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SUPPORTING INFORMATION

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General Considerations: All reactions were performed under an atmosphere of argon by using standard Schlenk or dry box techniques; solvents were dried over Na metal, or CaH_2. Reagents were of analytical grade, obtained from commercial suppliers and dried over 4Å molecular sieves. ^{1}H, and ^{13}C, ^{19}F NMR spectra were obtained with a Bruker Advance 300 MHz, and a Varian INOVA 500 MHz spectrometer. Chemical shifts (δ) were reported in parts per million (ppm) relative to TMS, and were referenced to the residual solvent peak. NMR multiplicities are abbreviated as follows: s = singlet, d = doublet, t = triplet, q = quartet, quin = quintet, sex =sextet, m = multiplet, br = broad signal. Melting points were measured with a MEL-TEMP apparatus. High-resolution mass spectrometry data was collected on an Agilent 6230 TOF-MS. IR spectra were recorded on thin films using a Jasco 4100 FTIR and attenuated total reflectance (ATR, 3 mm ZnSe plate).

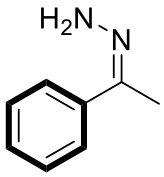
Catalytic procedure for the hydrohydrazination of alkynes: CuCl (0.024 mmol), KB_{Ar}^{F} (B_{Ar}^{F}: $B[C_6F_5]_4^-$) (0.024 mmol), and C_6D_6 (0.5 mL) were added to a J-Young NMR tube. Subsequently, the standard (hexamethylbenzene, 0.081 mmol), the desired alkyne (0.48 mmol), and the appropriate anhydrous hydrazine or amine (0.53 mmol) were added. Then, the tube was heated to 100 ^{o}C for the appropriate length of time. The reaction was monitored by ^{1}H NMR spectroscopy. After completion, the reaction mixture, in an inert argon atmosphere, was passed under a stream of argon through a small pipette column of 5 cm dry neutral alumina [(eluents for parent hydrazine hydrazones: 5 mL 1:10 benzene/pentane then 10 mL ether except in the case of **1g** wherein the eluents were 10 mL 6:4 ether/pentane then 10 mL THF)(eluents for amines and hydrazone derivatives: 15 mL 2:10 ether/pentane)].

Gram-scale synthesis: CuCl (0.49 mmol), KB_{Ar}^{F} (0.49 mmol), and C_6D_6 (10 mL) were added to a schlenk bomb. Subsequently, phenyl acetylene (9.8 mmol), and anhydrous hydrazine (10.8 mmol) were added. Then, the mixture was stirred at 100 ^{o}C for 12 hr. After completion, the reaction mixture, in an inert argon atmosphere, was passed under a stream of argon through a column of 15 cm dry neutral alumina (eluents: 30 mL pentane then 30 mL ether); yield: 1.17 g, 89%.

Catalytic procedure for the hydrohydrazination leading to symmetrical azines: CuCl (0.024 mmol), KB_{Ar}^{F} (0.024 mmol), and C_6D_6 (0.5 mL) were added to a J-Young NMR tube. Subsequently, the standard (hexamethylbenzene, 0.081 mmol), the desired alkyne (0.48 mmol), and the anhydrous hydrazine (0.24 mmol) were added. Then, the tube was heated to 100 ^{o}C for 12 hours. The reaction was monitored by ^{1}H NMR spectroscopy. After completion, the reaction mixture was passed under a stream of argon through a small pipette column of 5 cm neutral alumina (eluent: 20 mL 1:10 benzene/pentane).

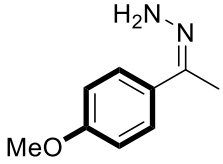
Catalytic procedure for the hydrohydrazination leading to unsymmetrical azines: CuCl (0.024 mmol), KB_{Ar}^{F} (0.024 mmol), and C_6D_6 (0.5 mL) were added to a J-Young NMR tube. Subsequently, the standard (hexamethylbenzene, 0.081 mmol), phenyl acetylene (0.48 mmol), and the anhydrous hydrazine (0.48 mmol) were added. Then, the tube was heated to 100 ^{o}C for 12 hours. Afterwards, the desired alkyne (0.48 mmol) was added to the reaction mixture and heated for another 12 hours at 100 ^{o}C. The reaction was monitored by ^{1}H NMR spectroscopy. After completion, the reaction mixture was passed under a stream of argon through a small pipette column of 5 cm neutral alumina (eluent: 20 mL 1:10 benzene/pentane).

1-phenylhydrazonoethane (1a):



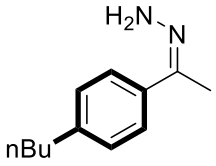
^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.66-7.63 (m, 2H), 7.39-7.30 (m, 3H), 5.35 (br s, 2H), 2.14 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 147.5 (C), 139.5 (C), 128.4 (CH), 128.2 (CH), 125.8 (CH), 11.8 (CH_3); IR (cm^{-1}): 3385 (br), 3302 (br), 2921 (vs) 1590 (vs, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_8H_{11}N_2]^+$, 135.0917; found, 135.0918; yield: 61 mg, 95%.

1-(4-methoxyphenyl)-hydrazonoethane (1b):



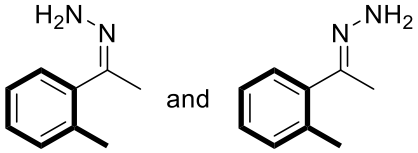
^{1}H NMR (300 MHz, C_6D_6): δ = 7.72 (d, *J* = Hz, 2H), 6.80 (d, *J* = Hz, 2H), 4.71 (br s, 2H), 3.30 (s, 3H), 1.60 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ =160.1 (C), 145.3 (C), 133 (C), 127.1 (CH), 113.9 (CH), 54.8 (CH_3), 10.7 (CH_3); IR: 3393 (s), 3305 (br), 3251 (br), 2836 (m), 1602 (C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_9H_{13}N_2O]^+$, 165.1024; found, 165.1022; yield: 68 mg, 86%.

1-(4-butylphenyl)-hydrazonoethane (1c):



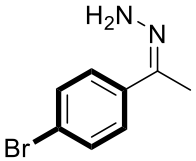
^{1}H NMR (300 MHz, C_6D_6): δ = 7.75 (d, *J* = 4.8 Hz, 2H), 7.08 (d, *J* = 4.8 Hz, 2H), 4.81 (br s, 2H), 2.46 (t, *J* = 4.5 Hz, 2H), 1.62 (s, 3H), 1.49 (quin, *J* = 4.5 Hz, 2H), 1.24 (sex, *J* = 4.5 Hz, 2H), 0.84 (t, *J* = 4.5 Hz, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 145.4 (C), 142.5 (C), 137.8 (C), 128.6 (CH), 125.8 (CH), 35.7 (CH_2), 33.9 (CH_2), 22.7 (CH_2), 14.2 (CH_3), 10.8 (CH_3); IR: 3373 (br m), 3299 (br, m), 3213 (br, m), 2926 (s), 1600 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{12}H_{19}N_2]^+$, 191.1540; found, 191.1543; yield: 79 mg, 87 %.

1-(2-methylphenyl)-hydrazonoethane (1d):



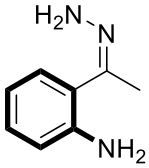
(1:1 mixture of cis/trans isomers) ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.29-7.27 (m, 3H), 7.26-7.15 (m, 4H), 7.08-7.04 (m, 1H), 5.28 (s br, 2H), 4.88 (s br, 2H), 2.35 (s, 3H), 2.26 (s, 3H), 2.14 (s, 3H), 2.11 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 150.0 (C), 149.7 (C), 140.8 (C), 135.5 (C), 135.4 (C), 135.2 (C), 130.7 (CH), 130.7 (CH), 128.8 (CH), 127.9 (CH), 126.8 (CH), 126.7 (CH), 125.8 (CH), 24.5 (CH_3), 20.4 (CH_3), 19.1 (CH_3), 15.7 (CH_3); IR: 3377 (br m), 3209 (br m), 1631 (s, C=N), 1603 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_9H_{13}N_2]^+$, 149.1073; found, 149.1072; yield: 59 mg, 83 %.

1-(4-bromophenyl)-hydrazonoethane (1e):



^{1}H NMR (300 MHz, $CDCl_3$): δ =7.53-7.45 (m, 4H), 5.38 (br s, 2H), 2.10 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 146.1 (C), 138.4 (C), 131.5 (CH), 131.5 (CH), 127.2 (CH), 127.2 (CH), 122.2 (C), 11.5 (CH_3); IR: 3391 (br m), 3219 (br, m), 2917 (m), 1597 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_8H_{10}BrN_2]^+$, 213.0022; found, 213.0021; yield: 75 mg, 73 %.

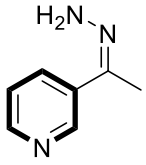
1-(2-aminophenyl)-hydrazonoethane (1f):



^{1}H NMR (300 MHz, C_6D_6): δ = 7.14 (d, *J* = Hz, 1H), 7.02 (t, *J* = , 1H), 6.68 (t, *J* = Hz, 1H), 6.44 (d, *J* = 4.5 Hz, 1H), 5.65 (br s, 2H), 4.46 (br s, 2H), 1.58 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 150.0 (C), 147.1 (C), 128.6 (CH), 128.4

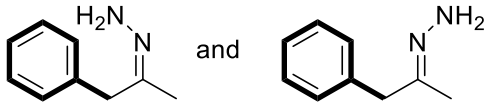
(CH), 121.2 (C), 116.5 (CH), 116.4 (CH), 12.0 (CH_3); IR: 3379 (br s), 3294 (br s), 1605 (vs, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_8H_{12}N_3]^+$, 150.1026; found, 150.1028; yield: 64 mg, 89%.

1-(3-Pyridinyl)-hydrazonoethane (1g):



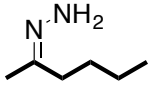
^{1}H NMR (300 MHz, $CDCl_3$): δ = 8.86 (d, *J* = 1.2 Hz, 1H), 8.52 (dd, *J* = 0.9 and 3 Hz, 1H), 7.95 (dt, *J* = 1.2 and 4.8 Hz, 1H), 7.26 (ddd, *J* = 0.6, 3, and 4.8 Hz, 1H), 5.48 (br s, 2H), 2.14 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 149.1 (CH), 147.2 (CH), 144.25 (C), 134.88 (C), 132.7 (CH), 123.3 (CH), 11.5 (CH_3); IR: 3372 (br m), 3310 (br m), 3205 (br m), 1599 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_7H_{10}N_3]^+$, 136.0869; found, 136.0870; yield: 46 mg, 71 %.

1-phenyl-2-hydrazonopropane (1h):



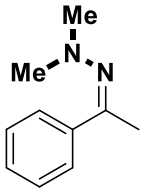
(mixture of isomers); ^{1}H NMR (300 MHz, C_6D_6): (major isomer) δ = 7.13-6.93 (m, 5H), 4.44 (s br, 2H), 3.42 (s, 2H), 1.19 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 149.2 (C), 148.9 (C), 138.7 (C), 136.5 (C), 129.2 (CH), 129.0 (CH), 128.8 (CH), 128.8 (CH), 126.8 (CH), 126.7 (CH), 45.5 (CH_2), 35.2 (CH_2), 23.8 (CH_3), 12.8 (CH_3); IR: 3350 (br m), 3325 (br m), 1642 (s, C=N), 1601 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_9H_{13}N_2]^+$, 149.1073; found, 149.1075; yield: 54 mg, 76 %.

2-hydrazonohexane (1i):



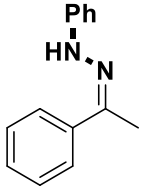
Despite several attempts, the product being very volatile and too unstable we were unable to isolate the pure product by the standard protocol. Hence, purification was performed by slow evaporation of the crude reaction below 5 °C. ^{1}H NMR (500 MHz, C_6D_6): δ = 4.16 (br s, 2H), 2.00 (bt, *J* = 7.5 Hz, 2H), 1.32-1.18 (m, 4H), 0.84 (bt, *J* = 7.5 Hz, 3H). ^{13}C NMR (125 MHz, C_6D_6): δ = 149.6 (C), 38.1 (CH_2), 28.6 (CH_2), 22.3 (CH_2), 13.6 (CH_3), 12.6 (CH_3). IR measurements could not be obtained. HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_6H_{15}N_2]^+$, 115.1230; found, 115.1230. Yield: 22% (NMR yield with internal standard)

1-phenyl-2,2-dimethylhydrazonoethane (1j):



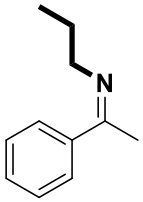
^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.75-7.72 (m, 2H), 7.37-7.35 (m, 3H), 2.6 (s, 6H), 2.36 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 162.2 (C), 139.3 (C), 129.4 (CH), 128.4 (CH), 126.5 (CH), 47.4 (CH_3), 15.7 (CH_3); IR: 2954 (m, NMe_2), 2855 (m, NMe_2), 1607 (m, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{10}H_{15}N_2]^+$, 163.1230; found, 163.1230; yield: 72 mg, 92 %.

1-phenyl-2-phenylhydrazonoethane (1k):



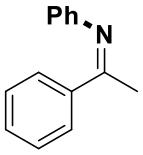
^{1}H NMR (300 MHz, C_6D_6): δ = 7.70 (d, *J* = 4.8 Hz, 1H), 7.21-7.13 (m, 3H), 7.13-7.10 (m, 4H), 6.84 (t, *J* = 4.8 Hz, 1H), 6.77 (br s, 1H), 1.41 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 145.8 (C), 141.6 (C), 139.6 (C), 129.6 (CH) 128.6 (CH), 128.2 (CH), 125.9 (CH), 120.6 (CH), 113.8 (CH), 11.2 (CH_3); IR: 3351 (br, NH), 2920 (m), 1599 (vs, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{14}H_{15}N_2]^+$, 211.1230; found, 211.1228; yield: 81 mg, 80 %.

***N*-(1-phenylethylidene)-1-propanamine (1l):**



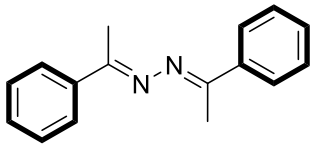
^{1}H NMR (300 MHz, C_6D_6): δ = 7.94-7.91 (m, 2H), 7.22-7.19 (m, 2H 3.24), 7.19-7.15 (m, 1H), (t, *J* = 6.9 Hz, 2H), 1.80 (sex, *J* = 7.2 Hz, 2H), 1.74 (s, 3H), 1.04 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, C_6D_6):163.2 (C), 141.6 (C), 129.4 (CH),128.4 (CH), 127.0 (CH), 54.0 (CH_2), 24.9 (CH_2), 14.5 (CH_3), 12.5 (CH_3); IR: 1633 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{11}H_{16}N]^+$, 162.1277; found, 162.1276; yield: 67 mg, 86 %.

***N*-(1-phenylethylidene)-benzenamine (1m):**



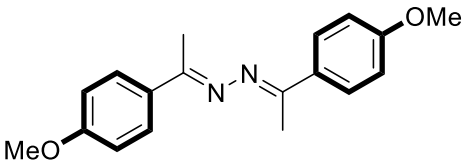
^{1}H NMR (300 MHz, C_6D_6): δ = 7.95-7.93 (m, 2H), 7.19-7.12 (m, 5H), 6.93 (t, *J* = 4.5 Hz, 1H), 6.75-6.73 (m, 2H), 1.81 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 164.4 (C), 152.6 (C), 152.2 (C), 139.9 (CH), 130.5 (CH), 129.3 (CH), 128.1 (CH), 127.7 (CH), 123.3 (CH), 119.7 (CH), 16.8 (CH_3); IR: 1600 (vs, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{16}H_{17}N_2]^+$, 196.1121; found, 196.1117; yield 73 mg, 78%.

1,2-bis-[1-phenylethylidene]hydrazine (2a):



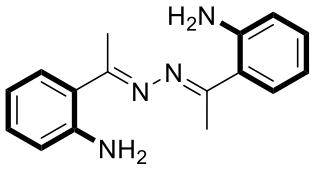
^{1}H NMR (300 MHz, C_6D_6): δ = 7.94-7.92 (m, 4H), 7.21-7.15 (m, 6H), 2.16 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 158.4 (C), 139.2 (C), 129.7 (CH), 128.5 (CH), 127.1 (CH), 14.8 (CH_3); IR: 1603 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{16}H_{17}N_2]^+$, 237.1386; found, 237.1385; yield: 52 mg, 92%.

1,2-bis-[1-(4-methoxyphenyl)ethylidene]hydrazine (2b):



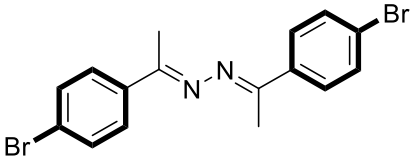
^{1}H NMR (500 MHz, C_6D_6): δ = 7.93 (d, *J* = 9 Hz, 2H), 6.83 (d, *J* = 9 Hz), 3.30 (s, 6H), 2.31 (s, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 161.4 (C), 158.6 (C), 132.1 (C), 128.6 (CH), 114.0 (CH), 54.8 (CH_3), 14.7 (CH_3); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{18}H_{21}N_2O_2]^+$, 297.1598; found, 297.1597; yield: 63 mg, 89 %.

1,2-bis-[1-(2-aminophenyl)ethylidene]hydrazine (2c):



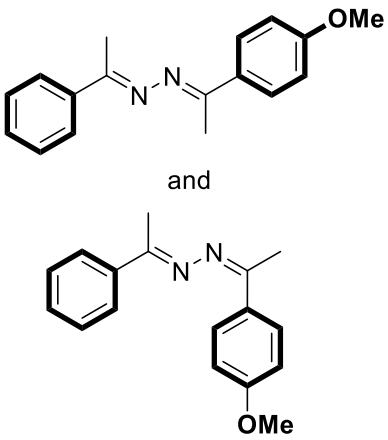
^{1}H NMR (300 MHz, C_6D_6): δ = 7.34 (dd, *J* = 0.9 and 4.8 Hz, 2H), 7.08 (td, *J* = 0.9 Hz and 4.5 Hz, 2H), (td, *J* = 0.9 and 4.5 Hz, 2H), 6.42 (dd, *J* = 0.9 and 4.8 Hz, 2H), 5.99 (br s, 4H), 2.13 (s, 3H); ^{13}C NMR (125 MHz, C_6D_6): δ = 162.2 (C), 148.64 (C), 130.6 (CH), 130.0 (CH), 119.4 (C), 116.8 (CH), 116.2 (CH), 15.8 (CH_3); IR: 3352 (sb), 1613 (s, C=N), 1565 (s); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{16}H_{19}N_4]^+$, 267.1604; found, 267.1607; yield: 58 mg, 90 %.

1,2-bis-[1-(4-bromophenyl)ethylidene]hydrazine (2d):



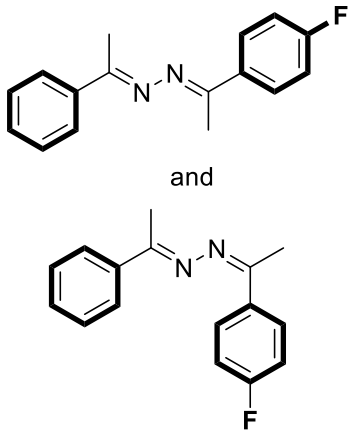
^{1}H NMR (300 MHz, C_6D_6): δ = 7.52 (d, *J* = 8.7 Hz, 4H), 7.32 (d, *J* = 8.7 Hz, 4H), 1.97 (s, 6H); ^{13}C NMR (125 MHz, C_6D_6): δ = 158.1 (C), 137.5 (C), 131.8 (CH), 128.6 (CH), 124.6 (C), 14.6 (CH_3);); IR: 2276 (s), 1618 (m); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{16}H_{15}Br_2N_2]^+$, 392.9597; found, 392.9596; yield: 80 mg, 85 %.

[1-(4-methoxyphenyl)-2-(1-phenyl)]ethylidenehydrazine (2e):



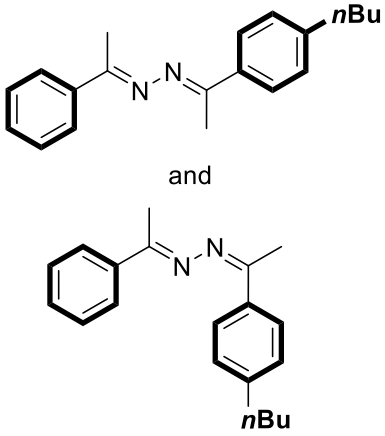
(mixture of isomers); ^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.42-7.41 (m, 3 H), 6.96-6.93 (m, 2H), 3.86 (s, 3H), 2.34-2.31 (m, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 161.0 (C), 161.0 (C), 158.1 (C), 158.0 (C), 157.8 (C), 157.8 (C), 138.8 (C), 138.6 (C), 128.5 (C), 128.5 (C), 128.3 (CH), 128.2 (CH), 126.8 (CH), 126.8 (CH), 113.8 (CH), 113.8 (CH), 55.5 (CH_3), 15.2 (CH_3), 15.1 (CH_3), 15.0 (CH_3), 15.0 (CH_3); IR: 3060 (m), 3026 (m), 1636 (vs, C=N), 1592 (vs); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{17}H_{19}N_2O]^+$, 267.1492; found, 267.1490; yield: 103 mg, 81 %.

[1-(4-fluorophenyl)-2-(1-phenyl)]ethylidenehydrazine (2f):



(1:1 mixture of isomers), ^{1}H NMR (500 MHz, $CDCl_3$): δ = 7.93-7.90 (m, 4H), 7.45-7.43 (m, 3H), 7.13-7.09 (m, 2H), 2.3 (m, 6H); ^{13}C NMR (125 MHz, $CDCl_3$): δ = 164.9 (C), 164.9 (C), 162.9 (C), 162.9 (C), 158.1 (d, $^{1}J_{CF}$ = 242 Hz, C), 157.5 (d, $^{1}J_{CF}$ = 242 Hz, C), 138.5 (C), 138.5 (C), 134.7 (d, $^{4}J_{CF}$ = 12.5 Hz, C), 134.7 (d, $^{4}J_{CF}$ = 12.5 Hz, C), 129.8 (CH), 129.8 (CH), 128.7 (d, $^{2}J_{CF}$ = 33.5 Hz, CH), 128.6 (d, $^{2}J_{CF}$ = 33.5 Hz, CH), 128.5 (CH), 126.7 (CH), 126.7 (CH), 115.4 (d, $^{3}J_{CF}$ = 86 Hz, CH), 115.4 (d, $^{3}J_{CF}$ = 86 Hz, CH), 15.2 (CH_3), 15.2 (CH_3), 15.2 (CH_3), 15.2 (CH_3); ^{19}F{^{1}H} NMR (300 MHz, $CDCl_3$): δ = -111.5 (m, C-F), -111.6 (m, C-F); IR: 1605 (vs, C=N), 1577 (s); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{16}H_{15}FN_2]^+$, 255.1292; found, 255.1294; yield: 92 mg, 76 %.

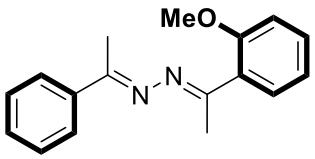
[1-(4-butylphenyl)-2-(1-phenyl)]ethylidenehydrazine (2g):



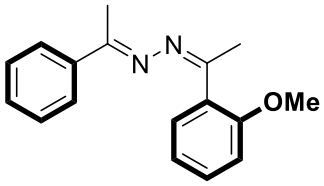
(1:1 mixture of isomers); ^{1}H NMR (300 MHz, C_6D_6): δ = 7.84-7.81 (m, 2H), 7.78-7.76 (m, 2H), 7.34-7.31 (m, 3H), 7.15-7.13 (m, 2H), 2.56 (t, *J* = 7.5 Hz, 2H), 2.23-2.22 (m, 6H), 1.54 (quin, *J* = 7.5 Hz, 2 H), 1.28 (sex, *J* = 7.5 Hz, 2H), 0.85 (t, *J* = 7.5 Hz, 3H); ^{13}C NMR (125 MHz, C_6D_6): 157.9 (C), 157.8, (C), 157.8 (C), 157.8 (C), 144.9 (C), 144.8 (C), 138.6 (C), 138.5 (C), 136.1 (C), 136.0 (C), 129.7 (CH), 129.7 (CH), 128.5 (CH), 128.5 (CH), 128.5 (CH), 128.4 (CH), 126.7 (CH), 126.7 (CH), 126.7 (CH), 126.7 (CH), 35.6 (CH_2), 33.65 (CH_2), 22.5 (CH_2), 15.2 (CH_3), 15.2 (CH_3),

15.1 (CH_3), 15.1 (CH_3), 14.1 (CH_3); IR: 2956 (m), 2927 (m), 1604 (s, C=N), 1565 (s); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{20}H_{25}N_2]^+$, 293.2012; found, 293.2012; yield: 119 mg, 85 %.

[1-(2-methoxyphenyl)-2-(1-phenyl)]ethylidenehydrazine (2h):



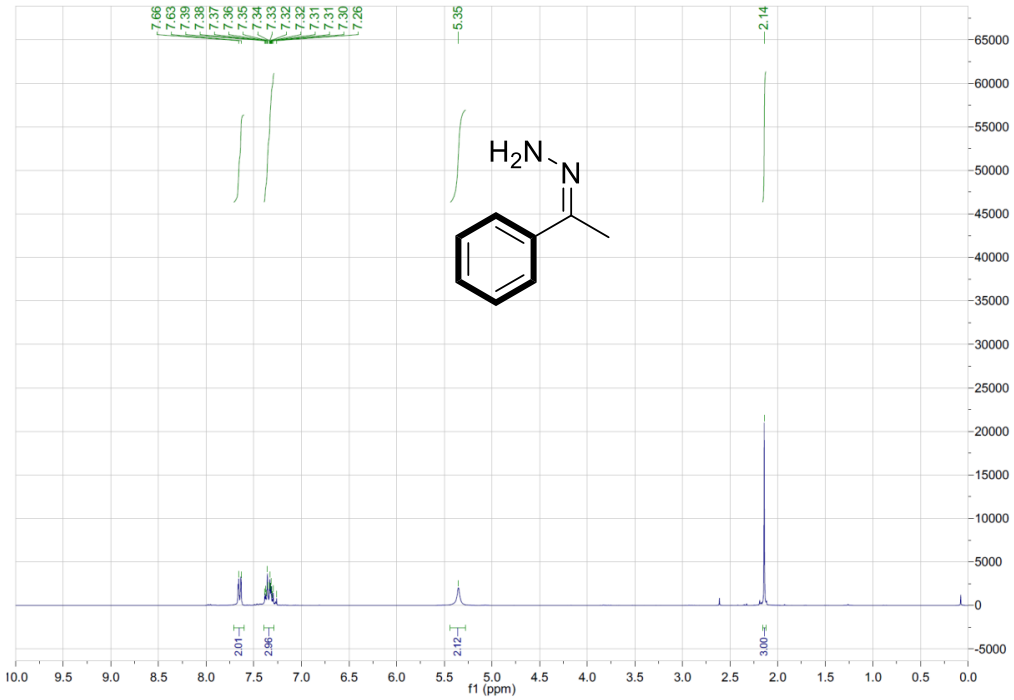
and

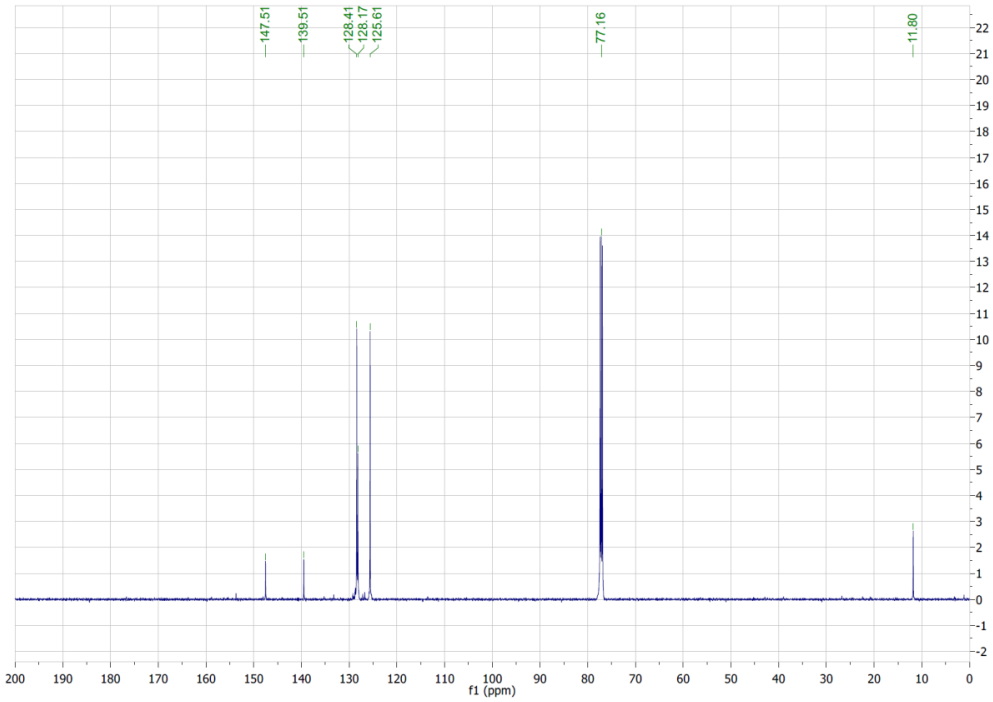


(mixture of isomers); ^1H NMR (300 MHz, $CDCl_3$): δ = 7.95-7.92 (m, 2H), 7.58-7.56 (m, 1H), 7.47-7.43 (m, 2H), 7.40-7.37 (m, 1H), 7.05-6.96 (m, 2H), 3.89 (s, 3H), 2.35-2.25 (m, 6H) ^{13}C NMR (125 MHz, $CDCl_3$): δ = 159.5 (C), 158.7 (C), 157.8 (C), 157.7 (C), 157.0 (C), 157.0 (C), 138.6 (C), 138.6 (C), 138.6 (C), 138.6 (C), 130.3 (CH), 130.2 (CH), 129.7 (CH), 129.7 (CH), 129.6 (CH), 129.6 (CH), 128.5 (CH), 128.4 (CH), 126.7 (CH), 120.8 (CH), 120.8 (CH), 111.3 (CH), 111.3 (CH), 55.6 (CH_3), 55.6 (CH_3), 19.1 (CH_3), 18.9 (CH_3), 15.2 (CH_3), 15.0 (CH_3); IR: 1604 (s, C=N); HRMS ESI (m/z): $[M+H]^+$ calcd. for $[C_{17}H_{19}N_2O]^+$, 267.1492; found, 267.1491; yield: 92 mg, 72 %.

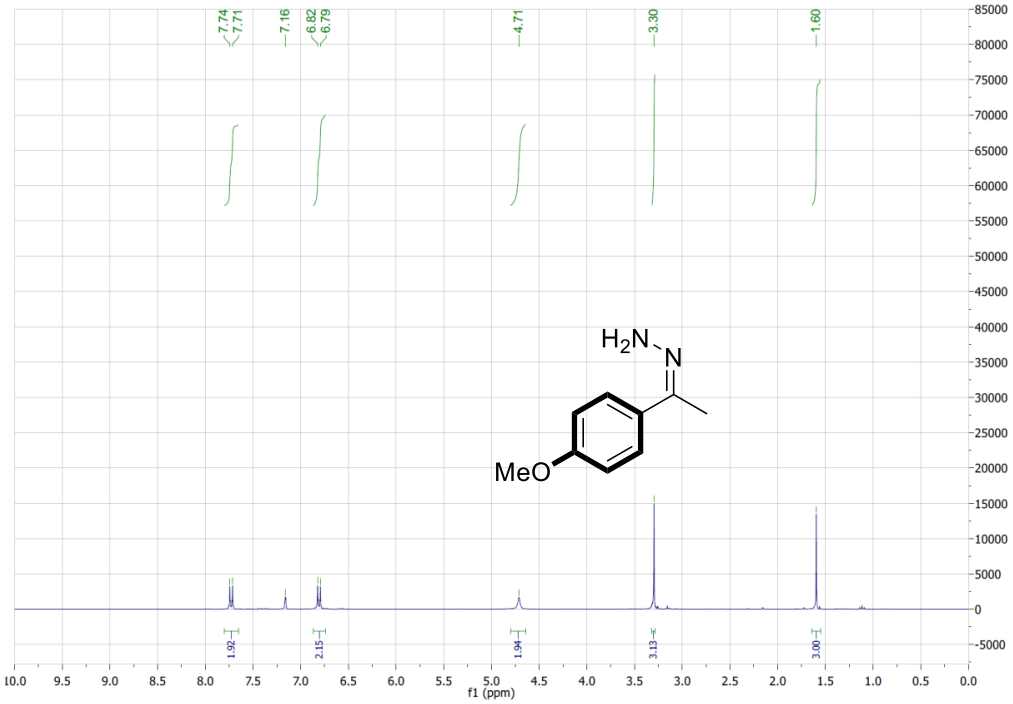
^{1}H and ^{13}C-{^{1}H} NMR spectra for compounds 1a-1i (Table 2):

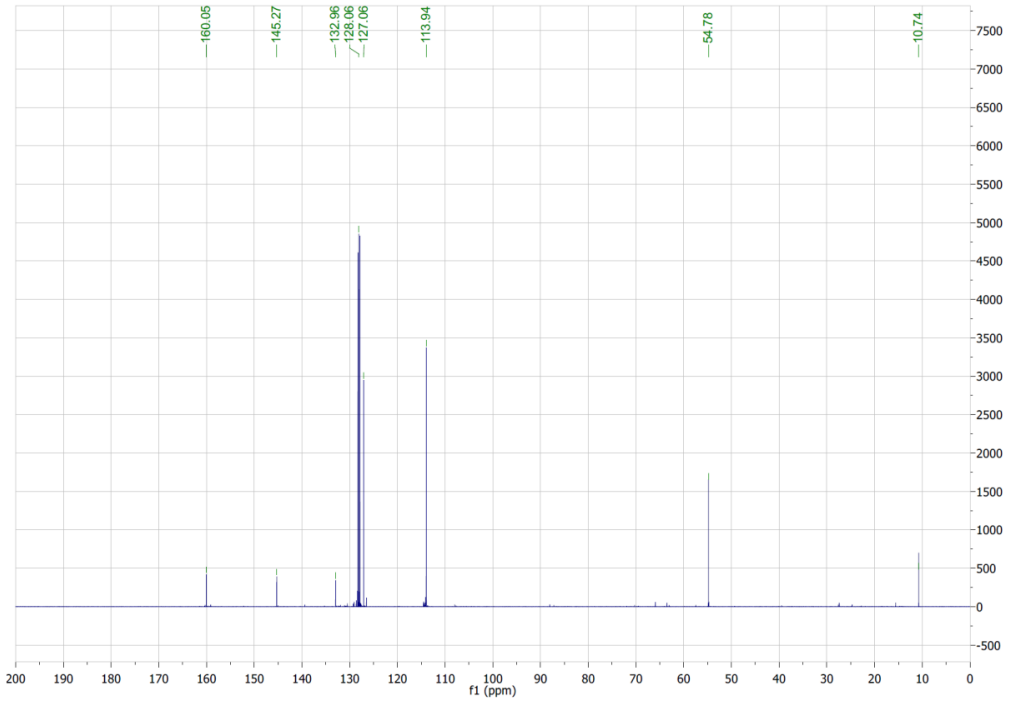
1-phenylhydrazonoethane (1a):



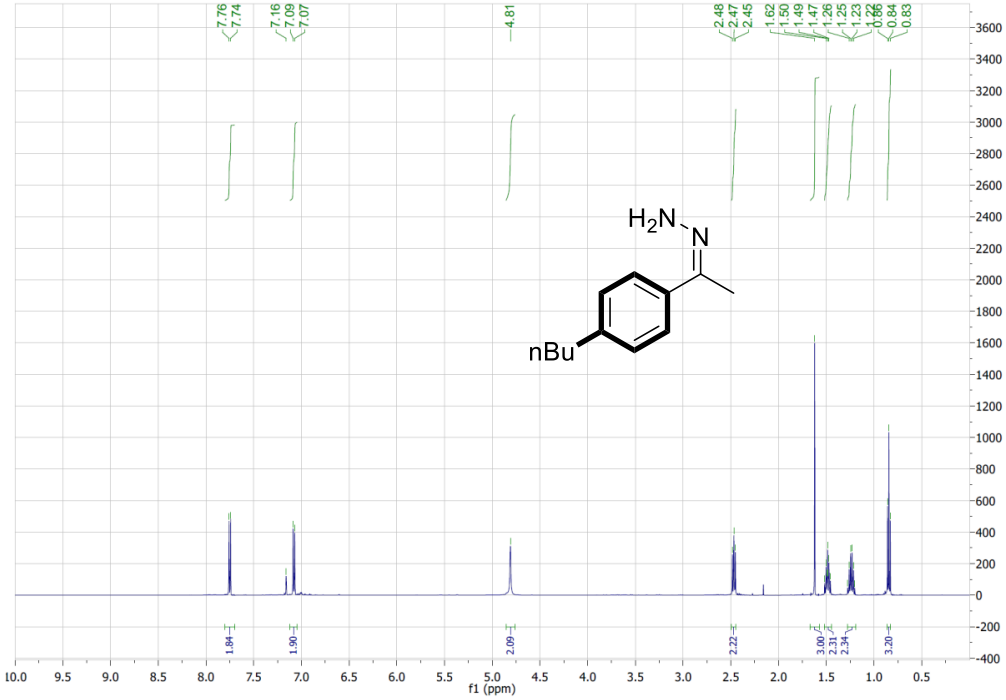


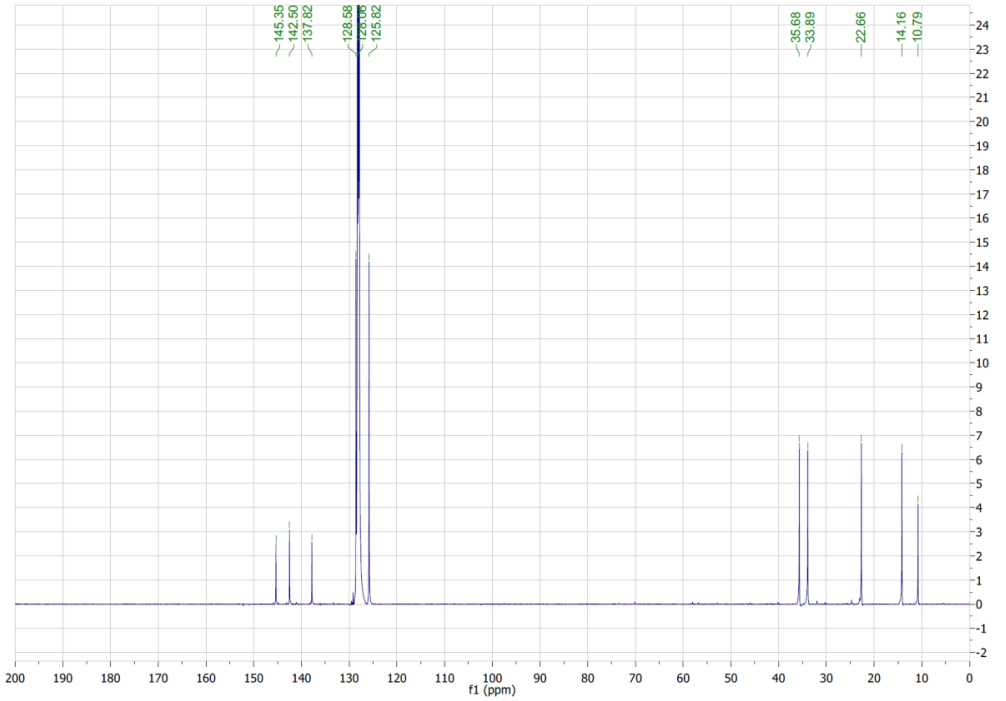
1-(4-methoxyphenyl)-hydrazonoethane (1b):



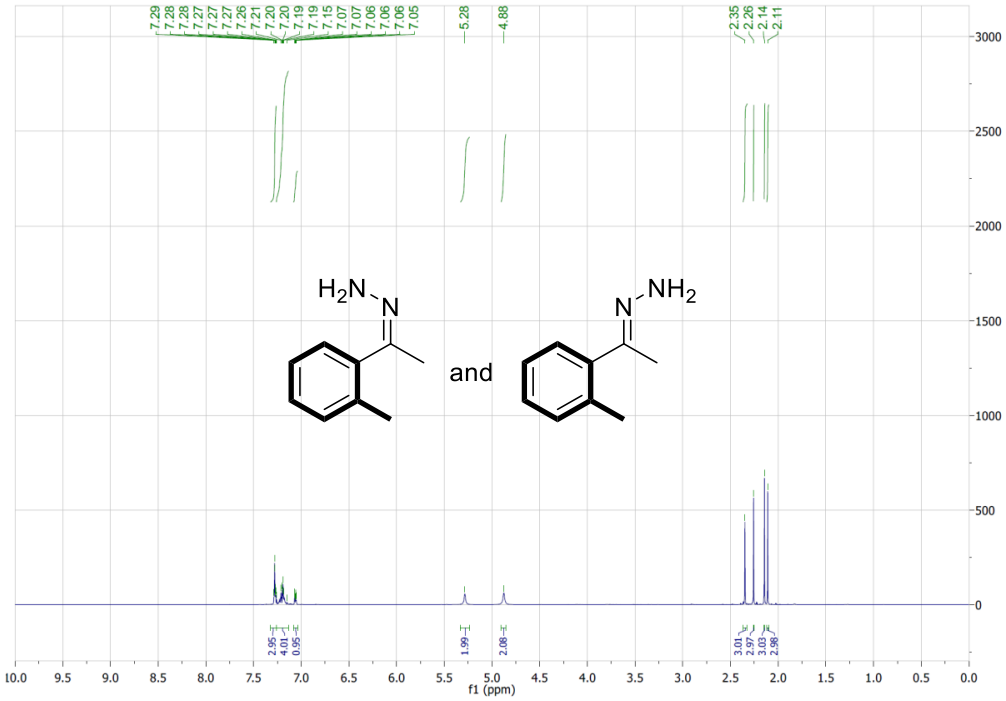


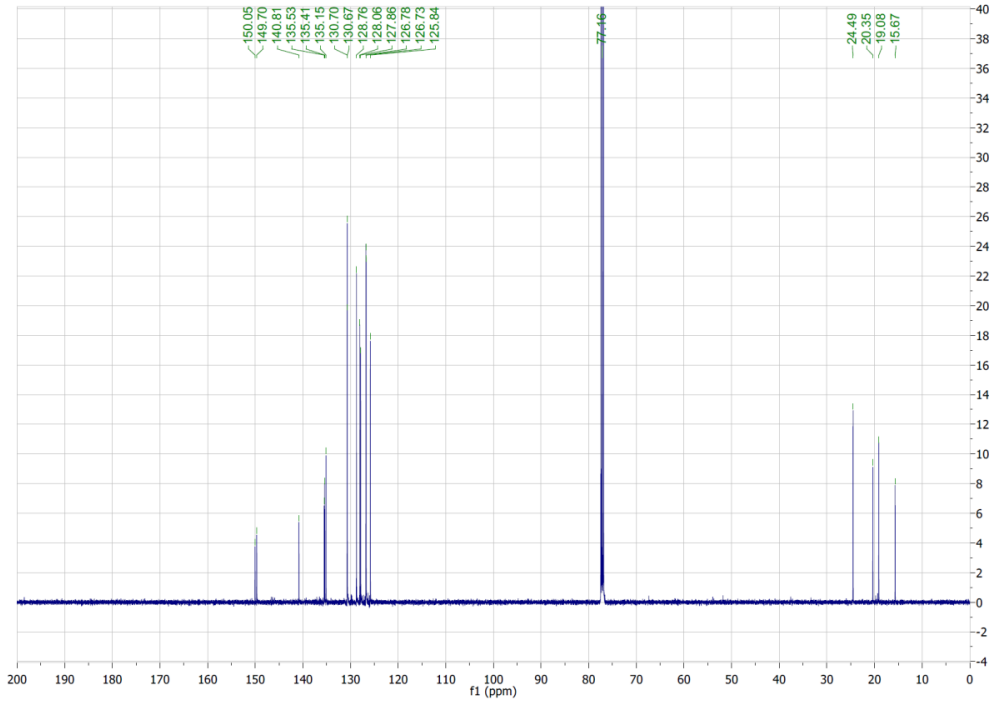
1-(4-butylphenyl)-hydrazonoethane (1c):



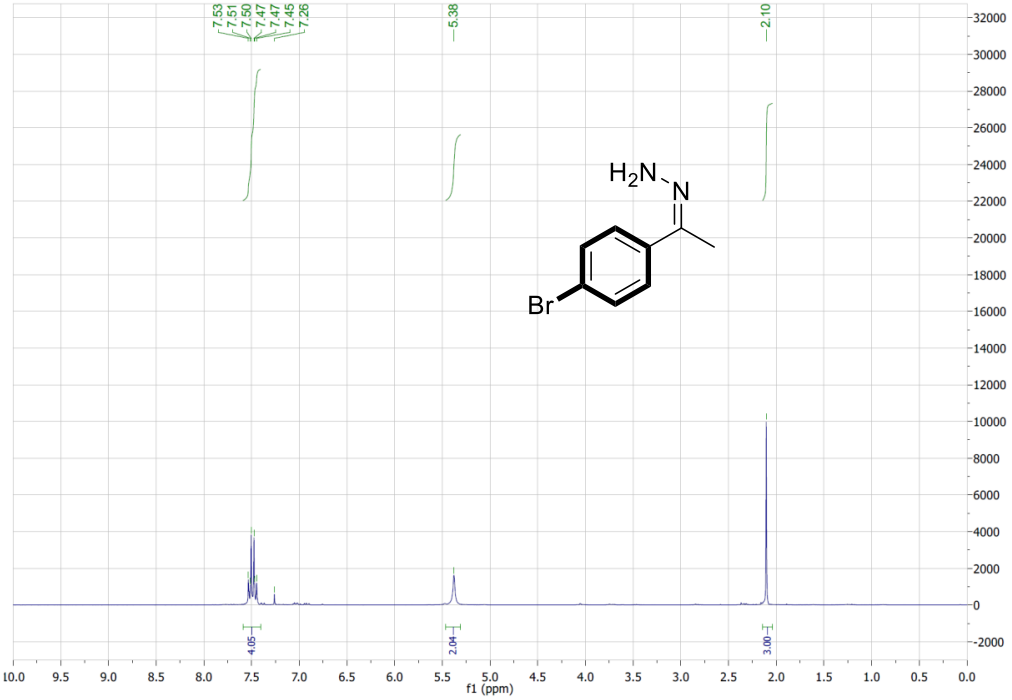


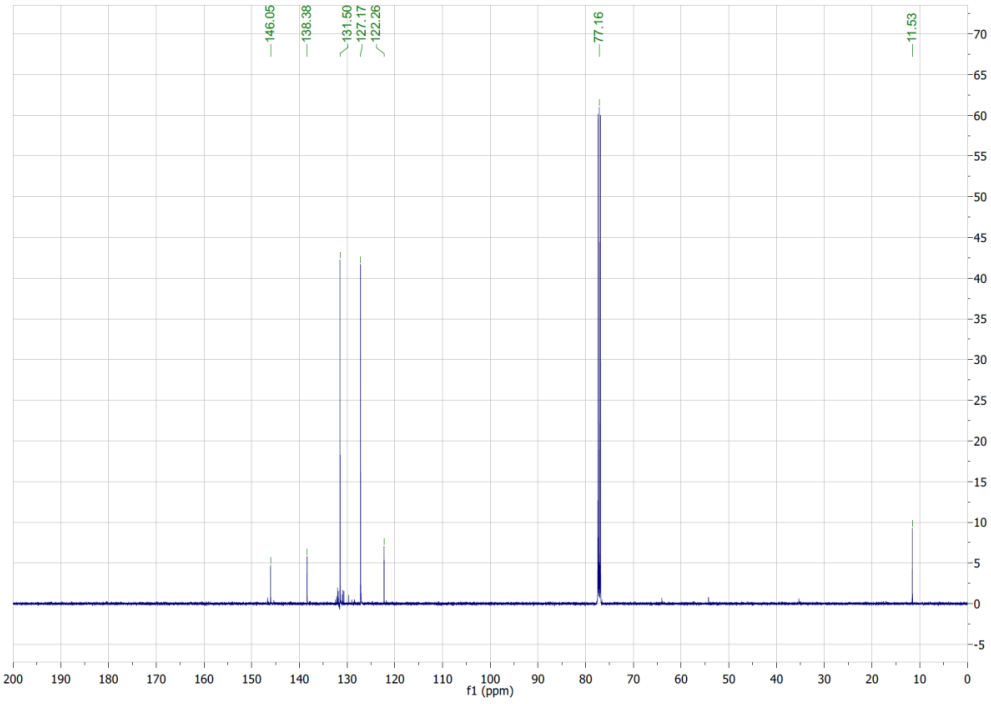
1-(2-methylphenyl)-hydrazonoethane (1d):



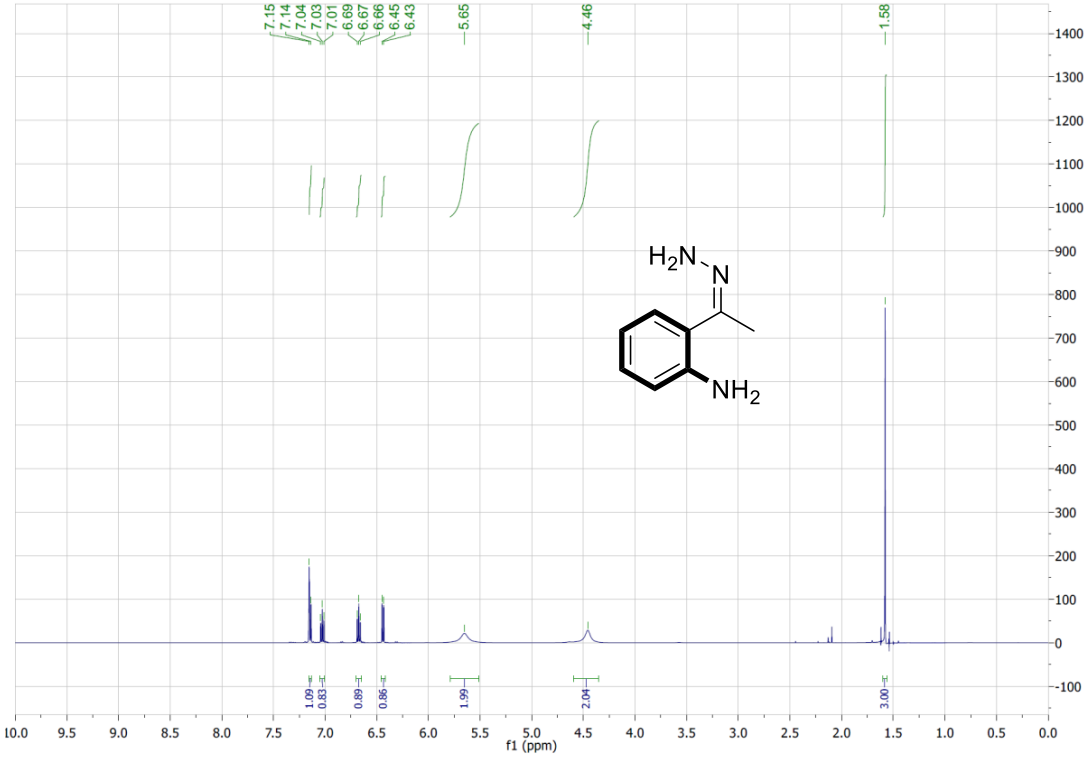


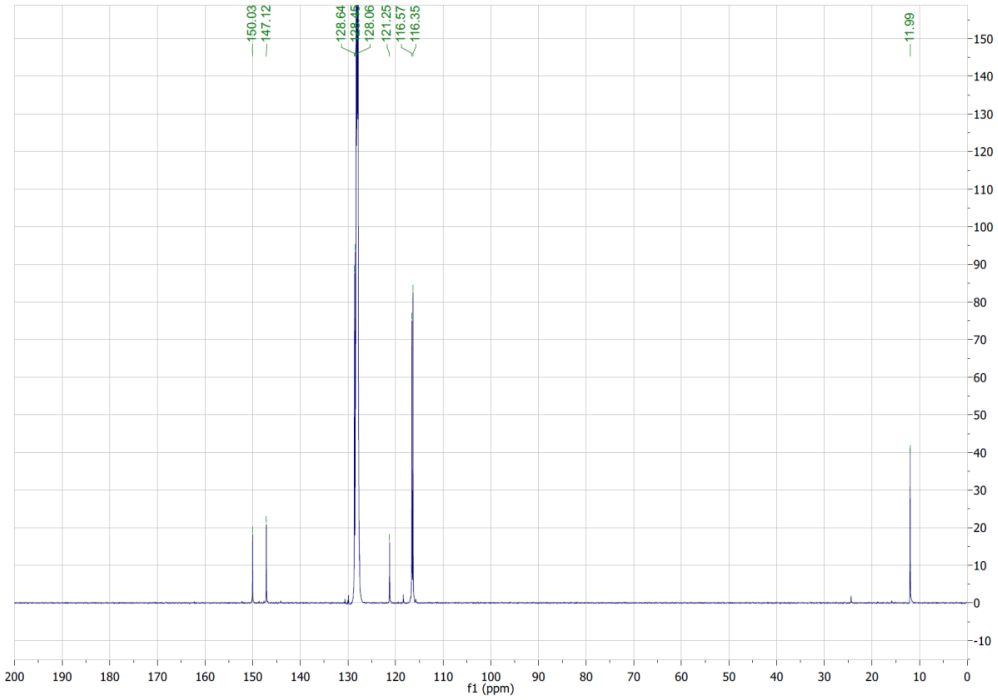
1-(4-bromophenyl)-hydrazonoethane (1e):



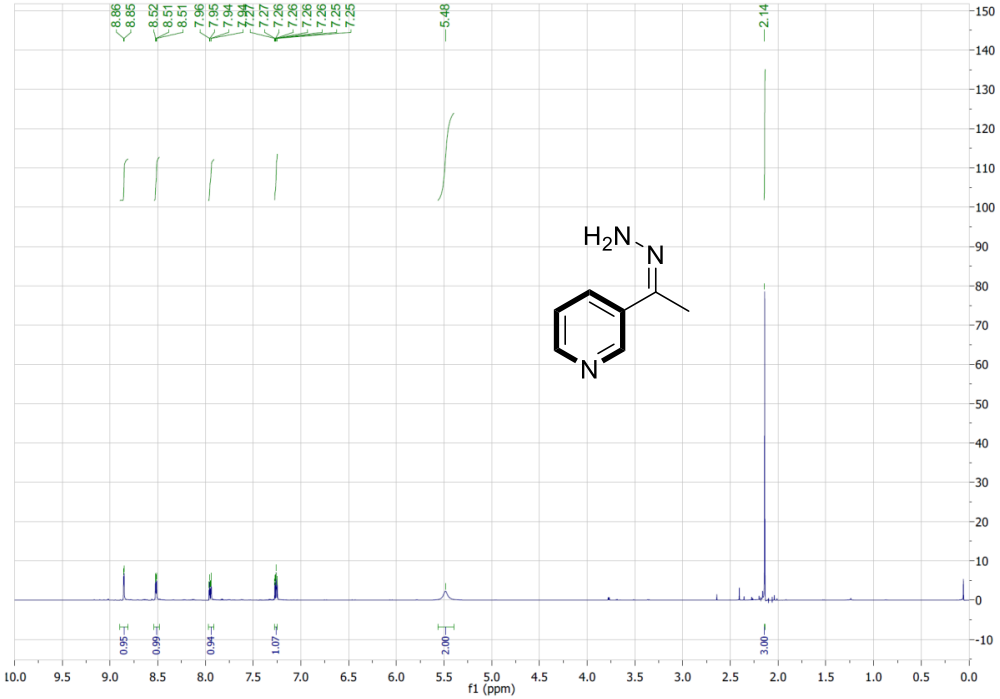


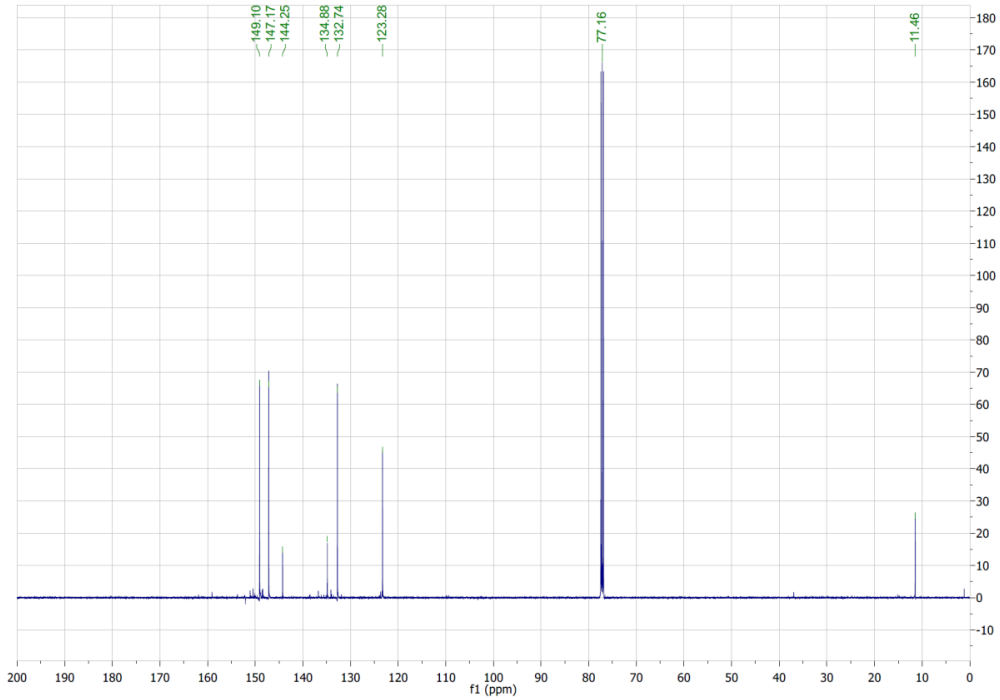
1-(2-aminophenyl)-hydrazonoethane (1f):



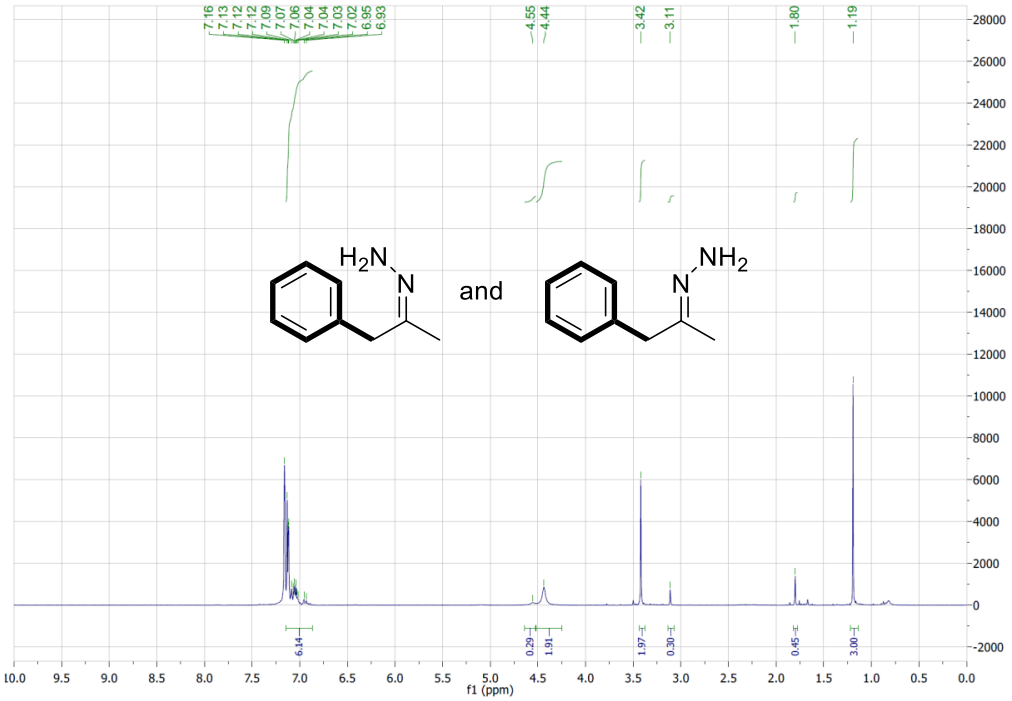


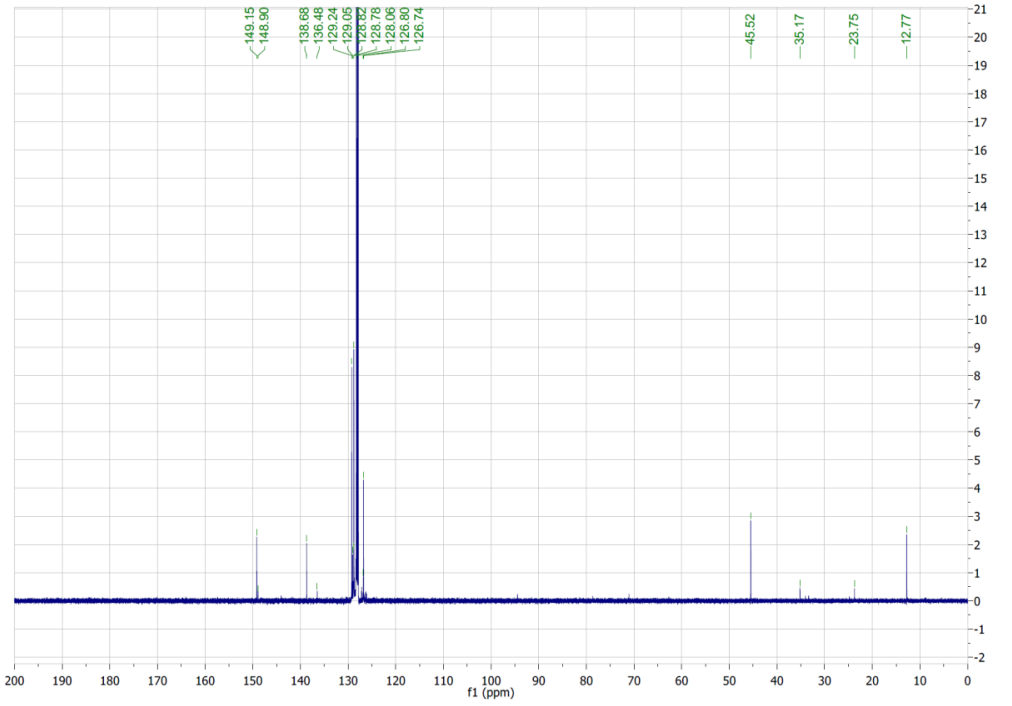
1-(3-Pyridinyl)-hydrazonoethane (1g):



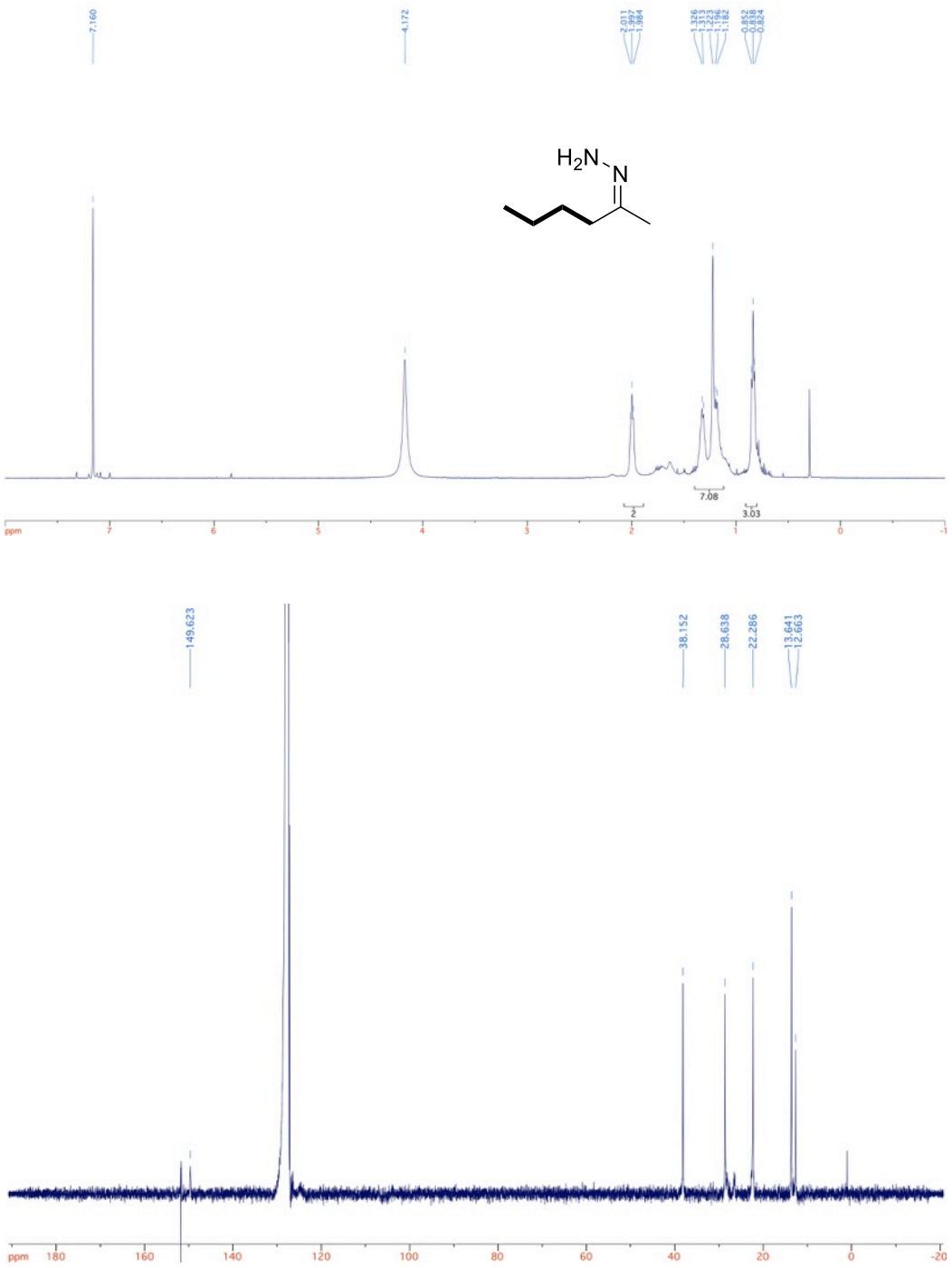


1-phenyl-2-hydrazonopropane (1h):



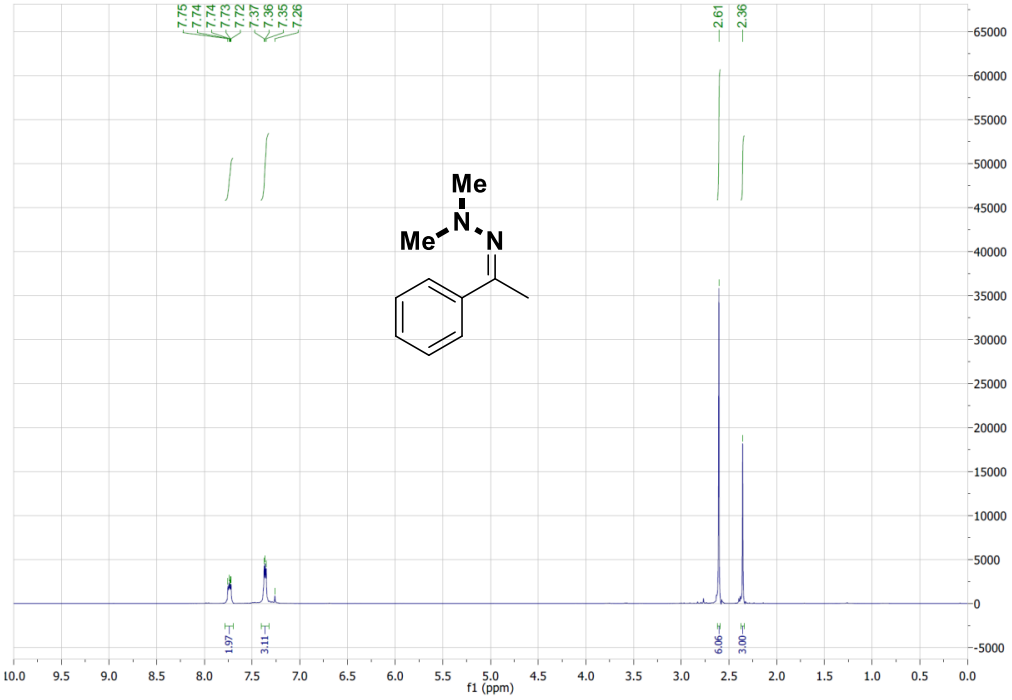


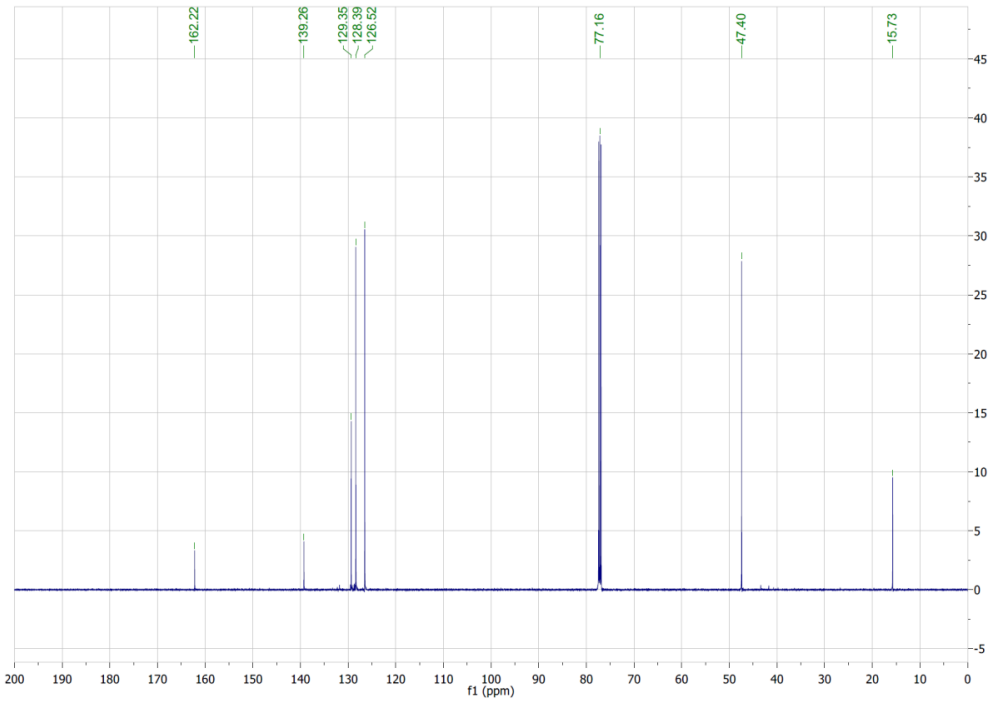
2-hydrazonohexane (1i):



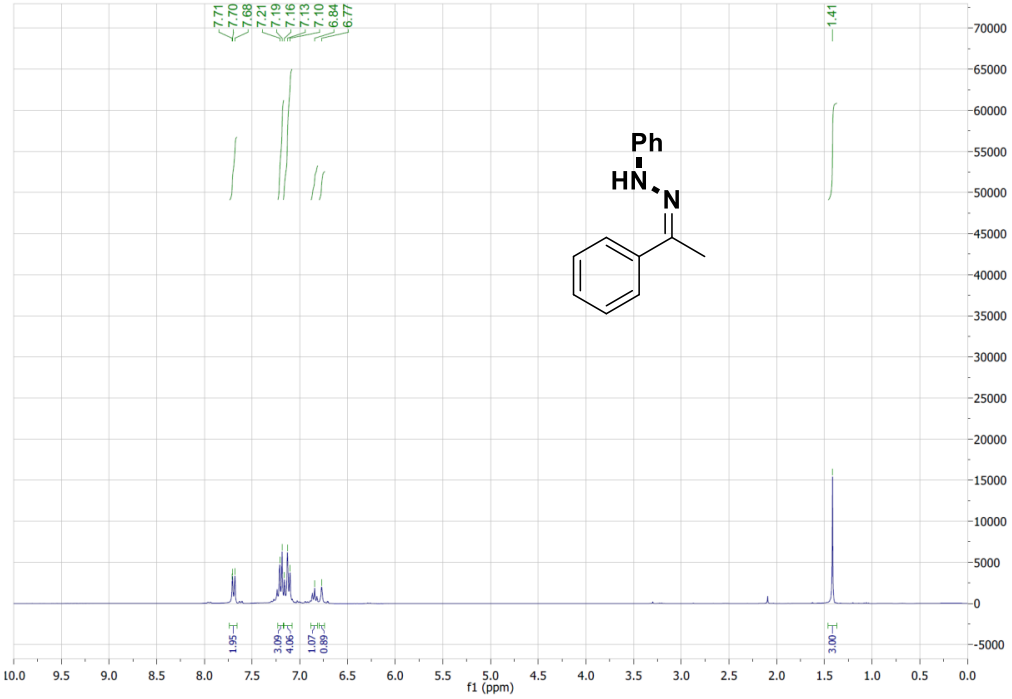
^{1}H and ^{13}C-{^{1}H} NMR spectra for compounds 1j-1m (Table 3):

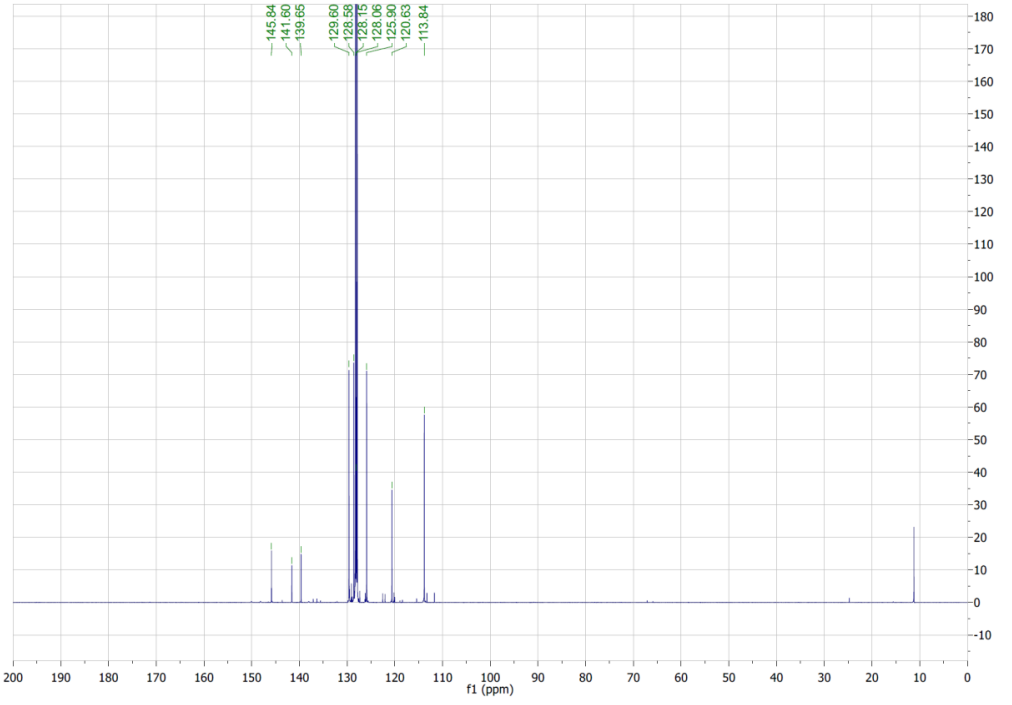
1-phenyl-2,2-dimethylhydrazonoethane (1j):



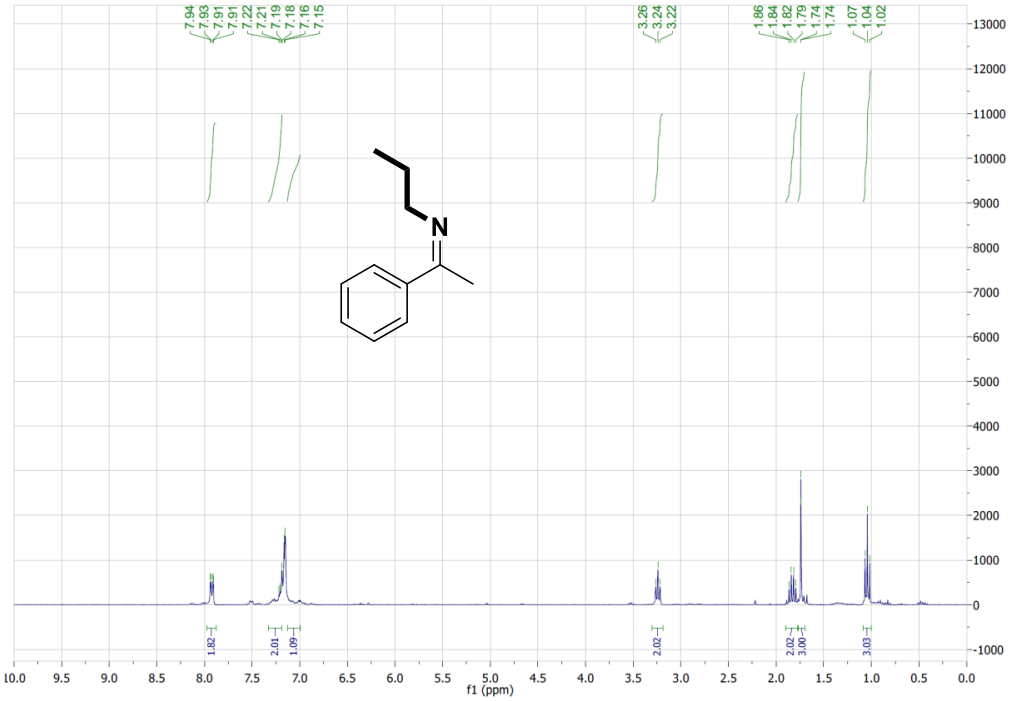


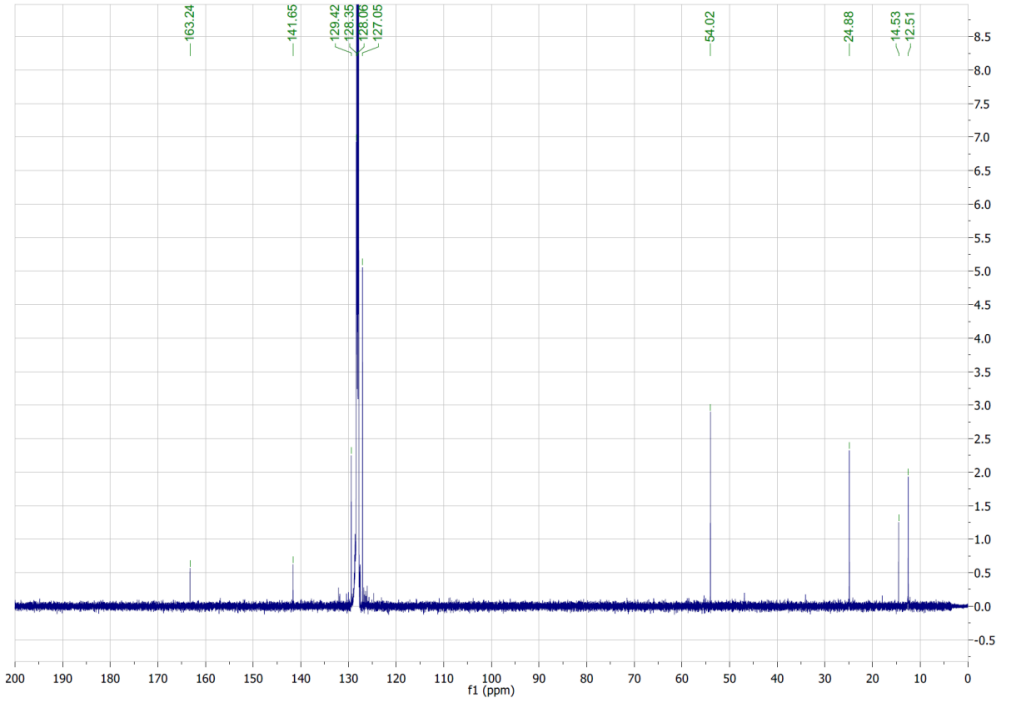
1-phenyl-2-phenylhydrazonoethane (1k):



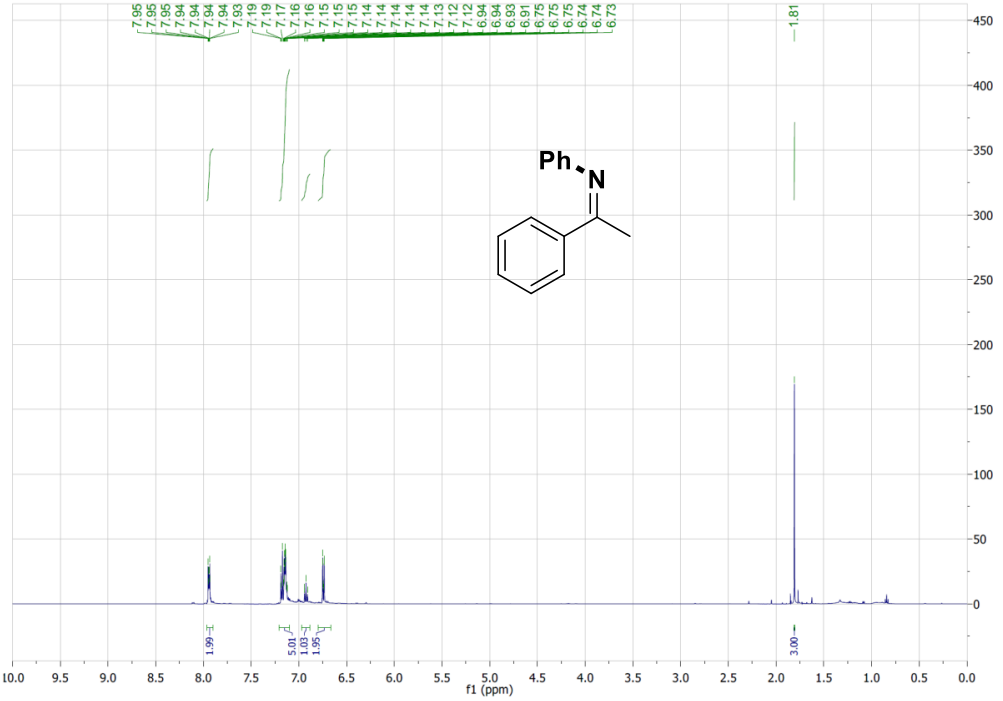


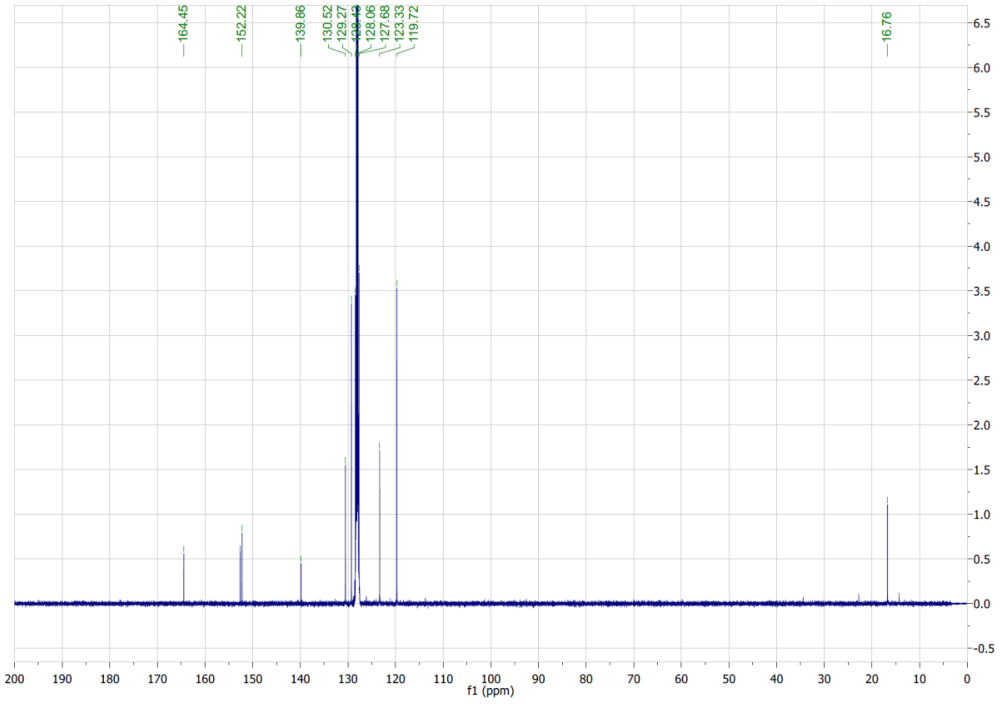
***N*-(1-phenylethylidene)-1-propanamine (1l):**





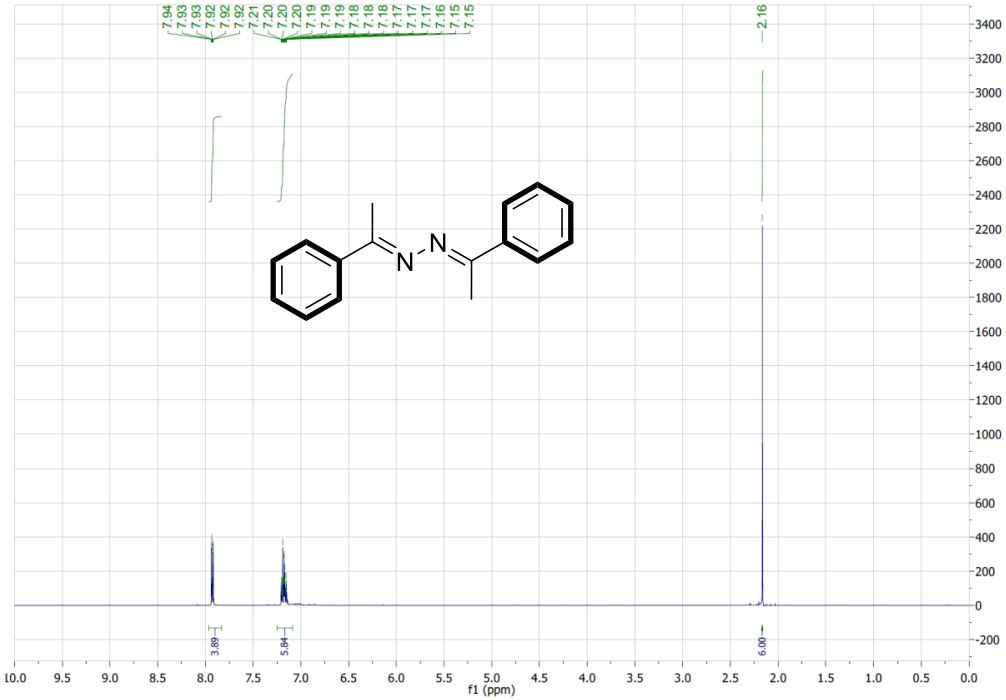
***N*-(1-phenylethylidene)-benzenamine (1m):**

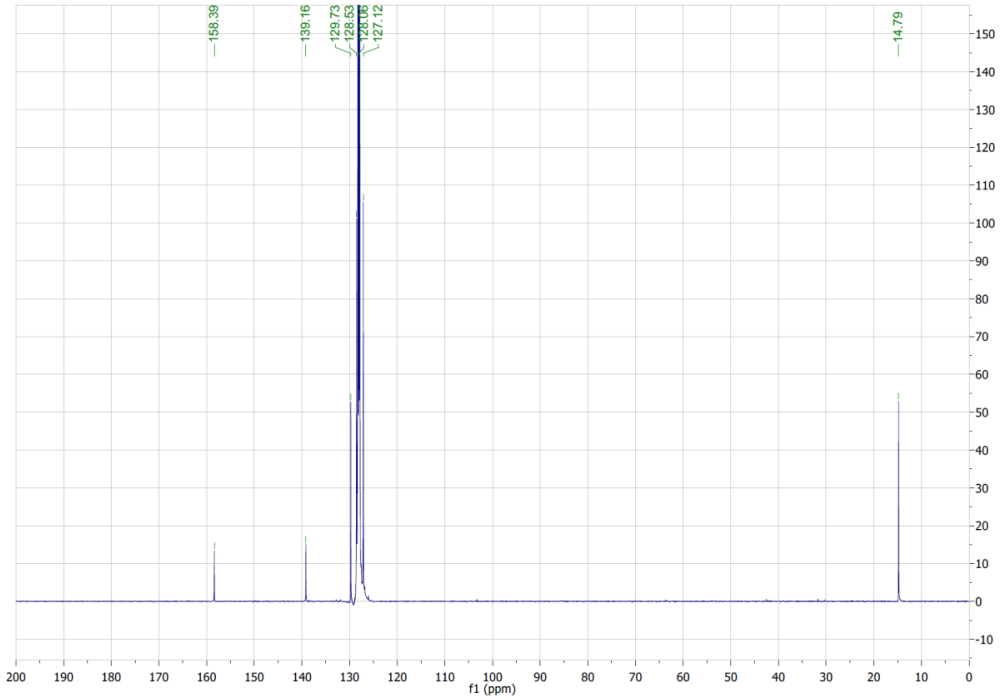




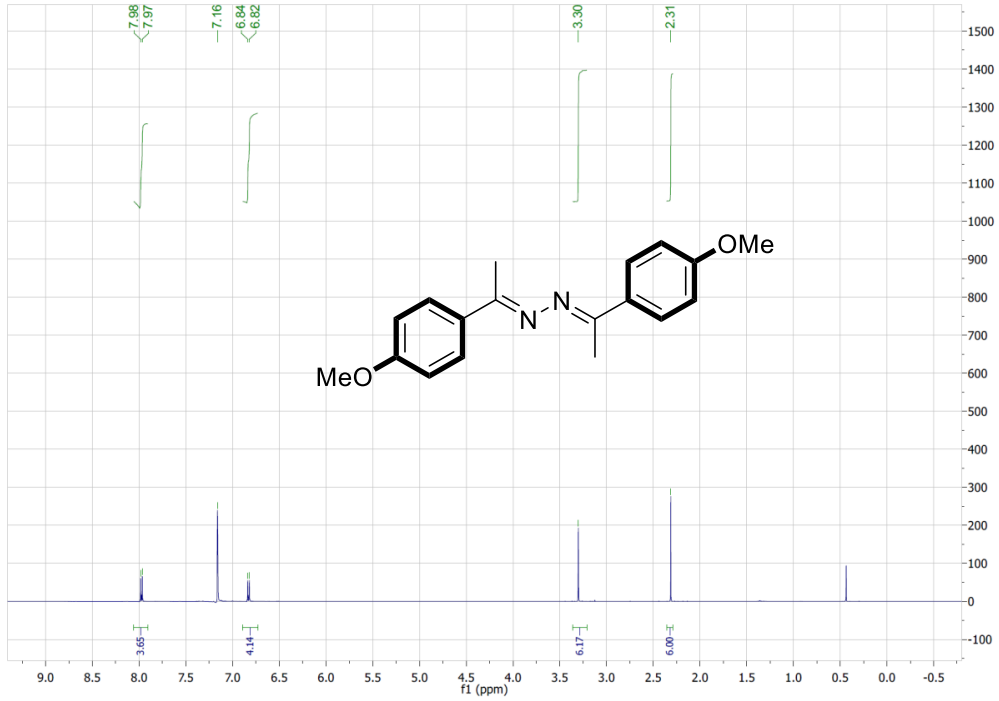
^{1}H and ^{13}C-{^{1}H} NMR spectra for compounds 2a-2d (Table 4):

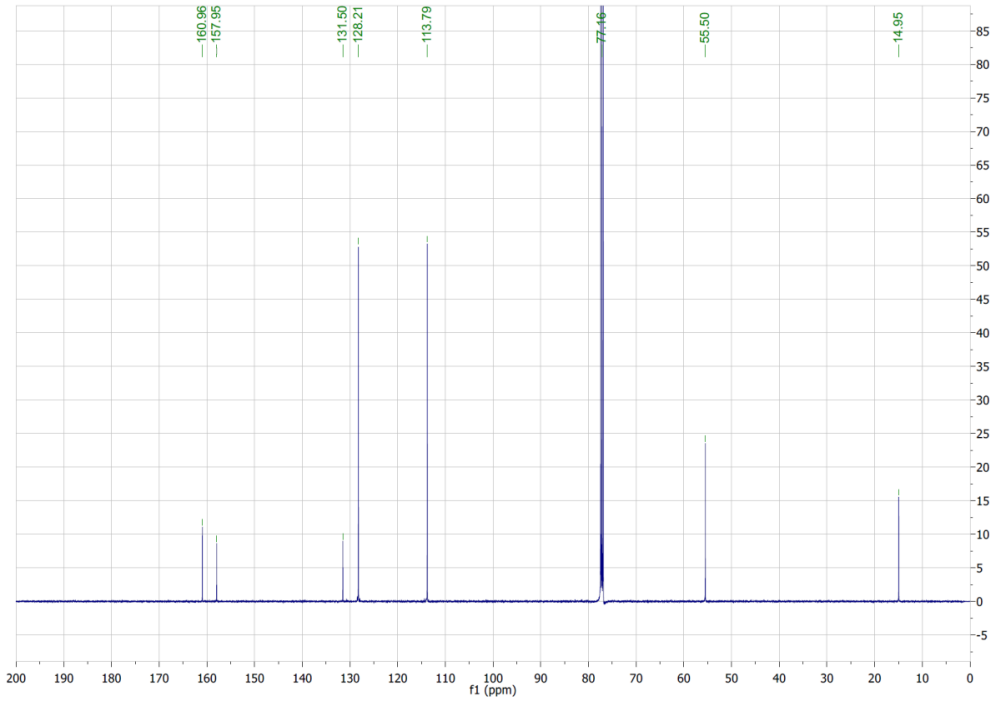
1,2-bis-[1-phenylethylidene]hydrazine (2a):



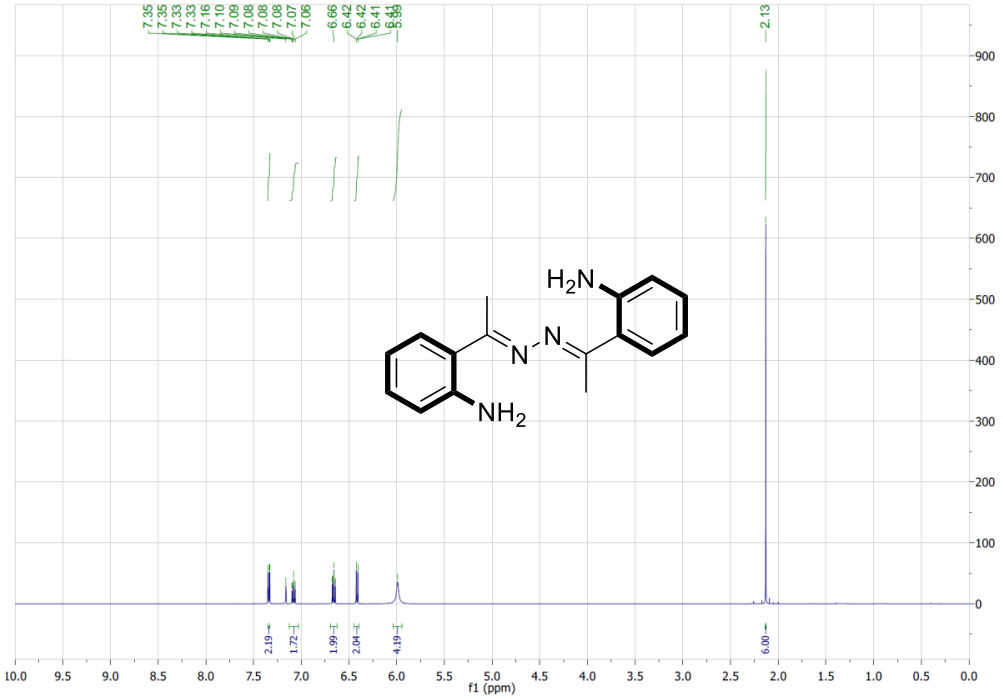


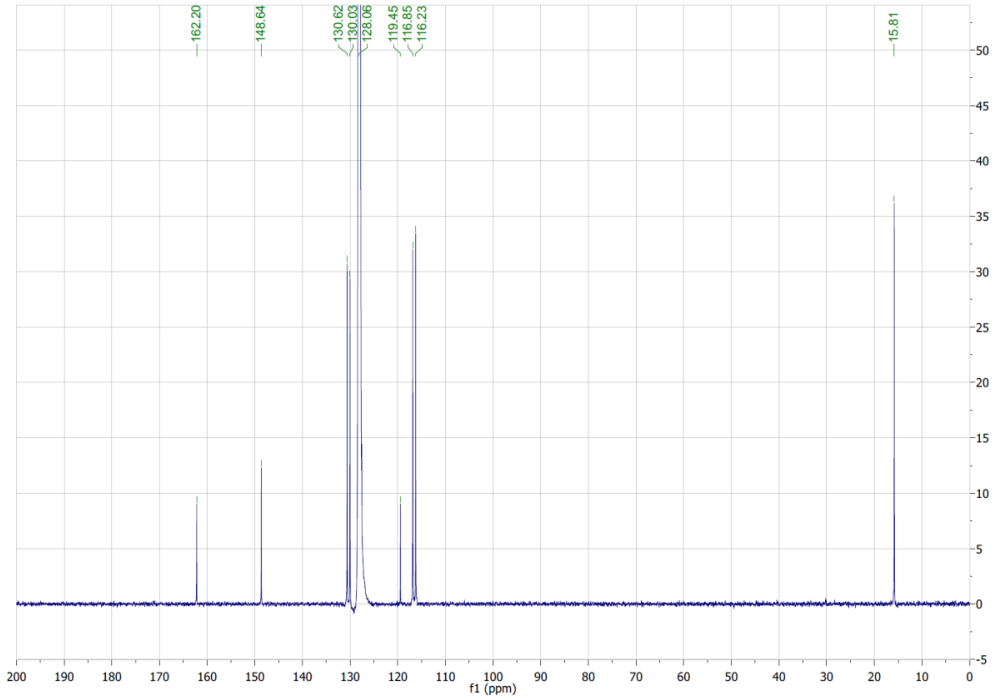
1,2-bis-[1-(4-methoxyphenyl)ethylidene]hydrazine (2b):



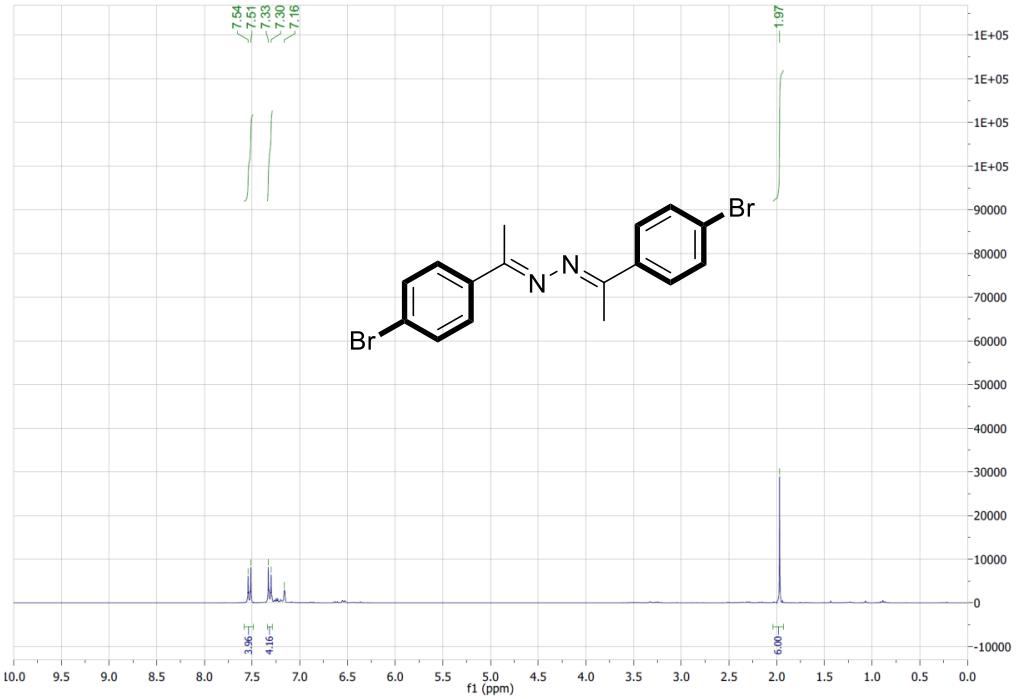


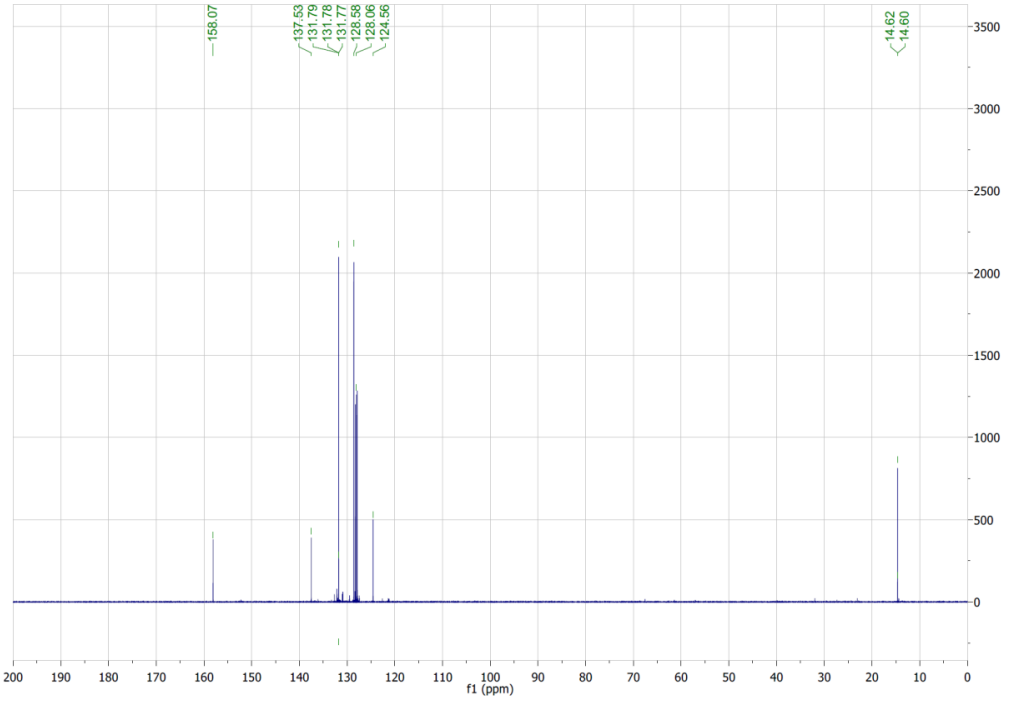
1,2-bis-[1-(2-aminophenyl)ethylidene]hydrazine (2c):





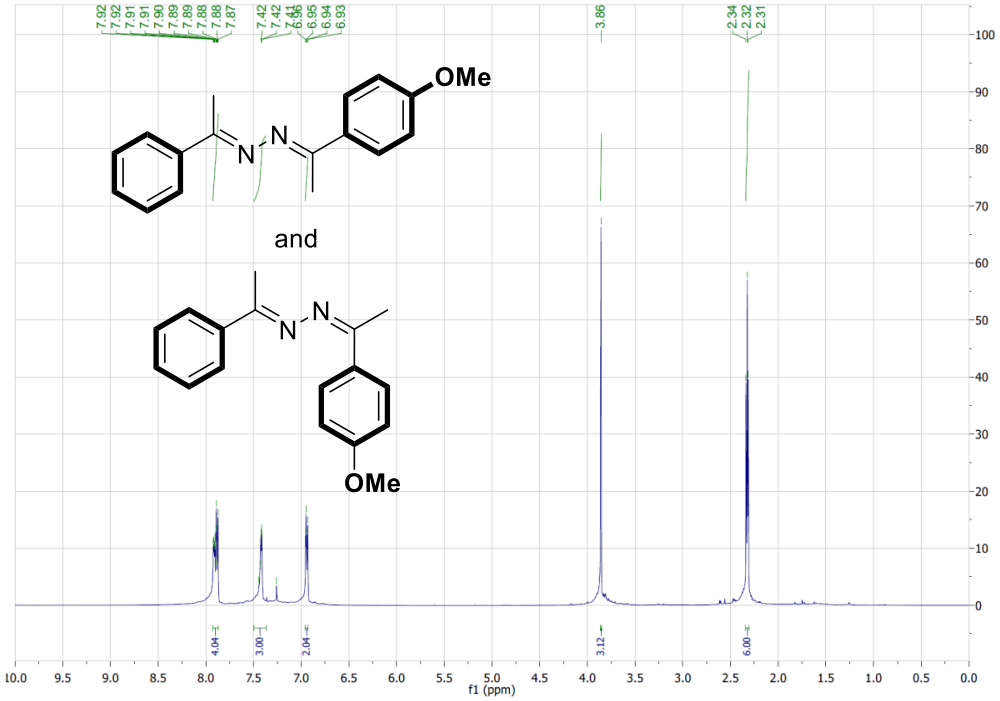
1,2-bis-[1-(4-bromophenyl)ethylidene]hydrazine (2d):

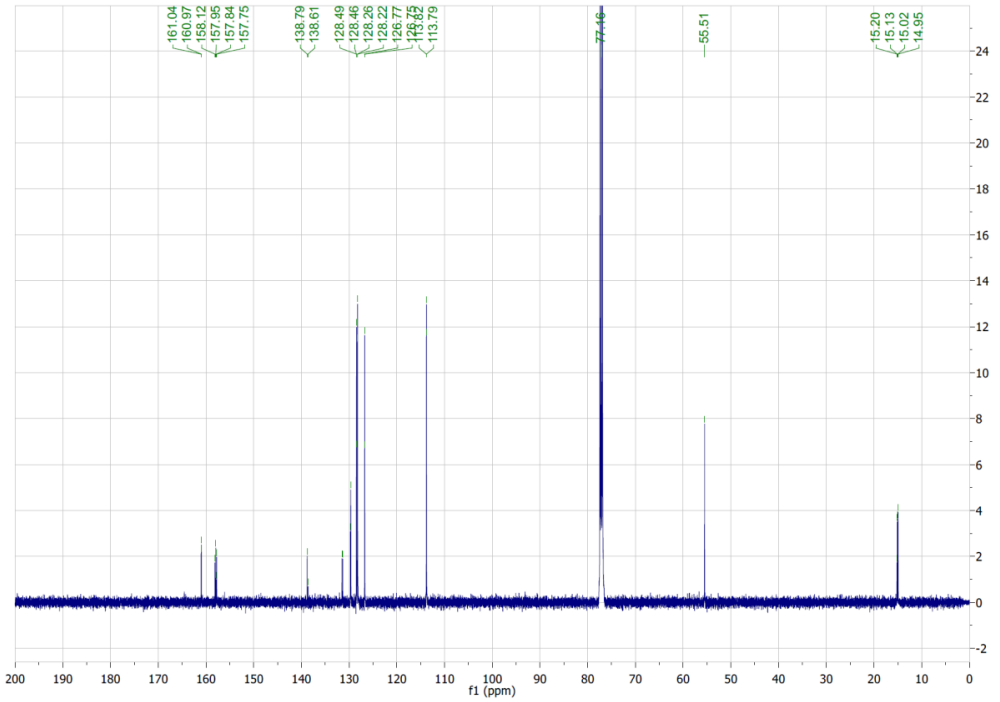

7.54
7.51
7.33
7.30
7.16
1.97
Br
Br
N
N
3.96
4.16
6.00
f1 (ppm)


158.07
137.53
131.79
131.78
131.77
128.58
128.06
124.56
14.62
14.60
f1 (ppm)

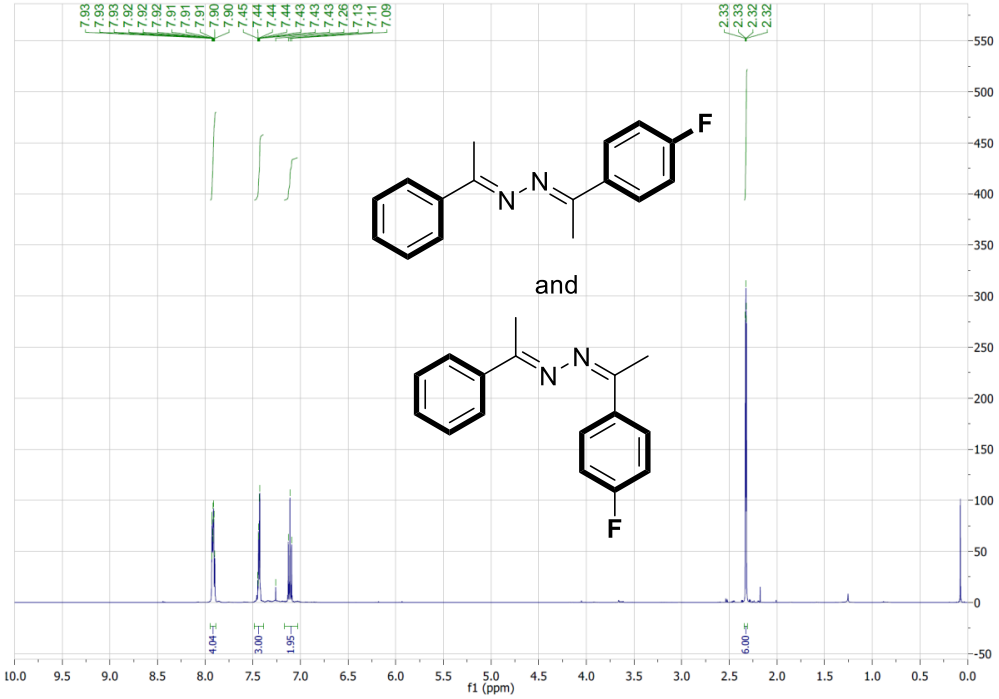
^{1}H and ^{13}C-{^{1}H} NMR spectra for compounds 2e-2h (Table 5):

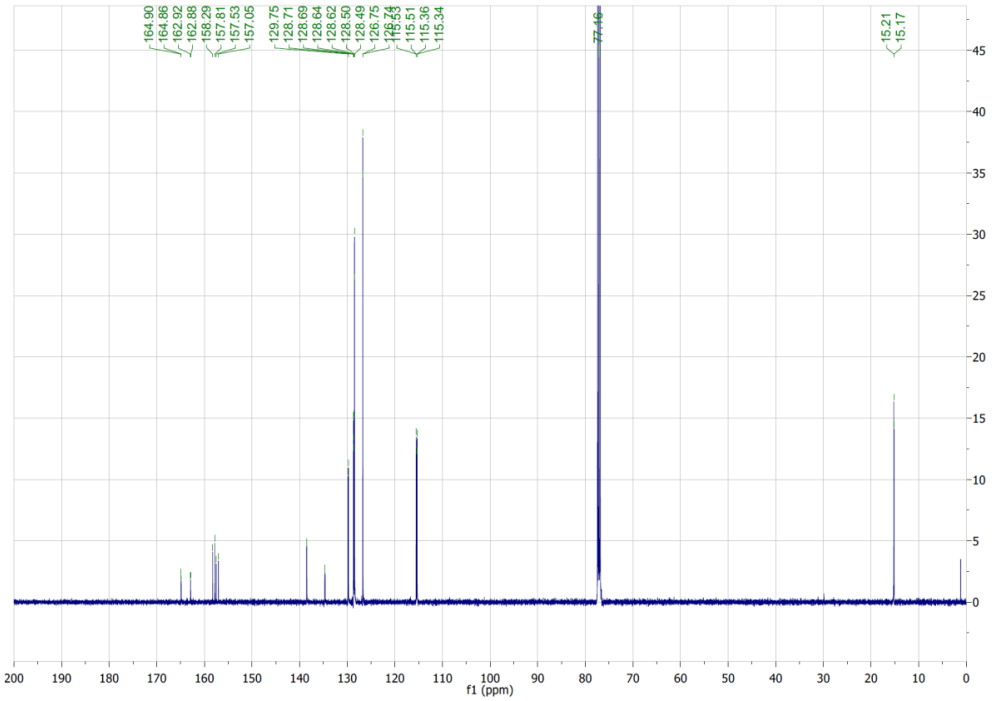
[1-(4-methoxyphenyl)-2-(1-phenyl)]ethylidenehydrazine (2e):



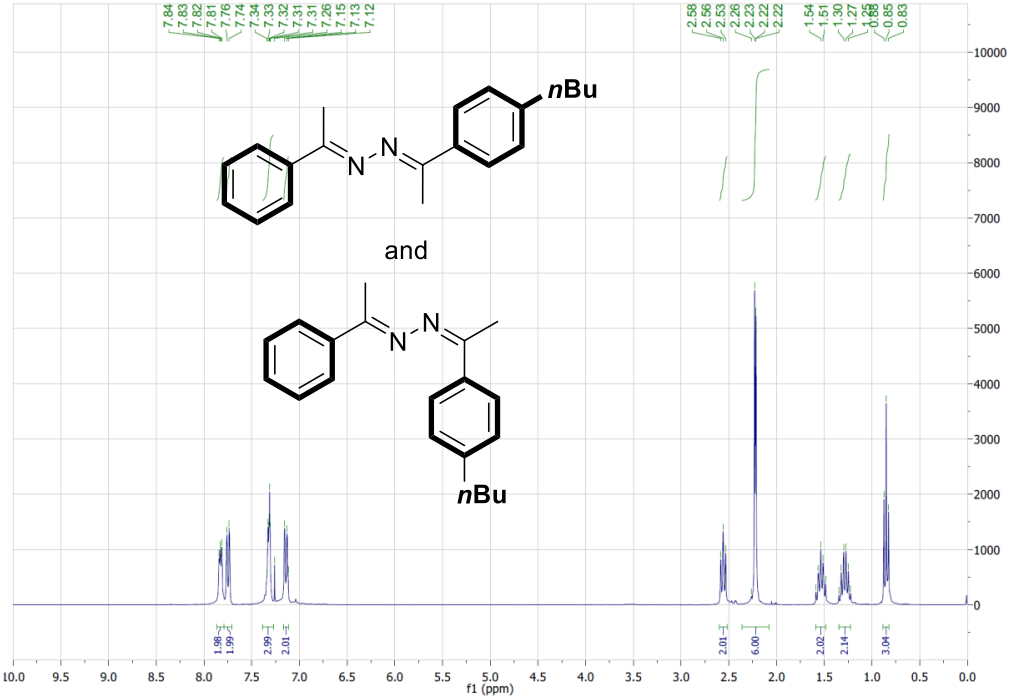


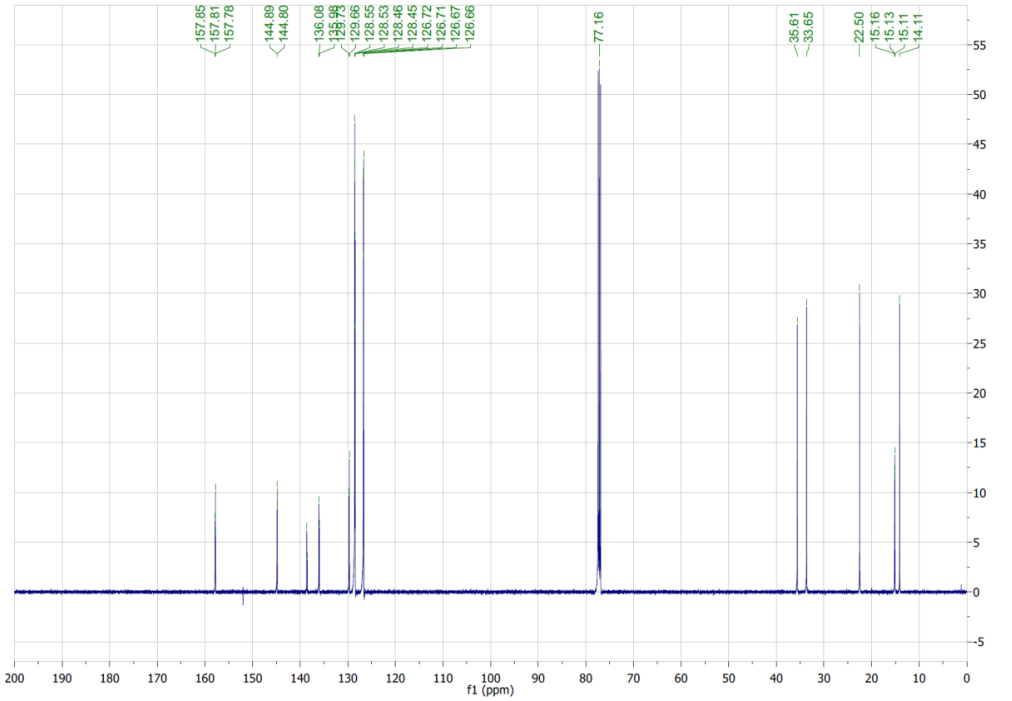
[1-(4-fluorophenyl)-2-(1-phenyl)]ethylidenehydrazine (2f):



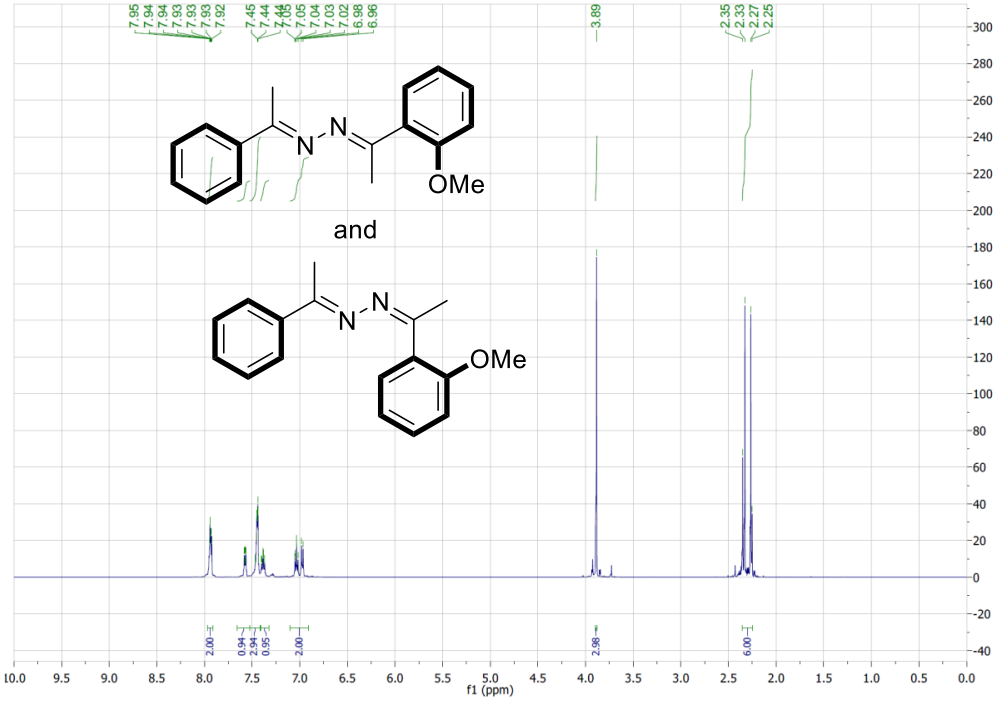


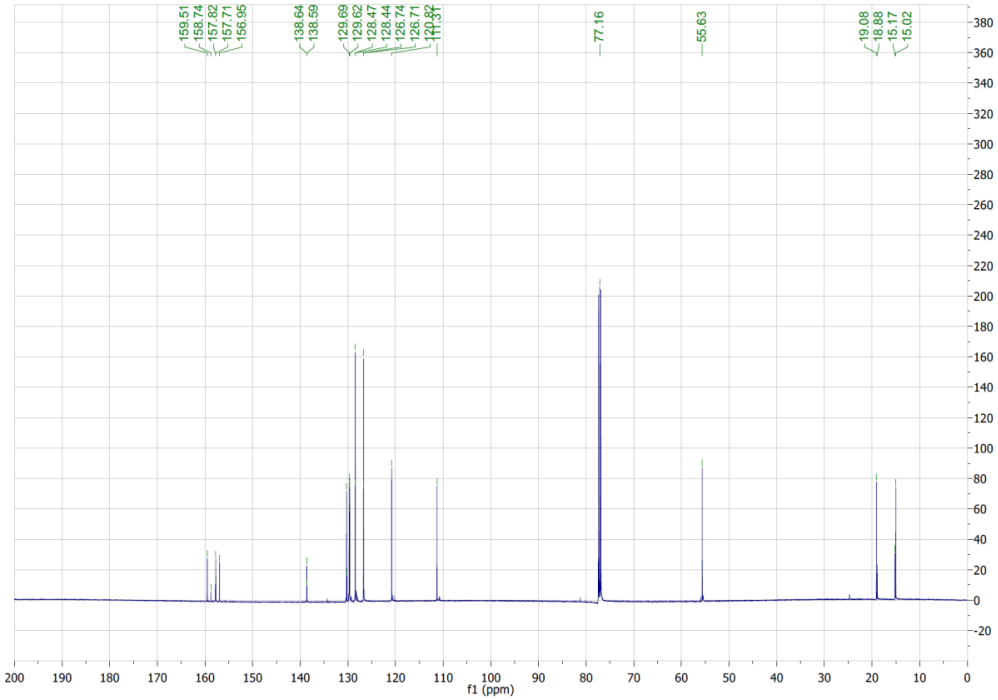
[1-(4-butylphenyl)-2-(1-phenyl)]ethylidenehydrazine (2g):





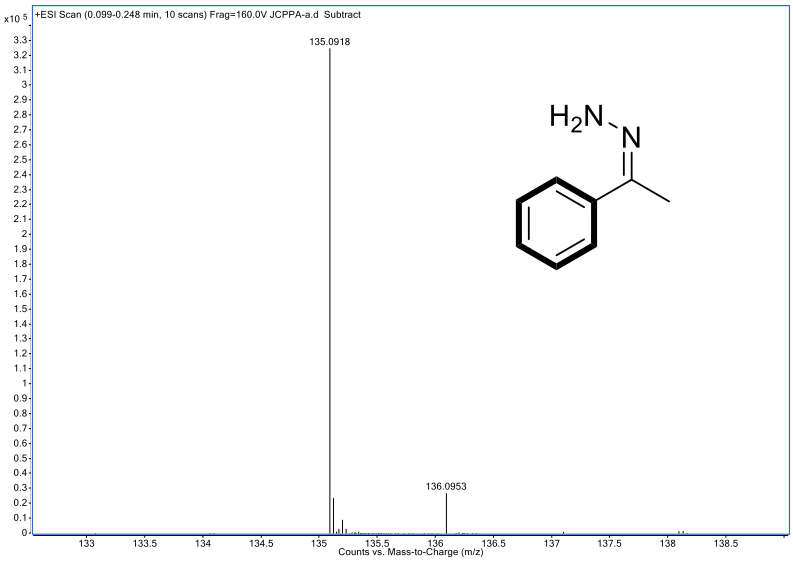
[1-(2-methoxyphenyl)-2-(1-phenyl)]ethylidenehydrazine (2h):



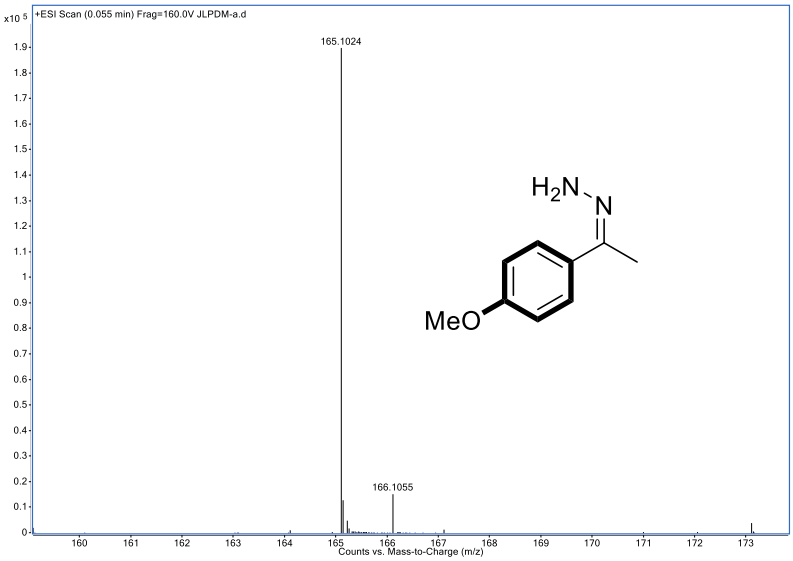


HR-MS spectra for compounds 1a-1i (Table 2):

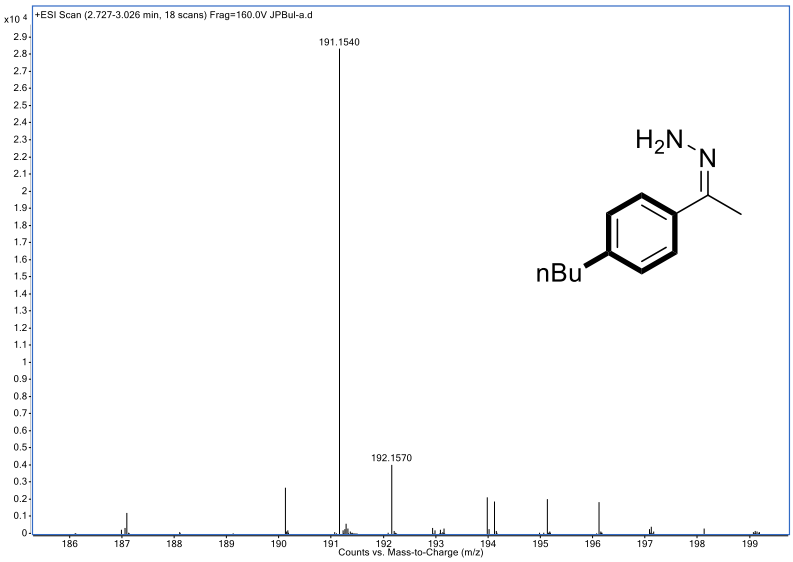
1-phenylhydrazonoethane (1a):



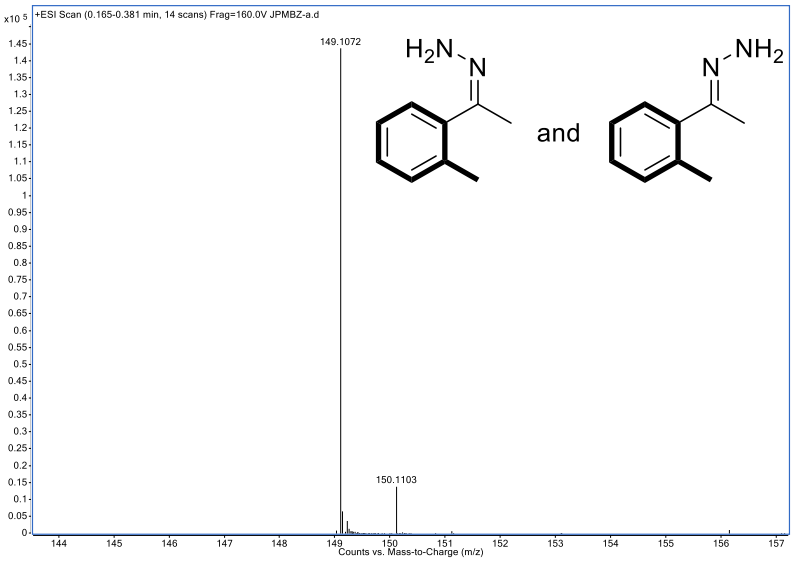
1-(4-methoxyphenyl)-hydrazonoethane (1b):



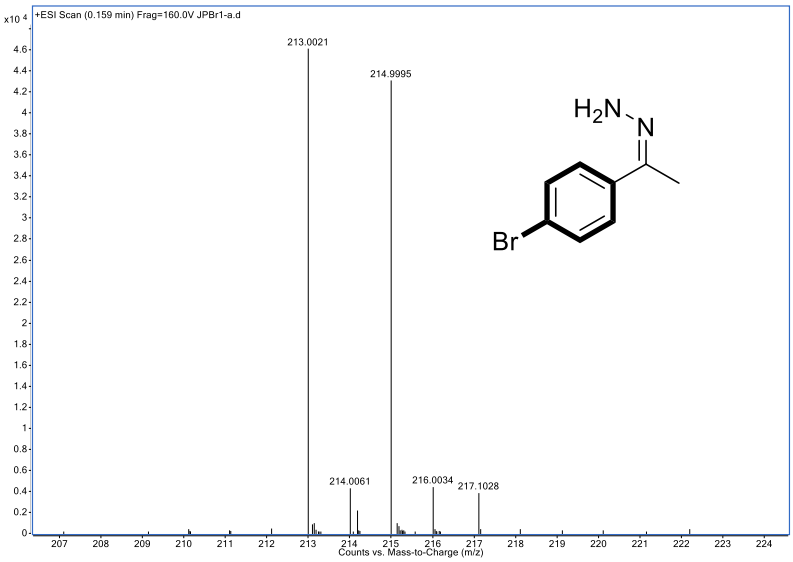
1-(4-butylphenyl)-hydrazonoethane (1c):



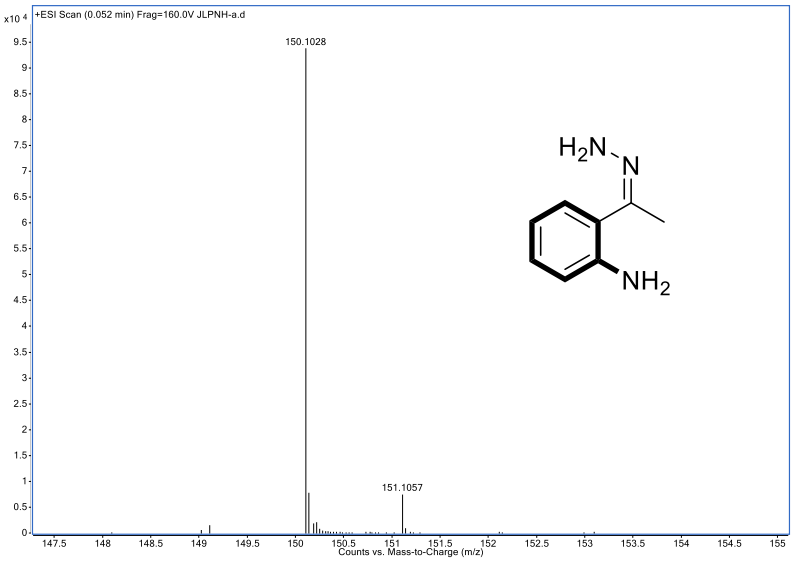
1-(2-methylphenyl)-hydrazonoethane (1d):



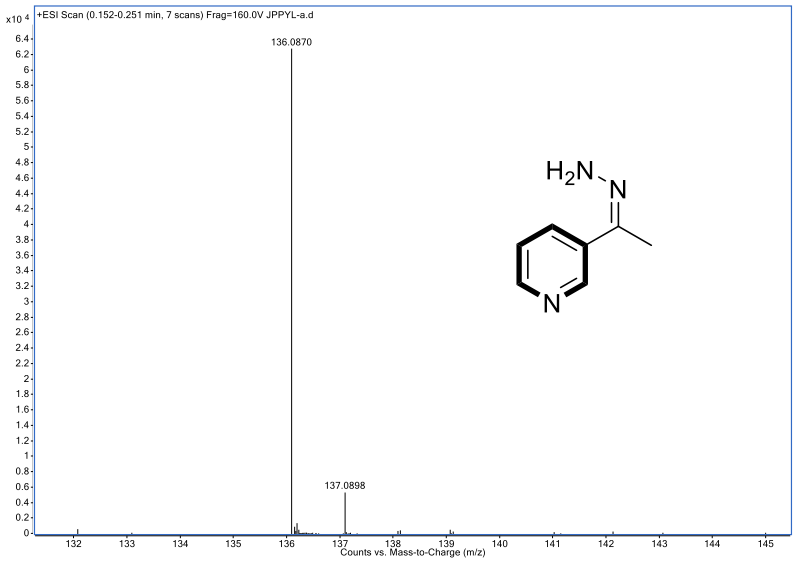
1-(4-bromophenyl)-hydrazonoethane (1e):



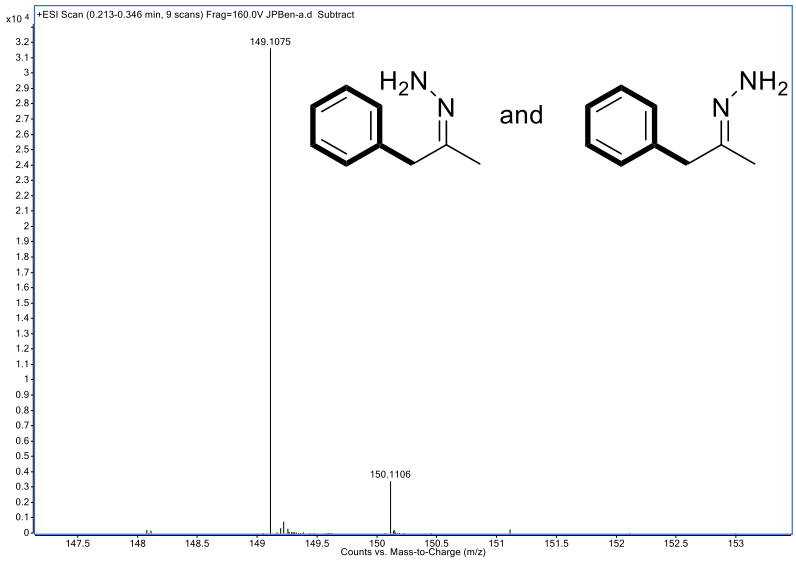
1-(2-aminophenyl)-hydrazonoethane (1f):



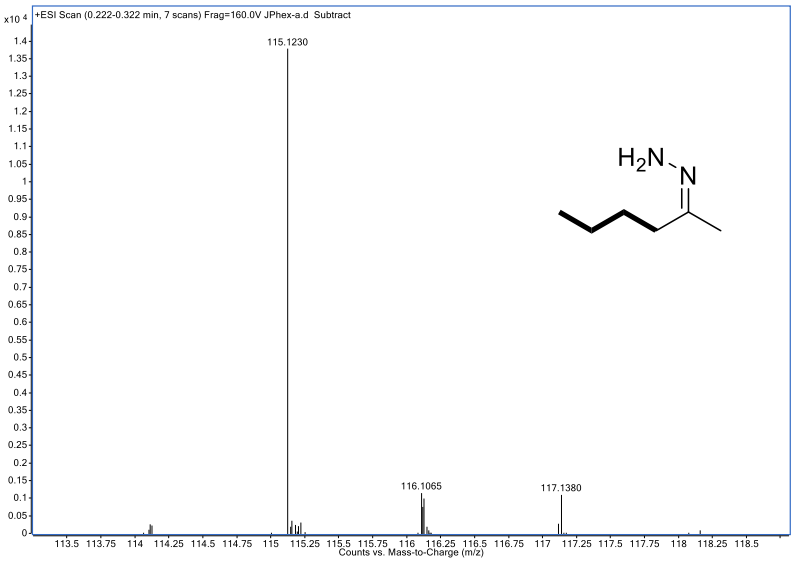
1-(3-Pyridinyl)-hydrazonoethane (1g):



1-phenyl-2-hydrazonopropane (1h):

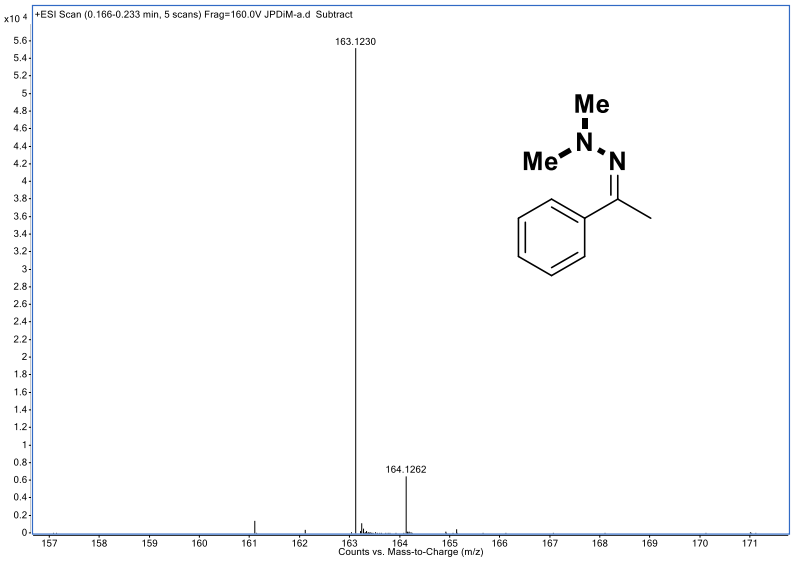


2-hydrazonohexane (1i):

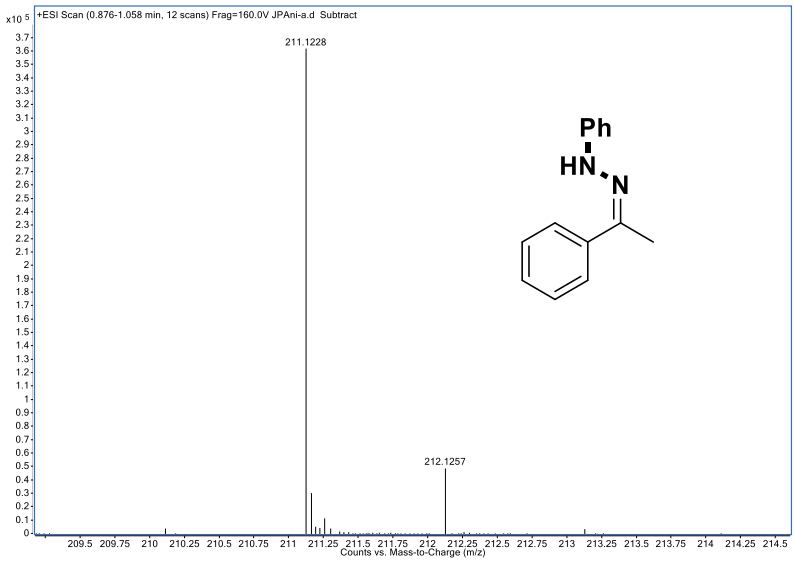


HR-MS spectra for compounds 1j-1m (Table 3):

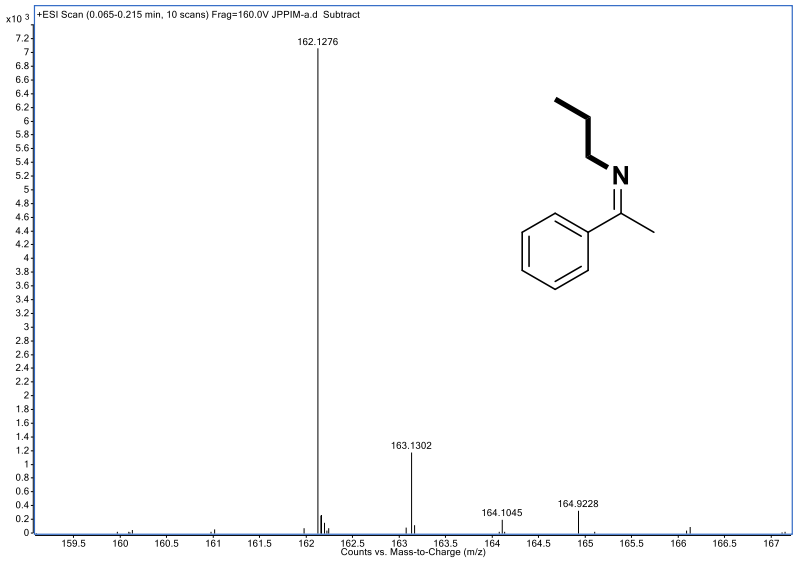
1-phenyl-2,2-dimethylhydrazonoethane (1j):



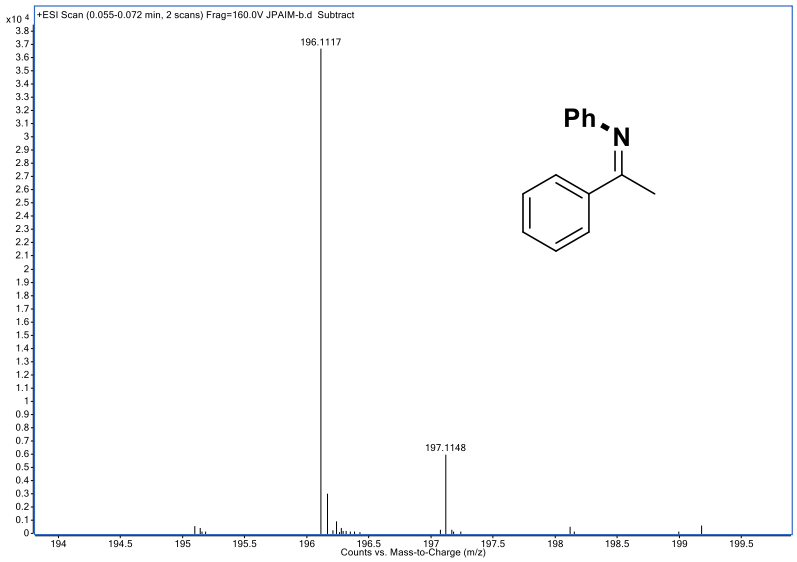
1-phenyl-2-phenylhydrazonoethane (1k):



N-(1-phenylethylidene)-1-propanamine (1l):

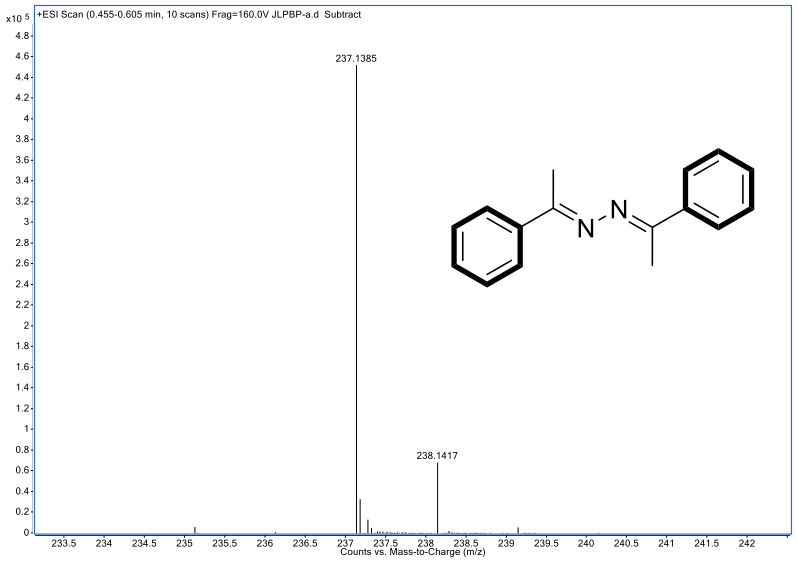


***N*-(1-phenylethylidene)-benzenamine (1m):**

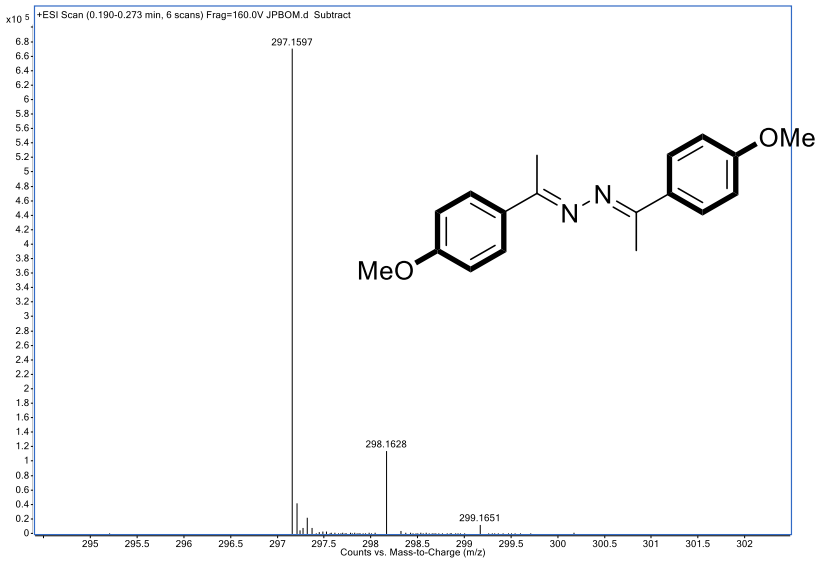


HR-MS spectra for compounds 2a-2d (Table 4):

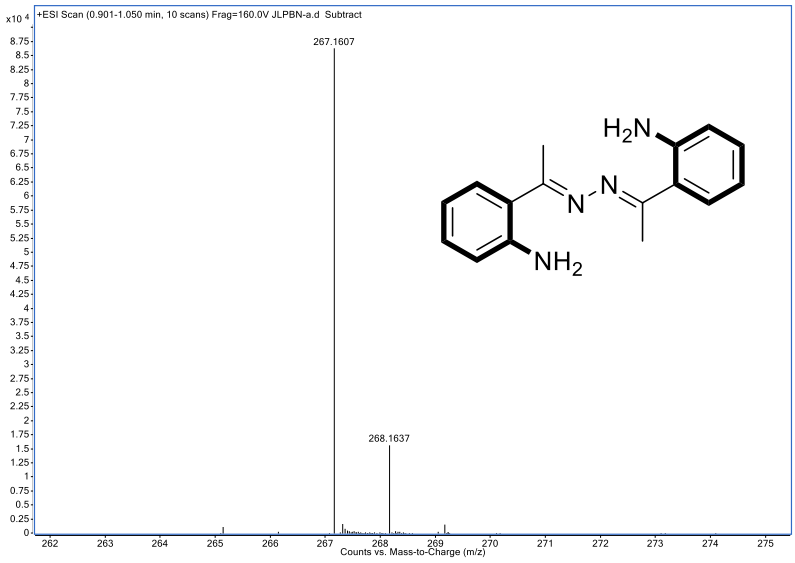
1,2-bis-[1-phenylethylidene]hydrazine (2a):



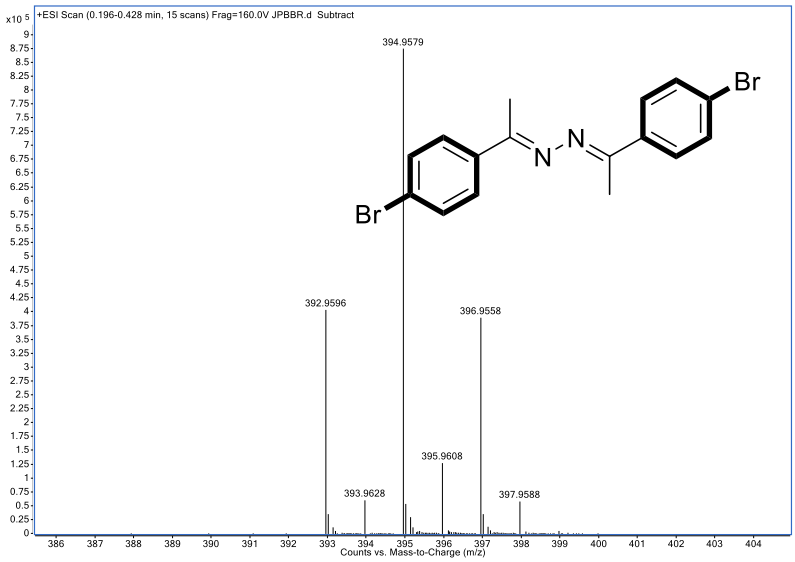
1,2-bis-[1-(4-methoxyphenyl)ethylidene]hydrazine (2b):



1,2-bis-[1-(2-aminophenyl)ethylidene]hydrazine (2c):

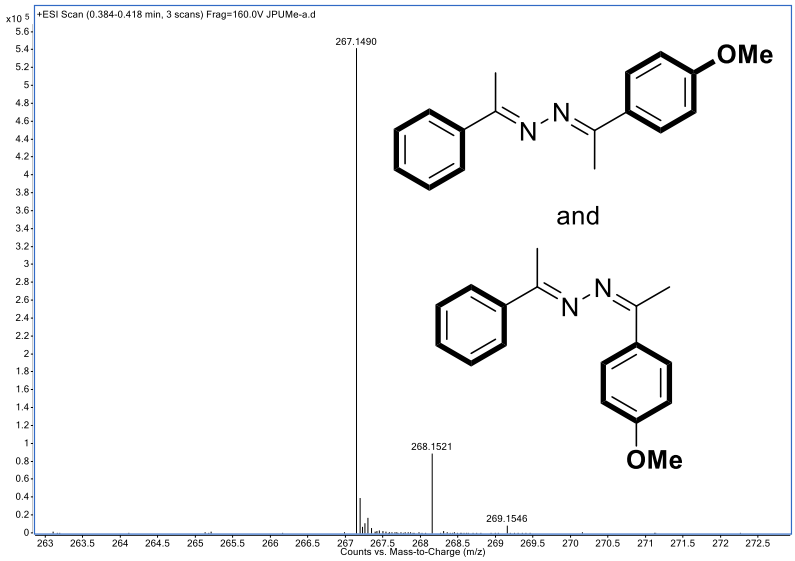


1,2-bis-[1-(4-bromophenyl)ethylidene]hydrazine (2d):

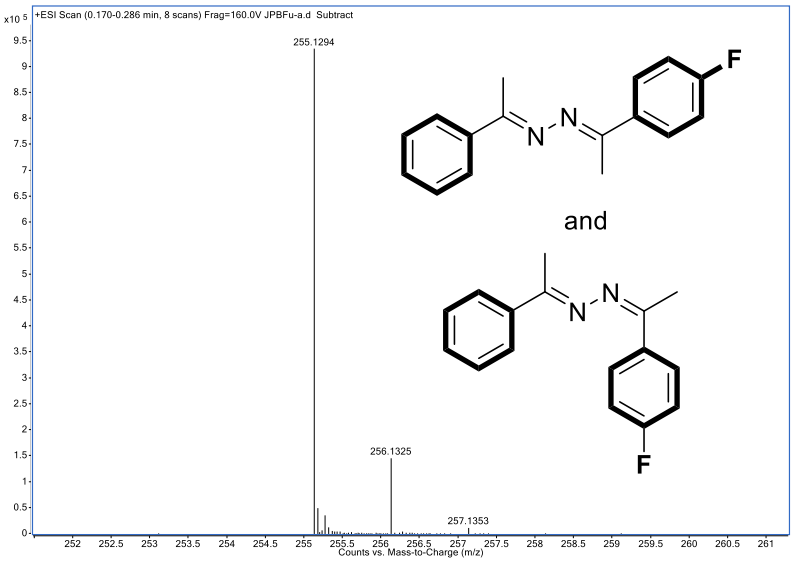


HR-MS spectra for compounds 2e-2h (Table 5):

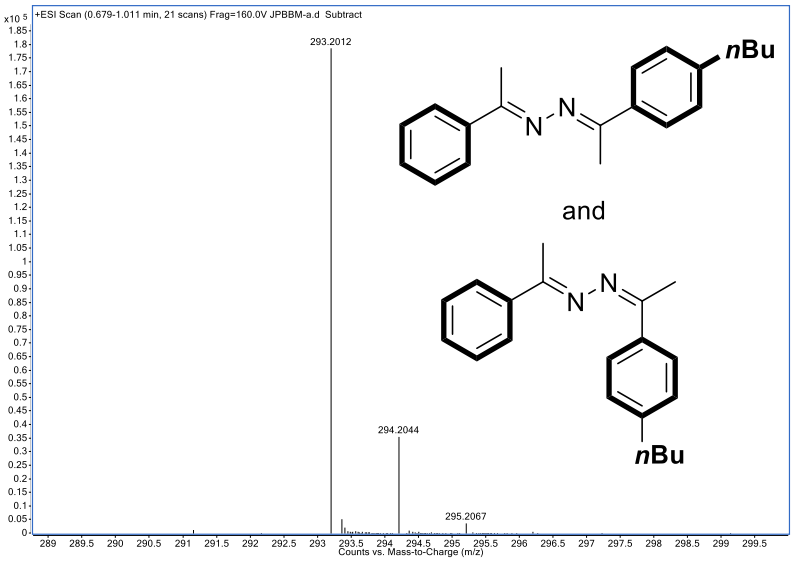
[1-(4-methoxyphenyl)-2-(1-phenyl)]ethylidenehydrazine (2e):



[1-(4-fluorophenyl)-2-(1-phenyl)]ethylidenehydrazine (2f):



[1-(4-butylphenyl)-2-(1-phenyl)]ethylidenehydrazine (2g):



[1-(2-methoxyphenyl)-2-(1-phenyl)]ethylidenehydrazine (2h):