Supporting Information for

Acetal initiated Prins bicyclization for the synthesis of furo[3,4-*c*]furan lignans and pyrano[3,4-*c*]pyran derivatives

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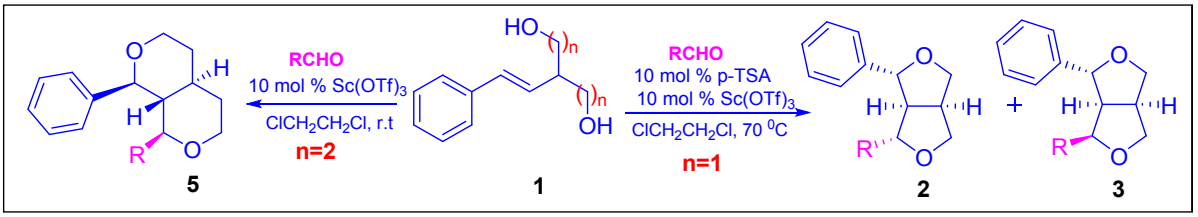

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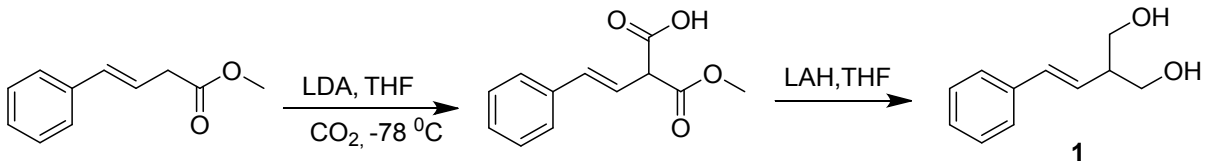
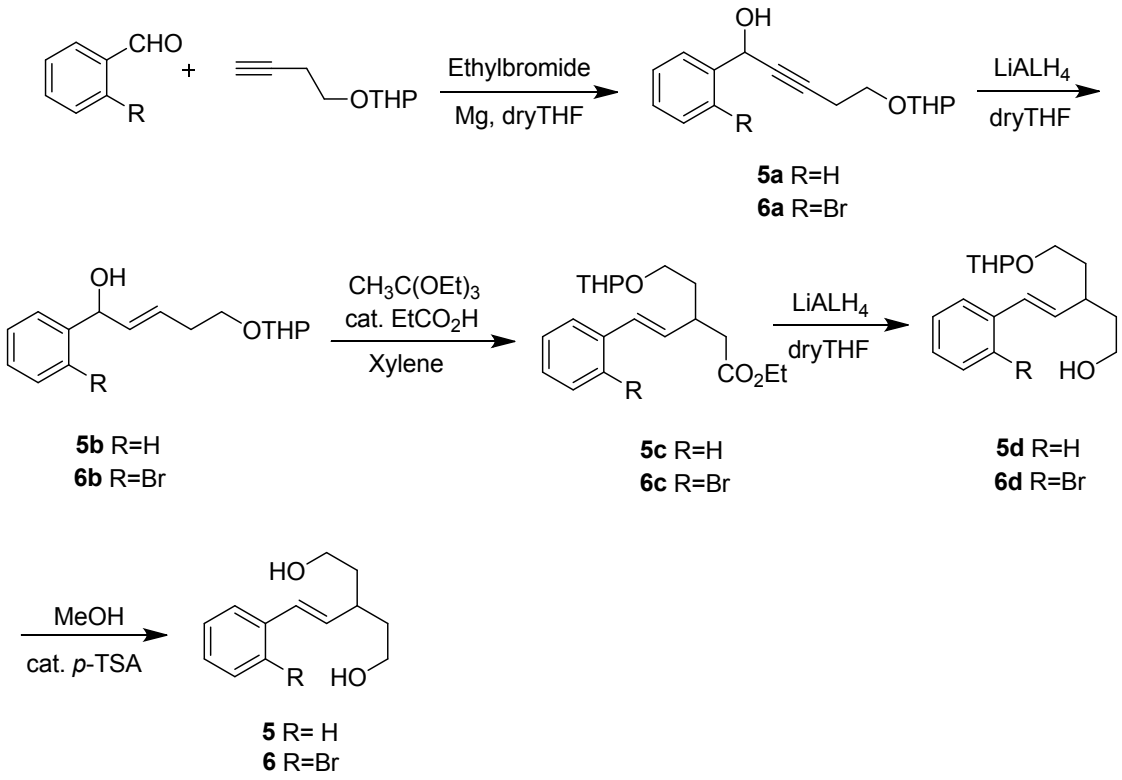
1. Preparation of starting materials:

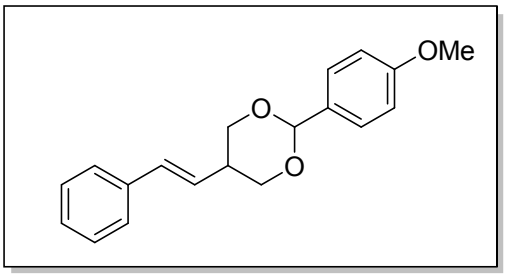
Scheme S1: Synthesis of (*E*)-2-styrylpropane-1,3-diol (1):[1]



Scheme S2: Synthesis of (*E*)-3-styrylpentane-1,5-diols (5&6):



Procedure for the synthesis of (*E*)-2-(4-methoxyphenyl)-5-styryl-1,3-dioxane (1a):

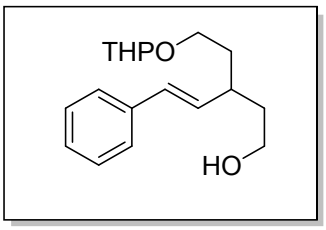


To a stirred solution of (*E*)-2-styrylpropane-1,3-diol (196 mg, 1mmol), and PMB-acetal (180 mg, 1mmol) in DCM (5 mL) at 0 ^{0}C was added a catalytic amount of CSA. The mixture was stirred 1 h at room temperature. After completion of the reaction, solid $NaHCO_3$ was added and the stirring was continued for 30 min. The solvent was removed under vacuum,

and a crude mixture was diluted with 10 mL CH_2Cl_2 and further washed with water and then concentrated to give the residue which was purified by silica gel chromatography to give the compound **1a** (260 mg, 90%) as a semi solid.

Semi solid; ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.46-7.40 (m, 3H), 7.36-7.28 (m, 3H), 6.93-6.87 (m, 2H), 6.53 (d, *J* = 16 Hz, 1H), 5.88 (dd, *J* = 8.0, *J* = 16 Hz, 1H), 5.43 (s, 1H), 4.30-4.09 (m, 2H), 3.81 (s, 6H), 2.33-2.29 (m, 1H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$): δ 160.0, 136.6, 133.0, 128.5, 127.7, 127.3, 126.1, 124.7, 113.6, 101.1, 71.4, 55.2, 38.5 ppm; MS (ESI): *m/z* ($[M+H]^+$): 297.

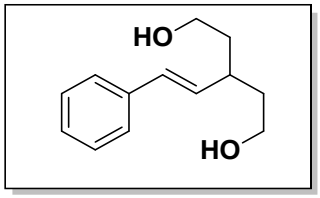
Procedure for the synthesis of (*E*)-5-phenyl-3-(2-(tetrahydro-2*H*-pyran-2-yloxy)ethyl)pent-4-en-1-ol (5d):



To a suspension of $LiAlH_4$ (228 mg, 6 mmol, 1eq.) in dry THF (6 mL) was added a solution of **5c** (1.9 g, 6.02 mmol) in 8 mL THF slowly at 0 °C under nitrogen atmosphere. The mixture was stirred 2 h at room temperature. After completion, a sat. solution of Na_2SO_4 was added slowly to quench the excess $LiAlH_4$ at 0 °C. The mixture was stirred for another 30 min and then filtered. The solid was washed with hot EtOAc (3x10 mL). The combined EtOAc solutions were dried over $MgSO_4$. The solvent was removed under vacuum and the residue was purified by silica gel chromatography to give the compound **5d** (1.5 g, 95%) as a liquid.

^{1}H NMR (500 MHz, $CDCl_3$): δ 7.36-7.18 (m, 10H), 6.44-6.37 (m, 1H), 5.99 (dd, *J* = 9.0, *J* = 15.8 Hz, 1H), 4.58-4.51 (m, 1H), 3.89-3.64 (m, 4H), 3.50-3.38 (m, 2H), 2.57-2.48 (m, 1H), 1.86-1.46 (m, 10H) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ 136.5, 133.1, 129.8, 127.8, 126.4, 125.3, 99.2, 98.5, 65.8, 65.7, 62.7, 62.3, 61.0, 38.6, 38.5, 37.7, 37.5, 35.7, 35.6, 31.3, 31.2, 26.0, 20.4, 20.2 ppm; MS (ESI): *m/z* ($[M+H]^+$): 291.

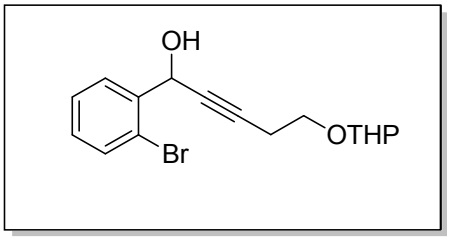
Procedure for the synthesis of (*E*)-3-styrylpentane-1,5-diol (5):



To a stirred solution of **5d** (290 mg, 1 mmol) in MeOH (3 mL) was added a catalytic amount CSA at 0 ^{0}C. The mixture was stirred 1h at room temperature. Up on completion, solid $NaHCO_3$ was added and then stirred it for another 30 min. The solvent was removed under vacuum and the resulting residue was diluted with 10 mL CH_2Cl_2 and further washed water, and then concentrated in vacuo to give the crude product which was purified by silica gel chromatography to give the compound **5** (180mg, 90%) as a liquid.

Liquid; ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.35-7.12 (m, 4H), 6.47 (d, *J* = 14.4 Hz, 1H), 5.93 (dd, *J* = 9.5, *J* = 15.8 Hz, 1H), 3.77-3.66 (m, 4H), 2.84-2.55 (m, 2H), 1.77-1.49 (m, 4H) ppm; ^{13}C NMR (75 MHz, $CDCl_3$): δ 136.4, 132.9, 129.9, 127.8, 126.5, 125.4, 62.5, 60.5, 38.2, 36.8 ppm; MS (ESI): *m/z* ([M+H]$^+$): 207.

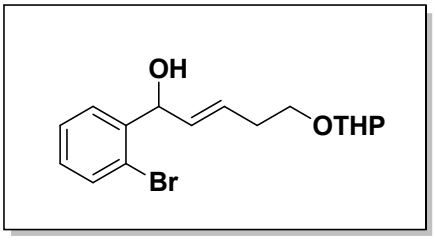
Procedure for the synthesis of 1-(2-bromophenyl)-5-(tetrahydro-2*H*-pyran-2-yloxy)pent-2-yn-1-ol (6a):



To a suspension of Mg turnings (26 mg, 1.2 mmol) in dry THF (2 mL) was added a solution of bromoethane (107 mg, 1 mmol) in 5 mL THF slowly under nitrogen atmosphere. After complete addition, the mixture was stirred for 1h after which, a solution of 2-(but-3-ynyloxy)tetrahydro-2*H*-pyran (154 mg, 1 mmol) in 5 mL THF was added slowly at 0 ^{0}C under nitrogen atmosphere. To this mixture, a solution of 2-bromobenzaldehyde (180 mg, 1mmol) in dry THF (5ml) was added dropwise at 0^{0}C under nitrogen atmosphere. After 3h, the reaction was quenched with a saturated solution of NH_4Cl (25 mL). The phases were separated and the aqueous phase was extracted with Et_2O (3 x 30 mL). The combined organic phases were dried over $MgSO_4$ and concentrated in vacuo to give the alcohol **6a** (300mg, 90%) as a yellow oil.

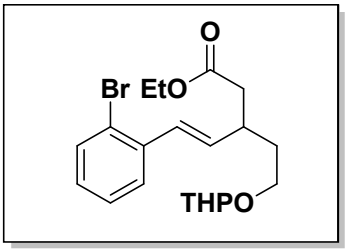
^{1}H NMR (300 MHz, $CDCl_3$): δ 7.80-7.76 (m, 1H), 7.57-7.52 (m, 1H), 7.37-7.32 (m, 1H), 7.20-7.14 (m, 1H), 5.78-5.75 (m, 1H), 4.76-4.73 (m, 1H), 4.67-4.62 (m, 1H), 3.89-3.80 (m, 2H), 3.61-3.45 (m, 2H), 2.60-2.54 (m, 2H), 1.85-1.77 (m, 1H), 1.73-1.66 (m, 1H), 1.62-1.45 (m, 5H) ppm; ^{13}C NMR (75 MHz, $CDCl_3$):δ 139.2, 131.7, 128.6, 127.5, 126.7, 121.7, 98.0, 83.4, 80.0, 65.3, 63.6, 61.9, 30.6, 25.6, 20.7, 19.6 ppm; MS (ESI): *m/z* ($[M+H]^+$): 339.

Procedure for the synthesis of (*E*)-1-(2-bromophenyl)-5-(tetrahydro-2*H*-pyran-2-yloxy)pent-2-en-1-ol (6b):



To a suspension of $LiAlH_4$ (119mg, 3.7mmol) in dry THF (15 mL) was added a solution of compound **6a** (1.2g, 3.7 mmol) in 10 mL THF slowly at 0 °C under nitrogen atmosphere. The mixture was stirred 2h at room temperature and then a sat. solution of Na_2SO_4 was added slowly to quench the excess $LiAlH_4$ at 0 °C. The resulting mixture was stirred for another 30 min and then filtered. The solid was washed with hot EtOAc (3x10mL). The combined EtOAc solutions were dried over $MgSO_4$ and concentrated in vacuo. The residue was purified by silica gel chromatography to give the compound **6b** (1.1g, 90%) as a colorless liquid.
^{1}H NMR (500 MHz, $CDCl_3$): δ 7.57-7.50 (m, 2H), 7.39-7.31 (m, 1H), 7.18-7.11 (m, 1H), 5.88-5.81 (m, 1H), 5.74-5.68 (m, 1H), 5.56 (d, *J* = 6.1 Hz, 1H), 4.59-4.55 (m, 1H), 3.85-3.73 (m, 2H), 3.50-3.41 (m, 2H), 2.36 (q, *J* = 6.7 Hz , 1H), 1.84-1.74 (m, 1H), 1.71-1.64 (m, 1H), 1.60-1.46 (m, 5H) ppm; ^{13}C NMR (75 MHz, $CDCl_3$):δ 131.8, 131.6, 128.4, 128.1, 127.1, 127.0, 98.3, 73.1, 66.5, 62.2, 33.1, 31.0, 25.9, 20.0 ppm; MS (ESI): *m/z* ($[M+H]^+$): 341.

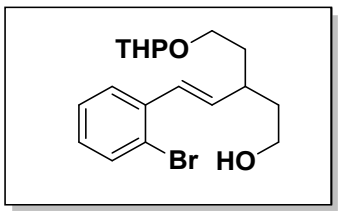
Procedure for the synthesis of (*E*)-ethyl 5-(2-bromophenyl)-3-((tetrahydro-2*H*-pyran-2-yloxy)methyl)pent-4-enoate (6c):



A mixture of (*E*)-1-(2-bromophenyl)-5-(tetrahydro-2*H*-pyran-2-yloxy)pent-2-en-1-ol (**6b**) (2.3 g, 7 mmol) and triethylorthoacetate (4.2 g, 35 mmol) in xylene (4 mL) was stirred at room temperature under nitrogen atmosphere. A catalytic amount of propionic acid was added to the above mixture and then heated at 140 ^{0}C for 24 h. The solvent was removed in vacuo and the crude product was purified by column chromatography on silica gel eluting with hexane to afford the ester **6c** (2.4g, 88%) as a liquid.

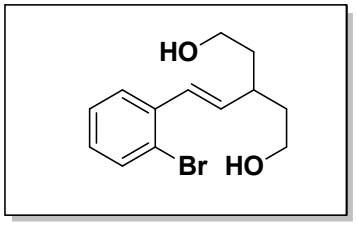
^{1}H NMR (300 MHz, $CDCl_3$): δ 7.55-7.44 (m, 2H), 7.36-7.20 (m, 2H), 7.10-7.03 (m, 1H), 6.76 (dd, *J* = 6.0, *J* = 15.6 Hz, 1H), 6.07-5.94 (m, 1H), 4.61-4.54 (m, 1H), 4.13 (q, *J* = 7.1 Hz, 1H), 3.91-3.74 (m, 2H), 3.53-3.37 (m, 3H), 3.01-2.89 (m, 1H), 2.58-2.34 (m, 3H), 1.91-1.45 (10H), 1.24 (q, *J* = 7.1 Hz, 3H) ppm; ^{13}C NMR (125 MHz, $CDCl_3$):δ 171.2, 136.6, 134.7, 132.2, 129.2, 129.0, 128.0, 126.8, 126.5, 122.8, 98.9, 98.3, 65.2, 62.3, 62.1, 60.4, 40.5, 37.4, 34.7, 30.8, 25.7, 19.9, 19.7, 14.6 ppm; MS (ESI): *m/z* ([M+H]$^+$): 397.

Procedure for the synthesis of (*E*)-5-(2-bromophenyl)-3-(2-(tetrahydro-2*H*-pyran-2-yloxy)ethyl)pent-4-en-1-ol (6d):



To a suspension of $LiAlH_4$ (119mg, 3.7mmol) in dryTHF (15mL) was added a solution of **6c** (1.5g, 3.7mmol) in 10 mL THF slowly at 0 °C under nitrogen atmosphere. The mixture was stirred 2 h at room temperature. Up on completion, a sat. solution of Na_2SO_4 was added slowly to quench the excess $LiAlH_4$ at 0 °C. The mixture was stirred for another 30 min and then filtered. The solid was washed with hot EtOAc (3x10 mL). The combined EtOAc solutions were dried over $MgSO_4$ and the solvent was removed under vacuum. The resulting residue was purified by silica gel chromatography to give the compound **6d** (1.2g, 90%) as a colorless liquid.

Procedure for the synthesis of (*E*)-3-(2-bromostyryl)pentane-1,5-diol (6):



To a solution of **6d** (360mg, 1mmol) in MeOH (3 mL) at 0 ^{0}C was added a catalytic amount *p*-TSA. The mixture was stirred 1 h at room temperature. After completion, the reaction was quenched with solid $NaHCO_3$ and then stirred it for 30 min. The solvent was removed under vacuum and the resulting crude compound was diluted with 10 mL CH_2Cl_2, further washed water, and concentrated to give the residue which was purified by silica gel chromatography to give the compound **6** (228mg, 80%) as a liquid.

References:

1. S. S. Khokhar; T. Wirth, *Eur. J. Org. Chem.* **2004,** 4567-4581.

2. X-ray Crystallography:

X-ray data for the compounds were collected at room temperature using a Bruker Smart Apex CCD diffractometer with graphite monochromated MoKα radiation (λ=0.71073Å) with ω-scan method[1]. Preliminary lattice parameters and orientation matrices were obtained from four sets of frames. Unit cell dimensions were determined using 9815 reflections for **2a** and 6522 reflections for **7a**.

Integration and scaling of intensity data were accomplished using SAINT program.[1] The structure was solved by direct methods using SHELXS97[2] and refinement was carried out by full-matrix least-squares technique using SHELXL97[2]. Anisotropic displacement parameters were included for all non-hydrogen atoms. All H atoms were positioned geometrically and treated as riding on their parent C atoms, with C-H distances of 0.93--0.96 Å, and with $U_{iso}(H) = 1.2U_{eq}$ (C). The absolute configuration of the procured material was known in advance and was confirmed by unambiguous refinement of the absolute structure parameter[3] for **7a**.

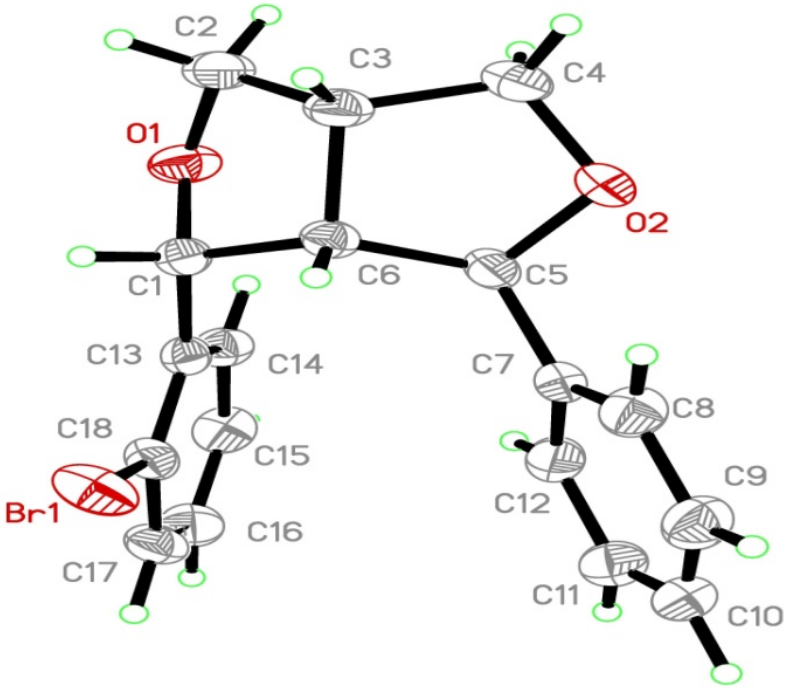


Figure 1. ORTEP diagram of **2a**

Crystal data for **2a**: $C_{18}H_{17}BrO_2$, M = 345.23, colorless block, 0.16 × 0.13 × 0.06 mm^3, orthorhombic, space group *Pbca* (No. 61), a = 9.6034(5), b = 15.7805(8), c = 19.7861(10) Å, V = 2998.5(3) Å^3, Z = 8, D_c = 1.529 g/cm^3, F_{000} = 1408, CCD Area Detector, MoKα radiation, λ = 0.71073 Å, T = 294(2)K, $2\theta_{max}$ = 50.0º, 26760 reflections collected, 2643

unique (R_{int} = 0.0397). Final *GooF* = 1.082, *R1* = 0.0314, *wR2* = 0.0824, *R* indices based on 2176 reflections with I>2σ(I) (refinement on F^2), 190 parameters, 0 restraints, μ = 2.743 mm^{-1}. CCDC 943366 contains supplementary Crystallographic data for the structure.

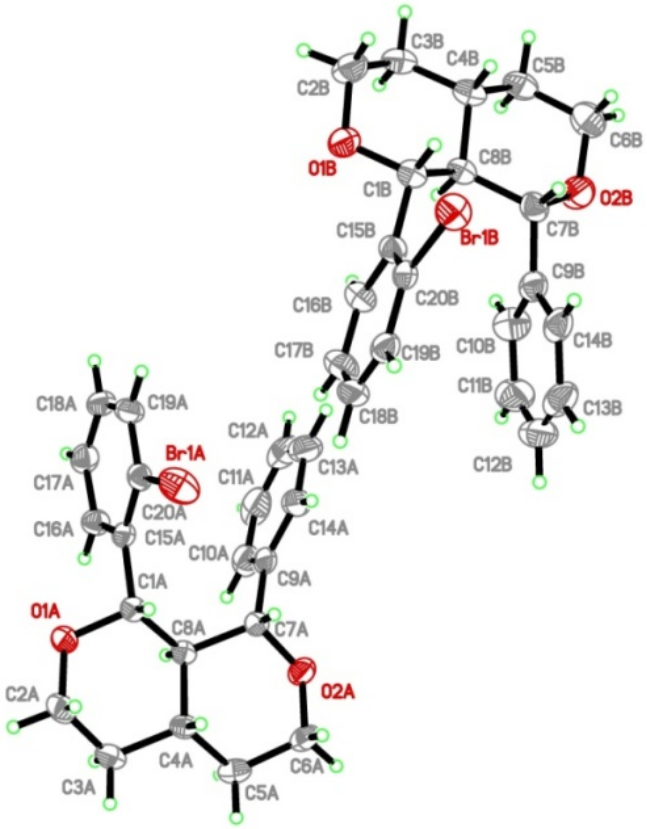


Figure 2. ORTEP diagram of **7a**

Crystal data for **7a**: $C_{20}H_{21}BrO_2$, *M* = 373.28, colorless needle, 0.18 × 0.11 × 0.09 mm^3, orthorhombic, space group $P2_12_12_1$ (No. 19), *a* = 11.2110(12), *b* = 16.7616(18), *c* = 18.3398(19) Å, *V* = 3446.3(6) $Å^3$, *Z* = 8, D_c = 1.439 g/cm^3, F_{000} = 1536, CCD Area Detector, MoKα radiation, λ = 0.71073 Å, *T* = 294(2)K, $2\theta_{max}$ = 50.0º, 31390 reflections collected, 6049 unique (R_{int} = 0.0507). Final *GooF* = 0.974, *R1* = 0.0344, *wR2* = 0.0881, *R* indices based on 5025 reflections with I>2σ(I) (refinement on F^2), 415 parameters, 0 restraints, μ = 2.393 mm^{-1}. Absolute structure parameter = 0.080(8). CCDC 943367 contains supplementary Crystallographic data for the structure. These data can be obtained free of charge at www.ccdc.cam.ac.uk/conts/retrieving.html [or from the Cambridge Crystallographic Data Centre (CCDC), 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44(0) 1223 336 033; email: deposit@ccdc.cam.ac.uk].

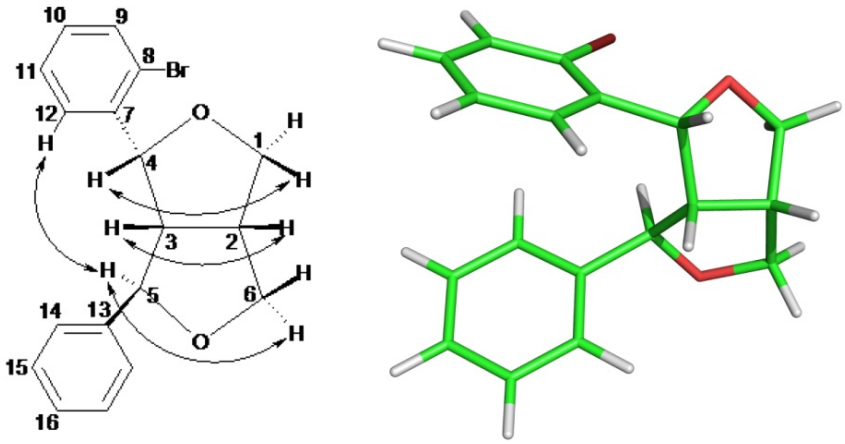
1. Bruker (2001). SAINT (Version 6.28a) & SMART (Version 5.625). Bruker AXS Inc., Madison, Wisconsin, USA.
2. Sheldrick GM. (2008**)** Acta Crystallogr A64: 112-122.
3. Flack, K. & Bernardinelli, G. (2000). J. Appl. Cryst. 33, 1143--1148.

Figure Caption

Fig.**1**. A view of **2a**, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 30% probability level and H atoms are represented by circles of arbitrary radii. The compound is crystallized in orthorhombic space group Pbca with one molecule in the asymmetric unit

Fig.**2**. A view of **7a**, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 30% probability level and H atoms are represented by circles of arbitrary radii. The compound is crystallized in orthorhombic space group $P2_12_12_1$ with two molecules in the asymmetric unit

3. 2D NMR experiments and NOE studies :

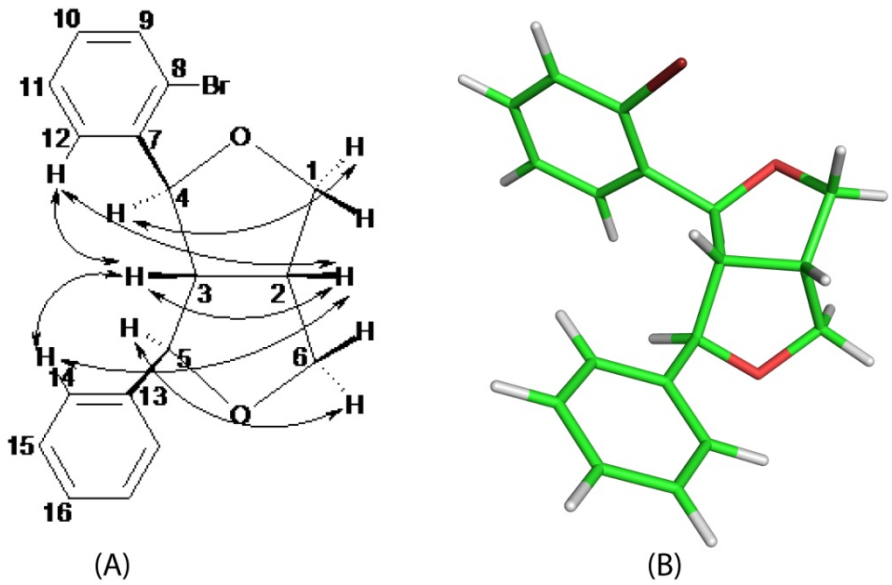


(A) (B)

(1*R*,3a*S*,6*R*,6a*R*)-1-(2-Bromophenyl)-6-phenylhexahydrofuro[3,4-c]furan (2a):

^{1}H NMR (500 MHz): 7.74 (dd, *J* = 1.9, 7.8 Hz, 1H, H8), 7.39 (dt, *J* = 1.2, 7.6 Hz, 1H, H9), 7.34 (dd, *J* = 1.2, 8.0 Hz, 1H,H11), 7.15 (dt, *J* = 1.8, 7.7 Hz, 1H, H10), 7.11 (m, 3H, Aro, H15, H15', H16), 6.68 (dd, *J* = 1.4, 7.8 Hz, 2H, H14, H14'), 5.02 (d, *J* = 6.0 Hz, 1H, H4), 4.38 (dd, *J* = 8.2, 8.9 Hz, 1H, H6), 4.18 (d, *J* = 7.0 Hz, 1H, H5), 4.06 (dd, *J* = 1.5, 9.2 Hz, 1H, H1'), 3.87 (dd, *J* = 6.4, 9.2 Hz, 1H, H1), 3.62 (dd, *J* = 7.0, 8.9 Hz, 1H, H6'), 3.44 (ddd, *J* = 6.0, 7.0, 9.4 Hz, 1H, H3), 3.25 (m, 1H, H2).

^{13}C NMR (125MHz): 140.7(C13), 137.7 (C7), 132.2(C11), 128.8(C10), 128.3 (C8), 128(C15, C15'), 127.3(C16), 127.1(C9), 126.3 (C14, C14'), 122.0 (C12), 82.6 (C5), 81.7 (C4), 74.2 (C6), 71.4 (C1), 55.0(C3), 46.8 (C2).

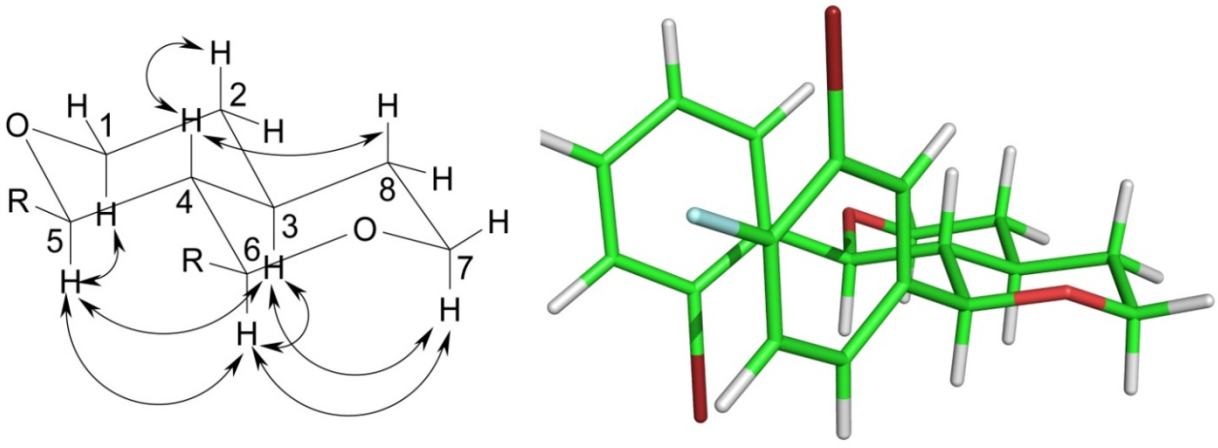


(A) (B)

(1*S*,3a*S*,6*R*,6a*R*)-1-(2-Bromophenyl)-6-phenylhexahydrofuro[3,4-c]furan (3a):

^{1}H NMR (500 MHz): 7.50 (dd, *J* = 1.3, 8.0 Hz, 1H, H9), 7.44 (dd, *J* = 1.8, 7.8 Hz, 1H, H12), 7.33 (m, 1H, H11), 7.33 (m, 2H, H15, H15'), 7.30 (dd, *J* = 1.6, 8.0 Hz, 2H, H14, H14'), 7.24 (m, 1H, H16), 7.11 (dt, *J* = 1.7, 7.7 Hz, 1H, H10), 5.31 (d, *J* = 4.3 Hz, 1H, H4), 5.23 (d, *J* = 4.6 Hz, 1H, H5), 4.35 (dd, *J* = 7.5, 8.9 Hz, 1H, H6), 4.19 (dd, *J* = 6.9, 9.0 Hz, 1H, H1), 3.90 (dd, *J* = 5.6, 8.9 Hz, 1H, H6'), 3.85 (dd, *J* = 4.1, 9.0 Hz, 1H, H1'), 3.18 (m, 1H, H2), 3.01 (ddd, *J* = 4.3, 4.6, 9.0, 1H, H3).

^{13}C NMR (125 MHz): 141.3, 141.1, 133.0, 129.0, 128.4, 127.5, 127.3, 126.2, 122.0, 85.5, 84.5, 73.4, 72.1, 61.1, 46.4.



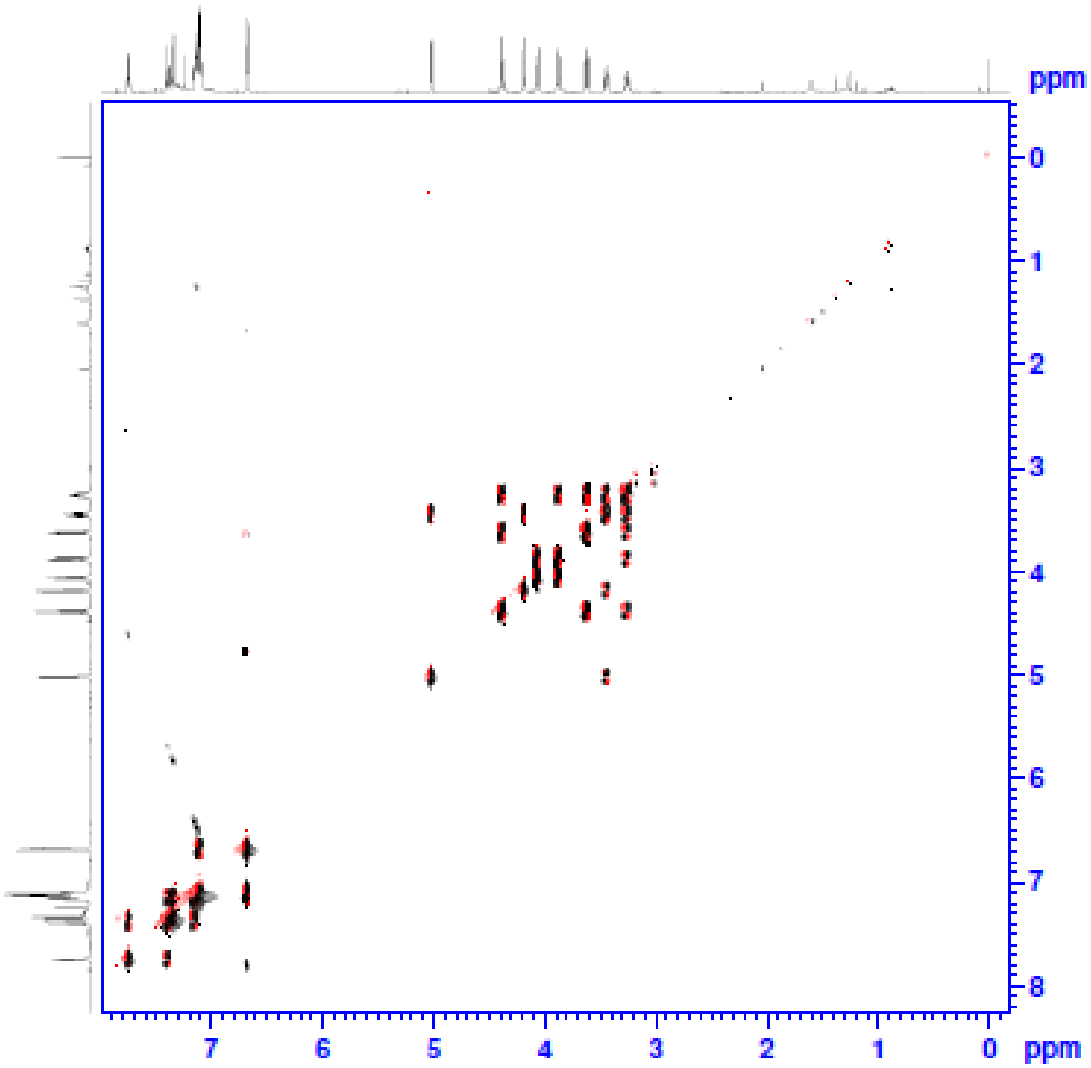
(A) (B)

(1*R*,4a*S*,8*S*,8a*R*)-1-(3-Bromo-4-fluorophenyl)-8-(2-bromophenyl)octahydropyrano[3,4-c]pyran (8d):

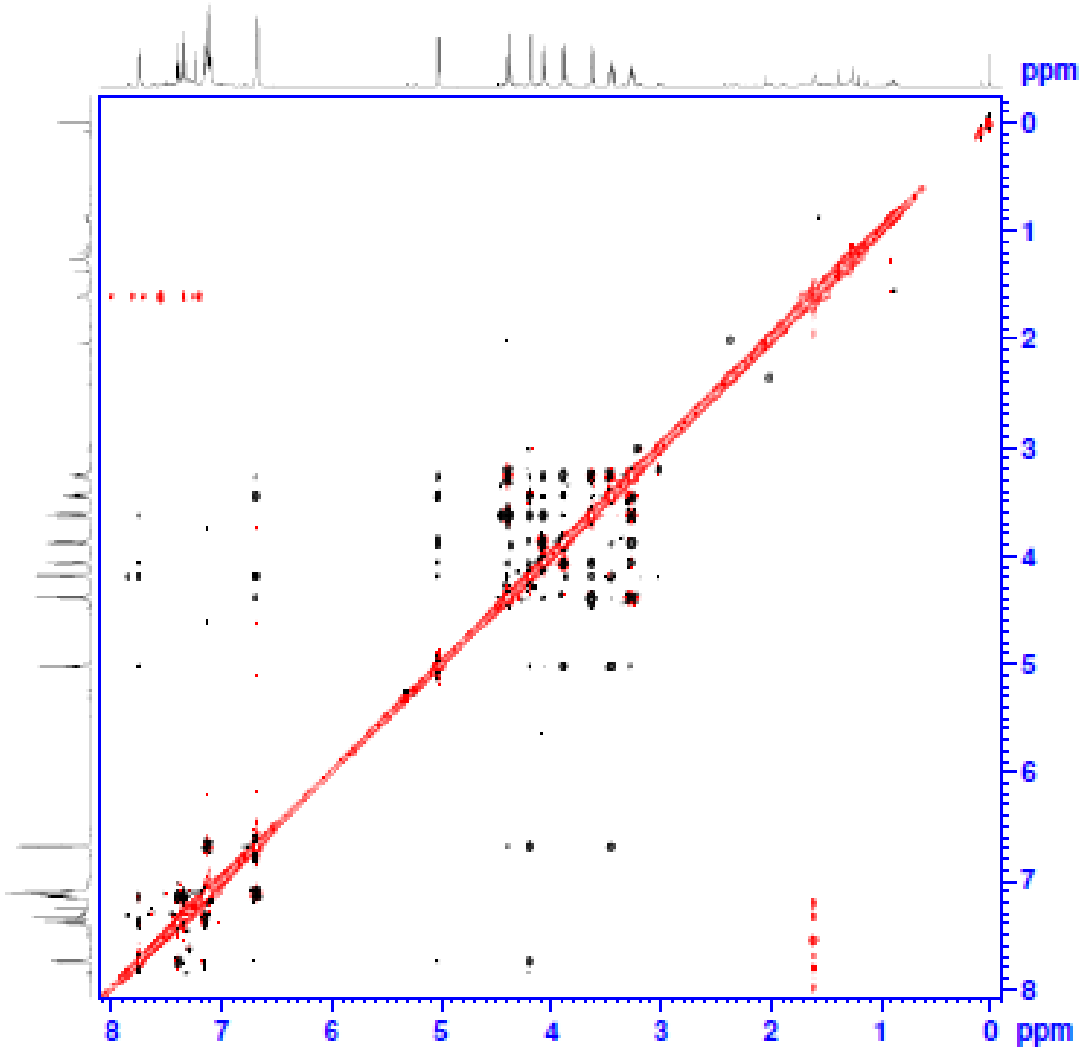
^{1}H NMR (500 MHz): 6.95-6.55 (m, 8H, Aromatic), 4.09 (m, 2H, H1', H7'), 4.02 (d, *J* = 10 Hz, 1H, H5), 4.00 (d, *J* = 10 Hz, 1H, H6), 3.69 (m, 2H, H1, H7), 2.08 (q, *J* = 10.0 Hz, 1H, H4), 1.86 (m, 1H, H3), 1.73-1.68 (m, 4H, H2, H2', H8, H8'), 1.68 (m, 2H, H).
^{13}C NMR (125 MHz): 158.7, 156.7, 139.9, 137.8, 133.3, 128.4, 128.4, 127.8, 127.7, 127.5, 115.4, 115.2, 108.1, 108.0, 82.7, 81.5, 68.5, 68.5, 51.0, 40.3, 33.1, 33.0.

The detailed structural studies were performed for compounds **2a**, **3a** and **8d** using 1D and 2D NMR experiments. The large coupling constant (JH2-H3=9.4 Hz for A and 9.0 for B) between H2-H3 protons and the presence of strong NOE cross peak between H2-H3 indicate that these protons are in cis orientation for both **2a** and **3a** compounds. Strong HMBC correlations between H12-C4, H4-C12, H14-C5, H5-C14 and strong NOE correlations between H12-H4, H14-H5 were used to assign the furan ring resonances unambiguously for compound **2a**. In the NOESY spectrum of compound **2a** the presence of NOE cross peak between H12-H5 and the coupling constant $^{3}J_{H3\text{-}H4}$= 6.0 Hz, $^{3}J_{H3\text{-}H5}$=7.0 Hz support the Bromo phenyl ring is down and the phenyl ring is upside the furan rings. Whereas for the compound **3a** the presence of NOE cross peaks between H2-H14, H3-H14, H2-H12 and the coupling constant $^{3}J_{H3\text{-}H4}$= 4.3 Hz, $^{3}J_{H3\text{-}H5}$=4.6 Hz support the both rings are upside the furan rings. For the compound **8d** the large coupling constants between H3-H4 ($^{3}J_{H3\text{-}H4}$=10.0 Hz), H4-H5 ($^{3}J_{H4\text{-}H5}$=10.0 Hz), H4-H6 ($^{3}J_{H4\text{-}H6}$=10.0 Hz) and the presence of strong NOE cross peaks between H3-H6, H3-H5 indicate H3-H4 are in trans orientation.

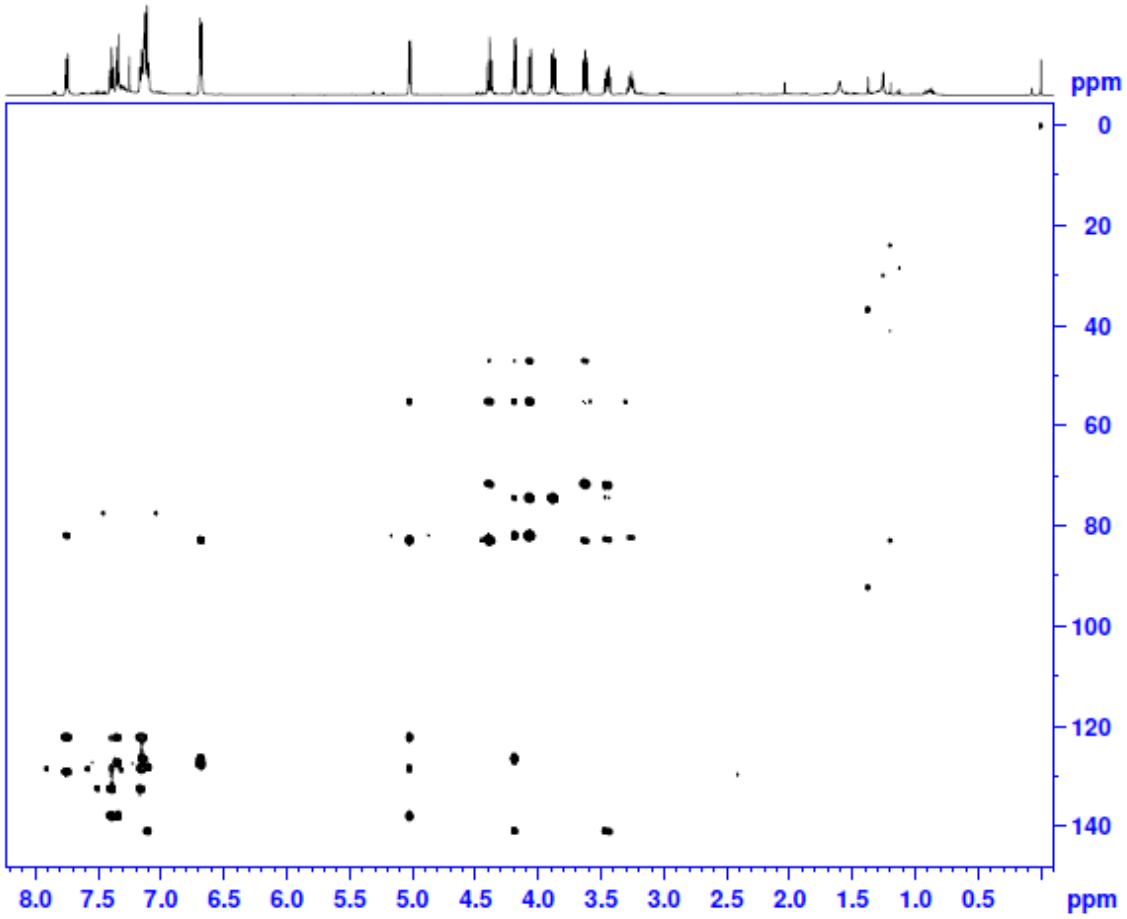
2D-DQFCOSY spectrum of (1*R*,3a*S*,6*R*,6a*R*)-1-(2-bromophenyl)-6-phenylhexahydrofuro[3,4-*c*]furan (2a):



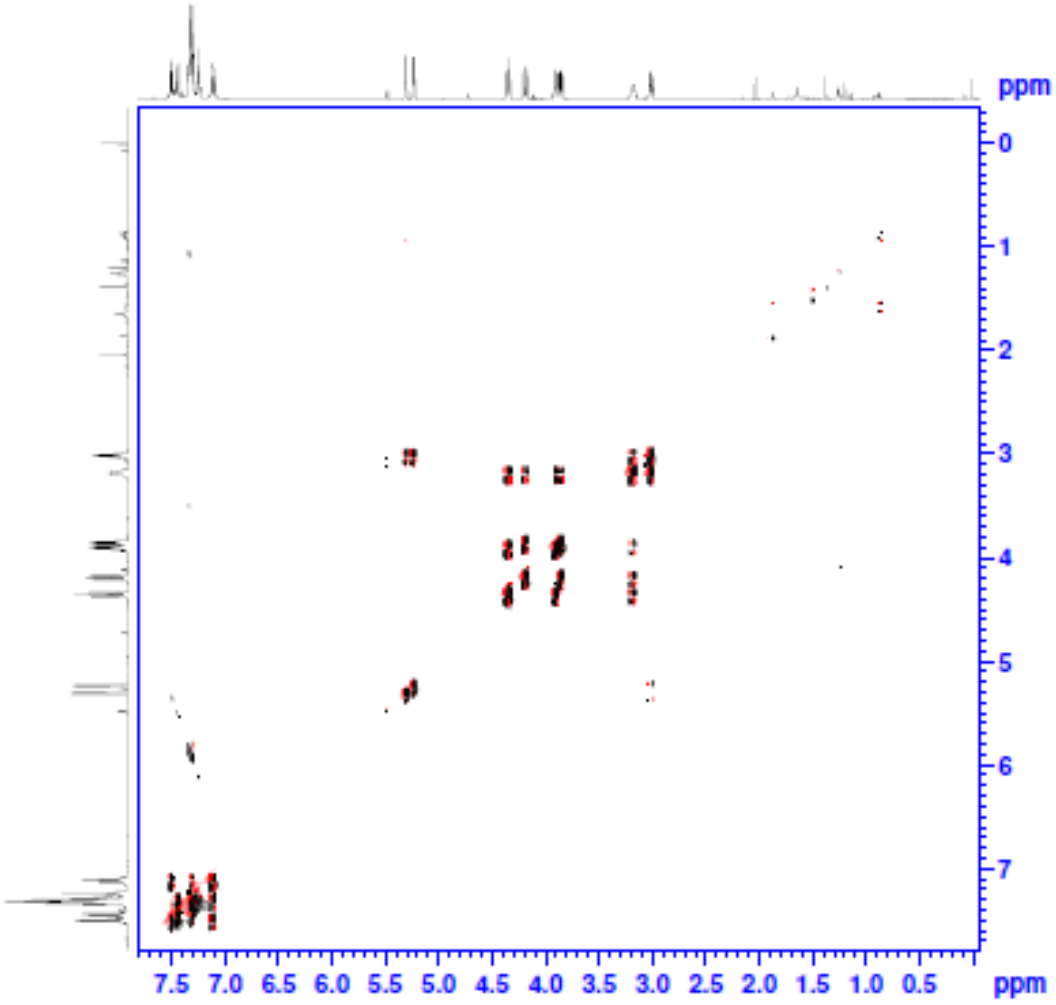
2D-NOESY spectrum of (1*R*,3a*S*,6*R*,6a*R*)-1-(2-bromophenyl)-6-phenylhexahydrofuro[3,4-*c*]furan (2a):



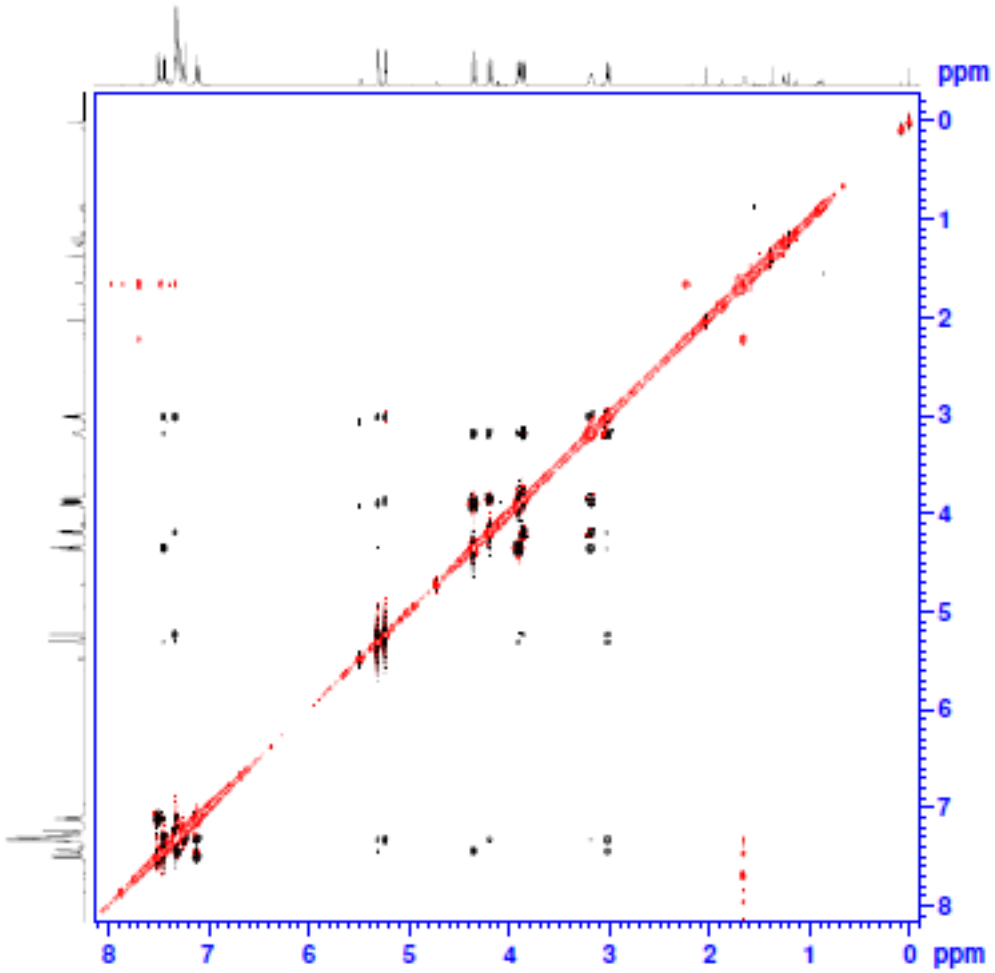
2D HMBC spectrum of (1*R*,3a*S*,6*R*,6a*R*)-1-(2-bromophenyl)-6-phenylhexahydrofuro[3,4-c]furan (2a):



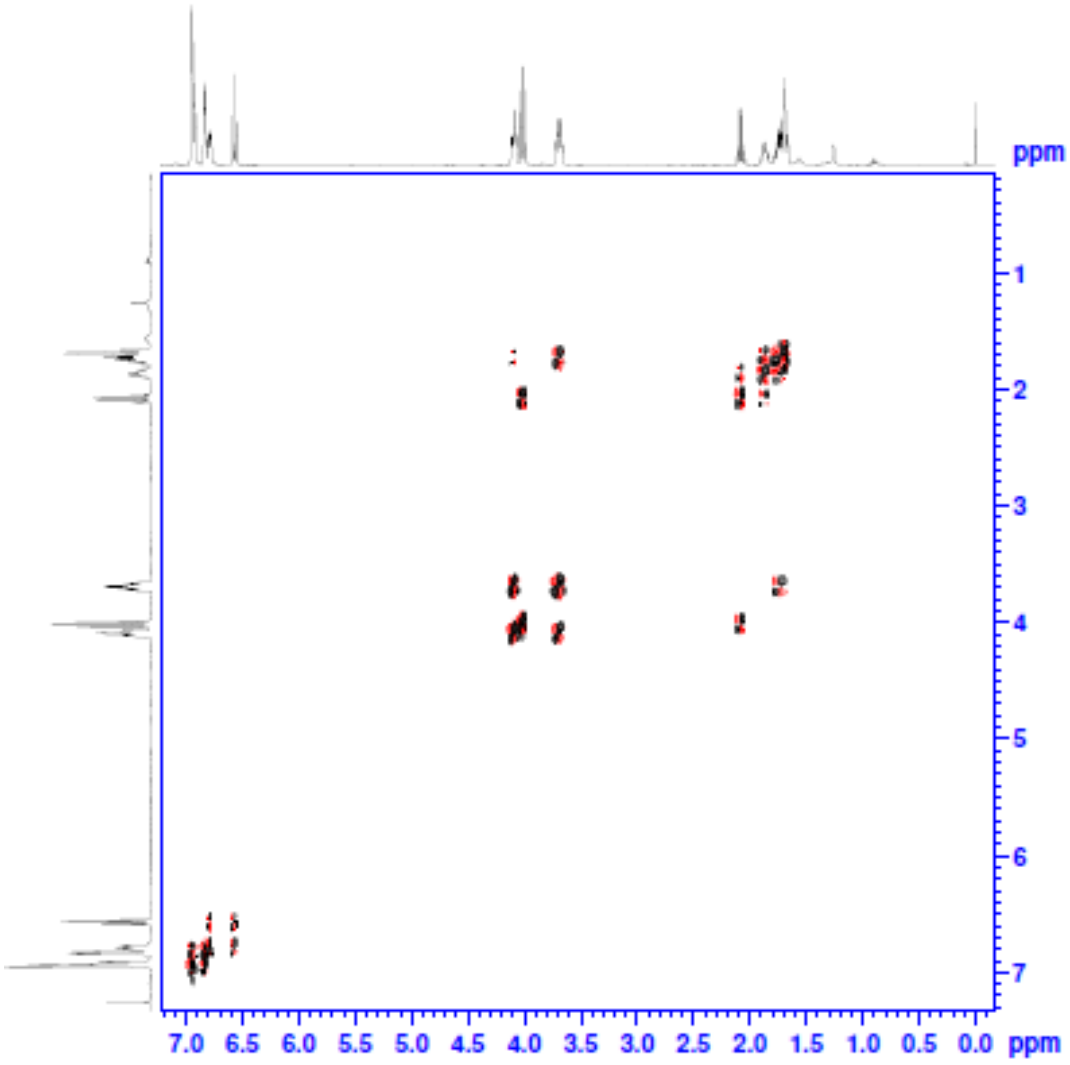
2D DQCOSY Spectrum of (1*S*,3a*S*,6*R*,6a*R*)-1-(2-bromophenyl)-6-phenylhexahydrofuro[3,4-*c*]furan (3a):



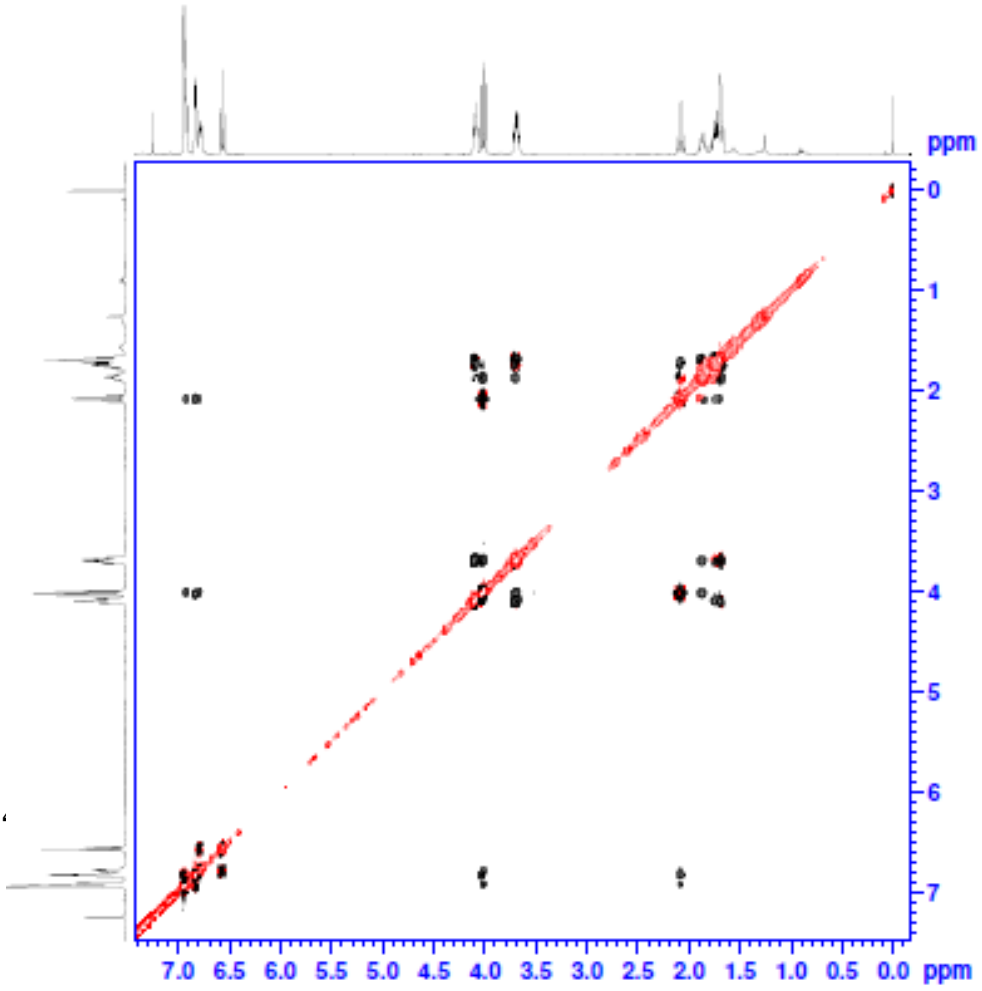
2D NOESY Spectrum of (1*S*,3a*S*,6*R*,6a*R*)-1-(2-bromophenyl)-6-phenylhexahydrofuro[3,4-*c*]furan (3a):



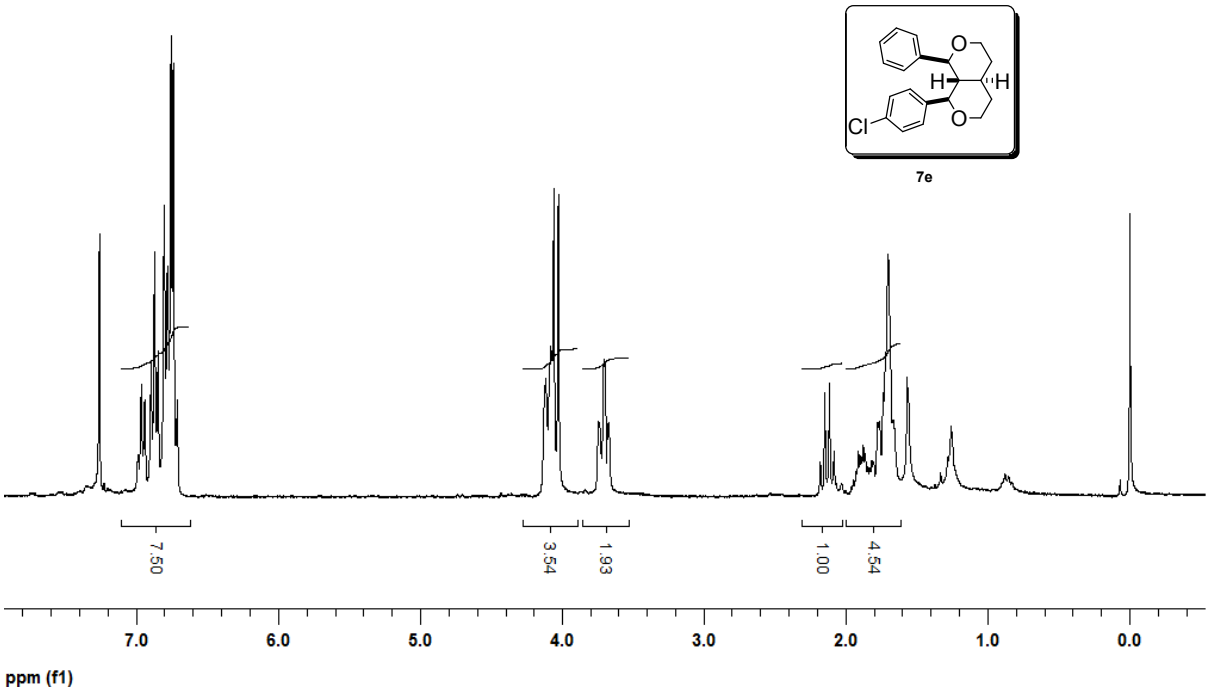
2D DQCOSY Spectrum of (1*R*,4a*S*,8*S*,8a*R*)-1-(3-bromo-4-fluorophenyl)-8-(2-bromophenyl)octahydropyrano[3,4-*c*]pyran (8d):



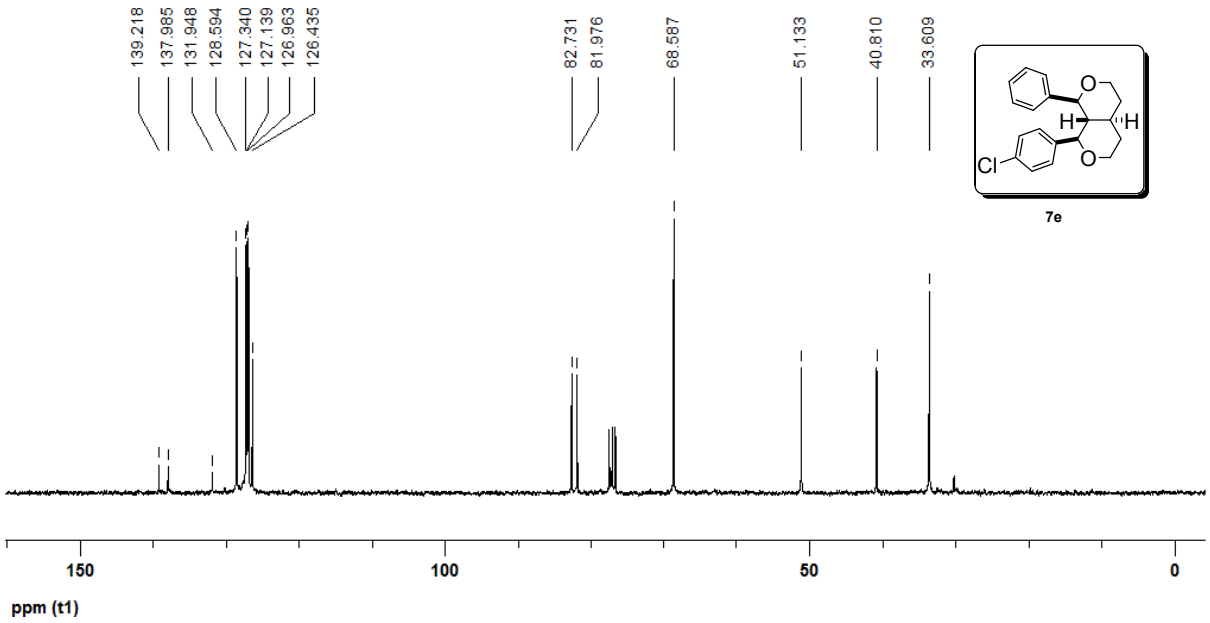
2D NOESY Spectrum of (1*R*,4a*S*,8*S*,8a*R*)-1-(3-bromo-4-fluorophenyl)-8-(2-bromophenyl)octahydropyrano[3,4-c]pyran (8d):



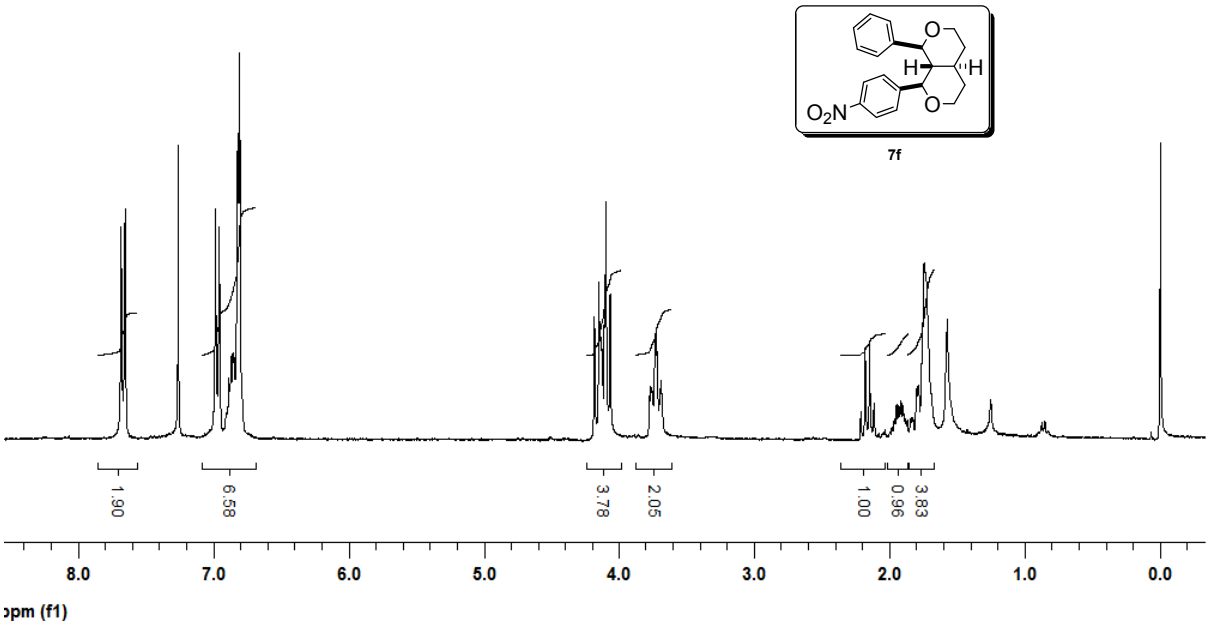
4. Copies of ^{1}H and ^{13}C NMR spectra:



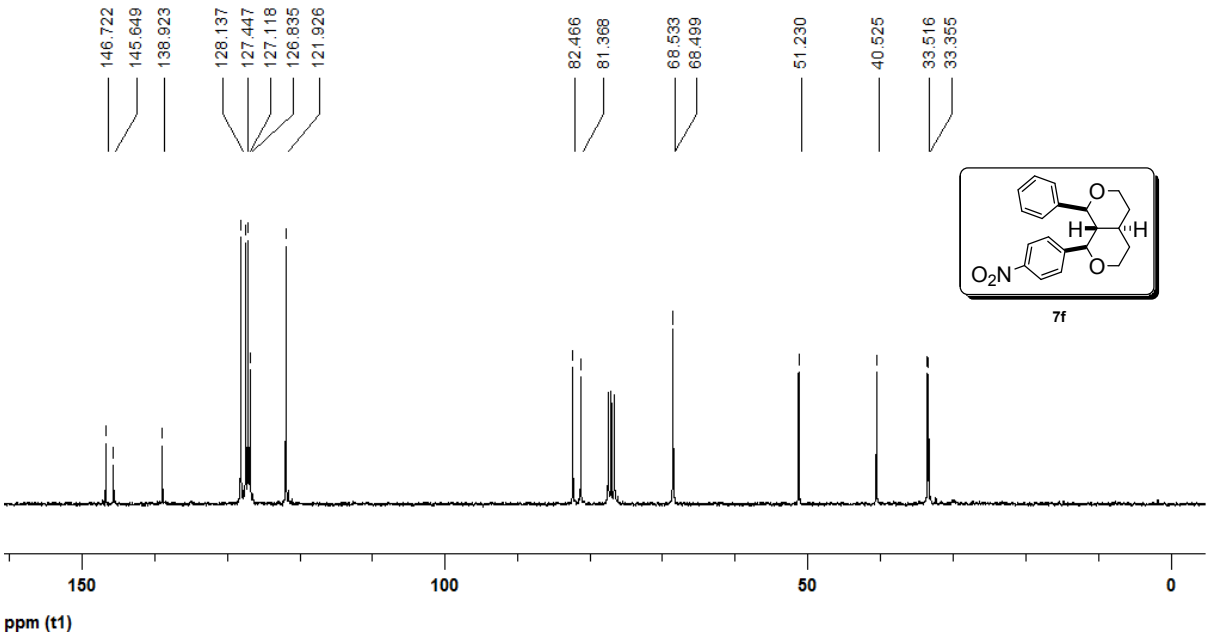
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 7e



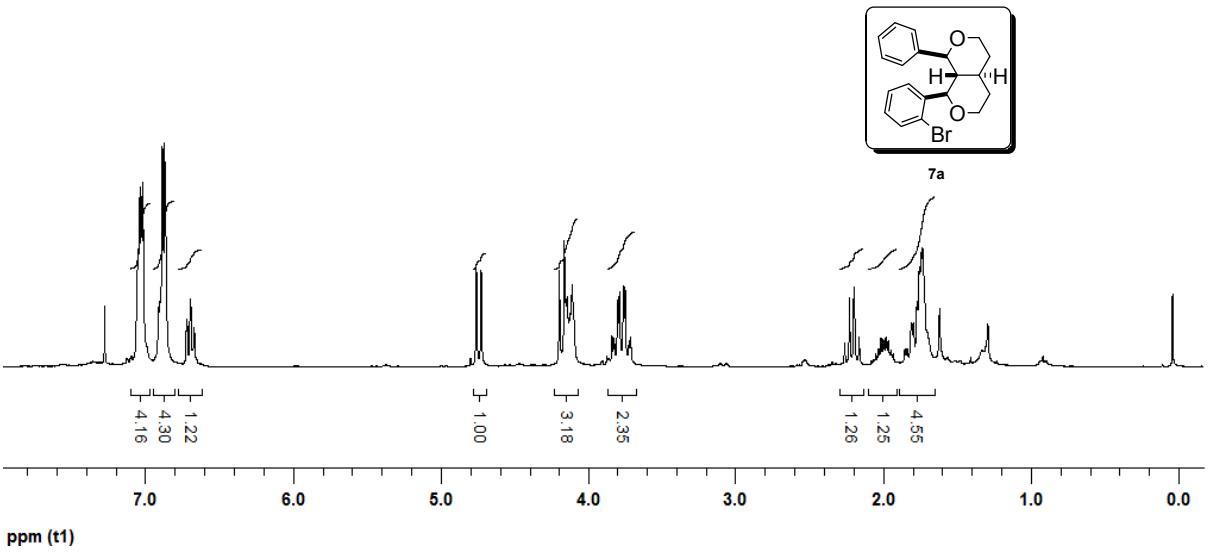
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 7e



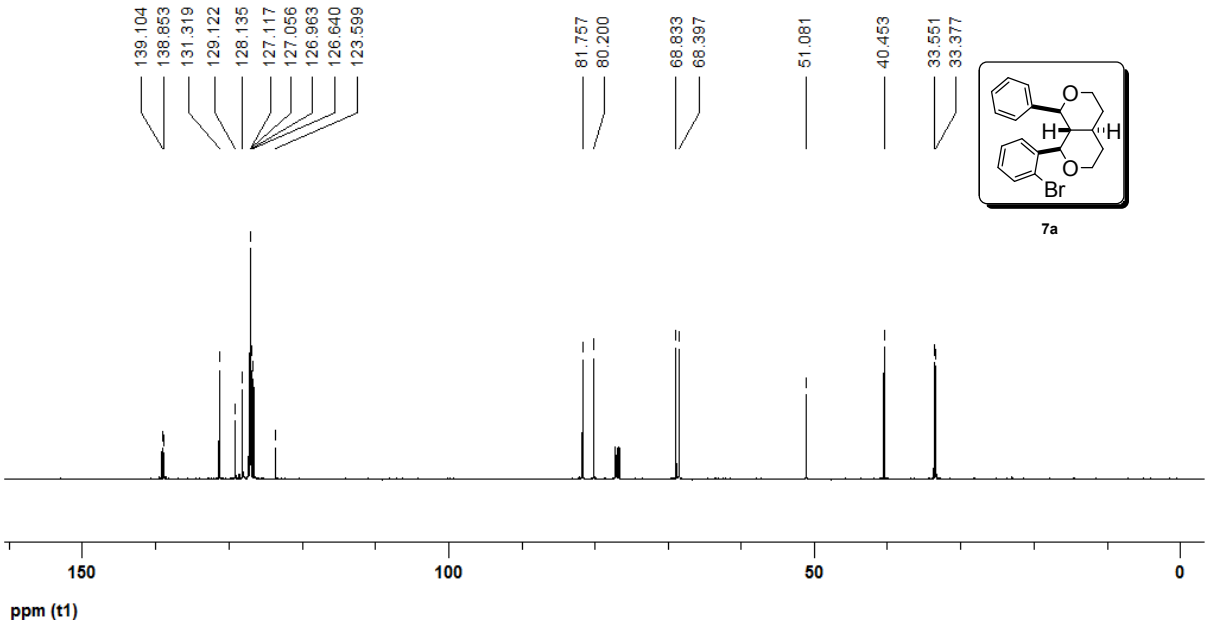
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 7f



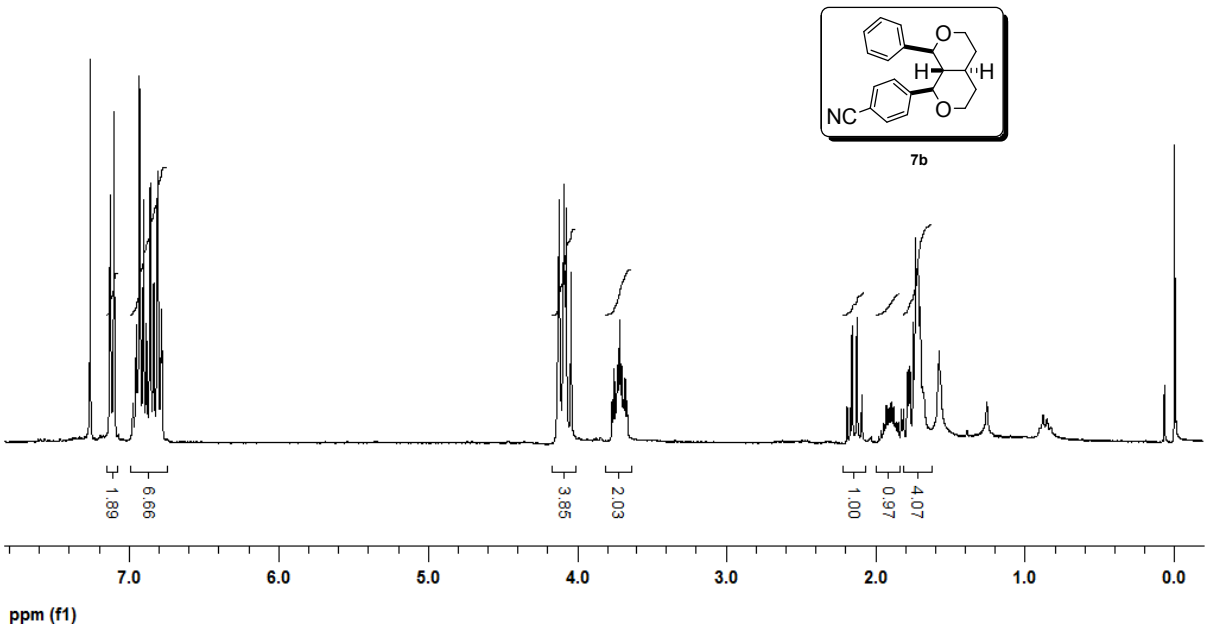
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 7f



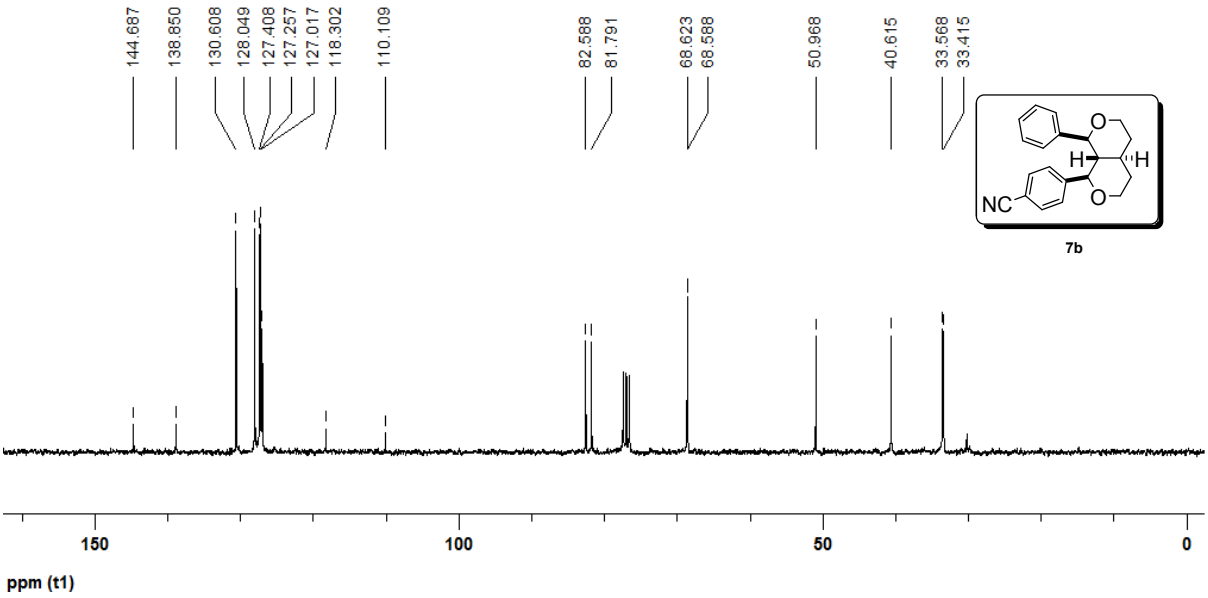
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 7a



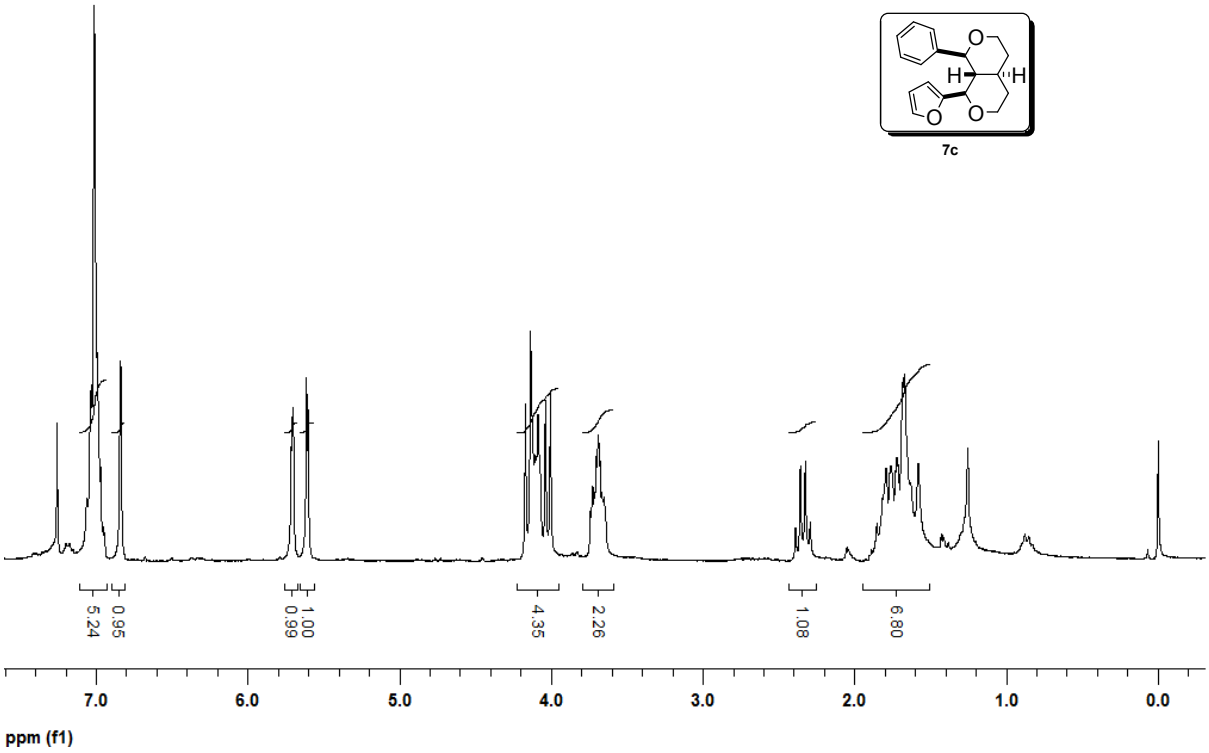
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 7a



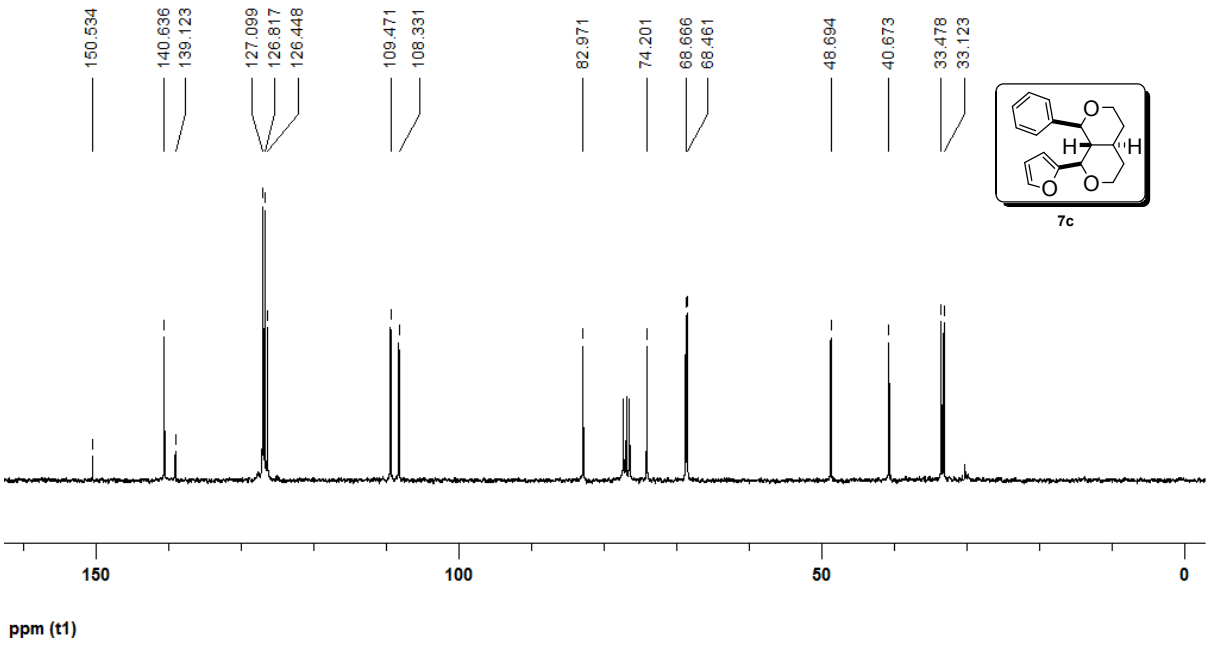
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 7b



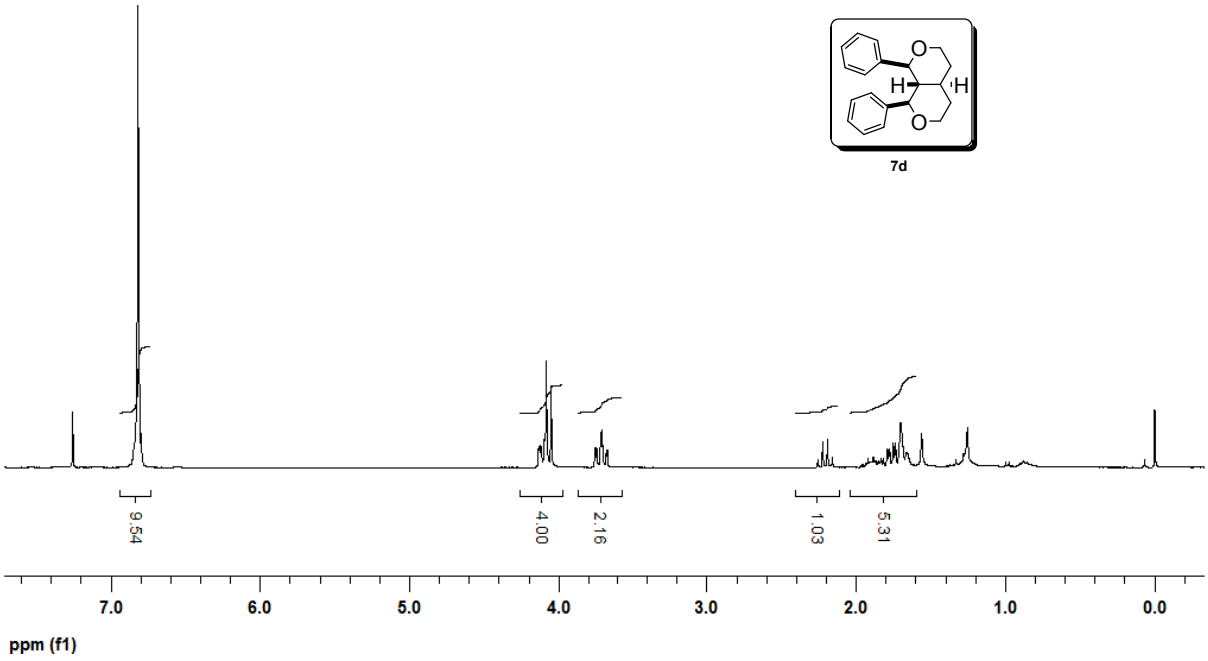
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 7b



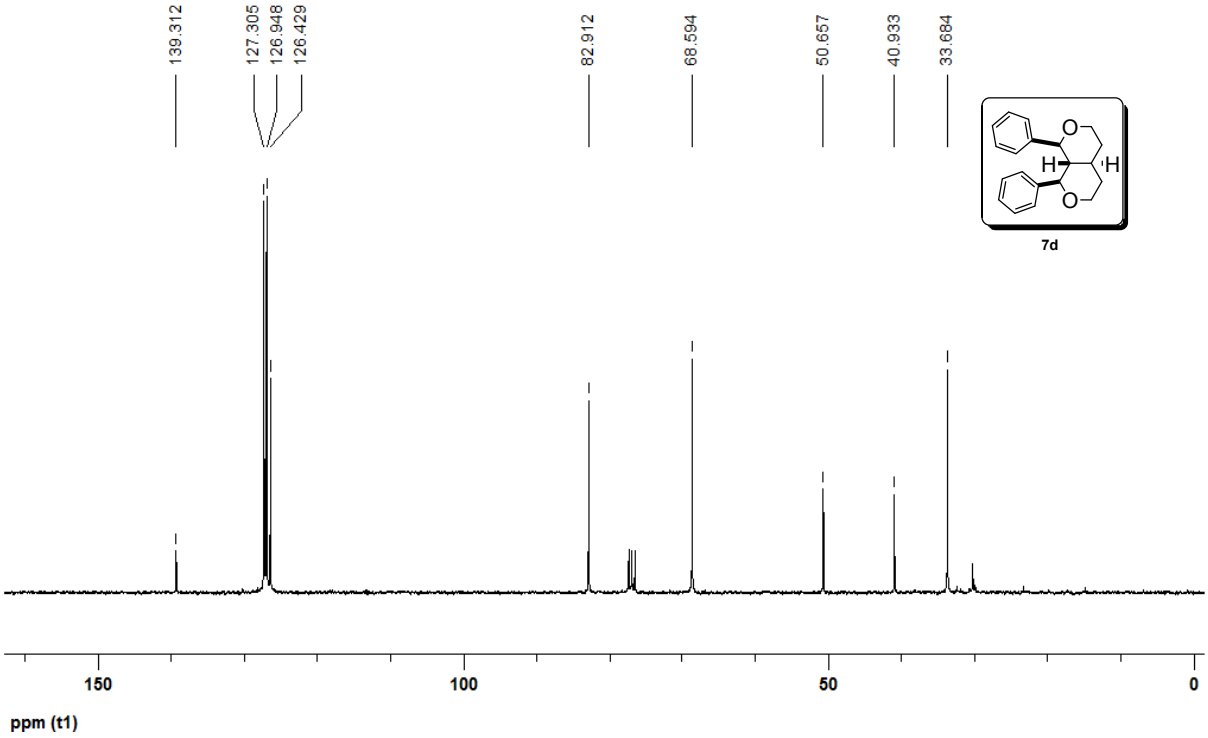
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 7c



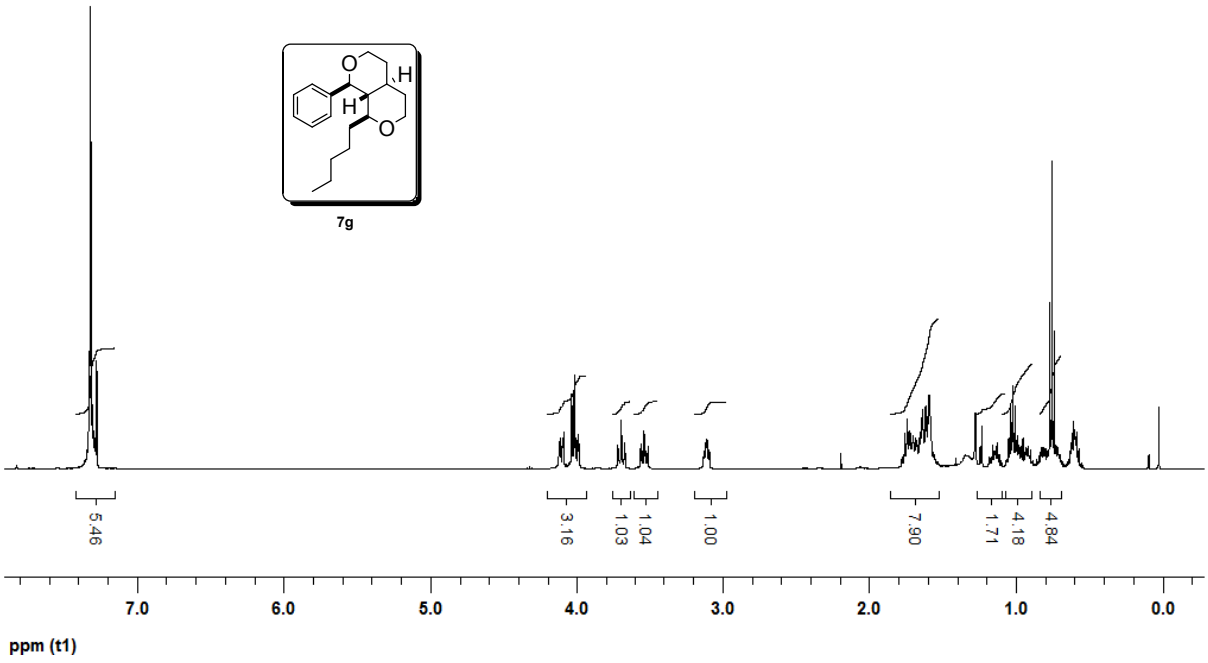
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 7c



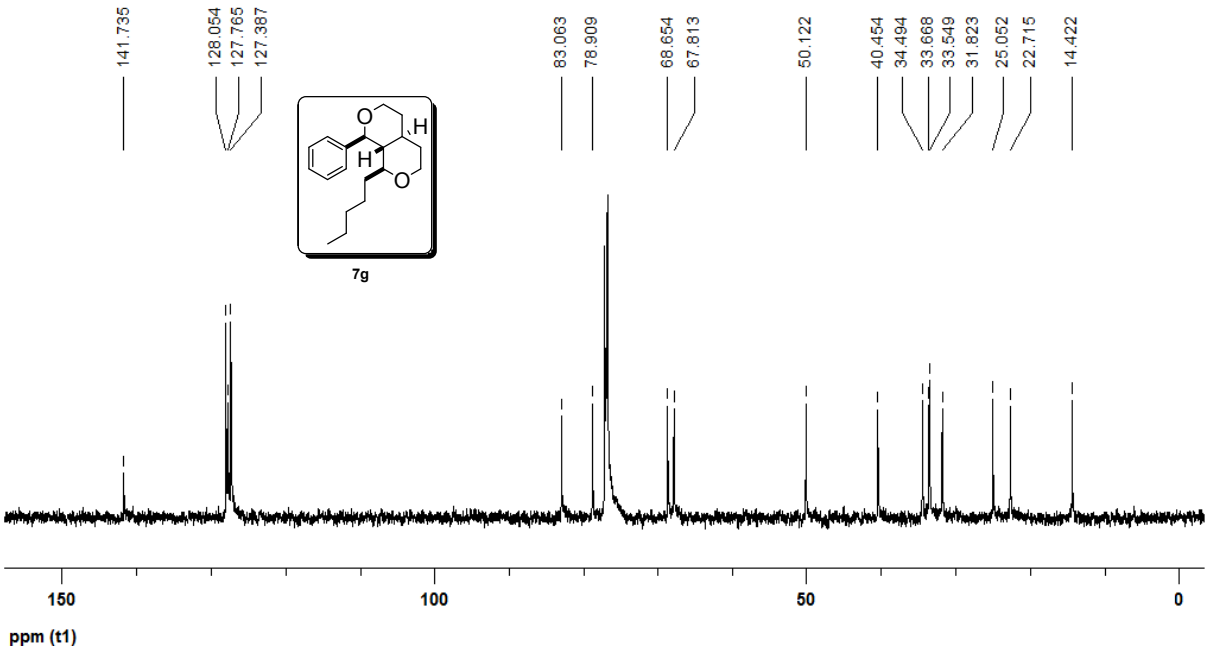
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 7d



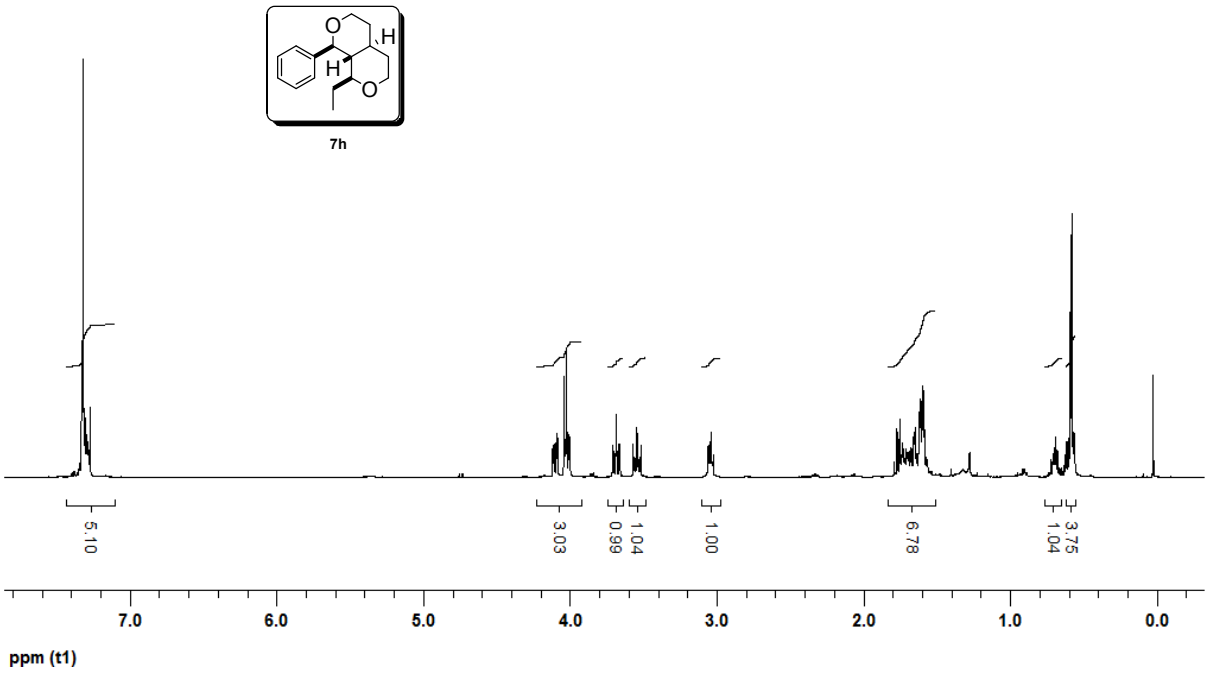
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 7d



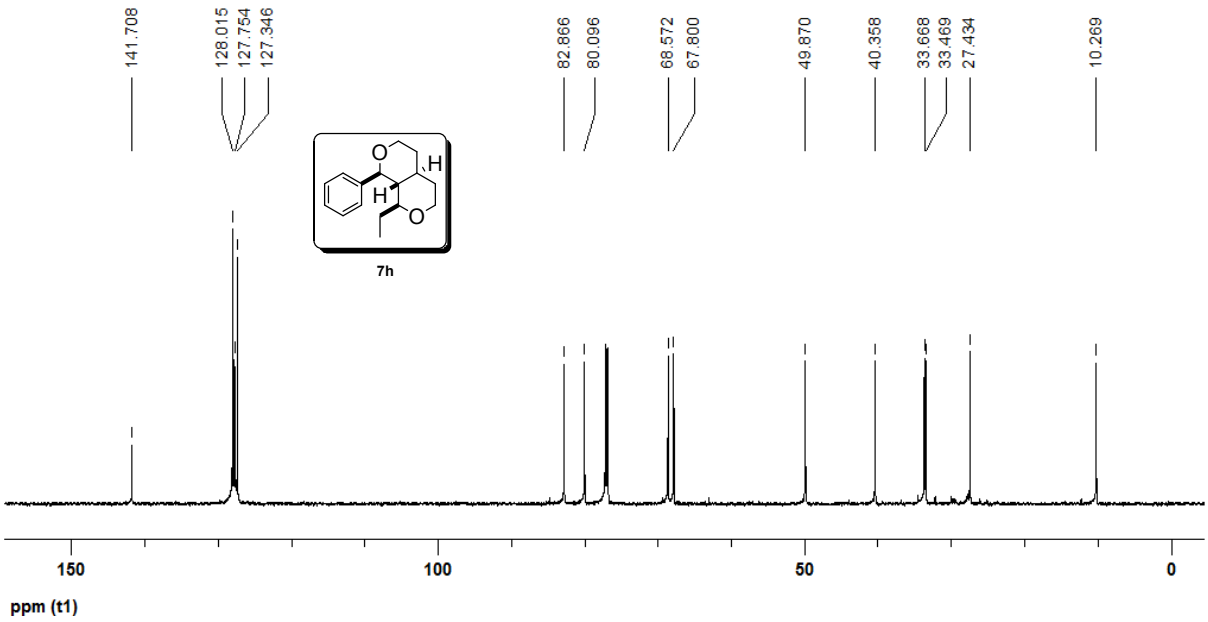
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 7g



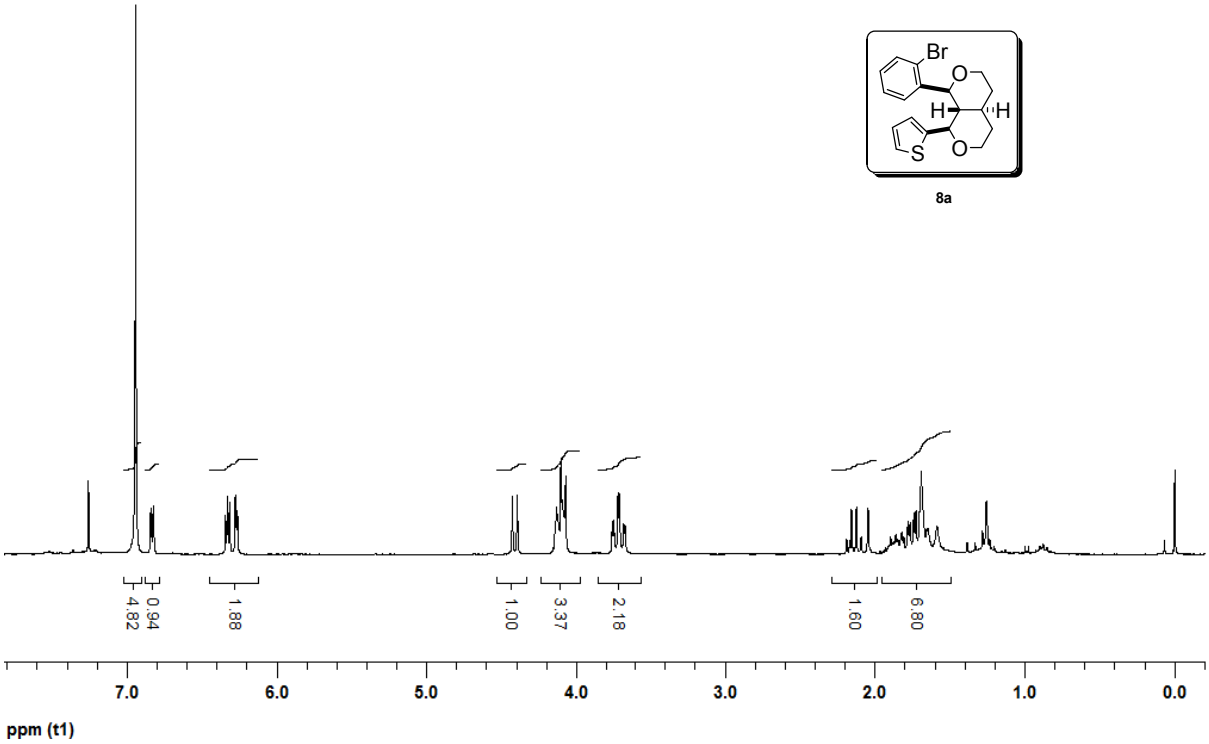
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 7g



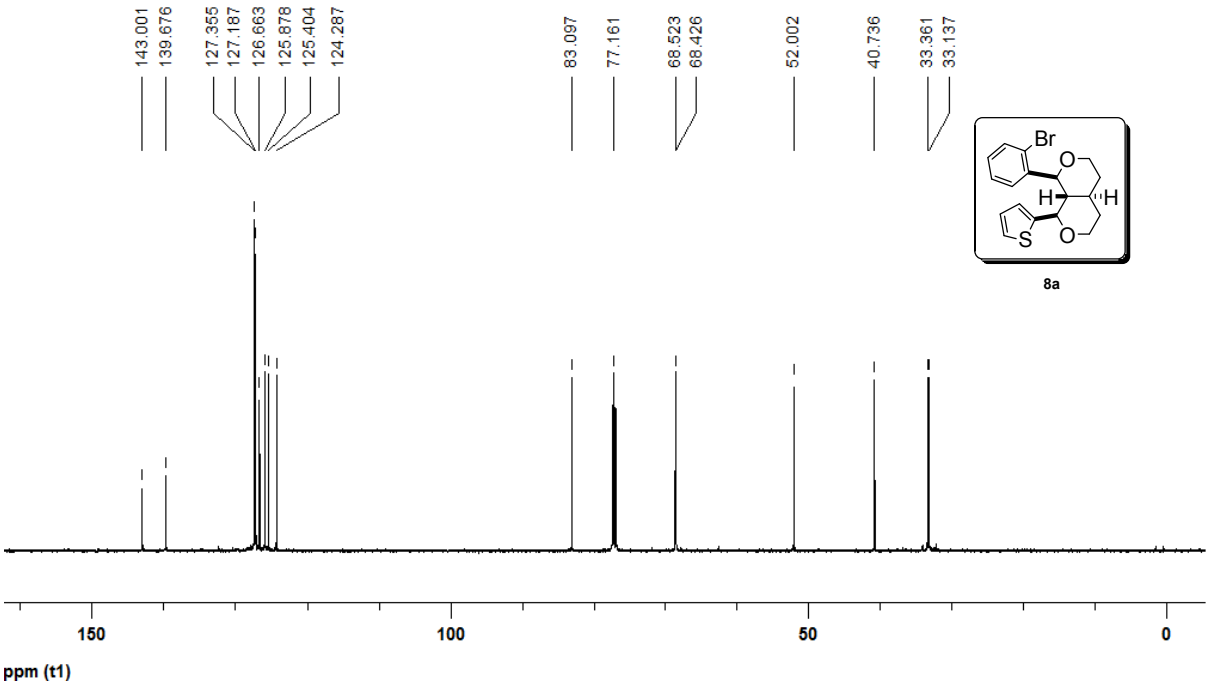
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 7h



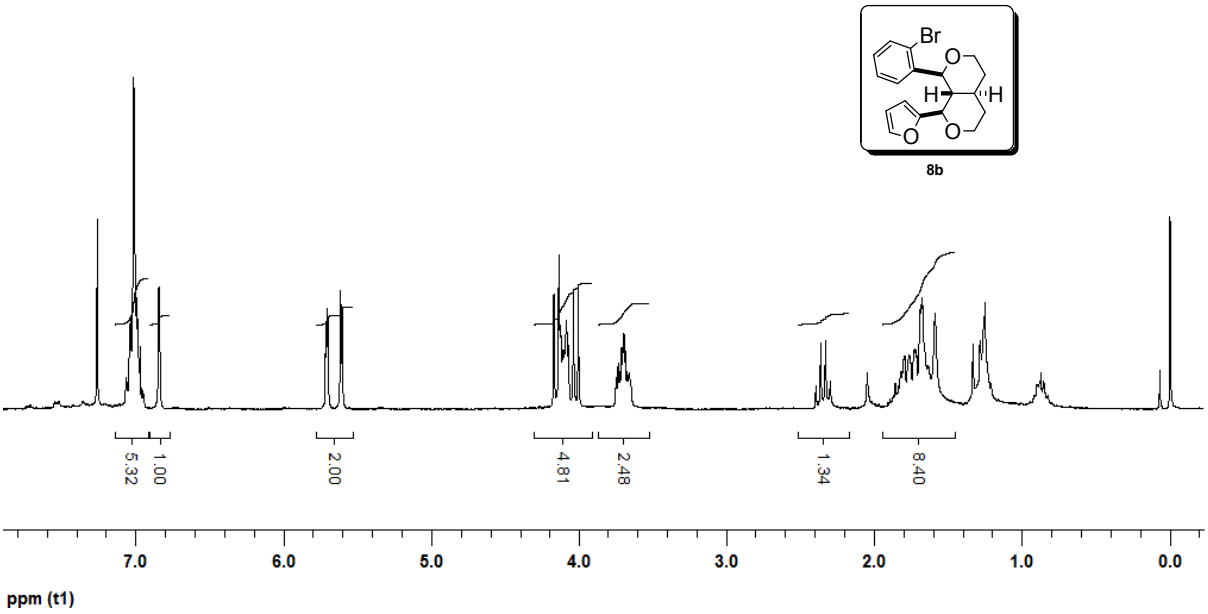
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 7h



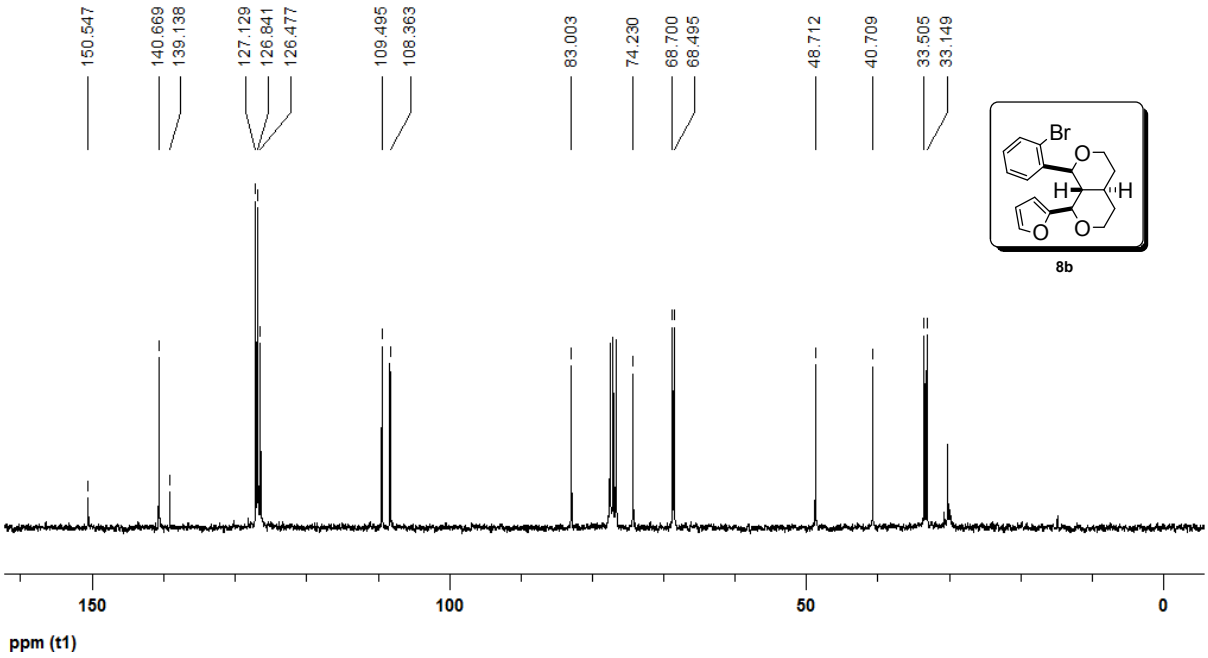
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 8a



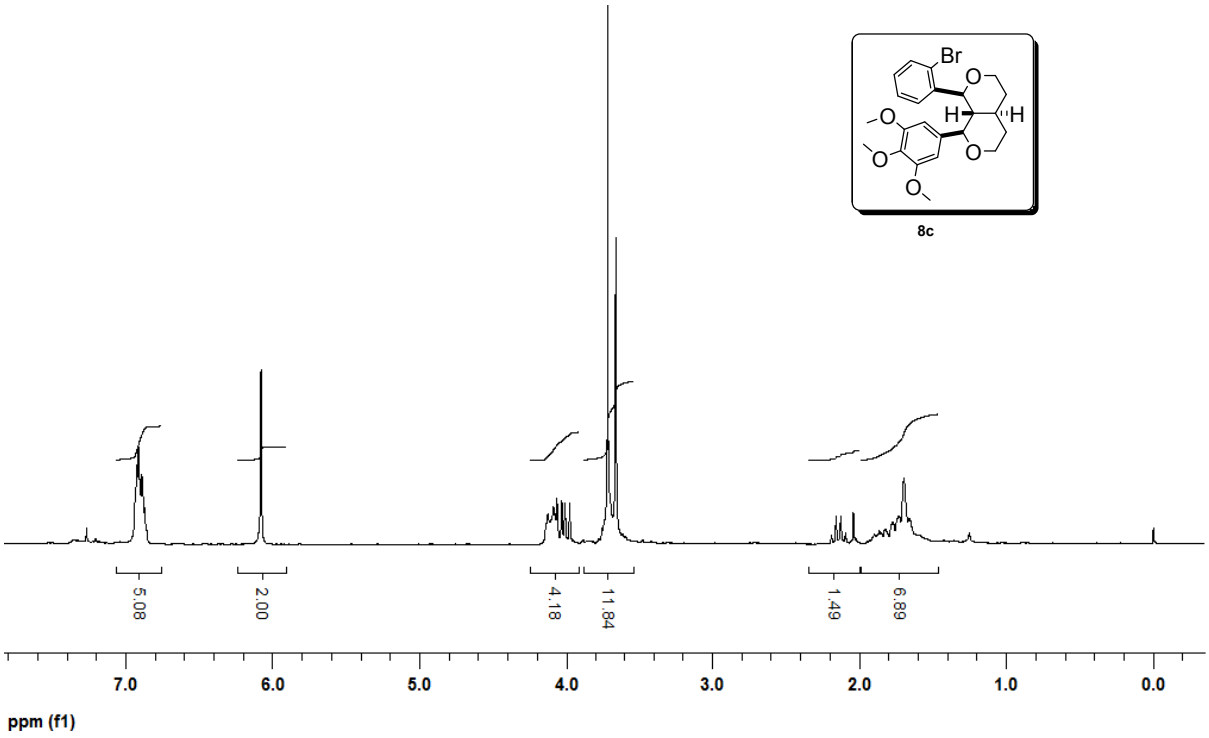
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 8a



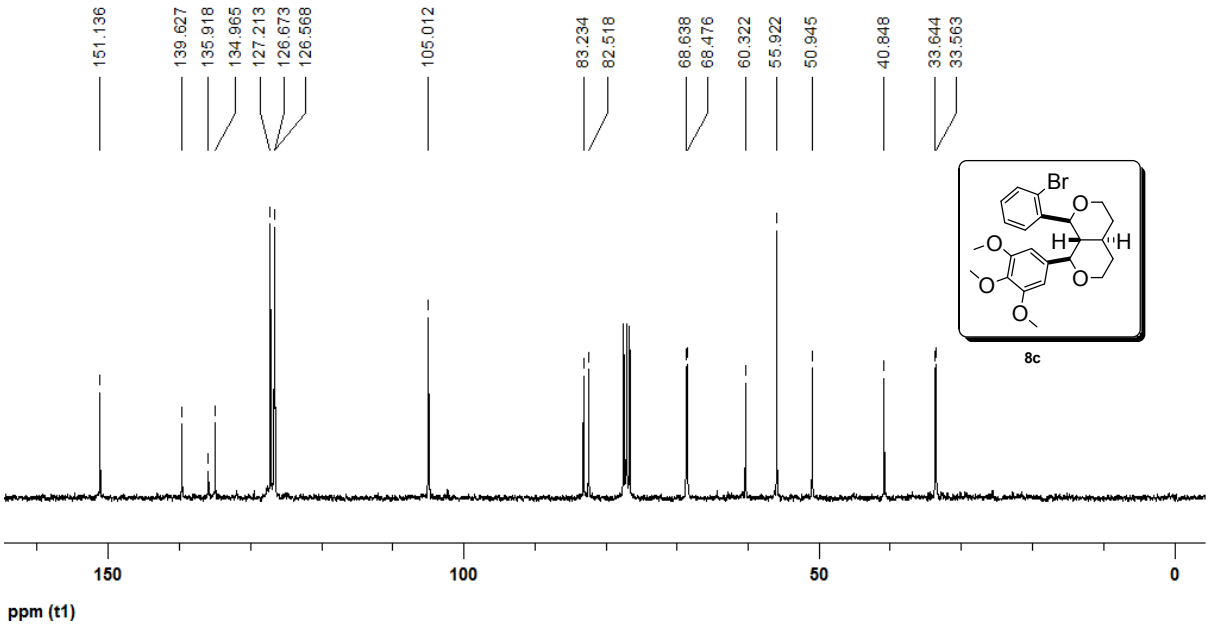
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 8b



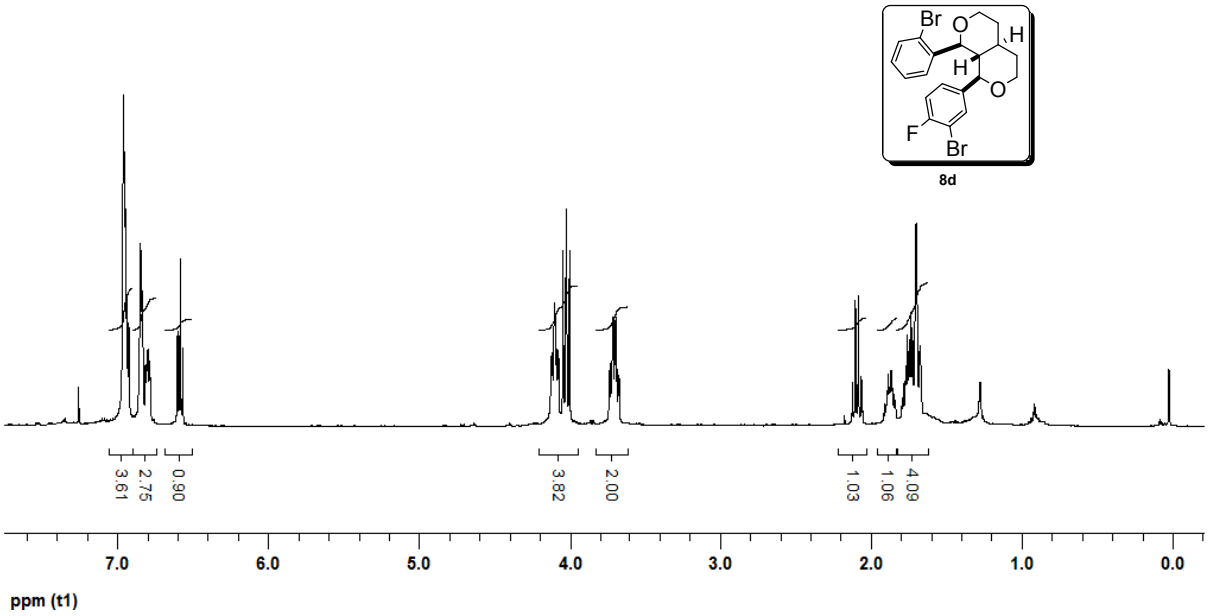
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 8b



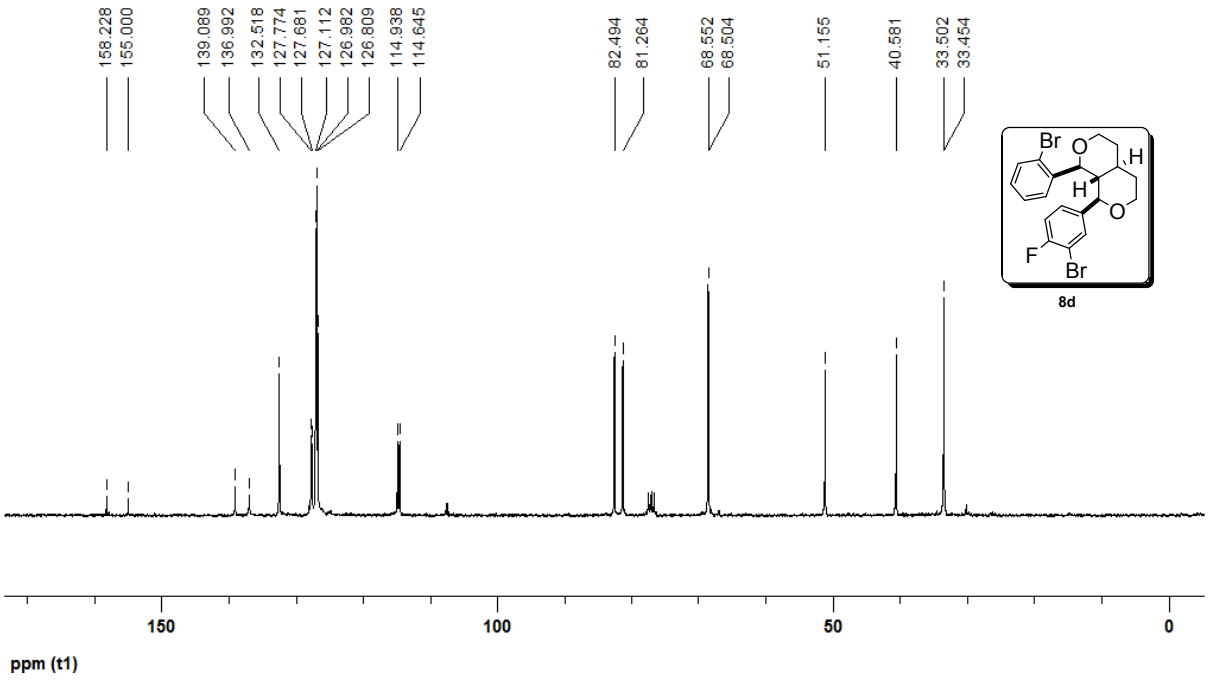
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 8c



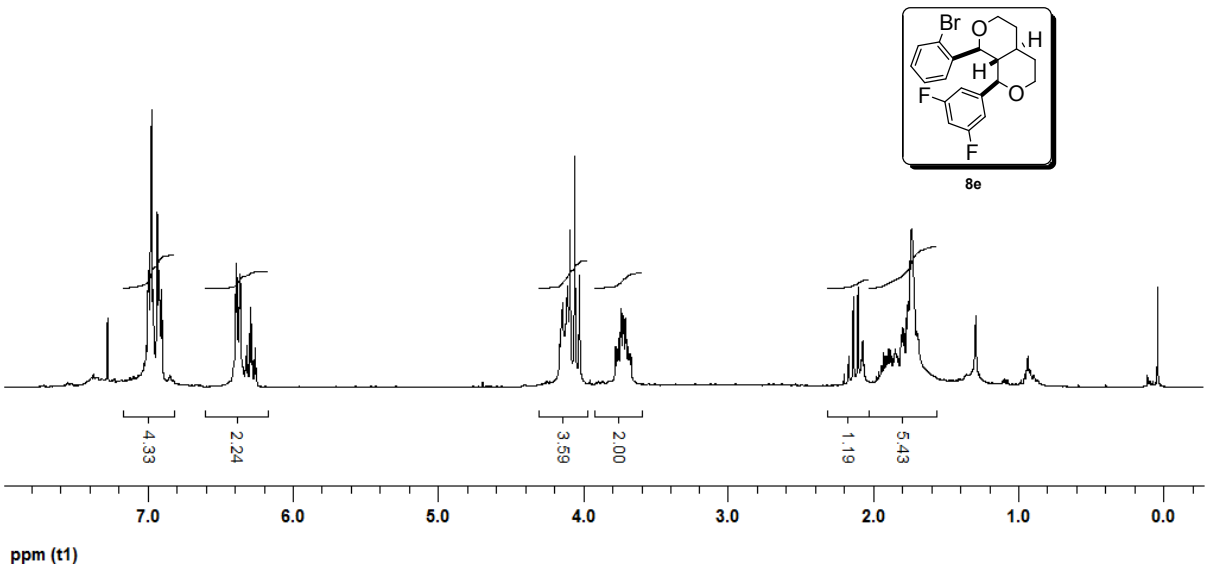
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 8c



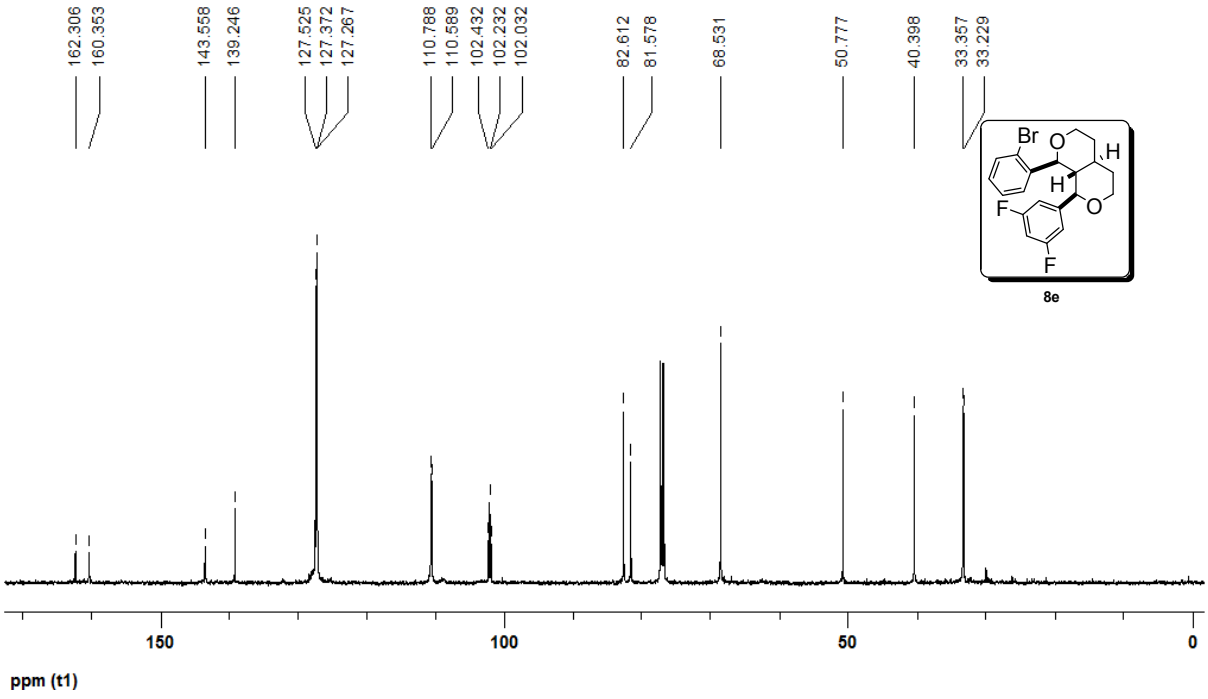
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 8d



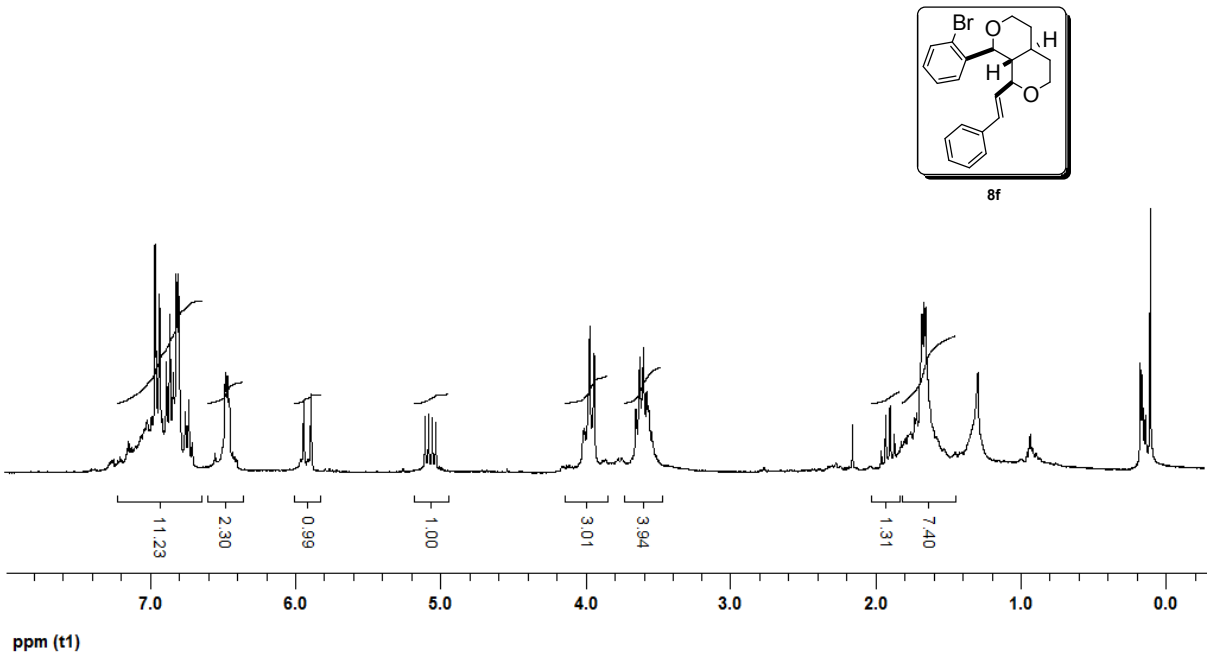
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 8d



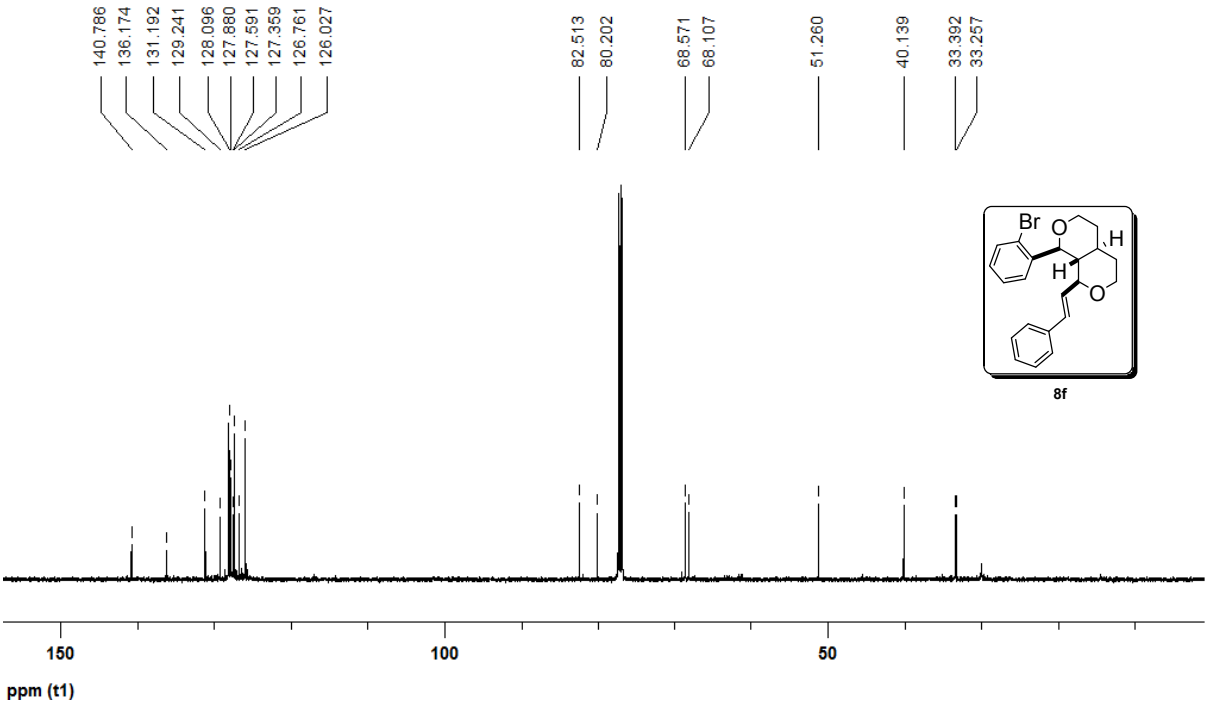
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 8e



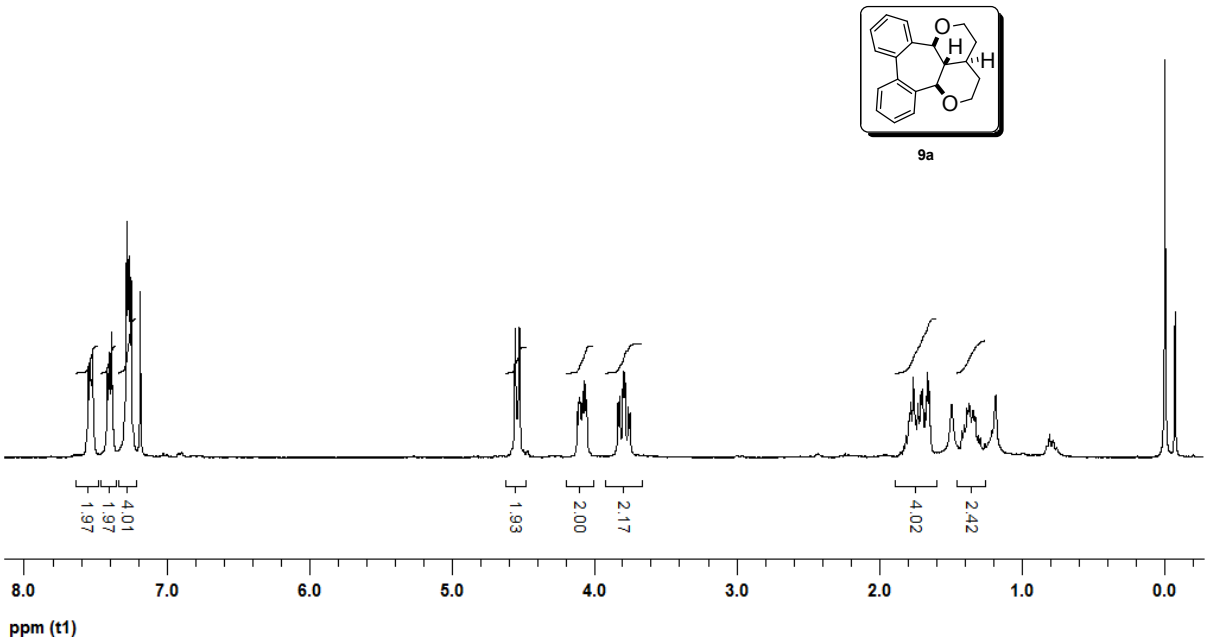
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 8e



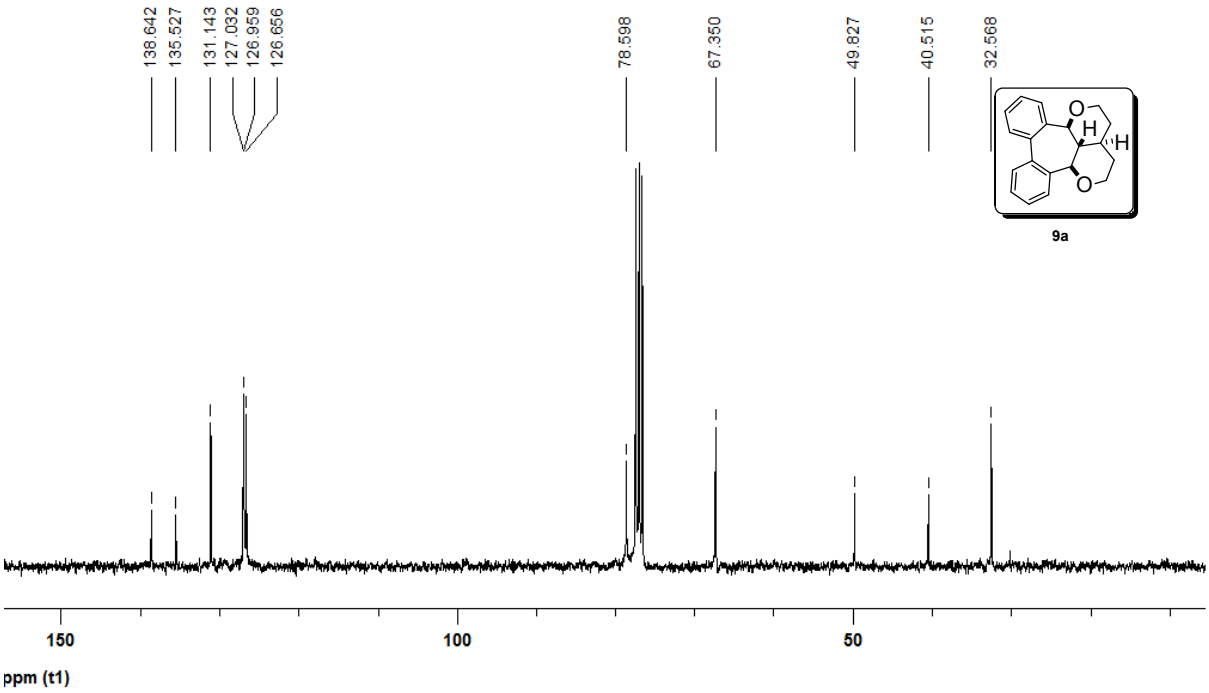
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 8f



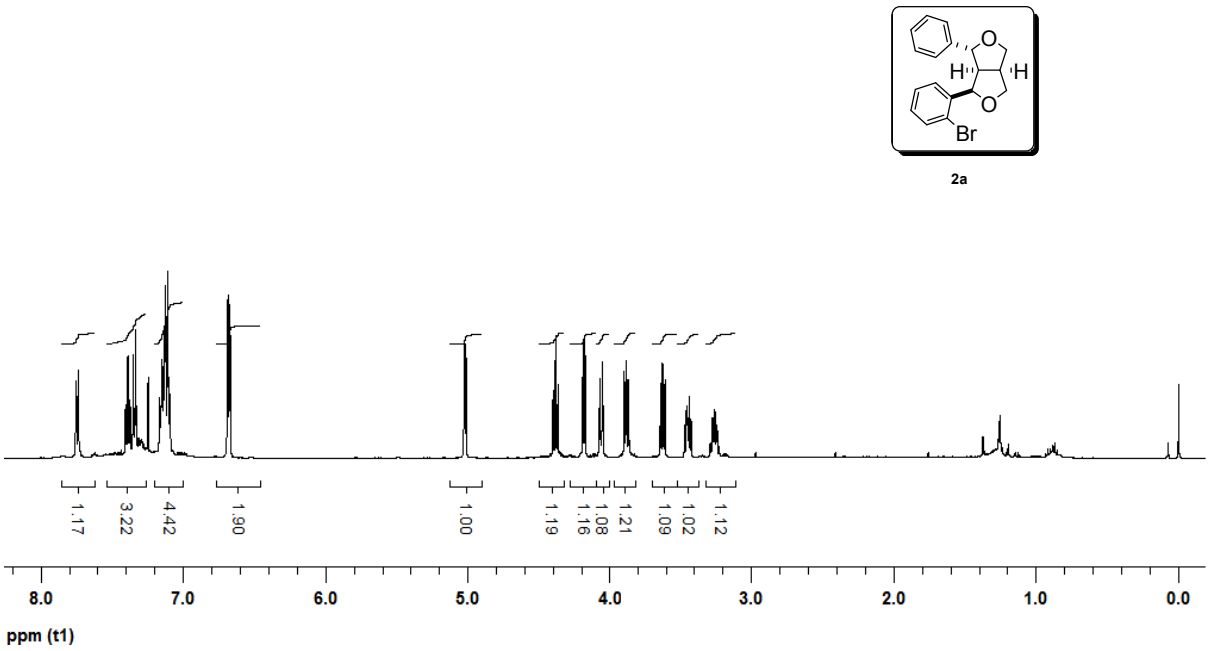
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 8f



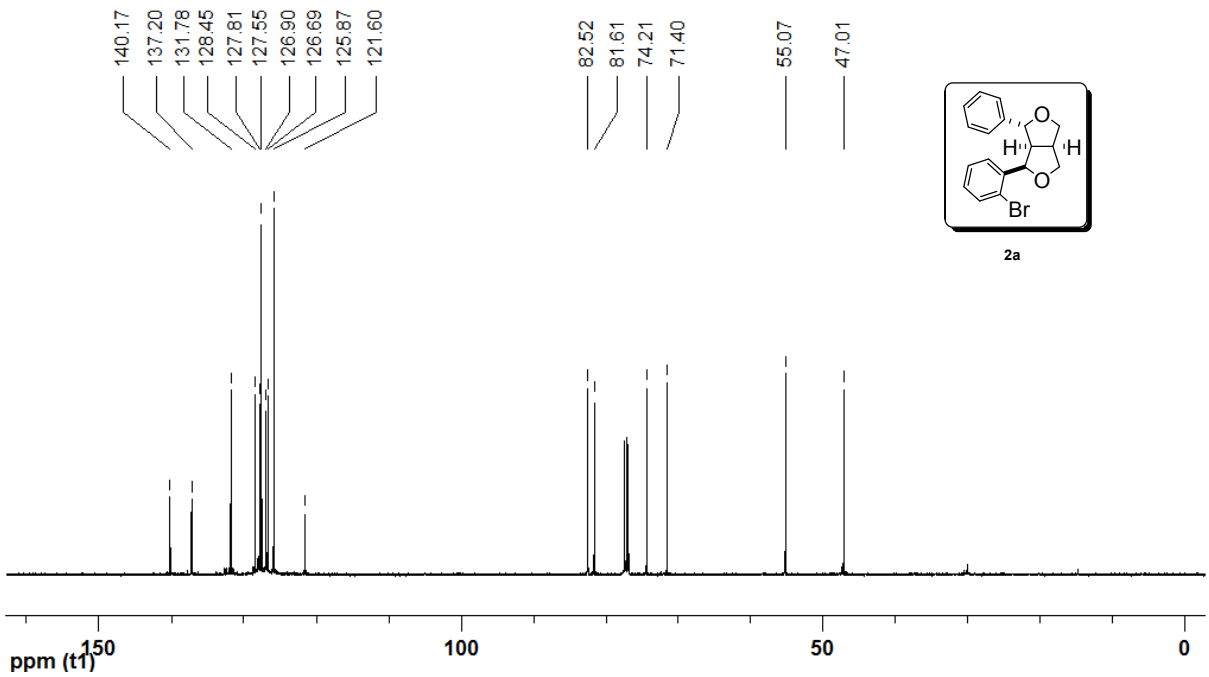
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 9a



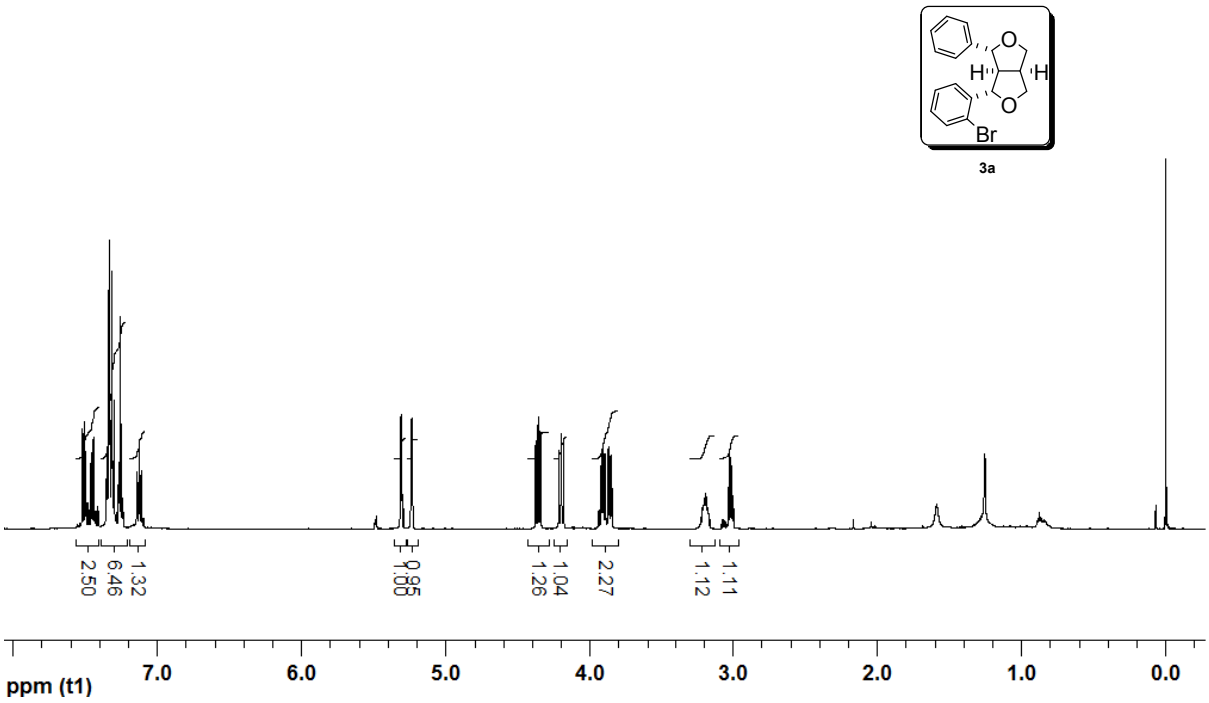
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 9a



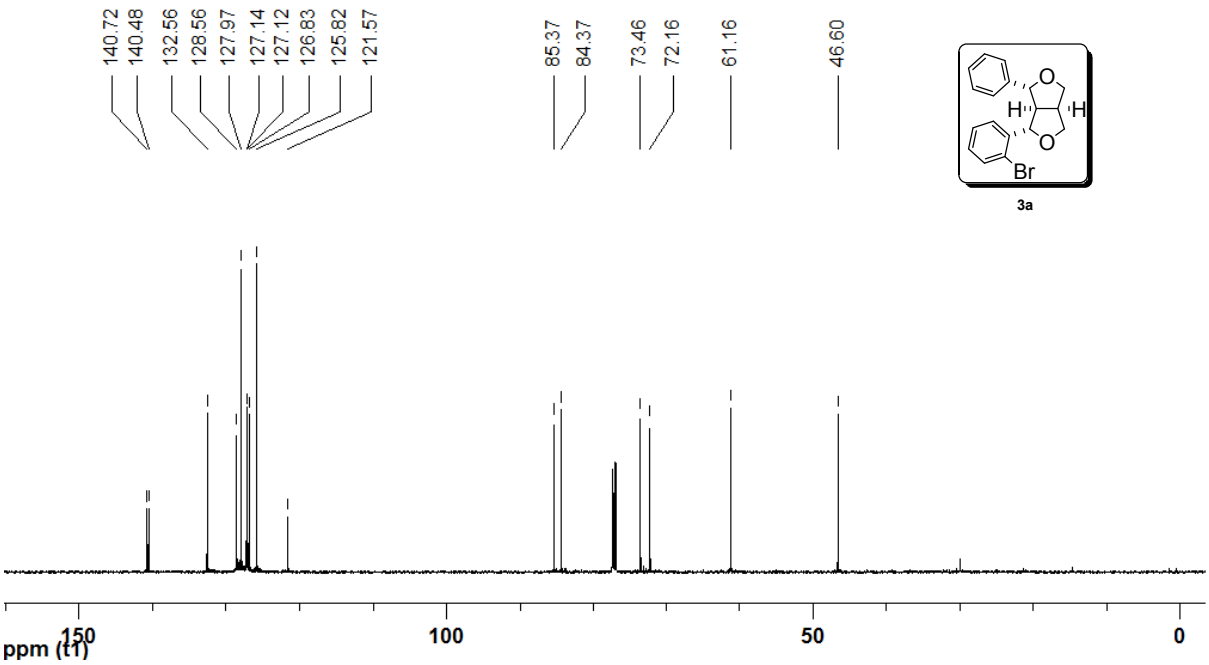
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 2a



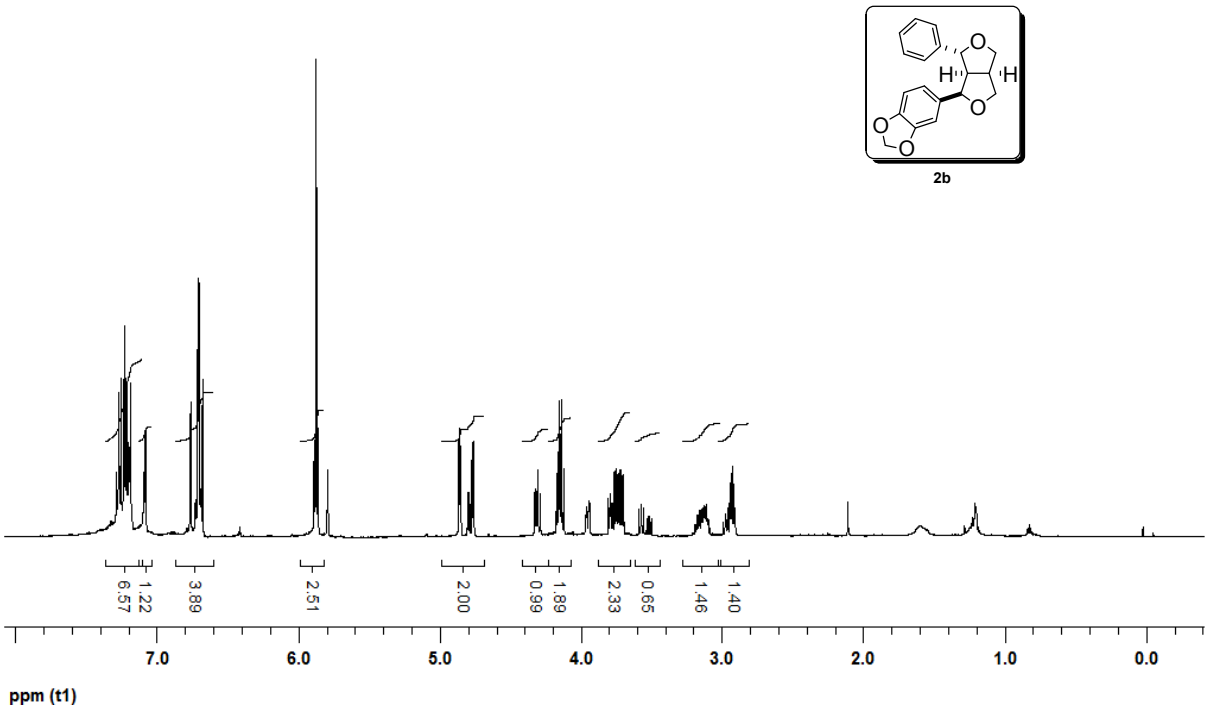
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 2a



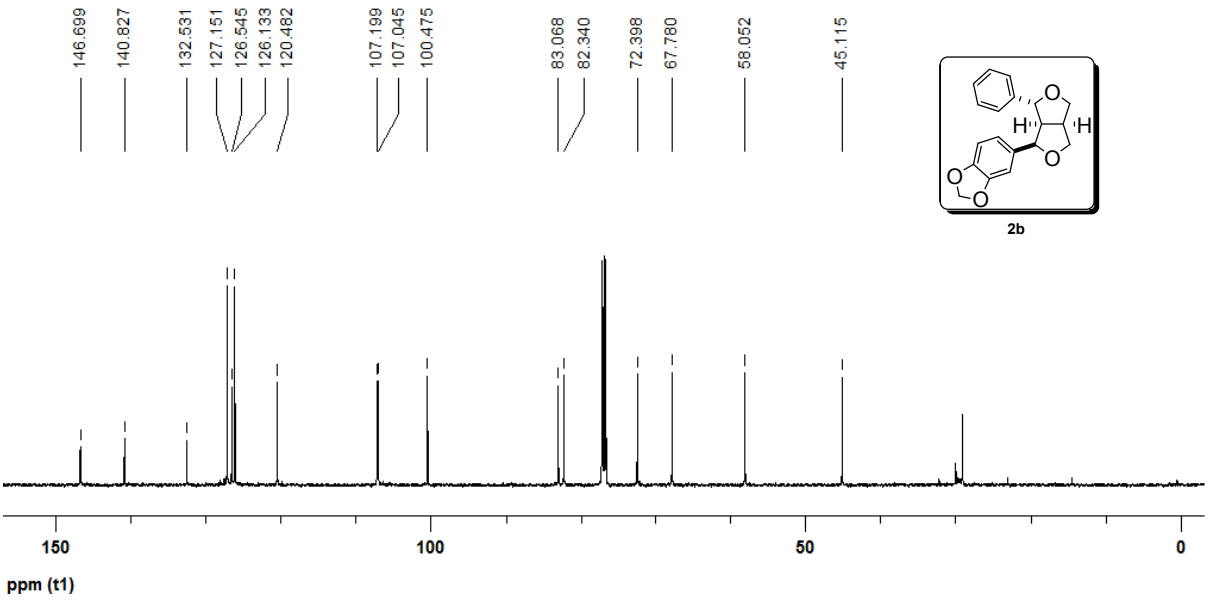
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 3a



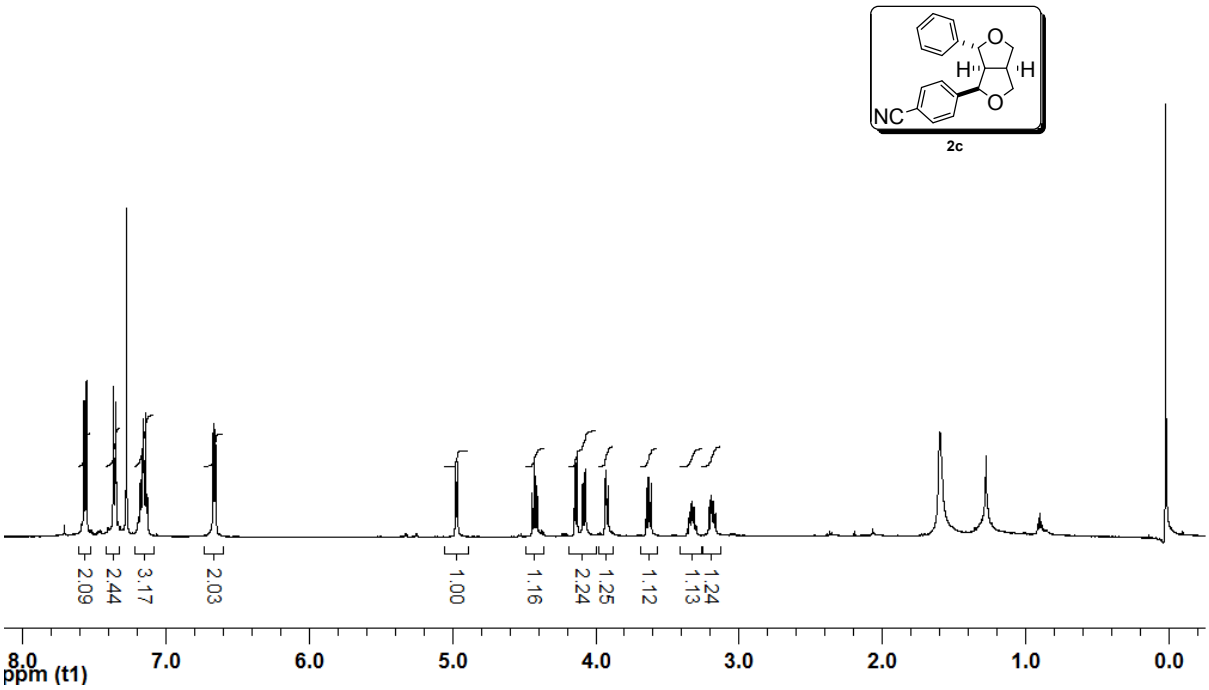
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 3a



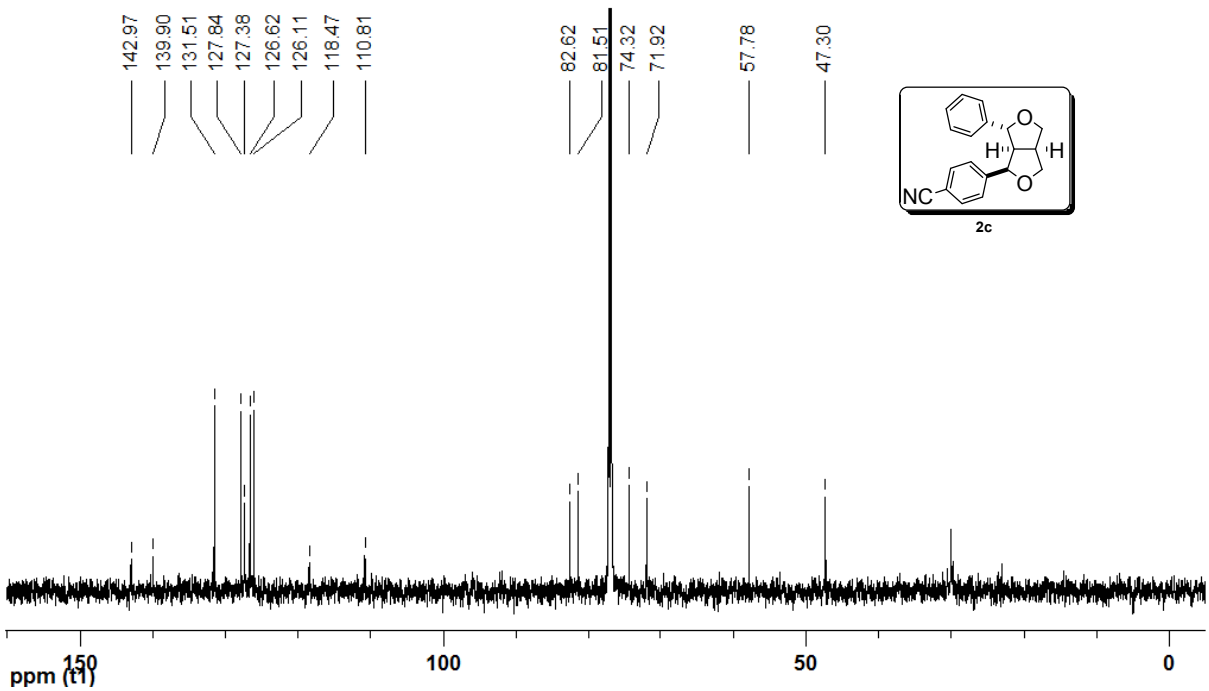
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2b



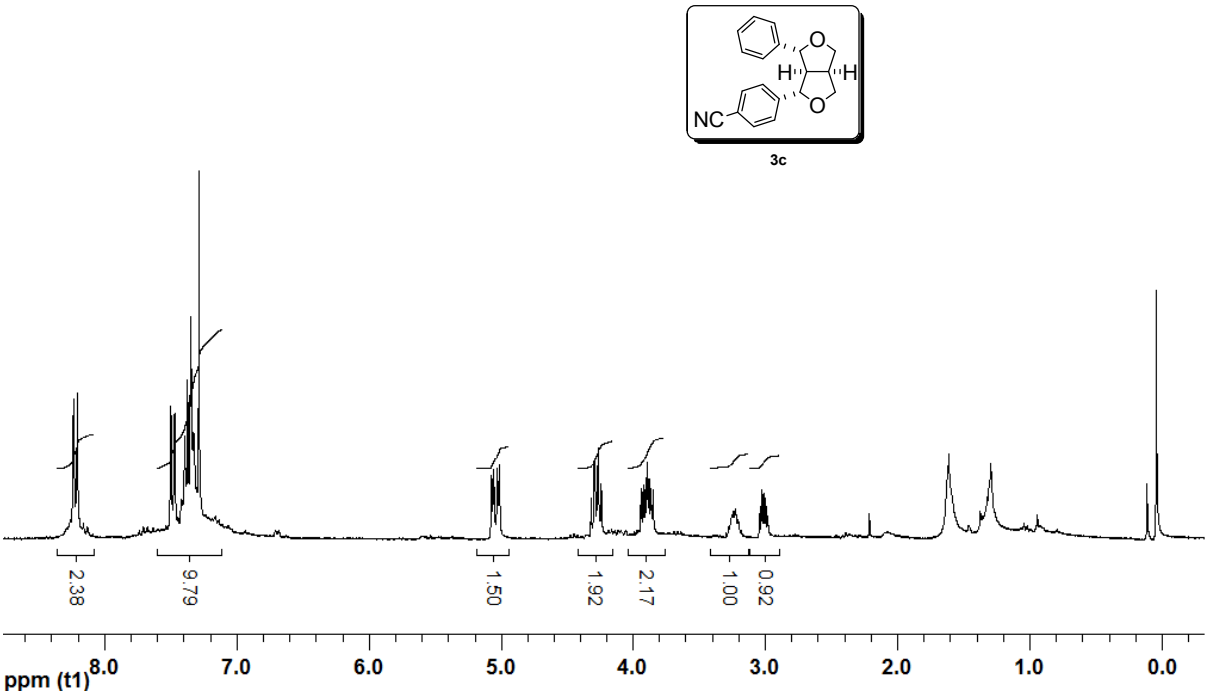
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 2b



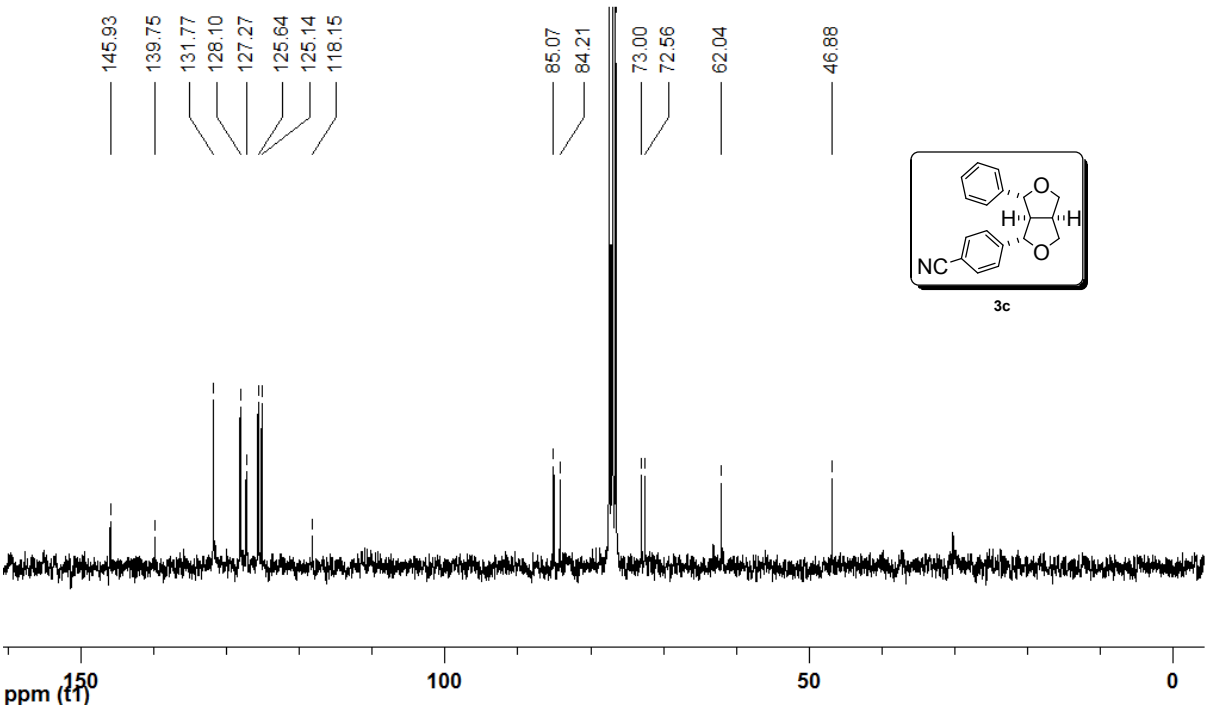
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2c



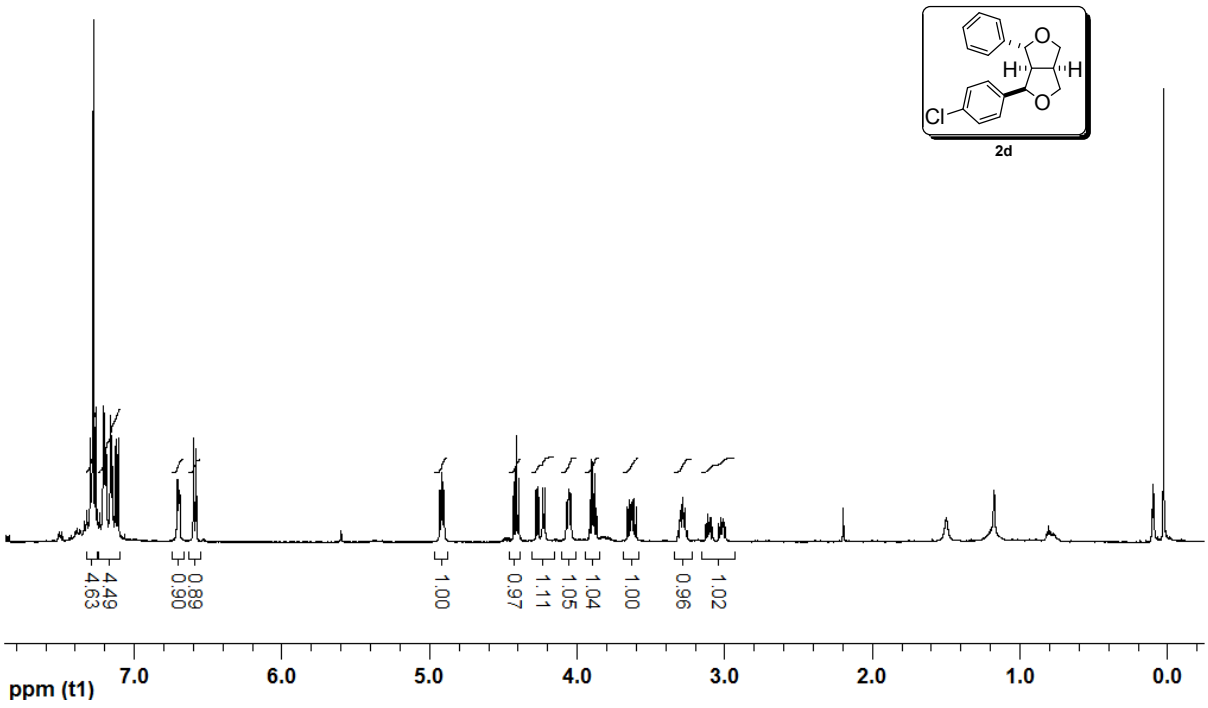
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 2c



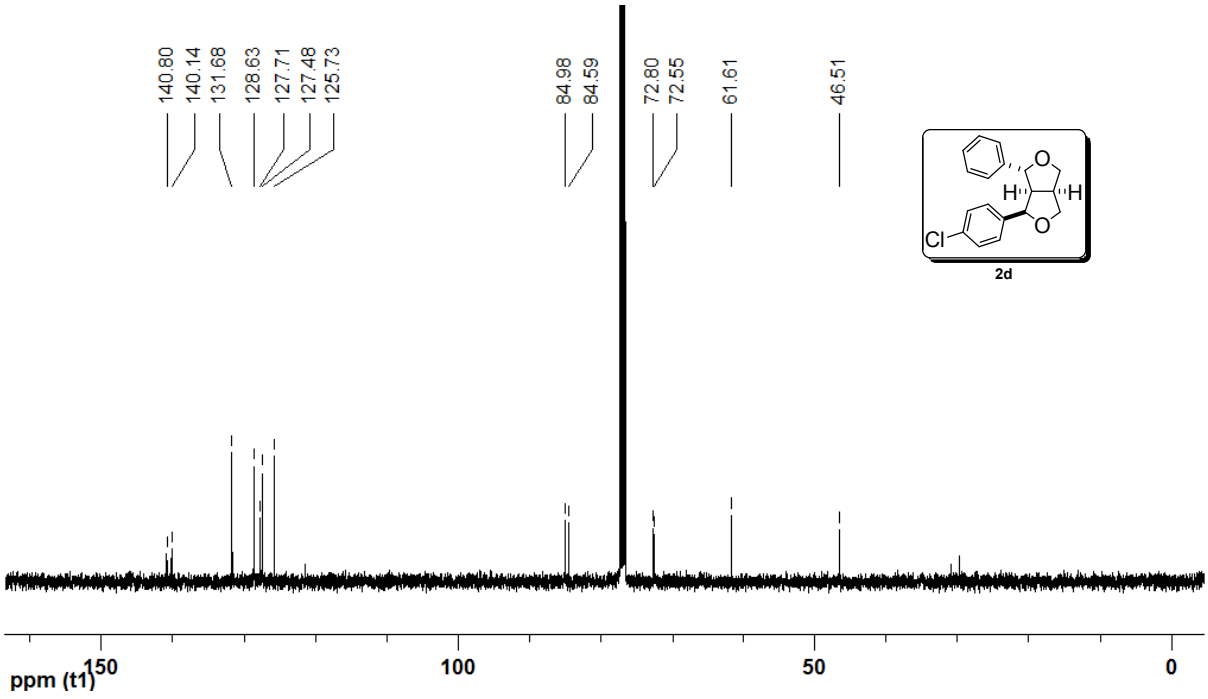
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 3c



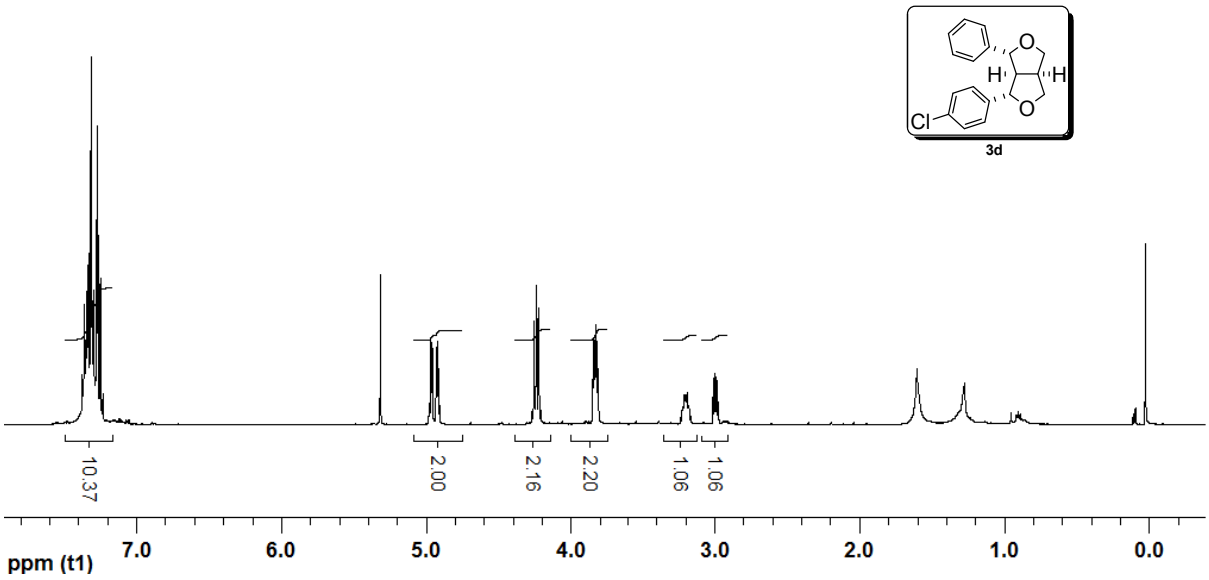
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 3c



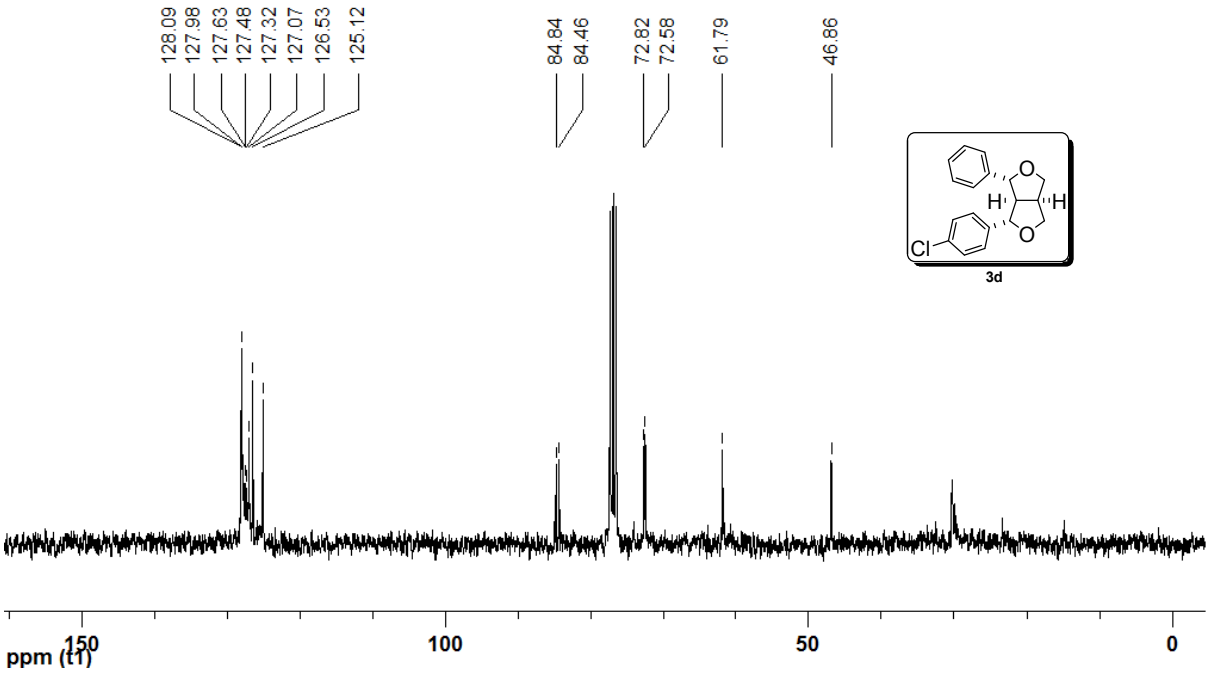
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2d



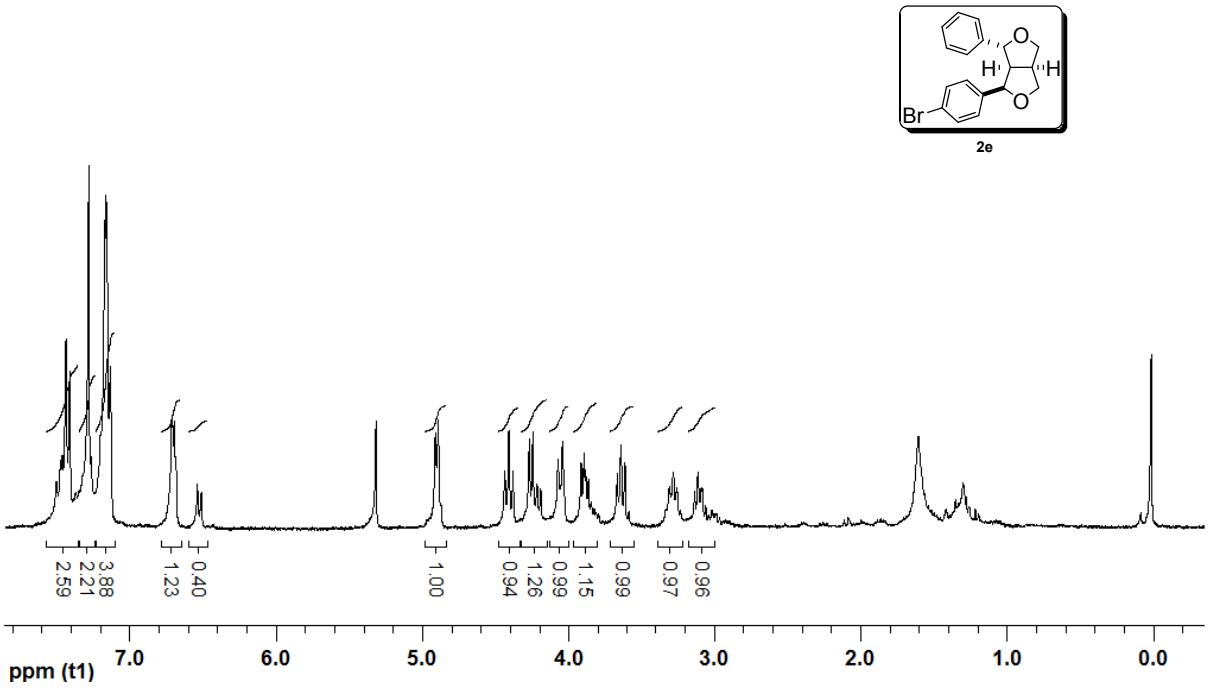
^{13}C NMR (100 MHz, $CDCl_3$) spectrum of compound 2d



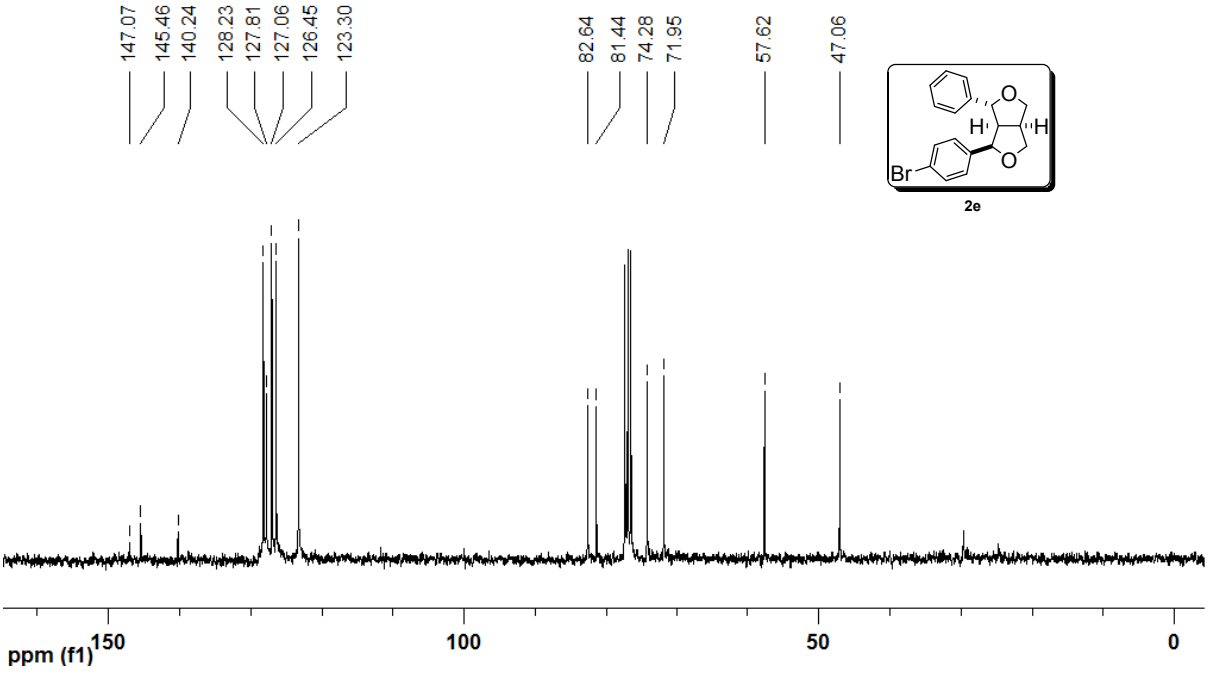
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 3d



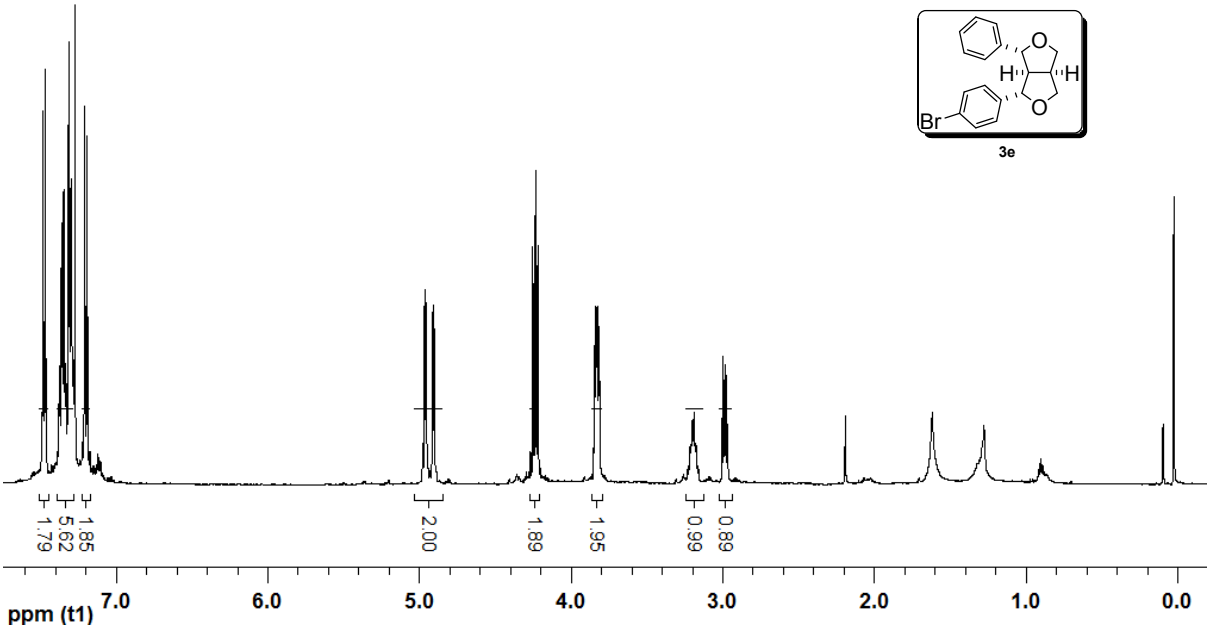
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 3d



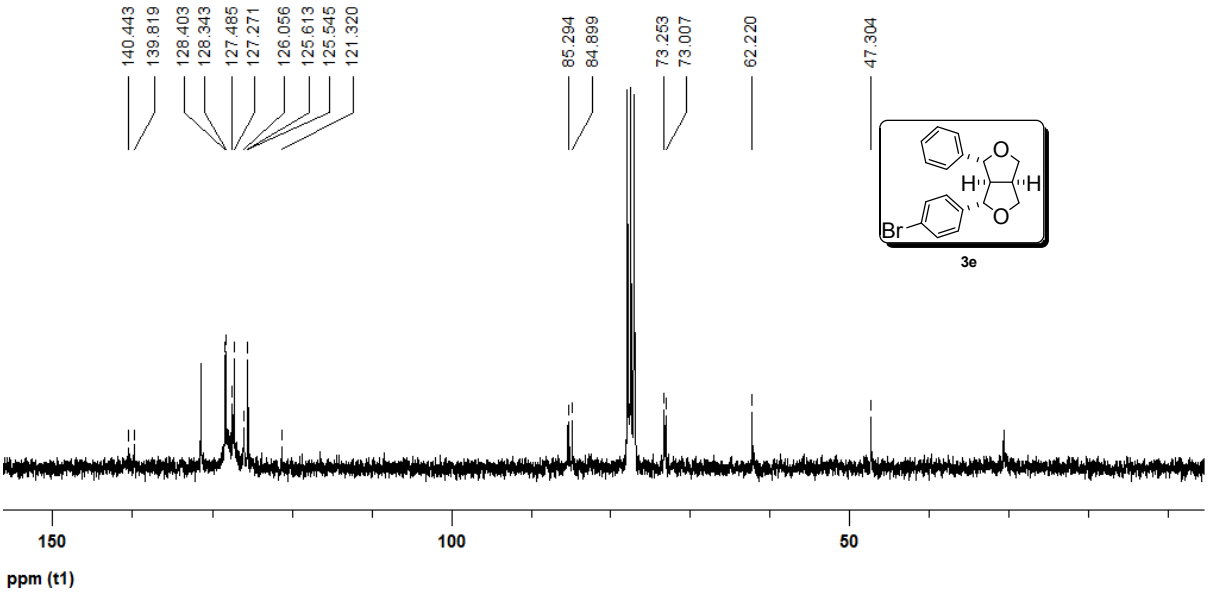
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 2e



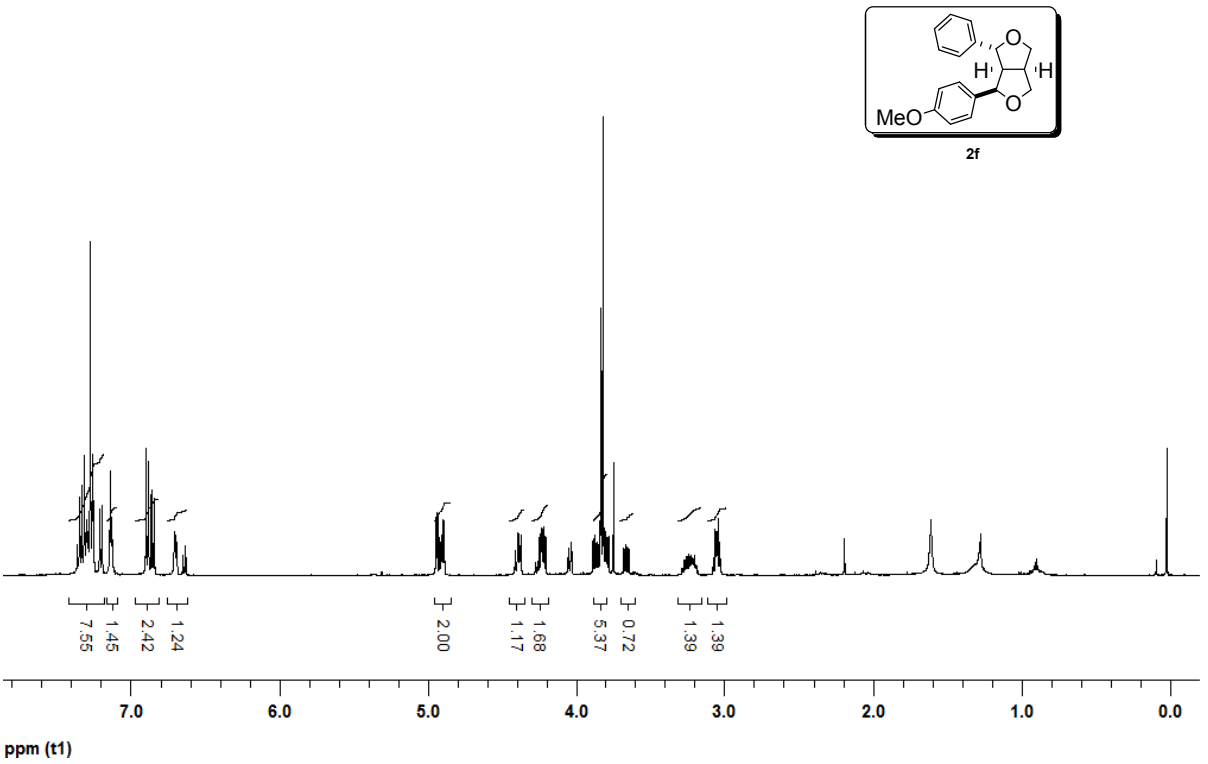
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 2e



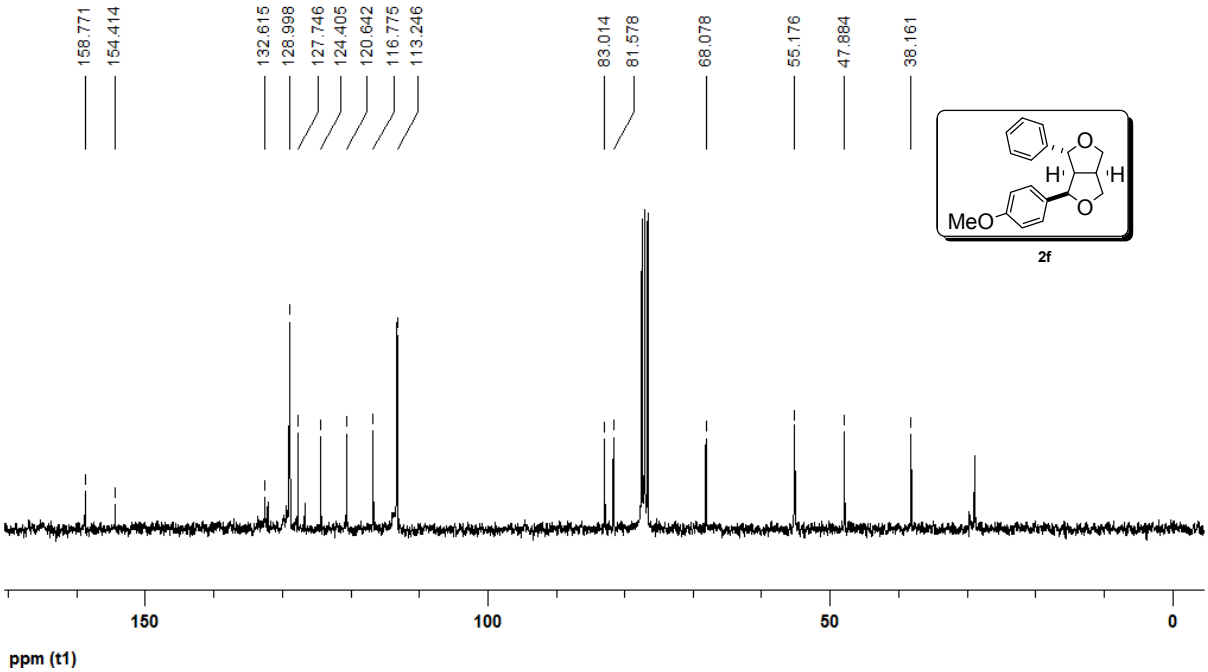
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 3e



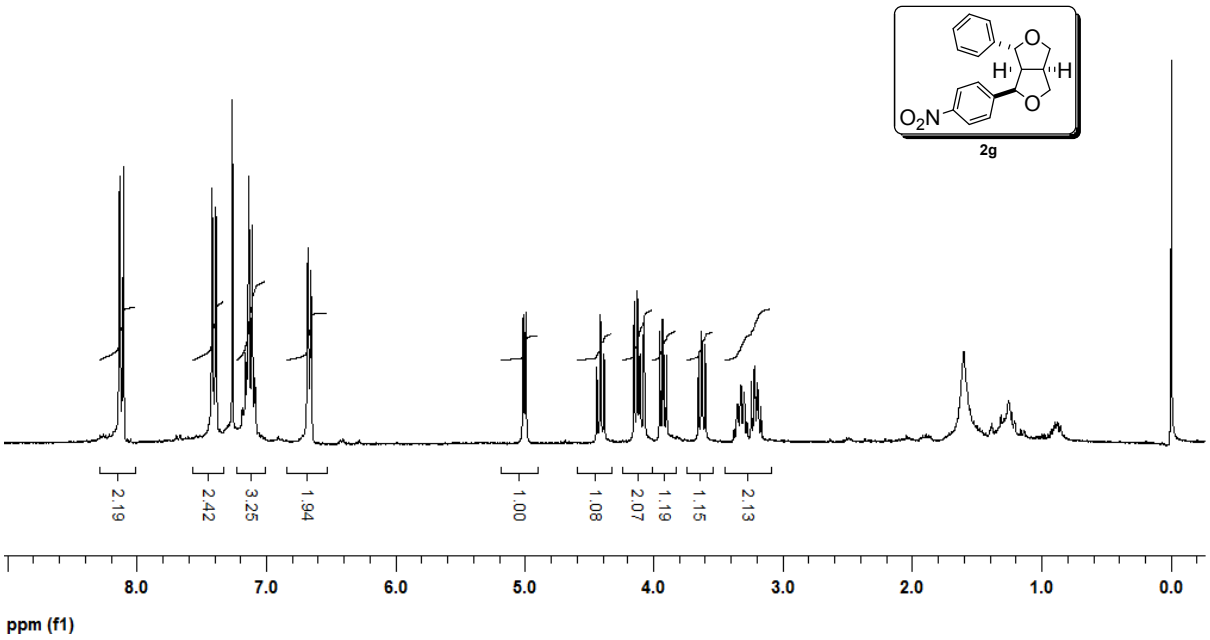
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 3e



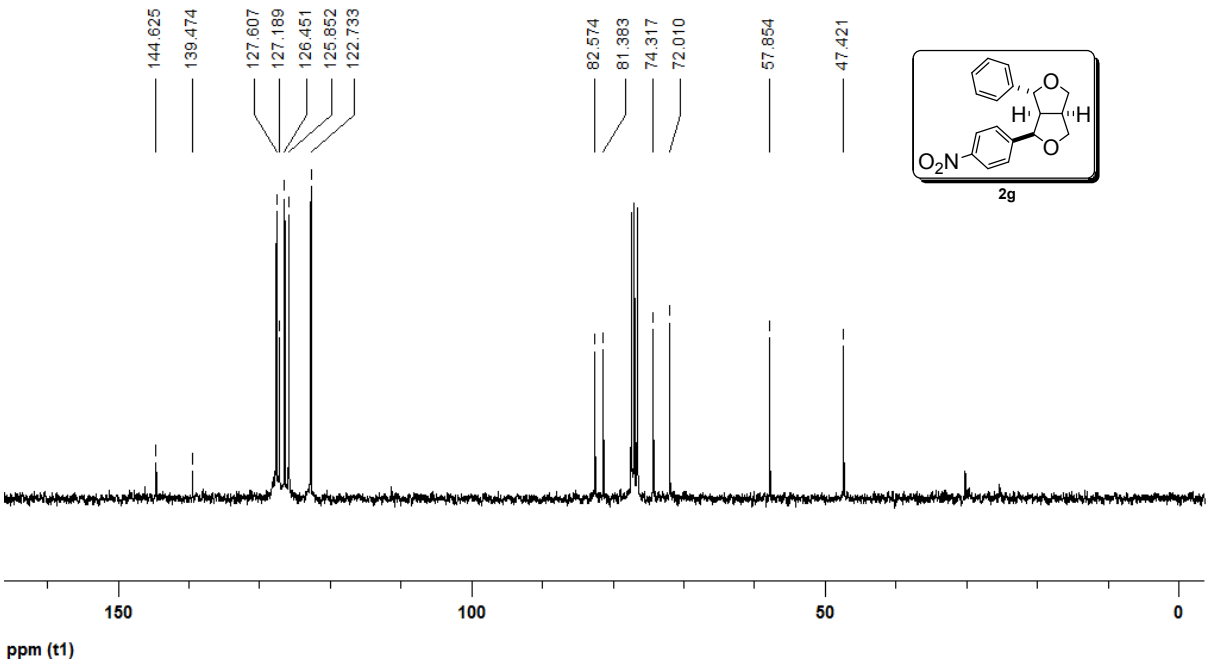
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2f



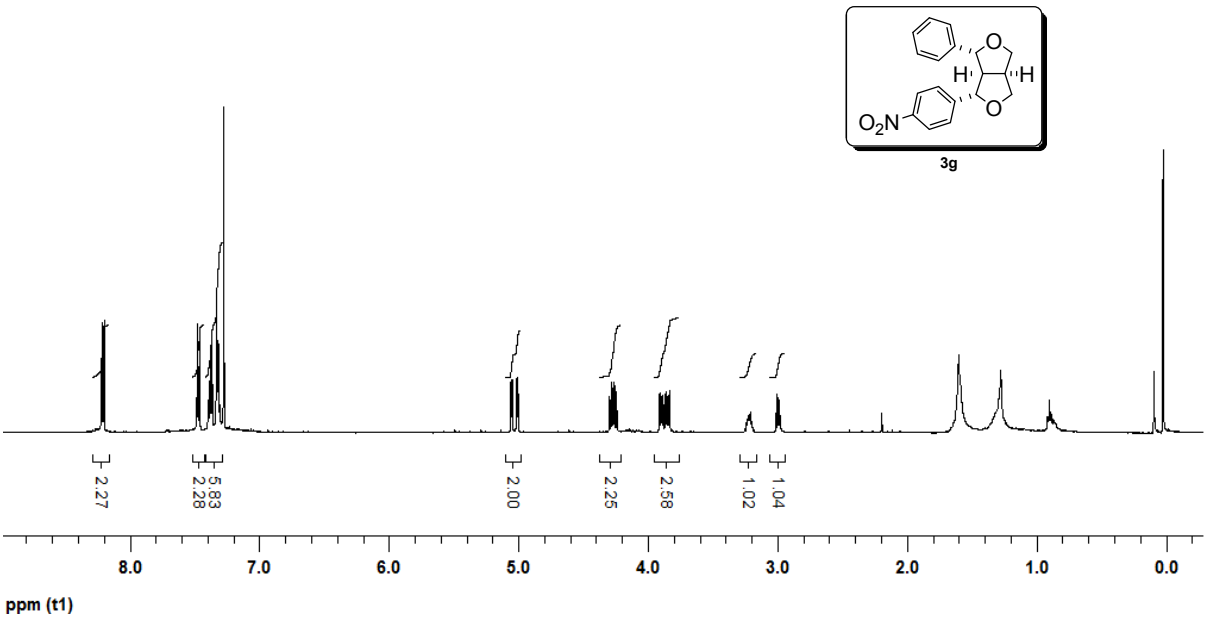
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 2f



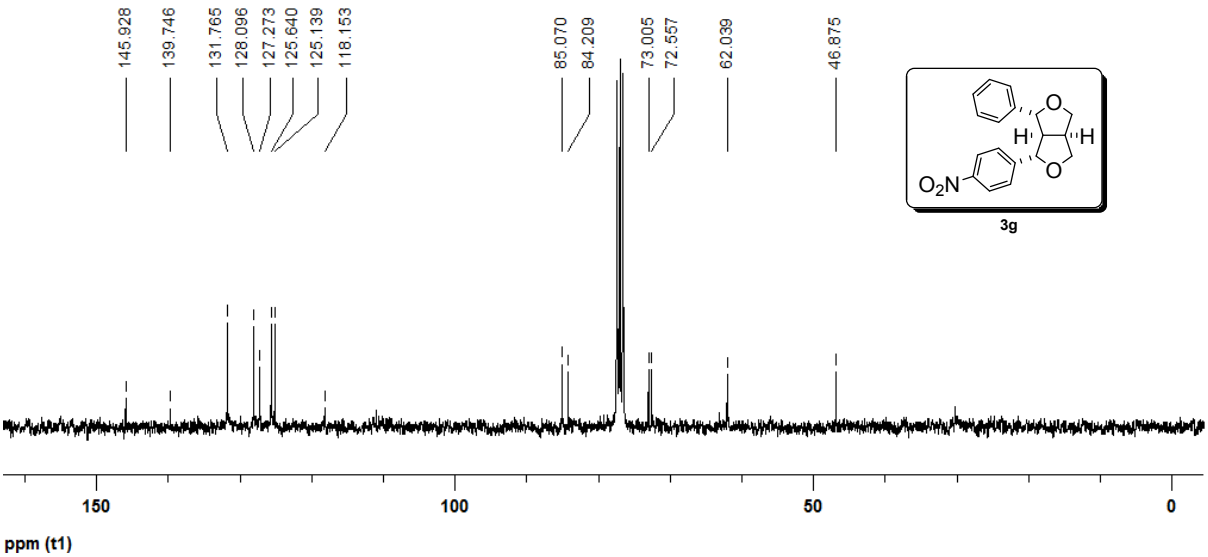
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 2g



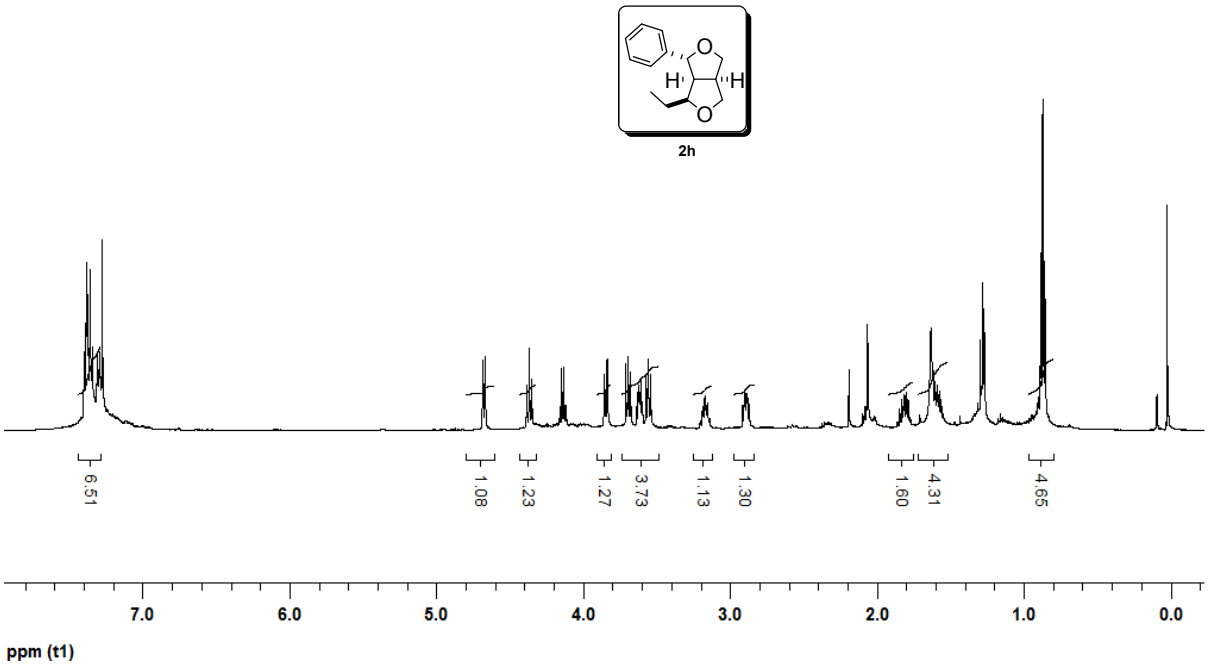
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 2g



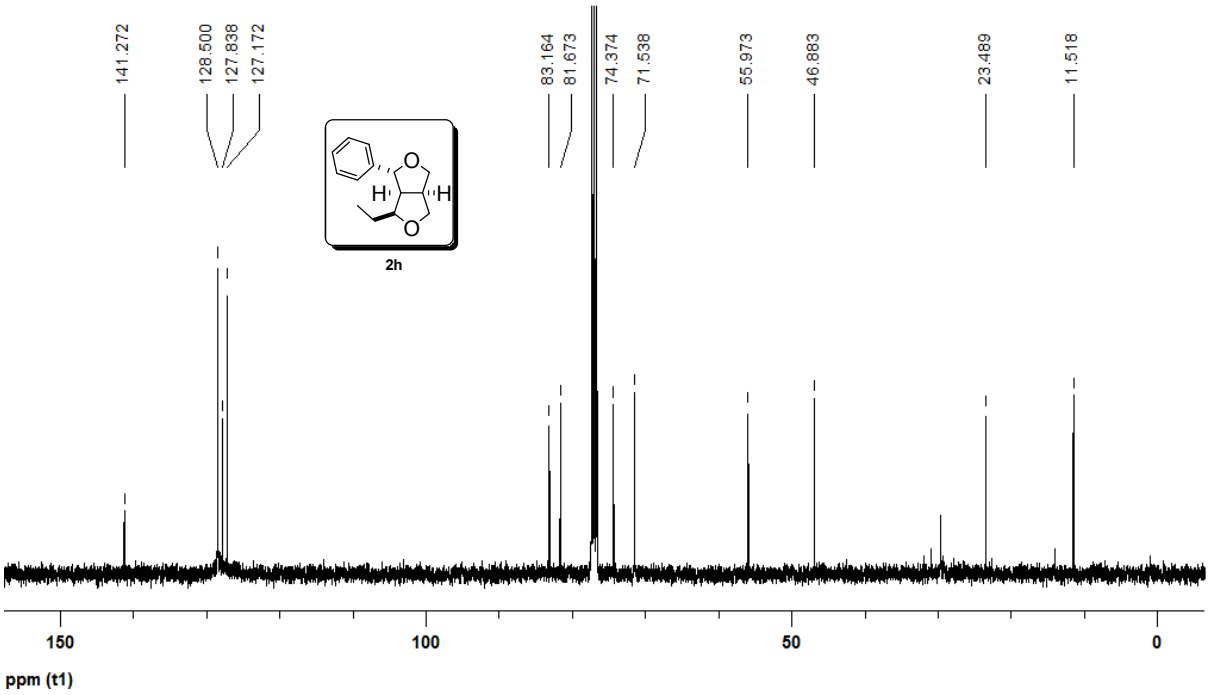
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 3g



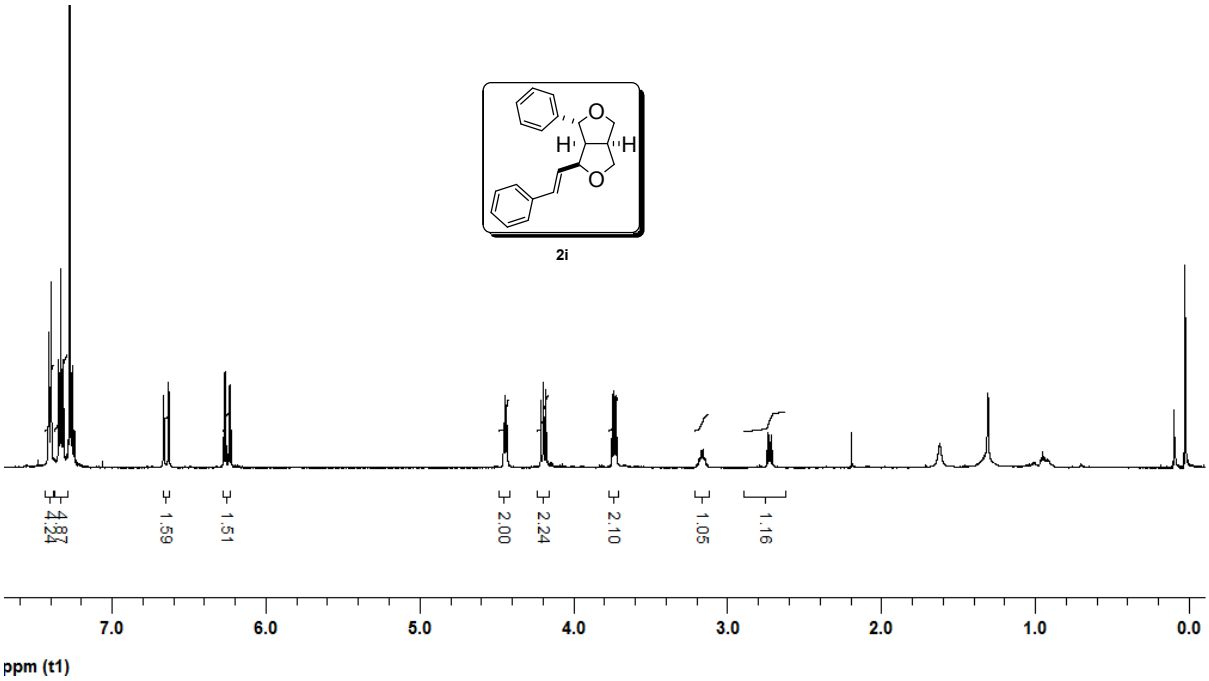
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 3g



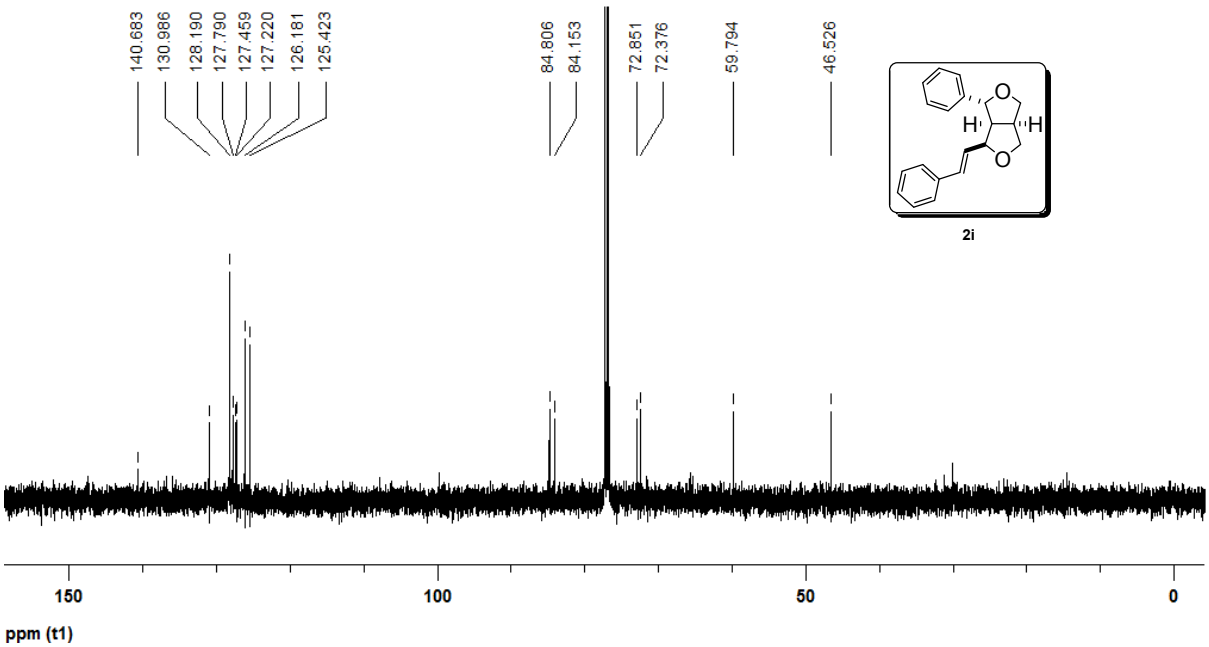
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2h



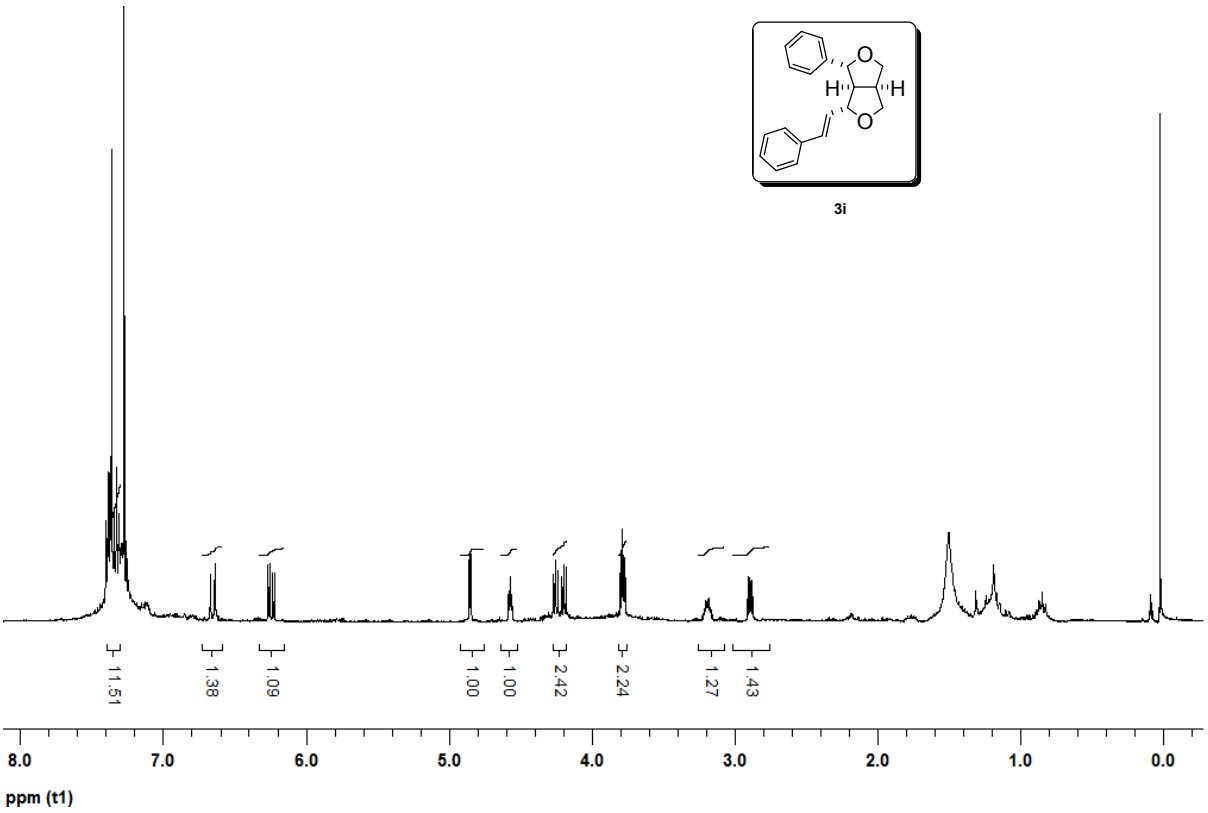
^{13}C NMR (100 MHz, $CDCl_3$) spectrum of compound 2h



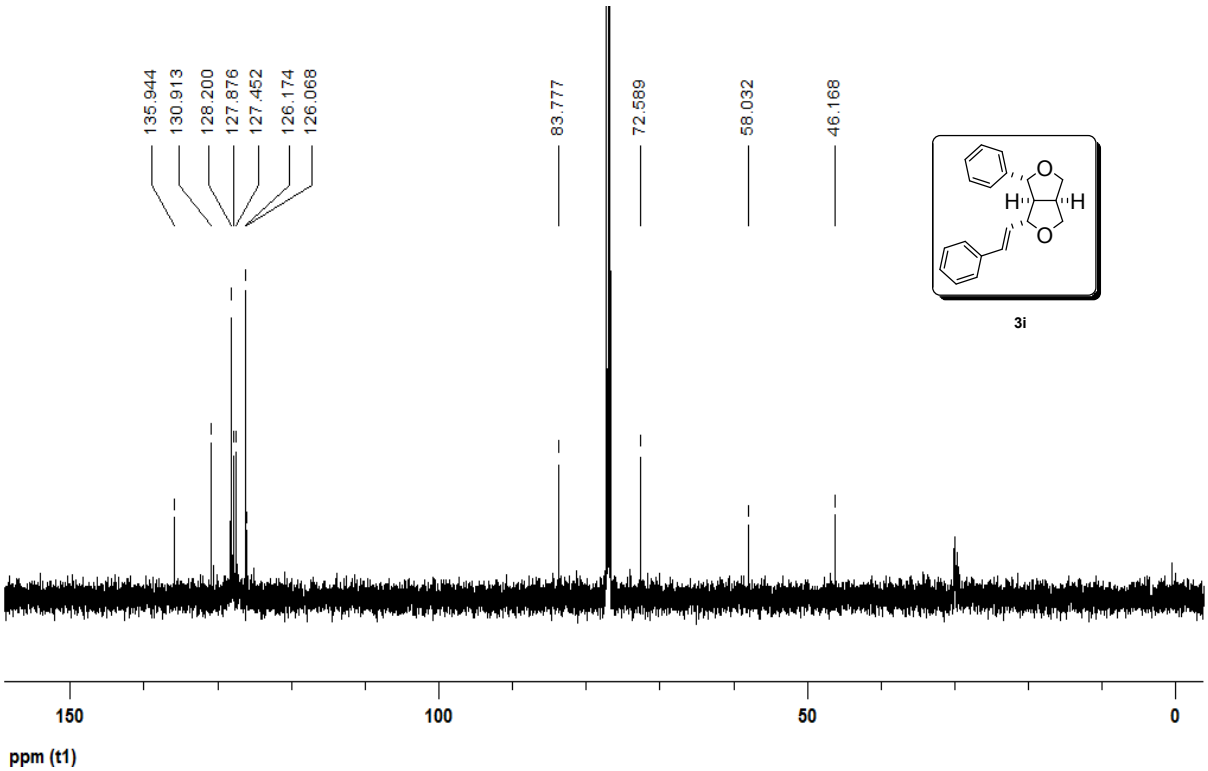
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2i



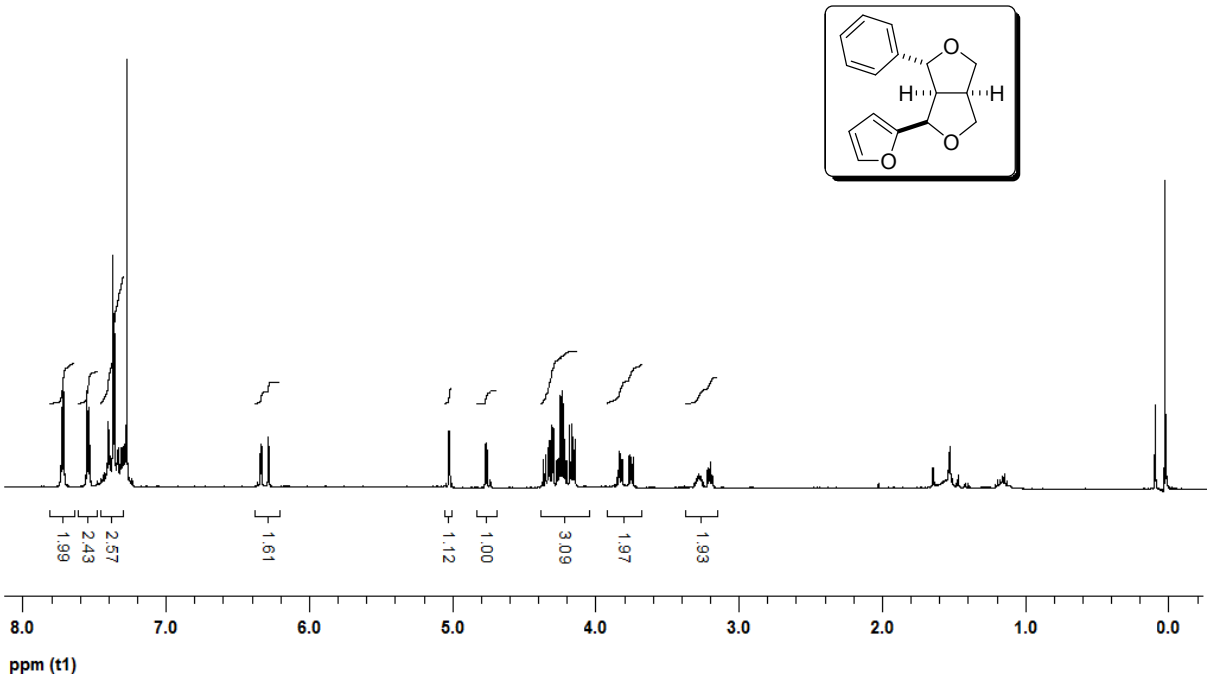
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 2i



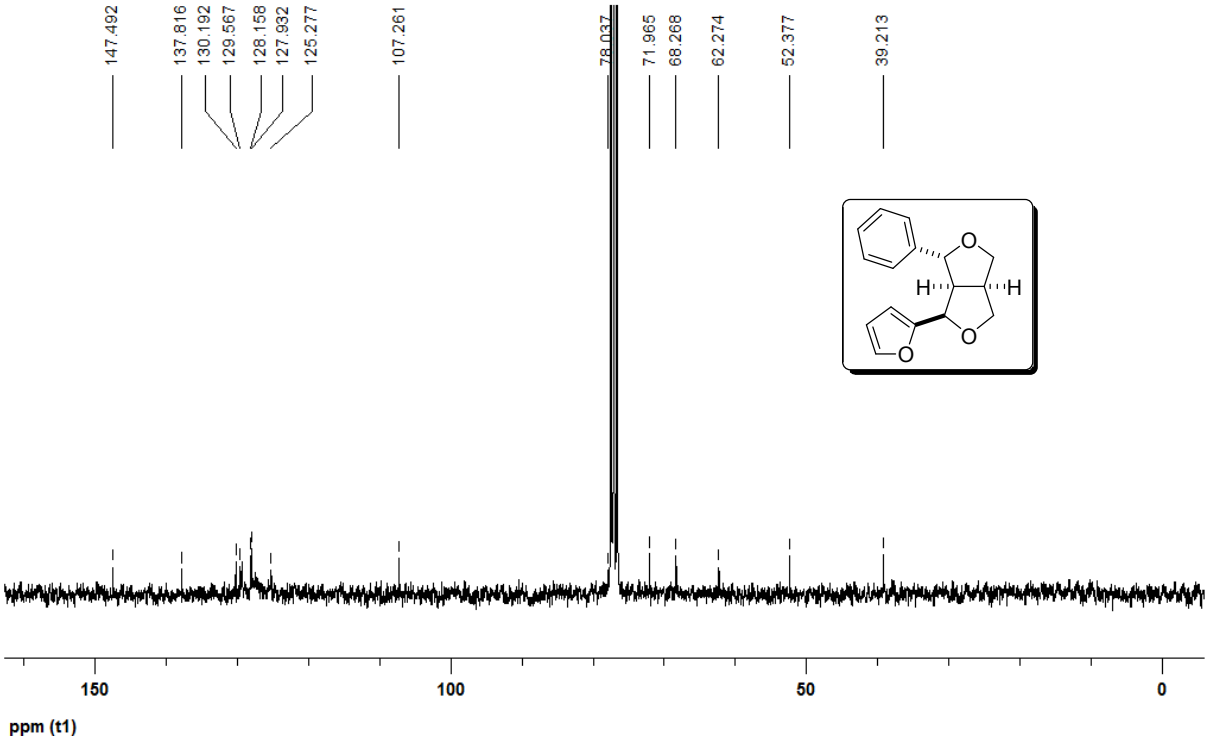
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 3i



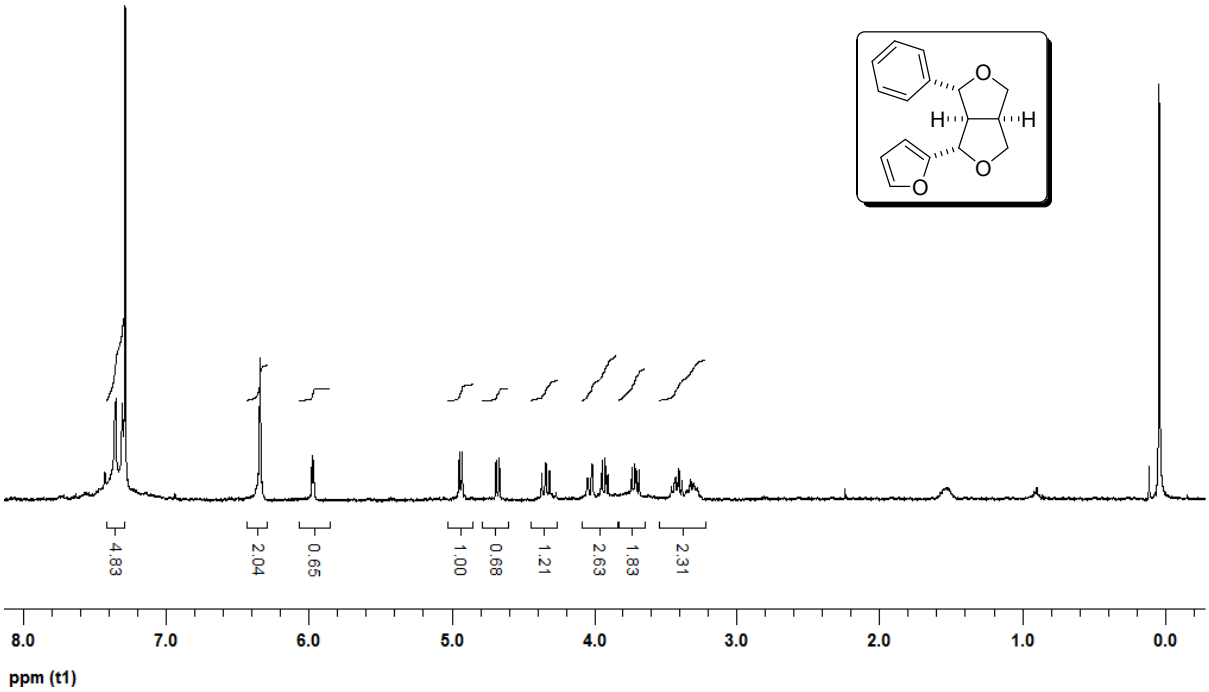
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 3i



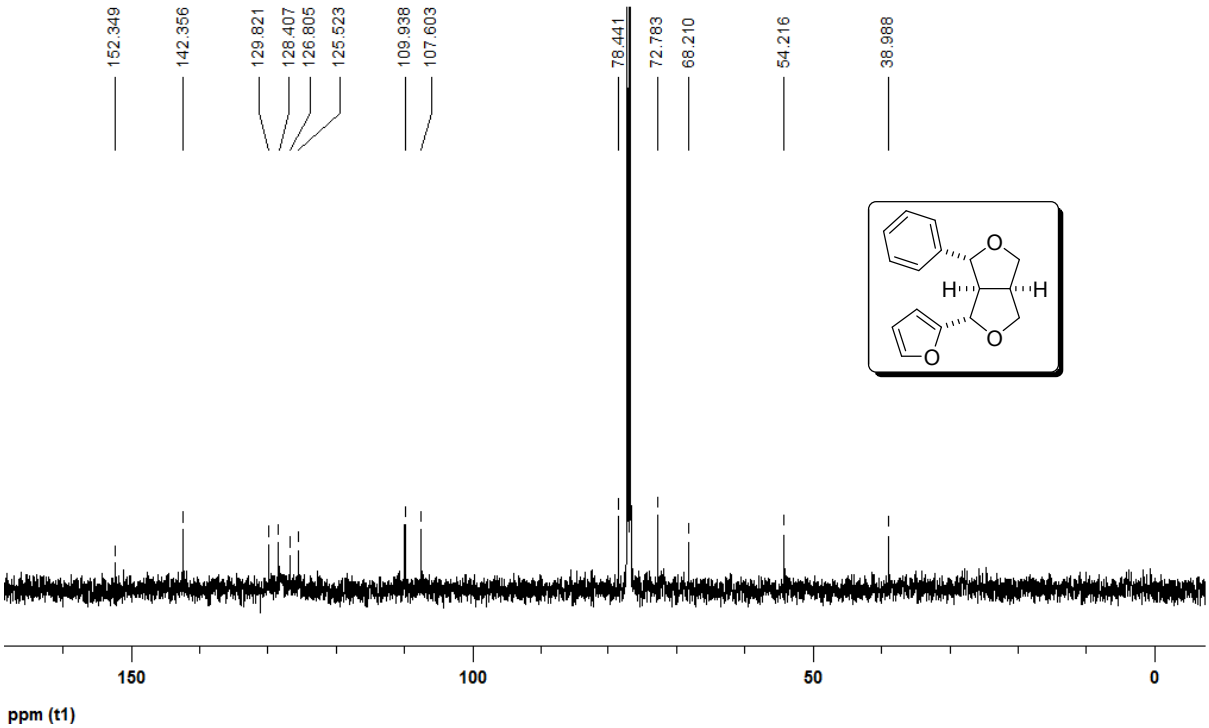
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 2j



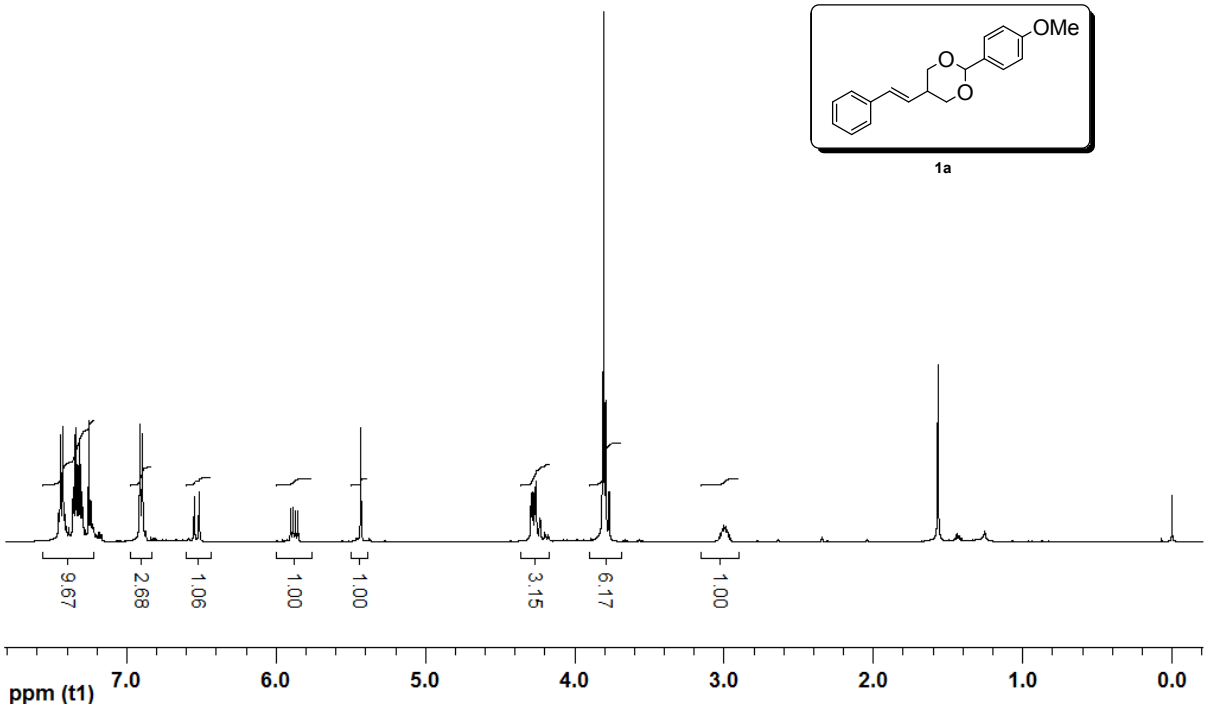
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 2j



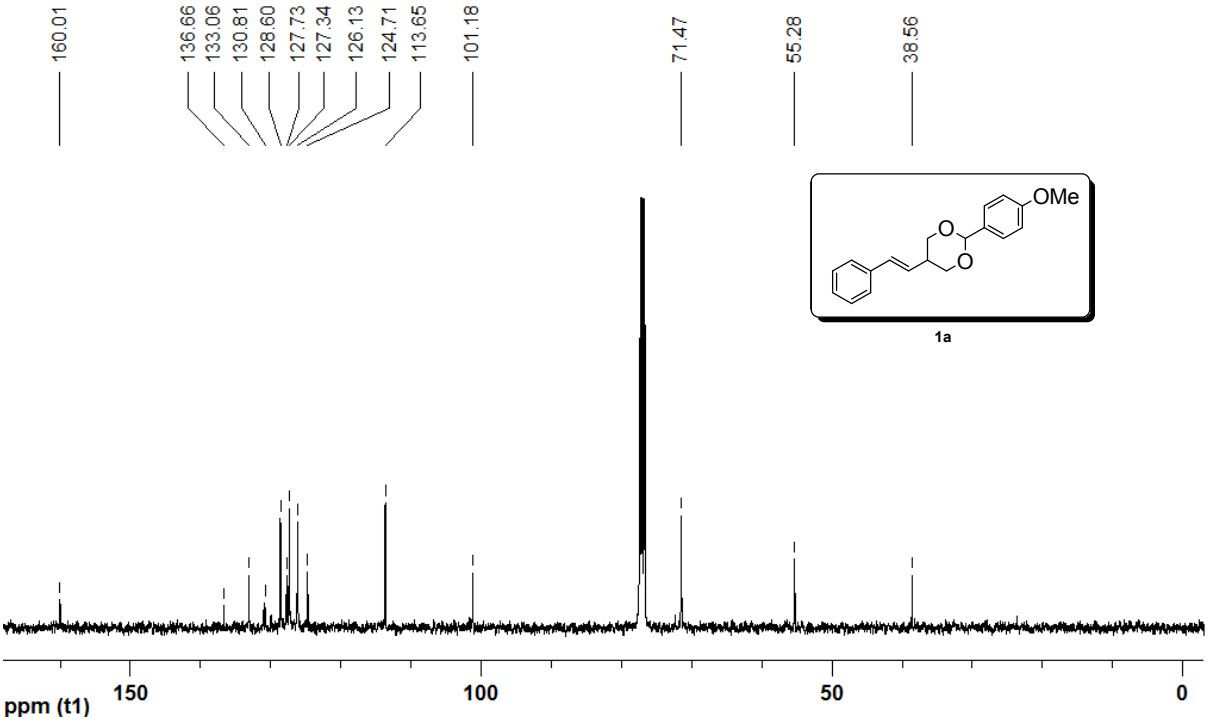
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 3j



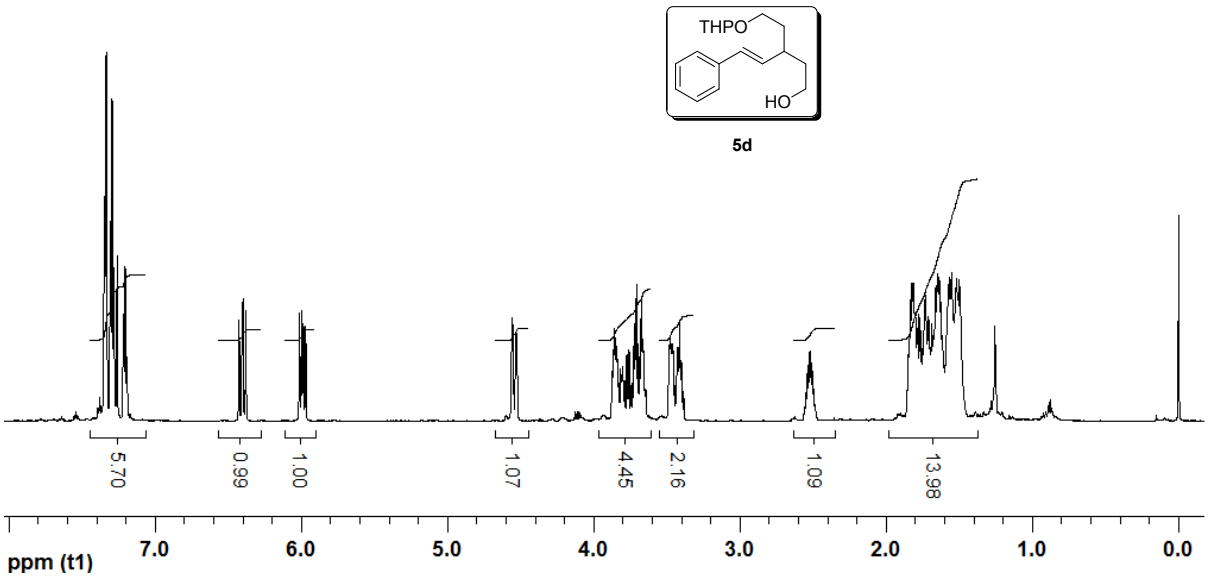
^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 3j



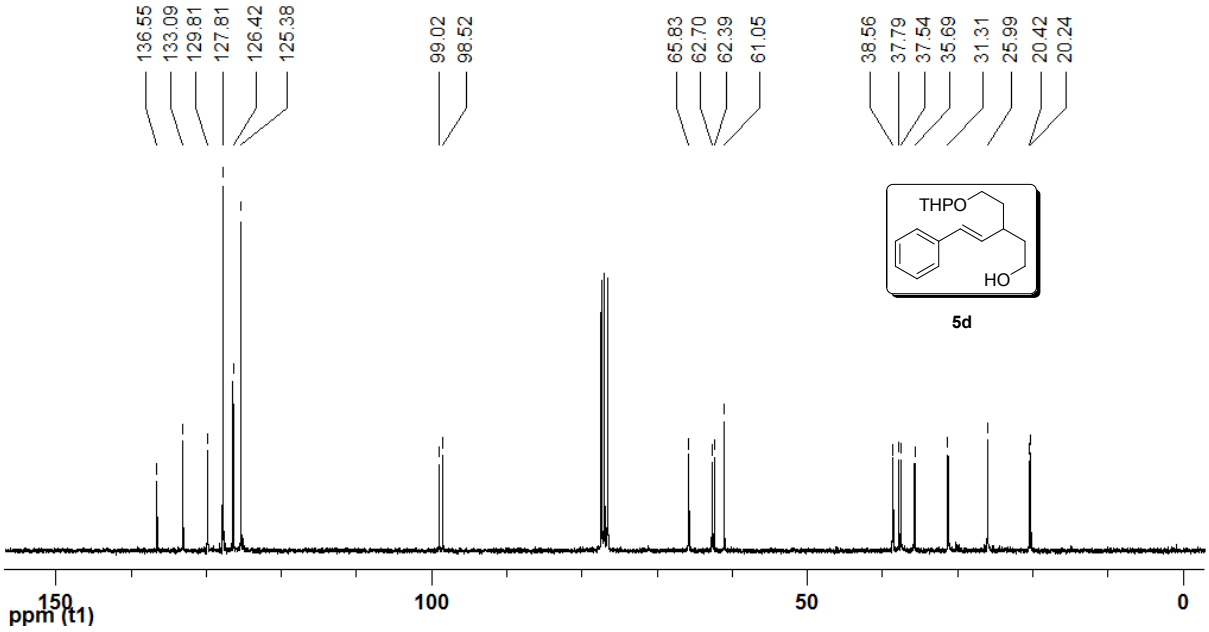
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 1a



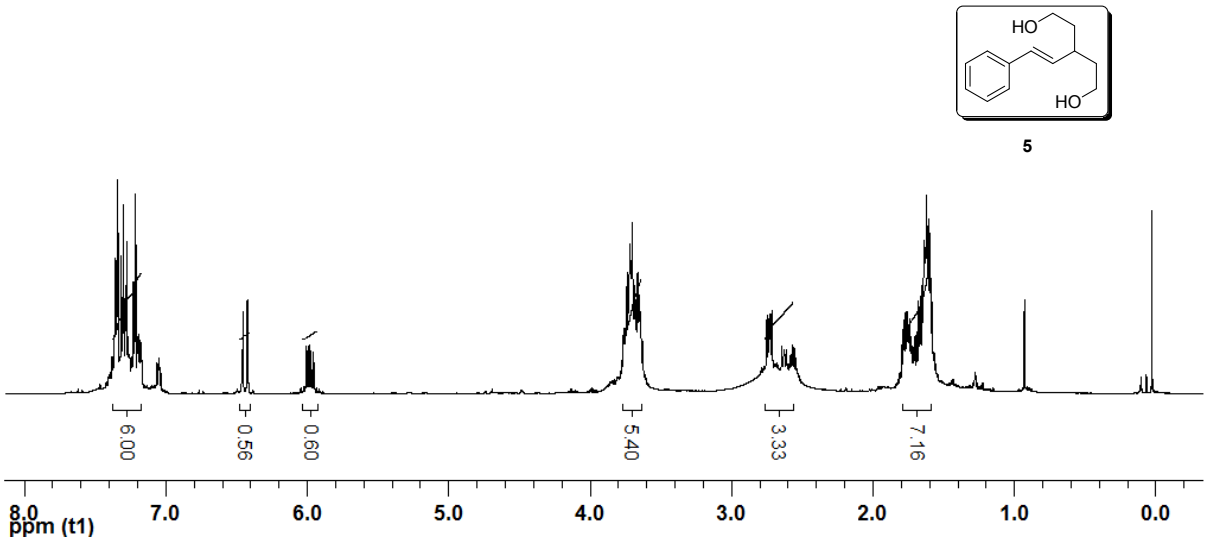
^{13}C NMR (100 MHz, $CDCl_3$) spectrum of compound 1a



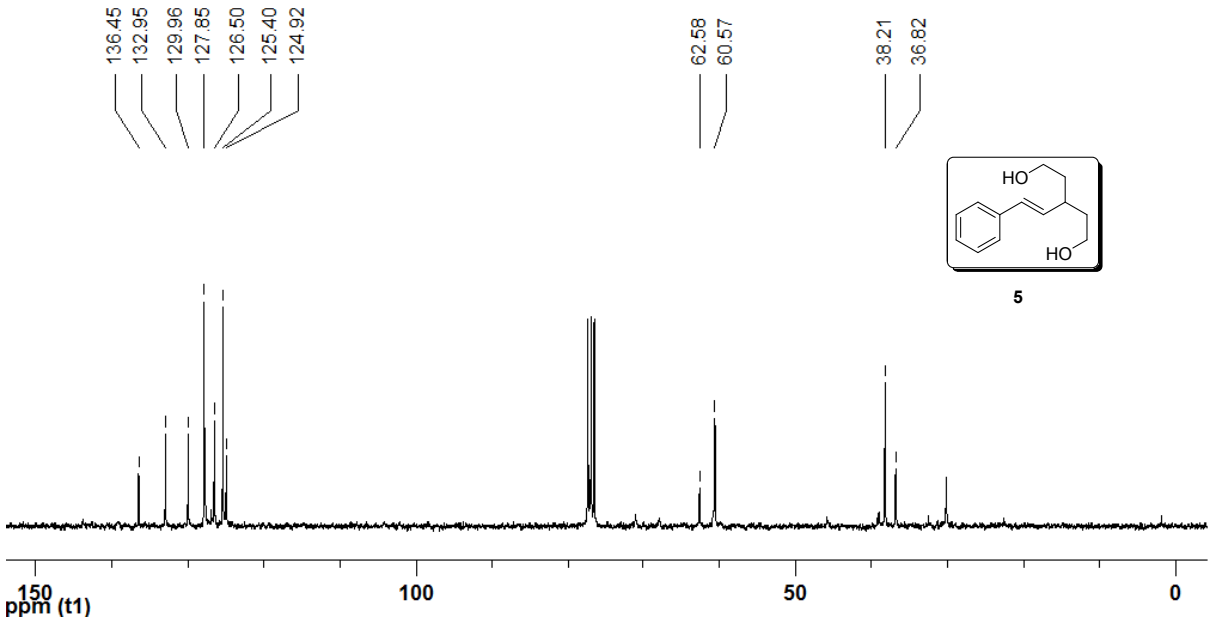
^{1}H NMR (600 MHz, $CDCl_3$) spectrum of compound 5d



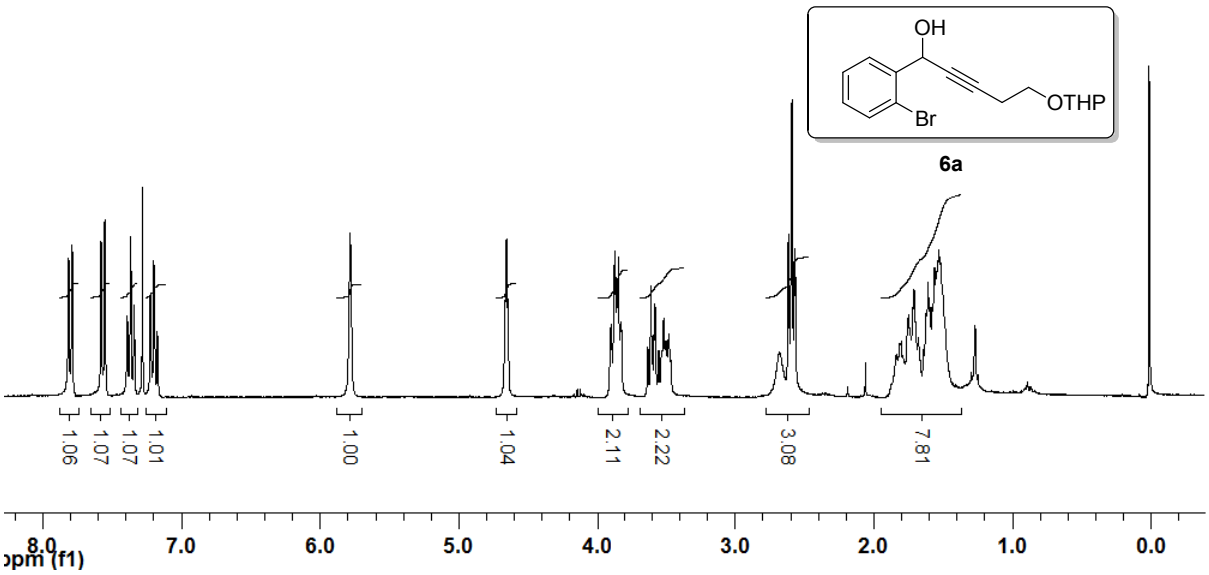
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 5d



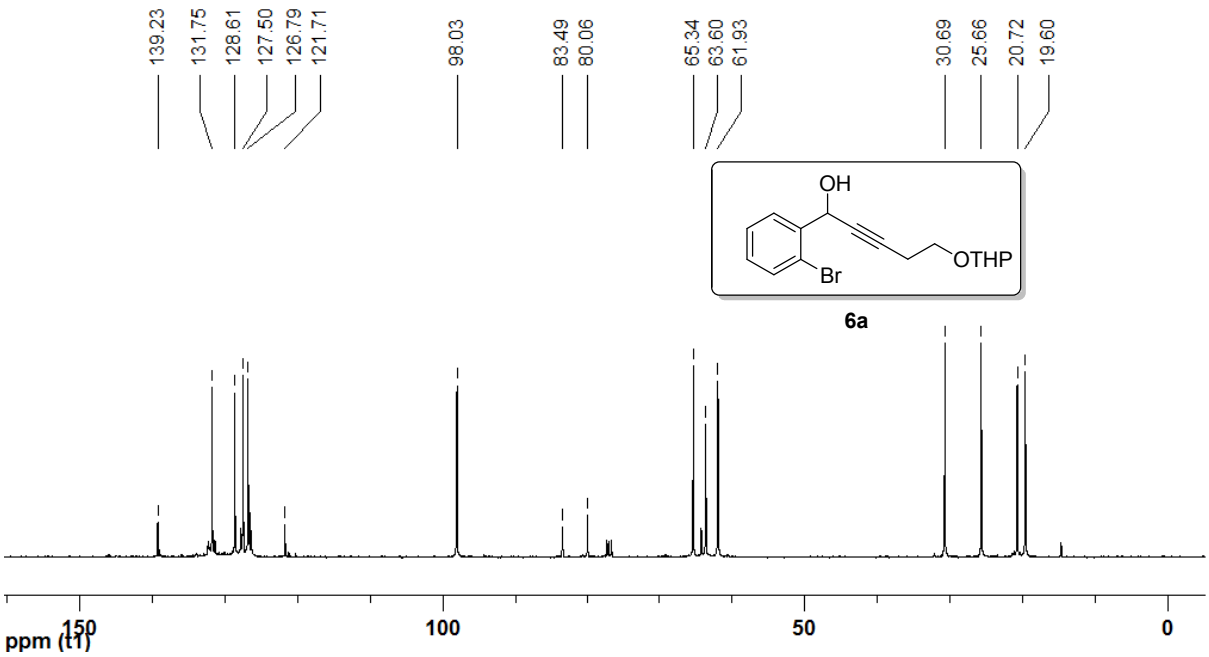
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 5



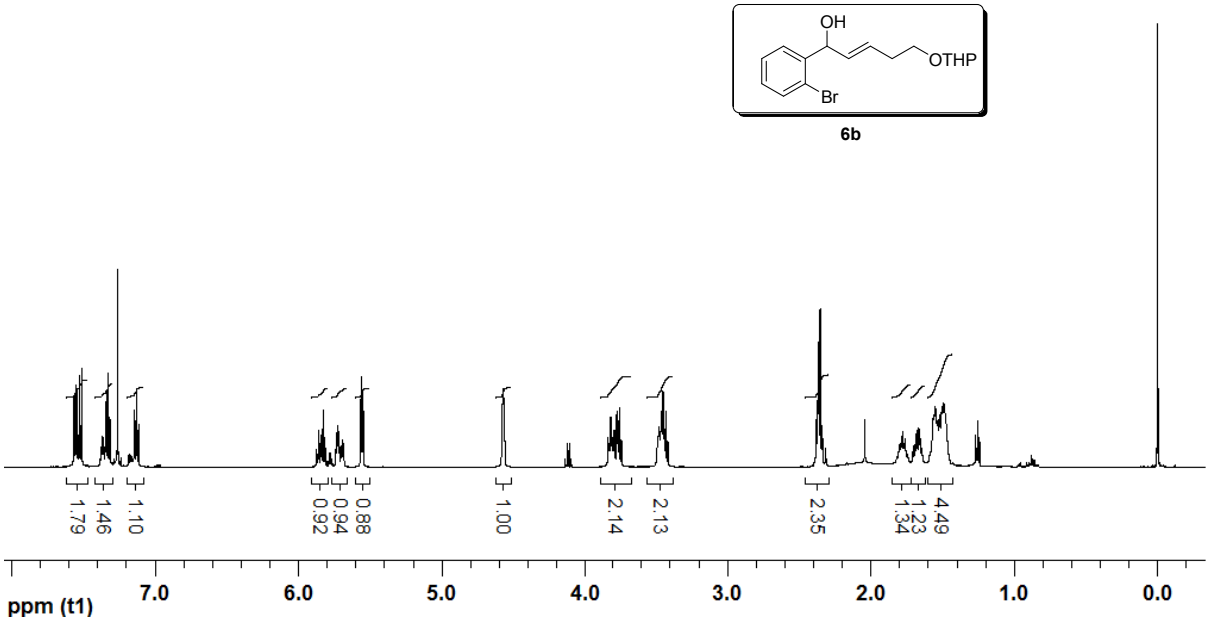
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 5



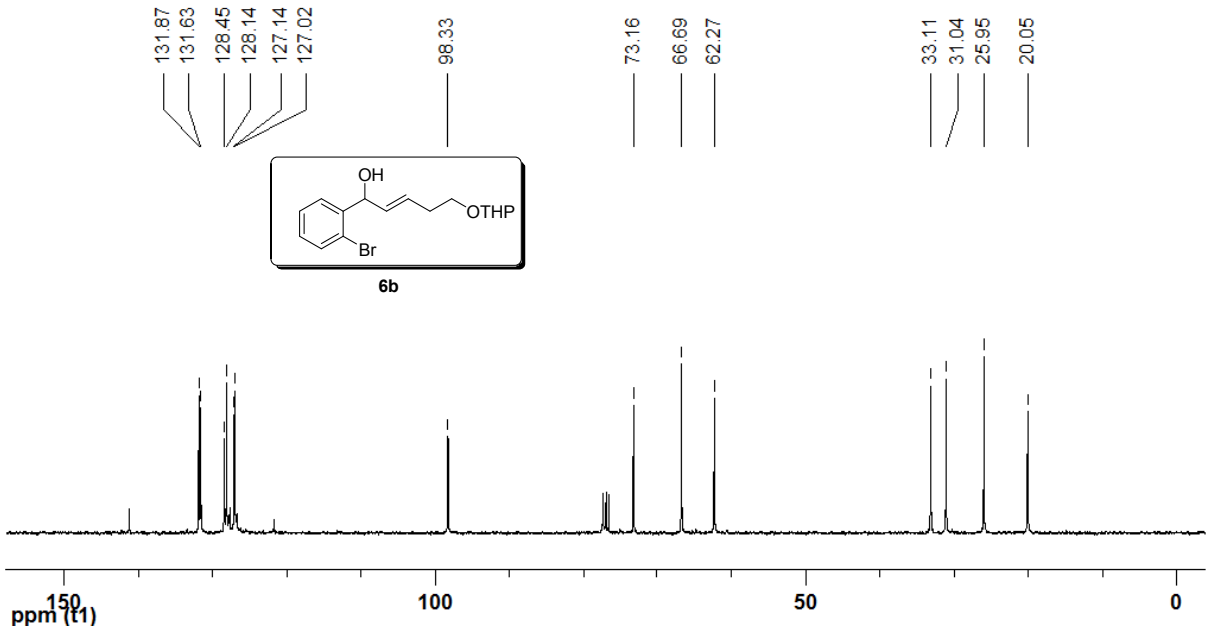
^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 6a



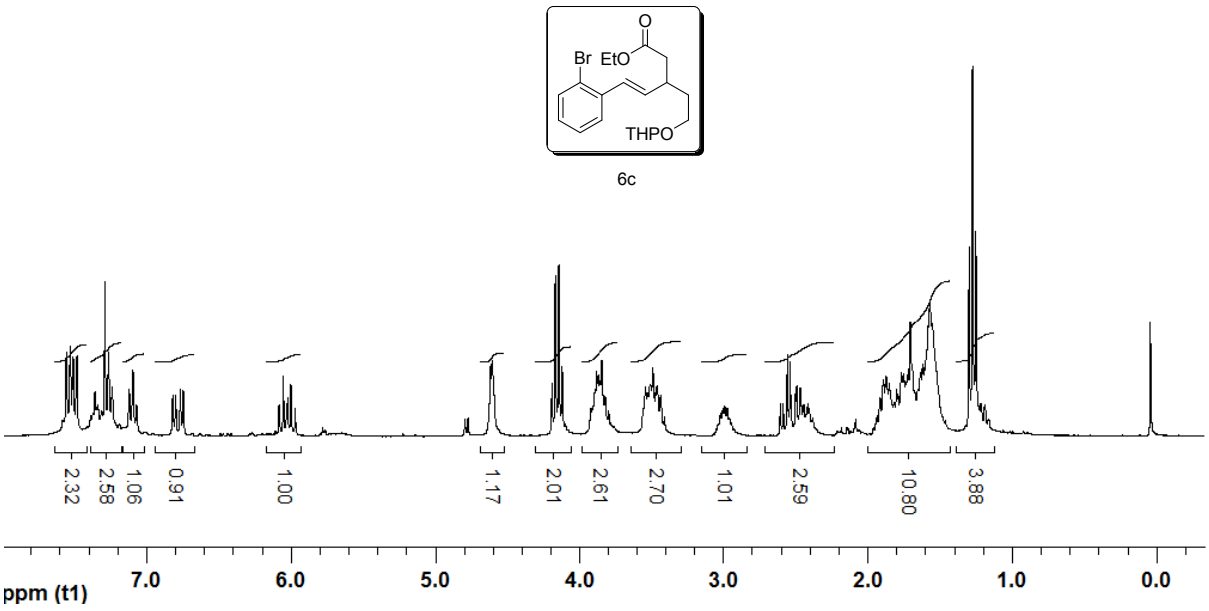
^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 6a



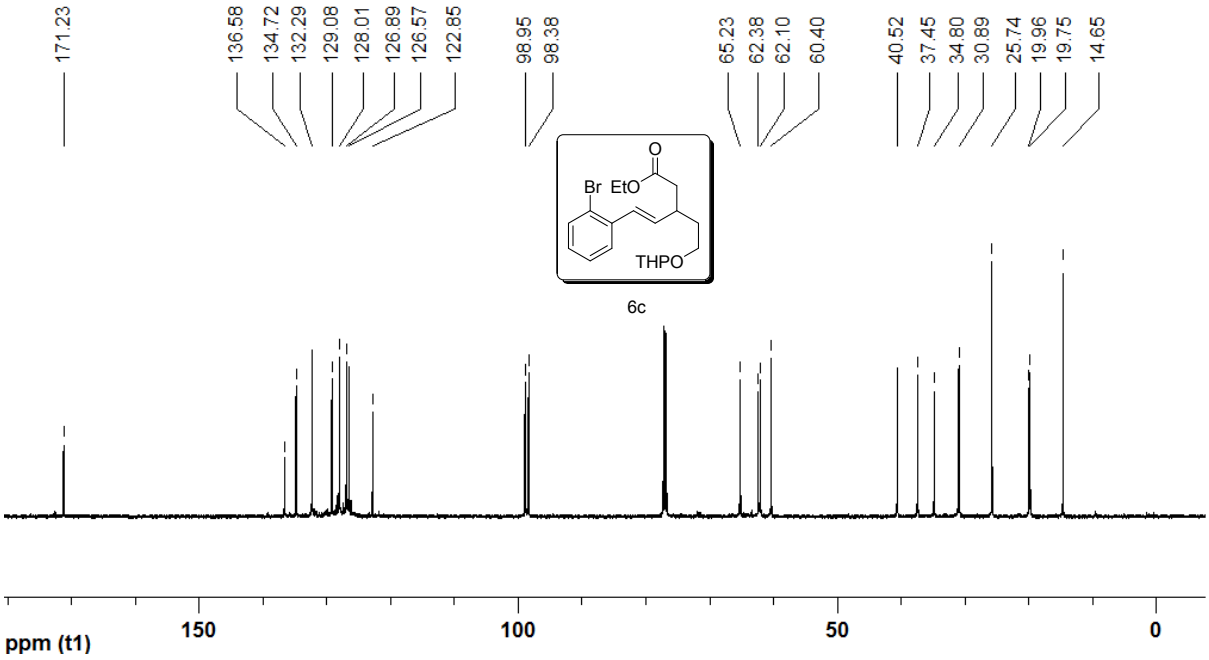
^{1}H NMR (500 MHz, $CDCl_3$) spectrum of compound 6b



^{13}C NMR (75 MHz, $CDCl_3$) spectrum of compound 6b



^{1}H NMR (300 MHz, $CDCl_3$) spectrum of compound 6c



^{13}C NMR (125 MHz, $CDCl_3$) spectrum of compound 6c