Electronic Supplementary Material (ESI) for ChemComm.

Supplementary Material

A formal [3 + 2] cycloaddition reaction of *N*-methylimidazole as a masked hydrogen cyanide: access to 1,3-disubstitued-1*H*-1,2,4-triazoles

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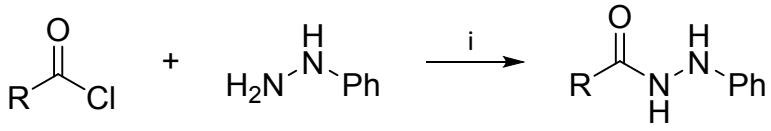
General experimental

All purchased solvents and chemicals were of analytical grade and used without further purification. Melting points: Electrothermal 9100 apparatus. IR spectra: Shimadzu-IR 460 spectrometer; ῡ in cm^{-1}. ^{1}H NMR, ^{13}C NMR, and ^{19}F NMR spectra: Bruker DRX-500 Avance instrument using $CDCl_3$ as solvent at 500.1, 125.7, and 471 MHz, respectively; δ in ppm, J in Hz. Mass spectra were recorded on a Finnigan-MAT-8430MS spectrometer; at an ionization potential of 70 eV; in *m/z* (rel. %). Elemental analyses for C, H, and N were performed using a Heraeus CHN-O-Rapid analyzer.

General procedure for the preparation of *N*'-phenylacylhydrazides 11a-11r

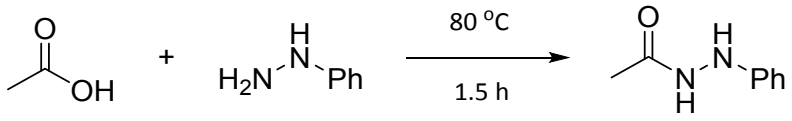
To a well-stirred solution of 2.16 g (20 mmol) phenyl hydrazine in 12 mL of pyridine at 0 ^{o}C was slowly added acyl chloride (22 mmol, 1.1 equiv.) within 10 minutes. Upon completion of addition, the stirring was continued allowing the reaction to reach rt. After 8 hours, water (100 mL) was added, the precipitate was collected, washed with water and recrystallized from methanol to give pure acylhydrazide **11**. This procedure was used for the preparation of acylhydrazides **11** except for **11j**, **11o**, **11p**, and **11q**. *N*'-Phenylisonicotino- (**11o**), *N*'-phenylnicotino- (**11p**), and *N*'-phenylpicolinohydrazide (**11q**) were purified by column chromatography (*n*-hexane: EtOAc, 1:1).



11

i) Py, 0 ^{o}C to rt

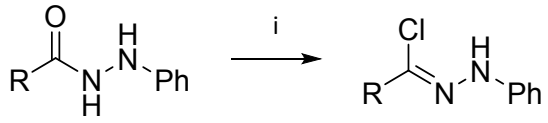
Literature procedure was used for the synthesis of **11j.**[1] Thus**,** phenylhydrazine (1.08 g, 10 mmol) was added to acetic acid (20 mL) at room temperature. The resulting mixture was heated at 80 °C for 1.5 hours. Most of the acetic acid was distilled off, the residue was cooled to room temperature, diethyl ether (10 mL) was added and a solid precipitated. The solid was then filtered and washed with diethyl ether (3 × 10 mL).



11j

General procedure for the preparation of hydrazonoyl chlorides 1a-1r

Literature procedure was used for the synthesis of hydrazonoyl chlorides **1a-1r.**[2] Carbon tetrachloride (2 g, 13.5 mmol) was added to a stirred suspension of compound *N*'-phenylacylhydrazides **11a-11r** (10 mmol) and Ph_3P (3.15 g, 12 mmol) in acetonitrile (20 mL). Upon completion of addition, the reaction mixture was stirred at room temperature for 8 h. Then, the solvent was removed under reduced pressure, and the crude product was subjected to flash column chromatography (*n*-hexane: EtOAc, 15:1) to afford hydrazonoyl chlorides **1a-1r**, except for **1o**, **1p**, and **1q**, which required different eluent (*n*-hexane: EtOAc, 3:1).

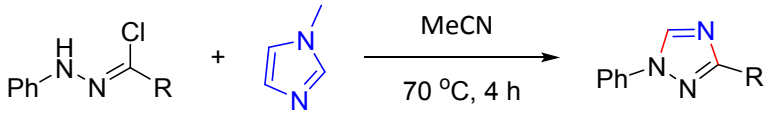


11a-11r **1a-1r**

i) CCl_4, PPh_3, CH_3CN, rt, 8 h.

General procedure for the synthesis of 1,3-disubstitued-1*H*-1,2,4-triazoles 3a-3r

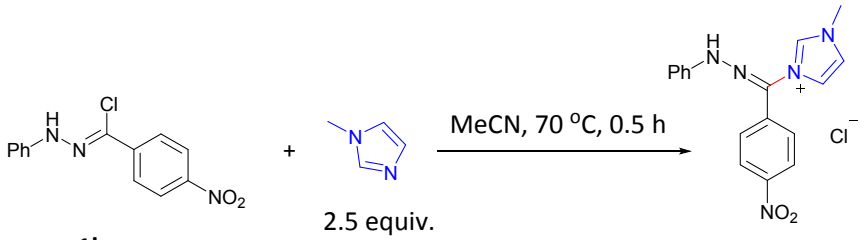
A mixture of **1** (0.5 mmol) and *N*-methylimidazole (103 mg, 1.25 mmol, 2.5 eq) in MeCN (2 mL) was stirred at 70 °C for 4 hours. Upon reaction completion (TLC monitoring), the solvent was removed under reduced pressure, and the product was purified by flash column chromatography on silica gel (Merck 230–400 mesh) using *n*-hexane: EtOAc (8:1) as eluent.



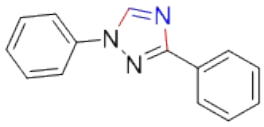
1a-1r **2.5 eq** **3a-3r**

General procedure for the isolation of stable intermediate 4h

A mixture of **1h** (0.5 mmol) and *N*-methylimidazole (103 mg, 1.25 mmol, 2.5 eq) in MeCN (2 mL) was stirred at 70 °C for 30 minutes. Upon reaction completion (TLC monitoring), the solvent was removed under reduced pressure, and the precipitate was washed with minimum amount of cold acetonitrile then with water to afford compound **4h** as a yellow solid in 83% yields.



Characteristic data



1,3-Diphenyl-1*H*-1,2,4-triazole (3a)

Yield 102 mg (92%); Pale yellow solid; mp 78-79 °C {Lit.[3] 80-81 °C}.

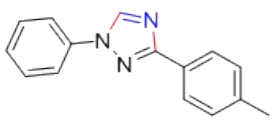
IR (KBr, cm^{-1}) 3062, 2924, 1599, 1525, 1504, 1438, 1330, 1246, 1066.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.60 (s, 1H), 8.24 (d, *J* = 7.2 Hz, 2H), 7.55-7.40 (m, 6H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 162.2, 140.4, 136.1, 129.6, 128.7, 128.5, 127.6, 127.0, 125.5, 118.8.

MS (*m/z*) 221 (100), 194 (31), 103 (12), 91 (92), 64 (30), 51 (11).

Anal. Calcd for $C_{14}H_{11}N_3$ (221.26) C, 76.00; H, 5.01; N, 18.99. Found C, 76.31; H, 5.03; N, 19.04.



1-Phenyl-3-(*p*-tolyl)-1*H*-1,2,4-triazole (3b)

Yield 106 mg (90%); Pale yellow solid; mp 65-66 °C {Lit.[4] 64-66 °C}.

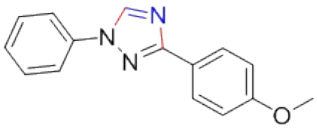
IR (KBr, cm^{-1}) 3046, 2920, 1599, 1500, 1439, 1323, 1246, 1064.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.58 (s, 1H), 8.13 (d, *J* = 7.9 Hz, 2H), 7.77 (d, *J* = 7.7 Hz, 2H), 7.54 (t, *J* = 7.8 Hz, 2H), 7.42 (t, *J* = 7.4 Hz, 1H), 7.31 (d, *J* = 7.7 Hz, 2H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 162.3, 140.3, 138.5, 136.2, 128.7, 128.3, 126.8, 127.0, 125.4, 118.8, 20.4.

MS (*m/z*) 235 (100), 208 (27), 117 (10), 91 (84), 64 (25).

Anal. Calcd for $C_{15}H_{13}N_3$ (235.29) C, 76.57; H, 5.57; N, 17.86. Found C, 76.86; H, 5.59; N, 17.90.



3-(4-Methoxyphenyl)-1-phenyl-1*H*-1,2,4-triazole (3c)

Yield 115 mg (91%); Pale yellow solid; mp 99-100 °C {Lit.[5] 98-100 °C}.

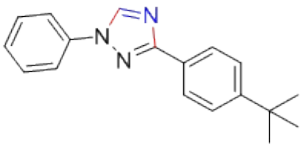
IR (KBr, cm^{-1}) 3121, 2972, 1615, 1599, 1508, 1436, 1289, 1244, 1024.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.56 (s, 1H), 8.16 (d, *J* = 8.3 Hz, 2H), 7.75 (d, *J* = 7.6 Hz, 2H), 7.52 (t, *J* = 7.5 Hz, 2H), 7.40 (t, *J* = 7.0 Hz, 1H), 7.02 (d, *J* = 8.3 Hz, 2H), 3.87 (s, 3H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 162.0, 159.7, 140.3, 136.2, 128.7, 126.9, 126.8, 122.3, 118.7, 113.0, 54.3.

MS (*m/z*) 251 (100), 236 (43), 209 (38), 151 (20), 91 (75), 64 (20).

Anal. Calcd for $C_{15}H_{13}N_3O$ (251.29) C, 71.70; H, 5.21; N, 16.72. Found C, 71.98; H, 5.22; N, 16.77.



3-(4-(*tert*-Butyl) phenyl)-1-phenyl-1*H*-1,2,4-triazole (3d)

Yield 122 mg (88%); Pale yellow solid; mp 102-104 °C

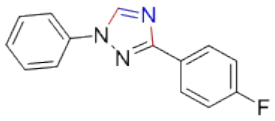
IR (KBr, cm^{-1}) 3111, 2953, 2864, 1600, 1510, 1325, 1262, 1123, 1063, 982.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.78 (s, 1H), 8.36 (d, *J* = 7.9 Hz, 2H), 7.95 (d, *J* = 7.5 Hz, 2H), 7.71 (t, *J* = 7.7 Hz, 4H), 7.59 (t, *J* = 7.0 Hz, 1H), 1.58 (s, 9H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 163.3, 152.8, 141.6, 137.3, 129.8, 128.0, 127.9, 126.4, 125.7, 119.9, 34.9, 31.4.

MS (*m/z*) 277 (3), 266 (100), 236 (9), 150 (70), 91 (77), 77 (18), 64 (21).

Anal. Calcd for $C_{18}H_{19}N_3$ (277.37) C, 77.95; H, 6.90; N, 15.15. Found C, 78.25; H, 6.91; N, 15.20.



3-(4-Fluorophenyl)-1-phenyl-1*H*-1,2,4-triazole (3e)

Yield 114 mg (95%); Pale yellow solid; mp 110-111 °C.

IR (KBr, cm^{-1}) 3111, 2924, 1603, 1512, 1444, 1320, 1220, 1158, 1110.

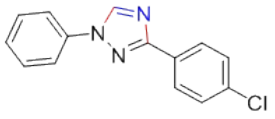
^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.58 (s, 1H), 8.21 (t, *J* = 7.9 Hz, 2H), 7.76 (d, *J* = 7.8 Hz, 2H), 7.54 (t, *J* = 7.6 Hz, 2H), 7.43 (t, *J* = 7.1 Hz, 1H), 7.18 (t, *J* = 8.5 Hz, 2H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 162.7 (d, $^1J_{CF}$ = 248.7 Hz), 161.4, 140.5, 136.0, 128.7, 127.4 (d, $^3J_{CF}$ = 8.4 Hz), 127.0, 125.8 (d, $^4J_{C\text{-}F}$ = 3.1 Hz), 118.7, 114.6 (d, $^2J_{CF}$ = 21.6 Hz).

^{19}F NMR (471 MHz, $CDCl_3$) δ -106.5.

MS (*m/z*) 239 (100), 212 (49), 121 (14), 91 (63), 64 (47).

Anal. Calcd for $C_{14}H_{10}FN_3$ (239.25) C, 70.28; H, 4.21; N, 17.56. Found C, 70.57; H, 4.23; N, 17.60.



3-(4-Chlorophenyl)-1-phenyl-1*H*-1,2,4-triazole (3f)

Yield 123 mg (95%); Pale yellow solid; mp 123-124 °C {Lit.[6] 124 °C}.

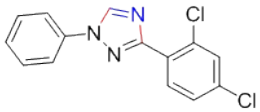
IR (KBr, cm^{-1}) 3112, 2946, 1601, 1557, 1517, 1495, 1339, 1258, 1241, 1161, 1103.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.57 (s, 1H), 8.15 (d, *J* = 7.9 Hz, 2H), 7.73 (d, *J* = 7.5 Hz, 2H), 7.52 (t, *J* = 7.2 Hz, 2H), 7.45-7.39 (m, 3H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 161.2, 140.5, 136.0, 134.4, 128.7, 128.17, 127.85, 127.09, 126.8, 118.81.

MS (*m/z*) 255 (100), 228 (22), 137 (7), 91 (63), 64 (30).

Anal. Calcd for $C_{14}H_{10}ClN_3$ (255.71) C, 65.76; H, 3.94; N, 16.43. Found C, 66.07; H, 3.95; N, 16.47



3-(2,4-Dichlorophenyl)-1-phenyl-1*H*-1,2,4-triazole (3g)

Yield 138 mg (95%); Pale yellow solid; mp 139-141 °C.

IR (KBr, cm^{-1}) 3114, 3043, 2918, 1593, 1502, 1426, 1318, 1121, 989.

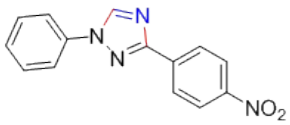
^1H NMR (500 MHz, Chloroform-*d*) δ 8.66 (s, 1H), 7.99 (d, *J* = 8.4 Hz, 1H), 7.77 (d, *J* = 7.8 Hz, 2H), 7.60 – 7.49 (m, 3H), 7.43 (t, *J* = 7.2 Hz, 1H), 7.37 (d, *J* = 8.4 Hz, 1H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 120.09, 127.26, 128.42, 129.95, 130.79, 132.31, 133.78, 135.71, 137.10, 141.14, 160.94.

^{13}C NMR (126 MHz, Chloroform-*d*) δ 160.9, 141.1, 137.1, 135.7, 135.5, 133.8, 132.3, 130.8, 129.9, 128.4, 127.3, 120.1.

MS (*m/z*) 289 (49), 262 (9), 171 (5), 91 (100), 77 (10), 64 (29).

Anal. Calcd for $C_{14}H_9Cl_2N_3$ (290.15) C, 57.95; H, 3.13; N, 14.48. Found C, 58.23; H, 3.15; N, 14.53.



3-(4-Nitrophenyl)-1-phenyl-1*H*-1,2,4-triazole (3h)

Yield 103 mg (77%); Pale yellow solid; mp 186-188 °C {Lit.[7] 188-189}.

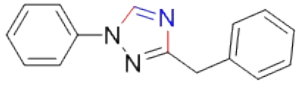
IR (KBr, cm^{-1}) 3094, 2923, 1594, 1523, 1512, 1459, 1414, 1319, 1243, 1100, 1067.

^1H NMR (500 MHz, Chloroform-d) δ 8.66 (s, 1H), 8.48 – 8.29 (m, 4H), 7.79 (d, *J* = 7.6 Hz, 2H), 7.58 (t, *J* = 7.8 Hz, 2H), 7.48 (t, *J* = 7.3 Hz, 1H), 7.29 (s, 1H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 161.4, 148.6, 142.1, 136.7, 130.0, 129.7, 128.7, 127.4, 124.1, 120.1.

MS (*m/z*) 266 (100), 236 (7), 220 (6), 159 (5), 91 (73), 77 (13), 64 (13).

Anal. Calcd for $C_{14}H_{10}N_4O_2$ (266.26) C, 63.15; H, 3.79; N, 21.04. Found C, 63.44; H, 3.81; N, 21.10.



3-Benzyl-1-phenyl-1*H*-1,2,4-triazole (3i)

Yield 107 mg (91%); Pale yellow solid; mp 89-91 °C {Lit.[8] 94.5 °C}.

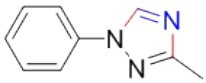
IR (KBr, cm^{-1}) 3107, 3028, 2927, 1599, 1522, 1493, 1336, 1349, 1231, 1061.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.66 (s, 1H), 7.85 (d, *J* = 7.7 Hz, 2H), 7.68 (t, *J* = 7.5 Hz, 2H), 7.64 – 7.50 (m, 5H), 7.45 (q, *J* = 7.3 Hz, 1H), 4.39 (s, 2H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 164.6, 141.5, 138.0, 137.2, 129.8, 129.1, 128.7, 128.0, 126.7, 120.0, 34.9.

MS (m/z) 235 (100), 207 (7), 159 (6), 132 (27), 104 (11), 91 (38), 77 (14), 64 (10).

Anal. Calcd for $C_{15}H_{13}N_3$ (235.29) C, 76.57; H, 5.57; N, 17.86. Found C, 76.88; H, 5.58; N, 17.90.



3-Methyl-1-phenyl-1*H*-1,2,4-triazole (3j)

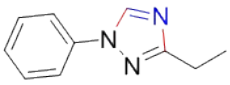
Yield 74 mg (93%); Pale yellow solid; mp 87-88 °C {Lit.[9] 89.5 °C}.

IR (KBr, cm^{-1}) 3098, 2972, 1599, 1528, 1498, 1309, 1243, 1232, 1065, 985.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.49 (s, 1H), 7.65 (d, *J* = 8.1 Hz, 2H), 7.50 (t, *J* = 7.5 Hz, 2H), 7.39 (t, *J* = 7.1 Hz, 1H), 2.52 (s, 3H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 162.1, 141.1, 137.2, 129.8, 127.9, 119.9, 14.0.
MS (*m/z*) 159 (100), 132 (50), 91 (99), 77 (29), 64 (68).

Anal. Calcd for $C_9H_9N_3$ (159.19) C, 67.90; H, 5.70; N, 26.40. Found C, 68.21; H, 5.71; N, 26.45.



3-Ethyl-1-phenyl-1*H*-1,2,4-triazole (3k)

Yield 77 mg (89%); Pale yellow oil.

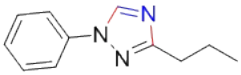
IR (KBr, cm^{-1}) 2971, 1598, 1523, 1411, 1249, 1223, 1054, 983.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.64 (s, 1H), 7.83 (d, *J* = 7.4 Hz, 2H), 7.67 (t, *J* = 6.8 Hz, 2H), 7.55 (t, *J* = 6.8 Hz, 1H), 3.04 (q, *J* = 7.2 Hz, 2H), 1.57 (t, *J* = 7.3 Hz, 3H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 167.0, 141.0, 137.2, 129.8, 127.8, 119.9, 21.9, 12.6.

MS (*m/z*) 173 (86), 145 (11), 91 (100), 77 (22), 64 (33).

Anal. Calcd for $C_{10}H_{11}N_3$ (173.22) C, 69.34; H, 6.40; N, 24.26. Found C, 69.64; H, 6.41; N, 24.31.



3-Propyl-1-phenyl-1*H*-1,2,4-triazole (3l)

Yield 81 mg (87%); Pale yellow oil.

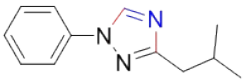
IR (KBr, cm^{-1}) 2959, 2927, 1601, 1599, 1522, 1457, 1402, 1328, 1246, 1102, 1059.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.66 (s, 1H), 7.85 (d, *J* = 7.8 Hz, 2H), 7.69 (t, *J* = 7.5 Hz, 2H), 7.57 (t, *J* = 7.1 Hz, 1H), 3.00 (t, *J* = 7.5 Hz, 2H), 2.05 (q, *J* = 7.4 Hz, 2H), 1.23 (t, *J* = 7.3 Hz, 3H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 165.9, 140.9, 137.2, 129.8, 127.8, 119.9, 30.4, 21.7, 13.9.

MS (*m/z*) 187 (28), 172 (42), 159 (100), 131 (8), 103 (25), 91 (69), 77 (38), 64 (31).

Anal. Calcd for $C_{11}H_{13}N_3$ (187.25.) C, 70.56; H, 7.00; N, 22.44. Found C, 70.87; H, 7.01; N, 22.48.



3-Isobutyl-1-phenyl-1*H*-1,2,4-triazole (3m)

Yield 81 mg (80%); Pale yellow oil.

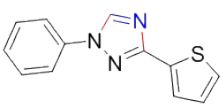
IR (KBr, cm^{-1}) 2956, 2927, 1600, 1521, 1456, 1416, 1319, 1247, 1228, 1060, 983.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.49 (s, 1H), 7.67 (d, *J* = 7.6 Hz, 3H), 7.50 (t, *J* = 7.6 Hz, 2H), 7.38 (t, *J* = 7.1 Hz, 1H), 2.71 (d, *J* = 7.1 Hz, 2H), 2.21 (m, 1H), 1.02 (d, *J* = 6.6 Hz, 6H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 165.2, 140.98, 137.2, 129.8, 127.9, 119.9, 37.4, 28.2, 22.5.

MS (*m/z*) 201 (15), 186 (23), 173 (52), 159 (100), 131 (4), 103 (11), 91 (67), 77 (20), 64 (16).

Anal. Calcd for $C_{11}H_{13}N_3$ (201.27.) C, 71.61; H, 7.51; N, 20.88. Found C, 71.90; H, 7.53; N, 20.93.



1-Phenyl-3-(thiophen-2-yl)-1*H*-1,2,4-triazole (3n)

Yield 103 mg (91%); Pale yellow solid; mp 114-116 °C {Lit.[6] 114 °C}.

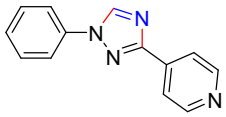
IR (KBr, cm^{-1}) 3090, 3020, 1601, 1596, 1564, 1465, 1318, 1296, 1216, 1076, 1057, 981.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.55 (s, 1H), 7.81 (s, 1H), 7.74 (d, *J* = 8.0 Hz, 2H), 7.52 (t, *J* = 7.5 Hz, 2H), 7.46-7.34 (m, 2H), 7.21 – 7.08 (m, 1H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 159.5, 141.5, 137.0, 133.5, 129.9, 128.2, 127.9, 127.0, 126.8, 120.0.

MS (*m/z*) 227 (98), 200 (26), 109 (9), 91 (100), 77 (13), 64 (36).

Anal. Calcd for $C_{12}H_9N_3S$ (227.29.) C, 63.41; H, 3.99; N, 18.49. Found C, 63.72; H, 4.00; N, 18.53.



4-(1-Phenyl-1*H*-1,2,4-triazol-3-yl)pyridine (3o)

Yield 102 mg (92%); Pale yellow solid; mp 142-143 °C.

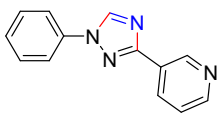
IR (KBr, cm^{-1}) 3106, 1599, 1500, 1419, 1346, 1258, 1073, 991.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.73 (d, *J* = 5.0 Hz, 2H), 8.62 (s, 1H), 8.05 (d, *J* = 5.0 Hz, 2H), 7.74 (d, *J* = 7.9 Hz, 2H), 7.54 (t, *J* = 7.8 Hz, 2H), 7.43 (t, *J* = 7.6 Hz, 1H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 161.3, 150.6, 142.1, 138.1, 137.0, 130.0, 128.6, 120.7, 120.8.

MS (*m/z*) 222 (88), 195 (16), 105 (10), 91 (100), 77 (21), 64 (48), 51 (15).

Anal. Calcd for $C_{13}H_{10}N_4$ (222.25) C, 70.26; H, 4.54; N, 25.21. Found C, 70.55; H, 4.55; N, 25.24.



3-(1-Phenyl-1*H*-1,2,4-triazol-3-yl)pyridine (3p)

Yield 100 mg (90%); Pale yellow solid; mp 101-102 °C.

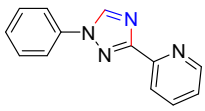
IR (KBr, cm^{-1}) 3090, 1598, 1589, 1499, 1419, 1335, 1256, 1237, 1073, 974.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 9.42 (s, 1H), 8.66 (s, 1H), 8.60 (d, *J* = 2.2 Hz, 1H), 8.44 (d, *J* = 7.8 Hz, 1H), 7.73 (d, *J* = 5.3 Hz, 2H), 7.52 (dt, *J* = 11.4, 5.6 Hz, 2H), 7.45 – 7.31 (m, 2H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 161.1, 150.5, 148.2, 141.9, 137.1, 133.9, 130.0, 128.4, 126.8, 123.6, 120.1.

MS (*m/z*) 222 (86), 195 (16), 105 (10), 91 (100), 77 (21), 64 (46), 51 (12).

Anal. Calcd for $C_{13}H_{10}N_4$ (222.25) C, 70.26; H, 4.54; N, 25.21. Found C, 70.56; H, 4.55; N, 25.23.



2-(1-Phenyl-1*H*-1,2,4-triazol-3-yl)pyridine (3q)

Yield 79 mg (71%); Pale yellow solid; mp 126-128 °C.

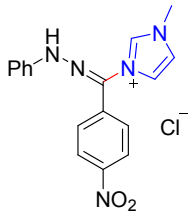
IR (KBr, cm^{-1}) 3090, 1598, 1589, 1499, 1385, 1335, 1256, 1237, 1073, 974.

^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.76 (s, 1H), 8.65 (s, 2H), 8.21 (d, *J* = 7.8 Hz, 2H), 7.78 (m, 3H), 7.54-7.44 (m, 2H), 7.38 (t, *J* = 7.5 Hz, 1H), 7.32 (t, *J* = 5 Hz, 1H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 162.9, 150.2, 149.6, 142.0, 137.1, 137.0, 129.8, 128.4, 124.2, 122.1, 120.3.

MS (*m/z*) 222 (78), 195 (17), 105 (19), 91 (100), 77 (19), 64 (43), 51 (10).

Anal. Calcd for $C_{13}H_{10}N_4$ (222.25) C, 70.26; H, 4.54; N, 25.21. Found C, 70.57; H, 4.54; N, 25.25.



1-Methyl-3-((4-nitrophenyl)(2-phenylhydrazineylidene)methyl)-1*H*-imidazol-3-ium chloride (4h)

Yield 148 mg (83%); yellow solid; mp 219 °C (decomposed).

IR (KBr, cm^{-1}) 3451, 3094, 2923, 1594, 1523, 1512, 1459, 1414, 1319, 1243, 1100, 1067.

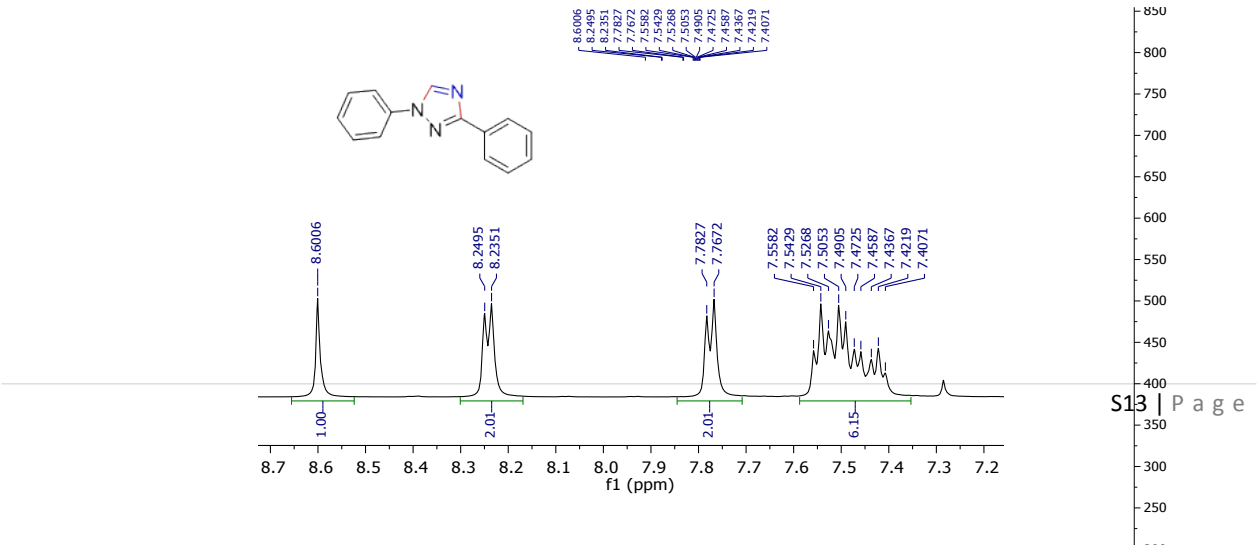
^{1}H NMR (500 MHz, DMSO-d_6) δ 11.35 (s, 1H), 9.74 (s, 1H), 8.24 (d, *J* = 9.0 Hz, 2H), 8.15 (s, 1H), 7.99 (s, 1H), 7.63 (d, *J* = 8.4 Hz, 2H), 7.55 (d, *J* = 8.0 Hz, 2H), 7.33 (t, *J* = 7.8 Hz, 2H), 6.98 (t, *J* = 7.4 Hz, 1H), 4.01 (s, 3H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 146.6, 143.6, 139.3, 138.9, 129.1, 125.5, 125.0, 124.1, 122.5, 122.3, 122.0, 114.5, 36.5.

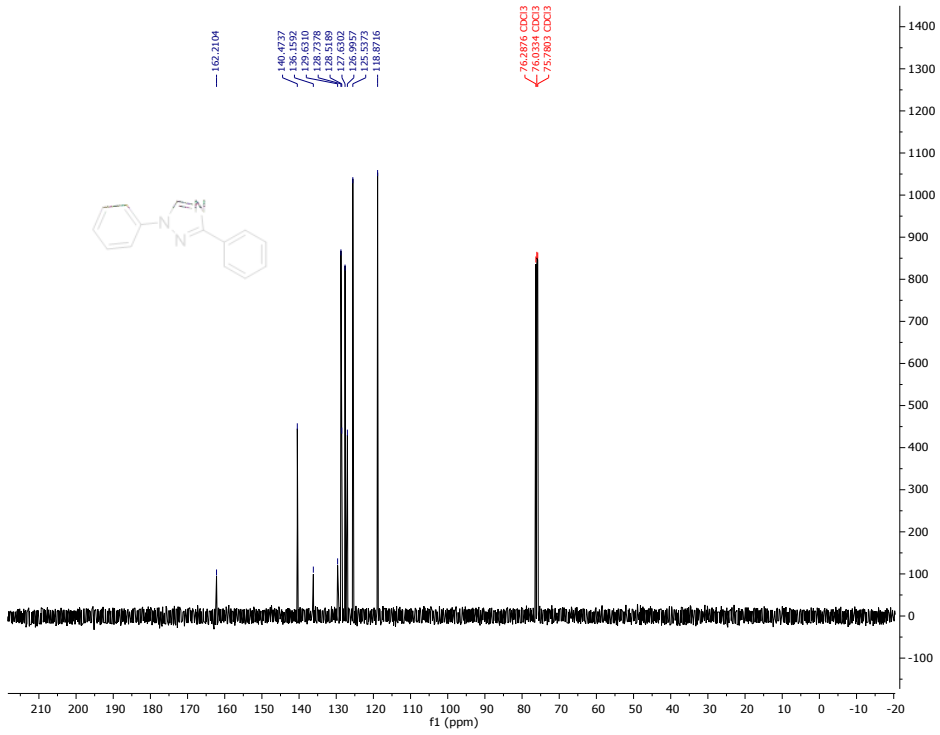
MS (*m/z*) 275 (57), 266 (73), 239 (16), 91 (100), 82 (50), 77 (34), 65 (57), 42 (15).

Anal. Calcd for $C_{17}H_{16}ClN_5O_2$ (357.80) C, 57.07; H, 4.51; N, 19.57. Found C, 57.36; H, 4.55; N, 25.29.

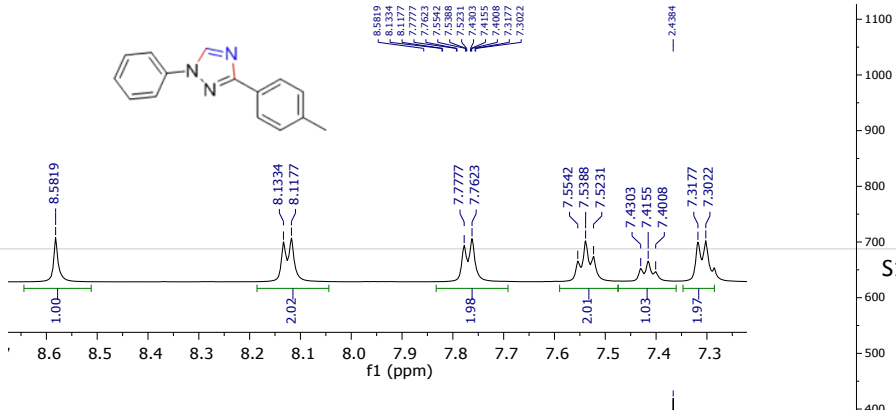
NMR spectra



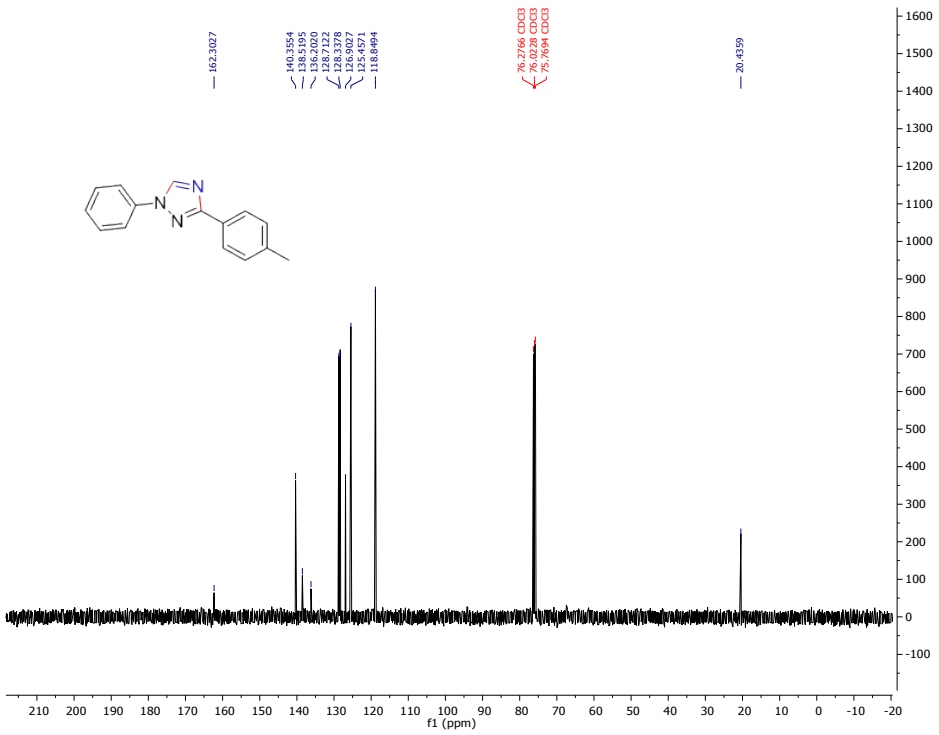
^{1}H NMR (500 MHz) of Compound **3a** in $CDCl_3$



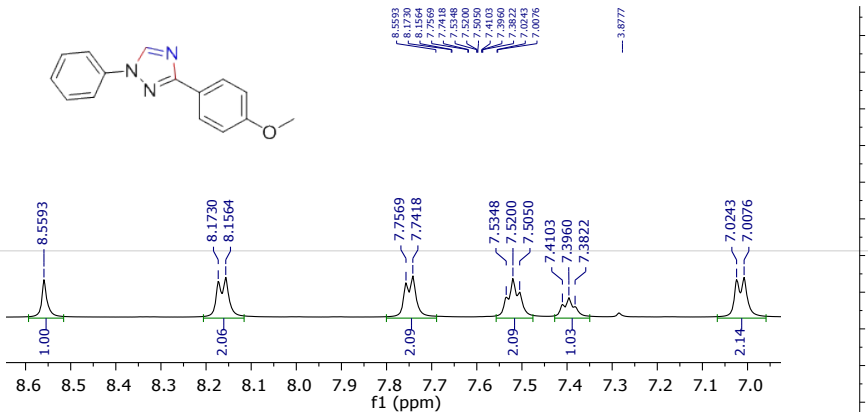
^{13}C NMR (126 MHz) of Compound **3a** in $CDCl_3$



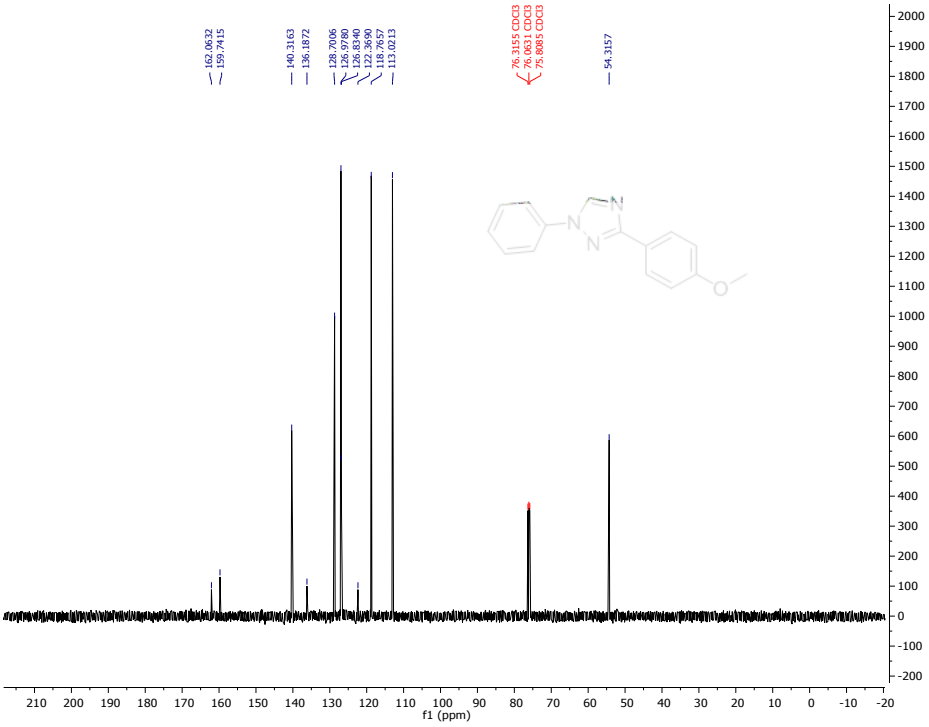
^{1}H NMR (500 MHz) of Compound **3b** in $CDCl_3$



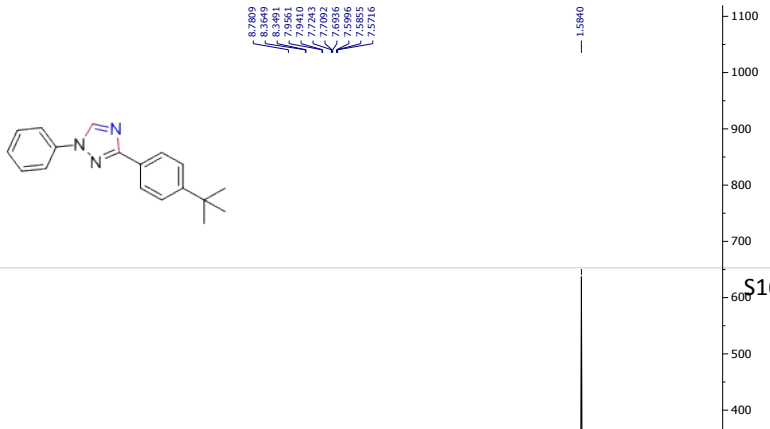
^{13}C NMR (126 MHz) of Compound **3b** in $CDCl_3$

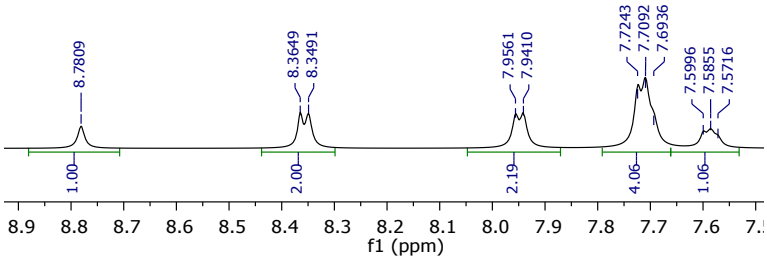


^{1}H NMR (500 MHz) of Compound **3c** in $CDCl_3$

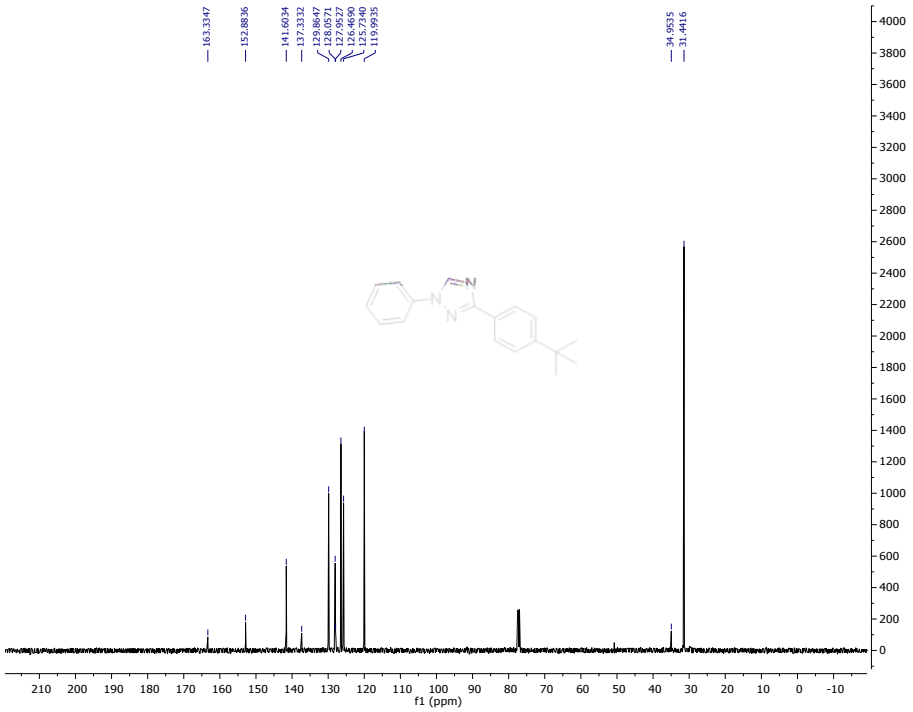


^{13}C NMR (126 MHz) of Compound **3c** in $CDCl_3$

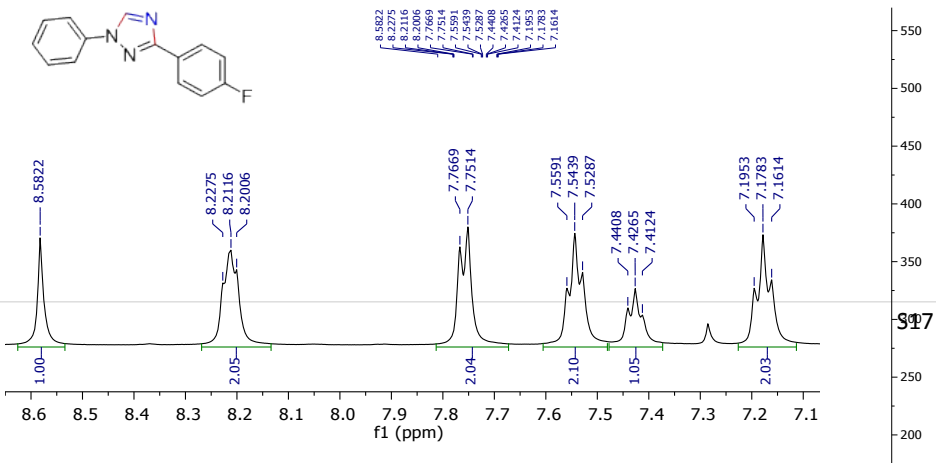




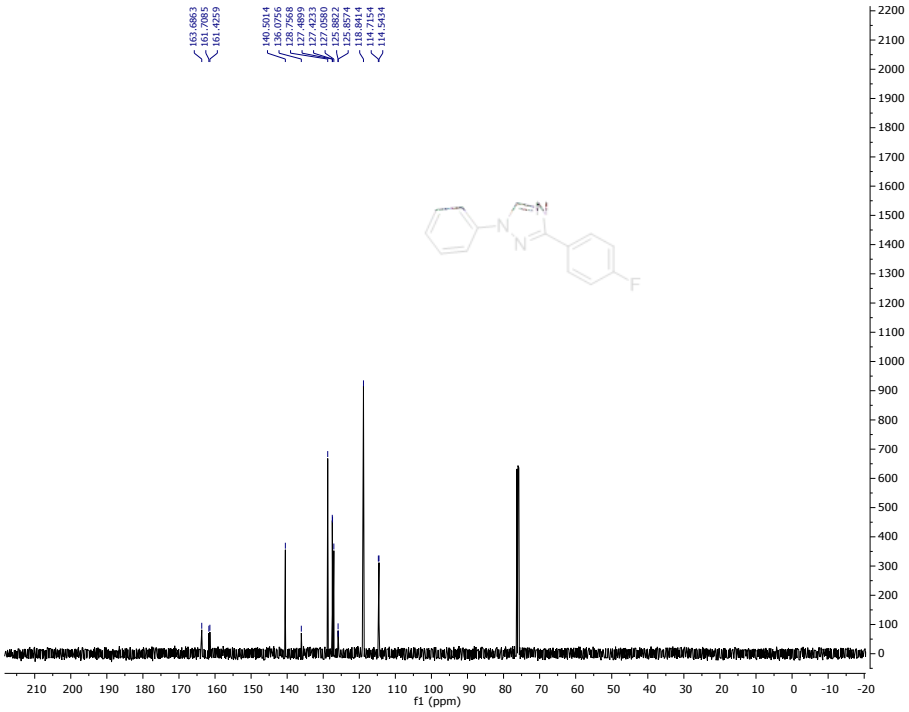
^{1}H NMR (500 MHz) of Compound **3d** in $CDCl_3$



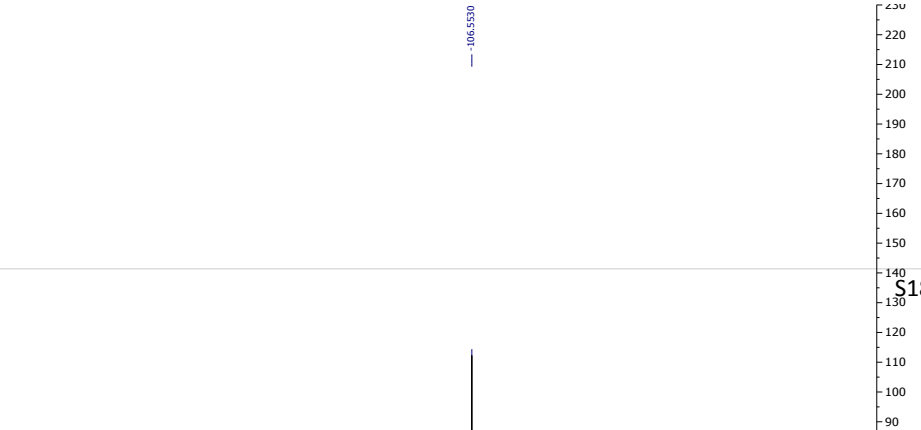
^{13}C NMR (126 MHz) of Compound **3d** in $CDCl_3$



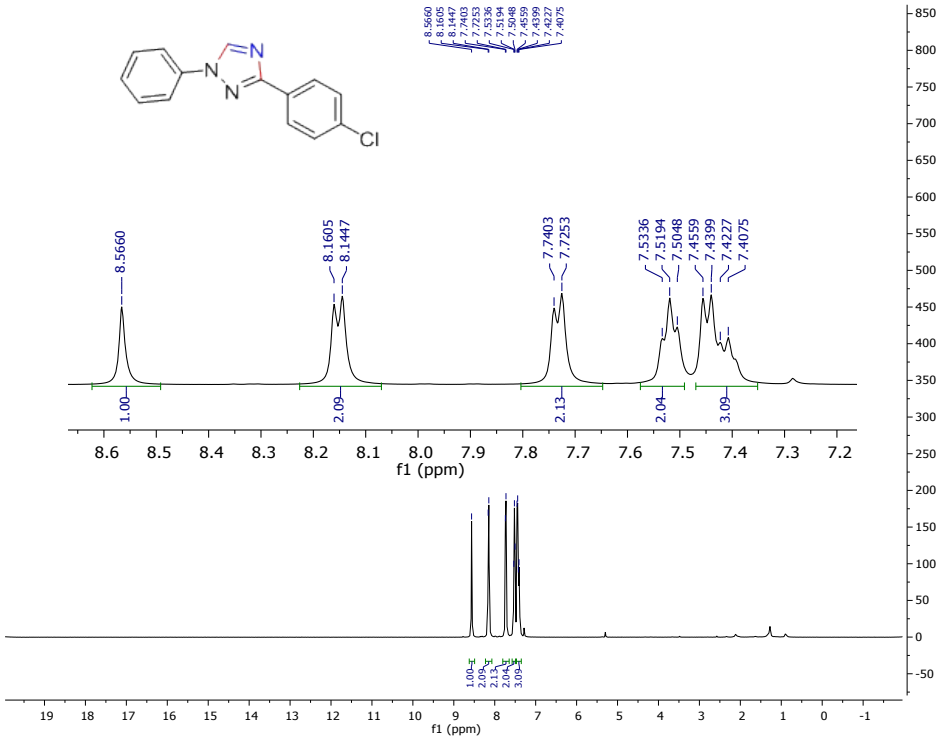
^{1}H NMR (500 MHz) of compound **3e** in $CDCl_3$



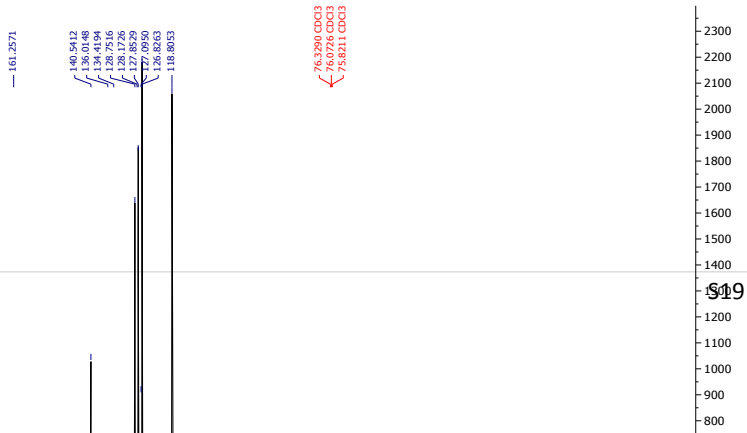
^{13}C NMR (126 MHz) of compound **3e** in $CDCl_3$



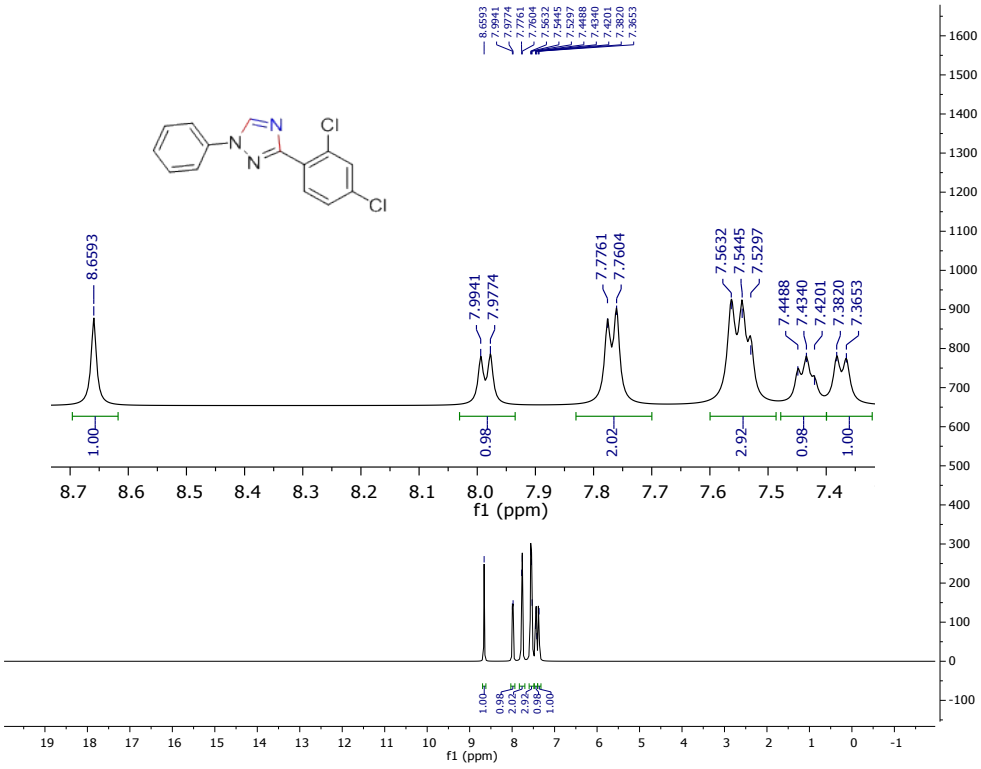
^{19}F NMR (471 MHz) of compound **3e** in $CDCl_3$



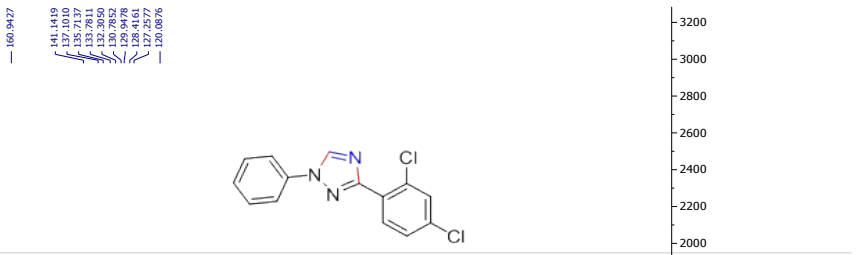
^{1}H NMR (500 MHz) of compound **3f** in $CDCl_3$



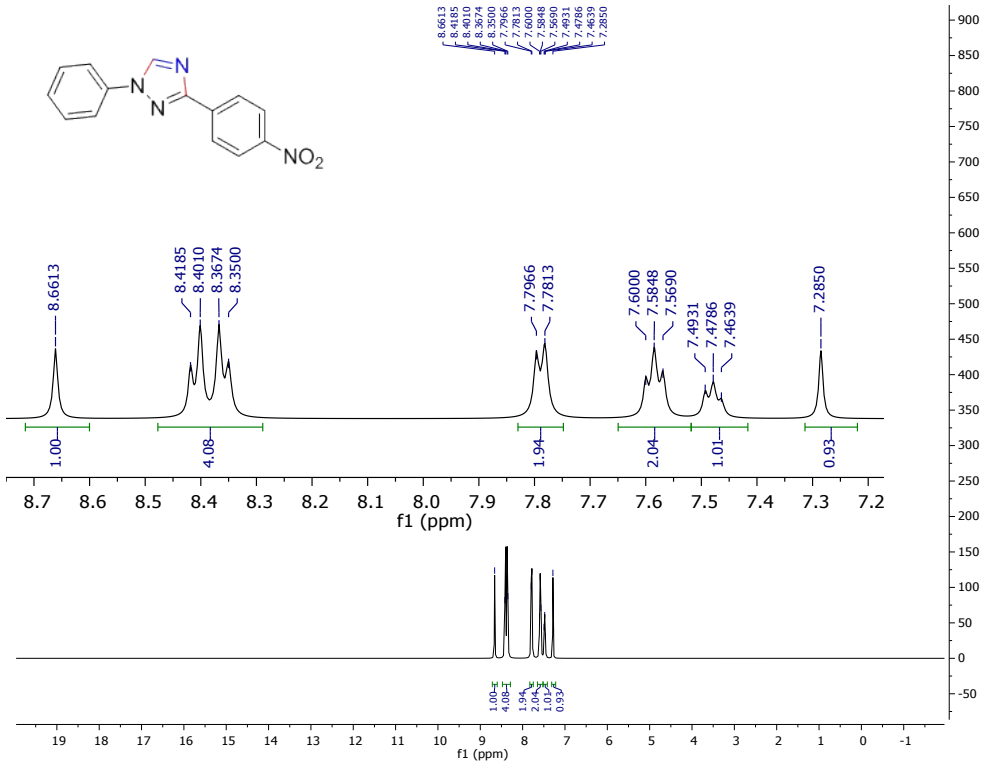
^{13}C NMR (126 MHz) of compound **3f** in $CDCl_3$



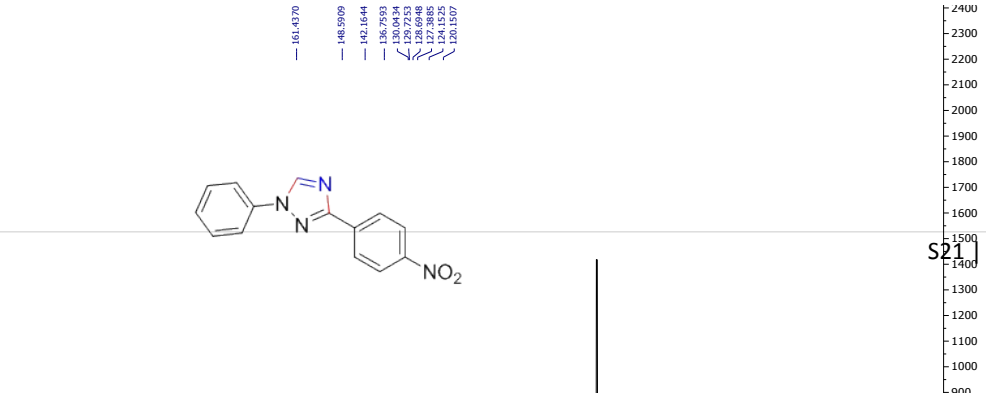
^{1}H NMR (500 MHz) of compound **3g** in $CDCl_3$



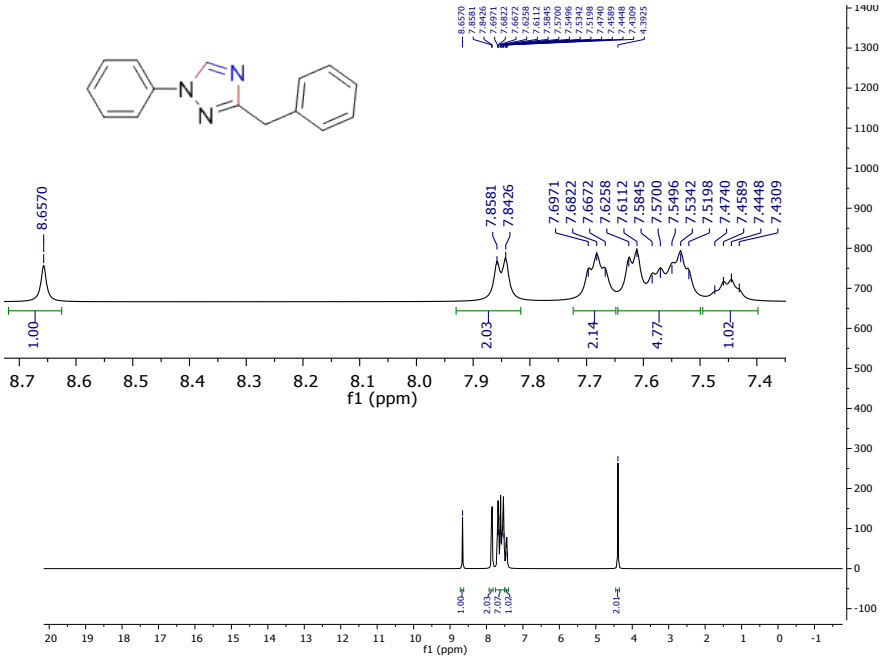
^{13}C NMR (126 MHz) of compound **3g** in $CDCl_3$



^{1}H NMR (500 MHz) of compound **3h** in $CDCl_3$

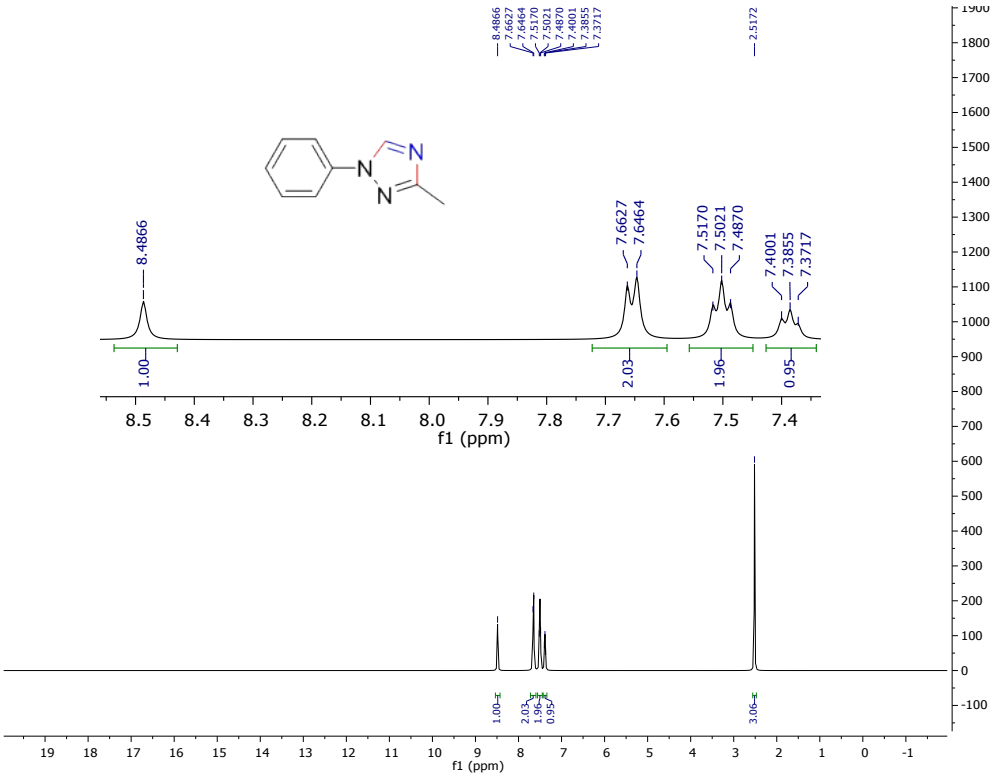


^{13}C NMR (126 MHz) of compound **3h** in $CDCl_3$

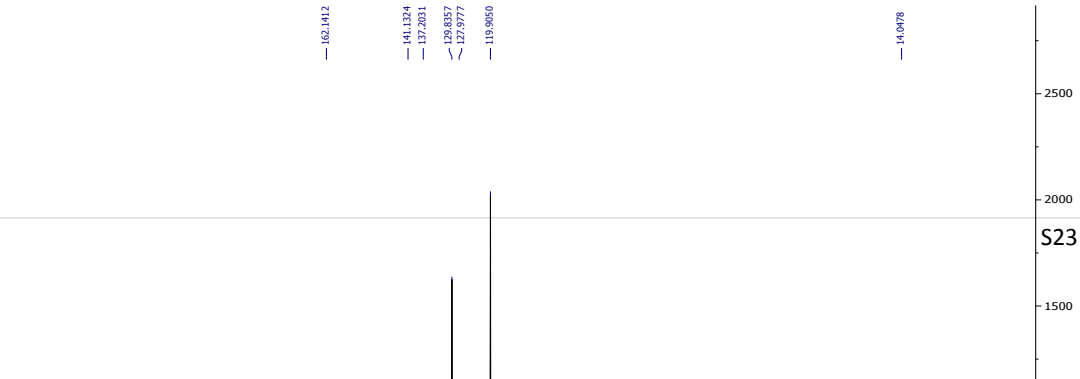


^{1}H NMR (500 MHz) of compound **3i** in $CDCl_3$

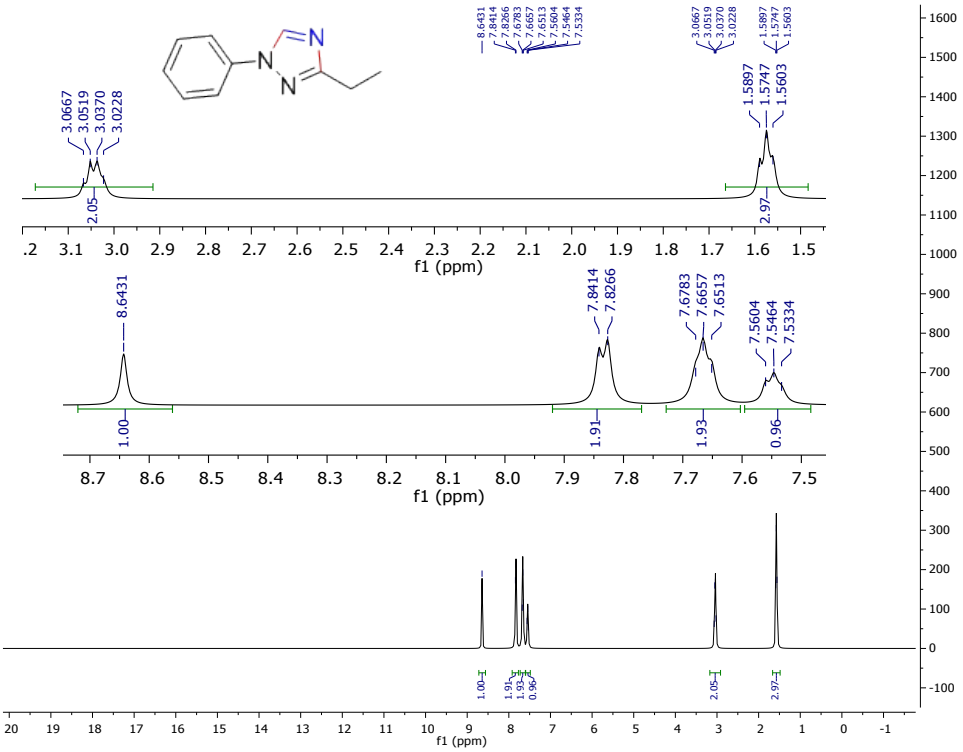
^{13}C NMR (126 MHz) of compound **3i** in $CDCl_3$



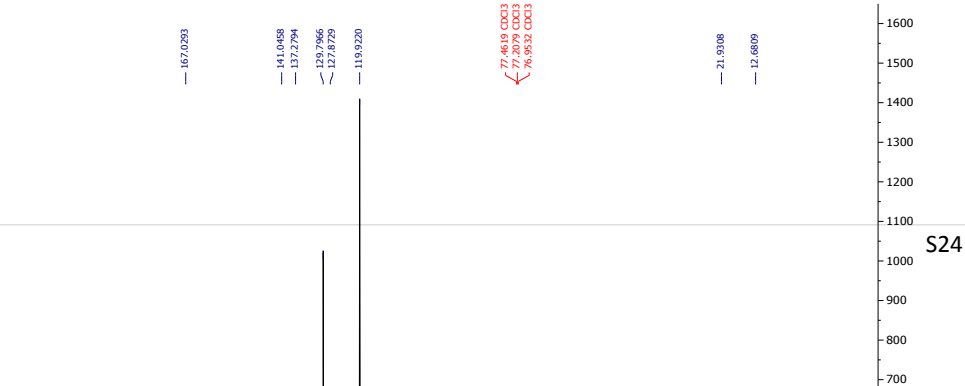
^{1}H NMR (500 MHz) of compound **3j** in $CDCl_3$



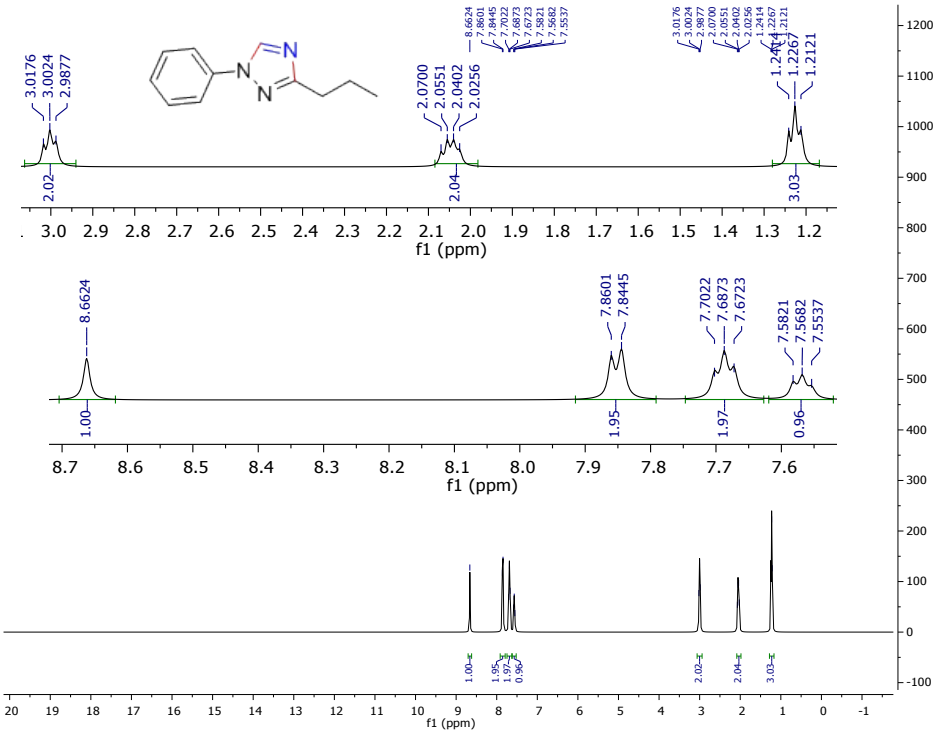
^{13}C NMR (126 MHz) of compound **3j** in $CDCl_3$



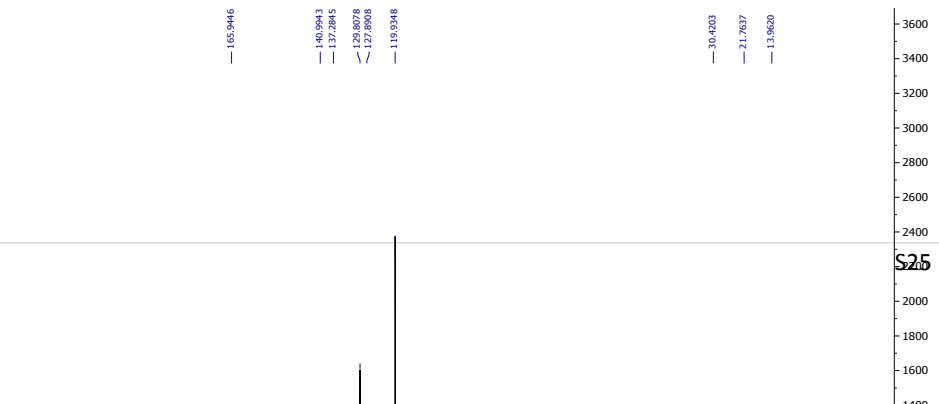
^{1}H NMR (500 MHz) of compound **3k** in $CDCl_3$



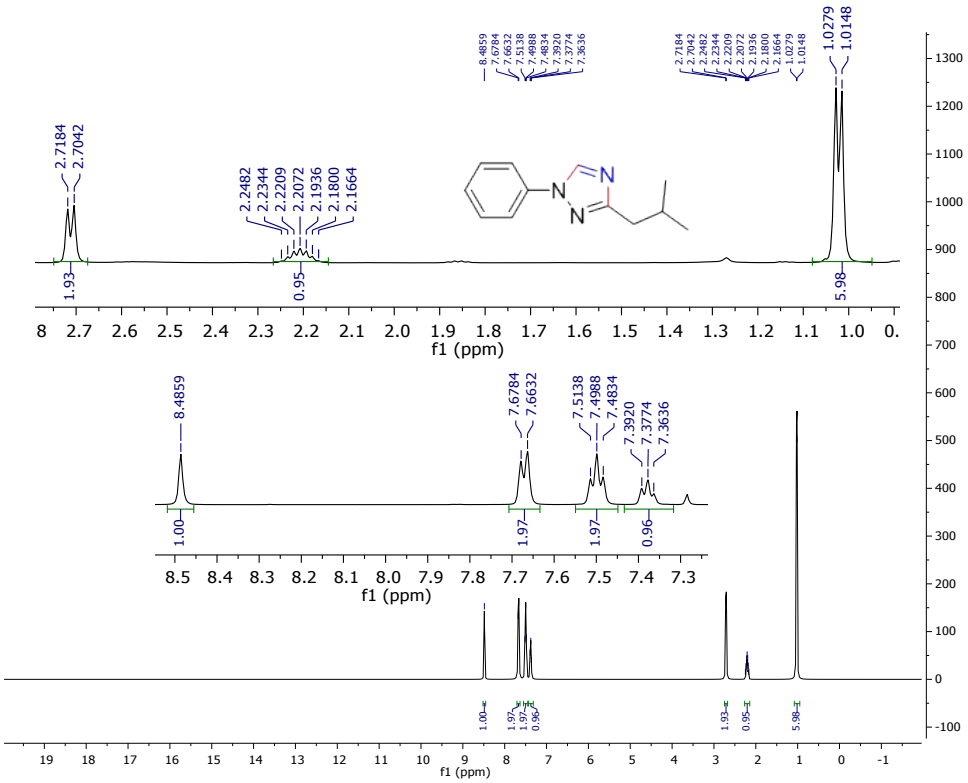
^{13}C NMR (126 MHz) of compound **3k** in $CDCl_3$



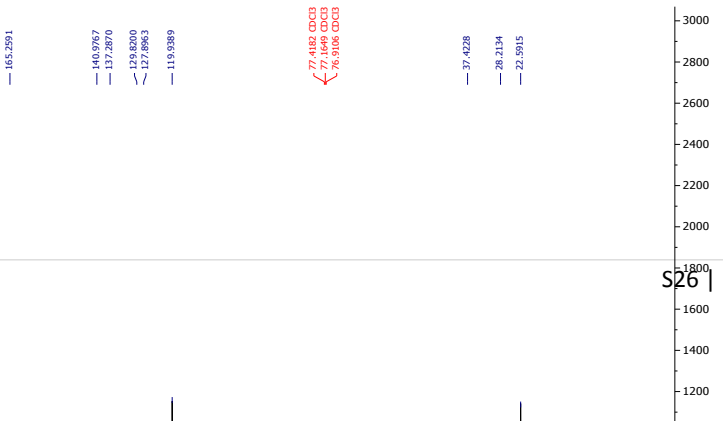
^{1}H NMR (500 MHz) of compound **3l** in $CDCl_3$



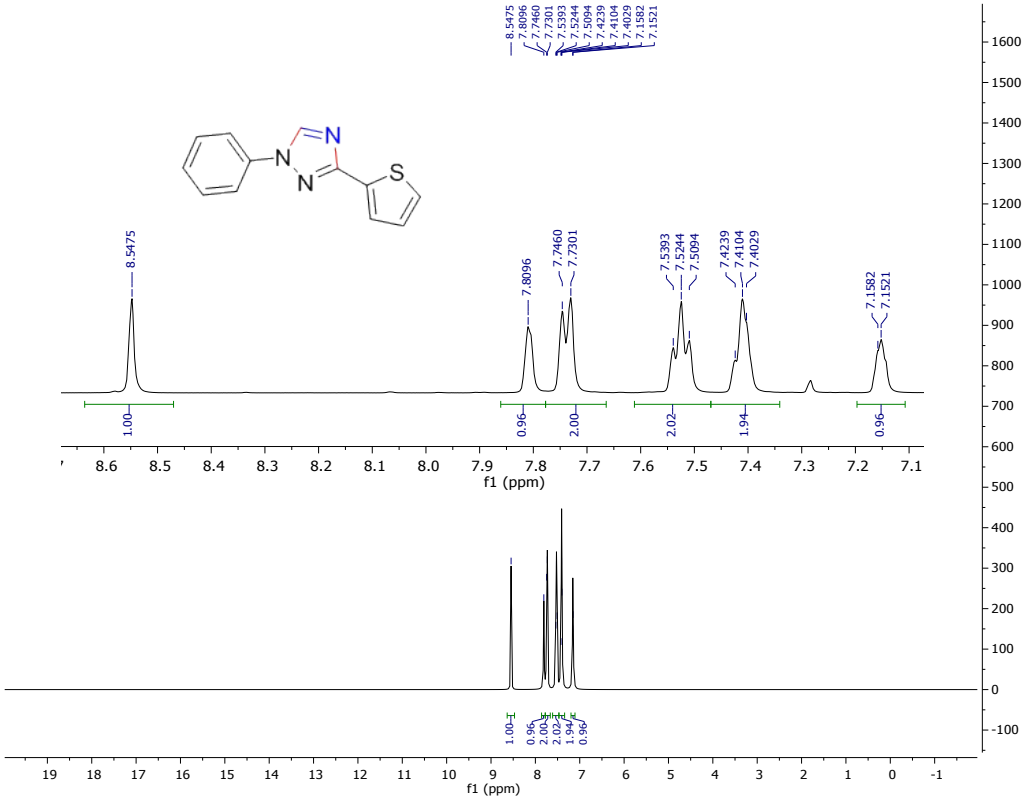
^{13}C NMR (126 MHz) of compound **3l** in $CDCl_3$



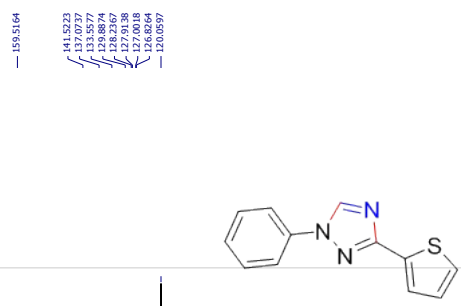
^{1}H NMR (500 MHz) of compound **3m** in $CDCl_3$



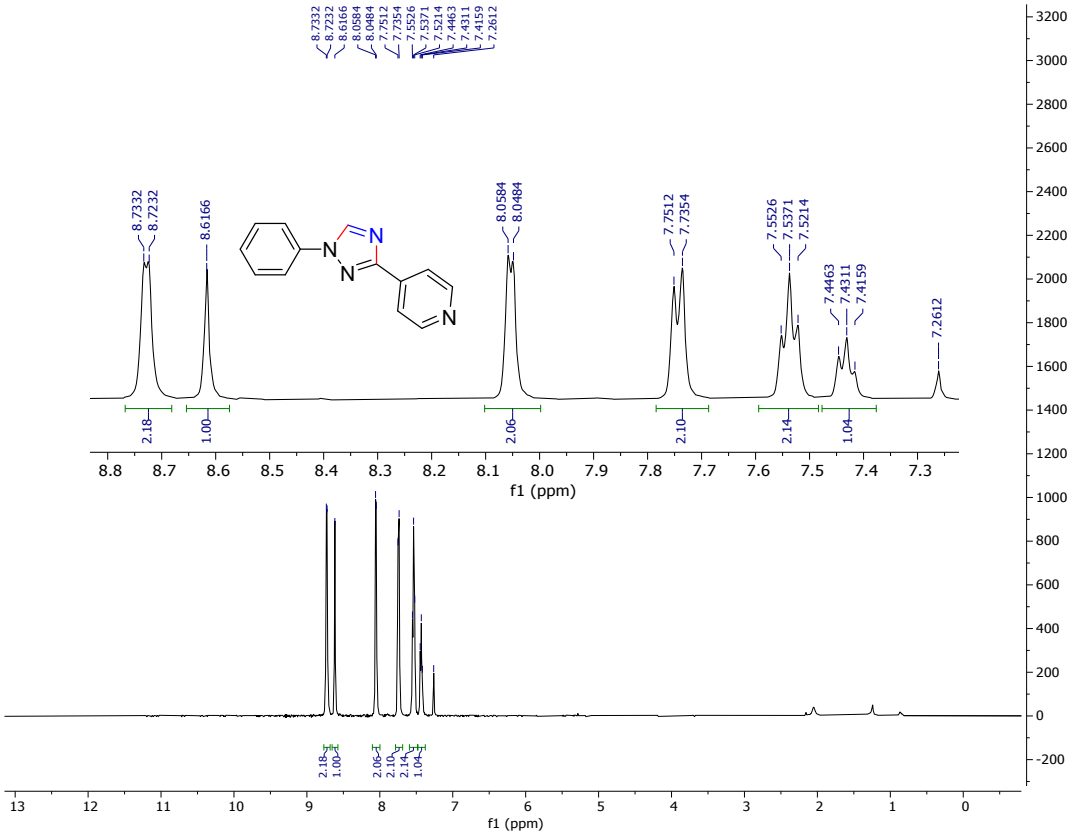
^{13}C NMR (126 MHz) of compound **3m** in $CDCl_3$



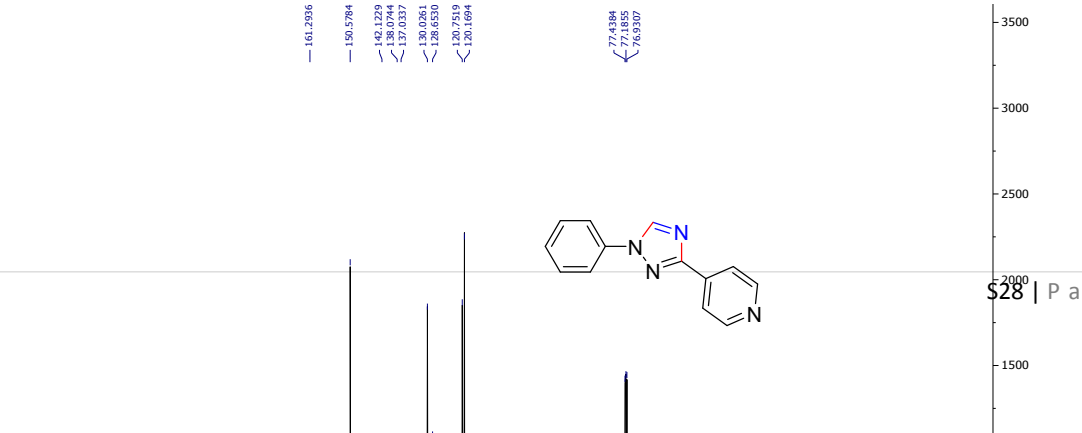
^{1}H NMR (500 MHz) of compound **3n** in $CDCl_3$



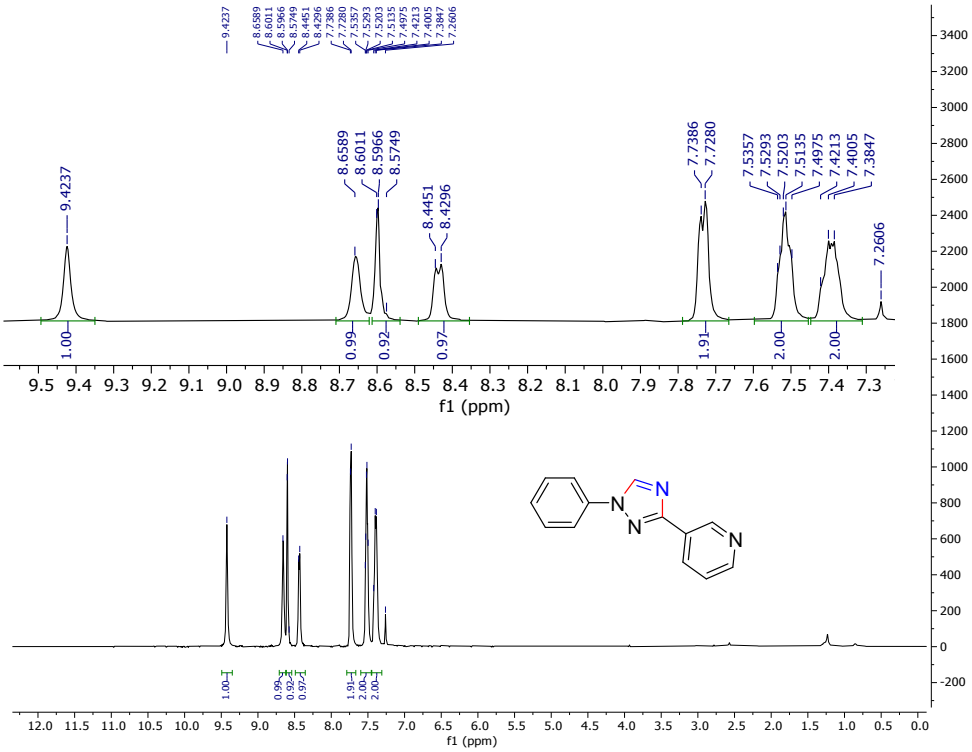
^{13}C NMR (126 MHz) of compound **3n** in $CDCl_3$



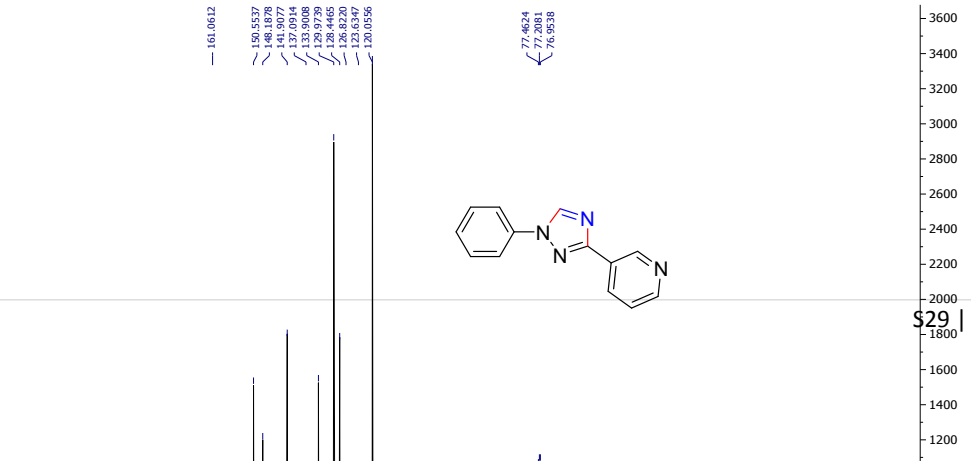
^{1}H NMR (500 MHz) of compound **3o** in $CDCl_3$



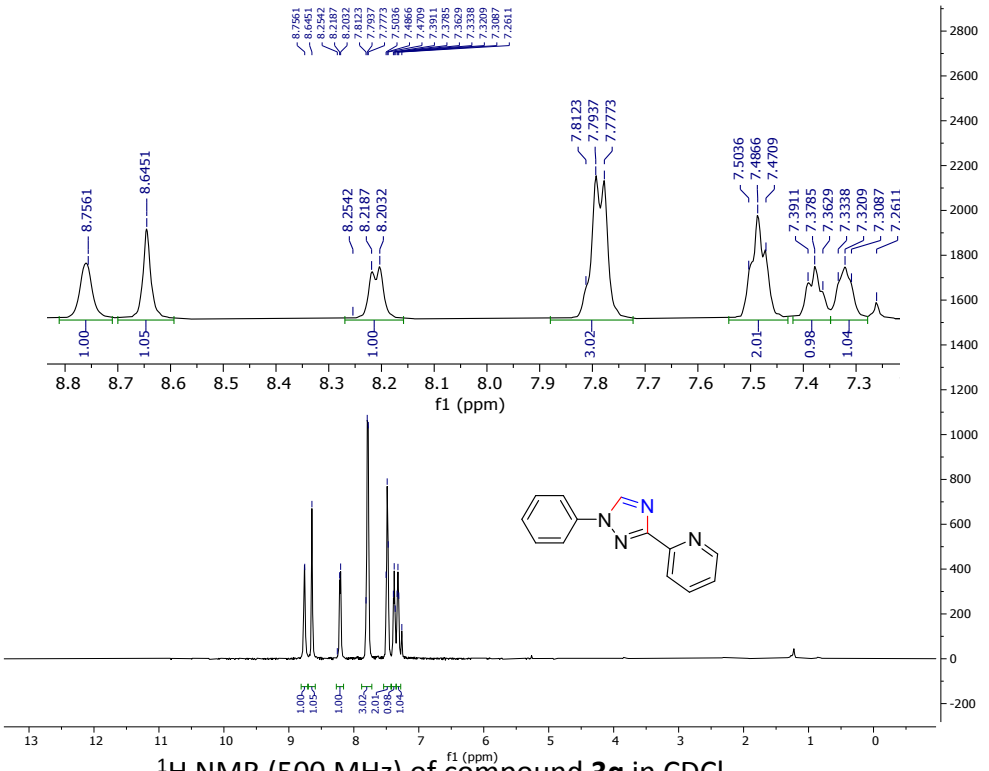
^{13}C NMR (126 MHz) of compound **3o** in $CDCl_3$



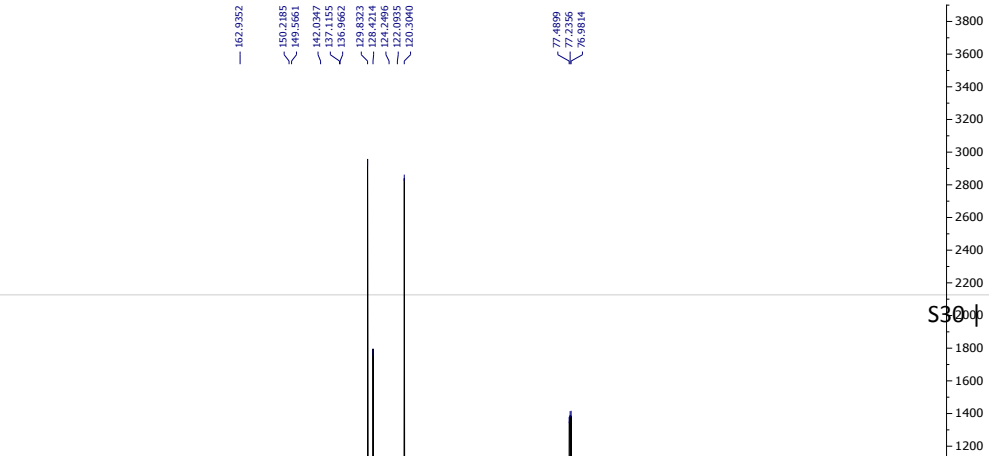
^{1}H NMR (500 MHz) of compound **3p** in $CDCl_3$

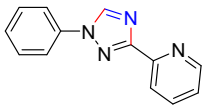


^{13}C NMR (126 MHz) of compound **3p** in $CDCl_3$

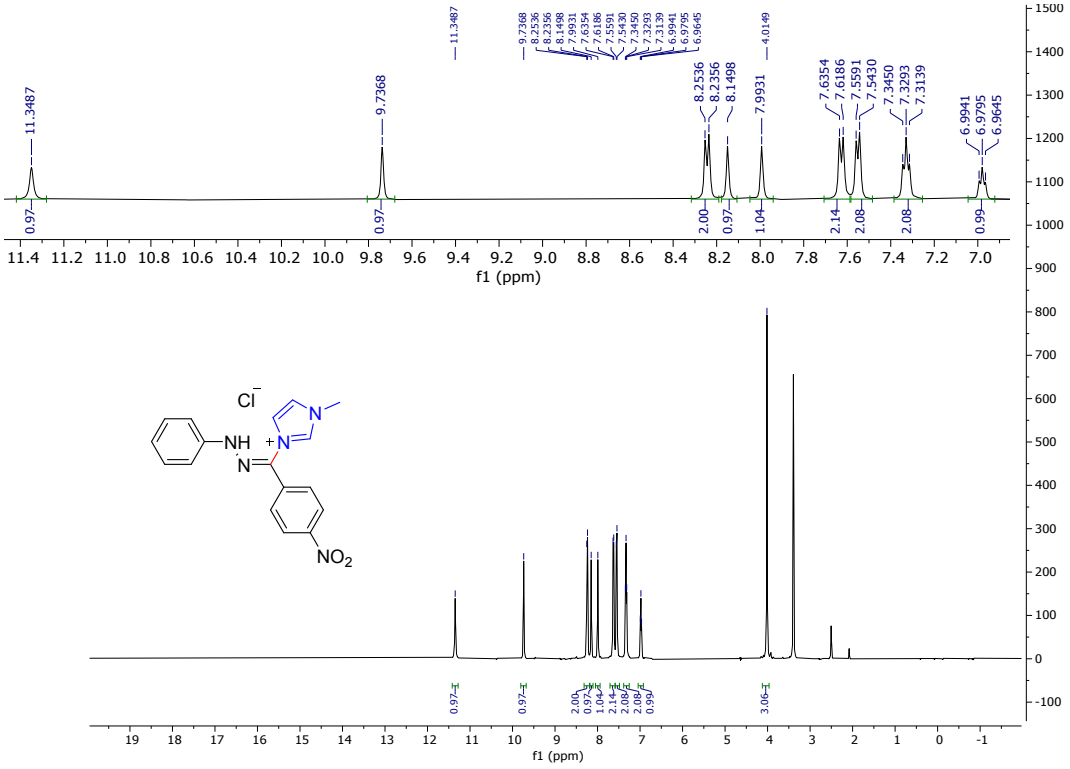


^{1}H NMR (500 MHz) of compound **3q** in $CDCl_3$

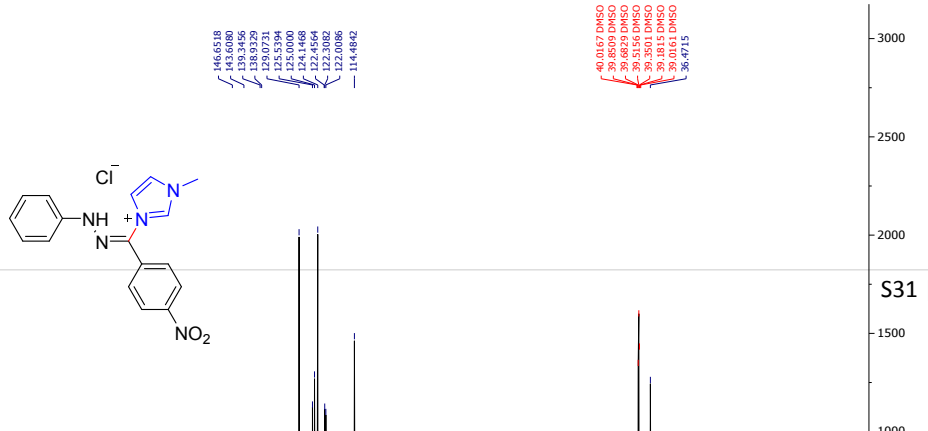




^{13}C NMR (126 MHz) of compound **3q** in $CDCl_3$



^{1}H NMR (500 MHz) of stable intermediate **4h** in DMSO-d_6



^{13}C NMR (126 MHz) of stable intermediate **4h** in DMSO-d_6

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