

Supporting Information

Highly Facile and Regio-Selective Synthesis of Pyrazolo[1,5-*a*]pyrimidines via Reactions of 1,2-Allenic Ketones with Aminopyrazoles

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I. General Experimental Information

All the commercial reagents and solvents were used without further purification. ^1H and ^{13}C NMR spectra were recorded at 400 and 100 MHz, respectively. Splitting patterns are designated as s (singlet), d (doublet), t (triplet), q (quartet), m (multiplet), and br. (broad). High-resolution mass spectra (HRMS) were obtained by using a MicroTOF mass spectrometer. All reactions were monitored by thin-layer chromatography (TLC) using silica gel plates (silica gel 60 F254 0.25 mm) and components were visualized by observation under UV light.

II. Experimental Procedures and Spectroscopic Data

2.1 General procedure for the preparation of pyrazolo[1,5-*a*]pyrimidines **3**, **4**, **7** and **11**

A mixture of 1,2-allenic ketone (1 mmol) and aminopyrazole **2** (1 mmol) in acetone (4 mL) was stirred at rt. Upon completion as indicated by TLC, the resulting mixture was concentrated under vacuum. The residue was purified by chromatography on silica gel to afford the title product.

5-methyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3a): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.60 (s, 3H), 6.61 (d, $J = 2.0$ Hz, 1H), 6.71 (s, 1H), 7.51 (t, $J = 3.2$ Hz, 3H), 7.94-7.97 (m, 2H), 8.07 (d, $J = 2.0$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.8, 95.9, 108.3, 128.7, 129.2, 130.9, 131.2, 144.7, 146.1, 149.5, 158.7. HRMS calcd for $\text{C}_{13}\text{H}_{12}\text{N}_3$: 210.1031 [M+H], found: 210.1037.

5-methyl-7-(4-(trifluoromethyl)phenyl)pyrazolo[1,5-*a*]pyrimidine (3b): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.58 (s, 3H), 6.59 (d, $J = 2.4$ Hz, 1H), 6.70 (s, 1H), 7.73 (d, $J = 8.4$ Hz, 2H), 8.02-8.06 (m, 3H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.7, 96.2, 108.6, 119.6, 122.3, 125.0, 125.5, 125.5, 125.6, 125.6, 127.8, 129.6, 132.0, 132.3, 132.6, 132.9, 134.6, 144.4, 144.7, 149.4, 158.8. HRMS calcd for $\text{C}_{14}\text{H}_{11}\text{F}_3\text{N}_3$: 278.0905 [M+H], found: 278.0911.

7-(4-bromophenyl)-5-methylpyrazolo[1,5-*a*]pyrimidine (3c): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.57 (s, 3H), 6.58 (d, $J = 2.0$ Hz, 1H), 6.66 (s, 1H), 7.59 (dd, $J_1 = 6.4$ Hz, $J_2 = 2.0$ Hz, 2H), 7.82 (dd, $J_1 = 6.4$ Hz, $J_2 = 2.0$ Hz, 2H), 8.02 (d, $J = 2.4$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.8, 96.0, 108.1, 125.4, 129.9, 130.7, 131.9, 144.6, 144.8, 149.5, 158.6. HRMS calcd for $\text{C}_{13}\text{H}_{11}\text{BrN}_3$: 288.0136 [M+H], found: 288.0145.

7-(4-methoxyphenyl)-5-methylpyrazolo[1,5-*a*]pyrimidine (3d): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.53 (s, 3H), 3.78 (s, 3H), 6.54 (d, $J = 2.0$ Hz, 1H), 6.63 (s, 1H), 6.95 (d, $J = 8.8$ Hz, 2H), 7.93 (d, $J = 8.8$ Hz, 2H), 8.02 (d, $J = 2.4$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.7, 55.4, 95.6, 107.4,

114.0, 123.2, 130.9, 144.4, 145.8, 149.6, 158.6, 161.6. HRMS calcd for C₁₄H₁₄N₃O: 240.1137 [M+H], found: 240.1131.

5-methyl-7-*p*-tolylpyrazolo[1,5-*a*]pyrimidine (3e): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.42 (s, 3H), 2.60 (s, 3H), 6.61 (d, *J* = 2.0 Hz, 1H), 6.71 (s, 1H), 7.32 (d, *J* = 8.0 Hz, 2H), 7.88 (d, *J* = 8.0 Hz, 2H), 8.07 (d, *J* = 2.0 Hz, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 21.6, 24.7, 95.7, 108.0, 128.2, 129.1, 129.3, 141.3, 144.6, 146.3, 149.5, 158.7. HRMS calcd for C₁₄H₁₄N₃: 224.1188 [M+H], found: 224.1181.

7-(3-chlorophenyl)-5-methylpyrazolo[1,5-*a*]pyrimidine (3f): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.56 (s, 3H), 6.58 (d, *J* = 2.4 Hz, 1H), 6.67 (s, 1H), 7.38-7.44 (m, 2H), 7.82 (d, *J* = 7.6 Hz, 1H), 7.93 (s, 1H), 8.03 (d, *J* = 2.0 Hz, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 24.8, 96.1, 108.4, 127.3, 129.2, 129.9, 130.9, 132.7, 134.6, 144.4, 144.7, 149.5, 158.7. HRMS calcd for C₁₃H₁₁ClN₃: 244.0642 [M+H], found: 244.0651.

7-(2-fluorophenyl)-5-methylpyrazolo[1,5-*a*]pyrimidine (3g): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.63 (s, 3H), 6.64 (d, *J* = 2.4 Hz, 1H), 6.76 (s, 1H), 7.26 (t, *J* = 9.2 Hz, 1H), 7.31 (t, *J* = 7.2 Hz, 1H), 7.50-7.54 (m, 1H), 7.75 (td, *J*₁ = 7.6 Hz, *J*₂ = 1.6 Hz, 1H), 8.07 (d, *J* = 2.4 Hz, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 24.8, 96.1, 110.2, 110.2, 116.3, 116.6, 119.2, 124.4, 124.4, 131.1, 131.1, 132.6, 132.6, 141.3, 144.7, 149.1, 158.4, 158.7, 161.2. HRMS calcd for C₁₃H₁₁FN₃: 228.0937 [M+H], found: 228.0945.

5-methyl-7-(naphthalen-1-yl)pyrazolo[1,5-*a*]pyrimidine (3h): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.63 (s, 3H), 6.69 (d, *J* = 2.0 Hz, 1H), 6.72 (s, 1H), 7.36-7.38 (m, 2H), 7.45-7.49 (m, 1H), 7.55 (t, *J* = 7.2 Hz, 1H), 7.63 (d, *J* = 6.4 Hz, 1H), 7.89 (d, *J* = 8.4 Hz, 1H), 7.98 (s, 1H), 8.01 (d, *J* = 2.0 Hz, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 24.8, 95.9, 110.6, 125.1, 125.2, 126.5, 127.0, 127.9, 128.7, 129.3, 130.7, 131.0, 133.6, 144.9, 146.0, 149.3, 158.6. HRMS calcd for C₁₇H₁₄N₃: 260.1188 [M+H], found: 260.1182.

5-methyl-7-(thiophen-2-yl)pyrazolo[1,5-*a*]pyrimidine (3i): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.61 (s, 3H), 6.61 (d, $J = 2.0$ Hz, 1H), 7.05 (s, 1H), 7.21 (t, $J = 4.4$ Hz, 1H), 7.66 (d, $J = 4.8$ Hz, 1H), 8.17 (d, $J = 1.6$ Hz, 1H), 8.26 (d, $J = 2.4$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.7, 95.8, 104.7, 127.6, 131.2, 131.5, 131.9, 139.4, 144.3, 149.5, 158.0. HRMS calcd for $\text{C}_{11}\text{H}_{10}\text{N}_3\text{S}$: 216.0595 [M+H], found: 216.0587.

7-benzyl-5-methylpyrazolo[1,5-*a*]pyrimidine (3j): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.45 (s, 3H), 4.42 (s, 2H), 6.19 (s, 1H), 6.56 (d, $J = 2.4$ Hz, 1H), 7.27-7.33 (m, 5H), 8.06 (d, $J = 2.0$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.7, 36.2, 95.9, 107.7, 127.4, 128.9, 129.7, 134.8, 144.3, 148.1, 148.6, 158.5. HRMS calcd for $\text{C}_{14}\text{H}_{14}\text{N}_3$: 224.1188 [M+H], found: 224.1192.

5-methyl-7-phenethylpyrazolo[1,5-*a*]pyrimidine (3k): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.46 (s, 3H), 3.10 (t, $J = 7.6$ Hz, 2H), 3.35 (t, $J = 7.6$ Hz, 2H), 6.36 (s, 1H), 6.53 (d, $J = 2.4$ Hz, 1H), 7.13-7.23 (m, 5H), 8.04 (d, $J = 2.0$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.7, 31.9, 32.2, 95.7, 107.4, 126.4, 128.4, 128.6, 140.1, 144.2, 147.8, 148.6, 158.3. HRMS calcd for $\text{C}_{15}\text{H}_{16}\text{N}_3$: 238.1344 [M+H], found: 238.1341.

5-ethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3l): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 1.41 (t, $J = 8.0$ Hz, 3H), 2.93 (q, $J = 7.6$ Hz, 2H), 6.68 (s, 1H), 6.79 (s, 1H), 7.56 (s, 3H), 8.01 (d, $J = 3.6$ Hz, 2H), 8.12 (s, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 13.1, 31.5, 96.0, 107.4, 128.7, 129.2, 130.9, 131.4, 144.7, 144.8, 149.6, 163.6. HRMS calcd for $\text{C}_{14}\text{H}_{14}\text{N}_3$: 224.1188 [M+H], found: 224.1185.

2,5-dimethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3m): yellow solid. ^1H NMR (400 MHz, CDCl_3) δ : 2.49 (s, 3H), 2.60 (s, 3H), 6.41 (s, 1H), 6.66 (s, 1H), 7.53 (t, $J = 2.8$ Hz, 3H), 8.00-8.03 (m, 2H); ^{13}C NMR (100 MHz, CDCl_3) δ : 14.8, 24.6, 95.2, 107.5, 128.6, 129.3, 130.8, 131.4, 145.6, 150.3, 154.9, 158.2. HRMS calcd for $\text{C}_{14}\text{H}_{14}\text{N}_3$: 224.1188 [M+H], found: 224.1185.

7-(2-fluorophenyl)-2,5-dimethylpyrazolo[1,5-*a*]pyrimidine (3n): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.44 (s, 3H), 2.57 (s, 3H), 6.40 (s, 1H), 6.64 (s, 1H), 7.20 (t, *J*=9.2 Hz, 1H), 7.26 (t, *J*=8.0 Hz, 1H), 7.44-7.48 (m, 1H), 7.75-7.78 (m, 1H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.8, 24.7, 95.4, 109.3, 109.3, 116.3, 116.5, 119.4, 119.5, 124.3, 124.3, 131.2, 131.2, 132.4, 132.4, 140.6, 149.9, 154.9, 157.9, 158.7, 161.2. HRMS calcd for C₁₄H₁₃FN₃: 242.1094 [M+H], found: 242.1099.

7-(3-chlorophenyl)-2,5-dimethylpyrazolo[1,5-*a*]pyrimidine (3o): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.50 (s, 3H), 2.61 (s, 3H), 6.43 (s, 1H), 6.66 (s, 1H), 7.47-7.51 (m, 2H), 7.92 (d, *J*=7.2 Hz, 1H), 8.01 (s, 1H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.8, 24.7, 95.5, 107.6, 127.4, 129.3, 129.9, 130.9, 133.0, 134.7, 144.1, 150.2, 155.2, 158.3. HRMS calcd for C₁₄H₁₃ClN₃: 258.0798 [M+H], found: 258.0805.

7-(4-bromophenyl)-2,5-dimethylpyrazolo[1,5-*a*]pyrimidine (3p): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.42 (s, 3H), 2.52 (s, 3H), 6.35 (s, 1H), 6.56 (s, 1H), 7.57 (d, *J* = 8.4 Hz, 2H), 7.84 (d, *J* = 8.8 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.8, 24.7, 95.4, 107.2, 125.3, 130.1, 130.8, 131.8, 144.2, 150.2, 154.9, 158.2. HRMS calcd for C₁₄H₁₃BrN₃: 302.0293 [M+H], found: 302.0299.

2,5-dimethyl-7-(4-(trifluoromethyl)phenyl)pyrazolo[1,5-*a*]pyrimidine (3q): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.46 (s, 3H), 2.58 (s, 3H), 6.41 (s, 1H), 6.64 (s, 1H), 7.76 (d, *J* = 8.4 Hz, 2H), 8.11 (d, *J* = 8.4 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.7, 24.7, 95.6, 107.8, 119.7, 122.4, 125.1, 125.5, 125.6, 125.6, 125.6, 127.9, 129.7, 131.9, 132.2, 132.6, 132.9, 134.8, 143.9, 150.3, 155.1, 158.3. HRMS calcd for C₁₄H₁₃F₃N₃: 292.1062 [M+H], found: 292.1068.

2,5-dimethyl-7-*p*-tolylpyrazolo[1,5-*a*]pyrimidine (3r): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.42 (s, 3H), 2.48 (s, 3H), 2.58 (s, 3H), 6.39 (s, 1H), 6.63 (s, 1H), 7.32 (d, *J* = 7.6 Hz, 2H), 7.91 (d, *J* = 8.4 Hz, 2H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.9, 21.6, 24.7, 95.1, 107.1, 128.4, 129.2, 129.3, 141.2, 145.7, 150.3, 154.8, 158.2. HRMS calcd for C₁₅H₁₆N₃: 238.1344 [M+H], found: 238.1341.

7-(3,4-dimethoxyphenyl)-2,5-dimethylpyrazolo[1,5-*a*]pyrimidine (3s): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.44 (s, 3H), 2.53 (s, 3H), 3.89 (s, 3H), 3.90 (s, 3H), 6.34 (s, 1H), 6.60 (s, 1H), 6.93 (d, *J* = 8.4 Hz, 1H), 7.59 (dd, *J*₁ = 8.4 Hz, *J*₂ = 1.6 Hz, 1H), 7.64 (d, *J*₁ = 1.2 Hz, 1H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.8, 24.6, 55.9, 56.0, 94.9, 106.7, 110.8, 112.1, 122.6, 123.5, 145.2, 148.5, 150.4, 151.1, 154.6, 158.1. HRMS calcd for C₁₆H₁₈N₃O₂: 284.1399 [M+H], found: 284.1403.

2,5-dimethyl-7-(naphthalen-1-yl)pyrazolo[1,5-*a*]pyrimidine (3t): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.37 (s, 3H), 2.59 (s, 3H), 6.48 (s, 1H), 6.60 (s, 1H), 7.33-7.37 (m, 1H), 7.42-7.47 (m, 2H), 7.53 (t, *J* = 8.0 Hz, 1H), 7.64 (d, *J* = 6.4 Hz, 1H), 7.87 (d, *J* = 8.4 Hz, 1H), 7.96 (d, *J* = 8.0 Hz, 1H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.9, 24.7, 95.4, 109.9, 125.3, 125.3, 126.4, 126.9, 127.9, 128.6, 129.4, 130.7, 130.8, 133.6, 145.3, 150.0, 155.1, 158.1. HRMS calcd for C₁₈H₁₆N₃: 274.1344 [M+H], found: 274.1341.

2,5-dimethyl-7-(thiophen-2-yl)pyrazolo[1,5-*a*]pyrimidine (3u): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.55 (s, 3H), 2.60 (s, 3H), 6.40 (s, 1H), 6.98 (s, 1H), 7.21-7.23 (m, 1H), 7.65 (d, *J* = 4.4 Hz, 1H), 8.31 (d, *J* = 2.0 Hz, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 14.9, 24.7, 95.1, 103.9, 127.6, 131.3, 131.6, 131.8, 138.8, 150.3, 154.8, 157.5. HRMS calcd for C₁₂H₁₂N₃S: 230.0752 [M+H], found: 230.0758.

7-benzyl-2,5-dimethylpyrazolo[1,5-*a*]pyrimidine (3v): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.37 (s, 3H), 2.47 (s, 3H), 4.36 (s, 2H), 6.03 (s, 1H), 6.32 (s, 1H), 7.22-7.30 (m, 5H); ¹³C NMR (100MHz, CDCl₃) δ: 14.7, 24.6, 36.1, 95.2, 106.7, 127.3, 128.8, 129.8, 134.9, 147.6, 149.3, 154.4, 158.1. HRMS calcd for C₁₅H₁₆N₃: 238.1344 [M+H], found: 238.1345.

2,5-dimethyl-7-phenethylpyrazolo[1,5-*a*]pyrimidine (3w): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 2.42 (s, 3H), 2.46 (s, 3H), 3.08 (t, *J* = 8.0 Hz, 2H), 3.31 (t, *J* = 8.0 Hz, 2H), 6.26 (s, 1H), 6.31 (s, 1H), 7.13-7.24 (m, 5H); ¹³C NMR (100MHz, CDCl₃) δ: 14.7, 24.6, 31.8, 32.0, 95.0, 106.3, 126.4, 128.4, 128.5, 140.2, 147.3, 149.3, 154.3, 157.9. HRMS calcd for C₁₆H₁₈N₃: 252.1501 [M+H], found: 252.1509.

5-ethyl-2-methyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3x): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 1.38 (t, $J = 8.0$ Hz, 3H), 2.50 (s, 3H), 2.87 (q, $J = 8.0$ Hz, 2H), 6.44 (s, 1H), 6.69 (s, 1H), 7.54 (t, $J = 3.2$ Hz, 3H), 8.02 (d, $J = 4.0$ Hz, 2H); ^{13}C NMR (100MHz, CDCl_3) δ : 13.2, 14.8, 31.5, 95.3, 106.5, 128.6, 129.2, 130.8, 131.5, 145.9, 150.3, 154.9, 163.2. HRMS calcd for $\text{C}_{15}\text{H}_{16}\text{N}_3$: 238.1344 [M+H], found: 238.1348.

2-cyclopropyl-5-methyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3y): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 0.85-0.87 (m, 2H), 0.95-0.99 (m, 2H), 2.06-2.10 (m, 1H), 2.50 (s, 3H), 6.20 (s, 1H), 6.56 (s, 1H), 7.44 (d, $J = 2.4$ Hz, 3H), 7.97-7.98 (m, 2H); ^{13}C NMR (100MHz, CDCl_3) δ : 9.3, 10.2, 24.6, 91.2, 107.2, 128.4, 129.3, 130.7, 131.3, 145.2, 150.4, 158.2, 161.3. HRMS calcd for $\text{C}_{16}\text{H}_{16}\text{N}_3$: 250.1344 [M+H], found: 250.1341.

7-(3-chlorophenyl)-2-cyclopropyl-5-methylpyrazolo[1,5-*a*]pyrimidine (3z): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 0.81-0.85 (m, 2H), 0.96-1.00 (m, 2H), 2.04-2.08 (m, 1H), 2.51 (s, 3H), 6.18 (s, 1H), 6.55 (s, 1H), 7.33-7.41 (m, 2H), 7.84 (d, $J = 7.2$ Hz, 1H), 7.98 (s, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 9.5, 10.2, 24.6, 91.4, 107.3, 127.4, 129.3, 129.7, 130.7, 132.9, 134.4, 143.6, 150.2, 158.2, 161.5. HRMS calcd for $\text{C}_{16}\text{H}_{15}\text{ClN}_3$: 284.0955 [M+H], found: 284.0953.

2-cyclopropyl-7-(4-methoxyphenyl)-5-methylpyrazolo[1,5-*a*]pyrimidine (3aa): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 0.81-0.84 (m, 2H), 0.93-0.97 (m, 2H), 2.03-2.08 (m, 1H), 2.46 (s, 3H), 3.75 (s, 3H), 6.14 (s, 1H), 6.50 (s, 1H), 6.91 (d, $J = 8.8$ Hz, 2H), 7.97 (d, $J = 8.8$ Hz, 2H); ^{13}C NMR (100MHz, CDCl_3) δ : 9.3, 10.2, 24.6, 55.3, 90.9, 106.3, 113.8, 123.3, 130.9, 144.9, 150.4, 158.0, 161.1, 161.5. HRMS calcd for $\text{C}_{17}\text{H}_{18}\text{N}_3\text{O}$: 280.1450 [M+H], found: 280.1453.

4-(2-cyclopropyl-5-methylpyrazolo[1,5-*a*]pyrimidin-7-yl)benzotrile (3bb): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 0.81-0.83 (m, 2H), 0.97-1.01 (m, 2H), 2.03-2.07 (m, 1H), 2.56 (s, 3H), 6.20 (s, 1H), 6.64 (s, 1H), 7.76 (d, $J = 7.6$ Hz, 2H), 8.13 (d, $J = 8.0$ Hz, 2H); ^{13}C NMR (100MHz, CDCl_3) δ : 9.5, 10.2,

24.7, 91.8, 107.7, 114.2, 118.2, 129.9, 132.2, 135.6, 143.0, 143.0, 150.2, 158.3, 161.8, 161.8. HRMS calcd for C₁₇H₁₅N₄: 275.1297 [M+H], found: 275.1295.

2-cyclopropyl-5-methyl-7-(thiophen-2-yl)pyrazolo[1,5-*a*]pyrimidine (3cc): yellow solid; ¹H NMR (400MHz, CDCl₃) δ: 0.98-1.07 (m, 4H), 2.12-2.15 (m, 1H), 2.53 (s, 3H), 6.27 (s, 1H), 6.91 (s, 1H), 7.15 (t, *J* = 4.0 Hz, 1H), 7.60 (d, *J* = 4.8 Hz, 1H), 8.19 (d, *J* = 4.0 Hz, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 9.7, 10.2, 24.7, 92.0, 103.3, 127.3, 130.9, 131.6, 131.9, 138.5, 150.1, 157.4, 161.0. HRMS calcd for C₁₄H₁₄N₃S: 256.0908 [M+H], found: 256.0903.

7-benzyl-2-cyclopropyl-5-methylpyrazolo[1,5-*a*]pyrimidine (3dd): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 0.90-0.94 (m, 2H), 1.01-1.06 (m, 2H), 2.11-2.15 (m, 1H), 2.41 (s, 3H), 4.38 (s, 2H), 6.06 (s, 1H), 6.20 (s, 1H), 7.26-7.35 (m, 5H); ¹³C NMR (100MHz, CDCl₃) δ: 9.3, 10.2, 24.6, 36.1, 91.5, 106.5, 127.3, 128.8, 129.9, 135.0, 147.6, 149.2, 158.1, 161.0. HRMS calcd for C₁₇H₁₈N₃: 264.1501 [M+H], found: 264.1505.

2-cyclopropyl-5-methyl-7-phenethylpyrazolo[1,5-*a*]pyrimidine (3ee): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 0.90-0.91 (m, 2H), 1.02 (d, *J* = 8.4 Hz, 2H), 2.09-2.12 (m, 1H), 2.44 (s, 3H), 3.10 (t, *J* = 8.4 Hz, 2H), 3.31 (t, *J* = 8.0 Hz, 2H), 6.18 (s, 1H), 6.28 (s, 1H), 7.17 (d, *J* = 6.8 Hz, 2H), 7.25 (t, *J* = 6.8 Hz, 3H); ¹³C NMR (100MHz, CDCl₃) δ: 9.4, 10.2, 24.6, 31.8, 32.1, 91.3, 106.2, 126.4, 128.4, 128.5, 140.4, 147.2, 149.3, 157.9, 160.9. HRMS calcd for C₁₈H₁₉N₃: 278.1657 [M+H], found: 278.1653.

2-cyclopropyl-5-ethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3ff): yellow solid; ¹H NMR (400MHz, CDCl₃) δ: 0.86-0.90 (m, 2H), 1.01-1.06 (m, 2H), 1.37 (t, *J* = 7.6 Hz, 3H), 2.11-2.15 (m, 1H), 2.86 (q, *J* = 7.6 Hz, 2H), 6.24 (s, 1H), 6.68 (s, 1H), 7.54 (t, *J* = 7.6 Hz, 3H), 8.04-8.06 (m, 2H); ¹³C NMR (100MHz, CDCl₃) δ: 9.4, 10.2, 13.2, 31.4, 91.1, 106.3, 128.5, 129.3, 130.8, 131.5, 145.7, 150.3, 161.6, 163.1. HRMS calcd for C₁₇H₁₈N₃: 264.1501 [M+H], found: 264.1508.

5-methyl-2,7-diphenylpyrazolo[1,5-*a*]pyrimidine (3gg): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.61 (s, 3H), 6.71 (s, 1H), 6.94 (s, 1H), 7.36 (t, $J = 7.2$ Hz, 1H), 7.44 (t, $J = 7.2$ Hz, 2H), 7.53-7.56 (m, 3H), 8.00-8.02 (m, 2H), 8.11-8.14 (m, 2H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.8, 92.5, 108.2, 126.6, 128.5, 128.7, 128.8, 129.5, 130.9, 131.1, 133.1, 145.6, 150.8, 155.6, 158.6. HRMS calcd for $\text{C}_{19}\text{H}_{16}\text{N}_3$: 286.1344 [M+H], found: 286.1348.

7-(4-bromophenyl)-5-methyl-2-phenylpyrazolo[1,5-*a*]pyrimidine (3hh): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.62 (s, 3H), 6.71 (s, 1H), 6.92 (s, 1H), 7.37 (t, $J = 7.6$ Hz, 1H), 7.44 (t, $J = 8.0$ Hz, 2H), 7.68-7.70 (m, 2H), 7.96-7.98 (m, 2H), 8.01-8.03 (m, 2H); ^{13}C NMR (100MHz, CDCl_3) δ : 24.8, 92.7, 107.9, 125.4, 126.5, 128.7, 128.9, 129.9, 131.0, 131.8, 132.9, 144.5, 150.7, 155.7, 158.6. HRMS calcd for $\text{C}_{19}\text{H}_{15}\text{BrN}_3$: 364.0449 [M+H], found: 364.0455.

5,6-dimethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3ii): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 2.14 (s, 3H), 2.62 (s, 3H), 6.58 (d, $J = 2.0$ Hz, 1H), 7.45-7.47 (m, 2H), 7.53-7.57 (m, 3H), 7.95 (d, $J = 2.0$ Hz, 1H); ^{13}C NMR (100MHz, CDCl_3) δ : 15.1, 24.1, 95.5, 114.9, 128.9, 129.4, 129.9, 130.9, 144.0, 147.4, 159.6. HRMS calcd for $\text{C}_{14}\text{H}_{14}\text{N}_3$: 224.1188 [M+H], found: 224.1185.

2,5,6-trimethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3jj): yellow solid. ^1H NMR (400 MHz, CDCl_3) δ : 2.08 (s, 3H), 2.39 (s, 3H), 2.57 (s, 3H), 6.35 (s, 1H), 7.44-7.54 (m, 5H); ^{13}C NMR (100 MHz, CDCl_3) δ : 14.9, 15.1, 24.1, 94.8, 113.9, 128.8, 129.6, 129.8, 131.0, 143.5, 148.2, 154.0, 159.1. HRMS calcd for $\text{C}_{15}\text{H}_{16}\text{N}_3$: 238.1344 [M+H], found: 238.1342.

2-cyclopropyl-5,6-dimethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3kk): yellow solid. ^1H NMR (400MHz, CDCl_3) δ : 0.74-0.77 (m, 2H), 0.93-0.96 (m, 2H), 2.01-2.03 (m, 1H), 2.08 (d, $J = 1.2$ Hz, 3H), 2.55 (d, $J = 2.0$ Hz, 3H), 6.11 (s, 1H), 7.45-7.54 (m, 5H); ^{13}C NMR (100MHz, CDCl_3) δ : 9.4, 10.2, 15.1, 24.0, 90.0, 113.7, 128.6, 129.7, 129.7, 130.9, 143.4, 148.2, 159.0, 160.7. HRMS calcd for $\text{C}_{17}\text{H}_{18}\text{N}_3$: 264.1501 [M+H], found: 264.1498.

6-ethyl-2,5-dimethyl-7-phenylpyrazolo[1,5-*a*]pyrimidine (3ll): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 1.05 (t, *J* = 7.6 Hz, 3H), 2.39 (s, 3H), 2.47 (q, *J* = 7.2 Hz, 2H), 2.64 (s, 3H), 6.36 (s, 1H), 7.41-7.43 (m, 2H), 7.53-7.56 (m, 3H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.8, 14.8, 21.7, 23.2, 94.7, 120.7, 128.9, 129.1, 129.6, 131.1, 143.9, 148.1, 154.3, 158.6. HRMS calcd for C₁₆H₁₈N₃: 252.1501 [M+H], found: 252.1503.

6,7-dimethyl-5-phenylpyrazolo[1,5-*a*]pyrimidine (4ii): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.32 (s, 3H), 2.86 (s, 3H), 6.67 (d, *J* = 2.0 Hz, 1H), 7.46-7.53 (m, 5H), 8.10 (d, *J* = 2.4 Hz, 1H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.1, 15.5, 96.7, 113.4, 128.4, 128.7, 128.7, 128.9, 139.7, 144.0, 146.9, 159.5. HRMS calcd for C₁₄H₁₄N₃: 224.1188 [M+H], found: 224.1181.

2,6,7-trimethyl-5-phenylpyrazolo[1,5-*a*]pyrimidine (4jj): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 2.26 (s, 3H), 2.52 (s, 3H), 2.80 (s, 3H), 6.43 (s, 1H), 7.43-7.51 (m, 5H); ¹³C NMR (100 MHz, CDCl₃) δ: 14.2, 14.7, 15.5, 95.8, 112.5, 128.3, 128.6, 128.7, 139.7, 143.4, 147.6, 154.2, 159.1. HRMS calcd for C₁₅H₁₆N₃: 238.1344 [M+H], found: 238.1351.

2-cyclopropyl-6,7-dimethyl-5-phenylpyrazolo[1,5-*a*]pyrimidine (4kk): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 0.88-0.92 (m, 2H), 1.02-1.07 (m, 2H), 2.12-2.16 (m, 1H), 2.24 (s, 3H), 2.79 (s, 3H), 6.25 (s, 1H), 7.40-7.50 (m, 5H); ¹³C NMR (100 MHz, CDCl₃) δ: 9.2, 10.2, 14.1, 15.5, 91.8, 112.3, 128.3, 128.6, 128.7, 139.8, 143.4, 147.6, 159.0, 160.8. HRMS calcd for C₁₇H₁₈N₃: 264.1501 [M+H], found: 264.1505.

6-ethyl-2,7-dimethyl-5-phenylpyrazolo[1,5-*a*]pyrimidine (4ll): yellow solid. ¹H NMR (400 MHz, CDCl₃) δ: 1.06 (t, *J* = 7.2 Hz, 3H), 2.54 (s, 3H), 2.68 (q, *J* = 7.2 Hz, 2H), 2.84 (s, 3H), 6.44 (s, 1H), 7.44-7.47 (m, 5H); ¹³C NMR (100 MHz, CDCl₃) δ: 13.9, 14.7, 15.1, 21.6, 95.8, 119.1, 128.2, 128.3, 128.5, 139.9, 143.3, 147.4, 154.4, 159.3. HRMS calcd for C₁₆H₁₈N₃: 252.1501 [M+H], found: 252.1504.

1-butyl-5-(5-methylpyrazolo[1,5-*a*]pyrimidin-7-yl)-4-(methylthio)pyrimidin-2(1*H*)-one (7): yellow solid. ¹H NMR (400MHz, CDCl₃) δ: 0.90 (t, *J* = 8.0 Hz, 3H), 1.35 (sext, *J* = 7.2 Hz, 2H), 1.73 (quint, *J* = 7.2 Hz, 2H), 2.47 (s, 3H), 2.58 (s, 3H), 3.86 (t, *J* = 7.2 Hz, 2H), 6.57 (d, *J* = 2.4 Hz, 1H), 6.86 (s, 1H), 7.99 (d, *J* = 2.4 Hz, 1H), 8.00 (s, 1H); ¹³C NMR (100MHz, CDCl₃) δ: 13.5, 13.7, 19.7, 24.8, 30.8, 51.2, 96.6, 107.1, 110.6, 138.5, 144.5, 146.9, 149.1, 153.1, 158.4, 175.6. HRMS calcd for C₁₆H₂₀N₅OS: 330.1389 [M+H], found: 330.1382.

((2*R*,3*S*,5*R*)-3-acetoxy-5-(5-(2,5-dimethylpyrazolo[1,5-*a*]pyrimidin-7-yl)-4-(methylthio)-2-oxopyrimidin-1(2*H*)-yl)tetrahydrofuran-2-yl)methyl acetate (11): yellowish solid. ¹H NMR (400 MHz, CDCl₃) δ: 1.54 (s, 3H), 2.00 (s, 3H), 2.13-2.20 (m, 1H), 2.32 (s, 3H), 2.42 (s, 3H), 2.49 (s, 3H), 2.71-2.77 (m, 1H), 4.17 (d, *J* = 2.8 Hz, 2H), 4.23-4.25 (m, 1H), 5.12-5.13 (m, 1H), 6.21 (t, *J* = 6.8 Hz, 1H), 6.30 (s, 1H), 6.70 (s, 1H), 8.28 (s, 1H); ¹³C NMR (100 MHz, CDCl₃) δ: 13.5, 14.6, 20.0, 20.8, 24.6, 38.8, 63.5, 74.0, 82.9, 87.3, 95.9, 107.7, 109.6, 137.9, 141.6, 149.7, 152.1, 154.8, 157.9, 169.9, 170.2, 176.4. HRMS calcd for C₂₂H₂₆N₅O₆S: 488.1604 [M+H], found: 488.1609.

2.2 General procedure for the preparation of nucleoside-pyrazolo[1,5-*a*]pyrimidine chimeras **8** and **12**

A sealed tube (50 mL) containing compound **7** or **11** (0.5 mmol), *i*-PrOH (5 mL), NH₃·H₂O (1 mL) and a magnetic stirring bar was heated to 90 °C for 8 h. The resulting mixture was then concentrated under vacuum, and the residue was purified by chromatography on silica gel to afford the nucleoside-pyrazolo[1,5-*a*]pyrimidine chimera **8** or **12**.

4-amino-1-butyl-5-(5-methylpyrazolo[1,5-*a*]pyrimidin-7-yl)pyrimidin-2(1*H*)-one (8): white solid. ¹H NMR (400 MHz, DMSO) δ: 0.88 (t, *J* = 7.2 Hz, 3H), 1.22-1.30 (m, 2H), 1.55-1.62 (m, 2H), 2.52 (s, 3H), 3.70 (t, *J* = 7.6 Hz, 2H), 6.58 (s, 1H), 6.94 (s, 1H), 8.09 (s, 1H), 8.13 (s, 1H); ¹³C NMR (100 MHz,

DMSO) δ : 14.1, 19.7, 24.7, 31.3, 49.1, 95.9, 98.1, 111.0, 140.3, 144.4, 149.0, 149.1, 155.2, 158.8, 162.8.

HRMS calcd for $C_{15}H_{19}N_6O$: 299.1620 [M+H], found: 299.1656.

4-amino-5-(2,5-dimethylpyrazolo[1,5-*a*]pyrimidin-7-yl)-1-((2R,4S,5R)-4-hydroxy-5-

(hydroxymethyl)tetrahydrofuran-2-yl)pyrimidin-2(1*H*)-one (12): white solid. 1H NMR (400 MHz,

DMSO) δ : 2.02-2.08 (m, 1H), 2.18-2.22 (m, 1H), 2.35 (s, 3H), 2.48 (s, 3H), 3.46-3.54 (m, 2H), 3.77 (s,

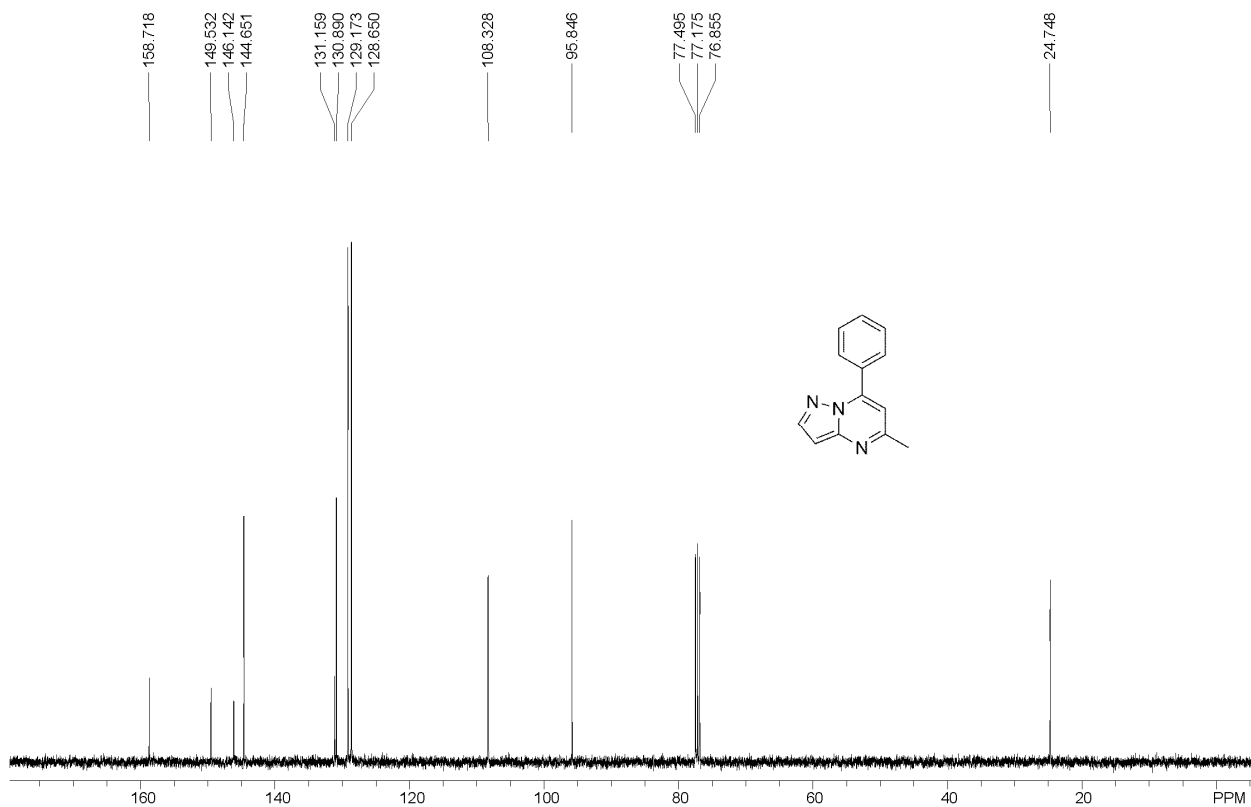
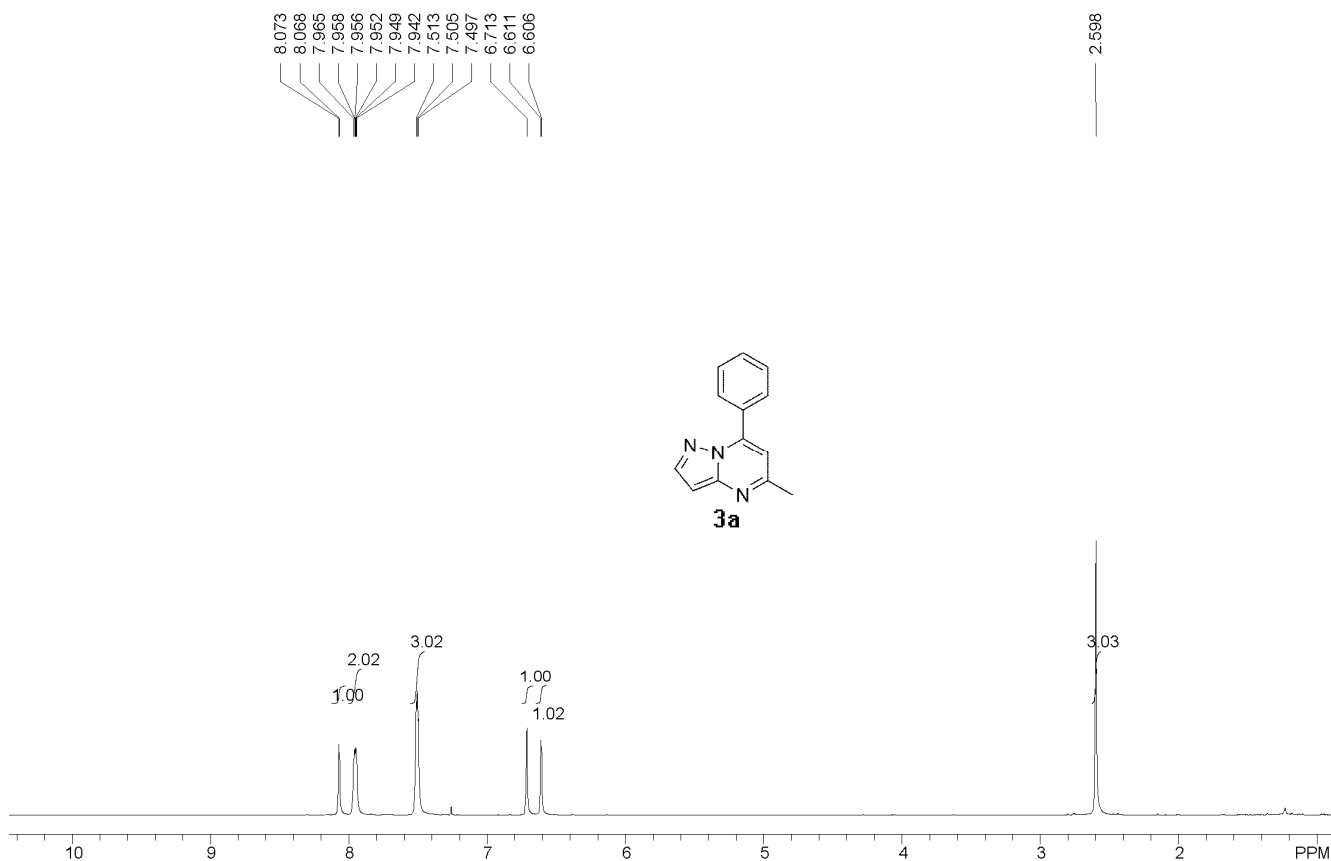
1H), 4.19 (s, 1H), 4.90 (t, $J = 4.4$ Hz, 1H), 5.22 (d, $J = 2.8$ Hz, 1H), 6.18 (t, $J = 6.0$ Hz, 1H), 6.37 (s, 1H),

6.69 (br., 1H), 6.81 (s, 1H), 7.48 (br., 1H), 8.28 (s, 1H); ^{13}C NMR (100 MHz, DMSO) δ : 14.9, 24.7, 41.2,

61.5, 70.5, 85.8, 87.9, 95.2, 99.3, 110.1, 139.8, 143.7, 149.8, 153.7, 154.5, 158.5, 162.5. MS: m/z 373

(MH) $^+$. HRMS calcd for $C_{17}H_{21}N_6O_4$: 373.1624 [M+H], found: 373.1628.

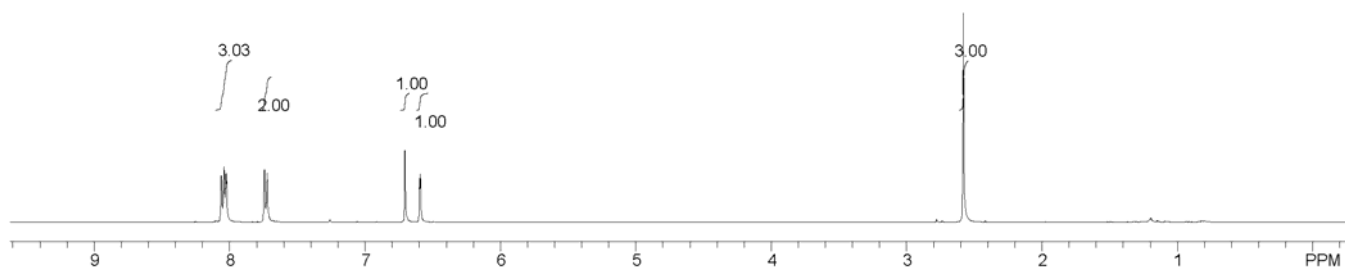
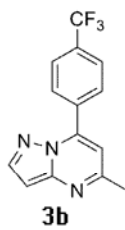
III. Copies of ^1H and ^{13}C NMR spectra of 3a-3ll, 4ii-4ll, 7, 8, 11, 12



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6.592

2.581



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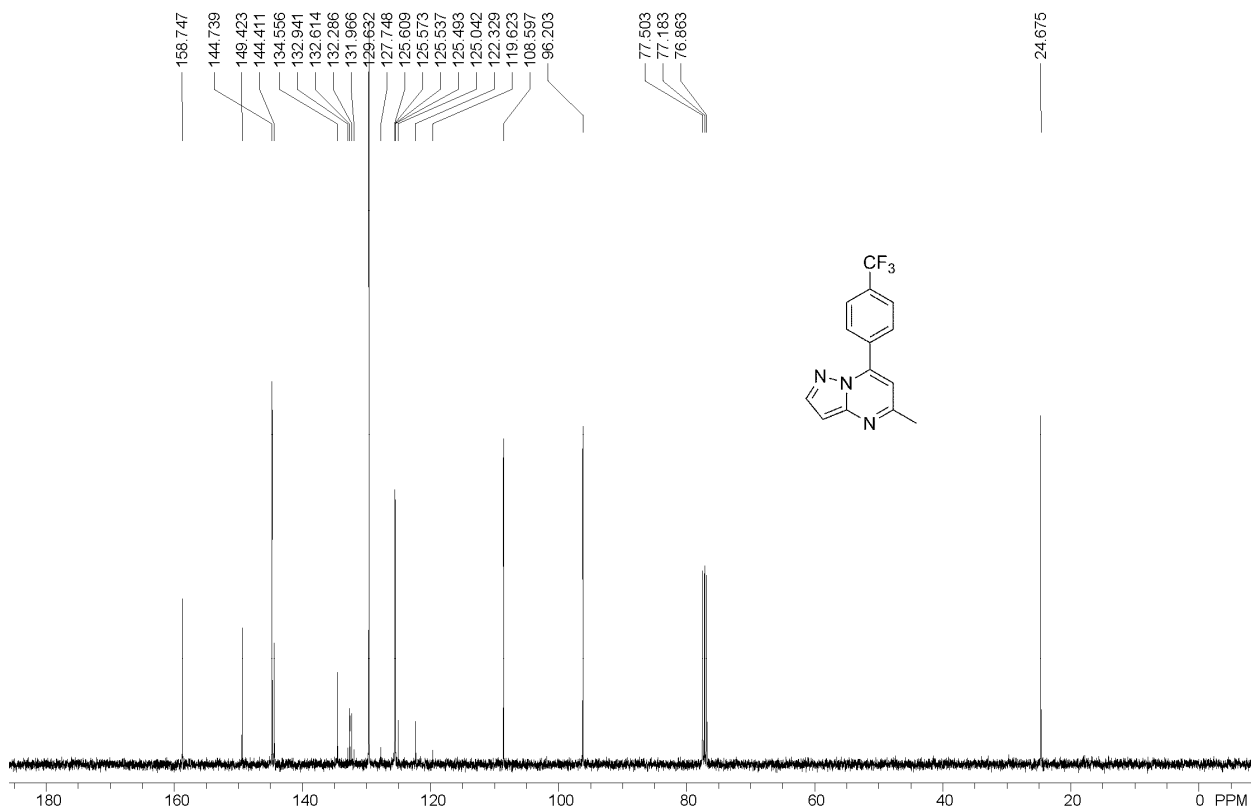
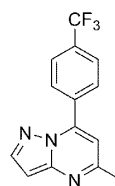
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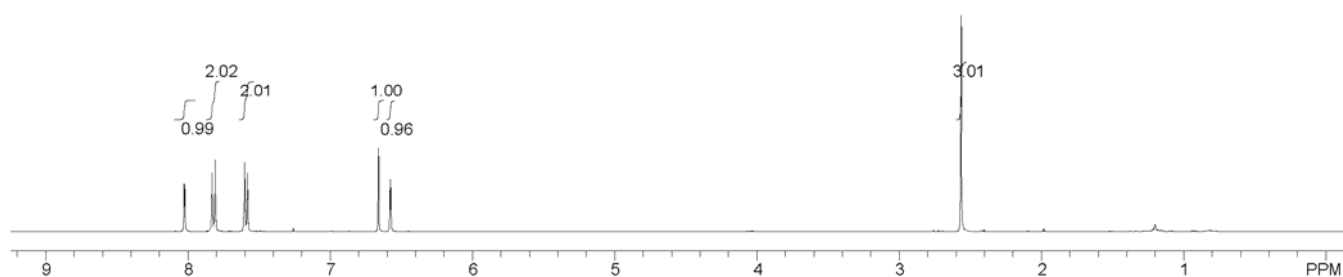
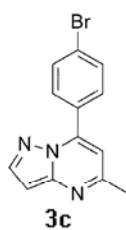
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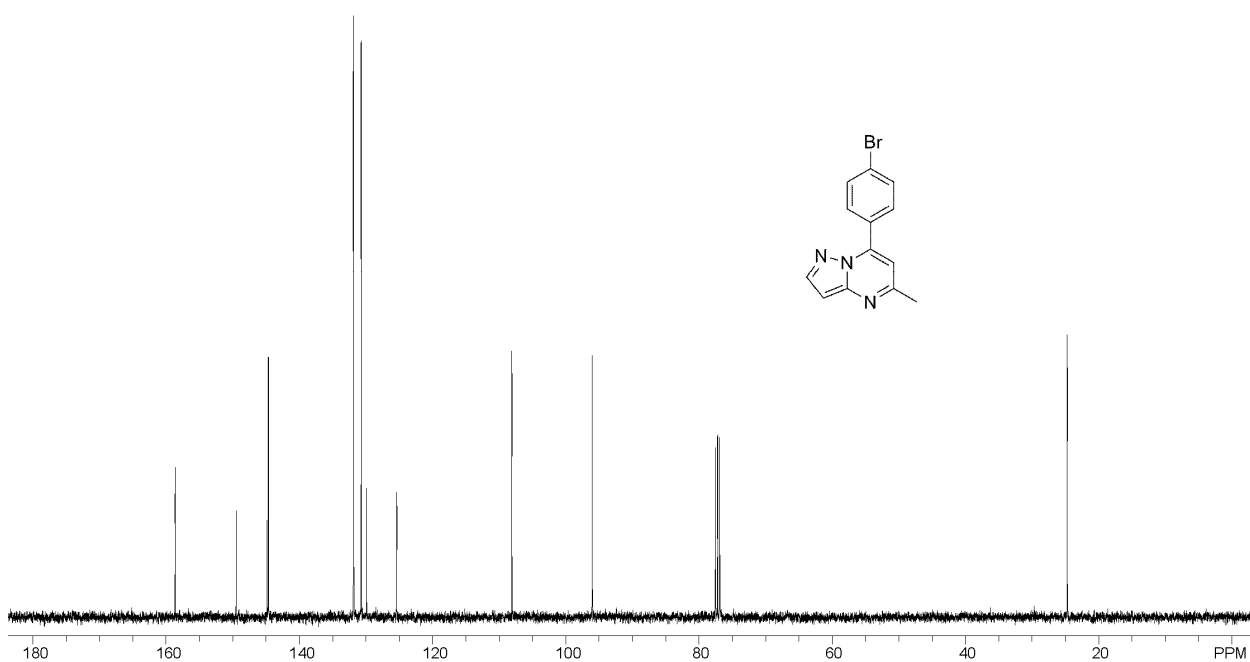
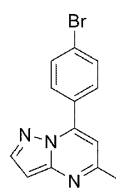
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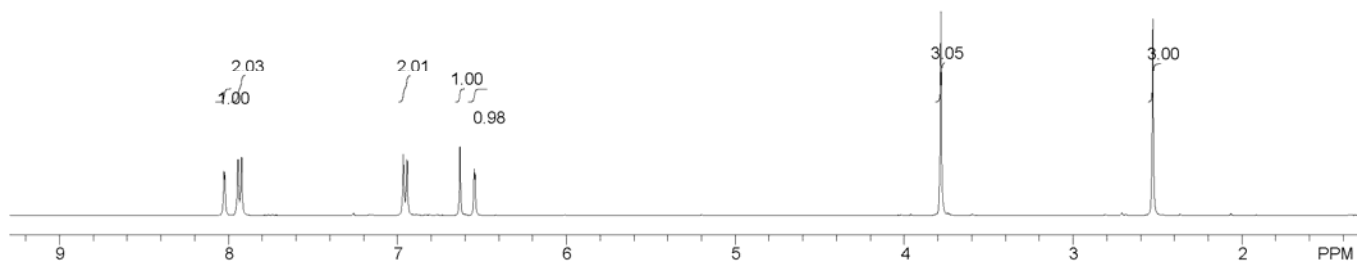
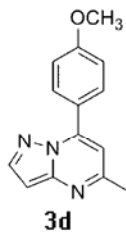


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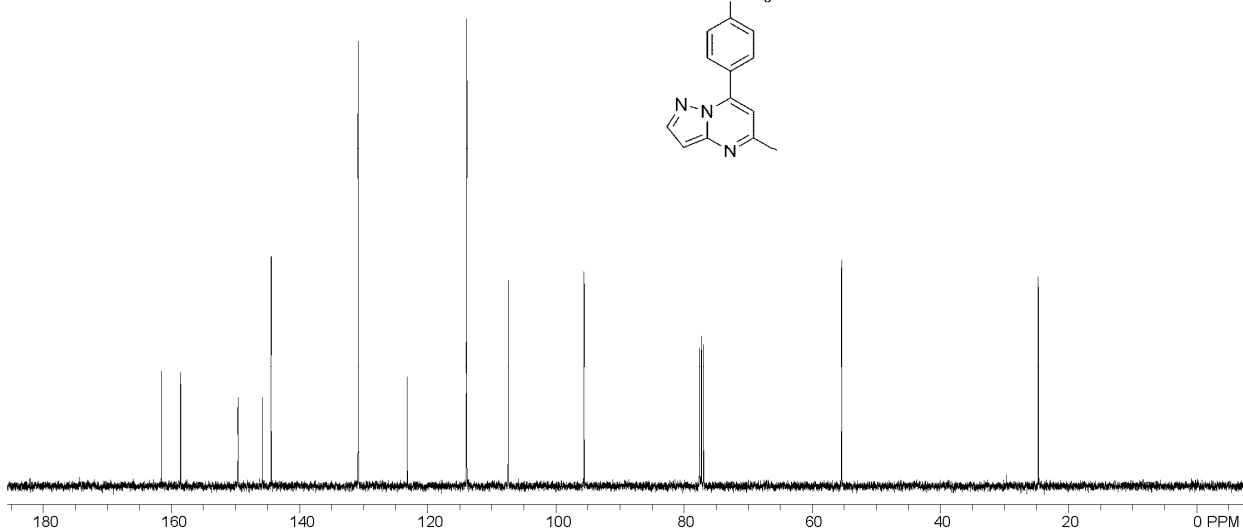
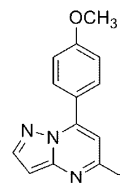
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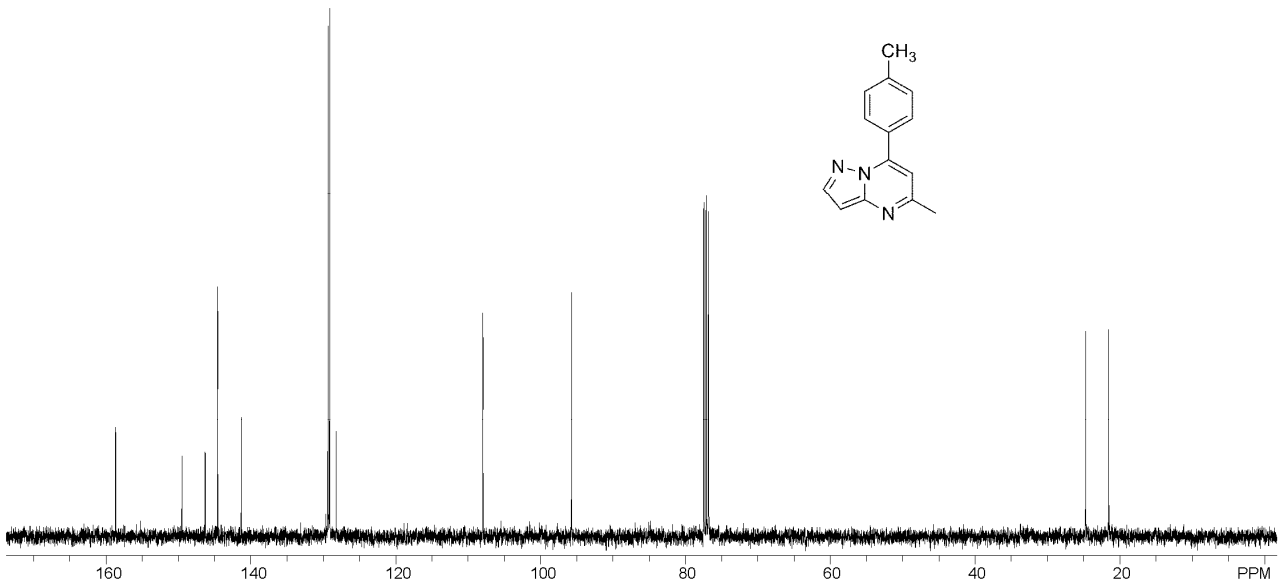
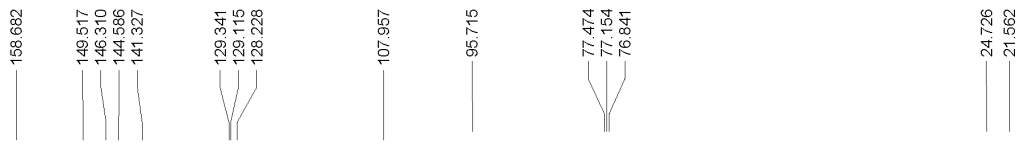
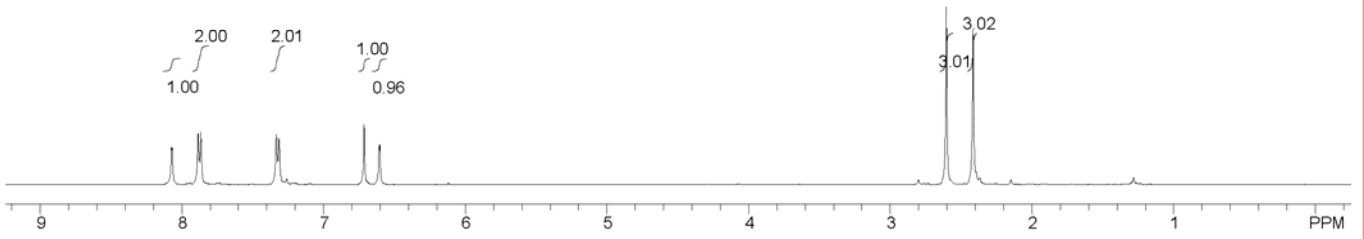
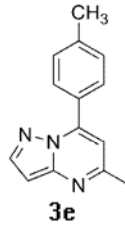
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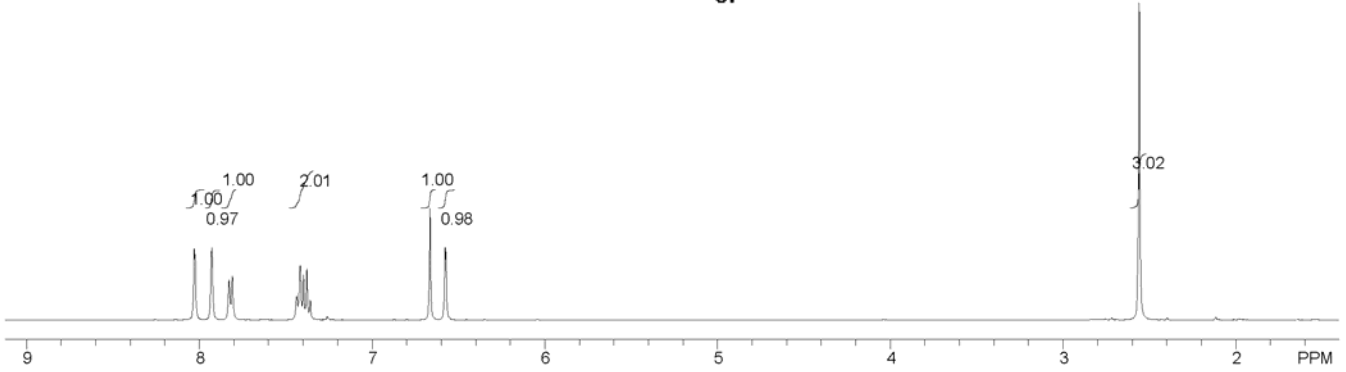
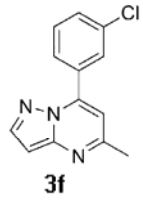
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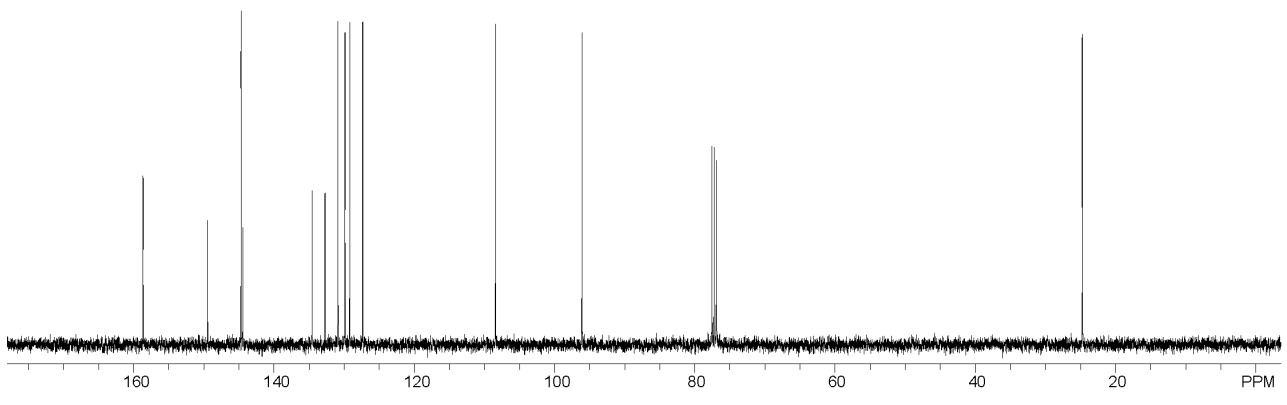
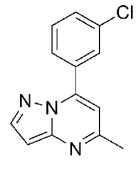


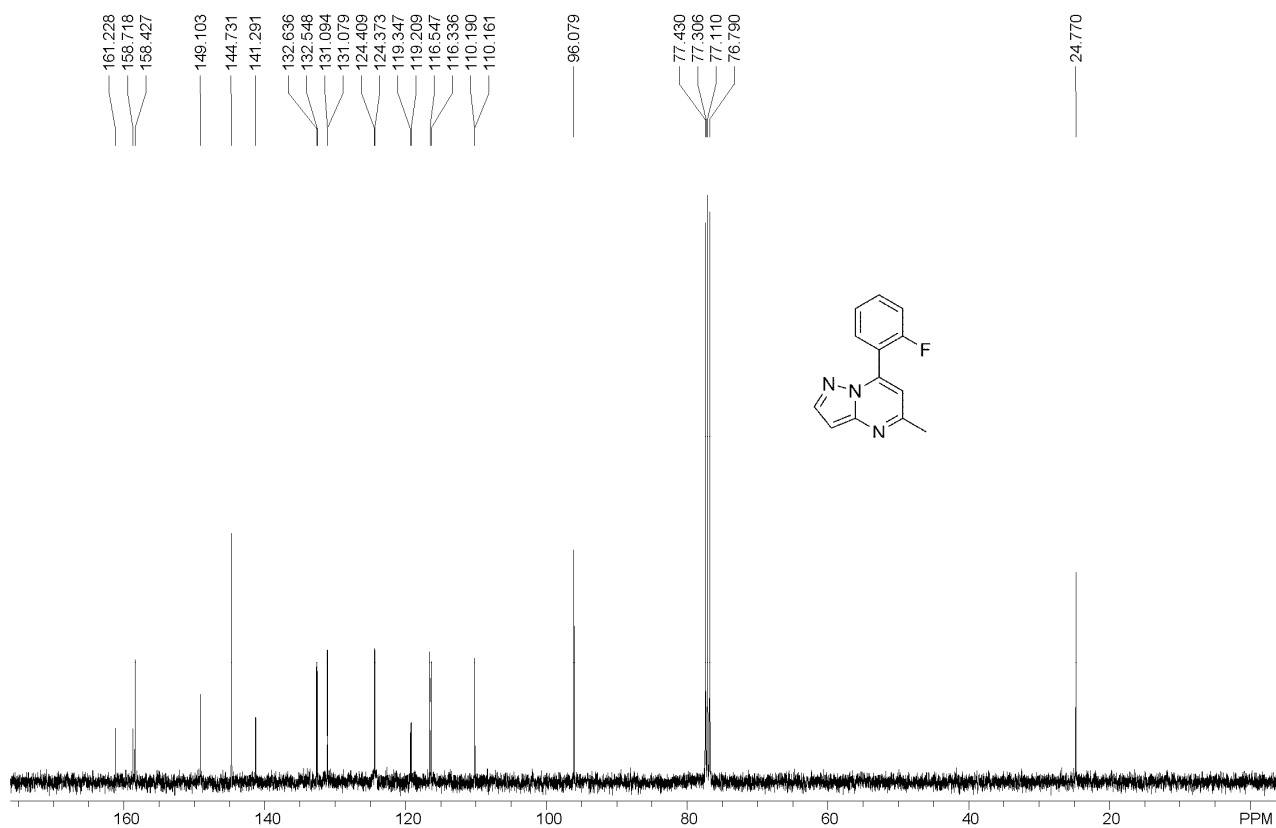
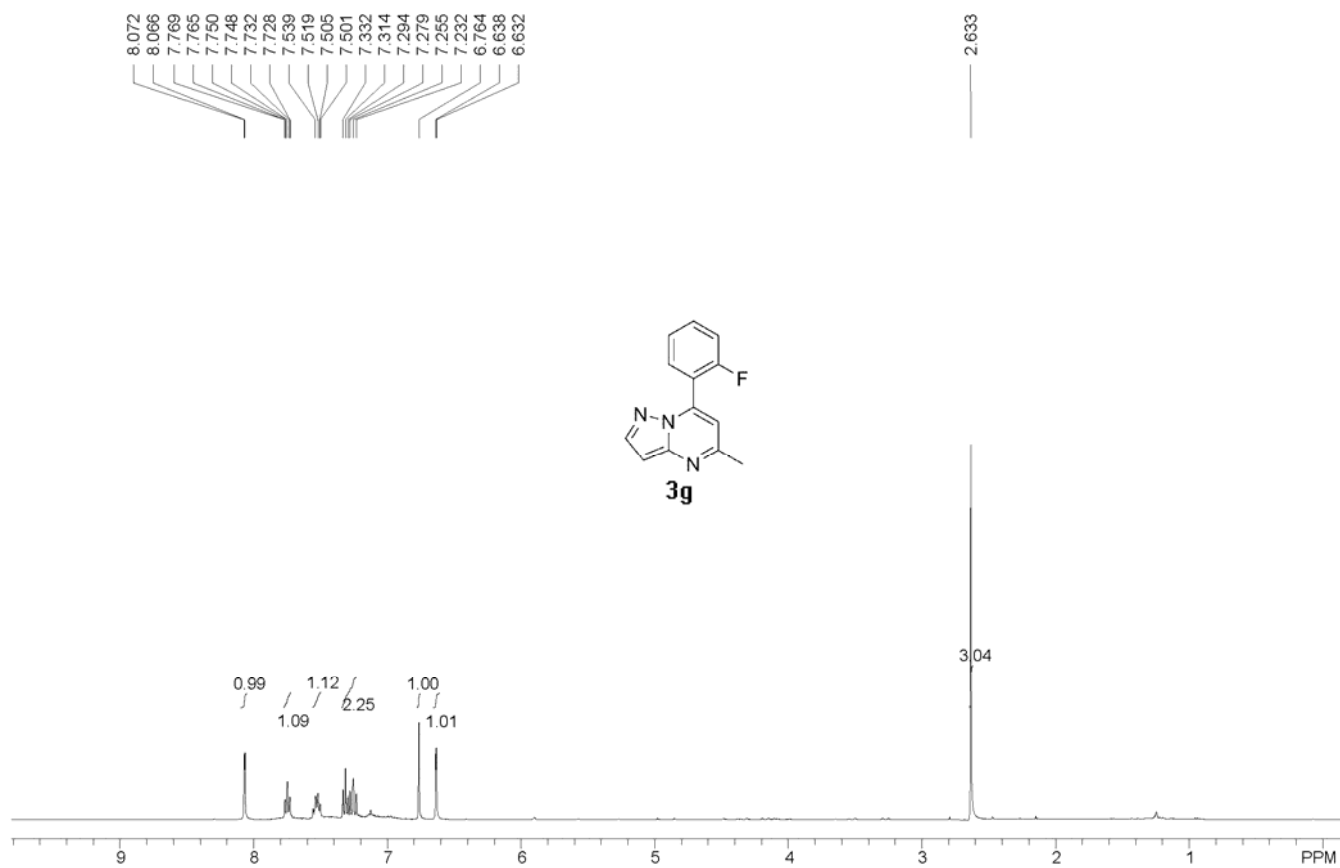
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2.559



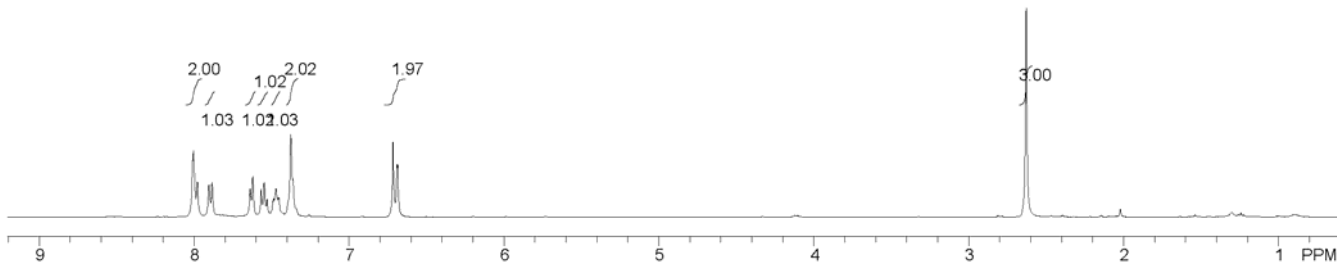
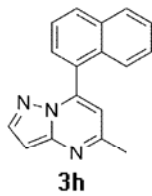
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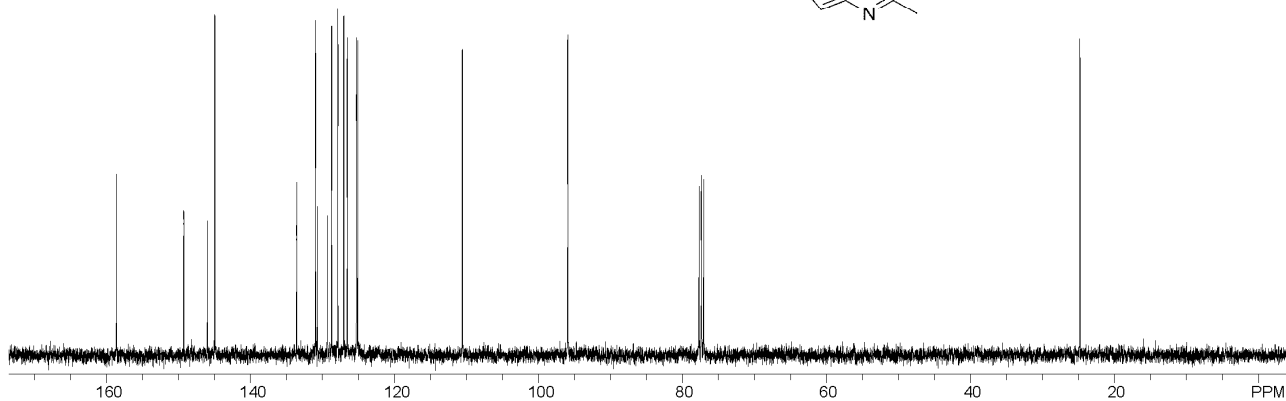
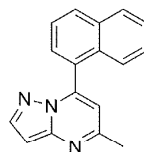


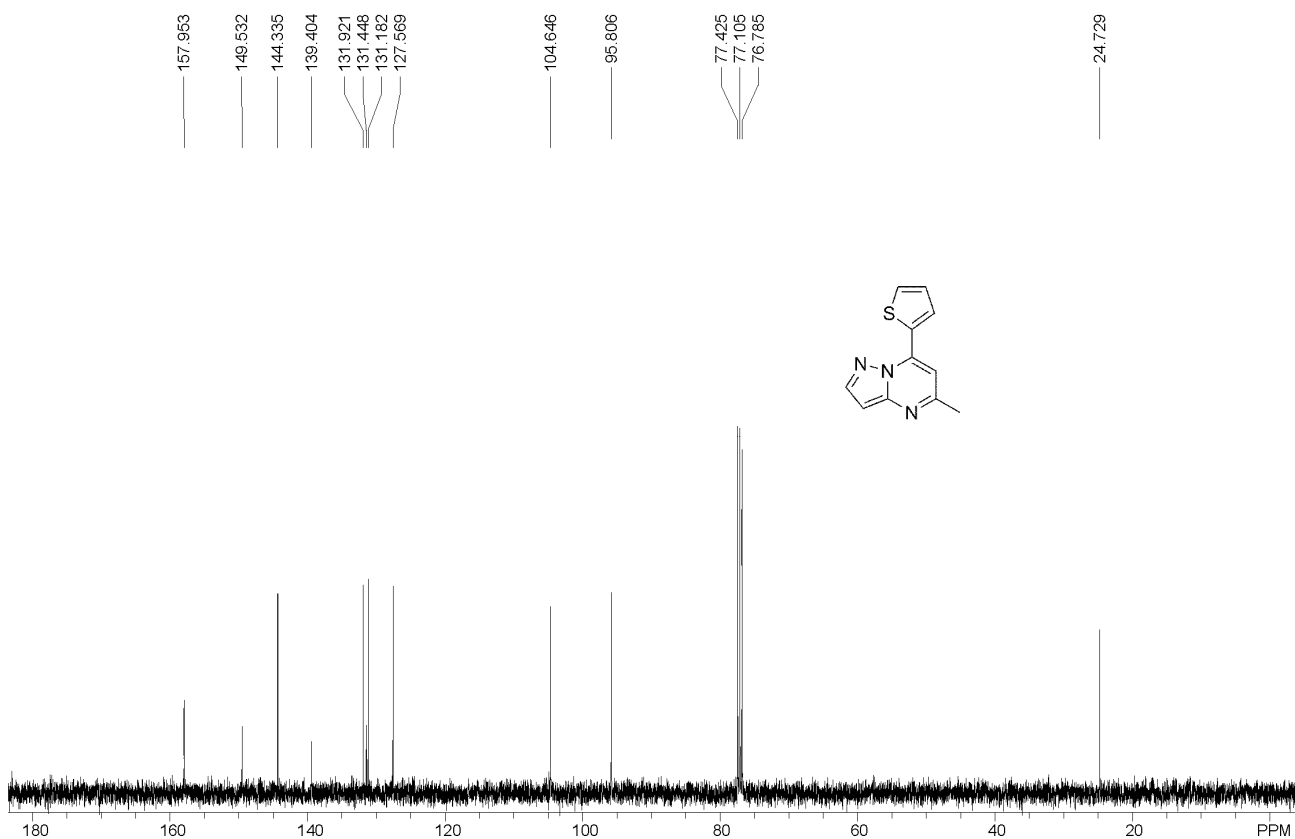
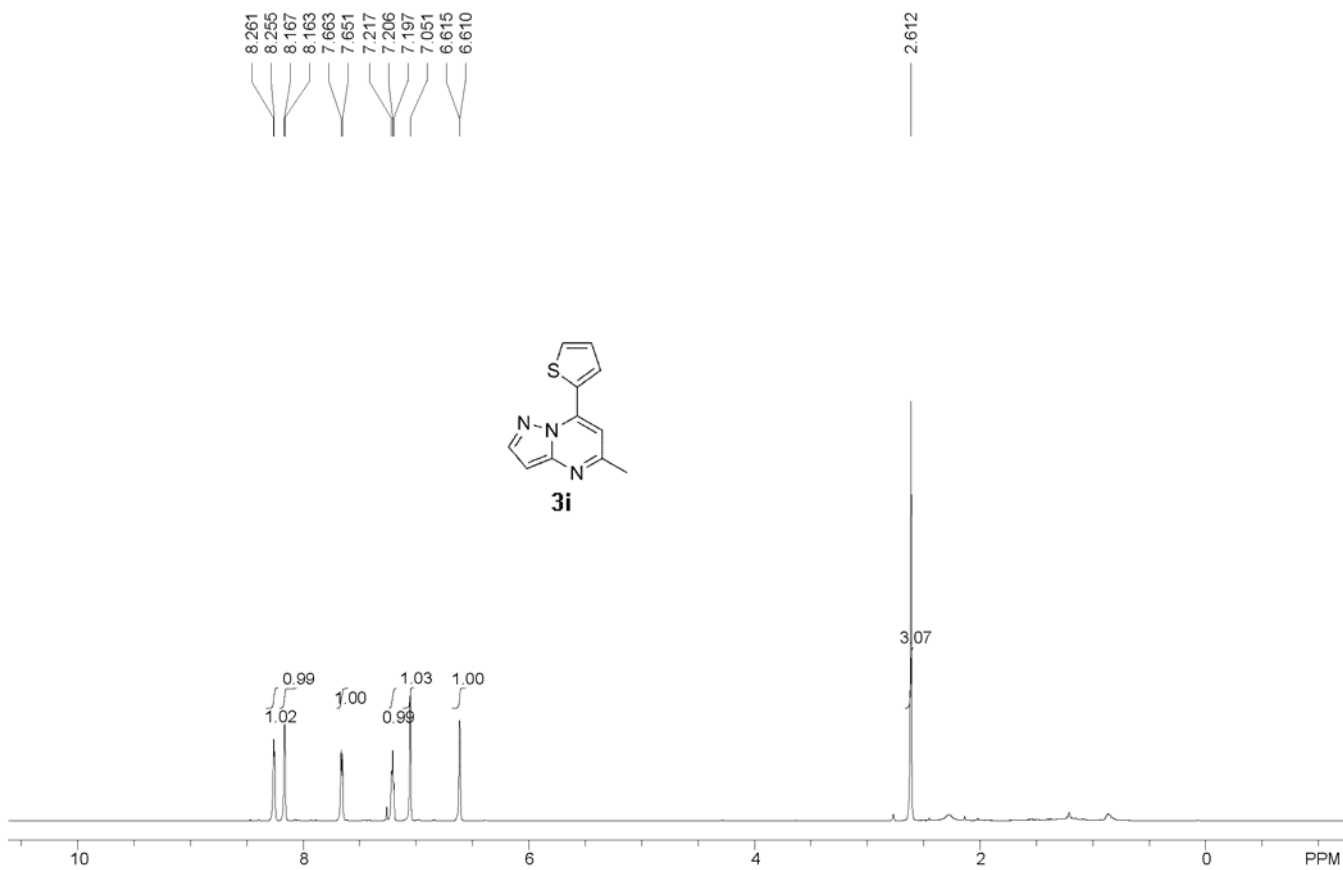
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125.100
110.561

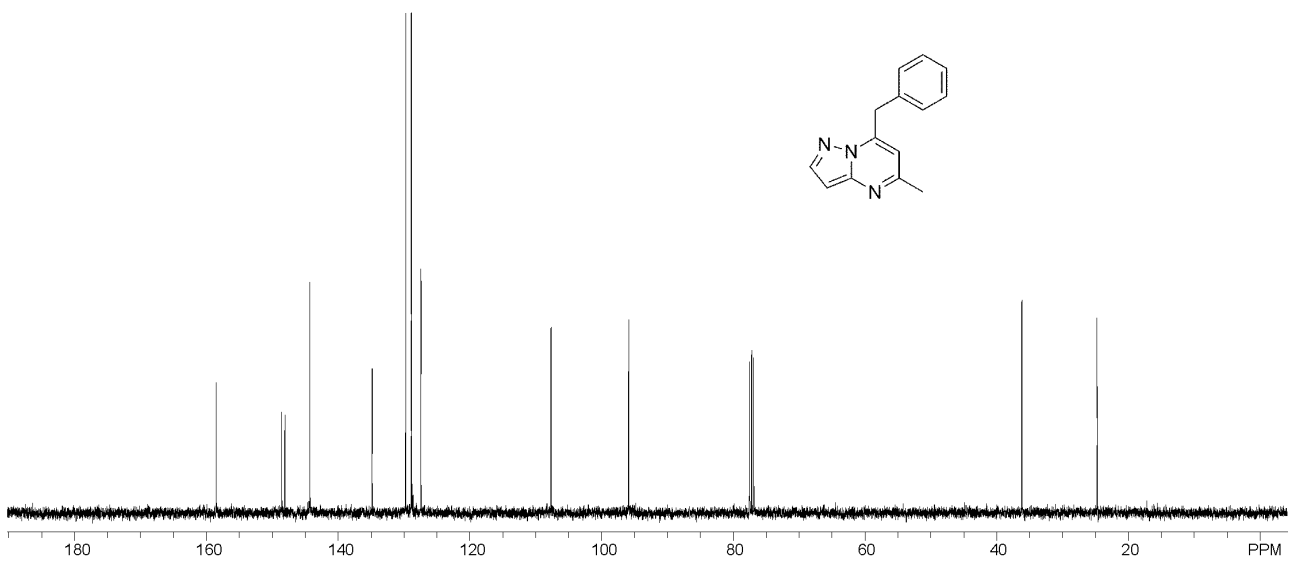
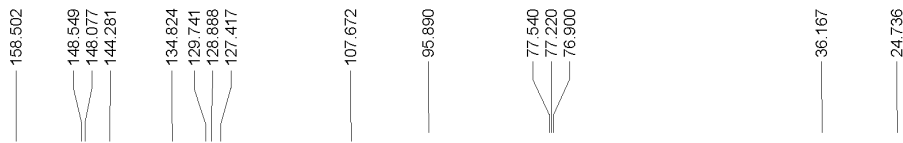
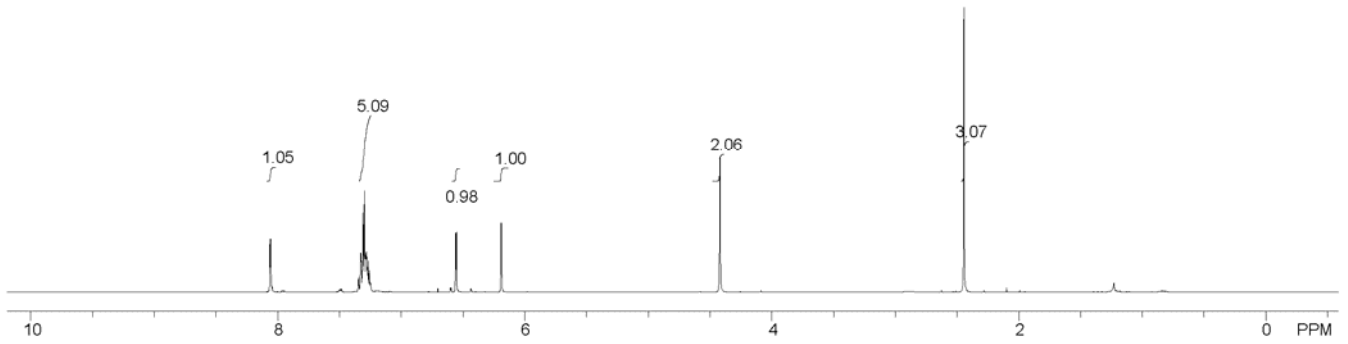
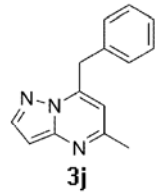
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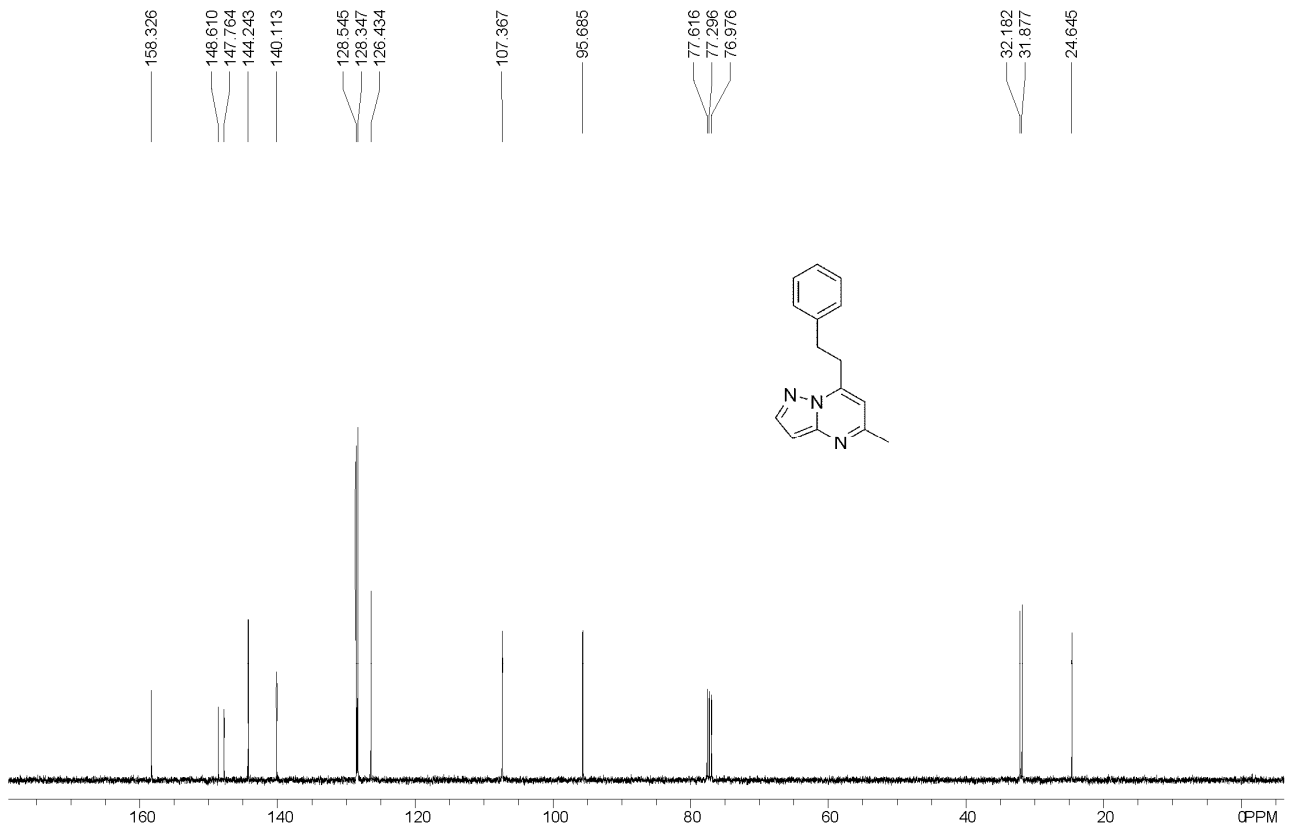
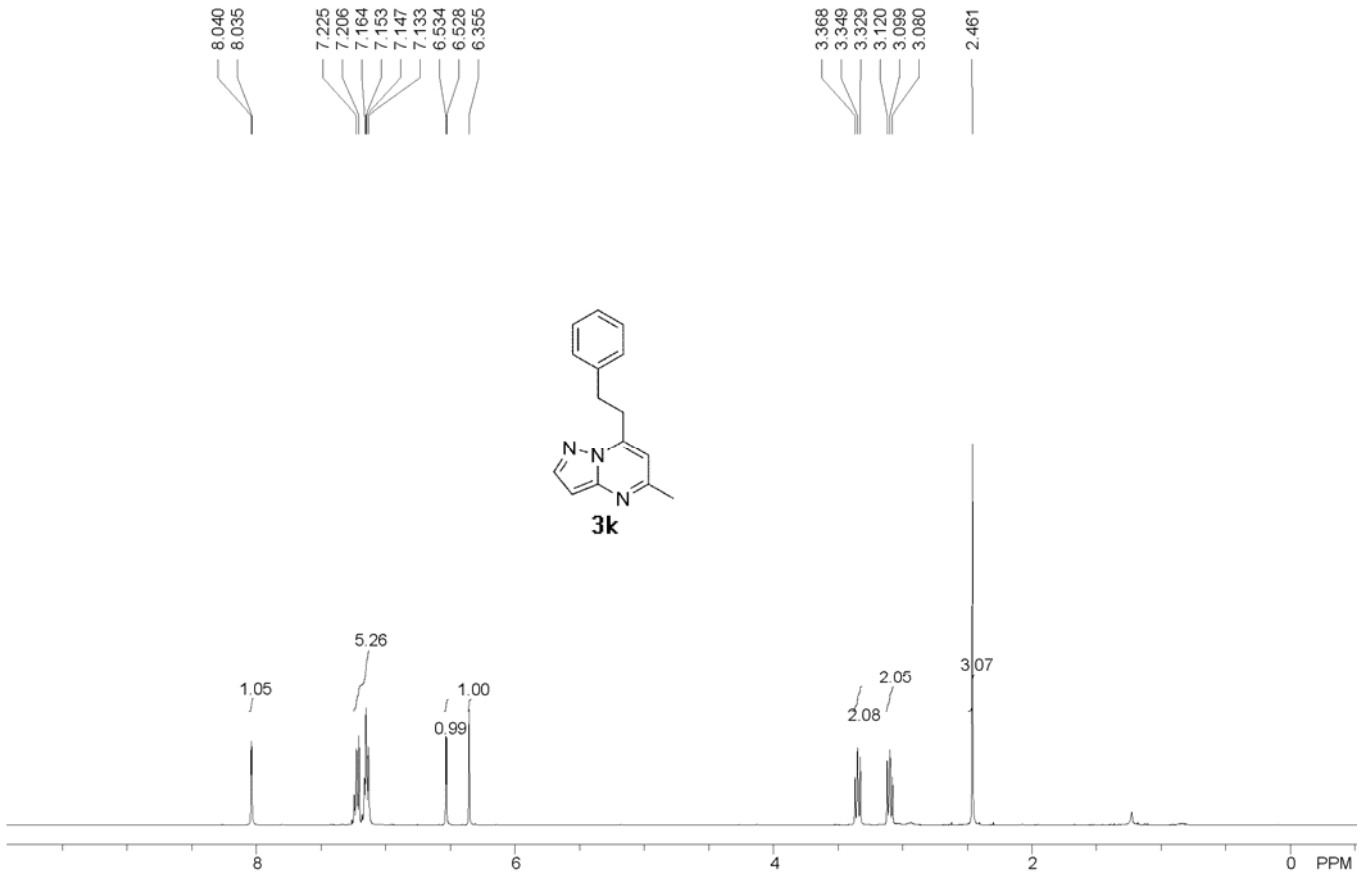
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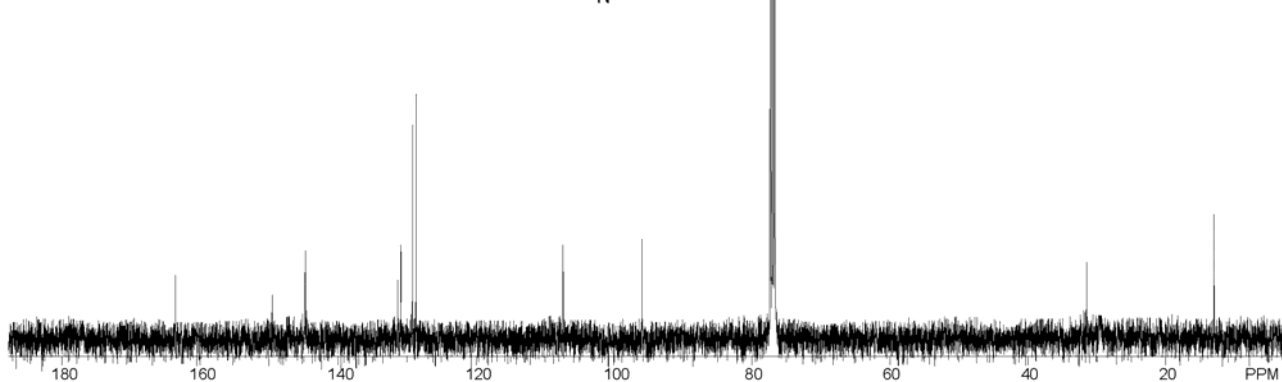
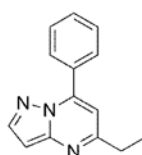
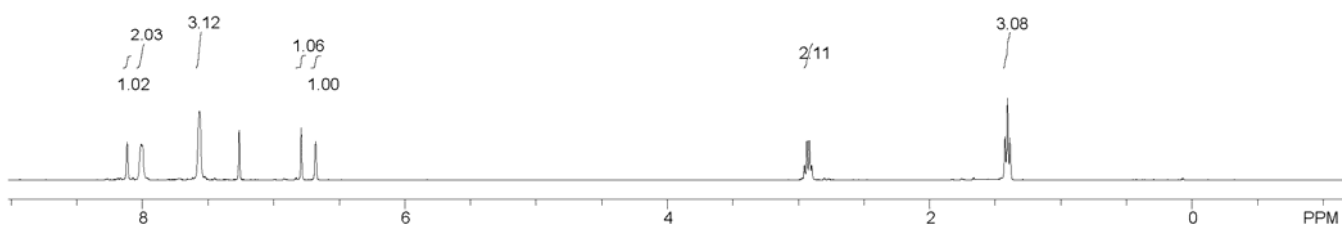
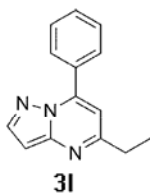
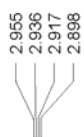
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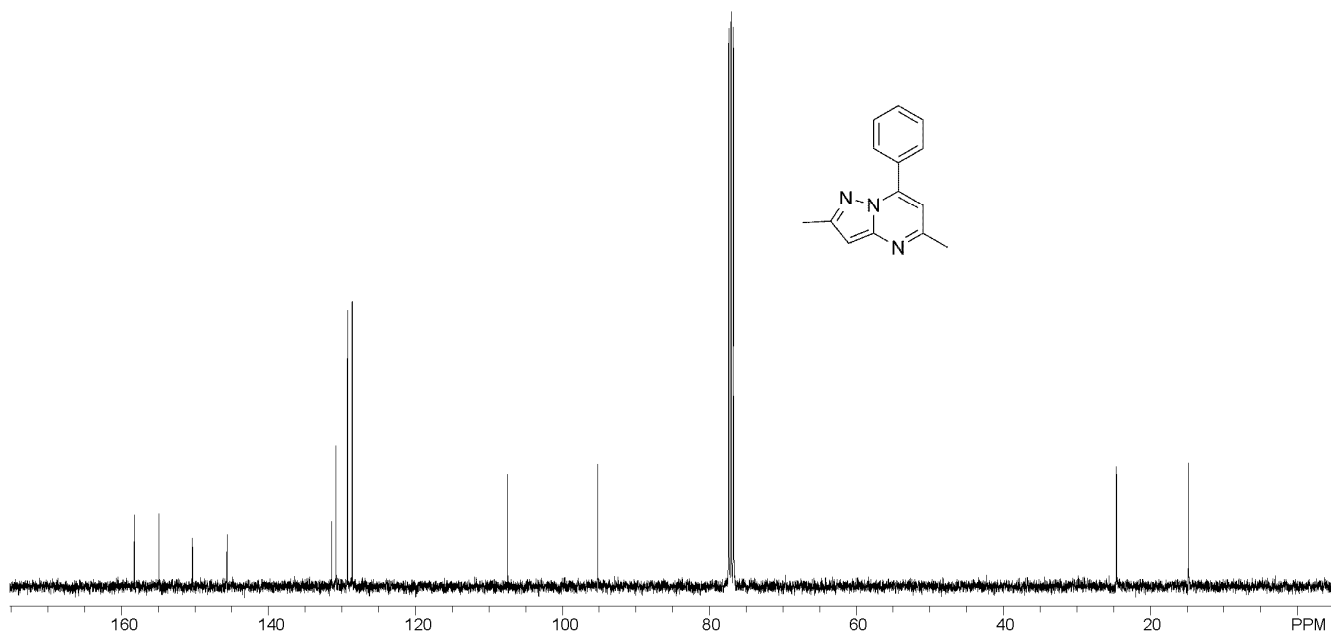
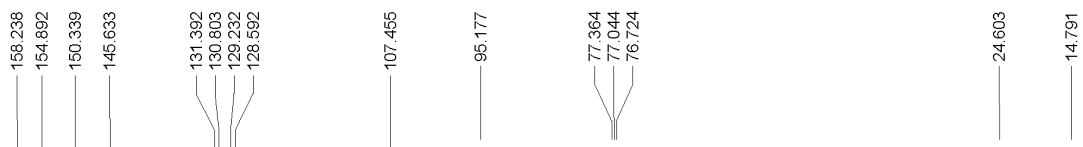
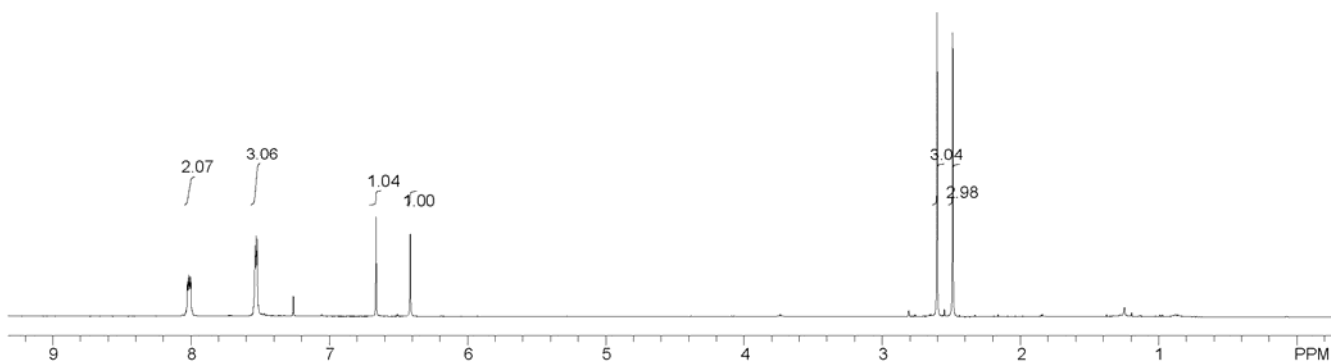
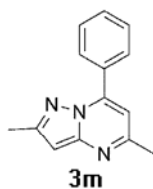






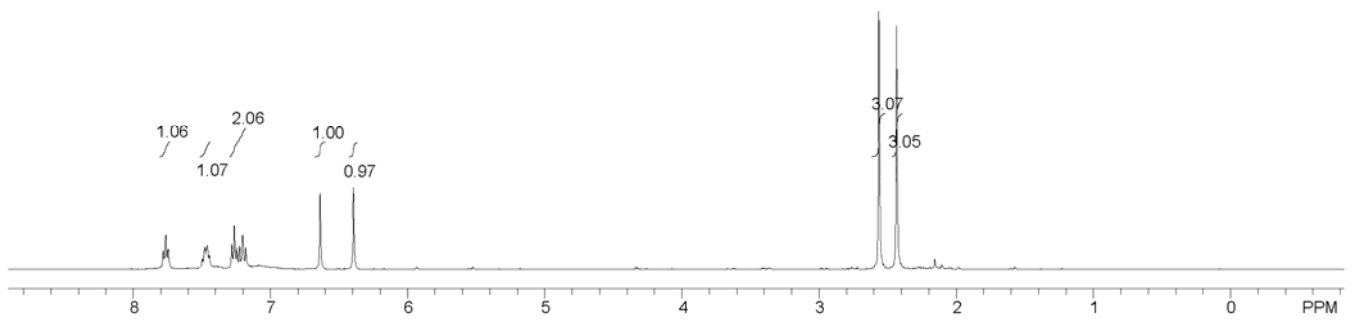
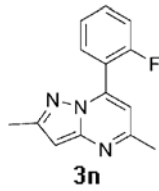






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2.565
2.435



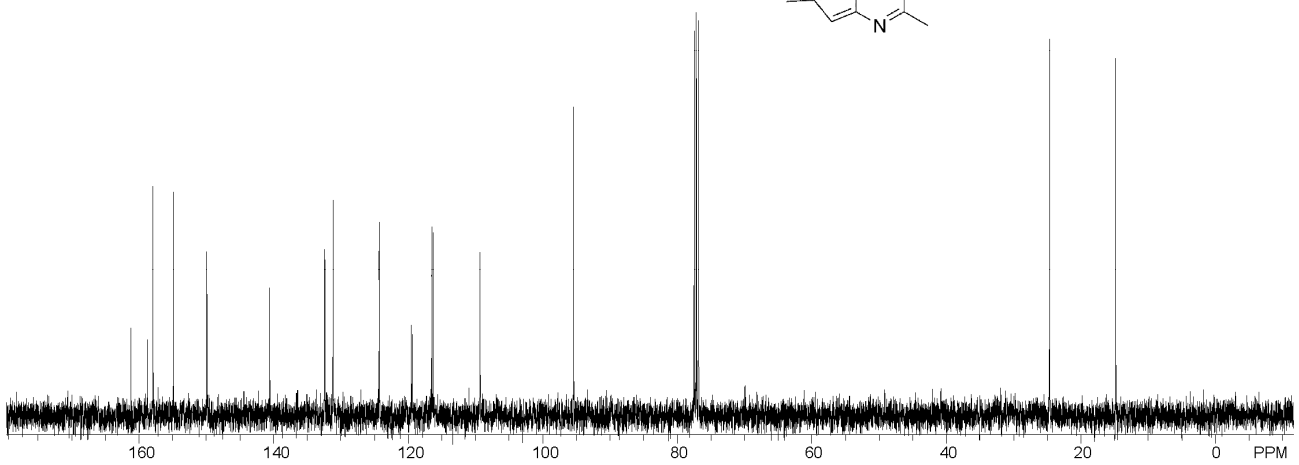
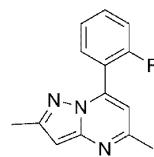
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119.376
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116.278
109.331
109.310

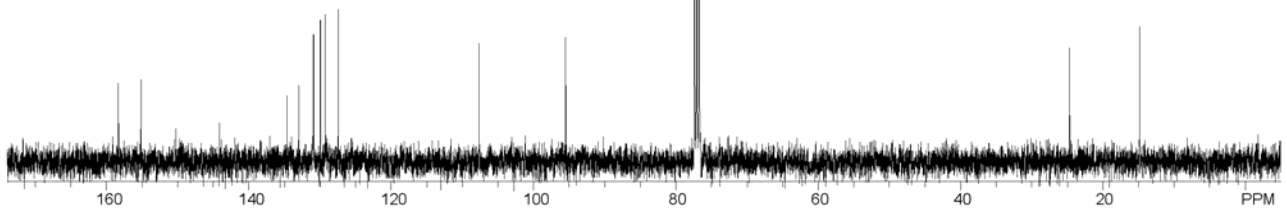
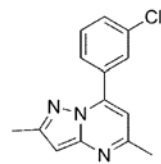
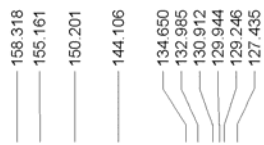
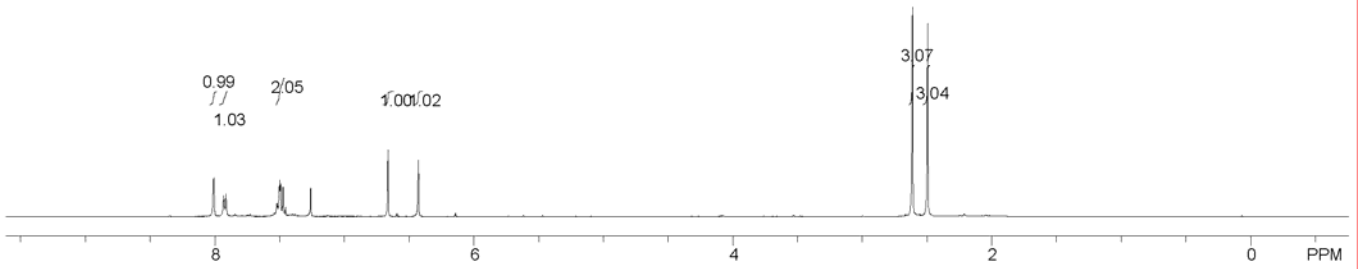
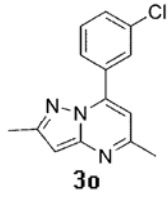
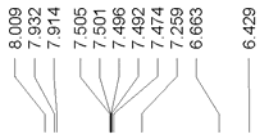
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24.661

14.834

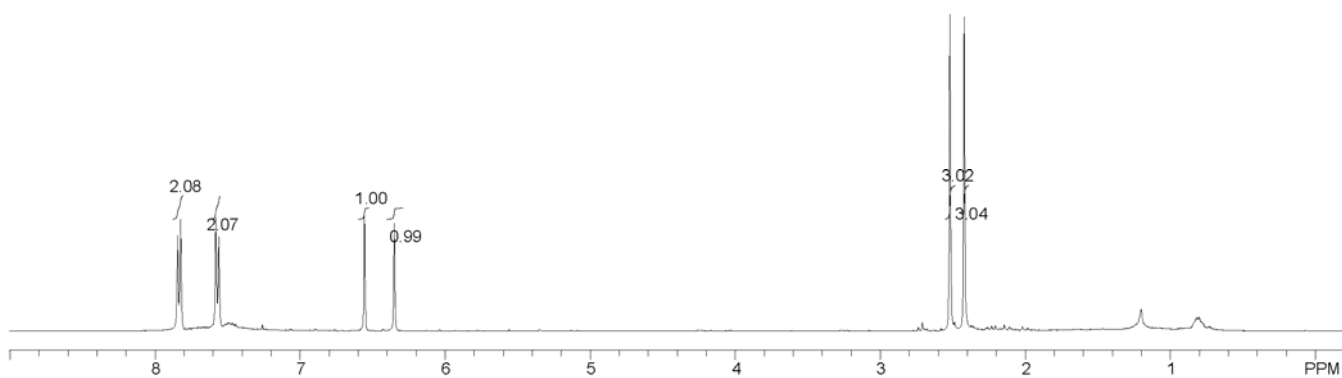
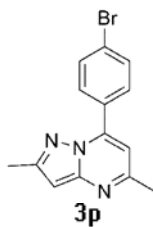




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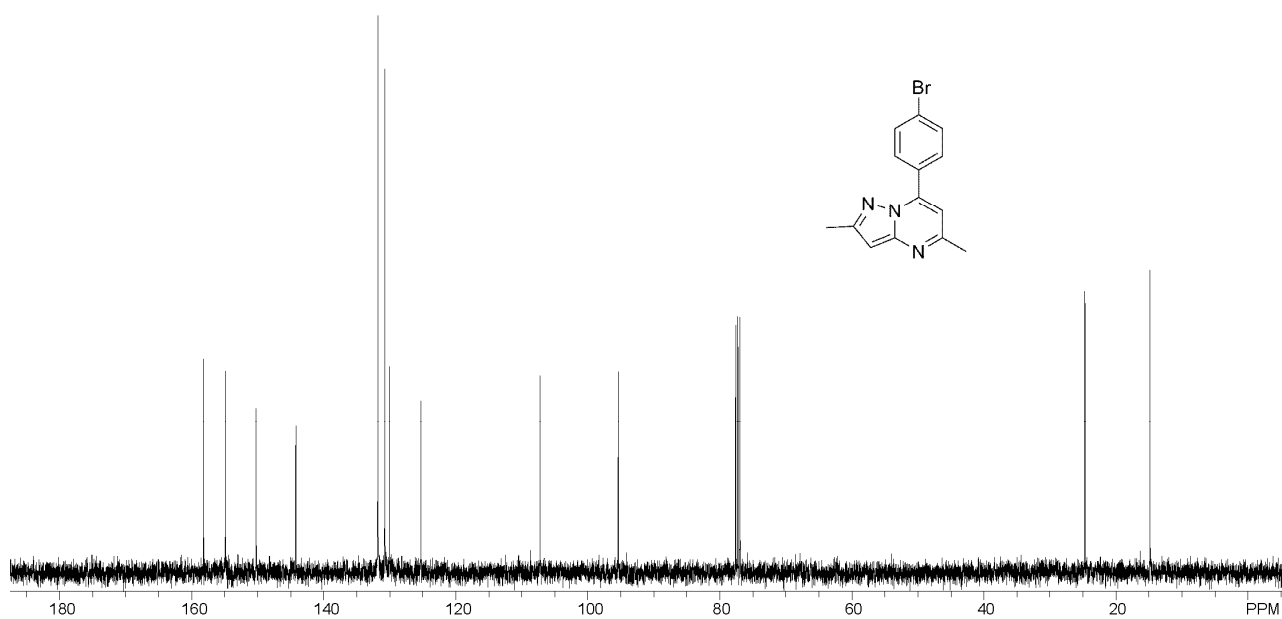
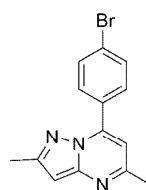
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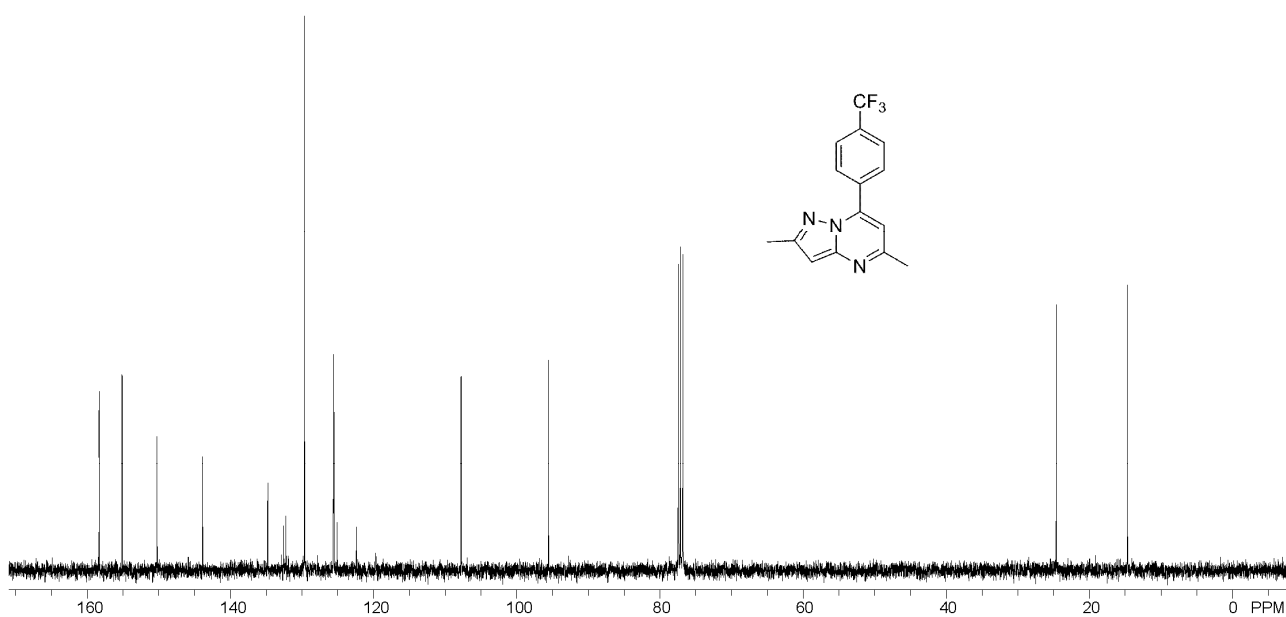
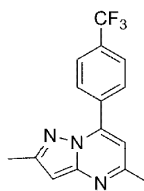
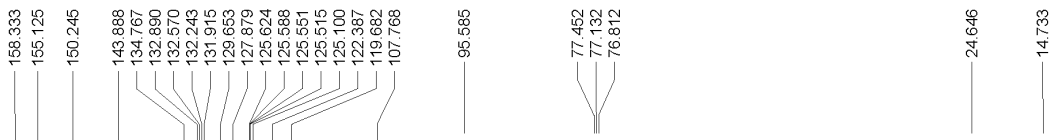
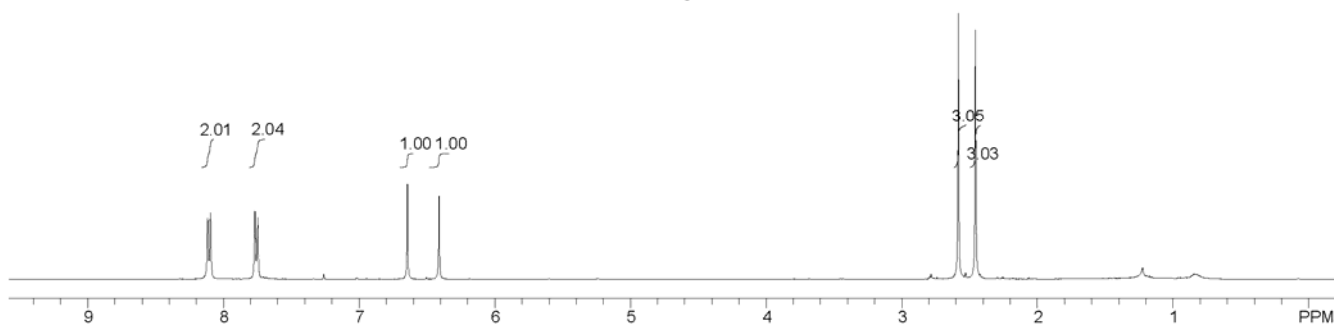
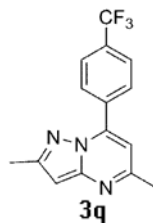
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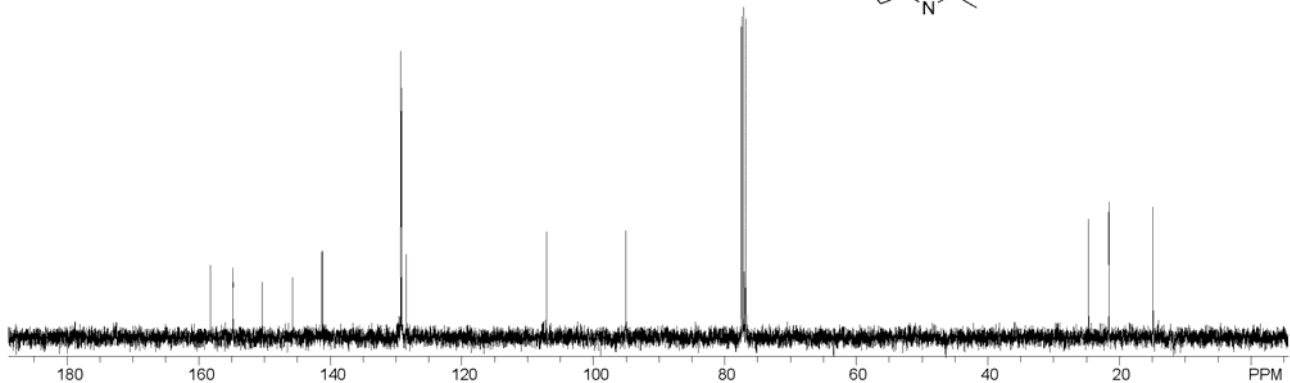
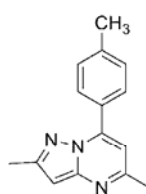
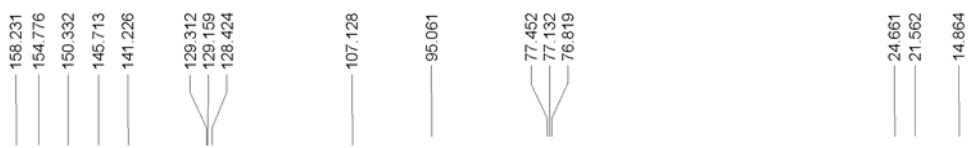
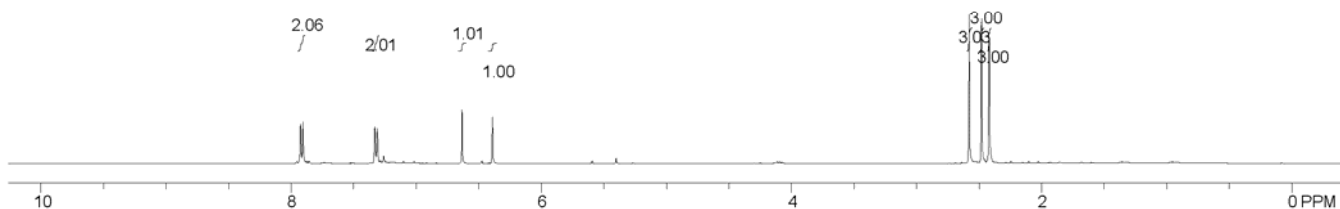
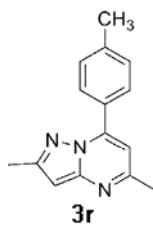
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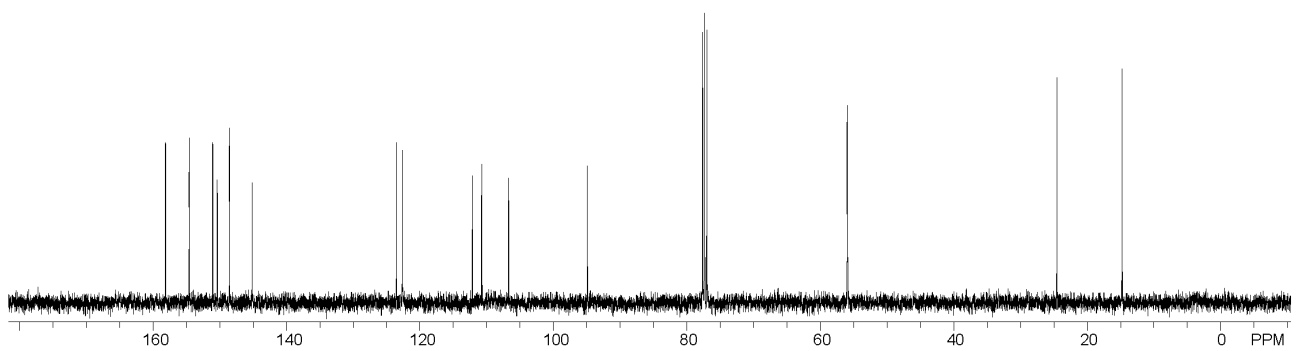
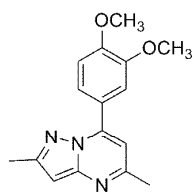
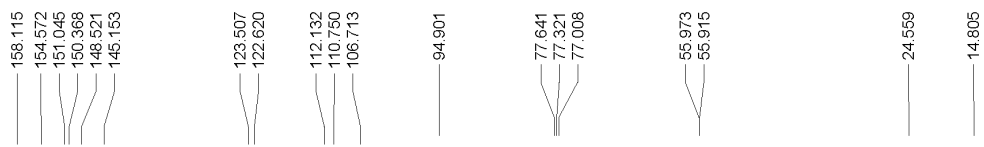
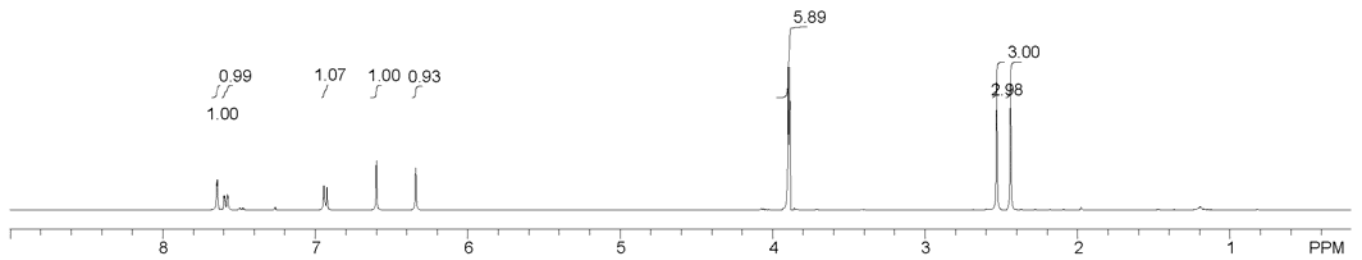
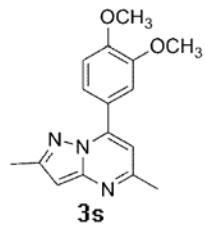
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14.820



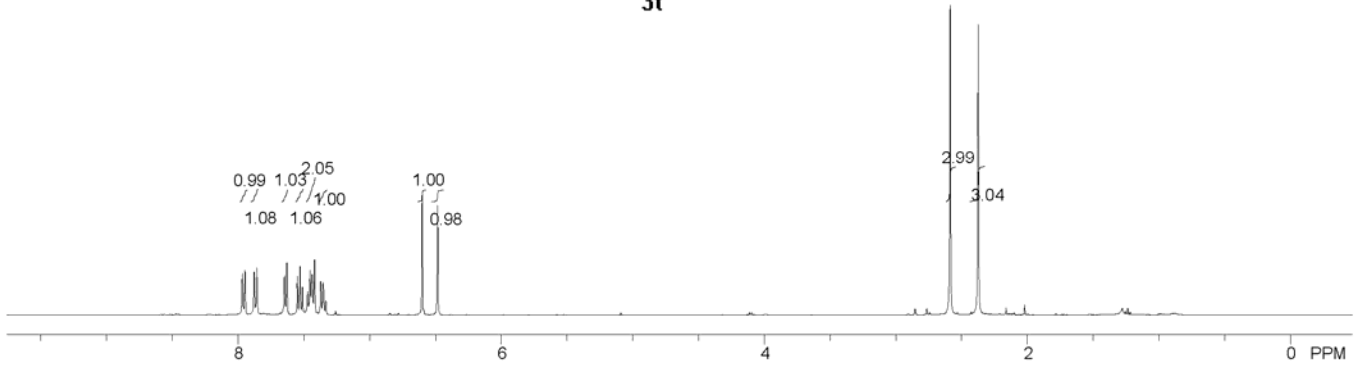
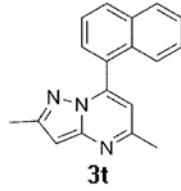






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2.373



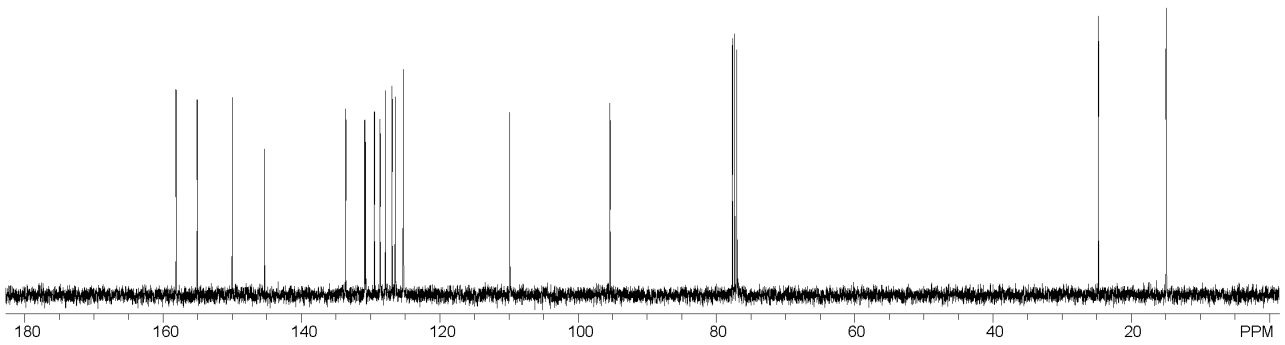
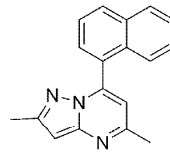
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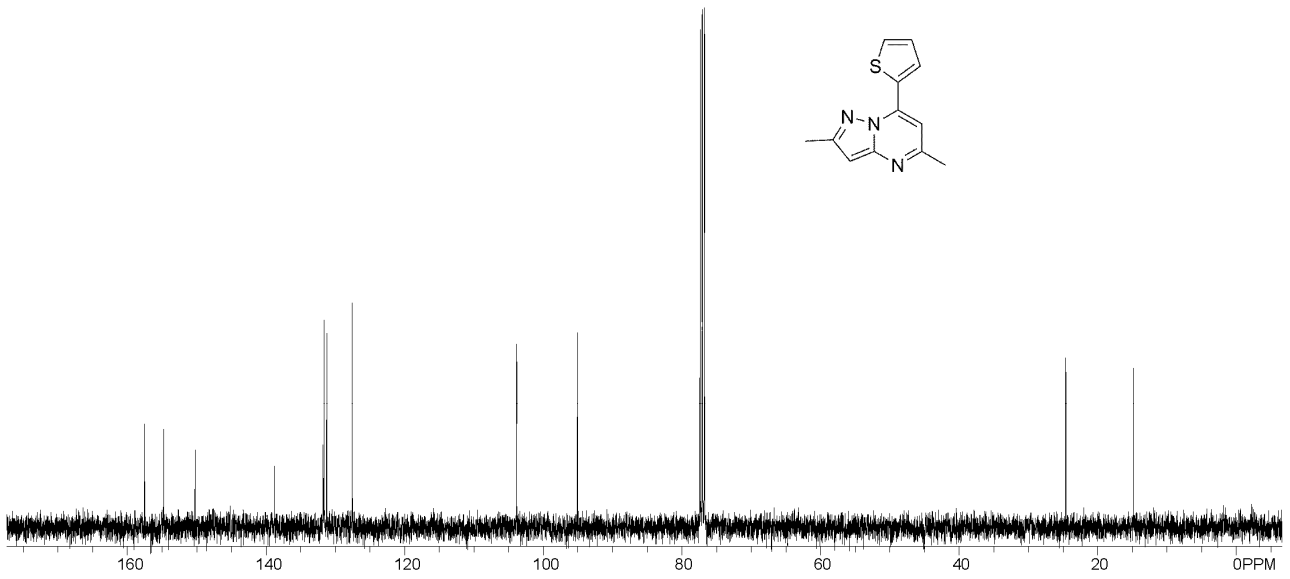
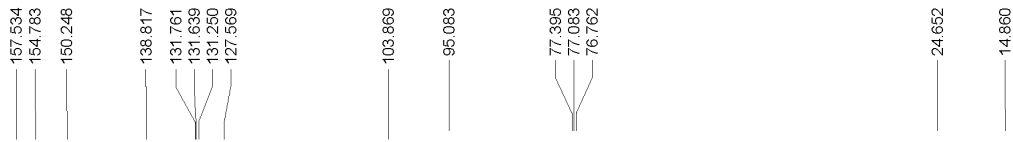
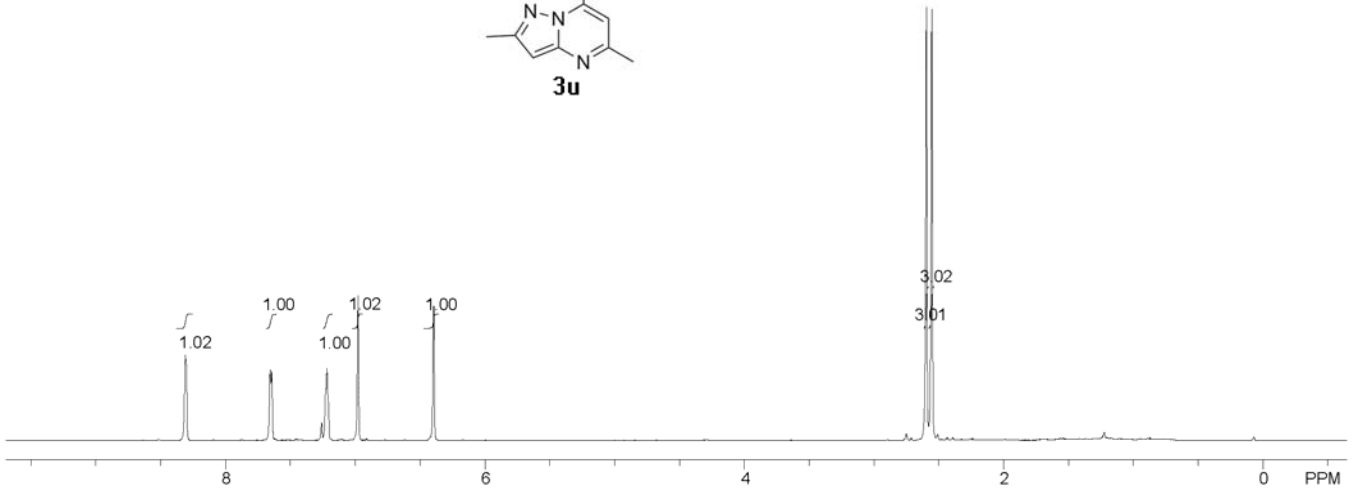
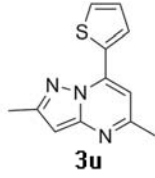
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24.741

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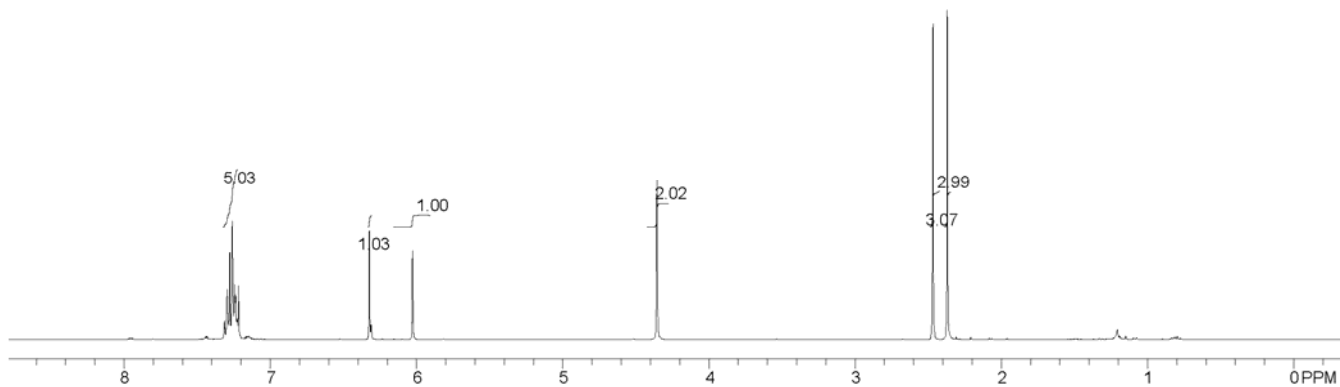
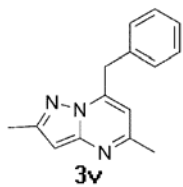


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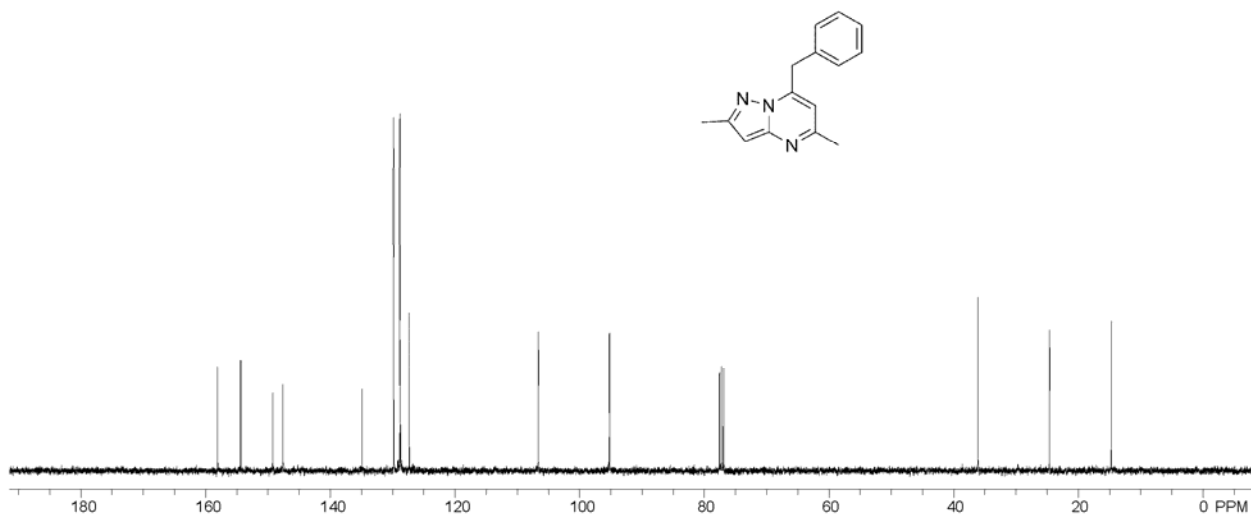
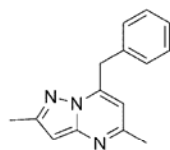
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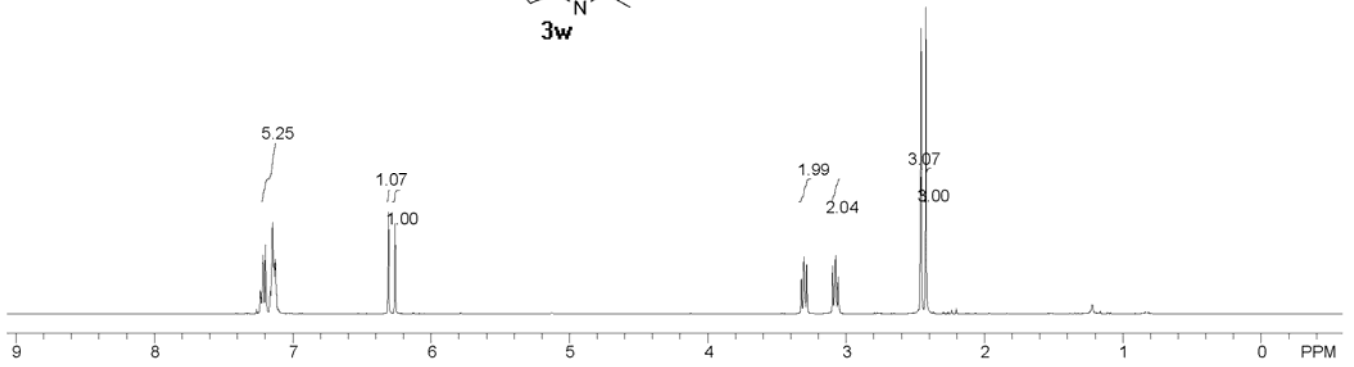
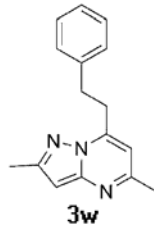
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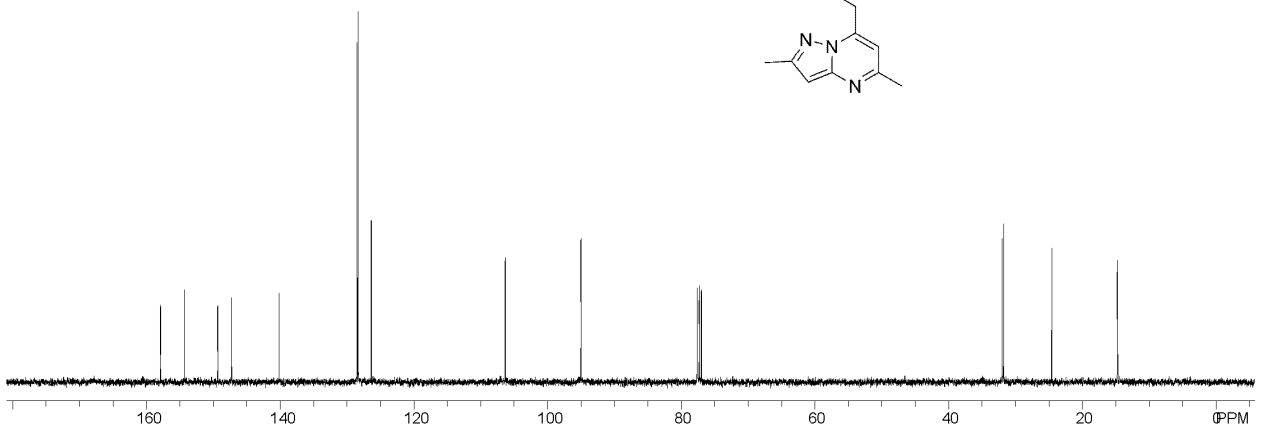
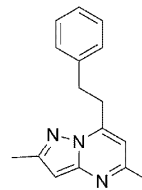
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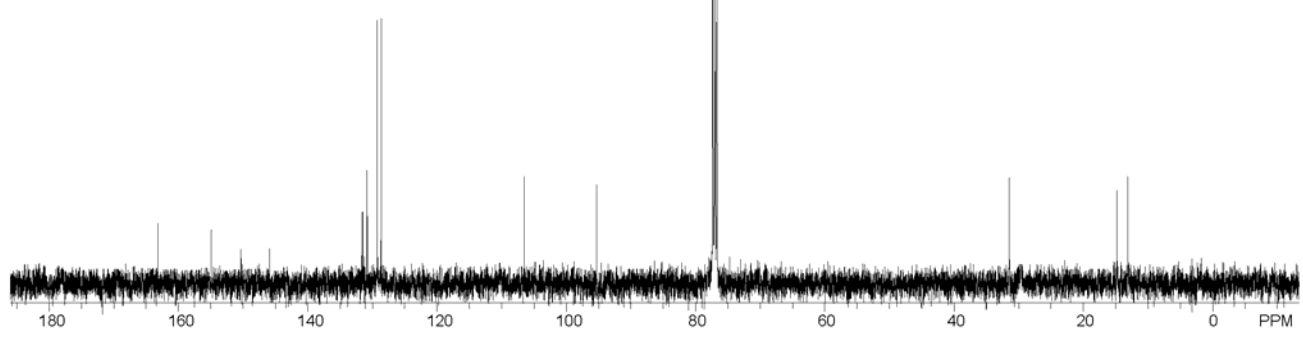
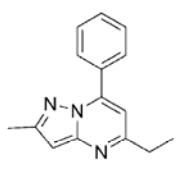
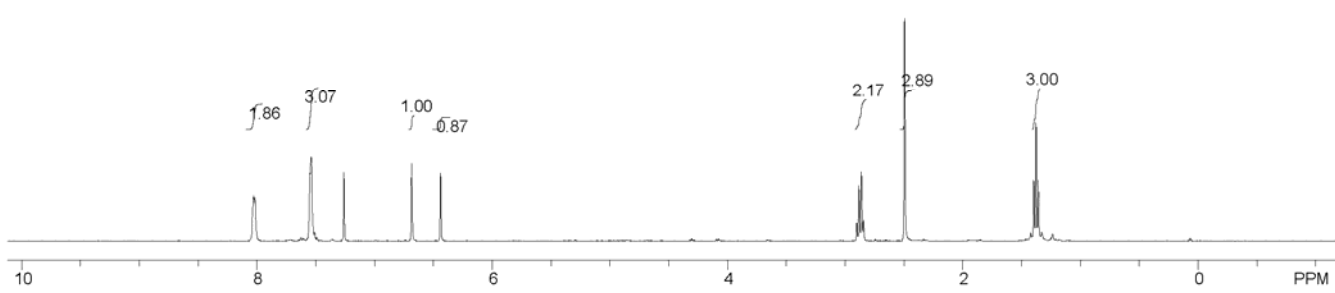
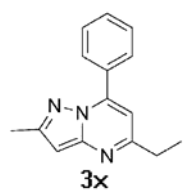
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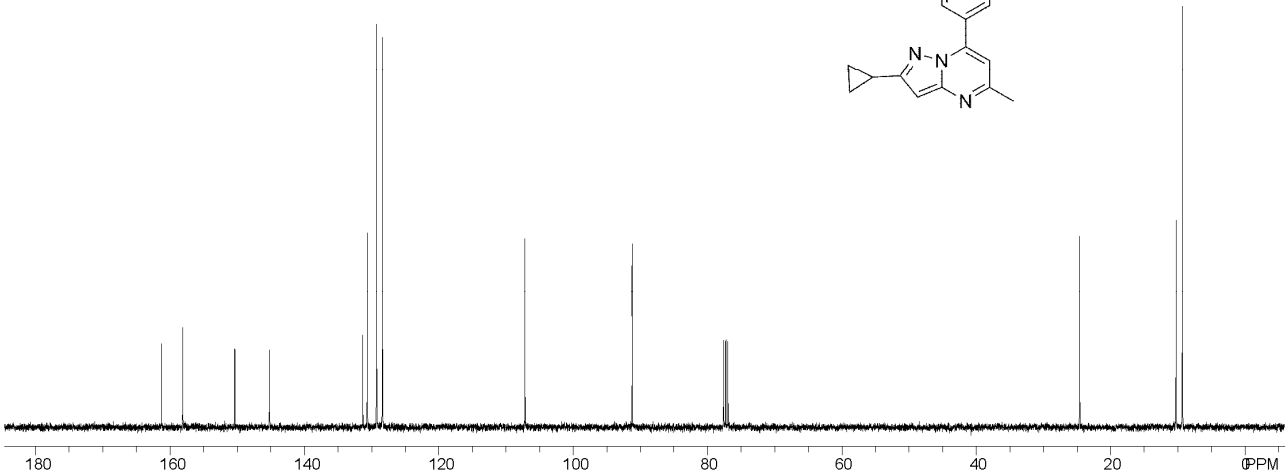
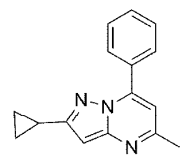
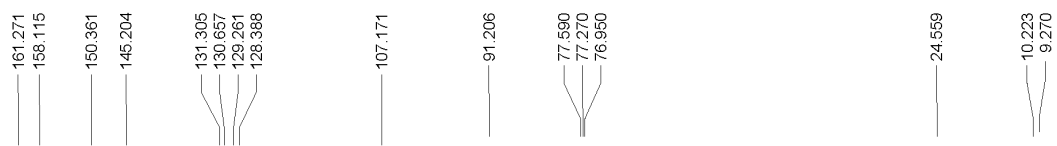
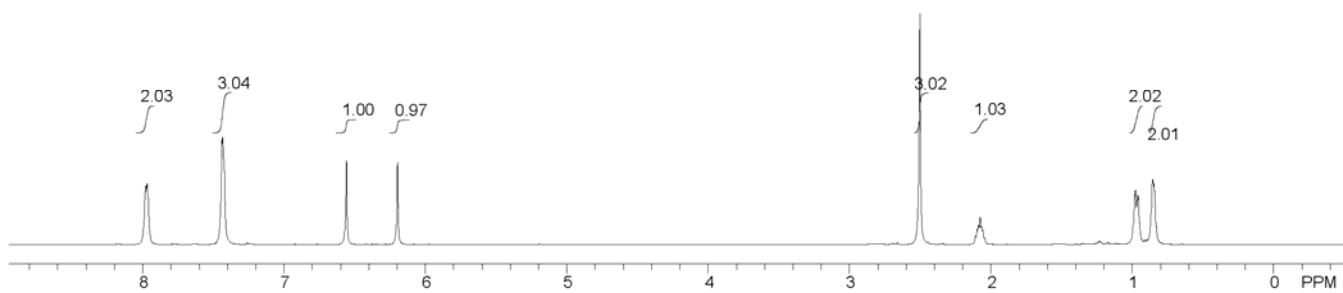
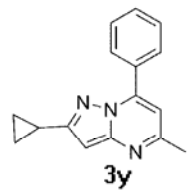
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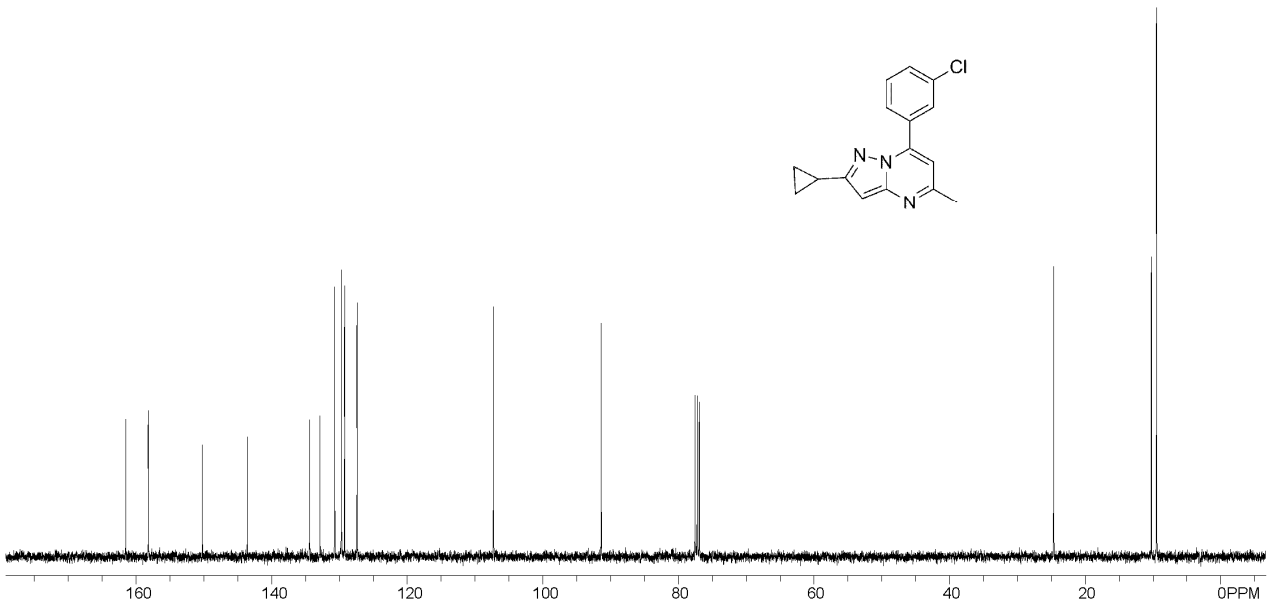
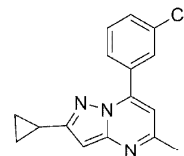
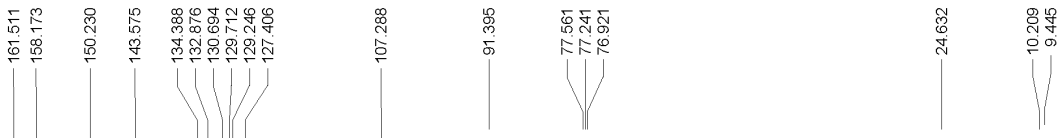
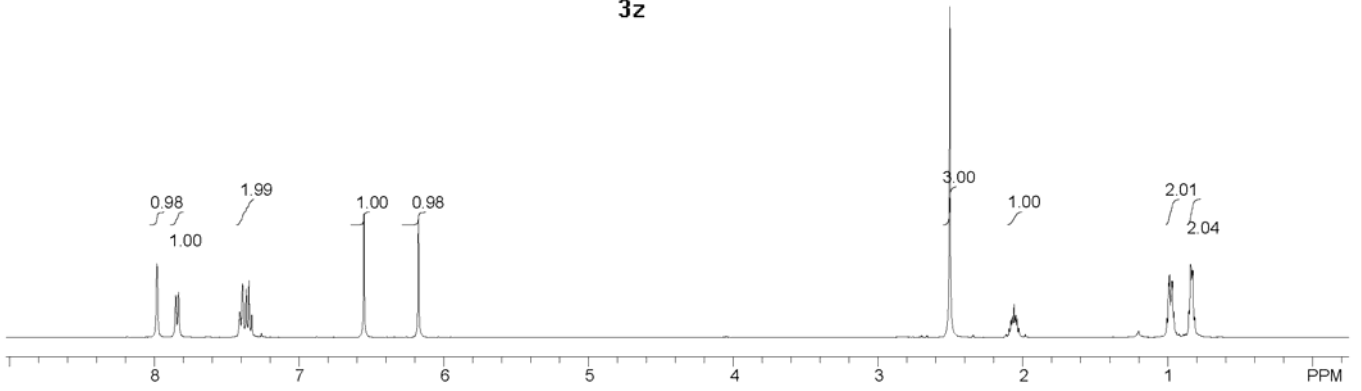
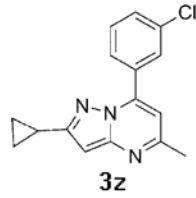
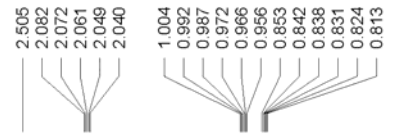
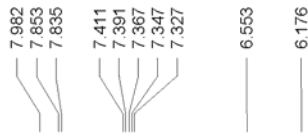
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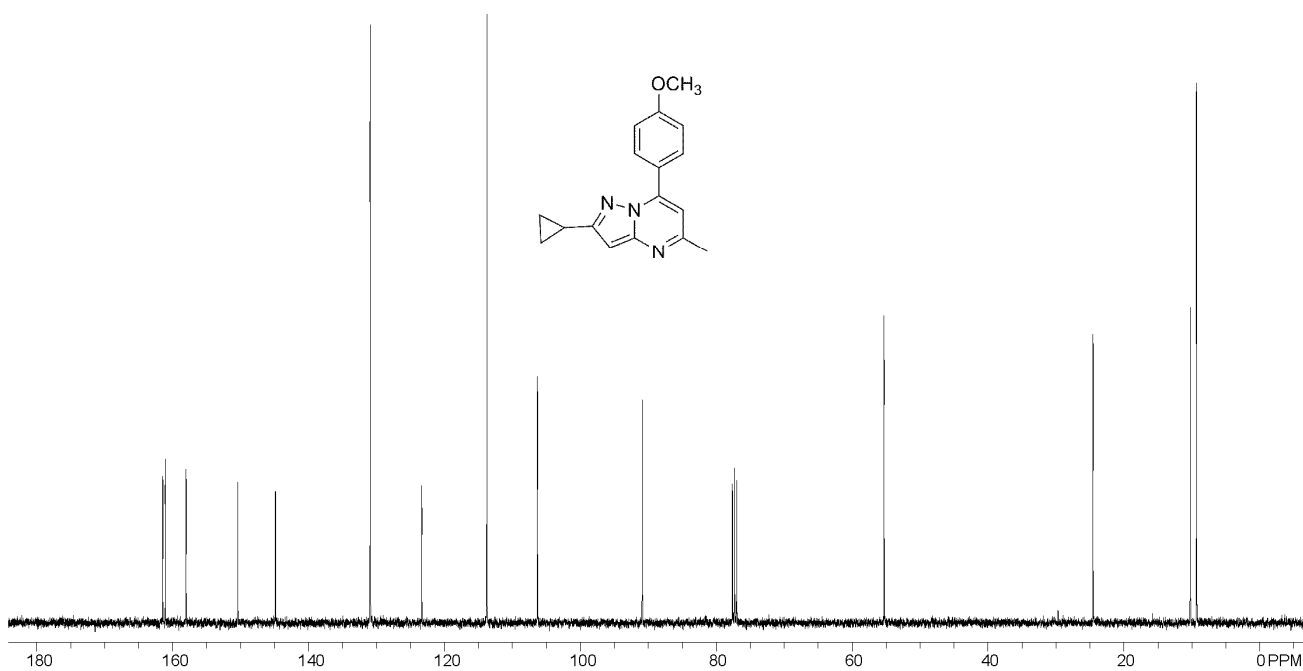
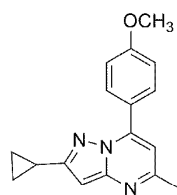
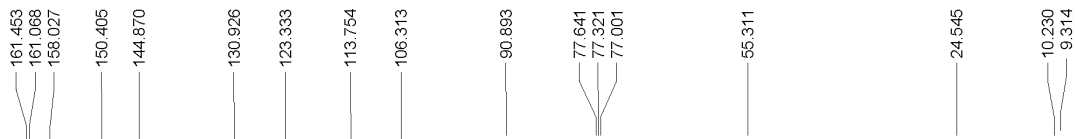
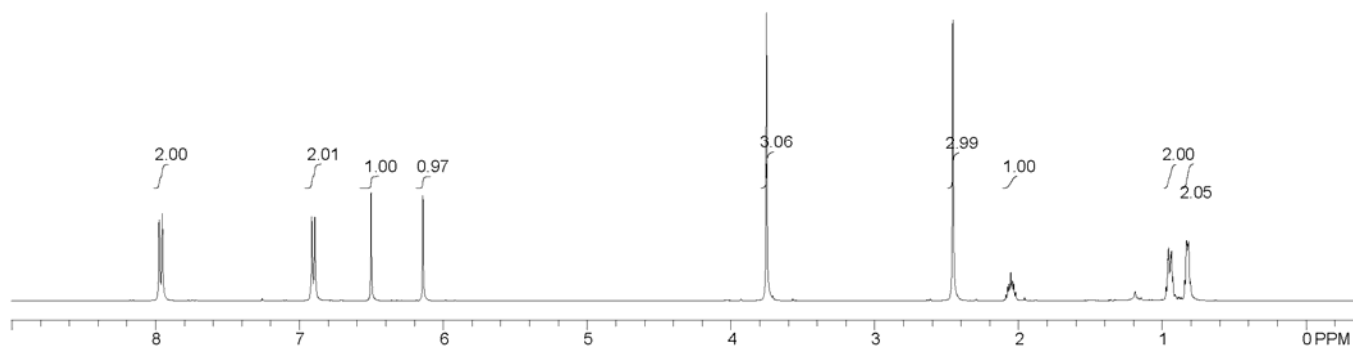
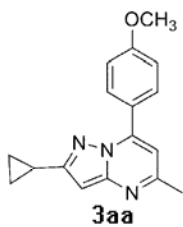
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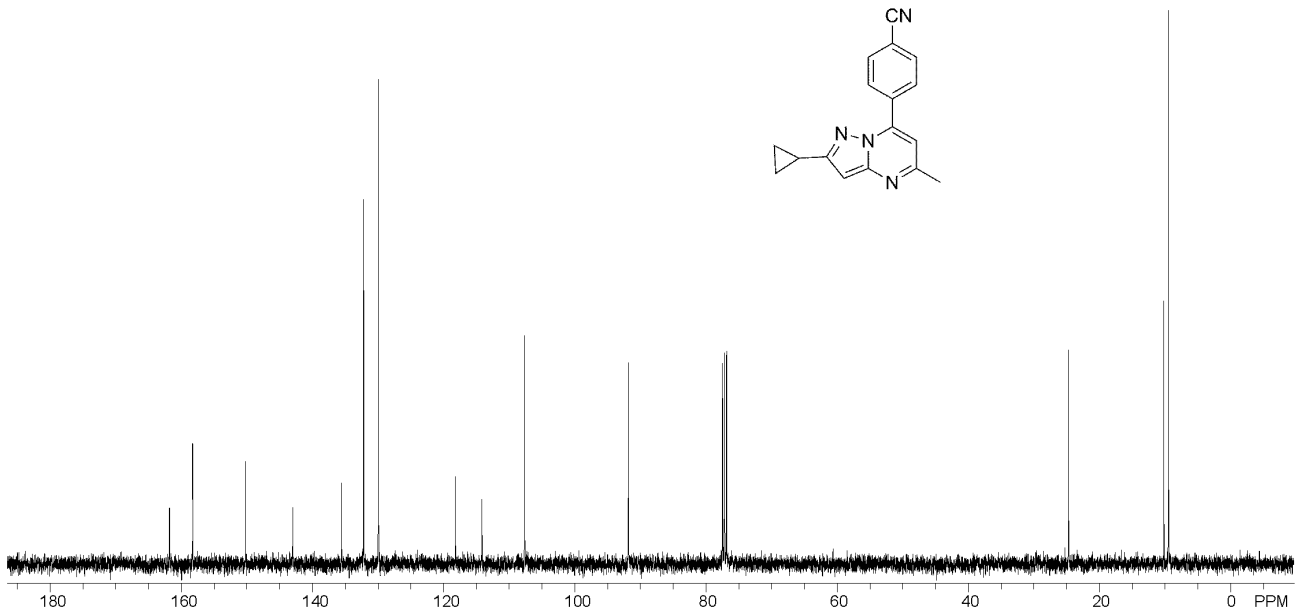
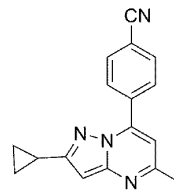
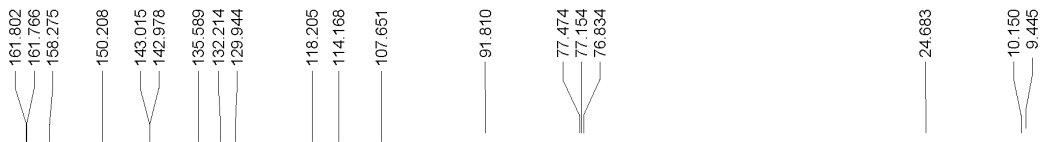
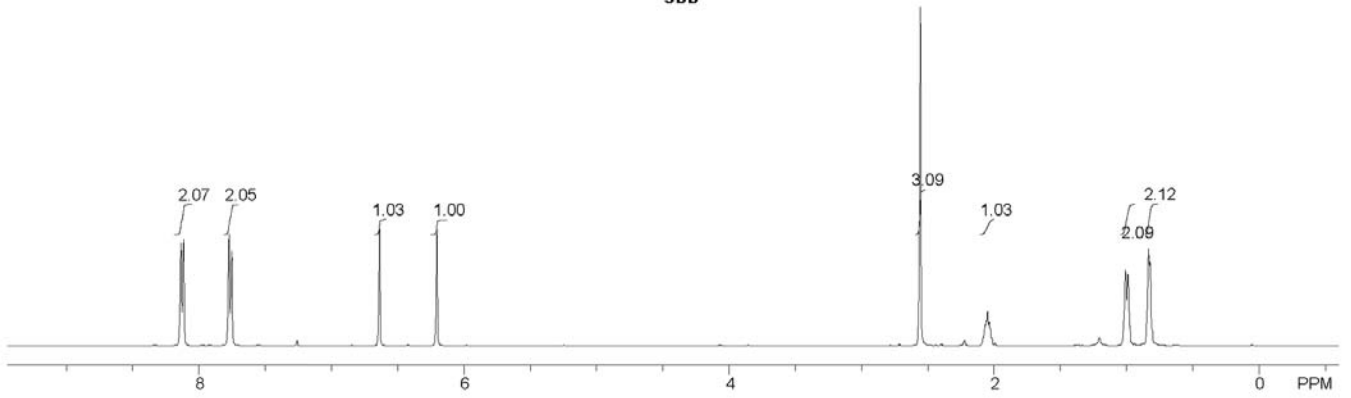
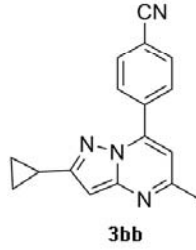


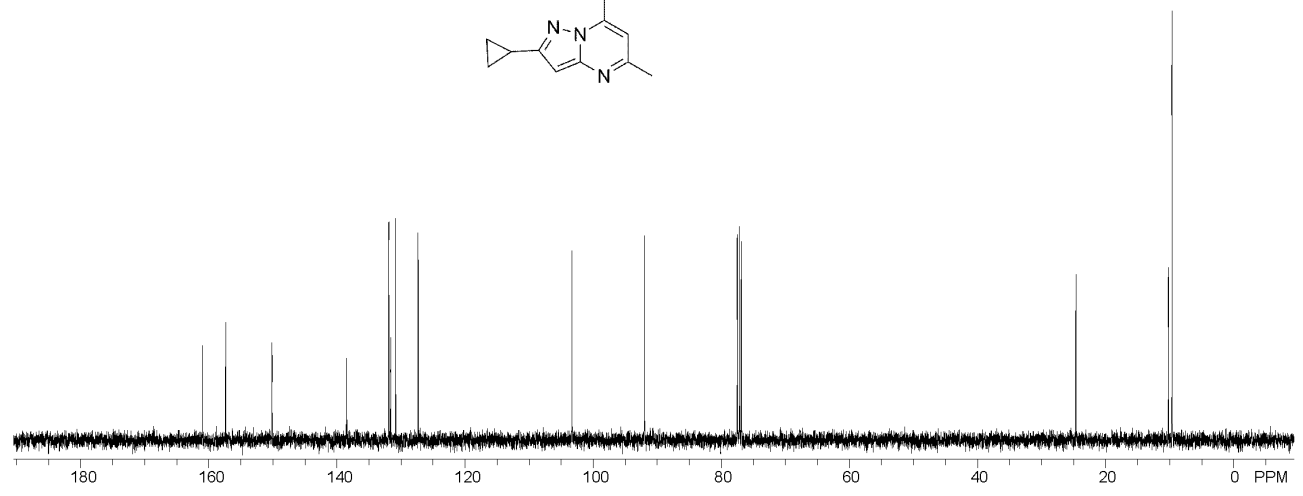
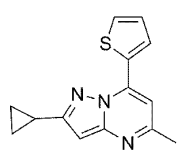
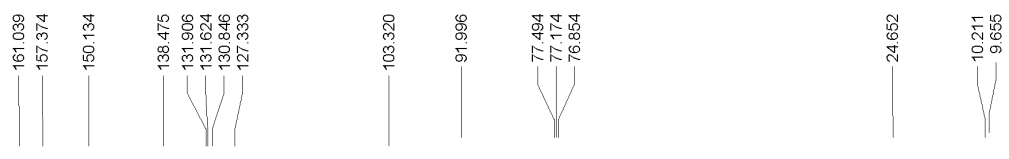
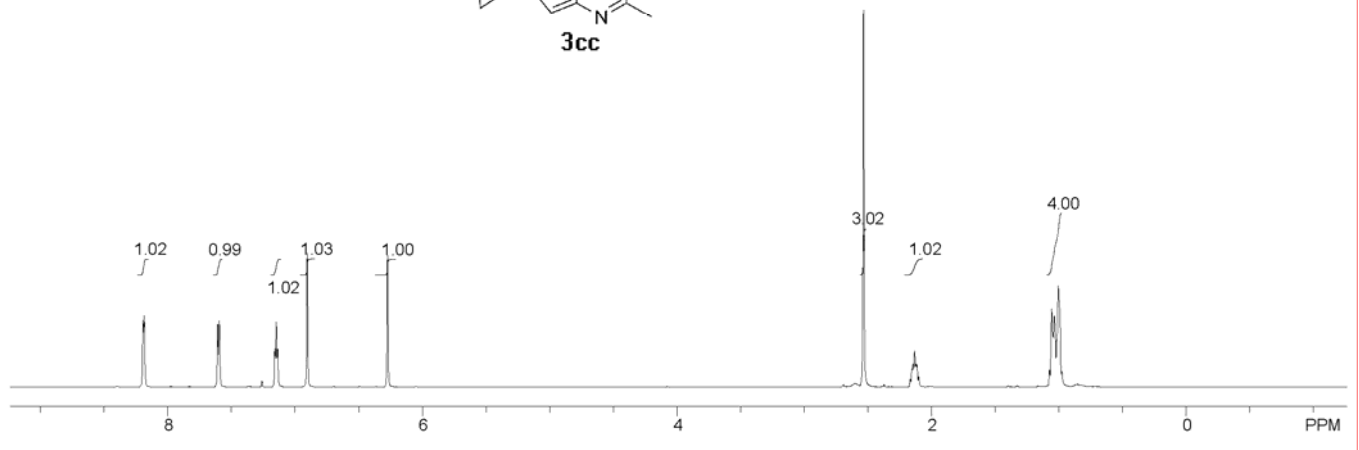
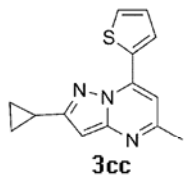
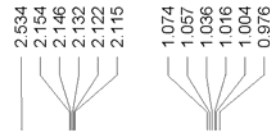
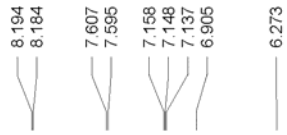


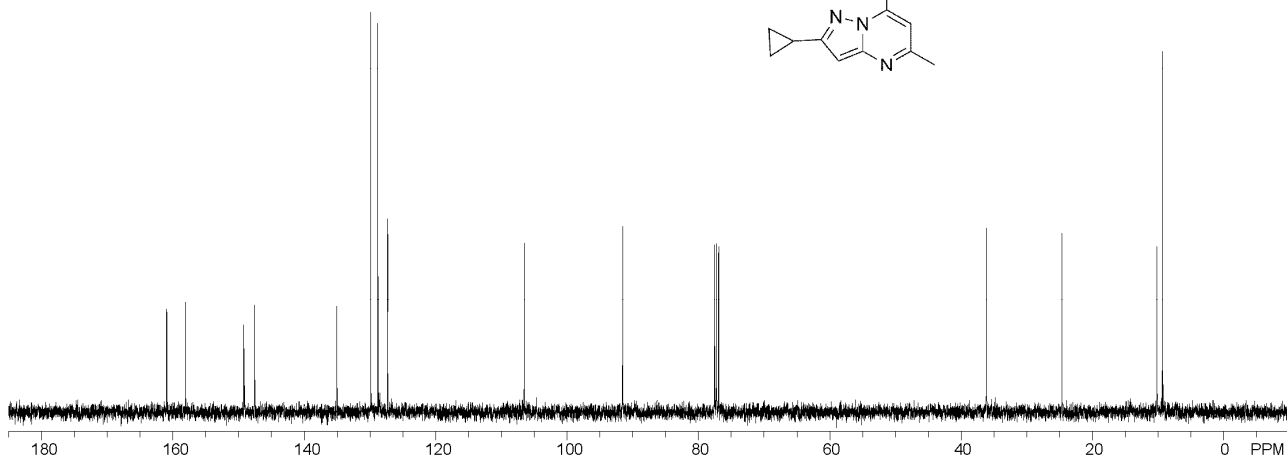
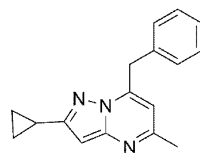
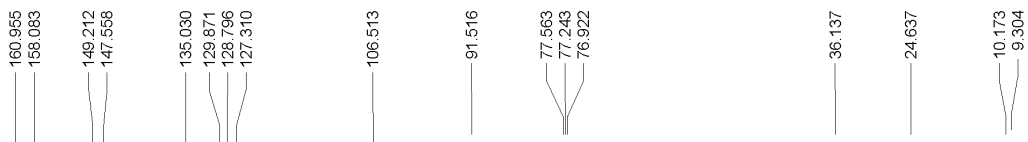
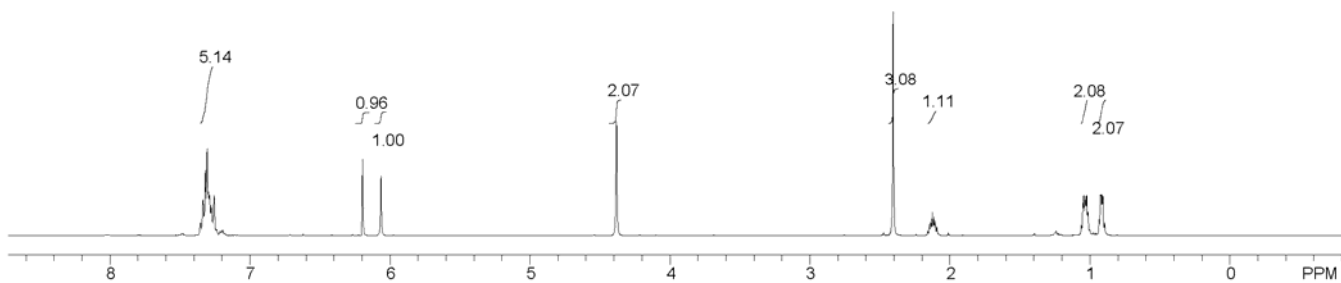
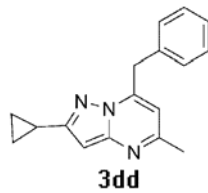












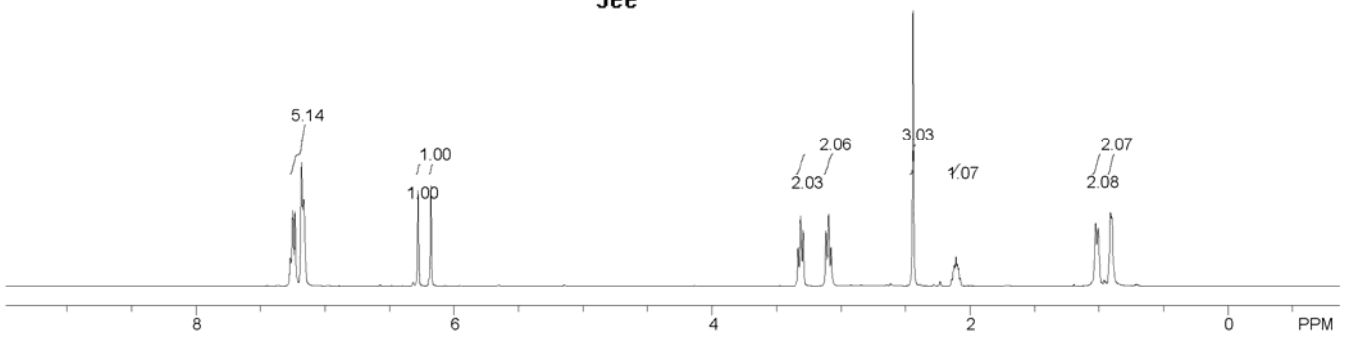
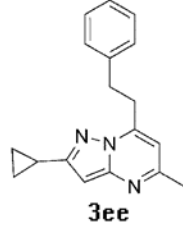
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6.180

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3.294
3.117
3.096
3.078

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2.120
2.110
2.088
2.089

1.026
1.005
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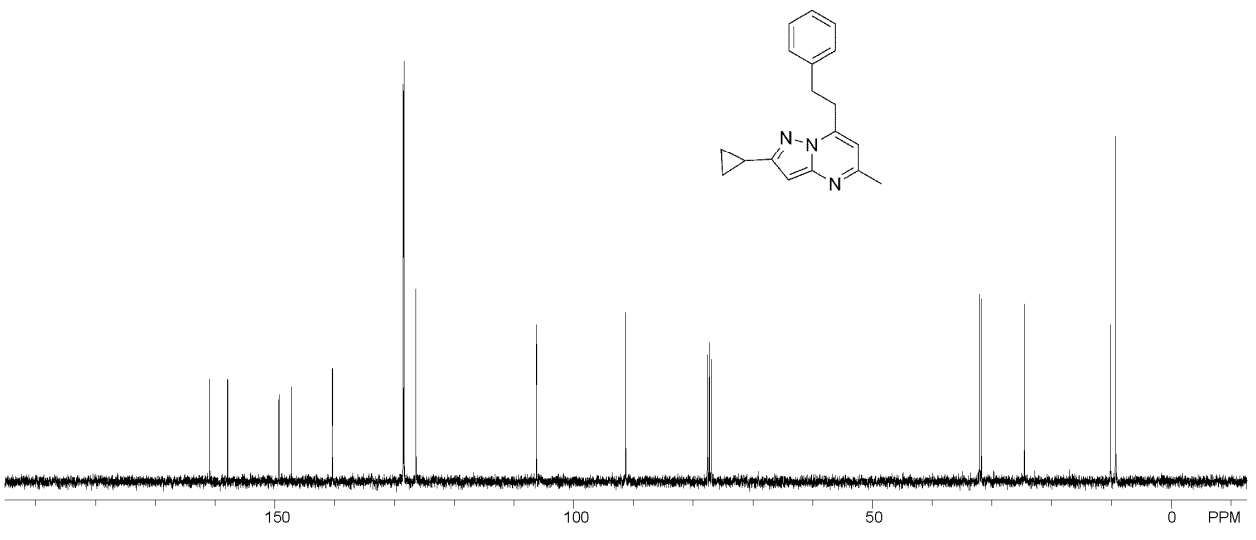
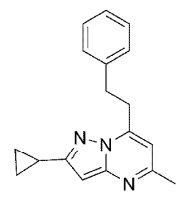
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31.816

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9.350



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8.050
8.047
8.040
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7.529
7.261

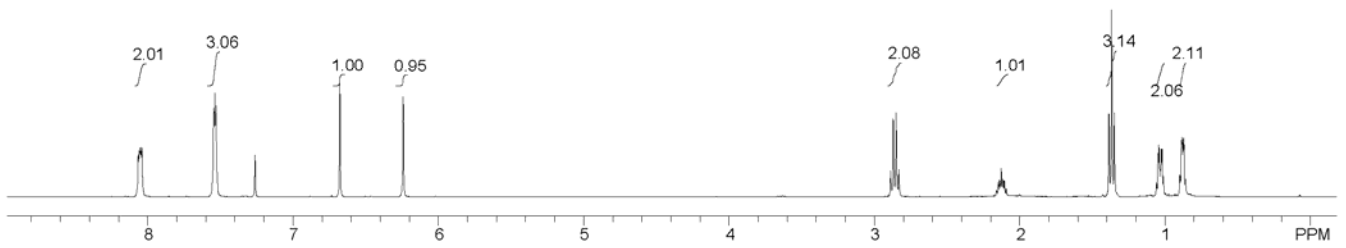
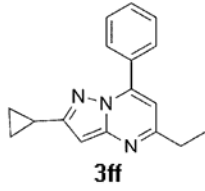
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2.139
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2.106

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1.043
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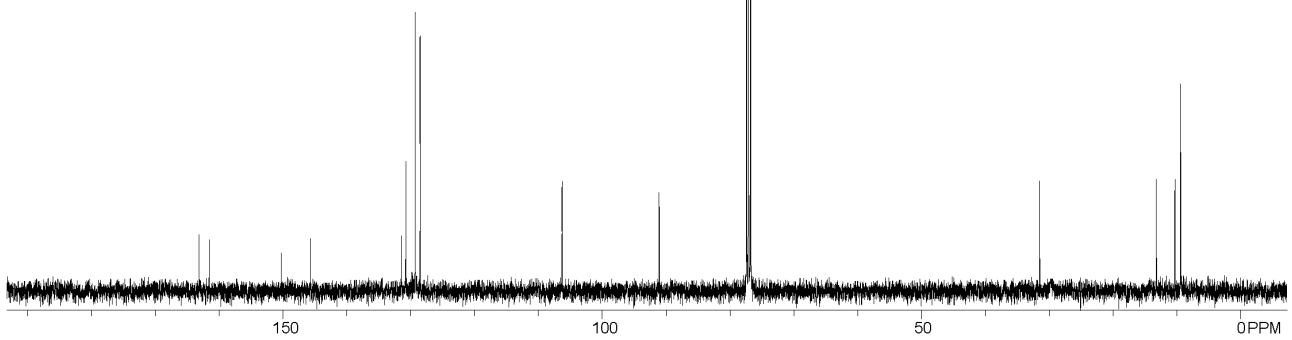
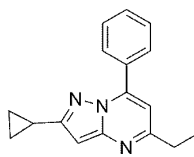
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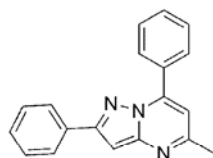
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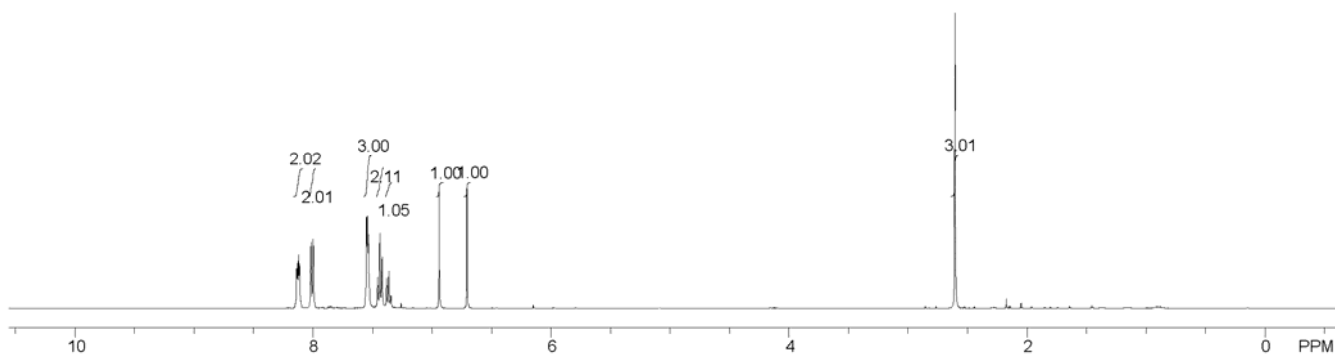


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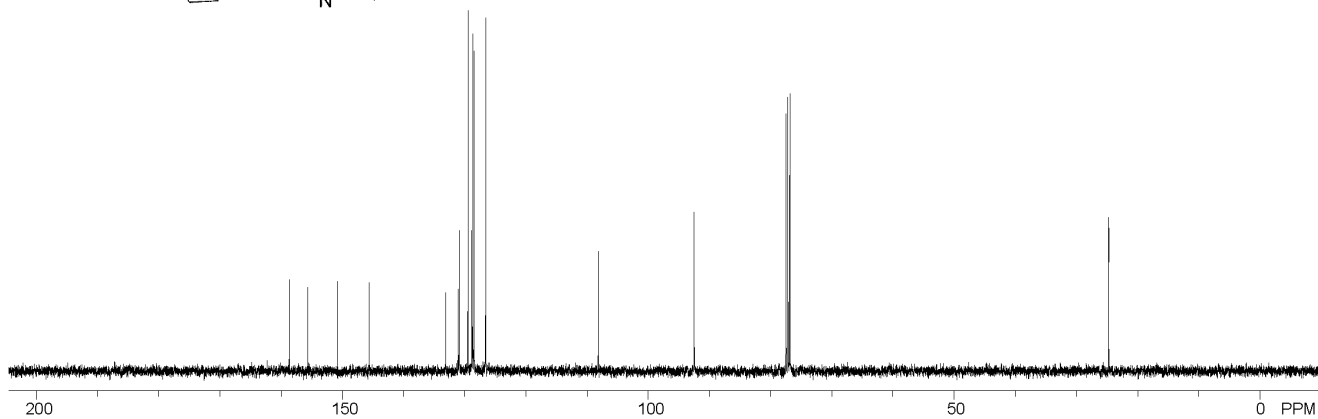
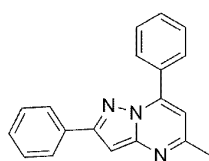
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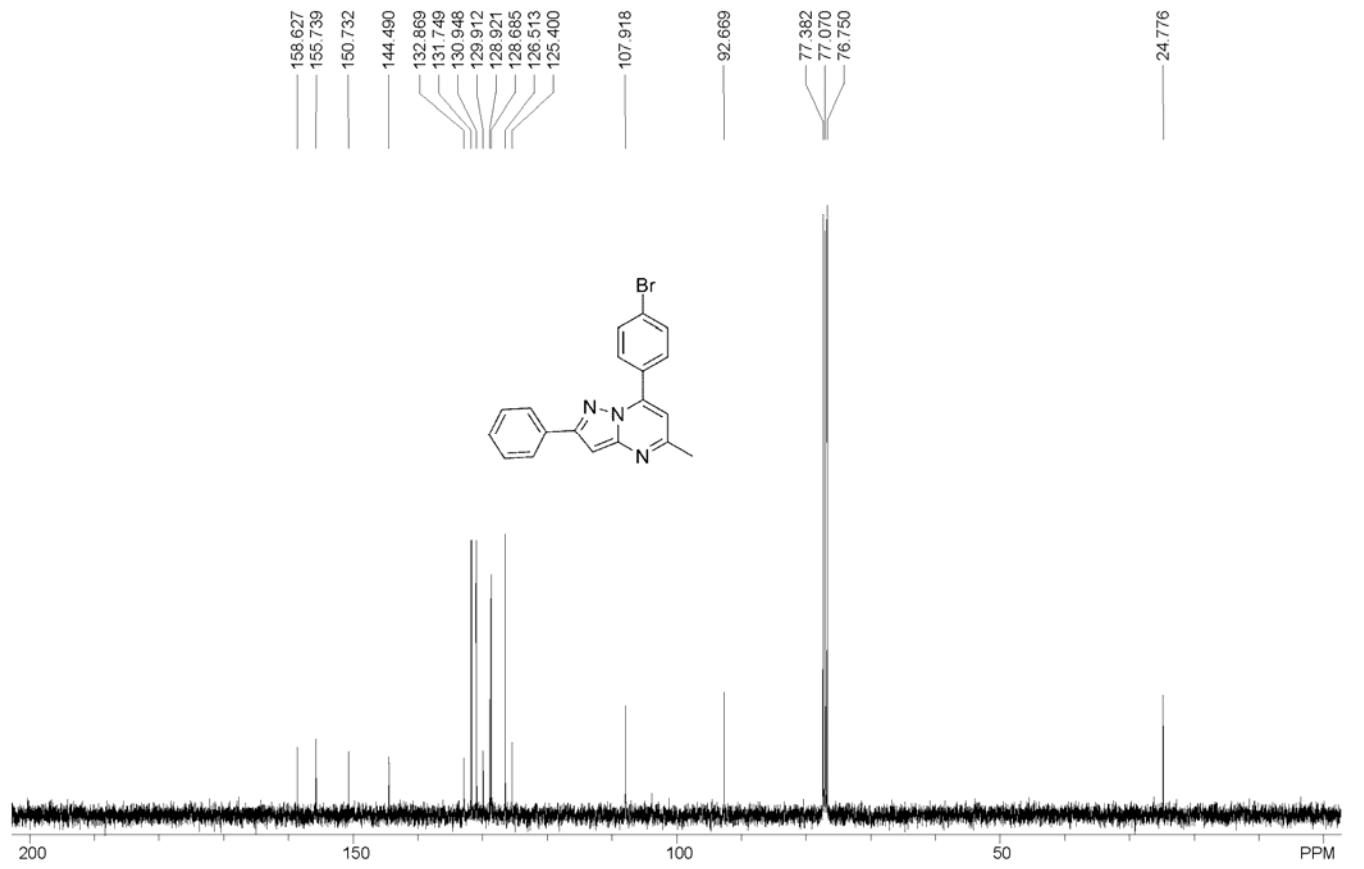
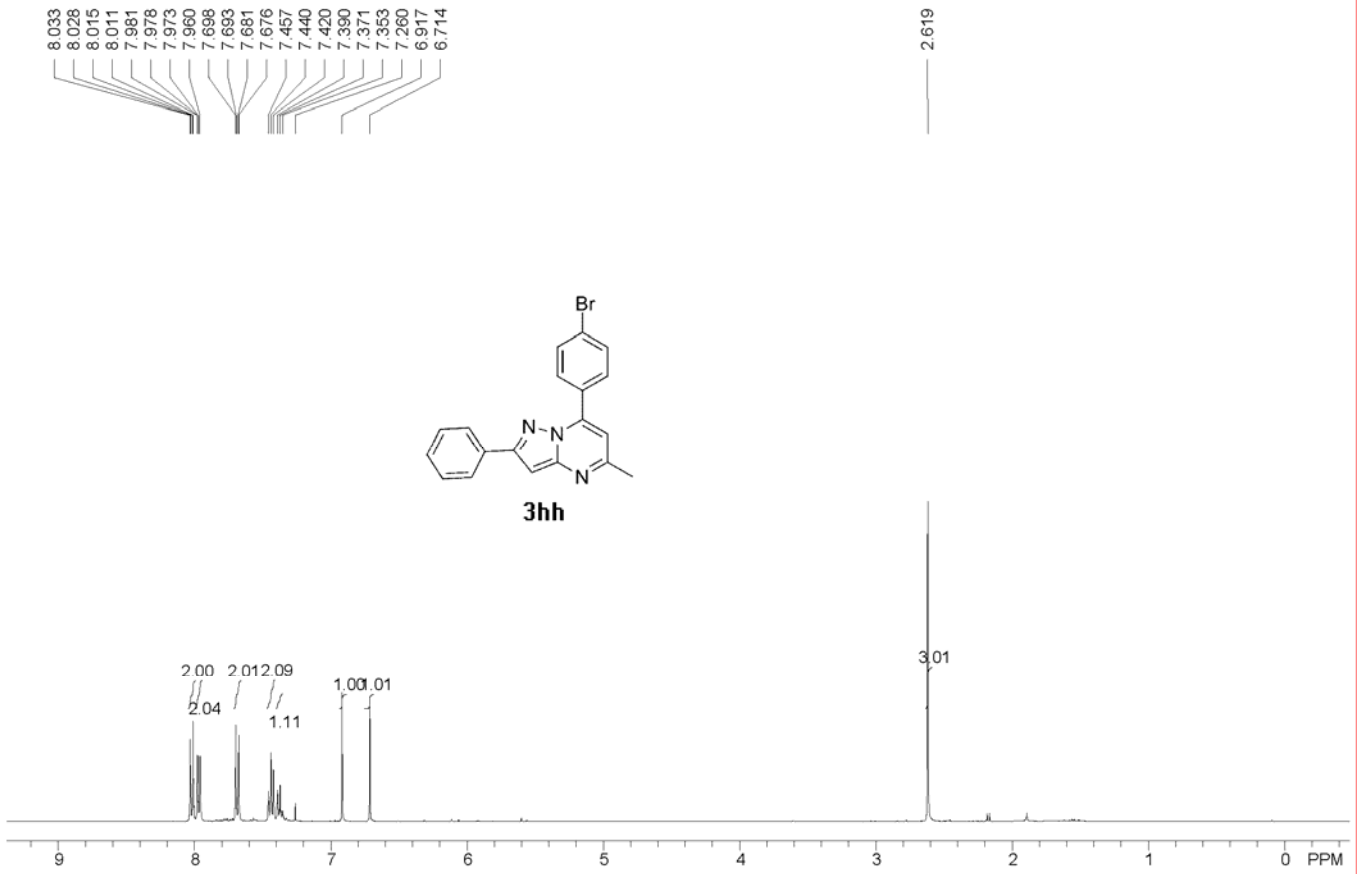
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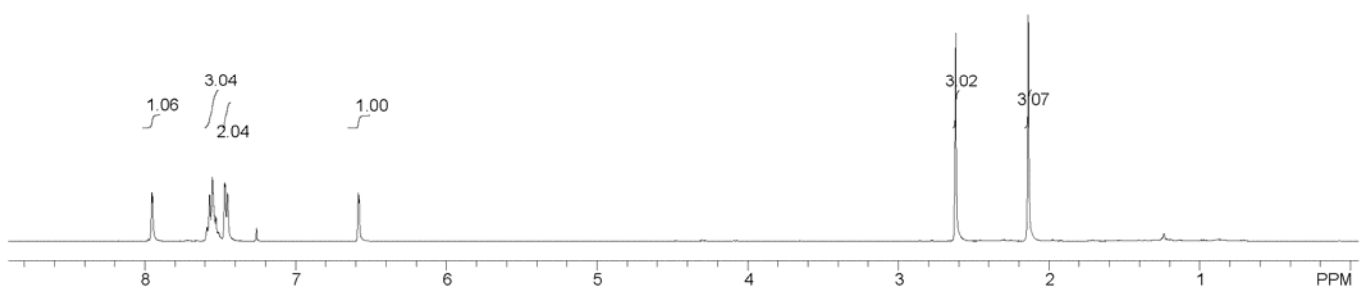
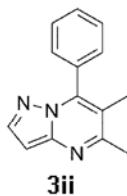
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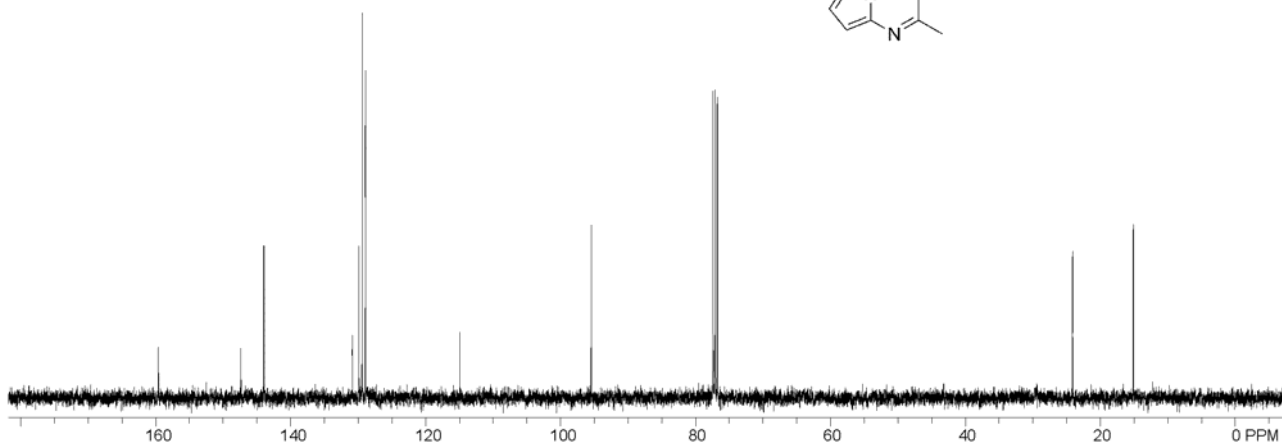
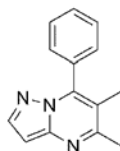


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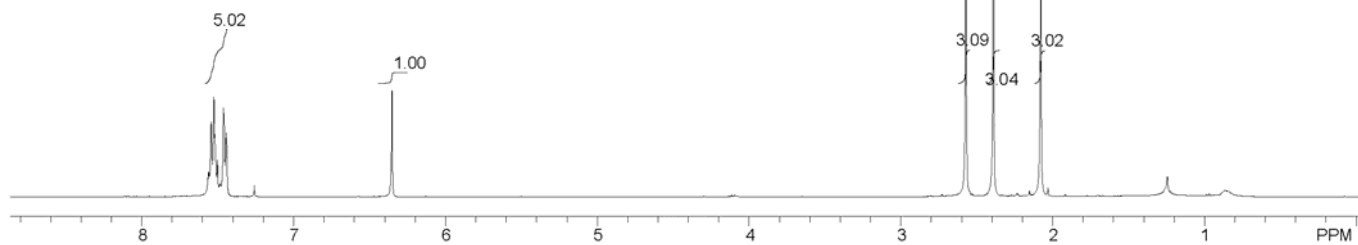
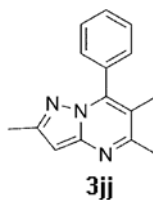
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2.078



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84.770

77.466

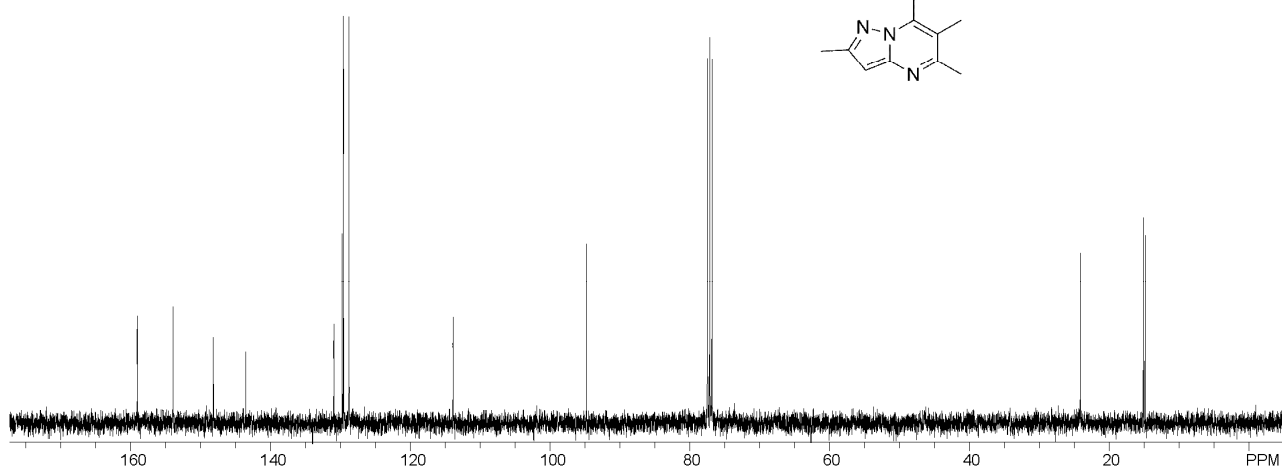
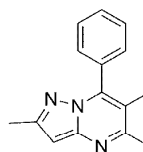
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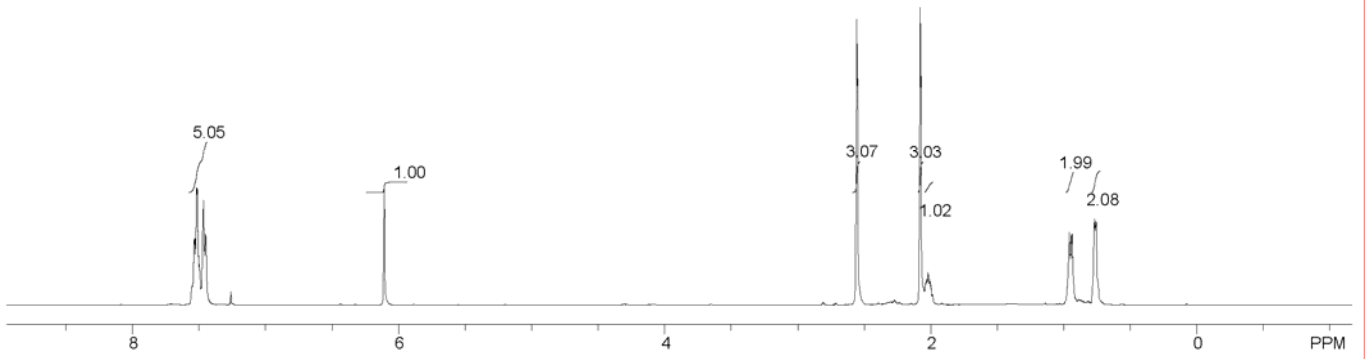
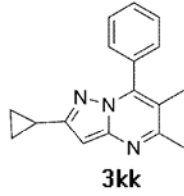
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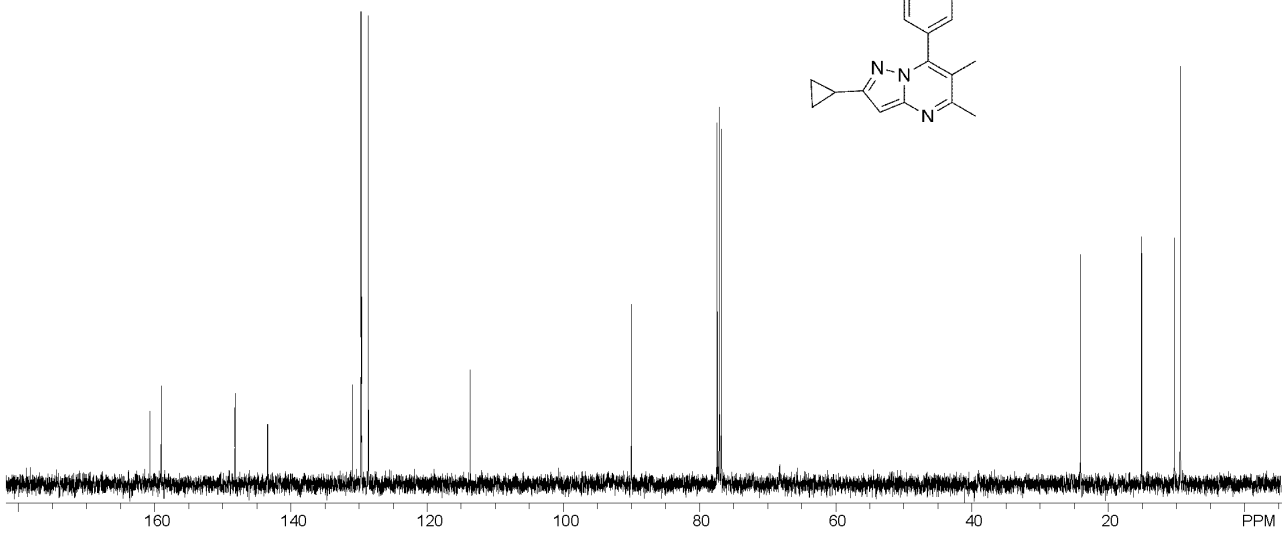
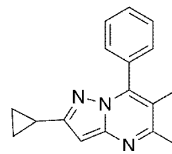


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2.075
2.032
2.028
2.020
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2.007
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0.744



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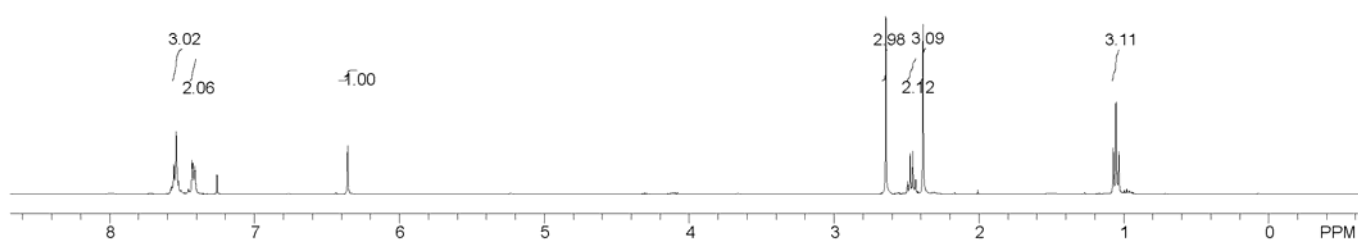
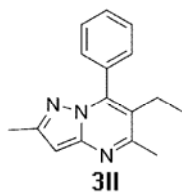


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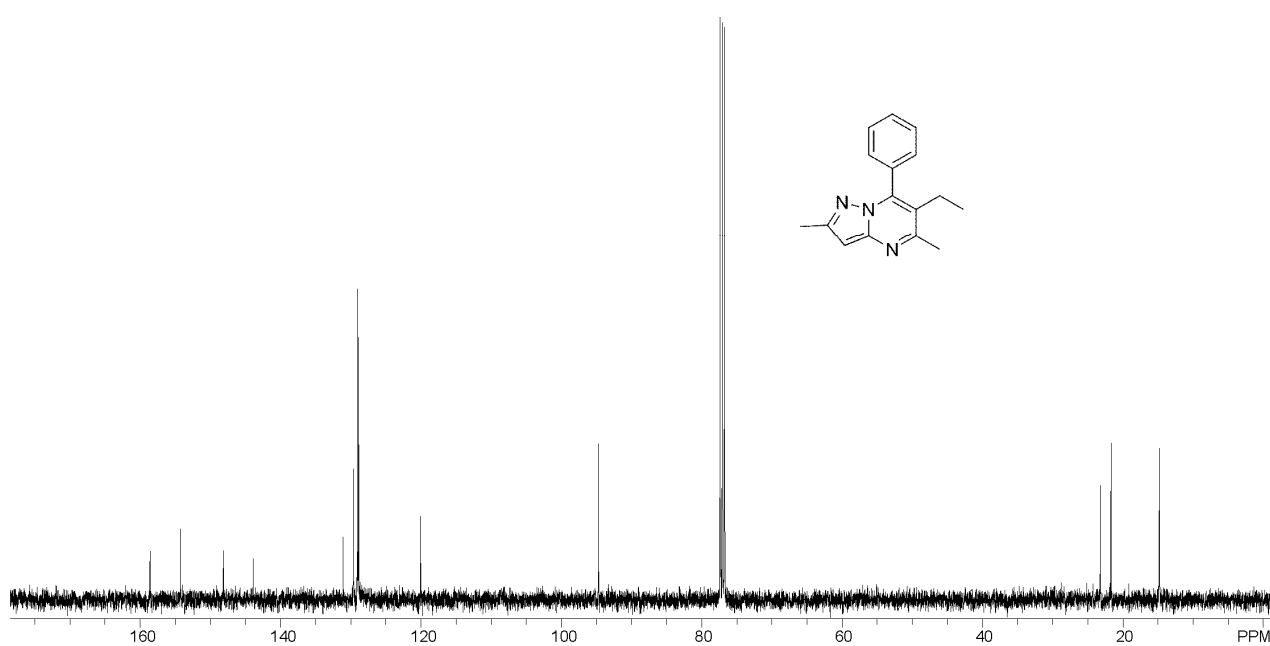
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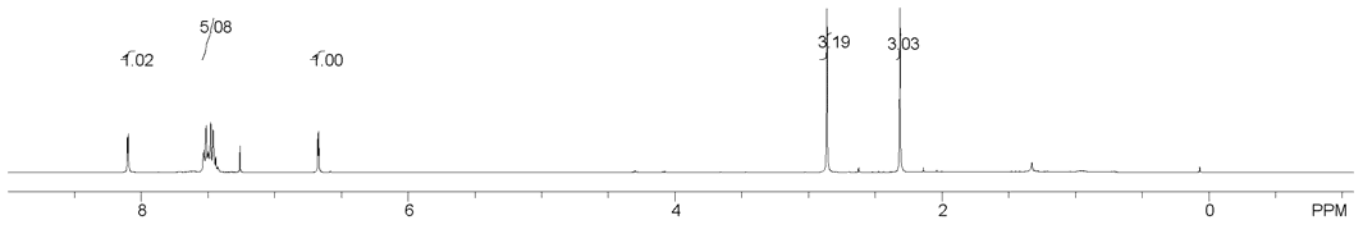
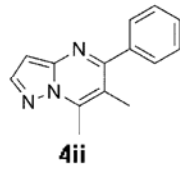
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2.315



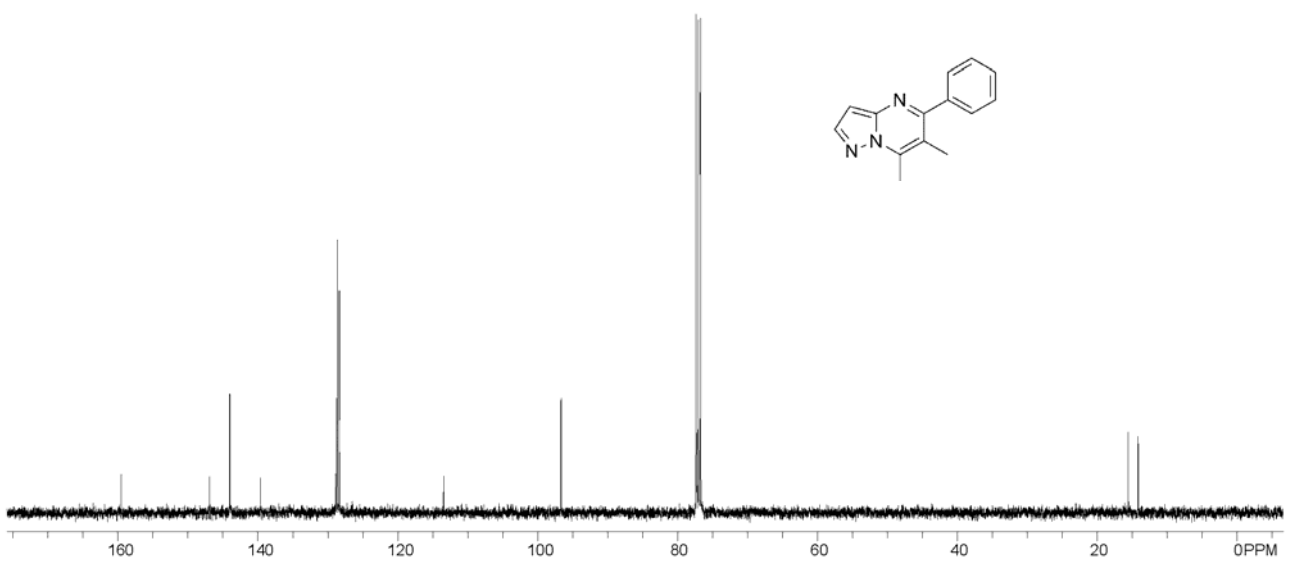
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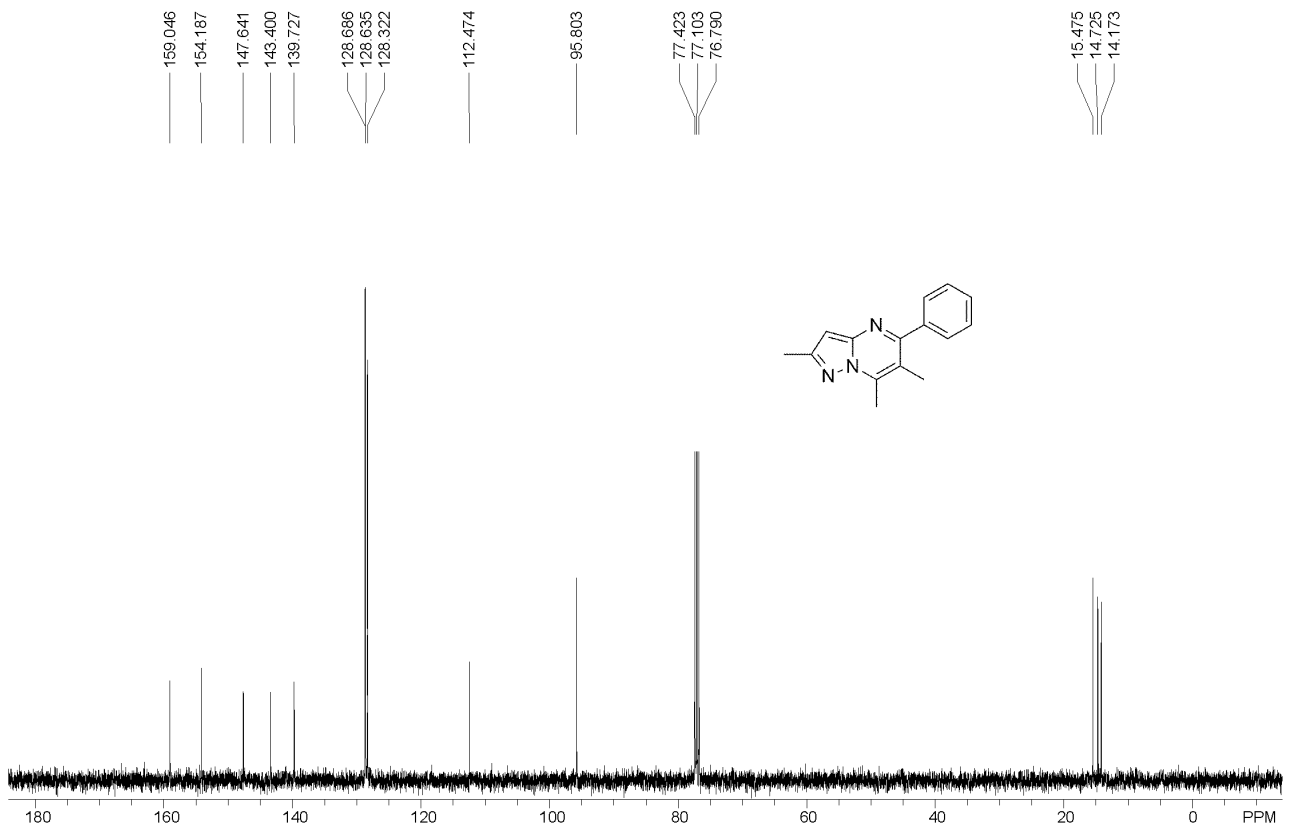
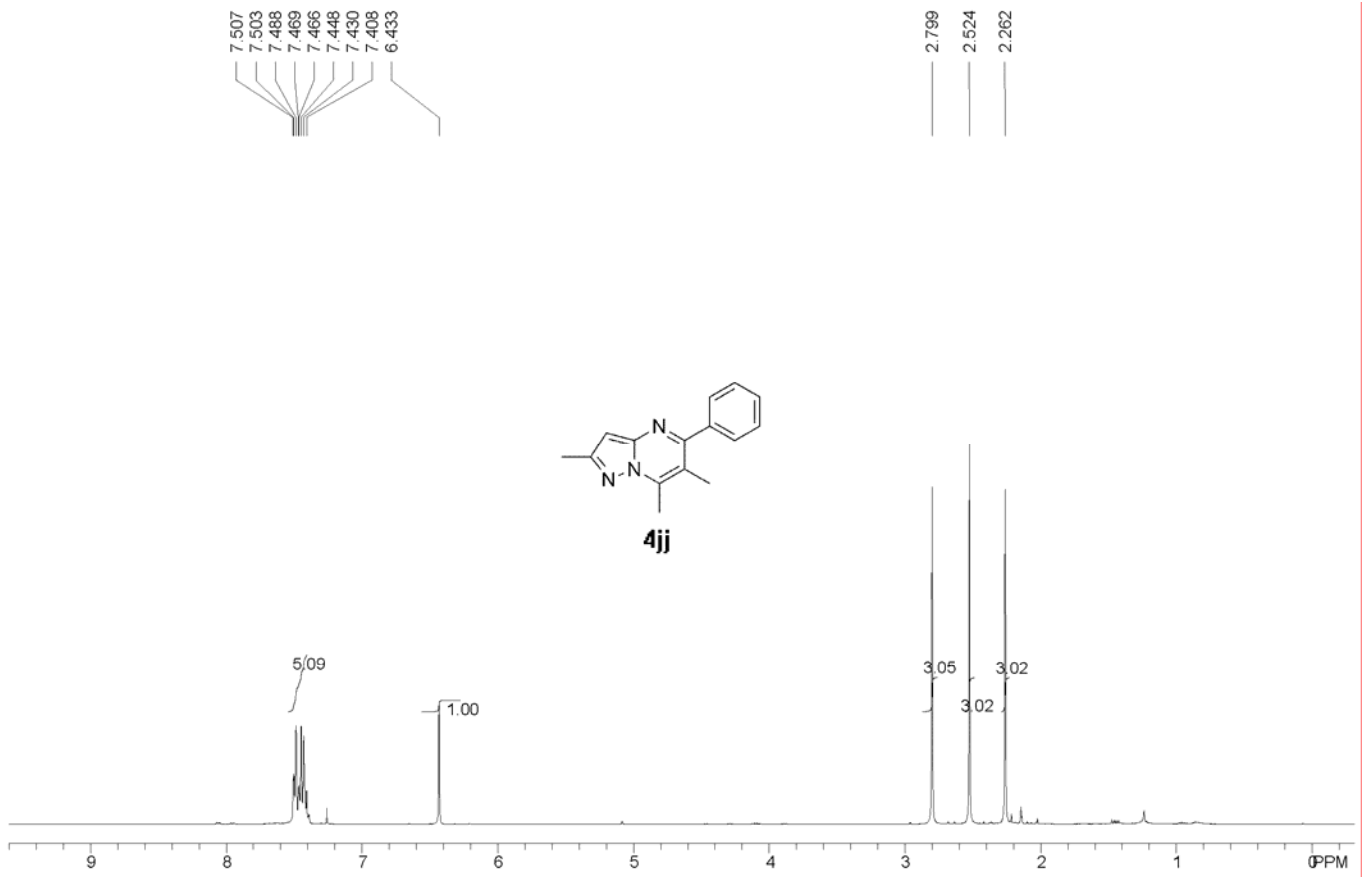
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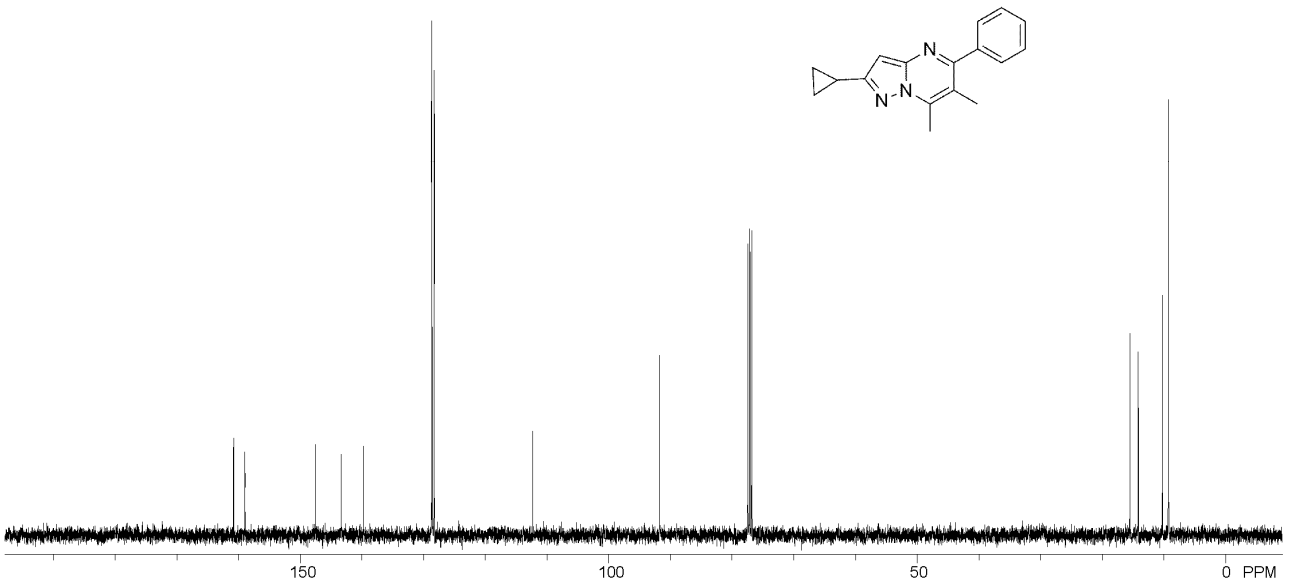
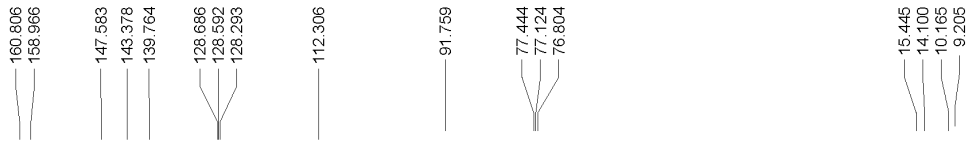
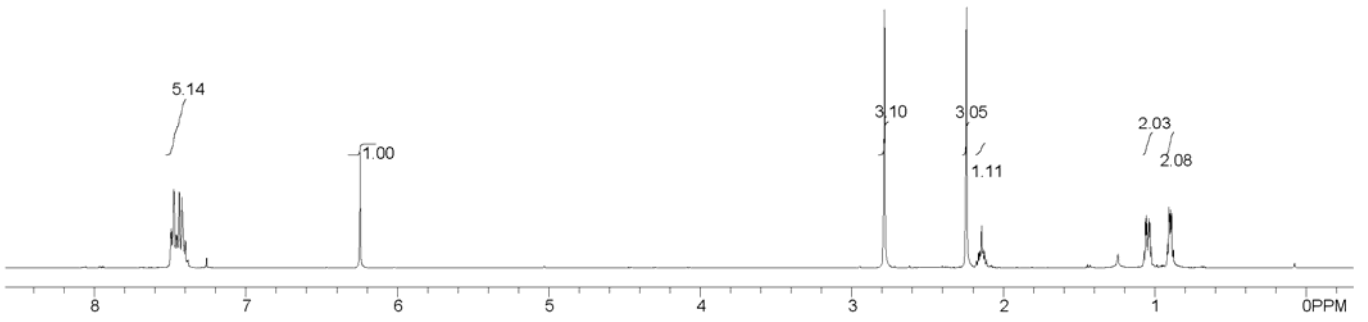
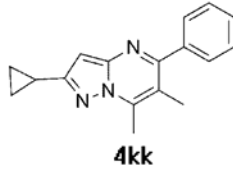
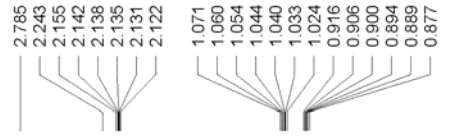
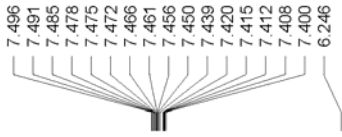
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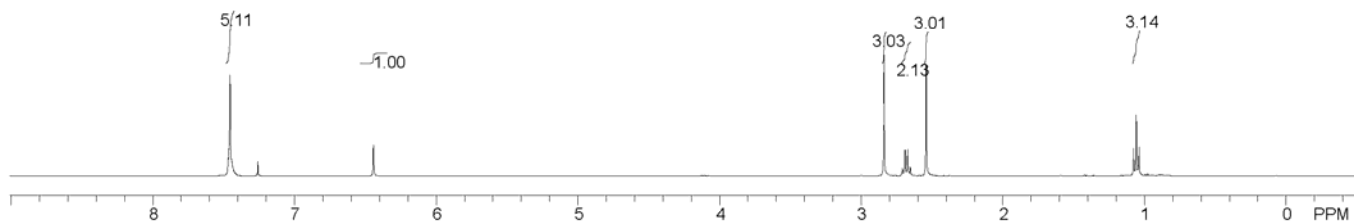
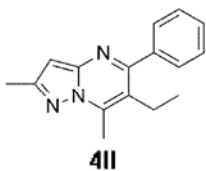


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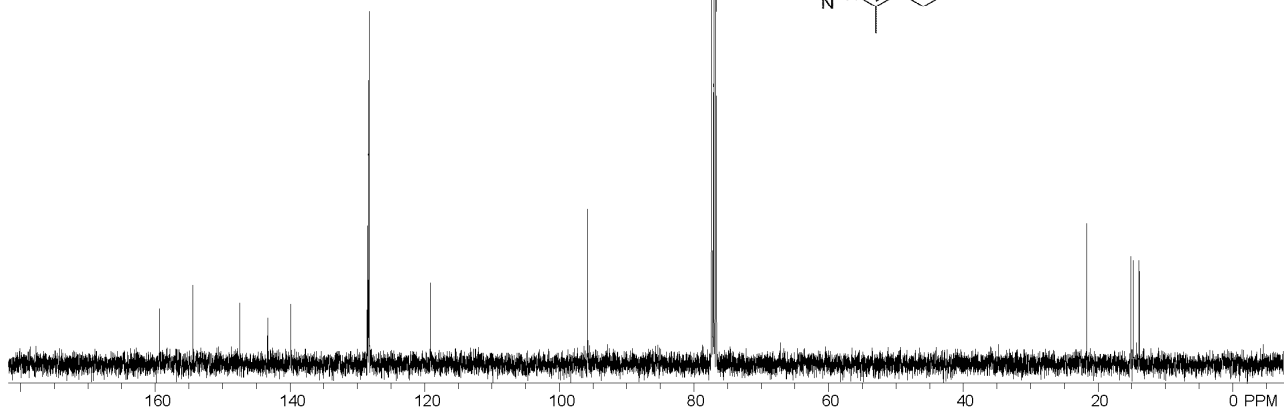
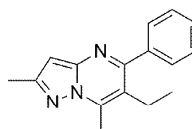
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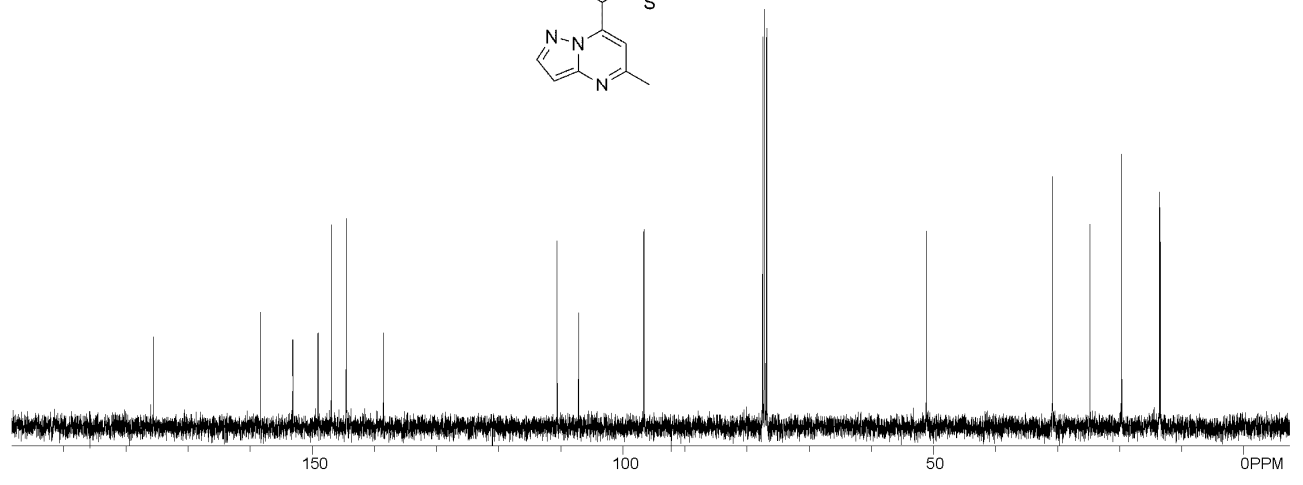
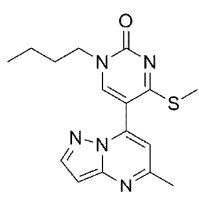
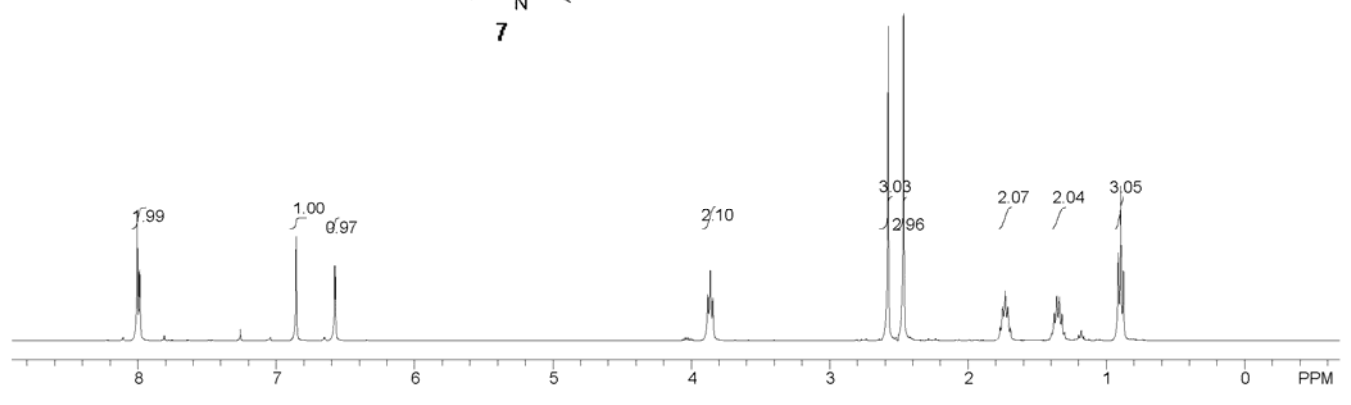
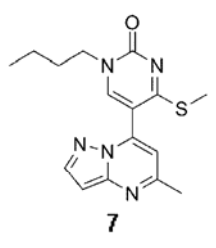
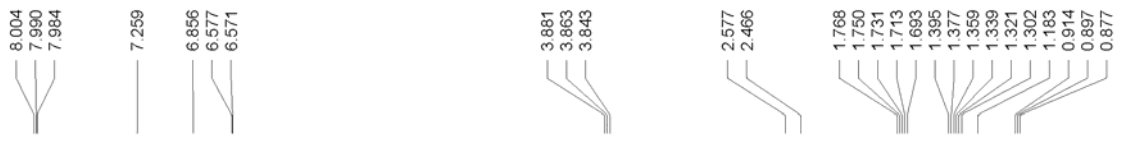
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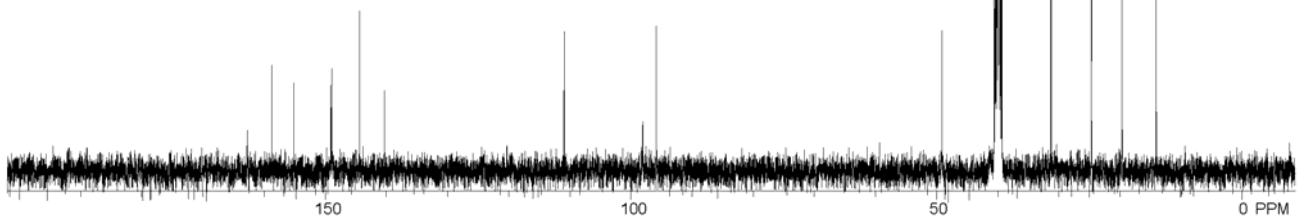
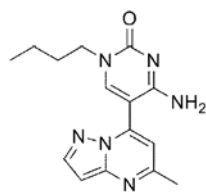
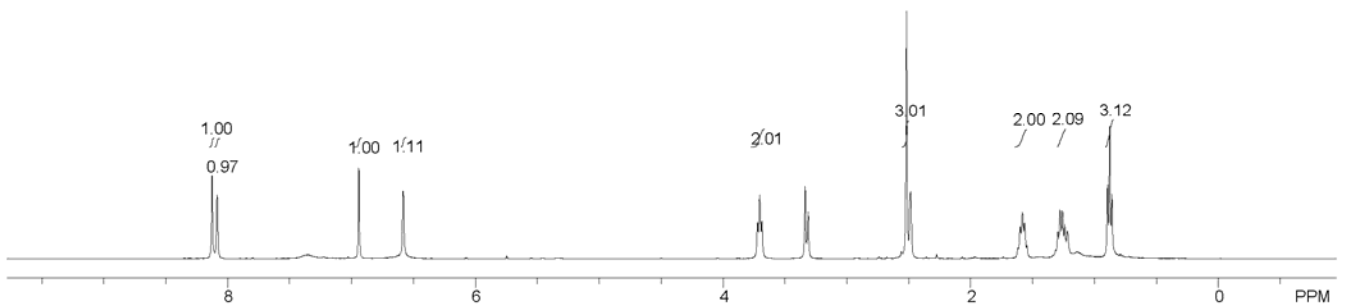
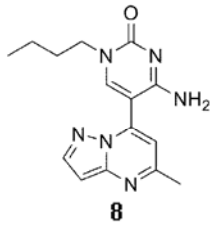
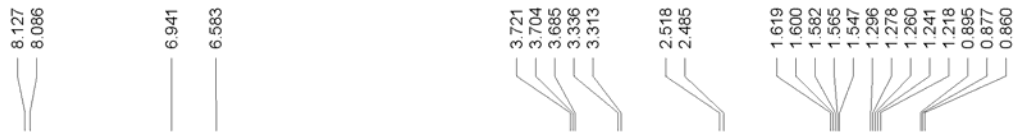
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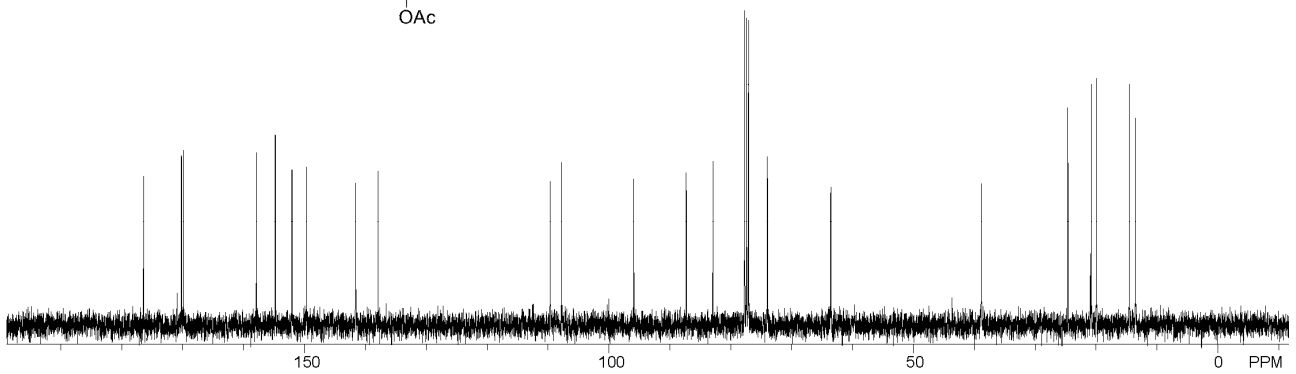
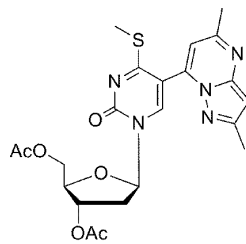
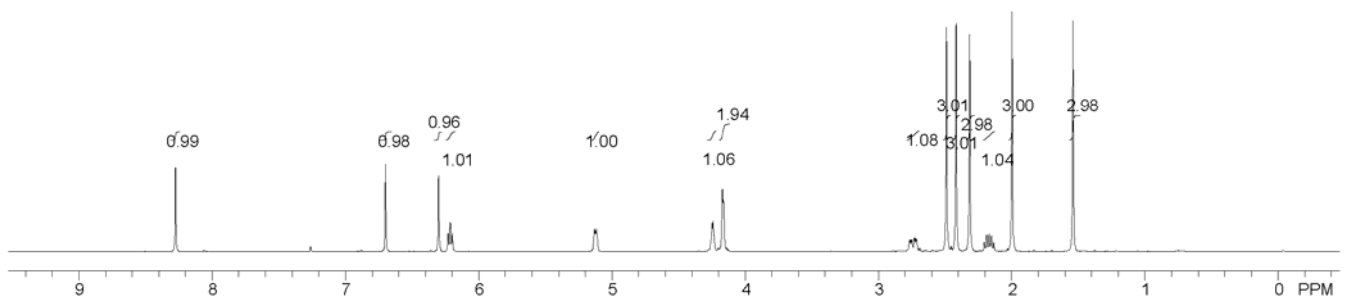
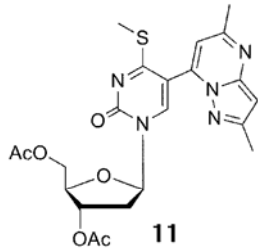
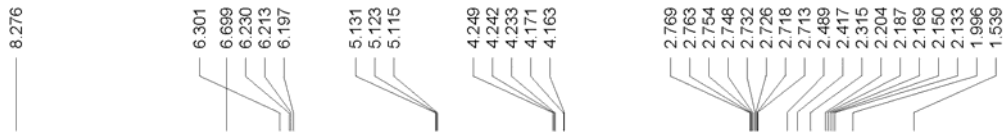
77.372
77.263
77.059
76.739

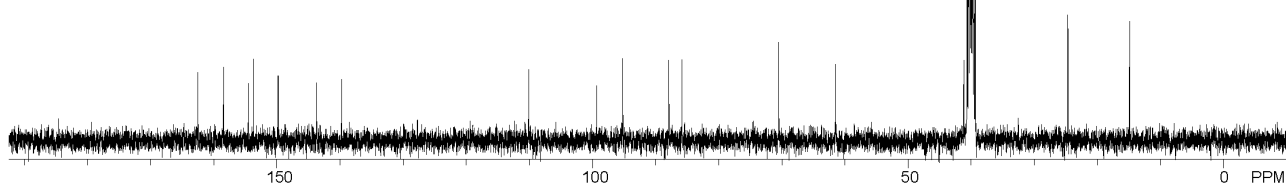
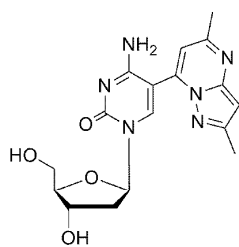
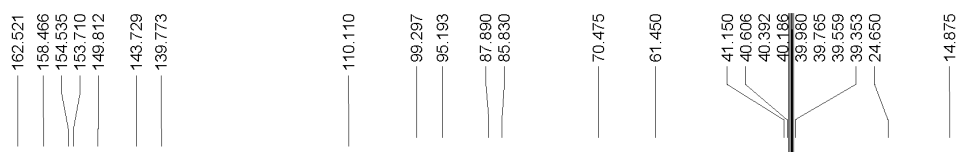
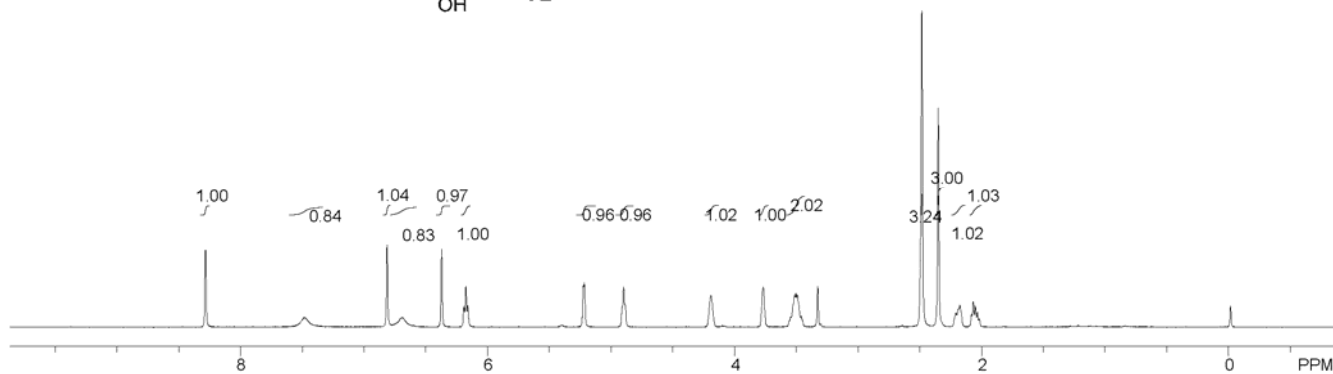
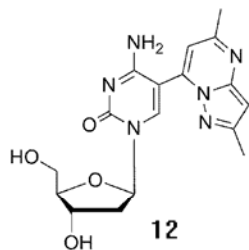
21.635
15.067
14.718
13.845











IV. X-ray crystal structures and crystal data of 3jj and 4jj

4.1 X-ray crystal structure and data of 3jj

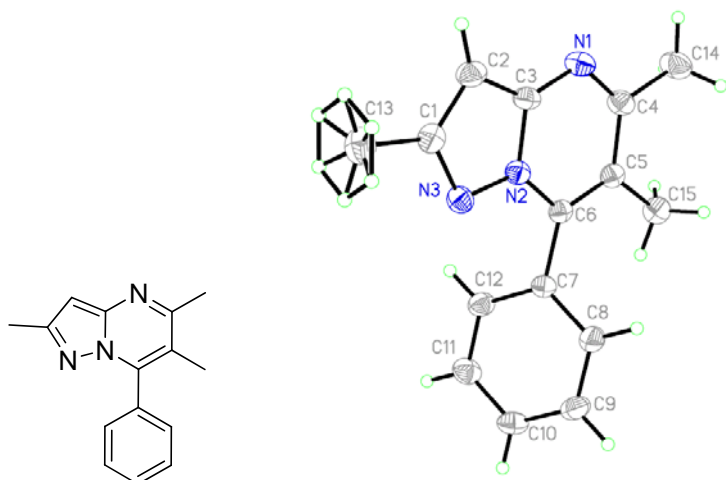


Table 1 Crystal data and structure refinement of **3jj**

Sum Formula	C ₁₅ H ₁₅ N ₃
Formula weight	237.30
Temperature	296(2) K
Radiation type, wavelength	Mo K α , 0.71073 Å
Crystal system, space group	Monoclinic, <i>P</i> 2 ₁ / <i>n</i>
Unit cell dimensions	$a = 5.784(3)$ Å $\alpha = 90^\circ$ $b = 16.411(9)$ Å $\beta = 92.703(7)^\circ$ $c = 13.211(7)$ Å $\gamma = 90^\circ$
Volume	1252.6(12) Å ³
Z	4
Calculated density	1.258 g/cm ³
Absorption coefficient	0.077 mm ⁻¹
F(000)	504
Crystal size	0.39 × 0.11 × 0.09 mm
Theta range for data collection	2.48° to 25.50°
Limiting indices	$-6 \leq h \leq 6$, $-19 \leq k \leq 19$, $-15 \leq l \leq 15$
Reflection collected/unique	7574/2284 [R(int) = 0.050]
Data completeness	0.985
Theta (max)	25.50

Absorption correction	Multi-scan
Refinement method	Full-matrix least-squares on F^2
Data/restraints/parameters	2284/0/165
Goodness-of-fit on F^2	1.033
Final R indices [$I > 2\sigma(I)$]	$R_1 = 0.0484$, $wR_2 = 0.1084$
R indices (all data)	$R_1 = 0.0804$, $wR_2 = 0.1257$
Largest diff. peak and hole	0.16 and -0.16 e. \AA^{-3}

4.2 X-ray crystal structure and data of 4jj

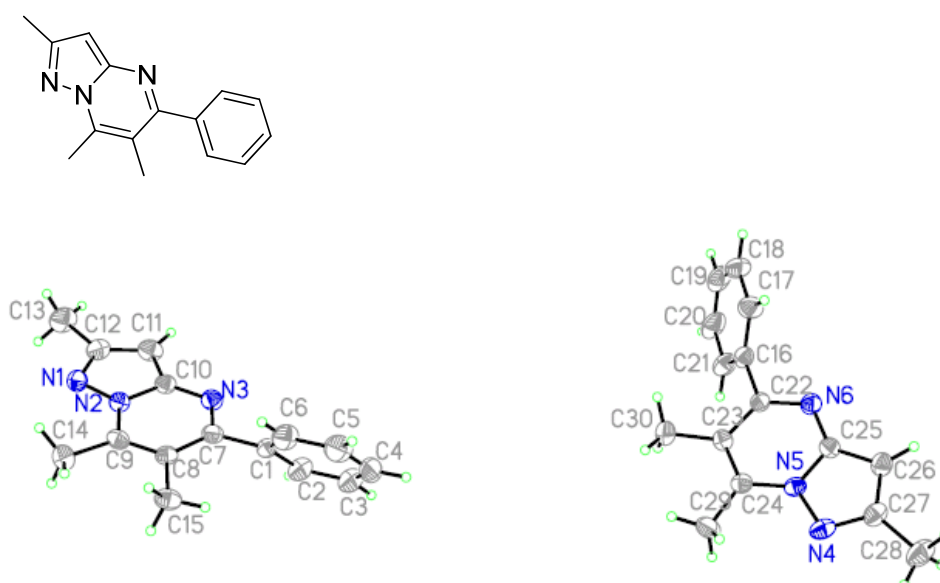


Table 2 Crystal data and structure refinement of **4jj**

Sum Formula	$C_{30}H_{30}N_6$
Formula weight	474.60
Temperature	296(2) K
Radiation type, wavelength	Mo $K\alpha$, 0.71073 \AA
Crystal system, space group	Monoclinic, $P2_1/c$
Unit cell dimensions	$a = 25.257(13) \text{\AA}$ $\alpha = 90^\circ$ $b = 7.226(4) \text{\AA}$ $\beta = 106.999(6)^\circ$ $c = 14.807(8) \text{\AA}$ $\gamma = 90^\circ$
Volume	$2584(2) \text{\AA}^3$
Z	4
Calculated density	1.220 g/cm^3

Absorption coefficient	0.075 mm ⁻¹
F(000)	1008
Crystal size	0.33 × 0.25 × 0.11 mm
Theta range for data collection	2.88° to 21.86°
Limiting indices	-30 ≤ h ≤ 30, -8 ≤ k ≤ 8, -17 ≤ l ≤ 17
Reflection collected/unique	4741/4741 [R(int) = 0.0000]
Data completeness	0.986
Theta (max)	25.50
Absorption correction	Multi-scan
Refinement method	Full-matrix least-squares on F ²
Data/restraints/parameters	4741/0/333
Goodness-of-fit on F ²	1.045
Final R indices [I > 2σ(I)]	R ₁ = 0.0519, wR ₂ = 0.1307
R indices (all data)	R ₁ = 0.0849, wR ₂ = 0.1477
Largest diff. peak and hole	0.14 and -0.16 e.Å ⁻³
