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Supporting Information

Fluorous Bispidine: A Bifunctional Reagent for Copper-catalyzed Oxidation and Knoevenagel Condensation Reaction in Water

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Dimethyl 7-(4-methoxybenzyl)-3-methyl-9-oxo-2,4-di(pyridin-2-yl)-3,7-diazabicyclo[3.3.1]nonane-1,5-dicarboxylate (L1). White solid. Mp: 188-189 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.43 (d, *J* = 3.2 Hz, 2H), 7.87 (d, *J* = 7.9 Hz, 2H), 7.49 (td, *J* = 7.7, 1.7 Hz, 2H), 7.26 (d, *J* = 8.5 Hz, 2H), 7.12 (dd, *J* = 6.8, 5.1 Hz, 2H), 6.93 (d, *J* = 8.5 Hz, 2H), 4.68 (s, 2H), 3.86 (s, 3H), 3.81 (s, 6H), 3.30 (s, 2H), 3.02 (d, *J* = 12.7 Hz, 2H), 2.50 (d, *J* = 12.2 Hz, 2H), 1.96 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 203.7, 168.5, 159.1, 158.5, 149.0, 136.0, 131.4, 129.0, 123.4, 122.8, 113.7, 73.7, 62.1, 61.6, 58.8, 55.4, 52.4, 43.2. HRMS (ESI): calcd for $C_{30}H_{33}N_4O_6$ $(M+H)^+$ 545.2395, found 545.2403.

(4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-Heptadecafluoroundecyloxy)phenyl)methanol (p2'). White solid. Mp: 83-84 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.29 (d, *J* = 8.7 Hz, 2H), 6.88 (d, *J* = 8.6 Hz, 2H), 4.61 (s, 2H), 4.04 (t, *J* = 5.9 Hz, 2H), 2.38 – 2.25 (m, 2H), 2.14 – 2.05 (m, 2H), 1.75 (s, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 158.2, 133.5, 128.7, 114.5, 66.4, 65.0, 27.9 (t, *J* = 22.7 Hz), 20.6. HRMS (EI): calcd for $C_{18}H_{13}F_{17}O_2$ (M^+) 584.0639, found 584.0642.

1-(Chloromethyl)-4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy)benzene (2). White solid. Mp: 64-65 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 (d, *J* = 8.6 Hz, 2H), 6.88 (d, *J* = 8.6 Hz, 2H), 4.57 (s, 2H), 4.04 (t, *J* = 5.9 Hz, 2H), 2.38 – 2.24 (m, 2H), 2.14 – 2.07 (m, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 158.7, 130.2, 130.1, 114.7, 66.4, 46.1, 27.9 (t, *J* = 22.8 Hz), 20.6. HRMS (EI): calcd for $C_{18}H_{12}F_{17}O_1{}^{35}Cl_1$ (M^+) 602.0300, found 602.0306.

Dimethyl 3-methyl-9-oxo-7-(4-(4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,11-heptadecafluoroundecyloxy)benzyl)-2,4-di(pyridin-2-yl)-3,7-diazabicyclo[3.3.1]nonane-1,5-dicarboxylate (FL1). Brown foamed solid. Mp: 74-75 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.43 (d, *J* = 4.0 Hz, 2H), 7.87 (d, *J* = 7.9 Hz, 2H), 7.49 (td, *J* = 7.7, 1.6 Hz, 2H), 7.26 (d, *J* = 8.5 Hz, 2H), 7.12 (dd, *J* = 6.9, 5.3 Hz, 2H), 6.91 (d, *J* = 8.5 Hz, 2H), 4.68 (s, 2H), 4.08 (t, *J* = 5.9 Hz, 2H), 3.80 (s, 6H), 3.32 (s, 2H), 3.04 (d, *J* = 12.5 Hz, 2H), 2.52 (d, *J* = 12.2 Hz, 2H), 2.41 – 2.31 (m, 2H), 2.16 (dd, *J* = 9.6, 5.7 Hz, 2H), 1.96 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 203.6, 168.5, 158.5, 158.1, 149.1, 136.0, 131.5, 129.4, 123.4, 122.8, 114.2, 73.7, 66.4, 62.1, 61.5, 58.7, 52.4, 43.2, 27.8 (t, *J* = 22.1 Hz), 20.6. HRMS (ESI): calcd for $C_{40}H_{36}F_{17}N_4O_6$ $(M+H)^+$ 991.2358, found 991.2351.

Dimethyl 3-methyl-9-oxo-7-(3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyl)-2,4-di(pyridin-2-yl)-3,7-diazabicyclo[3.3.1]nonane-1,5-dicarboxylate (FL2). White solid. Mp: 176-177 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.52 (d, *J* = 4.6 Hz, 2H), 7.96 (d, *J* = 7.8 Hz, 2H), 7.75 (t, *J* = 7.6 Hz, 2H), 7.24 – 7.18 (m, 2H), 4.75 (s, 2H), 3.83 (s, 6H), 3.03 (d, *J* = 11.7 Hz, 2H), 2.66 (dd, *J* = 15.8, 9.3 Hz, 4H), 2.27 (td, *J* = 18.4, 9.1 Hz, 2H), 2.06 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 202.9, 168.5, 158.6, 149.5, 136.2, 123.3, 123.1, 73.5, 62.2, 58.3, 52.6, 48.1, 43.2, 28.3 (t, *J* = 22.3 Hz). HRMS (ESI): calcd for $C_{32}H_{28}F_{17}N_4O_5$ $(M+H)^+$ 871.1783, found 871.1791.

9-Methoxy-1,5-diphenyl-3,7-bis(pyridin-2-ylmethyl)-3,7-diazabicyclo[3.3.1]nonane (L3). Brown foamed solid. Mp: 126-127 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.40 (d, *J* = 4.8 Hz, 1H), 8.31 (d, *J* = 4.8 Hz, 1H), 7.69 (ddd, *J* = 14.0, 10.1, 4.6 Hz, 2H), 7.63 – 7.56 (m, 5H), 7.48 (d, *J* = 7.8 Hz, 1H), 7.31 (t, *J* = 7.8 Hz, 4H), 7.23 – 7.18 (m, 3H), 7.14 (dd, *J* = 7.3, 5.0 Hz, 1H), 4.61 (s, 2H), 4.54 (d, *J* = 11.7 Hz, 2H), 4.40 (s, 1H), 4.35 (s, 2H), 3.67 – 3.59 (m, 4H), 3.48 (d, *J* = 12.4 Hz, 2H), 2.45 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 154.3, 151.7, 149.3, 149.2, 138.7, 137.1, 137.1, 128.7, 127.5, 126.1, 124.0, 123.9, 123.2, 122.9, 83.6, 62.0, 61.2, 61.2, 59.3, 55.2, 43.1. HRMS (ESI): calcd for $C_{32}H_{35}N_4O$ $(M+H)^+$ 491.2805, found 491.2810.

1,5-Diphenyl-9-(3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyloxy)-3,7-bis(pyridin-2-ylmethyl)-3,7-diazabicyclo[3.3.1]nonane (FL3). Brown foamed solid. Mp: 102-103 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.41 (d, *J* = 4.6 Hz, 1H), 8.31 (d, *J* = 4.7 Hz, 1H), 7.71 (td, *J* = 7.7, 1.6 Hz, 1H), 7.64 (d, *J* = 7.9 Hz, 6H), 7.46 (d, *J* = 7.8 Hz, 1H), 7.32 (t, *J* = 7.8 Hz, 4H), 7.20 (dd, *J* = 15.2, 7.6 Hz, 3H), 7.15 (dd, *J* = 7.0, 5.3 Hz, 1H), 4.84 (d, *J* = 11.3 Hz, 2H), 4.72 (s, 2H), 4.63 (s, 1H), 4.55 (t, *J* = 6.5 Hz, 1H), 4.48 (t, *J* = 6.5 Hz, 1H), 4.30 (s, 2H), 3.63 (d, *J* = 9.0 Hz, 2H), 3.54 (dd, *J* = 11.6, 5.4 Hz, 4H), 3.28 (d, *J* = 4.1 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 154.8, 151.2, 149.3, 149.3, 138.5, 137.2, 137.1, 128.9, 127.8, 126.1, 123.9, 123.8, 123.3, 122.9, 84.5, 65.3, 65.3, 62.0, 60.9, 59.1, 55.2, 43.3. HRMS (ESI): calcd for $C_{41}H_{36}F_{17}N_4O$ $(M+H)^+$ 923.2618, found 923.2620.

4-Methoxybenzaldehyde (7a). Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.88 (s, 1H), 7.84 (d, *J* = 8.8 Hz, 2H), 7.00 (d, *J* = 8.7 Hz, 2H), 3.89 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.8, 164.6, 132.0, 130.0, 114.3, 55.6. HRMS (EI): calcd for $C_8H_8O_2$ (M^+) 136.0519, found 136.0515.

Benzaldehyde (7b). Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.02 (s, 1H), 7.88 (dd, *J* = 8.1, 1.5 Hz, 2H), 7.62 (tt, *J* = 6.9, 1.4 Hz, 1H), 7.53 (t, *J* = 7.6 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 192.3, 136.4, 134.4, 129.7, 128.9. HRMS (EI): calcd for C_7H_6O (M^+) 106.0419, found 106.0415.

4-(Allyloxy)benzaldehyde (7c). Colorless oil. ^{1}H NMR (300 MHz, $CDCl_3$) δ 9.86 (s, 1H), 7.84 – 7.77 (m, 2H), 7.03 – 6.95 (m, 2H), 6.03 (ddt, *J* = 17.2, 10.5, 5.3 Hz, 1H), 5.36 (ddq, *J* = 27.3, 10.5, 1.5 Hz, 2H), 4.60 (dt, *J* = 5.3, 1.5 Hz, 2H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 190.7, 163.5, 132.2, 131.9, 130.0, 118.2, 114.9, 68.9. HRMS (EI): calcd for $C_{10}H_{10}O_2$ (M^+) 162.0675, found 162.0682.

4-Nitrobenzaldehyde (7d). Yellow solid. Mp: 106-108 ºC (lit.[1] mp 105-108 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.16 (s, 1H), 8.39 (d, *J* = 8.5 Hz, 2H), 8.08 (d, *J* = 8.3 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.2, 151.1, 140.0, 130.5, 124.3. HRMS (EI): calcd for $C_7H_5O_3N$ (M^+) 151.0264, found 151.0270.

3,4-Dimethoxybenzaldehyde (7e). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.81 (s, 1H), 7.41 (dd, *J* = 8.2, 1.8 Hz, 1H), 7.36 (d, *J* = 1.8 Hz, 1H), 6.94 (d, *J* = 8.2 Hz, 1H), 3.92 (s, 3H), 3.89 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.7, 154.4, 149.5, 130.0, 126.7, 110.3, 108.9, 56.0, 55.8. HRMS (EI): calcd for $C_9H_{10}O_3$ (M^+) 166.0625, found 166.0631.

2-Naphthaldehyde (7f). Pale-yellow solid. Mp: 61-62 ºC (lit.[2] mp 60-62 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.28 (s, 1H), 8.45 (s, 1H), 8.07 (ddd, *J* = 20.7, 16.2, 8.0 Hz, 4H), 7.74 (dt, *J* = 27.8, 7.1 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.2, 134.3, 132.4, 132.0, 130.5, 127.5, 127.0, 127.0, 126.0, 125.0, 120.6. HRMS (EI): calcd for $C_{11}H_8O$ (M^+) 156.0570, found 156.0574.

4-*tert*-Butylbenzaldehyde (7g). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.06 (s, 1H), 7.90 (d, *J* = 8.0 Hz, 2H), 7.63 (d, *J* = 8.0 Hz, 2H), 1.44 (s, 9H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.5, 156.9, 132.6, 128.2, 124.5, 33.8, 29.6. HRMS (EI): calcd for $C_{11}H_{14}O$ (M^+) 162.1039, found 162.1044.

***N*-(4-Formylphenyl)acetamide (7h).** Off-white solid. Mp: 150-151 ºC (lit.[3] mp 149-151 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.90 (s, 1H), 8.27 (s, 1H), 7.82 (d, *J* = 8.6 Hz, 2H), 7.72 (d, *J* = 8.2 Hz, 2H), 2.22 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 191.2, 169.1, 143.8, 132.1, 131.1, 119.3, 24.7. HRMS (EI): calcd for $C_9H_9O_2N$ (M^+) 163.0628, found 163.0633.

4-(Methylthio)benzaldehyde (7i). Pale-yellow oil. ^{1}H NMR (300 MHz, $CDCl_3$) δ 9.86 (s, 1H), 7.74 – 7.67 (m, 2H), 7.29 – 7.23 (m, 2H), 2.47 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 191.0, 147.7, 132.7, 129.8, 125.0, 14.4. HRMS (EI): calcd for C_8H_8OS (M^+) 152.0290, found 152.0288.

4-Hydroxybenzaldehyde (7j). Off-white solid. Mp: 109-111 ºC (lit.[4] mp 110-112 ºC). ^{1}H NMR (500 MHz, MeOD) δ 9.76 (s, 1H), 7.77 (d, *J* = 8.6 Hz, 2H), 6.91 (d, *J* = 8.6 Hz, 2H), 4.91 (s, 1H). ^{13}C NMR (126 MHz, MeOD) δ 192.8, 165.1, 133.4, 130.3, 116.9. HRMS (EI): calcd for $C_7H_6O_2$ (M^+) 122.0362, found 122.0361.

4-Formylbenzoic acid (7k). Off-white solid. Mp: 247-248 ºC (lit.[3] mp 245-247 ºC). ^{1}H NMR (500 MHz, DMSO) δ 13.39 (s, 1H), 10.11 (s, 1H), 8.13 (d, *J* = 7.7 Hz, 2H), 8.02 (d, *J* = 8.0 Hz, 2H). ^{13}C NMR (126 MHz, DMSO) δ 193.0, 166.6, 138.9, 135.6, 129.9, 129.5. HRMS (EI): calcd for $C_8H_6O_3$ (M^+) 150.0312, found 150.0307.

4-Chlorobenzaldehyde (7l). Off-white solid. Mp: 42-44 ºC (lit.[2] mp 43-45 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.97 (s, 1H), 7.80 (d, *J* = 7.8 Hz, 2H), 7.49 (d, *J* = 7.8 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.8, 140.9, 134.7, 130.8, 129.4. HRMS (EI): calcd for $C_7H_4O^{35}Cl$ (M^+) 138.9945, found 138.9951.

2-Chlorobenzaldehyde (7m). Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.39 (s, 1H), 7.83 (dd, *J* = 7.7, 1.7 Hz, 1H), 7.46 (td, *J* = 7.7, 1.8 Hz, 1H), 7.37 (d, *J* = 8.0 Hz, 1H), 7.31 (t, *J* = 7.9 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 189.5, 137.7, 134.9, 132.2, 130.4, 129.1, 127.1. HRMS (EI): calcd for $C_7H_4O^{35}Cl$ (M^+) 138.9945, found 138.9950.

2-Bromobenzaldehyde (7n). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.35 (s, 1H), 7.91 – 7.88 (m, 1H), 7.63 (dd, *J* = 7.4, 1.7 Hz, 1H), 7.45 – 7.39 (m, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 191.7, 135.3, 133.8, 133.5, 129.8, 127.8, 127.0. HRMS (EI): calcd for $C_7H_4O^{79}Br$ (M^+) 182.9440, found 182.9445.

2-Iodobenzaldehyde (7o). White solid. Mp: 30-31 ºC (lit.[5] mp 30-31 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.07 (s, 1H), 7.96 (d, *J* = 7.9 Hz, 1H), 7.88 (dd, *J* = 7.7, 1.8 Hz, 1H), 7.47 (t, *J* = 7.5 Hz, 1H), 7.32 – 7.26 (m, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 195.7, 140.6, 135.4, 135.1, 130.2, 128.7, 100.7. HRMS (EI): calcd for $C_7H_5O^{127}I$ (M^+) 231.9380, found 231.9383.

2-Aminobenzaldehyde (7p). Off-white solid. Mp: 36-37 ºC (lit.[6] mp 35-36 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.88 (s, 1H), 7.48 (d, *J* = 7.8 Hz, 1H), 7.31 (dd, *J* = 8.3, 7.1 Hz, 1H), 6.75 (t, *J* = 7.4 Hz, 1H), 6.65 (d, *J* = 8.3 Hz, 1H), 6.10 (s, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 194.0, 149.8, 135.6, 135.1, 118.8, 116.3, 115.9. HRMS (EI): calcd for C_7H_7NO (M^+) 121.0522, found 121.0528.

Thiophene-2-carbaldehyde (7q). Brown oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.02 (s, 1H), 7.85 (dd, *J* = 12.1, 3.9 Hz, 2H), 7.29 (t, *J* = 4.0 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 181.5, 142.6, 134.9, 133.8, 126.9. HRMS (EI): calcd for $C_5H_4O^{32}S$ (M^+) 111.9977, found 111.9976.

1*H*-indole-3-carbaldehyde (7r). Yellow solid. Mp: 192-193 ºC (lit.[7] mp 190-192 ºC). ^{1}H NMR (500 MHz, DMSO) δ 12.13 (s, 1H), 9.94 (s, 1H), 8.28 (s, 1H), 8.10 (d, *J* = 7.6 Hz, 1H), 7.51 (d, *J* = 8.0 Hz, 1H), 7.24 (dt, *J* = 14.8, 7.0 Hz, 2H). ^{13}C NMR (126 MHz, DMSO) δ 184.9, 138.4, 137.0, 124.1, 123.4, 122.1, 120.8, 118.2, 112.4. HRMS (EI): calcd for C_9H_7NO (M^+) 145.0522, found 145.0526.

5-Methylfuran-2-carbaldehyde (7s). Pale-brown oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.43 (s, 1H), 7.11 (d, *J* = 3.5 Hz, 1H), 6.17 (d, *J* = 3.5 Hz, 1H), 2.35 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 176.7, 159.6, 151.7, 123.7, 109.3, 13.8. HRMS (EI): calcd for $C_6H_6O_2$ (M^+) 110.0362, found 110.0363.

Cinnamaldehyde (7t). Pale-yellow oil. ^{1}H NMR (300 MHz, $CDCl_3$) δ 9.68 (d, *J* = 7.7 Hz, 1H), 7.58 – 7.50 (m, 2H), 7.50 – 7.37 (m, 4H), 6.70 (dd, *J* = 16.0, 7.7 Hz, 1H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 193.5, 152.6, 133.9, 131.1, 129.0, 128.4, 128.4. HRMS (EI): calcd for C_9H_8O (M^+) 132.0570, found 132.0572.

Geranial (7u). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.05 (d, *J* = 7.8 Hz, 1H), 5.94 (d, *J* = 7.6 Hz, 1H), 5.14 (t, *J* = 5.5 Hz, 1H), 2.31-2.26 (m, 4H), 2.23 (s, 3H), 1.75 (s, 3H), 1.68 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 189.5, 161.9, 131.2, 125.8, 120.9, 39.0, 24.1, 24.0, 16.0, 16.0. HRMS (EI): calcd for $C_{10}H_{16}O$ (M^+) 152.1196, found 152.1194.

Acetophenone (9a). Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.94 (dd, *J* = 8.3, 1.2 Hz, 2H), 7.54 (t, *J* = 7.4 Hz, 1H), 7.44 (t, *J* = 7.7 Hz, 2H), 2.58 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 198.0, 137.0, 133.0, 128.4, 128.2, 26.5. HRMS (EI): calcd for C_8H_8O 120.0575, found 120.0580.

1-(4-Iodophenyl)ethanone (9b). White solid. Mp: 82-83 ºC (lit.[8] mp 83-84 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.86 – 7.80 (m, 2H), 7.68 – 7.63 (m, 2H), 2.57 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 197.3, 137.9, 136.4, 129.7, 101.0, 26.4. HRMS (EI): calcd for $C_8H_7O^{127}I$ (M^+) 245.9536, found 245.9542.

3-Acetylpyridine (9c). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.09 (d, *J* = 1.8 Hz, 1H), 8.70 (dd, *J* = 4.8, 1.7 Hz, 1H), 8.15 (dt, *J* = 8.0, 2.0 Hz, 1H), 7.35 (dd, *J* = 7.9, 4.8 Hz, 1H), 2.57 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 196.5, 153.4, 149.8, 135.3, 132.1, 123.5, 26.5. HRMS (EI): calcd for C_7H_7NO (M^+) 121.0522, found 121.0527.

2-Acetylthiophene (9d). Dark-yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.68 (d, *J* = 3.7 Hz, 1H), 7.61 (d, *J* = 5.0 Hz, 1H), 7.12 – 7.09 (m, 1H), 2.54 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 190.6, 144.5, 133.7, 132.4, 128.0, 26.8. HRMS (EI): calcd for $C_6H_6O^{32}S$ (M^+) 126.0134, found 126.0136

4-(1-Hydroxyethyl)benzaldehyde (7v). Pale-yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.94 (s, 1H), 7.82 (d, *J* = 8.2 Hz, 2H), 7.51 (d, *J* = 8.1 Hz, 2H), 4.95 (q, *J* = 6.5 Hz, 1H), 2.59 (s, 1H), 1.49 (d, *J* = 6.5 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 192.0, 152.8, 135.4, 129.9, 125.8, 69.7, 25.2. HRMS (EI): calcd for $C_9H_{10}O_2$ (M^+) 150.0675, found 150.0676.

4-Acetylbenzaldehyde (7v'). Pale-yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 10.12 (s, 1H), 8.11 (d, *J* = 7.9 Hz, 2H), 7.99 (d, *J* = 7.1 Hz, 2H), 2.67 (s, 3H). HRMS (EI): calcd for $C_9H_8O_2$ (M^+) 148.0519, found 148.0524.

4-(3-Hydroxypropyl)benzaldehyde (7w). Pale-yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.93 (s, 1H), 7.77 (d, *J* = 8.1 Hz, 2H), 7.34 (d, *J* = 8.0 Hz, 2H), 3.66 (t, *J* = 6.3 Hz, 2H), 2.80 – 2.75 (m, 2H), 2.12 (s, 1H), 1.90 (tt, *J* = 12.9, 6.4 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 192.1, 149.5, 134.4, 129.9, 129.1, 61.7, 33.6, 32.2. HRMS (EI): calcd for $C_{10}H_{12}O_2$ (M^+) 164.0832, found 164.0834.

4-(3-Oxopropyl)benzaldehyde (7w'). Pale-yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 9.98 (s, 1H), 9.83 (s, 1H), 7.82 (d, *J* = 8.0 Hz, 2H), 7.37 (d, *J* = 8.0 Hz, 2H), 3.04 (t, *J* = 7.4 Hz, 2H), 2.84 (t, *J* = 7.4 Hz, 2H). HRMS (EI): calcd for $C_{10}H_{10}O_2$ (M^+) 162.0675, found 162.0677.

3-Phenylcyclohex-2-enone (11a). Pale yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.53 (dd, *J* = 6.7, 3.1 Hz, 2H), 7.43 – 7.39 (m, 3H), 6.42 (s, 1H), 2.80 – 2.74 (m, 2H), 2.48 (d, *J* = 7.0 Hz, 2H), 2.15 (dd, *J* = 12.8, 6.4 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 199.8, 159.8, 138.8, 129.9, 128.7, 126.1, 125.4, 37.2, 28.1, 22.8. HRMS (EI): calcd for $C_{12}H_{12}O$ 172.0888, found 172.0895.

3-Methylcyclohex-2-enone (11b). Pale yellow oil. ^{1}H NMR (300 MHz, $CDCl_3$) δ 5.88 (d, *J* = 1.2 Hz, 1H), 2.38 – 2.31 (m, 2H), 2.28 (t, *J* = 6.2 Hz, 2H), 2.04 – 1.97 (m, 2H), 1.96 (s, 3H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 199.8, 162.8, 126.7, 37.0, 30.9, 24.4, 22.5. HRMS (EI): calcd for $C_7H_{10}O$ 110.0732, found 110.0730.

3-Oxocyclohex-1-enecarbonitrile (11c). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 6.51 (s, 1H), 2.56 (td, *J* = 6.0, 1.9 Hz, 2H), 2.52 – 2.48 (m, 2H), 2.13 (td, *J* = 12.3, 6.1 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 196.3, 138.6, 130.9, 116.9, 37.2, 27.6, 22.0. HRMS (EI): calcd for C_7H_7NO 121.0528, found 121.0528.

3-Acetylcyclohex-2-enone (11d). Pale yellow solid. Mp: 46-47 ºC (lit.[9] mp 48-49 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 6.58 (t, *J* = 1.7 Hz, 1H), 2.51 (td, *J* = 6.1, 1.7 Hz, 2H), 2.49 – 2.45 (m, 2H), 2.40 (s, 3H), 2.03 (dt, *J* = 12.4, 6.1 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 201.4, 200.0, 154.6, 132.4, 37.9, 26.1, 23.4, 21.9. HRMS (EI): calcd for $C_8H_{10}O_2$ (M^+) 138.0675, found 138.0676.

3-Acetylcyclopent-2-enone (11e). Pale yellow solid. Mp: 58-59 ºC (lit.[10] mp 57.5-58.5 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 6.64 (t, *J* = 2.0 Hz, 1H), 2.82 – 2.78 (m, 2H), 2.54 – 2.51 (m, 2H), 2.49 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 210.1, 197.1, 169.1, 137.0, 35.4, 27.7, 26.3. HRMS (EI): calcd for $C_7H_8O_2$ (M^+) 124.0519, found 124.0521.

1-Phenylhexan-1-one (11f). Pale yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.96 (dd, *J* = 8.3, 1.2 Hz, 2H), 7.55 (d, *J* = 7.3 Hz, 2H), 7.45 (t, *J* = 7.7 Hz, 2H), 2.99 – 2.92 (m, 2H), 1.74 (dq, *J* = 14.8, 7.4 Hz, 2H), 1.37 (td, *J* = 7.2, 3.6 Hz, 4H), 0.91 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 200.5, 137.1, 132.8, 128.5, 128.0, 38.5, 31.5, 24.0, 22.5, 13.9. HRMS (EI): calcd for $C_{12}H_{16}O$ 176.1201, found 176.1206.

2,3-Dihydro-1*H*-inden-1-one (11g). Brown solid. Mp: 41-42 ºC (lit.[11] mp 40-42 ºC). ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.76 (d, *J* = 7.7 Hz, 1H), 7.61 – 7.53 (m, 1H), 7.48 (d, *J* = 7.7 Hz, 1H), 7.37 (t, *J* = 7.4 Hz, 1H), 3.18 – 3.11 (m, 2H), 2.73 – 2.67 (m, 2H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 207.1, 155.2, 137.1, 134.6, 127.3, 126.7, 123.7, 36.2, 25.8. HRMS (EI): calcd for C_9H_8O 132.0575, found 132.0570.

9*H*-Fluoren-9-one (11h). Yellow solid. Mp: 84-85 ºC (lit.[11] mp 84-86 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.65 (d, *J* = 7.4 Hz, 2H), 7.48 (ddd, *J* = 10.0, 8.5, 4.2 Hz, 4H), 7.28 (td, *J* = 7.3, 1.3 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 193.9, 144.4, 134.6, 134.1, 129.0, 124.3, 120.3. HRMS (EI): calcd for $C_{13}H_8O$ 180.0575, found 180.0575.

Methyl 2-oxo-2-phenylacetate (11i). Yellow oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.02 (dd, *J* = 8.3, 1.1 Hz, 2H), 7.67 (t, *J* = 7.5 Hz, 1H), 7.52 (t, *J* = 7.9 Hz, 2H), 3.98 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.0, 164.0, 135.0, 132.4, 130.1, 128.9, 52.8. HRMS (EI): calcd for $C_9H_8O_3$ 164.0473, found 164.0481.

Isochroman-1-one (11j). Brown oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.06 (d, *J* = 7.8 Hz, 1H), 7.52 (td, *J* = 7.5, 1.4 Hz, 1H), 7.37 (t, *J* = 7.5 Hz, 1H), 7.25 (d, *J* = 7.7 Hz, 1H), 4.53 – 4.49 (m, 2H), 3.04 (t, *J* = 6.0 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 165.0, 139.5, 133.6, 130.2, 127.6, 127.2, 125.2, 67.2, 27.7. HRMS (EI): calcd for $C_9H_8O_2$ 148.0524, found 148.0523.

1-Tosyl-2,3-dihydroquinolin-4(1*H*)-one (11k). Brown solid. Mp: 140-141 ºC (lit.[12] mp 141 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.94 (dd, *J* = 7.8, 1.3 Hz, 1H), 7.86 (d, *J* = 8.4 Hz, 1H), 7.59 – 7.53 (m, 3H), 7.26 (dd, *J* = 8.2, 7.0 Hz, 1H), 7.22 (d, *J* = 8.2 Hz, 2H), 4.23 (t, *J* = 6.5 Hz, 2H), 2.40 – 2.36 (m, 5H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 192.7, 144.5, 142.3, 136.8, 134.7, 130.1, 127.7, 126.8, 125.7, 125.6, 124.5, 46.2, 36.5, 21.6. HRMS (EI): calcd for $C_{16}H_{15}NO_3S$ 301.0773, found 301.0772.

***N*-(*tert*-Butyl)benzamide (11l).** Brown solid. Mp: 133-134 ºC (lit.[13] mp 134-136 ºC). ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.74 – 7.68 (m, 2H), 7.50 – 7.36 (m, 3H), 5.95 (s, 1H), 1.47 (s, 9H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 166.9, 135.9, 131.0, 128.4, 126.7, 51.6, 28.8. HRMS (EI): calcd for $C_{11}H_{15}NO$ (M^+) 177.1148, found 177.1155.

3-*tert*-Butylperoxy-1-phenylcyclohexene (11a'). Pale yellow oil. ^{1}H NMR (300 MHz, $CDCl_3$) δ 7.66 – 7.60 (m, 2H), 7.44 – 7.39 (m, 3H), 6.42 (s, 1H), 5.11 (t, *J* = 2.8 Hz, 1H), 2.89 – 2.66 (m, 2H), 2.47 (dd, *J* = 5.6, 1.8 Hz, 1H), 2.42 (d, *J* = 4.8 Hz, 1H), 2.17 – 2.03 (m, 2H), 1.21 (s, 9H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 163.2, 146.4, 130.0, 128.7, 128.4, 126.8, 80.2, 75.6, 32.4, 26.5, 26.1, 25.7.

2-Benzylidenemalononitrile (13a). White solid. Mp: 81-82 ºC (lit.[14] mp 82-83 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.91 (d, *J* = 7.6 Hz, 2H), 7.79 (s, 1H), 7.64 (t, *J* = 7.5 Hz, 1H), 7.54 (t, *J* = 7.8 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 159.9, 134.5, 130.8, 130.6, 129.5, 113.6, 112.5, 82.7. HRMS (EI): calcd for $C_{10}H_6N_2$ (M^+) 154.0526, found 154.0532.

2-(4-Methoxybenzylidene)malononitrile (13b). Yellow solid. Mp: 112-113 ºC (lit.[15] mp 114 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.90 (d, *J* = 8.9 Hz, 2H), 7.64 (s, 1H), 7.00 (d, *J* = 9.0 Hz, 2H), 3.91 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 164.7, 158.8, 133.4, 123.9, 115.0, 114.4, 113.3, 78.3, 55.7. HRMS (EI): calcd for $C_{11}H_8N_2O$ (M^+) 184.0631, found 184.0636.

2-(4-Nitrobenzylidene)malononitrile (13c). Pale yellow solid. Mp: 161-162 ºC (lit.[14] mp 160-161 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 (d, J = 8.8 Hz, 2H), 8.08 (d, J = 8.8 Hz, 2H), 7.90 (s, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 156.9, 150.3, 135.8, 131.3, 124.6, 112.6, 111.6, 87.5. HRMS (EI): calcd for $C_{10}H_5N_3O_2$ (M^+) 199.0376, found 199.0381.

2-(4-Chlorobenzylidene)malononitrile (13d). White solid. Mp: 161-162 ºC (lit.[14] mp 161-162 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.85 (d, *J* = 8.6 Hz, 2H), 7.74 (s, 1H), 7.52 (d, *J* = 8.6 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 158.3, 141.1, 131.8, 130.0, 129.2, 113.4, 112.3, 83.3. HRMS (EI): calcd for $C_{10}H_5N_2{}^{35}Cl$ (M^+) 188.0136, found 188.0142.

2-(4-Ethylbenzylidene)malononitrile (13e). White solid. Mp: 131-132 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.84 (d, *J* = 8.3 Hz, 2H), 7.73 (s, 1H), 7.36 (d, *J* = 8.3 Hz, 2H), 2.75 (q, *J* = 7.6 Hz, 2H), 1.27 (t, *J* = 7.6 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 159.9, 152.5, 131.1, 129.2, 128.7, 114.1, 112.9, 81.2, 29.2, 14.9. HRMS (EI): calcd for $C_{12}H_{10}N_2$ (M^+) 182.0839, found 182.0844.

2-(Benzo[d][1,3]dioxol-5-ylmethylene)malononitrile (13f). Yellow solid. Mp: 197-198 ºC (lit.[16] mp 196-197 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.60 (s, 2H), 7.32 (d, *J* = 8.2 Hz, 1H), 6.93 (d, *J* = 8.2 Hz, 1H), 6.13 (s, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 158.7, 153.5, 149.0, 130.0, 125.6, 114.2, 113.1, 109.1, 108.2, 102.7, 79.2. HRMS (EI): calcd for $C_{11}H_6N_2O_2$ (M^+) 198.0424, found 198.0421.

2-(4-Hydroxybenzylidene)malononitrile (13g). Yellow solid. Mp: 185-186 ºC (lit.[14] mp 186-188 ºC). ^{1}H NMR (500 MHz, MeOD) δ 7.90 (s, 1H), 7.86 (d, *J* = 8.8 Hz, 2H), 6.90 (d, *J* = 8.8 Hz, 2H), 4.93 (s, 1H). ^{13}C NMR (126 MHz, MeOD) δ 165.3, 161.0, 135.0, 124.5, 117.5, 115.9, 114.9, 77.3. HRMS (EI): calcd for $C_{10}H_6N_2O$ (M^+) 170.0475, found 170.0481.

2-(2-Chlorobenzylidene)malononitrile (13h). White solid. Mp: 92-93 ºC (lit.[15] mp 94 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.26 (s, 1H), 8.17 (d, *J* = 7.7 Hz, 1H), 7.55 (d, *J* = 3.8 Hz, 2H), 7.48 – 7.42 (m, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 156.0, 136.2, 135.0, 130.6, 129.4, 129.0, 127.7, 113.1, 111.8, 85.7. HRMS (EI): calcd for $C_{10}H_5N_2{}^{35}Cl$ (M^+) 188.0136, found 188.0132.

2-(3-Cyanobenzylidene)malononitrile (13i). White solid. Mp: 150-151 ºC (lit.[16] mp 150 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.20 (d, *J* = 7.9 Hz, 1H), 8.08 (s, 1H), 7.90 (d, *J* = 7.8 Hz, 1H), 7.81 (s, 1H), 7.71 (t, *J* = 7.9 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 157.0, 136.8, 133.9, 133.5, 131.7, 130.7, 116.9, 114.4, 112.7, 111.6, 86.3. HRMS (EI): calcd for $C_{11}H_5N_3$ (M^+) 179.0478, found 179.0482.

2-((1*H*-Indol-3-yl)methylene)malononitrile (13j). Yellow solid. Mp: 222-223 ºC (lit.[17] mp 223-224 ºC). ^{1}H NMR (500 MHz, DMSO) δ 12.71 (s, 1H), 8.67 (s, 1H), 8.51 (s, 1H), 8.03 (d, *J* = 7.1 Hz, 1H), 7.57 (d, *J* = 7.4 Hz, 1H), 7.33 – 7.25 (m, 2H). ^{13}C NMR (126 MHz, DMSO) δ 152.5, 136.2, 133.3, 126.7, 123.9, 122.6, 119.0, 115.9, 115.9, 113.0, 111.0, 69.3. HRMS (EI): calcd for $C_{12}H_7N_3$ (M^+) 193.0635, found 193.0640.

2-(Thiophen-2-ylmethylene)malononitrile (13k). Yellow solid. Mp: 93-94 ºC (lit.[18] mp 91-92 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.88 (d, *J* = 3.4 Hz, 2H), 7.80 (d, *J* = 3.6 Hz, 1H), 7.29 – 7.25 (m, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 151.1, 138.2, 136.9, 135.3, 129.0, 113.7, 112.9, 78.1. HRMS (EI): calcd for $C_8H_4N_2{}^{32}S$ (M^+) 160.0090, found 160.0096.

2-(Furan-2-ylmethylene)malononitrile (13l). Yellow solid. Mp: 71-72 ºC (lit.[19] mp 72-73 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.80 (s, 1H), 7.51 (s, 1H), 7.35 (d, *J* = 3.2 Hz, 1H), 6.72 – 6.68 (m, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 149.5, 148.0, 143.0, 123.5, 114.4, 113.7, 112.5, 77.4. HRMS (EI): calcd for $C_8H_4N_2O$ (M^+) 144.0318, found 144.0322.

(*E*)-2-(3-Phenylallylidene)malononitrile (13m). Yellow solid. Mp: 121-122 ºC (lit.[20] mp 121-122 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.63 – 7.57 (m, 3H), 7.51 – 7.43 (m, 3H), 7.32 – 7.22 (m, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 160.0, 150.4, 133.9, 132.0, 129.3, 128.9, 122.2, 113.5, 111.6, 82.8. HRMS (EI): calcd for $C_{12}H_8N_2$ (M^+) 180.0682, found 180.0685.

2-(2,2-Dimethylpropylidene)malononitrile (13n). White solid. Mp: 63-64 ºC (lit.[21] mp 64-65 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.21 (s, 1H), 1.30 (s, 9H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 177.5, 113.0, 111.0, 86.7, 36.9, 28.4. HRMS (EI): calcd for $C_8H_{10}N_2$ (M^+) 134.0839, found 134.0835.

2-(1-Phenylethylidene)malononitrile (13o). White solid. Mp: 92-93 ºC (lit.[22] mp 92 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.55 (dd, *J* = 6.5, 2.4 Hz, 3H), 7.51 (d, *J* = 6.1 Hz, 2H), 2.64 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 175.4, 135.9, 132.2, 129.1, 127.3, 112.7, 112.7, 84.7, 24.2. HRMS (EI): calcd for $C_{11}H_8N_2$ (M^+) 168.0682, found 168.0687.

2-Cyclohexylidenemalononitrile (13p). Colorless oil. ^{1}H NMR (500 MHz, $CDCl_3$) δ 2.68 – 2.62 (m, 4H), 1.80 (dt, *J* = 12.3, 6.0 Hz, 4H), 1.68 (ddd, *J* = 7.8, 5.8, 4.0 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 184.9, 111.6, 82.5, 34.7, 27.9, 25.0. HRMS (EI): calcd for $C_9H_{10}N_2$ (M^+) 146.0839, found 146.0839.

Ethyl 2-cyano-3-phenylacrylate (13q). White solid. Mp: 45-46 ºC (lit.[20] mp 46-47 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.23 (s, 1H), 7.97 (d, *J* = 7.4 Hz, 2H), 7.56 – 7.52 (m, 1H), 7.48 (dd, *J* = 8.1, 6.8 Hz, 2H), 4.37 (q, *J* = 7.1 Hz, 2H), 1.38 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 162.4, 154.9, 133.2, 131.4, 131.0, 129.2, 115.4, 102.9, 62.6, 14.1. HRMS (EI): calcd for $C_{12}H_{11}NO_2$ (M^+) 201.0784, found 201.0788.

***N*-Butyl-2-cyano-3-phenylacrylamide (13r).** White solid. Mp: 78-79 ºC (lit.[23] mp 79.2-80.2 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.33 (s, 1H), 7.92 (d, *J* = 7.1 Hz, 2H), 7.55 – 7.45 (m, 3H), 6.39 (s, 1H), 3.43 (dd, *J* = 13.5, 6.6 Hz, 2H), 1.62 – 1.56 (m, 2H), 1.45 – 1.36 (m, 2H), 0.96 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 160.1, 152.8, 132.6, 131.8, 130.5, 129.2, 117.1, 104.1, 40.3, 31.4, 20.0, 13.7. HRMS (EI): calcd for $C_{14}H_{16}N_2O$ (M^+) 228.1257, found 228.1254.

2-Benzylidene-4,4-dimethyl-3-oxopentanenitrile (13s). White solid. Mp: 70-71 ºC (lit.[24] mp 69-72 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.21 (s, 1H), 7.98 (d, *J* = 7.5 Hz, 2H), 7.55 – 7.45 (m, 3H), 1.42 (s, 9H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 198.1, 156.1, 132.9, 132.0, 131.1, 129.1, 118.2, 107.3, 44.5, 26.4. HRMS (EI): calcd for $C_{14}H_{15}NO$ (M^+) 213.1148, found 213.1144.

2-(3-Nitrophenyl)-3-phenylacrylonitrile (13t). Pale yellow solid. Mp: 167-168 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.53 (t, *J* = 2.0 Hz, 1H), 8.25 (dd, *J* = 8.2, 2.1 Hz, 1H), 8.02 (dd, *J* = 7.0, 1.7 Hz, 1H), 7.94 (dd, *J* = 6.4, 3.0 Hz, 2H), 7.70 – 7.63 (m, 2H), 7.51 (dd, *J* = 5.0, 1.9 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 148.8, 144.7, 136.3, 132.9, 132.0, 131.5, 130.2, 129.6, 129.2, 123.7, 120.5, 117.2, 109.3. HRMS (EI): calcd for $C_{15}H_{10}N_2O_2$ (M^+) 250.0737, found 250.0743.

Ethyl 2-cyano-3-(4-hydroxy-3-methoxyphenyl)acrylate (13u). Yellow solid. Mp: 109-110 ºC (lit.[25] mp 108-109 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.12 (s, 1H), 7.83 (d, *J* = 2.0 Hz, 1H), 7.38 (dd, *J* = 8.3, 2.0 Hz, 1H), 6.98 (d, *J* = 8.3 Hz, 1H), 6.36 (s, 1H), 4.36 (q, *J* = 7.1 Hz, 2H), 3.96 (s, 3H), 1.38 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 163.1, 154.8, 150.9, 146.8, 128.7, 124.2, 116.4, 114.9, 111.2, 98.9, 62.4, 56.1, 14.1. HRMS (EI): calcd for $C_{13}H_{13}O_4N$ (M^+) 247.0845, found 247.0846.

2-(3,4-Dimethoxybenzylidene)malononitrile (13v). Yellow solid. Mp: 144-145 ºC (lit.[26] mp 142 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.64 (d, *J* = 3.4 Hz, 2H), 7.38 (dd, *J* = 8.5, 2.1 Hz, 1H), 6.95 (d, *J* = 8.5 Hz, 1H), 3.96 (s, 3H), 3.91 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 159.1, 154.8, 149.4, 128.1, 124.2, 114.4, 113.5, 111.1, 110.8, 78.2, 56.2, 56.0. HRMS (EI): calcd for $C_{12}H_{10}N_2O_2$ (M^+) 214.0737, found 214.0737.

2-(4-*tert*-butylbenzylidene)malononitrile (13w). White solid. Mp: 91-92 ºC. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.86 (d, *J* = 8.5 Hz, 2H), 7.74 (s, 1H), 7.55 (d, *J* = 8.5 Hz, 2H), 1.35 (s, 9H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 159.6, 159.2, 130.8, 128.4, 126.6, 114.0, 112.8, 81.3, 35.5, 30.8. HRMS (EI): calcd for $C_{14}H_{14}N_2$ (M^+) 210.1152, found 210.1158.

2-(Naphthalen-2-ylmethylene)malononitrile (13x). Yellow solid. Mp: 141-142 ºC (lit.[16] mp 140.2-140.8 ºC). ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.25 (s, 1H), 8.05 (dd, *J* = 8.7, 1.8 Hz, 1H), 7.94 (t, *J* = 7.2 Hz, 2H), 7.90 (d, *J* = 8.2 Hz, 1H), 7.86 (s, 1H), 7.68 (t, *J* = 7.5 Hz, 1H), 7.61 (t, *J* = 7.5 Hz, 1H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 159.6, 135.8, 134.4, 132.5, 129.9, 129.6, 129.6, 128.5, 128.0, 127.7, 124.1, 113.9, 112.8, 82.1. HRMS (EI): calcd for $C_{14}H_8N_2$ (M^+) 204.0682, found 204.0684.

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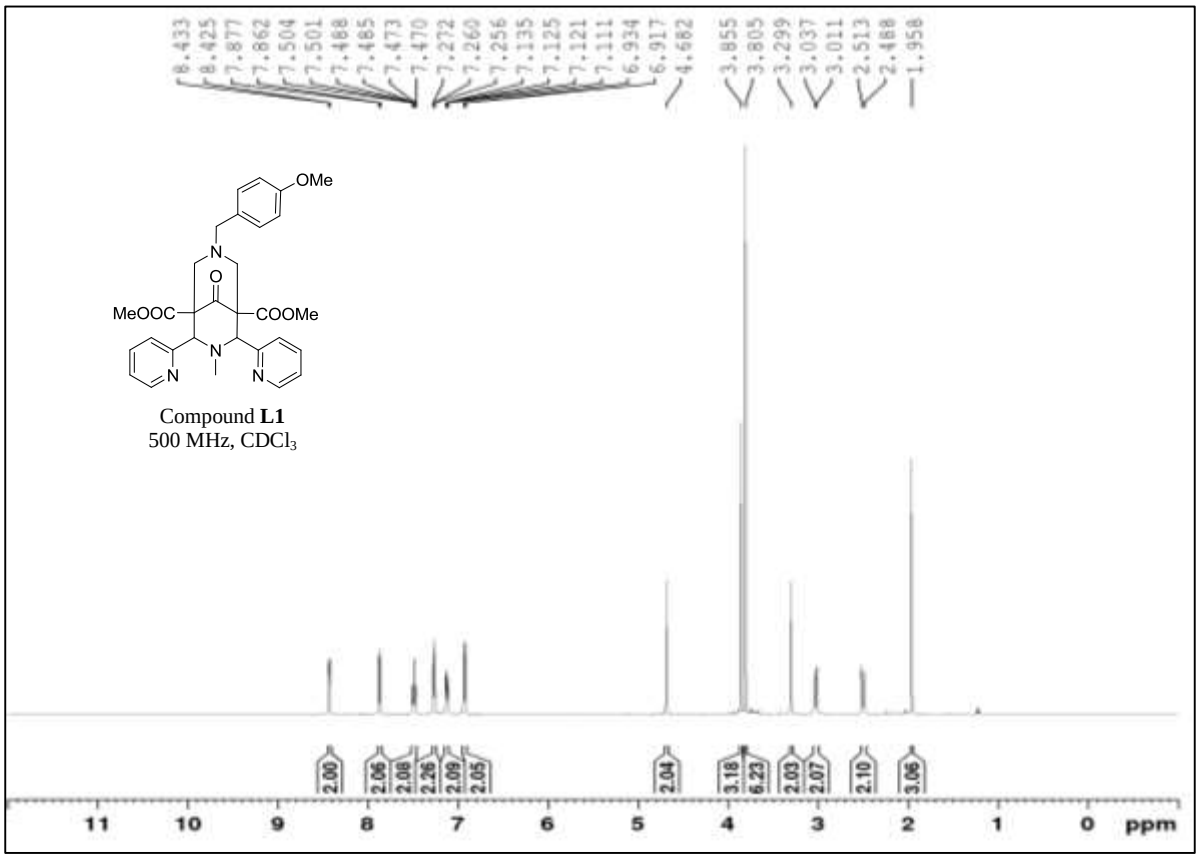
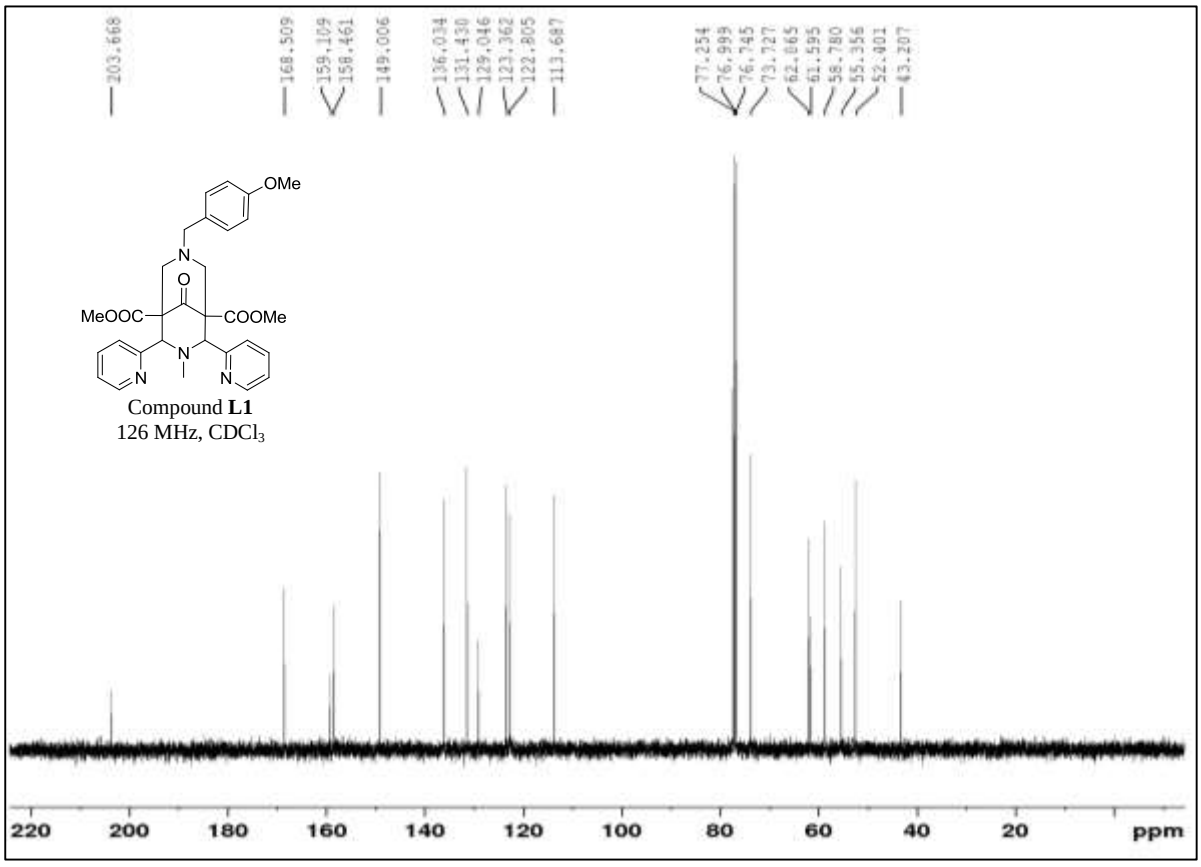
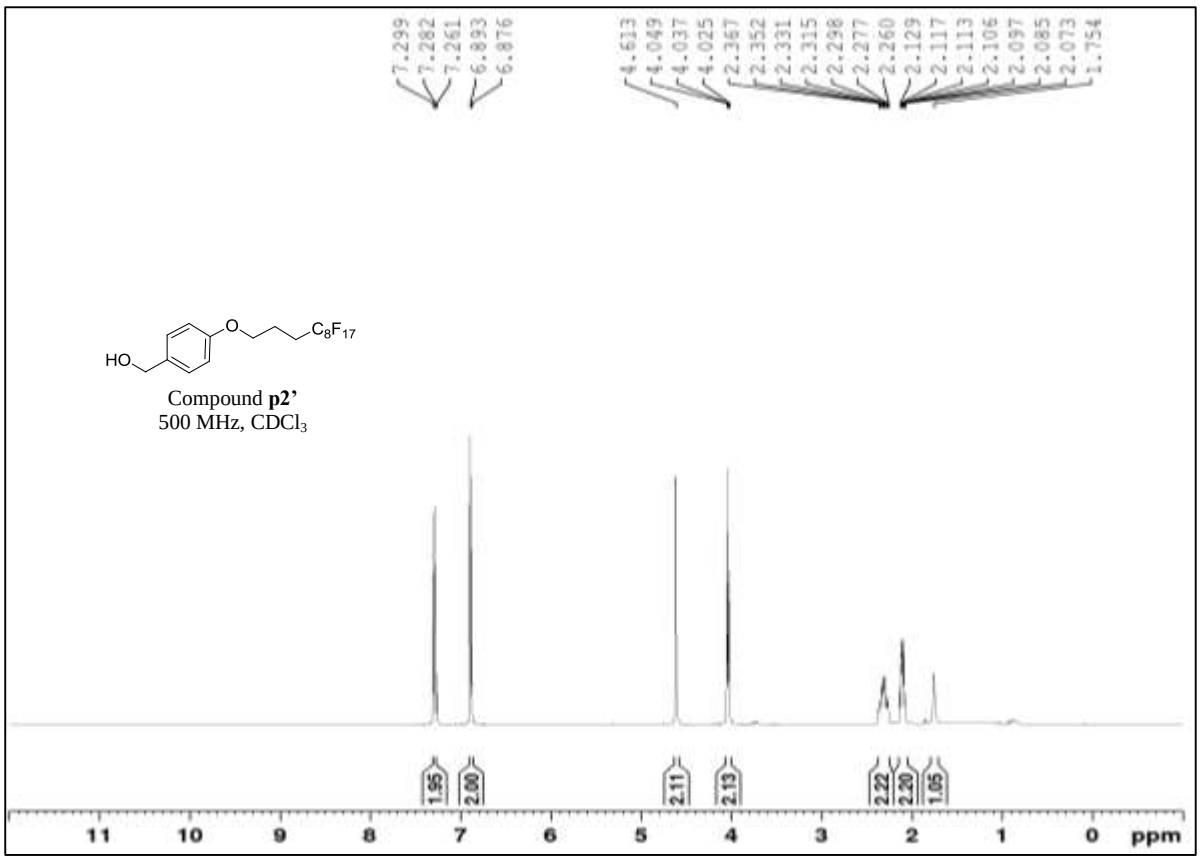
NMR spectra of new compounds:



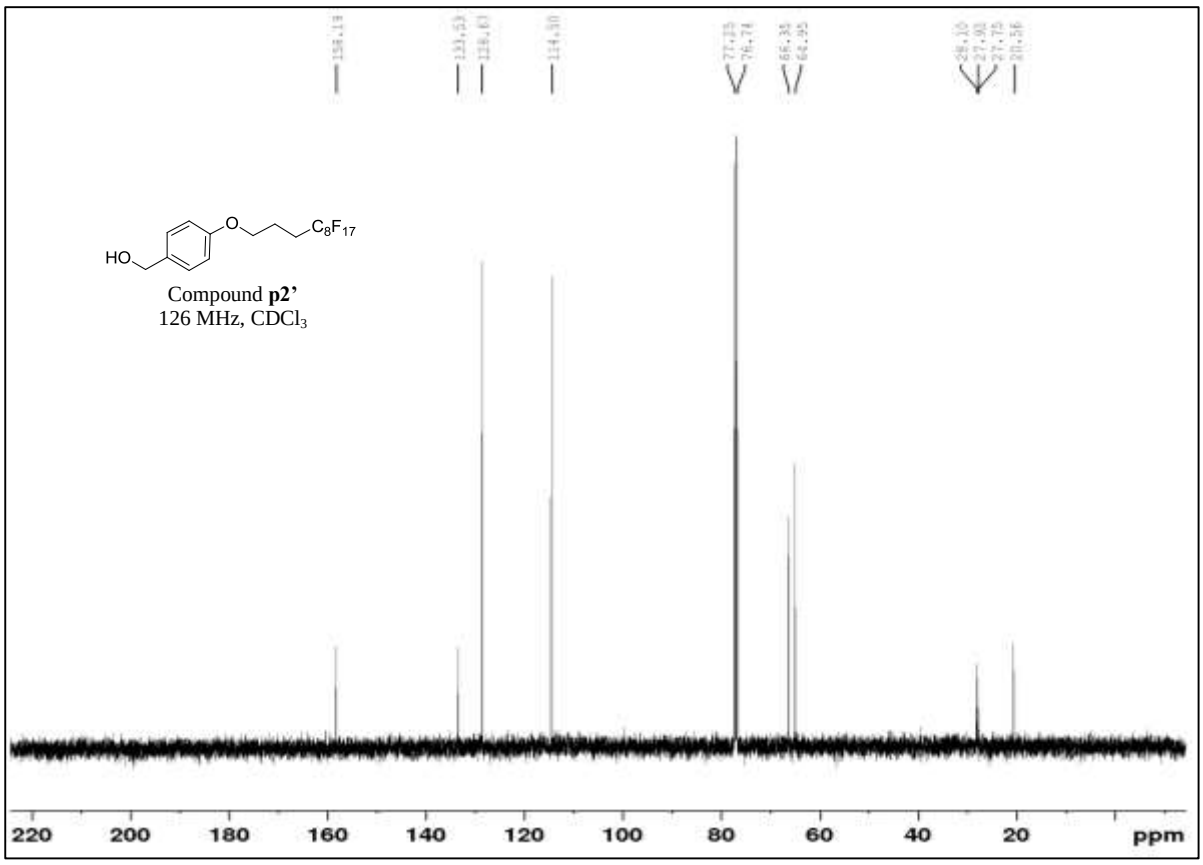
Compound **L1**
500 MHz, $CDCl_3$



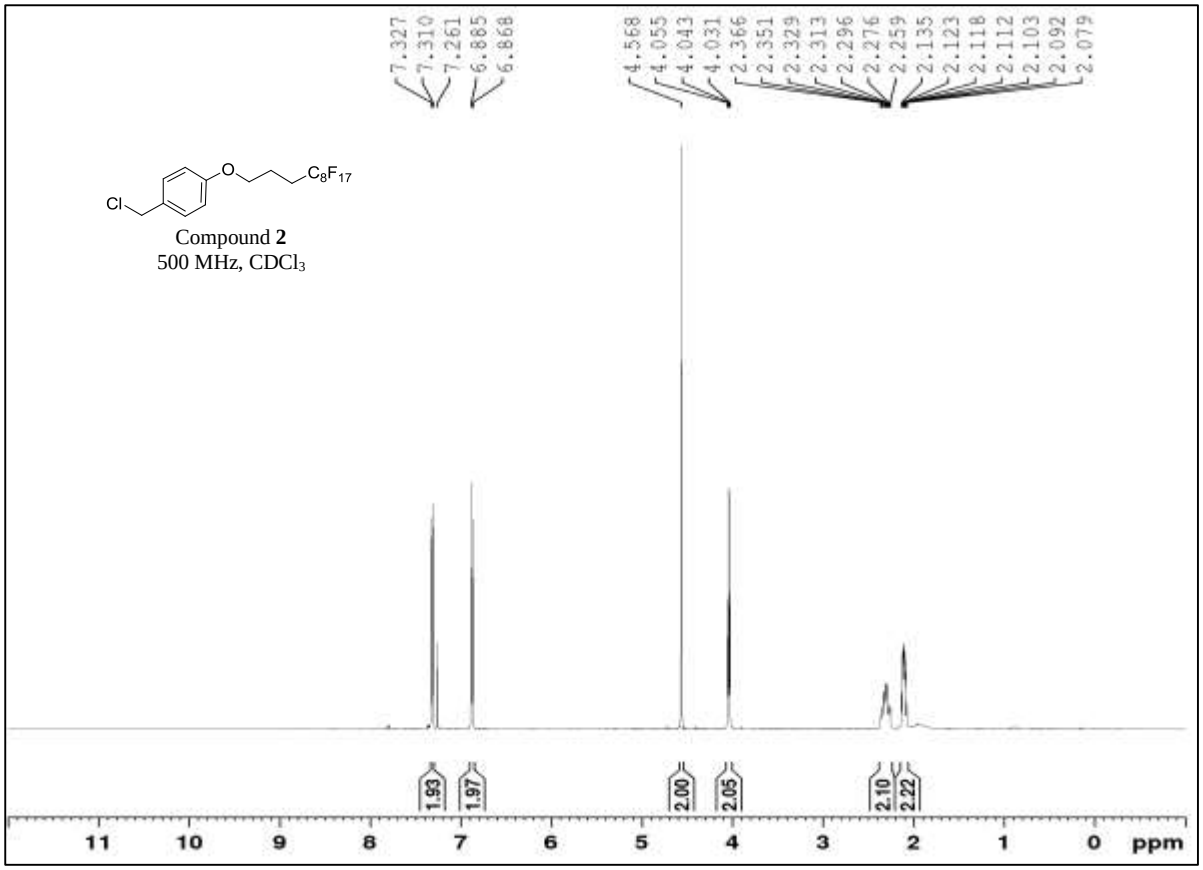
Compound **L1**
126 MHz, $CDCl_3$



Compound **p2'**
500 MHz, $CDCl_3$



Compound **p2'**
126 MHz, $CDCl_3$



Compound **2**
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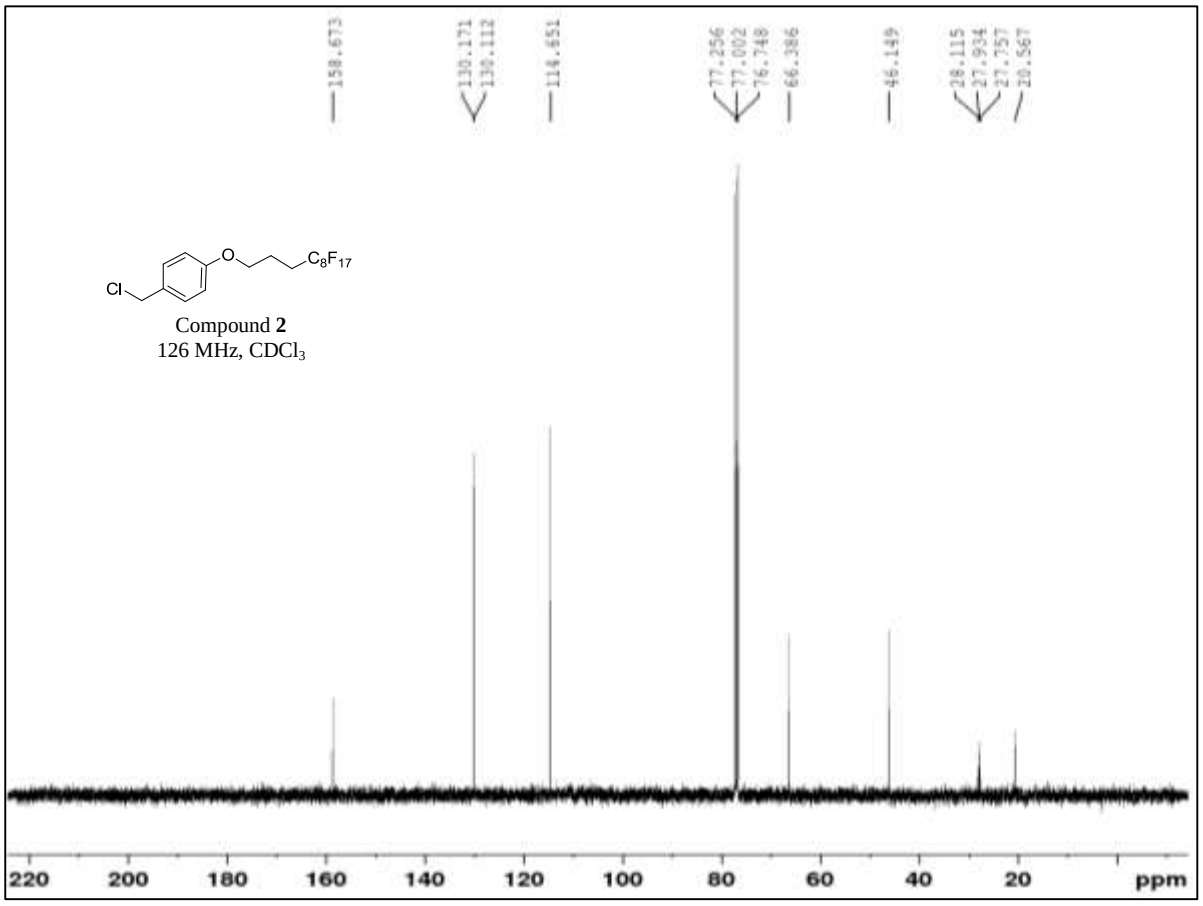

Compound **2**
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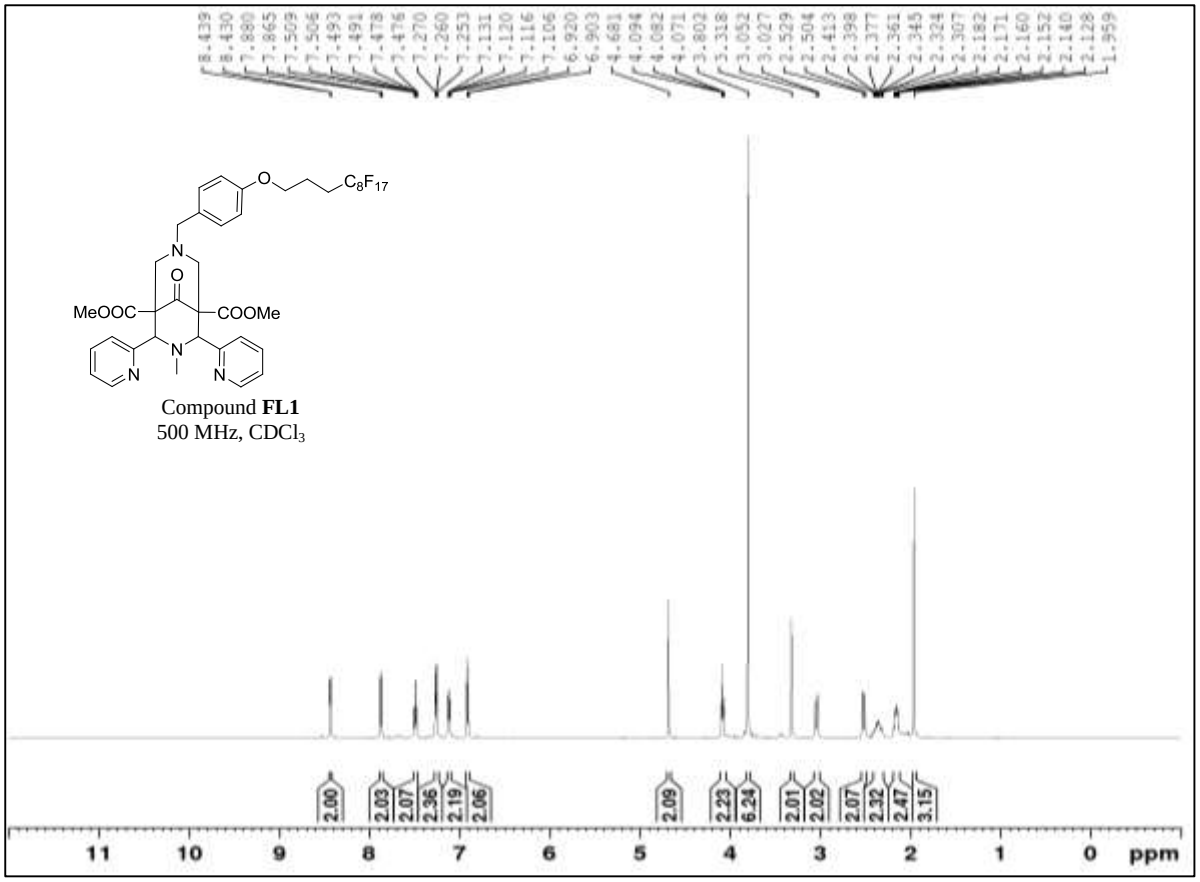

Compound **FL1**
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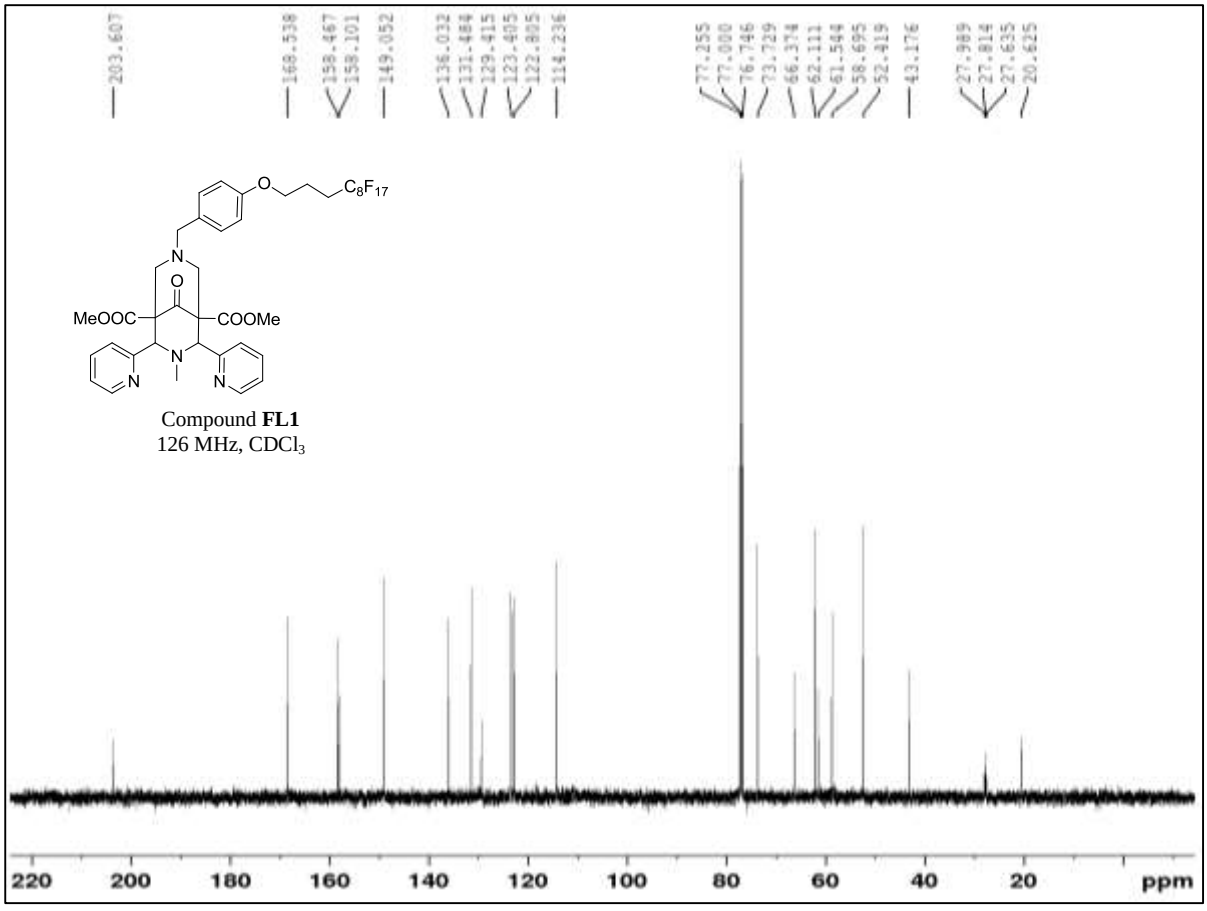
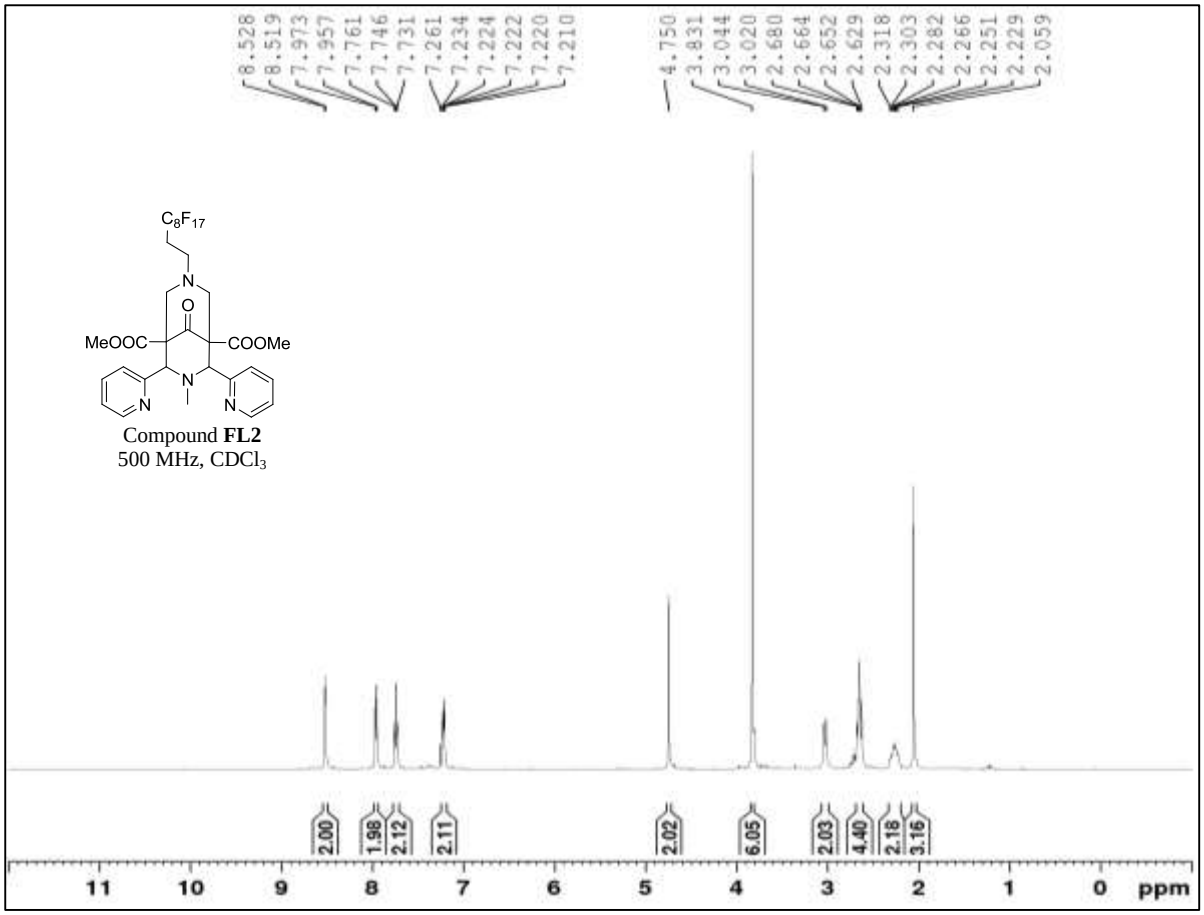


Compound **FL1**
126 MHz, $CDCl_3$



Compound **FL2**
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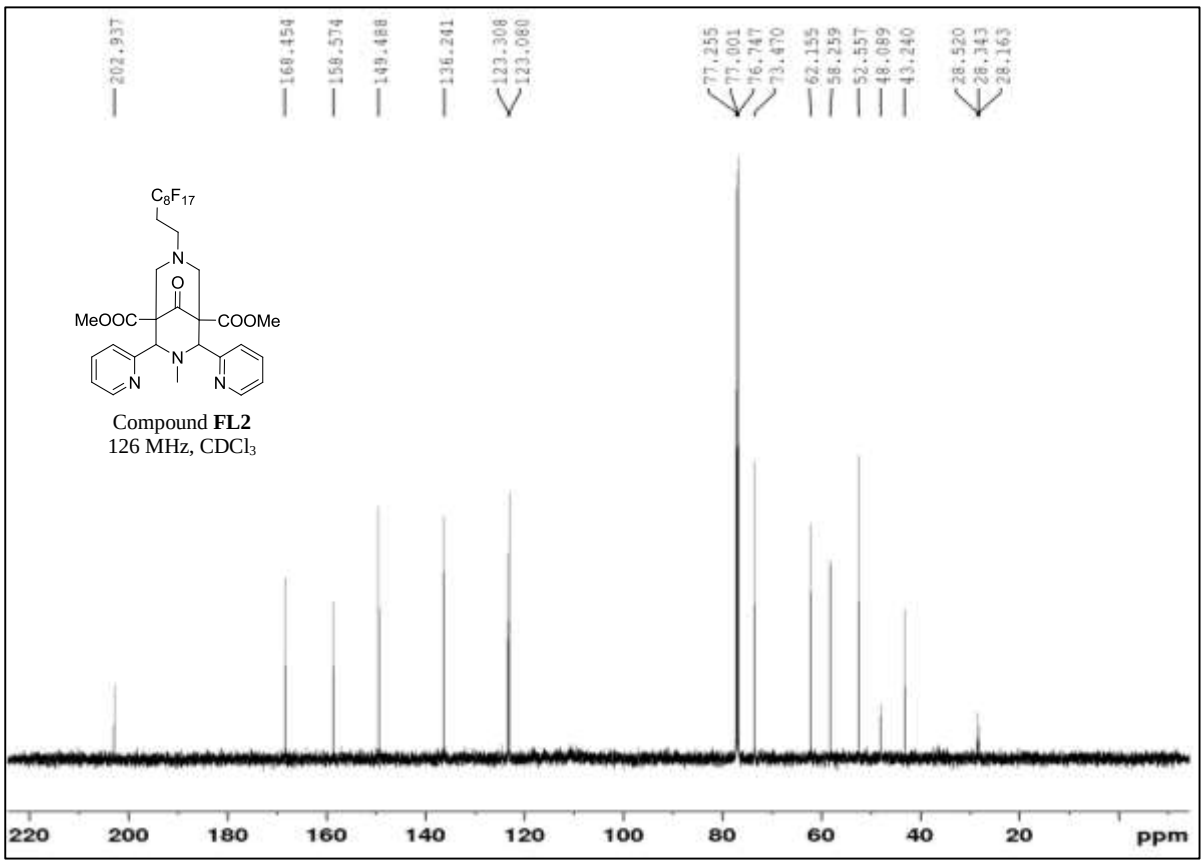
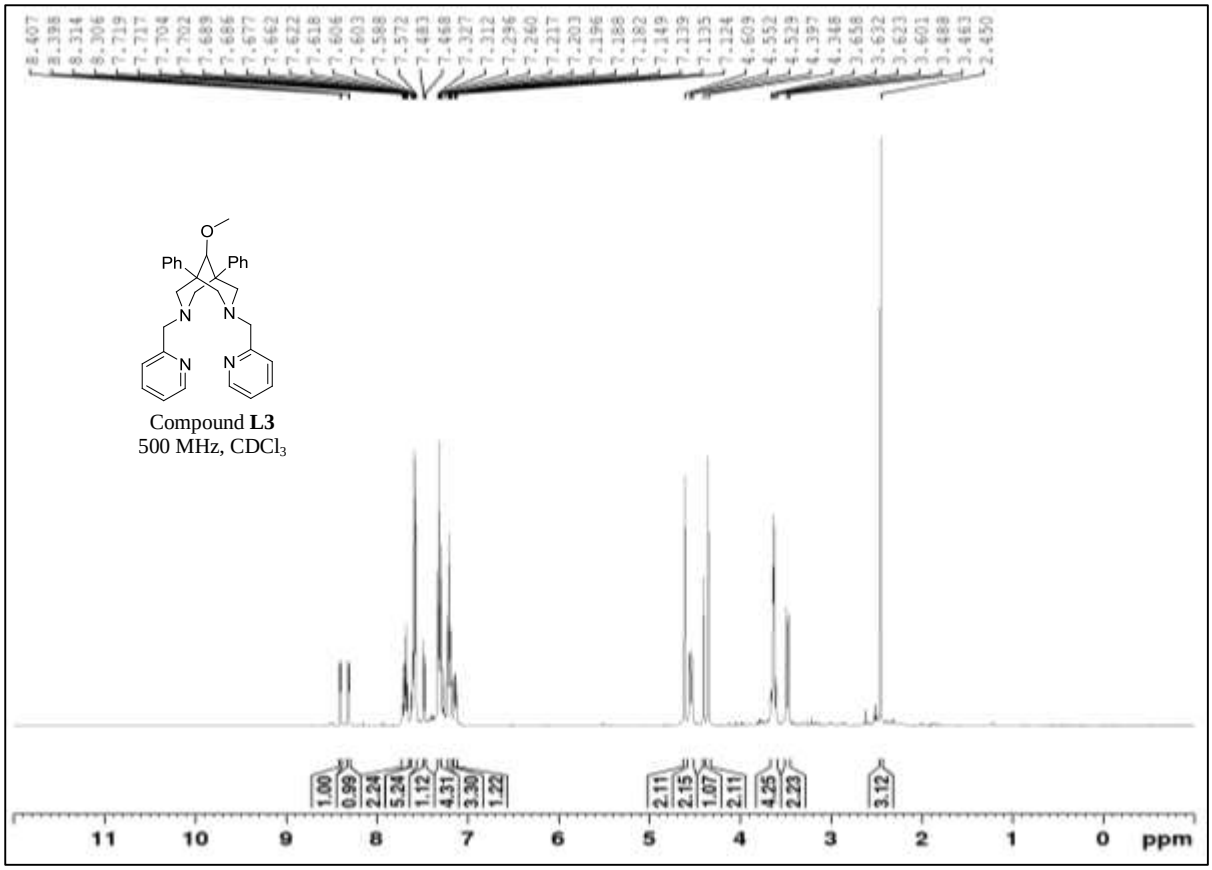
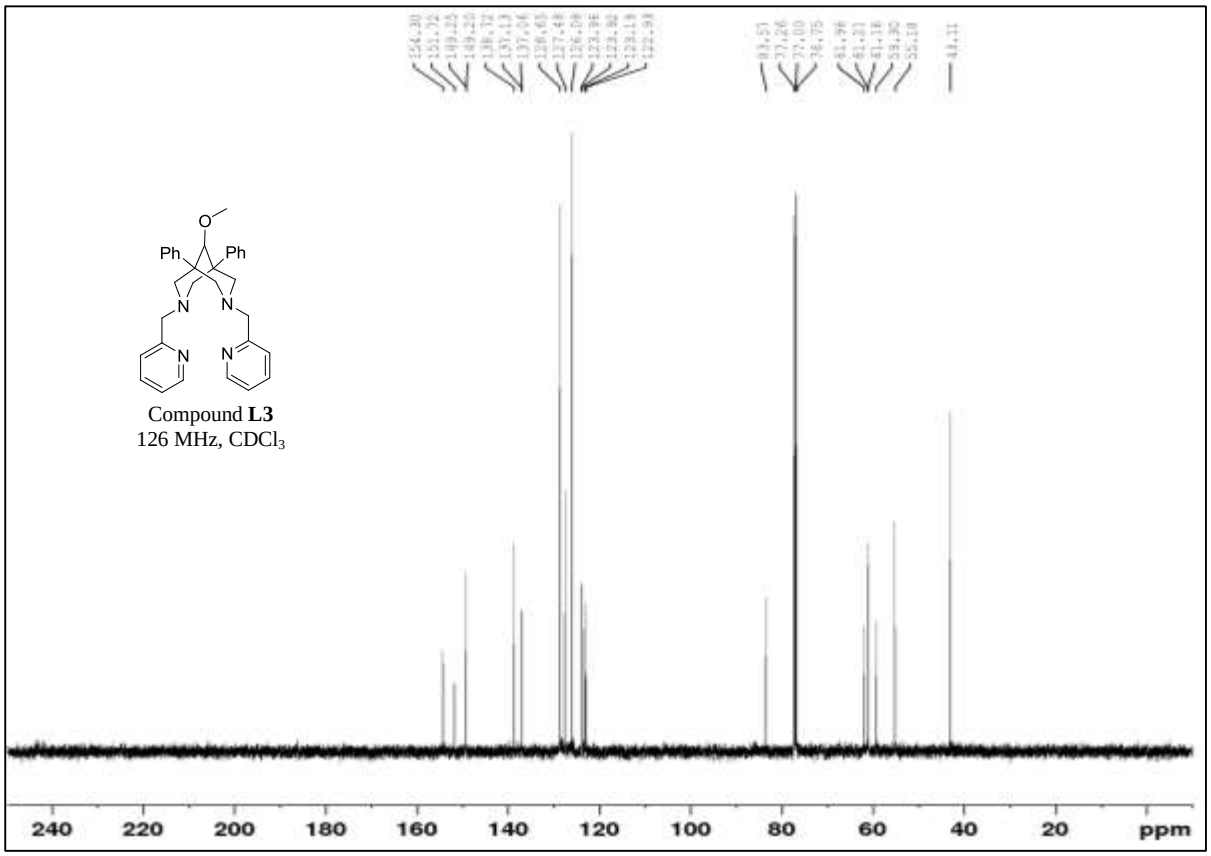


Compound **FL2**
126 MHz, $CDCl_3$



Compound **L3**
500 MHz, $CDCl_3$



Compound **L3**
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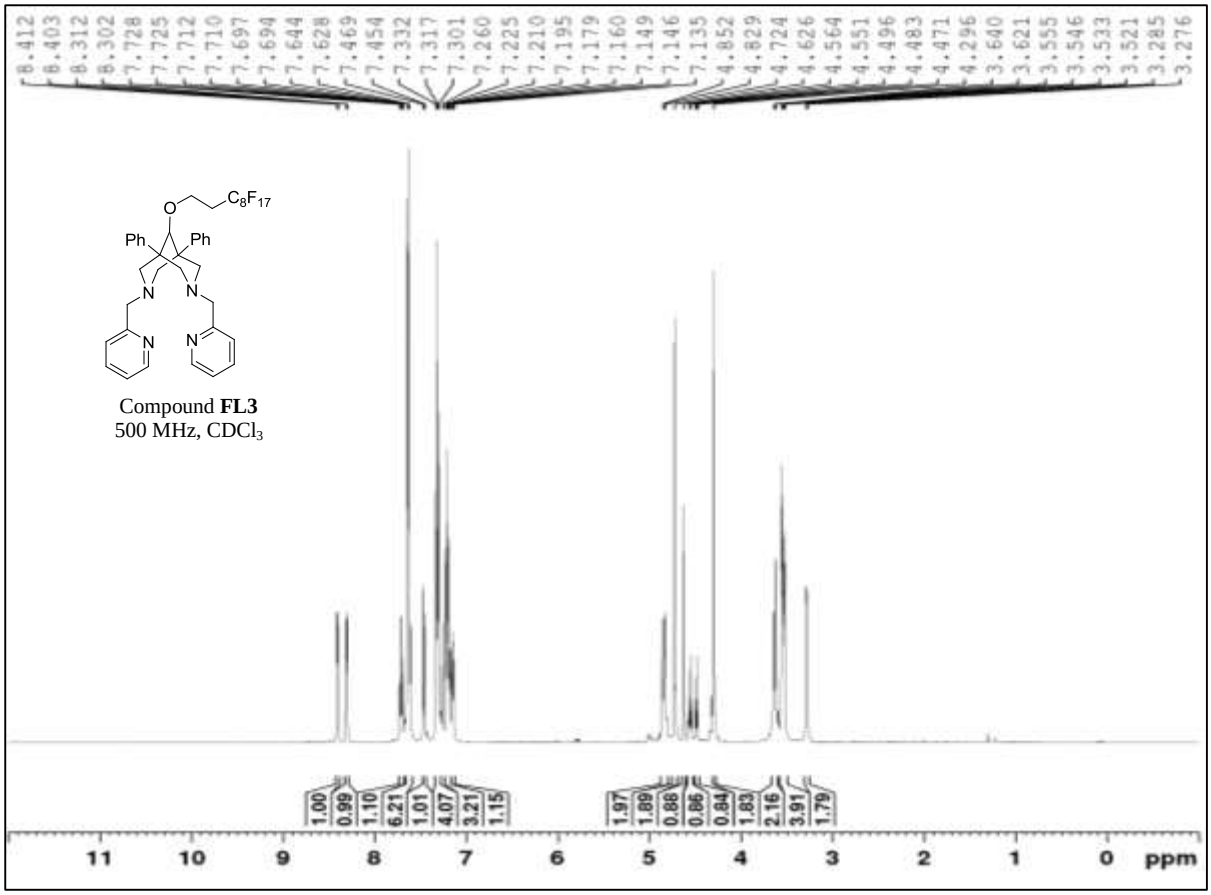
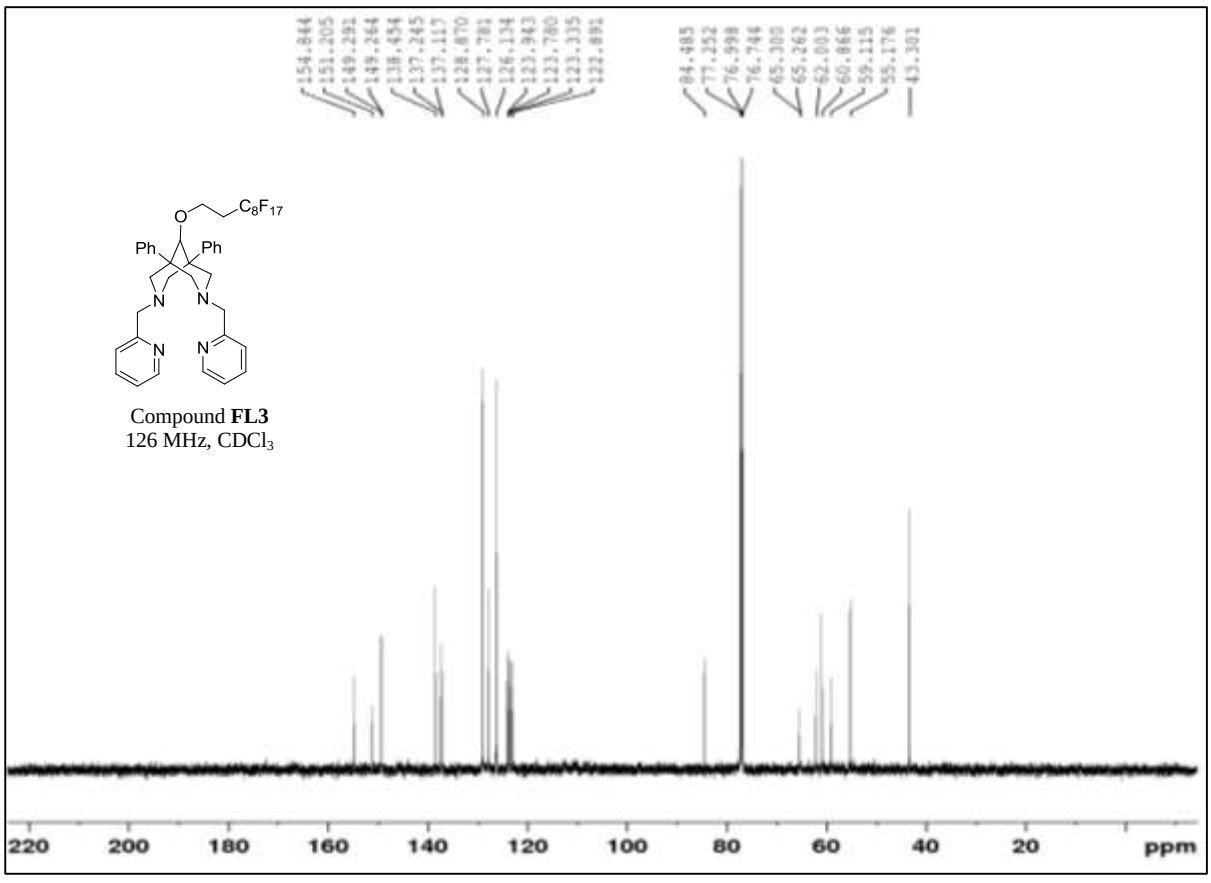
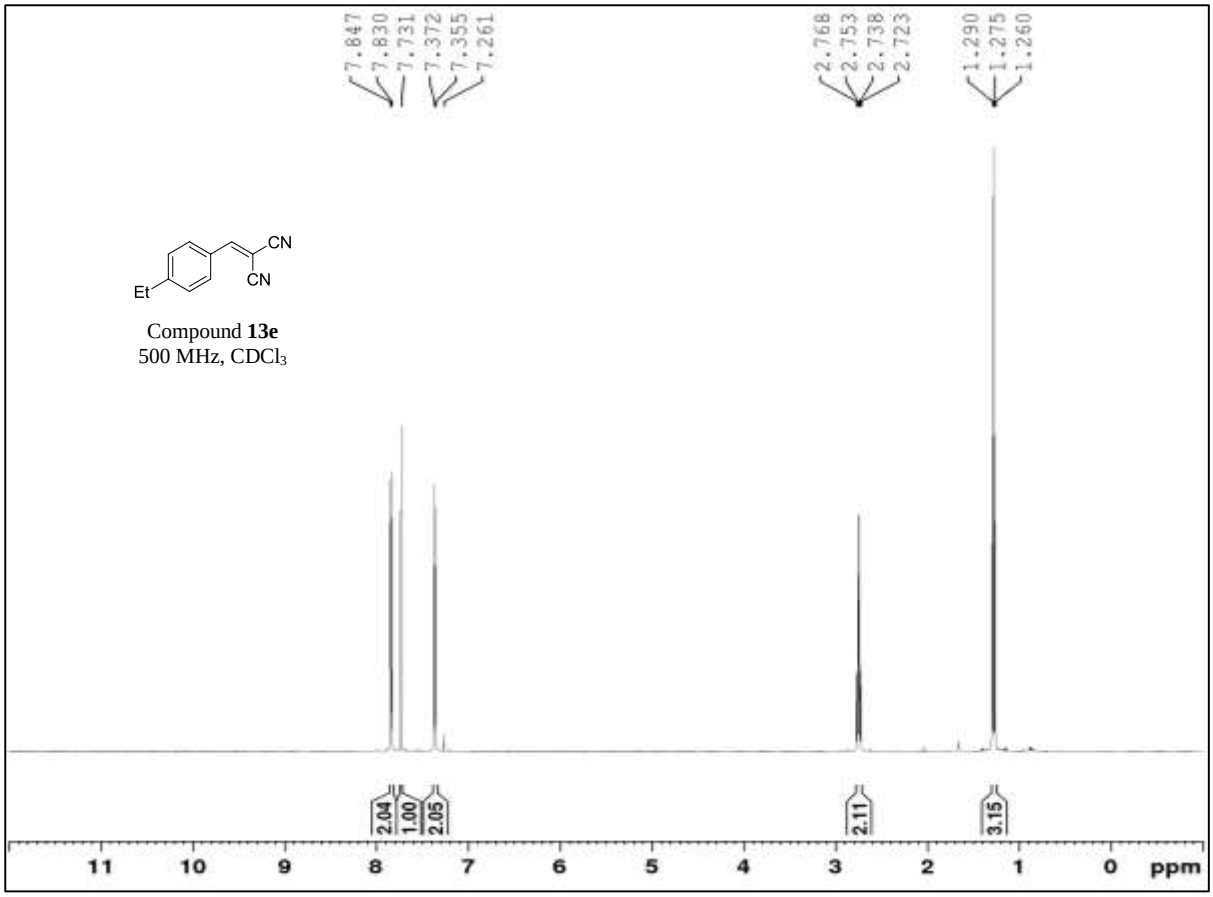


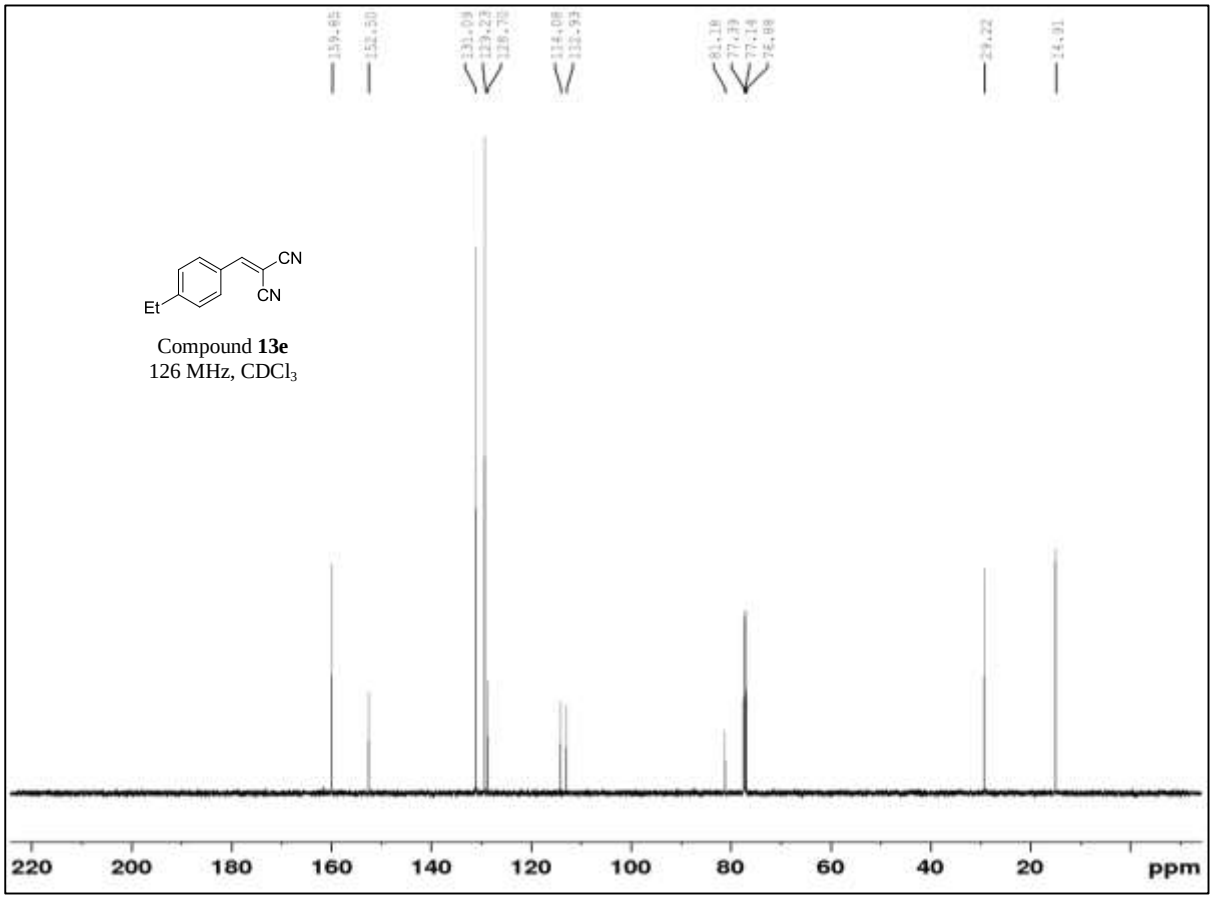
Compound **FL3**
500 MHz, $CDCl_3$



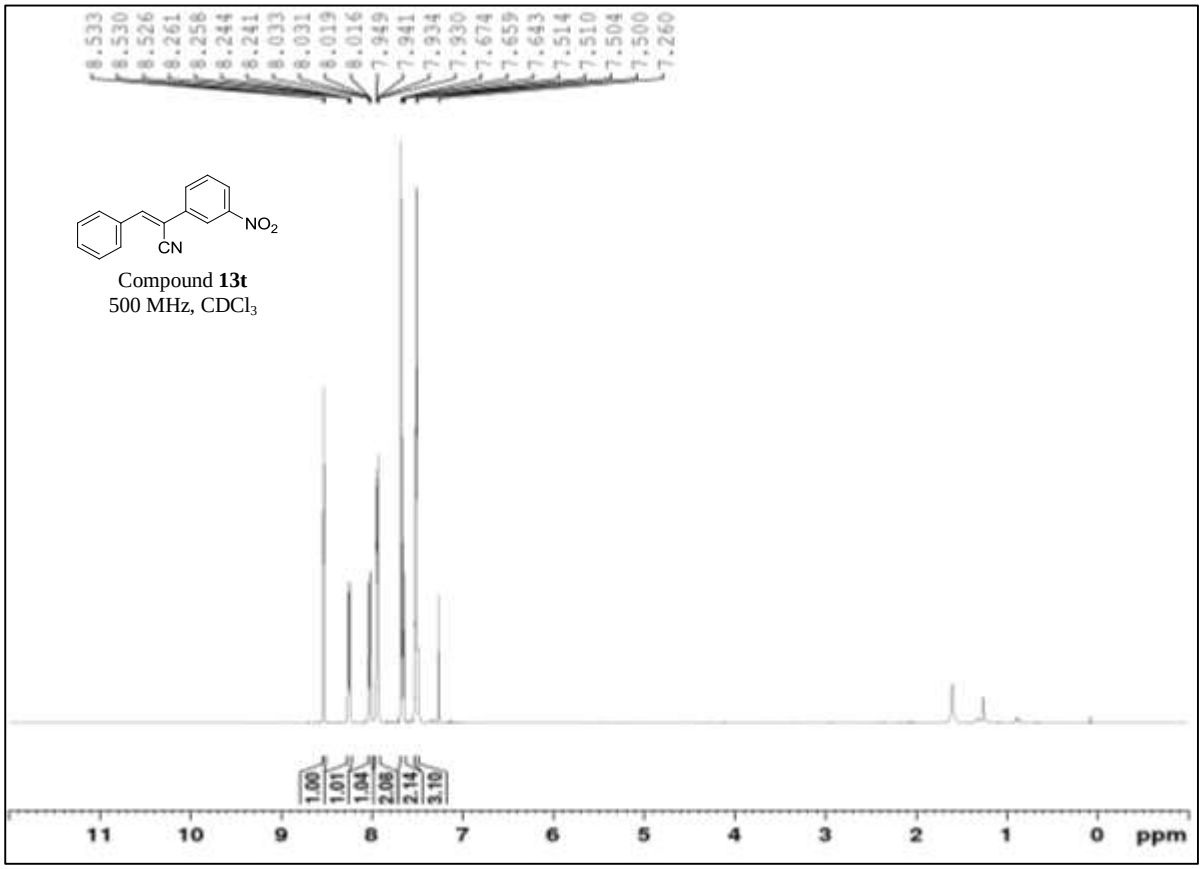
Compound **FL3**
126 MHz, $CDCl_3$



Compound **13e**
500 MHz, $CDCl_3$



Compound **13e**
126 MHz, $CDCl_3$



Compound **13t**
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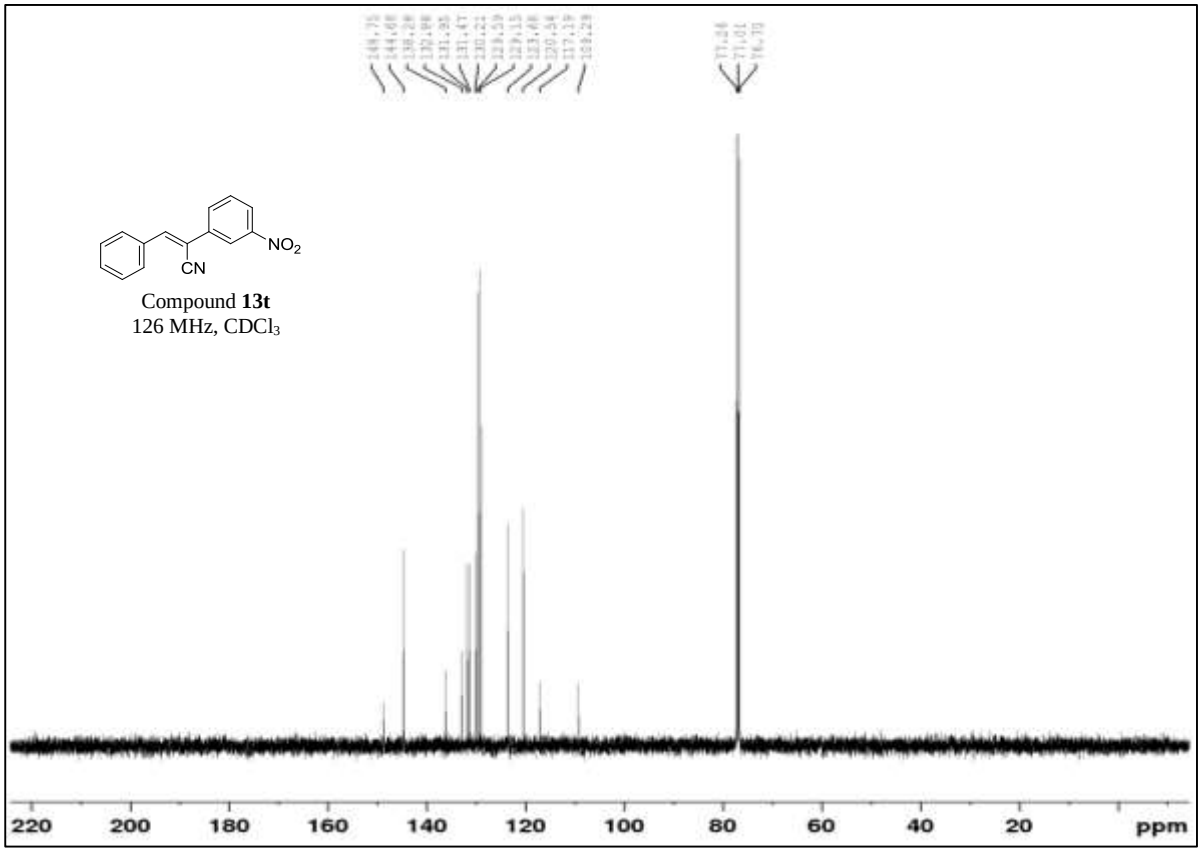
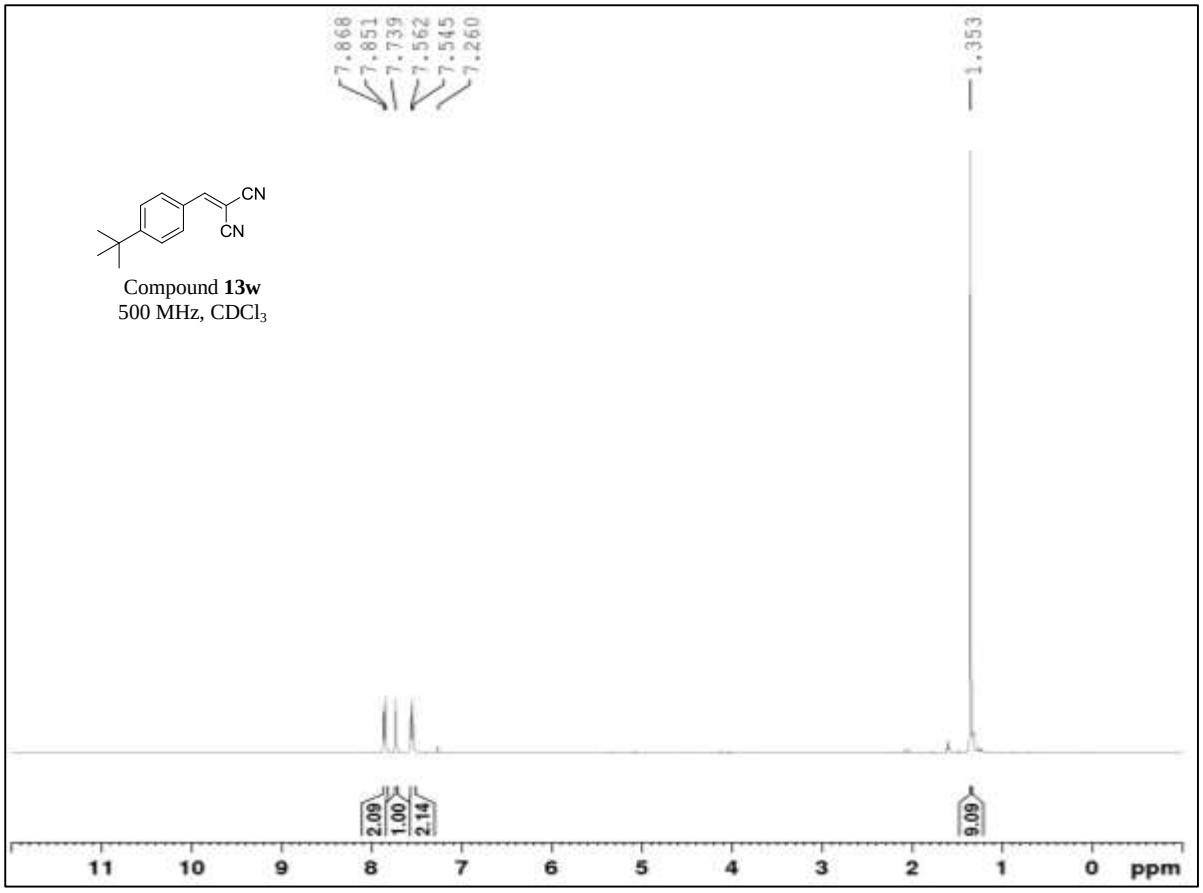


Compound **13t**
126 MHz, $CDCl_3$



Compound **13w**
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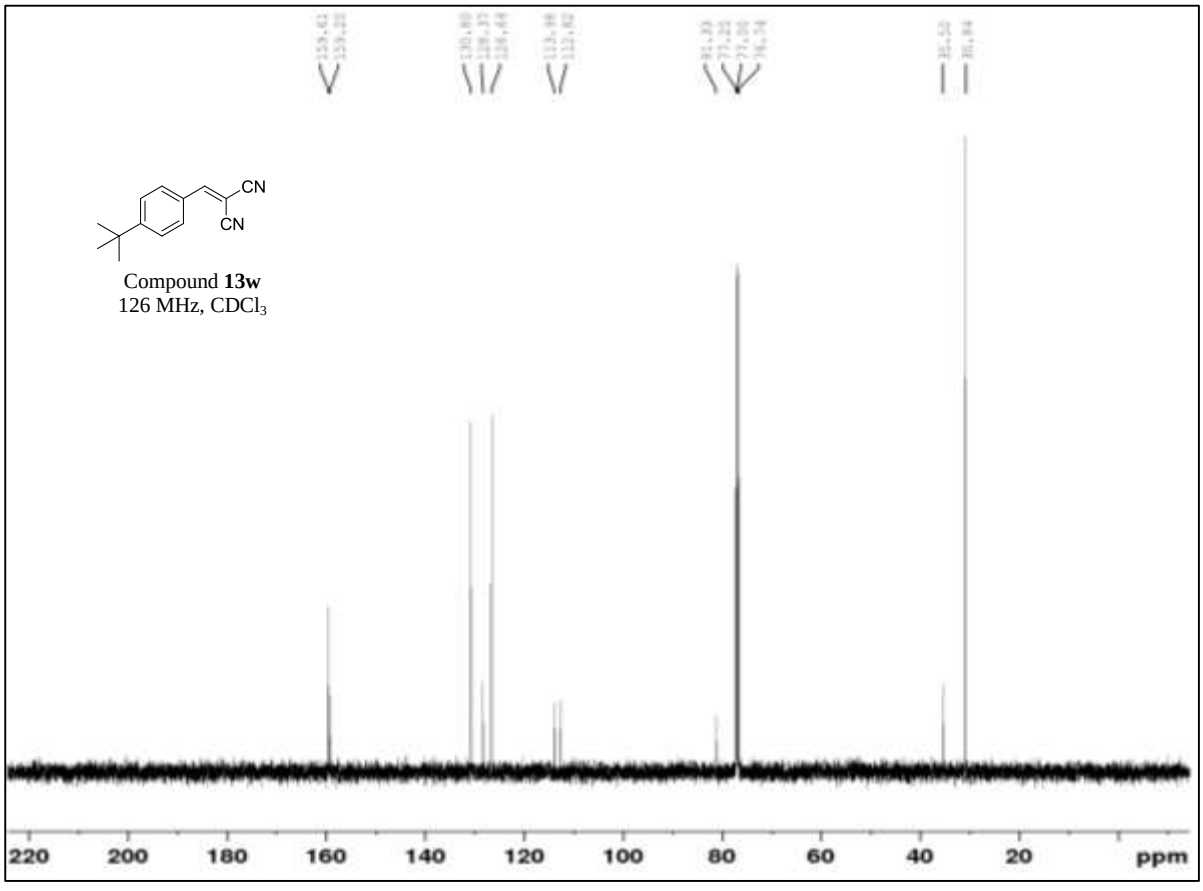
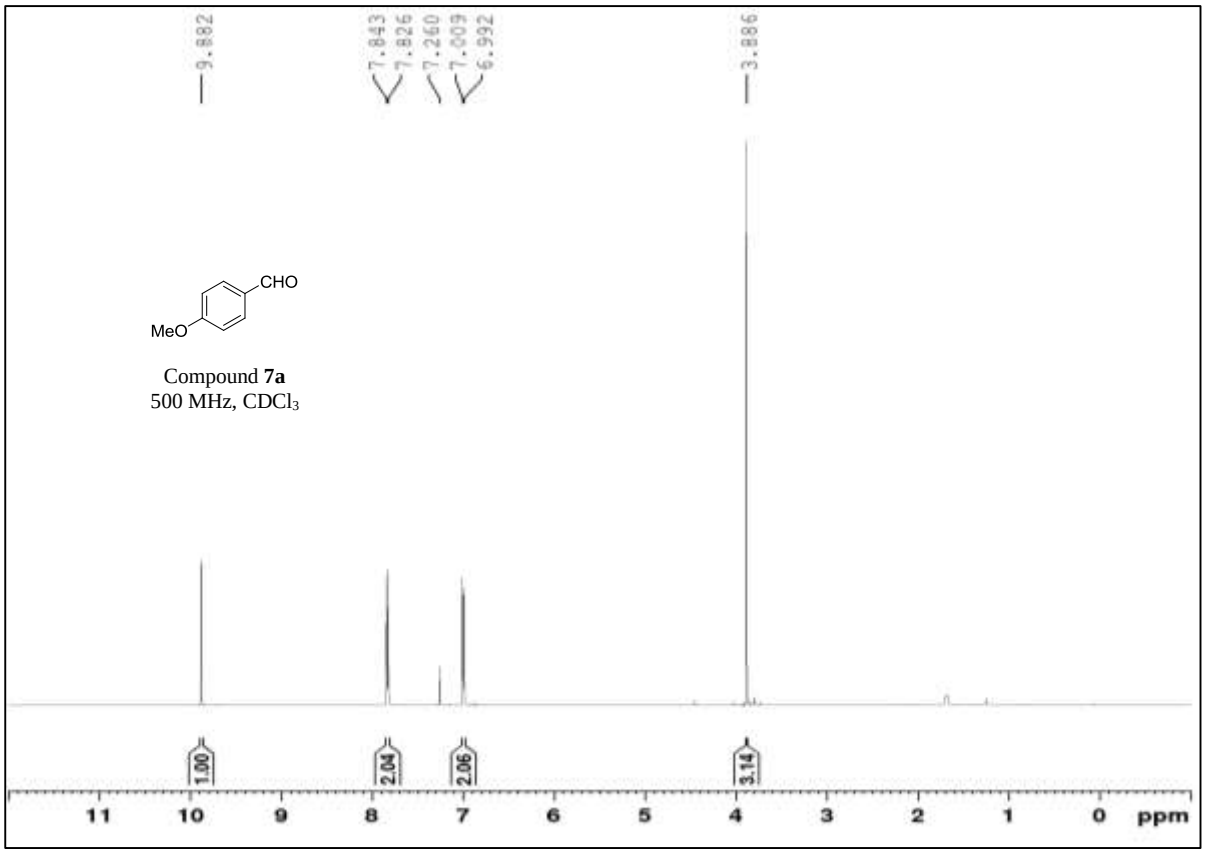
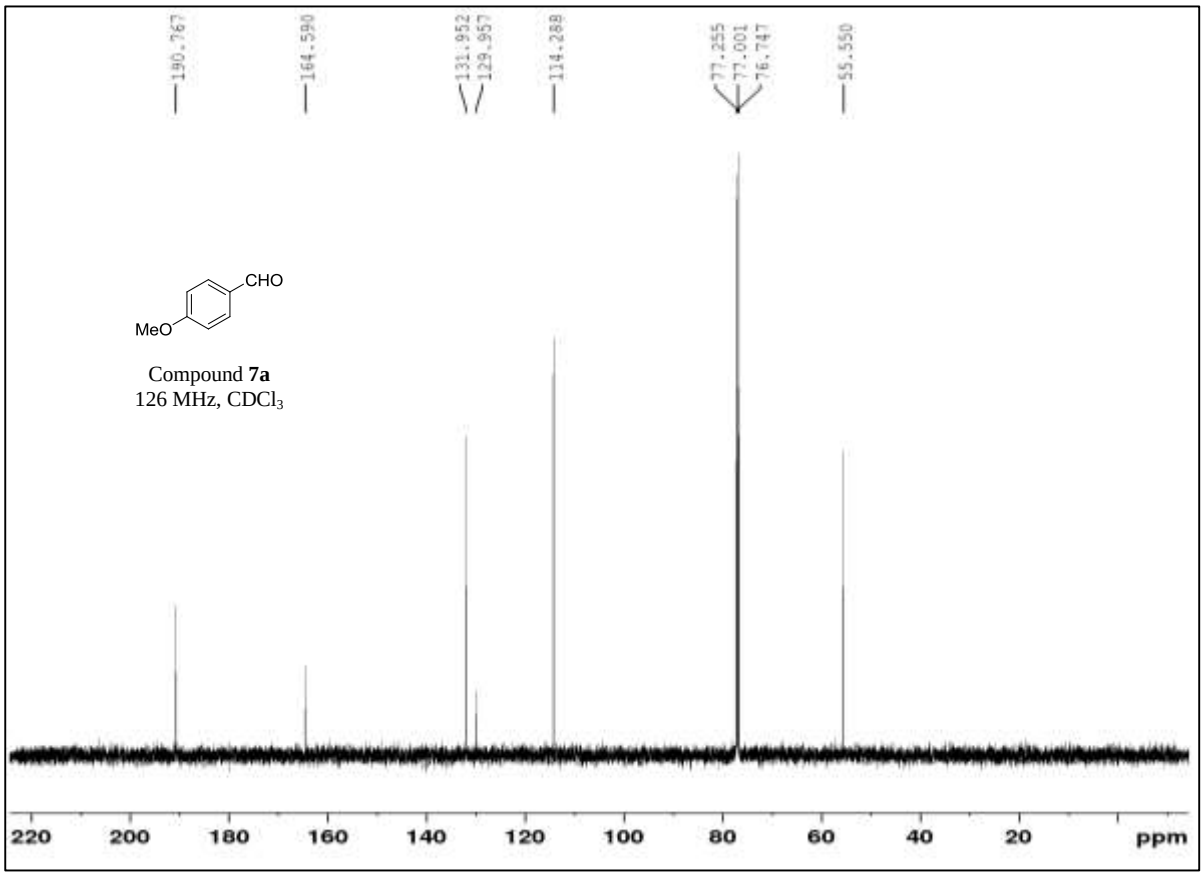


Compound **13w**
126 MHz, $CDCl_3$

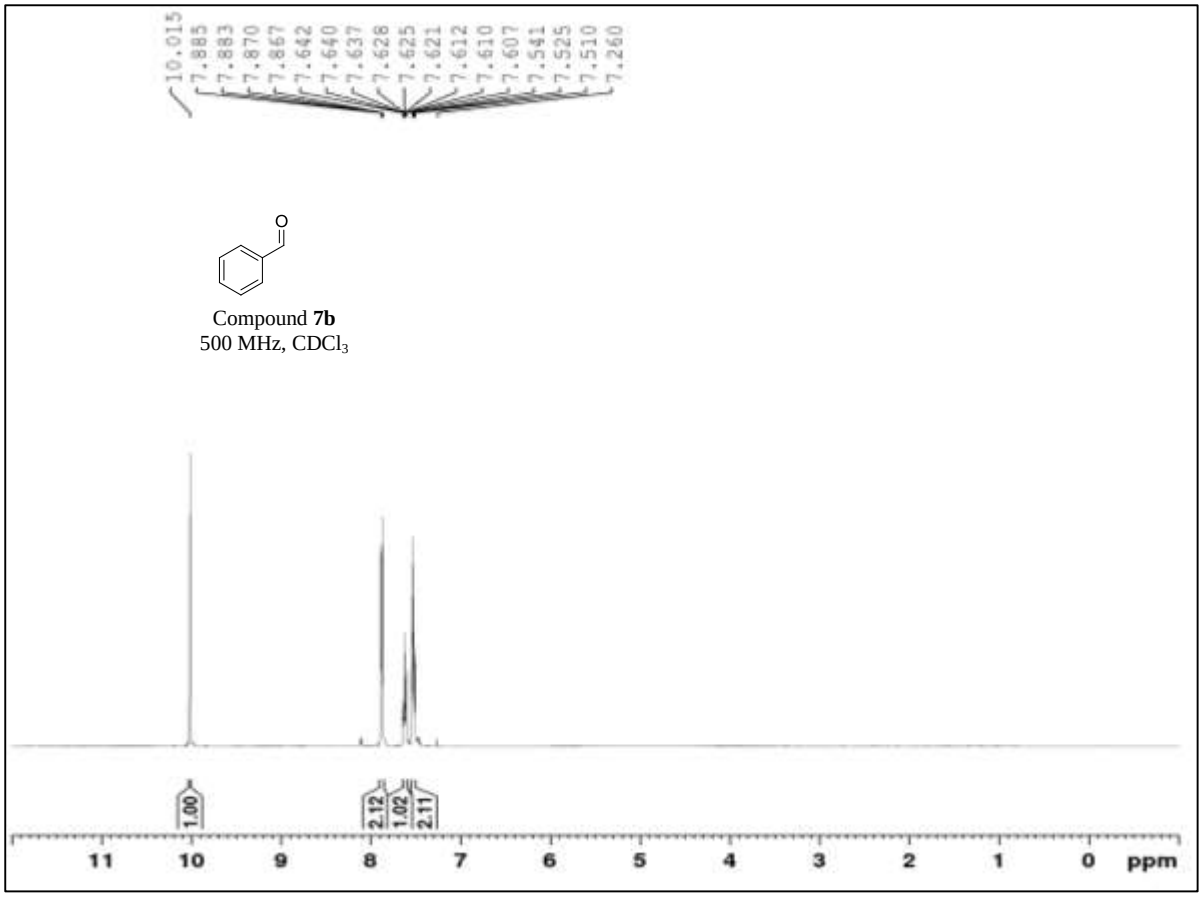
NMR spectra of known compounds:



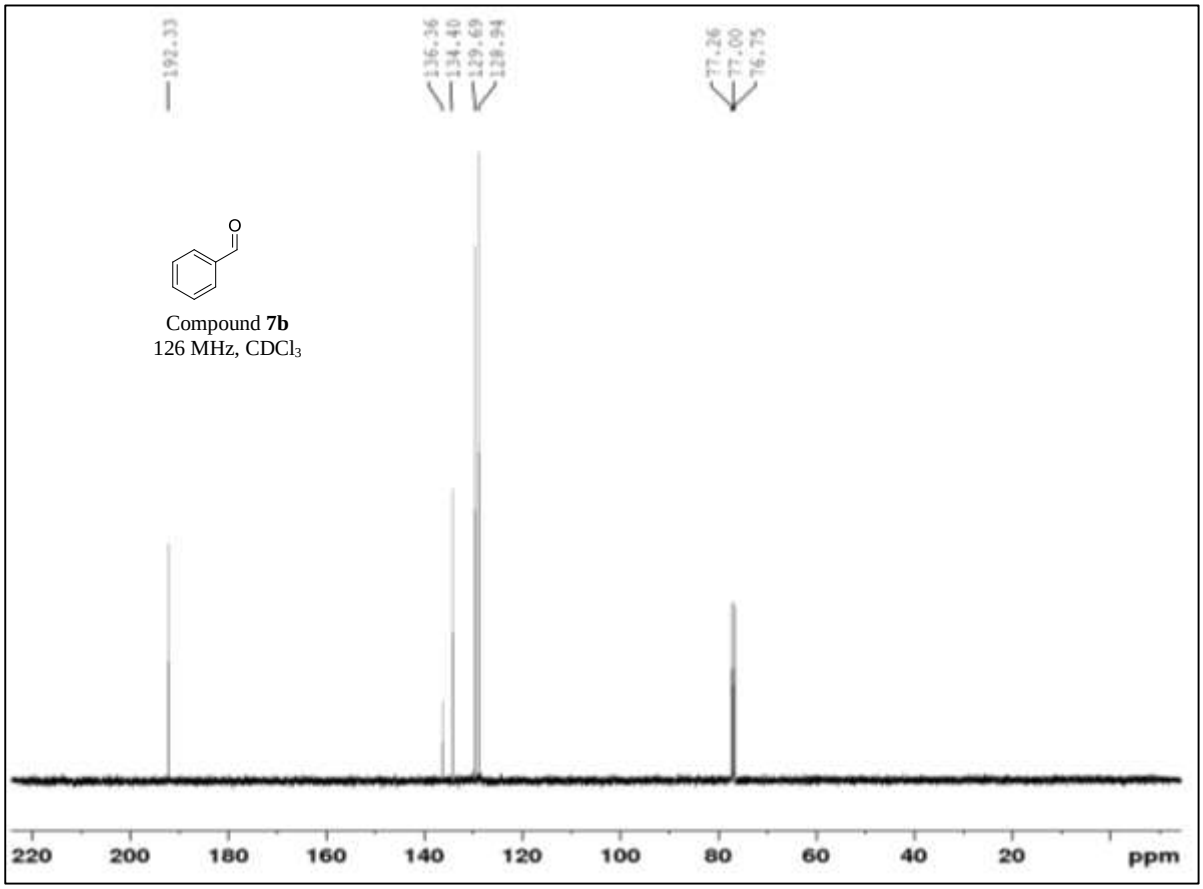
Compound **7a**
500 MHz, $CDCl_3$



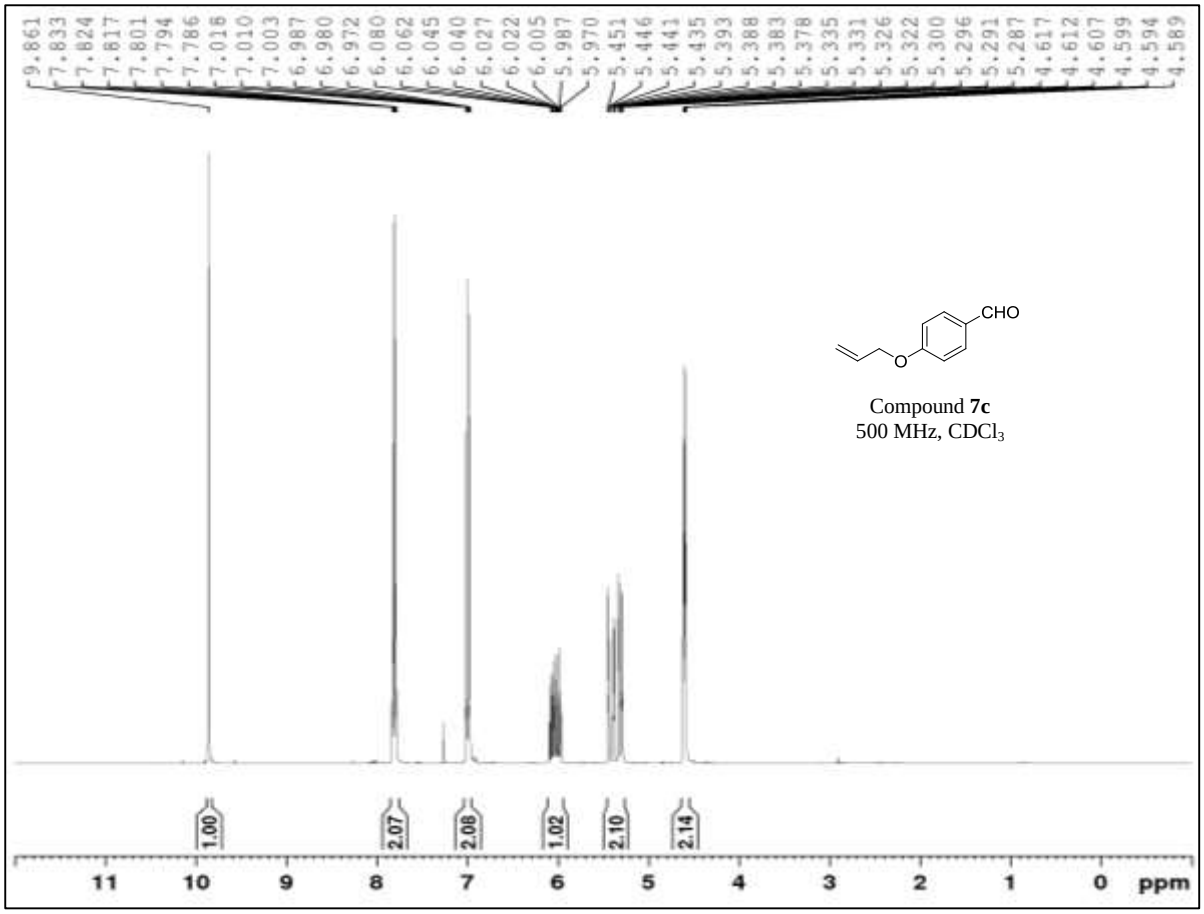
Compound **7a**
126 MHz, $CDCl_3$



Compound **7b**
500 MHz, $CDCl_3$



Compound **7b**
126 MHz, $CDCl_3$



Compound **7c**
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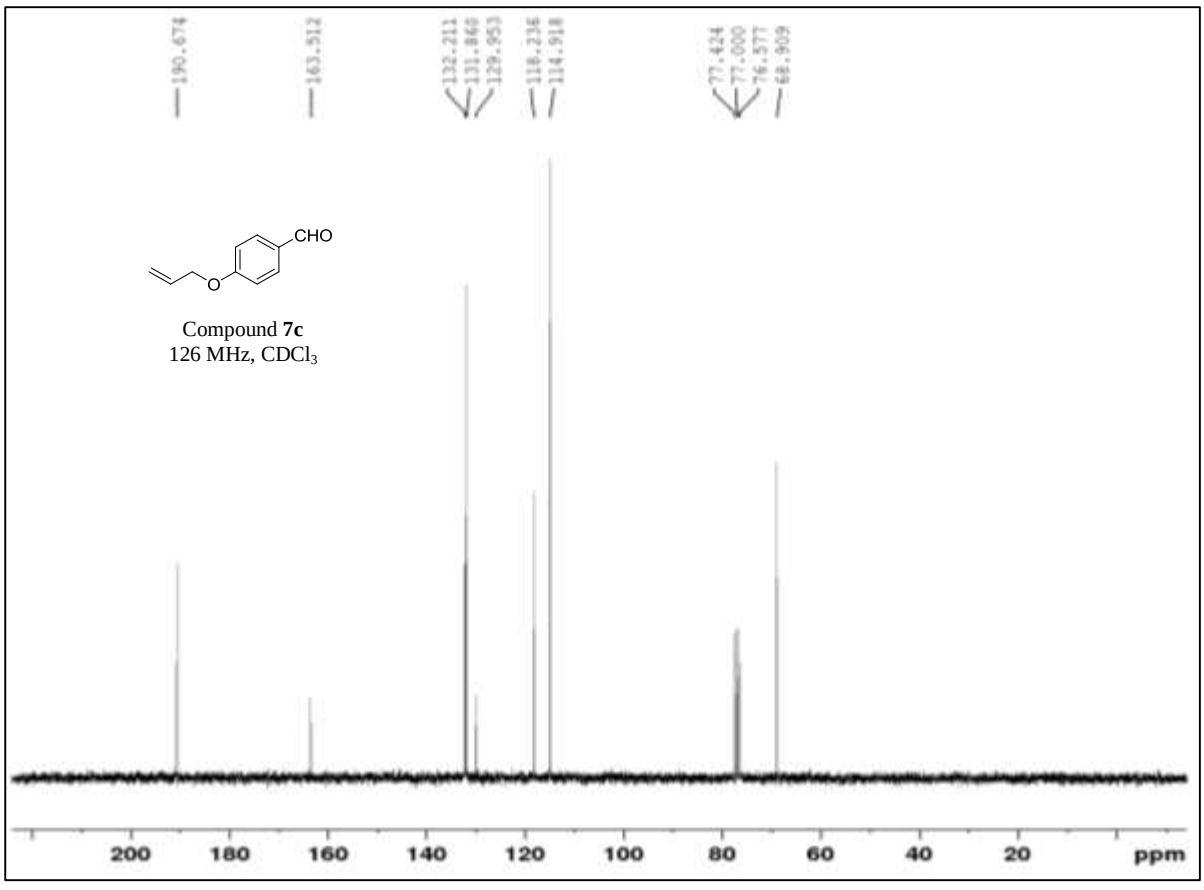
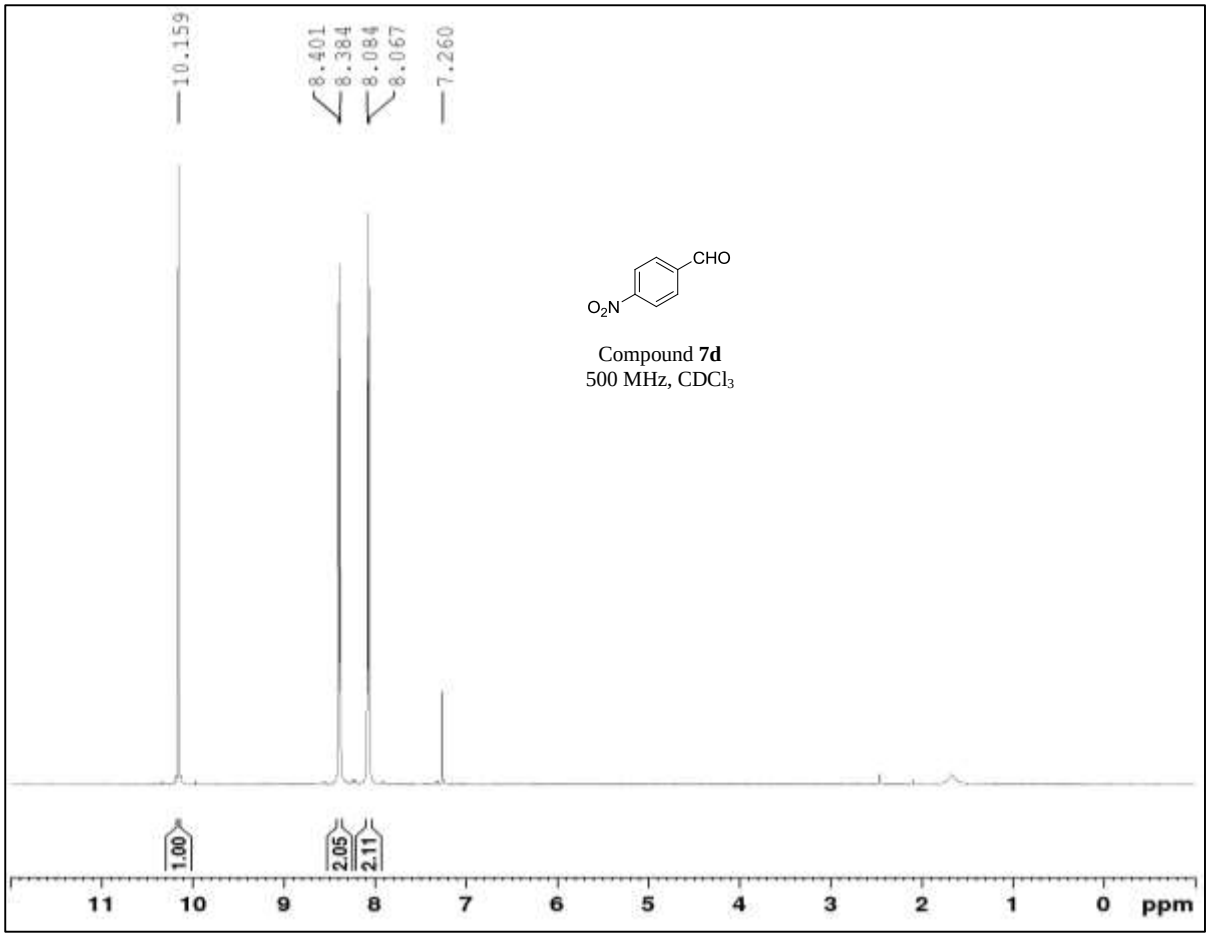
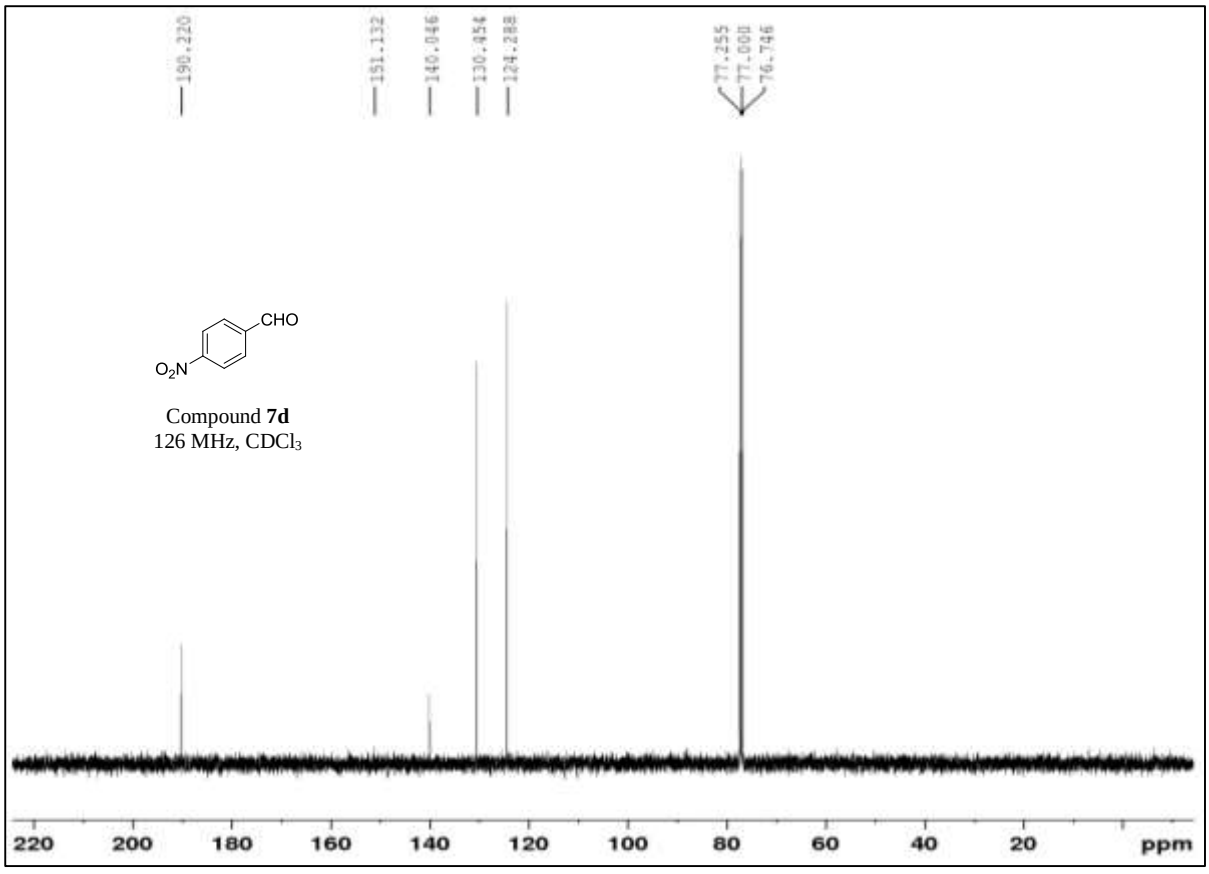


Compound **7c**
126 MHz, $CDCl_3$



Compound **7d**
500 MHz, $CDCl_3$



Compound **7d**
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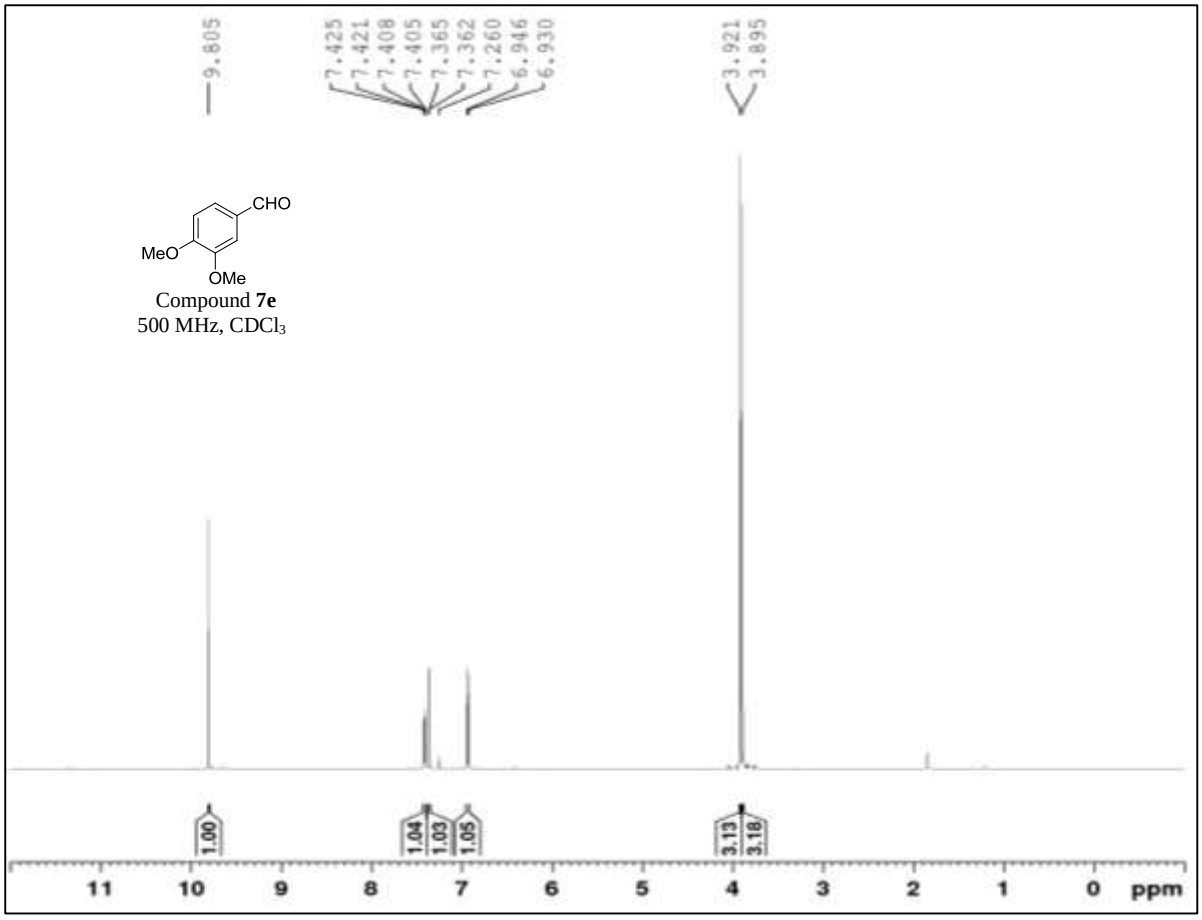

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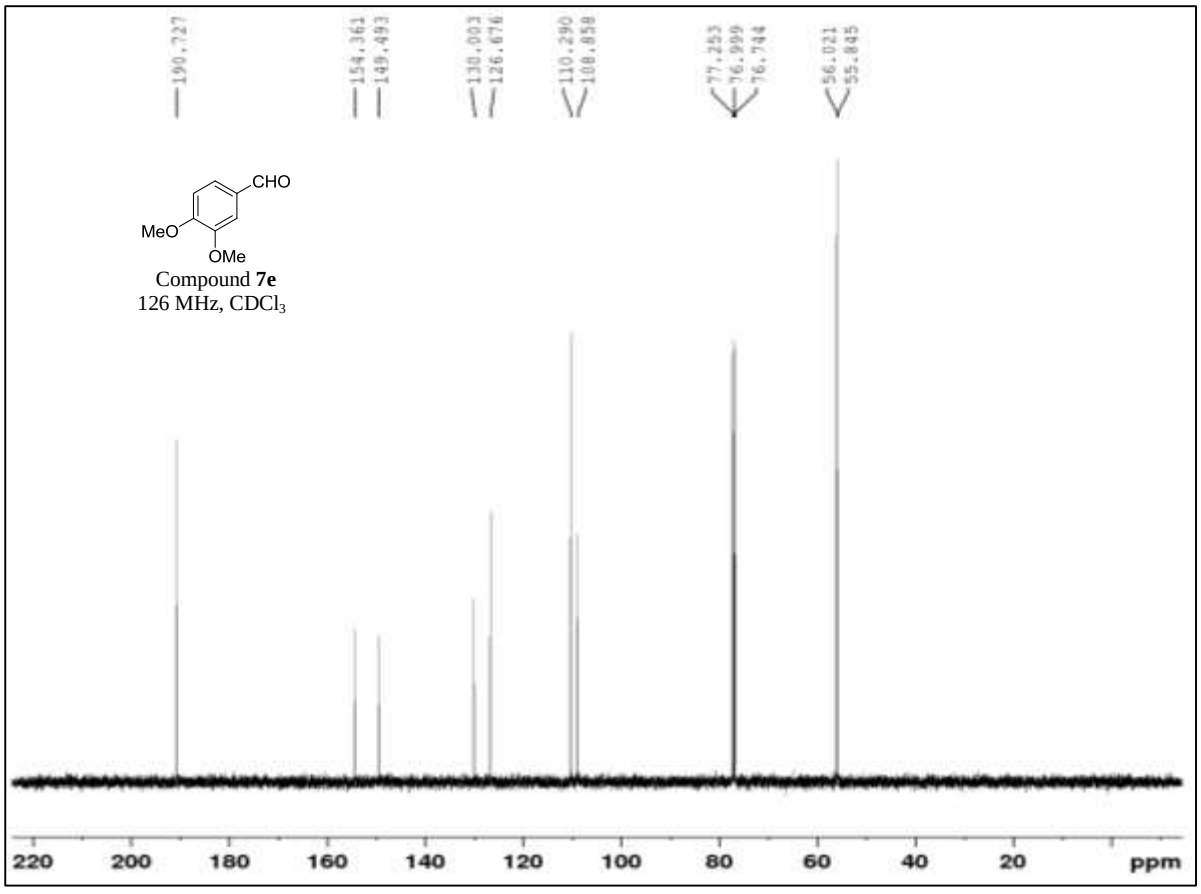

Compound **7e**
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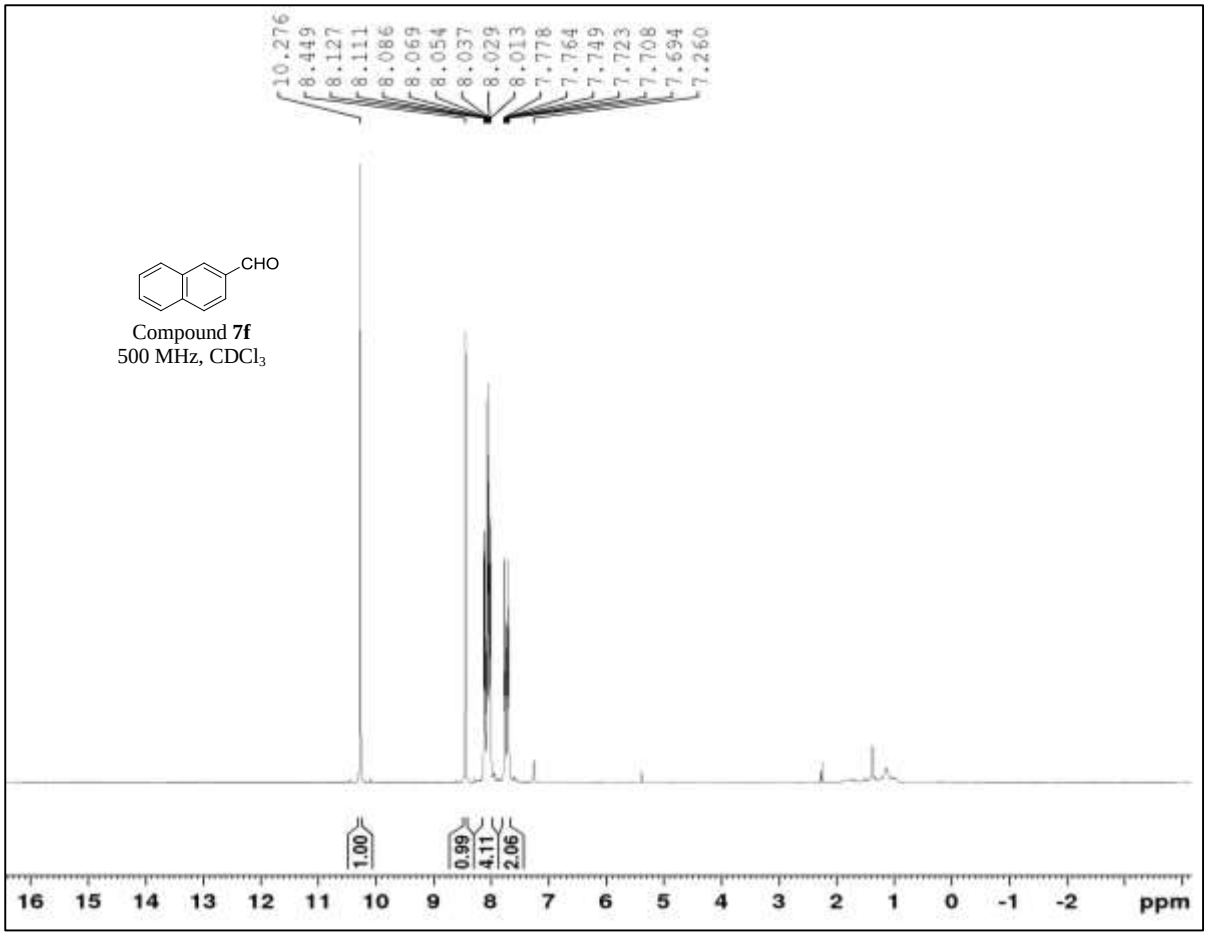

Compound **7f**
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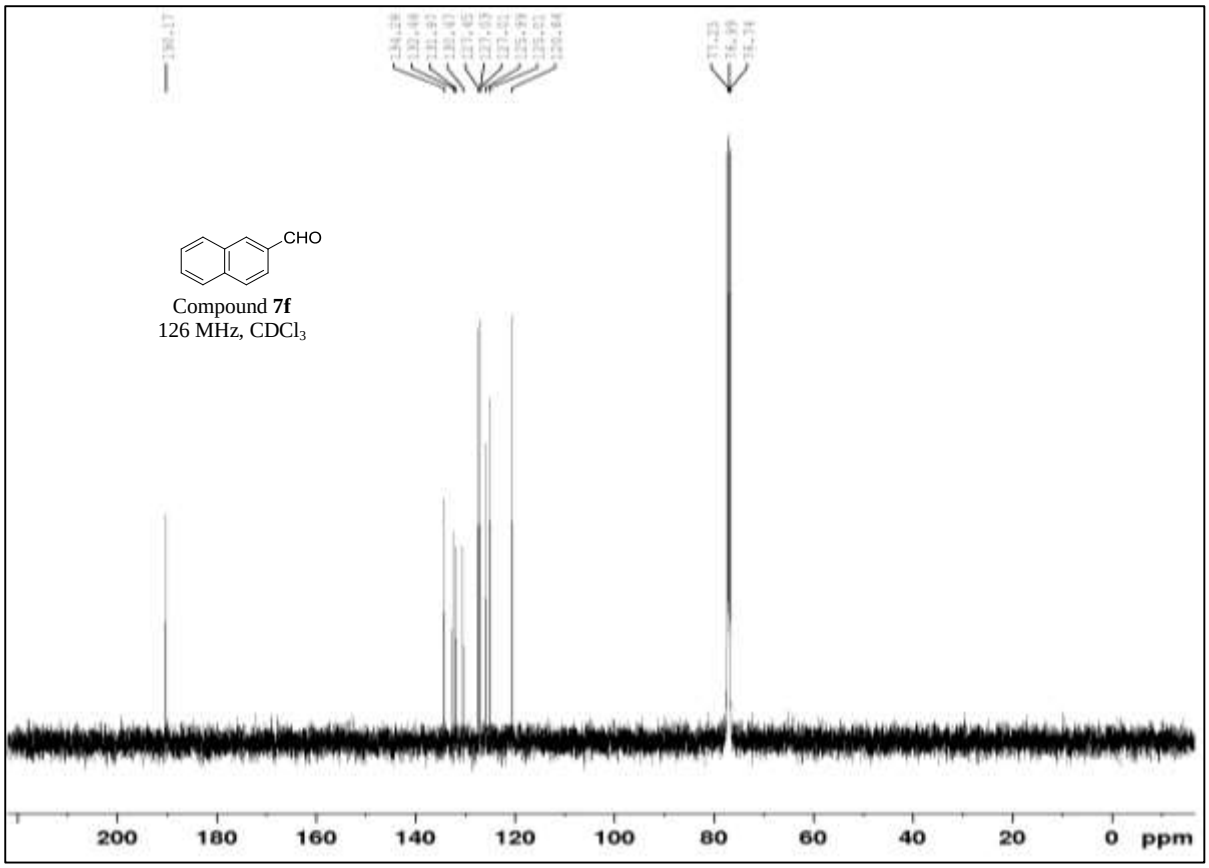

Compound **7f**
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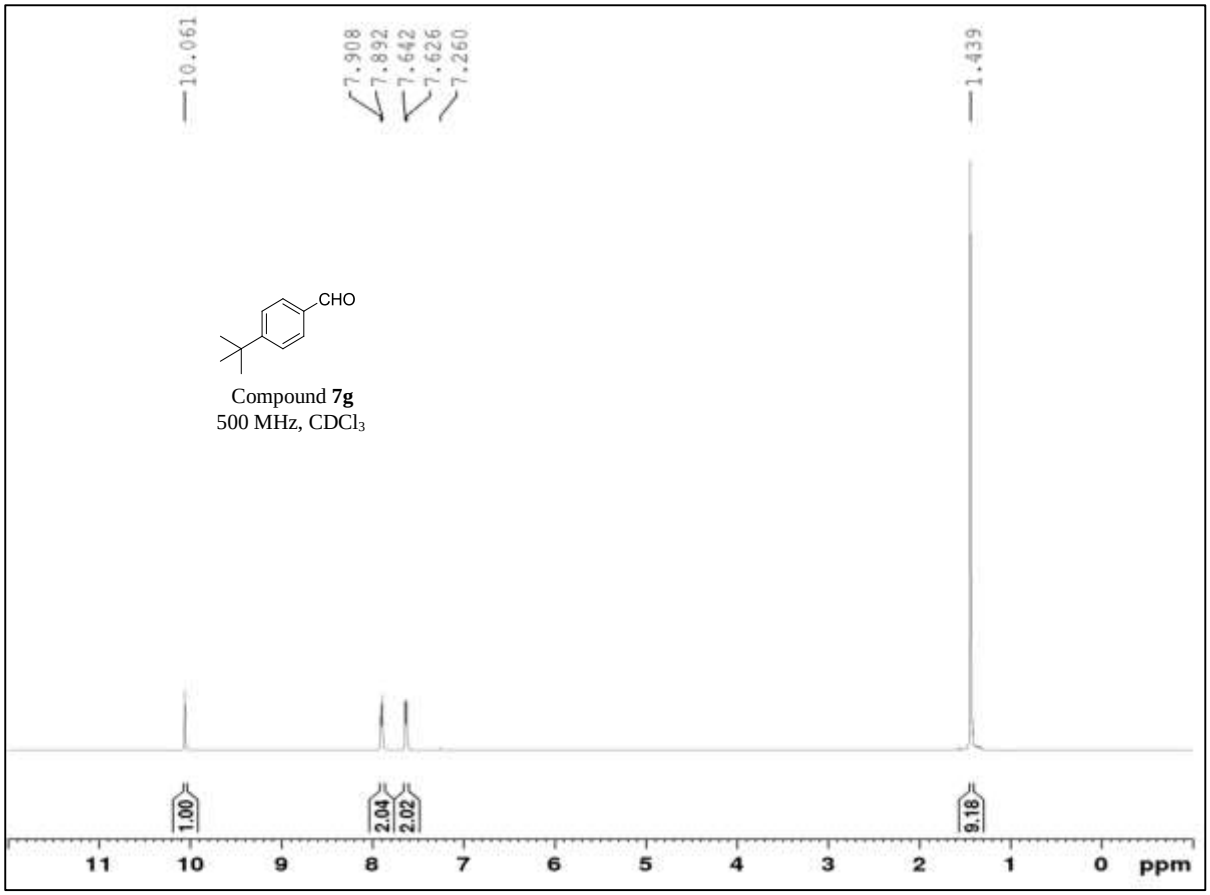

Compound **7g**
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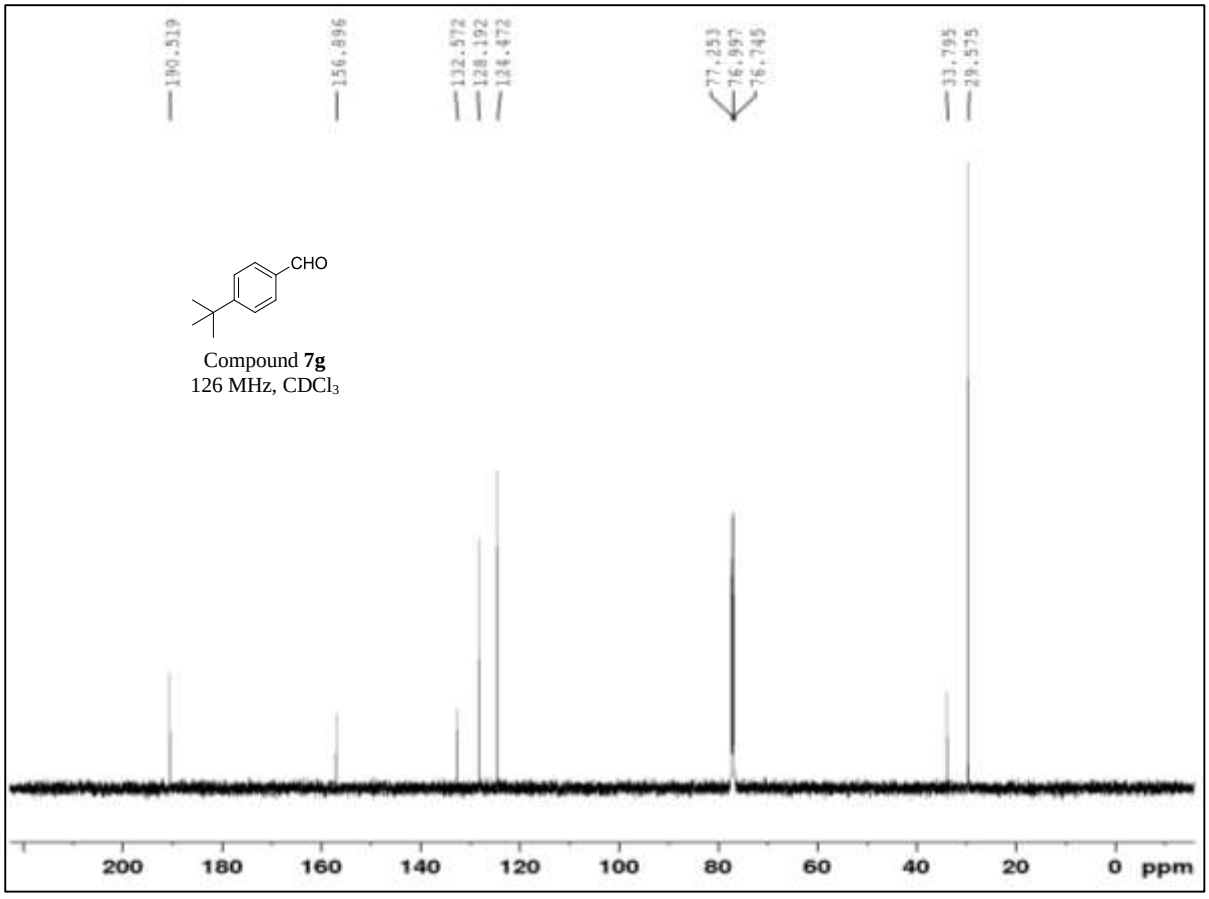

Compound **7g**
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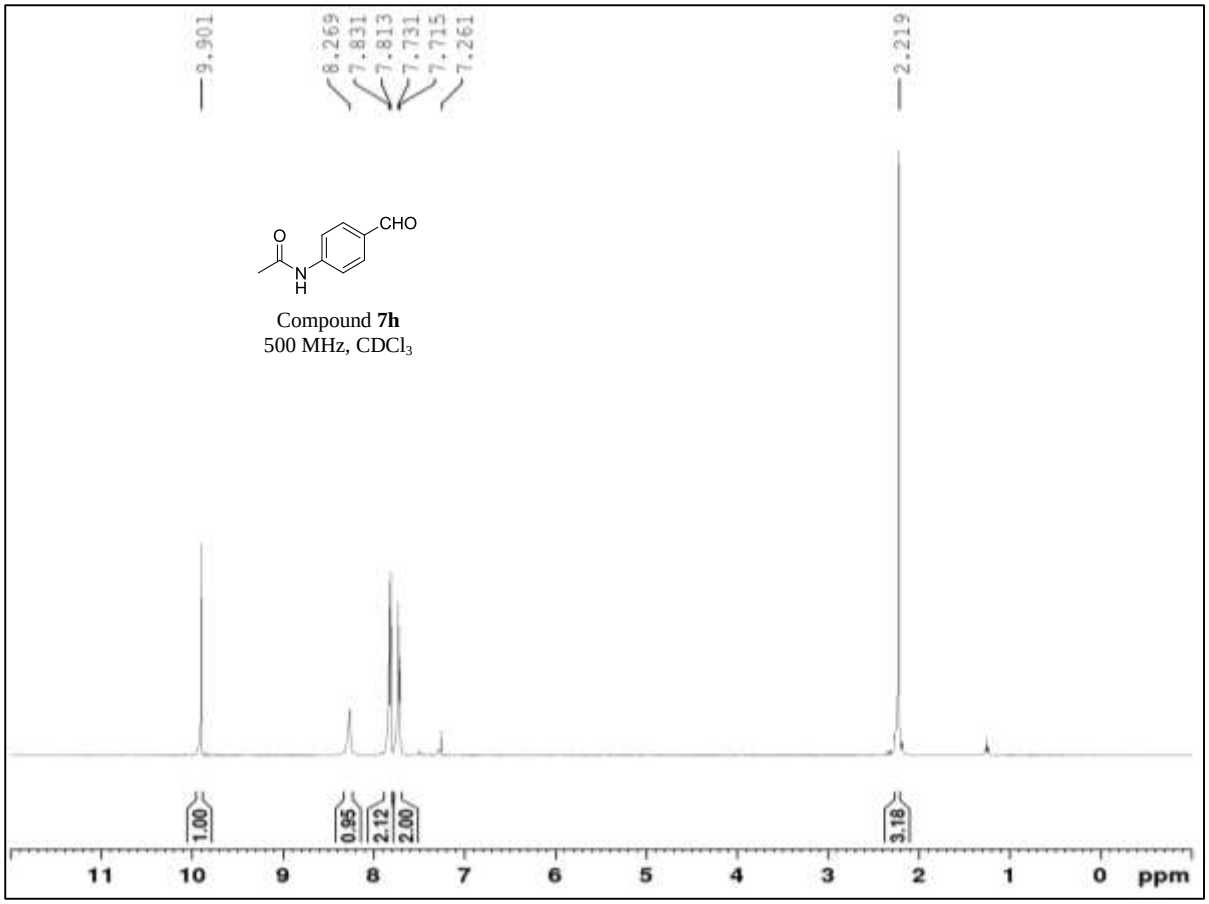

Compound **7h**
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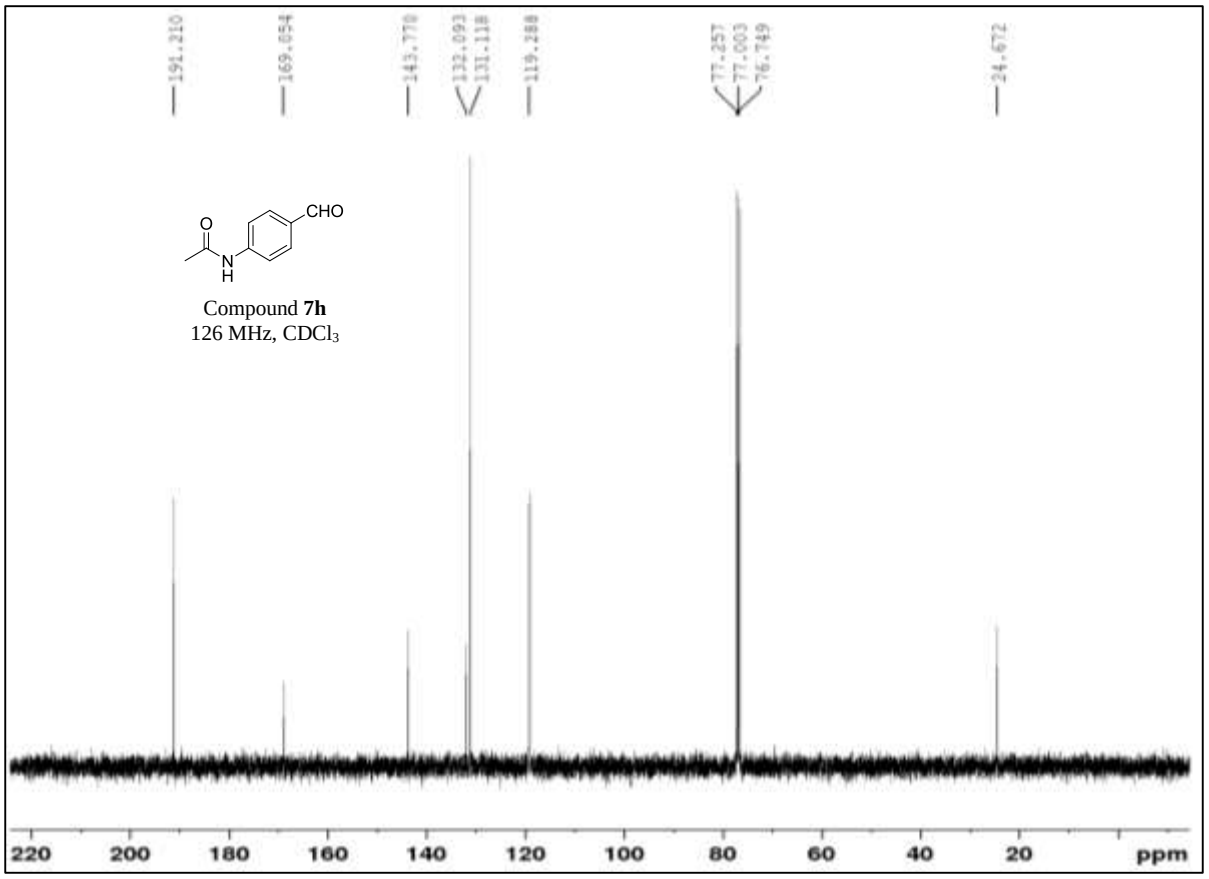

Compound **7h**
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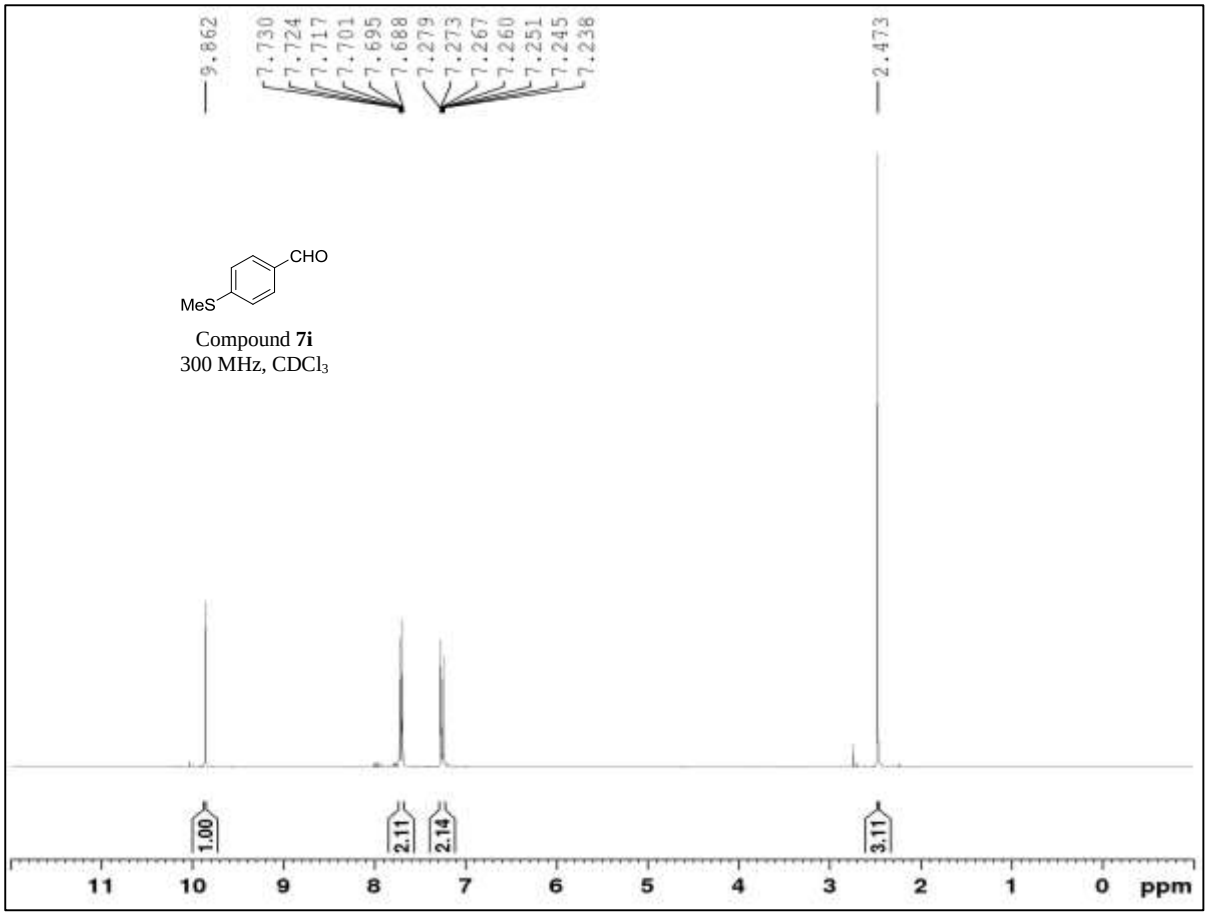
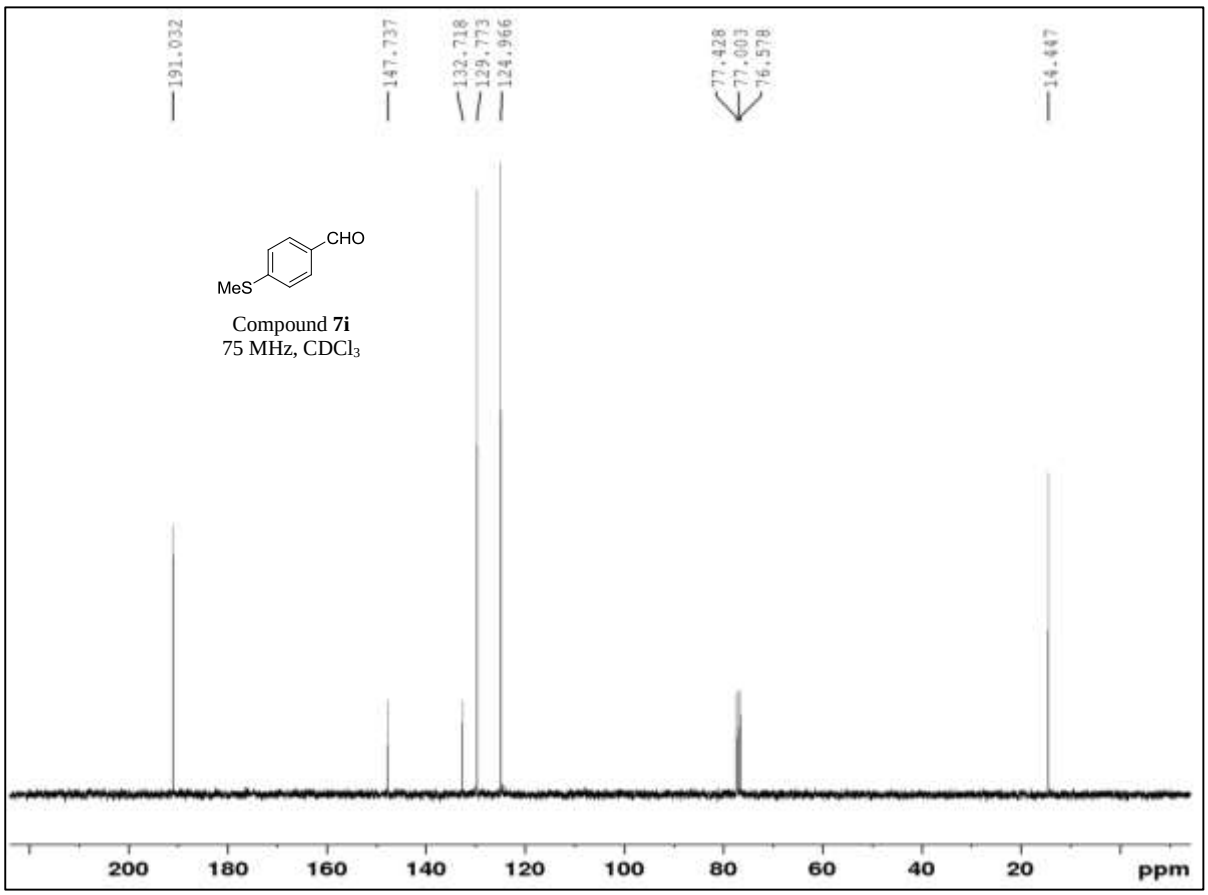
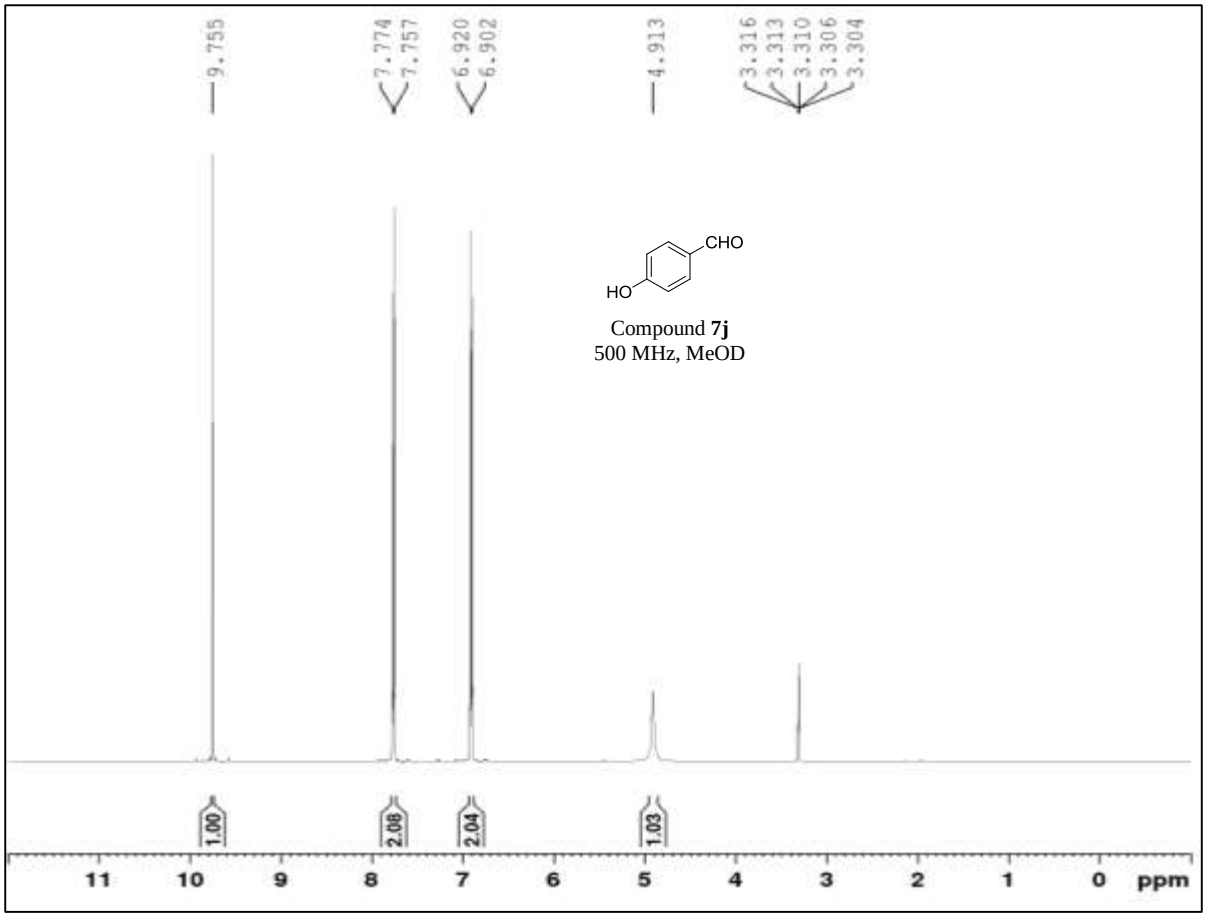


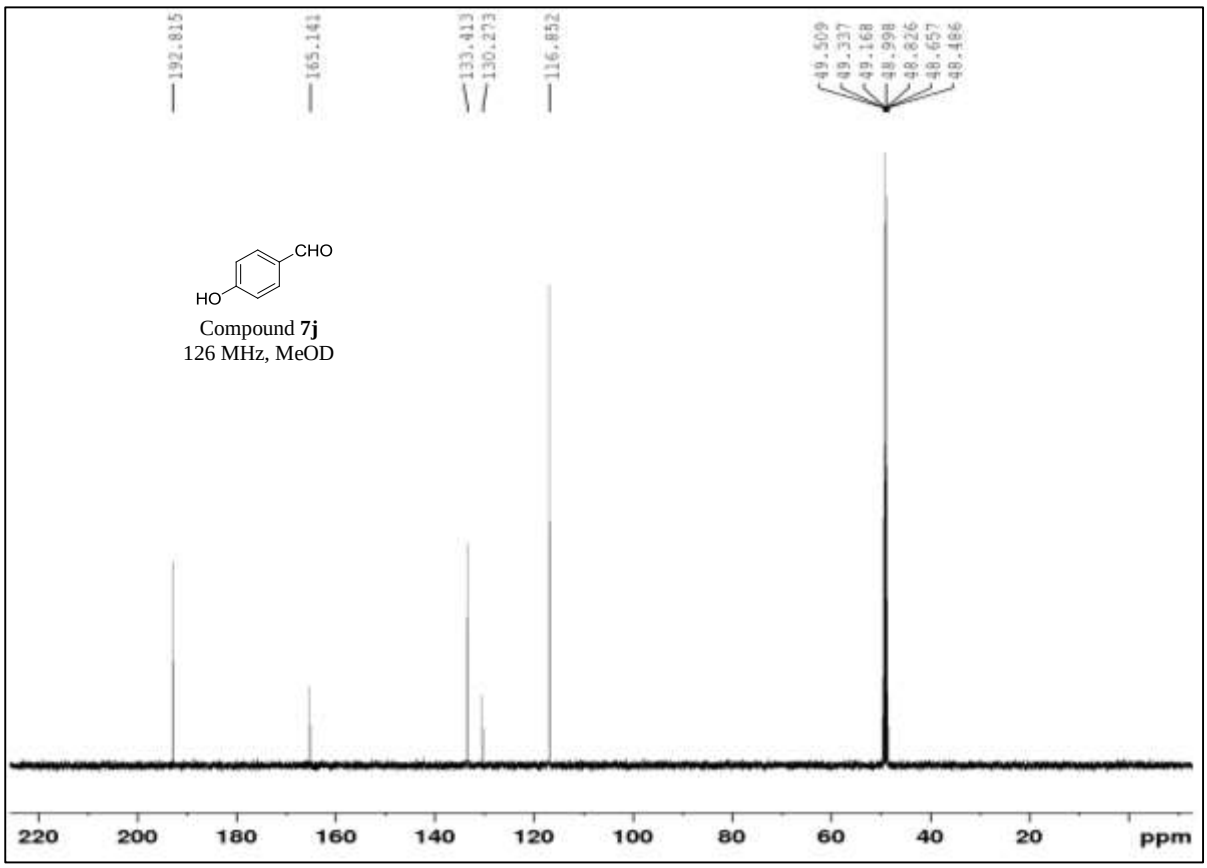
Compound **7i**
300 MHz, $CDCl_3$



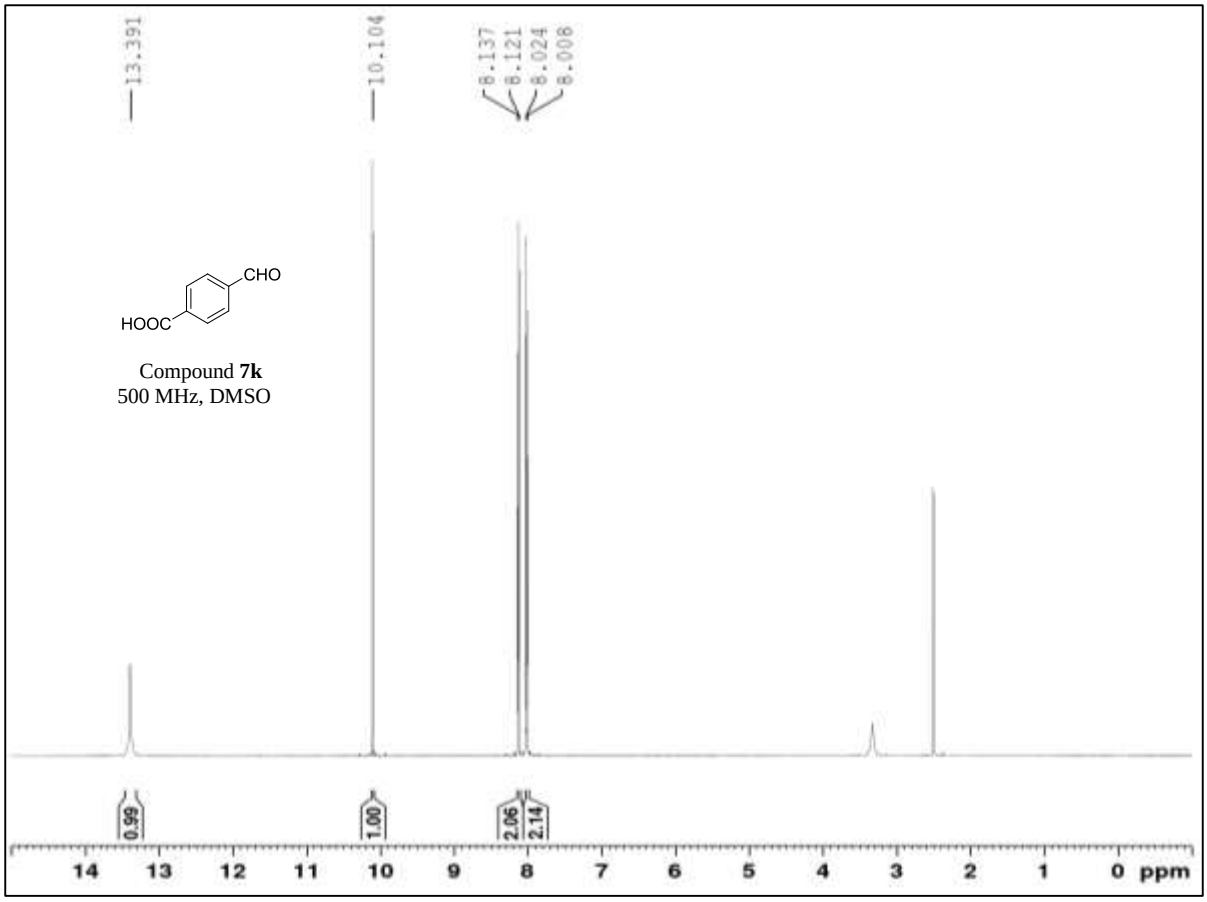
Compound **7i**
75 MHz, $CDCl_3$



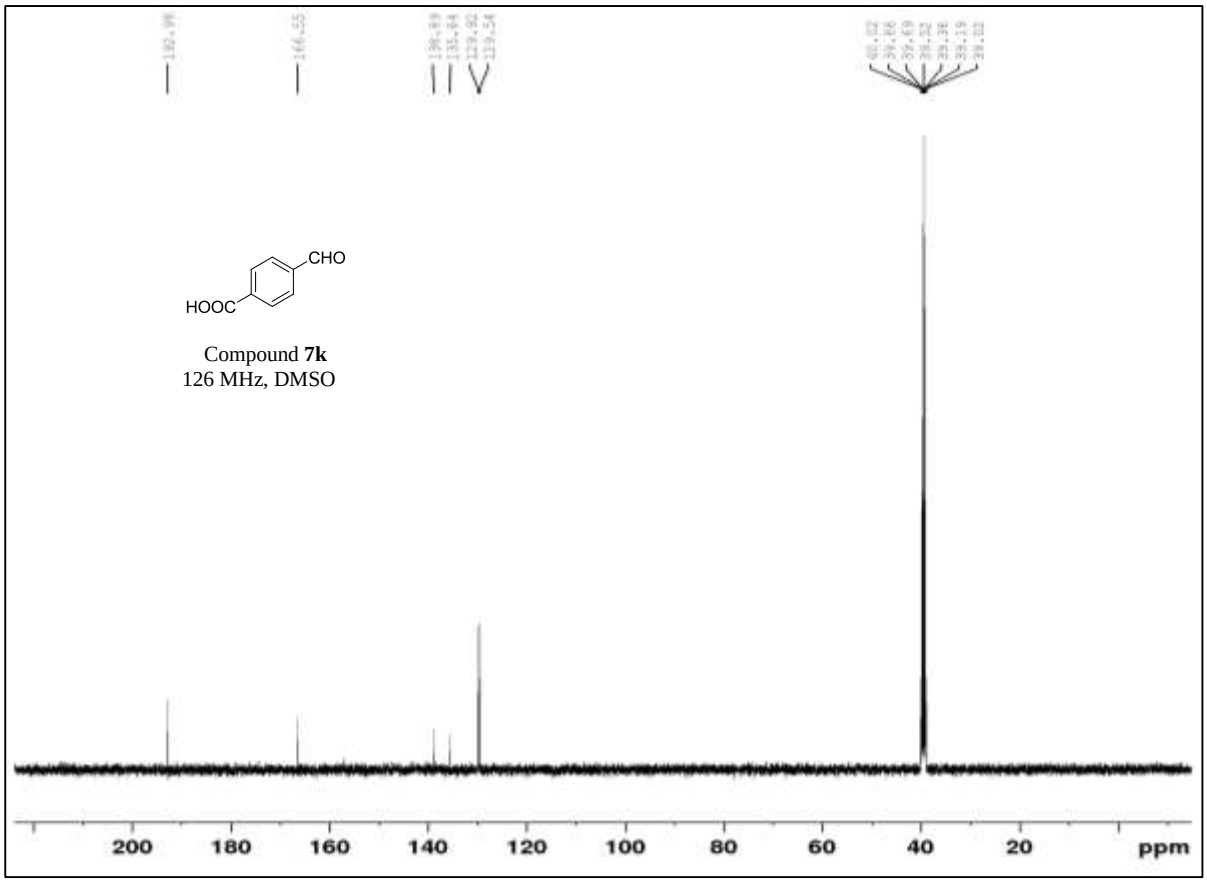
Compound **7j**
500 MHz, MeOD



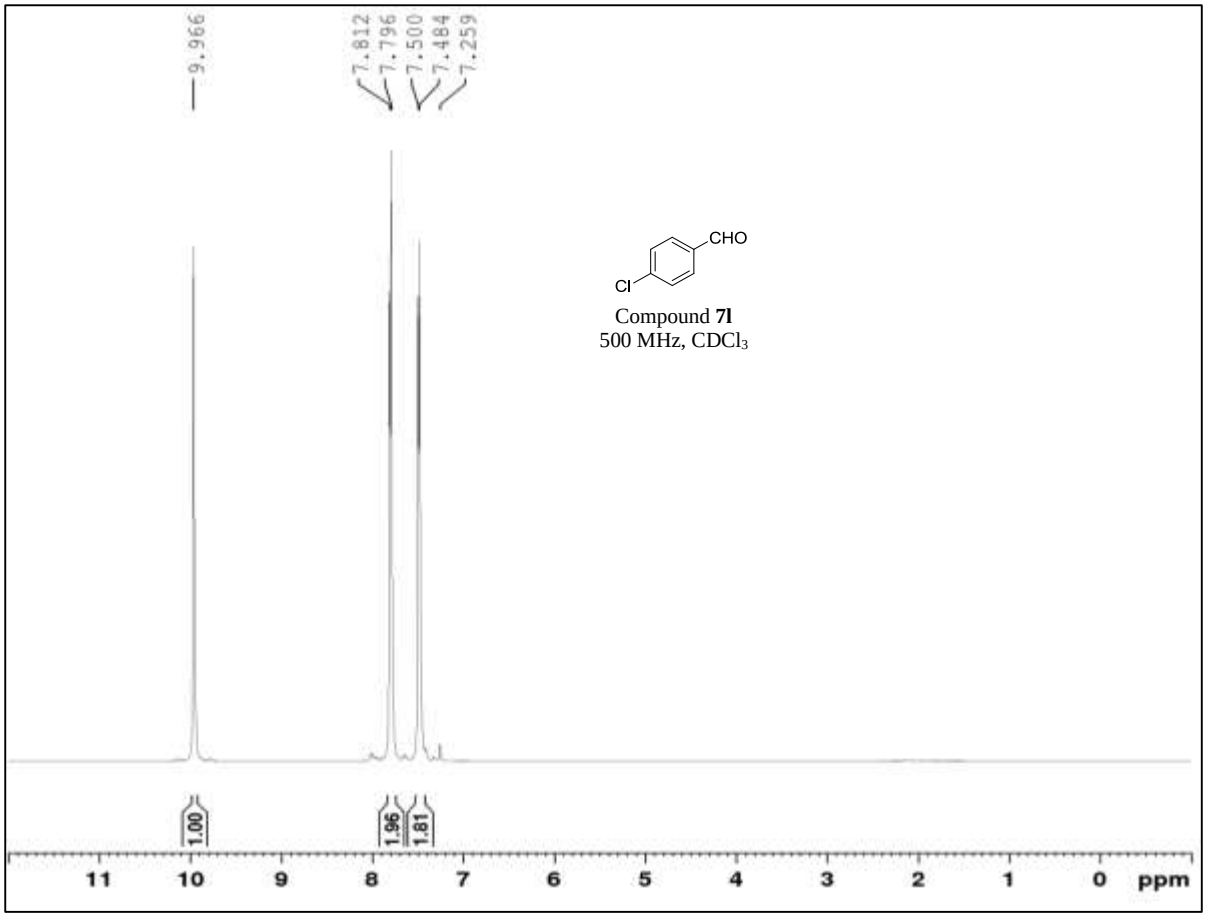
Compound **7j**
126 MHz, MeOD



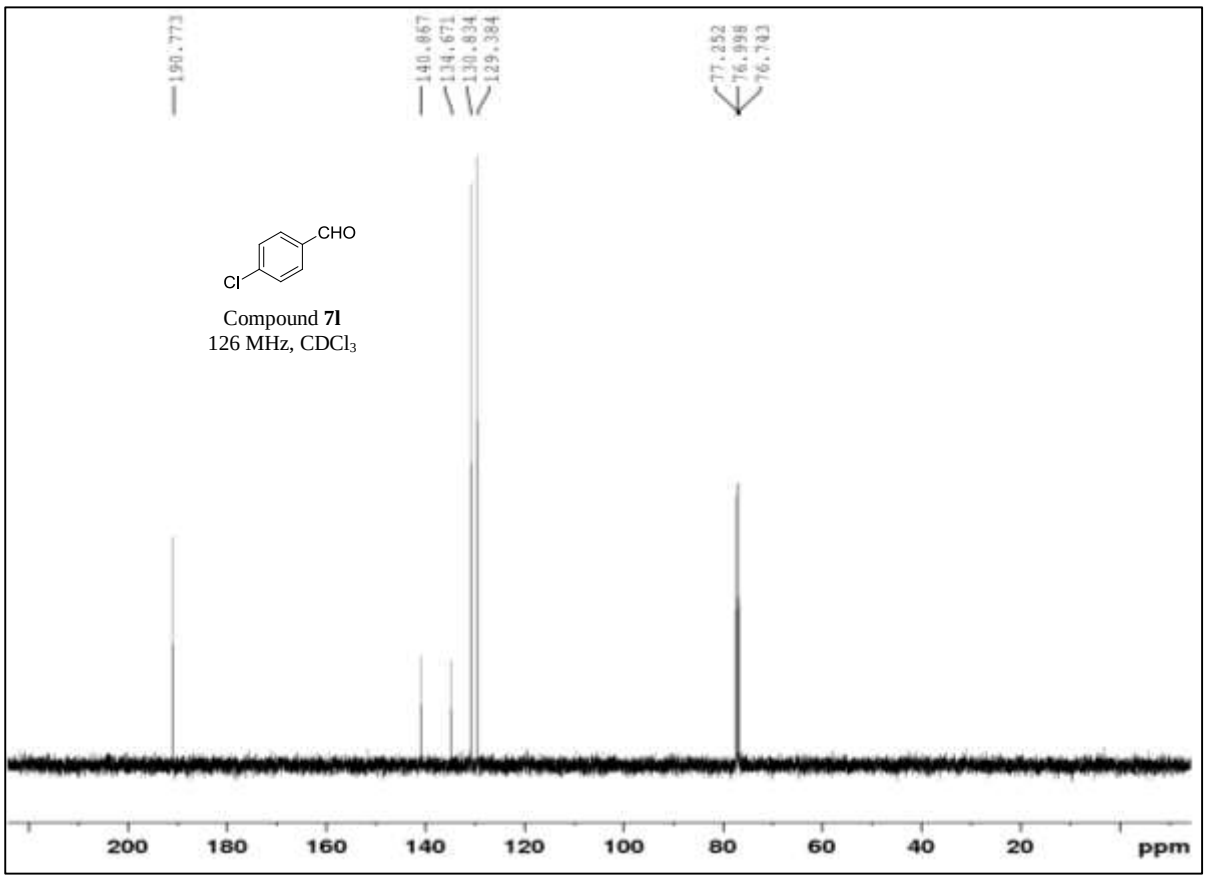
Compound **7k**
500 MHz, DMSO



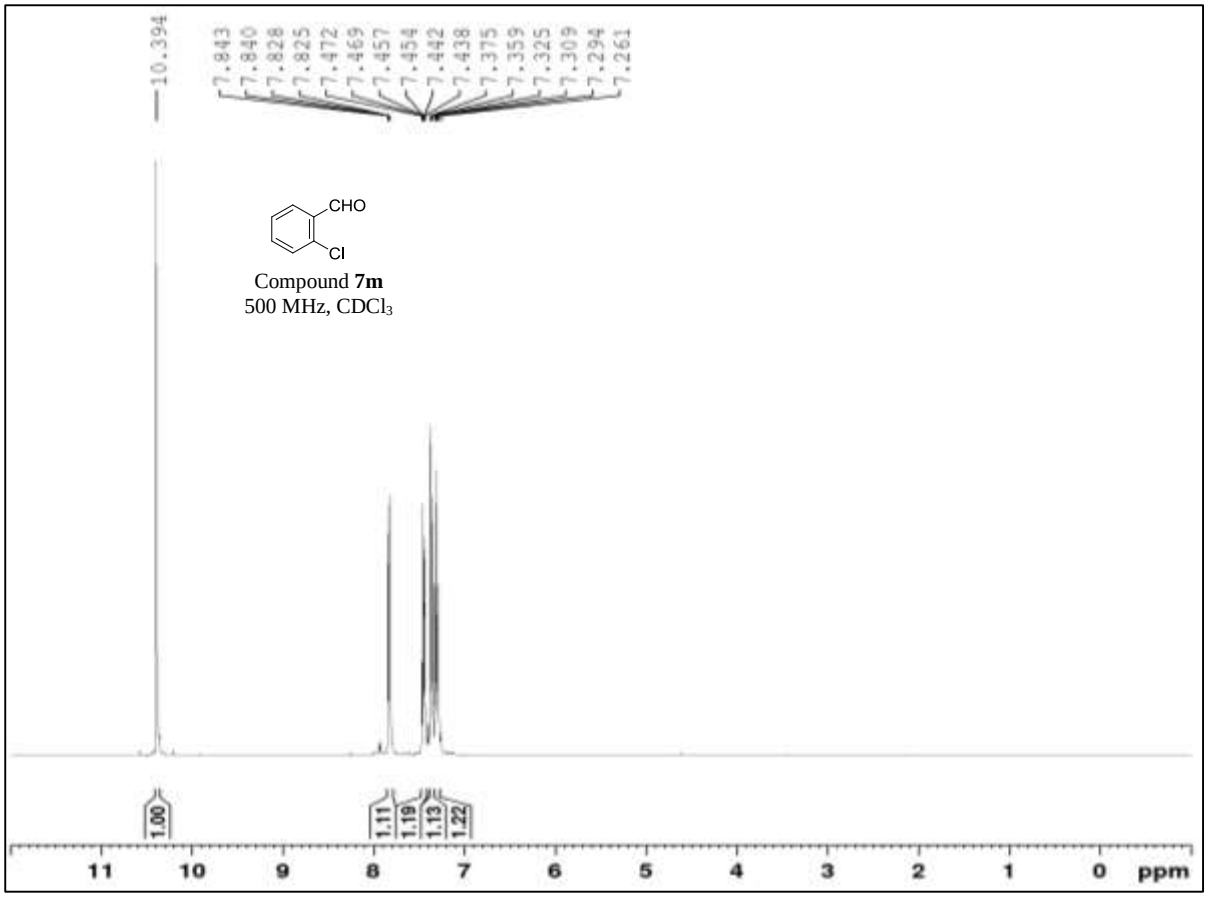
Compound **7k**
126 MHz, DMSO



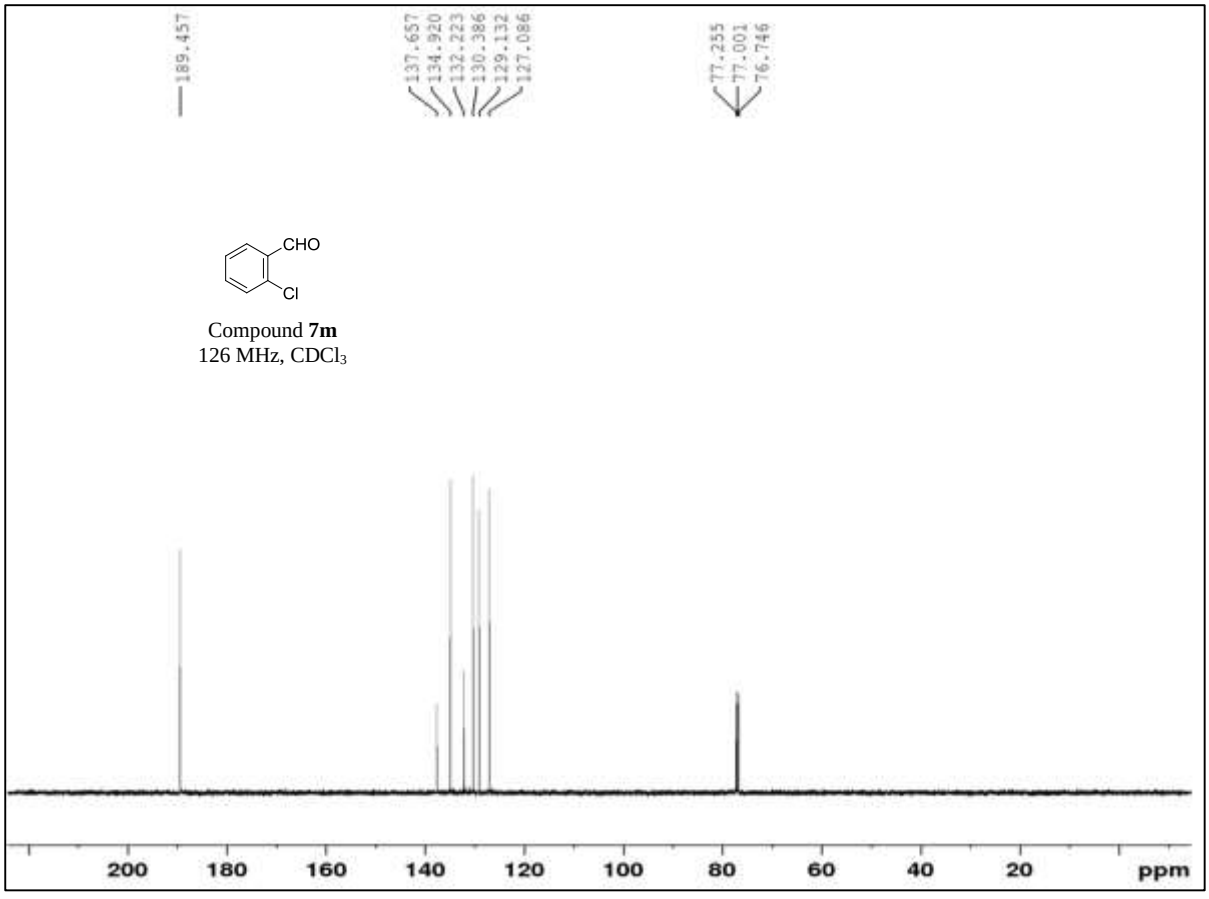
Compound **7l**
500 MHz, $CDCl_3$



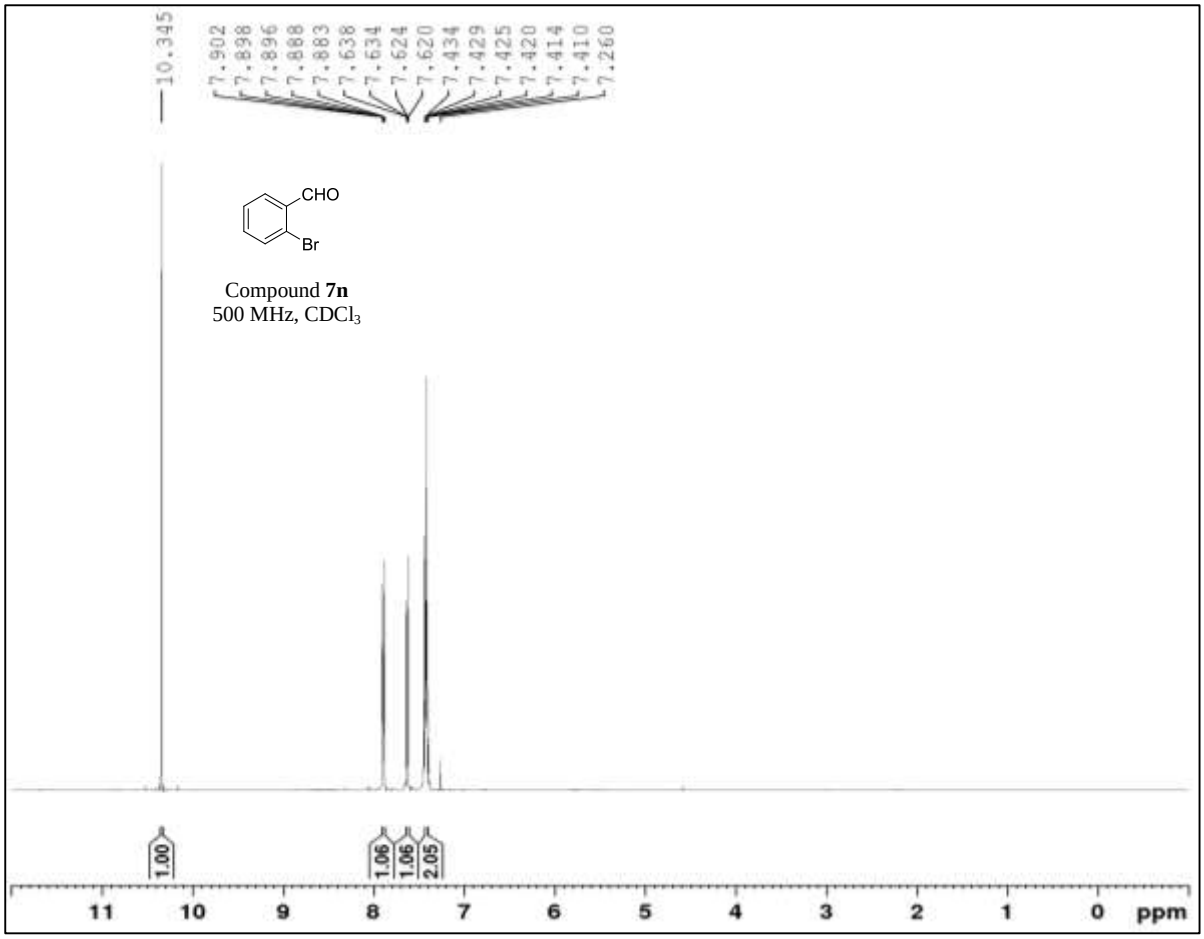
Compound **7l**
126 MHz, $CDCl_3$



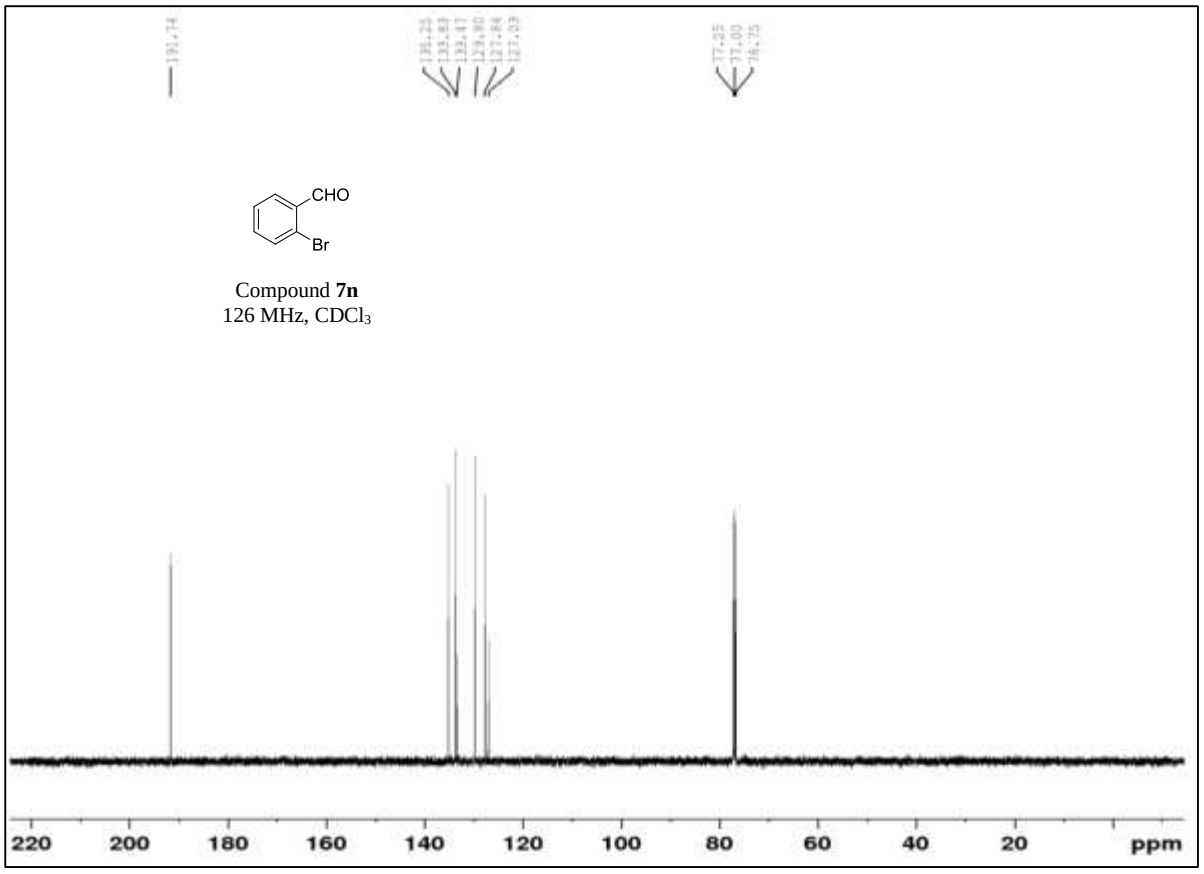
Compound **7m**
500 MHz, $CDCl_3$



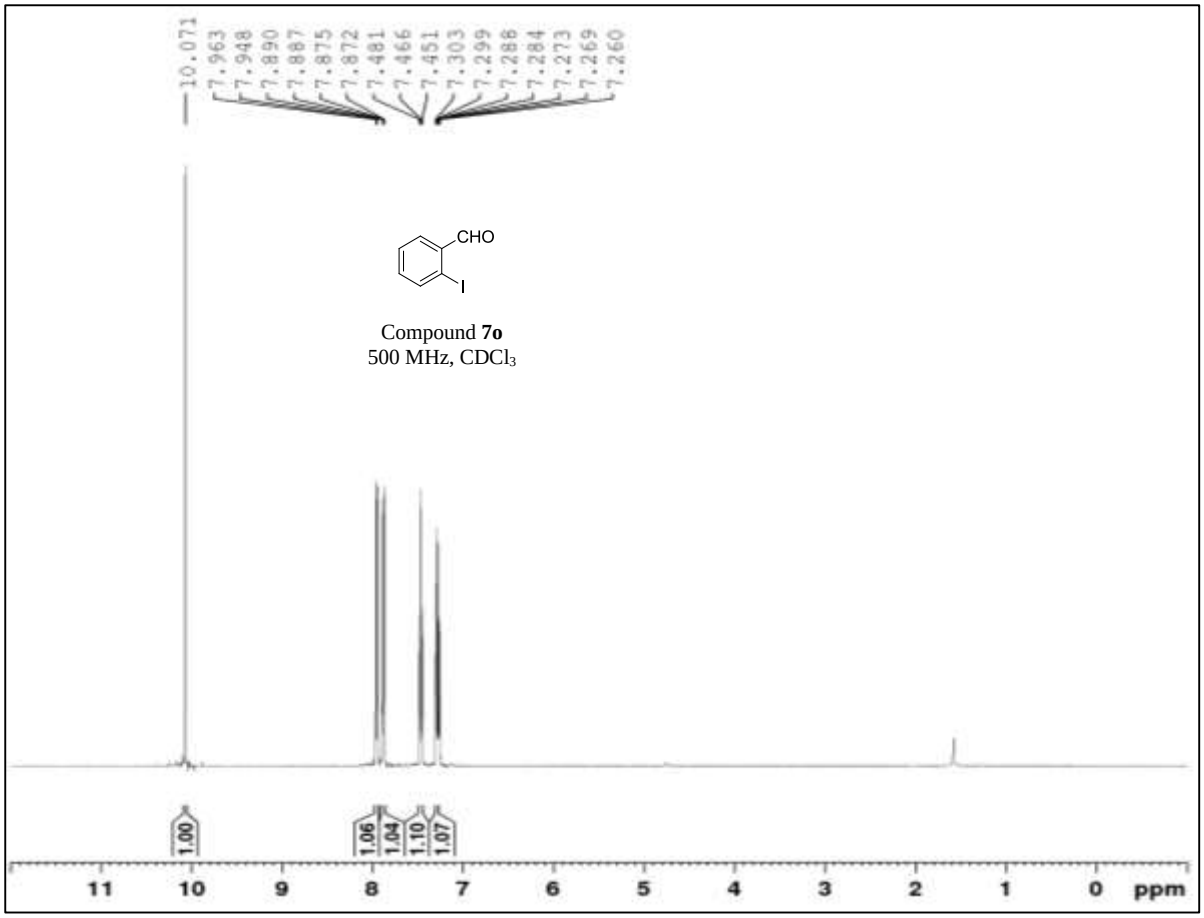
Compound **7m**
126 MHz, $CDCl_3$



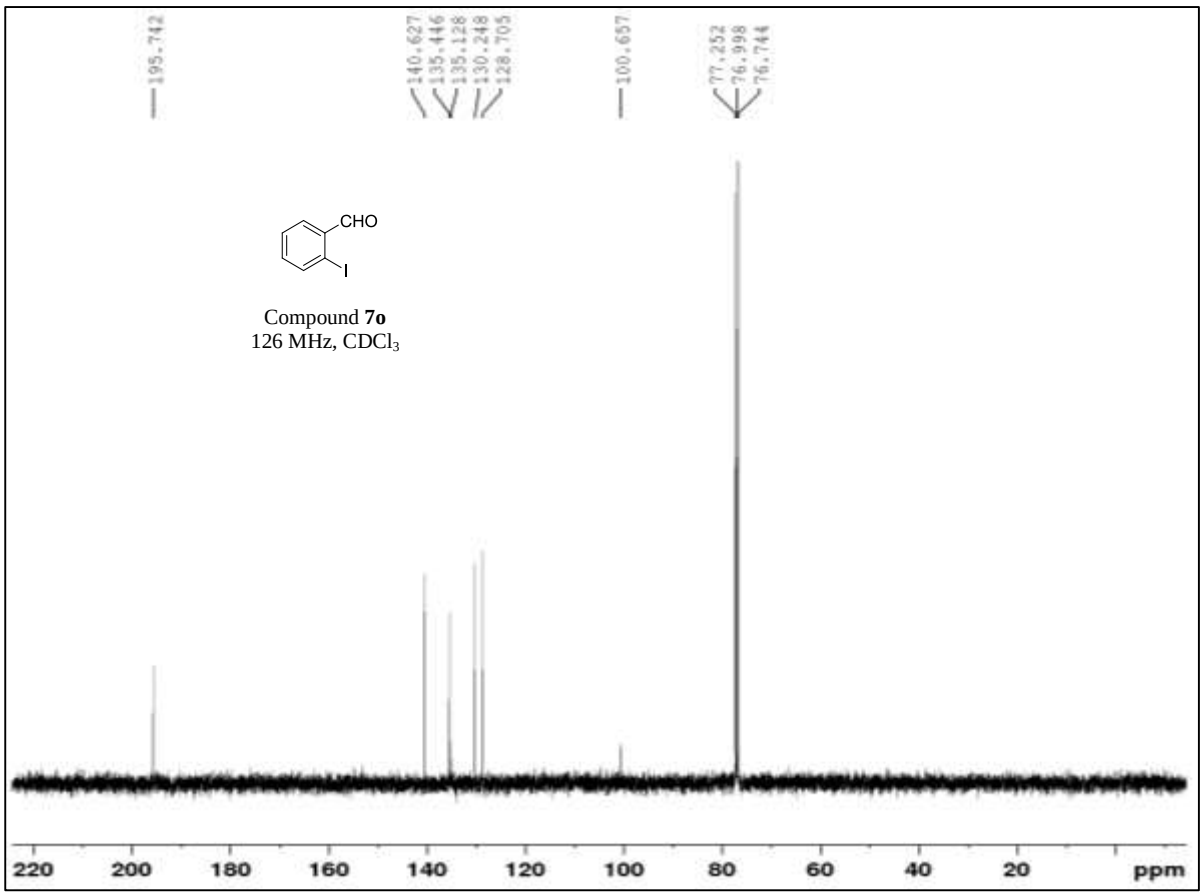
Compound **7n**
500 MHz, $CDCl_3$



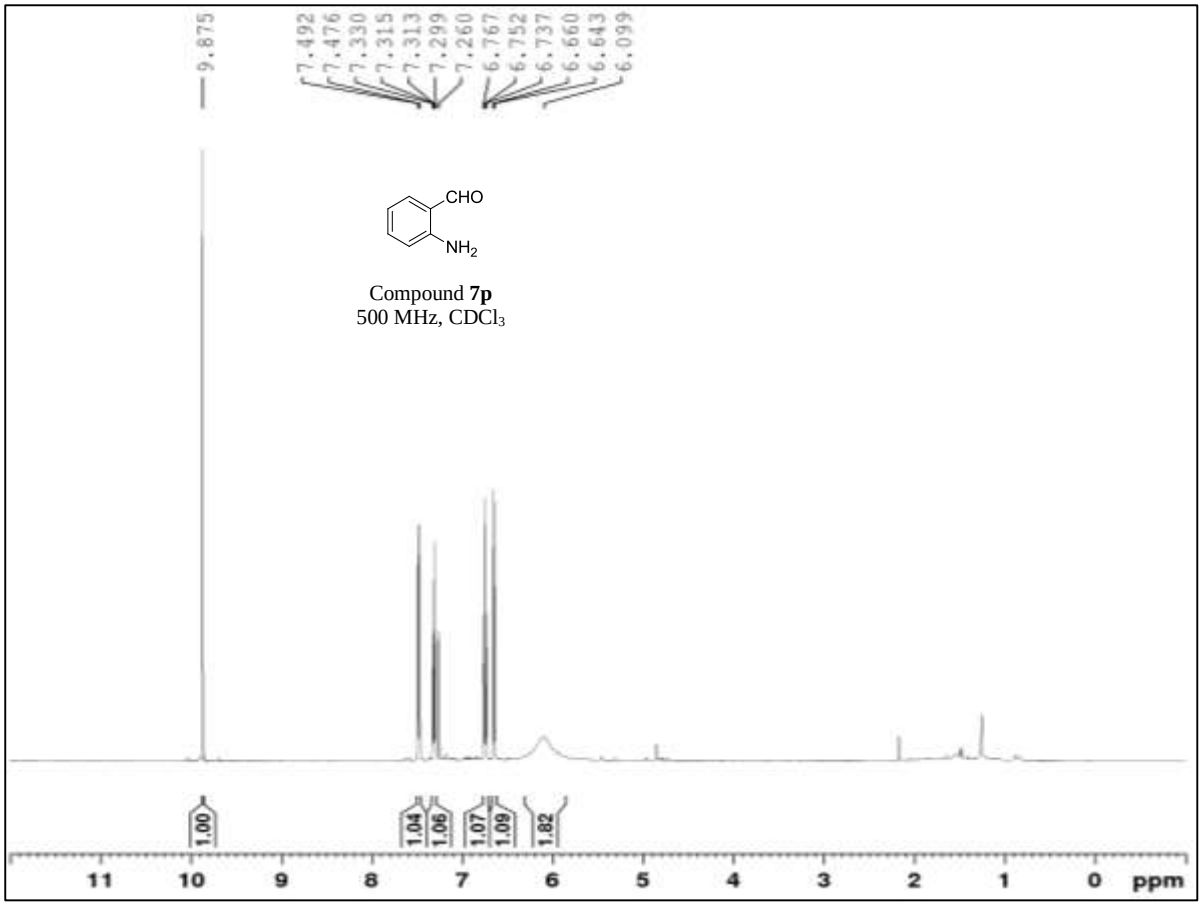
Compound **7n**
126 MHz, $CDCl_3$



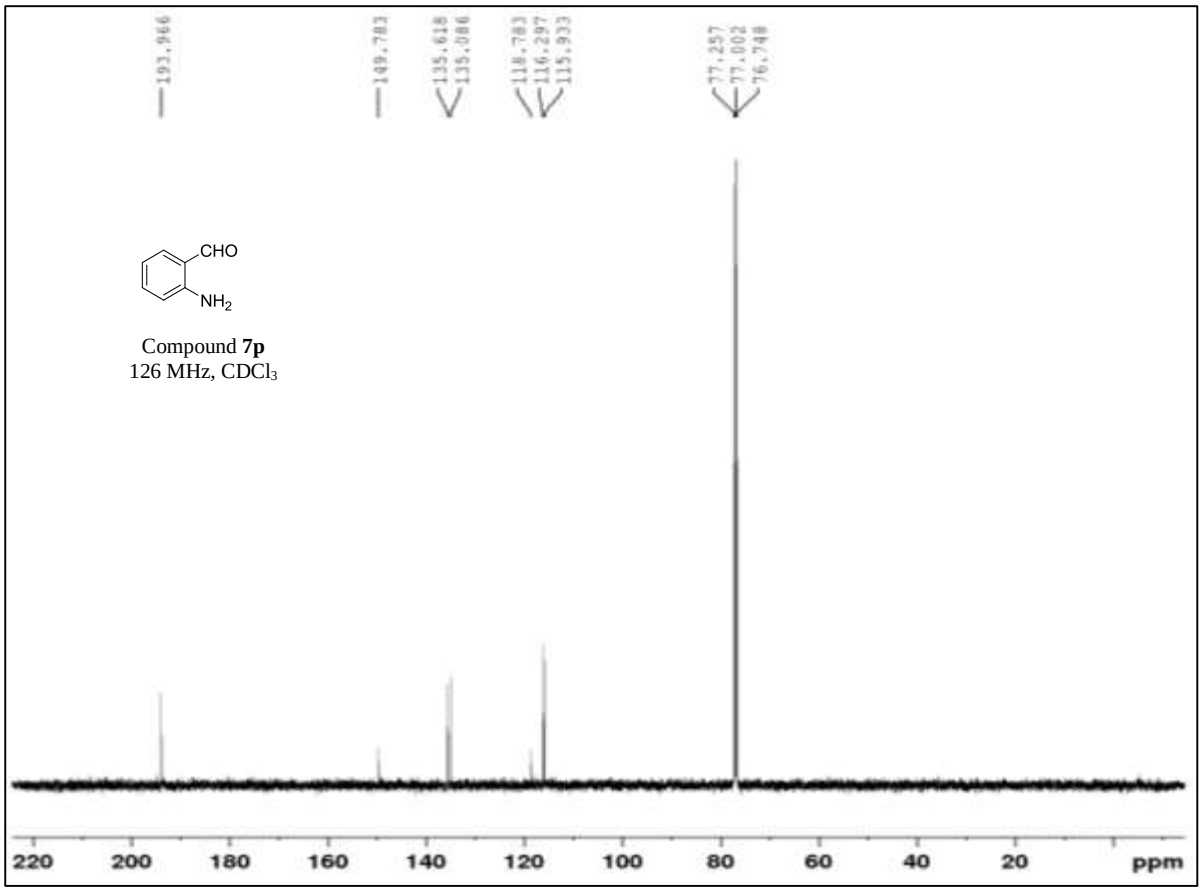
Compound **7o**
500 MHz, $CDCl_3$



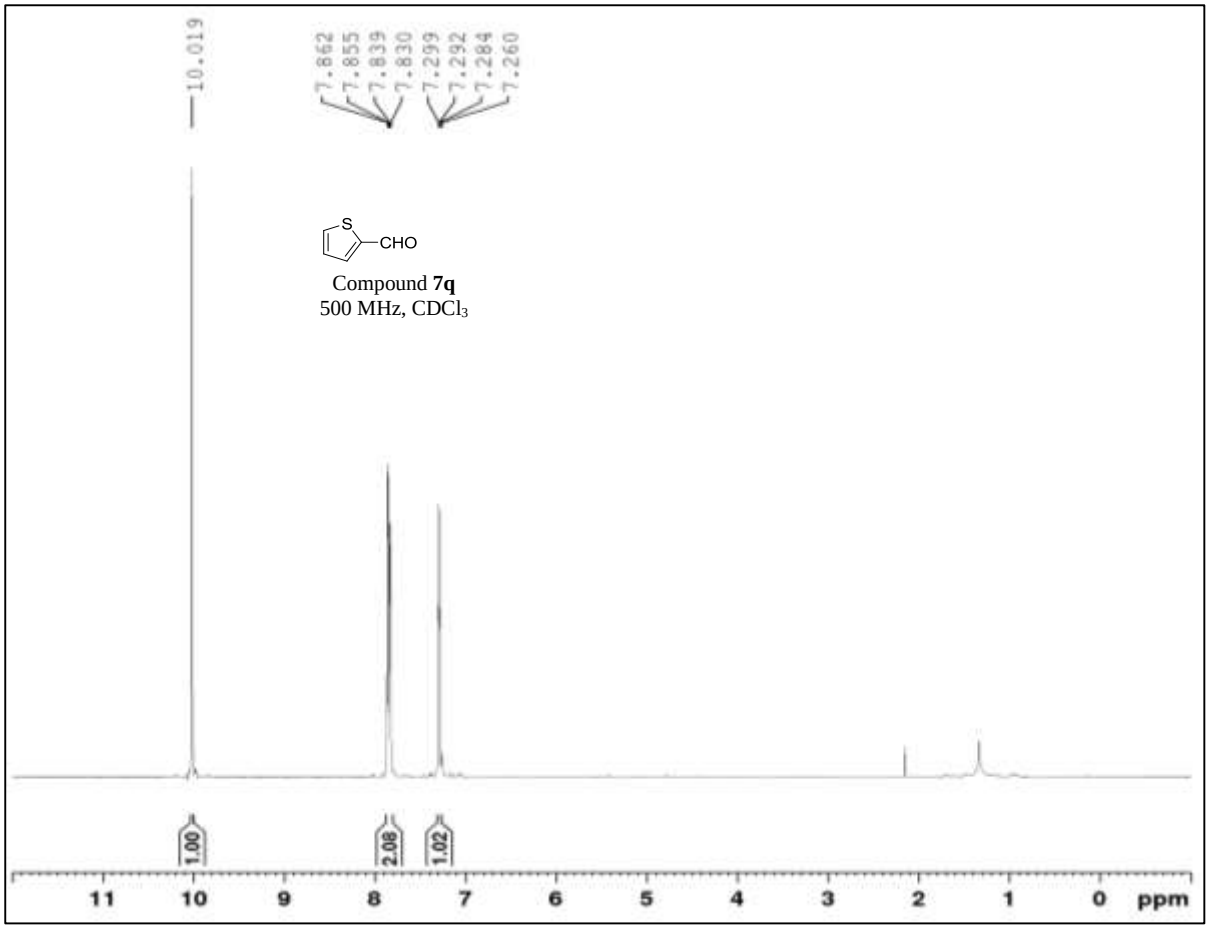
Compound **7o**
126 MHz, $CDCl_3$



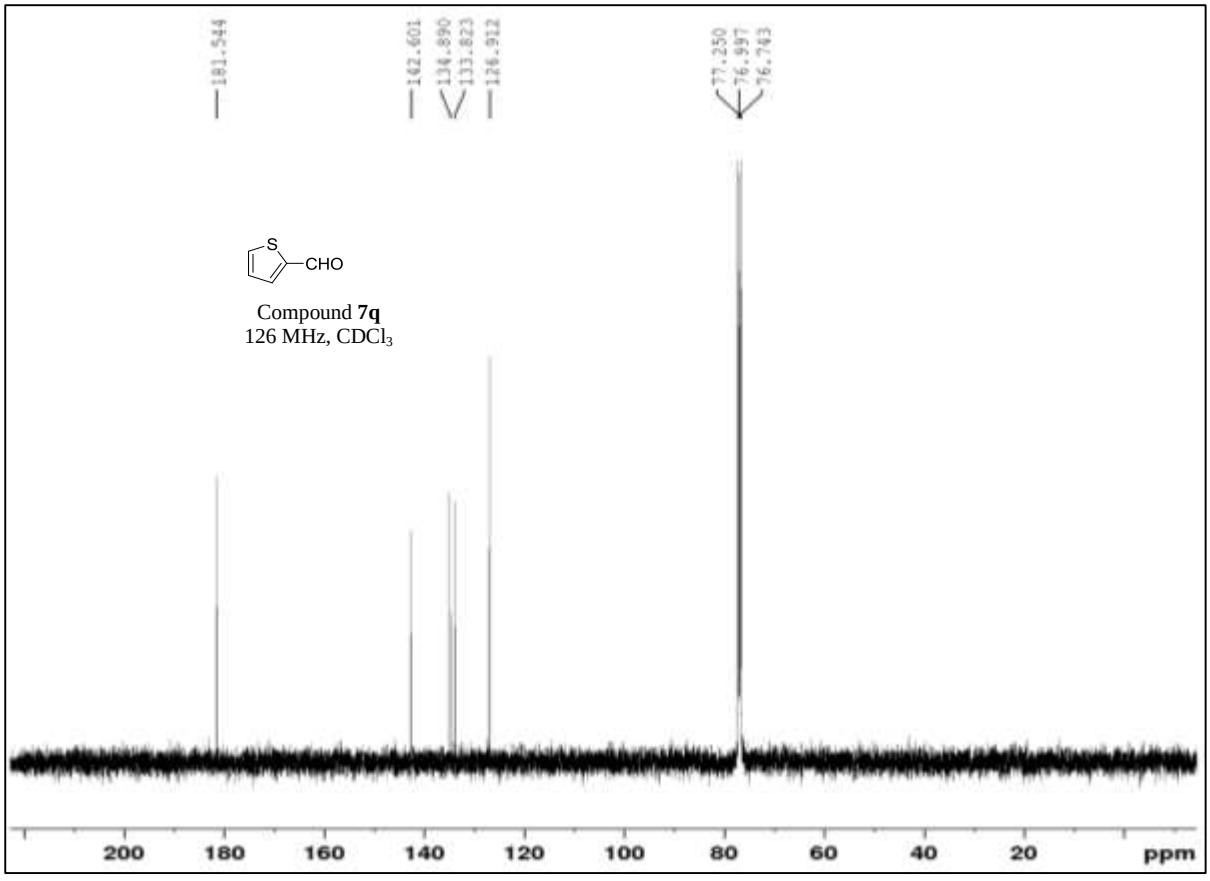
Compound **7p**
500 MHz, $CDCl_3$



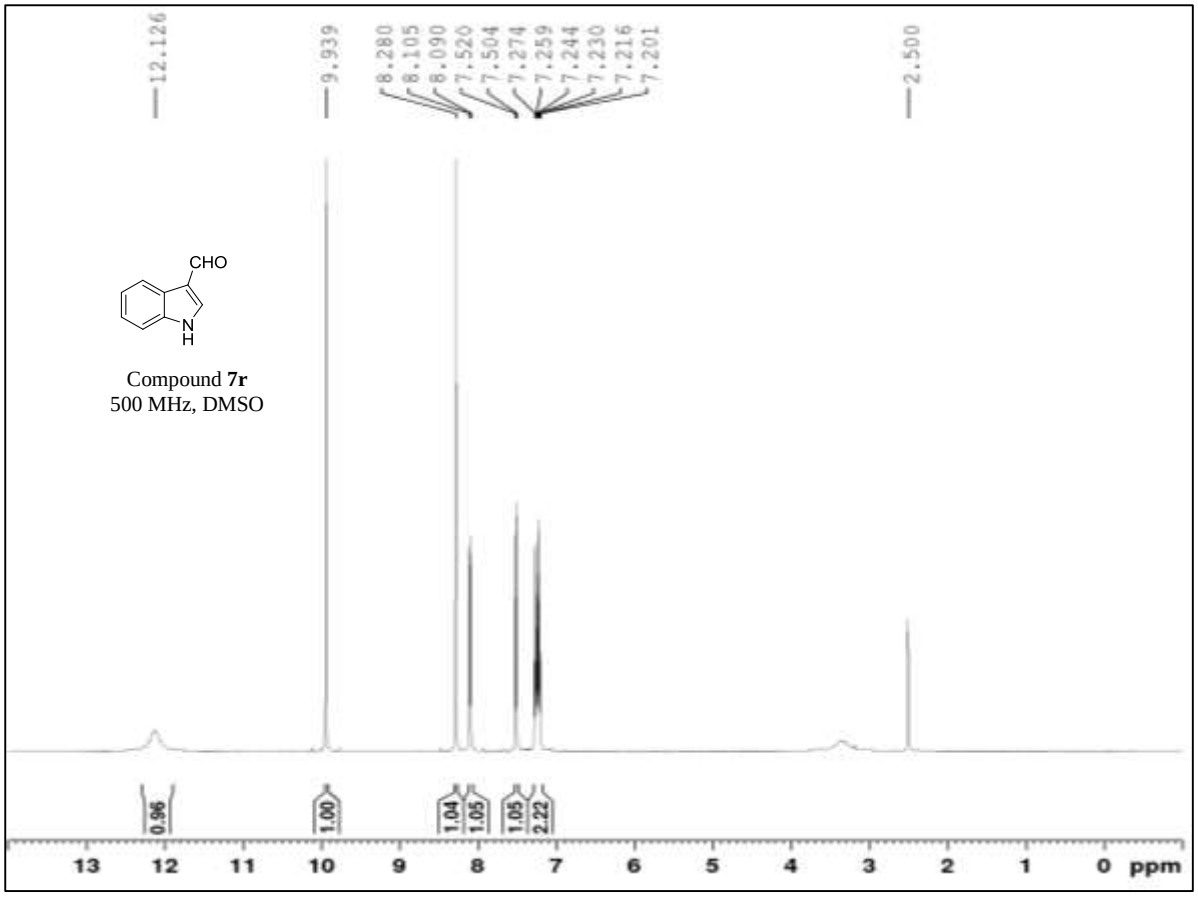
Compound **7p**
126 MHz, $CDCl_3$



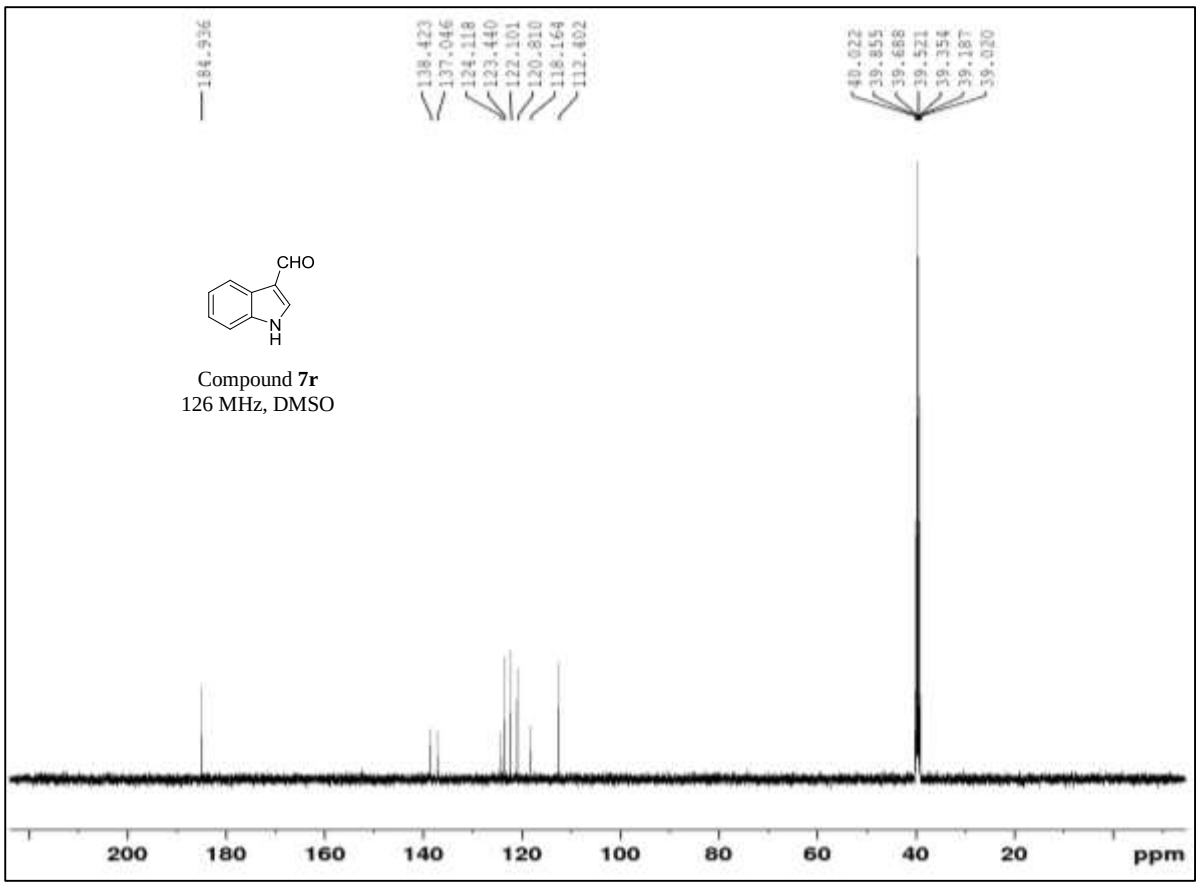
Compound **7q**
500 MHz, $CDCl_3$



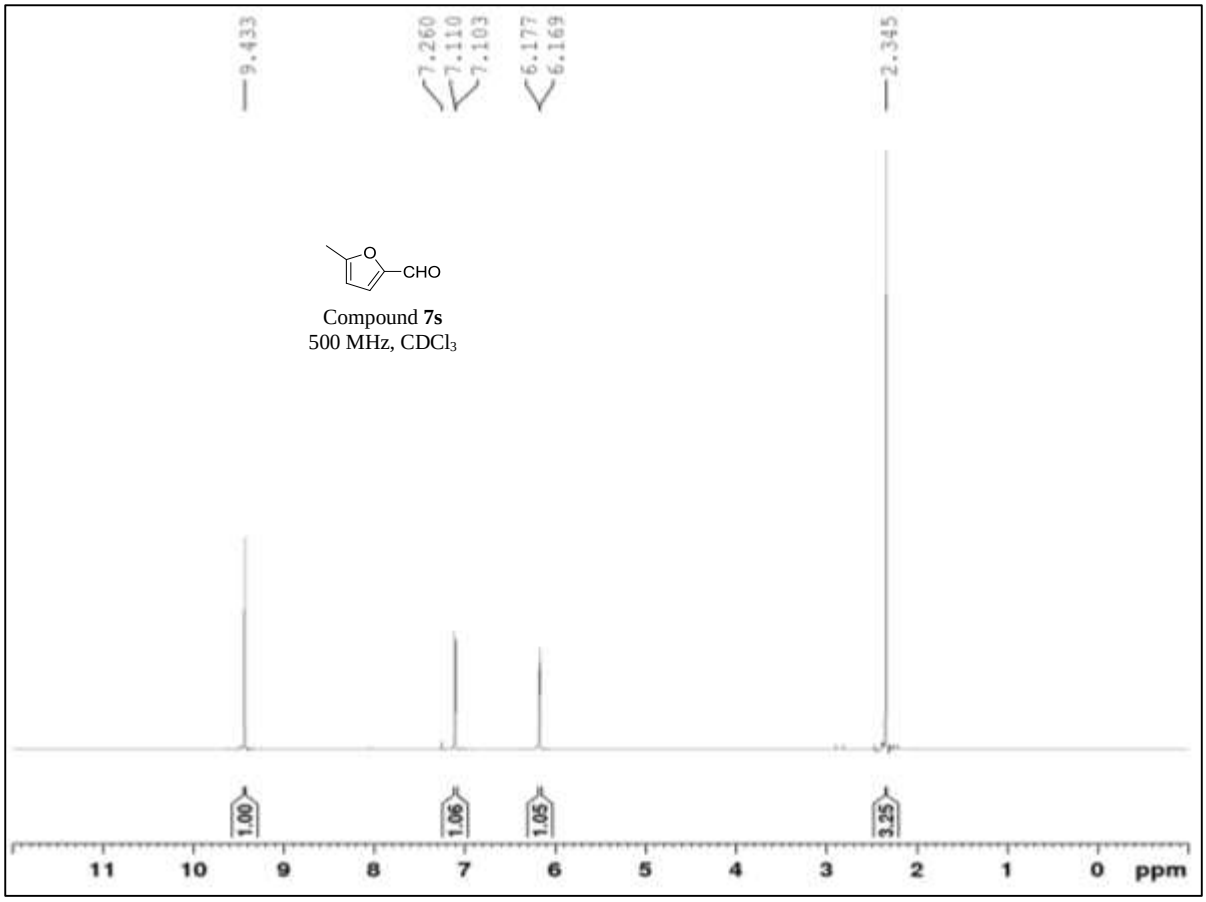
Compound **7q**
126 MHz, $CDCl_3$



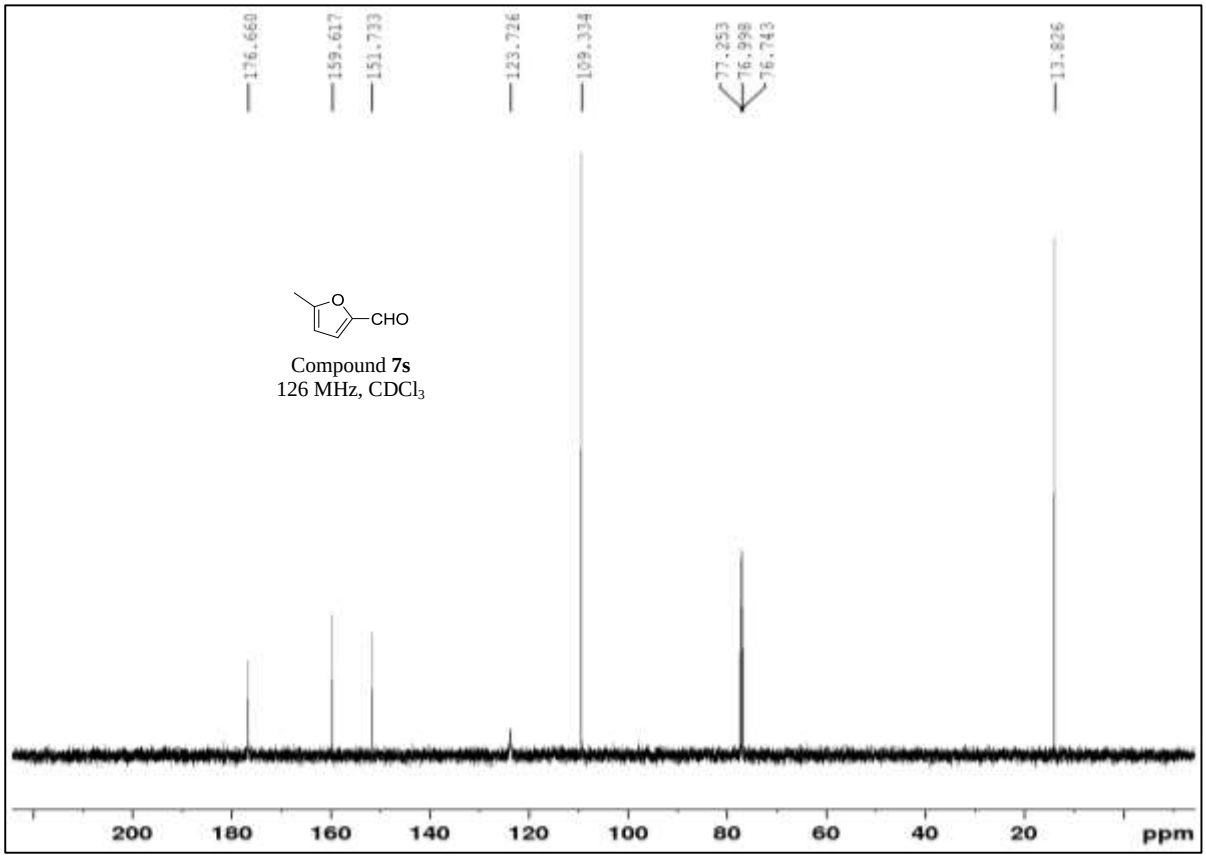
Compound **7r**
500 MHz, DMSO



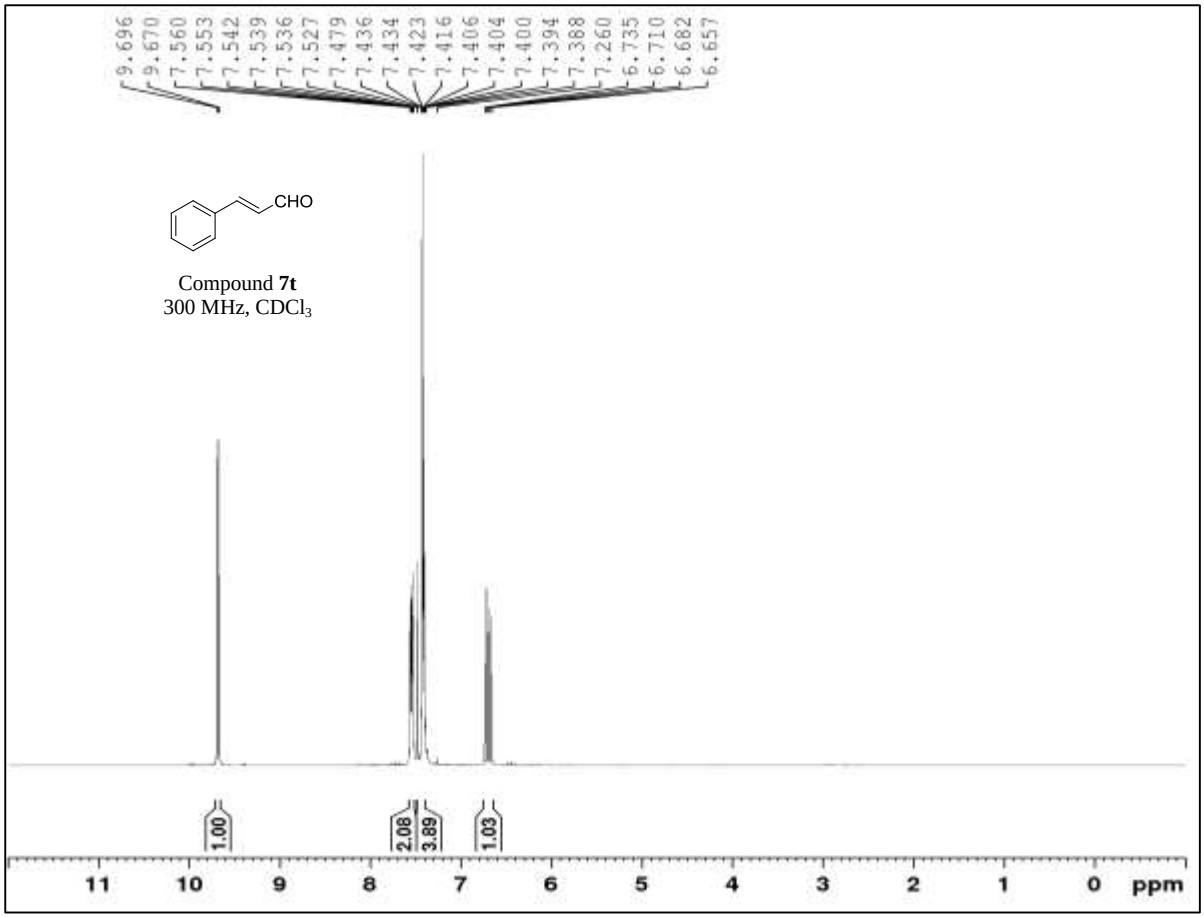
Compound **7r**
126 MHz, DMSO



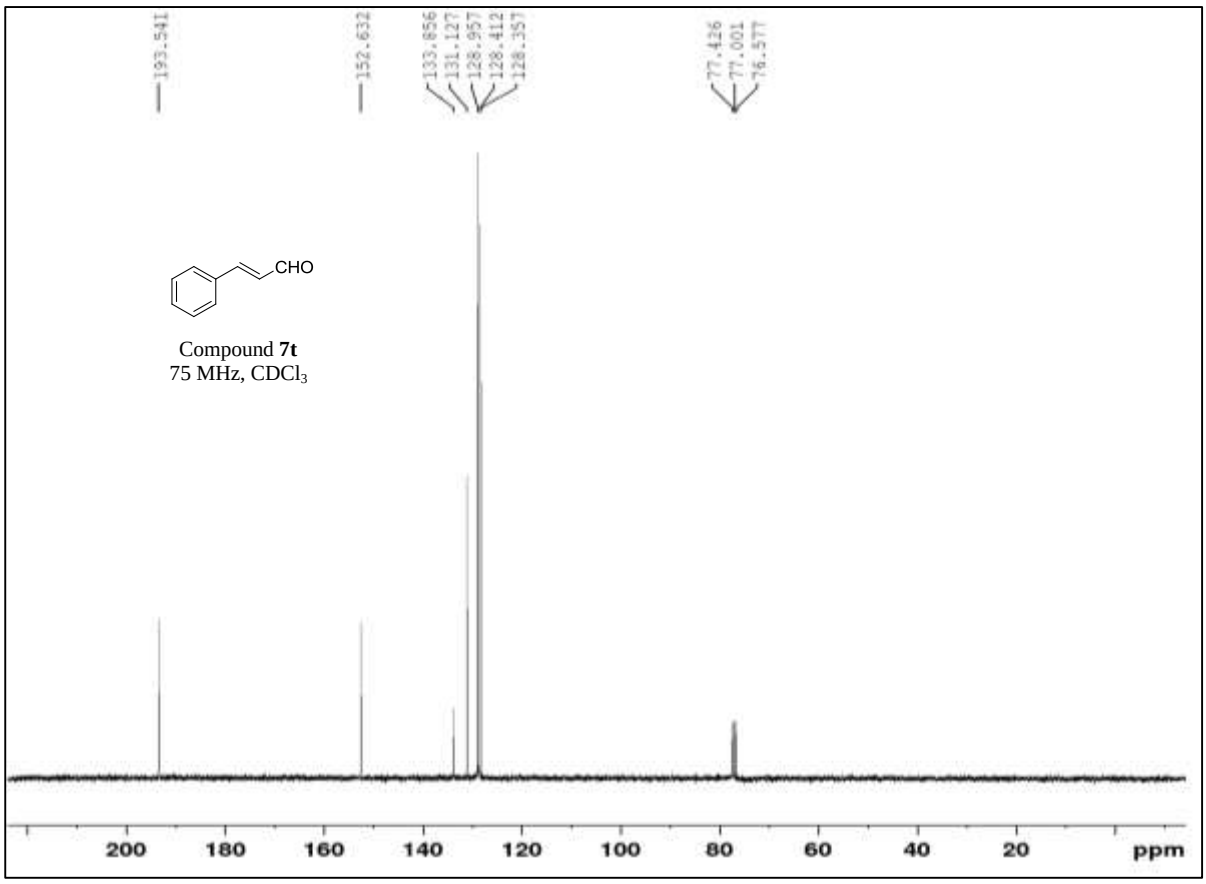
Compound **7s**
500 MHz, $CDCl_3$



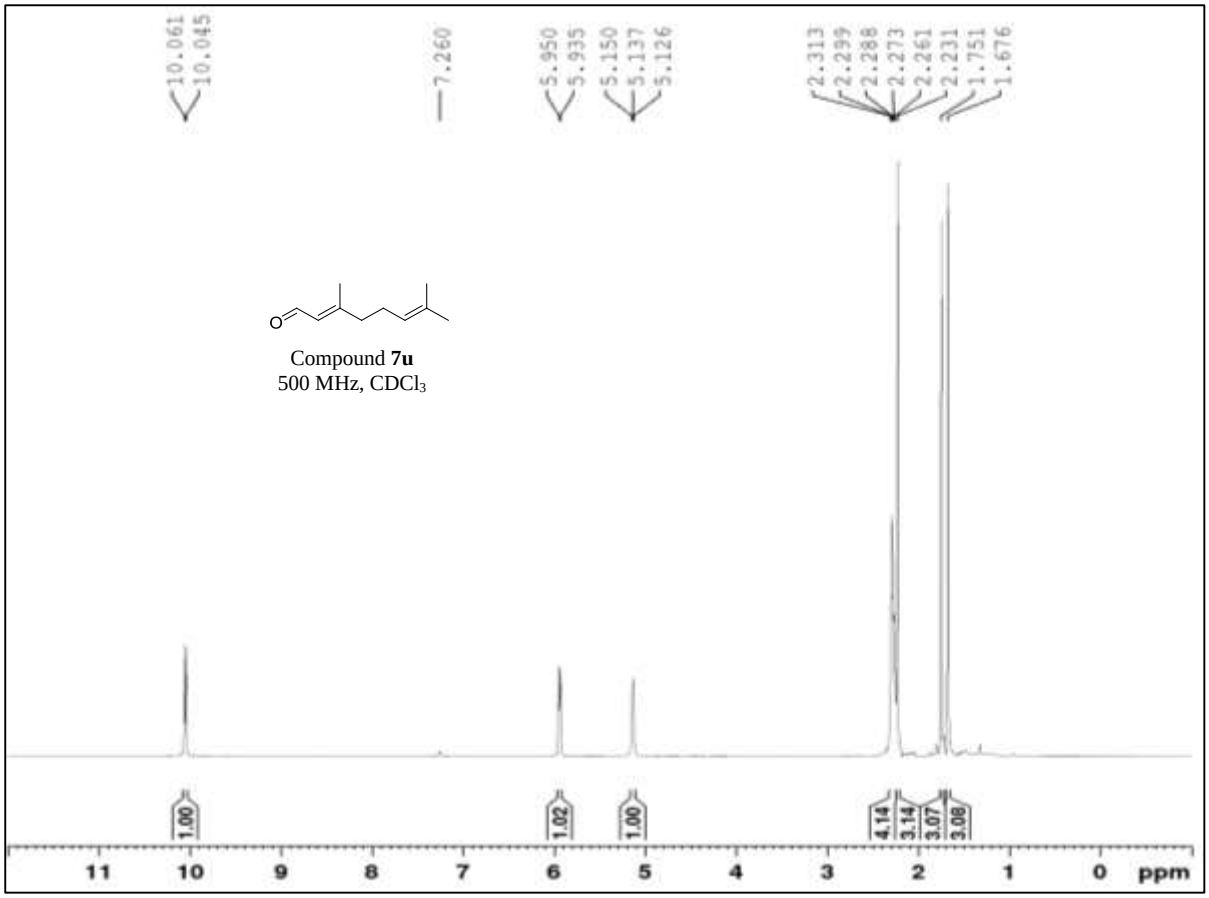
Compound **7s**
126 MHz, $CDCl_3$



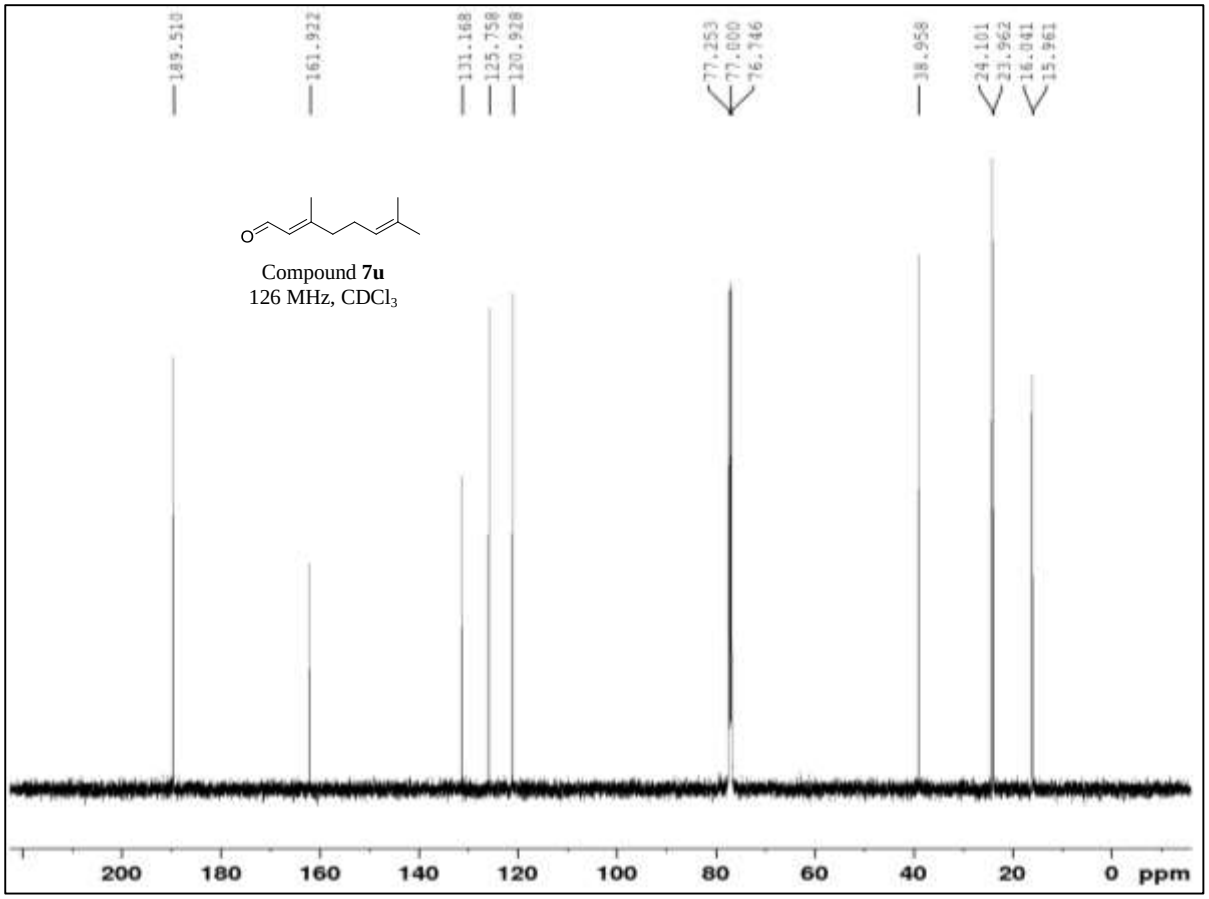
Compound **7t**
300 MHz, $CDCl_3$



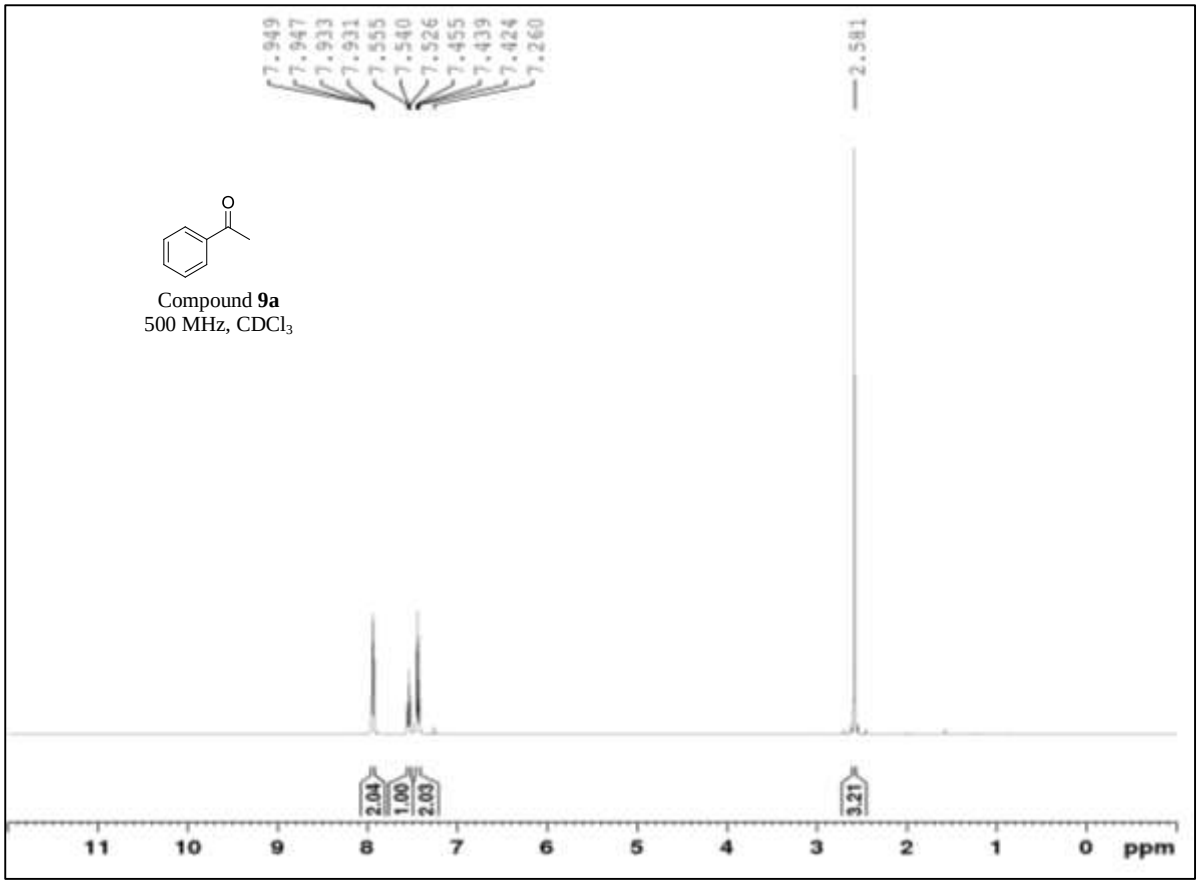
Compound **7t**
75 MHz, $CDCl_3$



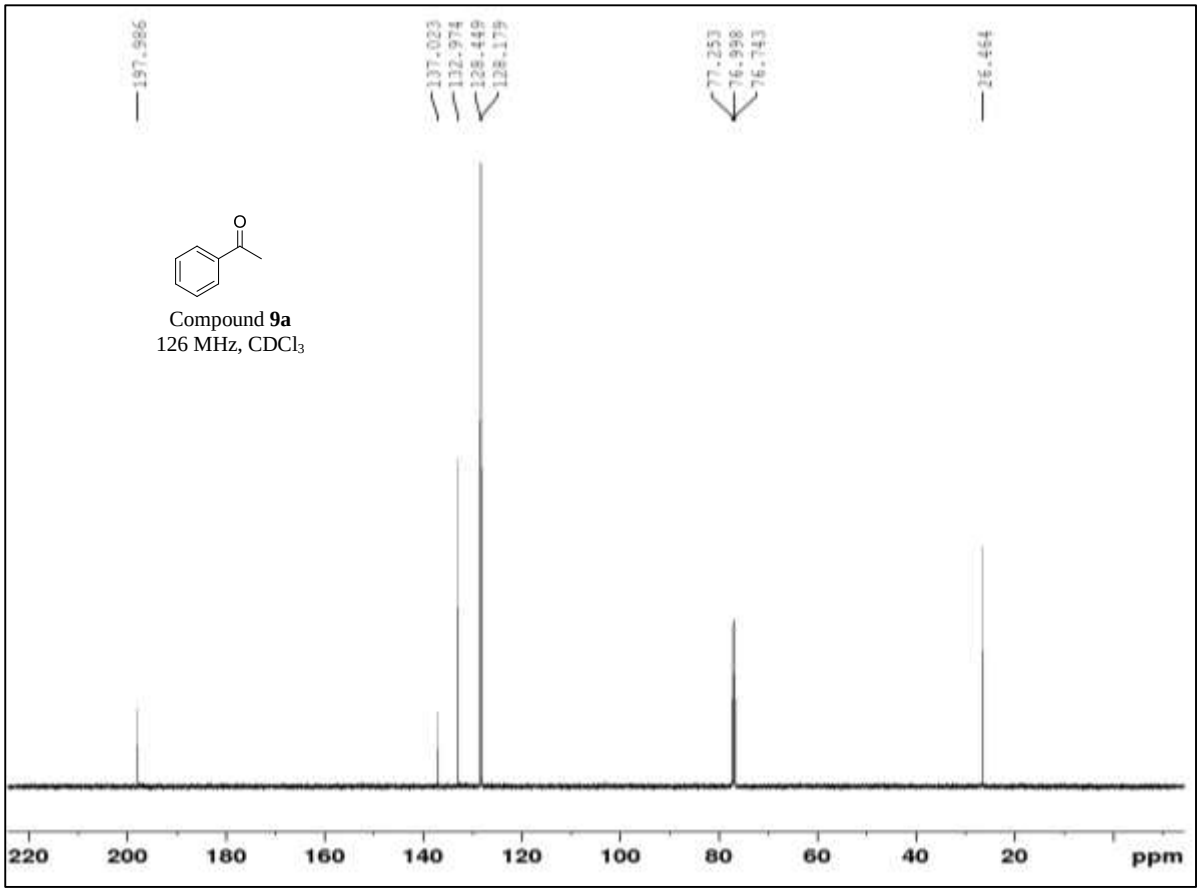
Compound **7u**
500 MHz, $CDCl_3$



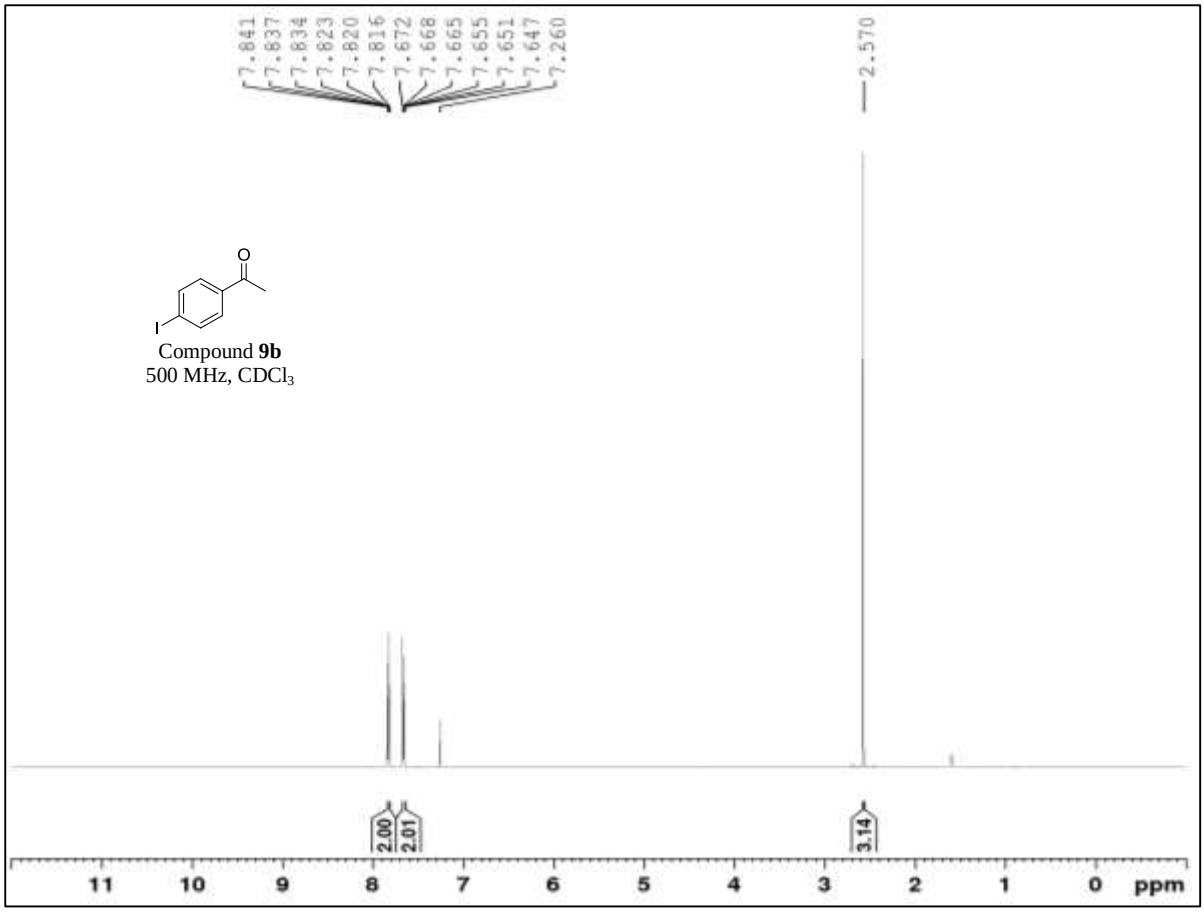
Compound **7u**
126 MHz, $CDCl_3$



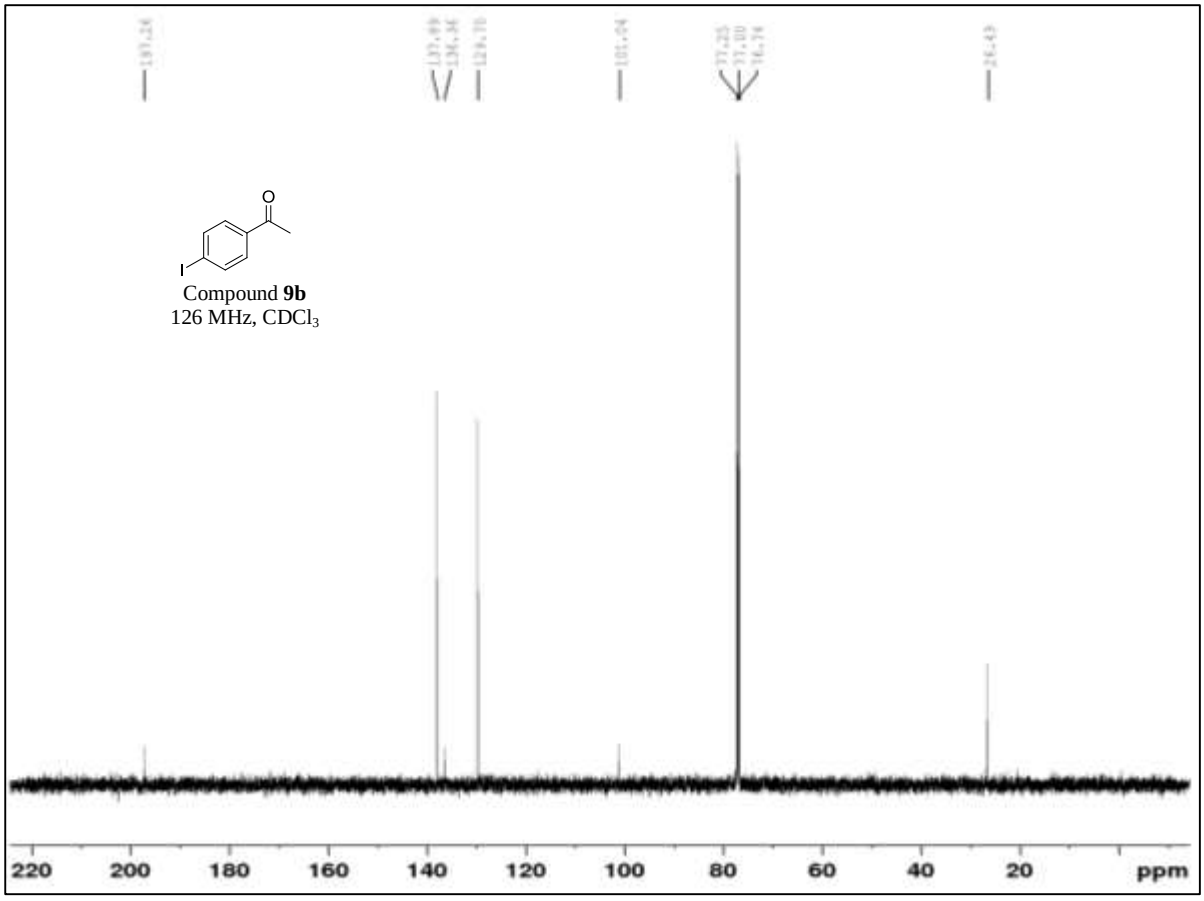
Compound **9a**
500 MHz, $CDCl_3$



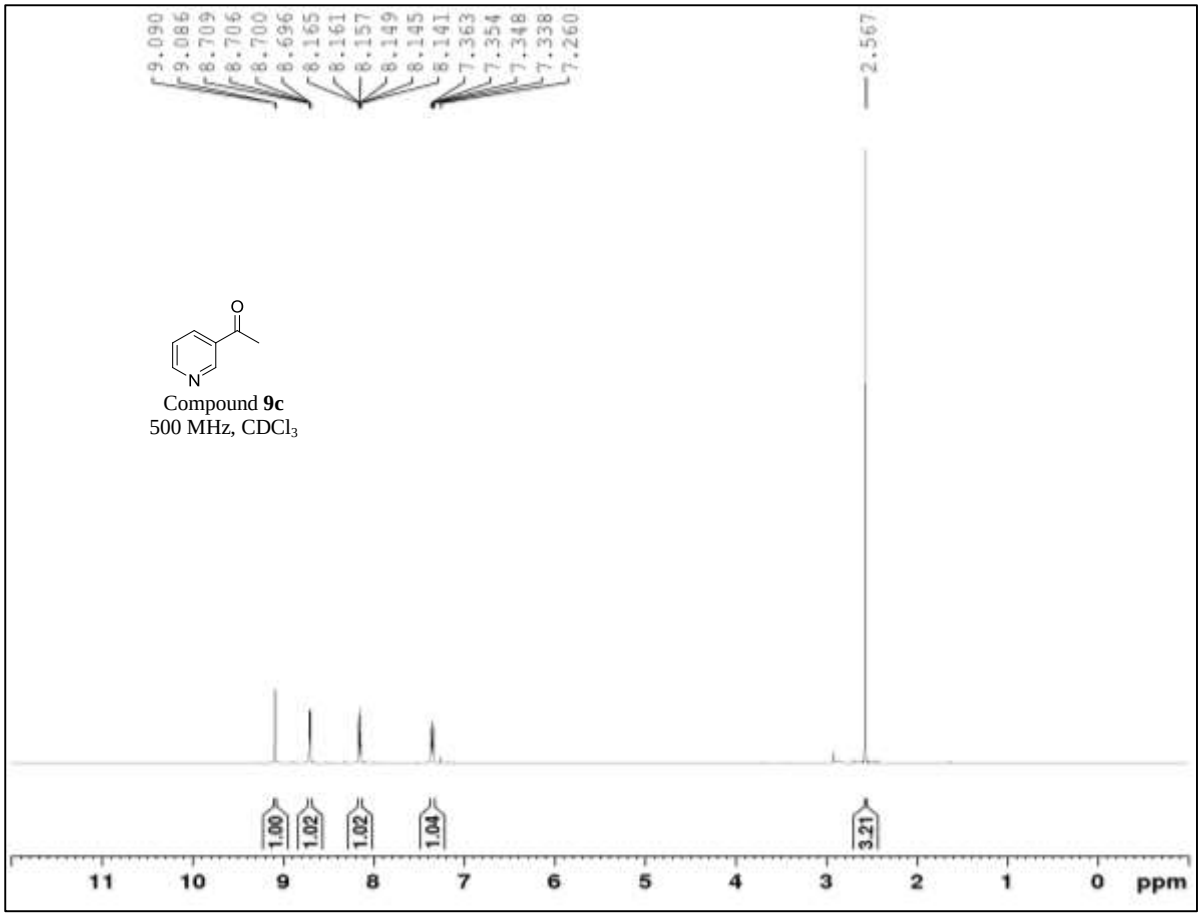
Compound **9a**
126 MHz, $CDCl_3$



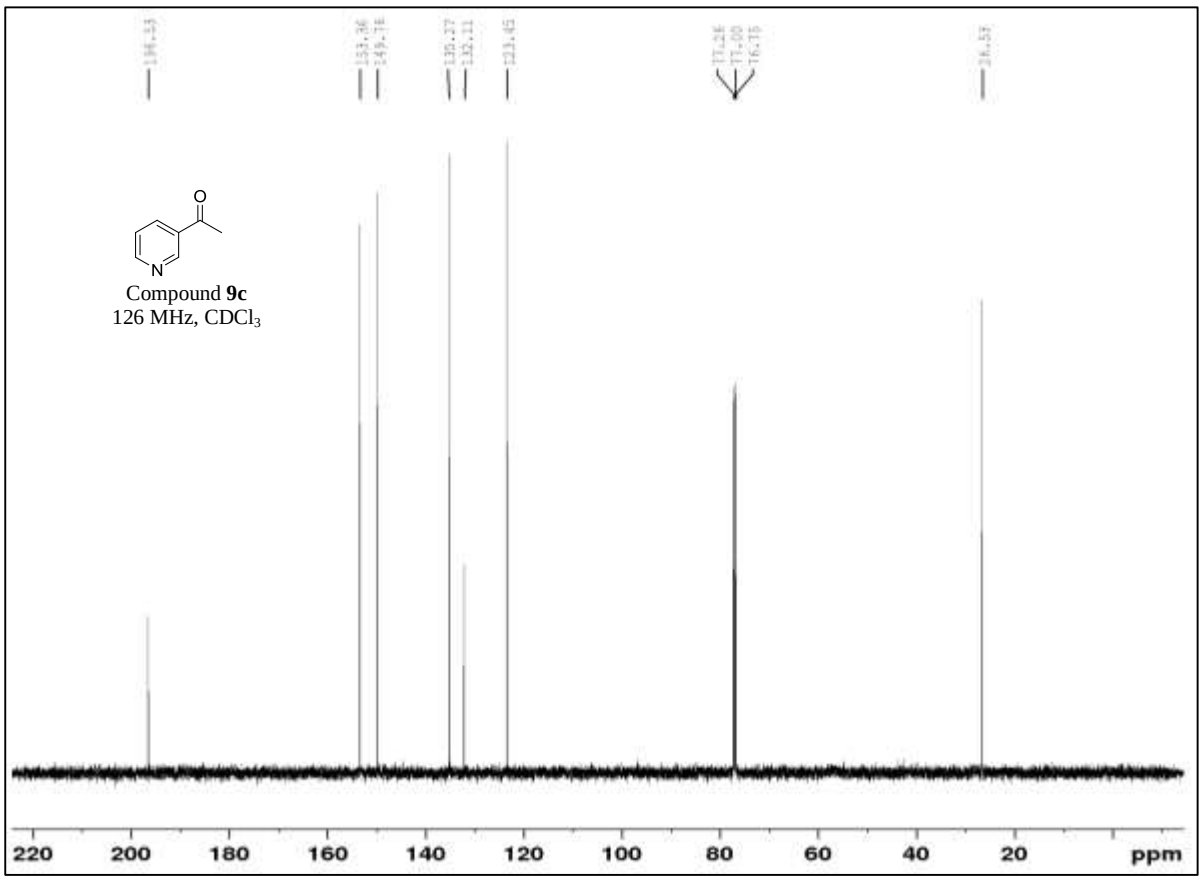
Compound **9b**
500 MHz, $CDCl_3$



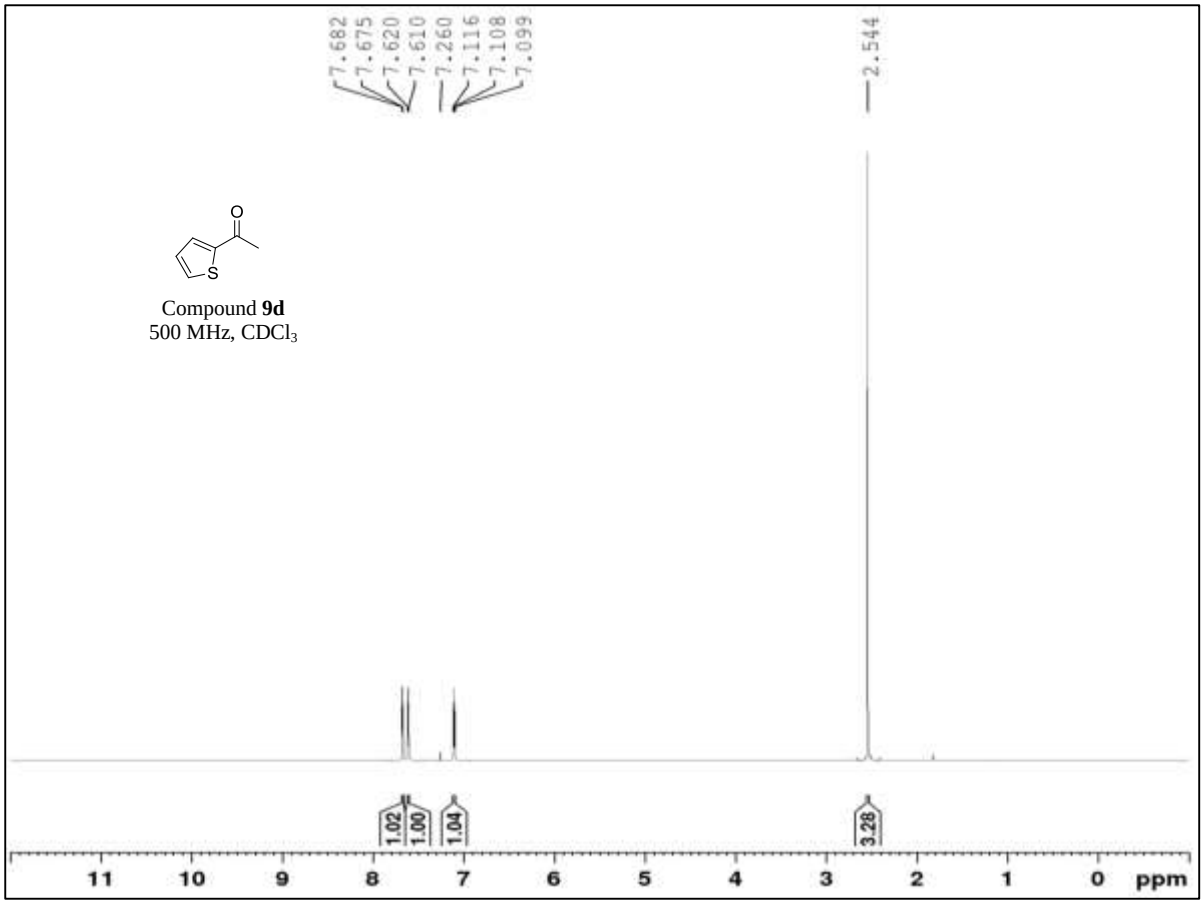
Compound **9b**
126 MHz, $CDCl_3$



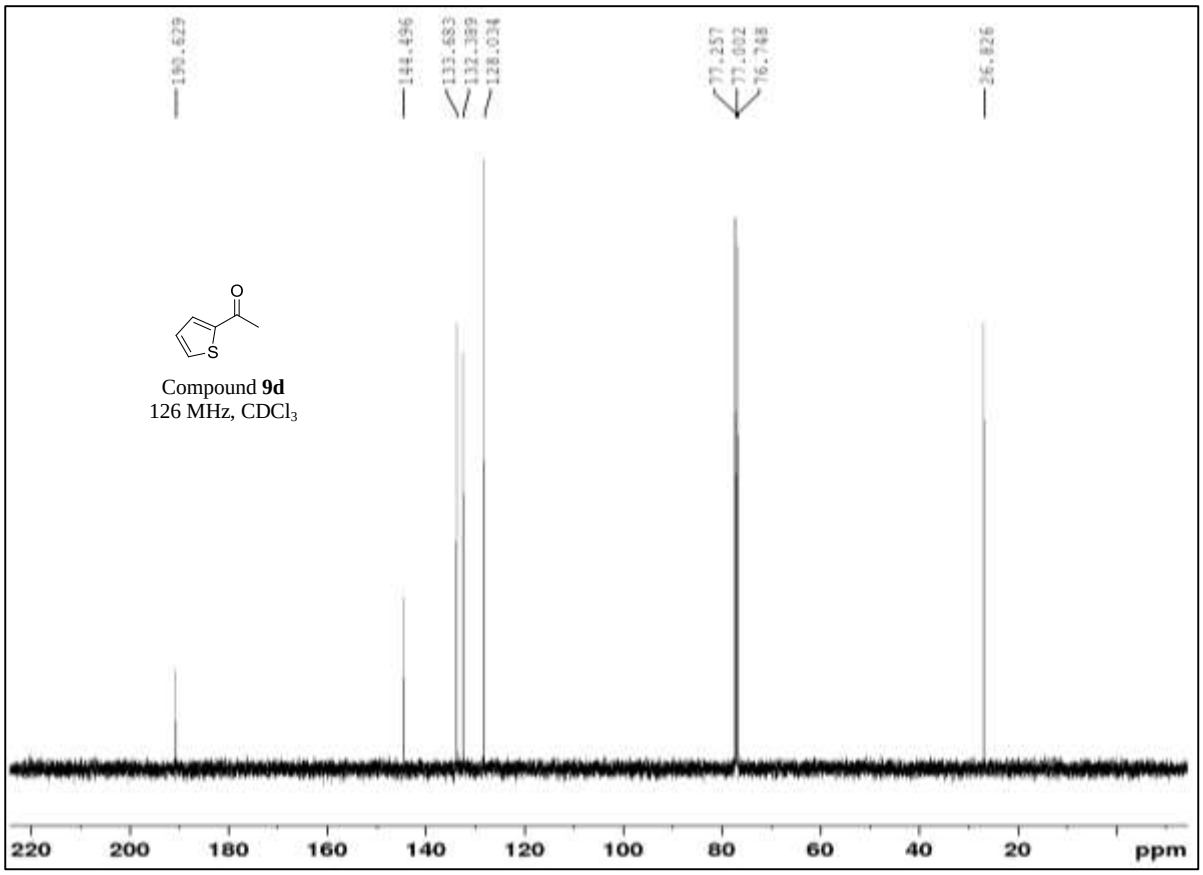
Compound **9c**
500 MHz, $CDCl_3$



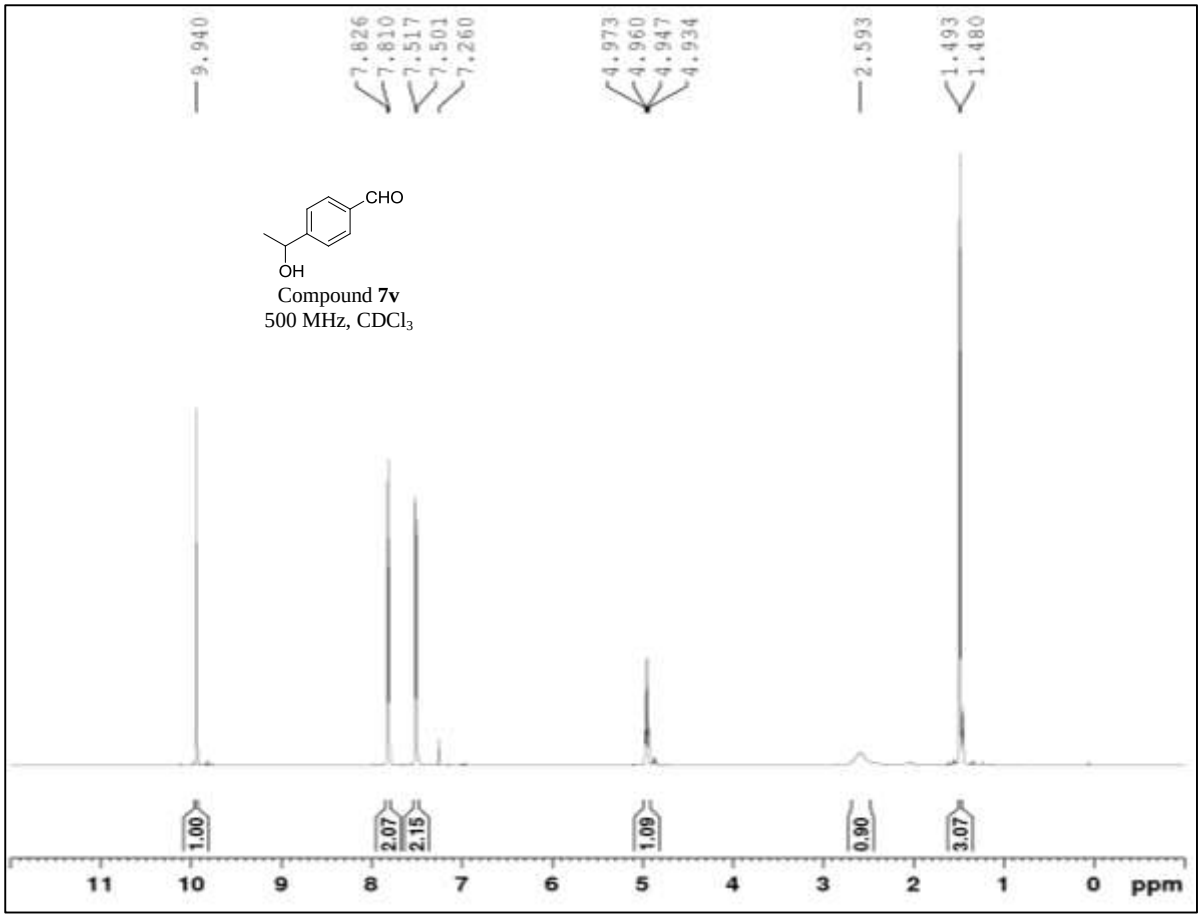
Compound **9c**
126 MHz, $CDCl_3$



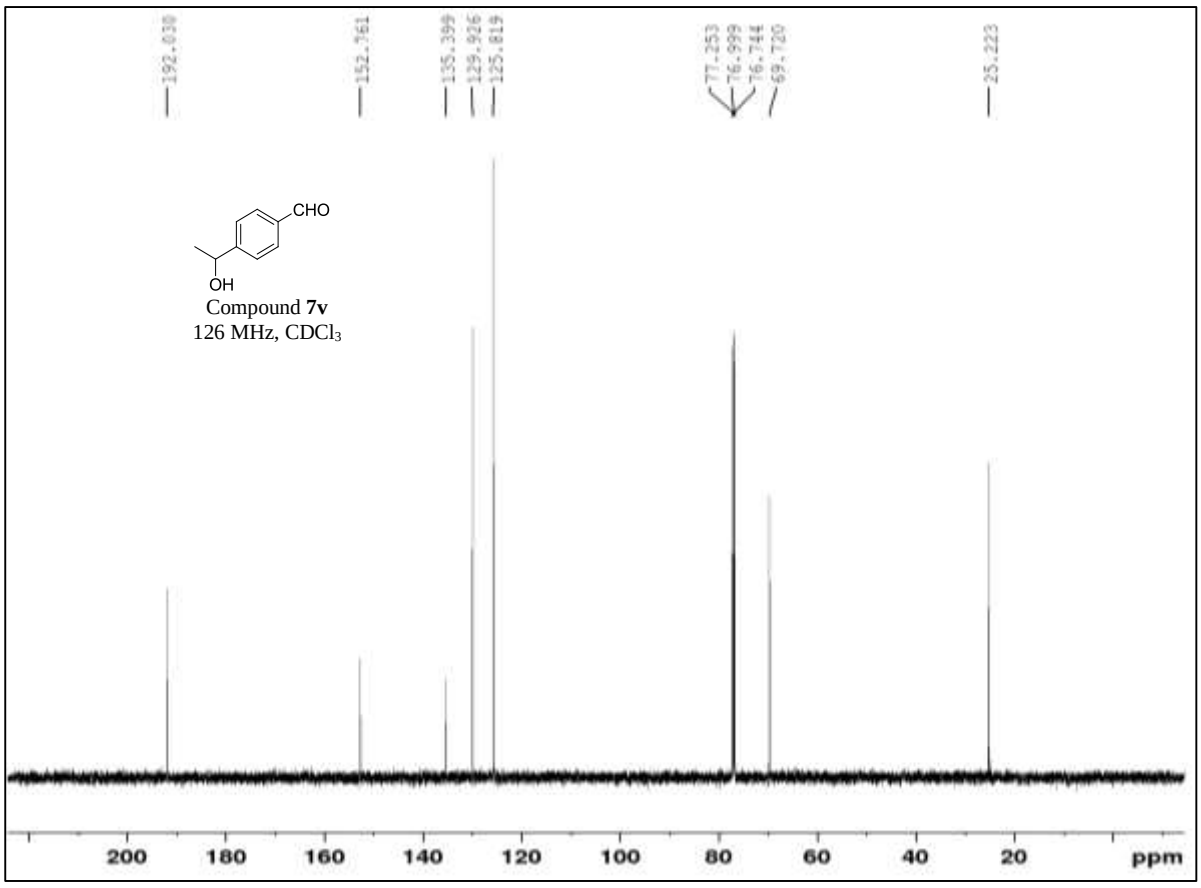
Compound **9d**
500 MHz, $CDCl_3$



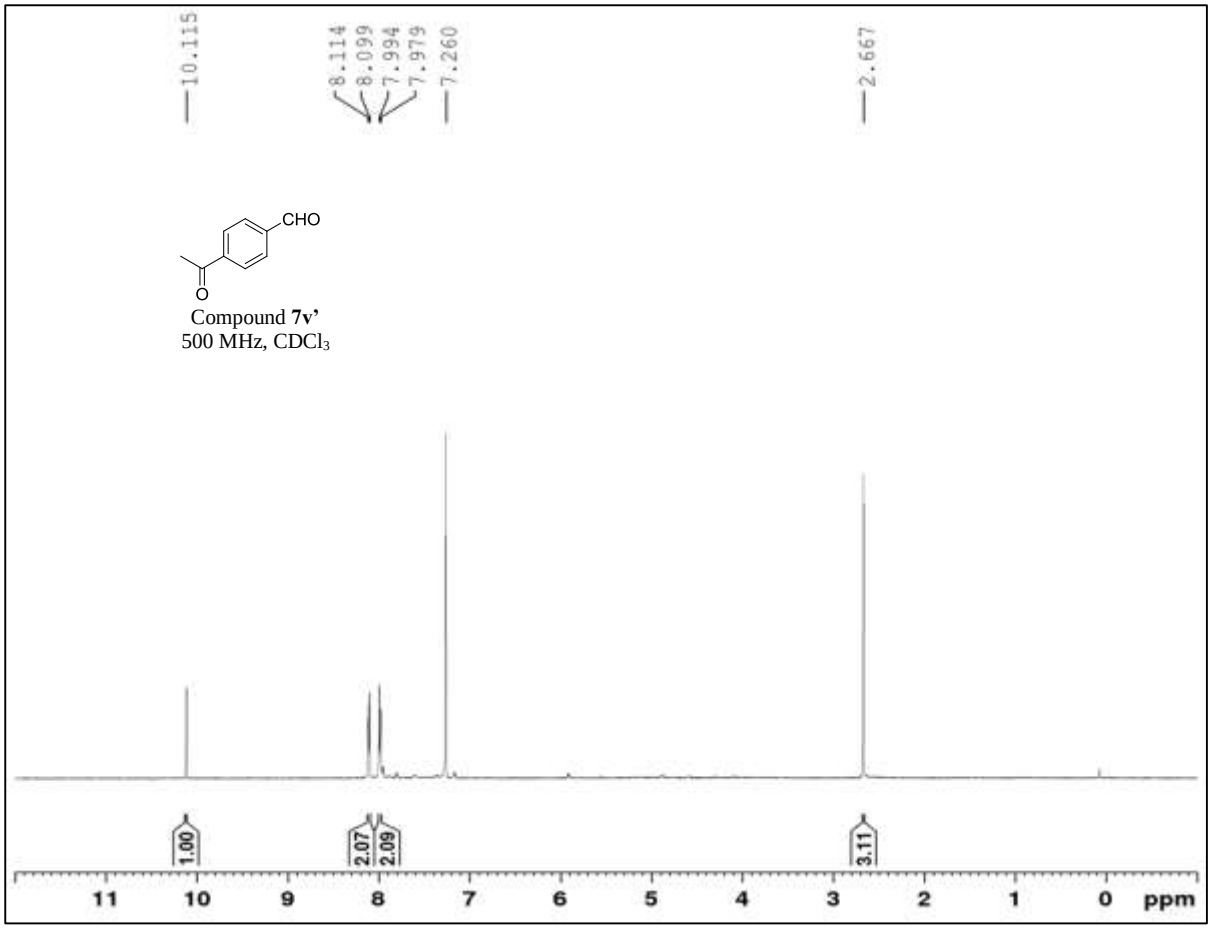
Compound **9d**
126 MHz, $CDCl_3$



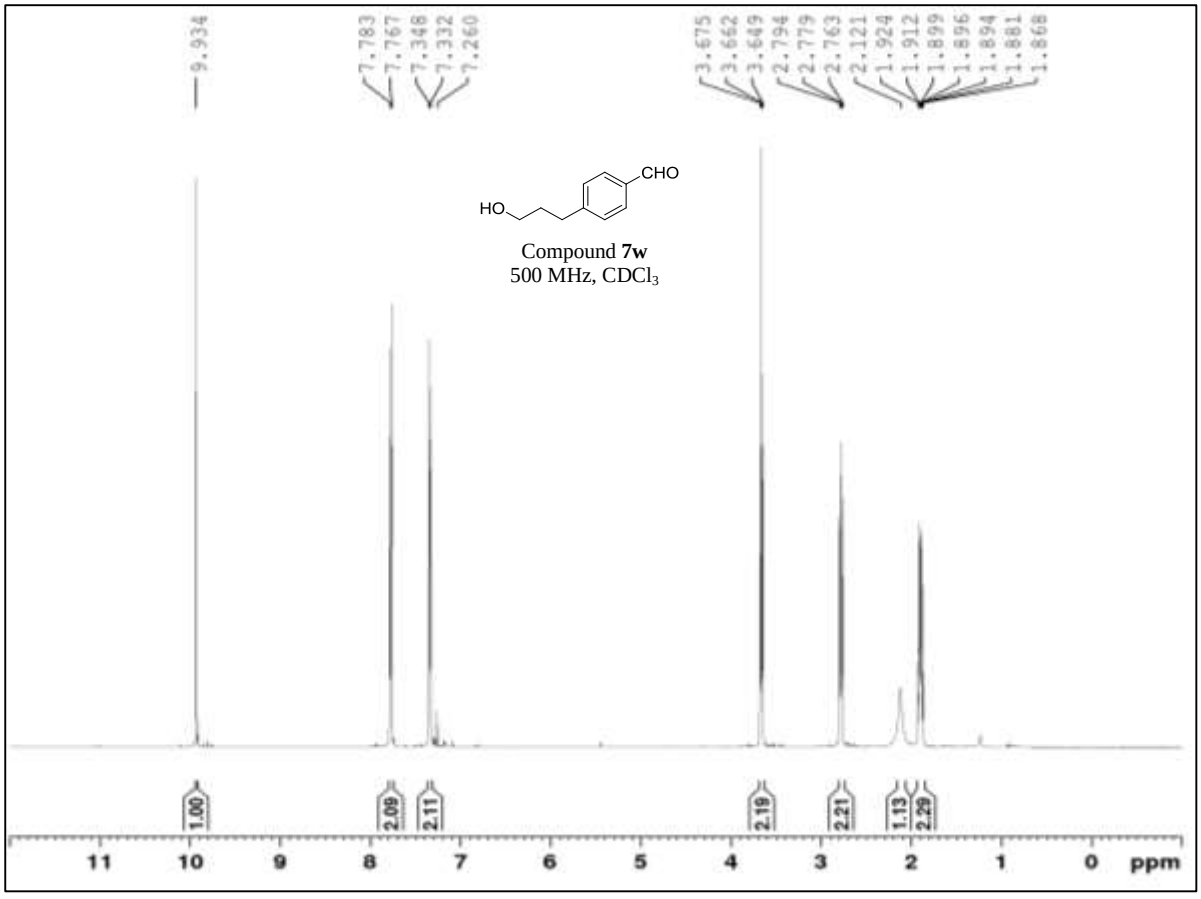
Compound **7v**
500 MHz, $CDCl_3$



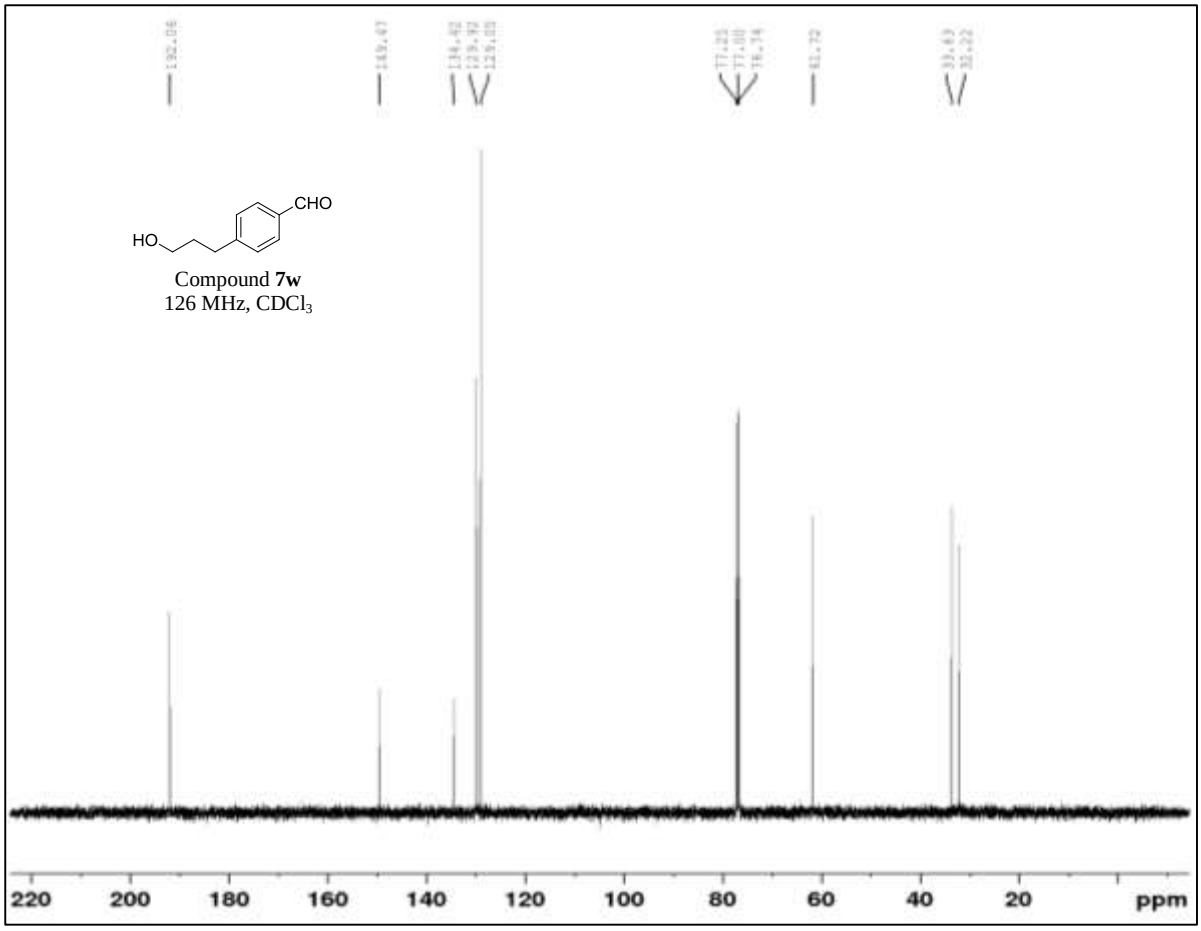
Compound **7v**
126 MHz, $CDCl_3$



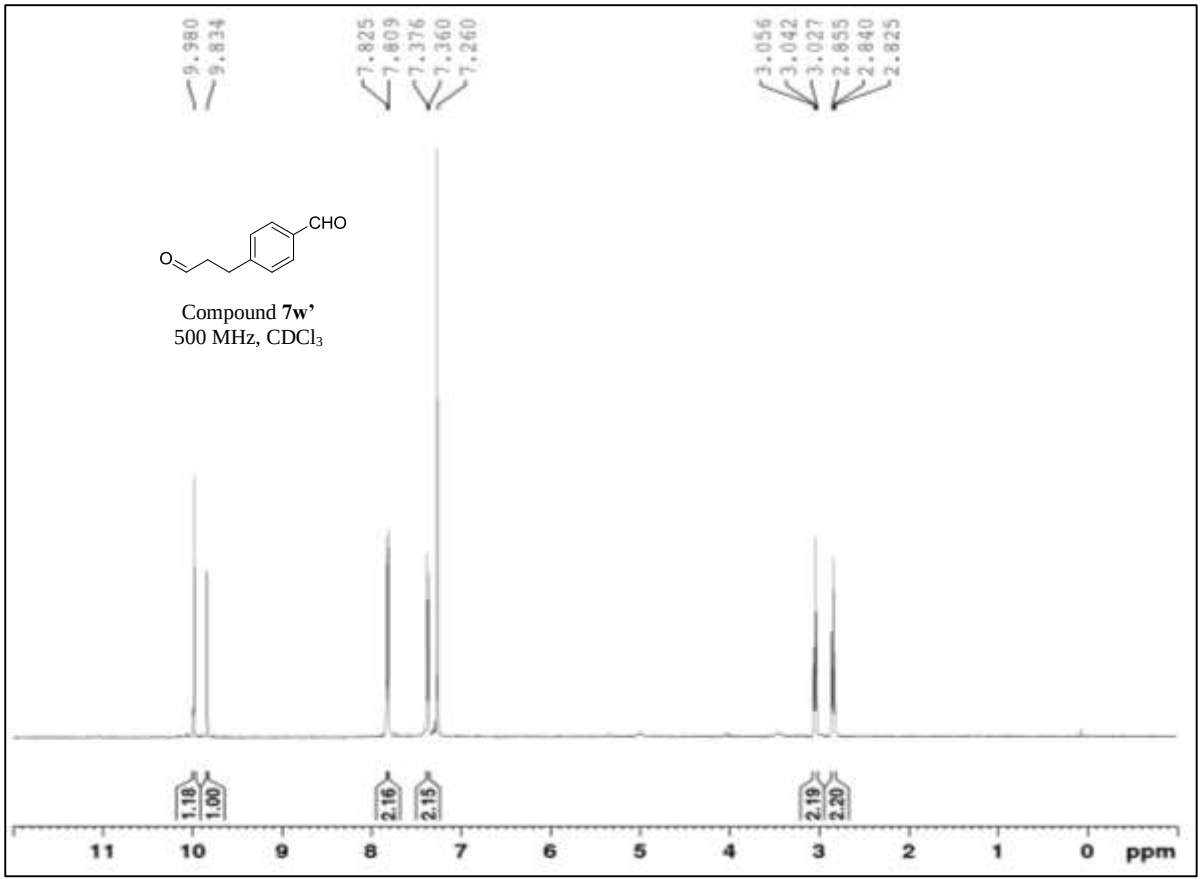
Compound **7v'**
500 MHz, $CDCl_3$



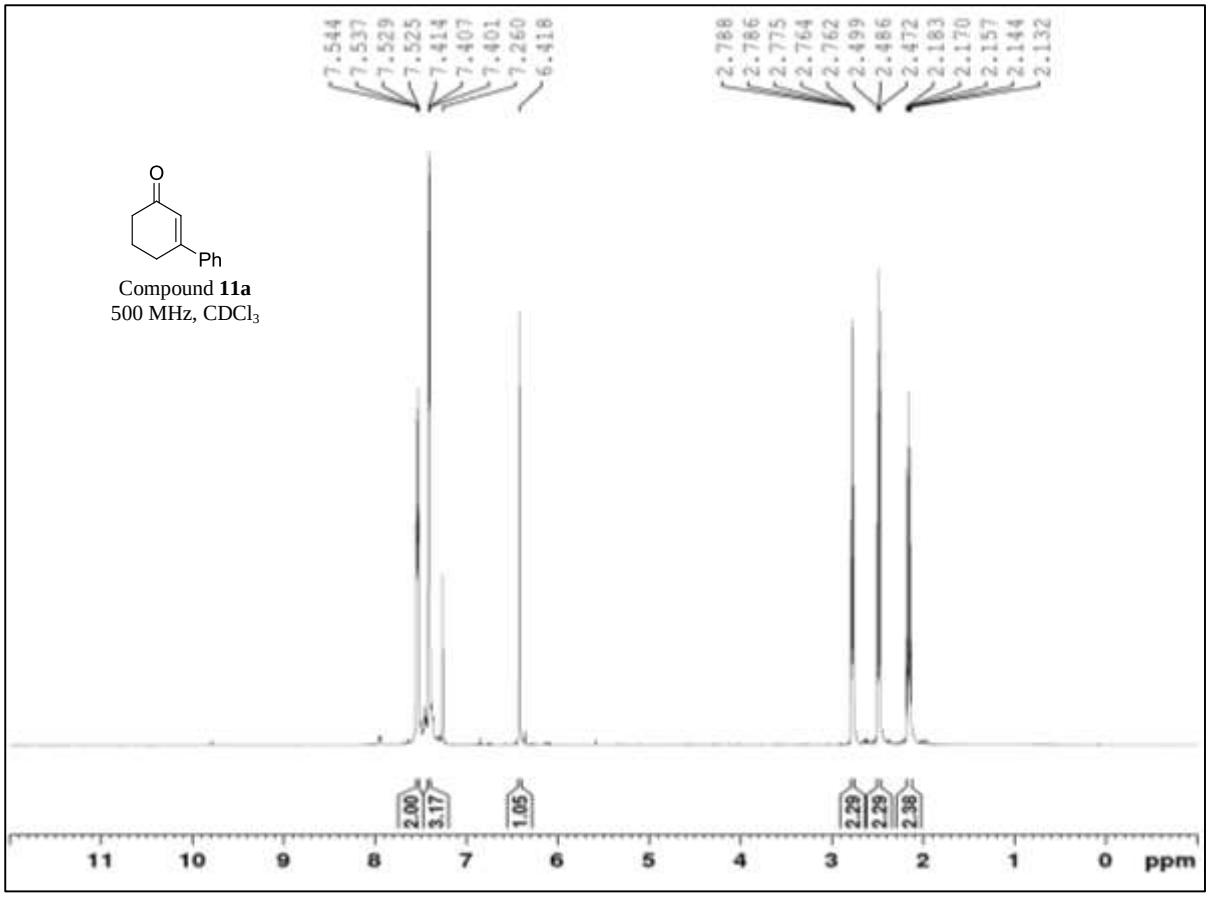
Compound **7w**
500 MHz, $CDCl_3$



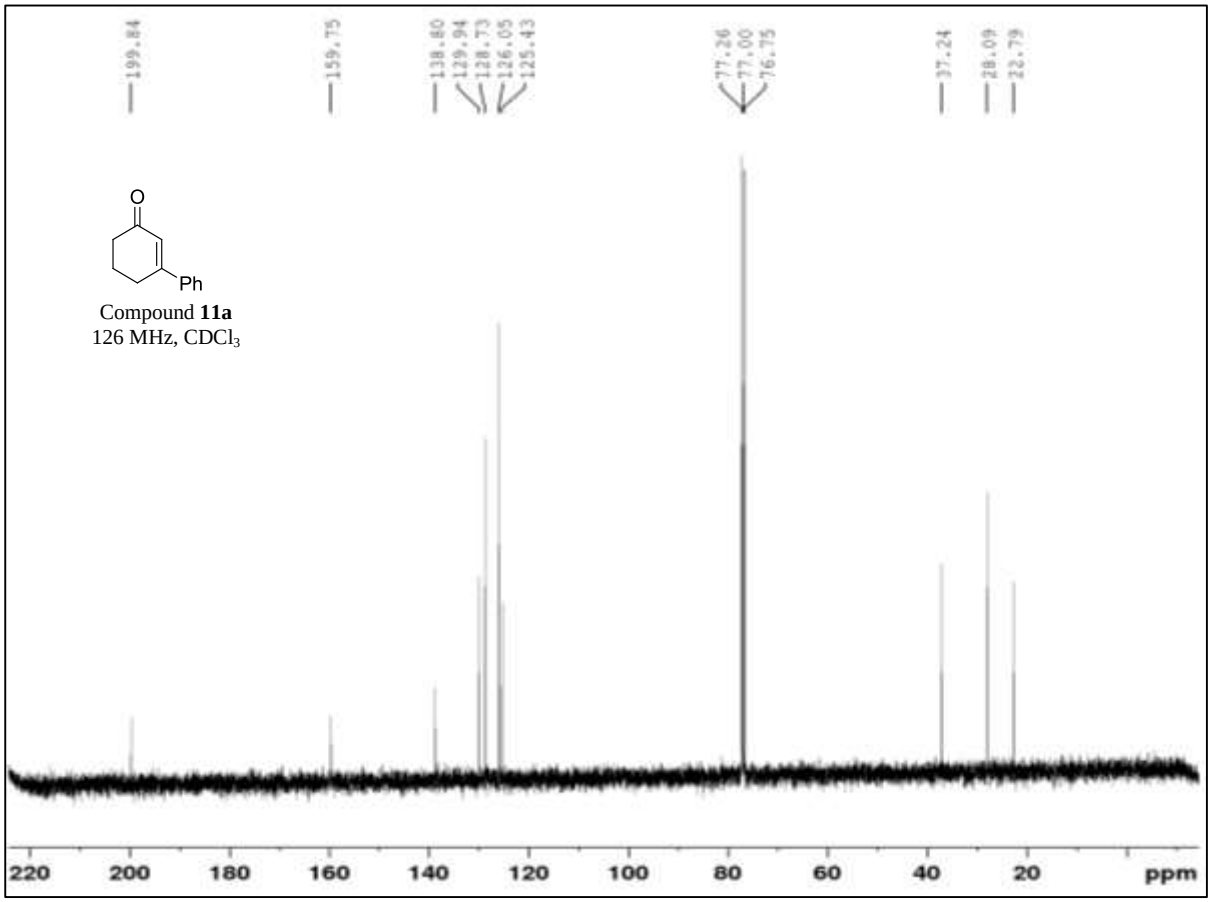
Compound **7w**
126 MHz, $CDCl_3$



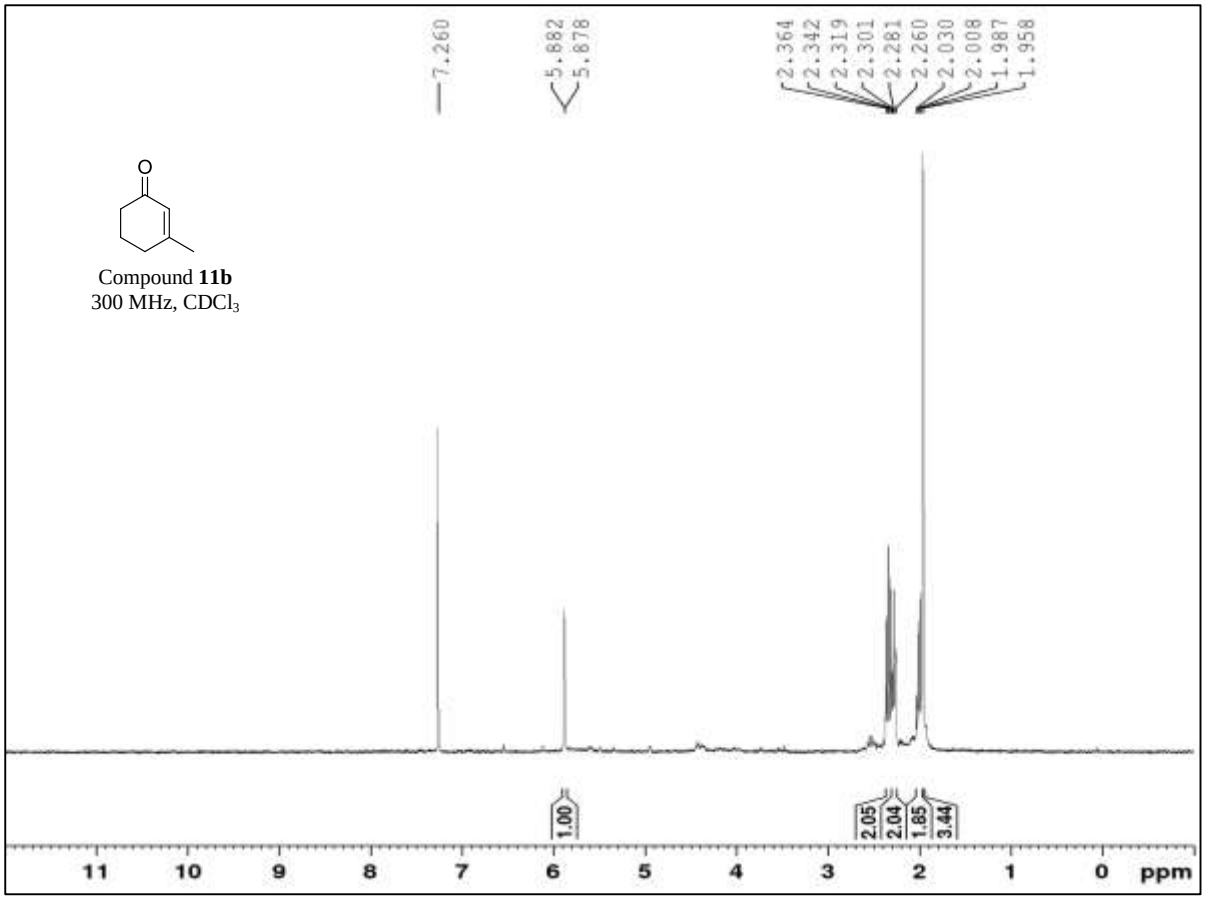
Compound **7w'**
500 MHz, $CDCl_3$



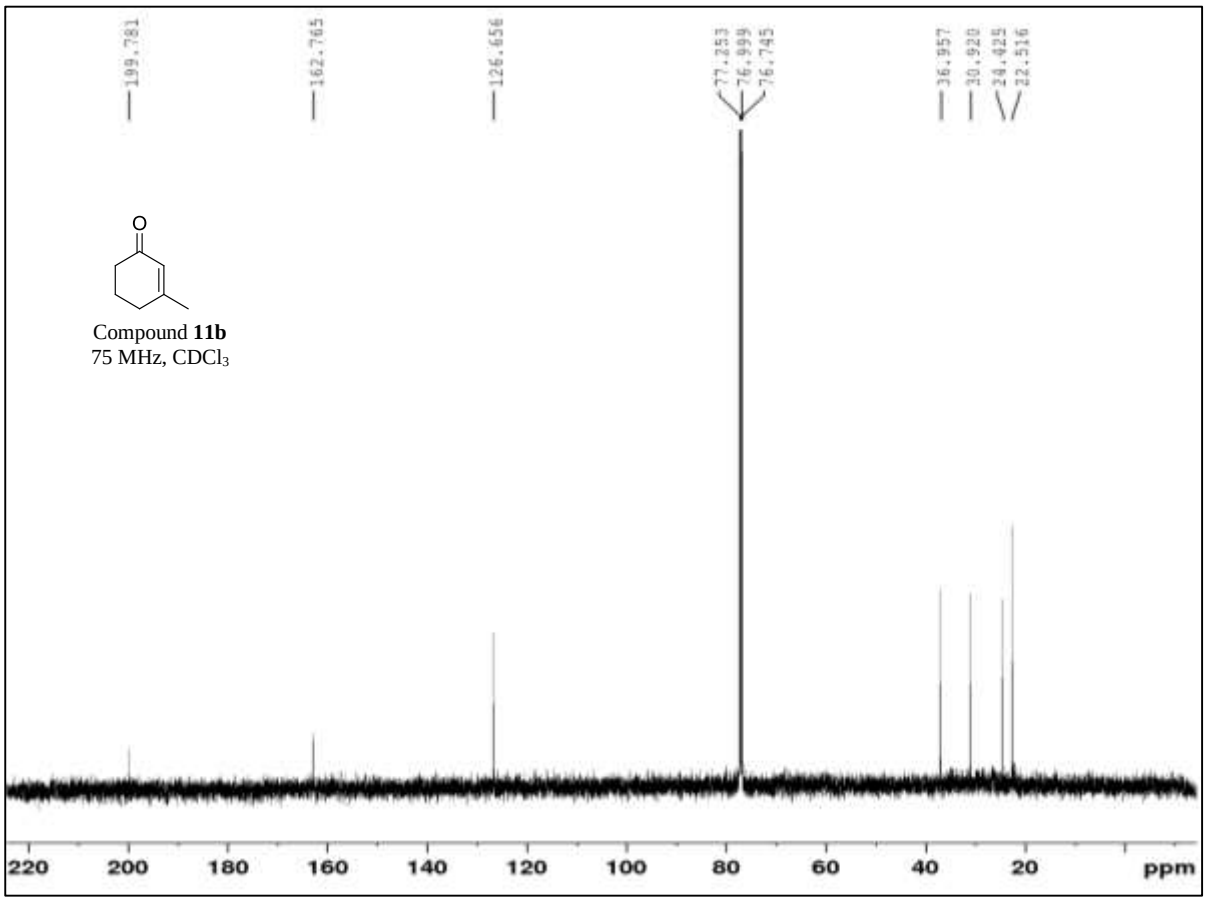
Compound **11a**
500 MHz, $CDCl_3$



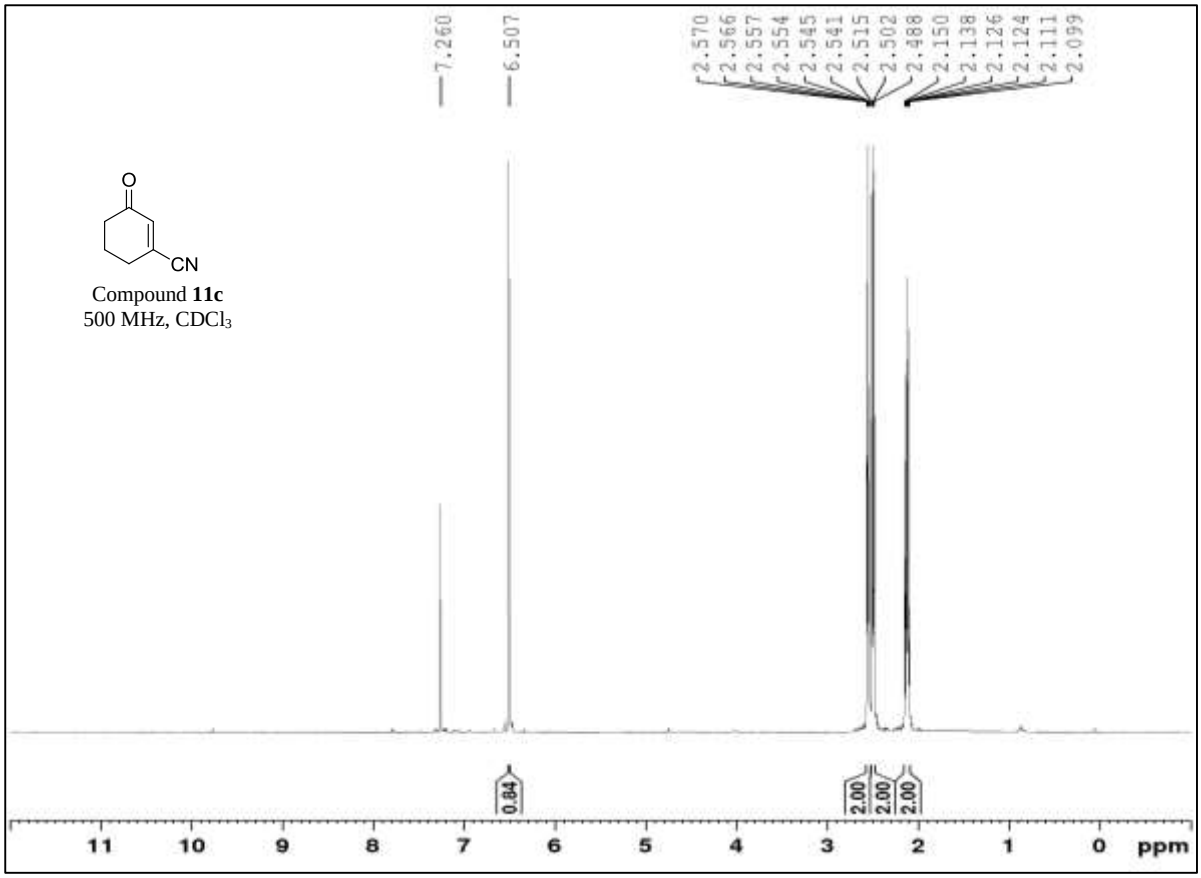
Compound **11a**
126 MHz, $CDCl_3$



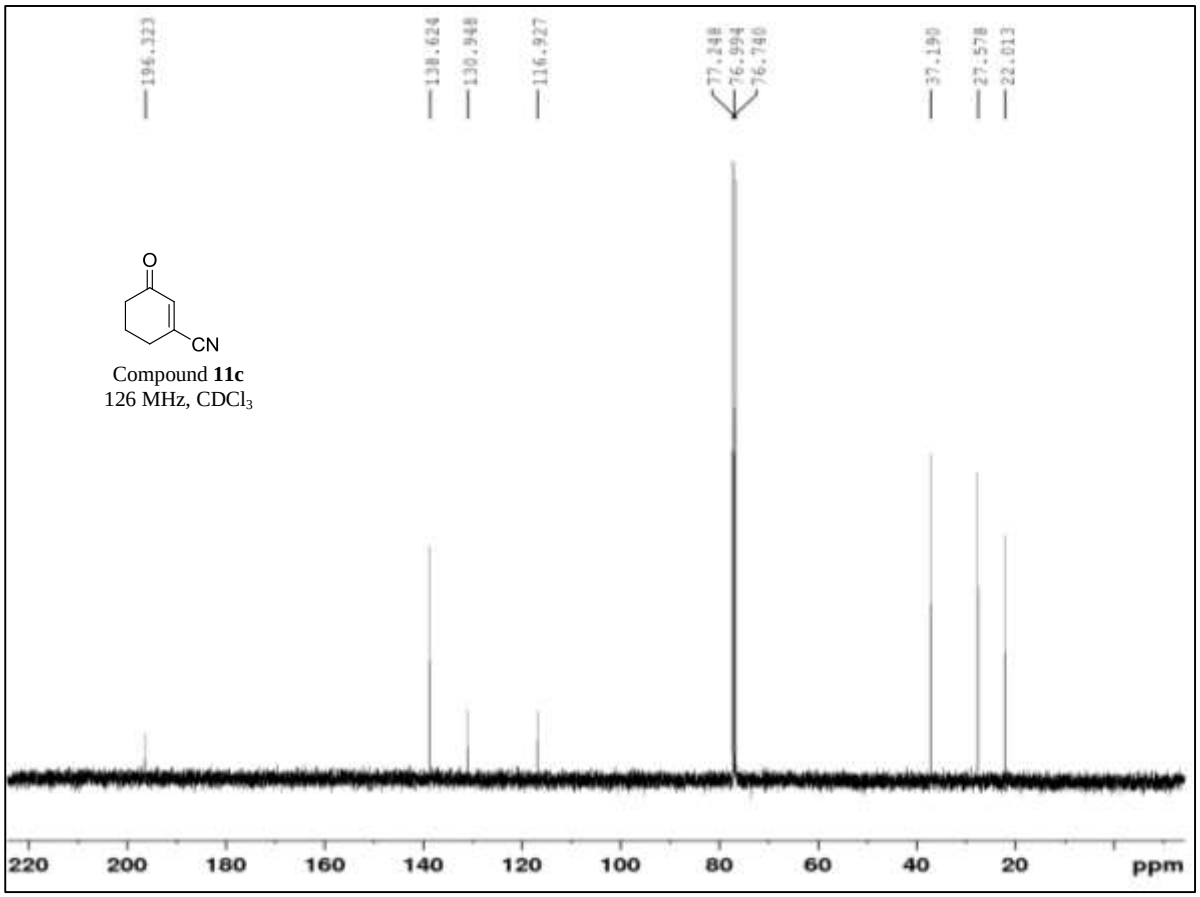
Compound **11b**
300 MHz, $CDCl_3$



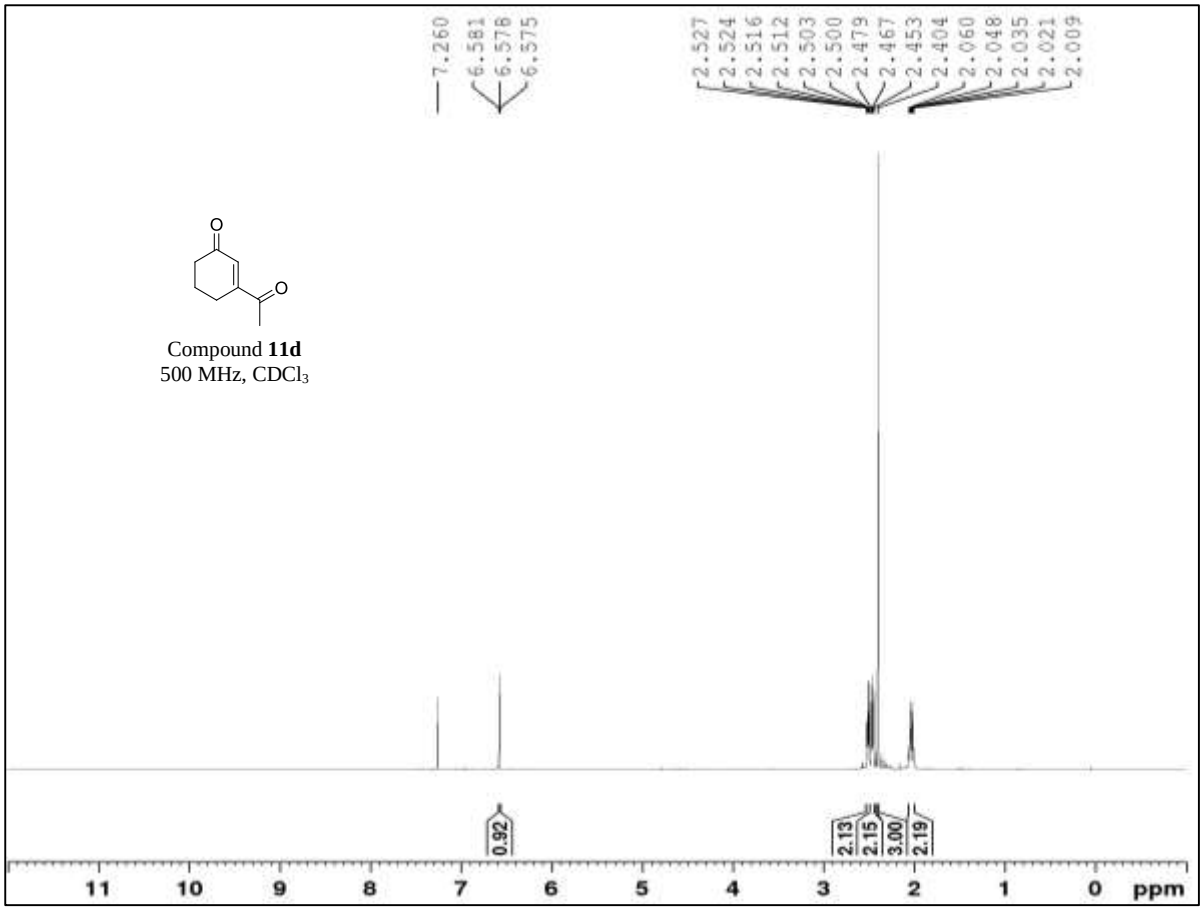
Compound **11b**
75 MHz, $CDCl_3$



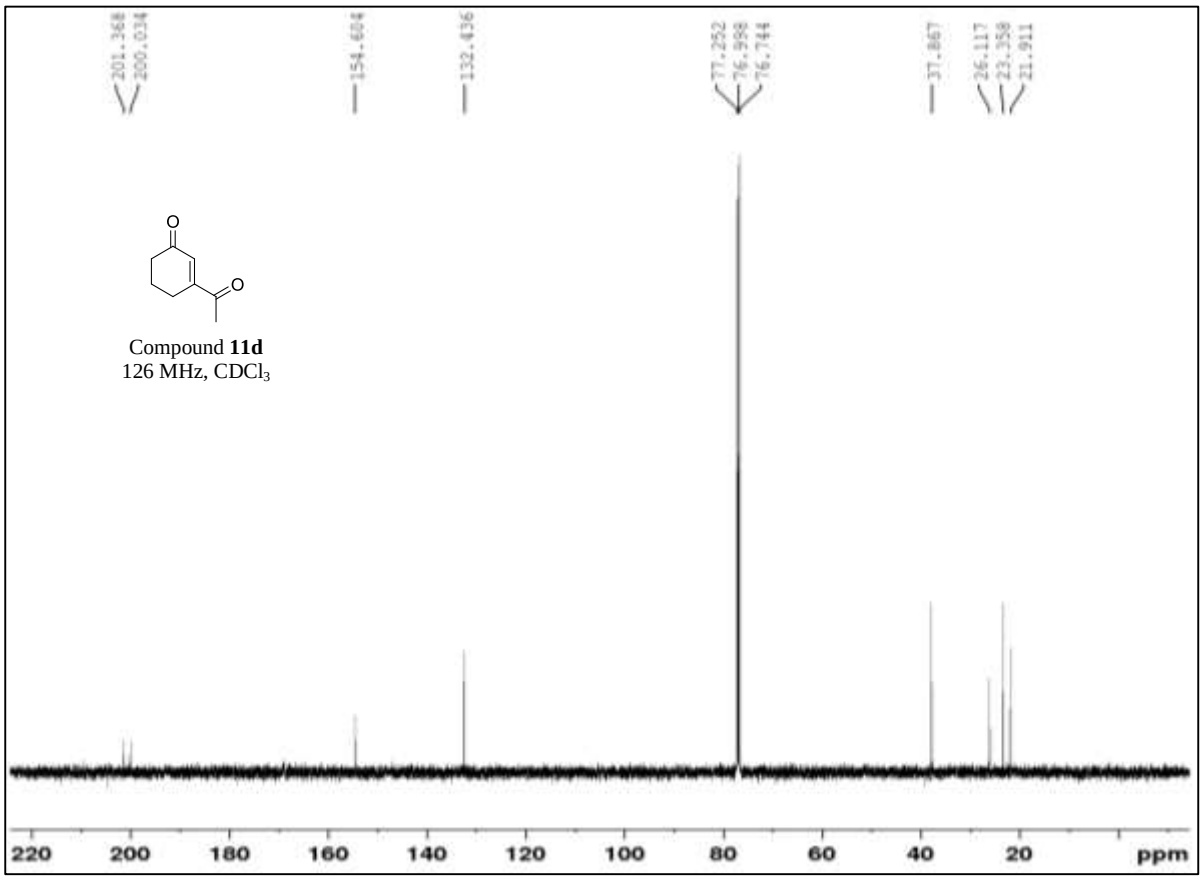
Compound **11c**
500 MHz, $CDCl_3$



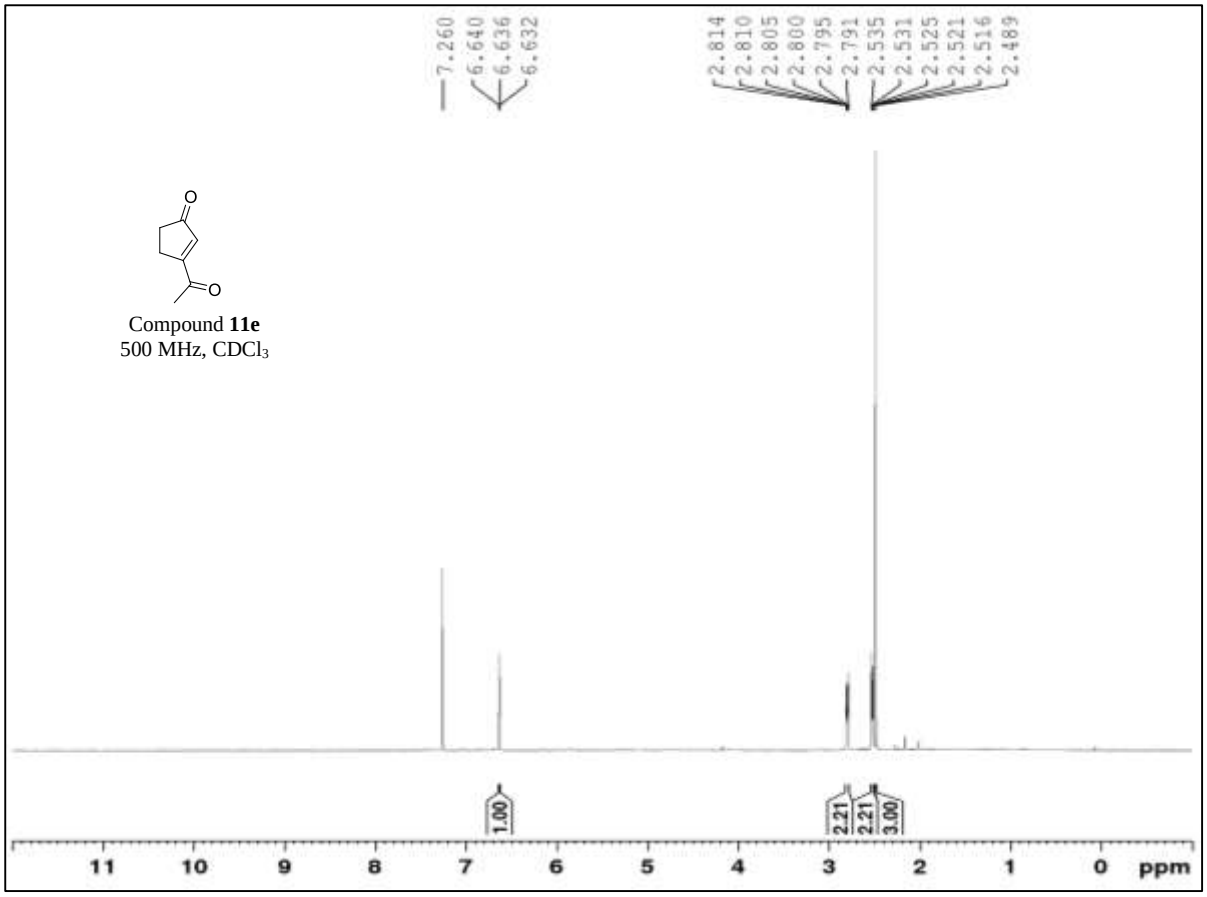
Compound **11c**
126 MHz, $CDCl_3$



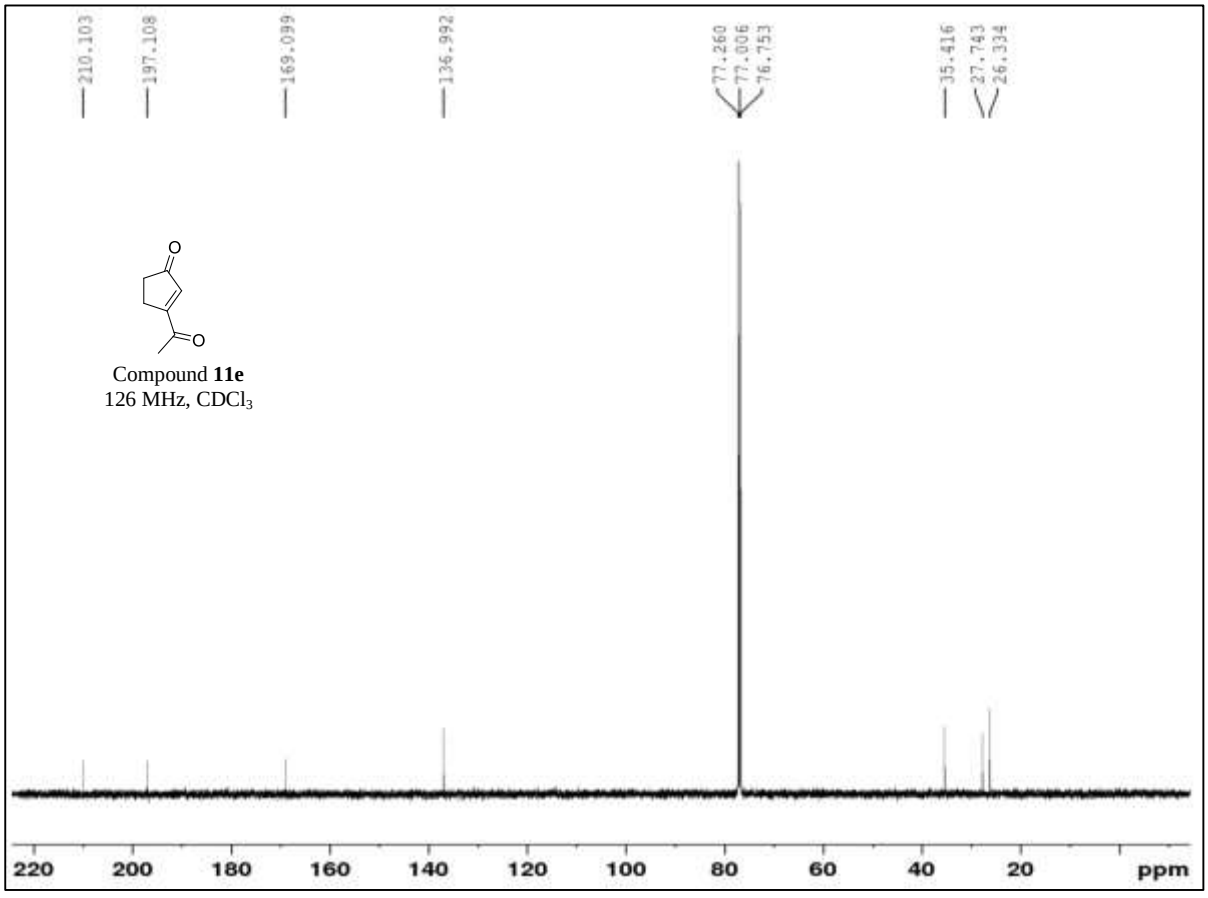
Compound **11d**
500 MHz, $CDCl_3$



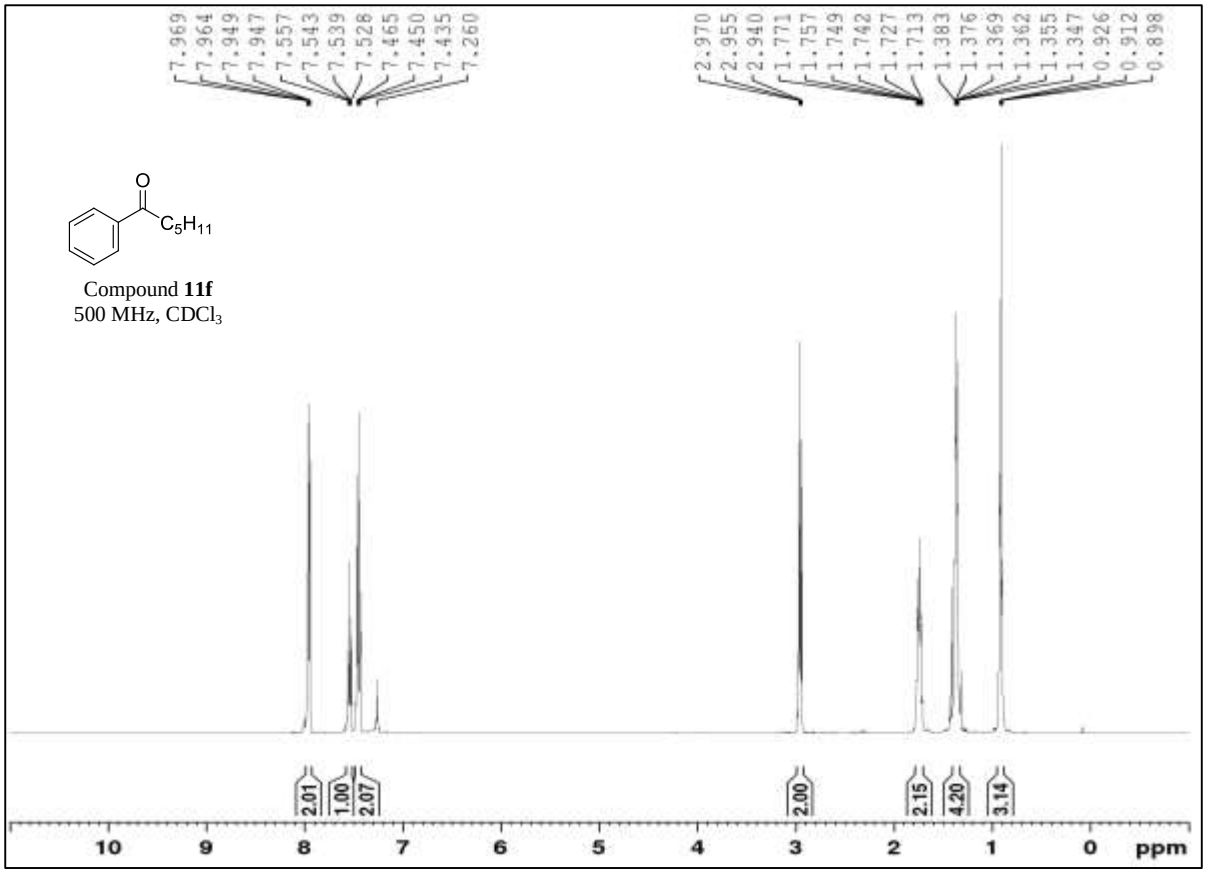
Compound **11d**
126 MHz, $CDCl_3$



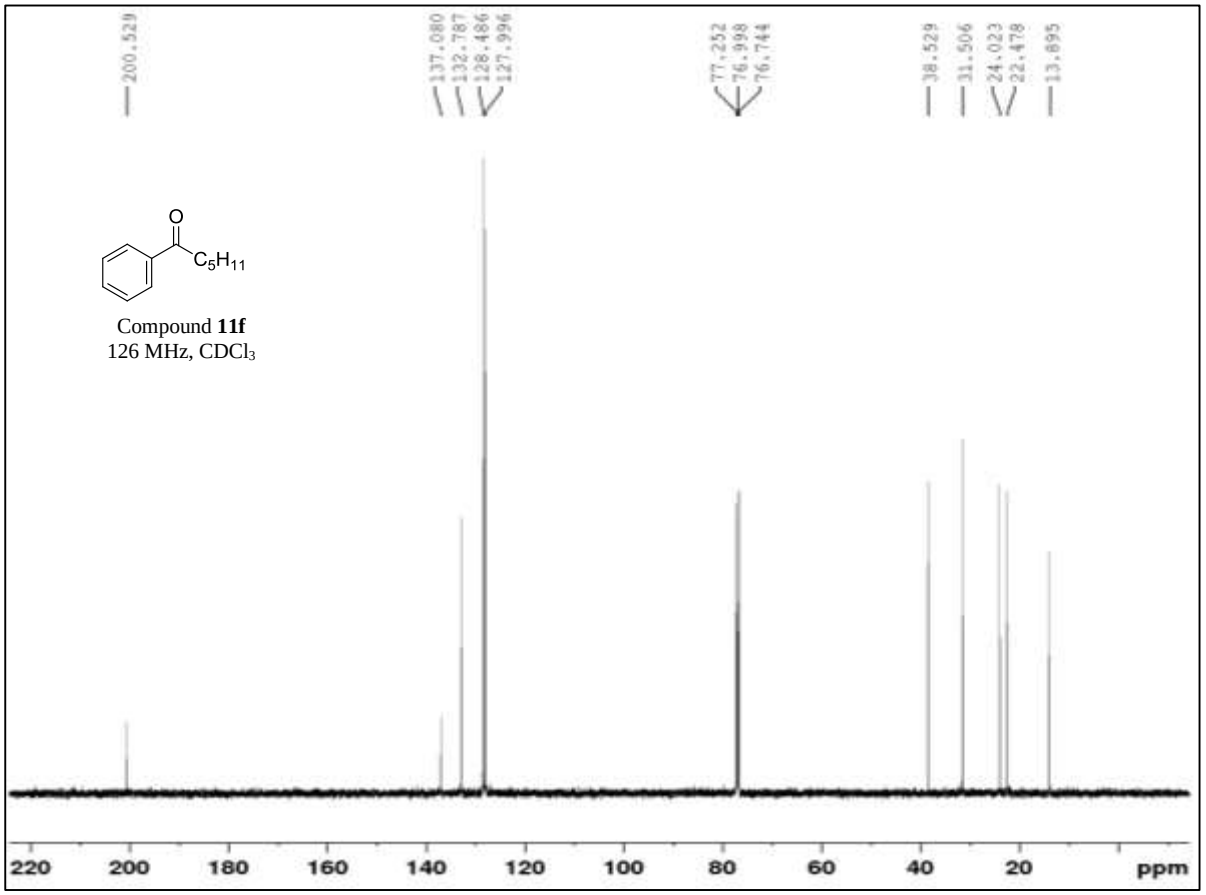
Compound **11e**
500 MHz, $CDCl_3$



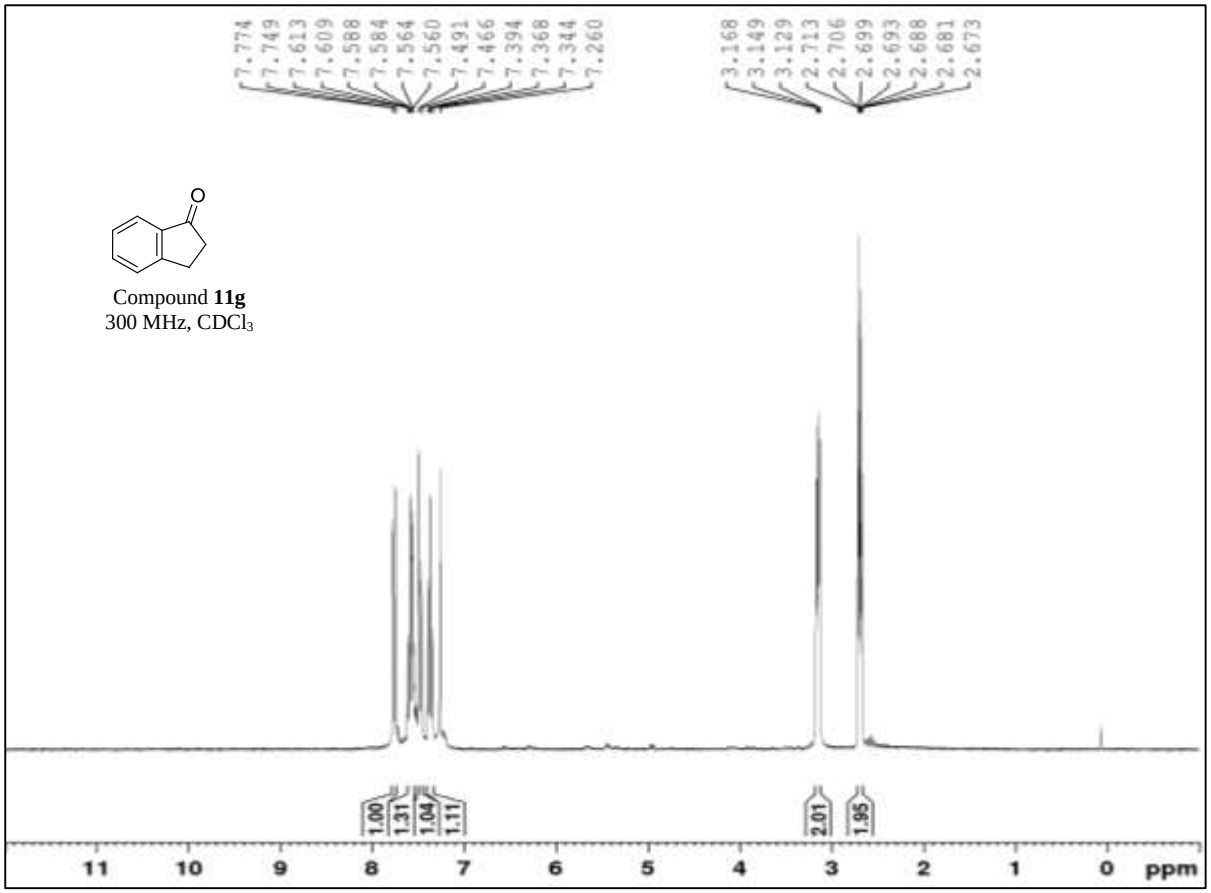
Compound **11e**
126 MHz, $CDCl_3$



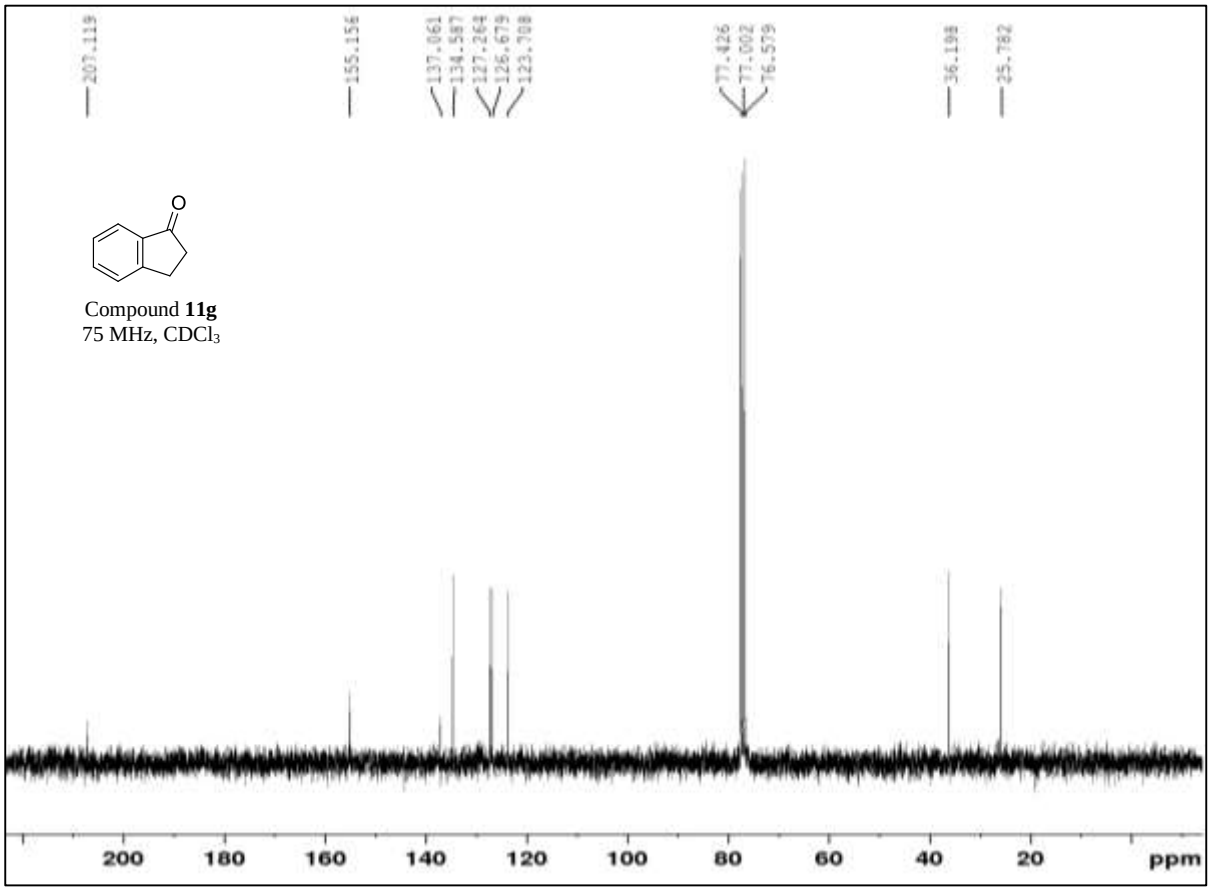
Compound **11f**
500 MHz, $CDCl_3$



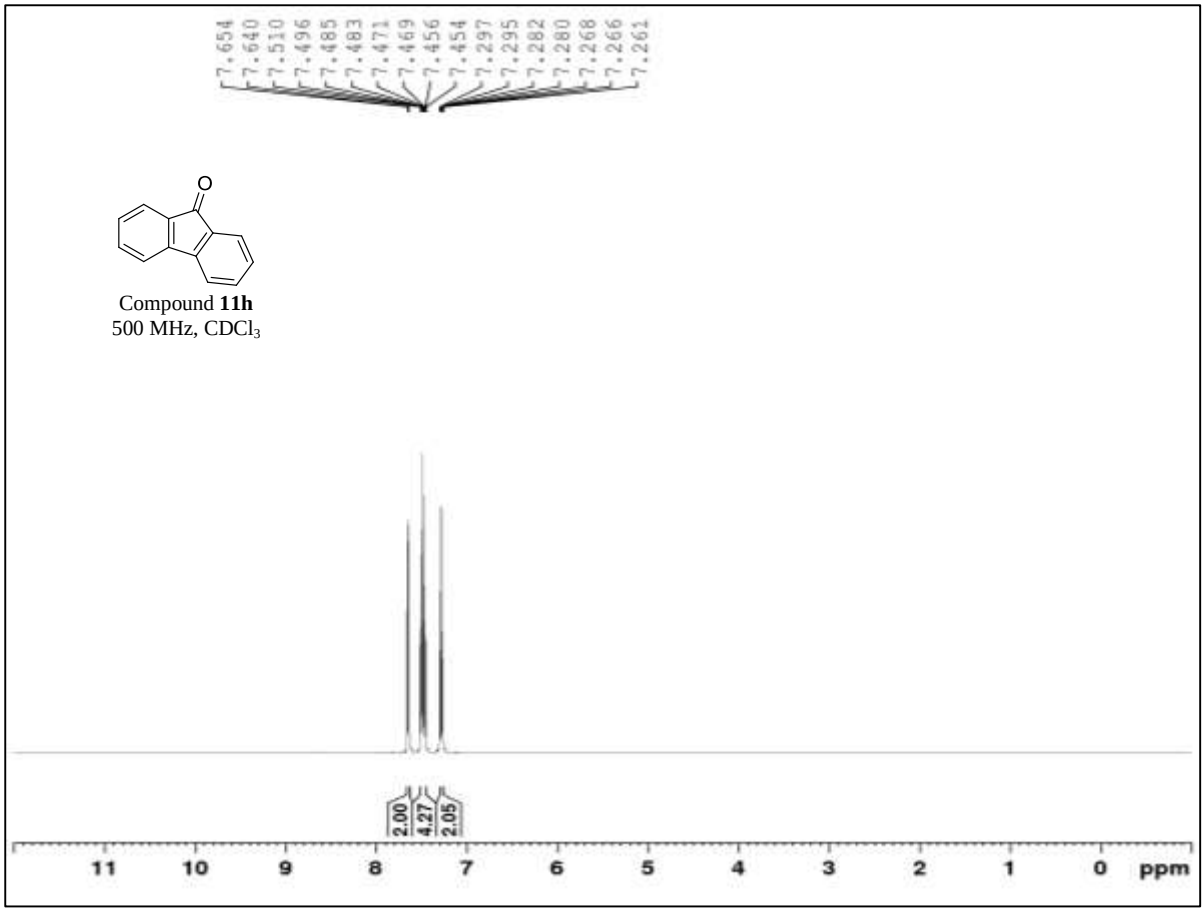
Compound **11f**
126 MHz, $CDCl_3$



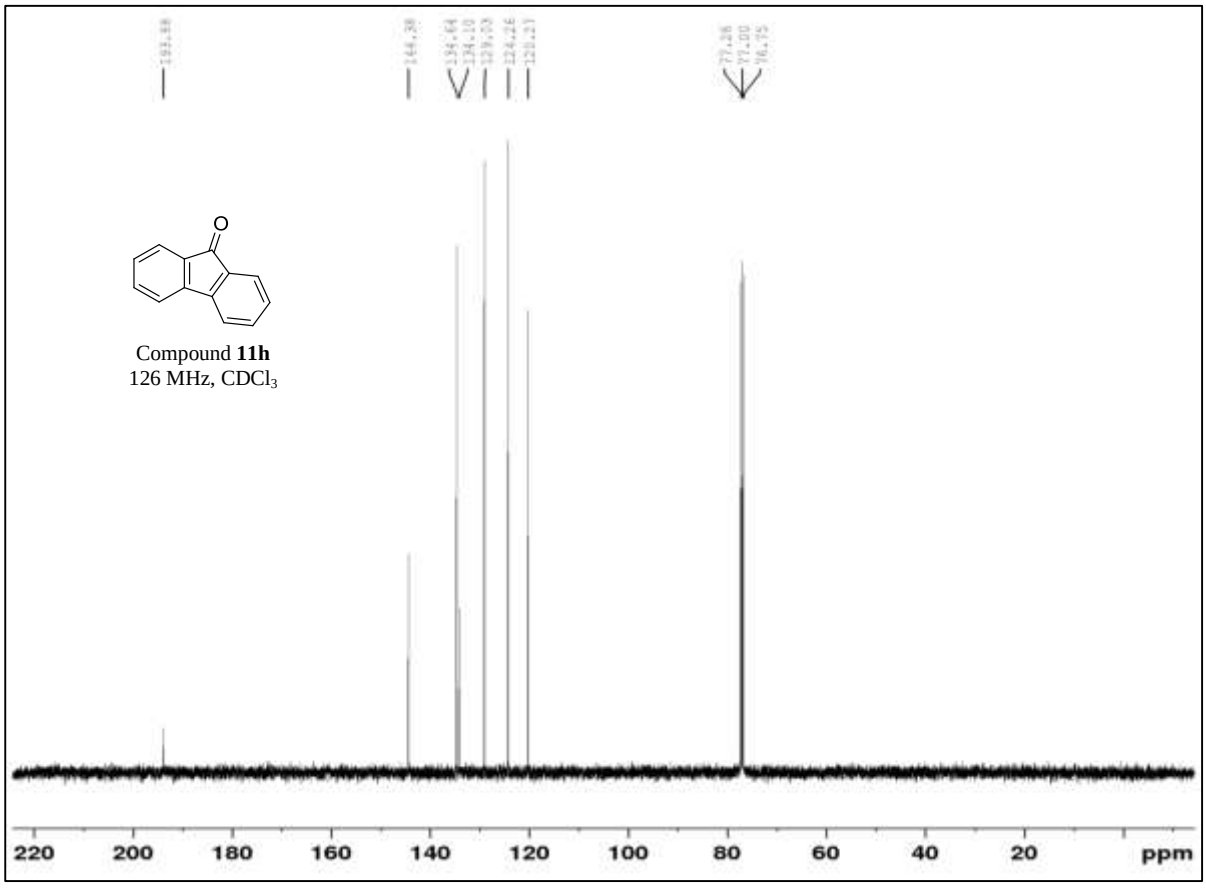
Compound **11g**
300 MHz, $CDCl_3$



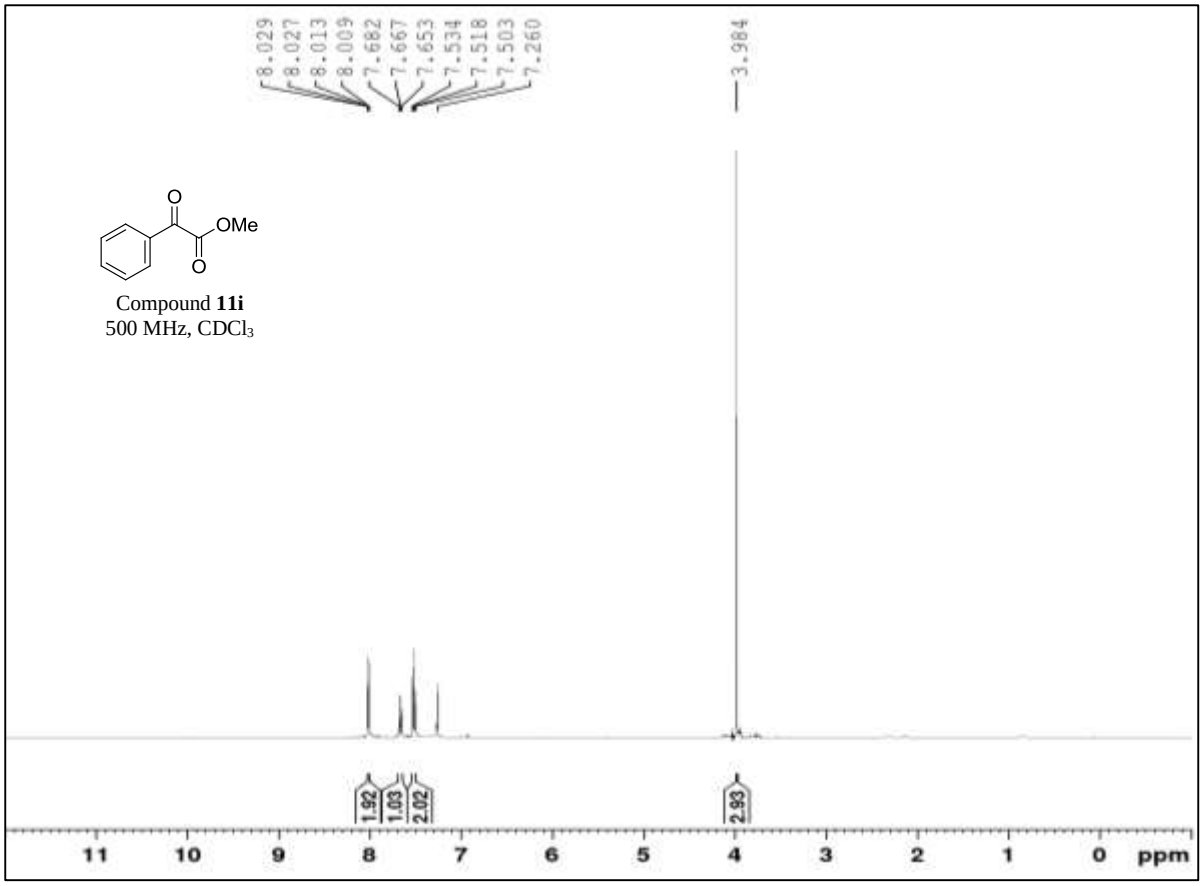
Compound **11g**
75 MHz, $CDCl_3$



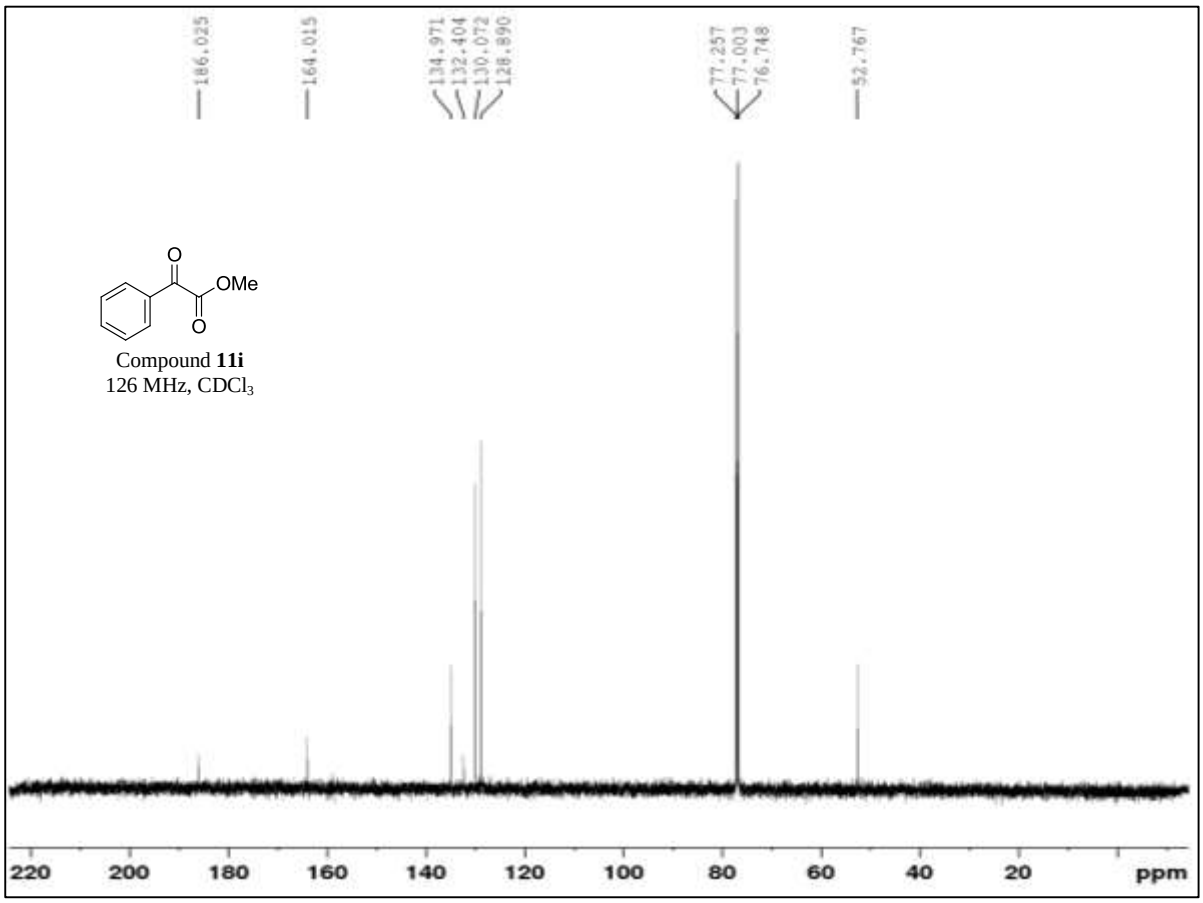
Compound **11h**
500 MHz, $CDCl_3$



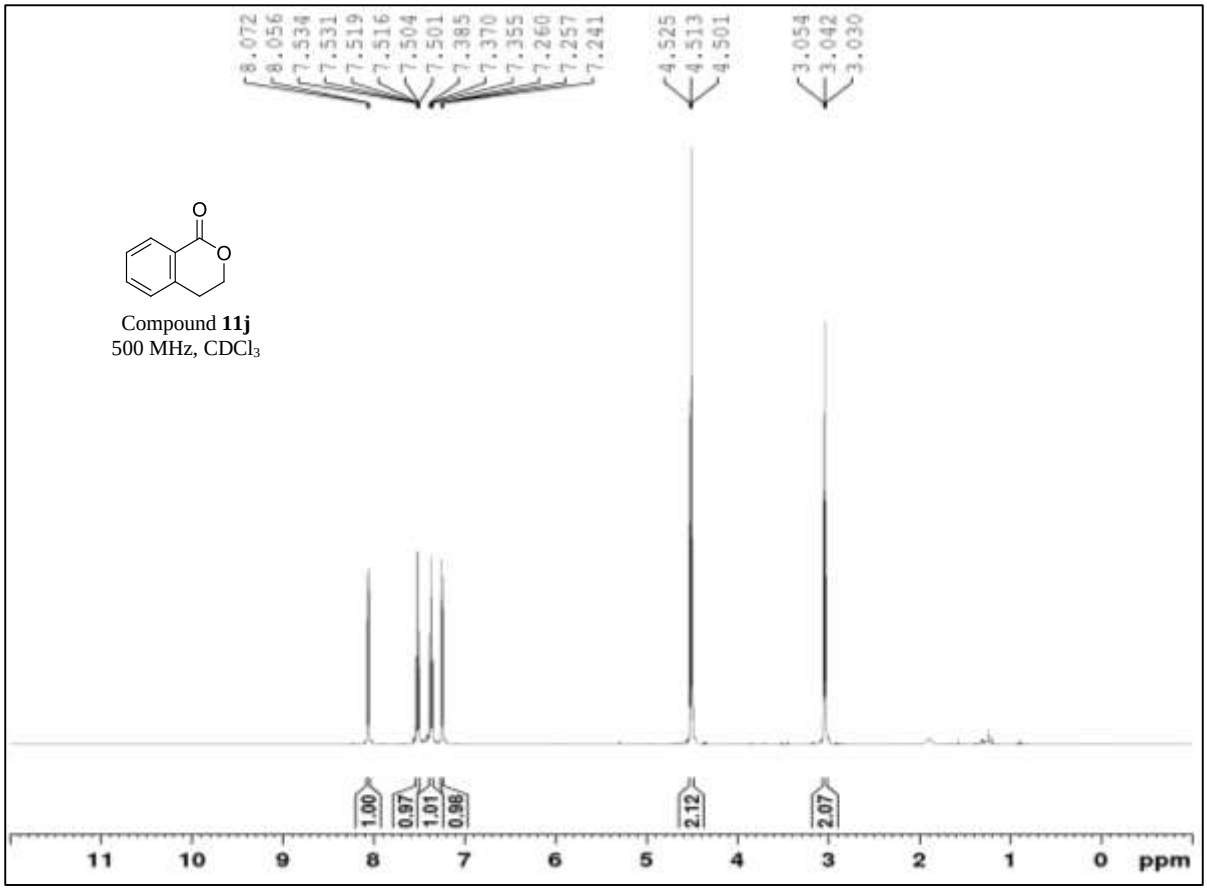
Compound **11h**
126 MHz, $CDCl_3$



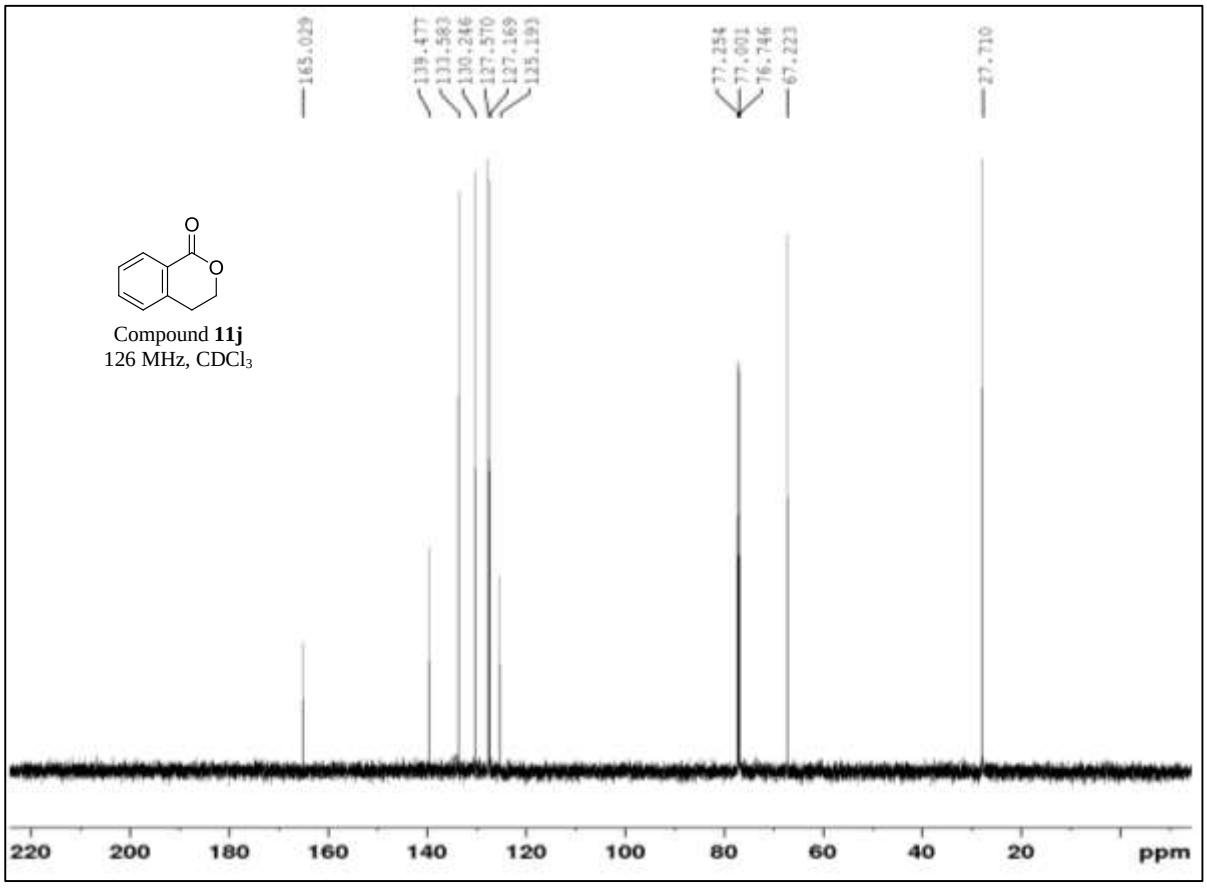
Compound **11i**
500 MHz, $CDCl_3$



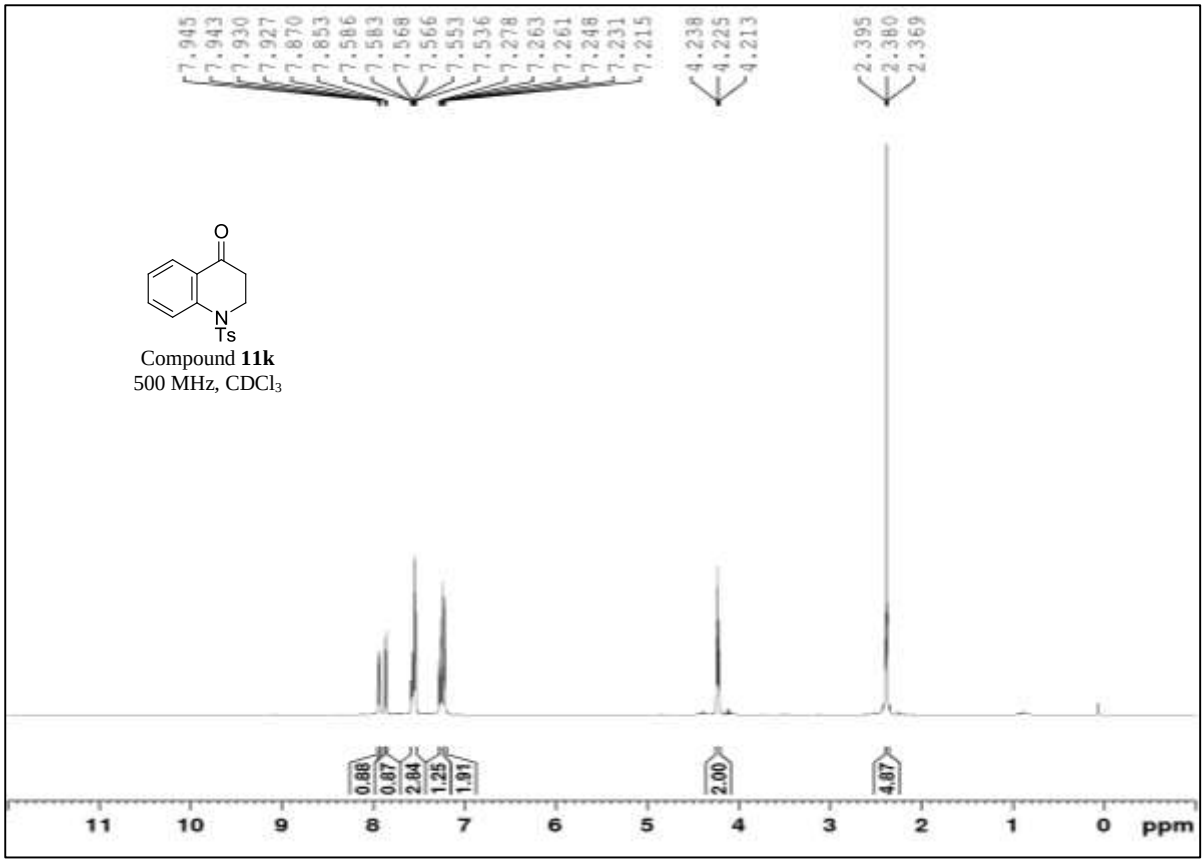
Compound **11i**
126 MHz, $CDCl_3$



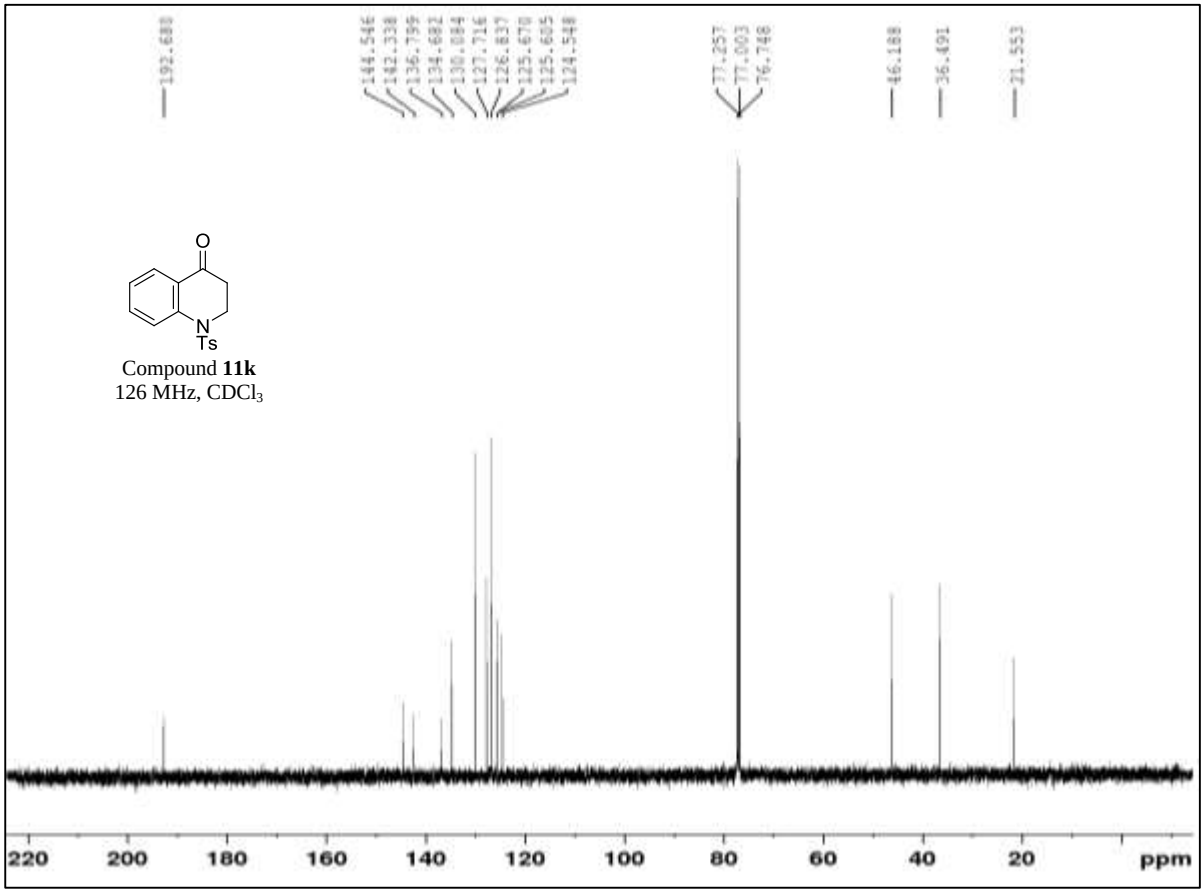
Compound **11j**
500 MHz, $CDCl_3$



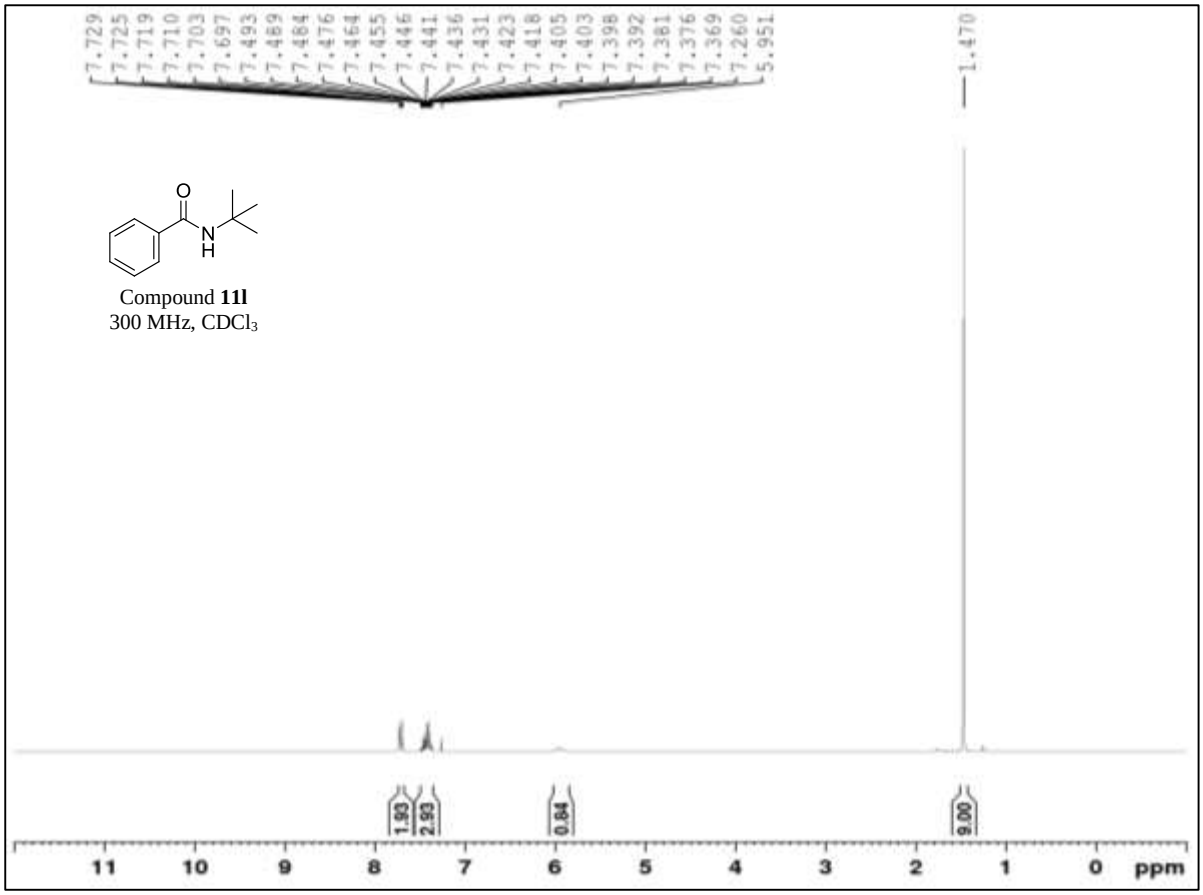
Compound **11j**
126 MHz, $CDCl_3$



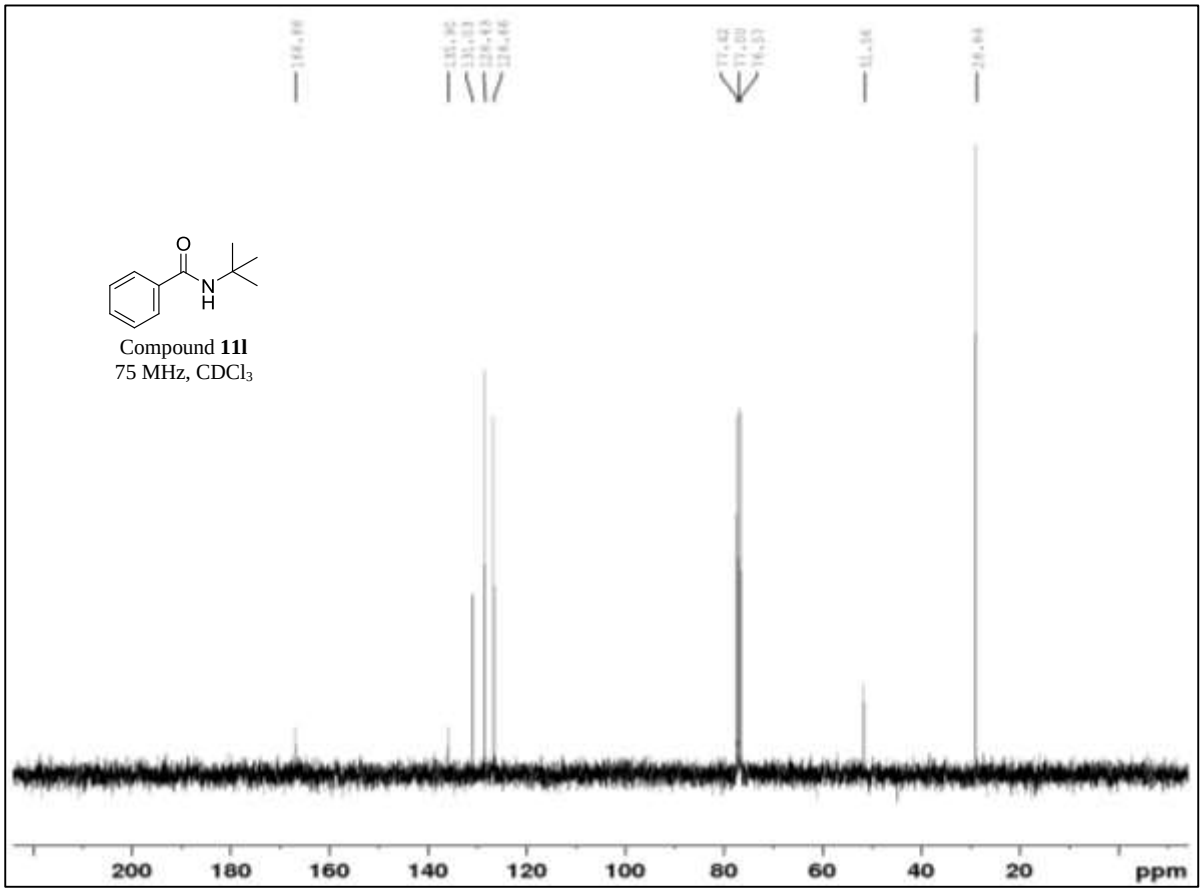
Compound **11k**
500 MHz, $CDCl_3$



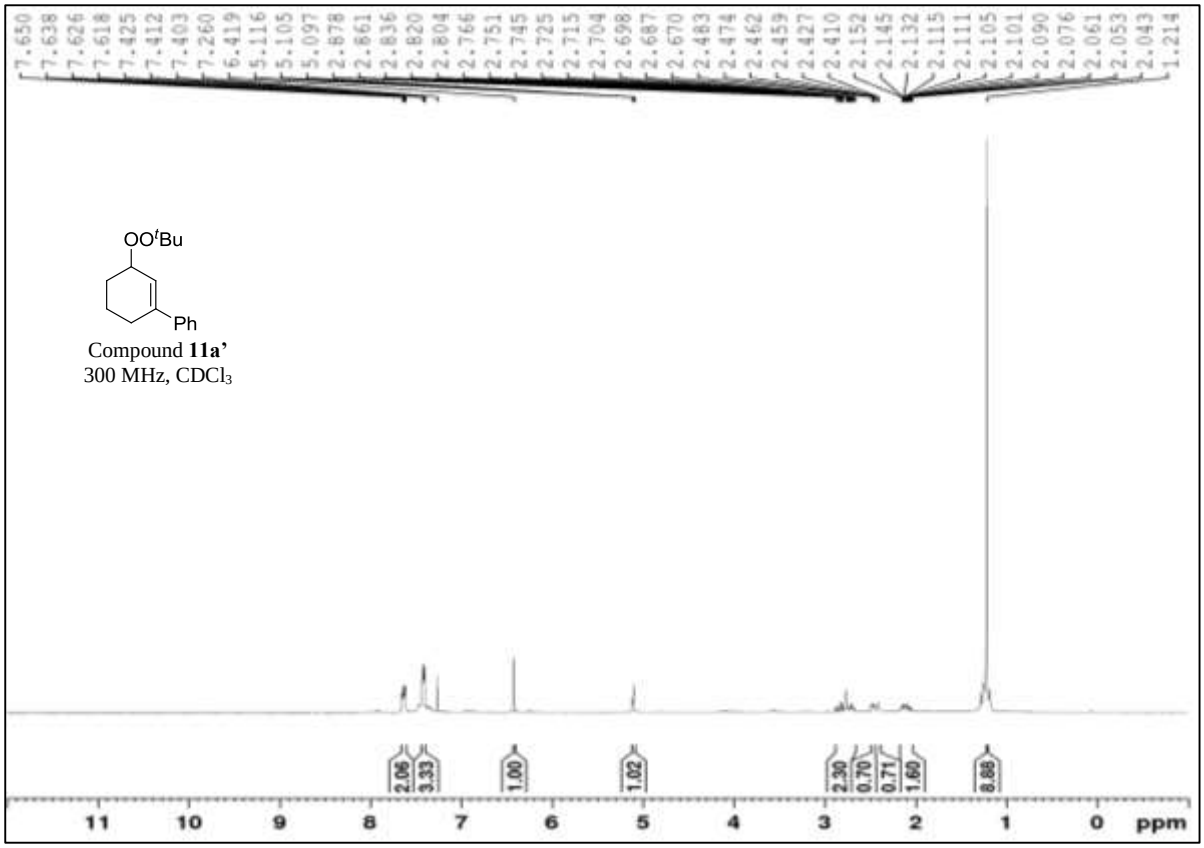
Compound **11k**
126 MHz, $CDCl_3$



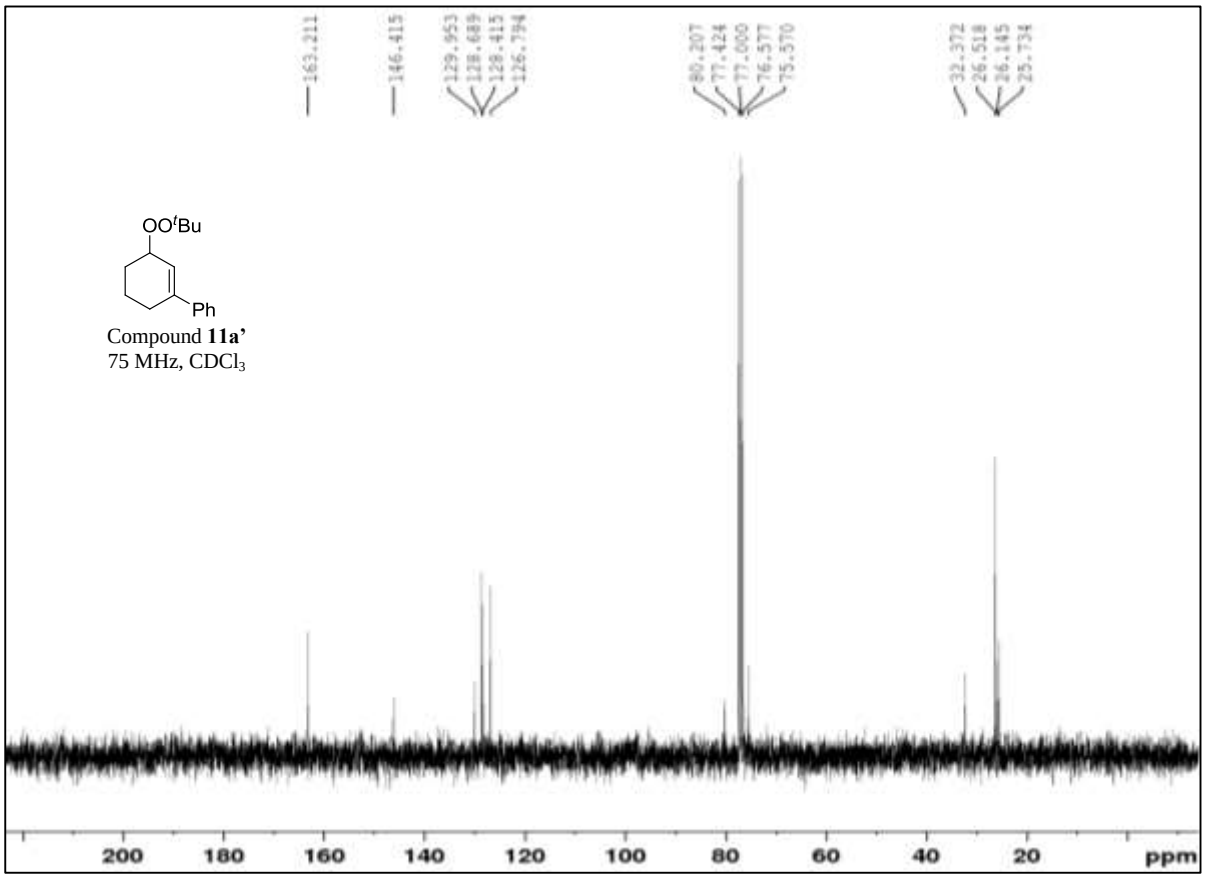
Compound **11l**
300 MHz, $CDCl_3$



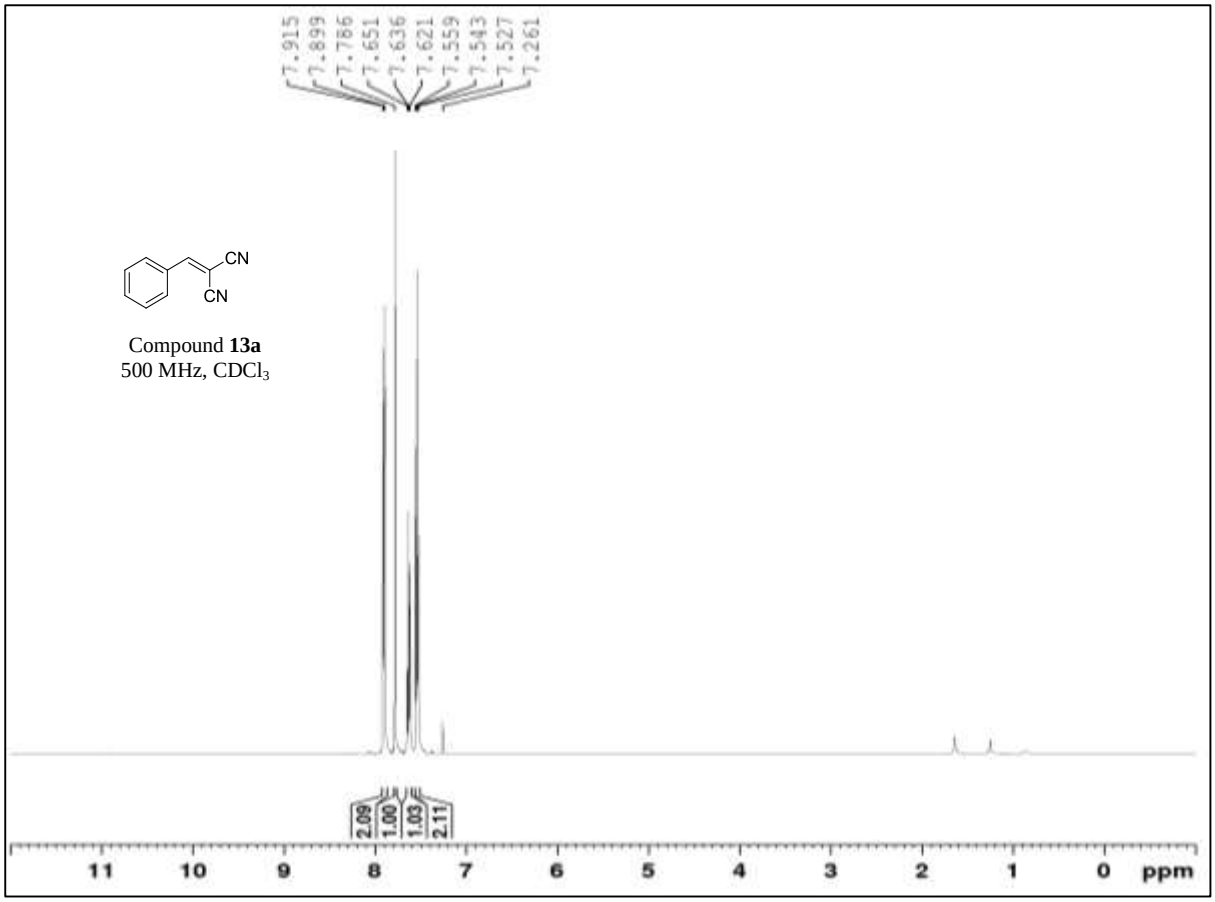
Compound **11l**
75 MHz, $CDCl_3$



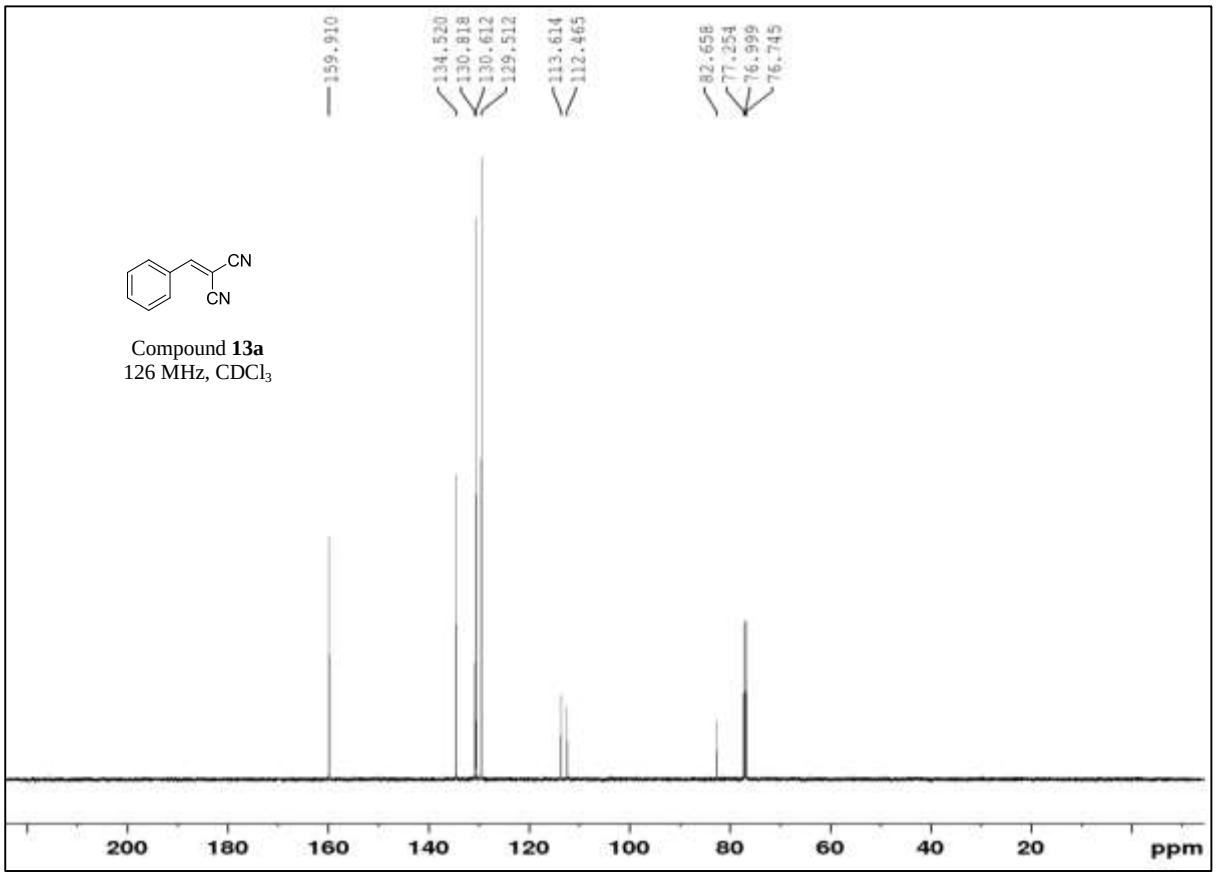
Compound **11a'**
300 MHz, $CDCl_3$



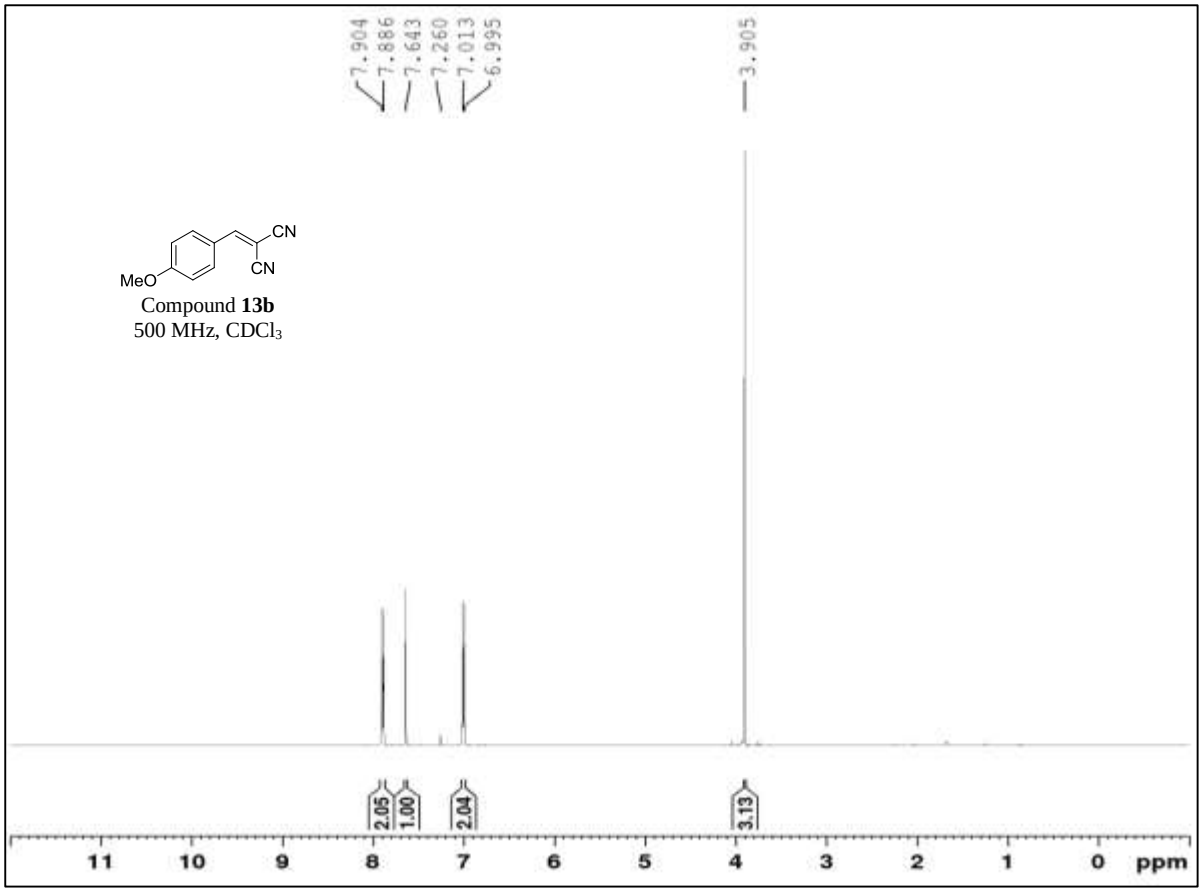
Compound **11a'**
75 MHz, $CDCl_3$



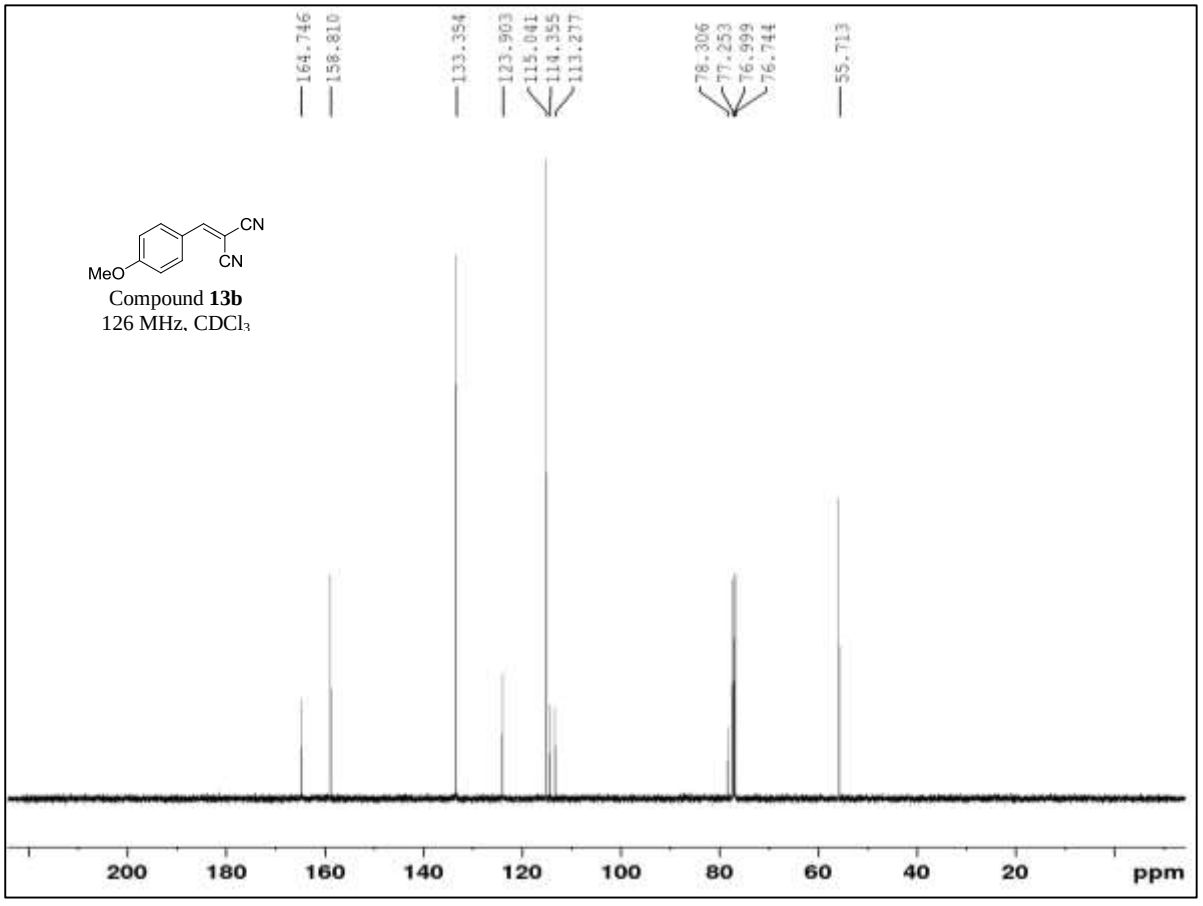
Compound **13a**
500 MHz, $CDCl_3$



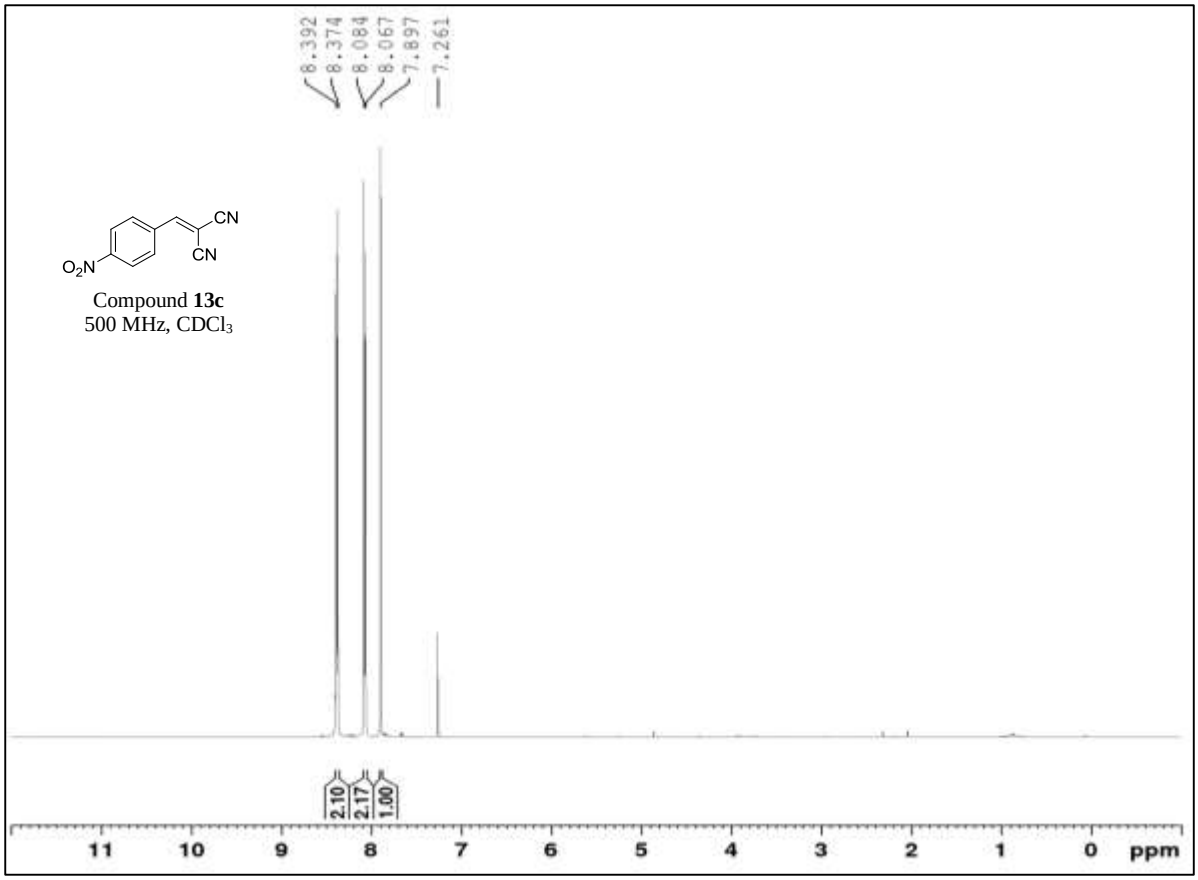
Compound **13a**
126 MHz, $CDCl_3$



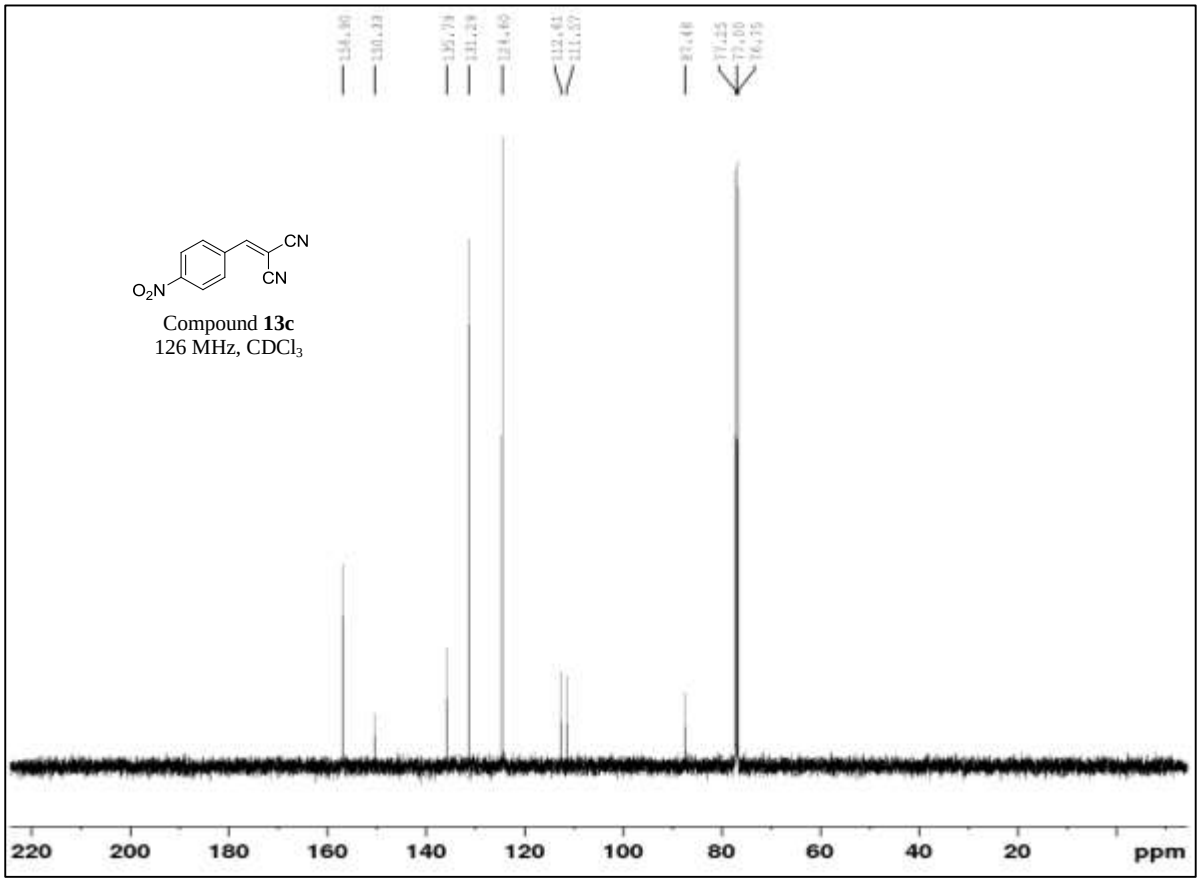
Compound **13b**
500 MHz, $CDCl_3$



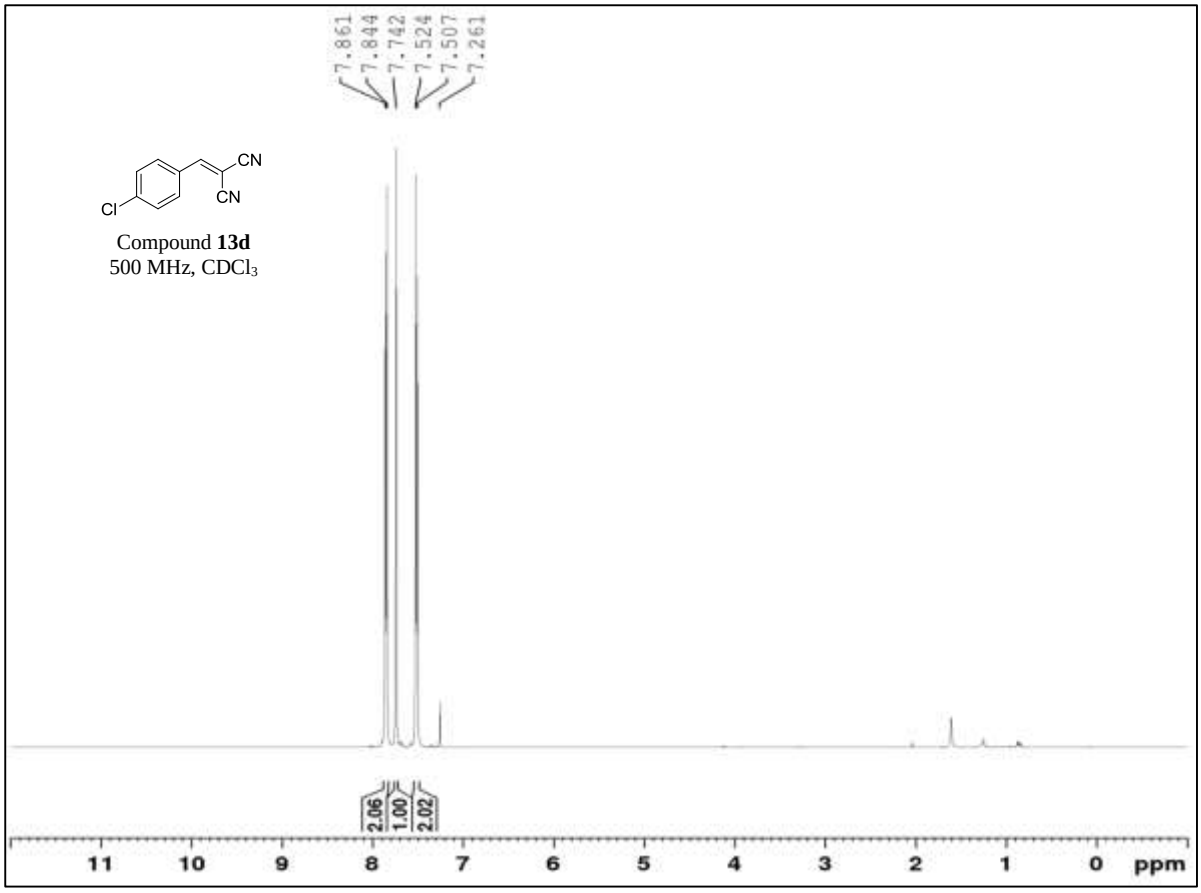
Compound **13b**
126 MHz, $CDCl_3$



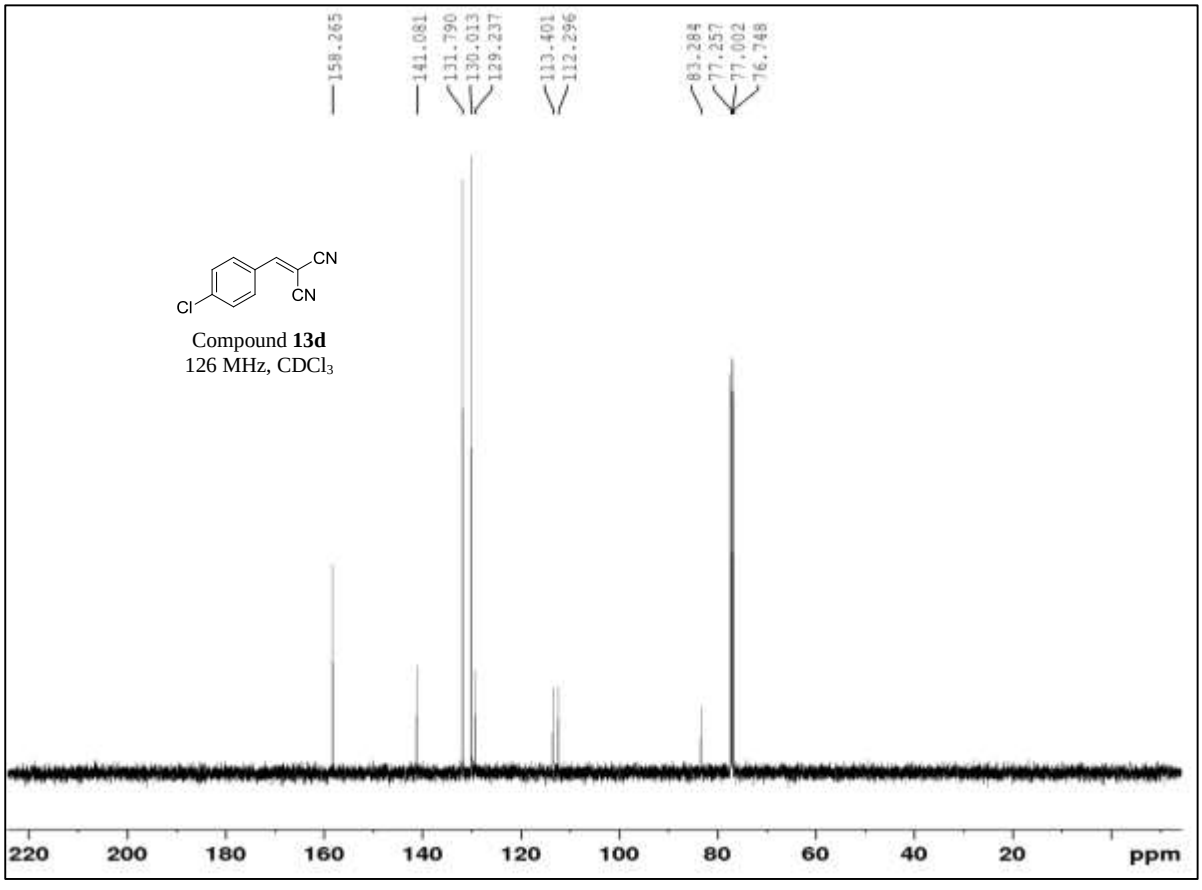
Compound **13c**
500 MHz, $CDCl_3$



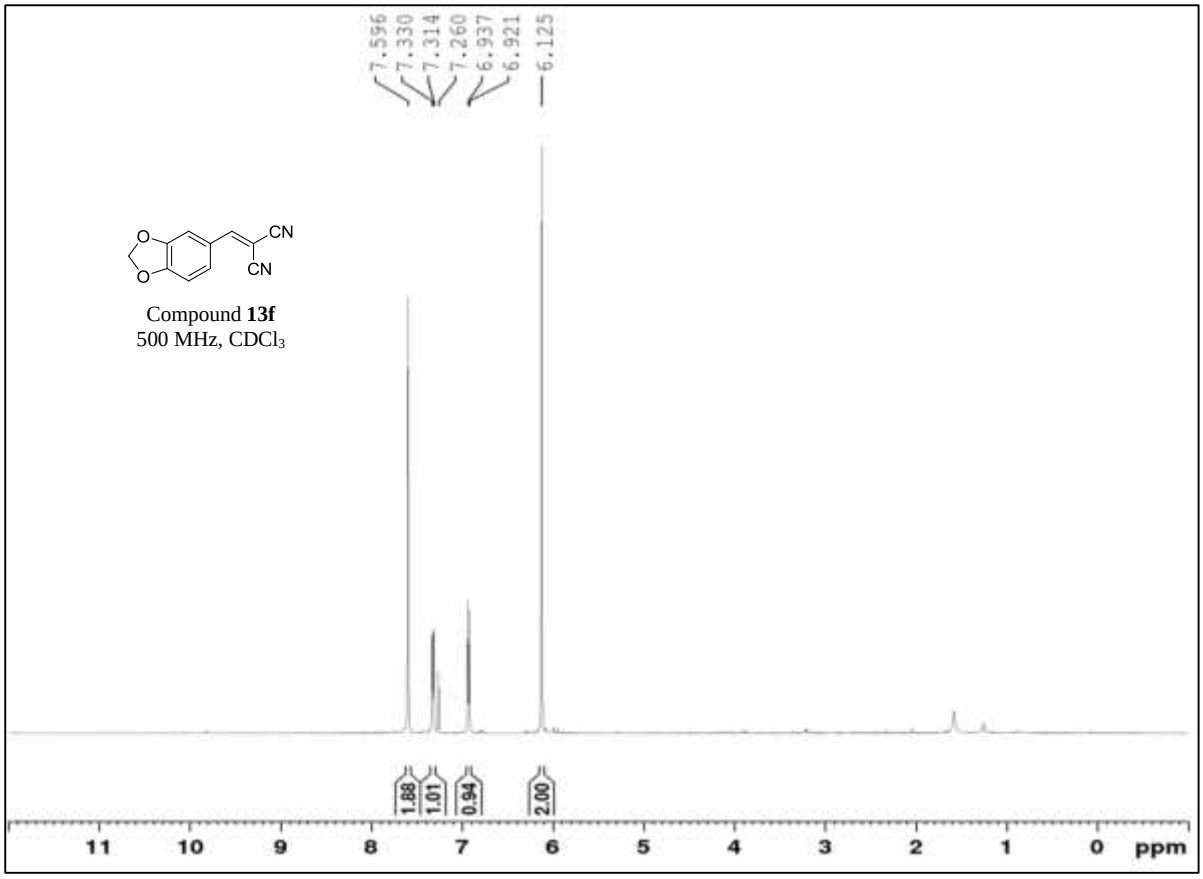
Compound **13c**
126 MHz, $CDCl_3$



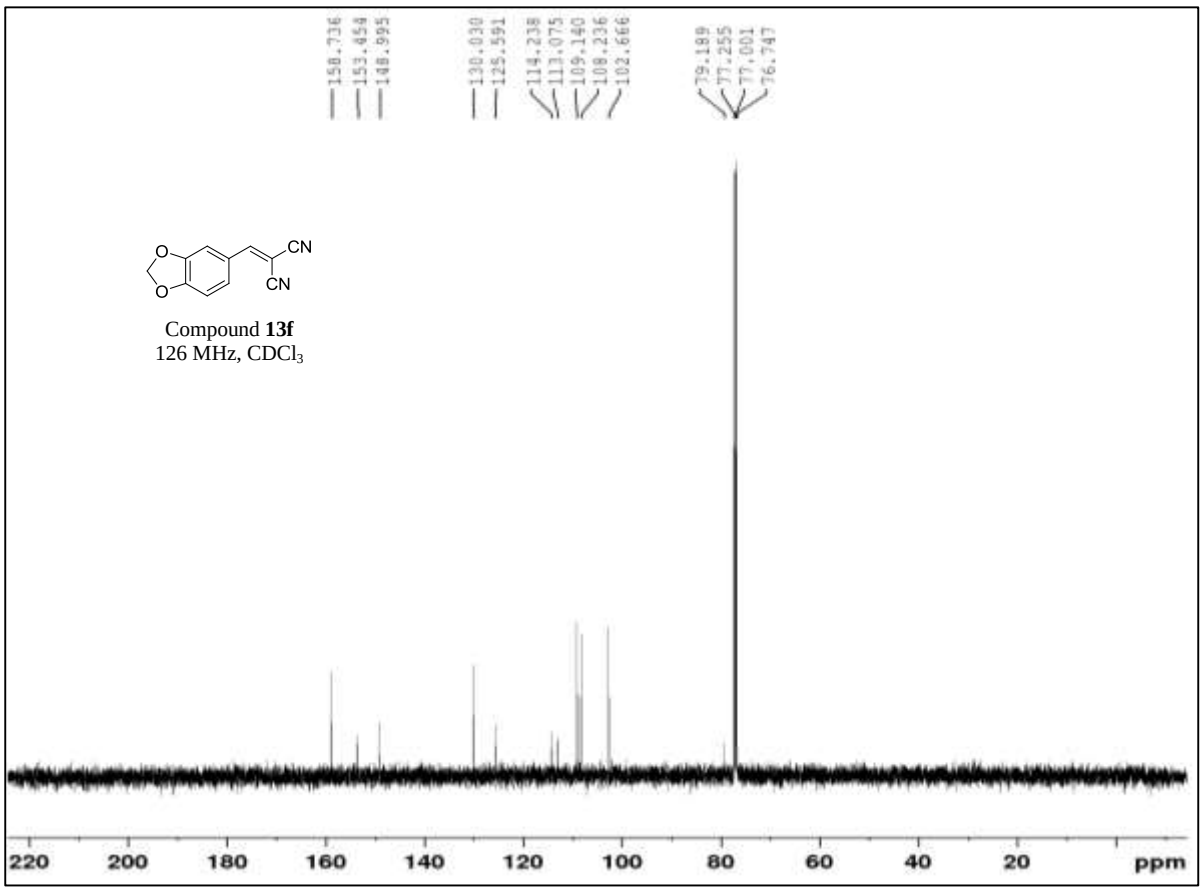
Compound **13d**
500 MHz, $CDCl_3$



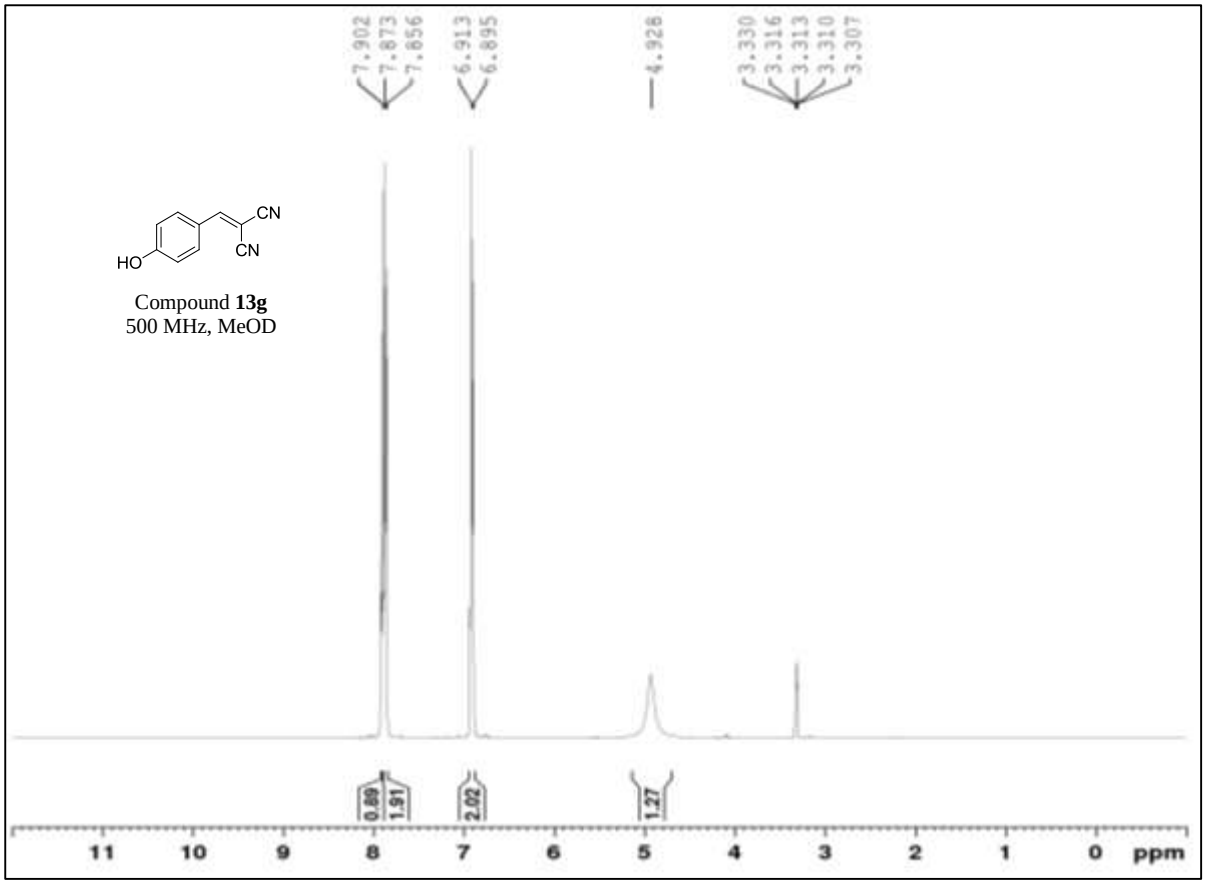
Compound **13d**
126 MHz, $CDCl_3$



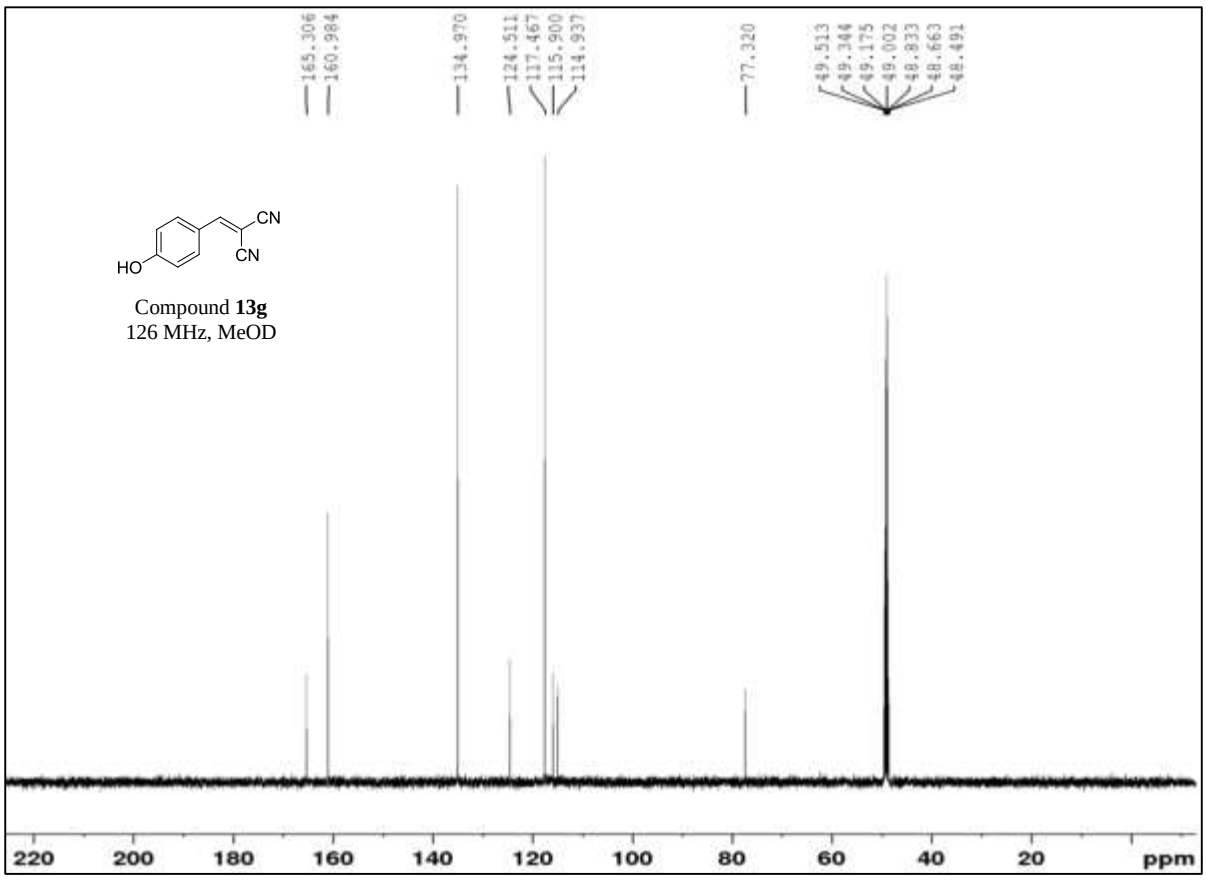
Compound **13f**
500 MHz, $CDCl_3$



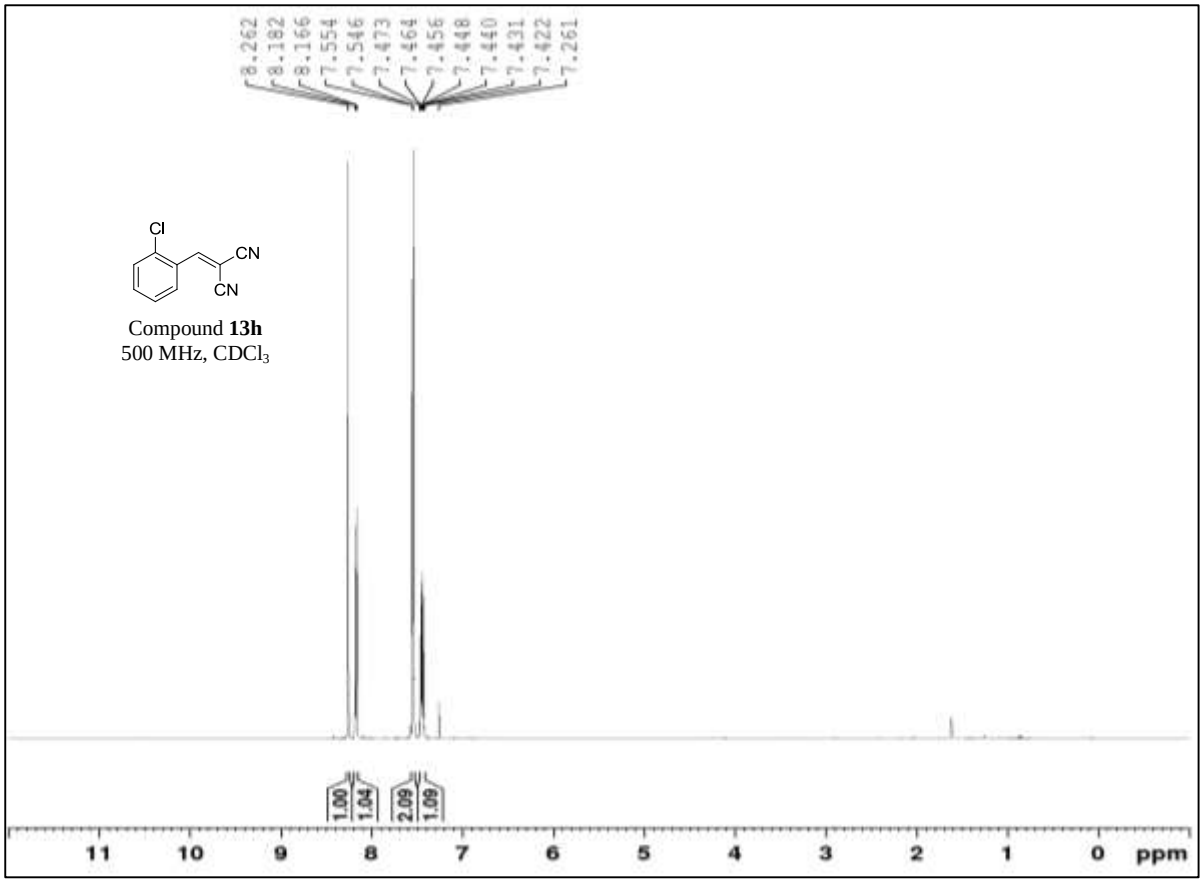
Compound **13f**
126 MHz, $CDCl_3$



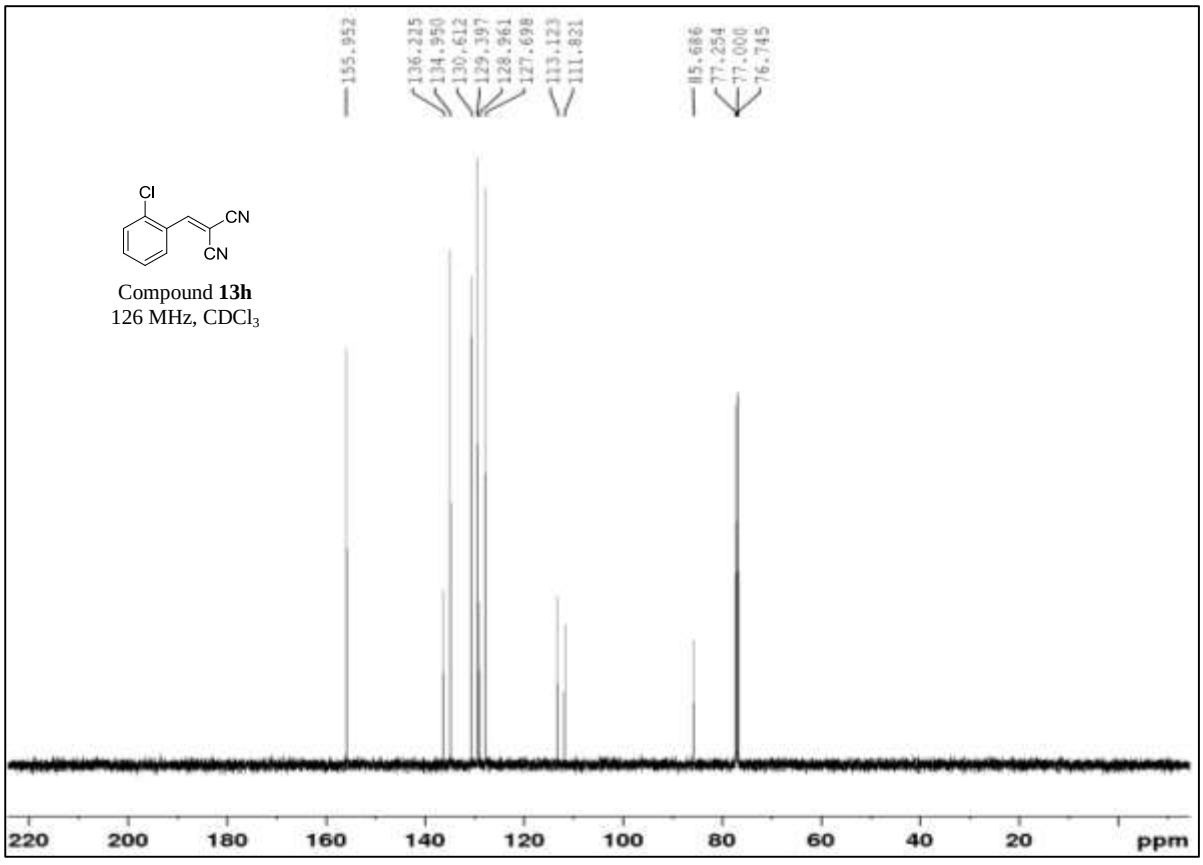
Compound **13g**
500 MHz, MeOD



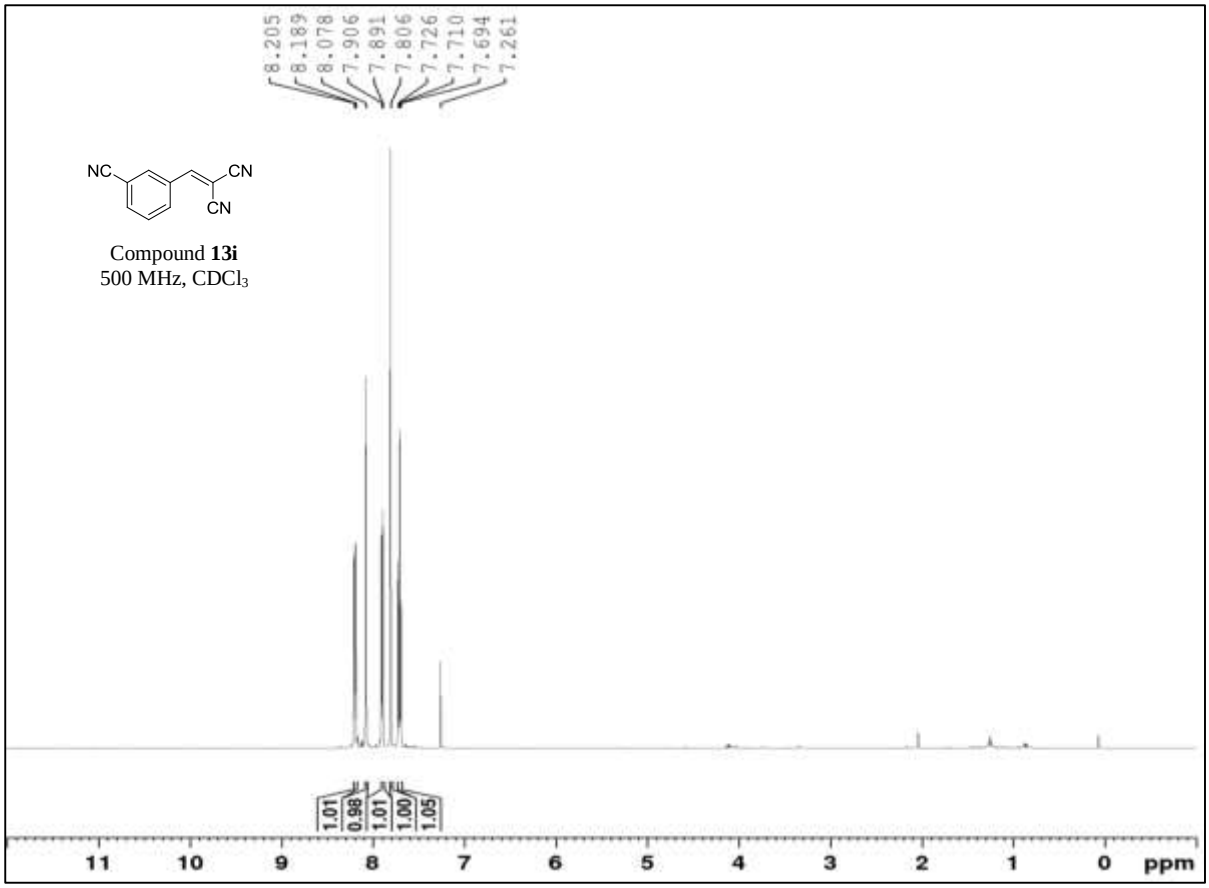
Compound **13g**
126 MHz, MeOD



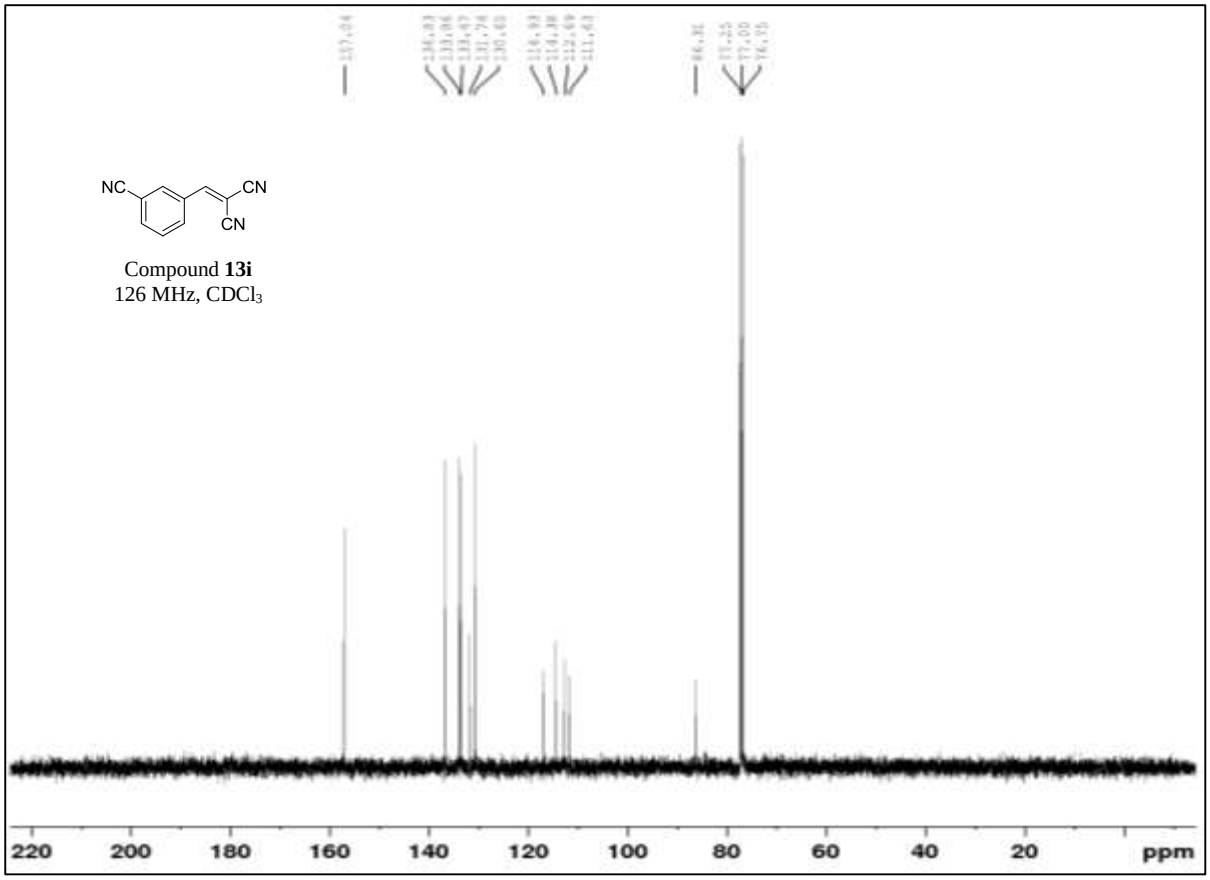
Compound **13h**
500 MHz, $CDCl_3$



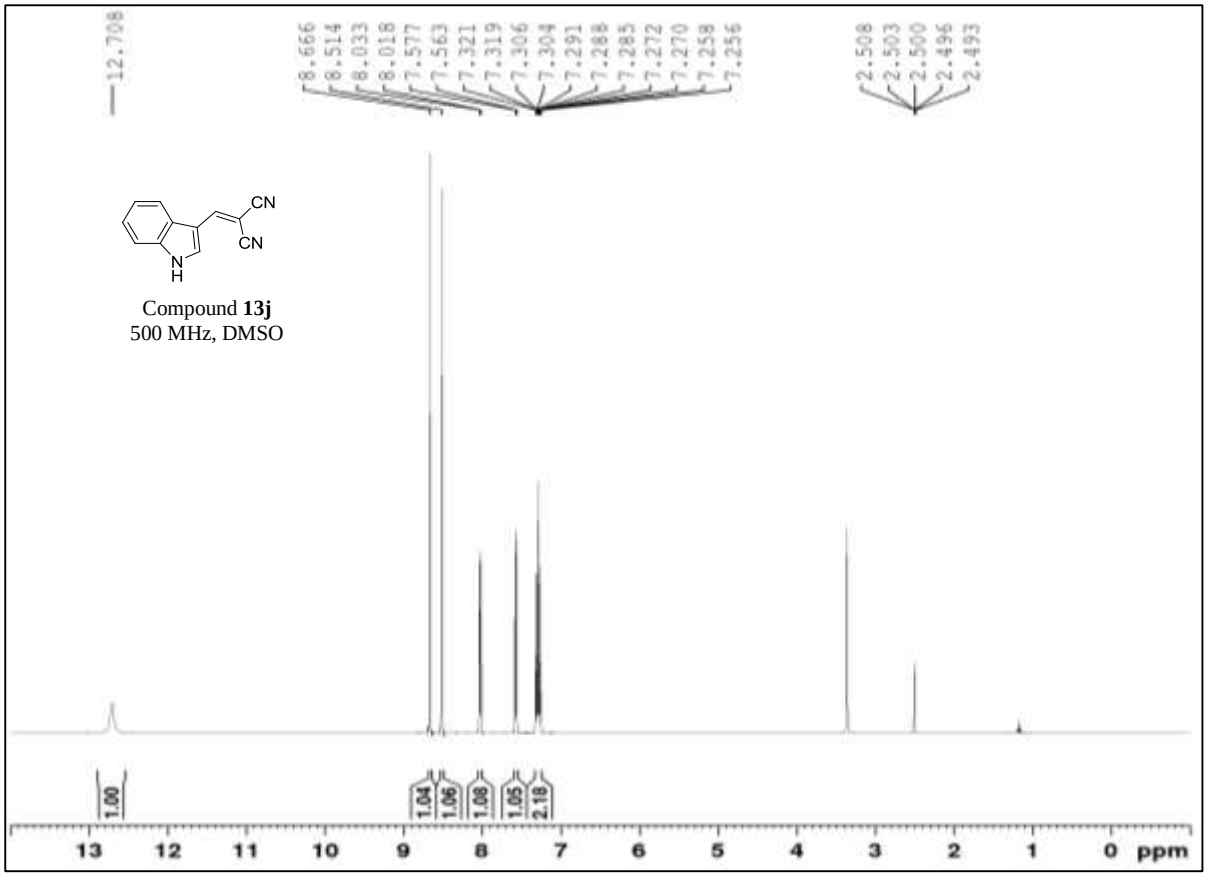
Compound **13h**
126 MHz, $CDCl_3$



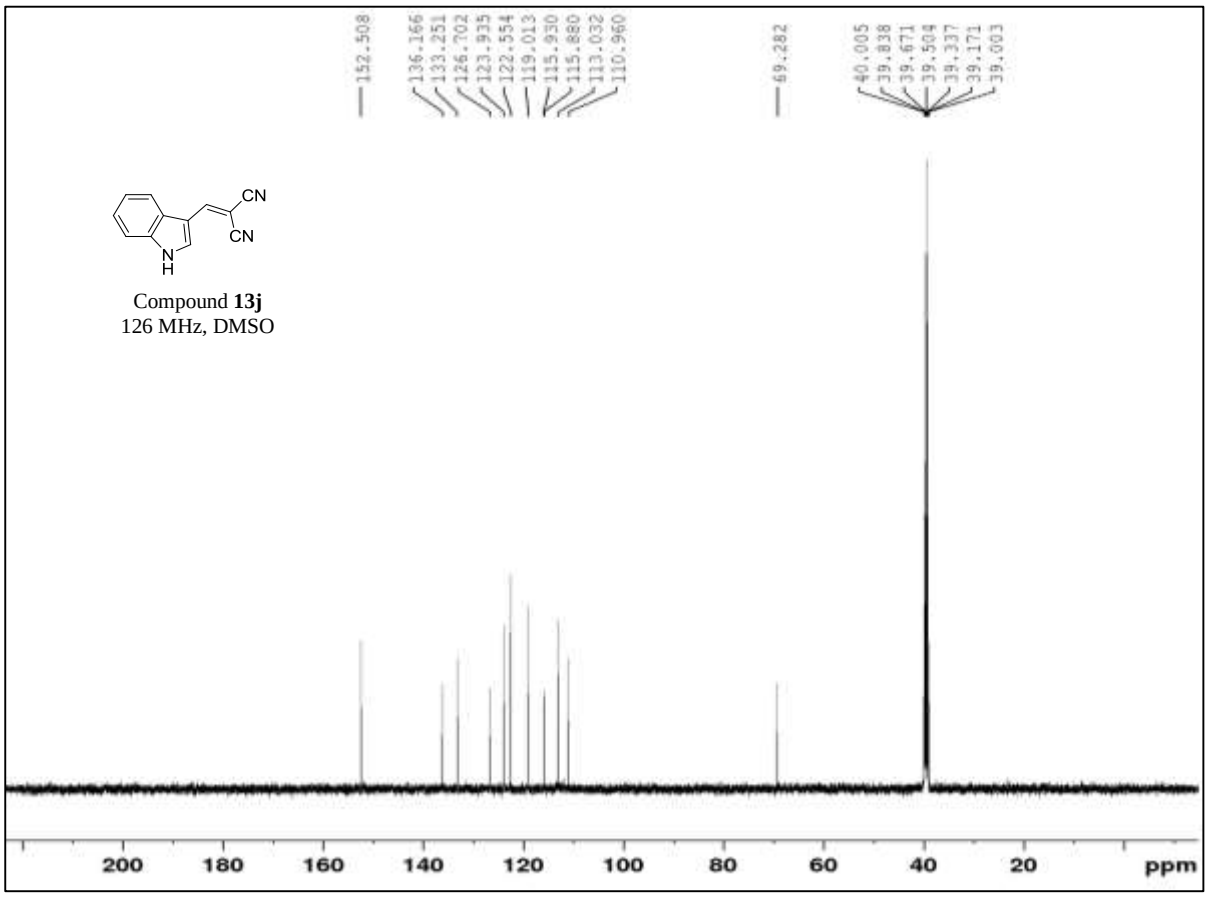
Compound **13i**
500 MHz, $CDCl_3$



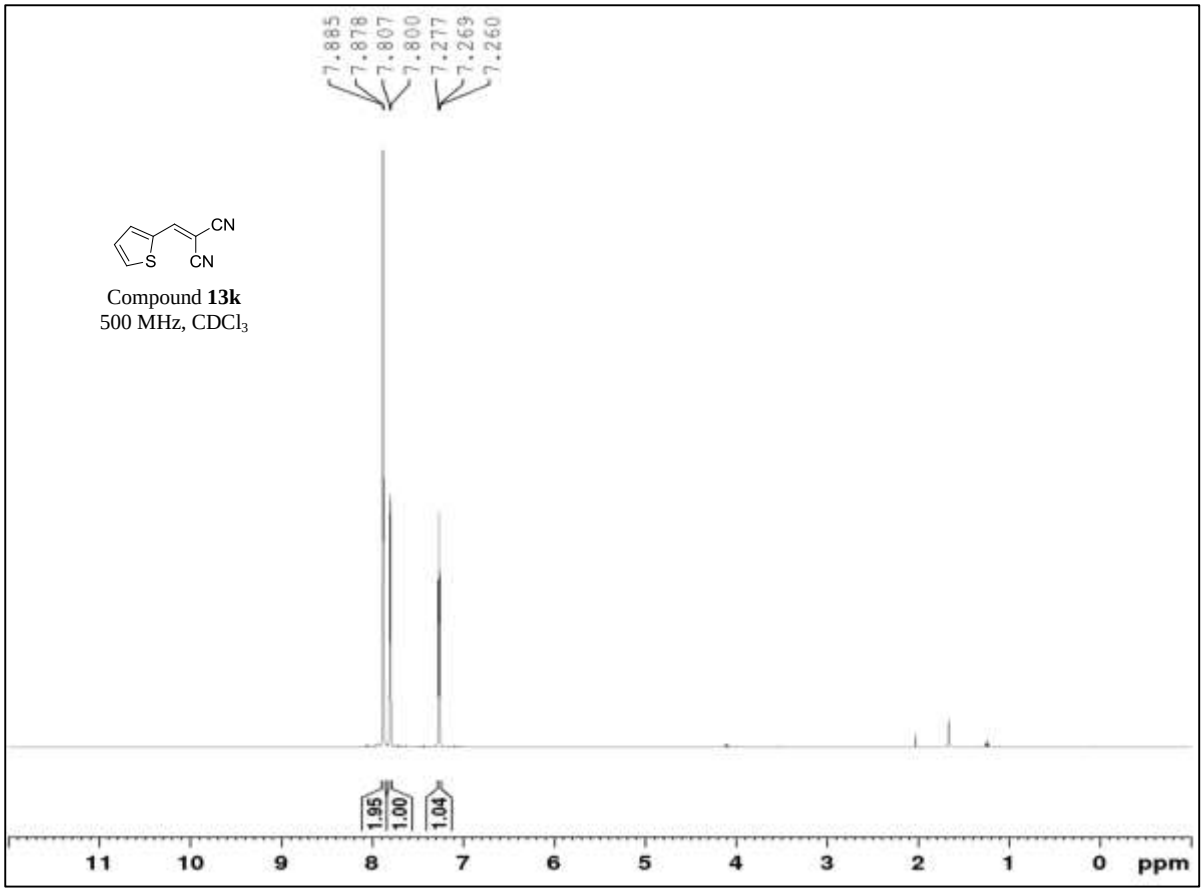
Compound **13i**
126 MHz, $CDCl_3$



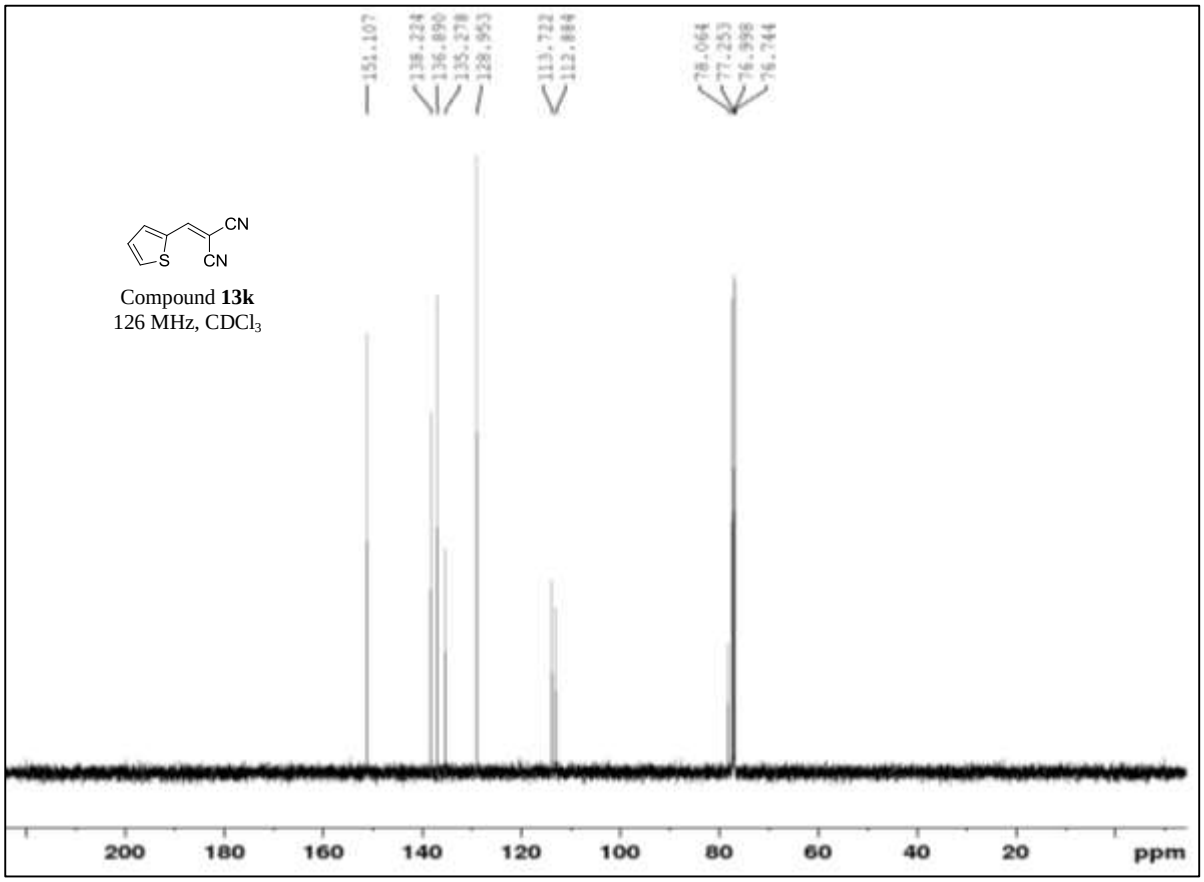
Compound **13j**
500 MHz, DMSO



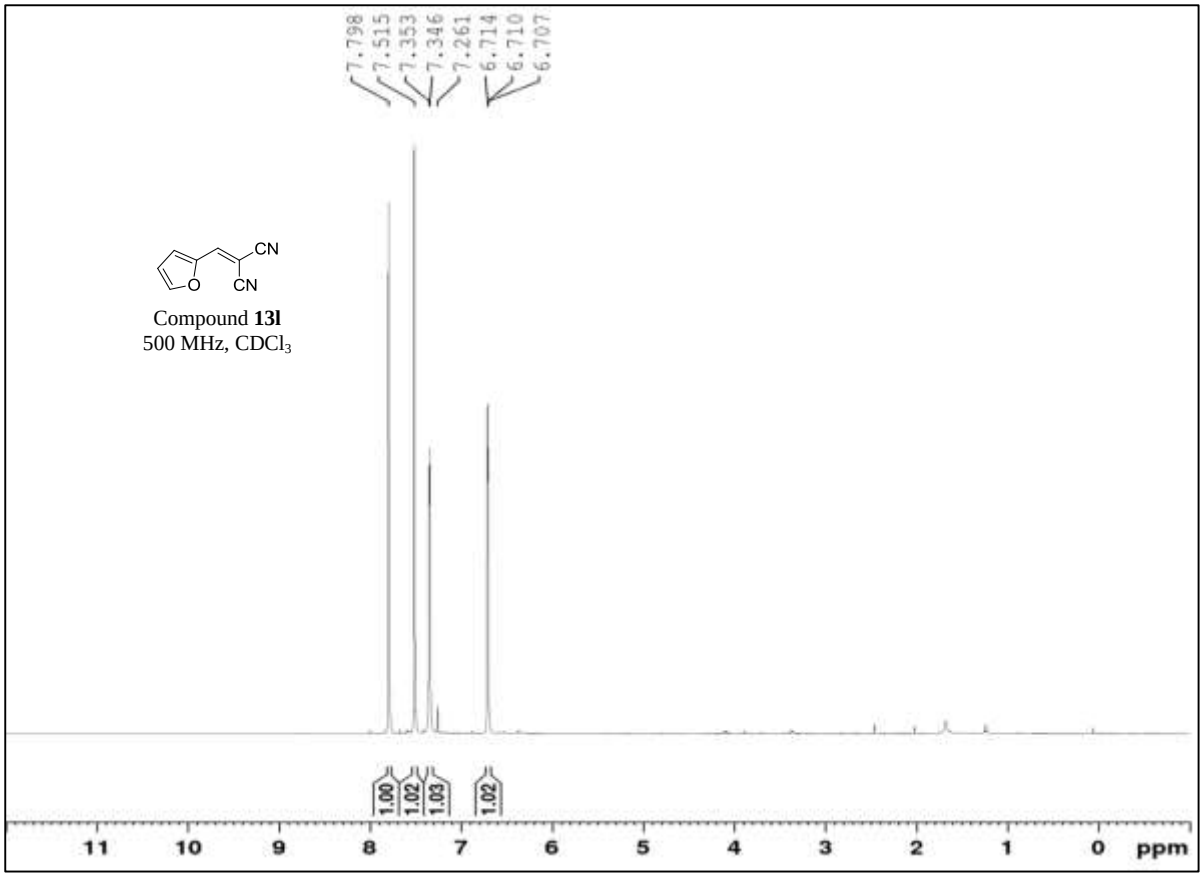
Compound **13j**
126 MHz, DMSO



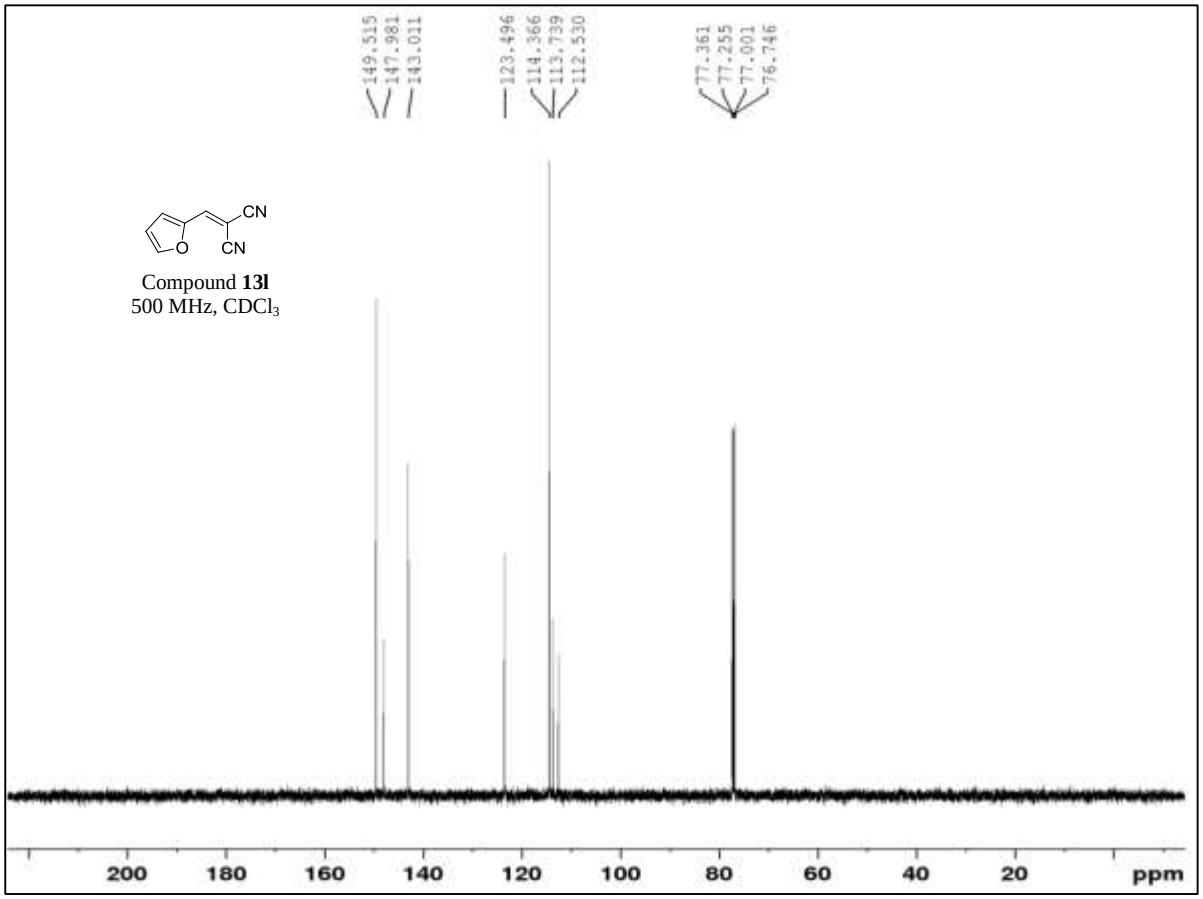
Compound **13k**
500 MHz, $CDCl_3$



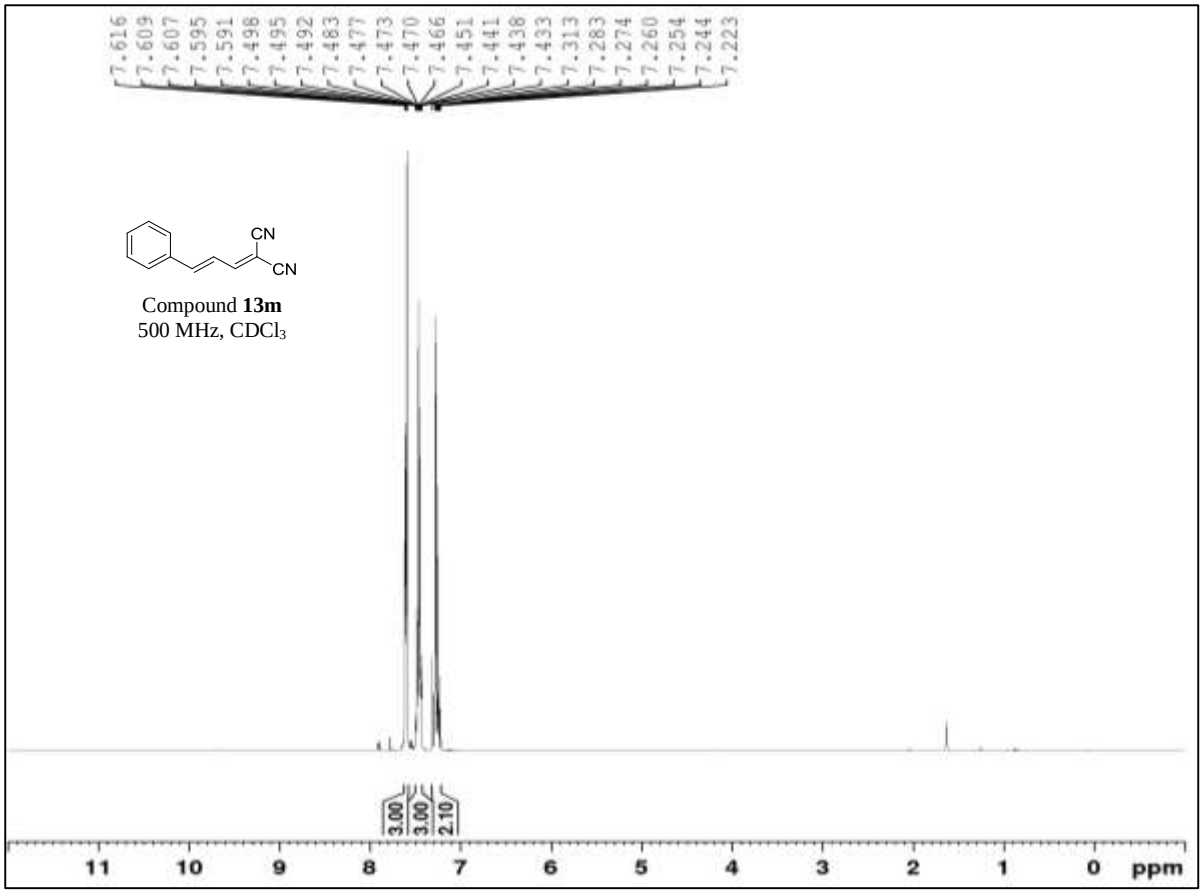
Compound **13k**
126 MHz, $CDCl_3$



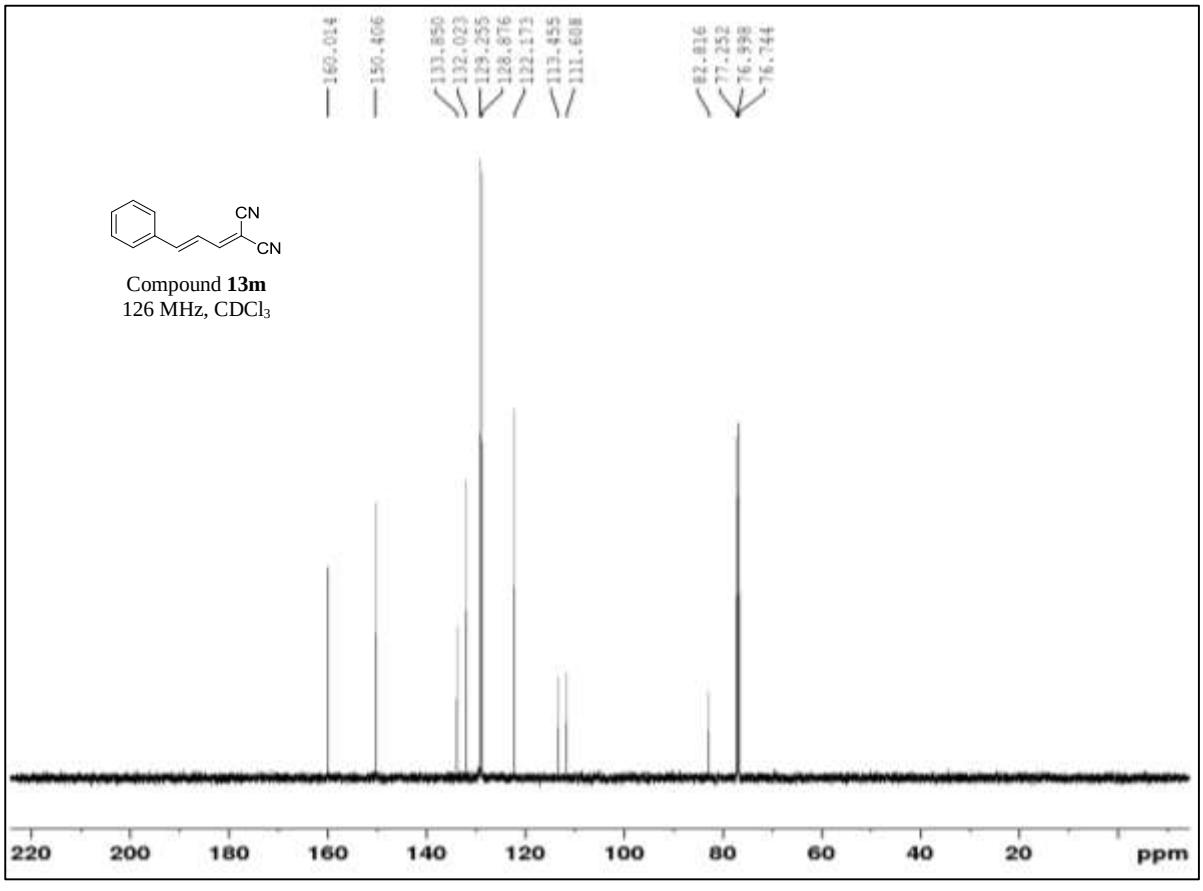
Compound **13l**
500 MHz, $CDCl_3$



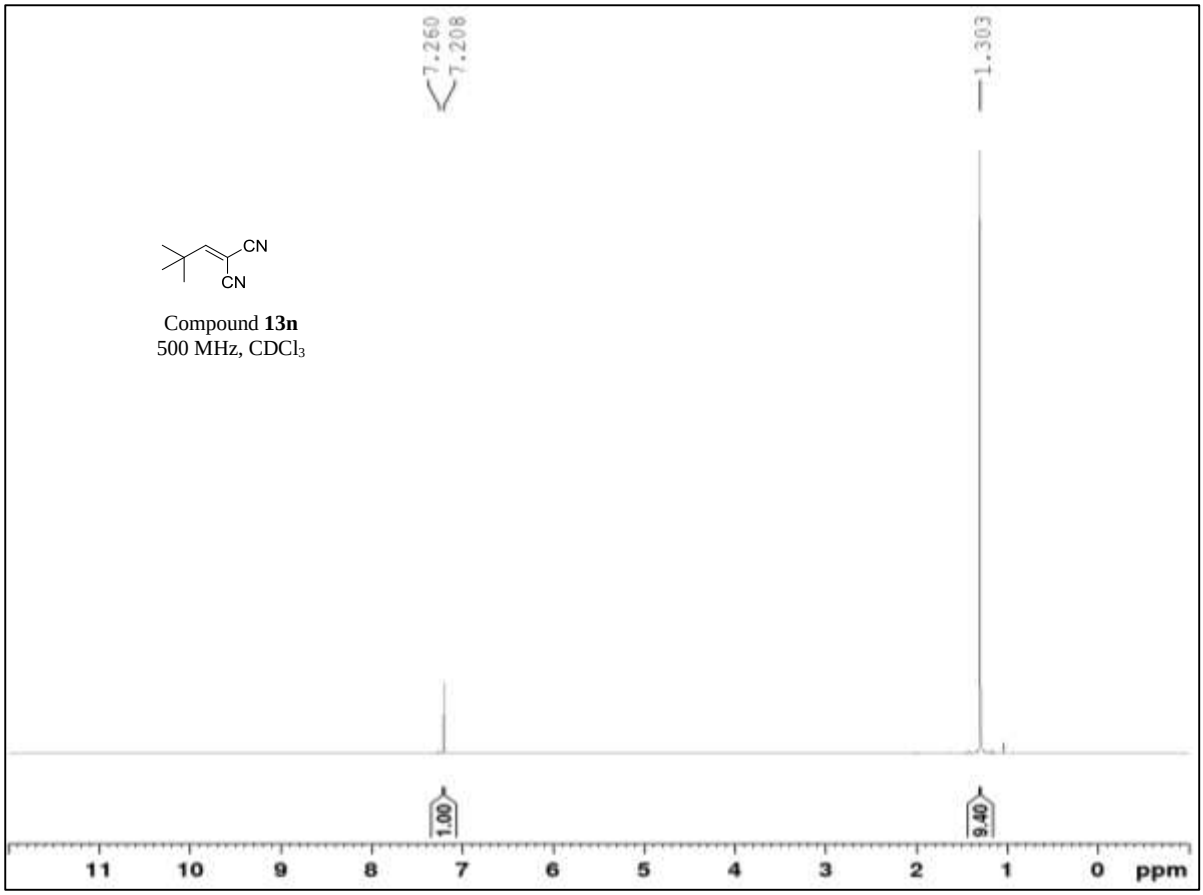
Compound **13l**
500 MHz, $CDCl_3$



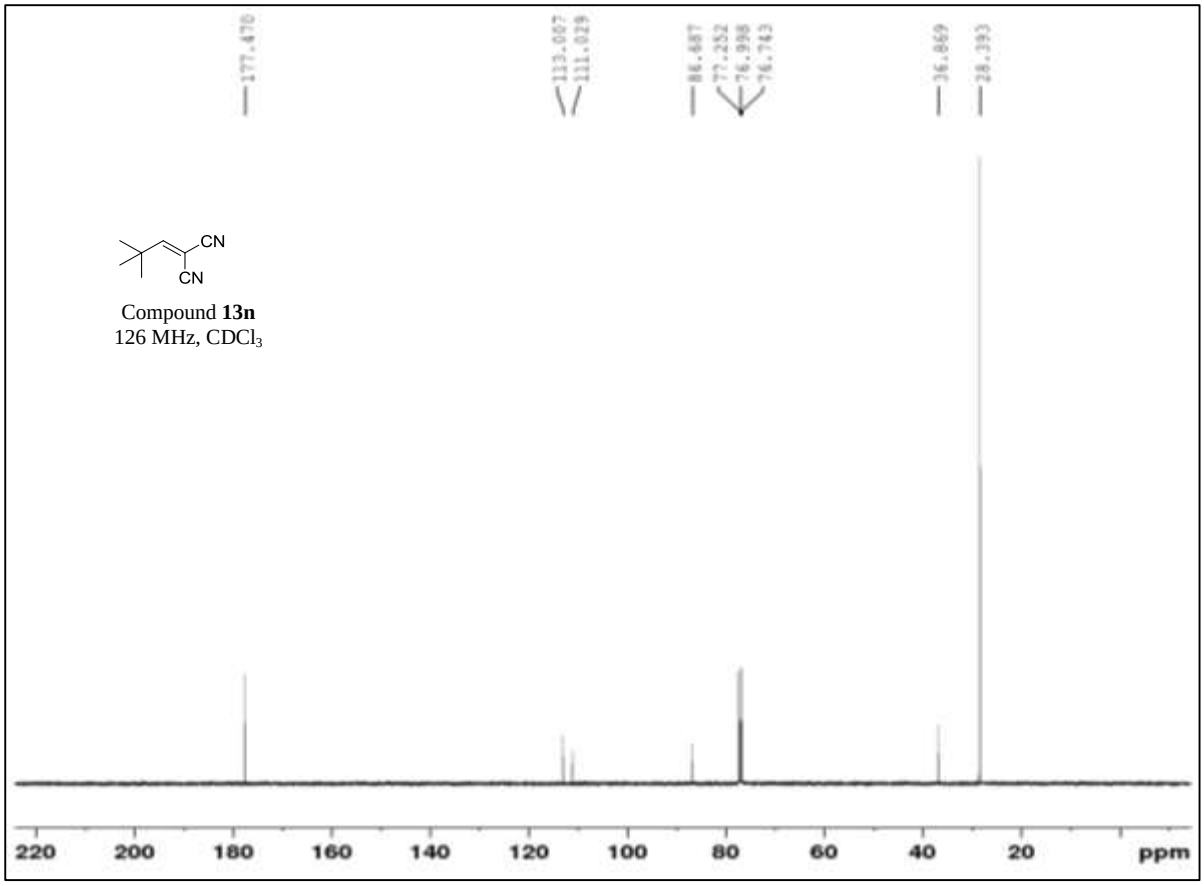
Compound **13m**
500 MHz, $CDCl_3$



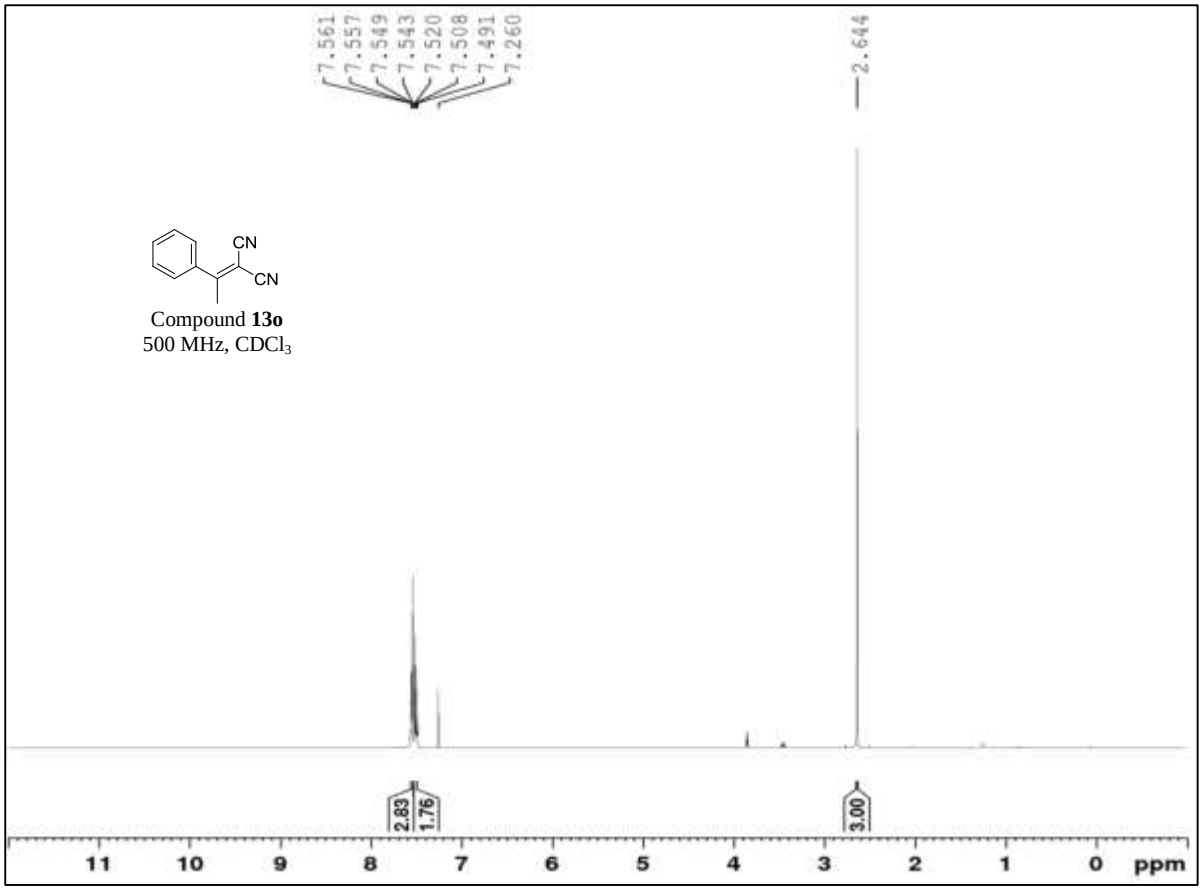
Compound **13m**
126 MHz, $CDCl_3$



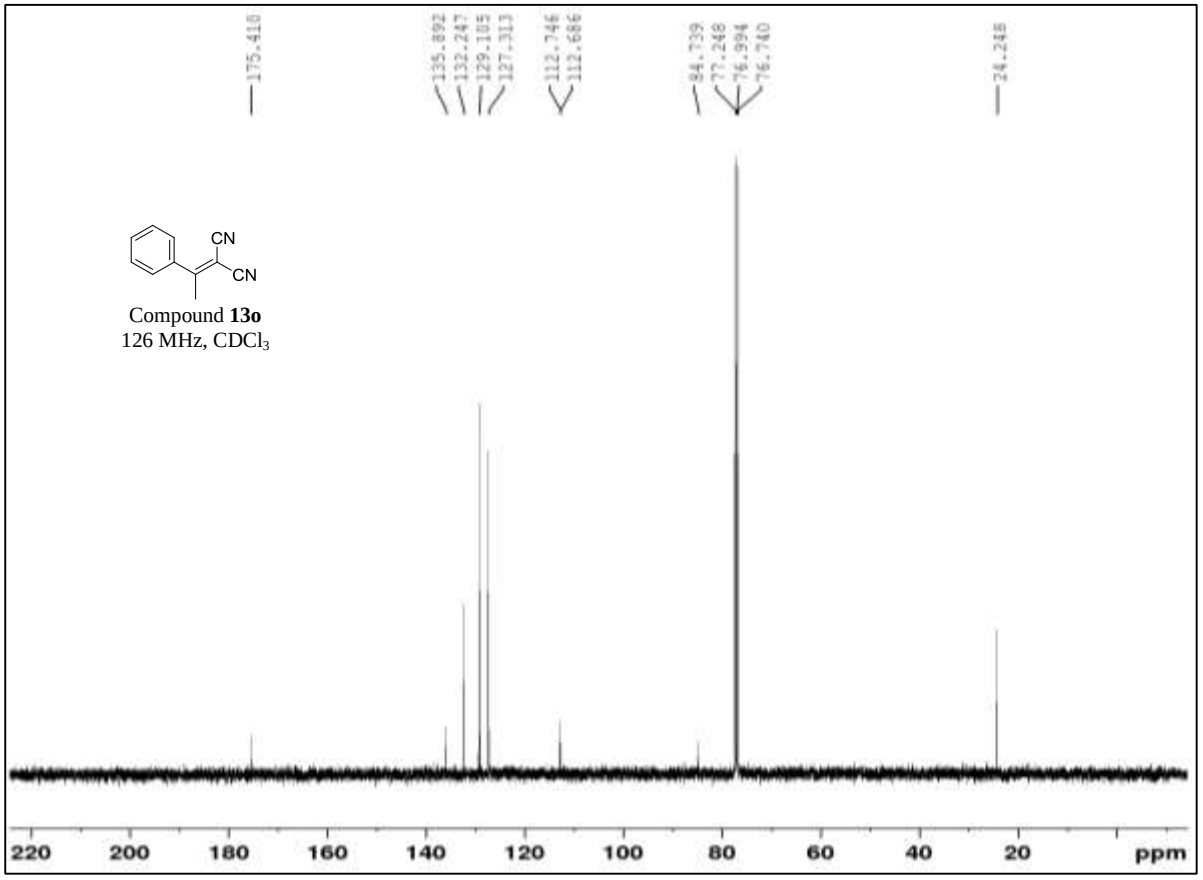
Compound **13n**
500 MHz, $CDCl_3$



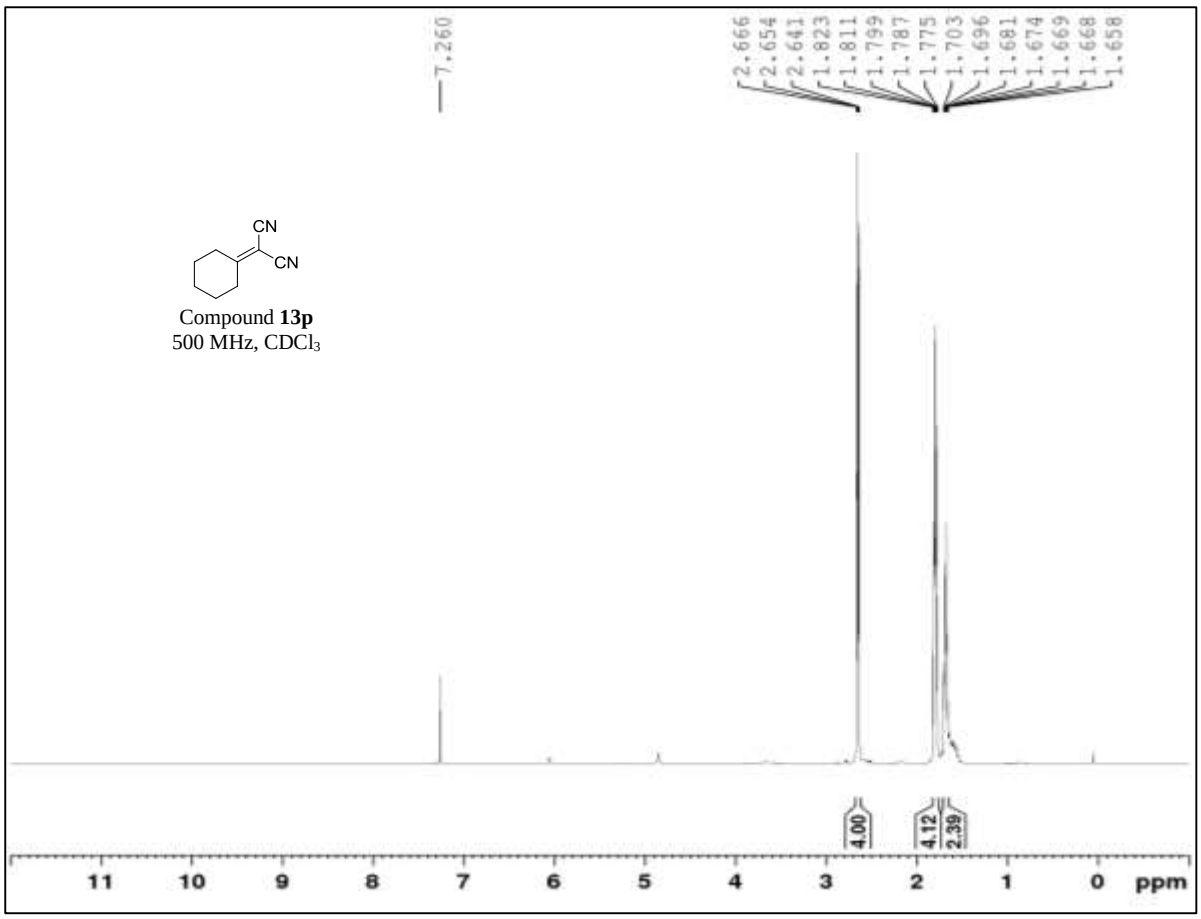
Compound **13n**
126 MHz, $CDCl_3$



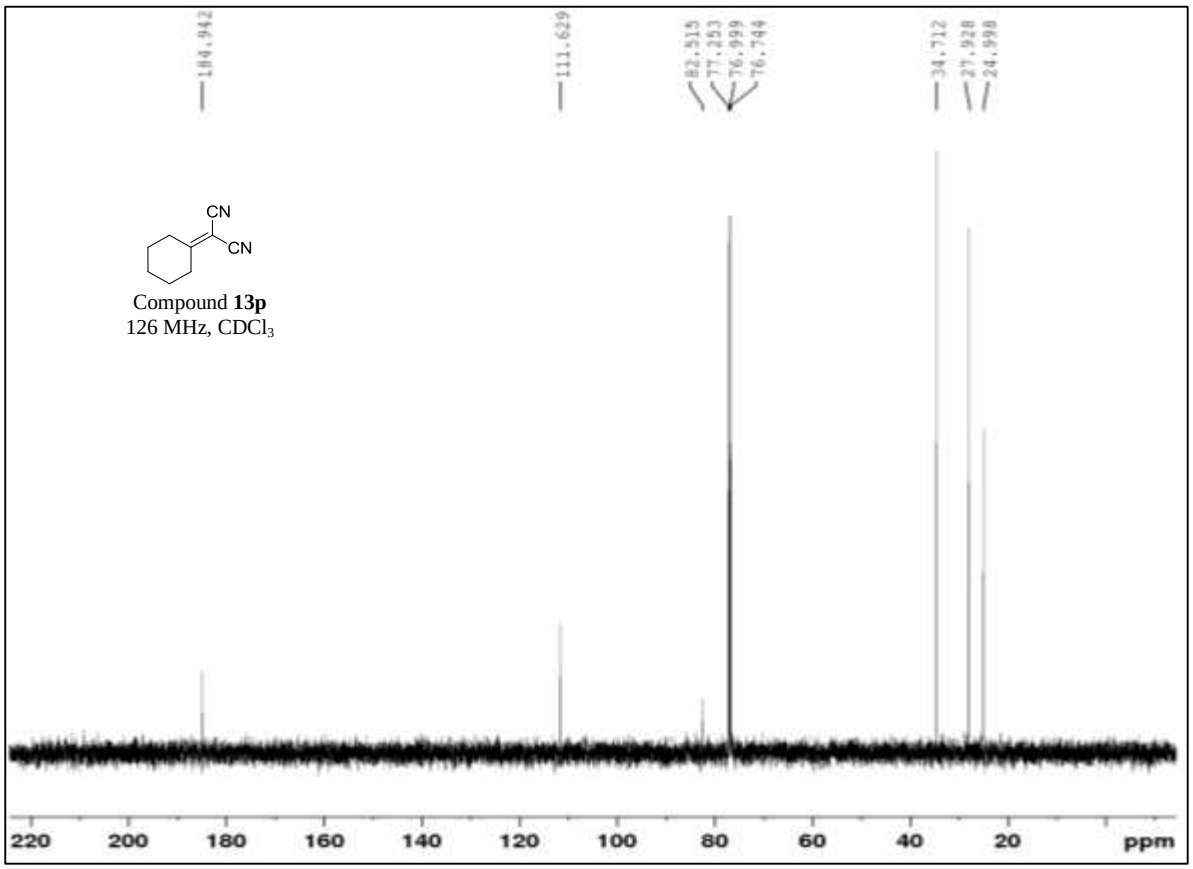
Compound **13o**
500 MHz, $CDCl_3$



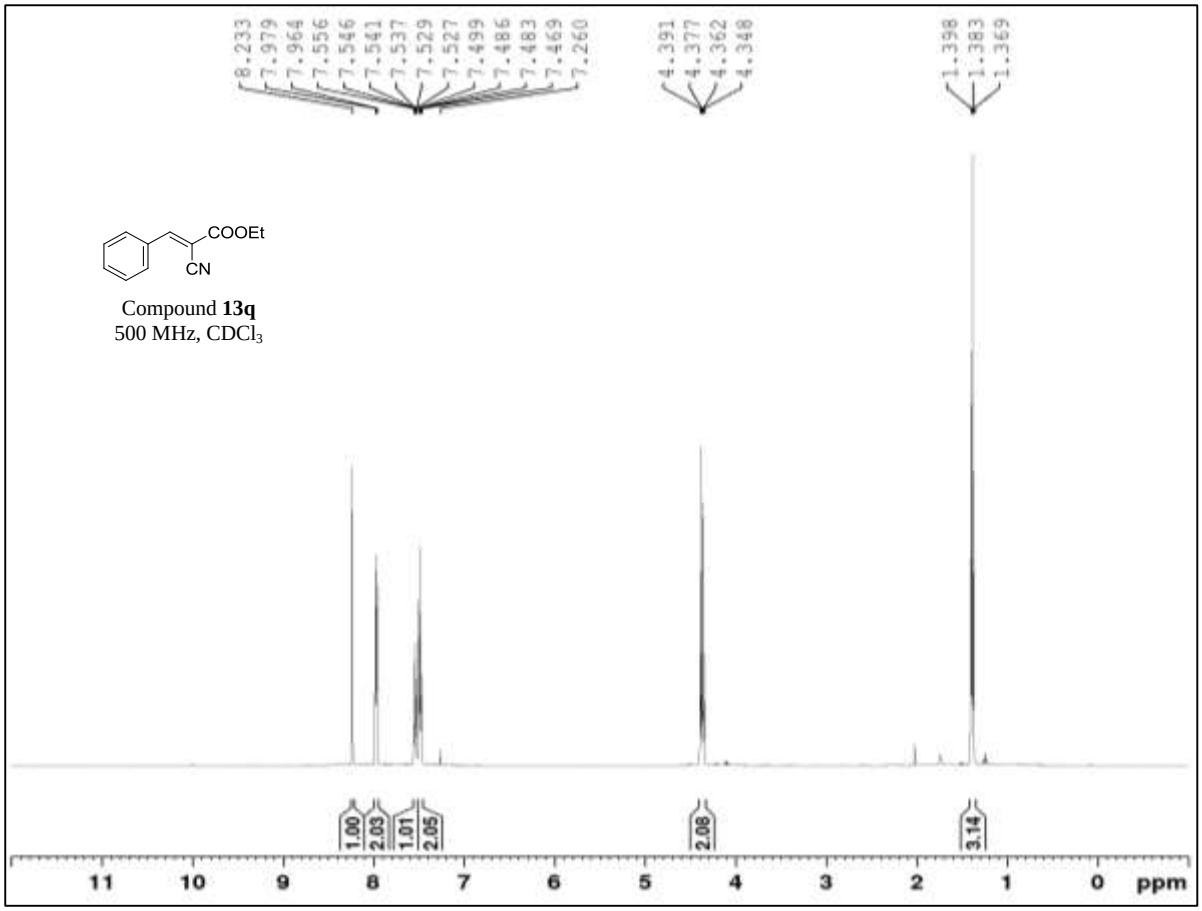
Compound **13o**
126 MHz, $CDCl_3$



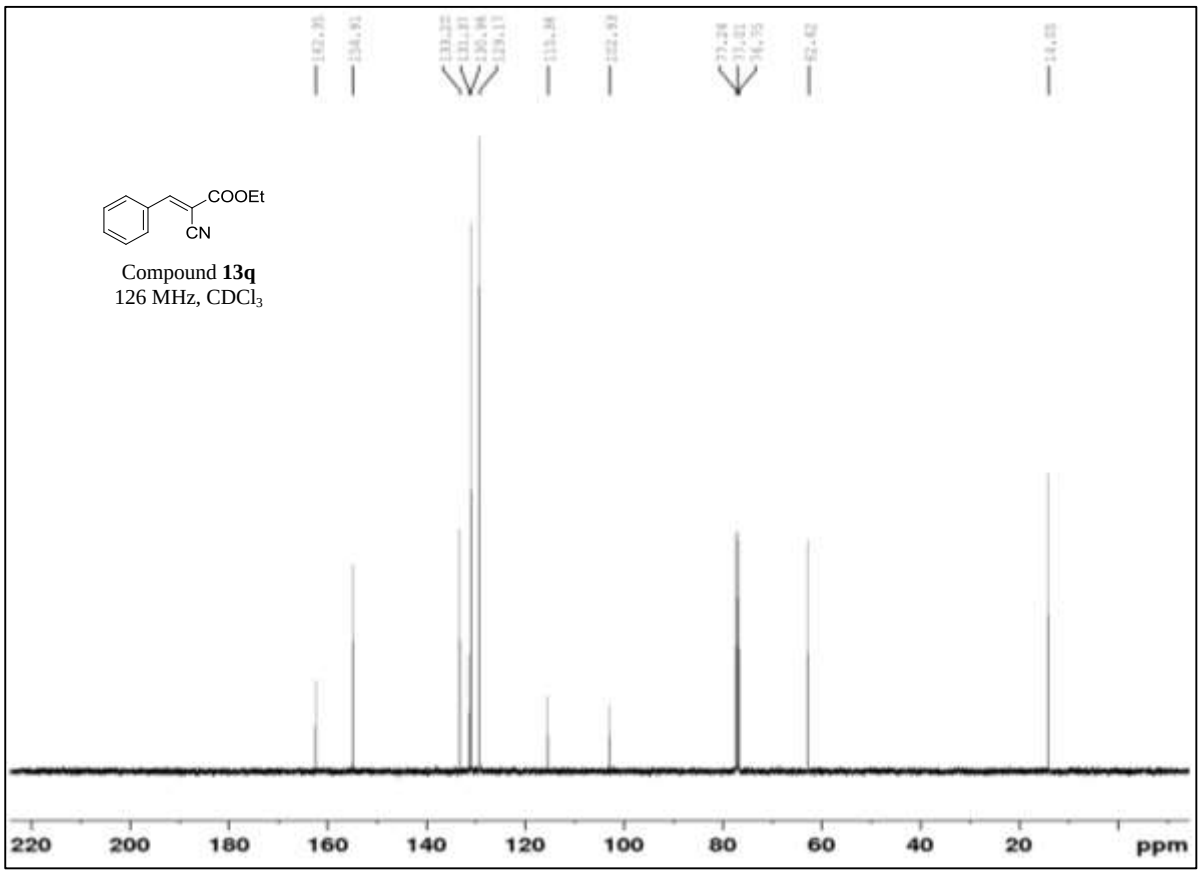
Compound **13p**
500 MHz, $CDCl_3$



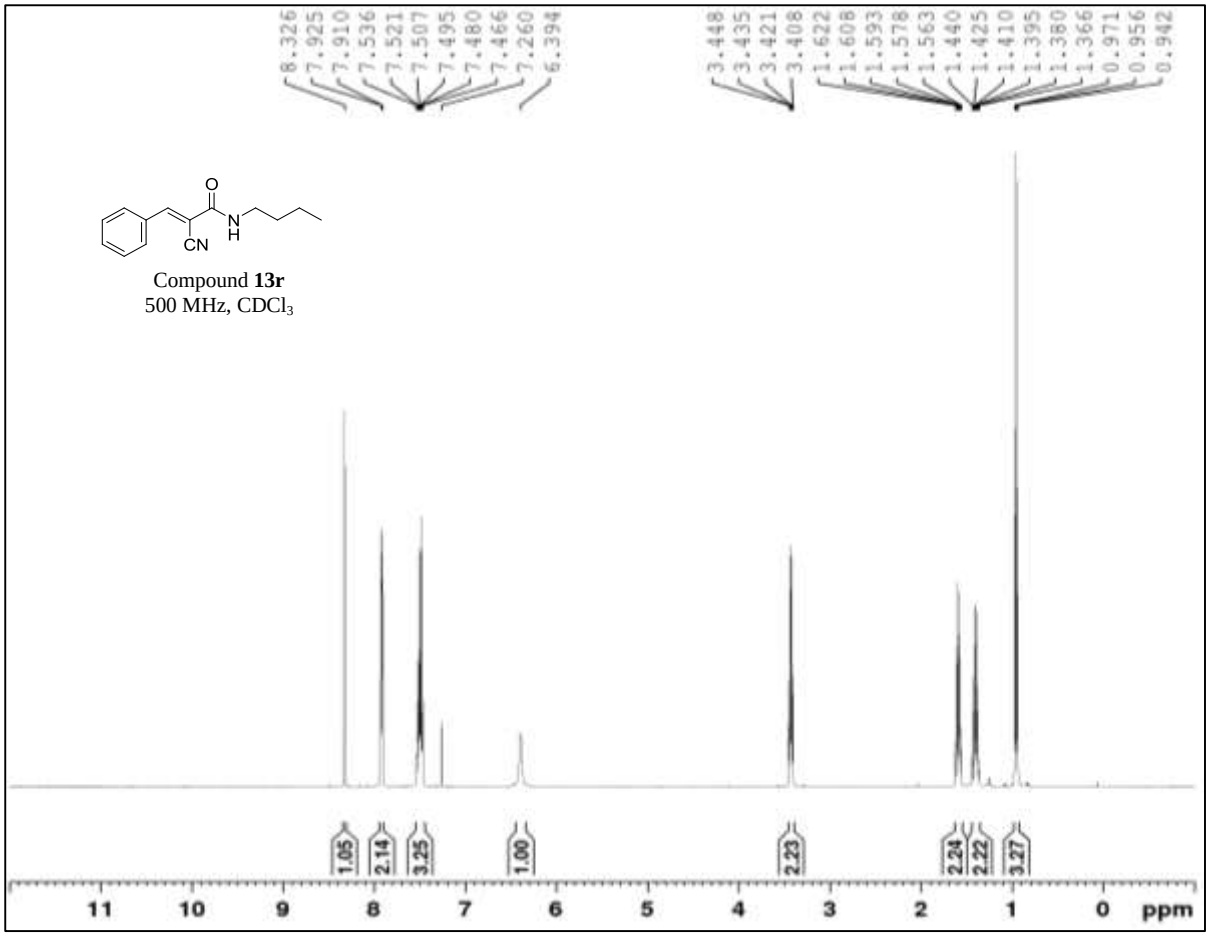
Compound **13p**
126 MHz, $CDCl_3$



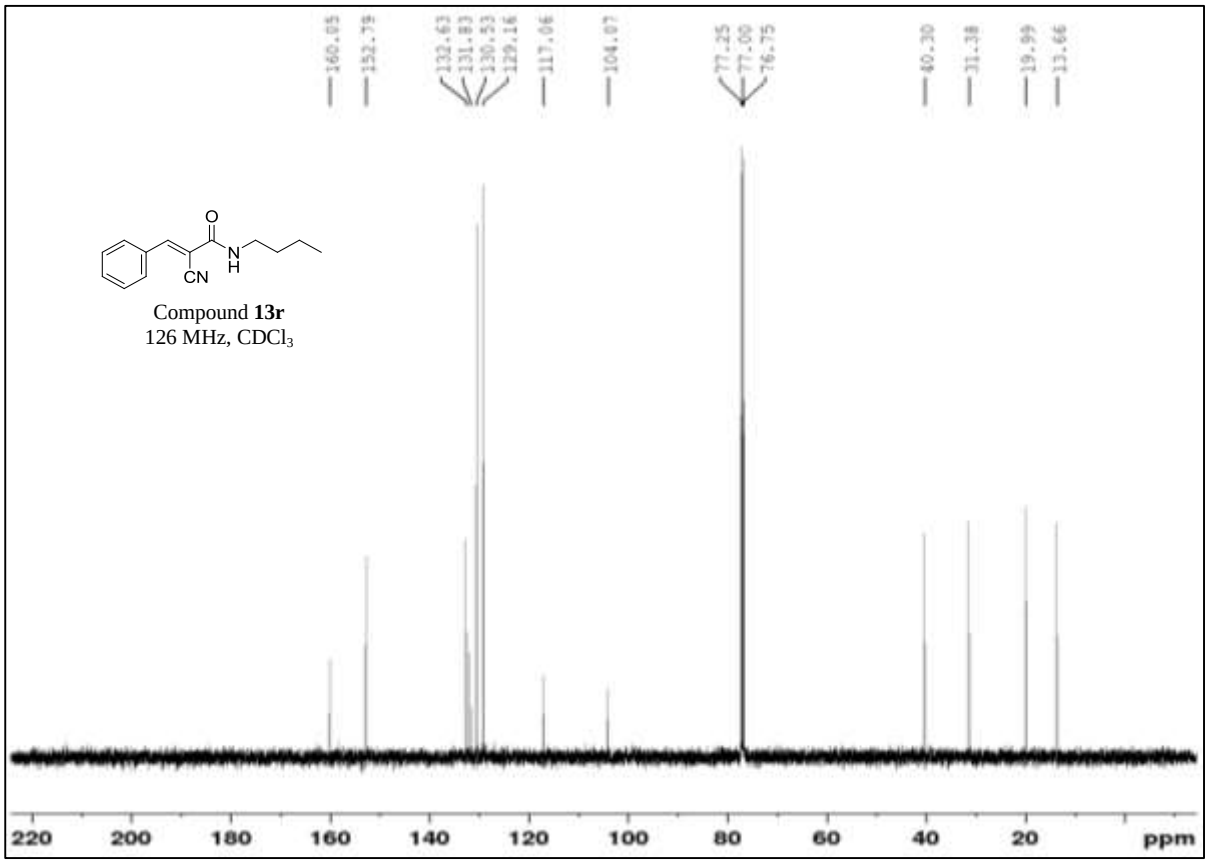
Compound **13q**
500 MHz, $CDCl_3$



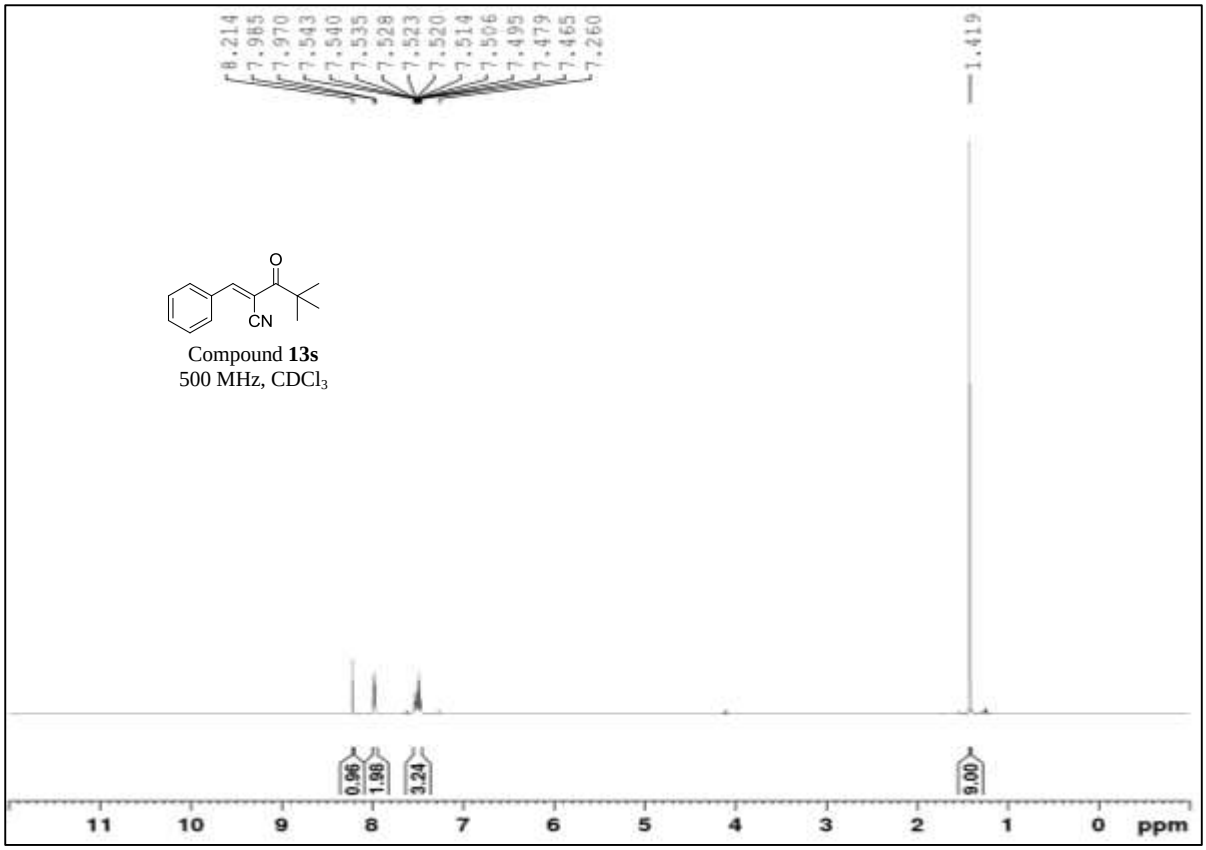
Compound **13q**
126 MHz, $CDCl_3$



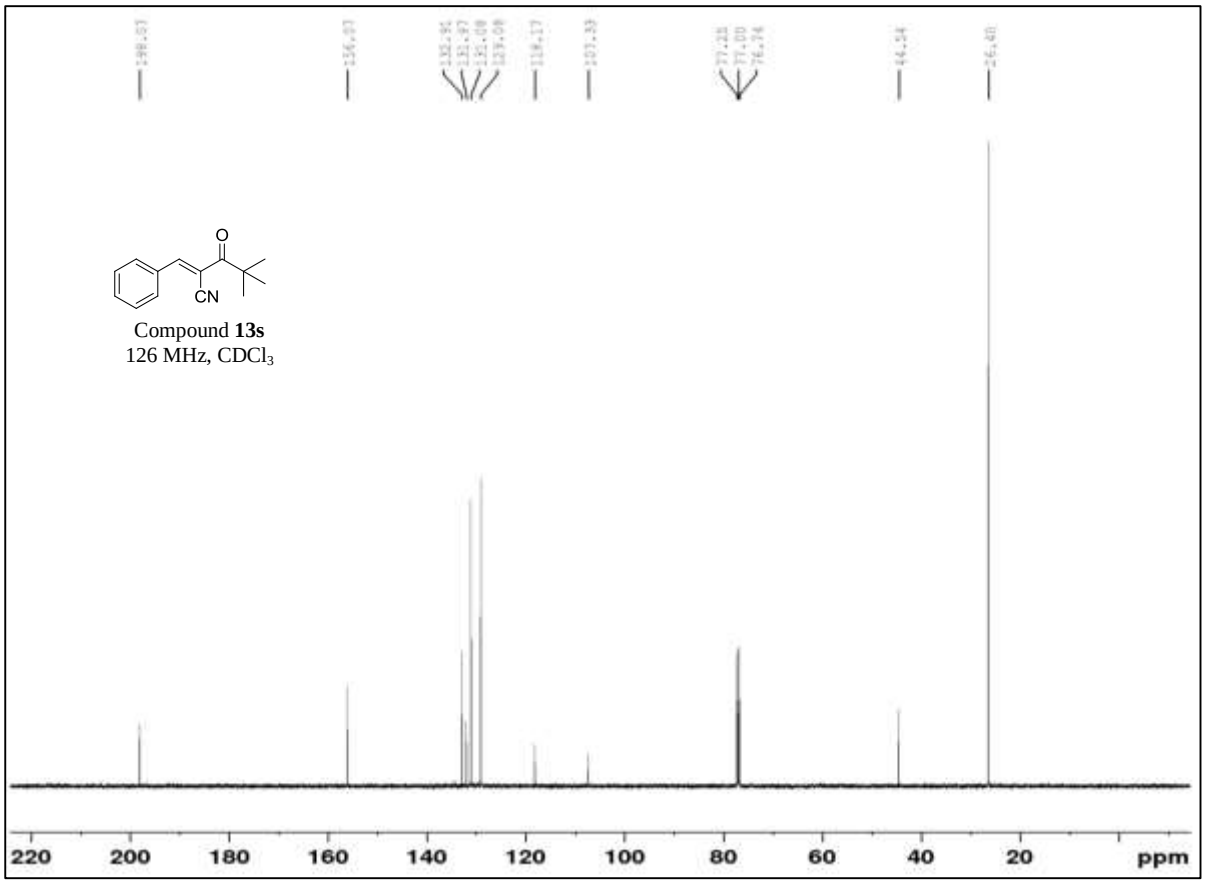
Compound **13r**
500 MHz, $CDCl_3$



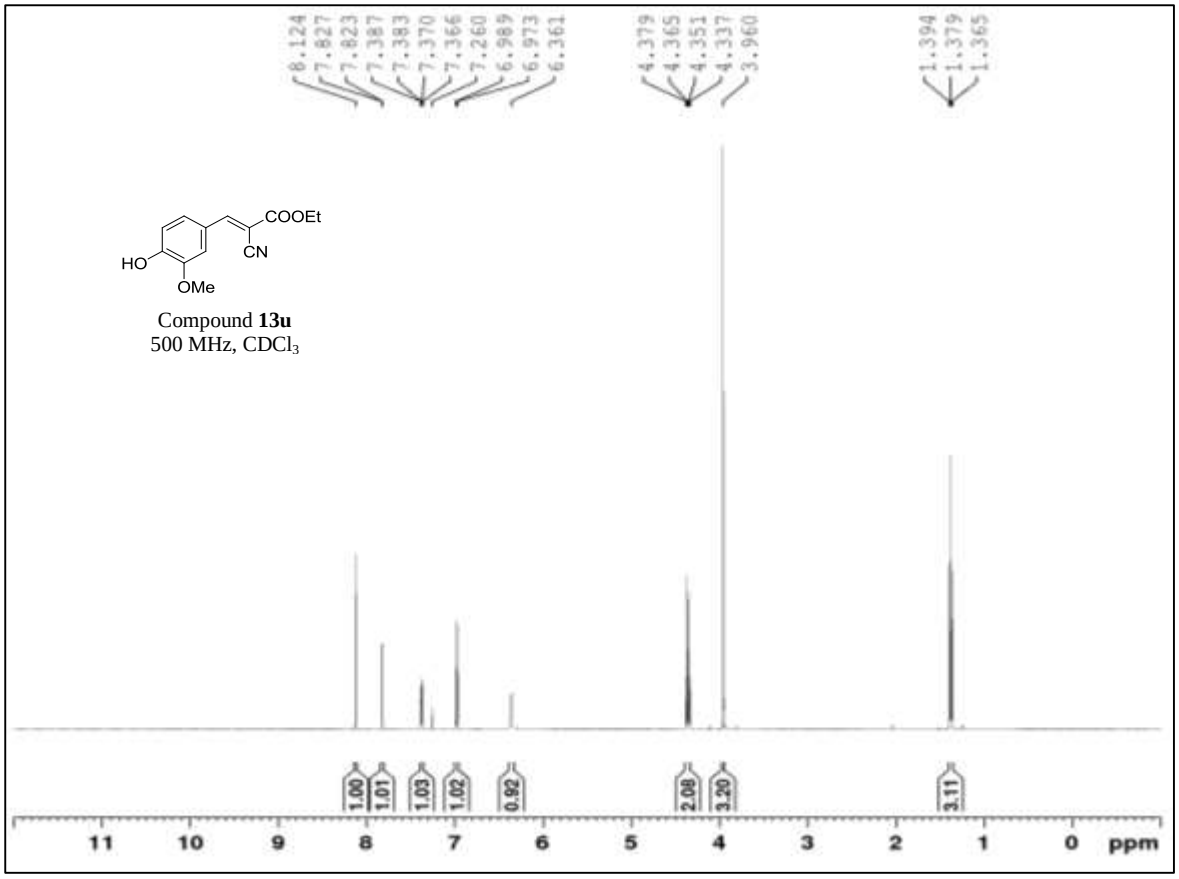
Compound **13r**
126 MHz, $CDCl_3$



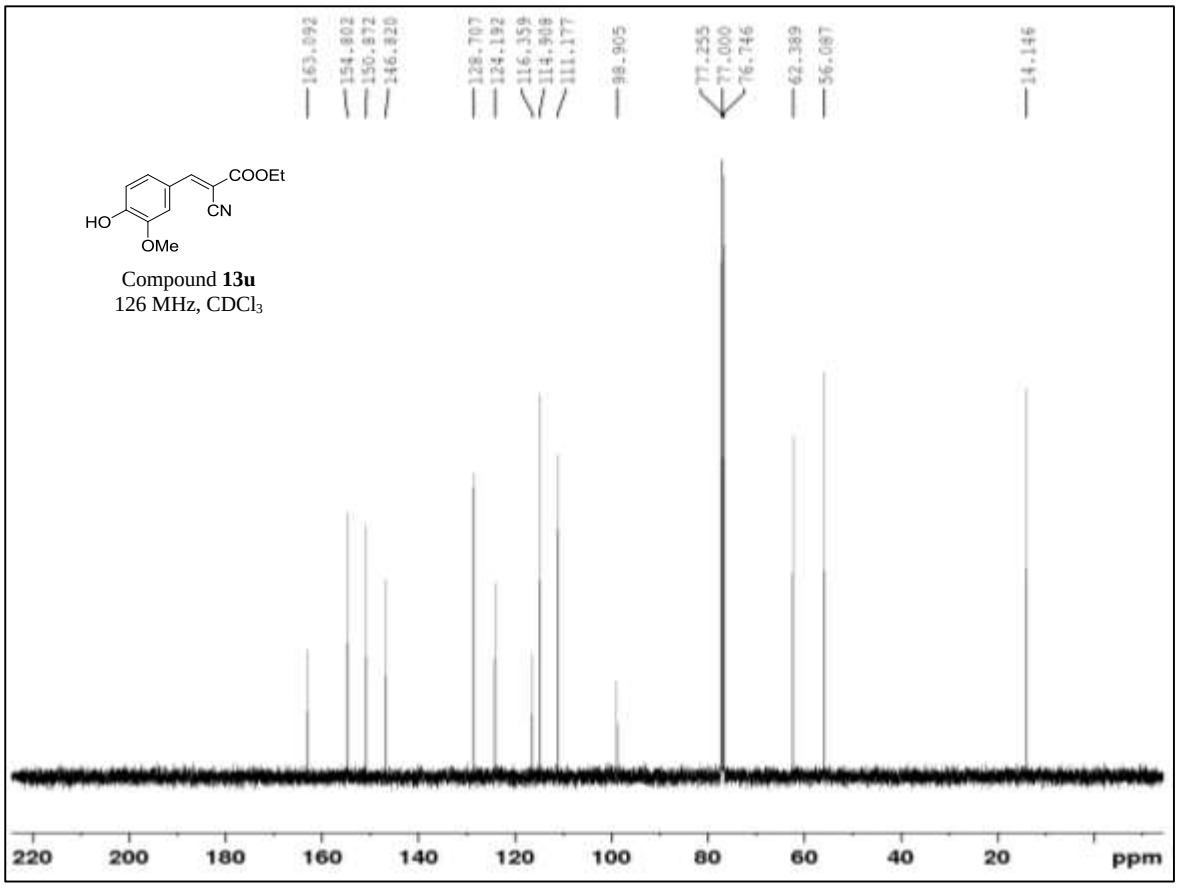
Compound **13s**
500 MHz, $CDCl_3$



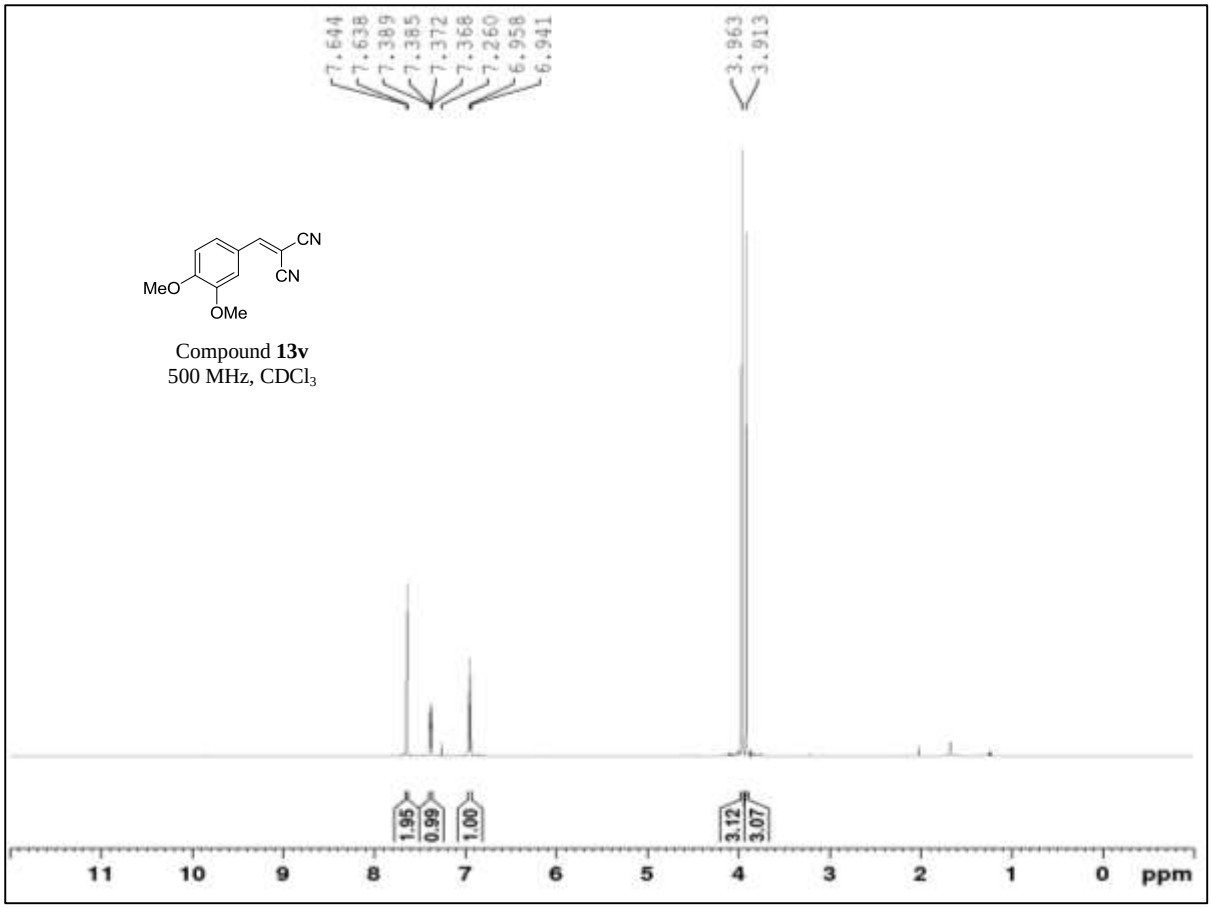
Compound **13s**
126 MHz, $CDCl_3$



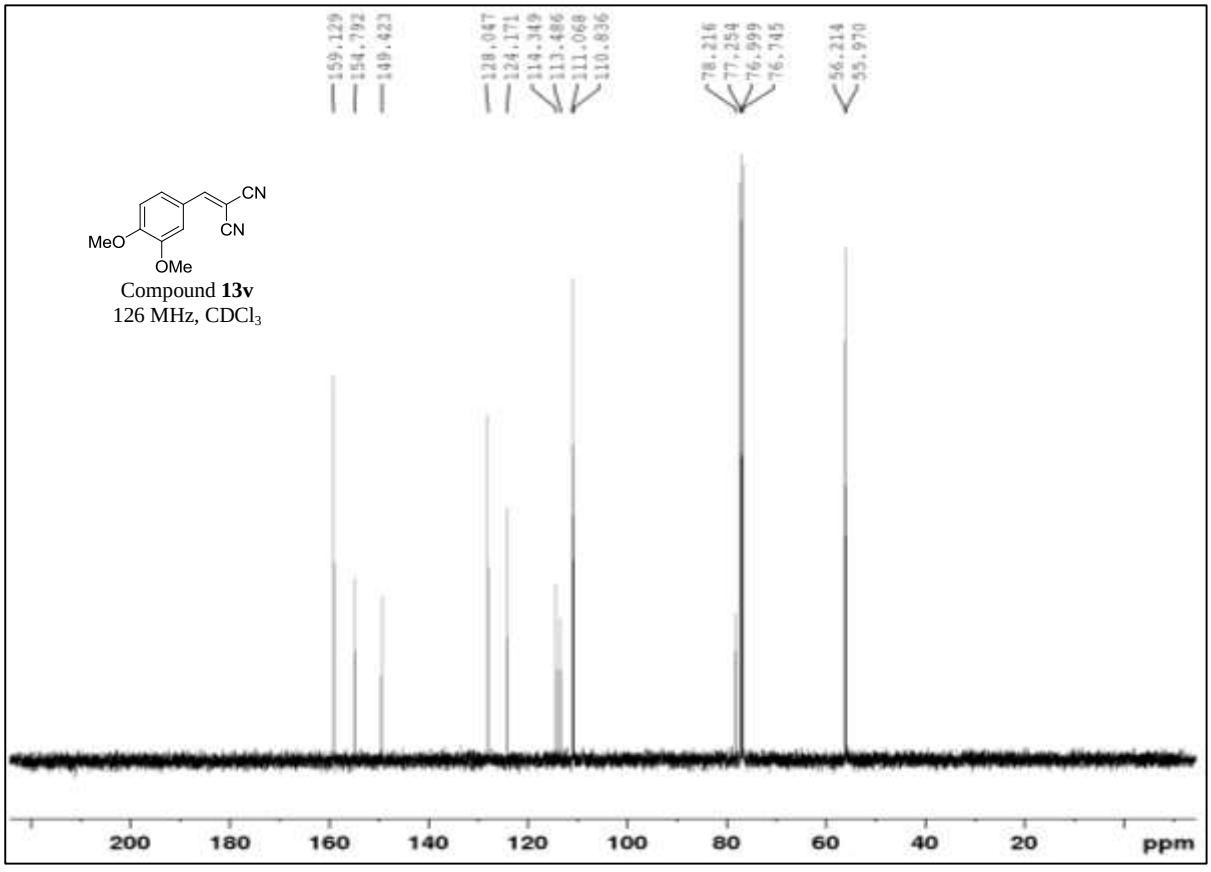
Compound **13u**
500 MHz, $CDCl_3$



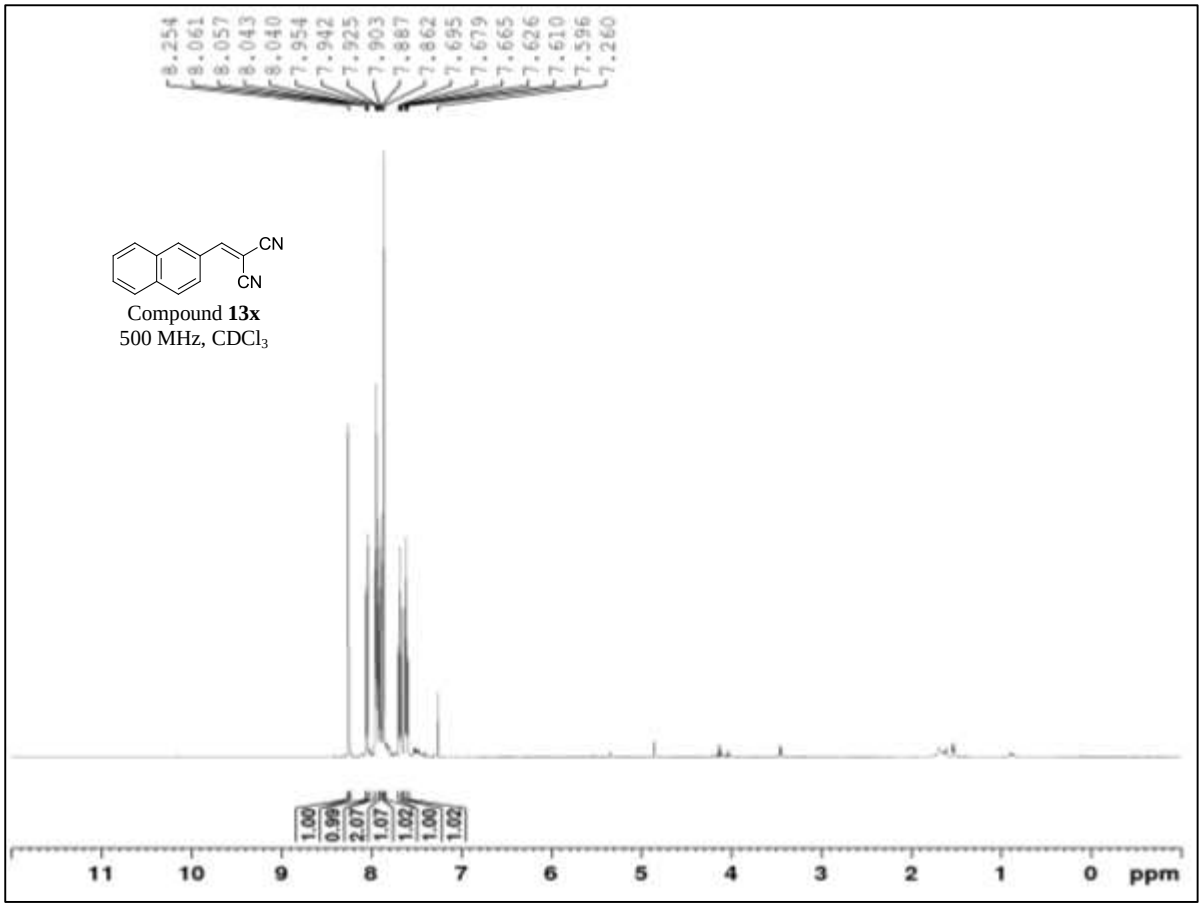
Compound **13u**
126 MHz, $CDCl_3$



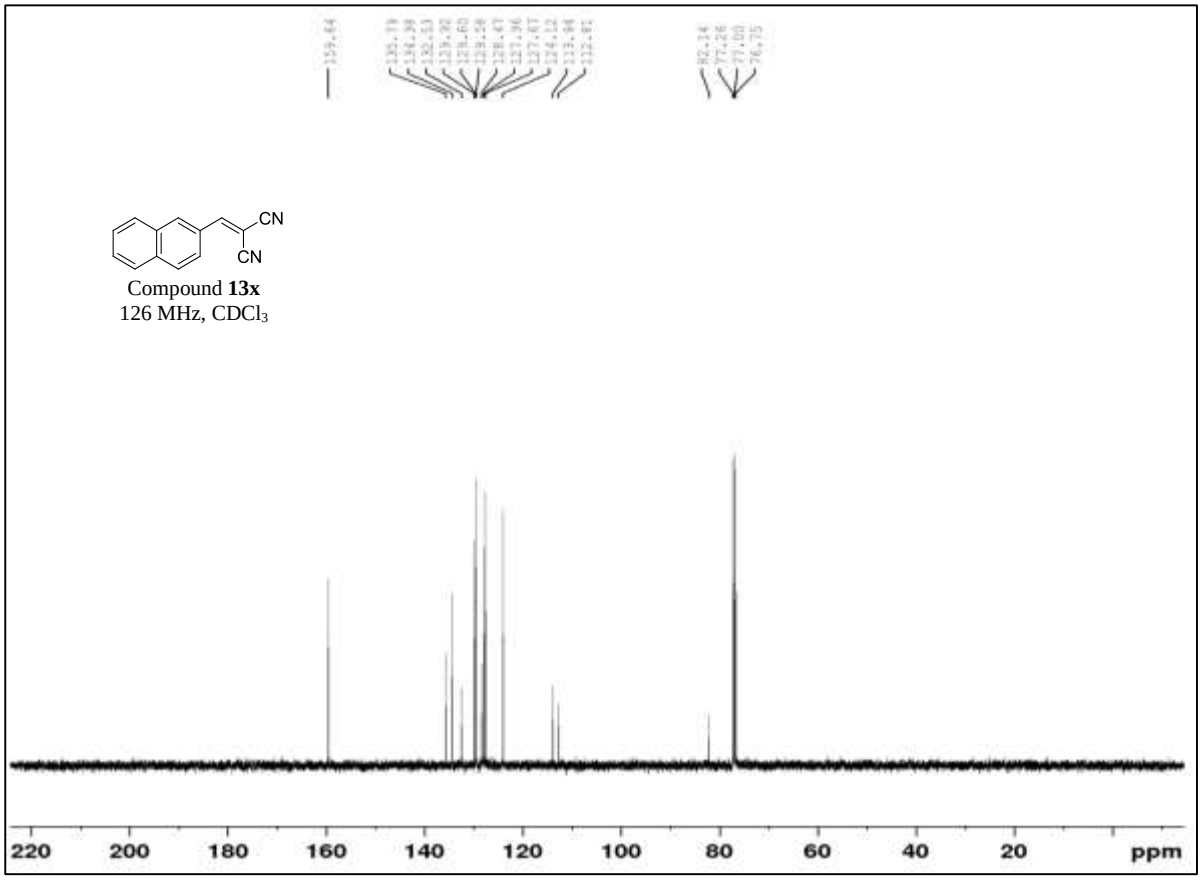
Compound **13v**
500 MHz, $CDCl_3$



Compound **13v**
126 MHz, $CDCl_3$



Compound **13x**
500 MHz, $CDCl_3$



Compound **13x**
126 MHz, $CDCl_3$