

Supplementary Materials for

Laccase-catalyzed 1,4-dioxane-mediated synthesis of belladine N-oxides with anti-influenza A virus activity.

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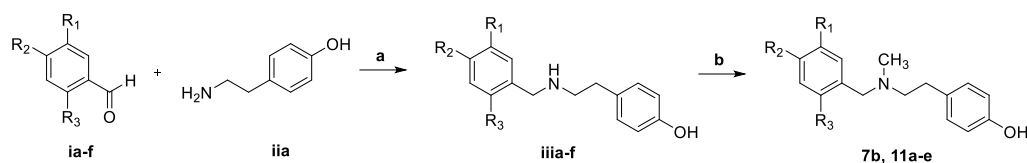
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SM #1. General procedures

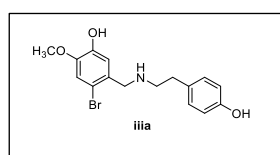
Reagents and solvents were obtained from commercial suppliers (Sigma-Aldrich Srl, Milan, Italy). Reactions were monitored using thin layer chromatography on precoated aluminium silica gel Merck 60 F254 plates and an UV lamp ($\lambda_{\text{max}} = 254 \text{ nm}$) was used for visualization. Merck silica gel 60 (230-400 mesh) was used for flash chromatography with the indicated solvent system. All products were dried in high vacuum (10-3 mbar). ¹H NMR and ¹³C-NMR and, DEPT-135 NMR were recorded on a Bruker Avance DRX400 (400 MHz/100 MHz) spectrometer. Chemical shifts for protons are reported in parts per million (δ scale) and internally referenced to the CD₃OD and DMSO-*d*₆ signal at δ 3.33 ppm and 2.50 ppm respectively. Coupling constants (*J*) are reported in Hz. Multiplicities are reported in the conventional form: s = singlet, d = doublet, t = triplet, td= triplet of doublets, q = quartet, ABq = AB quartet, m = multiplet, br = broad.

SM #2. Synthesis of compounds 7b and 11a-h

SM #2.1. Synthesis of compounds 7b and 11a-e

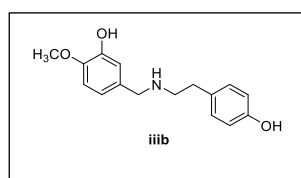


a: NaBH₄, MeOH, r.t. (90-96%); b: H₂CO, NaBH₄, MeOH, r.t. (93-98%).

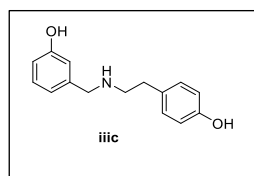


4-bromo-5-(((4-hydroxyphenethyl)amino)methyl)-2-methoxyphenol **iiia**:

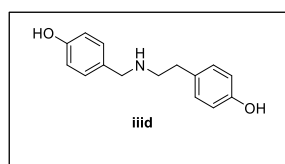
2-bromo-5-hydroxy-4-methoxybenzaldehyde **ia** (1.0 eq., 1.0 mmol) and tyramine **iia** (1.1 eq., 1.1 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 7:1:0.1) to afford **iiia** as a white powder (96%). ¹H-NMR (400 MHz, MeOD): 7.05 (s, 1H, ArH), 7.00 (d, 2H, ArH, J = 8.4 Hz), 6.84 (s, 1H, ArH), 6.71-6.69 (m, 2H, ArH), 3.83 (s, 3H, -OCH₃), 3.71 (s, 2H, ArCH₂N-), 2.80-2.76 (m, 2H, -NCH₂CH₂Ar), 2.73-2.70 (m, 2H, -NCH₂CH₂Ar) ppm. ¹³C-NMR (100 MHz, MeOD): 155.5, 147.8, 146.0, 130.2, 129.9, 129.2, 117.1, 115.4, 114.9, 111.7, 55.3, 52.2, 49.7, 34.2 ppm.



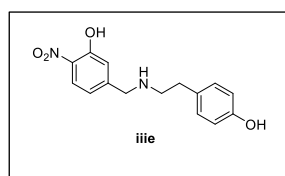
5-(((4-hydroxyphenethyl)amino)methyl)-2-methoxyphenol **iiib**: 3-hydroxy-4-methoxybenzaldehyde **ib** (1.0 eq., 1.0 mmol) and tyramine **iia** (1.1 eq., 1.1 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. Compound **iiib** was purified by crystallization in methanol to afford a colorless crystal (90%). ¹H-NMR (400 MHz, DMSO-d₆): 6.97 (d, 2H, ArH, J = 8.0 Hz), 6.81 (d, 1H, ArH, J = 8.0 Hz), 6.73 (s, 1H, ArH), 6.65 (m, 3H, ArH), 3.72 (s, 3H, -OCH₃), 3.54 (s, 2H, ArCH₂N-), 2.60-2.58 (brm, 4H, -NCH₂CH₂Ar), ppm. ¹³C-NMR (100 MHz, DMSO-d₆): 155.8, 146.7, 146.6, 134.0, 130.9, 129.8, 118.9, 115.8, 115.4, 112.4, 56.1, 53.0, 51.1, 35.4 ppm.



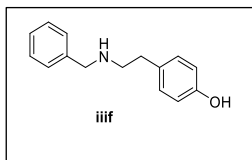
3-(((4-hydroxyphenethyl)amino)methyl)phenol **iiic**: 3-hydroxybenzaldehyde **ic** (1.0 eq., 1.0 mmol) and tyramine **iia** (1.1 eq., 1.1 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. Compound **iiic** was purified by crystallization in methanol to afford a colorless crystal (91%). ¹H-NMR (400 MHz, DMSO-d₆): 7.06 (t, 1H, ArH, J = 7.6), 6.98 (d, 2H, ArH, J = 8.0 Hz), 6.72-6.58 (m, 5H, ArH), 3.60 (s, 2H, ArCH₂N-), 2.62-2.61 (brd, 4H, -NCH₂CH₂Ar), ppm. ¹³C-NMR (100 MHz, DMSO-d₆): 157.7, 155.8, 142.9, 130.9, 129.8, 129.4, 118.9, 115.4, 115.1, 113.8, 53.3, 51.3, 35.4 ppm.



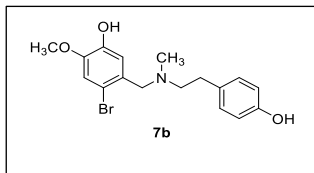
4-(2-(((4-hydroxybenzyl)amino)ethyl)phenol **iiid**: 4-hydroxybenzaldehyde **id** (1.0 eq., 1.0 mmol) and tyramine **iia** (1.1 eq., 1.1 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 7:1:0.1) to afford **iiid** as a white powder (94%). ¹H-NMR (400 MHz, MeOD): 7.13-7.11 (dd, 2H, ArH, J = 6.4, 2.0 Hz), 7.02-7.00 (dd, 2H, ArH, J = 6.4, 2.0 Hz), 6.75-6.70 (m, 4H, ArH), 3.67 (s, 2H, ArCH₂N-), 2.81-2.70 (m, 4H, -NCH₂CH₂Ar), ppm. ¹³C-NMR (100 MHz, MeOD): 156.5, 155.5, 130.0, 129.4, 129.3, 129.1, 114.9, 114.8, 52.3, 49.9, 34.0 ppm.



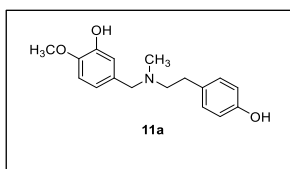
5-(((4-hydroxyphenethyl)amino)methyl)-2-nitrophenol **iiie**: 3-hydroxy-4-nitrobenzaldehyde **ie** (1.0 eq., 1.0 mmol) and tyramine **iia** (1.1 eq., 1.1 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. Compound **iiie** was purified by crystallization in methanol to afford an orange crystal (91%). ¹H-NMR (400 MHz, DMSO-d₆): 7.80 (d, 1H, ArH, J = 8.4 Hz), 7.00-6.98 (m, 3H, ArH), 6.78 (d, 2H, ArH, J = 8.4 Hz), 6.66 (d, 1H, ArH, J = 8.4 Hz), 3.70 (s, 2H, ArCH₂N-), 2.69-2.60 (m, 4H, -NCH₂CH₂Ar), ppm. ¹³C-NMR (100 MHz, DMSO-d₆): 155.9, 155.1, 149.0, 135.5, 130.4, 129.8, 125.7, 119.6, 117.7, 115.5, 52.3, 51.0, 35.0 ppm.



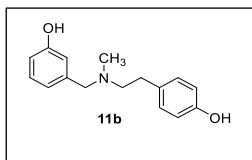
4-(2-(benzylamino)ethyl)phenol **iiif**: benzaldehyde **if** (1.0 eq., 1.0 mmol) and tyramine **ia** (1.1 eq., 1.1 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 15:1:0.1) to afford **iiif** as a white powder (96%). ¹H-NMR (400 MHz, DMSO-*d*₆): 7.28-7.17 (m, 5H, ArH), 6.97 (d, 2H, ArH, *J* = 8.4 Hz), 6.65 (d, 2H, ArH, *J* = 8.4 Hz), 3.68 (s, 2H, ArCH₂N-), 2.64-2.59 (brm, 4H, -NCH₂CH₂Ar), ppm. ¹³C-NMR (100 MHz, DMSO-*d*₆): 155.8, 141.4, 130.9, 129.8, 128.5, 128.3, 126.9, 115.4, 53.3, 51.2, 35.4 ppm.



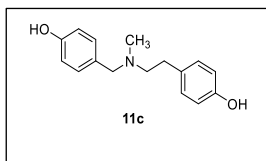
4-bromo-5-(((4-hydroxyphenethyl)(methyl)amino)methyl)-2-methoxyphenol **7b**: compound **iiia** (1.0 eq., 1.0 mmol) and formaldehyde (1.05 eq., 1.05 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 14:1:0.1) to afford **7b** as a white powder (98%). ¹H-NMR (400 MHz, MeOD): 7.07 (s, 1H, ArH), 7.01 (d, 2H, ArH, *J* = 8.8 Hz), 6.93 (s, 1H, ArH), 6.71-6.67 (m, 2H, ArH), 3.83 (s, 3H, -OCH₃), 3.58 (s, 2H, ArCH₂N-), 2.76-2.72 (m, 2H, -NCH₂CH₂Ar), 2.65-2.60 (m, 2H, -NCH₂CH₂Ar), 2.29 (s, 3H, -NCH₃) ppm. ¹³C-NMR (100 MHz, MeOD): 155.2, 147.7, 145.8, 130.8, 129.5, 129.1, 117.5, 115.3, 114.8, 112.8, 60.0, 59.5, 55.2, 40.9, 32.1 ppm.



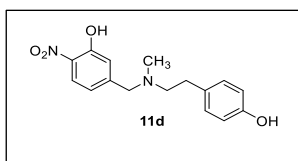
5-(((4-hydroxyphenethyl)(methyl)amino)methyl)-2-methoxyphenol **11a**: compound **iiib** (1.0 eq., 1.0 mmol) and formaldehyde (1.05 eq., 1.05 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 16:1:0.1) to afford **11a** as a white powder (97%). ¹H-NMR (400 MHz, MeOD): 7.01-6.98 (m, 2H, ArH), 6.90 (d, 1H, ArH, *J* = 8.4 Hz), 6.82 (s, 1H, ArH), 6.78-6.75 (dd, 1H, ArH, *J* = 8.0, 2 Hz), 6.72-6.68 (m, 2H, ArH), 3.86 (s, 3H, -OCH₃), 3.50 (s, 2H, ArCH₂N-), 2.76-2.72 (m, 2H, -NCH₂CH₂Ar), 2.60-2.56 (m, 2H, -NCH₂CH₂Ar), 2.29 (s, 3H, -NCH₃) ppm. ¹³C-NMR (100 MHz, MeOD): 155.3, 147.1, 146.0, 130.5, 129.9, 129.1, 120.8, 116.4, 114.8, 111.0, 60.9, 58.8, 55.0, 40.7, 31.8 ppm.



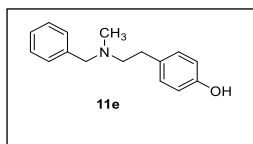
3-(((4-hydroxyphenethyl)(methyl)amino)methyl)phenol **11b**: compound **iiic** (1.0 eq., 1.0 mmol) and formaldehyde (1.05 eq., 1.05 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 14:1:0.1) to afford **11b** as a white powder (94%). ¹H-NMR (400 MHz, MeOD): 7.15 (t, 1H, ArH, *J* = 6.4, 1.6 Hz), 7.01-6.98 (m, 2H, ArH), 6.81-6.79 (m, 2H, ArH), 6.73-6.68 (m, 3H, ArH), 3.53 (s, 2H, ArCH₂N-), 2.76-2.72 (m, 2H, -NCH₂CH₂Ar), 2.61-2.57 (m, 2H, -NCH₂CH₂Ar), 2.29 (s, 3H, -NCH₃) ppm. ¹³C-NMR (100 MHz, MeOD): 157.1, 155.2, 138.8, 130.6, 129.2, 128.9, 120.6, 116.2, 114.9, 114.0, 61.4, 59.0, 41.0, 31.9 ppm.



4-(2-(((4-hydroxybenzyl)(methyl)amino)ethyl)phenol **11c**: compound **iiid** (1.0 eq., 1.0 mmol) was dissolved in methanol (2.0 mL) and formaldehyde (1.05 eq., 1.05 mmol) was added to the solution. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 14:1:0.1) to afford **11c** as a white powder. ¹H-NMR (400 MHz, MeOD): 7.15 (d, 2H, ArH, *J* = 8.4 Hz), 6.99 (d, 2H, ArH, *J* = 8.4 Hz), 6.77-6.74 (m, 2H, ArH), 6.71-6.67 (m, 2H, ArH), 3.52 (s, 2H, ArCH₂N-), 2.75-2.71 (m, 2H, -NCH₂CH₂Ar), 2.61-2.57 (m, 2H, -NCH₂CH₂Ar), 2.28 (s, 3H, -NCH₃) ppm. ¹³C-NMR (100 MHz, MeOD): 156.6, 155.3, 130.7, 130.4, 129.1, 127.5, 114.8, 114.6, 60.7, 58.7, 40.6, 31.7 ppm.

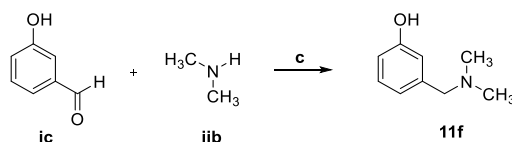


5-(((4-hydroxyphenethyl)(methyl)amino)methyl)-2-nitrophenol **11d**: compound **iiie** (1.0 eq., 1.0 mmol) and formaldehyde (1.05 eq., 1.05 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 12:1:0.1) to afford **11d** as a yellow powder (93%). ¹H-NMR (400 MHz, MeOD): 8.02 (d, 1H, ArH, *J* = 8.8 Hz), 7.10 (d, 1H, ArH, *J* = 1.6 Hz), 7.01-6.98 (d, 2H, ArH, *J* = 8.4 Hz), 6.96-6.93 (dd, 1H, ArH, *J* = 8.4, 1.6 Hz), 6.71-6.76 (m, 2H, ArH), 3.61 (s, 2H, ArCH₂N-), 2.76-2.72 (m, 2H, -NCH₂CH₂Ar), 2.64-2.60 (m, 2H, -NCH₂CH₂Ar), 2.31 (s, 3H, -NCH₃) ppm. ¹³C-NMR (100 MHz, MeOD): 155.3, 154.5, 149.0, 133.3, 130.6, 129.2, 124.7, 120.0, 119.6, 114.7, 60.7, 59.2, 41.1, 32.2 ppm.

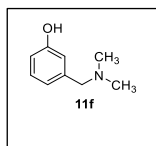


4-(2-(benzyl(methyl)amino)ethyl)phenol **11e**: compound **iiif** (1.0 eq., 1 mmol) and formaldehyde (1.05 eq., 1.05 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 20:1:0.1) to afford **11e** as a white powder (98%). ¹H-NMR (400 MHz, MeOD): 7.34-7.25 (m, 5H, ArH), 6.99-6.96 (m, 2H, ArH), 6.70-6.67 (m, 2H, ArH), 3.58 (s, 2H, ArCH₂N-), 2.75-2.71 (m, 2H, -NCH₂CH₂Ar), 2.59-2.57 (m, 2H, -NCH₂CH₂Ar), 2.27 (s, 3H, -NCH₃) ppm. ¹³C-NMR (100 MHz, MeOD): 155.3, 137.5, 130.7, 129.2, 129.1, 127.9, 127.0, 114.8, 61.5, 59.0, 40.9, 31.9 ppm.

SM #2.2. Synthesis and characterization of compound **11f**

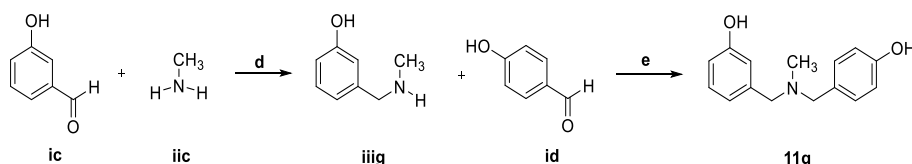


c: Ti(Oi-Pr)₄, NaBH₄, MeOH dry, r.t., argon (91%)

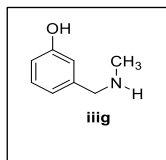


3-((dimethylamino)methyl)phenol **11f**: 3-hydroxybenzaldehyde **ic** (1.0 eq., 1.0 mmol) was dissolved in dry methanol (2.0 mL). Ti(Oi-Pr)₄ (1.3 eq., 1.3 mmol) and dimethylamine **iib** (1.1 eq., 1.1 mmol, 2M solution in methanol) was added and the mixture was stirred for 4h at room temperature under inert atmosphere. After this period, NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature, filtered over Celite® and evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH 10:1) to afford **11f** as a colorless oil (91%). ¹H-NMR (400 MHz, MeOD): 7.14 (t, 1H, ArH, J= 8.4, 8.0 Hz), 6.80-6.76 (m, 2H, ArH), 6.71-6.68 (dd, 1H, ArH, J= 8.0, 2.0 Hz), 3.63 (s, 2H, ArCH₂N-), 2.37 (s, 6H, -N(CH₃)₂) ppm. ¹³C-NMR (100 MHz, MeOD): 152.8, 135.6, 124.4, 114.5, 110.4, 109.2, 50.2, 29.3 ppm.

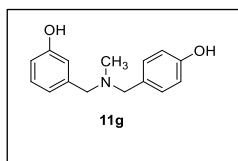
SM #2.3. synthesis and characterization of compound **11g**



d: NaBH₄, MeOH, r.t. (96%); e: Ti(Oi-Pr)₄, NaBH₄, MeOH dry, r.t., argon (82%)

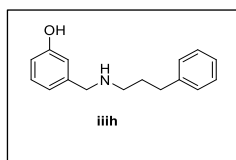
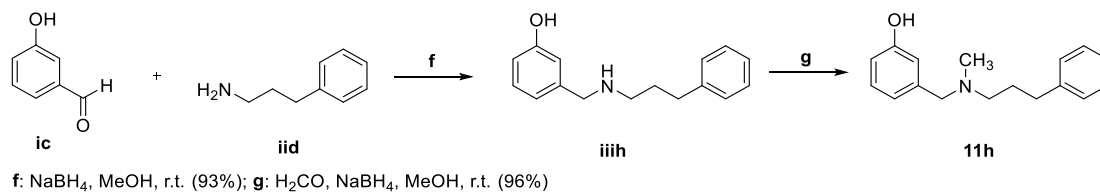


3-((methylamino)methyl)phenol **iiig**: 3-hydroxybenzaldehyde **ic** (1.0 eq., 1 mmol) and methylamine (1.1 eq., 1.1 mmol, 2M solution in methanol) were dissolved in methanol and the solution was stirred over night at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH, 7:1:0.1) to afford **iiig** as colorless oil (96%). ¹H-NMR (400 MHz, DMSO-d₆): 7.06 (t, 1H, ArH, J= 7.6 Hz), 6.71-6.69 (m, 2H, ArH), 6.60-6.58 (dd, 1H, ArH, J= 7.6, 1.2 Hz), 3.53 (s, 2H, ArCH₂N-), 2.23 (s, 3H, -NCH₃), ppm. ¹³C-NMR (100 MHz, DMSO-d₆): 157.1, 141.9, 128.8, 118.3, 114.6, 113.2, 54.8, 35.3 ppm

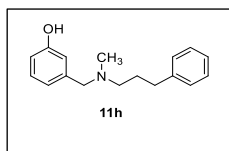


3-(((4-hydroxybenzyl)(methyl)amino)methyl)phenol **11g**: 4-hydroxybenzaldehyde **id** (1.0 eq., 1 mmol) was dissolved in dry methanol (2.0 mL). Then Ti(Oi-Pr)₄ (1.3 eq., 1.3 mmol) and **iiig** (1.1 eq., 1.1 mmol) were added and the mixture was stirred for 4h at room temperature under inert atmosphere. After this period, NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the solution was stirred for 5h at room temperature. The solution was then filtered over Celite® and the solvent was evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH 10:1) to afford **11g** as a white powder (82%). ¹H-NMR (400 MHz, MeOD): 7.17-7.12 (m, 3H, ArH), 6.81-6.75 (m, 4H, ArH), 6.72-6.70 (m, 1H, ArH), 3.45 (s, 4H, ArCH₂NCH₂Ar), 2.17 (s, 3H, -NCH₃), ppm. ¹³C-NMR (100 MHz, MeOD): 157.1, 156.5, 139.2, 130.4, 128.9, 128.2, 120.3, 115.9, 114.6, 113.9, 60.9, 60.6, 40.7 ppm

SM #2.4. synthesis and characterization of compound **11h**



3-((3-phenylpropyl)amino)methylphenol **iiih:** 3-hydroxybenzaldehyde **ic** (1.0 eq., 1 mmol) and 3-phenyl-1-propylamine **iid** were dissolved in methanol (2.0 mL) and the solution was stirred for 4h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the solution was filtered over Celite® and the solvent evaporated under reduced pressure. The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 20:1:0.1) to afford **iiih** as a white powder (93%). **¹H-NMR** (400 MHz, MeOD): 7.27-7.11 (m, 6H), 6.78-6.76 (m, 2H, ArH), 6.71-6.68 (m, 1H, ArH), 3.65 (s, 2H, ArCH₂N-), 2.65-2.58 (m, 4H, -NCH₂CH₂CH₂Ar), 1.87-1.80 (m, 2H, -NCH₂CH₂CH₂Ar) ppm. **¹³C-NMR** (100 MHz, MeOD): 157.3, 141.8, 140.5, 129.0, 128.0, 127.9, 125.4, 119.1, 115.0, 113.7, 52.9, 33.2, 30.7 ppm.

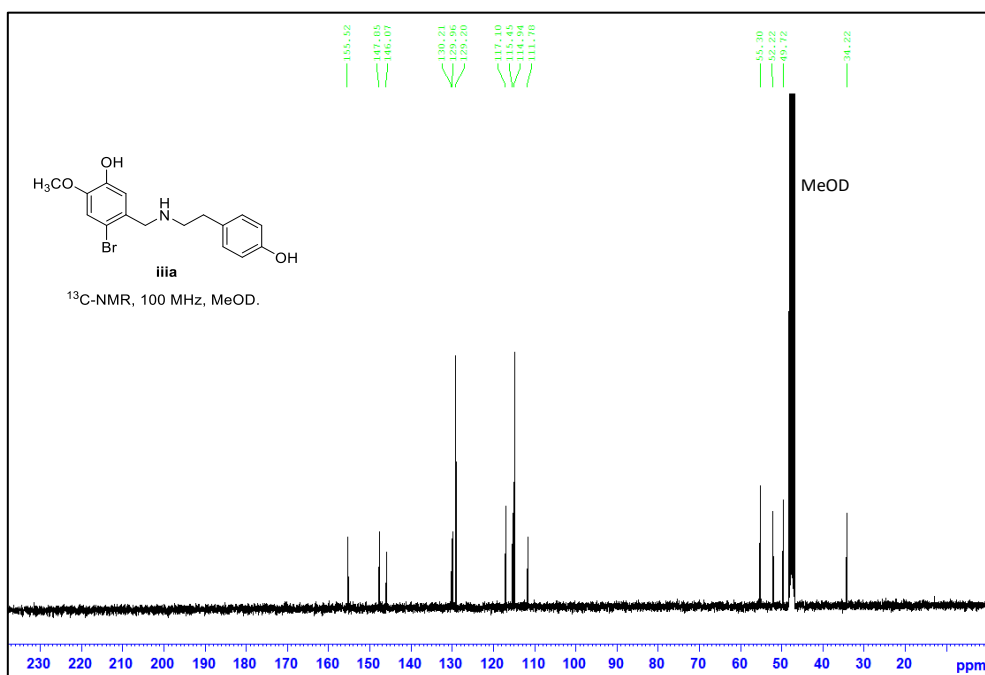
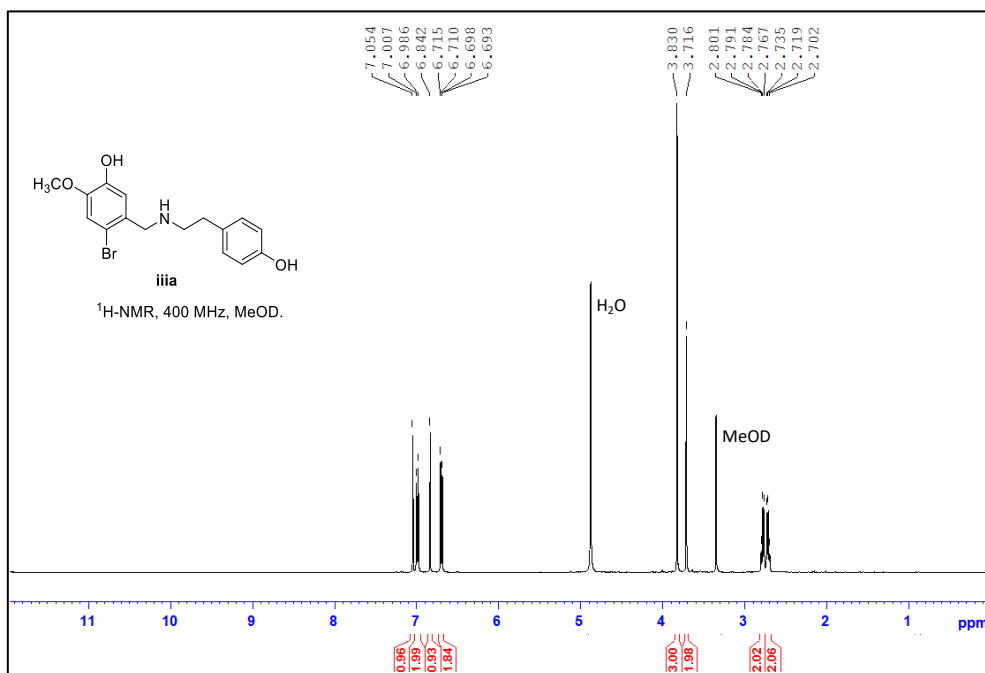


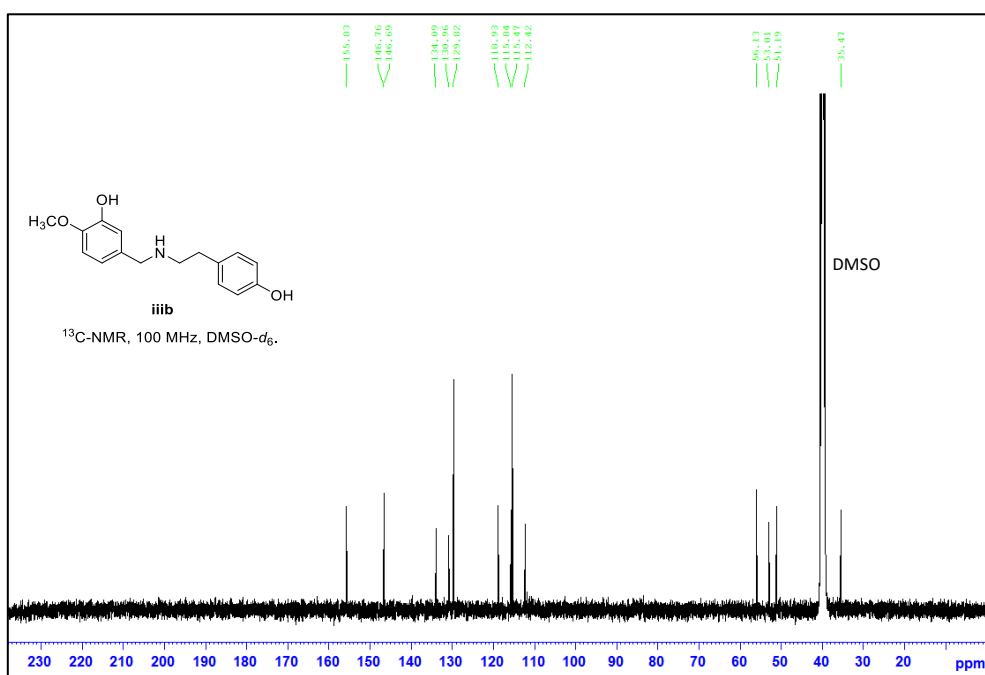
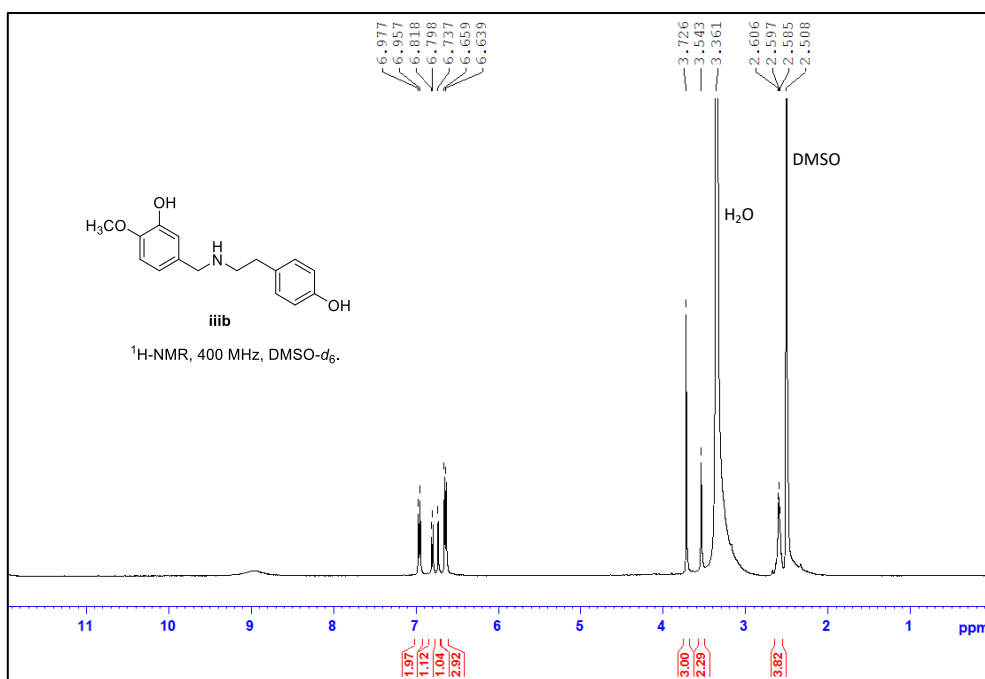
3-((methyl(3-phenylpropyl)amino)methyl)phenol **11h:** compound **iiih** (1.0 eq., 1 mmol) and formaldehyde (1.05 eq., 1.05 mmol) were dissolved in methanol (2.0 mL) and the solution was stirred for 5h at room temperature. Then NaBH₄ (1.1 eq., 1.1 mmol) was added at 0°C and the mixture was stirred for 5h at room temperature. After this period the mixture was filtered over Celite® and the solvent evaporated under reduced pressure.

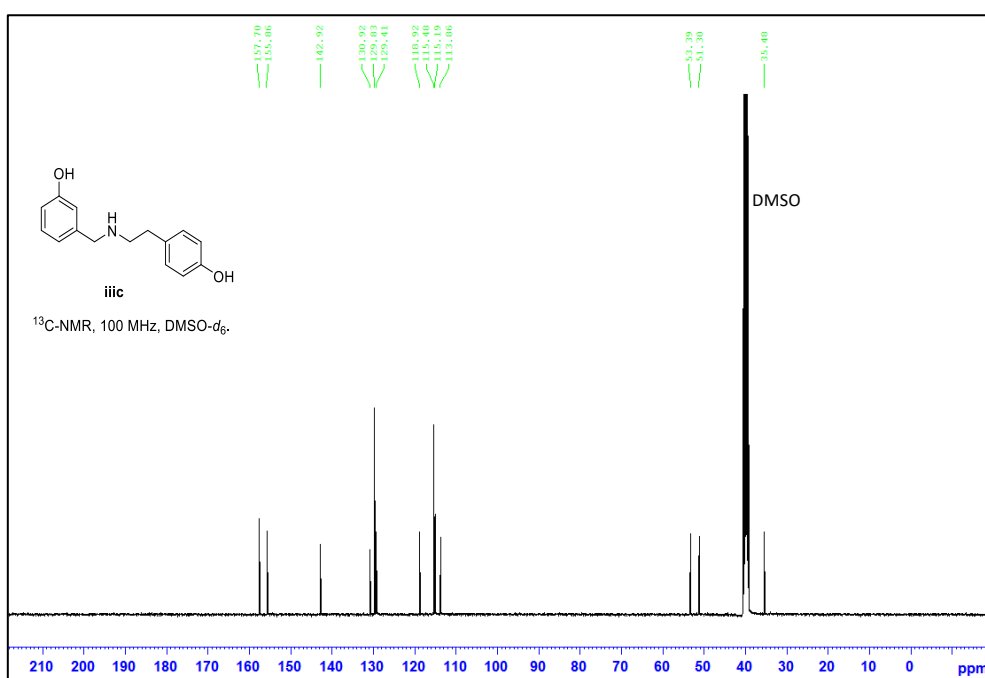
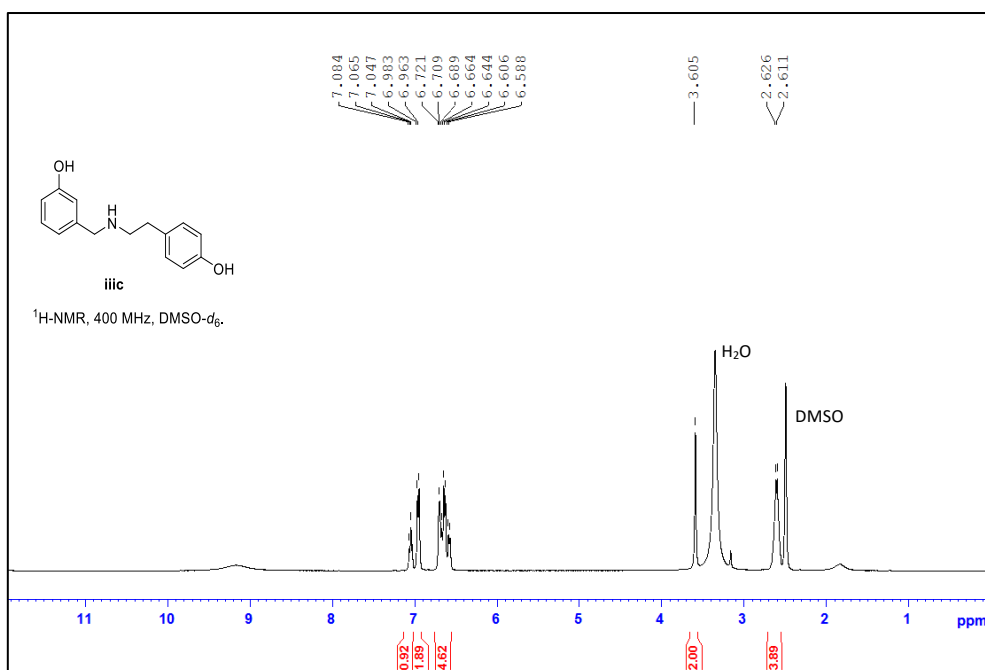
The crude mixture was purified by silica gel gradient column chromatography (DCM/MeOH/NH₄OH 9:1:0.1) to afford **11h** as a white powder (96%). **¹H-NMR** (400 MHz, MeOD): 7.27-7.23 (m, 2H, ArH), 7.19-7.11 (m, 4H, ArH), 6.78-6.76 (m, 2H, ArH), 6.72-6.69 (m, 1H, ArH), 3.47 (s, 2H, ArCH₂N-), 2.65-2.61 (t, 2H, -NCH₂CH₂CH₂Ar, J= 8.0 Hz), 2.46-2.42 (m, 2H, -NCH₂CH₂CH₂Ar), 2.22 (s, 3H, -NCH₃), 1.91-1.82 (m, 2H, -NCH₂CH₂CH₂Ar) ppm. **¹³C-NMR** (100 MHz, MeOD): 157.1, 141.8, 138.9, 128.8, 128.0, 127.9, 125.4, 120.4, 116.0, 113.9, 61.6, 56.3, 41.0, 33.1, 28.3 ppm.

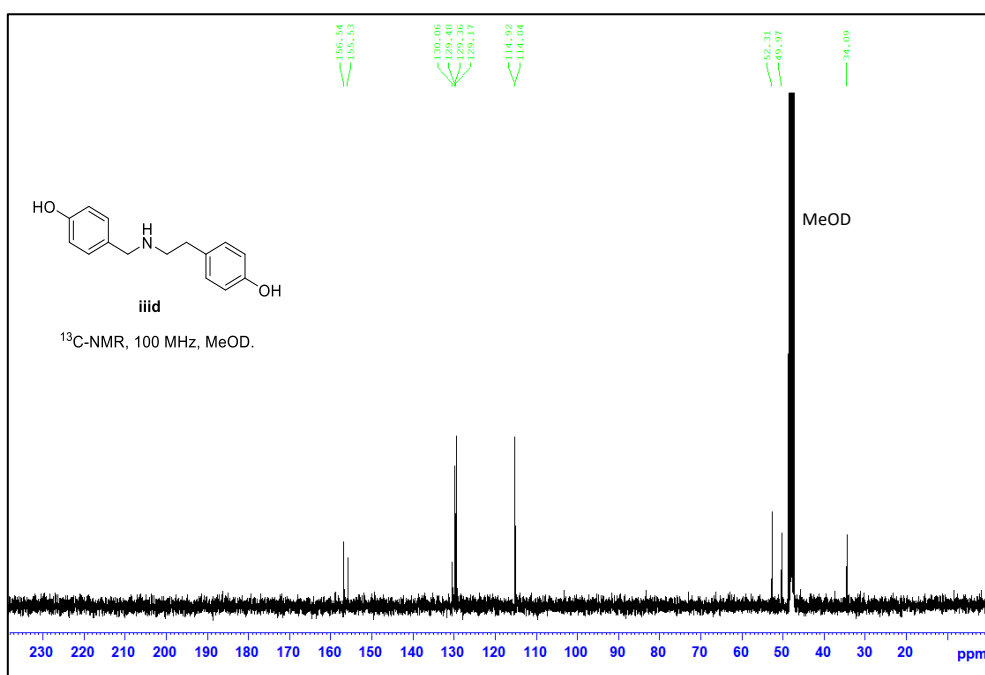
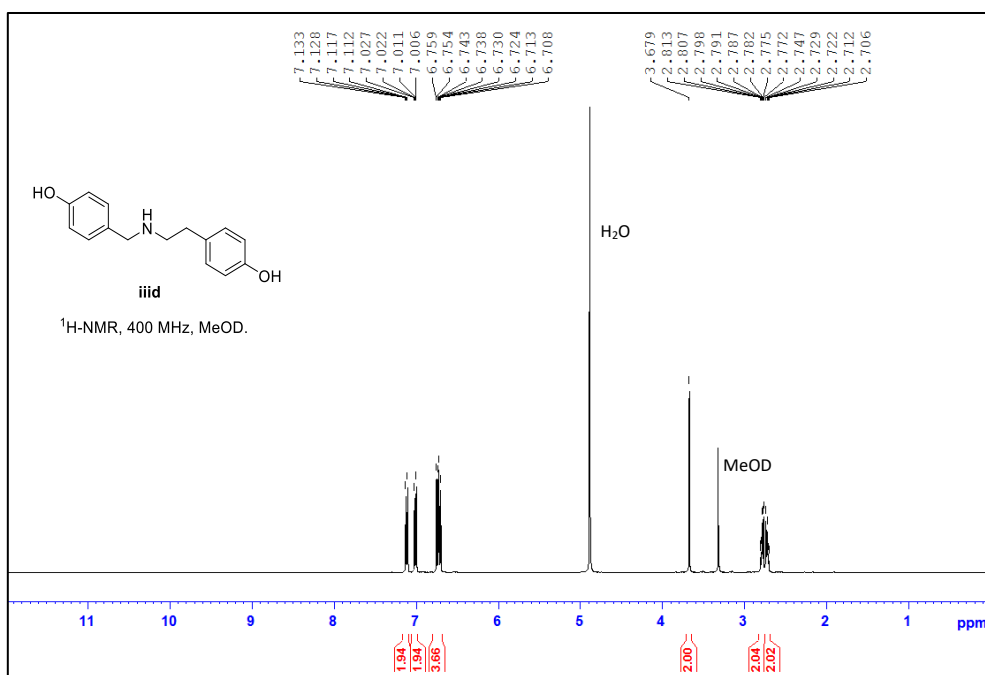
SM #3. NMR spectra.

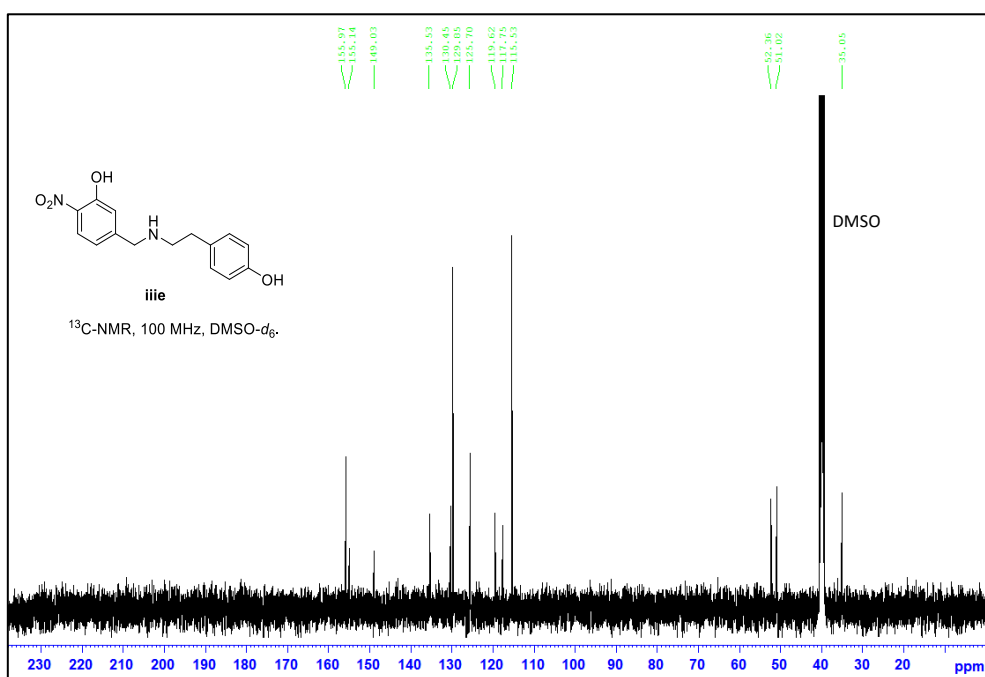
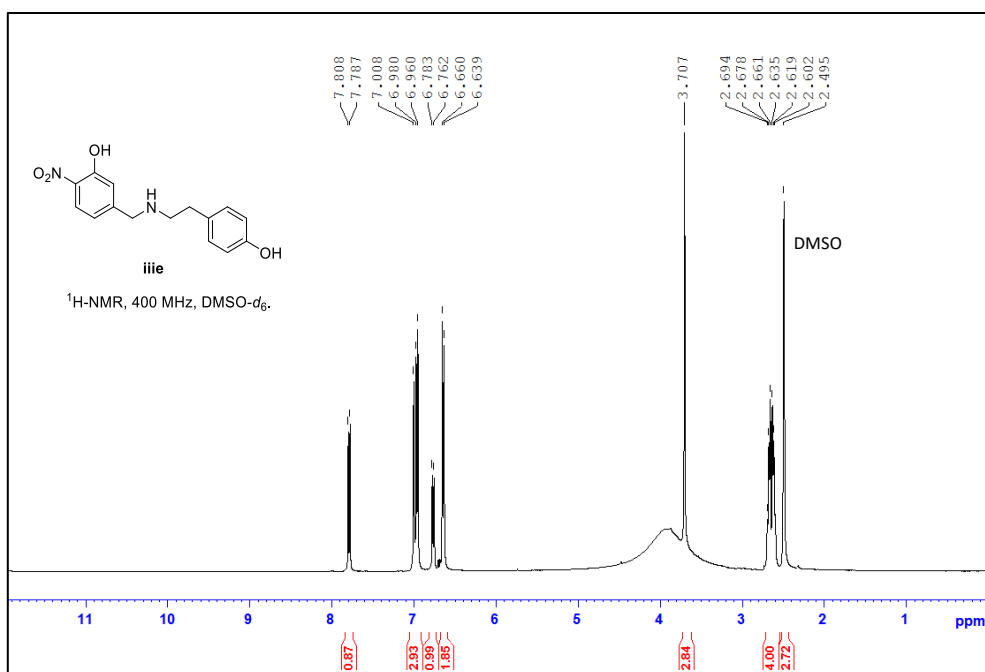
SM #3.1. NMR spectra of compounds *iii*a-h

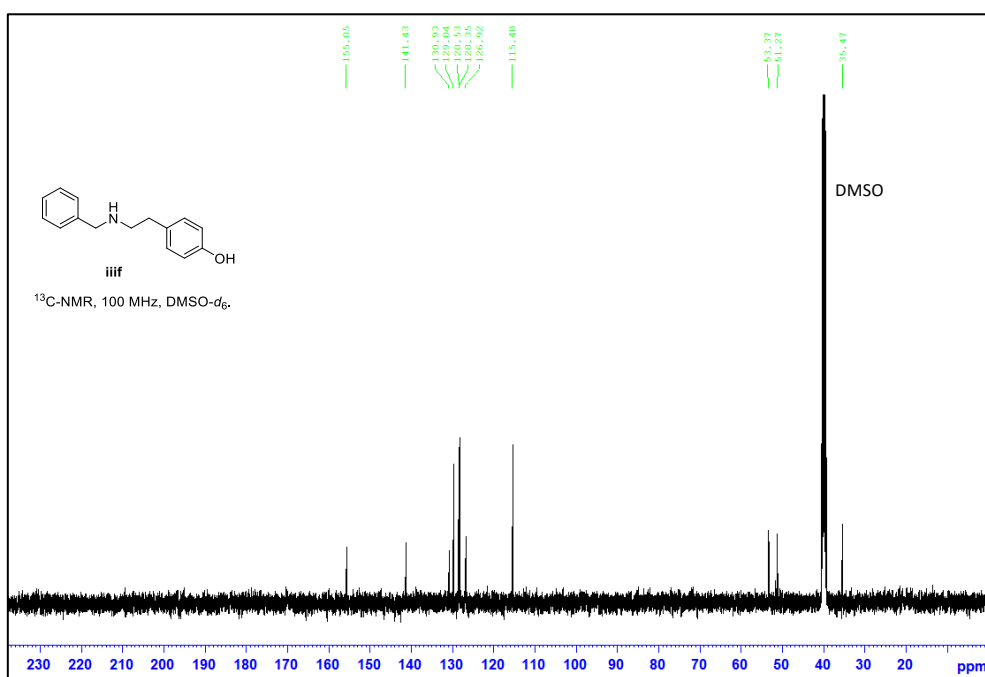
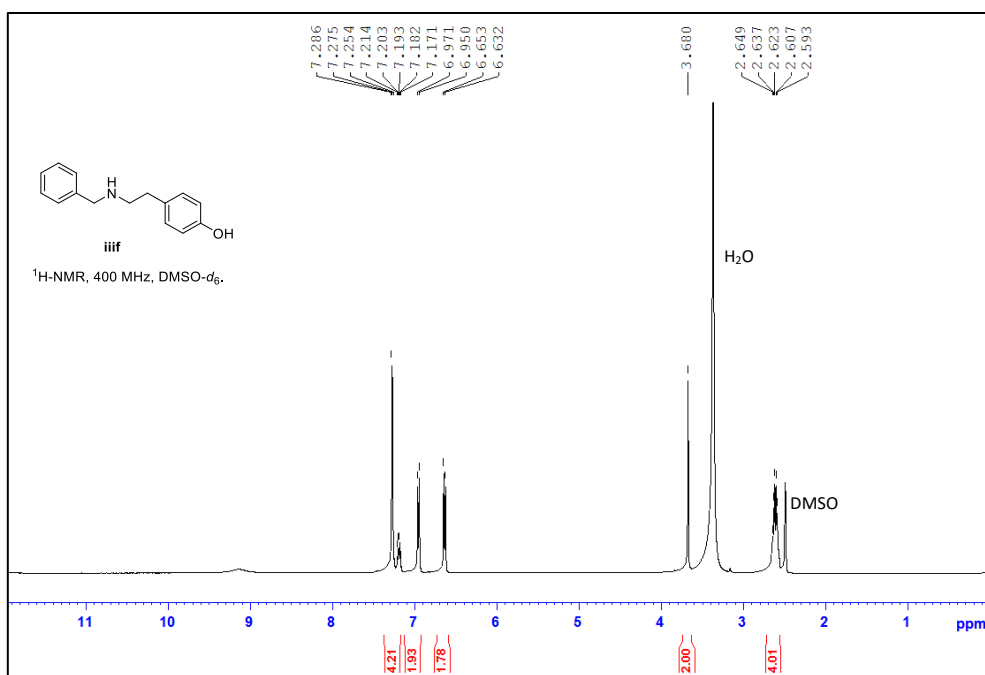


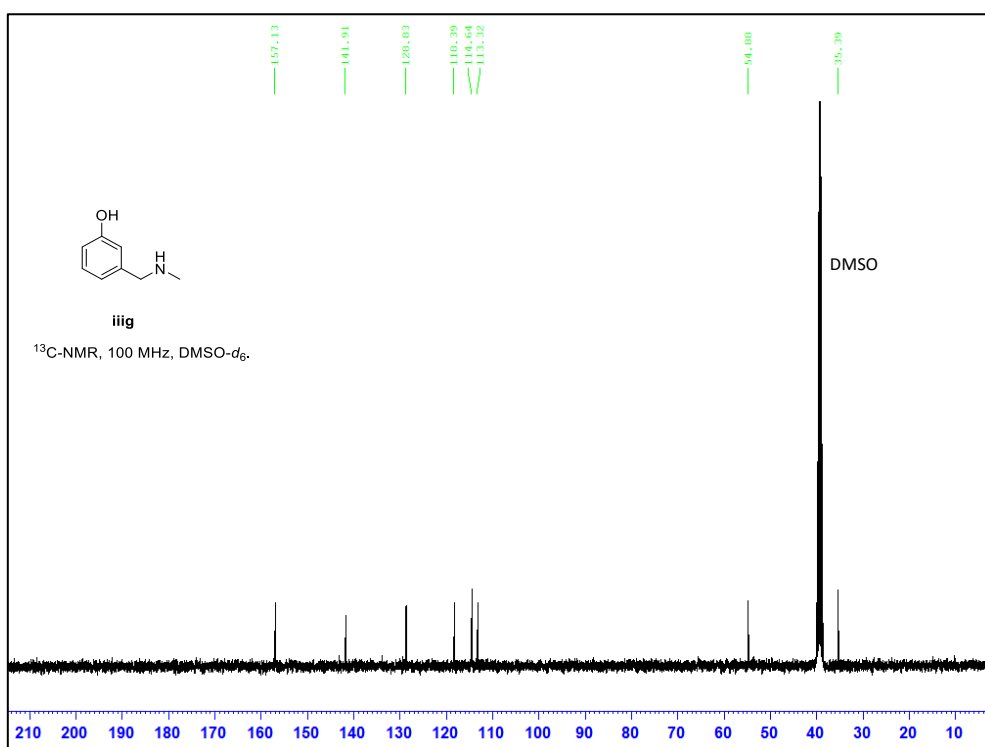
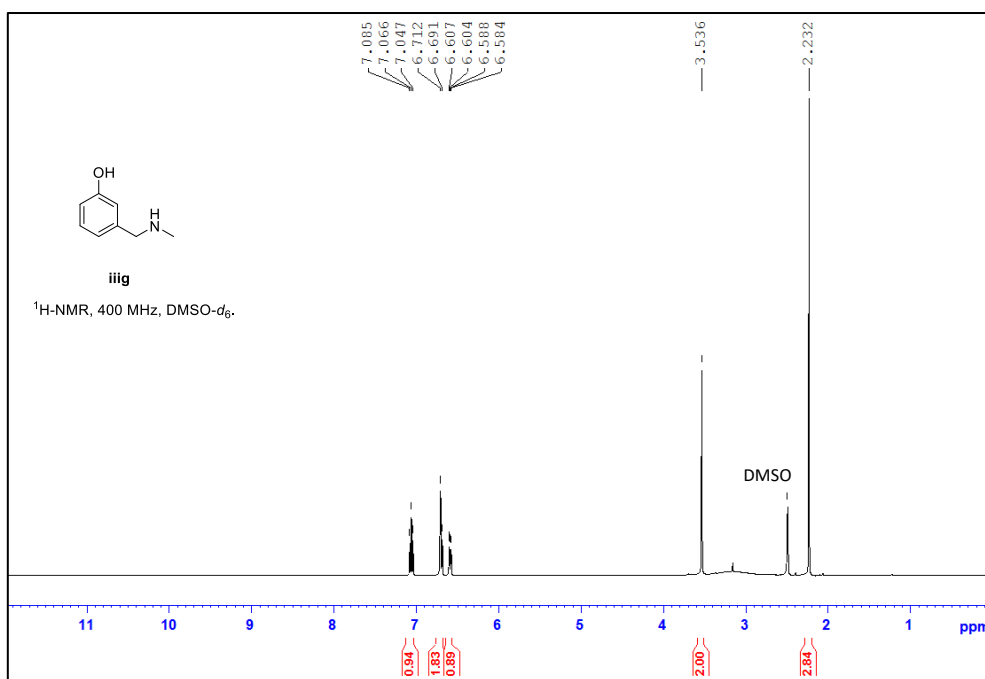


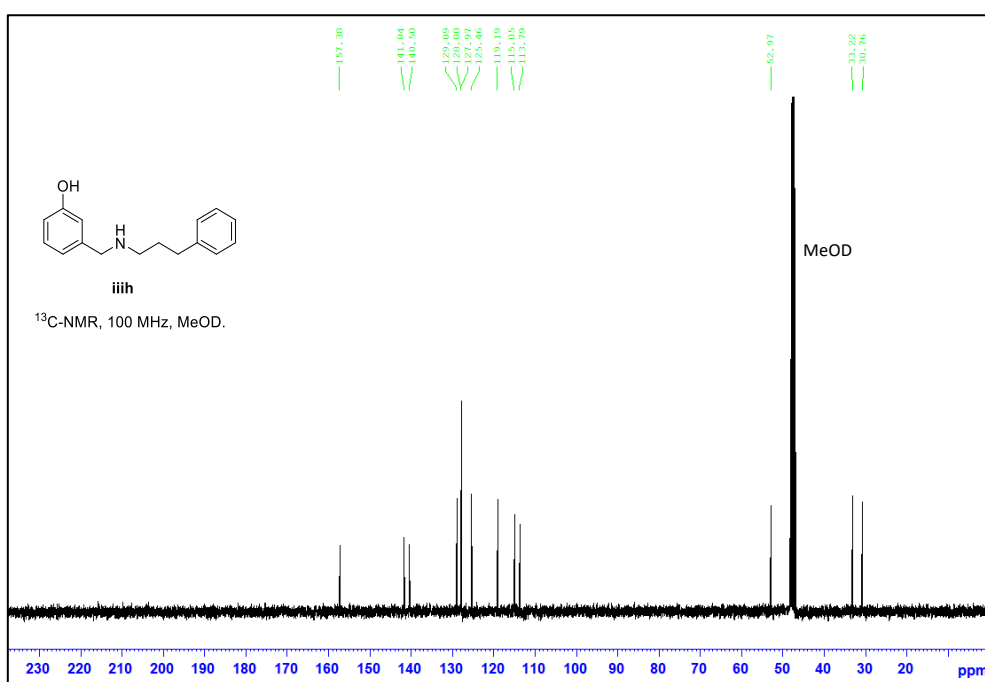
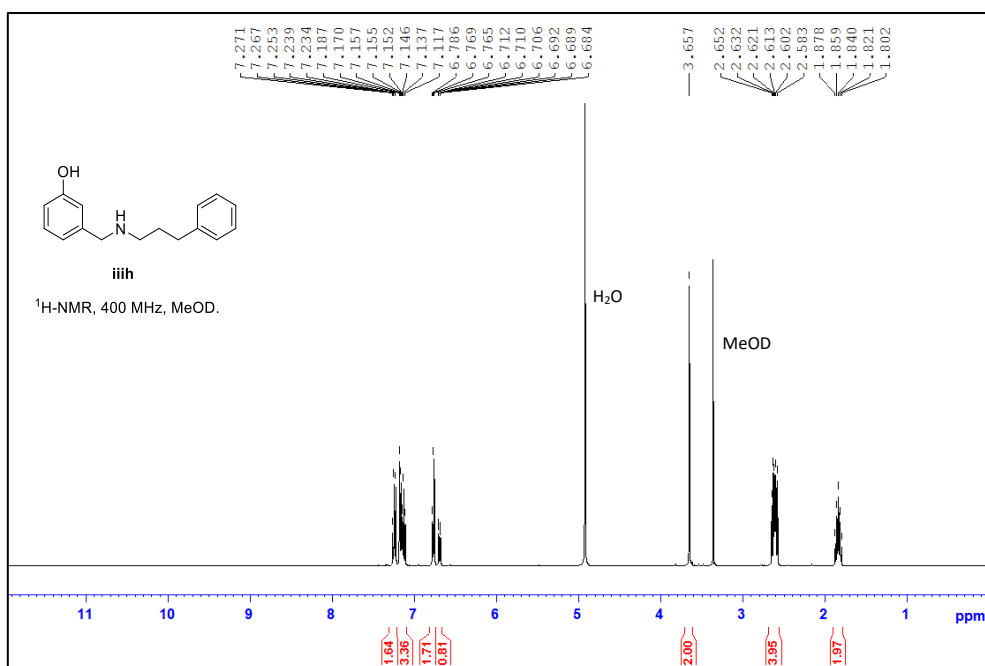




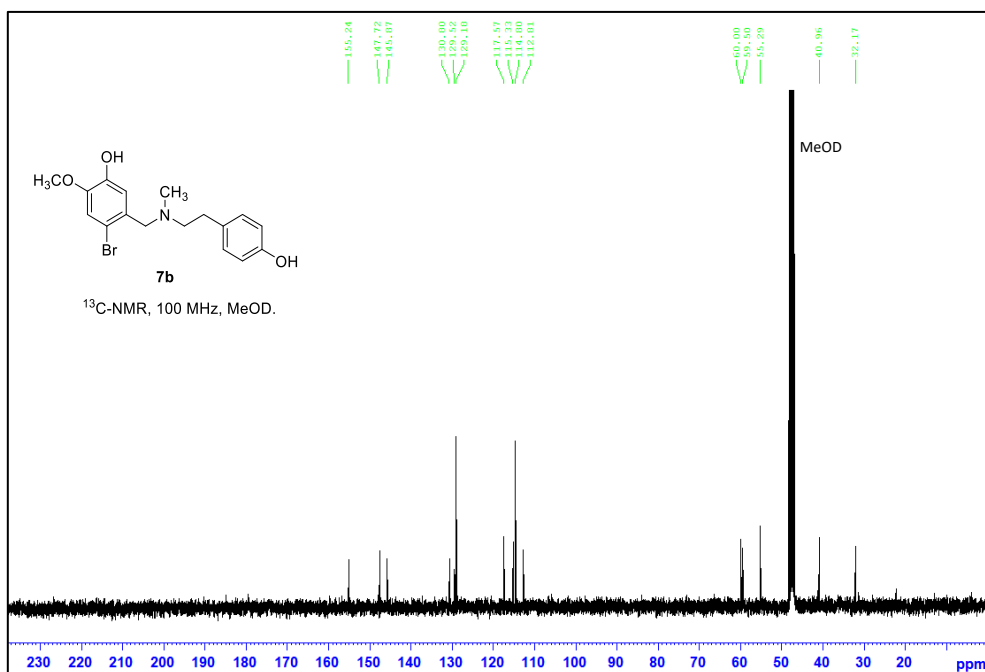
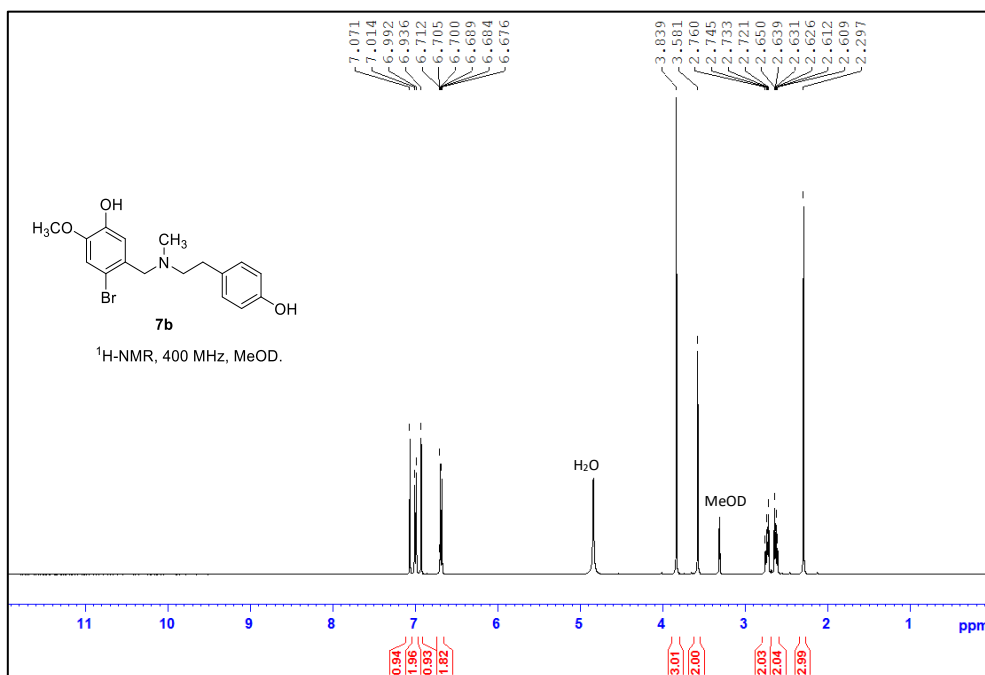


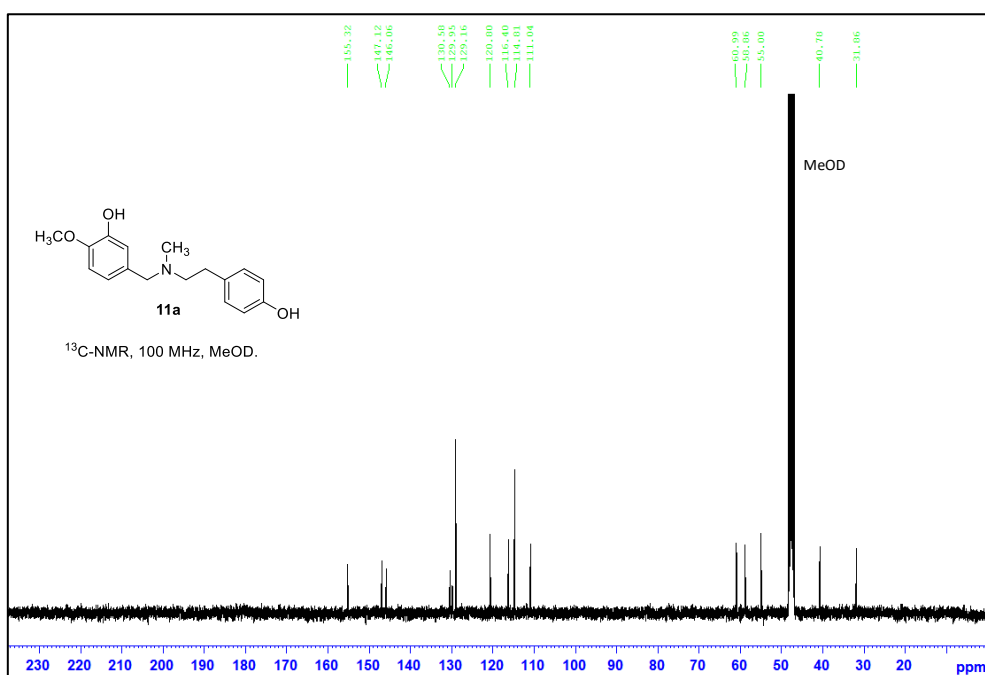
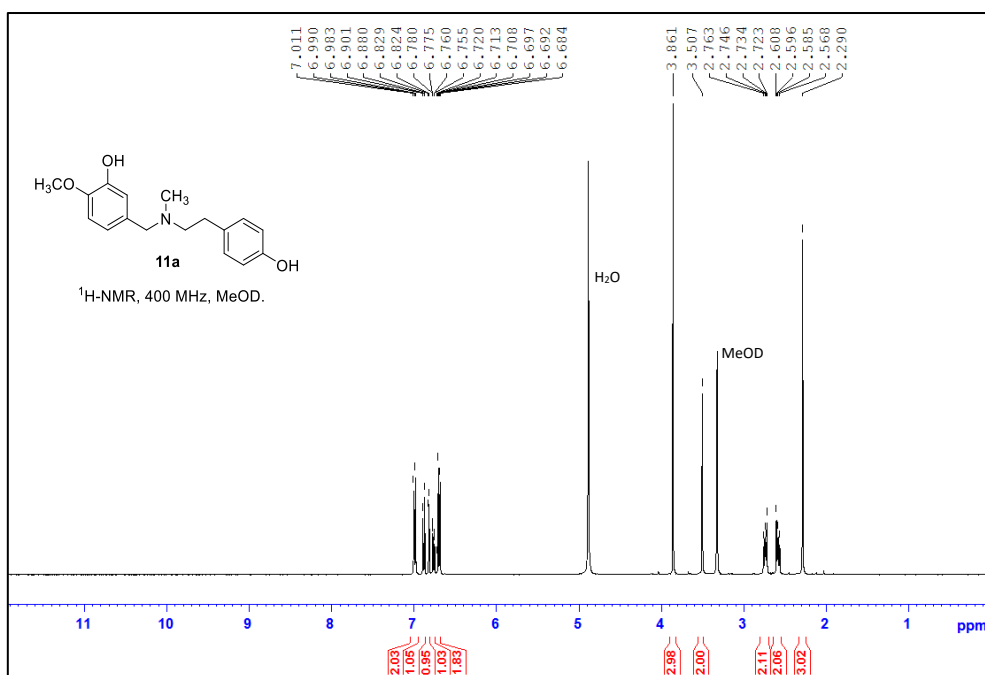


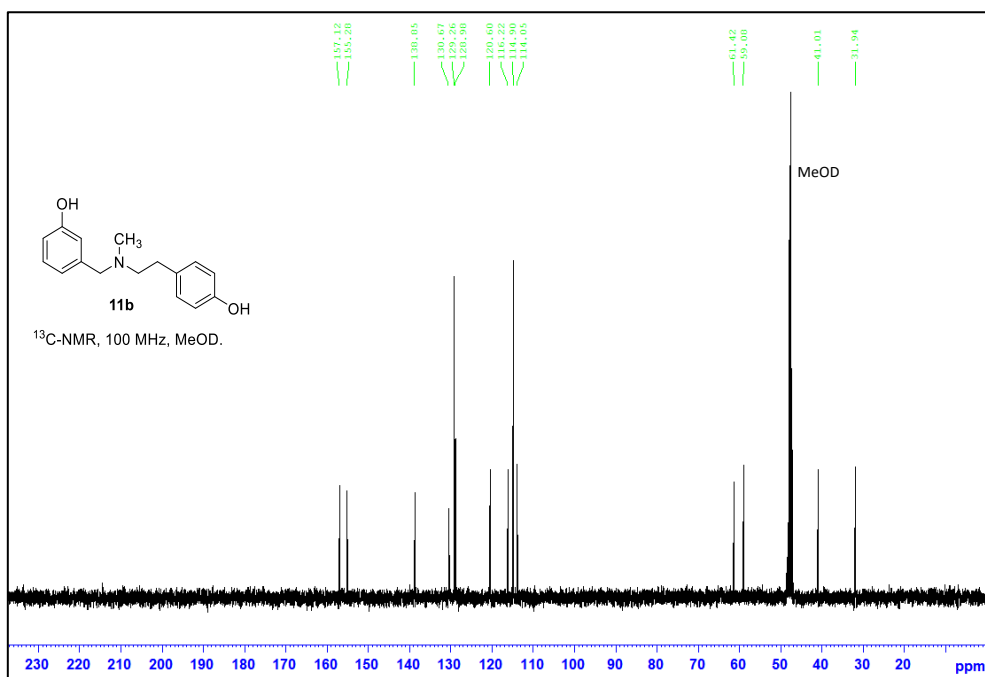
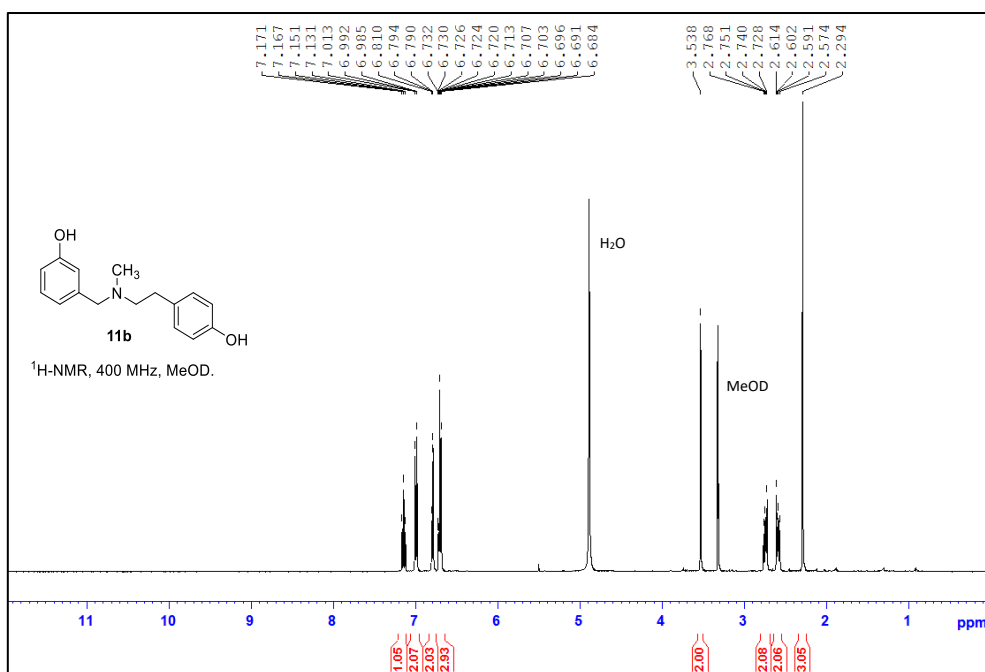


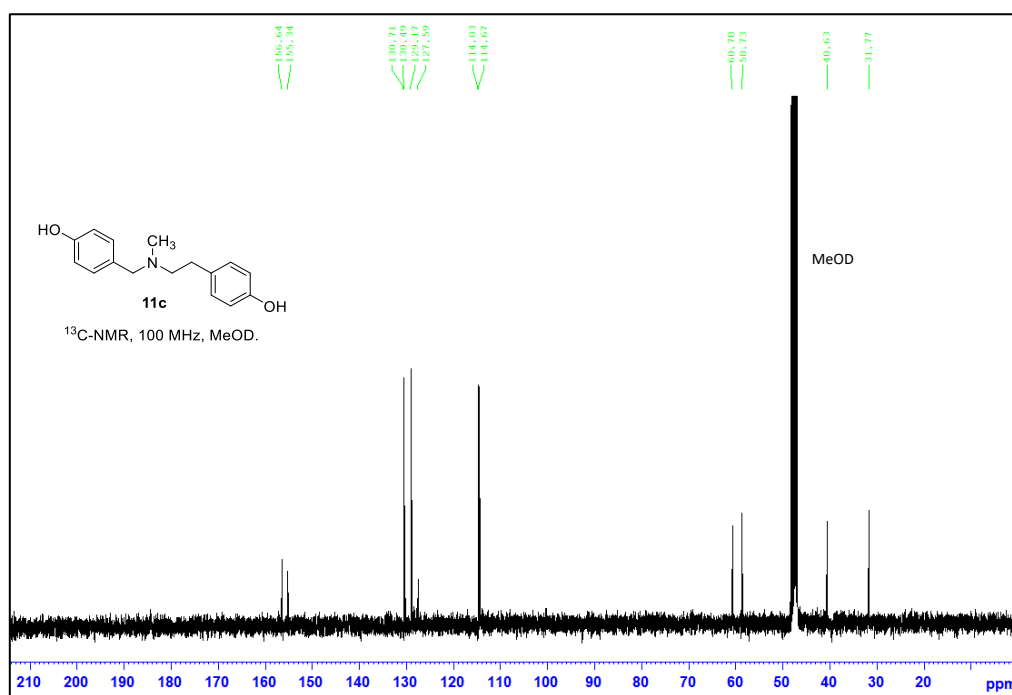
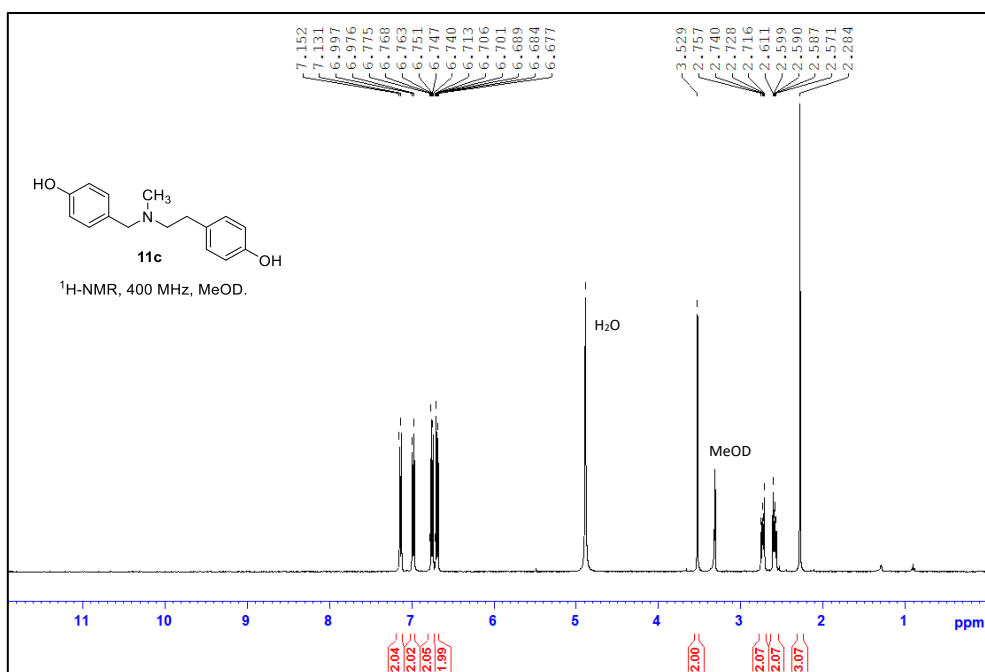


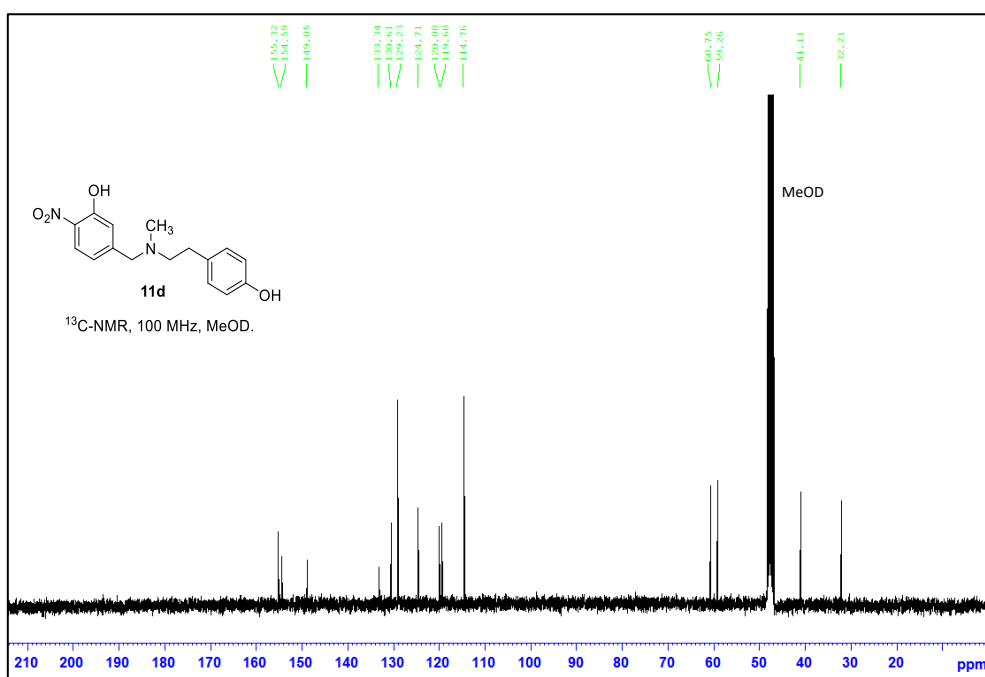
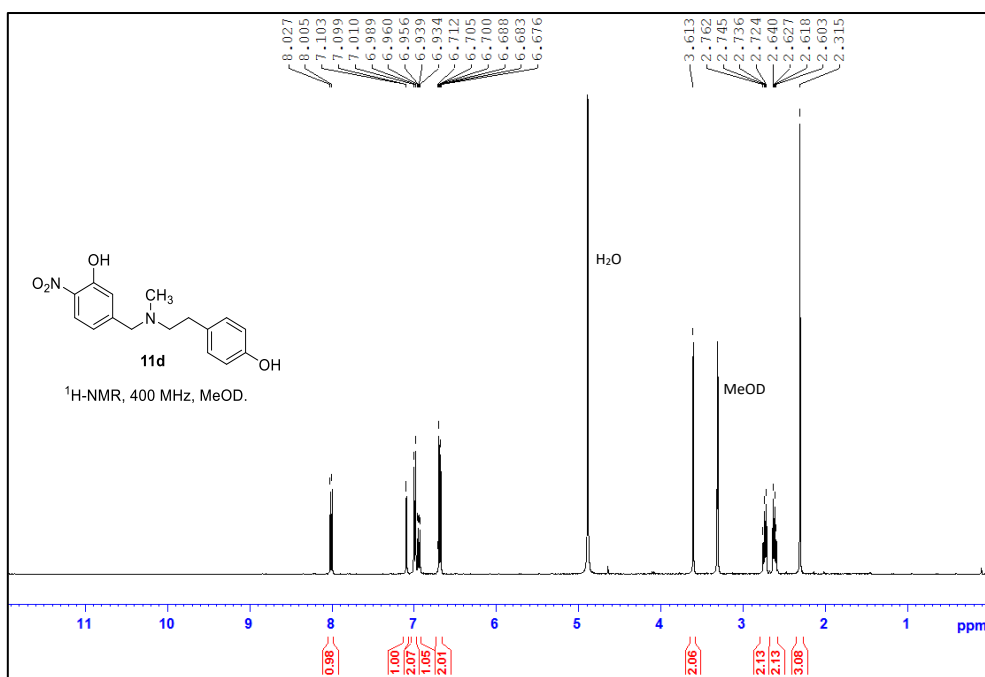
SM #3.2. NMR spectra of compounds **7b** and **11a-h**

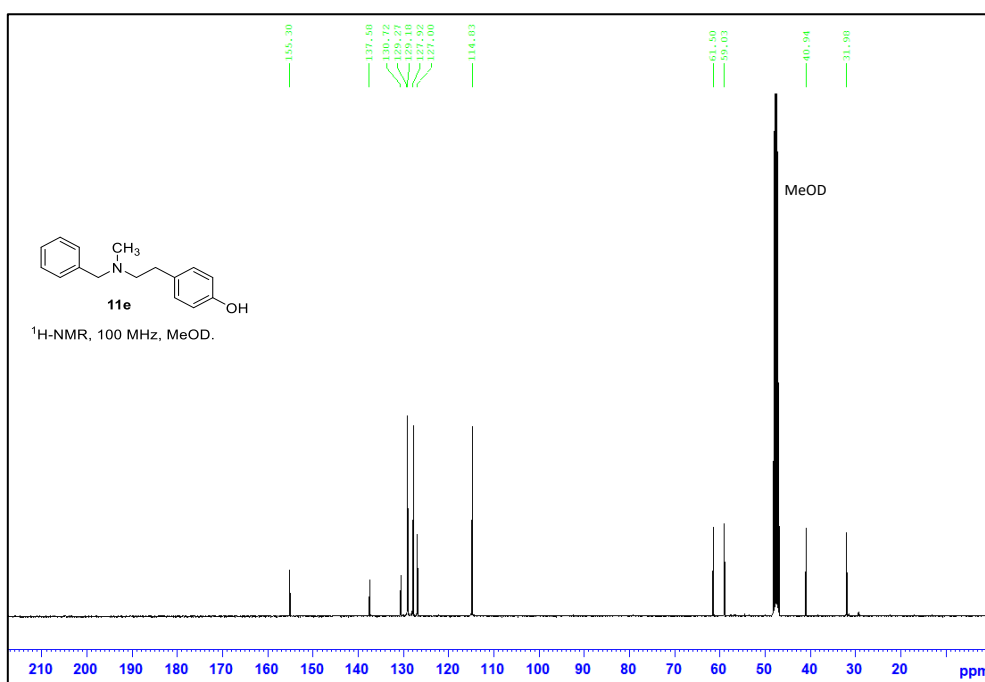
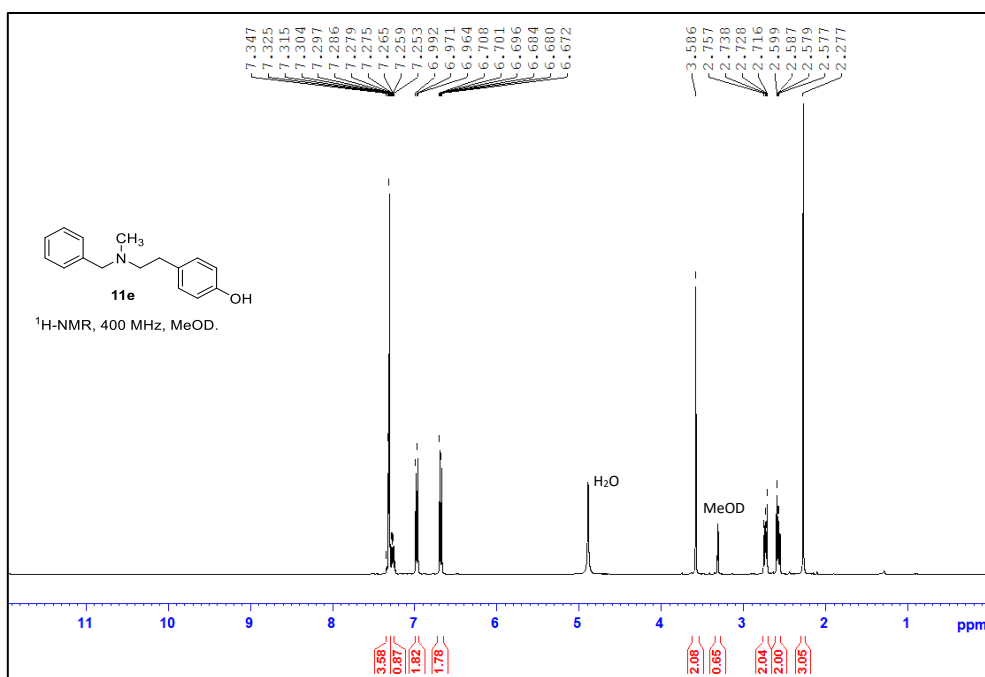


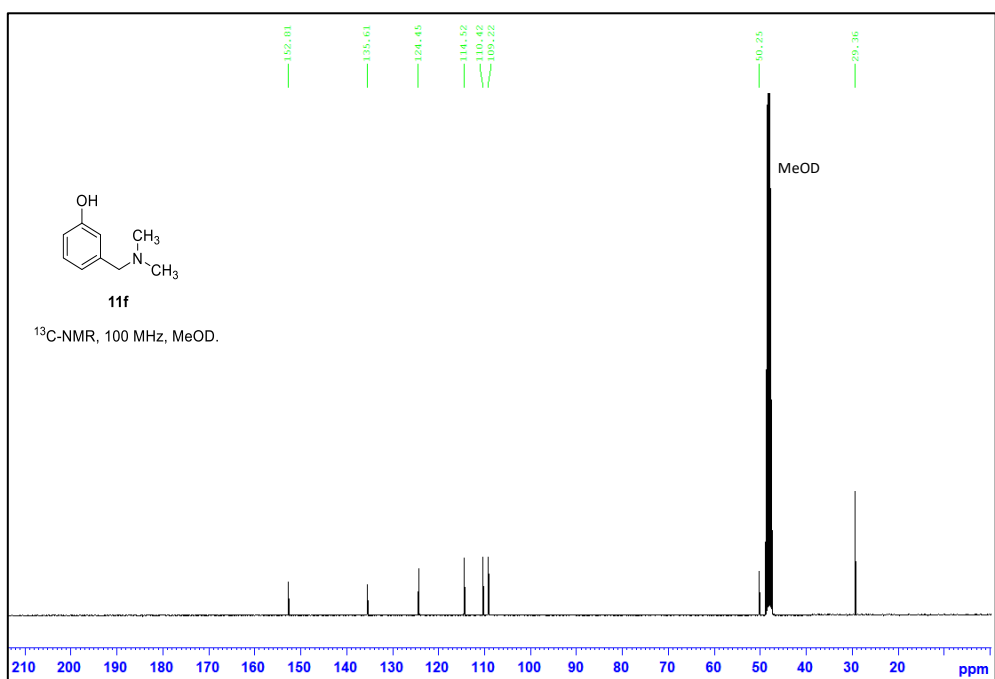
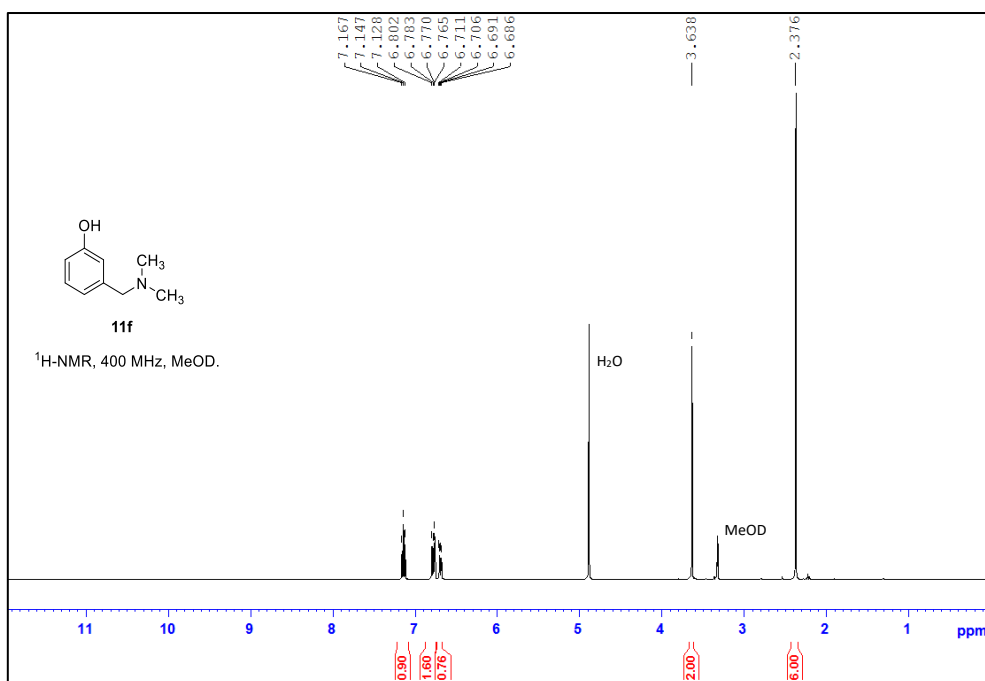


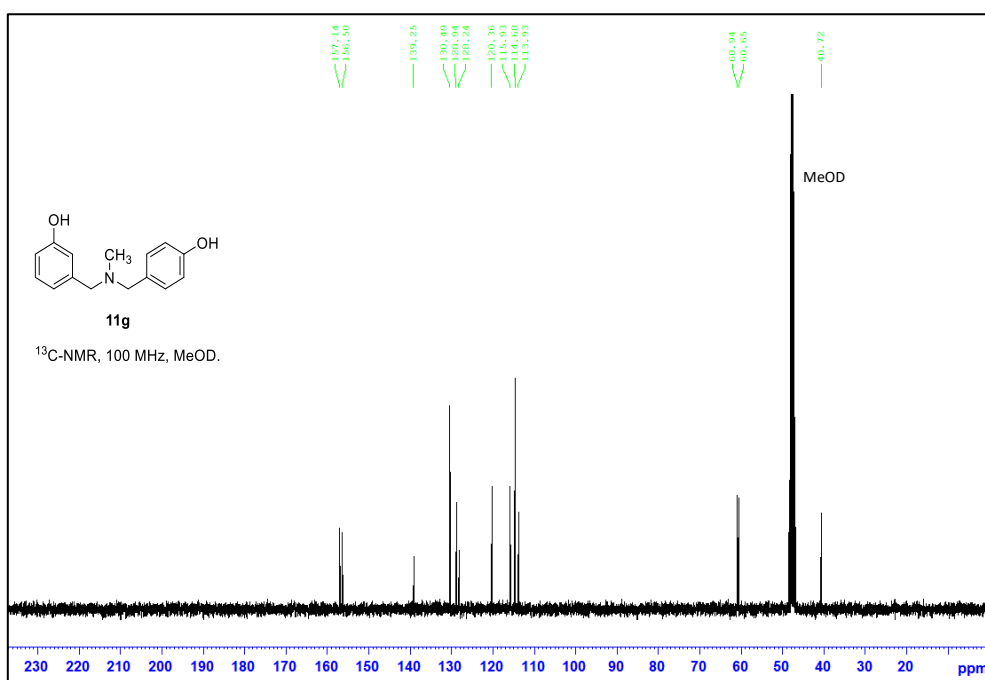
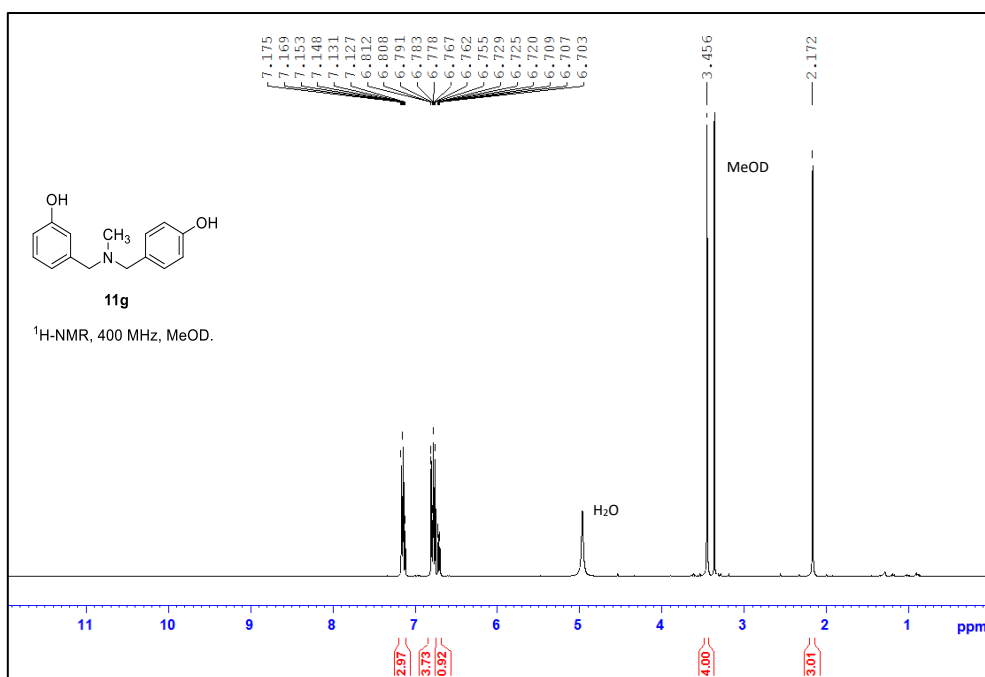


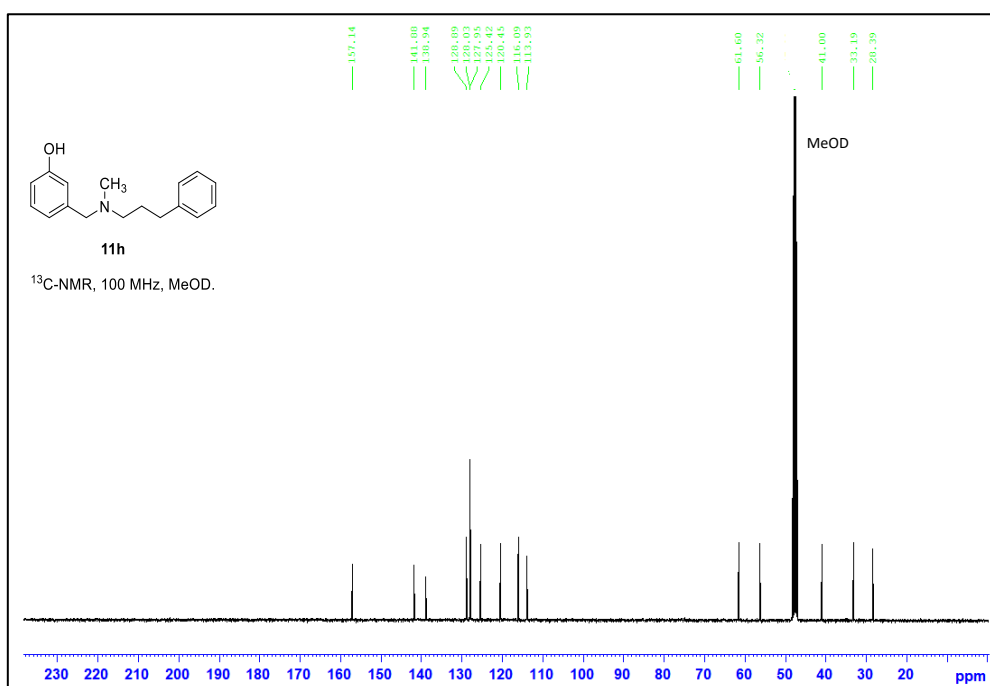
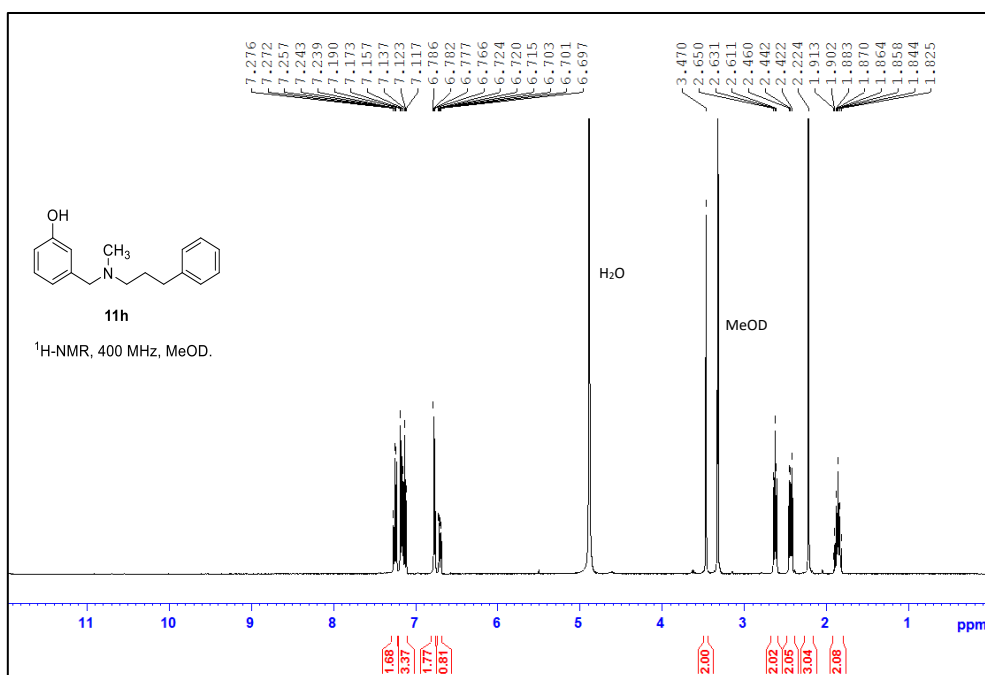




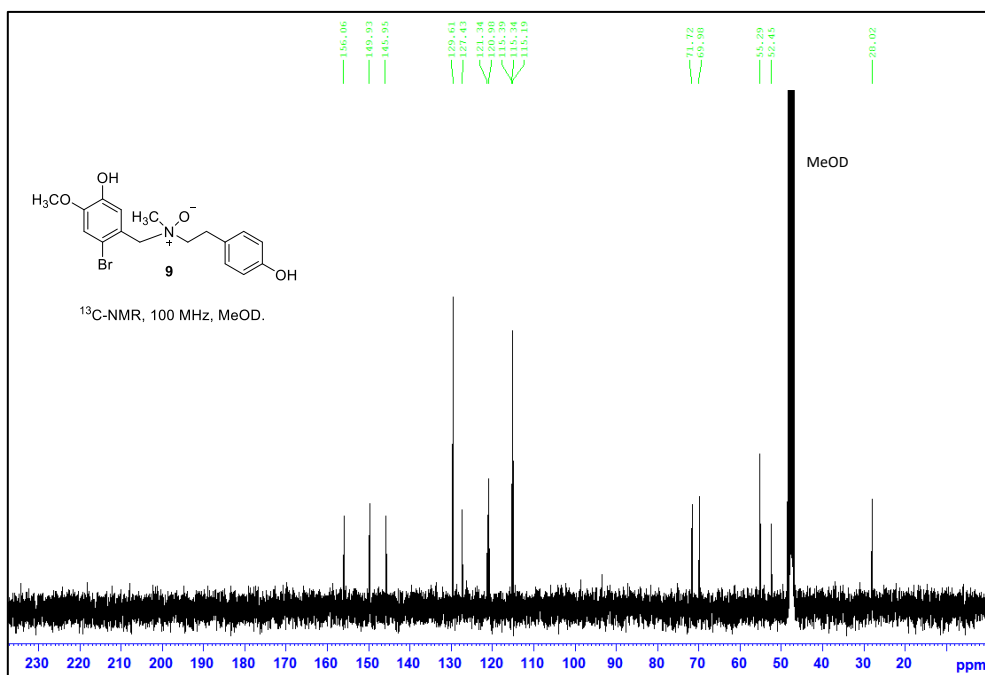
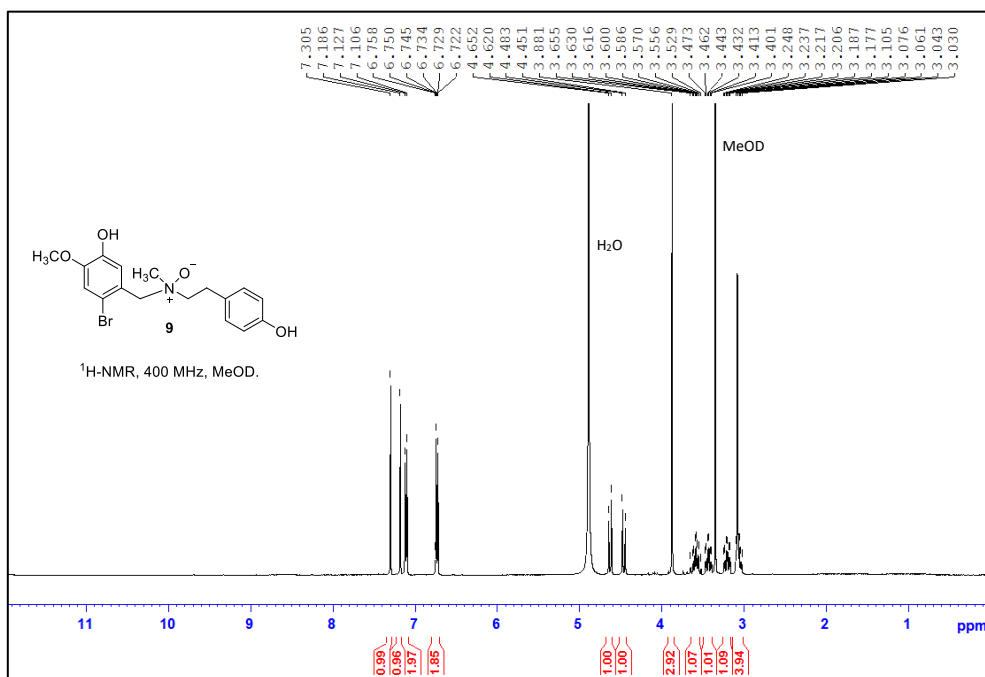


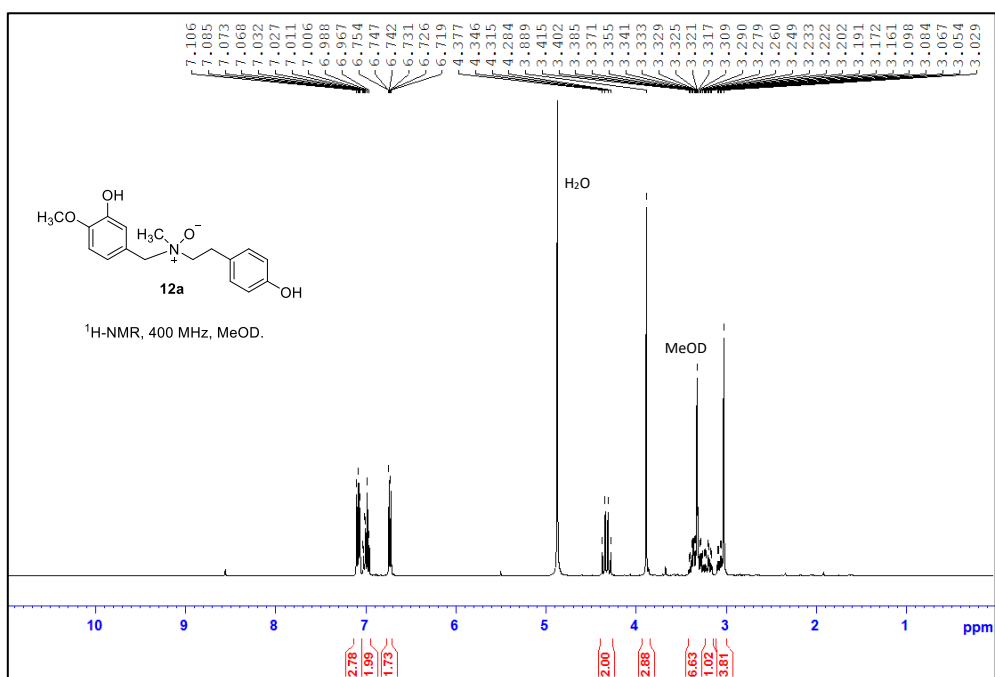
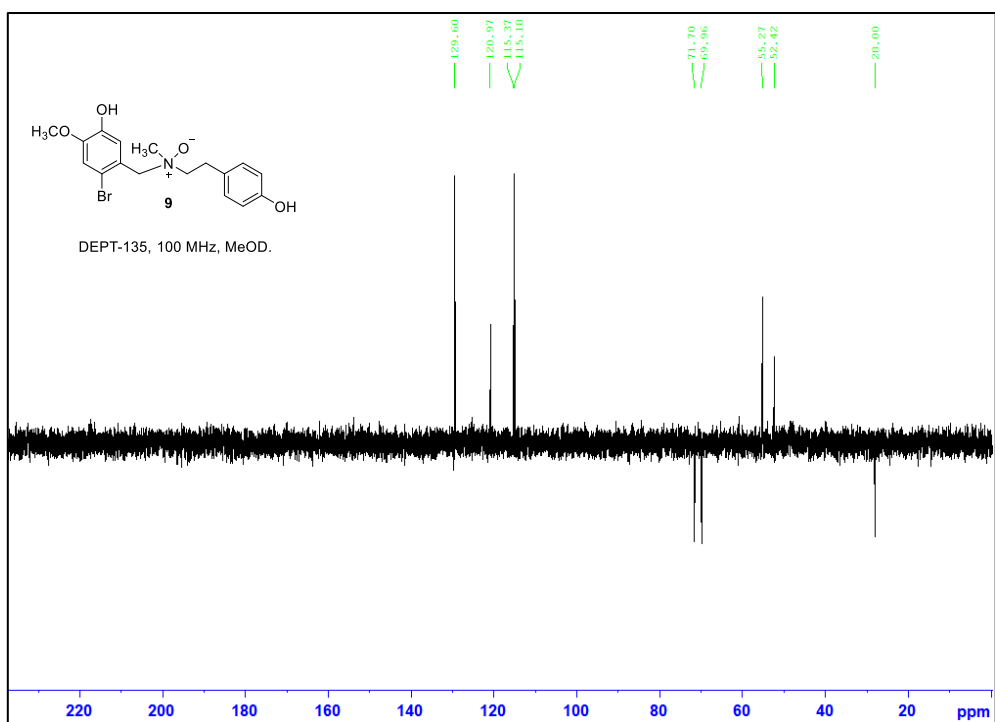


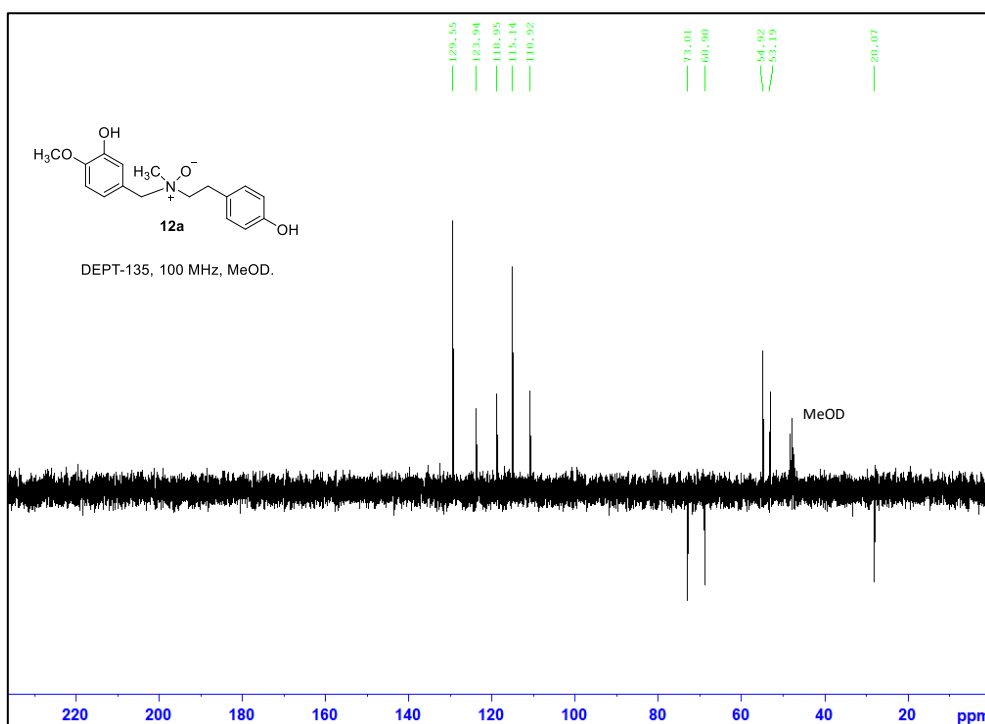
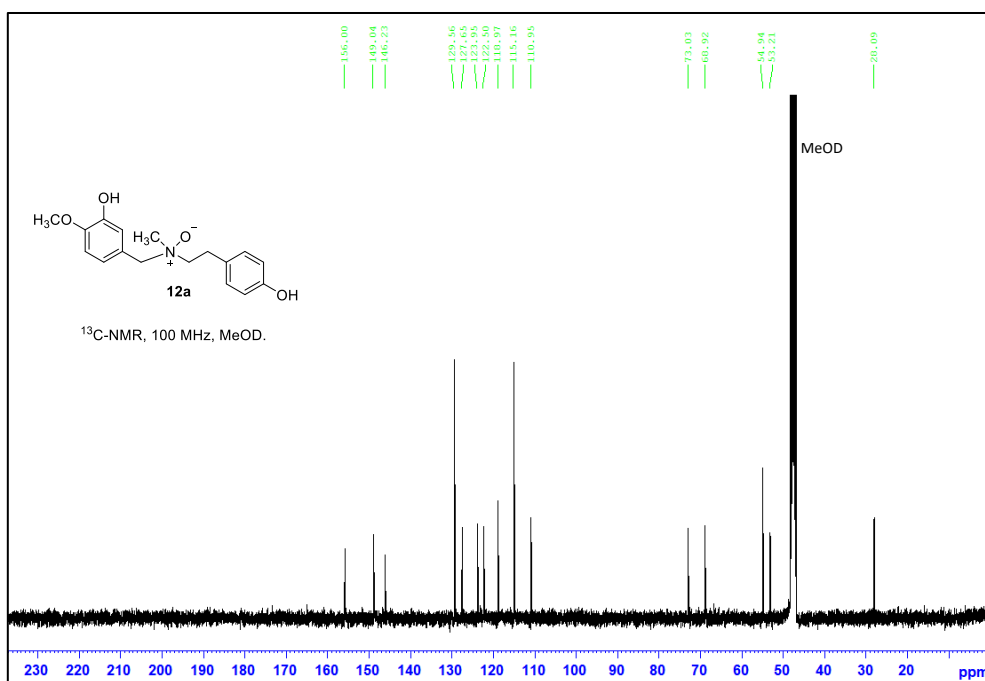


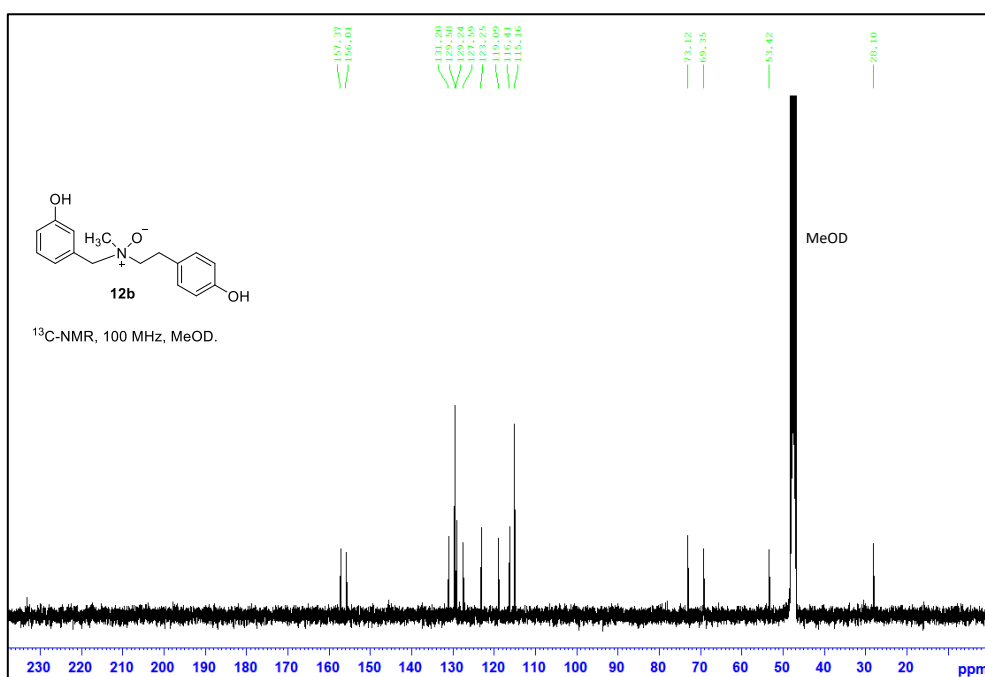
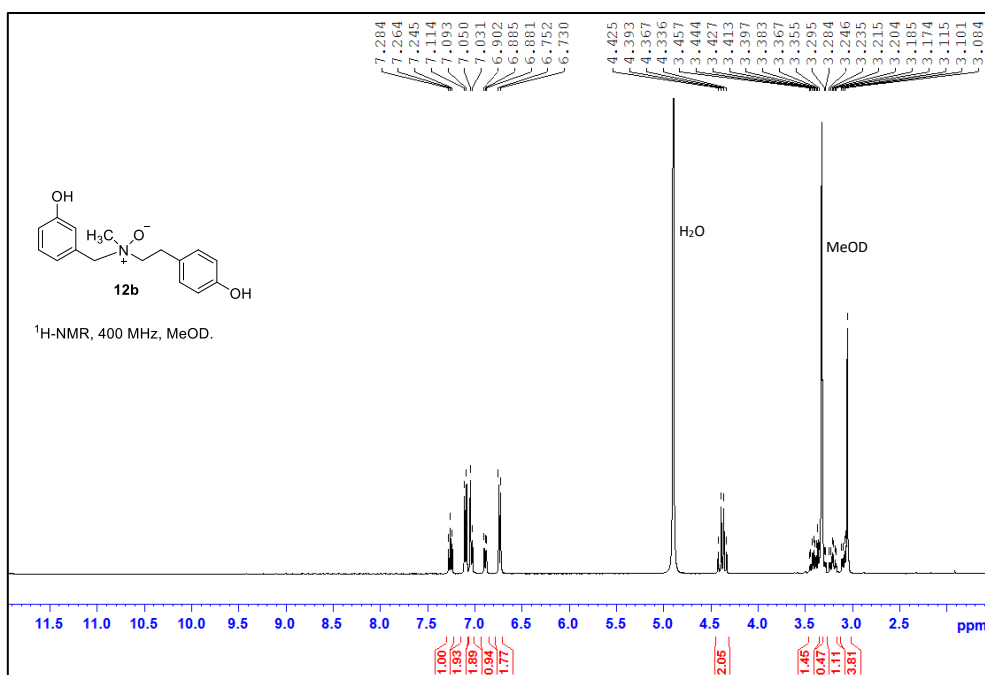


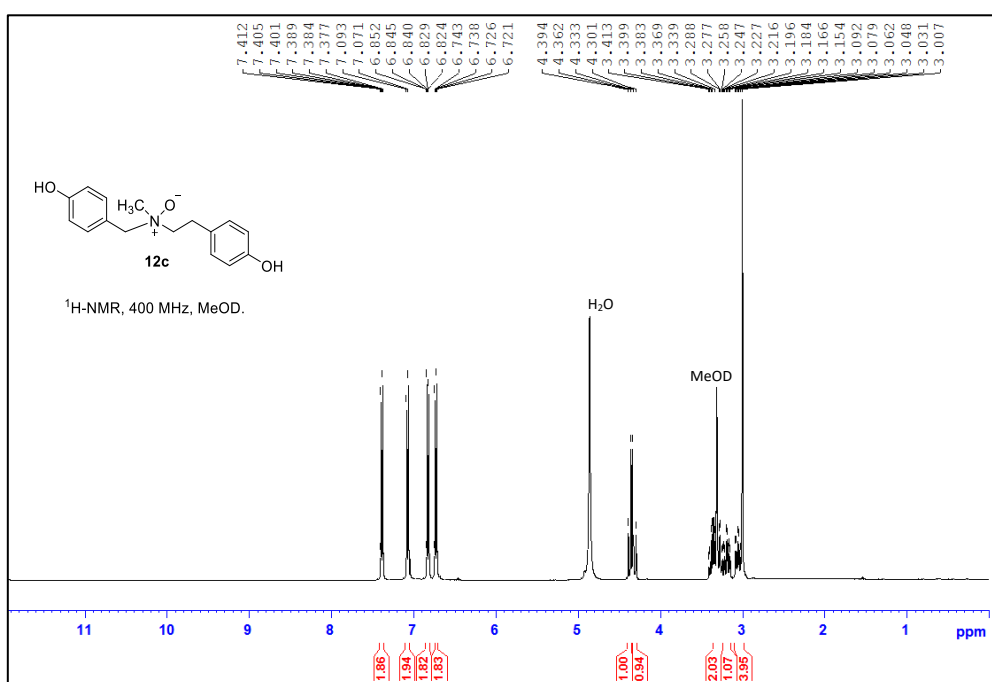
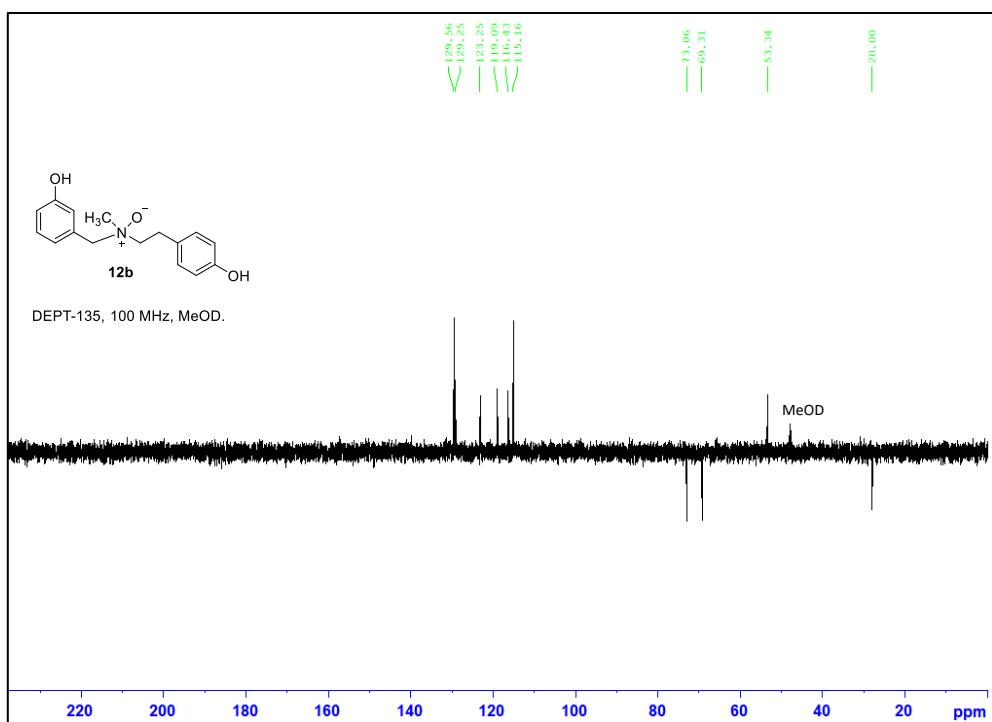
SM #3.3. NMR spectra of compounds **9** and **12a-h**

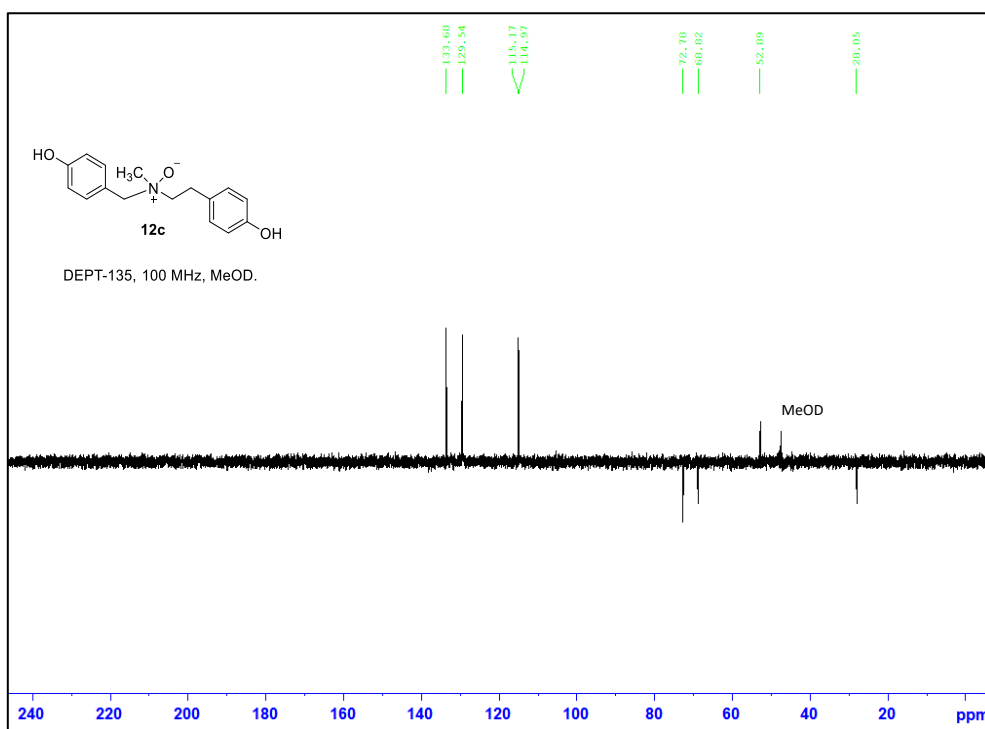
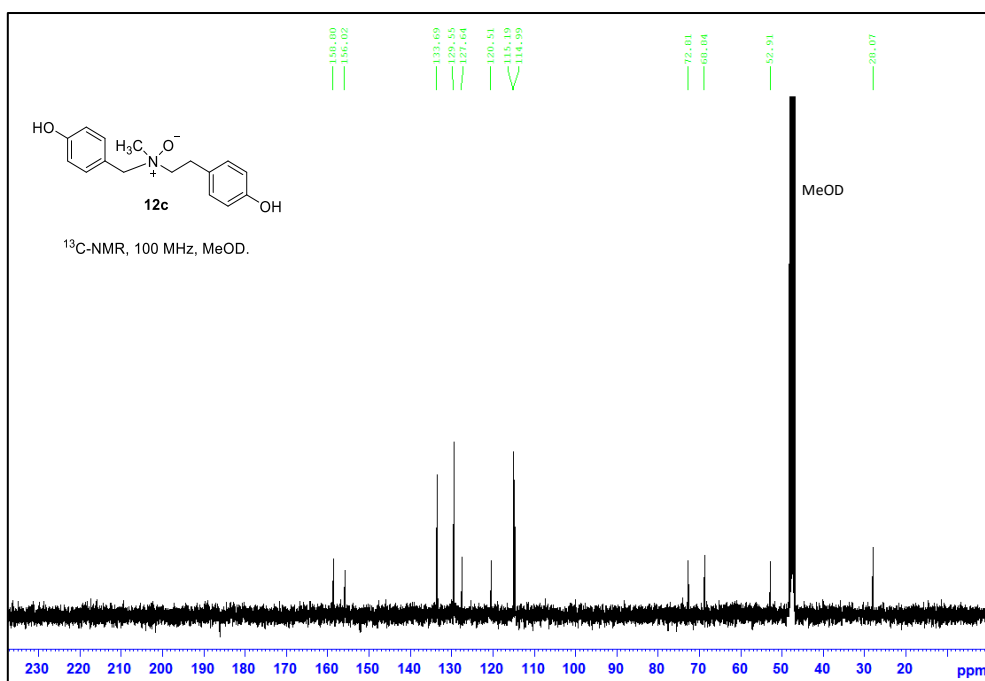


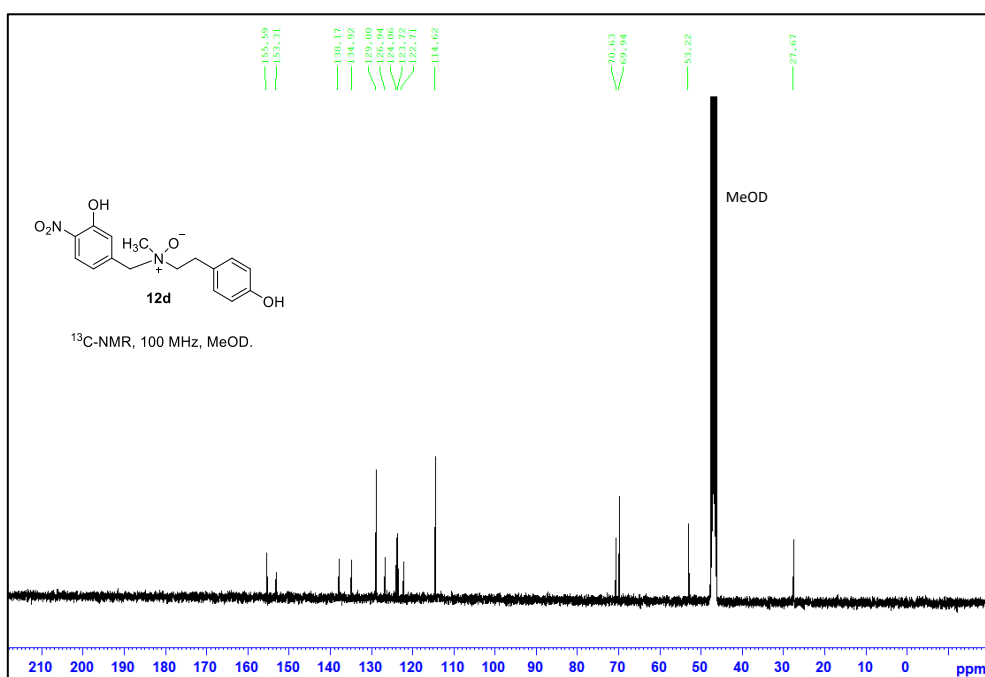
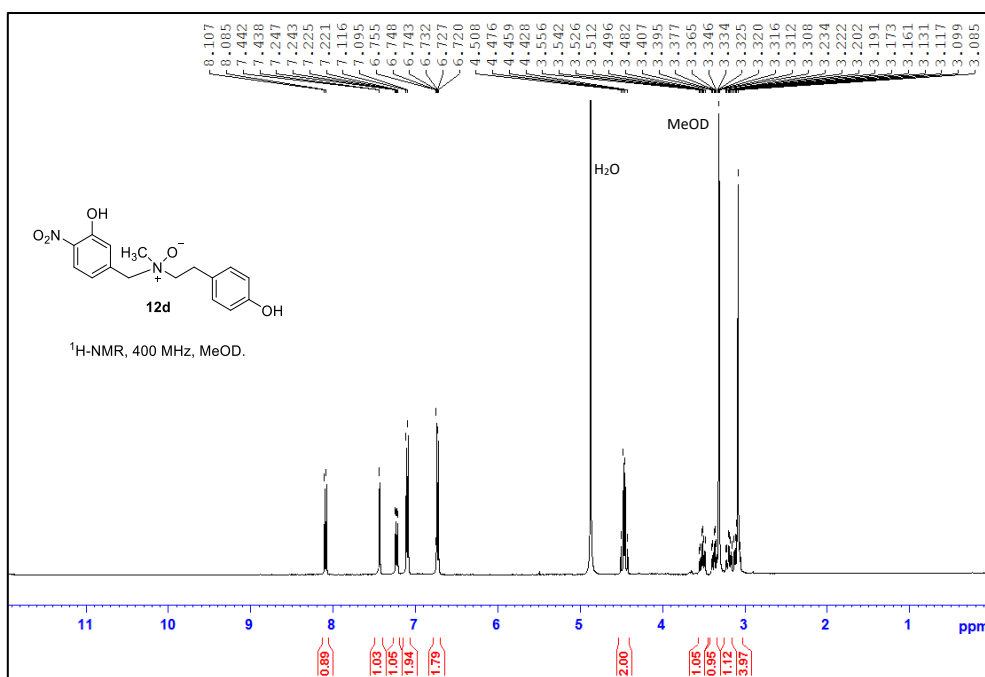


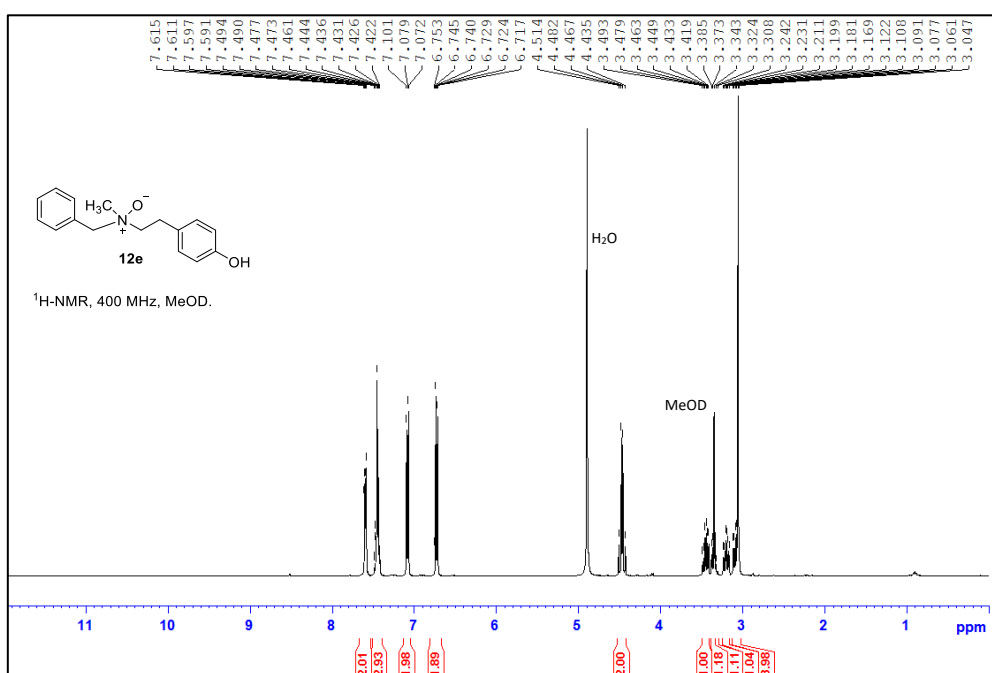
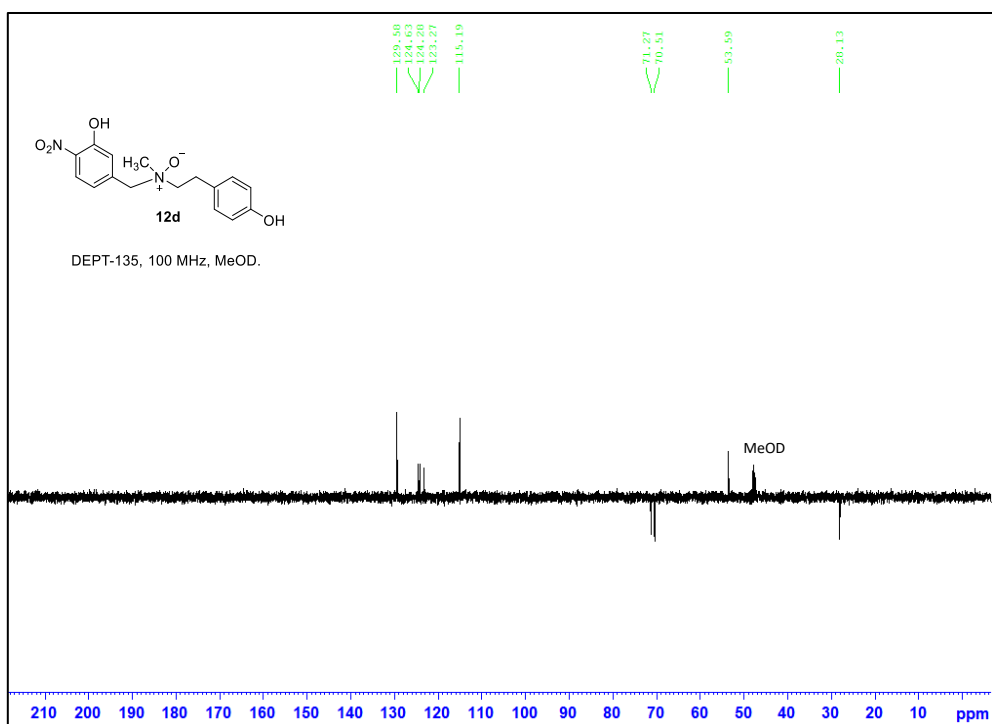


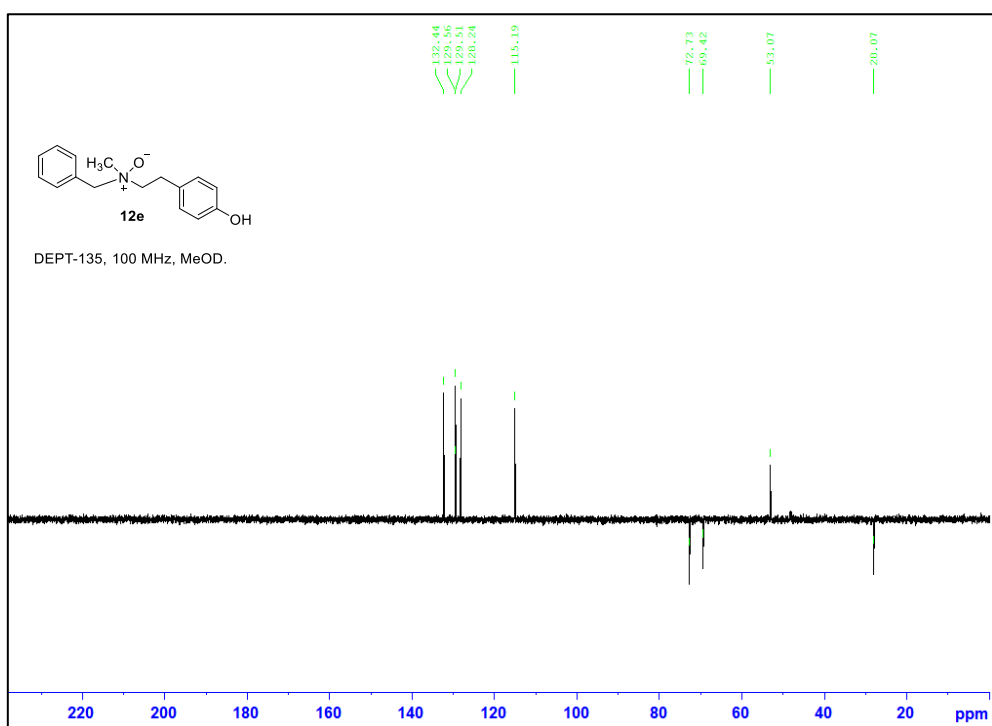
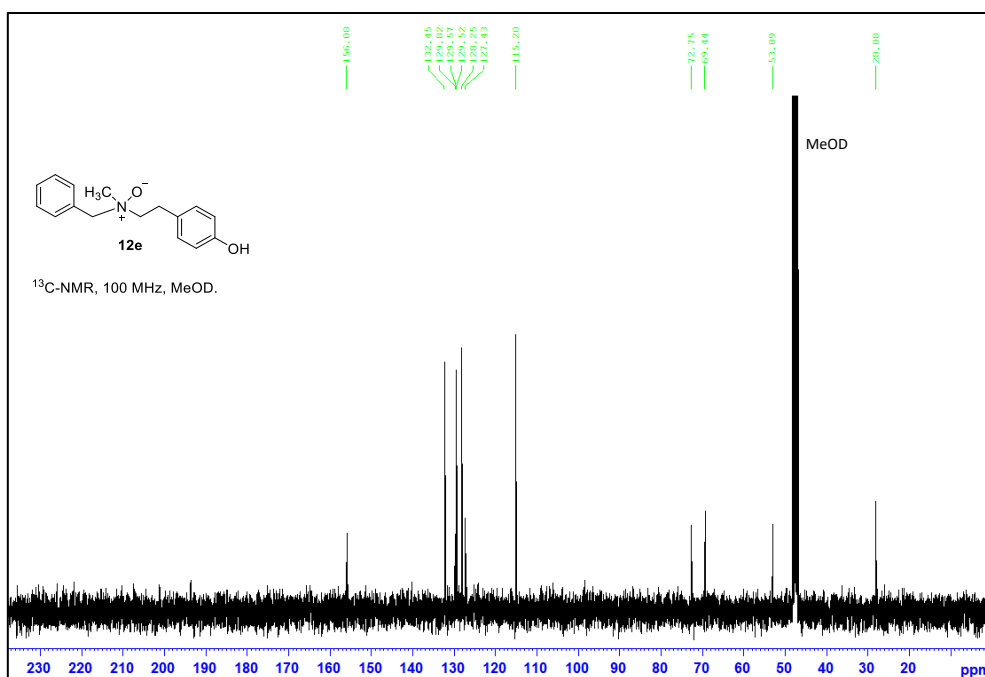


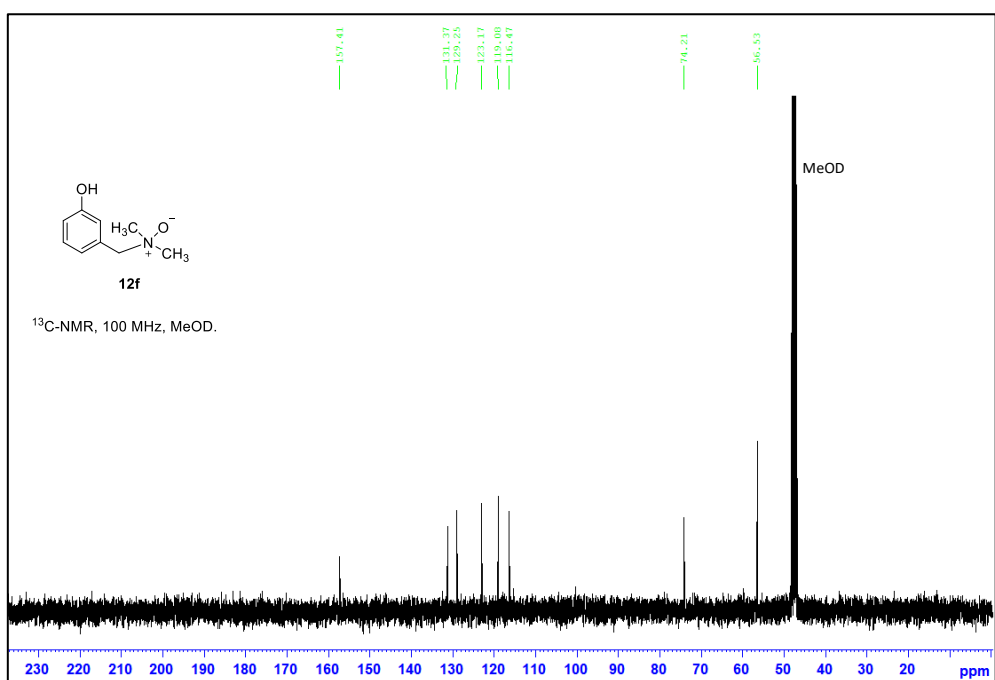
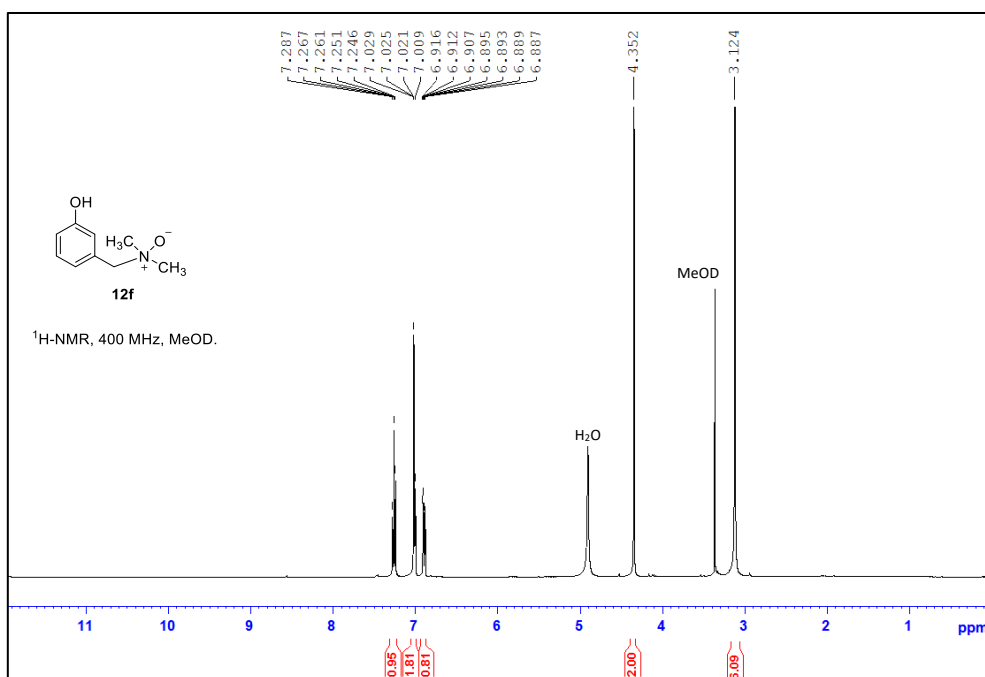


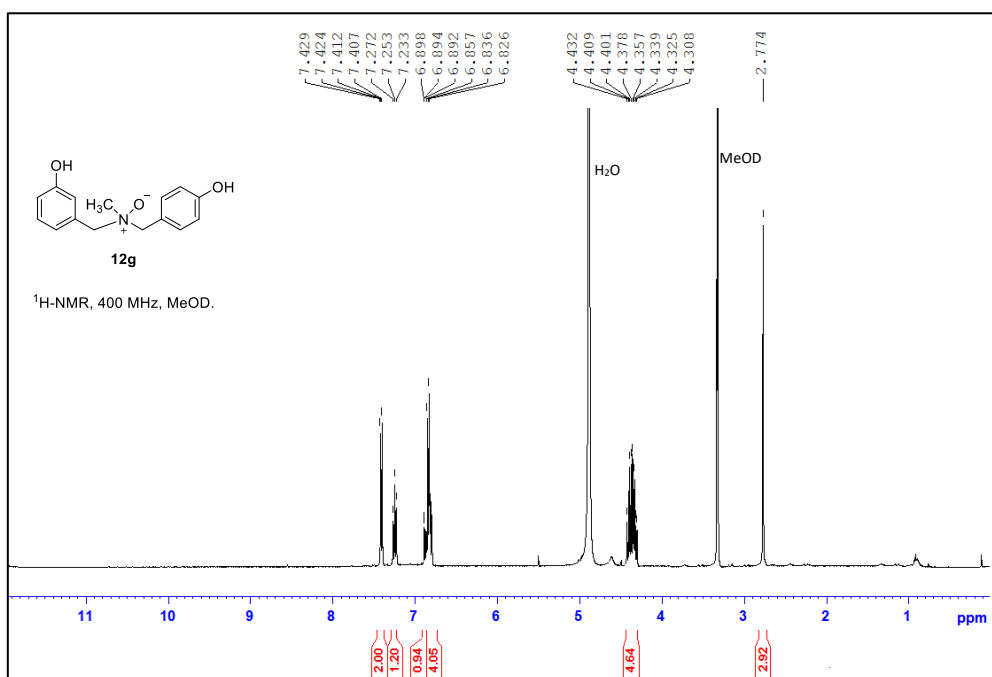
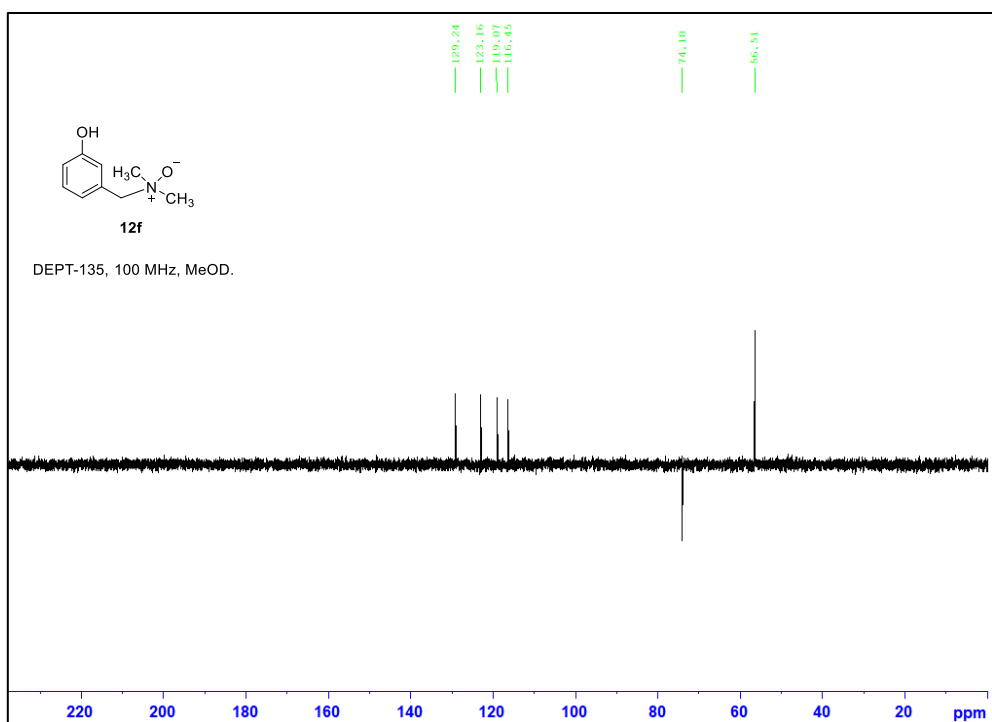


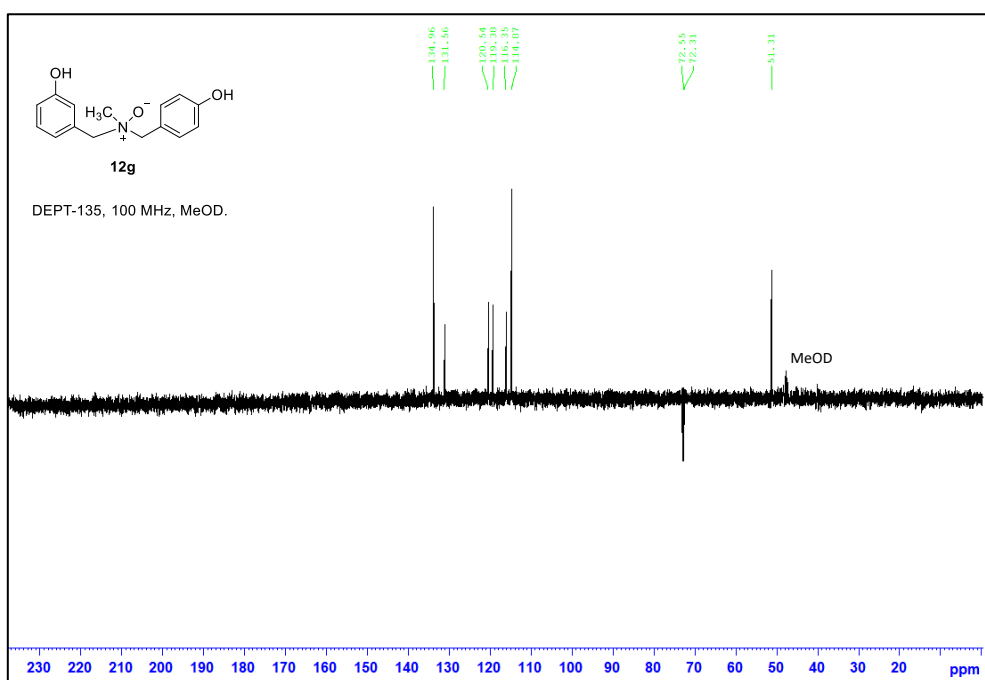
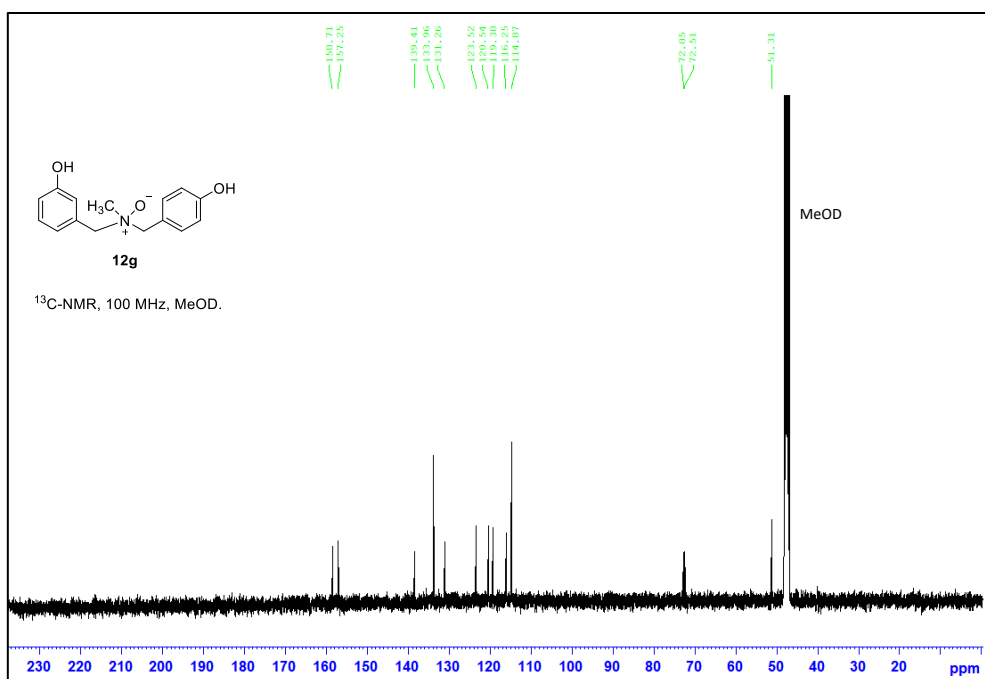


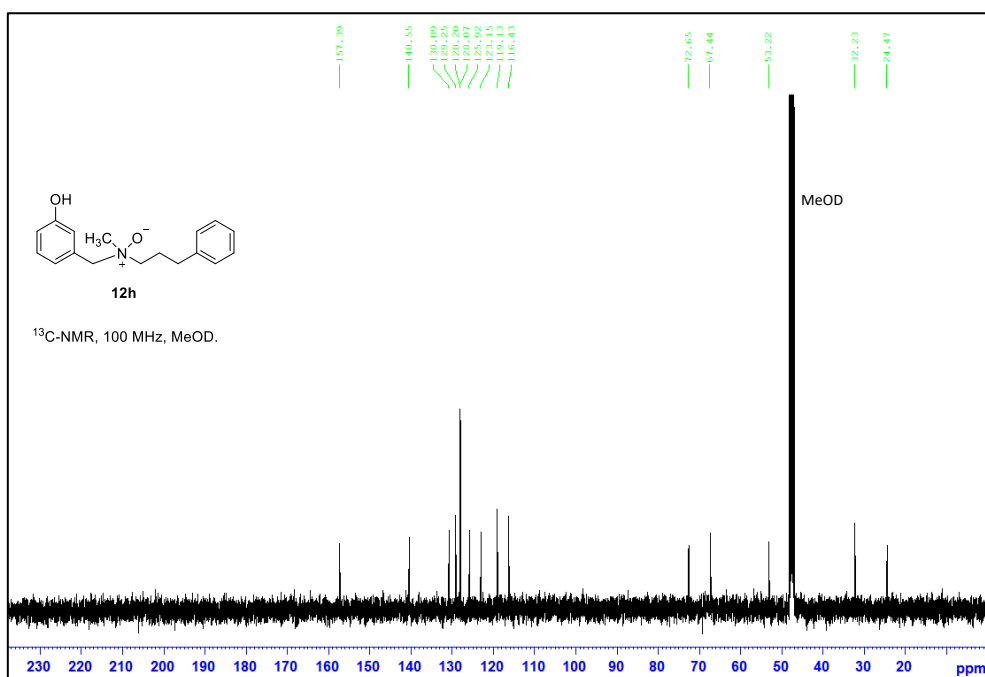
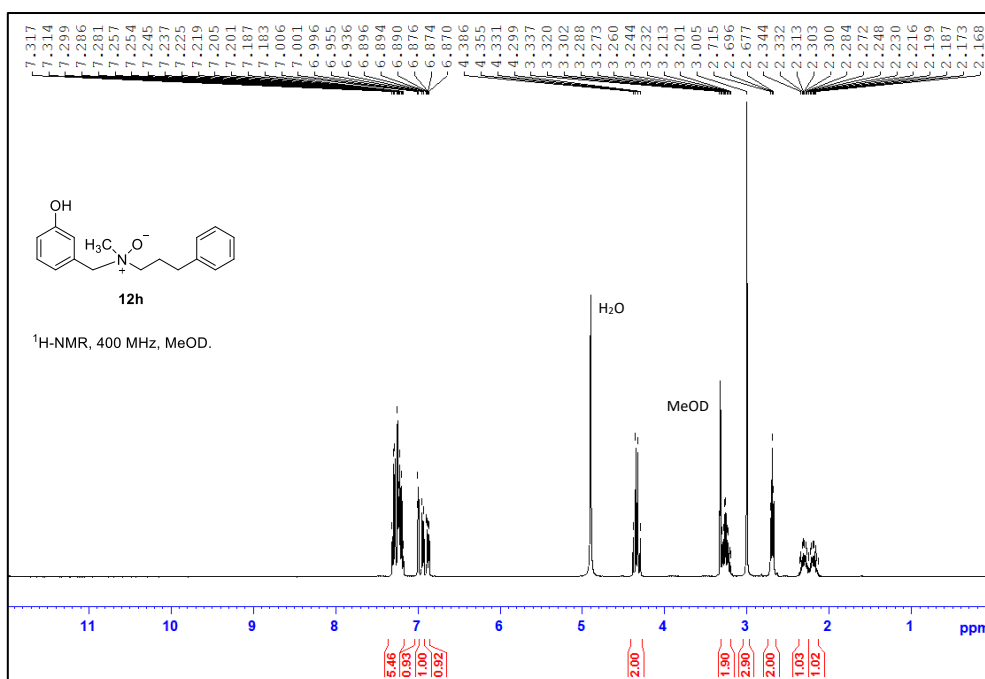


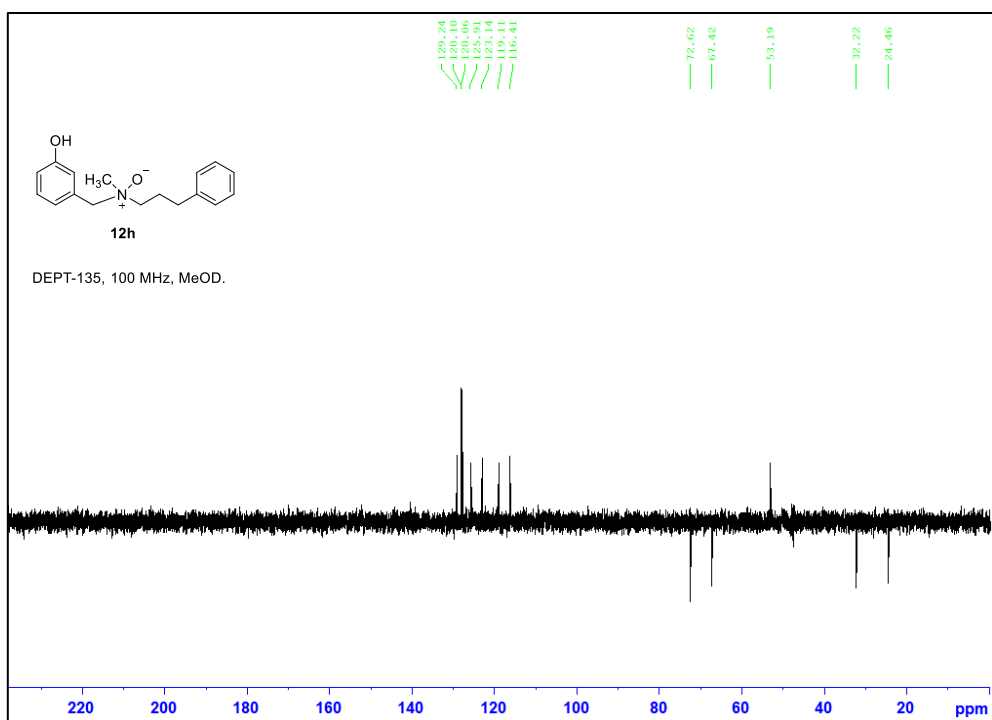












SM #3.1. NMR spectra of compound **10**

