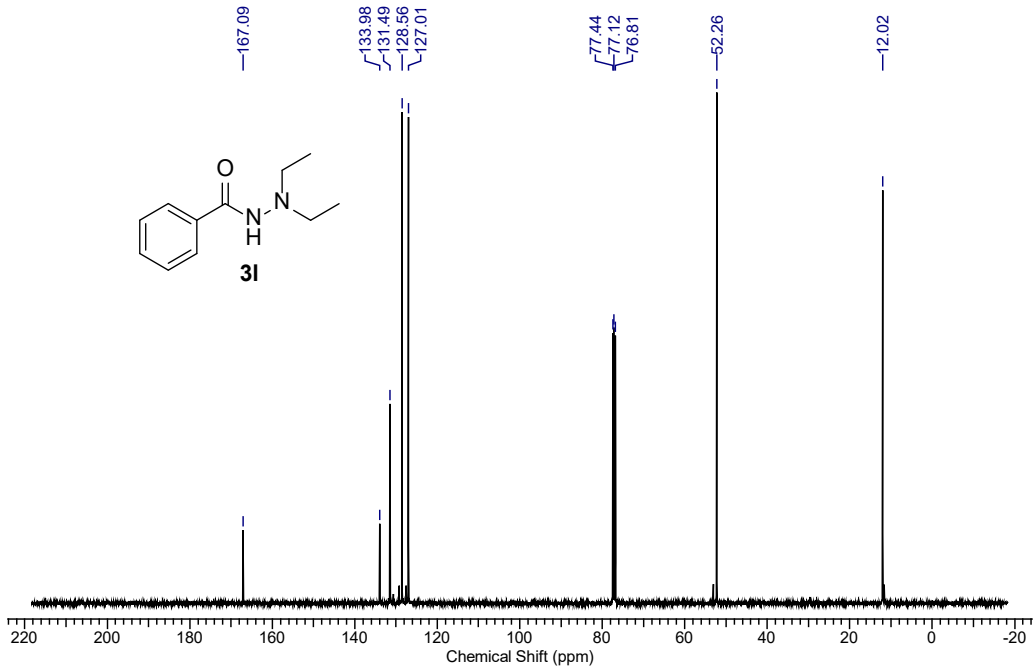


Figure S32. ^{13}C NMR of **3l** (100 MHz, $CDCl_3$)

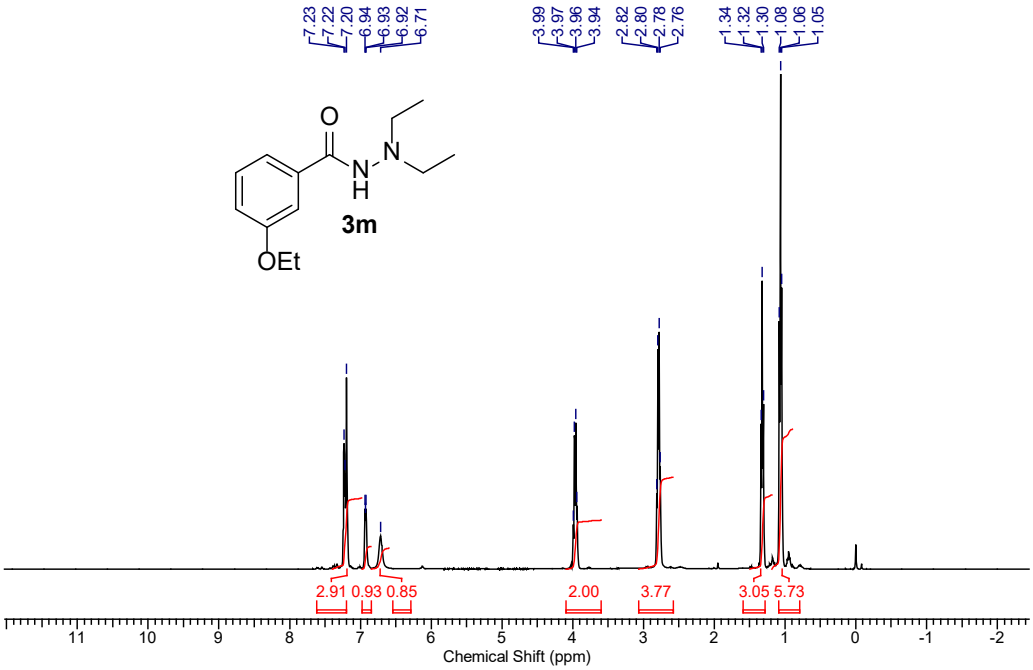


Figure S33. ^{1}H NMR of **3m** (400 MHz, $CDCl_3$)

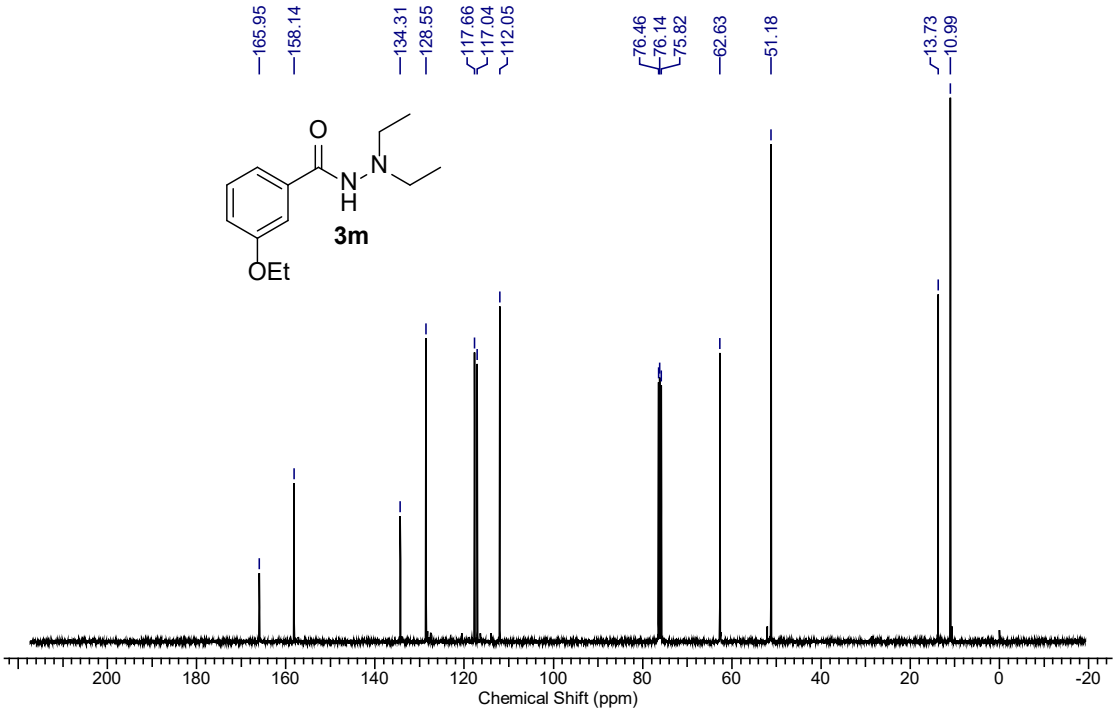


Figure S34. ^{13}C NMR of **3m** (100 MHz, $CDCl_3$)

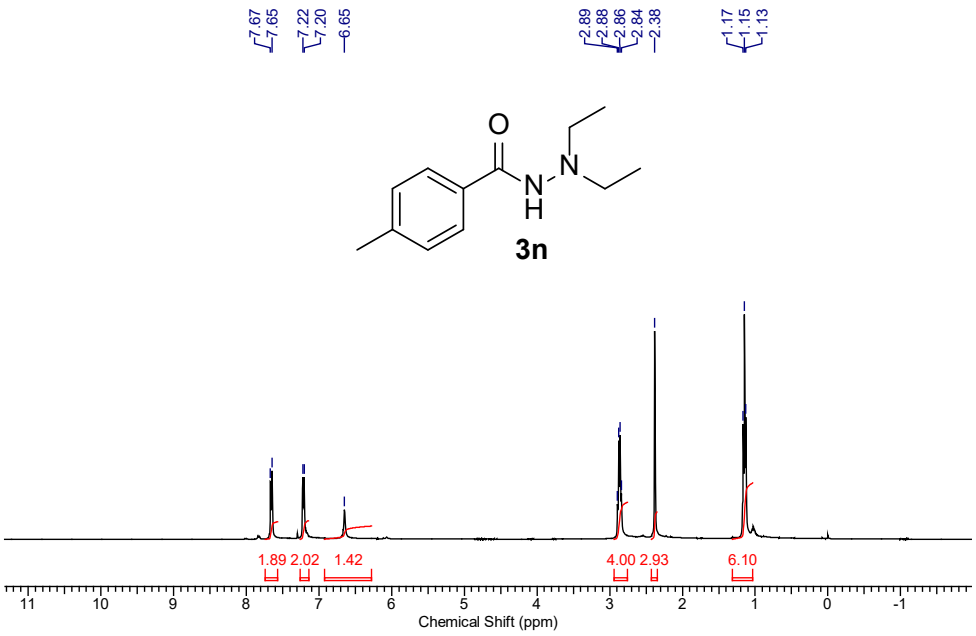


Figure S35. ^{1}H NMR of **3n** (400 MHz, $CDCl_3$)

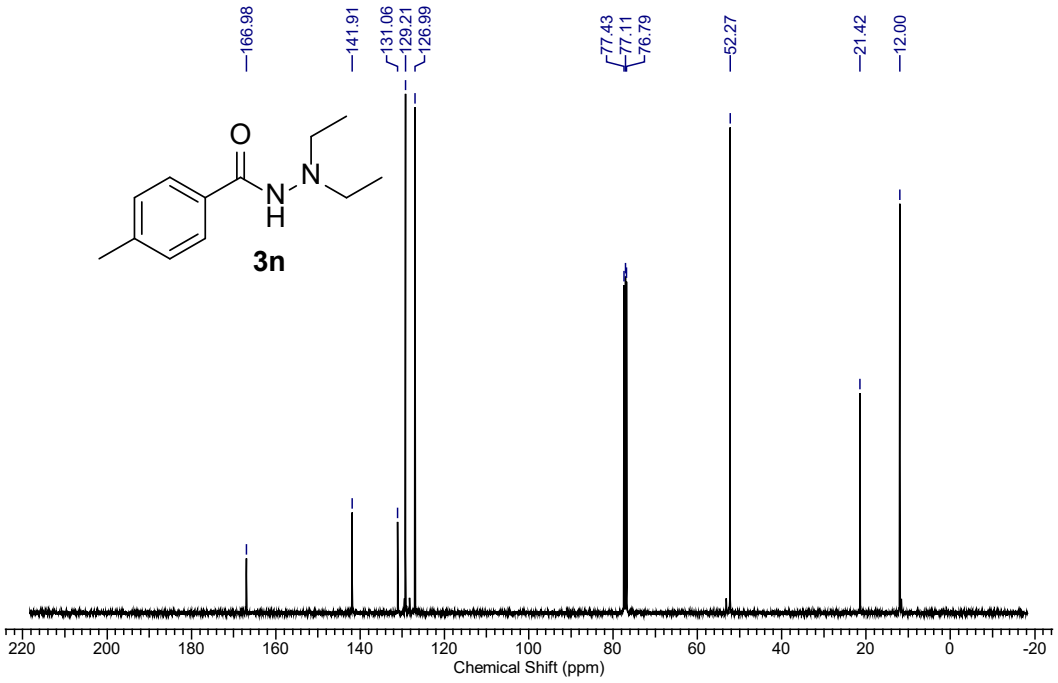


Figure S36. ^{13}C NMR of **3n** (100 MHz, $CDCl_3$)

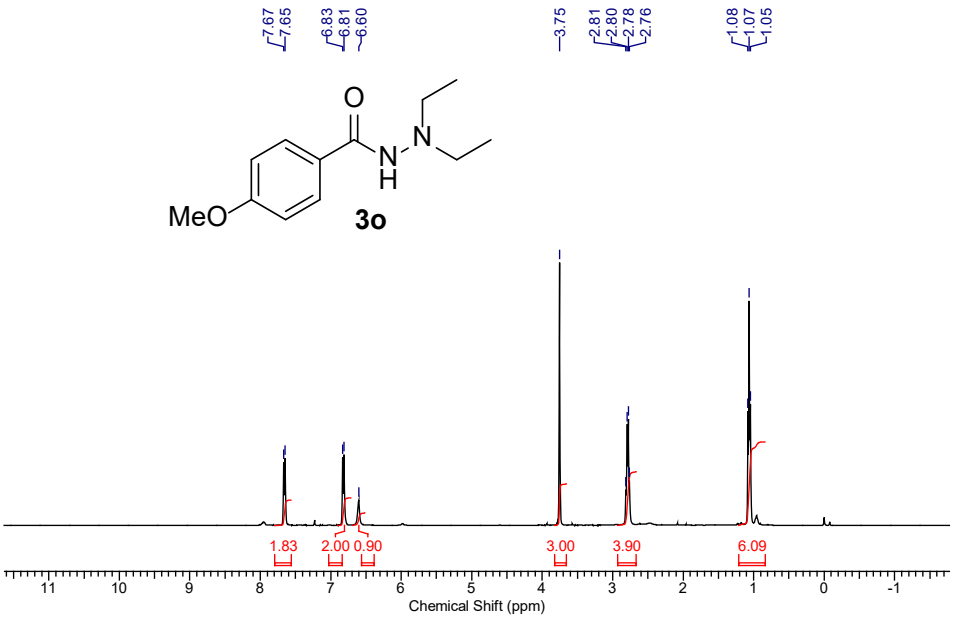


Figure S37. ^{1}H NMR of **3o** (400 MHz, $CDCl_3$)

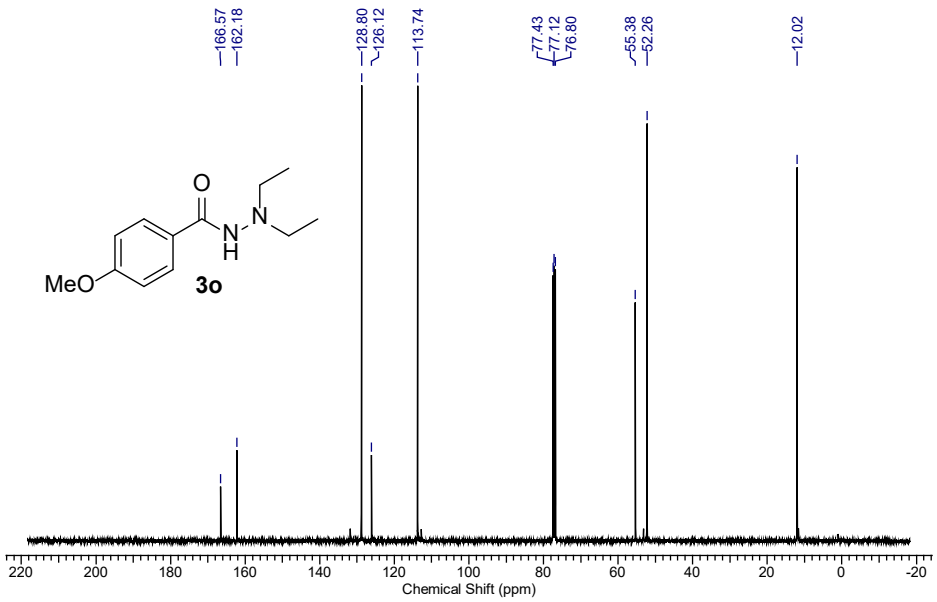


Figure S38. ^{13}C NMR of **3o** (100 MHz, $CDCl_3$)

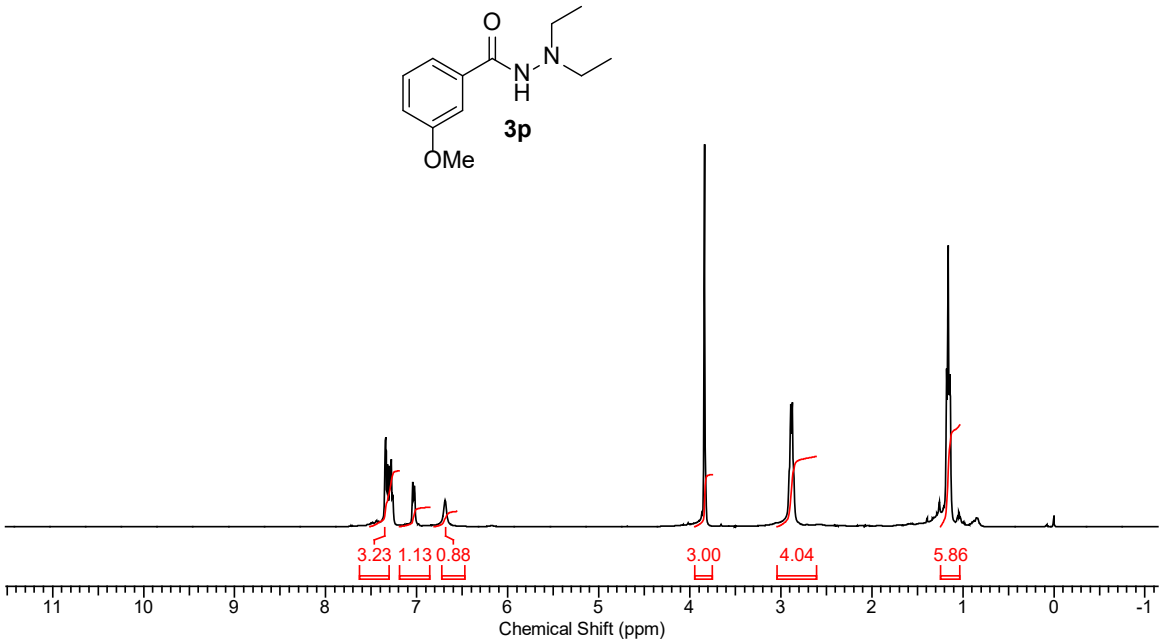


Figure S39. ^{1}H NMR of **3p** (400 MHz, $CDCl_3$)

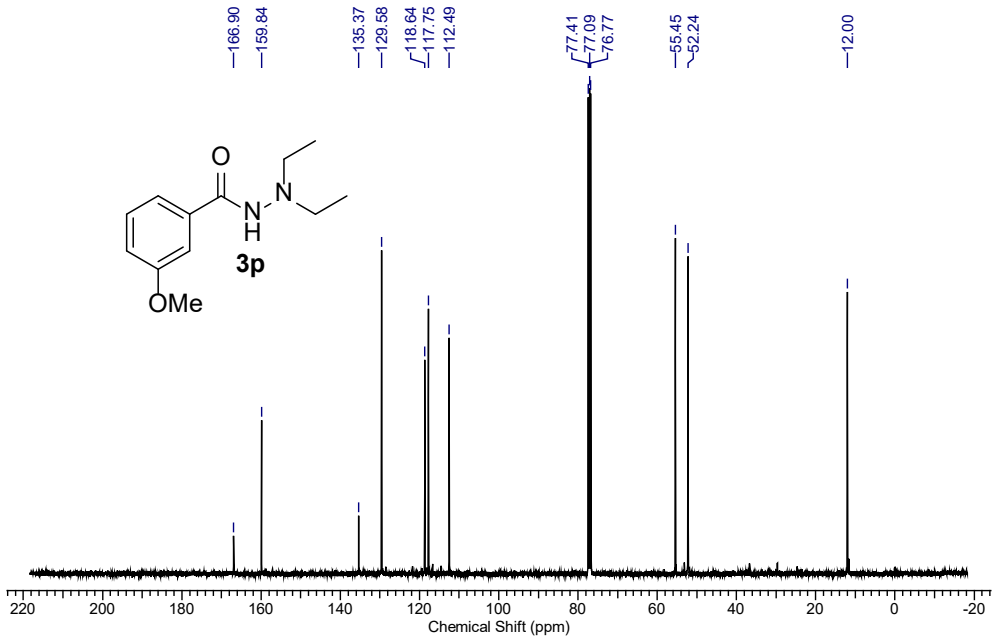


Figure S40. ^{13}C NMR of **3p** (100 MHz, $CDCl_3$)

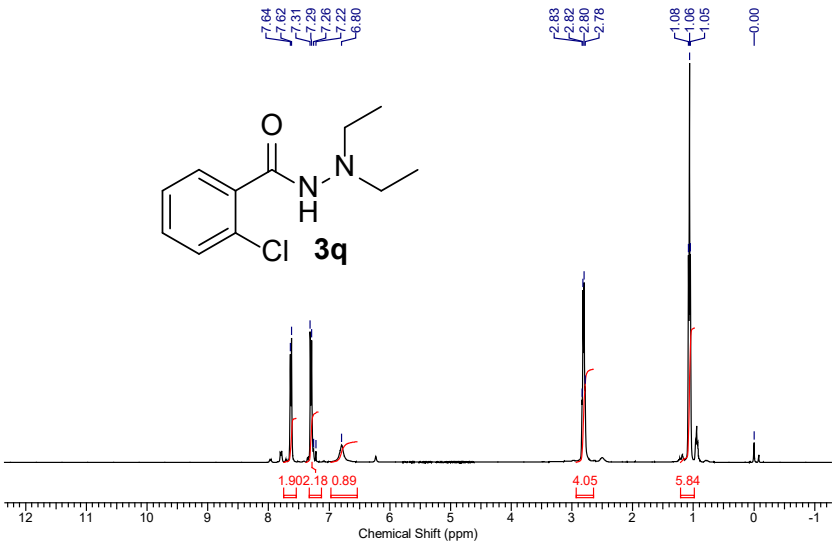


Figure S41. ^{1}H NMR of **3q** (400 MHz, $CDCl_3$)

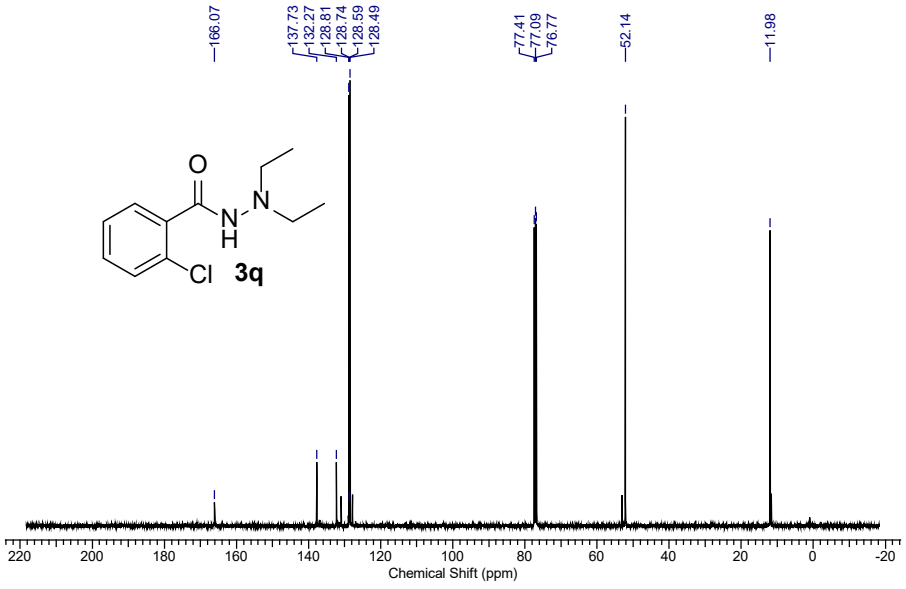


Figure S42. ^{13}C NMR of **3q** (100 MHz, $CDCl_3$)

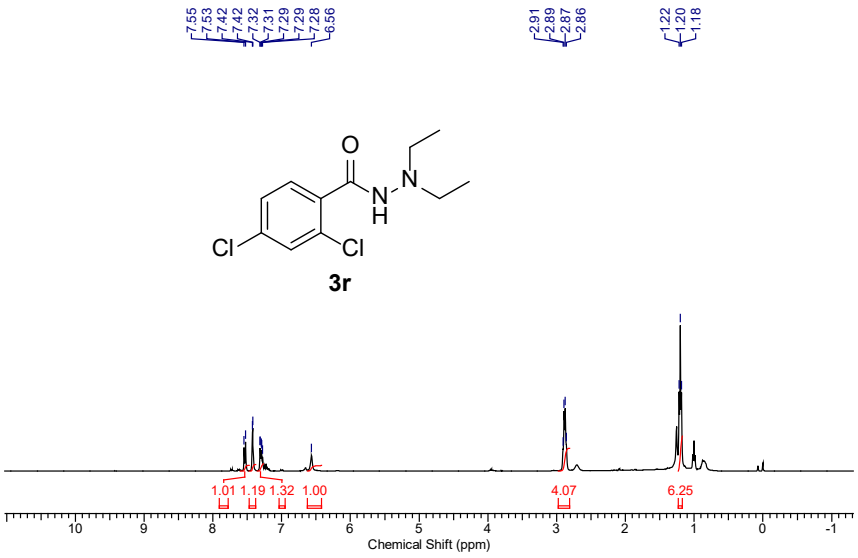


Figure S43. ^{1}H NMR of **3r** (400 MHz, $CDCl_3$)

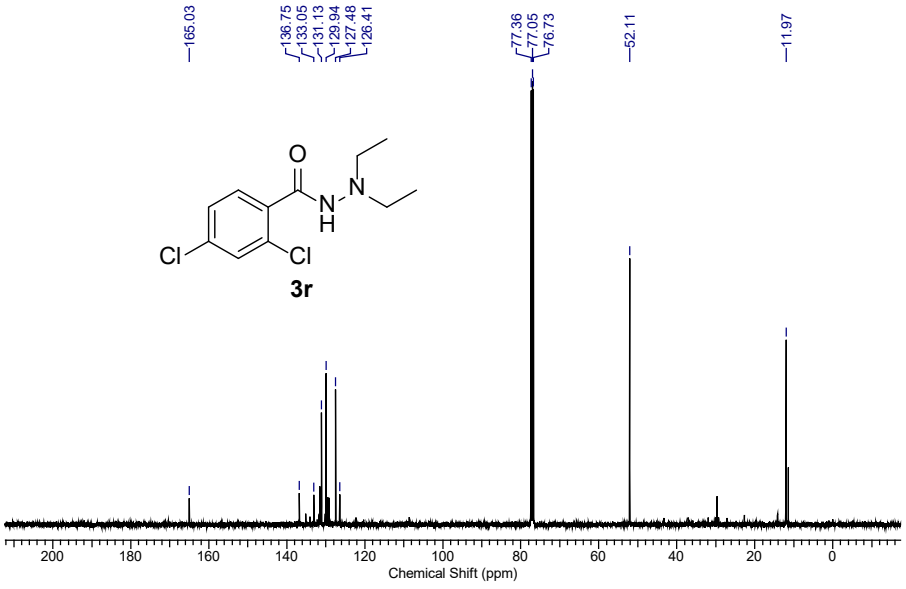


Figure S44. ^{13}C NMR of **3r** (100 MHz, $CDCl_3$)

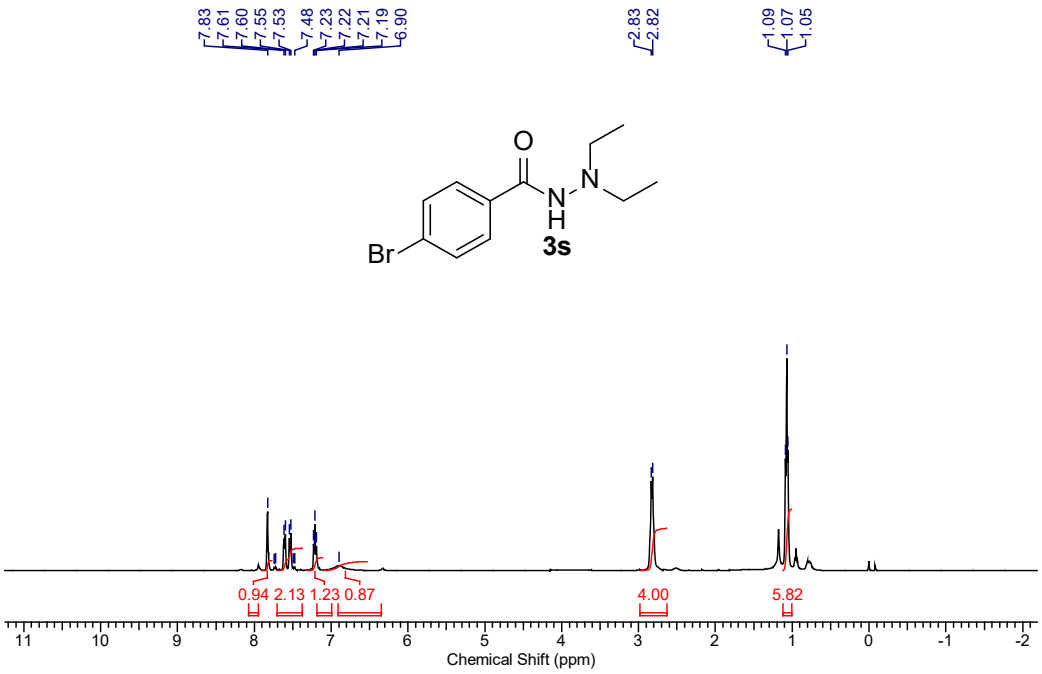


Figure S45. ^{1}H NMR of **3s** (400 MHz, $CDCl_3$)

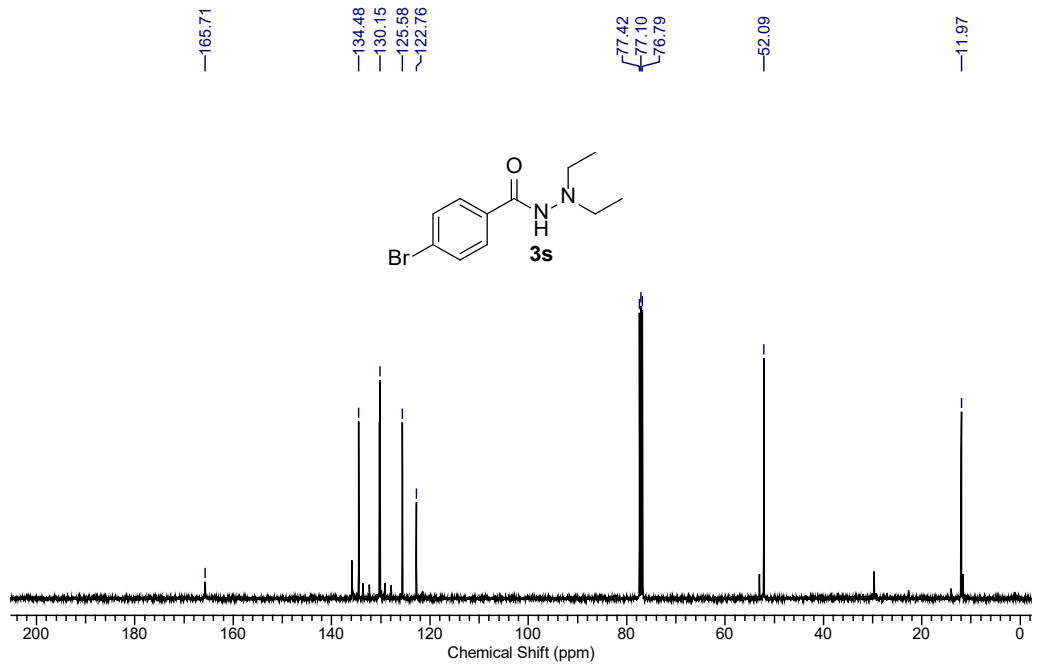


Figure S46. ^{13}C NMR of **3s** (100 MHz, $CDCl_3$)

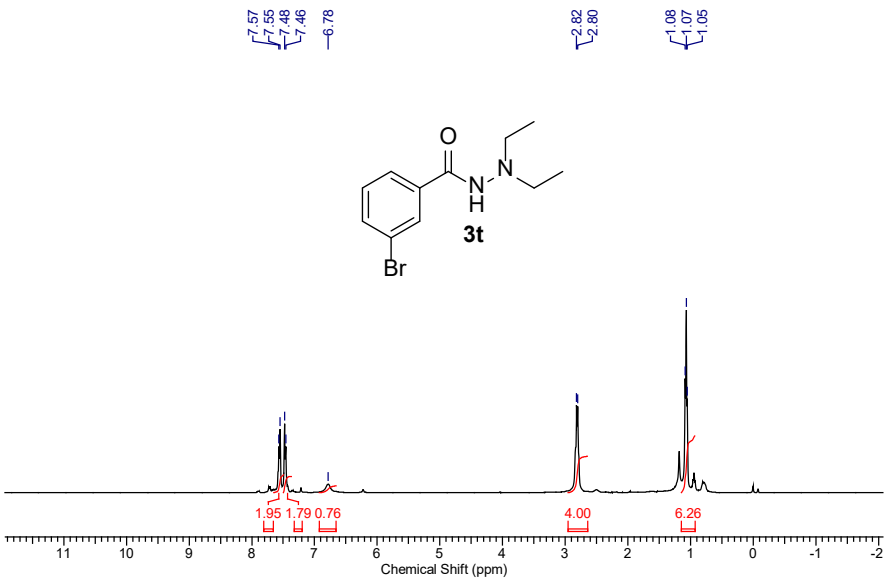


Figure S47. ^{1}H NMR of **3t** (400 MHz, $CDCl_3$)

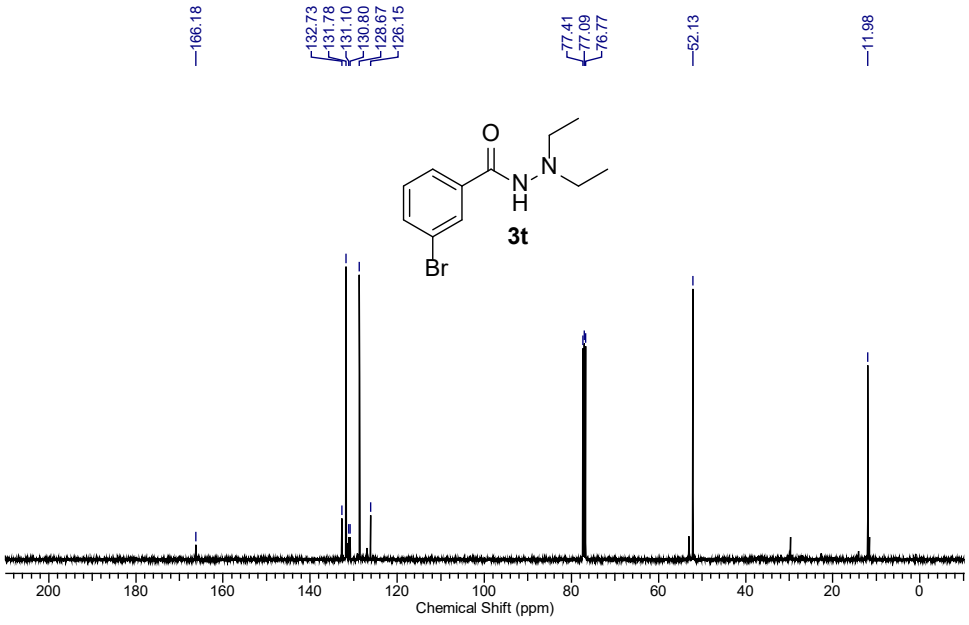


Figure S48. ^{13}C NMR of **3t** (100 MHz, $CDCl_3$)

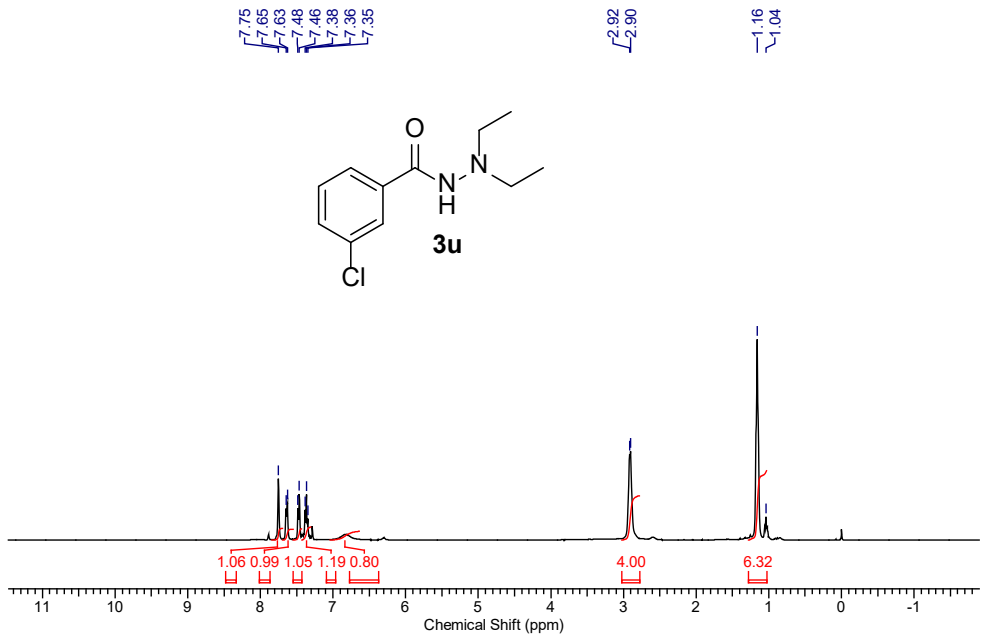


Figure S49. ^{1}H NMR of **3u** (400 MHz, $CDCl_3$)

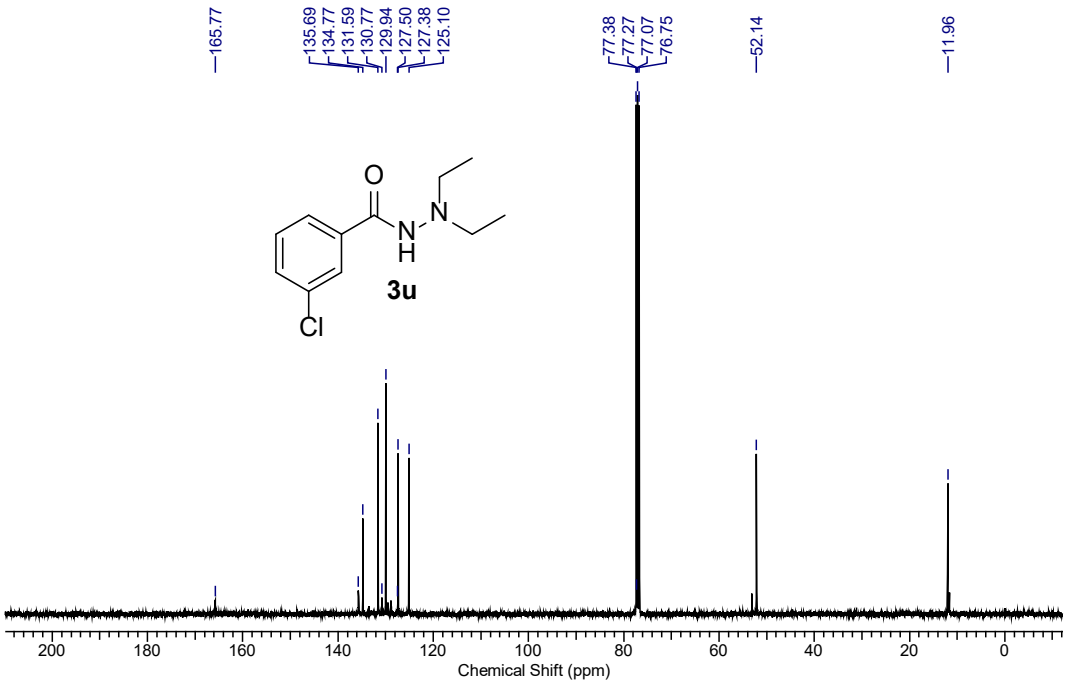


Figure S50. ^{13}C NMR of **3u** (100 MHz, $CDCl_3$)

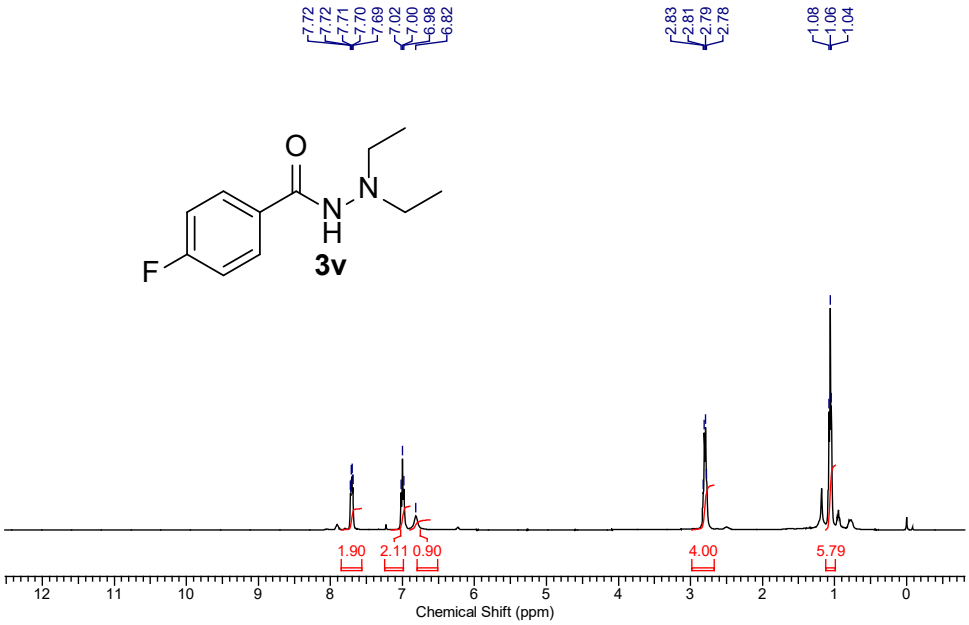


Figure S51. ^{1}H NMR of **3v** (400 MHz, $CDCl_3$)

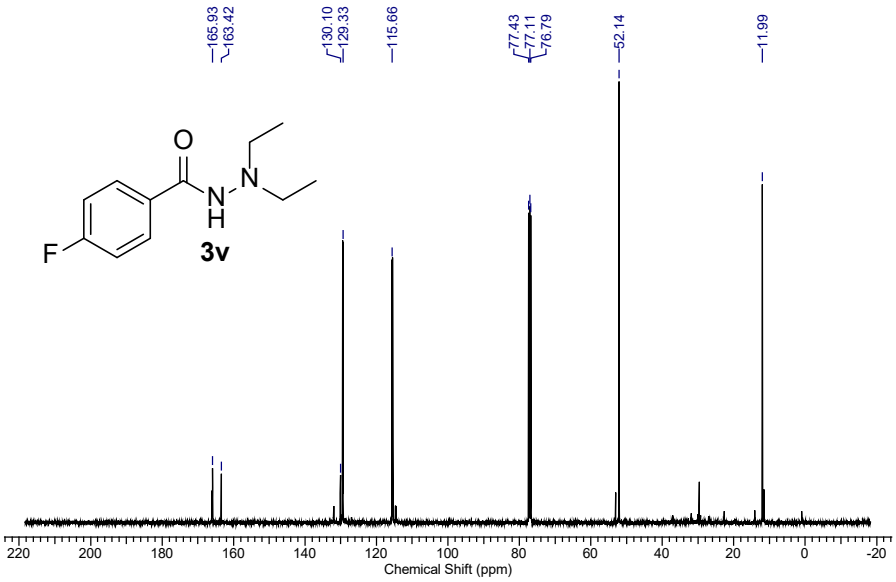


Figure S52. ^{13}C NMR of **3v** (100 MHz, $CDCl_3$)

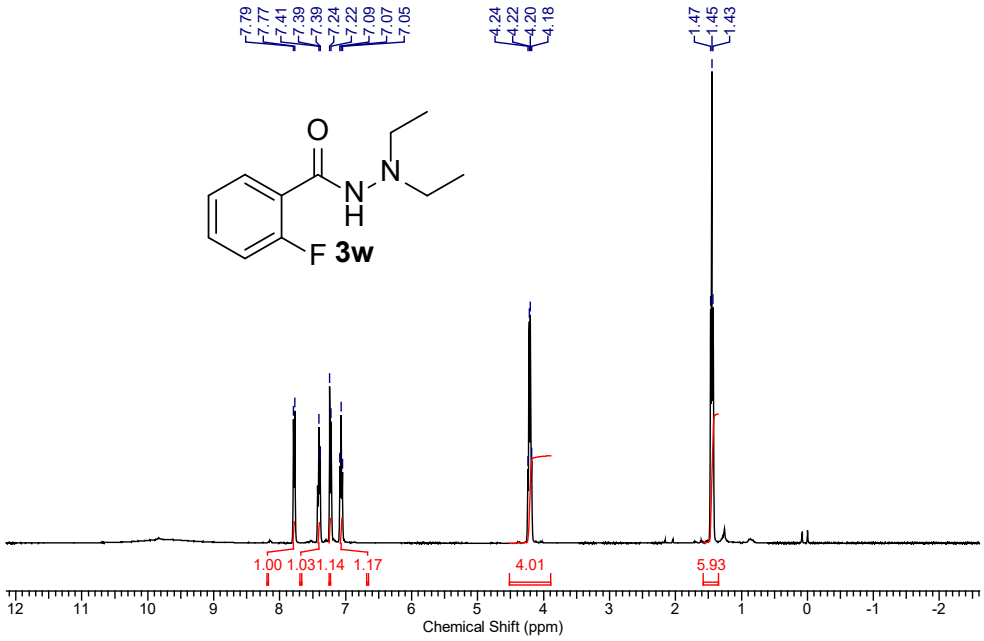


Figure S53. ^{1}H NMR of **3w** (400 MHz, $CDCl_3$)

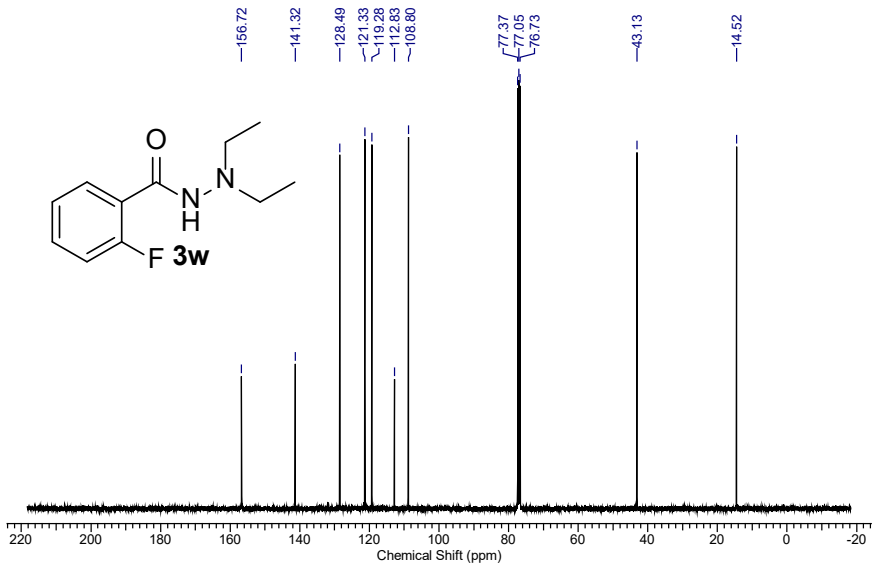


Figure S54. ^{13}C NMR of **3w** (400 MHz, $CDCl_3$)

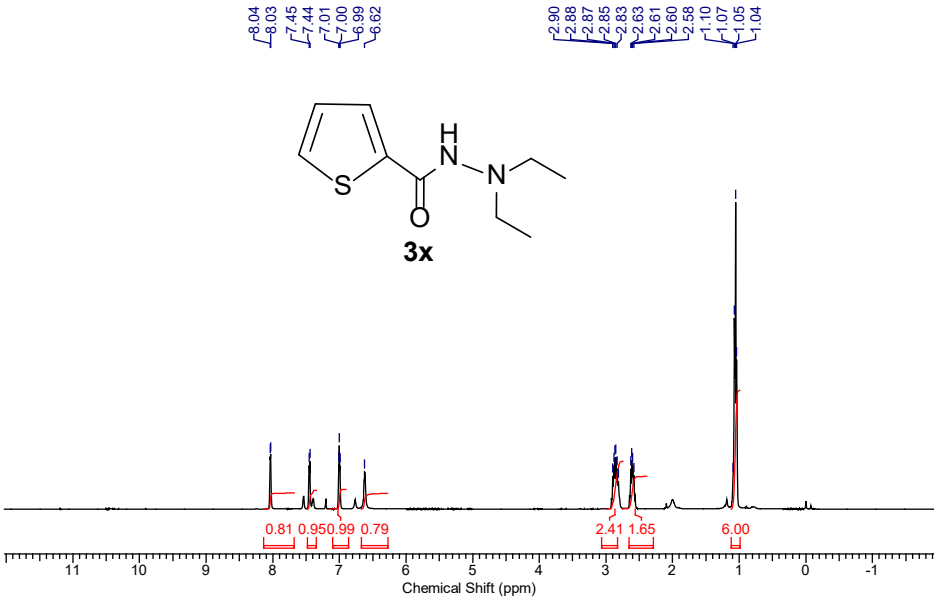


Figure S55. ^{1}H NMR of **3x** (400 MHz, $CDCl_3$)

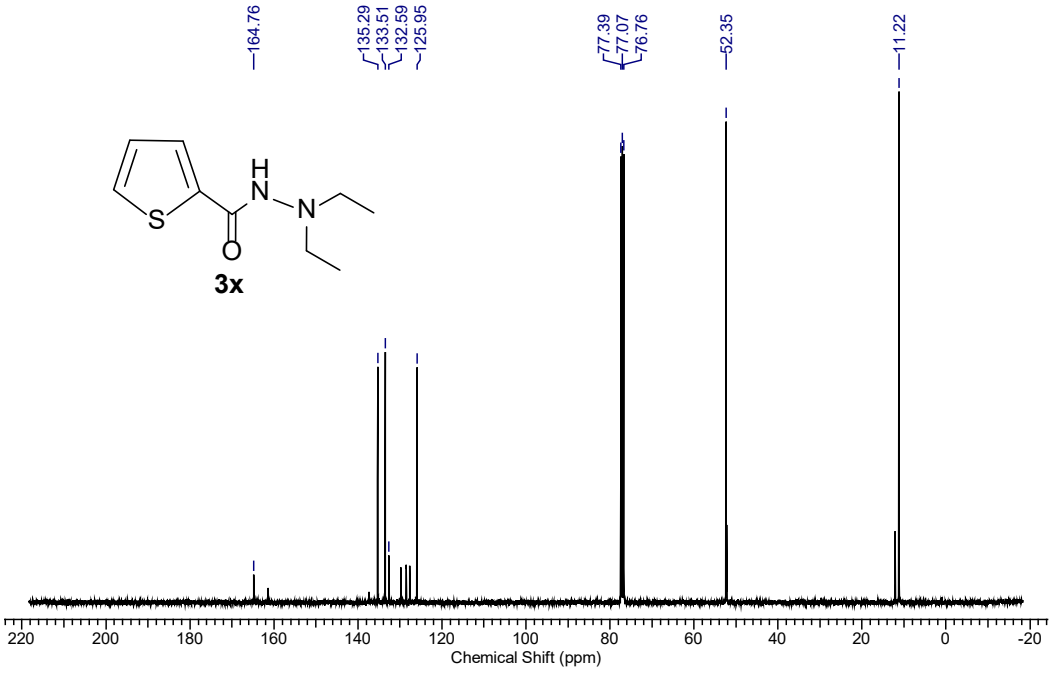


Figure S56. ^{13}C NMR of **3x** (100 MHz, $CDCl_3$)

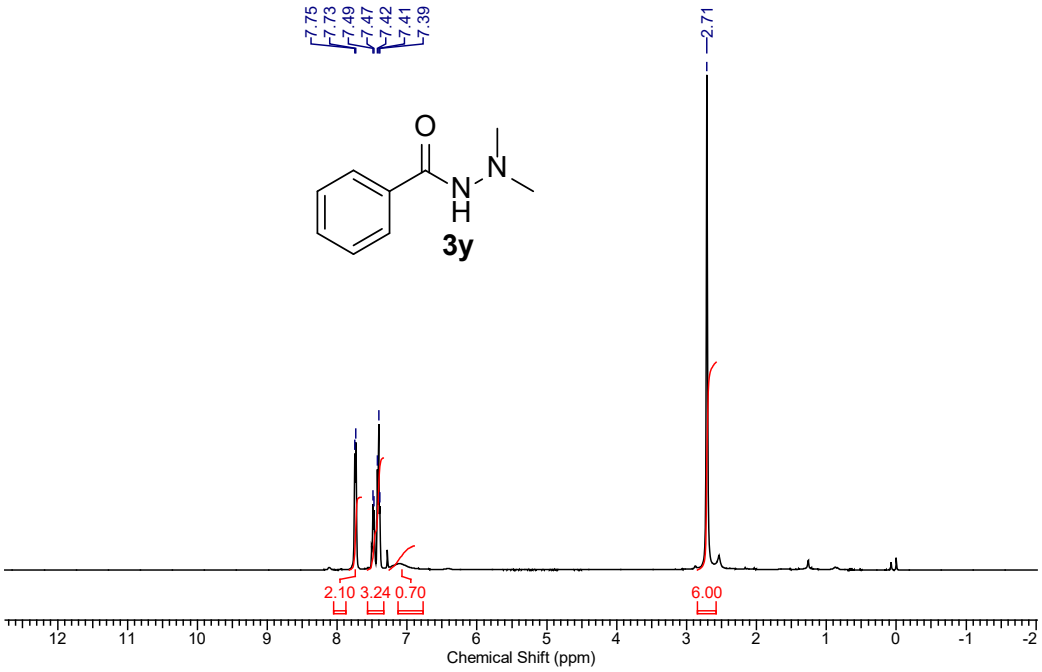


Figure S57. ^{1}H NMR of **3y** (400 MHz, $CDCl_3$)

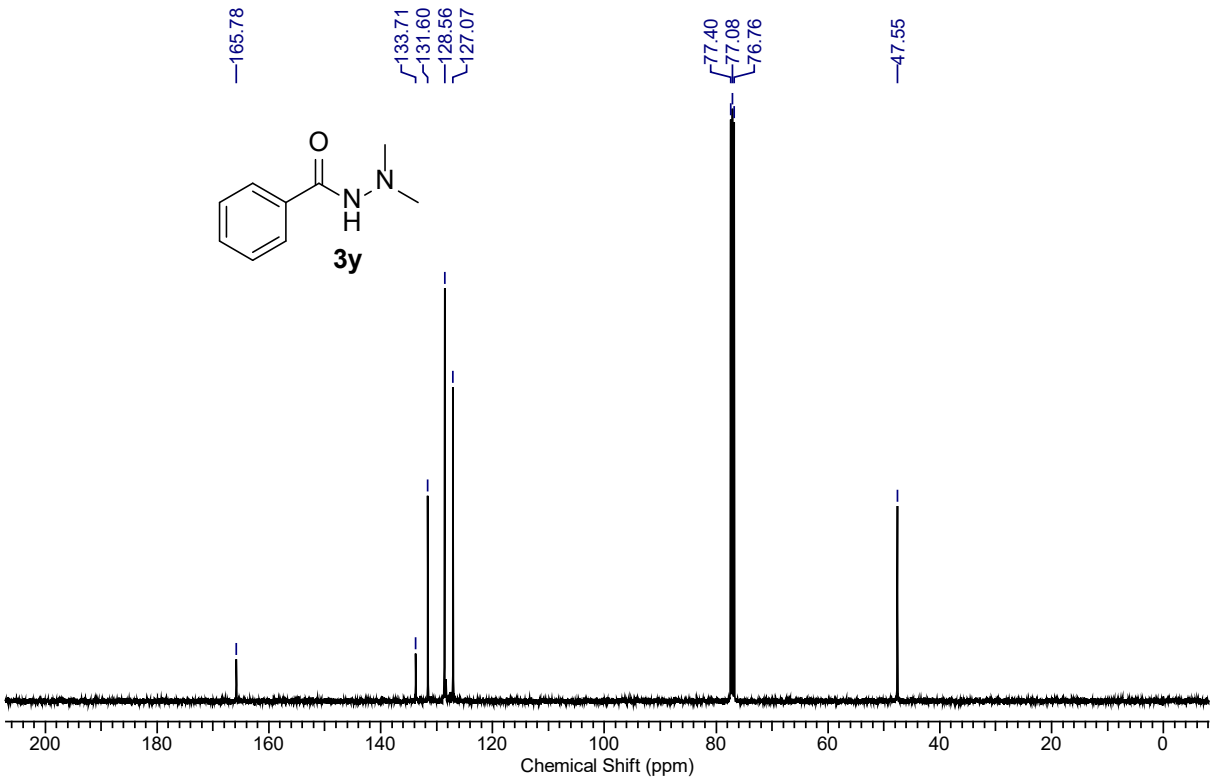


Figure S58. ^{13}C NMR of **3y** (100 MHz, $CDCl_3$)

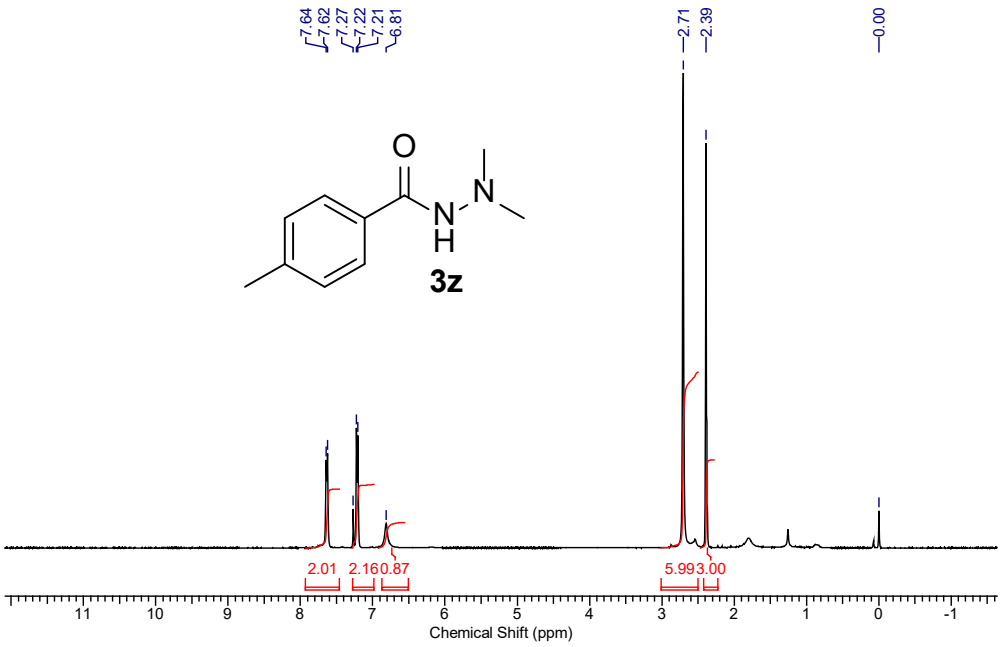


Figure S59. ^{1}H NMR of **3z** (400 MHz, $CDCl_3$)

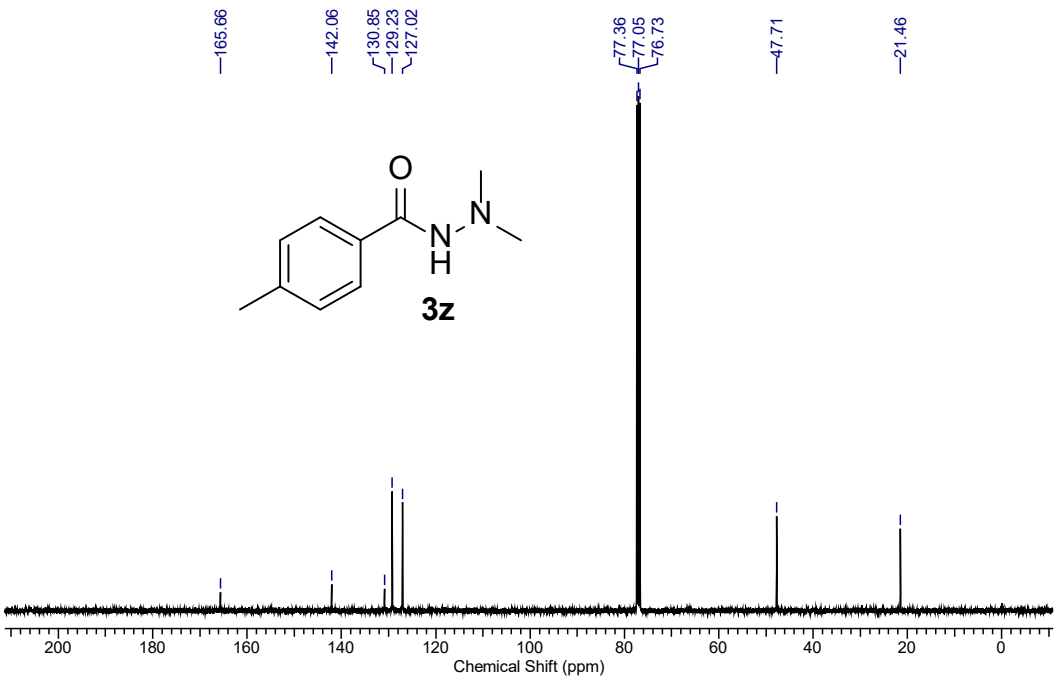


Figure S60. ^{13}C NMR of **3z** (100 MHz, $CDCl_3$)

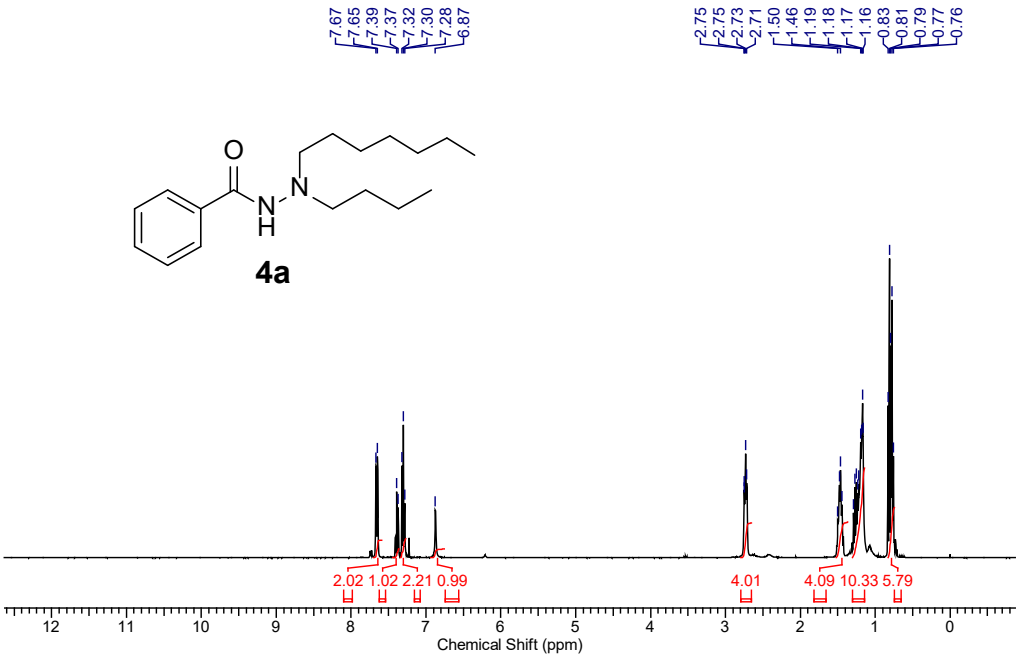


Figure S61. ^{1}H NMR of **4a** (400 MHz, $CDCl_3$)

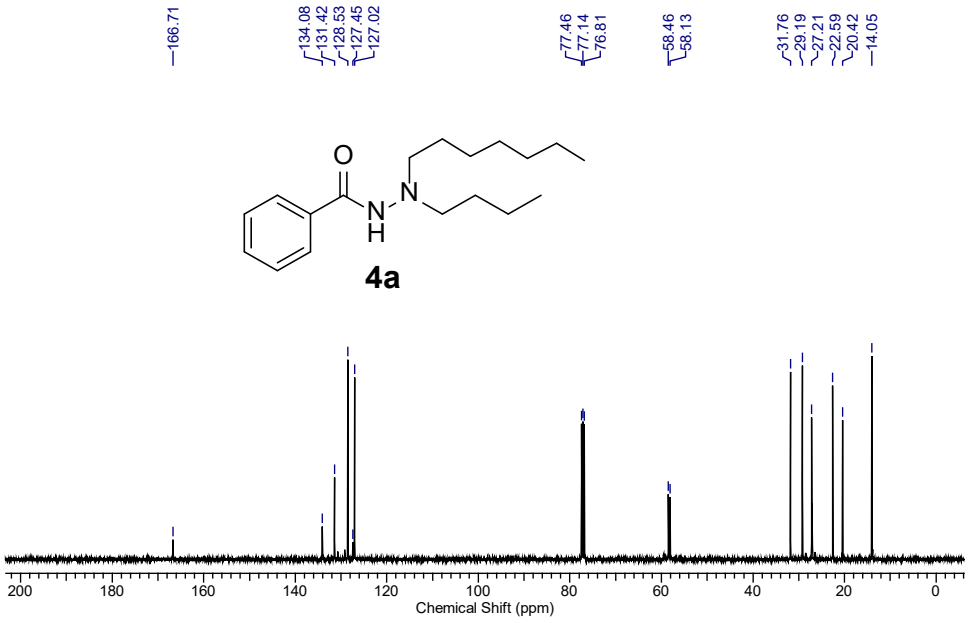


Figure S62. ^{13}C NMR of **4a** (100 MHz, $CDCl_3$)

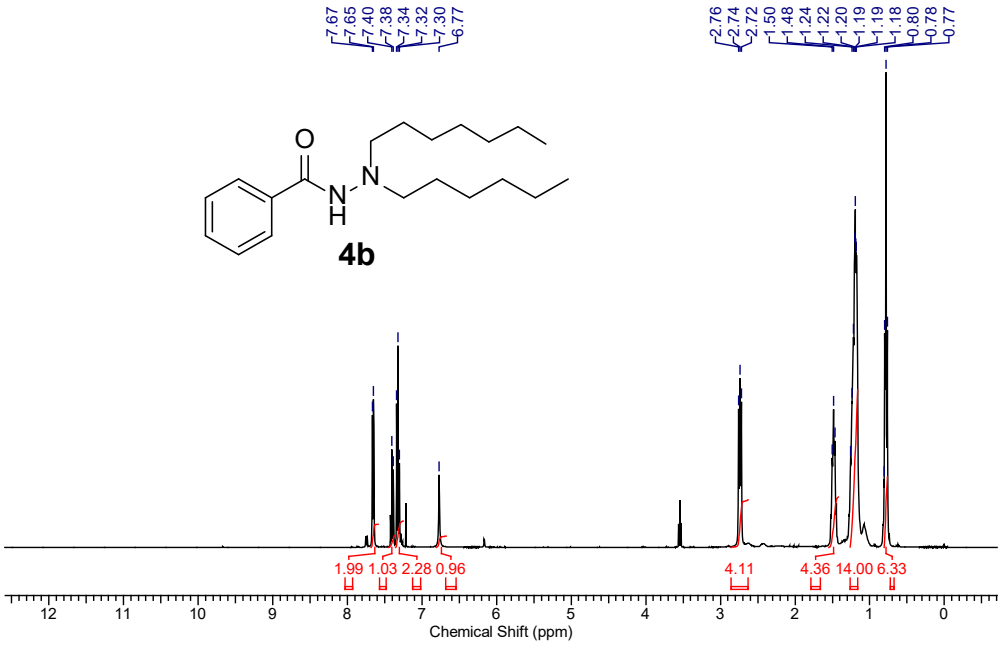


Figure S63. ^{1}H NMR of **4b** (400 MHz, $CDCl_3$)

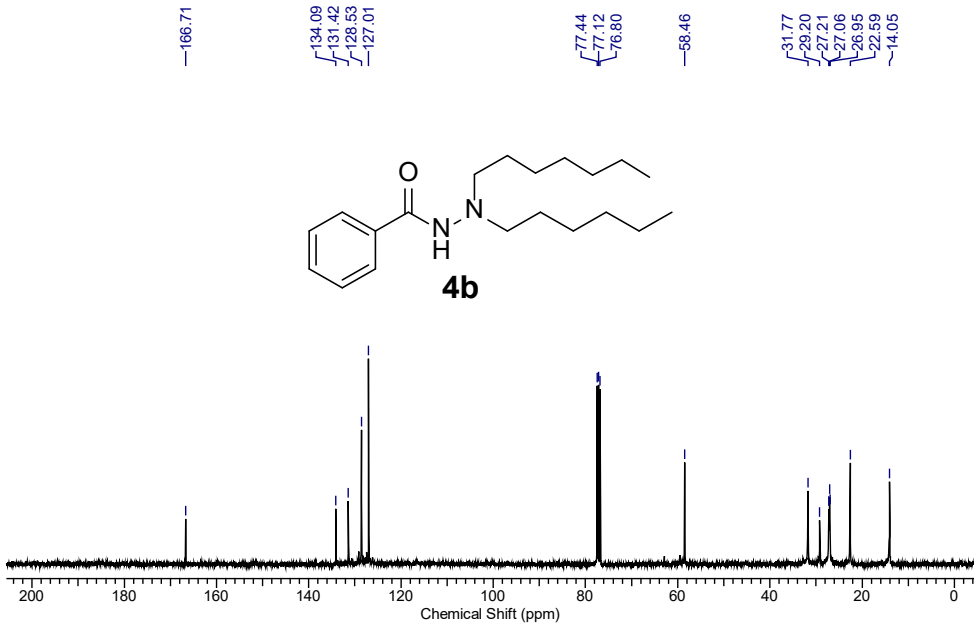


Figure S64. ^{13}C NMR of **4b** (100 MHz, $CDCl_3$)

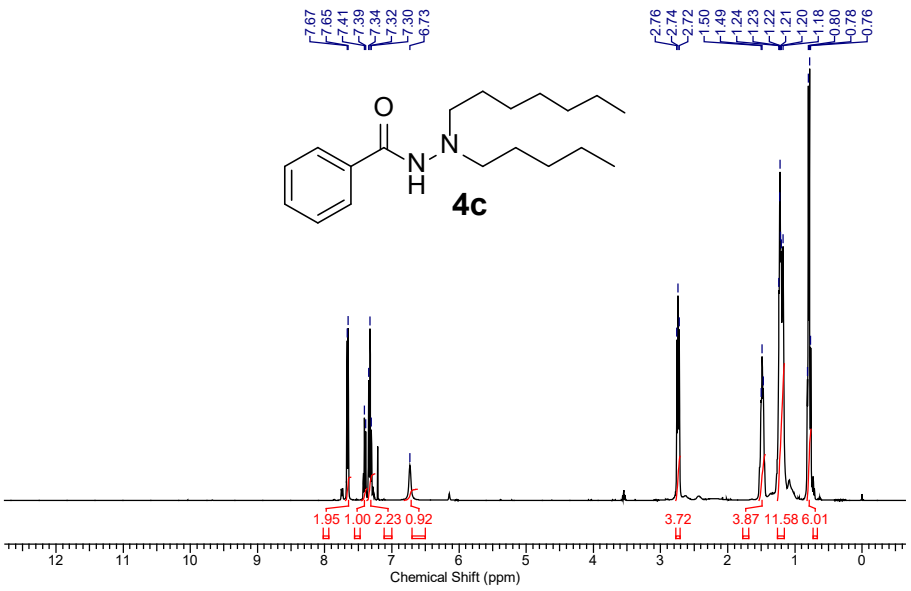


Figure S65. ^{1}H NMR of **4c** (400 MHz, $CDCl_3$)

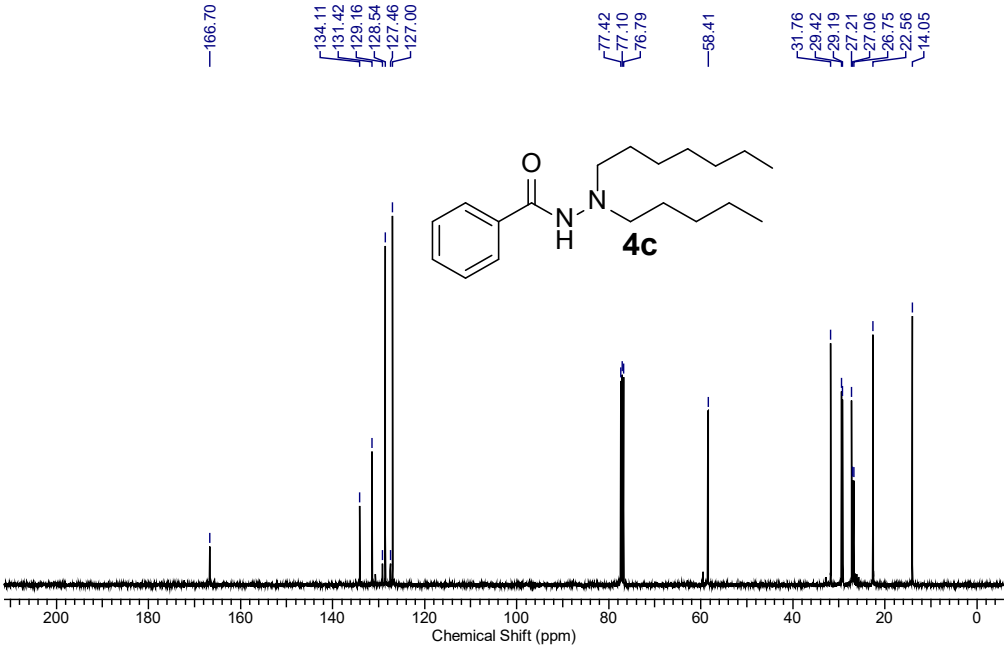


Figure S66. ^{13}C NMR of **4c** (100 MHz, $CDCl_3$)

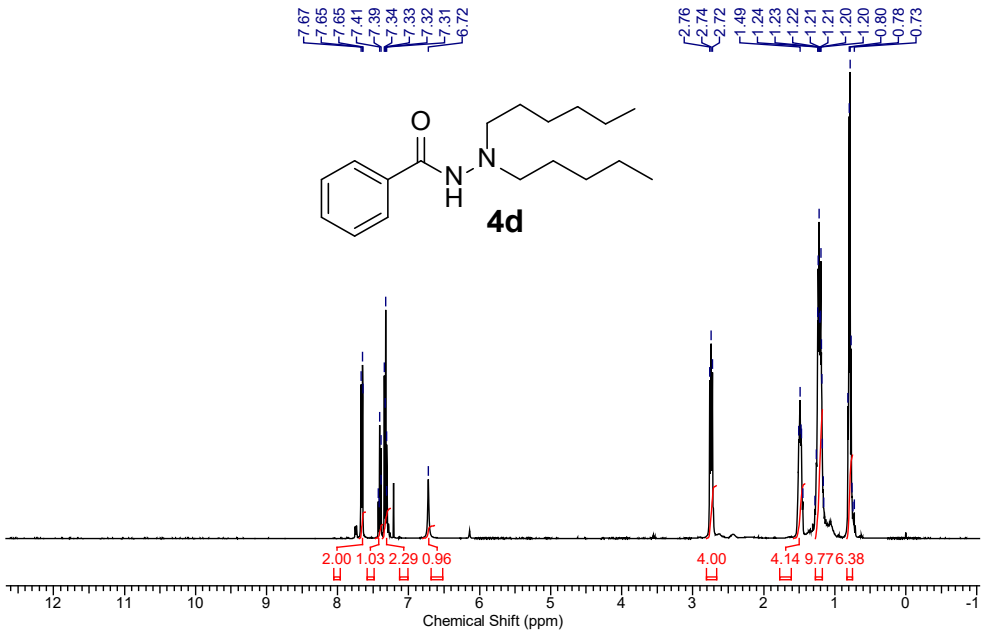


Figure S67. ^{1}H NMR of **4d** (400 MHz, $CDCl_3$)

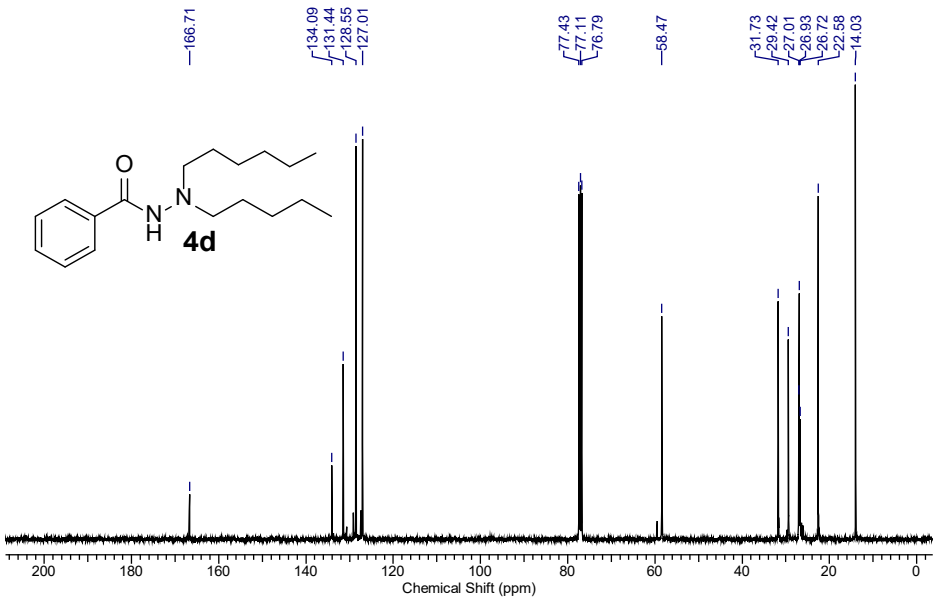


Figure S68. ^{13}C NMR of **4d** (100 MHz, $CDCl_3$)

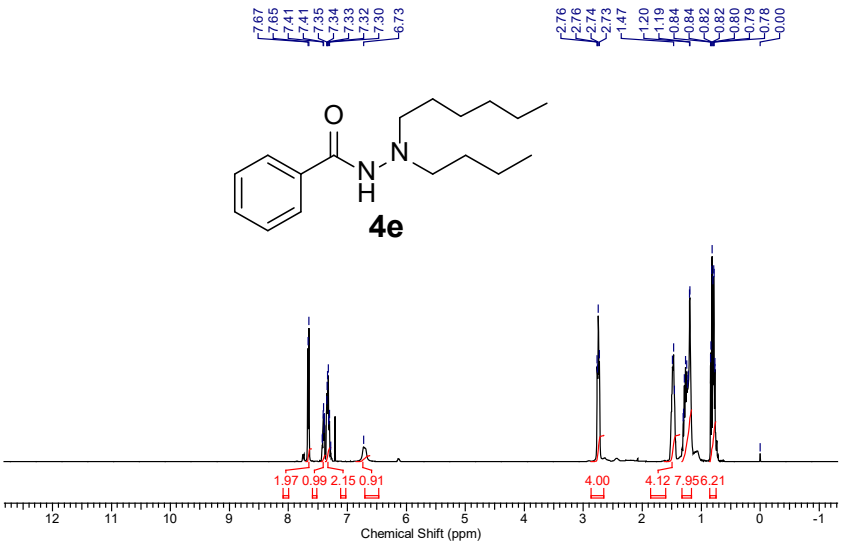


Figure S69. ^{1}H NMR of **4e** (400 MHz, $CDCl_3$)

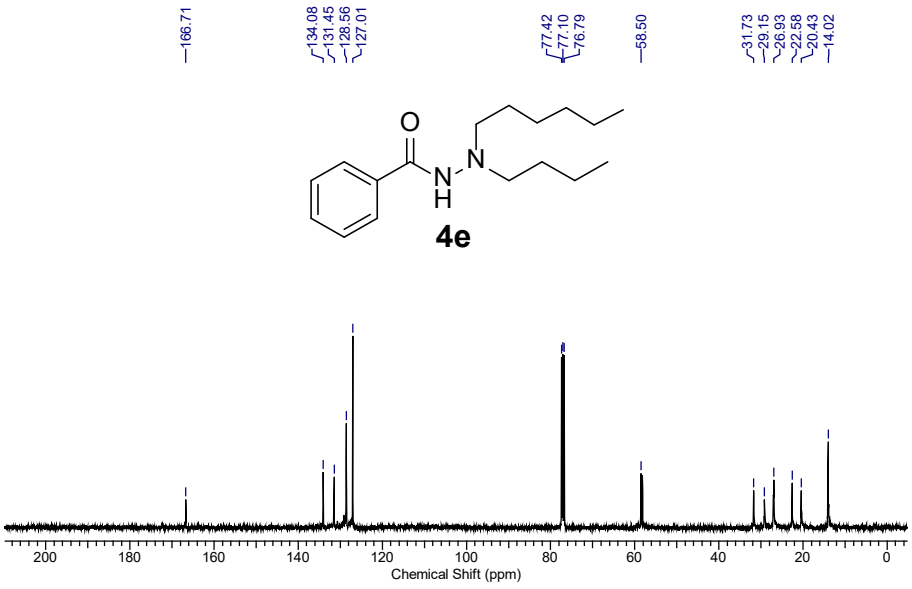


Figure S70. ^{13}C NMR of **4e** (100 MHz, $CDCl_3$)

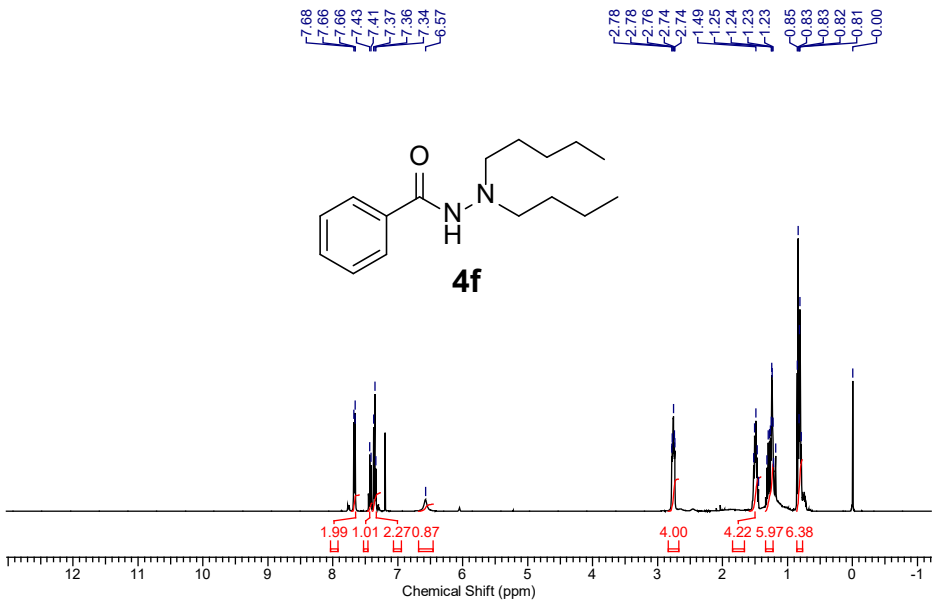


Figure S71. ^{1}H NMR of **4f** (400 MHz, $CDCl_3$)

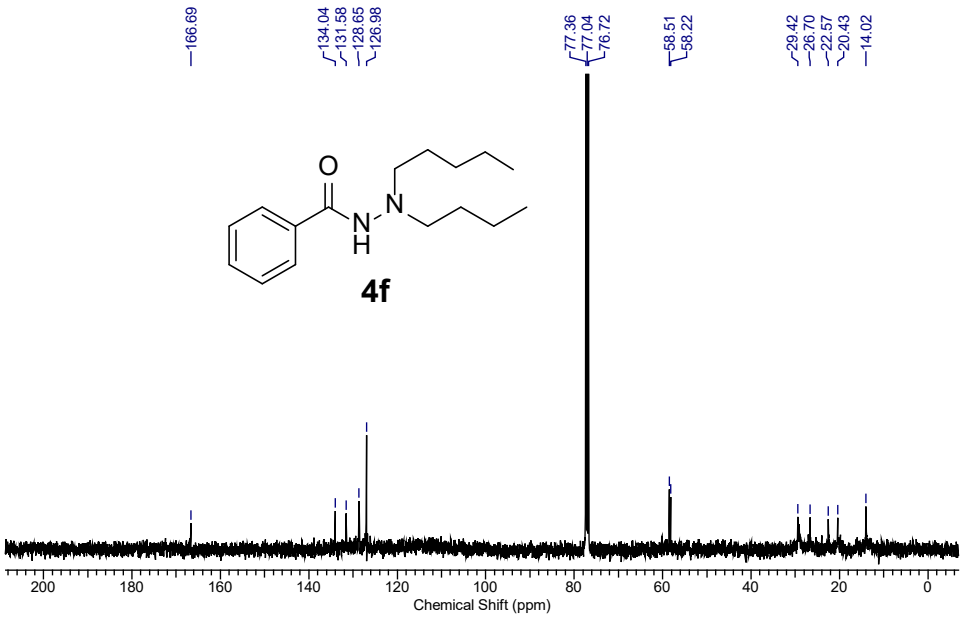


Figure S72. ^{13}C NMR of **4f** (100 MHz, $CDCl_3$)

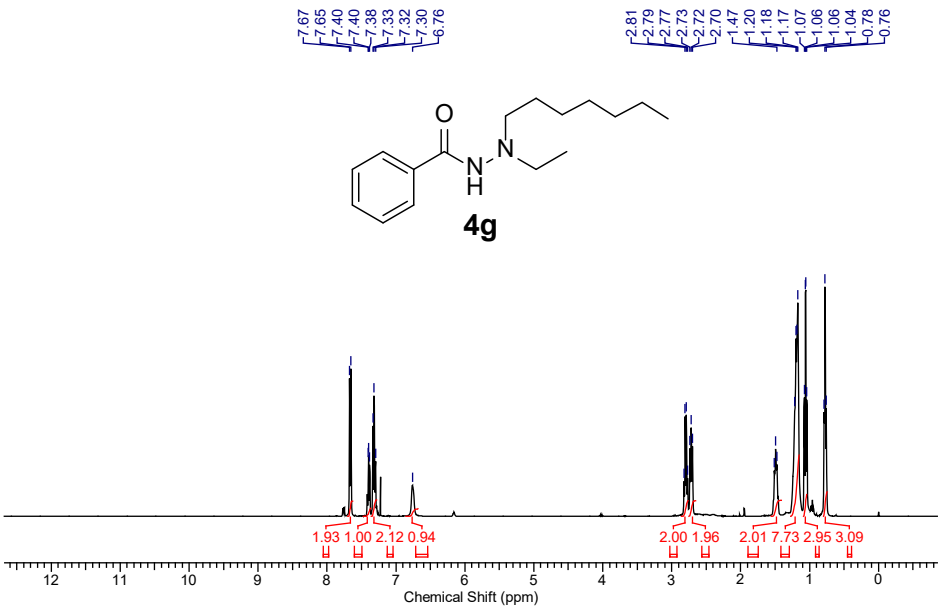


Figure S73. ^{1}H NMR of **4g** (400 MHz, $CDCl_3$)

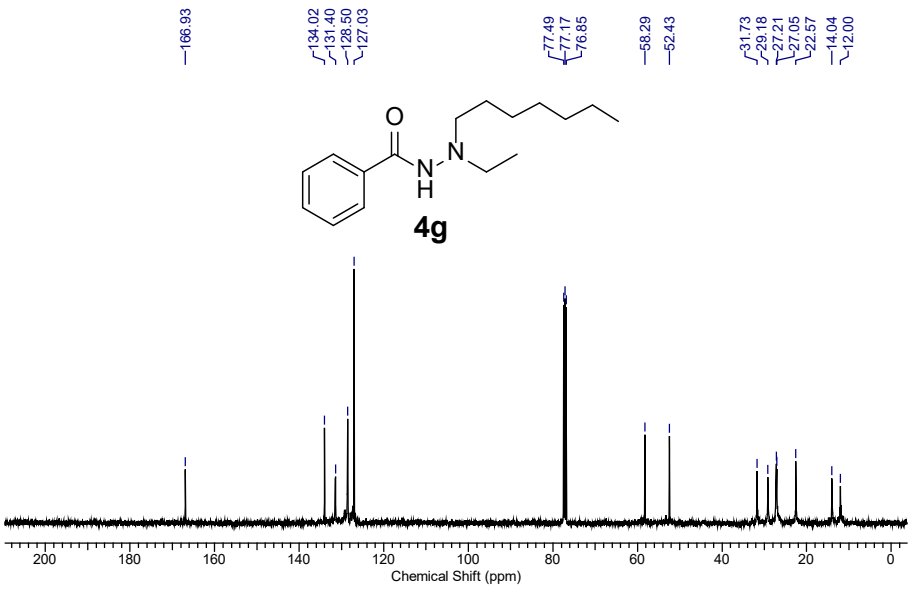


Figure S74. ^{13}C NMR of **4g** (100 MHz, $CDCl_3$)

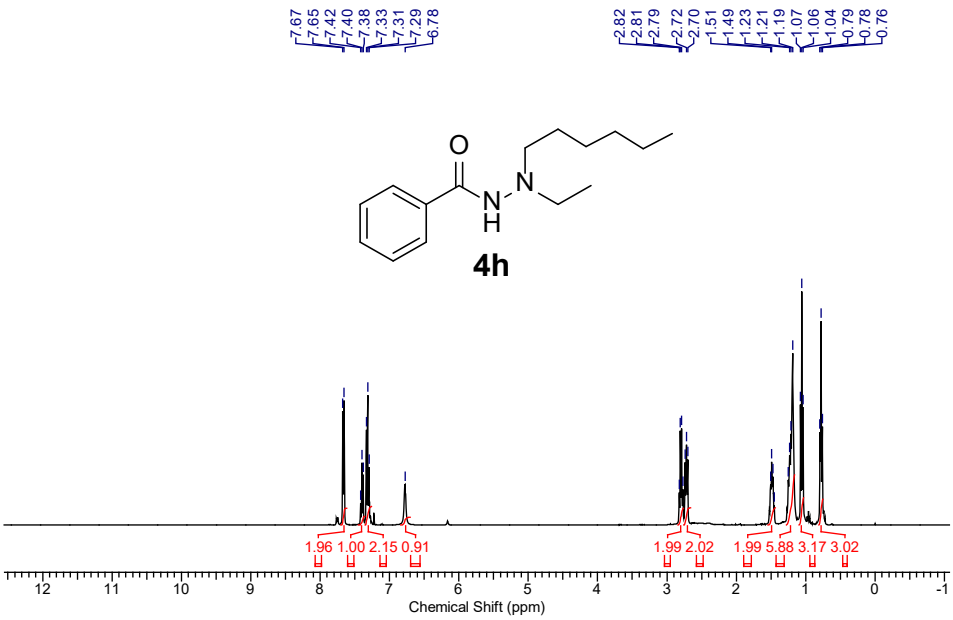


Figure S75. ^{1}H NMR of **4h** (400 MHz, $CDCl_3$)

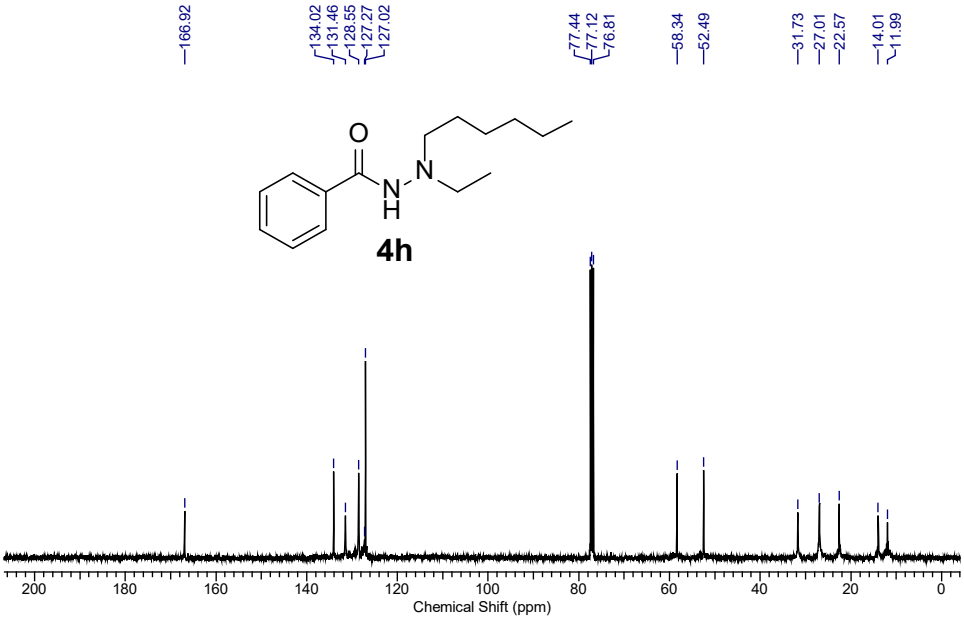


Figure S76. ^{13}C NMR of **4h** (100 MHz, $CDCl_3$)

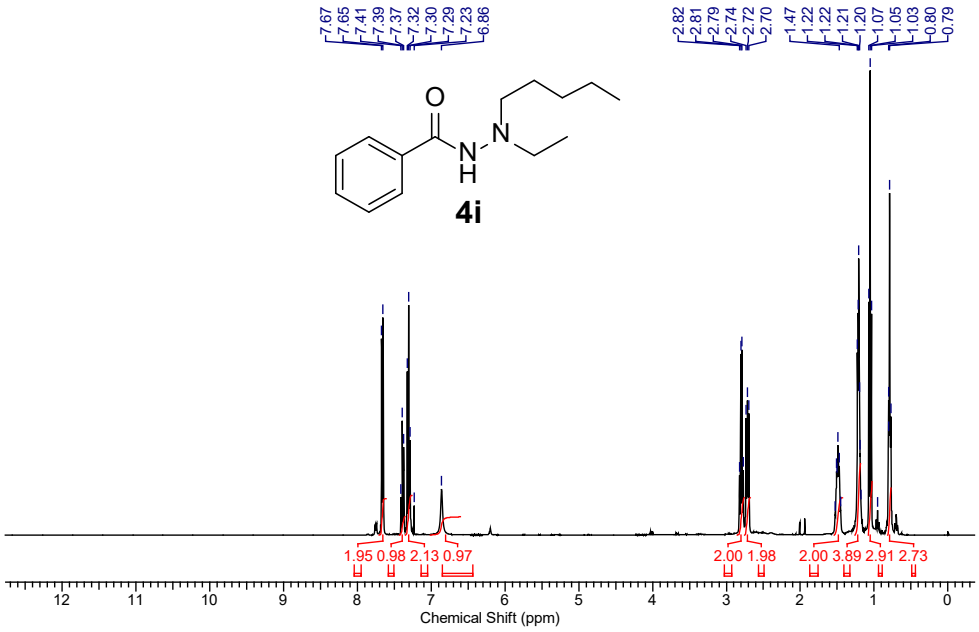


Figure S77. ^{1}H NMR of **4i** (400 MHz, $CDCl_3$)

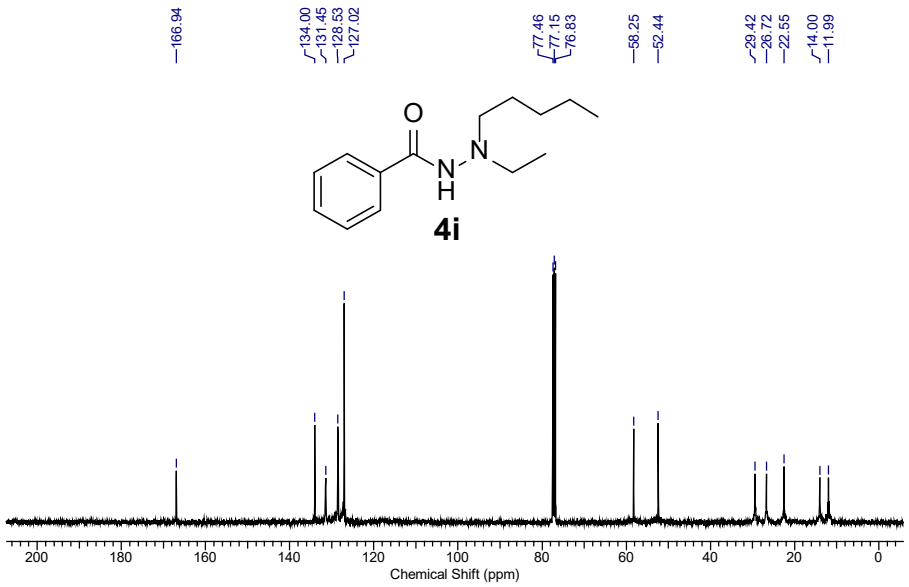


Figure S78. ^{13}C NMR of **4i** (100 MHz, $CDCl_3$)

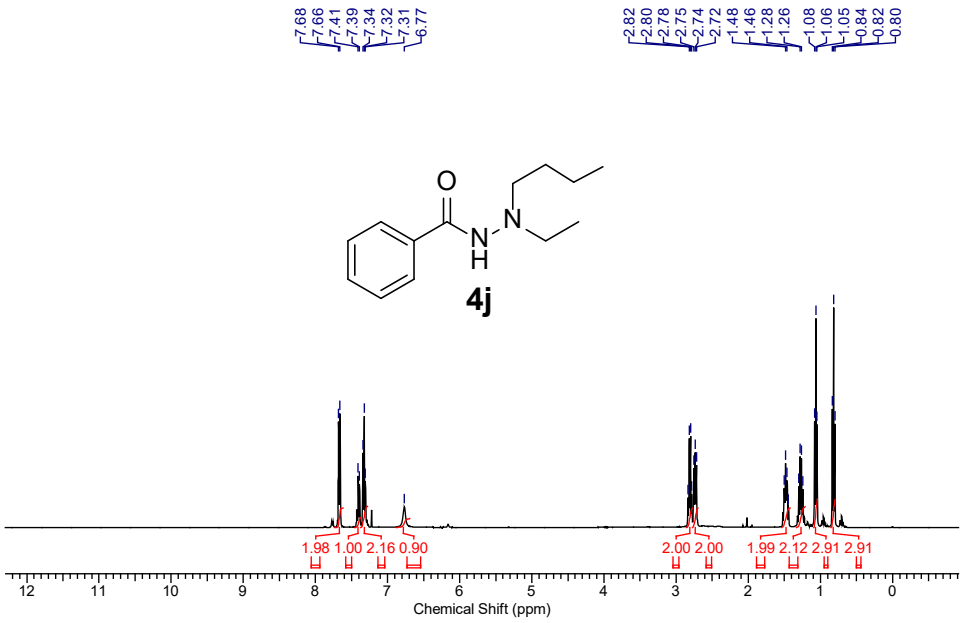


Figure S79. ^{1}H NMR of **4j** (400 MHz, $CDCl_3$)

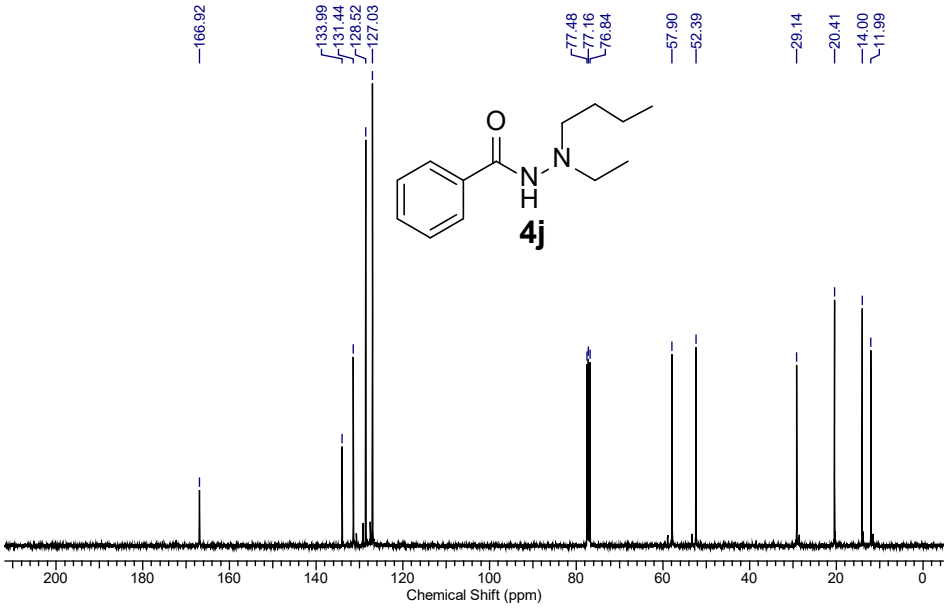


Figure S80. ^{13}C NMR of **4j** (100 MHz, $CDCl_3$)

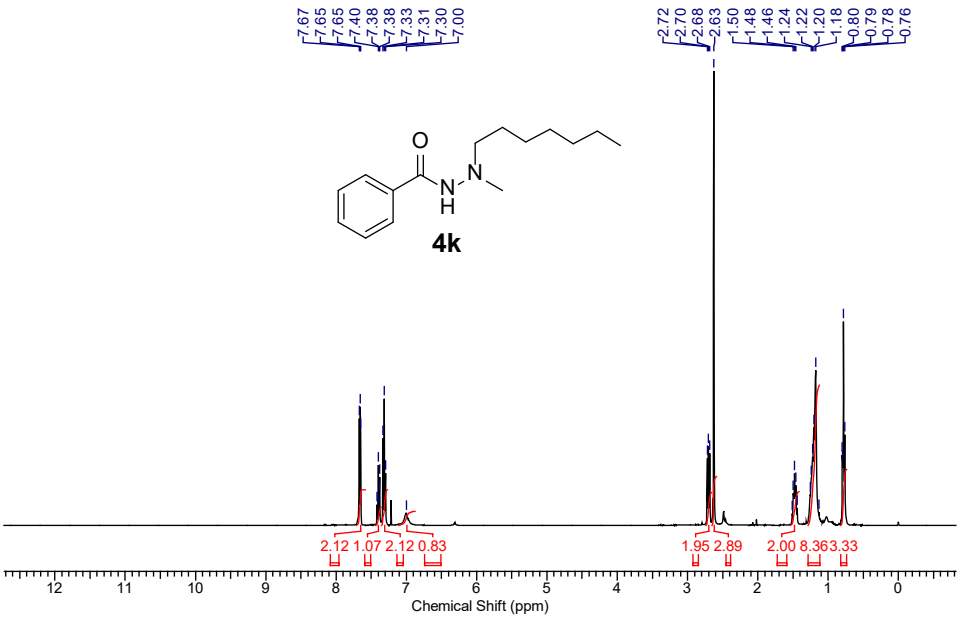


Figure S81. ^{1}H NMR of **4k** (400 MHz, $CDCl_3$)

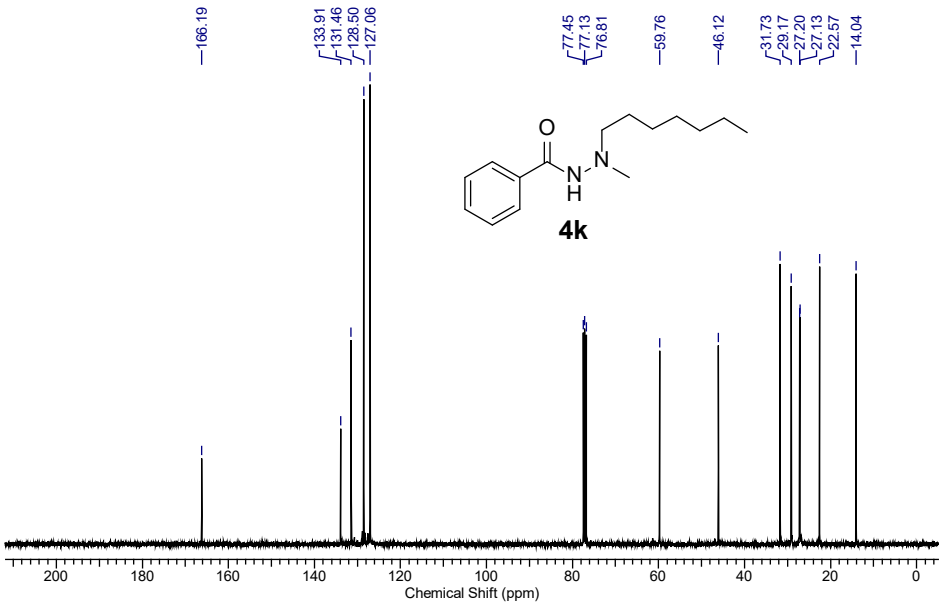


Figure S82. ^{13}C NMR of **4k** (100 MHz, $CDCl_3$)

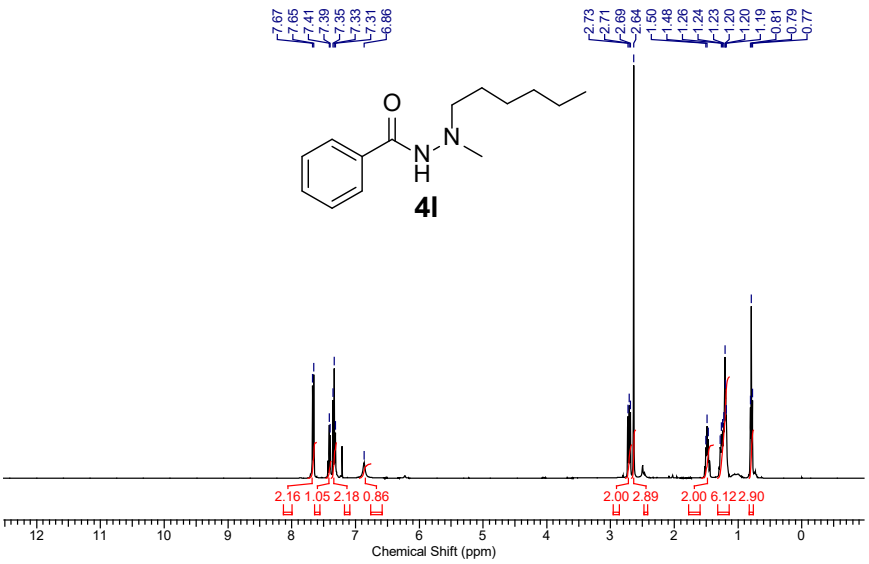


Figure S83. ^{1}H NMR of **4l** (400 MHz, $CDCl_3$)

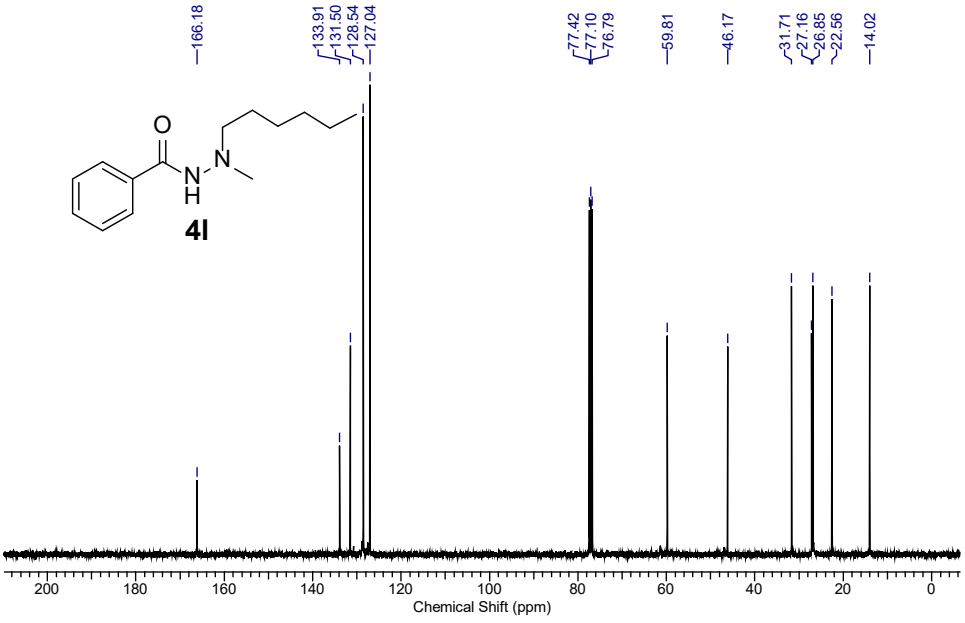


Figure S84. ^{13}C NMR of **4l** (100 MHz, $CDCl_3$)

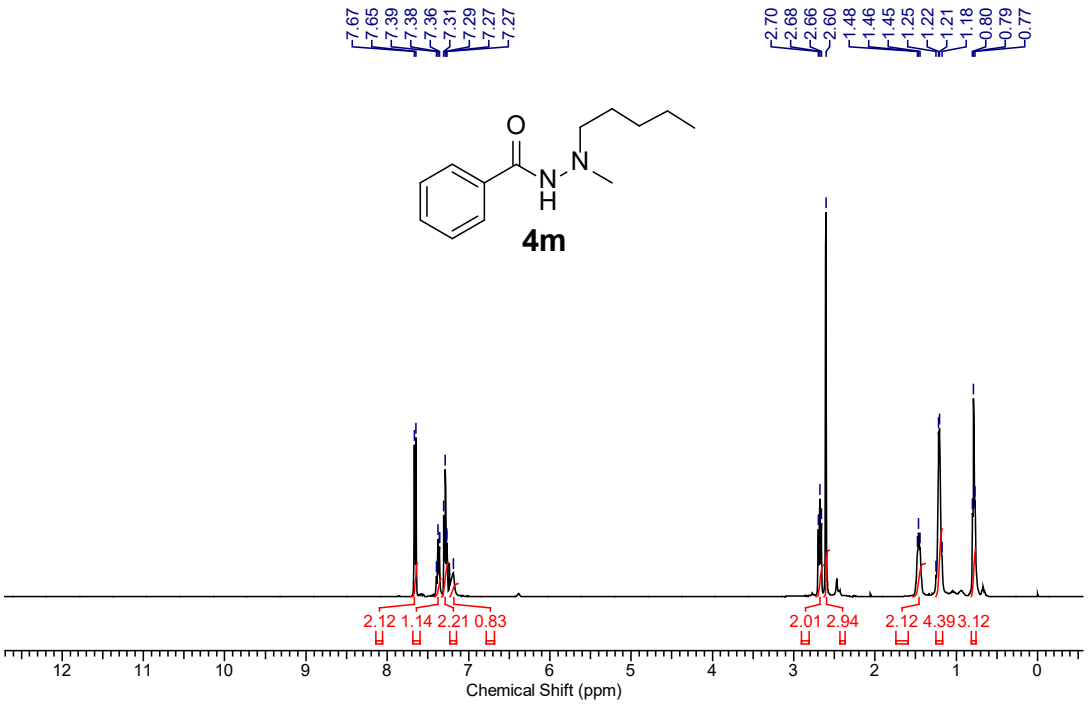


Figure S85. ^{1}H NMR of **4m** (400 MHz, $CDCl_3$)

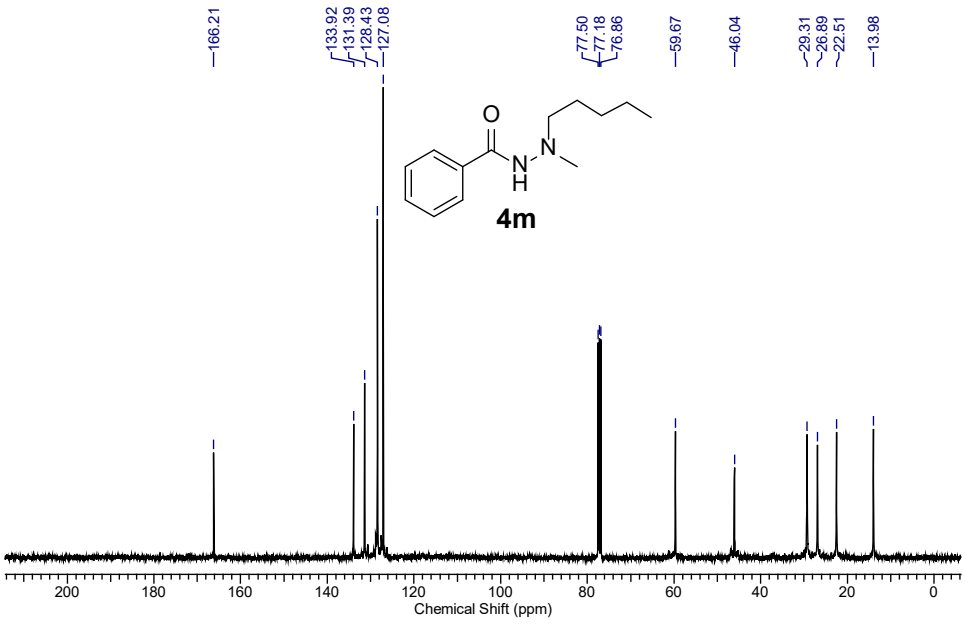


Figure S86. ^{13}C NMR of **4m** (100 MHz, $CDCl_3$)

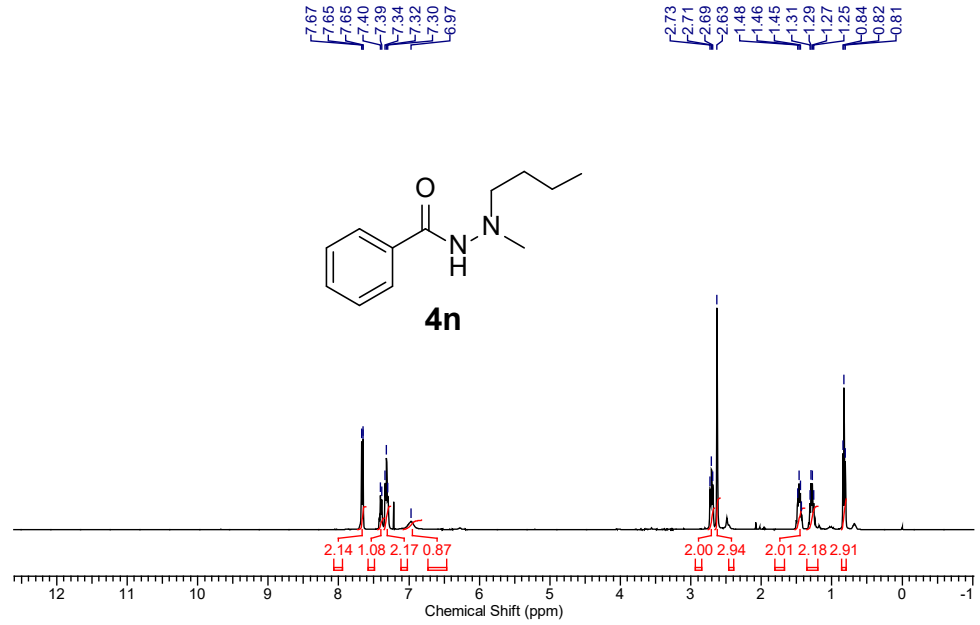


Figure S87. ^{1}H NMR of **4n** (400 MHz, $CDCl_3$)

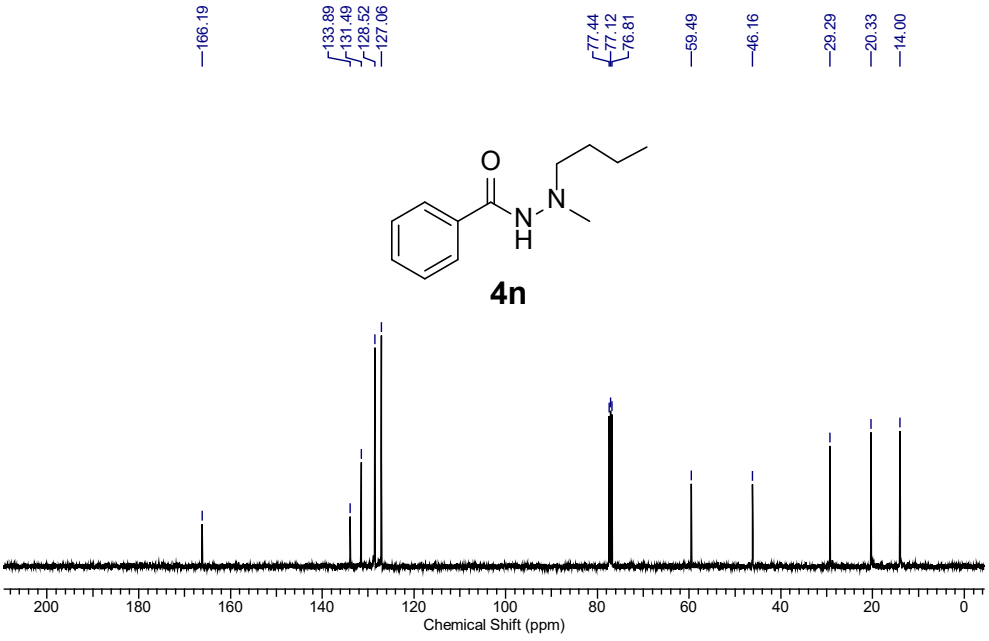


Figure S88. ^{13}C NMR of **4n** (100 MHz, $CDCl_3$)

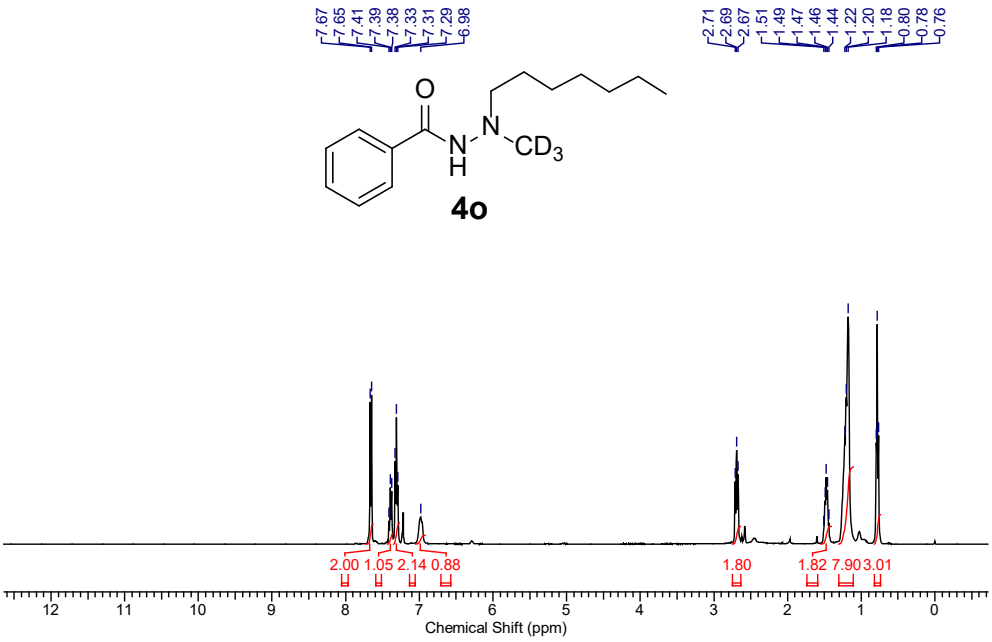


Figure S89. ^{1}H NMR of **4o** (400 MHz, $CDCl_3$)

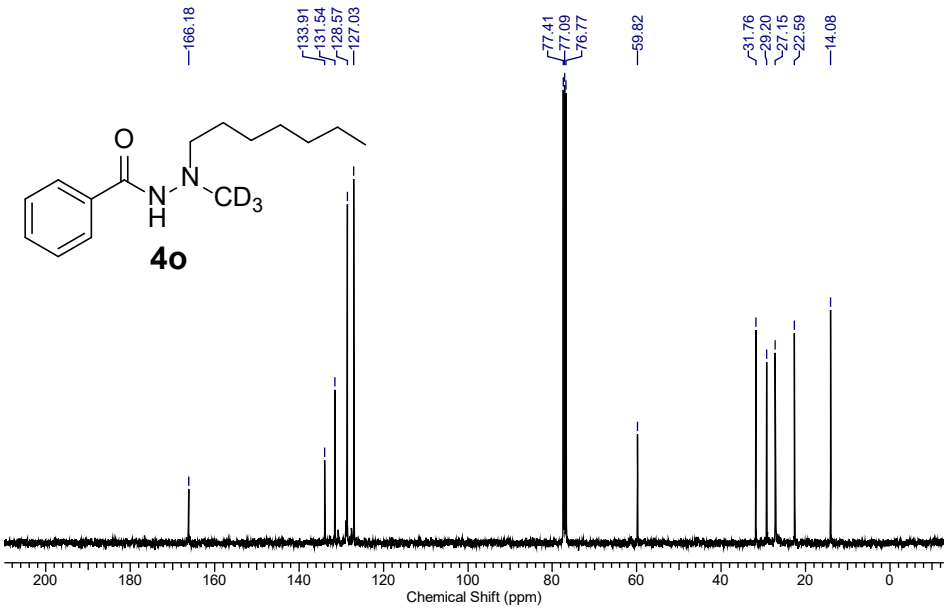


Figure S90. ^{13}C NMR of **4o** (100 MHz, $CDCl_3$)

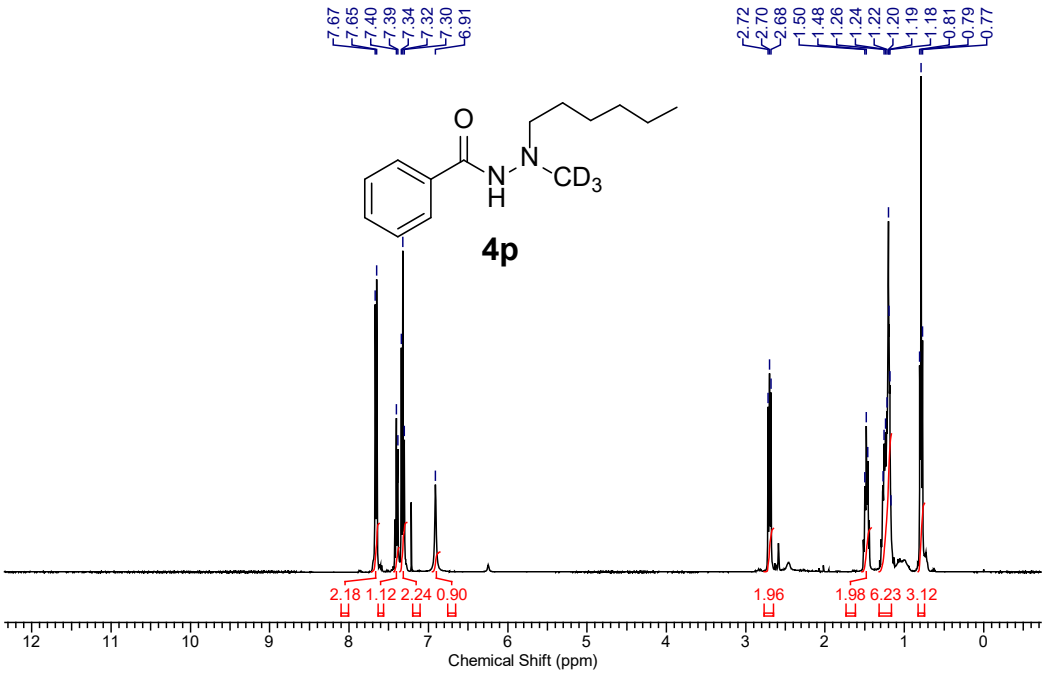


Figure S91. ^{1}H NMR of **4p** (400 MHz, $CDCl_3$)

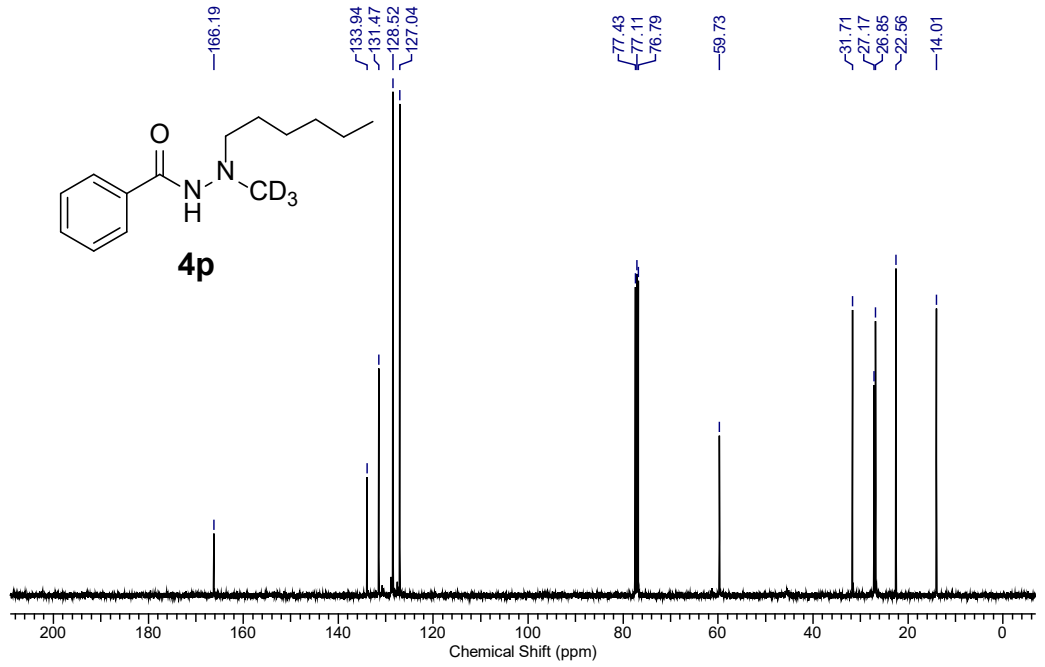


Figure S92. ^{13}C NMR of **4p** (100 MHz, $CDCl_3$)

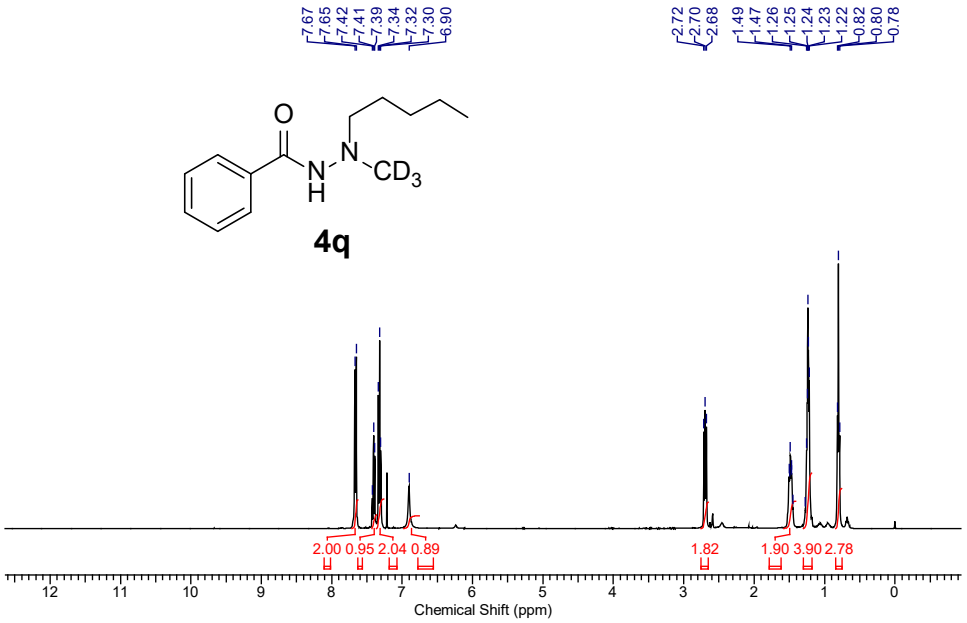


Figure S93. ^{1}H NMR of **4q** (400 MHz, $CDCl_3$)

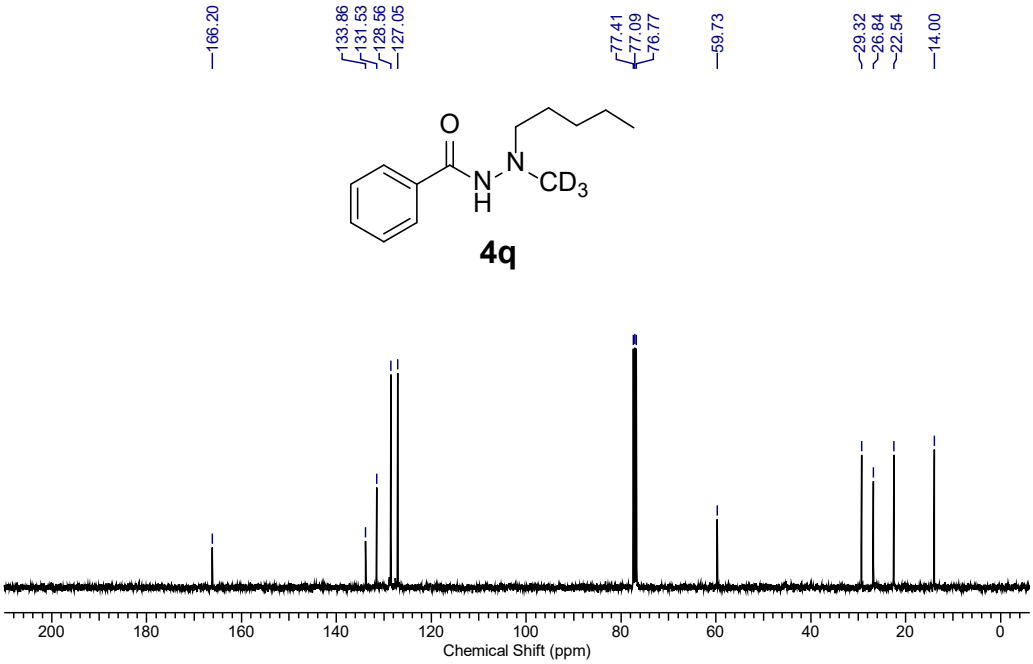


Figure S94. ^{13}C NMR of **4q** (100 MHz, $CDCl_3$)

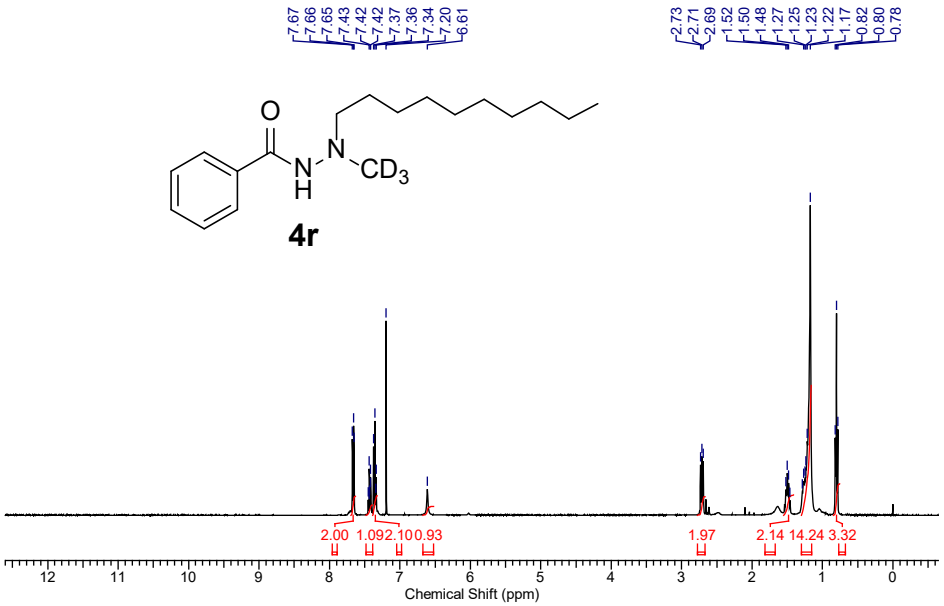


Figure S95. ^{1}H NMR of **4r** (400 MHz, $CDCl_3$)

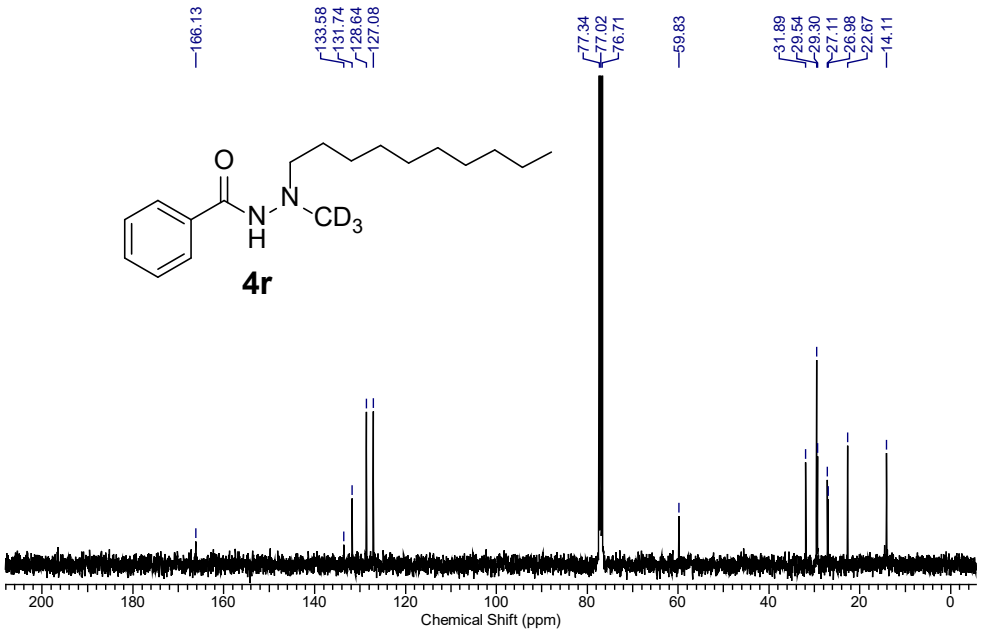


Figure S96. ^{13}C NMR of **4r** (100 MHz, $CDCl_3$)

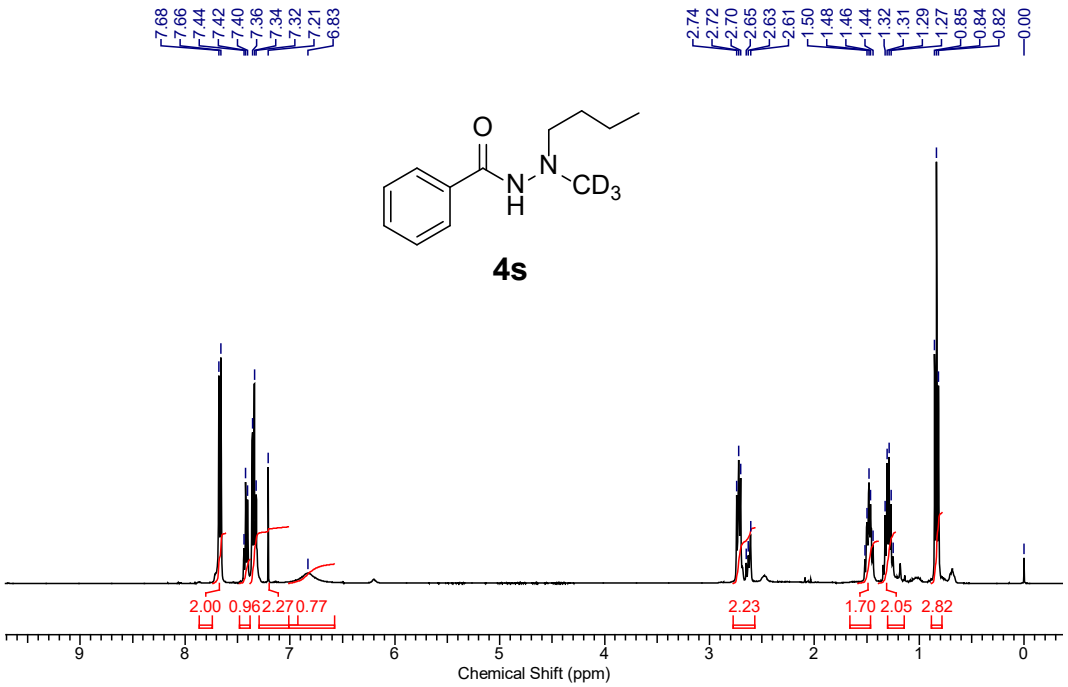


Figure S97. ^{1}H NMR of **4s** (400 MHz, $CDCl_3$)

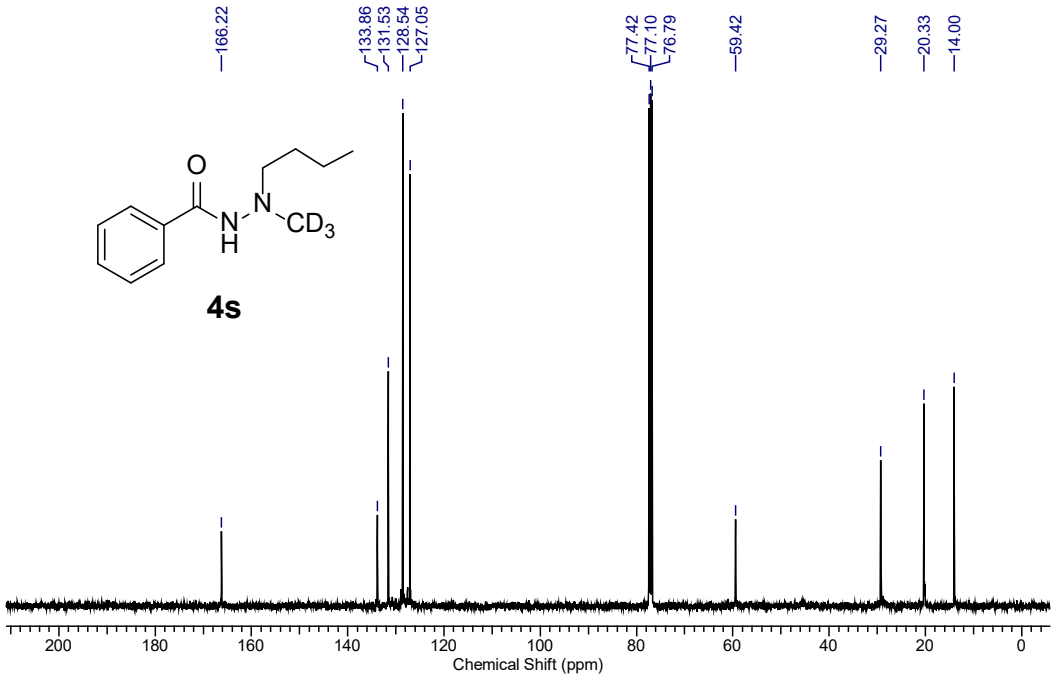


Figure S98. ^{13}C NMR of **4s** (100 MHz, $CDCl_3$)

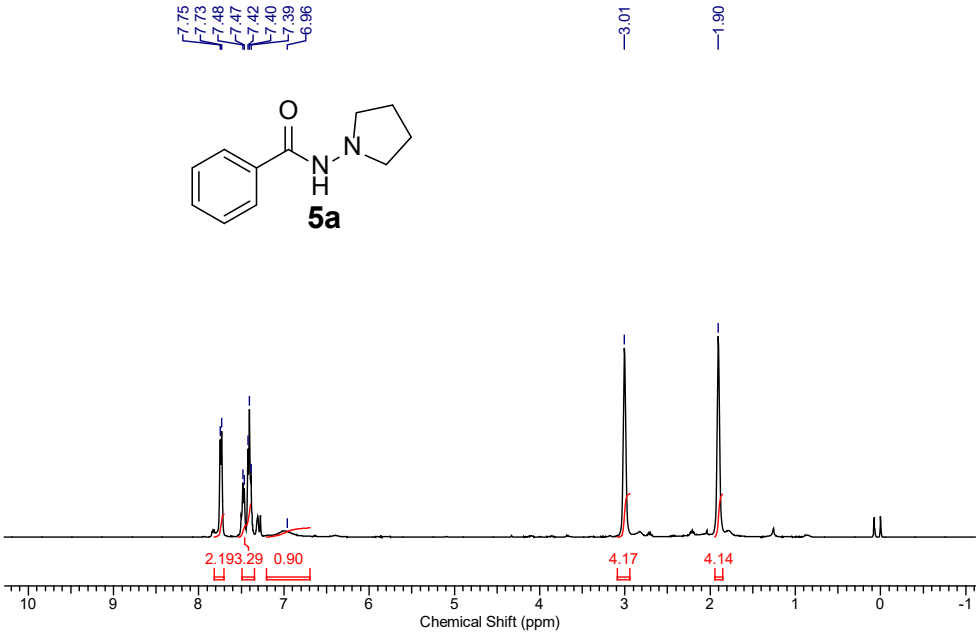


Figure S99. ^{1}H NMR of **5a** (400 MHz, $CDCl_3$)

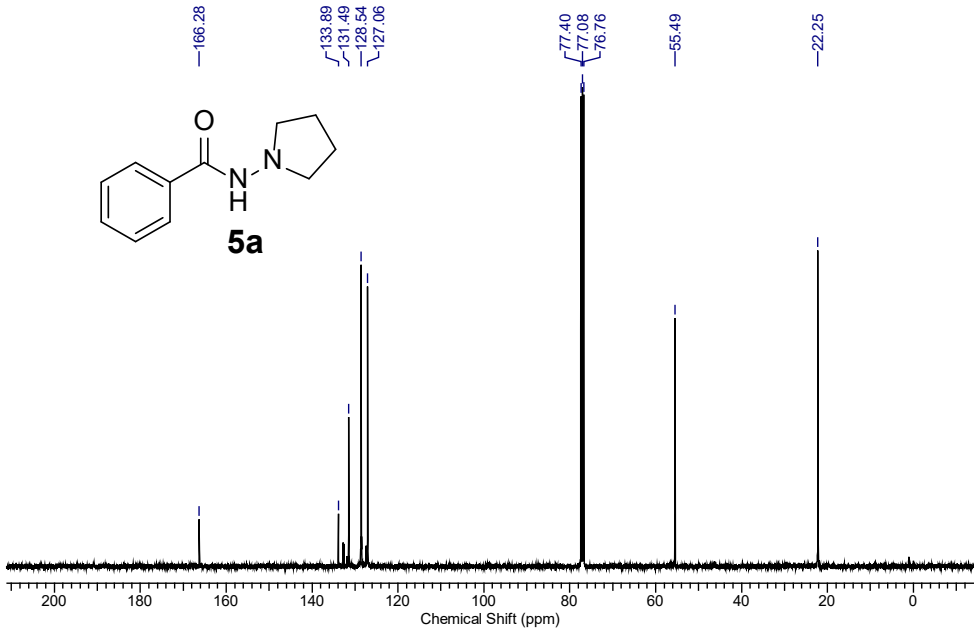


Figure S100. ^{13}C NMR of **5a** (100 MHz, $CDCl_3$)

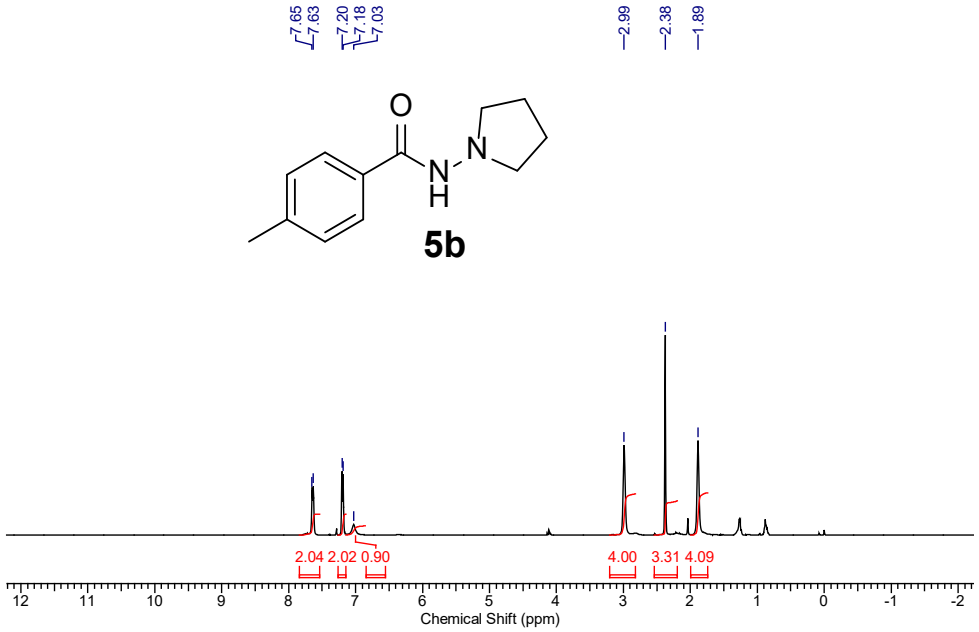


Figure S101. ^{1}H NMR of **5b** (400 MHz, $CDCl_3$)

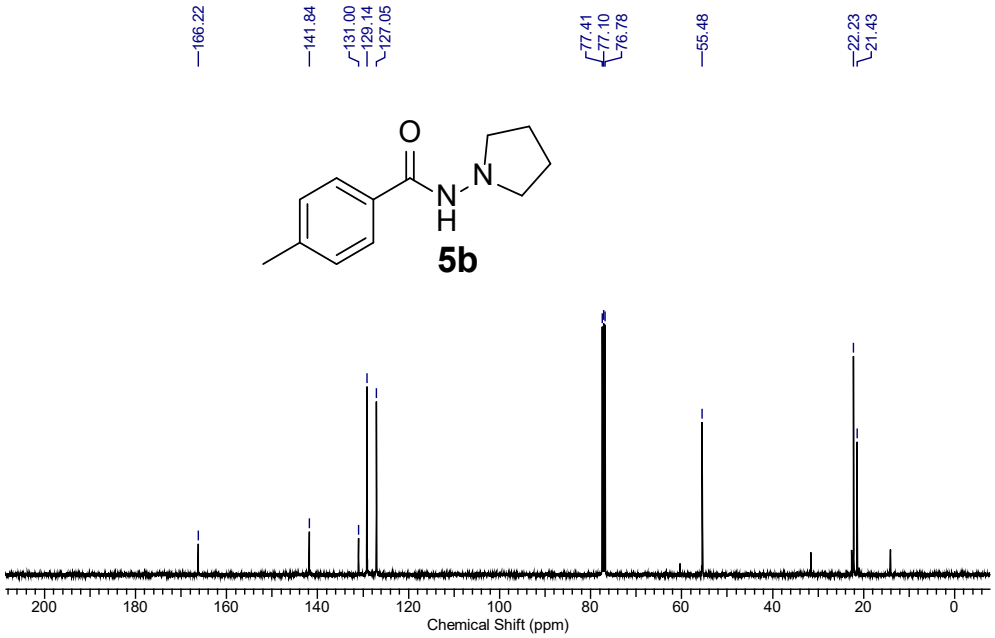


Figure S102. ^{13}C NMR of **5b** (100 MHz, $CDCl_3$)

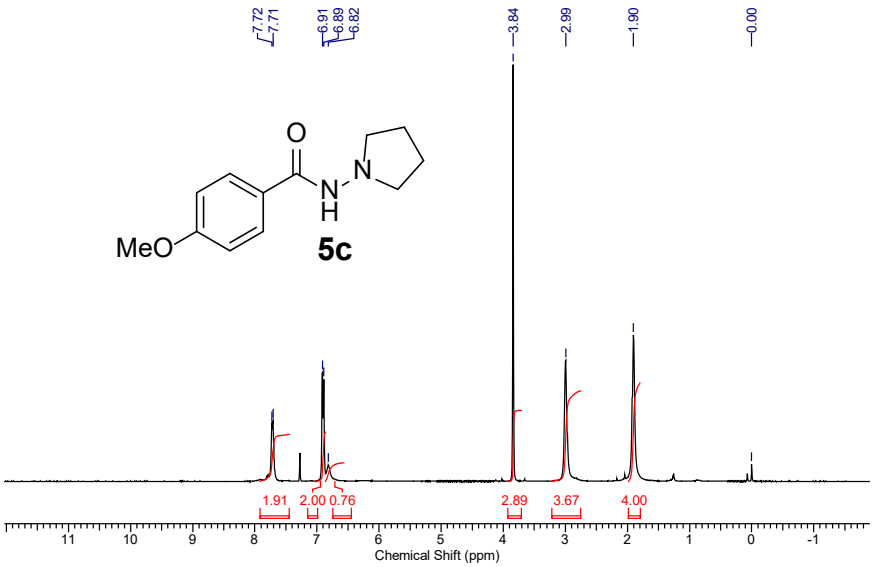


Figure S103. ^{1}H NMR of **5c** (400 MHz, $CDCl_3$)

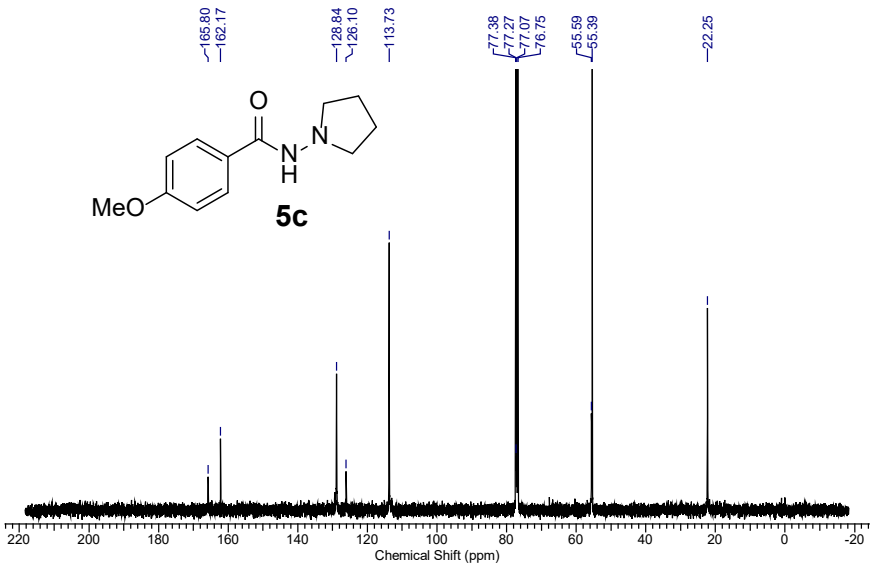


Figure S104. ^{13}C NMR of **5c** (100 MHz, $CDCl_3$)

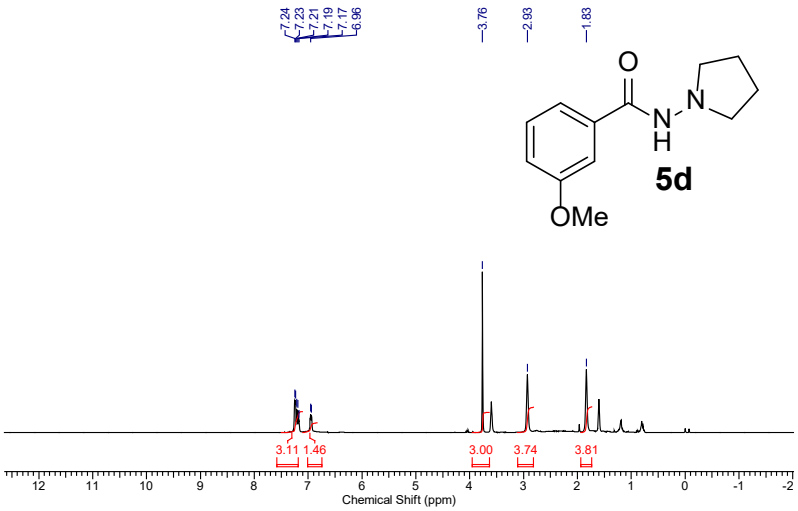


Figure S105. ^{1}H NMR of **5d** (400 MHz, $CDCl_3$)

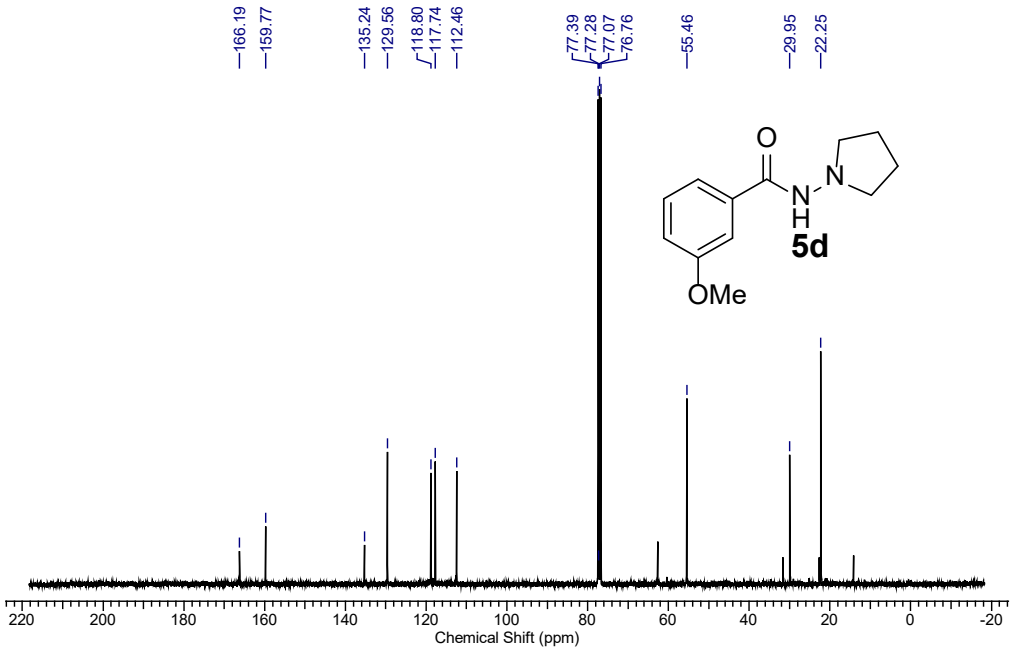


Figure S106. ^{13}C NMR of **5d** (100 MHz, $CDCl_3$)

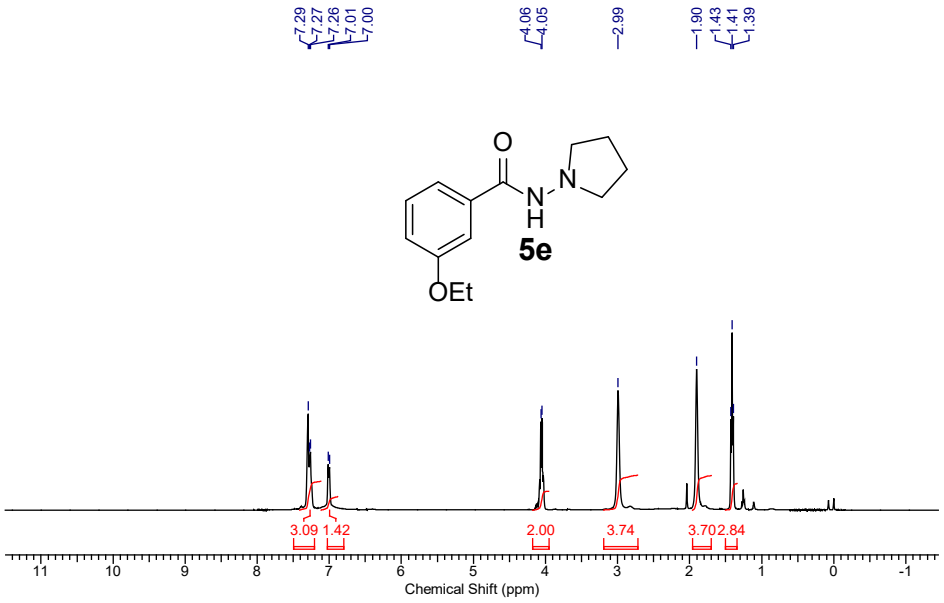


Figure S107. ^{1}H NMR of **5e** (400 MHz, $CDCl_3$)

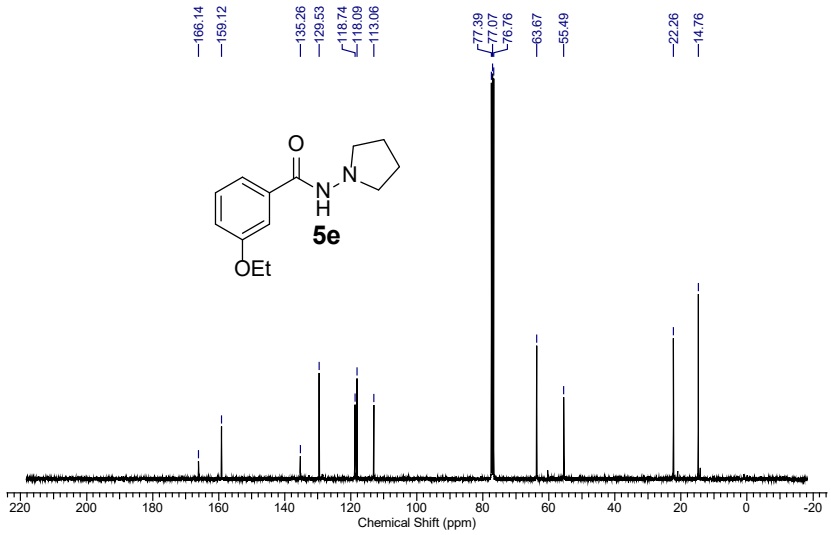


Figure S108. ^{13}C NMR of **5e** (100 MHz, $CDCl_3$)

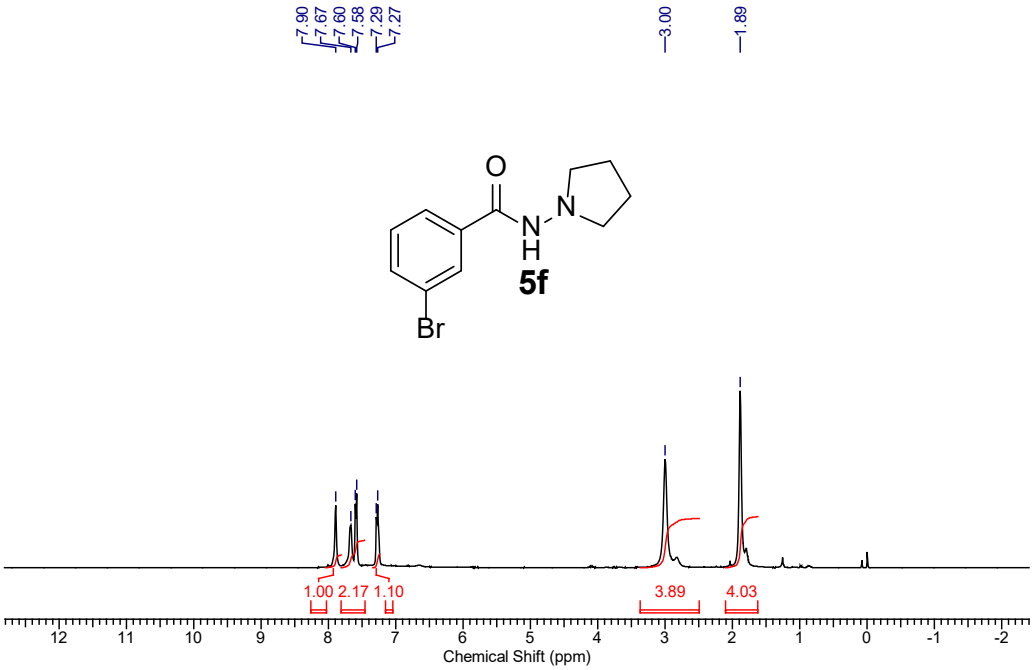


Figure S109. ^{1}H NMR of **5f** (400 MHz, $CDCl_3$)

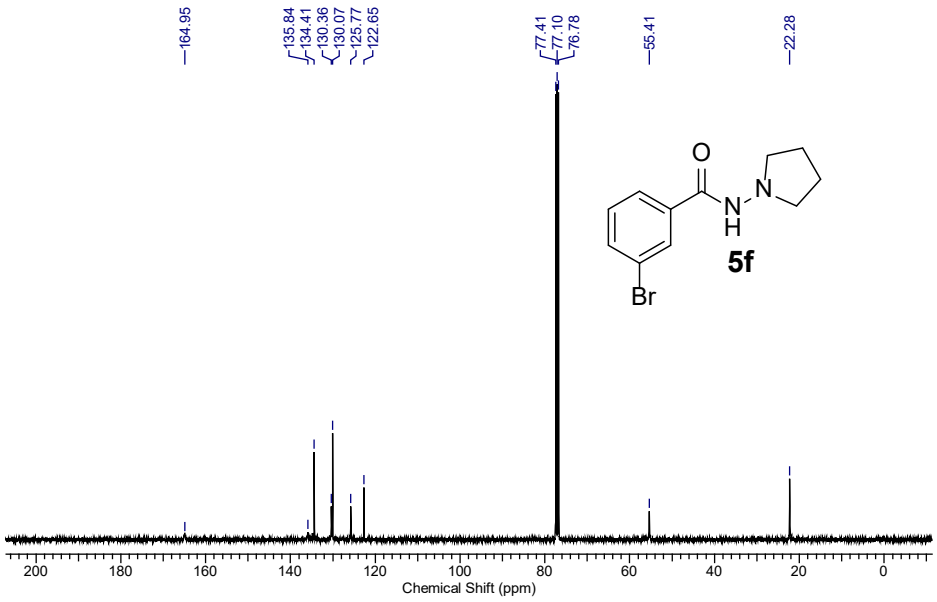


Figure S110. ^{13}C NMR of **5f** (100 MHz, $CDCl_3$)

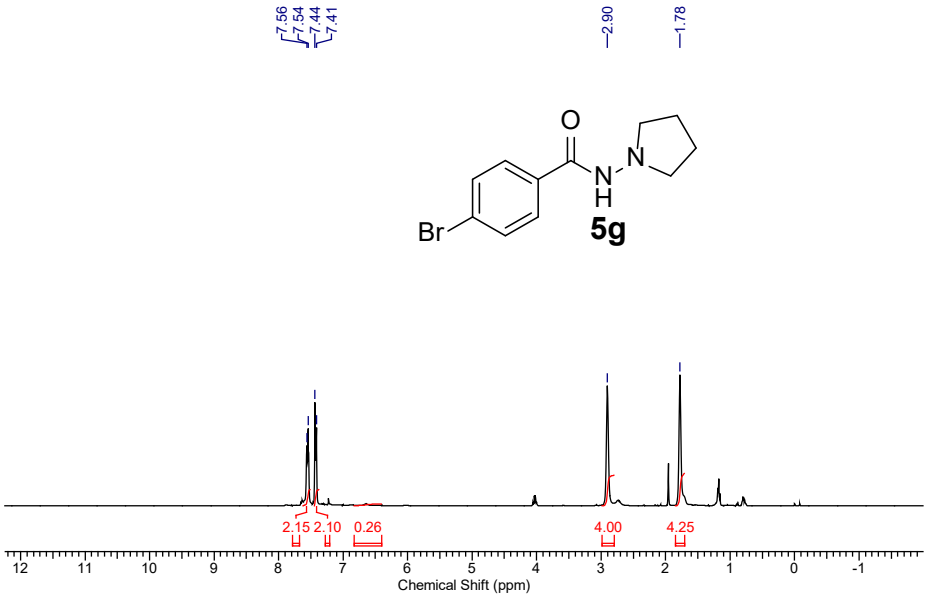


Figure S111. ^{1}H NMR of **5g** (400 MHz, $CDCl_3$)

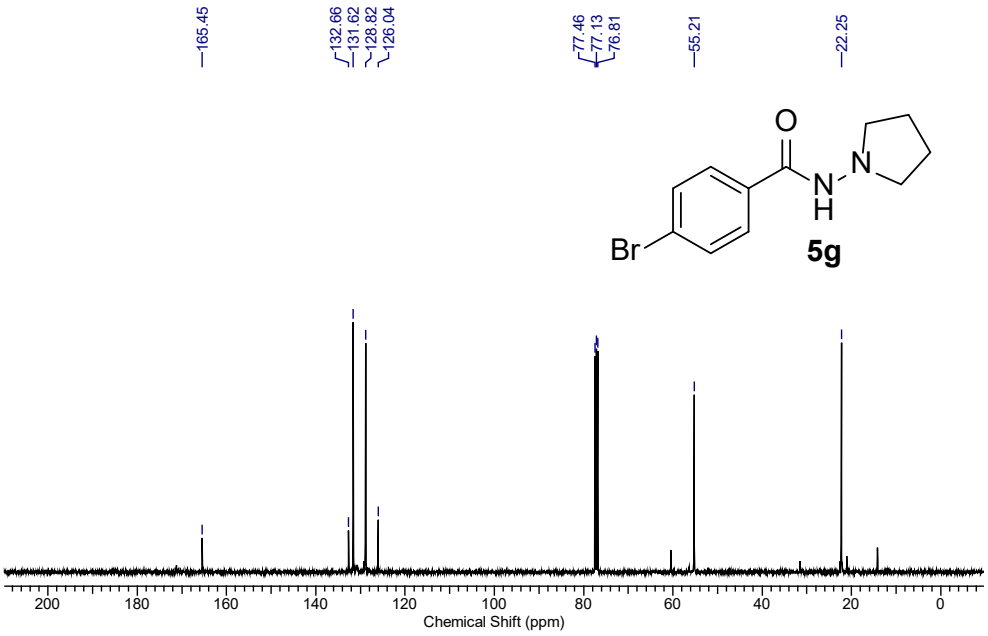


Figure S112. ^{13}C NMR of **5g** (100 MHz, $CDCl_3$)

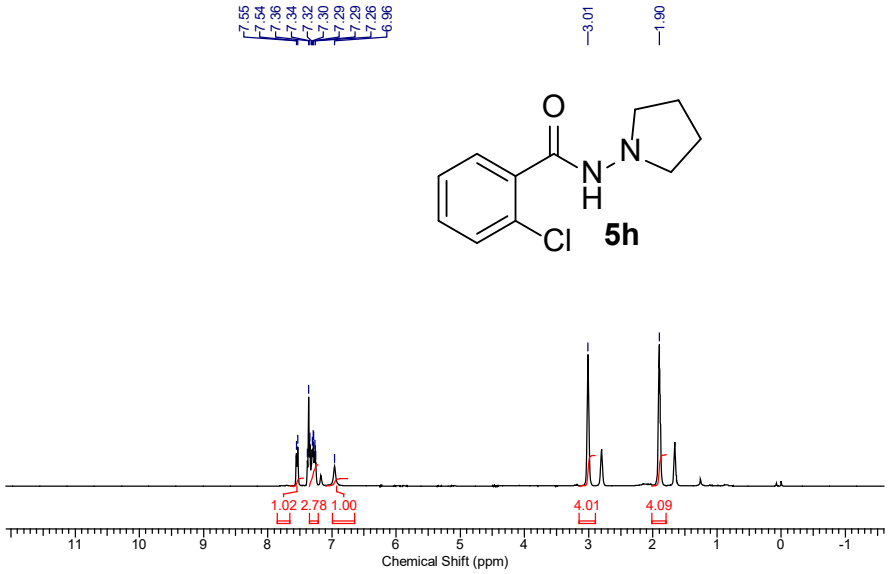


Figure S113. ^{1}H NMR of **5h** (400 MHz, $CDCl_3$)

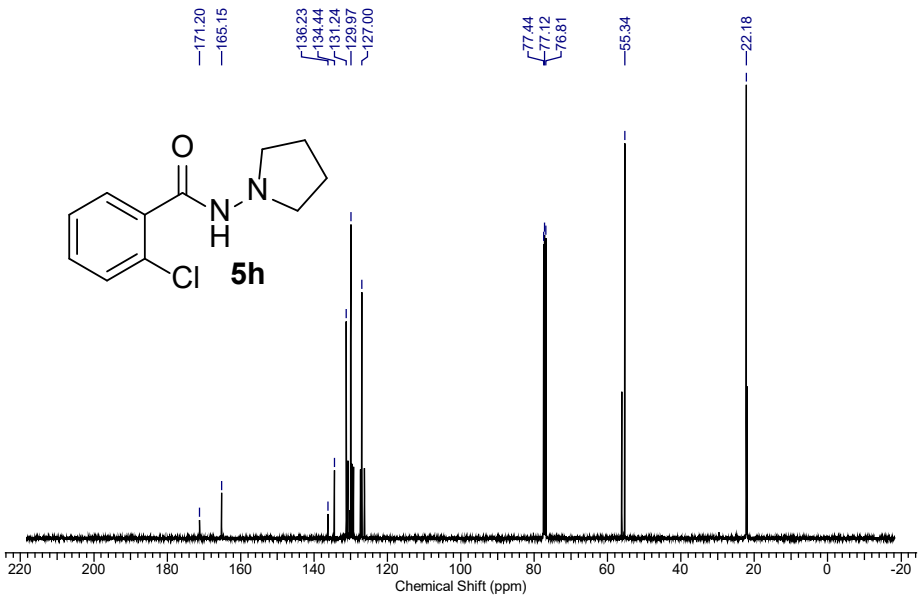


Figure S114. ^{13}C NMR of **5h** (100 MHz, $CDCl_3$)

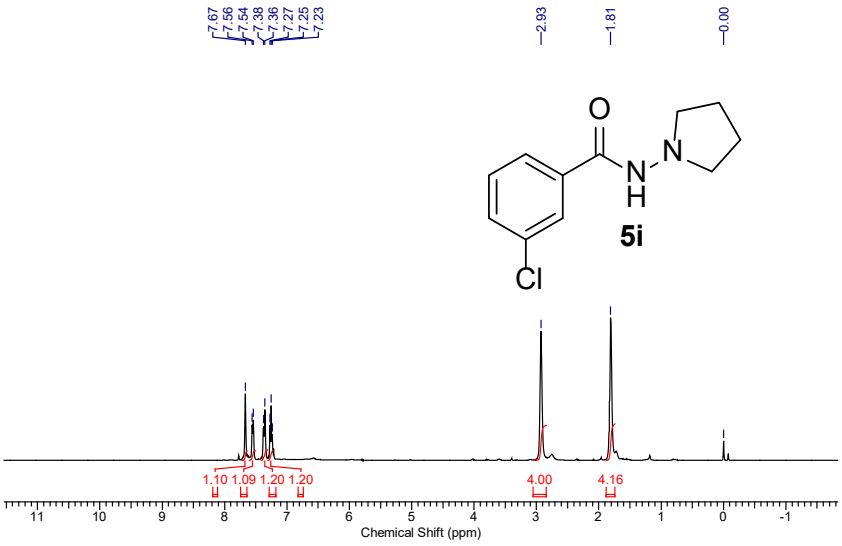


Figure S115. ^{1}H NMR of **5i** (400 MHz, $CDCl_3$)

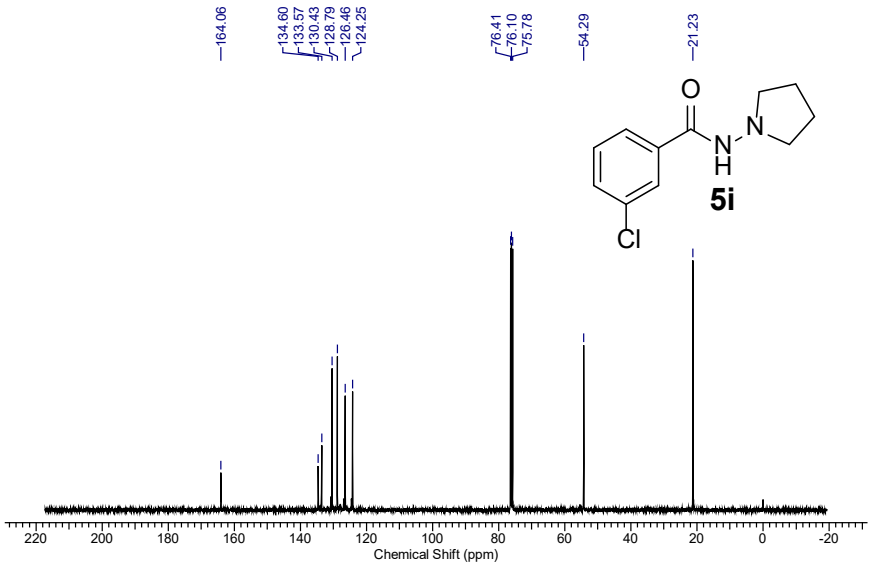


Figure S116. ^{13}C NMR of **5i** (100 MHz, $CDCl_3$)

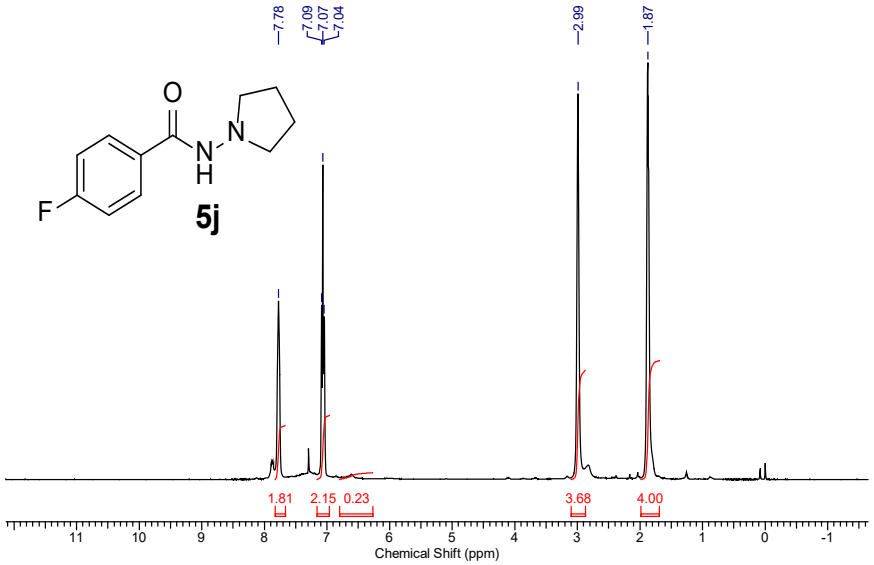


Figure S117. ^{1}H NMR of **5j** (400 MHz, $CDCl_3$)

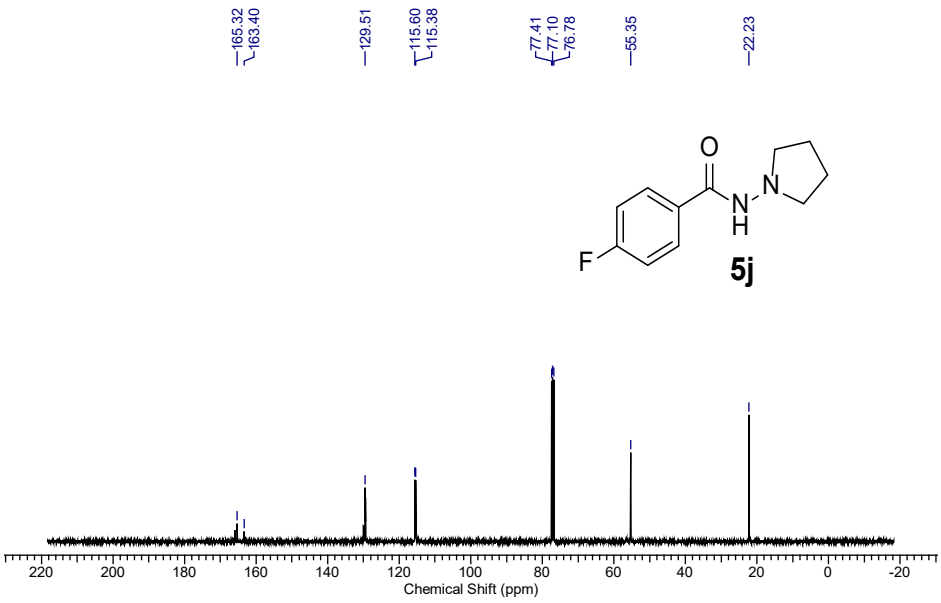


Figure S118. ^{13}C NMR of **5j** (100 MHz, $CDCl_3$)

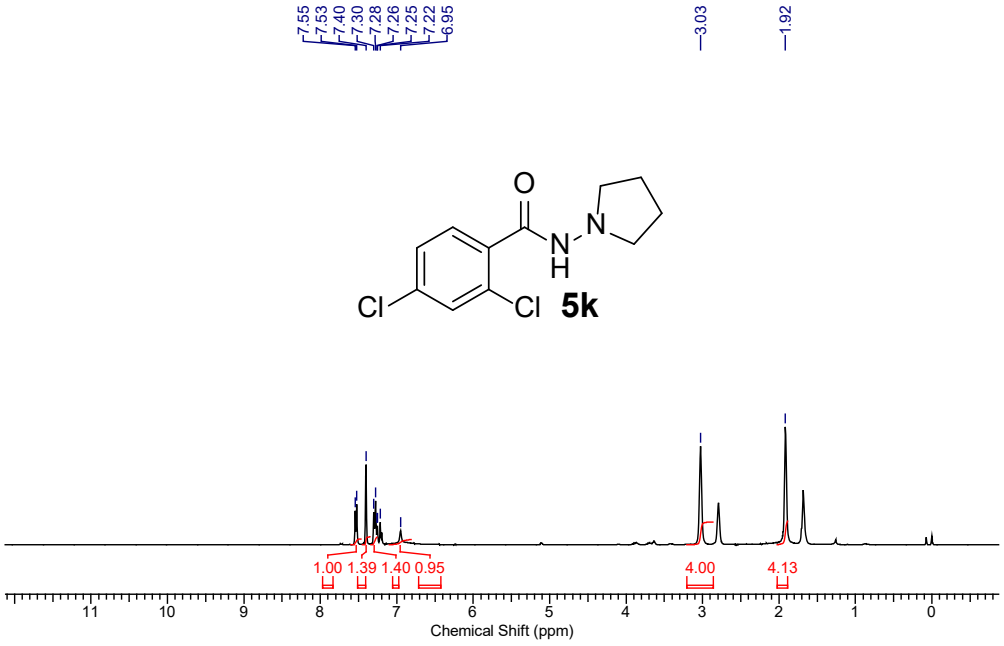


Figure S119. ^{1}H NMR of **5k** (400 MHz, $CDCl_3$)

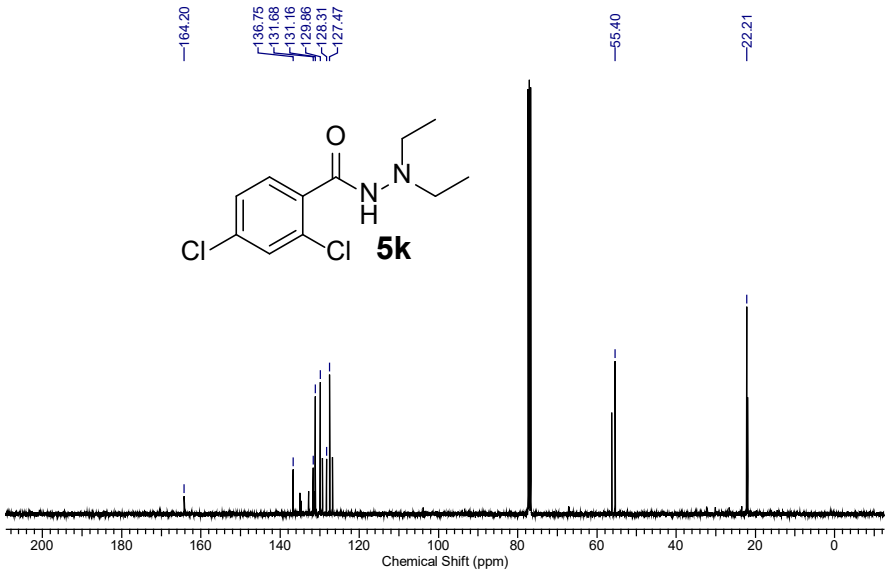


Figure S120. ^{13}C NMR of **5k** (100 MHz, $CDCl_3$)

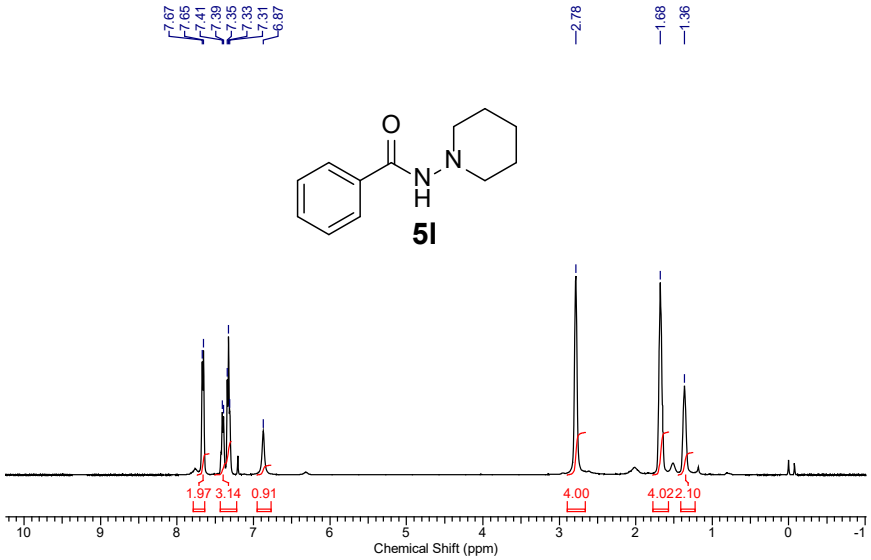


Figure S121. ^{1}H NMR of **5l** (400 MHz, $CDCl_3$)

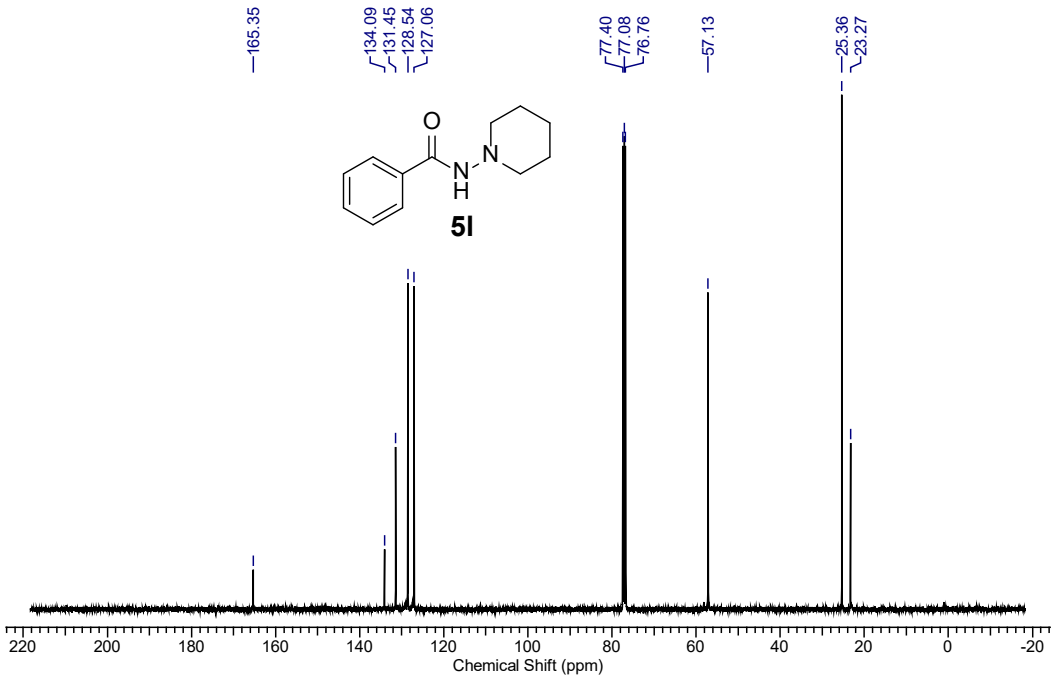


Figure S122. ^{13}C NMR of **5l** (100 MHz, $CDCl_3$)

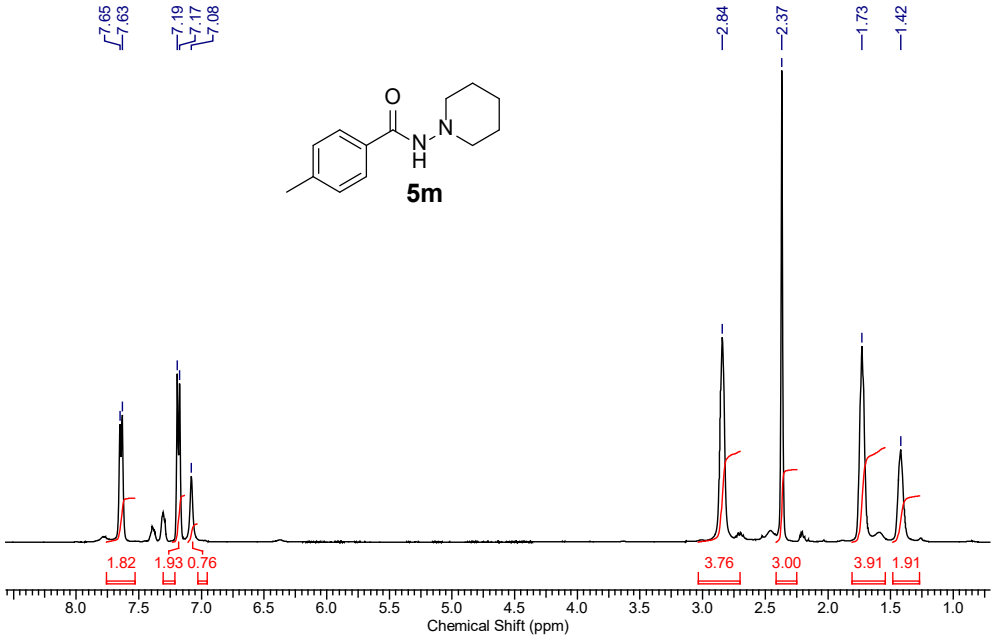


Figure S123. ^{1}H NMR of **5m** (400 MHz, $CDCl_3$)

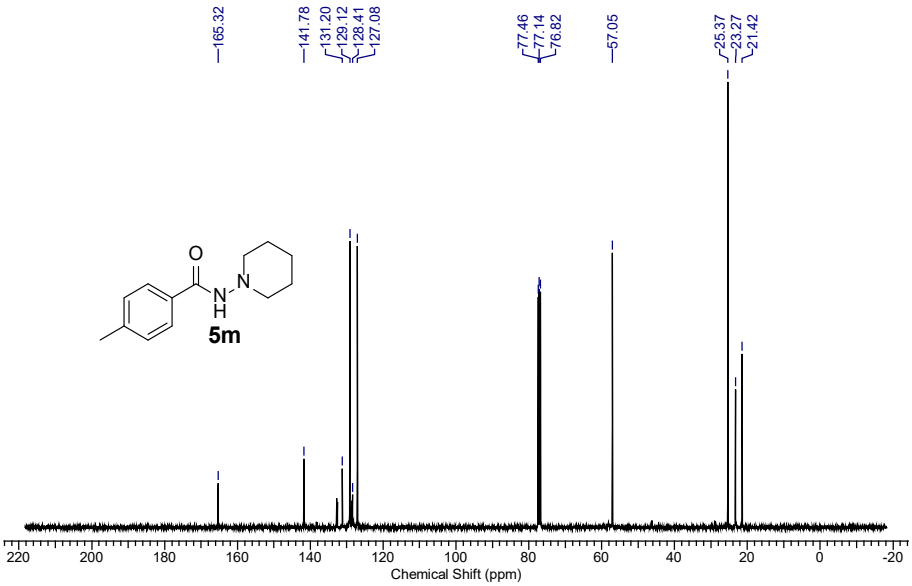


Figure S124. ^{13}C NMR of **5m** (100 MHz, $CDCl_3$)

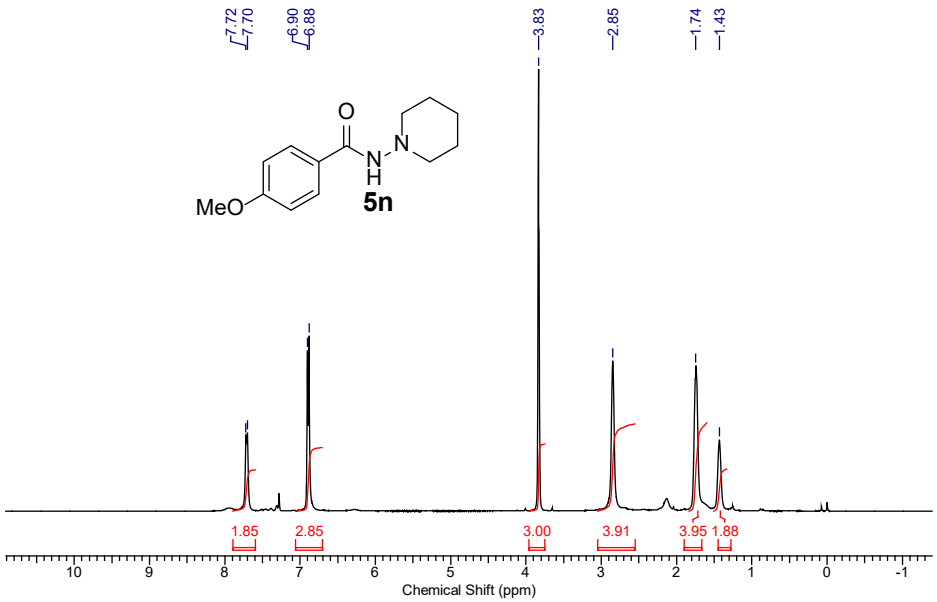


Figure S125. ^{1}H NMR of **5n** (400 MHz, $CDCl_3$)

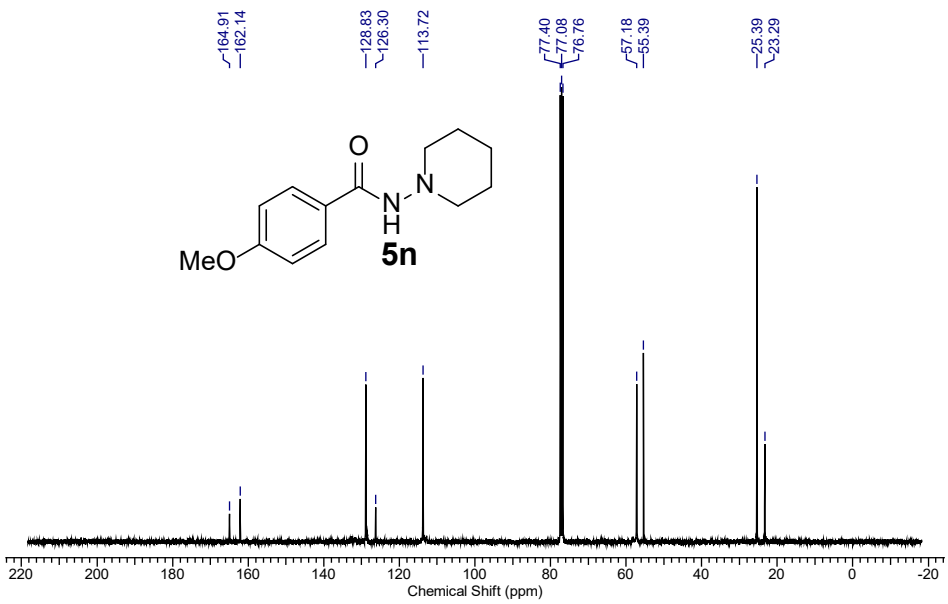


Figure S126. ^{13}C NMR of **5n** (100 MHz, $CDCl_3$)

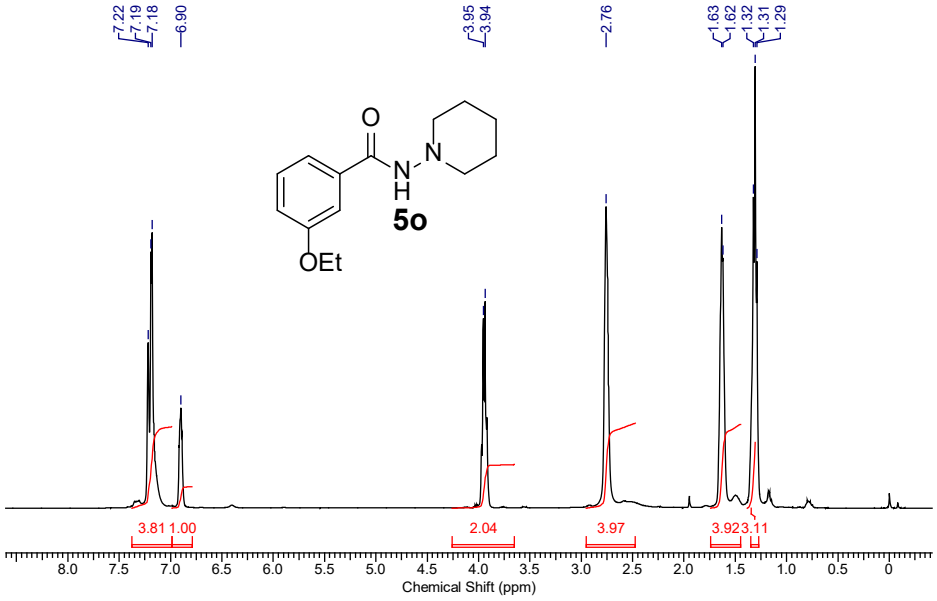


Figure S127. ^{1}H NMR of **5o** (400 MHz, $CDCl_3$)

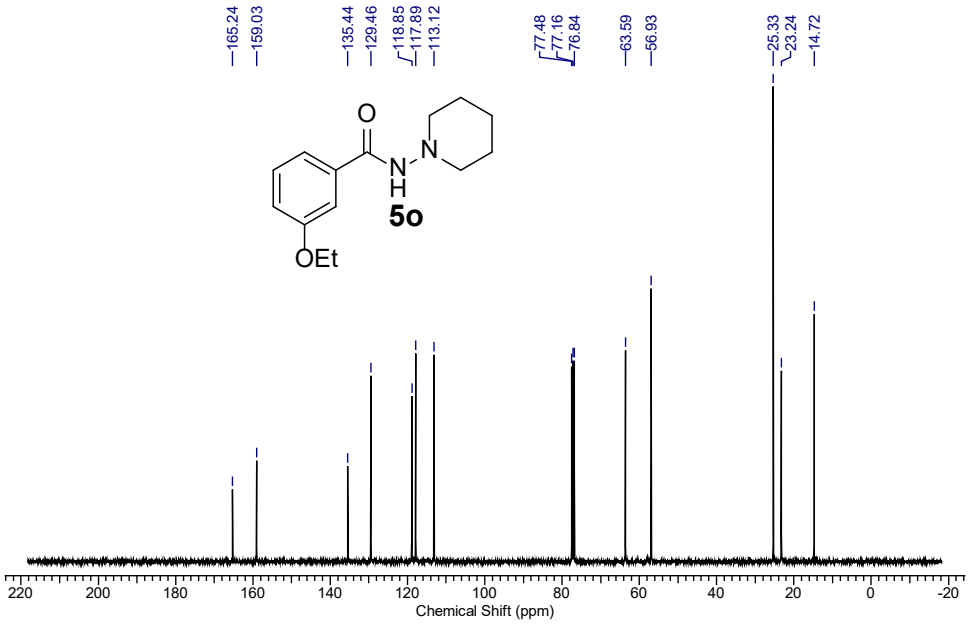


Figure S128. ^{13}C NMR of **5o** (100 MHz, $CDCl_3$)

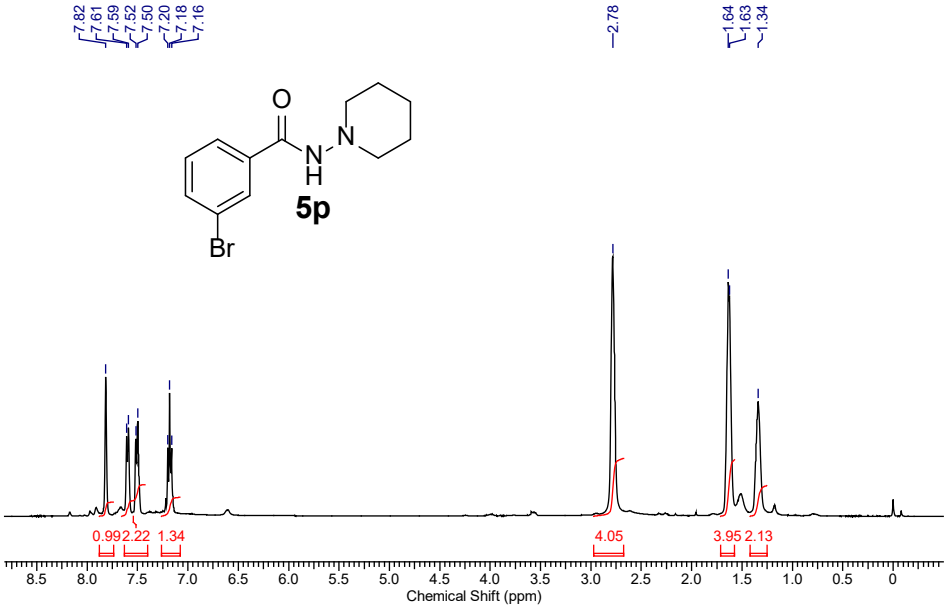


Figure S129. ^{1}H NMR of **5p** (400 MHz, $CDCl_3$)

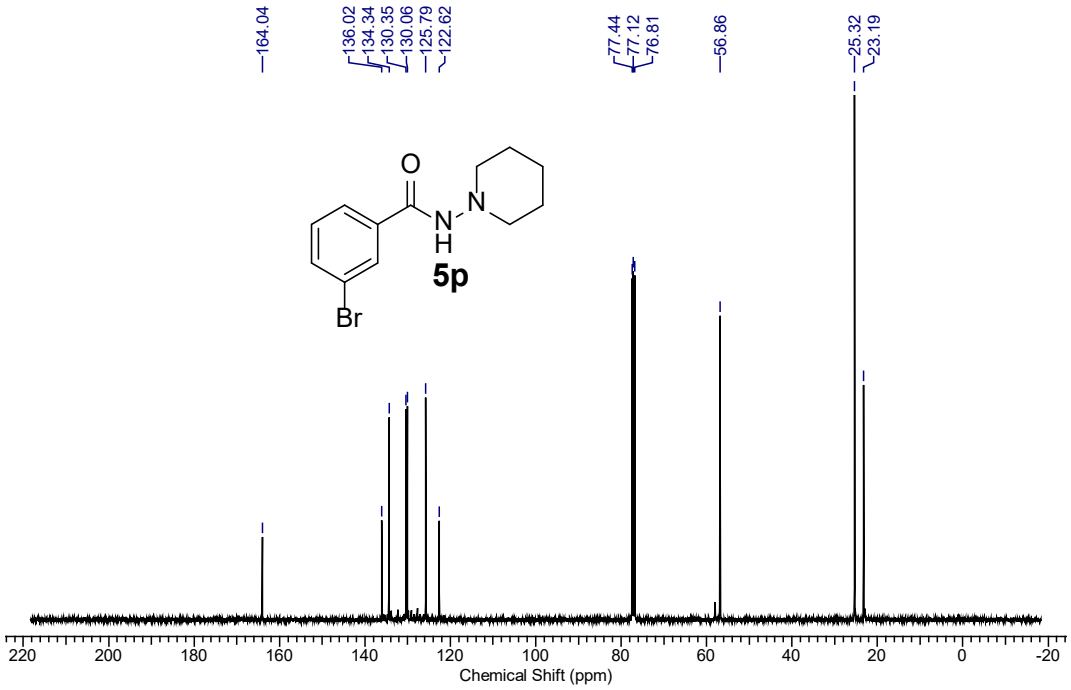


Figure S130. ^{13}C NMR of **5p** (100 MHz, $CDCl_3$

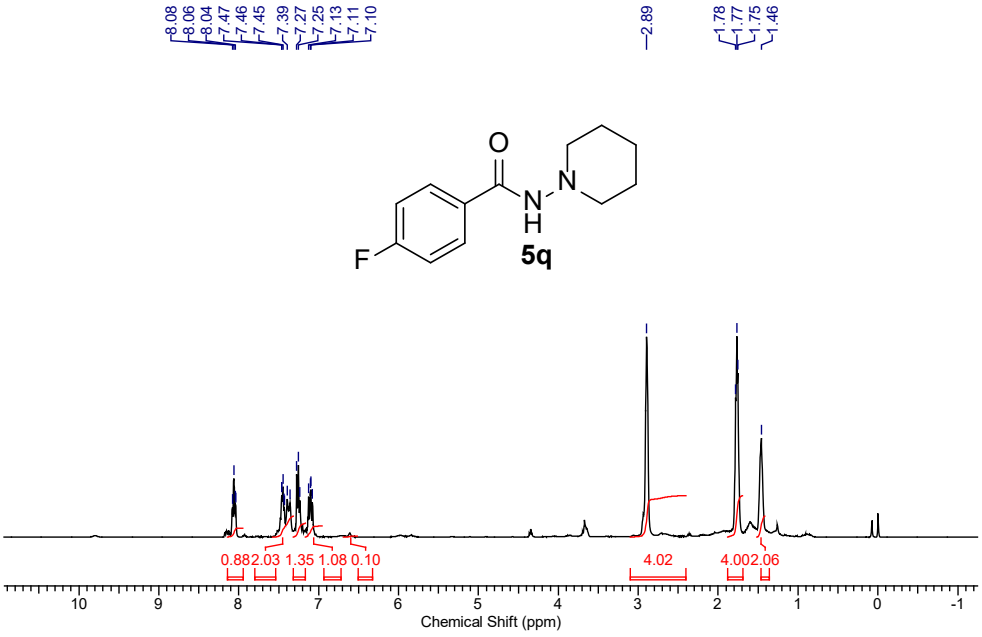


Figure S131. ^{1}H NMR of **5q** (400 MHz, $CDCl_3$)

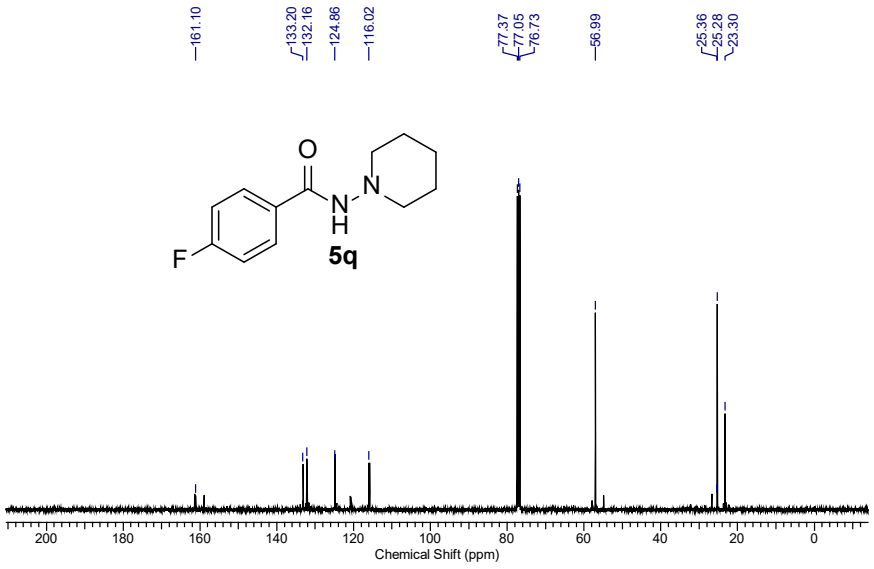


Figure S132. ^{13}C NMR of **5q** (100 MHz, $CDCl_3$)

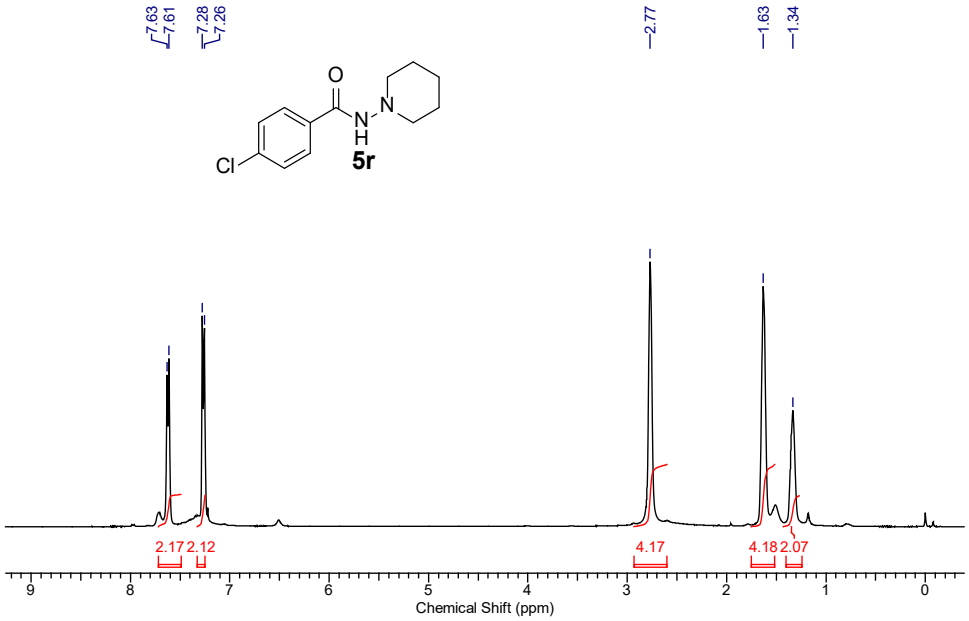


Figure S133. ^{1}H NMR of **5r** (400 MHz, $CDCl_3$)

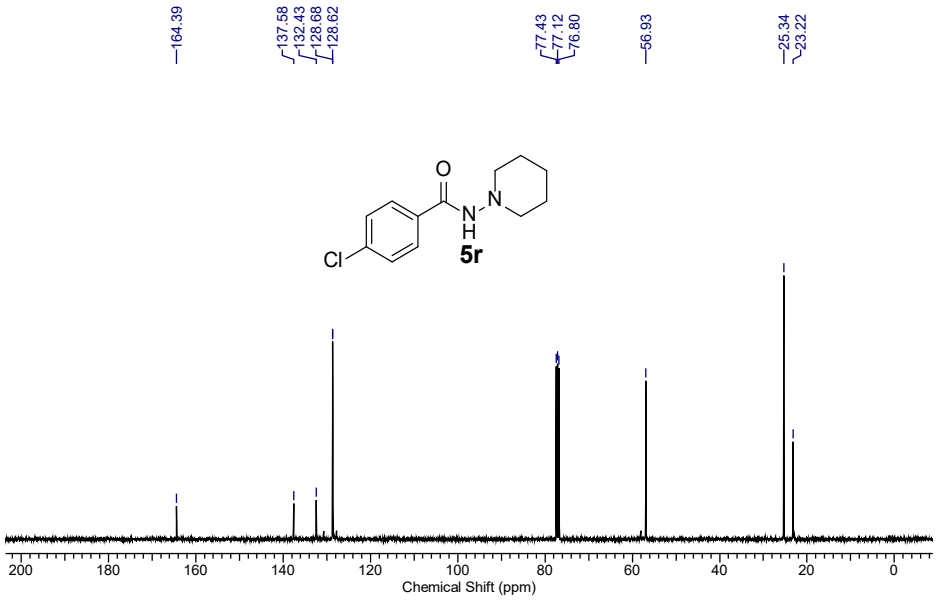


Figure S134. ^{13}C NMR of **5r** (100 MHz, $CDCl_3$)

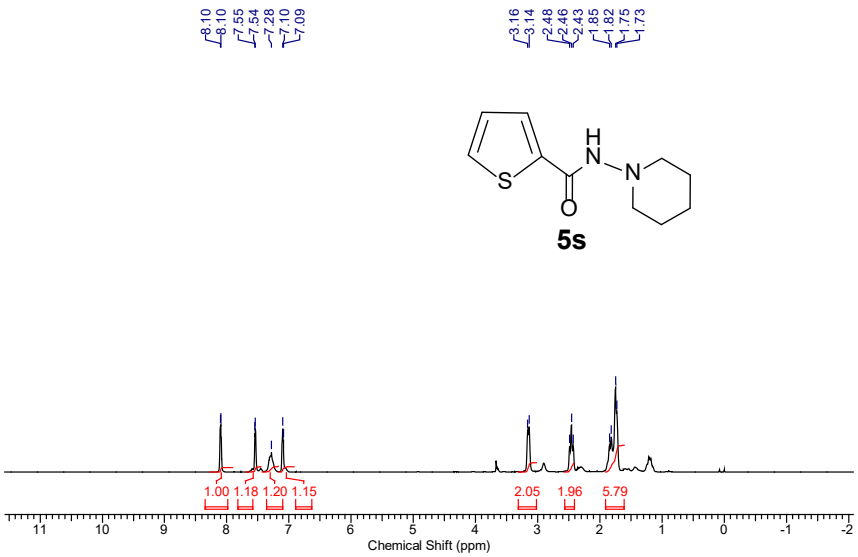


Figure S135. ^{1}H NMR of **5s** (400 MHz, $CDCl_3$)

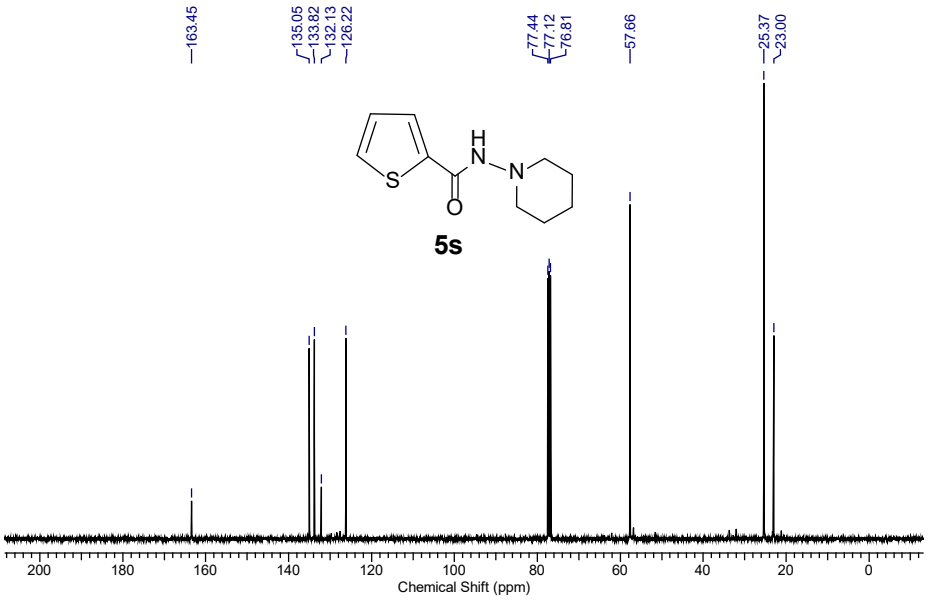


Figure S136. ^{13}C NMR of **5s** (100 MHz, $CDCl_3$)

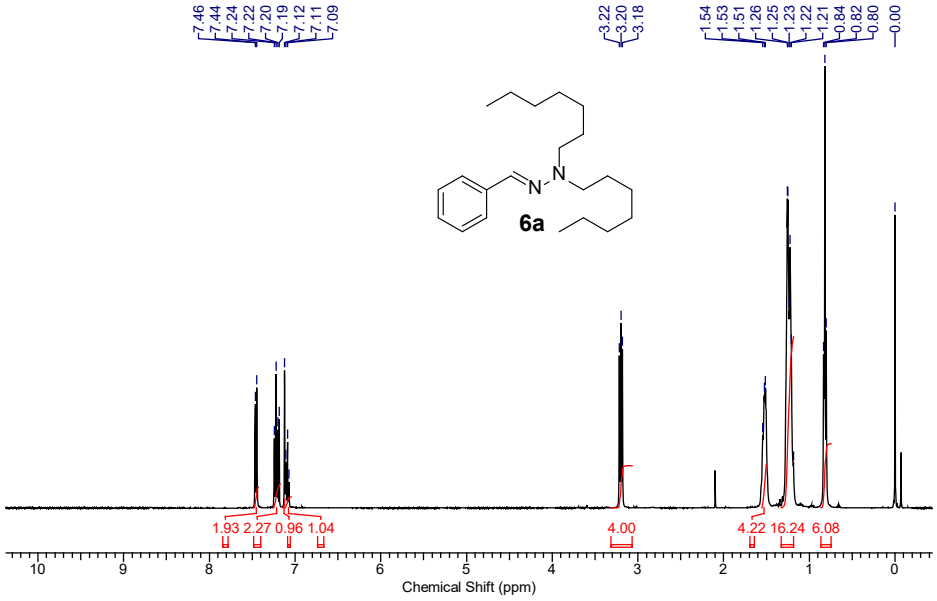


Figure S137. ^{1}H NMR of **6a** (400 MHz, $CDCl_3$)

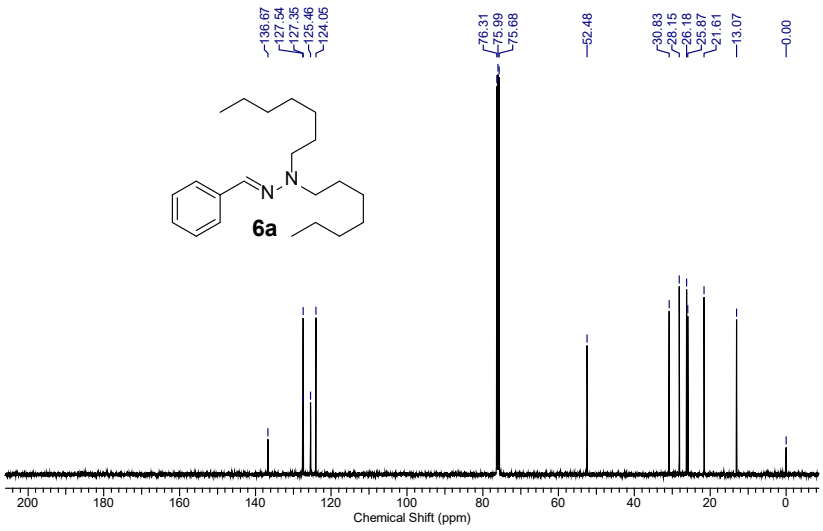


Figure S138. ^{13}C NMR of **6a** (100 MHz, $CDCl_3$)

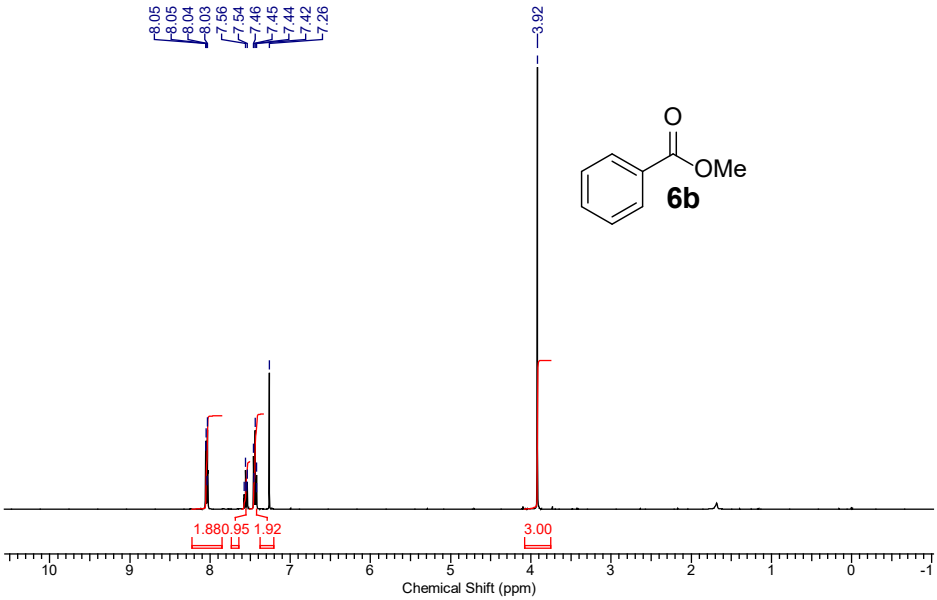


Figure S139. ^{1}H NMR of **6b** (100 MHz, $CDCl_3$)

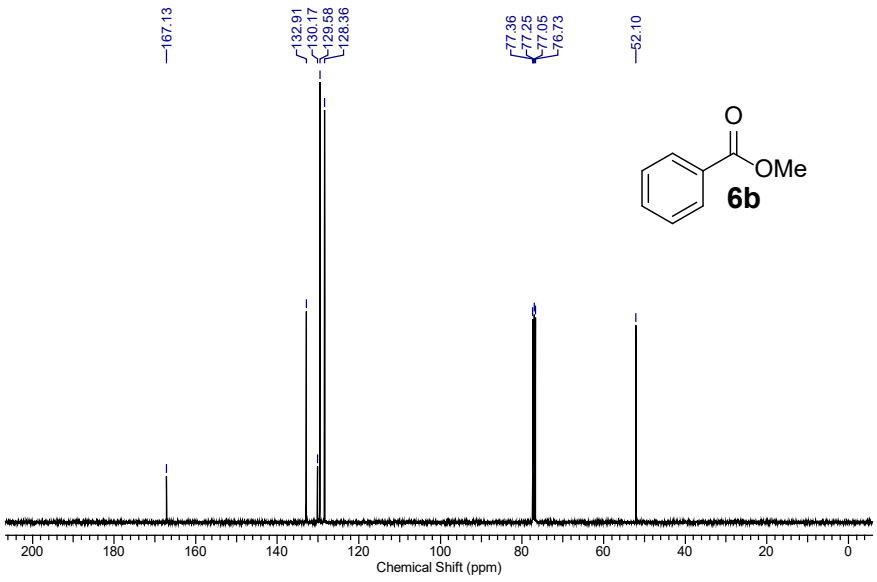


Figure S140. ^{13}C NMR of **6b** (100 MHz, $CDCl_3$)

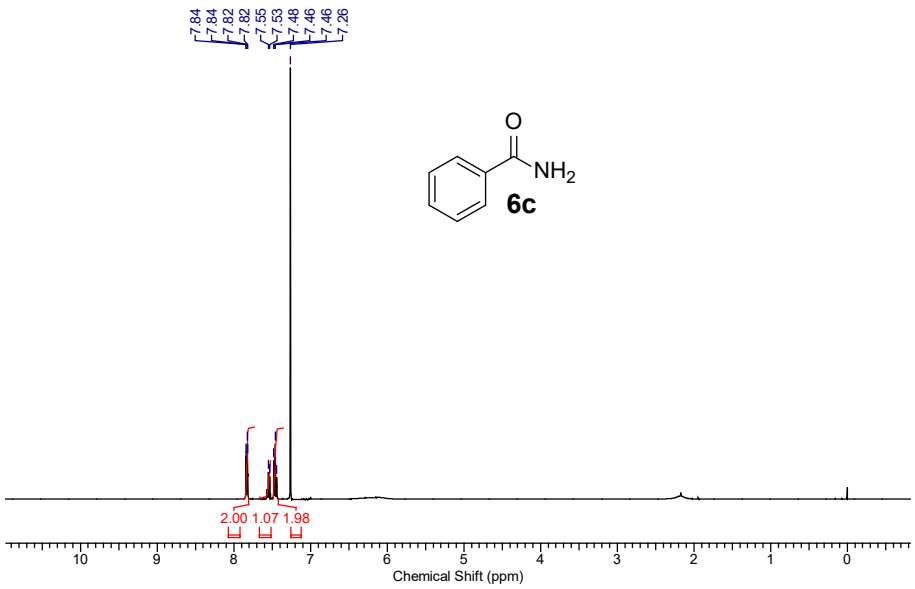


Figure S141. ^{1}H NMR of **6c** (100 MHz, $CDCl_3$)

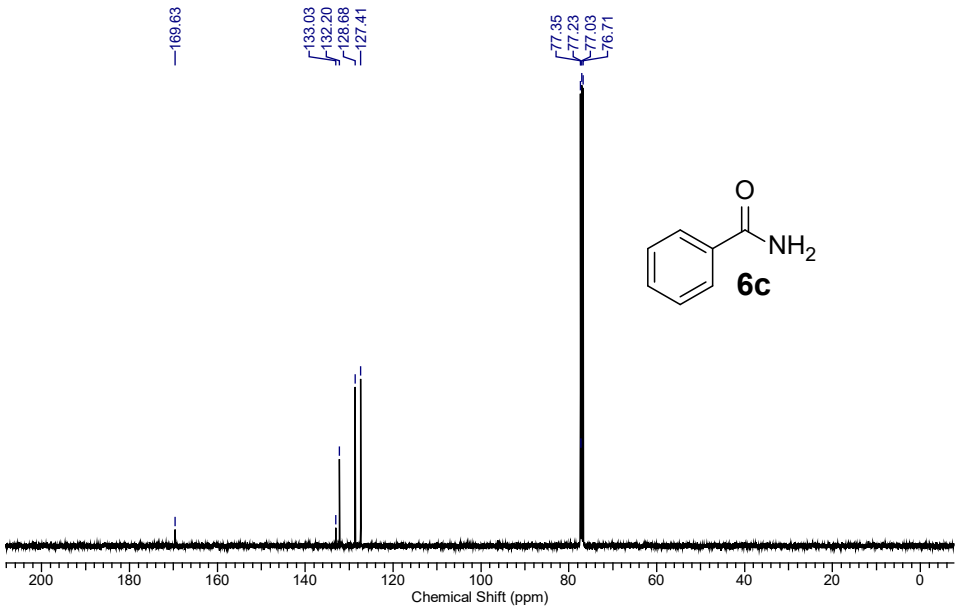


Figure S142. ^{13}C NMR of **6c** (100 MHz, $CDCl_3$)

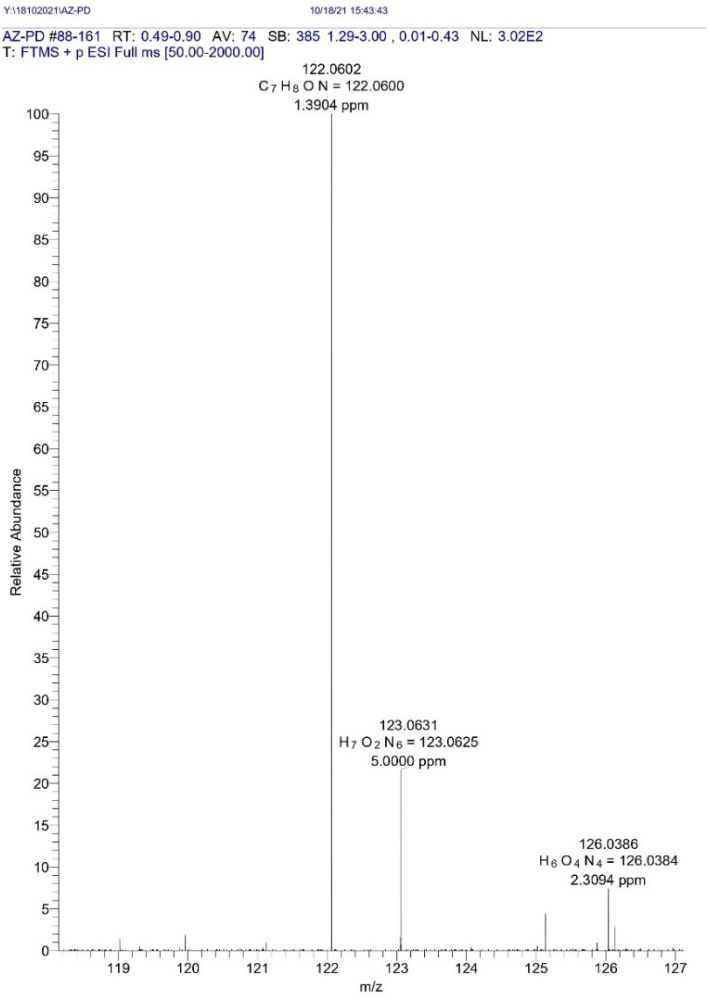


Figure S143. **HRMS data of benzamide 6c**

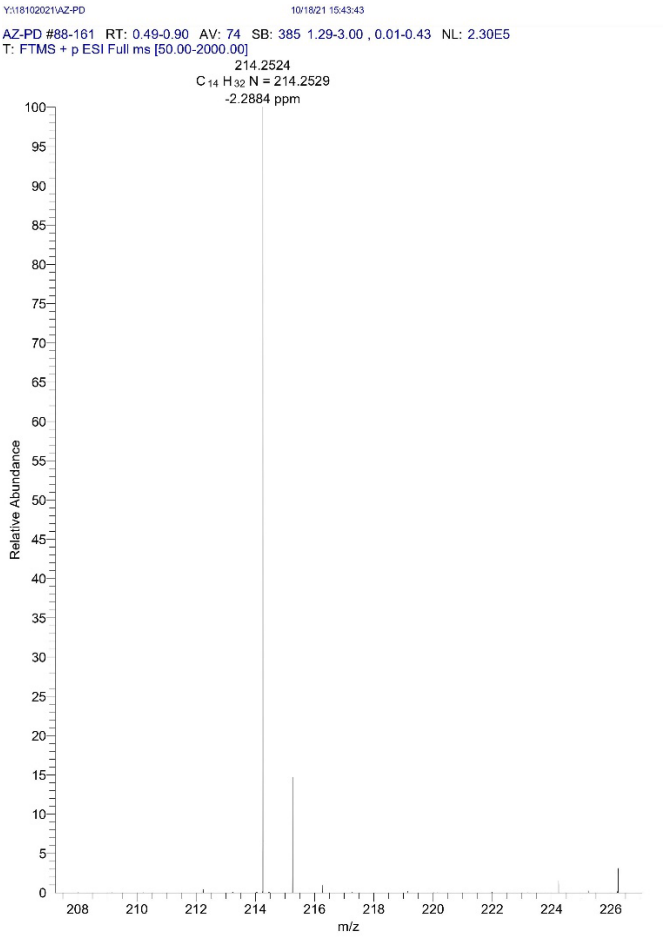


Figure S144. HRMS data of diheptylamine 6c'