

Metal-ligand cooperative strategy of Zn-catalyst for multicomponent synthesis of highly substituted pyrazolines, pyrimidines, and quinolines: A combined experimental and DFT optimized study

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1. Experimental

1.1 Materials used

All the chemicals and solvents were purchased from commercially available sources such as Aldrich, Merck, TCI, SRL, and ACROS organics and used as received. Analytical grade reagents and solvents were used for whole activities and spectroscopic studies. 2,6-Bis(1-phenylhydrazinyl) pyridine was synthesized according to literature.¹ UV-Vis. Spectra were recorded using a UV-Vis Spectrophotometer (UV-2600 from Shimadzu). IR spectra were recorded on an ALPHA II compact FT-IR spectrometer with the use of KBr pellets (16 scans in cm^{-1}). NMR spectra were recorded using Bruker Avance III 500 MHz (AV 500) and Jeol 500 MHz spectrometers in deuterated solvents. Chemical shift values in NMR are recorded in δ (ppm) values against TMS as an internal standard. NMR coupling constants (J) are represented in hertz (Hz) values. Multiplets in the chemical shift are given as s (singlet), d (doublet), t (triplet), q (quartet), dd (doublet of doublets), ddd (doublet of doublet of doublets), dt (doublet of triples), td (triplet of doublets). All the simulated HRMS data were obtained from the free software available (<https://www.envipat.eawag.ch/index.php>).

1.2 X-ray crystallography

The X-ray data collection and processing of complex 1 and complex 2 were performed on a Bruker Kappa (D8 QUEST) Apex-IV CMOS diffractometer by using graphite monochromated Mo-K α radiation ($\lambda = 0.71070 \text{ \AA}$) at 119 K and 105 K, respectively. The crystal structures were solved by SIR-92 GUI control methods. The Olex² software was used to solve the crystal structures and refine them using ShelXT and ShelXL, respectively.³ The bond lengths and bond angles were computed with the help of MERCURY software. Additionally, ORTEP⁴ images were generated using the same MERCURY program. The hydrogen atoms were placed in geometrically determined positions and refined using the riding model. The accompanying supplementary information (SI) files contain pertinent crystallographic parameters.

1.3 Computational methods

Geometry optimizations and frequency calculations were performed using the spin-restricted B3LYP functional, in combination with the def2-SVP basis set for all atoms except zinc, for which the def2-TZVP basis set was used. Dispersion corrections were included using the DFT-D4 approach. Solvent effects (toluene) were treated using the conductor-like polarizable continuum model (CPCM) within the self-consistent reaction field (SCRF) framework. Frequency calculations were conducted at 383.15 K to obtain thermochemical corrections. All transition states were confirmed to possess a single imaginary frequency, while all minima were verified to have no imaginary frequencies. Free energy changes (ΔG) were computed by incorporating electronic energy (ΔE), zero-point vibrational energy (ZPE), thermal enthalpy (ΔH), entropy contribution ($-T\Delta S$), and dispersion corrections.

2. Synthesis

2.1 Synthesis of Ligand L1 = (*E*)-2-((2-(pyridin-2-yl)hydrazineylidene)methyl)pyridine

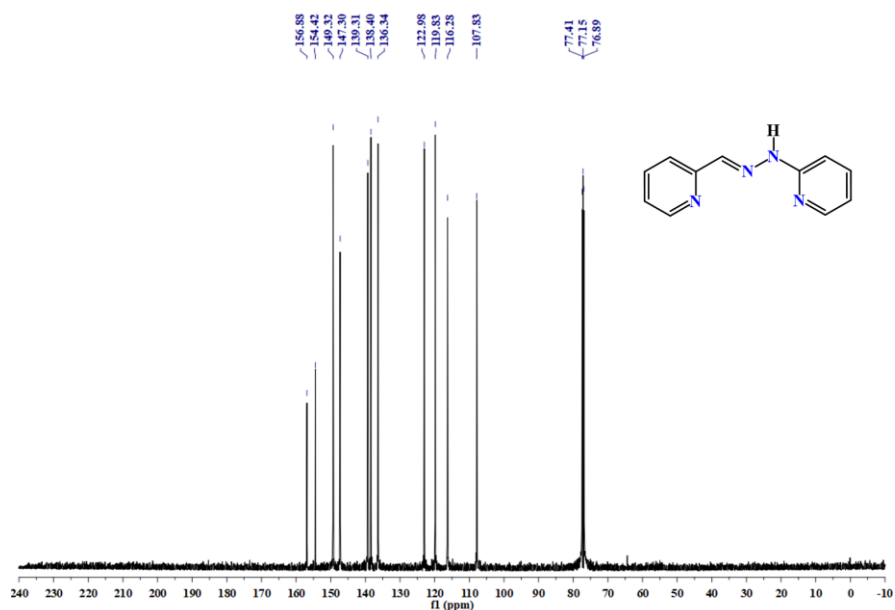
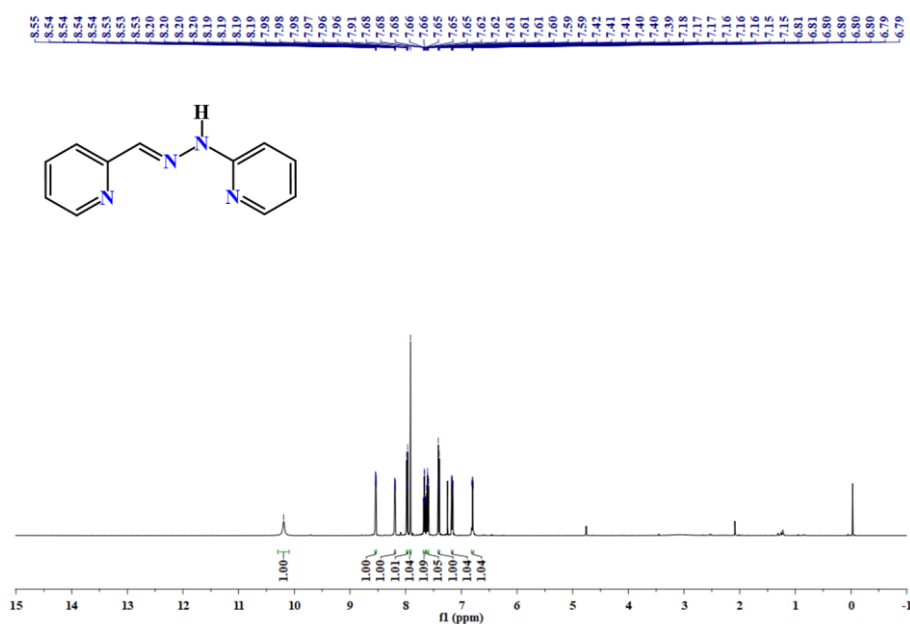
In a stirring solution of Pyridylhydrazine (436 mg, 4.0 mmol) in 10 mL methanol, a solution of pyridine-2-carboxaldehyde (428mg, 4 mmol) in 5 mL ethanol was added dropwise. The reaction mixture was stirred at room temperature for 6 hours, then filtered and recrystallized using ethanol. Yield - 697 mg (88%). Selected IR data (KBr, $\nu_{\text{max}}/\text{cm}^{-1}$): 3207 ($\nu_{\text{N-H}}$), 2988 ($\nu_{\text{C-H}}$), 1609 ($\nu_{\text{C=N}}$). UV-visible [DMSO, $\lambda_{\text{max}}/\text{nm}$ ($\epsilon/\text{M}^{-1}\text{cm}^{-1}$): 340 (43812.63). ¹H NMR (500 MHz, CDCl_3) δ 10.19 (s, 1H), 8.54 (ddd, J = 4.9, 1.7, 1.0 Hz, 1H), 8.19 (ddd, J = 5.0, 1.8, 0.8 Hz, 1H), 7.97 (dt, J = 8.0, 1.0 Hz, 1H), 7.91 (s, 1H), 7.68 – 7.64 (m, 1H), 7.61 (ddd, J = 8.5, 7.2, 1.8 Hz, 1H), 7.41 (dt, J = 8.4, 0.9 Hz, 1H), 7.16 (ddd, J = 7.4, 4.9, 1.2 Hz, 1H), 6.80 (ddd, J = 7.2, 5.0, 1.0 Hz, 1H). ¹³C{¹H} NMR (126 MHz, CDCl_3) δ 156.88, 154.42, 149.32, 147.30, 139.31, 138.40, 136.34, 122.98, 119.83, 116.28, 107.83.

2.2 Synthesis of Zn(II) complex C1.

To a stirring solution of the ligand L1 (99mg, 0.5 mmol) in 5 mL DCM, a solution of ZnCl₂ (68 mg, 0.5 mmol) in 2 mL MeOH was added dropwise. The reaction mixture was stirred at room temperature for 1 hour. A yellow product precipitates out and is filtered, washed with cold methanol, then air dried. Yield: = 150mg (90%), Selected IR data (KBr, $\nu_{\max}/\text{cm}^{-1}$): 3115 ($\nu_{\text{N-H}}$), 3019 ($\nu_{\text{C-H}}$), 1606 ($\nu_{\text{C=N}}$). UV-visible [DMSO, $\lambda_{\max}/\text{nm}$ ($\epsilon/\text{M}^{-1}\text{cm}^{-1}$): 340 (39380.53), 488 (3274.34). ¹H NMR (500 MHz, DMSO) δ 12.90 (s, 1H), 8.60 (s, 1H), 8.33 (s, 1H), 8.17 (d, J = 57.3 Hz, 2H), 7.90 (s, 2H), 7.67 (s, 1H), 7.09 (d, J = 62.0 Hz, 2H). ¹³C{¹H} NMR (126 MHz, DMSO) δ 151.56, 149.20, 147.39, 146.58, 141.57, 140.83, 134.74, 126.67, 125.45, 118.36, 110.21. Elemental analysis, C₁₁H₁₀Cl₂N₄Zn, calculated C, 39.50; H, 3.01; N, 16.75. Found: C, 40.22; H, 3.58; N, 17.36.

3. Characterization

3.1 Characterization of Ligand L1



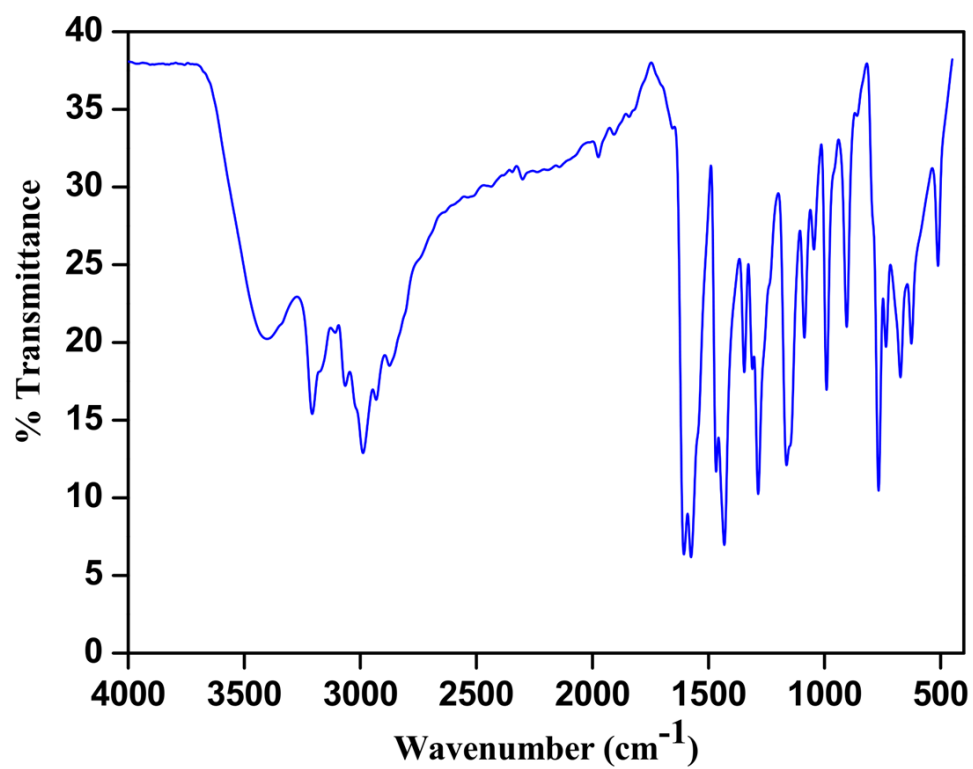


Figure S3 - IR spectrum of **L1**

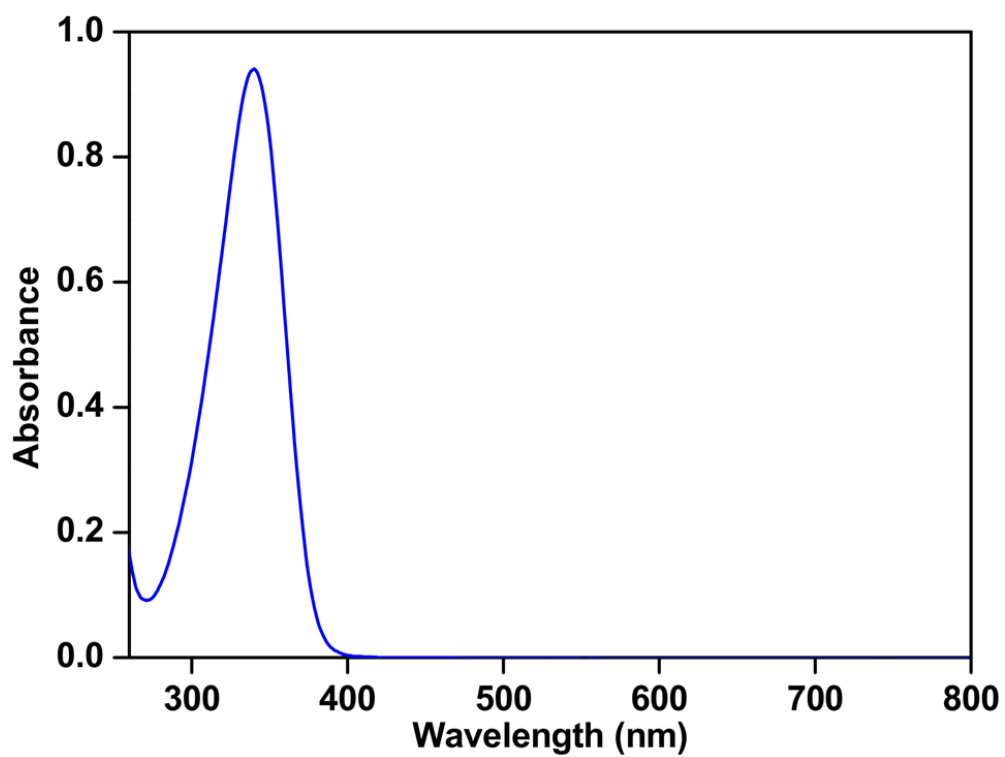


Figure S4 -UV-Visible spectrum of **L1**

User Spectrum Plot Report



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Inj. Vol. (ul)	2	IRM Status	Success		
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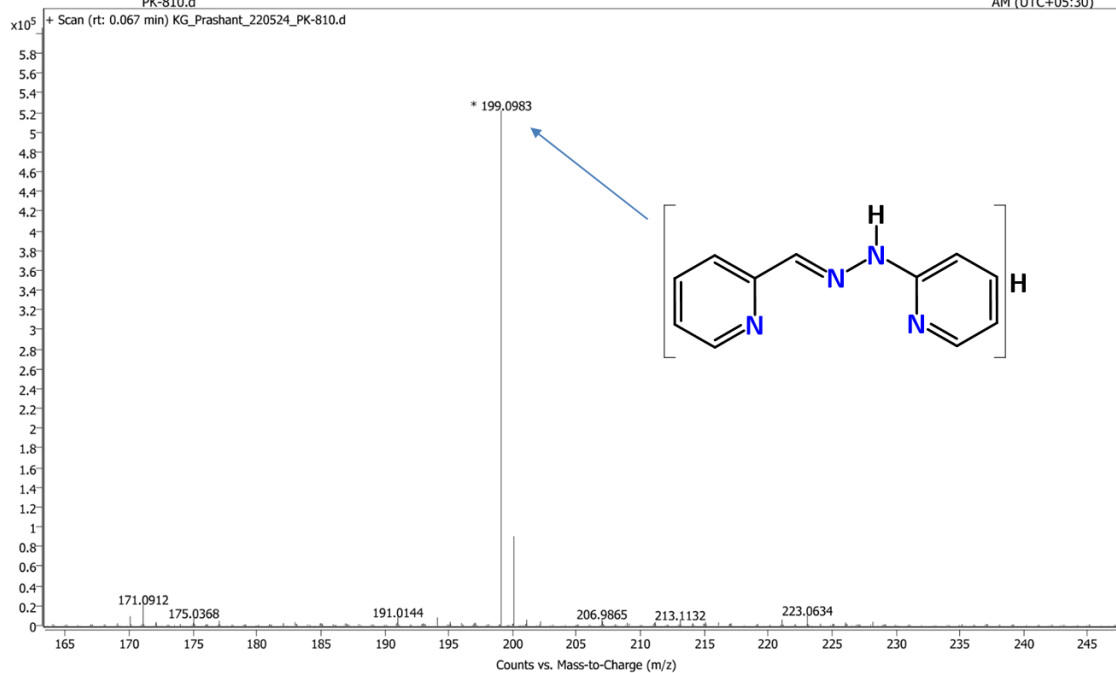


Figure S5 -HRMS spectrum of L1

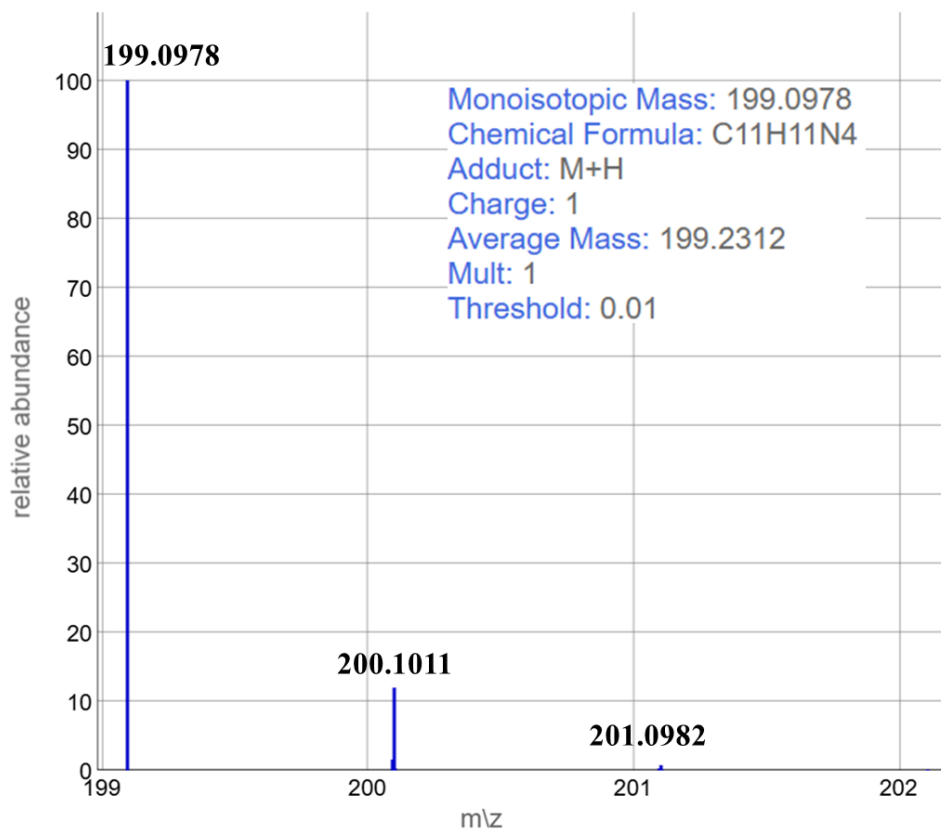
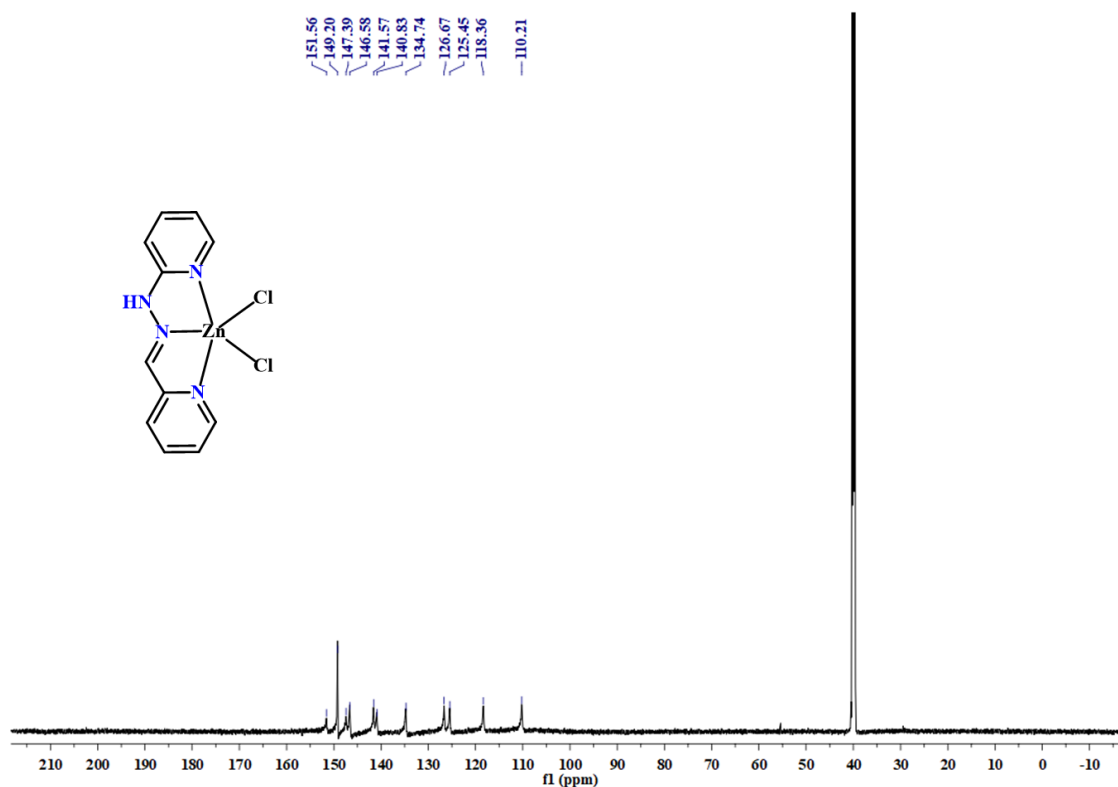
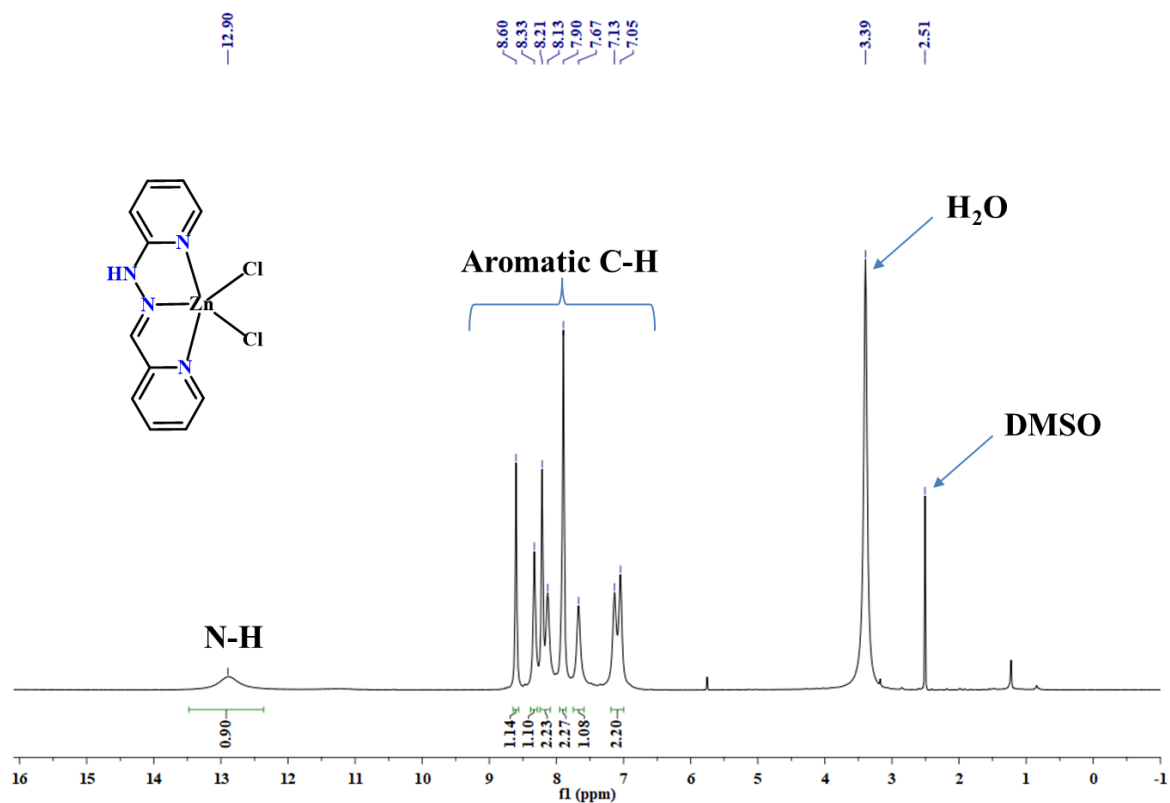


Figure S5b -Simulated HRMS spectrum of L1

3.2 Characterization of complexes $[\text{Zn(II)}(\text{Pyhzpy})\text{Cl}_2]$



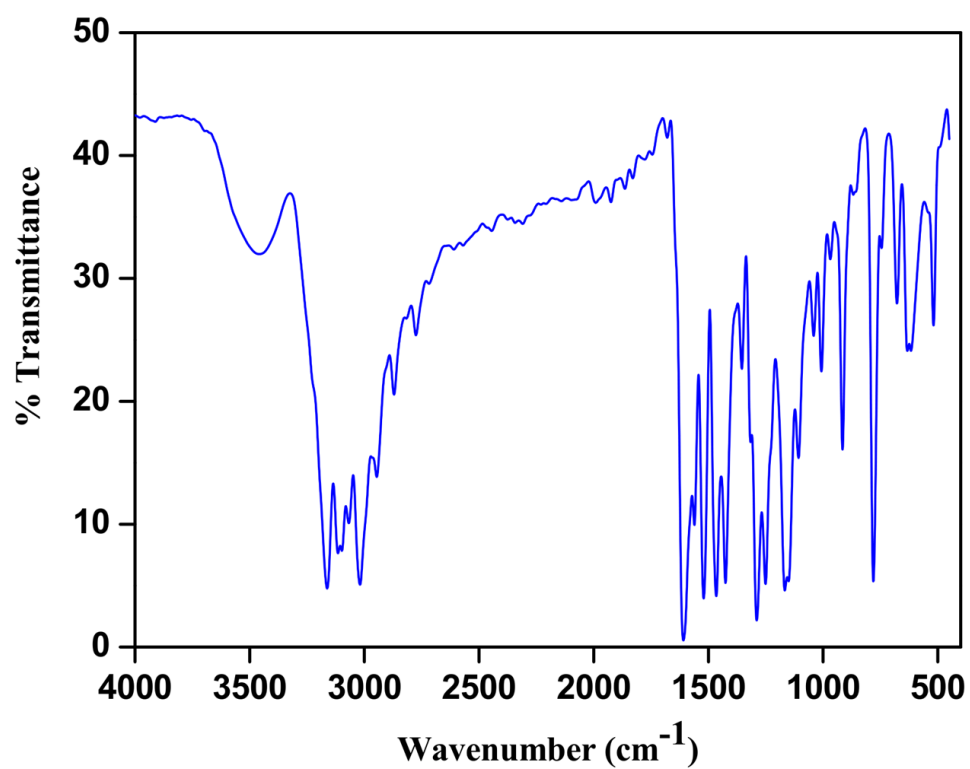


Figure S8 - IR spectrum of C1 [Zn(II)(L1)Cl₂]

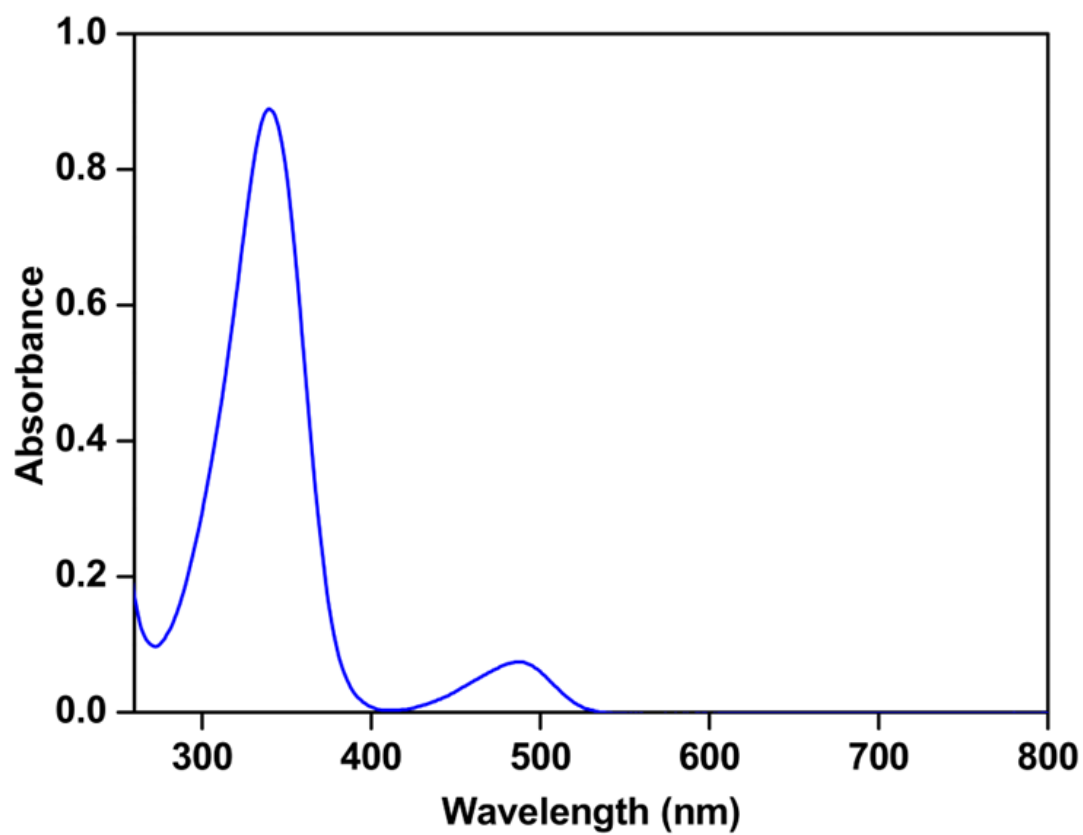


Figure S9 - UV visible spectrum of C1 [Zn(II)(L1)Cl₂]

Spectrum Plot Report

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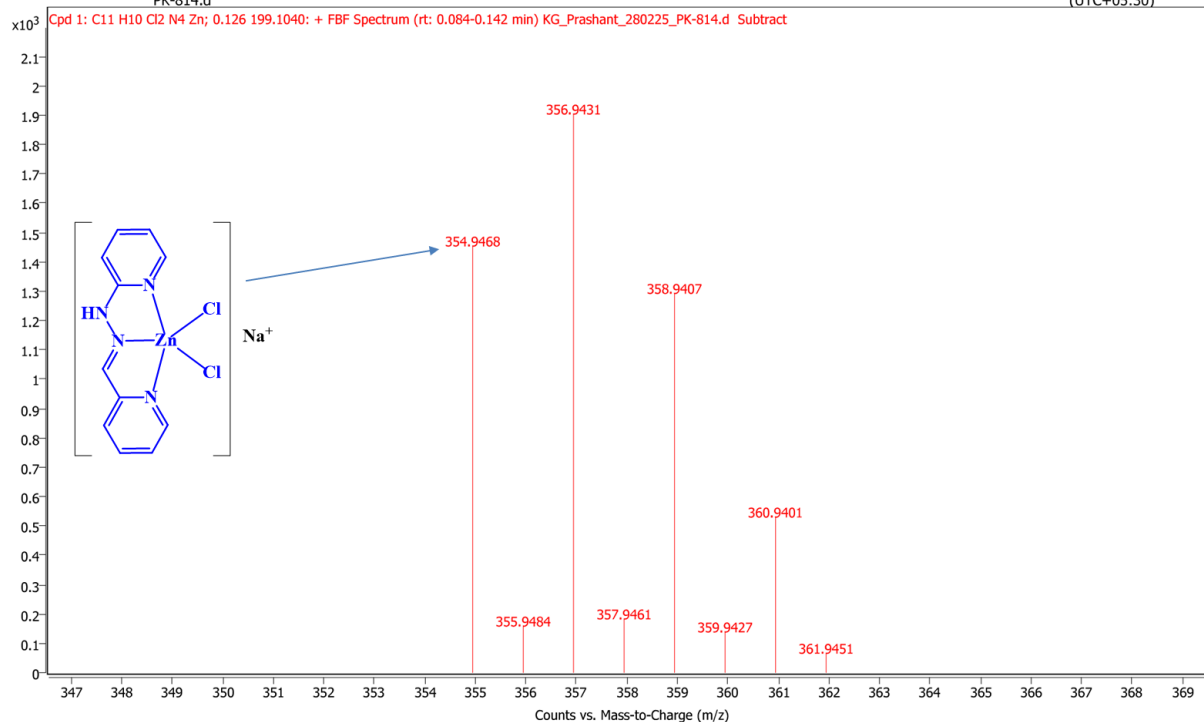


Figure S10 – HRMS spectrum of C1 [Zn(II)(L1)Cl₂]

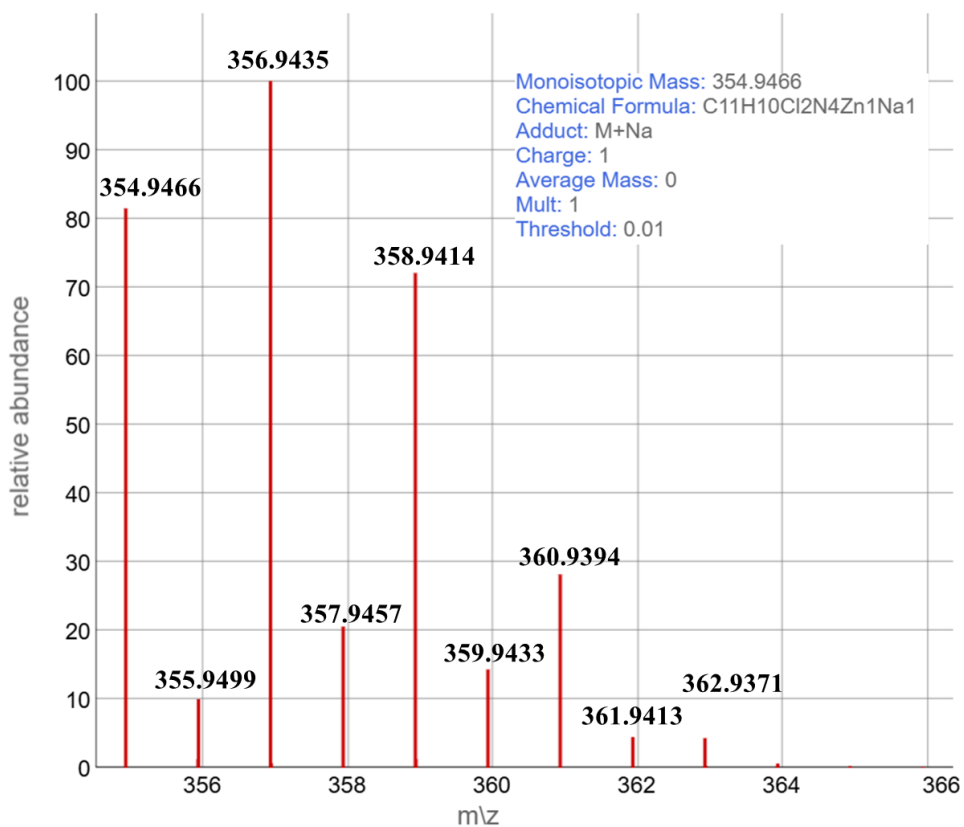


Figure S10b – Simulated HRMS spectrum of C1 [Zn(II)(L1)Cl₂]

4. Single-crystal XRD characterization

The molecular structure of complex C1 was elucidated through single-crystal X-ray diffraction (SC-XRD) studies. Single crystals suitable for X-ray diffraction were obtained by slow evaporation of a CH₂Cl₂/MeOH (1 : 1) solution. Complex C1 is found to crystallize in the orthorhombic crystal system characterized by the space group P-1. Figure 3 displays the ORTEP diagram of complex C1 and the selected bond lengths (in Å) adjacent to the zinc center include: Zn1–N2 = 2.2191(12) Å, Zn1–N3 = 2.1297(12) Å, Zn1–N4 = 2.1800(12) Å, Zn1–Cl1 = 2.2461(4) Å, and Zn1–Cl2 = 2.2763(4) Å. The geometric arrangement around the Zn(II) centre was analysed using the geometry index τ_5 , defined as $\tau_5 = (\beta - \alpha)/60$, where β and α denote the two largest coordination angles introduced by Reedijk and colleagues in 1984.⁵ The τ_5 value of 0.265 was computed for complex C1, suggesting a distorted square-pyramidal coordination geometry.

4.1 Crystal data collections and refinement parameters

Identification code	[Zn(L1)Cl ₂]
Empirical formula	C ₁₁ H ₁₀ Cl ₂ N ₄ Zn
Formula weight	334.50
Temperature/K	100.0
Crystal system	triclinic
Space group	P-1
a/Å	7.5445(2)
b/Å	7.6703(2)
c/Å	11.5765(3)
α /°	77.4140(10)
β /°	84.4980(10)
γ /°	83.2990(10)
Volume/Å ³	647.63(3)
Z	2
ρ_{calc} /cm ³	1.715
μ /mm ⁻¹	2.294
F(000)	336.0

Crystal size/mm ³	0.132 × 0.112 × 0.045
Radiation	MoK α (λ = 0.71073)
2 θ range for data collection/°	3.614 to 56.662
Index ranges	-10 ≤ h ≤ 10, -10 ≤ k ≤ 10, -15 ≤ l ≤ 15
Reflections collected	35430
Independent reflections	3235 [R_{int} = 0.0413, R_{sigma} = 0.0217]
Data/restraints/parameters	3235/0/167
Goodness-of-fit on F^2	1.055
Final R indexes [$I > 2\sigma(I)$]	R_1 = 0.0215, wR_2 = 0.0525
Final R indexes [all data]	R_1 = 0.0229, wR_2 = 0.0534

Table S1 – Crystal data collections and refinement parameters for **C1** [Zn(II)(L1)Cl₂]

4.2 Single crystal structure of **C1** [Zn(II)(L1)Cl₂]

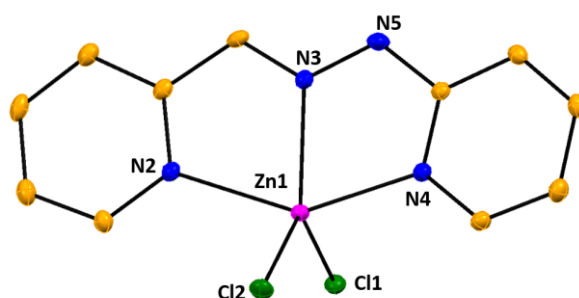


Figure S11 - ORTEP diagram (25% probability level) of **C1** [Zn(II)(L1)Cl₂]. Hydrogen atoms are omitted for clarity.

Bond	Bond Length/Å	Bond	Bond Length/Å
Zn1Cl1	2.2461(4)	N5C11	1.393(2)
Zn1Cl2	2.2763(4)	C1C2	1.391(2)
Zn1N2	2.2191(12)	C1C6	1.446(2)
Zn1N3	2.1297(12)	C2C3	1.386(2)
Zn1N4	2.1800(12)	C3C4	1.386(2)
N2C1	1.3479(19)	C4C4	1.390(2)
N2C5	1.337(2)	C7C8	1.378(2)
N3N5	1.3395(18)	C8C9	1.395(2)
N3C6	1.2983(19)	C9C10	1.374(2)
N4C7	1.3491(19)	C10C11	1.399(2)
N4C11	1.3358(19)		

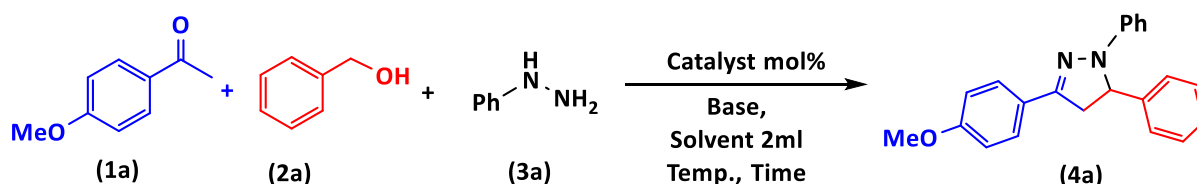
Table S2 – Selected bond lengths of (**C1**) [Zn(II)(L1)Cl₂]

Bond	Bond Angle (°)	Bond	Bond Angle (°)
Cl1Zn1Cl2	117.502(16)	C11N4C7	117.78(13)
N2Zn1Cl1	100.38(3)	N3N5C11	116.32(13)
N2Zn1Cl2	95.67(3)	N2C1C2	122.61(14)
N3Zn1Cl1	113.28(3)	N2C1C6	115.43(13)
N3Zn1Cl2	129.18(4)	C2C1C6	121.96(14)
N3Zn1N2	73.07(5)	C3C2C1	118.01(15)
N3Zn1N4	73.80(5)	C2C3C4	120.04(14)
N4Zn1Cl1	102.51(3)	C3C4C5	118.03(15)
N4Zn1Cl2	96.71(3)	N2C5C4	122.94(15)
N4Zn1N2	145.09(5)	N3C6C1	115.78(13)
C1N2Zn1	115.16(10)	N4C7C8	122.82(14)
C5N2Zn1	126.27(10)	C7C8C9	118.46(14)
C5N2C1	118.36(13)	C10C9C8	119.76(14)
N5N3Zn1	116.87(9)	C9C10C11	117.80(14)
C6N3Zn1	120.18(10)	N4C11N5	116.66(13)
C6N3N5	122.89(13)	N4C11C10	123.37(13)
C7N4Zn1	126.85(10)	N5C11C10	119.97(13)
C11N4Zn1	115.32(10)		

Table S3 – Selected bond angles of (C1) [Zn(II)(L1)Cl₂]

5. Optimization of reaction conditions

5.1. Optimization of reaction conditions for MCS of TriPyz



Entry	Catalyst Mol%	Base	Temperature	Time (hours)	Solvent	Yield (%)
1	L1 (2mol%)	^t BuOK 1.0eq	100	12	toluene	NR
2	L2 (2mol%)	^t BuOK 1.0eq	100	12	toluene	NR
3	ZnCl ₂ (2mol%)	^t BuOK 1.0eq	100	12	toluene	NR
4	C1 (2mol%)	^tBuOK 1.0eq	100	12	Toluene	86
5	C2 (2mol%)	^t BuOK 1.0eq	100	12	Toluene	64
6	C1 (2mol%)	No base	100	12	Toluene	NR
7	C1 (2mol%)	KOH 1.0eq	100	12	Toluene	67
8	C1 (2mol%)	NaOH 1.0eq	100	12	Toluene	55
9	C1 (2mol%)	CS ₂ CO ₃ 1.0eq	100	12	Toluene	38
10	C1 (2mol%)	Na ₂ CO ₃ 1.0eq	100	12	Toluene	NR
11	C1 (2mol%)	K ₂ CO ₃ 1.0eq	100	12	Toluene	NR
12	C1 (2mol%)	NaHCO ₃ 1.0eq	100	12	Toluene	NR
13	C1 (2mol%)	^t BuOK 1.0eq	100	12	Xylene	72
14	C1 (2mol%)	^t BuOK 1.0eq	100	12	Benzene	66
15	C1 (2mol%)	^t BuOK 1.0eq	100	12	DMF	NR
16	C1 (2mol%)	^t BuOK 1.0eq	100	12	DMSO	NR

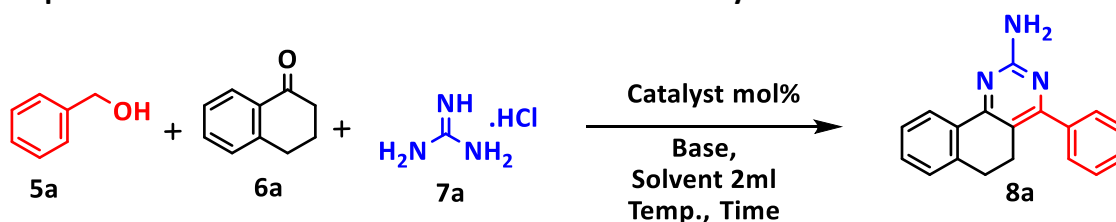
17	C1 (2mol%)	^t BuOK 1.0eq	100	12	1,4-dioxane	33
18	C1 (2mol%)	^t BuOK 1.0eq	100	12	<i>t</i> -amyl alcohol	NR
19	C1 (2mol%)	^t BuOK 0.5eq	100	12	Toluene	NR
20	C1 (2mol%)	^t BuOK 0.75eq	100	12	Toluene	56
21	C1 (2mol%)	^t BuOK 1.25eq	100	12	Toluene	86
22	C1 (2mol%)	^t BuOK 1.5eq	100	12	Toluene	86
23	C1 (1.0mol%)	^t BuOK 1.0eq	100	12	Toluene	46
24	C1 (1.5mol%)	^t BuOK 1.0eq	100	12	Toluene	73
25	C1 (2.5mol%)	^t BuOK 1.0eq	100	12	Toluene	86
26	C1 (3.0mol%)	^t BuOK 1.0eq	100	12	Toluene	86
27	C1 (2mol%)	^t BuOK 1.0eq	100	10	Toluene	63
28	C1 (2mol%)	^t BuOK 1.0eq	100	11	Toluene	71
29	C1 (2mol%)	^t BuOK 1.0eq	100	13	Toluene	86
30	C1 (2mol%)	^t BuOK 1.0eq	100	14	Toluene	86
31	C1 (2mol%)	^t BuOK 1.0eq	80	13	Toluene	58
32	C1 (2mol%)	^t BuOK 1.0eq	90	12	Toluene	77
33	C1 (2mol%)	^t BuOK 1.0eq	110	12	Toluene	86
34	C1 (2mol%)	^t BuOK 1.0eq	120	12	Toluene	86

Table S4: Reaction conditions and stoichiometry: 4-OMe-acetophenone (1a, 0.5 mmol), benzyl alcohol (2a, 0.5 mmol), Phenylhydrazine (3a, 0.6 mmol), catalyst loading (2-3 mol%), temperature: 80-120 °C; base: 0.5-1.5 equivalent, time 9-14 hour and Solvent (2ml) under N₂ atmosphere. Isolated yields after column chromatography.

5.2. General procedure for MCS of TriPyz

In a clean and dried 20 ml pressure tube filled with acetophenone derivatives (0.5 mmol), benzyl alcohol derivatives (0.5 mmol), phenylhydrazine (0.6 mmol), ^tBuOK (1.0 equivalent), and catalyst (C1 2.0 mol%). The vial was filled with 2.0 mL of dry toluene, and an inert atmosphere was created by purging N₂ into the reaction tube. The reaction mixture was heated in an oil bath at 100 °C for 12 hours. TLC was used to track the reaction's completion, and ethyl acetate (4 × 5 mL) was used to extract the reaction mixture. After being dried over anhydrous Na₂SO₄, the resultant organic layer of ethyl acetate was concentrated under reduced pressure. Hexane/ethyl acetate was used as an eluent in column chromatography on silica (100–200 mesh size) to purify the resultant organic mixture. ¹H and ¹³C{¹H} NMR spectroscopy was used to characterize the isolated products.

5.3. Optimization of reaction conditions for MCS of TetraPym



Entry	Catalyst Mol%	Base	Temperature	Time (hours)	Solvent	Yield (%)
1	L1 (2mol%)	^t BuOK 2.0eq	100	12	toluene	NR
2	L2 (2mol%)	^t BuOK 2.0eq	100	12	toluene	NR
3	ZnCl ₂ (2mol%)	^t BuOK 2.0eq	100	12	toluene	NR
4	C1 (2mol%)	^tBuOK 2.0eq	100	12	Toluene	88
5	C2 (2mol%)	^t BuOK 2.0eq	100	12	Toluene	68
6	C1 (2mol%)	No base	100	12	Toluene	NR
7	C1 (2mol%)	KOH 2.0eq	100	12	Toluene	71

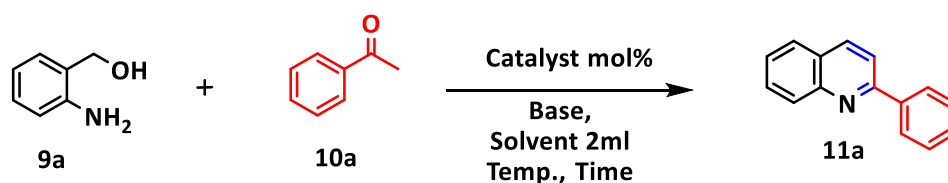
8	C1 (2mol%)	NaOH 2.0eq	100	12	Toluene	64
9	C1 (2mol%)	Na ₂ CO ₃ 2.0eq	100	12	Toluene	NR
10	C1 (2mol%)	K ₂ CO ₃ 2.0eq	100	12	Toluene	NR
11	C1 (2mol%)	Cs ₂ CO ₃ 2.0eq	100	12	Toluene	44
12	C1 (2mol%)	NaHCO ₃ 2.0eq	100	12	Toluene	NR
13	C1 (2mol%)	^t BuOK 2.0eq	100	12	Xylene	76
14	C1 (2mol%)	^t BuOK 2.0eq	100	12	Benzene	67
15	C1 (2mol%)	^t BuOK 2.0eq	100	12	DMF	NR
16	C1 (2mol%)	^t BuOK 2.0eq	100	12	DMSO	NR
17	C1 (2mol%)	^t BuOK 2.0eq	100	12	1,4-dioxane	32
18	C1 (2mol%)	^t BuOK 2.0eq	100	12	t-amyl alcohol	NR
19	C1 (2mol%)	^t BuOK 1.0eq	100	12	Toluene	NR
20	C1 (2mol%)	^t BuOK 1.5eq	100	12	Toluene	52
21	C1 (2mol%)	^t BuOK 2.0eq	100	12	Toluene	90
22	C1 (2mol%)	^t BuOK 2.5eq	100	12	Toluene	90
23	C1 (1.0mol%)	^t BuOK 2.0eq	100	12	Toluene	52
24	C1 (1.5mol%)	^t BuOK 2.0eq	100	12	Toluene	71
25	C1 (2.5mol%)	^t BuOK 2.0eq	100	12	Toluene	90
26	C1 (3.0mol%)	^t BuOK 2.0eq	100	12	Toluene	90
27	C1 (2mol%)	^t BuOK 2.0eq	100	10	Toluene	66
28	C1 (2mol%)	^t BuOK 2.0eq	100	11	Toluene	74
29	C1 (2mol%)	^t BuOK 2.0eq	100	13	Toluene	90
30	C1 (2mol%)	^t BuOK 2.0eq	100	14	Toluene	90
31	C1 (2mol%)	^t BuOK 2.0eq	80	13	Toluene	63
32	C1 (2mol%)	^t BuOK 2.0eq	90	12	Toluene	81
33	C1 (2mol%)	^t BuOK 2.0eq	110	12	Toluene	90
34	C1 (2mol%)	^t BuOK 2.0eq	120	12	Toluene	90

Table S5: Reaction conditions and stoichiometry: benzyl alcohol (5a, 0.5 mmol), α -tetralone (6a, 0.5 mmol), guanidine hydrochloride (7a, 0.6 mmol), catalyst loading (2-3 mol%), temperature: 80-120 °C; base: 1.0-2.5 equivalent, time 9-14 hour and Solvent (2ml) under N₂ atmosphere. Isolated yields after column chromatography.

5.4. General procedure for MCS of TetraPym

In a clean and dried 20 ml pressure tube filled with cyclic ketone derivatives (0.5 mmol), benzyl alcohol derivatives (0.5 mmol), amidine hydrochloride (0.6 mmol), ^tBuOK (2.0 equivalent), and catalyst (C1 2.0 mol%). The vial was filled with 2.0 mL of dry toluene, and an inert atmosphere was created by purging N₂ into the reaction tube. The reaction mixture was heated in an oil bath at 100 °C for 12 hours. TLC was used to track the reaction's completion, and ethyl acetate (4 × 5 mL) was used to extract the reaction mixture. After being dried over anhydrous Na₂SO₄, the resultant organic layer of ethyl acetate was concentrated under reduced pressure. Hexane/ethyl acetate was used as an eluent in column chromatography on silica (100–200 mesh size) to purify the resultant organic mixture. ¹H and ¹³C{¹H} NMR spectroscopy was used to characterize the isolated products.

5.5. Optimization of reaction conditions for MCS of TetraPym



Entry	Catalyst Mol%	Base	Temperature	Time (hours)	Solvent	Yield (%)
1	L1 (2mol%)	^t BuOK 1.0eq	100	10	toluene	NR
2	L2 (2mol%)	^t BuOK 1.0eq	100	10	toluene	NR
3	ZnCl ₂ (2mol%)	^t BuOK 1.0eq	100	10	toluene	NR
4	C1 (2mol%)	^tBuOK 1.0eq	100	10	Toluene	94
5	C2 (2mol%)	^t BuOK 1.0eq	100	10	Toluene	71
6	C1 (2mol%)	No base	100	10	Toluene	NR
7	C1 (2mol%)	KOH 1.0eq	100	10	Toluene	73
8	C1 (2mol%)	NaOH 1.0eq	100	10	Toluene	67
9	C1 (2mol%)	Na ₂ CO ₃ 1.0eq	100	10	Toluene	NR
10	C1 (2mol%)	K ₂ CO ₃ 1.0eq	100	10	Toluene	NR
11	C1 (2mol%)	Cs ₂ CO ₃ 1.0eq	100	10	Toluene	48
12	C1 (2mol%)	NaHCO ₃ 1.0eq	100	10	Toluene	NR
13	C1 (2mol%)	^t BuOK 1.0eq	100	10	Xylene	80
14	C1 (2mol%)	^t BuOK 1.0eq	100	10	Benzene	61
15	C1 (2mol%)	^t BuOK 1.0eq	100	10	DMF	NR
16	C1 (2mol%)	^t BuOK 1.0eq	100	10	DMSO	NR
17	C1 (2mol%)	^t BuOK 1.0eq	100	10	1,4-dioxane	42
18	C1 (2mol%)	^t BuOK 1.0eq	100	10	t-amyl alcohol	NR
19	C1 (2mol%)	^t BuOK 0.5eq	100	10	Toluene	47
20	C1 (2mol%)	^t BuOK 0.75eq	100	10	Toluene	75
21	C1 (2mol%)	^t BuOK 1.25eq	100	10	Toluene	94
22	C1 (2mol%)	^t BuOK 1.5eq	100	10	Toluene	94
23	C1 (1.0mol%)	^t BuOK 1.0eq	100	10	Toluene	56
24	C1 (1.5mol%)	^t BuOK 1.0eq	100	10	Toluene	77
25	C1 (2.5mol%)	^t BuOK 1.0eq	100	10	Toluene	94
26	C1 (3.0mol%)	^t BuOK 1.0eq	100	10	Toluene	94
27	C1 (2mol%)	^t BuOK 1.0eq	100	8	Toluene	68
28	C1 (2mol%)	^t BuOK 1.0eq	100	9	Toluene	81
29	C1 (2mol%)	^t BuOK 1.0eq	100	11	Toluene	94
30	C1 (2mol%)	^t BuOK 1.0eq	100	12	Toluene	94
31	C1 (2mol%)	^t BuOK 1.0eq	80	10	Toluene	53
32	C1 (2mol%)	^t BuOK 1.0eq	90	10	Toluene	85
33	C1 (2mol%)	^t BuOK 1.0eq	110	10	Toluene	94
34	C1 (2mol%)	^t BuOK 1.0eq	120	10	Toluene	94

Table S6: Reaction conditions and stoichiometry: 2-aminobenzyl alcohol (9a, 0.5 mmol), acetophenone (10a, 0.5 mmol), catalyst loading (2-3 mol%), temperature: 80-120 °C; base: 0.5-1.5 equivalent, time 8-12 hour and Solvent (2ml) under N₂ atmosphere. Isolated yields after column chromatography.

5.6. General procedure for quinoline synthesis

In a clean and dried 20 ml pressure tube filled with 2-amino benzyl alcohol derivative (0.5 mmol), acetophenone derivative (0.5 mmol), ^tBuOK (1.0 equivalent) and catalyst (C1 1.0 mol%). The charged vial was filled with 2.0 mL of dry toluene, and an inert atmosphere was created by purging N₂ to the reaction tube. The reaction mixture was heated in an oil bath at 100°C for the next 10 hours. TLC was used to track the reaction's completion, and ethyl acetate (4 × 5 mL) was used to extract the reaction mixture. After being dried over anhydrous Na₂SO₄, the resultant organic layer of ethyl acetate was concentrated under reduced pressure. Hexane/ethyl acetate was used as an eluent in column chromatography on silica (100–200 mesh size) to purify the resultant organic mixture. ¹H and ¹³C{¹H} NMR spectroscopy was used to characterize the isolated products.

6. Controlled experiments for quinoline synthesis

A few control experiments related to the synthesis of quinoline were performed, as outlined below. A reaction was performed using 2-aminobenzyl alcohol (9a), and 4-Me-acetophenone (11a) along with radical scavengers TEMPO and BHT. This results in no significant change in the yield of the respective quinoline product 11g, explaining a non-radical pathway for quinoline synthesis. The homogeneous character of the reaction was confirmed by the mercury drop test.

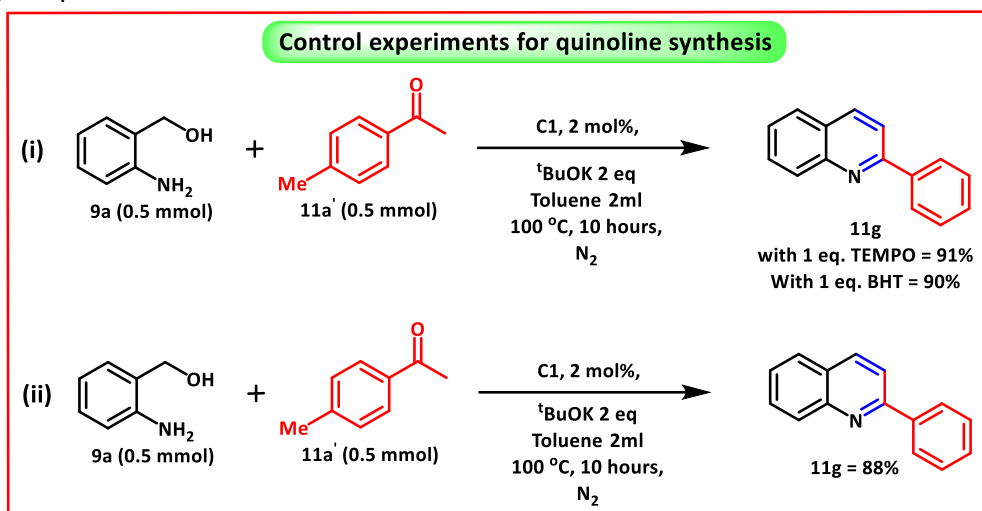


Figure S12- Controlled experiments for quinoline synthesis

7. Characterization of intermediates and side product (H₂) by GC-MS

7.1 Detection of H₂ via styrene reduction in the presence of 10% Pd-C and in situ formed α,β -unsaturated carbonyl compound

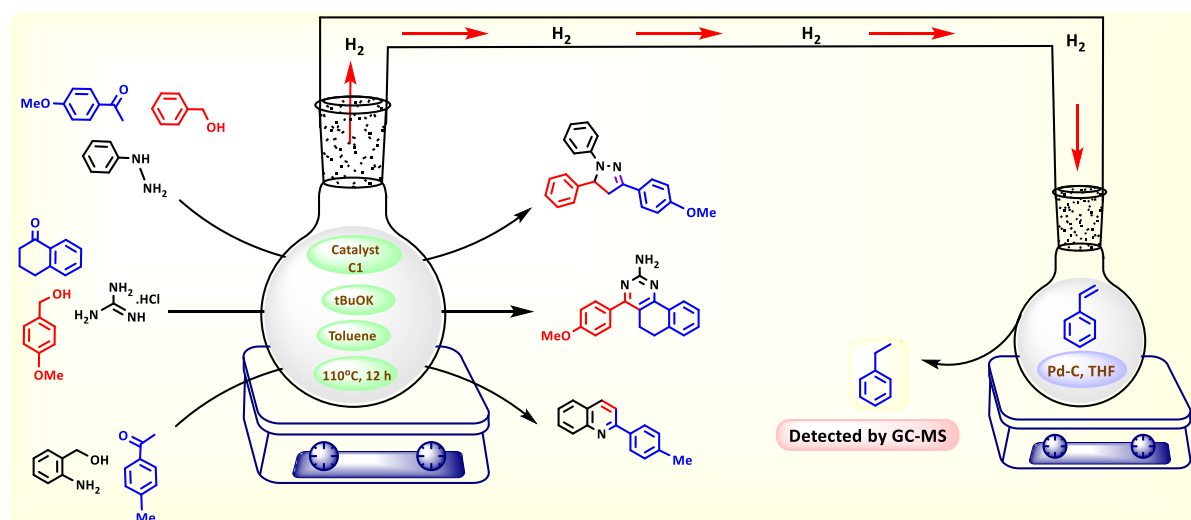


Figure S13- Diagrammatic representation of the experiment for the detection of H₂

The two 20 ml round bottom flask was connected through U-shaped tube the first flask was charged with catalyst base and substrates and second flask was charged with styrene, Pd-C

in THF at room temperature. The in situ produced H_2 travelled from first flask to second flask and styrene is reduced to ethylbenzene by H_2 in presence of Pd-C which is detected by GC-MS analysis.

Stoichiometry of reaction for MCS of pyrazoline: In the first round, bottom flask 4-methoxy acetophenone (0.5 mmol), benzyl alcohol (0.5 mmol), phenylhydrazine (0.6 mmol), $tBuOK$ (1.0 equivalent), and catalyst (C1 2.0 mol%). The vial was filled with 2.0 ml of toluene, and an inert atmosphere was created by purging N_2 into the reaction tube. The reaction mixture was heated in an oil bath at $100\text{ }^{\circ}C$ for the next 12 hours. In the second-round bottom flask, styrene 0.5 mmol, Pd-C 10 mol% in THF 10 ml.

Stoichiometry of reaction for MCS of pyrimidine: In the first round, bottom flask α -tetralone (0.5 mmol), 4-methoxy benzyl alcohol (0.5 mmol), guanidine hydrochloride (0.6 mmol), $tBuOK$ (2.0 equivalent), and catalyst (C1, 2.0 mol%). The vial was filled with 2.0 ml of toluene, and an inert atmosphere was created by purging N_2 into the reaction tube. The reaction mixture was heated in an oil bath at $100\text{ }^{\circ}C$ for the next 12 hours. In the second round, bottom flask, styrene 0.5 mmol, Pd-C 10 mol% in THF 10 ml.

Stoichiometry of reaction for quinoline synthesis: In the first-round bottom flask 2-aminobenzyl alcohol (0.5 mmol), 4-methyl acetophenone (0.5 mmol), $tBuOK$ (1.0 equivalent), and catalyst (C1 2.0 mol%). The vial was filled with 2.0 ml of toluene, and an inert atmosphere was created by purging N_2 into the reaction tube. The reaction mixture was heated in an oil bath at $100\text{ }^{\circ}C$ for the next 10 hours. In the second-round bottom flask, styrene 0.5 mmol, Pd-C 10 mol% in THF 10 ml.

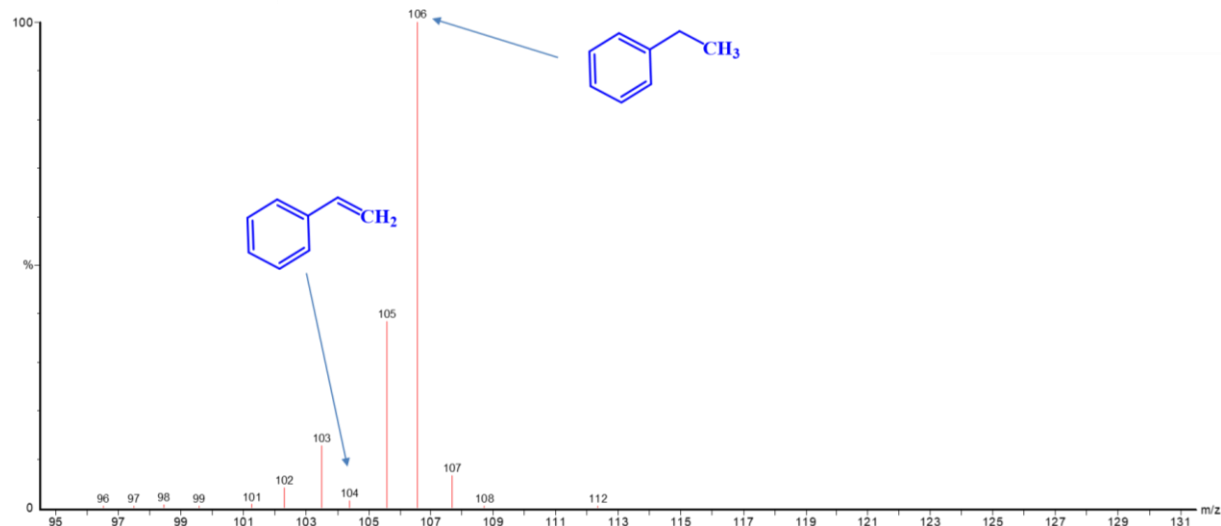


Figure S14- GC–MS spectrum of Styrene and ethylbenzene

7.2 Detection of in-situ formed α,β -unsaturated carbonyl compound during MCS of Tripyz

In a 10 ml pressure tube 4-methoxy acetophenone (0.5 mmol), benzyl alcohol (0.5 mmol), Phenylhydrazine (0.6 mmol), $tBuOK$ (1.0 equivalent), and catalyst (C1, 2 mol%) were taken in 2 ml toluene under an inert atmosphere of N_2 . The tube was sealed, heated at $100\text{ }^{\circ}C$ for the next 4 hours after the reaction mixture was cooled at room temperature, and extracted with

ethyl acetate and water. Formation of α,β -unsaturated carbonyl compound (*E*)-1-(4-methoxyphenyl)-3-phenylprop-2-en-1-one was then detected by HRMS analysis.

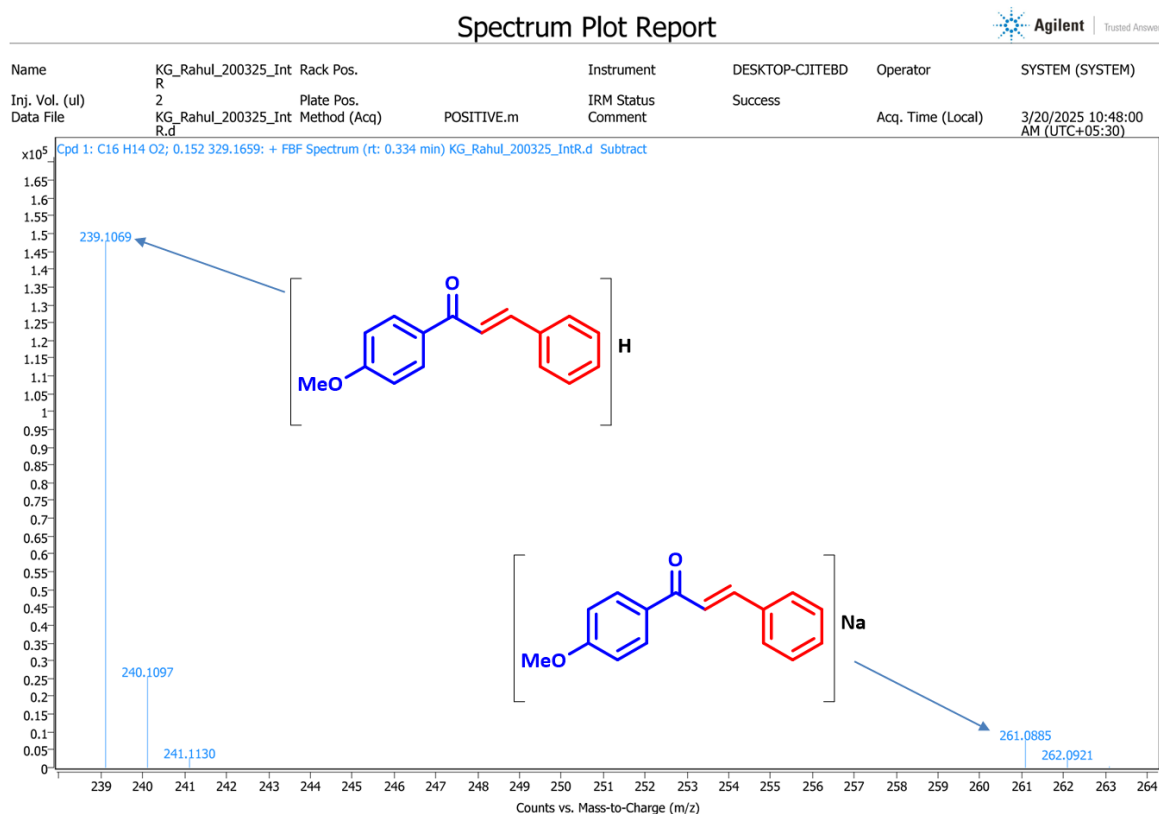


Figure S15- HRMS spectrum of (*E*)-1-(4-methoxyphenyl)-3-phenylprop-2-en-1-one

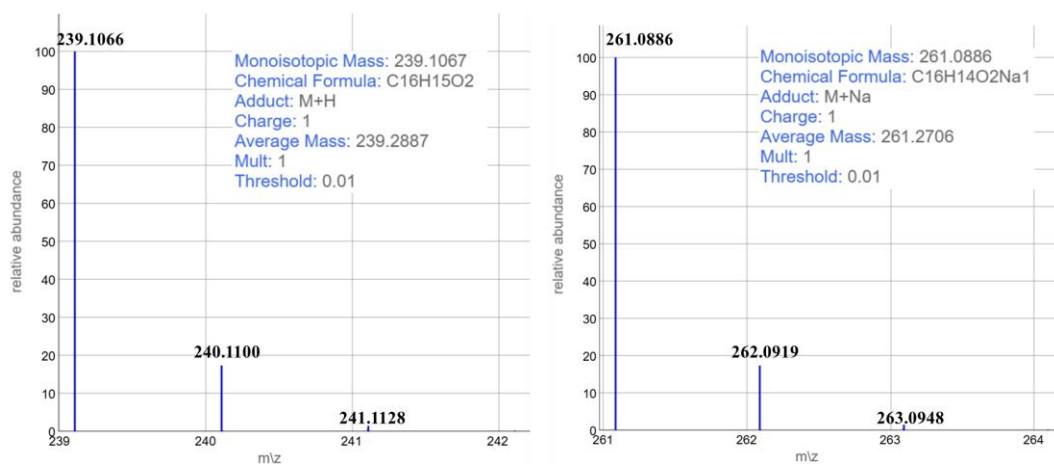


Figure S16- Simulated HRMS spectrum of (*E*)-1-(4-methoxyphenyl)-3-phenylprop-2-en-1-one

7.3 Detection of in-situ formed α,β -unsaturated carbonyl compound during MCS of TetraPym

In a 10 ml pressure tube 4-methoxy acetophenone (0.5 mmol), 4-methoxy benzyl alcohol (0.5 mmol), Phenylhydrazine (0.6 mmol), *t*BuOK (1.0 equivalent), and catalyst (C1, 2mol%) were taken in 2 ml toluene under an inert atmosphere of N₂. The tube was sealed, heated at 100

°C for the next 4 hours, after that reaction mixture was cooled at room temperature, extracted with ethyl acetate and water. Formation of α,β -unsaturated carbonyl compound (*E*)-2-(4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-one and intermediate 1-((*E*)-2-((*E*)-4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-ylidene)guanidine were then detected by HRMS analysis.

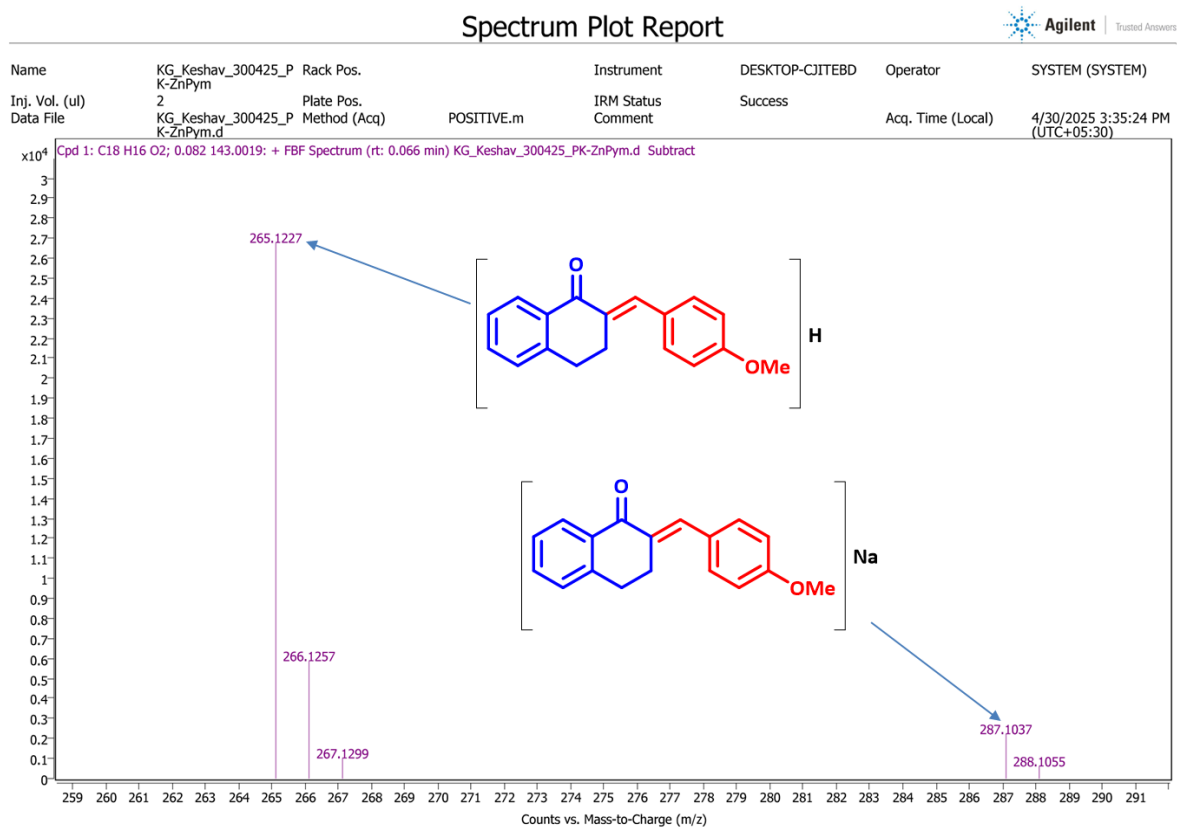


Figure S17- GC–MS spectrum of (*E*)-2-(4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-one

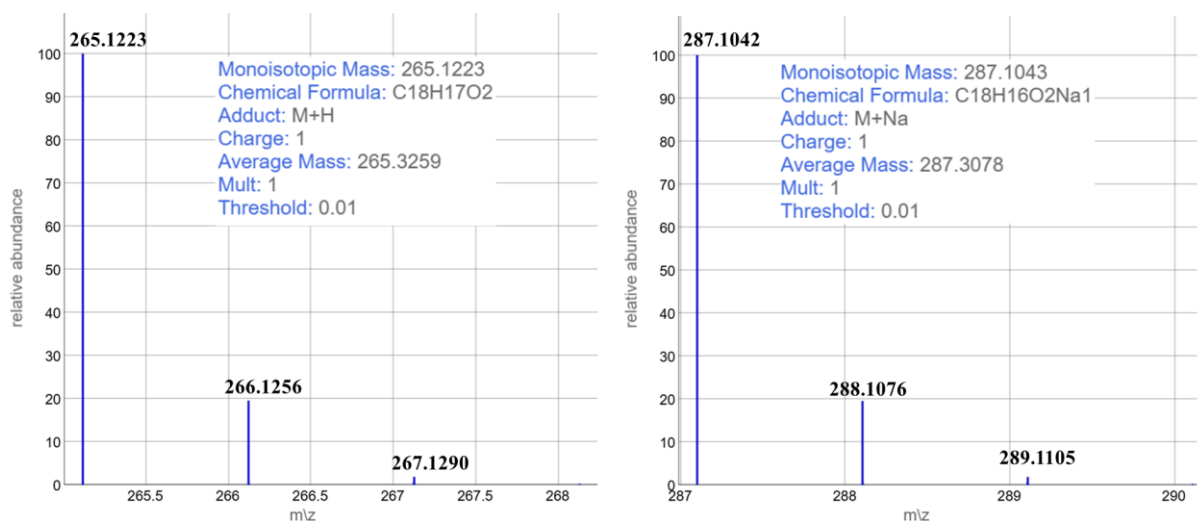


Figure S17b- Simulated HRMS spectrum of (*E*)-2-(4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-one

Spectrum Plot Report

Name	KG_Keshav_300425_P	Rack Pos.		Instrument	DESKTOP-CJITEBD	Operator	SYSTEM (SYSTEM)
Inj. Vol. (ul)	2	Plate Pos.		IRM Status	Success		
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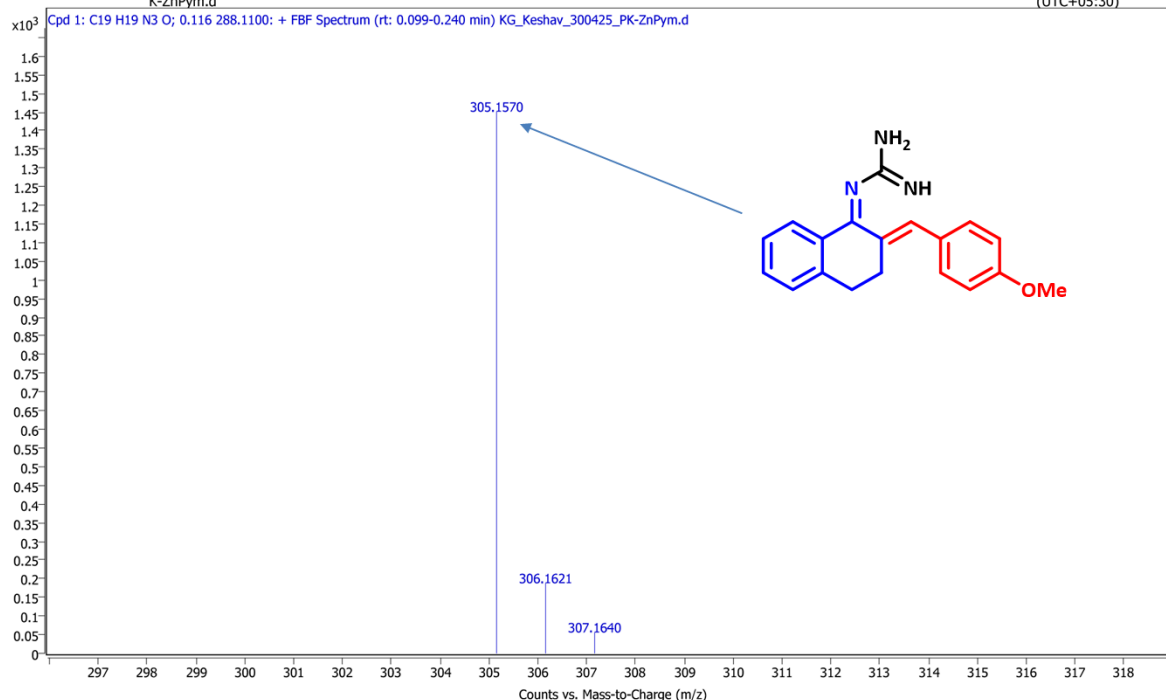


Figure S18- GC-MS spectrum of 1-((*E*)-2-((*E*)-4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-ylidene)guanidine

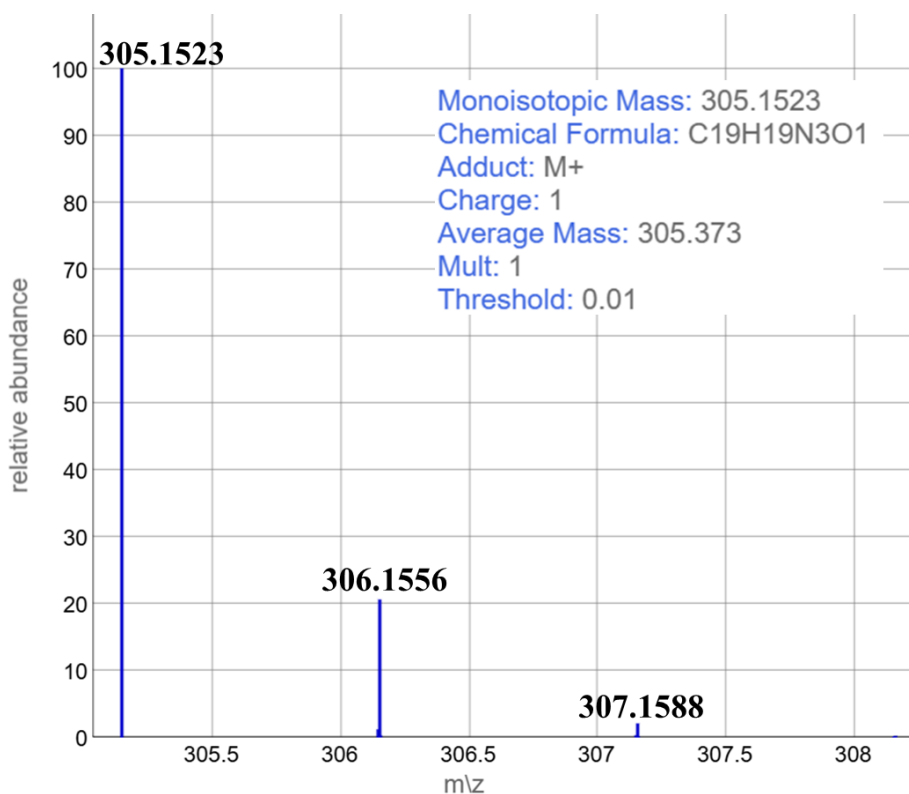


Figure S18b- GC-MS spectrum of 1-((*E*)-2-((*E*)-4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-ylidene)guanidine

7.4 Detection of in-situ formed α,β unsaturated carbonyl compound during Quinoline synthesis

In a 10 ml pressure tube, 4-methoxy acetophenone (0.5 mmol), 4-methoxy benzyl alcohol (0.5 mmol), Phenylhydrazine (0.6 mmol), t BuOK (1.0 equivalent), and catalyst (C1, 2 mol%) were taken in 2 ml toluene under an inert atmosphere of N_2 . The tube was sealed, heated at 100 °C for the next 4 hours after the reaction mixture was cooled at room temperature, extracted with ethyl acetate and water. Formation of α,β -unsaturated carbonyl compound (*E*)-3-(2-aminophenyl)-1-(*p*-tolyl)prop-2-en-1-one was detected by HRMS analysis.

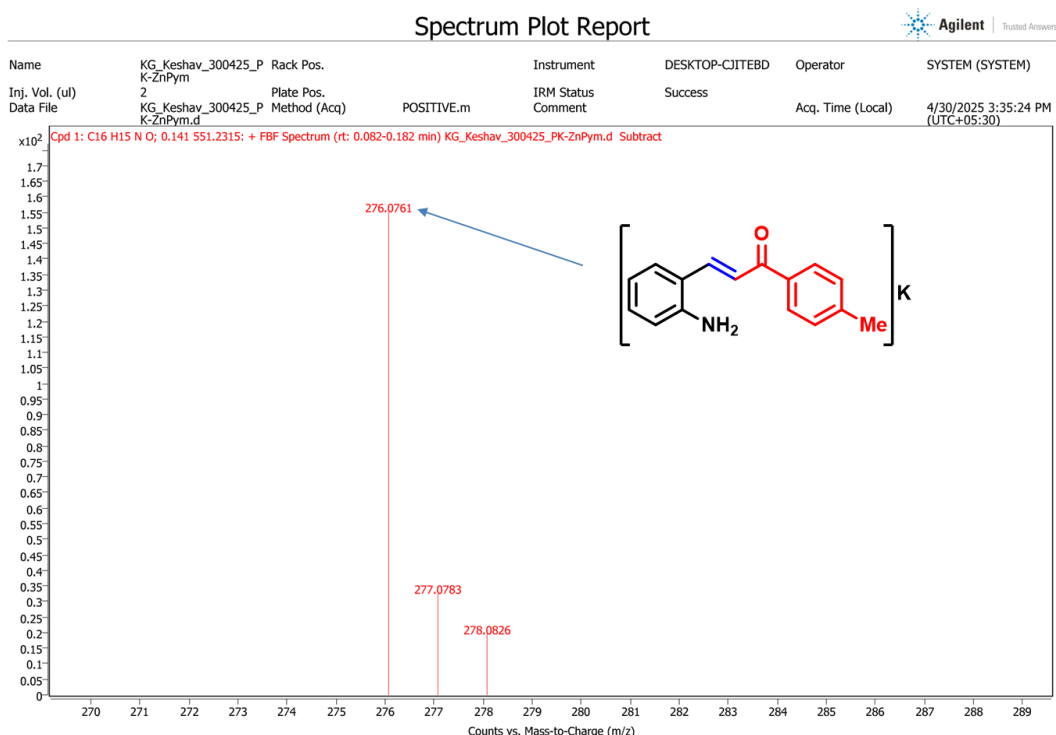


Figure S19- GC–MS spectrum of (*E*)-3-(2-aminophenyl)-1-(*p*-tolyl)prop-2-en-1-one

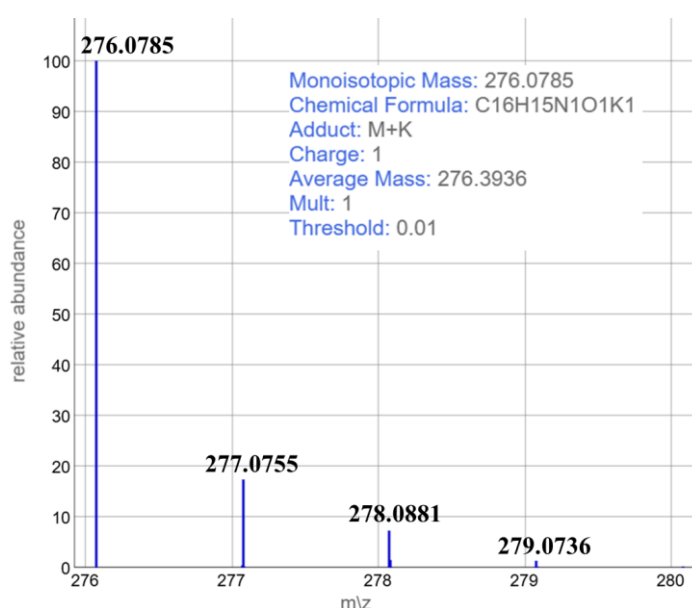


Figure S19b- HRMS spectrum of (*E*)-3-(2-aminophenyl)-1-(*p*-tolyl)prop-2-en-1-one

7.5 Characterization of Zn(II) intermediates involved during catalytic cycle

In a 10 ml pressure tube, benzyl alcohol (0.5 mmol), ^tBuOK (1.0 equivalent), and catalyst (C1 2mol%) were taken in 2 ml toluene under an inert atmosphere of N₂. The tube was sealed, heated at 100 °C for the next 4 hours, after that reaction mixture was cooled to room temperature, and the crude reaction mixture was analysed by HRMS studies. Zn(II) intermediates **C1A** and **C2A** were characterized by HRMS studies.

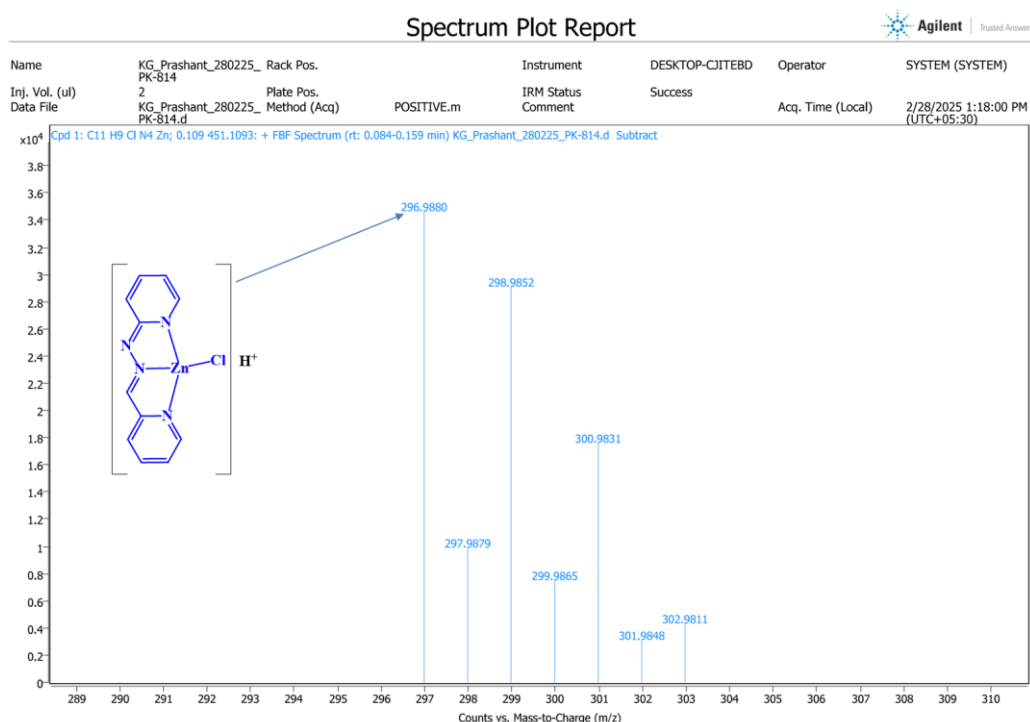


Figure S20- HRMS spectrum intermediate C1A

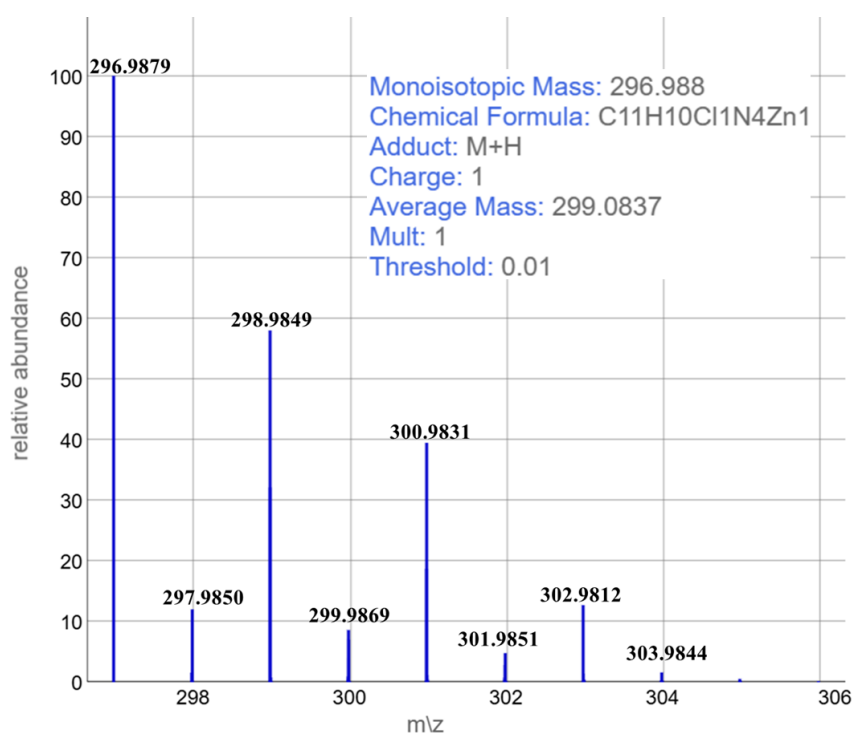


Figure S20b- Simulated HRMS spectrum intermediate C1A

Spectrum Plot Report

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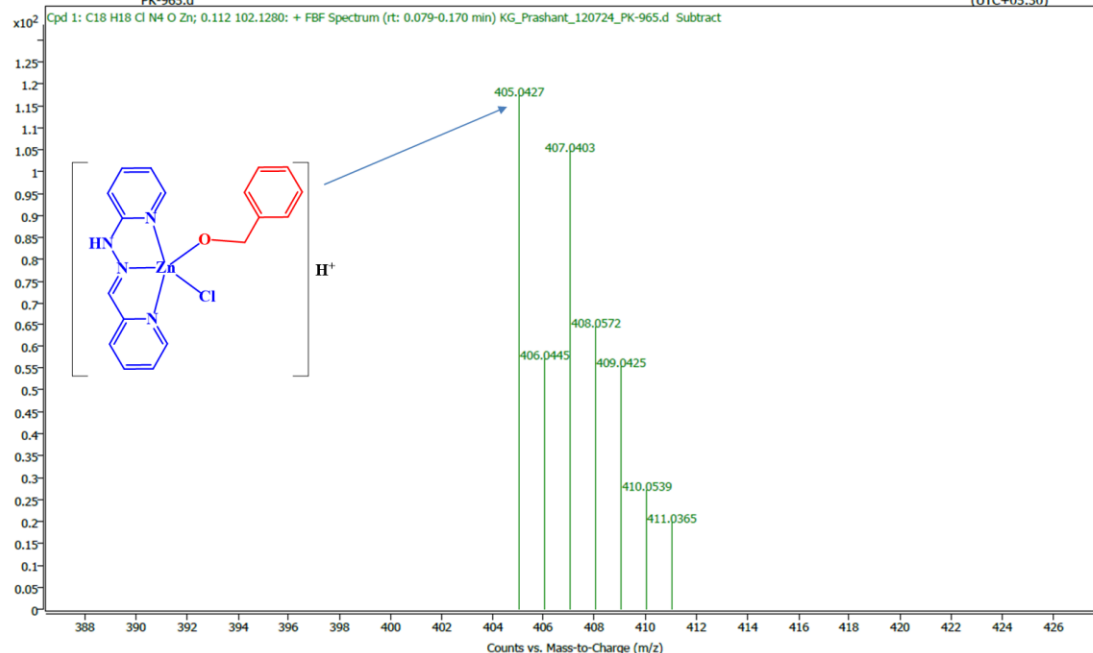


Figure S21- HRMS spectrum of C1B

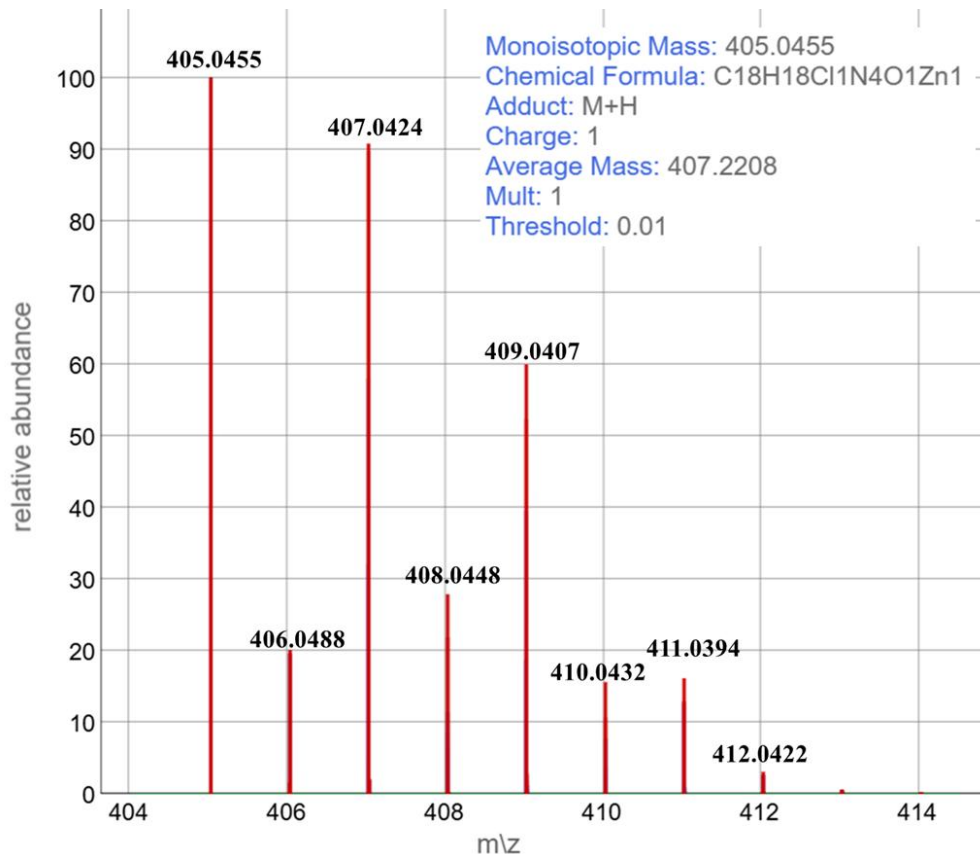


Figure S21b- Simulated HRMS spectrum of C1B

8. Proposed reaction pathway for acceptorless dehydrogenation of alcohol by C1

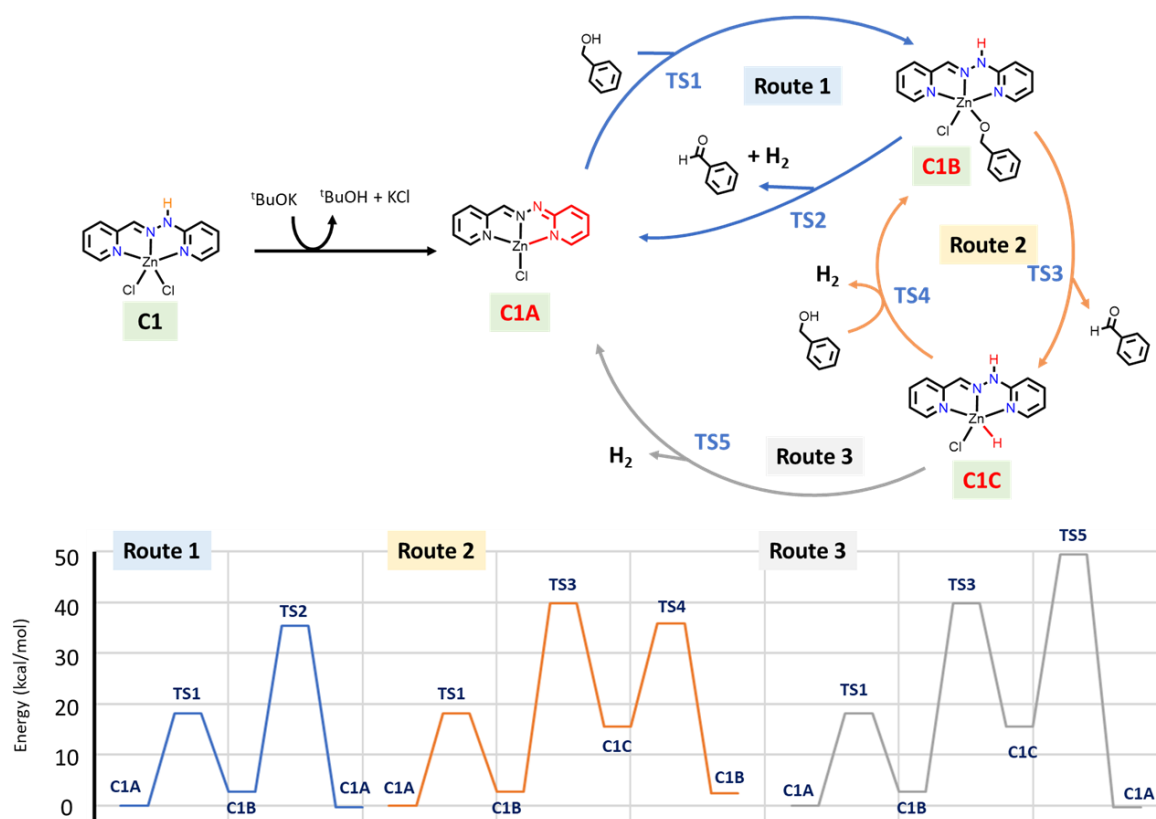


Figure S22- Proposed reaction pathway for AD of alcohol catalysed by C1

Route 1

Step	hartree	kcal/mol
C1A + <chem>c1ccccc1CO</chem> → C1B	0.00433	2.72
C1B → C1A + H_2 + <chem>c1ccccc1=O</chem>	-0.00489	-3.07

Table S7. Gibbs free energy change in each step of Route 1.

Route 2

Step	hartree	kcal/mol
C1A	0.00433	2.72
C1B	0.00204	12.8
C1C	-0.0209	-13.1

Table S8. Gibbs free energy change in each step of Route 2.

Route 3

Step	hartree	kcal/mol
C1A	0.00433	2.72
C1B	0.00204	12.8
C1C	-0.0252	-15.8

Table S9. Gibbs free energy change in each step of Route 3.

9. Coordinates for optimized structures

Table S10. Coordinates of C1A



26

E -2882.243760994419

Zn	-0.25034321665846	-0.59464106211889	1.50313092608157
N	1.49633710451025	-0.11188022618379	0.44568937232749
Cl	-0.47596133708767	0.97415731251488	3.07738040814674
N	-1.96494069637512	-1.85155110432081	1.87123684744615
C	2.32179326883004	0.91833671067893	0.69331877196537
C	1.73853425373775	-0.96170821308475	-0.58783446591567
C	2.87519798756421	-0.76093144051413	-1.41005892989037
C	3.45050506889353	1.17006509782822	-0.07278306811421
C	3.72295055817843	0.30092198254860	-1.14841114021351
N	0.90178268290450	-2.01711852100127	-0.85221469853489
H	3.04605637461403	-1.45795583552079	-2.23192289067801
H	2.04548475313569	1.55122679951743	1.54251806900898
H	4.09875208577562	2.01636873493697	0.15886491024150
H	4.60254240446916	0.46430301706874	-1.77701030306241
C	-2.87550282839690	-1.69185251029010	2.83448498072642
C	-2.06696019170207	-2.87664427856968	0.98696532768105
C	-3.13587666044390	-3.78753506301502	1.07903736748959
C	-3.96047747614937	-2.55542096588270	2.98526999796502
C	-4.08366331234600	-3.62163597238160	2.08466215486153
H	-3.20712359383458	-4.60843301616327	0.36240018965076
H	-2.71660857212182	-0.83711850822926	3.49942145116577

H	-4.68487914604427	-2.39414975273066	3.78510864331111
H	-4.91970400856472	-4.32085395467404	2.16880670726769
N	-0.10138791899965	-2.06322298841366	-0.01061593899077
C	-1.03509433970455	-2.97449167644615	-0.03037720309976
H	-1.04607224418411	-3.78053656555309	-0.77271648683718

Table S11. Coordinates of C1B



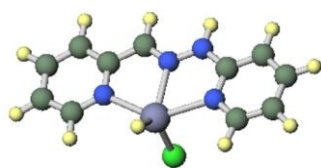
42

E -3228.611436577480

Zn	-0.60397701744628	-0.20890841368948	1.28437338783193
N	1.47452602168531	0.06300099631223	0.48457521806838
O	-1.70143894912903	0.52669252401839	-0.09680829698412
Cl	-0.30253255652329	0.73680593750266	3.32079175333169
N	-1.99153678088950	-1.86303387861711	1.87446240214328
C	2.19855408642831	1.17024610318623	0.69111033989369
C	1.88472690361949	-0.82535203727265	-0.42239002343460
C	3.04214016566170	-0.64339980130451	-1.19392454968759
C	3.36447030656754	1.44055314579523	-0.01659996935021
C	3.78257647598060	0.51409949475784	-0.97996616355887
N	1.09837065143927	-1.96774560477571	-0.56818587455287
H	3.34103247369182	-1.39063260416330	-1.93190772101193
H	1.81199801918080	1.84911945165256	1.45456639364367
H	3.92282848386077	2.35751298731553	0.17290210603729
H	4.68775305194682	0.69443392856539	-1.56491124538672

C	-2.98029045924179	-1.71438654790362	2.75300440239832
C	-1.90613864526594	-2.99453272420999	1.14711006517162
C	-2.84221317159369	-4.02681194993067	1.28249951468044
C	-3.95799894687720	-2.69587035012395	2.95101857324427
C	-3.88401420018052	-3.86944974647436	2.19889943159602
H	-2.74856037477152	-4.93279331349821	0.67986121960115
H	-2.98080710065832	-0.77759547044086	3.31854612829635
H	-4.75451127581528	-2.53542916322657	3.67982113952318
H	-4.62969083631909	-4.65829323981539	2.32490725059004
N	0.03218398718651	-2.08770953727627	0.20128956153017
H	1.35821004975759	-2.69029242555587	-1.23844315927620
C	-0.77727253585524	-3.08312096803015	0.21252538565932
H	-0.66141092840519	-3.96718017420437	-0.42996566813037
C	-1.20913110833192	0.80470086590208	-1.35200736881490
H	-2.05058860642907	1.04373182207186	-2.04622364776854
H	-0.69592690319764	-0.06409873770243	-1.83542657772115
C	-0.24223418660629	1.98250353770178	-1.42015781811158
C	-0.30188536054340	3.00082782028006	-0.45943670467787
C	0.71638495280863	2.07640369367205	-2.43840308623112
C	1.59389880813316	3.16224258417822	-2.50069973248931
C	0.57245473556634	4.08987598556005	-0.51756532370068
C	1.52476745287661	4.17575963619515	-1.53868156254838
H	0.78428715003135	1.27925936826473	-3.18619639010459
H	2.34174672096548	3.21408213973400	-3.29750435453056
H	-1.03679326911386	2.91494711456943	0.34366477577499
H	0.51325490493195	4.87517092151917	0.24177296021900
H	2.21278981087399	5.02466462946088	-1.58229677116273

Table S12. Coordinates of C1C



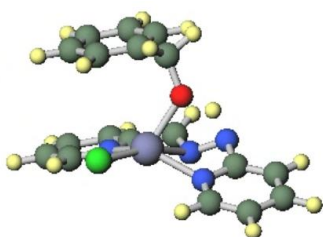
28

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Cl	-0.18263455766259	2.33425710773412	1.72919039702197
N	-2.23109343622263	-0.11846313982127	0.53083511718132
C	2.31042029067022	2.28500537181774	-1.47593286954344
C	2.33854343012378	0.13044170087760	-0.66648698034966
C	3.71199076171252	-0.01192019470701	-0.92982124613793
C	3.67040816033850	2.25030290456139	-1.77369288414306
C	4.37681933775458	1.07280739729102	-1.49043062224470
N	1.61358345228681	-0.91451458725141	-0.10497917115125
H	4.22918629419812	-0.94529003395833	-0.69732959667551
H	1.70371513883539	3.17379861782941	-1.67556758537226
H	4.16282142907075	3.11891200141272	-2.21351583303962
H	5.44542561560465	1.00174583475270	-1.70777515579733
C	-3.50055683313297	0.23534612721860	0.70817501824308
C	-1.83200467021254	-1.35528408143939	0.87866622547436
C	-2.71798819460604	-2.29135541590149	1.42953902548813
C	-4.45386849517025	-0.63248547023196	1.25388203547620
C	-4.04961994158929	-1.91813357562977	1.61946331579492
H	-2.36353501430074	-3.28792984788889	1.70183114838257
H	-3.76778028364989	1.25220602119302	0.40254479413039
H	-5.48524613041792	-0.30048964481871	1.38633270051390

H	-4.76418920136051	-2.62471609577776	2.04866464603361
N	0.31833141995506	-0.75399247799359	0.12797074458247
H	2.08381908932793	-1.78512170732546	0.13896256395154
C	-0.41802188626315	-1.66661523854707	0.65466949867224
H	-0.02517974202207	-2.65129175922064	0.94534249759158
H	-1.26130484578894	1.84586637850182	-1.61432676783914

Table S13. Coordinates of TS1



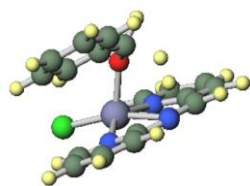
42

E -3228.584173200517

Zn	1.36652224921027	0.37676256980635	0.63630599580950
N	3.07518875847833	0.02482266248133	-0.67154710080011
O	0.62147310444973	1.47043189555797	-0.93138442494511
Cl	2.11437334938588	1.45387278706417	2.46403603885741
N	-0.16495408544000	-0.92477837642441	1.53877580496699
C	4.35810560691966	0.34805811648844	-0.48275041292196
C	2.64019849527369	-0.33096736203944	-1.89520092358808
C	3.50186189757390	-0.35192125255310	-2.99670015129704
C	5.28615068032140	0.33485242516861	-1.52302433661149
C	4.84063803864896	-0.01493229122376	-2.80269648169566
N	1.25293333001084	-0.60078609155108	-2.05816425875287
H	3.10878713290395	-0.64123770264201	-3.97234730789518
H	4.63526775121609	0.64641819411438	0.53300305418547
H	6.32714738087661	0.60274146384207	-1.33438524635851

H	5.53703660821042	-0.03086785864789	-3.64513596297309
C	-0.46242196102546	-0.99164167002210	2.83684023773679
C	-0.87603509449109	-1.64888739280094	0.64489966068279
C	-1.95948724555895	-2.43680765005785	1.04526601320596
C	-1.50929101479946	-1.78053715417724	3.31864684075986
C	-2.27299427407744	-2.50736764612784	2.40263100626082
H	-2.53128550830948	-2.99203582721543	0.29946845604028
H	0.15028878649774	-0.36938554757655	3.49623954636735
H	-1.72770623704630	-1.80481396093241	4.38779575355183
H	-3.10871570121847	-3.12379838760206	2.74291673528084
N	0.78520368104535	-1.14517462975027	-0.89029789870567
H	0.79692575364899	0.50109660698355	-1.84679460716110
C	-0.43749738933537	-1.50519097293578	-0.75223273937094
H	-1.14559555627374	-1.61361207795602	-1.58467193237768
C	-0.46278944148402	2.36449352409775	-0.89591420108500
H	-0.68968276784835	2.70909547331442	-1.92318925407706
H	-0.18686262723524	3.26119349003480	-0.30832415047968
C	-1.70164504414915	1.73779251924742	-0.29158907578797
C	-2.60822132957762	1.02582540977329	-1.09109695835964
C	-1.91843126676078	1.78424397790343	1.09312617119320
C	-3.01459959963711	1.13549624773641	1.66732772904422
C	-3.70672055392234	0.37556040291878	-0.52251343877966
C	-3.91010511467549	0.42691192939036	0.86040925991782
H	-1.20978997062508	2.32499629325897	1.72742466368237
H	-3.16936909368183	1.17972616443849	2.74841092405546
H	-2.44947838899901	0.98562750188281	-2.17317300354339
H	-4.40697982821019	-0.17282937848522	-1.15881580214574
H	-4.76743951028984	-0.08244142478241	1.30842577811367

Table S14. Coordinates of TS2



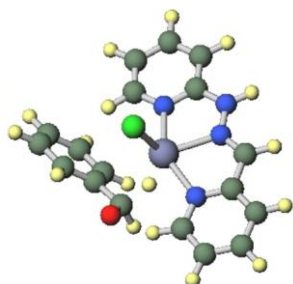
42

E -3228.552947701448

Zn	-0.45545940559373	-0.40846545677201	1.61994692129467
N	1.57887117438492	-0.14605306468021	0.94354302573531
O	-1.42531929971160	0.49576768578020	0.02620859364000
Cl	-0.51289373800588	0.79271915856962	3.52460733384594
N	-2.01709113011032	-1.90016517441849	1.92128234732091
C	2.47013002273185	0.72201086683385	1.43856963894027
C	1.75763104895655	-0.67359342414003	-0.28121645253056
C	2.85196617312401	-0.29722651721121	-1.08240074455609
C	3.58934576287075	1.12762944737349	0.72224355391247
C	3.76642009031579	0.61313112966709	-0.57186910045908
N	0.82608608865670	-1.58717876369748	-0.78271943621619
H	2.95754361961262	-0.73150751373769	-2.07764699844442
H	2.24357004953616	1.11419255551394	2.43473924746915
H	4.29327599753597	1.84002273438534	1.15486453816109
H	4.62299187671786	0.92278839644291	-1.17595348959780
C	-2.90817968105952	-1.91602162349252	2.91309187138954
C	-2.10183049003095	-2.80213531152112	0.91521417217556
C	-3.13530892127858	-3.75039162016881	0.88321341438824
C	-3.95862934283776	-2.83571013708061	2.95682632830590
C	-4.07049525803657	-3.76419132288437	1.91704293747676
H	-3.18994724265741	-4.46375812786422	0.05821445298732
H	-2.77262742209123	-1.15355703730346	3.68656127380774

H	-4.67287868892785	-2.81363480808467	3.78169180237852
H	-4.88230040394905	-4.49600251374086	1.91317372795070
N	-0.09385210620200	-1.91165851040245	0.12081018766598
H	0.08122191319075	-0.92181150586359	-1.74872102275834
C	-1.07951939025849	-2.70961806946111	-0.12641791490499
H	-1.17822414104485	-3.26657706586261	-1.06642076646768
C	-1.16925444590976	0.87628591478078	-1.15258124878253
H	-2.00771204576112	0.89031055897339	-1.87840013339853
H	-0.59963116495123	-0.30020147910524	-2.02265453091171
C	-0.10069795791500	1.89349674670202	-1.42943134022164
C	0.45519650480404	2.62246967979708	-0.37075915898910
C	0.30236863323225	2.17099215105769	-2.74499623337316
C	1.26637139113676	3.14487518441051	-2.99666738154271
C	1.42062822241789	3.60096246430968	-0.62378888961974
C	1.83259903734872	3.86085474531169	-1.93289997949122
H	-0.13072412177534	1.60034862932619	-3.57215821292514
H	1.58233324679408	3.34798593490318	-4.02334080557264
H	0.12129220514231	2.42634986100579	0.64938183030062
H	1.85190302479740	4.16259473216783	0.20862135613832
H	2.59082931480089	4.62366947018049	-2.12880171452171

Table S15. Coordinates of TS3

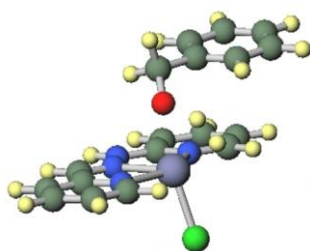


E -3228.549384719668

Zn	-0.22969418920271	-0.45716614994191	0.90088699932348
N	1.74516479026178	-0.20704392792481	-0.06870810968330
O	-2.45286636207553	1.49026628168738	0.81030120034247
Cl	0.34989729216103	0.20396079164401	2.96019953809780
N	-1.92884902788775	-1.76328407645980	1.15414557545027
C	2.34412663477638	0.97060122289975	-0.30085032651919
C	2.41209683490603	-1.34080212007505	-0.31187774766604
C	3.71994970901771	-1.35457713371138	-0.81946866965027
C	3.63616608100206	1.05921689977489	-0.80840342808282
C	4.32935901602310	-0.12974536586094	-1.07087487849120
N	1.74431768273725	-2.53170903490609	-0.03101996294429
H	4.23295233898750	-2.30057887403864	-1.00433487776025
H	1.76178011895031	1.86229919302120	-0.06280419242276
H	4.08540675283444	2.03656004612404	-0.98864482566446
H	5.34718349130560	-0.10210186085757	-1.46746116394104
C	-3.16162614744792	-1.34737016525776	1.46099769602903
C	-1.65182827135937	-3.08296904309014	1.07798942825689
C	-2.62471958899112	-4.05041558901396	1.34487973085428
C	-4.18971705946839	-2.25934323440285	1.73215898109764
C	-3.91243517179447	-3.62560106945452	1.68442340224205
H	-2.37386352048964	-5.11134288070601	1.28081120108853
H	-3.29062103576031	-0.25375559065585	1.44280748684734
H	-5.18759100241268	-1.89106414119321	1.97733037008078
H	-4.69262706526642	-4.35952955520598	1.90113184712502
N	0.50192211459046	-2.45587719535094	0.41522025232653
H	2.18943326235262	-3.42933095279650	-0.21605928570269
C	-0.28879908742174	-3.43770648093192	0.65861231175782

H	-0.00427657963371	-4.49131154958228	0.53561989955527
C	-1.84343136034540	1.37598280755969	-0.32213551566549
H	-2.45418212156929	1.10204460344911	-1.23247969455306
H	-1.07699247865863	0.37033911500134	-0.43081479928824
C	-0.84440532849669	2.46544974844765	-0.72778063604890
C	-0.48250321326328	3.44081415672915	0.20612700343166
C	-0.24990547764535	2.47806904039943	-1.99695849336173
C	0.70902173011646	3.43966558712935	-2.32351763897014
C	0.48279389251981	4.40262734615613	-0.11372359066768
C	1.08421776096807	4.40334025995641	-1.37743079534629
H	-0.53419890215319	1.71528882144970	-2.73055612637655
H	1.16962550195115	3.43952600774612	-3.31576526376128
H	-0.97349737359140	3.41303941576226	1.18143158120801
H	0.76630565175227	5.15771248901794	0.62538467441234
H	1.83690770772096	5.15582615746255	-1.62878915695955

Table S16. Coordinates of TS4



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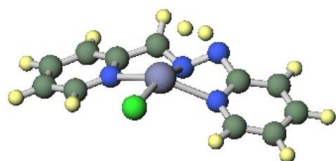
E -3229.744521412231

Zn	-0.33235384469494	-0.39797318578519	1.59838862547348
N	1.51368398093826	0.14019028960156	0.47141851133137
O	-1.58252591762102	0.43275110036937	-0.03234057904199
Cl	0.74888127776129	-0.87905912758324	3.56932601081740
N	-1.95057917289801	-1.90113280897420	1.84606348384301

C	2.19946720420458	1.27652090615876	0.64175612014999
C	1.97002213568220	-0.77338488045003	-0.38705992928405
C	3.14562148266032	-0.59286969291648	-1.13219357056051
C	3.37666162059209	1.54959523481683	-0.04531901537309
C	3.85054860192512	0.59094562895607	-0.94951441370722
N	1.21939168056343	-1.94018013357388	-0.51960699291927
H	3.48594795263846	-1.36435951141724	-1.82594118185381
H	1.77330028805219	1.98787924376294	1.35290085218491
H	3.90212616514904	2.49109525111528	0.11565155089953
H	4.76950842345269	0.76803209977768	-1.51348849857651
C	-2.98474410571518	-1.77494734474535	2.67339770140167
C	-1.83450858106029	-3.00774549260388	1.08537550326454
C	-2.78002475862819	-4.03938665270751	1.14188733622533
C	-3.97469136471758	-2.75647720572291	2.79143738843369
C	-3.86525371716482	-3.90785363350928	2.00985881774042
H	-2.65956335005169	-4.92521233262158	0.51459349540868
H	-3.02396610638508	-0.85330248055828	3.26192646969641
H	-4.80754365669955	-2.61319231994332	3.48213527042243
H	-4.61786191409374	-4.69748143501355	2.07434239507618
N	0.13682578677940	-2.07847844700507	0.22398047119882
H	1.50939607840346	-2.66401476314299	-1.17574324006350
C	-0.67328963379248	-3.07241237733971	0.18995144258824
H	-0.53937616136475	-3.93888680162744	-0.47252477275092
C	-1.23225248525011	0.76850369320705	-1.33856342940902
H	-2.14226106719597	1.03889903286349	-1.92221949481735
H	-0.80802404411078	-0.11700419662352	-1.85723107275441
C	-0.24435402426466	1.91336092985506	-1.49667463492126
C	-0.30914734602001	3.05287222213248	-0.68043902804104

C	0.73324152886292	1.86884774867748	-2.50025700222517
C	1.62396779254958	2.92956341383697	-2.68794890458524
C	0.58003532160172	4.11498024851832	-0.86256693762899
C	1.55127132907956	4.05886543640712	-1.86784607475166
H	0.80488210322505	0.98292675542915	-3.13866608305319
H	2.38427938386876	2.86930767958768	-3.47179951658435
H	-1.05622447006860	3.10558620659453	0.11411138818559
H	0.51486505718516	4.99278904814005	-0.21349375020229
H	2.24946943317997	4.88858508776228	-2.00743836708977
H	-1.18659419117340	1.32953440696421	1.95000141878846
H	-1.43825471538440	1.14372315933028	0.99037223706447

Table S17. Coordinates of TS5



28

E -2883.351639617781

Zn	-0.54876559795690	0.78285129199717	-0.83472637604291
N	1.54689551406937	1.14242139603807	-0.66478209658063
Cl	-1.49430869118910	2.80394068271203	-1.13339393869004
N	-2.00794857982552	-0.15378898219212	0.50650040023047
C	2.17618076562628	2.32131490771617	-0.77292327518427
C	2.25160107938427	-0.00920465939274	-0.70884691995795
C	3.63727483920836	-0.00767150696512	-0.90372276424733
C	3.55596572198811	2.40569701475752	-0.93082759296657
C	4.29379484452540	1.21621484208155	-1.00548621980189
N	1.50748819950036	-1.20942122876386	-0.67652632609001

H	4.16576917804592	-0.96058769722965	-0.95434855158649
H	1.53693121558193	3.20888927516311	-0.74877254921283
H	4.04014494529022	3.38099178300276	-1.00491423693852
H	5.37829952524036	1.24744468102073	-1.13810193443496
C	-3.12565175815879	0.42287408244200	0.95164926536933
C	-1.72594788618174	-1.43563836375857	0.84171430848454
C	-2.60964247209226	-2.19590800538379	1.61129460697925
C	-4.04750416575377	-0.26547278444221	1.74577274150089
C	-3.78565915308950	-1.59742734186273	2.07154115628619
H	-2.36583090817551	-3.23152718273322	1.85604699078270
H	-3.28554109286486	1.45711752821507	0.63206005453111
H	-4.95903660857742	0.23351062163899	2.07966385614674
H	-4.49036532232842	-2.16757270514939	2.68196650736044
N	0.47495043201209	-1.04417984281880	0.20708278382179
H	0.77758188071508	-0.98991945979968	-1.68511066657810
C	-0.45097607845936	-1.93033794116289	0.28742967642681
H	-0.36729525242451	-2.94899819144665	-0.12161573965159
H	-0.07840257411011	-0.47561321368374	-2.18861915995618

Table S18. Coordinates of BnOH

16

E -346.339139661818

C	-0.01673352124309	-0.01259612864034	-0.01548220143607
H	1.07796807239952	-0.00979244039621	-0.03086122758004
C	-0.71336415754933	1.20583892219054	0.01336997295400
C	-0.70620775660443	-1.22637264608441	-0.02586039685538
C	-2.10632385246644	-1.23903498427968	-0.00985991681592
C	-2.11290081177709	1.18720004278765	0.02877697211429

C	-2.80492402151515	-0.02947306784456	0.01663937035805
H	-0.14991886218258	-2.16749431144589	-0.04861958389485
H	-2.64763447265369	-2.18881267404452	-0.01946685557112
C	0.05574648713146	2.50718300412686	0.03718957491688
H	-2.65331478651944	2.13431900164143	0.04859425387620
H	-3.89847379962263	-0.02960779312039	0.02785472856198
H	0.66294788938761	2.54112883432726	0.96604677802331
H	0.77825718420124	2.50728033326860	-0.80526520086275
O	-0.82869216016119	3.60295036598370	-0.03904756210174
H	-0.30899343082472	4.41499454152992	0.01699229431317

Table S19. Coordinates of PhCHO

14

E -345.144730053970

C	-0.01566988571037	0.00560192412662	-0.20370827165694
H	1.06957036972376	0.02754442460116	-0.34187333573740
C	-0.71896364299463	1.21224246090869	-0.06724225066289
C	-0.69912384180408	-1.21153921555436	-0.16331200079612
C	-2.08696199156313	-1.22207207831258	0.01371984610066
C	-2.11350892340371	1.19607077771011	0.11029132475263
C	-2.79407917017587	-0.01857206633031	0.15044892950803
H	-0.15288124634341	-2.15203059964897	-0.26956774982984
H	-2.62361715552995	-2.17405750536645	0.04583030809633
C	0.01333028493441	2.49849900502771	-0.11200149739520
H	-2.63940813950945	2.14793463078796	0.21411424303081
H	-3.87817212989921	-0.03547414197720	0.28846221336761
H	1.11974970421867	2.39636474359588	-0.25449392421143
O	-0.50168423194303	3.59135764043176	-0.00411783456623

Table S20. Coordinates of H₂

2

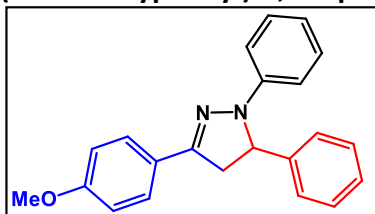
E -1.167355515050

H	-0.07078363338030	0.0000000000000000	0.0000000000000000
H	0.69078363338030	0.0000000000000000	0.0000000000000000

10. ¹H and ¹³C{¹H} NMR characterization of 1,3,5-trisubstituted pyrazolines derivatives

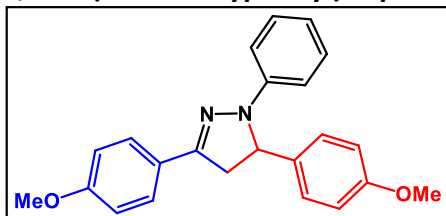
The NMR spectroscopic data for all the reported pyrazoline substrates are in agreement with the previously reported literatures.^{6–8}

3-(4-methoxyphenyl)-1,5-diphenyl-4,5-dihydro-1H-pyrazole (4a)



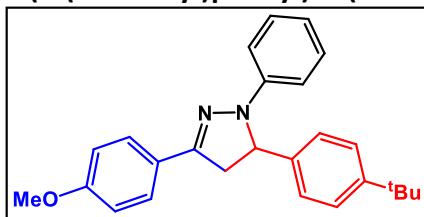
Brown liquid, Yield: 86%, 141 mg. ¹H NMR (500 MHz, CDCl₃) δ 7.68 – 7.64 (m, 2H), 7.33 (d, *J* = 4.4 Hz, 4H), 7.28 – 7.24 (m, 1H), 7.19 – 7.15 (m, 2H), 7.08 – 7.04 (m, 2H), 6.93 – 6.89 (m, 2H), 6.77 (tt, *J* = 7.3, 1.1 Hz, 2H), 5.21 (dd, *J* = 12.3, 7.3 Hz, 2H), 3.85 – 3.77 (m, 4H), 3.11 (dd, *J* = 17.0, 7.4 Hz, 1H). ¹³C{¹H} NMR (126 MHz, CDCl₃) δ 160.20, 146.87, 145.28, 142.86, 129.21, 128.97, 127.60, 127.32, 126.00, 125.60, 118.89, 114.09, 113.34, 64.57, 55.43, 43.90.

3,5-bis(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4b)



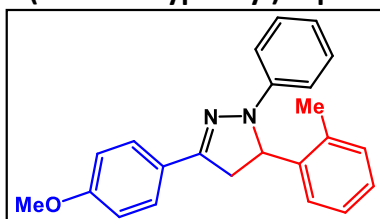
Brown liquid, Yield: 90%, 162 mg. ¹H NMR (500 MHz, CDCl₃) δ 7.69 – 7.64 (m, 2H), 7.26 – 7.22 (m, 2H), 7.22 – 7.10 (m, 2H), 7.06 (dd, *J* = 8.9, 1.2 Hz, 2H), 6.94 – 6.89 (m, 2H), 6.88 – 6.83 (m, 2H), 6.79 – 6.73 (m, 2H), 5.17 (dd, *J* = 12.4, 7.4 Hz, 1H), 3.85 – 3.80 (m, 4H), 3.77 (s, 3H), 3.08 (dd, *J* = 17.0, 7.4 Hz, 1H). ¹³C{¹H} NMR (126 MHz, CDCl₃) δ 160.17, 159.01, 146.89, 145.32, 134.92, 128.95, 127.29, 127.17, 125.69, 118.85, 114.54, 114.09, 113.38, 64.07, 55.44, 55.36, 43.95.

5-(4-(tert-butyl)phenyl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4c)



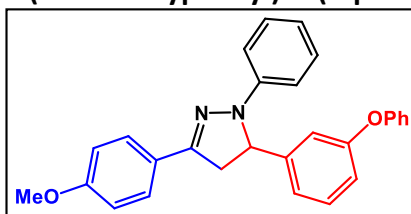
Brown liquid, Yield: 92%, 177 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.68 – 7.62 (m, 2H), 7.33 (dt, J = 8.9, 2.3 Hz, 2H), 7.26 – 7.23 (m, 2H), 7.22 – 7.12 (m, 2H), 7.06 (dt, J = 15.9, 5.2 Hz, 2H), 6.93 – 6.87 (m, 2H), 6.78 – 6.72 (m, 1H), 5.19 (dd, J = 12.2, 7.3 Hz, 1H), 3.83 (s, 3H), 3.78 (dd, J = 17.0, 12.2 Hz, 1H), 3.11 (dd, J = 17.0, 7.3 Hz, 1H), 1.29 (s, 9H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.14, 150.41, 146.92, 145.37, 139.77, 128.95, 127.29, 126.06, 125.62, 118.76, 114.06, 113.26, 64.21, 55.43, 43.89, 34.59, 31.43

3-(4-methoxyphenyl)-1-phenyl-5-(o-tolyl)-4,5-dihydro-1H-pyrazole (4d)



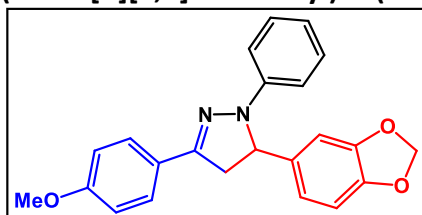
Brown liquid, Yield: 87%, 149 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.68 – 7.64 (m, 2H), 7.22 (t, J = 7.1 Hz, 2H), 7.19 – 7.14 (m, 3H), 7.10 (t, J = 7.7 Hz, 1H), 6.98 (dd, J = 8.8, 1.1 Hz, 2H), 6.92 – 6.89 (m, 2H), 6.76 (tt, J = 7.3, 1.1 Hz, 1H), 5.35 (dd, J = 12.4, 7.5 Hz, 1H), 3.88 – 3.84 (m, 1H), 3.83 (s, 3H), 2.98 (dd, J = 16.8, 7.5 Hz, 1H), 2.47 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.16, 146.76, 145.19, 140.30, 130.95, 129.02, 128.84, 127.38, 127.15, 126.96, 125.64, 123.66, 118.80, 114.06, 113.07, 55.43, 42.18, 19.62.

3-(4-methoxyphenyl)-5-(3-phenoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4e)



Brown liquid, Yield: 86%, 181 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.68 – 7.63 (m, 2H), 7.37 – 7.26 (m, 4H), 7.23 – 7.15 (m, 2H), 7.11 – 7.02 (m, 4H), 6.95 (ddd, J = 4.7, 3.2, 2.0 Hz, 2H), 6.93 – 6.89 (m, 2H), 6.88 – 6.84 (m, 1H), 6.81 – 6.77 (m, 1H), 5.18 (dd, J = 12.6, 7.4 Hz, 1H), 3.83 (s, 3H), 3.79 (dd, J = 12.6, 7.8 Hz, 1H), 3.13 (dd, J = 17.0, 7.4 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.24, 157.85, 156.99, 146.89, 145.19, 144.95, 130.62, 129.85, 128.98, 127.33, 125.52, 123.41, 120.74, 119.02, 118.90, 117.74, 116.64, 114.10, 113.45, 64.32, 55.44, 43.76.

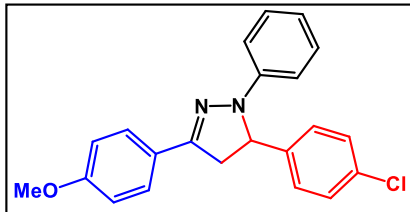
5-(benzo[d][1,3]dioxol-5-yl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4f)



Brown liquid, Yield: 88%, 164 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.67 – 7.63 (m, 2H), 7.20 – 7.15 (m, 2H), 7.08 – 7.05 (m, 2H), 6.93 – 6.89 (m, 2H), 6.78 (dddd, J = 7.9, 6.2, 5.6, 2.5 Hz, 4H), 5.91 (dd, J = 5.9, 1.4 Hz, 2H), 5.13 (dd, J = 12.2, 7.2 Hz, 1H), 3.83 (s, 3H), 3.76 (dd, J = 17.0, 12.2 Hz, 1H), 3.07 (dd, J = 17.0, 7.2 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.20,

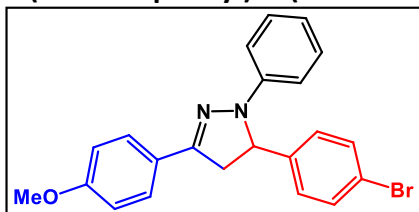
148.42, 147.04, 146.86, 145.20, 136.87, 128.96, 127.29, 125.57, 119.21, 118.93, 114.08, 113.33, 108.68, 106.35, 101.19, 64.36, 55.44, 43.92.

5-(4-chlorophenyl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4g)



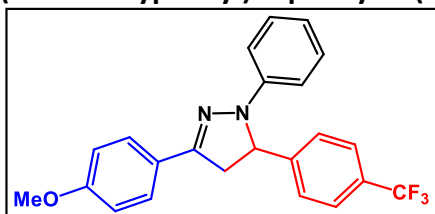
Brown liquid, Yield: 82%, 149 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.68 – 7.62 (m, 2H), 7.31 – 7.28 (m, 2H), 7.27 – 7.24 (m, 2H), 7.20 – 7.16 (m, 2H), 7.02 (dd, J = 8.7, 1.0 Hz, 2H), 6.93 – 6.89 (m, 2H), 6.78 (dt, J = 8.2, 0.9 Hz, 1H), 5.18 (dd, J = 12.2, 7.2 Hz, 1H), 3.83 (s, 3H), 3.79 (dd, J = 17.0, 12.3 Hz, 1H), 3.06 (dd, J = 17.0, 7.2 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.30, 146.88, 145.04, 141.33, 133.32, 129.40, 129.05, 127.46, 127.35, 125.36, 119.15, 114.13, 113.35, 63.91, 55.46, 43.79.

5-(4-bromophenyl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4h)



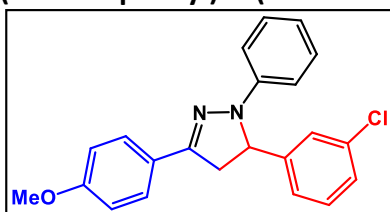
Brown liquid, Yield: 84%, 171 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.67 – 7.62 (m, 2H), 7.46 – 7.43 (m, 2H), 7.21 – 7.15 (m, 4H), 7.01 (dt, J = 9.0, 1.6 Hz, 2H), 6.93 – 6.89 (m, 2H), 6.78 (tt, J = 7.4, 1.0 Hz, 1H), 5.17 (dd, J = 12.2, 7.2 Hz, 1H), 3.83 (s, 3H), 3.79 (dd, J = 17.0, 12.3 Hz, 1H), 3.06 (dd, J = 17.0, 7.3 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.30, 146.88, 145.03, 141.86, 132.34, 129.05, 127.81, 127.35, 125.34, 121.41, 119.17, 114.13, 113.34, 63.96, 55.46, 43.74.

3-(4-methoxyphenyl)-1-phenyl-5-(4-(trifluoromethyl)phenyl)-4,5-dihydro-1H-pyrazole (4i)



Brown liquid, Yield: 85%, 169 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.68 – 7.63 (m, 2H), 7.62 – 7.54 (m, 2H), 7.44 (d, J = 8.8 Hz, 1H), 7.40 – 7.31 (m, 1H), 7.23 – 7.12 (m, 2H), 7.04 – 6.95 (m, 2H), 6.94 – 6.87 (m, 2H), 6.82 – 6.76 (m, 1H), 5.27 (dd, J = 12.8, 7.4 Hz, 1H), 3.85 (t, J = 6.3 Hz, 1H), 3.83 (s, 3H), 3.08 (dd, J = 17.0, 7.3 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.37, 146.86, 146.80, 144.96, 129.24, 129.09, 128.95, 127.36, 127.18, 126.27 (q, J = 3.8 Hz), 125.57, 125.53, 125.44, 125.21, 119.29, 114.14, 113.32, 64.07, 55.44, 43.71.

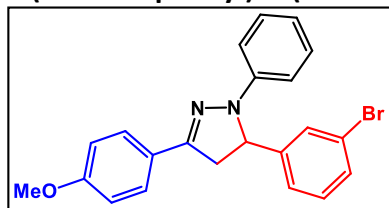
5-(3-chlorophenyl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4j)



Brown liquid, Yield: 86%, 156 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.32 – 7.25 (m, 4H), 7.24 – 7.13 (m, 5H), 7.04 – 7.00 (m, 2H), 6.87 (ddd, J = 8.2, 2.8, 1.2 Hz, 1H), 6.81 – 6.76 (m, 1H),

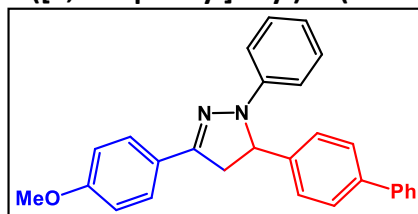
5.21 (dd, $J = 12.6, 7.2$ Hz, 1H), 3.84 (s, 3H), 3.79 (dd, $J = 17.0, 12.3$ Hz, 1H), 3.06 (dd, $J = 17.0, 7.2$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.95, 146.83, 144.77, 141.23, 134.09, 133.54, 129.79, 129.55, 129.17, 127.53, 119.62, 118.67, 115.05, 113.62, 110.82, 64.09, 55.53, 43.72.

5-(3-bromophenyl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4k)



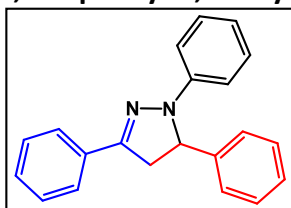
Brown liquid, Yield: 87%, 177 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.49 – 7.45 (m, 2H), 7.35 – 7.29 (m, 2H), 7.28 – 7.16 (m, 5H), 7.09 – 7.03 (m, 2H), 6.91 (ddd, $J = 8.2, 2.7, 1.1$ Hz, 1H), 6.85 – 6.79 (m, 1H), 5.24 (dd, $J = 12.6, 7.3$ Hz, 1H), 3.88 (s, 3H), 3.83 (dd, $J = 17.0, 12.3$ Hz, 1H), 3.09 (dd, $J = 17.1, 7.2$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.90, 146.81, 144.70, 141.73, 134.03, 132.47, 129.78, 129.16, 127.85, 121.59, 119.60, 118.64, 115.04, 113.58, 110.77, 64.09, 55.51, 43.64.

5-([1,1'-biphenyl]-4-yl)-3-(4-methoxyphenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4l)



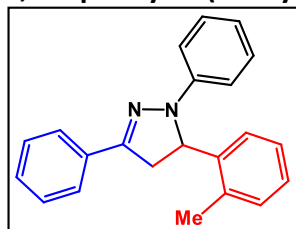
Brown liquid, Yield: 83%, 169 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.72 – 7.67 (m, 2H), 7.63 – 7.55 (m, 4H), 7.48 – 7.32 (m, 5H), 7.25 – 7.17 (m, 2H), 7.14 – 7.08 (m, 2H), 6.95 – 6.90 (m, 2H), 6.83 – 6.77 (m, 1H), 5.26 (dd, $J = 12.6, 7.5$ Hz, 1H), 3.86 – 3.81 (m, 4H), 3.15 (dd, $J = 17.1, 7.4$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.24, 146.97, 145.31, 141.92, 140.74, 140.53, 129.05, 128.89, 127.96, 127.44, 127.37, 127.15, 126.49, 125.60, 118.99, 114.13, 113.40, 64.32, 55.46, 43.93.

1,3,5-triphenyl-4,5-dihydro-1H-pyrazole (4m)



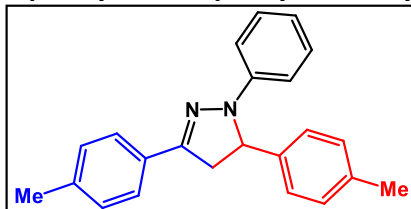
colourless liquid, Yield: 90%, 134 mg. ^1H NMR (500 MHz) δ 7.74 (ddd, $J = 4.1, 3.4, 1.7$ Hz, 2H), 7.42 – 7.38 (m, 2H), 7.37 – 7.32 (m, 5H), 7.30 – 7.26 (m, 1H), 7.23 – 7.18 (m, 2H), 7.13 – 7.09 (m, 2H), 6.81 (tt, $J = 7.3, 1.2$ Hz, 1H), 5.27 (dd, $J = 12.4, 7.2$ Hz, 1H), 3.84 (dd, $J = 17.0, 12.4$ Hz, 1H), 3.15 (dd, $J = 17.0, 7.2$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 146.83, 144.96, 142.71, 132.84, 129.26, 129.03, 128.71, 128.66, 127.69, 125.99, 125.85, 119.20, 113.48, 64.59, 43.67.

1,3-diphenyl-5-(o-tolyl)-4,5-dihydro-1H-pyrazole (4n)



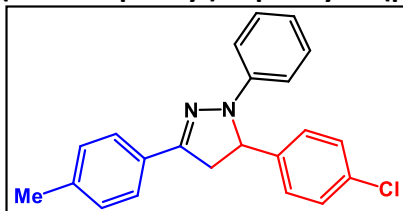
colourless liquid, Yield: 88%, 138 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.76 – 7.70 (m, 2H), 7.47 – 7.29 (m, 4H), 7.26 – 7.07 (m, 5H), 7.05 – 6.96 (m, 2H), 6.83 – 6.76 (m, 1H), 5.41 (dd, J = 12.9, 7.6 Hz, 1H), 3.88 (dd, J = 17.0, 12.5 Hz, 1H), 3.02 (dd, J = 17.1, 7.6 Hz, 1H), 2.49 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 146.72, 144.88, 140.14, 132.88, 131.00, 129.26, 129.06, 128.67, 128.63, 127.46, 126.99, 125.98, 125.81, 119.12, 113.23, 64.58, 41.99, 19.63.

1-phenyl-3,5-di-p-tolyl-4,5-dihydro-1H-pyrazole (4o)



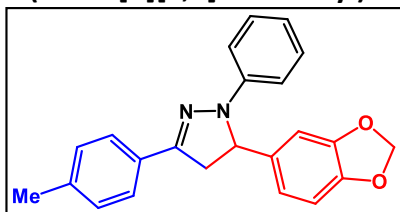
colourless liquid, Yield: 91%, 149 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.66 – 7.59 (m, 2H), 7.19 (dddd, J = 16.3, 11.1, 8.6, 4.4 Hz, 8H), 7.12 – 7.06 (m, 2H), 6.78 (tt, J = 7.3, 1.1 Hz, 1H), 5.21 (dd, J = 12.3, 7.3 Hz, 1H), 3.80 (dd, J = 17.0, 12.3 Hz, 1H), 3.11 (dd, J = 17.0, 7.3 Hz, 1H), 2.38 (s, 3H), 2.33 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 147.04, 145.18, 139.88, 138.72, 137.26, 130.13, 129.88, 129.35, 128.97, 125.92, 125.81, 118.96, 113.41, 64.33, 43.85, 21.51, 21.21.

5-(4-chlorophenyl)-1-phenyl-3-(p-tolyl)-4,5-dihydro-1H-pyrazole (4p)



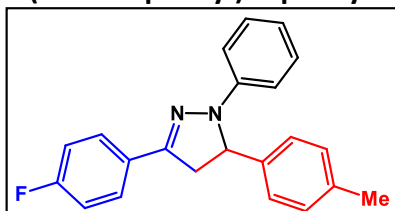
colourless liquid, Yield: 85%, 148 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.64 – 7.57 (m, 2H), 7.31 – 7.27 (m, 2H), 7.27 – 7.24 (m, 2H), 7.21 – 7.16 (m, 4H), 7.05 – 7.02 (m, 2H), 6.81 – 6.77 (m, 1H), 5.21 (dd, J = 12.3, 7.2 Hz, 1H), 3.81 (dd, J = 17.0, 12.3 Hz, 1H), 3.07 (dd, J = 17.0, 7.2 Hz, 1H), 2.37 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 147.03, 144.89, 141.28, 138.97, 133.35, 129.84, 129.40, 129.04, 127.44, 125.82, 119.28, 113.42, 63.90, 43.67, 21.50.

5-(benzo[d][1,3]dioxol-5-yl)-1-phenyl-3-(p-tolyl)-4,5-dihydro-1H-pyrazole (4q)



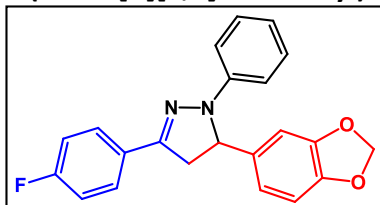
colourless liquid, Yield: 89%, 159 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.63 – 7.60 (m, 2H), 7.22 – 7.17 (m, 4H), 7.11 – 7.07 (m, 2H), 6.82 – 6.74 (m, 4H), 5.91 (dd, J = 6.0, 1.4 Hz, 2H), 5.15 (dd, J = 12.2, 7.2 Hz, 1H), 3.76 (dd, J = 17.0, 12.2 Hz, 1H), 3.09 (dd, J = 17.0, 7.2 Hz, 1H), 2.37 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 148.44, 147.06, 147.03, 145.04, 138.80, 136.83, 130.03, 129.36, 128.99, 125.80, 119.21, 119.07, 113.41, 108.70, 106.34, 101.20, 64.33, 43.80, 21.51.

3-(4-fluorophenyl)-1-phenyl-5-(p-tolyl)-4,5-dihydro-1H-pyrazole (4r)



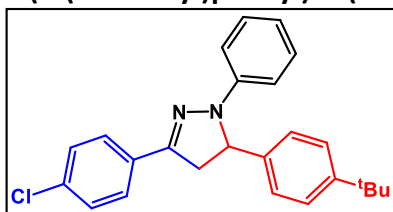
Brown liquid, Yield: 81%, 134 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.74 – 7.64 (m, 2H), 7.40 – 7.34 (m, 1H), 7.24 – 7.01 (m, 9H), 6.80 – 6.74 (m, 1H), 5.23 (dd, J = 12.6, 7.4 Hz, 1H), 3.78 (dd, J = 17.2, 12.3 Hz, 1H), 3.08 (dd, J = 17.1, 7.4 Hz, 1H), 2.32 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.03, 162.05, 145.89, 144.95, 139.59, 137.39, 129.91, 129.34, 129.00, 128.68, 127.53 (d, J = 8.3 Hz), 125.86, 119.17, 115.68 (d, J = 22.7 Hz), 113.41, 104.81, 64.46, 43.78, 21.20.

5-(benzo[d][1,3]dioxol-5-yl)-3-(4-fluorophenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4s)



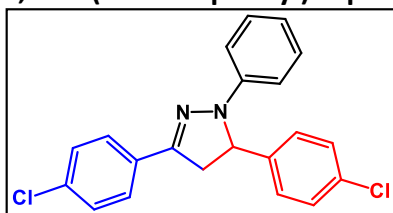
Brown liquid, Yield: 84%, 151 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.71 – 7.65 (m, 2H), 7.21 – 7.16 (m, 2H), 7.09 – 7.04 (m, 4H), 6.81 – 6.74 (m, 4H), 5.91 (dd, J = 5.8, 1.4 Hz, 2H), 5.17 (dd, J = 12.3, 7.2 Hz, 1H), 3.76 (dd, J = 17.0, 12.3 Hz, 1H), 3.07 (dd, J = 17.0, 7.2 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.06, 162.08, 148.48, 147.13, 145.88, 144.84, 136.55, 129.01, 127.54 (d, J = 8.4 Hz), 119.24 (d, J = 12.5 Hz), 115.7 (d, J = 22.7 Hz), 113.43, 108.74, 106.27, 101.22, 64.48, 43.75.

5-(4-(tert-butyl)phenyl)-3-(4-chlorophenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4t)



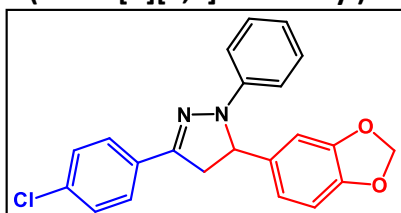
Yield: 90%, 175 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.66 – 7.61 (m, 2H), 7.36 – 7.33 (m, 4H), 7.25 – 7.18 (m, 4H), 7.12 – 7.07 (m, 2H), 6.82 – 6.78 (m, 1H), 5.26 (dd, J = 12.4, 7.1 Hz, 1H), 3.76 (dd, J = 17.0, 12.4 Hz, 1H), 3.09 (dd, J = 17.0, 7.2 Hz, 1H), 1.30 (s, 9H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 150.61, 145.68, 144.78, 139.37, 134.27, 131.45, 129.05, 128.83, 126.98, 126.17, 125.57, 119.30, 113.48, 64.31, 43.47, 34.62, 31.44.

3,5-bis(4-chlorophenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4u)



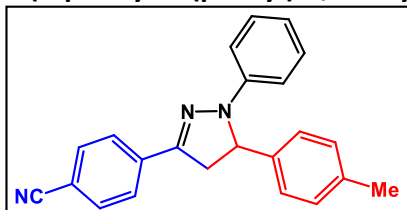
Brown liquid, Yield: 85%, 156 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.65 – 7.60 (m, 2H), 7.36 – 7.32 (m, 2H), 7.32 – 7.28 (m, 2H), 7.26 – 7.22 (m, 2H), 7.21 – 7.15 (m, 2H), 7.06 – 7.00 (m, 2H), 6.83 – 6.77 (m, 1H), 5.24 (dd, J = 12.7, 7.4 Hz, 1H), 3.78 (dd, J = 17.3, 12.4 Hz, 1H), 3.05 (dd, J = 17.1, 7.3 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 145.64, 144.46, 140.90, 134.53, 133.52, 131.15, 129.48, 129.10, 128.88, 127.37, 126.99, 119.68, 113.52, 64.05, 43.39.

5-(benzo[d][1,3]dioxol-5-yl)-3-(4-chlorophenyl)-1-phenyl-4,5-dihydro-1H-pyrazole (4v)



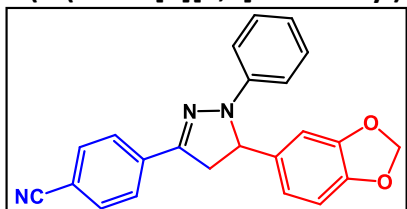
Brown liquid, Yield: 88%, 166 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.64 – 7.60 (m, 2H), 7.36 – 7.31 (m, 2H), 7.19 (tt, J = 7.5, 2.1 Hz, 2H), 7.11 – 7.05 (m, 2H), 6.83 – 6.70 (m, 4H), 5.91 (dd, J = 6.2, 1.5 Hz, 2H), 5.18 (dd, J = 12.7, 7.3 Hz, 1H), 3.73 (dd, J = 18.0, 12.3 Hz, 1H), 3.06 (dd, J = 17.2, 7.3 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 148.50, 147.18, 145.63, 144.62, 136.44, 134.35, 131.36, 129.04, 128.84, 126.96, 119.47, 119.20, 113.53, 108.76, 106.25, 101.24, 64.49, 43.51.

4-(1-phenyl-5-(p-tolyl)-4,5-dihydro-1H-pyrazol-3-yl)benzonitrile (4w)



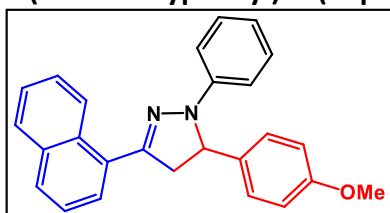
colourless liquid, Yield: 87%, 147 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.76 (dd, J = 33.8, 8.8 Hz, 4H), 7.40 – 7.33 (m, 1H), 7.20 – 7.06 (m, 7H), 6.80 (dd, J = 16.1, 8.8 Hz, 1H), 5.26 (dd, J = 13.0, 7.3 Hz, 1H), 3.78 (dd, J = 17.3, 12.5 Hz, 1H), 3.09 (dd, J = 17.2, 7.3 Hz, 1H), 2.30 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 169.17, 145.40, 144.35, 139.30, 137.50, 136.34, 132.57, 129.95, 129.34, 129.04, 128.67, 127.76, 125.80, 125.68, 119.63, 113.63, 64.45, 43.32, 21.18.

4-(5-(benzo[d][1,3]dioxol-5-yl)-1-phenyl-4,5-dihydro-1H-pyrazol-3-yl)benzonitrile (4x)



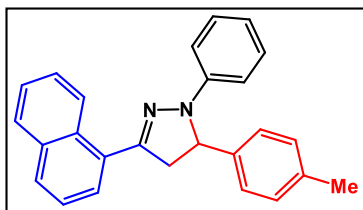
colourless liquid, Yield: 91%, 167 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.77 – 7.73 (m, 2H), 7.65 – 7.61 (m, 2H), 7.22 – 7.17 (m, 2H), 7.09 (dt, J = 8.9, 1.5 Hz, 2H), 6.86 – 6.81 (m, 1H), 6.78 – 6.74 (m, 2H), 6.73 – 6.71 (m, 1H), 5.93 – 5.90 (m, 2H), 5.28 (dd, J = 12.6, 7.0 Hz, 1H), 3.77 (dd, J = 17.1, 12.6 Hz, 1H), 3.08 (dd, J = 17.1, 7.0 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 148.58, 147.33, 144.35, 143.80, 137.14, 135.89, 132.38, 129.11, 125.87, 120.16, 119.17, 119.07, 113.78, 111.17, 108.83, 106.10, 101.31, 64.63, 42.94.

5-(4-methoxyphenyl)-3-(naphthalen-1-yl)-1-phenyl-4,5-dihydro-1H-pyrazole (4y)



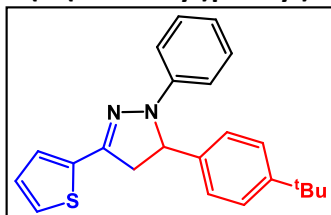
Yellow liquid, Yield: 94%, 178 mg. ^1H NMR (500 MHz, CDCl_3) δ 9.62 (d, J = 8.7 Hz, 1H), 7.90 (d, J = 8.1 Hz, 1H), 7.84 – 7.80 (m, 1H), 7.75 – 7.71 (m, 1H), 7.59 (t, J = 7.5 Hz, 1H), 7.45 – 7.41 (m, 2H), 7.32 – 7.26 (m, 4H), 7.19 (dd, J = 7.8, 0.8 Hz, 2H), 6.91 – 6.84 (m, 3H), 5.22 (dd, J = 12.2, 7.0 Hz, 1H), 3.98 (dd, J = 16.7, 12.2 Hz, 1H), 3.78 (s, 3H), 3.33 (dd, J = 16.7, 7.0 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.10, 147.45, 144.91, 134.70, 134.34, 130.71, 129.62, 129.20, 129.14, 128.77, 127.61, 127.46, 127.25, 126.85, 126.25, 125.04, 119.30, 114.64, 113.63, 62.88, 55.38, 46.22.

3-(naphthalen-1-yl)-1-phenyl-5-(p-tolyl)-4,5-dihydro-1H-pyrazole (4z)



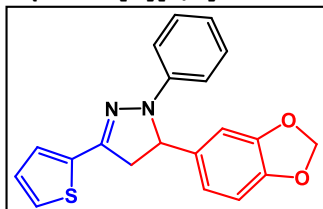
Yellow liquid, Yield: 91%, 165 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 9.62 (d, J = 9.0 Hz, 1H), 7.92 – 7.89 (m, 1H), 7.84 – 7.80 (m, 1H), 7.73 (ddd, J = 8.5, 6.8, 1.4 Hz, 1H), 7.59 (ddd, J = 8.1, 6.8, 1.2 Hz, 1H), 7.45 – 7.42 (m, 2H), 7.29 – 7.25 (m, 4H), 7.21 – 7.16 (m, 4H), 6.88 – 6.84 (m, 1H), 5.24 (dd, J = 12.2, 7.1 Hz, 1H), 4.01 (dd, J = 16.7, 12.2 Hz, 1H), 3.35 (dd, J = 16.7, 7.1 Hz, 1H), 2.35 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 147.42, 144.93, 139.72, 137.37, 134.33, 130.72, 129.96, 129.61, 129.20, 129.14, 128.74, 127.62, 127.45, 126.82, 126.24, 125.99, 125.02, 119.28, 113.59, 63.18, 46.24, 21.25.

5-(4-(tert-butyl)phenyl)-1-phenyl-3-(thiophen-2-yl)-4,5-dihydro-1H-pyrazole (4aa)



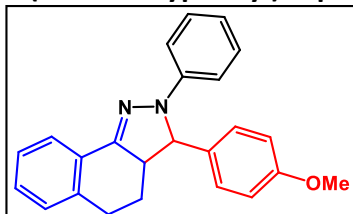
Colourless liquid, Yield: 84%, 155 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.36 – 7.33 (m, 2H), 7.29 (dd, J = 4.9, 1.2 Hz, 1H), 7.26 – 7.23 (m, 2H), 7.20 – 7.16 (m, 2H), 7.08 – 7.04 (m, 2H), 7.03 (dd, J = 3.6, 1.2 Hz, 1H), 7.00 (dd, J = 4.9, 3.6 Hz, 1H), 6.78 (tt, J = 7.3, 1.1 Hz, 1H), 5.23 (dd, J = 12.3, 7.2 Hz, 1H), 3.81 (dd, J = 16.8, 12.3 Hz, 1H), 3.13 (dd, J = 16.8, 7.2 Hz, 1H), 1.30 (s, 9H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 150.58, 144.86, 143.03, 139.35, 136.82, 128.97, 127.40, 126.54, 126.13, 125.93, 125.62, 119.17, 113.51, 64.39, 44.43, 34.61, 31.44.

5-(benzo[d][1,3]dioxol-5-yl)-1-phenyl-3-(thiophen-2-yl)-4,5-dihydro-1H-pyrazole (4ab)



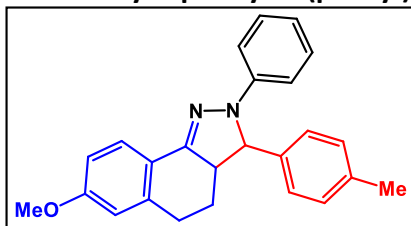
Colourless liquid, Yield: 86%, 150 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 7.37 – 7.28 (m, 2H), 7.21 – 7.16 (m, 2H), 7.07 – 7.02 (m, 2H), 7.01 (dd, J = 5.1, 3.6 Hz, 1H), 6.82 – 6.74 (m, 4H), 5.91 (dd, J = 6.6, 1.5 Hz, 2H), 5.16 (dd, J = 12.5, 7.3 Hz, 1H), 3.78 (dd, J = 17.1, 12.2 Hz, 1H), 3.10 (dd, J = 17.0, 7.3 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 148.49, 147.16, 144.69, 142.99, 136.70, 136.42, 128.99, 127.43, 126.65, 126.00, 119.34, 119.25, 113.56, 108.74, 106.32, 101.24, 64.53, 44.45.

3-(4-methoxyphenyl)-2-phenyl-3,3a,4,5-tetrahydro-2H-benzo[g]indazole (4ac)



brown liquid, Yield: 84%, 149 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.10 – 8.05 (m, 1H), 7.41 – 7.37 (m, 2H), 7.27 – 7.24 (m, 2H), 7.19 – 7.13 (m, 3H), 7.12 – 7.08 (m, 2H), 6.95 – 6.92 (m, 2H), 6.85 – 6.80 (m, 1H), 4.59 (d, J = 12.0 Hz, 1H), 3.82 (s, 3H), 3.24 (td, J = 12.7, 4.9 Hz, 1H), 2.95 – 2.88 (m, 2H), 2.29 (ddd, J = 9.0, 7.8, 3.9 Hz, 1H), 1.99 – 1.89 (m, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.21, 150.57, 147.09, 138.15, 134.24, 129.14, 129.00, 128.78, 128.38, 127.36, 126.73, 124.47, 120.01, 114.98, 114.69, 72.94, 56.41, 55.41, 29.43, 27.45.

7-methoxy-2-phenyl-3-(p-tolyl)-3,3a,4,5-tetrahydro-2H-benzo[g]indazole (4ad)

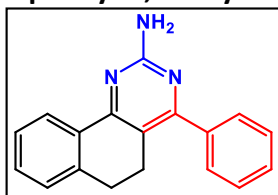


Brown liquid, Yield: 81%, 149 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.00 (d, J = 8.7 Hz, 1H), 7.37 – 7.35 (m, 2H), 7.20 (d, J = 7.8 Hz, 2H), 7.18 – 7.13 (m, 2H), 7.09 – 7.06 (m, 2H), 6.84 – 6.78 (m, 2H), 6.66 (d, J = 2.5 Hz, 1H), 4.53 (d, J = 12.1 Hz, 1H), 3.82 (s, 3H), 3.21 (td, J = 12.8, 4.9 Hz, 1H), 2.94 – 2.83 (m, 2H), 2.37 (s, 3H), 2.28 (dtd, J = 12.1, 4.6, 2.7 Hz, 1H), 1.97 – 1.88 (m, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.45, 150.66, 147.42, 140.04, 139.38, 137.40, 129.97, 128.76, 126.17, 121.27, 119.79, 114.93, 113.42, 113.31, 73.08, 56.57, 55.38, 29.80, 27.38, 21.25.

11. ^1H and ^{13}C NMR characterization of 2,4,5,6-tetrasubstituted pyrimidines derivatives

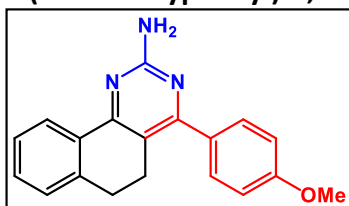
The NMR spectroscopic data for all the reported Tetrapyrimidine derivatives are in agreement with the previously reported literatures.^{9–12}

4-phenyl-5,6-dihydrobenzo[h]quinazolin-2-amine (8a)



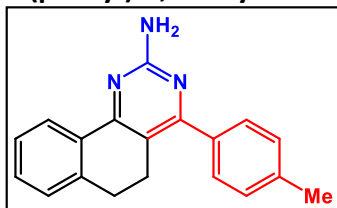
Colourless liquid, Yield: 88%, 120 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.30 – 8.25 (m, 1H), 7.56 – 7.53 (m, 2H), 7.48 – 7.43 (m, 3H), 7.39 – 7.34 (m, 2H), 7.23 – 7.20 (m, 1H), 5.27 (s, 2H), 2.85 (ddd, J = 7.9, 5.1, 2.0 Hz, 2H), 2.82 – 2.78 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.62, 161.82, 161.24, 139.66, 138.41, 133.28, 130.68, 129.07, 128.71, 128.39, 127.81, 127.15, 125.71, 116.18, 28.41, 24.19.

4-(4-methoxyphenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8b)



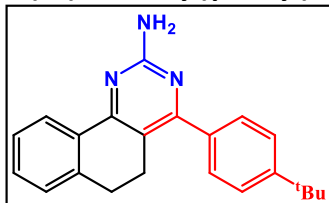
White solid, Yield: 92%, 140 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.28 – 8.22 (m, 1H), 7.54 – 7.50 (m, 2H), 7.38 – 7.33 (m, 2H), 7.22 – 7.19 (m, 1H), 7.00 – 6.96 (m, 2H), 5.19 (s, 2H), 3.85 (s, 3H), 2.89 – 2.85 (m, 2H), 2.82 – 2.78 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.19, 161.77, 161.10, 160.36, 139.57, 133.40, 130.82, 130.56, 130.30, 127.74, 127.12, 125.66, 116.05, 113.75, 55.46, 28.46, 24.40.

4-(p-tolyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8c)



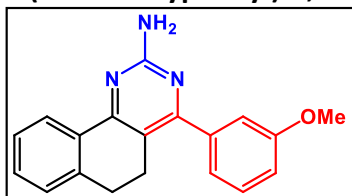
White solid, Yield: 91%, 131 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.28 – 8.24 (m, 1H), 7.45 (dt, J = 8.3, 2.0 Hz, 2H), 7.38 – 7.33 (m, 2H), 7.27 (dd, J = 8.5, 0.7 Hz, 2H), 7.22 – 7.19 (m, 1H), 5.32 (s, 2H), 2.85 (ddd, J = 8.7, 5.9, 1.7 Hz, 2H), 2.82 – 2.77 (m, 2H), 2.41 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.64, 161.84, 161.10, 139.65, 139.08, 135.52, 133.37, 130.59, 129.04, 128.69, 127.78, 127.12, 125.67, 116.11, 28.44, 24.27, 21.48.

4-(4-(tert-butyl)phenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8d)



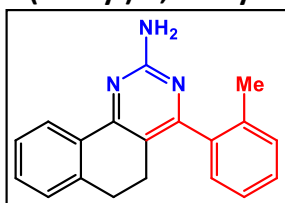
White solid, Yield: 93%, 153 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.29 – 8.24 (m, 1H), 7.52 – 7.47 (m, 4H), 7.38 – 7.34 (m, 2H), 7.23 – 7.20 (m, 1H), 5.23 (s, 2H), 2.91 – 2.87 (m, 2H), 2.80 (dd, J = 8.6, 5.7 Hz, 2H), 1.35 (s, 9H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.63, 161.80, 161.09, 152.25, 139.66, 135.52, 133.37, 130.60, 128.47, 127.79, 127.13, 125.66, 125.35, 116.21, 34.86, 31.39, 28.46, 24.28.

4-(3-methoxyphenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8e)



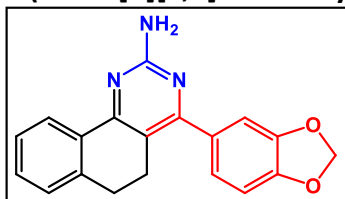
White solid, Yield: 90%, 137 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.28 – 8.23 (m, 1H), 7.39 – 7.33 (m, 3H), 7.23 – 7.20 (m, 1H), 7.11 – 7.08 (m, 2H), 7.01 – 6.95 (m, 1H), 5.24 (s, 2H), 3.85 (s, 3H), 2.84 (ddd, J = 7.8, 4.9, 2.0 Hz, 2H), 2.82 – 2.78 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.47, 161.73, 161.28, 159.60, 139.68, 133.23, 130.71, 129.44, 127.81, 127.14, 125.70, 121.07, 116.25, 114.92, 114.05, 55.45, 28.39, 24.18.

4-(o-tolyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8f)



White solid, Yield: 88%, 127 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.32 – 8.27 (m, 1H), 7.39 – 7.34 (m, 2H), 7.33 – 7.30 (m, 1H), 7.29 – 7.24 (m, 2H), 7.23 – 7.18 (m, 2H), 5.45 (d, J = 17.8 Hz, 2H), 2.80 (t, J = 7.3 Hz, 2H), 2.52 (d, J = 3.6 Hz, 2H), 2.21 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 166.79, 161.84, 160.79, 139.84, 138.03, 135.35, 133.16, 130.76, 130.40, 128.61, 128.05, 127.98, 127.16, 125.94, 125.67, 116.75, 28.27, 23.21, 19.52.

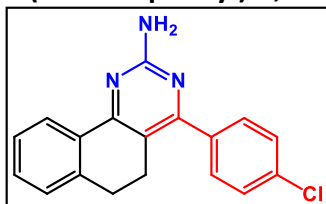
4-(benzo[d][1,3]dioxol-5-yl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8g)



White solid, Yield: 87%, 138 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.27 – 8.21 (m, 1H), 7.35 (pd, J = 7.5, 2.0 Hz, 2H), 7.24 – 7.18 (m, 1H), 7.08 – 7.02 (m, 2H), 6.89 (d, J = 8.0 Hz, 1H), 6.00 (s, 2H), 5.16 (s, 2H), 2.89 – 2.84 (m, 2H), 2.82 – 2.76 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ

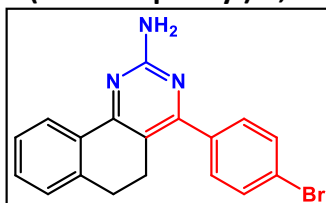
164.99, 161.68, 161.27, 148.36, 147.68, 139.58, 133.30, 132.31, 130.65, 127.77, 127.14, 125.68, 123.00, 116.07, 109.37, 108.20, 101.40, 28.42, 24.38.

4-(4-chlorophenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8h)



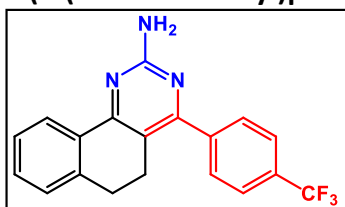
White solid, Yield: 84%, 129 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.24 (ddd, J = 9.6, 3.7, 2.0 Hz, 1H), 7.51 – 7.47 (m, 2H), 7.45 – 7.40 (m, 2H), 7.40 – 7.31 (m, 2H), 7.24 – 7.19 (m, 1H), 5.13 (s, 2H), 2.83 (dd, J = 6.5, 3.1 Hz, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.32, 161.73, 161.48, 139.56, 136.79, 135.20, 133.10, 130.83, 130.18, 128.63, 127.84, 127.22, 125.72, 116.18, 28.32, 24.18.

4-(4-bromophenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8i)



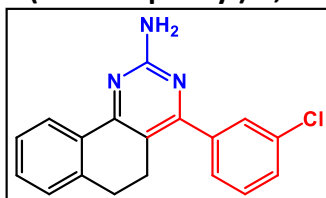
Brown liquid, Yield: 88%, 155mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.28 – 8.23 (m, 1H), 7.61 – 7.58 (m, 2H), 7.44 – 7.41 (m, 2H), 7.37 (ddd, J = 6.6, 4.2, 1.8 Hz, 2H), 7.23 – 7.20 (m, 1H), 5.12 (s, 2H), 2.83 – 2.80 (m, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.34, 161.73, 161.51, 139.55, 137.27, 133.09, 131.58, 130.84, 130.44, 127.83, 127.21, 125.73, 123.48, 116.14, 28.31, 24.16.

4-(4-(trifluoromethyl)phenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8j)



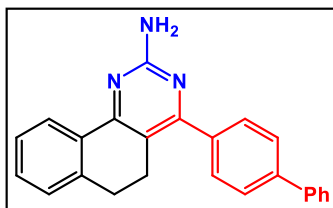
Brown liquid, Yield: 86%, 147 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.30 – 8.24 (m, 1H), 7.74 (s, 2H), 7.66 (d, J = 8.0 Hz, 2H), 7.41 – 7.35 (m, 2H), 7.24 – 7.21 (m, 1H), 5.25 (s, 2H), 2.82 (s, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.05, 161.82, 161.67, 141.95, 139.58, 132.98, 131.17, 130.95, 129.17, 127.89, 127.26, 125.40 (q, J = 3.8 Hz), 125.17, 123.01, 116.20, 28.26, 24.05.

4-(3-chlorophenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8k)



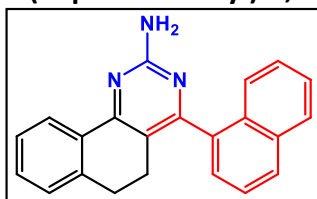
Brown liquid, Yield: 89%, 137 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.28 – 8.24 (m, 1H), 7.55 (td, J = 1.8, 0.6 Hz, 1H), 7.43 – 7.33 (m, 5H), 7.23 – 7.20 (m, 1H), 5.26 (s, 2H), 2.81 (s, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.04, 161.81, 161.55, 140.18, 139.60, 134.43, 133.07, 130.86, 129.67, 129.16, 128.90, 127.86, 127.20, 126.91, 125.75, 116.13, 28.31, 24.09.

4-([1,1'-biphenyl]-4-yl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8l)



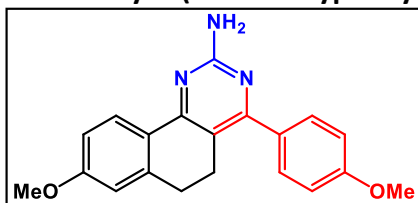
White solid, Yield: 85%, 149 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.31 – 8.28 (m, 1H), 7.70 (dq, J = 3.8, 1.4 Hz, 2H), 7.66 – 7.63 (m, 4H), 7.49 – 7.45 (m, 2H), 7.40 – 7.36 (m, 3H), 7.25 – 7.22 (m, 1H), 5.29 (s, 2H), 2.94 – 2.90 (m, 2H), 2.83 (dd, J = 8.5, 5.7 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.23, 161.87, 161.30, 141.96, 140.65, 139.67, 137.31, 133.31, 130.71, 129.28, 128.98, 127.83, 127.74, 127.29, 127.18, 127.12, 125.74, 116.26, 28.44, 24.31.

4-(naphthalen-1-yl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8m)



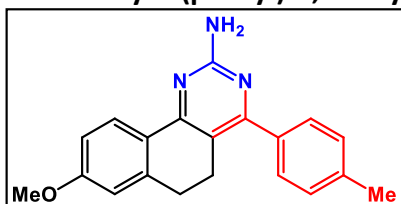
Colourless liquid, Yield: 86%, 139 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.39 – 8.34 (m, 1H), 7.92 (t, J = 7.4 Hz, 2H), 7.67 (d, J = 8.2 Hz, 1H), 7.56 (dd, J = 8.2, 7.0 Hz, 1H), 7.52 – 7.47 (m, 2H), 7.43 (ddd, J = 8.2, 6.8, 1.3 Hz, 1H), 7.41 – 7.35 (m, 2H), 7.20 – 7.17 (m, 1H), 5.54 (s, 2H), 2.74 (t, J = 7.3 Hz, 2H), 2.47 (dd, J = 13.9, 6.9 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.70, 162.05, 160.89, 139.94, 136.07, 133.65, 133.16, 130.86, 130.82, 129.08, 128.58, 128.03, 127.21, 126.73, 126.26, 126.17, 125.80, 125.47, 125.42, 117.66, 28.28, 23.58.

8-methoxy-4-(4-methoxyphenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8n)



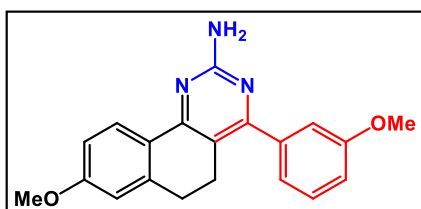
Brown liquid, Yield: 92%, 153 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.19 (d, J = 8.6 Hz, 1H), 7.52 – 7.49 (m, 2H), 6.99 – 6.95 (m, 2H), 6.87 (dd, J = 8.6, 2.6 Hz, 1H), 6.72 (d, J = 2.5 Hz, 1H), 5.19 (s, 2H), 3.84 (s, 3H), 3.83 (s, 3H), 2.87 – 2.83 (m, 2H), 2.76 (dd, J = 8.5, 5.6 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.69, 161.70, 161.59, 161.06, 160.25, 141.63, 130.95, 130.25, 127.51, 126.31, 115.07, 113.71, 112.94, 112.61, 55.45, 55.41, 28.83, 24.40.

8-methoxy-4-(p-tolyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8o)



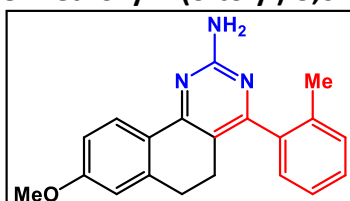
Brown liquid, Yield: 90%, 143 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.20 (d, J = 8.6 Hz, 1H), 7.44 – 7.42 (m, 2H), 7.27 – 7.24 (m, 2H), 6.87 (dd, J = 8.6, 2.6 Hz, 1H), 6.72 (d, J = 2.6 Hz, 1H), 5.29 (s, 2H), 3.83 (s, 3H), 2.85 – 2.81 (m, 2H), 2.78 – 2.74 (m, 2H), 2.40 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.14, 161.81, 161.78, 161.61, 161.06, 141.70, 138.91, 135.66, 128.99, 128.67, 127.54, 126.31, 115.11, 112.97, 112.60, 55.41, 28.81, 24.28, 21.46.

8-methoxy-4-(3-methoxyphenyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8p)



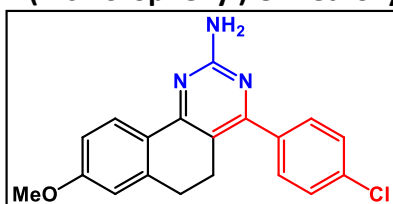
Brown liquid, Yield: 87%, 145 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.20 (d, J = 8.6 Hz, 1H), 7.35 (ddd, J = 8.0, 7.1, 0.9 Hz, 1H), 7.09 – 7.07 (m, 2H), 6.96 (ddd, J = 8.3, 2.4, 1.2 Hz, 1H), 6.87 (dd, J = 8.6, 2.6 Hz, 1H), 6.72 (d, J = 2.5 Hz, 1H), 5.12 (s, 2H), 3.84 (d, J = 0.9 Hz, 6H), 2.83 (ddd, J = 8.3, 5.5, 1.7 Hz, 2H), 2.79 – 2.75 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.98, 161.71, 161.62, 161.24, 159.59, 141.74, 139.85, 129.41, 127.57, 126.15, 121.04, 115.31, 114.81, 114.01, 112.96, 112.69, 55.47, 55.43, 28.76, 24.19.

8-methoxy-4-(*o*-tolyl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8q)



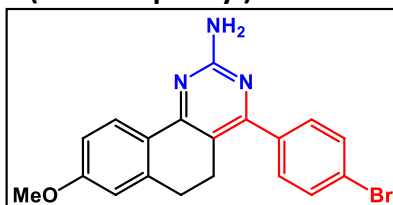
White solid, Yield: 91%, 145 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.22 (d, J = 8.6 Hz, 1H), 7.31 – 7.23 (m, 3H), 7.20 – 7.17 (m, 1H), 6.87 (dd, J = 8.7, 2.6 Hz, 1H), 6.71 (d, J = 2.6 Hz, 1H), 5.32 (s, 2H), 3.83 (s, 3H), 2.76 (t, J = 7.3 Hz, 2H), 2.50 (dd, J = 11.4, 6.3 Hz, 2H), 2.19 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 166.25, 161.73, 161.71, 160.75, 160.75, 141.91, 138.15, 135.34, 130.36, 128.52, 128.05, 127.50, 126.06, 125.90, 115.81, 113.12, 112.68, 55.43, 28.65, 23.27, 19.51.

4-(4-chlorophenyl)-8-methoxy-5,6-dihydrobenzo[h]quinazolin-2-amine (8r)



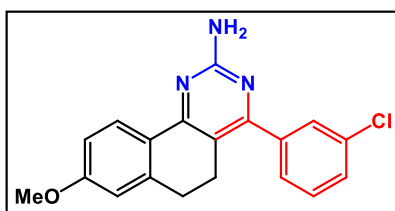
Pale yellow solid, Yield: 85%, 144 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.19 (d, J = 8.6 Hz, 1H), 7.49 – 7.45 (m, 2H), 7.43 – 7.40 (m, 2H), 6.86 (dd, J = 8.7, 2.6 Hz, 1H), 6.72 (d, J = 2.6 Hz, 1H), 5.28 (s, 2H), 3.83 (s, 3H), 2.81 – 2.74 (m, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.79, 161.77, 161.75, 161.41, 141.65, 136.94, 135.02, 130.17, 128.57, 127.62, 126.02, 115.09, 112.99, 112.72, 55.43, 28.70, 24.18.

4-(4-bromophenyl)-8-methoxy-5,6-dihydrobenzo[h]quinazolin-2-amine (8s)



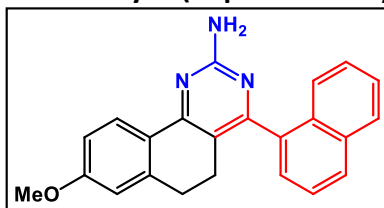
Pale yellow solid, Yield: 86%, 165 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.19 (d, J = 8.6 Hz, 1H), 7.59 – 7.56 (m, 2H), 7.42 – 7.39 (m, 2H), 6.87 (dd, J = 8.6, 2.6 Hz, 1H), 6.72 (d, J = 2.6 Hz, 1H), 5.19 (s, 2H), 3.84 (s, 3H), 2.81 – 2.75 (m, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.84, 161.78, 161.72, 161.43, 141.64, 137.43, 131.54, 130.43, 127.62, 126.01, 123.31, 115.11, 112.98, 112.74, 55.44, 28.69, 24.17.

4-(3-chlorophenyl)-8-methoxy-5,6-dihydrobenzo[h]quinazolin-2-amine (8t)



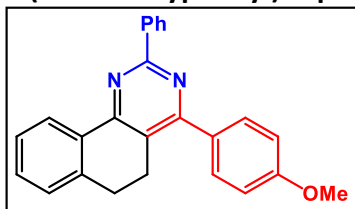
Colourless liquid, Yield: 88%, 149 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.20 (d, J = 8.7 Hz, 1H), 7.59 – 7.49 (m, 1H), 7.42 – 7.34 (m, 2H), 6.87 (dd, J = 8.8, 2.8 Hz, 1H), 6.72 (d, J = 2.9 Hz, 1H), 5.20 (s, 2H), 3.84 (s, 3H), 2.88 – 2.68 (m, 4H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.55, 161.81, 161.70, 161.49, 141.68, 140.32, 134.39, 129.64, 129.04, 128.86, 127.63, 126.88, 125.96, 115.19, 112.99, 112.75, 55.44, 28.67, 24.10.

8-methoxy-4-(naphthalen-1-yl)-5,6-dihydrobenzo[h]quinazolin-2-amine (8u)



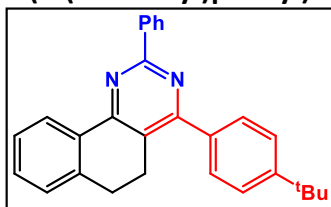
White solid, Yield: 86%, 152 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.28 (d, J = 8.7 Hz, 1H), 7.92 – 7.89 (m, 2H), 7.66 (d, J = 8.4 Hz, 1H), 7.55 (dd, J = 8.2, 7.0 Hz, 1H), 7.50 – 7.40 (m, 3H), 6.90 (dd, J = 8.7, 2.6 Hz, 1H), 6.70 (d, J = 2.6 Hz, 1H), 5.34 (s, 2H), 3.83 (s, 3H), 2.71 (t, J = 7.3 Hz, 2H), 2.45 (td, J = 6.7, 3.1 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.13, 161.83, 160.87, 141.99, 136.16, 133.64, 130.83, 128.95, 128.51, 127.63, 126.65, 126.18, 126.10, 126.01, 125.47, 125.40, 116.82, 113.13, 112.75, 55.43, 28.63, 23.61.

4-(4-methoxyphenyl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8v)



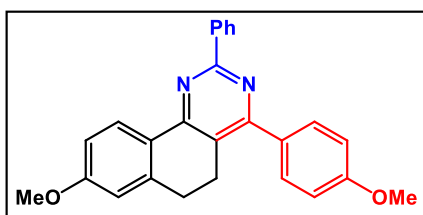
White solid, Yield: 94%, 171 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.69 (dd, J = 6.8, 1.4 Hz, 2H), 8.61 (d, J = 7.4 Hz, 1H), 7.74 – 7.71 (m, 2H), 7.55 – 7.40 (m, 5H), 7.25 (d, J = 7.0 Hz, 1H), 7.07 – 7.03 (m, 2H), 3.88 (s, 3H), 3.11 – 3.06 (m, 2H), 2.90 – 2.86 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.88, 162.08, 160.62, 160.09, 139.15, 138.46, 133.61, 130.94, 130.86, 130.81, 130.29, 128.49, 128.27, 127.80, 127.34, 126.12, 123.28, 113.79, 55.52, 27.97, 25.07.

4-(4-(tert-butyl)phenyl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8w)



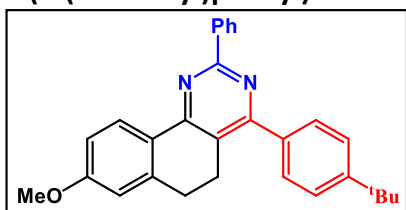
White solid, Yield: 92%, 180 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.70 – 8.67 (m, 2H), 8.63 – 8.60 (m, 1H), 7.72 – 7.69 (m, 2H), 7.58 – 7.55 (m, 2H), 7.54 – 7.47 (m, 3H), 7.46 – 7.40 (m, 2H), 7.28 – 7.25 (m, 1H), 3.14 – 3.10 (m, 2H), 2.92 – 2.87 (m, 2H), 1.41 (s, 9H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.30, 162.16, 160.10, 152.54, 139.22, 138.41, 135.58, 133.58, 130.84, 130.28, 129.14, 128.47, 128.28, 127.82, 127.35, 126.13, 125.38, 123.52, 34.94, 31.45, 27.98, 24.97.

8-methoxy-4-(4-methoxyphenyl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8x)



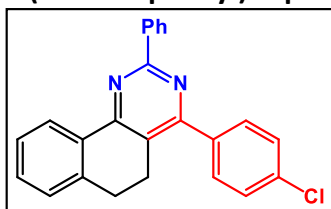
White solid, Yield: 91%, 180 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.66 – 8.63 (m, 2H), 8.53 (d, J = 8.6 Hz, 1H), 7.72 – 7.68 (m, 2H), 7.52 – 7.45 (m, 3H), 7.05 – 7.01 (m, 2H), 6.96 (dd, J = 8.6, 2.6 Hz, 1H), 6.75 (d, J = 2.5 Hz, 1H), 3.87 (s, 3H), 3.86 (s, 3H), 3.08 – 3.04 (m, 2H), 2.87 – 2.82 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.43, 161.94, 161.80, 160.50, 160.03, 141.17, 138.58, 131.03, 130.87, 130.15, 128.42, 128.21, 127.94, 126.51, 122.21, 113.74, 112.93, 112.86, 55.50, 55.45, 28.34, 25.04.

4-(4-(tert-butyl)phenyl)-8-methoxy-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8y)



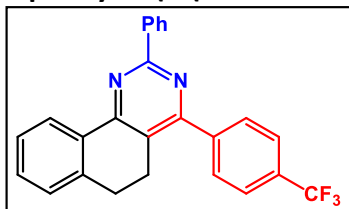
White solid, Yield: 94%, 198 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.69 – 8.64 (m, 2H), 8.55 (d, J = 8.6 Hz, 1H), 7.70 – 7.67 (m, 2H), 7.56 – 7.53 (m, 2H), 7.53 – 7.47 (m, 3H), 6.97 (dd, J = 8.6, 2.6 Hz, 1H), 6.76 (d, J = 2.5 Hz, 1H), 3.87 (s, 3H), 3.11 – 3.07 (m, 2H), 2.87 – 2.83 (m, 2H), 1.41 (s, 9H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.84, 162.02, 161.83, 160.04, 152.37, 141.25, 138.56, 135.73, 130.16, 129.10, 128.41, 128.25, 127.96, 126.50, 125.33, 122.46, 112.96, 112.88, 55.46, 34.91, 31.45, 28.34, 24.95.

4-(4-chlorophenyl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8z)



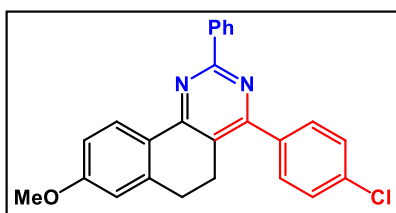
White solid, Yield: 85%, 157 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.68 – 8.64 (m, 2H), 8.60 (dd, J = 7.4, 1.7 Hz, 1H), 7.68 – 7.65 (m, 2H), 7.54 – 7.48 (m, 5H), 7.44 (pd, J = 7.6, 1.7 Hz, 2H), 7.26 – 7.23 (m, 1H), 3.02 (dd, J = 8.5, 5.9 Hz, 2H), 2.90 – 2.86 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.11, 162.28, 160.40, 139.12, 138.16, 136.84, 135.49, 133.31, 131.05, 130.77, 130.47, 128.64, 128.54, 128.26, 127.87, 127.42, 126.20, 123.46, 27.82, 24.84.

2-phenyl-4-(4-(trifluoromethyl)phenyl)-5,6-dihydrobenzo[h]quinazoline (8aa)



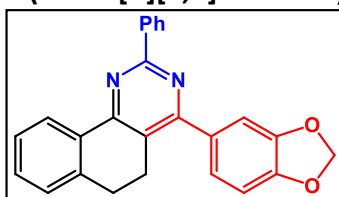
White solid, Yield: 86%, 173 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.66 – 8.62 (m, 2H), 8.62 – 8.59 (m, 1H), 7.81 (d, J = 8.4 Hz, 2H), 7.67 (d, J = 8.0 Hz, 1H), 7.53 – 7.42 (m, 5H), 7.26 (dd, J = 5.7, 1.5 Hz, 1H), 6.74 (t, J = 56.4 Hz, 1H), 3.08 – 3.01 (m, 2H), 2.93 – 2.88 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.36, 162.35, 160.46, 140.89, 140.87, 140.84, 139.16, 138.10, 135.30, 135.12, 134.94, 133.26, 131.10, 130.49, 129.69, 128.54, 128.25, 127.89, 127.44, 126.19, 125.67 (q, 6.3 Hz), 123.62, 116.48, 114.58, 112.68, 27.80, 24.76.

4-(4-chlorophenyl)-8-methoxy-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ab)



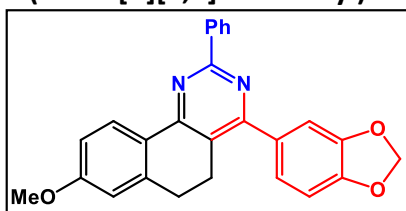
White solid, Yield: 83%, 166 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.64 – 8.58 (m, 2H), 8.53 (d, J = 8.6 Hz, 1H), 7.68 – 7.62 (m, 2H), 7.52 – 7.44 (m, 5H), 6.96 (dd, J = 8.6, 2.6 Hz, 1H), 6.75 (d, J = 2.5 Hz, 1H), 3.87 (s, 3H), 3.01 (dd, J = 8.7, 5.9 Hz, 2H), 2.88 – 2.84 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 162.68, 162.16, 161.99, 160.33, 141.17, 138.29, 137.01, 135.32, 130.71, 130.32, 128.59, 128.46, 128.20, 128.04, 126.23, 122.38, 113.05, 112.91, 55.47, 28.20, 24.83.

4-(benzo[d][1,3]dioxol-5-yl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ac)



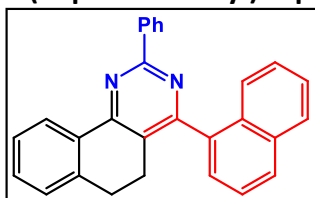
White solid, Yield: 87%, 165 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.69 – 8.65 (m, 2H), 8.63 – 8.57 (m, 1H), 7.55 – 7.40 (m, 5H), 7.30 (d, J = 1.6 Hz, 1H), 7.25 (d, J = 7.0 Hz, 1H), 7.21 (dd, J = 8.0, 1.6 Hz, 1H), 6.94 (d, J = 8.0 Hz, 1H), 6.04 (s, 2H), 3.09 – 3.05 (m, 2H), 2.91 – 2.86 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.70, 162.08, 160.24, 148.65, 147.83, 139.14, 138.32, 133.52, 132.40, 130.87, 130.35, 128.50, 128.25, 127.80, 127.35, 126.14, 123.69, 123.30, 109.97, 108.15, 101.52, 27.94, 25.08.

4-(benzo[d][1,3]dioxol-5-yl)-8-methoxy-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ad)



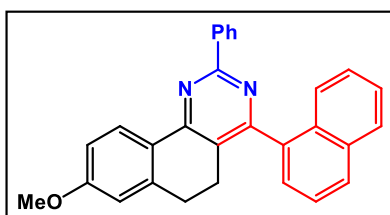
White solid, Yield: 88%, 180 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.64 – 8.58 (m, 2H), 8.51 (d, J = 8.6 Hz, 1H), 7.54 – 7.44 (m, 3H), 7.26 (d, J = 1.5 Hz, 1H), 7.19 (dd, J = 8.0, 1.7 Hz, 1H), 6.97 – 6.92 (m, 2H), 6.76 (d, J = 2.5 Hz, 1H), 6.04 (s, 2H), 3.87 (s, 3H), 3.08 – 3.04 (m, 2H), 2.88 – 2.84 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 163.30, 161.98, 161.85, 160.18, 148.52, 147.78, 141.18, 138.42, 132.57, 130.20, 128.42, 128.18, 127.98, 126.44, 123.57, 122.23, 112.96, 112.87, 109.92, 108.12, 101.45, 55.47, 28.32, 25.06.

4-(naphthalen-1-yl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ae)



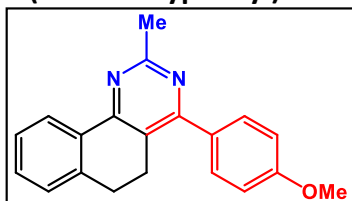
White solid, Yield: 89%, 171 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.69 (dd, J = 7.6, 1.3 Hz, 1H), 8.66 – 8.61 (m, 2H), 7.97 (dd, J = 14.0, 8.2 Hz, 2H), 7.69 (dd, J = 8.4, 0.9 Hz, 1H), 7.61 (dd, J = 8.1, 7.0 Hz, 1H), 7.57 (dd, J = 7.0, 1.4 Hz, 1H), 7.54 – 7.42 (m, 8H), 2.84 (t, J = 7.3 Hz, 2H), 2.72 – 2.66 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.00, 162.48, 159.81, 139.46, 138.33, 136.13, 133.75, 133.31, 131.11, 131.08, 130.37, 129.27, 128.61, 128.51, 128.39, 128.02, 127.41, 126.80, 126.72, 126.23, 126.15, 125.54, 125.40, 125.33, 27.72, 24.27.

8-methoxy-4-(naphthalen-1-yl)-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8af)



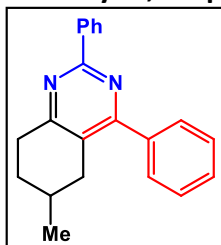
White solid, Yield: 87%, 180 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.66 – 8.60 (m, 3H), 7.96 (ddd, J = 12.9, 8.3, 1.0 Hz, 2H), 7.71 (ddd, J = 11.0, 7.1, 2.3 Hz, 1H), 7.61 (dd, J = 8.3, 7.0 Hz, 1H), 7.57 – 7.42 (m, 6H), 7.00 (dd, J = 8.8, 2.8 Hz, 1H), 6.75 (d, J = 2.8 Hz, 1H), 3.88 (s, 3H), 2.84 – 2.78 (m, 2H), 2.71 – 2.63 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.48, 162.36, 162.04, 159.78, 141.54, 138.49, 136.29, 133.76, 131.16, 130.27, 129.18, 128.59, 128.48, 128.38, 128.02, 126.77, 126.67, 126.24, 126.20, 125.61, 125.41, 124.28, 113.10, 113.06, 55.49, 28.13, 24.32.

4-(4-methoxyphenyl)-2-methyl-5,6-dihydrobenzo[h]quinazoline (8ag)



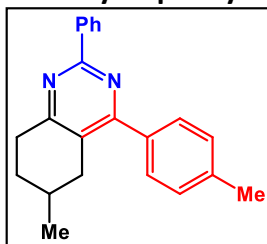
White solid, Yield: 91%, 138 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.41 – 8.35 (m, 1H), 7.59 – 7.53 (m, 2H), 7.40 – 7.36 (m, 2H), 7.23 – 7.20 (m, 1H), 7.02 – 6.98 (m, 2H), 3.85 (s, 3H), 2.98 (dd, J = 8.4, 6.1 Hz, 2H), 2.85 – 2.81 (m, 2H), 2.80 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 165.72, 163.94, 160.49, 159.96, 139.19, 133.26, 130.78, 130.60, 130.56, 127.79, 127.36, 125.91, 122.28, 113.87, 55.48, 27.95, 26.27, 24.74.

6-methyl-2,4-diphenyl-5,6,7,8-tetrahydroquinazoline (8ah)



White solid, Yield: 70%, 105 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.50 – 8.45 (m, 2H), 7.69 – 7.65 (m, 2H), 7.53 – 7.43 (m, 6H), 3.14 (ddd, J = 19.2, 6.4, 3.1 Hz, 1H), 3.08 – 2.94 (m, 1H), 2.84 – 2.76 (m, 1H), 2.50 – 2.38 (m, 1H), 2.08 – 1.97 (m, 1H), 1.85 – 1.74 (m, 1H), 1.64 – 1.52 (m, 1H), 1.06 (d, J = 6.7 Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 166.70, 165.13, 161.55, 138.76, 138.26, 130.12, 129.19, 129.10, 128.50, 128.32, 128.16, 125.01, 35.38, 32.68, 30.69, 29.34, 21.77.

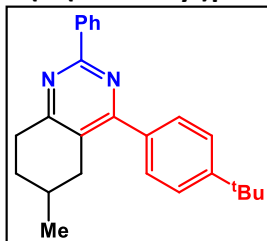
6-methyl-2-phenyl-4-(p-tolyl)-5,6,7,8-tetrahydroquinazoline (8ai)



White solid, Yield: 72%, 113 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.50 – 8.43 (m, 2H), 7.60 – 7.56 (m, 2H), 7.48 – 7.41 (m, 3H), 7.33 – 7.28 (m, 2H), 3.13 (ddd, J = 18.6, 5.9, 2.7 Hz, 1H), 3.05 – 2.96 (m, 1H), 2.82 (ddd, J = 16.7, 4.4, 1.9 Hz, 1H), 2.48 (t, J = 8.3 Hz, 1H), 2.44 (s, 3H), 2.02 (dddd, J = 11.5, 9.1, 5.1, 2.7 Hz, 1H), 1.85 – 1.74 (m, 1H), 1.58 (dtd, J = 13.1, 11.3, 6.0 Hz, 1H), 1.06 (d, J = 6.6 Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 166.53, 165.09, 161.48, 139.13,

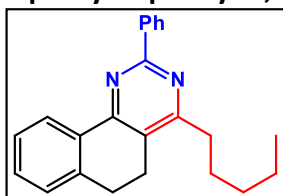
138.33, 135.91, 130.05, 129.18, 129.00, 128.46, 128.13, 124.91, 35.49, 32.67, 30.71, 29.38, 21.78, 21.51.

4-(4-(tert-butyl)phenyl)-6-methyl-2-phenyl-5,6,7,8-tetrahydroquinazoline (8aj)



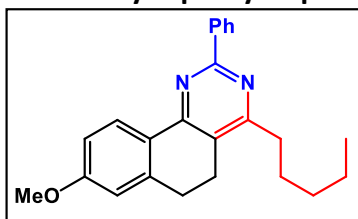
White solid, Yield: 75%, 134 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.46 (dt, $J = 4.3, 2.4$ Hz, 2H), 7.65 – 7.61 (m, 2H), 7.53 – 7.50 (m, 2H), 7.47 – 7.42 (m, 3H), 3.13 (ddd, $J = 18.5, 5.9, 2.7$ Hz, 1H), 3.06 – 2.95 (m, 1H), 2.86 (ddd, $J = 16.6, 4.3, 1.8$ Hz, 1H), 2.49 (dd, $J = 16.7, 10.6$ Hz, 1H), 2.05 – 1.98 (m, 1H), 1.85 – 1.74 (m, 1H), 1.64 – 1.54 (m, 1H), 1.38 (s, 9H), 1.07 (d, $J = 6.6$ Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 166.53, 164.95, 161.48, 152.23, 138.33, 135.84, 130.03, 129.01, 128.44, 128.14, 125.27, 124.95, 77.39, 77.13, 76.88, 35.49, 34.88, 32.66, 31.41, 30.72, 29.39, 21.77.

4-pentyl-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ak)



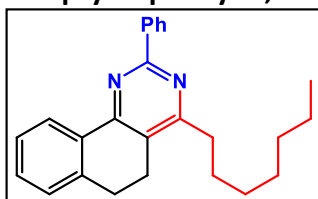
White solid, Yield: 74%, 122 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.65 – 8.57 (m, 2H), 8.57 – 8.54 (m, 1H), 7.53 – 7.44 (m, 3H), 7.43 – 7.37 (m, 2H), 7.27 – 7.24 (m, 1H), 3.01 – 2.92 (m, 4H), 2.91 – 2.83 (m, 2H), 1.82 (dt, $J = 15.3, 7.5$ Hz, 2H), 1.48 – 1.37 (m, 4H), 0.93 (t, $J = 7.0$ Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 168.01, 161.93, 158.43, 138.99, 138.69, 133.42, 130.60, 130.07, 128.47, 128.12, 127.84, 127.28, 125.97, 123.81, 34.83, 31.87, 28.08, 27.66, 22.89, 22.71, 14.20.

8-methoxy-4-pentyl-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8al)



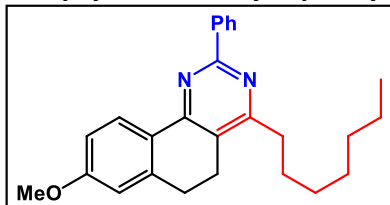
White solid, Yield: 76%, 136 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.63 (dd, $J = 11.0, 4.7$ Hz, 2H), 8.50 (d, $J = 8.6$ Hz, 1H), 7.58 – 7.45 (m, 3H), 6.93 (d, $J = 8.6$ Hz, 1H), 6.74 (s, 1H), 3.85 (s, 3H), 2.92 (s, 4H), 2.84 (t, $J = 7.8$ Hz, 2H), 1.89 – 1.77 (m, 2H), 1.43 (dd, $J = 10.9, 7.5$ Hz, 4H), 0.97 (d, $J = 6.3$ Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 167.45, 161.78, 161.62, 158.42, 141.00, 138.89, 129.99, 128.45, 128.13, 127.77, 126.33, 122.72, 112.92, 112.83, 55.41, 34.79, 31.93, 28.11, 28.01, 22.91, 22.75, 14.25.

4-heptyl-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8am)



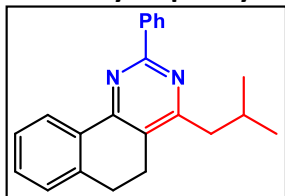
White solid, Yield: 73%, 130 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.69 – 8.63 (m, 2H), 8.59 (d, J = 7.2 Hz, 1H), 7.59 – 7.48 (m, 3H), 7.47 – 7.34 (m, 2H), 7.25 (d, J = 6.6 Hz, 1H), 3.03 – 2.91 (m, 4H), 2.91 – 2.84 (m, 2H), 1.88 – 1.78 (m, 2H), 1.51 – 1.31 (m, 8H), 0.99 – 0.91 (m, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 130.61, 130.11, 128.50, 128.18, 127.86, 127.28, 126.00, 123.82, 34.88, 32.02, 29.73, 29.41, 28.41, 27.66, 22.89, 22.86, 14.32.

4-heptyl-8-methoxy-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8an)



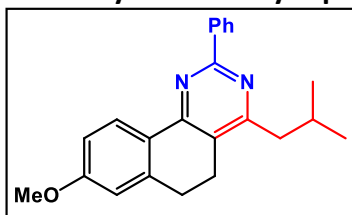
White solid, Yield: 75%, 145 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.62 (t, J = 8.9 Hz, 2H), 8.50 (d, J = 8.6 Hz, 1H), 7.56 – 7.45 (m, 3H), 6.93 (d, J = 8.6 Hz, 1H), 6.74 (s, 1H), 3.85 (s, 3H), 2.97 – 2.88 (m, 4H), 2.84 (t, J = 7.7 Hz, 2H), 1.80 (dd, J = 14.8, 7.8 Hz, 2H), 1.47 – 1.31 (m, 8H), 0.92 (t, J = 6.3 Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 167.46, 161.78, 161.62, 158.42, 141.00, 138.87, 129.98, 128.44, 128.12, 127.77, 126.34, 122.71, 112.92, 112.82, 55.41, 34.83, 32.00, 29.72, 29.38, 28.44, 28.01, 22.92, 22.83, 14.29.

4-isobutyl-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ao)



White solid, Yield: 71%, 112 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.64 (ddd, J = 4.3, 3.6, 2.0 Hz, 2H), 8.59 (dd, J = 7.1, 1.9 Hz, 1H), 7.57 – 7.48 (m, 3H), 7.46 – 7.39 (m, 2H), 7.28 – 7.24 (m, 1H), 2.97 (s, 4H), 2.78 (d, J = 7.2 Hz, 2H), 2.38 – 2.28 (m, 1H), 1.06 (dd, J = 6.6, 1.7 Hz, 6H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 167.24, 161.79, 158.57, 139.05, 138.74, 133.48, 130.65, 130.12, 128.51, 128.17, 127.86, 127.30, 126.02, 124.34, 43.41, 28.49, 27.69, 23.17, 22.80.

4-isobutyl-8-methoxy-2-phenyl-5,6-dihydrobenzo[h]quinazoline (8ap)

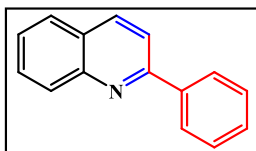


White solid, Yield: 74%, 128 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.70 – 8.62 (m, 2H), 8.54 (d, J = 8.5 Hz, 1H), 7.59 – 7.48 (m, 3H), 6.95 (dd, J = 8.6, 2.5 Hz, 1H), 6.74 (s, 1H), 3.84 (s, 3H), 2.91 (s, 4H), 2.75 (d, J = 7.2 Hz, 2H), 2.39 – 2.28 (m, 1H), 1.07 (d, J = 6.6 Hz, 6H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 166.68, 161.67, 161.62, 158.57, 141.04, 138.92, 130.04, 128.48, 128.17, 127.82, 126.38, 123.27, 112.91, 112.88, 55.41, 43.37, 28.49, 27.98, 23.20, 22.83.

12. ^1H and $^{13}\text{C}\{^1\text{H}\}$ NMR characterization of quinoline derivatives

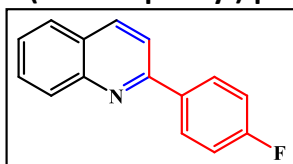
The NMR spectroscopic data for all the reported quinoline derivatives are in agreement with the previously reported literatures.^{13–15}

2-phenylquinoline (11a)



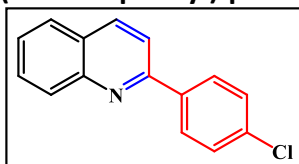
White solid, Yield: 94%, 96.5 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.22 – 8.16 (m, 4H), 7.85 (d, J = 8.6 Hz, 1H), 7.81 (dd, J = 8.1, 1.5 Hz, 1H), 7.73 (ddd, J = 8.5, 7.0, 1.5 Hz, 1H), 7.56 – 7.50 (m, 3H), 7.49 – 7.46 (m, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.48, 148.38, 139.78, 136.91, 129.82, 129.79, 129.45, 128.97, 127.70, 127.60, 127.29, 126.40, 119.14.

2-(4-fluorophenyl)quinoline (11b)



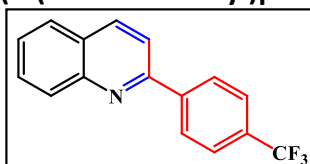
Pale yellow solid, Yield: 86%, 96 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.19 (d, J = 8.6 Hz, 1H), 8.17 – 8.12 (m, 3H), 7.81 (d, J = 8.6 Hz, 2H), 7.72 (ddd, J = 8.4, 6.9, 1.4 Hz, 1H), 7.52 (ddd, J = 8.1, 6.9, 1.2 Hz, 1H), 7.23 – 7.17 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 164.88, 162.90, 156.33, 148.30, 137.01, 135.90, 135.88, 129.79 (d, J = 22.7 Hz), 129.50 (d, J = 8.5 Hz), 127.57, 127.16, 126.44, 118.72, 115.85 (d, J = 21.8 Hz).

2-(4-chlorophenyl)quinoline (11c)



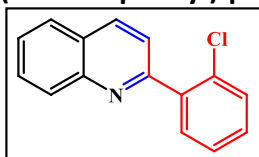
White solid, Yield: 90%, 108 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.21 (d, J = 8.2 Hz, 1H), 8.15 (ddd, J = 8.5, 1.8, 0.8 Hz, 1H), 8.12 – 8.09 (m, 2H), 7.84 – 7.81 (m, 2H), 7.73 (ddd, J = 8.4, 6.9, 1.5 Hz, 1H), 7.53 (ddd, J = 8.1, 6.9, 1.2 Hz, 1H), 7.50 – 7.47 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 156.11, 148.30, 138.13, 137.08, 135.62, 129.95, 129.76, 129.11, 128.91, 127.59, 127.30, 126.60, 118.68.

2-(4-(trifluoromethyl)phenyl)quinoline (11d)



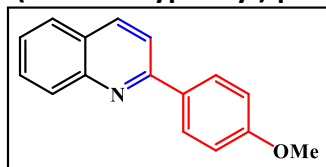
White solid, Yield: 92%, 126 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.26 (d, J = 8.1 Hz, 2H), 8.20 (dd, J = 15.1, 8.5 Hz, 2H), 7.83 (t, J = 8.0 Hz, 2H), 7.75 (ddd, J = 8.4, 5.6, 2.1 Hz, 3H), 7.55 (ddd, J = 8.0, 6.9, 1.1 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 155.71, 148.33, 143.00, 137.21, 131.26, 131.01, 130.08, 129.92, 127.91, 127.62, 127.51, 126.94, 125.82 (q, J = 3.8 Hz), 125.39, 123.23, 118.84.

2-(2-chlorophenyl)quinoline (11e)



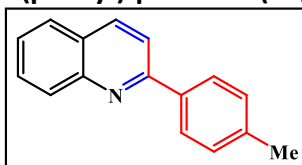
White solid, Yield: 87%, 104 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.23 (d, J = 8.4 Hz, 2H), 7.88 (dd, J = 8.2, 1.2 Hz, 1H), 7.80 – 7.75 (m, 2H), 7.73 (dd, J = 7.5, 1.8 Hz, 1H), 7.62 – 7.57 (m, 1H), 7.54 (dd, J = 7.6, 1.5 Hz, 1H), 7.42 (dq, J = 15.0, 7.4, 1.6 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.44, 148.11, 139.69, 135.69, 132.39, 131.74, 130.12, 129.91, 129.71, 127.59, 127.21, 127.15, 126.80, 122.80.

2-(4-methoxyphenyl)quinoline (11f)



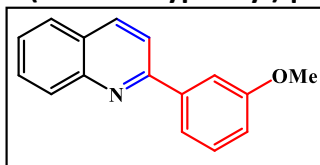
White solid, Yield: 96%, 113 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.16 – 8.12 (m, 4H), 7.80 (d, J = 8.6 Hz, 1H), 7.78 (dd, J = 8.1, 1.3 Hz, 1H), 7.70 (ddd, J = 8.4, 6.9, 1.5 Hz, 1H), 7.48 (ddd, J = 8.1, 6.9, 1.2 Hz, 1H), 7.05 – 7.02 (m, 2H), 3.86 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.91, 157.00, 148.37, 136.73, 132.33, 129.67, 129.58, 128.99, 127.54, 127.00, 126.00, 118.65, 114.32, 55.47.

2-(p-tolyl)quinoline (11g)



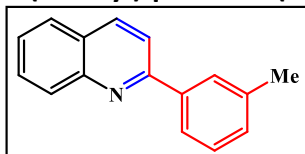
Colourless liquid, Yield: 95%, 104 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.18 (ddd, J = 8.2, 3.5, 2.6 Hz, 2H), 8.10 – 8.06 (m, 2H), 7.84 (d, J = 8.6 Hz, 1H), 7.80 (dd, J = 8.1, 1.4 Hz, 1H), 7.72 (ddd, J = 8.4, 6.9, 1.5 Hz, 1H), 7.50 (ddd, J = 8.1, 6.9, 1.2 Hz, 1H), 7.35 – 7.31 (m, 2H), 2.44 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.43, 148.38, 139.50, 136.97, 136.77, 129.74, 129.68, 127.55, 127.20, 126.18, 118.97, 21.45.

2-(3-methoxyphenyl)quinoline (11h)



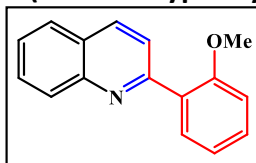
Colourless liquid, Yield: 94%, 111 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.21 – 8.17 (m, 2H), 7.85 (d, J = 8.6 Hz, 1H), 7.81 (dd, J = 8.1, 1.4 Hz, 1H), 7.77 (dd, J = 2.5, 1.7 Hz, 1H), 7.74 – 7.69 (m, 2H), 7.54 – 7.50 (m, 1H), 7.45 – 7.41 (m, 1H), 7.02 (ddd, J = 8.2, 2.6, 0.9 Hz, 1H), 3.92 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.23, 157.23, 148.30, 141.25, 136.86, 129.91, 129.83, 129.83, 129.76, 127.55, 127.36, 126.42, 120.11, 119.21, 115.47, 112.81, 55.51.

2-(m-tolyl)quinoline (11i)



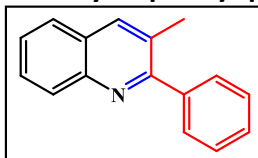
Colourless liquid, Yield: 93%, 102mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.21 – 8.17 (m, 2H), 8.00 (ddd, J = 2.4, 1.8, 0.6 Hz, 1H), 7.92 (dddd, J = 7.8, 1.8, 1.1, 0.6 Hz, 1H), 7.86 (d, J = 8.6 Hz, 1H), 7.81 (dd, J = 8.1, 1.5 Hz, 1H), 7.72 (ddd, J = 8.4, 6.9, 1.5 Hz, 1H), 7.52 (ddd, J = 8.1, 6.9, 1.2 Hz, 1H), 7.41 (t, J = 7.6 Hz, 1H), 7.30 – 7.26 (m, 1H), 2.48 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.70, 148.35, 139.74, 138.62, 136.82, 130.21, 129.77, 129.73, 128.83, 128.37, 127.55, 127.26, 126.32, 124.81, 119.28, 21.69.

2-(2-methoxyphenyl)quinoline (11j)



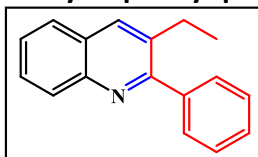
Colourless liquid, Yield: 91%, 107 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.17 (ddd, $J = 8.5, 1.8, 0.8$ Hz, 1H), 8.13 (d, $J = 8.6$ Hz, 1H), 7.87 (d, $J = 8.5$ Hz, 1H), 7.83 (td, $J = 7.6, 1.6$ Hz, 2H), 7.70 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.52 (ddd, $J = 8.1, 6.9, 1.2$ Hz, 1H), 7.42 (ddd, $J = 8.3, 7.4, 1.8$ Hz, 1H), 7.12 (td, $J = 7.5, 1.1$ Hz, 1H), 7.03 (dd, $J = 8.3, 0.9$ Hz, 1H), 3.85 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.28, 157.23, 148.39, 135.19, 131.57, 130.42, 129.81, 129.69, 129.31, 127.48, 127.13, 126.27, 123.55, 121.36, 111.52, 55.74.

3-methyl-2-phenylquinoline (11k)



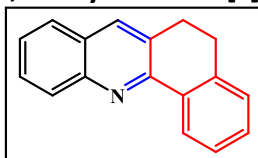
White solid, Yield: 92%, 101 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.12 (ddd, $J = 8.5, 1.9, 0.8$ Hz, 1H), 8.01 – 8.00 (m, 1H), 7.77 (dd, $J = 8.1, 1.4$ Hz, 1H), 7.65 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.60 – 7.57 (m, 2H), 7.52 – 7.46 (m, 3H), 7.45 – 7.41 (m, 1H), 2.45 (d, $J = 1.0$ Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.63, 146.70, 140.95, 136.83, 129.36, 129.31, 128.93, 128.84, 128.39, 128.27, 127.69, 126.79, 126.50, 20.70.

3-ethyl-2-phenylquinoline (11l)



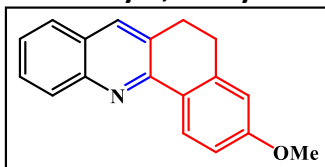
White solid, Yield: 94%, 110 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.14 (d, $J = 8.7$ Hz, 1H), 8.04 (s, 1H), 7.80 (dd, $J = 8.1, 1.3$ Hz, 1H), 7.66 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.56 – 7.46 (m, 5H), 7.45 – 7.41 (m, 1H), 2.79 (qd, $J = 7.5, 0.8$ Hz, 2H), 1.19 (t, $J = 7.5$ Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 160.74, 146.45, 141.02, 135.39, 135.02, 129.37, 128.89, 128.82, 128.39, 128.17, 127.84, 127.03, 126.47, 26.11, 14.82.

5,6-dihydrobenzo[c]acridine (11m)



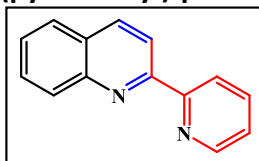
Yellow liquid, Yield: 90%, 104 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.61 – 8.57 (m, 1H), 8.15 (ddt, $J = 8.5, 1.2, 0.8$ Hz, 1H), 7.89 (d, $J = 0.9$ Hz, 1H), 7.73 (dd, $J = 8.1, 1.4$ Hz, 1H), 7.65 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.49 – 7.42 (m, 2H), 7.37 (td, $J = 7.4, 1.5$ Hz, 1H), 7.29 – 7.26 (m, 1H), 3.11 (ddd, $J = 5.8, 3.4, 1.0$ Hz, 2H), 3.00 (dd, $J = 8.4, 5.5$ Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 153.50, 147.73, 139.53, 134.81, 133.81, 130.70, 129.79, 129.49, 128.76, 128.06, 127.97, 127.43, 127.04, 126.16, 28.92, 28.50.

3-methoxy-5,6-dihydrobenzo[c]acridine (11n)



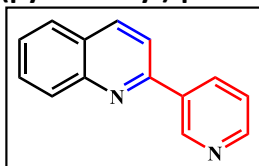
Yellow liquid, Yield: 93%, 122 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.51 (d, $J = 8.6$ Hz, 1H), 8.09 (ddt, $J = 8.5, 1.2, 0.7$ Hz, 1H), 7.85 (d, $J = 0.8$ Hz, 1H), 7.70 (dd, $J = 8.1, 1.4$ Hz, 1H), 7.62 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.43 (ddd, $J = 8.1, 6.9, 1.2$ Hz, 1H), 6.96 (dd, $J = 8.6, 2.6$ Hz, 1H), 6.78 (d, $J = 2.6$ Hz, 1H), 3.86 (s, 3H), 3.10 – 3.06 (m, 2H), 2.96 (dd, $J = 8.4, 5.6$ Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz) δ 161.00, 153.51, 147.73, 141.37, 133.60, 130.12, 129.19, 128.66, 127.88, 127.85, 127.64, 127.01, 125.71, 55.43, 28.97, 28.84.

2-(pyridin-2-yl)quinoline (11o)



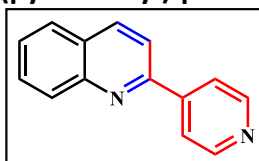
Colourless solid, Yield: 90%, 93 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.76 (ddd, $J = 4.8, 1.7, 0.9$ Hz, 1H), 8.68 (dt, $J = 8.0, 1.0$ Hz, 1H), 8.59 (d, $J = 8.6$ Hz, 1H), 8.29 (d, $J = 8.6$ Hz, 1H), 8.21 (d, $J = 8.6$ Hz, 1H), 7.91 – 7.83 (m, 2H), 7.75 (ddd, $J = 8.4, 6.9, 1.4$ Hz, 1H), 7.56 (ddd, $J = 8.0, 6.9, 1.1$ Hz, 1H), 7.36 (ddd, $J = 7.4, 4.8, 1.2$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 156.36, 156.18, 149.19, 147.95, 136.95, 136.82, 129.84, 129.57, 128.27, 127.64, 126.77, 124.04, 121.85, 118.98.

2-(pyridin-3-yl)quinoline (11p)



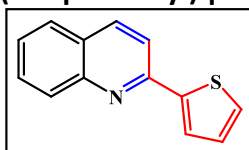
Colourless liquid, Yield: 92%, 95 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 9.34 (dd, $J = 2.4, 1.0$ Hz, 1H), 8.69 (dd, $J = 4.9, 1.8$ Hz, 1H), 8.50 (ddd, $J = 8.0, 2.4, 1.7$ Hz, 1H), 8.23 (dd, $J = 8.8, 1.1$ Hz, 1H), 8.16 (ddd, $J = 8.5, 2.1, 0.8$ Hz, 1H), 7.87 – 7.80 (m, 2H), 7.73 (ddd, $J = 8.6, 6.9, 1.6$ Hz, 1H), 7.54 (ddd, $J = 8.2, 6.9, 1.3$ Hz, 1H), 7.44 (ddd, $J = 8.0, 4.8, 1.0$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz) δ 154.57, 150.00, 148.64, 148.42, 137.31, 135.27, 132.13, 130.12, 129.80, 127.65, 127.46, 126.92, 123.87, 118.60.

2-(pyridin-4-yl)quinoline (11q)



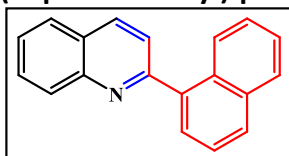
Colourless liquid, Yield: 93%, 96 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.75 (dd, $J = 4.5, 1.6$ Hz, 2H), 8.25 (d, $J = 8.6$ Hz, 1H), 8.17 (d, $J = 8.5$ Hz, 1H), 8.04 (dd, $J = 4.5, 1.7$ Hz, 2H), 7.85 (dd, $J = 15.5, 8.3$ Hz, 2H), 7.74 (ddd, $J = 7.9, 6.7, 1.1$ Hz, 1H), 7.56 (ddd, $J = 8.0, 7.0, 1.0$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 154.46, 150.46, 148.33, 146.78, 137.39, 130.22, 130.03, 127.90, 127.64, 127.32, 121.74, 118.51.

2-(thiophen-2-yl)quinoline (11r)



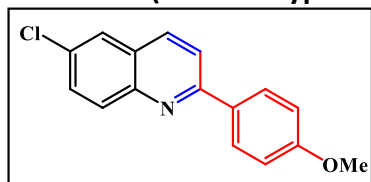
Brown liquid, Yield: 91%, 96 mg. $^1\text{H NMR}$ (500 MHz, CDCl_3) δ 8.13 – 8.10 (m, 1H), 8.08 (ddd, $J = 8.5, 1.8, 0.8$ Hz, 1H), 7.78 (d, $J = 8.6$ Hz, 1H), 7.76 (dd, $J = 8.1, 1.5$ Hz, 1H), 7.72 (dd, $J = 3.7, 1.1$ Hz, 1H), 7.68 (ddd, $J = 8.4, 6.9, 1.5$ Hz, 1H), 7.50 – 7.44 (m, 2H), 7.15 (dd, $J = 5.0, 3.7$ Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 152.42, 148.19, 145.46, 136.72, 129.91, 129.34, 128.67, 128.17, 127.57, 127.27, 126.19, 125.94, 117.75.

2-(naphthalen-1-yl)quinoline (11s)



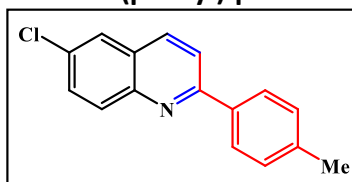
Colourless liquid, Yield: 93%, 119 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.27 (d, J = 8.4 Hz, 1H), 8.24 (dd, J = 8.5, 0.7 Hz, 1H), 8.14 (d, J = 8.3 Hz, 1H), 7.95 (t, J = 8.6 Hz, 2H), 7.90 (dd, J = 8.1, 1.2 Hz, 1H), 7.78 (ddd, J = 8.4, 6.9, 1.4 Hz, 1H), 7.73 (d, J = 7.1 Hz, 1H), 7.70 (d, J = 8.4 Hz, 1H), 7.62 – 7.58 (m, 2H), 7.54 – 7.45 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.53, 148.22, 138.82, 136.38, 134.11, 131.36, 129.89, 129.81, 129.23, 128.52, 127.86, 127.70, 127.11, 126.70, 126.08, 125.80, 125.50, 123.38.

6-chloro-2-(4-methoxyphenyl)quinoline (11t)



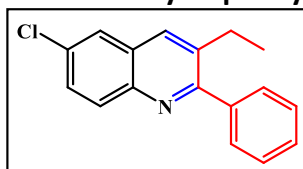
White solid, Yield: 91%, 123 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.12 – 8.08 (m, 2H), 8.04 (dd, J = 8.7, 3.5 Hz, 2H), 7.80 (d, J = 8.7 Hz, 1H), 7.74 (d, J = 2.3 Hz, 1H), 7.61 (dd, J = 9.0, 2.4 Hz, 1H), 7.05 – 7.01 (m, 2H), 3.87 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 161.07, 157.16, 146.72, 135.77, 131.81, 131.51, 131.17, 130.53, 128.94, 127.50, 126.20, 119.40, 114.35, 55.49.

6-chloro-2-(p-tolyl)quinoline (11u)



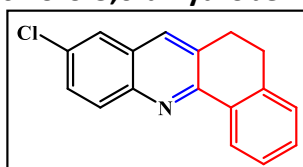
White solid, Yield: 88%, 112 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.08 (dd, J = 8.7, 4.4 Hz, 2H), 8.04 (d, J = 8.2 Hz, 2H), 7.86 (d, J = 8.7 Hz, 1H), 7.77 (d, J = 2.3 Hz, 1H), 7.63 (dd, J = 9.0, 2.4 Hz, 1H), 7.32 (d, J = 7.9 Hz, 2H), 2.43 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.61, 146.74, 139.82, 136.47, 135.82, 131.75, 131.32, 130.56, 129.73, 127.72, 127.46, 126.20, 119.73, 21.45.

6-chloro-3-ethyl-2-phenylquinoline (11v)



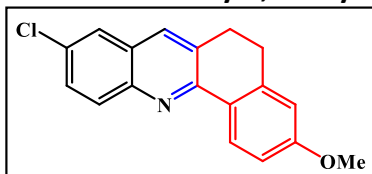
Colourless liquid, Yield: 87%, 117 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.07 – 8.02 (m, 1H), 7.95 (d, J = 0.4 Hz, 1H), 7.78 (d, J = 2.3 Hz, 1H), 7.58 (dd, J = 8.9, 2.3 Hz, 1H), 7.54 – 7.51 (m, 2H), 7.49 – 7.42 (m, 3H), 2.78 (qd, J = 7.5, 0.8 Hz, 2H), 1.18 (t, J = 7.5 Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 161.00, 144.77, 140.58, 136.47, 134.06, 132.09, 130.98, 129.80, 128.74, 128.46, 128.37, 125.68, 26.11, 14.68.

9-chloro-5,6-dihydrobenzo[c]acridine (11w)



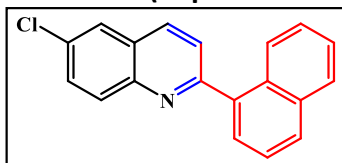
Colourless liquid, Yield: 88%, 117 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.53 (dd, J = 7.6, 1.5 Hz, 1H), 8.04 (d, J = 8.9 Hz, 1H), 7.79 (s, 1H), 7.69 (d, J = 2.3 Hz, 1H), 7.56 (dd, J = 8.9, 2.4 Hz, 1H), 7.42 (td, J = 7.5, 1.4 Hz, 1H), 7.37 (td, J = 7.4, 1.5 Hz, 1H), 7.26 (dd, J = 7.4, 0.8 Hz, 1H), 3.12 – 3.08 (m, 2H), 3.01 – 2.97 (m, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 153.77, 146.05, 139.47, 134.44, 132.81, 131.69, 131.06, 130.02, 129.60, 128.49, 128.10, 127.48, 126.11, 125.69, 28.89, 28.30.

9-chloro-3-methoxy-5,6-dihydrobenzo[c]acridine (11x)



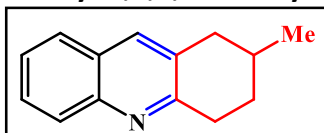
Colourless liquid, Yield: 90%, 133 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.45 (d, J = 8.6 Hz, 1H), 7.98 (d, J = 8.9 Hz, 1H), 7.70 (s, 1H), 7.63 (d, J = 2.4 Hz, 1H), 7.53 (dd, J = 8.9, 2.4 Hz, 1H), 6.93 (dd, J = 8.7, 2.6 Hz, 1H), 6.76 (d, J = 2.6 Hz, 1H), 3.85 (s, 3H), 3.06 – 3.02 (m, 2H), 2.93 (dd, J = 8.3, 5.6 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 161.16, 153.76, 146.05, 141.34, 132.59, 131.14, 131.09, 130.74, 129.45, 128.15, 127.87, 127.45, 125.68, 113.21, 113.02, 55.43, 28.91, 28.63.

6-chloro-2-(naphthalen-1-yl)quinoline (11y)



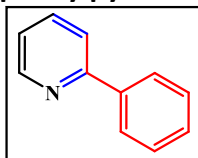
White solid, Yield: 86%, 125 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.16 (d, J = 8.6 Hz, 2H), 8.12 (dd, J = 8.5, 0.9 Hz, 1H), 7.97 – 7.92 (m, 2H), 7.87 (d, J = 2.5 Hz, 1H), 7.73 – 7.68 (m, 3H), 7.59 (dd, J = 8.2, 7.1 Hz, 1H), 7.52 (ddd, J = 8.2, 6.8, 1.4 Hz, 1H), 7.48 (ddd, J = 8.2, 6.8, 1.5 Hz, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.79, 146.59, 138.36, 135.45, 134.11, 132.37, 131.43, 131.21, 130.80, 129.47, 128.59, 127.95, 127.65, 126.82, 126.35, 126.17, 125.61, 125.49, 124.21.

2-methyl-1,2,3,4-tetrahydroacridine (11z)



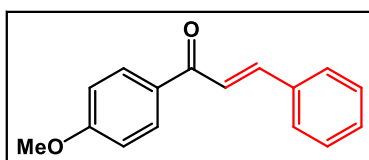
Colourless liquid, Yield: 76%, 75 mg. ^1H NMR (500 MHz, CDCl_3) δ 7.96 (d, J = 8.5 Hz, 1H), 7.76 (s, 1H), 7.68 (dd, J = 8.1, 0.7 Hz, 1H), 7.58 (ddd, J = 8.4, 6.9, 1.4 Hz, 1H), 7.44 – 7.39 (m, 1H), 3.21 (ddd, J = 17.9, 5.7, 3.4 Hz, 1H), 3.08 (ddd, J = 17.8, 11.4, 6.2 Hz, 1H), 3.00 (ddd, J = 16.4, 4.6, 1.0 Hz, 1H), 2.58 (dd, J = 16.3, 10.8 Hz, 1H), 2.08 – 2.03 (m, 1H), 2.00 – 1.93 (m, 1H), 1.59 (dtd, J = 13.0, 11.2, 5.7 Hz, 1H), 1.11 (d, J = 6.6 Hz, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 159.11, 146.72, 135.05, 130.69, 128.59, 128.32, 127.22, 126.97, 125.61, 77.37, 77.12, 76.86, 37.88, 33.17, 31.49, 29.18, 21.76.

2-phenylpyridine (11aa)



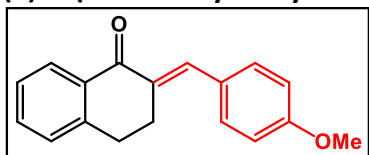
Colourless liquid, Yield: 54%, 42 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.69 (dt, J = 4.8, 1.4 Hz, 1H), 8.01 – 7.98 (m, 2H), 7.71 – 7.68 (m, 2H), 7.49 – 7.45 (m, 2H), 7.43 – 7.38 (m, 1H), 7.21 – 7.16 (m, 1H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 157.52, 149.76, 139.49, 136.86, 129.07, 128.87, 127.02, 122.21, 120.67.

(E)-1-(4-methoxyphenyl)-3-phenylprop-2-en-1-one (3b)



White solid, Yield: 89%, 106 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.05 – 8.01 (m, 2H, 7.80 (d, J = 15.6 Hz, 1H), 7.62 – 7.56 (m, 3H), 7.53 – 7.48 (m, 2H), 7.43 (d, J = 15.6 Hz, 1H), 6.96 – 6.92 (m, 2H), 3.84 (s, 3H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 190.57, 161.69, 144.73, 138.48, 132.61, 130.28, 128.59, 128.44, 127.57, 119.70, 114.43, 55.41.

(E)-2-(4-methoxybenzylidene)-3,4-dihydronaphthalen-1(2H)-one (6b)



White solid, Yield: 92%, 122 mg. ^1H NMR (500 MHz, CDCl_3) δ 8.11 (dd, J = 7.8, 1.3 Hz, 1H), 7.84 (s, 1H), 7.45 (t, J = 7.4 Hz, 1H), 7.41 (d, J = 8.6 Hz, 2H), 7.33 (t, J = 7.6 Hz, 1H), 7.22 (d, J = 7.6 Hz, 1H), 6.93 (d, J = 8.6 Hz, 2H), 3.82 (s, 3H), 3.11 (t, J = 6.5 Hz, 2H), 2.91 (t, J = 6.4 Hz, 2H). $^{13}\text{C}\{^1\text{H}\}$ NMR (126 MHz, CDCl_3) δ 187.91 (s), 160.07, 143.17, 136.77, 133.72, 133.59, 133.22, 131.88, 128.44, 128.21, 127.06, 114.05, 55.42, 28.84, 27.31.

13. ^1H and $^{13}\text{C}\{^1\text{H}\}$ NMR spectra of characterized products

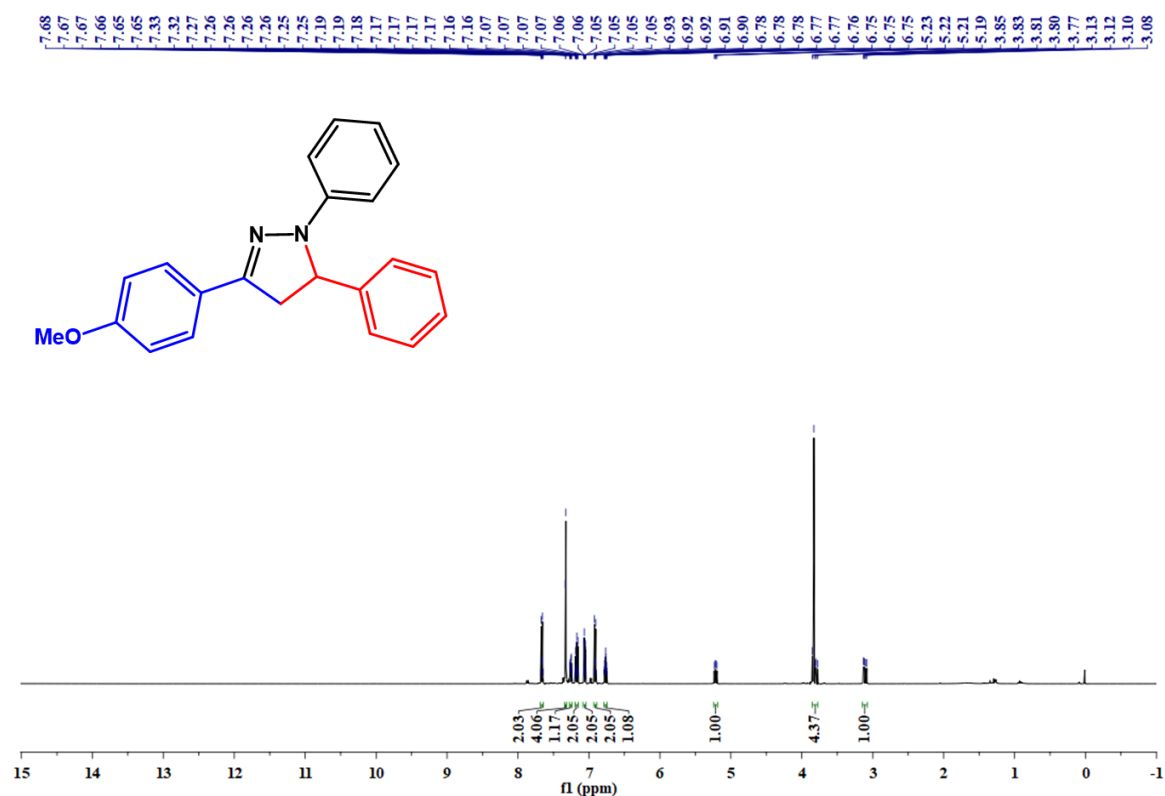


Figure S23: ^1H NMR spectrum of **4a** in CDCl_3 at 500 MHz.

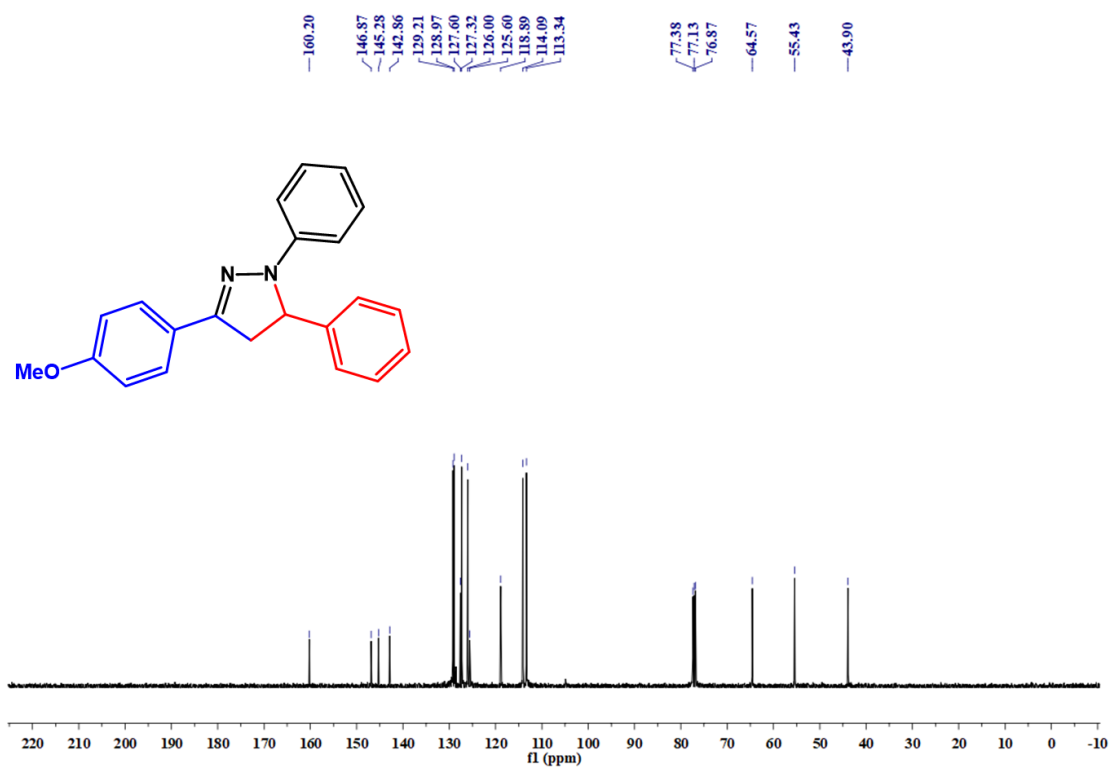


Figure S24: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4a** in CDCl_3 at 126 MHz.

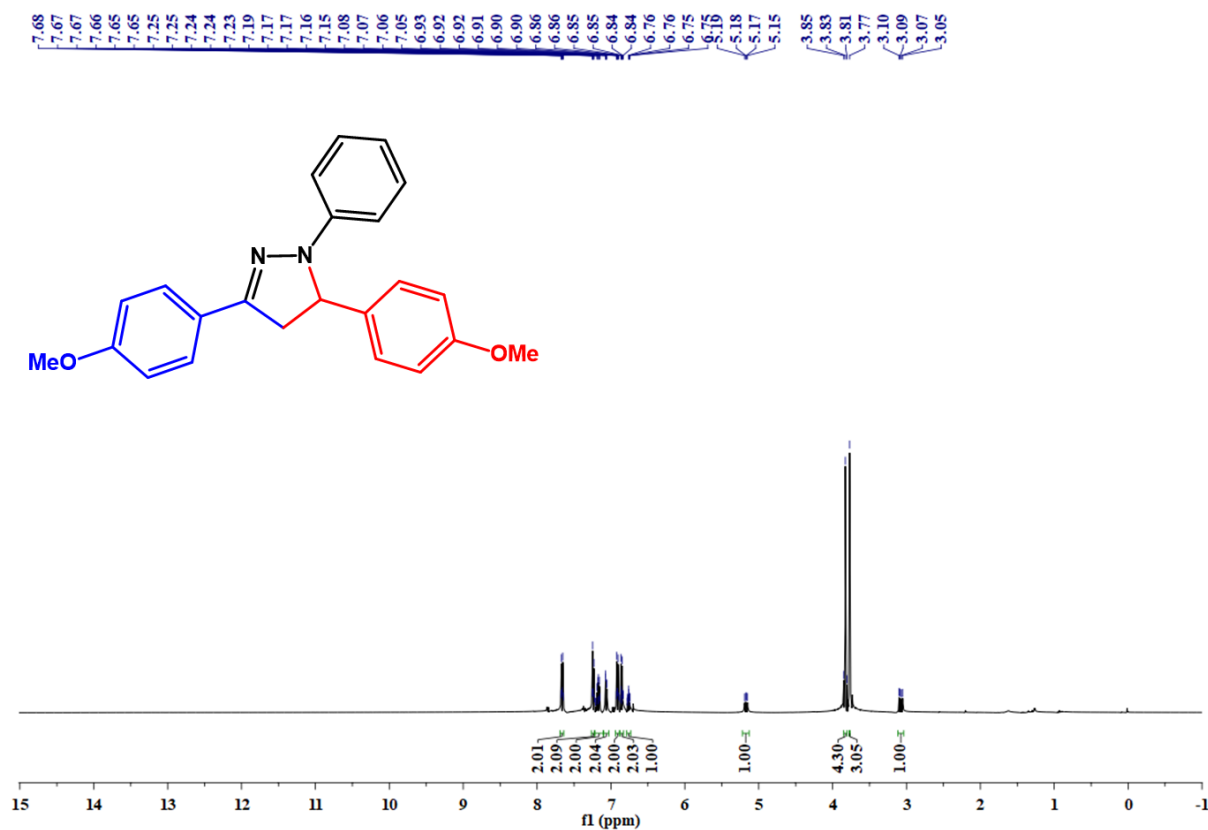


Figure S25: ^1H NMR spectrum of **4b** in CDCl_3 at 500 MHz.

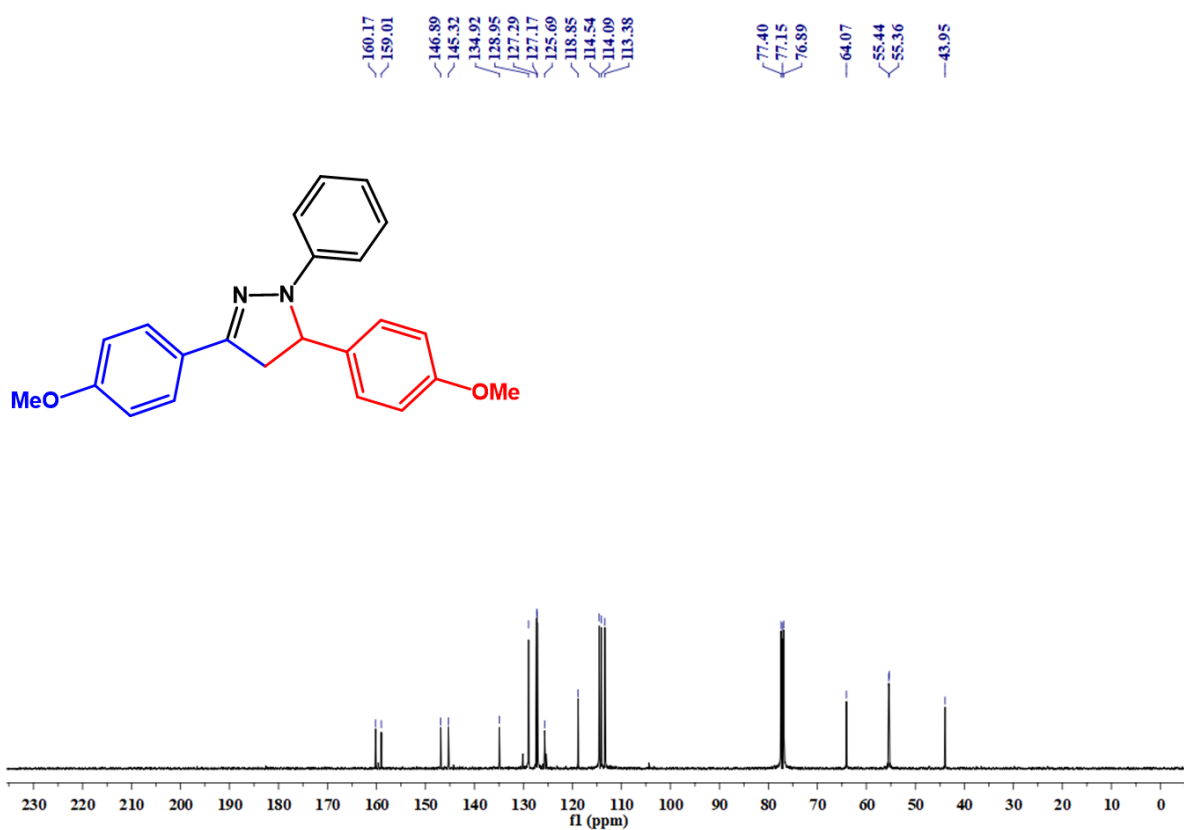


Figure S26: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4b** in CDCl_3 at 126 MHz.

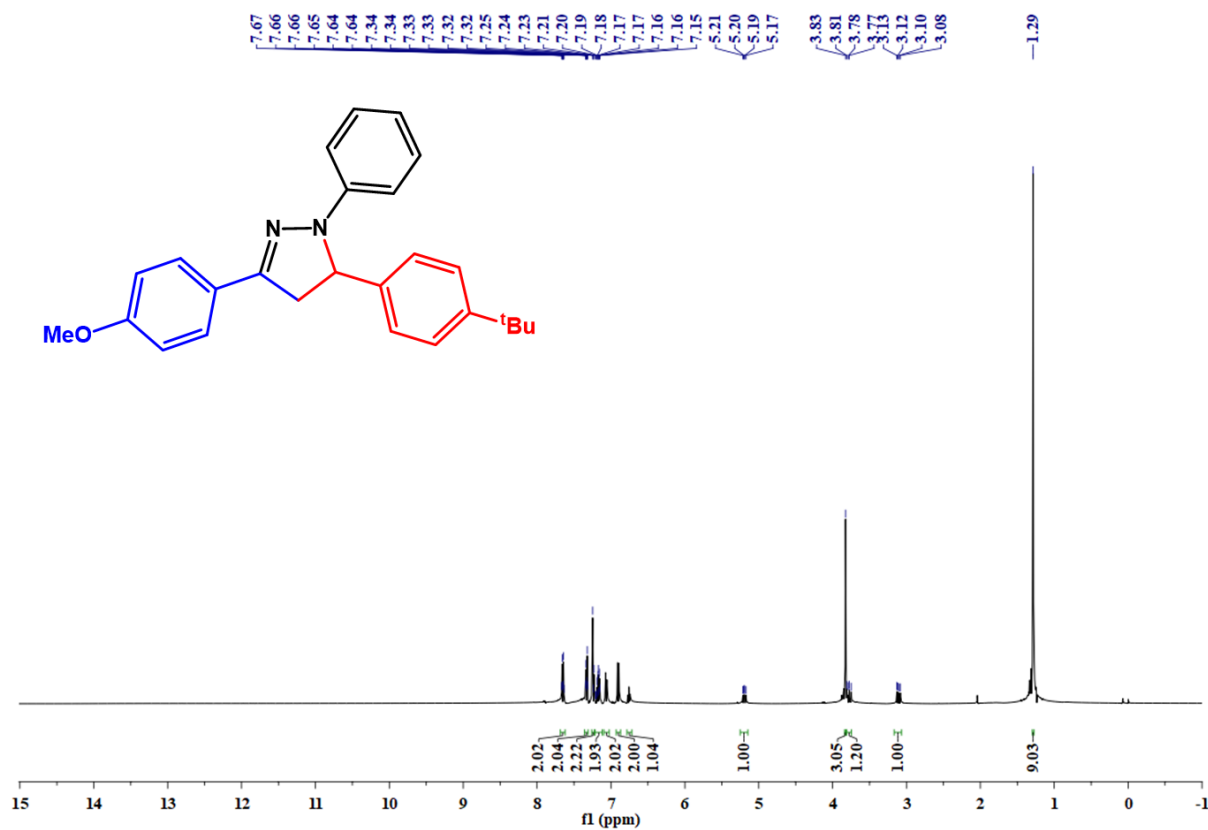


Figure S27: ^1H NMR spectrum of **4c** in CDCl_3 at 500 MHz.

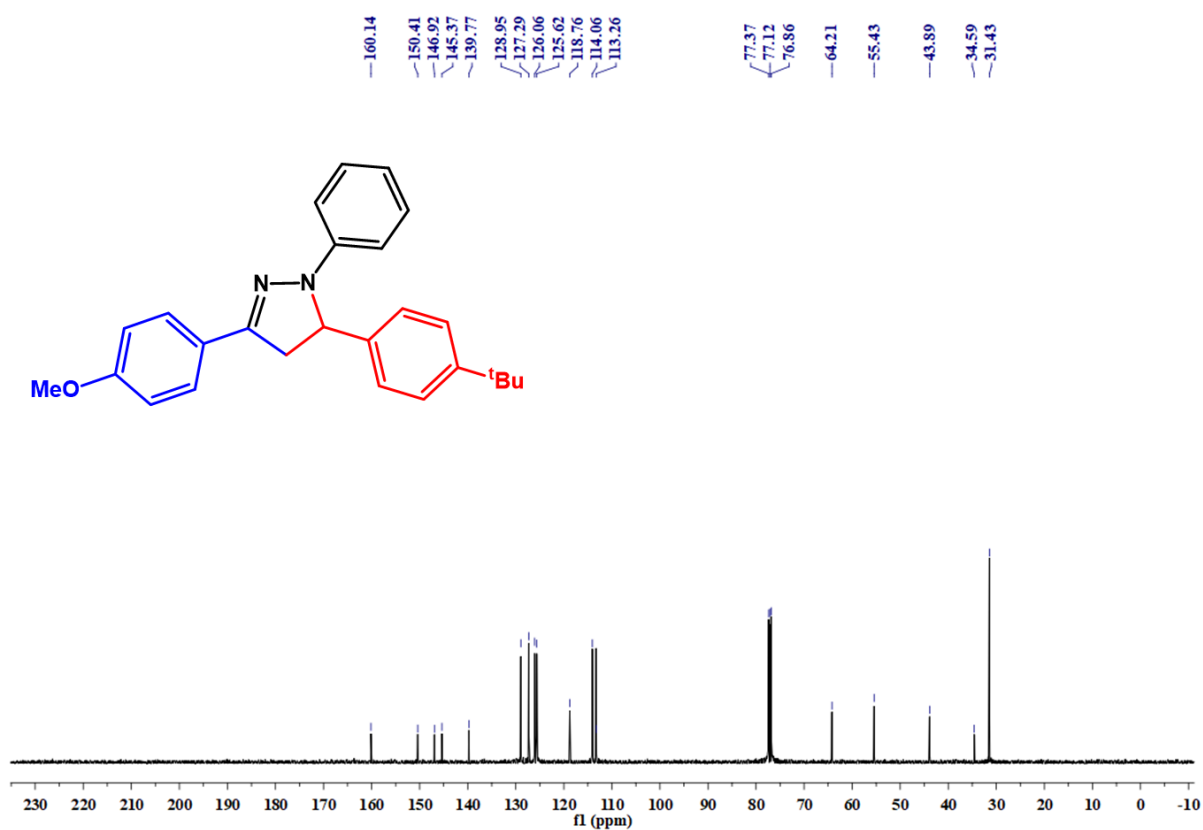


Figure S28: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4c** in CDCl_3 at 126 MHz.

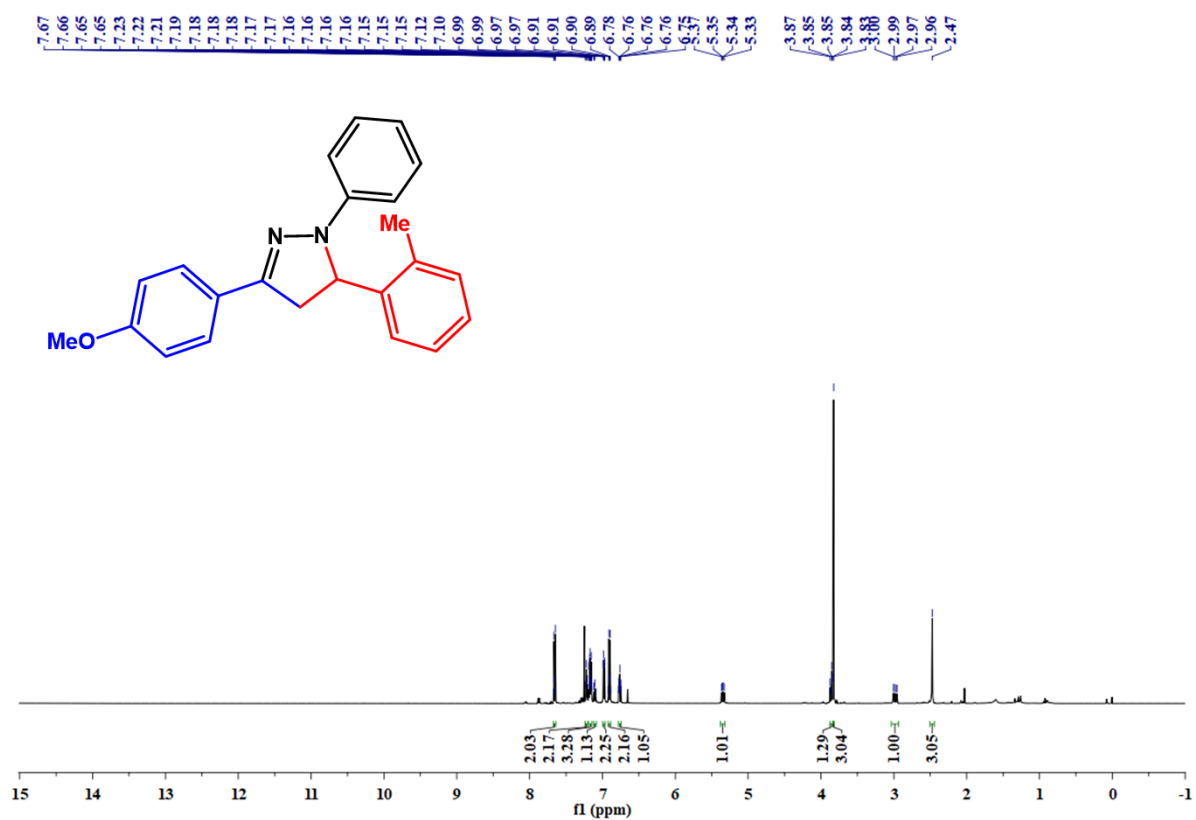


Figure S29: ^1H NMR spectrum of **4d** in CDCl_3 at 500 MHz.

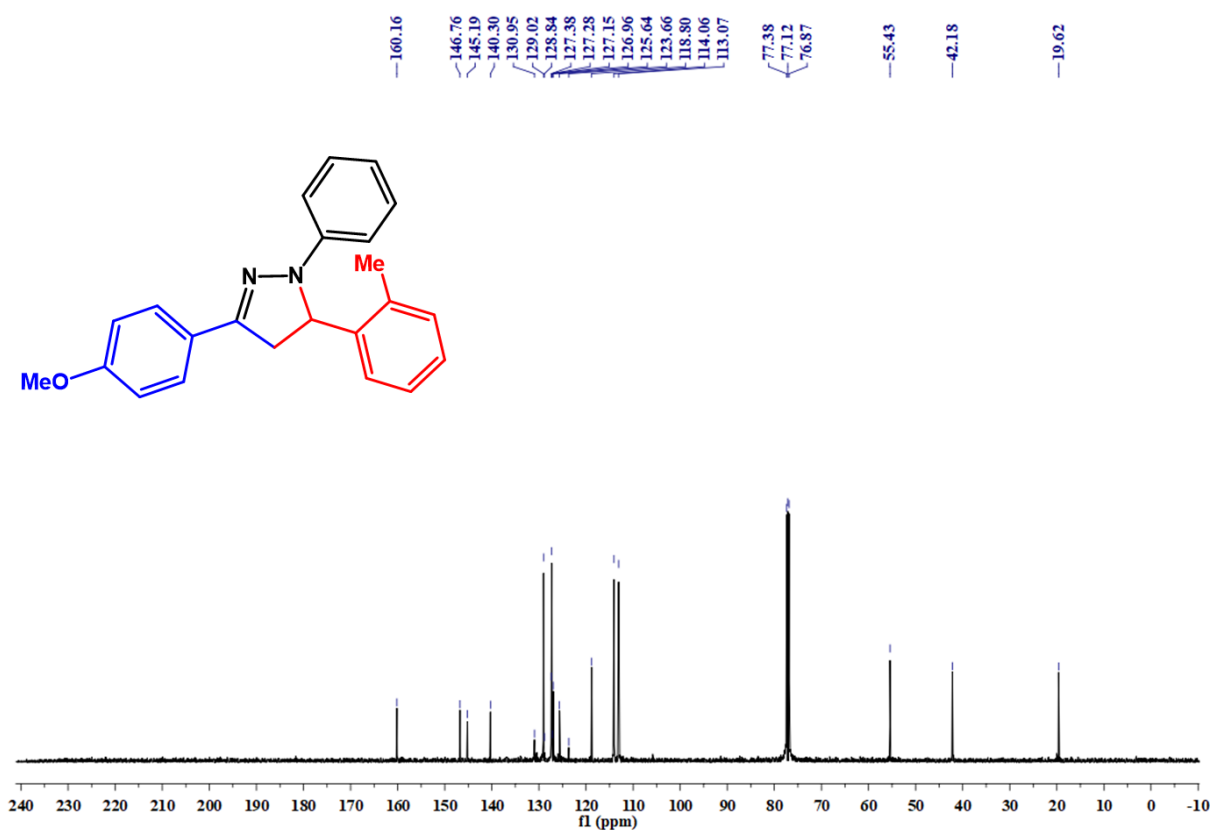


Figure S30: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4d** in CDCl_3 at 126 MHz.

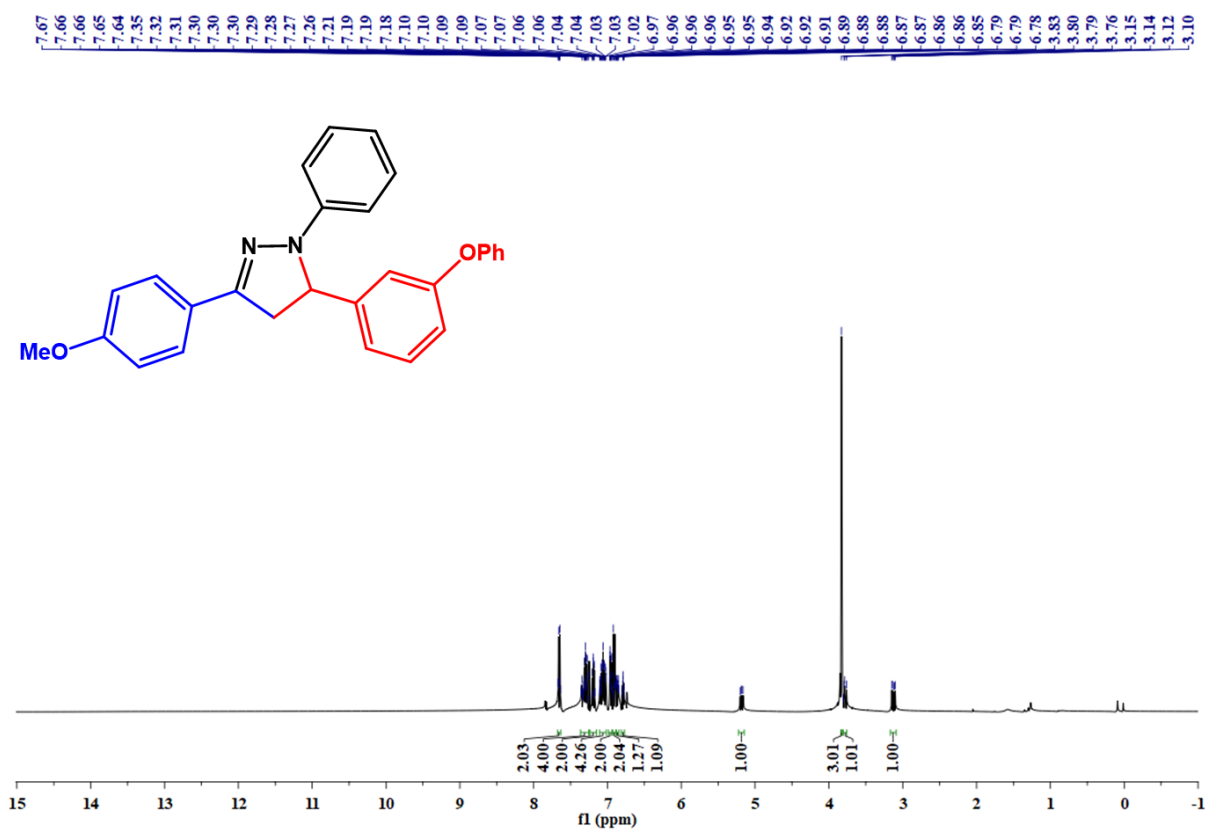


Figure S31: ^1H NMR spectrum of **4e** in CDCl_3 at 500 MHz.

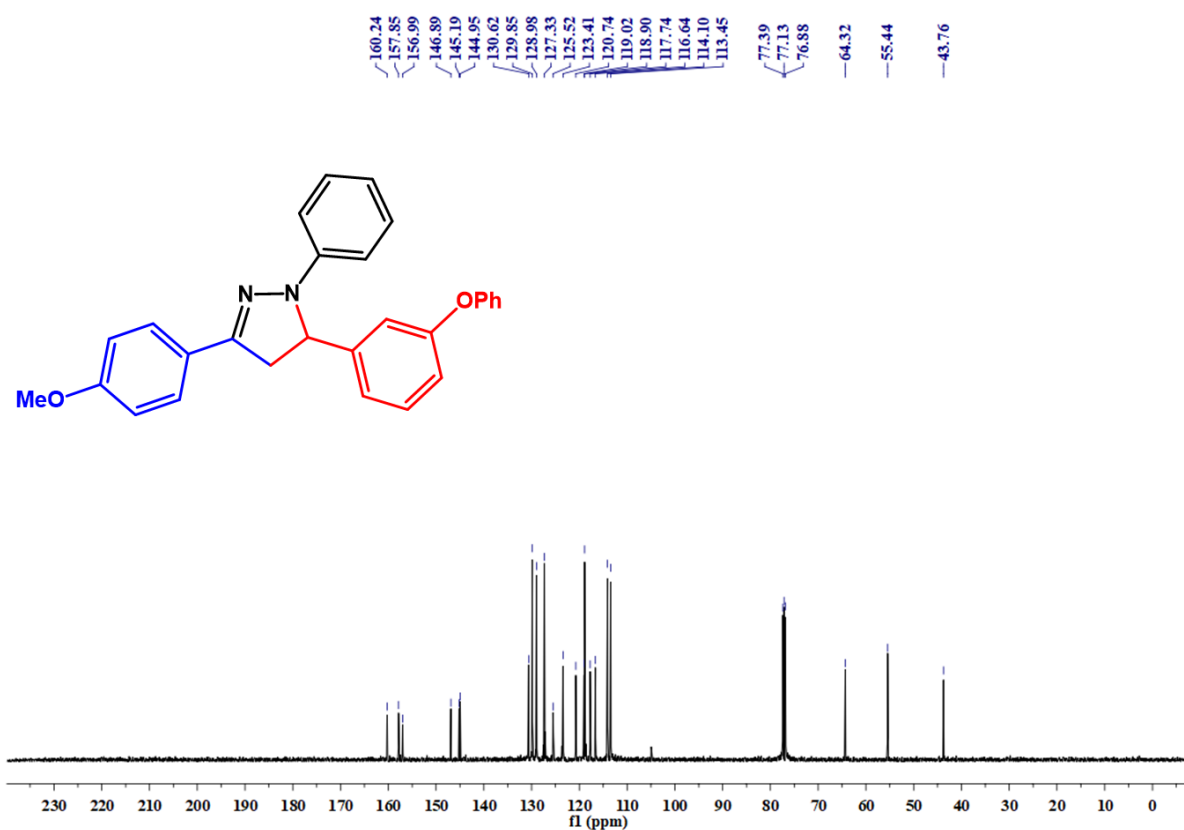


Figure S32: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4e** in CDCl_3 at 126 MHz.

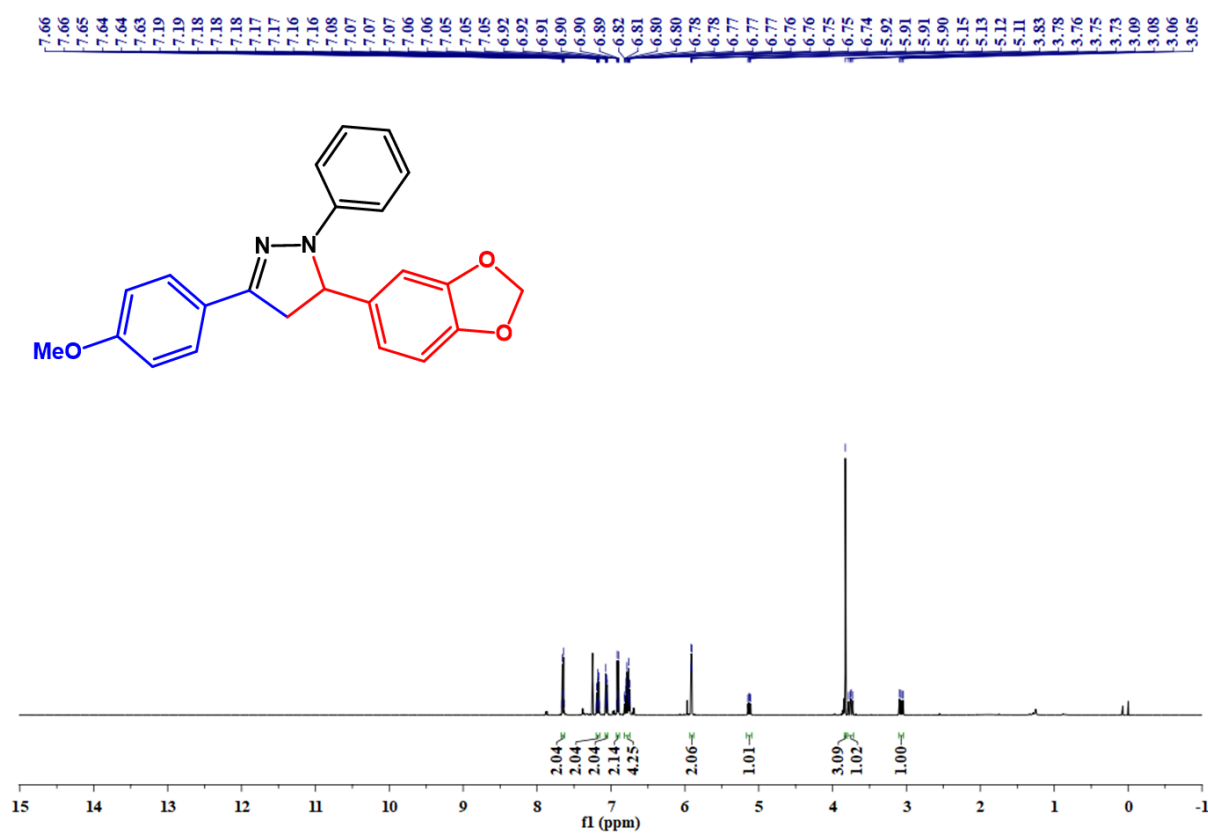


Figure S33: ^1H NMR spectrum of **4f** in CDCl_3 at 500 MHz.

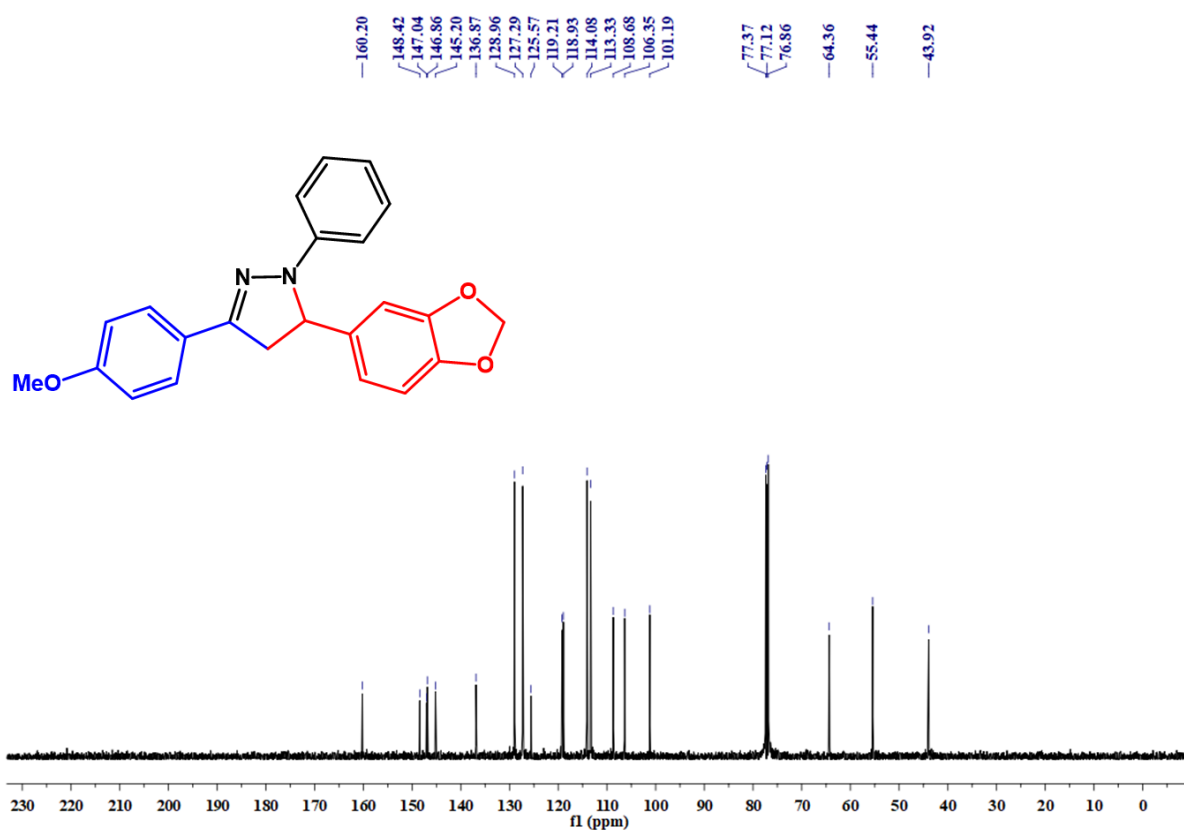


Figure S34: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4f** in CDCl_3 at 126 MHz.

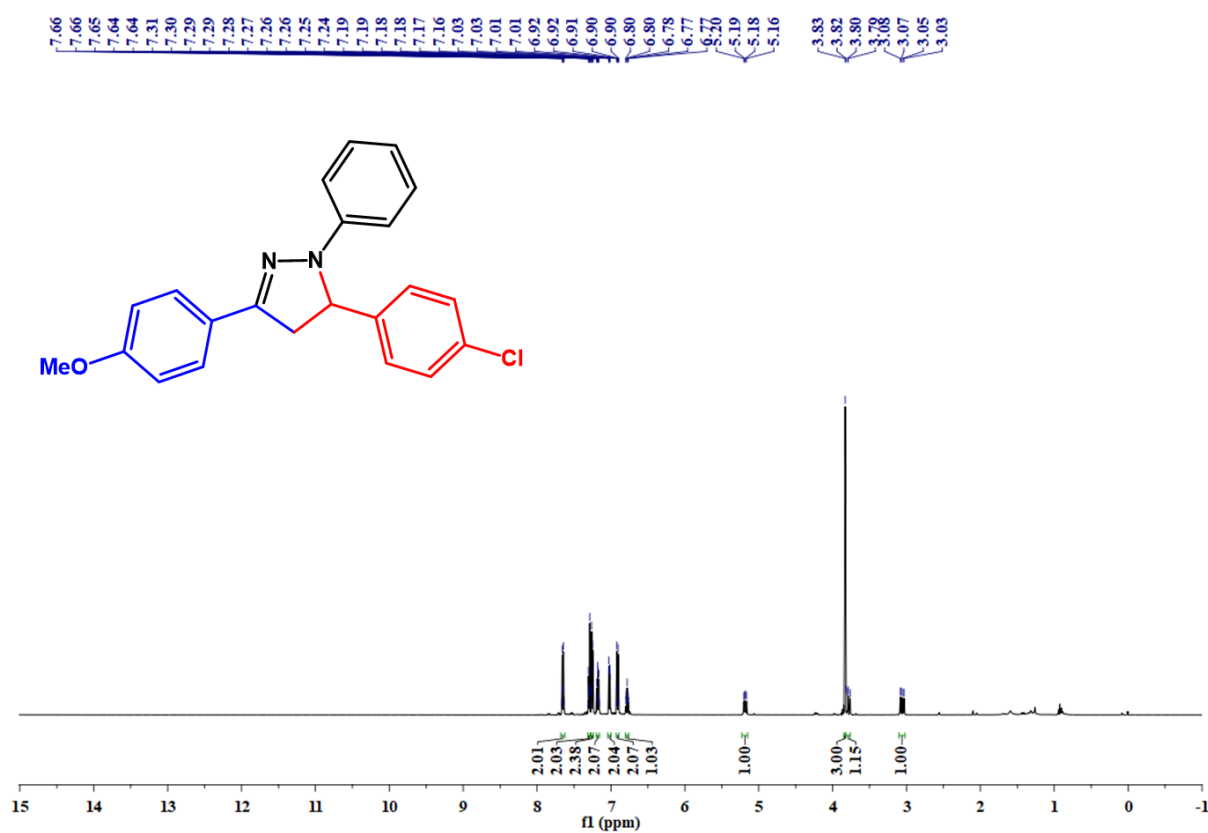


Figure S35: ^1H NMR spectrum of **4g** in CDCl_3 at 500 MHz.

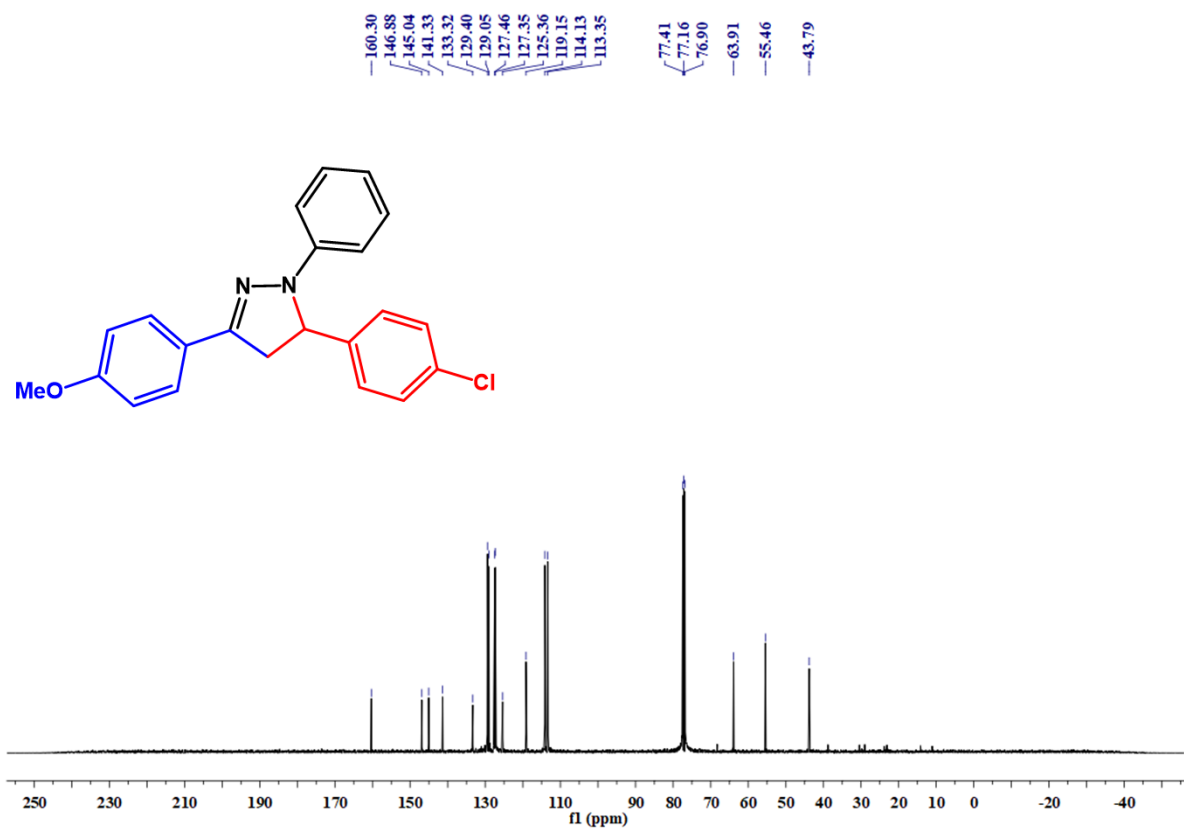


Figure S36: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4g** in CDCl_3 at 126 MHz.

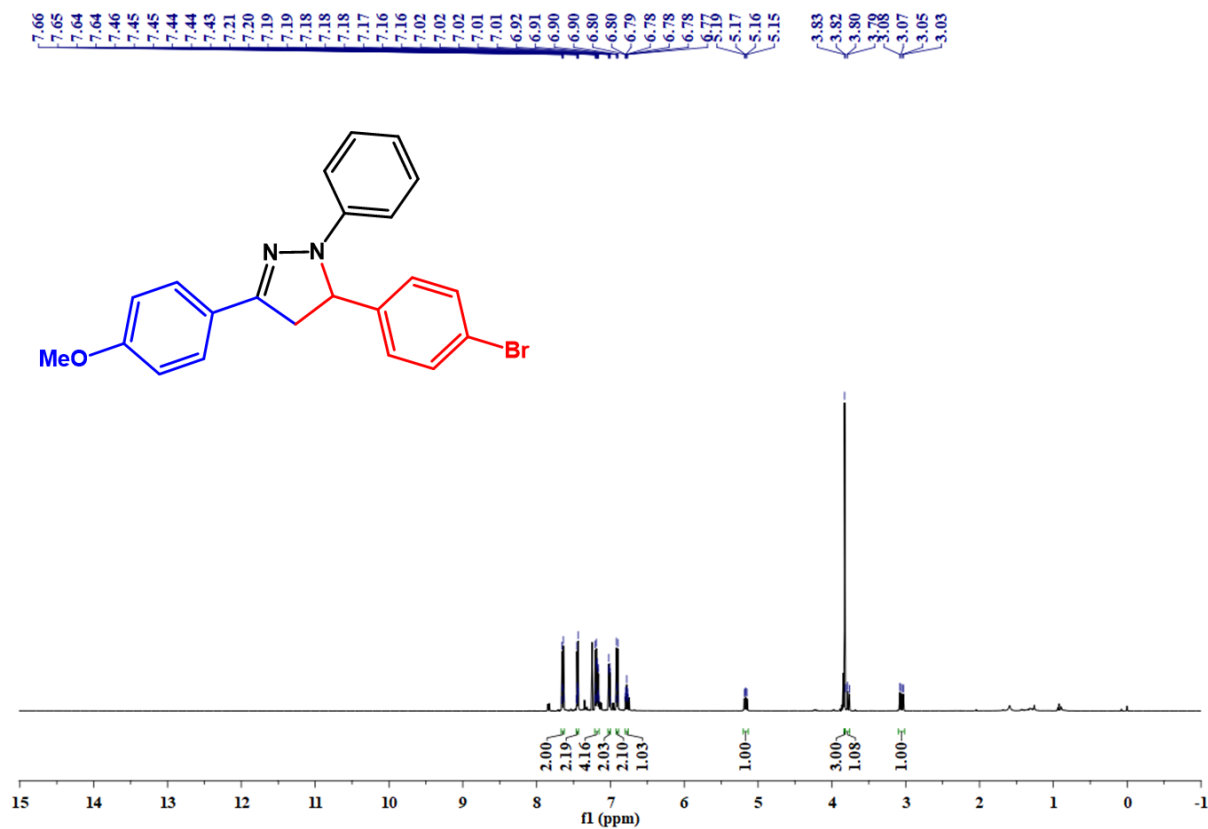


Figure S37: ^1H NMR spectrum of **4h** in CDCl_3 at 500 MHz.

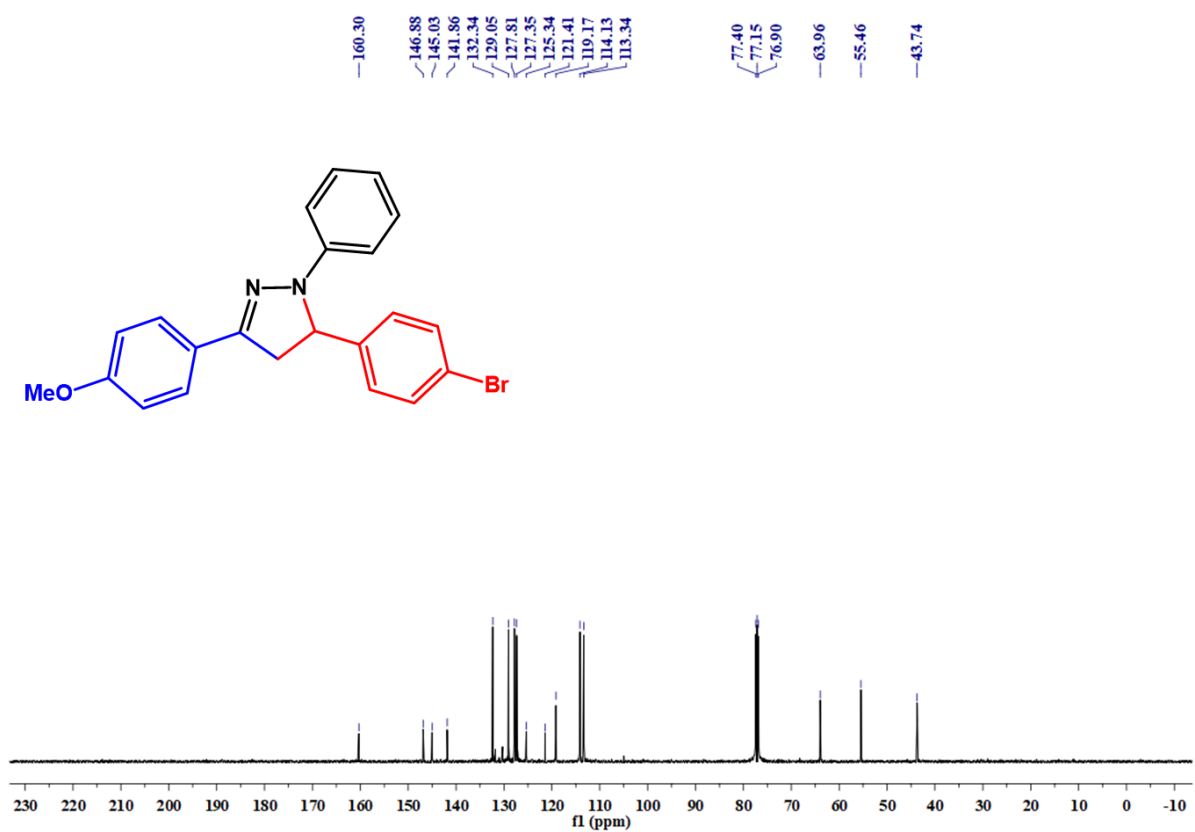


Figure S38: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4h** in CDCl_3 at 126 MHz.

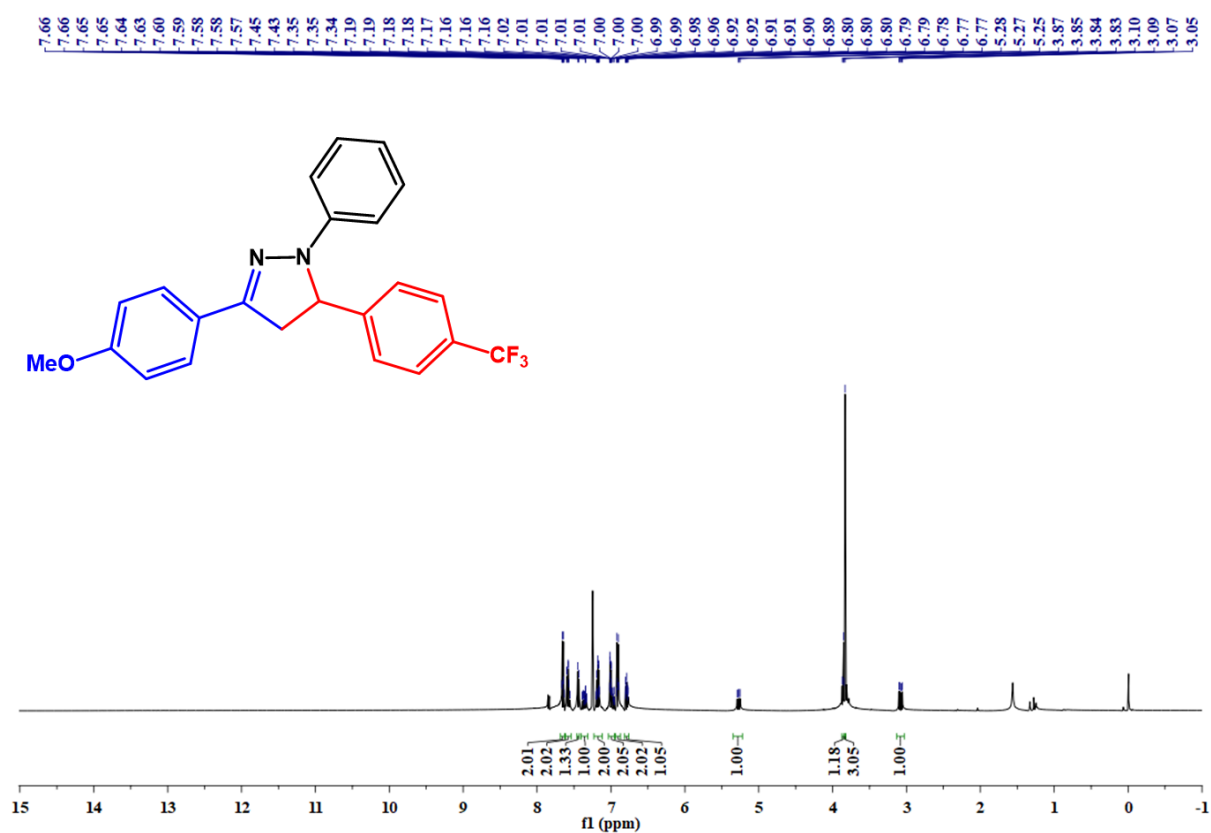


Figure S39: ^1H NMR spectrum of **4i** in CDCl_3 at 500 MHz.

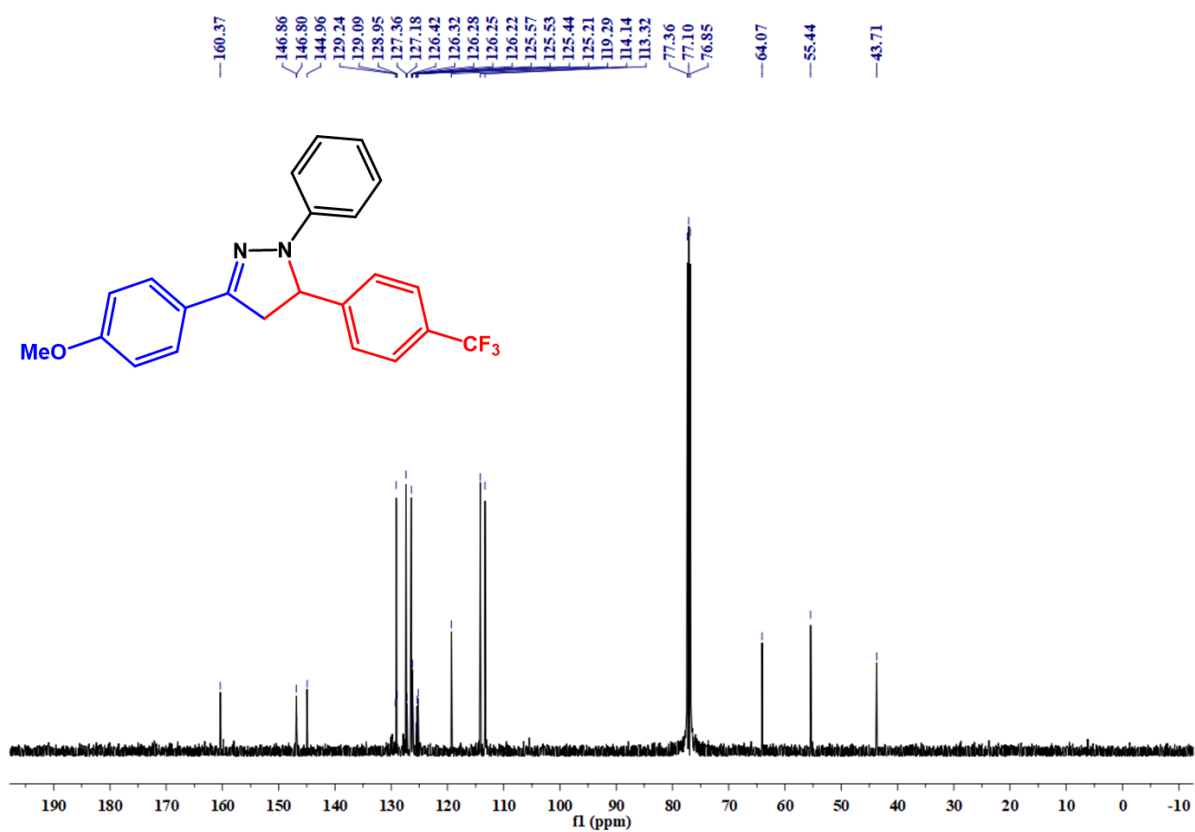


Figure S40: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4i** in CDCl_3 at 126 MHz.

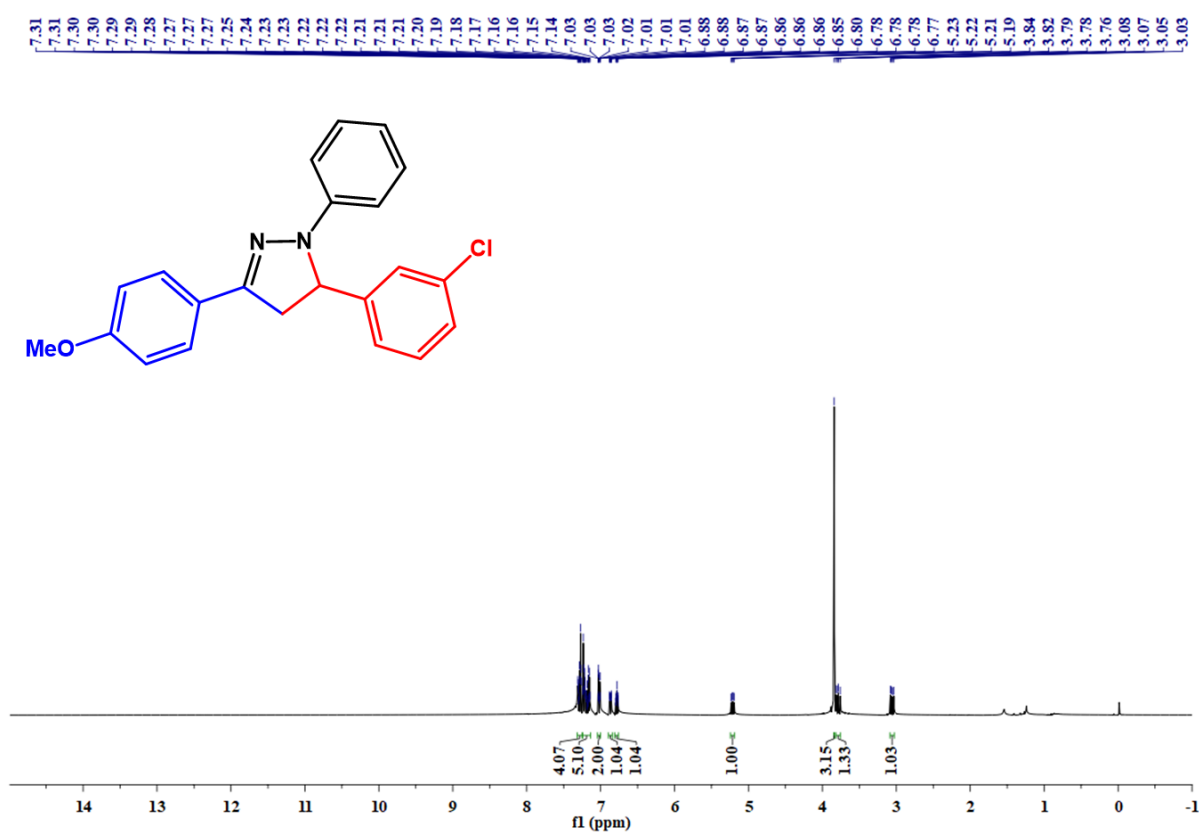


Figure S41: ^1H NMR spectrum of **4j** in CDCl_3 at 500 MHz.

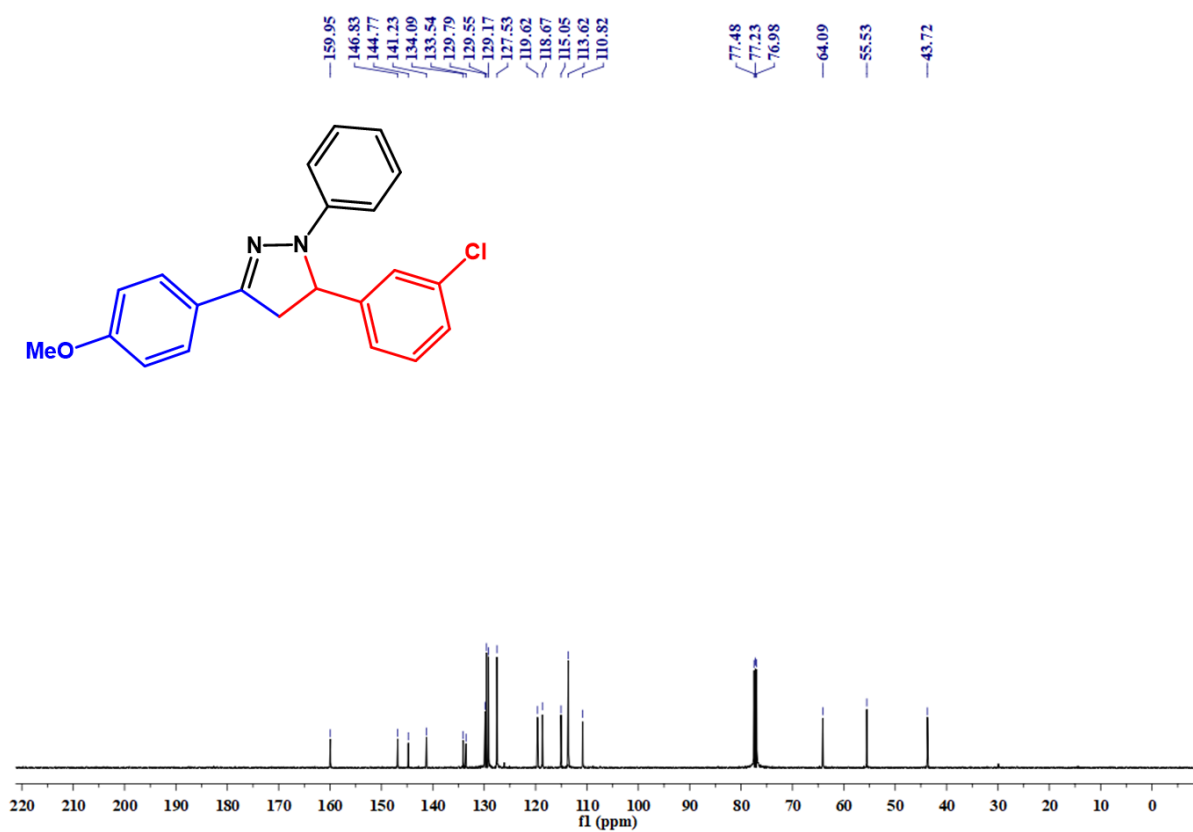


Figure S42: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4j** in CDCl_3 at 126 MHz.

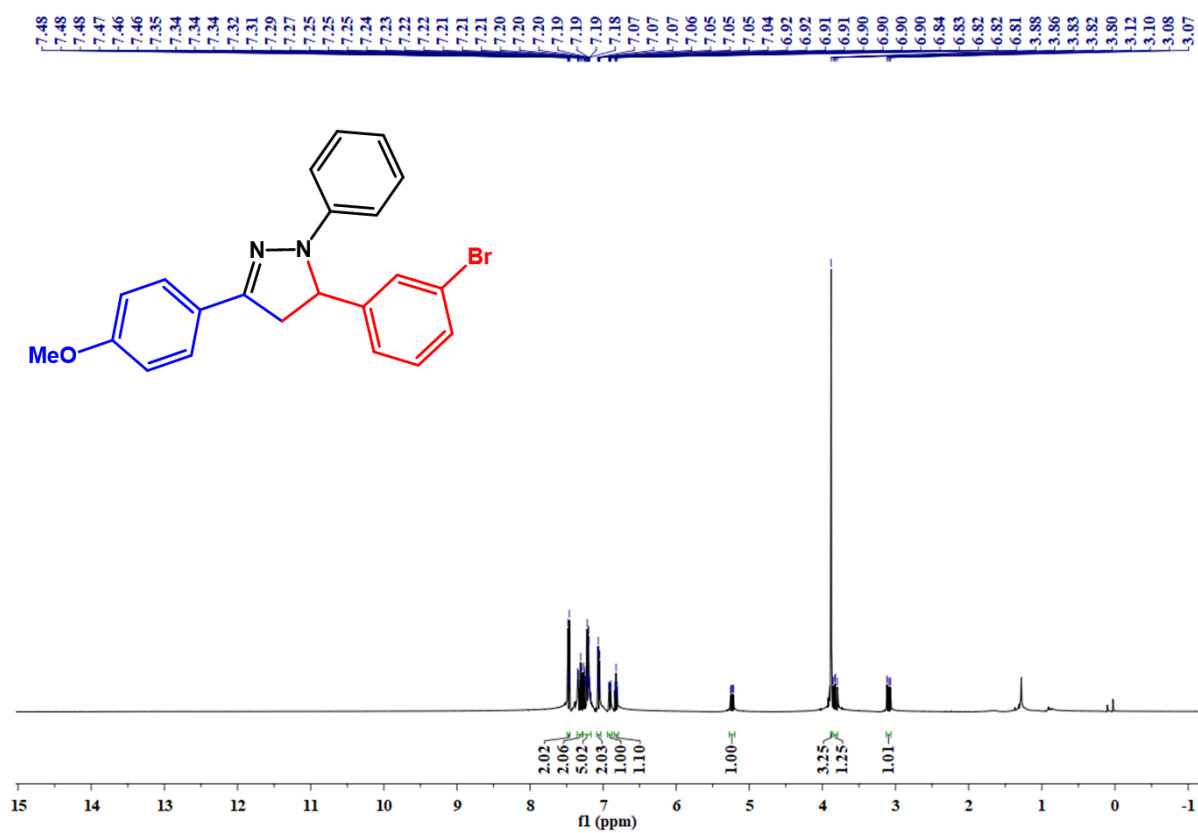


Figure S43: ^1H NMR spectrum of **4k** in CDCl_3 at 500 MHz.

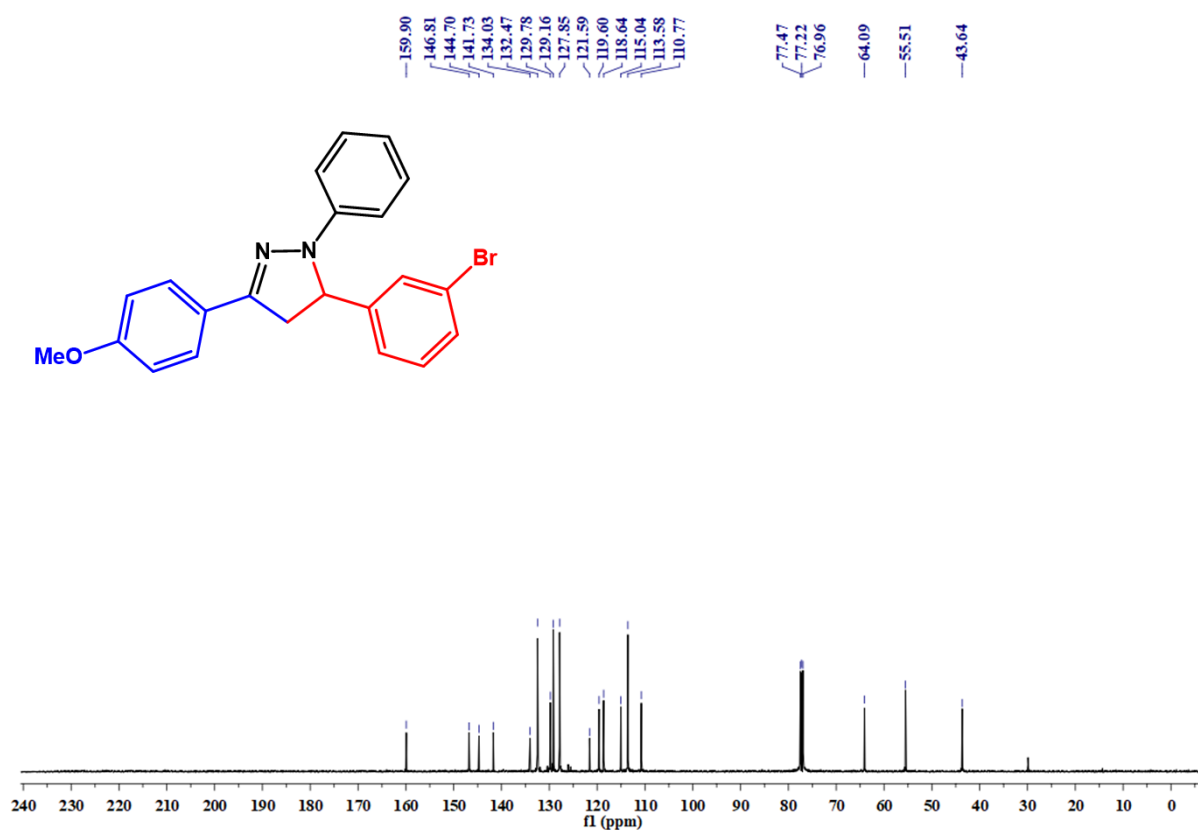


Figure S44: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4k** in CDCl_3 at 126 MHz.

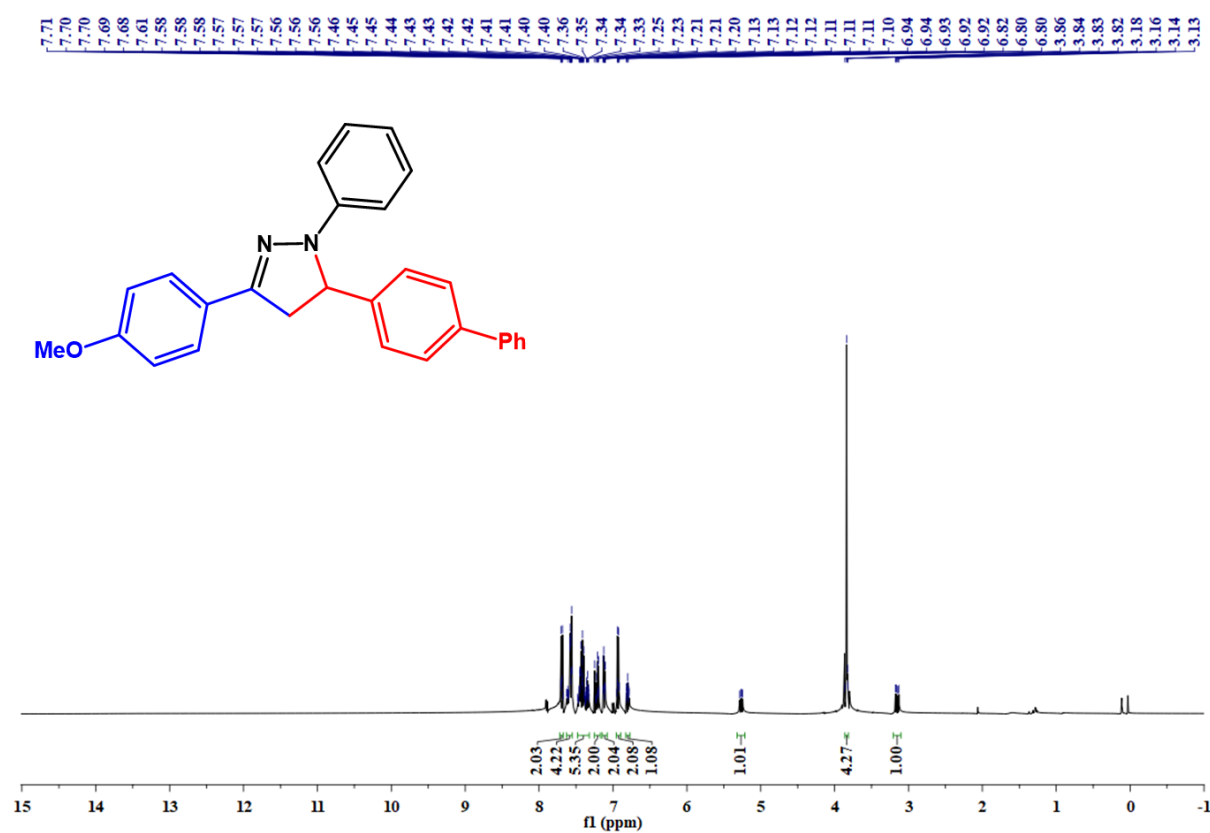


Figure S45: ^1H NMR spectrum of **4l** in CDCl_3 at 500 MHz.

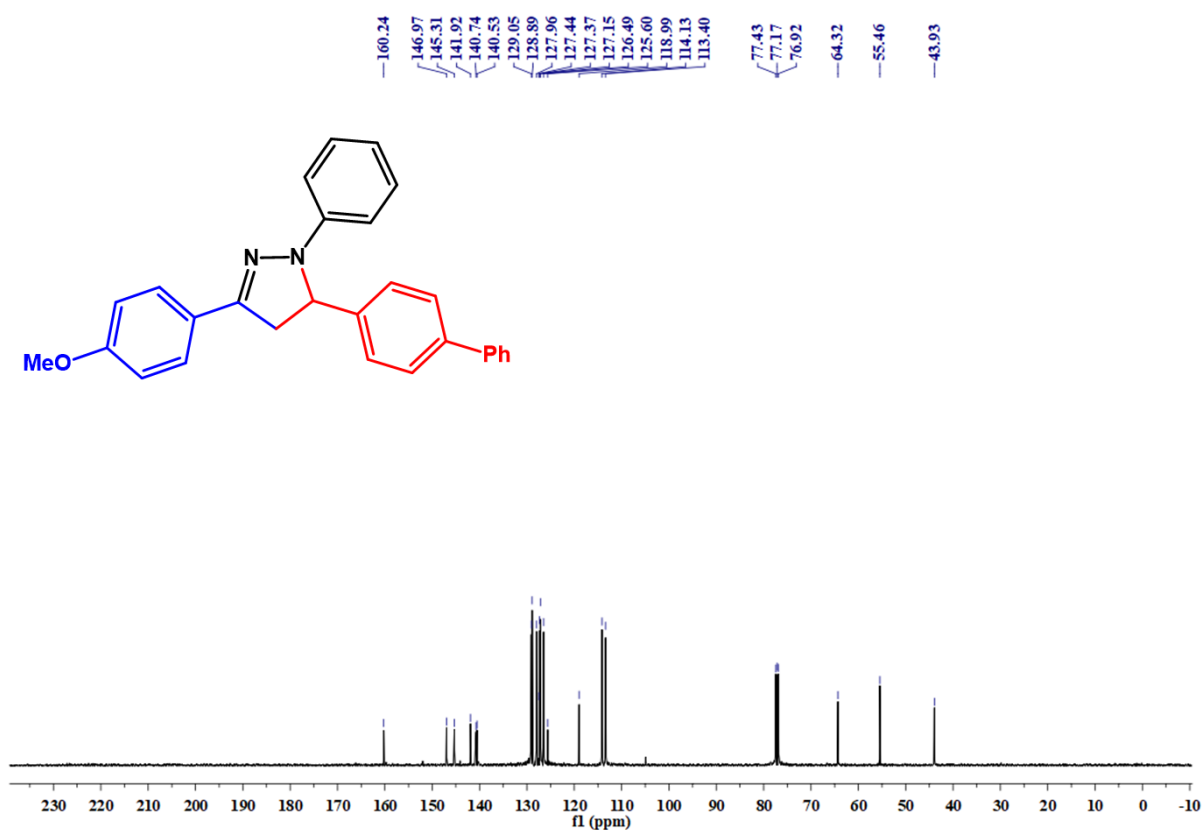


Figure S46: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4l** in CDCl_3 at 126 MHz.

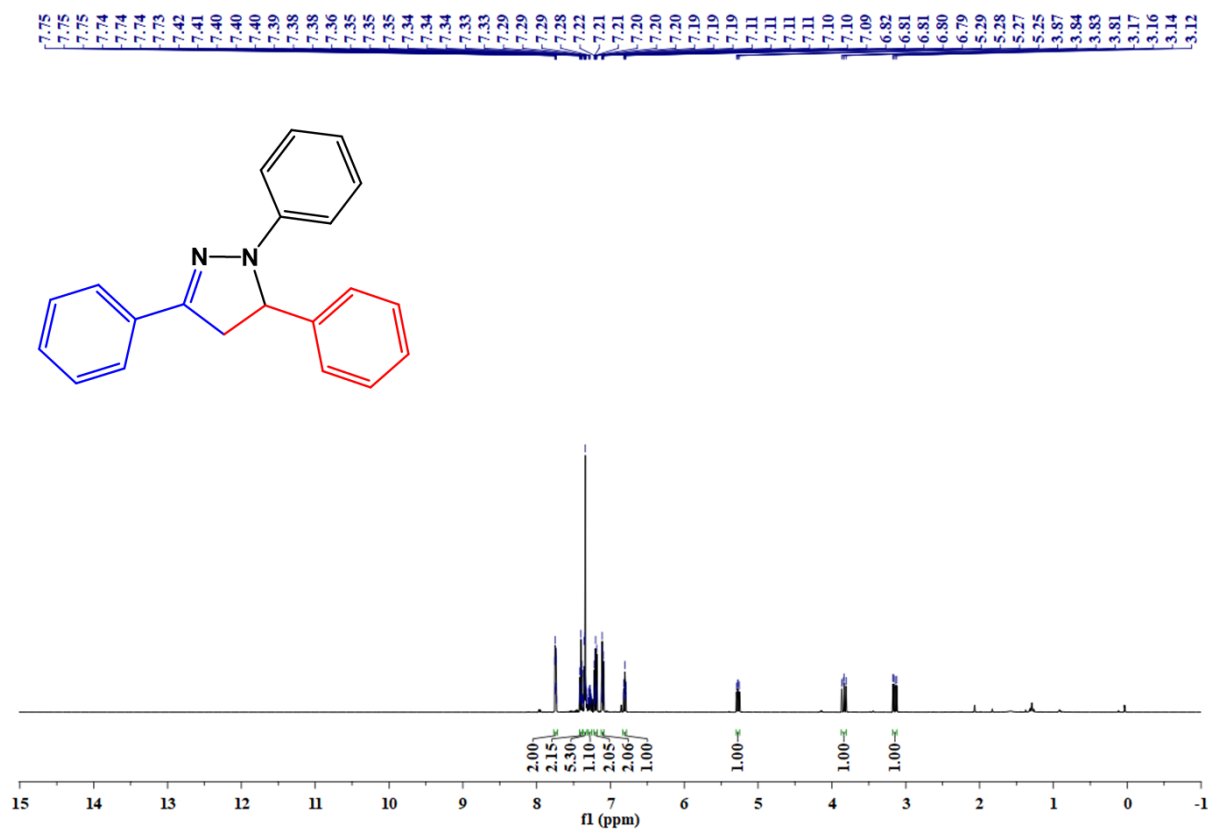


Figure S47: ^1H NMR spectrum of **4m** in CDCl_3 at 500 MHz.

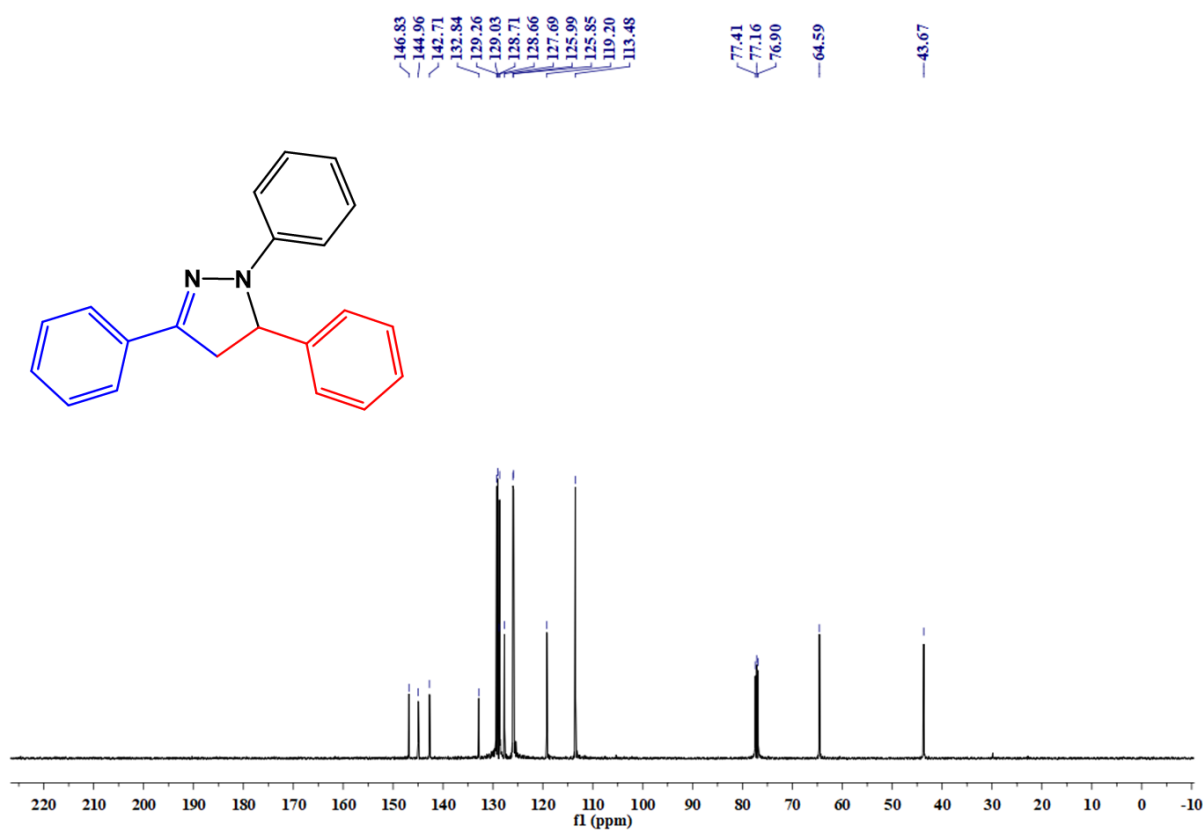


Figure S48: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4m** in CDCl_3 at 126 MHz.

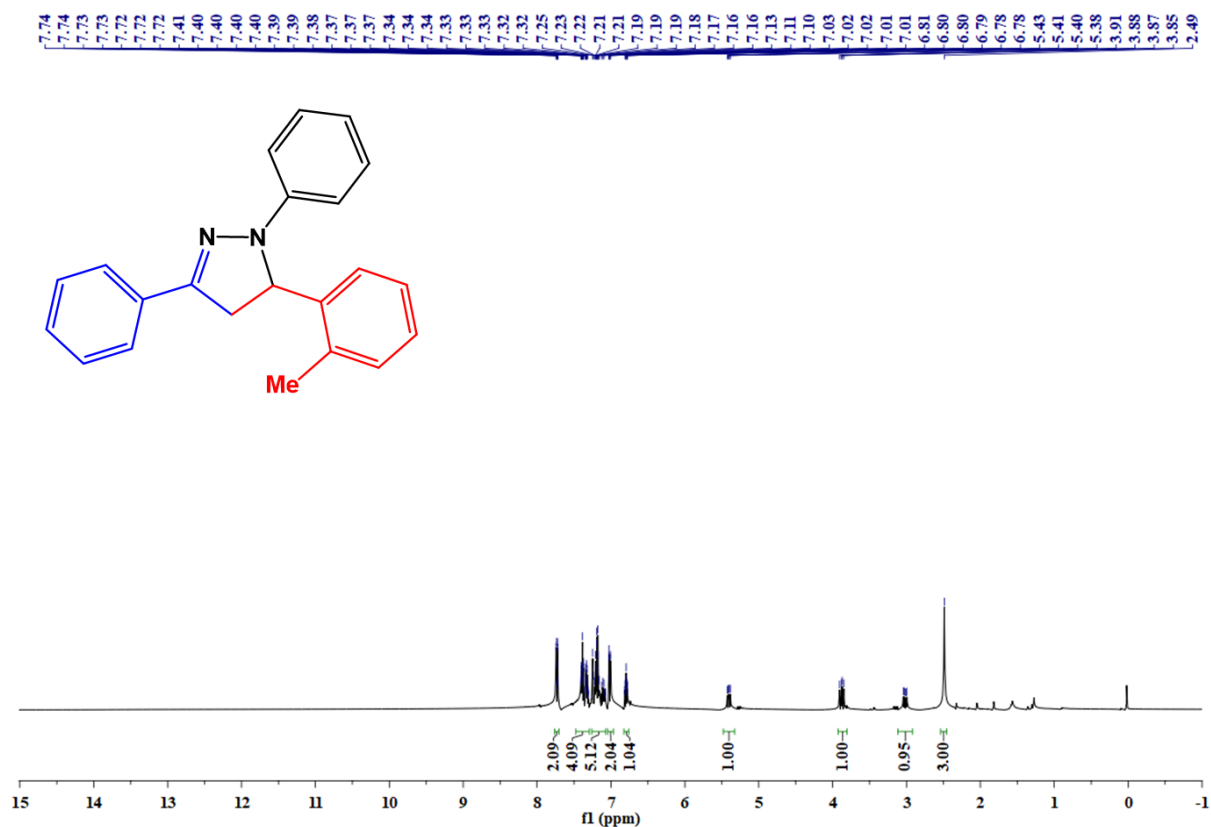


Figure S49: ^1H NMR spectrum of **4n** in CDCl_3 at 500 MHz.

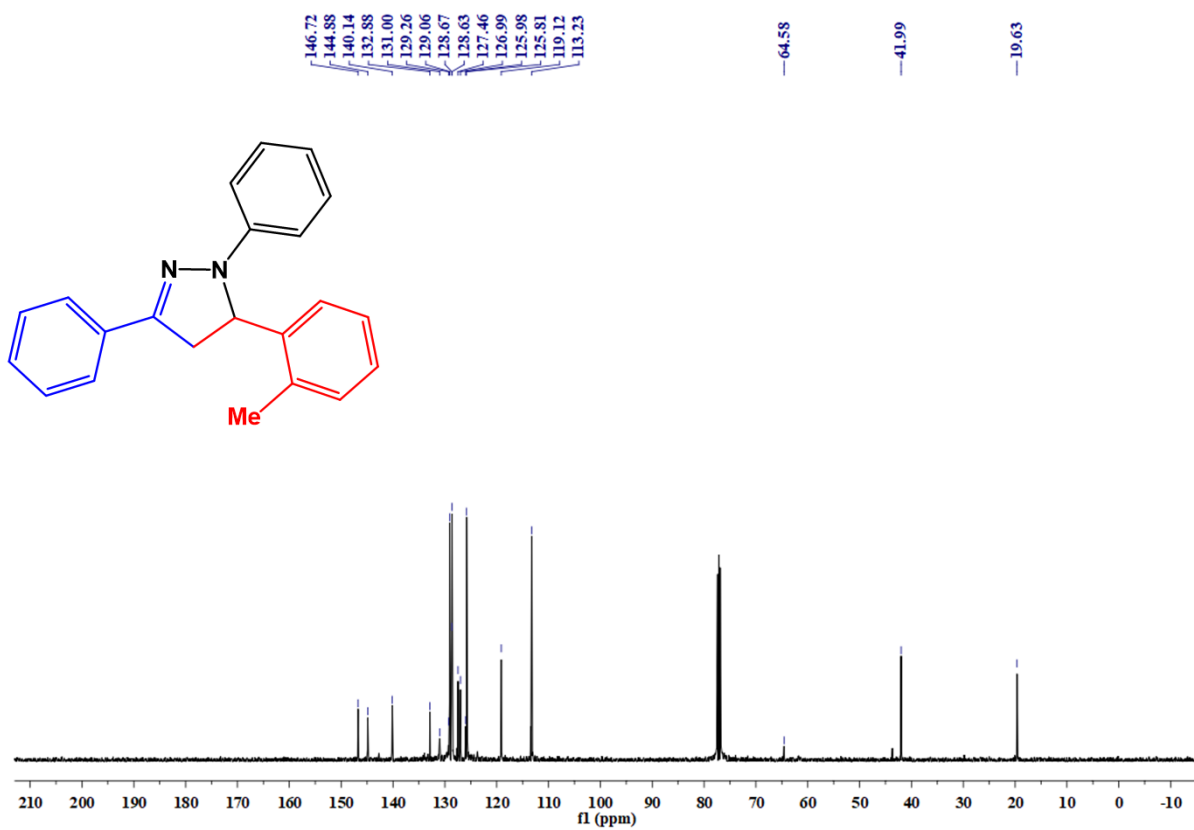


Figure S50: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4n** in CDCl_3 at 126 MHz.

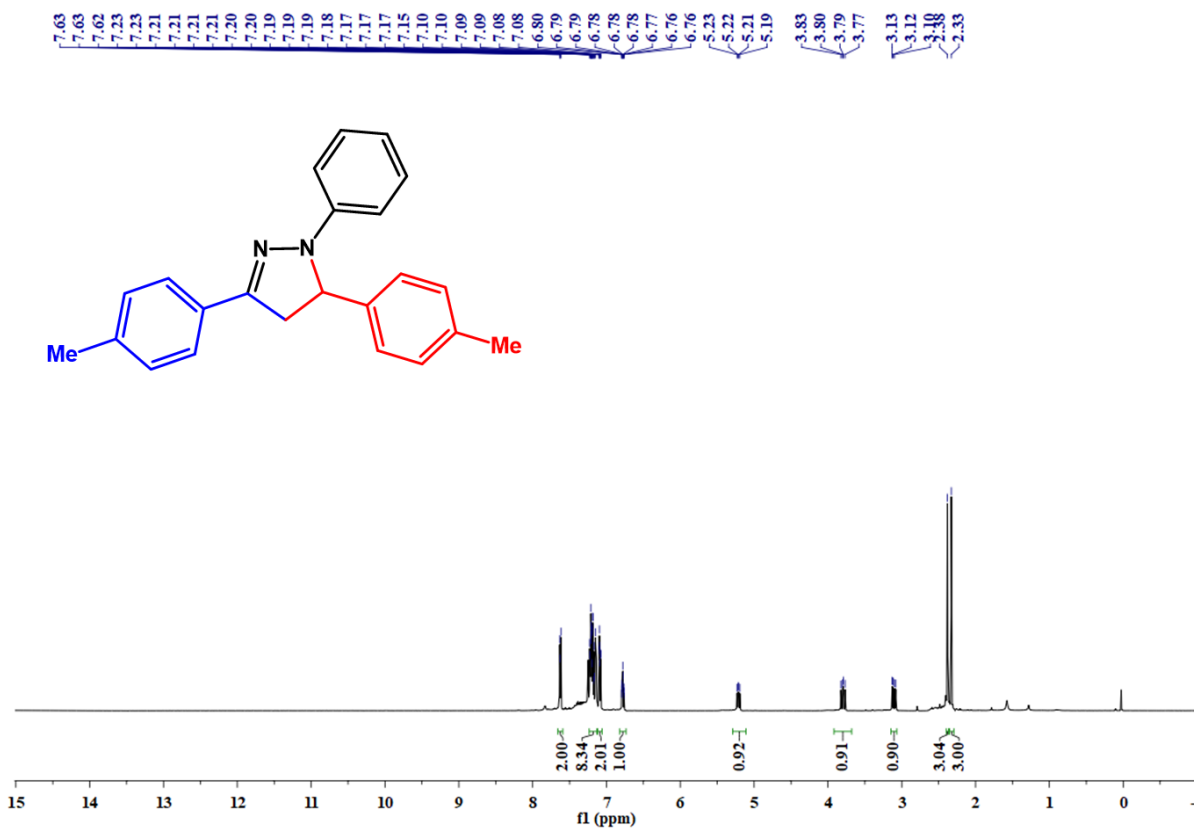


Figure S51: ^1H NMR spectrum of **4o** in CDCl_3 at 500 MHz.

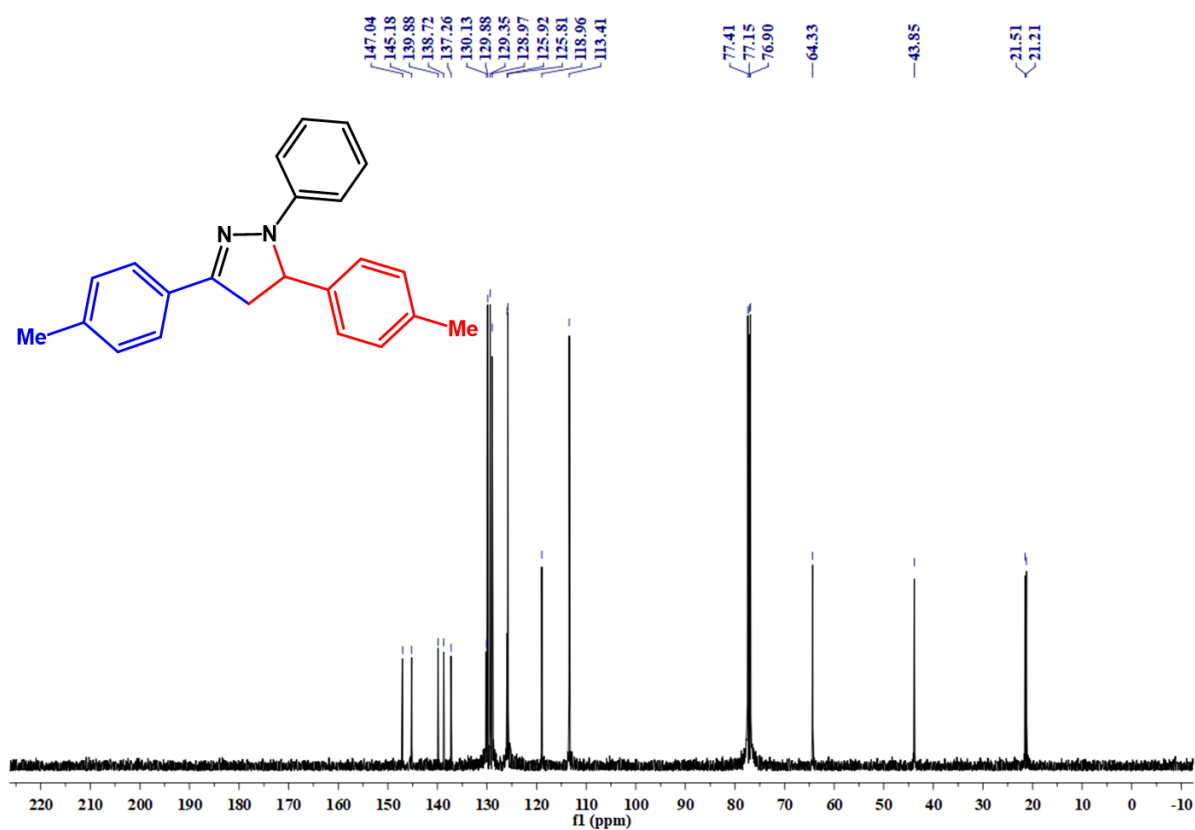


Figure S52: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4o** in CDCl_3 at 126 MHz.

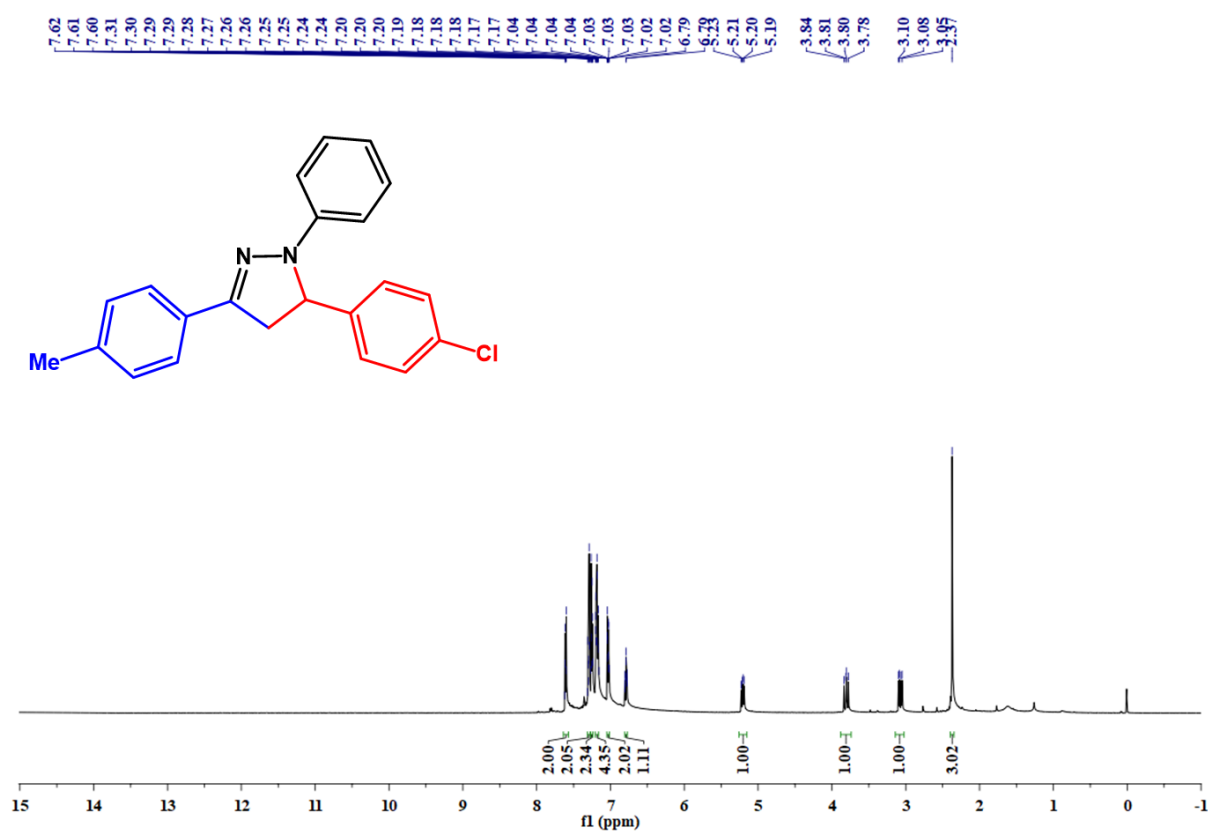


Figure S53: ^1H NMR spectrum of **4p** in CDCl_3 at 500 MHz.

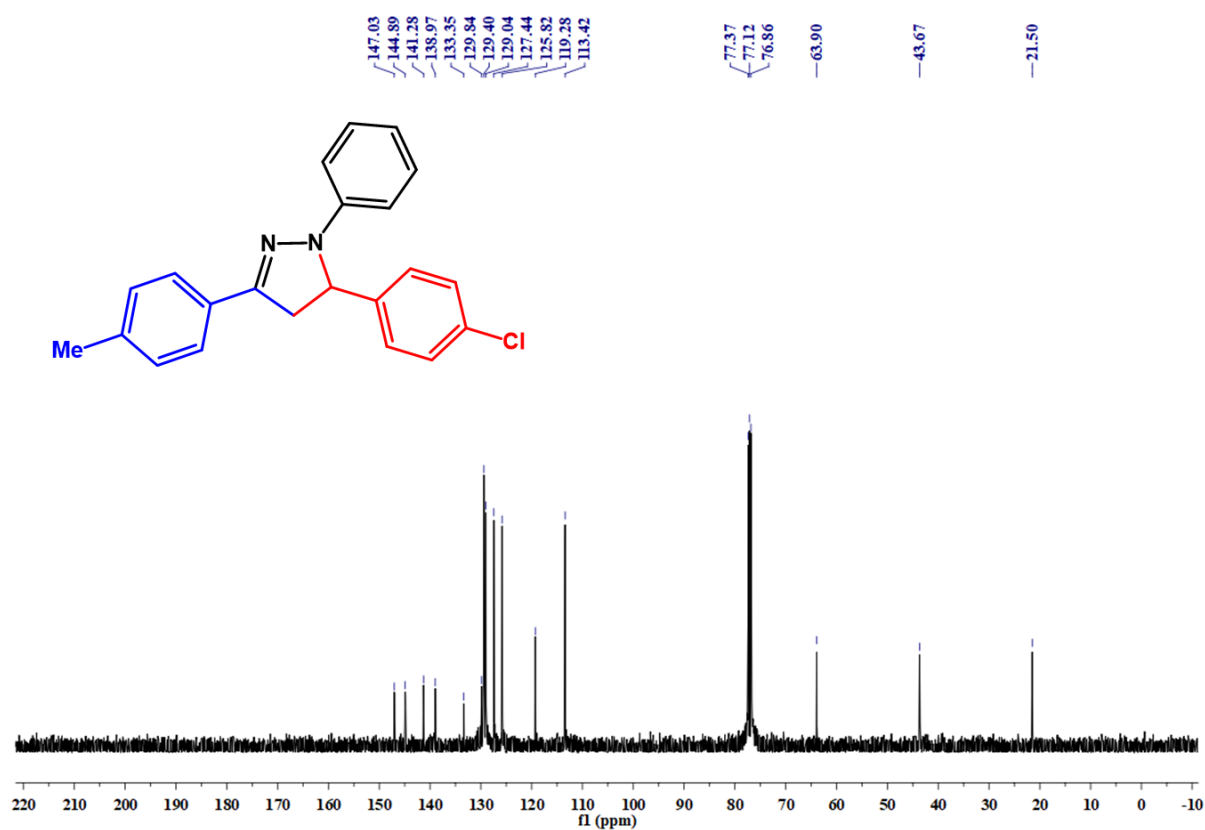


Figure S54: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4p** in CDCl_3 at 126 MHz.

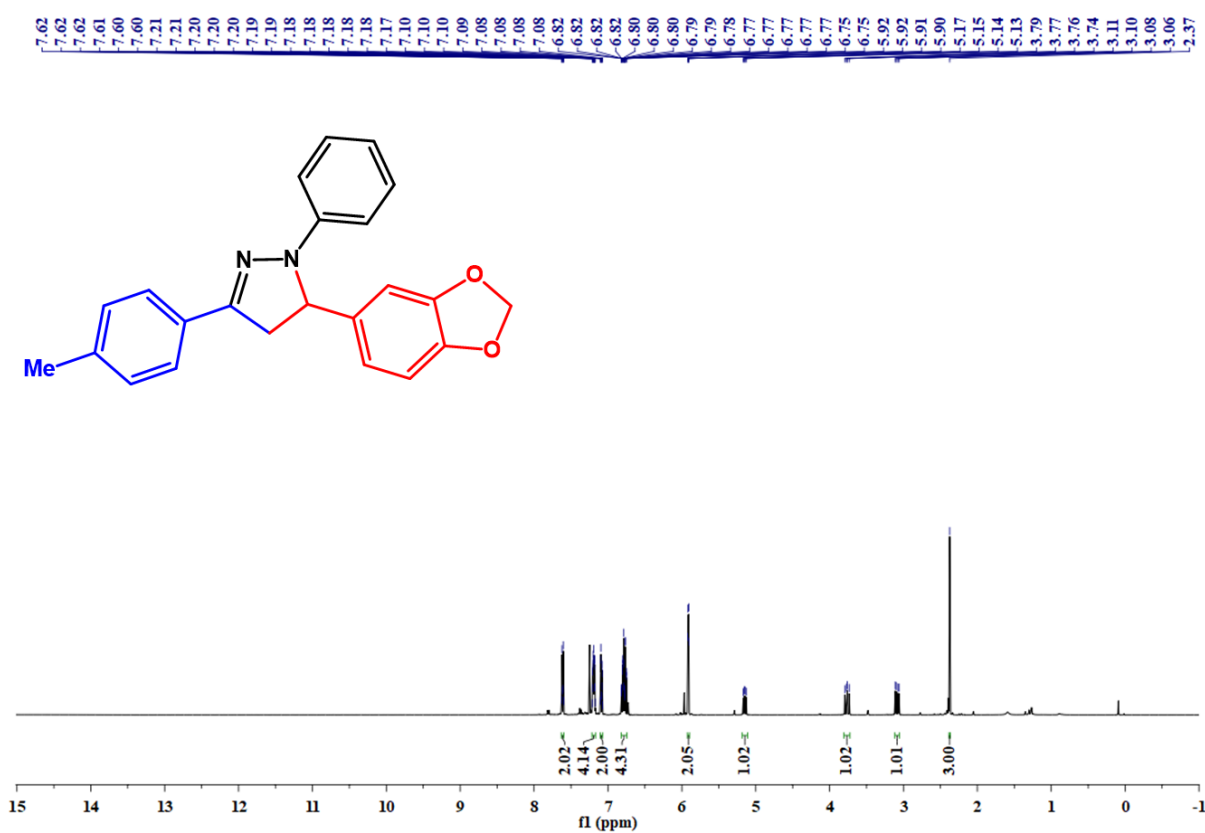


Figure S55: ^1H NMR spectrum of **4q** in CDCl_3 at 500 MHz.

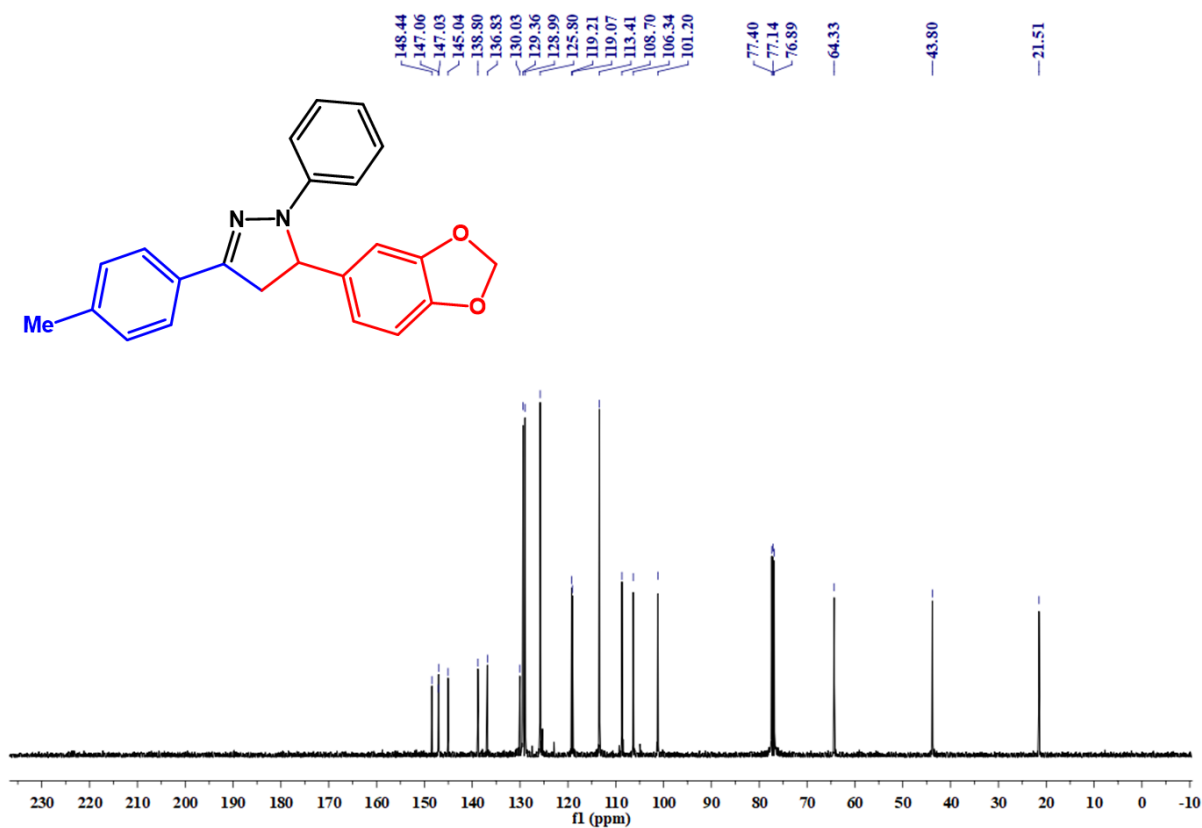


Figure S56: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4q** in CDCl_3 at 126 MHz.

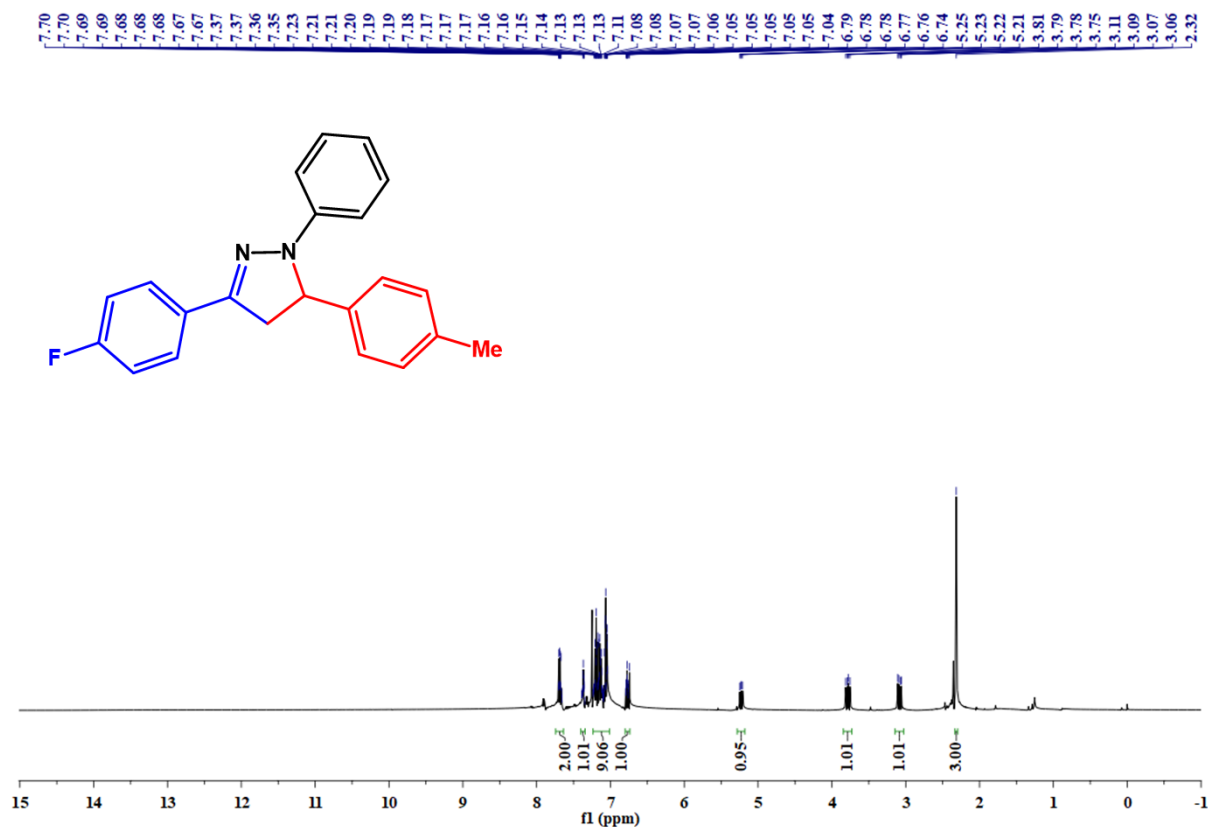


Figure S57: ^1H NMR spectrum of **4r** in CDCl_3 at 500 MHz.

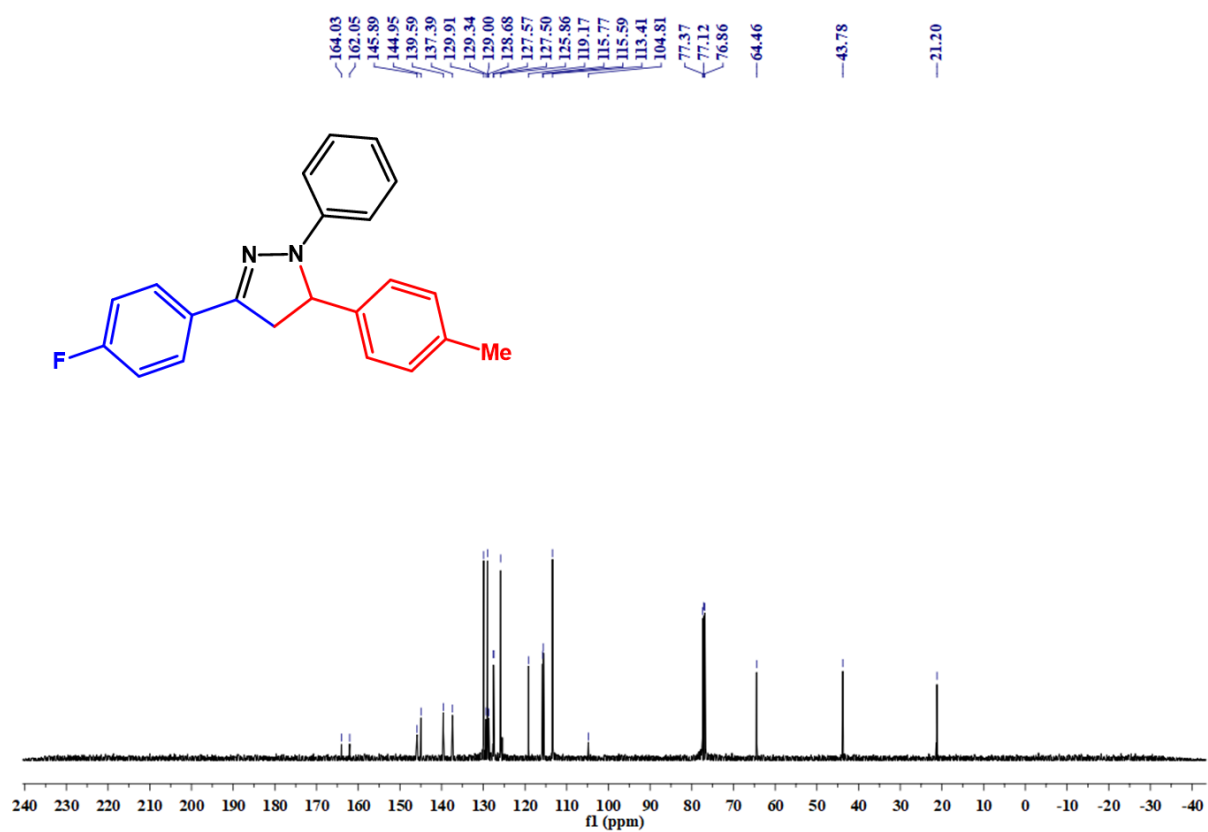


Figure S58: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4r** in CDCl_3 at 126 MHz.

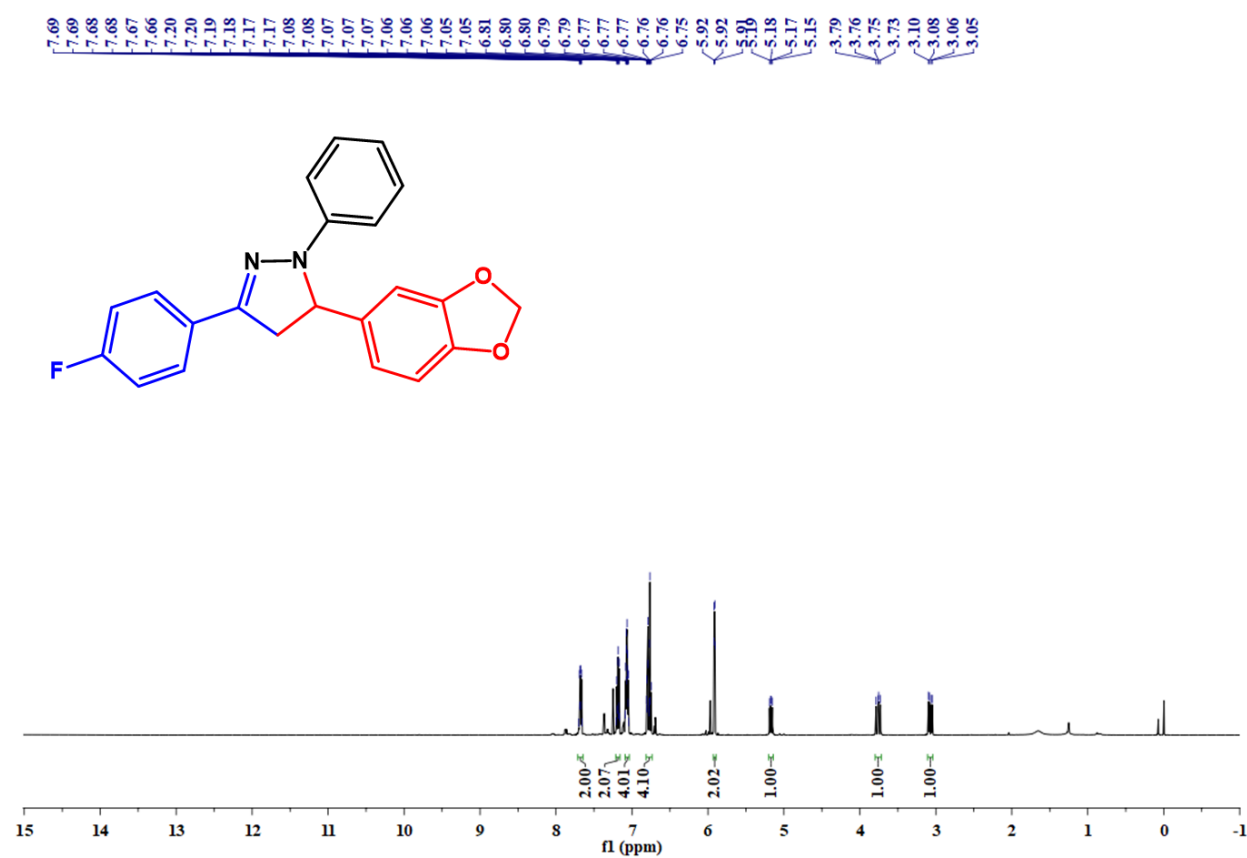


Figure S59: ^1H NMR spectrum of **4s** in CDCl_3 at 500 MHz.

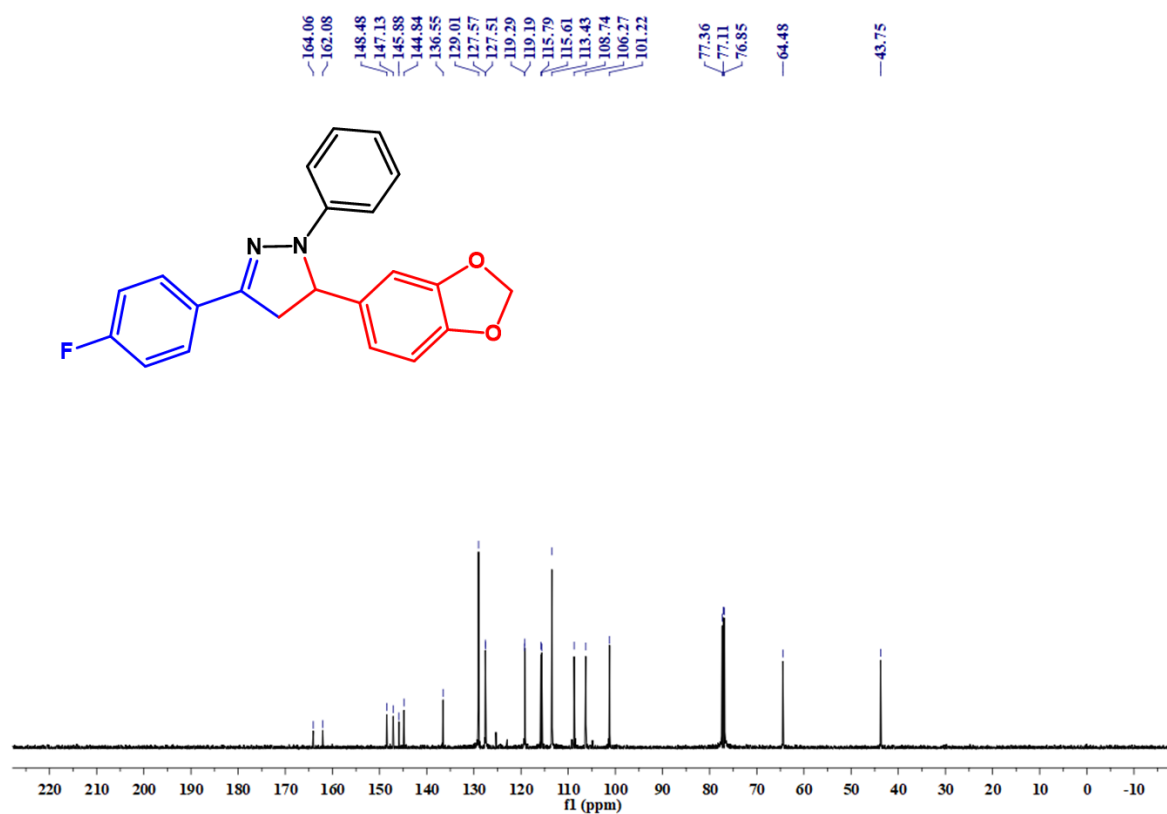


Figure S60: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4s** in CDCl_3 at 126 MHz.

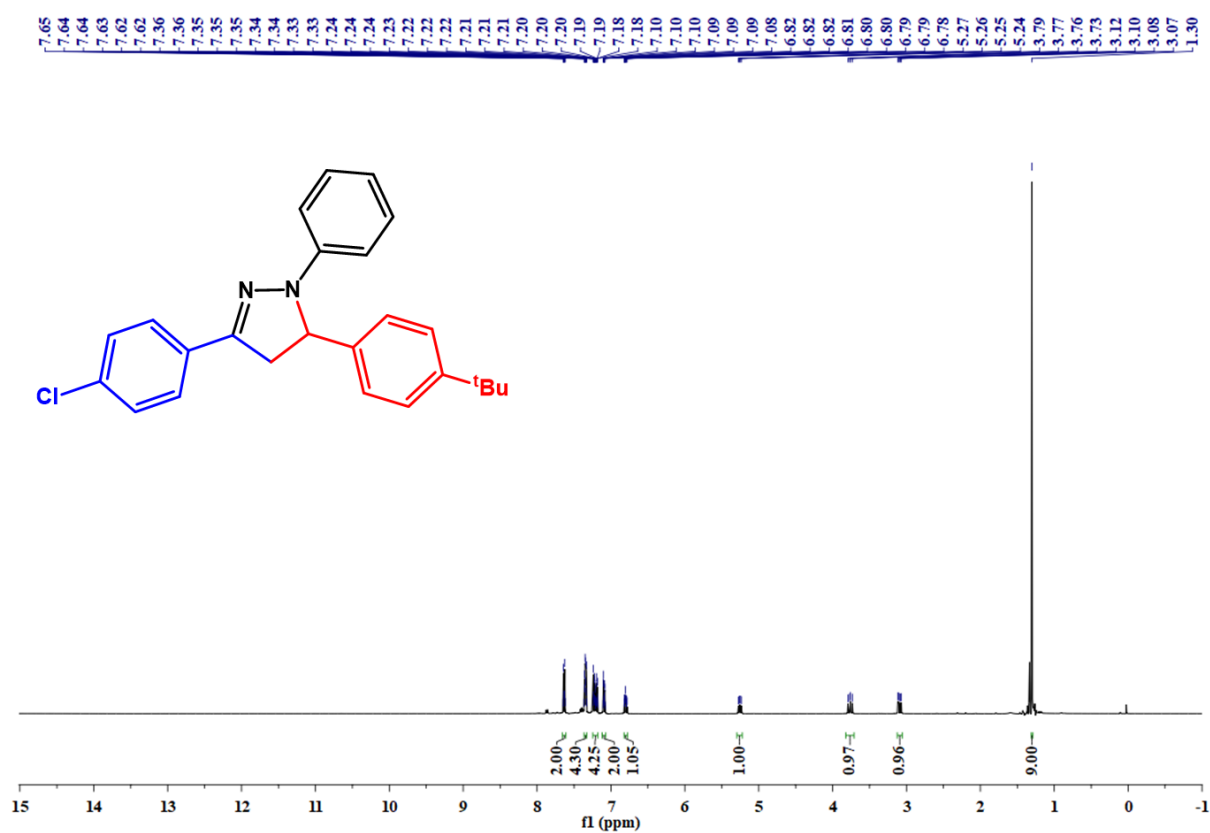


Figure S61: ^1H NMR spectrum of **4t** in CDCl_3 at 500 MHz.

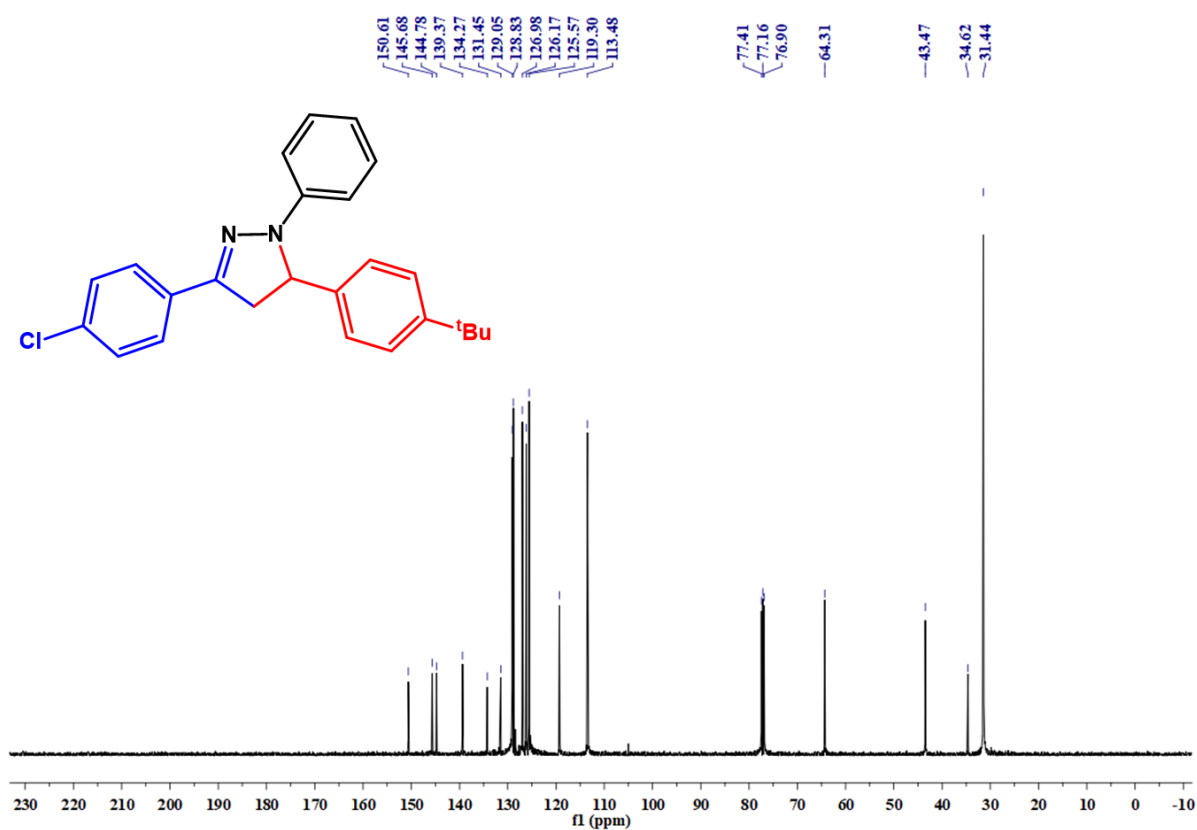


Figure S62: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4t** in CDCl_3 at 126 MHz.

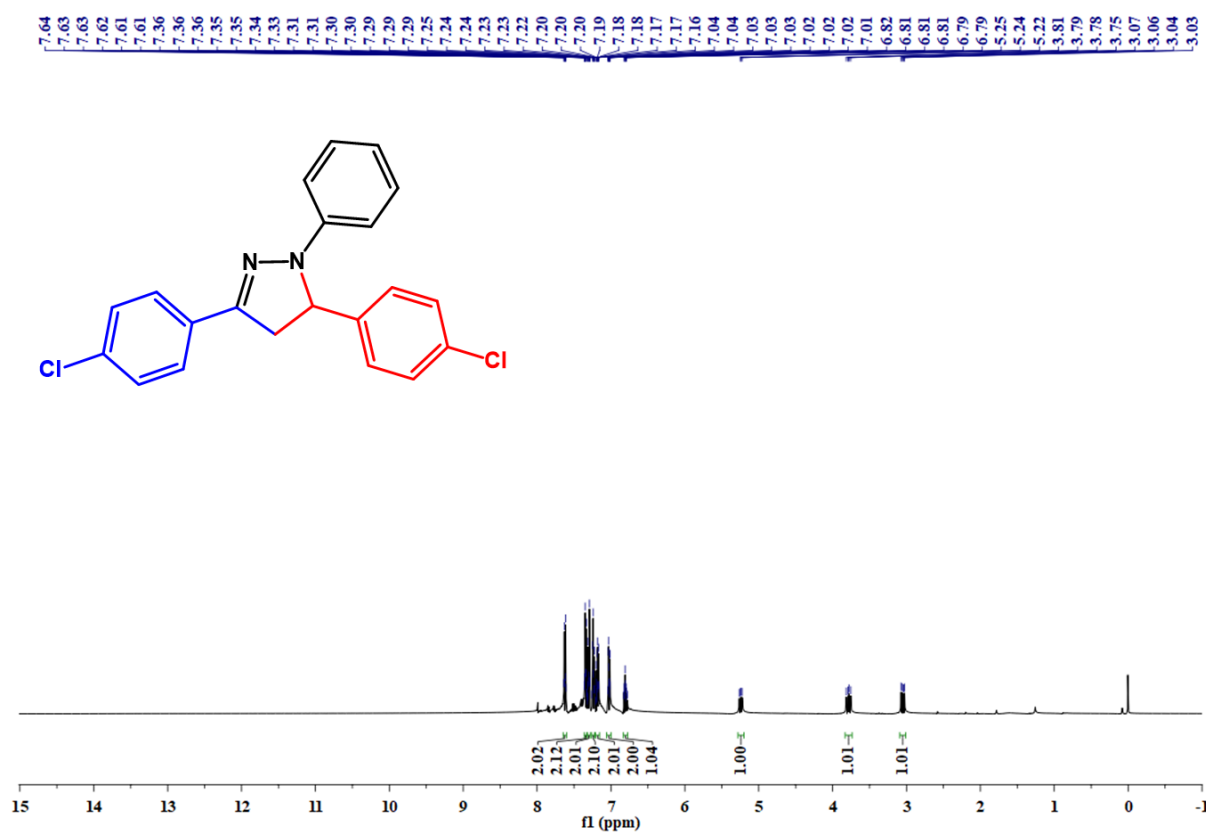


Figure S63: ^1H NMR spectrum of **4u** in CDCl_3 at 500 MHz.

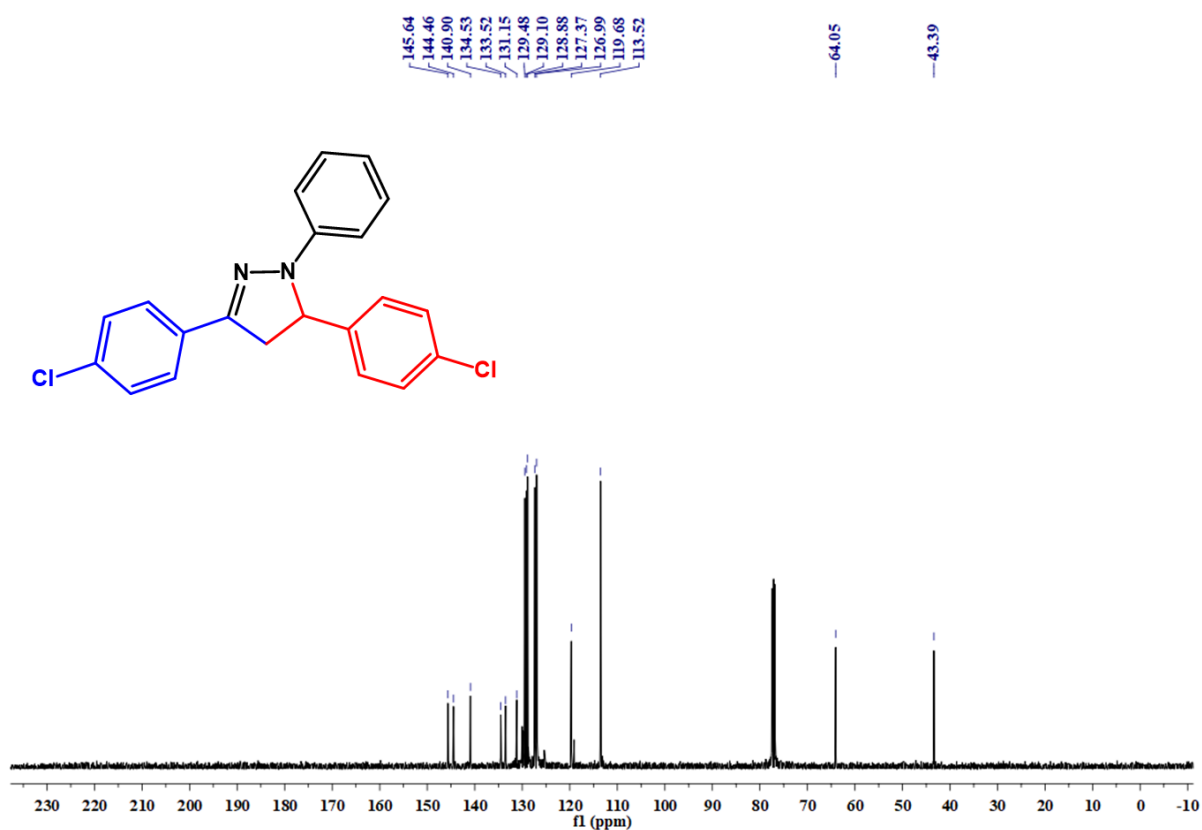


Figure S64: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4u** in CDCl_3 at 126 MHz.

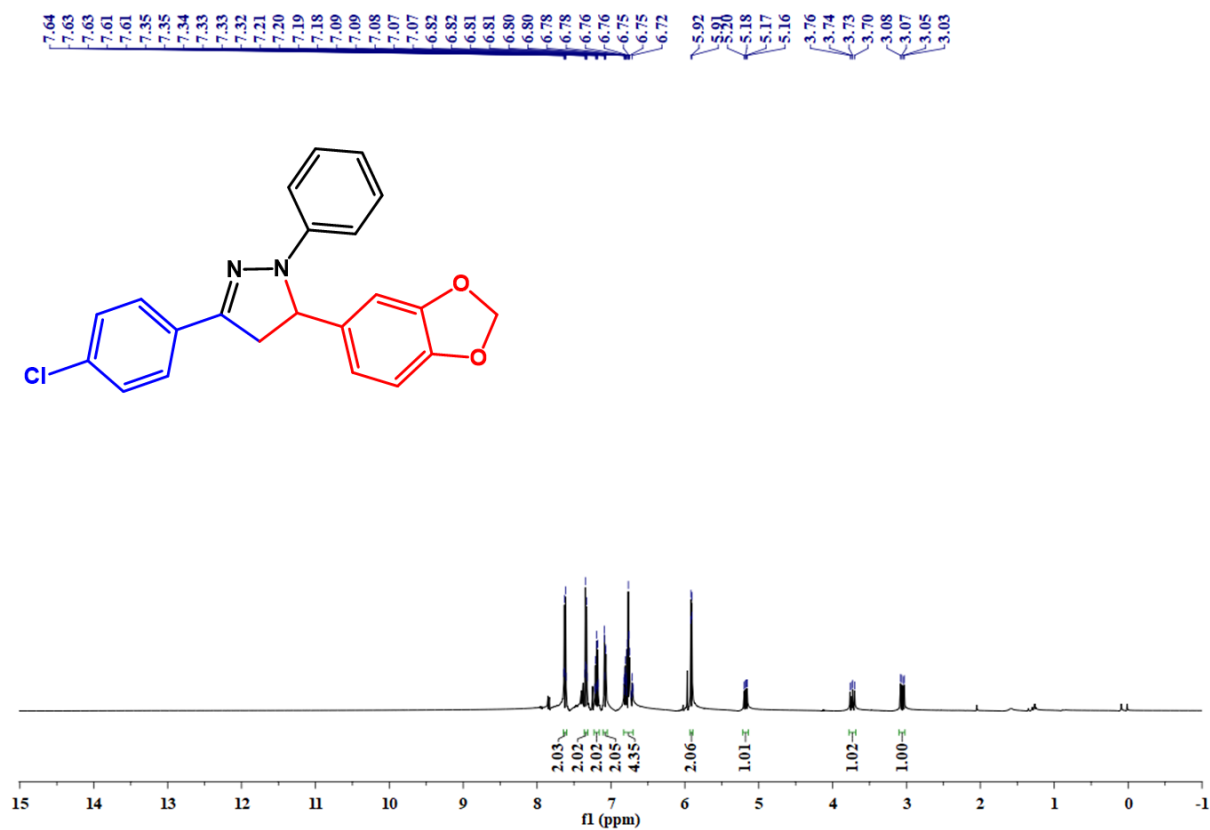


Figure S65: ^1H NMR spectrum of **4v** in CDCl_3 at 500 MHz.

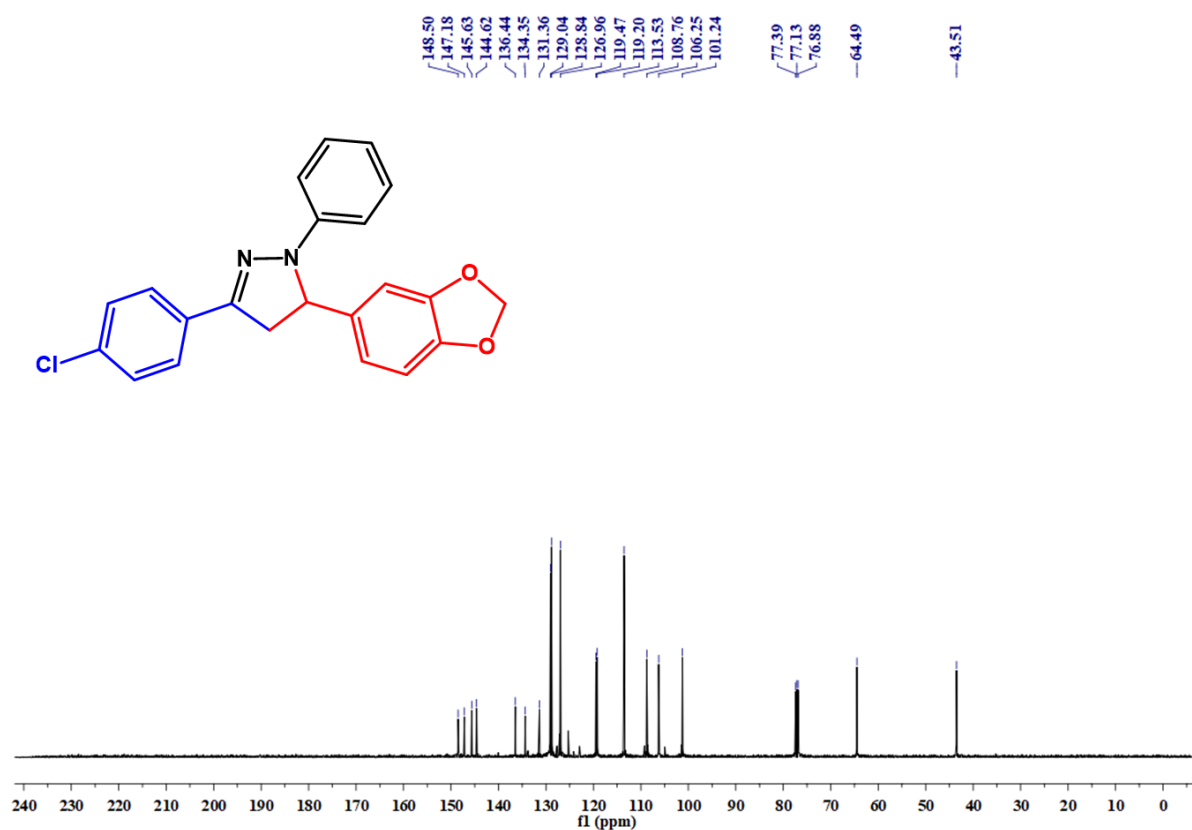


Figure S66: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4v** in CDCl_3 at 126 MHz.

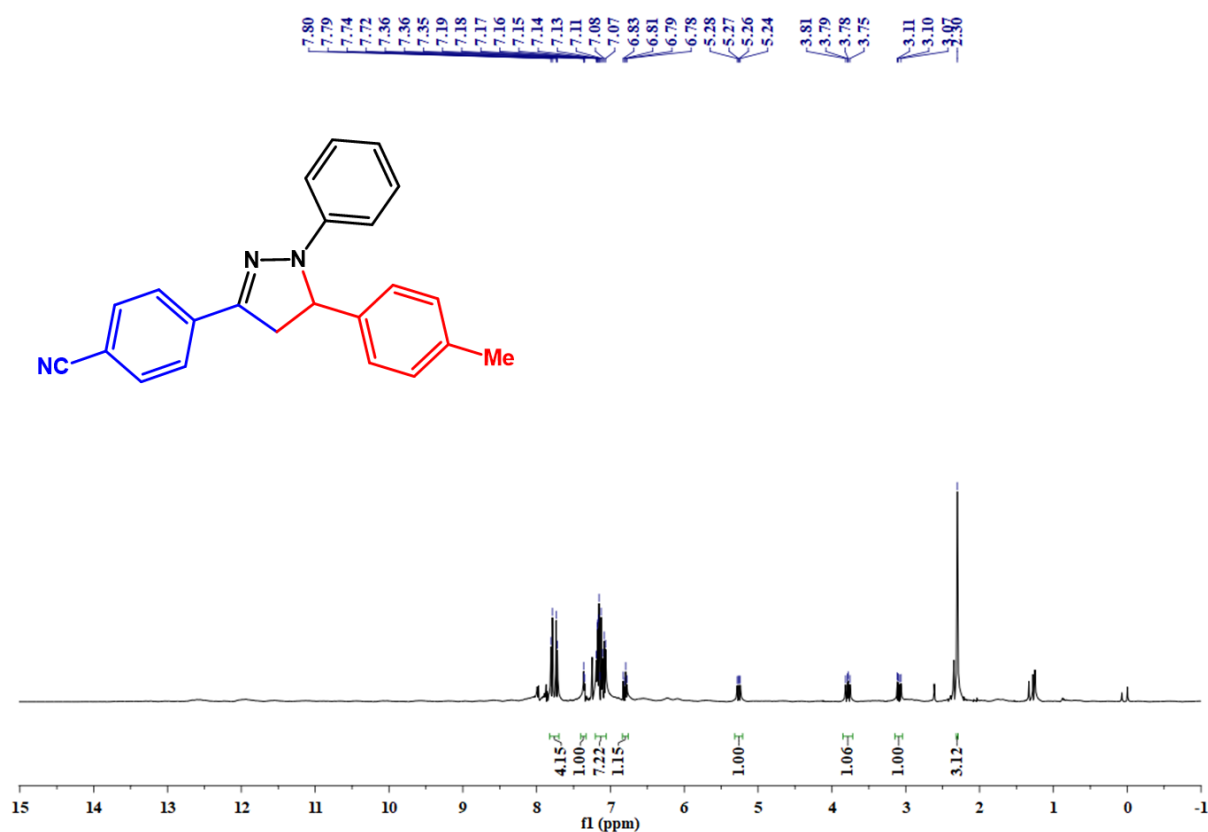


Figure S67: ^1H NMR spectrum of **4w** in CDCl_3 at 500 MHz.

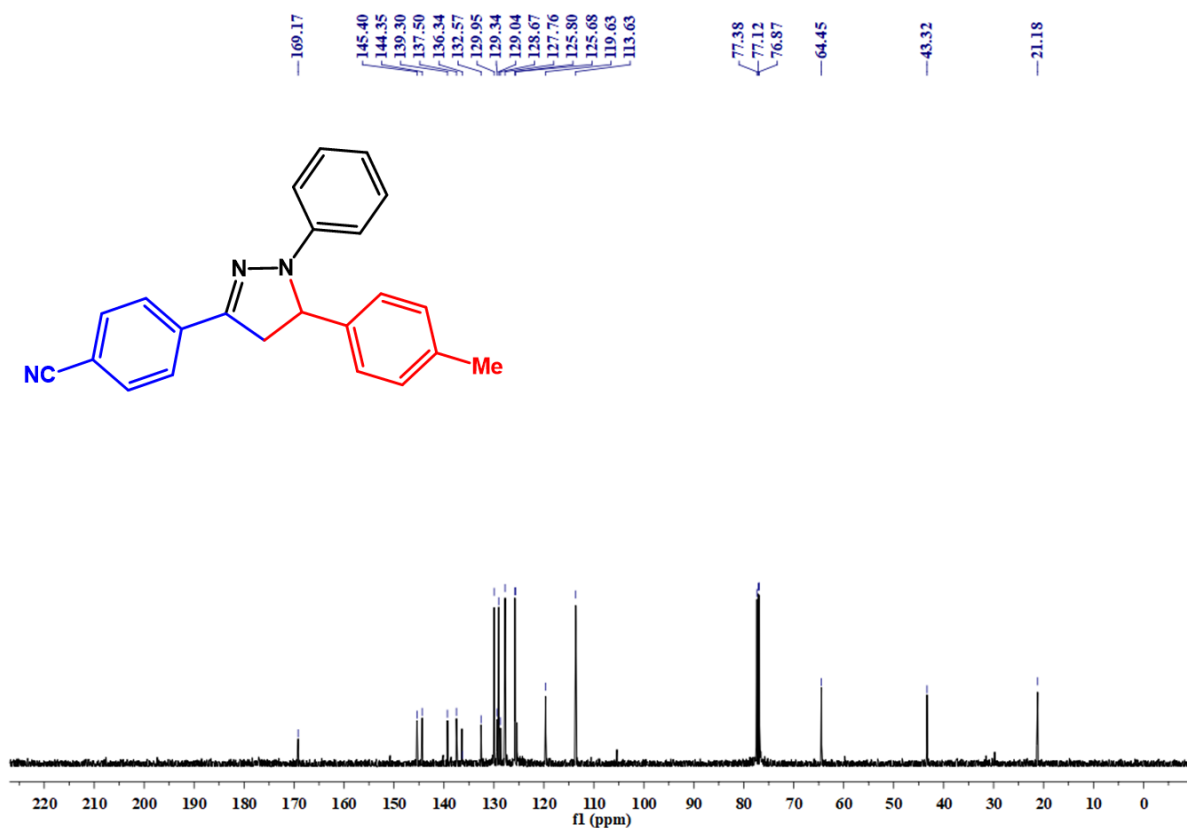


Figure S68: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4w** in CDCl_3 at 126 MHz.

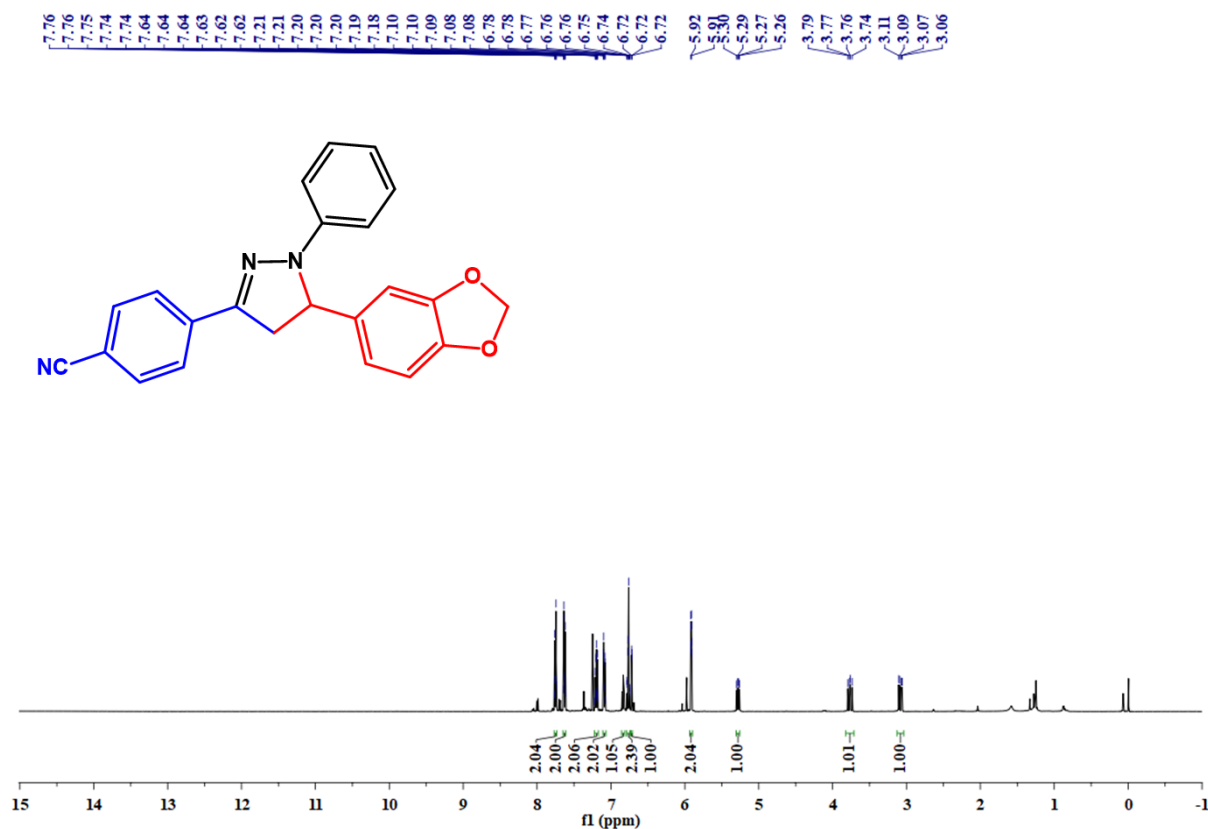


Figure S69: ^1H NMR spectrum of **4x** in CDCl_3 at 500 MHz.

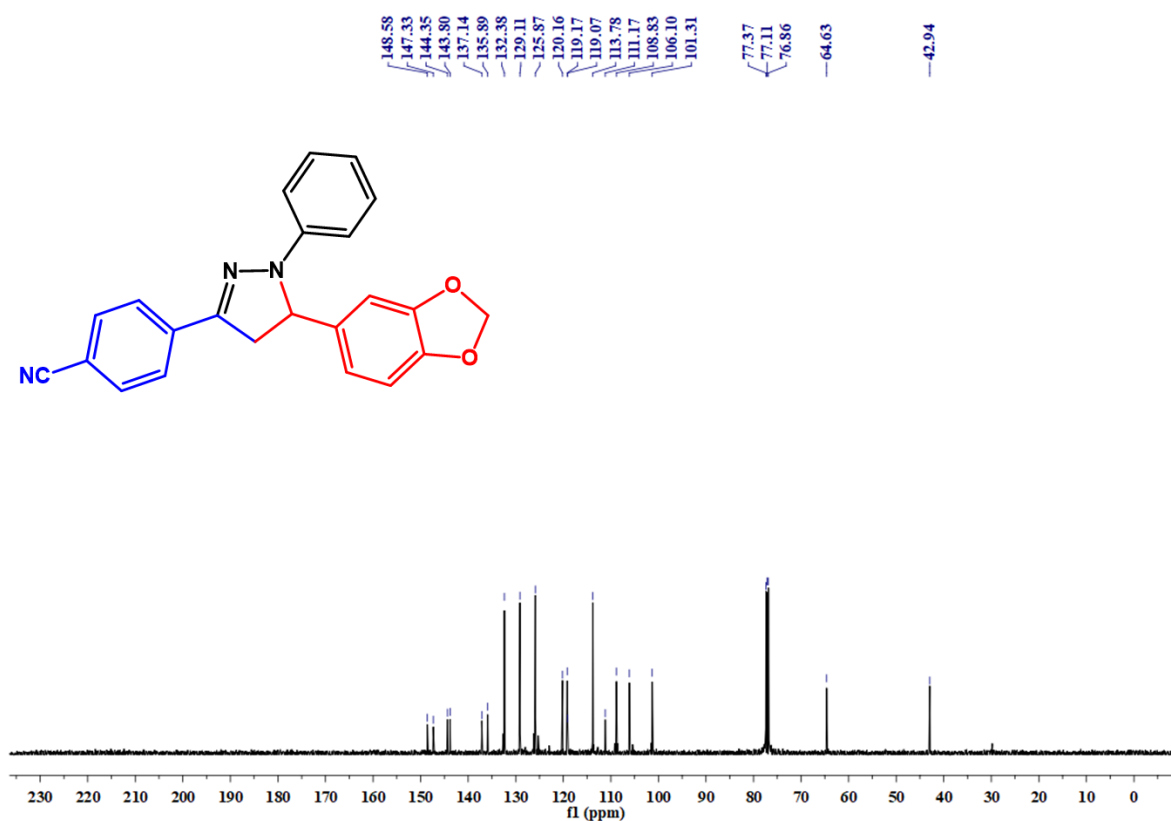


Figure S70: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4x** in CDCl_3 at 126 MHz.

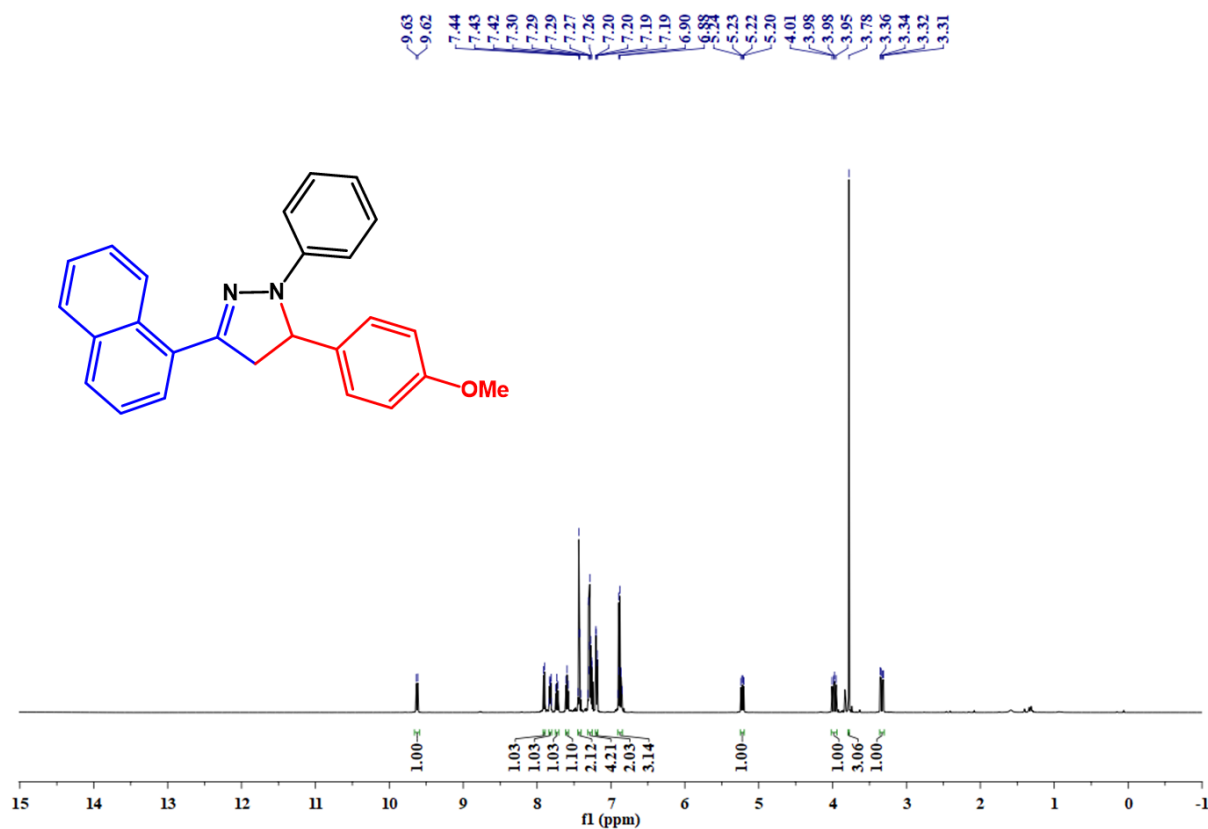


Figure S71: ^1H NMR spectrum of **4y** in CDCl_3 at 500 MHz.

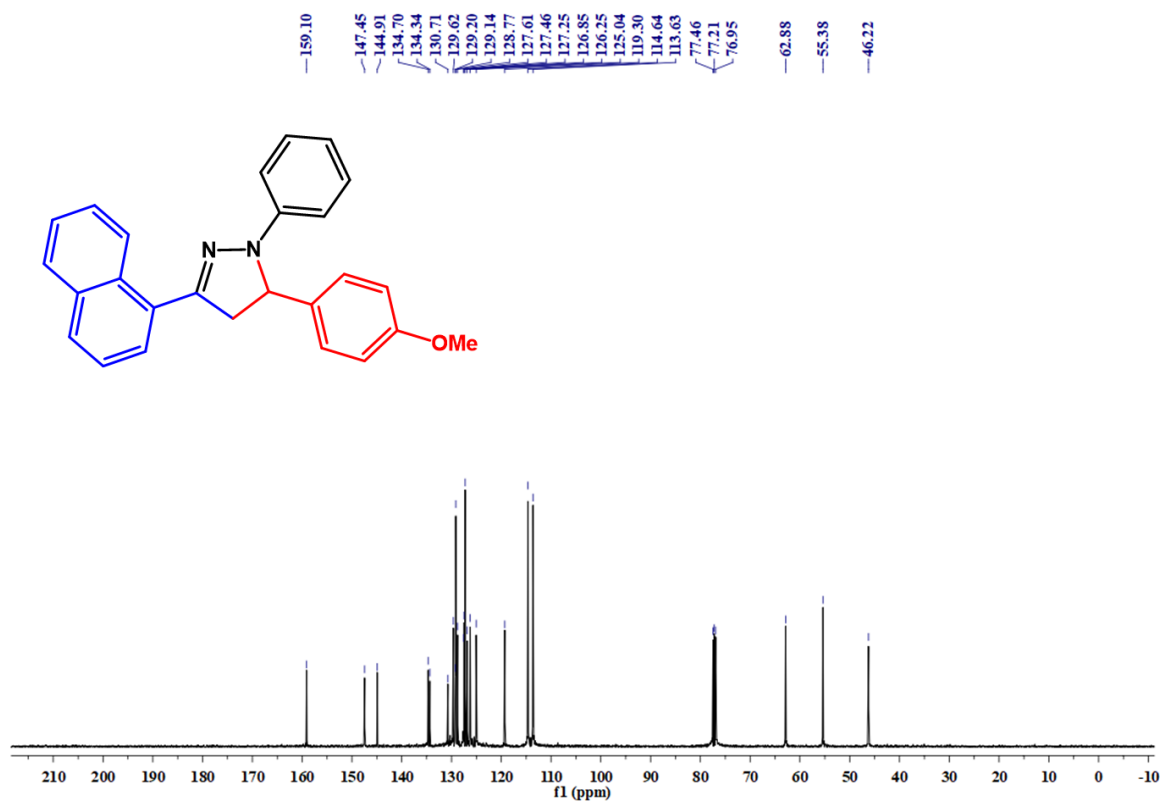


Figure S72: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4y** in CDCl_3 at 126 MHz.

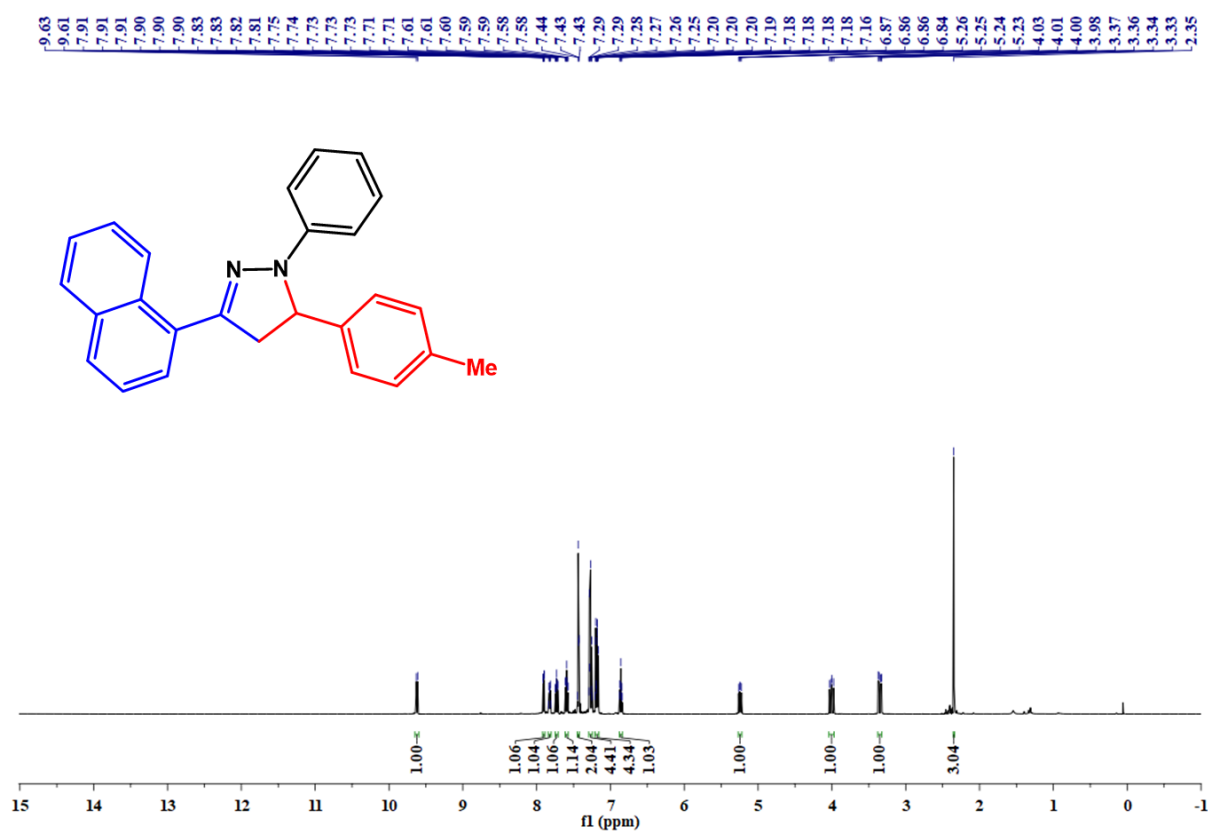


Figure S73: ^1H NMR spectrum of **4z** in CDCl_3 at 500 MHz.

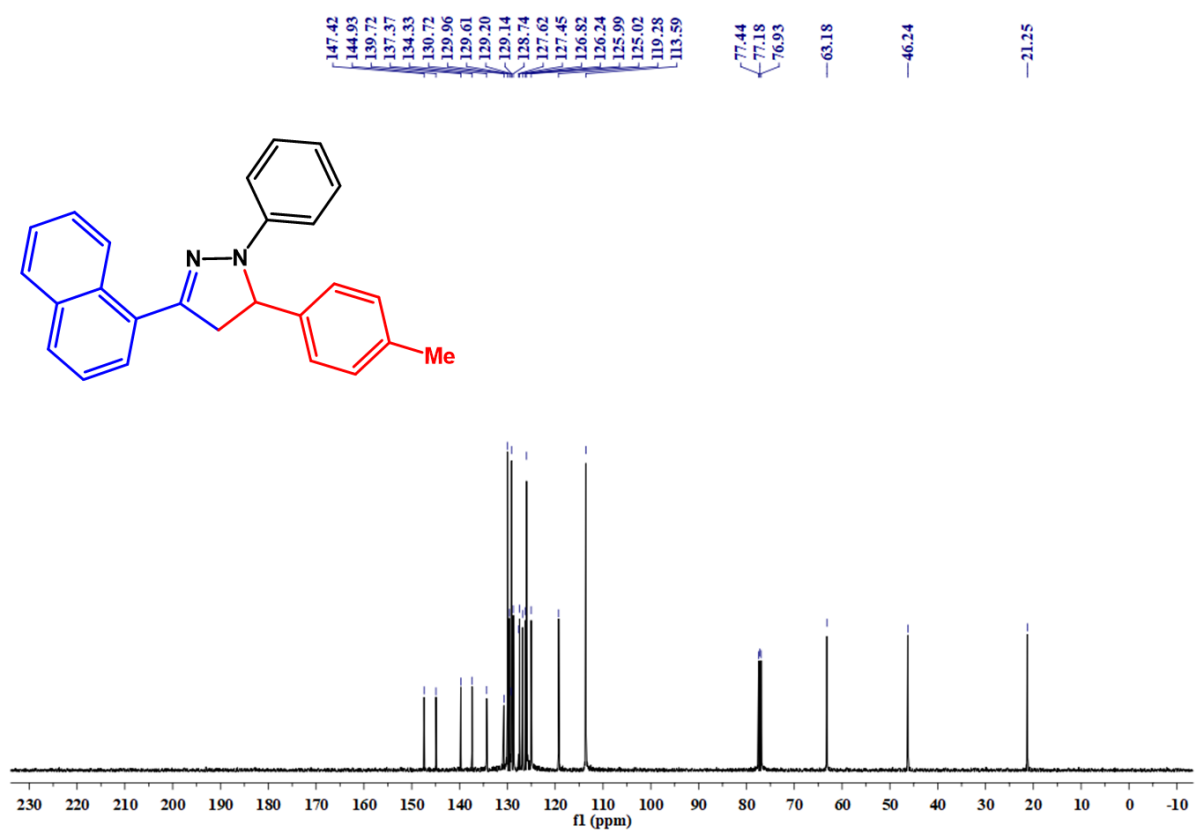


Figure S74: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4z** in CDCl_3 at 126 MHz.

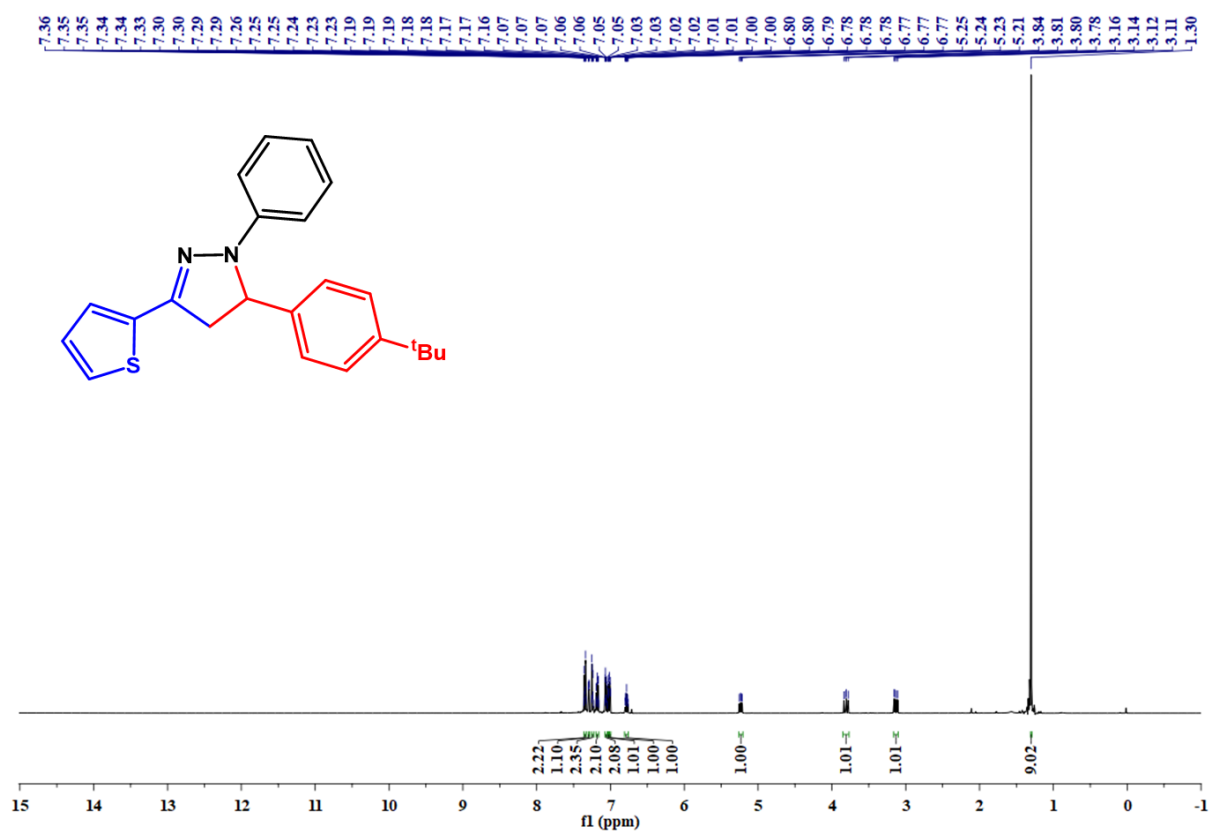


Figure S75: ^1H NMR spectrum of **4aa** in CDCl_3 at 500 MHz.

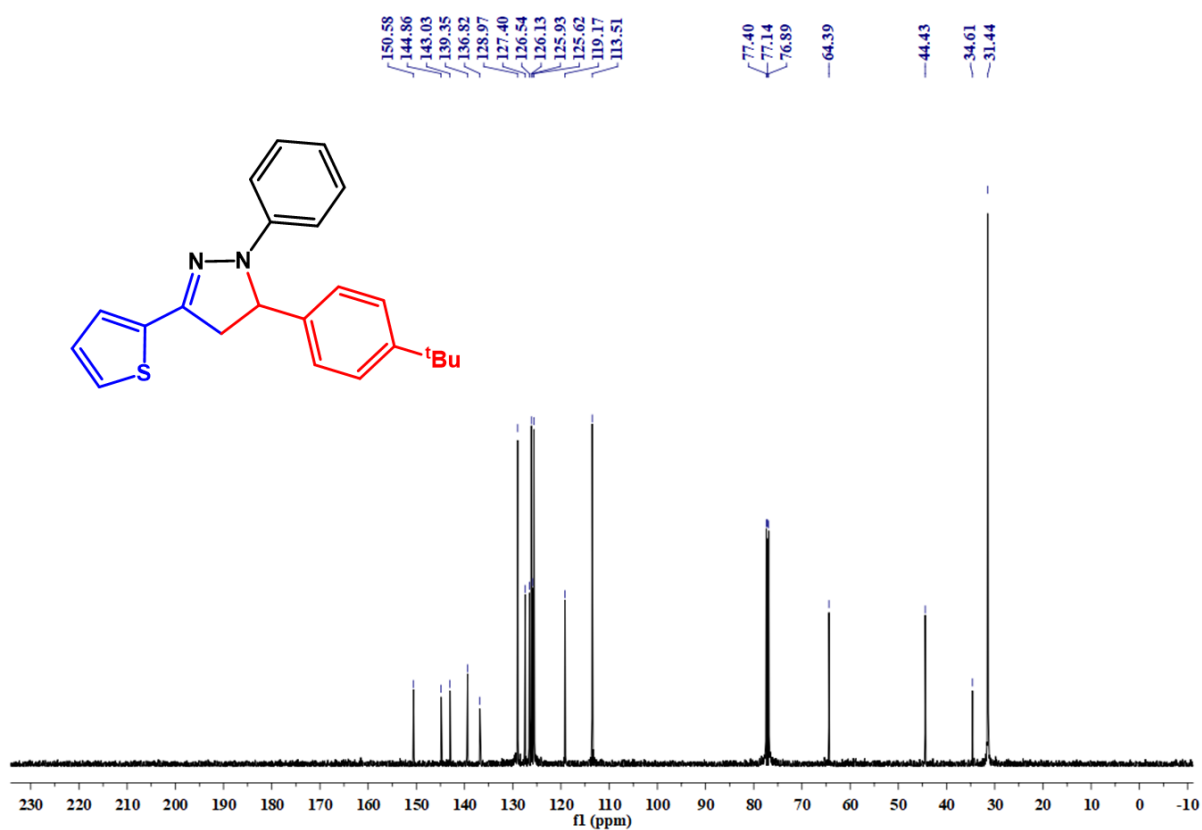


Figure S76: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4aa** in CDCl_3 at 126 MHz.

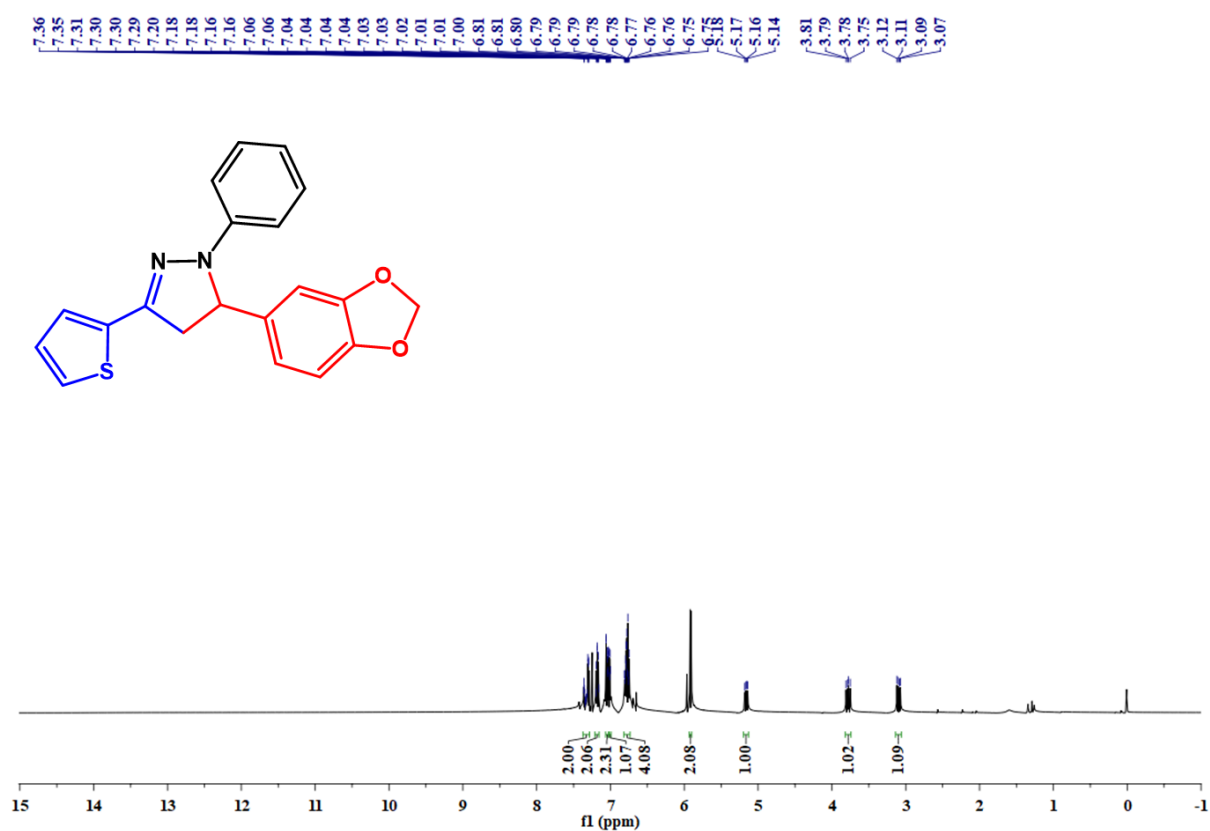


Figure S77: ^1H NMR spectrum of **4ab** in CDCl_3 at 500 MHz.

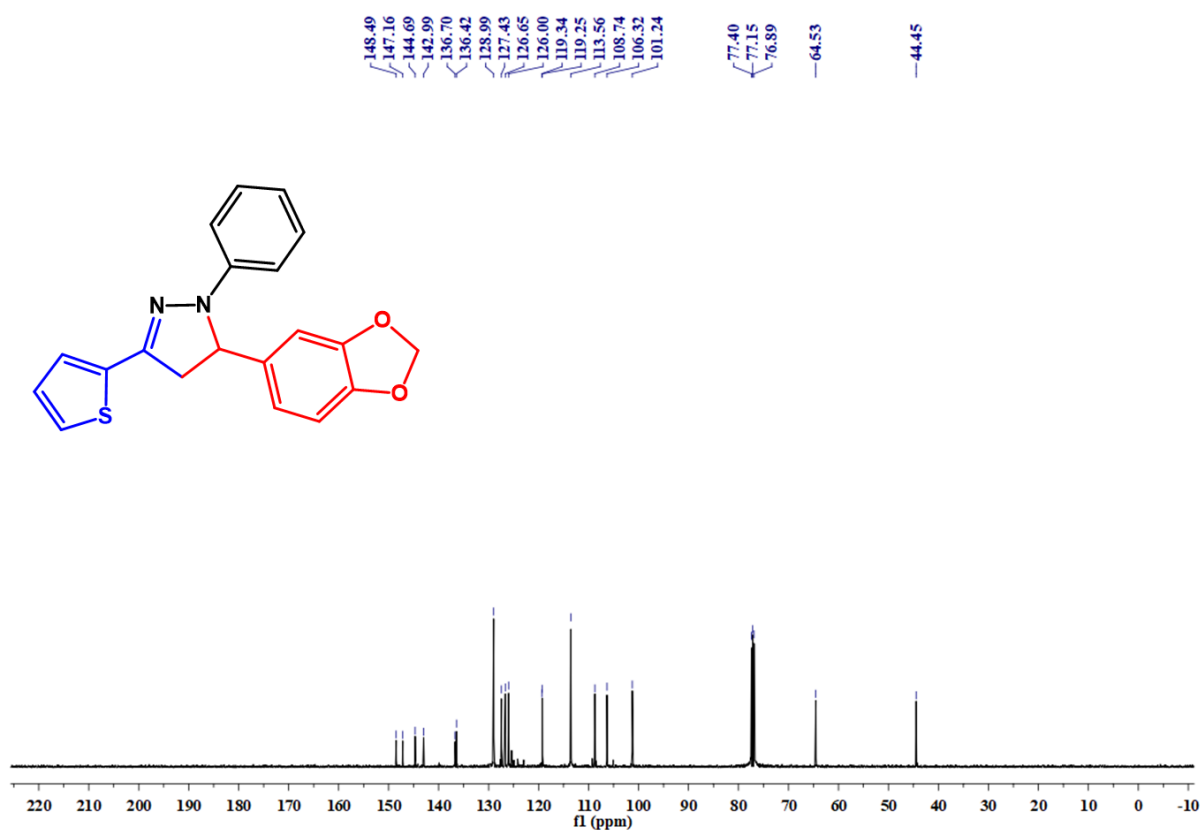


Figure S78: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4ab** in CDCl_3 at 126 MHz.

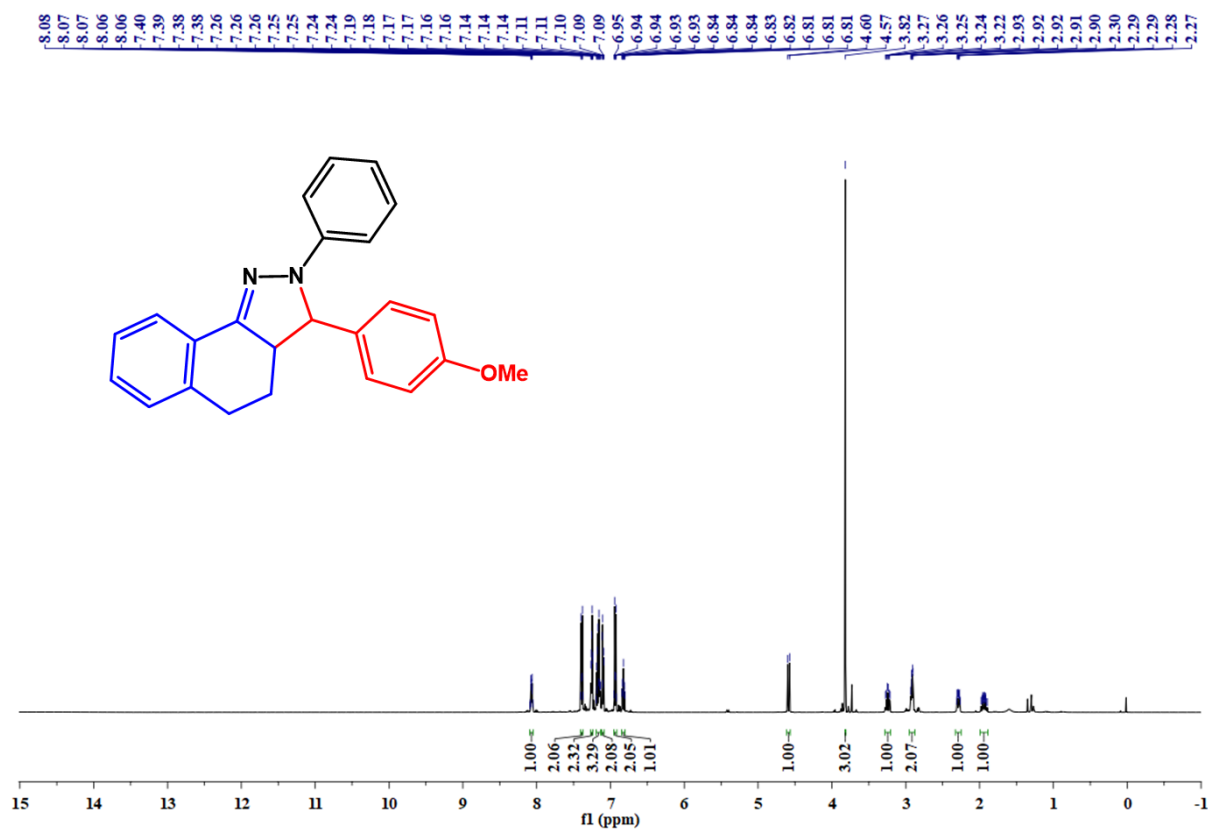


Figure S79: ^1H NMR spectrum of **4ac** in CDCl_3 at 500 MHz.

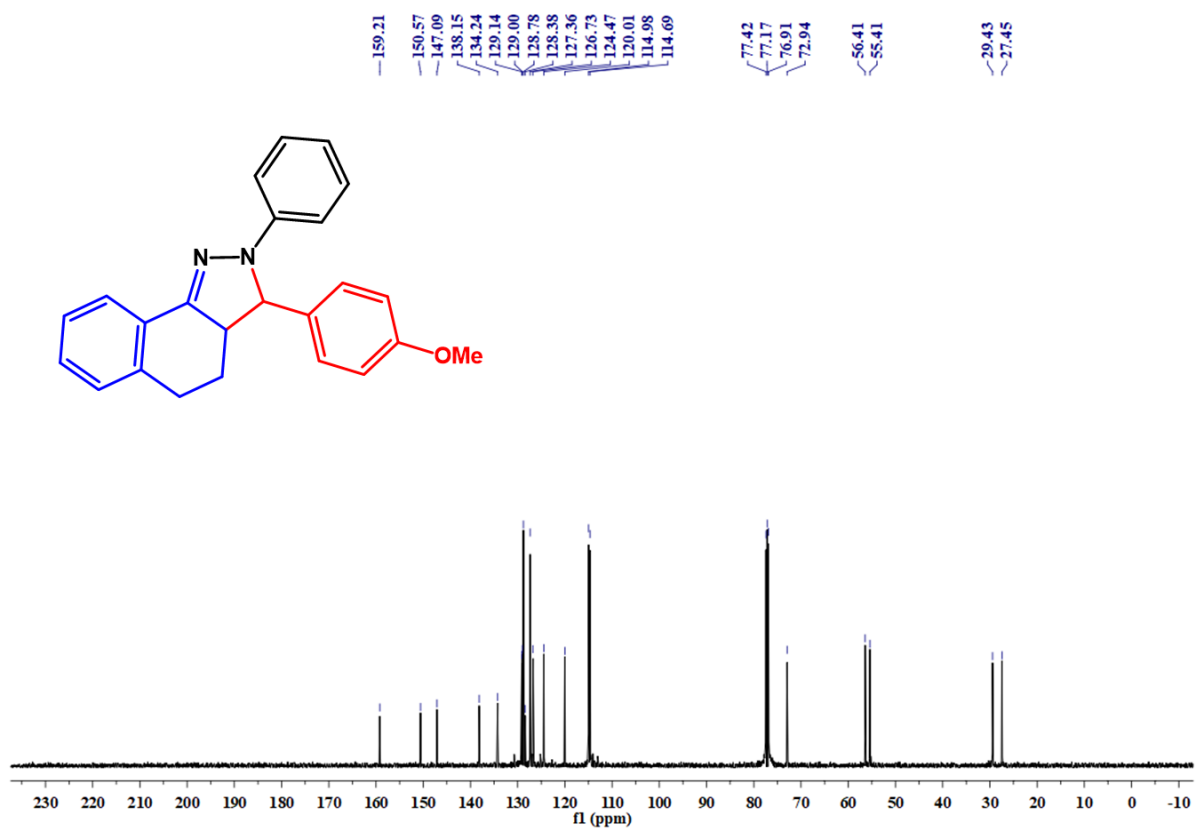


Figure S80: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **4ac** in CDCl_3 at 126 MHz.

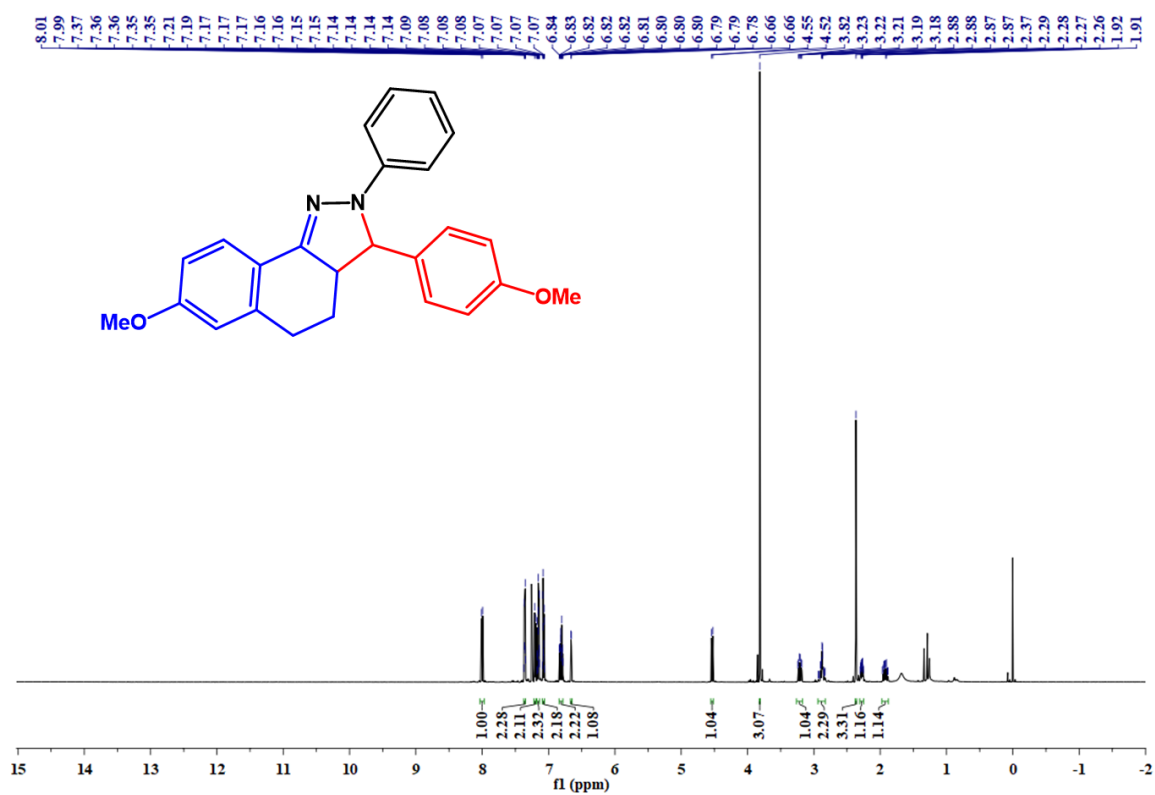
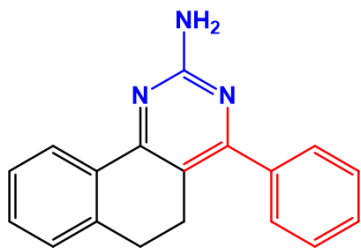
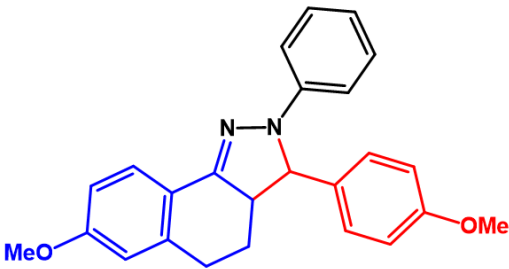


Figure S81: ^1H NMR spectrum of **4ad** in CDCl_3 at 500 MHz.



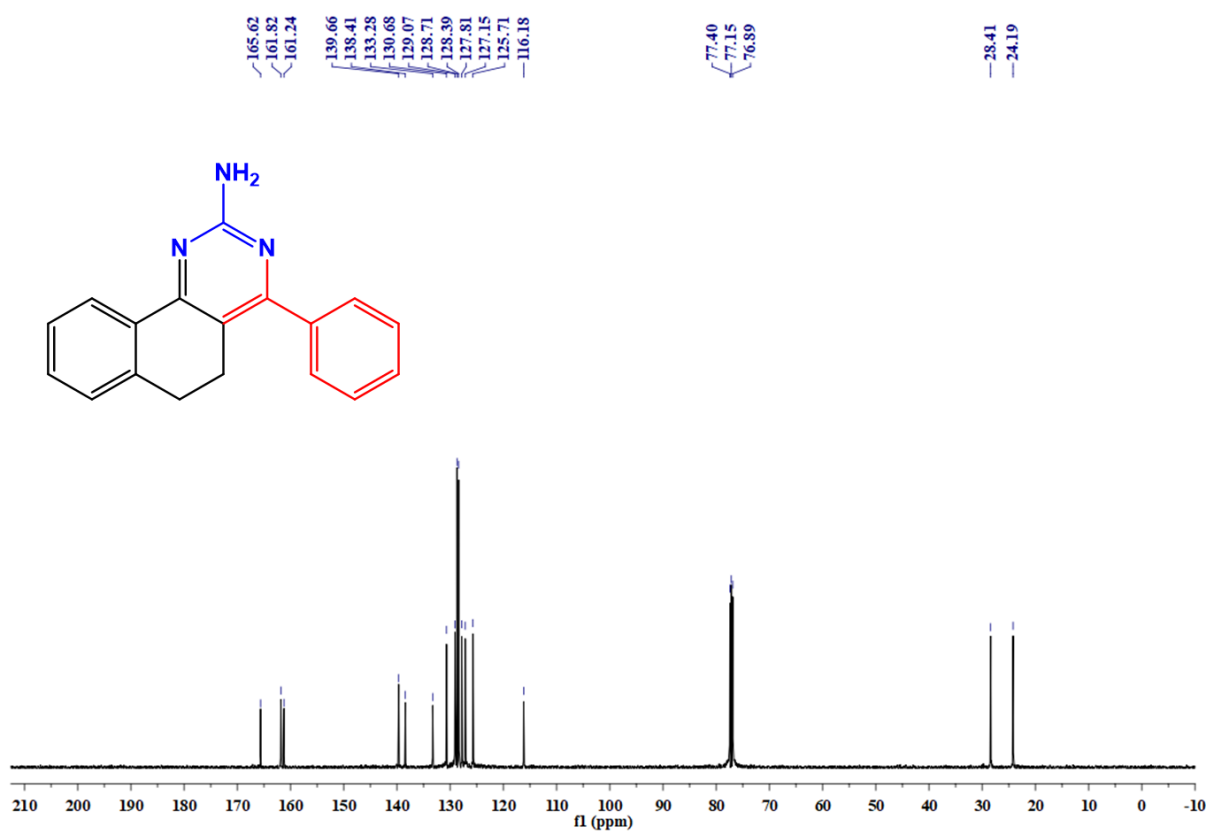


Figure S84: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8a** in CDCl_3 at 126 MHz.

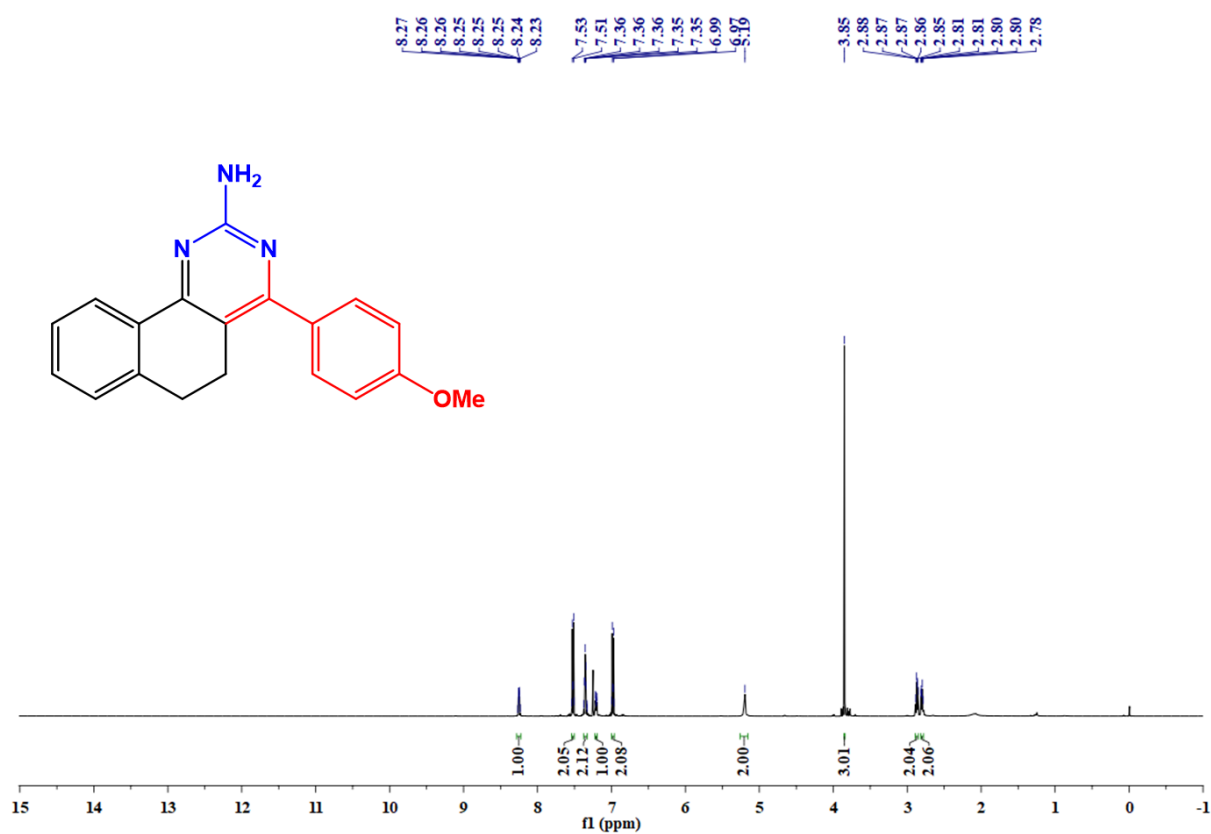


Figure S85: ^1H NMR spectrum of **8b** in CDCl_3 at 500 MHz.

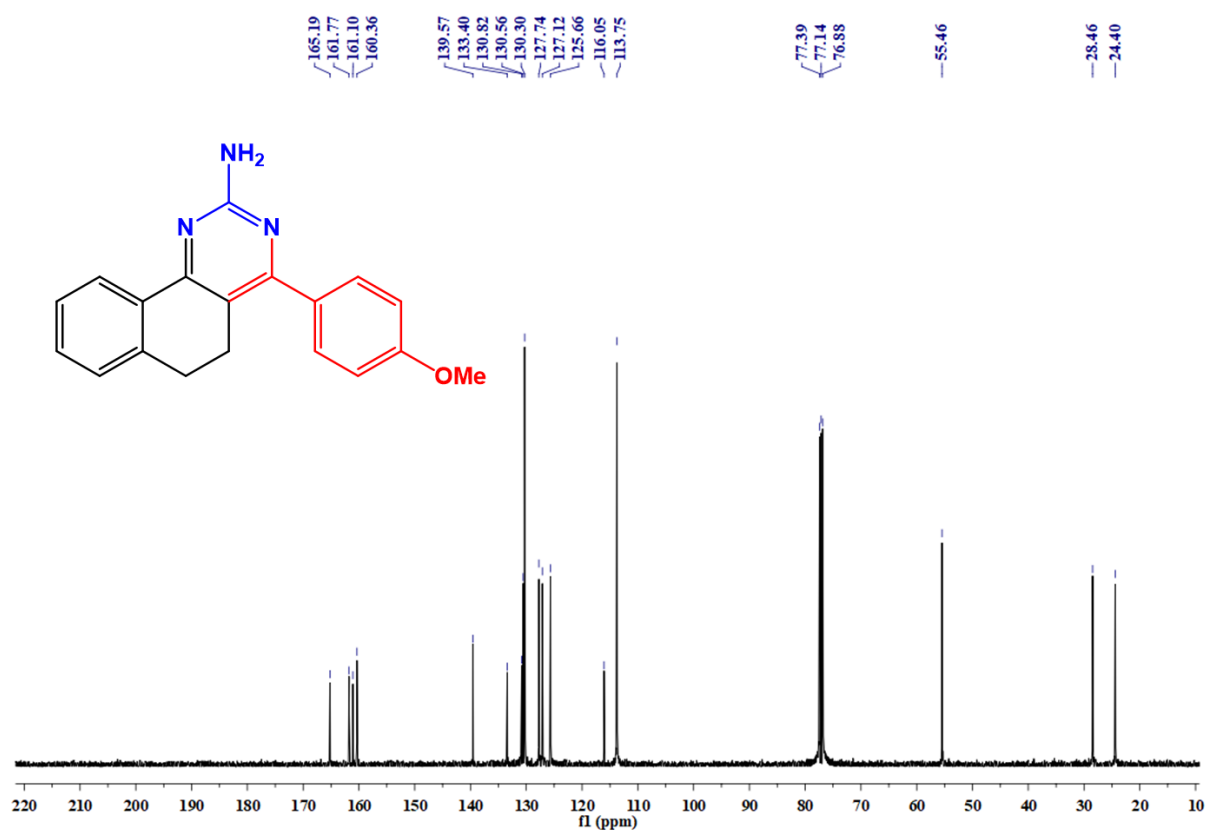


Figure S86: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8b** in CDCl_3 at 126 MHz.

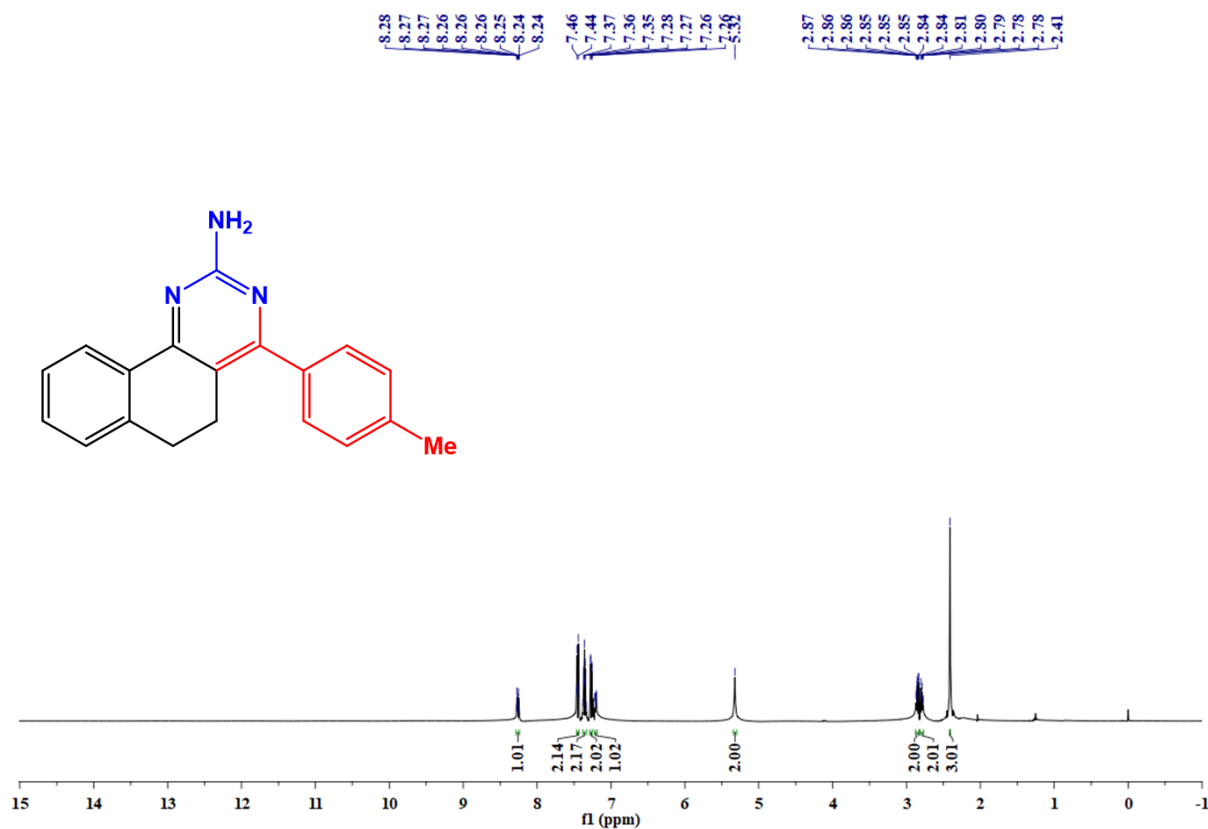


Figure S87: ^1H NMR spectrum of **8c** in CDCl_3 at 500 MHz.

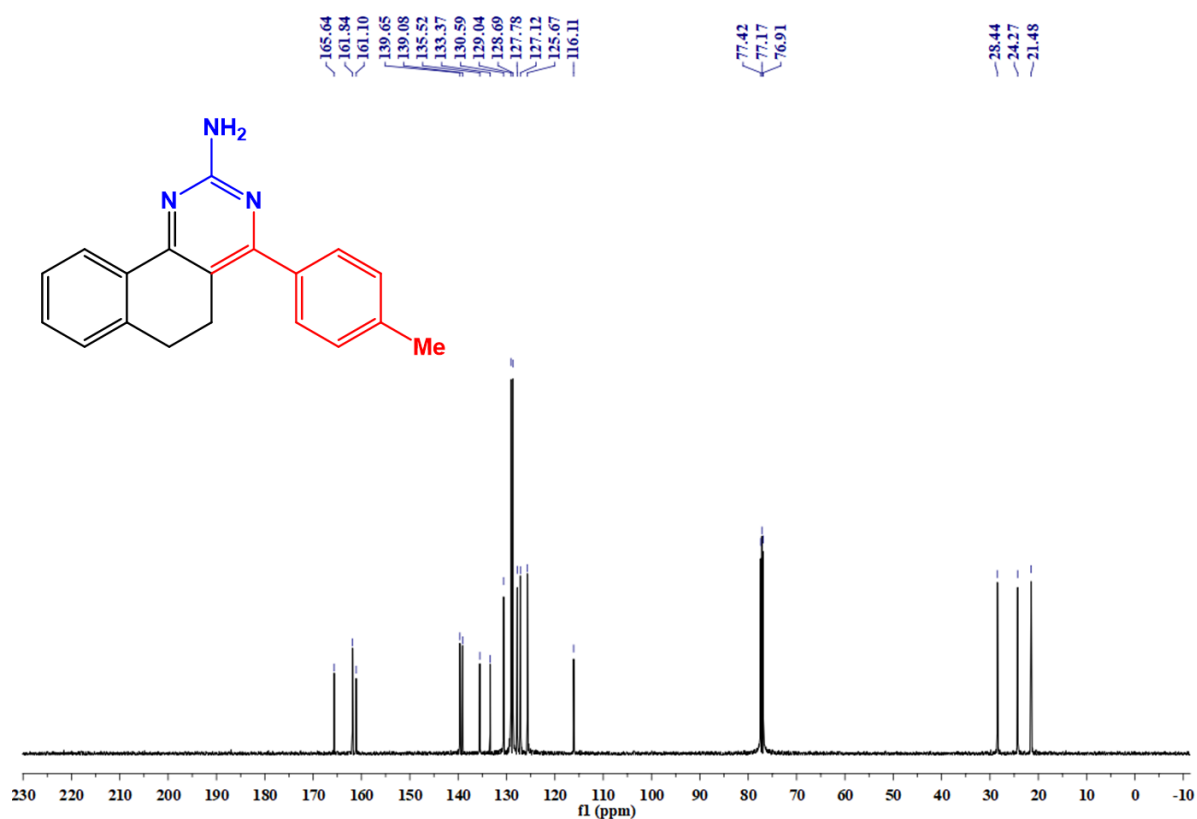


Figure S88: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8c** in CDCl_3 at 126 MHz.

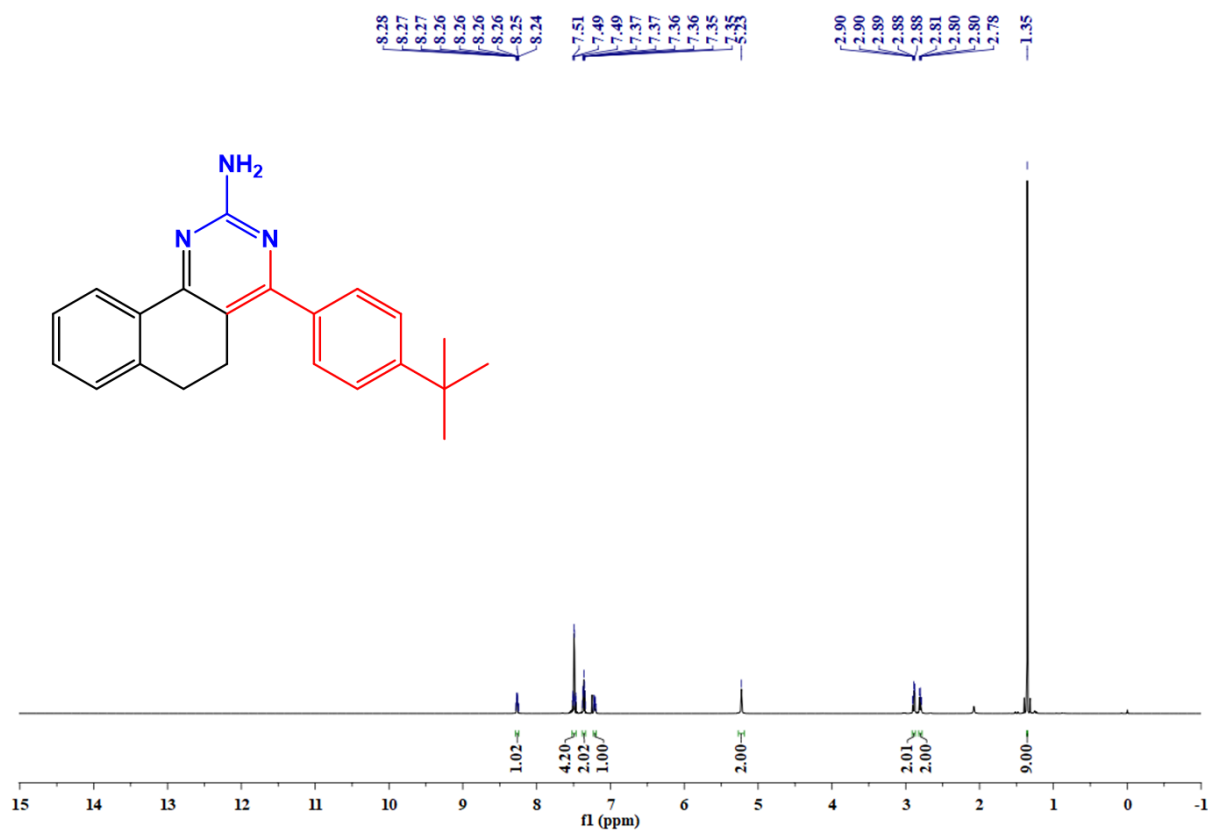


Figure S89: ^1H NMR spectrum of **8d** in CDCl_3 at 500 MHz.

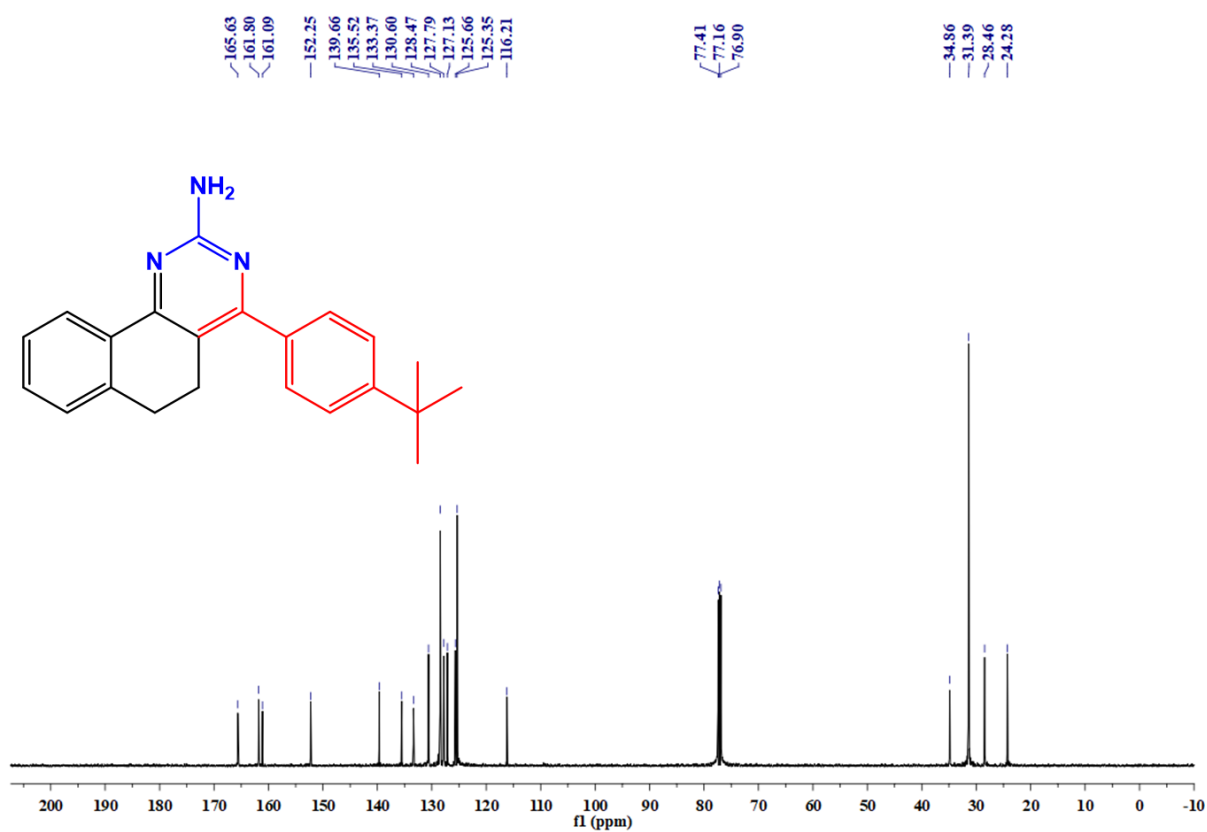


Figure S90: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8d** in CDCl_3 at 126 MHz.

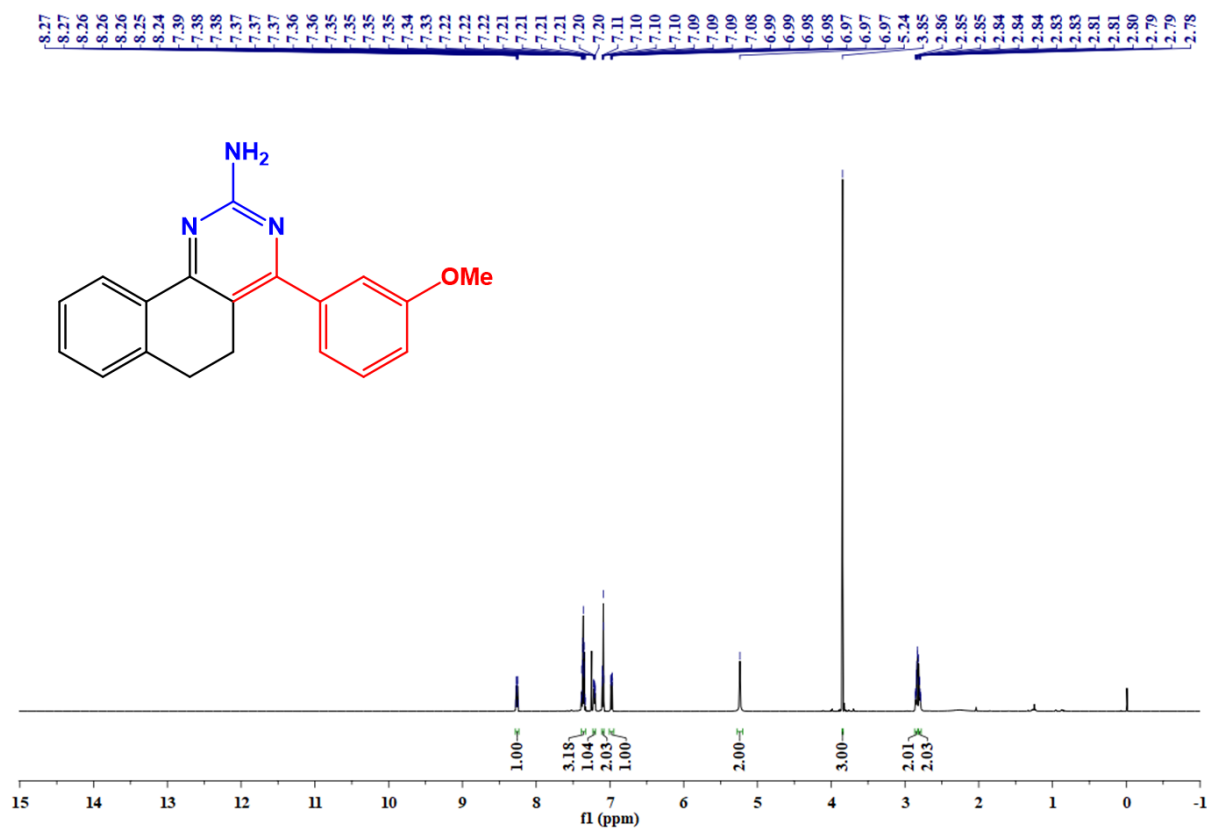


Figure S91: ^1H NMR spectrum of **8e** in CDCl_3 at 500 MHz.

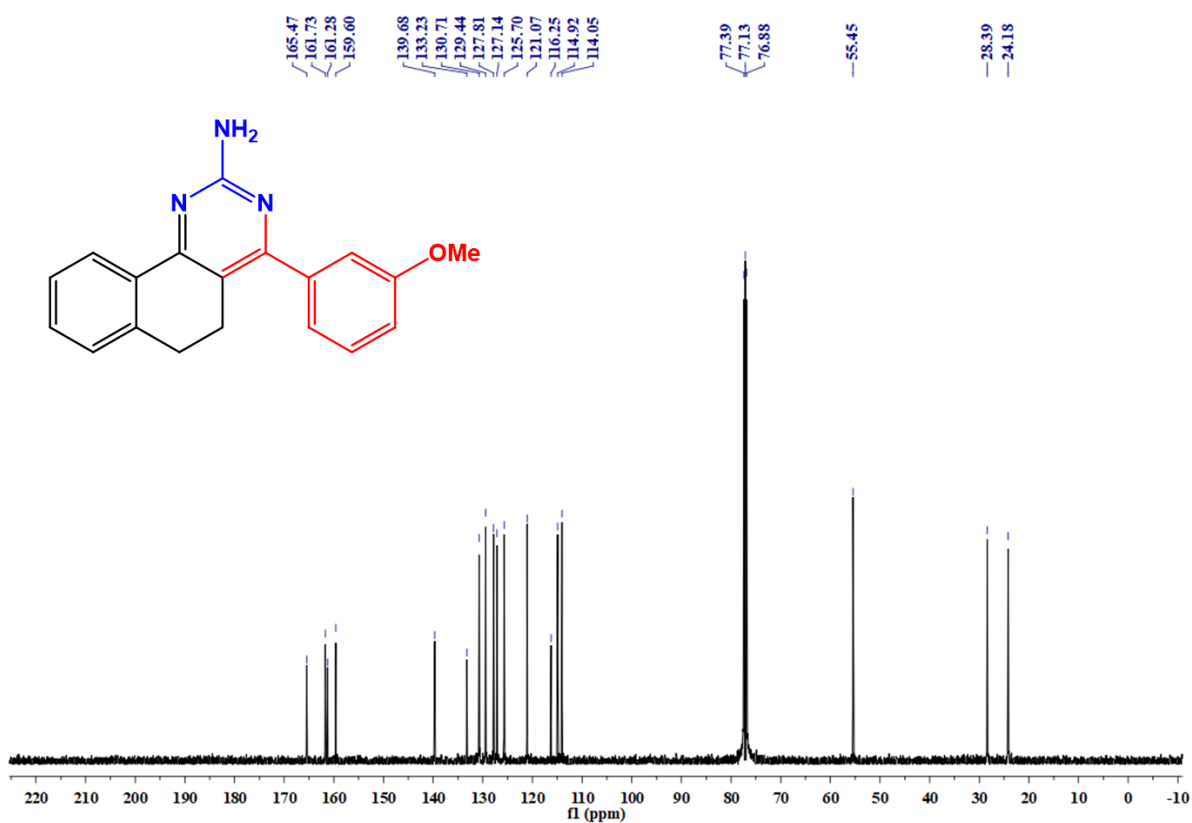


Figure S92: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8e** in CDCl_3 at 126 MHz.

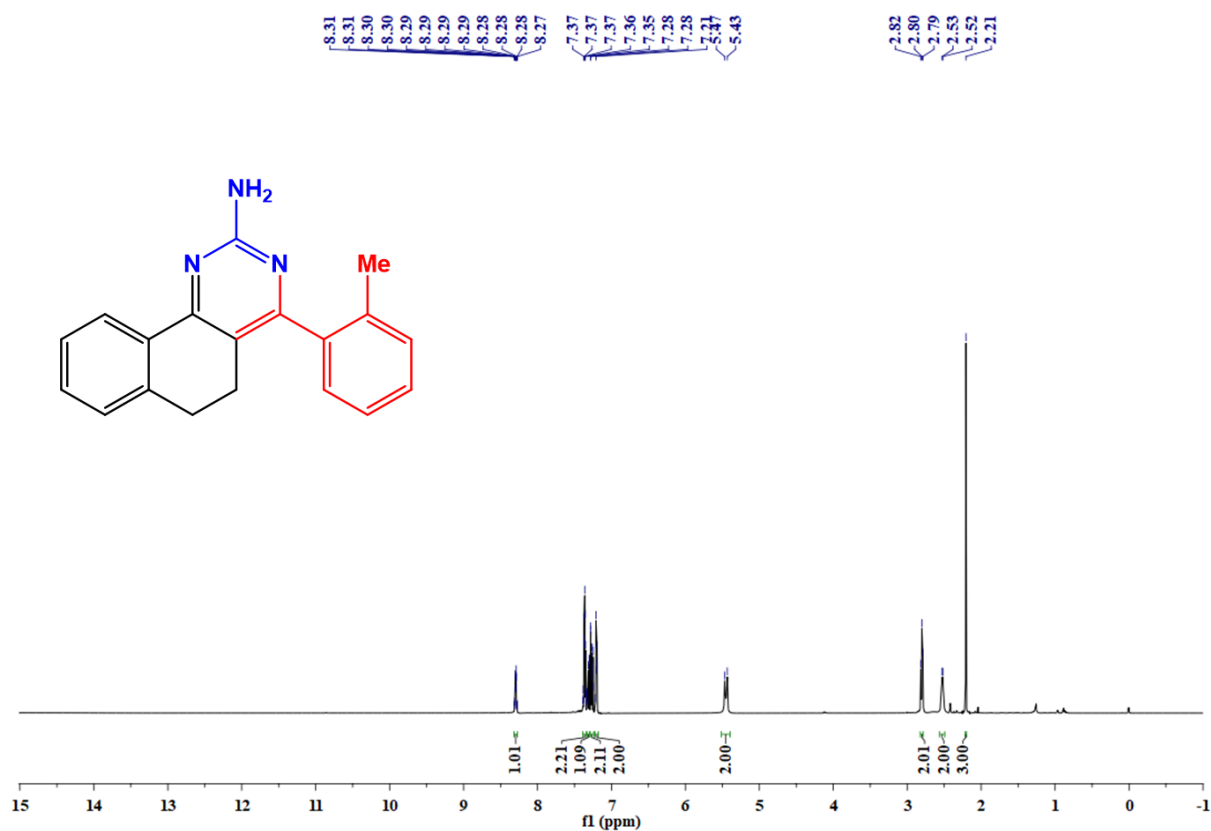


Figure S93: ^1H NMR spectrum of **8f** in CDCl_3 at 500 MHz.

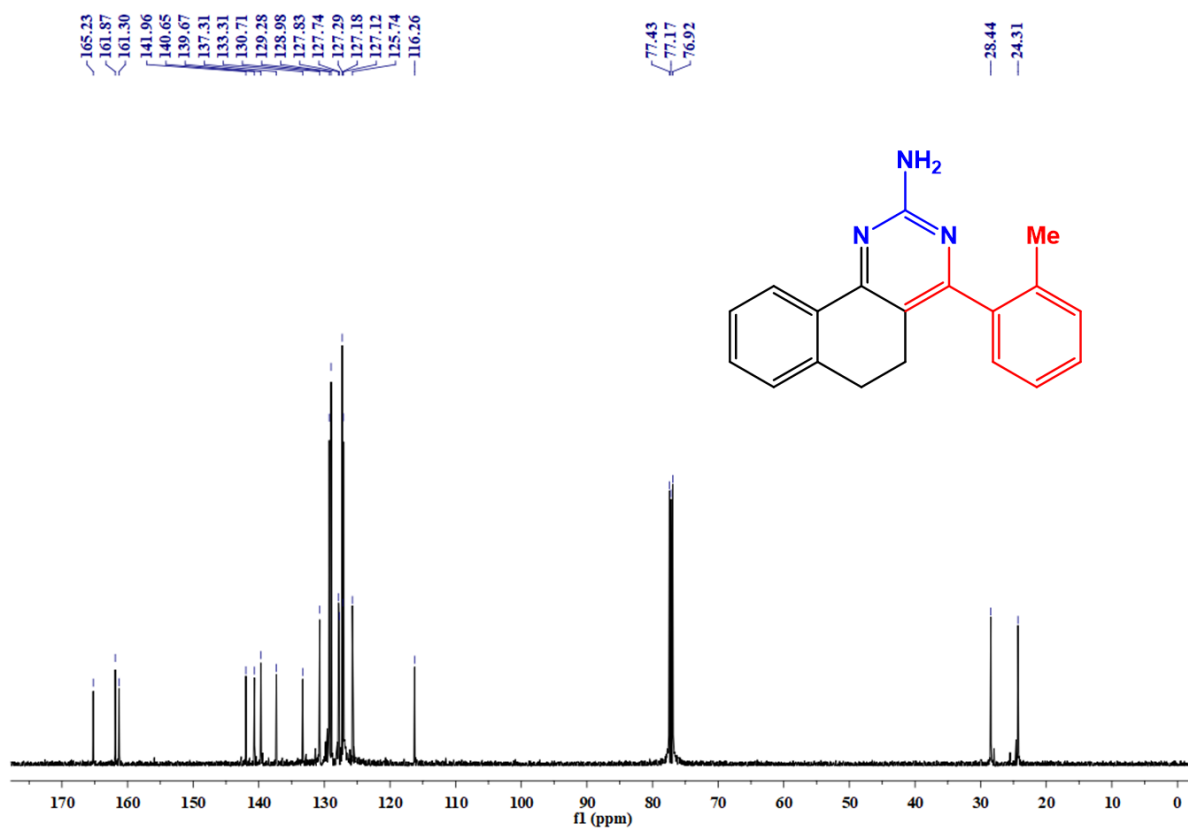


Figure S94: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8f** in CDCl_3 at 126 MHz.

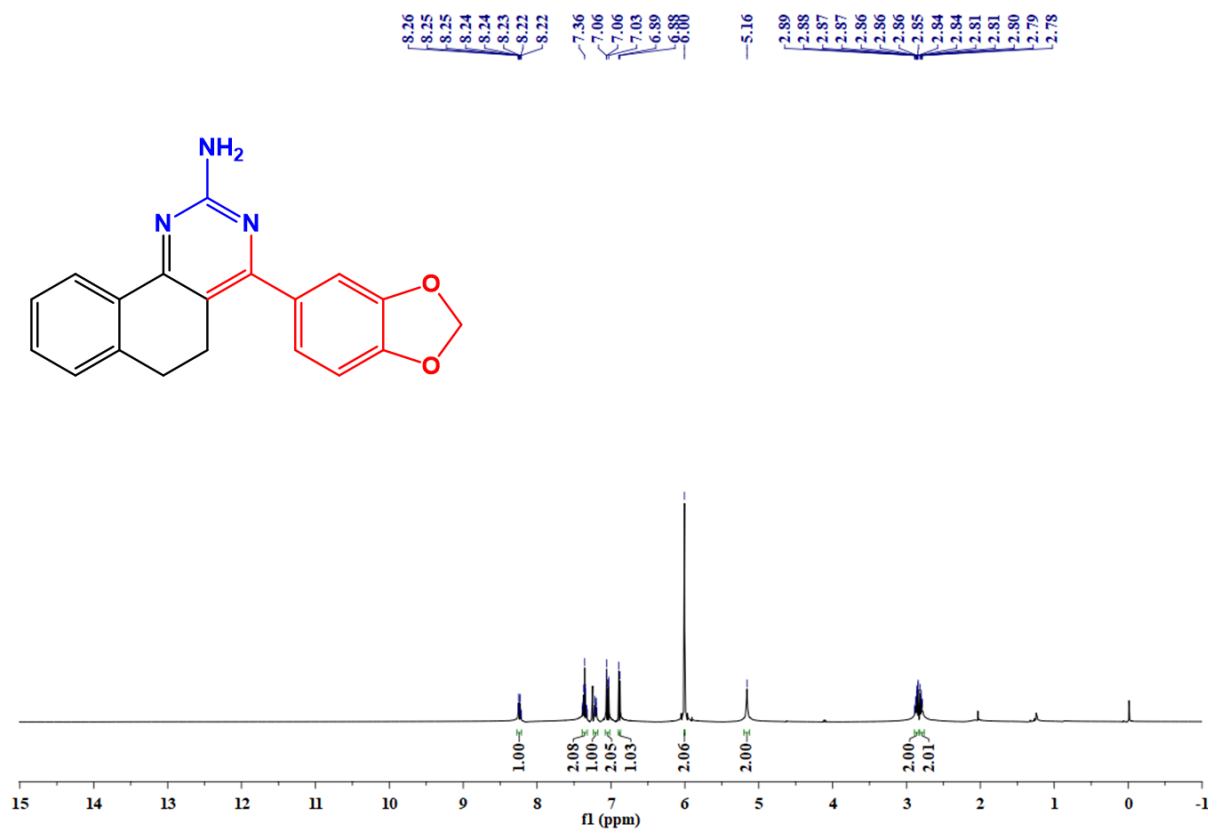


Figure S95: ^1H NMR spectrum of **8g** in CDCl_3 at 500 MHz.

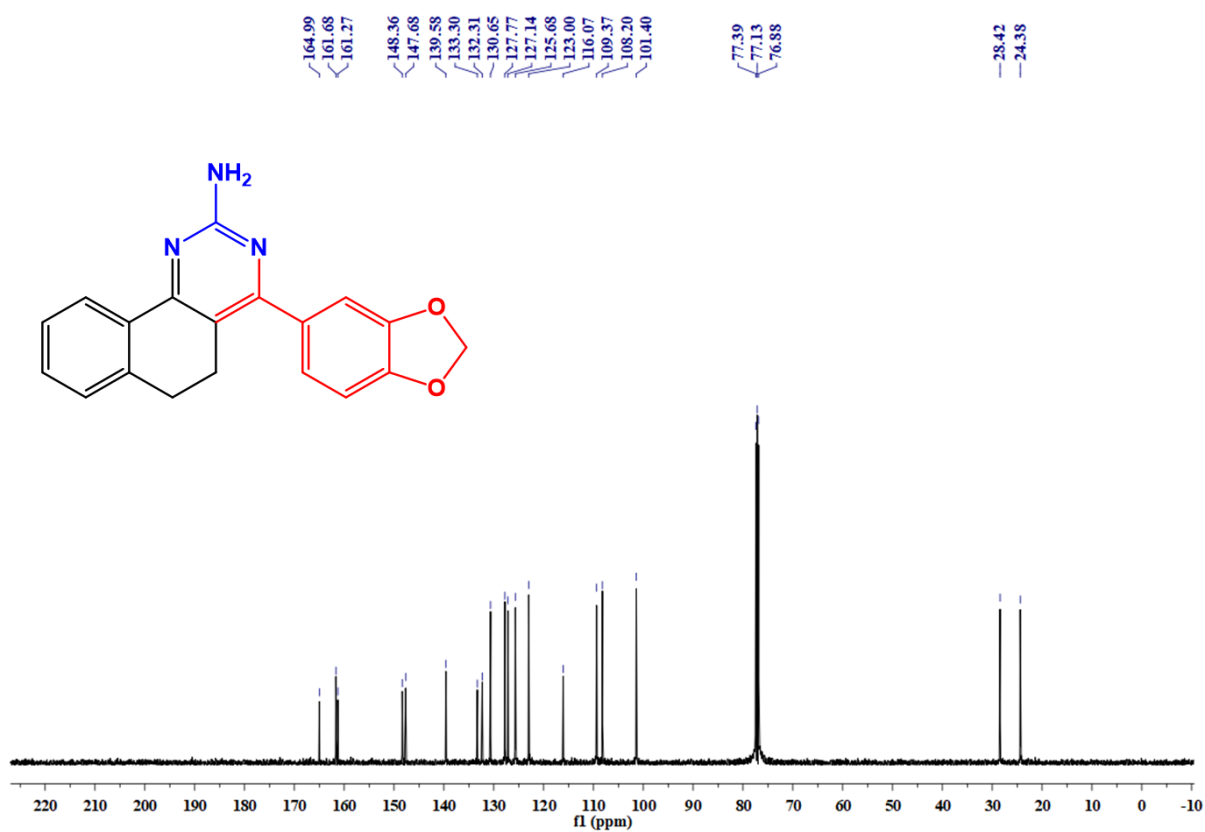


Figure S96: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8g** in CDCl_3 at 126 MHz.

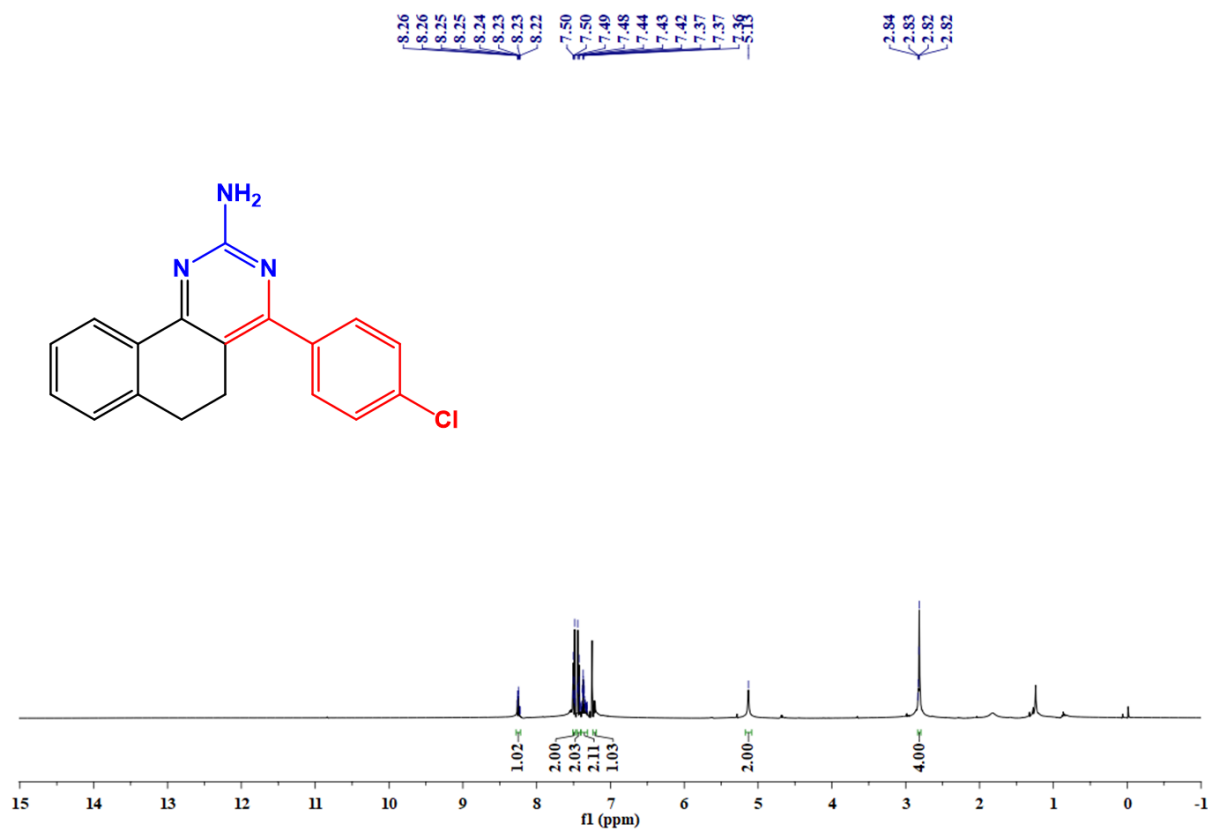


Figure S97: ^1H NMR spectrum of **8h** in CDCl_3 at 500 MHz.

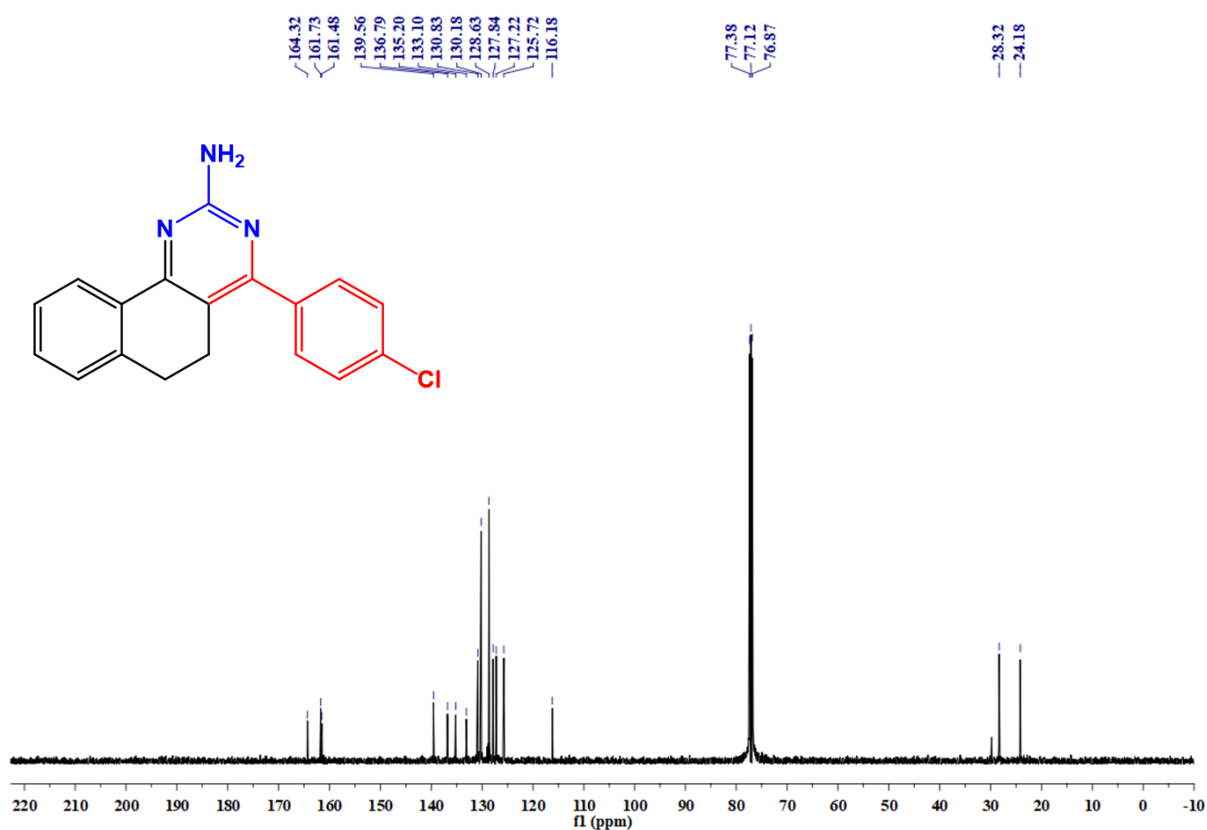


Figure S98: ¹³C{¹H} NMR spectrum of **8h** in CDCl₃ at 126 MHz.

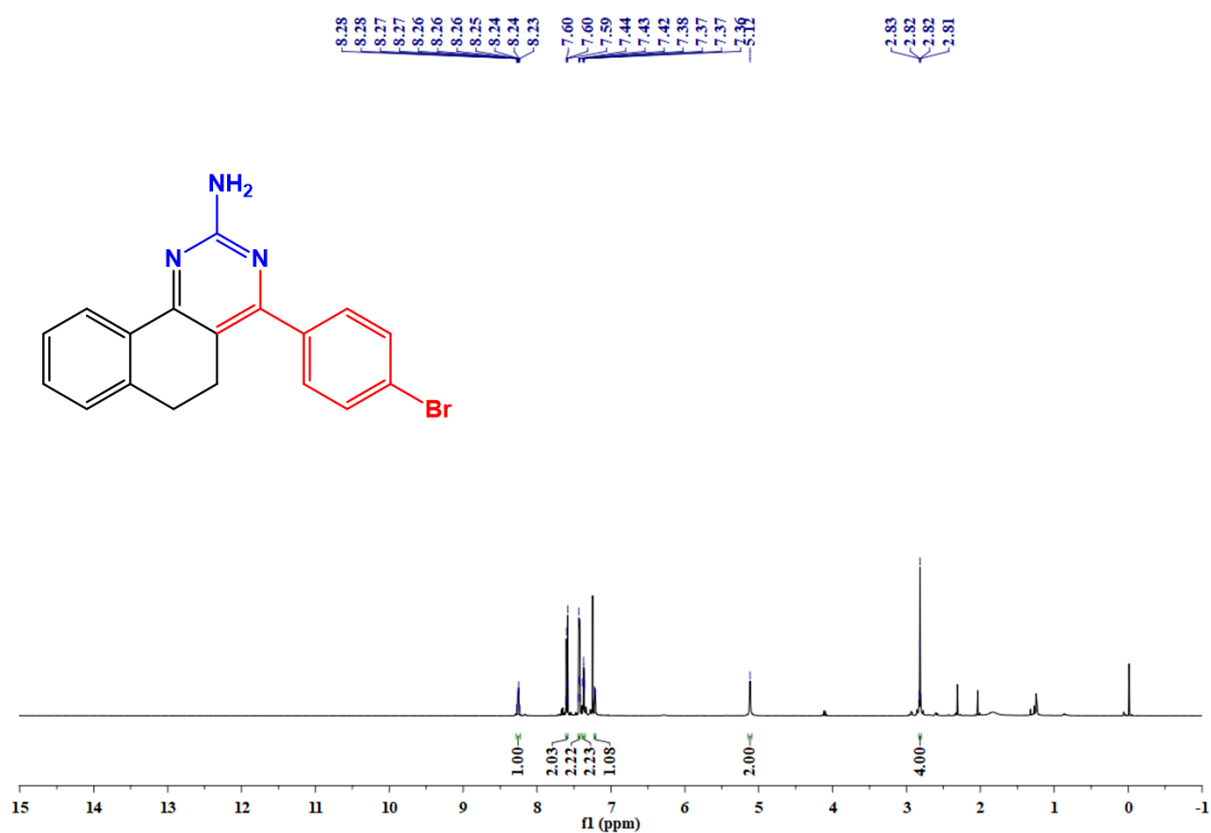


Figure S99: ¹H NMR spectrum of **8i** in CDCl₃ at 500 MHz.

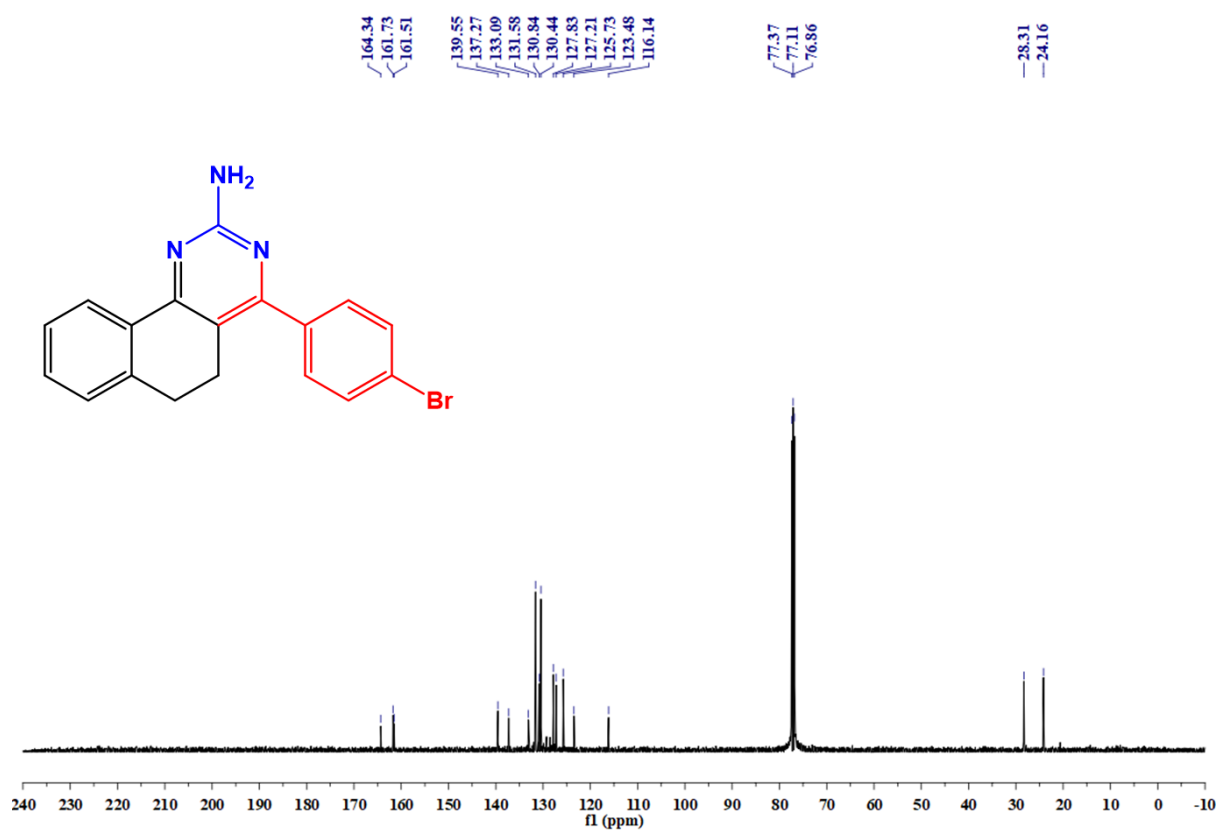


Figure S100: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8i** in CDCl_3 at 126 MHz.

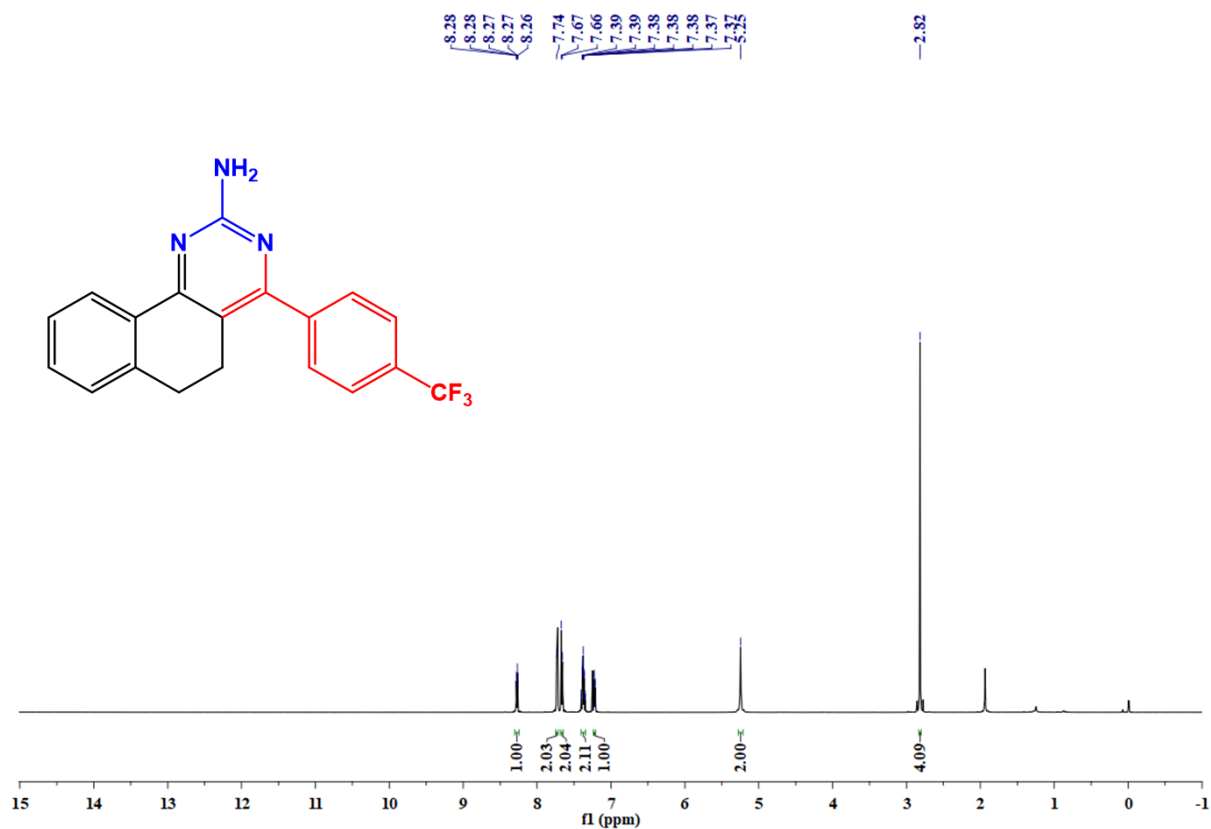


Figure S101: ^1H NMR spectrum of **8j** in CDCl_3 at 500 MHz.

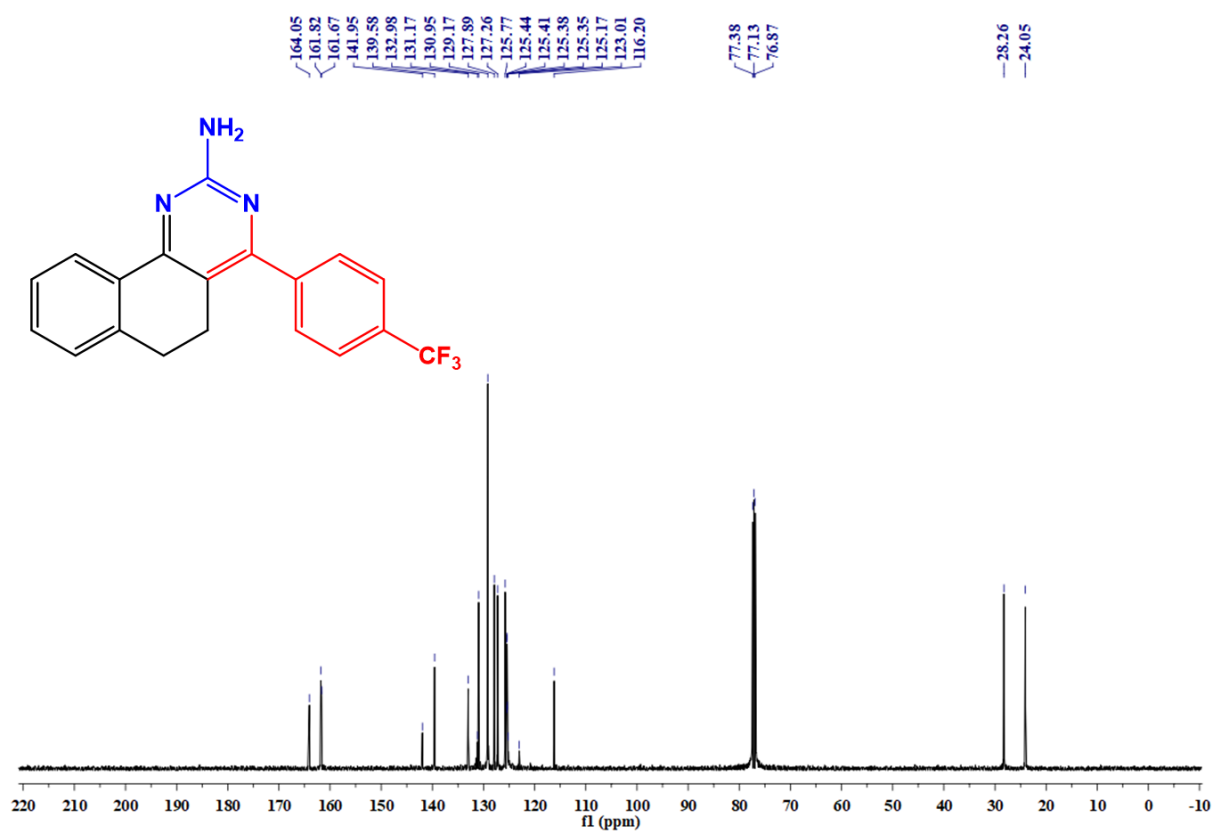


Figure S102: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8j** in CDCl_3 at 126 MHz.

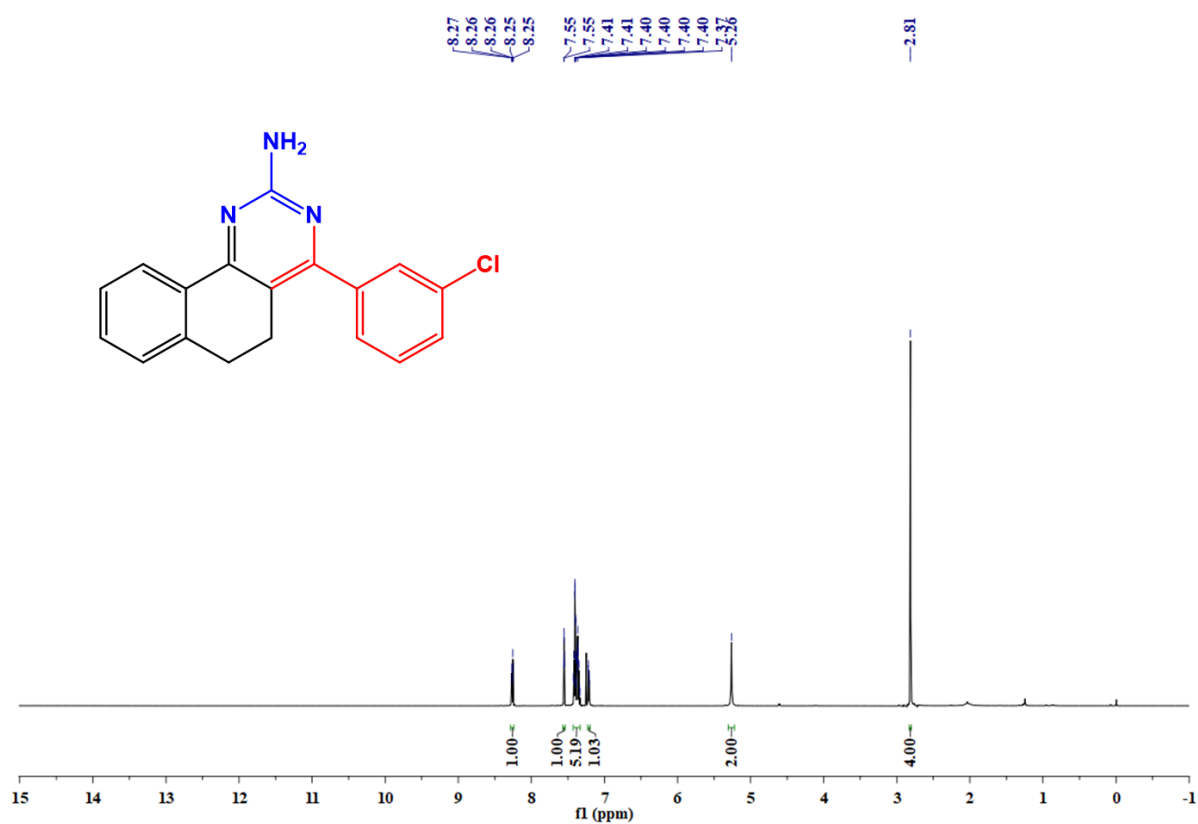


Figure S103: ^1H NMR spectrum of **8k** in CDCl_3 at 500 MHz.

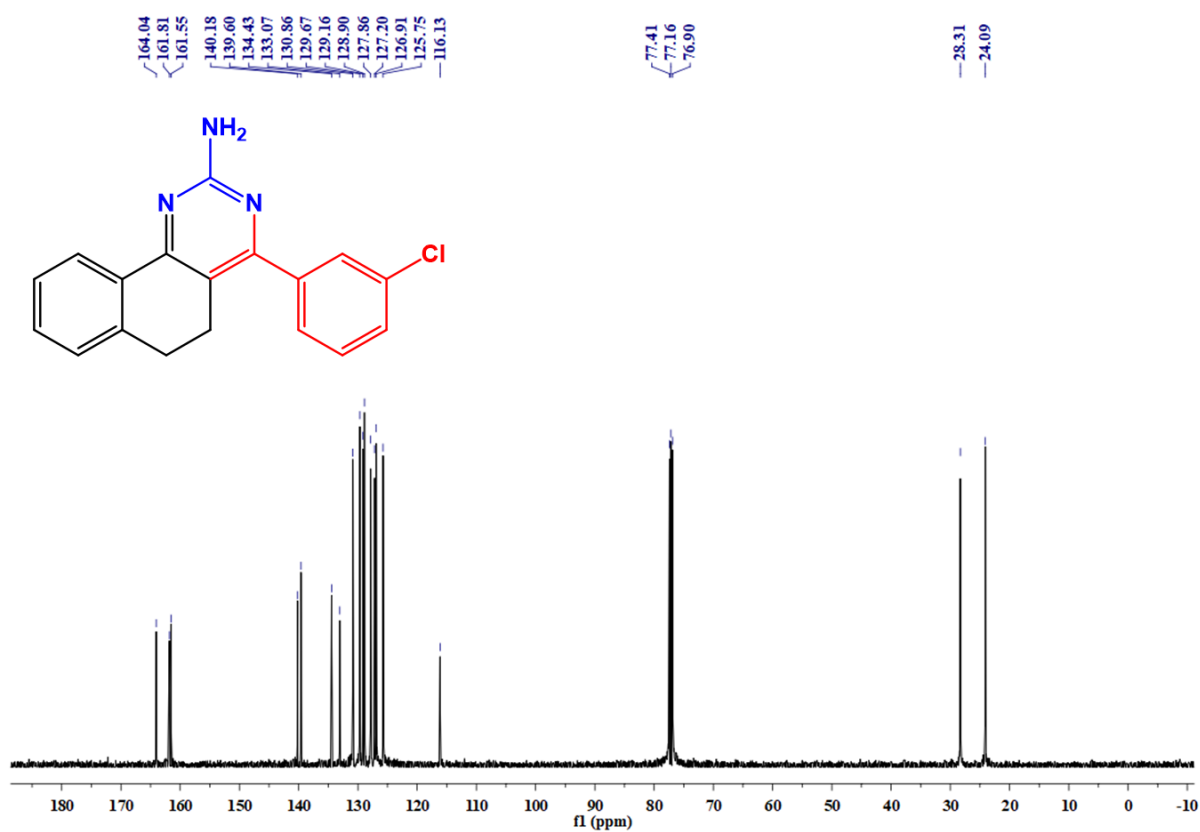


Figure S104: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8k** in CDCl_3 at 126 MHz.

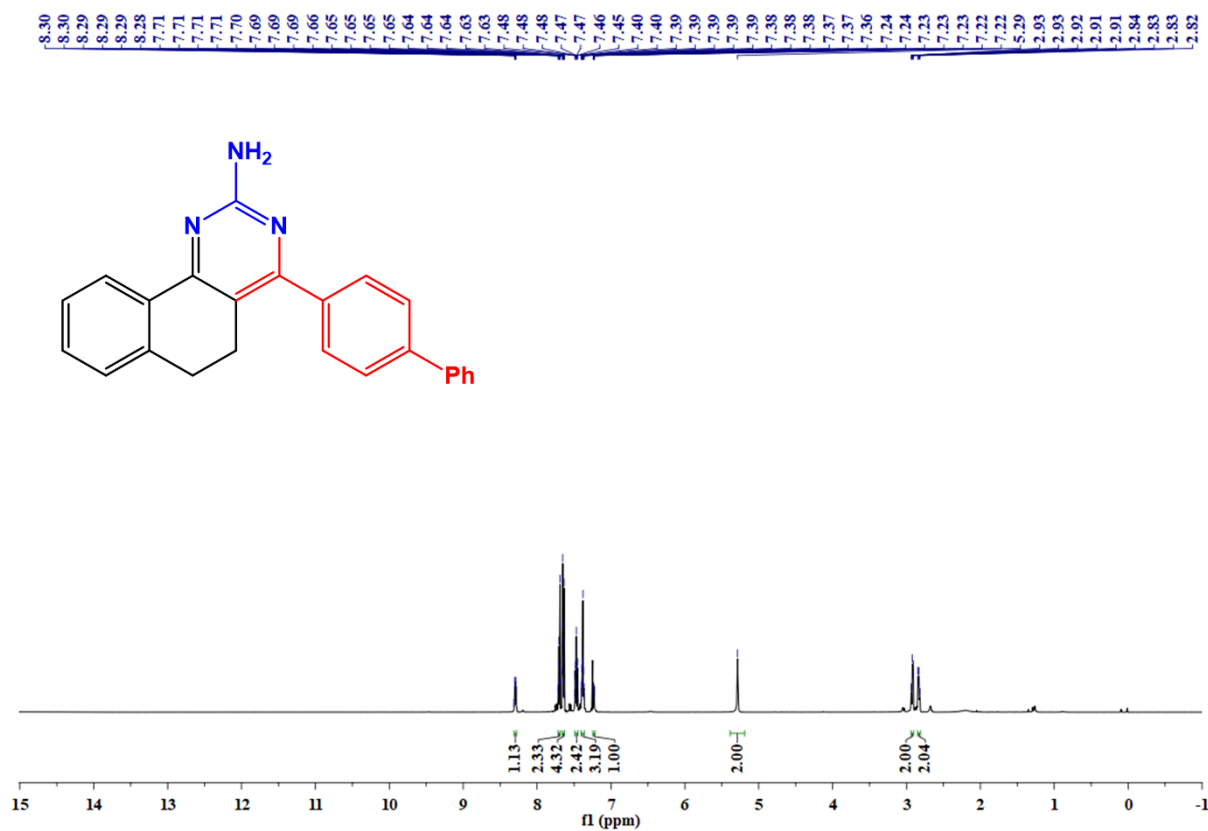


Figure S105: ^1H NMR spectrum of **8l** in CDCl_3 at 500 MHz.

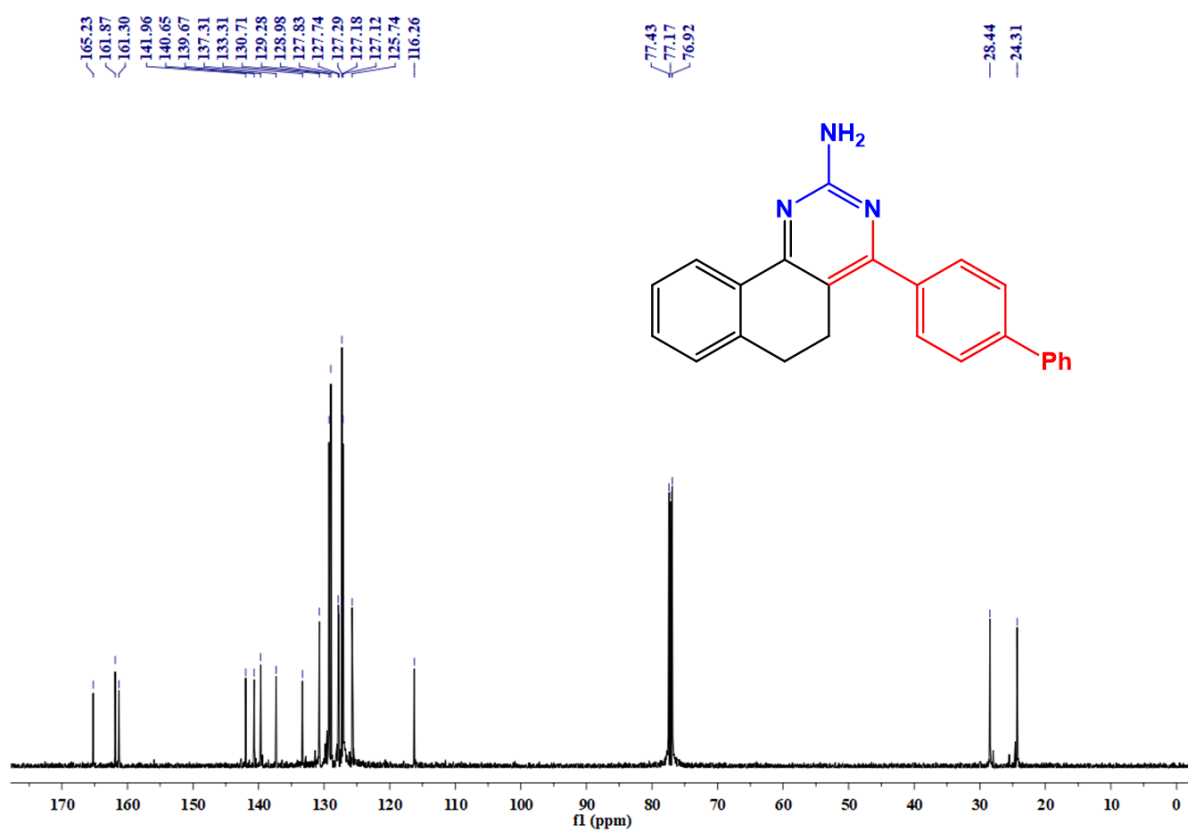


Figure S106: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8l** in CDCl_3 at 126 MHz.

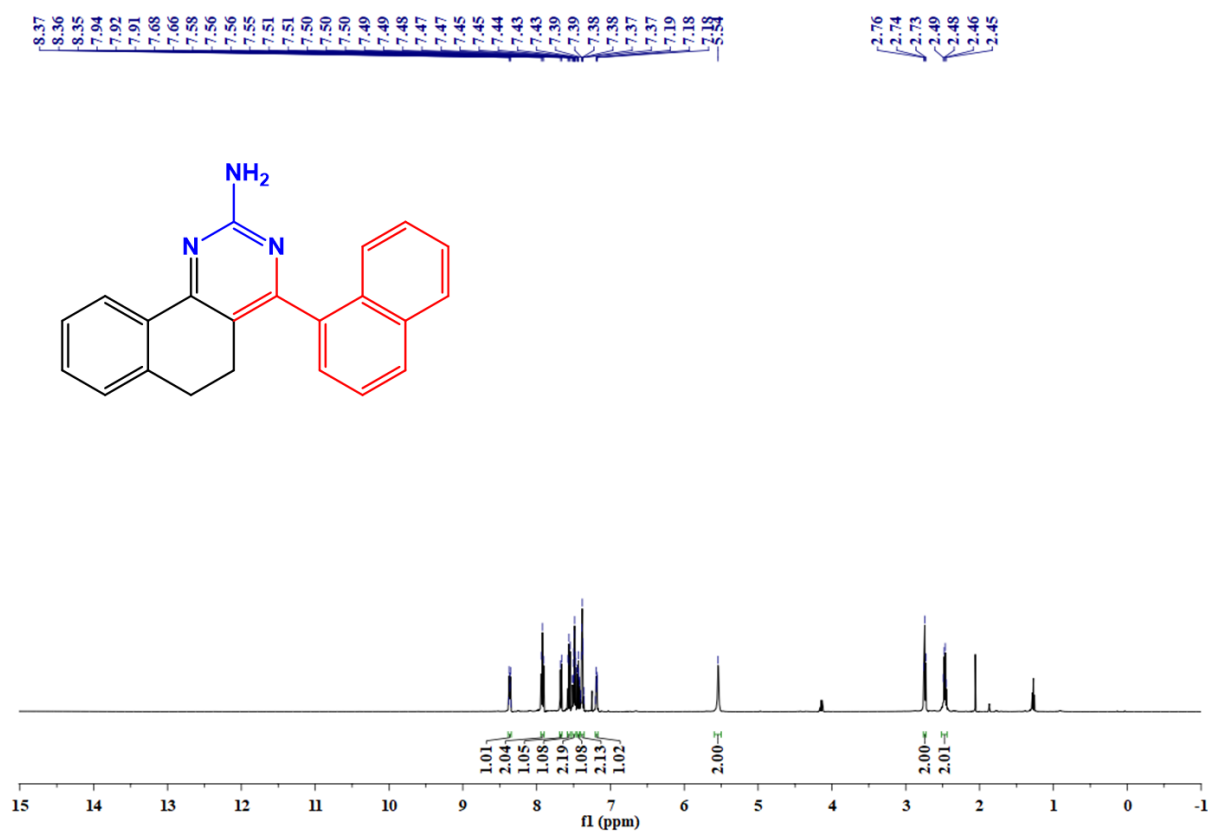


Figure S107: ^1H NMR spectrum of **8m** in CDCl_3 at 500 MHz.

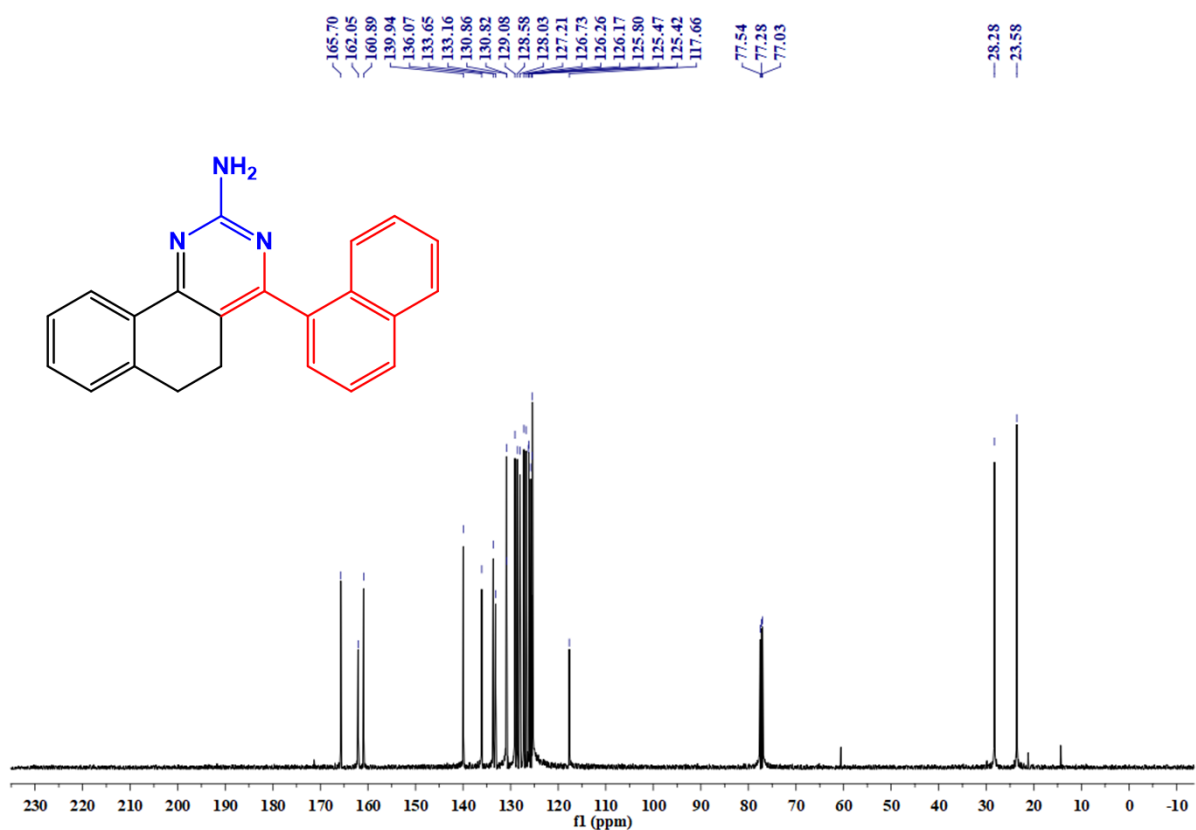


Figure S108: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8m** in CDCl_3 at 126 MHz.

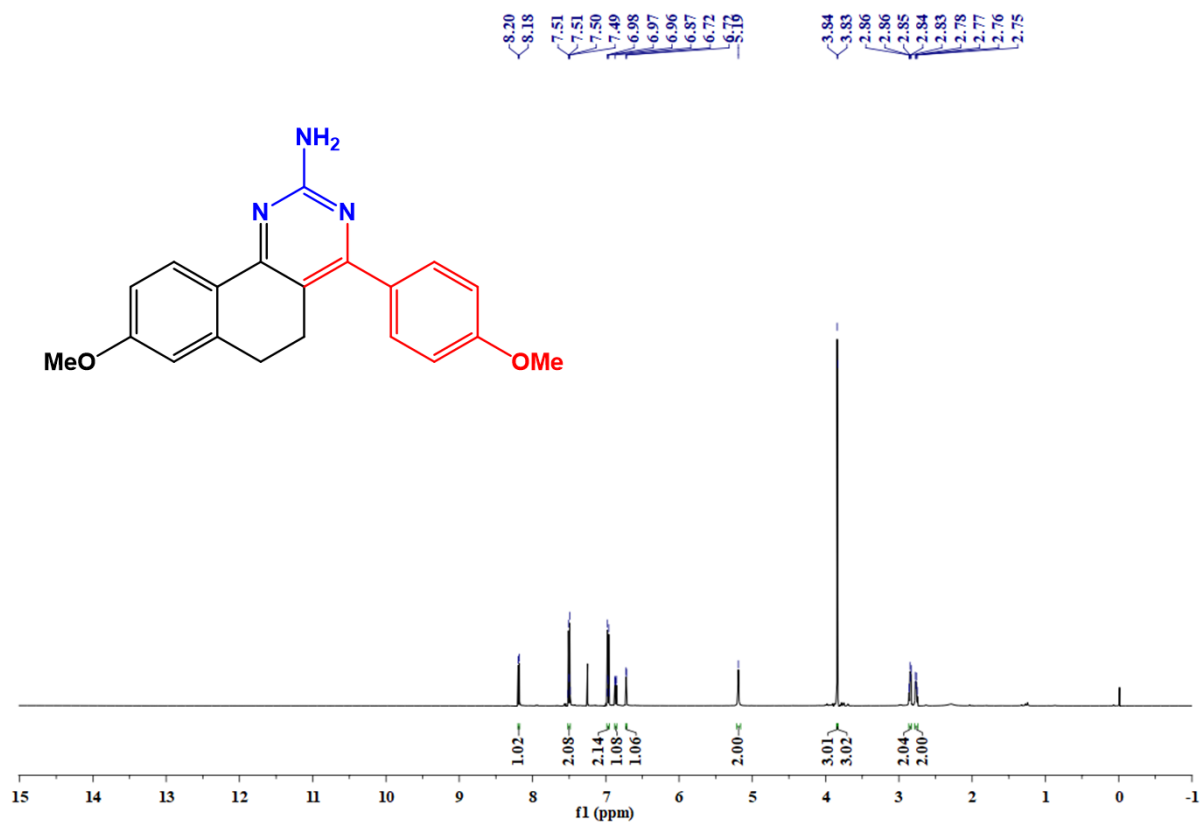


Figure S109: ^1H NMR spectrum of **8n** in CDCl_3 at 500 MHz.

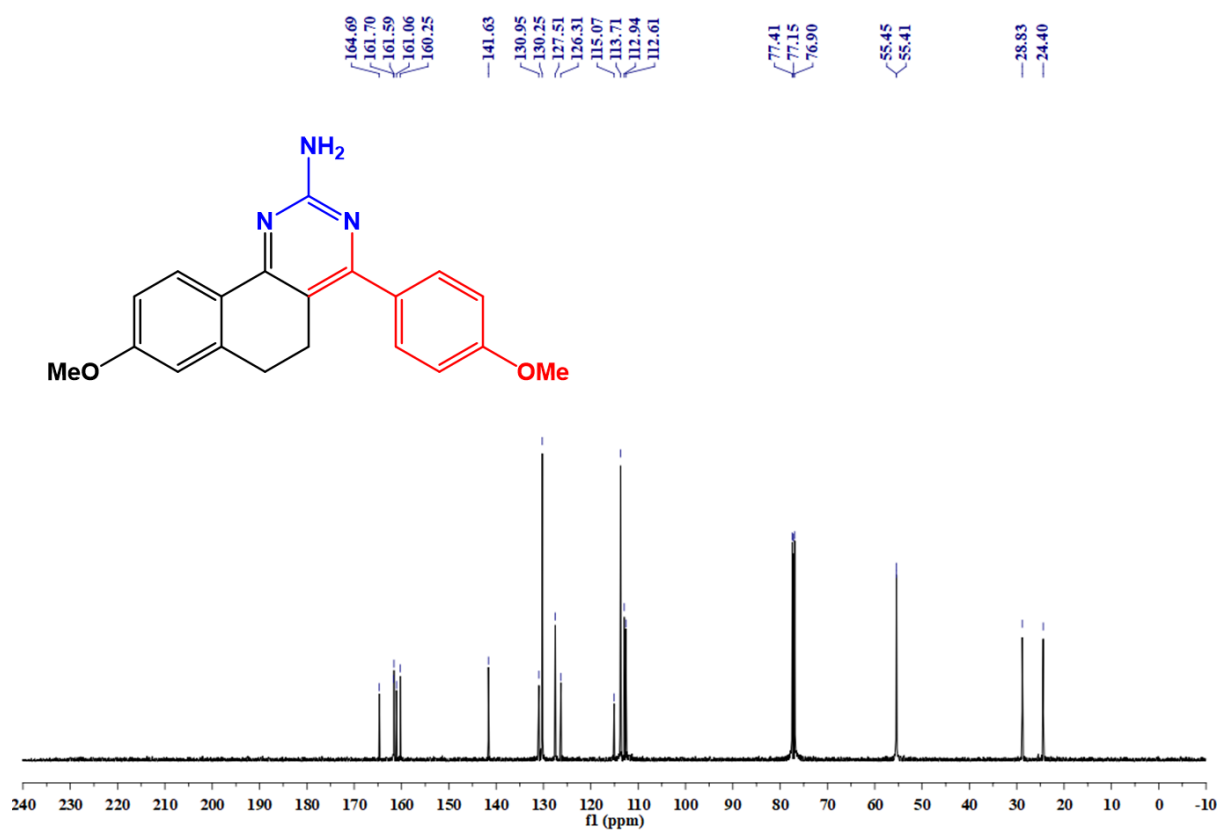


Figure S110: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8n** in CDCl_3 at 126 MHz.

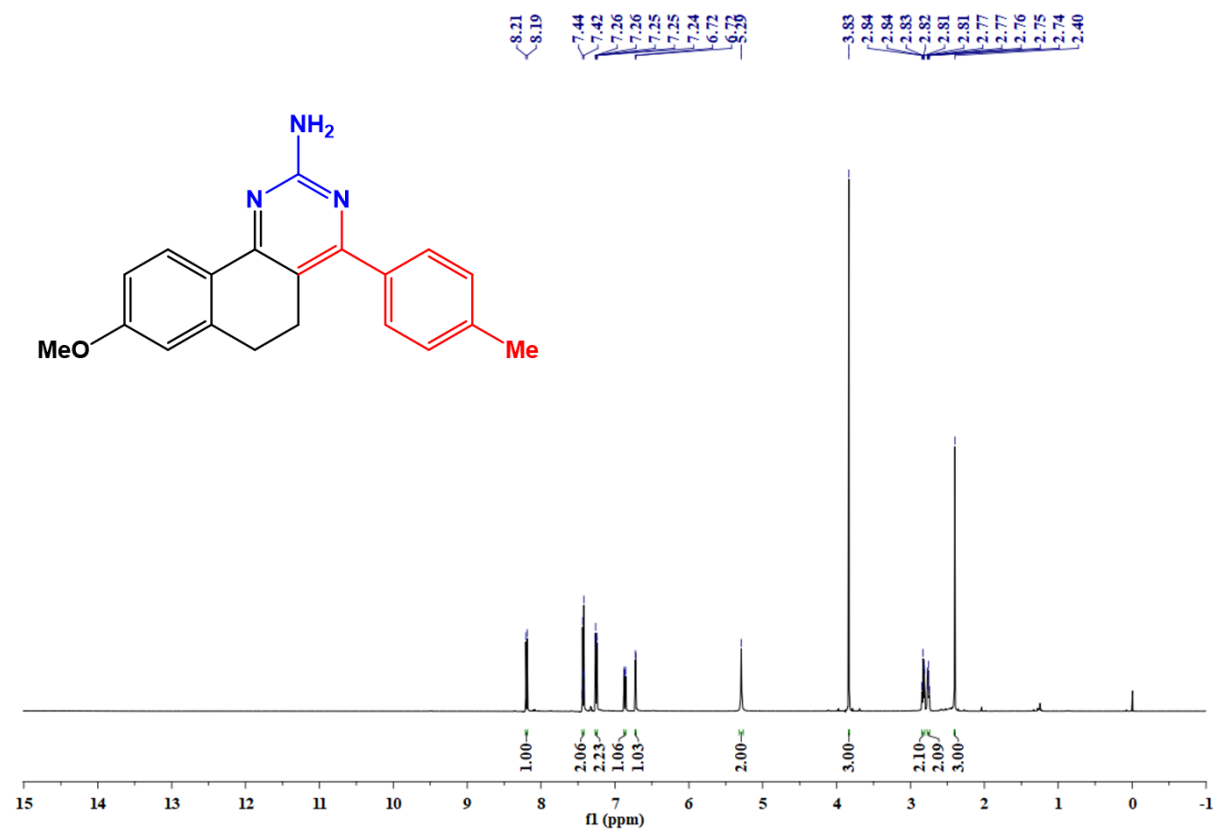


Figure S111: ^1H NMR spectrum of **8o** in CDCl_3 at 500 MHz.

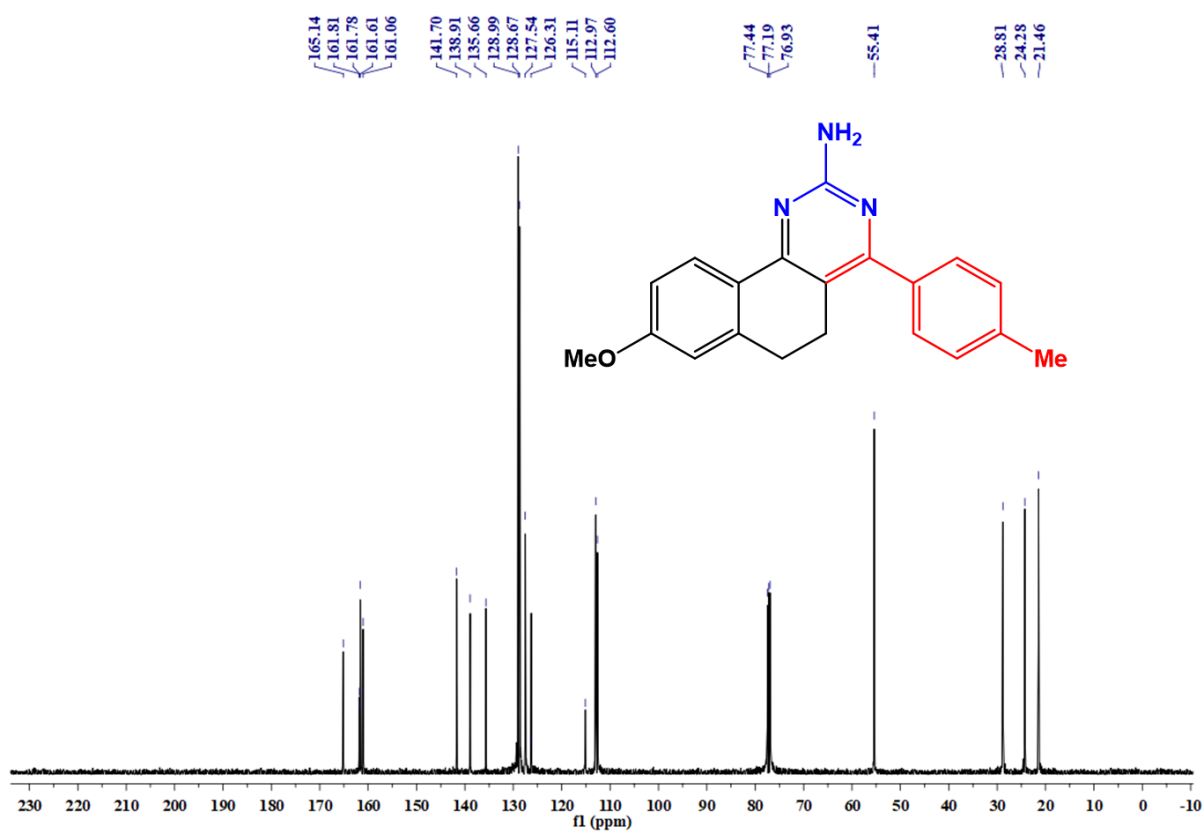


Figure S112: ¹³C{¹H} NMR spectrum of **8o** in CDCl₃ at 126 MHz.

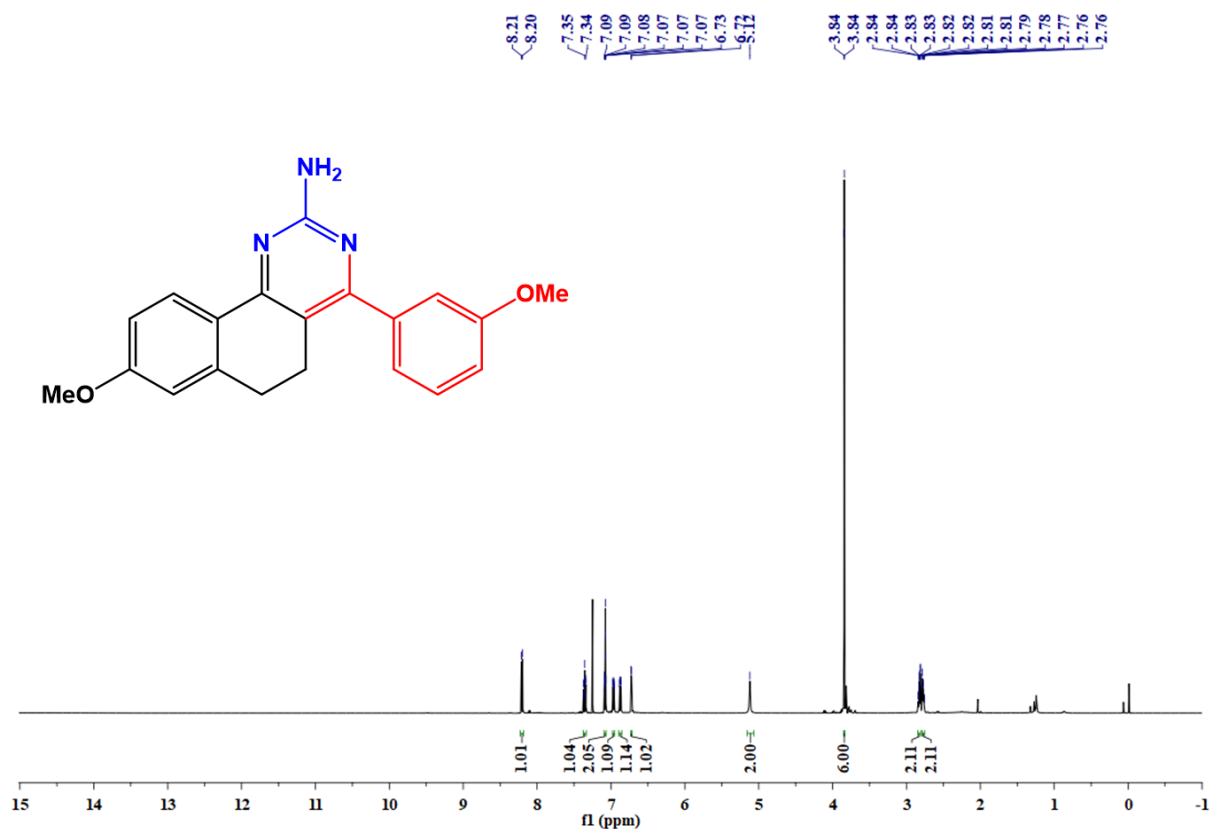


Figure S113: ¹H NMR spectrum of **8p** in CDCl₃ at 500 MHz.

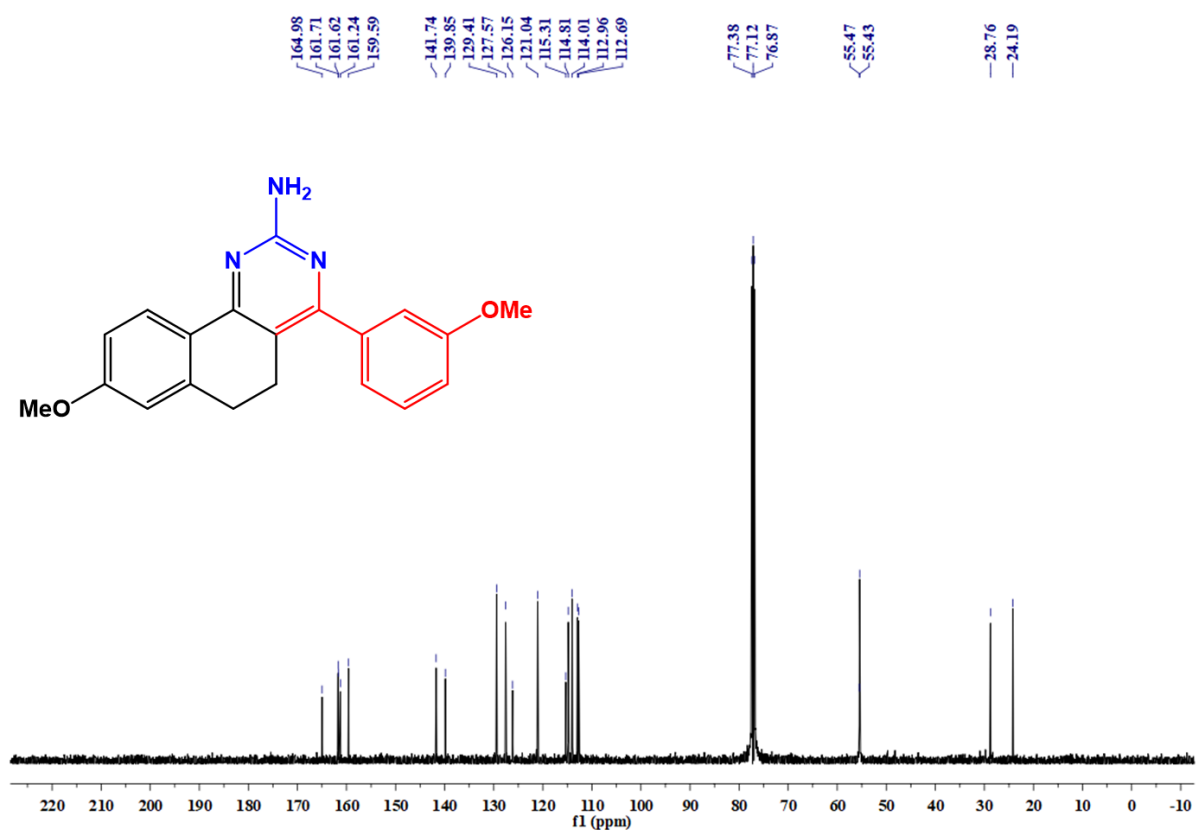


Figure S114: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8p** in CDCl_3 at 126 MHz.

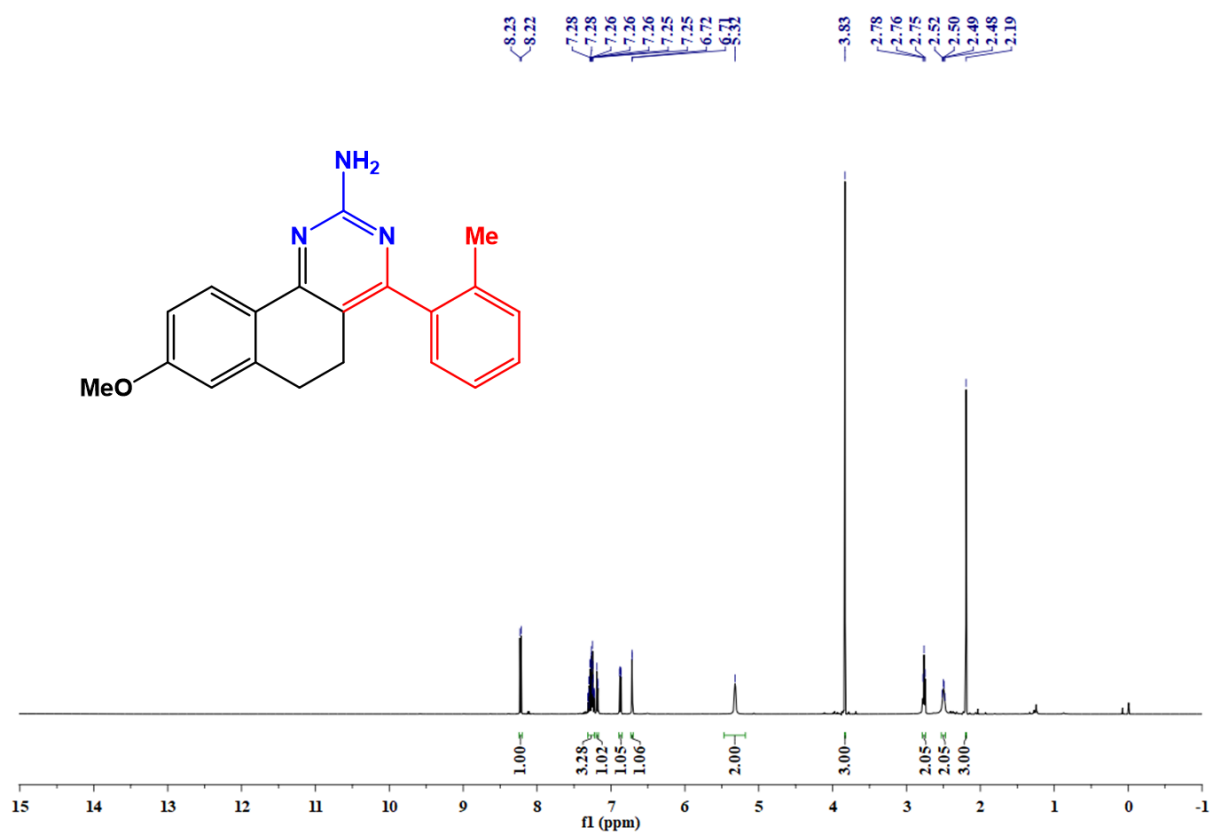


Figure S115: ^1H NMR spectrum of **8q** in CDCl_3 at 500 MHz.

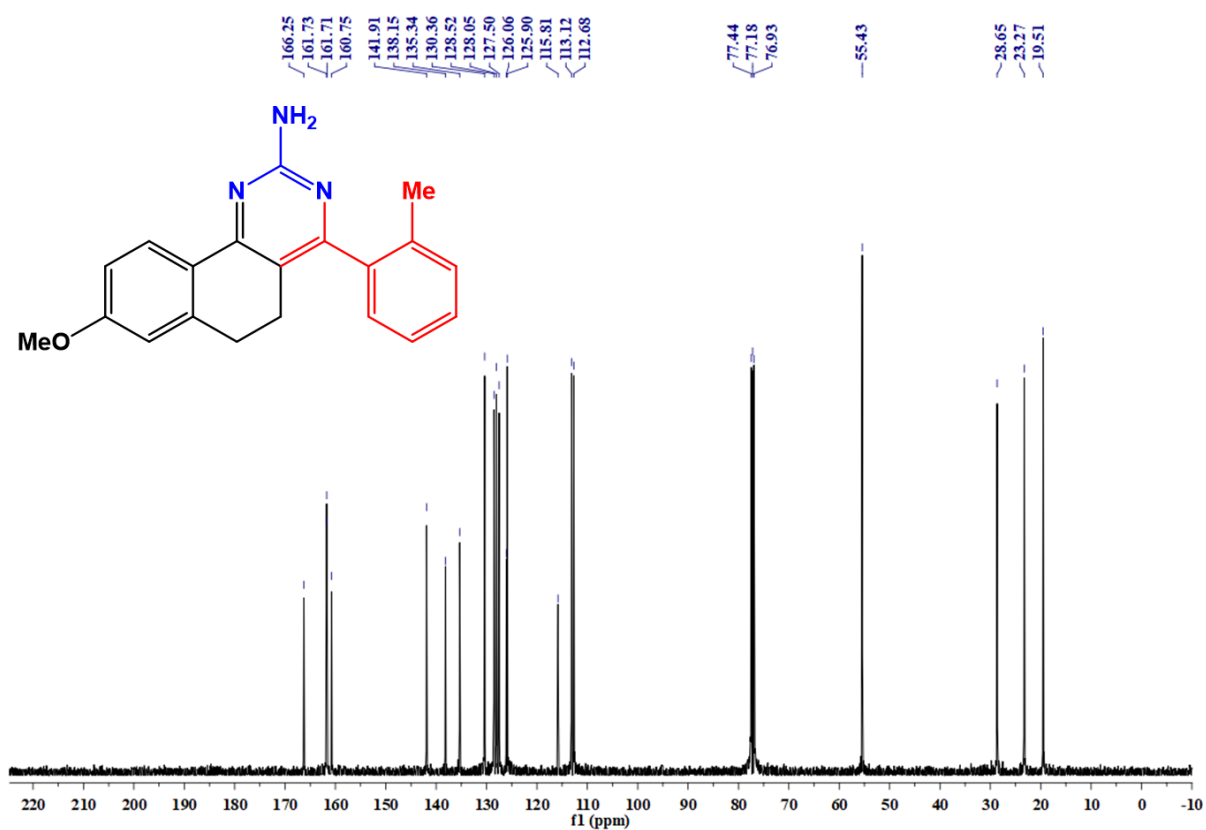


Figure S116: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8q** in CDCl_3 at 126 MHz.

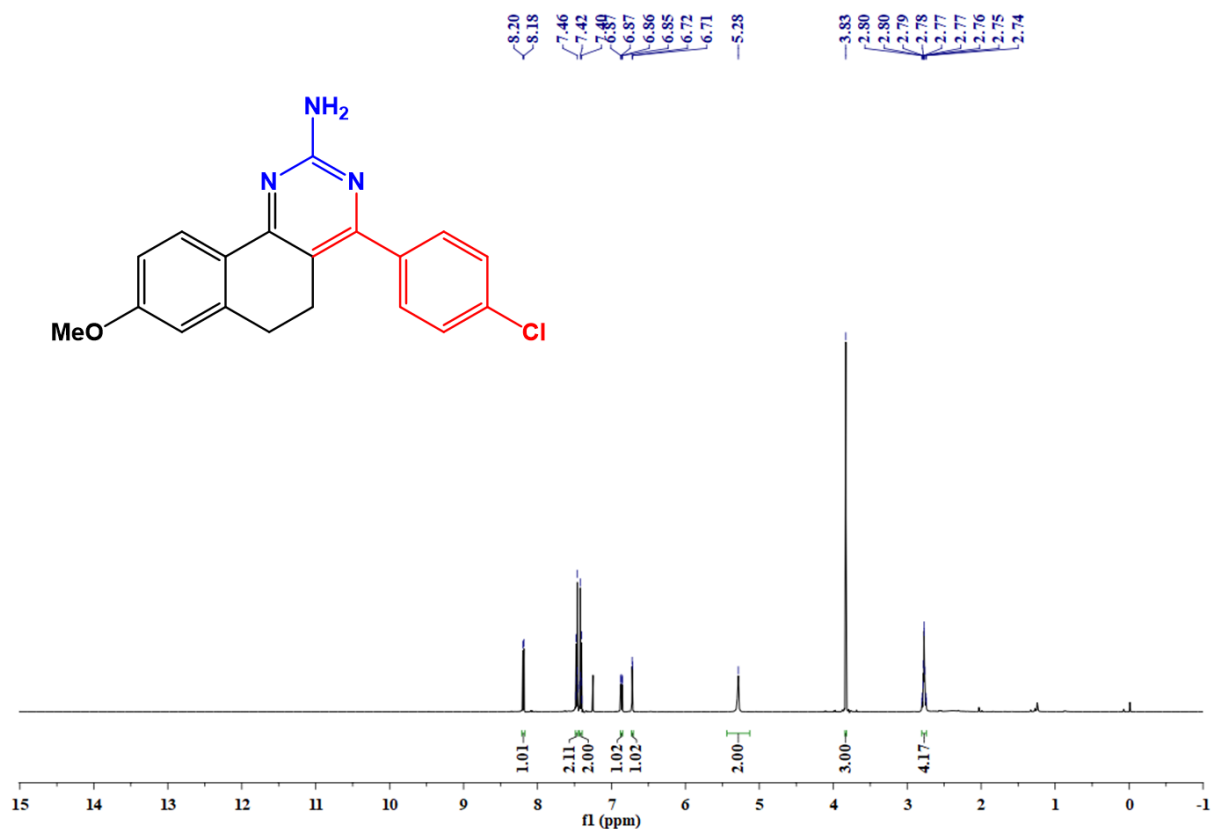


Figure S117: ^1H NMR spectrum of **8r** in CDCl_3 at 500 MHz.

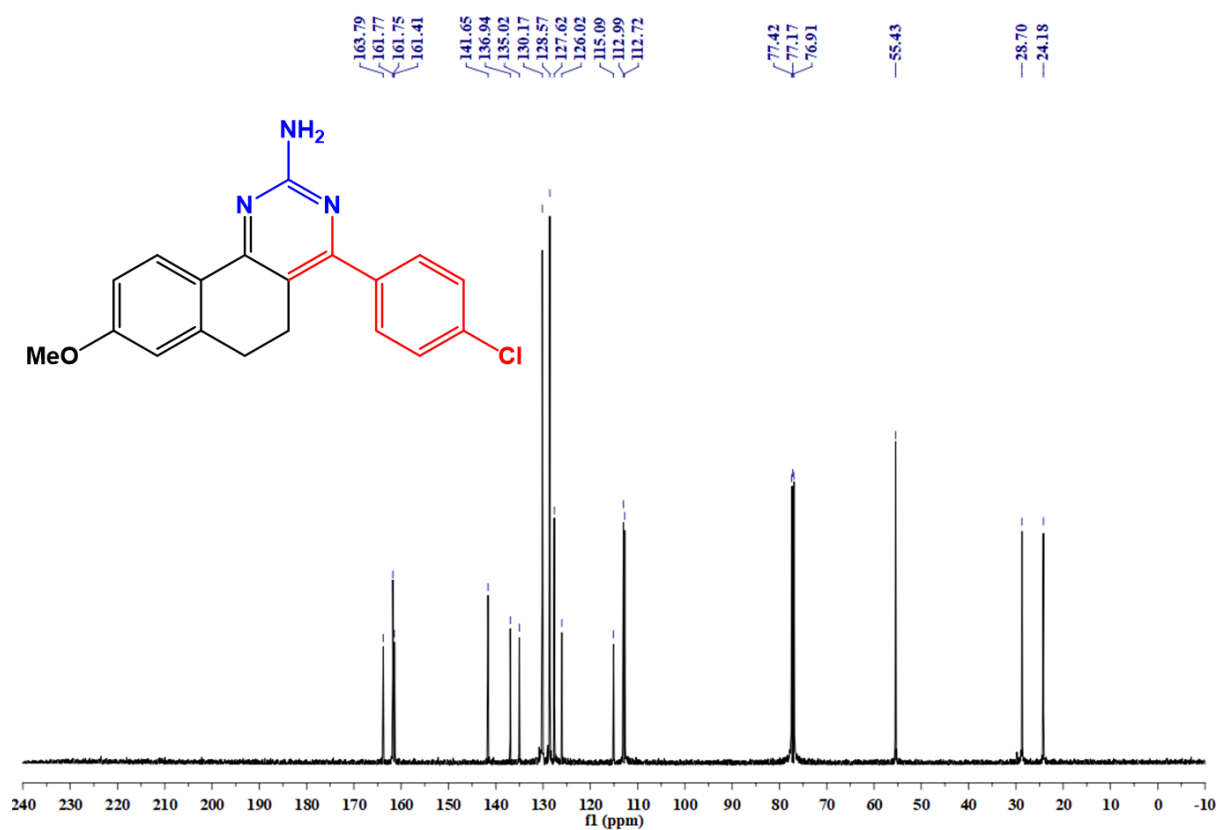


Figure S118: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8r** in CDCl_3 at 126 MHz.

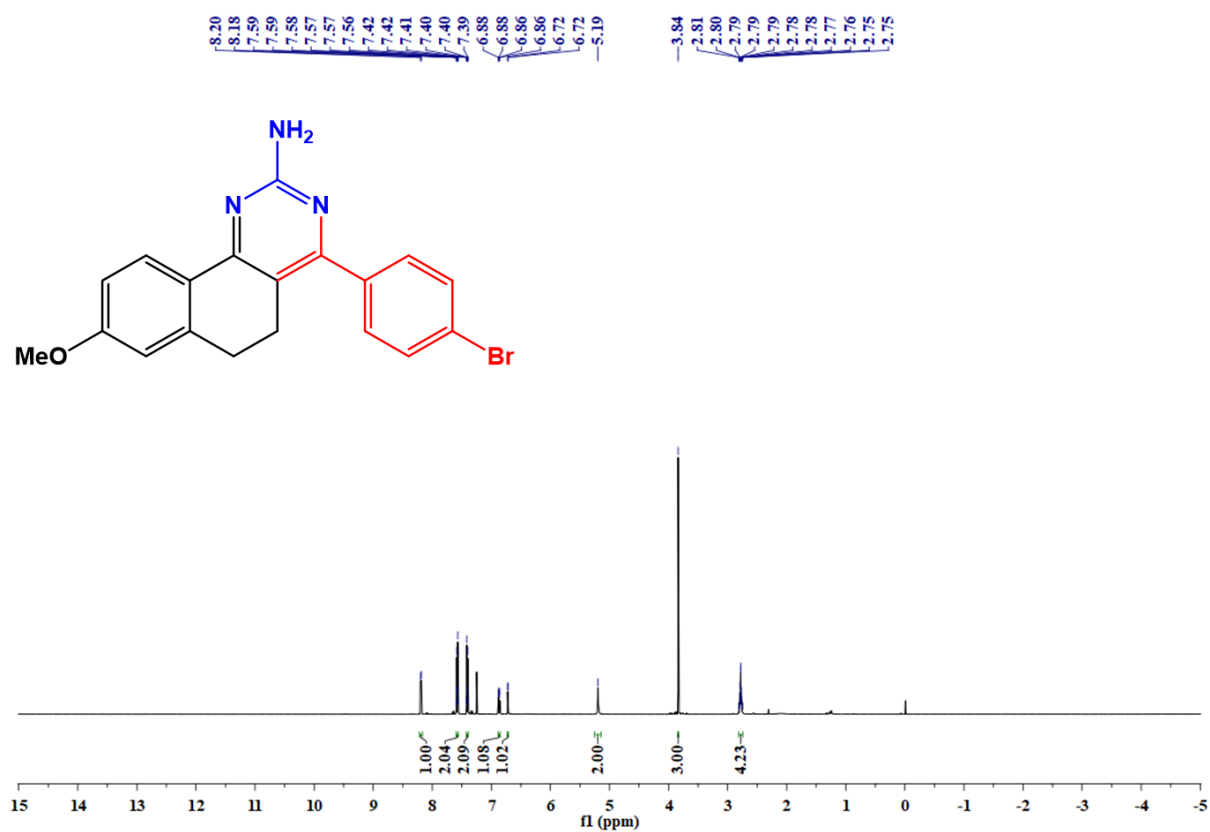


Figure S119: ^1H NMR spectrum of **8s** in CDCl_3 at 500 MHz.

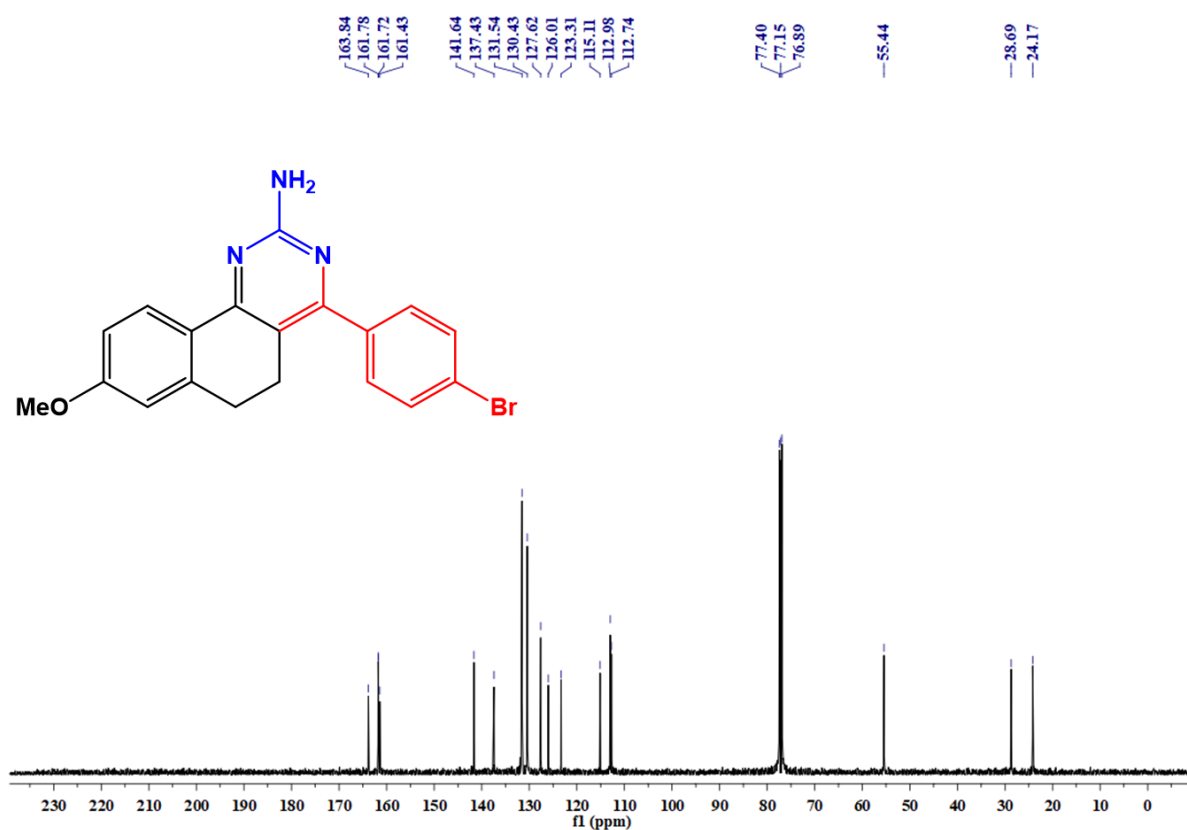


Figure S120: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8s** in CDCl_3 at 126 MHz.

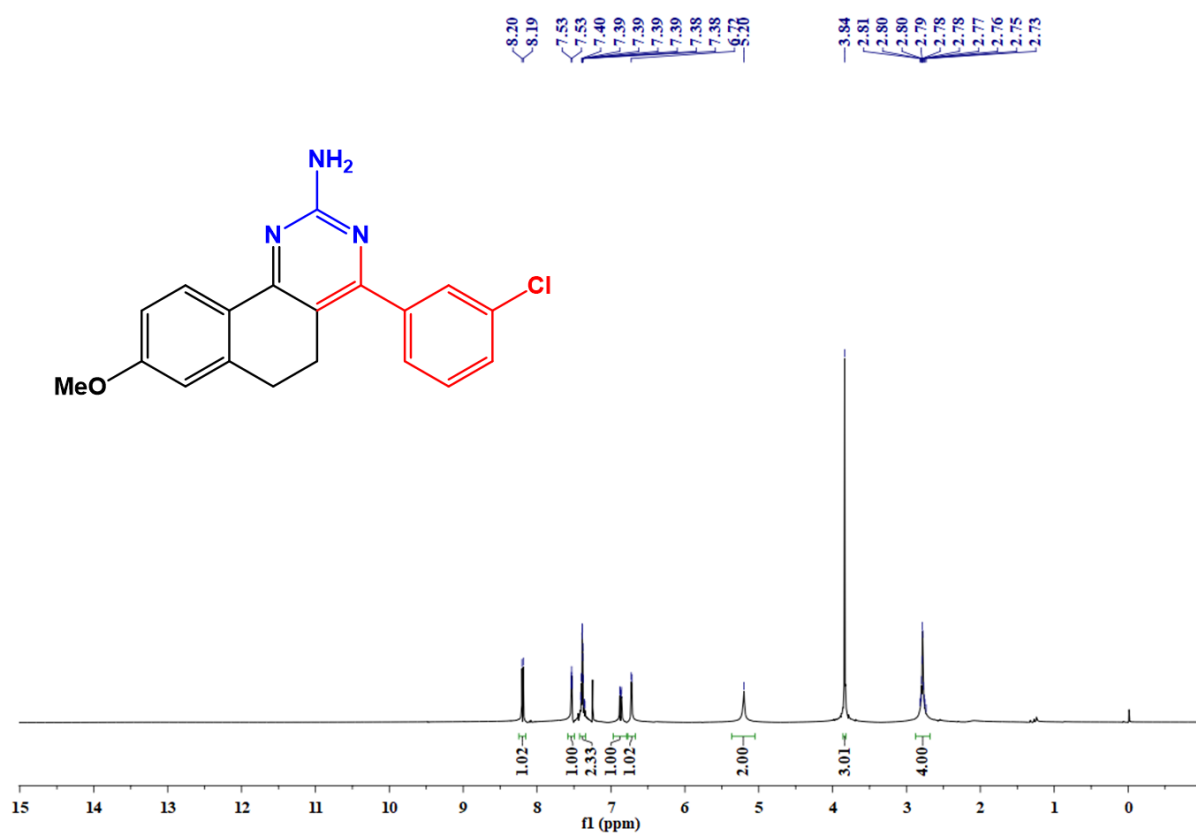


Figure S121: ^1H NMR spectrum of **8t** in CDCl_3 at 500 MHz.

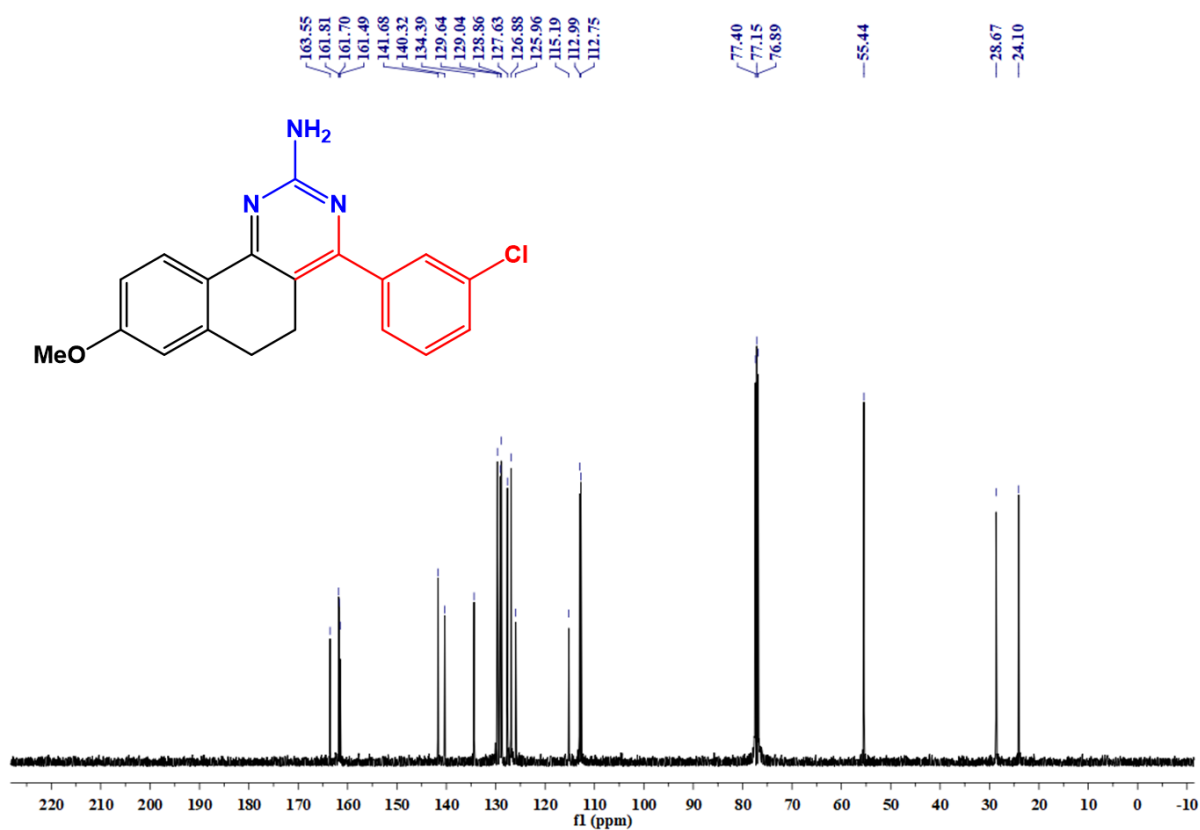


Figure S122: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8t** in CDCl_3 at 126 MHz.

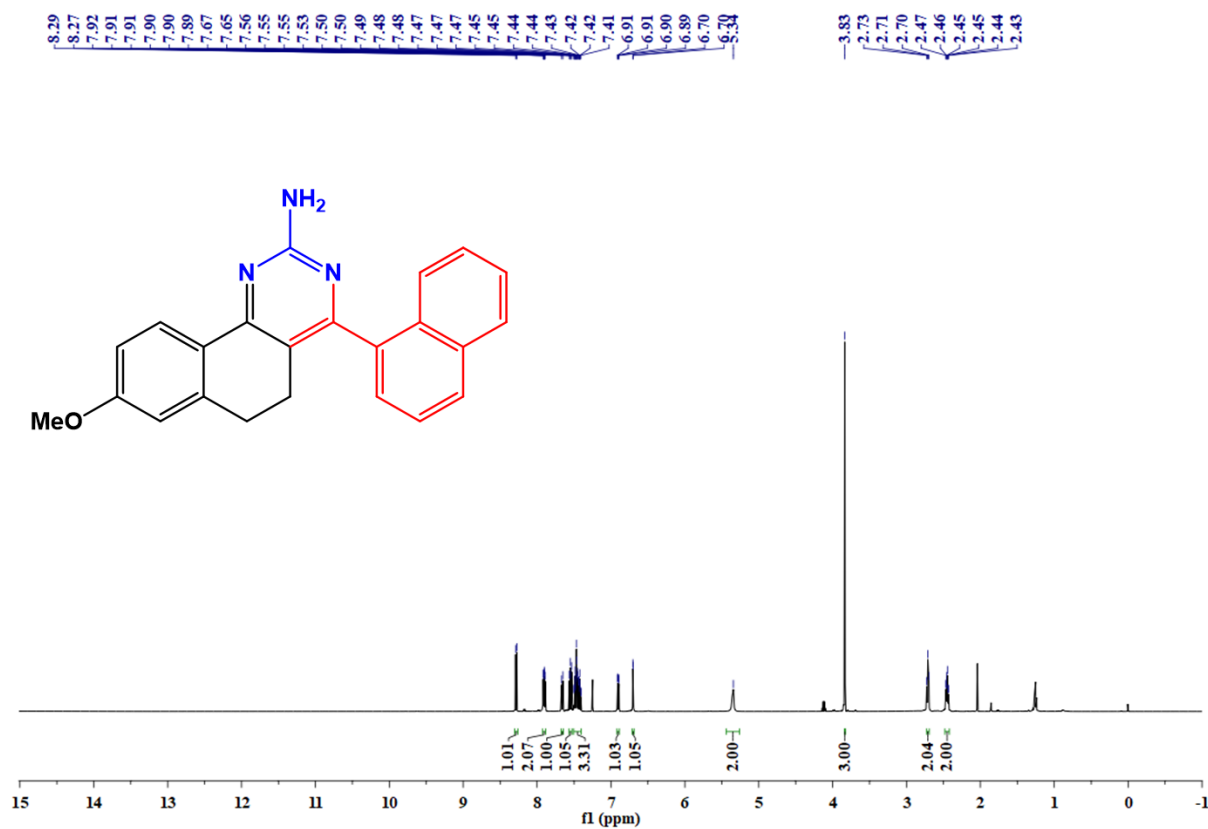
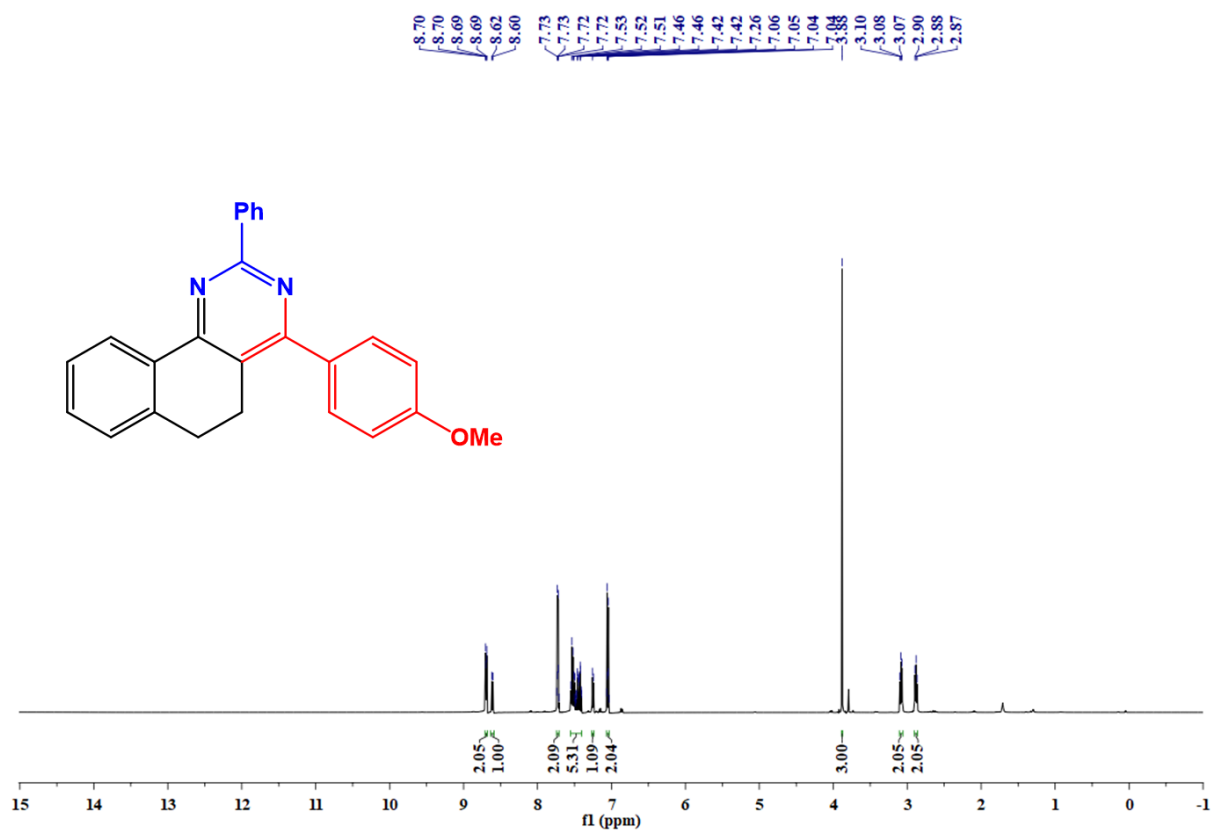
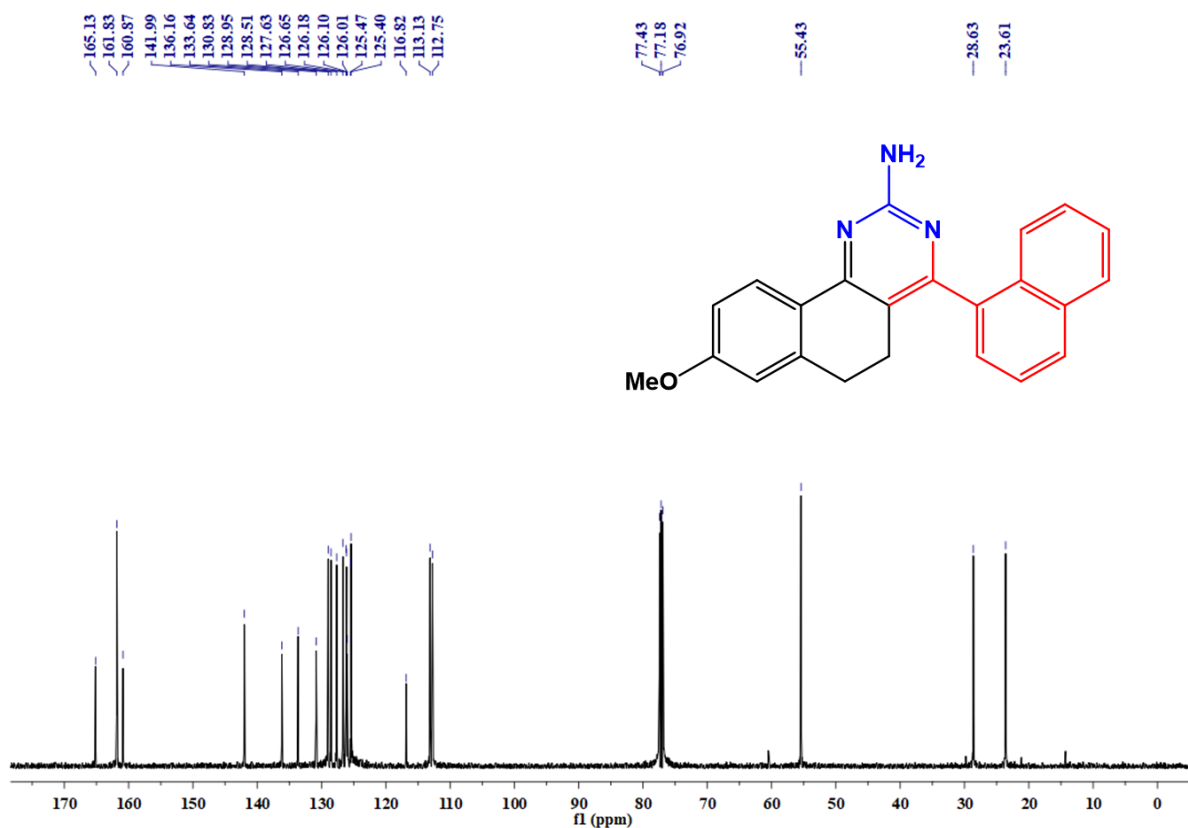
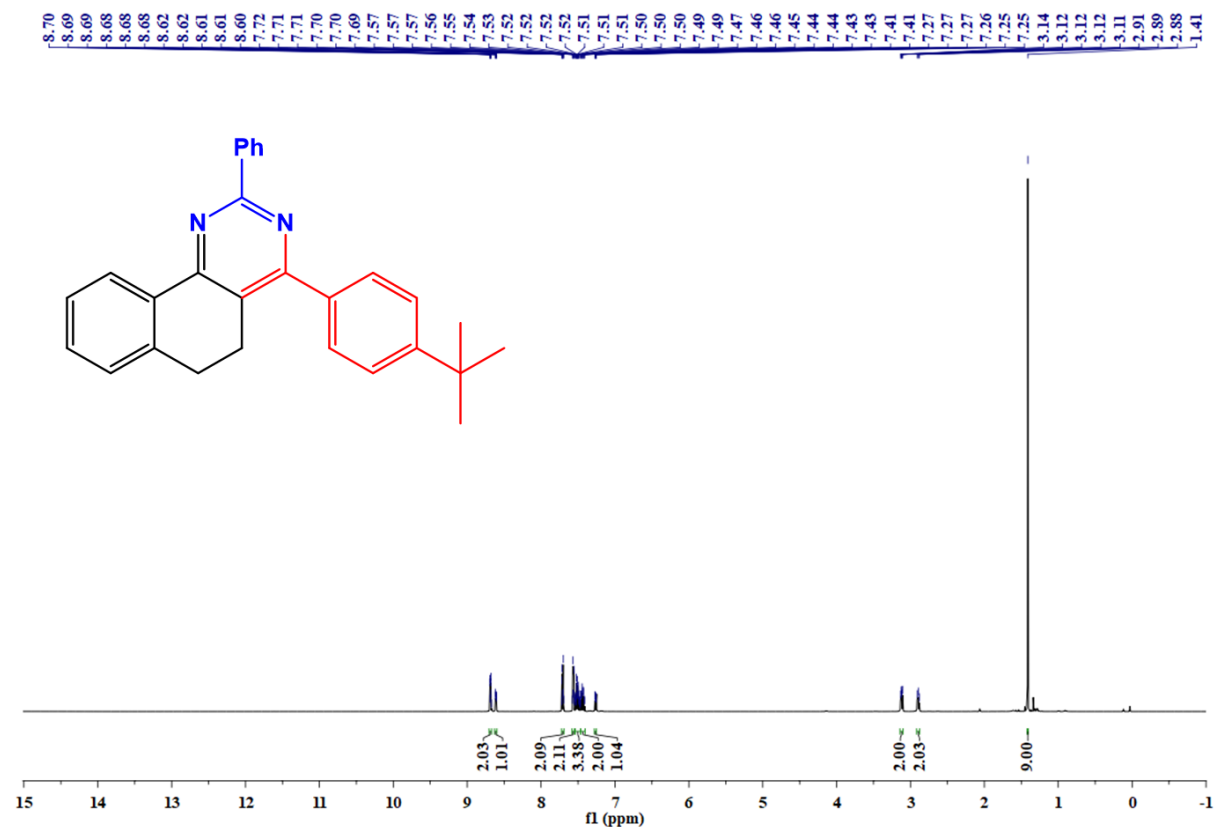
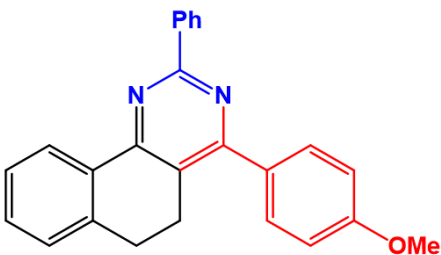


Figure S123: ^1H NMR spectrum of **8u** in CDCl_3 at 500 MHz.





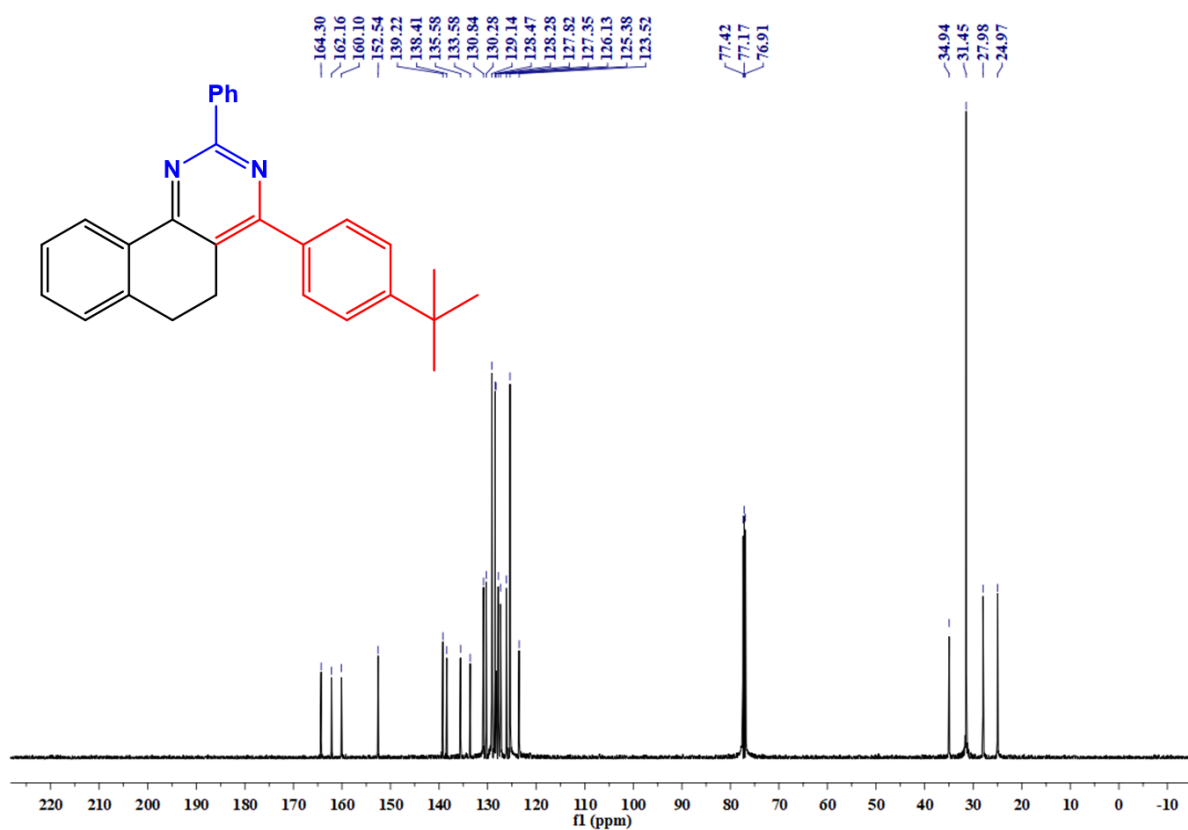


Figure S128: ¹³C{¹H} NMR spectrum of **8w** in CDCl₃ at 126 MHz.

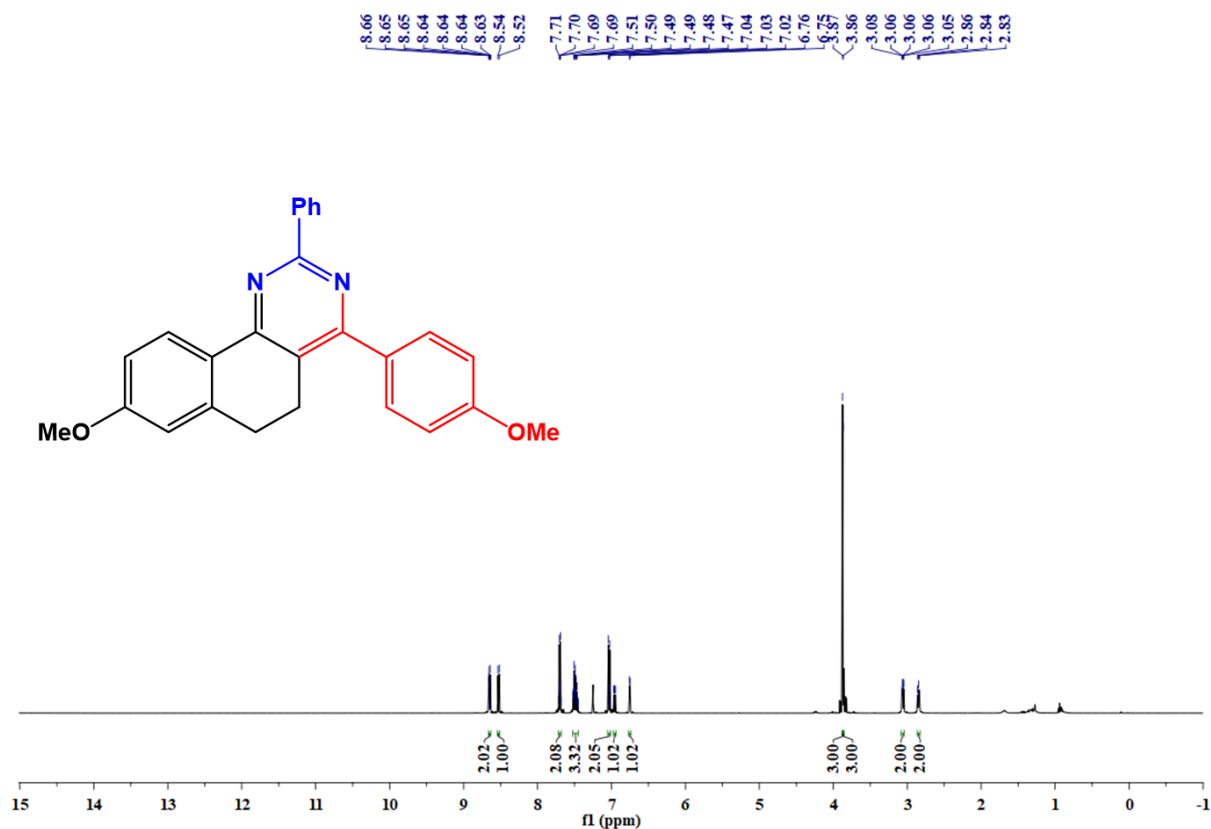


Figure S129: ¹H NMR spectrum of **8x** in CDCl₃ at 500 MHz.

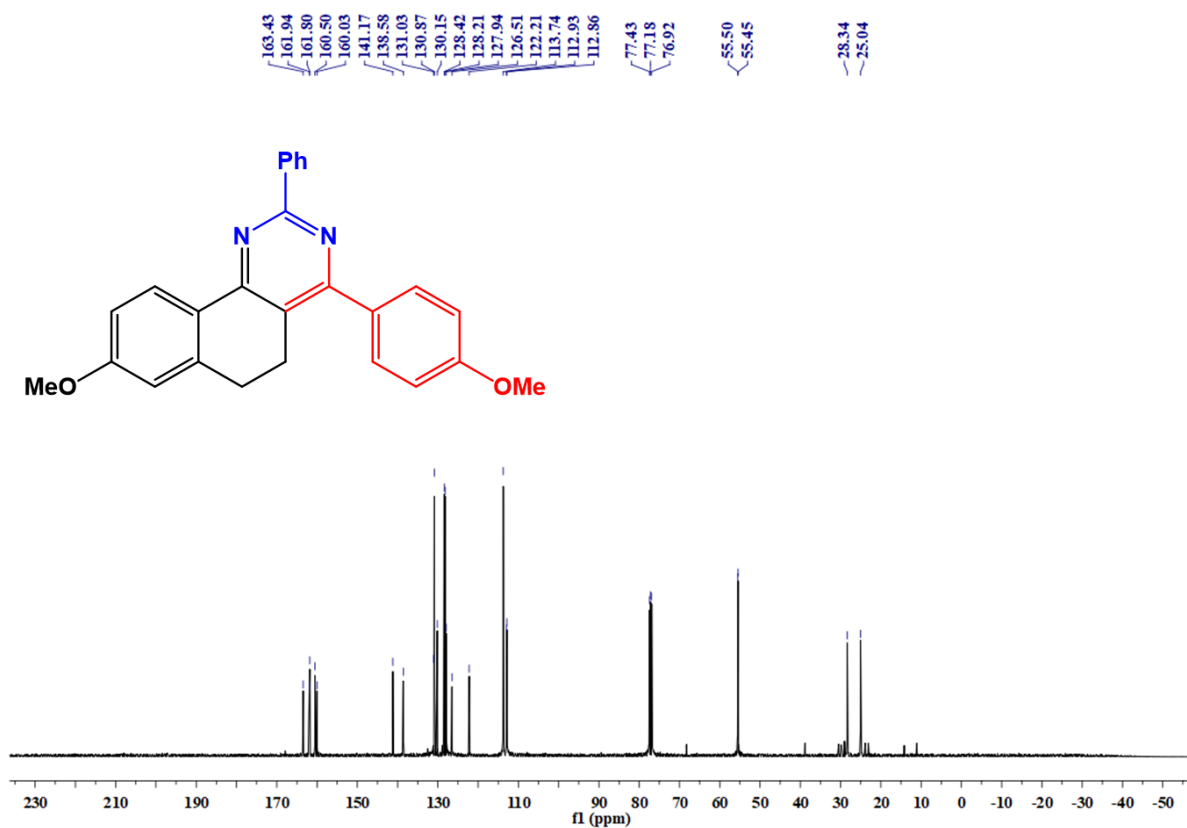


Figure S130: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8x** in CDCl_3 at 126 MHz.

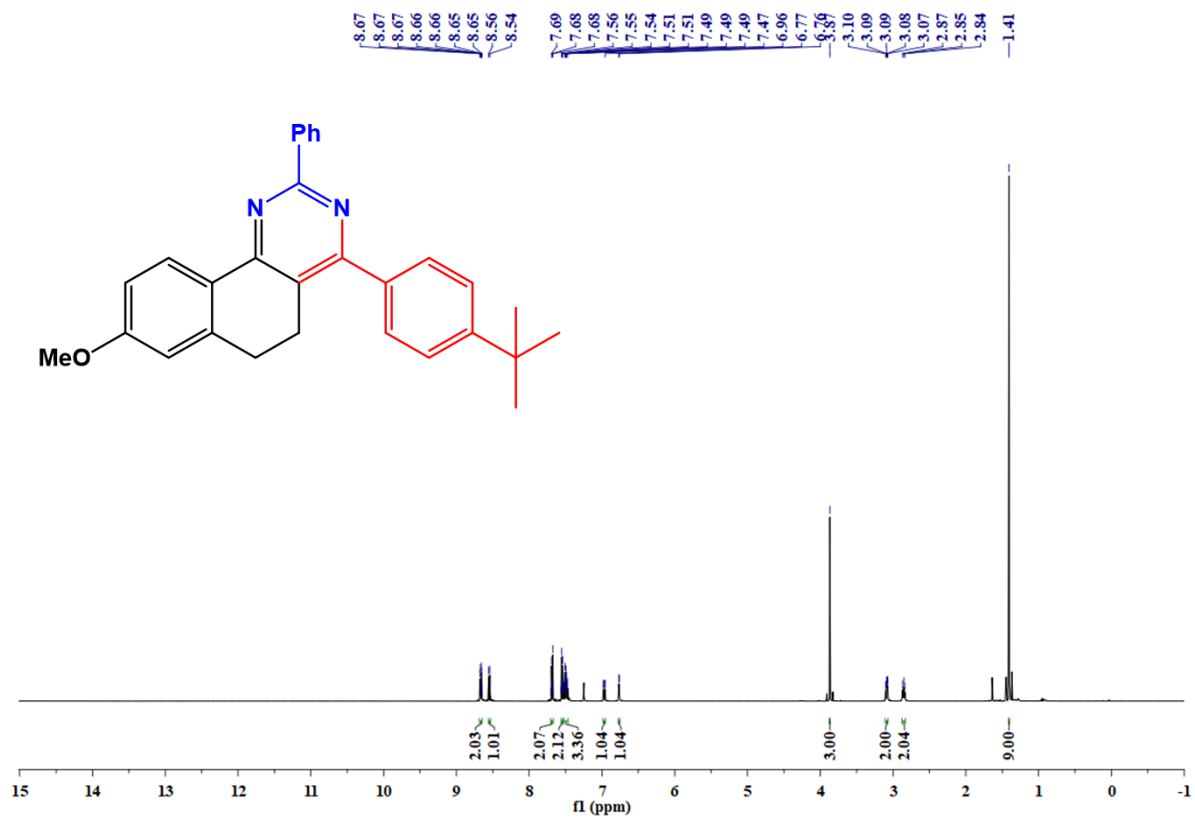


Figure S131: ^1H NMR spectrum of **8y** in CDCl_3 at 500 MHz.

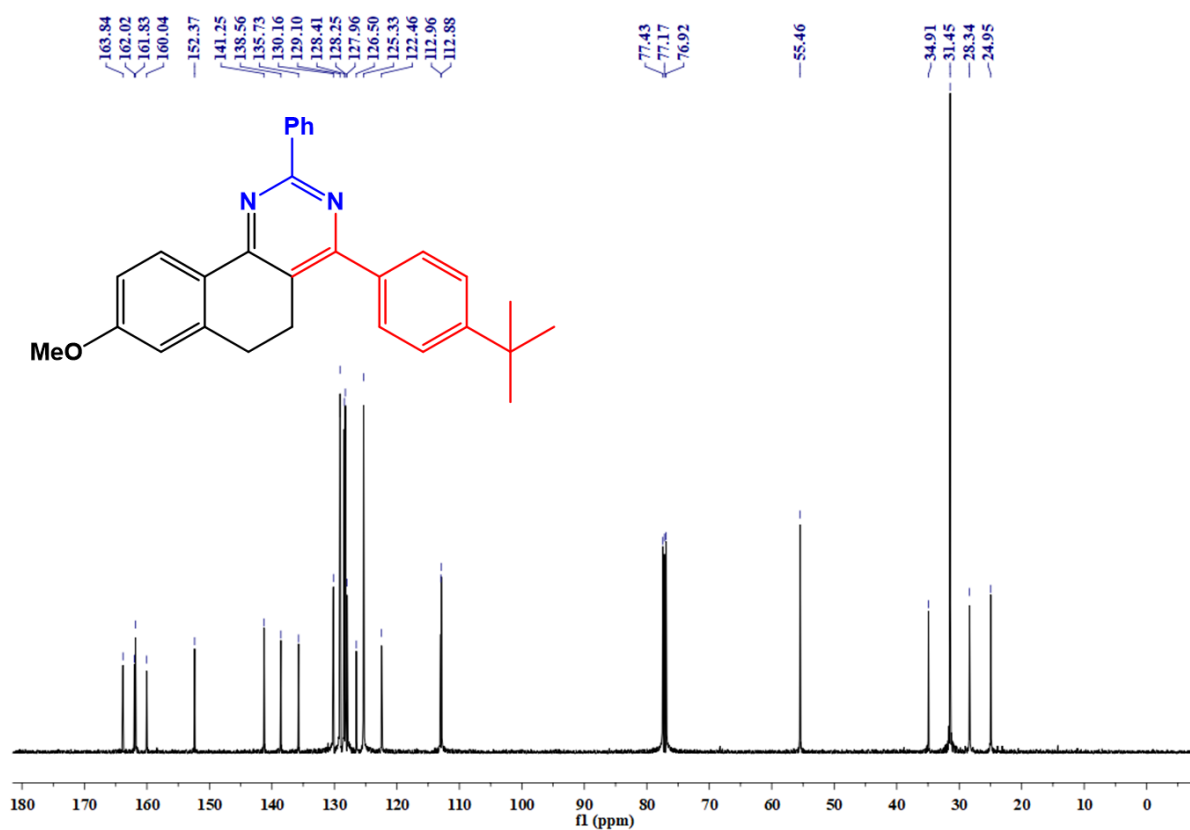


Figure S132: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8y** in CDCl_3 at 126 MHz.

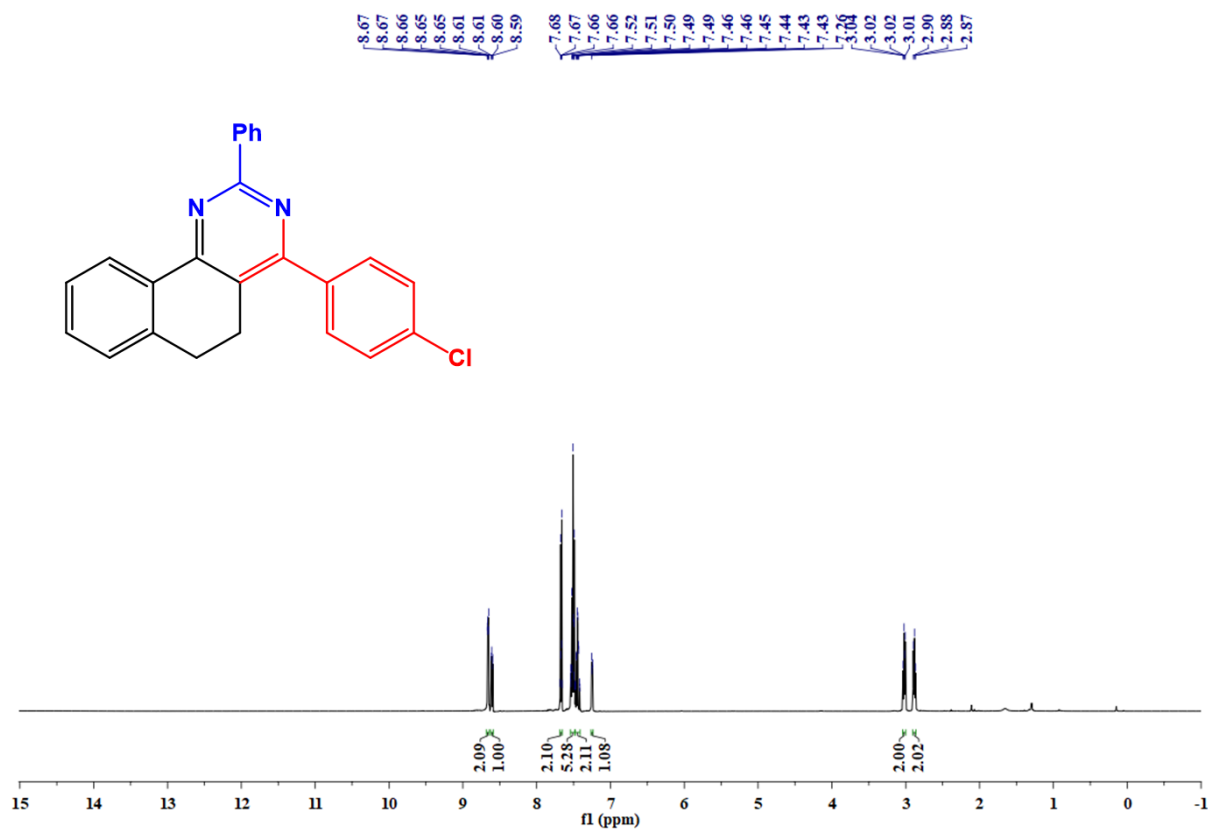


Figure S133: ^1H NMR spectrum of **8z** in CDCl_3 at 500 MHz.

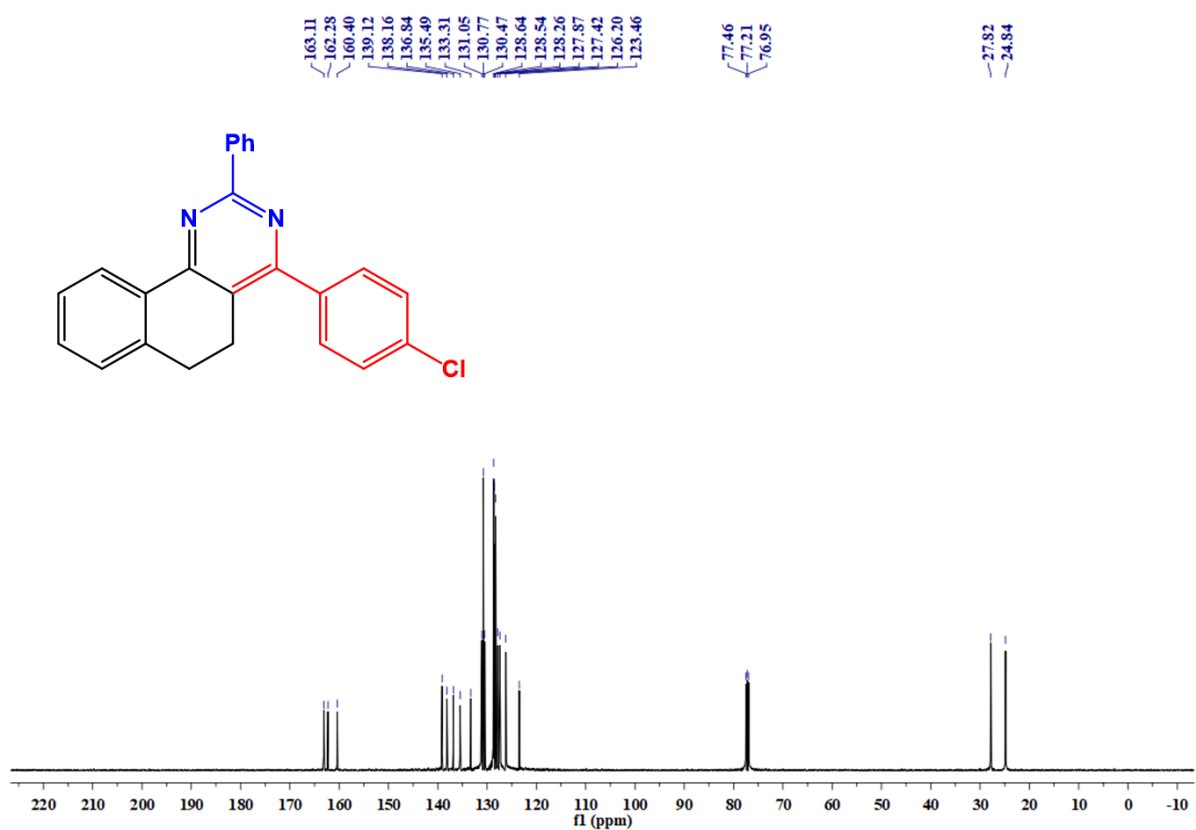


Figure S134: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8z** in CDCl_3 at 126 MHz.

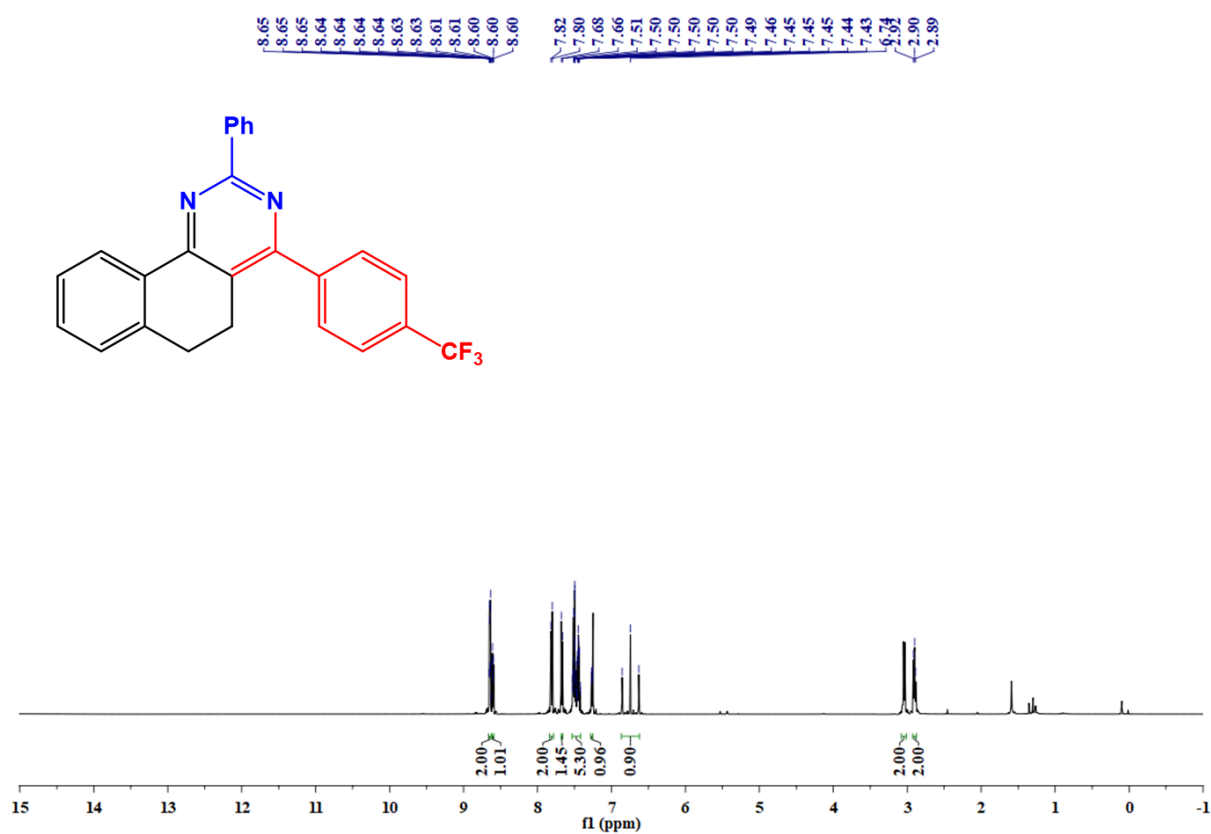


Figure S135: ^1H NMR spectrum of **8aa** in CDCl_3 at 500 MHz.

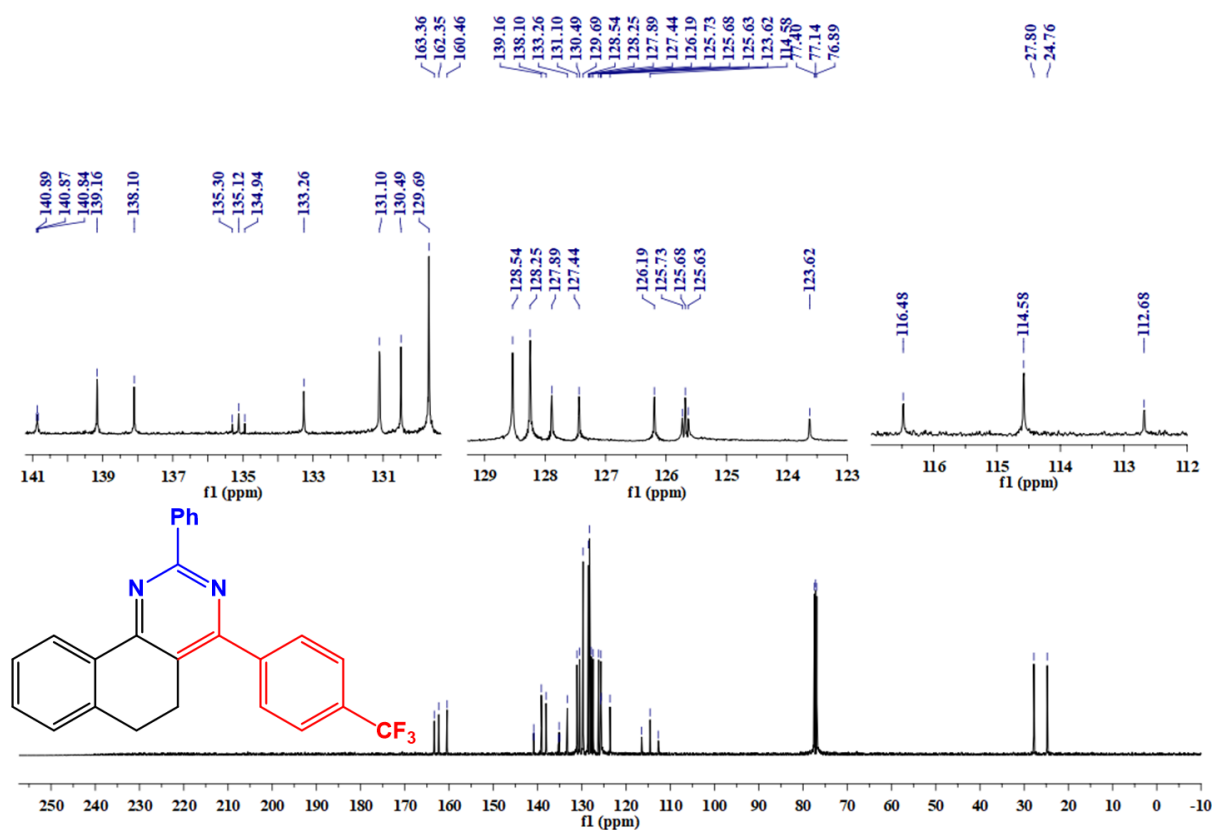


Figure S136: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8aa** in CDCl_3 at 126 MHz.

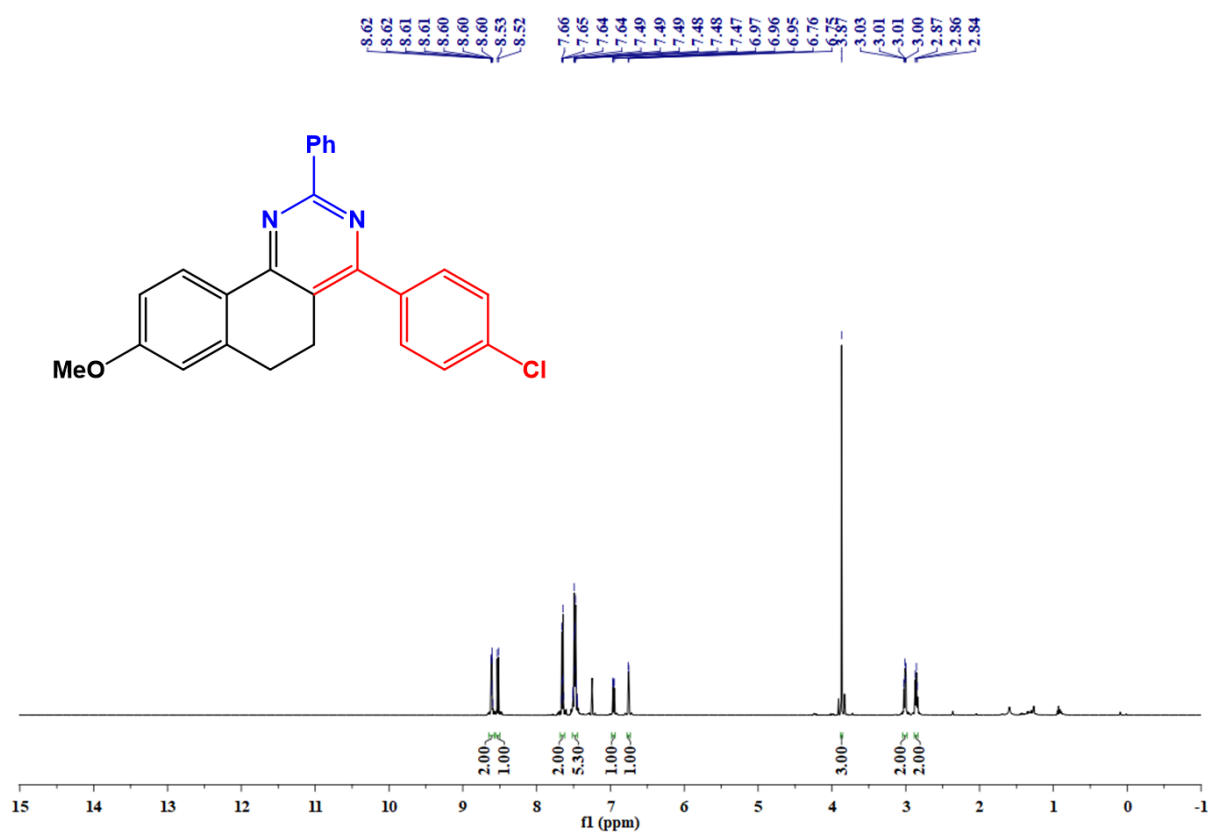


Figure S137: ^1H NMR spectrum of **8ab** in CDCl_3 at 500 MHz.

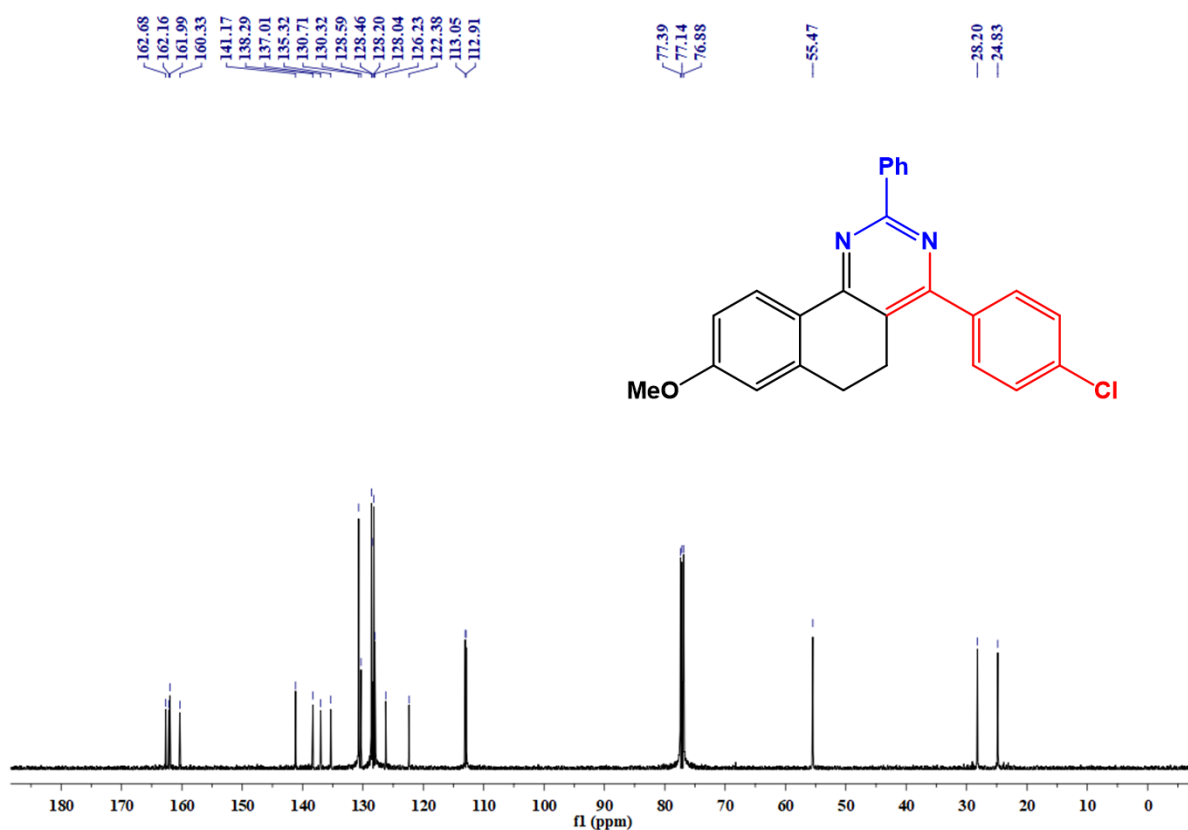


Figure S138: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8ab** in CDCl_3 at 126 MHz.

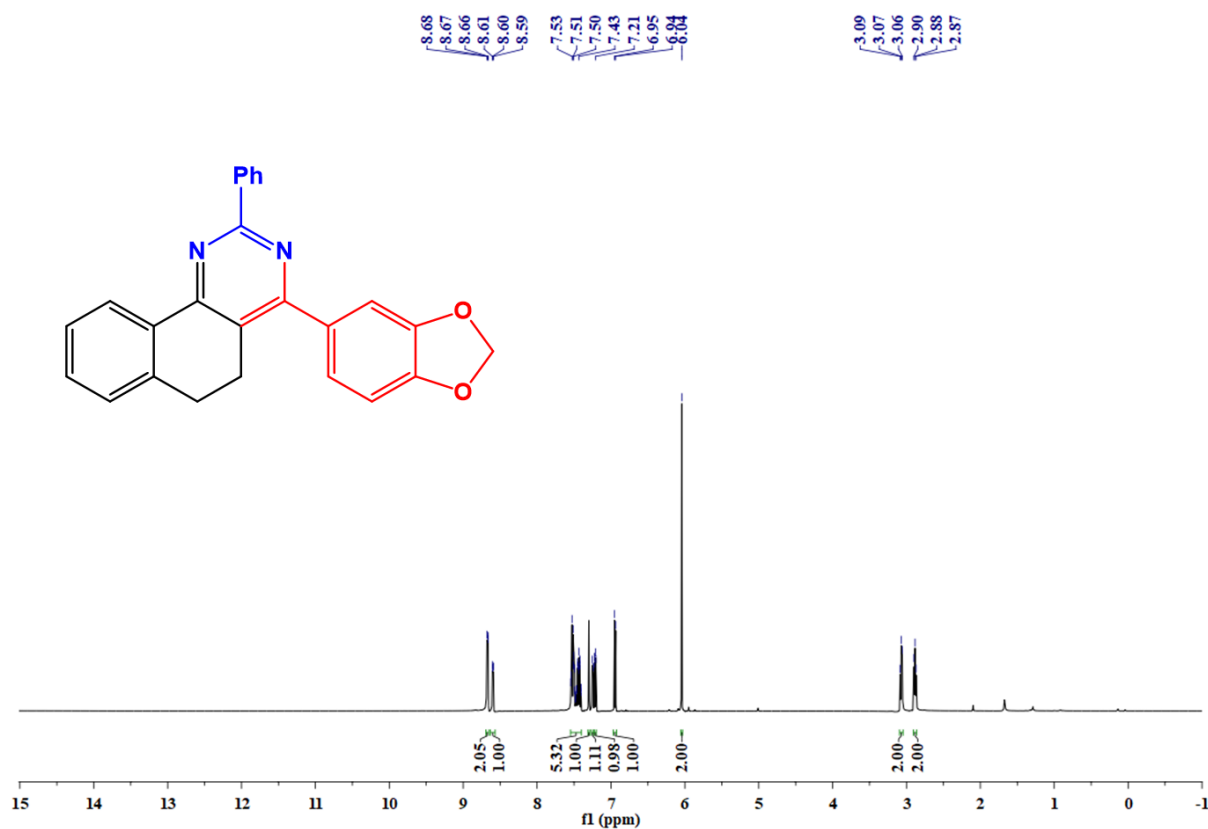
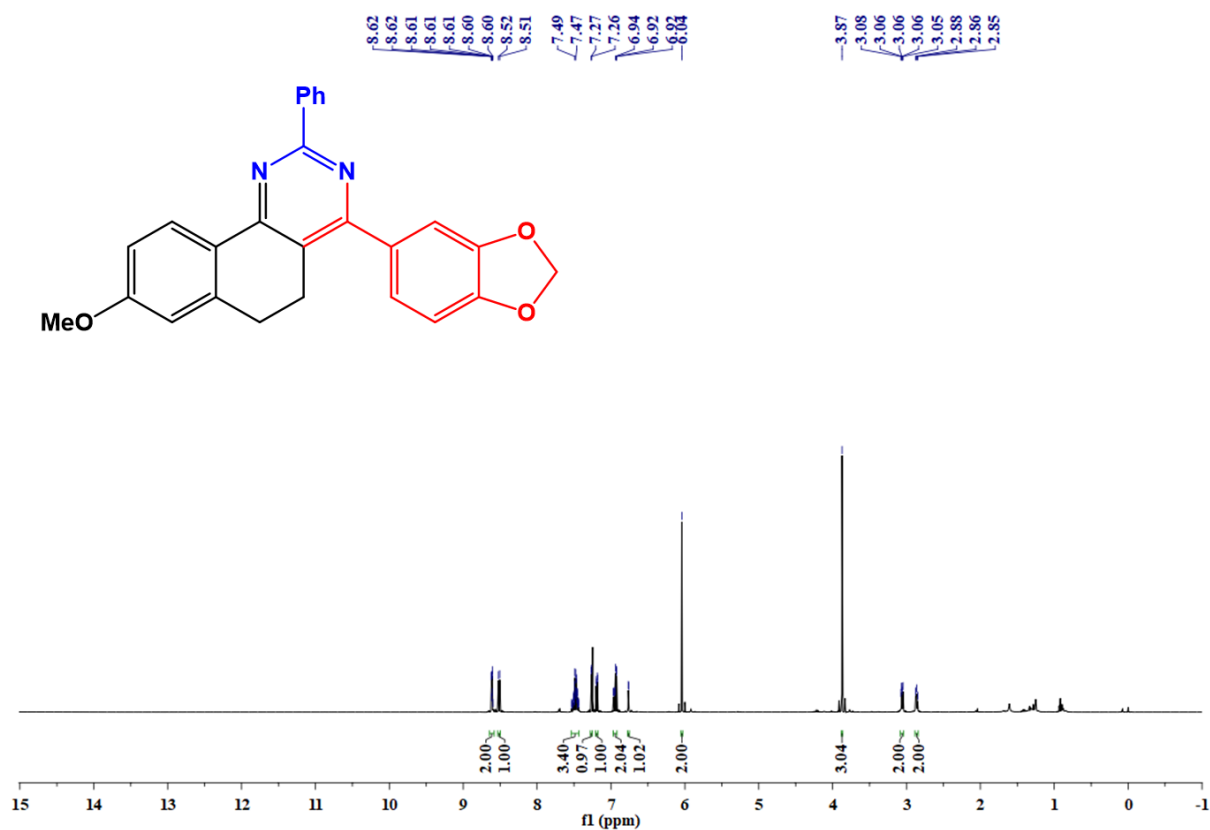
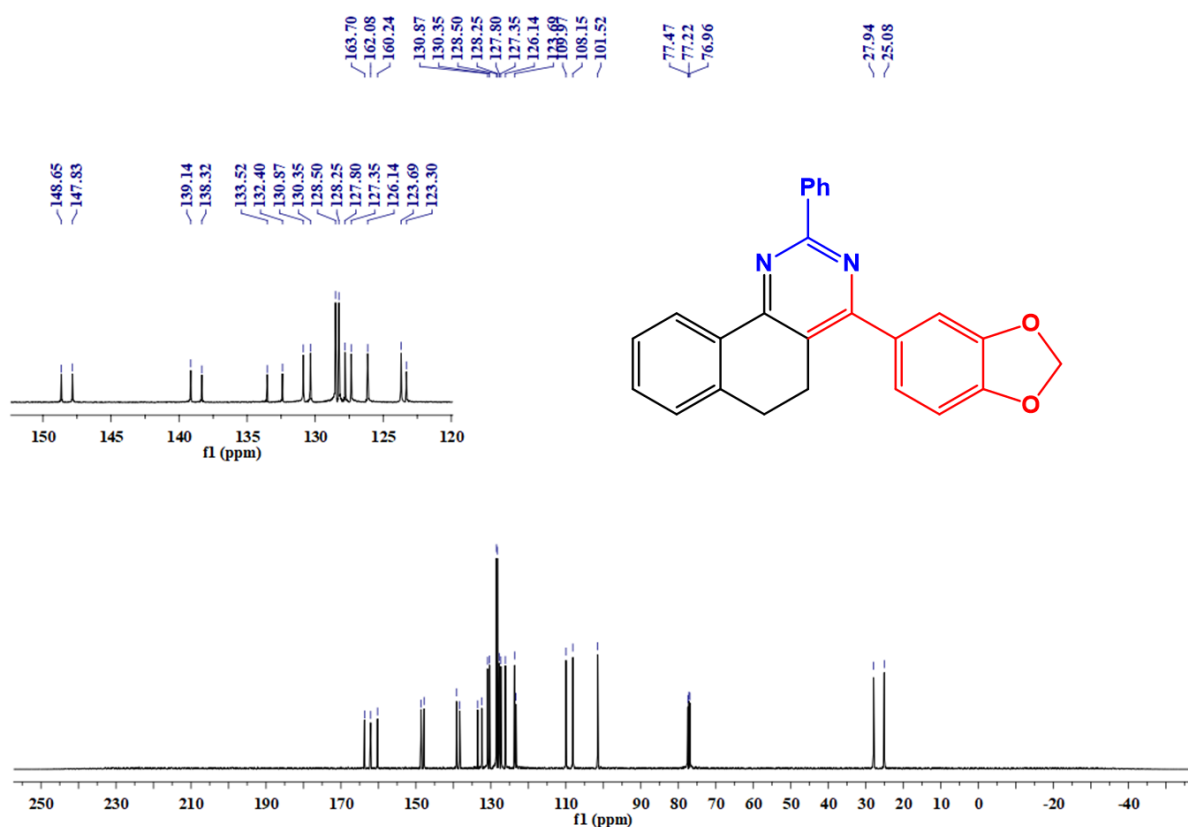
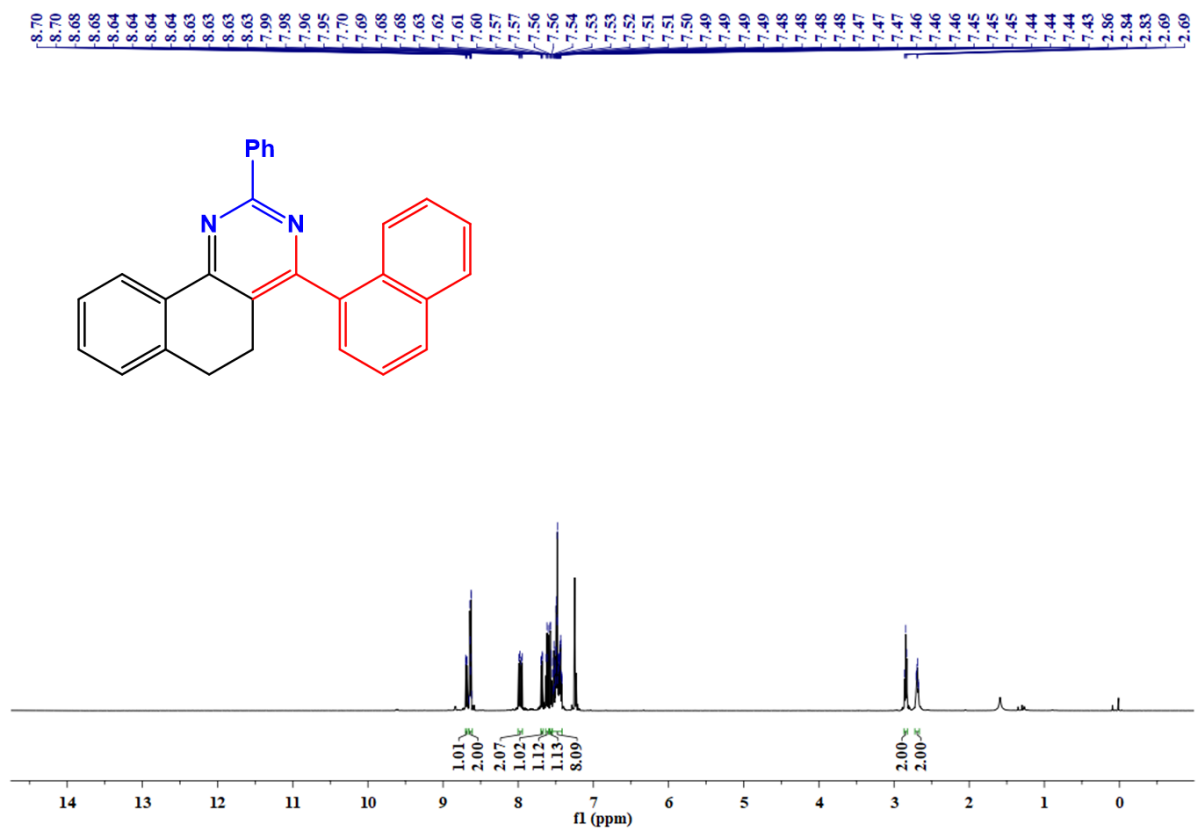
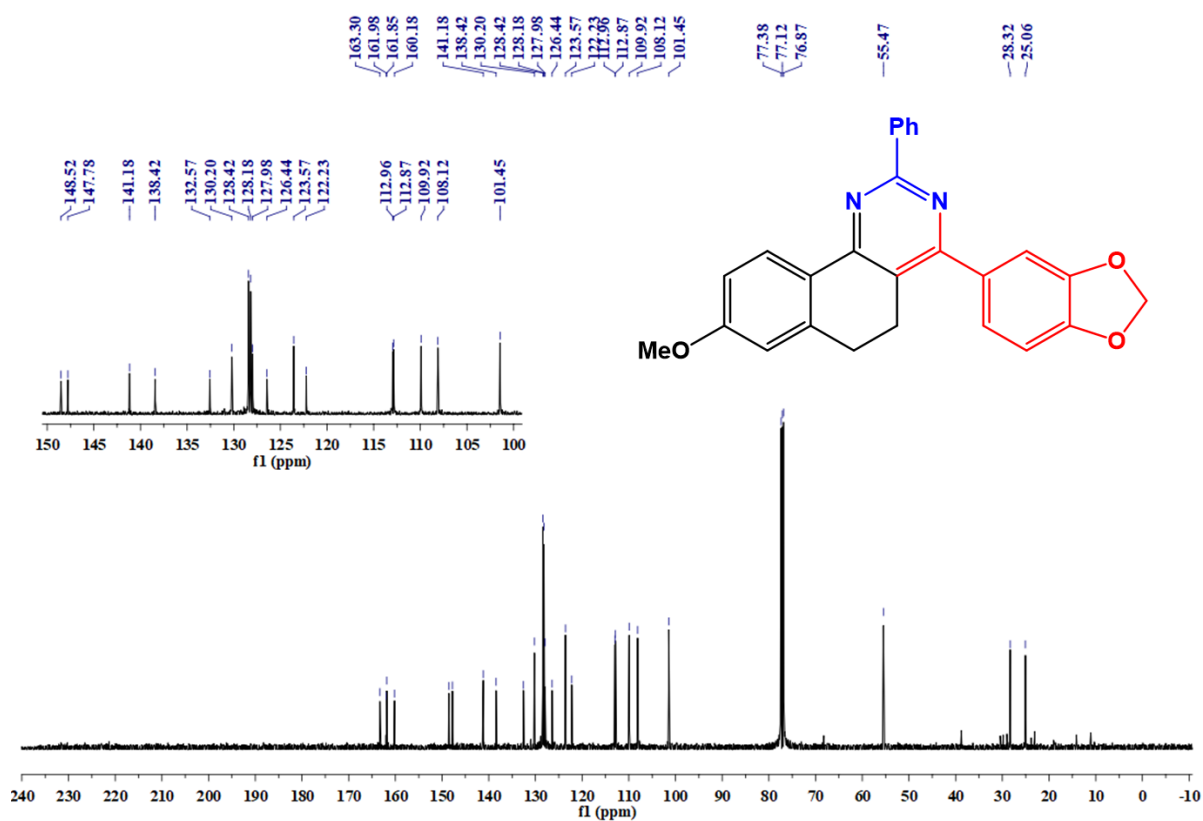


Figure S139: ^1H NMR spectrum of **8ac** in CDCl_3 at 500 MHz.





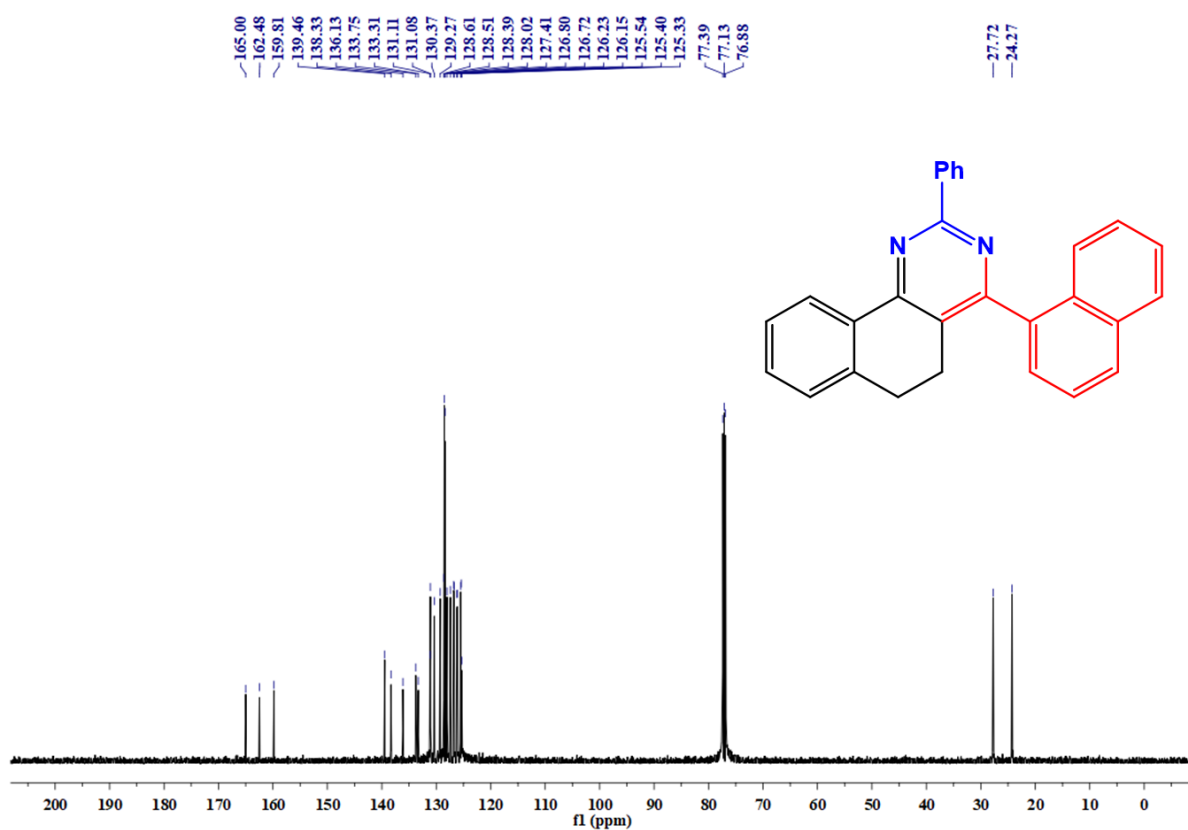


Figure S144: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8ae** in CDCl_3 at 126 MHz.

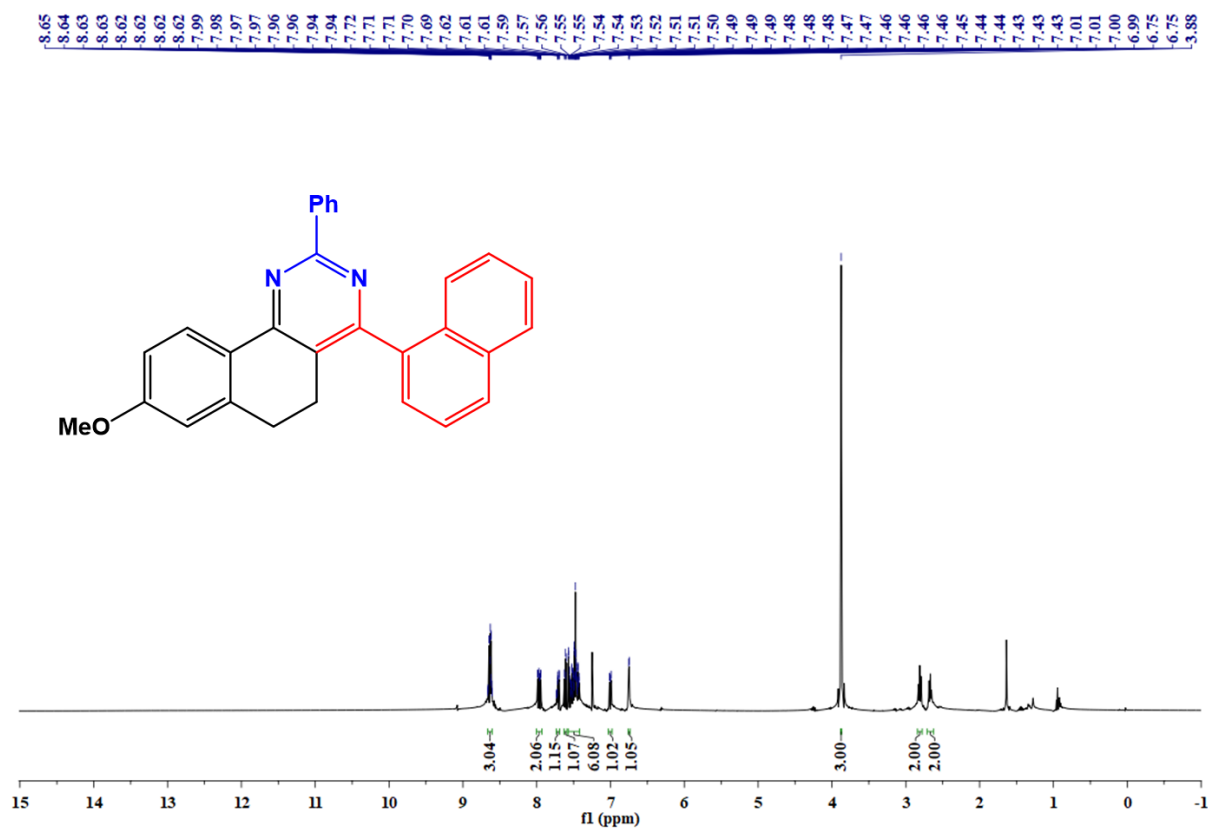
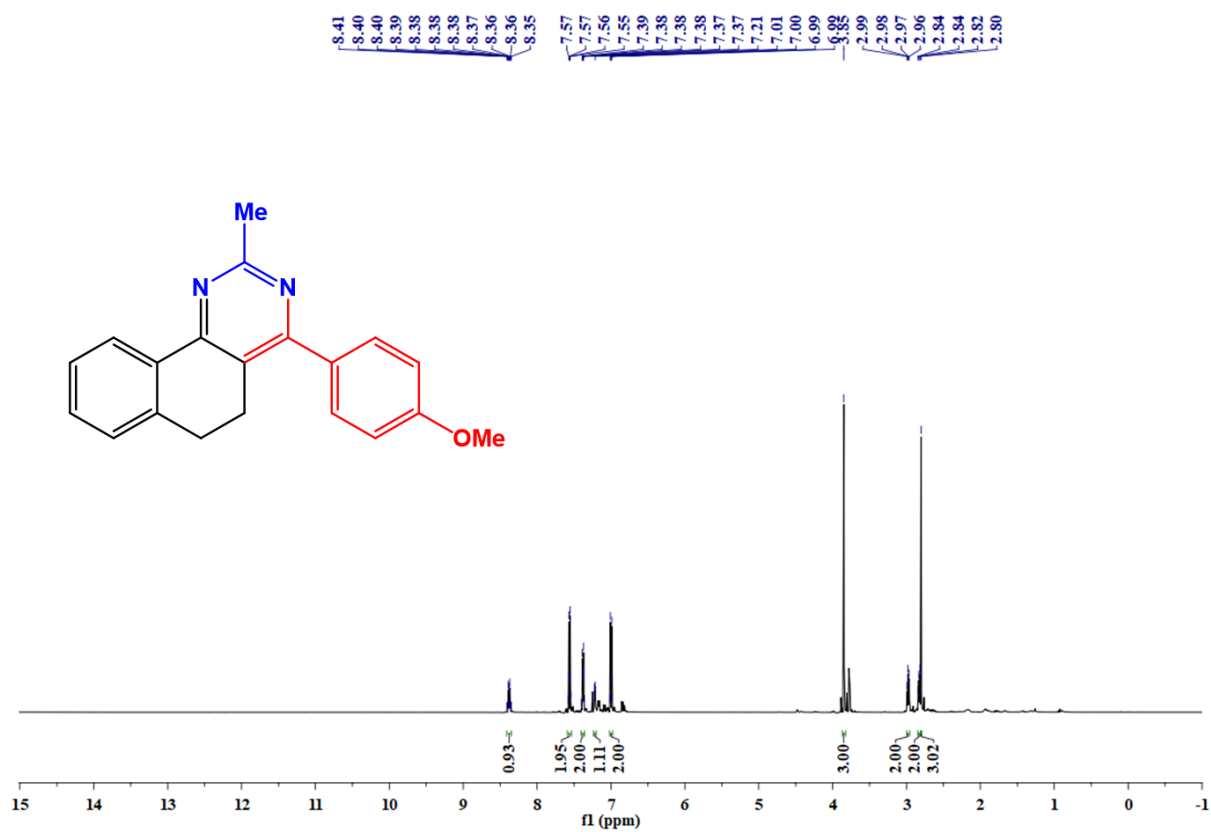
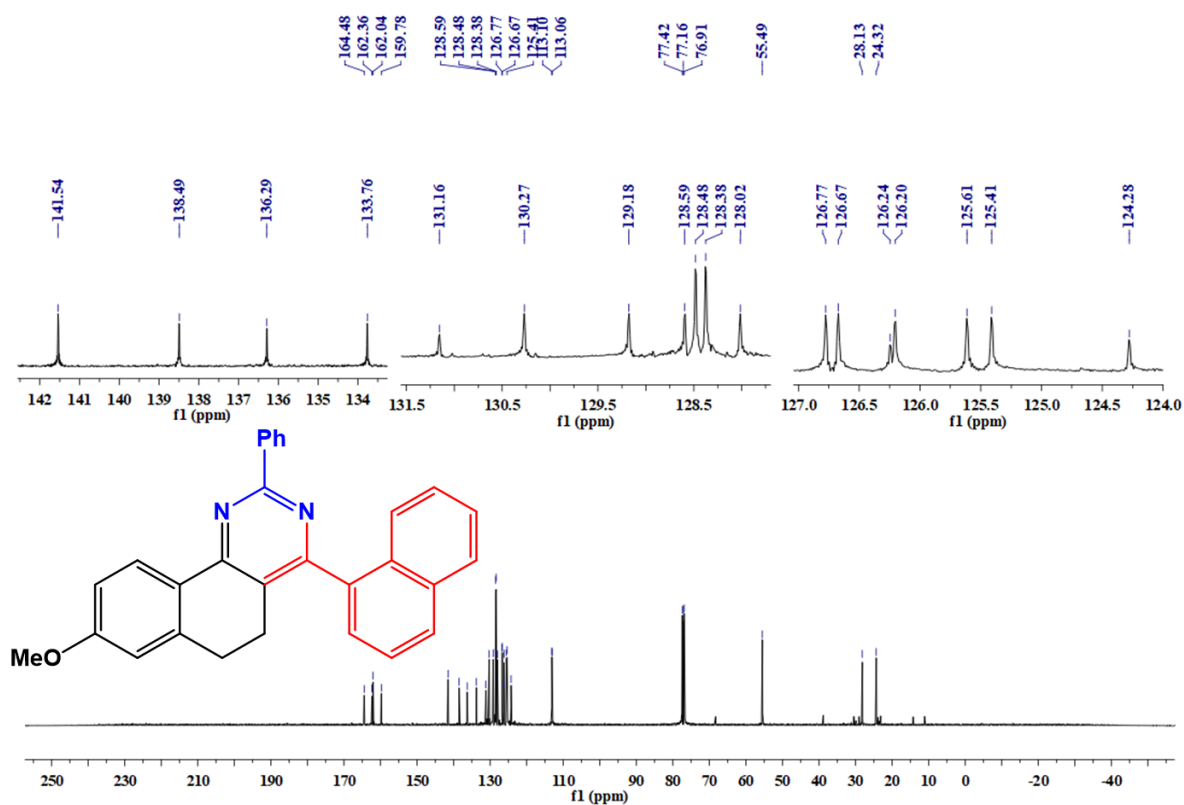


Figure S145: ^1H NMR spectrum of **8af** in CDCl_3 at 500 MHz.



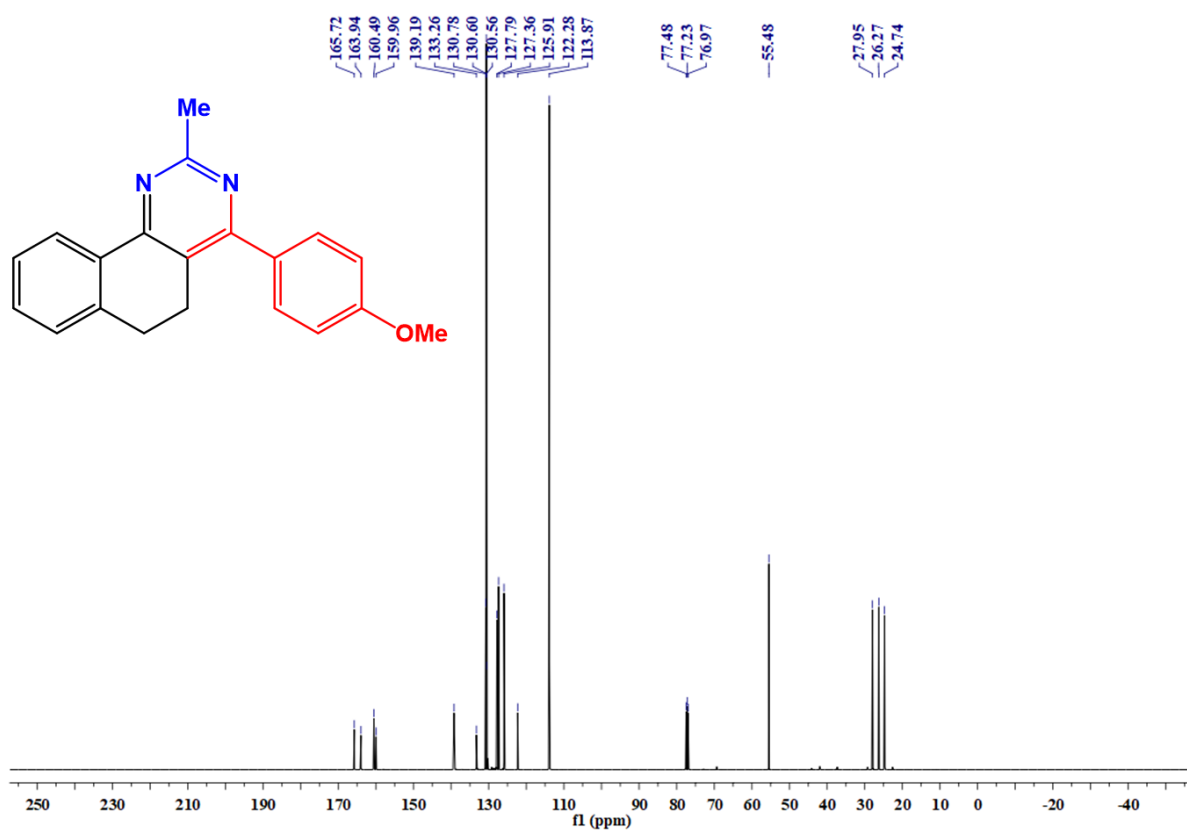


Figure S148: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8ag** in CDCl_3 at 126 MHz.

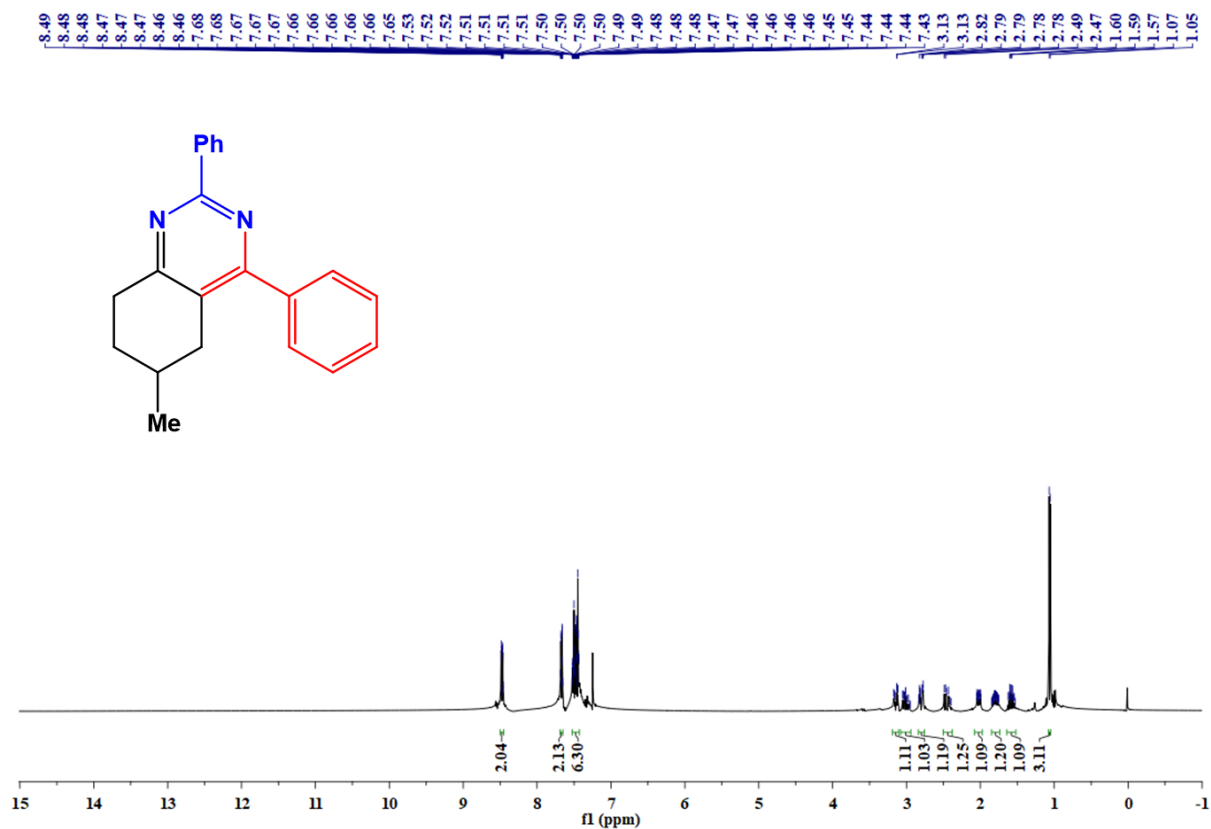


Figure S149: ^1H NMR spectrum of **8ah** in CDCl_3 at 500 MHz.

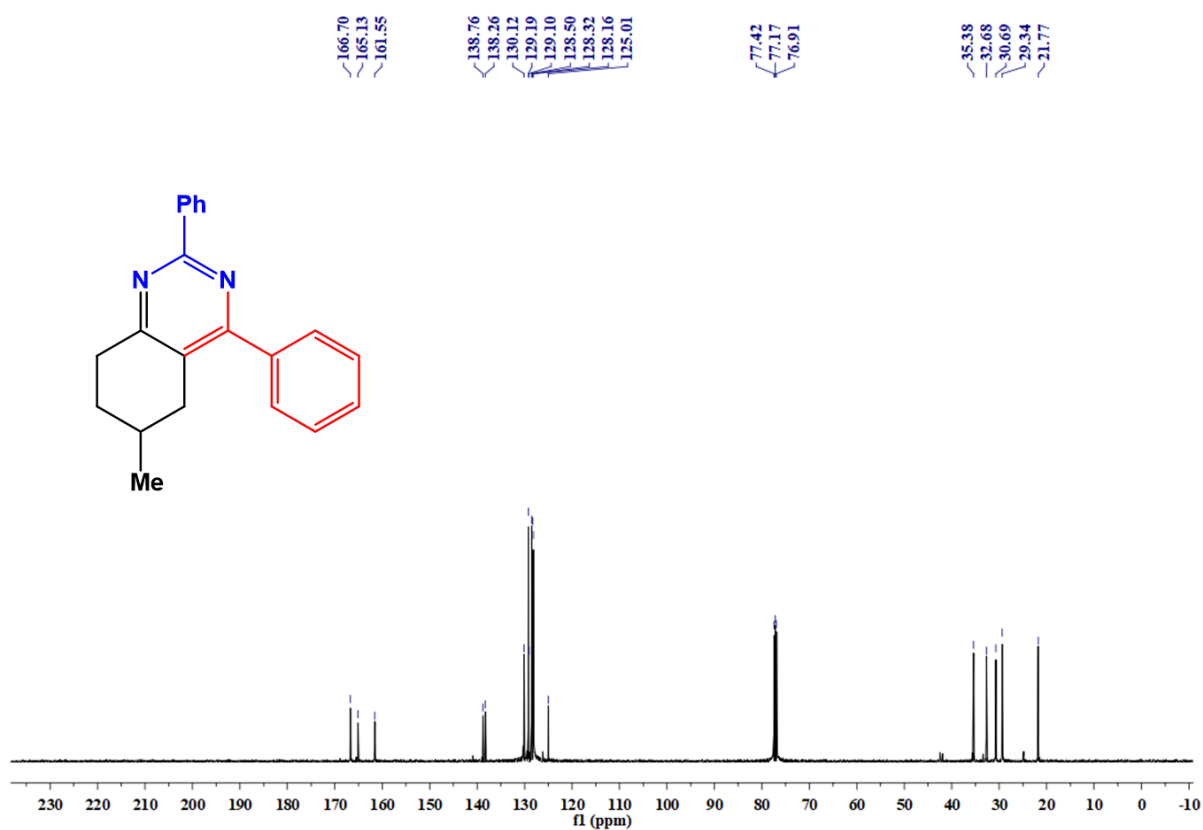


Figure S150: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8ah** in CDCl_3 at 126 MHz.

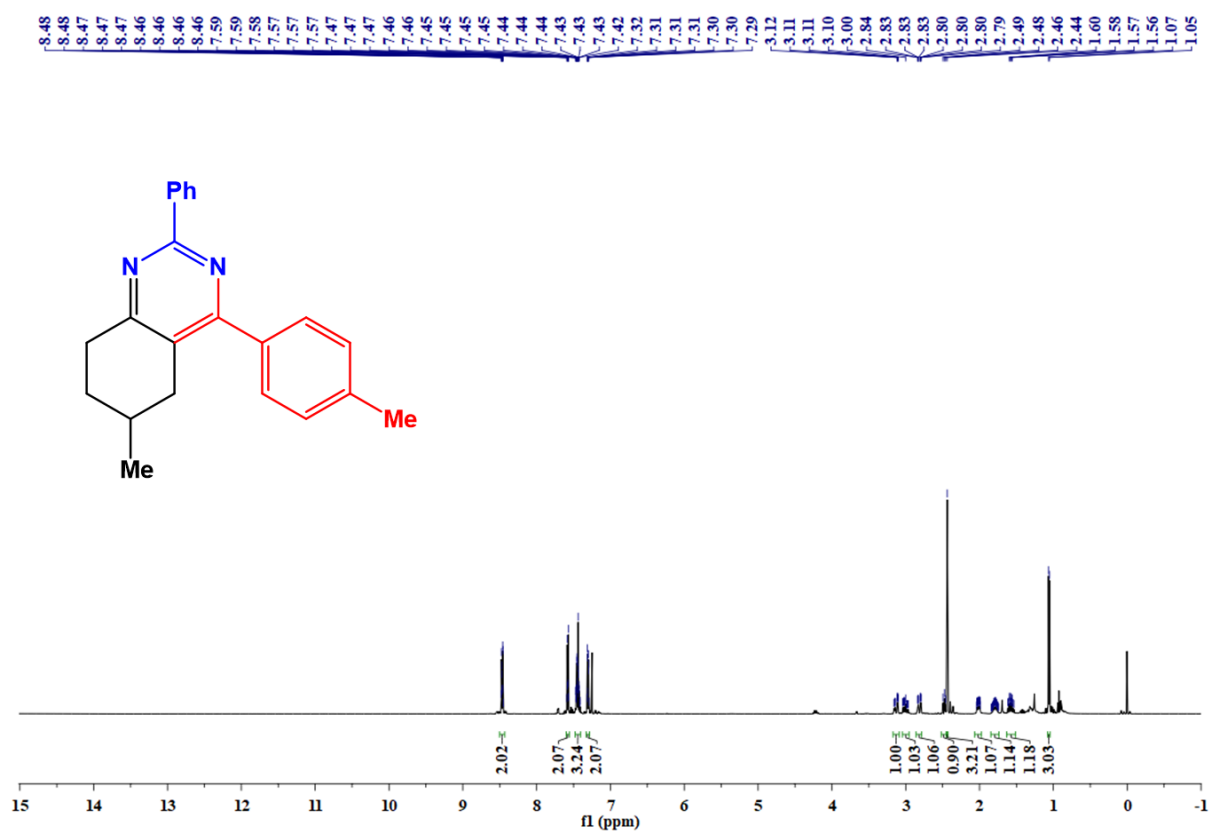
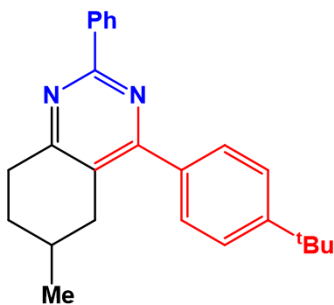
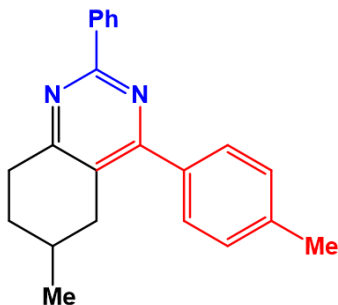


Figure S151: ^1H NMR spectrum of **8ai** in CDCl_3 at 500 MHz.



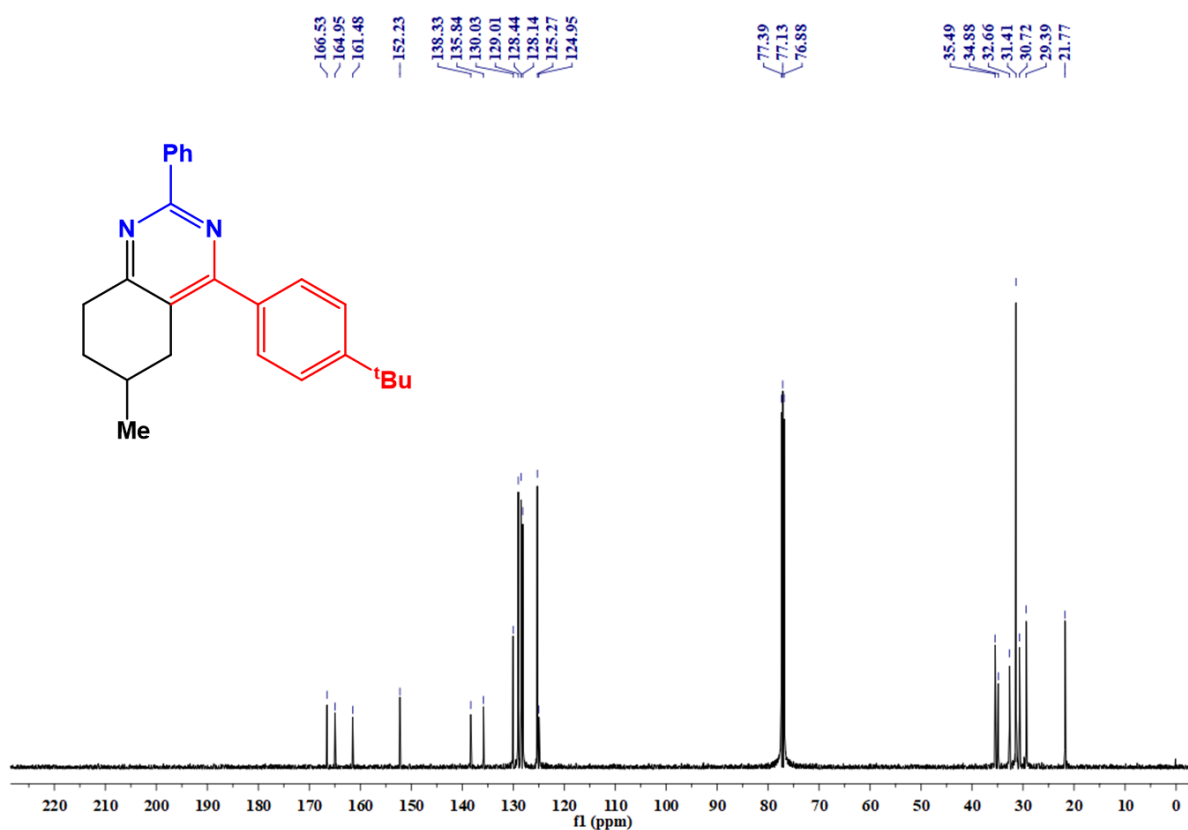


Figure S154: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8aj** in CDCl_3 at 126 MHz.

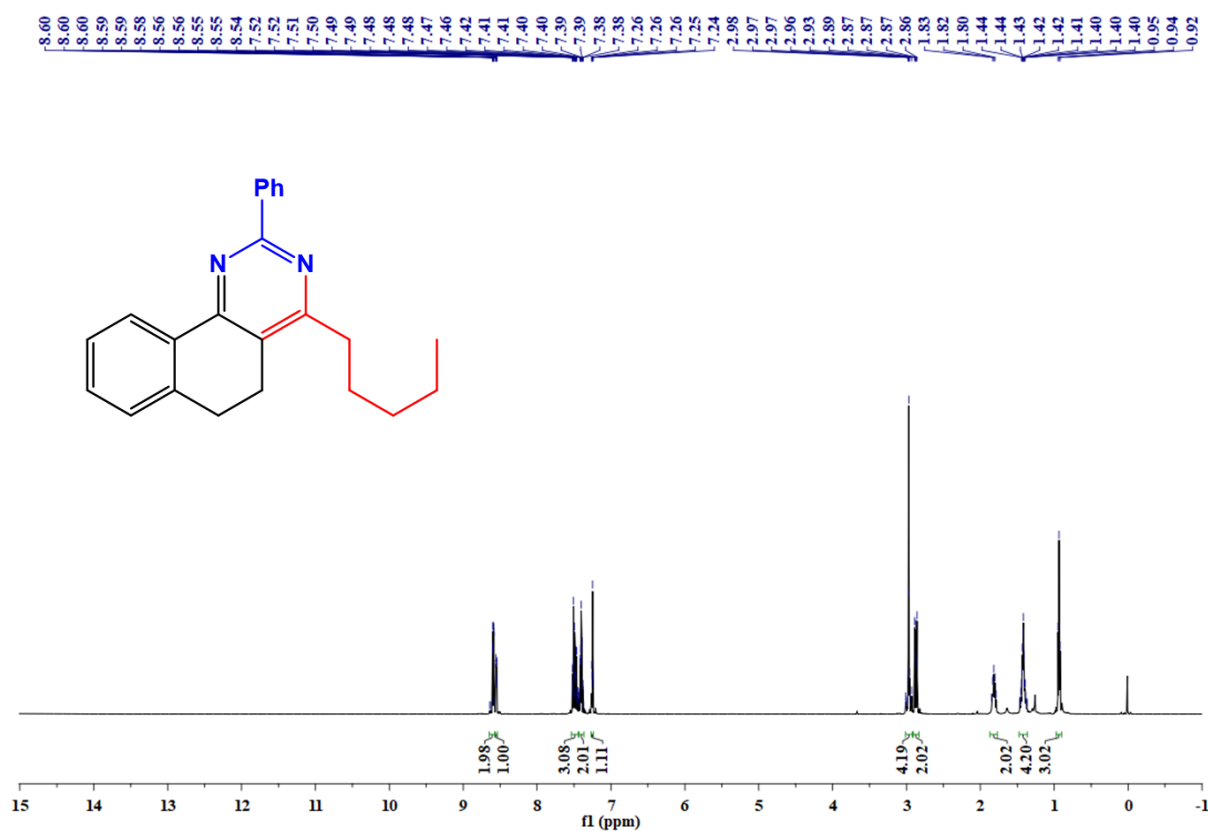


Figure S155: ^1H NMR spectrum of **8ak** in CDCl_3 at 500 MHz.

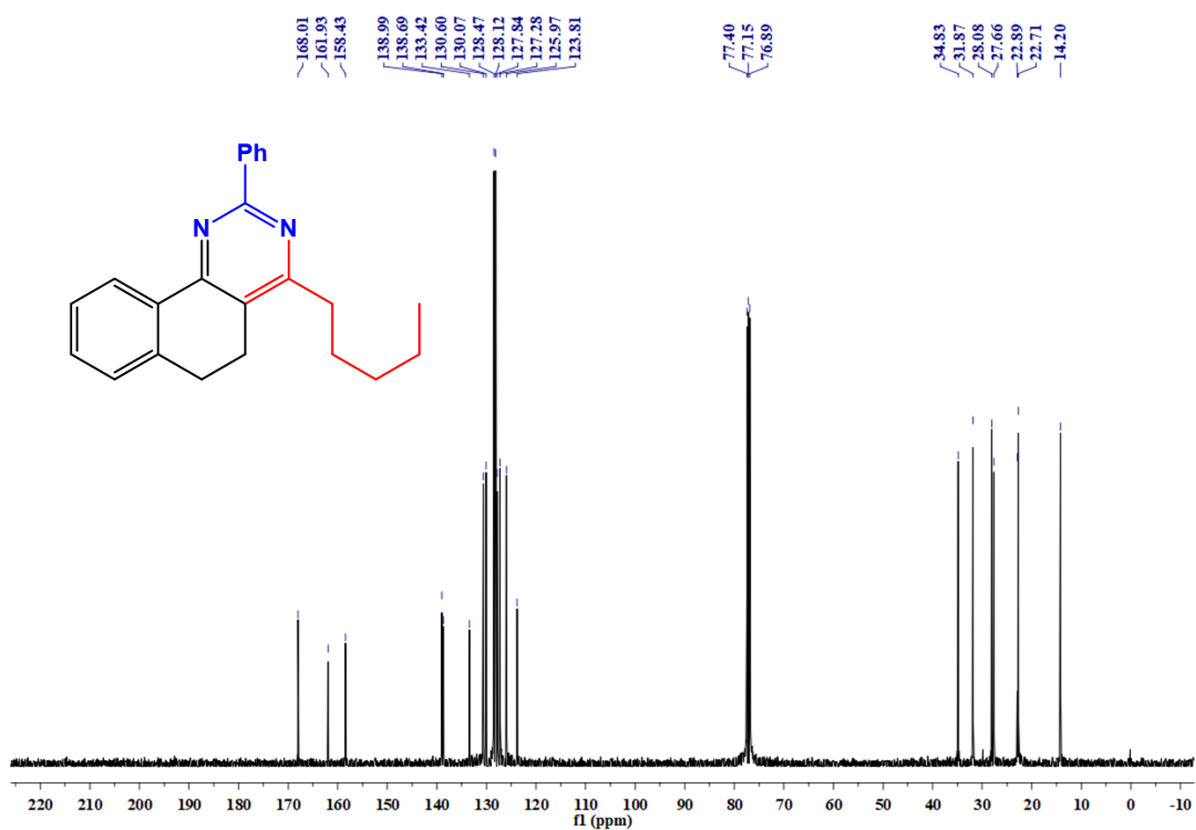


Figure S156: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8ak** in CDCl_3 at 126 MHz.

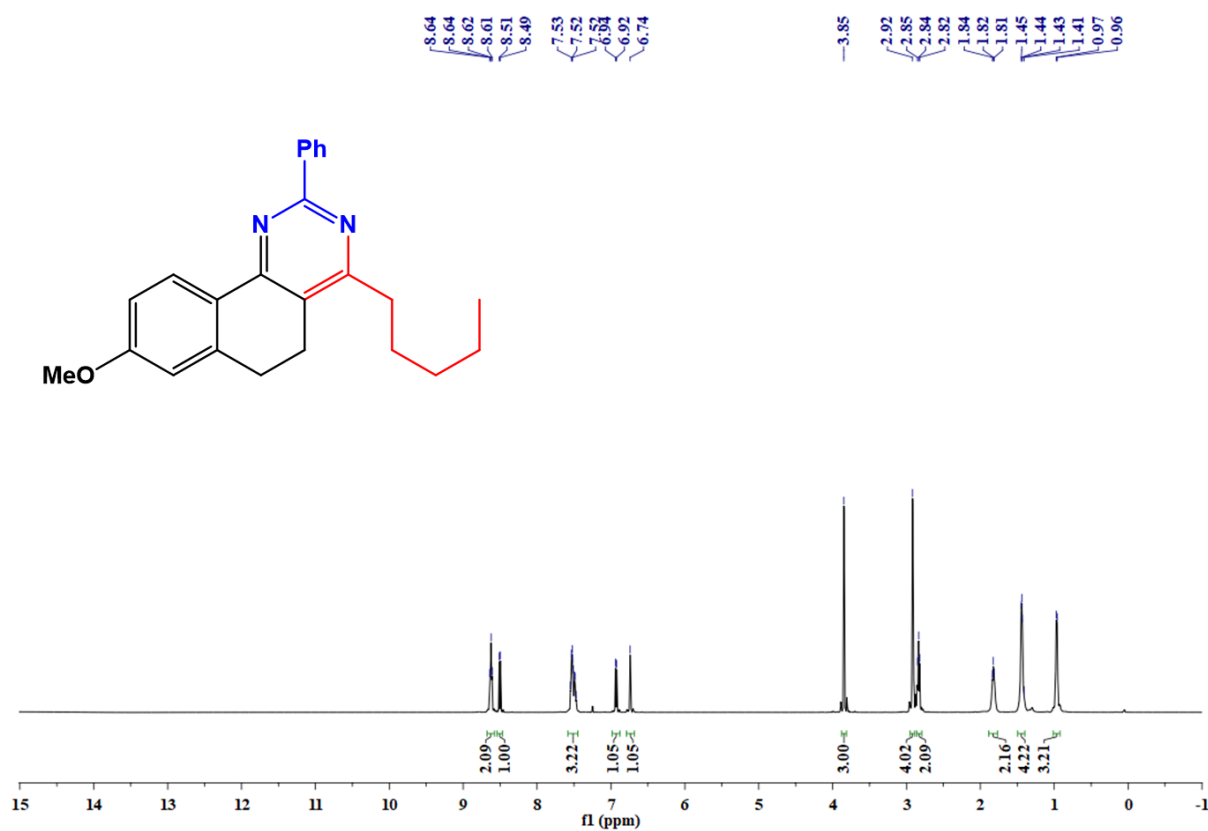


Figure S157: ^1H NMR spectrum of **8al** in CDCl_3 at 500 MHz.

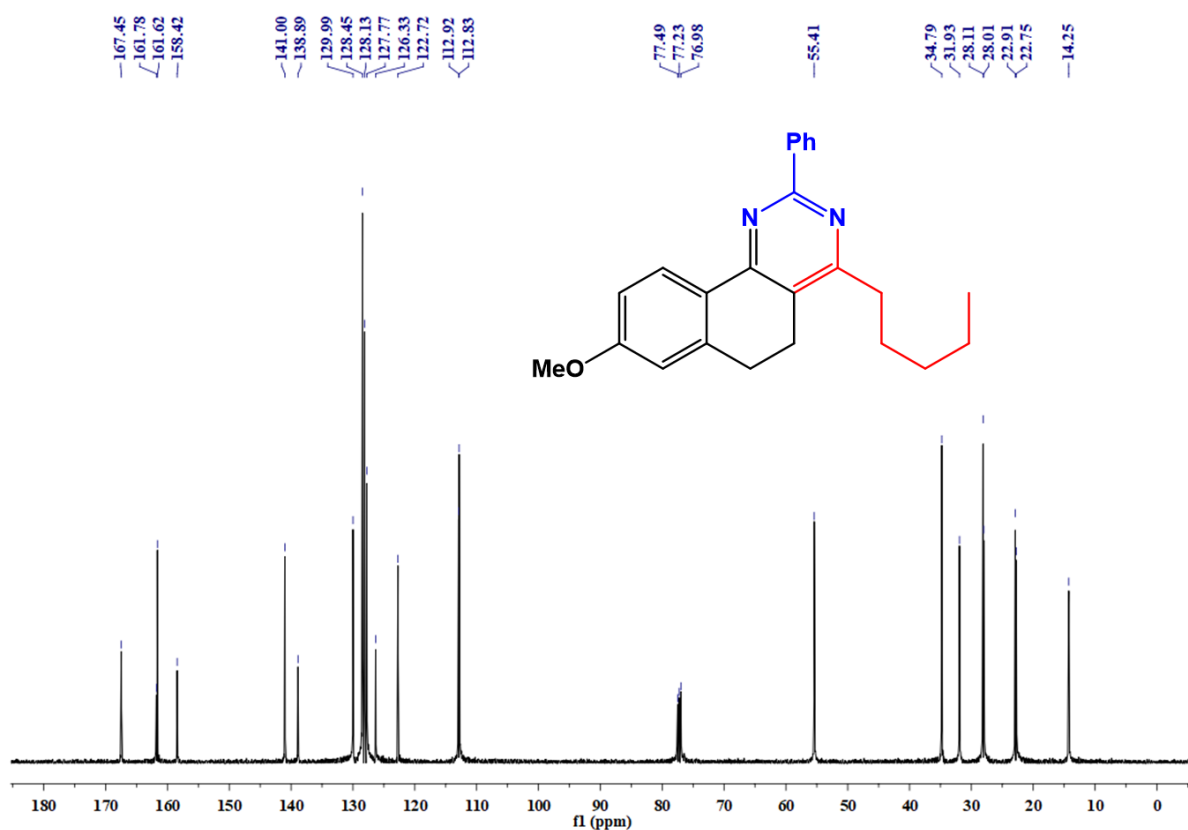


Figure S158: ¹³C{¹H} NMR spectrum of **8al** in CDCl₃ at 126 MHz.

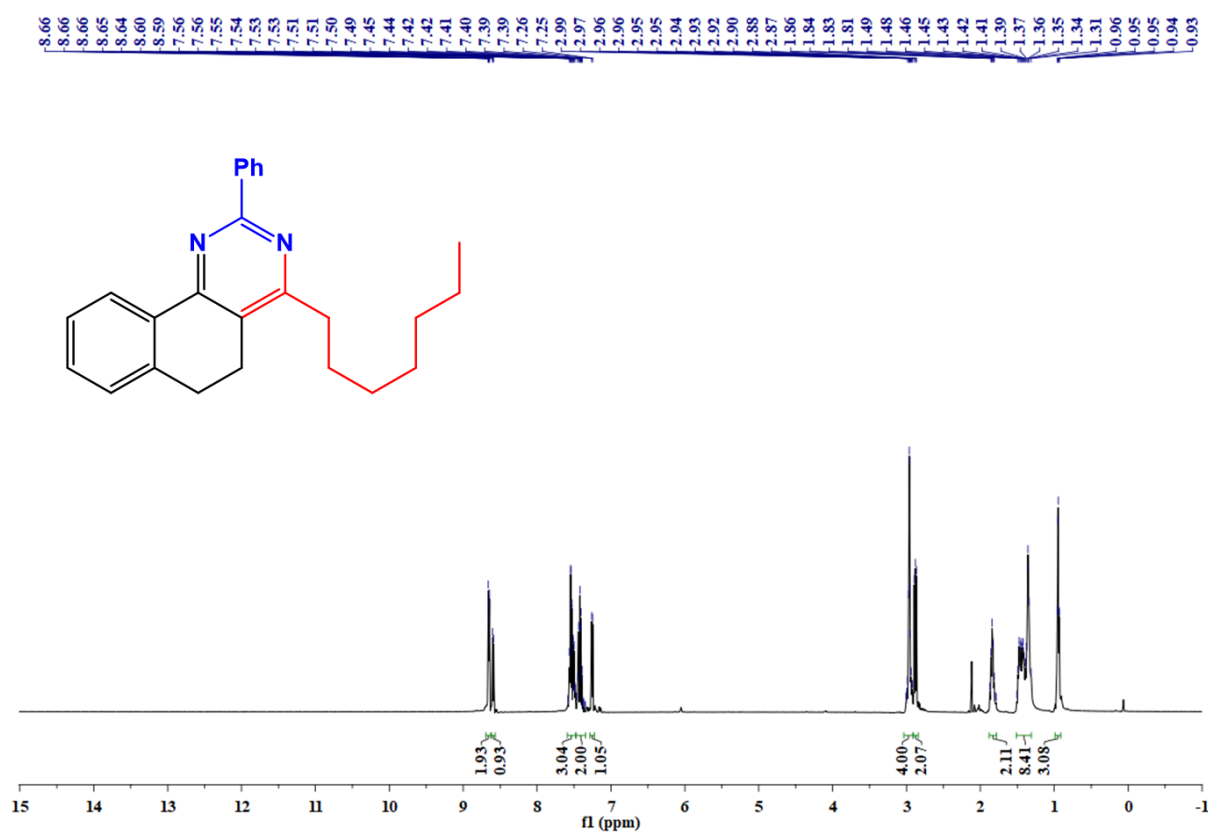


Figure S159: ¹H NMR spectrum of **8am** in CDCl₃ at 500 MHz.

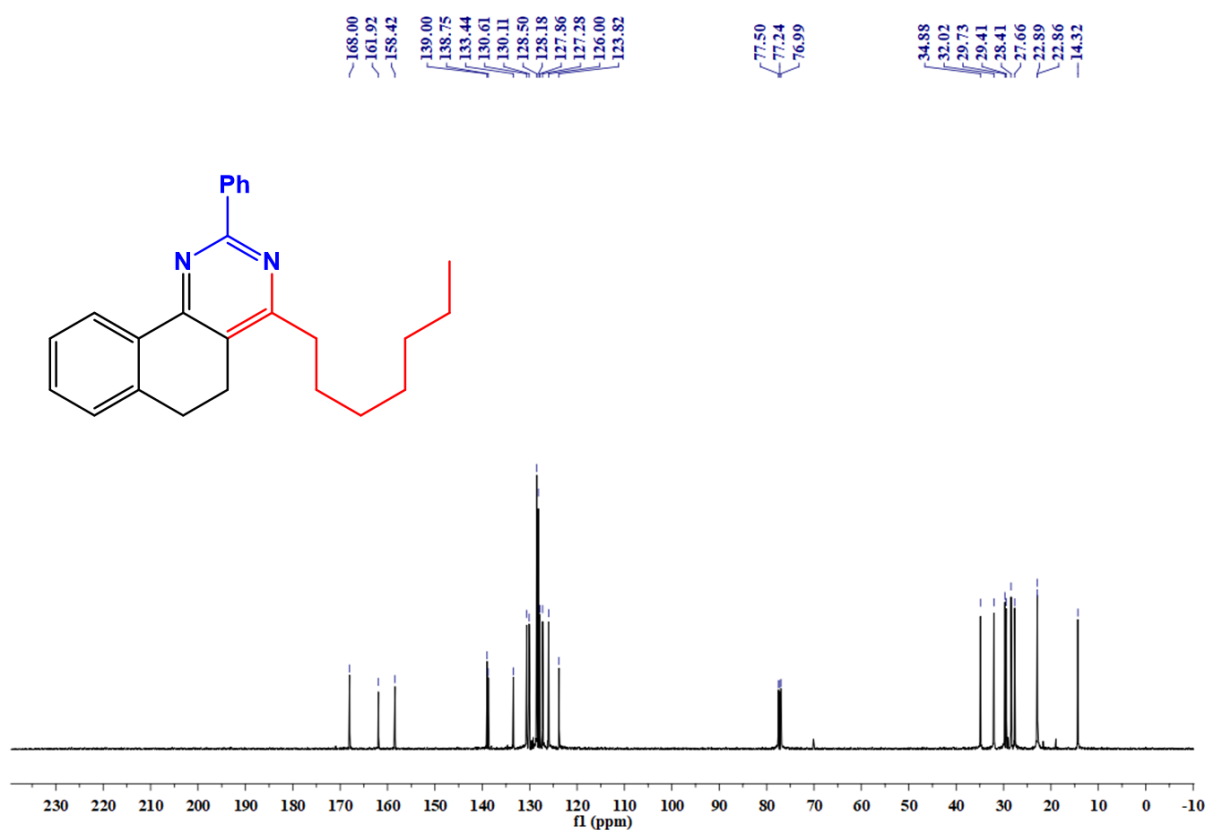


Figure S160: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8am** in CDCl_3 at 126 MHz.

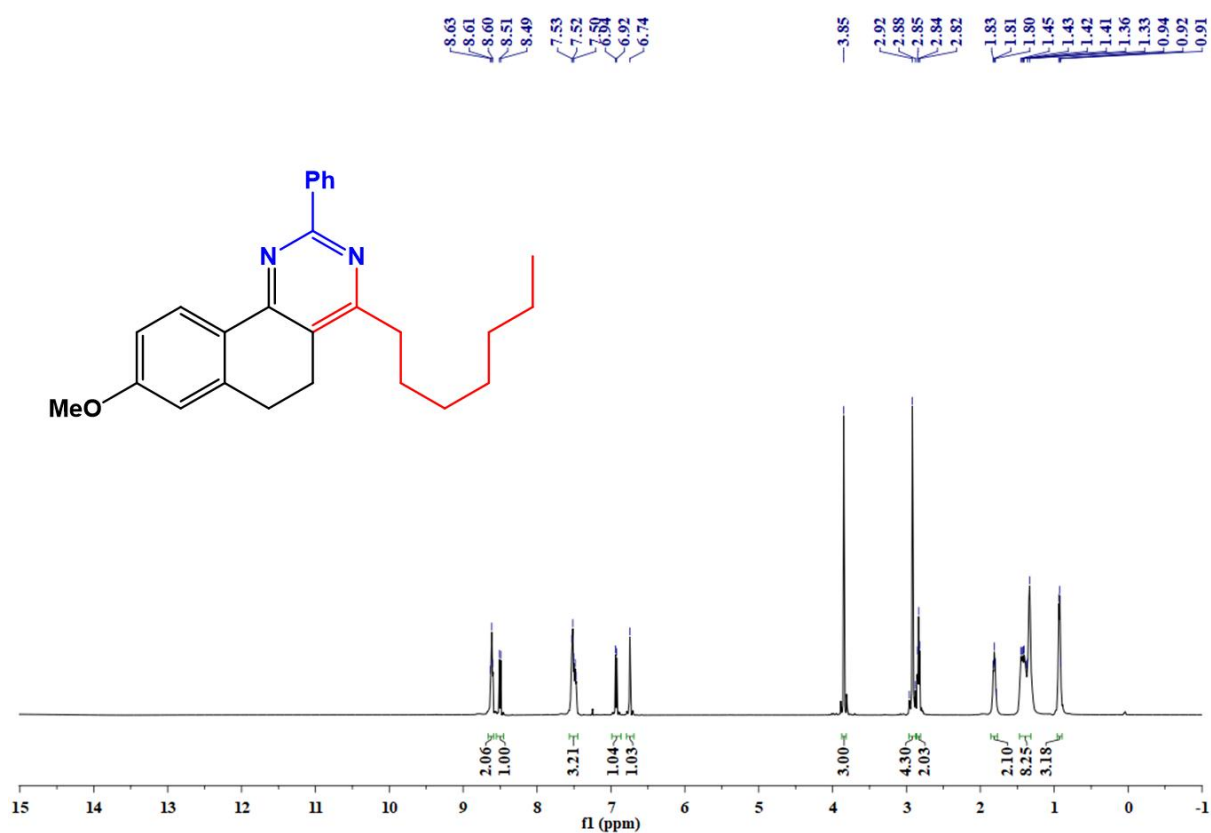


Figure S161: ^1H NMR spectrum of **8an** in CDCl_3 at 500 MHz.

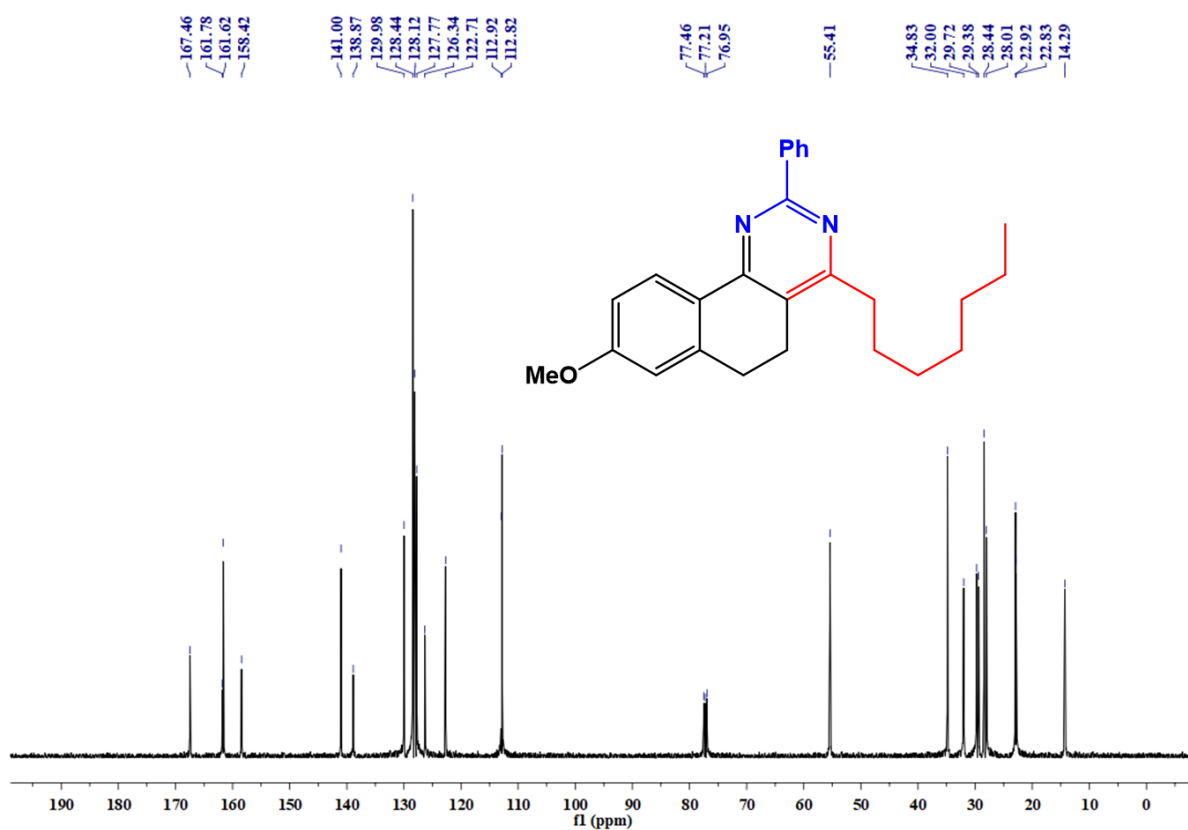


Figure S162: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8an** in CDCl_3 at 126 MHz.

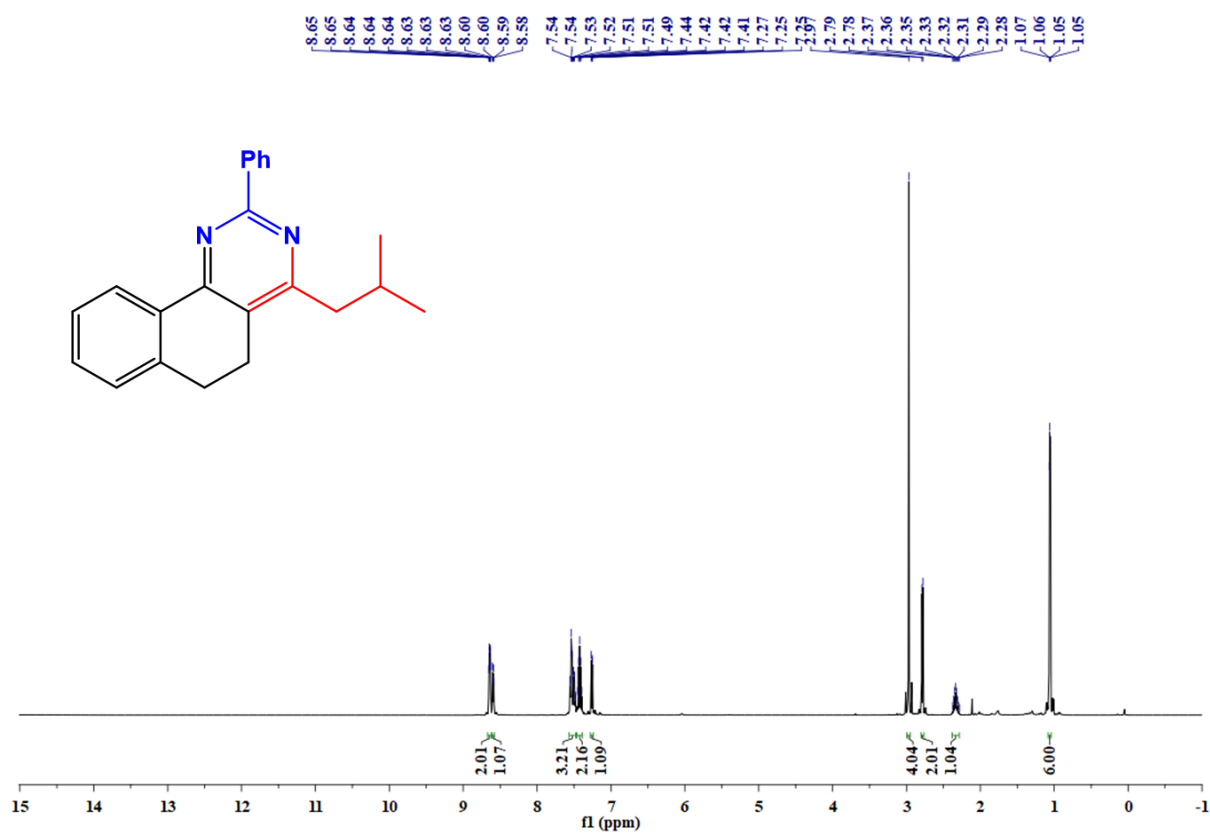


Figure S163: ^1H NMR spectrum of **8ao** in CDCl_3 at 500 MHz.

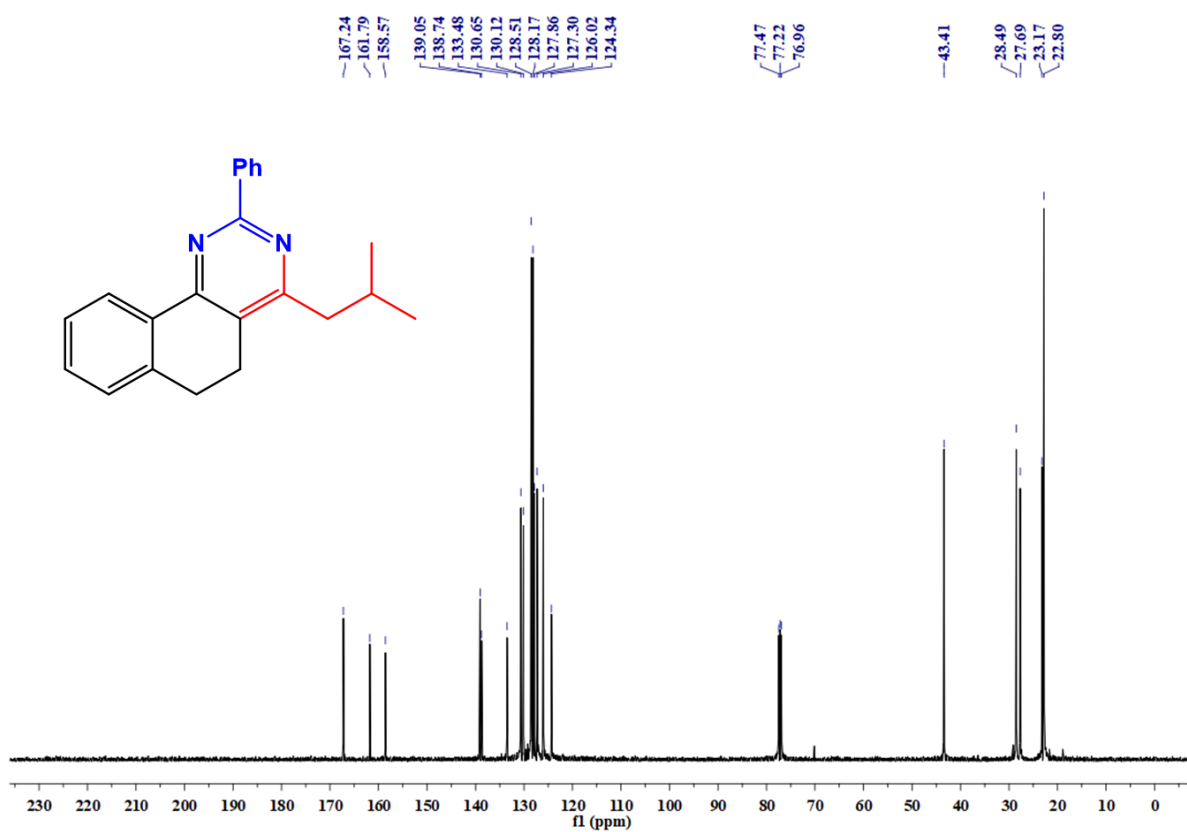


Figure S164: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **8ao** in CDCl_3 at 126 MHz.

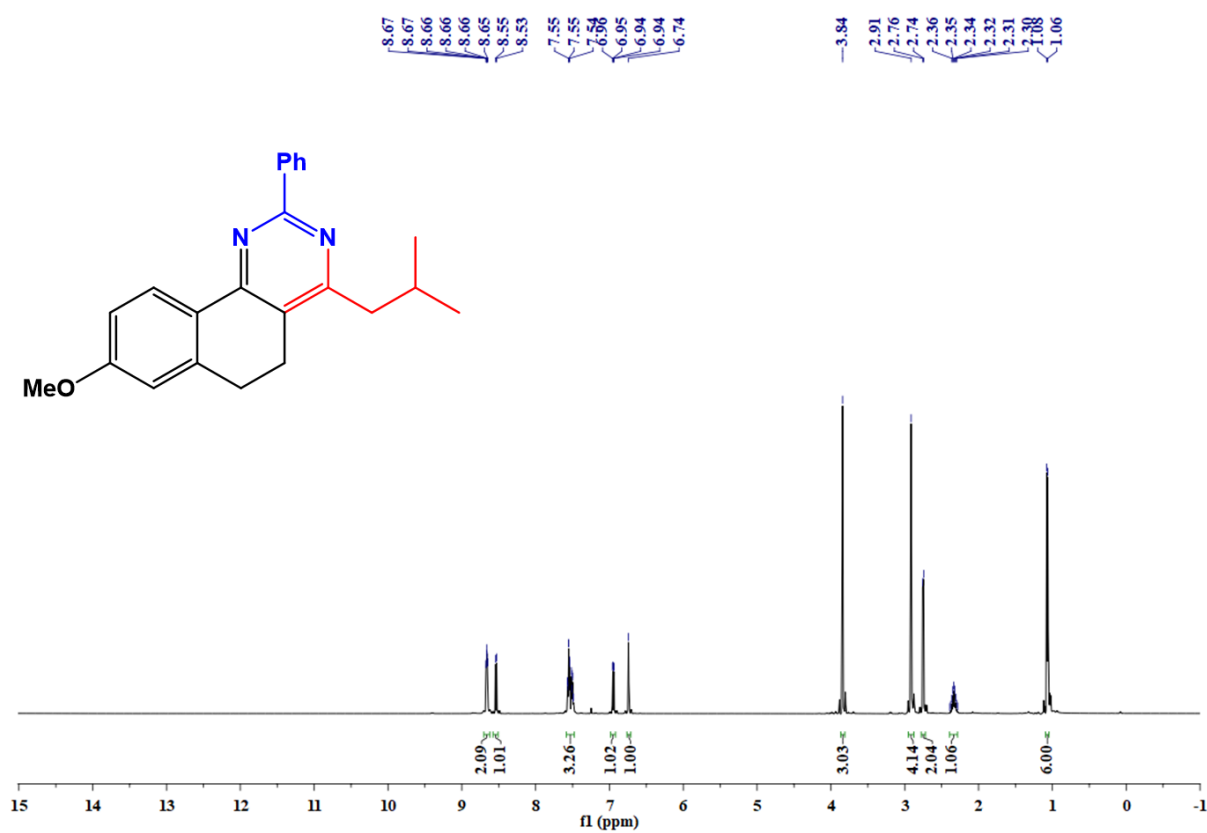


Figure S165: ^1H NMR spectrum of **8ap** in CDCl_3 at 500 MHz.

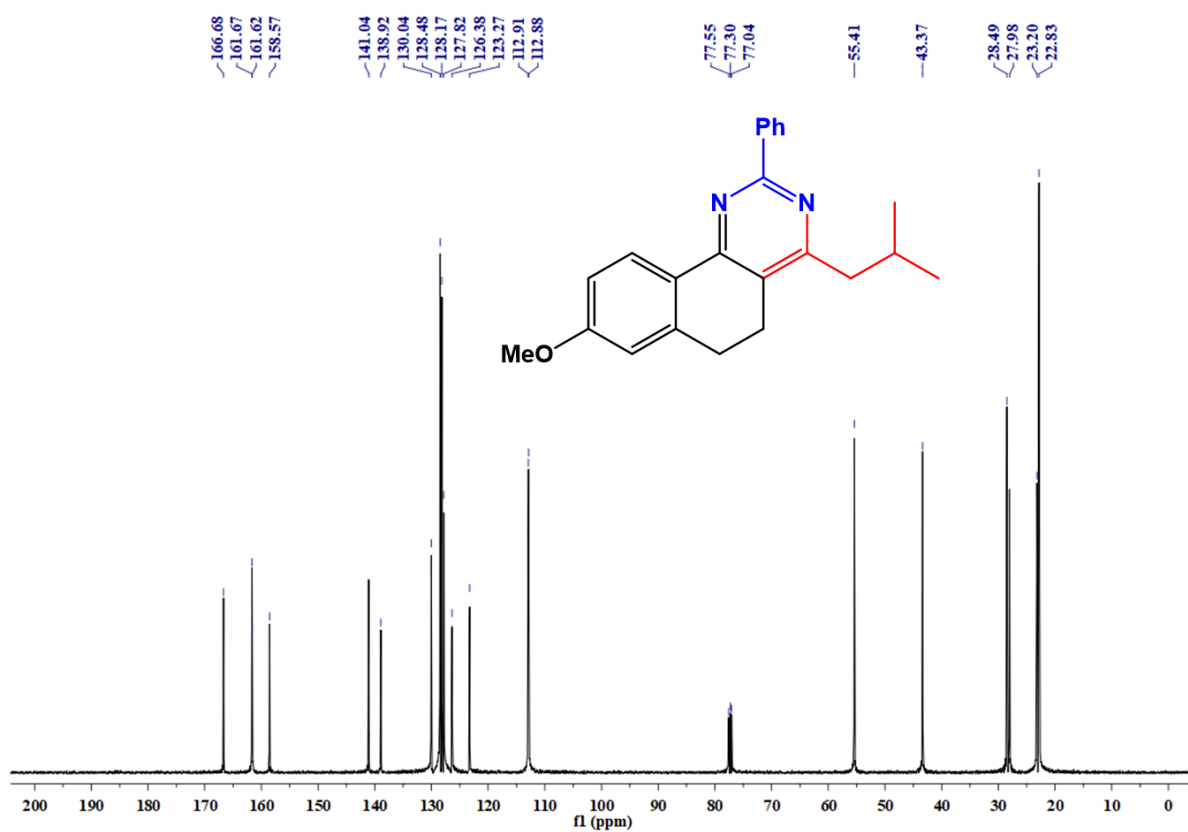


Figure S166: ¹³C{¹H} NMR spectrum of **8ap** in CDCl₃ at 126 MHz.

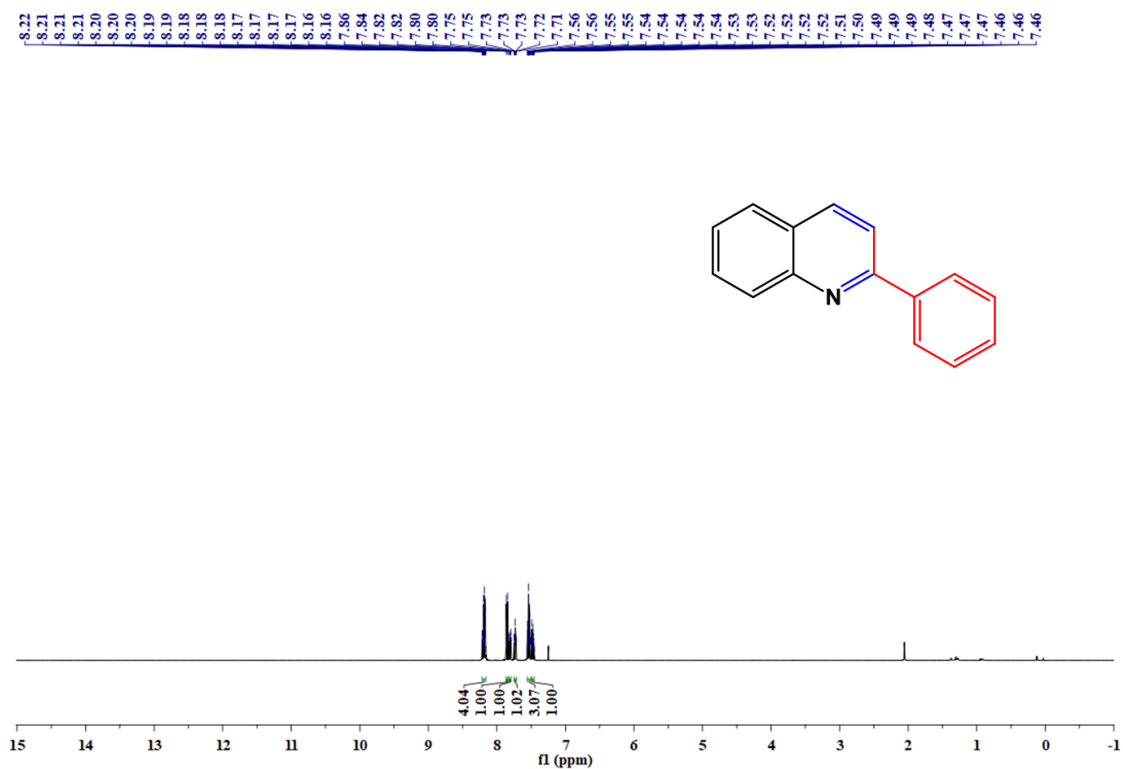


Figure S167: ¹H NMR spectrum of **11a** in CDCl₃ at 500 MHz.

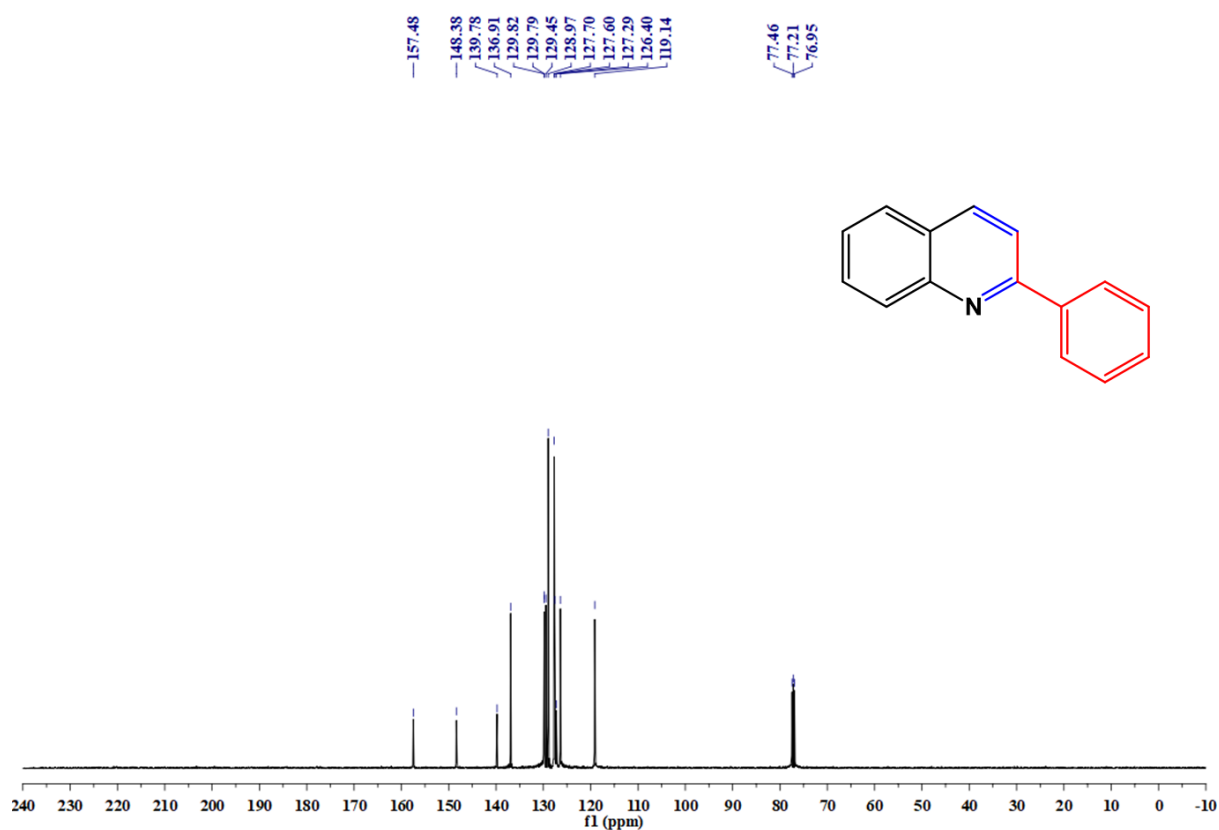


Figure S168: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11a** in CDCl_3 at 126 MHz.

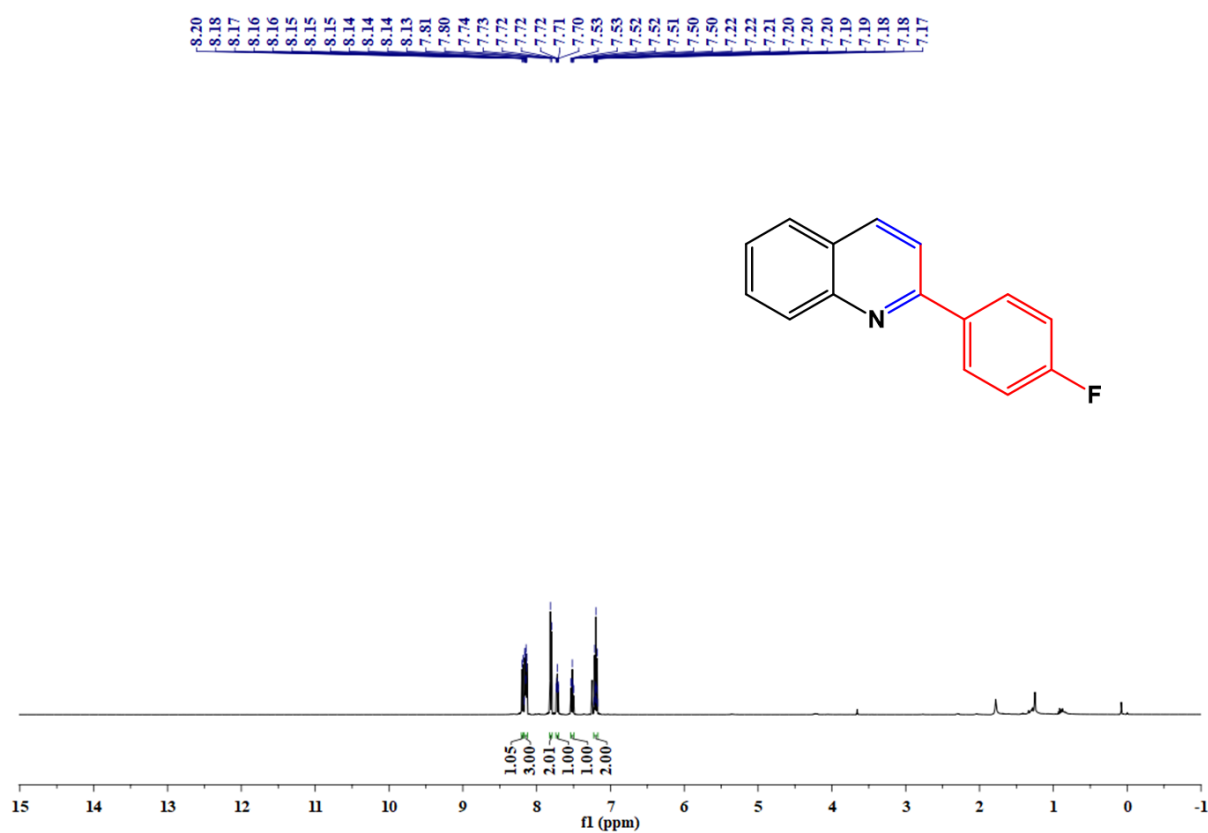


Figure S169: ^1H NMR spectrum of **11b** in CDCl_3 at 500 MHz.

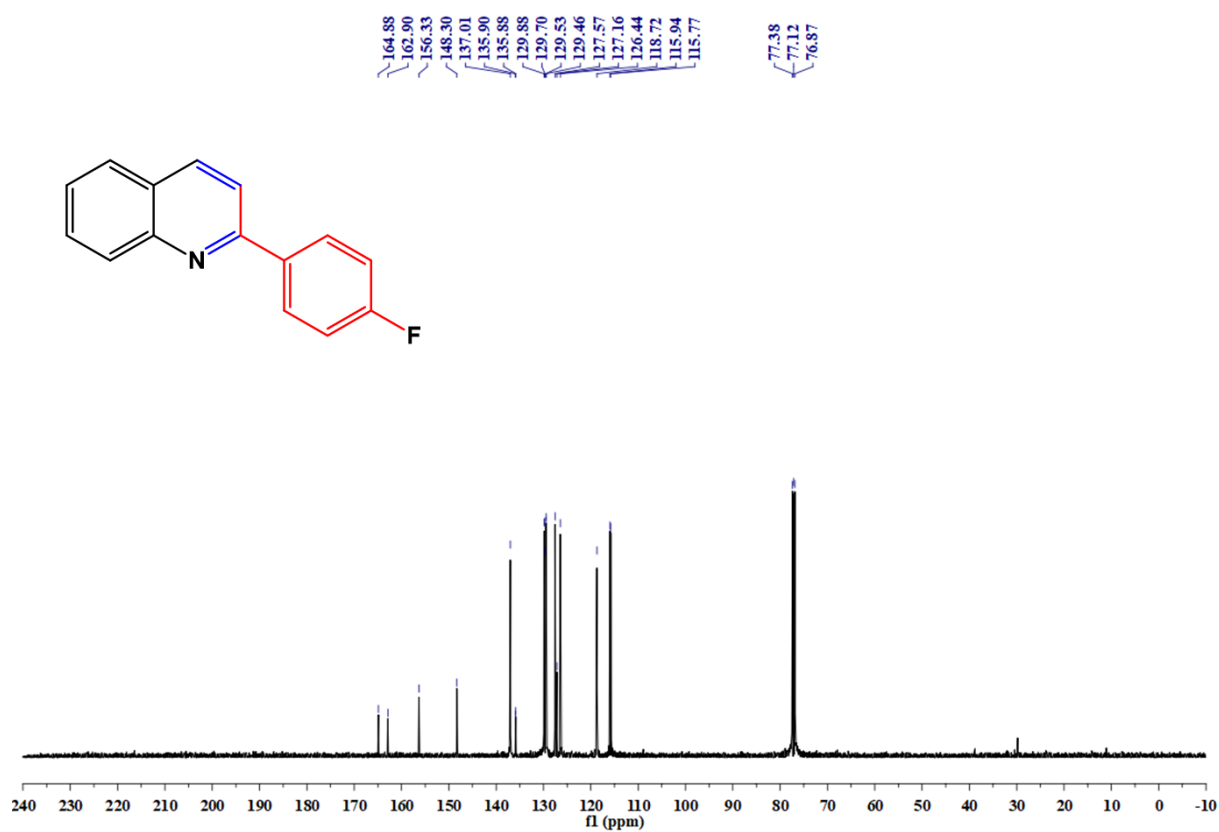


Figure S170: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11b** in CDCl_3 at 126 MHz.

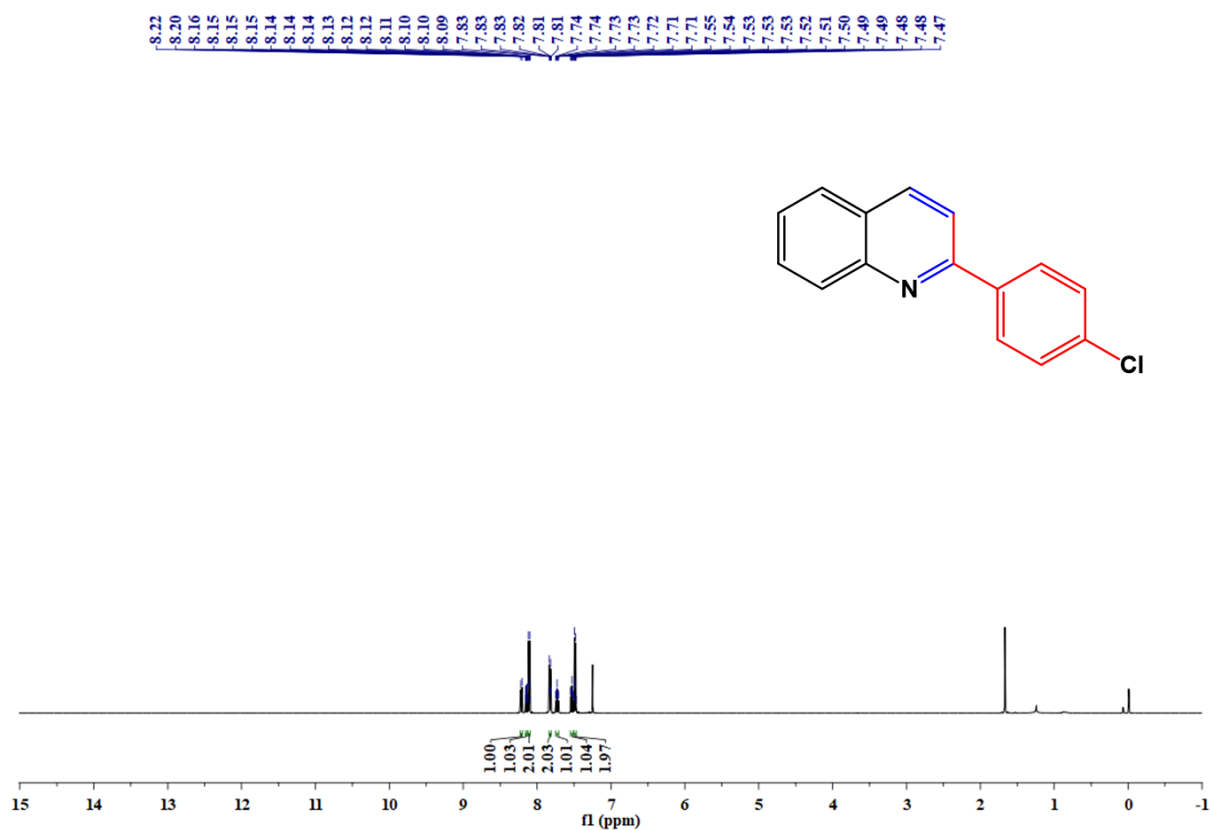


Figure S171: ^1H NMR spectrum of **11c** in CDCl_3 at 500 MHz.

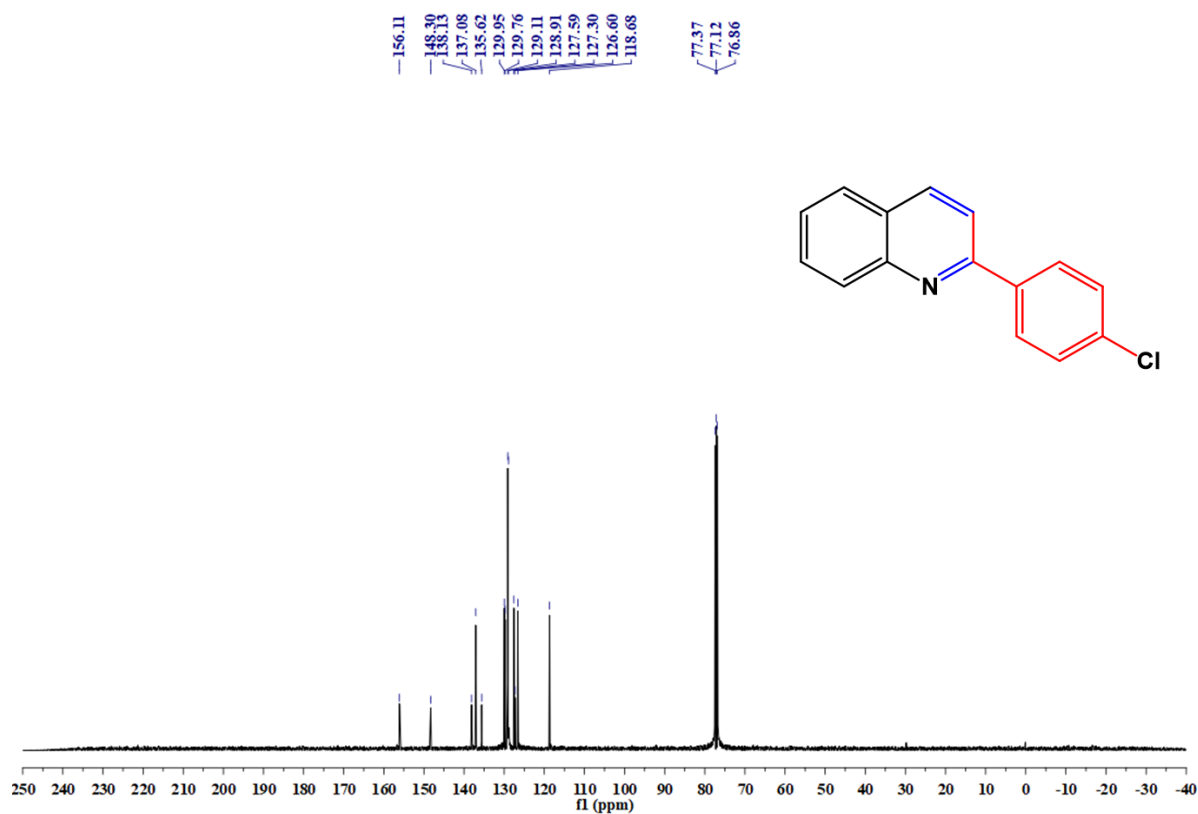


Figure S172: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11c** in CDCl_3 at 126 MHz.

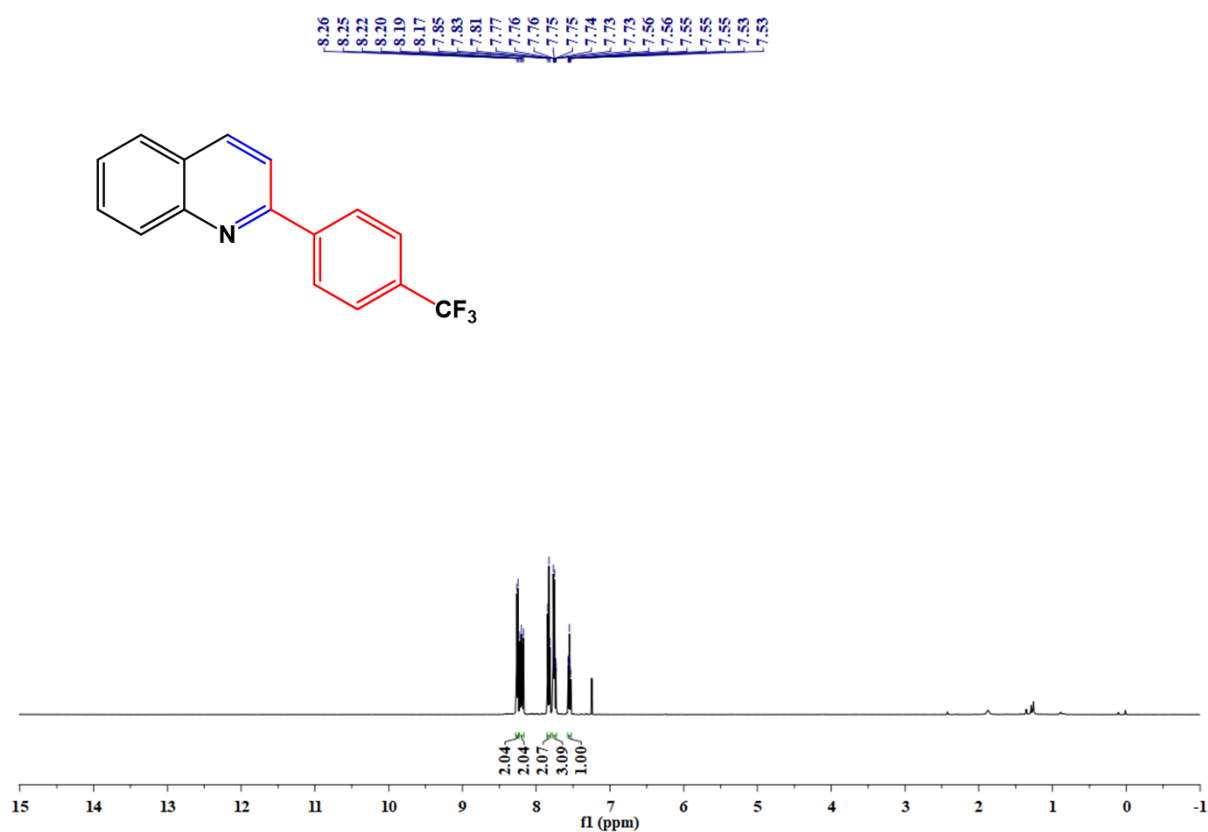


Figure S173: ^1H NMR spectrum of **11d** in CDCl_3 at 500 MHz.

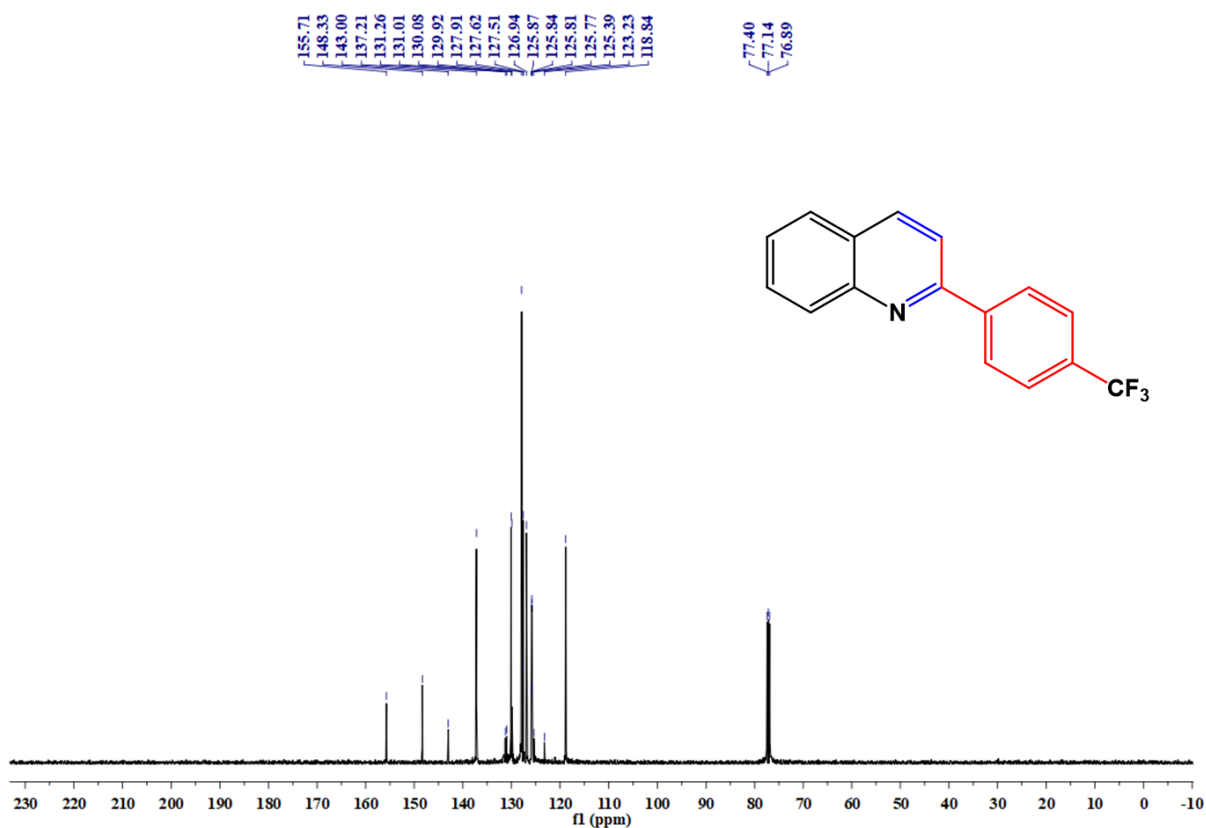


Figure S174: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11d** in CDCl_3 at 126 MHz.

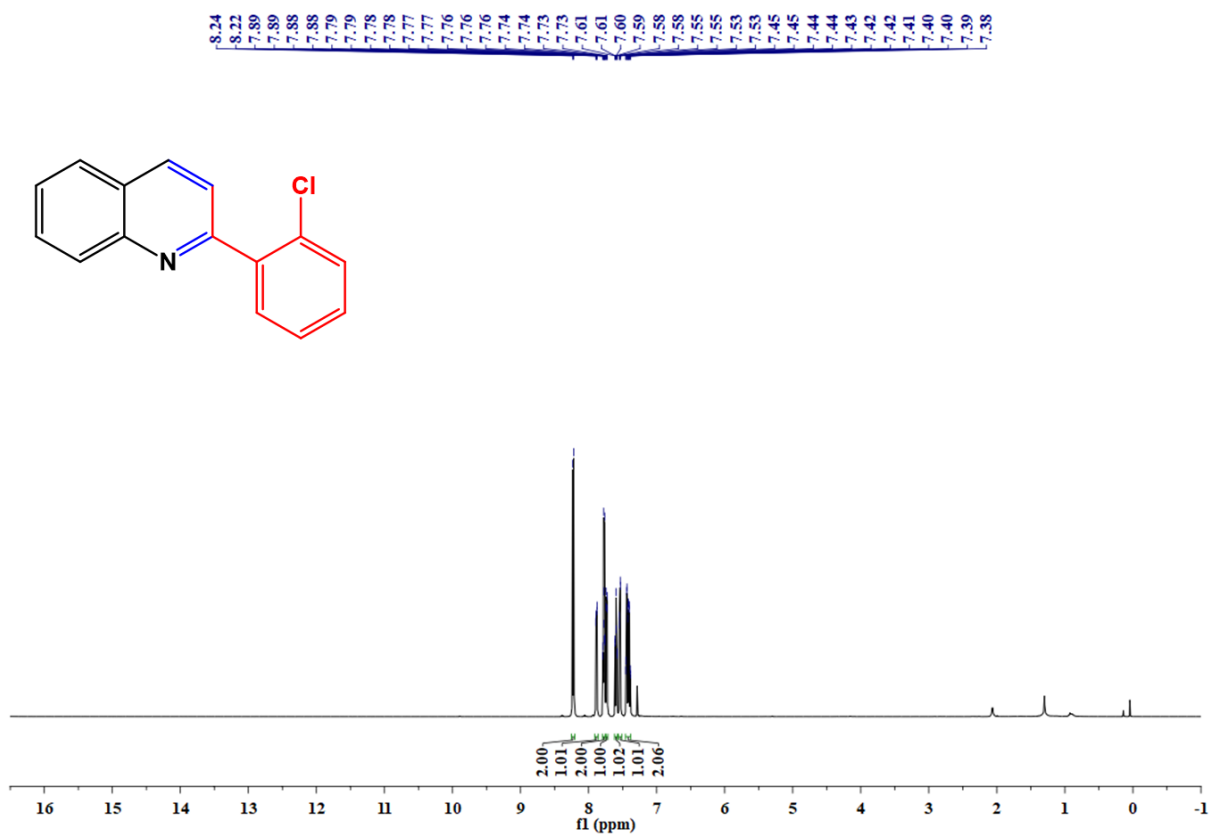


Figure S175: ^1H NMR spectrum of **11e** in CDCl_3 at 500 MHz.

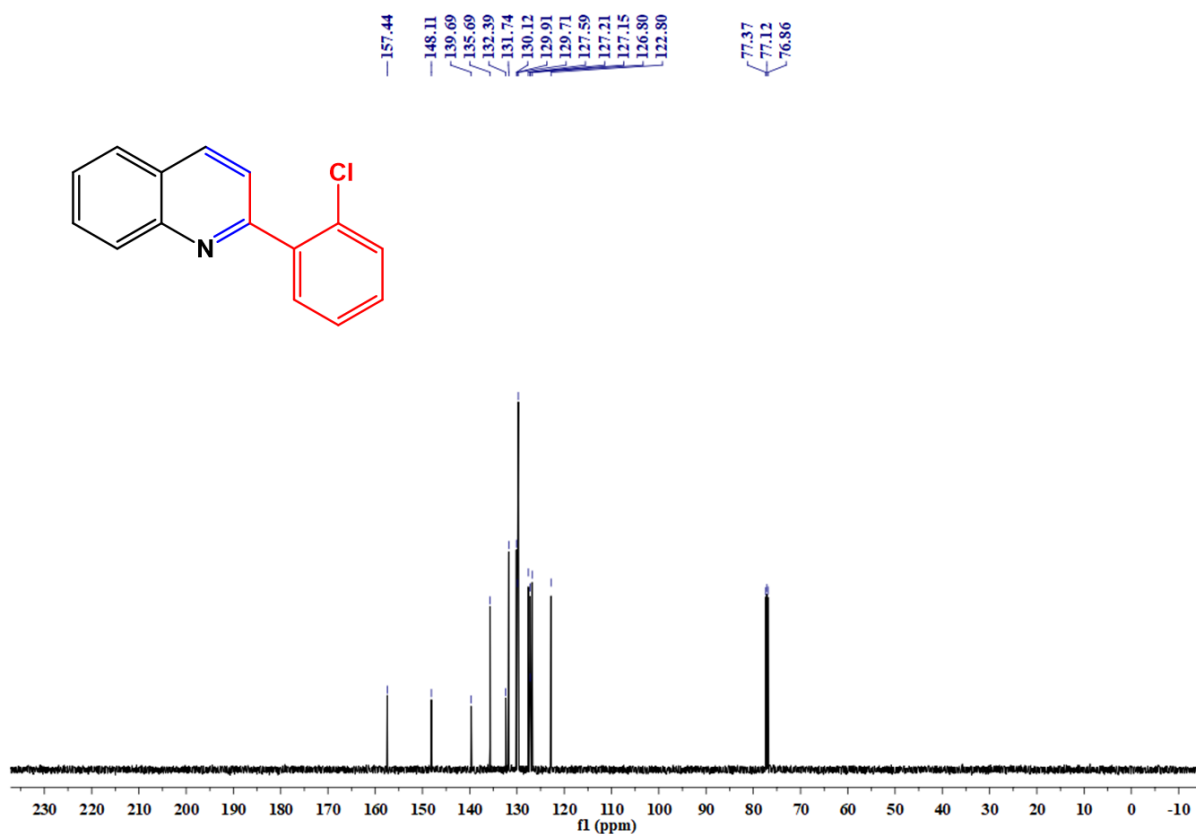


Figure S176: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11e** in CDCl_3 at 126 MHz.

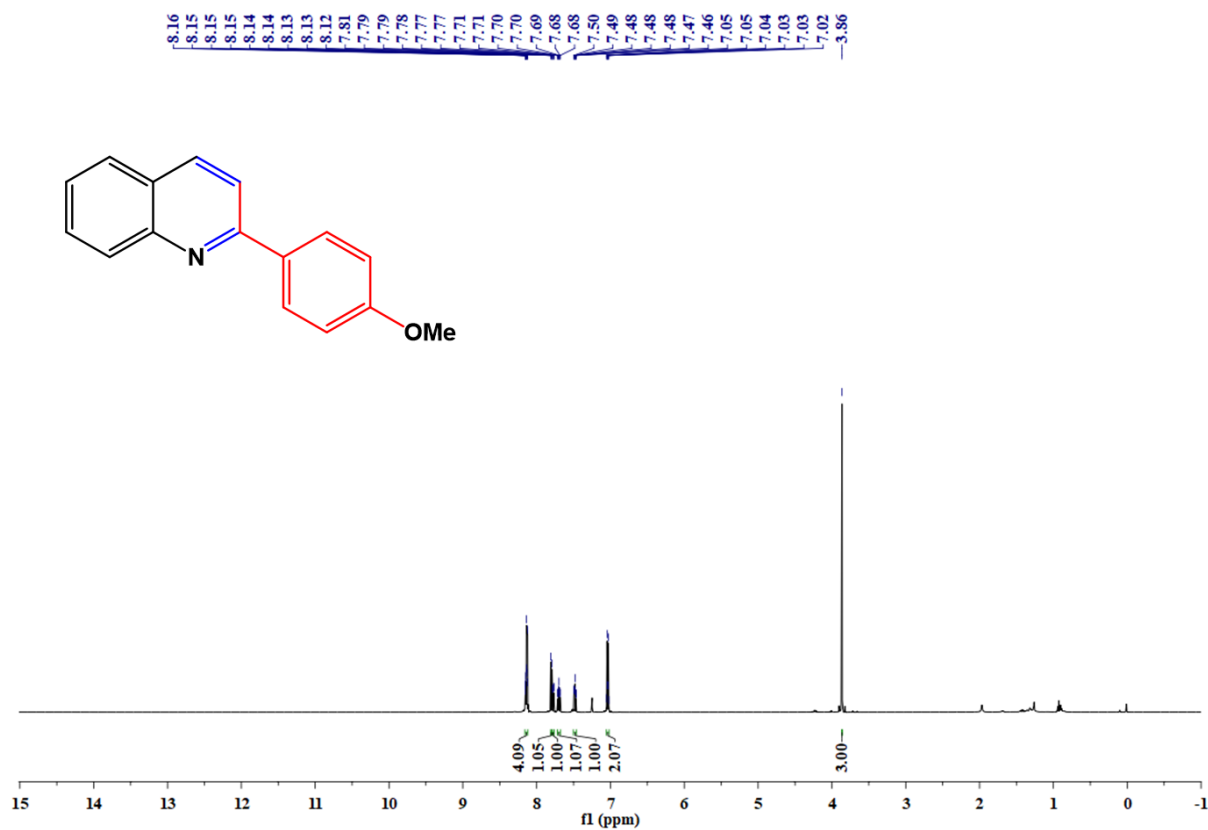


Figure S177: ^1H NMR spectrum of **11f** in CDCl_3 at 500 MHz.

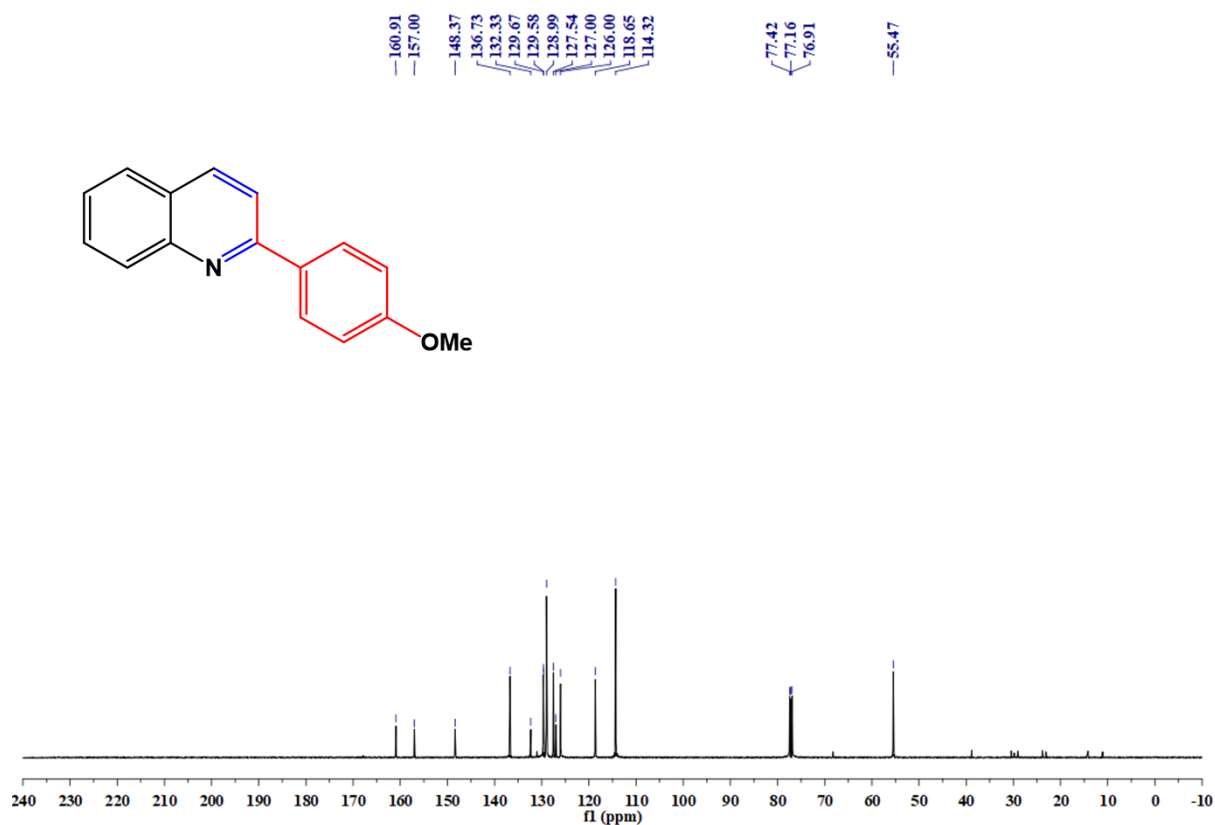


Figure S178: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11f** in CDCl_3 at 126 MHz.

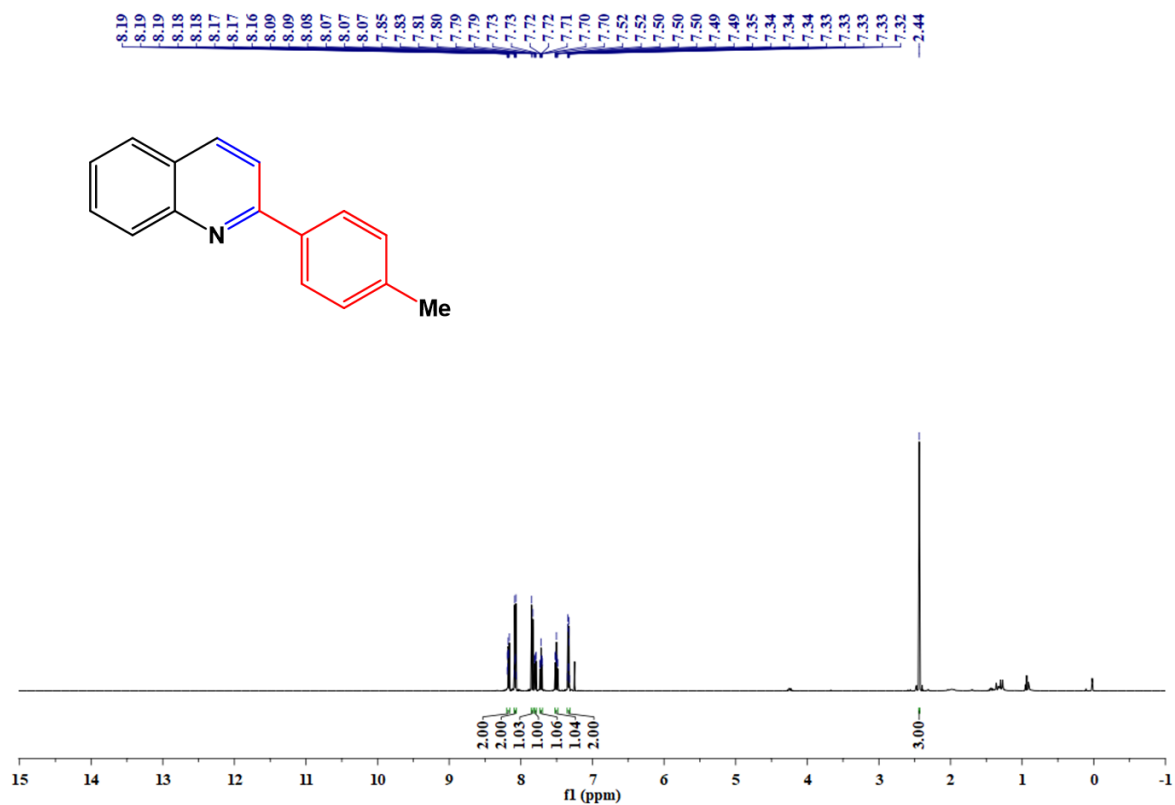


Figure S179: ^1H NMR spectrum of **11g** in CDCl_3 at 500 MHz.

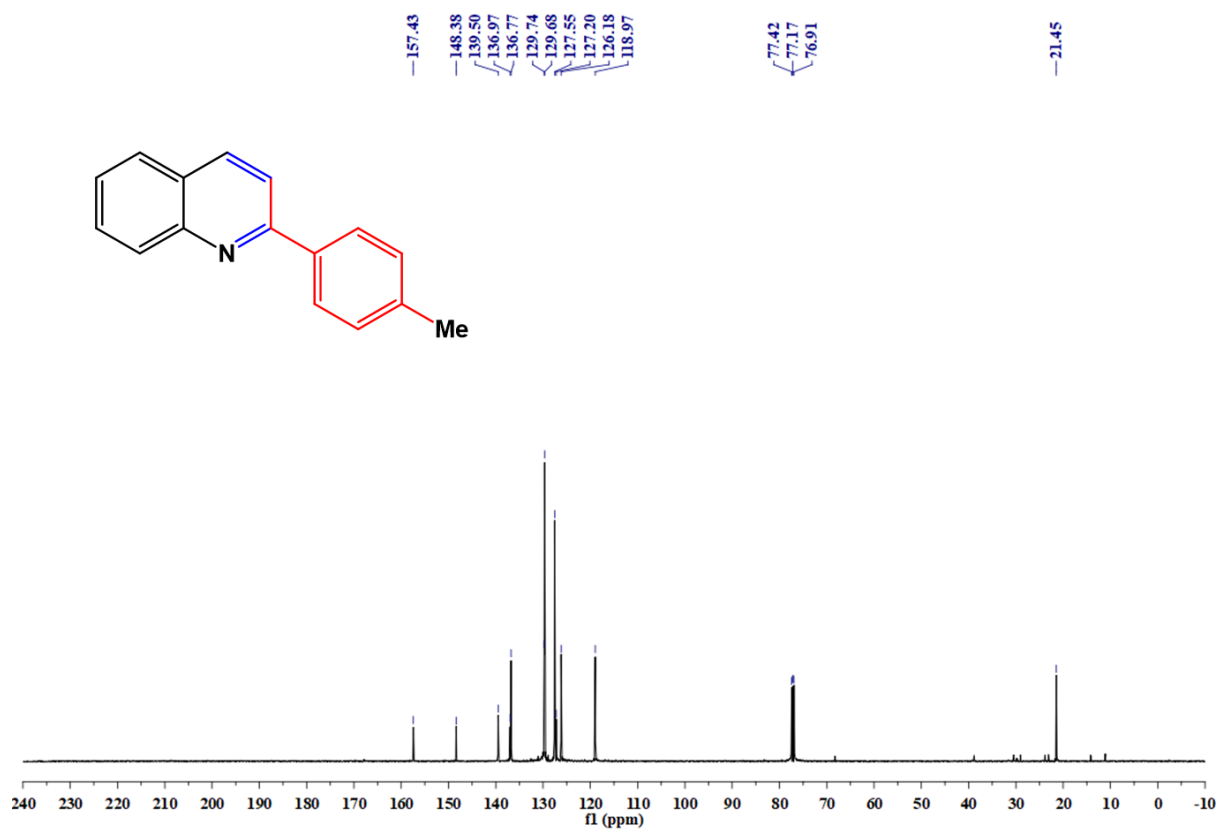


Figure S180: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11g** in CDCl_3 at 126 MHz.

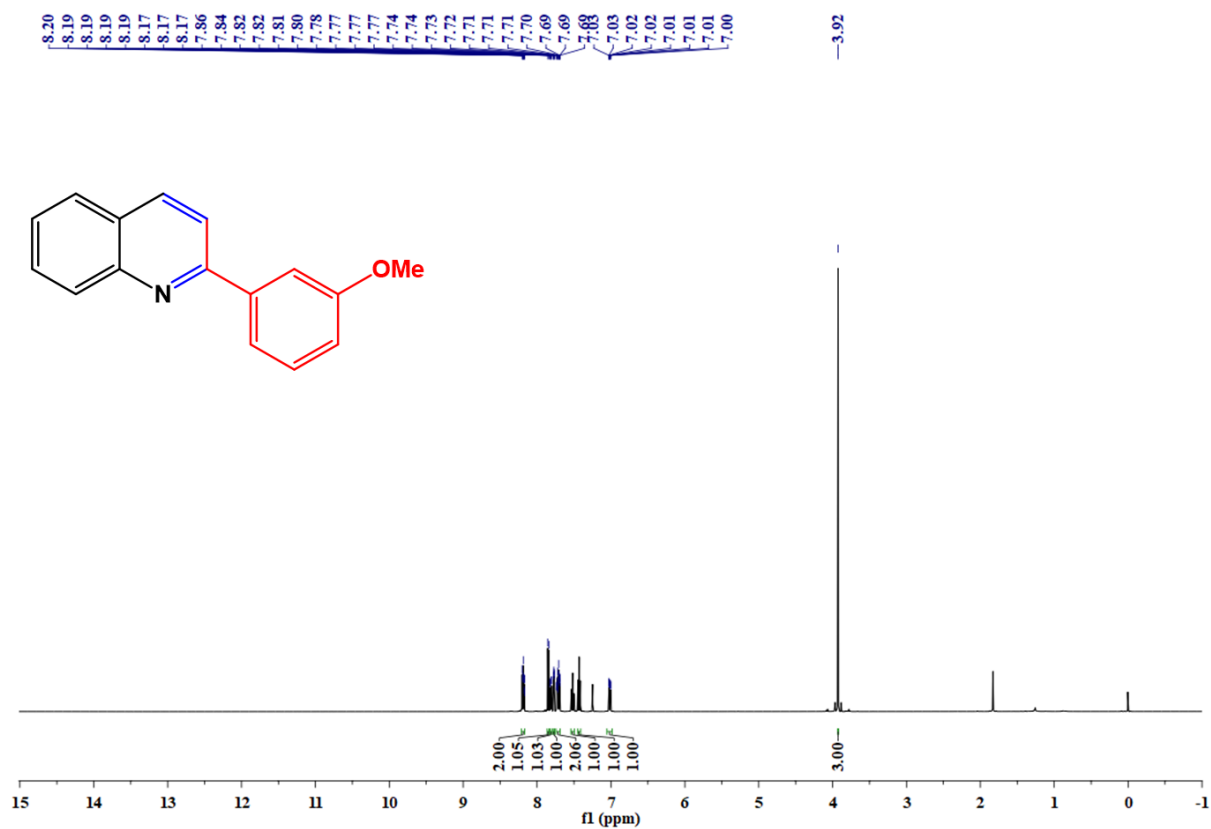


Figure S181: ^1H NMR spectrum of **11h** in CDCl_3 at 500 MHz.

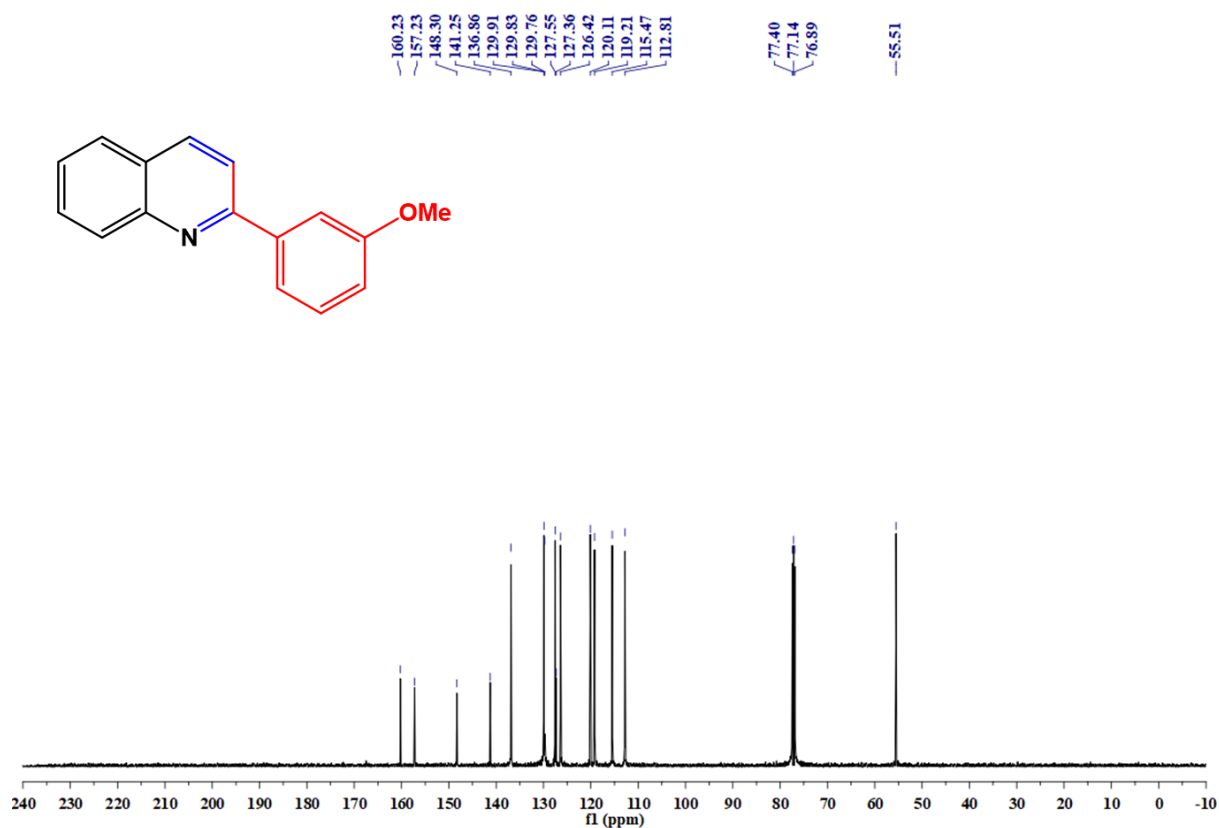


Figure S182: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11h** in CDCl_3 at 126 MHz.

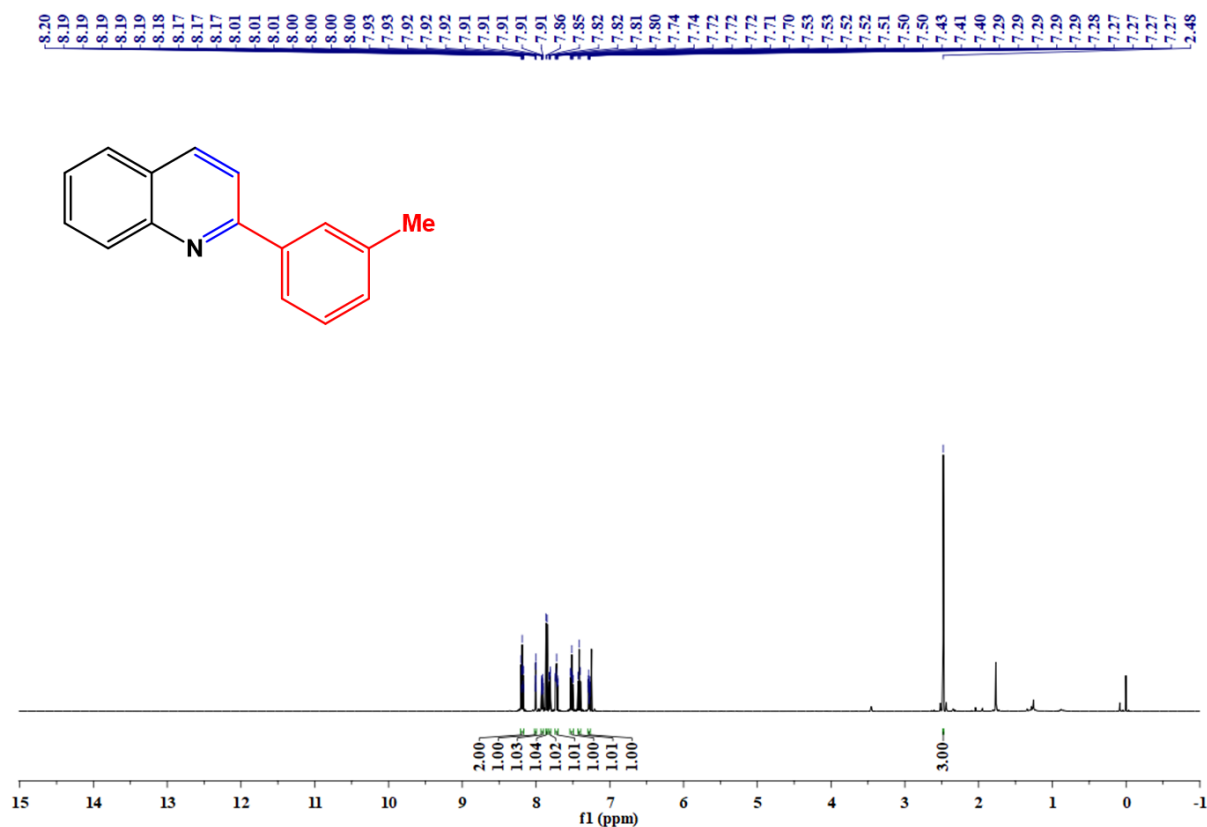


Figure S183: ^1H NMR spectrum of **11i** in CDCl_3 at 500 MHz.

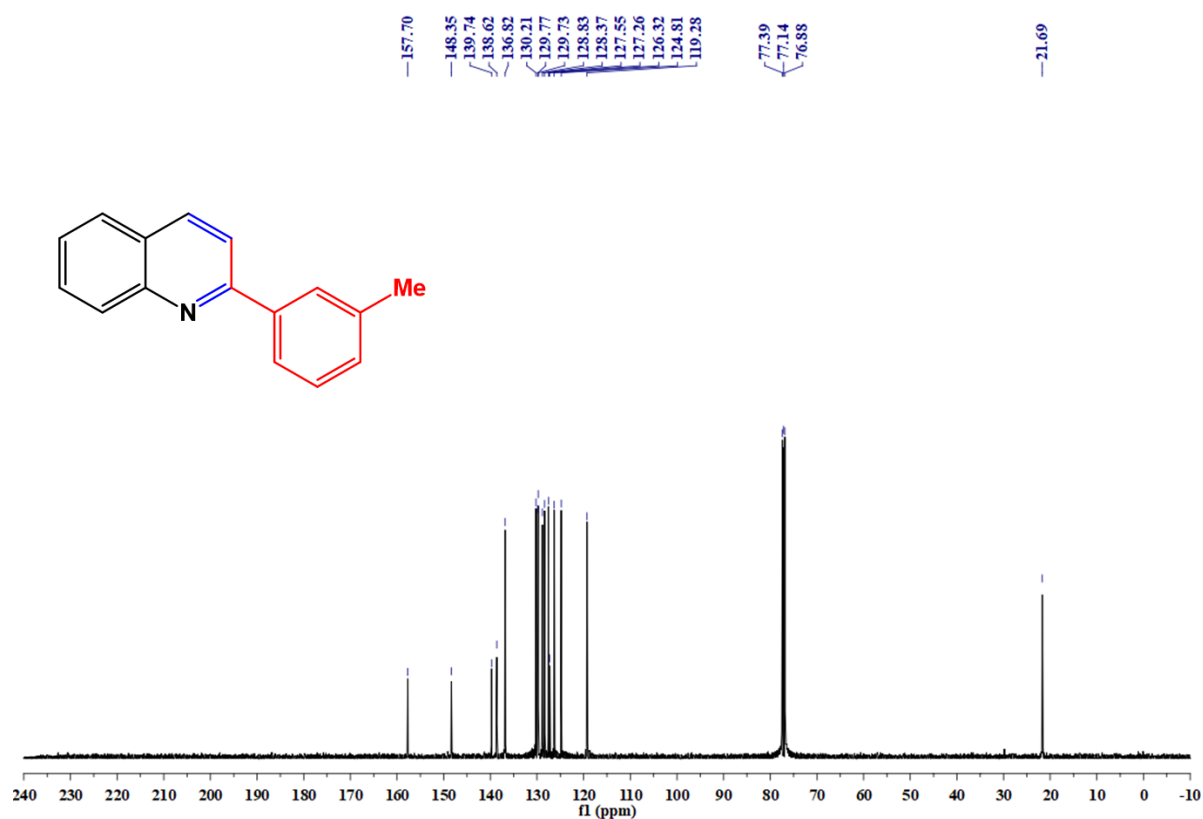


Figure S184: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11i** in CDCl_3 at 126 MHz.

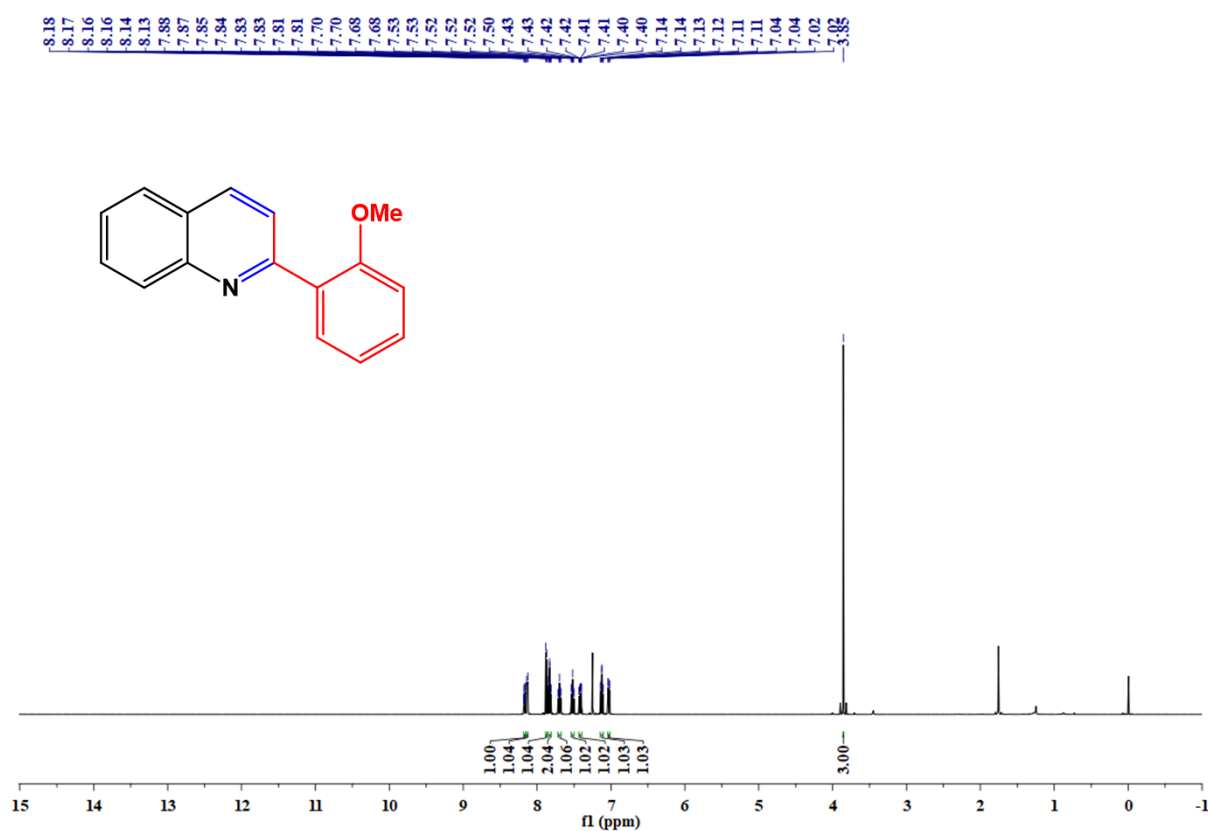


Figure S185: ^1H NMR spectrum of **11j** in CDCl_3 at 500 MHz.

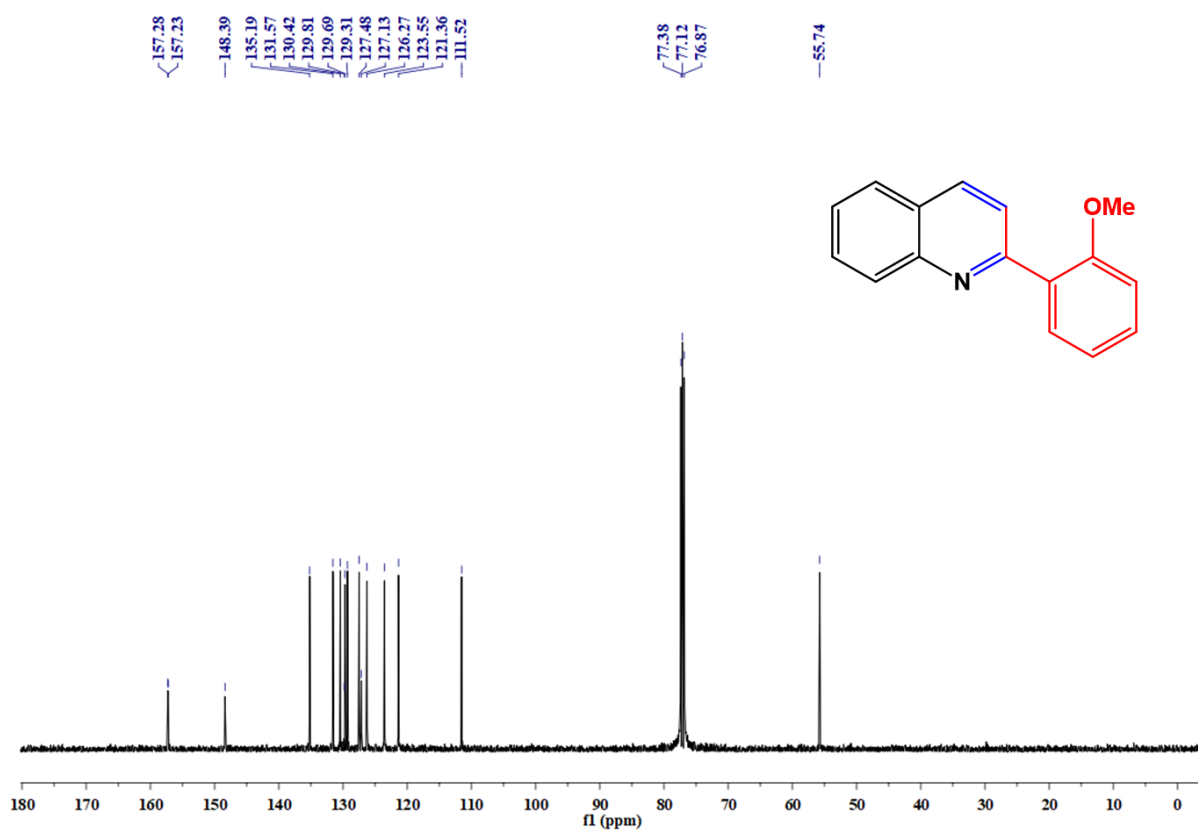


Figure S186: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11j** in CDCl_3 at 126 MHz.

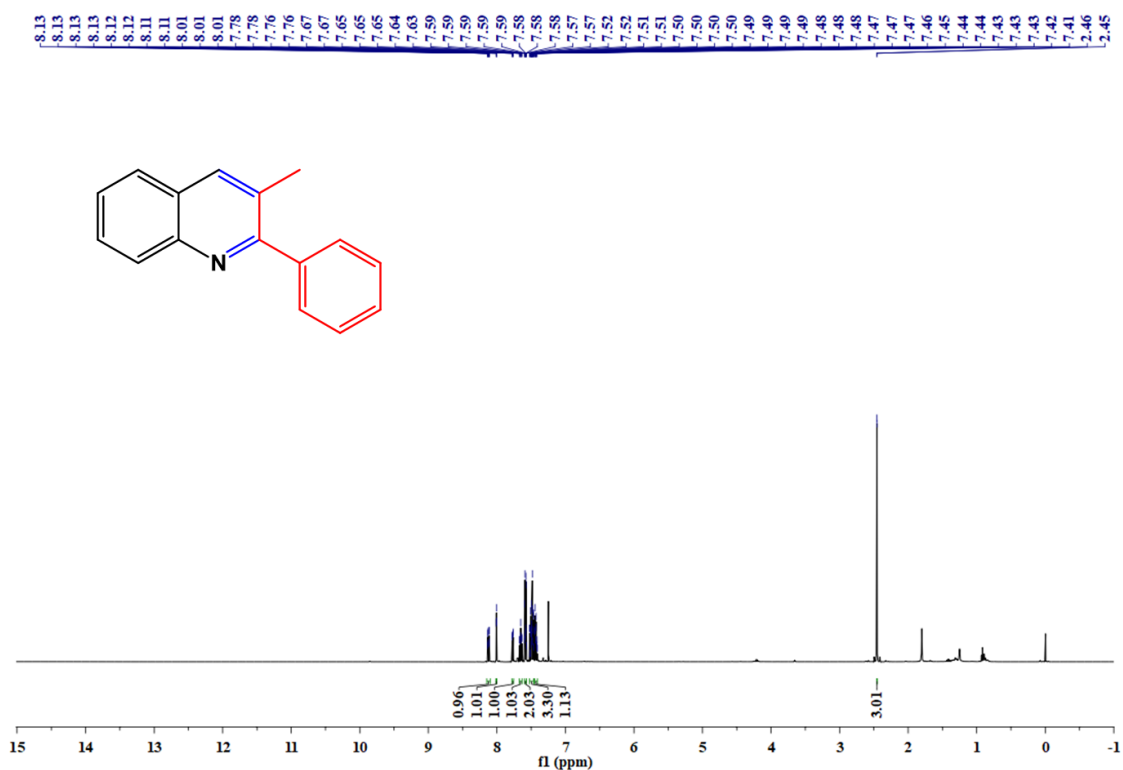


Figure S187: ^1H NMR spectrum of **11k** in CDCl_3 at 500 MHz.

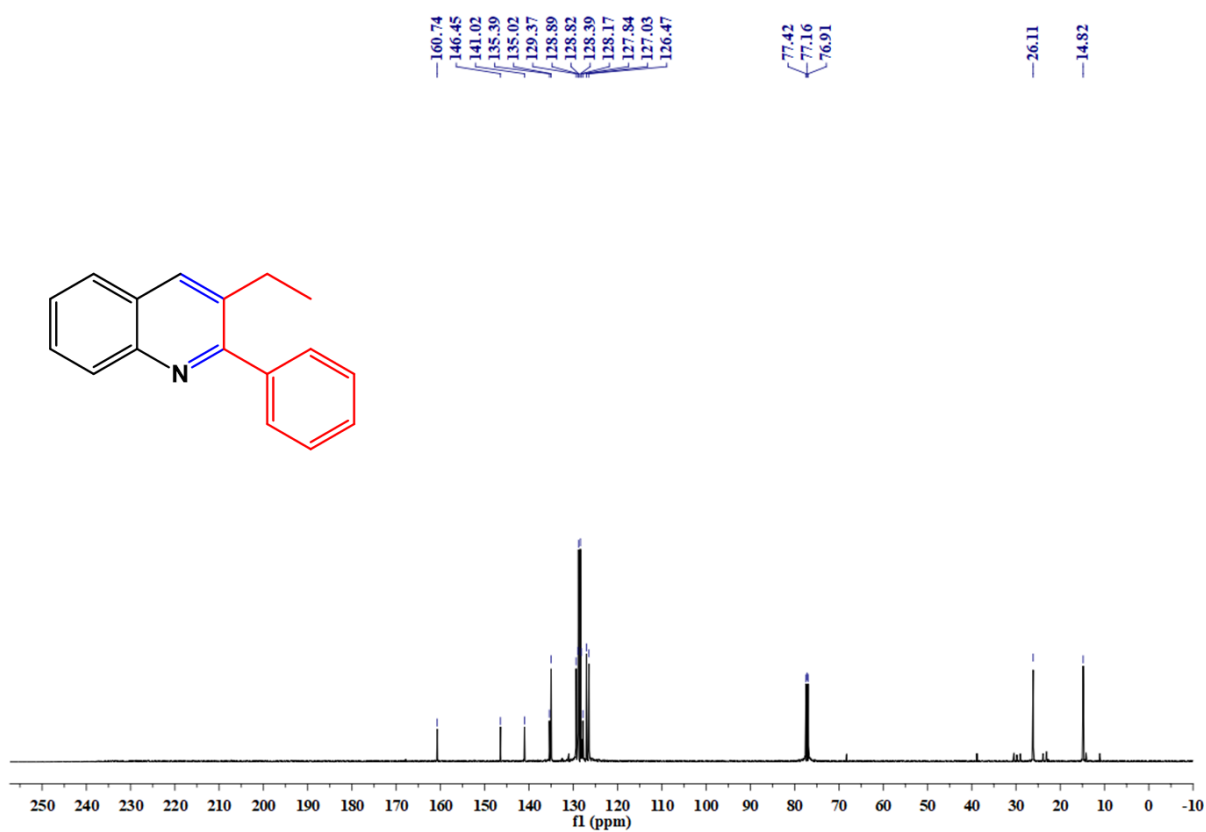


Figure S190: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11l** in CDCl_3 at 126 MHz.

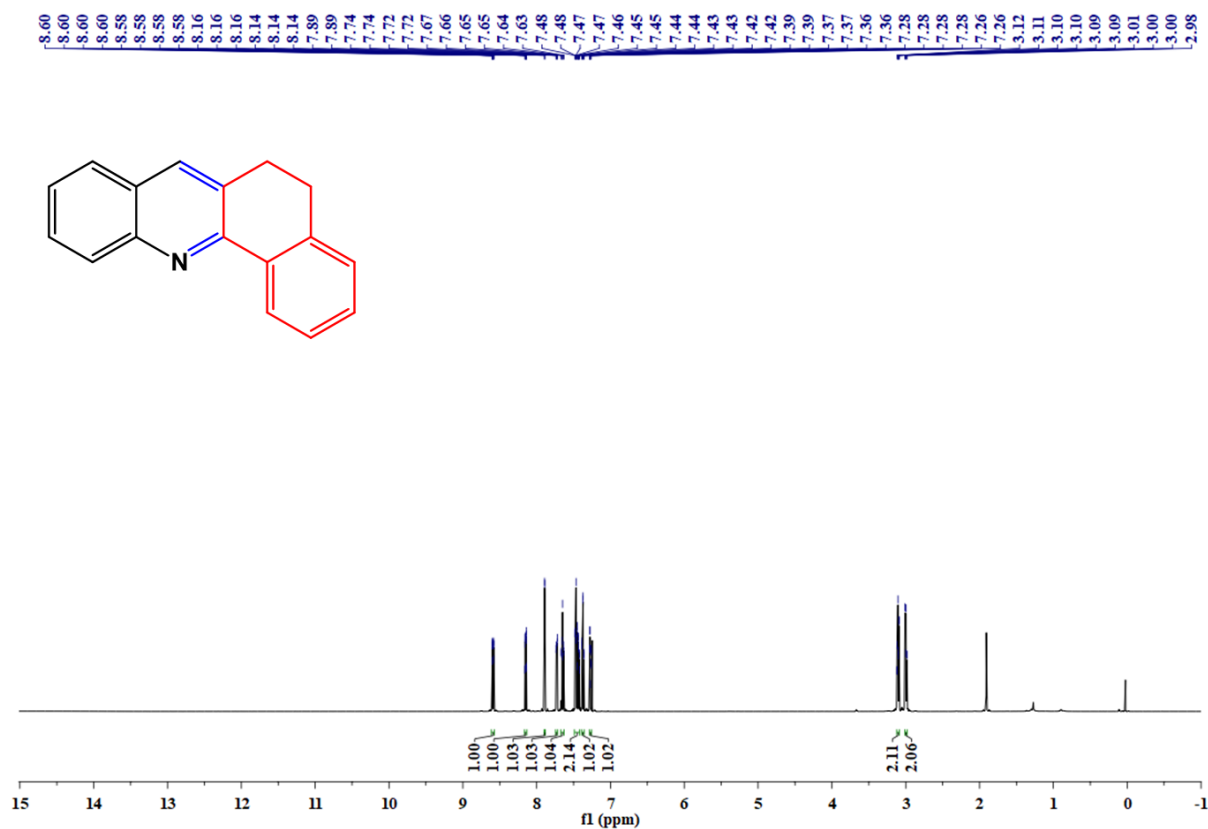


Figure S191: ^1H NMR spectrum of **11m** in CDCl_3 at 500 MHz.

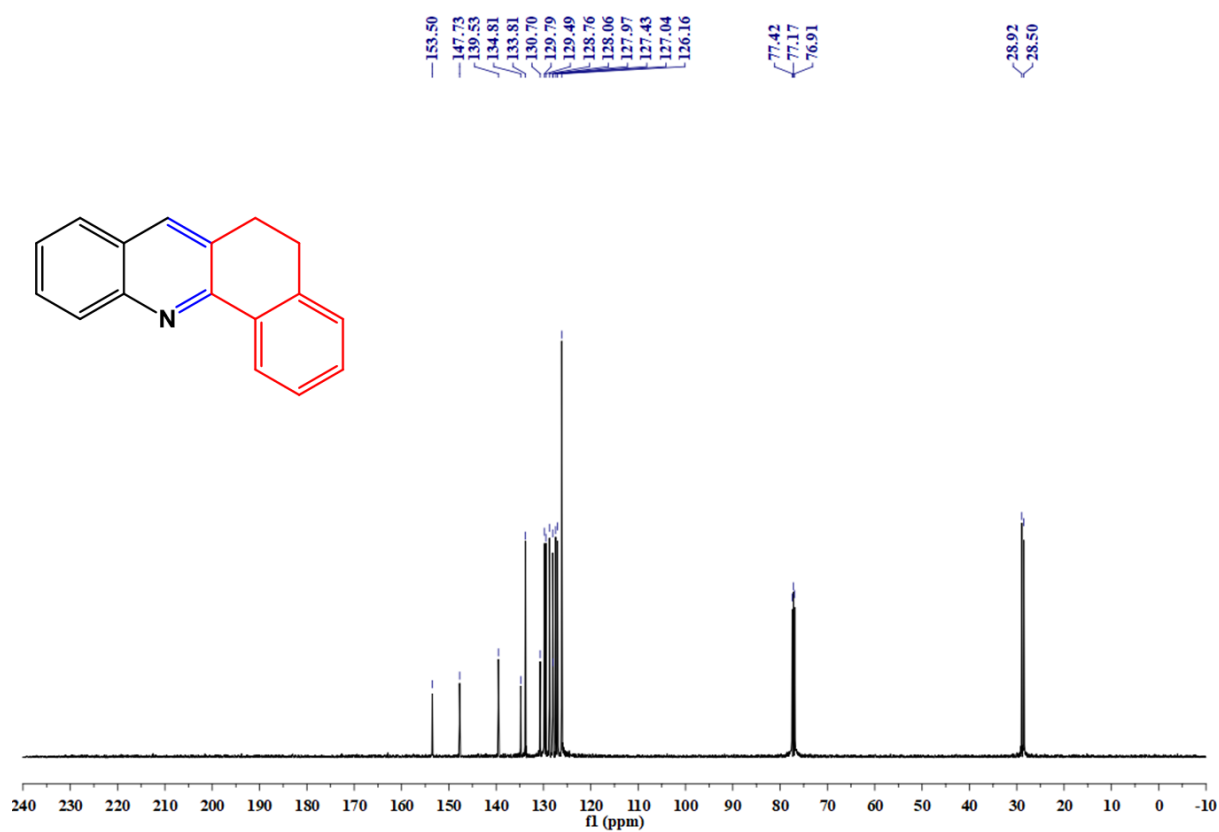


Figure S192: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11m** in CDCl_3 at 126 MHz.

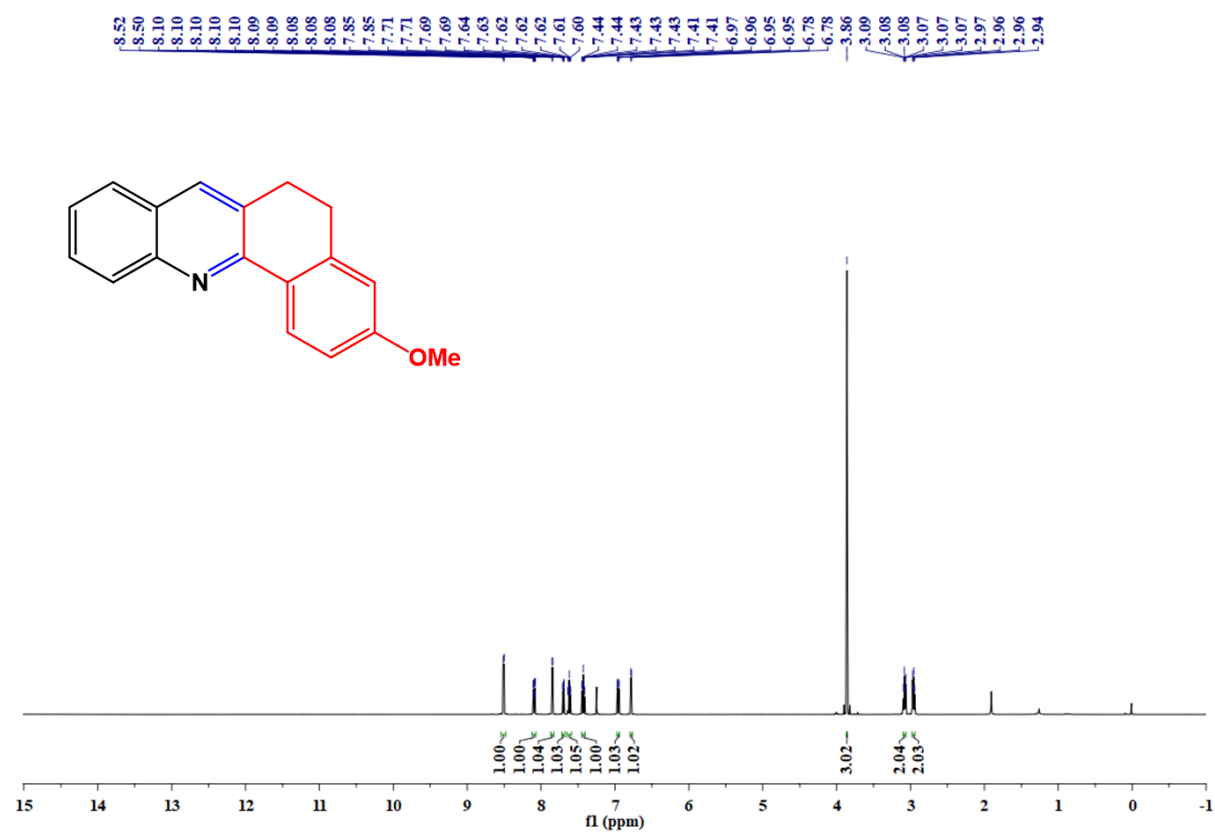


Figure S193: ^1H NMR spectrum of **11n** in CDCl_3 at 500 MHz.

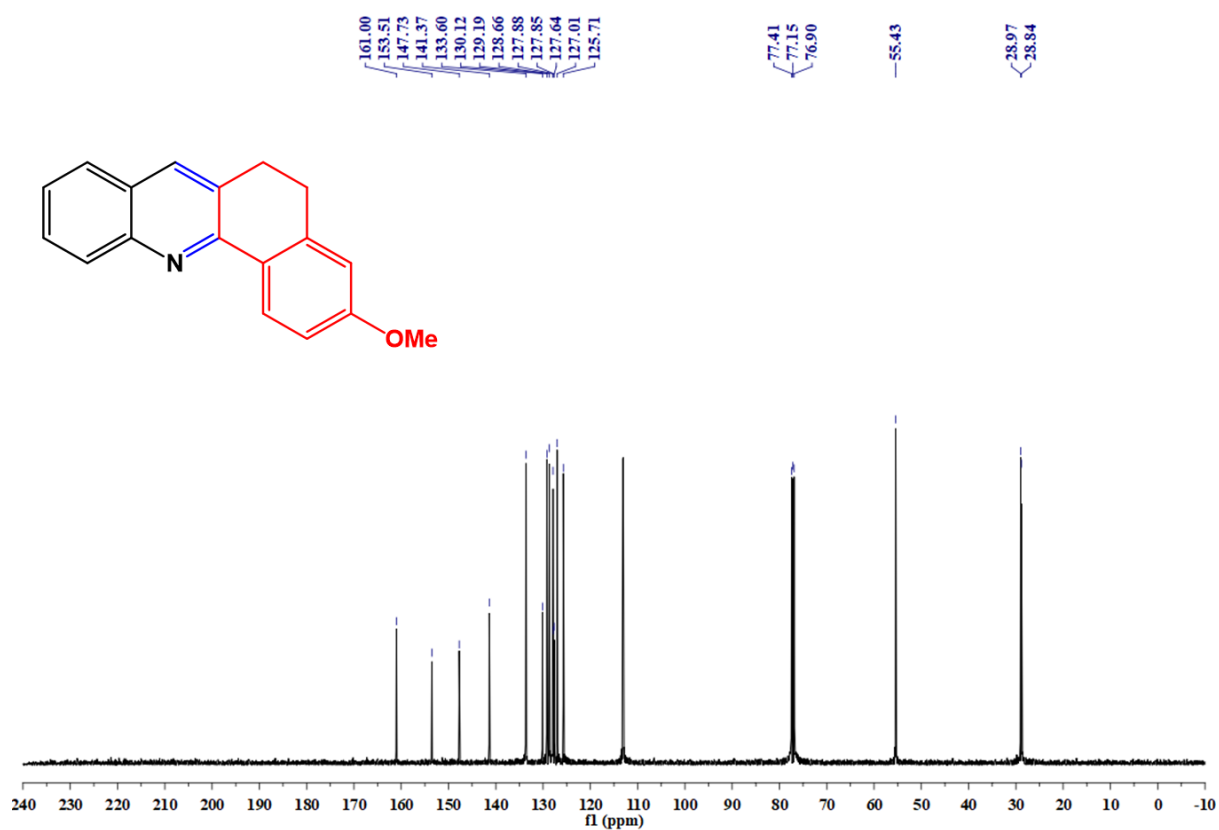


Figure S194: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11n** in CDCl_3 at 126 MHz.

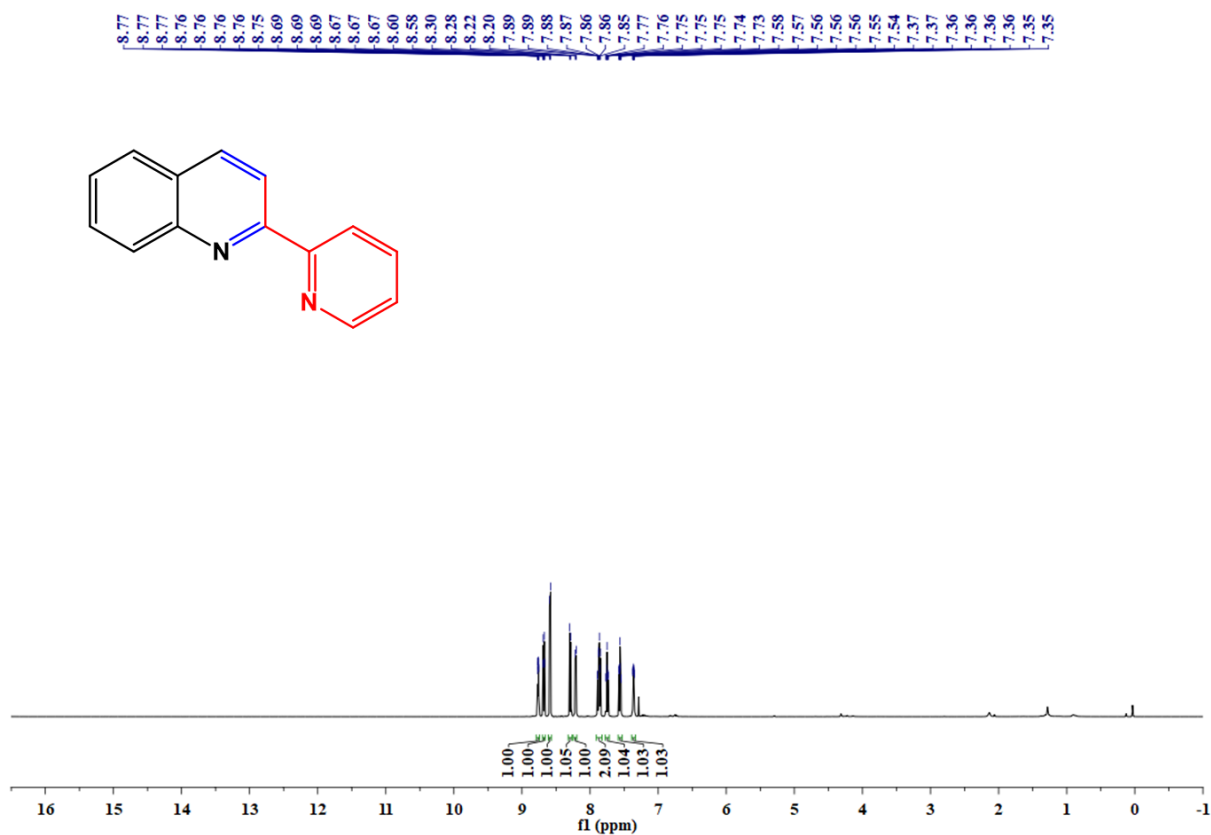
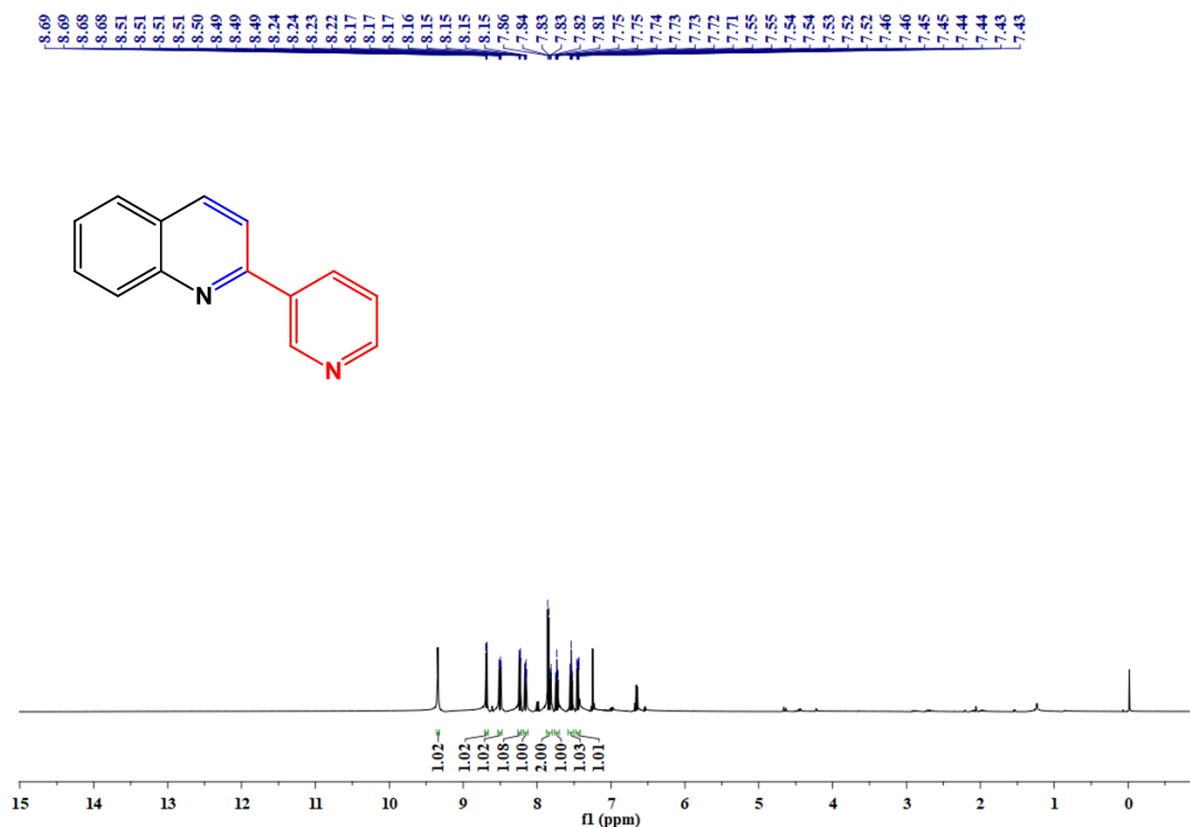
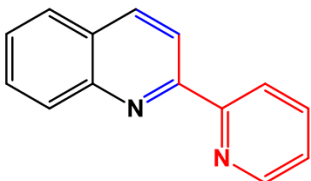


Figure S195: ^1H NMR spectrum of **11o** in CDCl_3 at 500 MHz.



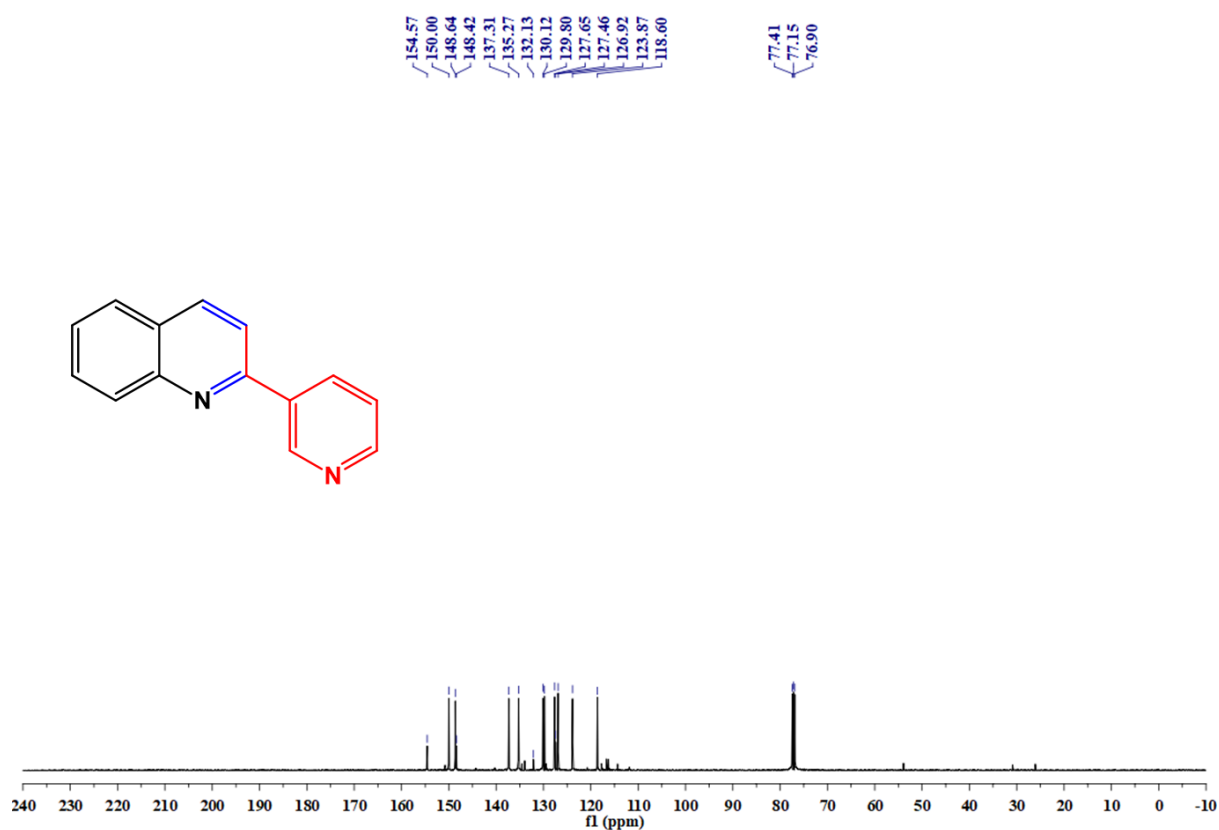


Figure S198: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11p** in CDCl_3 at 126 MHz.

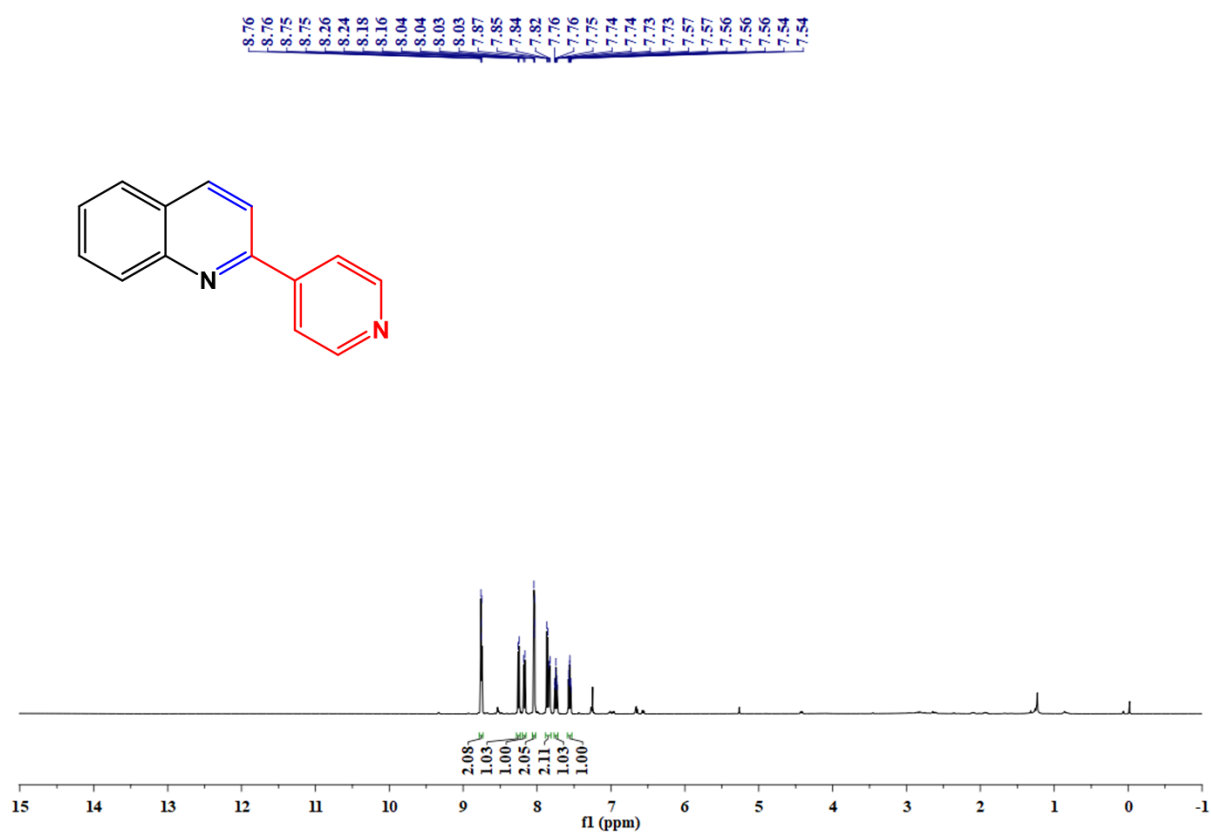


Figure S199: ^1H NMR spectrum of **11q** in CDCl_3 at 500 MHz.

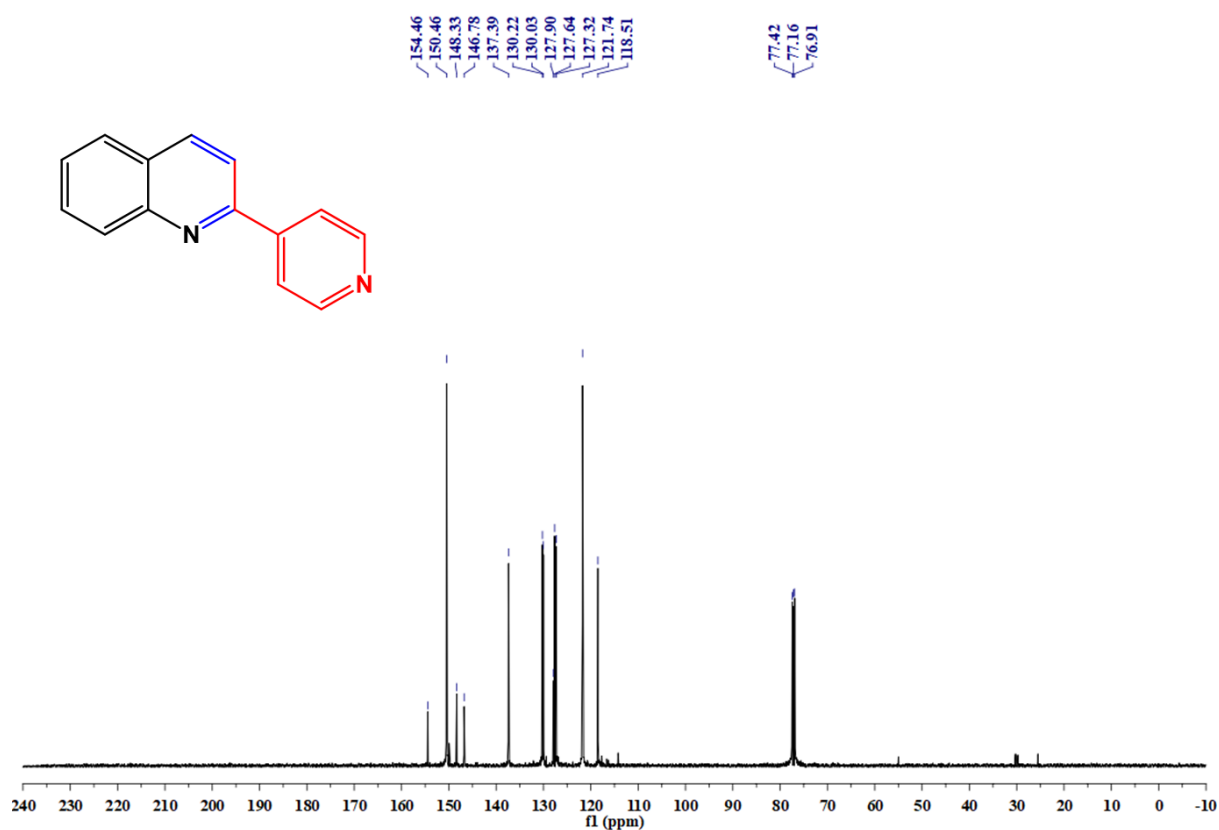


Figure S200: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11q** in CDCl_3 at 126 MHz.

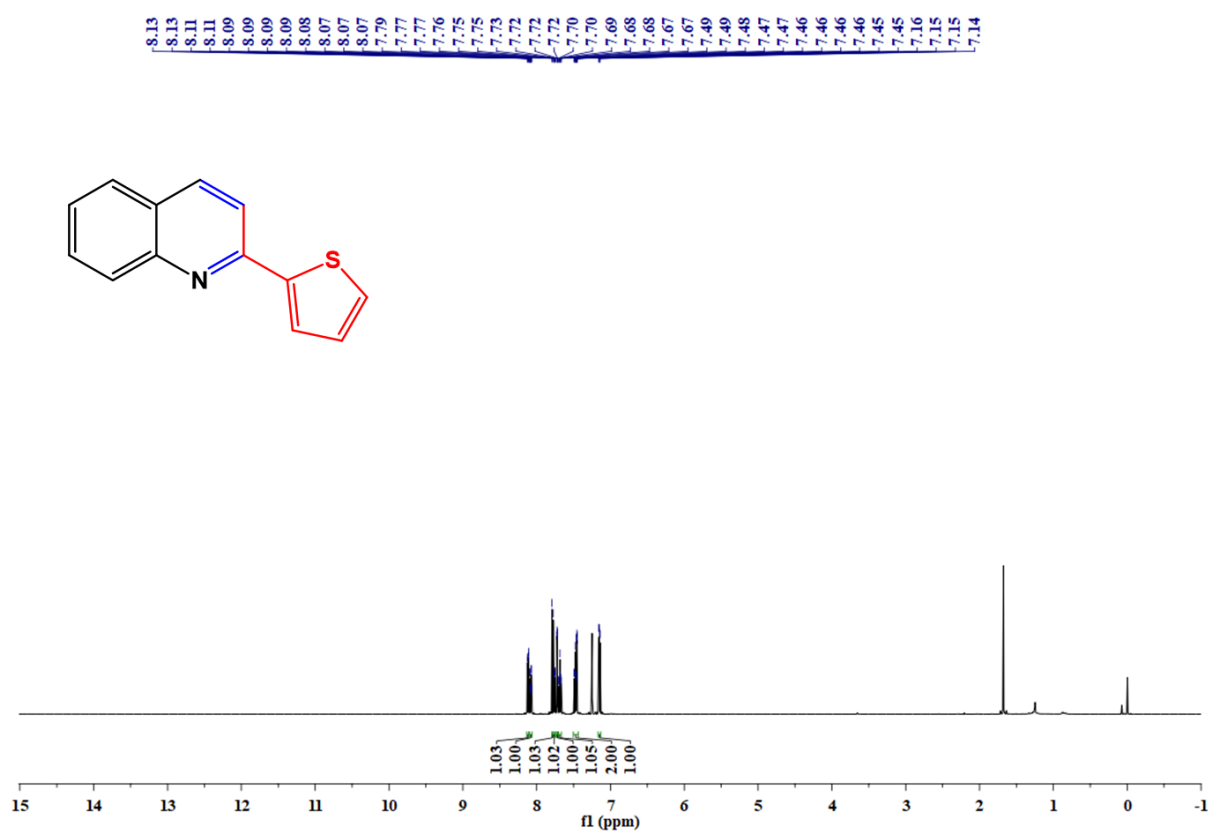


Figure S201: ^1H NMR spectrum of **11r** in CDCl_3 at 500 MHz.

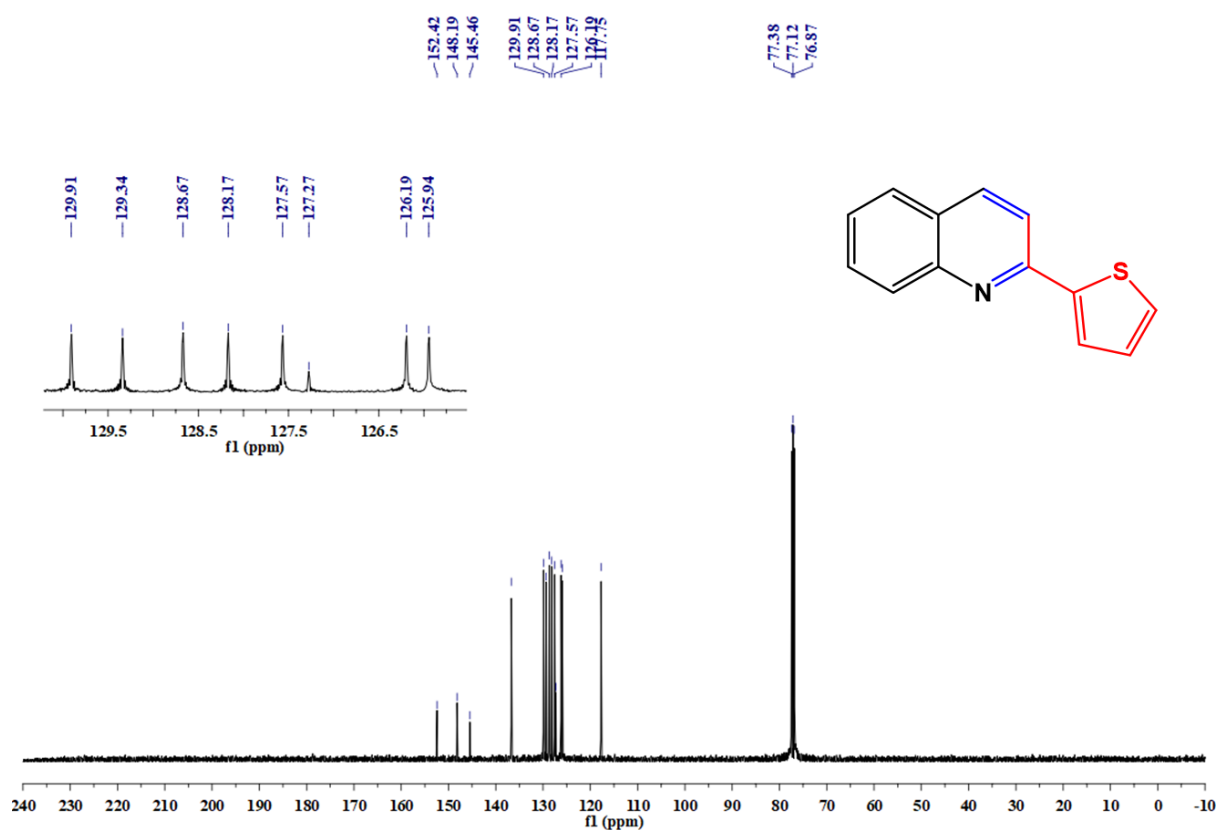


Figure S202: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11r** in CDCl_3 at 126 MHz.

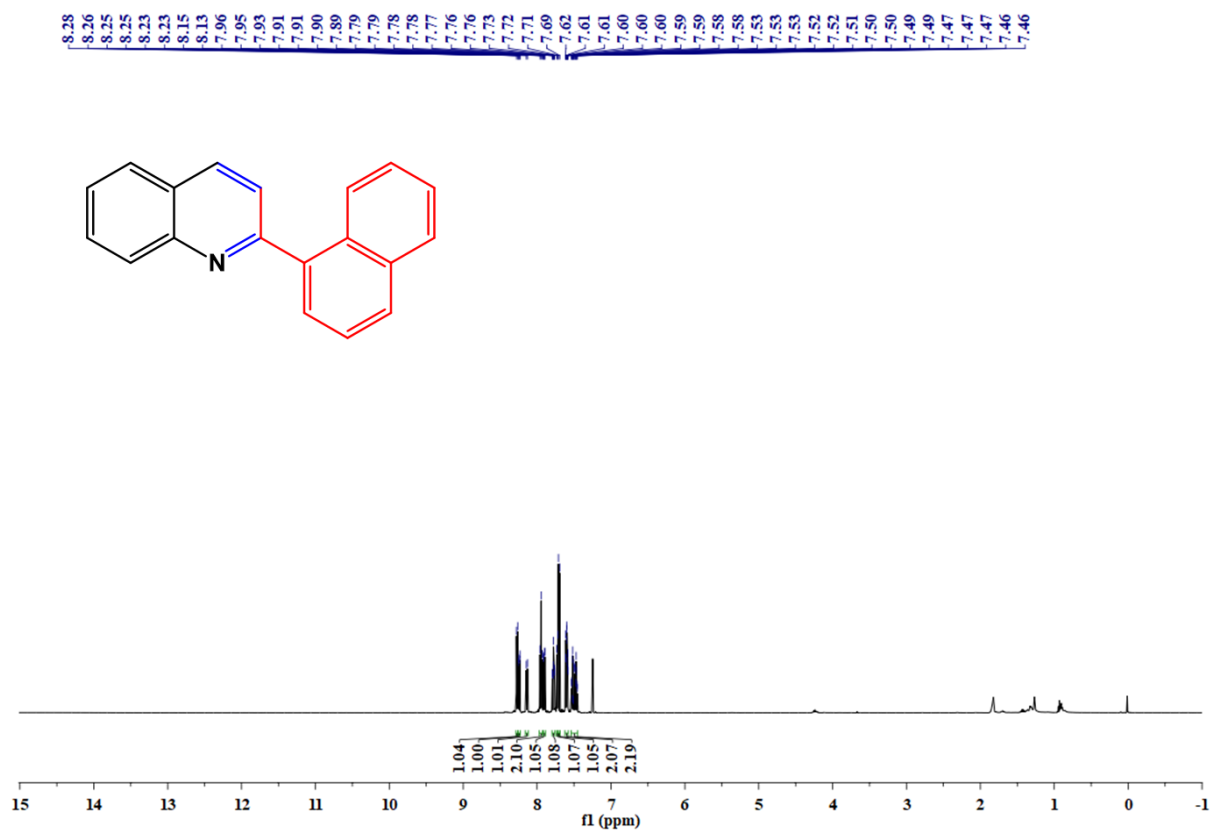


Figure S203: ^1H NMR spectrum of **11s** in CDCl_3 at 500 MHz.

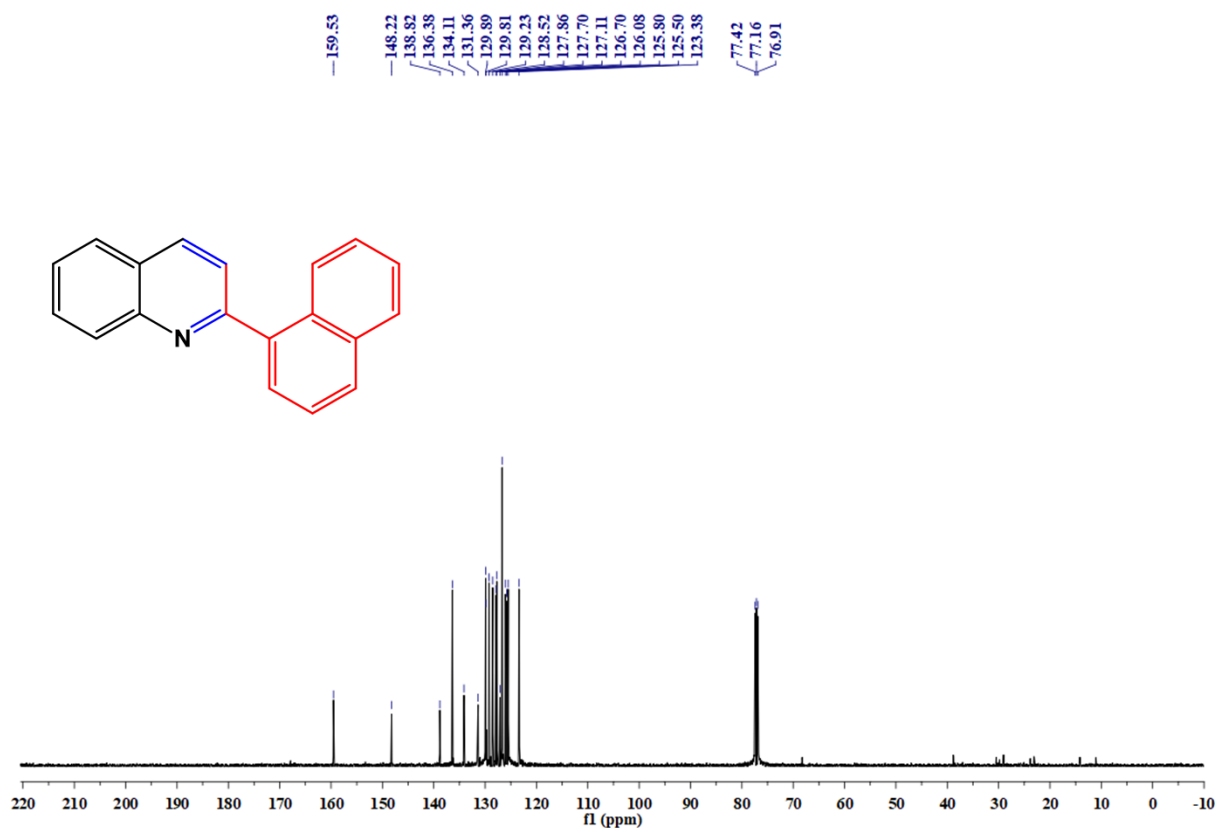


Figure S204: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11s** in CDCl_3 at 126 MHz.

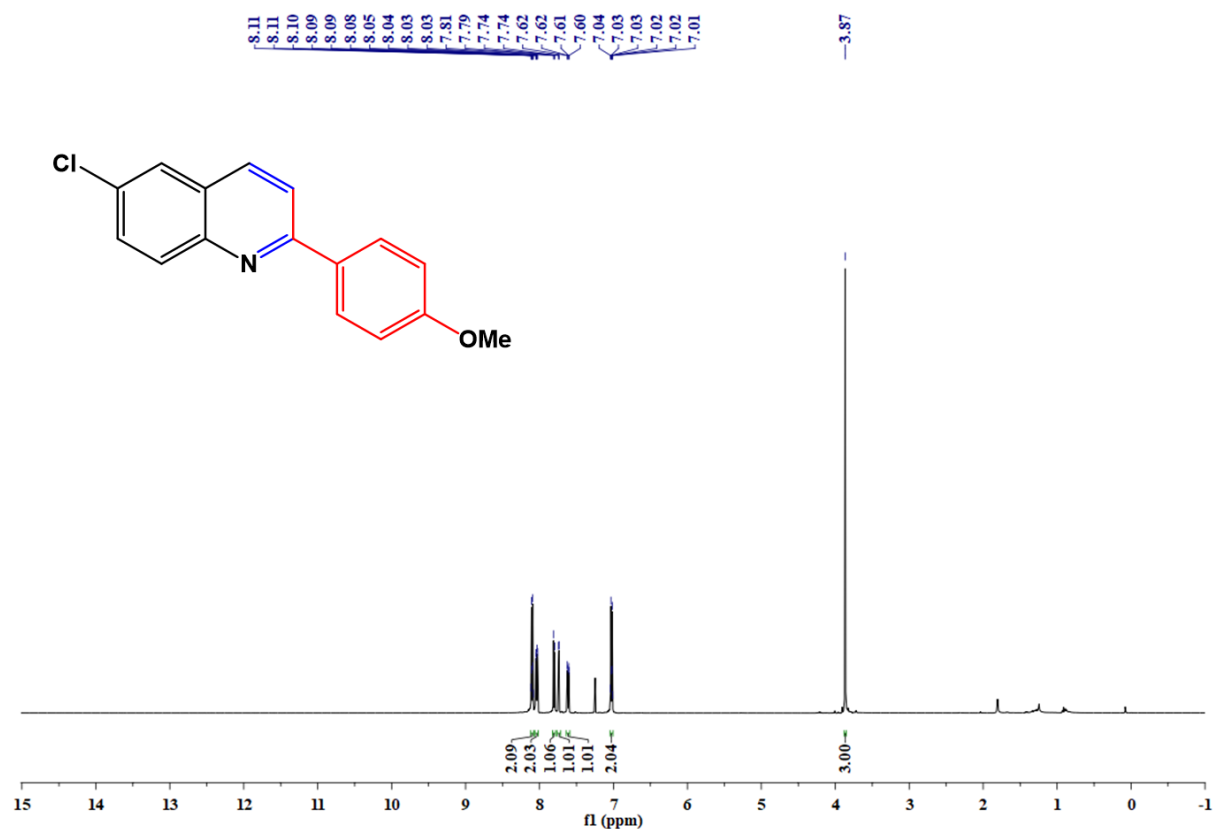


Figure S205: ^1H NMR spectrum of **11t** in CDCl_3 at 500 MHz.

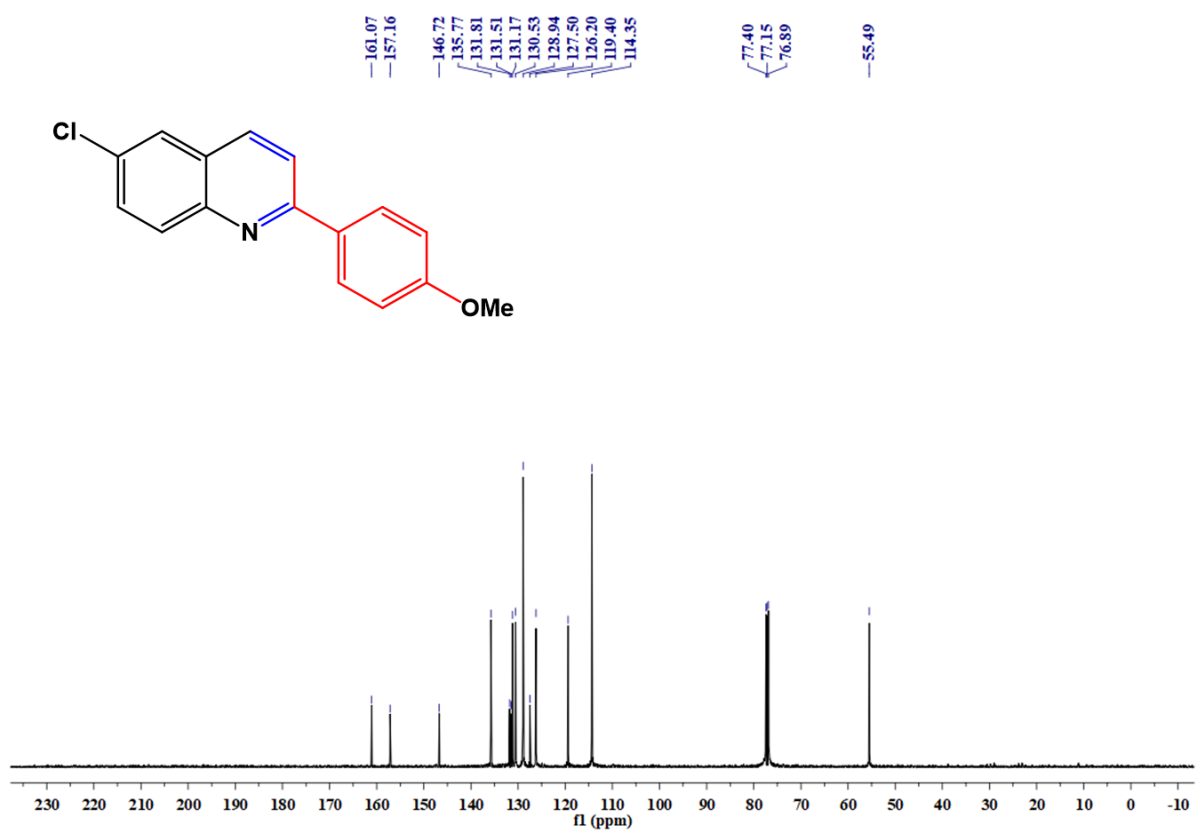


Figure S206: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11t** in CDCl_3 at 126 MHz.

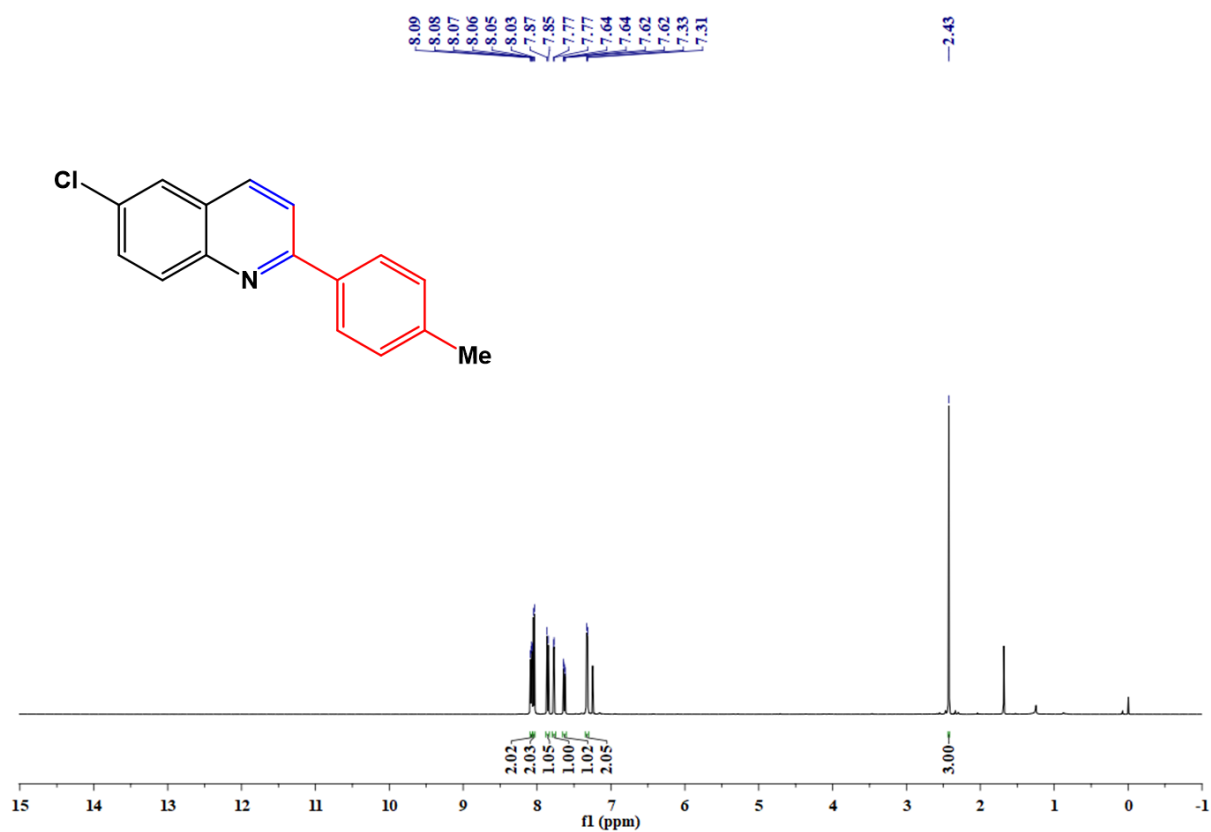


Figure S207: ^1H NMR spectrum of **11u** in CDCl_3 at 500 MHz.

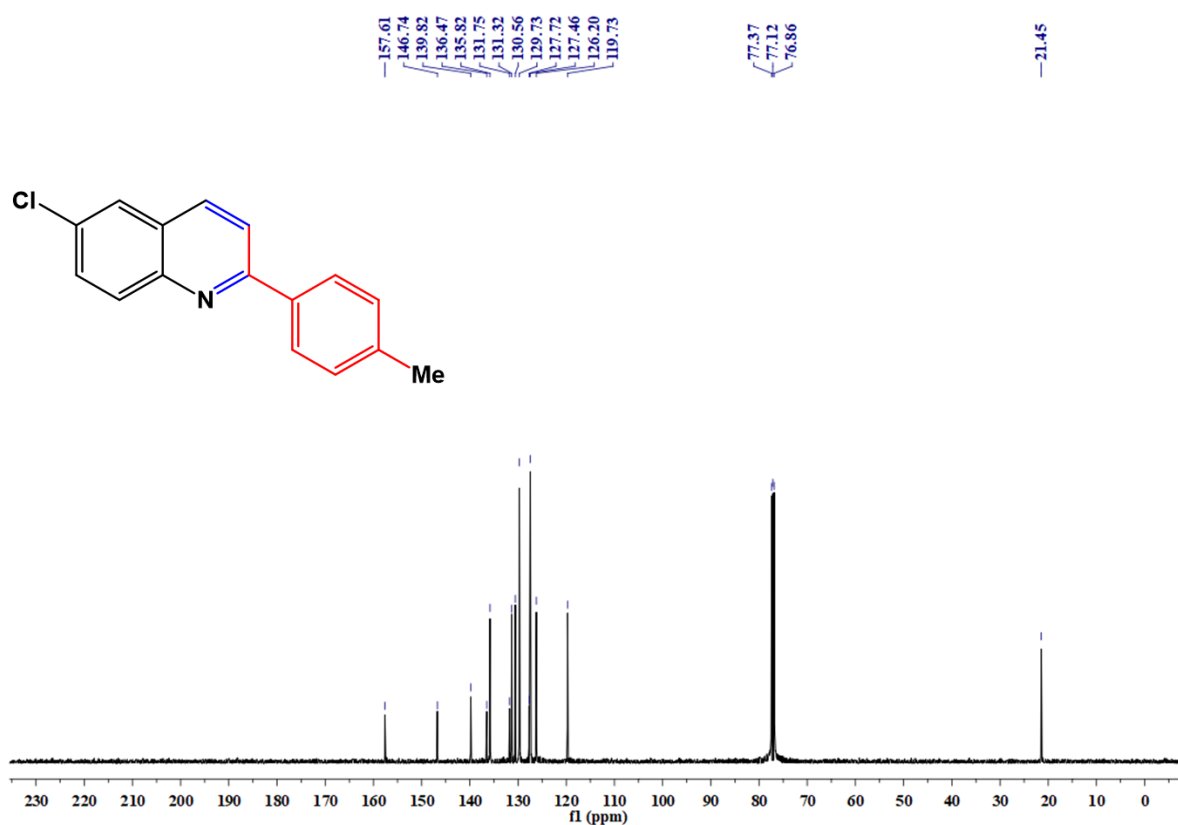


Figure S208: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11u** in CDCl_3 at 126 MHz.

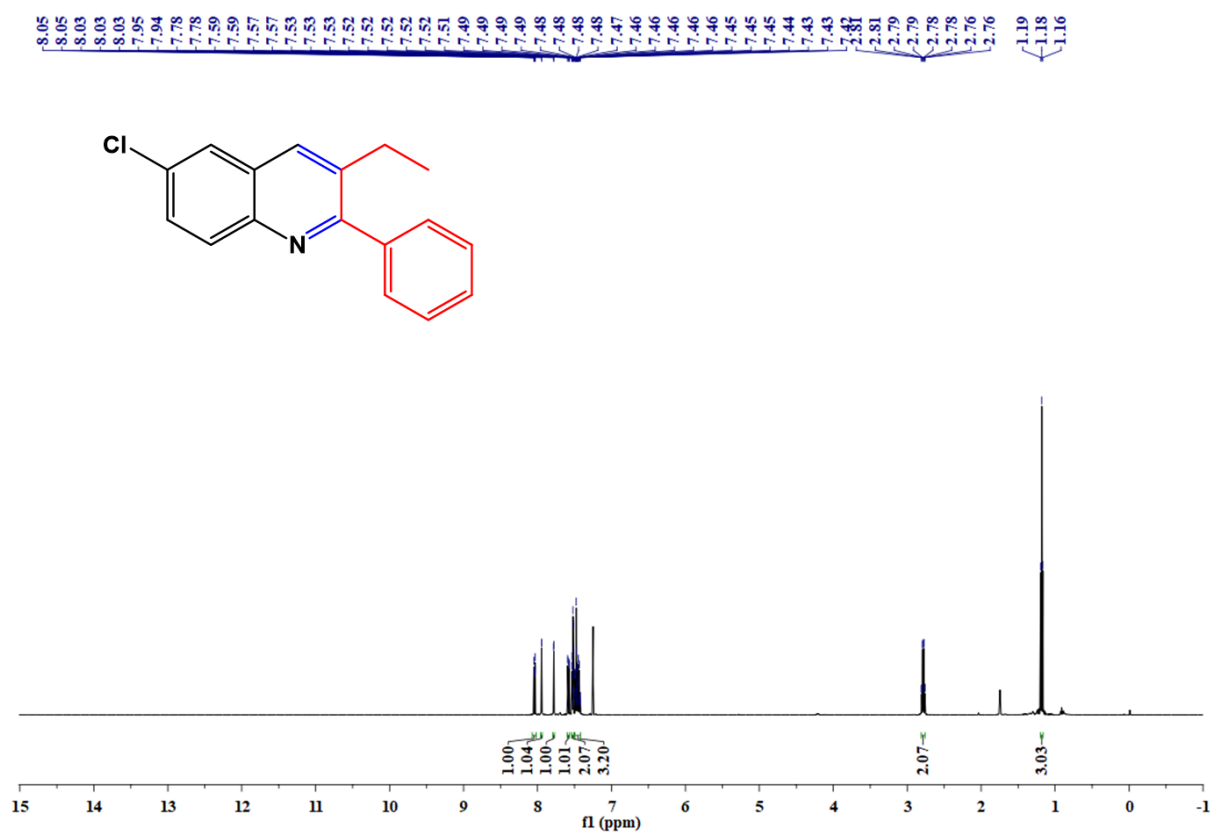


Figure S209: ^1H NMR spectrum of **11v** in CDCl_3 at 500 MHz.

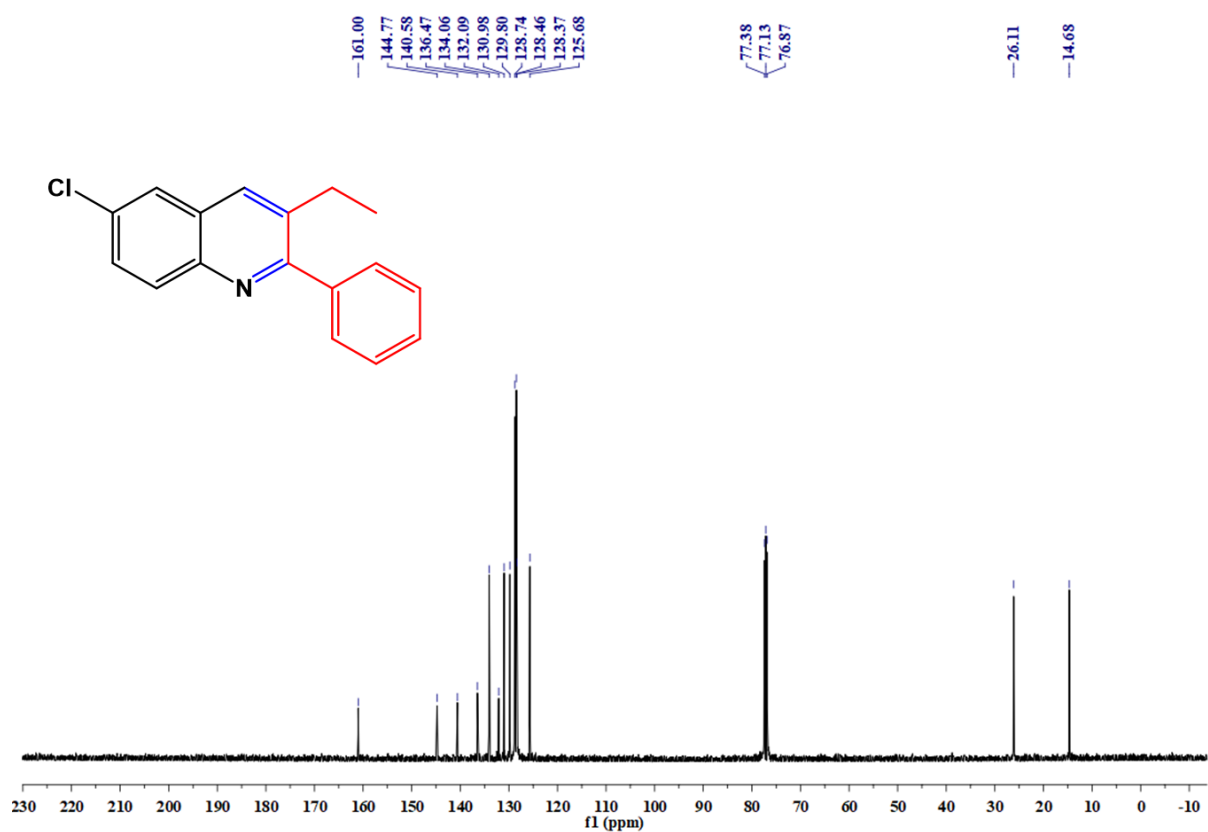


Figure S210: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11v** in CDCl_3 at 126 MHz.

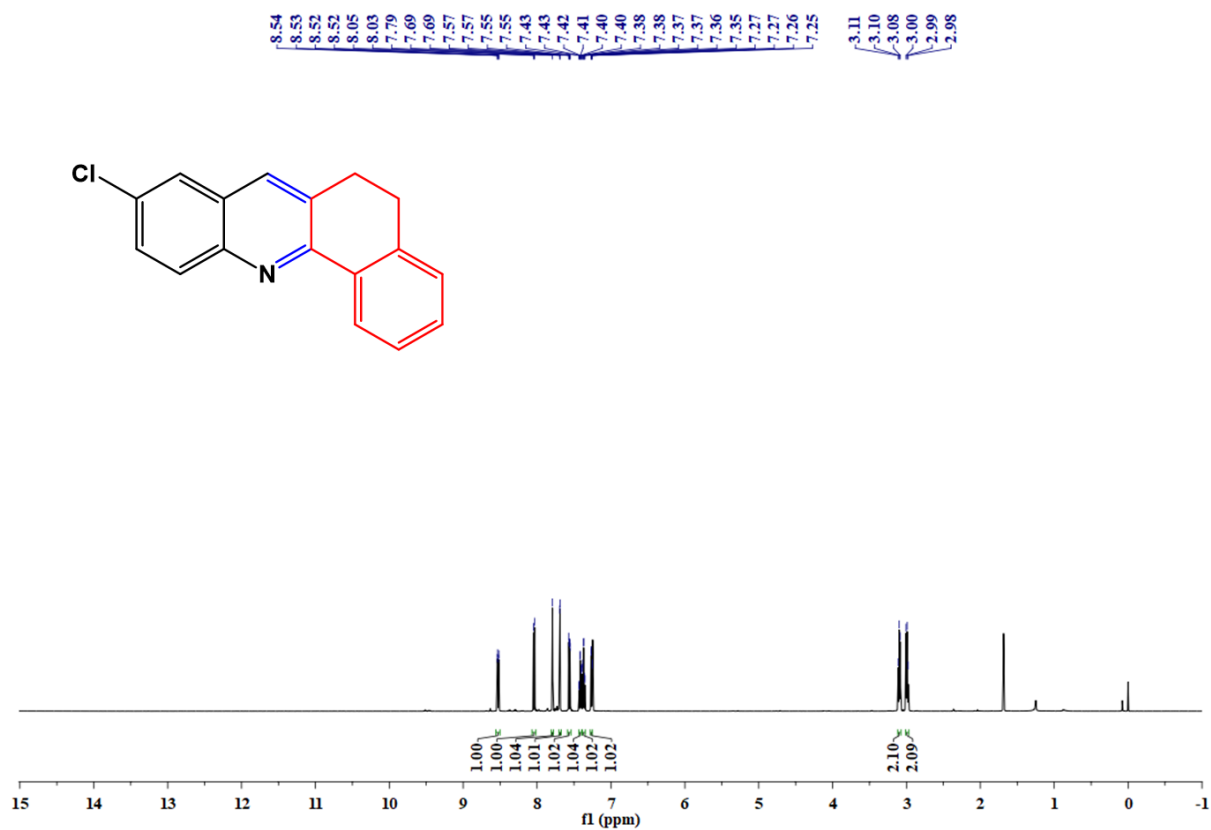


Figure S211: ^1H NMR spectrum of **11w** in CDCl_3 at 500 MHz.

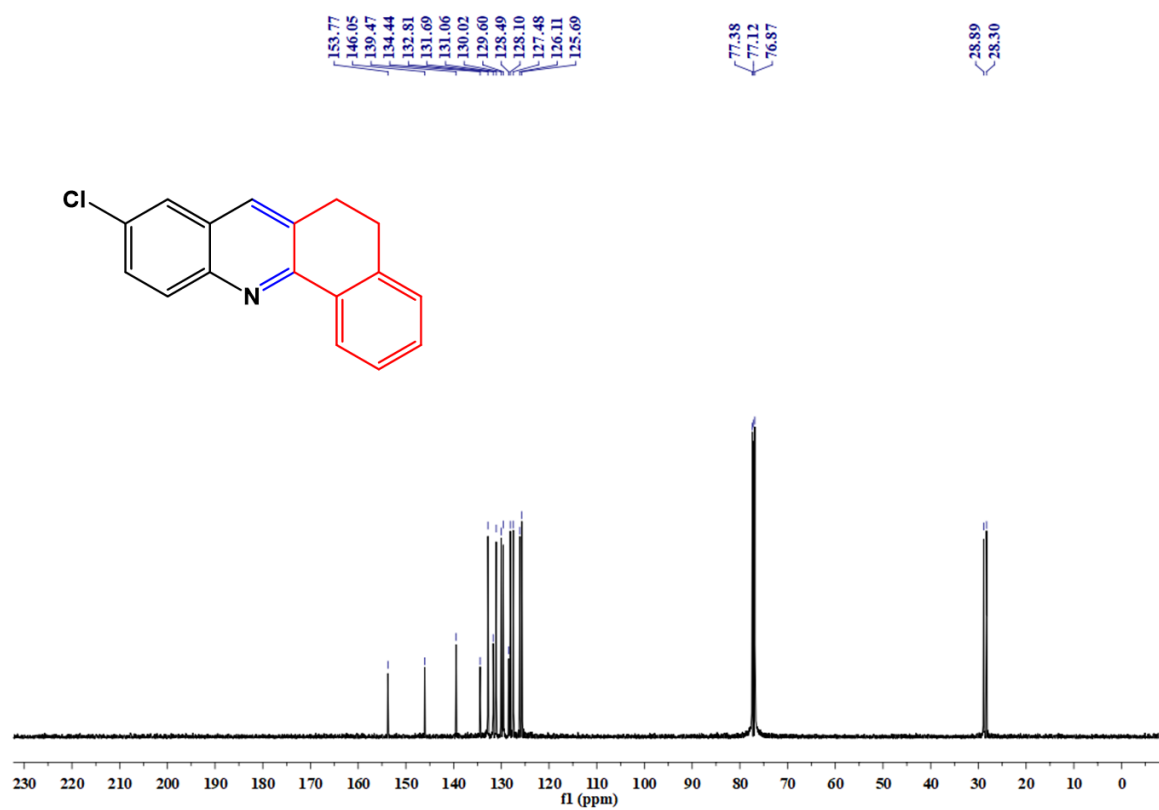


Figure S212: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11w** in CDCl_3 at 126 MHz.

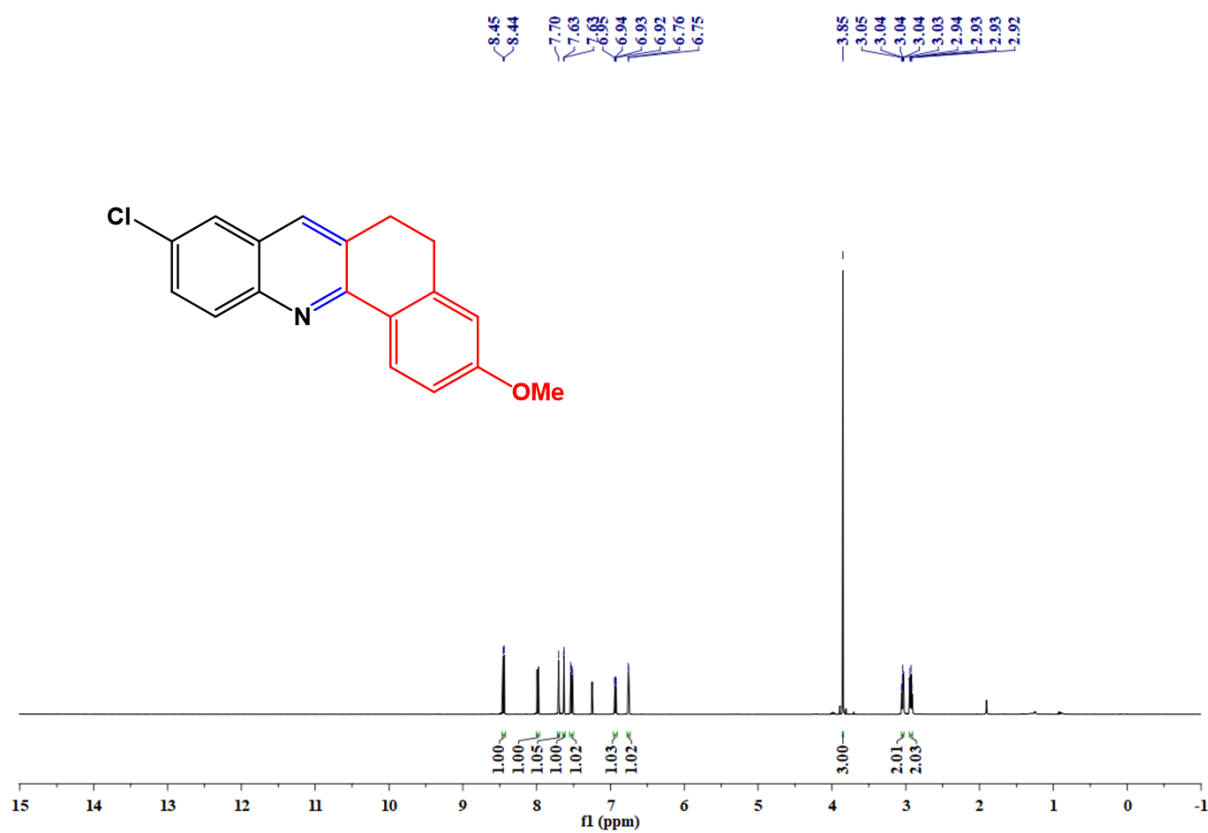


Figure S213: ^1H NMR spectrum of **11x** in CDCl_3 at 500 MHz.

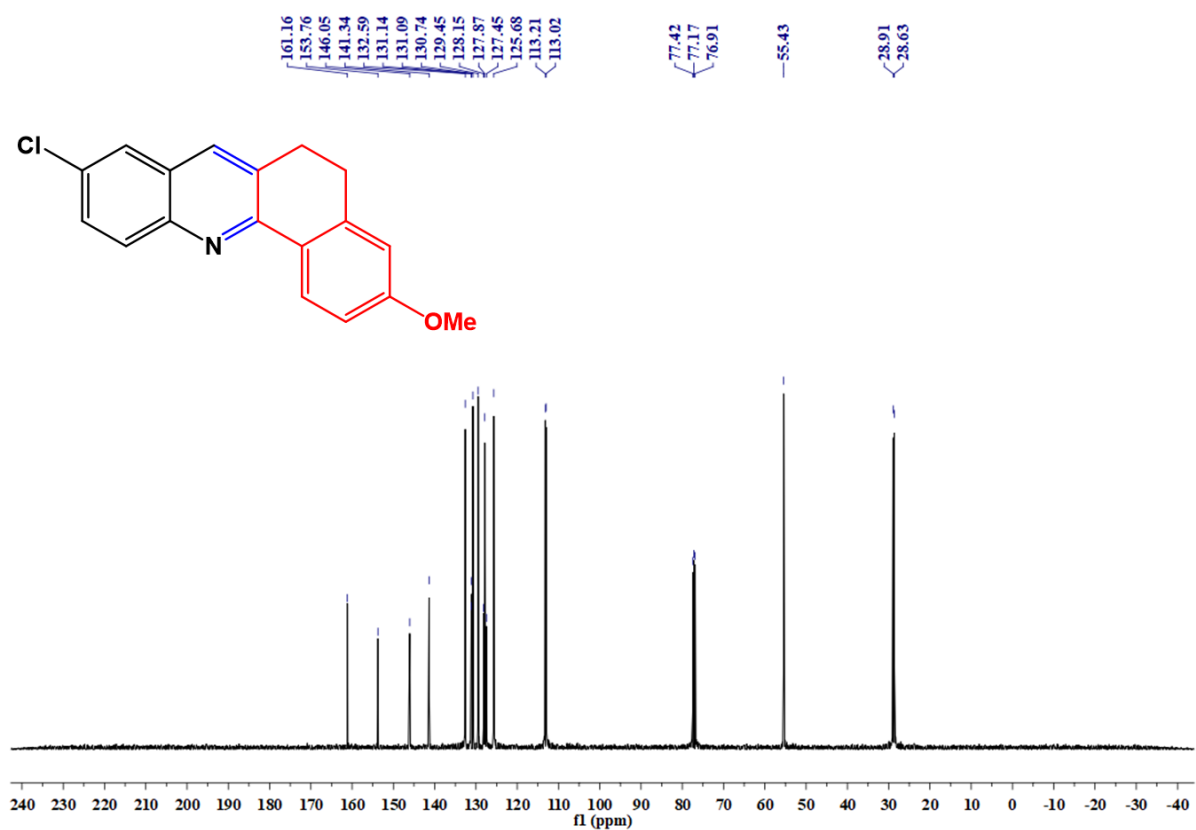


Figure S214: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11x** in CDCl_3 at 126 MHz.

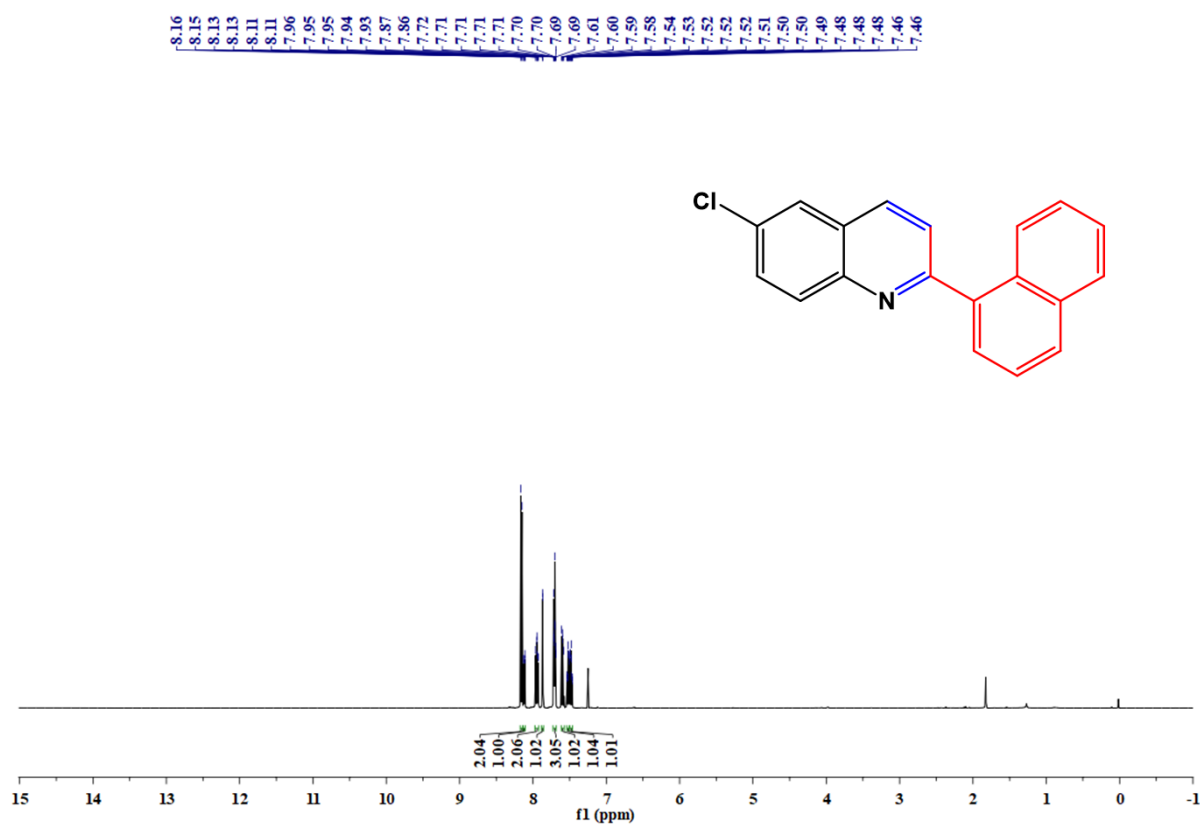


Figure S215: ^1H NMR spectrum of **11y** in CDCl_3 at 500 MHz.

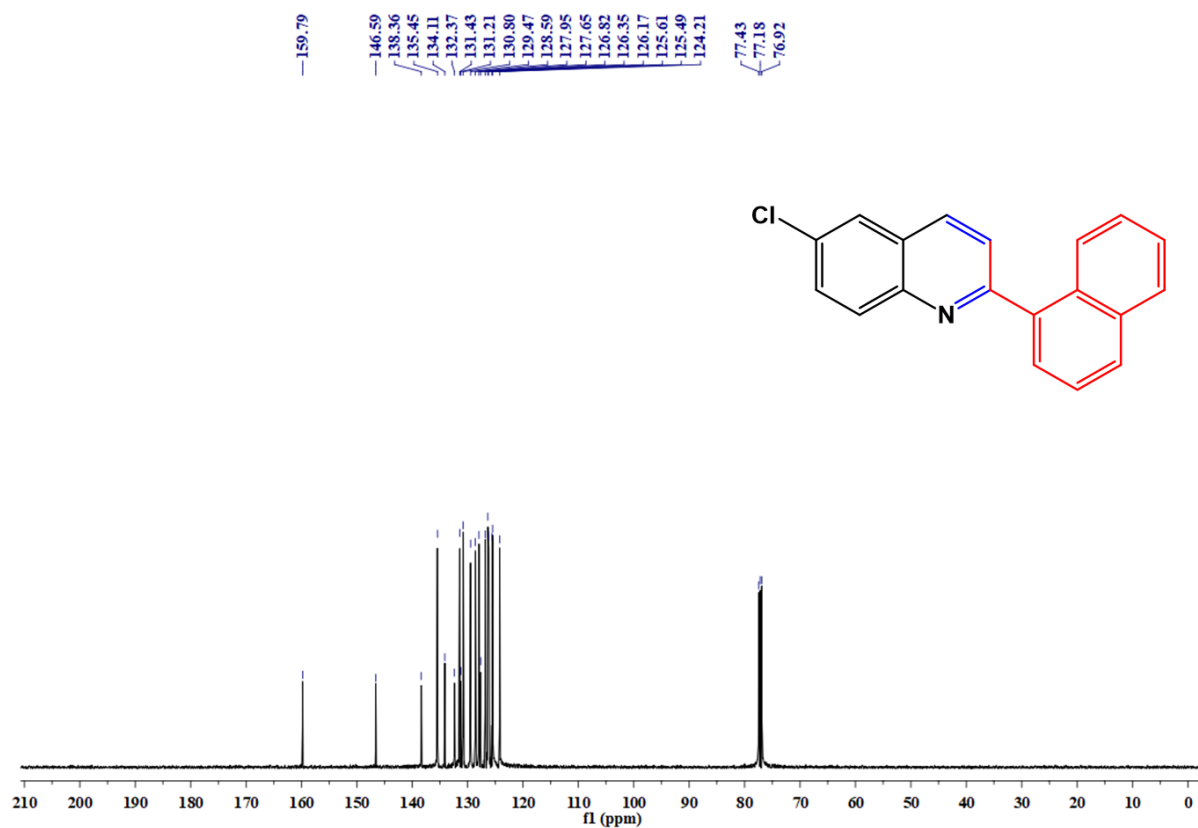


Figure S216: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11y** in CDCl_3 at 126 MHz.

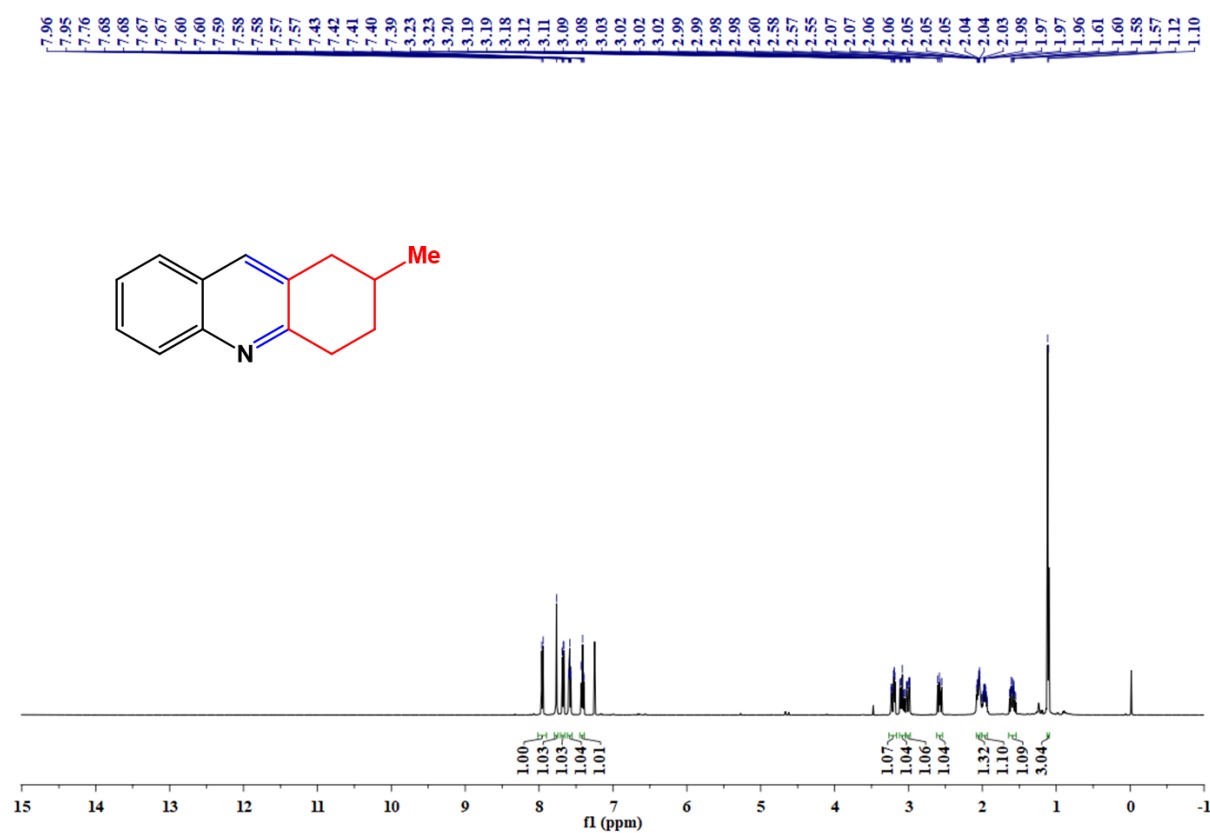


Figure S217: ^1H NMR spectrum of **11z** in CDCl_3 at 500 MHz.

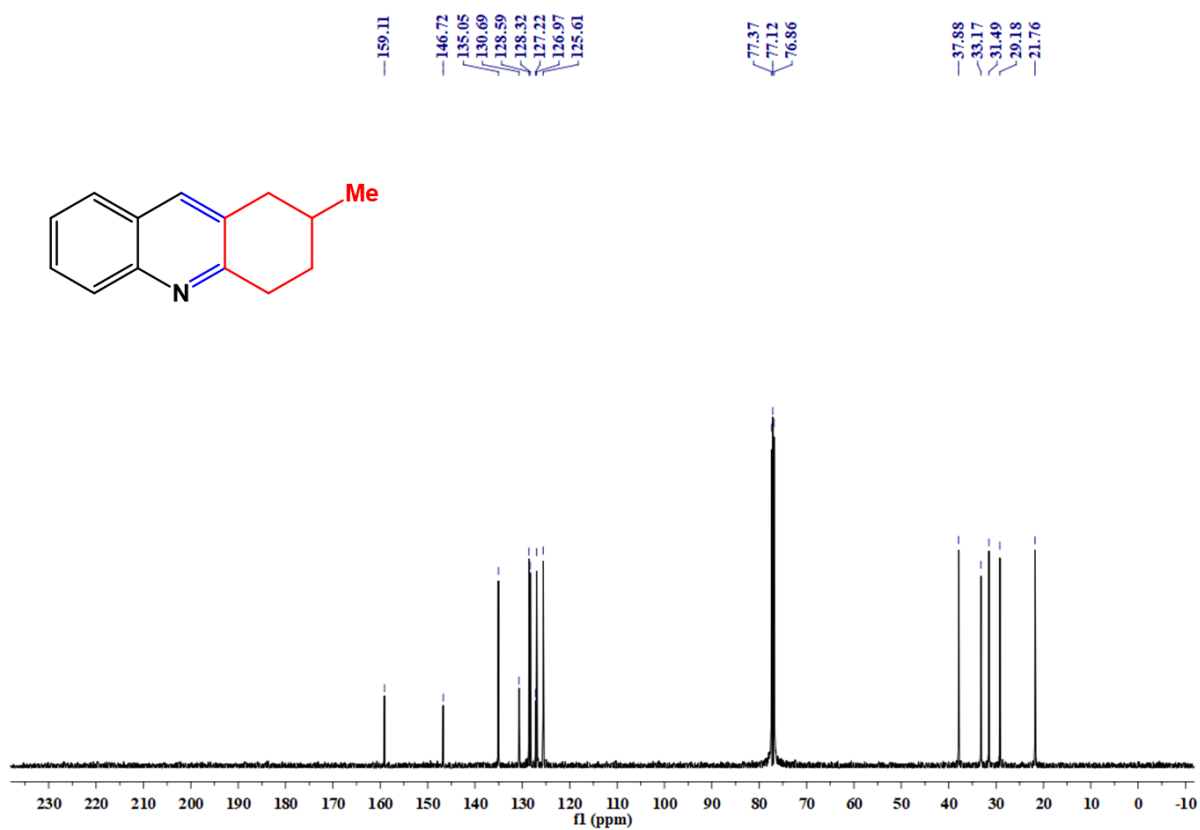


Figure S218: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **11z** in CDCl_3 at 126 MHz.

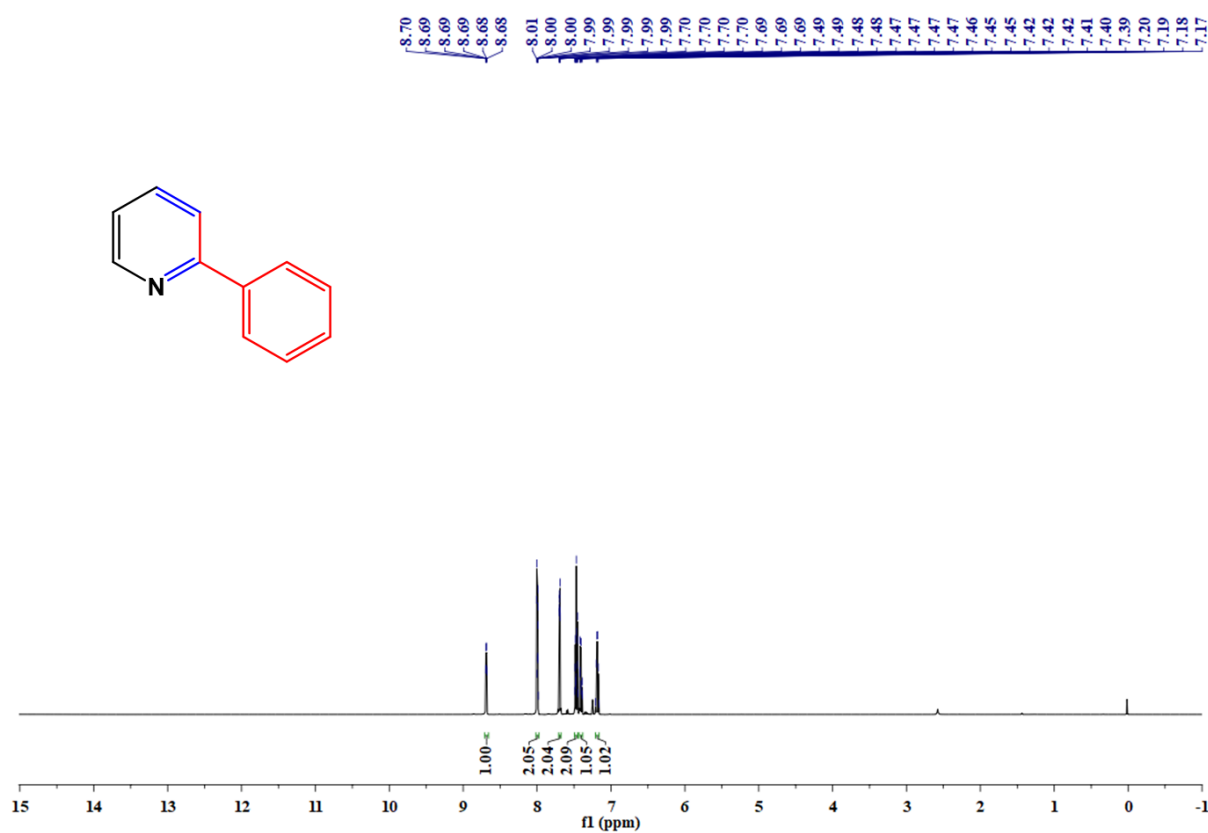


Figure S219: ^1H NMR spectrum of **11aa** in CDCl_3 at 500 MHz.

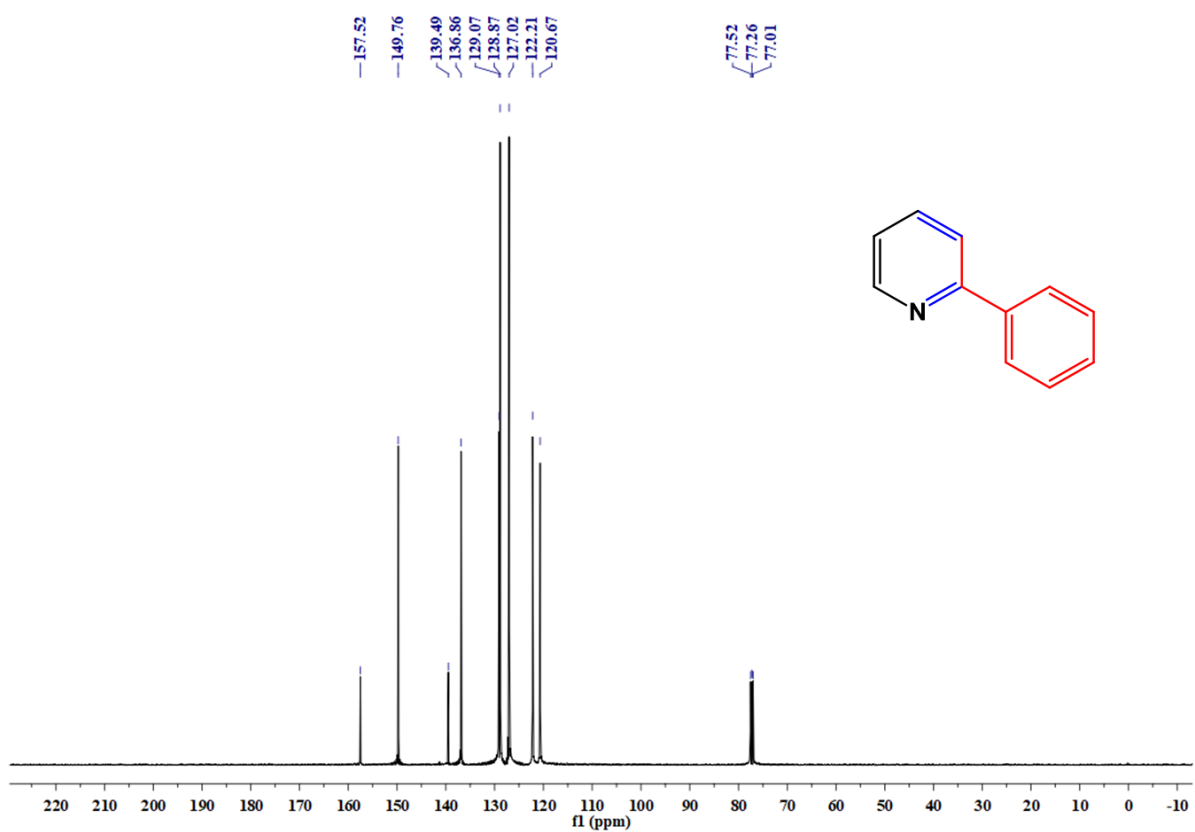


Figure S220: ¹³C{¹H} NMR spectrum of **11aa** in CDCl₃ at 126 MHz.

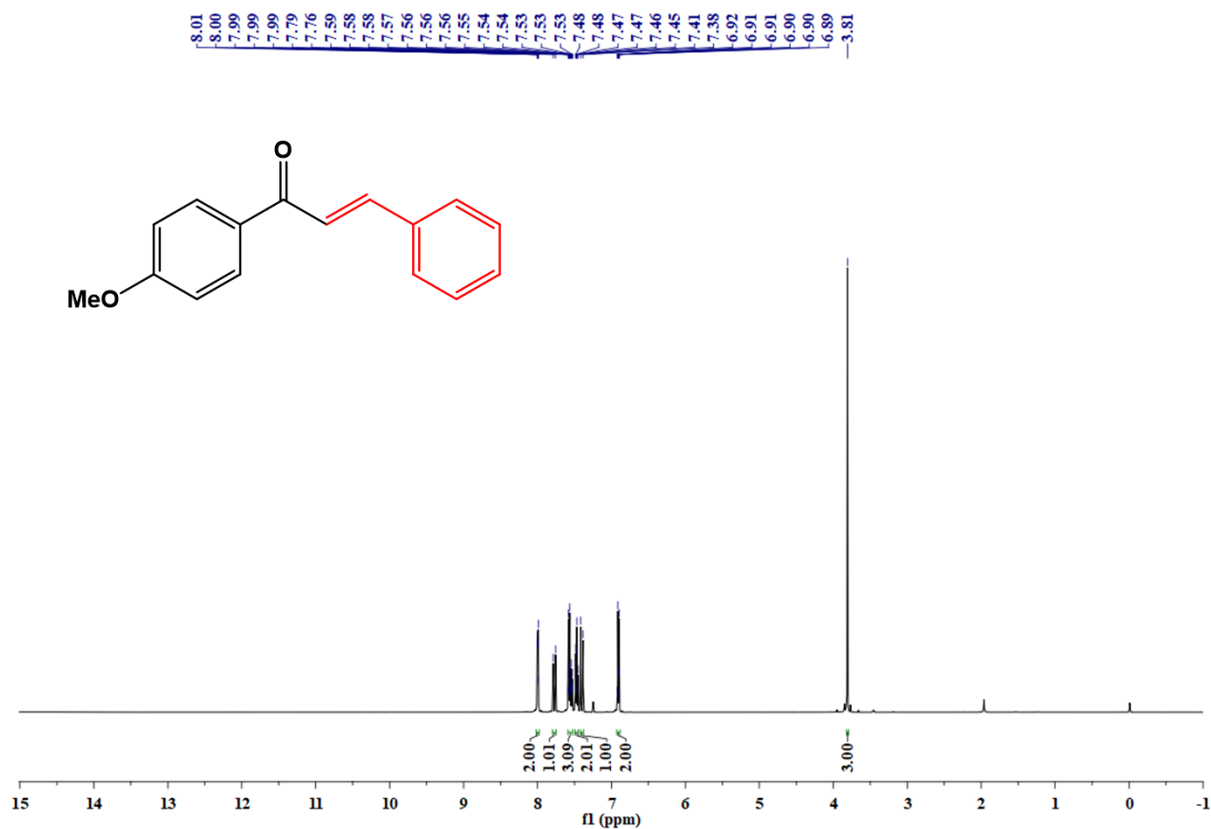


Figure S221: ¹H NMR spectrum of **3b** in CDCl₃ at 500 MHz.

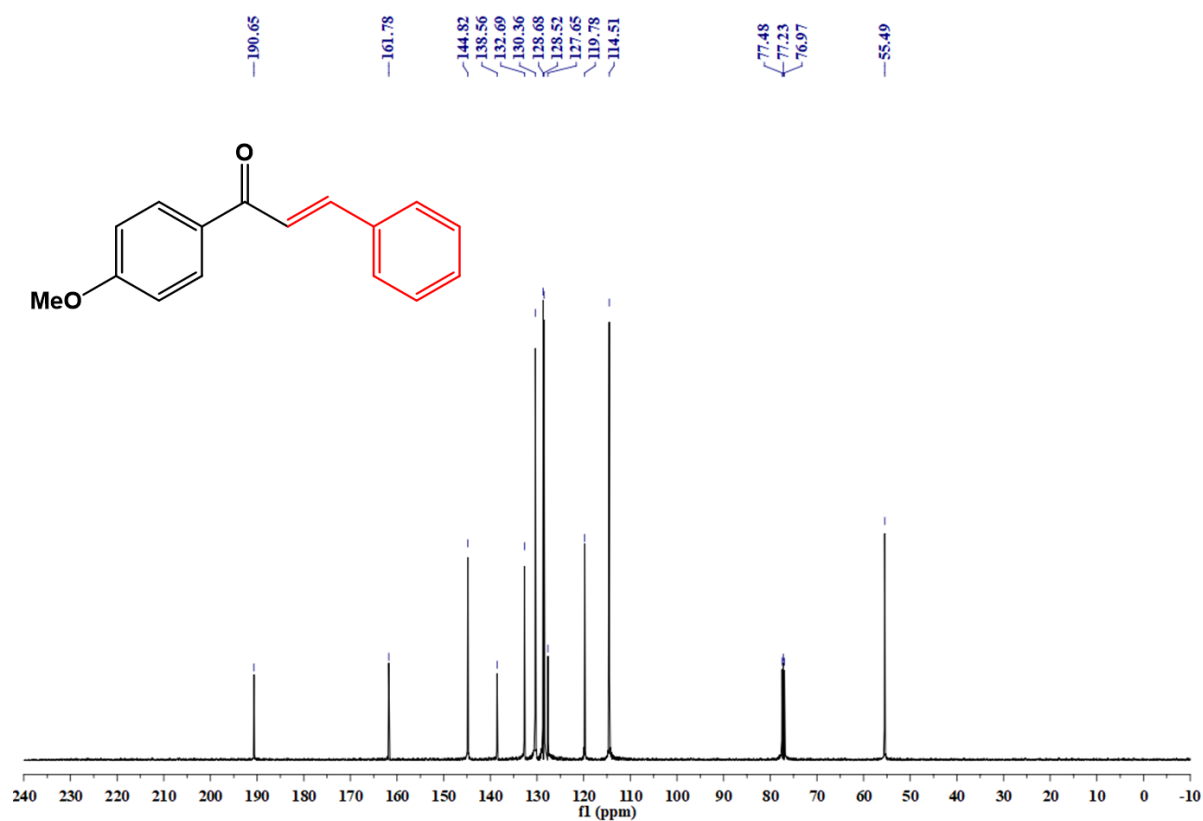


Figure S222: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **3b** in CDCl_3 at 126 MHz.

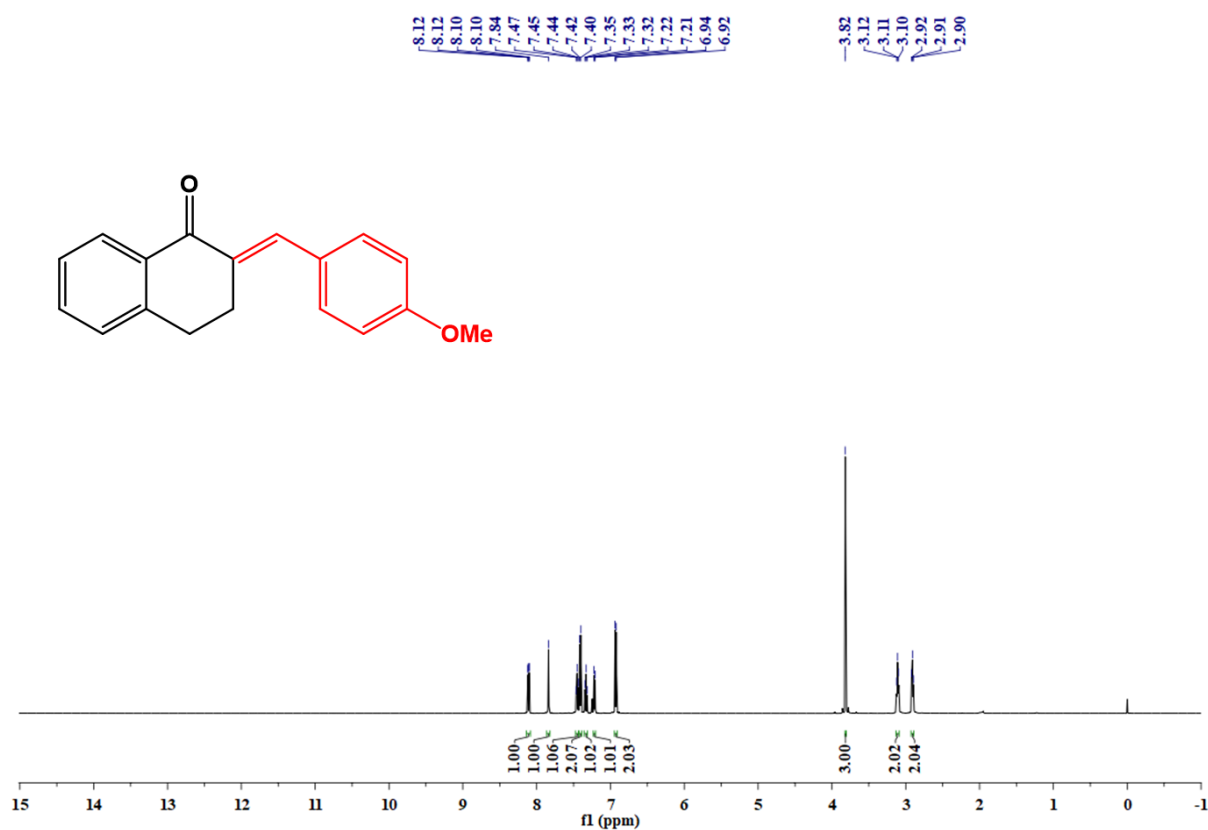


Figure S223: ^1H NMR spectrum of **6b** in CDCl_3 at 500 MHz.

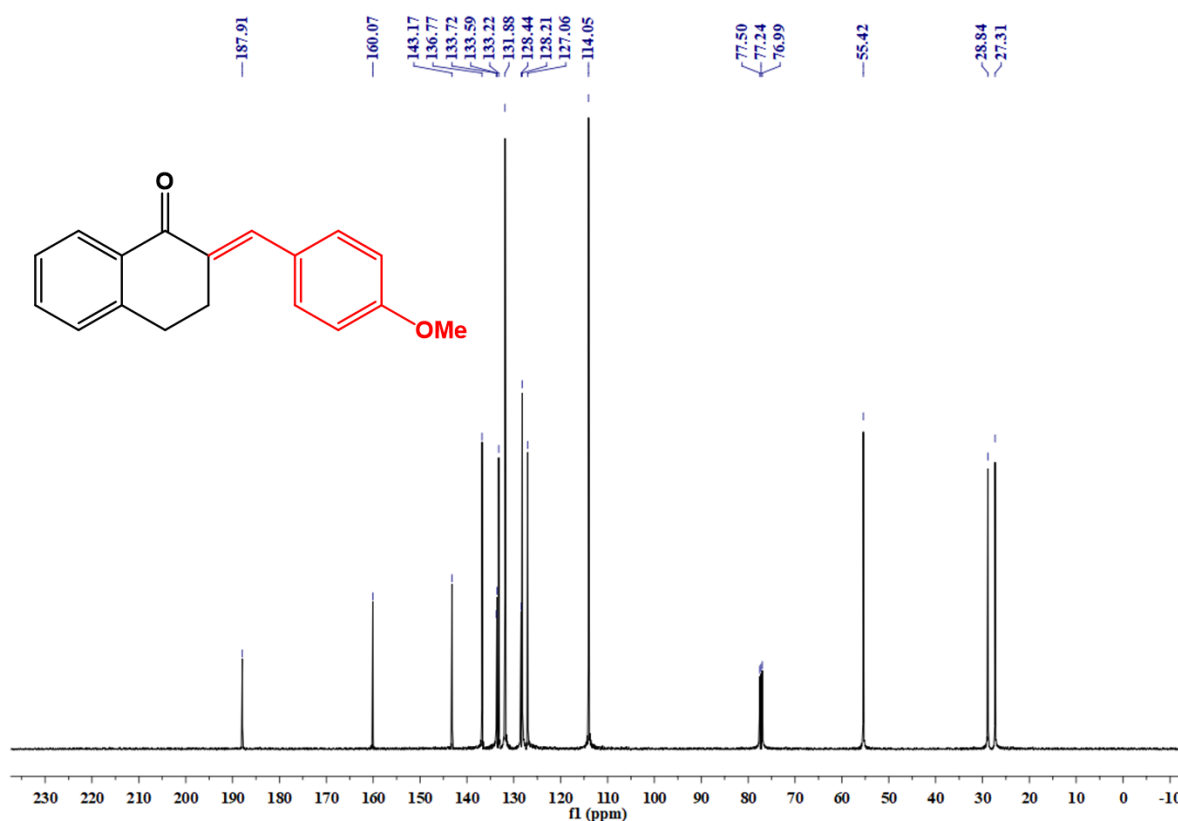


Figure S224: $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of **6b** in CDCl_3 at 126 MHz.

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