Supporting Information

Direct Catalytic Symmetrical, Unsymmetrical N,N-Dialkylation and Cyclization of Acylhydrazides Using Alcohols

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Index

General Experimental: Unless otherwise noted, all catalytic reactions were performed under inert atmosphere using standard Schlenk techniques. All stoichiometric reactions were performed in nitrogen atmosphere MBRAUN glove box. Ru-MACHO [Carbonylchlorohydrido{bis[2-(diphenylphosphinomethyl)ethyl]amino}ethyl]amino}ruthenium(II)] and KO^{t}Bu were purchased from Sigma-Aldrich and stored inside glove box. Acylhydrazides were prepared from previous reported procedure.[1] Chemicals were purchased from Sigma-Aldrich, Acros, Alfa-aesar, Himedia and TCI Chemicals and used without further purification. Dry solvents were prepared according to standard procedures. Infrared (IR) spectra were recorded in Thermo-Nicolet FT-IR spectrophotometers. High-resolution mass spectra (HRMS) were obtained on Bruker micrOTOF-Q II Spectrometer and are reported as m/z (relative intensity). Accurate masses are reported for the molecular ion $[M+Na]^+$, $[M+H]^+$, $[M]^+$. Nuclear magnetic resonance spectra (^{1}H NMR and ^{13}C NMR) were recorded at Bruker AV-400 (^{1}H at 400 MHz, ^{13}C at 100.6 MHz). ^{1}H NMR chemical shifts are referenced in parts per million (ppm) with respect to tetramethyl silane (TMS) (δ 0.00 ppm) and ^{13}C {^{1}H} NMR chemical shifts are referenced in parts per million (ppm) with respect to $CDCl_3$ (δ 77.160 ppm). Coupling constants are reported in Hertz (Hz). ^{1}H NMR spectroscopy abbreviations: s, Singlet; d, doublet; t, triplet; q, quartet; dd, doublet of doublets; dt, doublet of triplets; dq, doublet of quartets; td, triplet of doublets; qd, quartets of doublets; ddd, doublets of doublets of doublets; m, multiplet; br, broad. Assignment of spectra was done based on one-dimensional (dept-135) NMR techniques.

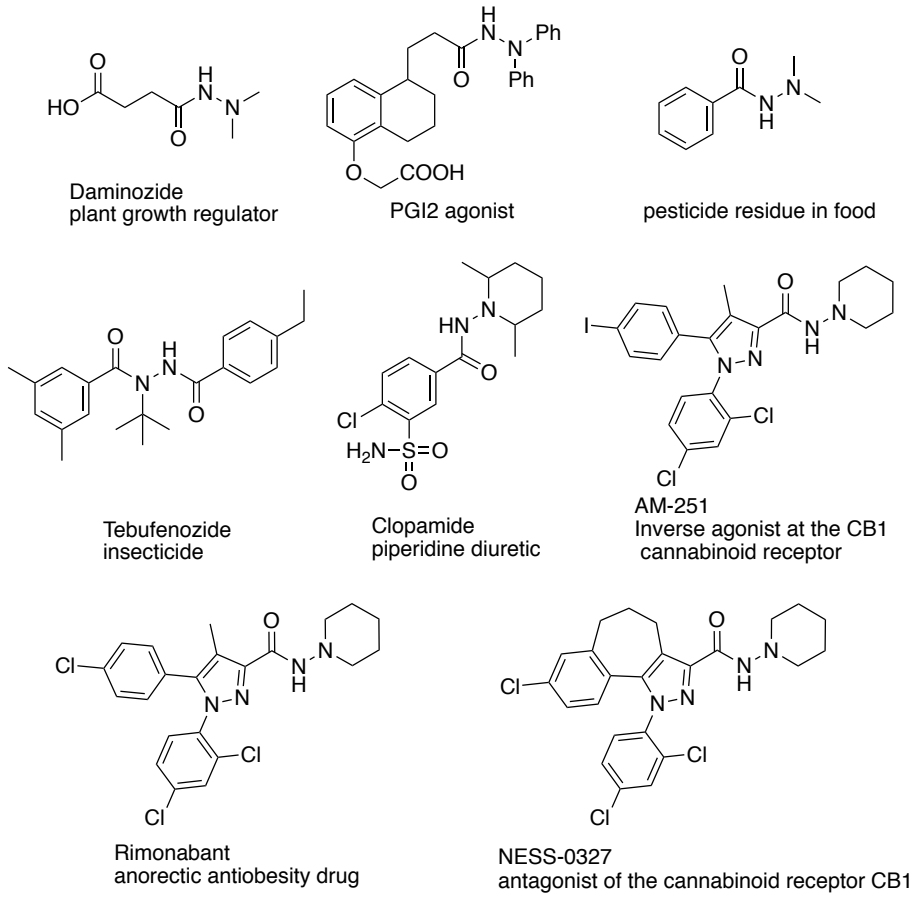


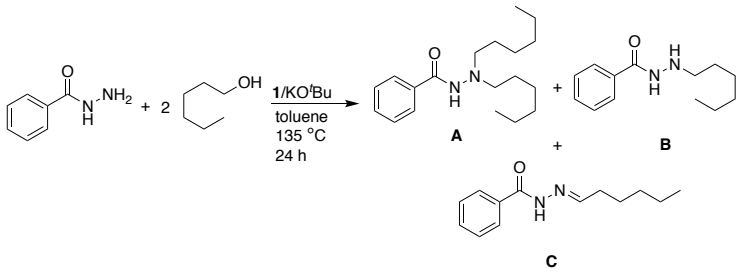
Figure S1. Biologically Active N,N-Disubstituted Acylhydrazides

Optimization of Experimental Conditions for the N,N-Dialkylation of Acylhydrazides:

Initial investigations were set on the optimization of reaction conditions required for direct catalytic N,N-dialkylation of acylhydrazides. Thus, reaction of benzohydrazide (0.5 mmol), 1-hexanol in toluene with ruthenium pincer catalyst **1** and KO^{t}Bu at 135 °C was performed. Interestingly, challenging formation of N,N-dialkylbenzohydrazide was observed predominantly and the corresponding hydrazone compound (**C**) also observed in minor amount (entry 1, Table S1). Notably, no mono-alkylation product (**B**) was observed under this condition. Increasing 1-hexanol to 2.2 equivalents resulted in slight enhancement of dialkylation product; however, mono-alkylation and hydrazone formation were observed (entry 2, Table S1). Decreasing the base load to 5 mol% provided 78% of the dialkylation product (entries 3,4, Table S1). Upon, increasing catalyst load to 2 mol% with base load at 5 mol%, the complete dialkylation occurred and the product *N,N*-di(1-hexyl)-

benzohydrazide was isolated in 98% yield (entry 4, Table S1). Further, varying temperature and base resulted in diminished yields (entries 5-7, Table S1). No dialkylation was observed on changing the solvent from toluene to polar solvent such as *tert*-amyl alcohol (entry 8, Table S1). Notably, no product formation was observed in the absence of catalyst as well as in the absence of catalyst and base (entries 9,10, Table S1).

Table S1. Optimization of Reaction Conditions[a]



entry	cat. (mol%)	base (mol%)	alcohol (equiv)	yield (%)[b]		
				A	B	C
1	1	100	2	60	-	30
2	1	100	2.2	68	5	20
3	1	5	2.2	78	10	4
4	**2**	**5**	**2.2**	**98**	**-**	**-**
5[c]	2	5	2.2	80	2	6
6	2	2	2.2	63	18	-
7[d]	2	5	2.2	93	2	-
8[e]	2	5	2.2	-	10	5
9[f]	-	5	2.2	-	-	5
10[g]	-	-	2.2	-	-	-

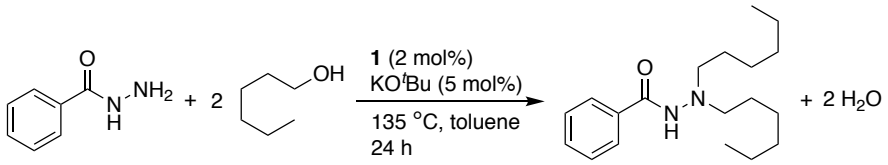
[a]Reaction conditions: catalyst **1**, KO^{t}Bu, benzohydrazide (0.5 mmol, 1 equiv.), 1-hexanol and toluene (2 mL) were heated at 135 °C in a Schlenk flask for 24 h. [b]Isolated yield after column chromatography. [c]Reaction was performed at 125 °C. [d]5 mol% of NaO^{t}Bu was used as a base. [e]The reaction was performed using *tert*-amyl alcohol as a solvent. [f]Only 5 mol% of KO^{t}Bu was used. [g]Reaction was performed without catalyst **1** and base.

General Procedure for Optimization of N,N-Dialkylation of Acylhydrazides Using Alcohols:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.01-0.02 mmol), base (1-0.02 mmol), benzohydrazide (0.5 mmol, 1 equiv), 1-hexanol (1.1 mmol, 2.2 equiv) and dry toluene (2 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring

in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC analysis. After 24 hours the reaction was stopped and cooled to room temperature and the solvent was evaporated. Further, the crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Yields were calculated for isolated pure products.

1 mmol Scale Reaction of Symmetrical N,N-Dialkylation of Benzohydrazide Using 1-Hexanol:



2d, 97% yield

Procedure:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.02 mmol), base (0.05 mmol), benzohydrazide (1 mmol, 1 equiv), 1-hexanol (2.2 mmol, 2.2 equiv) and dry toluene (2 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC analysis. After 24 hours the reaction was stopped and cooled to room temperature and the solvent was evaporated. Further, the crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). The reaction provided the product **2d** in 97% (295 mg) yield.

General Procedure for Symmetrical N,N-Dialkylation of Acylhydrazides Using Alcohols:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.02 mmol), KO^{t}Bu (0.05 mmol), acylhydrazide (0.5 mmol, 1 equiv), alcohol (1.1 mmol, 2.2 equiv) and dry toluene (2 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC

analysis. After 24 hours the reaction was stopped and cooled to room temperature. Further, the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Yields were calculated for isolated pure products.

General Procedure for N,N-Diethylation of Acylhydrazides Using Ethanol:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.02 mmol), KO^{t}Bu (0.05 mmol), acylhydrazide (0.5 mmol, 1 equiv), ethanol (5 mmol, 10 equiv) and dry toluene (1.5 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC analyses. After 24 hours the reaction was stopped and cooled to room temperature. Further, the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Yields were calculated for isolated pure products.

General Procedure for N,N-Dimethylation of Acylhydrazides Using Methanol:

An oven-dried pressure tube (35 mL) was equipped with a stir bar, catalyst **1** (0.05 mmol), KO^{t}Bu (0.5 mmol), acylhydrazide (0.5 mmol, 1 equiv) and dry methanol (2 mL) under nitrogen atmosphere in a glove box. The pressure tube was taken out of the glove box and the solution was heated at 150 °C (oil bath temperature) with stirring for 48 h. After 48 hours the reaction was stopped and cooled to room temperature. Further, pressure was released and the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Yields were calculated for isolated pure products.

General Procedure for Unsymmetrical N,N-Dialkylation of Acylhydrazides Using Alcohols:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.01 mmol), KO^{t}Bu (0.05 mmol), acylhydrazide (0.5 mmol, 1 equiv), alcohol (0.55 mmol, 1.1 equiv) and dry toluene (1.5 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 12 h. After 12 hours, catalyst **1** (0.01 mmol), base (0.05 mmol), alcohol (0.55 mmol, 1.1 equiv) and toluene (1 mL) were taken in a separate vial from glove box and the solution was added inside the reaction mixture under argon atmosphere. Further, the reaction continued for another 12 hours and the completion of the reaction was monitored using TLC analysis. After the completion the reaction was stopped and cooled to room temperature. Then, the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Yields were calculated for isolated pure products.

General Procedure for the Intermolecular Cyclization of Acylhydrazides Using Alcohols:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.02 mmol), KO^{t}Bu (0.05 mmol), acylhydrazide (0.5 mmol, 1 equiv), alcohol (1 mmol, 2 equiv) and dry toluene (2 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC analysis. After 24 hours the reaction was stopped and cooling to room temperature. Further, the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Yields were calculated for isolated pure products.

Procedure for One-Pot Synthesis of N,N-Dialkylated Acylhydrazide From Methyl Benzoate:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, methyl benzoate (0.5 mmol), hydrazine hydrate (5 mmol) and ethanol solvent (2 mL) refluxed for 4 hours. Further, the solvent was

evaporated and the flask was taken inside glove box. Catalyst **1** (0.02 mmol), KO^{t}Bu (0.05 mmol), acylhydrazide (0.5 mmol, 1 equiv), 1-hexanol (1.1 mmol, 2.2 equiv) and dry toluene (2 mL) were added to the same flask under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC analysis. After 24 hours the reaction was stopped and cooling to room temperature. Further, the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Product **2d** was isolated in 93% yield.

Spectral Data of N,N-Dialkyl Acylhydrazides:

3-Methoxy-*N*',*N*'-dipropylbenzohydrazide (2a): Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 124 mg, 99%. IR (DCM): 3263, 3052, 2962, 2874, 2836, 1654, 1582, 1484, 1265, 1041, 895, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.26-7.13 (m, 3H, ArC*H*), 6.96-6.90 (m, 1H, ArC*H*), 6.86 (s, N*H*), 3.74 (s, 3H, OCH_3), 2.70 (t, J = 8 Hz, 4H, CH_2), 1.55-1.45 (m, 4H, CH_2), 0.84 (t, J = 7.4 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.6, 159.7, 135.3, 129.5, 118.6, 117.6, 112.4, 60.1, 55.4, 20.3, 11.6. HRMS (ESI) m/z calcd for $C_{14}H_{22}N_2O_2$ $(M+H)^+$: 251.1754, found: 251.1775.

***N*',*N*'-Dibutylbenzohydrazide (2b):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 115 mg, 93%. IR (DCM): 3243, 2958, 2864, 1651, 1468, 1275, 748 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.68-7.63 (m, 2H, ArC*H*), 7.48-7.39 (m, 1H, ArC*H*), 7.38-7.30 (m, 2H, ArC*H*), 6.63 (s, 1H, N*H*), 2.76 (t, J = 7.6 Hz, 4H, CH_2), 1.48 (ddd, J_1 = 15.2 Hz, J_2 = 8.7 Hz, J_3 = 6.4 Hz, 4H, CH_2), 1.30 (dt, J_1 = 15.1 Hz, J_2 = 7.4 Hz, 4H, CH_2), 0.83 (t, J = 7.4 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.7, 134.2, 131.5, 128.6, 127.0, 58.2, 29.2, 20.5, 14.0. HRMS (ESI) m/z calcd for $C_{15}H_{24}N_2O$ $(M+H)^+$: 249.1961, found: 249.1971.

4-Ethoxy-*N*',*N*'-diisopentylbenzohydrazide (2c): Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 149 mg, 93%. IR (DCM): 3237, 3047, 2955, 2869, 1643, 1607, 1467, 1252, 1045, 765 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.63 (d, J = 8.7 Hz, 2H, ArC*H*), 6.83 (d, J = 7.8 Hz, 2H, ArC*H*), 6.52 (s, 1H, N*H*), 3.99 (tt, J_1 = 6.9 Hz, J_2 = 3.5 Hz, 2H, OCH_2), 2.76 (t, J = 8 Hz, 4H, CH_2), 1.55 (dt, J_1 = 13.2 Hz, J_1 = 6.6 Hz, 2H, C*H*), 1.37 (ddd, J_1 = 14.0 Hz, J_2 = 11.8 Hz, J_3 = 7.0 Hz, 7H, CH_2 & CH_3), 0.81 (d, J = 6.6 Hz, 12H, CH_3). $^{13}C\{^1H\}$ NMR

(100.6 MHz, $CDCl_3$): δ 166.3, 161.6, 128.8, 126.0, 114.3, 63.7, 57.0, 35.8, 26.4, 22.8, 14.7. HRMS (ESI) m/z calcd for $C_{19}H_{32}N_2O_2$ $(M+H)^+$: 321.2537, found: 321.2533.

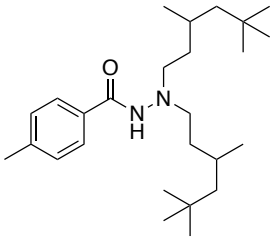
***N*',*N*'-Dihexylbenzohydrazide (2d):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 149 mg, 98% (for 0.5 mmol scale) and 295 mg, 97% (for 1 mmol scale). IR (DCM): 3430, 3053, 2956, 2930, 1672, 1457, 1265, 895, 741 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.66 (d, *J* = 7.3 Hz, 2H, ArC*H*), 7.43 (t, *J* = 7.3 Hz, 1H, ArC*H*), 7.35 (t, *J* = 7.4 Hz, 2H, ArC*H*), 6.57 (s, 1H, N*H*), 2.75 (t, *J* = 7.6 Hz, 4H, CH_2), 1.59-1.38 (m, 4H, CH_2), 1.27-1.19 (dd, J_1 = 26.5 Hz, J_1 = 8.0 Hz, 12H, CH_2), 0.79 (t, *J* = 6.7 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.7, 134.2, 131.6, 128.7, 127.0, 58.5, 31.8, 27.1, 27.0, 22.7, 14.1. HRMS (ESI) m/z calcd for $C_{19}H_{32}N_2O$ $(M+H)^+$: 305.2587, found: 305.2569.

***N*',*N*'-Di(hex-5-en-1-yl)benzohydrazide (2e):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 130 mg, 87%. IR (DCM): 3427, 3270, 3053, 2933, 2857, 1663, 1487, 1265, 1072, 914, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.66-7.62 (m, 2H, ArC*H*), 7.40 (dd, J_1 = 10.6 Hz, J_2 = 4.1 Hz, 1H, ArC*H*), 7.31 (t, *J* = 7.4 Hz, 2H, ArC*H*), 6.94 (s, 1H, N*H*), 5.69 (ddt, J_1 = 16.9 Hz, J_2 = 10.2 Hz, J_3 = 6.7 Hz, 2H, olefinic-C*H*), 4.86 (ddd, J_1= 13.7 Hz, J_2= 11.0 Hz, J_3= 1.2 Hz, 4H, olefinic-C*H*), 2.76 (t, *J* = 7.2 Hz, 4H, CH_2), 1.97 (dt, J_1 = 13.9 Hz, J_2 = 7.1 Hz, 4H, CH_2), 1.55-1.44 (m, 4H, CH_2), 1.35 (dt, J_1 = 14.9 Hz, J_2 = 7.4 Hz, 4H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.7, 138.6, 134.0, 131.4, 128.5, 127.0, 114.5, 57.9, 33.5, 26.5, 26.4. HRMS (ESI) m/z calcd for $C_{19}H_{28}N_2O$ $(M+H)^+$: 301.2274, found: 301.2290.

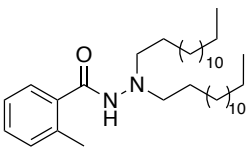
4-(*tert*-Butyl)-*N*',*N*'-dinonylbenzohydrazide (2f): Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 215 mg, 97%. IR (DCM): 3429, 3269, 3051, 2926, 2854, 1654, 1466, 1264, 1053, 895,

742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.61 (d, *J* = 8.3 Hz, 2H, ArC*H*), 7.35 (d, *J* = 8.2 Hz, 2H, ArC*H*), 6.66 (s, 1H, N*H*), 2.74 (t, *J* = 7.6 Hz, 4H, CH_2), 1.47 (dd, J_1 = 14.3 Hz, J_2 = 7.3 Hz, 4H, CH_2), 1.25 (s, 9H, 3×CH_3), 1.20-1.17 (m, 24H, CH_2), 0.79 (t, *J* = 6.7 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.6, 155.0, 131.2, 126.9, 125.5, 58.5, 34.9, 31.9, 31.2, 29.6, 29.3, 27.3, 27.1, 22.7, 14.1. HRMS (ESI) m/z calcd for $C_{29}H_{52}N_2O$ $(M+H)^+$: 445.4152, found: 445.4135.

4-Methyl-*N*',*N*'-bis(3,5,5-trimethylhexyl)benzohydrazide (2g): Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 197 mg, 98%. IR (DCM): 3246, 3043, 2954, 2866, 1644, 1466, 1264, 834, 741 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.55 (d, *J* = 7.8 Hz, 2H, ArC*H*), 7.14 (d, *J* = 7.8 Hz, 2H, ArC*H*), 6.55 (s, 1H, N*H*), 2.75 (t, *J* = 12.8 Hz, 4H, CH_2), 2.31 (s, 3H, CH_3), 1.50 (dd, J_1 = 18.1 Hz, J_1 = 8.1 Hz, 4H, CH_2), 1.40-1.27 (m, 2H, C*H*), 1.13 (dd, J_1 = 13.8 Hz, J_2 = 2.4 Hz, 2H, CH_2), 0.97 (dd, J_1 = 14.0 Hz, J_2 = 6.0 Hz, 2H, CH_2), 0.84 (t, *J* = 5.8 Hz, 6H, CH_3), 0.79 (s, 18H, 3×CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.6, 141.9, 131.3, 129.3, 127.0, 56.7, 51.4, 36.4, 31.1, 30.0, 27.6, 22.8, 21.5. HRMS (ESI) m/z calcd for $C_{26}H_{46}N_2O$ $(M+H)^+$: 403.3683, found: 403.3692.



2-Methyl-*N*',*N*'-ditetradecylbenzohydrazide (2h): Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 263 mg, 97%. IR (DCM): 3417, 3206, 3053, 2919, 2849, 1651, 1447, 1265, 895, 748 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.22 (q, *J* = 7.0 Hz, 2H, ArC*H*), 7.12 (dd, J_1 = 16.4 Hz, J_2 = 8.3 Hz, 2H, ArC*H*), 6.14 (s, 1H, N*H*), 2.72 (t, *J* = 7.6 Hz, 4H, CH_2), 2.37 (s, 3H, CH_3), 1.59-1.48 (m, 4H, CH_2), 1.35-1.07 (m, 44H, CH_2), 0.81 (t, *J* = 6.7 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 168.9, 136.4, 135.6, 131.0, 130.0, 126.7, 125.7, 58.6, 32.0, 29.8, 29.8, 29.7, 29.7, 29.7, 29.4, 27.3, 27.1, 22.8, 19.7, 14.2. HRMS (ESI) m/z calcd for $C_{36}H_{66}N_2O$ $(M+H)^+$: 543.5248, found: 543.5254.



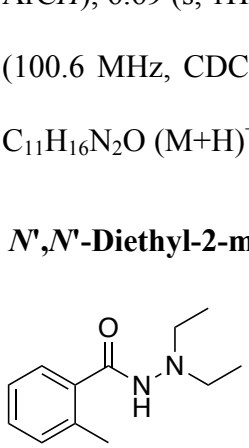
***N'*,*N'*-bis(4-Phenylbutyl)-1-naphthohydrazide (2i):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 209 mg, 93%. IR (DCM): 3413, 3317, 3053, 2937, 2859, 1676, 1452, 1265, 895, 738 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 8.15-8.07 (m, 1H, ArC*H*), 7.82-7.71 (m, 2H, ArC*H*), 7.45-7.39 (m, 2H, ArC*H*), 7.31-7.19 (m, 2H, ArC*H*), 7.15 (t, J = 7.4 Hz, 4H, ArC*H*), 7.06 (dd, J_1 = 12.8 Hz, J_2 = 7.0 Hz, 6H, ArC*H*), 6.35 (s, 1H, N*H*), 2.74 (t, J = 6.7 Hz, 4H, CH_2), 2.55 (t, J = 7.1 Hz, 4H, CH_2), 1.70-1.50 (m, 8H, CH_2). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 100.6 MHz): δ 168.5, 142.3, 133.6, 133.3, 130.6, 130.3, 128.5, 128.3, 128.3, 128.2, 127.2, 126.5, 125.7, 125.3, 124.8, 124.6, 58.1, 35.8, 28.9, 26.7. HRMS (ESI) m/z calcd for $C_{31}H_{34}N_2O$ $(M+H)^+$: 451.2744, found: 451.2758.

***N'*,*N'*-Bis(3,7-dimethyloct-6-en-1-yl)benzohydrazide (2j):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. Colorless liquid. Yield: 179 mg, 87%. IR (DCM): 3422, 3238, 3056, 2958, 2925, 2856, 1650, 1461, 1267, 740 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.65 (d, J = 7.4 Hz, 2H, ArC*H*), 7.42 (t, J = 7.3 Hz, 1H, ArC*H*), 7.34 (t, J = 7.5 Hz, 2H, ArC*H*), 6.57 (s, 1H, N*H*), 4.99 (t, J = 7.0 Hz, 2H, olefinic-C*H*), 2.78 (tt, J_1 = 16.2 Hz, J_2 = 8.1 Hz, 4H, CH_2), 1.99-1.77 (m, 4H, CH_2), 1.59 (s, 6H, CH_3), 1.49 (s, 6H, CH_3), 1.30 (dddd, J_1 = 15.2 Hz, J_2 = 12.0 Hz, J_3 = 11.1 Hz, J_4 = 6.1 Hz, 8H, CH_2 &C*H*), 1.16-1.02 (m, 2H, CH_2), 0.82 (d, J = 6.5 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.7, 134.2, 131.5, 131.2, 128.7, 127.0, 124.8, 56.6, 37.2, 34.0, 30.8, 25.7, 25.5, 19.7, 17.7. HRMS (ESI) m/z calcd for $C_{27}H_{44}N_2O$ $(M+H)^+$: 413.3526, found: 413.3529.

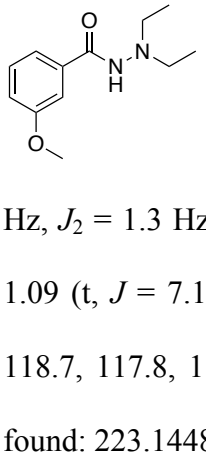
***N'*,*N'*-Diethylbenzohydrazide (2k):** Purified by silica gel column chromatography using ethyl acetate/hexane (30:70) mixture as an eluent. White solid. Yield: 85 mg, 89%. IR (DCM): 3245, 2957, 2865, 1656, 1466, 1265, 741 cm^{-1}. ^1H NMR (400 MHz,

$CDCl_3$): δ 7.68 (d, *J* = 7.3 Hz, 2H, ArC*H*), 7.43 (t, *J* = 7.3 Hz, 1H, ArC*H*), 7.35 (t, *J* = 7.4 Hz, 2H, ArC*H*), 6.69 (s, 1H, N*H*), 2.83 (q, *J* = 7.1 Hz, 4H, CH_2), 1.09 (t, *J* = 7.1 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 167.1, 134.0, 131.6, 128.7, 127.0, 52.2, 12.0. HRMS (ESI) m/z calcd for $C_{11}H_{16}N_2O$ $(M+H)^+$: 193.1335, found: 193.1329.

***N*',*N*'-Diethyl-2-methylbenzohydrazide (2l):** Purified by silica gel column chromatography using ethyl acetate/hexane (30:70) mixture as an eluent. White solid. Yield: 96 mg, 93%. IR (DCM): 3243, 2958, 2867, 1657, 1465, 1265, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.23 (t, *J* = 7.2 Hz, 2H, ArC*H*), 7.12 (dd, J_1 = 16.9 Hz, J_2 = 8.2 Hz, 2H, ArC*H*), 6.11 (s, 1H, N*H*), 2.77 (q, *J* = 7.0 Hz, 4H, CH_2), 2.37 (s, 3H, CH_3), 1.12 (t, *J* = 7.1 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 169.2, 136.2, 135.5, 130.9, 129.9, 126.6, 125.6, 52.3, 19.6, 12.1. HRMS (ESI) m/z calcd for $C_{12}H_{18}N_2O$ $(M+H)^+$: 207.1492, found: 207.1491.



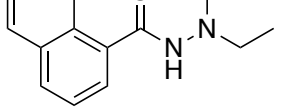
***N*',*N*'-Diethyl-3-methoxybenzohydrazide (2m):** Purified by silica gel column chromatography using ethyl acetate/hexane (30:70) mixture as an eluent. White solid. Yield: 108 mg, 97%. IR (DCM): 3266, 3053, 2967, 2878, 1647, 1583, 1487, 1265, 1043, 895, 740 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.31-7.16 (m, 3H, ArC*H*), 6.96 (dd, J_1 = 7.9 Hz, J_2 = 1.3 Hz, 1H, ArC*H*), 6.60 (s, 1H, N*H*), 3.77 (s, 3H, OCH_3), 2.81 (q, *J* = 7.1 Hz, 4H, CH_2), 1.09 (t, *J* = 7.1 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.9, 159.9, 135.4, 129.6, 118.7, 117.8, 112.5, 55.5, 52.3, 12.0. HRMS (ESI) m/z calcd for $C_{12}H_{18}N_2O_2$ $(M+H)^+$: 223.1441, found: 223.1448.



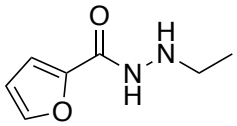
4-Amino-*N*',*N*'-diethylbenzohydrazide (2n): Purified by silica gel column chromatography using ethyl acetate/hexane (70:30) mixture as an eluent. Pale yellow solid. Yield: 65 mg, 63%. IR (DCM): 3466, 3265, 3052, 2950, 2874, 1647, 1487, 1265, 1041, 896, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.54 (d, *J* = 8.5 Hz, 2H, ArC*H*), 6.59 (t, 3H, ArC*H* & N*H*), 3.93 (s, 2H, NH_2), 2.85 (dd, J_1 = 14.2 Hz, J_2 = 7.1

Hz, 4H, CH_2), 1.09 (t, J = 7.1 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.9, 149.9, 128.7, 122.9, 114.2, 52.0, 11.9. HRMS (ESI) m/z calcd for $C_{11}H_{17}N_3O$ $(M+H)^+$: 208.1444, found: 208.1459.

***N*',*N*'-Diethyl-1-naphthohydrazide (2o):** Purified by silica gel column chromatography using ethyl acetate/hexane (30:70) mixture as an eluent. White solid. Yield: 120 mg, 99%. IR (DCM): 3266, 3054, 2963, 2874, 2835, 1653, 1484, 1265, 895, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 8.19 (d, J = 8.2 Hz, 1H, ArC*H*), 7.86-7.74 (m, 2H, ArC*H*), 7.50-7.42 (m, 3H, ArC*H*), 7.35 (t, J = 7.6 Hz, 1H, ArC*H*), 6.33 (s, 1H, N*H*), 2.83 (q, J = 7.0 Hz, 4H, CH_2), 1.18 (t, J = 7.1 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 168.8, 133.7, 133.4, 130.8, 130.4, 128.3, 127.4, 126.6, 125.4, 124.8, 124.6, 52.5, 12.2. HRMS (ESI) m/z calcd for $C_{15}H_{18}N_2O$ $(M+H)^+$: 243.1492, found: 243.1477.



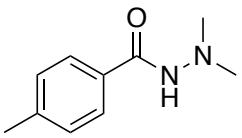
***N*'-Ethylfuran-2-carbohydrazide (2p):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 58 mg, 75%. IR (DCM): 3263, 3052, 2987, 2854, 1672, 1447, 1267, 1047, 895, 747 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.38 (d, J = 0.8 Hz, 1H, ArC*H*), 7.07 (d, J = 3.4 Hz, 1H, ArC*H*), 6.43 (dd, J_1 = 3.5 Hz, J_2 = 1.7 Hz, 1H, ArC*H*), 5.27 (s, 1H, N*H*), 2.91 (q, J = 7.1 Hz, 2H, CH_2), 1.07 (t, J = 7.2 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 158.2, 146.9, 144.2, 114.8, 112.1, 46.7, 13.0. HRMS (ESI) m/z calcd for $C_7H_{10}N_2O_2$ $(M+H)^+$: 155.0815, found: 155.0811.



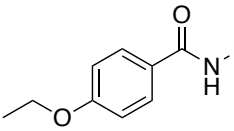
***N*',*N*'-Dimethylbenzohydrazide (2q):** Purified by silica gel column chromatography using ethyl acetate/hexane (50:50) mixture as an eluent. White solid. Yield: 76 mg, 93%. IR (DCM): 3263, 3052, 2962, 2850, 1657, 1580, 1484, 1267, 1041, 895, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.66 (d, J = 7.4 Hz, 2H, ArC*H*), 7.41 (d, J = 7.1 Hz,

1H, ArC*H*), 7.33 (dd, J_1 = 11.4 Hz, J_2 = 4.2 Hz, 2H, ArC*H*), 6.97 (s, 1H, N*H*), 2.63 (s, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.8, 133.8, 131.6, 128.6, 127.1, 47.7. HRMS (ESI) m/z calcd for $C_9H_{12}N_2O$ $(M+H)^+$: 165.1022, found: 165.1022.

***N*',*N*',4-Trimethylbenzohydrazide (2r):** Purified by silica gel column chromatography using ethyl acetate/hexane (75:25) mixture as an eluent. White solid. Yield: 76.5 mg, 86%. IR (DCM): 3263, 3052, 2962, 2874, 2836, 1654, 1265, 1041, 895, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.58 (d, *J* = 7.1 Hz, 2H, ArC*H*), 7.16 (t, *J* = 13.4 Hz, 3H, ArC*H* & N*H*), 2.66 (s, 6H, CH_3), 2.32 (s, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.8, 142.2, 130.7, 129.3, 127.1, 47.7, 21.5. HRMS (ESI) m/z calcd for $C_{10}H_{14}N_2O$ $(M+H)^+$: 179.1178, found: 179.1184.



4-Ethoxy-*N*',*N*'-dimethylbenzohydrazide (2s): Purified by silica gel column chromatography using ethyl acetate/hexane (50:50) mixture as an eluent. White solid. Yield: 90 mg, 87%. IR (DCM): 3264, 3052, 2987, 2850, 1660, 1447, 1260, 895, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.63 (d, *J* = 8.1 Hz, 2H, ArC*H*), 6.87-6.76 (m, 3H, ArC*H* & N*H*), 3.99 (q, *J* = 7.0 Hz, 2H, CH_2), 2.63 (s, 6H, CH_3), 1.35 (t, *J* = 7.0 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.4, 161.7, 128.9, 125.8, 114.3, 63.7, 47.8, 14.7. HRMS (ESI) m/z calcd for $C_{11}H_{16}N_2O_2$ $(M+H)^+$: 209.1286, found: 209.1290.



4-Bromo-*N*',*N*'-dimethylbenzohydrazide (2t): Purified by silica gel column chromatography using ethyl acetate/hexane (70:30) mixture as an eluent. White solid. Yield: 103 mg, 85%. IR (DCM): 3265, 3052, 2966, 2878, 1657, 1480, 1267, 1045, 895, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.55 (d, *J* = 8.5 Hz, 2H, ArC*H*), 7.48 (d, *J* = 8.4 Hz, 2H, ArC*H*), 7.06 (s, 1H, N*H*), 2.64 (s, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 164.9, 132.5, 131.9, 128.8, 127.1, 126.4, 47.6. HRMS (ESI) m/z calcd for $C_9H_{11}BrN_2O$ $(M+H)^+$: 243.0127, found: 243.0142

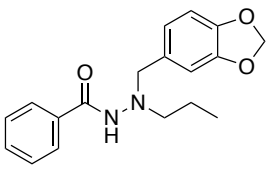
***N*',*N*'-Dimethyl-4-(methylamino)benzohydrazide (2u):** Purified by silica gel column chromatography using ethyl acetate solvent (100%) as an eluent. White solid. Yield: 72 mg, 75%. IR (DCM): 3440, 3260, 3050, 2988, 2853, 1661, 1447, 1265, 1047, 895, 743 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.54 (d, *J* = 7.2 Hz, 2H, ArC*H*), 6.74 (s, 1H, amide-N*H*), 6.52-6.43 (m, 2H, ArC*H*), 3.37 (s, 1H, amine-N*H*), 2.78 (s, 3H, CH_3), 2.61 (s, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.8, 152.1, 128.8, 121.4, 111.4, 47.9, 30.3. HRMS (ESI) m/z calcd for $C_{10}H_{15}N_3O$ $(M+H)^+$: 194.1287, found: 194.1290.

***N*',*N*'-Dimethyl-1-naphthohydrazide (2v):** Purified by silica gel column chromatography using ethyl acetate/hexane (75:25) mixture as an eluent.White solid. Yield: 96 mg, 90%. IR (DCM): 3250, 3059, 2980, 2857, 1650, 1447, 1266, 1047, 897, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 8.27-8.17 (m, 1H, ArC*H*), 7.91-7.79 (m, 2H, ArC*H*), 7.58-7.44 (m, 3H, ArC*H*), 7.44-7.34 (m, 1H, ArC*H*), 6.84 (s, 1H, N*H*), 2.76-2.72 (m, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 167.4, 133.7, 132.8, 130.9, 130.4, 128.4, 127.3, 126.6, 125.2, 124.7, 47.7. HRMS (ESI) m/z calcd for $C_{13}H_{14}N_2O$ $(M+H)^+$: 215.1178, found: 215.1189.

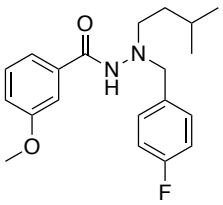
***N*'-Butyl-*N*'-(4-isopropylbenzyl)-3-methoxybenzohydrazide (3a):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 166 mg, 94%. IR (DCM): 3250, 3056, 2987, 2857, 1640, 1447, 1267, 1048, 897, 741 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.19 (t, *J* = 5.0 Hz, 2H, ArC*H*), 7.16 (d, *J* = 7.9 Hz, 1H, ArC*H*), 7.13-7.06 (m, 3H, ArC*H*), 7.01-6.96 (m, 1H, ArC*H*), 6.91 (ddd, J_1 = 8.3 Hz, J_2 = 2.6 Hz, J_3 = 0.9 Hz, 1H, ArC*H*), 6.76 (s, 1H, N*H*), 4.05 (s, 2H, CH_2), 3.72 (s, 3H, OCH_3), 2.83 (ddd, J_1 = 20.7 Hz, J_2 = 14.3 Hz, J_3 = 7.1 Hz, 3H, CH_2&C*H*), 1.48 (ddd, J_1 = 14.9 Hz, J_2 = 8.5 Hz, J_3 = 6.3 Hz, 2H, CH_2), 1.40-1.24 (m, 2H, CH_2), 1.15 (d, *J* = 6.9 Hz, 6H, CH_3), 0.82 (t, *J* = 7.3 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.7, 159.8,

148.3, 135.6, 133.6, 129.6, 129.6, 126.5, 118.7, 117.8, 112.3, 60.7, 56.1, 55.4, 33.8, 29.6, 24.0, 20.4, 14.1. HRMS (ESI) m/z calcd for $C_{22}H_{30}N_2O_2$ $(M+H)^+$: 355.2380, found: 355.2403.

***N*'-(Benzo[*d*][1,3]dioxol-5-ylmethyl)-*N*'-propylbenzohydrazide (3b):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 131 mg, 84%. IR (DCM): 3251, 3057, 2980, 2856, 1647, 1440, 1268, 1047, 897, 747 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.56-7.51 (m, 2H, ArC*H*), 7.40 (t, *J* = 7.4 Hz, 1H, ArC*H*), 7.31 (t, *J* = 7.5 Hz, 2H, ArC*H*), 6.83 (s, 1H, ArC*H*), 6.76 (s, 1H, N*H*), 6.70 (dd, J_1 = 19.2 Hz, J_2 = 8.4 Hz, 2H, ArC*H*), 5.87 (s, 2H, OCH_2), 4.01 (s, 2H, CH_2), 2.90-2.77 (m, 2H, CH_2), 1.53 (dt, J_1 = 14.8 Hz, J_2 = 7.4 Hz, 2H, CH_2), 0.87 (t, *J* = 7.4 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.9, 147.8, 147.1, 134.0, 131.6, 130.3, 128.7, 126.9, 122.8, 109.9, 108.1, 101.0, 60.7, 57.9, 20.8, 11.7. HRMS (ESI) m/z calcd for $C_{18}H_{20}N_2O_3$ $(M+H)^+$: 313.1547, found: 313.1570.



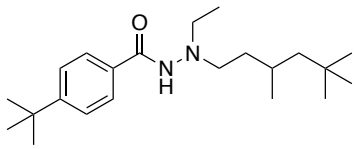
***N*'-(4-Fluorobenzyl)-*N*'-isopentyl-3-methoxybenzohydrazide (3c):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 110 mg, 64%. IR (DCM): 3255, 3059, 2960, 2888, 1657, 1440, 1264, 1049, 890, 745 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.26 (dd, J_1 = 8.3 Hz, J_2 = 5.6 Hz, 2H, ArC*H*), 7.20-7.14 (m, 1H, ArC*H*), 7.08 (s, 1H, ArC*H*), 6.97 (d, *J* = 7.6 Hz, 1H, ArC*H*), 6.94-6.89 (m, 3H, ArC*H*), 6.80 (s, 1H, N*H*), 4.04 (s, 2H, CH_2), 3.72 (s, 3H, OCH_3), 3.04-2.79 (m, 2H, CH_2), 1.59 (td, J_1 = 13.4 Hz, J_2 = 6.7 Hz, 1H, C*H*), 1.39 (dd, J_1 = 14.9 Hz, J_2 = 7.0 Hz, 2H, CH_2), 0.80 (d, *J* = 6.6 Hz, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.8, 163.5, 161.1, 159.8, 135.4, 132.6, 131.0, 131.0, 129.7, 128.8, 128.7, 118.6, 117.8, 115.3, 115.1, 112.3, 60.3, 55.4, 54.7, 36.3, 26.1, 22.7. HRMS (ESI) m/z calcd for $C_{20}H_{25}FN_2O_2$ $(M+H)^+$: 345.1973, found: 345. 1991.



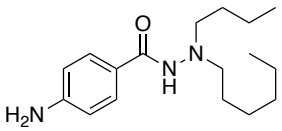
***N*'-Hexyl-*N*'-(3-(pyridin-2-yl)propyl)benzohydrazide (3d):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. Pale yellow solid. Yield: 136 mg, 80%. IR (DCM): 3257, 3050, 2969, 2887, 1658, 1447, 1260, 1045, 890, 747 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 8.40 (d, *J* = 4.8 Hz, 1H, ArC*H*), 7.70-7.64 (m, 2H, ArC*H*), 7.49 (td, J_1 = 7.7 Hz, J_2 = 1.7 Hz, 1H, ArC*H*), 7.41 (t, *J* = 7.4 Hz, 1H, ArC*H*), 7.33 (t, *J* = 7.4 Hz, 2H, ArC*H*), 7.11 (d, *J* = 7.8 Hz, 1H, ArC*H*), 7.01 (dd, J_1 = 6.9 Hz, J_2 = 5.4 Hz, 1H, ArC*H*), 6.91 (s, 1H, N*H*), 2.87-2.74 (m, 6H, CH_2), 1.98-1.86 (m, 2H, CH_2), 1.53-1.39 (m, 2H, CH_2), 1.30-1.14 (m, 6H, CH_2), 0.78 (t, *J* = 6.8 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.8, 161.8, 149.0, 136.5, 134.1, 131.5, 128.6, 127.1, 123.2, 121.1, 58.3, 57.3, 35.7, 31.8, 27.1, 27.1, 26.9, 22.6, 14.1. HRMS (ESI) m/z calcd for $C_{21}H_{29}N_3O$ $(M+H)^+$: 340.2383, found: 340.2400.

***N*'-(4-Aminobenzyl)-4-ethoxy-*N*'-heptylbenzohydrazide (3e):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. Pale yellow solid. Yield: 149 mg, 78%. IR (DCM): 3422, 3255, 3056, 2955, 2886, 1647,1480, 1268, 1040, 890, 748 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.50 (d, *J* = 8.4 Hz, 2H, ArC*H*), 7.03 (d, *J* = 7.9 Hz, 2H, ArC*H*), 6.77 (d, *J* = 8.5 Hz, 2H, ArC*H*), 6.64 (s, 1H, N*H*), 6.55 (d, *J* = 7.9 Hz, 2H, ArC*H*), 4.02-3.90 (m, 4H, CH_2), 3.66 (s, 2H, NH_2), 2.86-2.72 (m, 2H, CH_2), 1.47 (dd, J_1 = 13.9 Hz, J_2 = 7.1 Hz, 2H, CH_2), 1.32 (dd, J_1 = 15.3 Hz, J_2 = 8.4 Hz, 3H, CH_3), 1.30-1.07 (m, 10H, CH_2), 0.78 (t, *J* = 6.2 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.1, 161.6, 145.9, 131.0, 128.7, 126.1, 125.6, 115.0, 114.2, 63.6, 60.6, 56.2, 31.8, 29.2, 27.5, 27.2, 22.6, 14.7, 14.1. HRMS (ESI) m/z calcd for $C_{23}H_{33}N_3O_2$ $(M+H)^+$: 384.2646, found: 384.2797.

4-(*tert*-Butyl)-*N*'-ethyl-*N*'-(3,5,5-trimethylhexyl)benzohydrazide (3f): Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 166 mg, 96%. IR (DCM): 3255, 3052, 2986, 2852, 1647, 1447, 1265, 1047, 897, 740 cm^{-1}. ^1H NMR

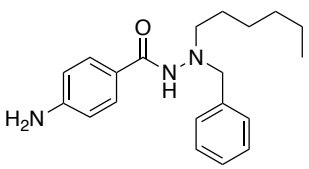


(400 MHz, $CDCl_3$): δ 7.61 (d, *J* = 8.3 Hz, 2H, ArC*H*), 7.37 (d, *J* = 8.3 Hz, 2H, ArC*H*), 6.51 (s, 1H, N*H*), 2.90-2.64 (m, 4H, CH_2), 1.57-1.42 (m, 2H, CH_2), 1.41-1.30 (m, 1H, C*H*), 1.25 (s, 9H, 3×CH_3), 1.14 (dd, J_1 = 13.8 Hz, J_2 = 3.2 Hz, 1H, CH_2), 1.07 (t, *J* = 7.1 Hz, 3H, CH_3), 0.98 (dd, J_1 = 14.0 Hz, J_2 = 6.1 Hz, 1H, CH_2), 0.85 (d, *J* = 6.4 Hz, 3H, CH_3), 0.80 (s, 9H, 3×CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.8, 155.1, 131.2, 126.9, 125.6, 56.5, 52.6, 51.4, 36.4, 35.0, 31.2, 31.1, 30.0, 27.6, 22.8, 12.1. HRMS (ESI) m/z calcd for $C_{22}H_{38}N_2O$ $(M+H)^+$: 347.3057, found: 347.3076.



4-Amino-*N*'-butyl-*N*'-hexylbenzohydrazide (3g): Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 98 mg, 67%. IR (DCM): 3476. 3256, 3056, 2960, 2887, 1647, 1442, 1265, 1040, 890, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.50 (d, *J* = 8.3 Hz, 2H, ArC*H*), 6.57 (d, *J* = 8.4 Hz, 3H, ArC*H* & N*H*), 3.92 (s, 2H, NH_2), 2.79-2.68 (m, 4H, CH_2), 1.45 (dd, J_1 = 13.2 Hz, J_2 = 6.5 Hz, 4H, CH_2), 1.32-1.13 (m, 8H, CH_2), 0.86-0.74 (m, 6H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.5, 149.8, 128.7, 123.3, 114.2, 58.7, 58.3, 31.8, 29.2, 27.0, 22.6, 20.5, 14.1. HRMS (ESI) m/z calcd for $C_{17}H_{29}N_3O$ $(M+H)^+$: 292.2383, found: 292.2390.

4-Amino-*N*'-benzyl-*N*'-hexylbenzohydrazide (3h): Purified by silica gel column chromatography



using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 91 mg, 56%. IR (DCM): 3422, 3052, 2986, 2856, 1650, 1448, 1265, 1047, 895, 743 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.36 (d, *J* = 8.3 Hz, 2H, ArC*H*), 7.23 (dq, J_1 = 13.5 Hz, J_2 = 7.4 Hz, 5H, ArC*H*), 6.70 (s, 1H, N*H*), 6.51 (d, *J* = 8.5 Hz, 2H, ArC*H*), 4.08 (s, 2H, CH_2), 3.90 (s, 2H, NH_2), 2.93-2.81 (m, 2H, CH_2), 1.48 (dt, J_1 = 14.7 Hz, J_2 = 7.3 Hz, 2H, CH_2), 1.32-1.14 (m, 6H, CH_2), 0.78 (t, *J* = 6.7 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.6, 149.8, 136.5, 129.7, 128.6, 128.4, 127.5, 123.3, 114.2, 61.1, 56.4, 31.8, 27.5, 26.9, 22.7, 14.1. HRMS (ESI) m/z calcd for $C_{20}H_{27}N_3O$ $(M+H)^+$: 326.2227, found: 326.2236.

***N*'-Butyl-*N*'-cyclohexylbenzohydrazide (3i):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 131 mg, 96%. IR (DCM): 3256, 3052, 2967, 2886, 1649, 1447, 1265, 1046, 880, 747 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.68 (d, *J* = 6.5 Hz, 2H, ArC*H*), 7.42 (d, *J* = 6.5 Hz, 1H, ArC*H*), 7.36 (t, *J* = 6.7 Hz, 2H), 6.60 (s, 1H, N*H*), 2.74 (d, *J* = 30.1 Hz, 3H, CH_2 & C*H*), 1.92 (s, 2H, CH_2), 1.73 (s, 2H, CH_2), 1.44-1.61 (m, 3H, CH_2), 1.24 (dt, J_1 = 48.8 Hz, J_2 = 14.8 Hz, 7H, CH_2), 0.83 (t, *J* = 6.7 Hz, 3H, CH_3). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 167.1, 134.2, 131.5, 128.7, 127.1, 64.4, 54.3, 29.3, 29.0, 26.0, 25.3, 20.5, 14.1. HRMS (ESI) m/z calcd for $C_{17}H_{26}N_2O$ $(M+H)^+$: 275.2118, found: 275.2099.

***N*'-Cycloheptyl-4-methyl-*N*'-(3-phenylpropyl)benzohydrazide (3j):** Purified by silica gel column chromatography using ethyl acetate/hexane (10:90) mixture as an eluent. White solid. Yield: 146 mg, 80%. IR (DCM): 3255, 3056, 2967, 2880, 1647, 1449, 1266, 1046, 890, 744 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.54 (d, *J* = 8.0 Hz, 2H, ArC*H*), 7.20-7.05 (m, 7H, ArC*H*), 6.47 (s, 1H, N*H*), 3.00-2.79 (m, 1H, C*H*), 2.70 (t, *J* = 7.2 Hz, 2H, CH_2), 2.63 (t, *J* = 7.6 Hz, 2H, CH_2), 2.32 (s, 3H, CH_3), 1.91 (dd, J_1 = 13.0 Hz, J_2 = 4.1 Hz, 2H, CH_2), 1.81 (dd, J_1 = 14.8 Hz, J_2 = 7.5 Hz, 2H, CH_2), 1.67-1.54 (m, 2H, CH_2), 1.41 (ddd, J_1 = 26.0 Hz, J_2 = 12.2 Hz, J_3 = 6.0 Hz, 8H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 167.0, 142.2, 142.0, 131.4, 129.3, 128.6, 128.4, 127.1, 125.8, 66.4, 54.6, 33.5, 29.8, 29.2, 28.1, 25.2, 21.5. HRMS (ESI) m/z calcd for $C_{24}H_{32}N_2O$ $(M+H)^+$: 365.2587, found: 365.2630.

4-Methyl-*N*-(pyrrolidin-1-yl)benzamide (4a): Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 92 mg, 90%. IR (DCM): 3250, 3059, 2960, 2886, 1650, 1447, 1267, 1058, 898, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.57 (d, *J* = 7.7 Hz, 2H, ArC*H*), 7.14 (d, *J* = 7.9 Hz, 2H, ArC*H*), 3.14-2.83 (m, 4H, CH_2), 2.31 (s, 3H, CH_3), 1.96-1.77 (m, 4H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$):

δ 166.3, 142.1, 131.0, 129.3, 127.1, 55.7, 22.4, 21.5. HRMS (ESI) m/z calcd for $C_{12}H_{16}N_2O$ $(M+H)^+$: 205.1335, found: 205.1330.

4-(*tert*-Butyl)-*N*-(pyrrolidin-1-yl)benzamide (4b): Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 106 mg, 86%. IR (DCM): 3267, 3010, 2966, 2889, 1657, 1447, 1270, 1046, 890, 739 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.61 (d, *J* = 8.0 Hz, 2H, ArC*H*), 7.35 (d, *J* = 8.1 Hz, 2H, ArC*H*), 3.11-2.76 (m, 4H, CH_2), 1.97-1.70 (m, 4H, CH_2), 1.25 (s, 9H, 3×CH_3) $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.2, 155.2, 130.9, 127.0,125.6, 55.6, 35.0, 31.2, 22.4. (ESI) m/z calcd for $C_{15}H_{22}N_2O$ $(M+H)^+$: 247.1805, found: 247.1817.

***N*-(Pyrrolidin-1-yl)-1-naphthamide (4c):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 100 mg, 83%. IR (DCM): 3250, 3077, 2960, 2889, 1650, 1448, 1260, 1049, 890, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 8.15 (d, *J* = 7.6 Hz, 1H, ArC*H*), 7.75 (dt, J_1 = 19.0 Hz, J_1 = 9.4 Hz, 2H, ArC*H*), 7.47-7.37 (m, 3H, ArC*H*), 7.29 (t, *J* = 7.6 Hz, 1H, ArC*H*), 6.88 (s, 1H, N*H*), 3.04-2.80 (m, 4H, CH_2), 1.94-1.62 (m, 4H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 167.9, 133.6, 133.0, 130.6, 130.3, 128.3, 127.1, 126.4, 125.3, 125.1, 124.6, 55.5, 22.2. (ESI) m/z calcd for $C_{15}H_{16}N_2O$ $(M+H)^+$: 241.1335, found: 241.1325.

***N*-(Piperidin-1-yl)benzamide (4d):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 77 mg, 75%. IR (DCM): 3300, 3109, 2956, 2887, 1651, 1447, 1265, 1049, 890, 747 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.66 (d, *J* = 7.1 Hz, 2H, ArC*H*), 7.47-7.38 (m, 1H, ArC*H*), 7.34 (t, *J* = 7.3 Hz, 2H, ArC*H*), 6.86 (s, 1H, N*H*), 2.88-2.63 (m, 4H, CH_2), 1.78-1.52 (m, 4H, CH_2), 1.44-1.24 (m, 2H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.4, 134.1, 131.6, 128.6, 127.1, 57.2, 25.4, 23.3. (ESI) m/z calcd for $C_{12}H_{16}N_2O$ $(M+H)^+$: 205.1335, found: 205.1336.

3-Methoxy-*N*-(piperidin-1-yl)benzamide (4e): Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 84 mg, 72%. IR (DCM): 3250, 3053, 2956, 2878, 1649, 1449, 1262, 1047, 898, 745 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.29-7.13 (m, 4H, ArC*H* & N*H*), 6.95 (dd, J_1 = 8.2 Hz, J_2 = 2.4 Hz, 1H, ArC*H*), 3.77 (s, 3H, OCH_3), 2.91-2.75 (m, 4H, CH_2), 1.69 (dt, J_1 = 11.1 Hz, J_2 = 5.6 Hz, 4H, CH_2), 1.38 (dd, J_1 = 14.9 Hz, J_2 = 9.9 Hz, 2H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.3, 159.9, 135.5, 129.6, 118.9, 117.7, 112.6, 57.2, 55.5, 25.4, 23.3. (ESI) m/z calcd for $C_{13}H_{18}N_2O_2$ $(M+H)^+$: 235.1441, found: 235.1464.

4-Ethoxy-*N*-(piperidin-1-yl)benzamide (4f): Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 87 mg, 70%. IR (DCM): 3266, 3156, 2960, 1652, 1447, 1269, 1040, 890, 751 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.63 (d, *J* = 8.2 Hz, 2H, ArC*H*), 6.81 (d, *J* = 8.6 Hz, 2H, ArC*H*), 3.99 (q, *J* = 7.0 Hz, 2H, CH_2), 2.95-2.54 (m, 4H, CH_2), 1.85-1.51 (m, 4H, CH_2), 1.35 (t, *J* = 7.0 Hz, 5H, CH_3 & CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.0, 161.7, 128.9, 126.0, 114.3, 63.7, 57.3, 25.4, 23.3, 14.8. (ESI) m/z calcd for $C_{14}H_{20}N_2O_2$ $(M+H)^+$: 249.1597, found: 249.1603.

4-Fluoro-*N*-(piperidin-1-yl)benzamide (4g): Purified by silica gel column chromatography using ethyl acetate/hexane (45:55) mixture as an eluent. White solid. Yield: 70 mg, 63%. IR (DCM): 3253, 3052, 2956, 2881, 1649, 1447, 1269, 1047, 890, 742 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ 7.85 (s, 1H, N*H*), 7.72-7.66 (m, 2H, ArC*H*), 7.02 (t, *J* = 8.4 Hz, 2H, ArC*H*), 2.93-2.67 (m, 4H, CH_2), 1.81-1.62 (m, 4H, CH_2), 1.44-1.28 (m, 2H, CH_2). $^{13}C\{^1H\}$ NMR (100.6 MHz, $CDCl_3$): δ 166.1, 164.5, 163.6, 130.0, 129.6, 129.5, 115.8, 115.6, 57.3, 25.3, 23.2. (ESI) m/z calcd for $C_{12}H_{15}FN_2O$ $(M+H)^+$: 223.1241, found: 223.1235.

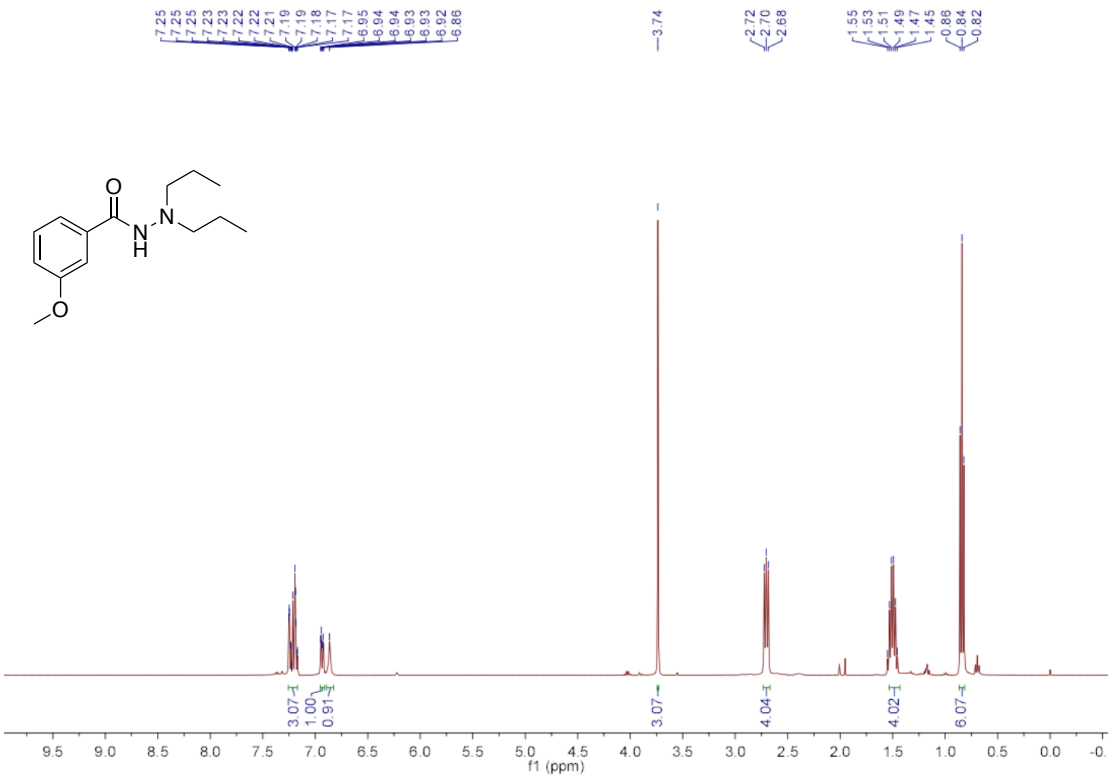
***N*-(Azepan-1-yl)benzamide (4h):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 71 mg, 65%. IR

(DCM): 3266, 3046, 2950, 2856, 1640, 1402, 1276, 1049, 892, 745 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.66 (d, *J* = 7.3 Hz, 2H, ArC*H*), 7.41 (t, *J* = 6.9 Hz, 1H, ArC*H*), 7.33 (t, *J* = 7.4 Hz, 2H, ArC*H*), 3.11-3.08 (m, 4H, CH_2), 1.76-1.67 (m, 4H, CH_2), 1.57-1.52 (m, 4H, CH_2). $^{13}C\{^{1}H\}$ NMR (100.6 MHz, $CDCl_3$): δ 165.7, 134.1, 131.5, 128.6, 127.0, 58.2, 27.1, 26.2. (ESI) m/z calcd for $C_{13}H_{18}N_2O$ $(M+H)^+$: 219.1492, found: 219.1510.

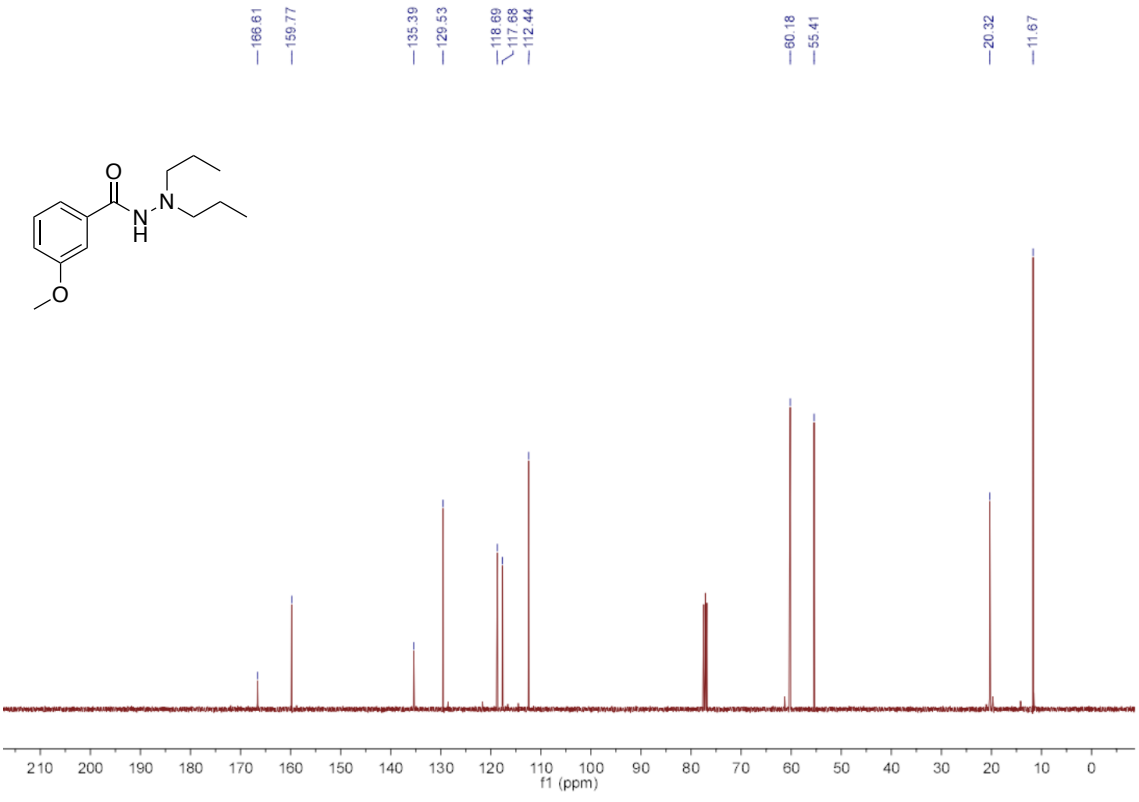
***N*-(Azepan-1-yl)-2-methylbenzamide (4i):** Purified by silica gel column chromatography using ethyl acetate/hexane (40:60) mixture as an eluent. White solid. Yield: 80 mg, 70%. IR (DCM): 3266, 3076, 2966, 2887, 1640, 1449, 1267, 890, 743 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.22 (dd, J_1 = 13.4 Hz, J_2 = 6.4 Hz, 2H, ArC*H*), 7.12 (dd, J_1 = 13.6 Hz, J_1 = 6.5 Hz, 2H, ArC*H*), 6.81 (s, 1H, N*H*), 3.14-3.05 (m, 4H, CH_2), 2.36 (s, 3H, CH_3), 1.76-1.60 (m, 4H, CH_2), 1.60-1.54 (m, 4H, CH_2). $^{13}C\{^{1}H\}$ NMR (100.6 MHz, $CDCl_3$): δ 167.8, 136.3, 135.3, 130.9, 130.0, 126.9, 125.7, 58.4, 27.0, 26.1, 19.6. (ESI) m/z calcd for $C_{14}H_{20}N_2O$ $(M+H)^+$: 233.1648, found: 233.1640.

^{1}H and ^{13}C NMR Spectra of *N,N*-Dialkyl Acylhydrazides:

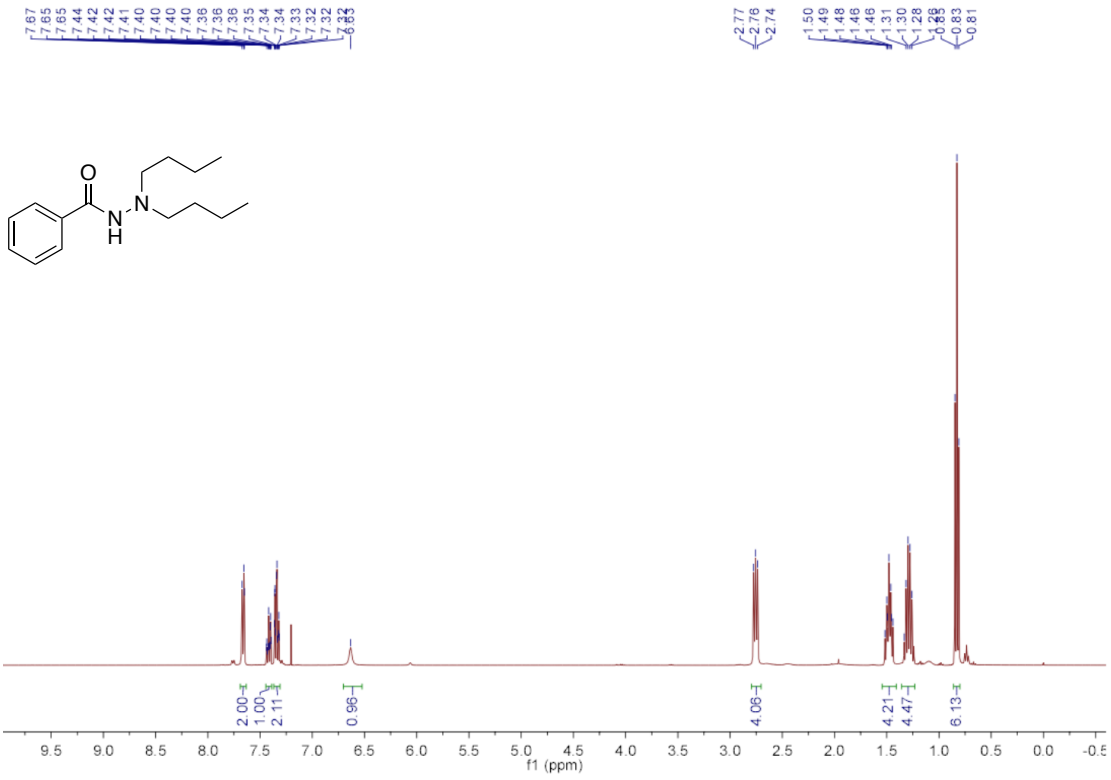
^{1}H NMR spectrum of 3-Methoxy-*N*',*N*'-dipropylbenzohydrazide **(2a):** (400 MHz, $CDCl_3$)



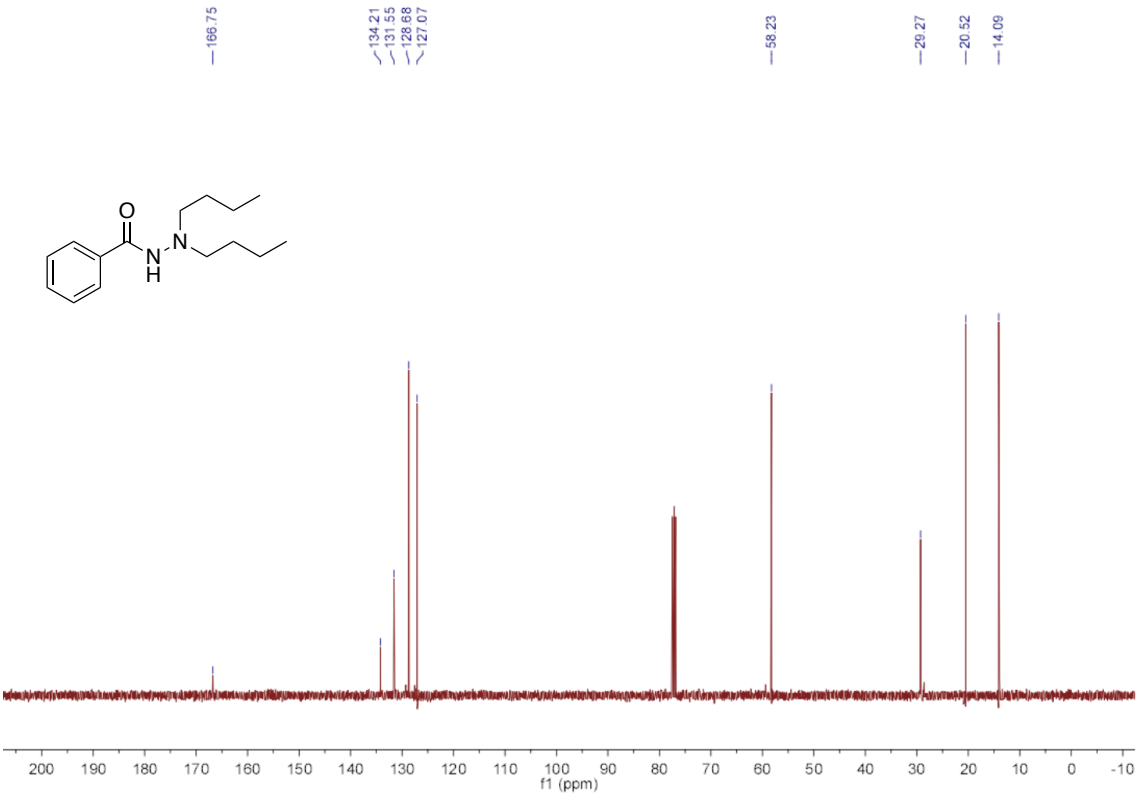
^{13}C NMR spectrum of 3-Methoxy-*N*',*N*'-dipropylbenzohydrazide **(2a):** (100.6 MHz, $CDCl_3$)



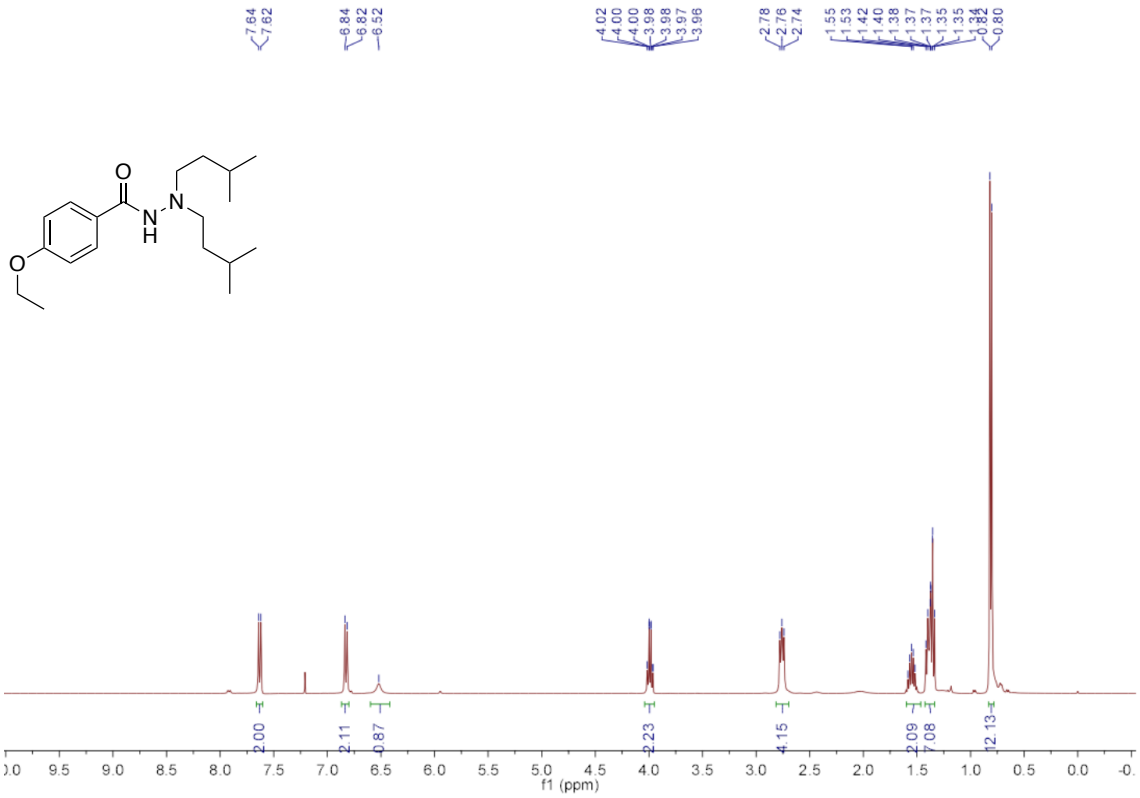
^{1}H NMR spectrum of *N*',*N*'-Dibutylbenzohydrazide **(2b):** (400 MHz, $CDCl_3$)



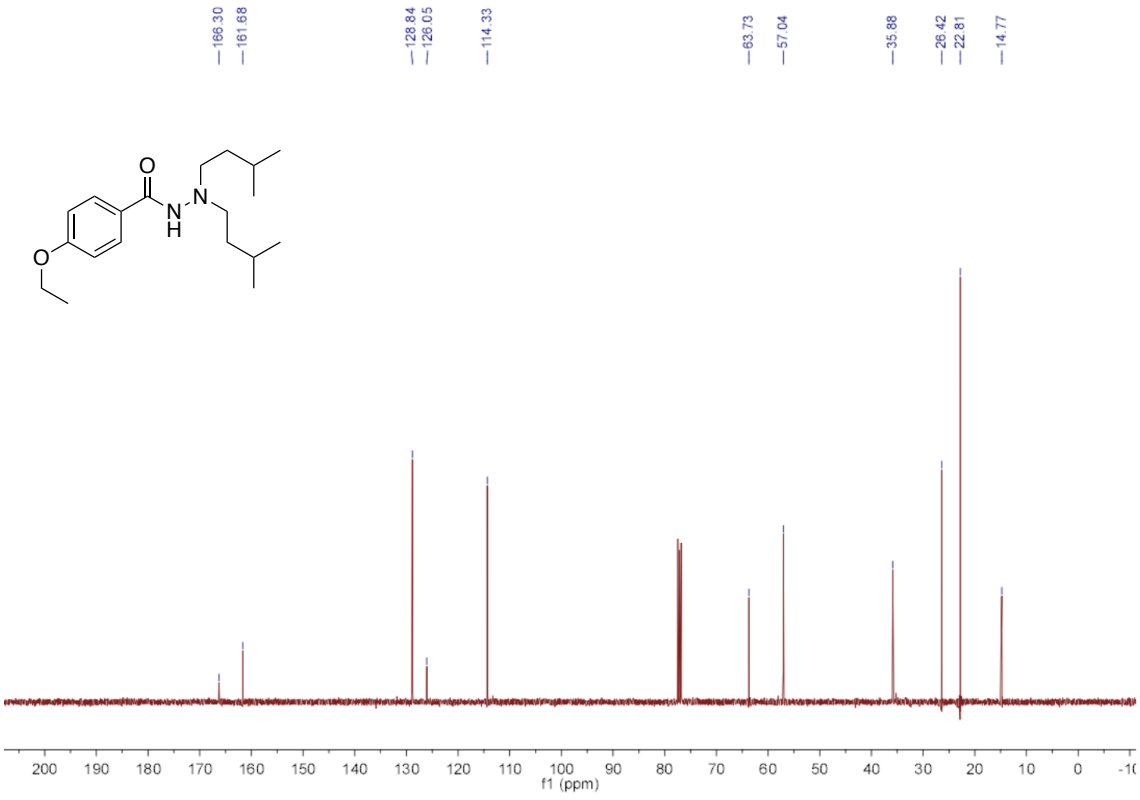
^{13}C NMR spectrum of *N*',*N*'-Dibutylbenzohydrazide **(2b):** (100.6 MHz, $CDCl_3$)



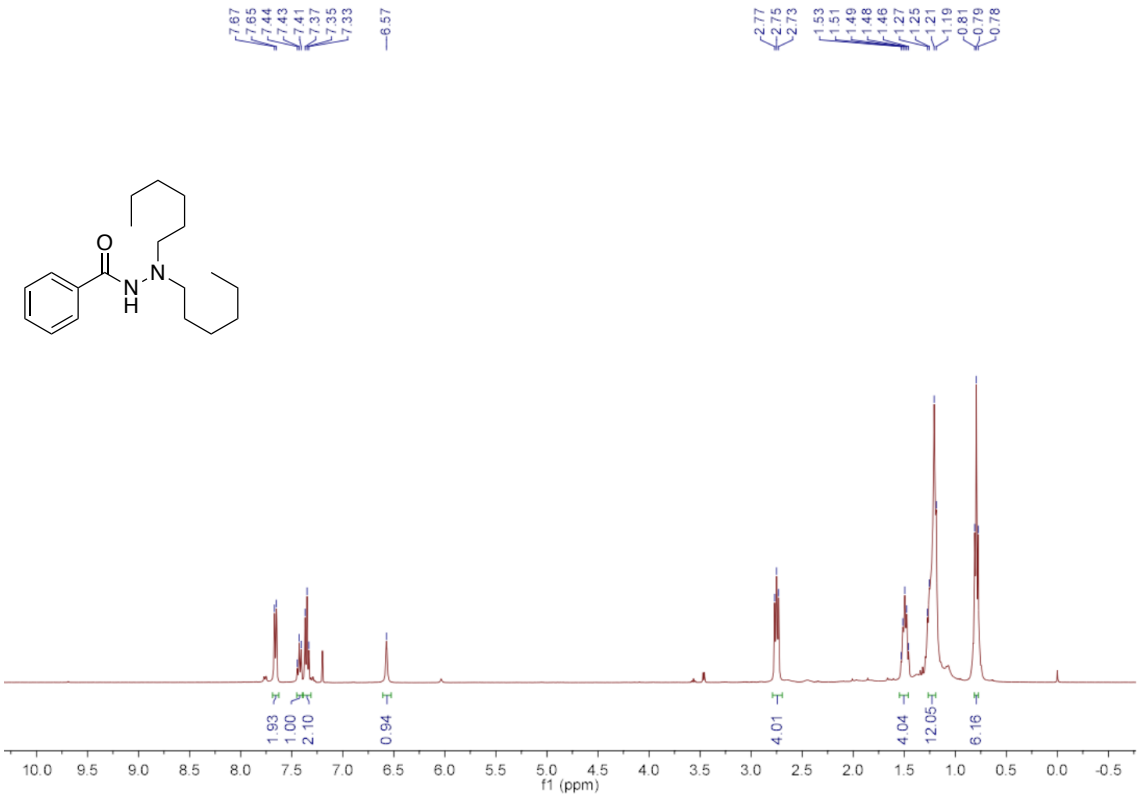
^{1}H NMR spectrum of 4-ethoxy-*N*',*N*'-diisopentylbenzohydrazide **(2c):** (400 MHz, $CDCl_3$)



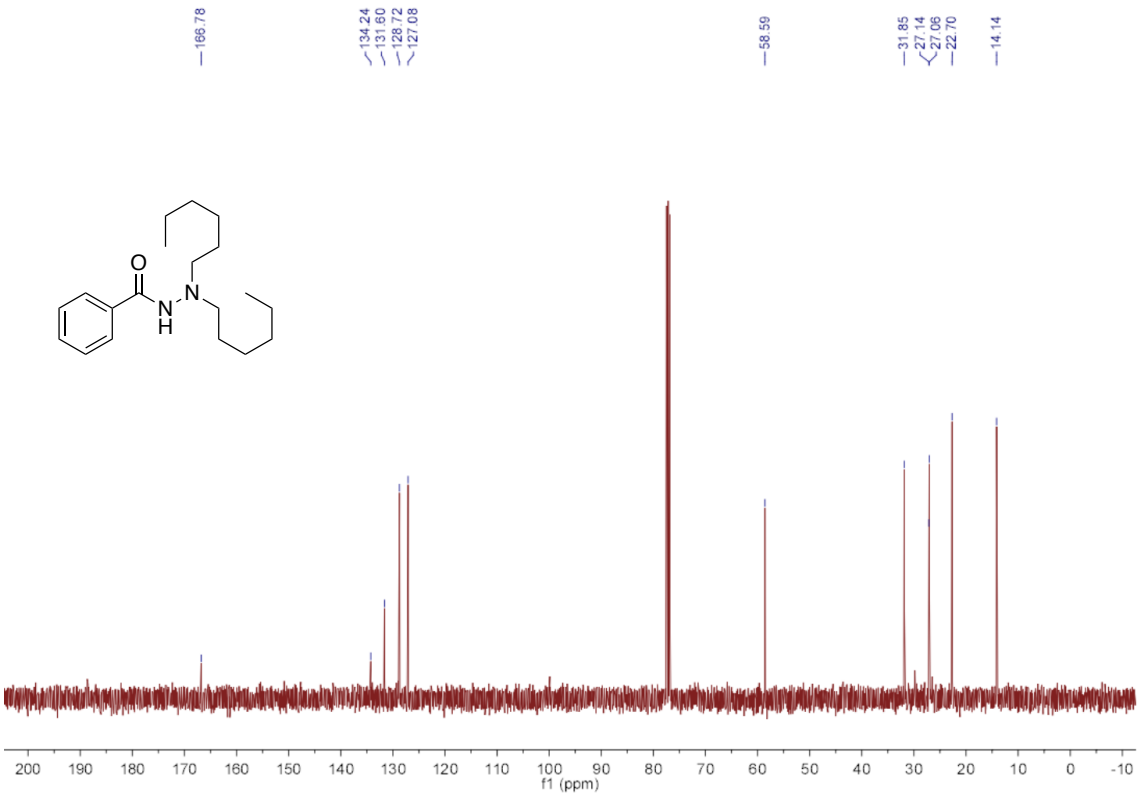
^{13}C NMR spectrum of 4-ethoxy-*N*',*N*'-diisopentylbenzohydrazide **(2c):** (100.6 MHz, $CDCl_3$)



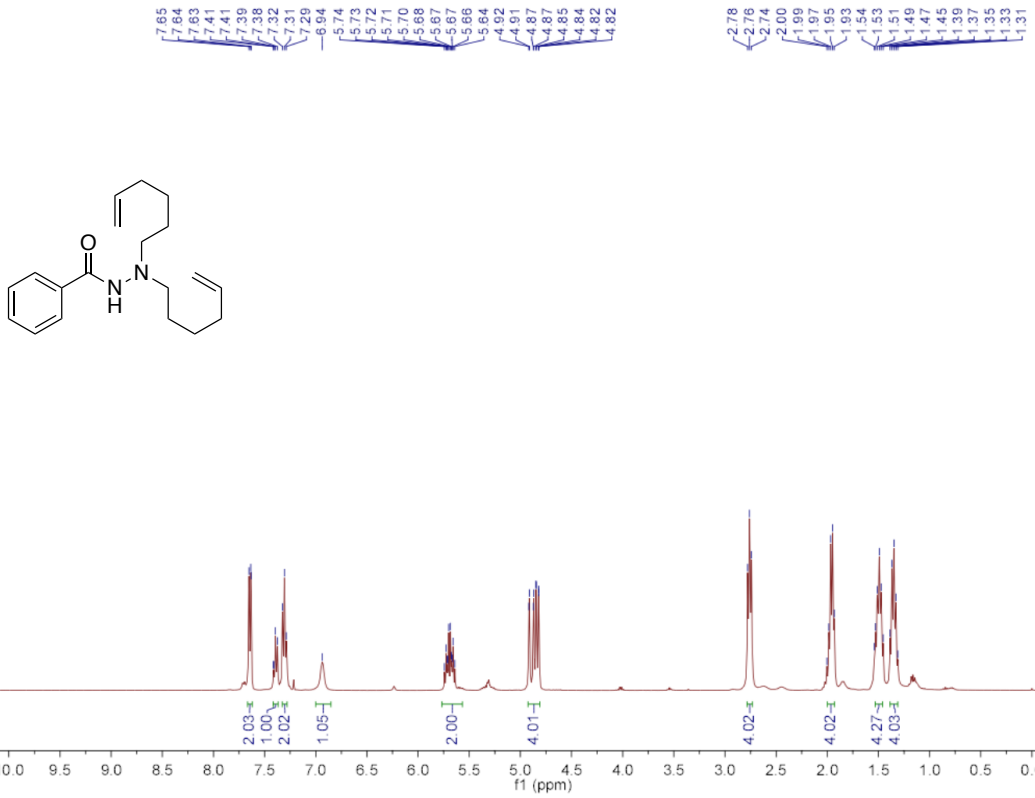
^{1}H NMR spectrum of *N*',*N*'-Dihexylbenzohydrazide **(2d):** (400 MHz, $CDCl_3$)



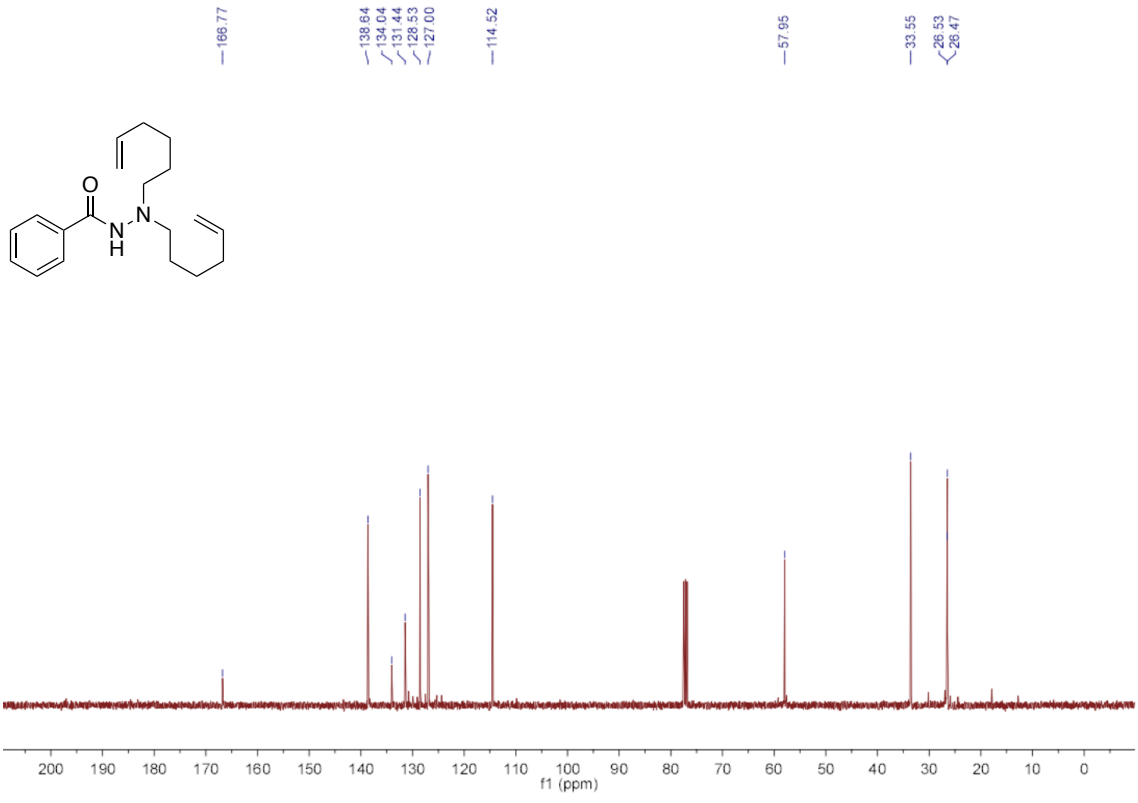
^{13}C NMR spectrum of *N*',*N*'-Dihexylbenzohydrazide **(2d):** (100.6 MHz, $CDCl_3$)



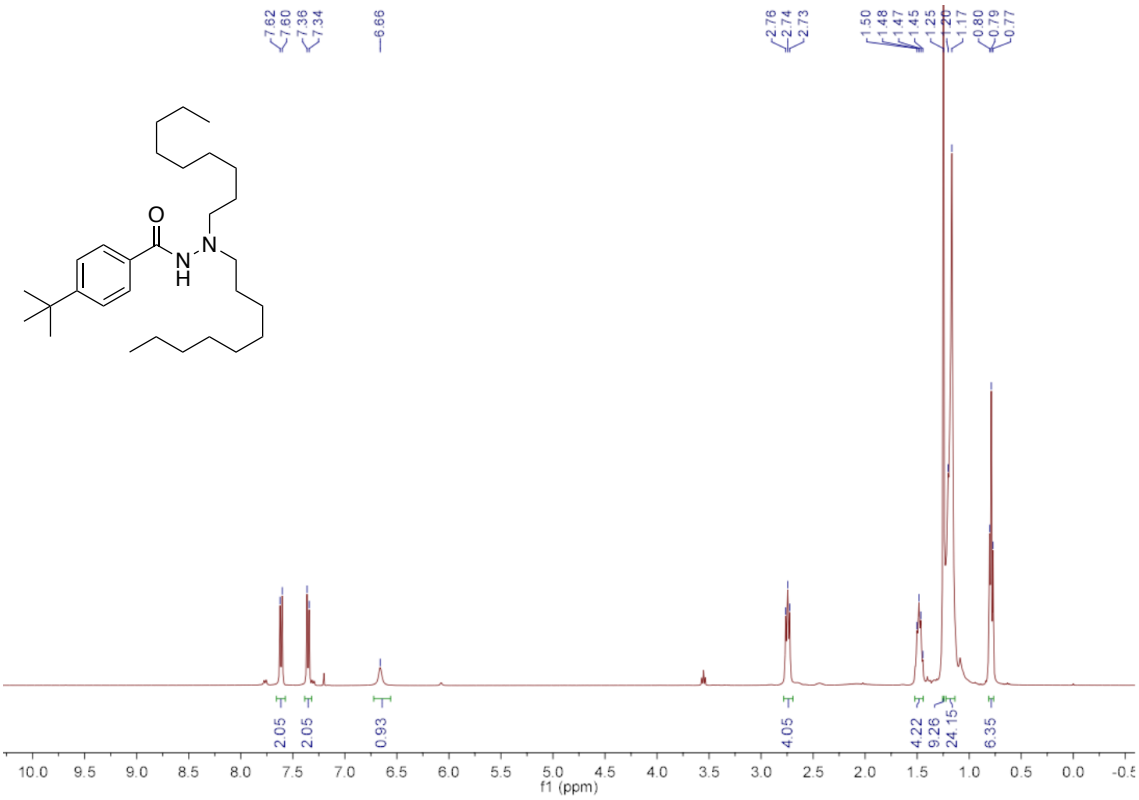
^{1}H NMR spectrum of *N*',*N*'-Di(hex-5-en-1-yl)benzohydrazide **(2e):** (400 MHz, $CDCl_3$)



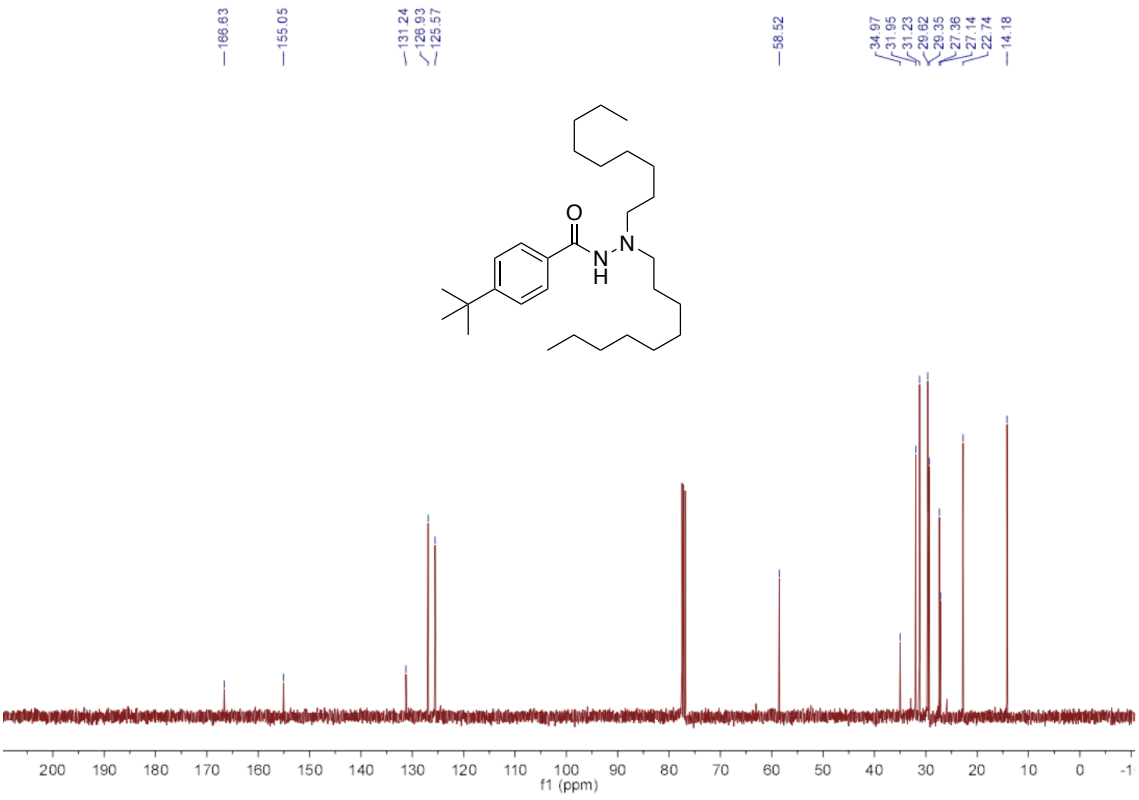
^{13}C NMR spectrum of *N*',*N*'-Di(hex-5-en-1-yl)benzohydrazide **(2e):** (100.6 MHz, $CDCl_3$)



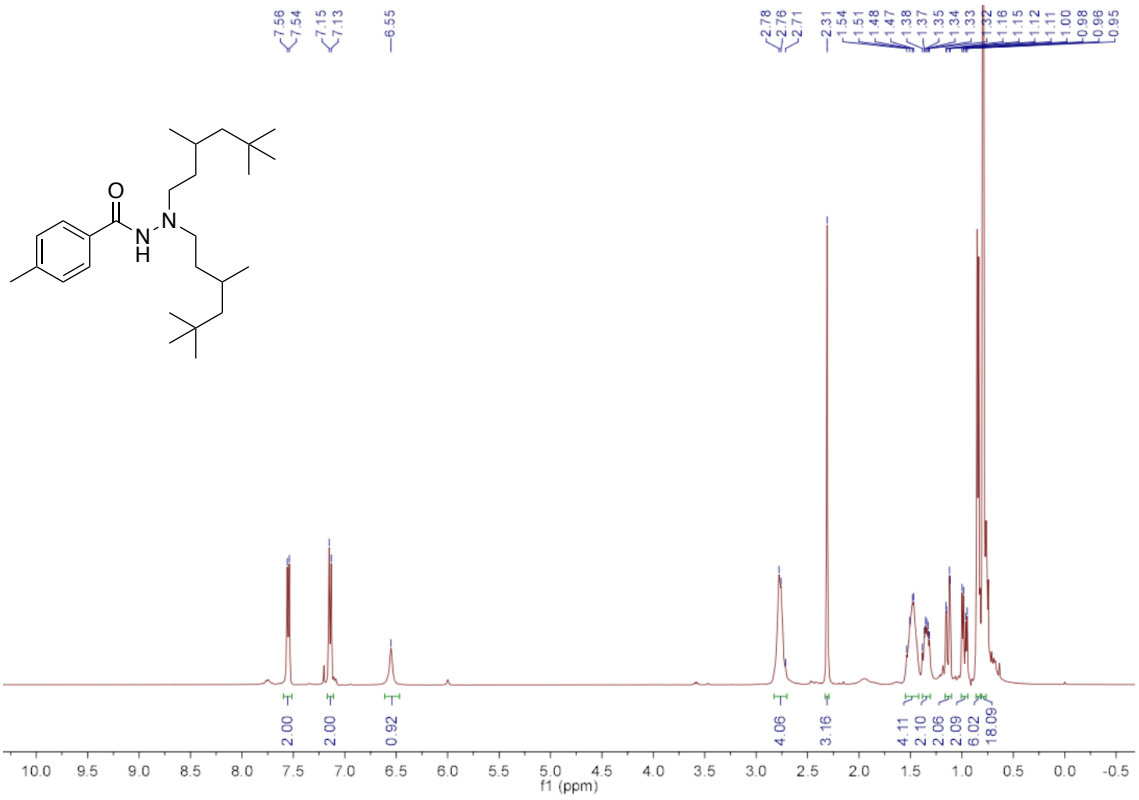
^{1}H NMR spectrum of 4-(*tert*-Butyl)-*N*',*N*'-dinonylbenzohydrazide **(2f):** (400 MHz, $CDCl_3$)



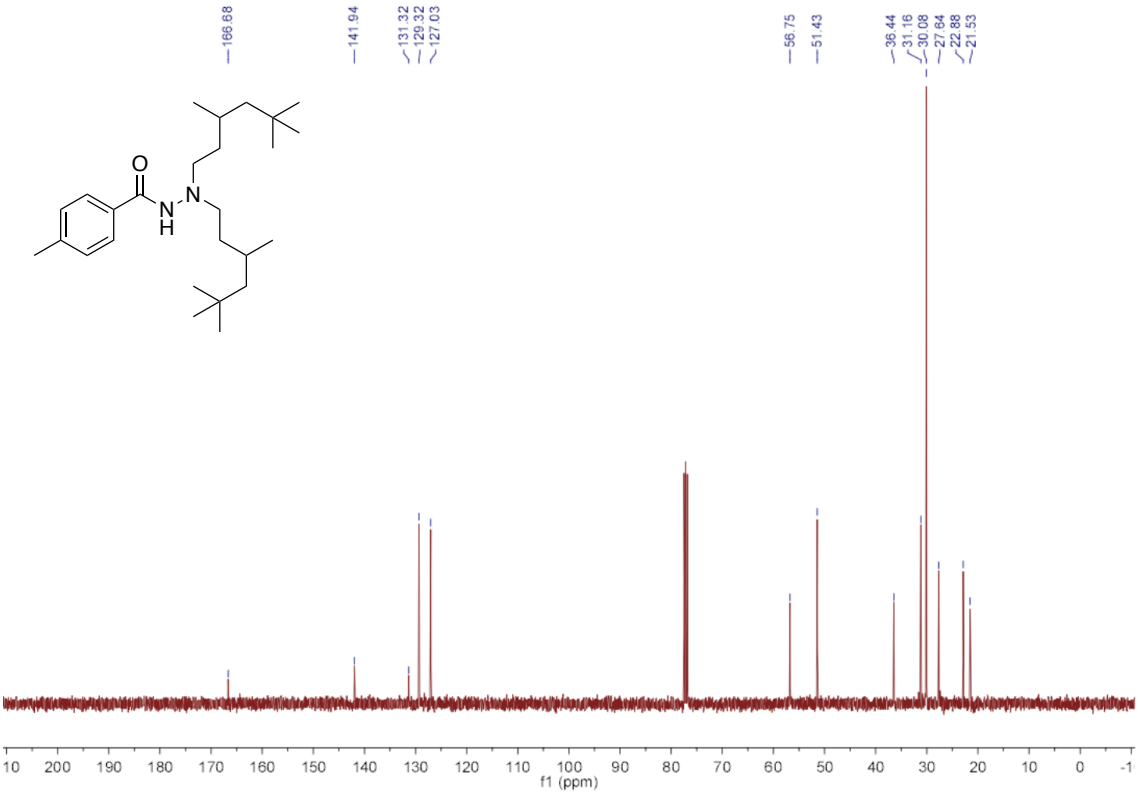
^{13}C NMR spectrum of 4-(*tert*-Butyl)-*N*',*N*'-dinonylbenzohydrazide **(2f):** (100.6 MHz, $CDCl_3$)



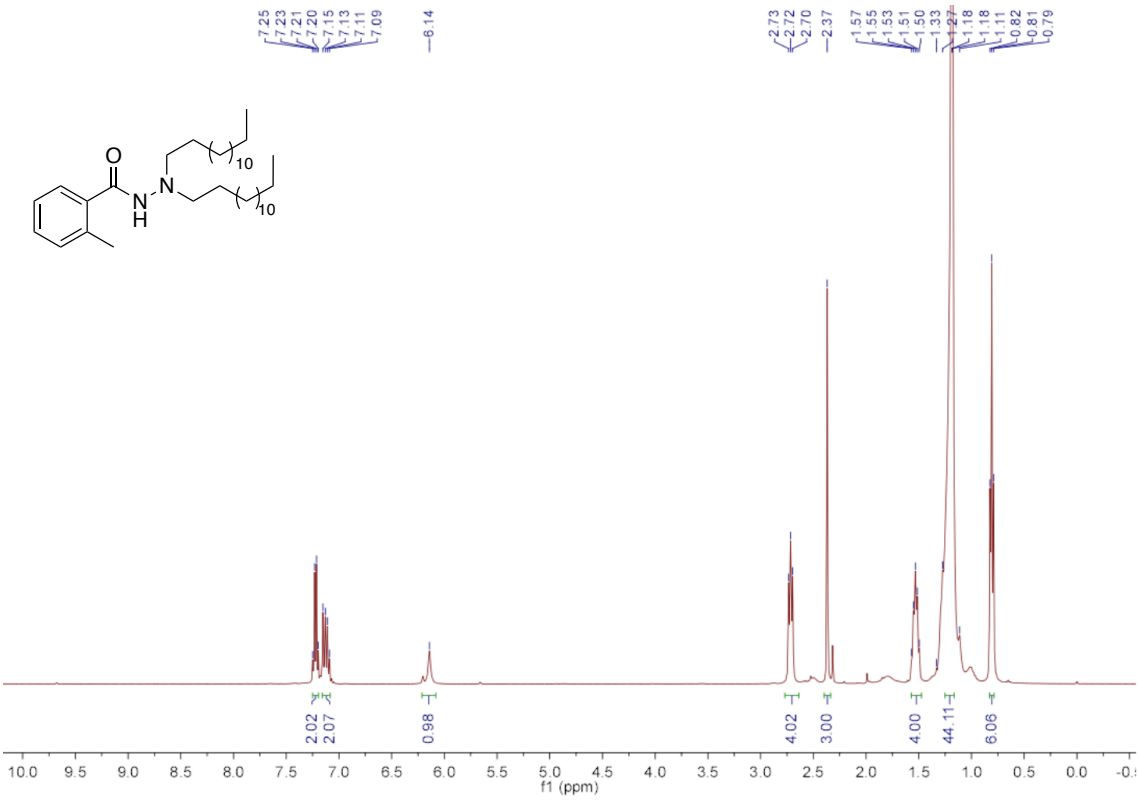
^{1}H NMR spectrum of 4-methyl-*N*',*N*'-bis(3,5,5-trimethylhexyl)benzohydrazide **(2g):** (400 MHz, $CDCl_3$)



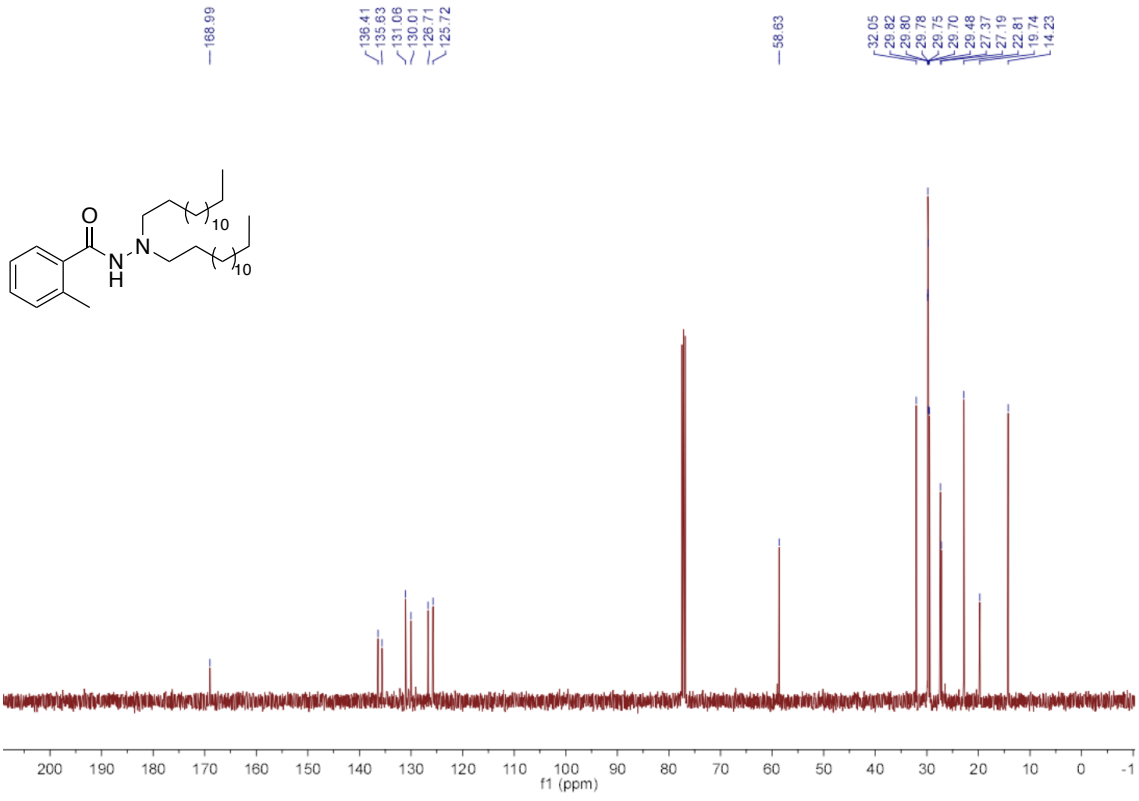
^{13}C NMR spectrum of 4-methyl-*N*',*N*'-bis(3,5,5-trimethylhexyl)benzohydrazide **(2g):** (100.6 MHz, $CDCl_3$)



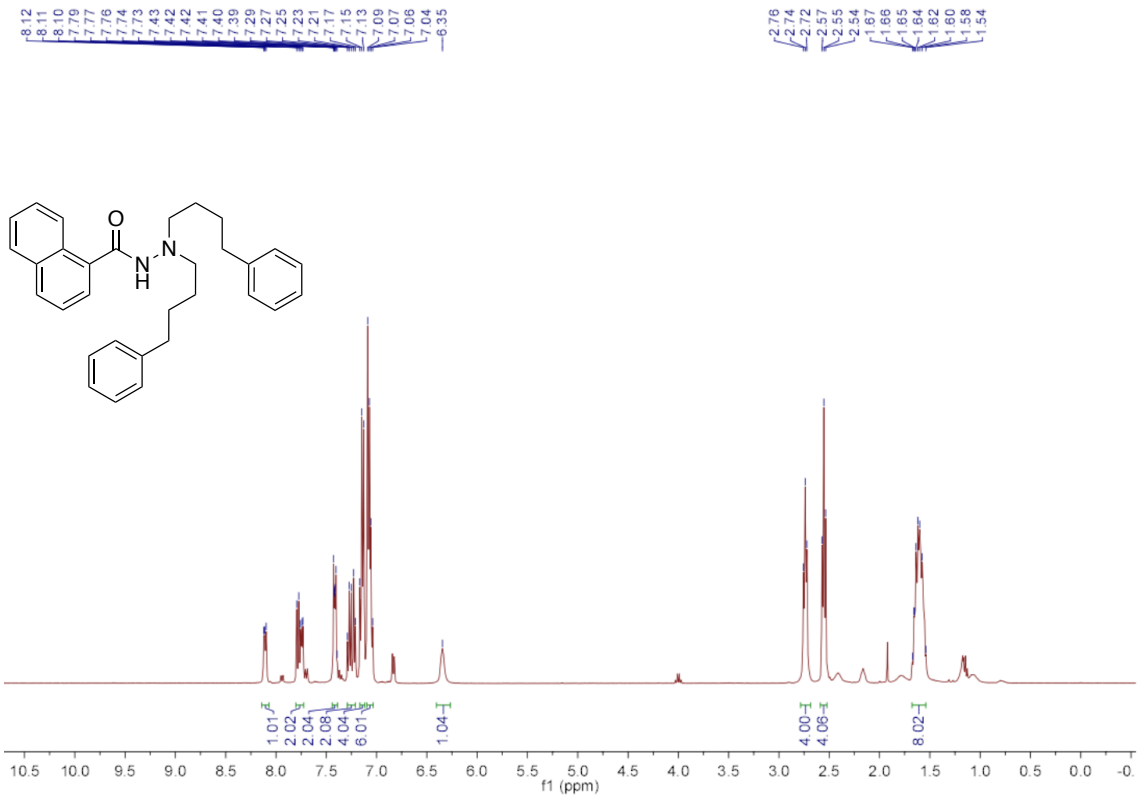
^{1}H NMR spectrum of 2-Methyl-*N*',*N*'-ditetradecylbenzohydrazide **(2h):** (400 MHz, $CDCl_3$)



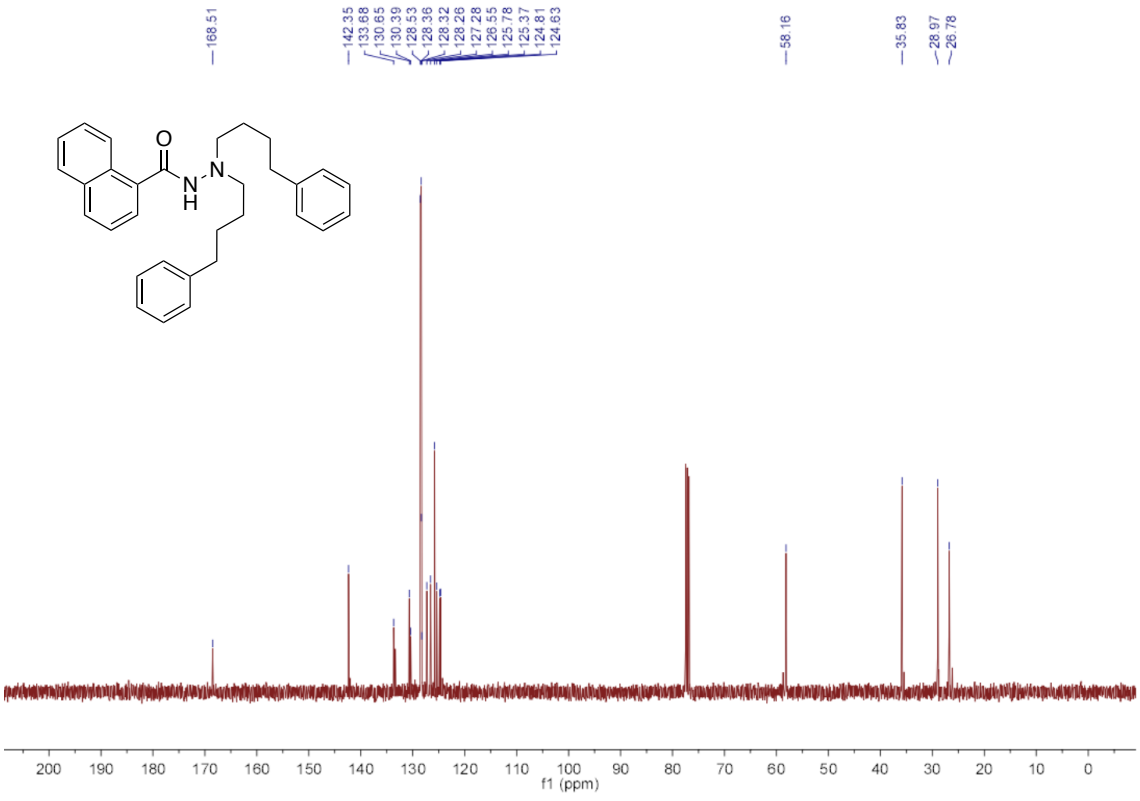
^{13}C NMR spectrum of 2-Methyl-*N*',*N*'-ditetradecylbenzohydrazide **(2h):** (100.6 MHz, $CDCl_3$)



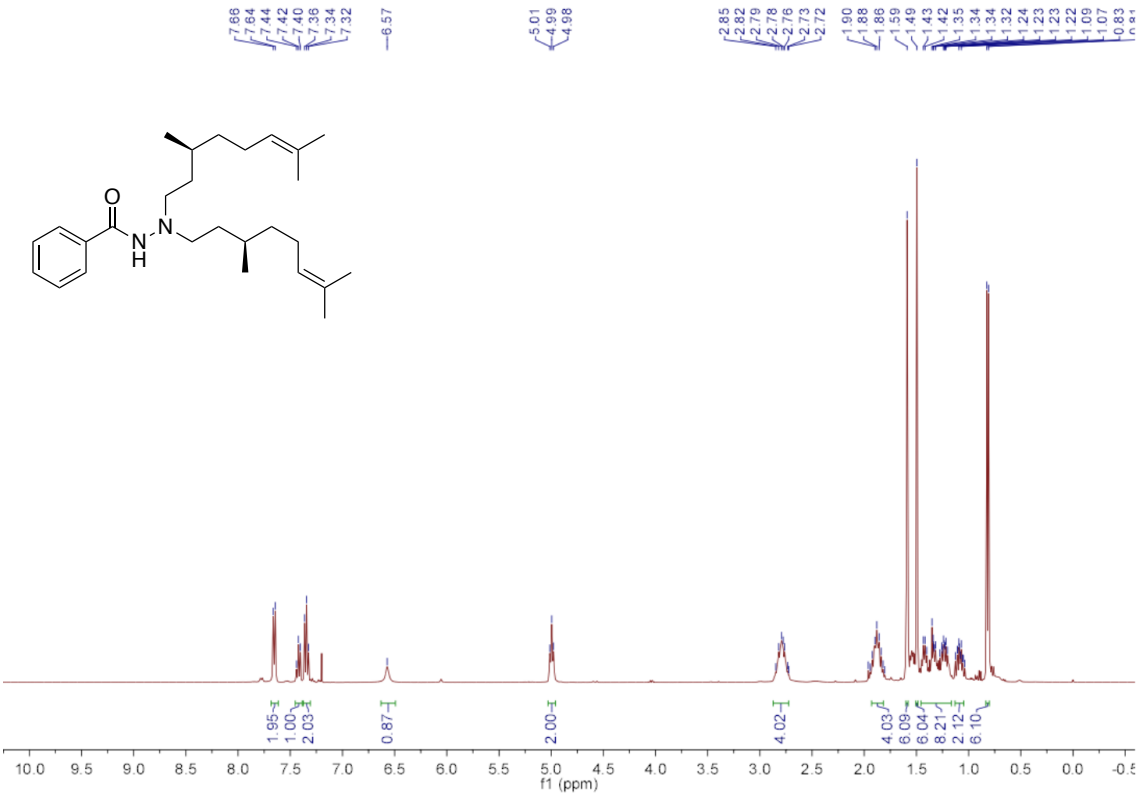
^{1}H NMR spectrum of *N*',*N*'-bis(4-Phenylbutyl)-1-naphthohydrazide **(2i):** (400 MHz, $CDCl_3$)



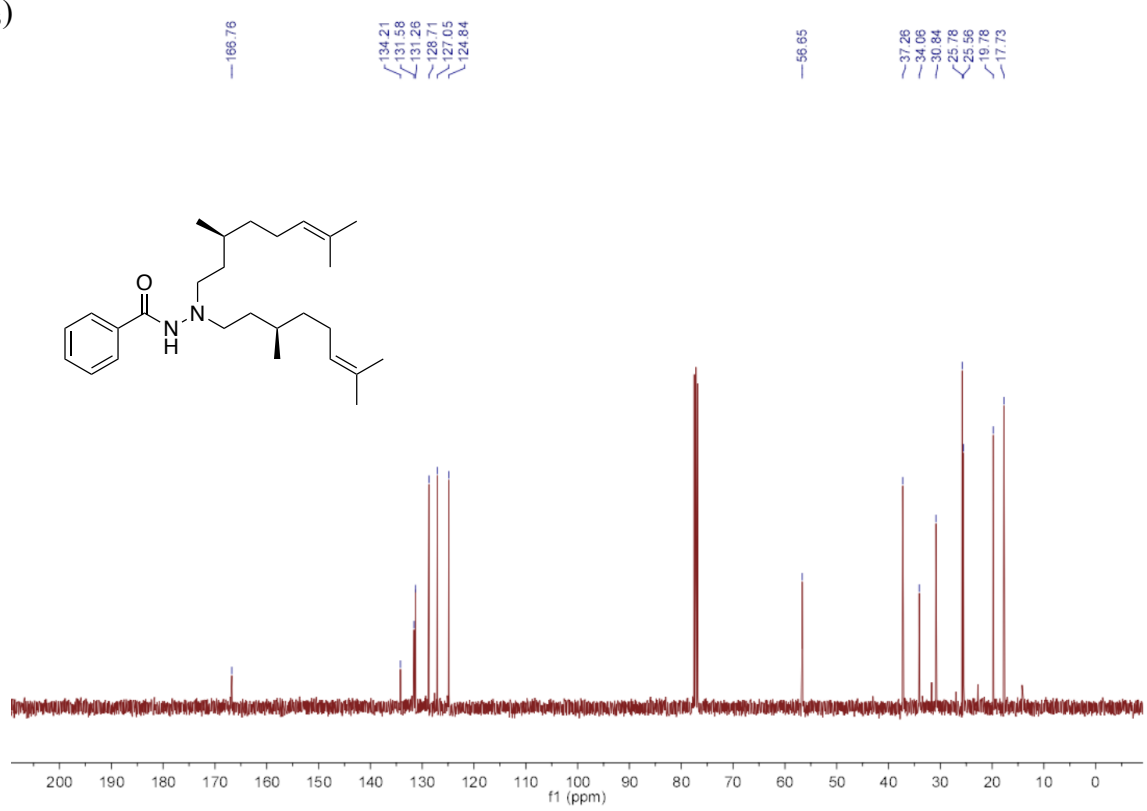
^{13}C NMR spectrum of *N*',*N*'-bis(4-Phenylbutyl)-1-naphthohydrazide **(2i):** (100.6 MHz, $CDCl_3$)



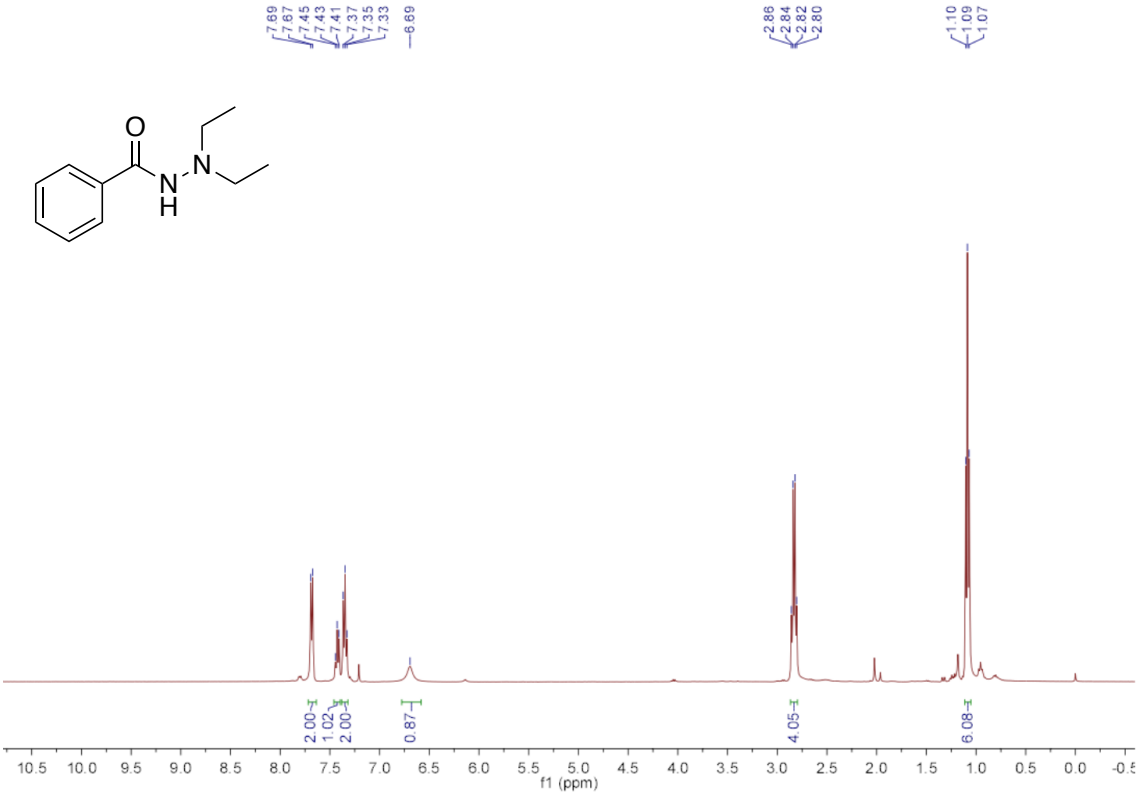
^{1}H NMR spectrum of *N*',*N*'-Bis(3,7-dimethyloct-6-en-1-yl)benzohydrazide **(2j):** (400 MHz, $CDCl_3$)



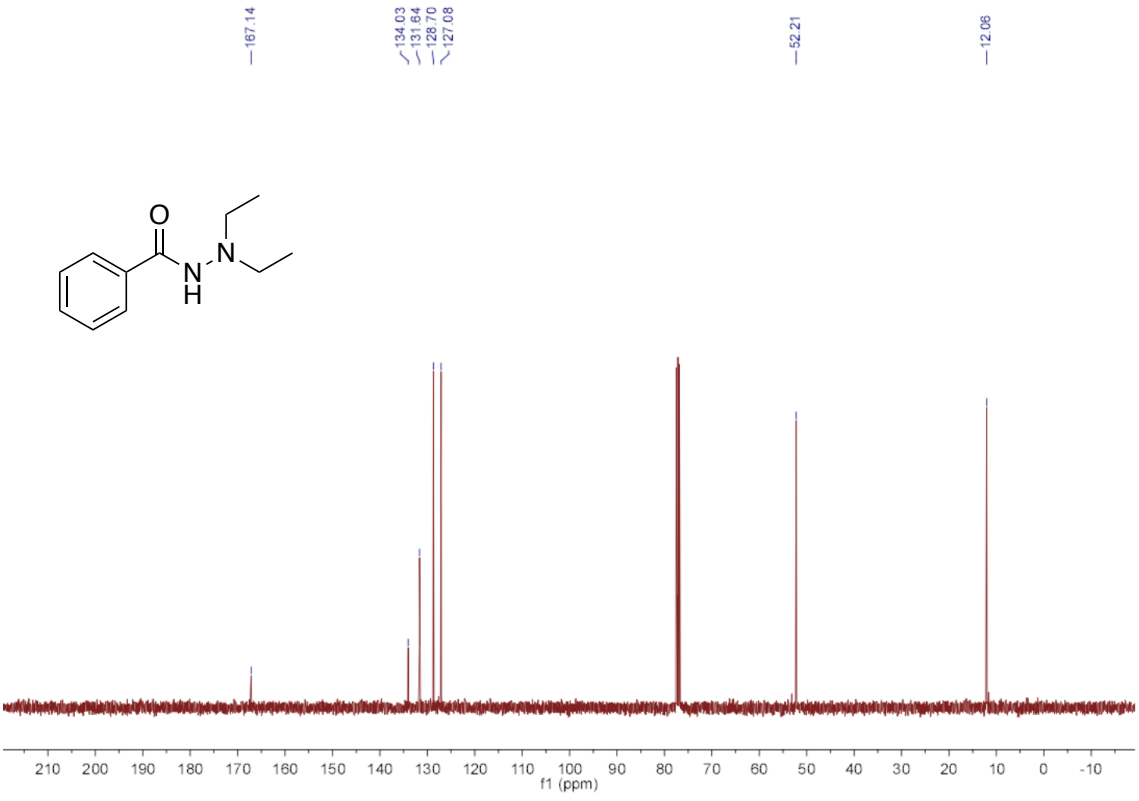
^{13}C NMR spectrum of *N*',*N*'-Bis(3,7-dimethyloct-6-en-1-yl)benzohydrazide **(2j):** (100.6 MHz, $CDCl_3$)



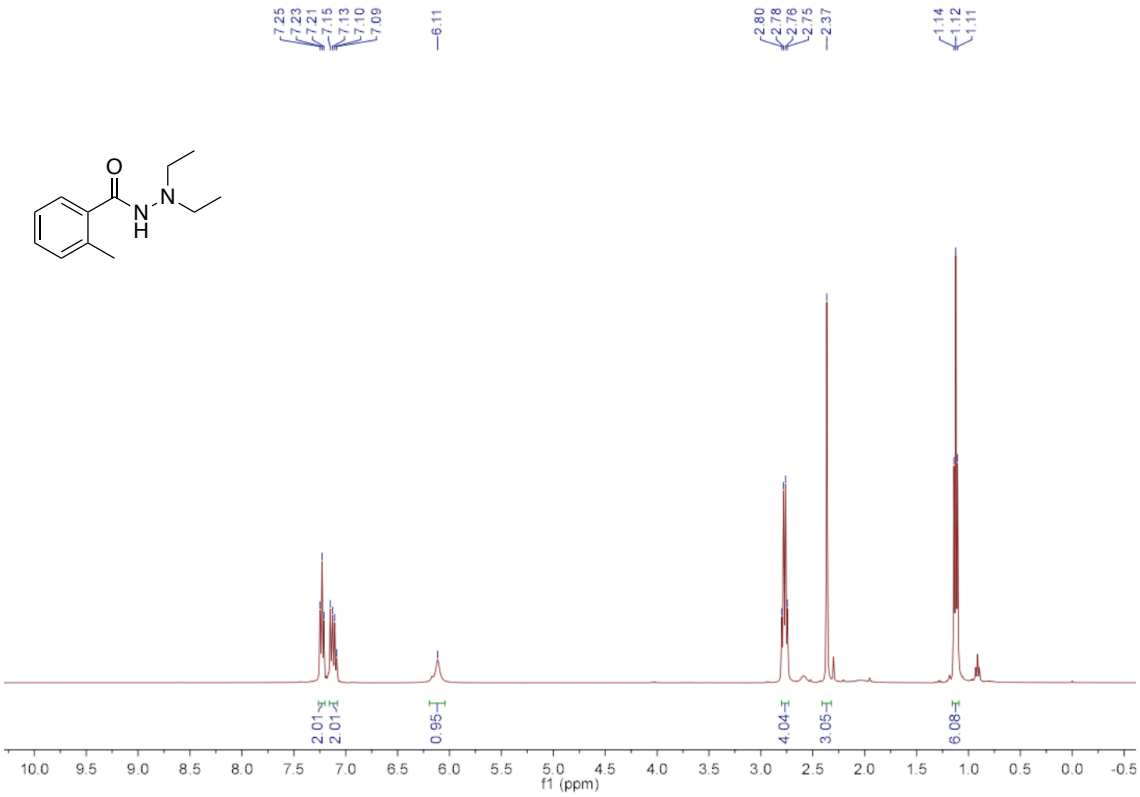
^{1}H NMR spectrum of *N*',*N*'-Diethylbenzohydrazide **(2k):** (400 MHz, $CDCl_3$)



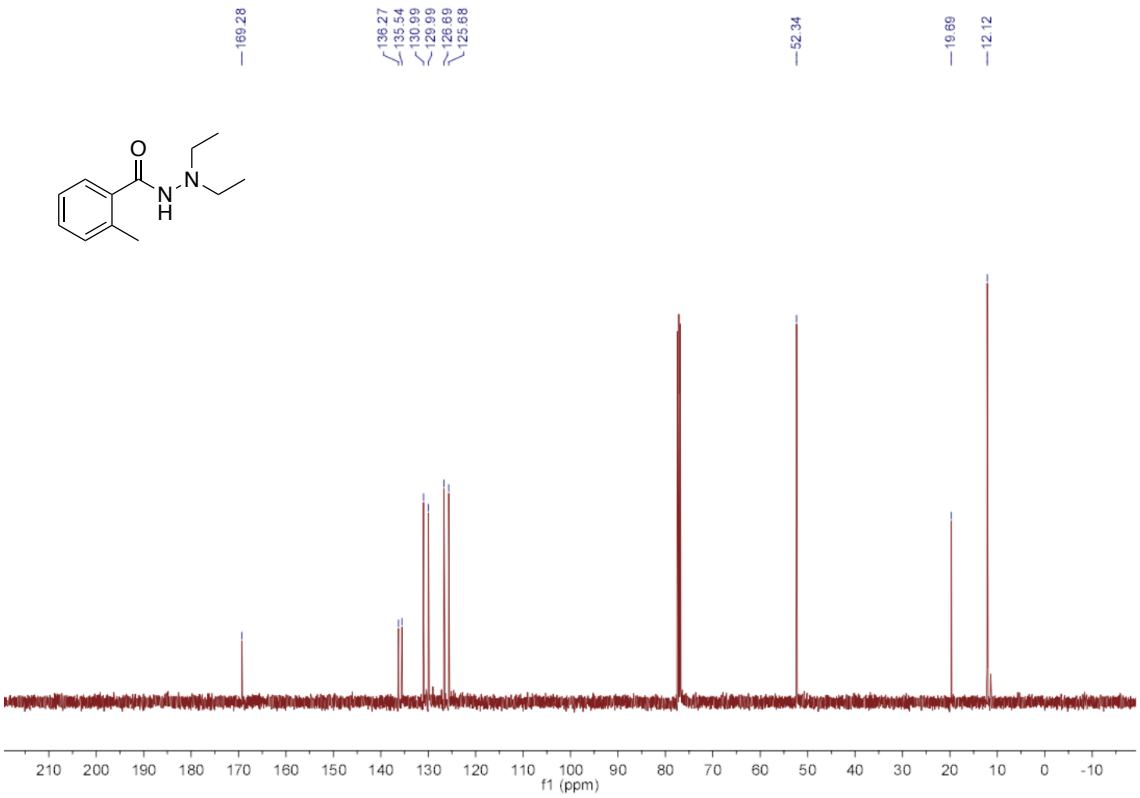
^{13}C NMR spectrum of *N*',*N*'-Diethylbenzohydrazide **(2k):** (100.6 MHz, $CDCl_3$)



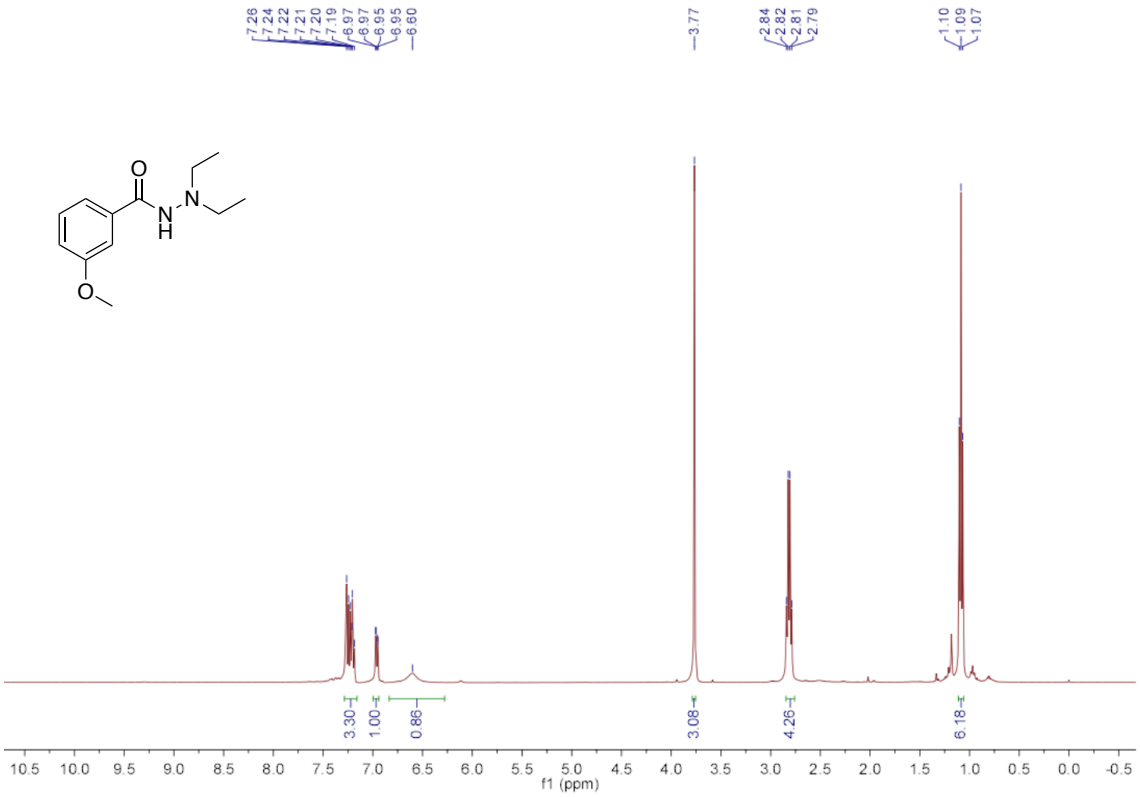
^{1}H NMR spectrum of *N*',*N*'-diethyl-2-methylbenzohydrazide **(2l):** (400 MHz, $CDCl_3$)



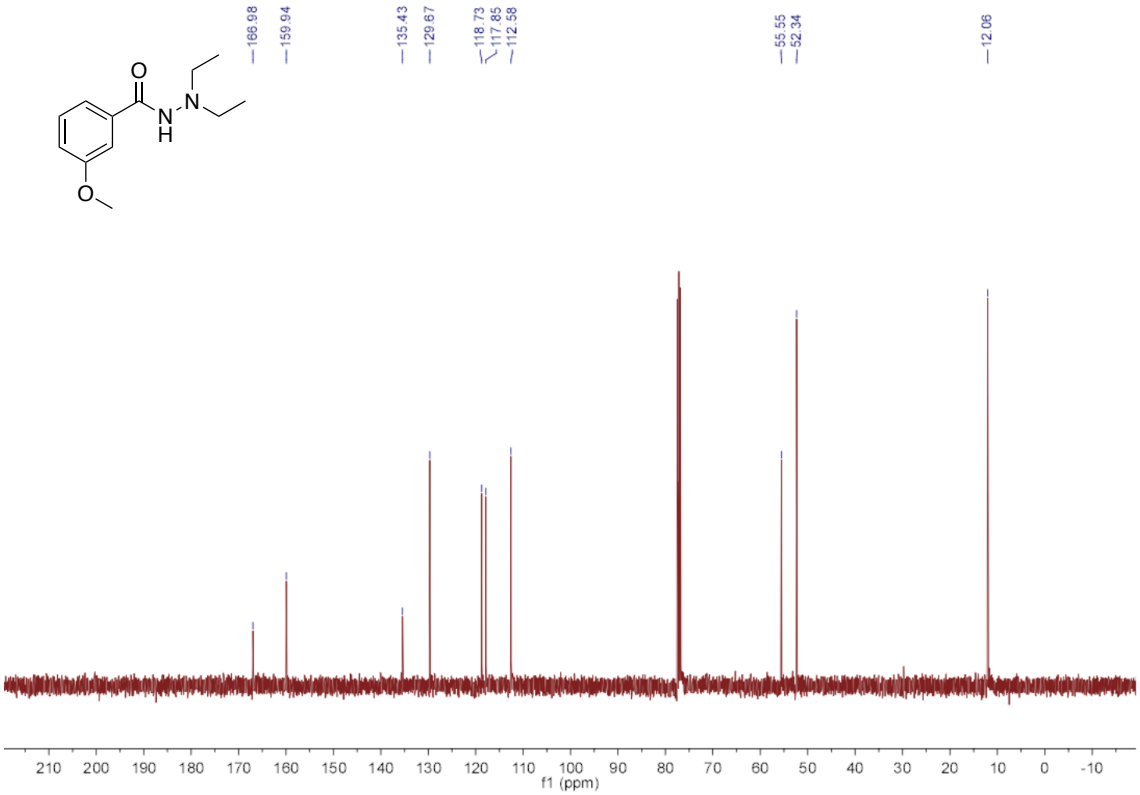
^{13}C NMR spectrum of *N*',*N*'-diethyl-2-methylbenzohydrazide **(2l):** (100.6 MHz, $CDCl_3$)



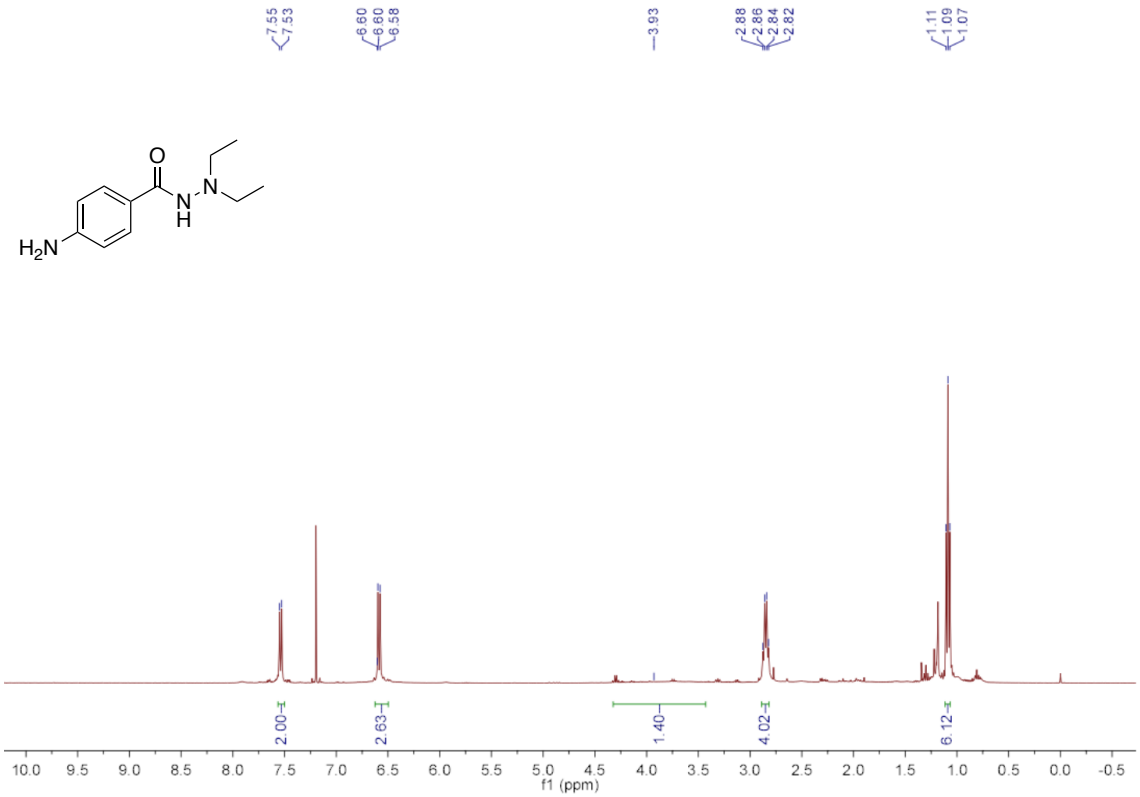
^{1}H NMR spectrum of *N*',*N*'-diethyl-3-methoxybenzohydrazide **(2m):** (400 MHz, $CDCl_3$)



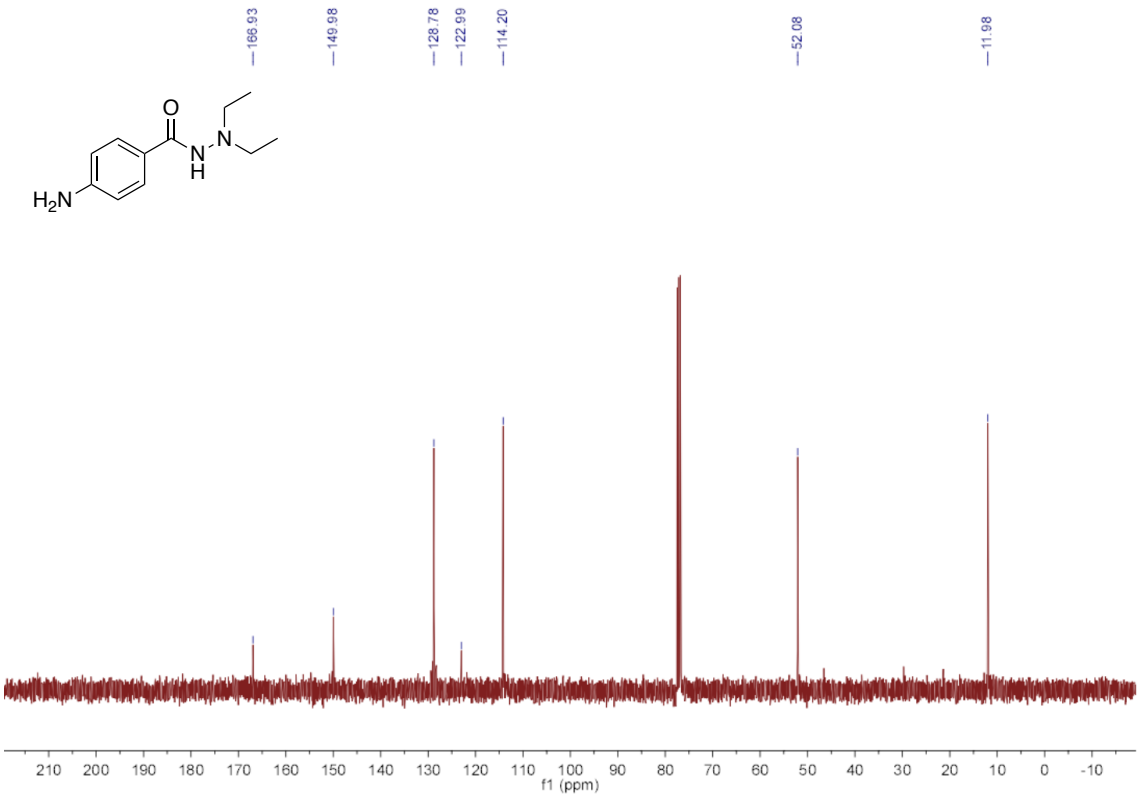
^{13}C NMR spectrum of *N*',*N*'-diethyl-3-methoxybenzohydrazide **(2m):** (100.6 MHz, $CDCl_3$)



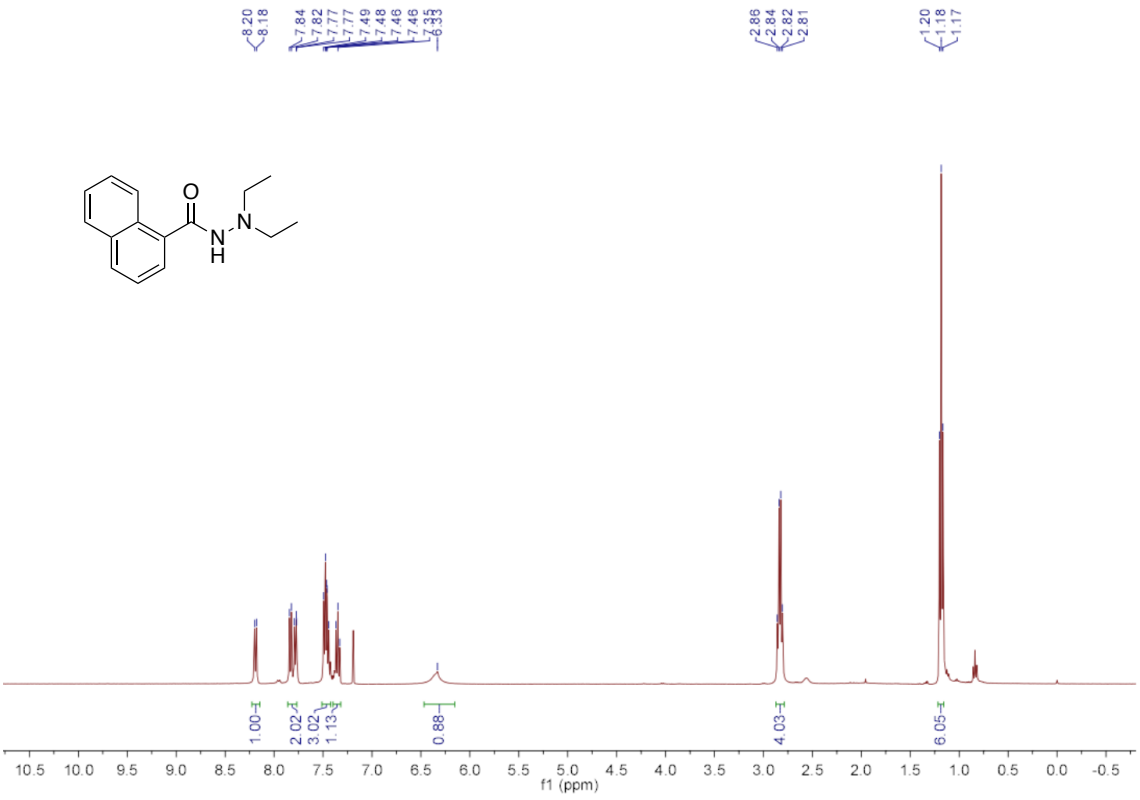
^{1}H NMR spectrum of 4-amino-*N*',*N*'-diethylbenzohydrazide **(2n):** (400 MHz, $CDCl_3$)



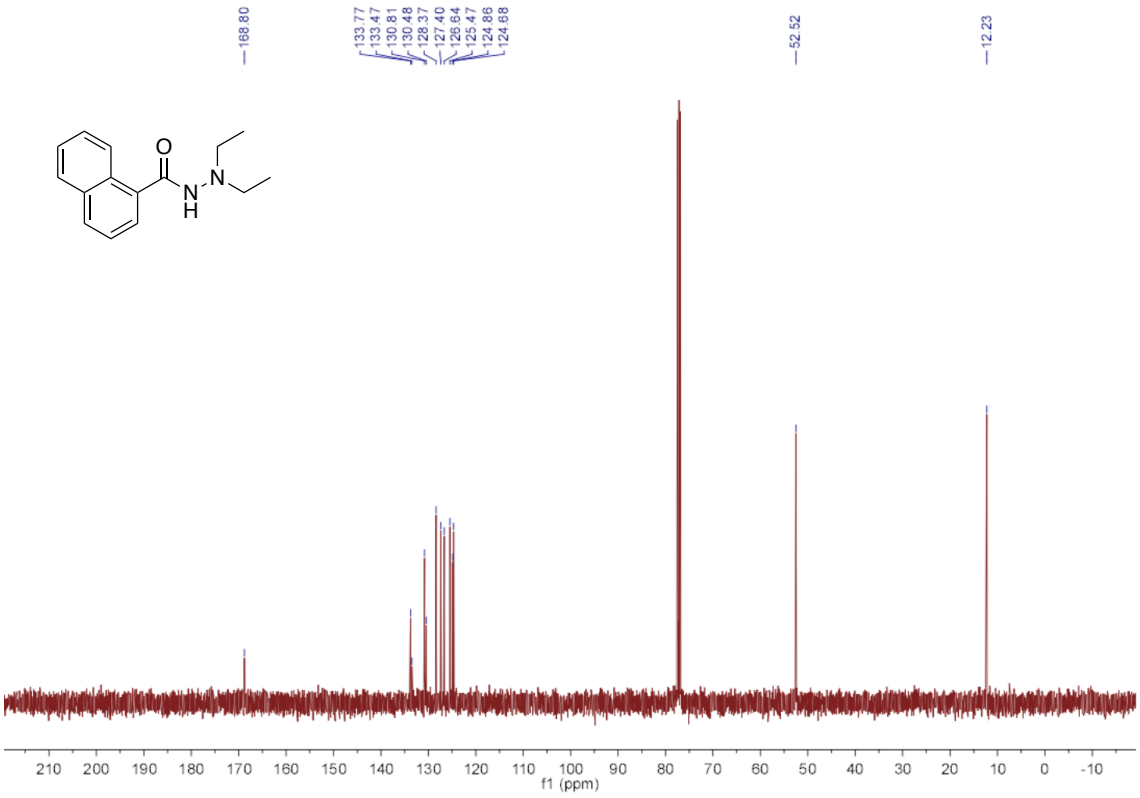
^{13}C NMR spectrum of 4-amino-*N*',*N*'-diethylbenzohydrazide **(2n):** (100.6 MHz, $CDCl_3$)



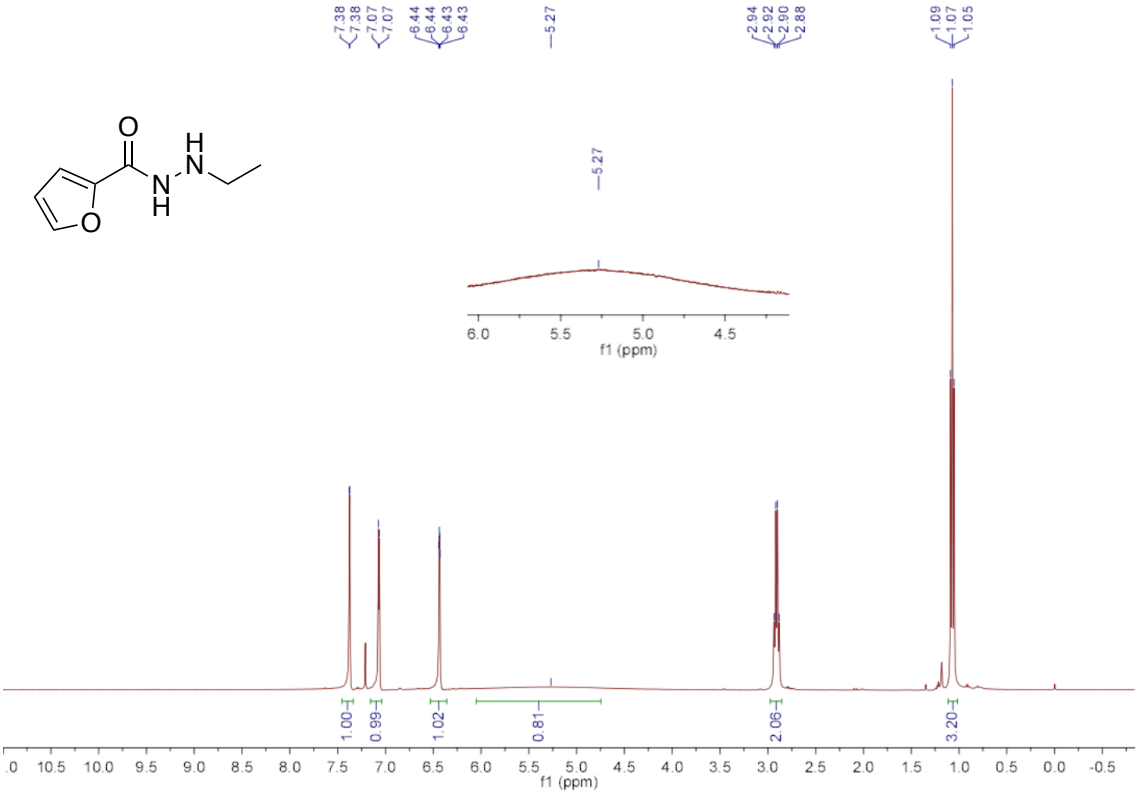
^{1}H NMR spectrum of *N*',*N*'-diethyl-1-naphthohydrazide **(2o):** (400 MHz, $CDCl_3$)



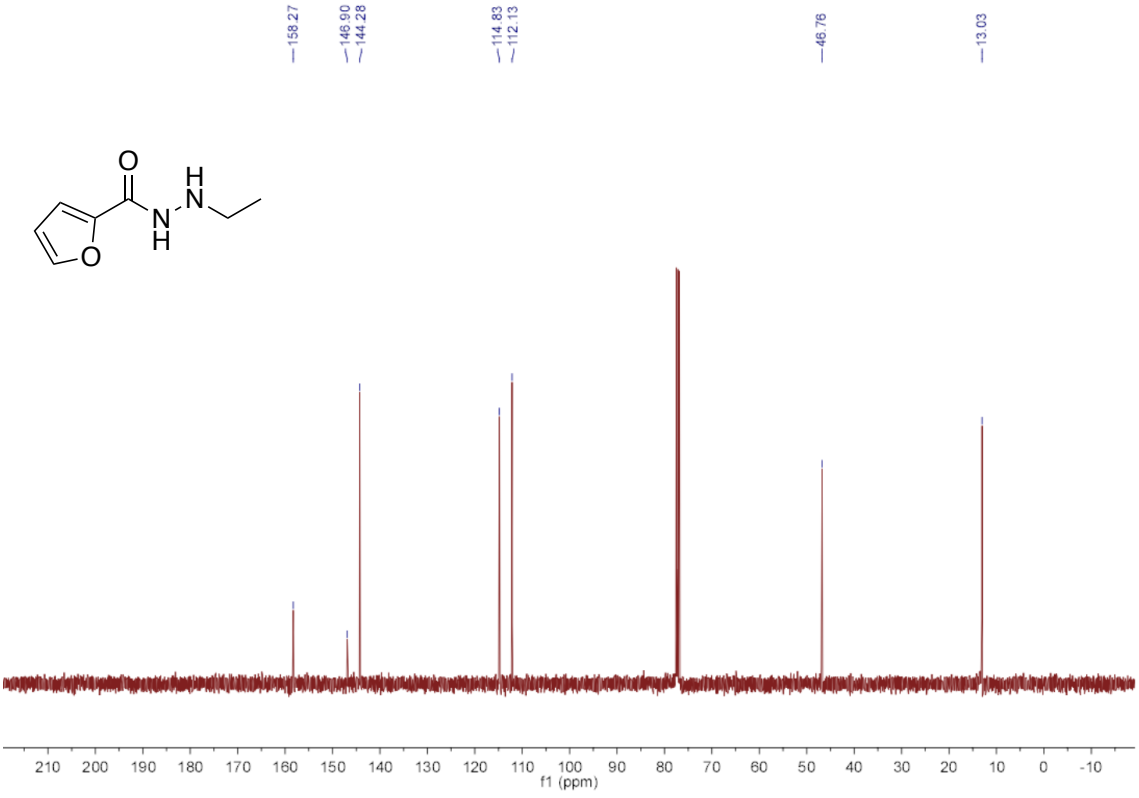
^{13}C NMR spectrum of *N*',*N*'-diethyl-1-naphthohydrazide **(2o):** (100.6 MHz, $CDCl_3$)



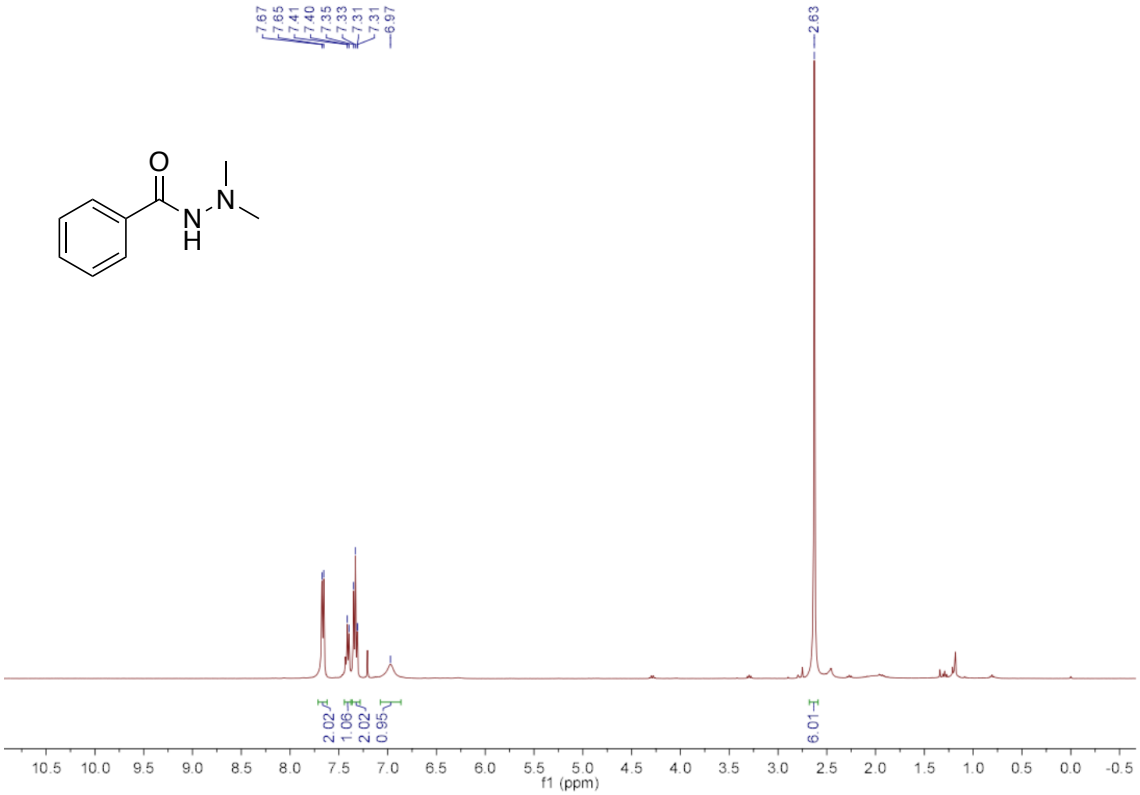
^{1}H NMR spectrum of *N*'-ethylfuran-2-carbohydrazide **(2p):** (400 MHz, $CDCl_3$)



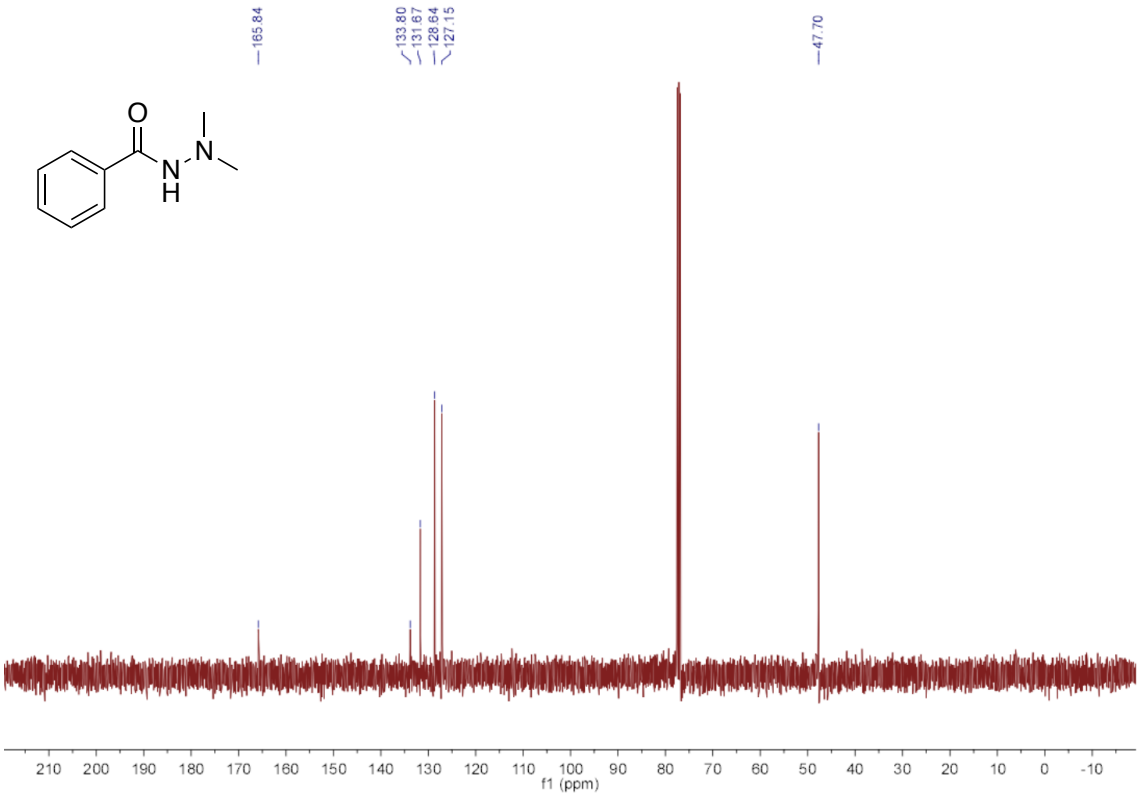
^{13}C NMR spectrum of *N*'-ethylfuran-2-carbohydrazide **(2p):** (100.6 MHz, $CDCl_3$)



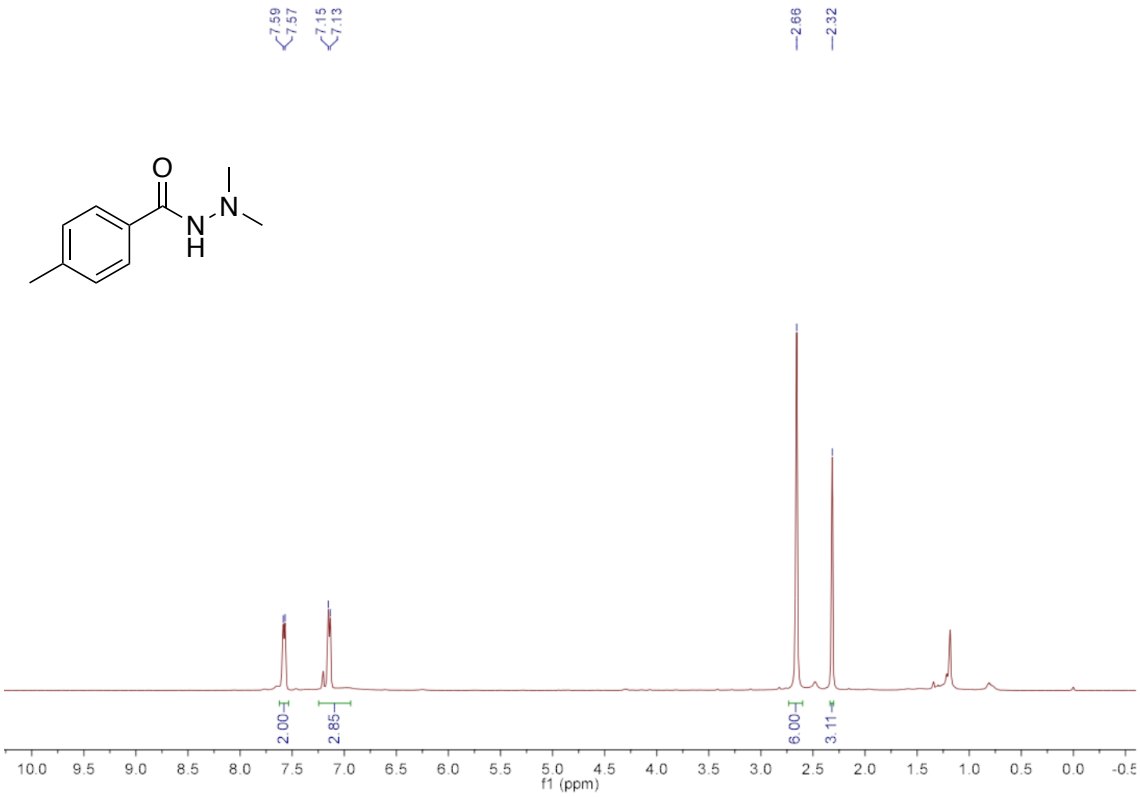
^{1}H NMR spectrum of *N*',*N*'-Dimethylbenzohydrazide **(2q):** (400 MHz, $CDCl_3$)



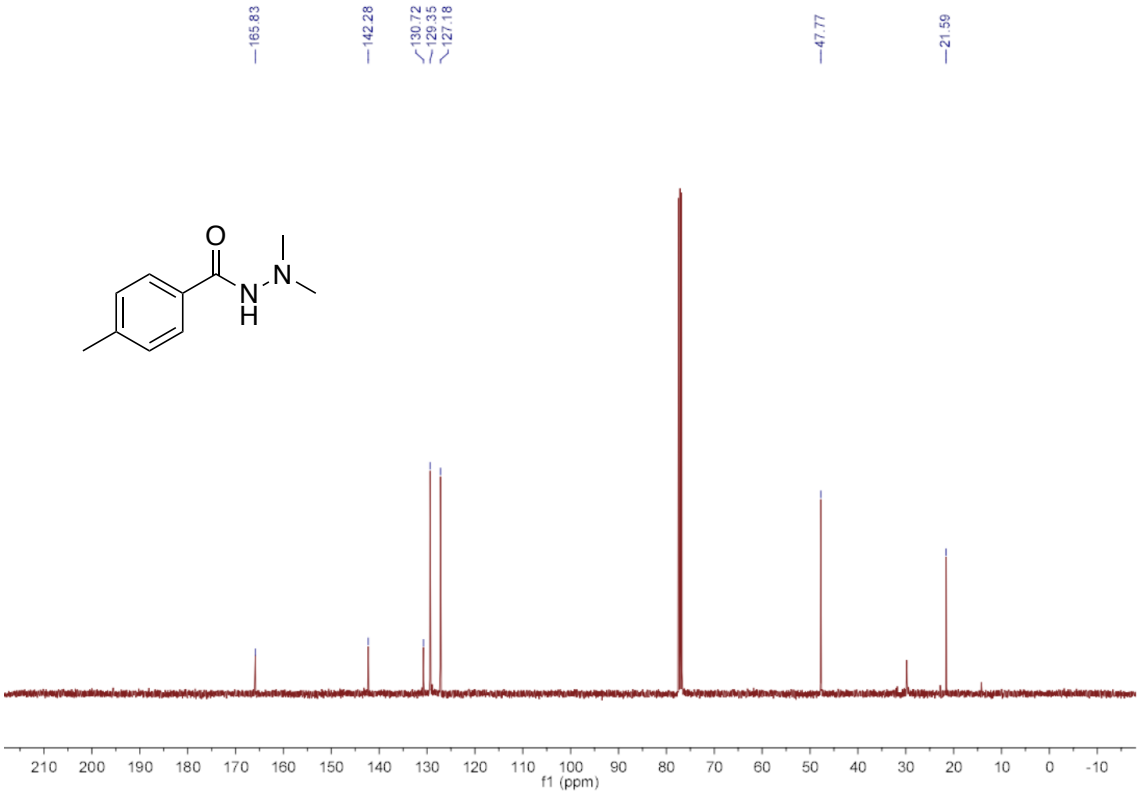
^{13}C NMR spectrum of *N*',*N*'-Dimethylbenzohydrazide **(2q):** (100.6 MHz, $CDCl_3$)



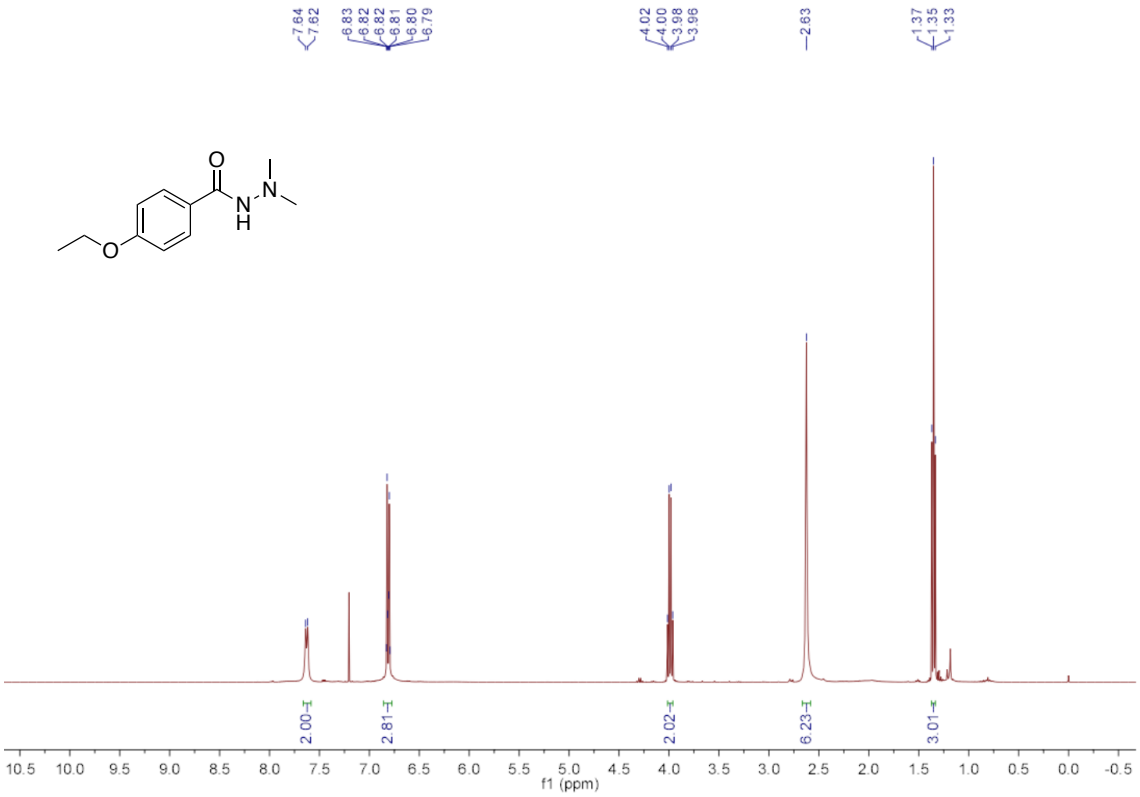
^{1}H NMR spectrum of *N*',*N*',4-trimethylbenzohydrazide **(2r):** (400 MHz, $CDCl_3$)



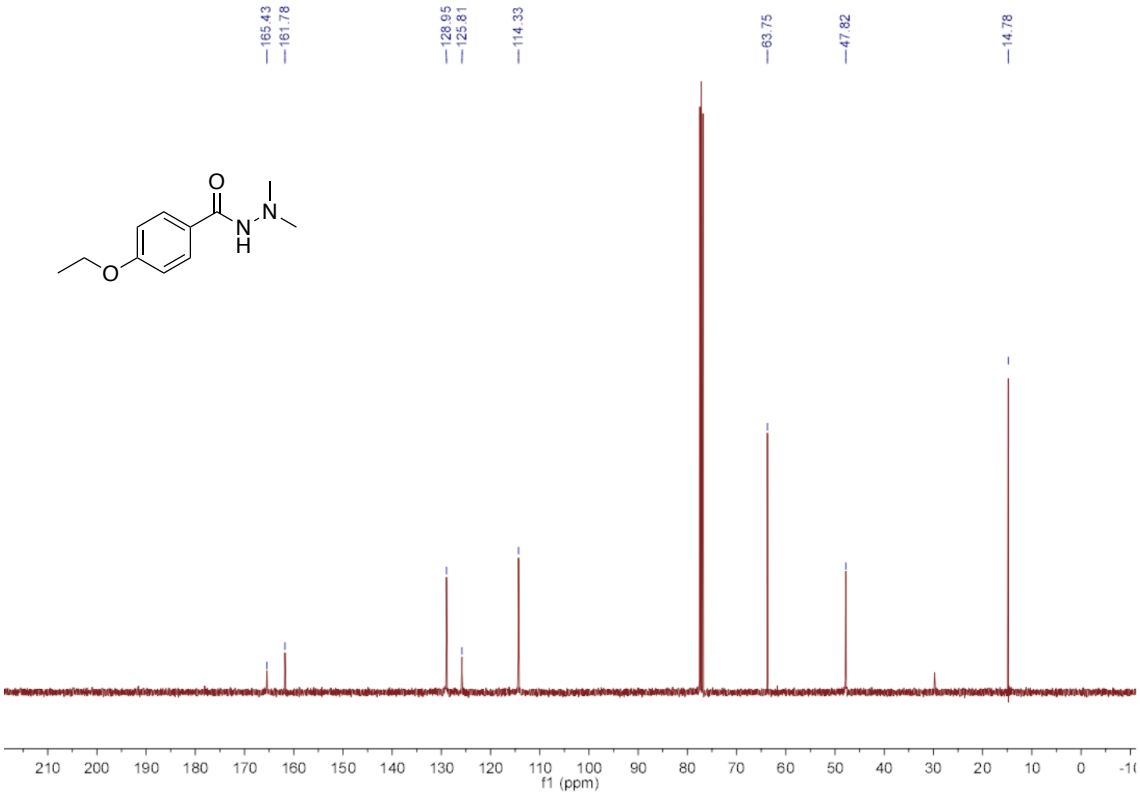
^{13}C NMR spectrum of *N*',*N*',4-trimethylbenzohydrazide **(2r):** (100.6 MHz, $CDCl_3$)



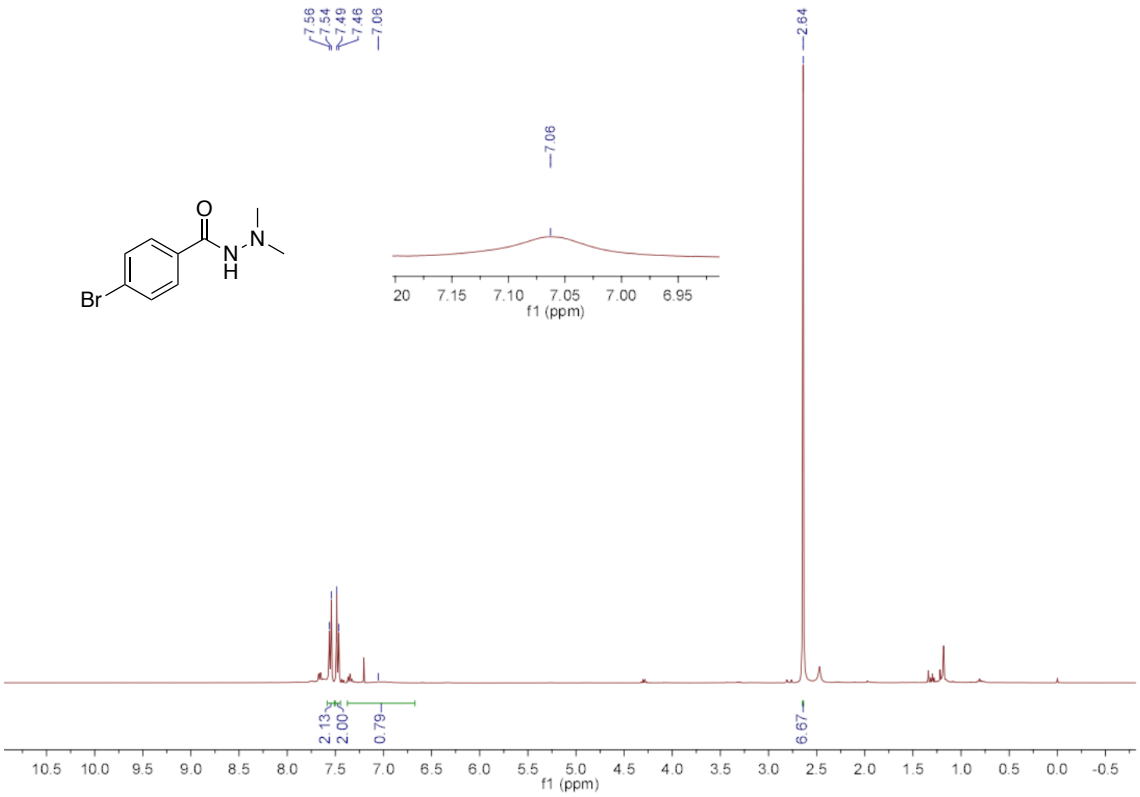
^{1}H NMR spectrum of 4-ethoxy-*N*',*N*'-dimethylbenzohydrazide **(2s):** (400 MHz, $CDCl_3$)



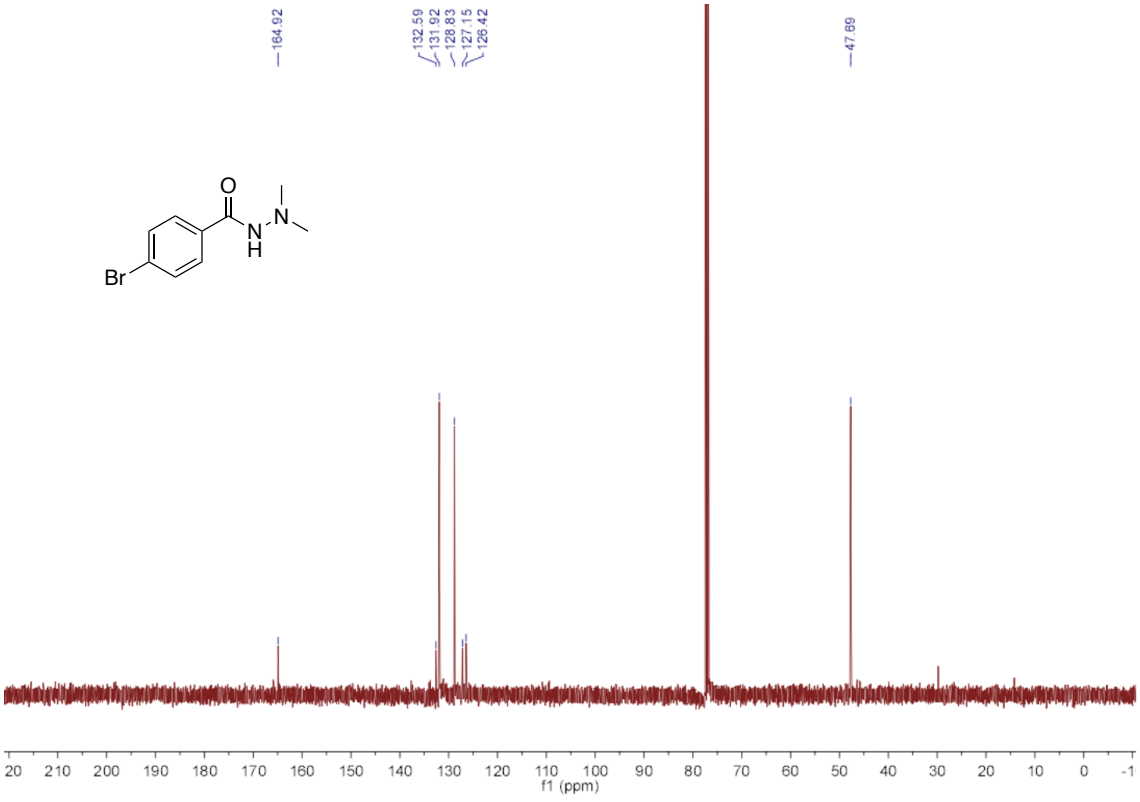
^{13}C NMR spectrum of 4-ethoxy-*N*',*N*'-dimethylbenzohydrazide **(2s):** (100.6 MHz, $CDCl_3$)



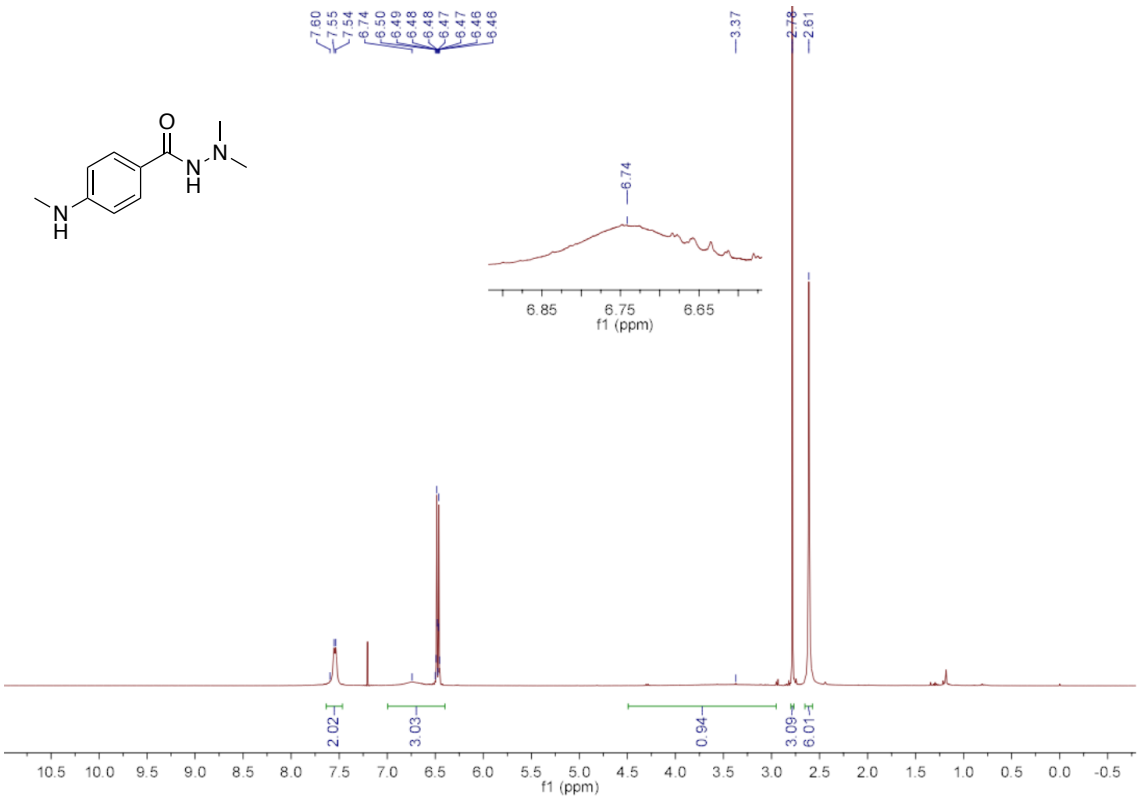
^{1}H NMR spectrum of 4-bromo-*N*',*N*'-dimethylbenzohydrazide **(2t):** (400 MHz, $CDCl_3$)



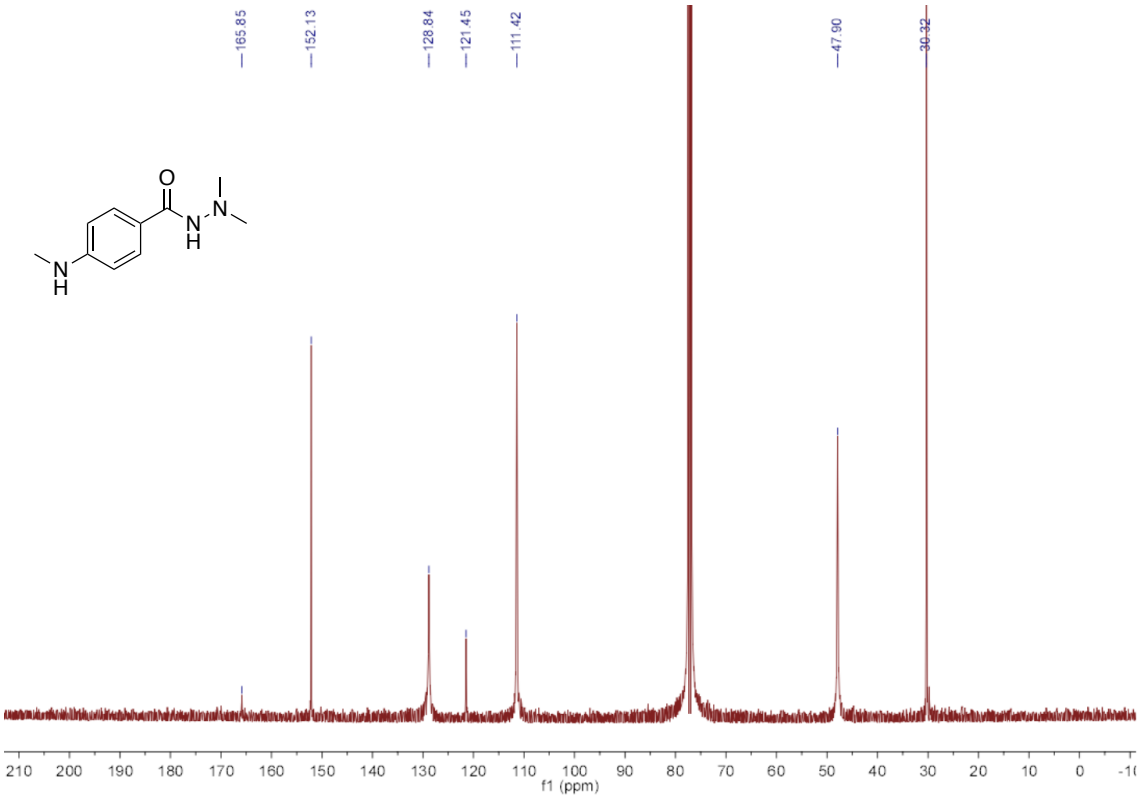
^{13}C NMR spectrum of 4-bromo-*N*',*N*'-dimethylbenzohydrazide **(2t):** (100.6 MHz, $CDCl_3$)



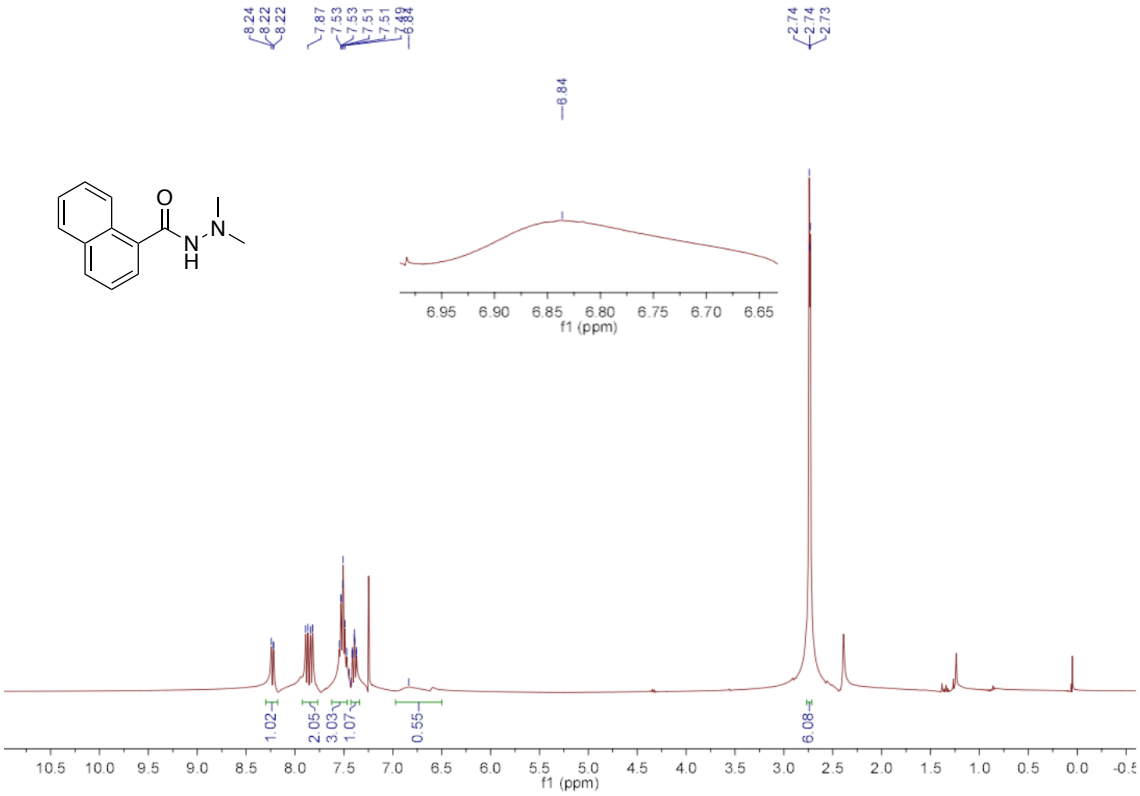
^{1}H NMR spectrum of *N*',*N*'-dimethyl-4-(methylamino)benzohydrazide **(2u):** (400 MHz, $CDCl_3$)



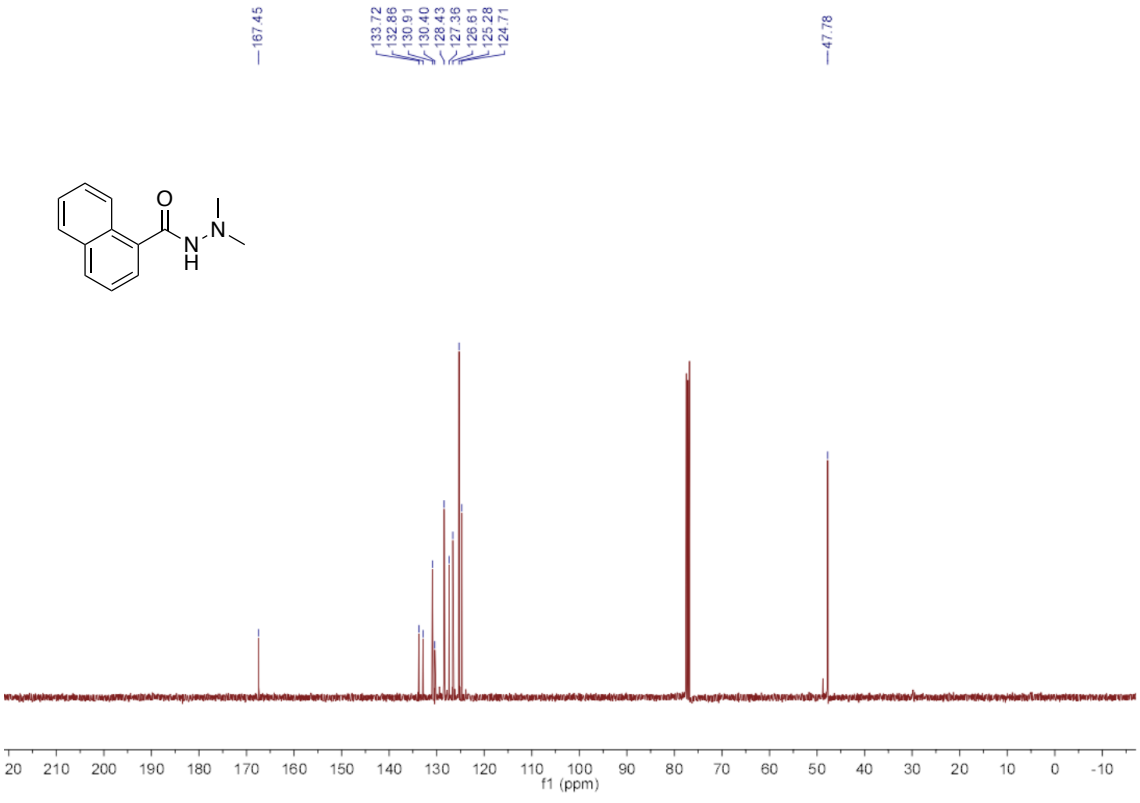
^{13}C NMR spectrum of *N*',*N*'-dimethyl-4-(methylamino)benzohydrazide **(2u):** (100.6 MHz, $CDCl_3$)



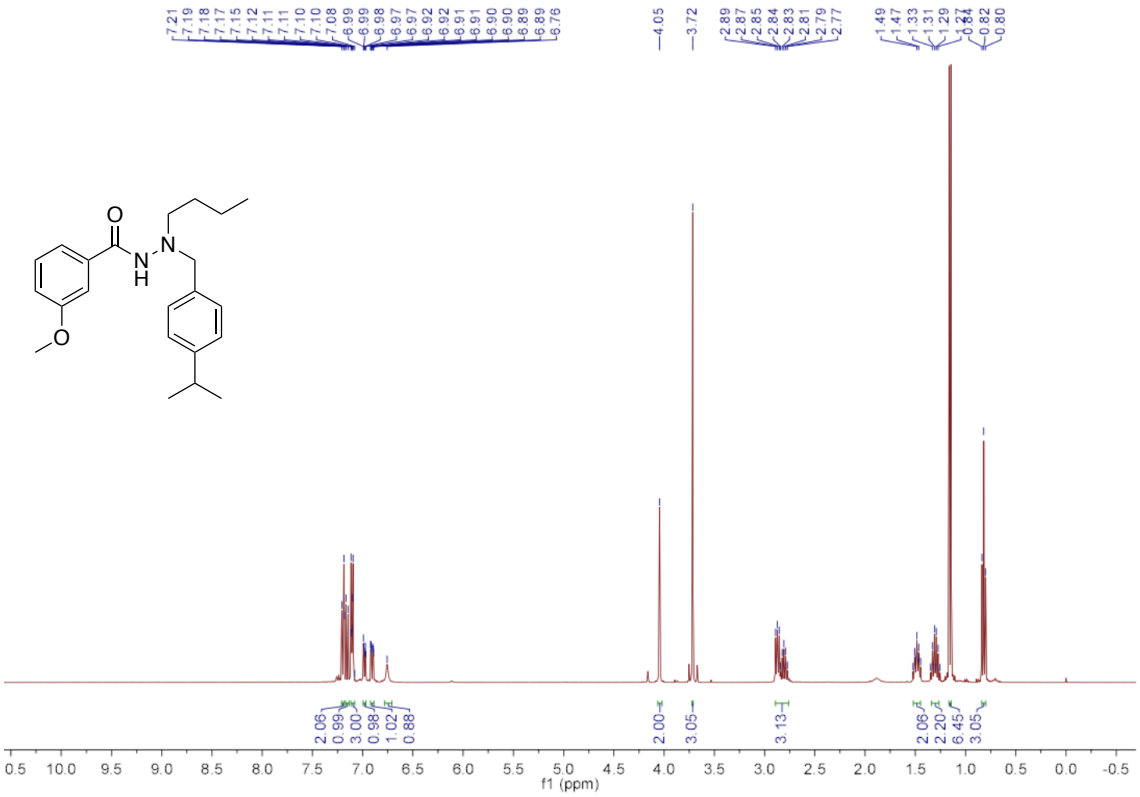
^{1}H NMR spectrum of *N*',*N*'-dimethyl-1-naphthohydrazide **(2v):** (400 MHz, $CDCl_3$)



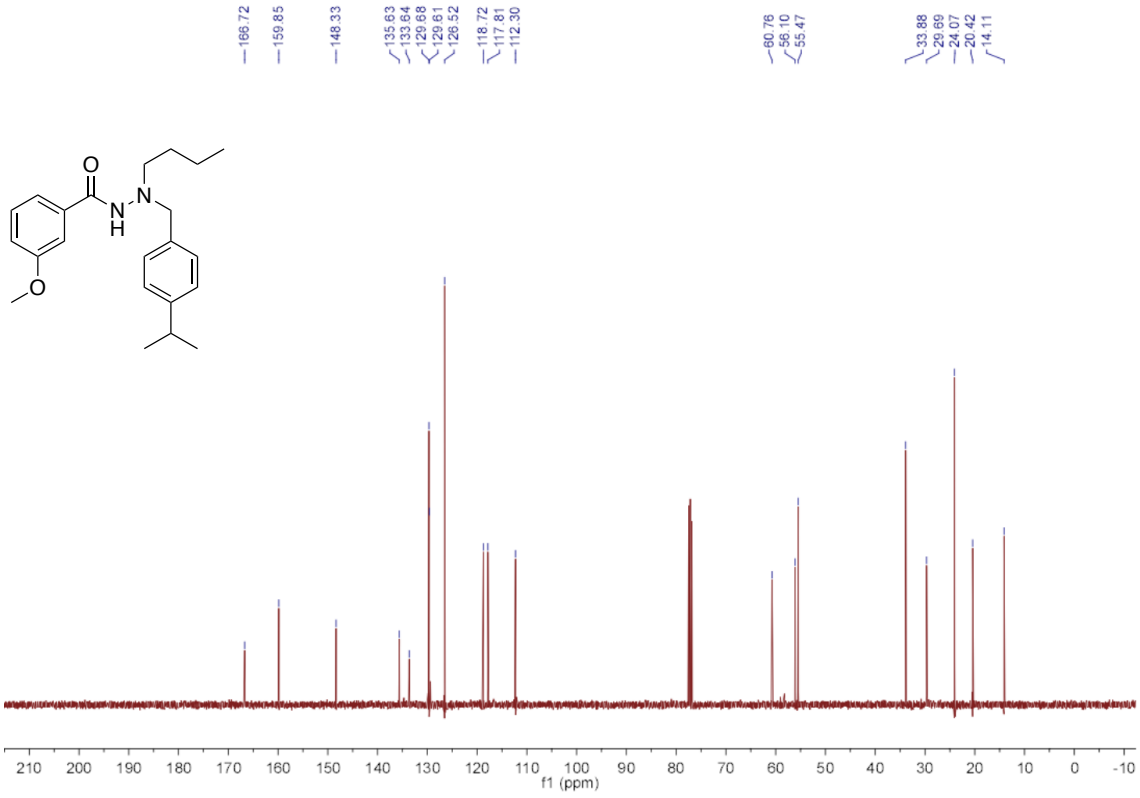
^{13}C NMR spectrum of *N*',*N*'-dimethyl-1-naphthohydrazide **(2v):** (100.6 MHz, $CDCl_3$)



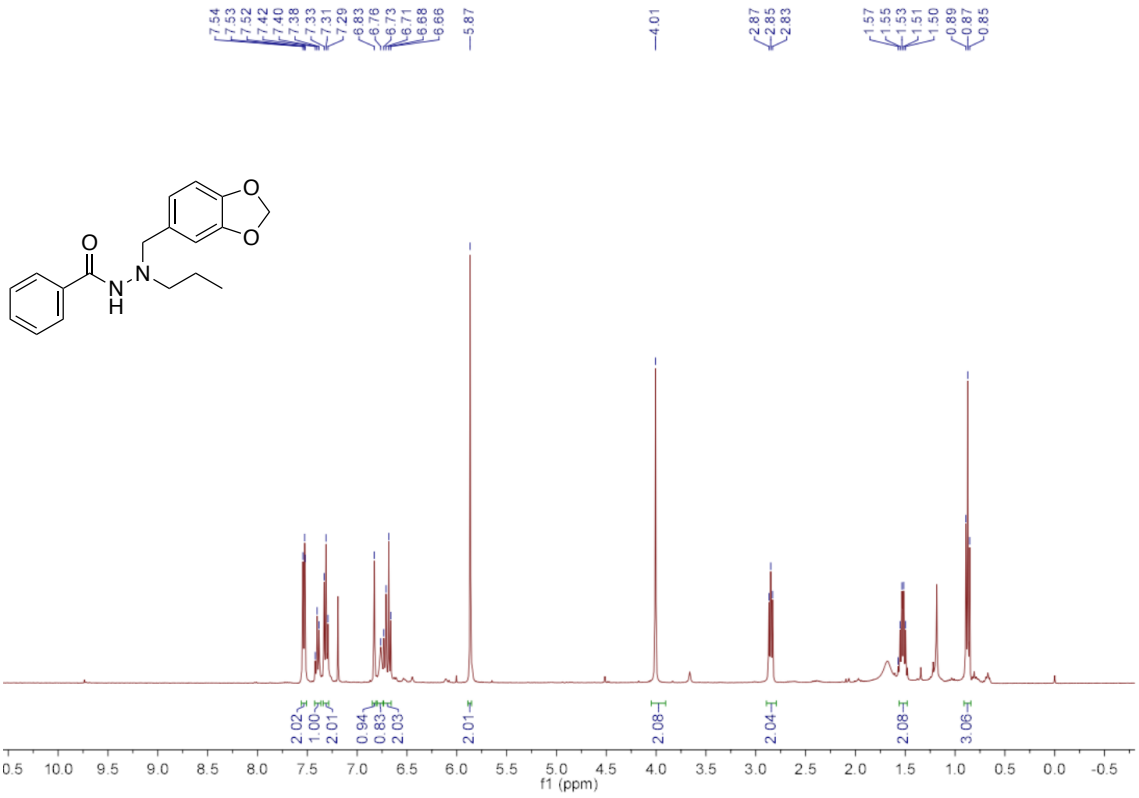
^{1}H NMR spectrum of *N*'-butyl-*N*'-(4-isopropylbenzyl)-3-methoxybenzohydrazide **(3a):** (400 MHz, $CDCl_3$)



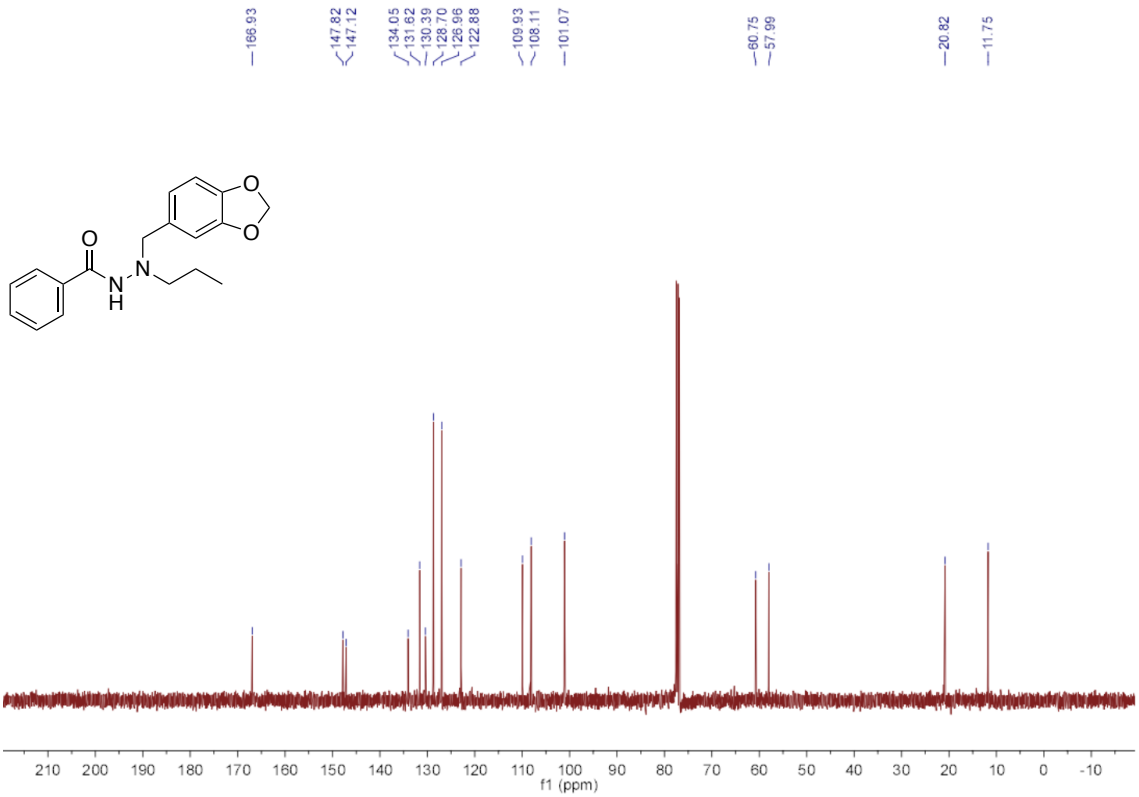
^{13}C NMR spectrum of *N*'-butyl-*N*'-(4-isopropylbenzyl)-3-methoxybenzohydrazide **(3a):** (100.6 MHz, $CDCl_3$)



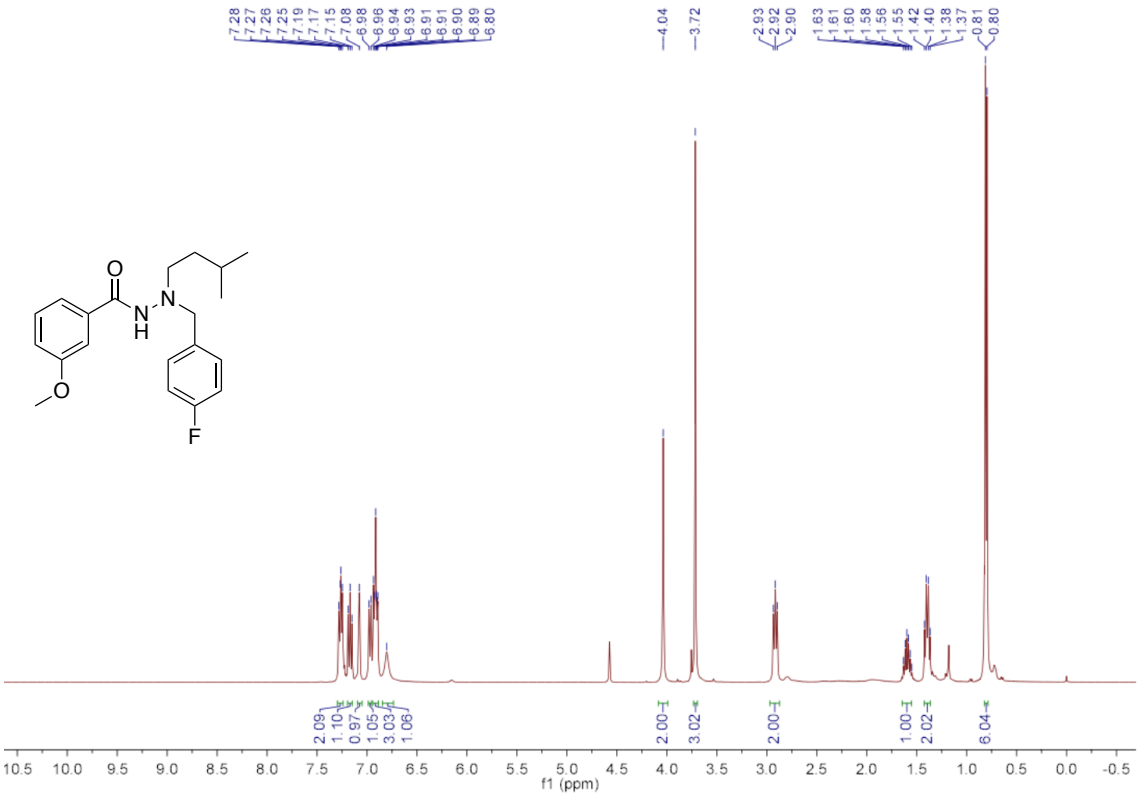
^{1}H NMR spectrum of *N*'-(benzo[*d*][1,3]dioxol-5-ylmethyl)-*N*'-propylbenzohydrazide **(3b):** (400 MHz, $CDCl_3$)



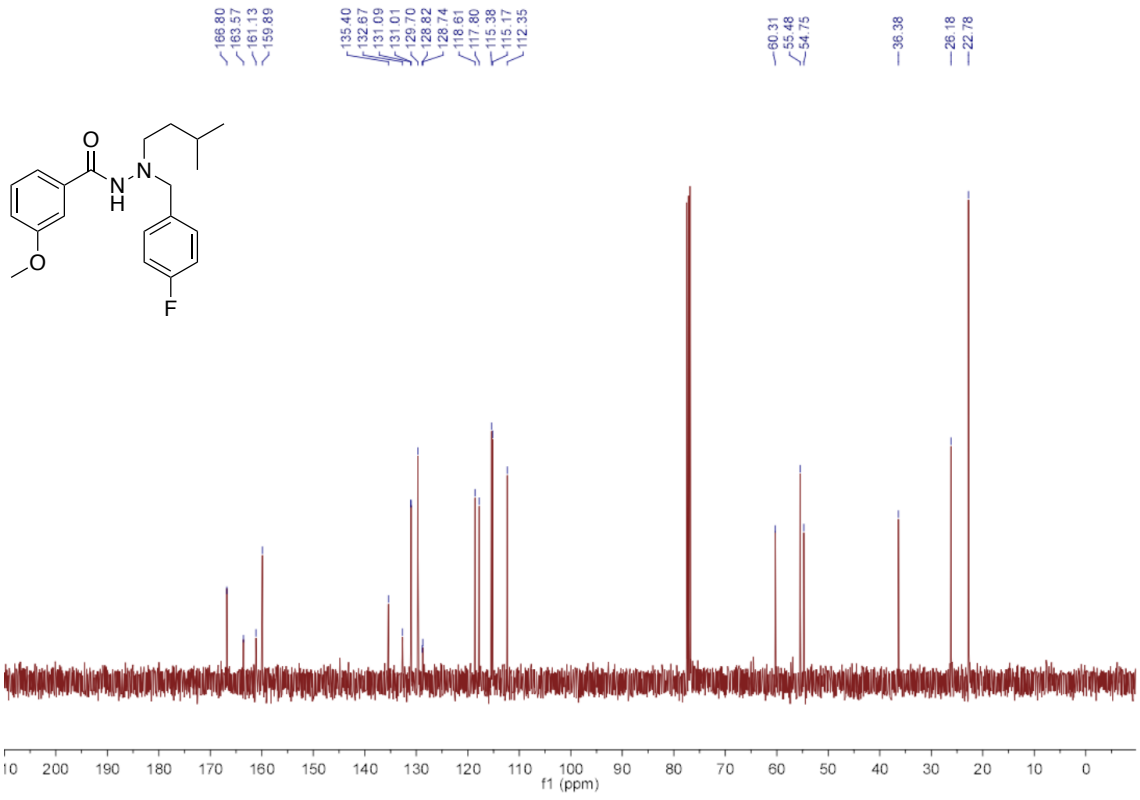
^{13}C NMR spectrum of *N*'-(benzo[*d*][1,3]dioxol-5-ylmethyl)-*N*'-propylbenzohydrazide **(3b):** (100.6 MHz, $CDCl_3$)



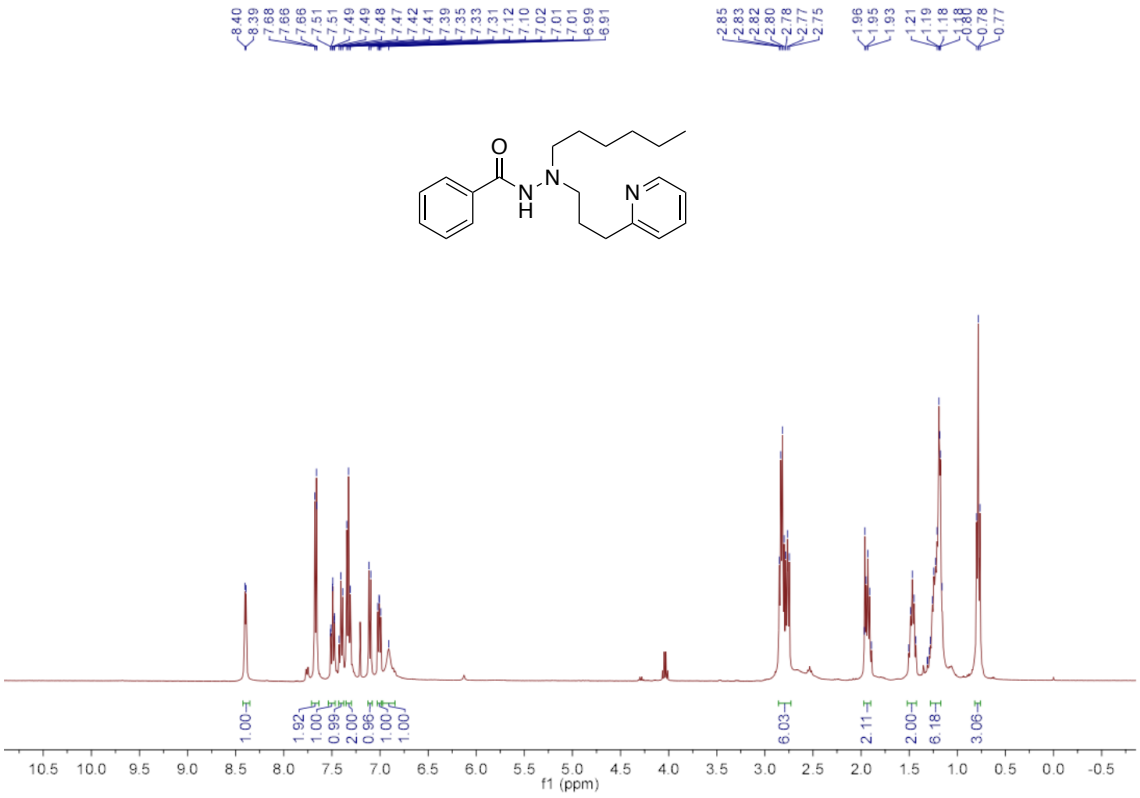
^{1}H NMR spectrum of *N*'-(4-fluorobenzyl)-*N*'-isopentyl-3-methoxybenzohydrazide **(3c):** (400 MHz, $CDCl_3$)



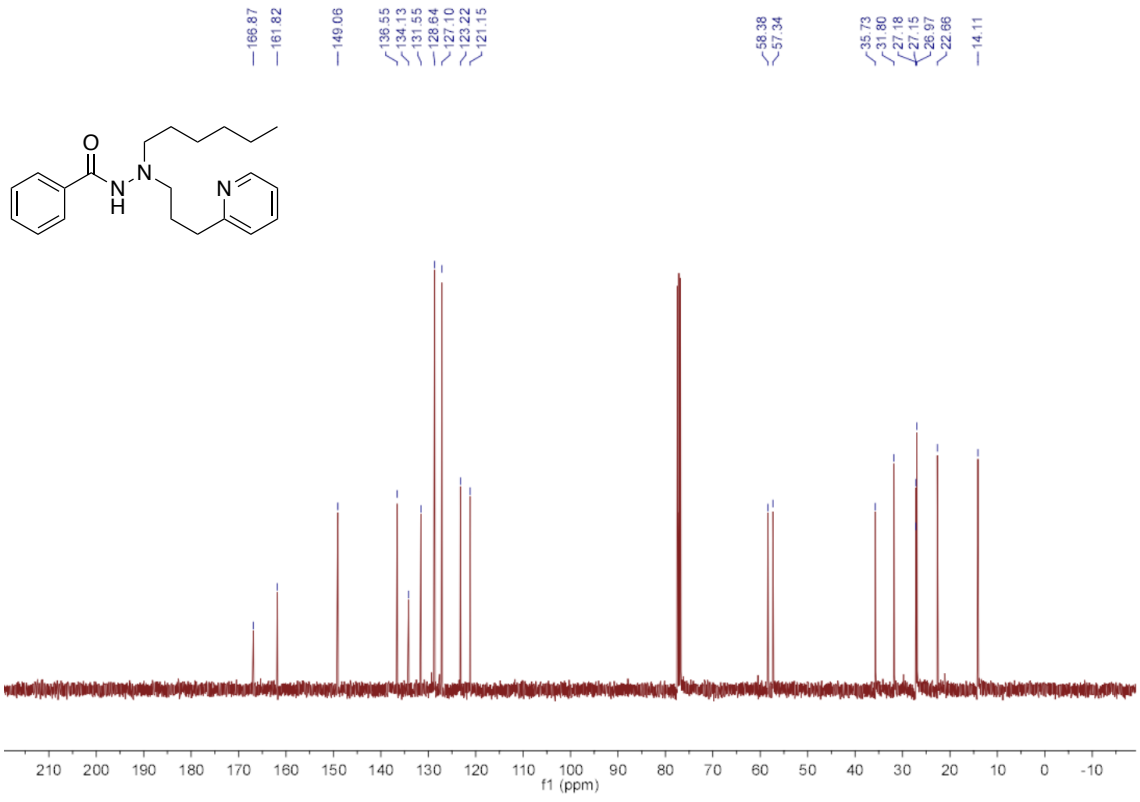
^{13}C NMR spectrum of *N*'-(4-fluorobenzyl)-*N*'-isopentyl-3-methoxybenzohydrazide **(3c):** (100.6 MHz, $CDCl_3$)



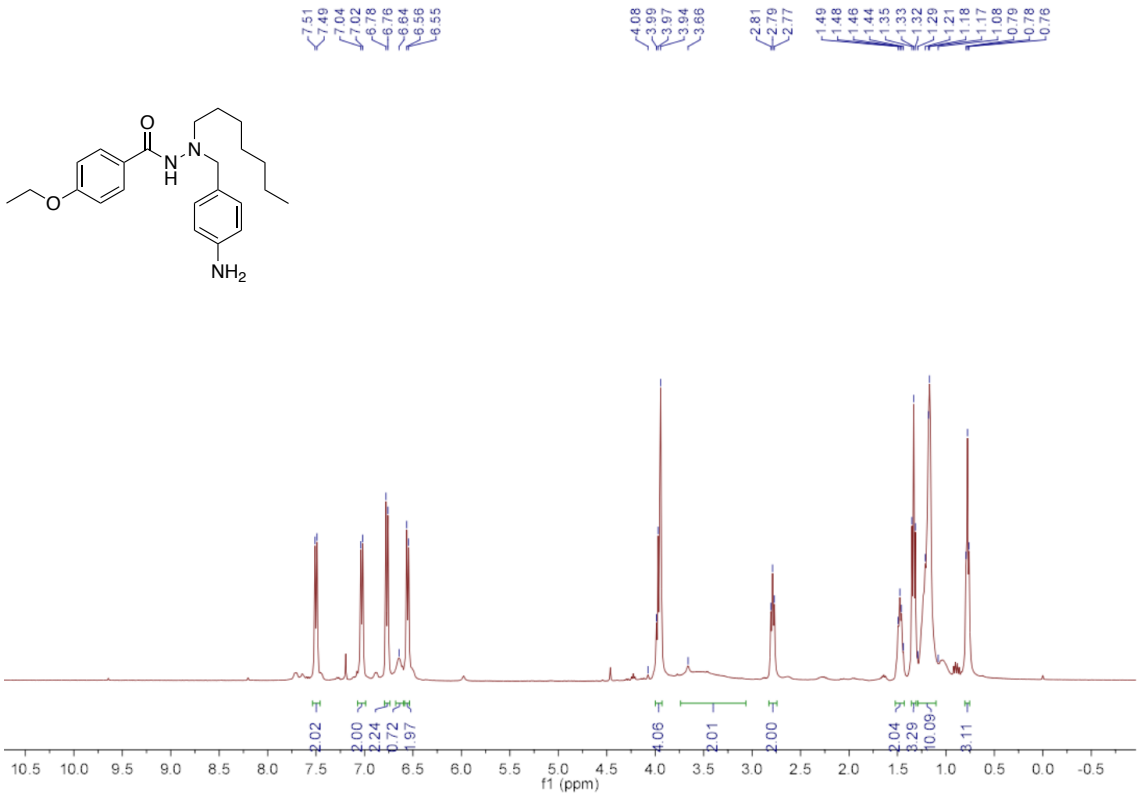
^{1}H NMR spectrum of *N*'-hexyl-*N*'-(3-(pyridin-2-yl)propyl)benzohydrazide **(3d):** (400 MHz, $CDCl_3$)



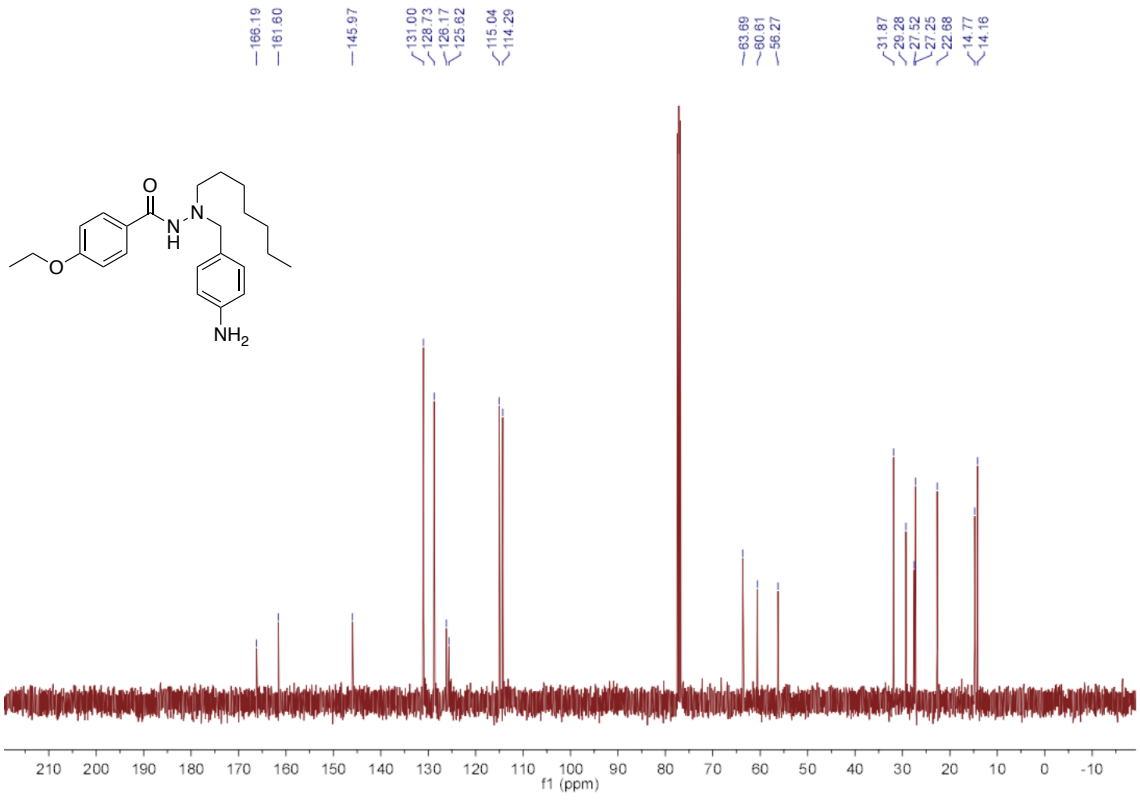
^{13}C NMR spectrum of *N*'-hexyl-*N*'-(3-(pyridin-2-yl)propyl)benzohydrazide **(3d):** (100.6 MHz, $CDCl_3$)



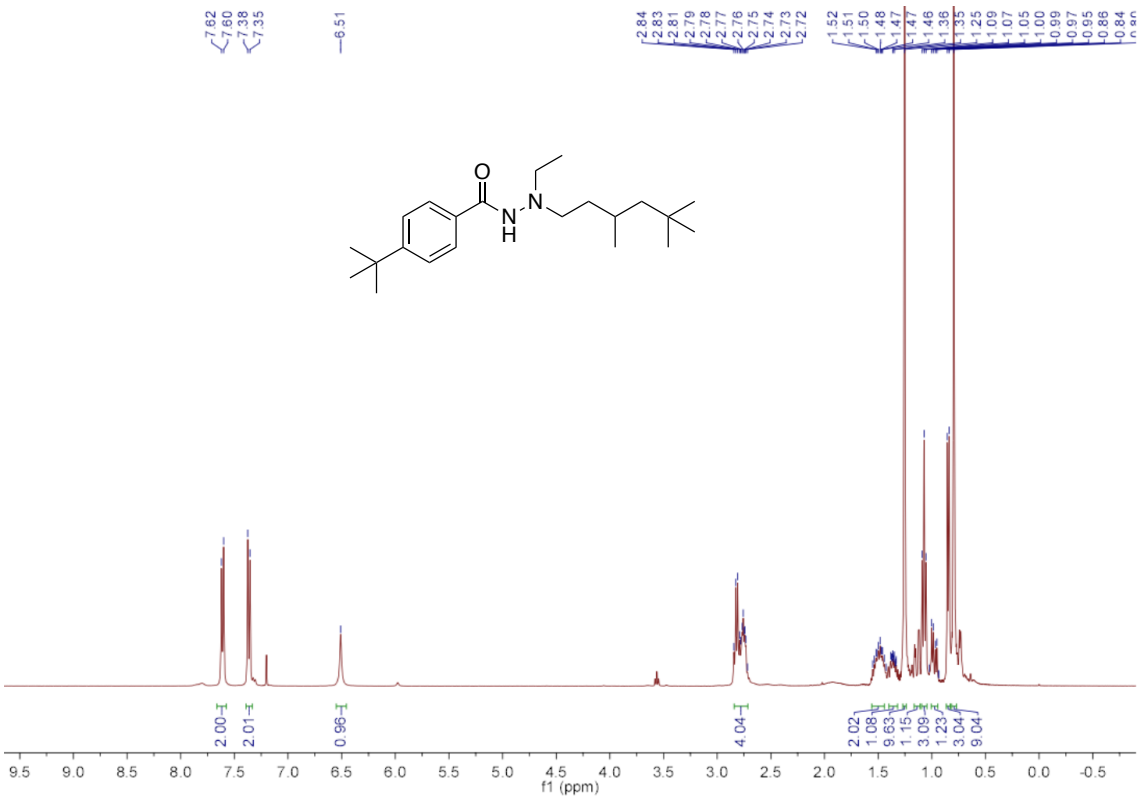
^{1}H NMR spectrum of *N*'-(4-aminobenzyl)-4-ethoxy-*N*'-heptylbenzohydrazide **(3e):** (400 MHz, $CDCl_3$)



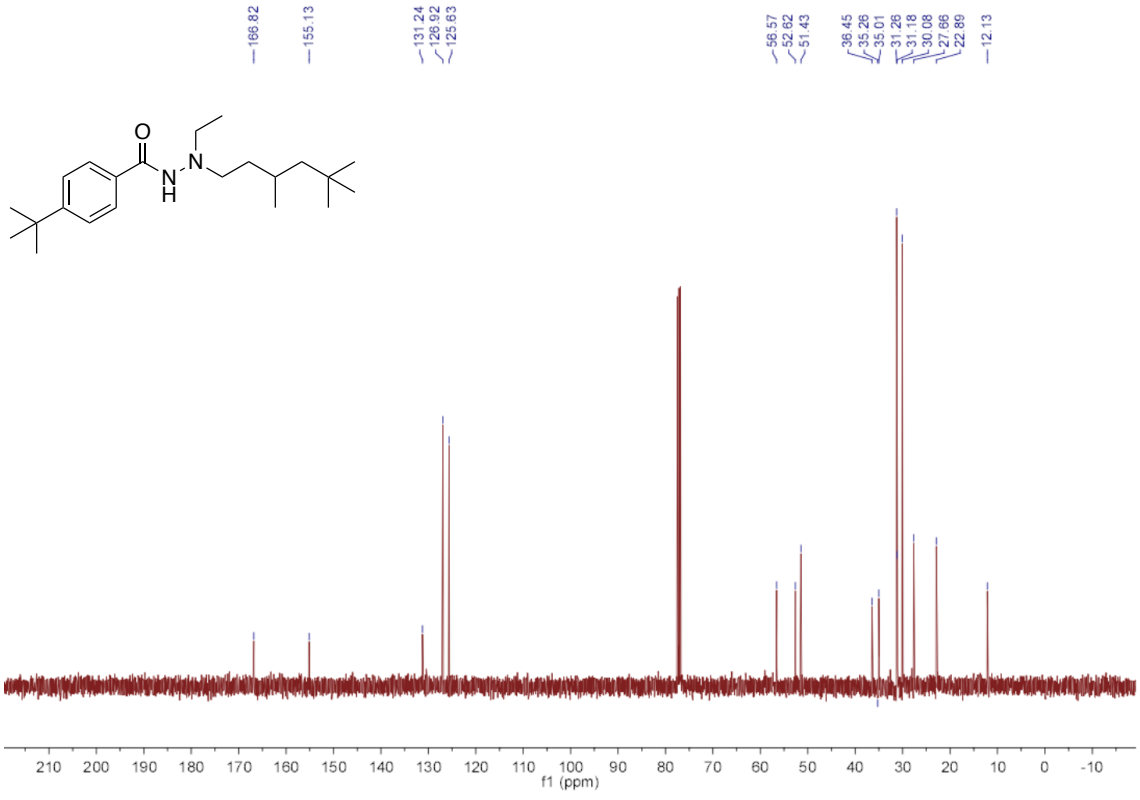
^{13}C NMR spectrum of *N*'-(4-aminobenzyl)-4-ethoxy-*N*'-heptylbenzohydrazide **(3e):** (100.6 MHz, $CDCl_3$)



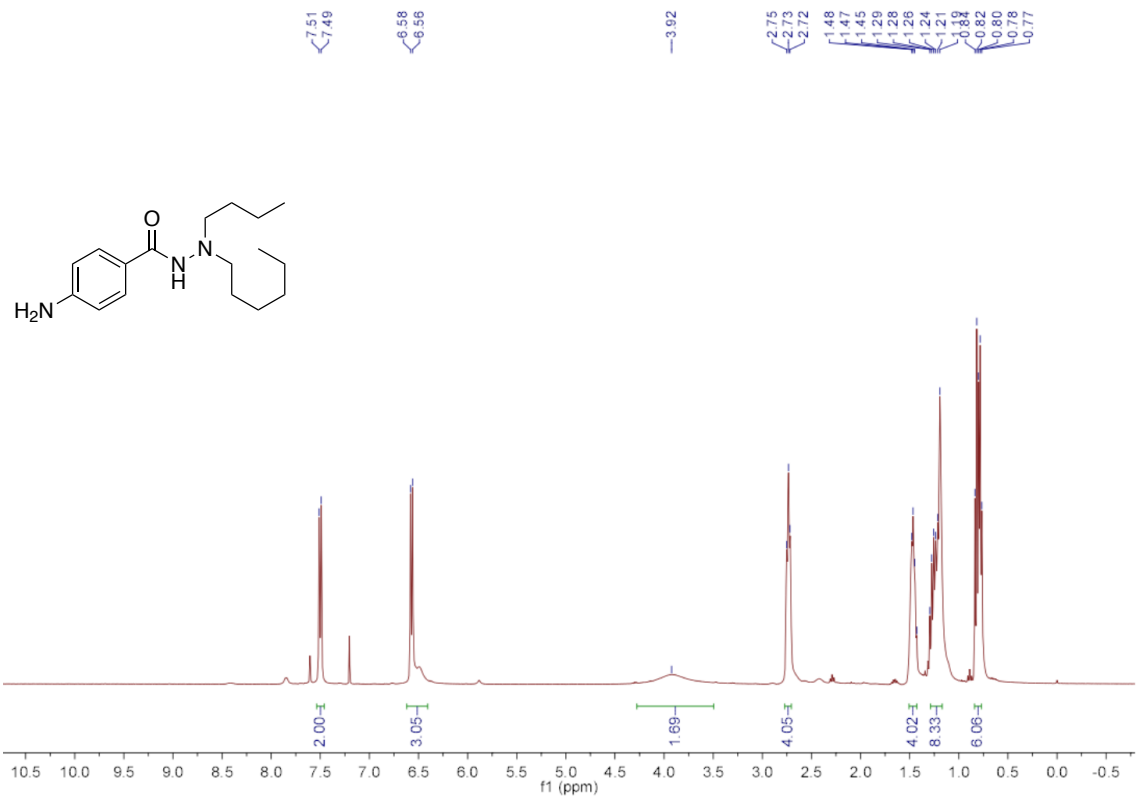
^{1}H NMR spectrum of 4-(*tert*-butyl)-*N*'-ethyl-*N*'-(3,5,5-trimethylhexyl)benzohydrazide **(3f):** (400 MHz, $CDCl_3$)



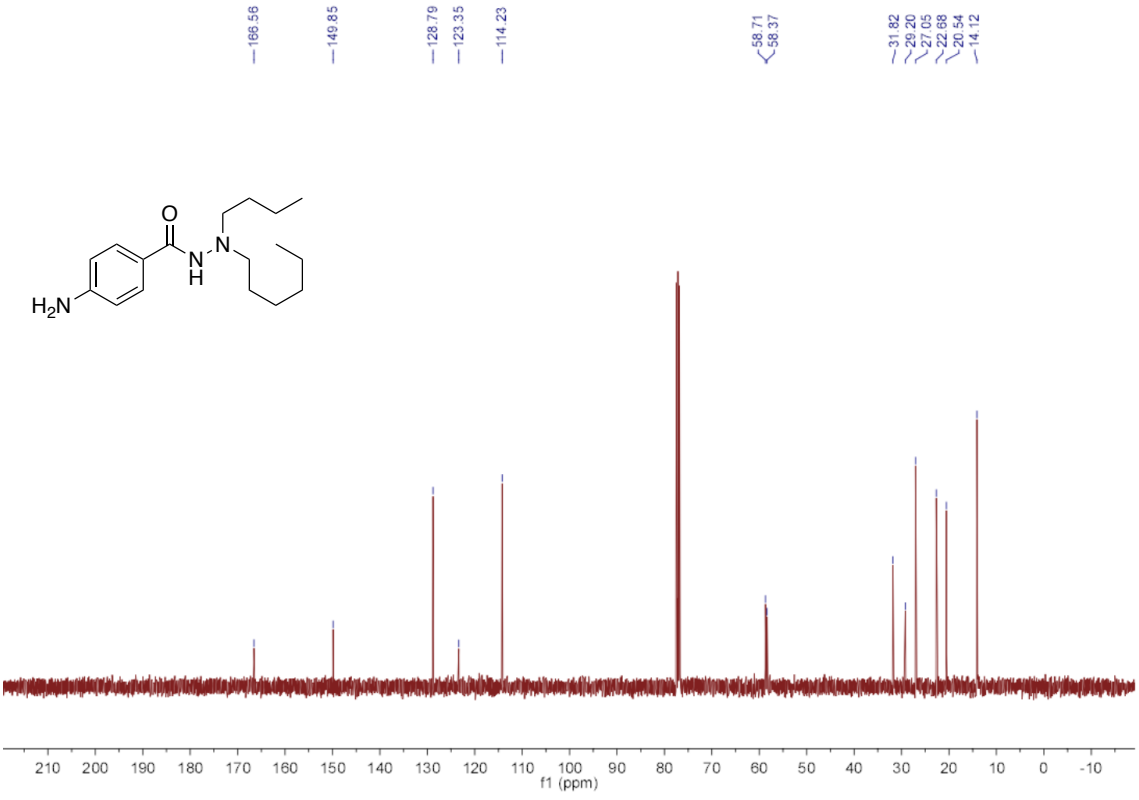
^{13}C NMR spectrum of 4-(*tert*-butyl)-*N*'-ethyl-*N*'-(3,5,5-trimethylhexyl)benzohydrazide **(3f):** (100.6 MHz, $CDCl_3$)



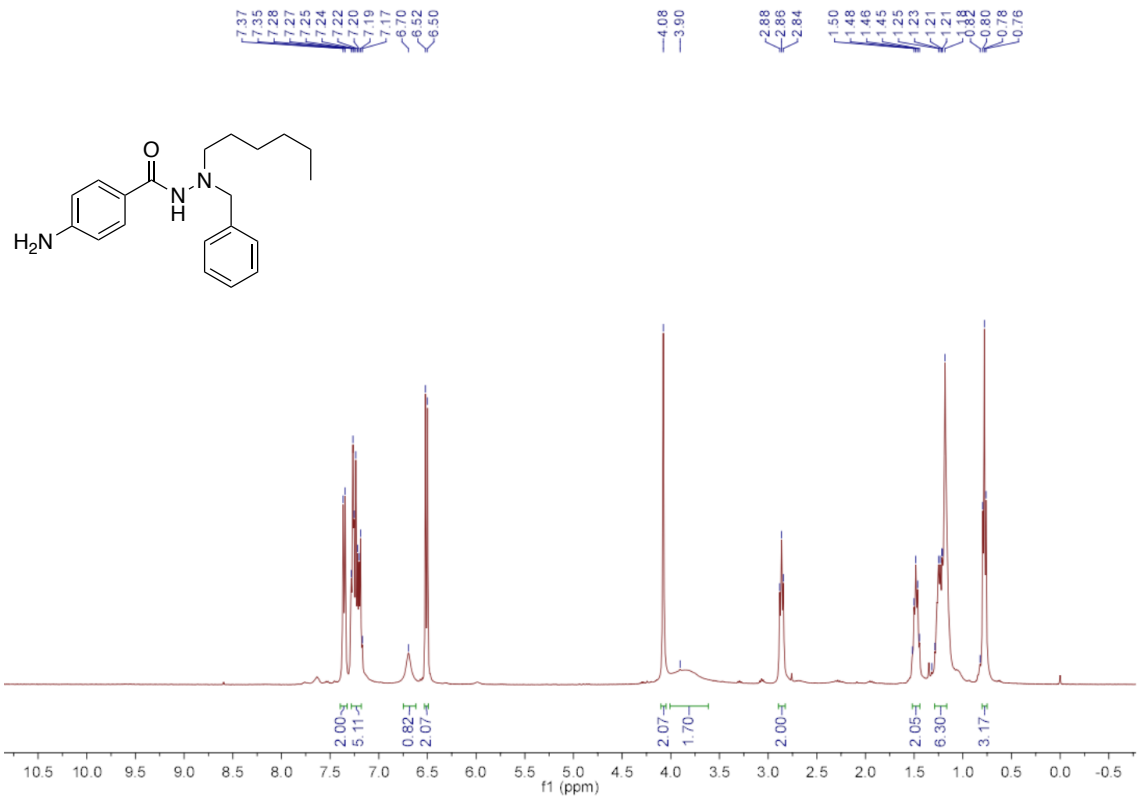
^{1}H NMR spectrum of 4-amino-*N*'-butyl-*N*'-hexylbenzohydrazide **(3g):** (400 MHz, $CDCl_3$)



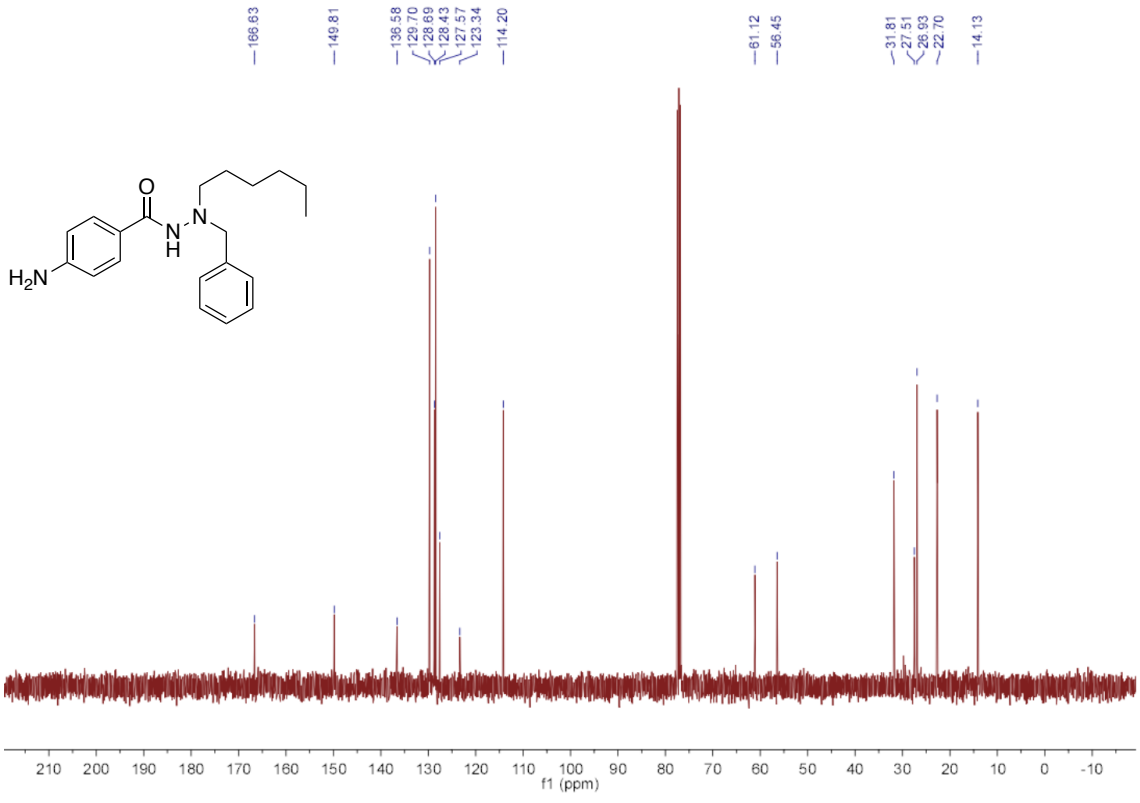
^{13}C NMR spectrum of 4-amino-*N*'-butyl-*N*'-hexylbenzohydrazide **(3g):** (100.6 MHz, $CDCl_3$)



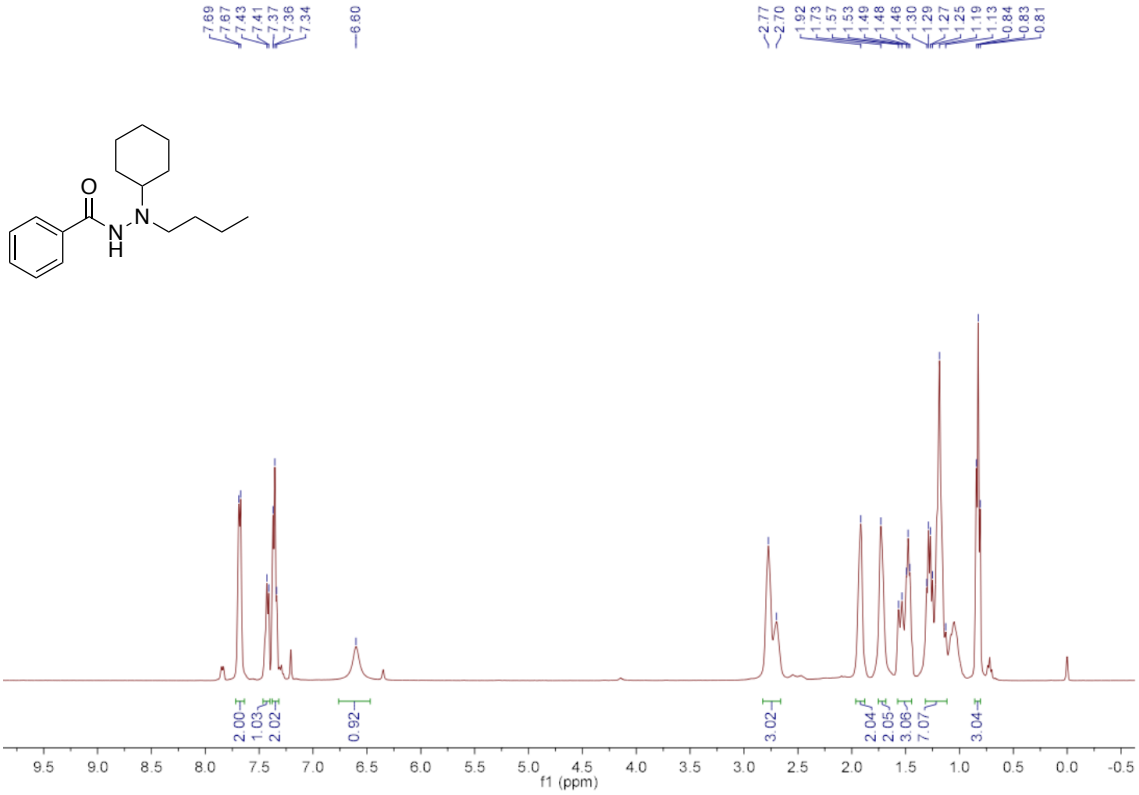
^{1}H NMR spectrum of 4-amino-*N*'-benzyl-*N*'-hexylbenzohydrazide **(3h):** (400 MHz, $CDCl_3$)



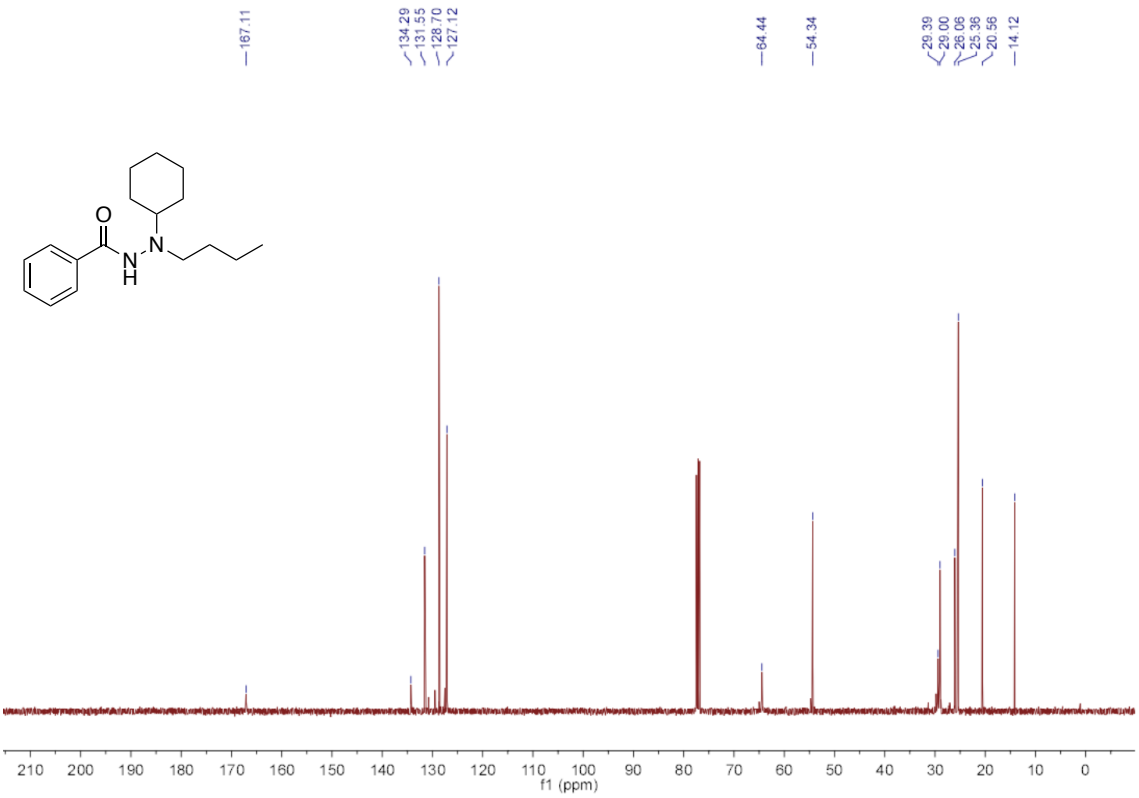
^{13}C NMR spectrum of 4-amino-*N*'-benzyl-*N*'-hexylbenzohydrazide **(3h):** (100.6 MHz, $CDCl_3$)



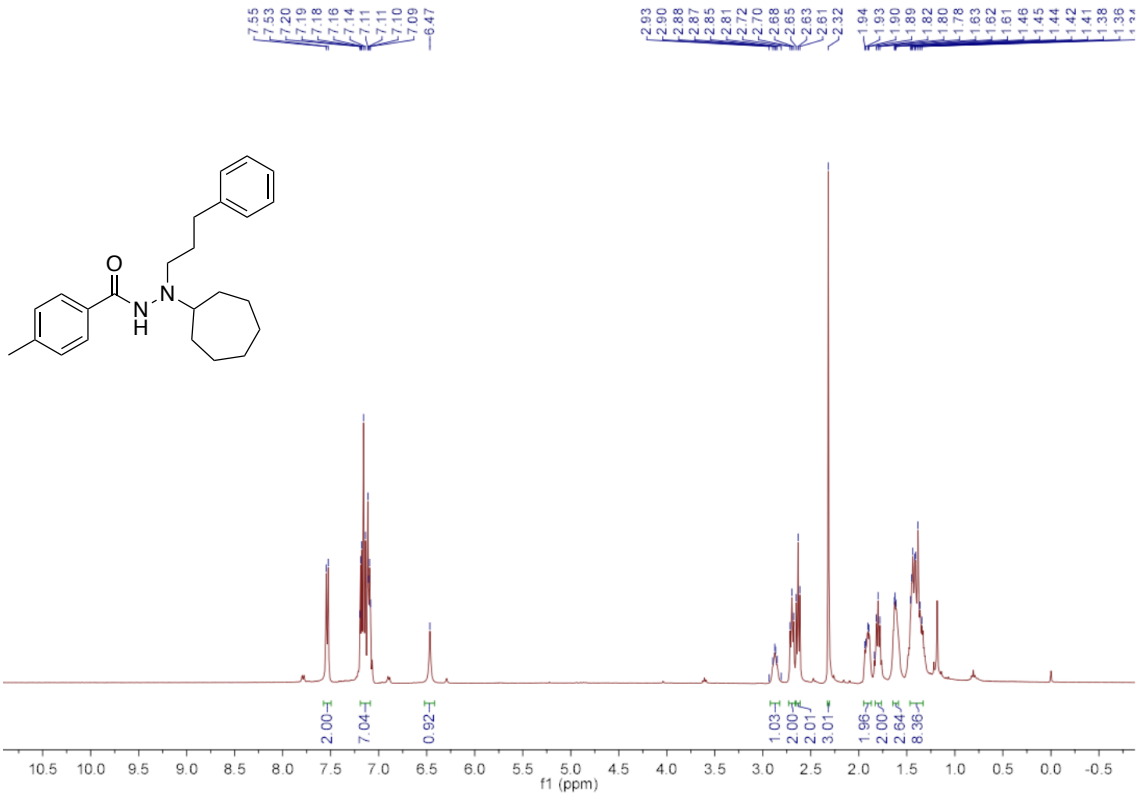
^{1}H NMR spectrum of *N*'-butyl-*N*'-cyclohexylbenzohydrazide **(3i):** (400 MHz, $CDCl_3$)



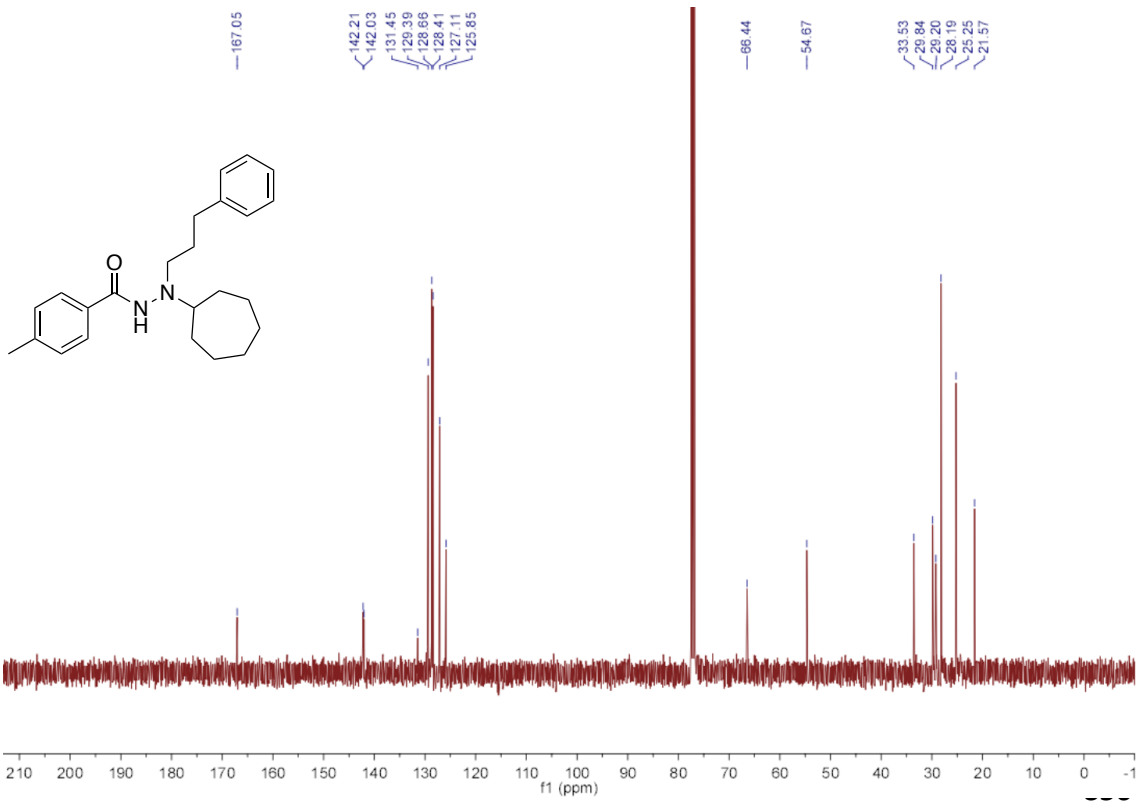
^{13}C NMR spectrum of *N*'-butyl-*N*'-cyclohexylbenzohydrazide **(3i):** (100.6 MHz, $CDCl_3$)



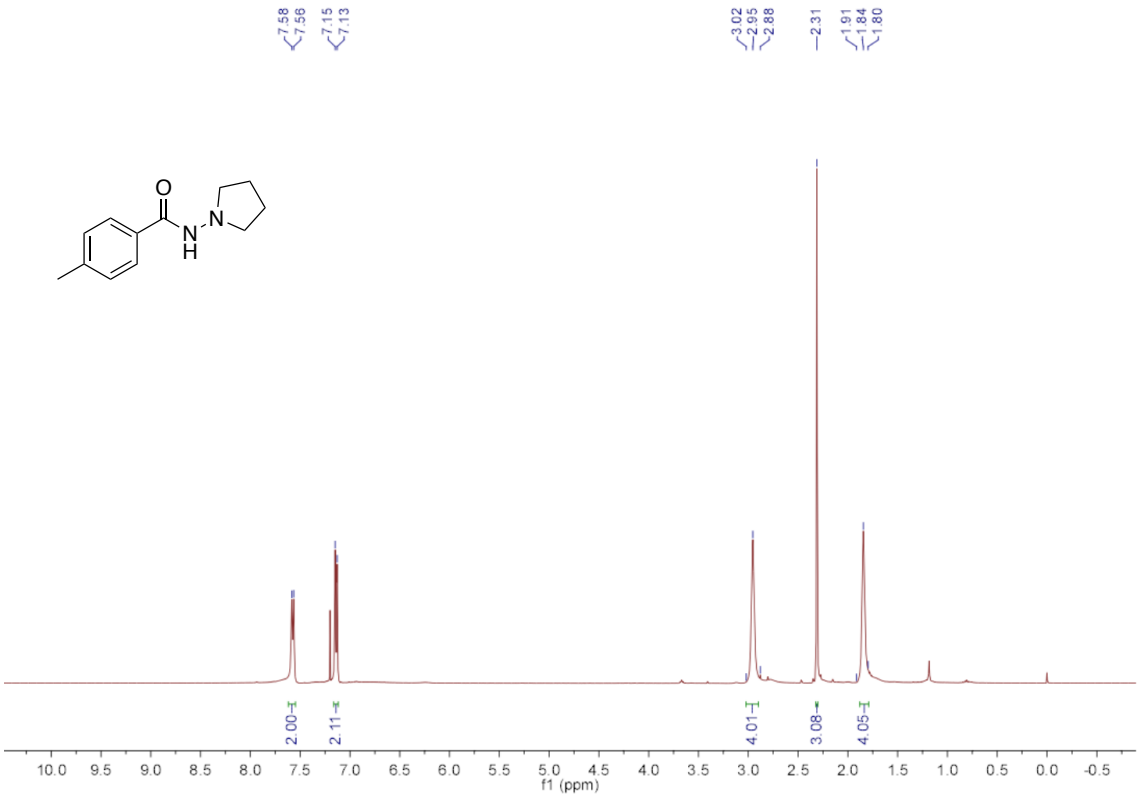
^{1}H NMR spectrum of *N*'-cycloheptyl-4-methyl-*N*'-(3-phenylpropyl)benzohydrazide **(3j):** (400 MHz, $CDCl_3$)



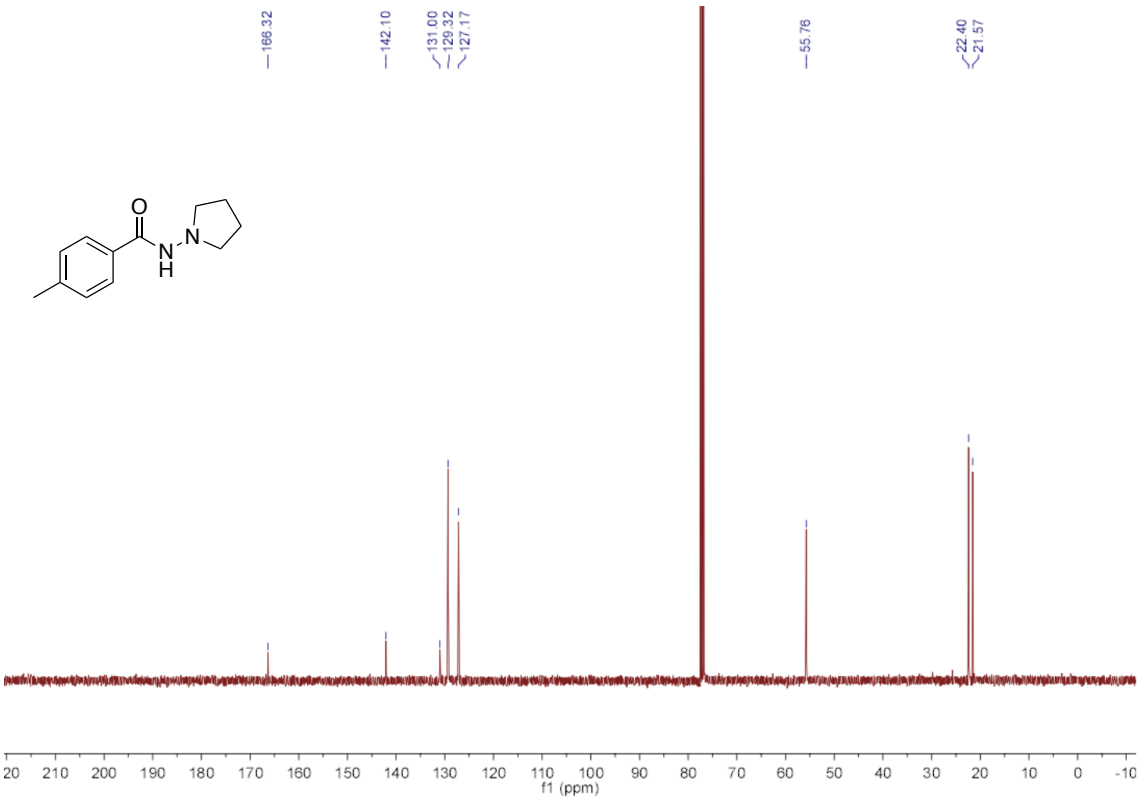
^{13}C NMR spectrum of *N*'-cycloheptyl-4-methyl-*N*'-(3-phenylpropyl)benzohydrazide **(3j):** (100.6 MHz, $CDCl_3$)



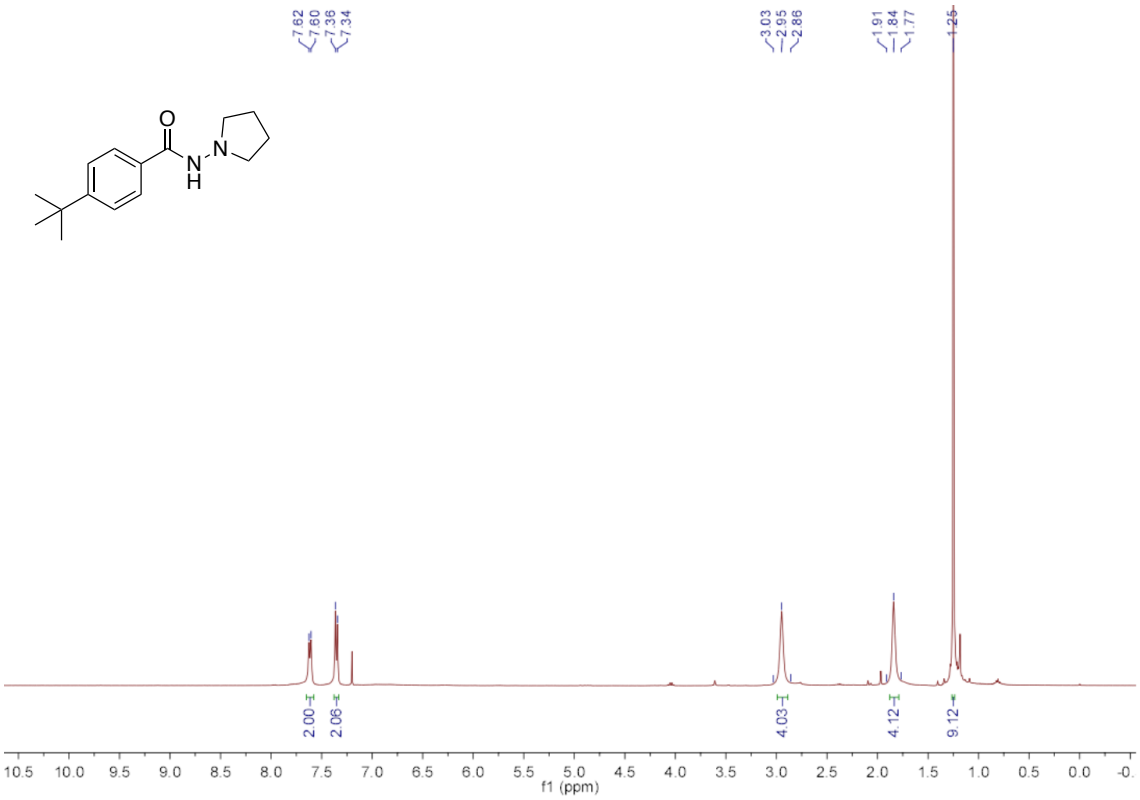
^{1}H NMR spectrum of 4-methyl-*N*-(pyrrolidin-1-yl)benzamide **(4a):** (400 MHz, $CDCl_3$)



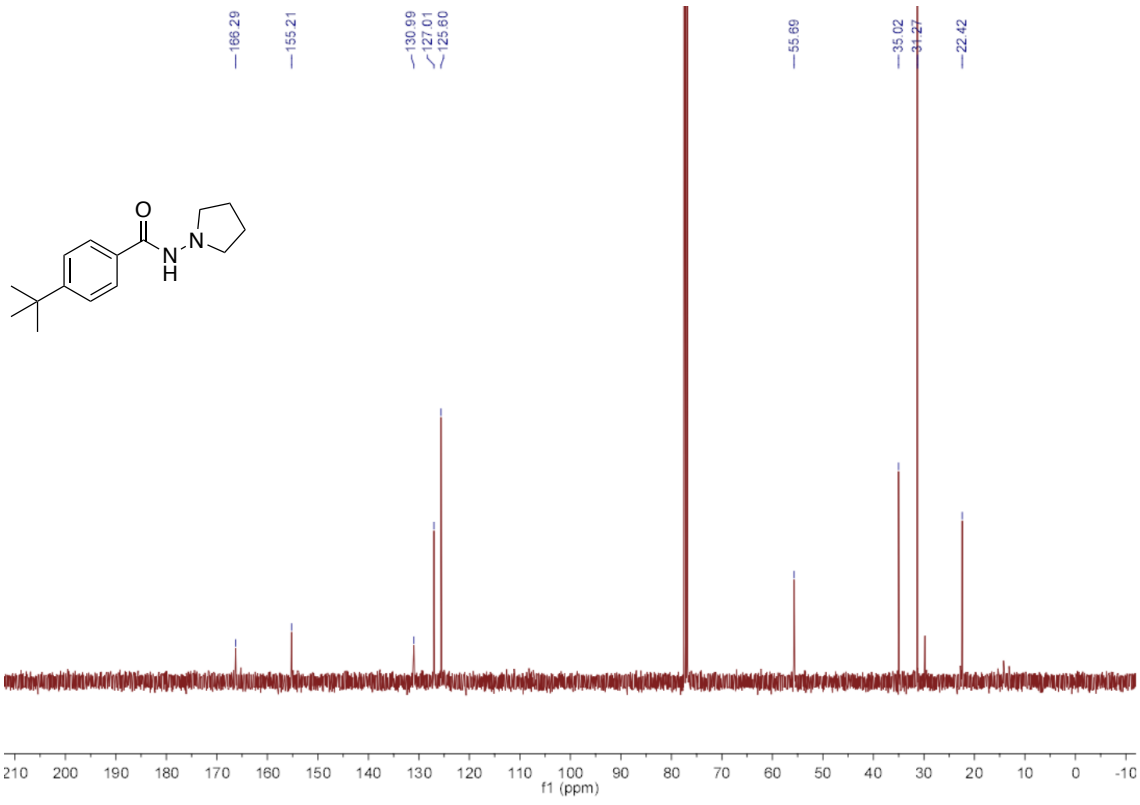
^{13}C NMR spectrum of 4-methyl-*N*-(pyrrolidin-1-yl)benzamide **(4a):** (100.6 MHz, $CDCl_3$)



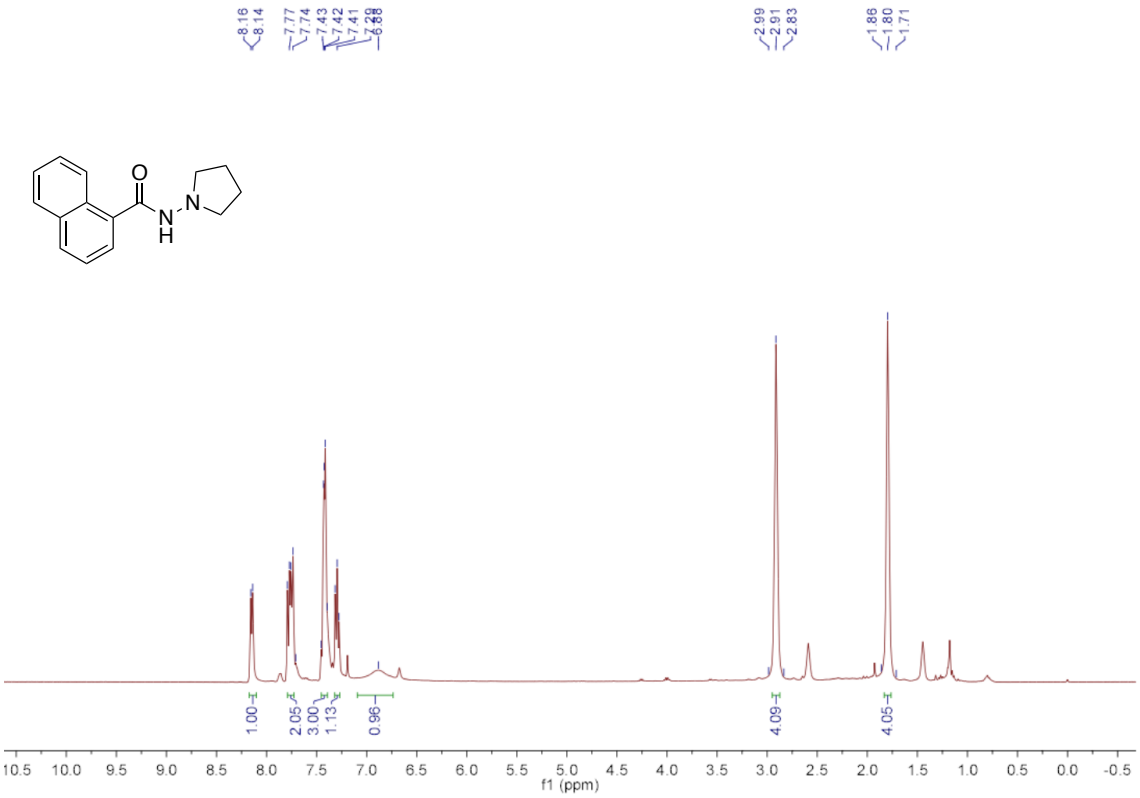
^{1}H NMR spectrum of 4-(*tert*-butyl)-*N*-(pyrrolidin-1-yl)benzamide **(4b):** (400 MHz, $CDCl_3$)



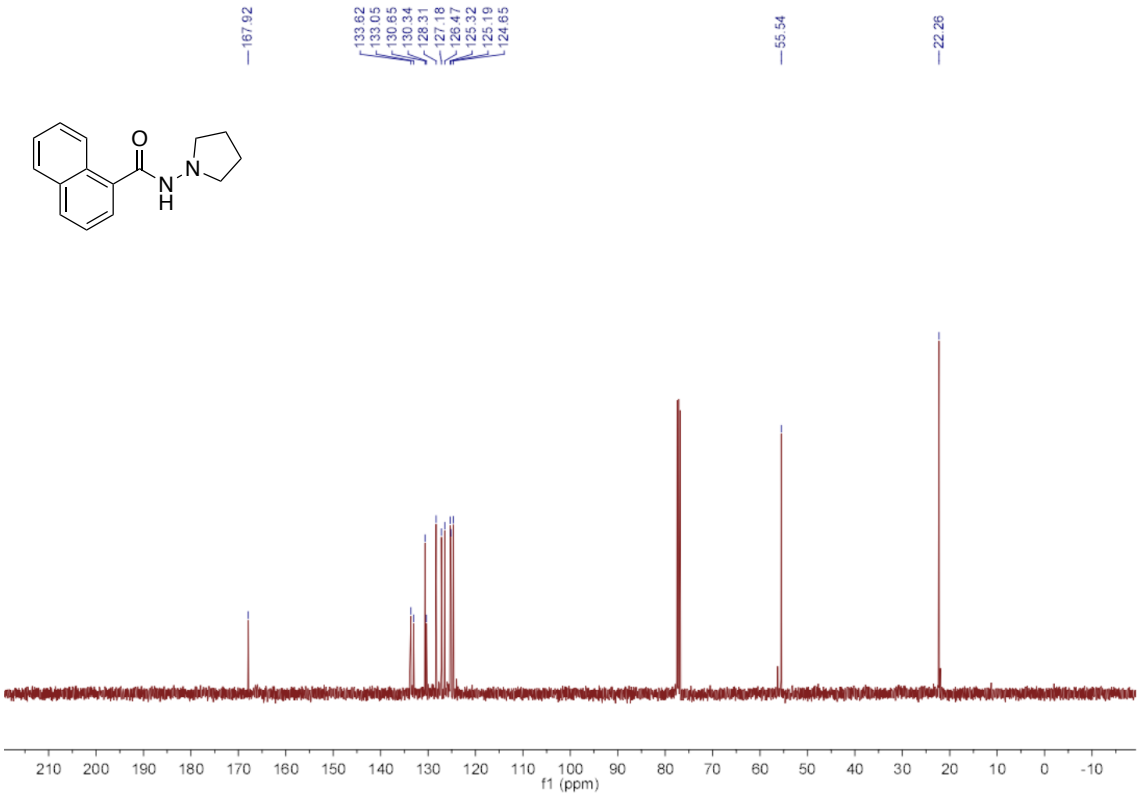
^{13}C NMR spectrum of 4-(*tert*-butyl)-*N*-(pyrrolidin-1-yl)benzamide **(4b):** (100.6 MHz, $CDCl_3$)



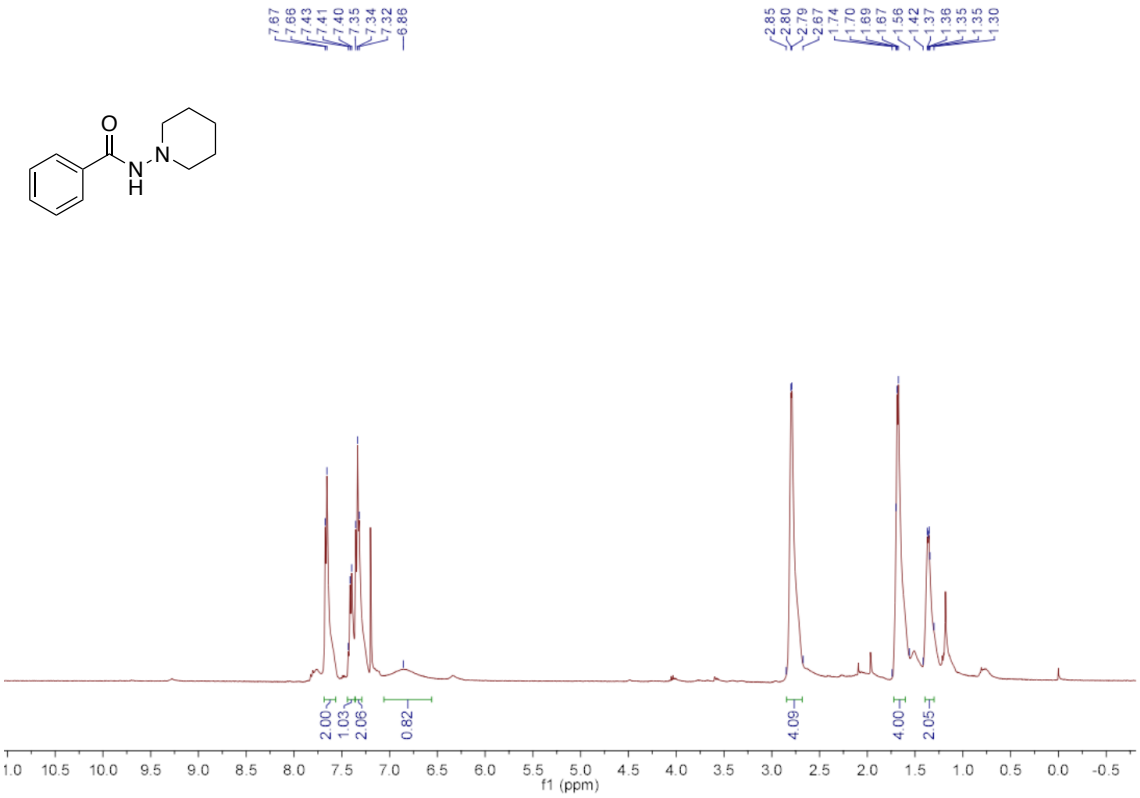
^{1}H NMR spectrum of *N*-(pyrrolidin-1-yl)-1-naphthamide **(4c):** (400 MHz, $CDCl_3$)



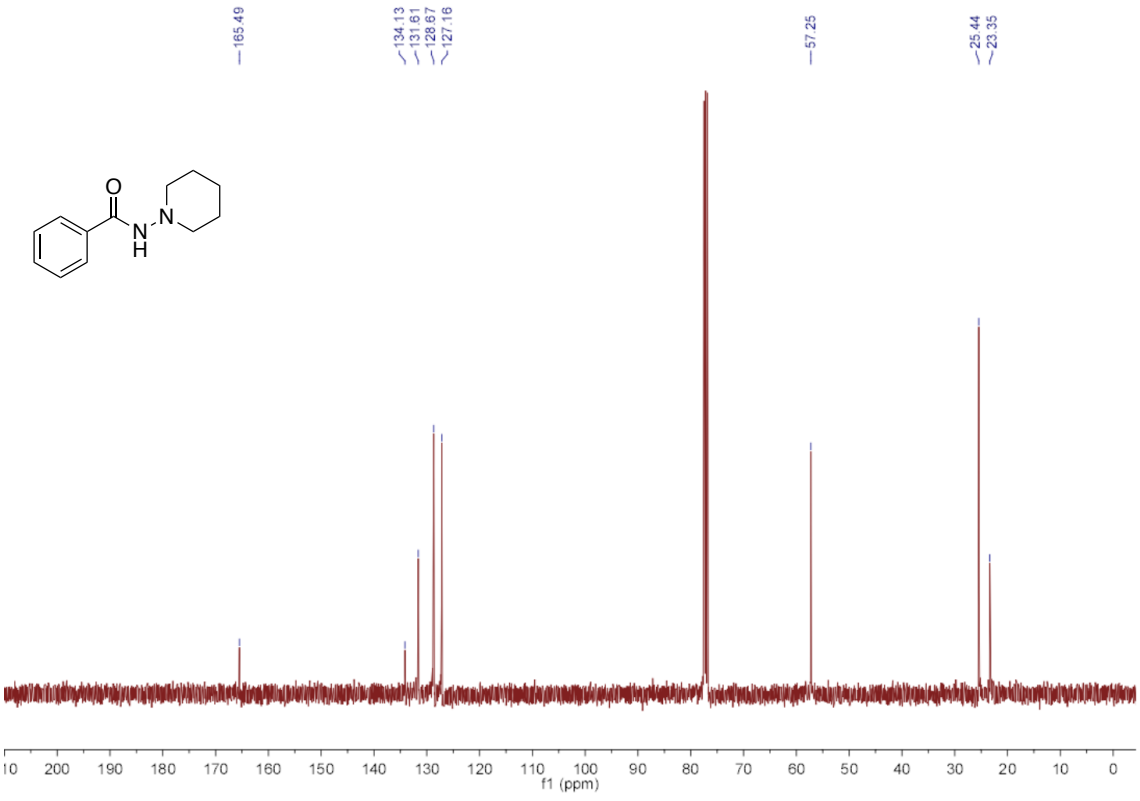
^{13}C NMR spectrum of *N*-(pyrrolidin-1-yl)-1-naphthamide **(4c):** (100.6 MHz, $CDCl_3$)



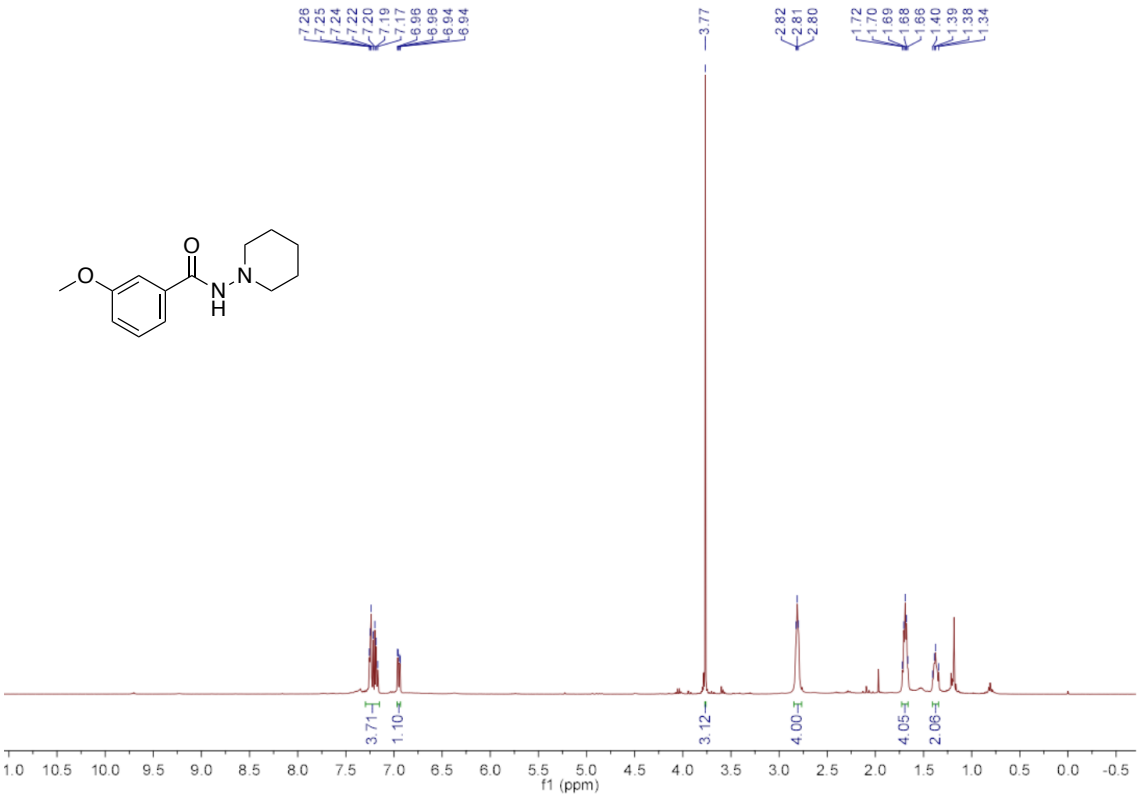
^{1}H NMR spectrum of *N*-(piperidin-1-yl)benzamide **(4d):** (400 MHz, $CDCl_3$)



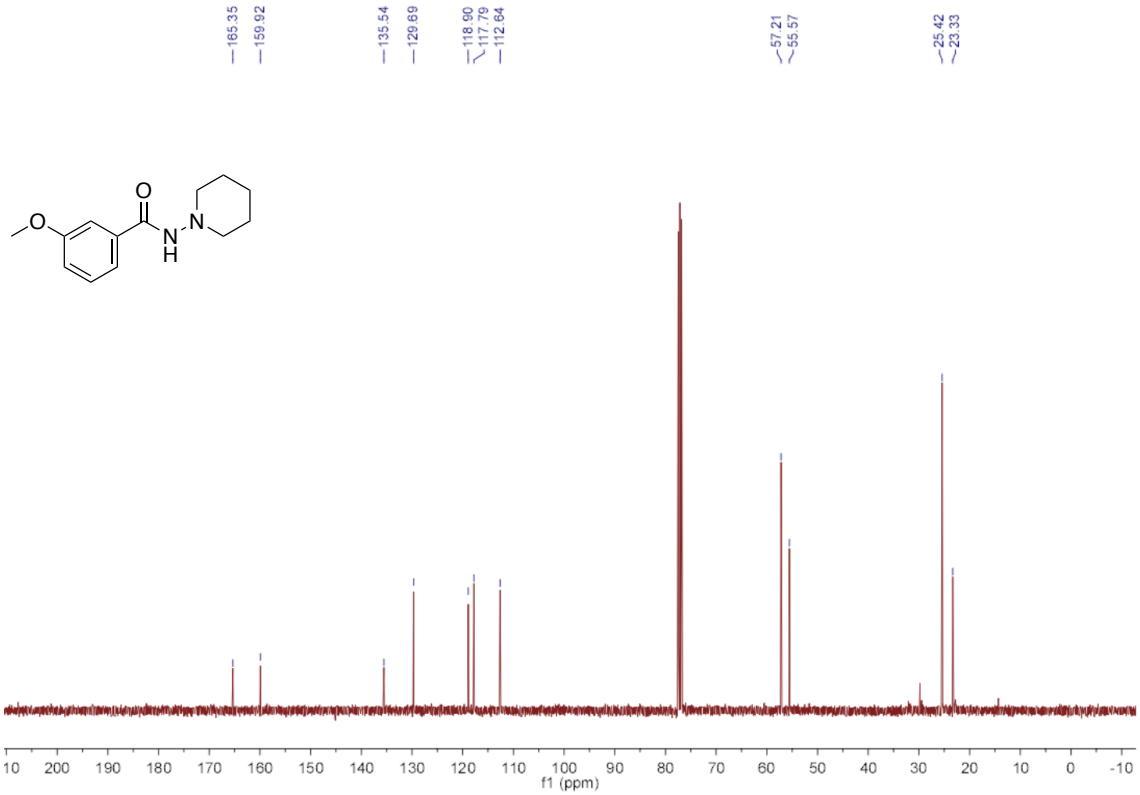
^{13}C NMR spectrum of *N*-(piperidin-1-yl)benzamide **(4d):** (100.6 MHz, $CDCl_3$)



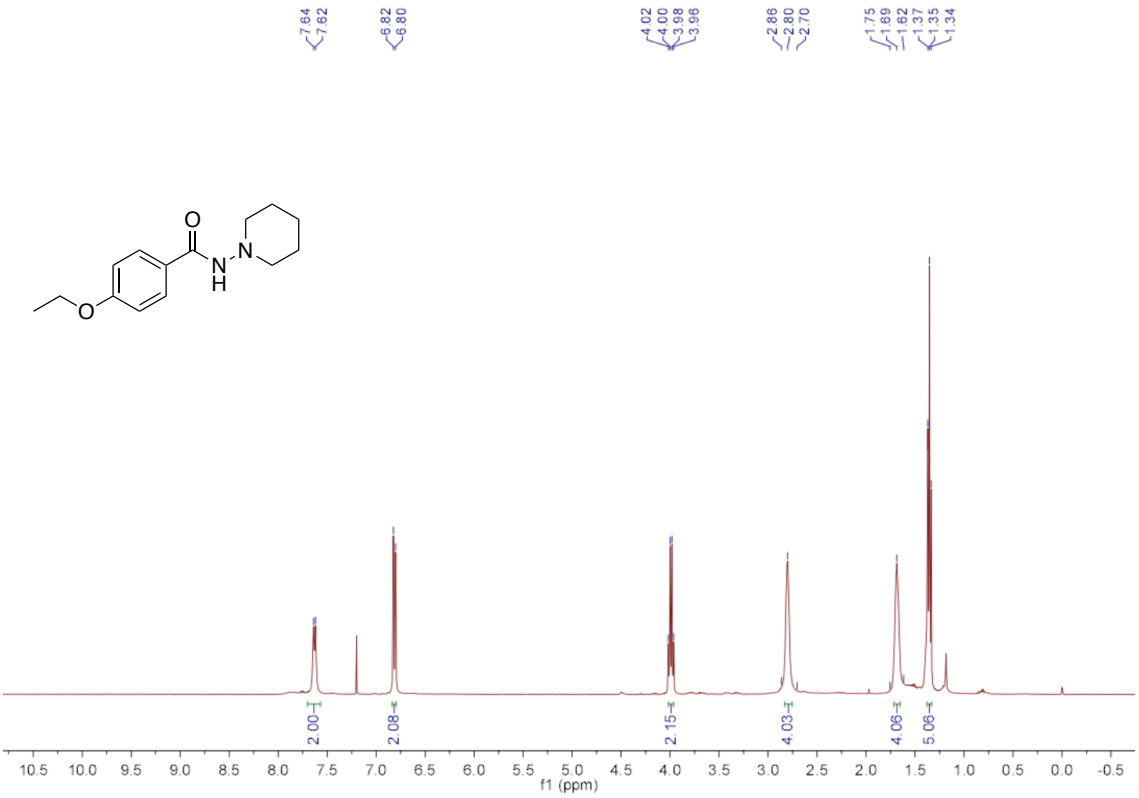
^{1}H NMR spectrum of 3-methoxy-*N*-(piperidin-1-yl)benzamide **(4e):** (400 MHz, $CDCl_3$)



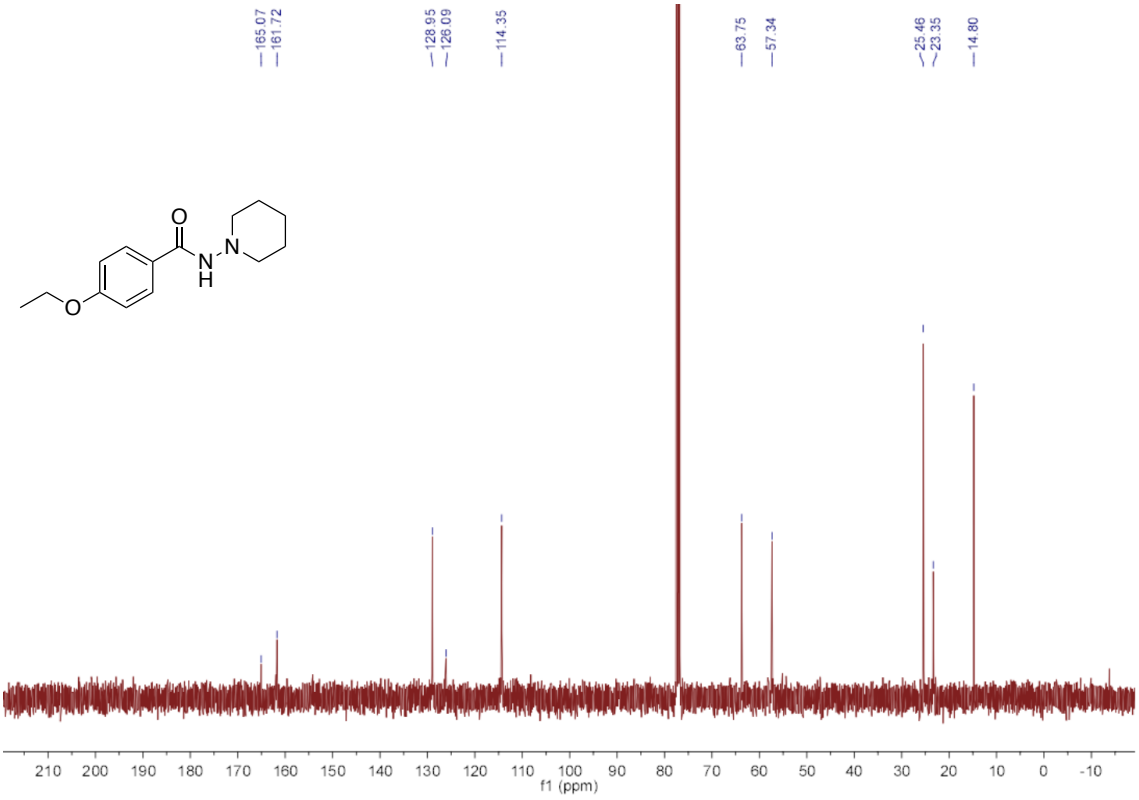
^{13}C NMR spectrum of 3-methoxy-*N*-(piperidin-1-yl)benzamide **(4e):** (100.6 MHz, $CDCl_3$)



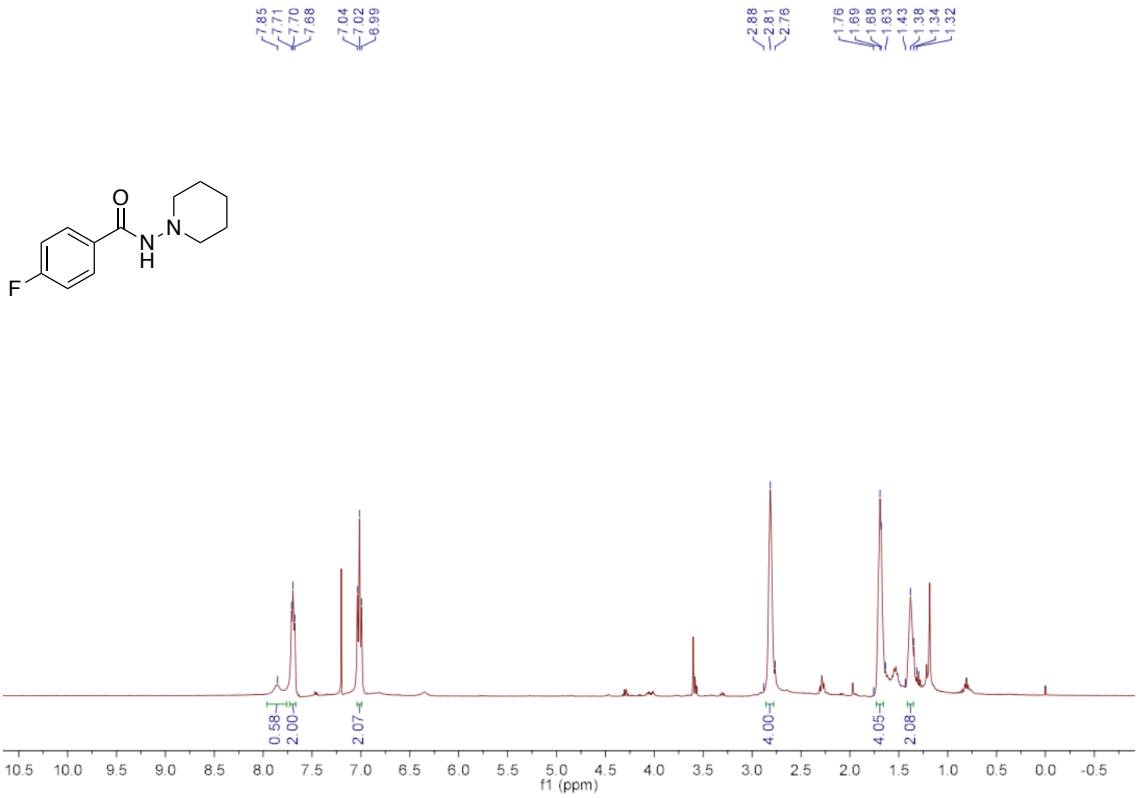
^{1}H NMR spectrum of 4-ethoxy-*N*-(piperidin-1-yl)benzamide **(4f):** (400 MHz, $CDCl_3$)



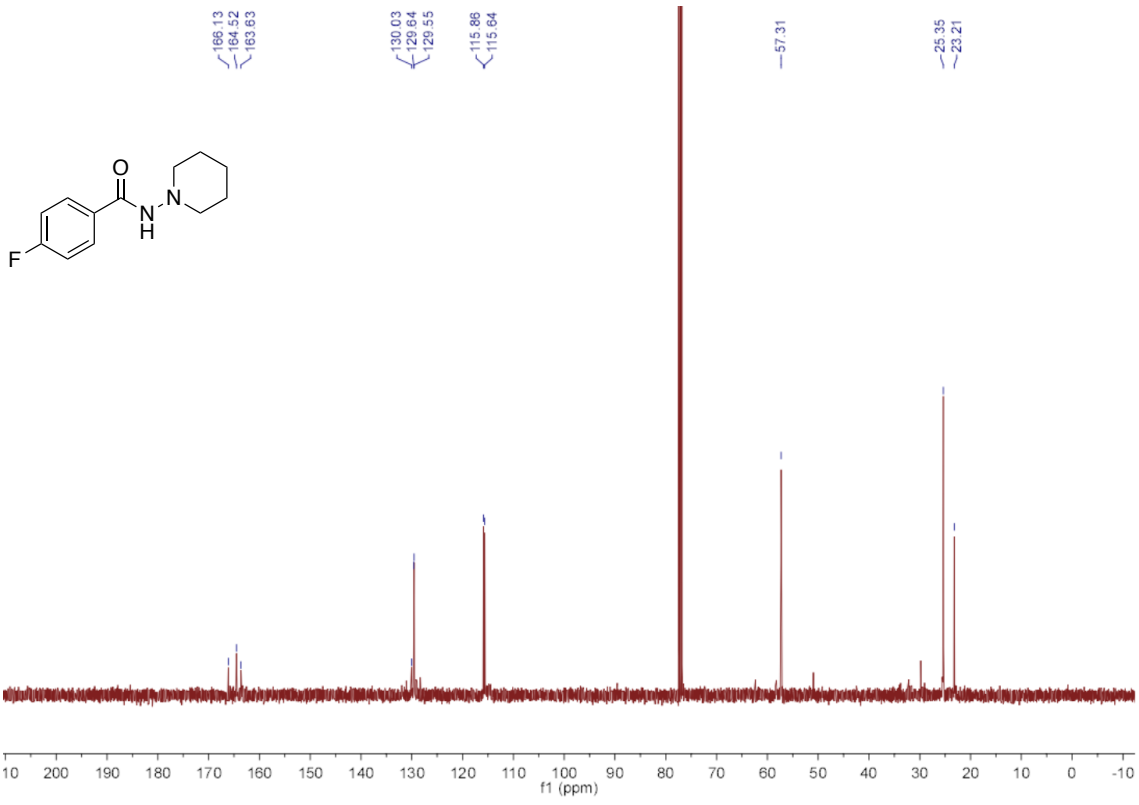
^{13}C NMR spectrum of 4-ethoxy-*N*-(piperidin-1-yl)benzamide **(4f):** (100.6 MHz, $CDCl_3$)



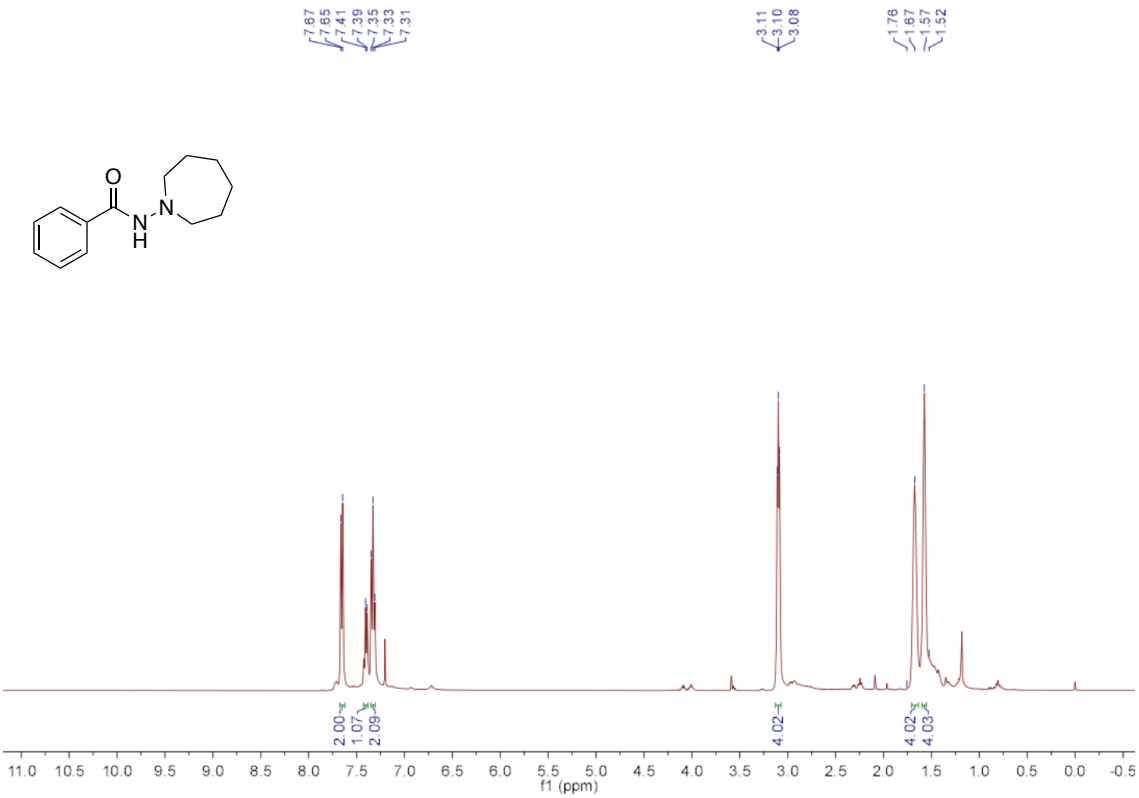
^{1}H NMR spectrum of 4-fluoro-*N*-(piperidin-1-yl)benzamide **(4g):** (400 MHz, $CDCl_3$)



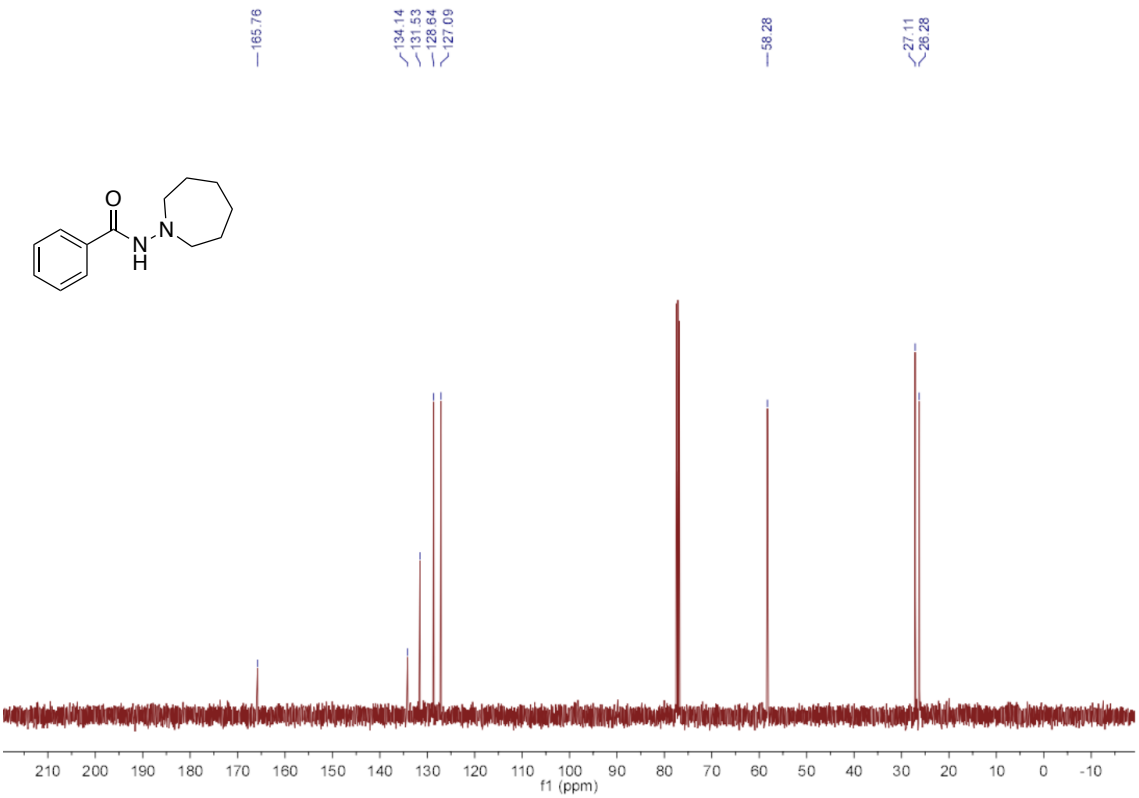
^{13}C NMR spectrum of 4-fluoro-*N*-(piperidin-1-yl)benzamide **(4g):** (100.6 MHz, $CDCl_3$)



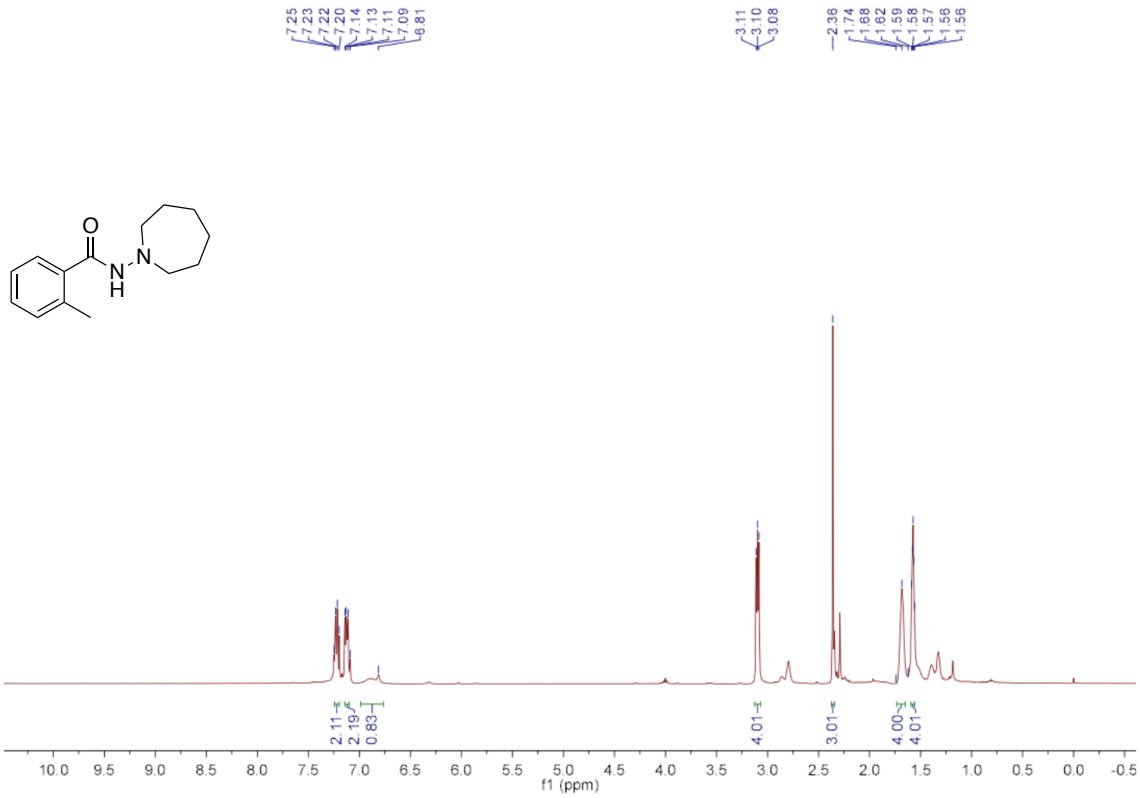
^{1}H NMR spectrum of *N*-(azepan-1-yl)benzamide **(4h):** (400 MHz, $CDCl_3$)



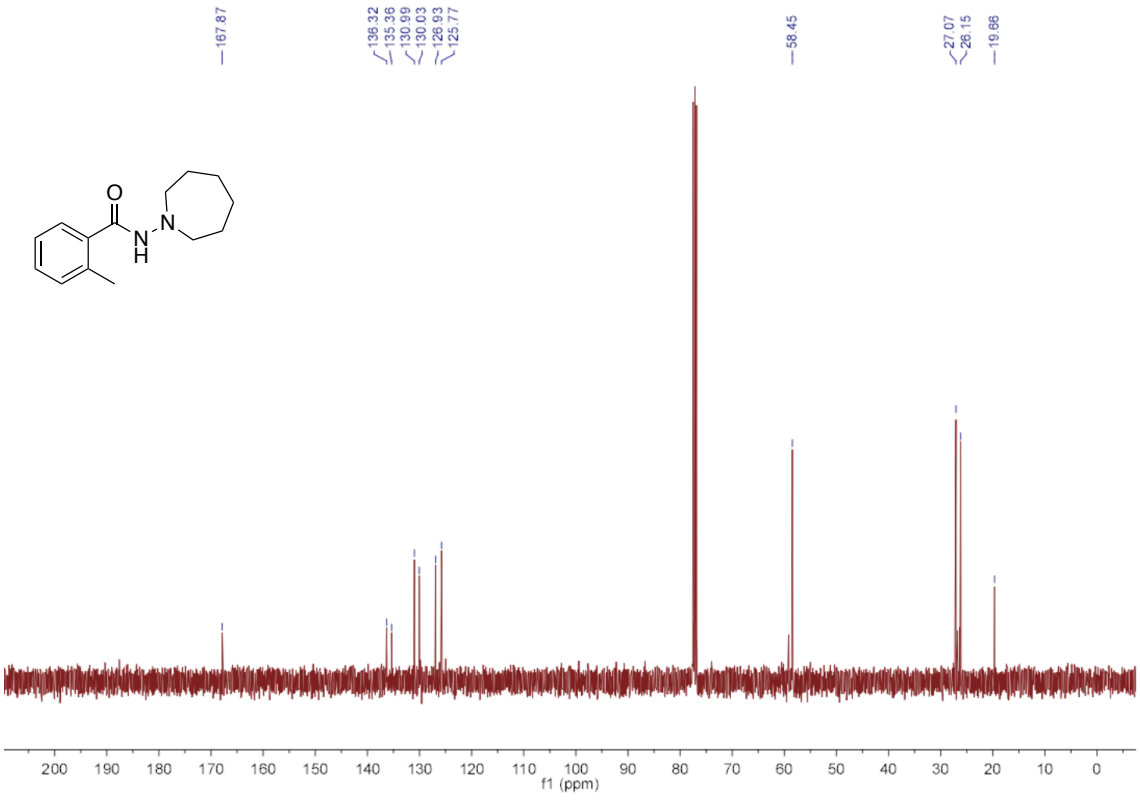
^{13}C NMR spectrum of *N*-(azepan-1-yl)benzamide **(4h):** (100.6 MHz, $CDCl_3$)



^{1}H NMR spectrum of *N*-(azepan-1-yl)-2-methylbenzamide **(4i):** (400 MHz, $CDCl_3$)



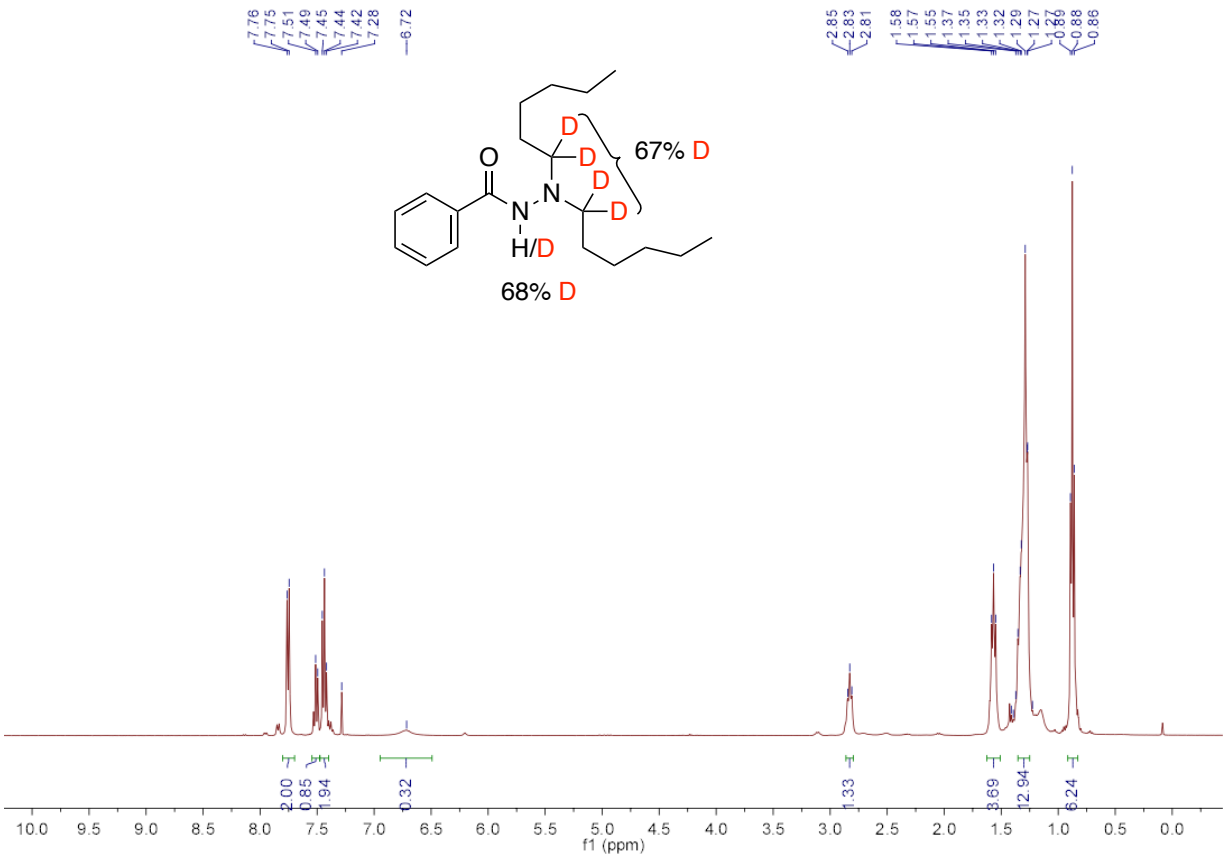
^{13}C NMR spectrum of *N*-(azepan-1-yl)-2-methylbenzamide **(4i):** (100.6 MHz, $CDCl_3$)



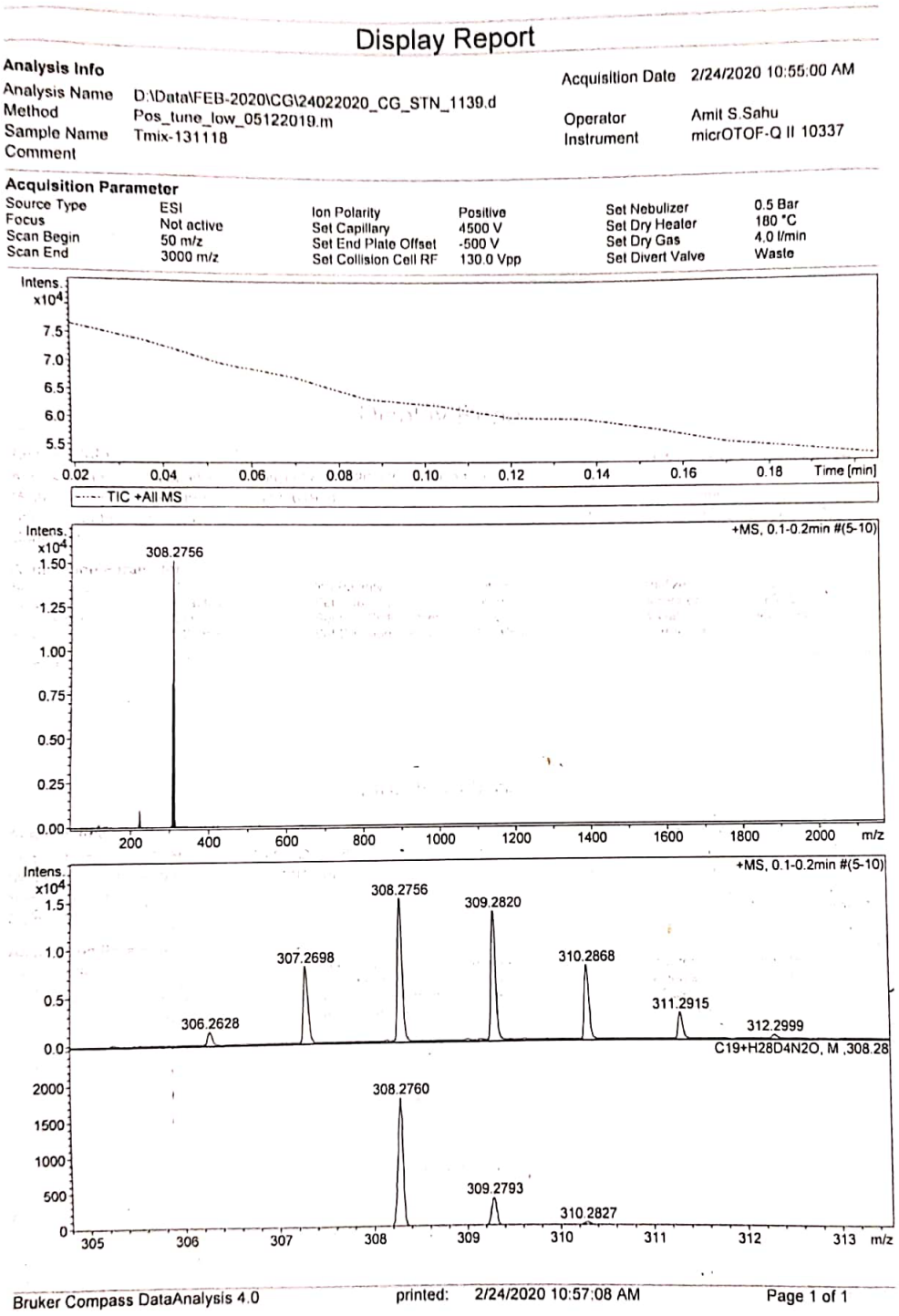
Deuterium Labeling Experiment for *N,N*-Dialkylation of Acylhydrazides Using Alcohols:

An oven-dried Schlenk flask (25 mL) was equipped with a stir bar, catalyst **1** (0.02 mmol), KO^{t}Bu (0.05 mmol), benzohydrazide (0.5 mmol, 1 equiv), 1-hexanol-D3 (1.1 mmol, 2.2 equiv) and dry toluene (2 mL) under nitrogen atmosphere in a glove box. The flask was taken out of the glove box, equipped with a condenser and the solution was heated at 135 °C (oil bath temperature) with stirring in an open system under a flow of argon for 24 h. The completion of the reaction was monitored using TLC analysis. After 24 hours the reaction was stopped and cooled to room temperature. Further, the solvent was evaporated and crude reaction mixture was purified by column chromatography over silica-gel (100-200 mesh) using ethyl acetate / hexane mixture as an eluent (deactivated silica gel by Et_3N). Product **5a** is isolated 90% yield.

^{1}H NMR of the product **5a:** (400 MHz, $CDCl_3$)



HRMS spectrum of the product **5a:**



References:

1) Yu, B.; Chen, Y.; Hong, M.; Duan, P.; Gan, S.; Chao, H.; Zhao, Z.; Zhao, J. Rhodium-Catalyzed C–H Activation of Hydrazines Leads to Isoquinolones with Tunable Aggregation-Induced Emission Properties. *Chem. Commun.* **2015,** *51***,** 14365-14368.