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A green polyol approach for the synthesis of Cu_2O NPs adhered on graphene oxide: A robust and efficient catalyst for 1,2,4-triazoles and imidazo[1,2-a]pyridines synthesis

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Contents

Experimental Section

Graphite mesh, sulfuric acid, sodium nitrate, potassium permanganate, hydrogen peroxide (30%), $CuNO_3$. $6H_2O$ and ethylene glycol were purchased from Alfa Aesar, TCI, Sigma-Aldrich and Loba. FTIR spectroscopic analysis was recorded on a Perkin Elmer spectrometer between 400 and 4000 cm−1 using KBr pellets. FT-Raman spectroscopic analysis was recorded on Bruker RFS 27: stand-alone FT-Raman spectrometer between 50 and 4000 cm−1 with 2 cm−1 resolutions. The surface morphology was observed by field emission microscope (FESEM) of model FESEM Supra 55 (Carl Zeiss, Germany). The energy-dispersive X-ray spectroscopy (EDX) analysis was carried out on an electron probe microscope (Oxford X-Maxn). Powdered X-ray diffraction (PXRD) of the catalyst was done by PANalytical's X'Pert PRO with CuKα radiation (λ = 0.15418 nm). High-resolution transmission electron microscope (HRTEM) image was observed using model Jeol/JE 2100. X-ray photoelectron (XPS) analysis was performed on a PHI 5000 Versa Probe II, FEI Inc. The thermogravimetric analysis was done using EXSTAR, SI 6300 EXSTAR, Inductively coupled plasma–atomic emission spectroscopy (ICP-MS) was carried out using ICP-MS: Thermo Fisher iCAP RQ ICP-MS, Mass spectroscopy was carried out using the ESI method on the Bruker instrument.

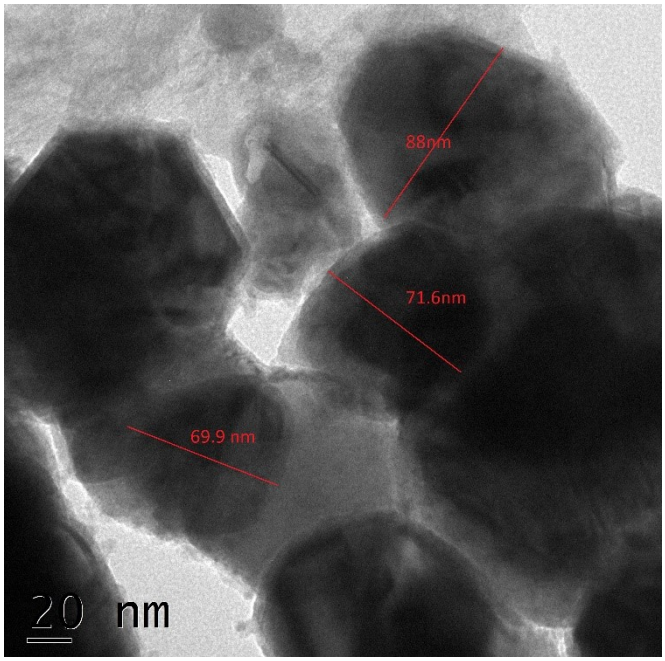
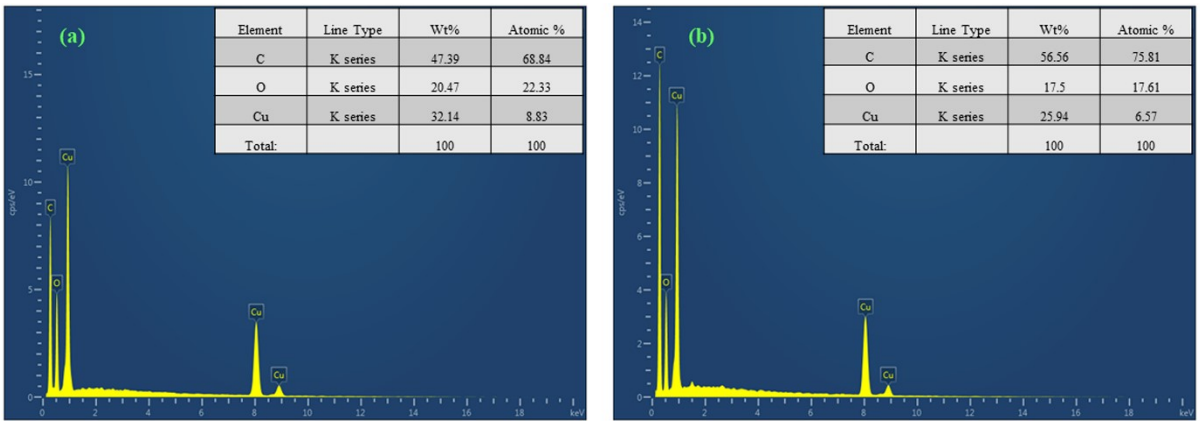


Fig. S1 HRTEM micrograph of Cu_2O-GO



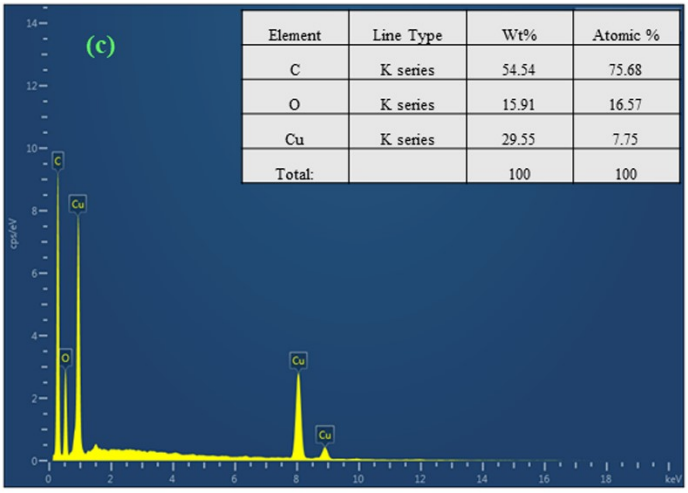


Fig. S2 EDX spectrum of (a) fresh Cu_2O@GO, (b) Cu_2O-GO for 1,2,4- triazole after the fifth cycle and (c) Cu_2O-GO for imidazo-[1,2a]pyridines after the fifth cycle

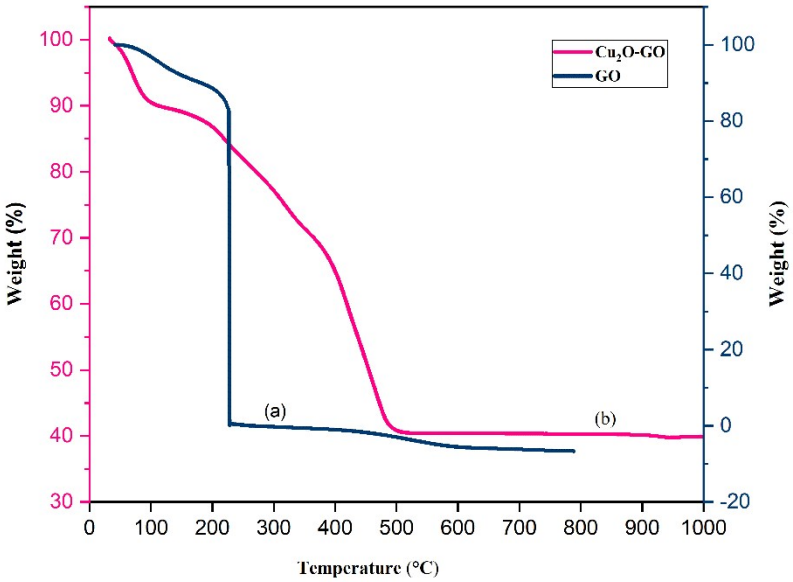


Fig. S3 TGA thermogram (a) Cu_2O@GO and (b) GO

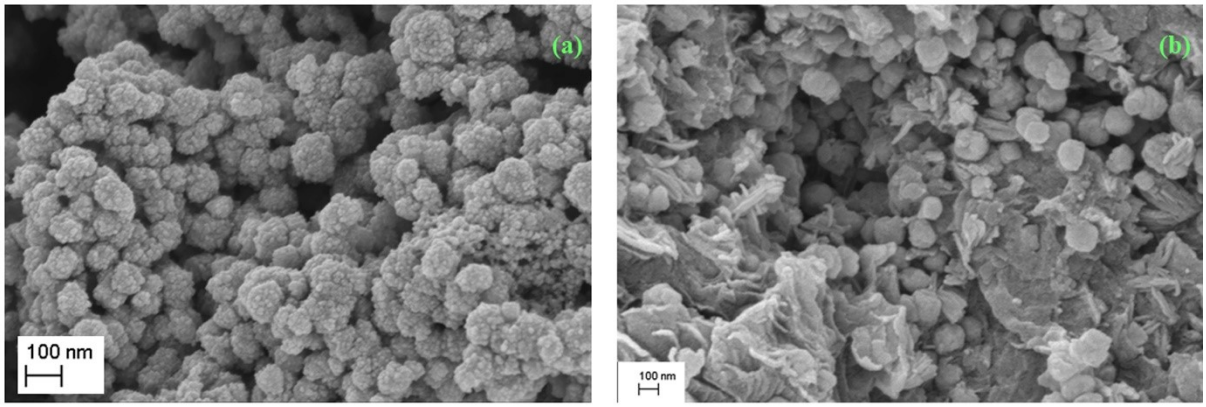


Fig. S4 FESEM image of Cu_2O-GO after five consecutive cycles (a) 1,2,4-triazoles and (b) imidazo-[1,2a]pyridine

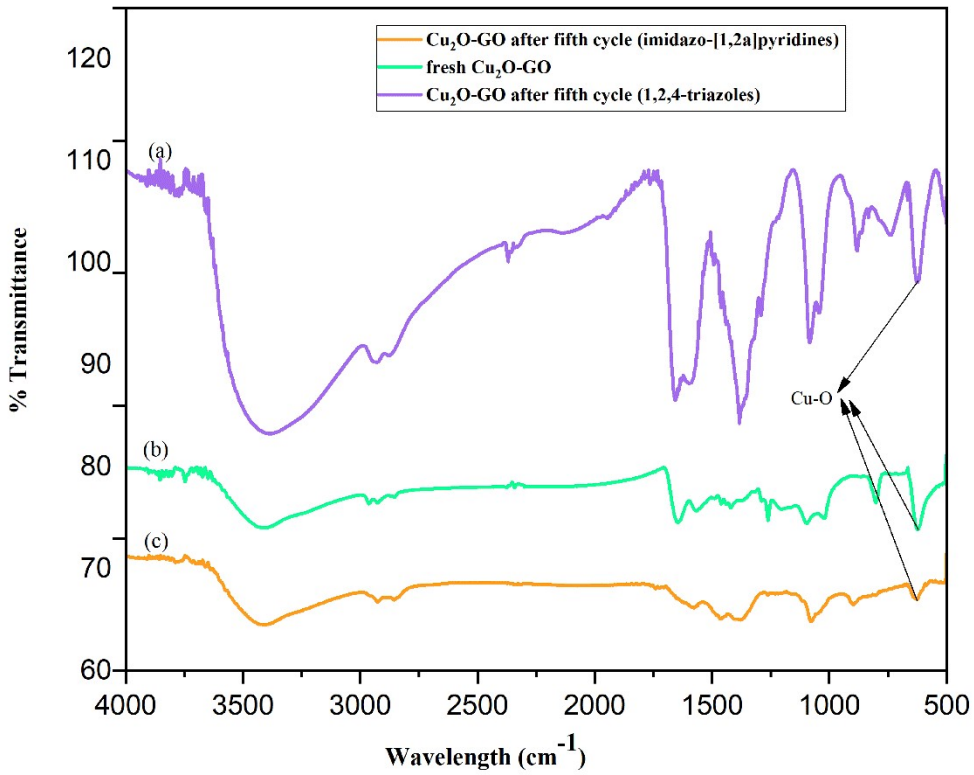
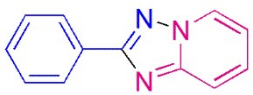


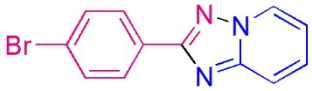
Fig. S5 FT-IR plot (a) Cu_2O-GO after the fifth cycle synthesis of 1,2,4-triazole, (b) Fresh Cu_2O-GO and (c) Cu_2O-GO after the fifth cycle synthesis of imidaz-[1,2a]pyridines.

Physical and spectral data 1,2,4-triazoles



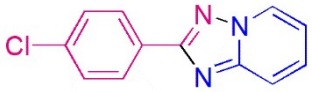
2-Phenyl-[1,2,4]triazolo[1,5-a]pyridine[1] (3a)

White solid, m.p. 134-135 °C;) δ_H (400 MHz, $CDCl_3$) 8.60 (1 H, d, *J* 6.8), 8.25 – 8.22 (2 H, m), 7.76 (1 H, d, *J* 9.0), 7.58 – 7.42 (4 H, m), 7.05 – 7.01 (1 H, m). ^{13}C NMR (100 MHz, $CDCl_3$) δ 164.13, 151.65, 130.72, 130.08, 129.50, 128.68, 128.29, 127.29, 116.35, 113.59.



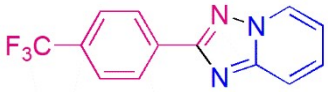
2-(4-Bromophenyl)-[1,2,4]triazolo[1,5-a]pyridine[2] (3b)

White solid, m.p. 229-230 °C; δ_H (400 MHz, $CDCl_3$) 8.52 (1 H, d, *J* 6.8), 8.09 (3 H, d, *J* 7.4), 7.69 (1 H, d, *J* 8.9), 7.56 (2 H, d, *J* 7.5), 7.49 – 7.43 (2 H, m), 6.99 – 6.94 (1 H, m). δ_C (100 MHz, $CDCl_3$) 163.29, 151.71, 131.95, 129.78, 129.74, 128.83, 128.37, 124.54, 116.47, 113.89.



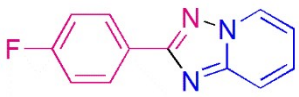
2-(4-Chlorophenyl)-[1,2,4]triazolo[1,5-a]pyridine[3] (3c)

White solid, 221-222 °C; δ_H (400 MHz, $CDCl_3$) 8.57 (2 H, d, *J* 6.8), 8.21 (3 H, d, *J* 8.6), 7.74 (2 H, d, *J* 9.0), 7.55 – 7.47 (3 H, m), 7.02 – 7.00 (1 H, m). ^{13}C NMR (100 MHz, $CDCl_3$) 163.21, 151.68, 136.09, 129.67, 129.28, 128.93, 128.56, 128.31, 116.40, 113.78.



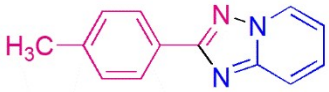
2-(4-(Trifluoromethyl)phenyl)-[1,2,4]triazolo[1,5-a]pyridine[3] (3d)

White solid, m. p. 235-236 °C; δ_H (400 MHz, $CDCl_3$) 8.61 (1 H, d, *J* 6.8), 8.39 (2 H, d, *J* 8.1), 7.80 – 7.72 (4 H, m), 7.57 – 7.51 (1 H, m), 7.08 – 7.02 (1 H, m). ^{13}C NMR (100 MHz, $CDCl_3$) 162.82, 151.75, 134.20, 131.93, 129.94, 128.46, 127.56, 125.68, 122.72, 116.65, 114.14, 109.45, 109.41.



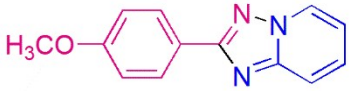
2-(4-Fluorophenyl)-[1,2,4]triazolo[1,5-a]pyridine[1] (3e)

White solid, m.p. 173-174; δ_H (400 MHz, $CDCl_3$) 8.58 (3 H, d, *J* 6.8), 8.27 (7 H, dd, *J* 8.9, 5.5), 7.74 (3 H, d, *J* 8.9), 7.53 – 7.50 (1 H, m), 7.18 – 7.16 (2 H, m), 7.02 – 6.99 (1 H, m). δ_C (100 MHz, $CDCl_3$) 165.35, 163.38, 151.73, 129.69, 129.33, 128.34, 127.02, 116.37, 115.68, 113.73.



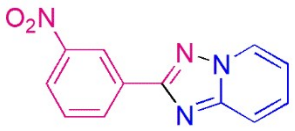
2-*p*-Tolyl-[1,2,4]triazolo[1,5-a]pyridine[4] (3f)

White solid, m.p. 167-168 °C, δ_H (400 MHz, $CDCl_3$) 8.58 (1 H, d, *J* 6.8), 8.17 (2 H, d, *J* 8.1), 7.74 (1 H, d, *J* 8.7), 7.51 – 7.46 (1 H, m), 7.30 (2 H, d, *J* 8.0), 7.00 – 6.96 (1 H, m), 2.42 (3 H, s). δ_C (100 MHz, $CDCl_3$) 164.32, 151.68, 140.27, 129.67, 129.48, 128.30, 127.94, 127.23, 116.31, 113.52, 21.52.



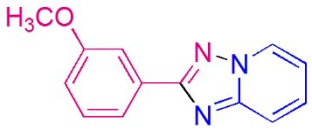
2-(4-Methoxyphenyl)-[1,2,4]triazolo[1,5-a]pyridine[1] (3g)

White solid, 135-136 °C, δ_H (400 MHz, $CDCl_3$) 8.48 (1 H, d, *J* 6.8), 8.14 (2 H, d, *J* 6.9), 7.64 (1 H, d, *J* 8.1), 7.41 – 7.37 (1 H, m), 6.93 (2 H, d, *J* 8.9), 6.90 – 6.86 (1 H, m), 3.79 (3 H, s). δ_C (100 MHz, $CDCl_3$) 163.90, 161.04, 151.56, 129.38, 128.76, 128.18, 123.32, 116.07, 114.08, 113.33, 55.43.



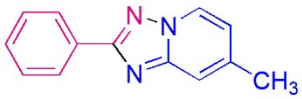
2-(3-Nitrophenyl)[1,2,4]triazolo[1,5-*a*]pyridine[5] (3h)

White solid, mp 281-282 °C, δ_H (400 MHz, $CDCl_3$) 9.11 – 9.07 (1 H, m), 8.59 – 8.54 (2 H, m), 8.28 – 8.24 (1 H, m), 7.74 (1 H, d, *J* 9.0), 7.64 – 7.59 (1 H, m), 7.54 – 7.49 (1 H, m), 7.05 – 7.00 (1 H, m). δ_C (100 MHz, $CDCl_3$) 162.08, 151.82, 132.99, 132.71, 130.13, 129.77, 128.53, 124.61, 122.35, 116.74, 114.35.



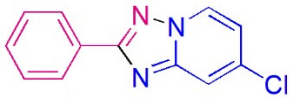
2-(3-Methoxyphenyl)-[1,2,4]triazolo[1,5-a]pyridine[1] (3i)

White solid, m.p. 106-108 °C, δ_H (400 MHz, $CDCl_3$) 8.49-8.46 (1 H, m), 7.79 (1 H, d, *J* 7.6), 7.66 – 7.63 (1 H, m), 7.35 (2 H, d, *J* 8.9), 7.33 – 7.31 (1 H, m), 6.95-6.93 (2 H, m), 6.84 (2 H, t, *J* 7.9), 3.81 (3 H, s). δ_C (100 MHz, $CDCl_3$) 163.90, 159.86, 151.51, 132.00, 129.73, 129.53, 128.25, 119.73, 116.67, 116.23, 113.64, 111.72, 55.35.



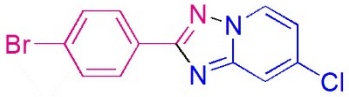
7-Methyl-2-phenyl[1,2,4]triazolo[1,5-*a*]pyridine[4] (3j)

White solid, m.p. 142-144 °C, δ_H (400 MHz, $CDCl_3$) 8.45 (1 H, d, *J* 6.9), 8.26 (2 H, dd, *J* 8.0, 1.6), 7.50 – 7.46 (4 H, m), 6.81 (1 H, dd, *J* 7.0, 1.6), 2.48 (3 H, s). δ_C (100 MHz, $CDCl_3$) 164.20, 151.91, 141.06, 130.91, 130.00, 128.70, 127.25, 116.12, 115.00, 21.63.



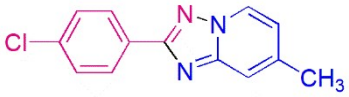
7-Chloro-2-phenyl-[1,2,4]triazolo[1,5-a]pyridine[5] (3k)

White solid, m.p. 189-190°C δ_H (400 MHz, $CDCl_3$) 8.48 (1 H, d, *J* 7.9), 8.25 – 8.23 (2 H, m), 7.73 (1 H, d, *J* 2.7), 7.49 – 7.47 (3 H, m), 6.96 (1 H, dd, *J* 7.2, 2.2). δ_C (100 MHz, $CDCl_3$) 165.23, 151.83, 136.21, 130.42, 128.78, 128.47, 127.37, 115.45, 115.18



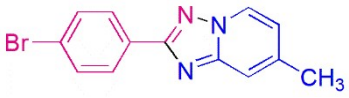
2-(4-Bromophenyl)-7-chloro-[1,2,4]triazolo[1,5-a]pyridine (3l) new

White solid, m.p. 195-197 °C, δ_H (400 MHz, $CDCl_3$) 8.49 (1 H, d, *J* 7.2), 8.13 – 8.11 (2 H, m), 7.73 (1 H, dd, *J* 2.1, 0.6), 7.64 – 7.61 (2 H, m), 7.01 (1 H, dd, *J* 7.2, 2.1). δ_C (100 MHz, $CDCl_3$) 164.38, 151.87, 136.47, 132.02, 129.31, 128.89, 128.49, 124.89, 115.44. HRMS (M+H) 309.940 calculated for $C_{12}H_7BrClN_3$: 309.5685; found 309.9401



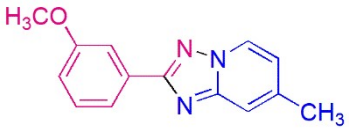
2-(4-Chlorophenyl)-7-methyl[1,2,4]triazolo[1,5-*a*]pyridine[5] (3m)

White solid, m.p. 169-171 °C, δ_H (400 MHz, $CDCl_3$) 8.39 (2 H, d, *J* 7.0), 8.18 – 8.15 (3 H, m), 7.46 – 7.41 (5 H, m), 6.79 (2 H, dd, *J* 6.9, 1.6), 2.44 (3 H, s). δ_C (100 MHz, $CDCl_3$) 163.17, 151.87, 141.26, 135.94, 129.45, 128.90, 128.51, 127.25, 116.28, 114.96, 21.61.



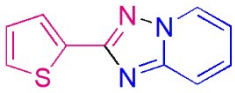
2-(4-Bromophenyl)-7-methyl[1,2,4]triazolo[1,5-*a*]pyridine[6] (3n)

White solid, m.p. 179-181 °C, δ_H (400 MHz, $CDCl_3$) 8.43-8.38 (1 H, m), 8.12 – 8.08 (2 H, m), 7.61 – 7.45 (3 H, m), 6.81 (1 H, dd, *J* 6.9, 1.7), 2.46 (3 H, s). δ_C (100 MHz, $CDCl_3$) 163.22, 151.87, 141.33, 131.87, 129.88, 128.98, 127.76, 124.36, 116.34, 114.98, 113.42, 21.64. HRMS-ESI: *m/z* [M + H]+ calcd for C13H10BrN3: 288.0131; found: 288.0034



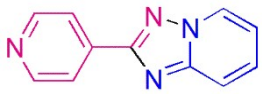
2-(3-Methoxyphenyl)-7-methyl[1,2,4]triazolo[1,5-a]pyridine [7] (3o)

White solid, m.p. 143-144 °C δ_H (400 MHz, $CDCl_3$) 8.54 (1 H, d, *J* 6.8), 7.82 (1 H, d, *J* 7.6), 7.76 – 7.75 (1 H, m), 7.70 (2 H, d, *J* 8.9), 7.47 – 7.43 (2 H, m), 7.34 (2 H, t, *J* 7.9), 6.97 – 6.92 (3 H, m), 3.85 (3 H, s). δ_C (100 MHz, $CDCl_3$) 164.10, 159.94, 151.66, 132.03, 129.82, 129.62, 128.36, 119.82, 116.92, 116.44, 113.73, 111.67, 55.47.



2-(Thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyridine[5] (3p)

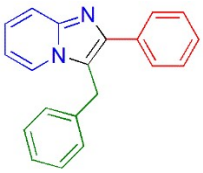
White solid, m.p. 155-156 °C, δ_H (400 MHz, $CDCl_3$) 8.55 (1 H, d, *J* 6.8), 7.88 (1 H, dd, *J* 3.6, 1.2), 7.71 (1 H, d, *J* 9.0), 7.56 – 7.48 (1 H, m), 7.46 (1 H, dd, *J* 5.0, 1.2), 7.15 (1 H, dd, *J* 5.0, 3.7), 6.98 (1 H, td, *J* 6.9, 1.2). δ_C (100 MHz, $CDCl_3$) 160.23, 151.50, 133.54, 129.89, 128.23, 128.04, 127.91, 127.73, 116.18, 113.81.



2-(Pyridin-4-yl)-[1,2,4]triazolo[1,5-a]pyridine[2] (3q)

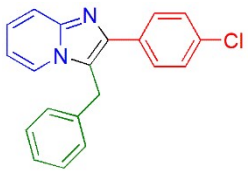
White solid m.p. 190-192 °C, δ_H (400 MHz, $CDCl_3$) 8.73 (3 H, d, *J* 6.0), 8.60 (1 H, d, *J* 6.8), 8.12 (2 H, d, *J* 6.0), 7.78 (1 H, d, *J* 9.0), 7.57 – 7.54 (1 H, m), 7.08 – 7.05 (1 H, m). δ_C (100 MHz, $CDCl_3$) 161.89, 151.75, 150.40, 138.29, 130.13, 128.53, 121.35, 116.85, 114.46, 110.57.

Physical and spectral data imidazo-[1,2a]pyridines



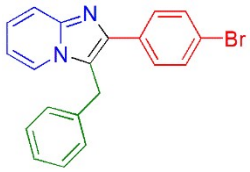
3-Benzyl-2-phenylimidazo[1,2-*a*]pyridine[8] (4a)

White solid, m.p. 118-119 °C, δ_H (400 MHz, $CDCl_3$) 7.79 (2 H, d, *J* 6.9), 7.75 – 7.69 (2 H, m), 7.45 – 7.41 (2 H, m), 7.39 – 7.24 (5 H, m), 7.23 – 7.10 (4 H, m), 6.75 (1 H, t, *J* 7.0), 4.51 (2 H, s). δ_C (100 MHz, $CDCl_3$) 144.61, 143.61, 136.61, 133.95, 129.11, 128.73, 128.43, 128.26, 127.71, 127.00, 124.70, 123.49, 117.74, 117.39, 112.355, 29.85.



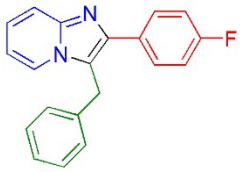
3-benzyl-2-(4-chlorophenyl)H-imidazo[1,2-a]pyridine[9] (4b)

White solid, m.p. 145-146 °C δ_H (400 MHz, $CDCl_3$) 7.72 – 7.67 (4 H, m), 7.38 (2 H, d, *J* 8.5), 7.30 – 7.24 (3 H, m), 7.18 – 7.15 (1 H, m), 7.11 (2 H, d, *J* 6.9), 6.72 – 6.68 (1 H, m), 4.44 (2 H, s). δ_C (100 MHz, $CDCl_3$) 144.90, 142.91, 136.50, 133.71, 133.00, 129.41, 128.86, 127.65, 127.07, 124.53, 123.44, 117.87, 117.54, 112.44, 29.81.



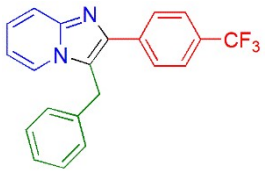
3-benzyl-2-(4bromophenyl)H-imidazo[1,2-a]pyridine[8] (4c)

White solid, m.p. 160-161 °C, δ_H (400 MHz, $CDCl_3$) 7.70 – 7.64 (4 H, m), 7.54 (2 H, d, *J* 8.5), 7.31 – 7.25 (3 H, m), 7.20 – 7.17 (1 H, m), 7.10 (2 H, s, *J* 6.9), 6.73 – 6.69 (1 H, m), 4.45 (2 H, s). δ_C (100 MHz, $CDCl_3$) 144.95, 143.00, 136.49, 133.51, 131.81, 129.70, 129.14, 127.65, 127.07, 124.50, 123.44, 121.94, 117.88, 117.59, 112.42, 29.84.



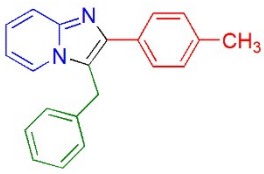
3-Benzyl-2-(4-fluorophenyl)imidazo[1,2-*a*]pyridine[10] (4d)

Yellow oil, δ_H (400 MHz, $CDCl_3$) 7.74 – 7.65 (4 H, m), 7.28 – 7.22 (3 H, m), 7.16 – 7.14 (1 H, m), 7.10 – 7.06 (4 H, m), 6.68 – 6.65 (1 H, m), 4.42 (2 H, s). δ_C (100 MHz, $CDCl_3$) 163.81, 144.79, 143.10, 136.58, 130.56, 129.92, 129.84, 129.13, 127.64, 127.04, 124.48, 123.44, 117.40, 1015.73, 112.38, 29.74.



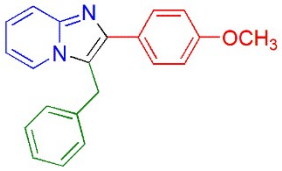
3-benzyl-2-(4-(trifluoromethyl)phenyl) imidazo[1,2-a]pyridine[9] (4e)

White solid, δ_H (400 MHz, $CDCl_3$) 7.90 (1 H, d, *J* 6.9), 7.67 (2 H, d, *J* 9.1), 7.46 – 7.39 (1 H, m), 7.34 – 7.32 (1 H, m), 7.28 (3 H, d, *J* 7.4), 7.14 (2 H, d, *J* 7.1), 7.12 – 7.08 (2 H, m), 6.73 (1 H, d, *J* 0.9), 4.48 (2 H, s). δ_C (100 MHz, $CDCl_3$) 144.99, 142.41, 136.25,129.19, 128.34, 127.60, 127.16, 125.62, 124.92, 123.52, 118.59, 117.68, 112.72, 29.79.



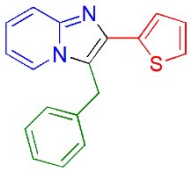
3-Benzyl-2-*p*-tolylimidazo[1,2-*a*]pyridine[9] (4f)

White solid, m.p. 160-161°C; δ_H (400 MHz, $CDCl_3$) 7.73 (2 H, d, *J* 9.1), 7.68 (5 H, d, *J* 8.1), 7.29 – 7.25 (6 H, m), 7.15 – 7.13 (3 H, m, *J* 7.0), 6.70 (2 H, t, *J* 6.3), 4.47 (2 H, s), 2.38 (3 H, s). δ_C (100 MHz, $CDCl_3$) 144.67, 143.73, 137.64, 136.77, 131.21, 129.43, 129.04, 128.17, 127.71, 126.92, 124.46, 123.49, 117.45, 117.34, 112.36, 29.75, 21.31.



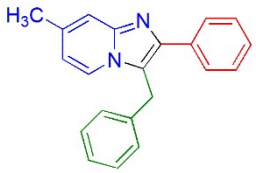
3-benzyl-2-(4-methoxyphenyl)H-imidazo[1,2-a]pyridine[10] (4g)

White solid, δ_H (400 MHz, $CDCl_3$) 7.76 (2 H, d, *J* 8.2), 7.71 – 7.69 (2 H, m), 7.31 – 7.26 (4 H, m), 7.22 – 7.12 (4 H, m), 6.96 (2 H, d, *J* 7.7), 6.75 – 6.72 (1 H, m), 4.68 (2 H, s), 3.83 (3 H, s). δ_C (100 MHz, $CDCl_3$) 159.54, 144.48, 136.64, 129.52, 129.10, 127.00, 126.23, 123.36, 117.08, 114.20, 112.57, 55.32, 29.81.



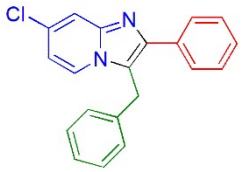
3-Benzyl-2-(2-thiophenyl)imidazo[1,2-*a*]pyridine[10] (4h)

Yellow oil, δ_H (400 MHz, $CDCl_3$) 7.72 (1 H, d, *J* 6.9), 7.66 (2 H, d, *J* 9.1), 7.41 – 7.40 (1 H, m), 7.35 – 7.34 (1 H, m), 7.27 – 7.23 (3 H, m, *J* 9.3, 5.6), 7.17 – 7.15 (3 H, m, *J* 7.1), 7.10 – 7.09 (1 H, m), 6.73 – 6.69 (1 H, m), 4.54 (2 H, s). δ_C (100 MHz, $CDCl_3$) 144.79, 138.53, 137.49, 136.37, 129.04, 127.80, 127.00, 125.62, 124.59, 124.55, 123.18, 117.38, 112.44, 29.90.



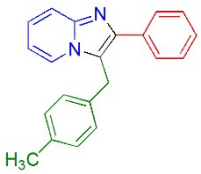
3-Benzyl-7-methyl-2-phenylimidazo[1,2-*a*]pyridine[8] (4i)

White solid, m.p. 145-146°C; δ_H (400 MHz, $CDCl_3$) 7.77 – 7.74 (2 H, m), 7.55 (1 H, d, *J* 6.5), 7.53 – 7.36(4 H, m), 7.34 – 7.20 (4 H, m), 7.11 (2 H, d, *J* 7.0), 6.53 – 6.51 (1 H, m), 4.40 (2 H, s), 2.35 (3 H, s). δ_C (100 MHz, $CDCl_3$) 145.06, 142.98, 136.81, 135.96, 133.93, 129.52, 129.05,128.67, 127.83, 127.70, 126.94, 122.71, 117.17, 115.60, 115.26, 29.69, 21.33.



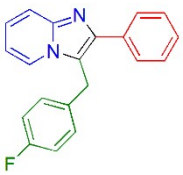
3-Benzyl-7-chloro-2-phenylimidazo[1,2-*a*]pyridine[8] (4j)

Light yellow solid, m.p. 142-143; δ_H (400 MHz, $CDCl_3$) 7.77 (2 H, d, *J* 7.1), 7.65 (1 H, d, *J* 1.6), 7.55 (1 H, d, *J* 7.3), 7.41 (2 H, t, *J* 7.4), 7.34 (1 H, d, *J* 7.1), 7.39 – 7.25 (3 H, m), 7.10 (2 H, d, *J* 6.9), 6.64 (1 H, dd, *J* 7.3, 2.0), 4.43 (2 H, s). δ_C (100 MHz, $CDCl_3$) 148.59, 145.08, 136.07, 133.90, 129.23, 128.08, 127.64, 127.24, 123.58, 118.86, 117.78, 112.84, 29.79.



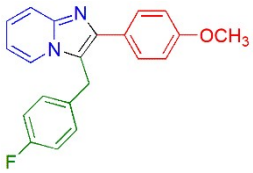
3-(4-Methylbenzyl)-2-phenylimidazo[1,2-*a*]pyridine[9] (4k)

White solid. Mp 125-126 °C. δ_H (400 MHz, $CDCl_3$) 7.80 (2 H, d, *J* 7.2), 7.77 – 7.66 (2 H, m), 7.43 (2 H, t, *J* 7.5), 7.34 (1 H, t, *J* 7.4), 7.19 – 7.14 (1 H, m), 7.11 (2 H, d, *J* 7.8), 7.02 (2 H, d, *J* 7.9), 6.68 (1 H, t, *J* 6.8), 4.44 (2 H, s), 2.32 (3 H, s). δ_C (100 MHz, $CDCl_3$) 144.81, 143.89, 136.55, 134.41, 133.59, 129.75, 128.67, 128.26, 127.77, 127.61, 124.31, 123.49, 117.95, 117.47, 112.27, 29.45, 21.06.



3-(4-Fluorobenzyl)-2-phenylimidazo[1,2-*a*]pyridine[9] (4l)

Yellow oil, δ_H (400 MHz, $CDCl_3$) 7.86 – 7.79 (2 H, m), 7.72 (1 H, d, *J* 6.8), 7.41 (1 H, d, *J* 7.6), 7.34 – 7.29 (1 H, m), 7.18 – 7.12 (1 H, m), 7.10 – 7.05 (2 H, m), 6.98 (2 H, t, *J* 8.6), 6.76 – 6.73 (1 H, m), 4.43 (2 H, s). δ_C (100 MHz, $CDCl_3$) 160.63, 144.73, 143.56, 132.08, 129.10, 128.75, 127.92, 124.92, 123.31, 117.56, 117.41, 115.86, 112.74, 29.01.



3-(4-fluorobenzyl)-2-(4-methoxyphenyl)imidazo[1,2-a]pyridine[11] (4m)

Yellow semi solid, δ_H (400 MHz, $CDCl_3$) 7.79 – 7.66 (4 H, m), 7.32 – 7.25 (1 H, m), 7.08 (2 H, dd, *J* 8.3, 5.5), 6.99 – 6.95 (4 H, m), 6.75 (1 H, t, *J* 6.1), 4.43 (2 H, s), 3.83 (3 H, s). δ_C (100 MHz, $CDCl_3$) 163.05, 160.62, 159.49, 144.68, 143.68, 132.32, 129.42, 129.17, 129.09, 126.52, 124.50, 117.30, 116.82, 116.05, 115.83, 114.19, 112.44, 55.32, 29.09.

^{1}H and ^{13}C NMR spectra of organic products

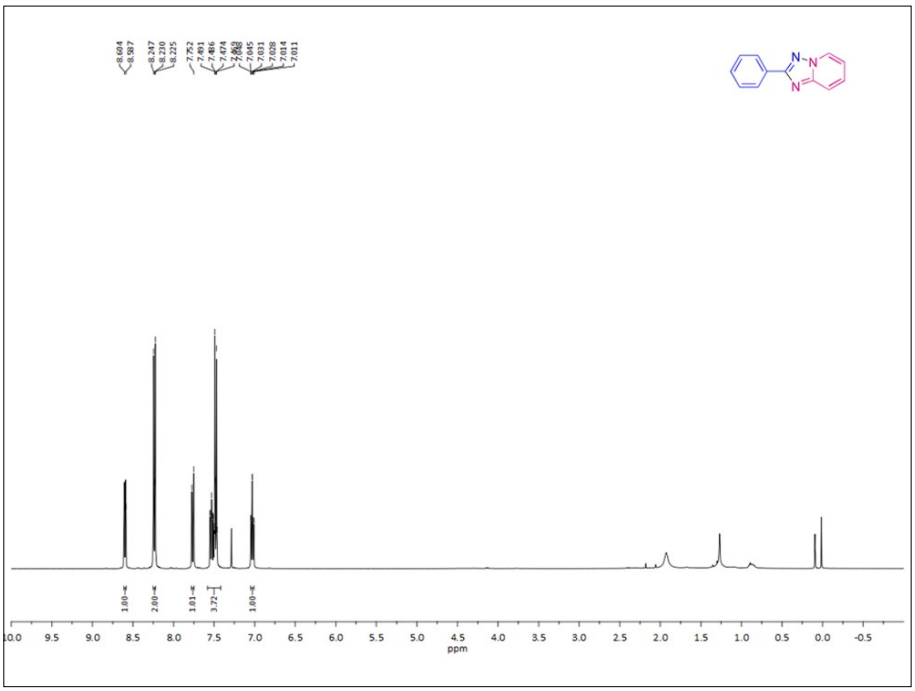


Fig. S6 ^{1}H NMR of 2-Phenyl-[1,2,4]triazolo[1,5-a]pyridine (3a)

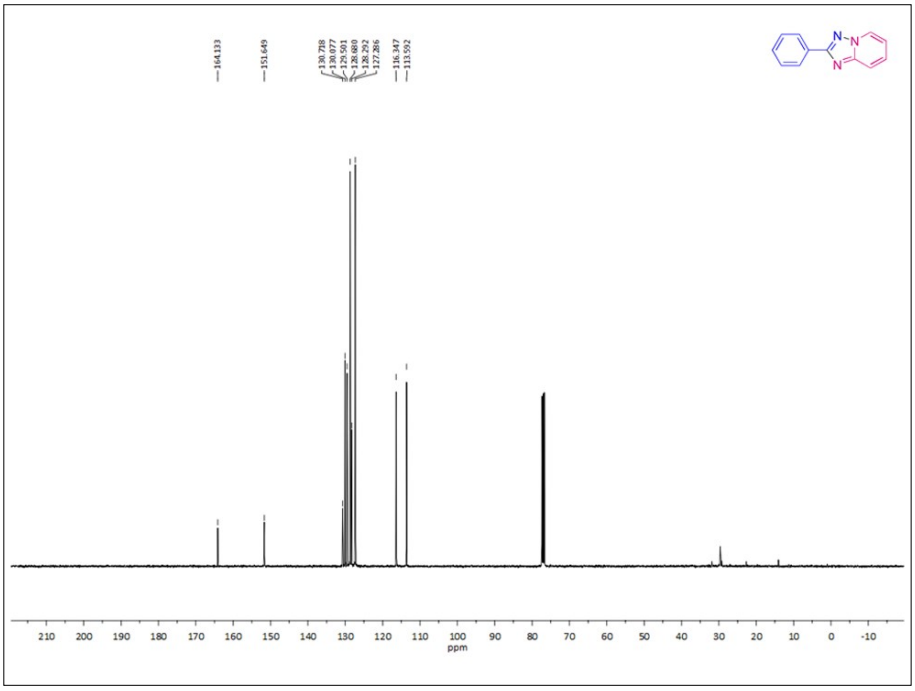


Fig. S7 ^{13}C NMR of 2-Phenyl-[1,2,4]triazolo[1,5-a]pyridine (3a)

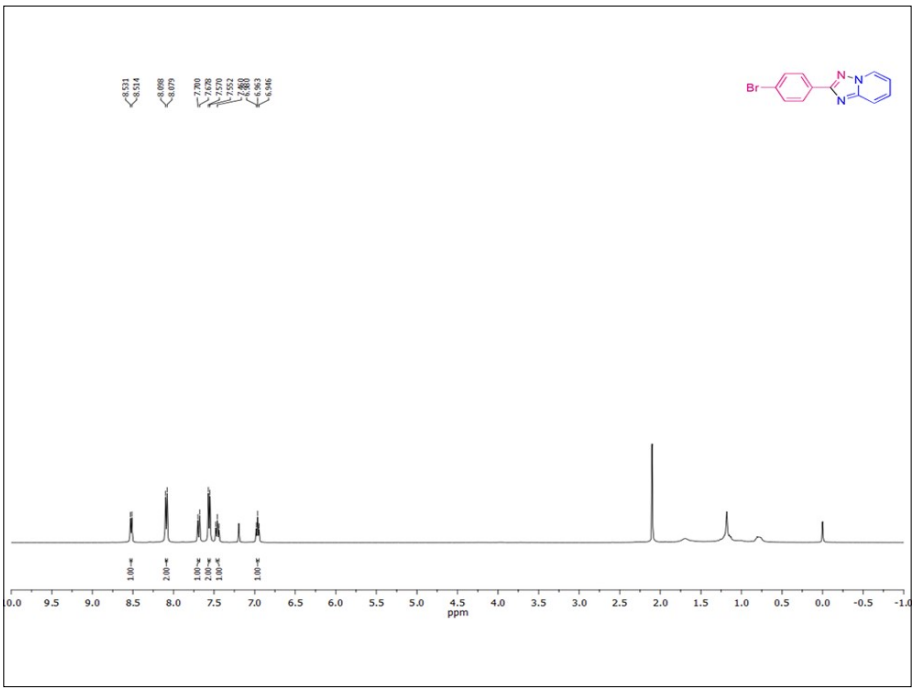


Fig. S8 ^{1}H NMR of 2-(4-Bromophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3b)

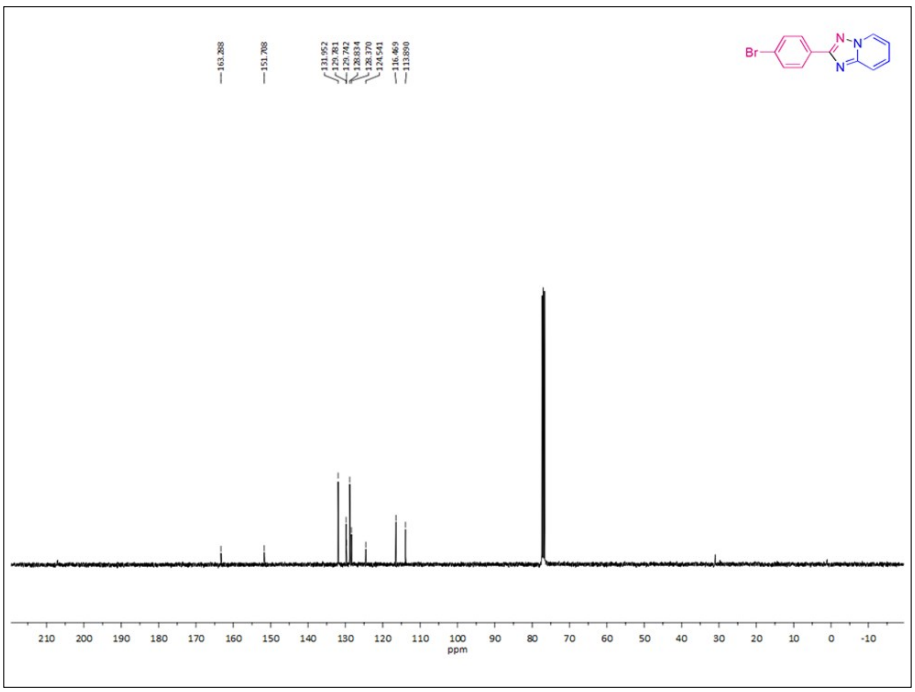


Fig. S9 ^{13}C NMR of 2-(4-Bromophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3b)

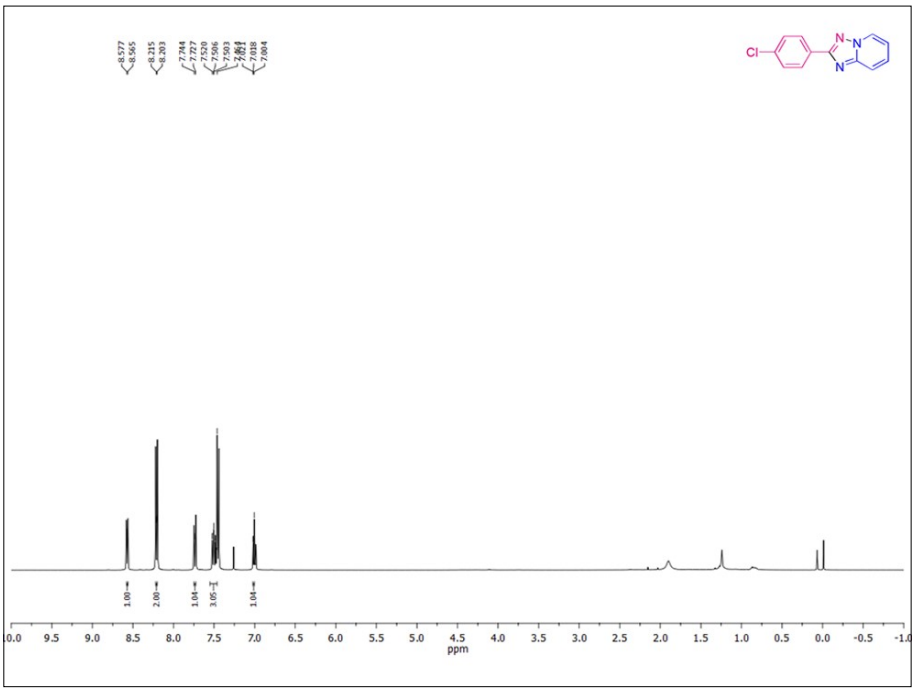


Fig. S10 ^{1}H NMR of 2-(4-Chlorophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3c)

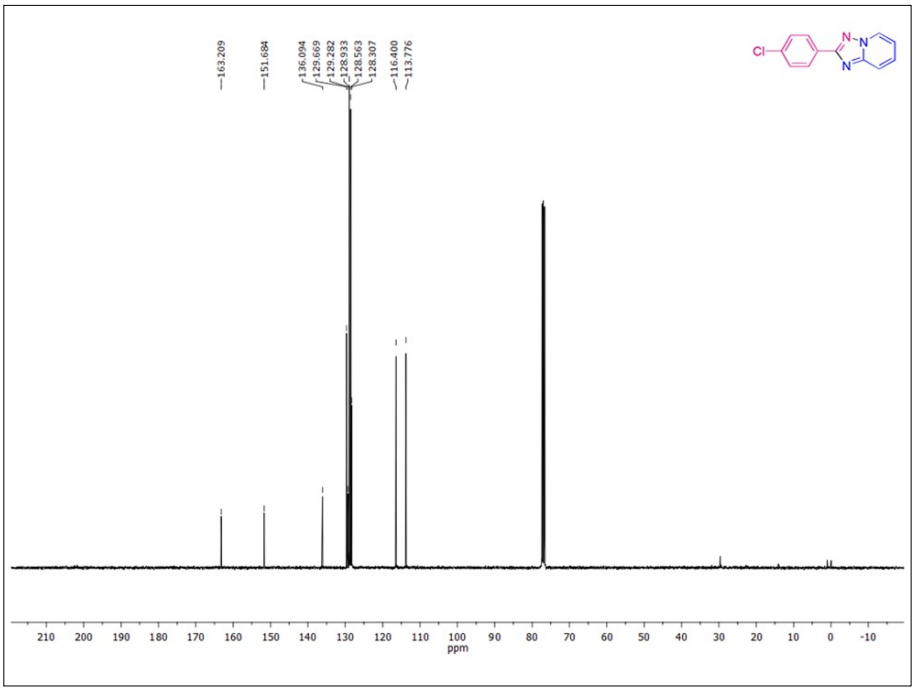


Fig. S11 ^{13}C NMR of 2-(4-Chlorophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3c)

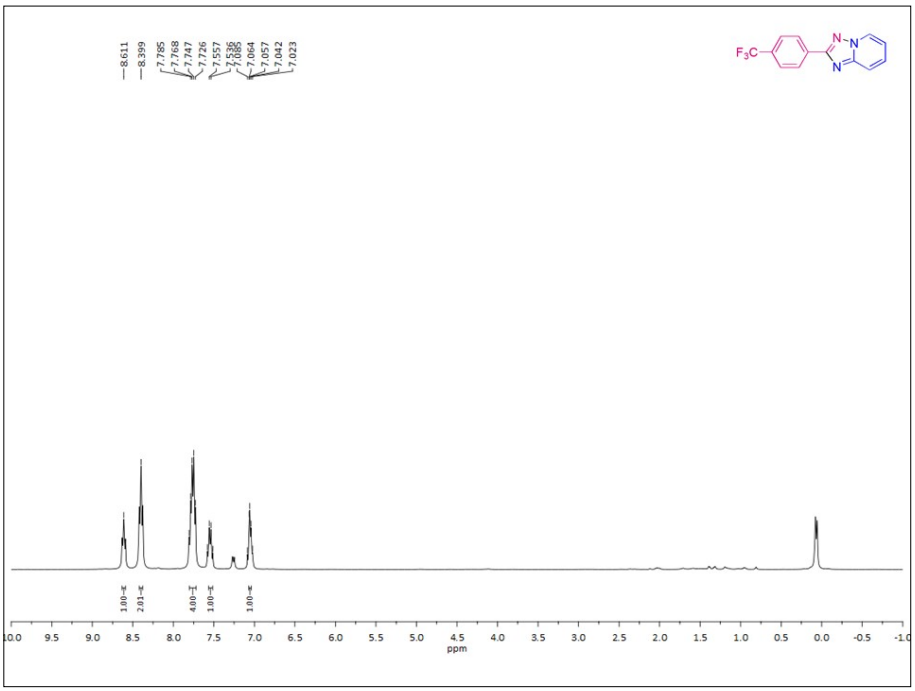


Fig. S12 ^{1}H NMR of 2-(4-(Trifluoromethyl)phenyl)-[1,2,4]triazolo[1,5-a]pyridine (3d)

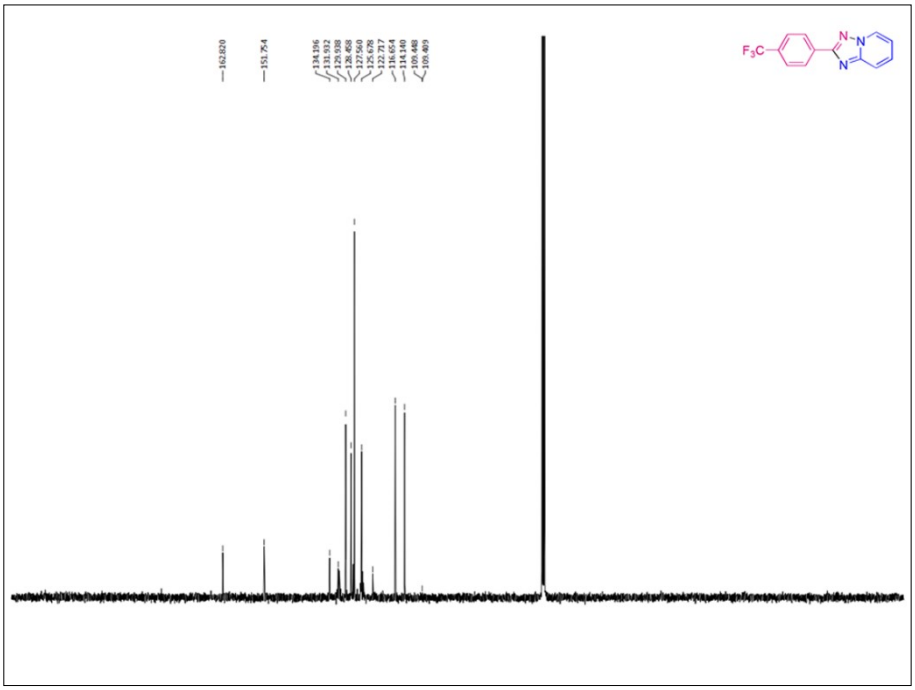


Fig. S13 ^{13}C NMR of 2-(4-(Trifluoromethyl)phenyl)-[1,2,4]triazolo[1,5-a]pyridine (3d)

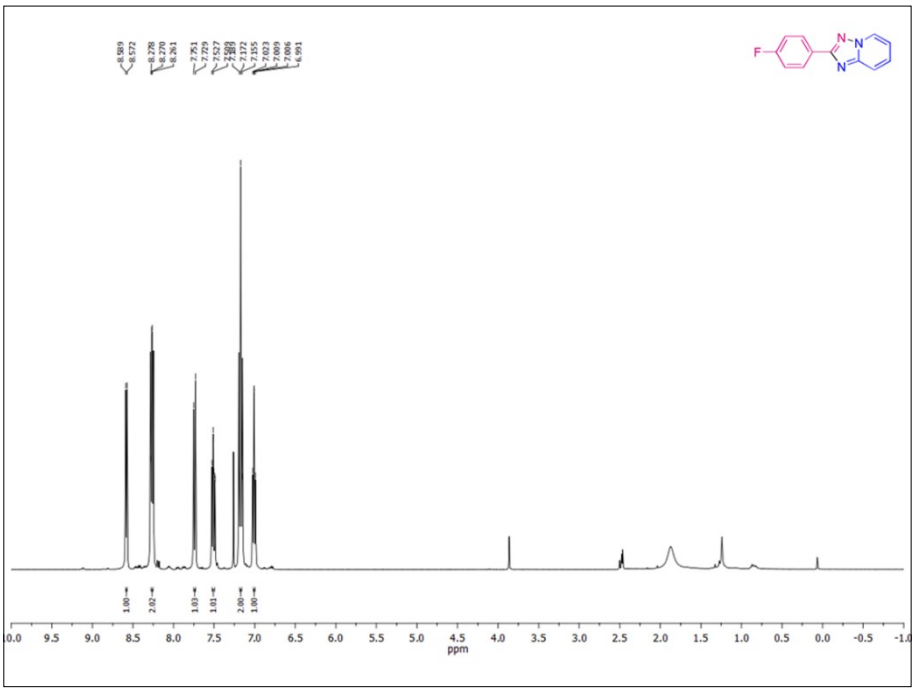


Fig. S14 ^{1}H NMR of 2-(4-Fluorophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3e)

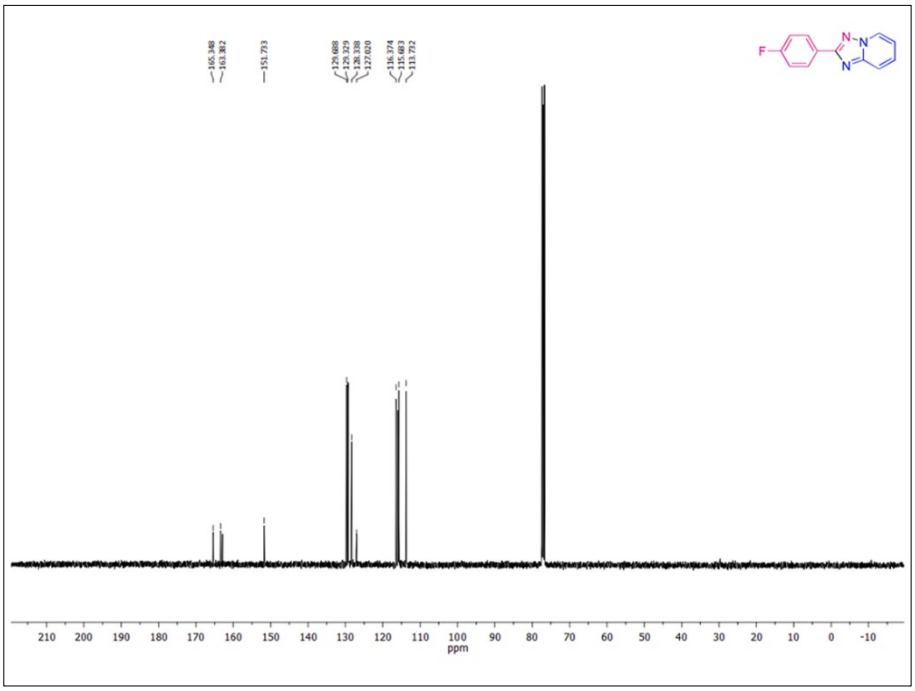


Fig. S15 ^{13}C 2-(4-Fluorophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3e)

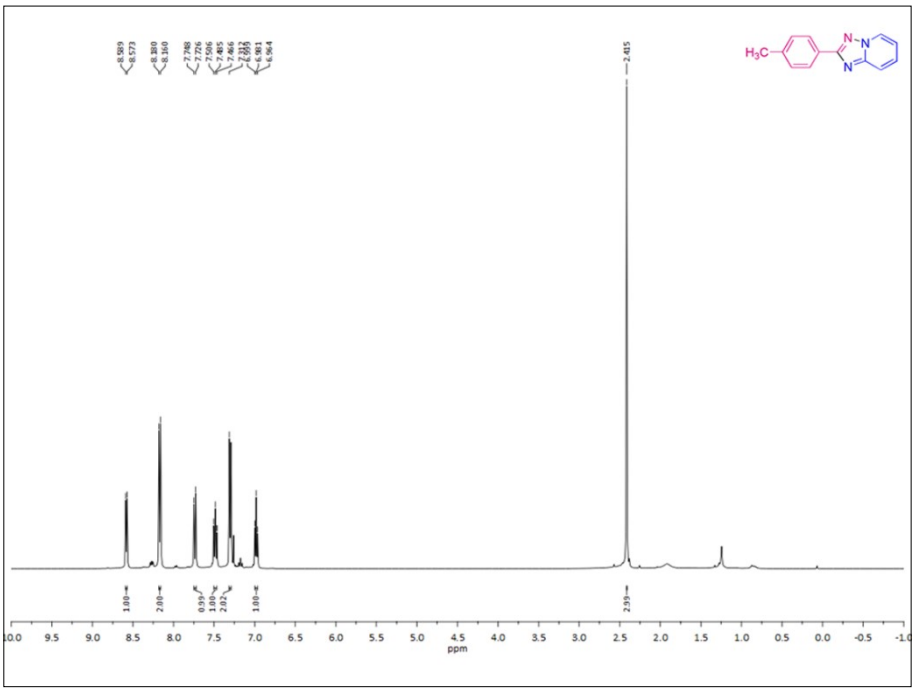


Fig. S16 ^{1}H NMR of 2-*p*-Tolyl-[1,2,4]triazolo[1,5-a]pyridine (3f)

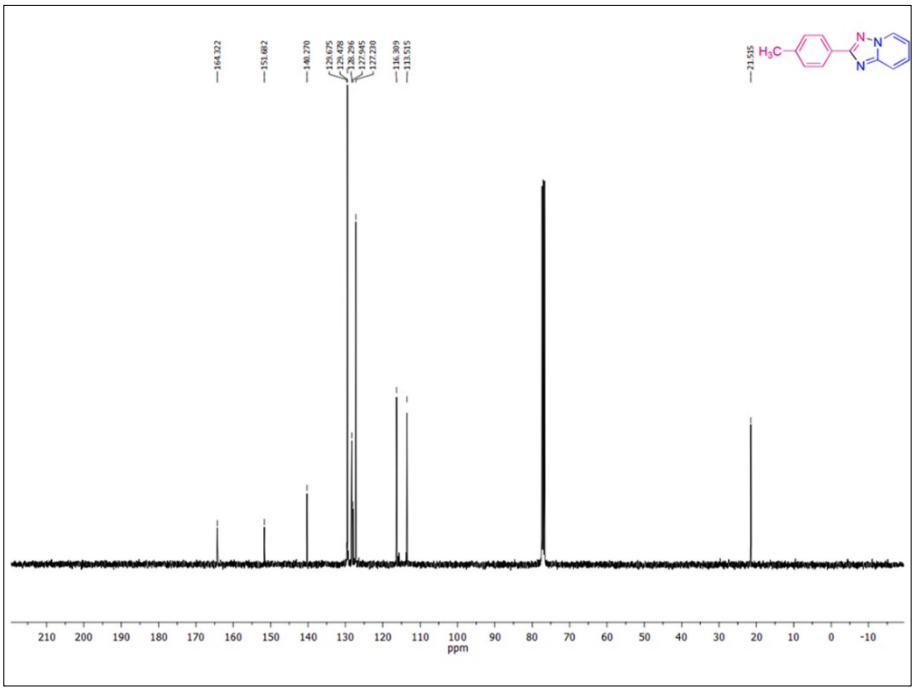


Fig. S17 ^{13}C NMR of 2-*p*-Tolyl-[1,2,4]triazolo[1,5-a]pyridine (3f)

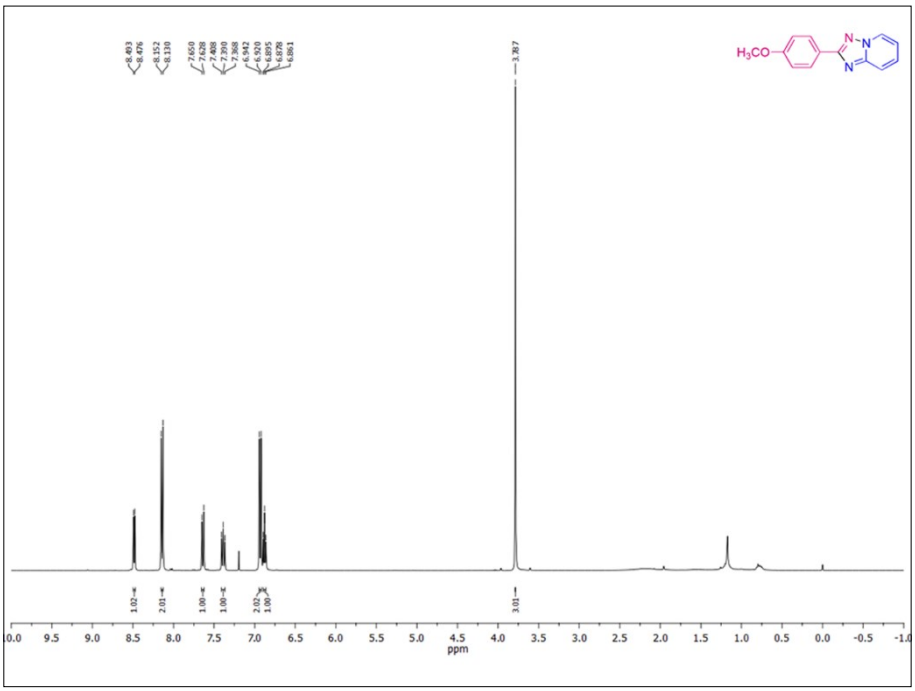


Fig. S18 ^{1}H NMR of 2-(4-Methoxyphenyl)-[1,2,4]triazolo[1,5-a]pyridine (3g)

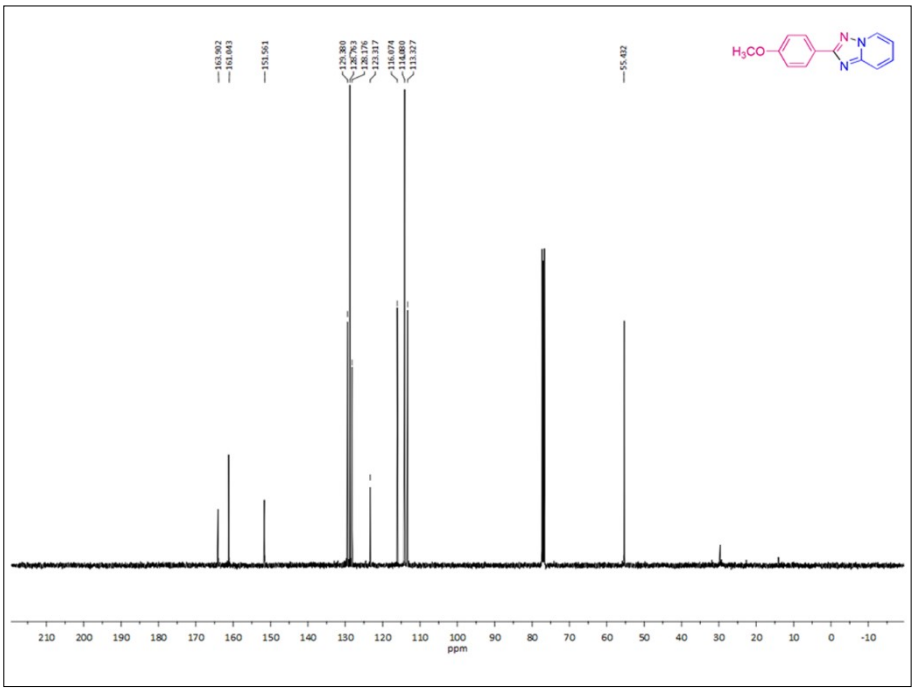


Fig. S19 ^{13}C NMR of 2-(4-Methoxyphenyl)-[1,2,4]triazolo[1,5-a]pyridine (3g)

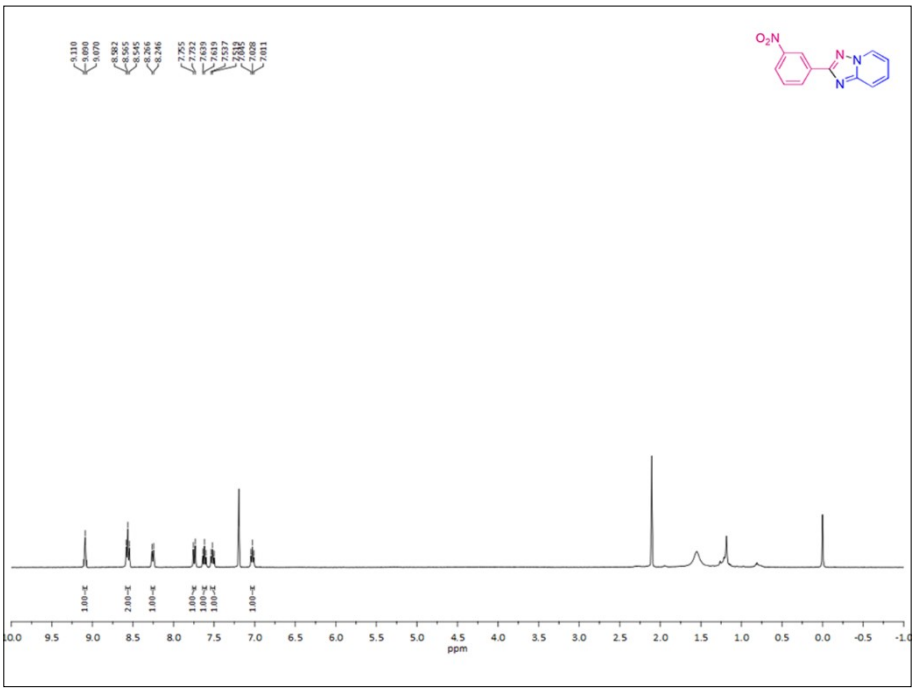


Fig. S20 ^{1}H NMR of 2-(3-Nitrophenyl)[1,2,4]triazolo[1,5-*a*]pyridine (3h)

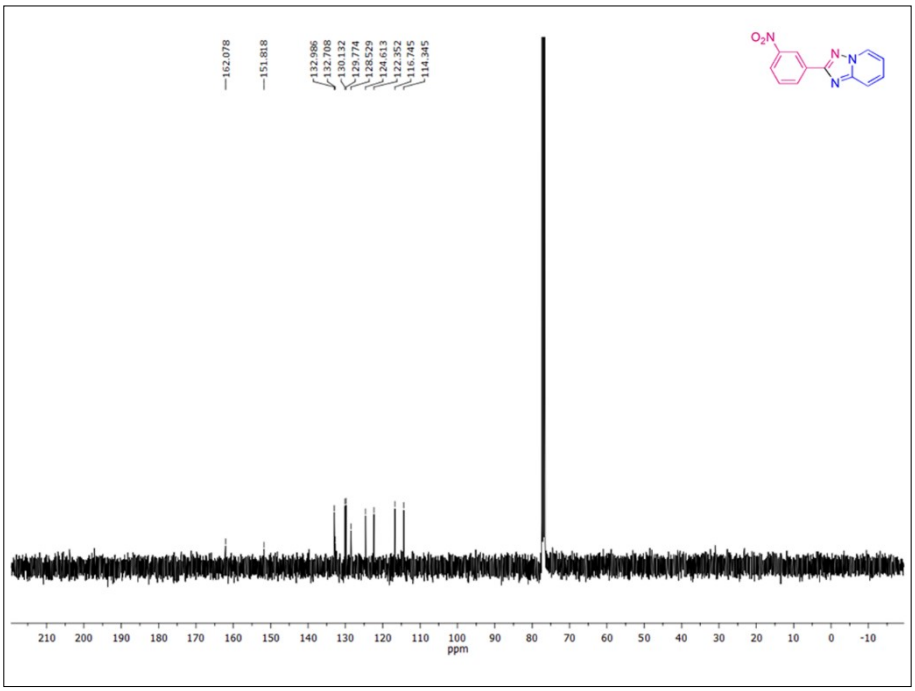


Fig. S21 ^{13}C NMR of 2-(3-Nitrophenyl)[1,2,4]triazolo[1,5-*a*]pyridine (3h)

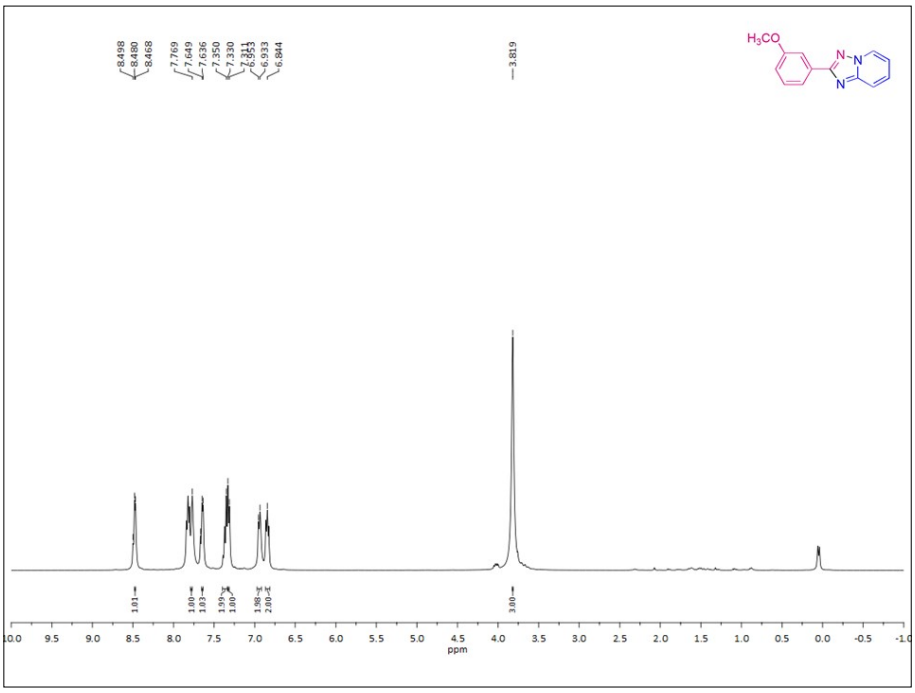


Fig. S22 ^{1}H NMR of 2-(3-Methoxyphenyl)-[1,2,4]triazolo[1,5-a]pyridine (3i)

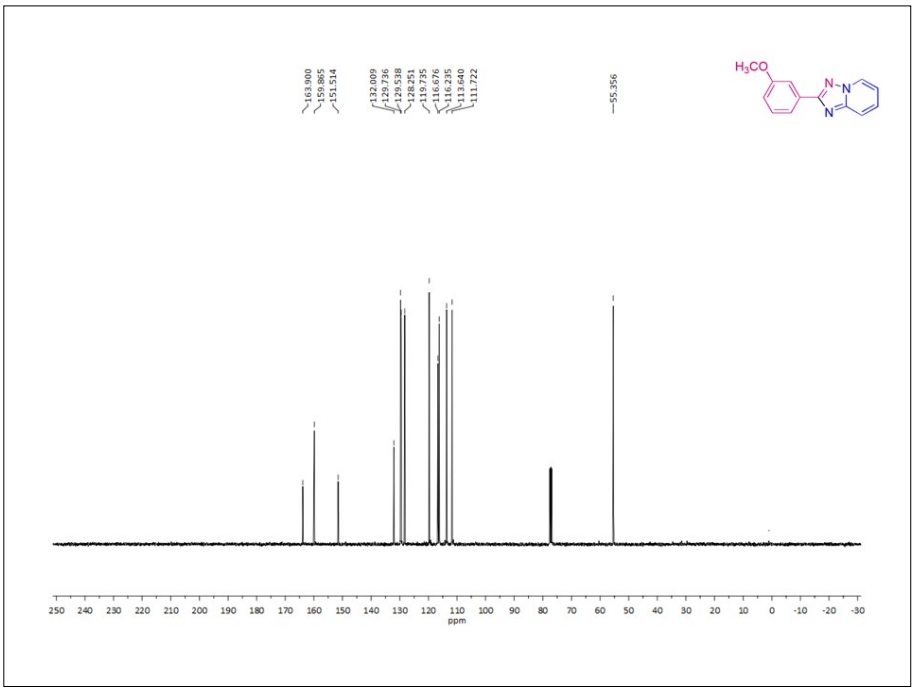


Fig. S23 ^{13}C NMR of 2-(3-Methoxyphenyl)-[1,2,4]triazolo[1,5-a]pyridine (3i)

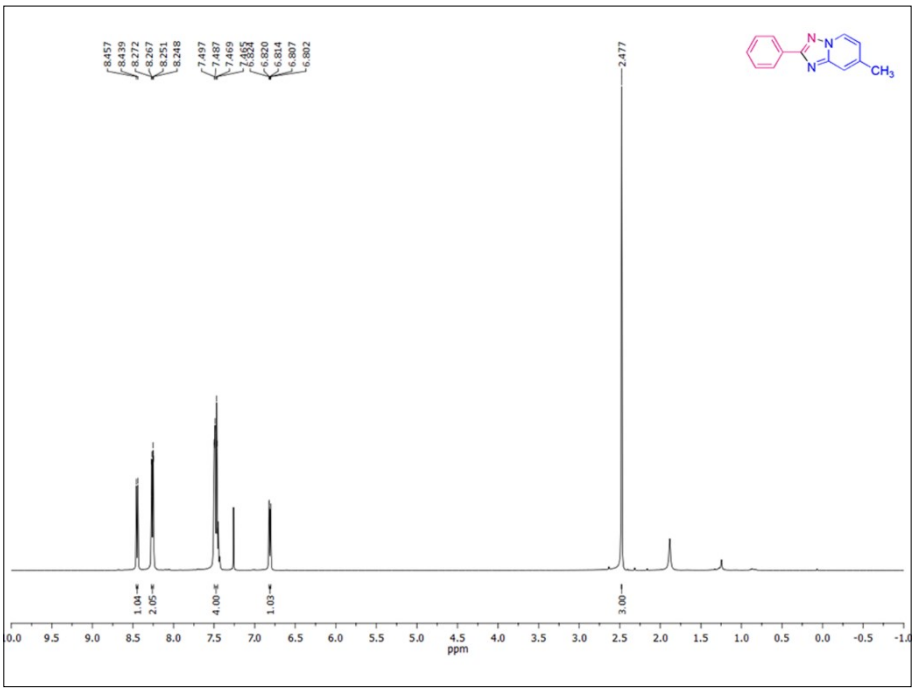


Fig. S24 ^{1}H NMR of 7-Methyl-2-phenyl[1,2,4]triazolo[1,5-*a*]pyridine (3j)

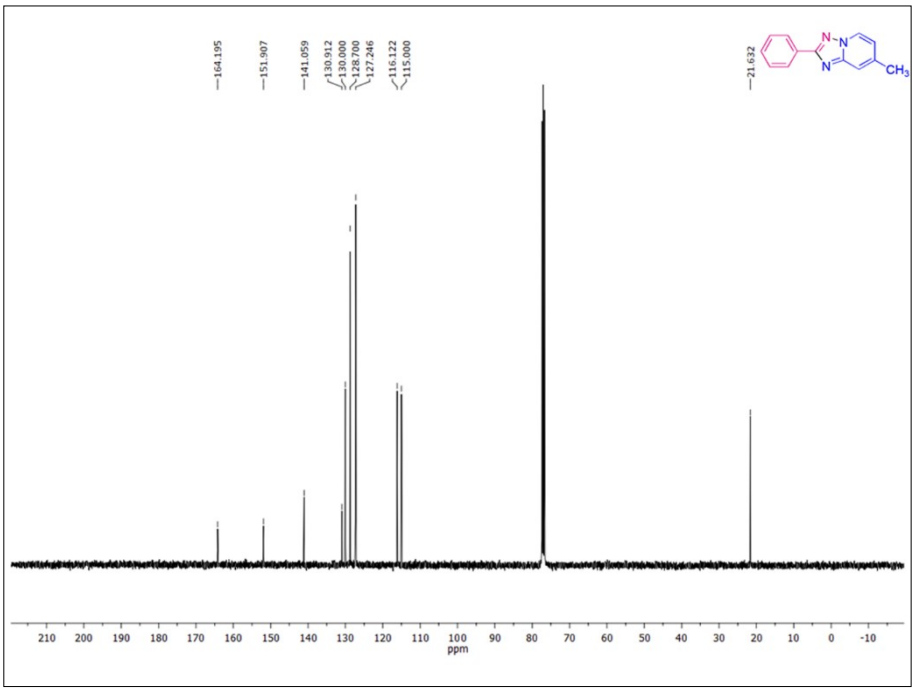


Fig. S25 ^{13}C NMR of 7-Methyl-2-phenyl[1,2,4]triazolo[1,5-*a*]pyridine (3j)

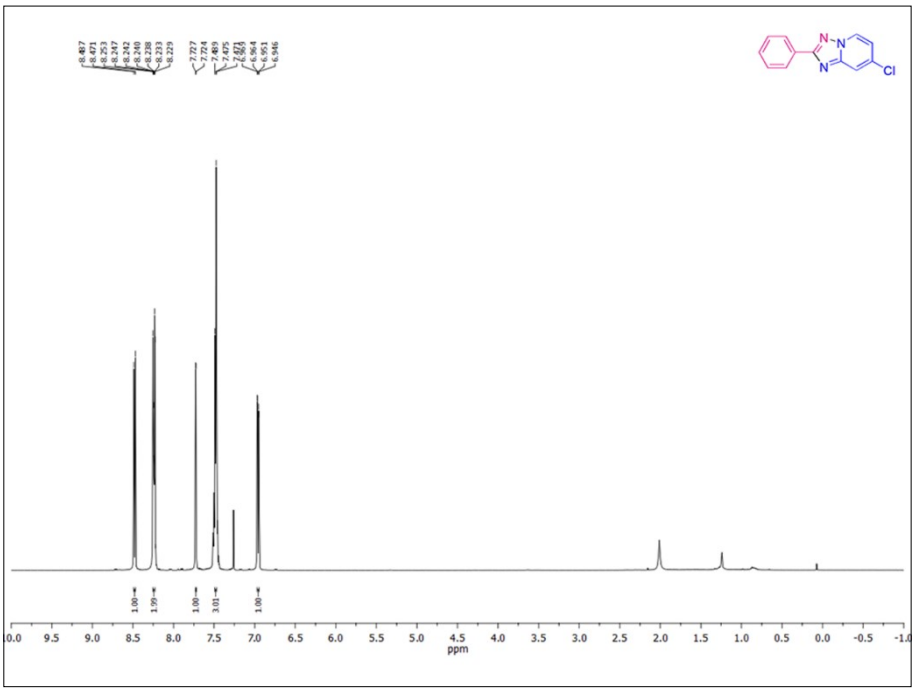


Fig. S26 ^{1}H NMR of 7-Chloro-2-phenyl-[1,2,4]triazolo[1,5-a]pyridine (3k)

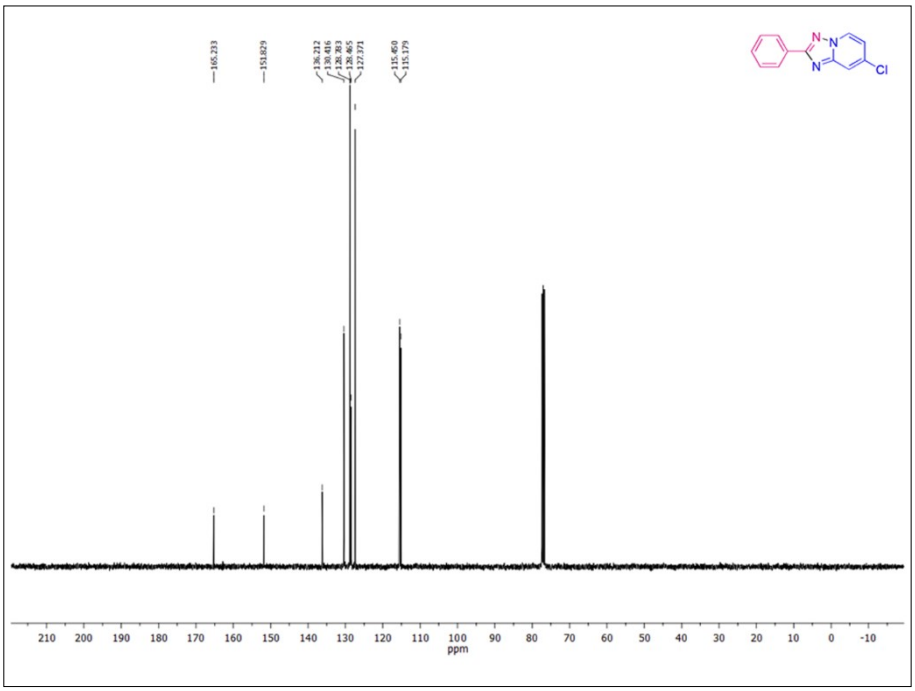


Fig. S27 ^{13}C NMR of 7-Chloro-2-phenyl-[1,2,4]triazolo[1,5-a]pyridine (3k)

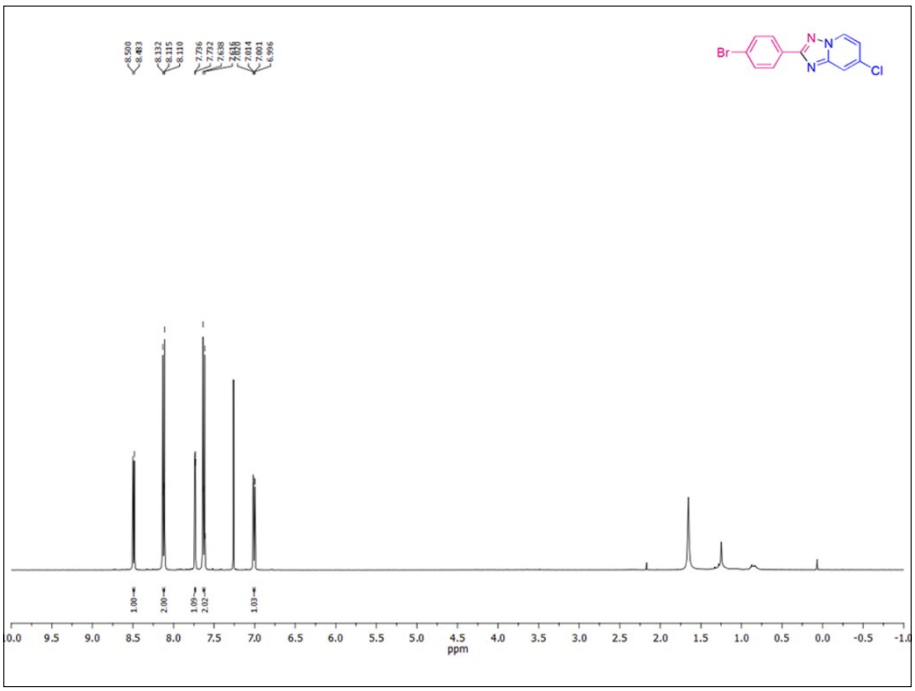


Fig. S28 ^{1}H NMR of 2-(4-Bromophenyl)-7-chloro-[1,2,4]triazolo[1,5-a]pyridine (3l)

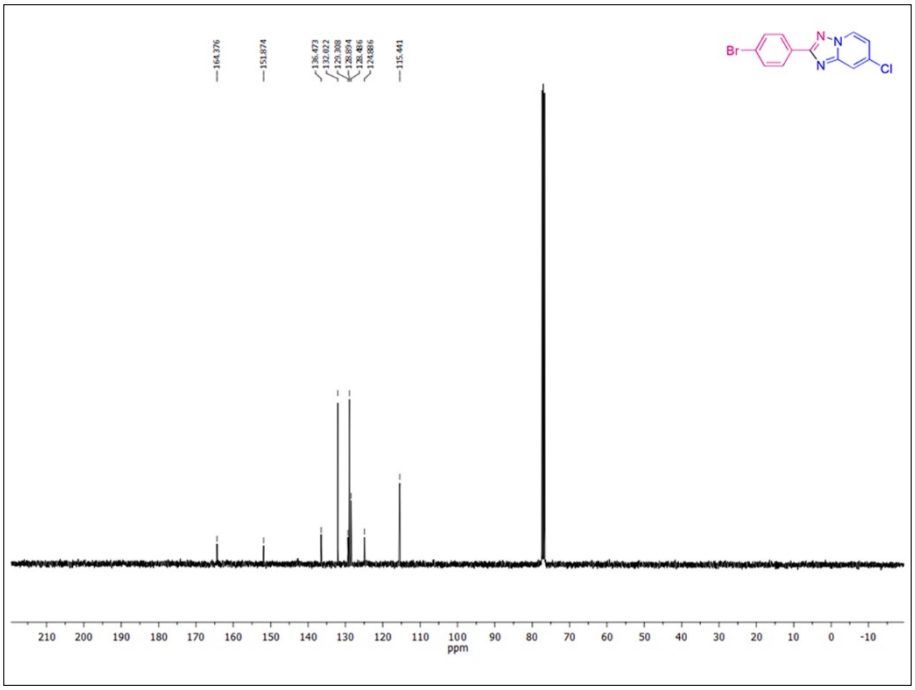


Fig. S29 ^{13}C NMR of 2-(4-Bromophenyl)-7-chloro-[1,2,4]triazolo[1,5-a]pyridine (3l)

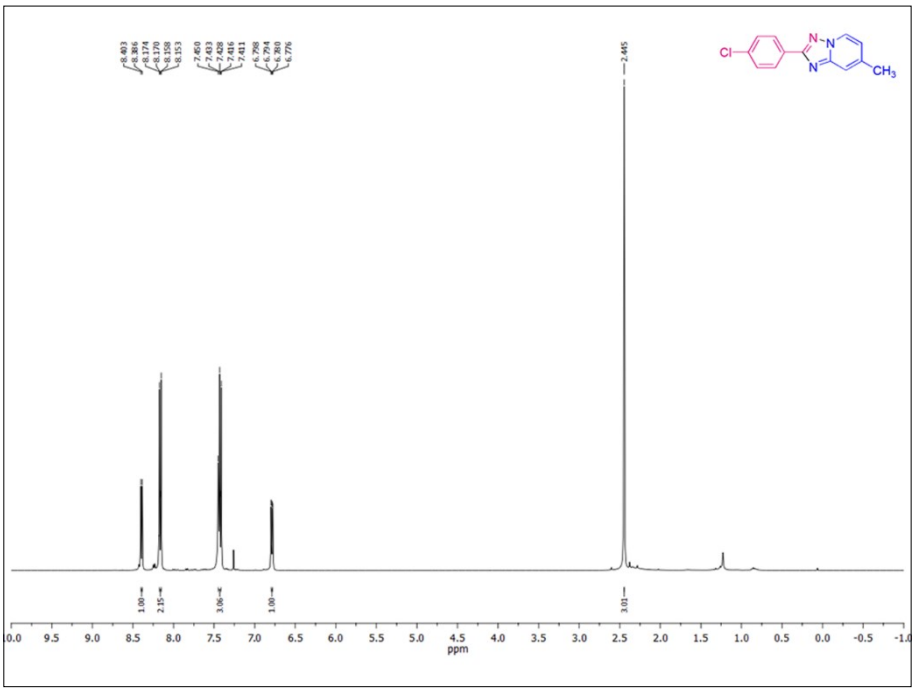


Fig. S30 ^{1}H NMR of 2-(4-Chlorophenyl)-7-methyl[1,2,4]triazolo[1,5-*a*]pyridine (3m)

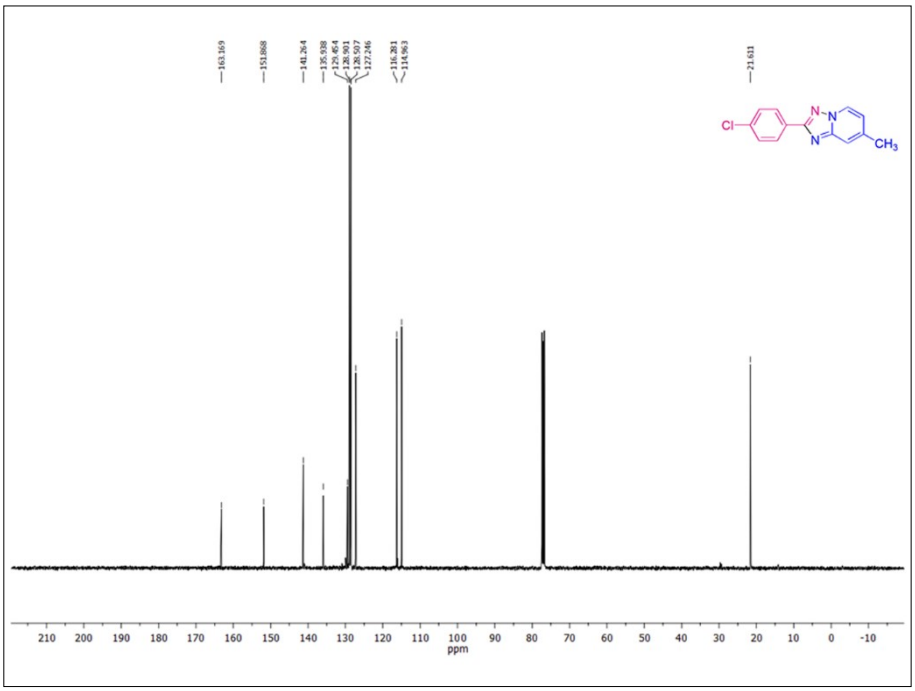


Fig. S31 ^{13}C NMR of 2-(4-Chlorophenyl)-7-methyl[1,2,4]triazolo[1,5-*a*]pyridine (3m)

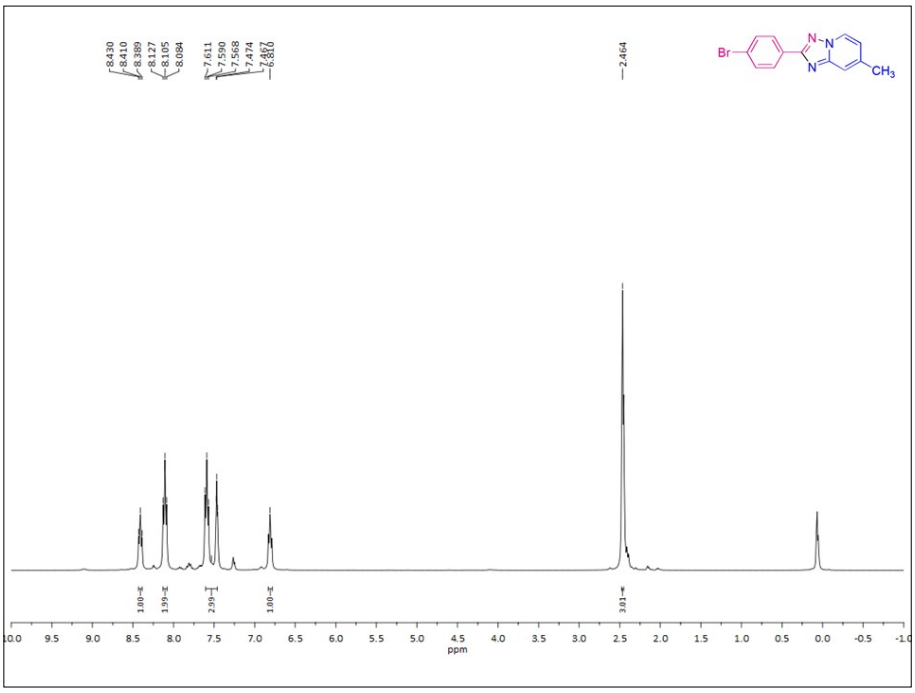


Fig. S32 ^{1}H NMR of 2-(4-Bromophenyl)-7-methyl[1,2,4]triazolo[1,5-*a*]pyridine (3n)

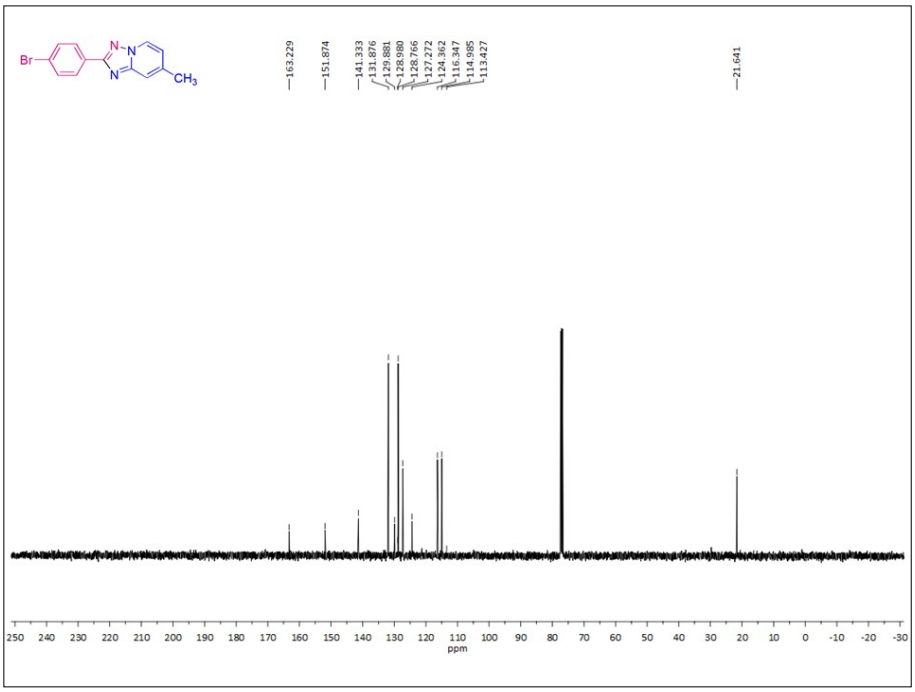


Fig. S33 ^{13}C NMR of 2-(4-Bromophenyl)-7-methyl[1,2,4]triazolo[1,5-*a*]pyridine (3n)

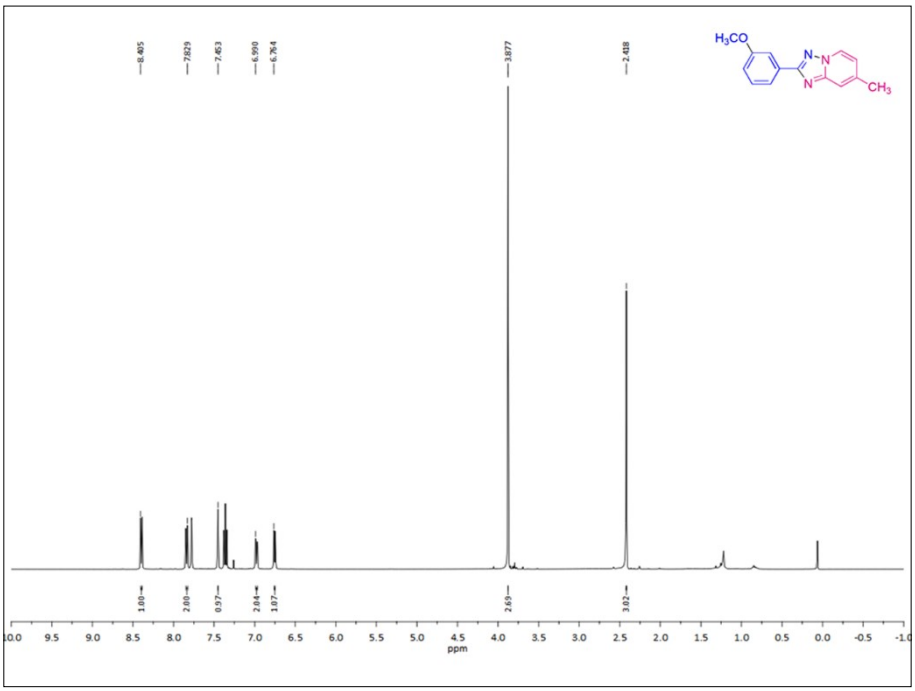


Fig. S34 ^{1}H NMR of 2-(3-Methoxyphenyl)-7-methyl[1,2,4]triazolo[1,5-a]pyridine (3o)

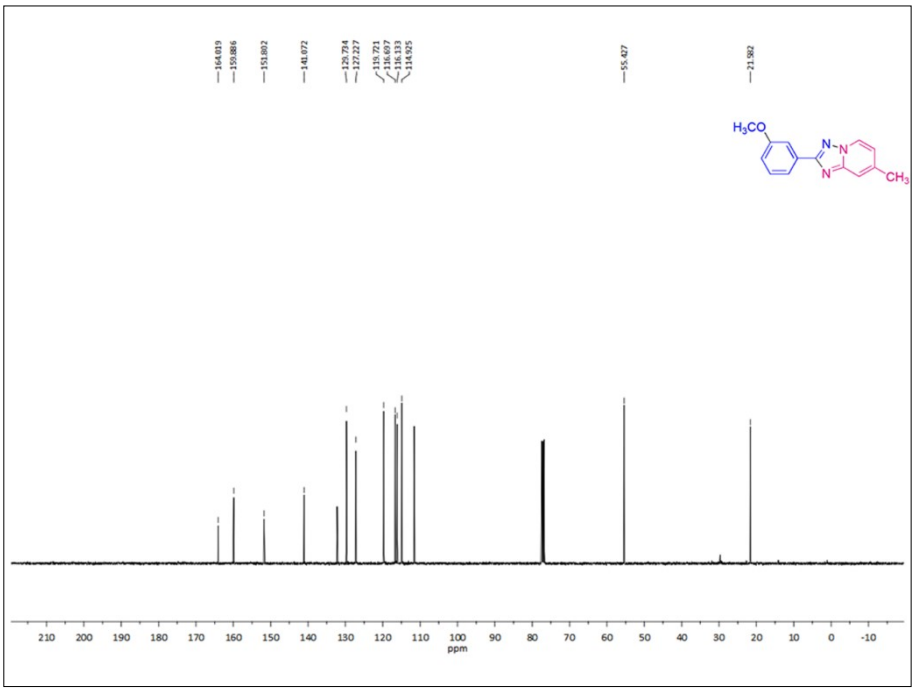


Fig. S35 ^{13}C NMR of 2-(3-Methoxyphenyl)-7-methyl[1,2,4]triazolo[1,5-a]pyridine (3o)

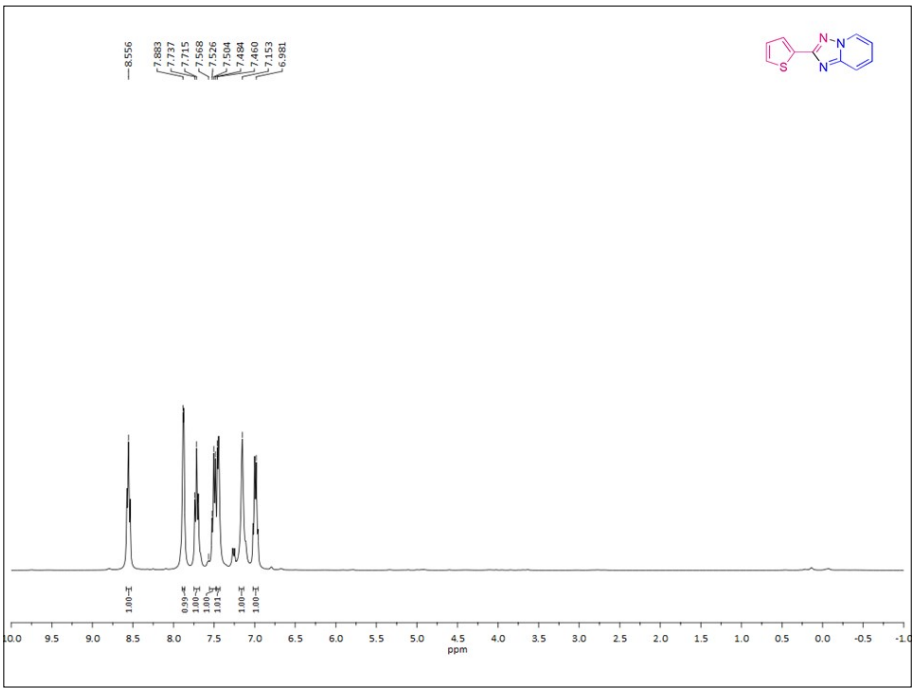


Fig. S36 ^{1}H NMR of 2-(2-Thiophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3p)

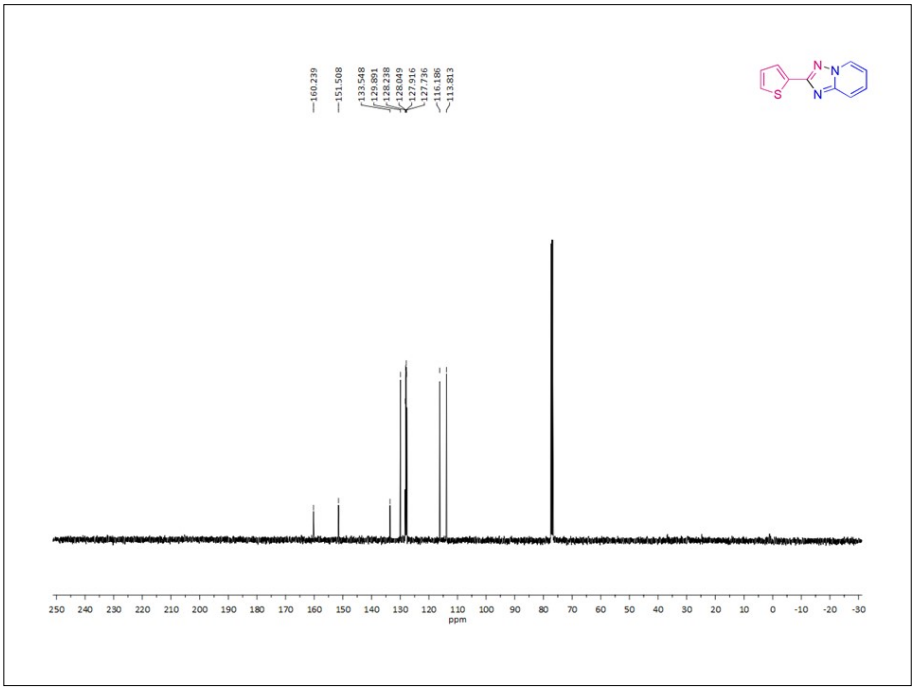


Fig. S37 ^{13}C NMR of 2-(2-Thiophenyl)-[1,2,4]triazolo[1,5-a]pyridine (3p)

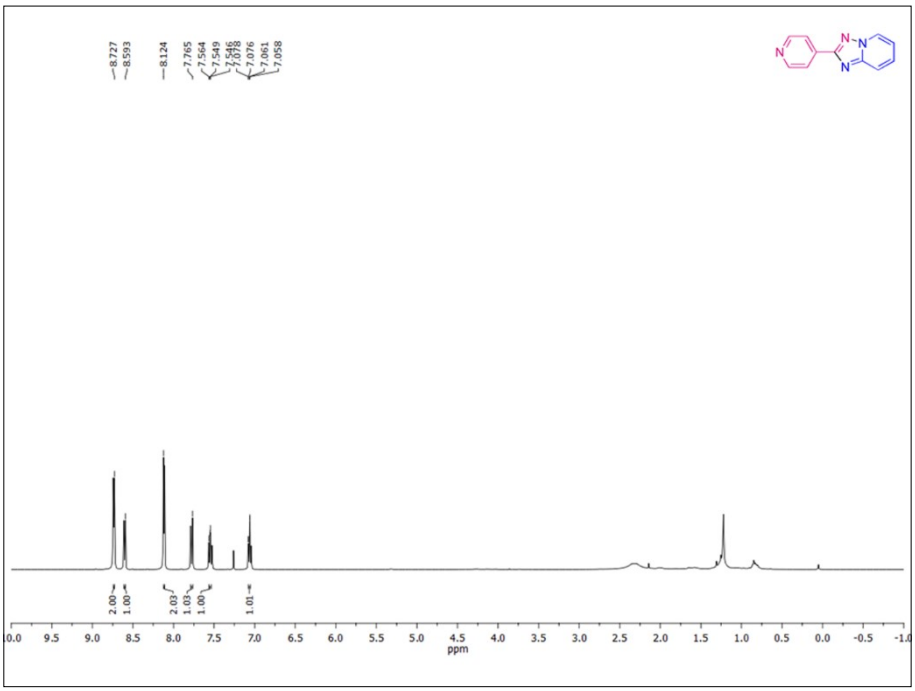


Fig. S38 ^{1}H NMR of 2-(Pyridin-4-yl)-[1,2,4]triazolo[1,5-a]pyridine (3q)

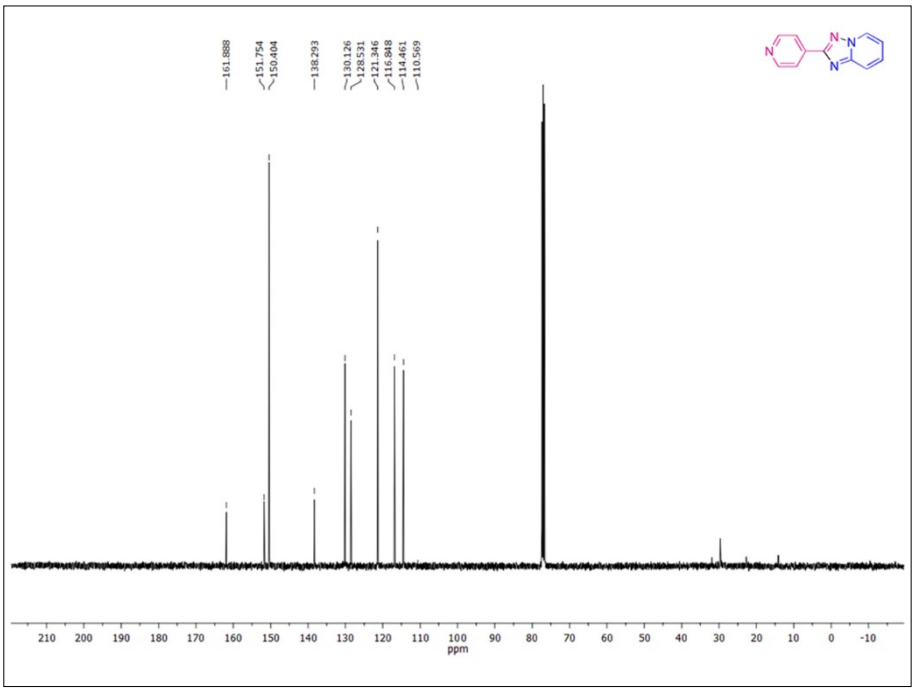


Fig. S39 ^{13}C NMR of 2-(Pyridin-4-yl)-[1,2,4]triazolo[1,5-a]pyridine (3q)

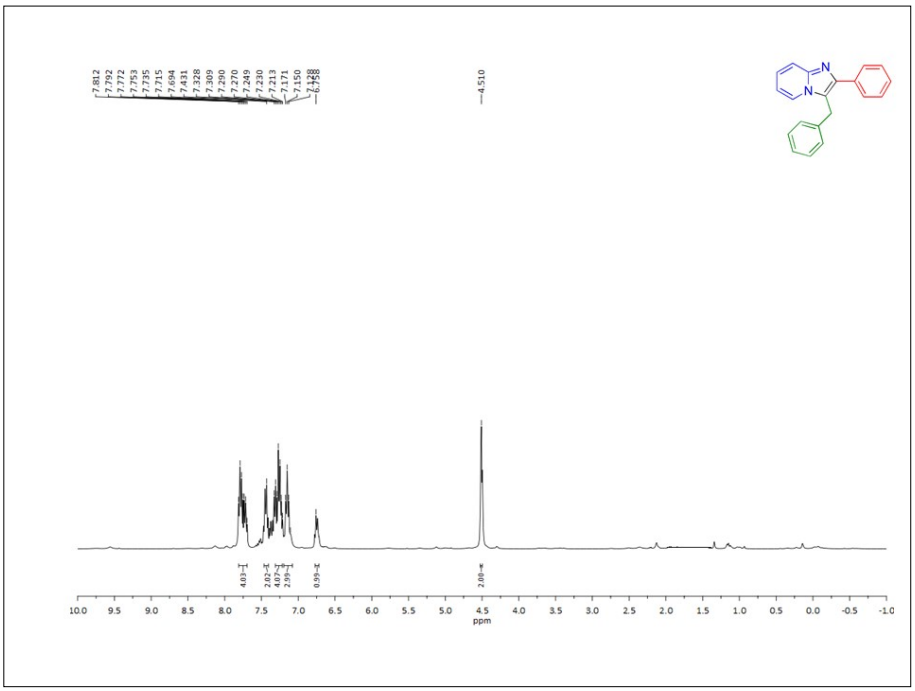


Fig. S40 ^{1}H NMR of 3-Benzyl-2-phenylimidazo[1,2-*a*]pyridine (4a)

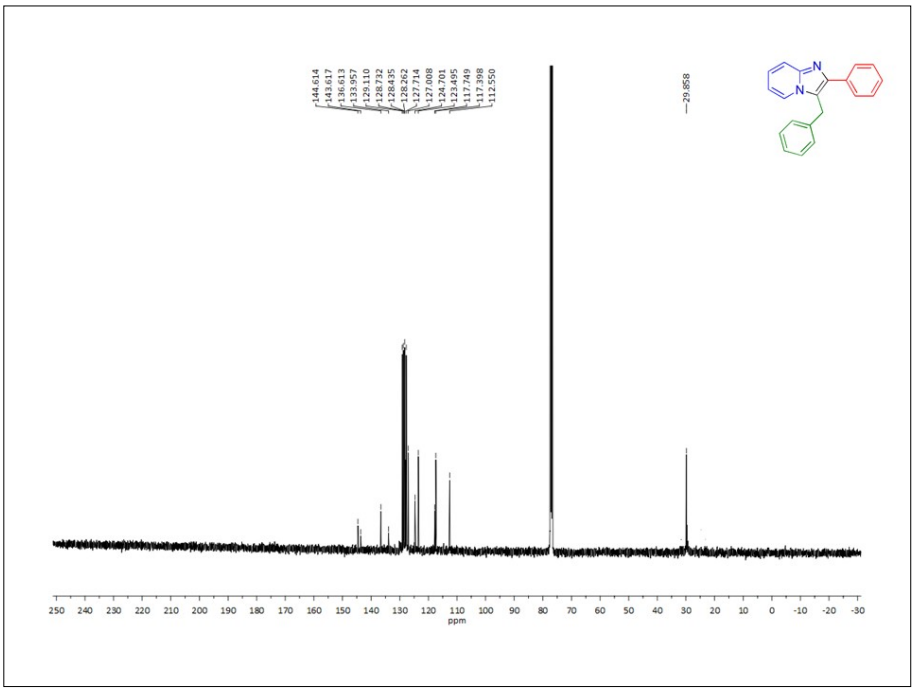


Fig. S41 ^{13}C NMR of 3-Benzyl-2-phenylimidazo[1,2-*a*]pyridine (4a)

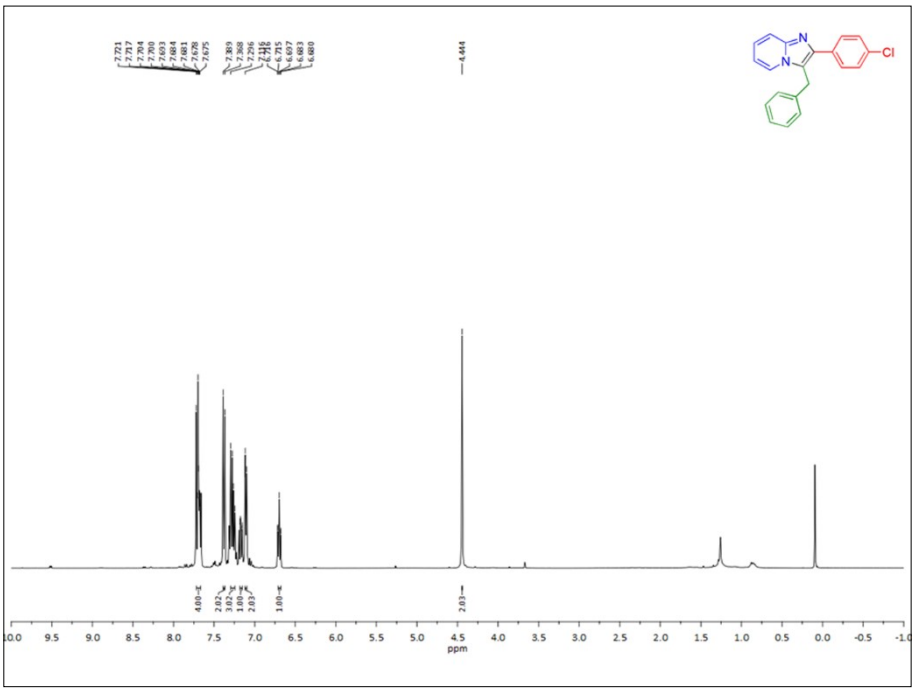


Fig. S42 ^{1}H NMR of 3-benzyl-2-(4-chlorophenyl)H-imidazo[1,2-a]pyridine (4b)

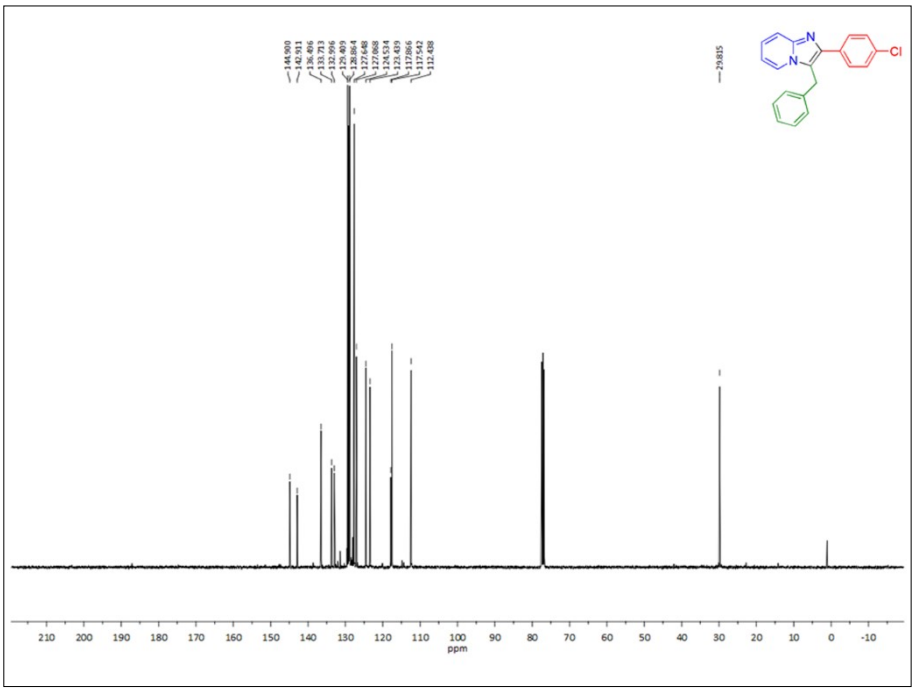


Fig. S43 ^{13}C NMR of 3-benzyl-2-(4-chlorophenyl)H-imidazo[1,2-a]pyridine (4b)

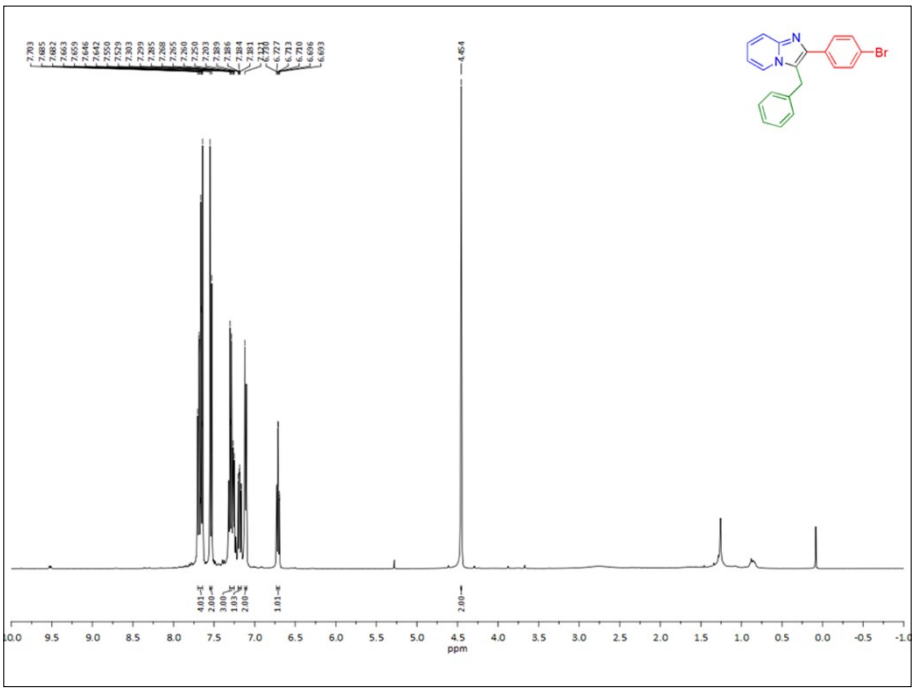


Fig. S44 ^{1}H NMR of 3-benzyl-2-(4bromophenyl)H-imidazo[1,2-a]pyridine (4c)

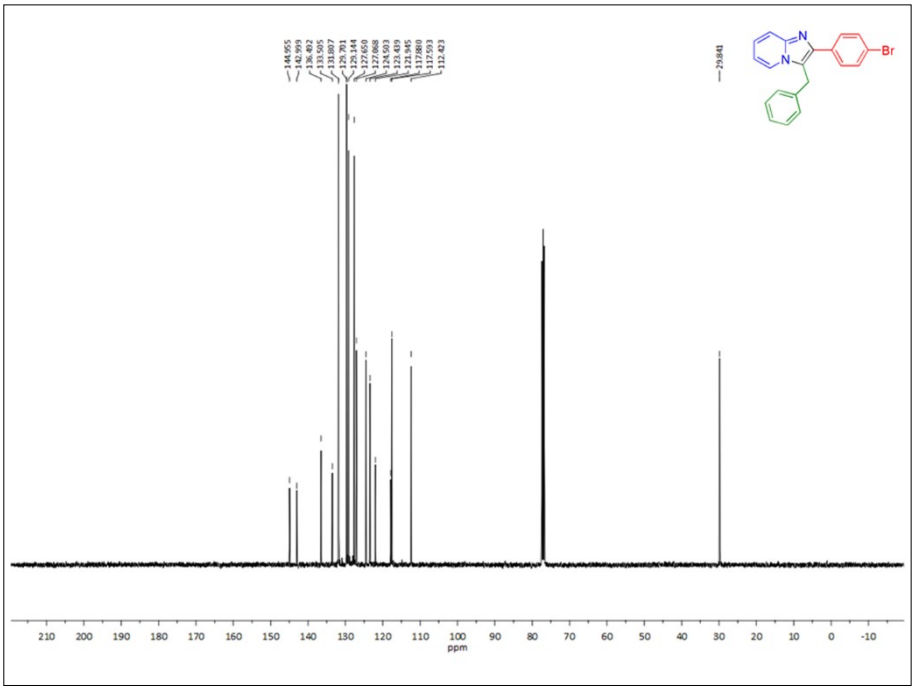


Fig. S45 ^{13}C NMR of 3-benzyl-2-(4bromophenyl)H-imidazo[1,2-a]pyridine (4c)

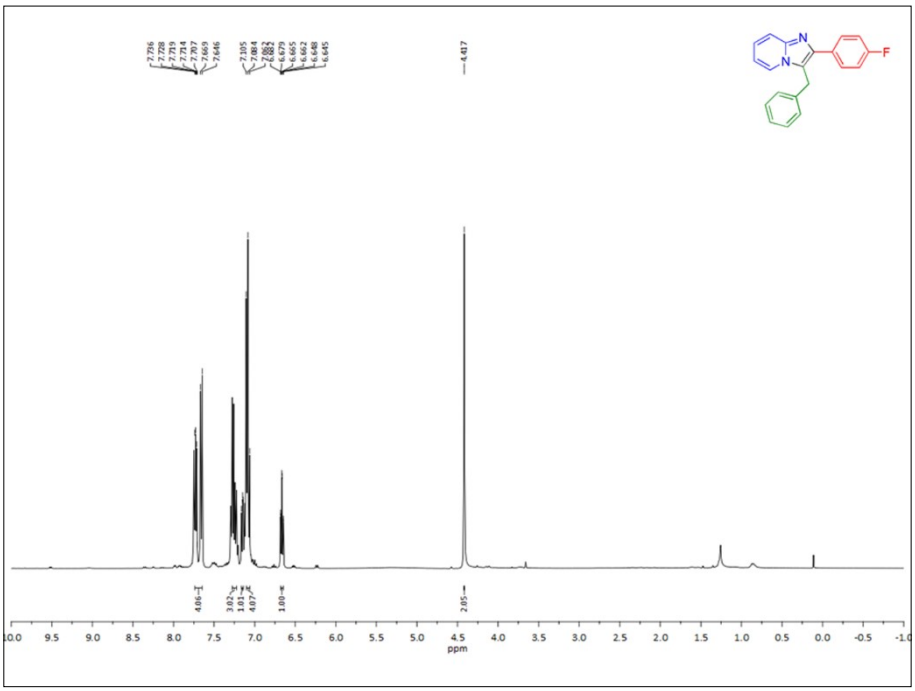


Fig. S46 ^{1}H NMR of 3-Benzyl-2-(4-fluorophenyl)imidazo[1,2-*a*]pyridine (4d)

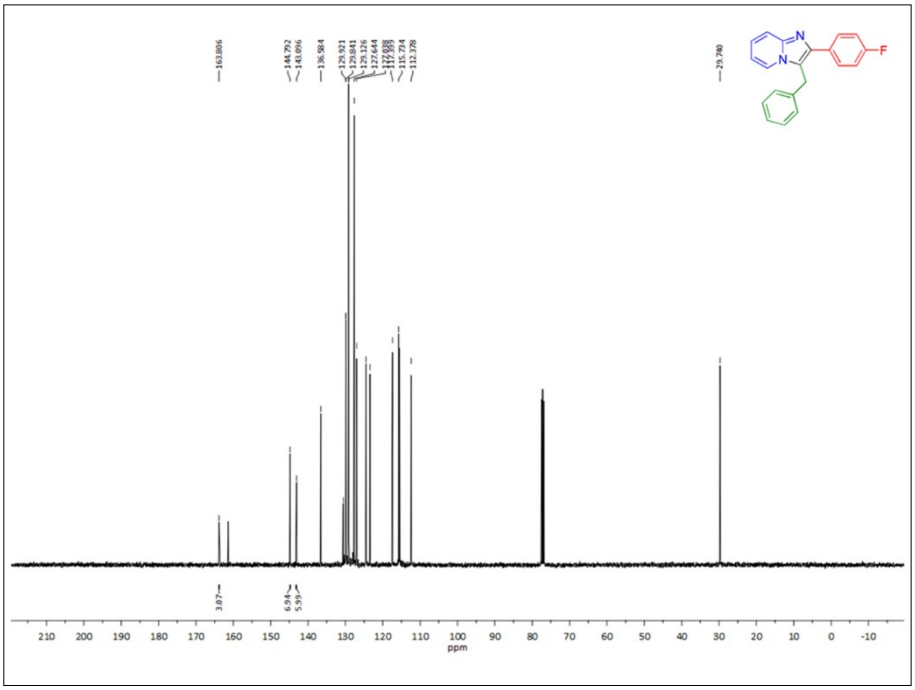


Fig. S47 ^{13}C NMR of 3-Benzyl-2-(4-fluorophenyl)imidazo[1,2-*a*]pyridine (4d)

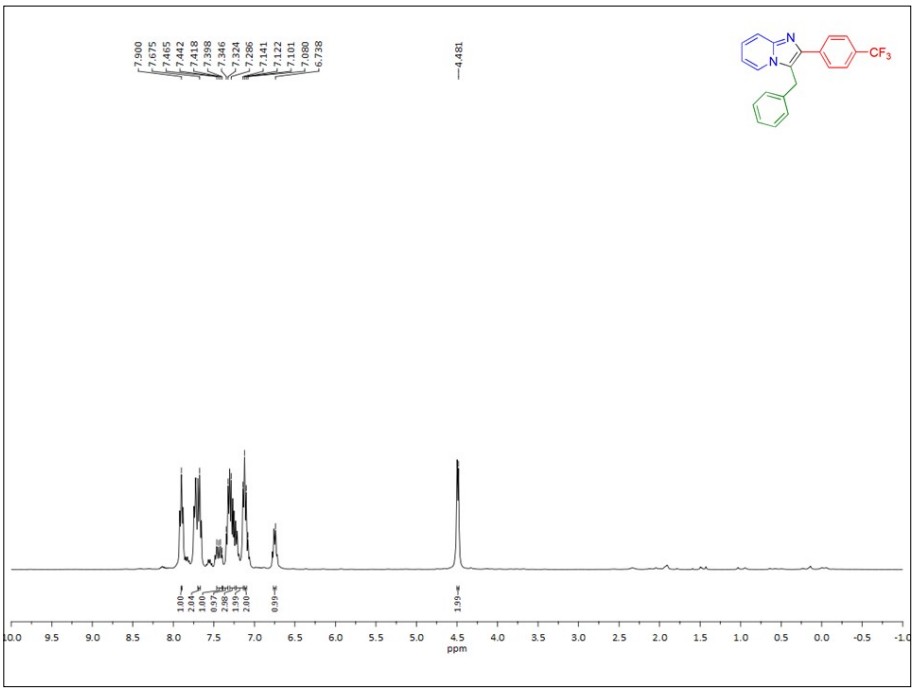


Fig. S48 ^{1}H NMR of 3-benzyl-2-(4-(trifluoromethyl)phenyl)imidazo[1,2-a]pyridine (4e)

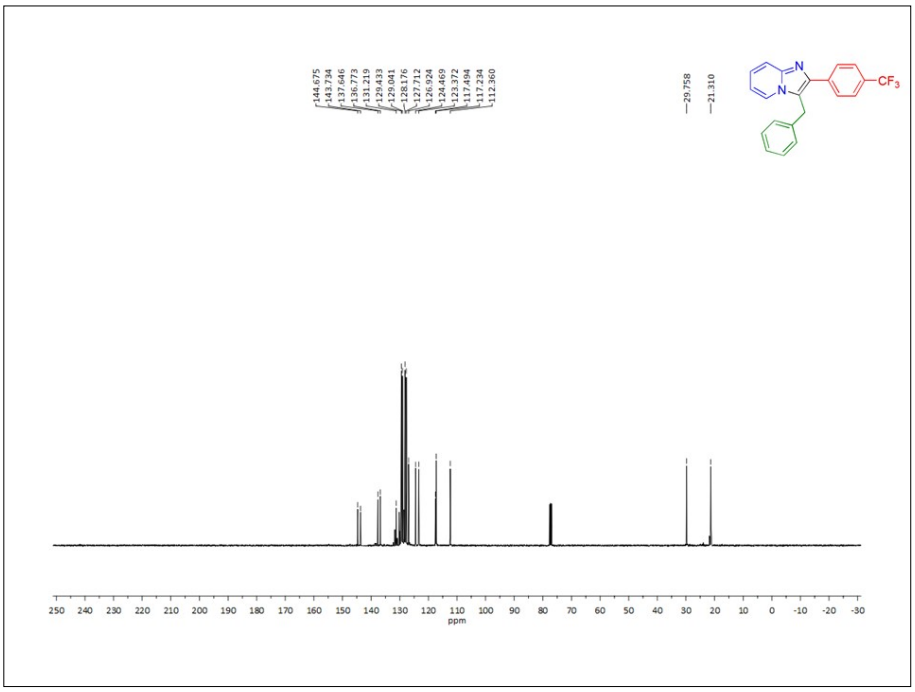


Fig. S49 ^{13}C NMR of 3-benzyl-2-(4-(trifluoromethyl)phenyl)imidazo[1,2-a]pyridine (4e)

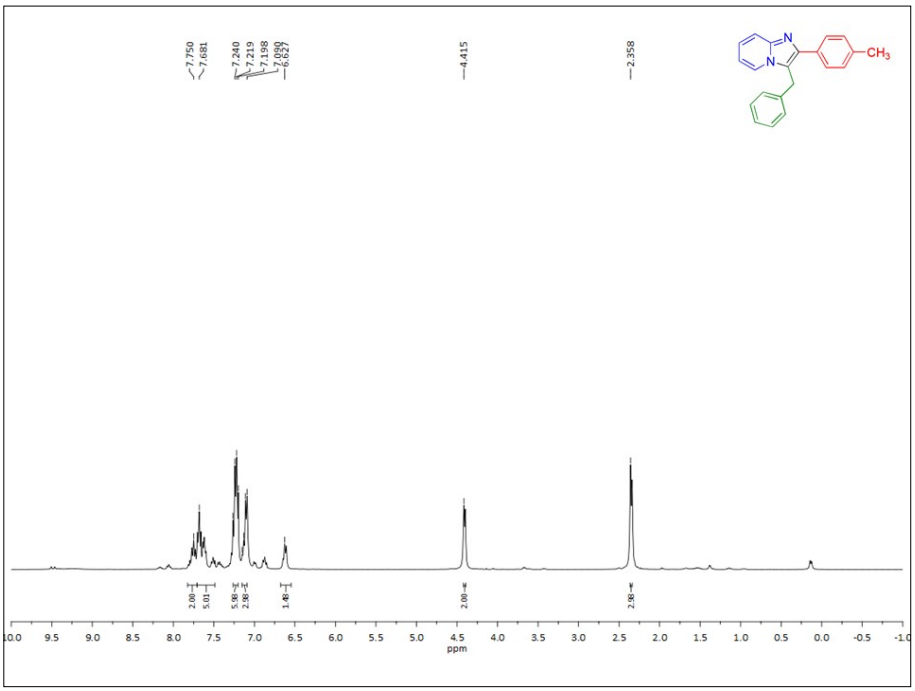


Fig. S50 ^{1}H NMR of 3-Benzyl-2-*p*-tolylimidazo[1,2-*a*]pyridine (4f)

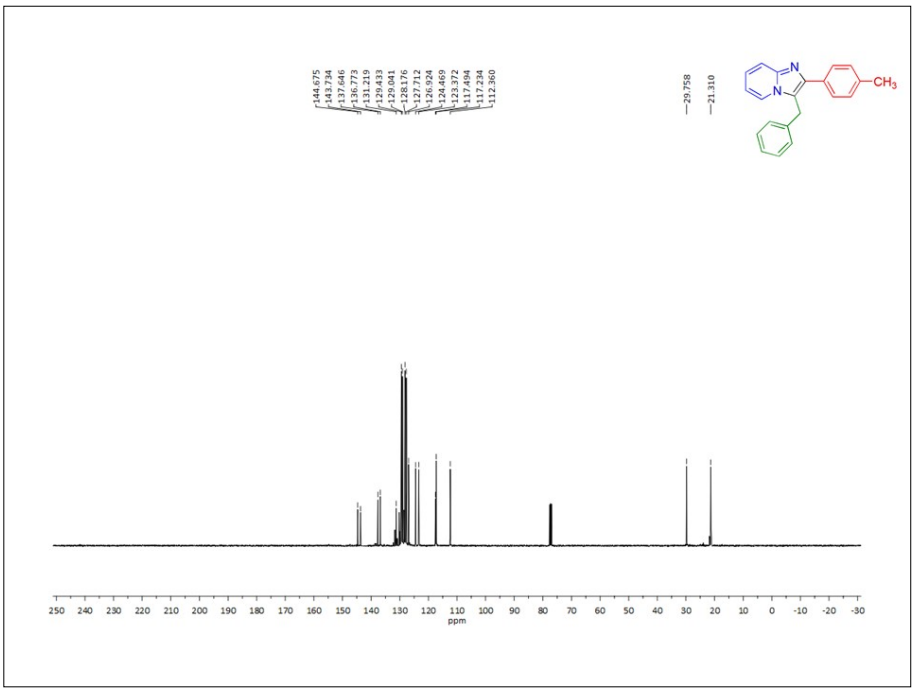


Fig. S51 ^{13}C NMR of 3-Benzyl-2-*p*-tolylimidazo[1,2-*a*]pyridine (4f)

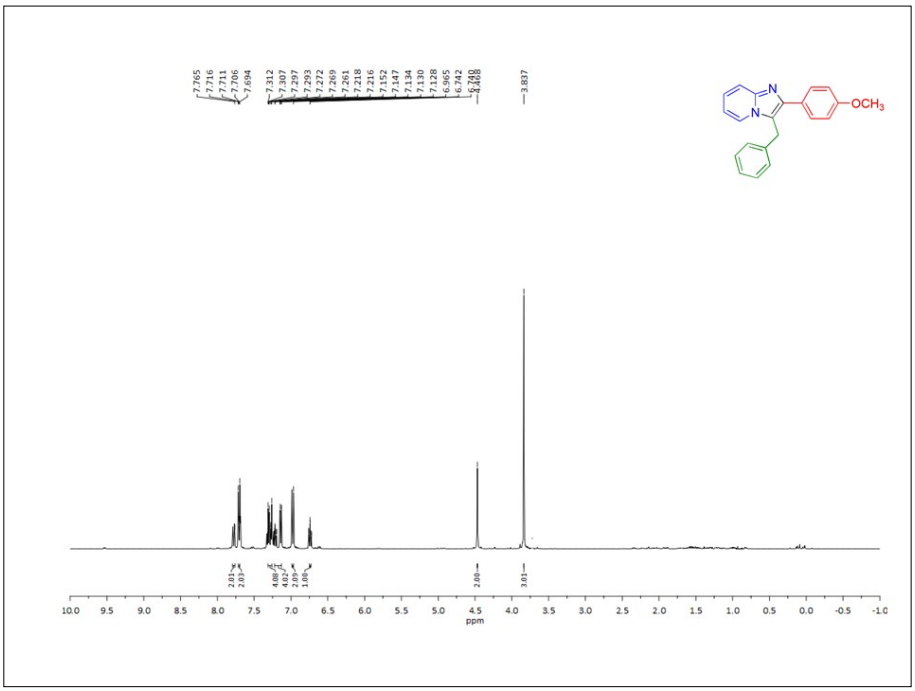


Fig. S52 ^{1}H NMR of 3-benzyl-2-(4-methoxyphenyl) imidazo[1,2-a]pyridine (4g)

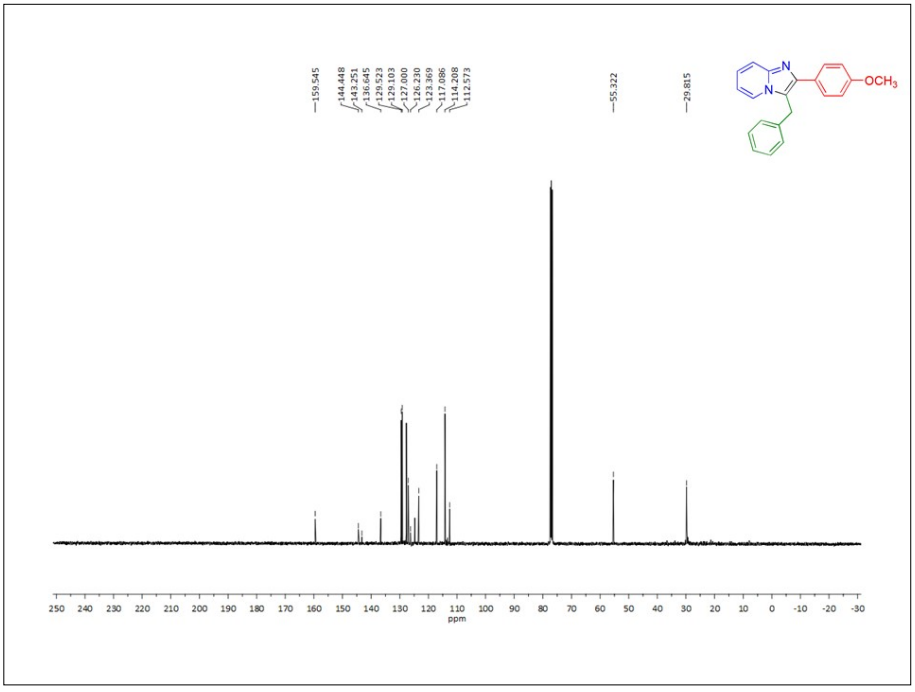


Fig. S53 ^{13}C NMR of 3-benzyl-2-(4-methoxyphenyl)imidazo[1,2-a]pyridine (4g)

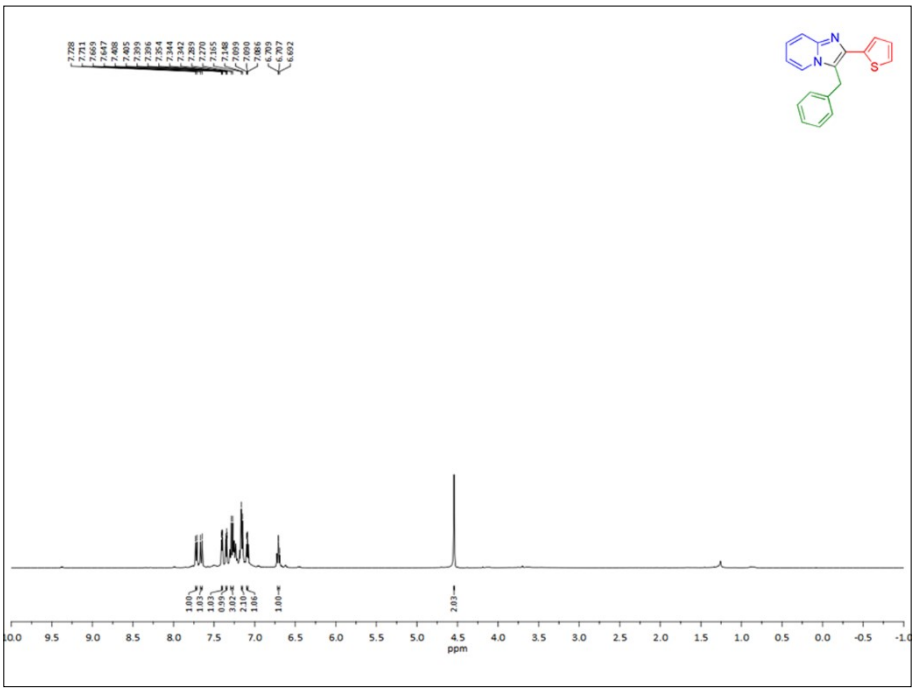


Fig. S54 ^{1}H NMR of 3-Benzyl-2-(2-thiophenyl)imidazo[1,2-*a*]pyridine (4h)

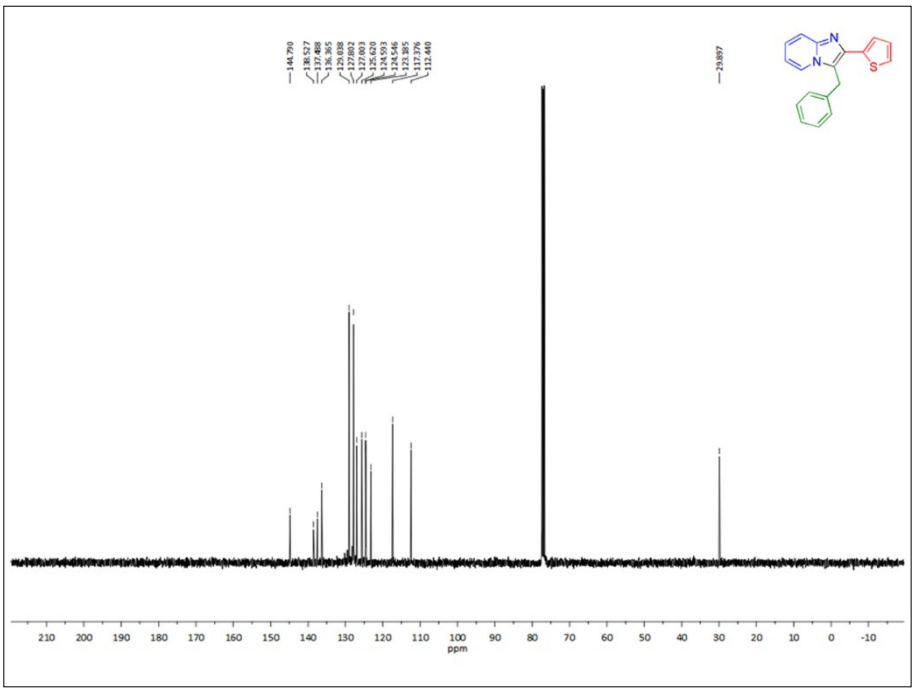


Fig. S55 ^{13}C NMR of 3-Benzyl-2-(2-thiophenyl)imidazo[1,2-*a*]pyridine (4h)

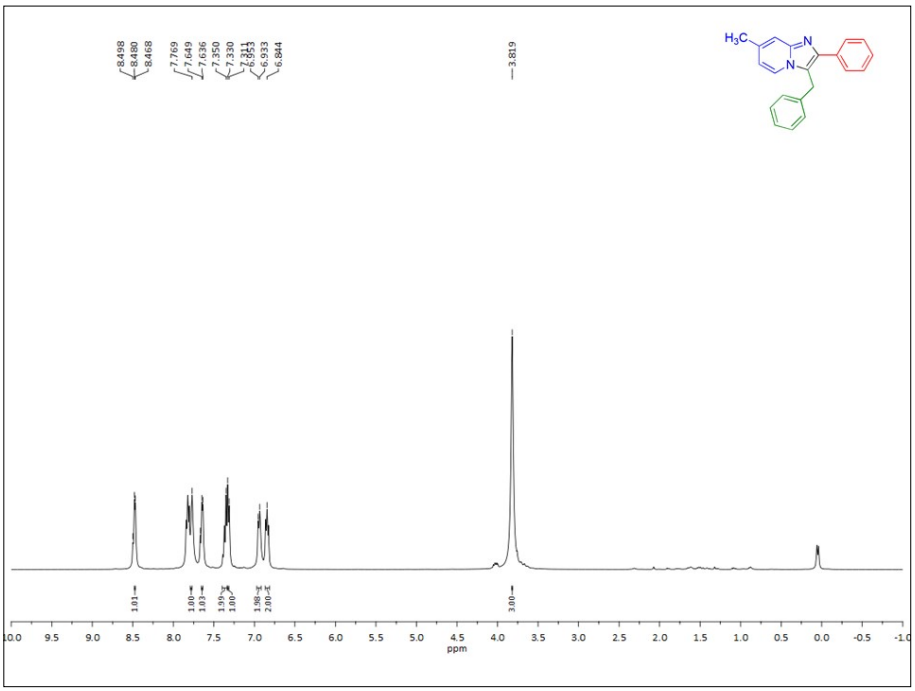


Fig. S56 ^{1}H NMR of 3-Benzyl-7-methyl-2-phenylimidazo[1,2-*a*]pyridine (4i)

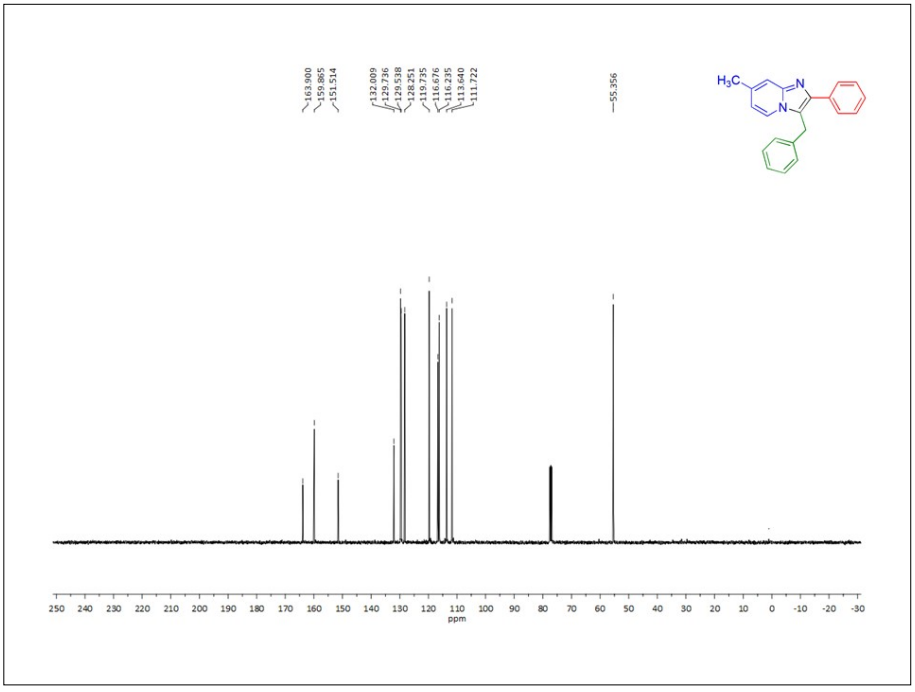


Fig. S57 ^{13}C NMR of 3-Benzyl-7-methyl-2-phenylimidazo[1,2-*a*]pyridine (4i)

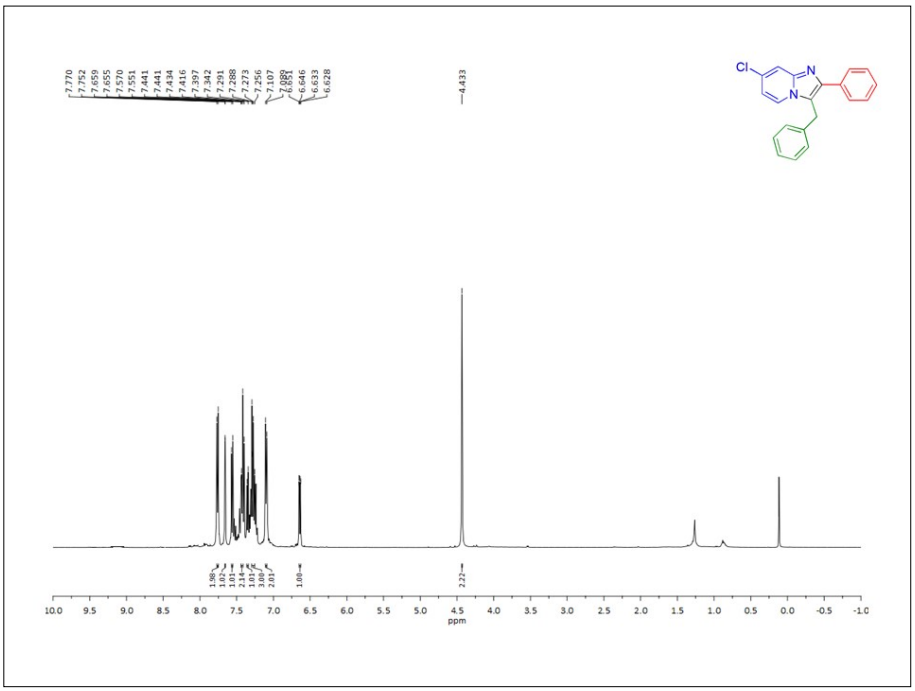


Fig. S58 ^{1}H NMR of 3-Benzyl-7-chloro-2-phenylimidazo[1,2-*a*]pyridine (4j)

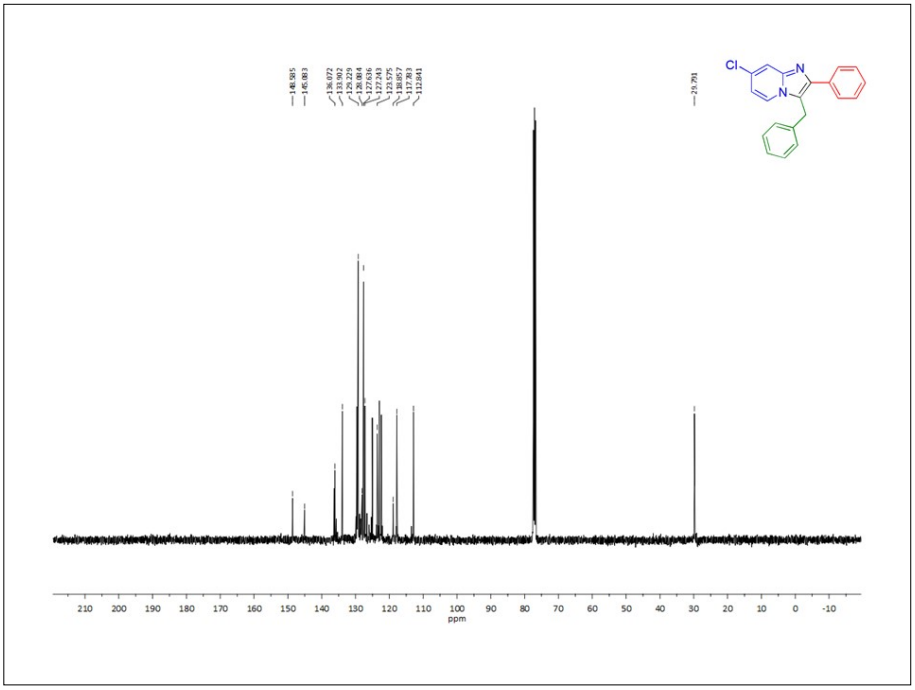


Fig. S59 ^{13}C NMR of 3-Benzyl-7-chloro-2-phenylimidazo[1,2-*a*]pyridine (4j)

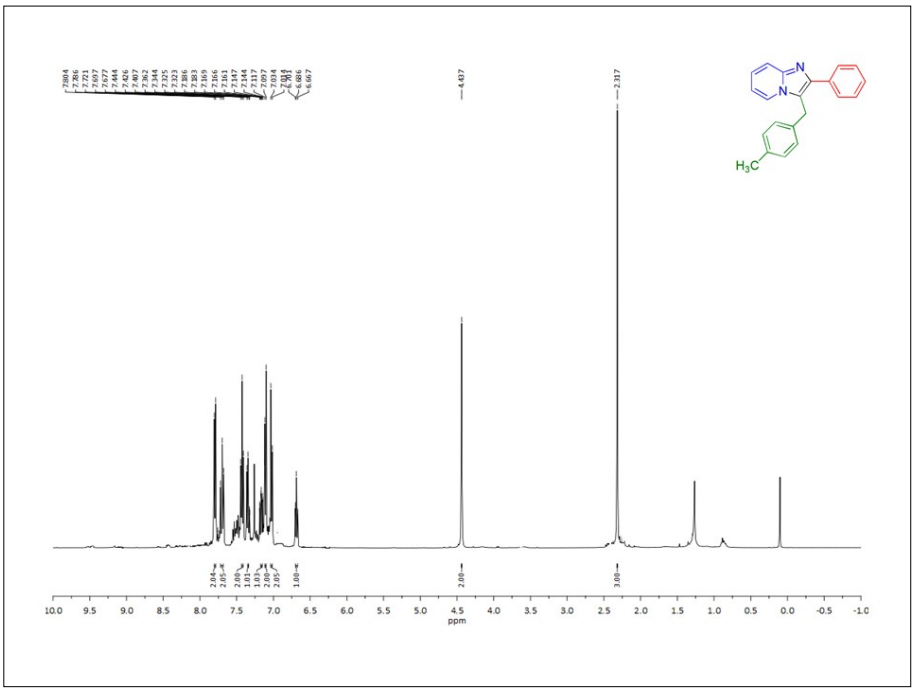


Fig. S60 ^{1}H NMR of 3-(4-Methylbenzyl)-2-phenylimidazo[1,2-*a*]pyridine (4k)

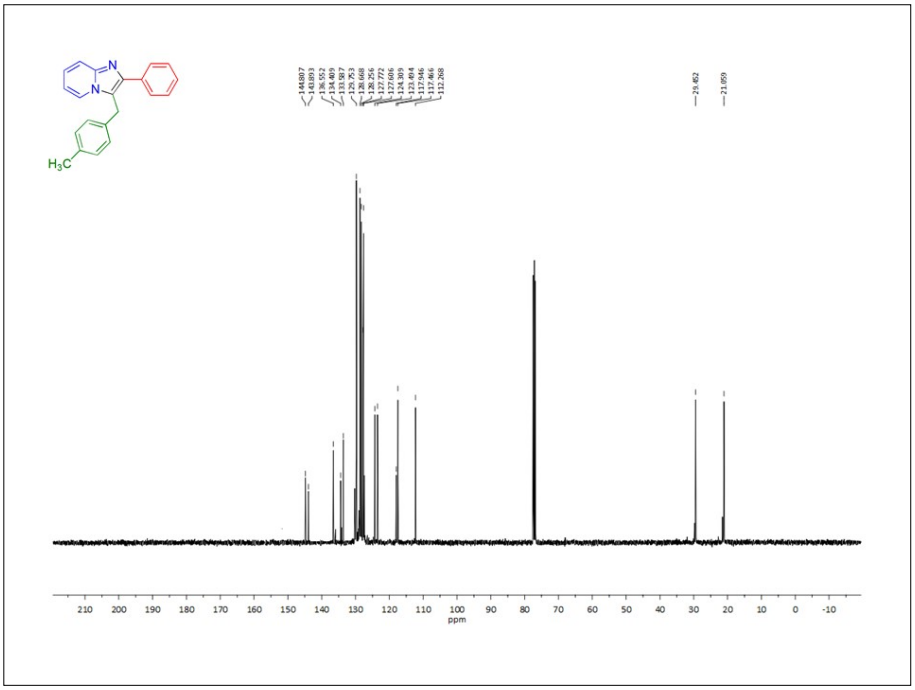


Fig. S61 ^{13}C NMR of 3-(4-Methylbenzyl)-2-phenylimidazo[1,2-*a*]pyridine (4k)

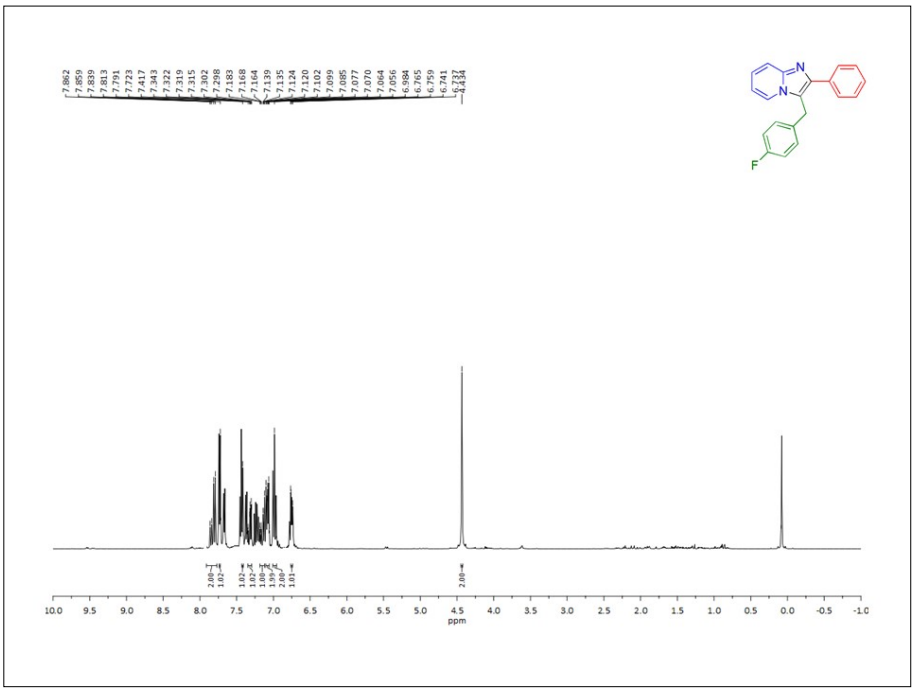


Fig. S62 ^{1}H NMR of 3-(4-Fluorobenzyl)-2-phenylimidazo[1,2-*a*]pyridine (4l)

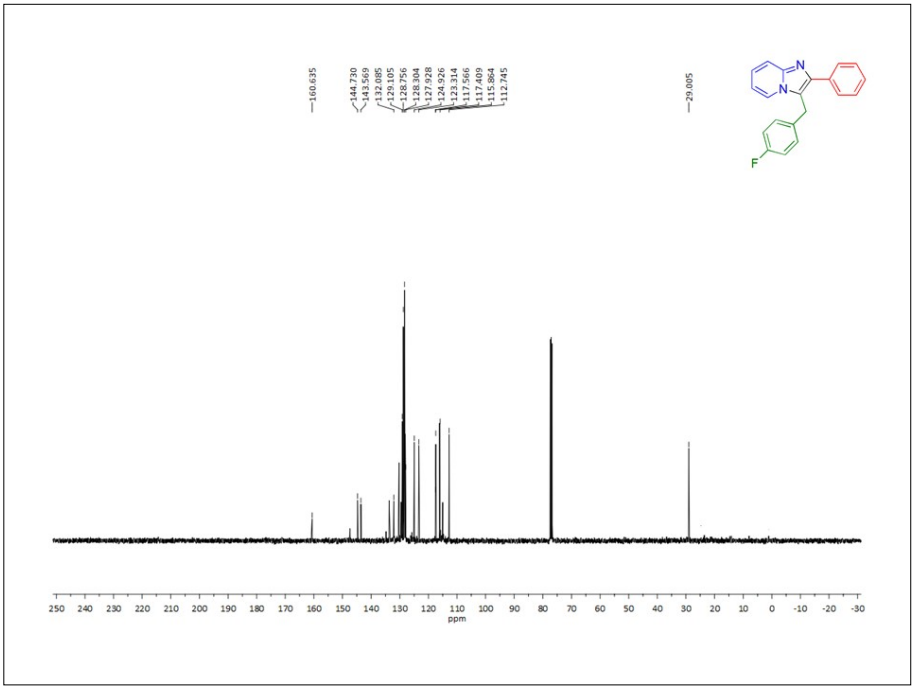


Fig. S63 ^{13}C NMR of 3-(4-Fluorobenzyl)-2-phenylimidazo[1,2-*a*]pyridine (4l)

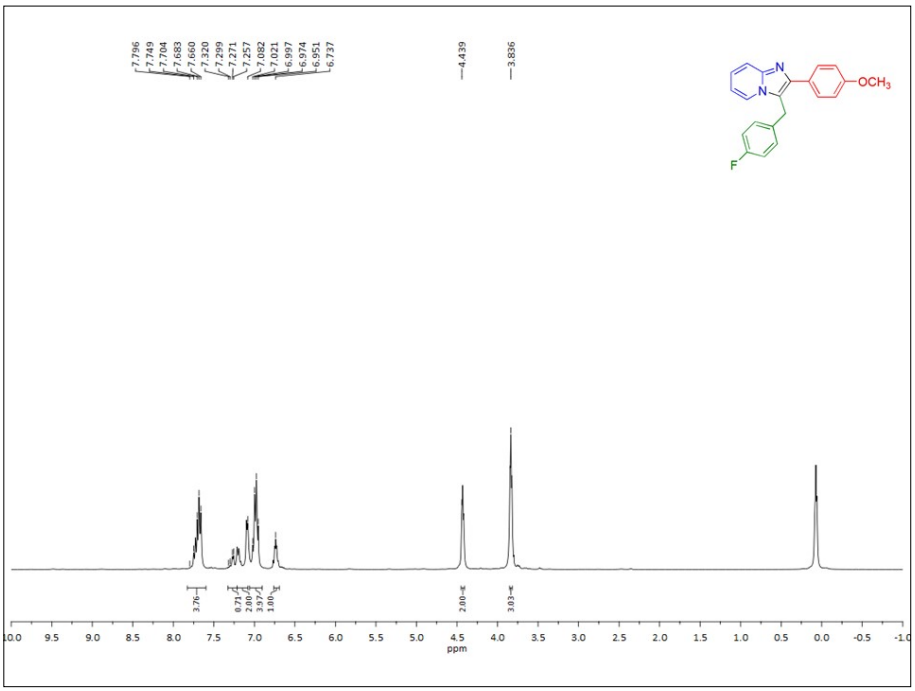


Fig. S64 ^{1}H NMR of 3-(4-fluorobenzyl)-2-(4-methoxyphenyl)imidazo[1,2-a]pyridine (4m)

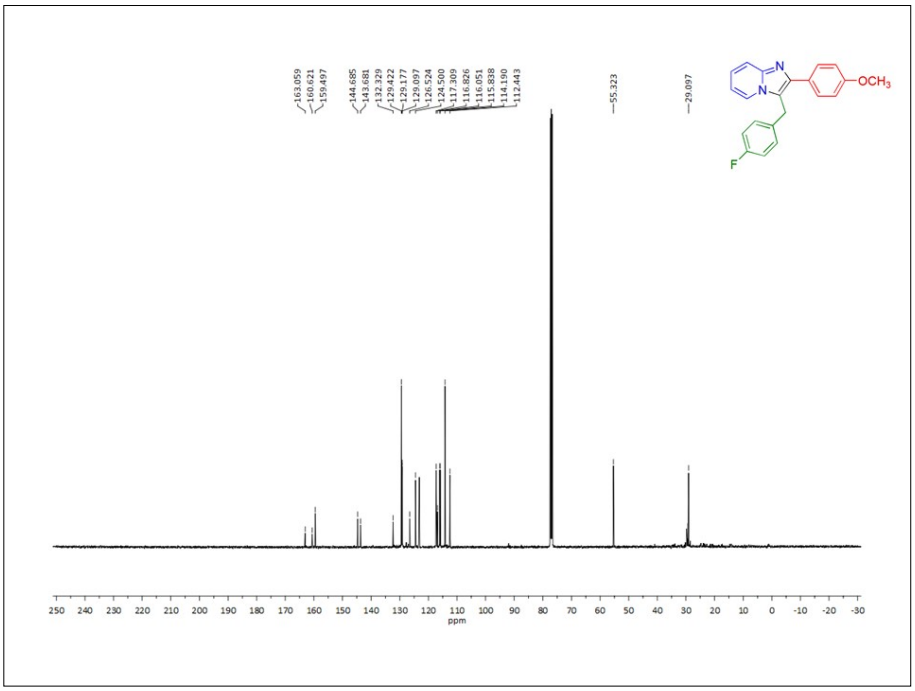


Fig. S65 ^{1}H NMR of 3-(4-fluorobenzyl)-2-(4-methoxyphenyl)imidazo[1,2-a]pyridine (4m)

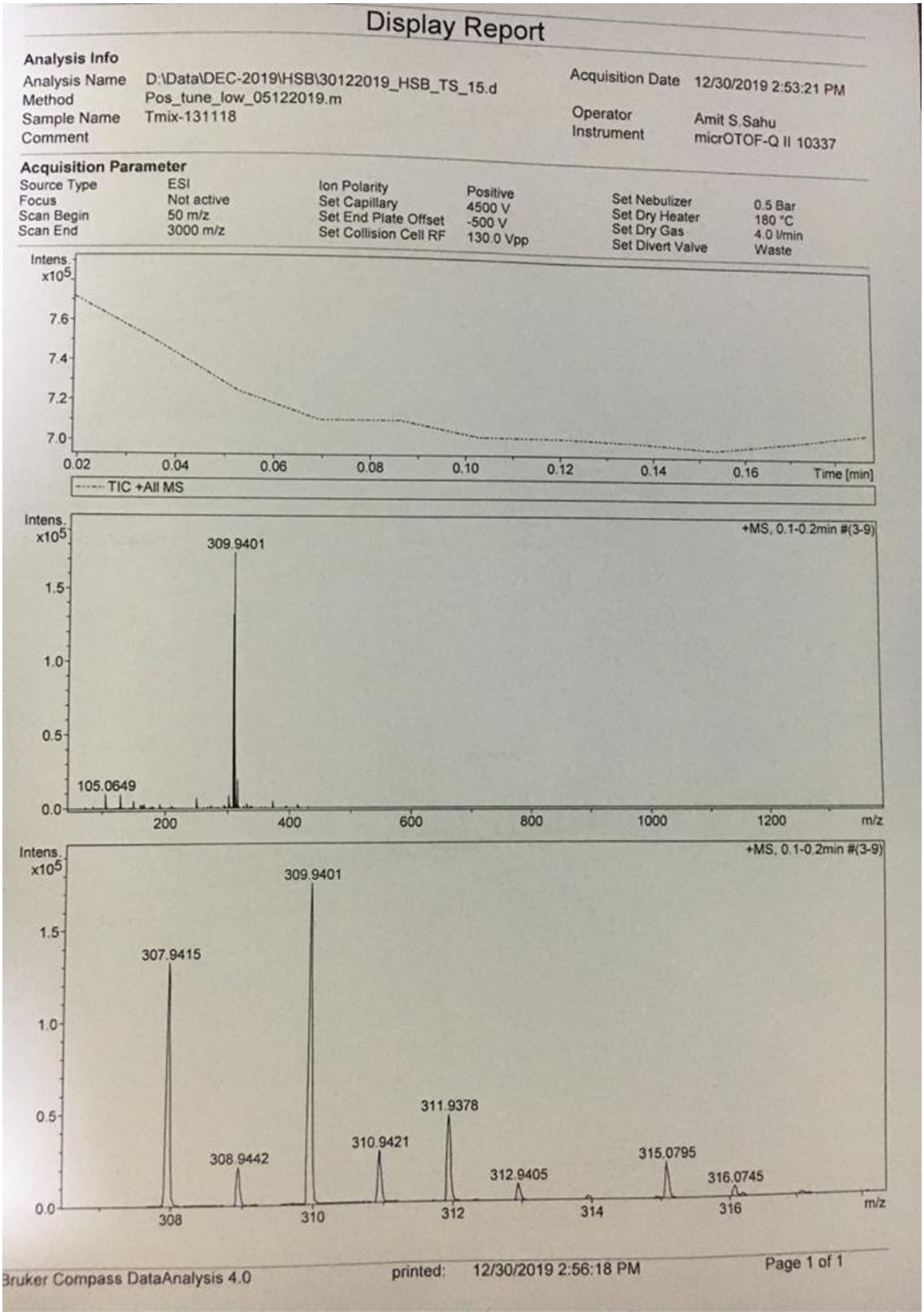


Fig. S66 HRMS of 2-(4-Bromophenyl)-7-chloro-[1,2,4]triazolo[1,5-a]pyridine (3l)

[1] S. Ueda and H. Nagasawa, *J. Am. Chem.Soc.*, 2009, ***131***, 15080-1581.
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[3] X. Meng, C. Yu and P. Zhao, *RSC Adv.*, 2014, **4**, 8612- 8616.

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