Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Copper catalyzed room temperature lactonization of aromatic C-H bond: A novel and efficient approach for the synthesis of dibenzopyranones

Raju Singha, Shubhendu Dhara, Munmun Ghosh and Jayanta K. Ray*

Department of Chemistry, Indian Institute of Technology, Kharagpur721302, India

* Corresponding author. Tel.: +91 3222283326; fax: +91 3222282252.

E-mail address: jkray@chem.iitkgp.ernet.in (J. K. Ray).

Table of contents **Page**

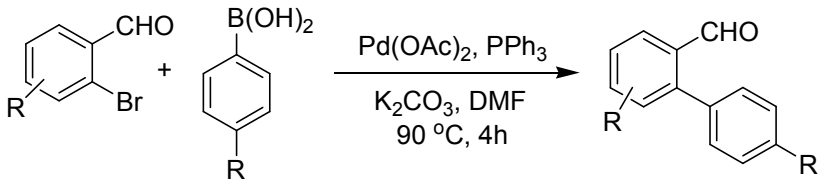
1. General methods:

High quality reagents were purchased from Sigma Aldrich. Analytical grade commercial reagents and solvents were purified by standard procedures prior to use. Chromatographic purification was done with 60-120 mesh silica gel (Merck). For reaction monitoring, pre-coated silica gel 60 F254 sheets (Merck) were used. ^{1}H NMR (200 MHz) spectra were recorded on a BRUCKER-AC 200 MHz spectrometer. Chemical shifts are reported in ppm from tetramethylsilane with the solvent resonance as the internal standard (deuterochloroform: 7.26 ppm). Data are reported as follows: chemical shifts, multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet, dd = double doublet, bs = broad singlet), coupling constant (Hz). ^{13}C NMR (50 MHz) spectra were recorded on a BRUKER-AC 200 MHz. Spectrometer with complete proton decoupling. Chemical shifts are reported in ppm from tetramethylsilane with the solvent resonance as the internal standard (deuterochloroform: 77.23 ppm). HRMS (ESI) spectra were taken using Waters Xevo G2 QTof mass spectrometer.

2. General procedures

2.1 *General procedure for the synthesis biphenyl-2-carbaldehydes:* ***GP-1***

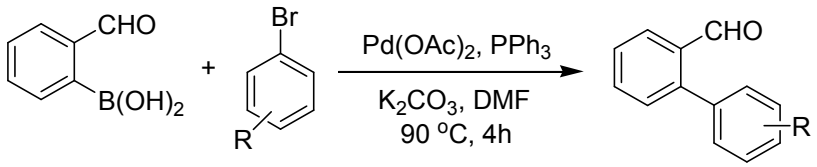
The compounds were synthesized using our previously reported Suzuki-Miyaura cross coupling procedure.[1]



The compound 2-bromobenzaldehyde (0.5 mmol), phenylboronic acid (0.6 mmol), K_2CO_3 (0.5 mmol) and PPh_3 (0.25 equiv.) were taken in two-neck round bottomed flask and flashed with nitrogen gas. Then 3 mL of dry DMF was added and degassed with N_2 for 15 min. Then

the catalyst $Pd(OAc)_2$ (5 mol%) was added and the reaction ,mixture was heated at 90 °C for 4h. After completion of the reaction, the reaction mixture was allowed to cool to room temperature and then diluted with water and extracted with ethyl acetate (3 x 20 mL). The combined organic layer was dried over anhydrous Na_2SO_4, evaporated under reduced pressure. Then the crude product was purified by column chromatography using silica gel (60-120 mesh) and hexane/EtOAc as eluent.

2.2 General procedure for the synthesis of biphenyl-2-carbaldehydes: ***GP-2***



The substrate 2-formylphenylboronic acid (0.5 mmol), arylbromide (0.6 mmol), K_2CO_3 (0.5 mmol) and PPh_3 (0.25 equiv.) were taken in two-neck round bottomed flask and flashed with nitrogen gas. Then 3 mL of dry DMF was added and degassed with N_2 for 15 min. Then the catalyst $Pd(OAc)_2$ (5 mol%) was added and the reaction ,mixture was heated at 90 °C for 4h. After completion of the reaction, the reaction mixture was allowed to cool to room temperature and then diluted with water and extracted with ethyl acetate (3 x 20 mL). The combined organic layer was dried over anhydrous Na_2SO_4, evaporated under reduced pressure. Then the crude product was purified by column chromatography using silica gel (60-120 mesh) and hexane/EtOAc as eluent.

2.3 General lactonization procedure for synthesis of dibenzopyranones: ***GP-3***

The substrate *o*-arylbenzaldehyde (0.25 mmol) and CuCl (5 mol%) were taken in a single neck round bottomed flask and then 2.0 mL DMSO was added. The reaction mixture was stirred and then TBHP (70% in water) (6 equiv.) was added drop wise. The stirring was

continued at room temperature for 4h. After completion of the reaction, the reaction mixture was diluted with water and extracted with ethyl acetate (3 x 20 mL). The combined organic layer was evaporated under reduced pressure and the crude product was purified by column chromatography using silica gel (60-120 mesh) and hexane/ethyl acetate as eluent.

2.4 general procedure for the synthesis of 20phenylbenzylalcohol: ***GP-4***

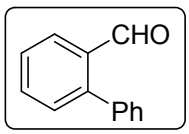
Acetonitrile (4 mL) was added to the substrate biphenyl-2-carbaldehyde (4a) (1 mmol) and $NaBH_4$ (2 mmol) taken in a round bottomed flask. The reaction mixture was stirred at room temperature for 3h. After completion of the reaction, the reaction mixture was diluted with water and extracted with ethyl acetate (3 x 20 mL). The combined organic layer was evaporated under reduced pressure and the crude product was purified by column chromatography using silica gel (60-120 mesh) and hexane/ethyl acetate as eluent.

1.5 General procedure for the synthesis of biphenyl-2-carboxylic acid: ***GP-5***

Acetonitrile (4 mL) was added to the substrate biphenyl-2-carbaldehyde (4a) (1 mmol) taken in a round bottomed flask and cooled to 0 °C. A solution of NaH_2PO_4 (1 mmol) in 1mL of water was added. Then few drops of H_2O_2 and $NaClO_2$ (1.5 mmol dissolved in minimum amount of water) were added. The reaction mixture was stirred at 0 °C to room temperature for 2h. After completion of the reaction, the reaction mixture was diluted with water and extracted with ethyl acetate (3 x 20 mL). The combined organic layer was evaporated under reduced pressure and the crude product was purified by column chromatography using silica gel (60-120 mesh) and hexane/ethyl acetate as eluent.

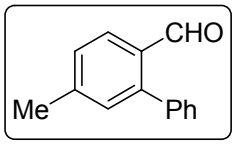
1. Spectroscopic data of starting materials

Biphenyl-2-carbaldehyde (4a):



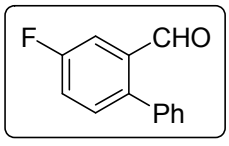
According to ***GP-1*** with 2-bromobenzaldehyde and phenylboronic acid afforded biphenyl-2-carbaldehyde (4a) in 90% yield as a Colourless liquid; ^{1}H NMR ($CDCl_3$, 200 MHz) δ: 7.34-7.67 (8H, m), 8.05 (1H, d, *J* = 7.4 Hz), 10.0 (1H, s); ^{13}C NMR ($CDCl_3$, 50 MHz) δ: 127.6, 127.8, 128.1, 128.4 (2 x CH), 130.1 (2 x CH), 130.8, 133.6, 133.7, 137.7, 145.9, 192.3. The spectral data are in well agreement with the literature reported data.[1]

5-Methyl-biphenyl-2-carbaldehyde (4b):



According to the ***GP-1*** with 4-methyl-2-bromobenzaldehyde and phenylboronic acid afforded 5-methyl-biphenyl-2-carbaldehyde (4b) in 91% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.45 (3H, s), 7.24-7.53 (7H, m), 7.96 (1H, d, *J* = 8.0 Hz), 9.95 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 21.8, 127.7, 128.0, 128.4 (2 x CH), 128.7, 130.1 (2 x CH), 131.4, 131.5, 137.9, 144.5, 146.1, 192.0. Spectral data are in well agreement with the literature reported data.[2]

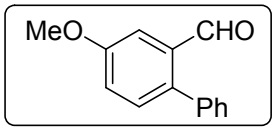
4-Fluoro-biphenyl-2-carbaldehyde (4c):



According to the ***GP-1*** with 5-fluoro-2-bromobenzaldehyde and phenylboronic acid affordd 4-fluoro-biphenyl-2-carbaldehyde in 93% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.32-7.51 (7H, m), 7.69 (1H, dd, *J* = 8.8, 2.6 Hz), 9.92 (1H, d, *J* = 3.2 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 113.5 (CH, d, *J* = 22.0 Hz), 120.7 (CH, d, *J* = 21.5 Hz), 128.3, 128.5 (2 x CH), 130.1 (2 x CH), 132.7 (CH, d, *J*

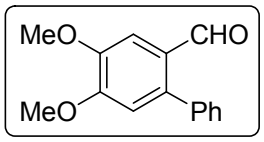
= 7.0 Hz), 135.2 (C, d, *J* = 6.5 Hz), 136.7, 142.0 (C, d, *J* = 3.5 Hz), 162.1 (CF, d, *J* = 247.5 Hz), 190.9. Spectral data are in well agreement with the literature reported data.[2]

4-Methoxy-biphenyl-2-carbaldehyde (4d):



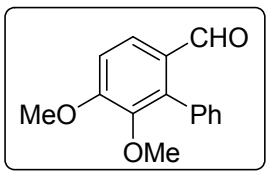
According to ***GP-1*** with 5-methoxybenzaldehyde and phenylboronic acid afforded 4-methoxybiphenyl-2-carbaldehyde (4d) in 87% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.82 (3H, s), 7.10-7.54 (8H, m), 9.92 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.3, 109.8, 121.1, 127.7, 128.3 (2 x CH), 130.1 (2 x CH), 131.9, 134.4, 137.4, 138.8, 159.0, 191.9. Spectral data are in well agreement with the literature reported data.[3]

4,5-Dimethoxy-biphenyl-2-carbaldehyde (4e):



According to the GP-1 with 4,5-dimethoxy-2-bromobenzaldehyde and phenylboronic acid afforded 4,5-dimethoxy-biphenyl-2-carbaldehyde (4e) in 86% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.86 (3H, s), 3.87 (3H, s), 6.78 (1H, s), 7.30-7.39 (5H, m), 7.44 (1H, s), 9.72 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.9, 56.0, 108.4, 112.5, 126.7, 127.8, 128.2 (2 x CH), 130.1 (2 x CH), 137.4, 141.3, 148.6, 153.3, 190.8. The spectral data are in well agreement with the literature reported data.[2]

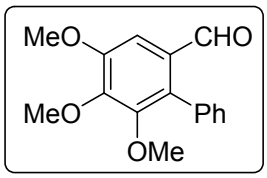
5,6-Dimethoxy-biphenyl-2-carbaldehyde (4f):



According to the ***GP-1*** with 2-bromo-3,4-dimethoxybenzaldehyde and phenylboronic acid afforded 5,6-dimethoxybiphenyl-2-carbaldehyde in 76% yield as colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.56 (3H, s), 3.97 (3H, s), 7.05 (1H, d, *J* = 8.8 Hz), 7.32-7.49 (5H, m), 7.85 (1H, d, *J* = 8.6 Hz), 9.61 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 56.1, 60.8, 111.5, 124.8, 128.0 (3 x CH), 130.6

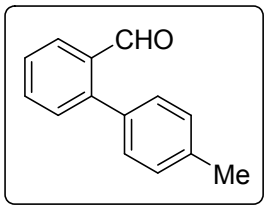
(2 x CH), 133.0, 140.5, 146.1, 156.3, 157.8, 191.4. **HRMS** (ESI) m/z $[M+H]^+$ for $C_{15}H_{15}O_3^+$ calculated: 243.1016, found 243.1018.

4,5,6-Trimethoxy-biphenyl-2-carbaldehyde (4g):



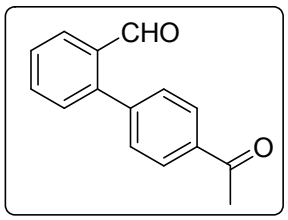
According to the ***GP-1*** with 3,4,5-trimethoxy-2-bromobenzaldehyde and phenylboronic acid afforded the compound (4g) in 78% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.57 (3H, s), 3.90 (3H, s), 3.97 (3H, s), 7.28-7.46 (6H, m), 9.62 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 56.0, 61.0 (2 x CH_3), 105.2, 127.8, 127.9 (2 x CH), 129.6, 131.0 (2 x CH), 132.7, 134.5, 147.6, 151.0, 153.1, 191.2. The spectral data are in well agreement with the literature reported data.[4]

4/-Methyl-biphenyl-2-carbaldehyde (4h):



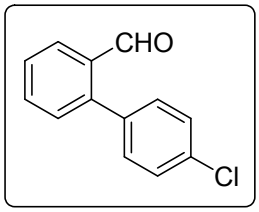
According to the ***GP-2*** with 2-formaylphenylboronic acid and *p*-bromotoluene afforded the compound (4h) in 85% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.44 (3H, s), 7.29-7.68 (7H, m), 8.03 (1H, dd, *J* = 7.4, 0.8 Hz), 10.01 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 21.3, 127.7 (2 x CH), 129.3 (2 x CH), 130.2 (2 x CH), 130.9, 133.7, 133.9, 134.9, 138.2, 146.1, 192.7. Spectral data are in well agreement with the literature reported data.[2]

4/-Acetyl-biphenyl-2-carbaldehyde (4i):



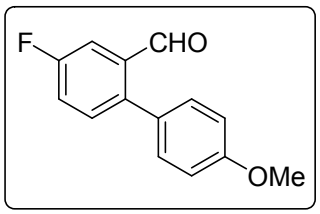
According to the ***GP-2*** with 2-formylphenylboronic acid and 4-bromoacetophenone afforded 4/-acetyl-biphenyl-2-carbaldehyde in 88% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.62 (3H, s), 7.38-7.67 (5H, m), 7.96-8.08 (3H, m), 9.91 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 26.7, 128.1, 128.4 (2 x CH), 128.5, 130.3 (2 x CH), 130.6, 133.6, 133.8, 136.6, 142.6, 144.5, 191.6, 197.5. Spectral data are in well agreement with the literature reported data.[5]

4′-Chloro-biphenyl-2-carbaldehyde (4j):



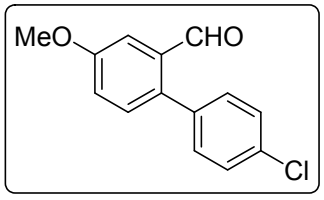
According to the ***GP-1*** with 2-bromobenzaldehyde and 4-chlorophenylboronic acid afforded 4′-chlorobiphenyl-2-carbaldehyde in 90% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.30-7.67 (7H, m), 8.03 (1H, dd, *J* = 7.6, 1.2 Hz), 9.98 (1H, d, *J* = 0.6 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 128.0, 128.2, 128.7 (2 x CH), 130.7, 131.3 (2 x CH), 133.7 (1CH+1C), 134.5, 136.3, 144.5, 191.8. Spectral data are in well agreement with the literature reported data.[1]

4-Fluoro-4′-methoxy-biphenyl-2-carbaldehyde (4k):



According to the ***GP-1*** with 2-bromo-5-fluorobenzaldehyde and 4-methoxyphenylboronic acid afforded the compound (4k) in 92% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.87 (3H, s), 7.01 (2H, d, *J* = 8.4 Hz), 7.25-7.48 (4H, m), 7.65 (1H, dd, *J* = 8.8, 6.6 Hz), 9.92 (1H, d, *J* = 3.6 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.3, 113.5 (CH, d, *J* = 22.0 Hz), 114.1 (2 x CH), 120.7 (CH, d, *J* = 22.0 Hz), 129.0, 131.3 (2 x CH), 132.8 (CH, d, *J* = 7.0 Hz), 135.2 (C, d, *J* = 6.0 Hz), 141.9, 159.9, 162.0 (CF, d, *J* = 247.0 Hz), 191.3. **HRMS** (ESI) m/z $[M+H]^+$ for $C_{14}H_{12}FO_2^+$ calculated: 231.0816, found 231.0813.

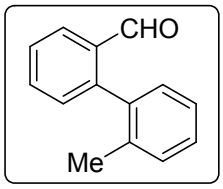
4′-Chloro-4-methoxy-biphenyl-2-carbaldehyde (4l):



According to the ***GP-1*** with 2-bromo-5-methoxybenzaldehyde and 4-chlorophenylboronic acid afforded 4′-Chloro-4-methoxy-biphenyl-2-carbaldehyde (4l) in 86% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.87 (3H, s), 7.14-7.49 (7H, m), 9.91 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.5, 110.3, 121.3, 128.6 (2 x CH), 131.4 (2 x CH), 131.9, 134.0, 134.5, 136.0,

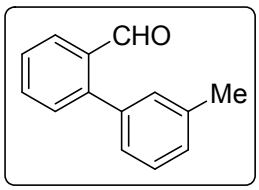
137.5, 159.4, 191.6. **HRMS** (ESI) m/z $[M+H]^+$ for $C_{14}H_{12}ClO_2^+$ calculated: 247.0520 , found 247.0526.

2/-Methyl-biphenyl-2-carbaldehyde (4m):



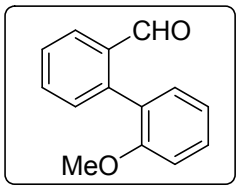
According to the ***GP-2*** with 2-formylphenylboronic acid and 2-bromotoluene afforded 2/-methyl-biphenyl-2-carbaldehyde in 81% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.12 (3H, s), 7.15-7.54 (6H, m), 7.61-7.69 (1H, m), 8.05 (1H, dd, *J* = 7.8, 1.2 Hz), 9.77 (1H, d, *J* = 0.6 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 20.4, 125.8, 127.2, 127.9, 128.4, 130.2, 130.3, 130.9, 133.9, 134.0, 136.3, 137.6, 145.8, 192.4. Spectral data are in well agreement with the literature reported data.[2]

3/-Methyl-biphenyl-2-carbaldehyde (4n):



According to the ***GP-2*** with 2-formylphenylboronic acid and 3-bromotluene afforded 3/-methyl-biphenyl-2-carbaldehyde in 85% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.44 (3H, s), 6.98-7.68 (7H, m), 8.03 (1H, dd, *J* = 7.6, 1.0 Hz), 10.0 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 21.6, 127.4, 127.6, 127.8, 128.5, 129.0, 130.9, 131.0, 133.7, 133.9, 137.9, 138.3, 146.3, 192.8. The spectral data are in well agreement with the literature reported data.[2]

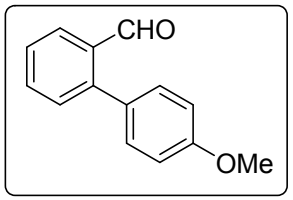
2/-Methoxy-biphenyl-2-carbaldehyde (4o):



According to the GP-2 with 2-formylphenylboronic acid and 1-bromo-2-methoxybenzene afforded 2/-Methoxy-biphenyl-2-carbaldehyde (4o) in 76% yield as a colourless liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.73 (3H, s), 6.97-7.13 (2H, m), 7.28-7.51 (4H, m), 7.60-7.68 (1H, m), 8.03 (1H, dd, *J* = 7.6, 1.2 Hz), 9.83 (1H, s); **^{13}H NMR** ($CDCl_3$, 50 MHz) δ: 55.3, 110.7, 121.5, 126.6, 126.8, 127.8,

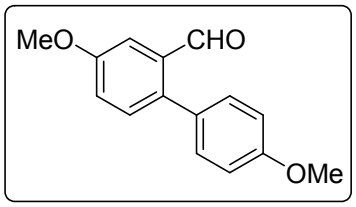
130.0, 131.2, 131.4, 133.7, 134.0, 141.8, 156.5, 192.6. The spectral data are in well agreement with the literature reported data.[6]

4/-Methoxy-biphenyl-2-carbaldehyde (4p):



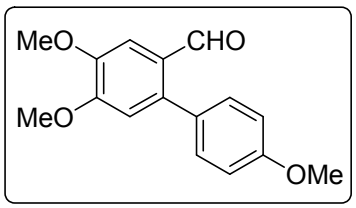
According to the ***GP-1*** with 2-bromobenzaldehyde and 4-methoxyphenylboronic acid afforded4/-Methoxy-biphenyl-2-carbaldehyde (4p) in 82% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.86 (3H, s), 7.10 (2H, d, *J* = 8.8 Hz), 7.29-7.33 (2H, m), 7.41-7.49 (2H, m), 7.57-7.65 (1H, m), 8.03 (1H, d, *J* = 8.2 Hz), 10.02 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.3, 113.9 (2 x CH), 127.3, 127.6, 129.9, 130.8, 131.3 (2 x CH), 133.5, 133.7, 145.6, 159.7, 192.4. Spectral data are in well agreement with the literature reported data.[2]

4,4/-Dimethoxy-biphenyl-2-carbaldehyde (4q):



According to the ***GP-1*** with 2-bromo-5-methoxybenzaldehyde and 4-methoxyphenylboronic acid afforded 4,4/-Dimethoxy-biphenyl-2-carbaldehyde (4q) in 84% yield as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.78 (3H, s), 3.80 (3H, s), 6.92 (2H, d, *J* = 8.8 Hz), 7.07-7.29 (4H, m), 7.44 (1H, d, *J* = 2.8 Hz), 9.90 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.0, 55.2, 109.8, 113.7 (2 x CH), 121.0, 129.5, 131.2 (2 x CH), 131.9, 134.3, 138.5, 158.7, 159.3, 192.0. Spectral data are in well agreement with the literature reported data.[7]

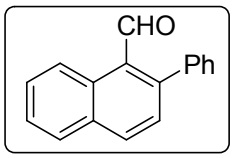
4,4/,5-Trimethoxy-biphenyl-2-carbaldehyde (4r):



According to the ***GP-1*** with 2-bromo-4,5-dimethoxybenzaldehyde and 4-methoxyphenylboronic acid afforded 4,4/,5-Trimethoxy-biphenyl-2-carbaldehyde (4r) in 85% yield as white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.74 (3H, s), 3.84 (3H, s), 3.86 (3H,

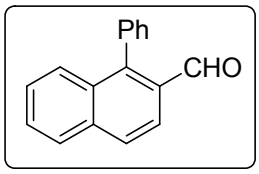
s), 6.75 (1H, s), 6.85-6.89 (2H, m), 7.16-7.21 (2H, m), 7.40 (1H, s), 9.72 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.0, 55.7, 55.8, 108.3, 112.4, 113.5 (2 x CH), 126.6, 129.5, 131.1 (2 x CH), 140.9, 148.3, 153.2, 159.3, 190.8**. HRMS** (ESI) m/z $[M+H]^+$ for $C_{16}H_{17}O_4^+$ calculated: 273.1121, found 273.1123.

2-Phenyl-1-naphthaldehyde (4s):



According to the ***GP-1*** with 2-bromo-1-naphthaldehyde and phenylboronic acid afforded 2-phenyl-1-naphthaldehyde (4s) in 92% yield as a yellow liquid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.42-7.63 (7H, m), 7.67-7.75 (1H, m), 7.90 (1H, d, J = 8.0 Hz), 8.04 (1H, d, J = 8.4 Hz), 9.33 (1H, d, J = 8.6 Hz), 10.2 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 125.8, 126.8, 128.2 (3 x CH), 128.3 (2 x CH), 128.8, 129.2, 130.3, 130.6 (2 x CH), 133.0, 133.9, 138.7, 147.9, 194.6. This compound has been reported in literature.[8]

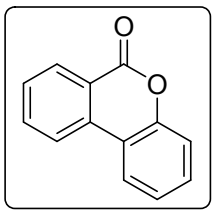
1-Phenyl-2-naphthaldehyde (4t):



According to the ***GP-1*** with 1-bromo-2-naphthaldehyde and phenylboronic acid afforded 1-phenyl-2-naphthaldehyde in 88% yield as a yellow solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.33-7.70 (8H, m), 7.90 (2H, d, J = 8.6 Hz), 8.10 (1H, d, J = 8.8 Hz), 9.95 (1H, s); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 122.1, 126.8, 127.6, 128.1(3 x CH), 128.2, 128.3, 128.7, 130.9 (2 x CH), 131.1, 132.3, 135.1, 136.0, 146.4, 192.4. This compound has been reported in literature.[9]

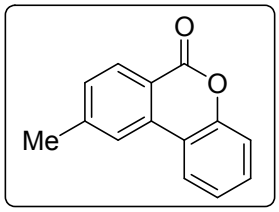
2. Spectroscopic data of products

6*H*-benzo[*c*]chromen-6-one (5a):



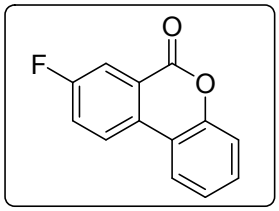
According to the ***GP-3*** with Biphenyl-2-carbaldehyde (4a) afforded 6*H*-benzo[*c*]chromen-6-one (5a) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.30-7.39 (2H, m), 7.45-7.63 (2H, m), 7.79-7.87 (1H, m), 8.04-8.14 (2H, m), 8.40 (1H, d, *J* = 8 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ:117.9, 118.2, 121.4, 121.9, 122.9, 124.7, 129.1, 130.6, 130.8, 134.9, 135.0, 151.5, 161.4. **HRMS** (ESI) for $C_{13}H_8O_2$: Calculated 197.0603 (M^++H) ; Found: 197.0609. The spectral data are in well agreement with our previously reported data.[10]

9-Methyl-6*H*-benzo[*c*]chromen-6-one (5b):



According to the ***GP-3*** with 5-methyl-biphenyl-2-carbaldehyde (4b) afforded 9-methyl-6*H*-benzo[*c*]chromen-6-one (5b) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.55 (3H, s), 7.27-7.46 (4H, m), 7.89 (1H, s), 8.03 (1H, d, *J* = 8.2 Hz), 8.27 (1H, d, *J* = 8.2 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 22.5, 117.9, 118.3, 119.0, 122.0, 122.9, 124.6, 130.3, 130.5, 130.7, 134.9, 146.1, 151.6, 161.5. The spectral data are in well agreement with our previously reported data.[10]

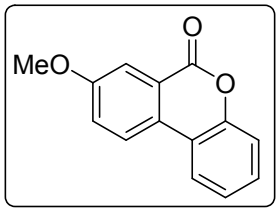
8-Fluoro-6*H*-benzo[*c*]chromen-6-one (5c):



According to the ***GP-3*** with 4-fluoro-biphenyl-2-carbaldehyde (4c) afforded 8-fluoro-6*H*-benzo[*c*]chromen-6-one (5c) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.31-7.40 (2H, m), 7.45-7.60 (2H, m), 7.99-8.17 (3H, m); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 116.4 (d, *J* = 23 Hz), 117.6, 118.1, 122.8, 123.2 (d, *J* = 9 Hz), 123.5, 124.5 (d, *J* = 7.5 Hz), 125.0, 130.6, 131.5, 151.1, 160.3 (d,

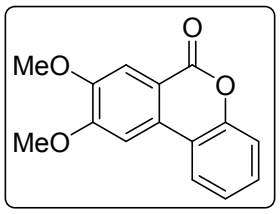
J = 13.5 Hz), 165.2. The spectral data are in well agreement with our previously reported data.[10]

8-Methoxy-6*H*-benzo[*c*]chromen-6-one (5d):



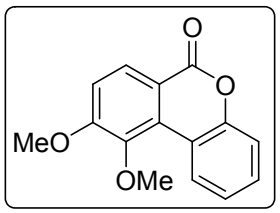
According to the ***GP-3*** with 4-methoxy-biphenyl-2-carbaldehyde (4d) afforded 8-methoxy-6*H*-benzo[*c*]chromen-6-one (5d) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.92 (3H, s), 7.27-7.46 (4H, m), 7.78 (1H, d, J = 2.8 Hz), 7.94-8.03 (2H, m); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.9, 111.4, 117.7, 118.3, 122.3, 122.6, 123.6, 124.4, 124.7, 128.3, 129.5, 150.6, 160.2, 161.4. The spectral data are in well agreement with the literature reported data.[11]

8,9-Dimethoxy-6*H*-benzo[*c*]chromen-6-one (5e):



According to the ***GP-3*** with 4,5-dimethoxy-biphenyl-2-carbaldehyde (4e) afforded 8,9-dimethoxy-6*H*-benzo[*c*]chromen-6-one (5e) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 4.00 (3H, s), 4.09 (3H, s), 7.26-7.45 (4H, m), 7.75 (1H, s), 7.94 (1H, d, J = 8 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 56.5 (2C), 102.9, 110.8, 114.7, 117.9, 118.3, 122.3, 124.6, 129.8, 130.1, 150.4, 151.2, 155.4, 161.3. The spectral data are in well agreement with our previously reported data.[10]

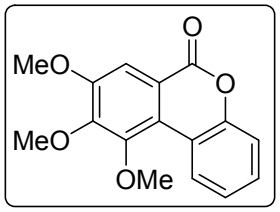
9,10-Dimethoxy-6*H*-benzo[*c*]chromen-6-one (5f):



According to the ***GP-3*** with 5,6-dimethoxybiphenyl-2-carbaldehyde (4f) afforded 9,10-dimethoxy-6*H*-benzo[*c*]chromen-6-one (5f) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.94 (3H, s), 4.04 (3H, s), 7.13 (1H, d, J = 11.2 Hz), 7.20-7.37 (2H, m), 7.44-7.53 (1H, m), 8.28 (1H, d, J = 8.8 Hz), 8.95 (1H, d, J = 8.4 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 56.4, 60.2, 112.8, 115.3, 117.7,

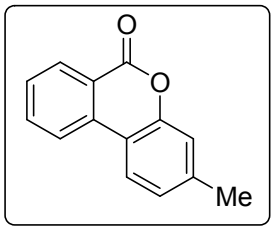
117.8, 124.9, 127.9, 128.4 (1CH+1C), 130.3, 146.0, 151.3, 158.7, 161.3. **HRMS** (ESI) calculated for $C_{15}H_{13}O_2$ $[M + H]^+$: 257.0808; found: 257.0802.

8,9,10-Trimethoxy-6*H*-benzo[*c*]chromen-6-one (5g):



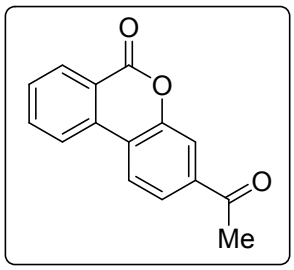
According to the ***GP-3*** with 4,5,6-trimethoxy-biphenyl-2-carbaldehyde (4g) afforded 8,9,10-trimethoxy-6*H*-benzo[*c*]chromen-6-one (5g) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.98 (3H, s), 4.00 (3H, s), 4.04 (3H, s), 7.31-7.49 (3H, m), 7.77 (1H, s), 8.85 (1H, dd, *J* = 7.6 Hz, 1.0 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 56.5, 60.8, 61.4, 108.4, 117.6, 117.7, 117.8, 122.9, 124.9, 126.8, 129.4, 149.3, 150.6, 151.5, 154.0, 161.3. The spectral data are in well agreement with our previously reported data.[10]

3-Methyl-6*H*-benzo[*c*]chromen-6-one (5h):



According to the ***GP-3*** with 4/-methyl-biphenyl-2-carbaldehyde (4h) afforded 3-methyl-6*H*-benzo[*c*]chromen-6-one (5h) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.45 (3H, s), 7.13-7.17 (2H, m), 7.50-7.58 (1H, m), 7.76-7.84 (1H, m), 7.93 (1H, d, *J* = 8.4 Hz), 8.07 (1H, d, *J* = 8.0 Hz), 8.38 (1H, dd, *J* = 8.0, 1.2 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 21.6, 115.6, 118.1, 121.1, 121.6, 122.7, 125.9, 128.6, 130.7, 135.0, 135.2, 141.5, 151.5, 161.7. The spectral data are in well agreement with the literature reported data.[12]

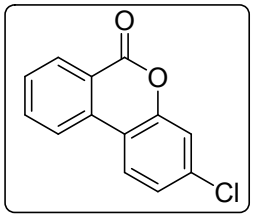
3-Acetyl-6*H*-benzo[*c*]chromen-6-one (5i):



According to the ***GP-3*** with 4/-acetyl-biphenyl-2-carbaldehyde (4i) afforded 3-acetyl-6*H*-benzo[*c*]chromen-6-one (5i) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.66 (3H, s), 7.62-7.71 (1H, m), 7.84-7.94 (3H, m), 8.13-8.19 (2H, m), 8.43 (1H, dd, *J* = 7.8, 0.8 Hz);

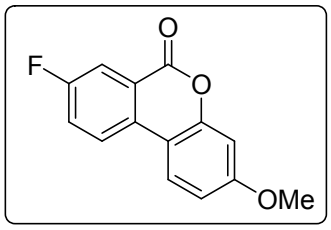
^{13}C NMR ($CDCl_3$, 50 MHz) δ: 26.9, 118.1, 122.0, 122.2, 122.6, 123.4, 124.1, 130.3, 131.0, 133.8, 135.3, 138.6, 151.3, 160.8, 196.7. The spectral data are in well agreement with the literature reported data.[13]

3-Chloro-6*H*-benzo[*c*]chromen-6-one (5j):



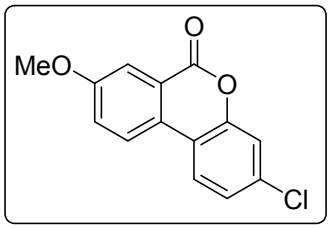
According to the ***GP-3*** with 4′-chloro-biphenyl-2-carbaldehyde (4j) afforded 3-chloro-6*H*-benzo[*c*]chromen-6-one (5j) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.28-7.37 (2H, m), 7.55-7.63 (1H, m), 7.83 (1H, ddd, *J* = 8.6, 7.4, 1.4 Hz), 7.95-8.08 (2H, m), 8.38 (1H, dd, *J* = 8.0, 1.2 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 116.9, 118.2, 121.1, 121.9, 124.0, 125.2, 129.4, 130.9, 134.2, 135.3, 136.1, 151.7, 160.7. The spectral data are in well agreement with the literature reported data.[13]

8-Fluoro-3-methoxy-6*H*-benzo[*c*]chromen-6-one (5k):



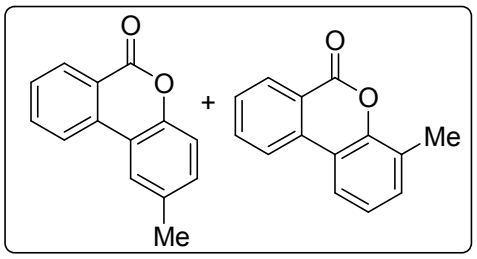
According to the ***GP-3*** with 4-fluoro-4′-methoxy-biphenyl-2-carbaldehyde (4k) afforded 8-fluoro-3-methoxy-6*H*-benzo[*c*]chromen-6-one (5k) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.88 (3H, s), 6.85-6.95 (2H, m), 7.45-7.55 (1H, m), 7.88 (1H, d, *J* = 8.8 Hz), 7.96-8.03 (2H, m); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.9, 101.9, 110.7, 112.9, 116.2 (d, *J* = 23.0 Hz), 121.8 (d, *J* = 8.0 Hz), 123.3 (d, *J* = 23.0 Hz), 123.7 (d, *J* = 6.5 Hz), 123.8, 131.9, 152.3, 161.2 (C, *J* = 43.5 Hz), 161.7, 161.9 (CF, d, *J* = 248.0 Hz). **HRMS** (ESI) calculated for $C_{14}H_{10}FO_3$ $[M + H]^+$: 245.0608; found: 245.0613.

3-Chloro-8-methoxy-6*H*-benzo[*c*]chromen-6-one (5l):



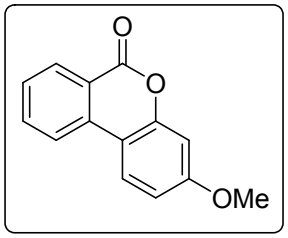
According to the ***GP-3*** with 4′-chloro-4-methoxy-biphenyl-2-carbaldehyde (4l) afforded 3-chloro-8-methoxy-6*H*-benzo[*c*]chromen-6-one (5l) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.92 (3H, s), 7.24-7.33 (2H, m), 7.37 (1H, dd, *J* = 9.0, 2.8 Hz), 7.74 (1H, d, *J* = 2.8 Hz), 7.85 (1H, d, *J* = 8.6 Hz), 7.93 (1H, d, *J* = 8.8 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 56.0, 111.6, 117.0, 117.9, 122.3, 123.3, 123.6, 124.6, 125.2, 127.5, 134.8, 150.8, 160.4, 160.8. The spectral data are in well agreement with the literature reported data.[14]

2-Methyl-6*H*-benzo[*c*]chromen-6-one and 4-methyl-6*H*-benzo[*c*]chromen-6-one in 3:2 mixture (5n):



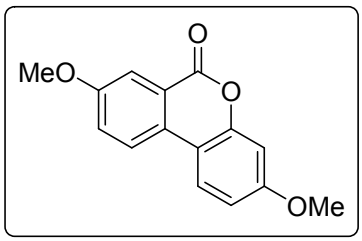
According to the ***GP-3*** with 3′-Methyl-biphenyl-2-carbaldehyde (4n) afforded a mixture of 2-methyl-6*H*-benzo[*c*]chromen-6-one and 4-methyl-6*H*-benzo[*c*]chromen-6-one in 3:2 mixture (5n) as a white solid, **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.45 (1.5), 2.53 (1.0), 7.26-7.38 (2.48), 7.57-7.64 (1.45), 7.81-7.95 (2.0), 8.30 (1H, d, *J* = 8.2 Hz), 8.41-8.46 (1.0); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 21.1, 117.5, 117.6, 117.7, 120.4, 121.1, 121.3, 121.6, 121.9, 122.7, 124.0, 127.1, 128.7, 130.5, 130.6, 131.3, 131.8, 134.1, 134.7, 134.8, 135.1, 149.4, 149.7, 161.2, 161.4. **HRMS** (ESI) calculated for $C_{14}H_{11}O_2$ $[M + H]^+$: 211.0754; found: 211.0748.

3-Methoxy-6*H*-benzo[*c*]chromen-6-one (5p):



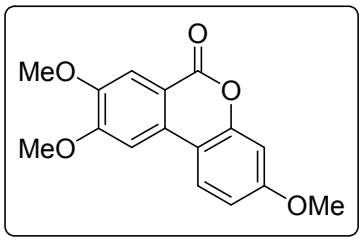
According to the ***GP-3*** with 4′-Methoxy-biphenyl-2-carbaldehyde (4p) afforded 3-methoxy-6*H*-benzo[*c*]chromen-6-one (5p) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.88 (3H, s), 6.86-6.94 (2H, m), 7.46-7.54 (1H, m), 7.73-7.82 (1H, m), 7.92-8.02 (2H, m), 8.35 (1H, d, *J* = 8.0 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.9, 101.9, 112.4, 112.7, 120.2, 121.3, 124.0, 127.9, 130.8, 135.1, 135.4, 152.9, 161.7 (2 x C). The spectral data are in well agreement with the literature reported data.[12]

3,8-Dimethoxy-6*H*-benzo[*c*]chromen-6-one (5q):



According to the ***GP-3*** with 4,4′-Dimethoxy-biphenyl-2-carbaldehyde (4q) afforded 3,8-dimethoxy-6*H*-benzo[*c*]chromen-6-one (5q) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.87 (3H, s), 3.92 (3H, s), 6.82-6.94 (2H, m), 7.35 (1H, dd, *J* = 8.8, 2.8 Hz), 7.74-7.93 (3H, m); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.8, 55.9, 101.8, 111.2, 111.5, 112.6, 121.2, 123.0, 123.3, 124.6, 128.8, 151.8, 159.4, 160.9, 161.8. The spectral data are in well agreement with the literature reported data.[15]

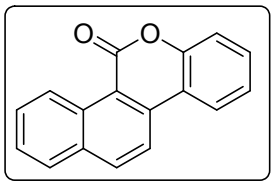
3,8,9-Trimethoxy-6*H*-benzo[*c*]chromen-6-one (5r):



According to the ***GP-3*** with 4,4′,5-trimethoxy-biphenyl-2-carbaldehyde (4r) afforded 3,8,9-trimethoxy-6*H*-benzo[*c*]chromen-6-one (5r) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 3.86 (3H, s), 3.97 (3H, s), 4.01 (3H, s), 6.81-6.91 (2H, m), 7.29 (1H, s), 7.67 (1H, s), 7.80 (1H, d, *J* = 8.8 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 55.8, 56.4 (2 x CH_3), 101.7, 102.3, 110.6, 111.4, 112.5, 113.2, 123.3, 130.6, 149.5,

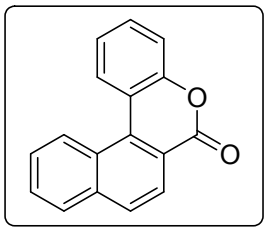
152.4, 155.4, 161.0, 161.6. **HRMS** (ESI) calculated for $C_{16}H_{15}O_5$ $[M + H]^+$: 287.0914; found: 287.0822.

5*H*-naphtho[1,2-*c*]chromen-5-one (5s):



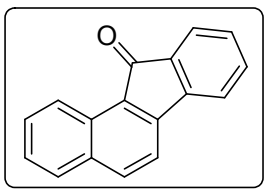
According to the ***GP-3*** with 2-phenyl-1-naphthaldehyde (4s) afforded 5*H*-naphtho[1,2-*c*]chromen-5-one (5s) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.35-7.69 (4H, m), 7.74-7.83 (1H, m), 7.92-7.94 (1H, dd, *J* = 7.8 Hz, 1.2 Hz), 8.17-8.29 (3H, m), 9.82 (1H, dt, *J* = 8.8 Hz, 0.6 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 115.5, 117.5, 118.4, 119.0, 123.7, 124.5, 127.5, 127.6, 128.9, 129.9, 131.1, 132.3, 133.5, 136.7, 136.9, 151.9, 160.7. The spectral data are in well agreement with our previously reported data.[10]

6*H*-naphtho[2,1-*c*]chromen-6-one (5t):



According to the ***GP-3*** with 1-phenyl-2-naphthaldehyde (4t) afforded 6*H*-naphtho[2,1-*c*]chromen-6-one (5t) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.37-7.56 (3H, m), 7.70-7.75 (2H, m), 7.95-8.04 (2H, m), 8.34 (1H, d, *J* = 8.3 Hz), 8.54 (1H, d, *J* = 8.3 Hz), 8.87-8.92 (1H, m); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 118.2, 119.1, 120.4, 124.4,124.7 127.4, 127.7, 128.1, 128.6, 129.2, 129.4, 129.9, 130.3, 134.6, 137.4, 151.9, 161.9. The spectral data are in well agreement with our previously reported data.[10]

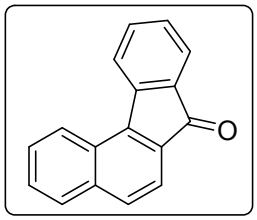
11*H*-benzo[*a*]fluoren-11-one (6a):



According to the ***GP-3*** with 2-phenyl-1-naphthaldehyde (4s) afforded 11*H*-benzo[*a*]fluoren-11-one (6a) as a yellow solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.26-7.30 (1H, m), 7.39-7.49 (3H, m), 7.54-7.65 (3H, m), 7.78 (1H, d, *J* = 8.4 Hz), 7.98 (1H, d, *J* = 8.2 Hz), 8.95 (1H, d, *J* = 8.6 Hz); **^{13}C NMR**

($CDCl_3$, 50 MHz) δ: 118.3, 120.1, 124.0, 124.5, 126.6, 127.0, 128.7, 129.4, 129.6, 130.4, 134.4, 134.6, 134.8, 136.1, 144.1, 146.3, 195.6. The spectral data are in well agreement with our previously reported data.[16]

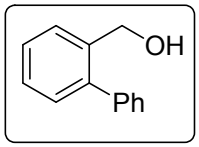
7*H*-benzo[*c*]flouren-7-one (6b):



According to the ***GP-3*** with 1-phenyl-2-naphthaldehyde (4t) afforded 7*H*-benzo[*c*]flouren-7-one (6b) as a yellow solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.31-7.42 (1H, m), 7.46-7.79 (7H, m), 8.04 (1H, d, *J* = 7.6 Hz), 8.49 (1H, d, *J* = 7.0 Hz); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 120.0, 123.6, 124.3, 125.1, 128.0, 128.1, 128.5, 129.0, 129.1, 129.9, 130.1, 132.1, 134.7, 138.3, 143.1, 145.3, 194.7. The spectral data are in well agreement with our previously reported data.[16]

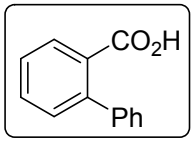
5. Spectroscopic data of compound 7 and 8

2-Phenylbenzylalcohol (7):



According to the ***GP-4*** with biphenyl-2-carbaldehyde(4a) afforded 2-phenylbenzylalcohol(7) as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 2.47 (1H, bs), 4.60 (2H, s), 7.32-7.60 (9H, m); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 62.9, 127.3, 127.6, 127.7, 128.3 (2 x CH), 128.4, 129.2 (2 x CH), 130.1, 138.1, 140.7, 141.2. The spectral data are in well agreement with literature reported data.

Biphenyl-2-carboxylic acid (8):



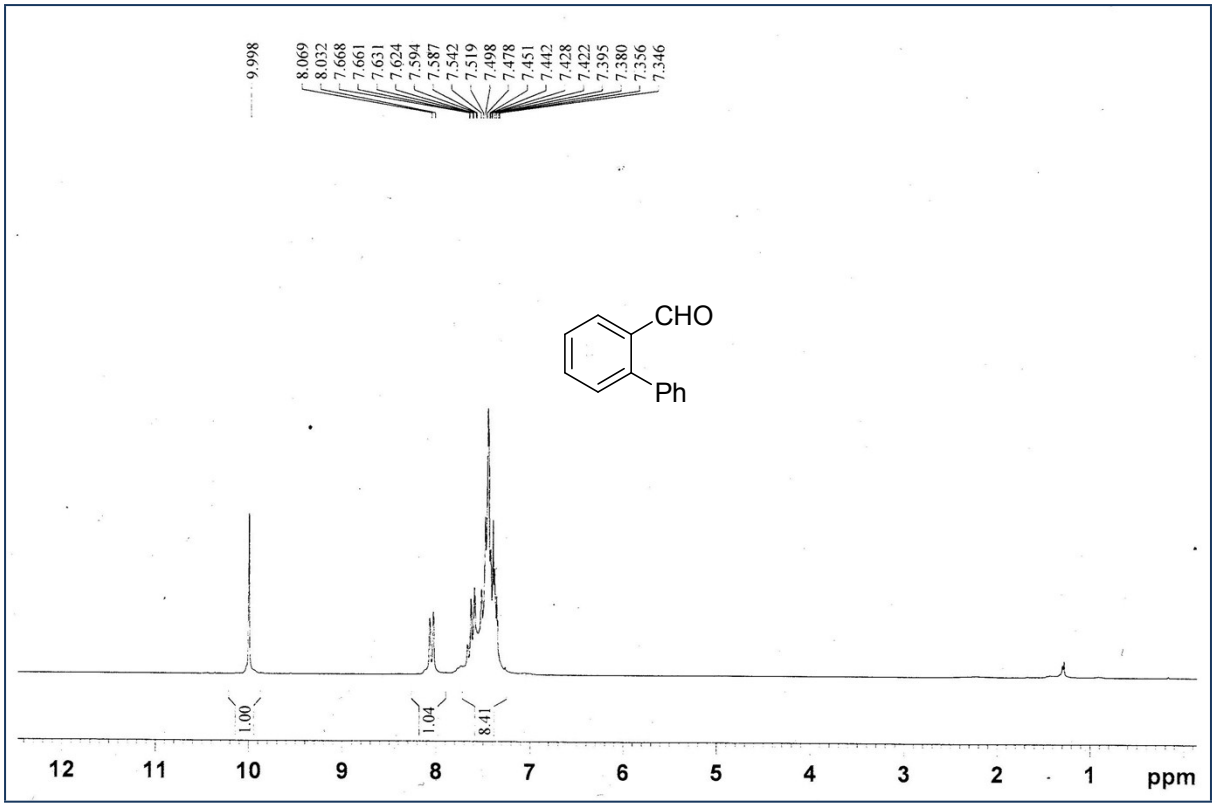
According to the ***GP-5*** with biphenyl-2-carbaldehyde(4a) afforded biphenyl-2-carboxylic acid as a white solid; **^{1}H NMR** ($CDCl_3$, 200 MHz) δ: 7.45-7.60 (8H, m), 8.05-8.09 (1H, m), 10.11 (1H, bs); **^{13}C NMR** ($CDCl_3$, 50 MHz) δ: 127.2,

127.4, 128.1 (2 x CH), 128.5 (2 x CH), 129.5, 130.7, 131.2, 132.1, 141.1, 143.4, 173.8. The spectral data are in well agreement with literature reported data.

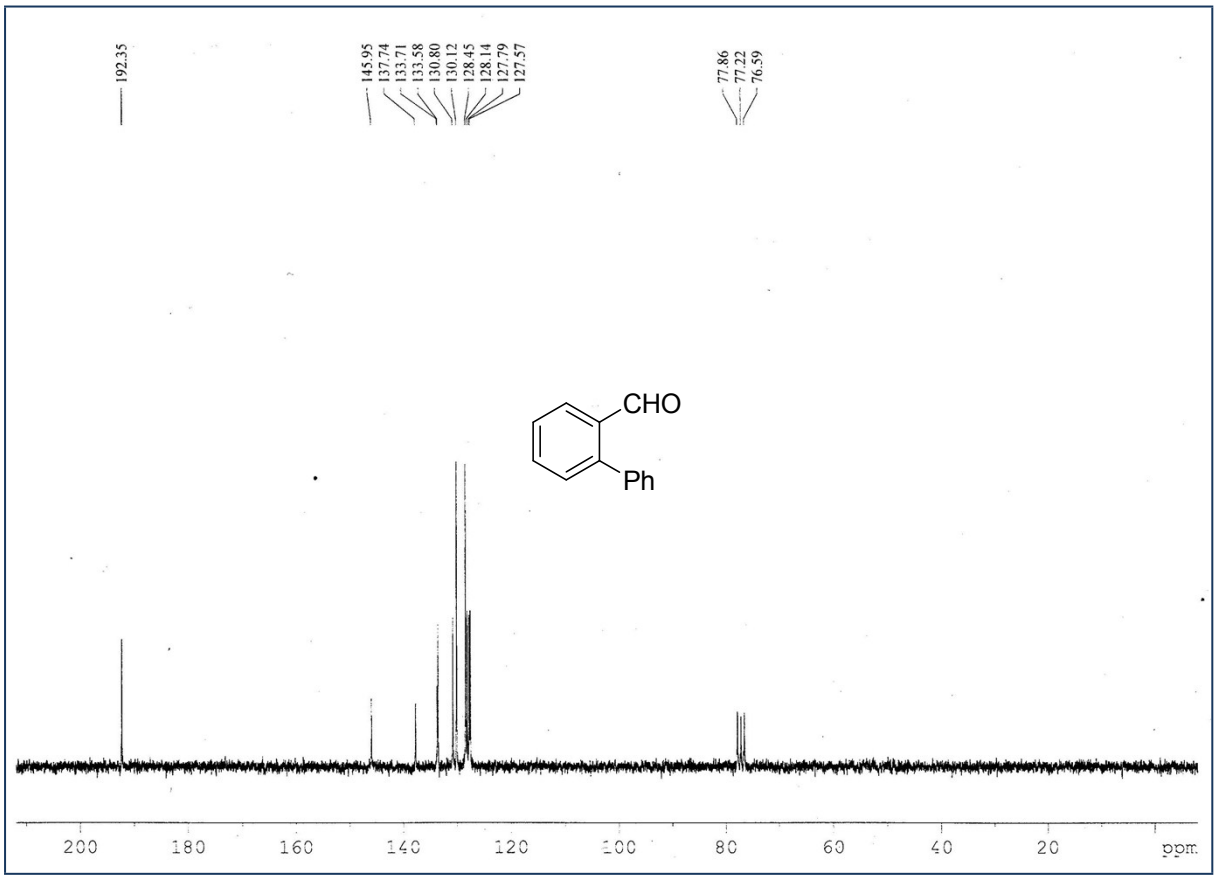
6. References:

1. M. L. Hossain, F. Ye, Z. Liu, Y. Xia, Y. Shi, L. Zhou and Y. Zhang. *J. Org. Chem.*, 2014, **79**, 8689.
2. S. Wertz, D. Leifert and A. Studer, *Org. Lett.* 2013, **15**, 928.
3. J. Zhao, D. Yue, M. A. Campo and R. C. Larock, *J. Am. Chem. Soc*., 2007, **129**, 5288.
4. A. S. Kumar, M. A. Reddy, N. Jain, C. Kishor, T. R. Murthy, D. Ramesh, B. Supriya, A. Addlagatta, S. V. Kalivendi and B. Sreedhar, *Eur. J. Med. Chem*., 2013, **60**, 305.
5. V. Farina, B. Krishna, D. R. Marshall and G. P. Roth, *J. Org. Chem*., 1993, **58**, 5434.
6. T. E. Barder, S. D. Walker, J. R. Martinelli and S. L. Buchwald, *J. Am. Chem. Soc.*, 2005, **127**, 4685.
7. T. A. Chen, T. J. Lee, M. Y. Lin, S. M. A. Sohel, E. W. G. Diau, S. F. Lush and R. S. Liu, *Chem. Eur. J*., 2010, **16**, 1826.
8. T. G. Gant and A. I. Meyers, *J. Am. Chem. Soc.* 1992, **114**, 1010.
9. L. Zhang, G. Y. Ang and S, Chiba, *Org. Lett*., 2010, **12**, 3682.
10. R. Singha, S. Roy, S. Nandi, P. Ray and J. K. Ray, *Tetrahedron Lett.*, 2013, **54**, 657.
11. K. Vishnumurthy and A. Makriyannis, *J. Comb. Chem.*, 2010, **12**, 664.
12. P. Gao and Y. Wei, *Synthesis*, 2014, **46**, 343.
13. Y. Wang, A. V. Gulevich and V. Gevorgyan, *Chem. Eur. J.*, 2013, **19**, 15836.
14. K. Inamoto, J. Kadokawa and Y. Kondo, *Org. Lett.,* 2013, **15**, 3962.
15. P. Nealmongkol, K. Tangdenpaisal, S. Sitthimonchai, S. Ruchirawat and N. Thasana, *Tetrahedron*, 2013, **69**, 9277.
16. S. Paul, S. Samanta, J. K. Ray, *Tetrahedron Lett*., 2010, 51, 5604.

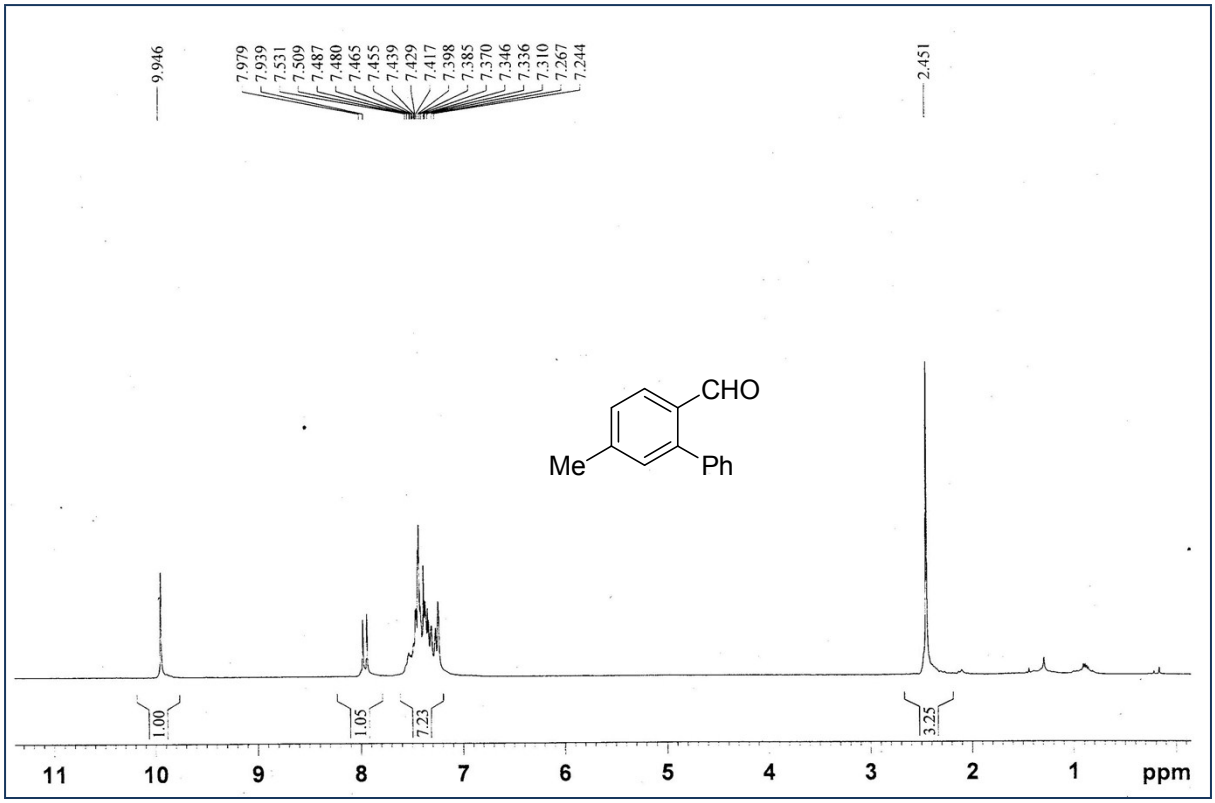
^{1}H NMR of compound 4a



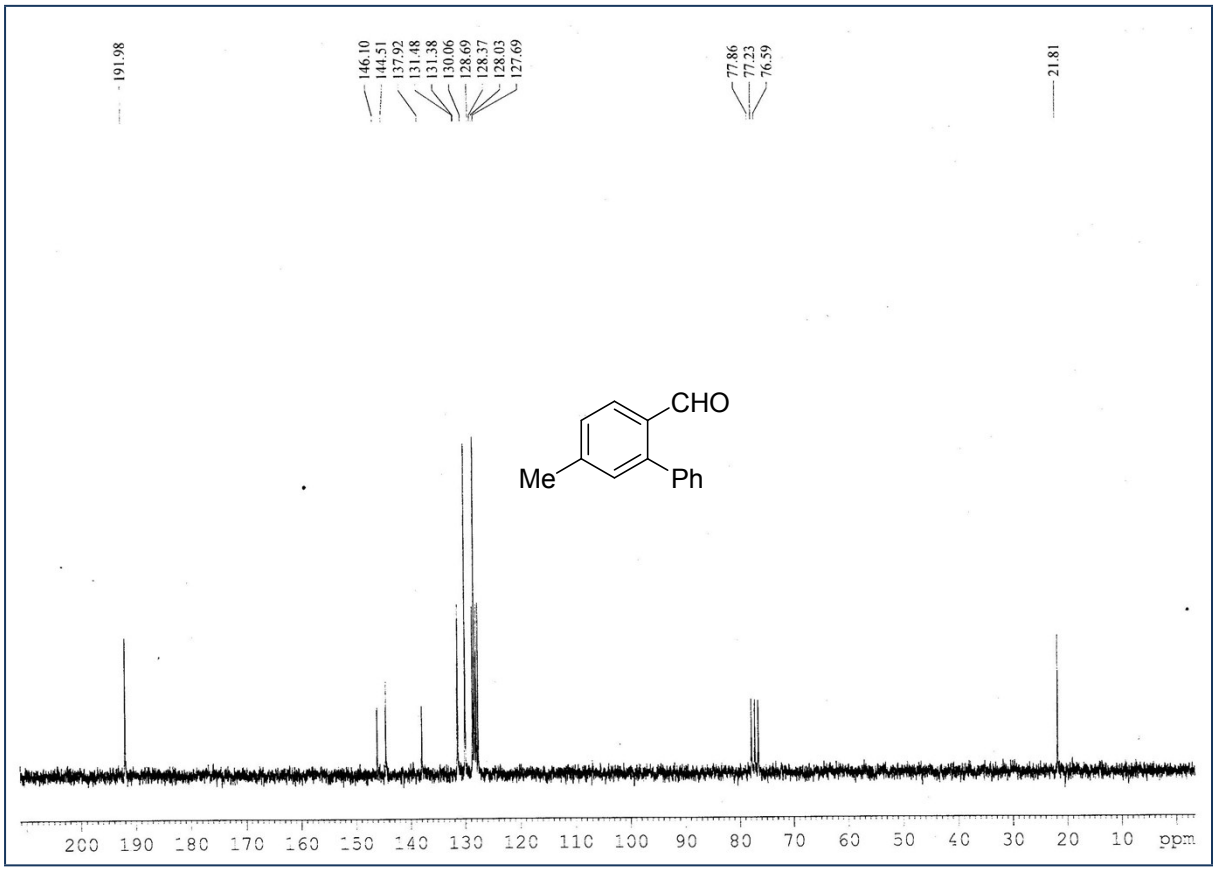
^{13}C NMR of compound 4a



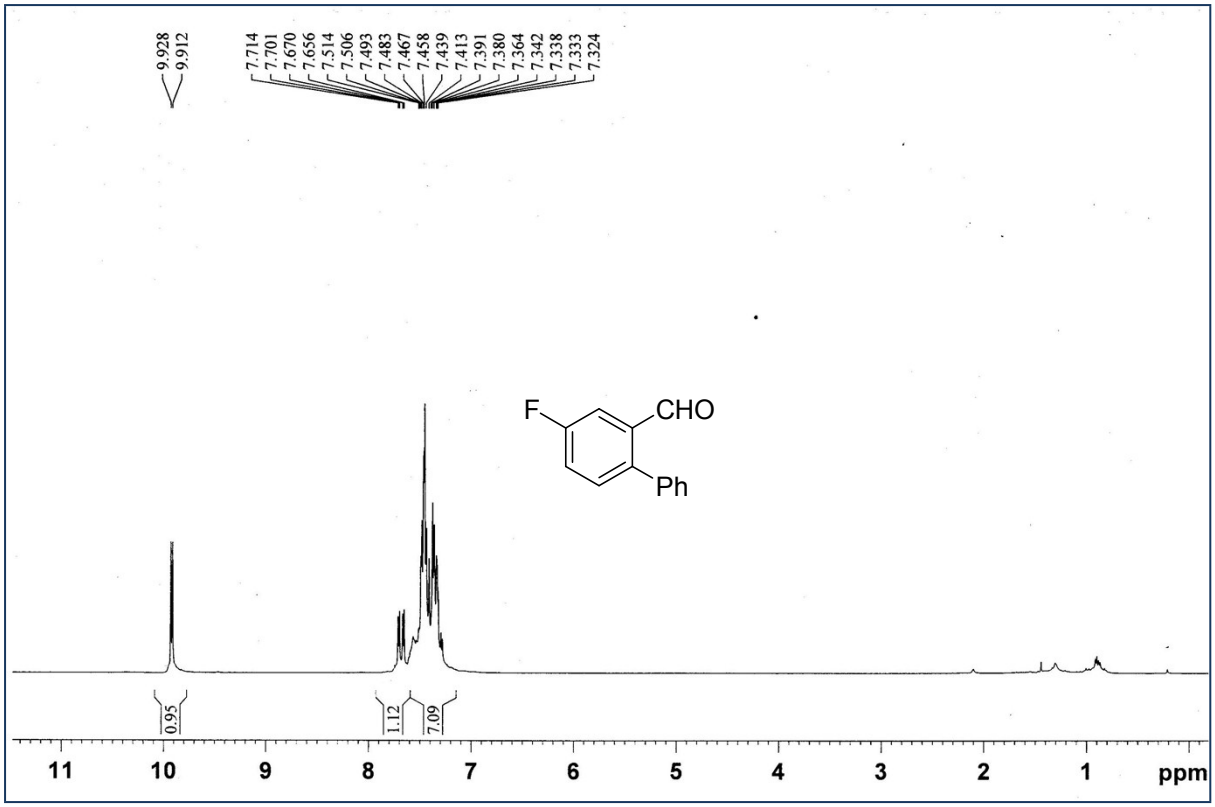
^{1}H NMR of compound of 4b



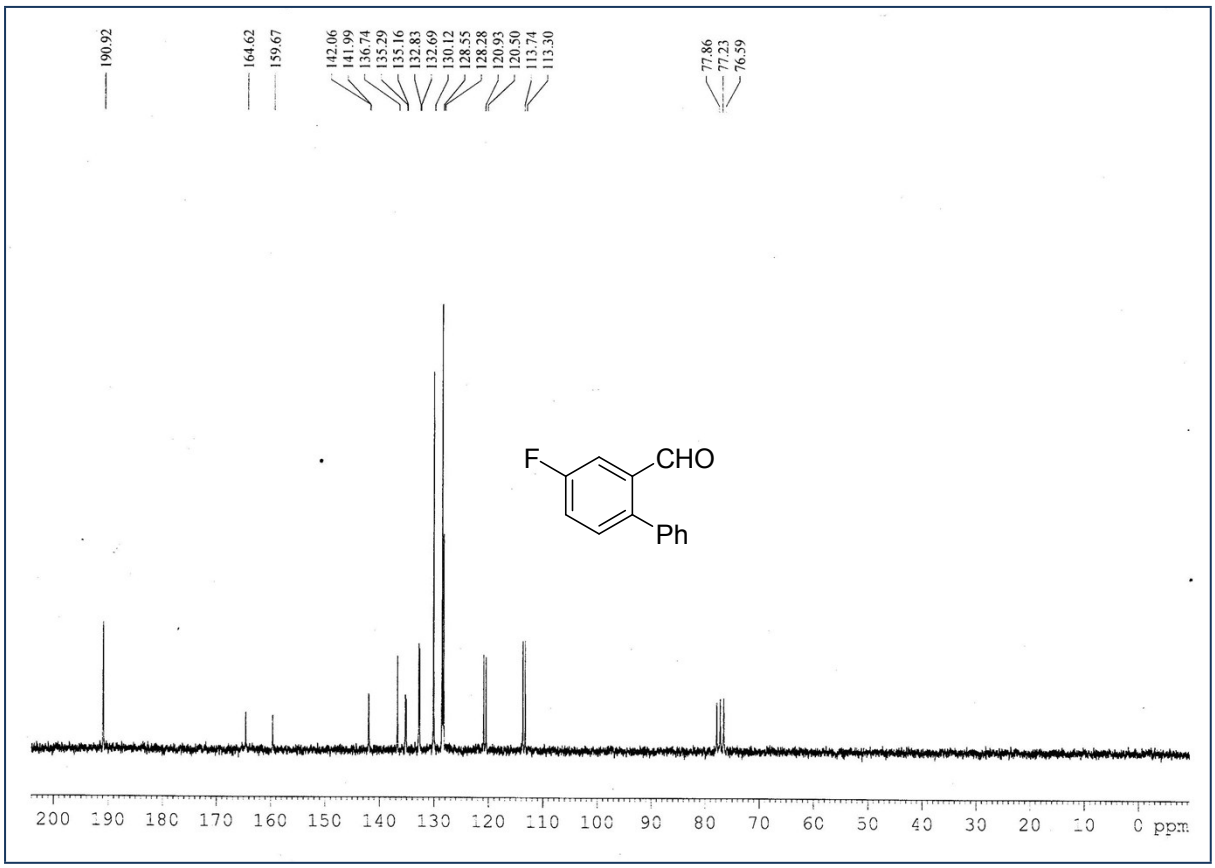
^{13}C NMR of compound 4b



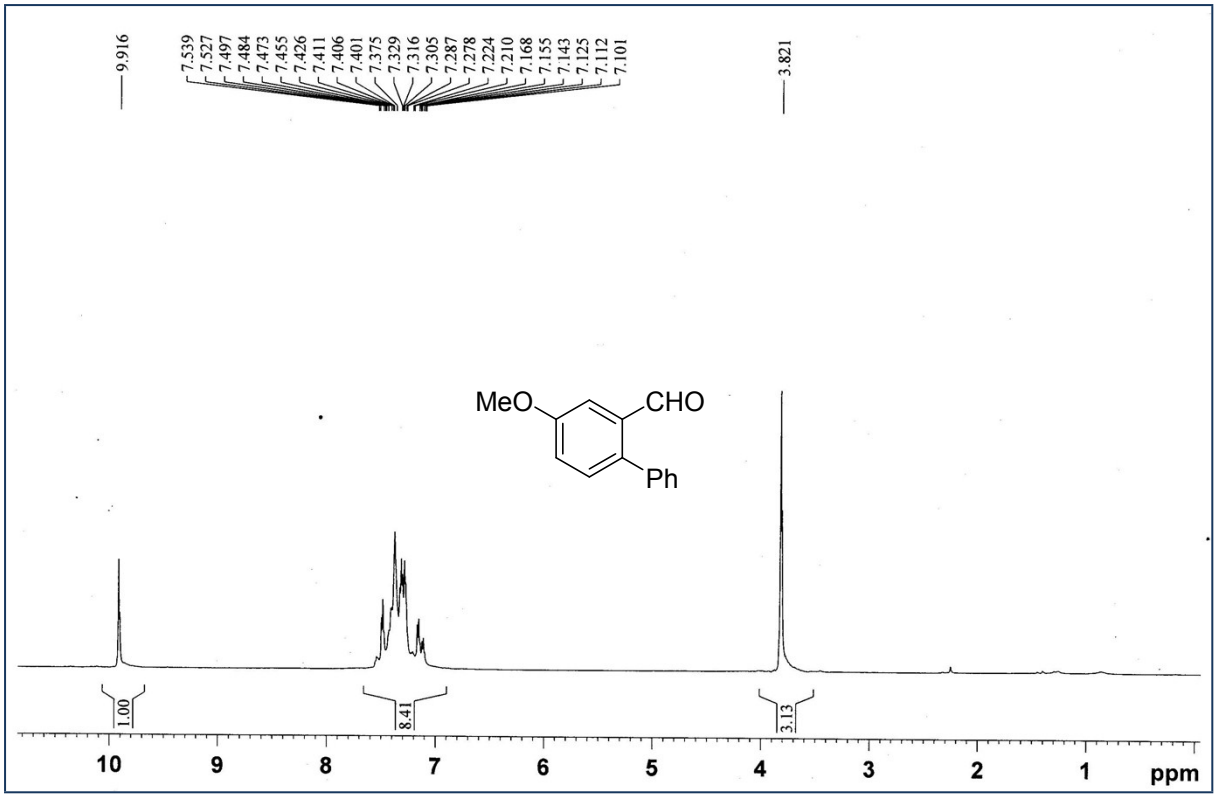
^{1}H NMR of compound 4c



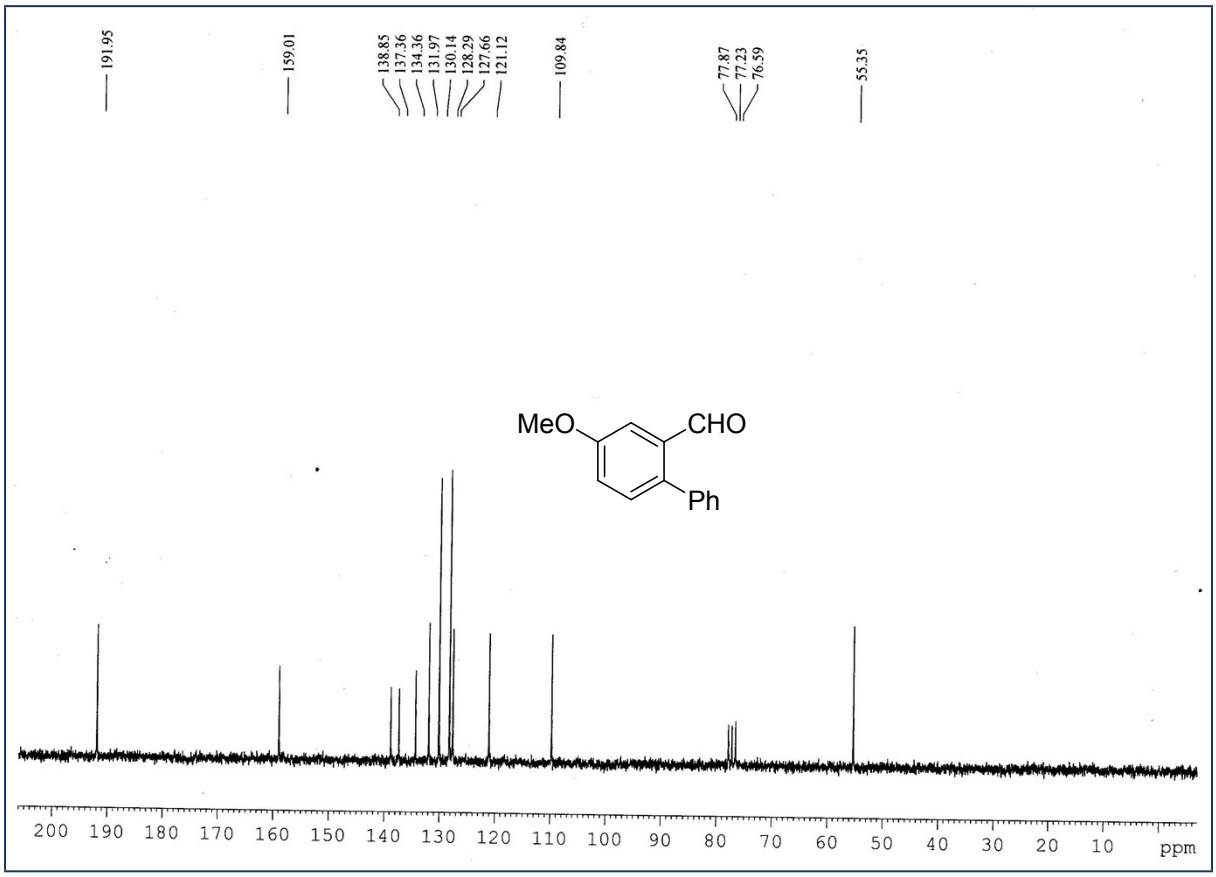
^{13}C NMR of compound 4c



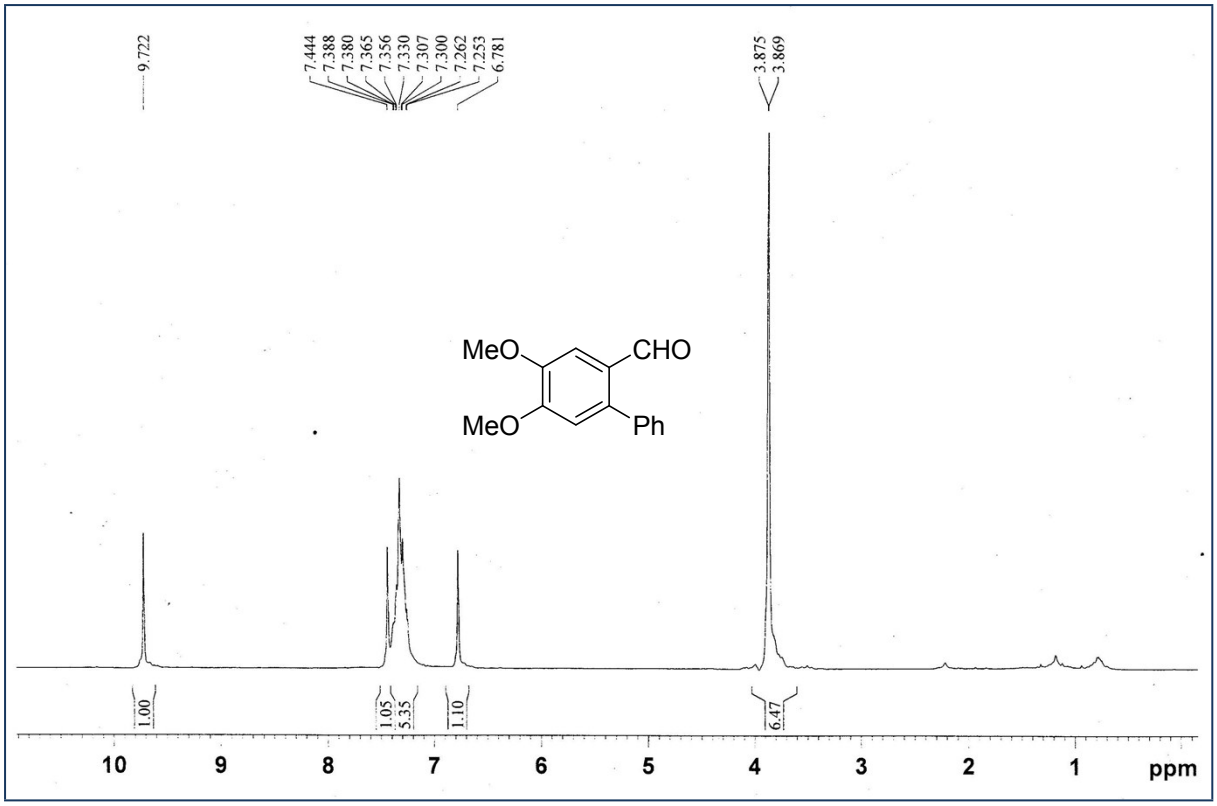
^{1}H NMR of compound 4d



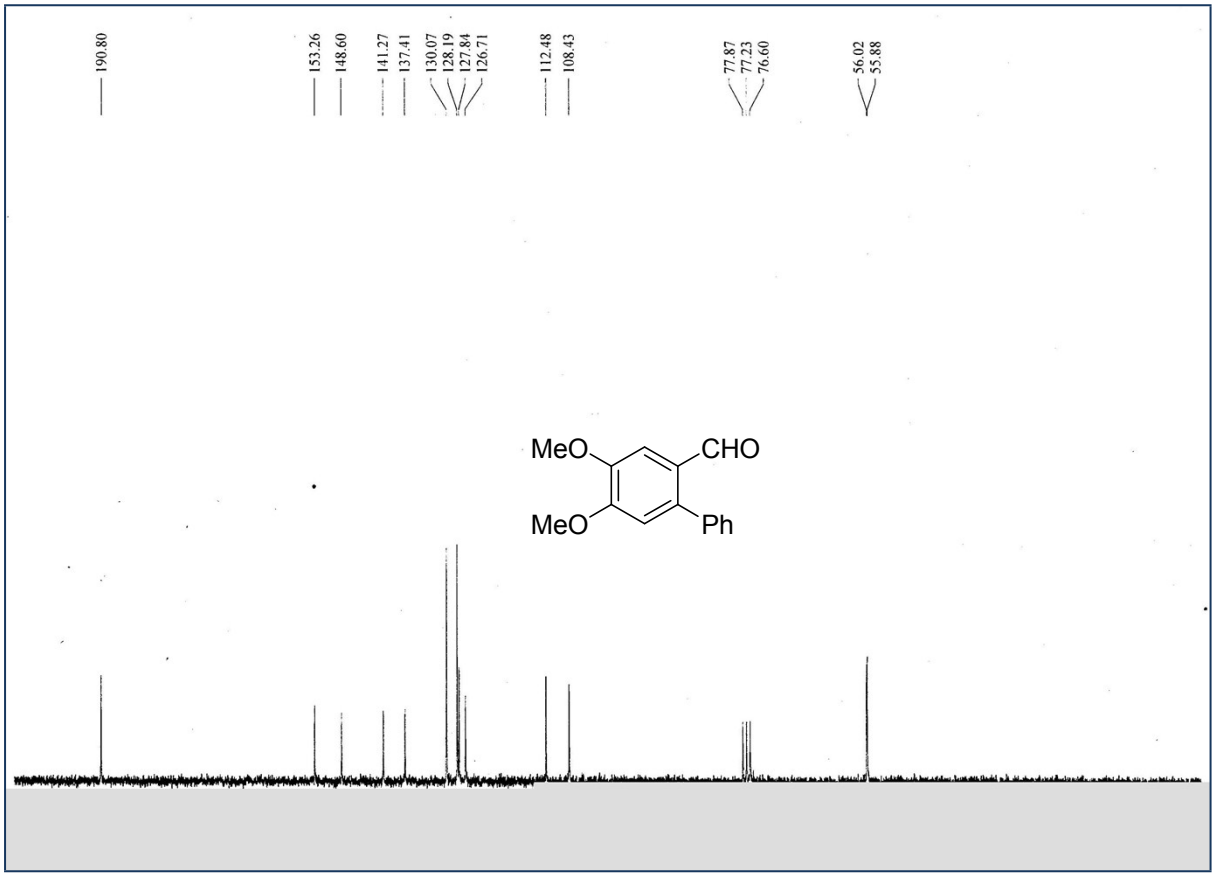
^{13}C NMR of compound 4d



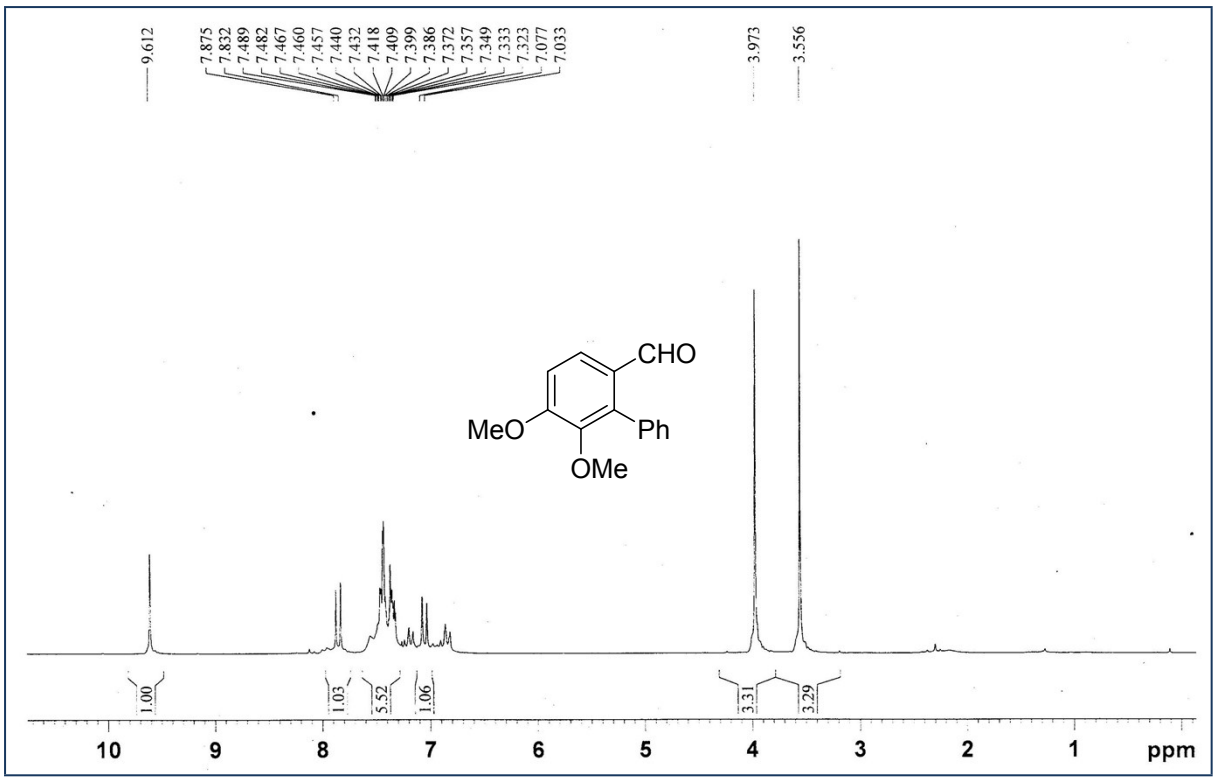
^{1}H NMR of compound 4e



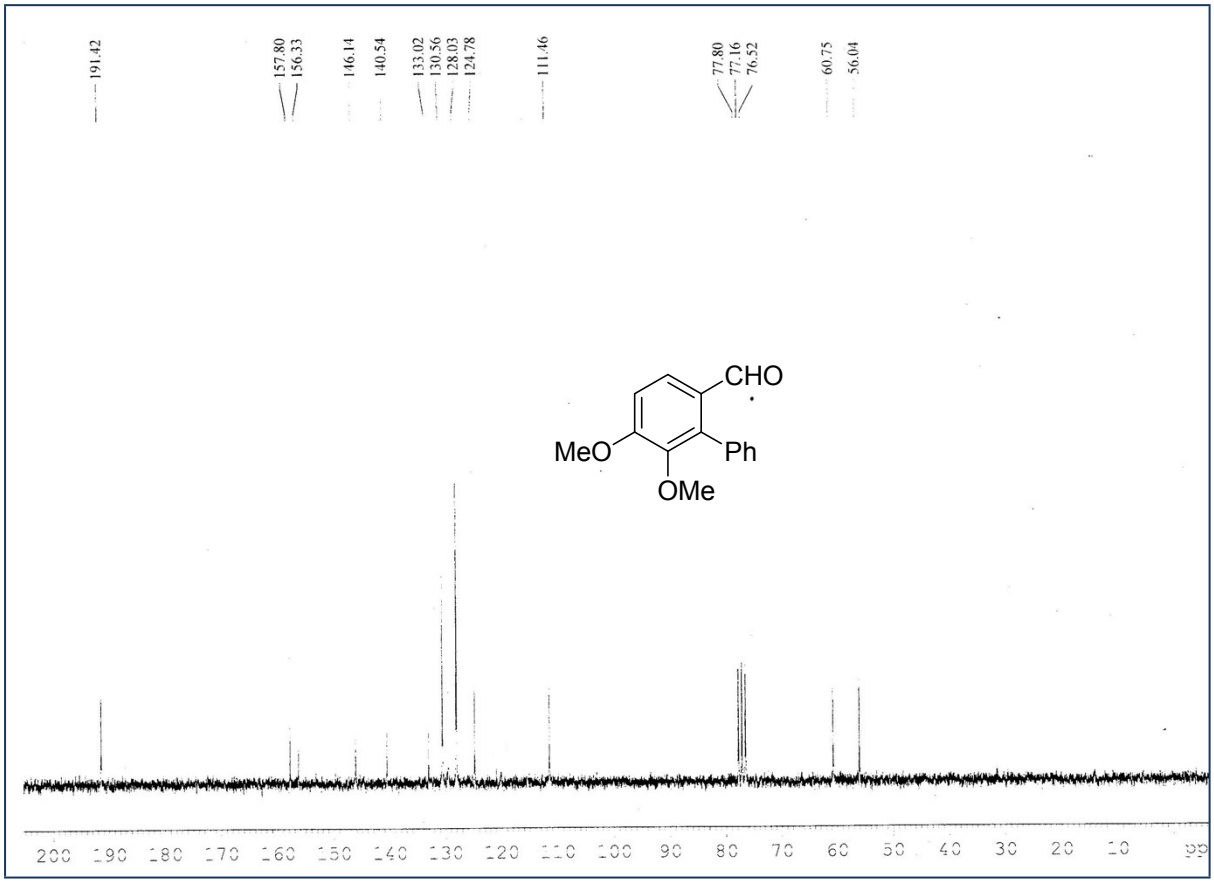
^{13}C NMR of compound 5e



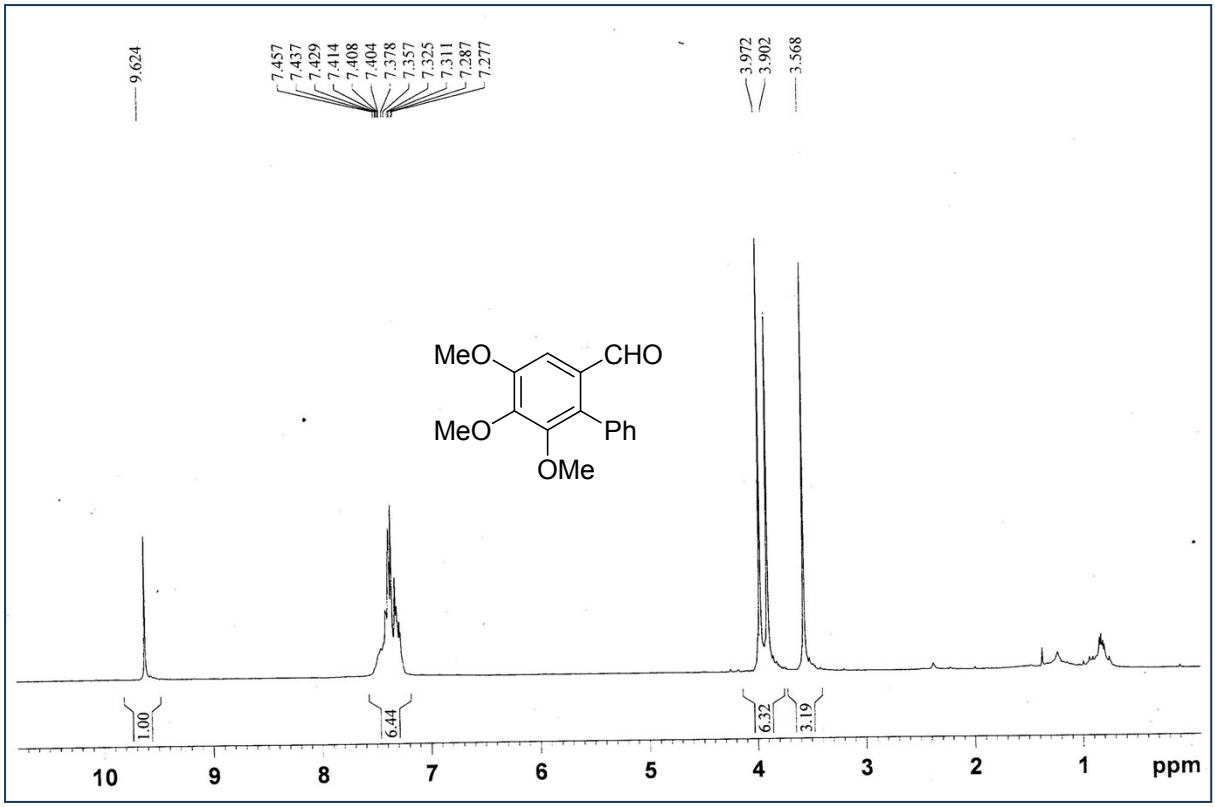
^{1}H NMR of compound 4f



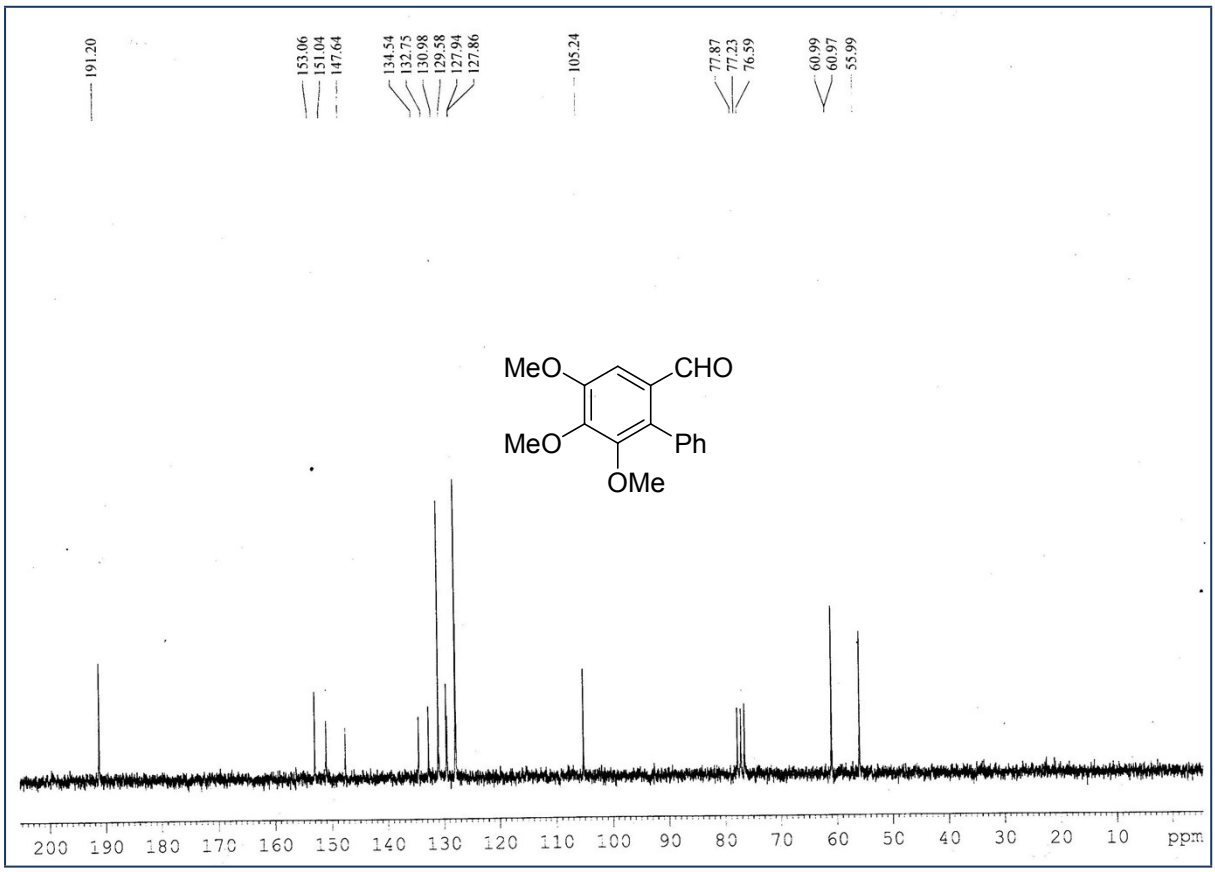
^{13}C NMR of compound 4f



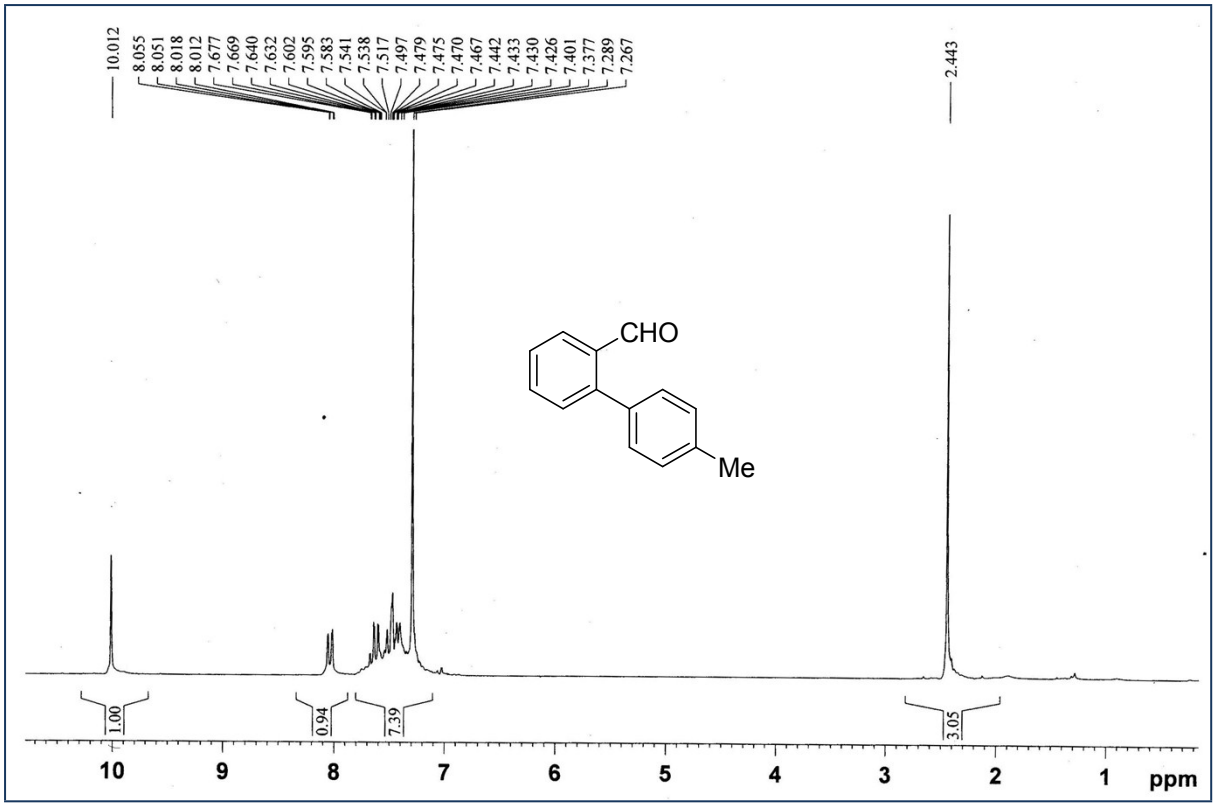
^{1}H NMR of compound 4g



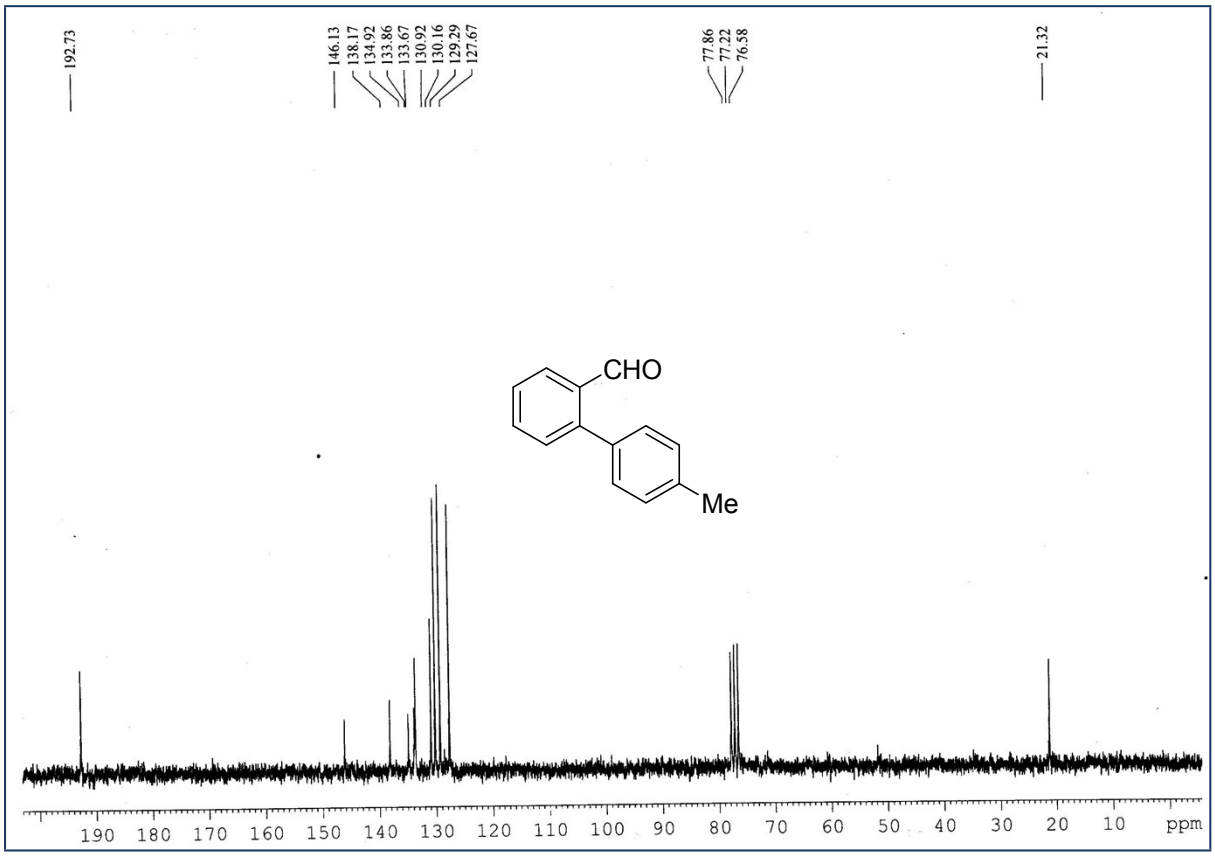
^{13}C NMR of compound 4g



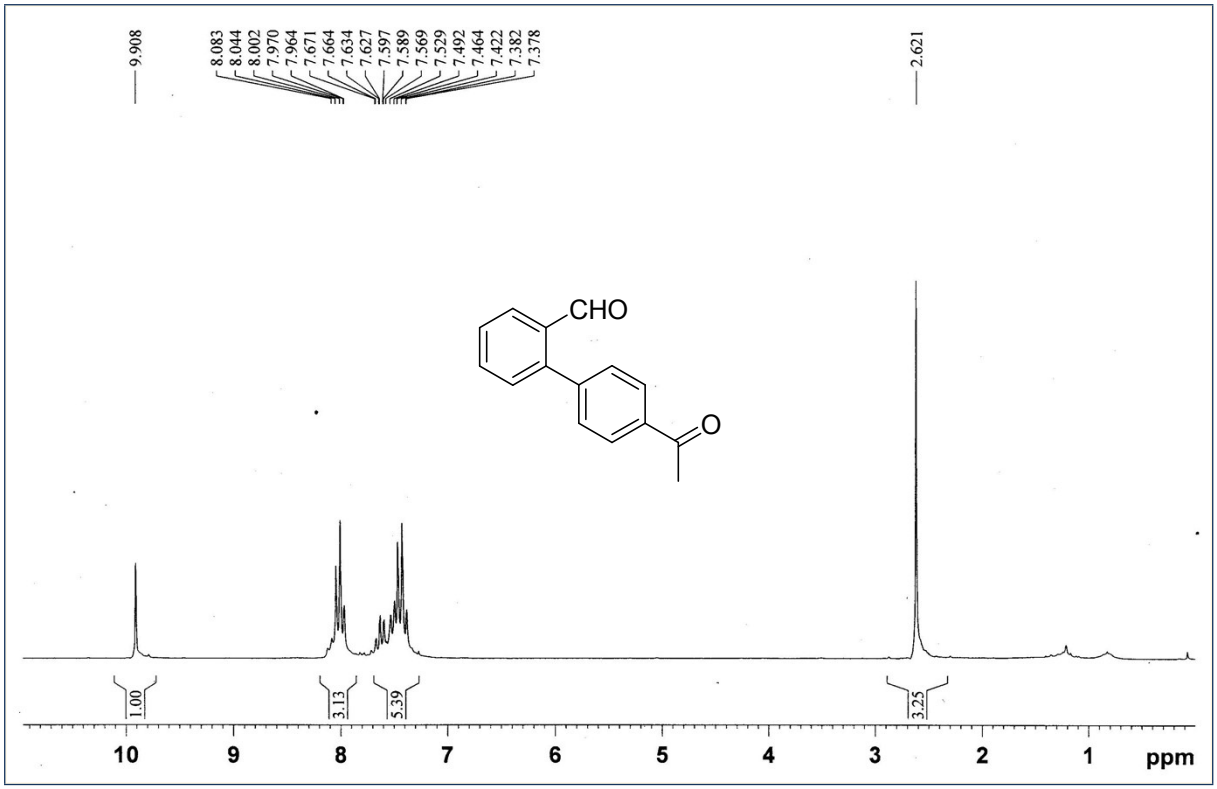
^{1}H NMR of compound 4h



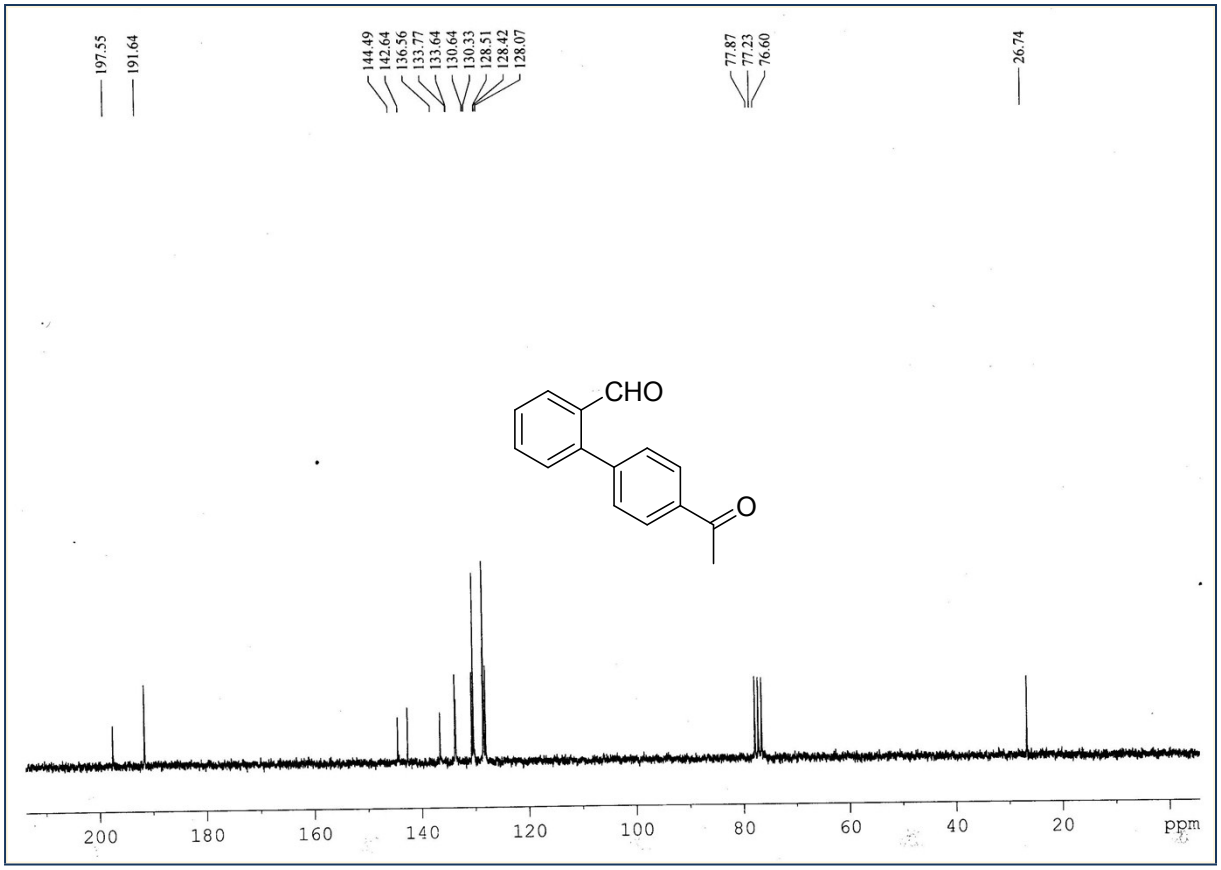
13C NMR of compound 4h



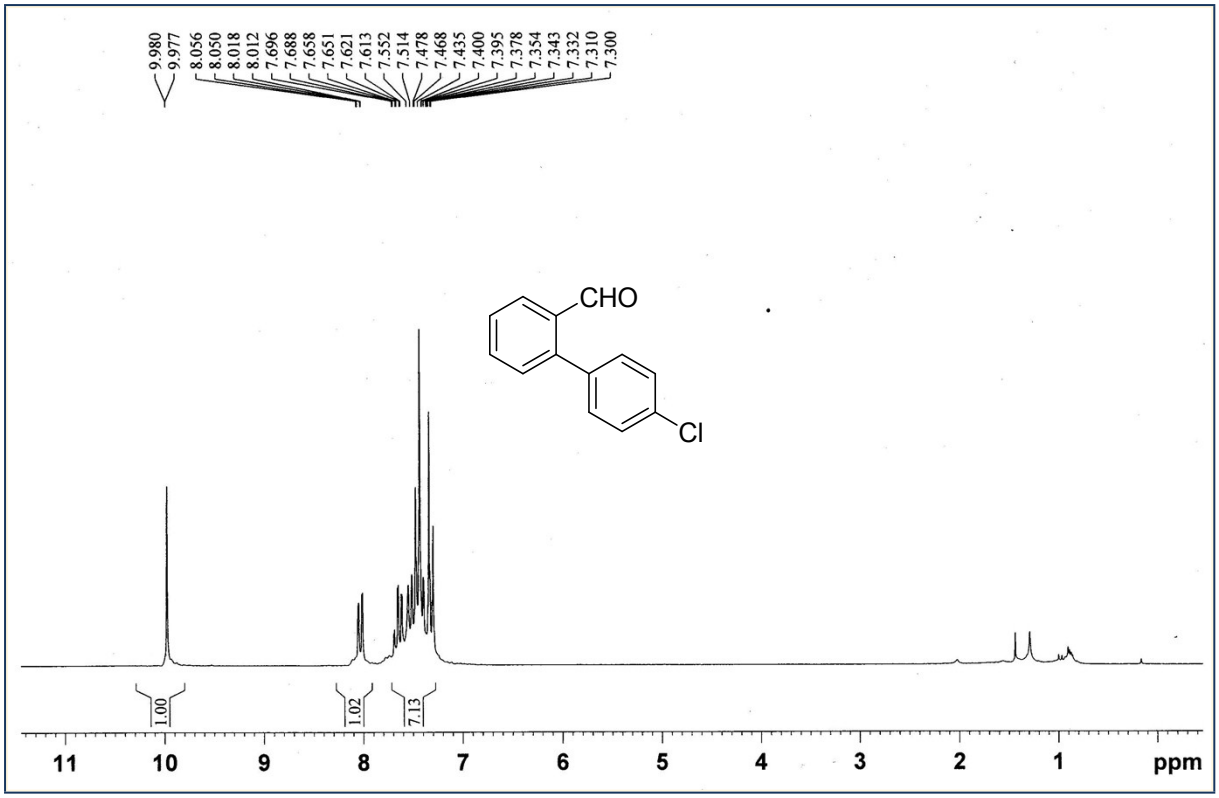
^{1}H NMR of 4i



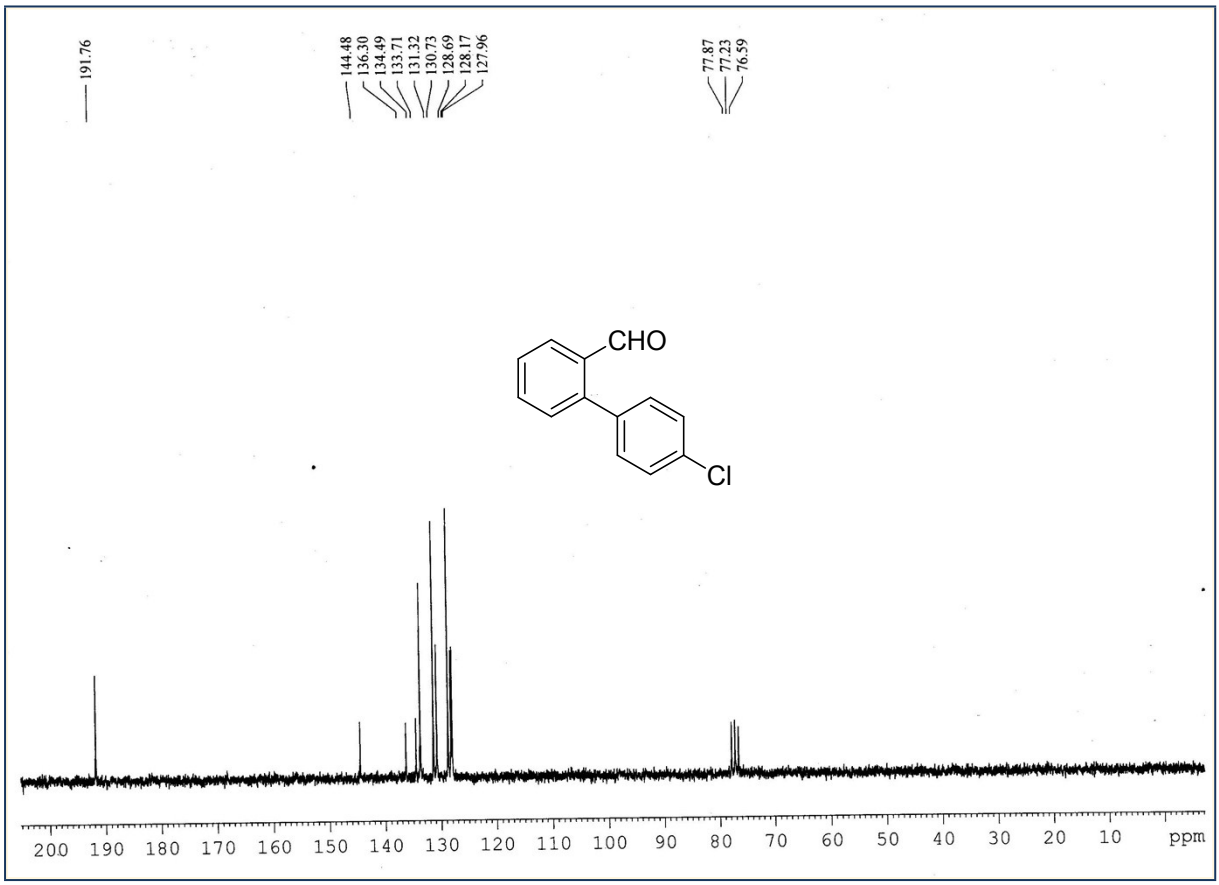
^{13}C NMR of compound 4i



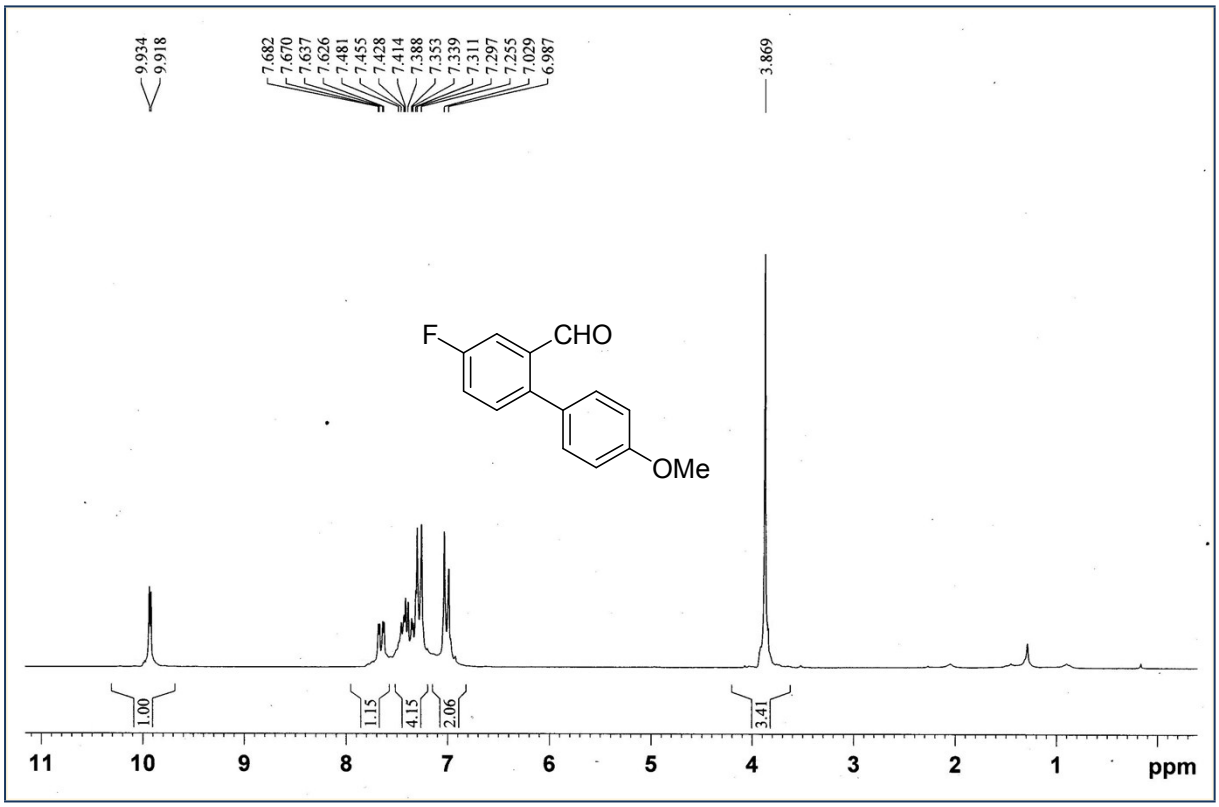
^{1}H NMR of compound 4j



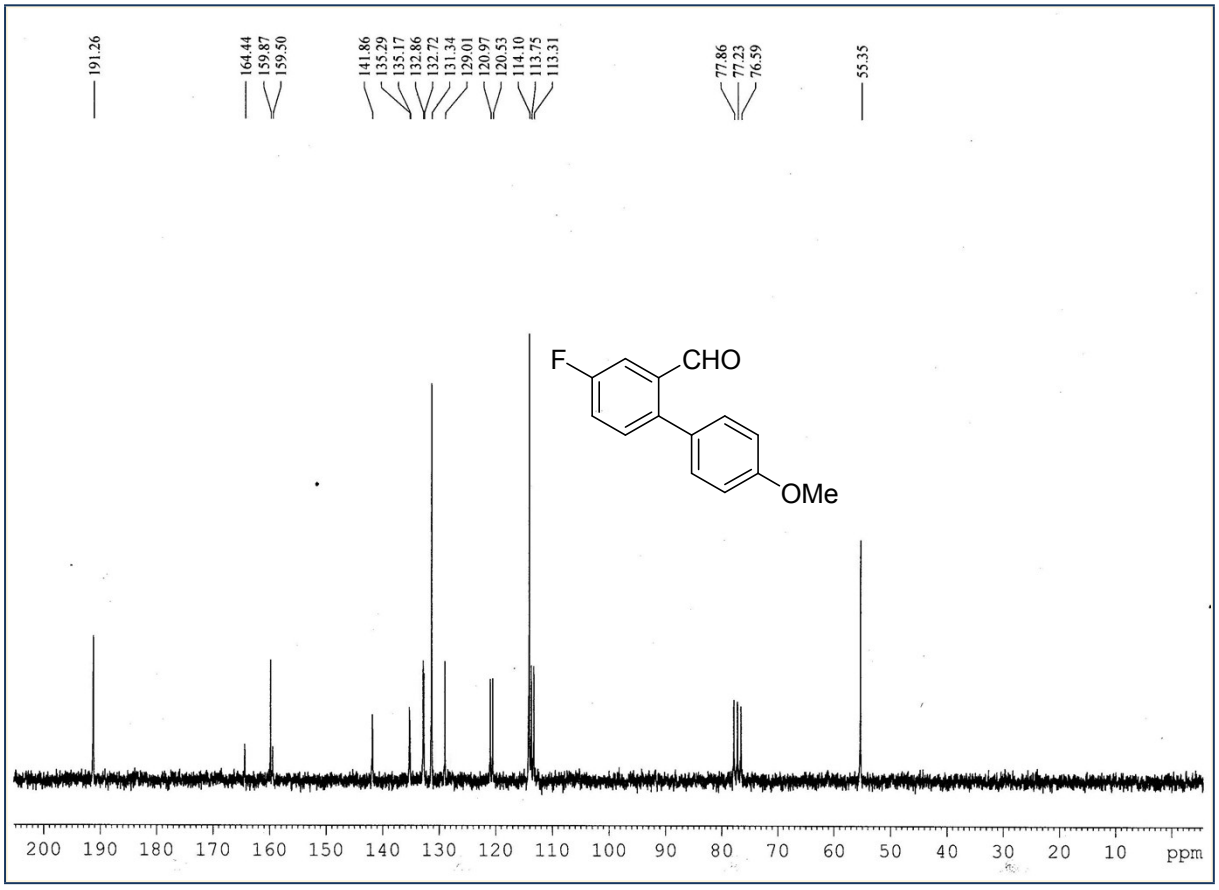
^{13}C NMR of compound 4j



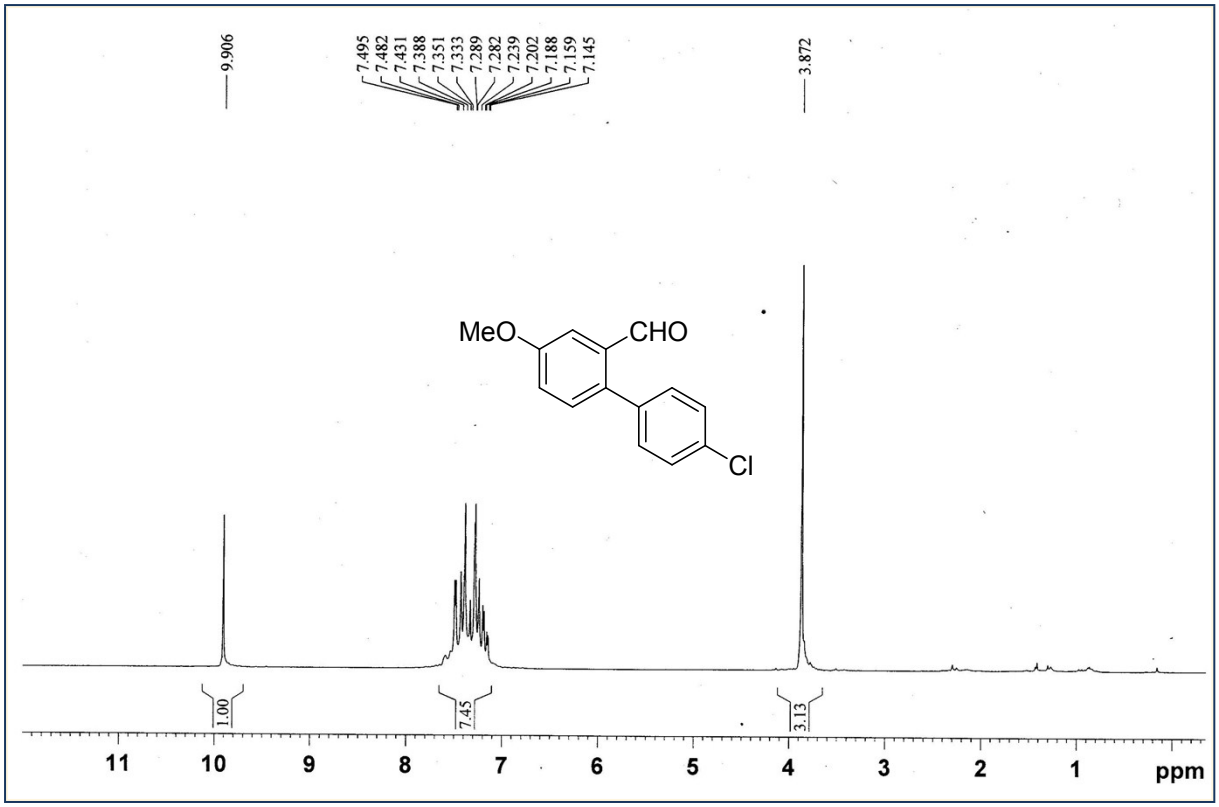
^{1}H NMR of compound 4k



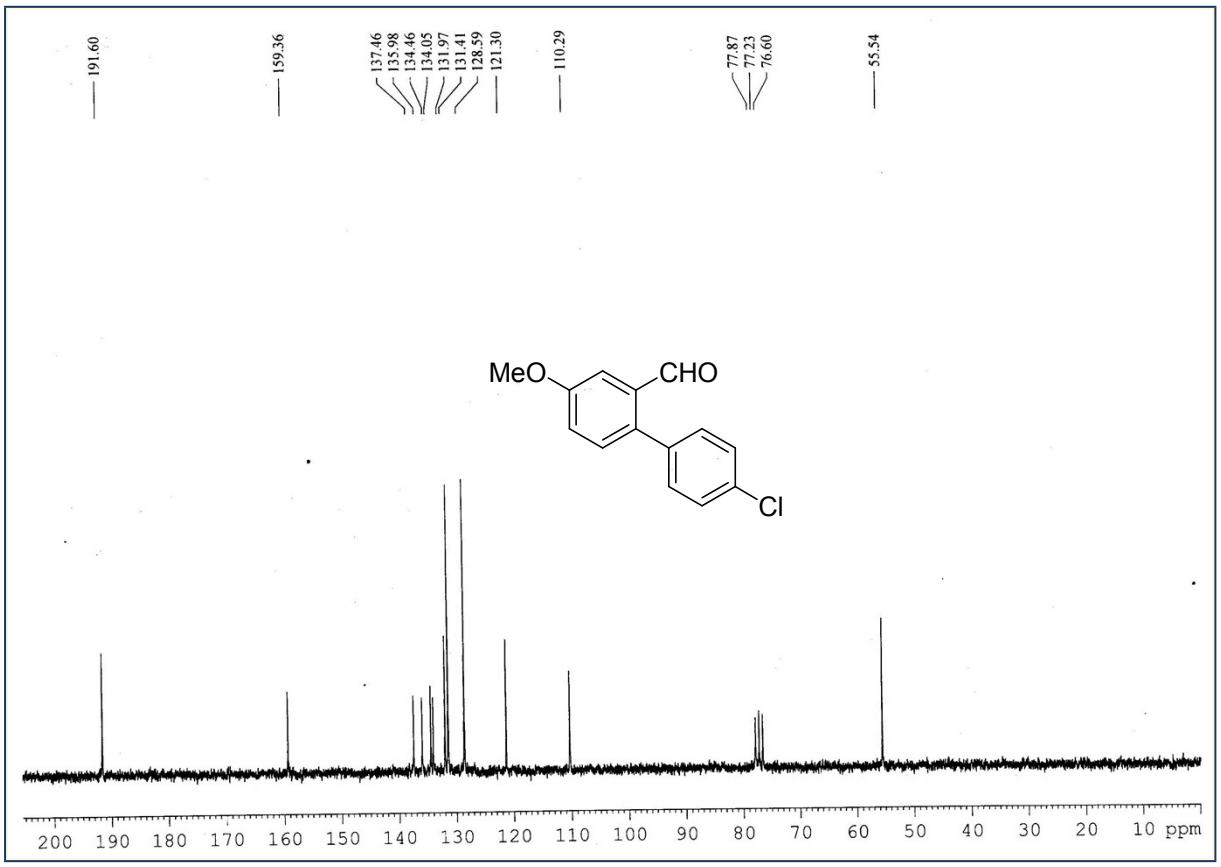
^{13}C NMR of compound 4k



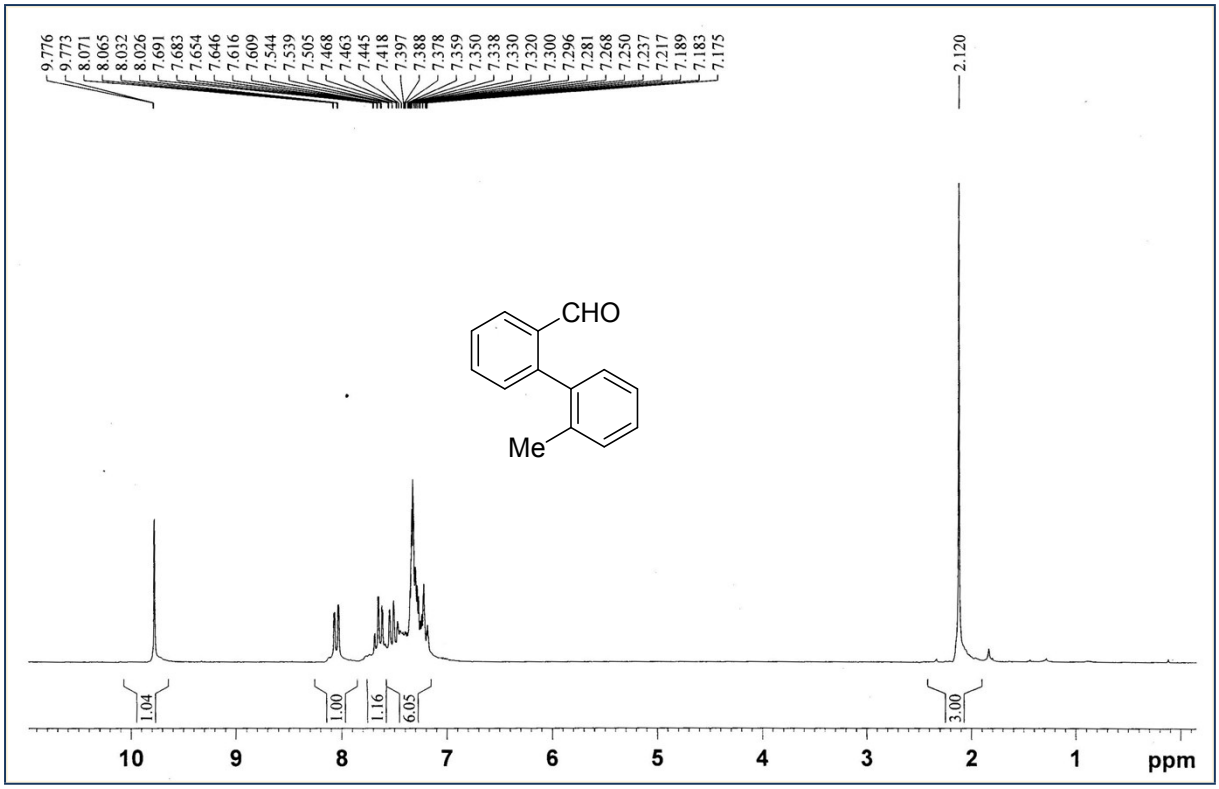
^{1}H NMR of compound 4l



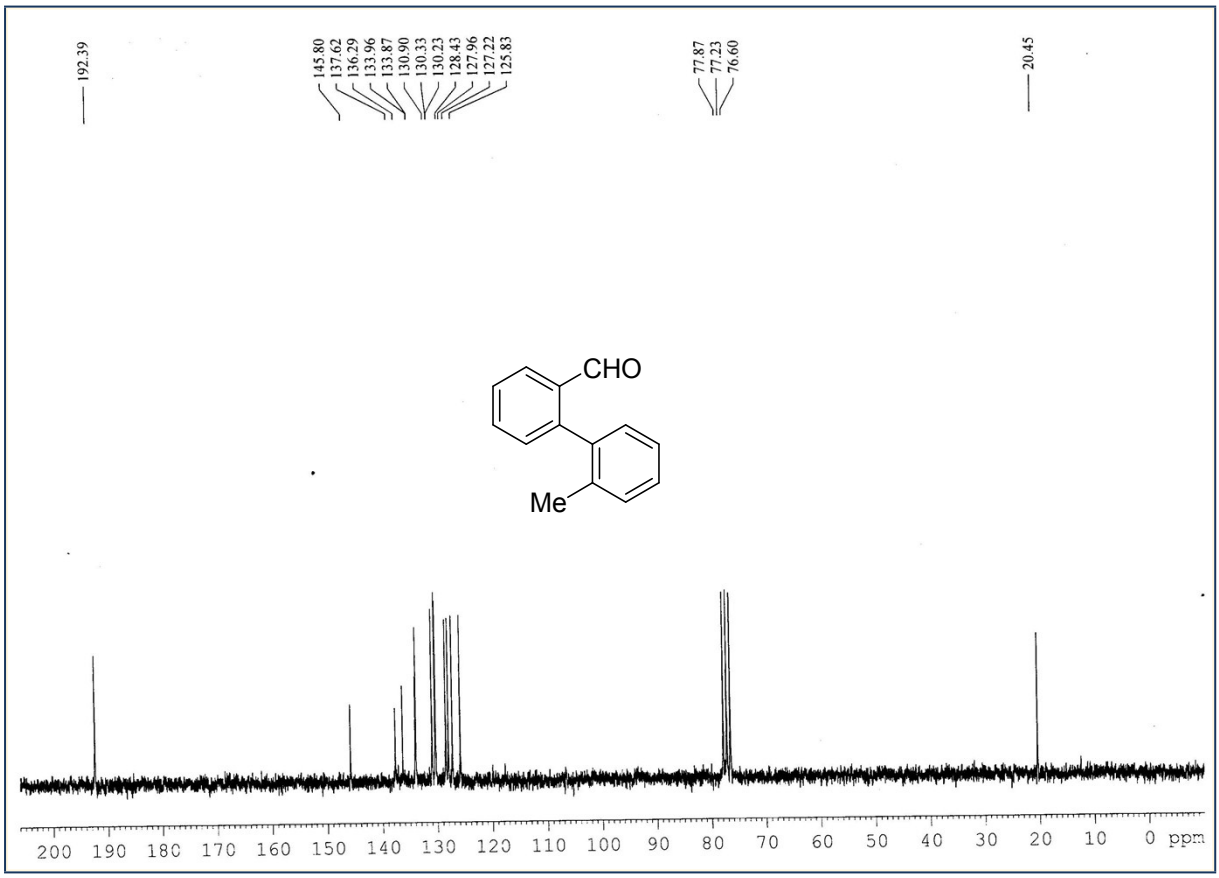
^{13}C NMR of compound 4l



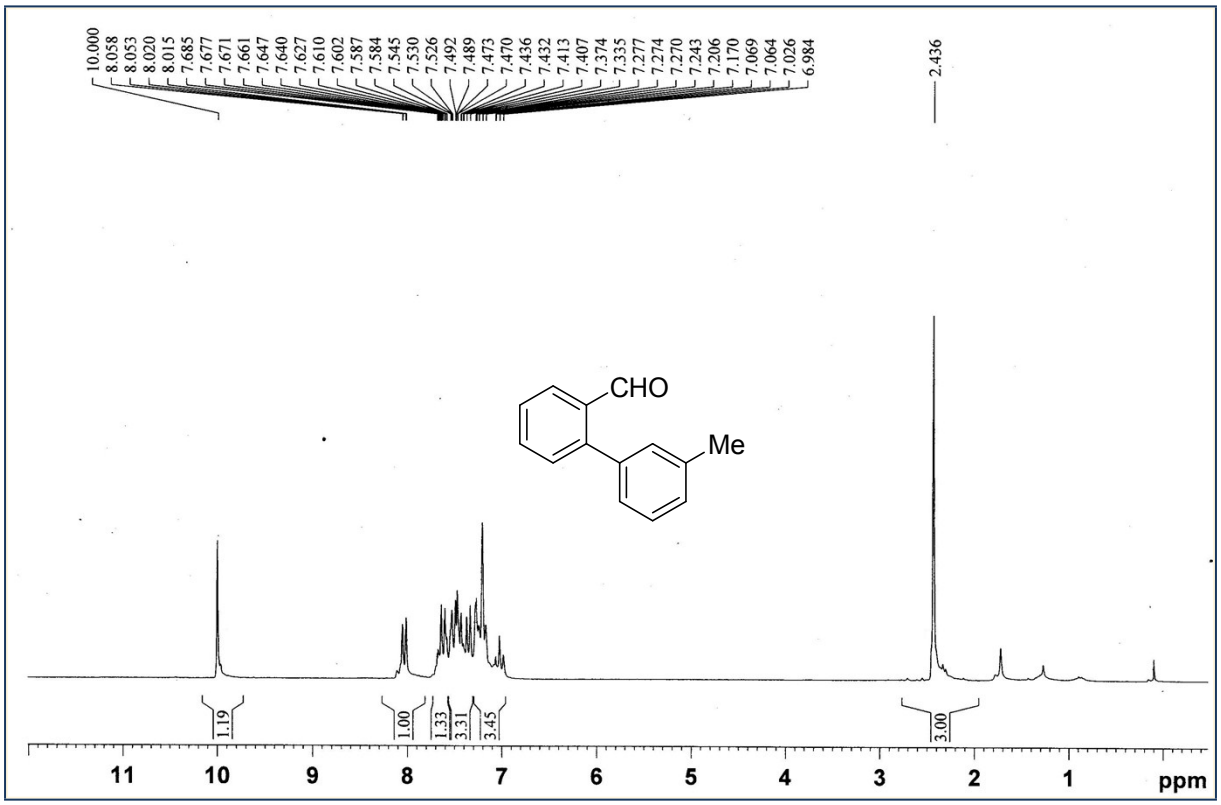
^{1}H NMR of compound 4m



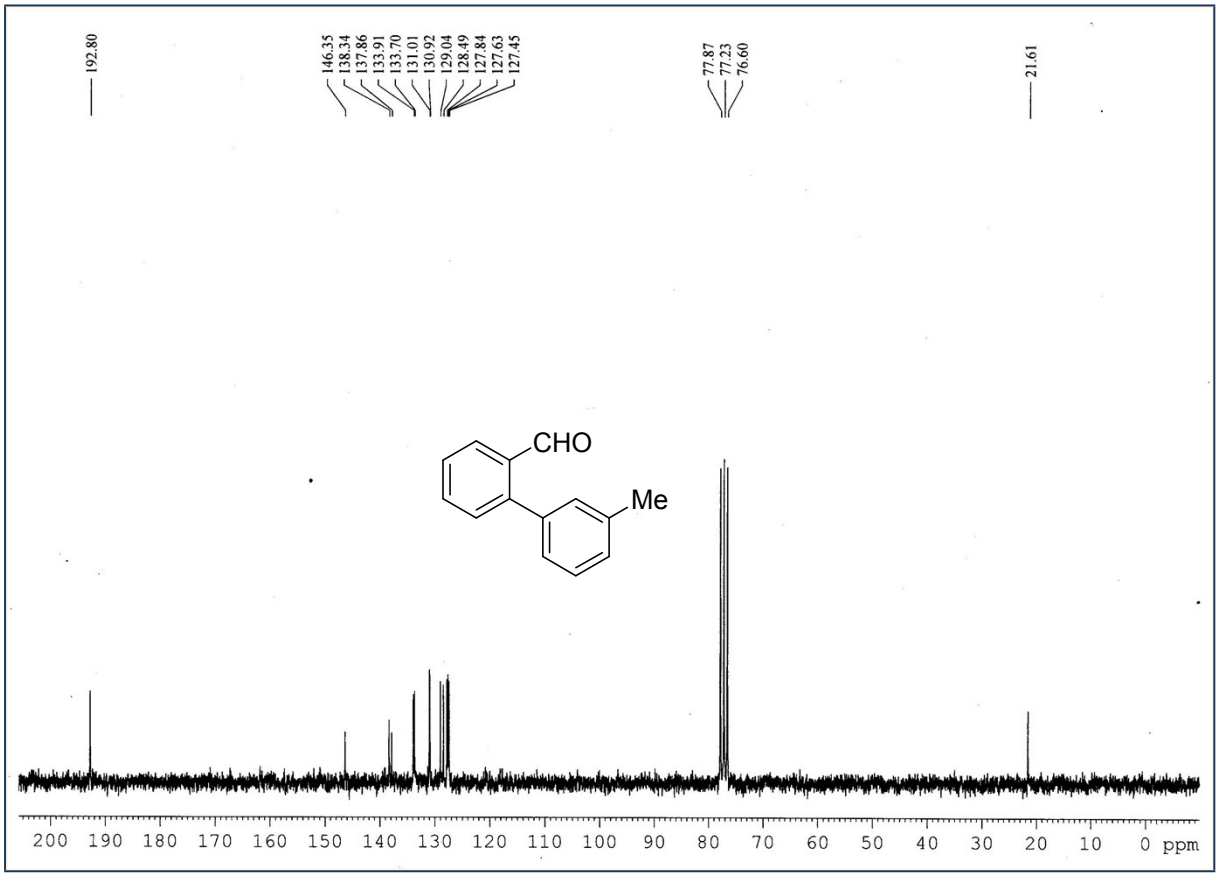
^{13}C NMR of compound 4m



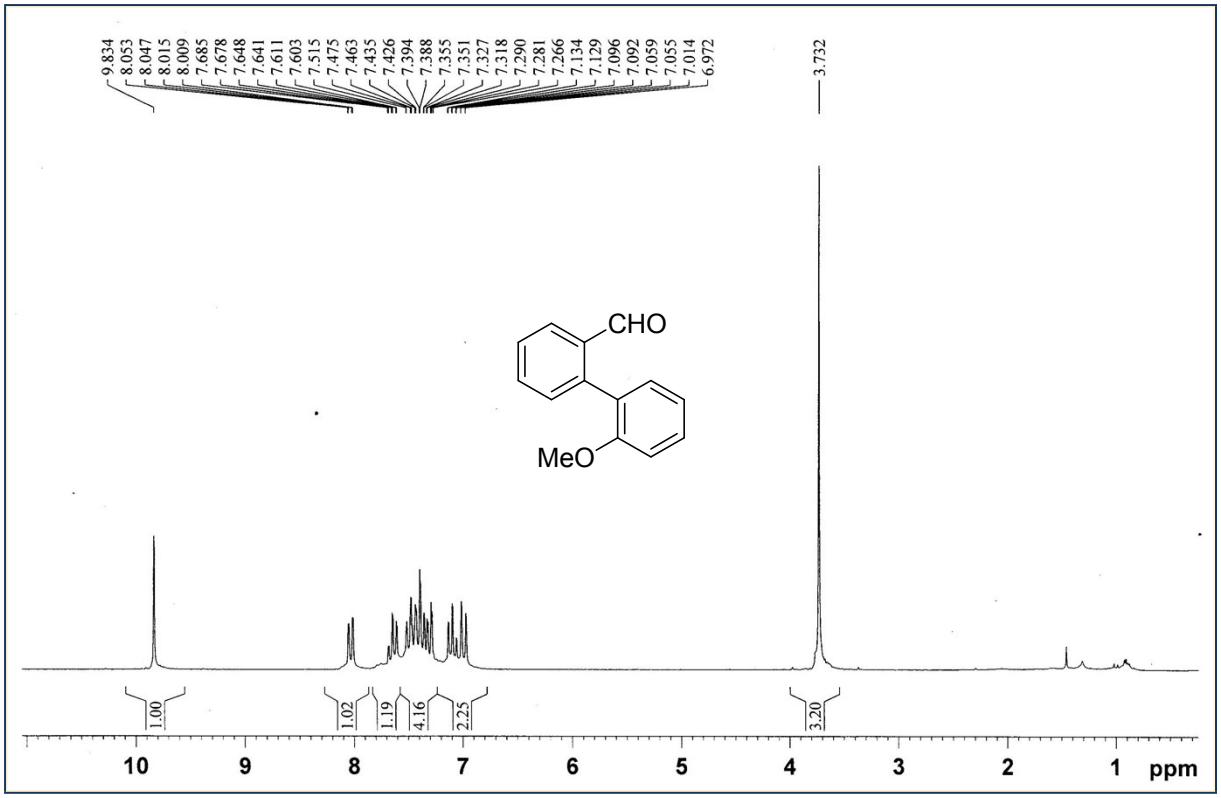
^{1}H NMR of compound 4n



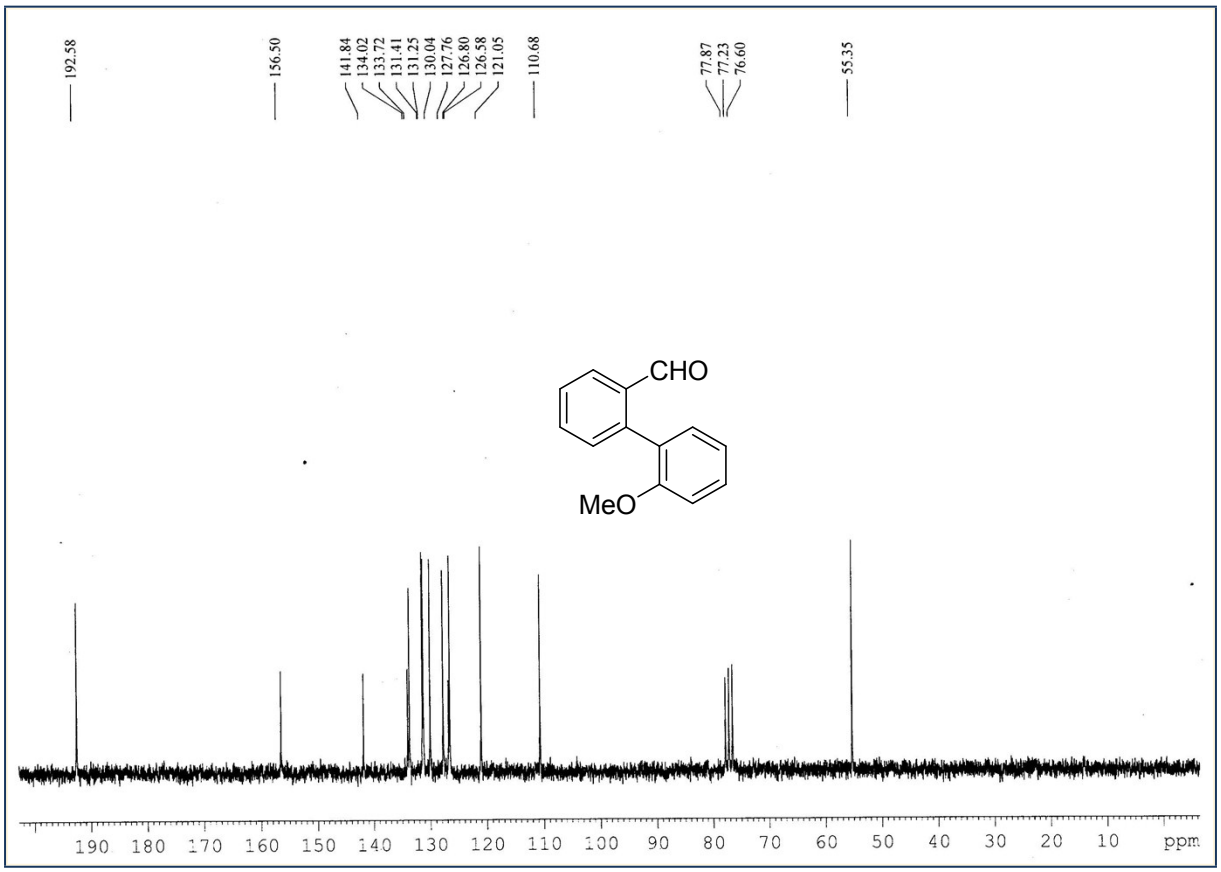
^{13}C NMR of compound 4n



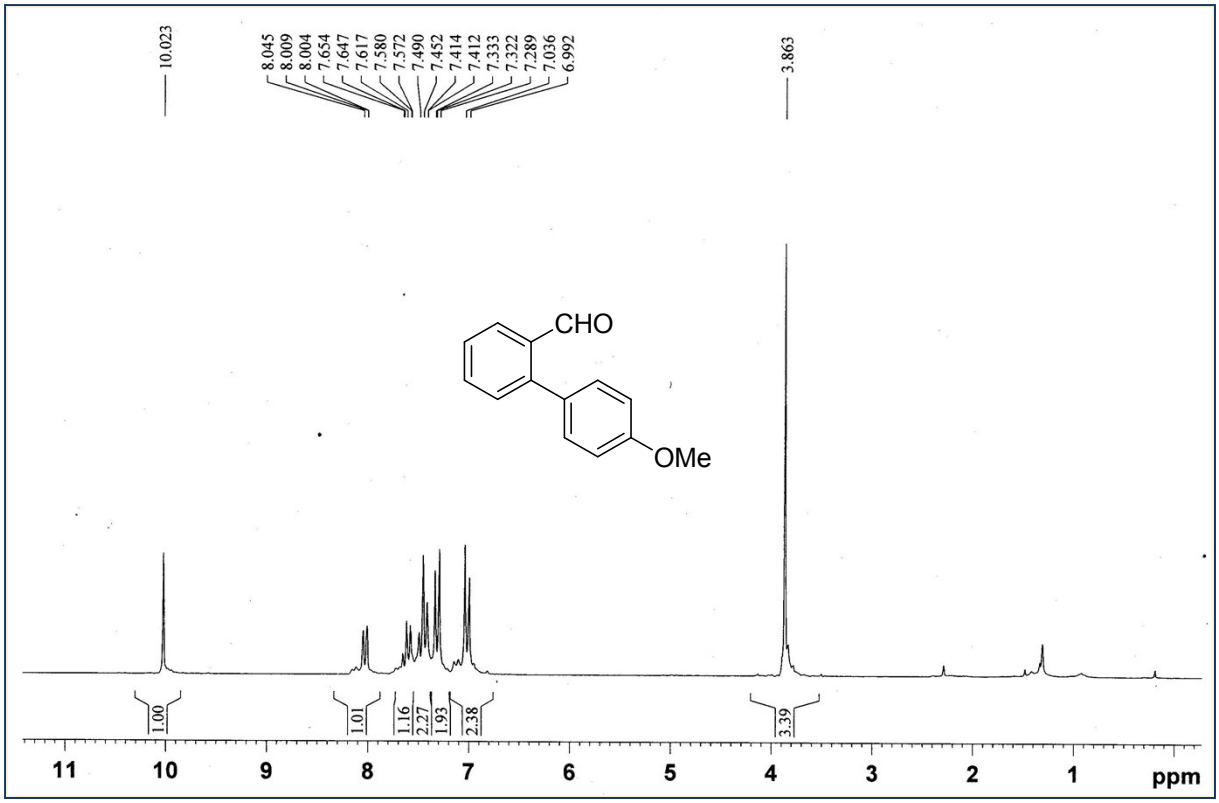
^{1}H NMR of compound 4o



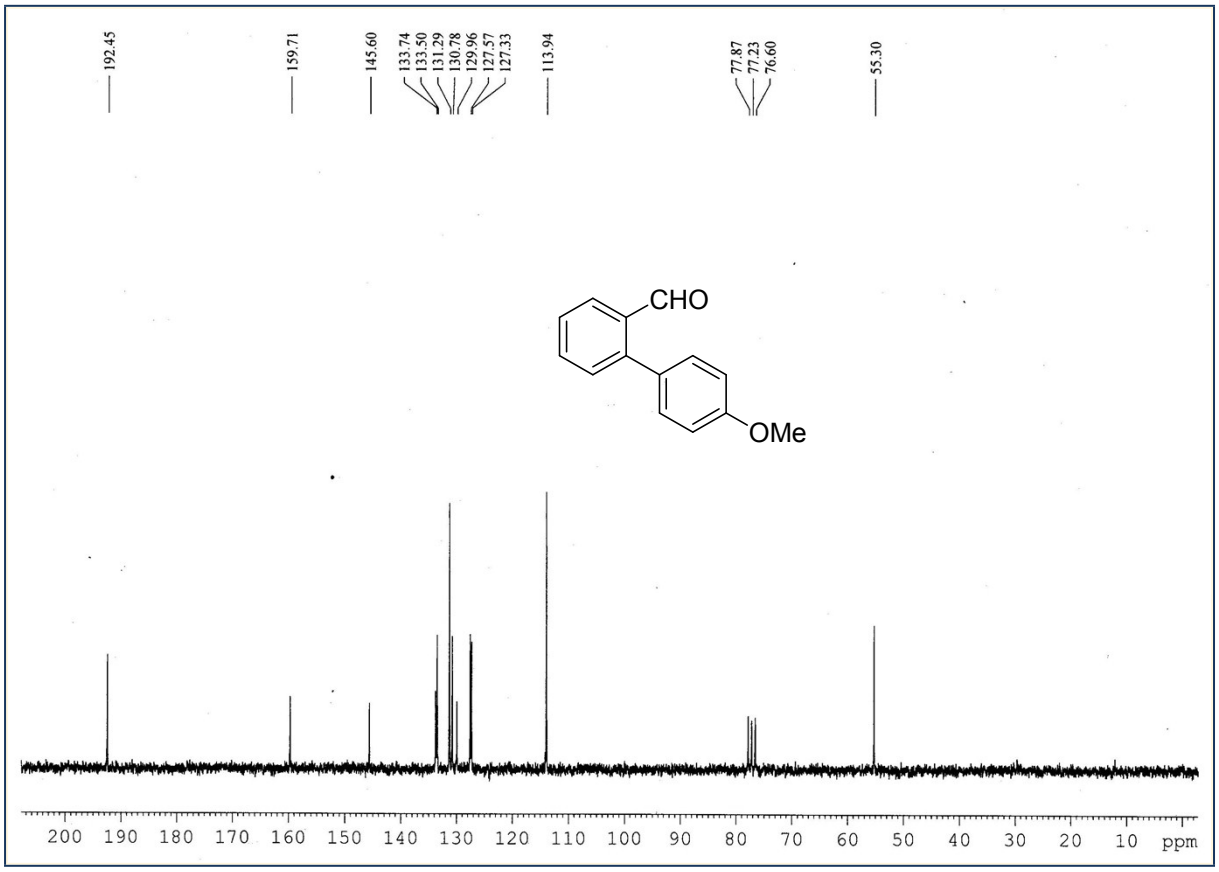
^{13}C NMR of compound 4o



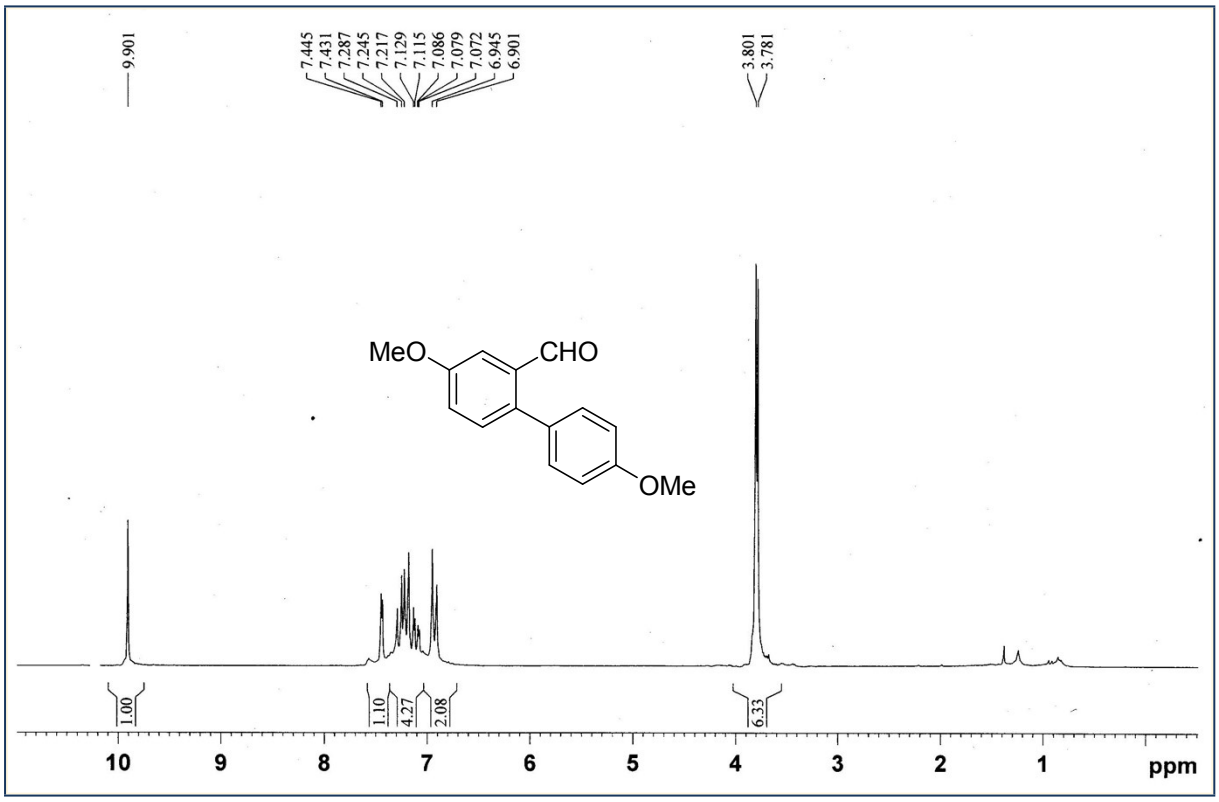
^{1}H NMR of compound 4p



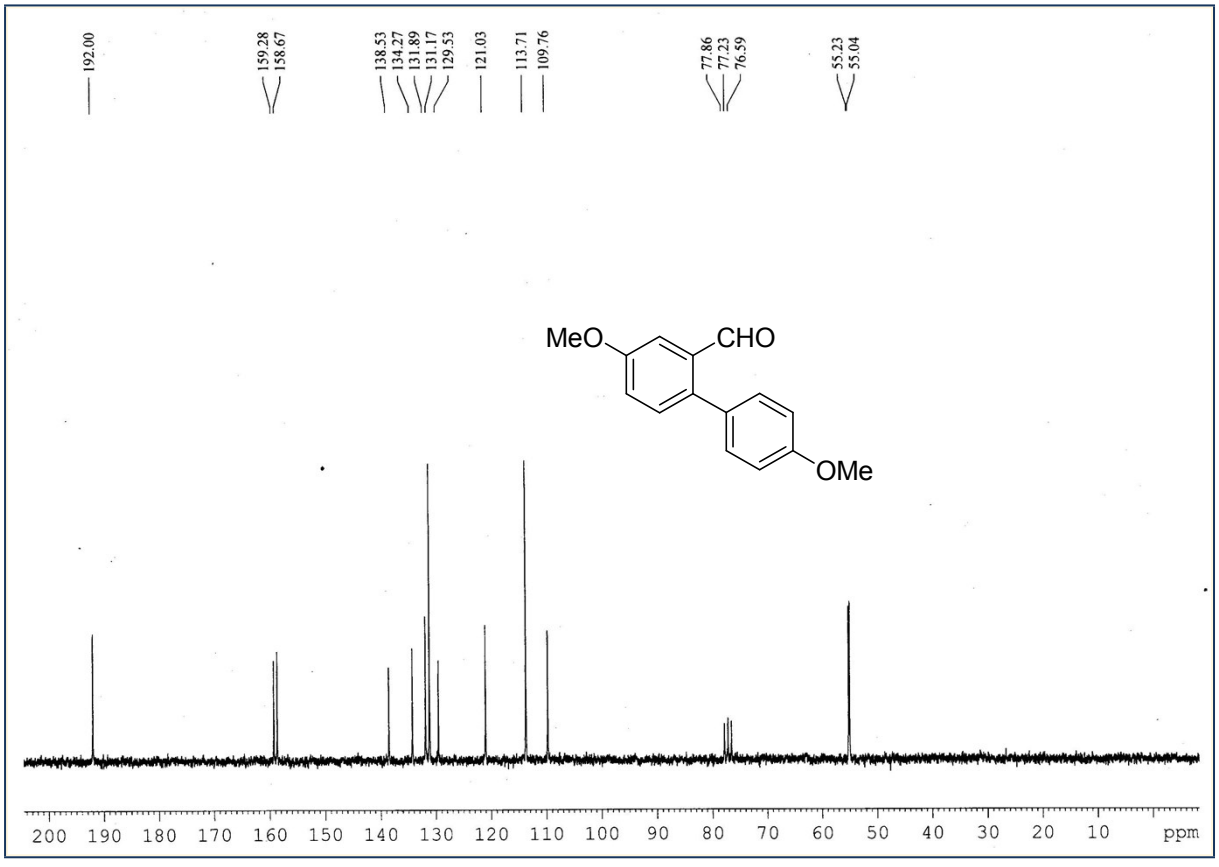
^{13}C NMR of compound 4p



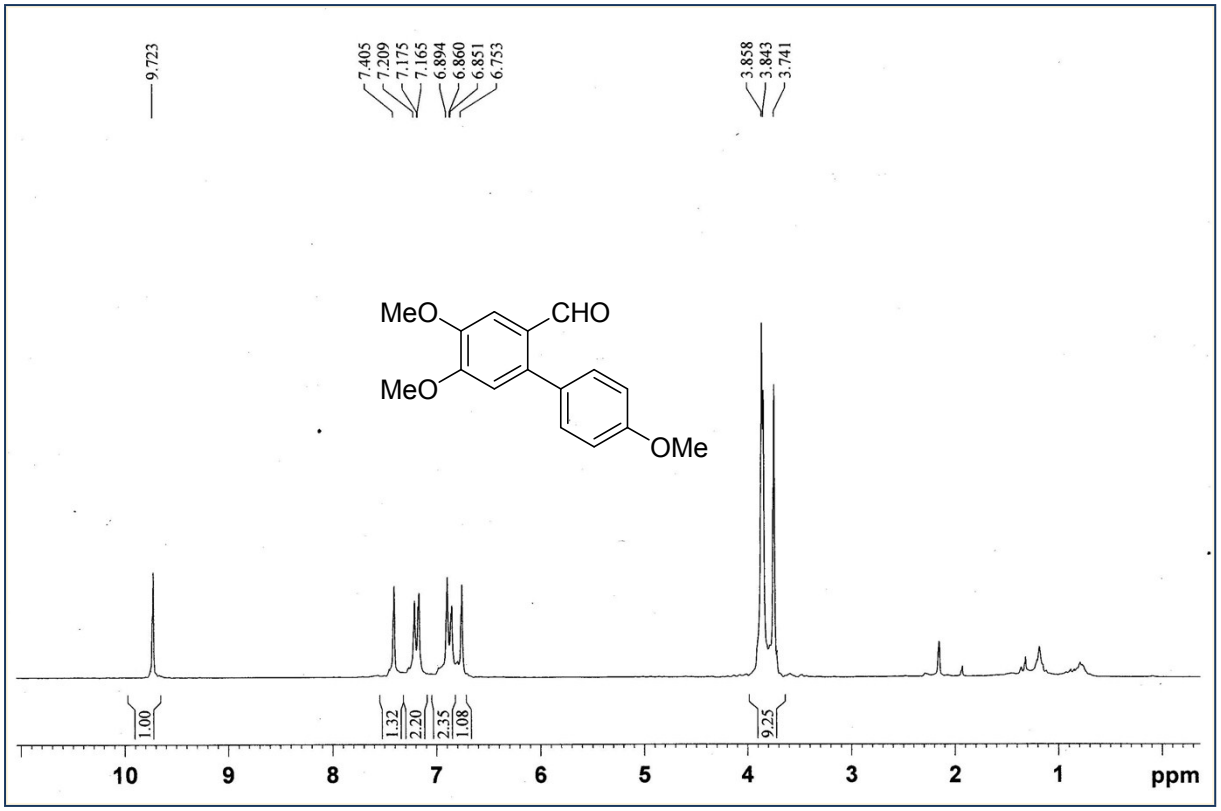
^{1}H NMR of compound 4q



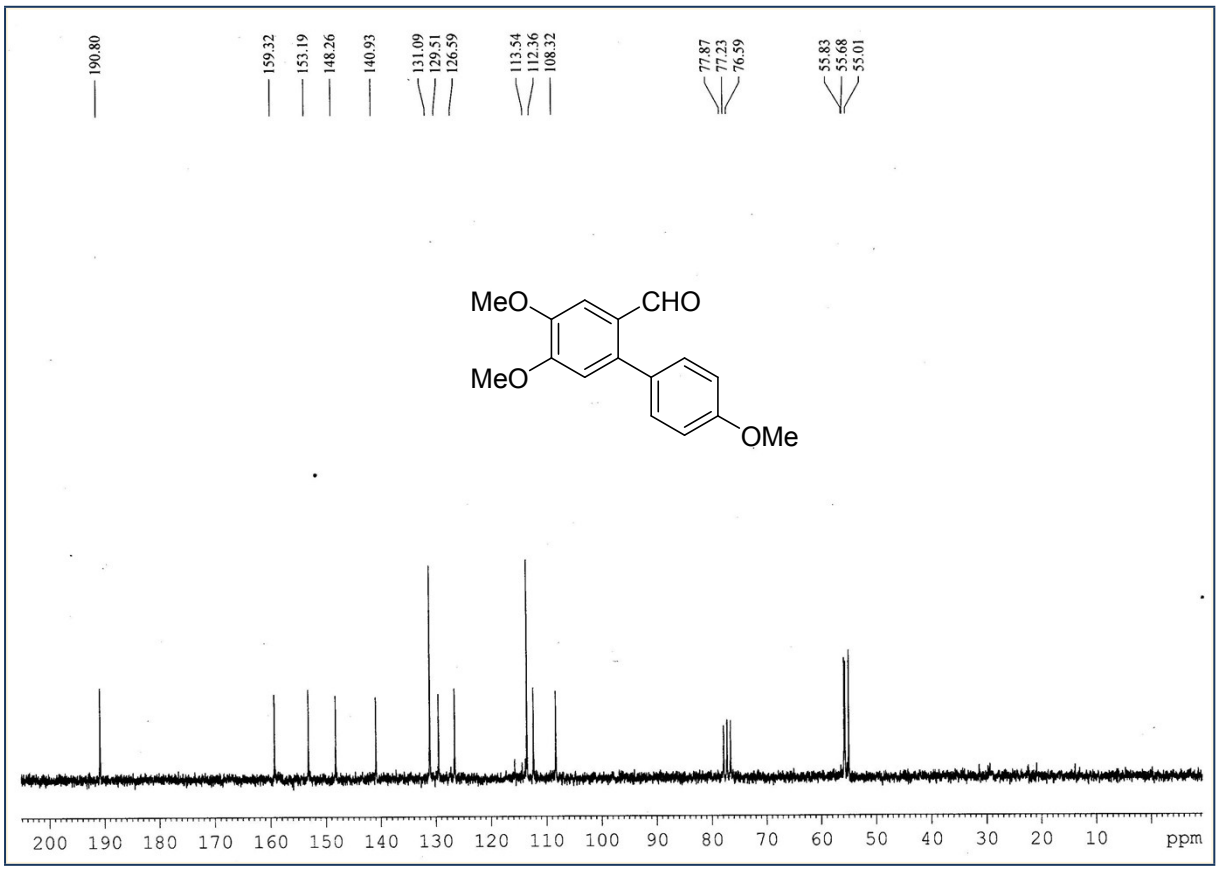
^{13}C NMR of compound 4q



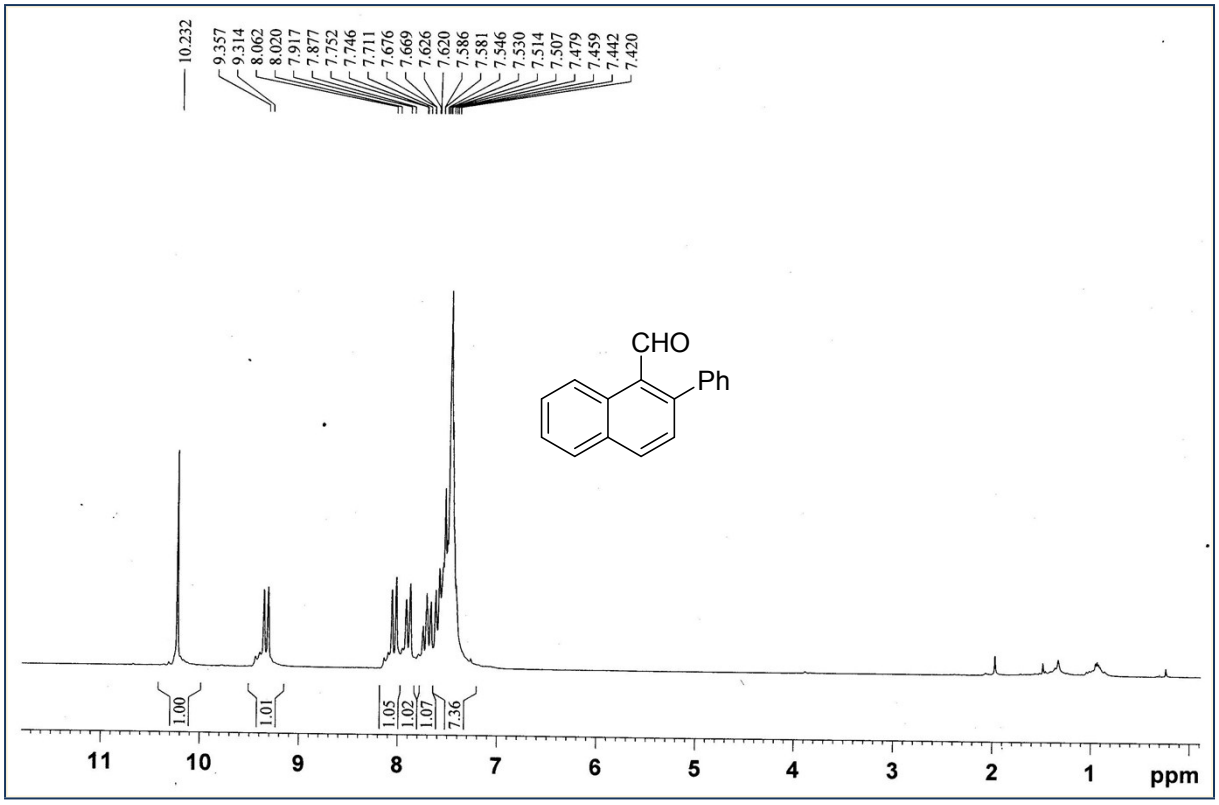
^{1}H NMR of compound 4r



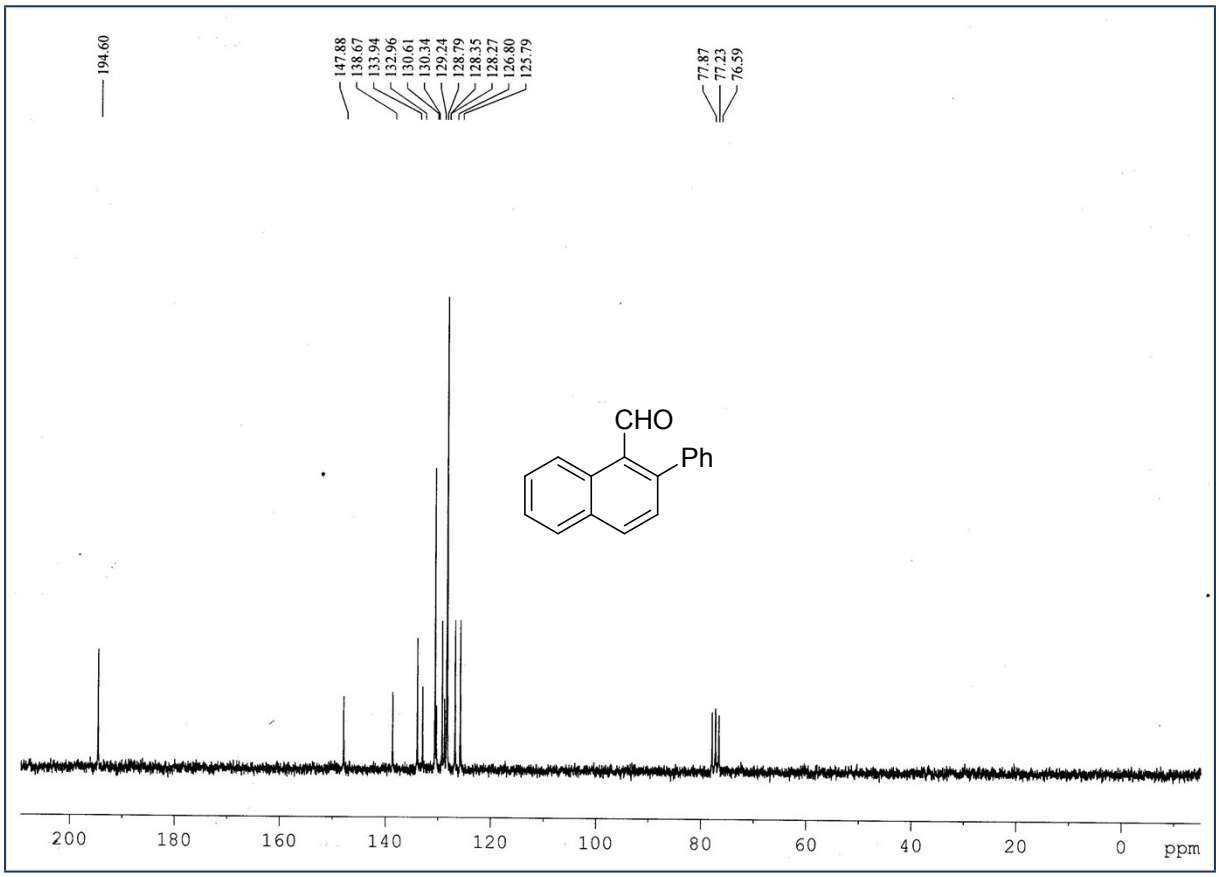
^{13}C NMR of compound 4r



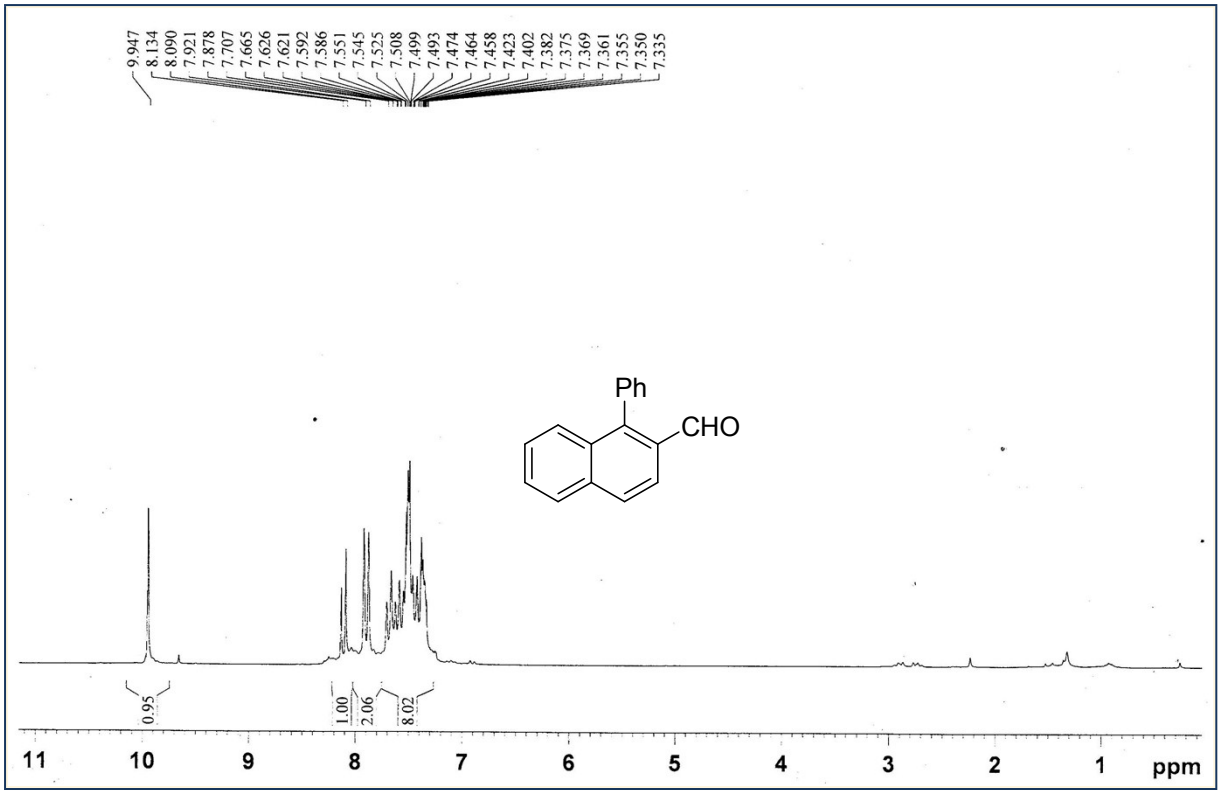
^{1}H NMR of compound 4s



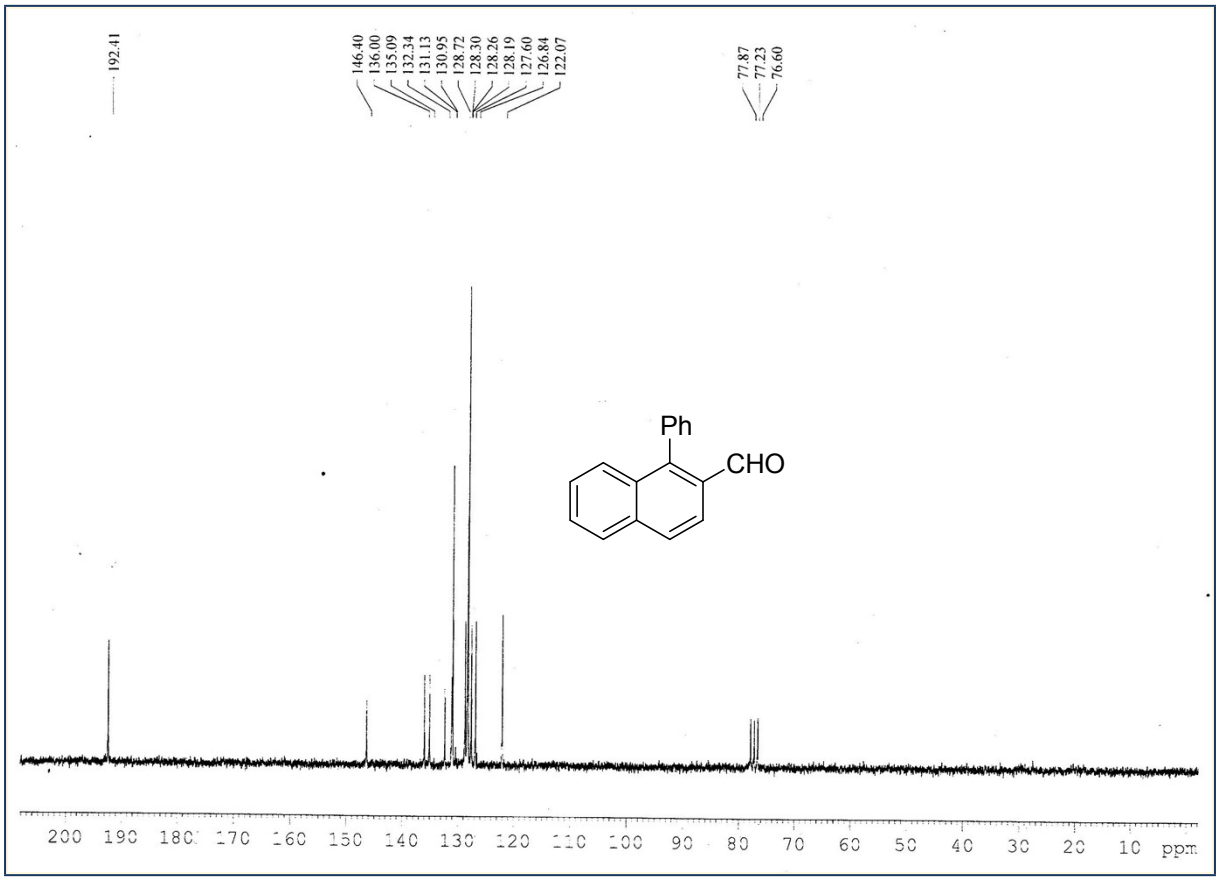
^{13}C NMR of compound 4s



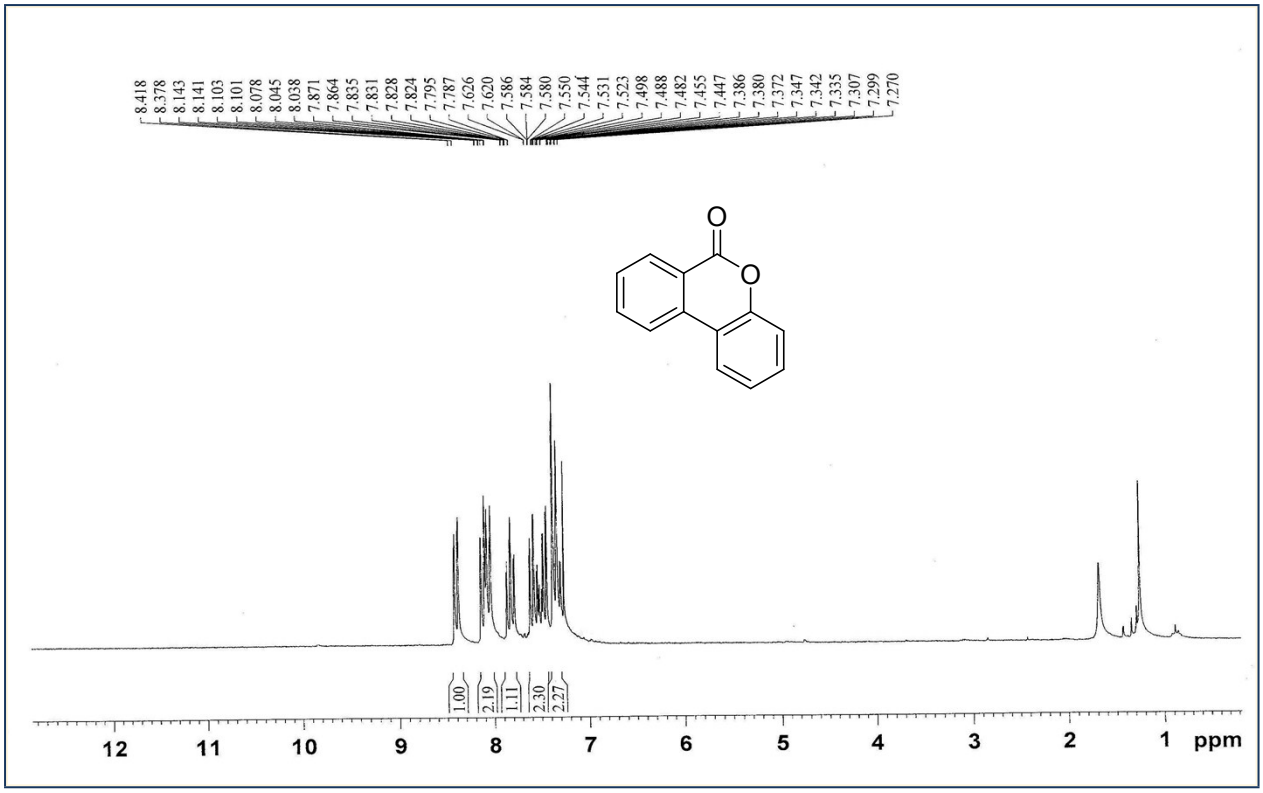
^{1}H NMR of compound 4t



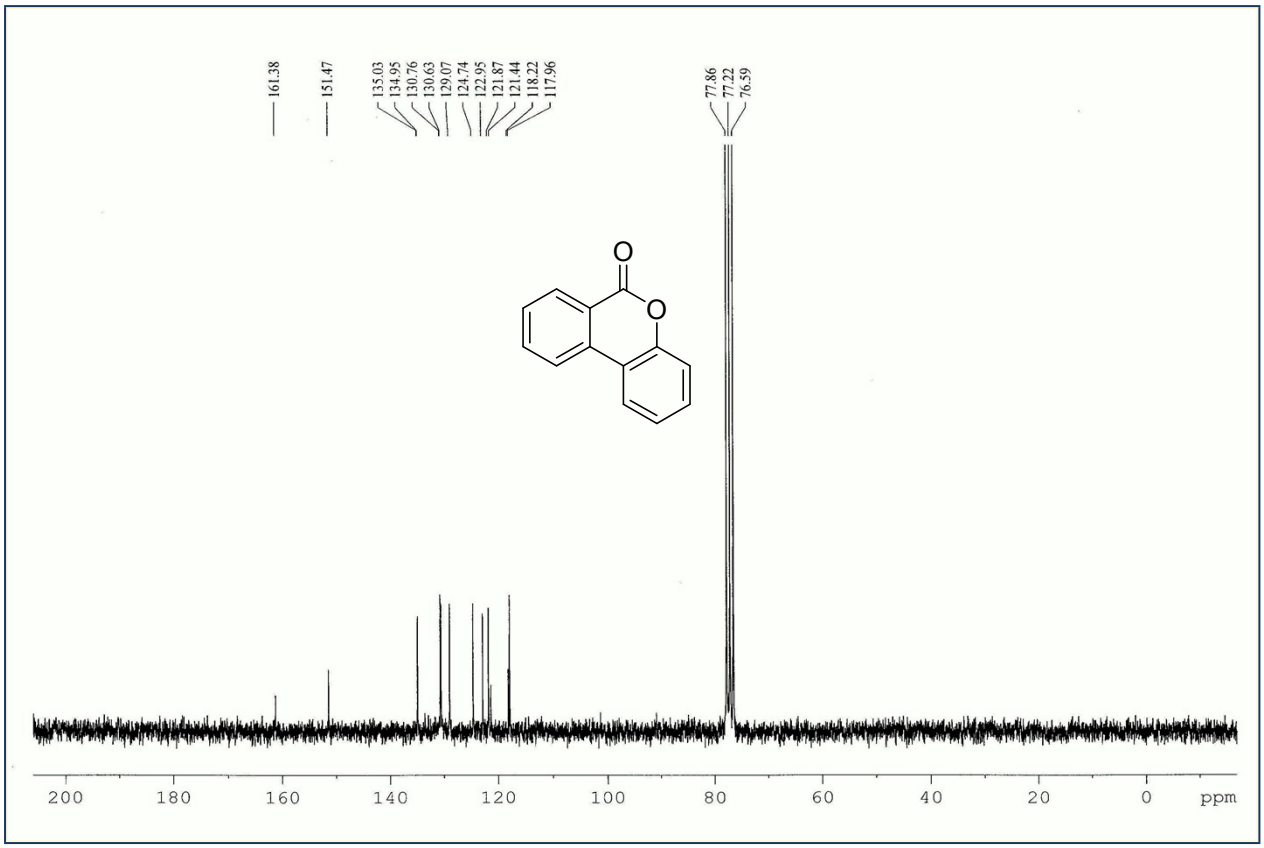
^{13}C NMR of compound 4t



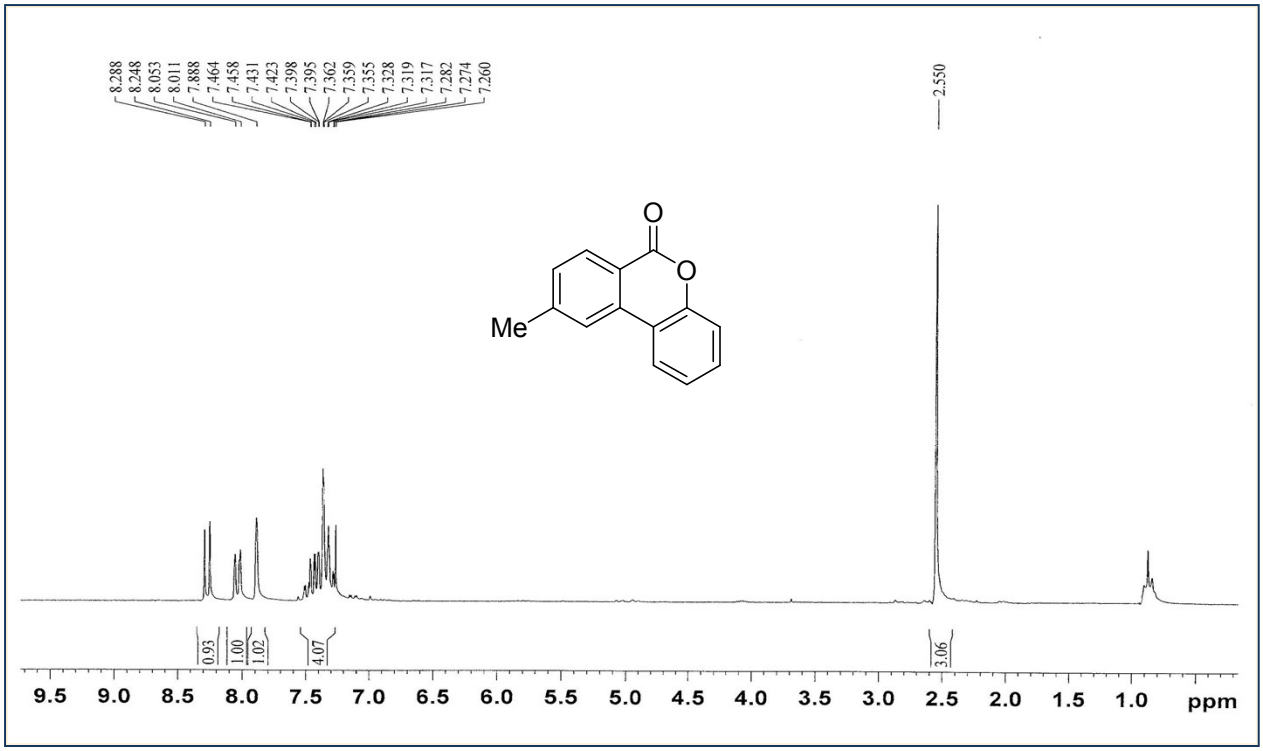
^{1}H NMR of compound 5a



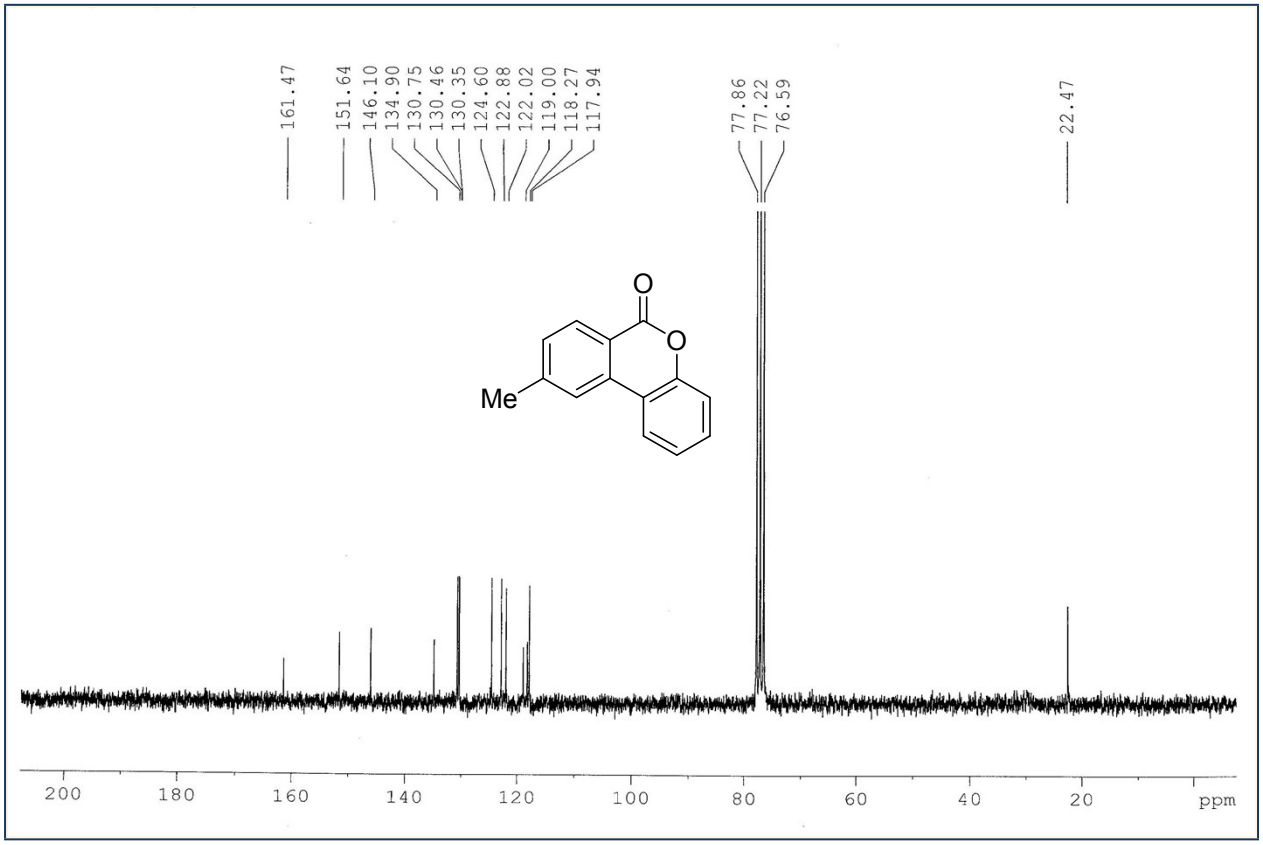
^{13}C NMR of compound 5a



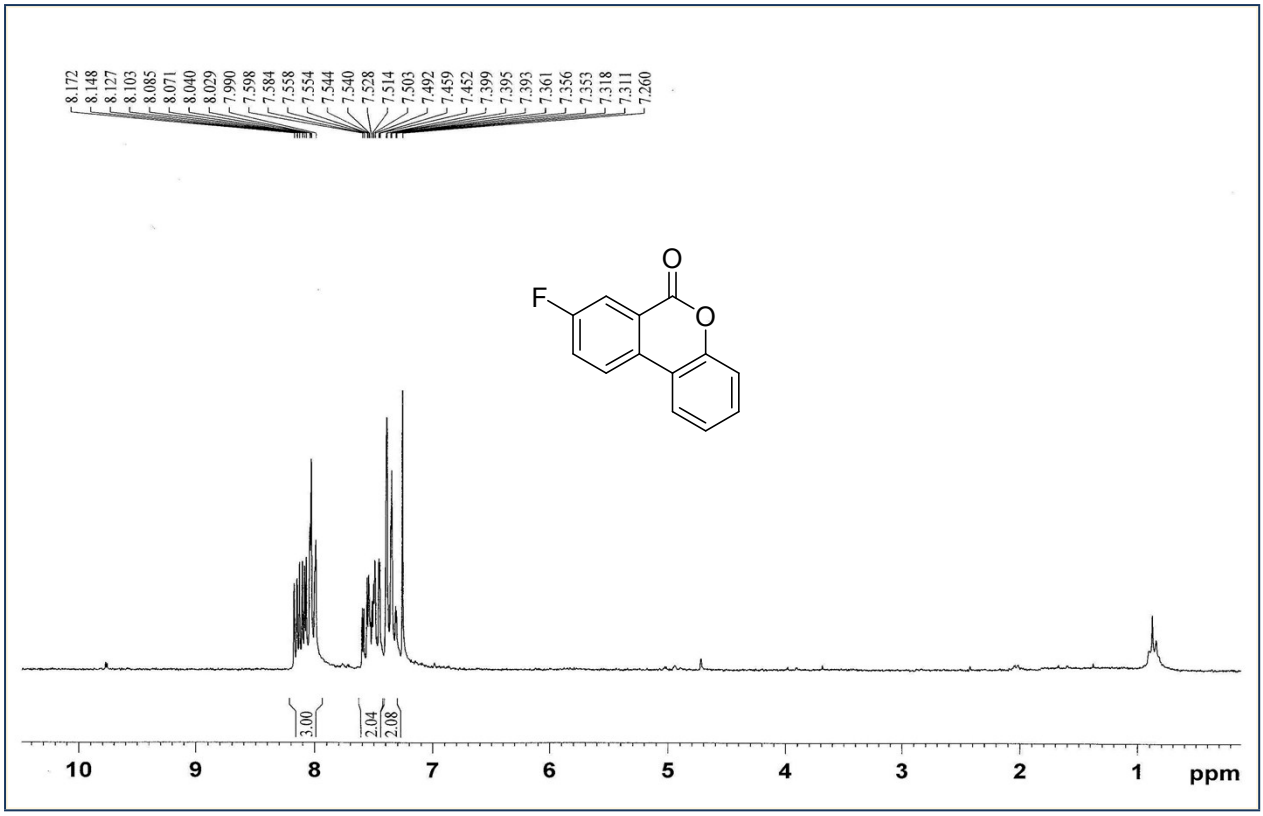
^{1}H NMR of compound 5b



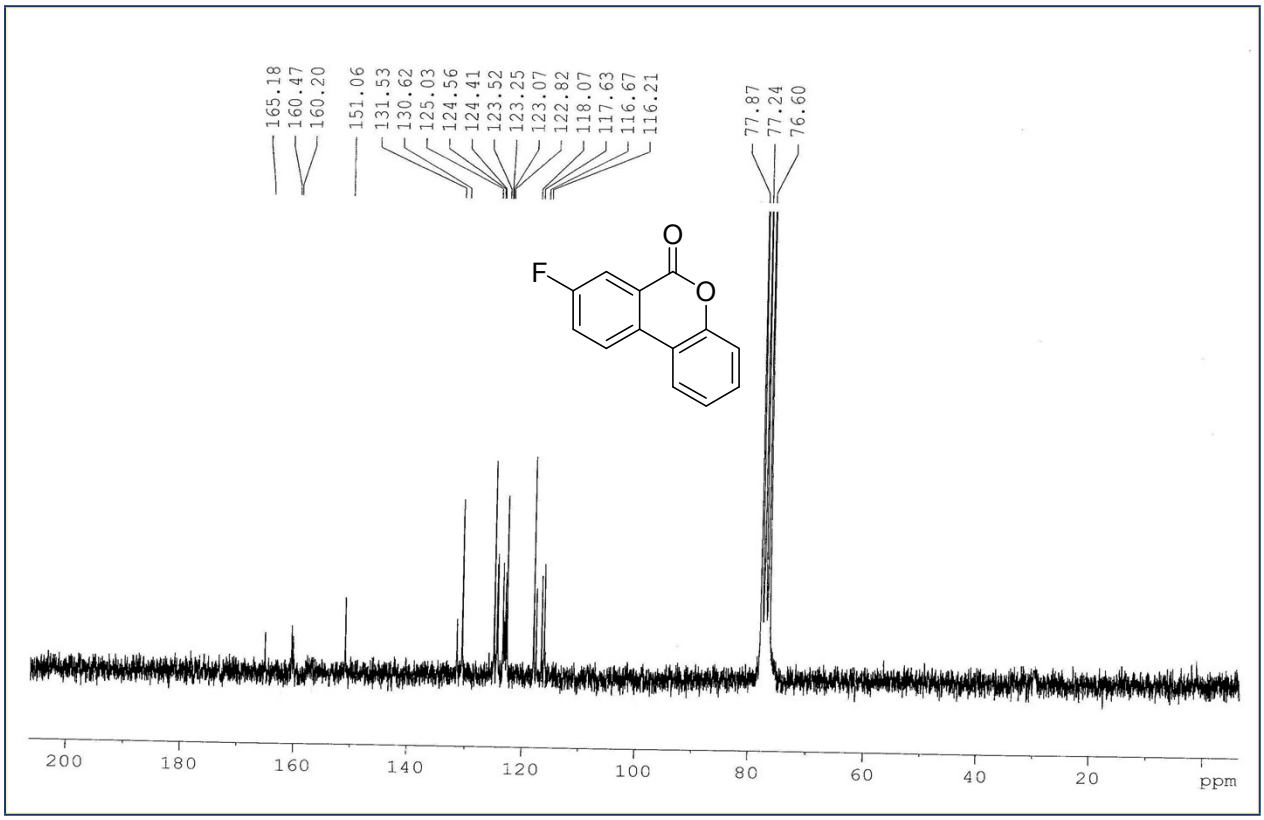
^{13}C NMR of compound 5b



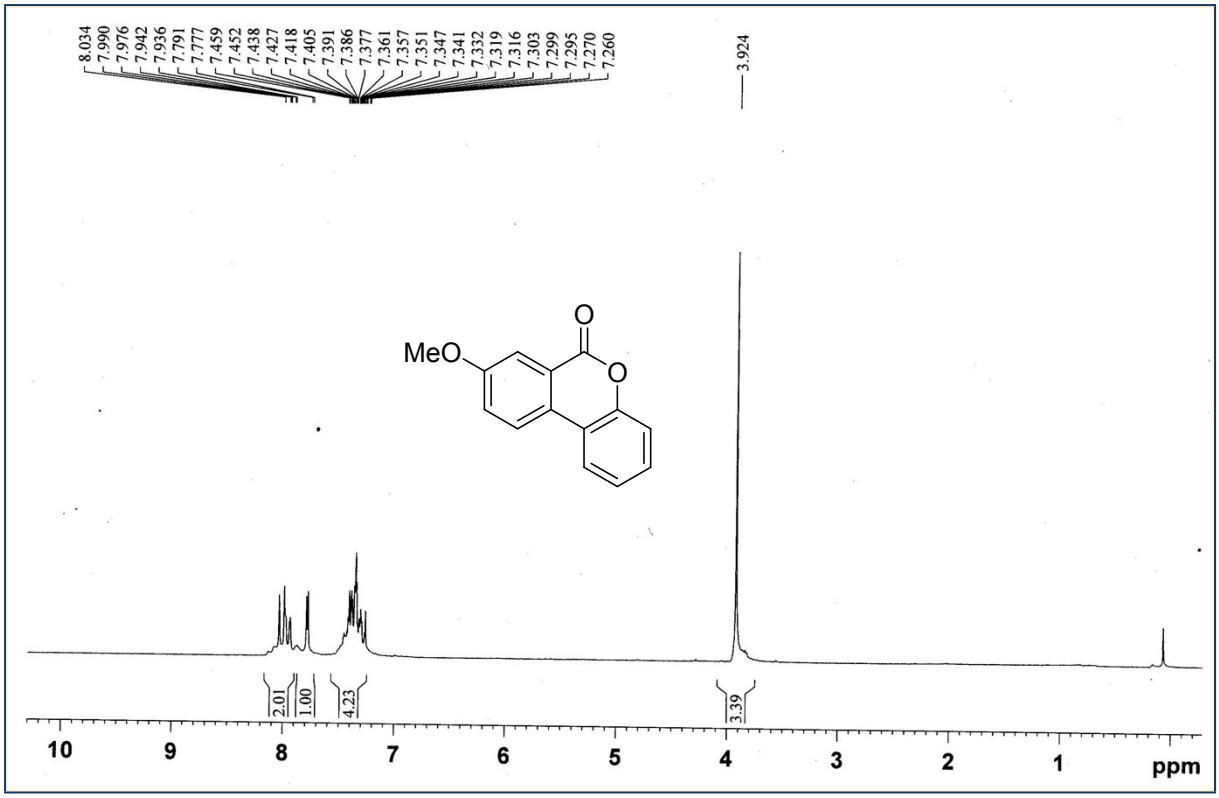
^{1}H NMR of compound 5c



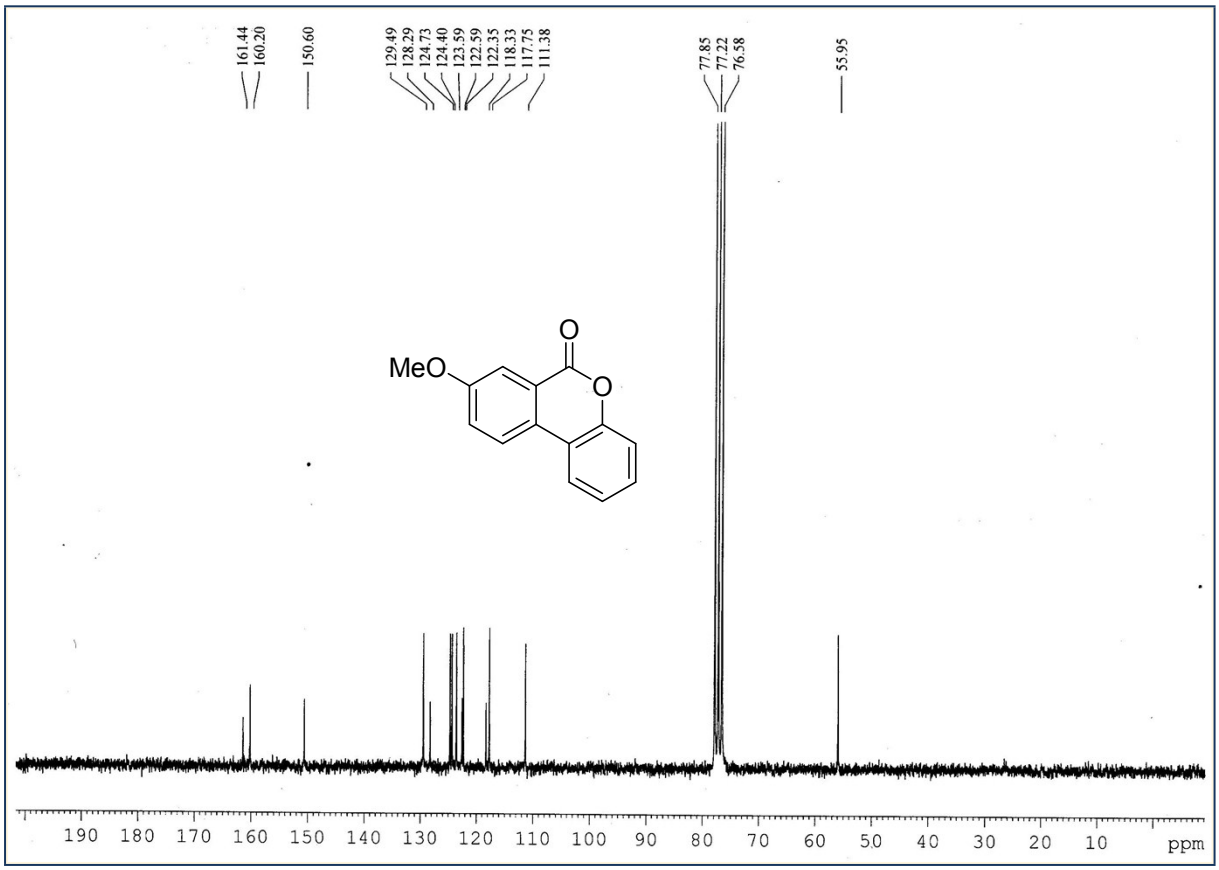
^{13}C NMR of compound 5c



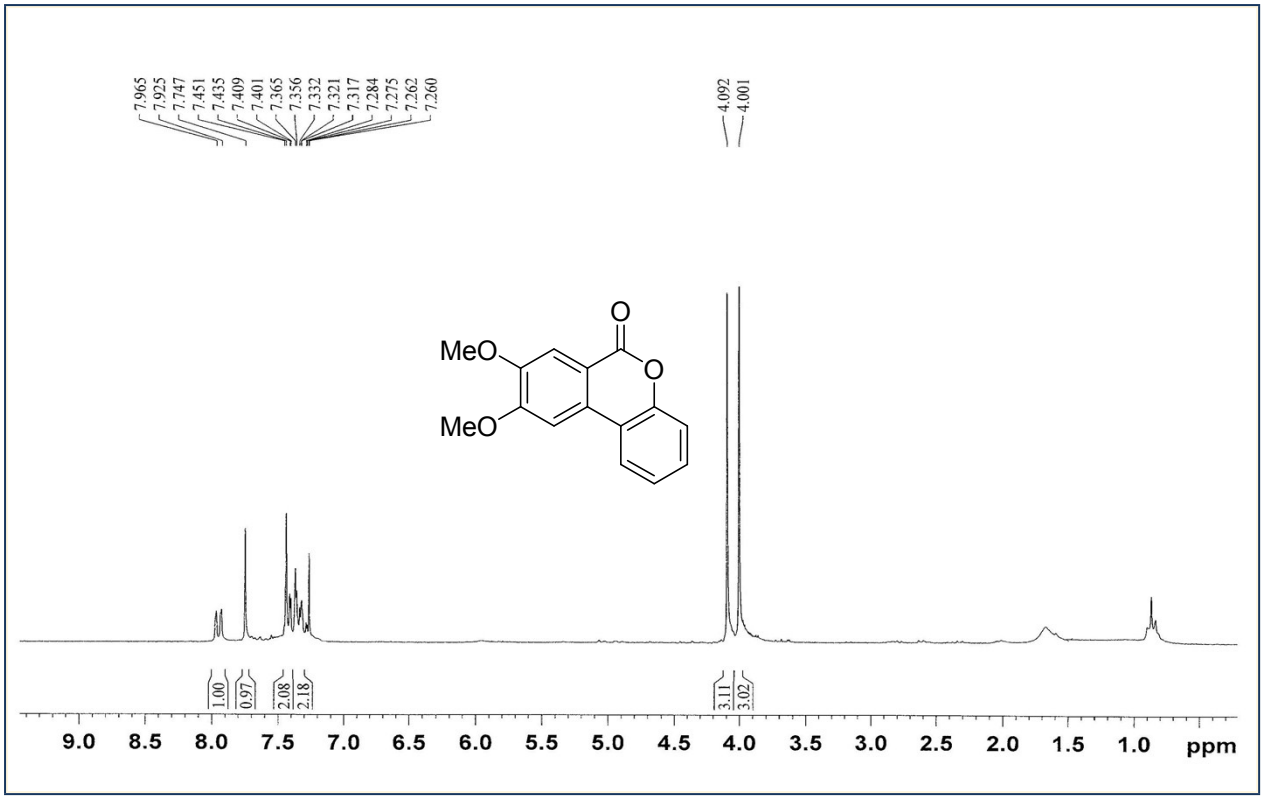
^{1}H NMR of compound 4d



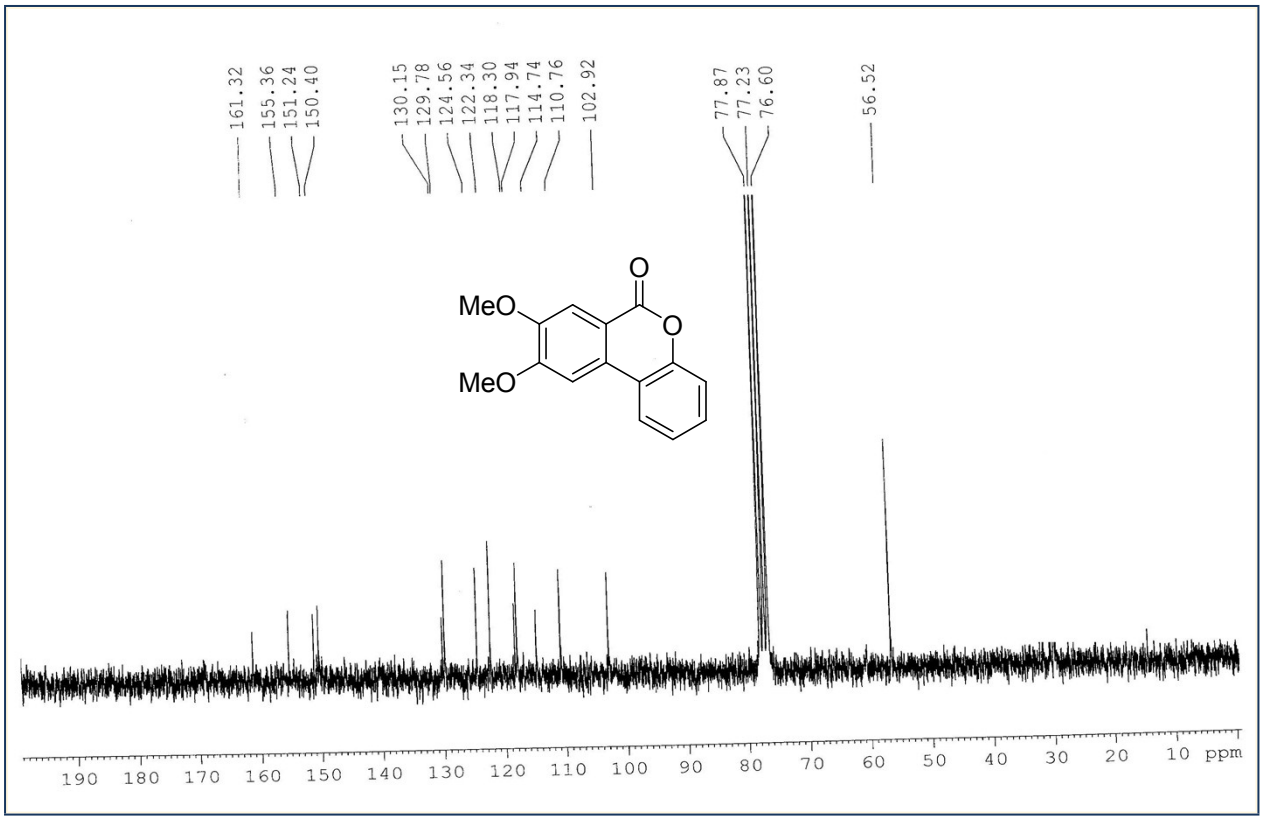
^{13}C NMR of compound 5d



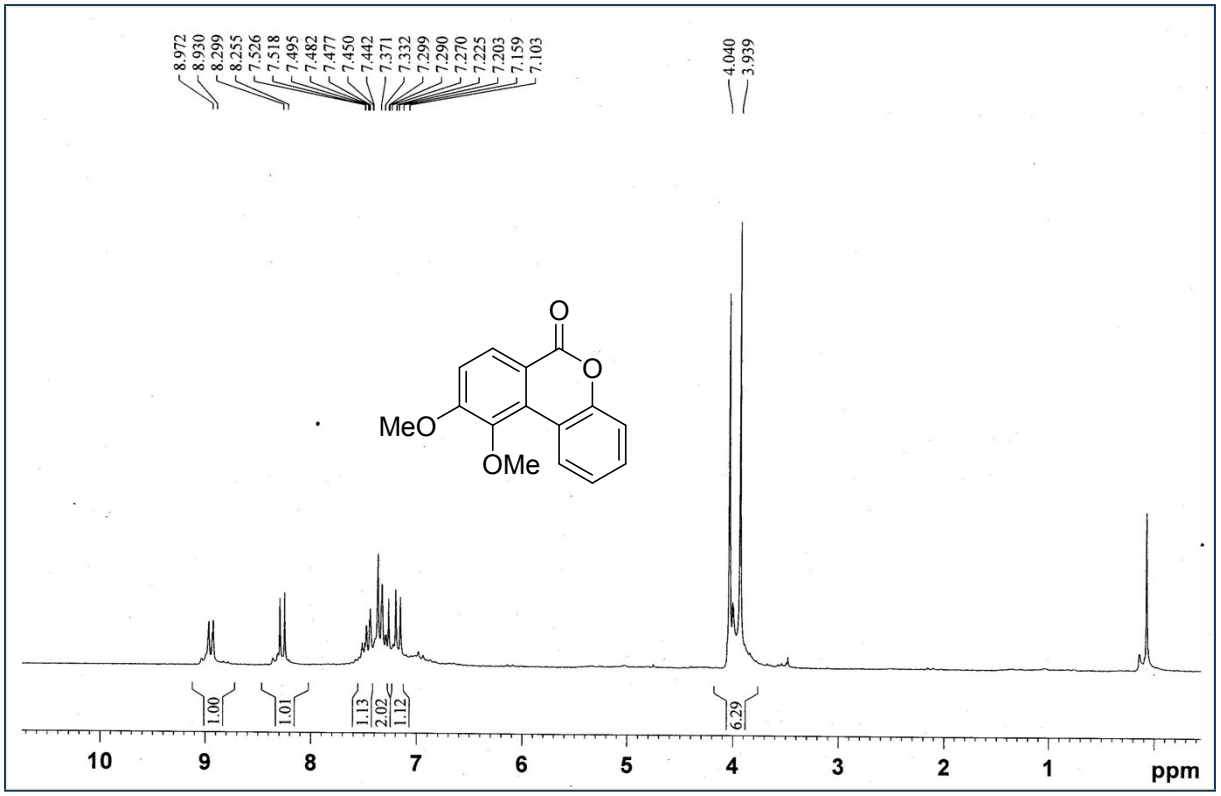
^{1}H NMR of compound 5e



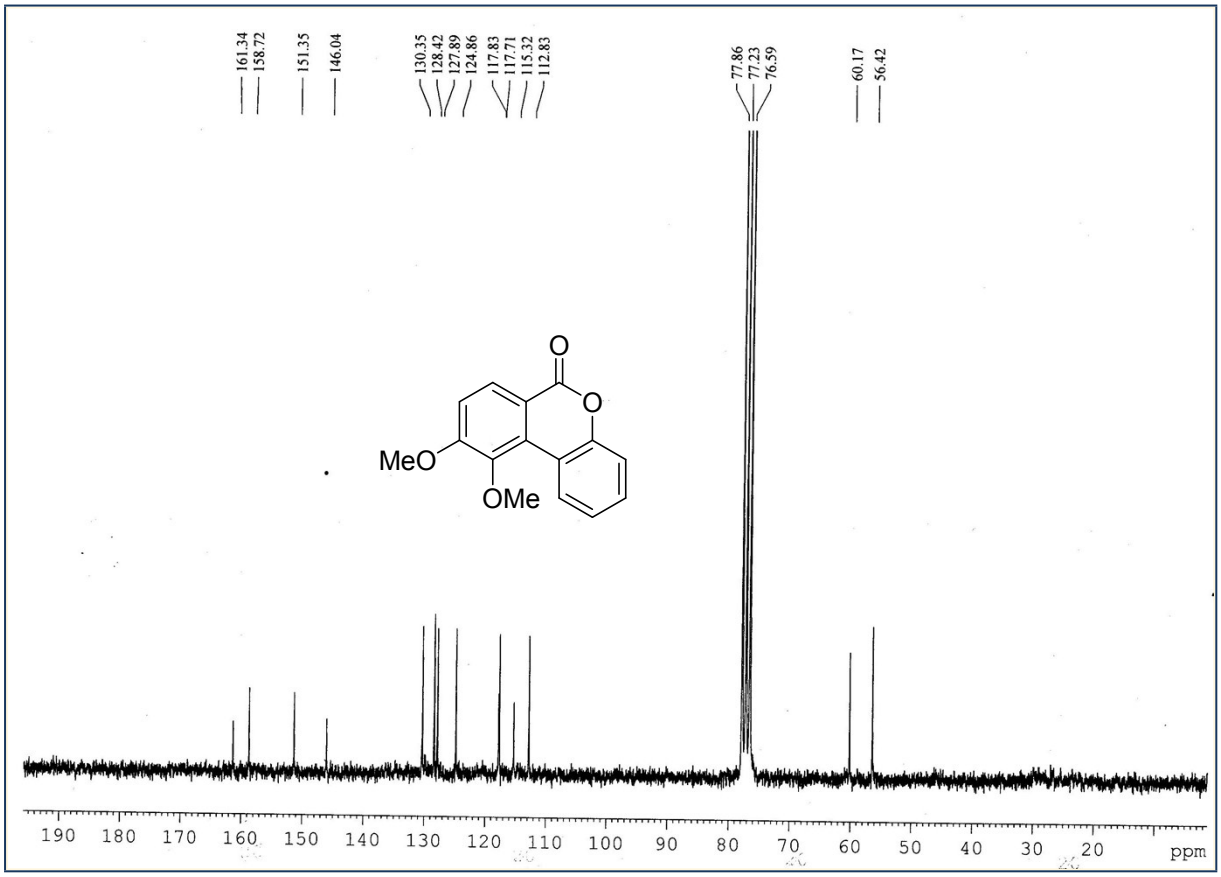
^{13C} NMR of compound 5e



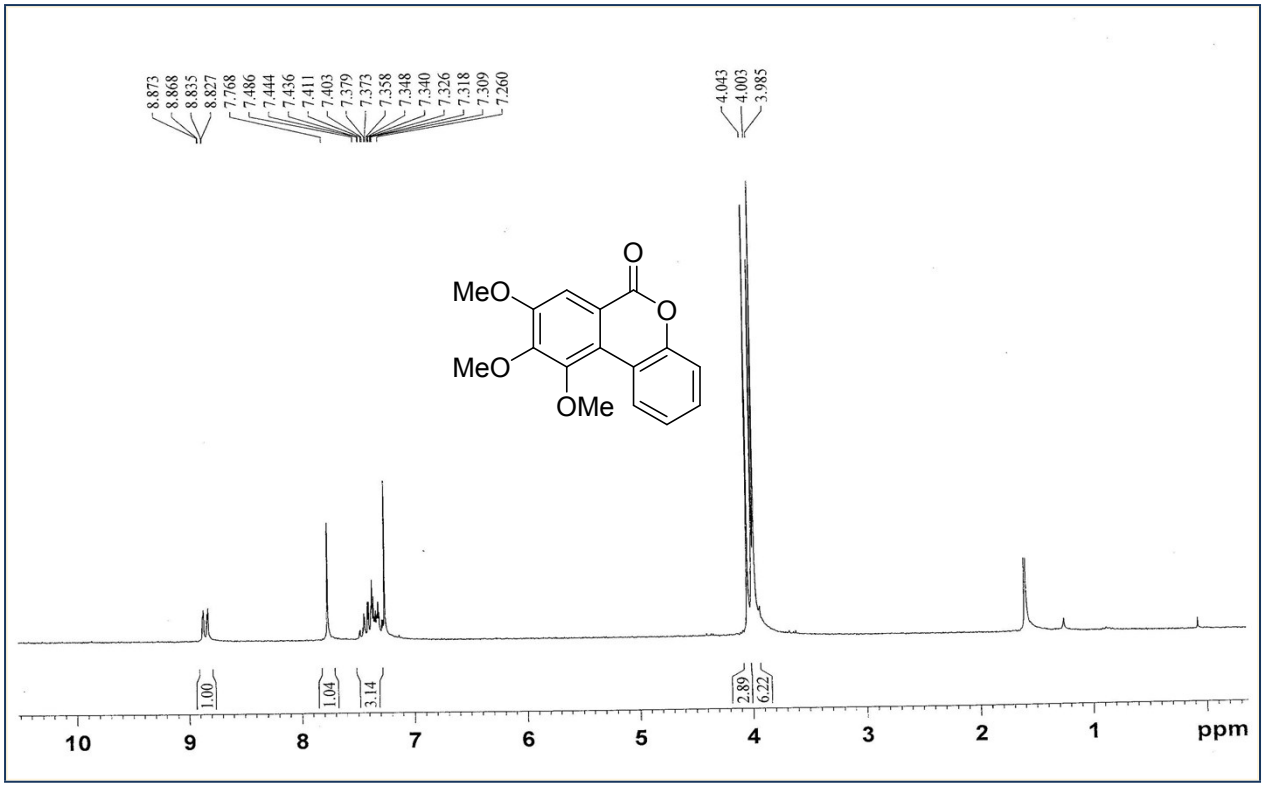
^{1}H NMR of compound 5f



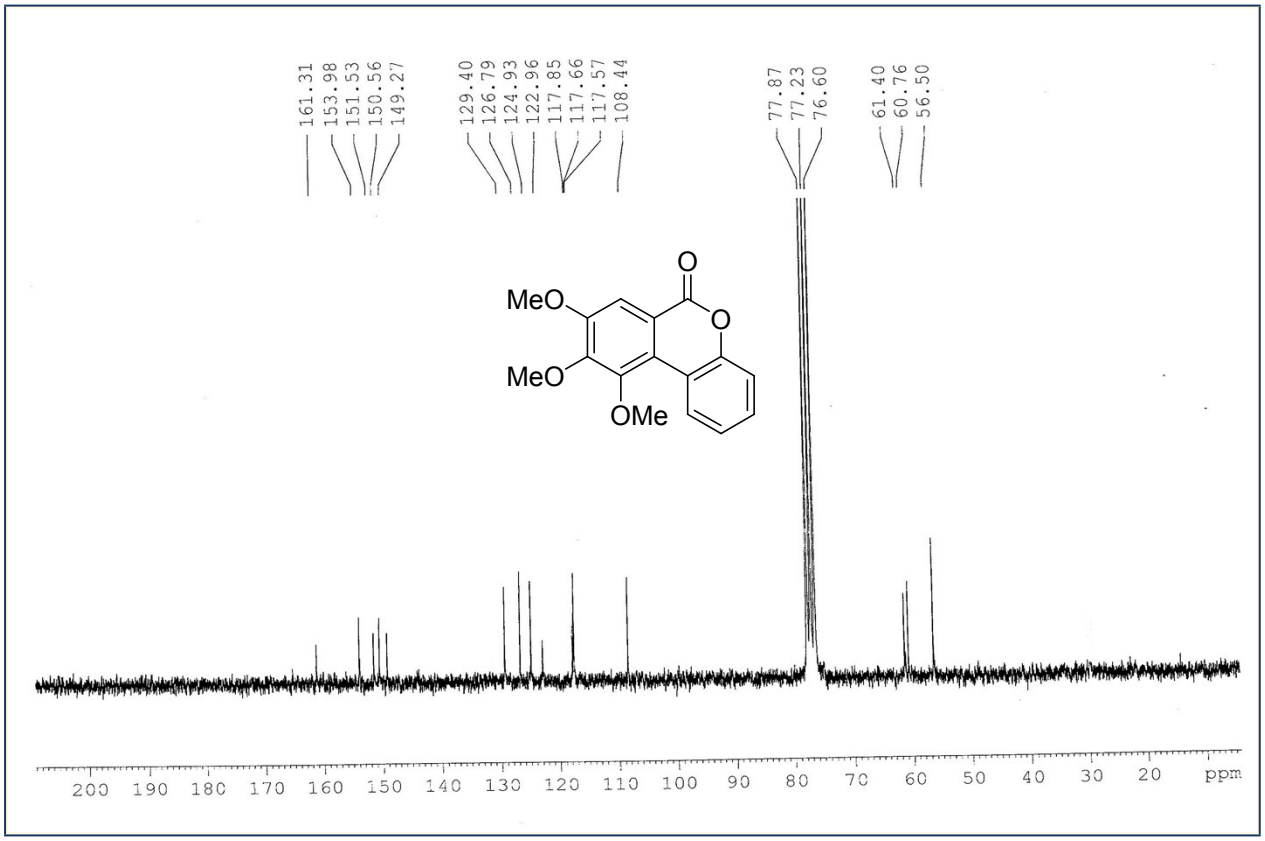
^{13}C NMR of compound 5f



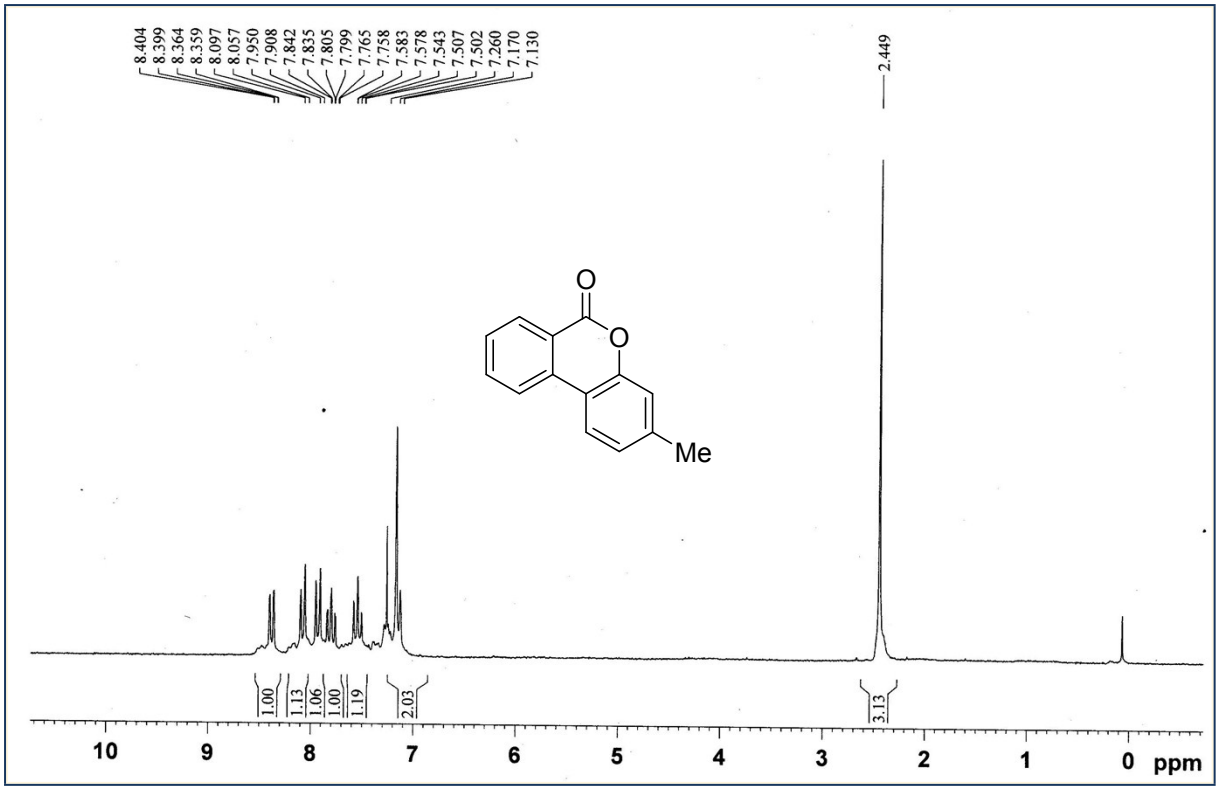
^{1}H NMR of compound 5g



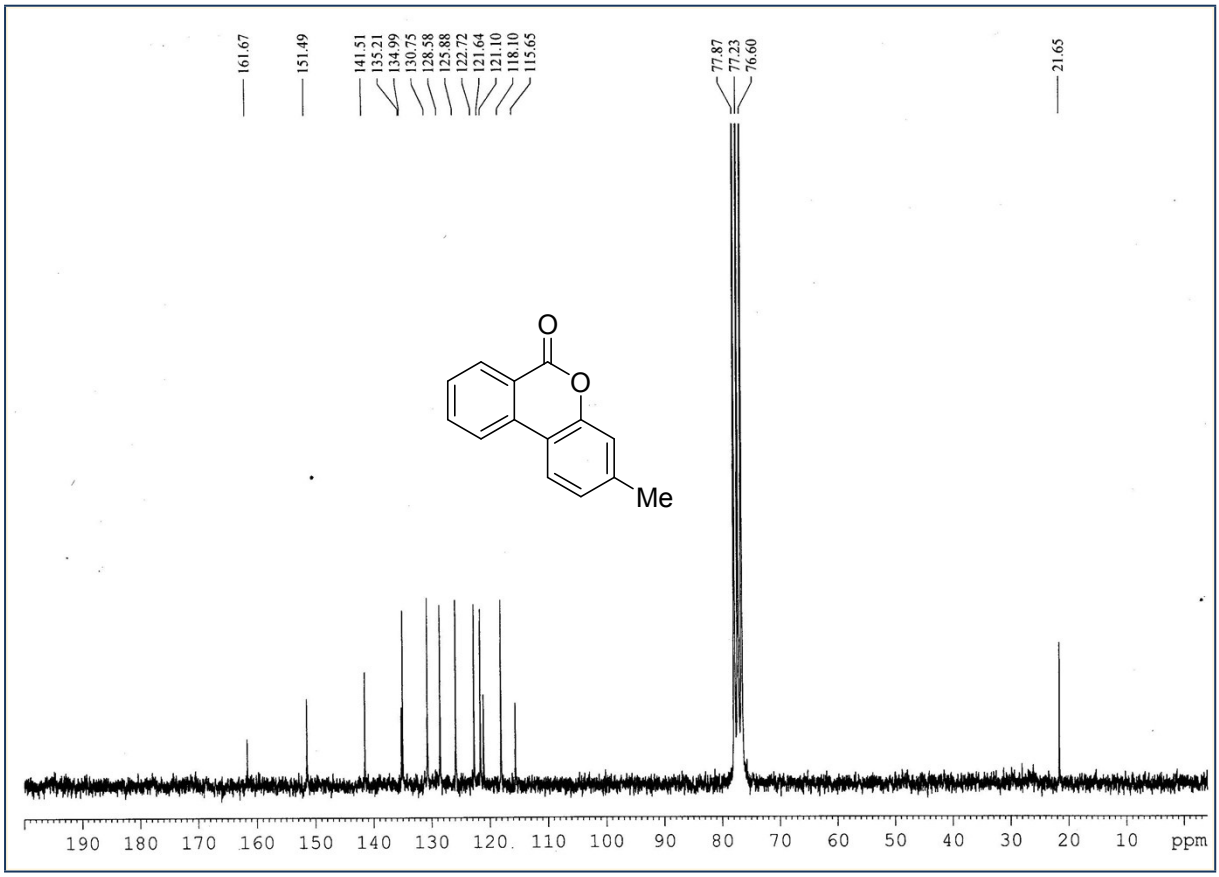
^{13}C NMR of compound 5g



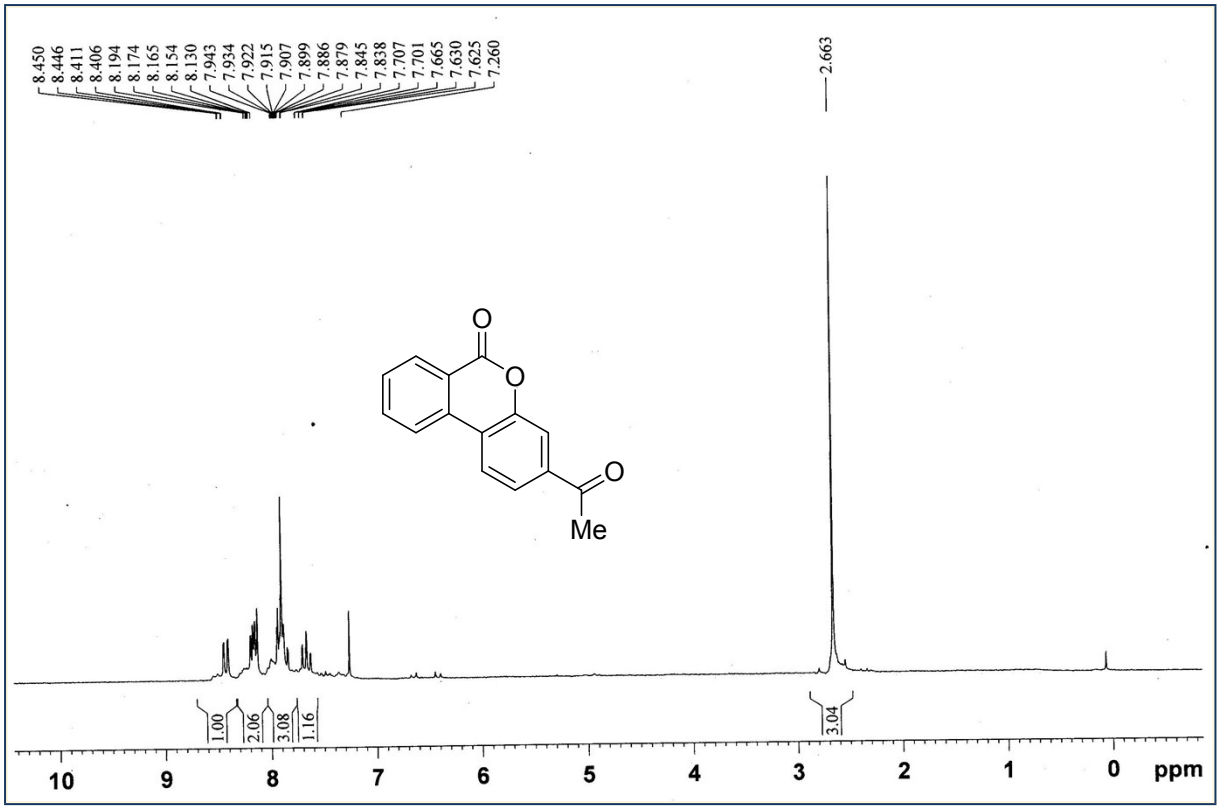
^{1}H NMR of compound 5h



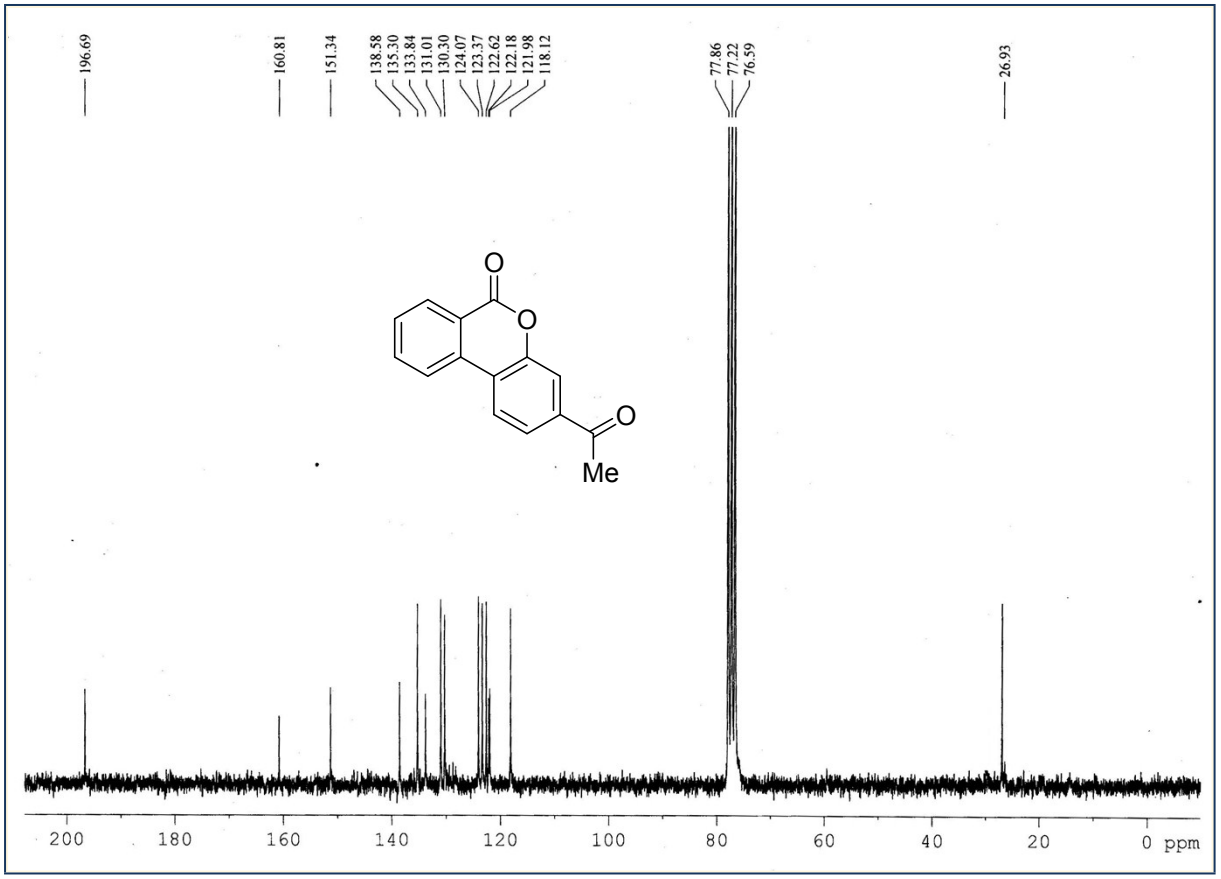
^{13}C NMR of compound 5h



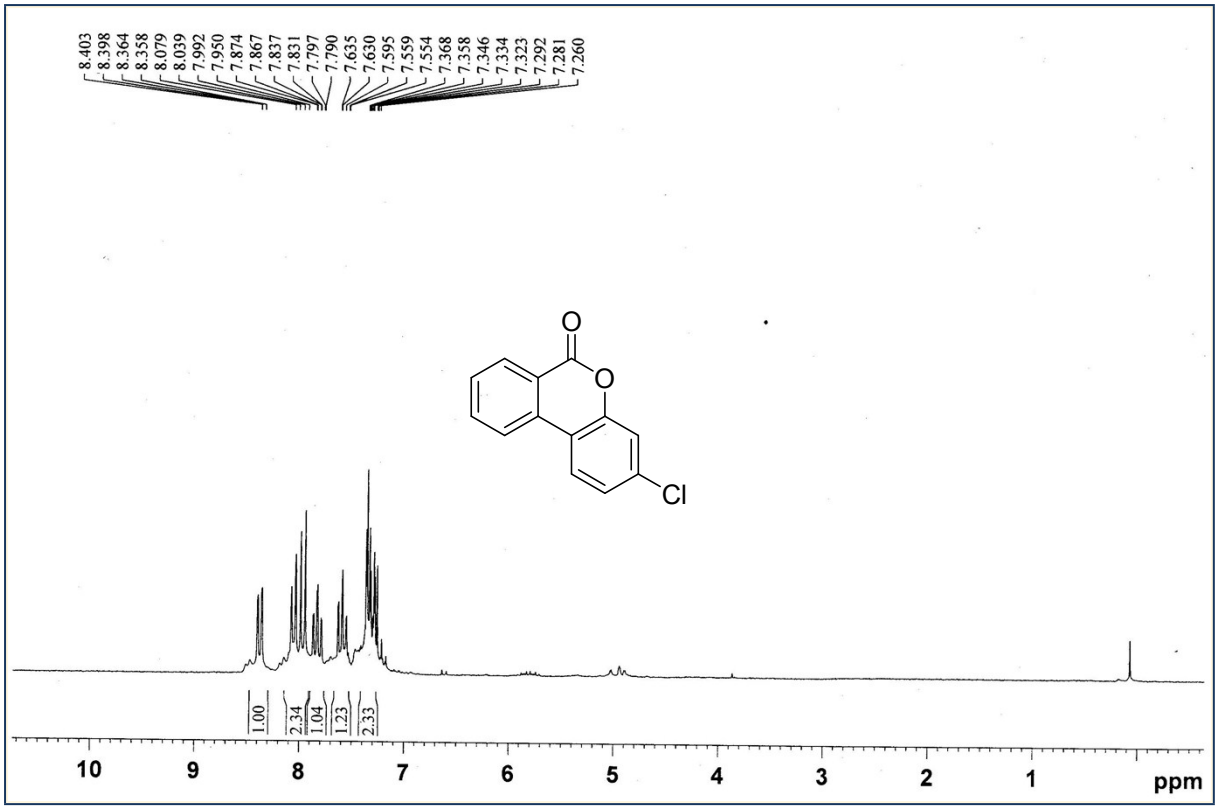
^{1}H NMR of compound 5i



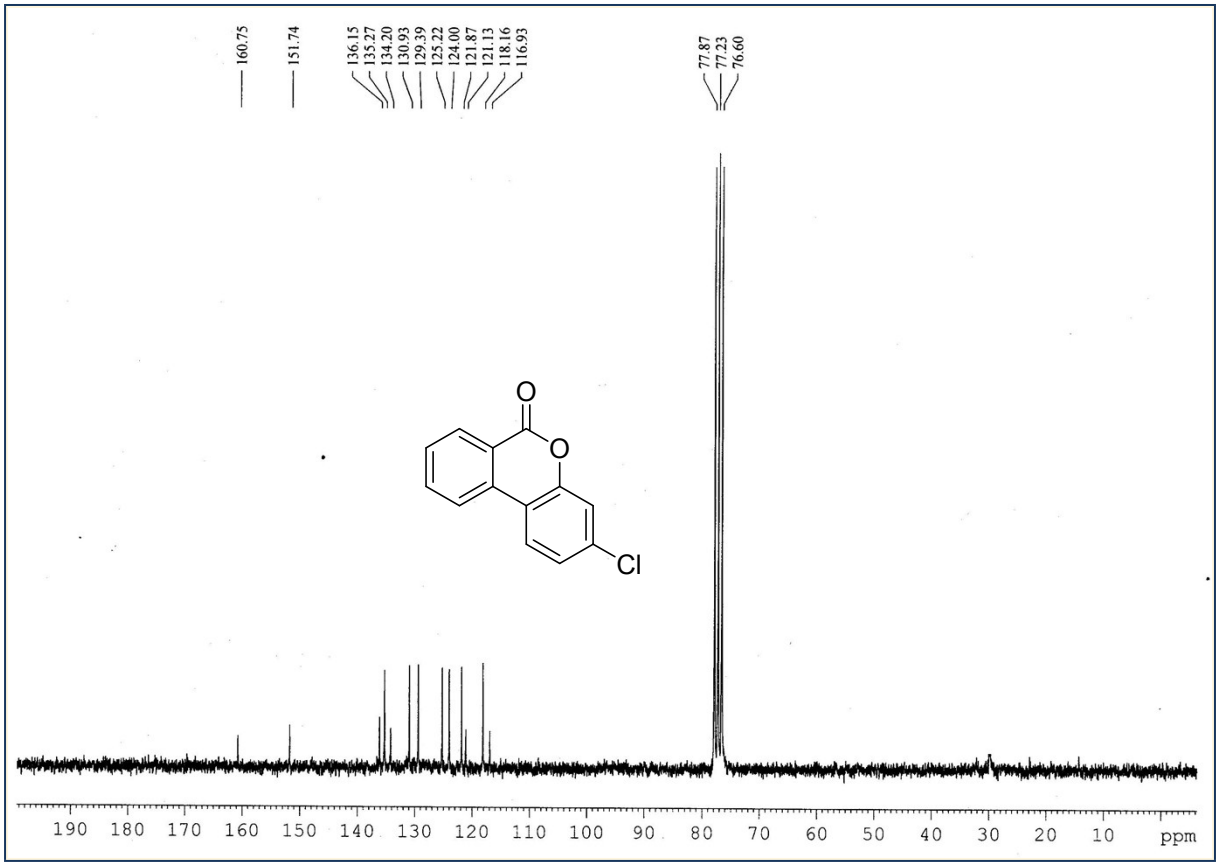
^{13}C NMR of compound 5i



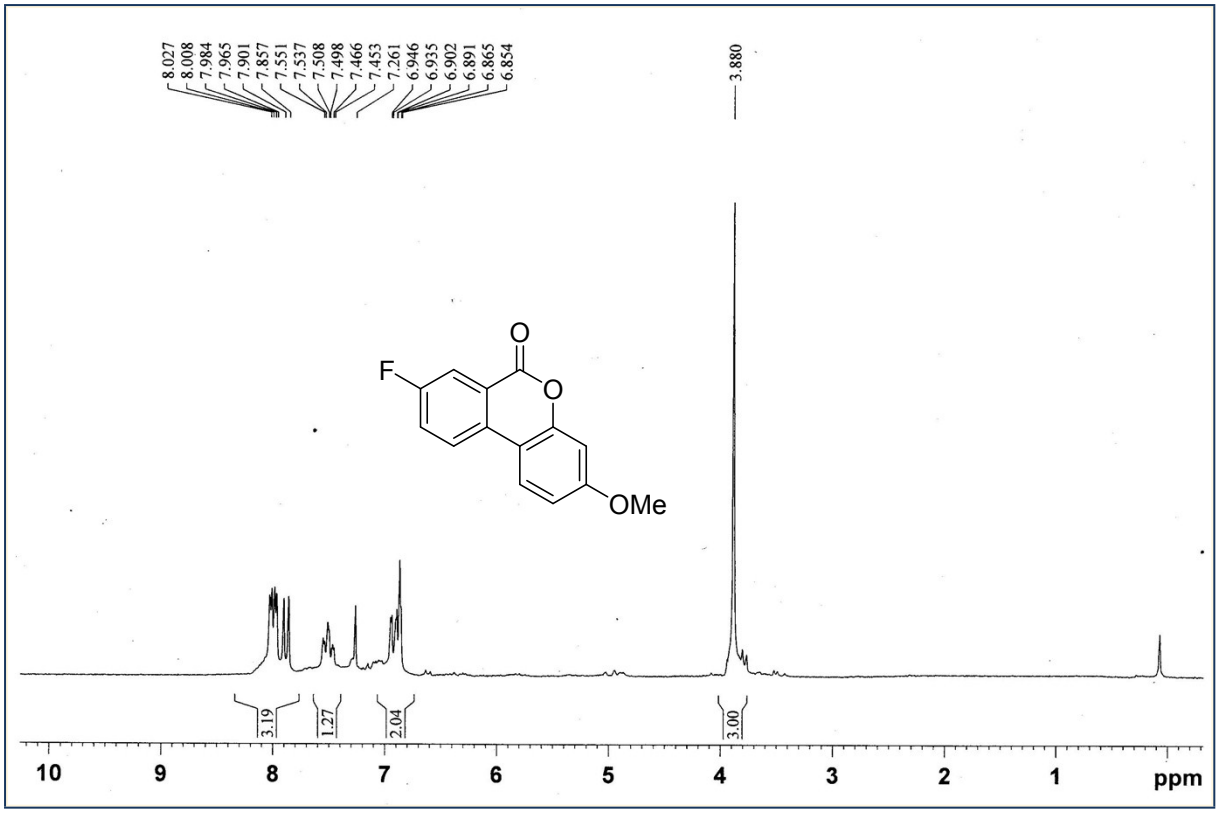
^{1}H NMR of compound 5j



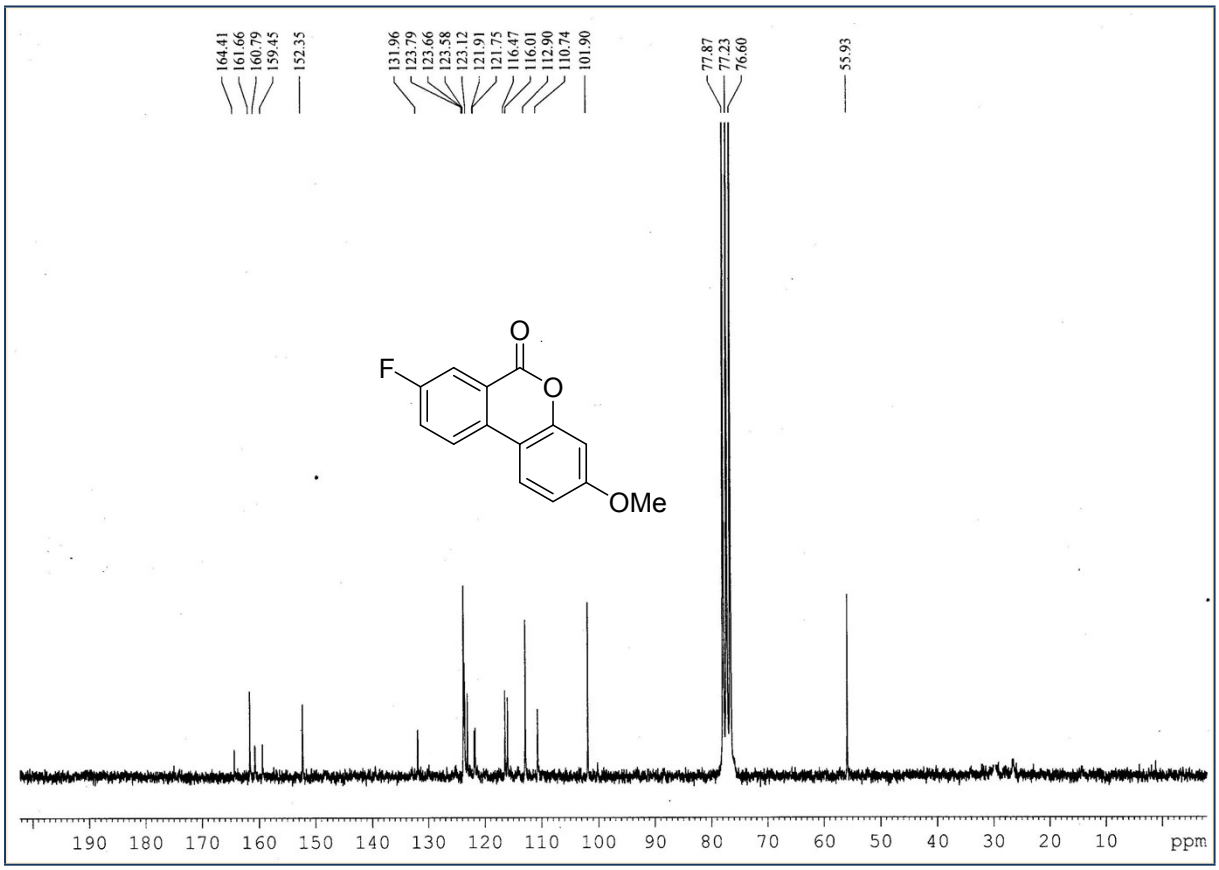
^{13}C NMR of compound 5j



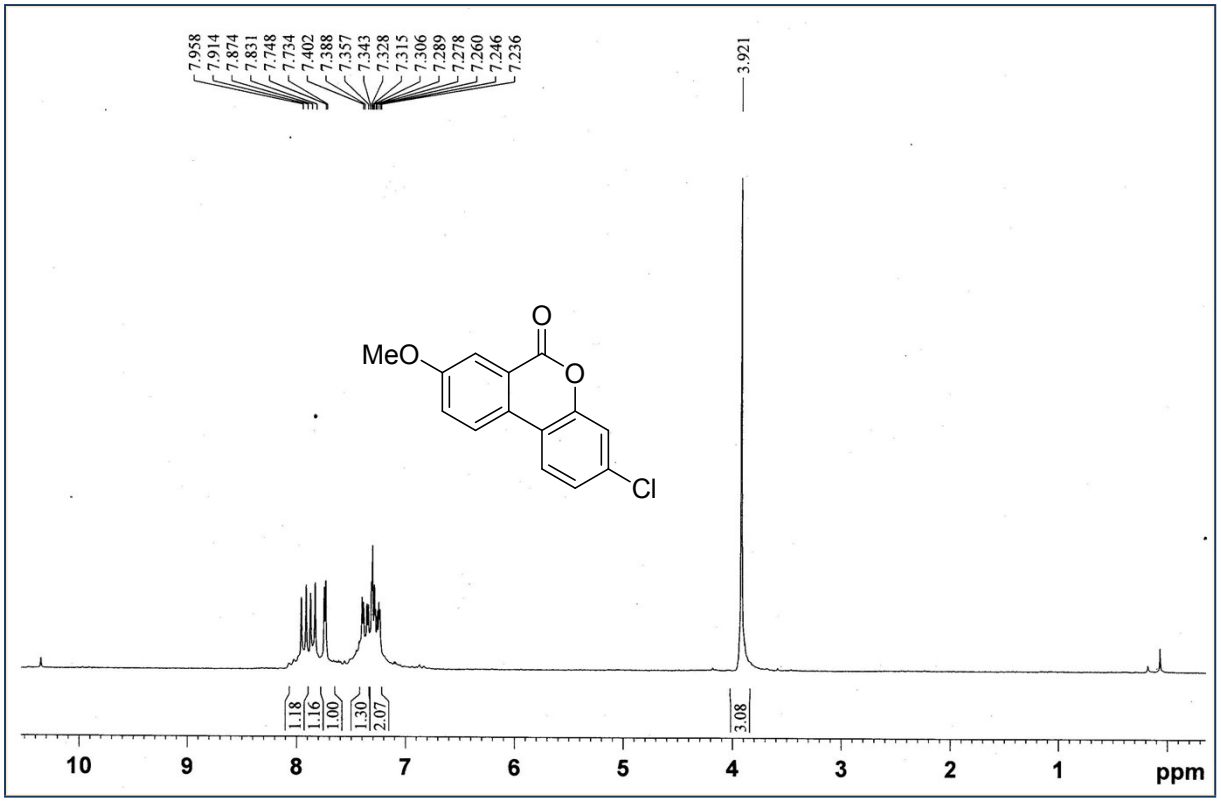
^{1}H NMR of compound 5k



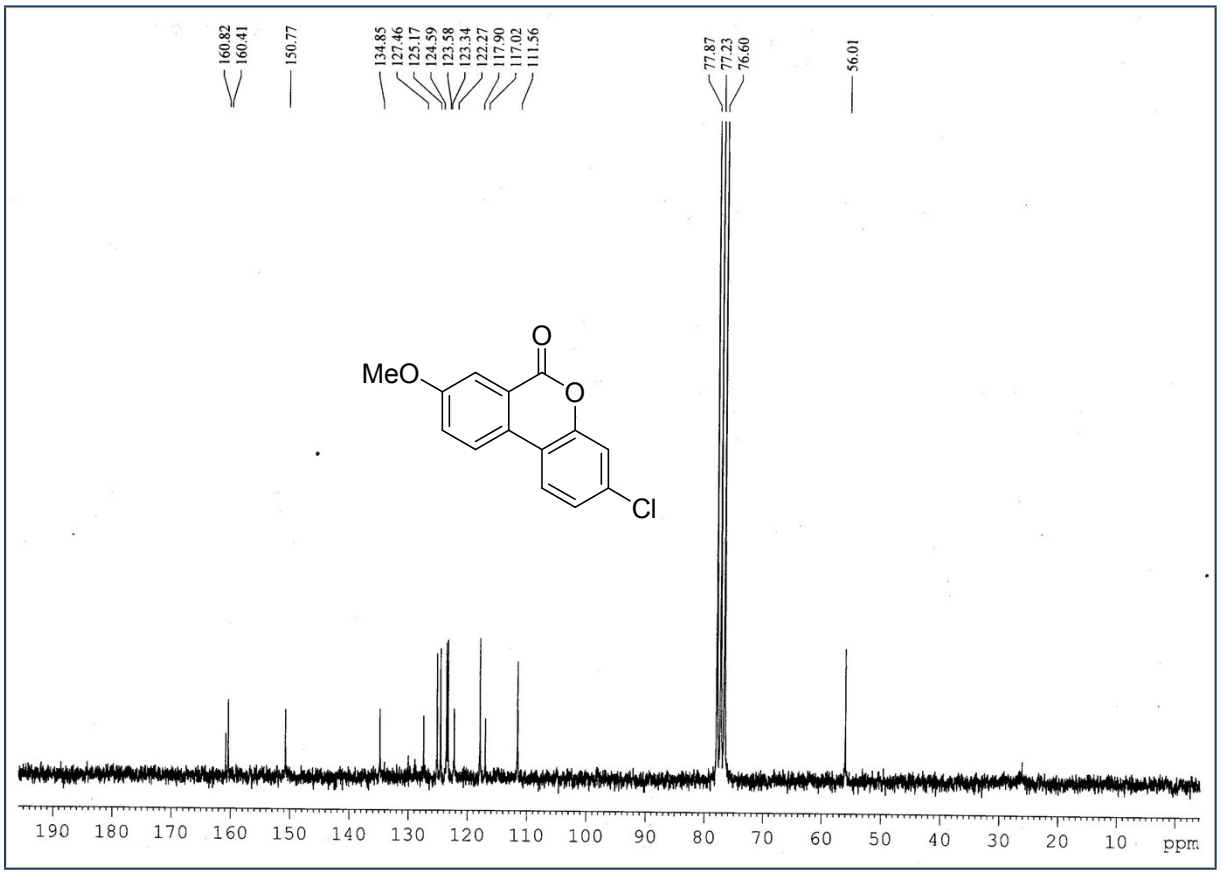
^{13}C NMR of compound 5k



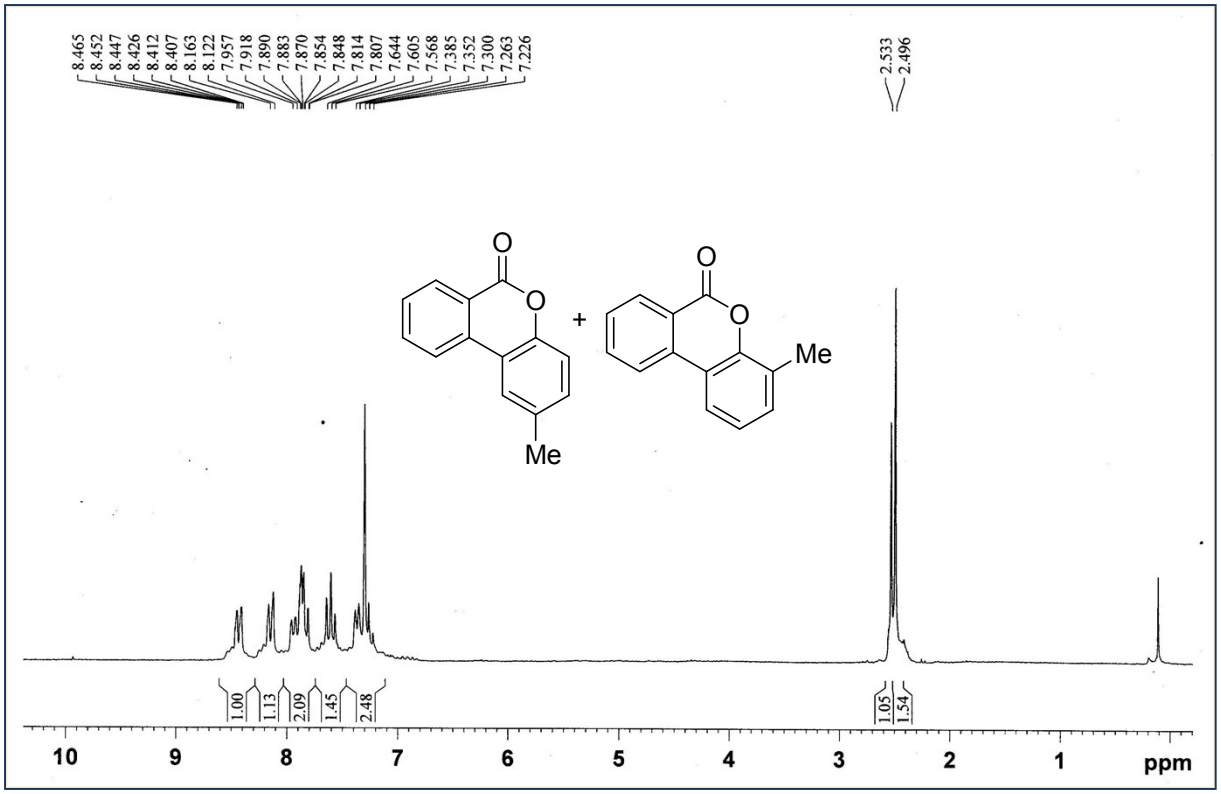
^{1}H NMR of compound 5l



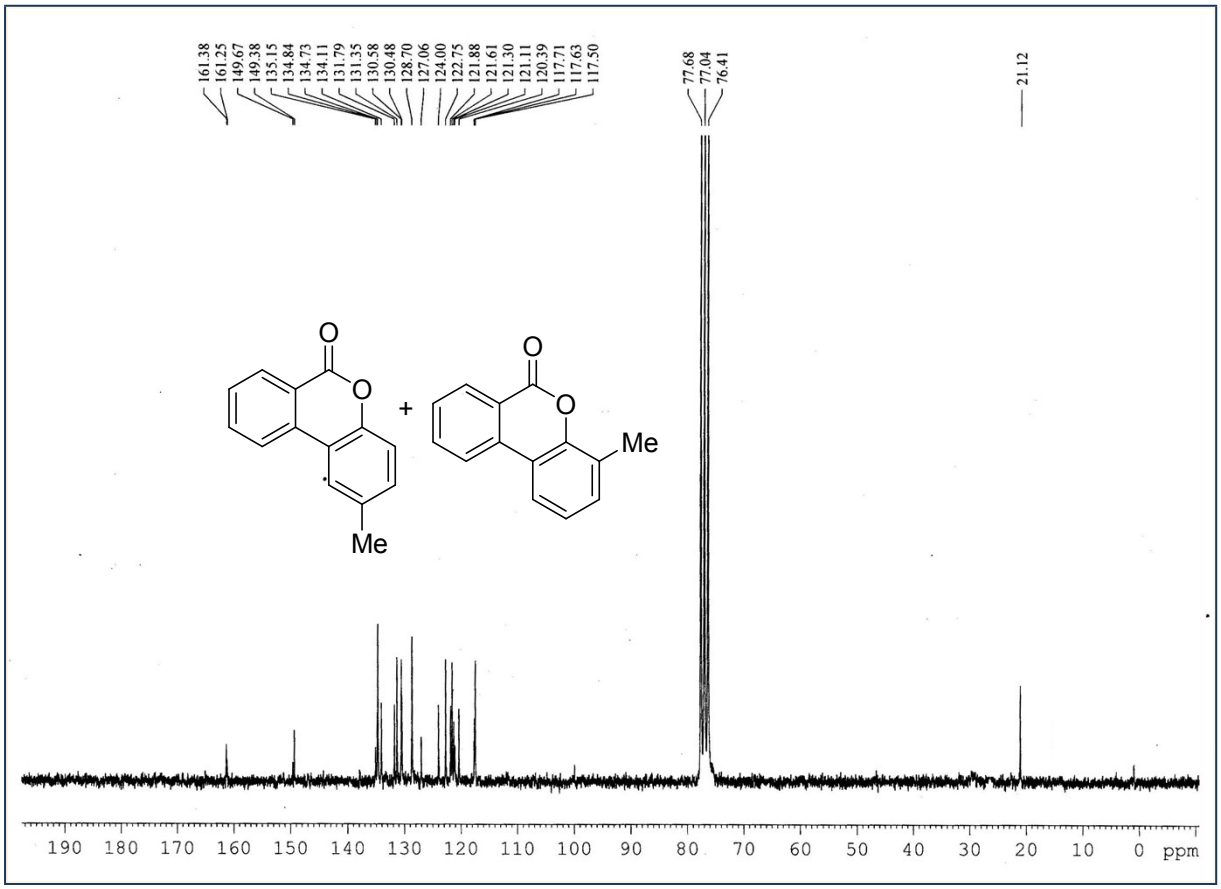
^{13}C NMR of compound 5l



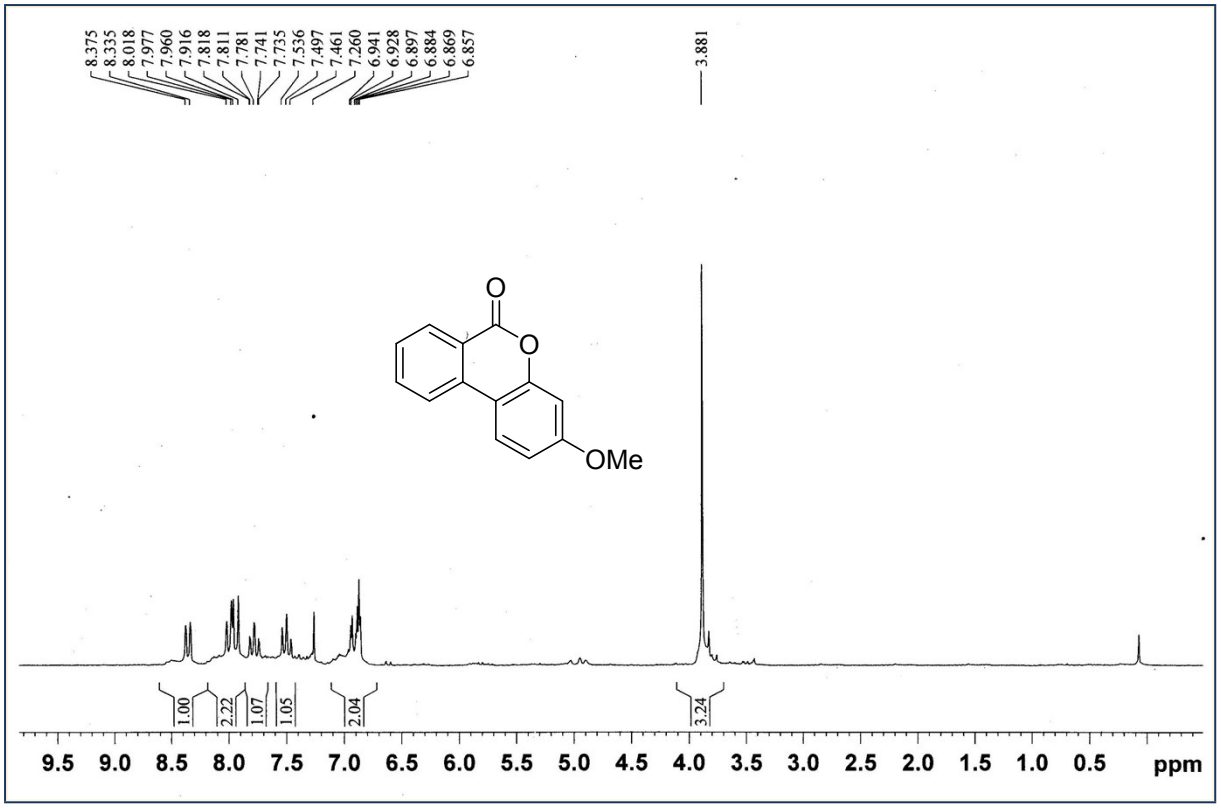
^{1}H NMR of compound 5n



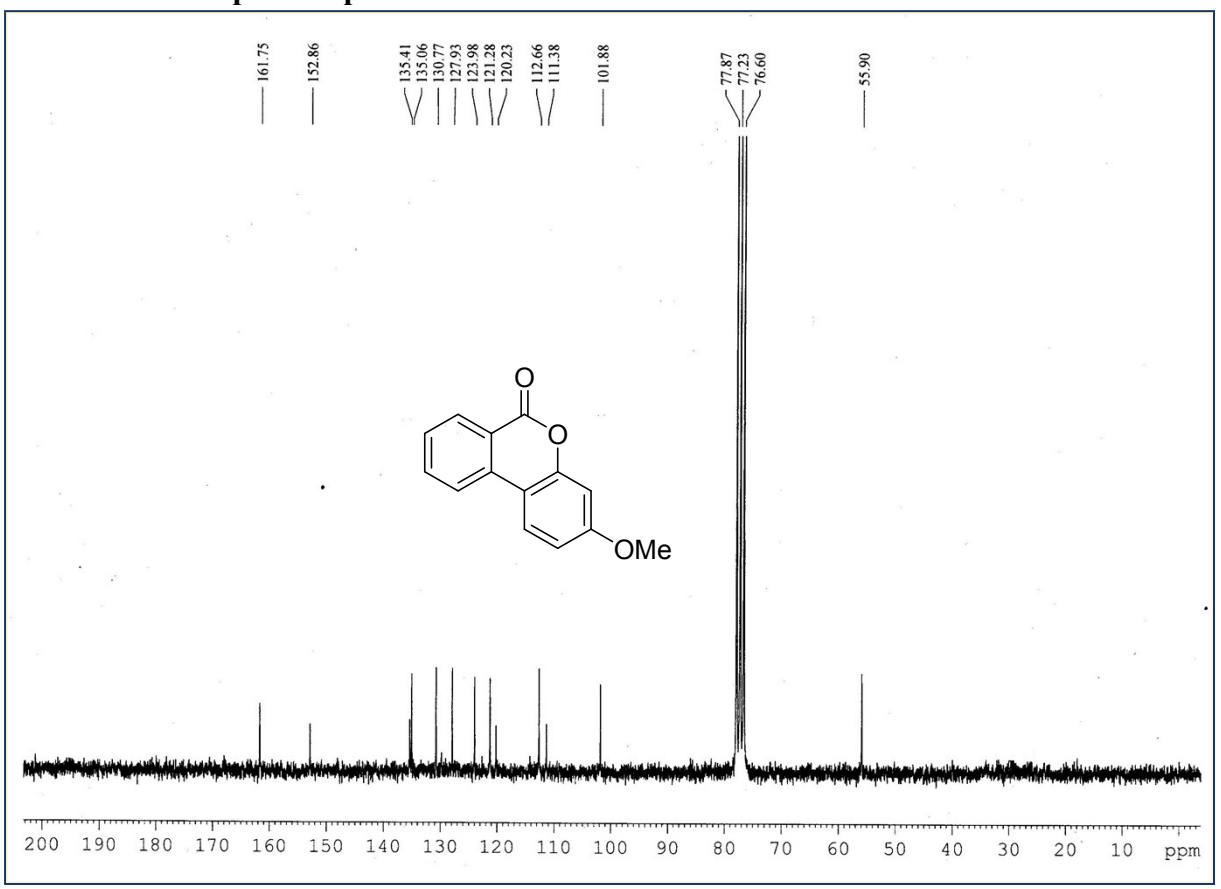
^{13}C NMR of compound 5n



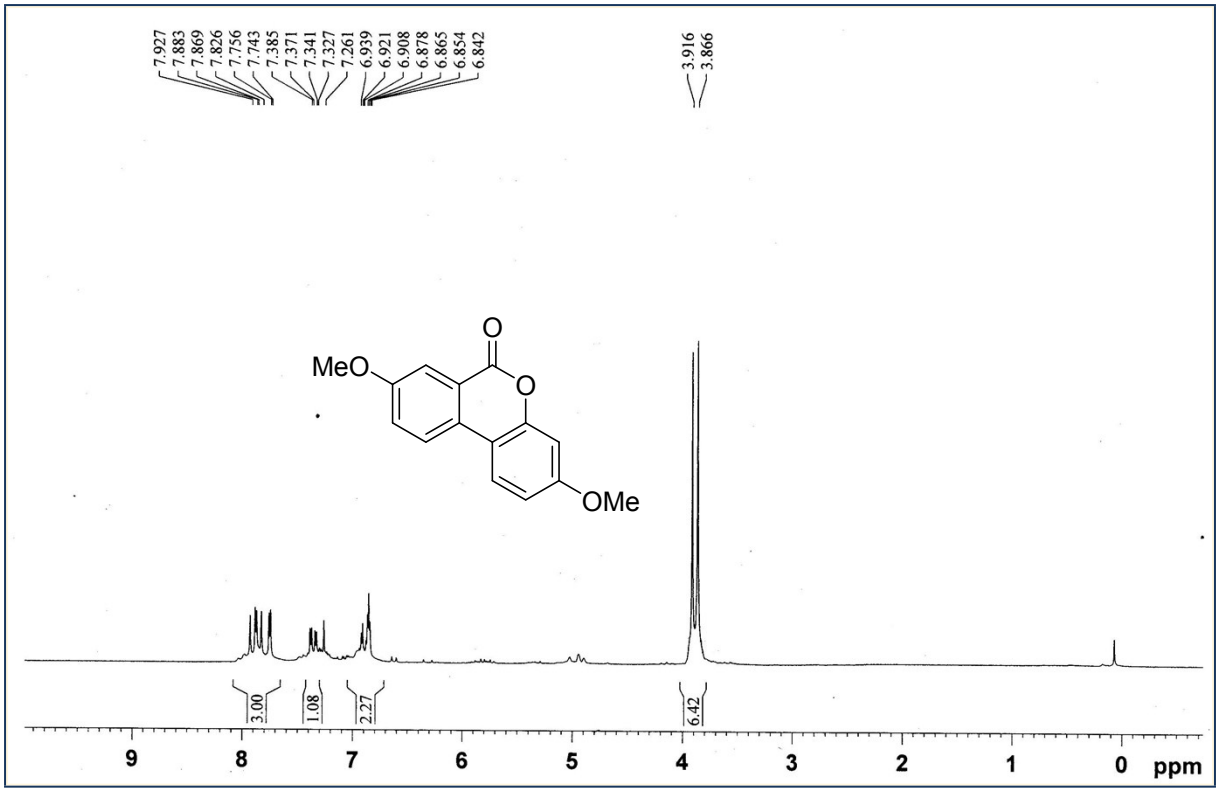
^{1}H NMR of compound 5p



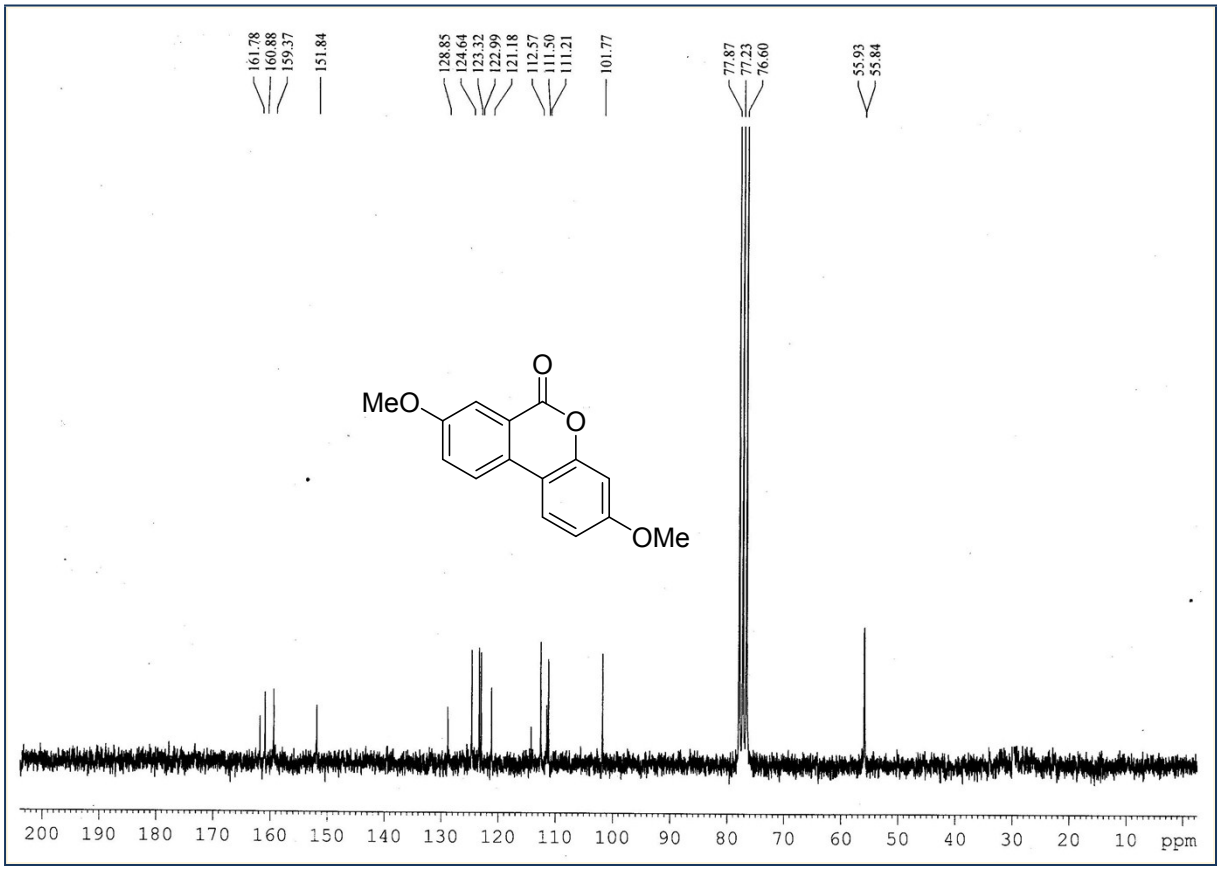
^{13}C NMR of compound 5p



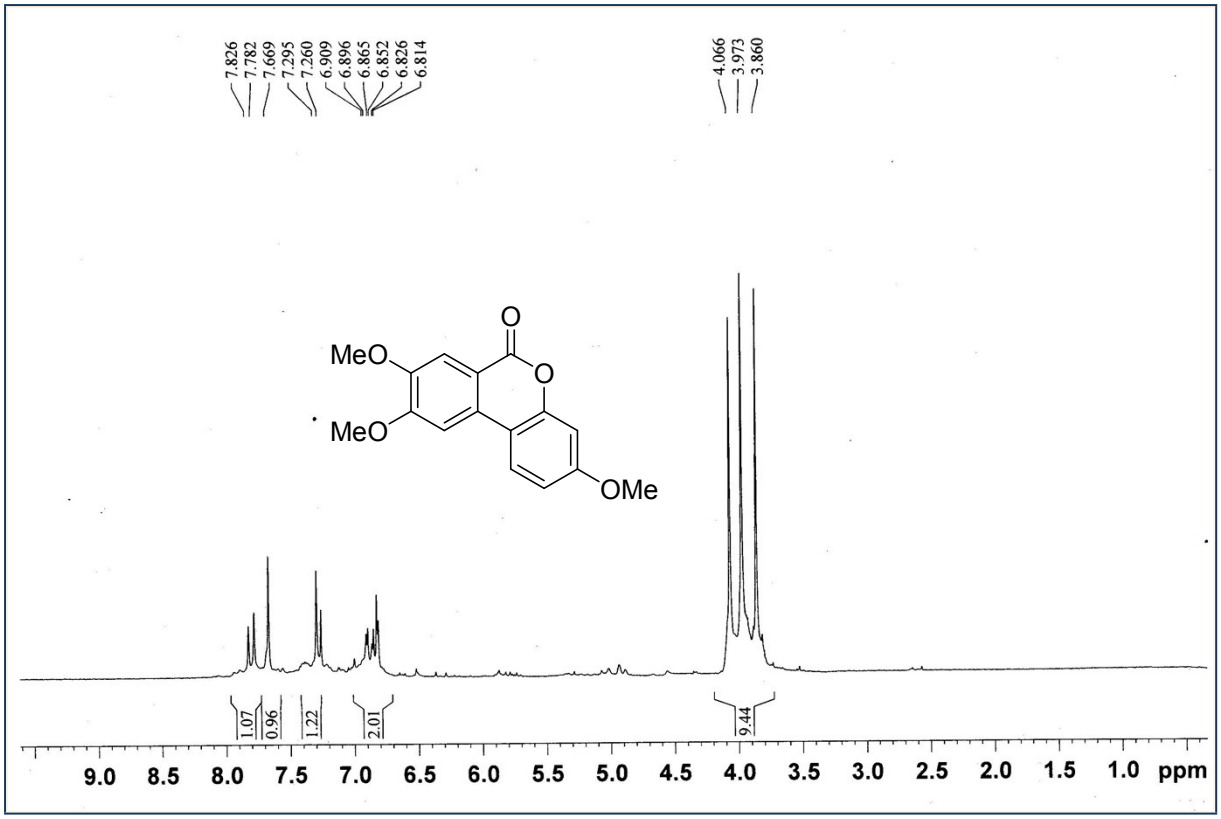
^{1}H NMR of compound 5q



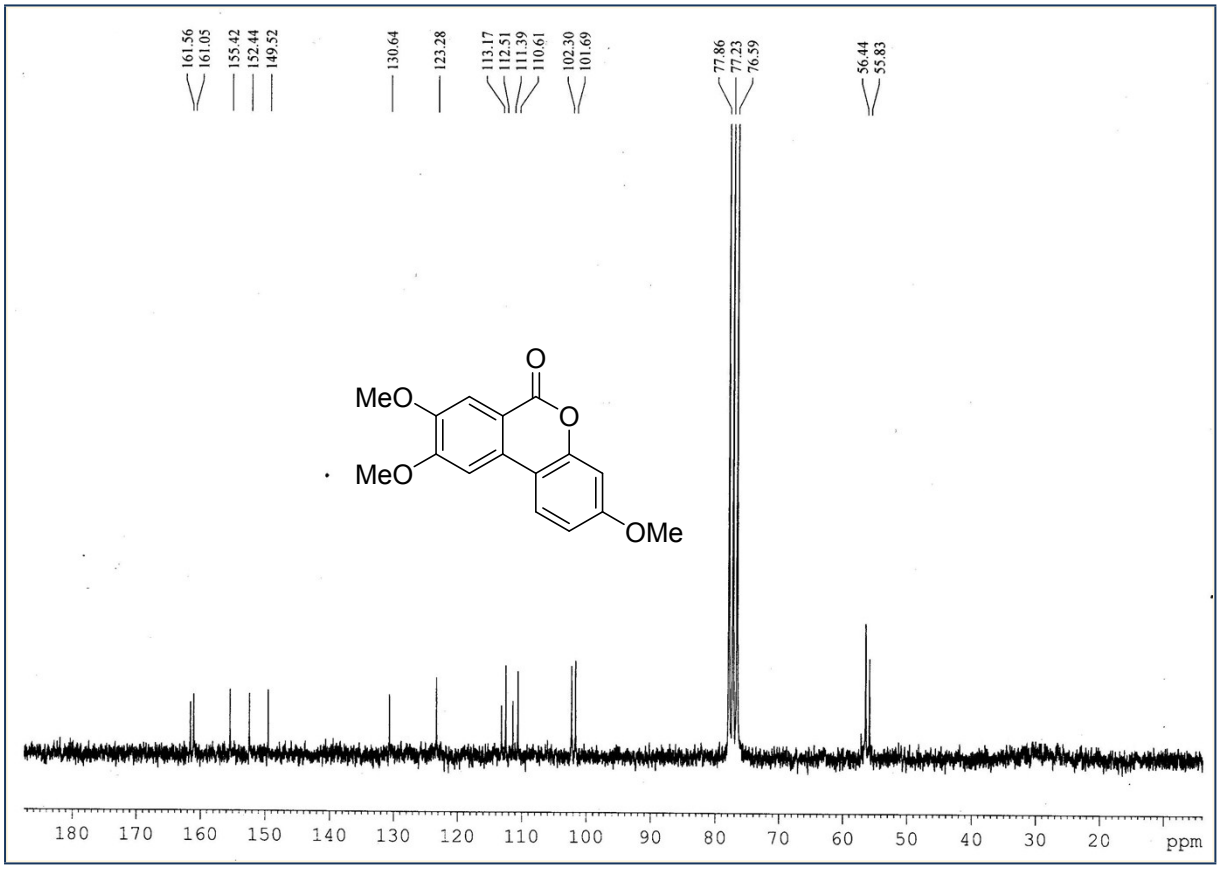
^{13}C NMR of compound 5q



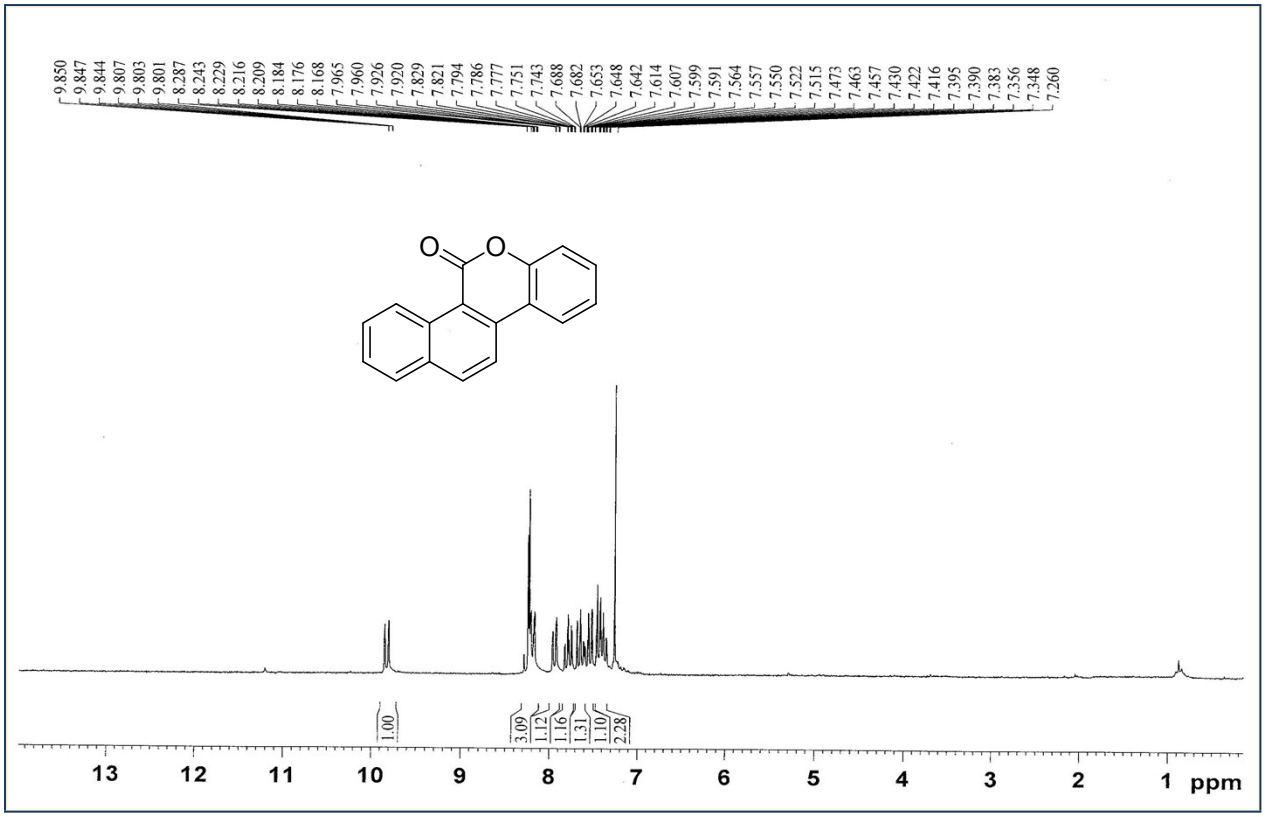
^{1}H NMR of compound 5r



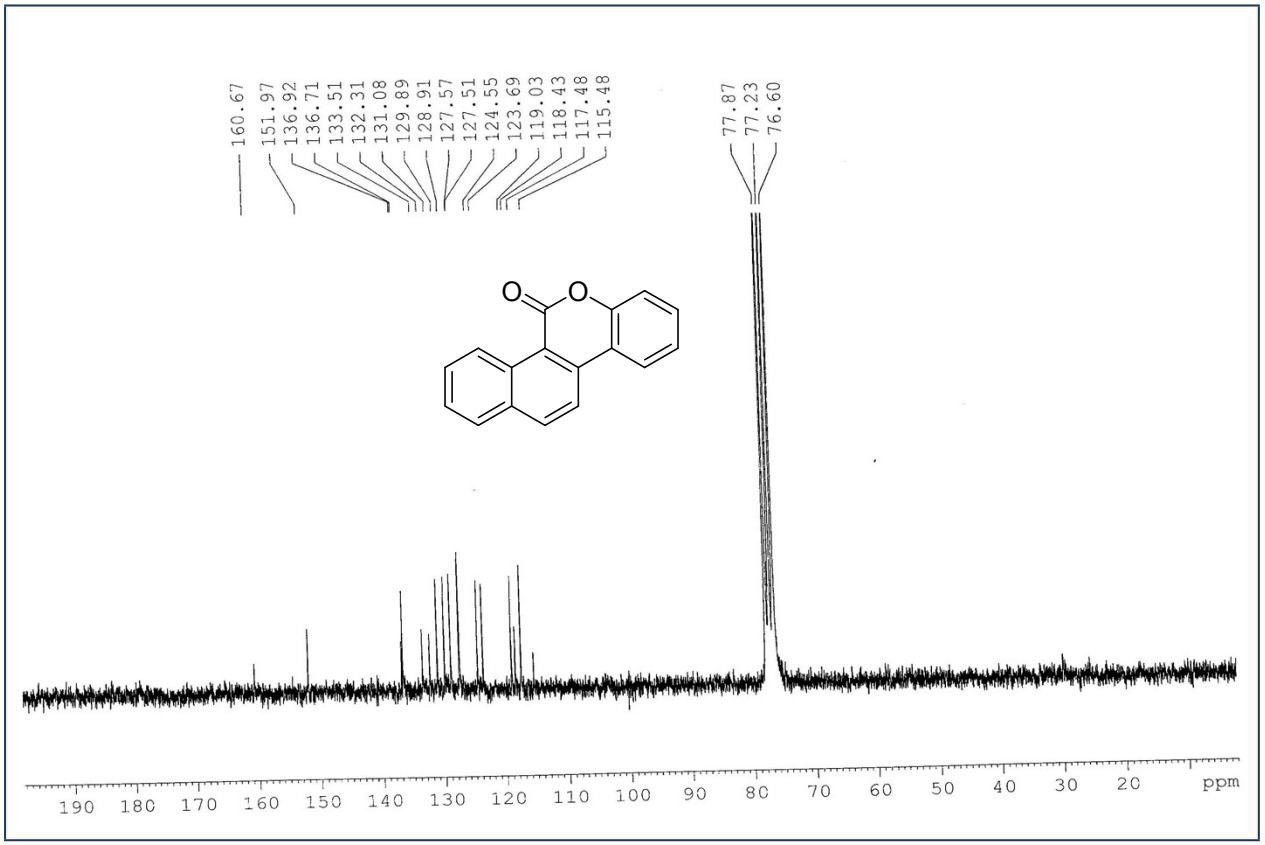
^{13}C NMR of compound 5r



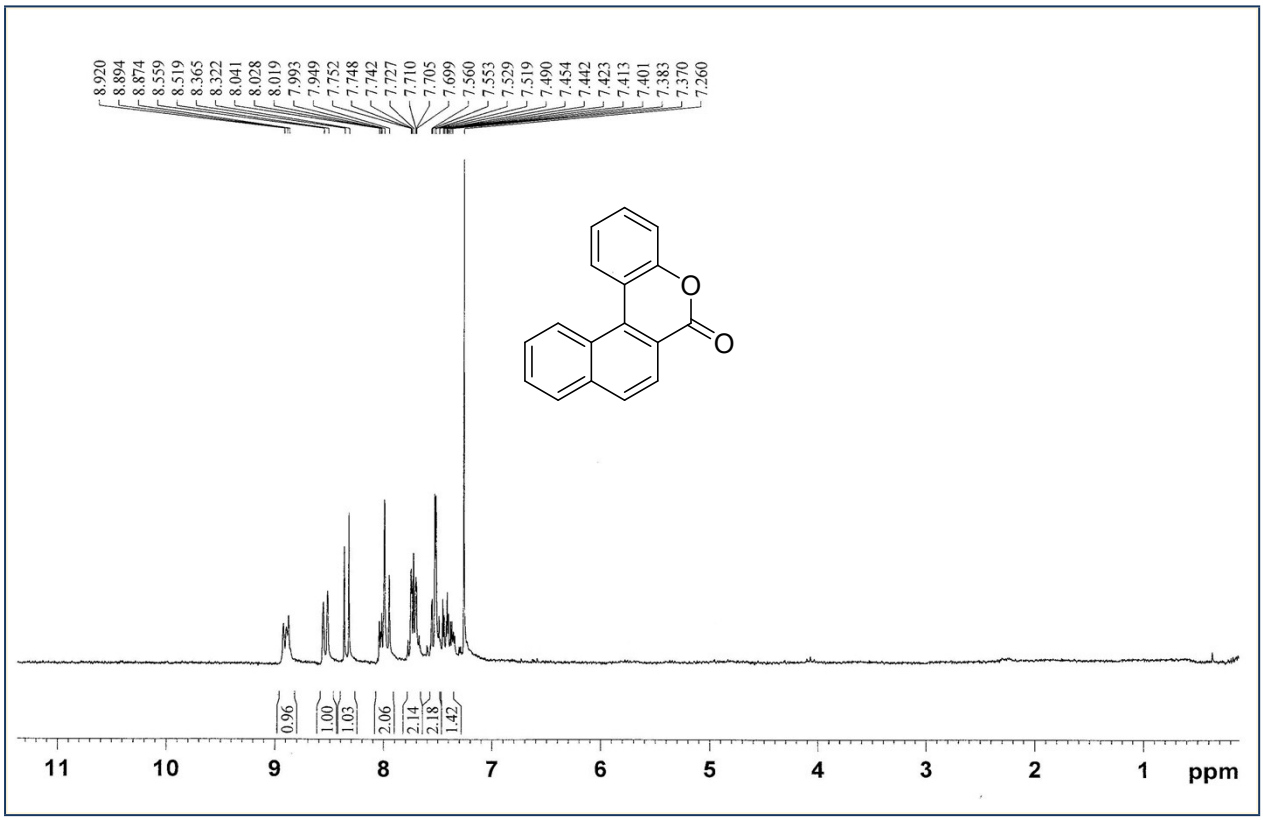
^{1}H NMR of compound 5s



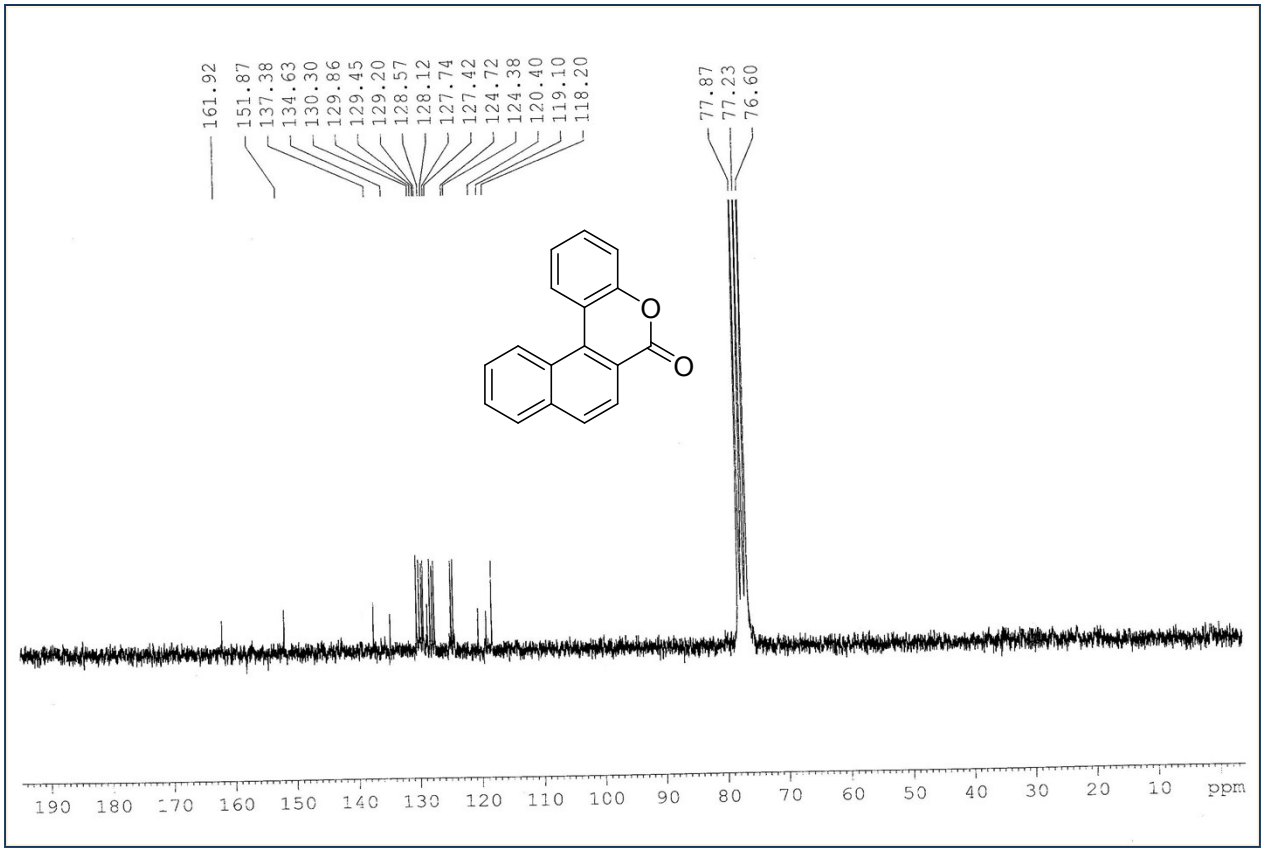
^{13}C NMR of compound 5s



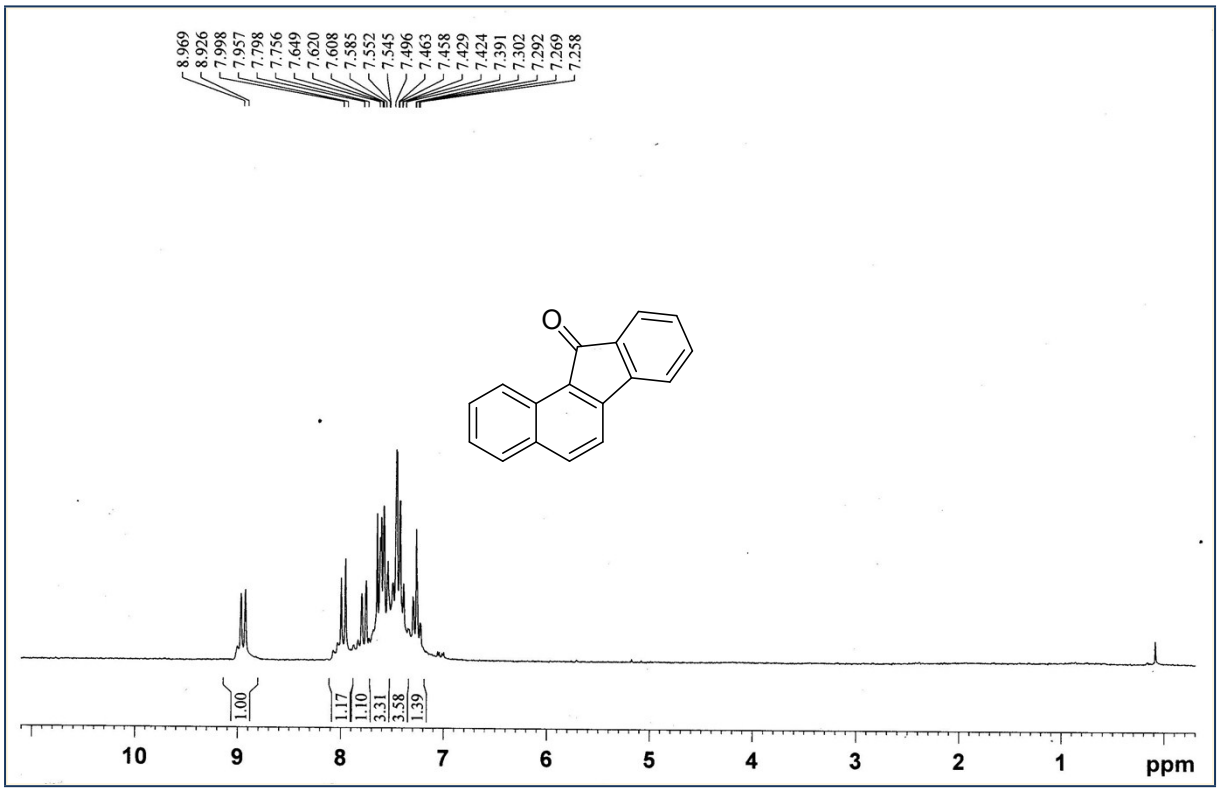
^{1}H NMR of compound 5t



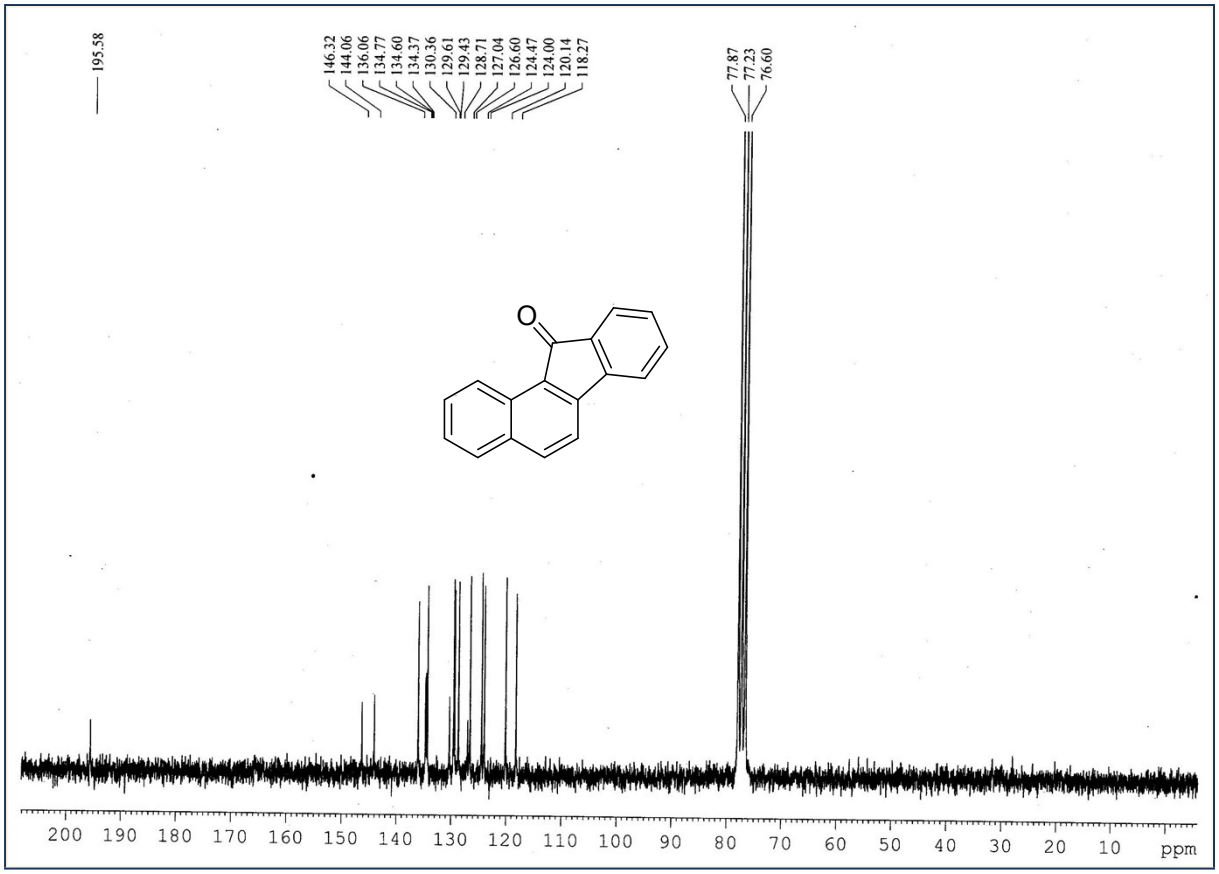
^{13}C NMR of compound 5t



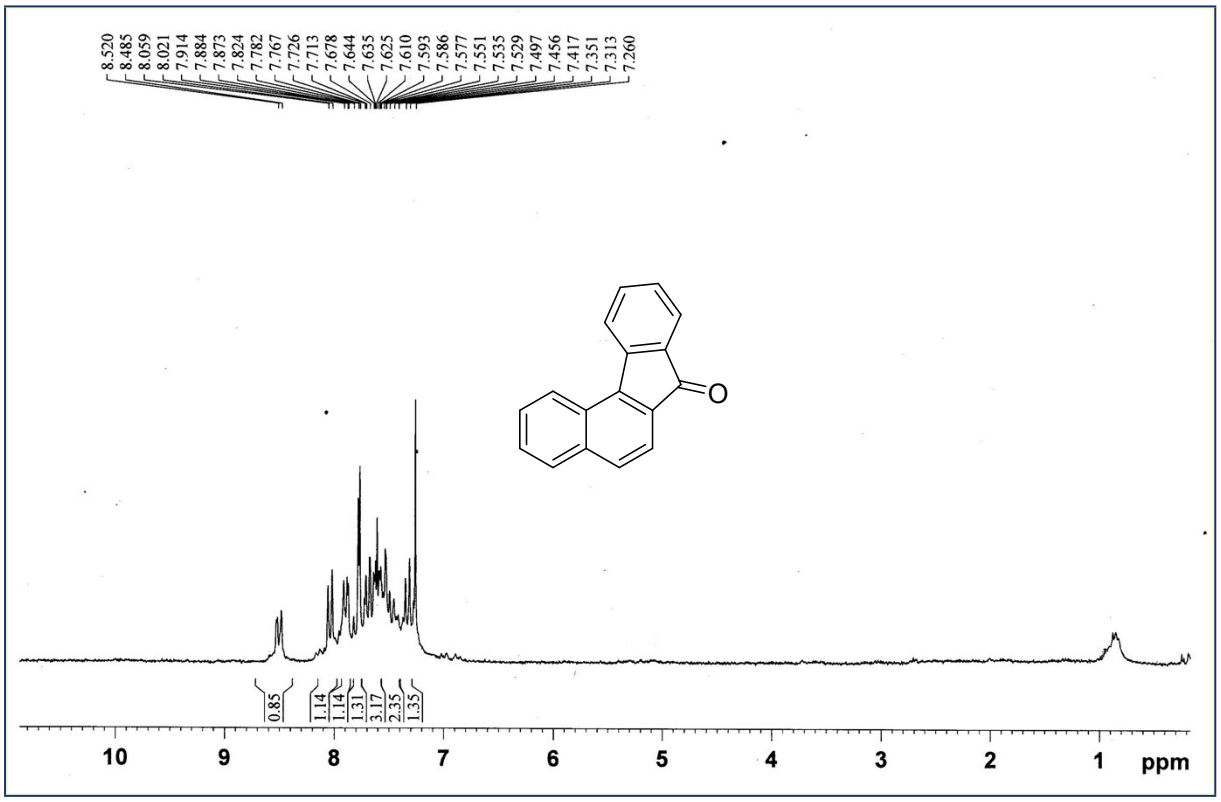
^{1}H NMR of compound 6a



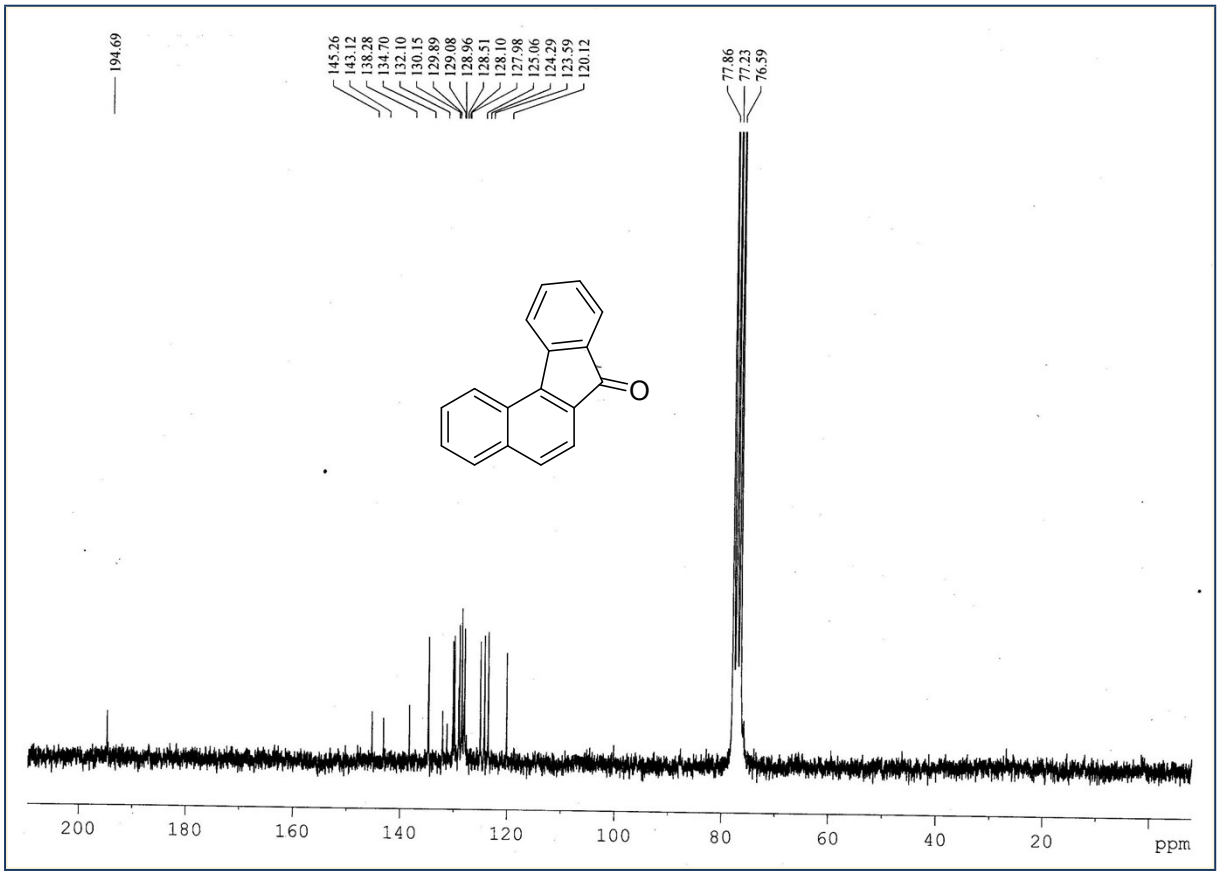
^{13}C NMR of compound 6a



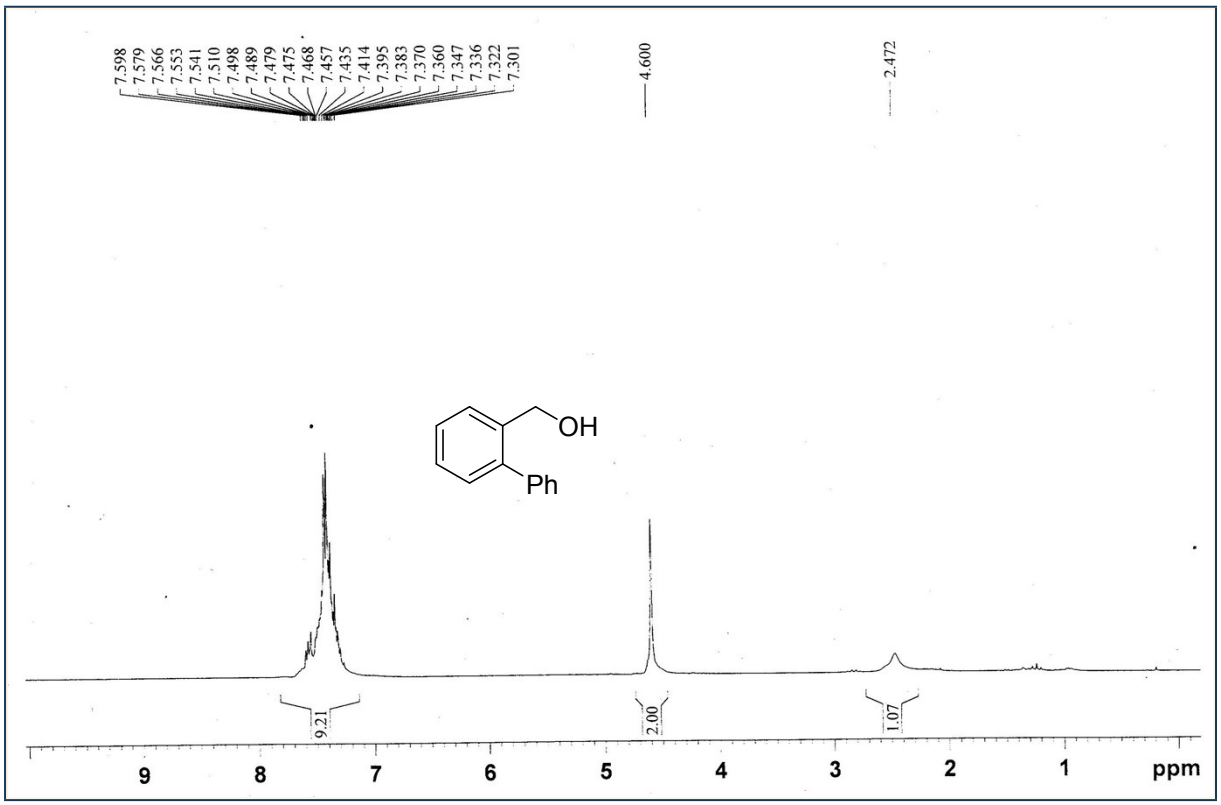
^{1}H NMR of compound 6b



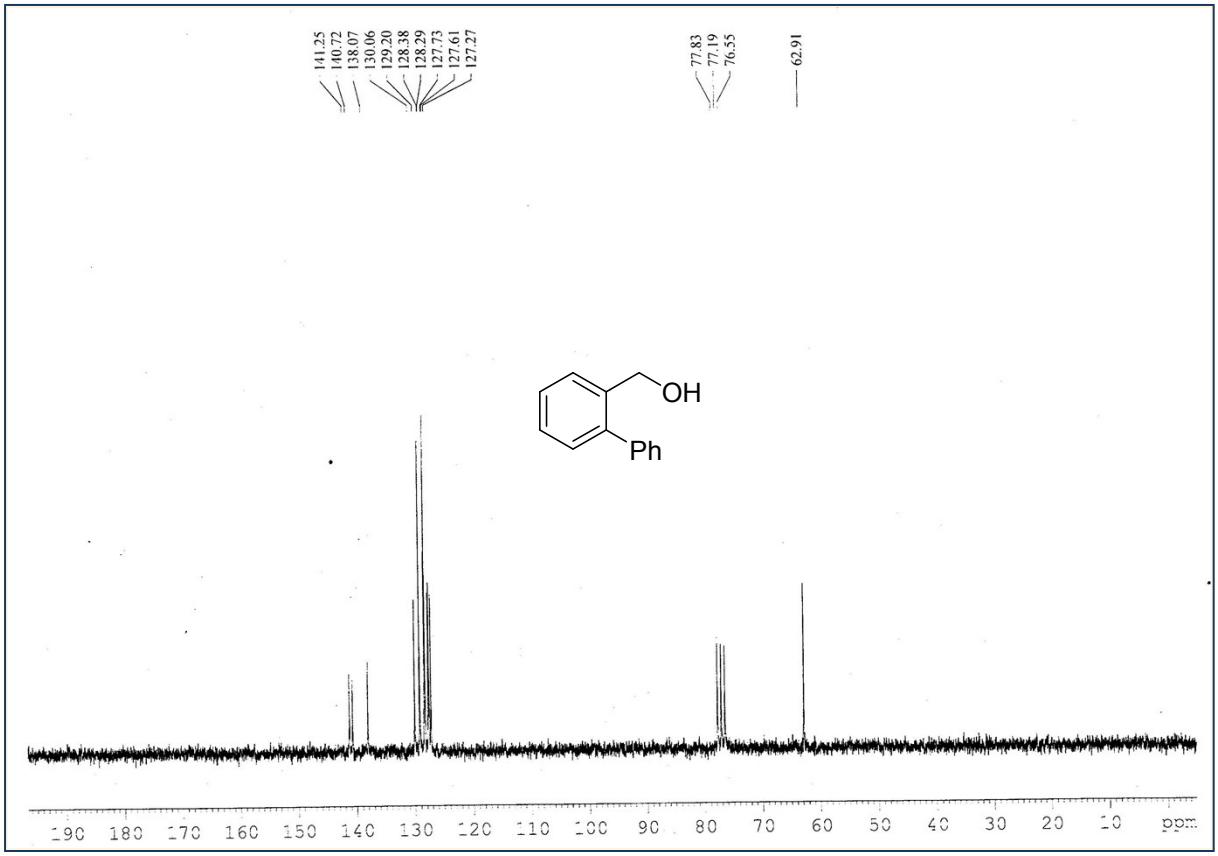
^{13}C NMR of compound 6b



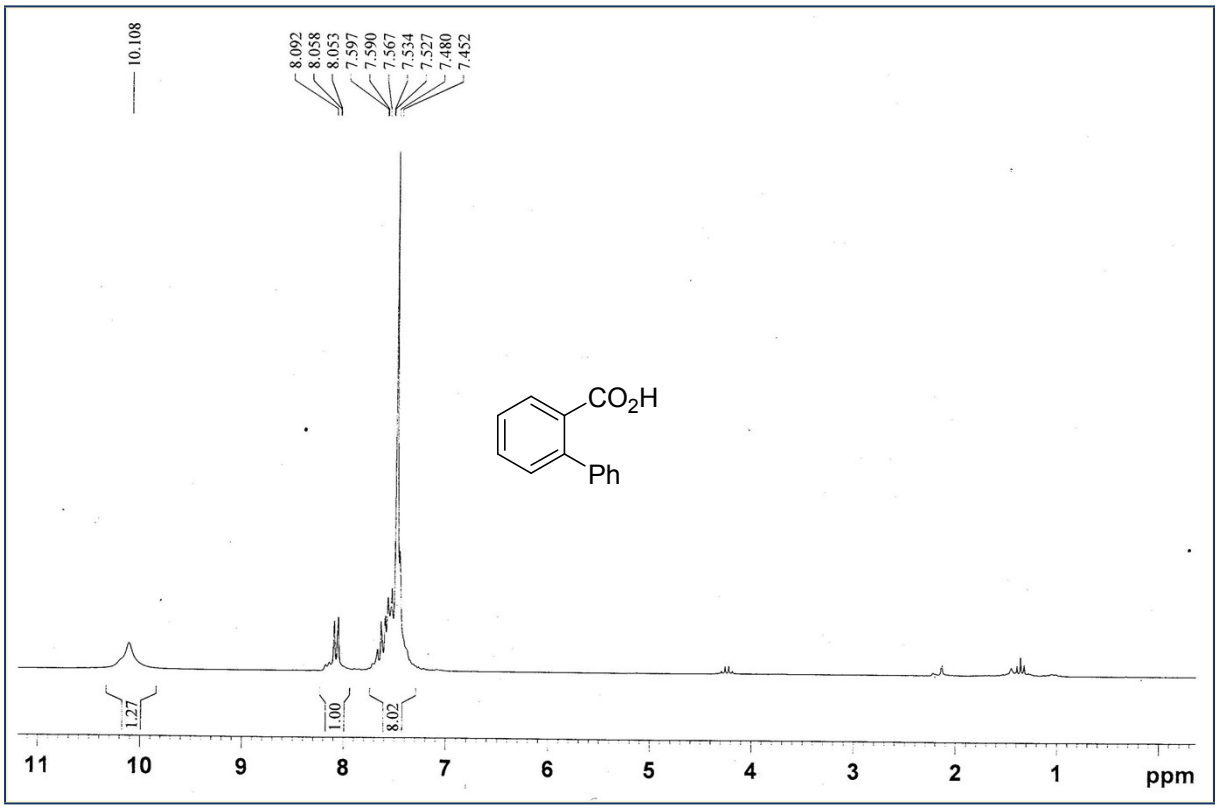
[1]H NMR of compound 7



[13]C NMR of compound 7



^{1}H NMR of compound 8



^{13}C NMR of compound 8