Supporting Information

C70 Fullerene Catalyzed Photo-induced Aerobic Oxidation of Benzyl-amines to Imines and Aldehydes

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Table of Contents

1.1. Experimental Setup for photocatalytic reaction

Experimental setup used for carrying out C_{70} fullerene photocatalyzed aerobic oxidation of benzylic amines is shown in figure 1. Here we used two Kessil LED Photoredox Light PR160 470 nm for photo irradiation with 100% intensity of light. We used Fisher brand Disposable Borosilicate Glass Tubes to carry out reactions and reaction vials were placed at a distance of 6 cm from the light source. Also, during the light irradiation, the temperature of the reaction setup was reached upto 35°C. Figure shows only one reaction vial under irradiation; however, we used six reaction vials at a time under this setup at 0.2 mmol scale.

Figure S1: Experimental setup for 0.2 mmol scale reaction.

2. Optimization Study: Table S1

Entry	Catalyst (mol%)	hv	O_2/Air	Solvent	Time	3a NMR Yield (%)
1	C_{70} (0.05)	Blue Light	O_2	$CHCl_3$	24h	36
2	C_{70} (0.05)	Blue Light	O_2	MeOH	24h	28
3	C_{70} (0.05)	Blue Light	O_2	1,4-Dioxane	24h	32
4	C_{70} (0.05)	Blue Light	O_2	THF	24h	20
5	C_{70} (0.05)	Blue Light	O_2	Toluene	24h	16
6	C_{70} (0.05)	Blue Light	O_2	Acetone	24h	38
7	C_{70} (0.05)	Blue Light	O_2	ACN	12h	64
8	C_{70} (0.05)	Blue Light	O_2	H_2O	24h	Traces
9	C_{70} (0.05)	Blue Light	O_2	HFIP	24h	n.d.

3. Mechanistic Study

3.1. Measurement of λ_{max} of C_{70} fullerene catalyst

Ultraviolet–visible absorption experiments were performed using Thermo Scientific NanoDrop 2000 UV-Vis spectrophotometer. In experiment, the sample was combined in $CHCl_3$ in 1.0 cm quartz cuvette.

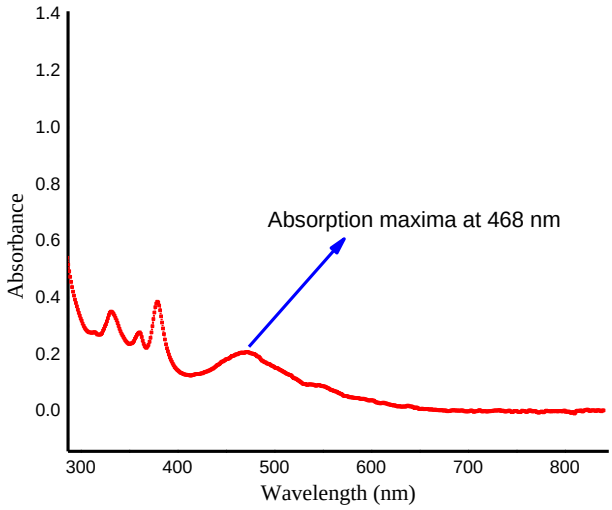


Figure 5: UV-Visible absorption spectra of C70 fullerene.

3.2. Reactive oxygen species (ROS) quenching experiments

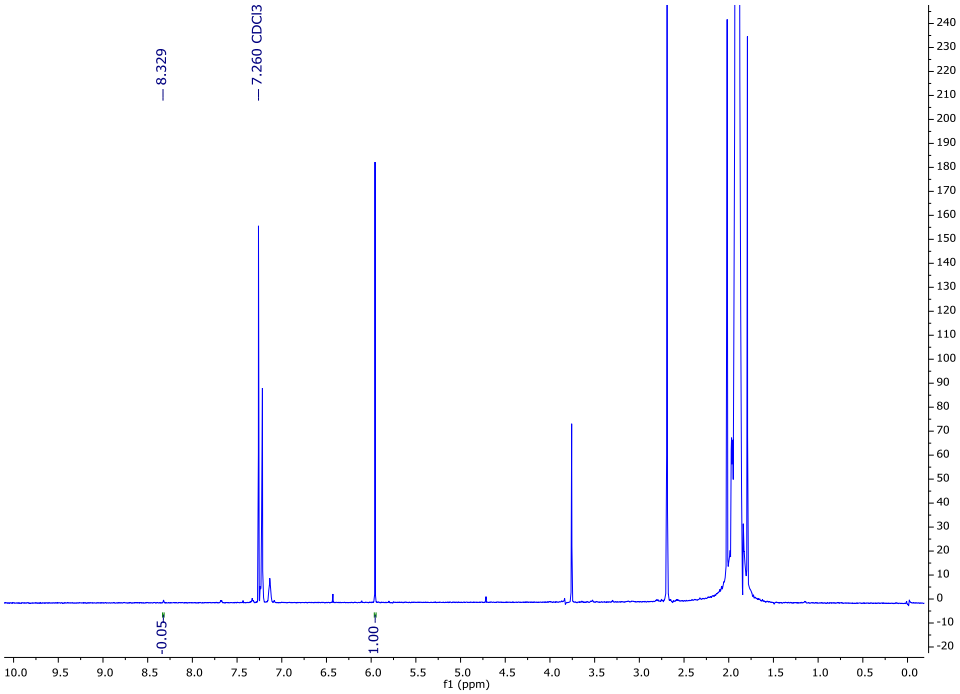


Figure S2: NMR spectrum of crude reaction mixture while using **DABCO** as ROS quencher.

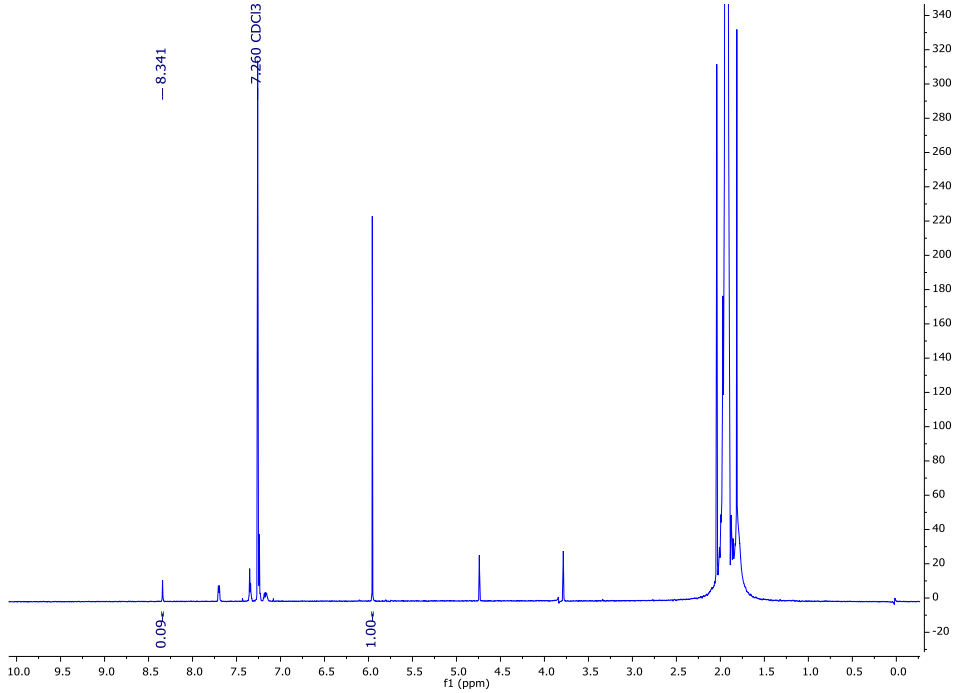


Figure S3: NMR spectrum of crude reaction mixture while using **benzoquinone** as ROS quencher.

3.2. Radical quenching quenching experiments

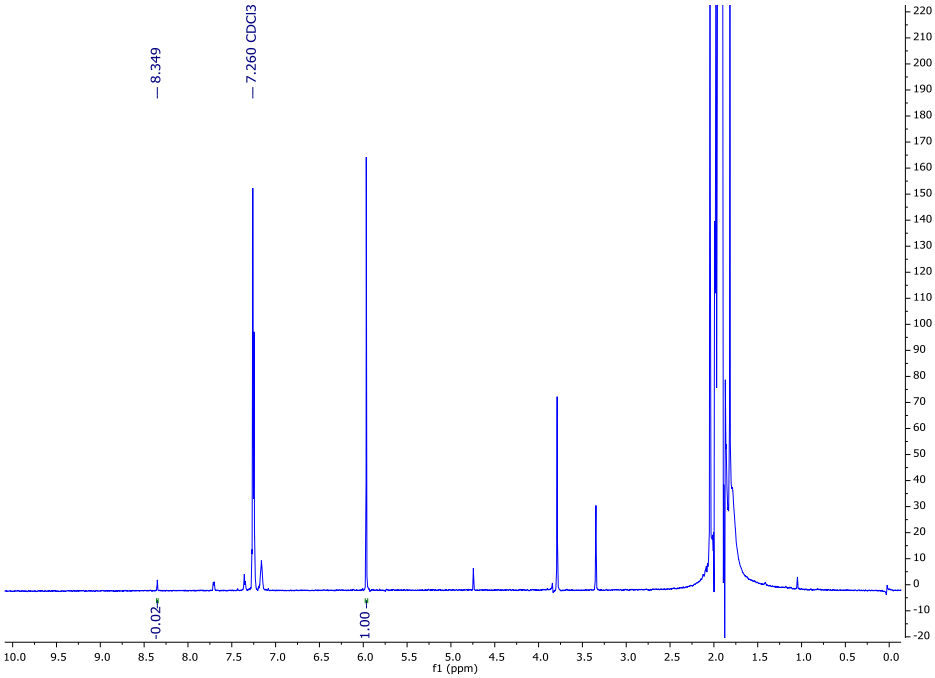


Figure S4: NMR spectrum of crude reaction mixture while using **TEMPO** as radical quencher.

3.3. ^{18}O-Labeling experiment

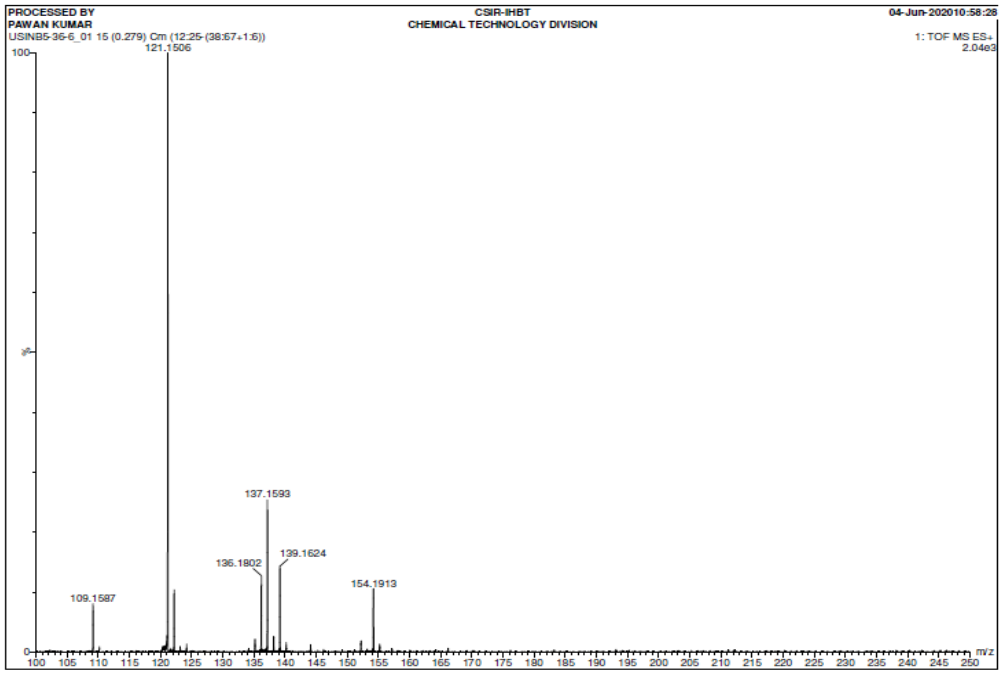


Figure S5: LC-MS chromatogram of crude reaction showing the incorporation of ^{18}O in oxidized product.

3.4. Reaction with different Composition of reaction solvents CH_3CN and $CHCl_3$

To the oven-dried screw cap reaction vials charged with a spin vane magnetic stir-bar, 84 µL of C_{70} solution (1 mg/ 2 mL in toluene) (0.05 mol%) was added in three different vials and dried over rotary evaporator. To this, primary benzylic amine (10.9 µL, 0.1 mmol) and different composition of CH_3CN+$CHCl_3$ (2 mL) were added. The reaction vials were closed with screw cap, purged with oxygen and kept for vigorous stirring under blue LED light at room temperature for 24 h. After completion, the reactions were dried over rotary evaporator and crude reactions were analyzed with the help of NMR using tetrachloroethane as an internal standard.

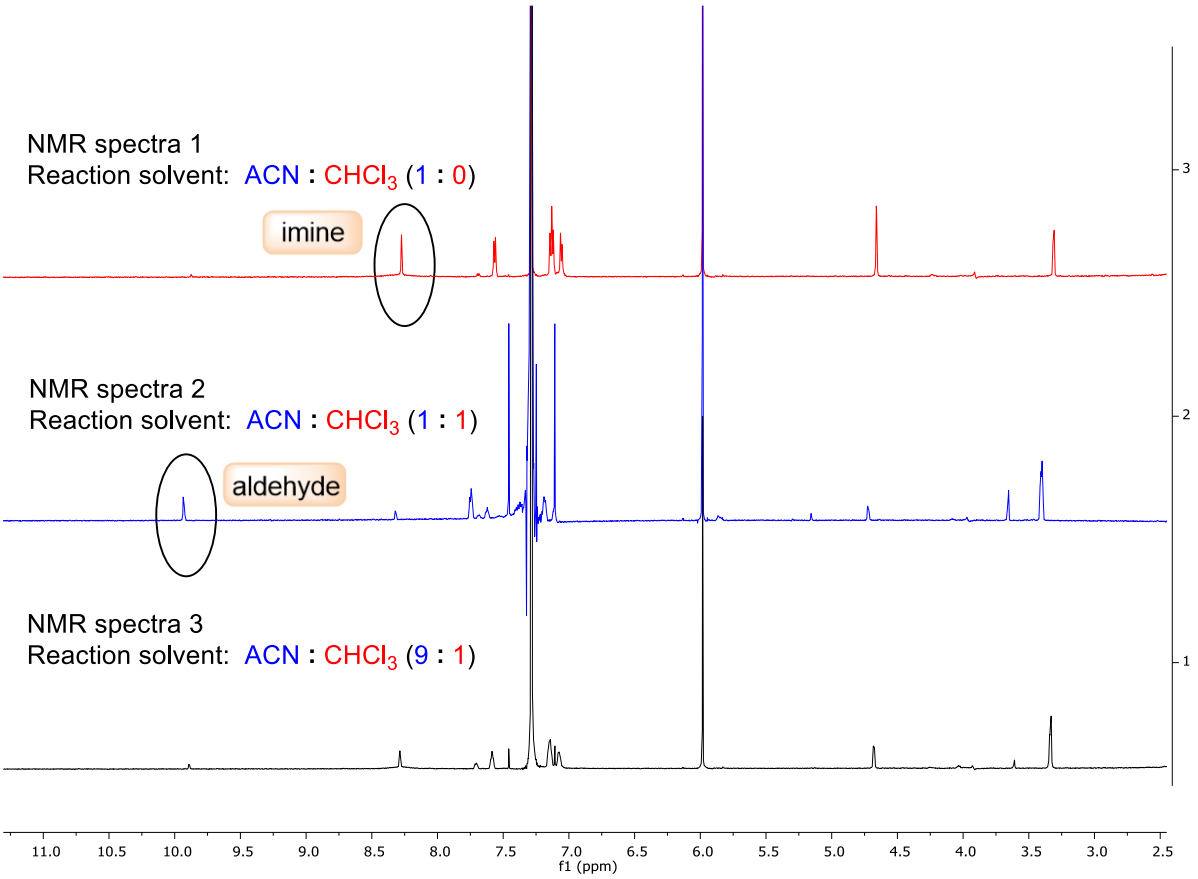


Figure S6. NMR yield comparison of 3a and 4a with different ratio of solvents CH_3CN and $CHCl_3$.

3.5. Reaction of imine product 3p under standard condition using $CHCl_3$ solvent.

To the oven-dried screw cap reaction vial charged with a spin vane magnetic stir-bar, 84 µL of C_{70} solution (1 mg/ 2 mL in toluene) (0.05 mol%) was added in vial and dried over rotary evaporator. To this, imine product **3p** (39.5 mg, 0.1 mmol) and $CHCl_3$ (2 mL) were added. The reaction vial was closed with screw cap, purged with oxygen and kept for vigorous stirring under blue LED light at room temperature for 24 h. After completion, the reaction was dried over rotary evaporator and crude reaction was analyzed with the help of NMR using tetrachloroethane as an internal standard.

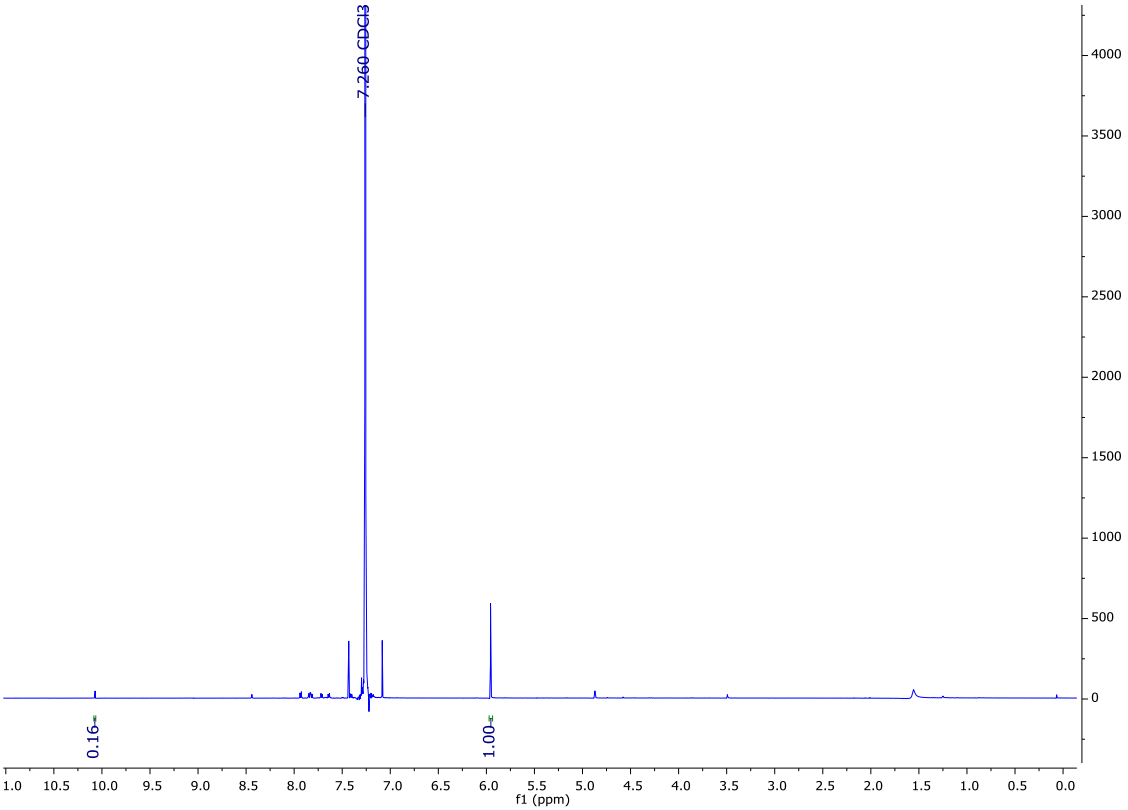


Figure S7. NMR spectrum of the reaction of imine product **3p** in solvent $CHCl_3$.

4. Greeen Metrics

The following formulae were used for calculating Atom Economy (AE), Atom Efficiency (AEf), Carbon Efficiency (CE), Reaction Mass Efficiency (RME), Optimum Efficiency (OE), Mass Productivity (MP), Mass Intensity (MI) and Process Mass Intensity (PMI), E factor, Solvent and Water Intensity. These calculations not includes the solvents used for the isolation of the compounds.

AE = **Molecular weight of product /Total molecular weight of reactants X 100**

AEf = **AE X yield**%

CE =**Amount of carbon in the product /Total carbon present in reactants X 100**

RME = **Mass of isolated product/Total mass of reactants X 100**

OE =**RME AE X 100**

MI =**Total mass of input material in a process or process step/Mass of product**

PMI = Total mass of input material in the whole process/Mass of product

MP = 1 PMI X 100

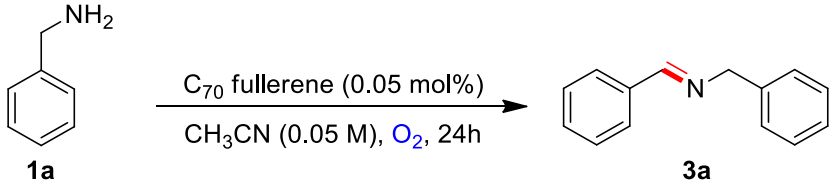
E Factor = PMI − 1

SI = Total mass of solvents excl.water in the whole process/Mass of product

WI = Total mass of water used in the whole process/Mass of product

Reaction 1: C_{70} fullerene catalyzed synthesis of imines from corresponding primary benzylic amines

To an oven-dried screw cap reaction vial charged with a spin vane magnetic stir-bar, 168 µL of C_{70} solution (1 mg/ 2 mL in toluene) (0.05 mol%) was added and dried over rotary evaporator. To this, primary benzylic amine (21.9 µL, 0.2 mmol) and acetonitrile (4 mL) were added. The reaction vial was closed with screw cap, purged with oxygen and kept for vigorous stirring under blue LED light at room temperature for 24 h. After completion, the reaction mixture was dried over rotary evaporator and purified by column chromatography using silica gel (60-120 mesh size) as stationary phase and *n*-hexane: EtOAc as eluent. Additionally, triethylamine was added to the solvent system to avoid any kind of degradation of imine product.



Reaction 2: C_{70} fullerene catalyzed synthesis of benzaldehydes from corresponding benzylic amines

To an oven-dried screw cap reaction vial charged with a spin vane magnetic stir-bar, 168 µL of C_{70} solution (1 mg/ 2 mL in toluene) (0.05 mol%) was added and dried over rotary evaporator. To this, primary benzylic amine (21.9 µL, 0.2 mmol) and chloroform (4 mL) were added. The reaction vial was closed with screw cap, purged with oxygen and kept for vigorous stirring under blue LED light at room temperature for 24 h. After completion, the reaction mixture was dried over rotary evaporator and purified by flash chromatography using silica gel (230-400 mesh size) as stationary phase and *n*-hexane: EtOAc as eluent.

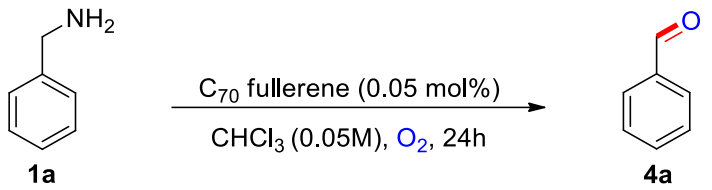


Table S2: Green Metrics.

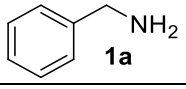
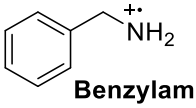
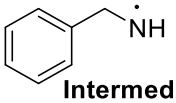
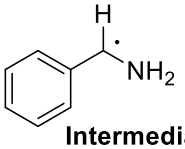
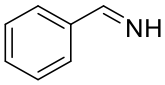
Green parameters	**Imine Synthesis**	**Benzaldehyde synthesis**
Steps	1	1
Molecular weight of Product	195.26	106.12
yield%	98	55
Molecular weight of reactants	214.30	214.3
Atom Economy	**91%**	**49%**
Atom Efficiency	**89.3**	**27.2**
Carbon in Product	0.001372	0.00077
Total Carbon in Reactants	0.0014	0.0014
Carbon Efficiency	**98%**	**55%**
Mass of Product	0.0191 g	0.01167 g
Total Mass of Reactants	0.02142 g	0.02142 g
Reaction Mass Efficiency	**89.16%**	**54.48%**
Optimum efficiency	**97.86%**	**110.02%**
Total mass of input Material	4.067 g	5.113 g
Mass of Product	0.0191 g	0.0116 g
Mass Intensity	**212.95**	**438.17**
Process Mass Intensity	**78.35**	**189.7**

Mass Productivity	**0.47%**	**0.23%**
E-Factor	**211.95**	**437.17**
Total mass of solvent	3.144	4.190
Water mass	0	0
Solvent Intensity	**164.60**	**359.04**
Water Intensity	**0**	**0**

5. Computational Study

5.1 General Consideration: DFT calculations were done with Gaussian 16 program.[1] All energies were computed through B3LYP method in which atoms were specified by 6-31+G (d, p) basis set.[2] Energy of each structure were taken after performing opt+freq job in Guassian software. All optimization were performed in gas phase without considering any solvent model. Free enthalpy of all strucutres has been mentioned in Table S3.

Table S3. Computational data of various proposed reaction intermediates.

Structures	ΔG (Hartree)	ΔG (Kcal/mol)	Relative ΔG (Kcal/mol)
 **1a**	-326.825118	-205085.901691	0
 **Benzylamine radical cation**	-326.529380	-204900.32325	+185.57
 **Intermediate A**	-326.173194	-204676.81312	+409.09
 **Intermediate B**	-326.204170	-204696.25085	+389.65
 **Intermediate I**	-325.630134	-204336.03775	+749.86

5.2 Cartesian Coordinates, Number of Imaginary Frequencies and other Absolute Energy Values for proposed reaction intermediates

5.2.1 Benzyl amine (1a)

Zero point energies, enthalpies, and free energies

Zero-point correction= 0.145941 (Hartree/Particle)
Thermal correction to Energy= 0.153126
Thermal correction to Enthalpy= 0.154070
Thermal correction to Gibbs Free Energy= 0.114086
Sum of electronic and zero-point Energies= -326.793264
Sum of electronic and thermal Energies= -326.786078
Sum of electronic and thermal Enthalpies= -326.785134
Sum of electronic and thermal Free Energies= -326.825118

Optimized geometry for structures 1a

C	-1.42243300	-1.33195700	0.05511900
C	-0.05548000	-1.07893000	0.18662700
C	0.43264000	0.23620400	0.15780800
C	-0.47744500	1.28764300	-0.01263300
C	-1.84732300	1.03745000	-0.14103800
C	-2.32422300	-0.27483500	-0.10700600
H	-1.78535200	-2.35595900	0.07920700
H	0.64898000	-1.89668200	0.30236900
H	-0.11215300	2.31171900	-0.04735500
H	-2.53767000	1.86586700	-0.27394100
H	-3.38724600	-0.47340700	-0.21035600
C	1.91258800	0.52039800	0.34034000
H	2.16613400	0.43394800	1.40559400
H	2.10581000	1.56853700	0.05869200
N	2.74738200	-0.45148100	-0.37704700
H	2.62855700	-0.35941200	-1.38286800
H	3.73132800	-0.31008400	-0.16731100

5.2.2 Benzyl amine radical cation

Zero point energies, enthalpies, and free energies

Zero-point correction= 0.145093 (Hartree/Particle)

Thermal correction to Energy= 0.152493
Thermal correction to Enthalpy= 0.153437
Thermal correction to Gibbs Free Energy= 0.112802
Sum of electronic and zero-point Energies= -326.497090

Sum of electronic and thermal Energies= -326.489690
Sum of electronic and thermal Enthalpies= -326.488746
Sum of electronic and thermal Free Energies= -326.529380

Optimized geometry for Benzyl amine radical cation

C	-1.57515200	1.22510800	0.07182200
C	-0.20990400	1.22995000	-0.15905100
C	0.48123300	0.00024300	-0.32629500
C	-0.20959900	-1.22976500	-0.15932900
C	-1.57476100	-1.22540500	0.07157100
C	-2.26002600	-0.00021700	0.19426300
H	-2.11836800	2.16049500	0.15529700
H	0.32434000	2.17030100	-0.26308300
H	0.32501500	-2.16988500	-0.26361700
H	-2.11785000	-2.16087700	0.15487000
H	-3.32785900	-0.00048900	0.39050700
C	1.97073300	0.00044800	-0.61683900
H	2.30256500	-0.89056900	-1.15076700
H	2.30241000	0.89227400	-1.14965400
N	2.50535800	-0.00027200	0.73175900
H	2.51947200	-0.85969300	1.27285400
H	2.51761800	0.85817800	1.27443900

5.2.3 Reaction Intermediate A

Zero point energies, enthalpies, and free energies

Zero-point correction= 0.130923 (Hartree/Particle)
Thermal correction to Energy= 0.138223
Thermal correction to Enthalpy= 0.139167
Thermal correction to Gibbs Free Energy= 0.098142
Sum of electronic and zero-point Energies= -326.140413
Sum of electronic and thermal Energies= -326.133114
Sum of electronic and thermal Enthalpies= -326.132170
Sum of electronic and thermal Free Energies= -326.173194

Optimized geometry for Reaction Intermediate A

C	-1.60969800	1.18008300	0.09227000
C	-0.23300100	1.22598400	-0.14335800
C	0.50971900	0.04485900	-0.28225200
C	-0.15553100	-1.18646600	-0.18144700
C	-1.53133200	-1.23546600	0.05465100

C	-2.26221700	-0.05169300	0.19253700
H	-2.17199100	2.10432400	0.19086300
H	0.26729300	2.18821800	-0.22678400
H	0.40890200	-2.10908500	-0.29015900
H	-2.03257000	-2.19640400	0.12831000
H	-3.33260000	-0.08903700	0.37281500
C	2.01328900	0.07994400	-0.48970600
H	2.32227600	-0.65780800	-1.23945800
H	2.30237600	1.06962000	-0.88815600
N	2.78934200	-0.14778300	0.71805300
H	2.32354000	0.38118400	1.47003300

5.2.4 Reaction Intermediate B

Zero point energies, enthalpies, and free energies
Zero-point correction= 0.132310 (Hartree/Particle)

Thermal correction to Energy= 0.139478
Thermal correction to Enthalpy= 0.140422
Thermal correction to Gibbs Free Energy= 0.100603
Sum of electronic and zero-point Energies= -326.172464
Sum of electronic and thermal Energies= -326.165296
Sum of electronic and thermal Enthalpies= -326.164352
Sum of electronic and thermal Free Energies= -326.204170

Optimized geometry for structures B

C	1.36360200	1.34649000	-0.00666600
C	-0.00143300	1.08094200	-0.02663400
C	-0.48838900	-0.26072000	-0.01712900
C	0.48532800	-1.30501900	-0.00113000
C	1.84244700	-1.02489600	0.01689300
C	2.30053100	0.30454200	0.01774900
H	1.70436600	2.37855800	-0.01728500
H	-0.69589500	1.91534700	-0.07062500
H	0.14593100	-2.33808400	0.00124300
H	2.55722800	-1.84336900	0.03055200
H	3.36416200	0.52062800	0.03184000
C	-1.86360500	-0.58736700	-0.01301800
H	-2.17279800	-1.62513900	-0.07081200
N	-2.89005400	0.34209500	-0.03771200

H	-3.80821300	0.00687600	0.21555400
H	-2.69529200	1.26668300	0.32312600

5.2.5 Reaction Intermediate I

Zero point energies, enthalpies, and free energies

Zero-point correction= 0.122450 (Hartree/Particle)
Thermal correction to Energy= 0.128898
Thermal correction to Enthalpy= 0.129843
Thermal correction to Gibbs Free Energy= 0.091784
Sum of electronic and zero-point Energies= -325.599469
Sum of electronic and thermal Energies= -325.593020
Sum of electronic and thermal Enthalpies= -325.592076
Sum of electronic and thermal Free Energies= -325.630134

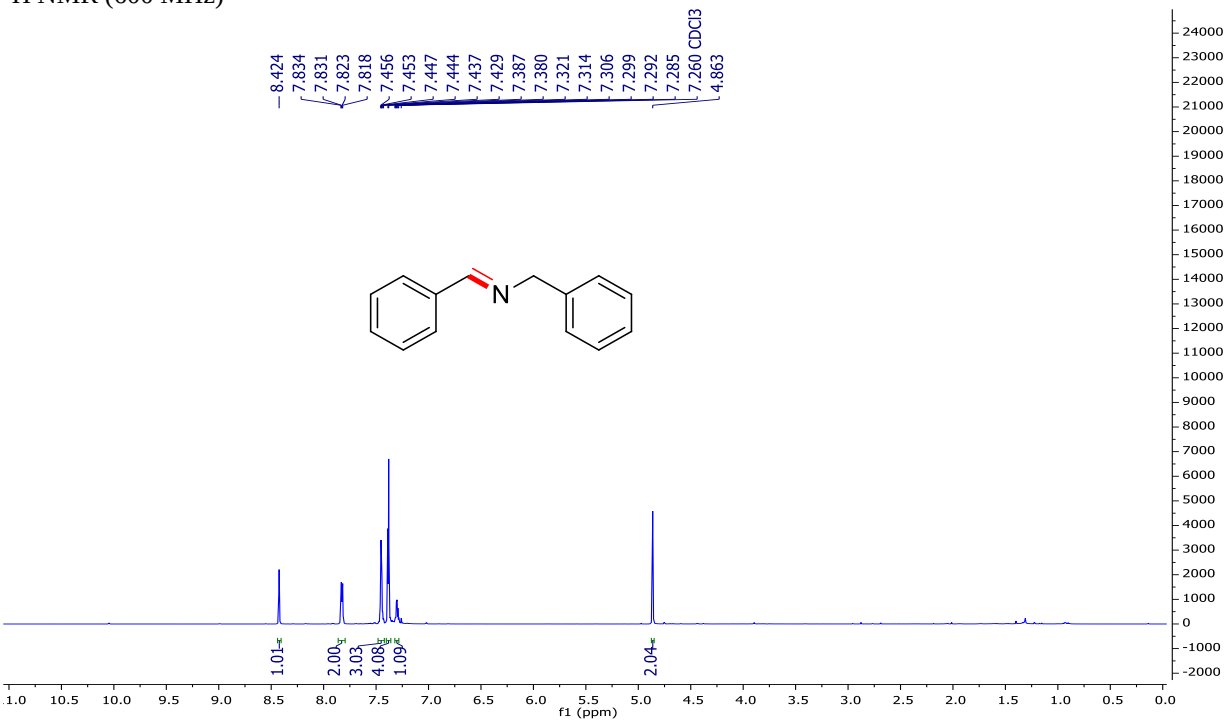
Optimized geometry for Reaction Intermediate I

C	-1.57515200	1.22510800	0.07182200
C	-0.20990400	1.22995000	-0.15905100
C	0.48123300	0.00024300	-0.32629500
C	-0.20959900	-1.22976500	-0.15932900
C	-1.57476100	-1.22540500	0.07157100
C	-2.26002600	-0.00021700	0.19426300
H	-2.11836800	2.16049500	0.15529700
H	0.32434000	2.17030100	-0.26308300
H	0.32501500	-2.16988500	-0.26361700
H	-2.11785000	-2.16087700	0.15487000
H	-3.32785900	-0.00048900	0.39050700
C	1.97073300	0.00044800	-0.61683900
H	2.30256500	-0.89056900	-1.15076700
H	2.30241000	0.89227400	-1.14965400
N	2.50535800	-0.00027200	0.73175900
H	2.51947200	-0.85969300	1.27285400
H	2.51761800	0.85817800	1.27443900

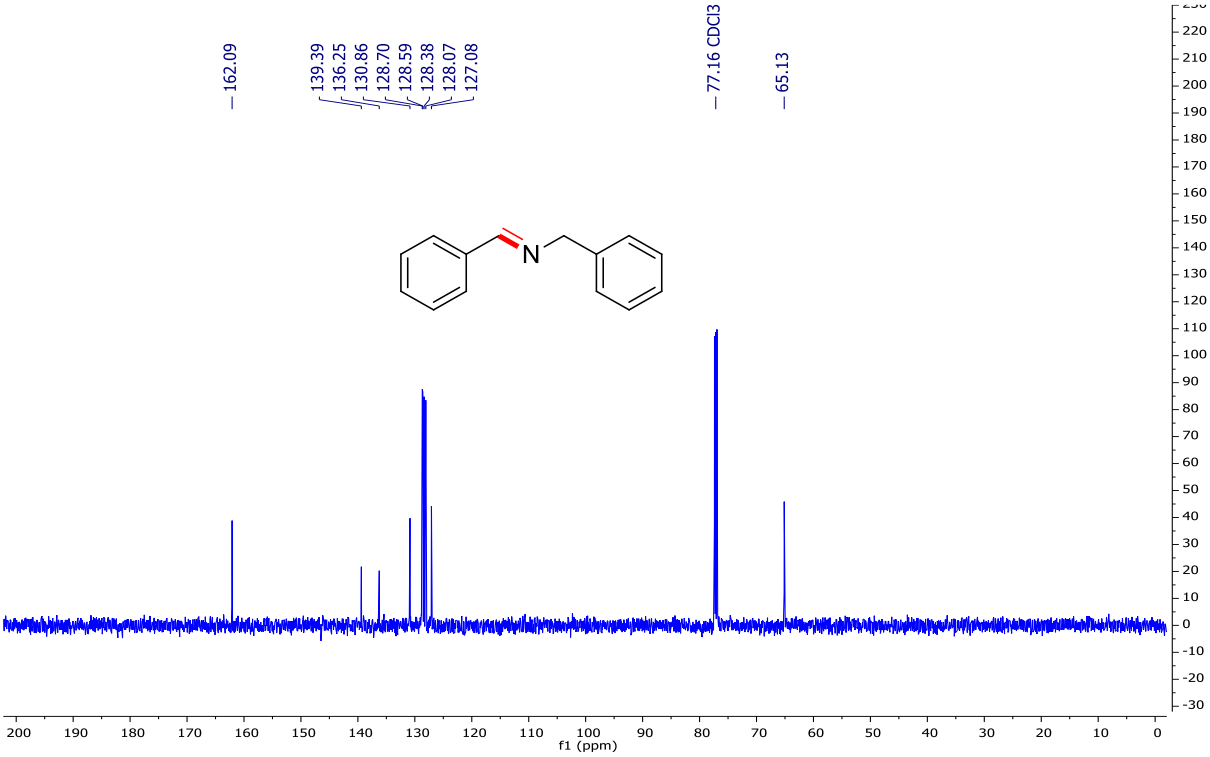
6. NMR spectra of synthesized compounds

(E)-N-benzylidene-1-phenylmethanamine (Table 2, entry **3a**)

^{1}H NMR (600 MHz)

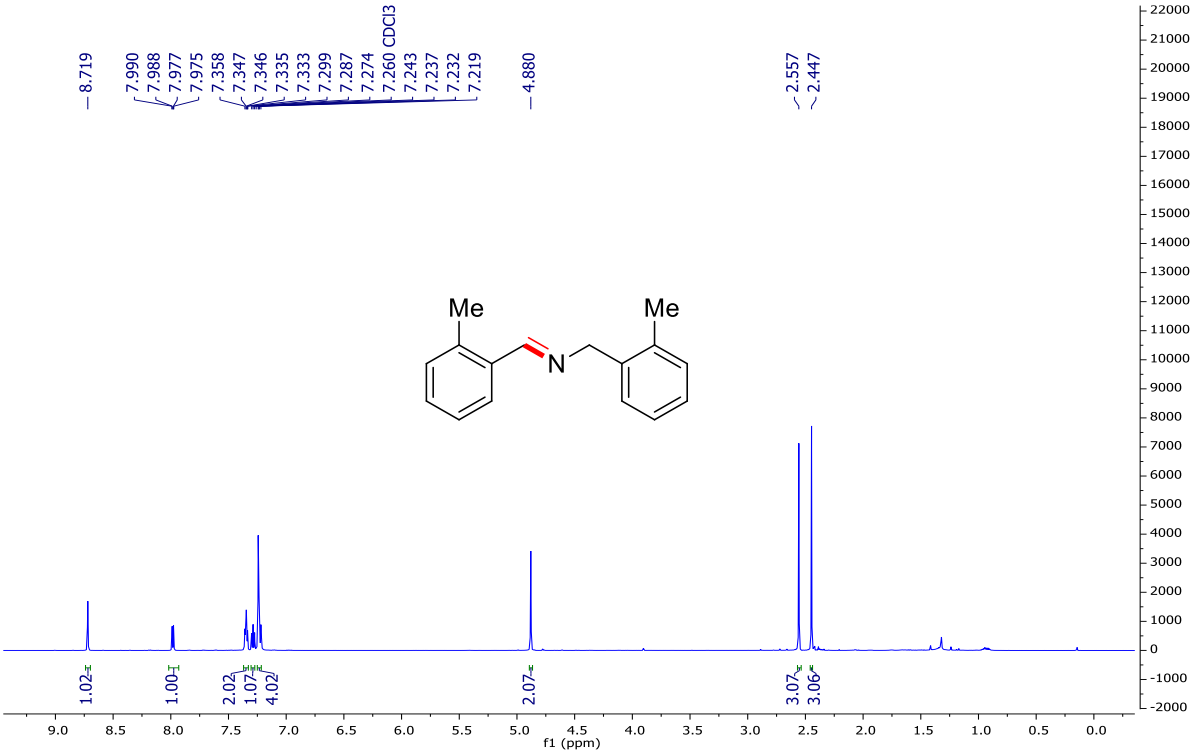


^{13}C{^{1}H} NMR (150 MHz)

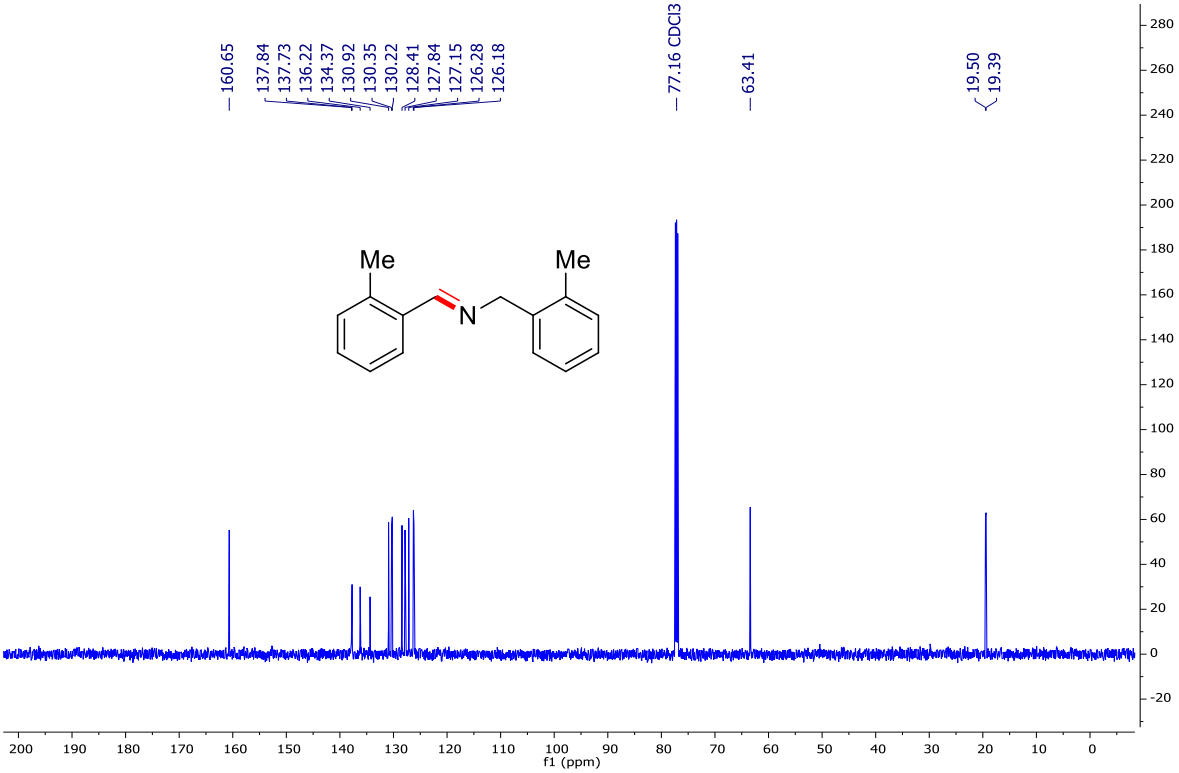


(E)-N-(2-methylbenzylidene)-1-(o-tolyl)methanamine (Table 2, entry **3b**)

^{1}H NMR (600 MHz)

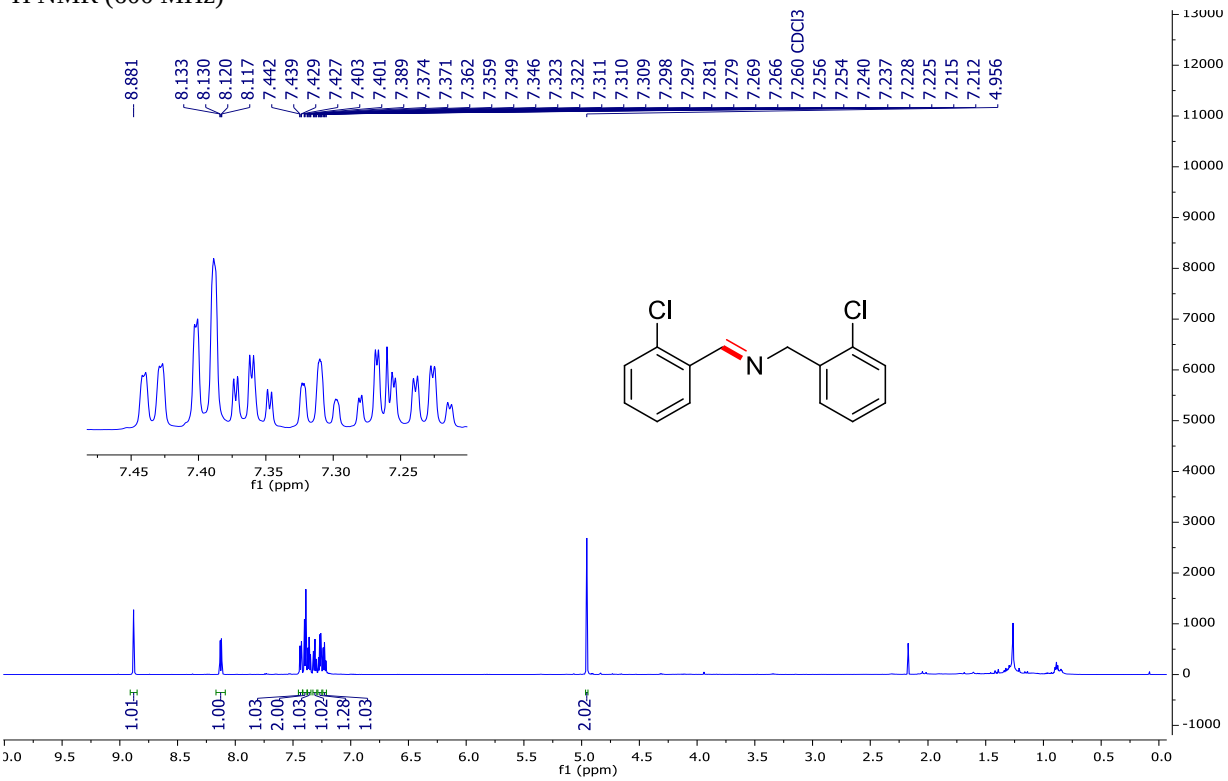


^{13}C{^{1}H} NMR (150 MHz)

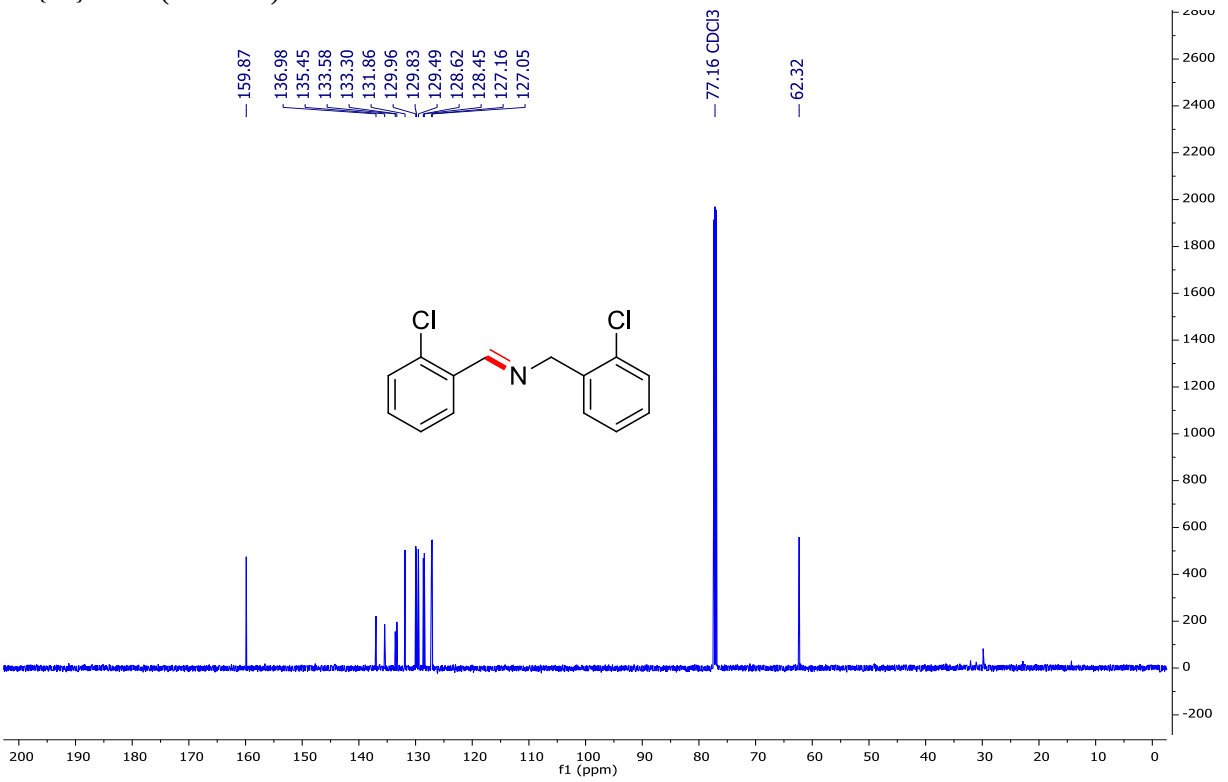


(E)-N-(2-chlorobenzylidene)-1-(2-chlorophenyl)methanamine (Table 2, entry **3c**)

^{1}H NMR (600 MHz)

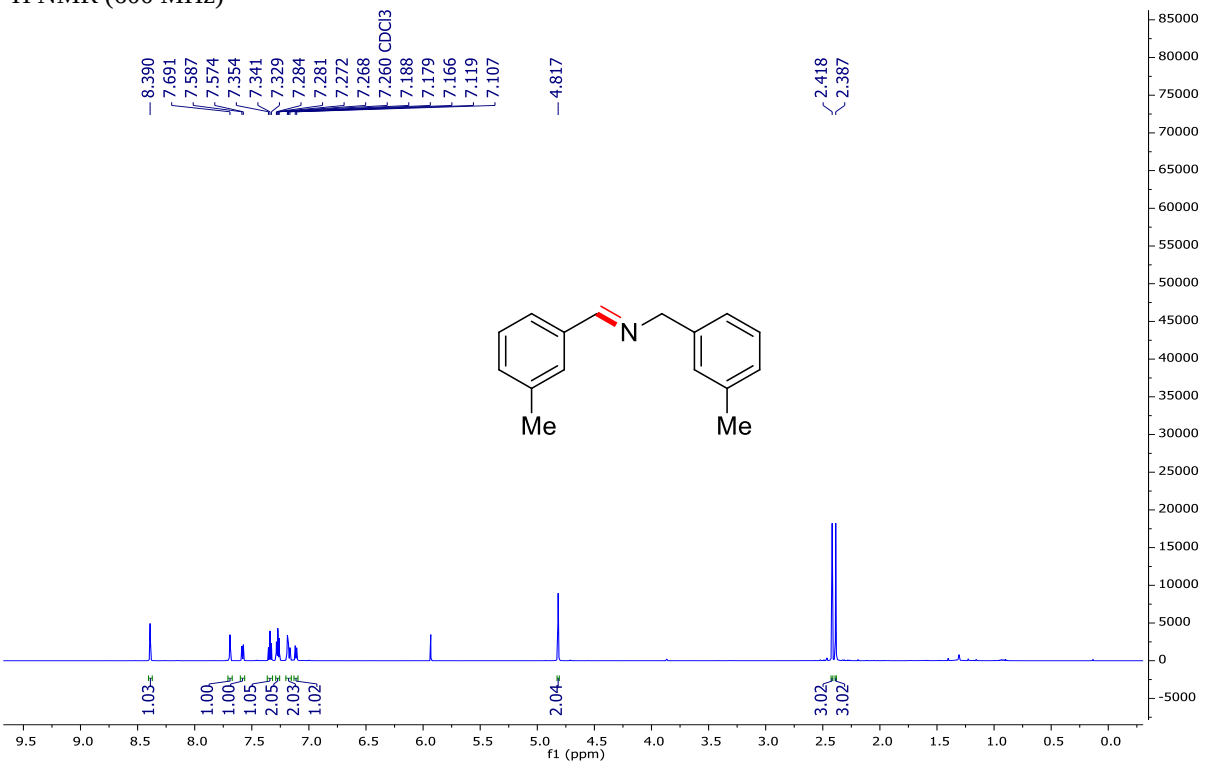


^{13}C{^{1}H} NMR (150 MHz)

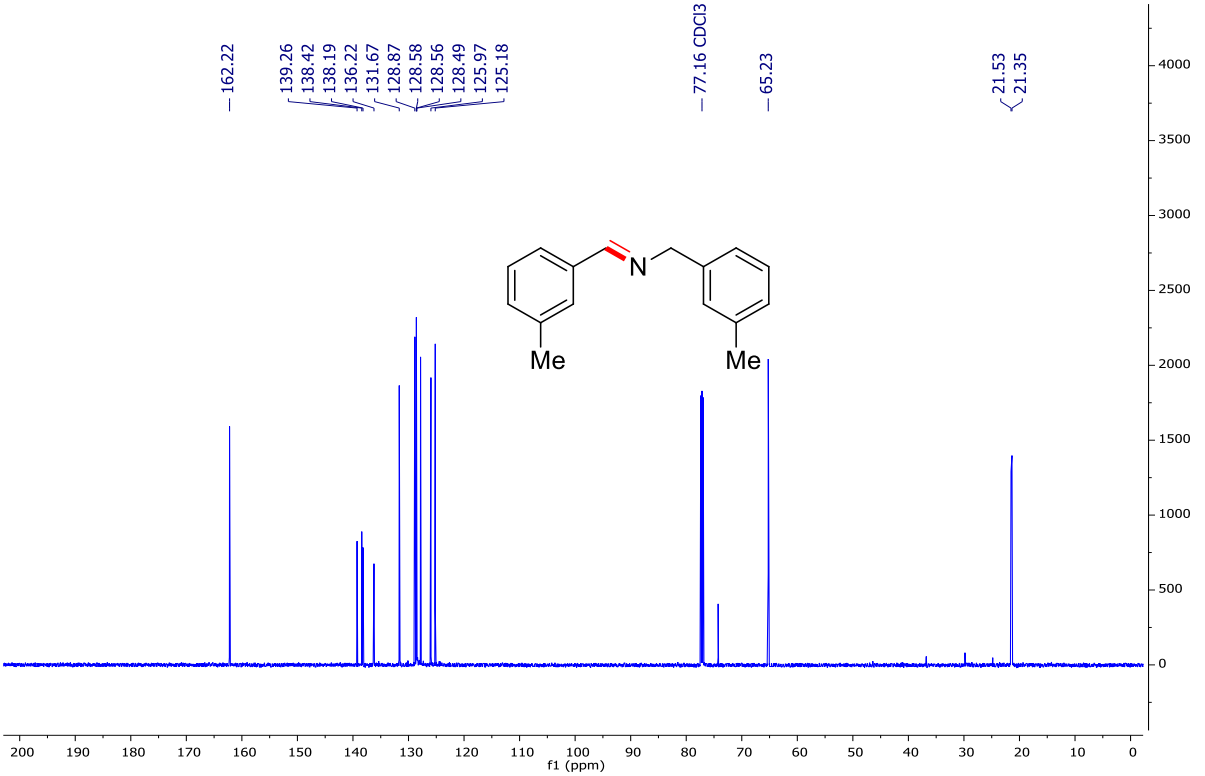


(E)-N-(3-methylbenzylidene)-1-(m-tolyl)methanamine (Table 2, entry **3d**)

^{1}H NMR (600 MHz)

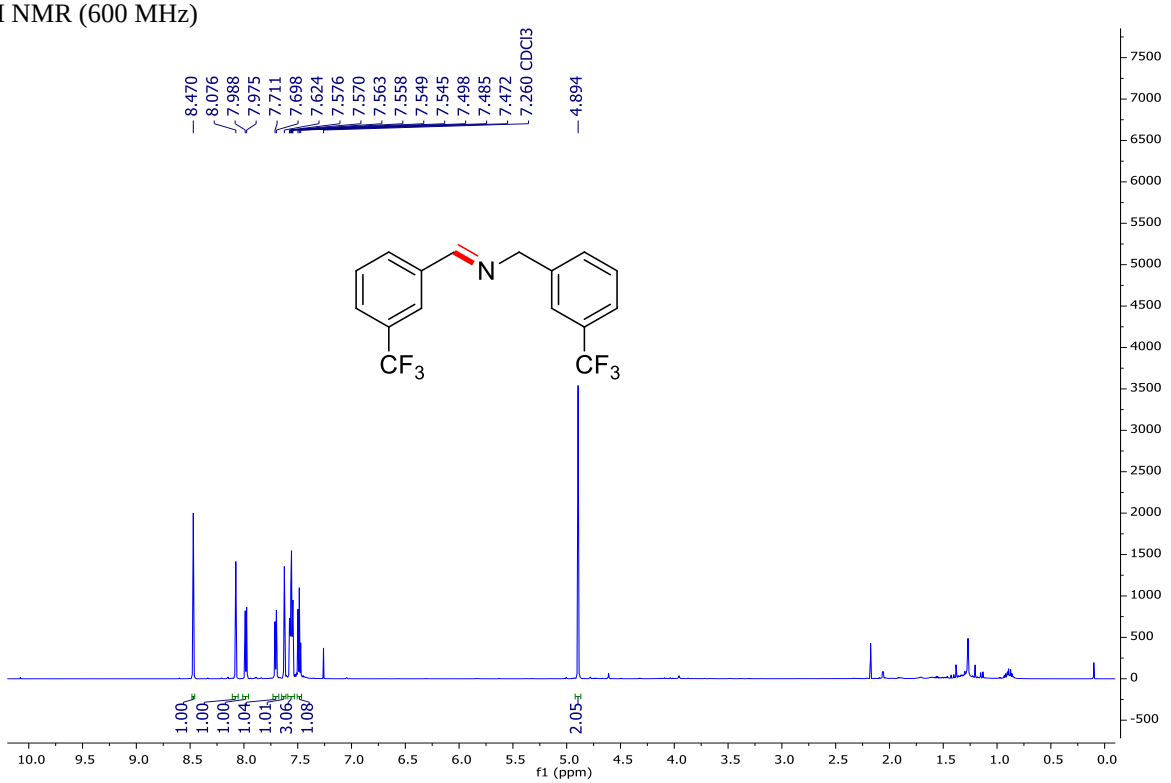


^{13}C{^{1}H} NMR (150 MHz)

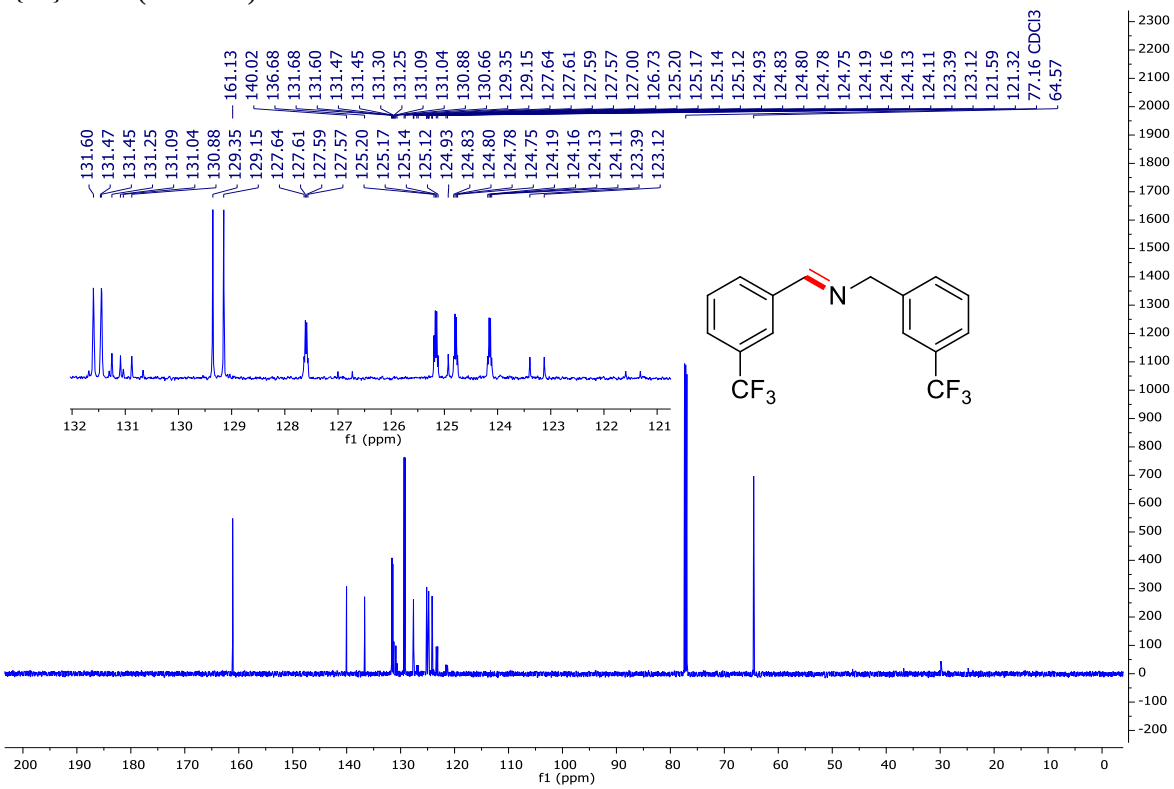


(E)-N-(3-(trifluoromethyl)benzylidene)-1-(3-(trifluoromethyl)phenyl)methanamine (Table 2, entry **3e**)

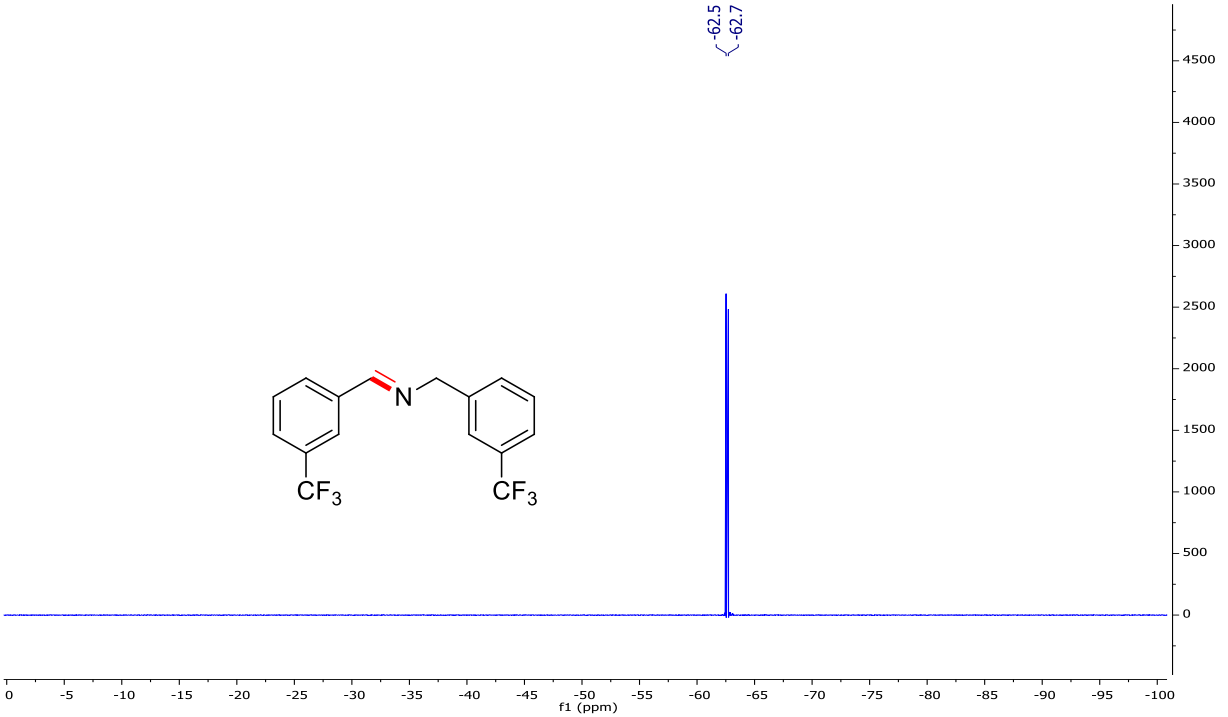
^{1}H NMR (600 MHz)



^{13}C{^{1}H} NMR (150 MHz)

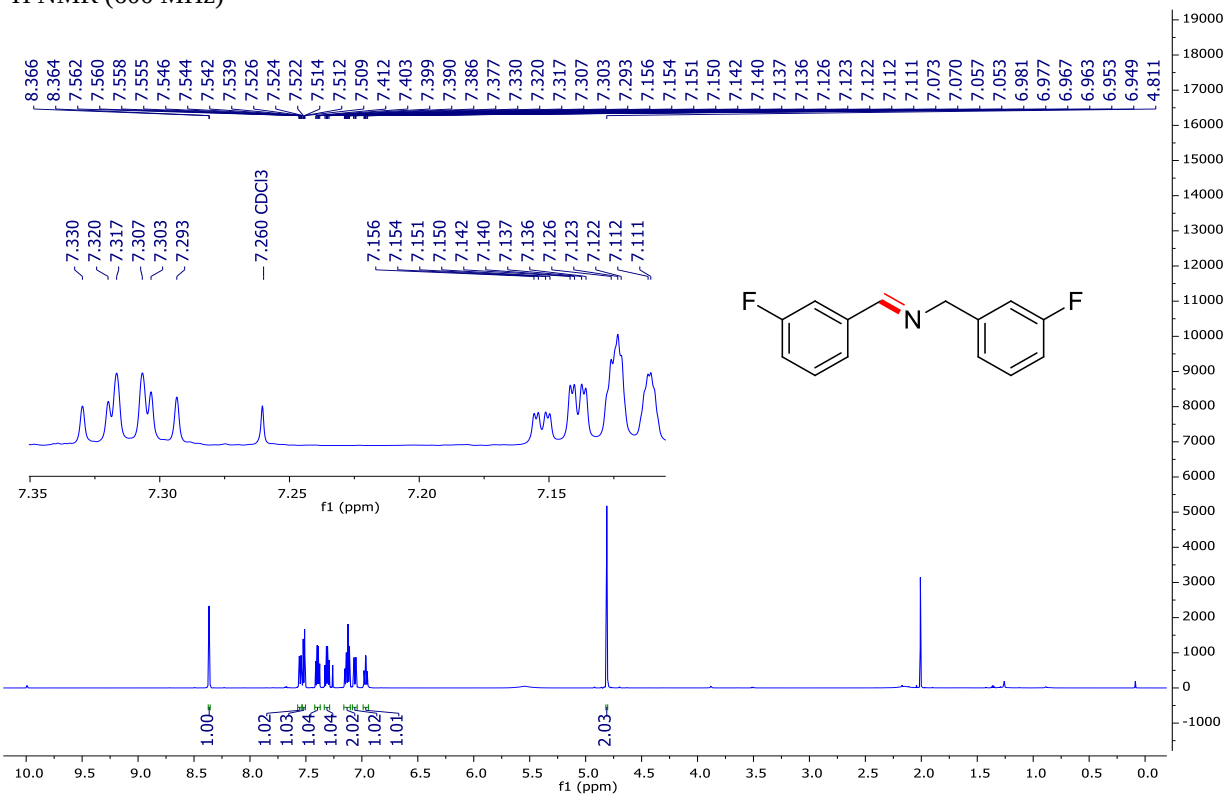


^{19}F NMR (564 MHz)

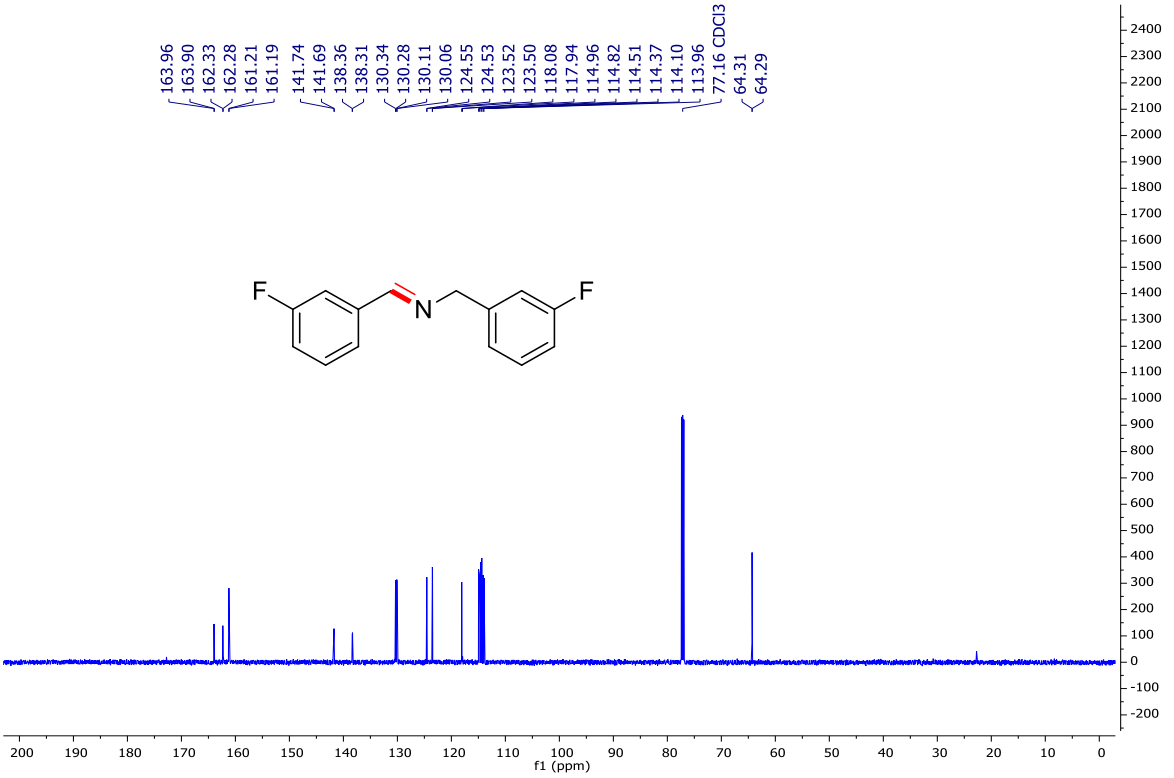


(E)-N-(3-fluorobenzylidene)-1-(3-fluorophenyl)methanamine (Table 2, entry **3f**)

^{1}H NMR (600 MHz)

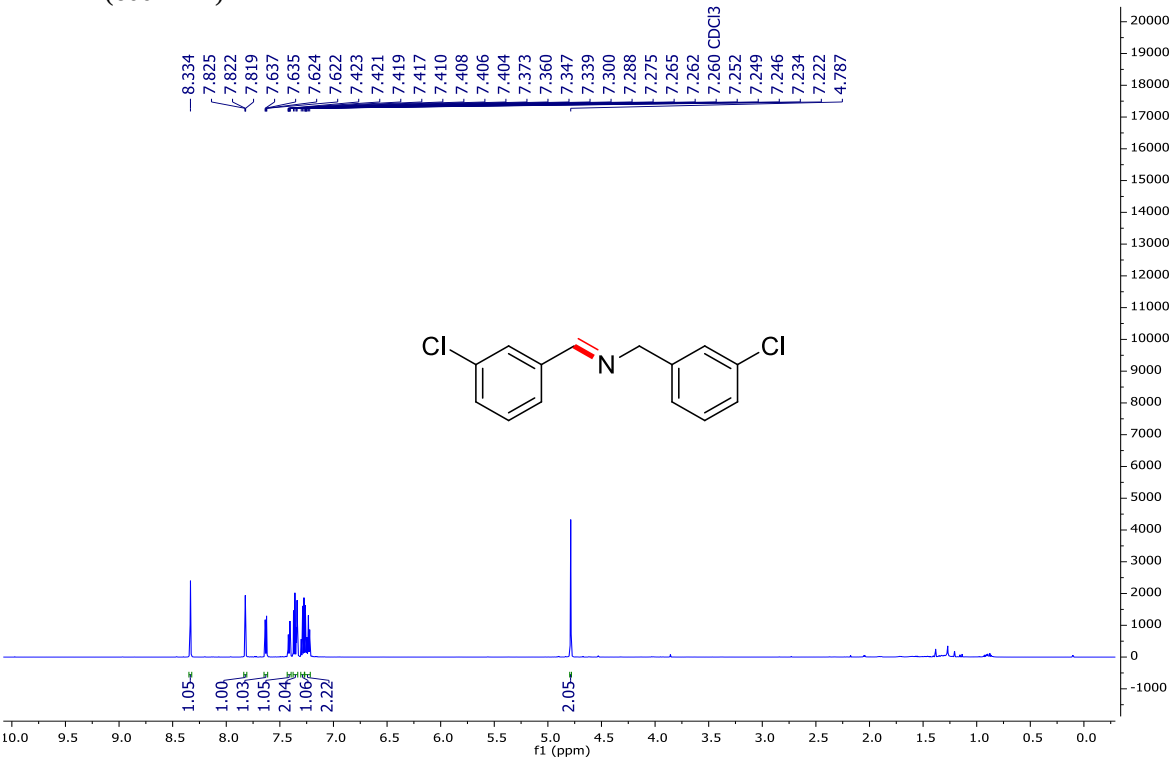


$^{13}C\{^{1}H\}$ NMR (150 MHz)

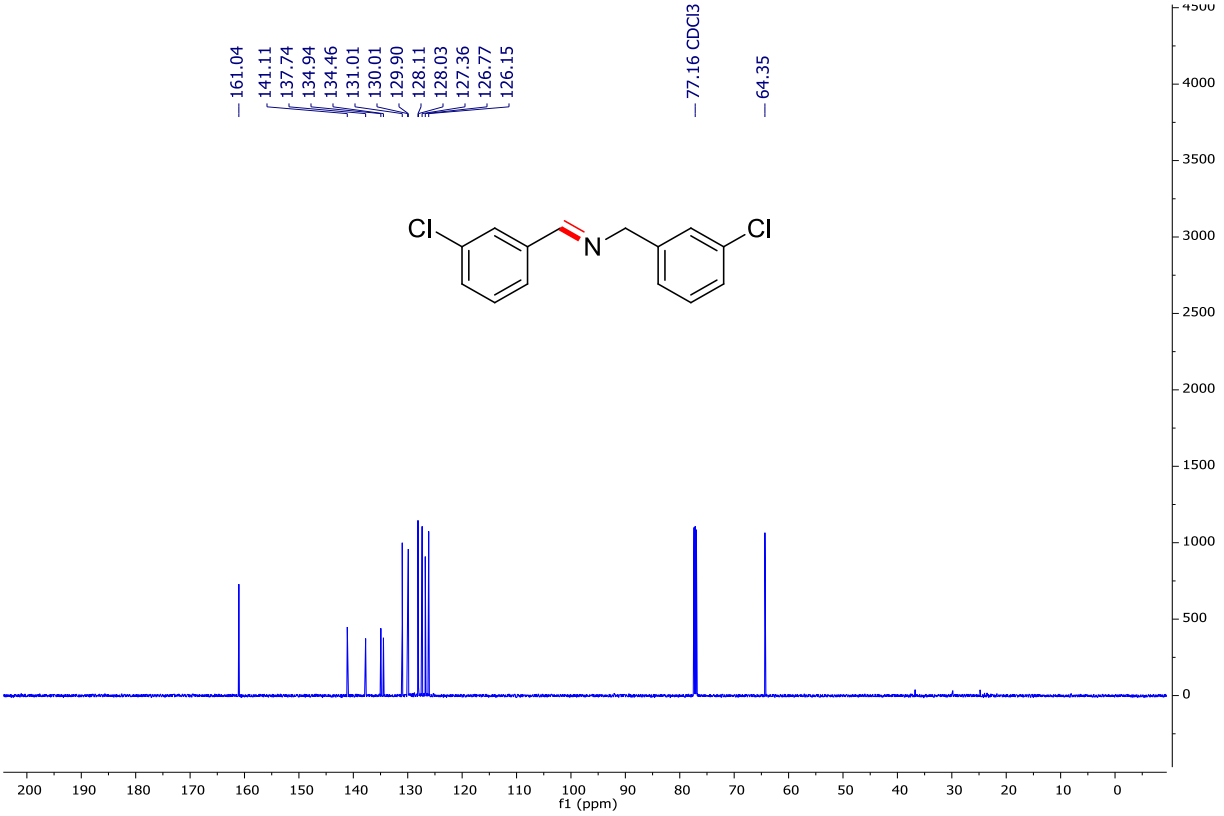


(E)-N-(3-chlorobenzylidene)-1-(3-chlorophenyl)methanamine (Table 2, entry **3g**)

^{1}H NMR (600 MHz)

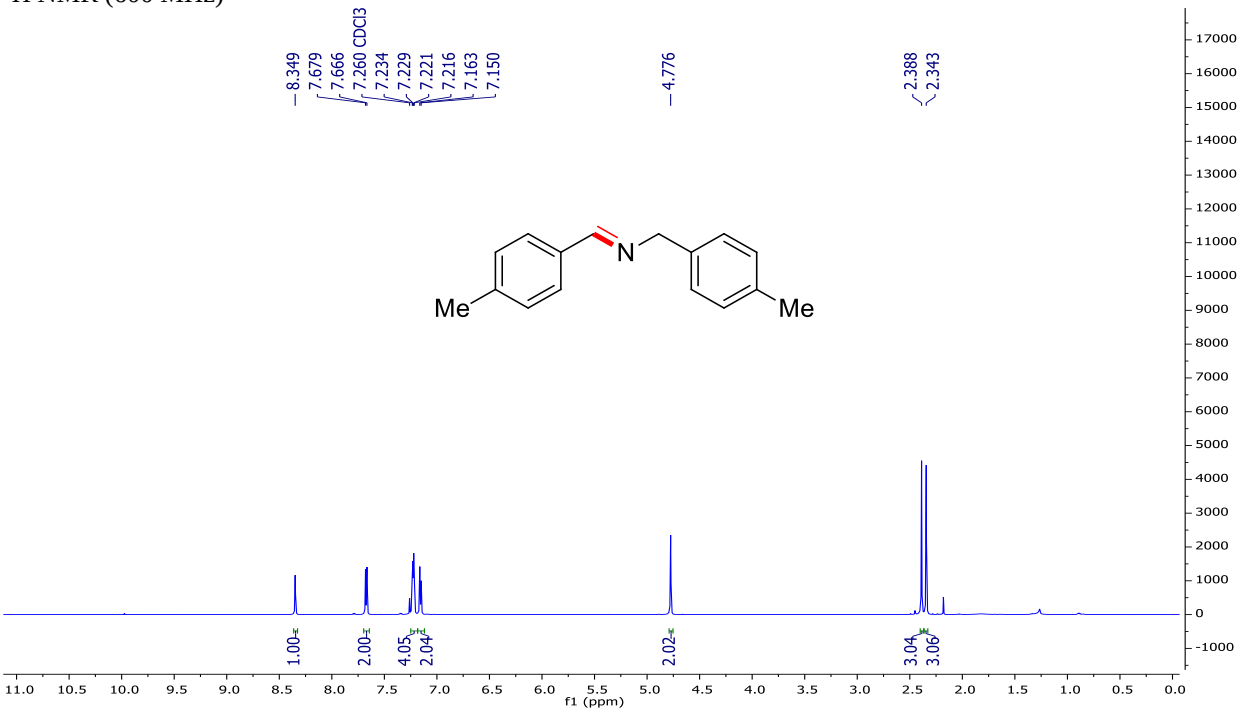


$^{13}C\{^{1}H\}$ NMR (150 MHz)

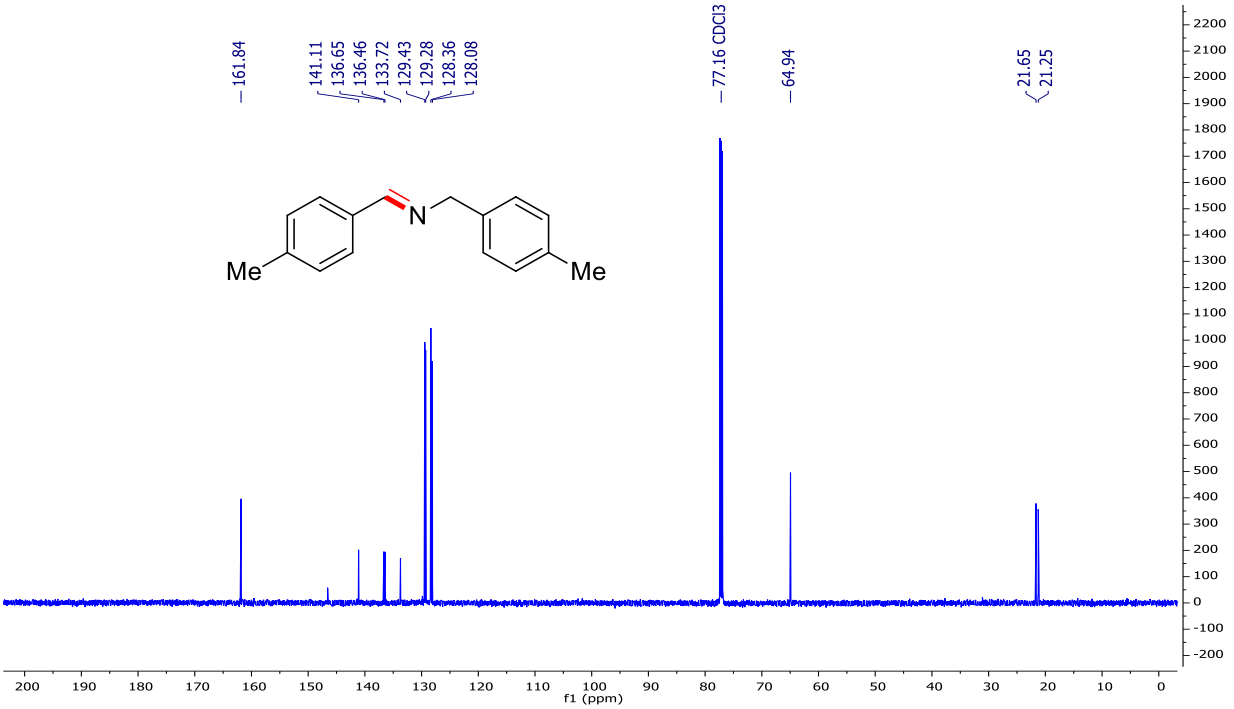


(E)-N-(4-methylbenzylidene)-1-(p-tolyl)methanamine (Table 2, entry **3h**)

^{1}H NMR (600 MHz)

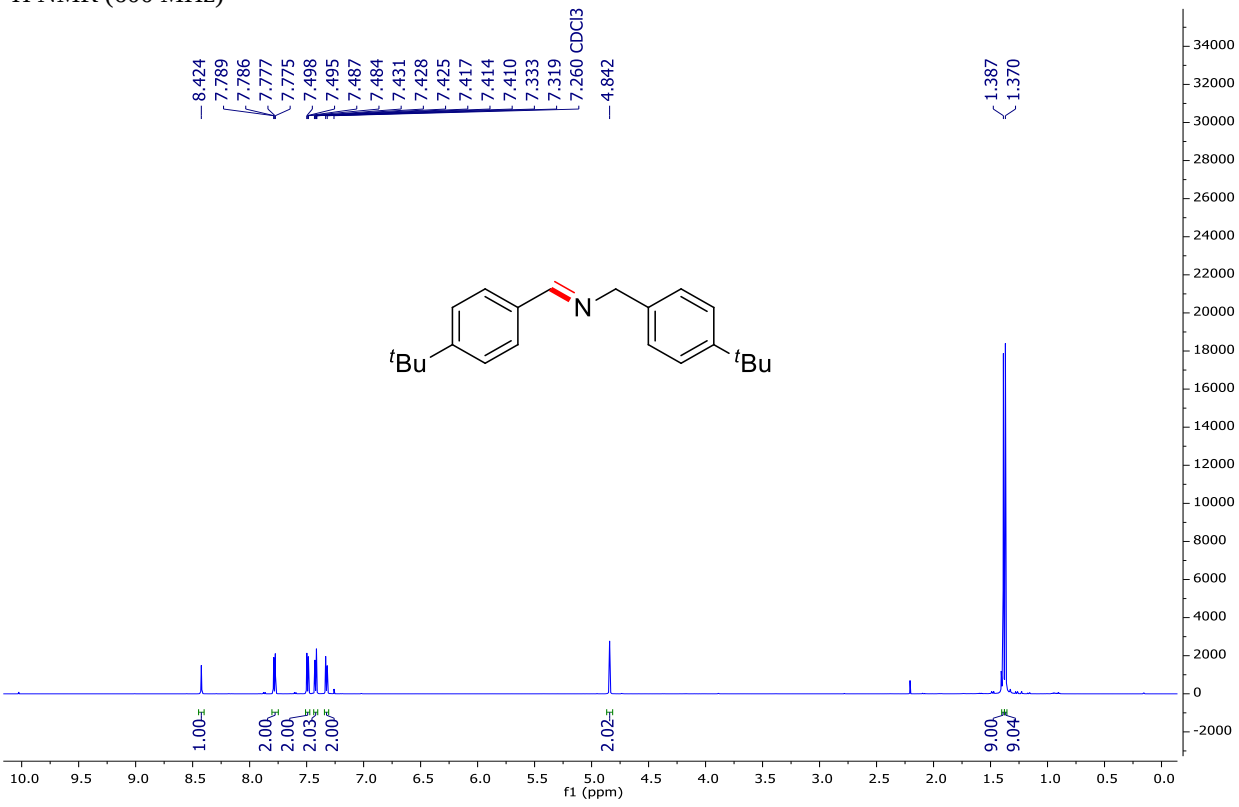


$^{13}C\{^{1}H\}$ NMR (150 MHz)

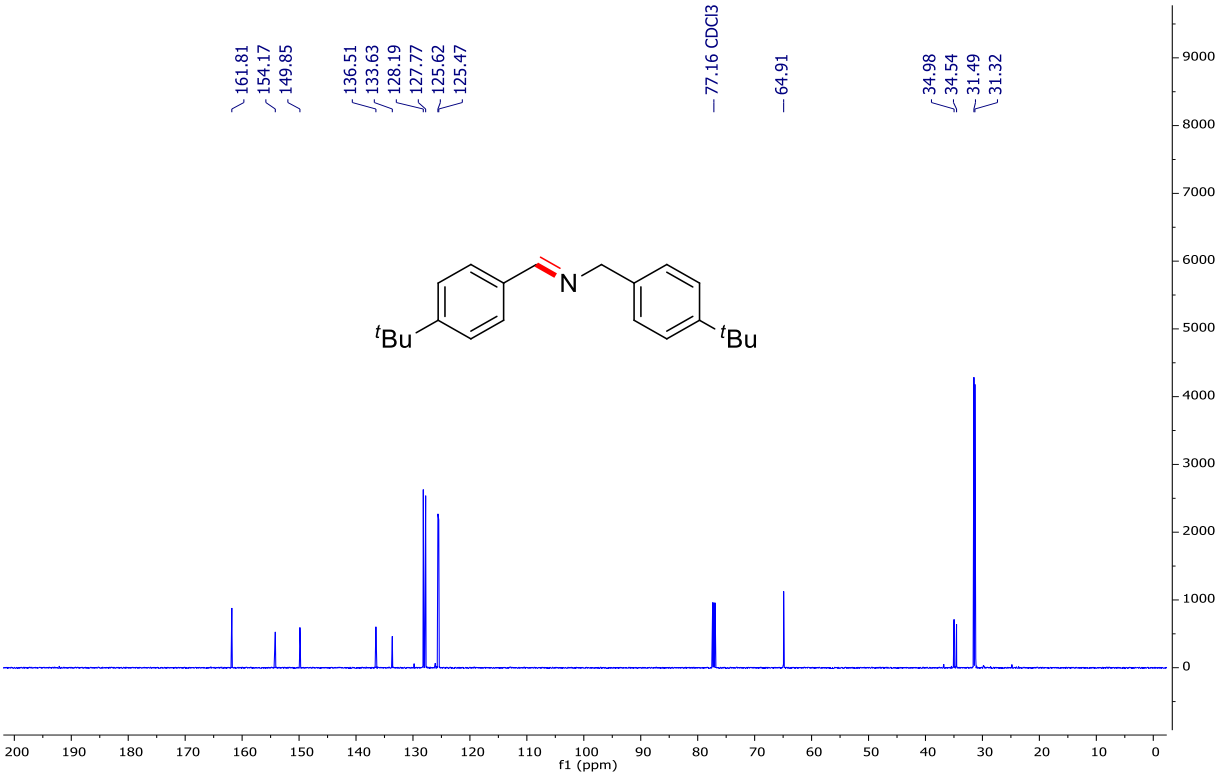


(E)-N-(4-(tert-butyl)benzylidene)-1-(4-(tert-butyl)phenyl)methanamine (Table 2, entry **3i**)

^{1}H NMR (600 MHz)

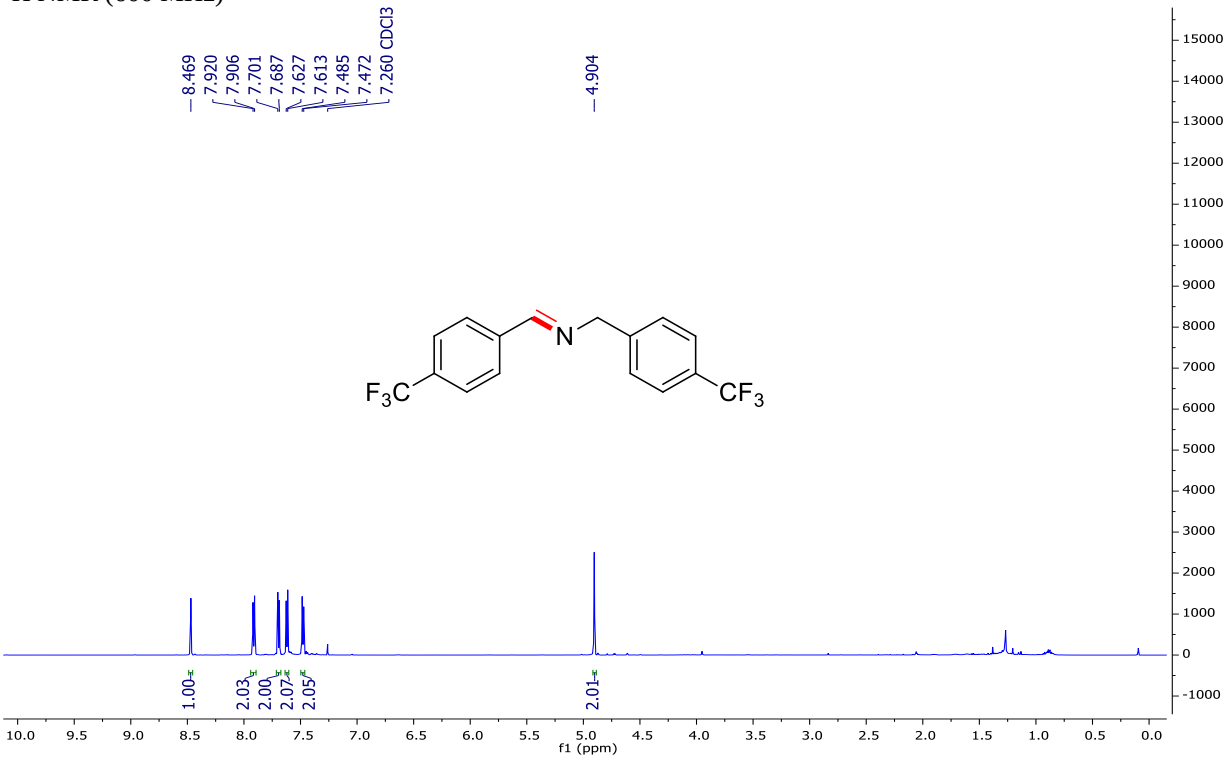


^{13}C{^{1}H} NMR (150 MHz)

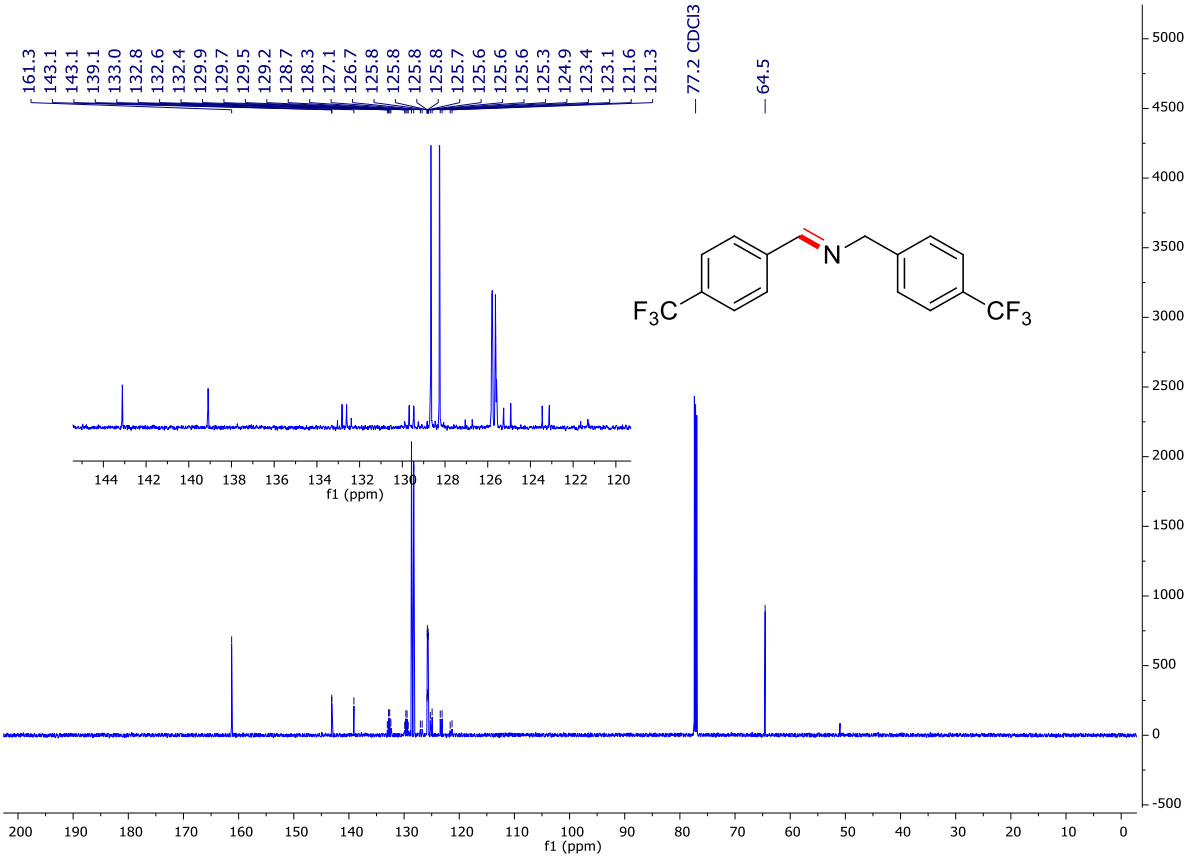


(E)-N-(4-(trifluoromethyl)benzylidene)-1-(4-(trifluoromethyl)phenyl)methanamine (Table 2, entry **3j**)

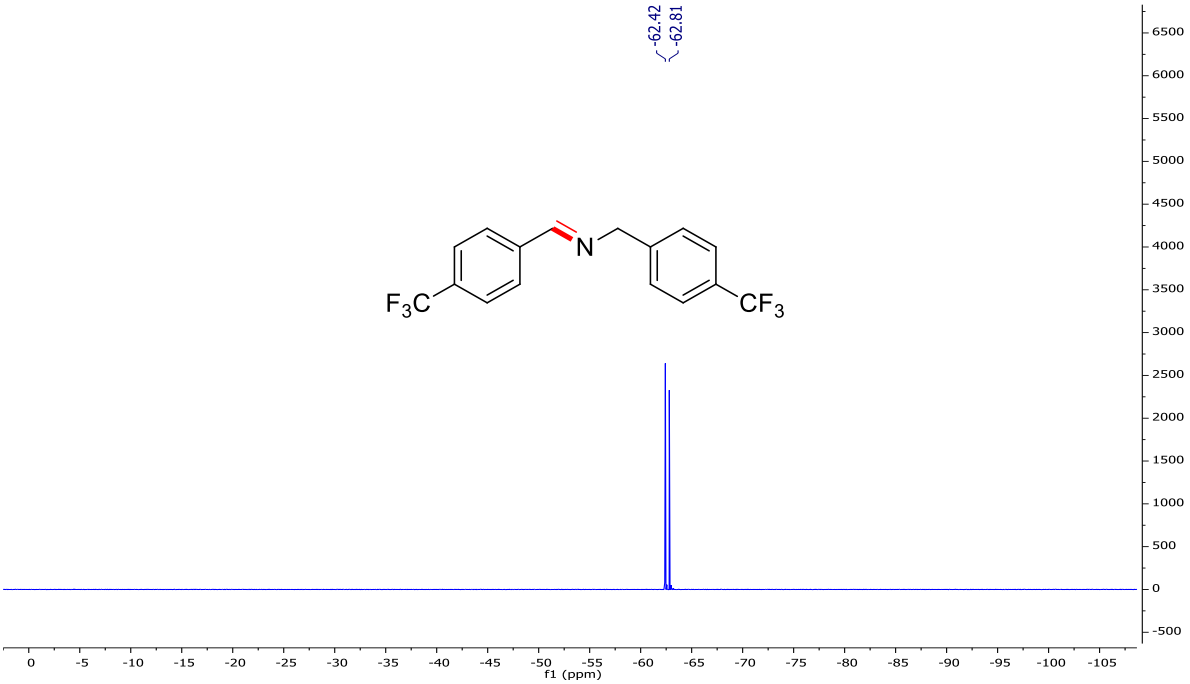
^{1}H NMR (600 MHz)



^{13}C{^{1}H} NMR (150 MHz)

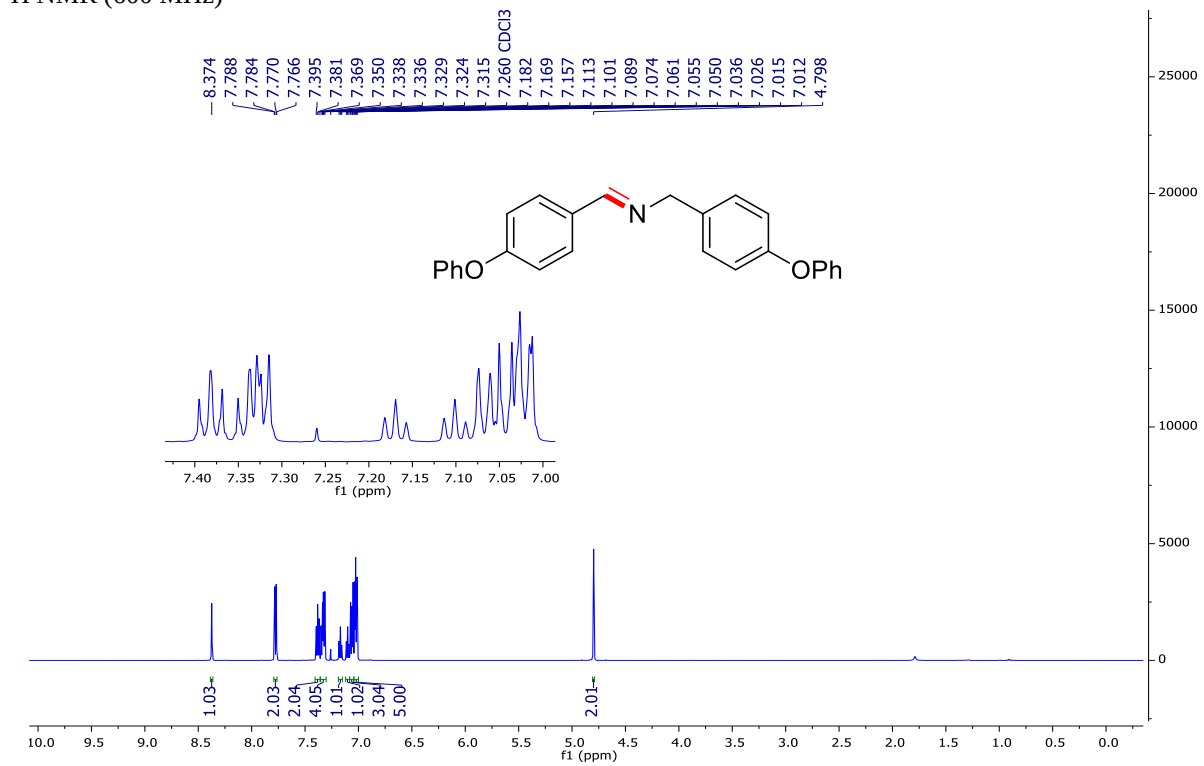


^{19}F NMR (564 MHz)

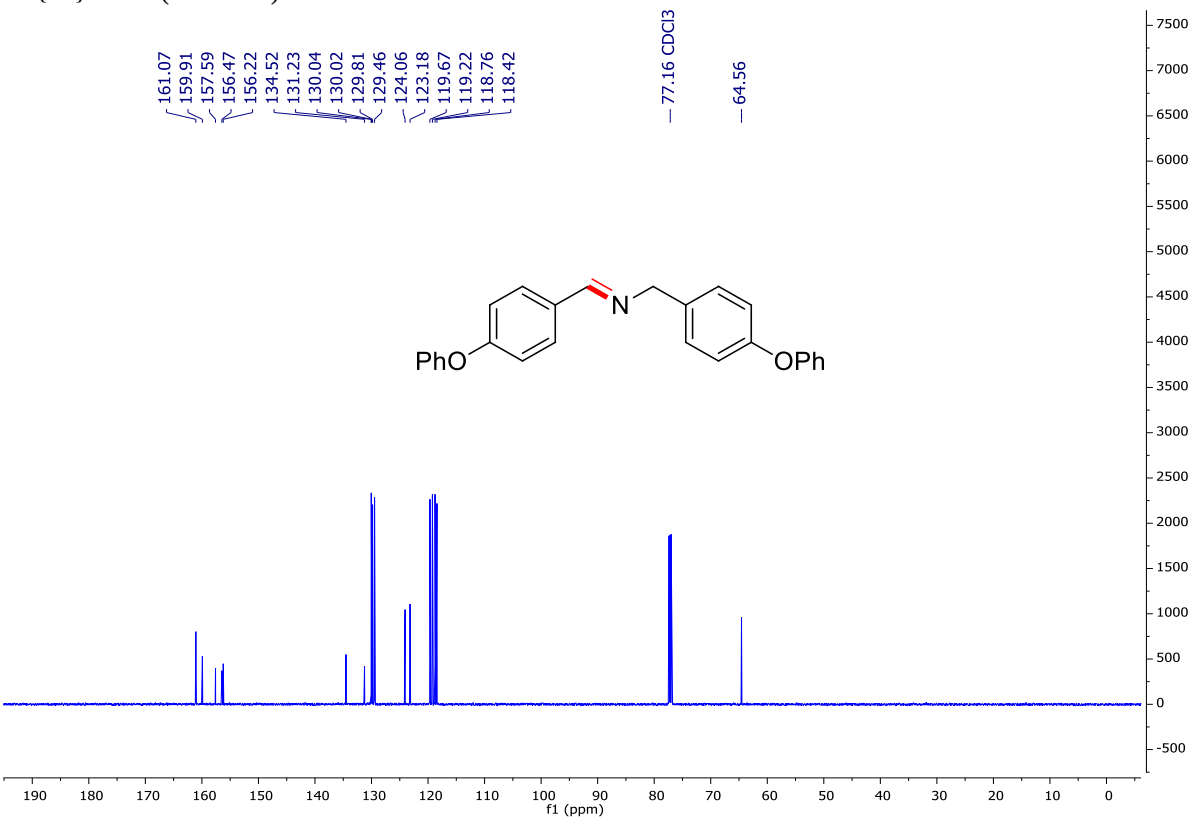


(E)-N-(4-phenoxybenzylidene)-1-(4-phenoxyphenyl)methanamine (Table 2, entry **3k**)

^{1}H NMR (600 MHz)

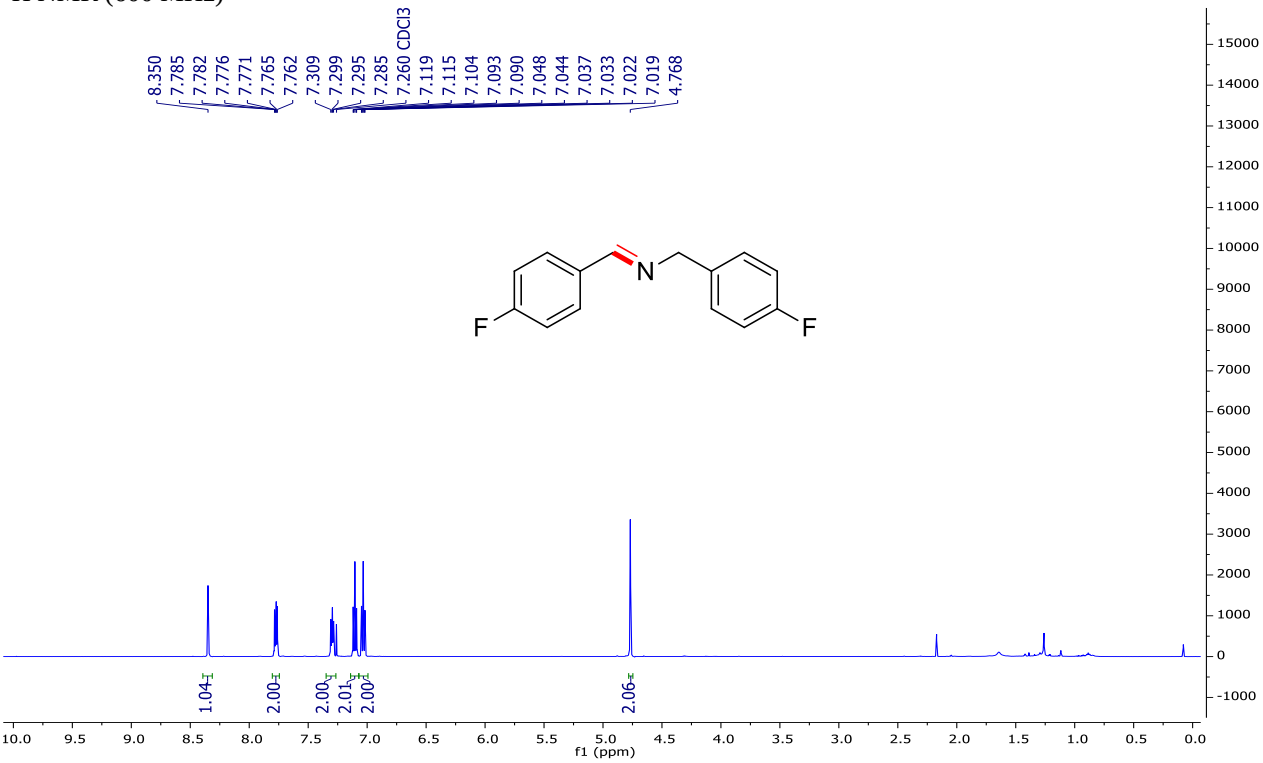


^{13}C{^{1}H} NMR (150 MHz)

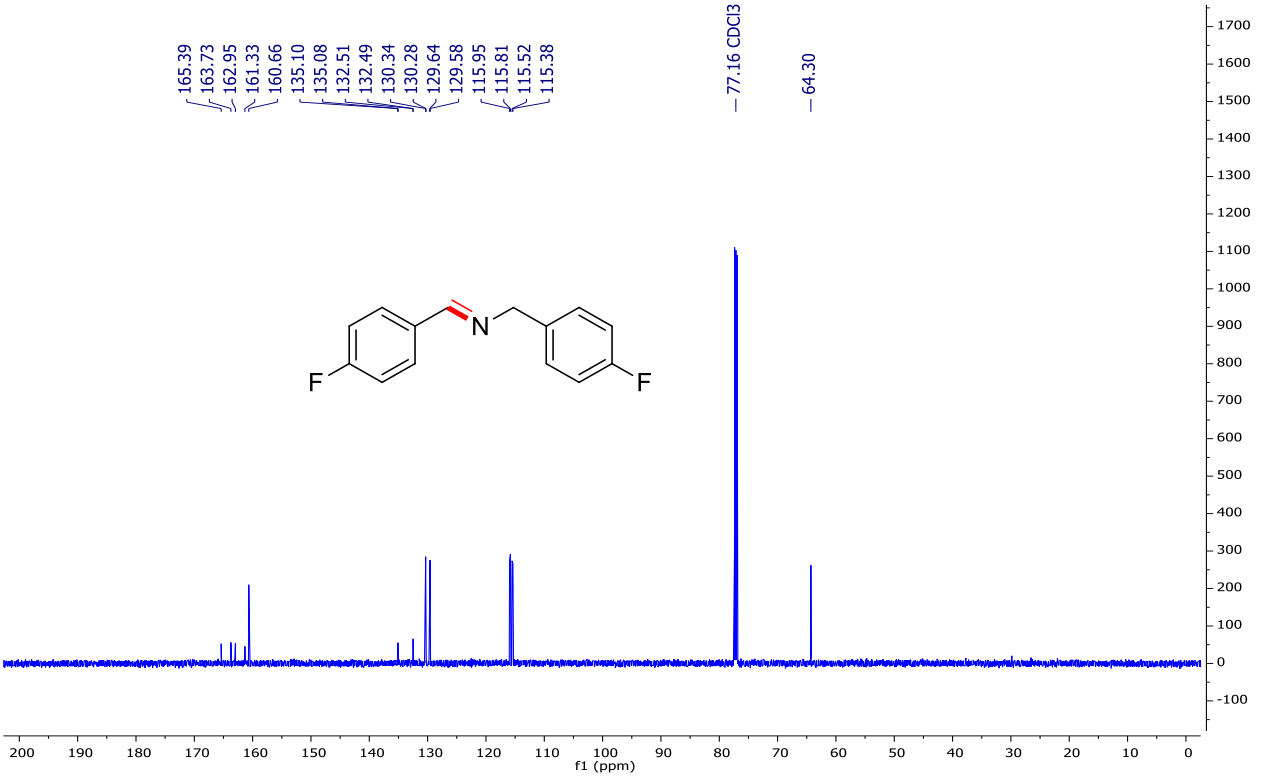


(E)-N-(4-fluorobenzylidene)-1-(4-fluorophenyl)methanamine (Table 2, entry **3l**)

^{1}H NMR (600 MHz)

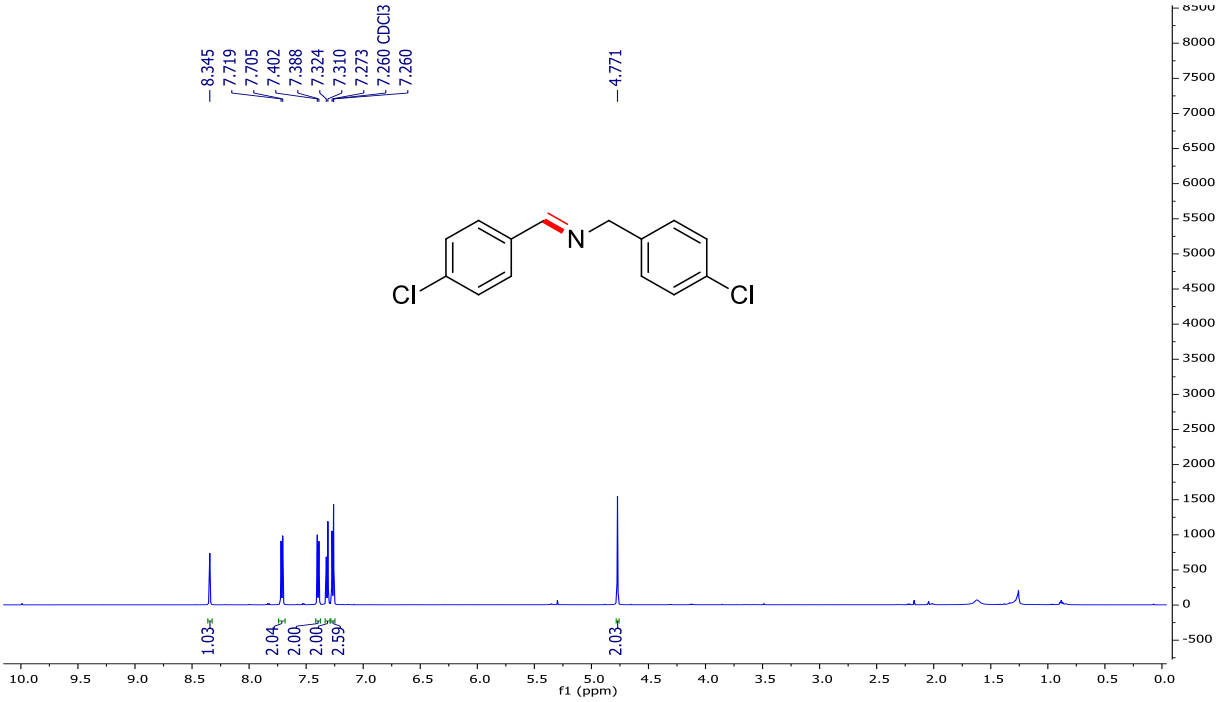


^{13}C{^{1}H} NMR (150 MHz)

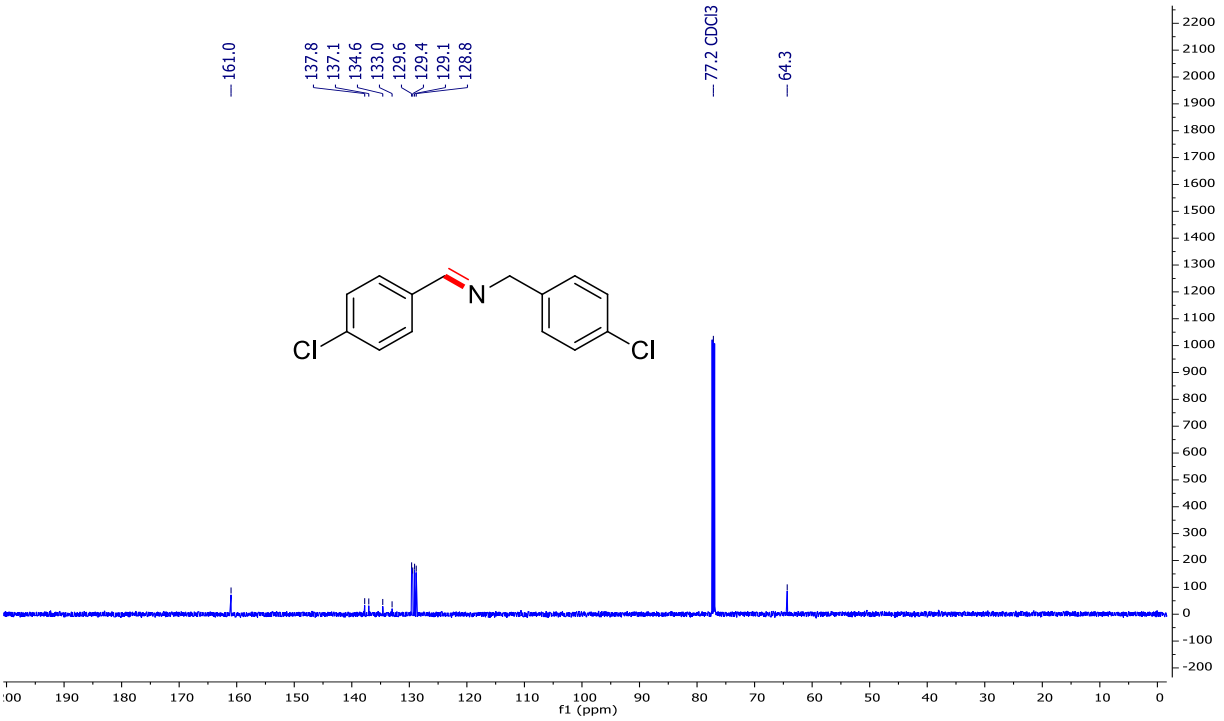


(E)-N-(4-chlorobenzylidene)-1-(4-chlorophenyl)methanamine (Table 2, entry **3m**)

^{1}H NMR (600 MHz)

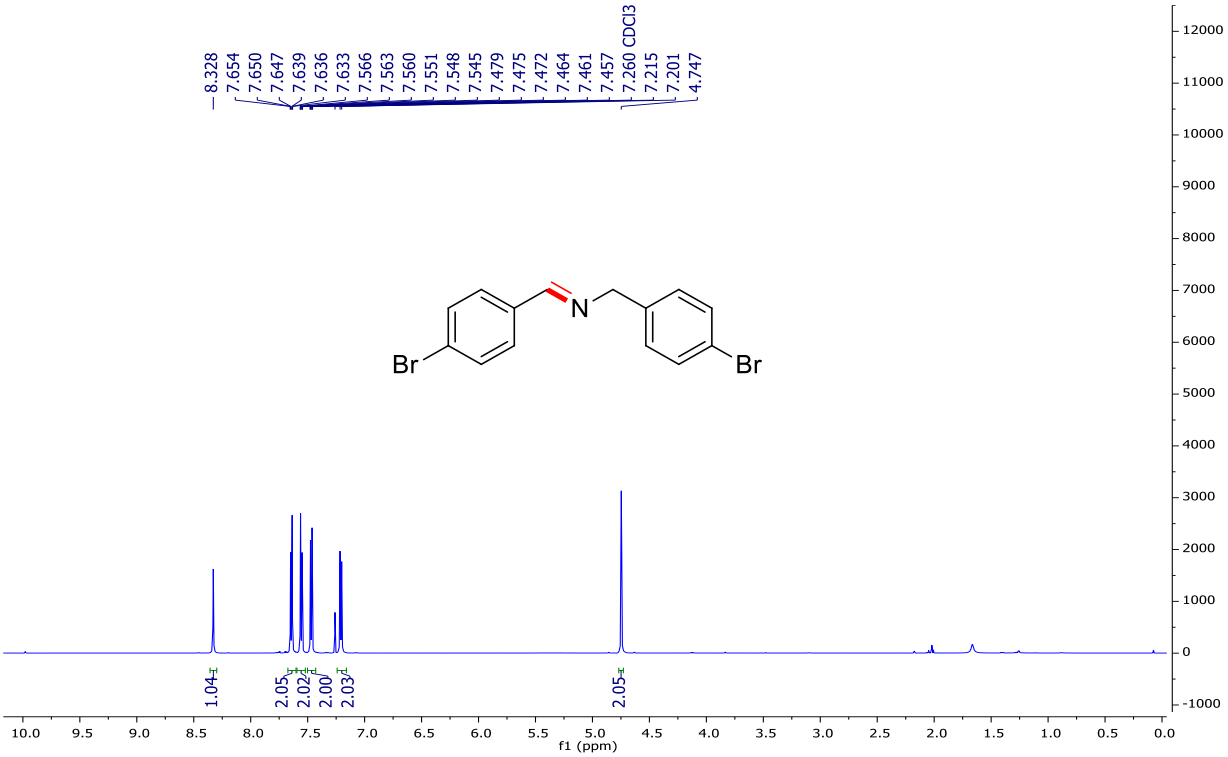


$^{13}C\{^{1}H\}$ NMR (150 MHz)

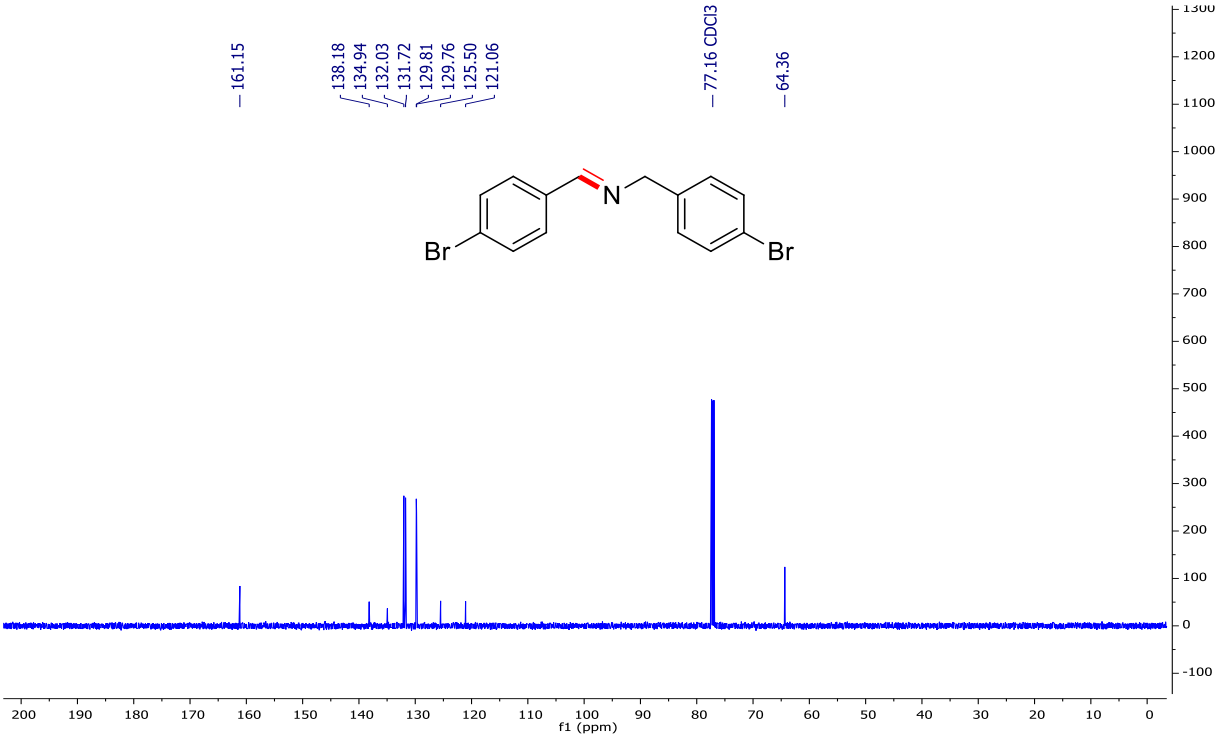


(E)-N-(4-bromobenzylidene)-1-(4-bromophenyl)methanamine (Table 2, entry **3n**)

^{1}H NMR (600 MHz)



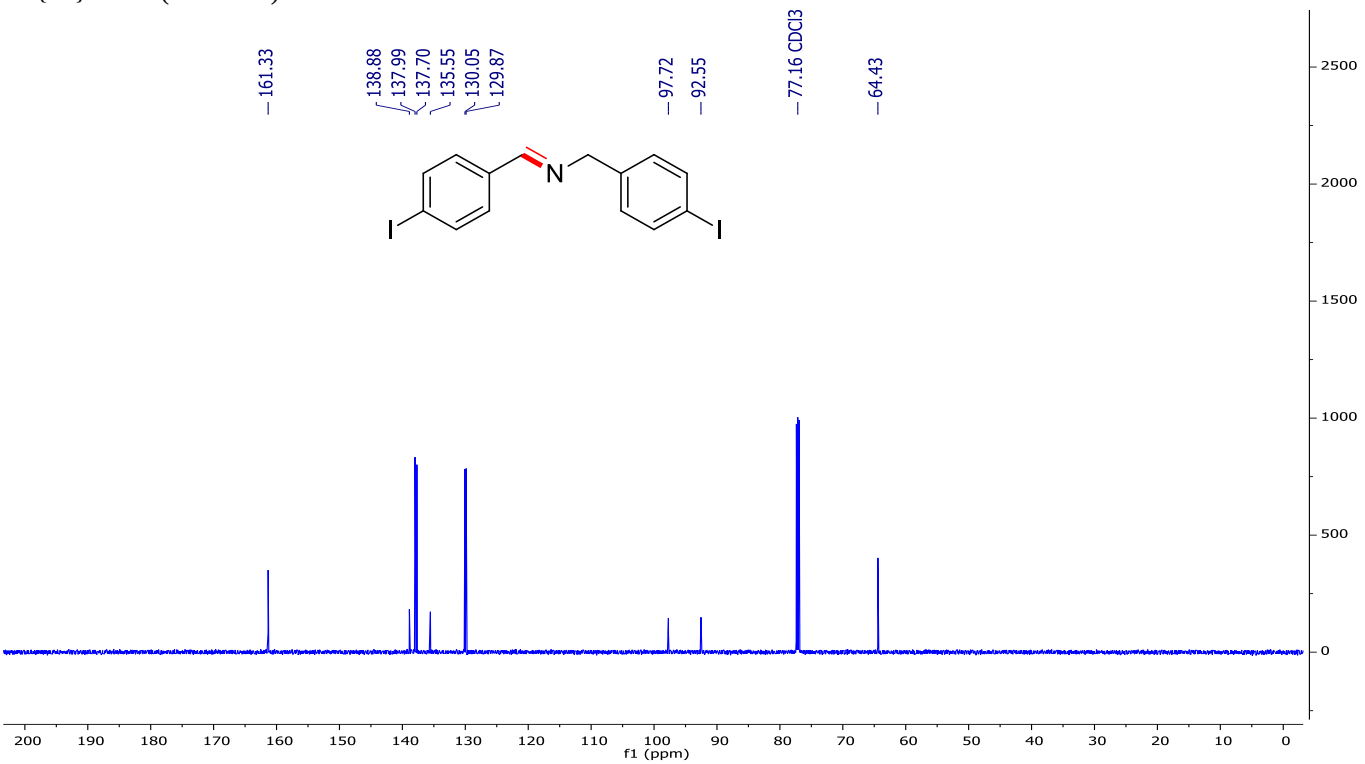
$^{13}C\{^{1}H\}$ NMR (150 MHz)



(E)-N-(4-iodobenzylidene)-1-(4-iodophenyl)methanamine (Table 2, entry **3o**)

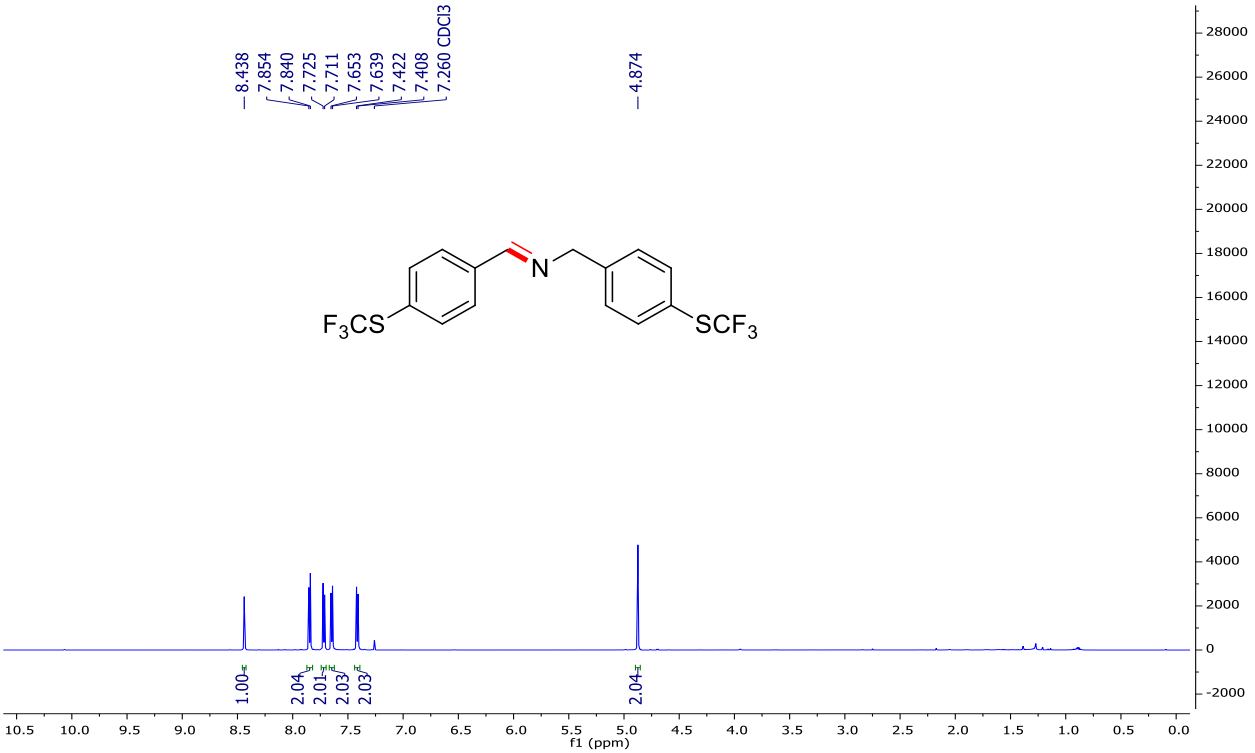
^{1}H NMR (600 MHz)

$^{13}C\{^{1}H\}$ NMR (150 MHz)

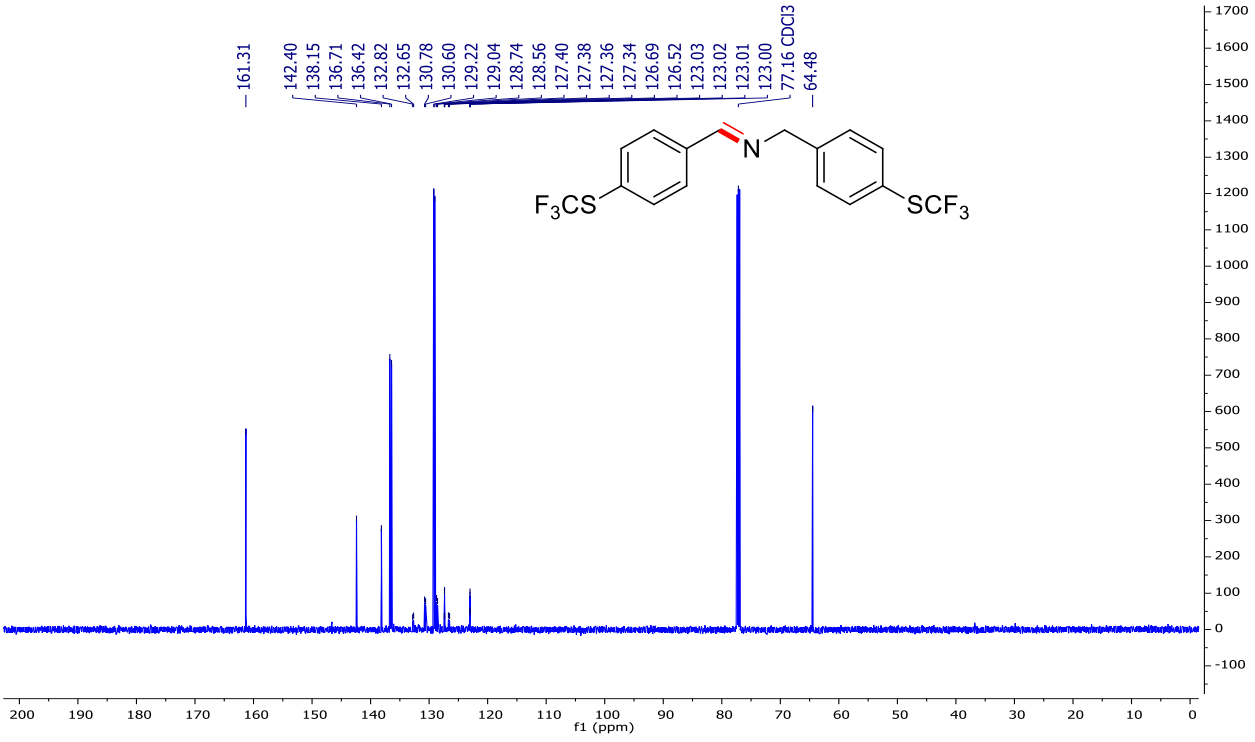


(E)-N-(4-((trifluoromethyl)thio)benzylidene)-1-(4-((trifluoromethyl)thio)phenyl)methanamine (Table 2, entry **3p**)

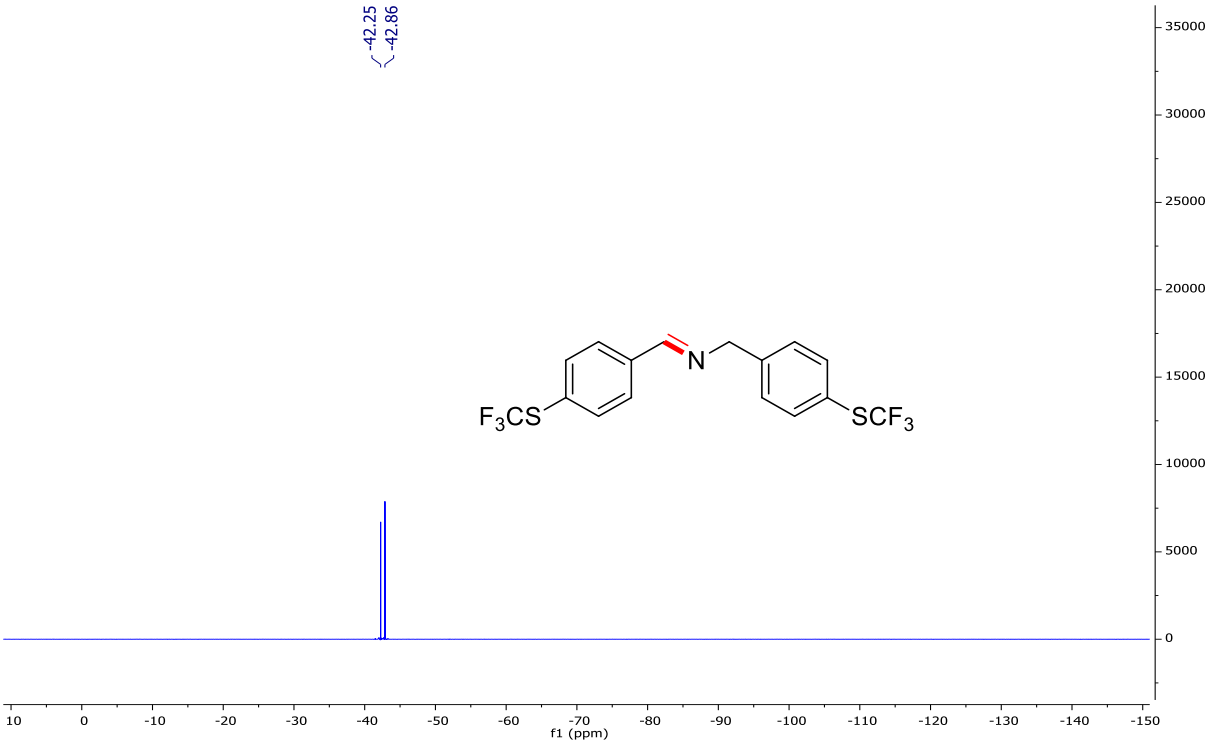
^{1}H NMR (600 MHz)



$^{13}C\{^{1}H\}$ NMR (150 MHz)

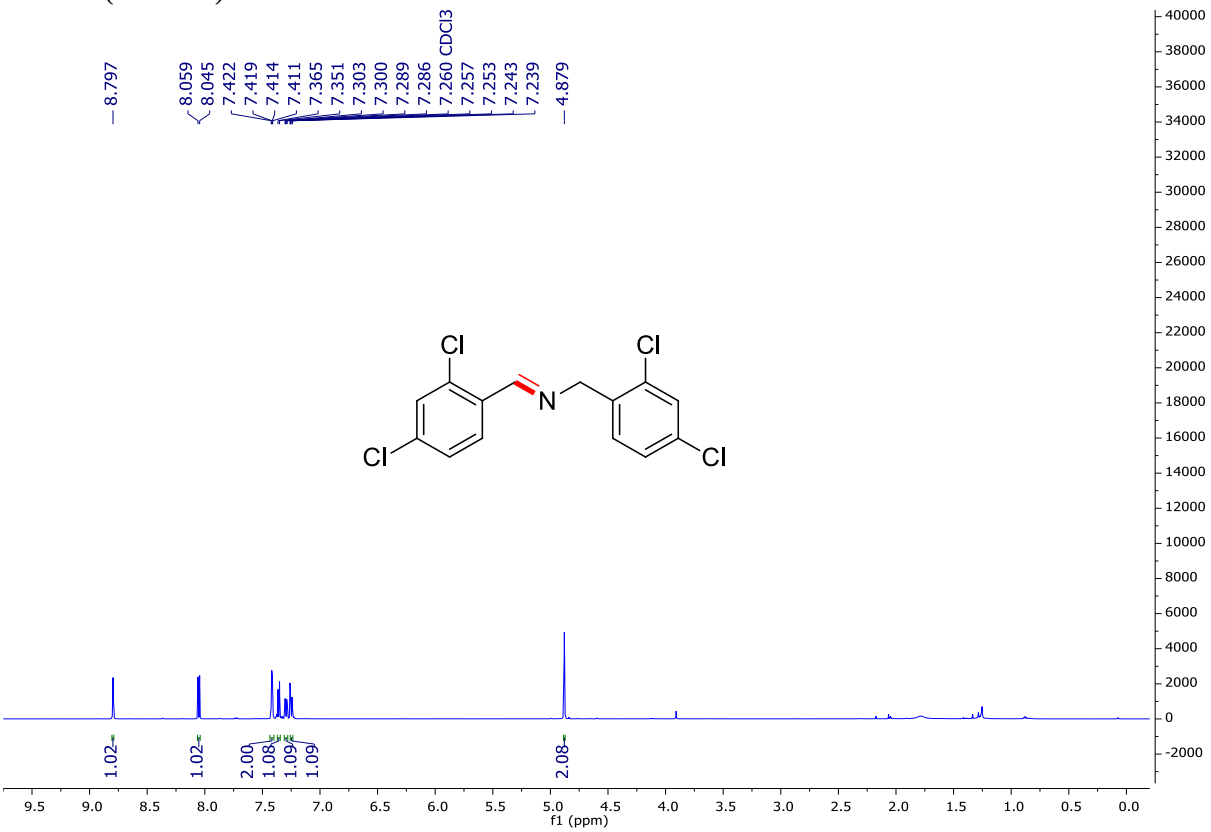


^{19}F NMR (564 MHz)

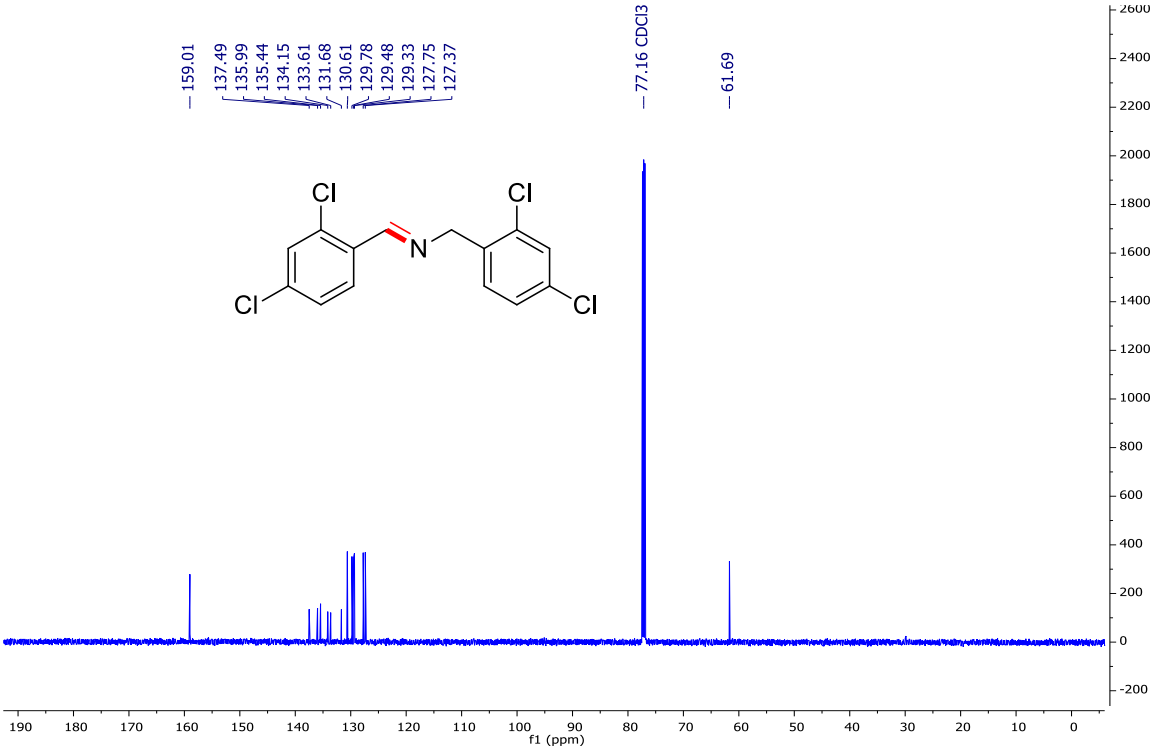


(E)-N-(2,4-dichlorobenzylidene)-1-(2,4-dichlorophenyl)methanamine (Table 2, entry **3q**)

^{1}H NMR (600 MHz)

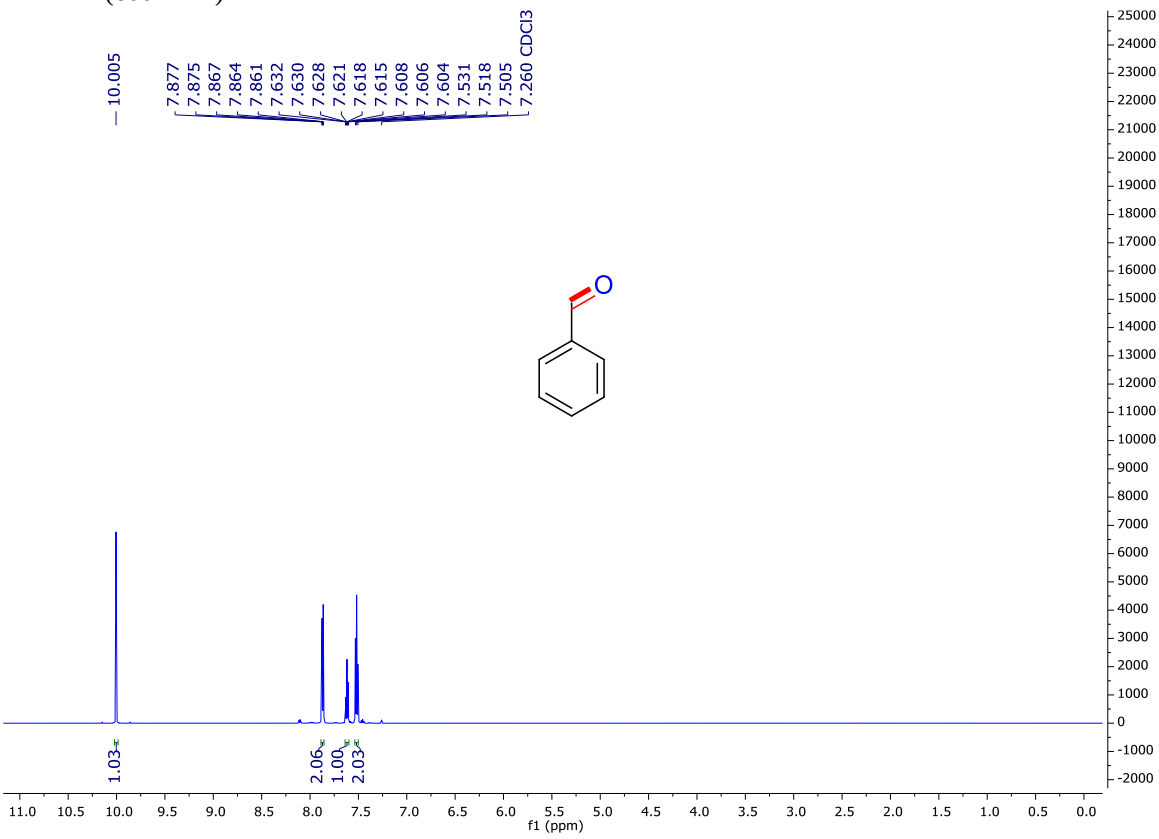


^{13}C{^{1}H} NMR (150 MHz)

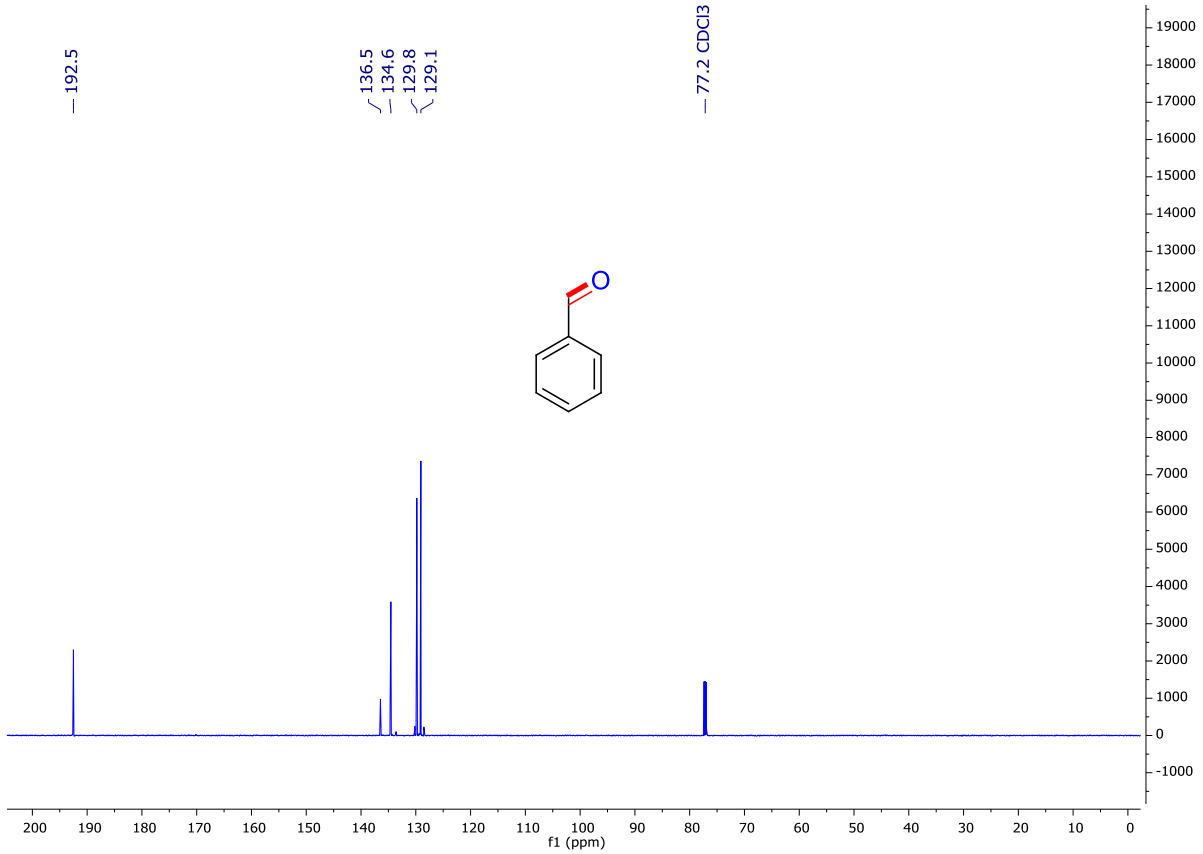


Benzaldehyde (Table 3, entry **4a**)

^{1}H NMR (600 MHz)

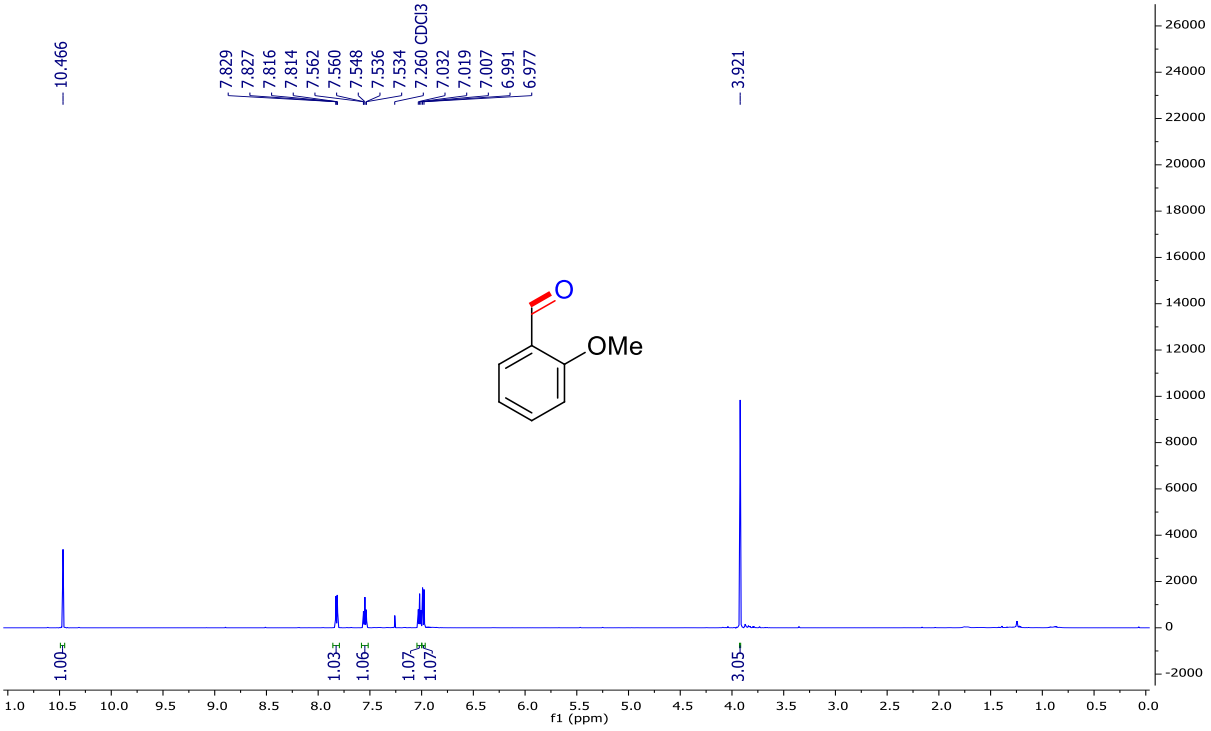


^{13}C{^{1}H} NMR (150 MHz)

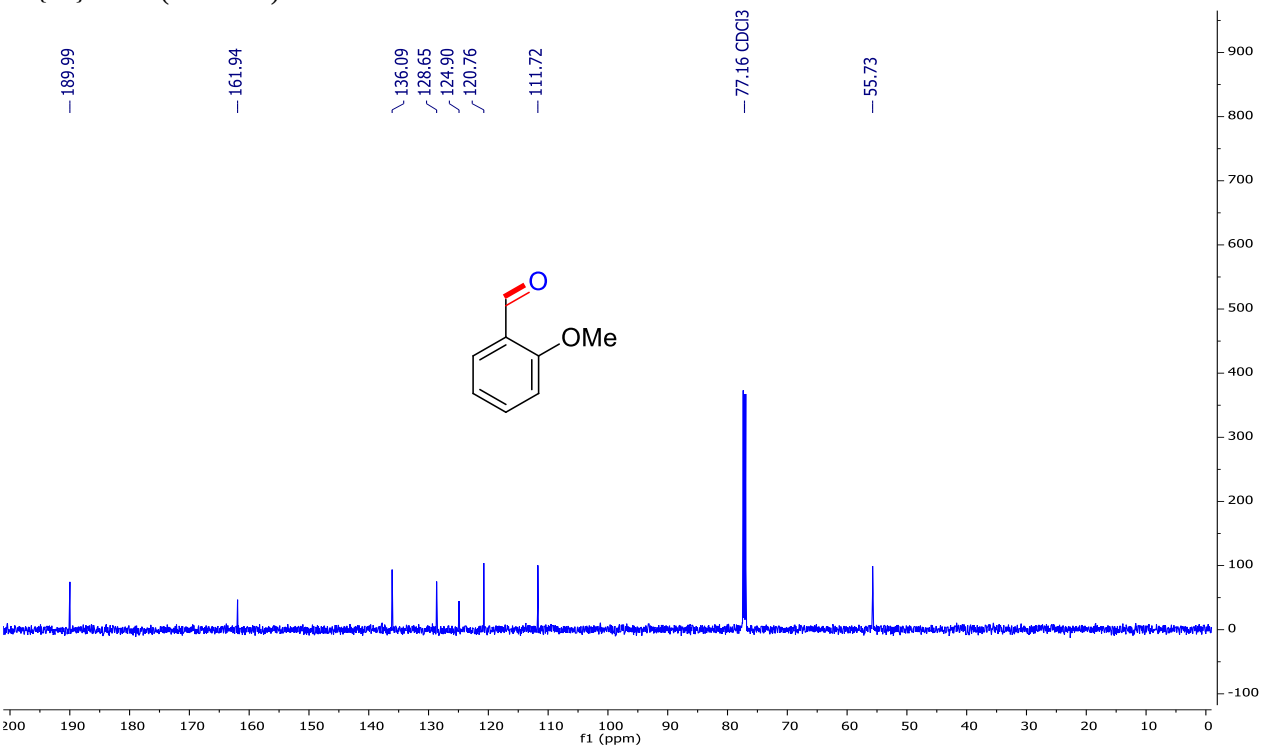


2-methoxybenzaldehyde (Table 3, entry **4b**)

^{1}H NMR (600 MHz)

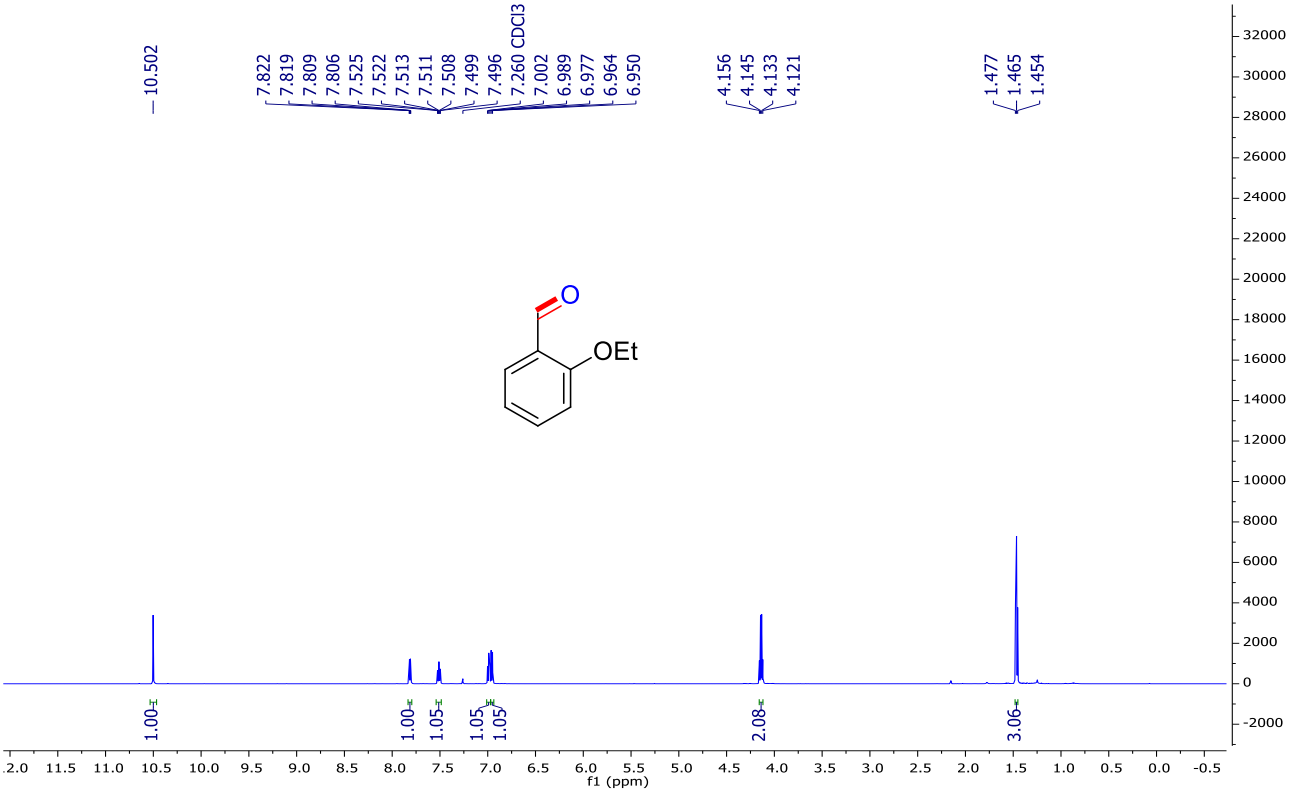


$^{13}C\{^{1}H\}$ NMR (150 MHz)

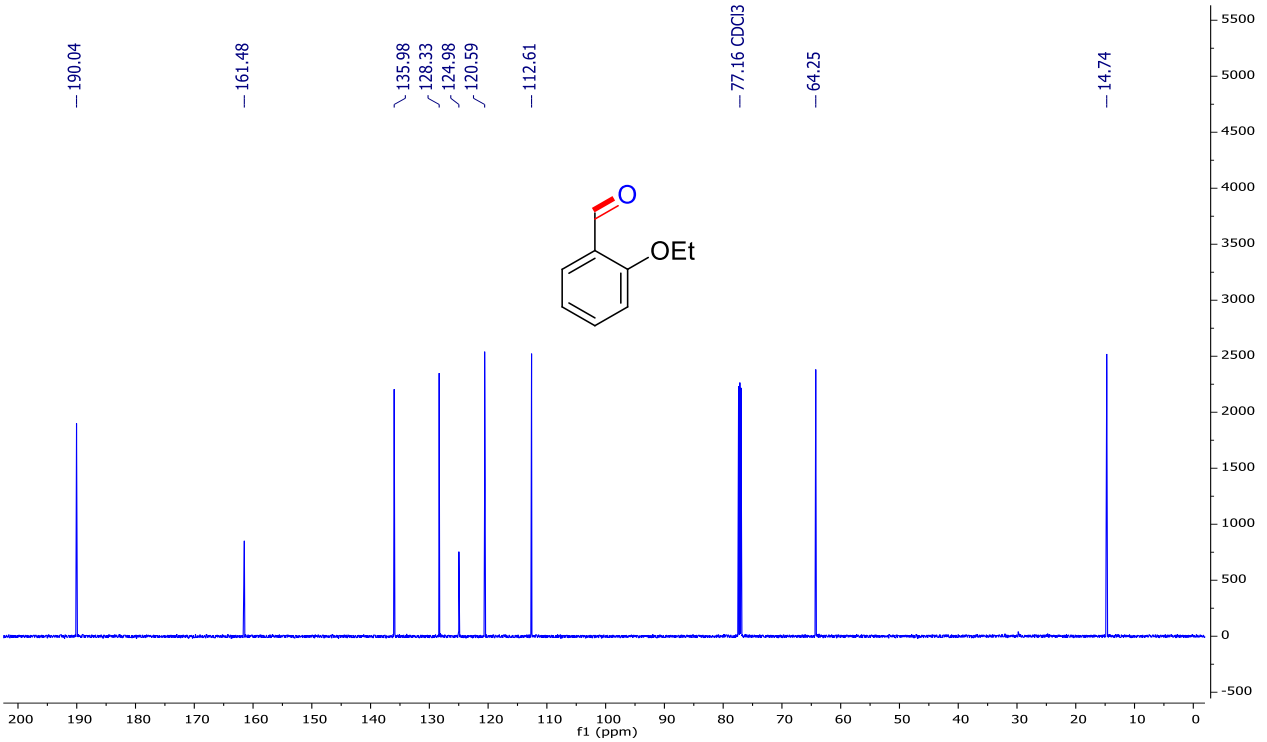


2-ethoxybenzaldehyde (Table 3, entry **4c**)

^{1}H NMR (600 MHz)

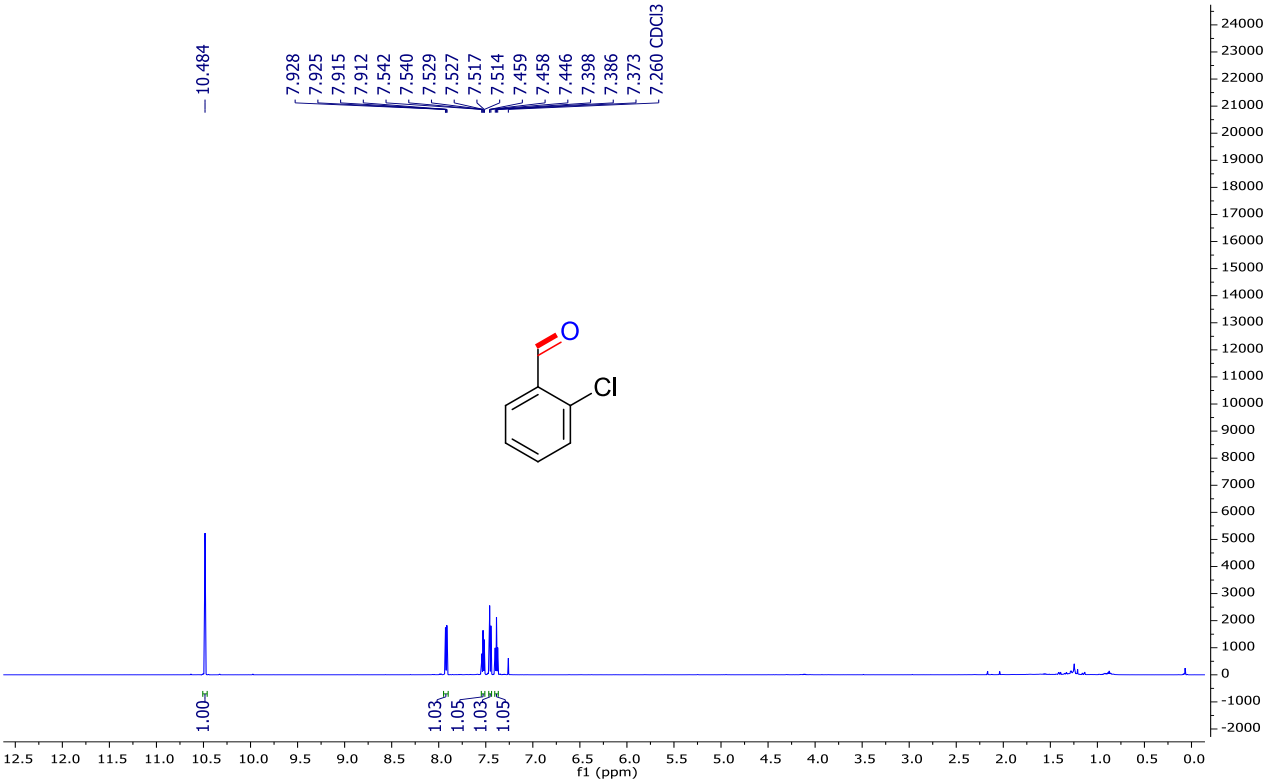


$^{13}C\{^{1}H\}$ NMR (150 MHz)

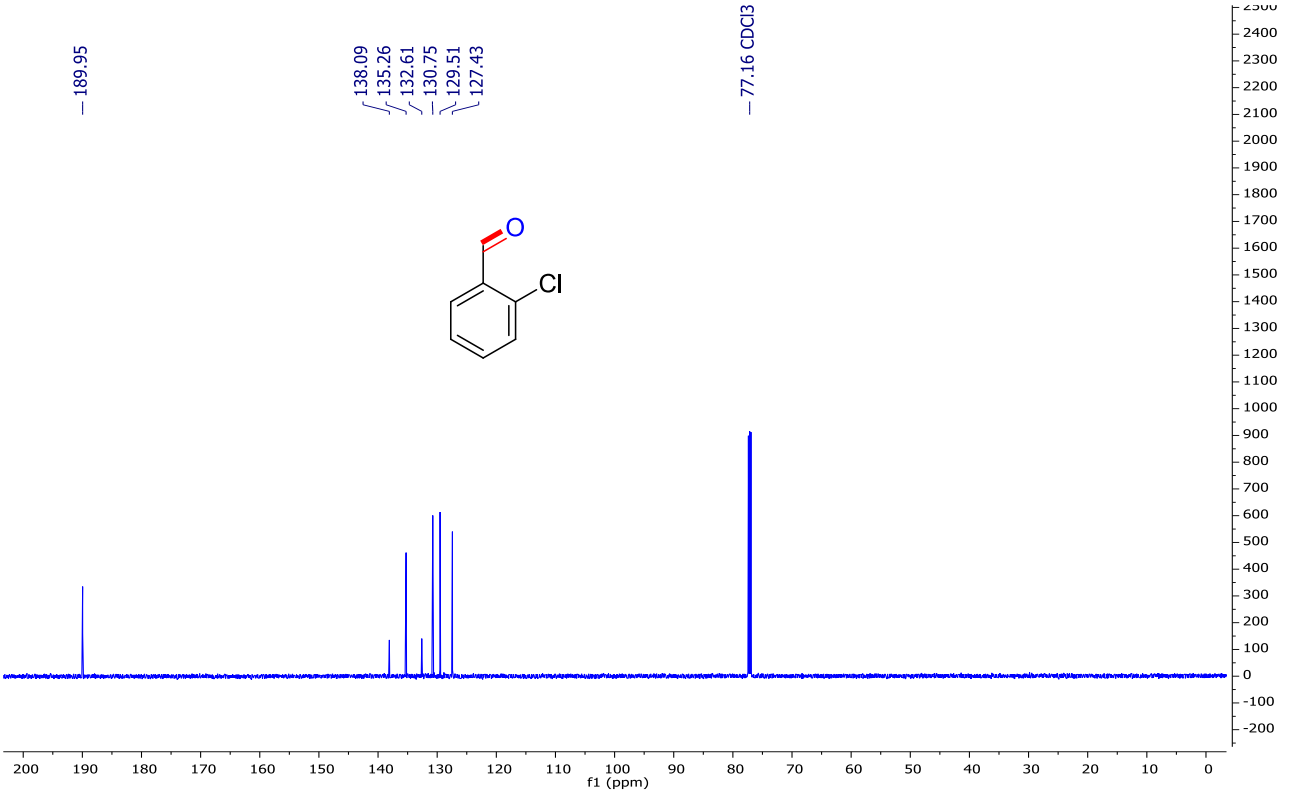


2-chlorobenzaldehyde (Table 3, entry **4d**)

^{1}H NMR (600 MHz)

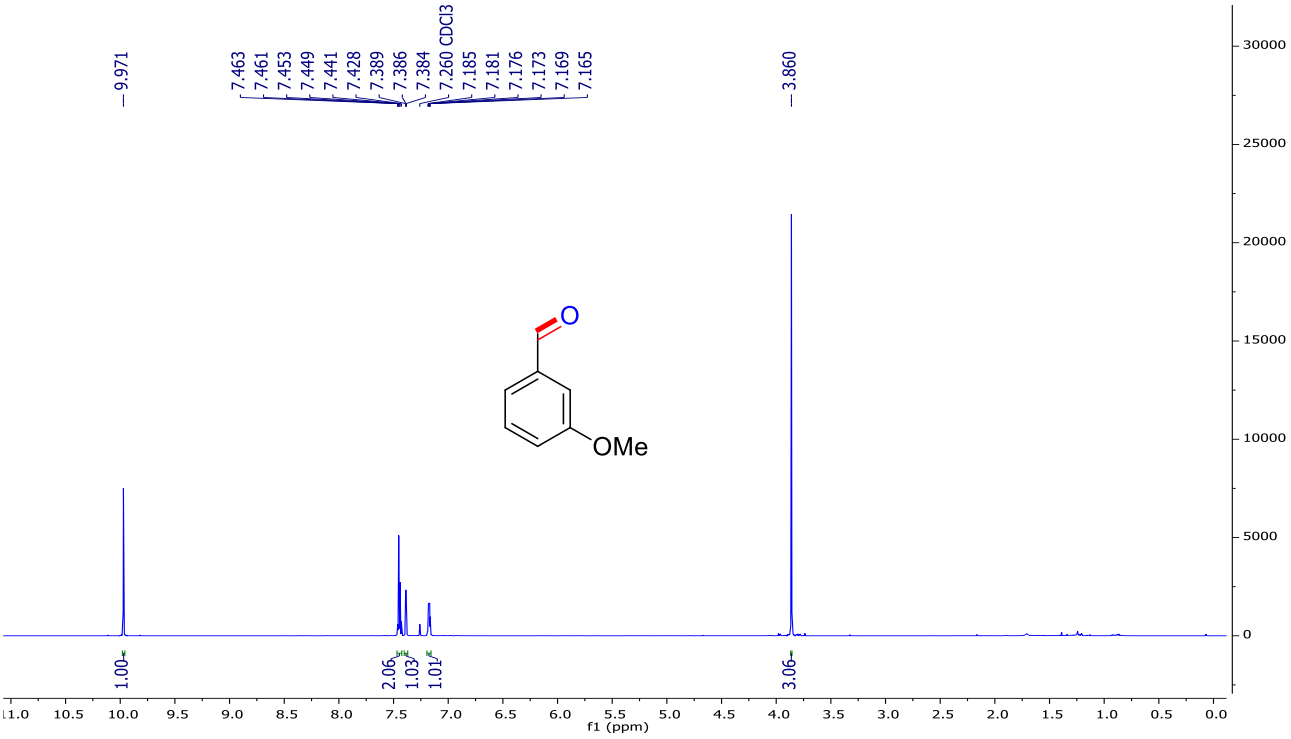


^{13}C{^{1}H} NMR (150 MHz)

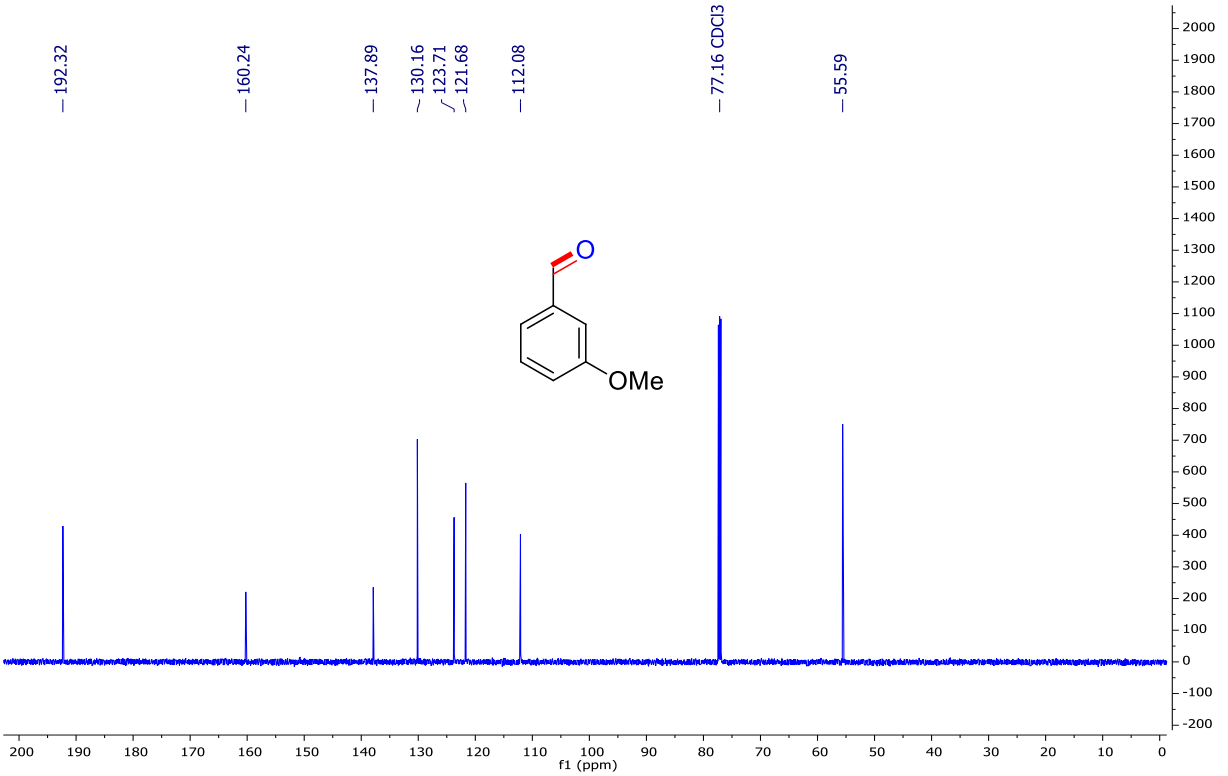


3-methoxybenzaldehyde (Table 3, entry **4e**)

^{1}H NMR (600 MHz)

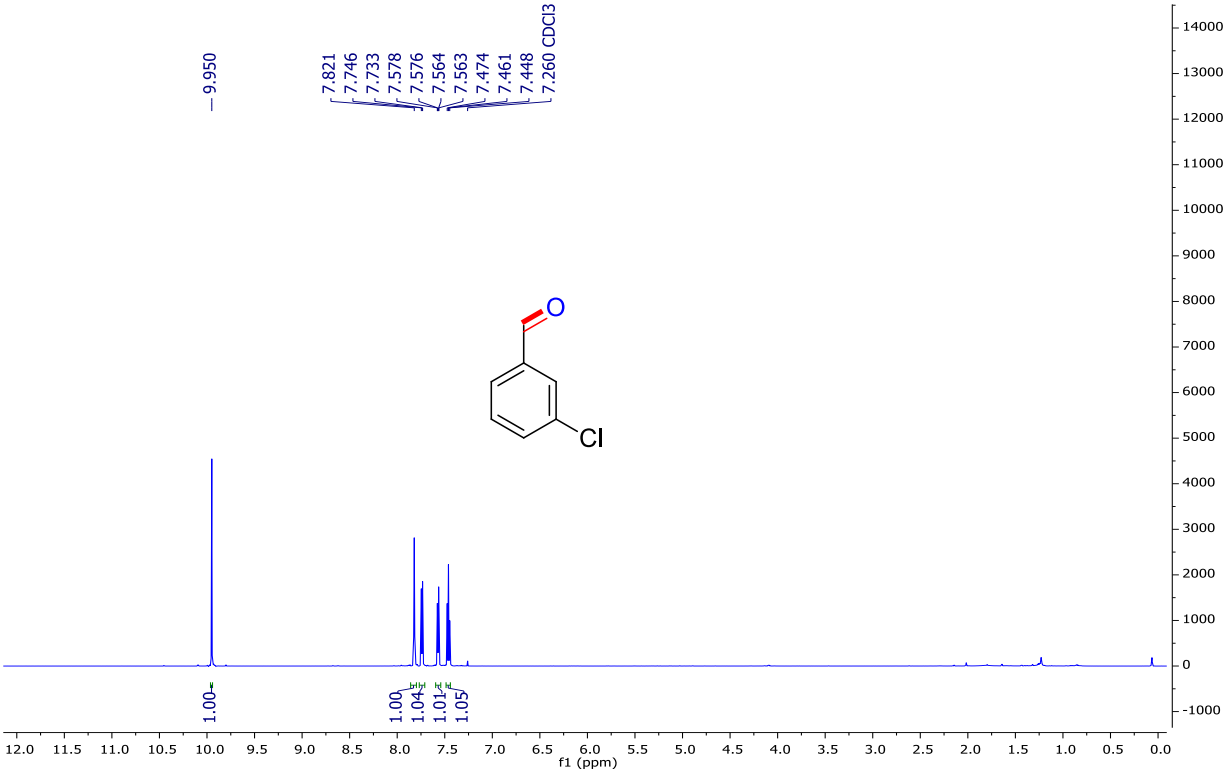


^{13}C{^{1}H} NMR (150 MHz)

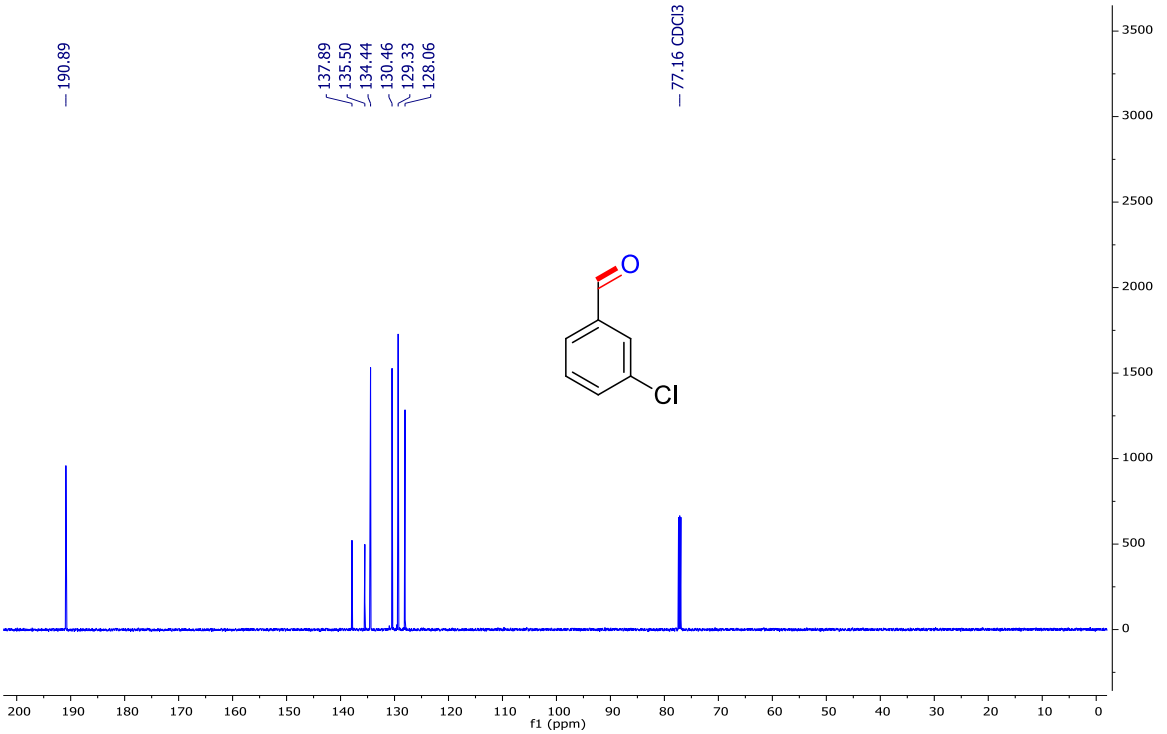


3-chlorobenzaldehyde (Table 3, entry **4f**)

^{1}H NMR (600 MHz)

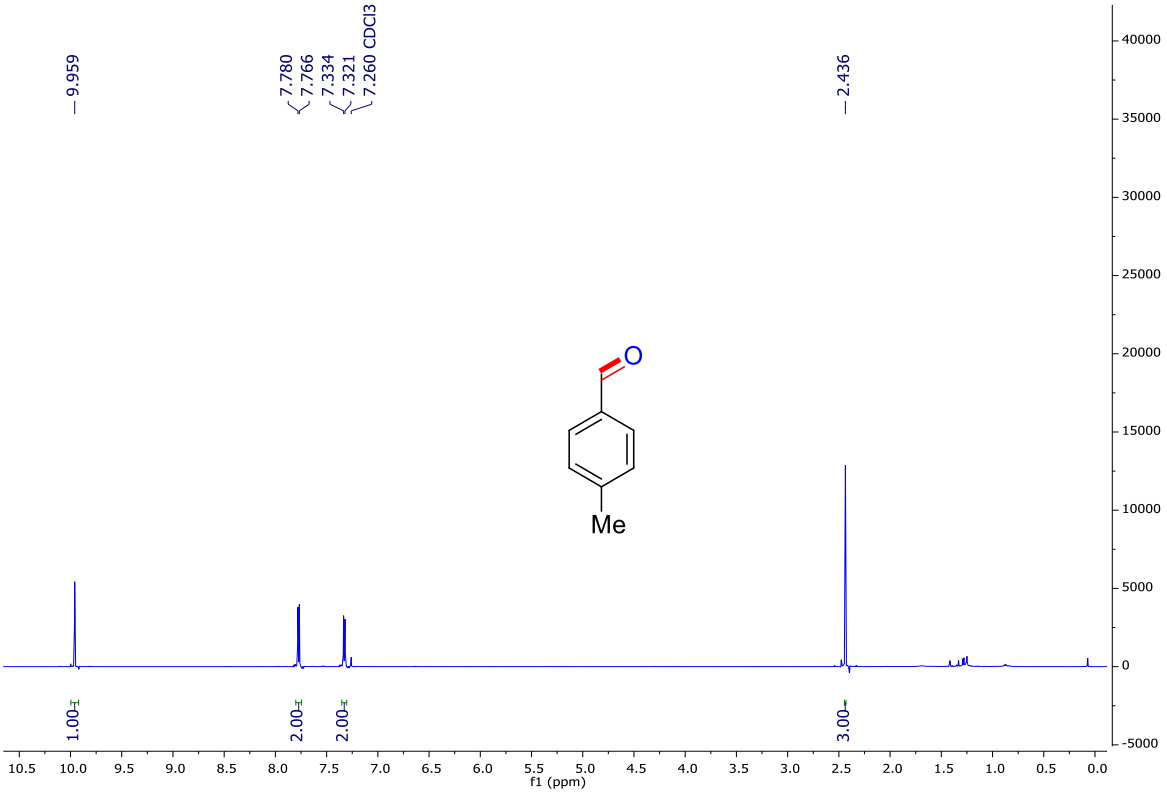


^{13}C{^{1}H} NMR (150 MHz)

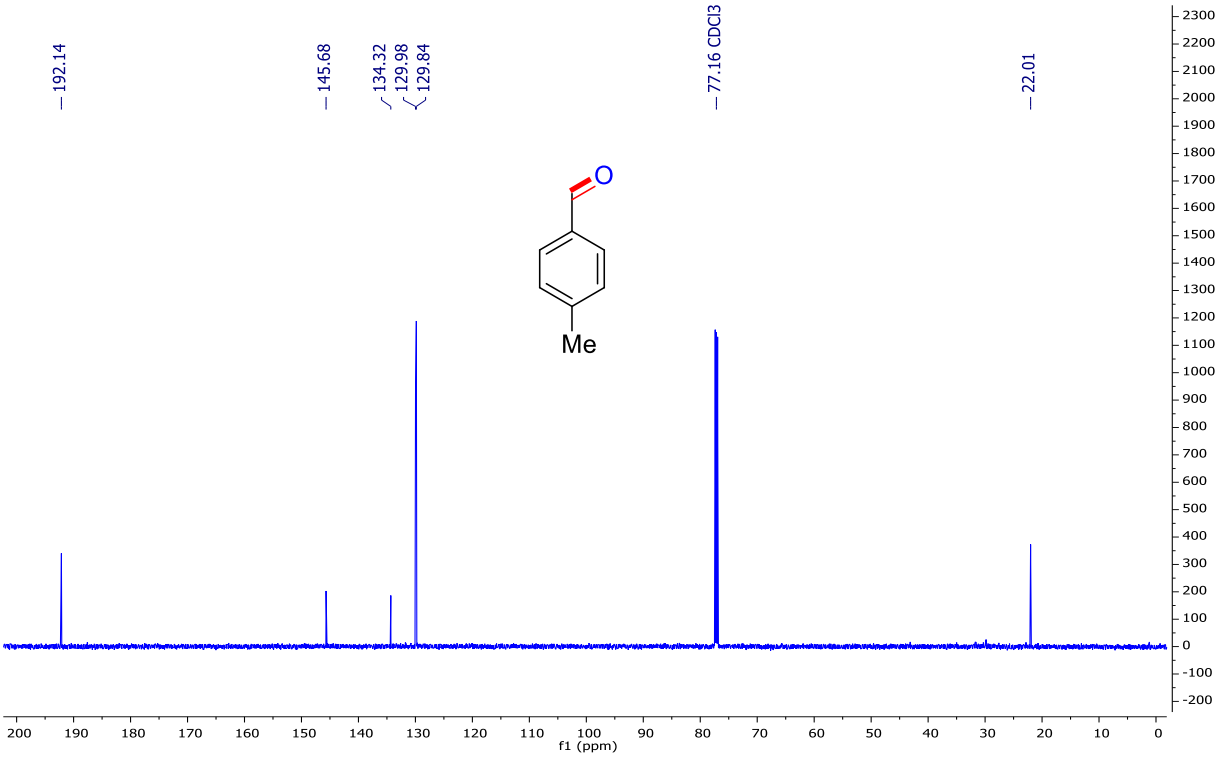


4-methylbenzaldehyde (Table 3, entry **4g**)

^{1}H NMR (600 MHz)

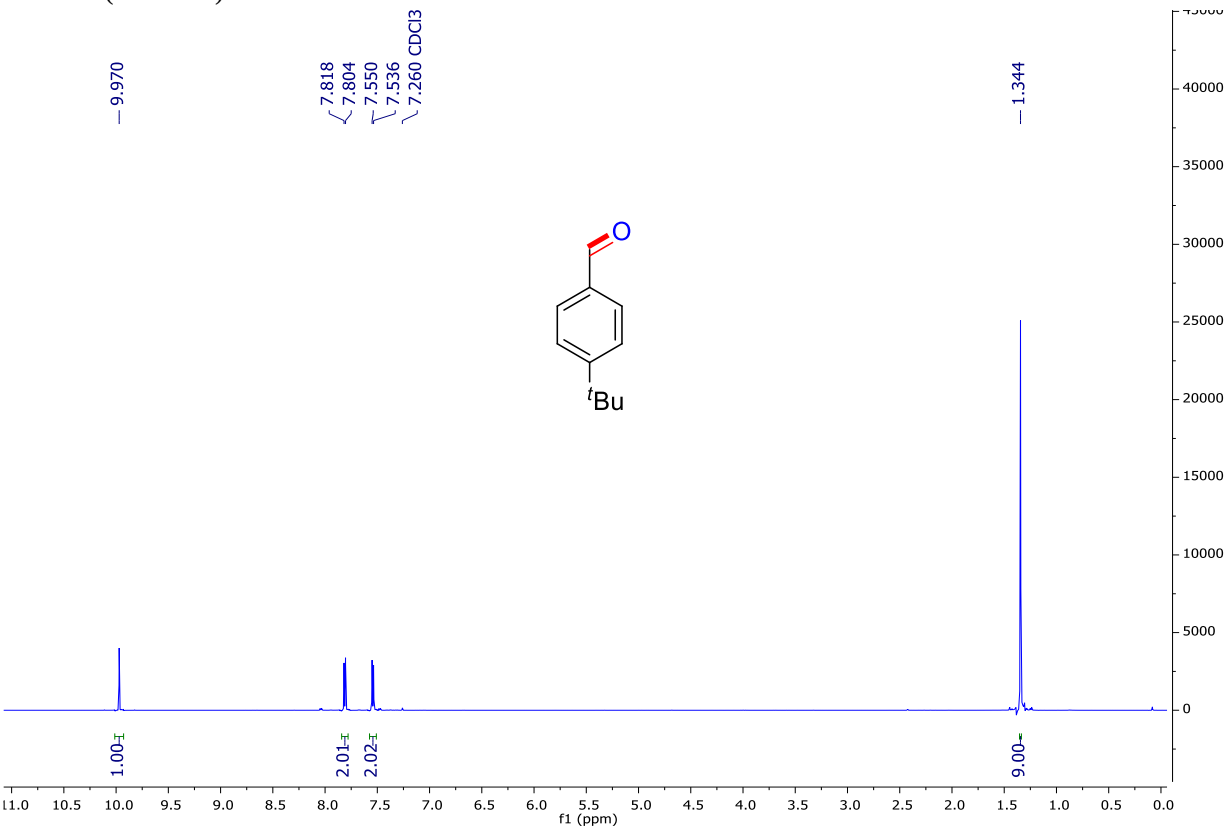


^{13}C{^{1}H} NMR (150 MHz)

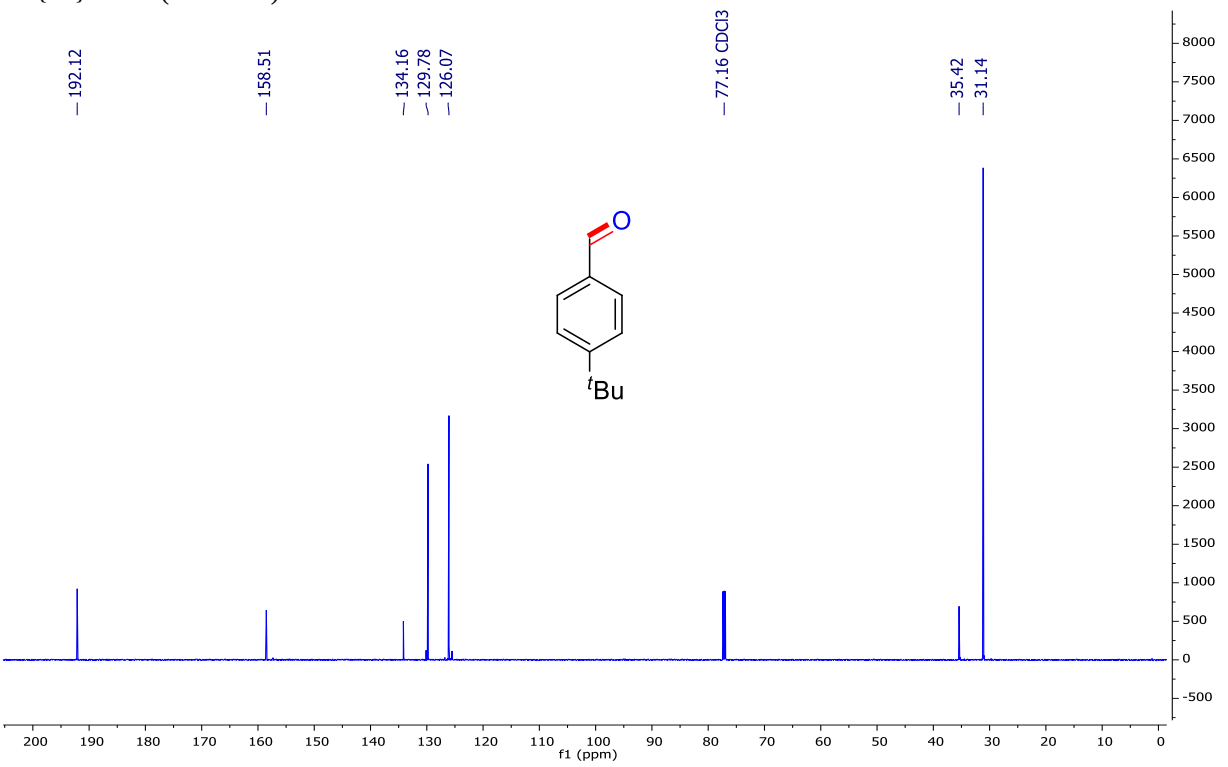


4-(tert-butyl)benzaldehyde (Table 3, entry **4h**)

^{1}H NMR (600 MHz)

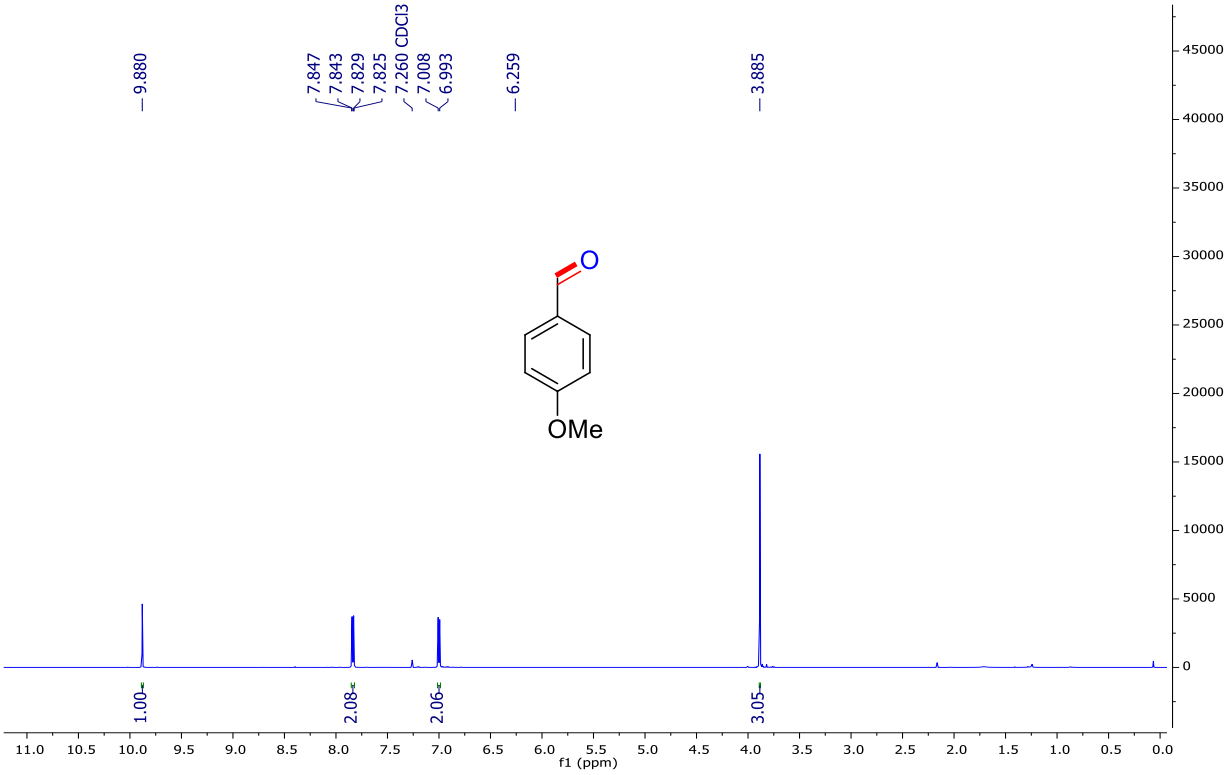


$^{13}C\{^{1}H\}$ NMR (150 MHz)

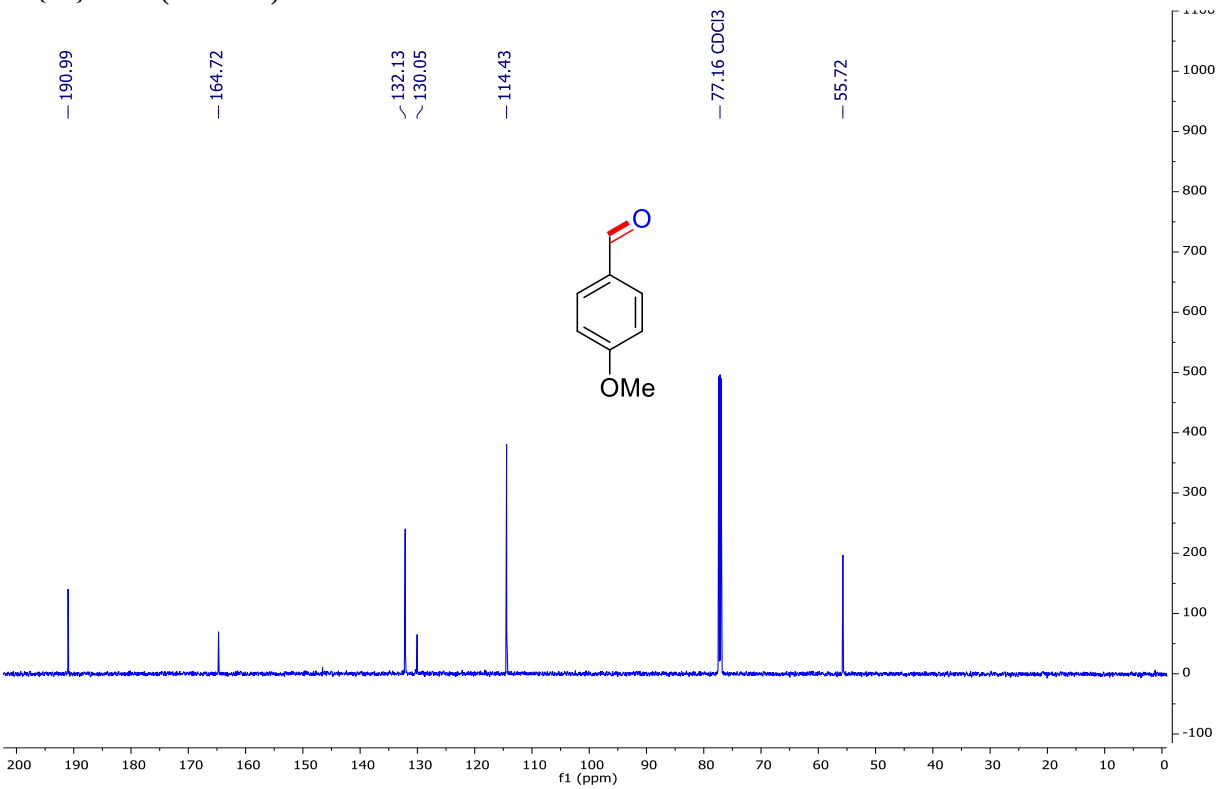


4-methoxybenzaldehyde (Table 3, entry **4i**)

^{1}H NMR (600 MHz)

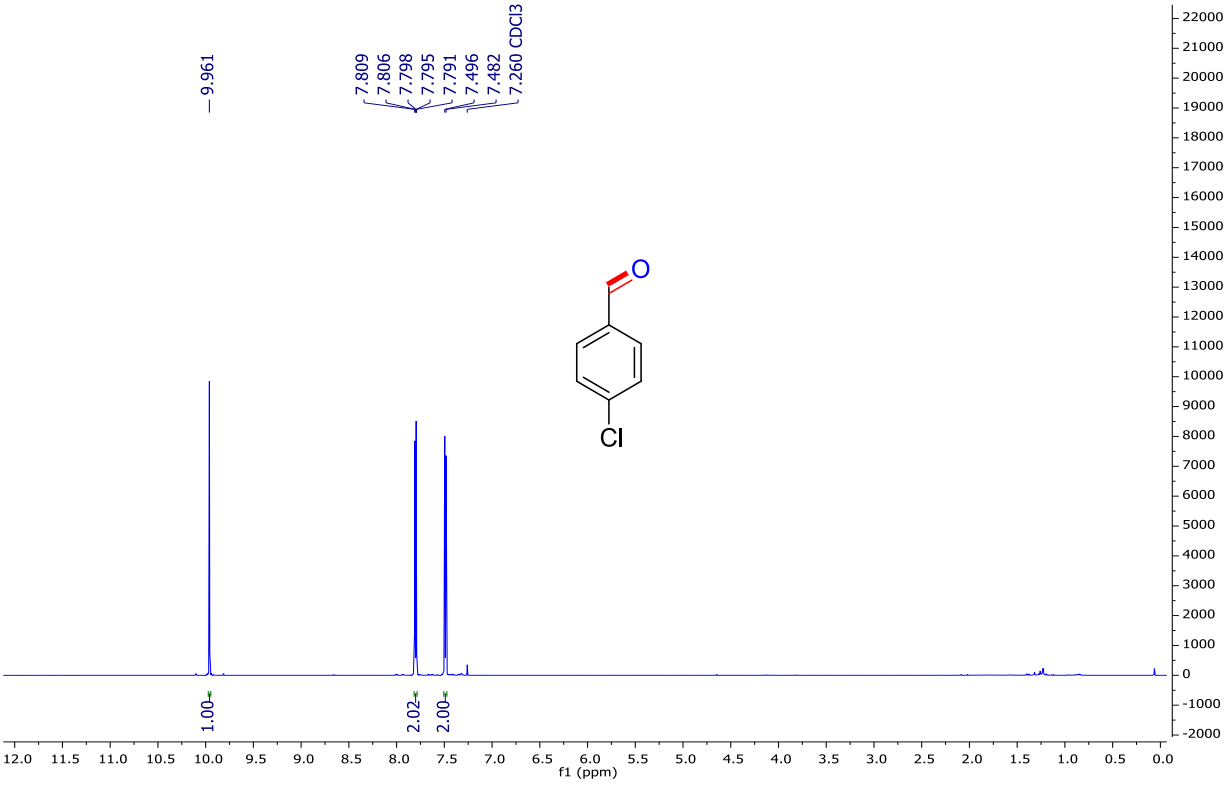


$^{13}C\{^{1}H\}$ NMR (150 MHz)

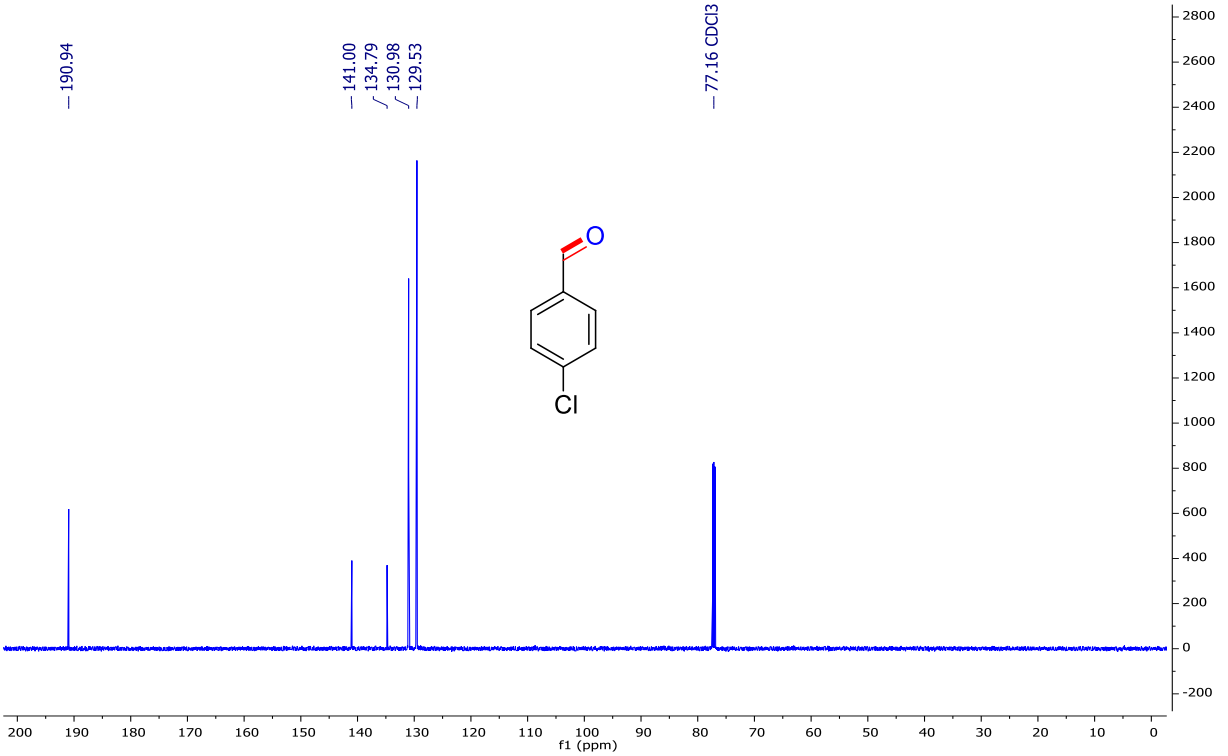


4-chlorobenzaldehyde (Table 3, entry **4j**)

^{1}H NMR (600 MHz)

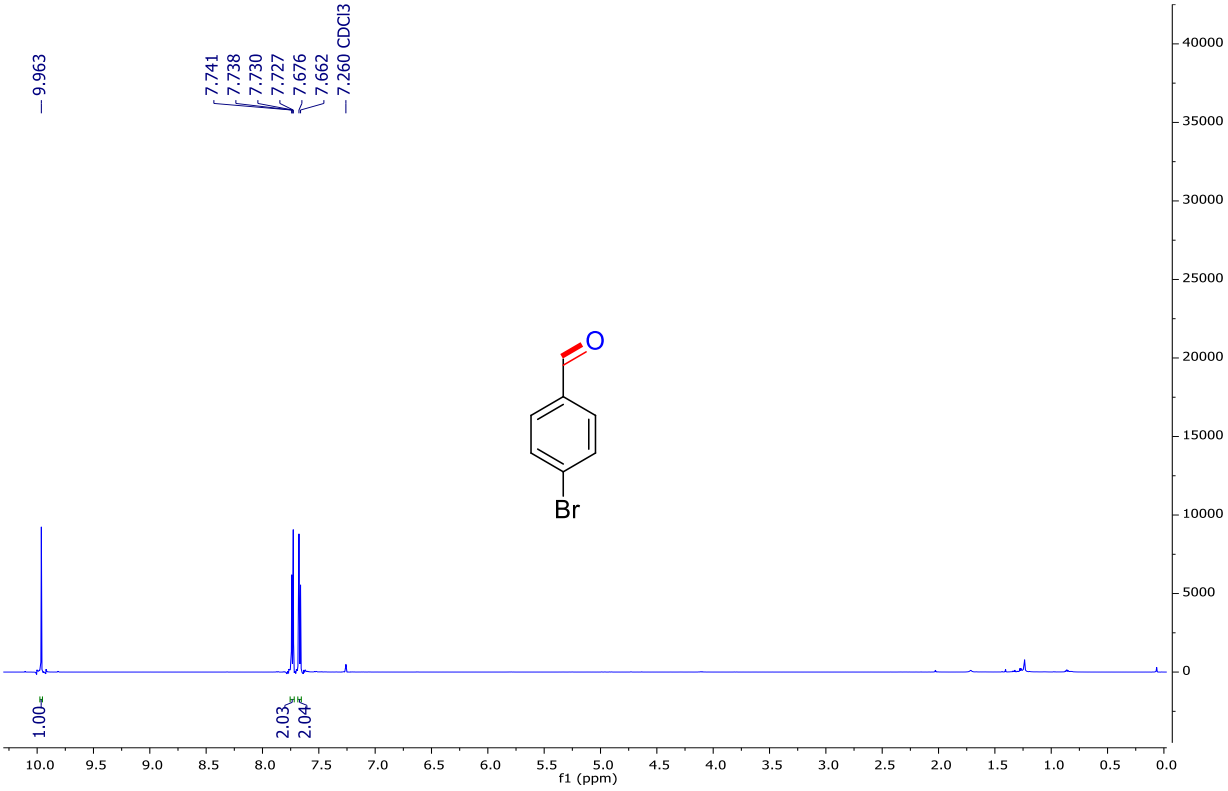


$^{13}C\{^{1}H\}$ NMR (150 MHz)

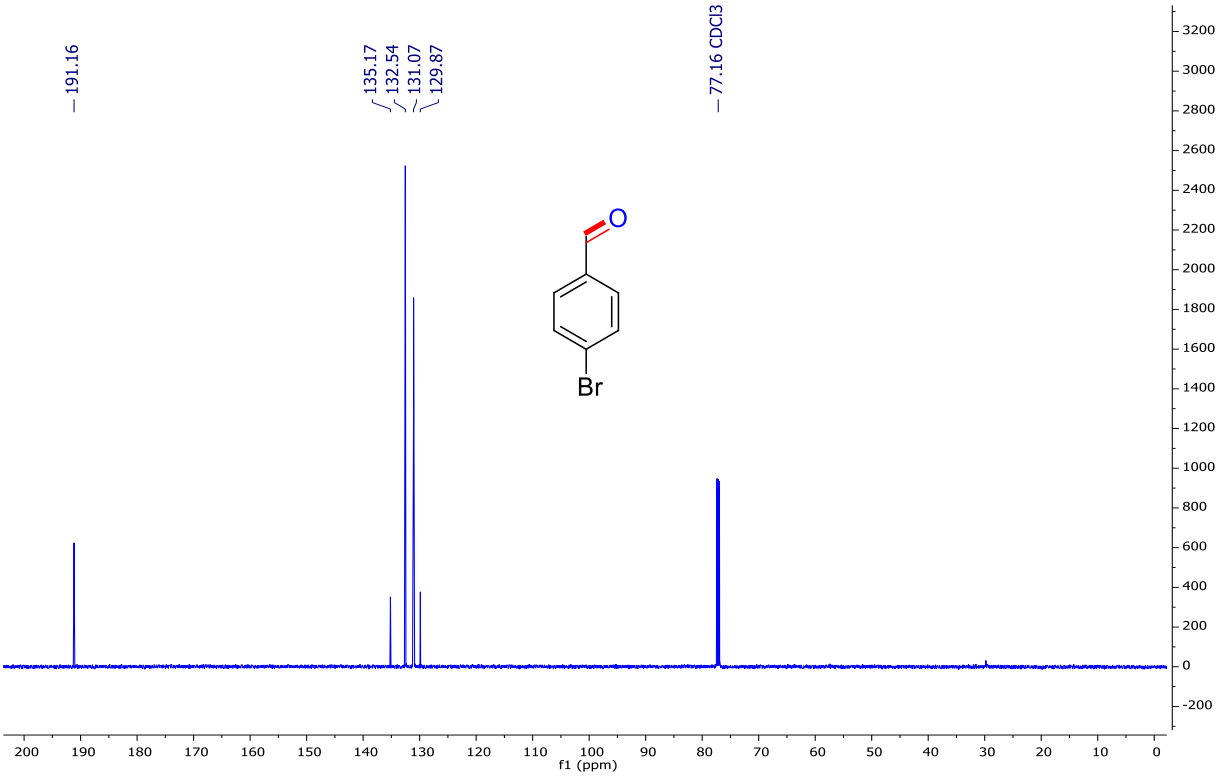


4-bromobenzaldehyde (Table 3, entry **4k**)

^{1}H NMR (600 MHz)

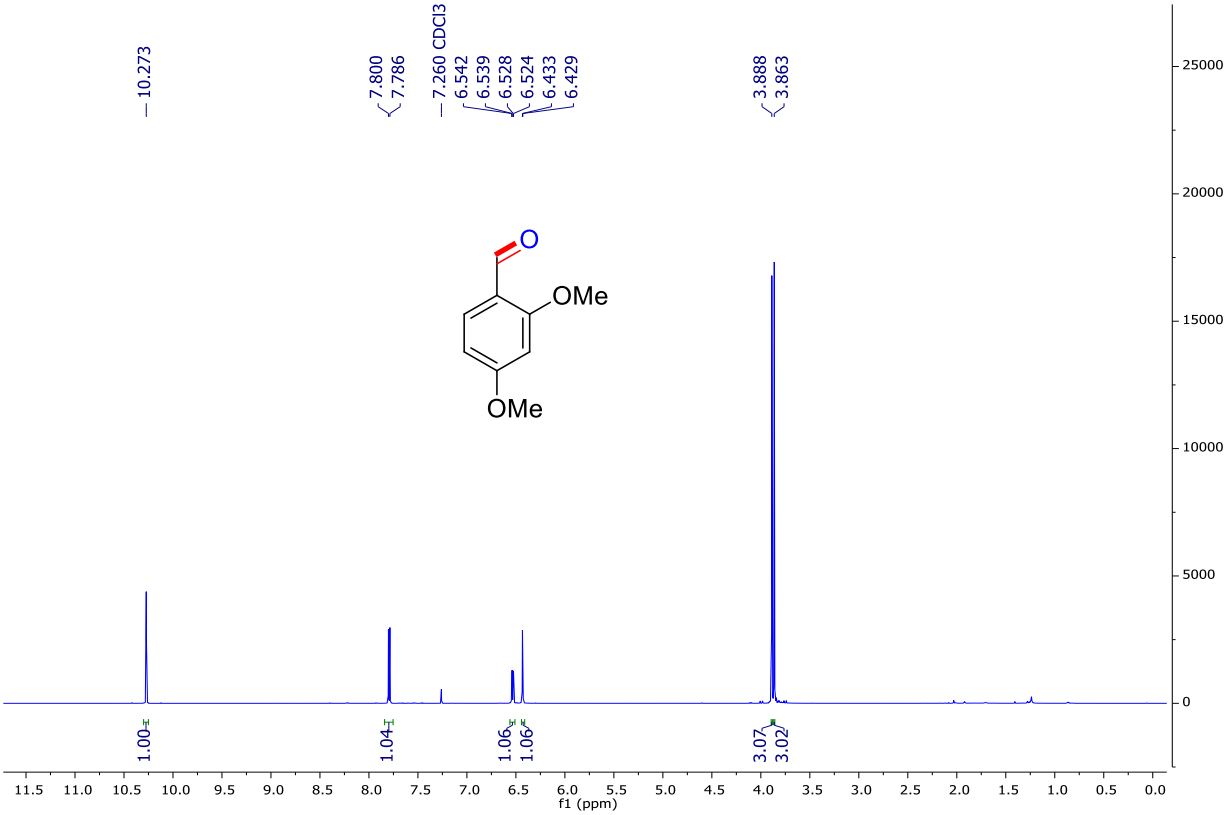


$^{13}C\{^{1}H\}$ NMR (150 MHz)

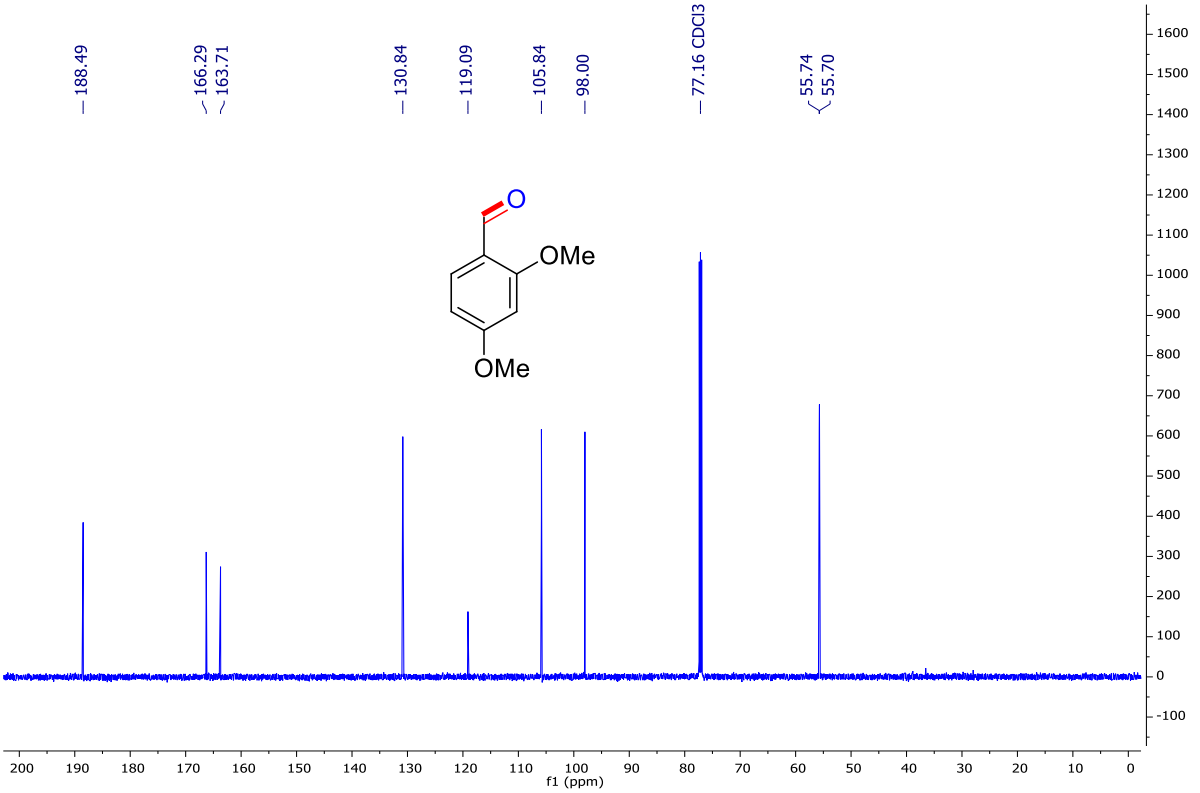


2,4-dimethoxybenzaldehyde (Table 3, entry **4l**)

^{1}H NMR (600 MHz)

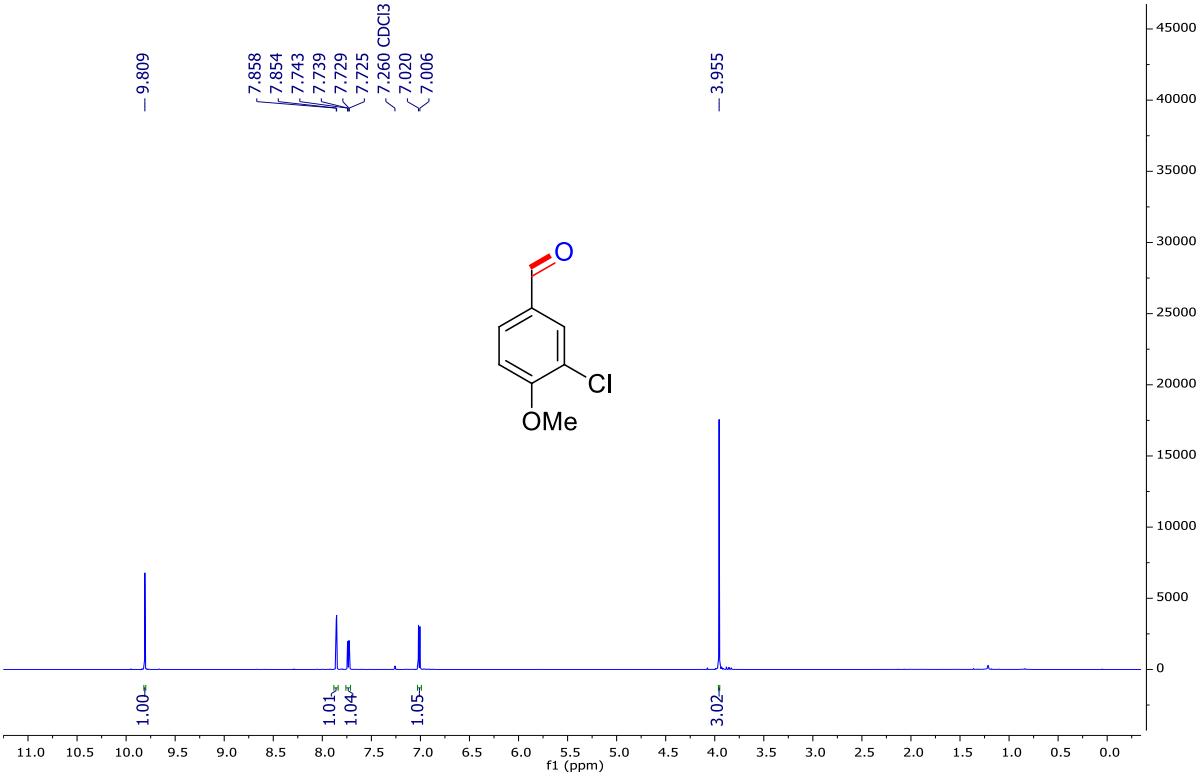


$^{13}C\{^{1}H\}$ NMR (150 MHz)

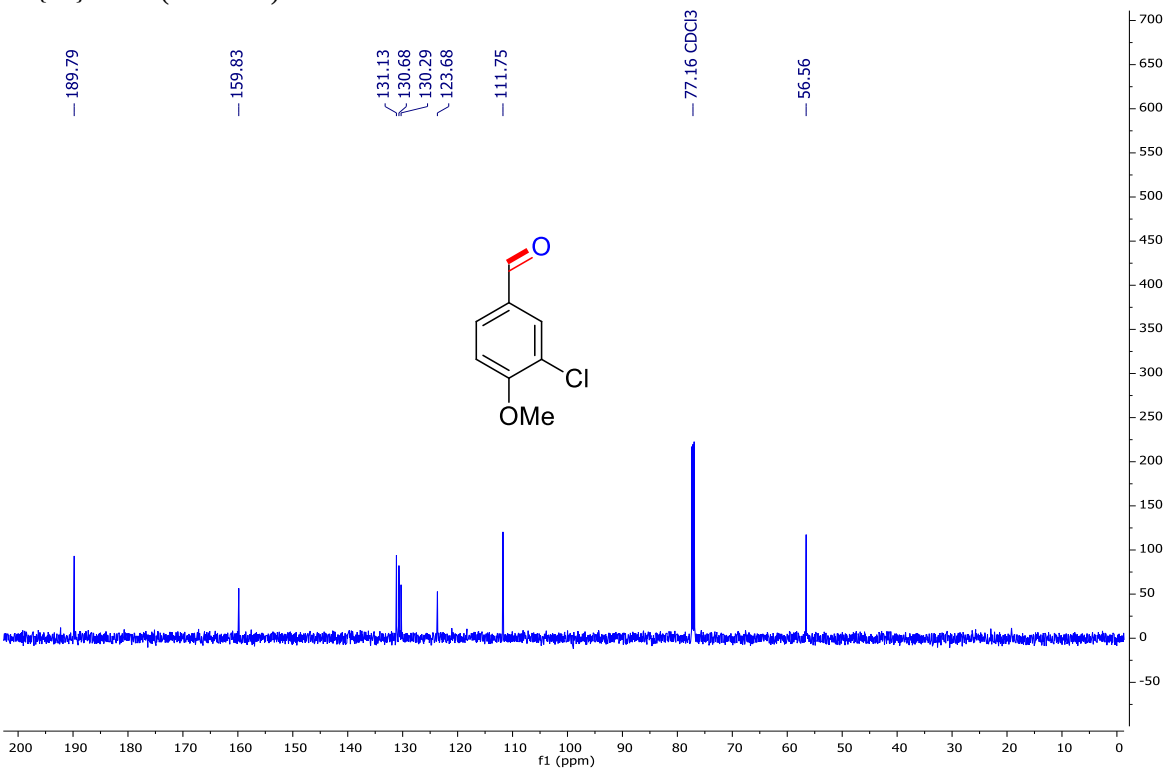


2-chloro-4-methoxybenzaldehyde (Table 3, entry **4m**)

^{1}H NMR (600 MHz)

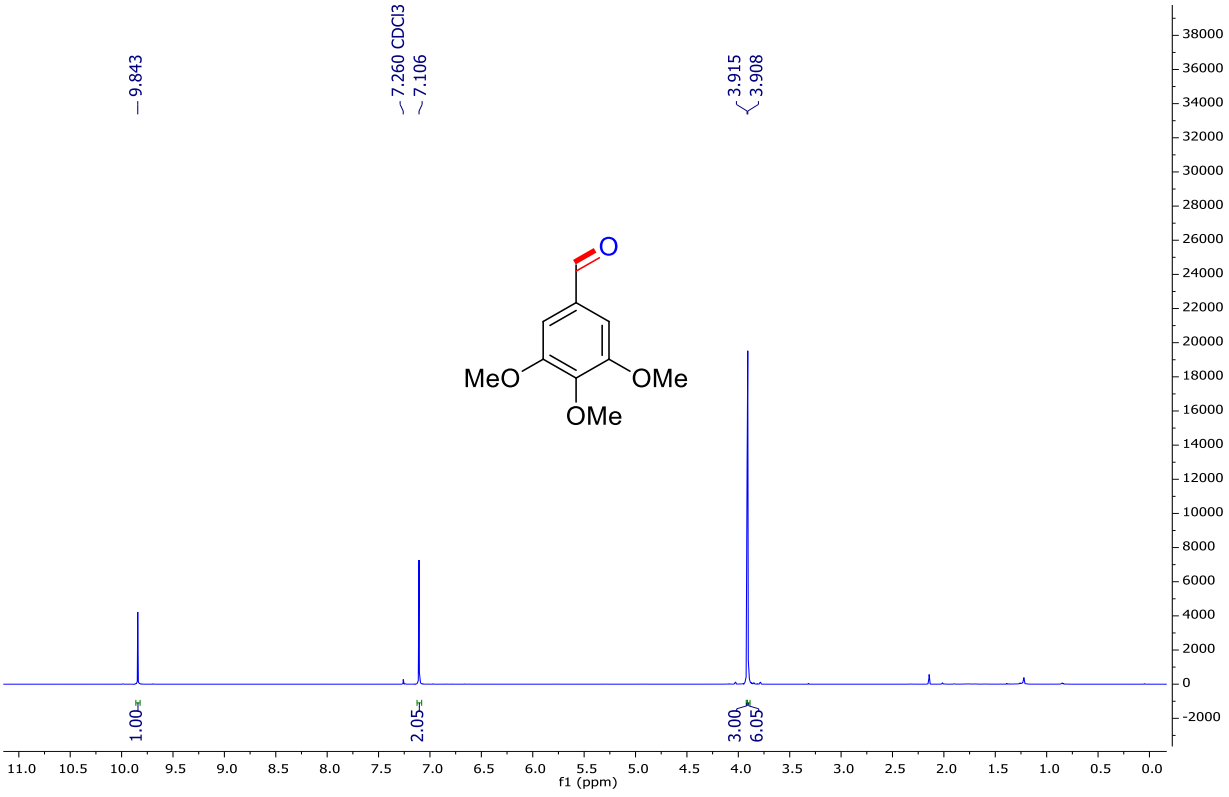


$^{13}C\{^{1}H\}$ NMR (150 MHz)

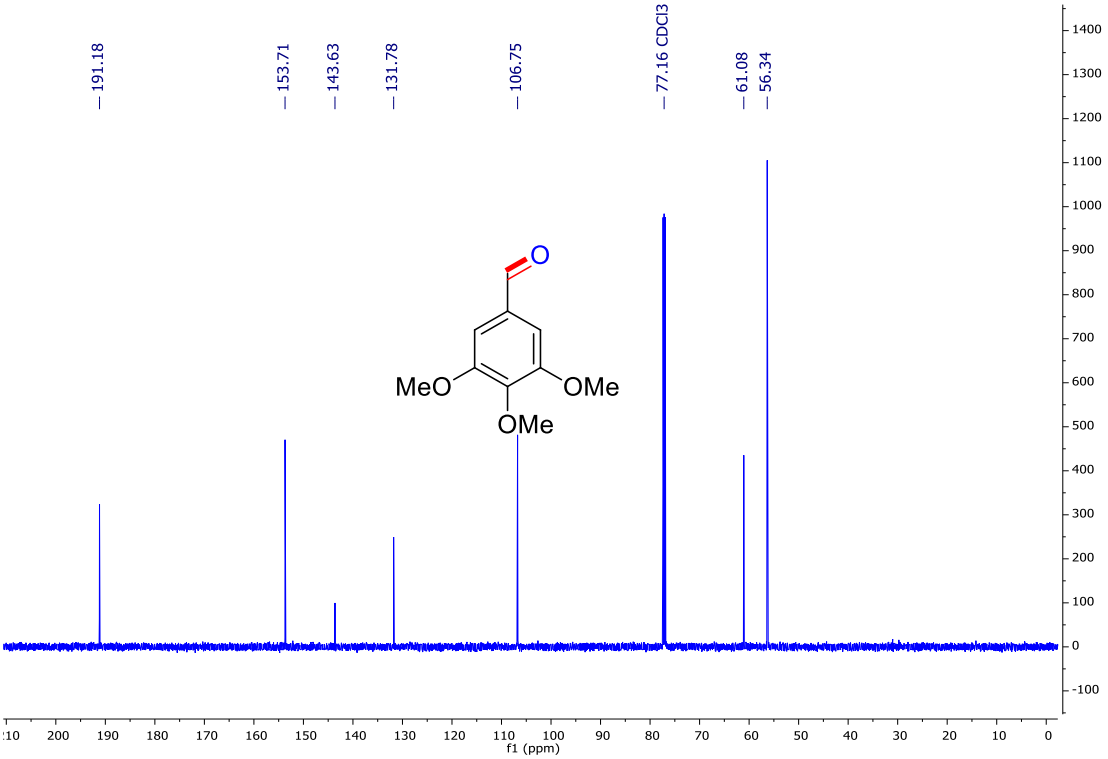


3,4,5-trimethoxybenzaldehyde (Table 3, entry **4n**)

^{1}H NMR (600 MHz)

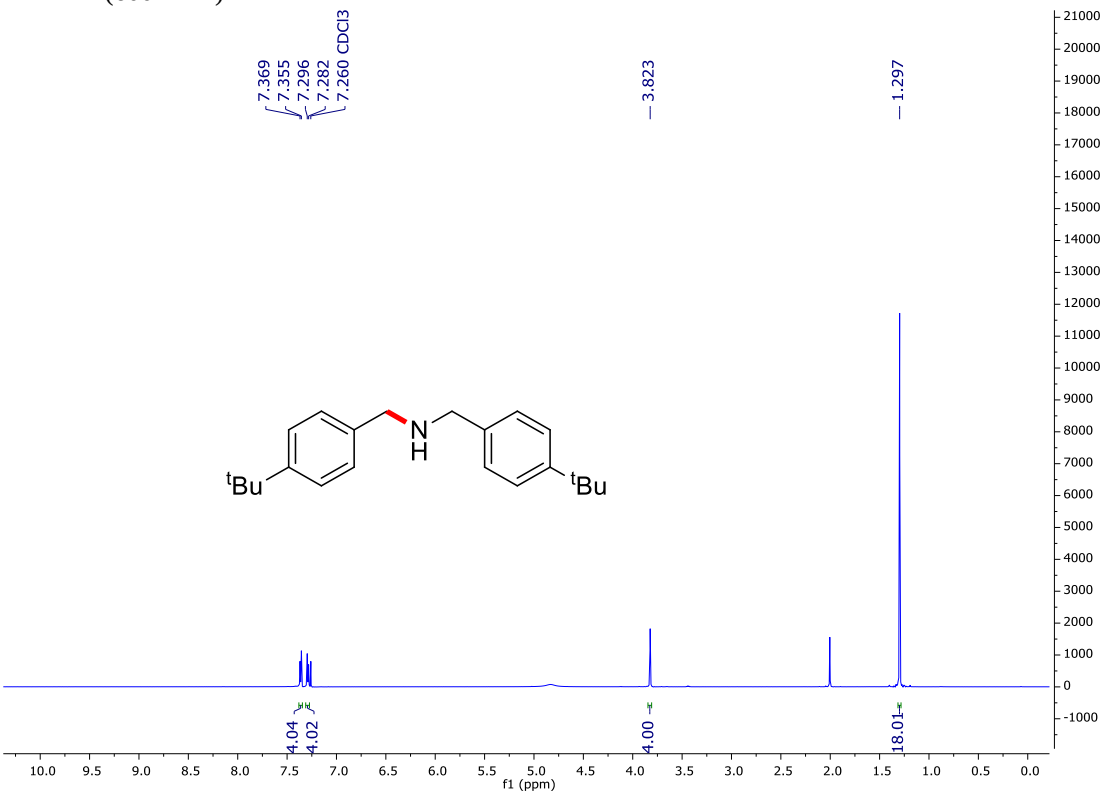


$^{13}C\{^{1}H\}$ NMR (150 MHz)

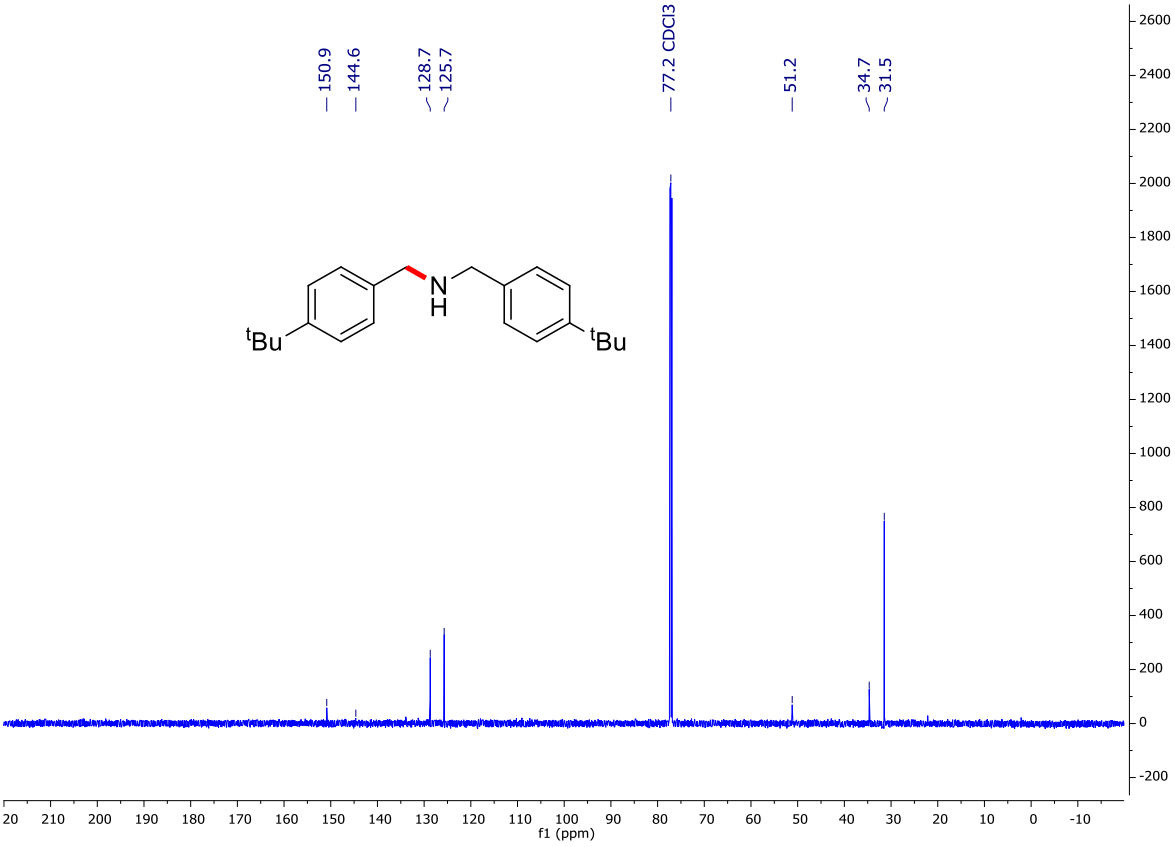


bis(4-(tert-butyl)benzyl)amine (Scheme 3, **6a**)

^{1}H NMR (600 MHz)



$^{13}C\{^{1}H\}$ NMR (150 MHz)



7. References:

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2. a) A. D. Becke, J. Chem. Phys. 1993, 98, 5648; b) C. Lee, W. Yang, R. G. Parr, *Phys. Rev. B* **1988**, 37, 785.