

*Supplementary Information*

# Ionic Organic Solid 1,3-bis(Sulfomethyl)Imidazolium As an Effective Metal-Free Catalyst for Sustainable Organic Syntheses

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## Detailed Experimental Protocols

### Procedure for the preparation of bsmim

Glyoxal (40% aq., 5 mmol, 0.57 mL), formaldehyde (37% aq., 5 mmol, 0.37 mL) and aminomethanesulfonic acid (10 mmol, 1.11 g) were added to a round-bottom flask along with water (2 mL) and sulfuric acid (catalytic, 100  $\mu$ L) and stirred at 95 °C for 20 minutes, after which it was quickly put in an ice-brine bath at -15 °C. Then, acetone (20 mL) was added and the mixture was vigorously stirred (1200 rpm), which caused the precipitation of a fluffy brown solid over the next 20 minutes. The solid was then filtered, affording 628 mg of pure **bsmim** (49% yield).

### General procedure for the Friedländer synthesis of quinolines promoted by bsmim

2-Aminobenzophenone (0.5 mmol), ketone (2.5 mmol) and **bsmim** (10 mol%, 13 mg) were added to a reaction tube. The reaction was then stirred at 80 °C for 16 hours, after which ethyl acetate was added (1 mL). After filtering the organic phase to remove the catalyst and removing the solvent under vacuum, the crude product was purified by column chromatography using mixtures of hexane and ethyl acetate.

### General procedure for the allylation of heterocycles with bsmim

In a glass tube, precisely weighed allyl alcohol (0.5 mmol, 105 mg), heterocycle (0.5 mmol) and **bsmim** (10 mol%, 13 mg) were added. The mixture was then stirred at 80 °C until completion (monitored by GC-MS), after which the crude reaction mixture was diluted with ethyl acetate (1 mL) and filtered through a thin plug of silica to remove the catalyst. After evaporation of the solvent under reduced pressure, the corresponding allyl heterocycles were obtained.

## Spectral data for all compounds

**1,3-Bis(sulfomethyl)imidazole (1):** Light brown solid, 49% yield; m.p. = 291-294 °C; <sup>1</sup>H NMR (400 MHz, D<sub>2</sub>O) δ<sub>H</sub> = 9.26 (s, 1H, NCHN), 7.76 (d, *J* = 1.6 Hz, 2H, NCHCHN), 5.43 (s, 4H, CH<sub>2</sub>); <sup>13</sup>C NMR (100 MHz, D<sub>2</sub>O) δ<sub>C</sub> = 138.3, 123.5, 62.6. MS (EI, 70 eV) *m/z* (%): 66 (5), 64 (100), 48 (40); MS/MS (ESI<sup>+</sup>) [256]: 261 (11), 242 (26), 238 (20), 193 (13), 176 (100), 163 (27).

**3-Acetyl-2-methyl-4-phenylquinoline (3):** Yellow solid, purification by column chromatography (hexane/ethyl acetate 7:3), 99% yield; m.p. 120-122 °C (lit. 117-119 °C)<sup>1</sup>; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.13 (d, *J* = 8.4 Hz, 1H, CH<sub>Ar</sub>), 7.74 (ddd, *J* = 8.4, 6.9, 1.4 Hz, 1H, CH<sub>Ar</sub>), 7.65-7.62 (m, 1H, CH<sub>Ar</sub>), 7.53-7.46 (m, 4H, CH<sub>Ar</sub>), 7.38-7.35 (m, 2H, CH<sub>Ar</sub>), 2.73 (s, 3H, NCCH<sub>3</sub>), 2.00 (s, 3H, COCH<sub>3</sub>); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 205.5, 153.7, 147.1, 144.7, 135.2, 134.9, 130.6, 130.1, 129.2, 128.9, 128.6, 126.9, 126.4, 125.2, 32.1, 23.7; MS (EI, 70 eV) *m/z* (%): 261 (M<sup>+</sup>, 49), 247 (19), 246 (100), 219 (7), 218 (38), 217 (31), 176 (22), 151 (8).

**3-(Ethoxycarbonyl)-2-methyl-4-phenylquinoline (4):** Yellow solid, purification by column chromatography (hexane/ethyl acetate 9:1), 98% yield; m.p. 99-100 °C (lit. 98-100 °C)<sup>1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.06 (d, *J* = 8.1 Hz, 1H, CH<sub>Ar</sub>), 7.68 (ddd, *J* = 8.4, 6.8, 1.4 Hz, 1H, CH<sub>Ar</sub>), 7.56 (dd, *J* = 8.4, 0.9 Hz, 1H, CH<sub>Ar</sub>), 7.47-7.43 (m, 3H, CH<sub>Ar</sub>), 7.41-7.33 (m, 3H, CH<sub>Ar</sub>), (q, *J* = 7.1 Hz, 2H, CH<sub>2</sub>), 2.78 (s, 3H, NCCH<sub>3</sub>), 0.92 (t, *J* = 7.1 Hz, 3H, COCCH<sub>3</sub>); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 168.5, 154.6, 147.7, 146.4, 135.8, 130.3, 129.4, 128.9, 128.5, 128.3, 127.5, 126.5, 126.5, 125.2, 61.4, 23.8, 13.7; MS (EI, 70 eV) *m/z* (%): 292 (M<sup>+1</sup>, 20), 291 (M<sup>+</sup>, 96), 263 (8), 262 (8), 247 (20), 246 (100), 245 (34), 219 (10), 218 (44), 217 (41), 216 (14), 177 (8), 176 (23), 151 (8).

**3-acetyl-6-chloro-2-methyl-4-phenylquinoline (5):** Yellow solid, purification by column chromatography (hexane/ethyl acetate 85:15), 93% yield; m.p. 160-161 °C (lit. 149-152 °C)<sup>1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.06 (d, *J* = 8.9 Hz, 1H, CH<sub>Ar</sub>), 7.66 (dd, *J* = 8.9, 2.3 Hz, 1H, CH<sub>Ar</sub>), 7.58 (d, *J* = 2.3 Hz, 1H, CH<sub>Ar</sub>), 7.55-7.52 (m, 3H, CH<sub>Ar</sub>), 7.35-7.32 (m, 2H, *J* = 8.9, 2.3 Hz, 1H, CH<sub>Ar</sub>), 2.69 (s, 3H, NCCH<sub>3</sub>), 1.99 (s, 3H, COCH<sub>3</sub>); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 204.9, 154.1, 145.3, 143.9, 135.7, 134.4, 132.9, 131.5, 130.0, 129.5, 129.1, 126.1, 125.1, 31.9, 23.6; MS (EI, 70 eV) *m/z* (%): 297 (M<sup>+2</sup>, 16), 296 (M<sup>+1</sup>, 10), 295 (M<sup>+</sup>, 46), 282 (33), 281 (19), 280 (100), 254 (7), 252 (22), 218 (8), 217 (32), 216 (13), 189 (8), 176 (23).

**9-Phenyl-1,2,3,4-tetrahydroacridine (6):** Yellow solid, purification by column chromatography (hexane/ethyl acetate 8:2), 94% yield; m.p. 151-153 °C (lit. 152-155 °C)<sup>2</sup>; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.13 (d, *J* = 8.5 Hz, 1H, CH<sub>Ar</sub>), 7.66-7.60 (m, 1H, CH<sub>Ar</sub>), 7.56-7.45 (m, 3H, CH<sub>Ar</sub>), 7.35-7.33 (m, 2H, CH<sub>Ar</sub>), 7.25-7.22 (m, 2H, CH<sub>Ar</sub>), 3.27 (t, *J* = 6.6 Hz, 2H, NCCH<sub>2</sub>), 2.62 (t, *J* = 6.4 Hz, 2H, PhCCCH<sub>2</sub>), 2.02-1.93 (m, 2H, NCCCH<sub>2</sub>), 1.84-1.76 (m, 2H, PhCCCCH<sub>2</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 158.9, 147.7, 145.4, 136.9, 129.1, 129.0, 128.8, 128.7, 128.1, 127.7, 126.9, 126.0, 125.9, 33.8, 28.1, 23.0, 22.8; MS (EI, 70 eV) *m/z* (%): 260 (M<sup>+1</sup>, 19), 259 (M<sup>+</sup>, 100), 258 (M<sup>-1</sup>, 66), 244 (8), 230 (14), 182 (8).



**9-Phenyl-2,3-dihydro-1*H*-cyclopenta[*b*]quinoline (7):** Faint yellow solid, purification by column chromatography (hexane/ethyl acetate 98:2), 93% yield, m.p. 138–141 °C (lit. 138–139 °C)<sup>2</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.02–8.00 (m, 1H, CH<sub>Ar</sub>), 7.57–7.53 (m, 2H, CH<sub>Ar</sub>), 7.47–7.37 (m, 3H, CH<sub>Ar</sub>), 7.32–7.28 (m, 3H, CH<sub>Ar</sub>), 3.17 (t, J = 7.7 Hz, 2H, NCCH<sub>2</sub>), 2.83 (t, J = 7.4 Hz, 2H, PhCCCH<sub>2</sub>), 2.13–2.07 (m, 2H, NCCCH<sub>2</sub>); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 167.5, 147.9, 142.9, 136.9, 133.8, 129.4, 128.9, 128.6, 128.4, 128.1, 126.4, 125.8, 125.7, 35.3, 30.5, 23.7; MS (EI, 70 eV) *m/z* (%): 246 (M<sup>+</sup>+1, 18), 245 (M<sup>+</sup>, 100), 244 (M<sup>+</sup>-1, 80), 243 (12), 242 (12), 241 (7), 217 (9), 168 (15), 167 (10), 108 (7).

**12-Phenyl-6,7,8,9,10,11-hexahydrocycloocta[*b*]quinoline (8):** Faint yellow solid, purification by column chromatography (hexane/ethyl acetate 95:5), 96% yield; m.p. 209–211 °C (lit. 214–215 °C)<sup>1</sup>; <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.01–7.98 (m, 1H, CH<sub>Ar</sub>), 7.54–7.48 (m, 1H, CH<sub>Ar</sub>), 7.46–7.36 (m, 3H, CH<sub>Ar</sub>), 7.27–7.11 (m, 4H, CH<sub>Ar</sub>), 3.17–3.13 (m, 2H, NCCH<sub>2</sub>), 2.69–2.65 (m, 2H, PhCCCH<sub>2</sub>), 1.88–1.84 (m, 2H, CH<sub>2</sub>), 1.42–1.27 (m, 6H, 3xCH<sub>2</sub>); <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 163.5, 146.7, 146.3, 137.7, 131.9, 129.4, 128.5, 128.4, 127.7, 127.3, 126.2, 125.5, 36.3, 31.3, 31.3, 28.2, 26.8, 25.9; MS (EI, 70 eV) *m/z* (%): 288 (M<sup>+</sup>+1, 20), 287 (M<sup>+</sup>, 93), 286 (M<sup>+</sup>-1, 100), 272 (9), 260 (8), 259 (17), 258 (40), 256 (9), 245 (8), 244 (25), 243 (9), 242 (9), 232 (19), 231 (14), 230 (16), 217 (14), 216 (7), 202 (8), 189 (10).

**9-Phenyl-3,4-dihydroacridin-1(2*H*)-one (9):** Yellow solid, purification by column chromatography (hexane/ethyl acetate 8:2), 96% yield; m.p. 163–165 °C (lit. 161–162 °C)<sup>2</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 8.18 (d, J = 8.4 Hz, 1H, CH<sub>Ar</sub>), 7.80 (ddd, J = 8.4, 6.6, 1.6 Hz, 1H, CH<sub>Ar</sub>), 7.53–7.41 (m, 5H, CH<sub>Ar</sub>), 7.19–7.17 (m, 2H, CH<sub>Ar</sub>), 3.45 (t, J = 6.3 Hz, 2H, NCCH<sub>2</sub>), 2.72 (t, J = 6.1 Hz, 2H, COCH<sub>2</sub>), 2.28–2.25 (m, 2H, COCCH<sub>2</sub>); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 197.6, 162.1, 152.5, 147.7, 137.4, 132.4, 128.4, 128.3, 128.1, 127.9, 127.7, 126.9, 124.0, 40.7, 34.1, 21.4; MS (EI, 70 eV) *m/z* (%): 274 (M<sup>+</sup>+1, 19), 273 (M<sup>+</sup>, 100), 272 (M<sup>+</sup>-1, 94), 246 (7), 245 (47), 244 (97), 217 (32), 216 (31), 214 (8), 190 (9), 189 (16), 176 (7).

**2,9-Diphenyl-1,2,3,4-tetrahydroacridine (10):** Yellow solid, purification by column chromatography (hexane/ethyl acetate 9:1), 97% yield; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 7.98–7.96 (m, 1H, CH<sub>Ar</sub>), 7.57–7.52 (m, 1H, CH<sub>Ar</sub>), 7.41–7.35 (m, 3H, CH<sub>Ar</sub>), 7.25–7.10 (m, 9H, CH<sub>Ar</sub>), 3.37–3.22 (m, 2H, CH<sub>Aik</sub>), 2.95 (tdd, J = 11.4, 4.7, 3.0 Hz, 1H, CH<sub>Aik</sub>), 2.83 (ddd, J = 17.1, 4.9, 2.0 Hz, 1H, CH<sub>Aik</sub>), 2.68 (dd, J = 17.1, 4.9 Hz, 1H, CH<sub>Aik</sub>), 2.25–2.19 (m, 1H, CH<sub>Aik</sub>), 2.13–2.02 (m, 1H, CH<sub>Aik</sub>); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 158.4, 146.9, 146.6, 145.9, 136.9, 129.2, 129.0, 128.9, 128.8, 128.7, 128.5, 127.9, 127.8, 127.0, 126.8, 126.5, 126.0, 125.7, 40.9, 36.2, 34.4, 30.3; MS (EI, 70 eV) *m/z* (%): 336 (M<sup>+</sup>+1, 14), 335 (M<sup>+</sup>, 65), 334 (M<sup>+</sup>-1, 100), 231 (8), 230 (19), 91 (8).

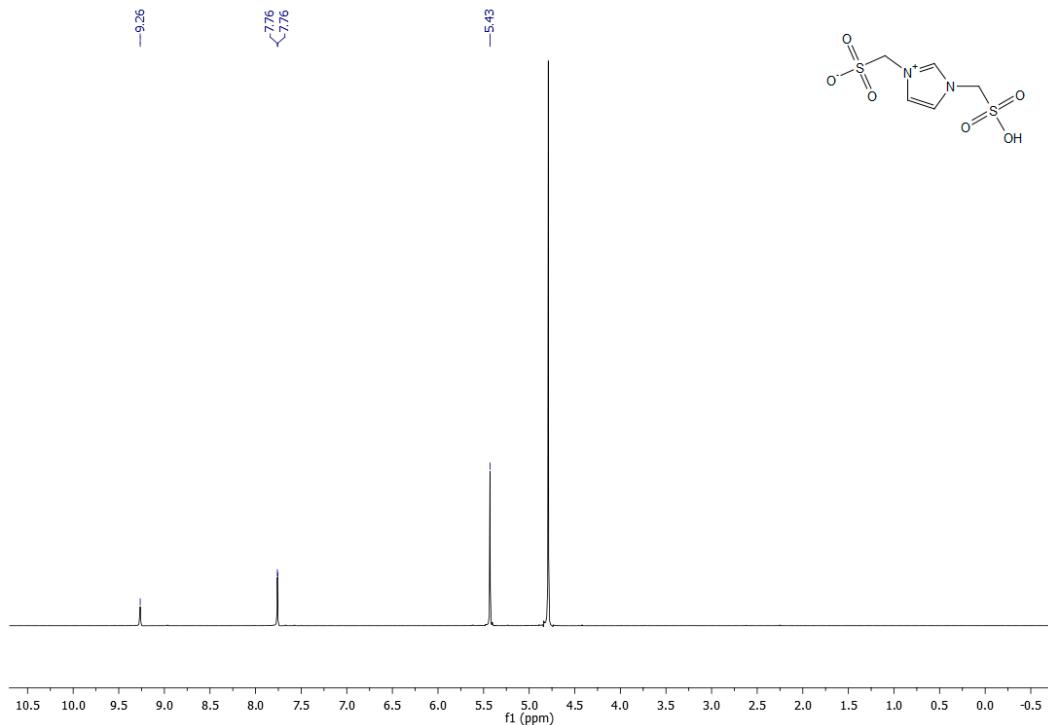
**(E)-3-(1,3-Diphenylallyl)-1*H*-indole (11):** Yellow oil, obtained pure, 99% yield; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ<sub>H</sub> = 7.96 (br s, 1H, NH), 7.51 (d, J = 8.0 Hz, 1H, CH<sub>Ar</sub>), 7.45–7.22 (m, 12H, CH<sub>Ar</sub>), 7.10 (ddd, J = 8.0, 7.1, 1.0 Hz, 1H, CH<sub>Ar</sub>), 6.93 (d, J = 1.3 Hz, 1H, CH<sub>Ar</sub>), 6.80 (dd, J = 15.9, 7.4 Hz, 1H, PhC=CH), 6.52 (br d, J = 15.9 Hz, 1H, C=CHPh), 5.19 (br d, J = 7.4 Hz, 1H, C=CCH); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ<sub>C</sub> = 143.5, 137.6, 136.8, 132.7, 130.7, 128.6, 128.6, 127.3, 126.9, 126.5, 126.5, 122.7, 122.2, 120.0, 119.6, 118.8, 111.2, 46.3; MS (EI, 70 eV) *m/z* (%): 310 (M<sup>+</sup>+1, 25), 309 (M<sup>+</sup>, 100), 308 (M<sup>+</sup>-1, 39), 294 (10), 233 (7), 232 (36), 231 (7), 230 (20), 218 (15), 217 (17), 206 (28), 205 (8), 204 (22), 202 (8), 192 (14), 191 (16), 178 (9), 130 (19), 115 (17).

**(E)-3-(1,3-Diphenylallyl)-2-phenyl-1*H*-indole (12):** Faint orange oil, purified by preparative TLC (hexane/ethyl acetate 9:1), 60% yield;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{H}} = 8.02$  (br s, 1H, NH), 7.52-7.48 (m, 2H,  $\text{CH}_{\text{Ar}}$ ), 7.44-7.12 (m, 16H,  $\text{CH}_{\text{Ar}}$ ), 6.98 (ddd,  $J = 8.0, 7.1, 1.0$  Hz, 1H,  $\text{CH}_{\text{Ar}}$ ), 6.88 (dd,  $J = 15.8, 7.3$  Hz, 1H,  $\text{PhC=CH}$ ), 6.39 (dd,  $J = 15.8, 1.0$  Hz, 1H,  $\text{C=CHPh}$ ), 5.27 (br d,  $J = 7.3$  Hz, 1H,  $\text{C=CCH}$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{C}} = 143.6, 137.6, 136.3, 135.7, 133.0, 132.4, 131.2, 128.9, 128.7, 128.6, 128.4, 128.2, 128.0, 127.2, 126.4, 126.2, 122.2, 121.3, 119.8, 113.9, 111.1, 45.2$ ; MS (EI, 70 eV)  $m/z$  (%): 386 ( $\text{M}^{+}+1$ , 30), 385 ( $\text{M}^{+}$ , 100), 384 ( $\text{M}^{+}-1$ , 19), 309 (12), 308 (46), 307 (7), 306 (17), 304 (11), 295 (21), 294 (96), 293 (29), 292 (9), 291 (11), 282 (17), 281 (8), 280 (18), 278 (8), 230 (14), 218 (9), 217 (13), 207 (9), 206 (47), 205 (8), 204 (23), 203 (8), 202 (9), 194 (9), 193 (45), 192 (22), 191 (22), 189 (8), 178 (9), 176 (7), 165 (14), 153 (9), 152 (10), 146 (9), 115 (9), 91 (7).

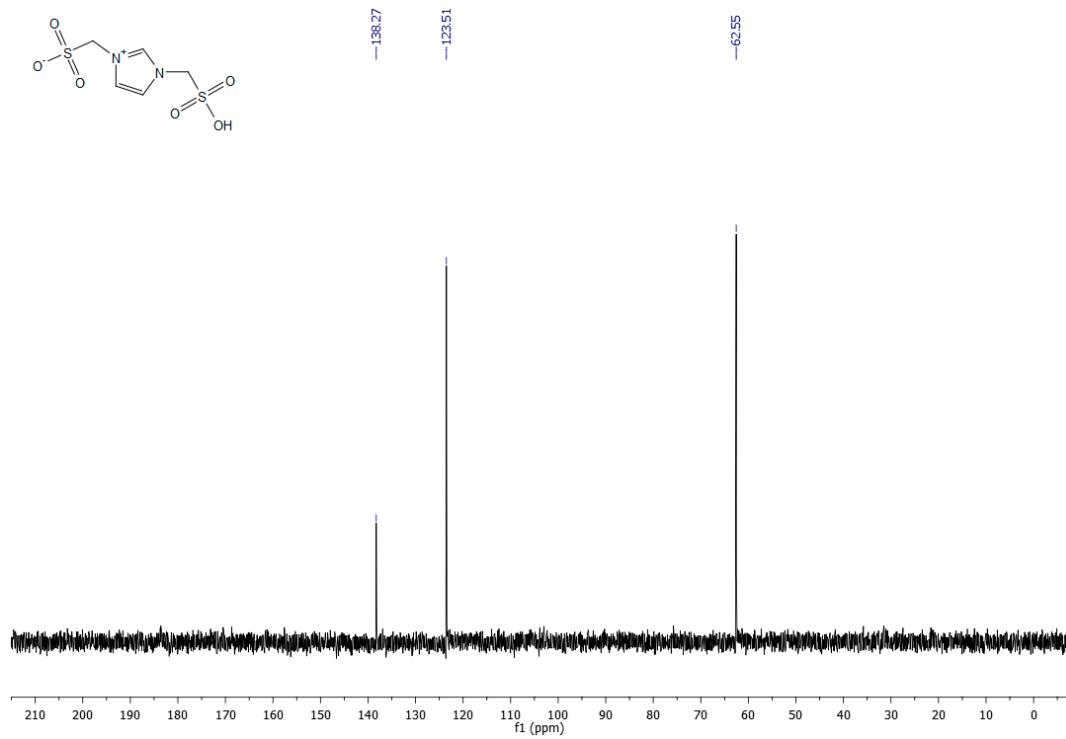
**(E)-3-(1,3-Diphenylallyl)-9-ethyl-1*H*-indole (13):** Reddish oil, obtained pure, 99% yield;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ )  $\delta_{\text{H}} = 7.89$  (br s, 1H, NH), 7.36-7.15 (m, 11H,  $\text{CH}_{\text{Ar}}$ ), 7.02-6.97 (m, 2H, CHN,  $\text{CH}_{\text{Ar}}$ ), 6.84 (dd,  $J = 2.4, 0.9$  Hz, 1H,  $\text{CH}_{\text{Ar}}$ ), 6.72 (dd,  $J = 15.8, 7.4$  Hz, 1H,  $\text{PhC=CH}$ ), 6.43 (br d,  $J = 15.8$  Hz, 1H,  $\text{C=CHPh}$ ), 5.10 (br d,  $J = 7.4$  Hz, 1H,  $\text{C=CCH}$ ), 2.82 (q,  $J = 7.6$  Hz, 2H,  $\text{CH}_2$ ), 1.34 (t,  $J = 7.6$  Hz, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{C}} = 143.6, 137.6, 135.6, 132.7, 130.6, 128.6, 128.5, 127.3, 126.7, 126.6, 126.4, 122.3, 120.7, 119.8, 119.2, 117.8, 46.4, 24.1, 13.9$ ; MS (EI, 70 eV)  $m/z$  (%): 338 ( $\text{M}^{+}+1$ , 27), 337 ( $\text{M}^{+}$ , 100), 336 ( $\text{M}^{+}-1$ , 34), 334 (9), 333 (28), 332 (9), 308 (20), 261 (7), 260 (33), 234 (23), 231 (9), 230 (24), 218 (9), 217 (14), 204 (10), 192 (17), 191 (20), 189 (7), 158 (15), 130 (12), 115 (11); HRMS (QTOF) calculated for  $\text{C}_{25}\text{H}_{23}\text{N} = 337.1830$ , observed = 337.1823.

**(E)-1-(1,3-Diphenylallyl)-1*H*-pyrazole (14):** Colorless oil, obtained pure, 99% yield;  $^1\text{H}$  NMR (300 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{H}} = 7.60$  (d,  $J = 1.4$  Hz, 1H,  $\text{CH}_{\text{Ar}}$ ), 7.45 (d,  $J = 2.2$  Hz, 1H,  $\text{CH}_{\text{Ar}}$ ), 7.39-7.20 (m, 10H,  $\text{CH}_{\text{Ar}}$ ), 6.71 (dd,  $J = 16.0, 6.9$  Hz, 1H,  $\text{PhC=CH}$ ), 6.43 (br d,  $J = 16.0$  Hz, 1H,  $\text{C=CHPh}$ ), 6.30-6.28 (m, 1H,  $\text{CH}_{\text{Ar}}$ ), 6.18 (br d,  $J = 6.9$  Hz, 1H,  $\text{C=CCH}$ );  $^{13}\text{C}$  NMR (75 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{C}} = 139.6, 139.4, 136.1, 133.8, 128.9, 128.7, 128.2, 127.4, 127.3, 126.8, 105.7, 67.6$ ; MS (EI, 70 eV)  $m/z$  (%): 261 ( $\text{M}^{+}+1$ , 15), 260 ( $\text{M}^{+}$ , 78), 259 ( $\text{M}^{+}-1$ , 24), 232 (7), 193 (28), 192 (43), 191 (55), 190 (11), 189 (23), 184 (8), 183 (55), 179 (7), 178 (33), 169 (31), 165 (21), 157 (18), 156 (36), 144 (21), 143 (8), 117 (9), 115 (100), 91 (27), 89 (7), 77 (8).

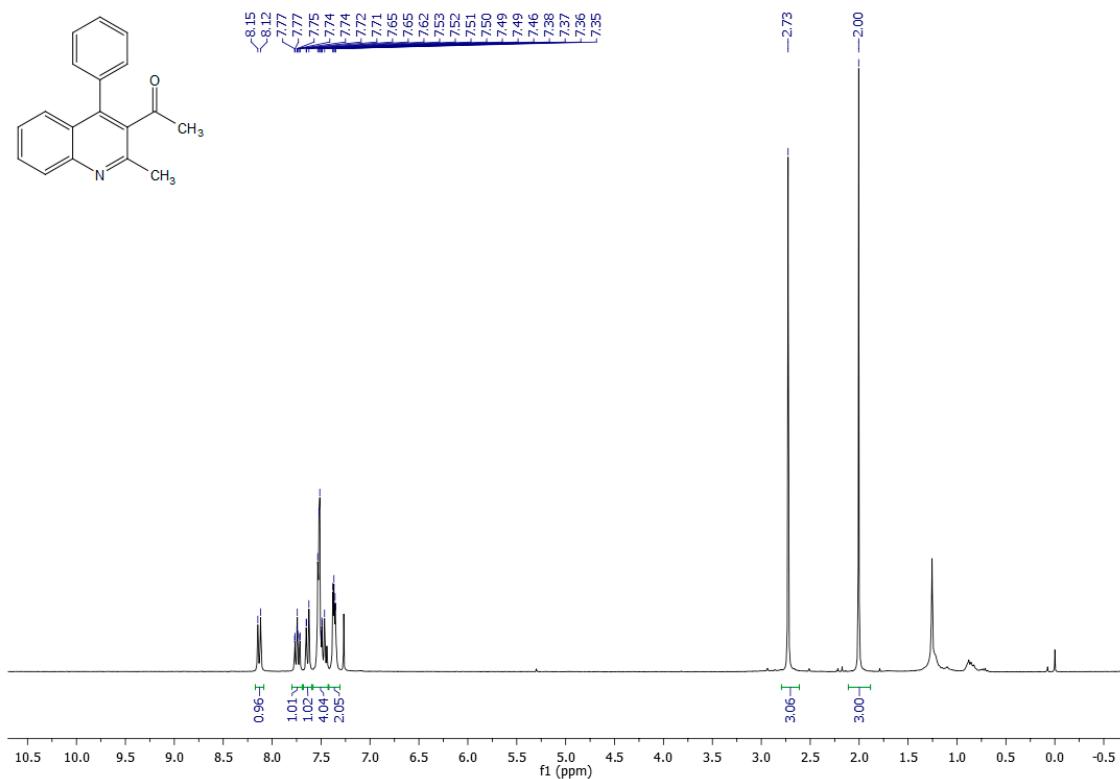
**(E)-1-(1,3-Diphenylallyl)-3,5-dimethyl-1*H*-pyrazole (15):** White solid, obtained pure, 99% yield; m.p. 108-110 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{H}} = 7.43$ -7.41 (m, 2H,  $\text{CH}_{\text{Ar}}$ ), 7.33-7.23 (m, 6H,  $\text{CH}_{\text{Ar}}$ ), 7.16-7.14 (m, 2H,  $\text{CH}_{\text{Ar}}$ ), 6.88 (dd,  $J = 15.9, 7.5$  Hz, 1H,  $\text{PhC=CH}$ ), 6.51 (br d,  $J = 15.9$  Hz, 1H,  $\text{C=CHPh}$ ), 5.98 (br d,  $J = 7.5$  Hz, 1H,  $\text{C=CCH}$ ), 5.87 (s, 1H, MeCCH), 2.27 (s, 3H,  $\text{CH}_3$ ), 2.17 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta_{\text{C}} = 147.8, 140.2, 139.2, 136.5, 133.1, 128.8, 128.6, 128.0, 127.9, 127.7, 126.9, 126.9, 105.9, 64.5, 13.8, 11.5$ ; MS (EI, 70 eV)  $m/z$  (%): 289 ( $\text{M}^{+}+1$ , 10), 288 ( $\text{M}^{+}$ , 50), 287 ( $\text{M}^{+}-1$ , 18), 273 (8), 212 (16), 211 (100), 194 (7), 193 (38), 192 (14), 185 (9), 184 (9), 178 (30), 170 (12), 165 (3), 116 (10), 115 (91), 108 (8), 91 (25).

**NMR spectra**


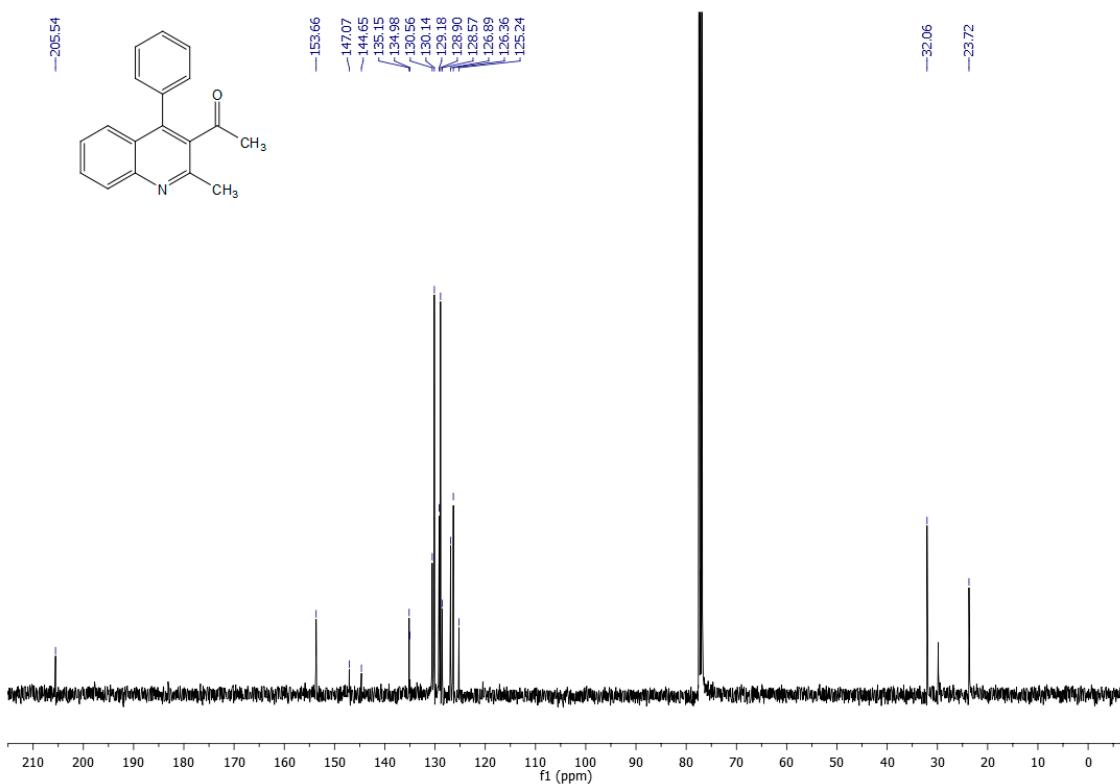
**Figure SI.1.**  $^1\text{H}$  NMR spectrum of 1,3-bis(sulfomethyl)imidazole (1)



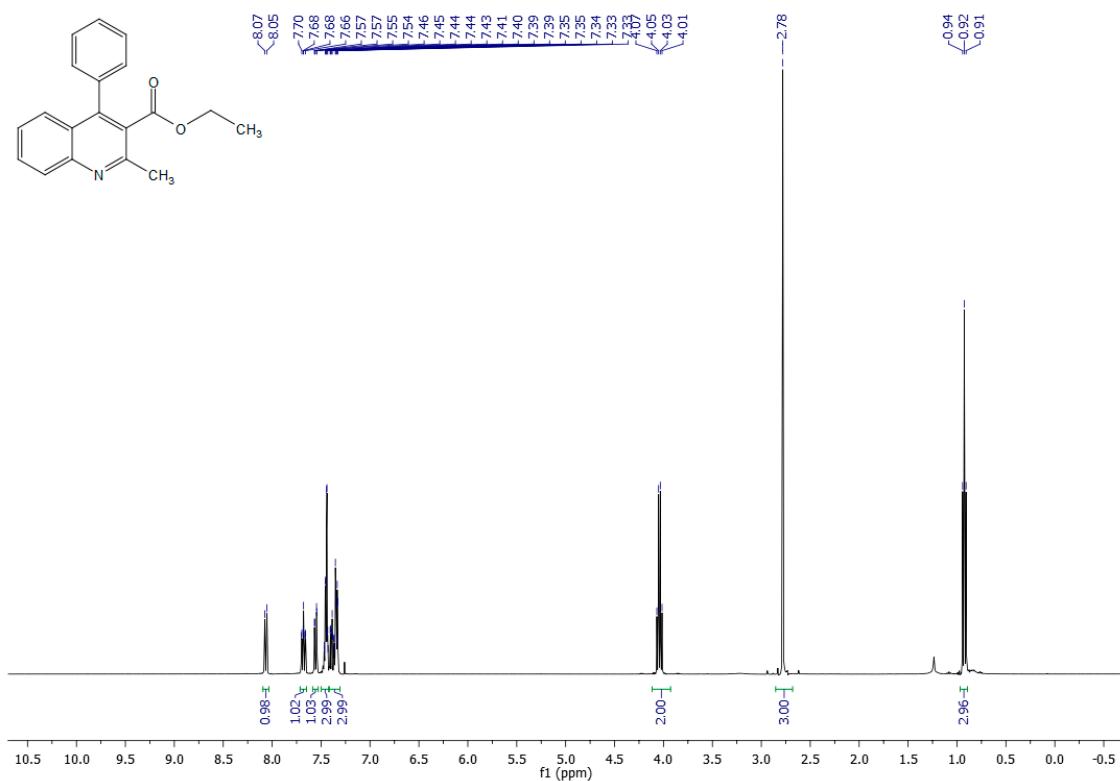
**Figure SI.2.**  $^{13}\text{C}$  NMR spectrum of 1,3-bis(sulfomethyl)imidazole (1)



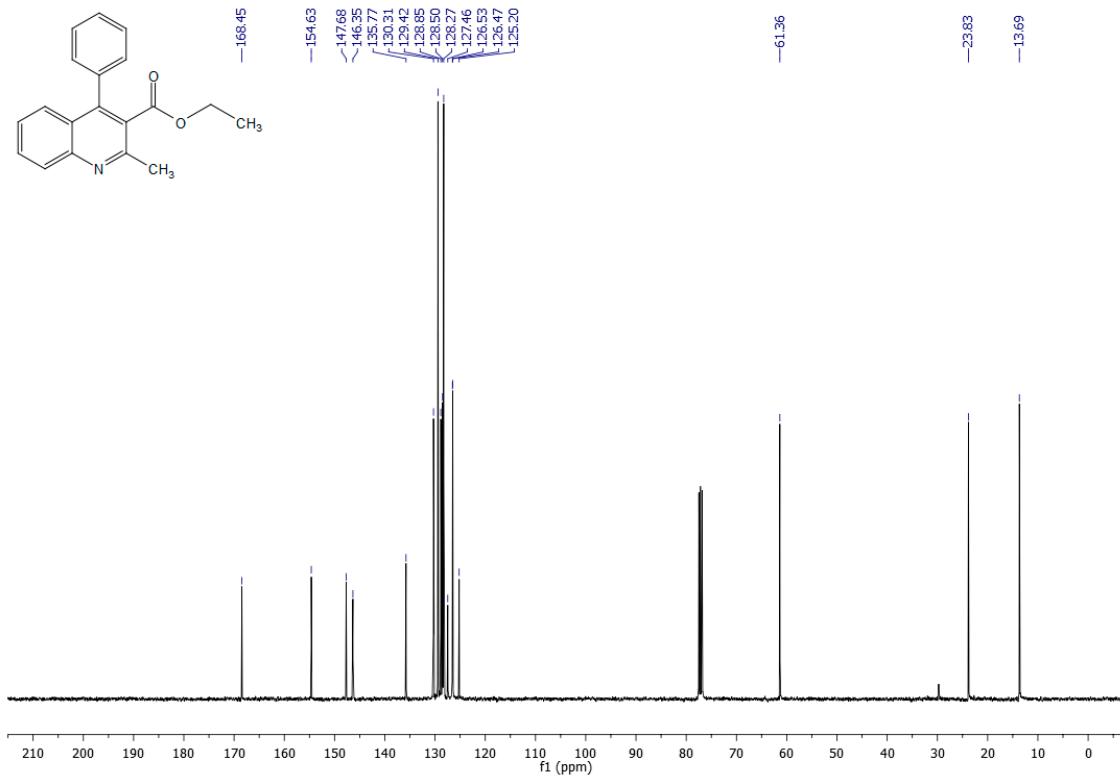
**Figure SI.3.** <sup>1</sup>H NMR spectrum of 3-acetyl-2-methyl-4-phenylquinoline (3)



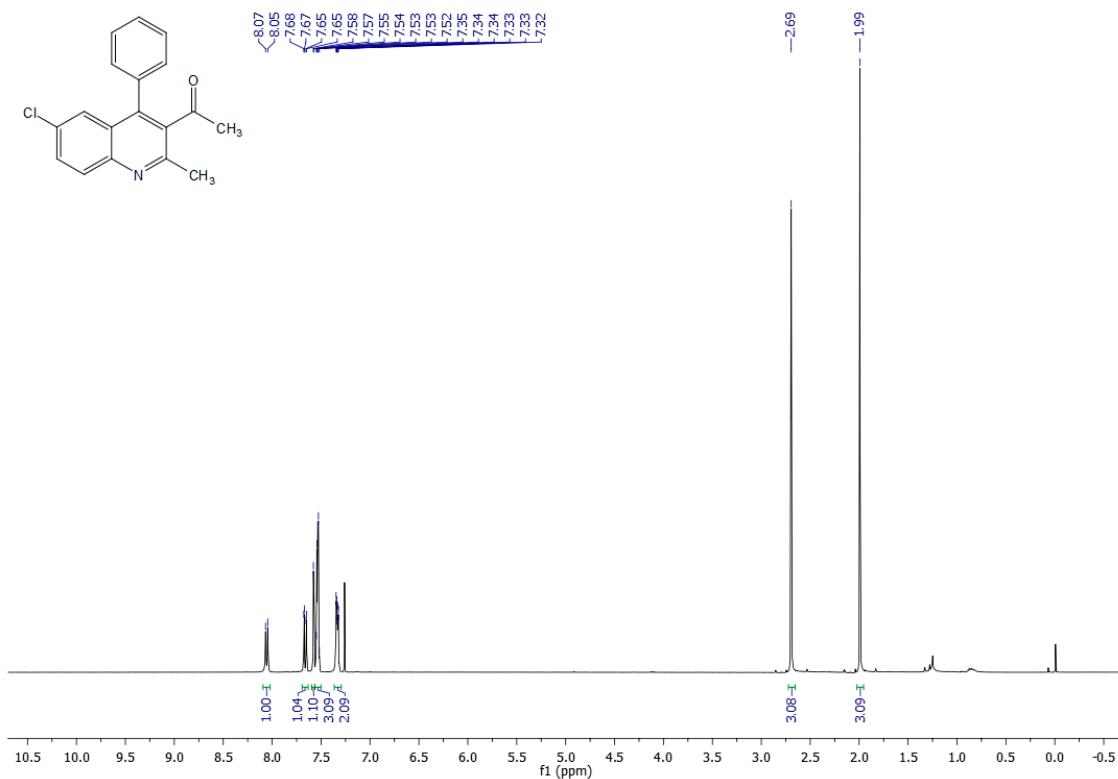
**Figure SI.4.** <sup>13</sup>C NMR spectrum of 3-acetyl-2-methyl-4-phenylquinoline (3)



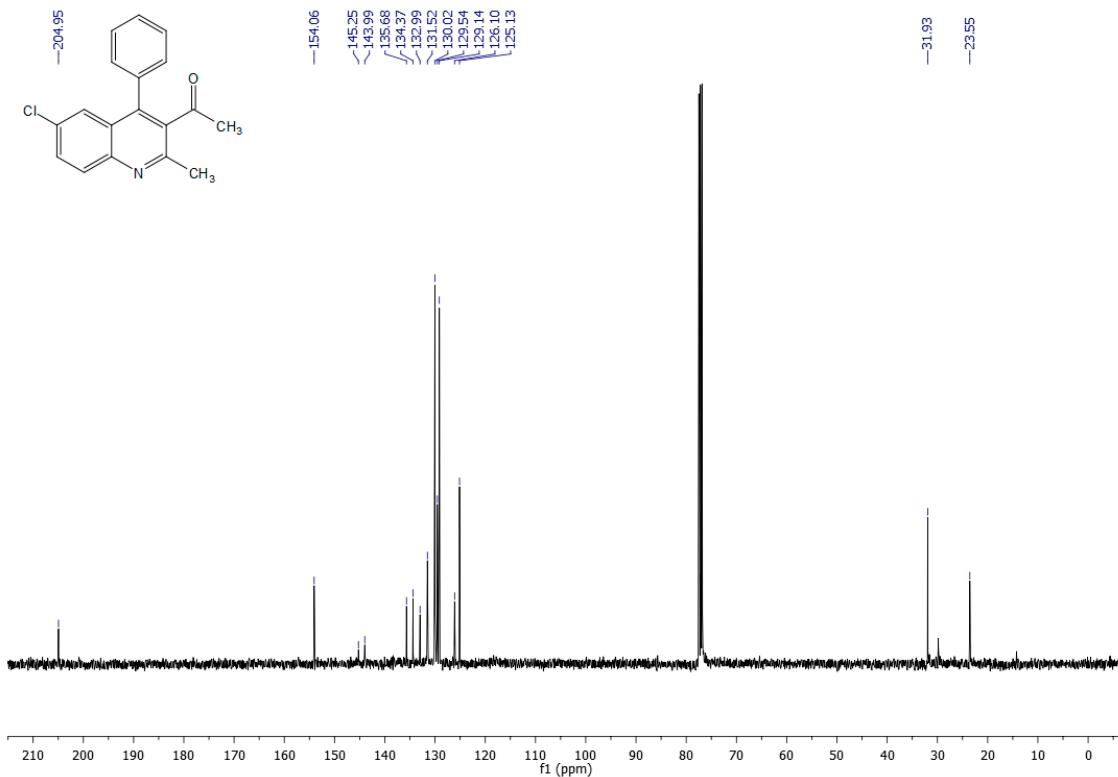
**Figure SI.5.** <sup>1</sup>H NMR spectrum of 3-(ethoxycarbonyl)-2-methyl-4-phenylquinoline (4)



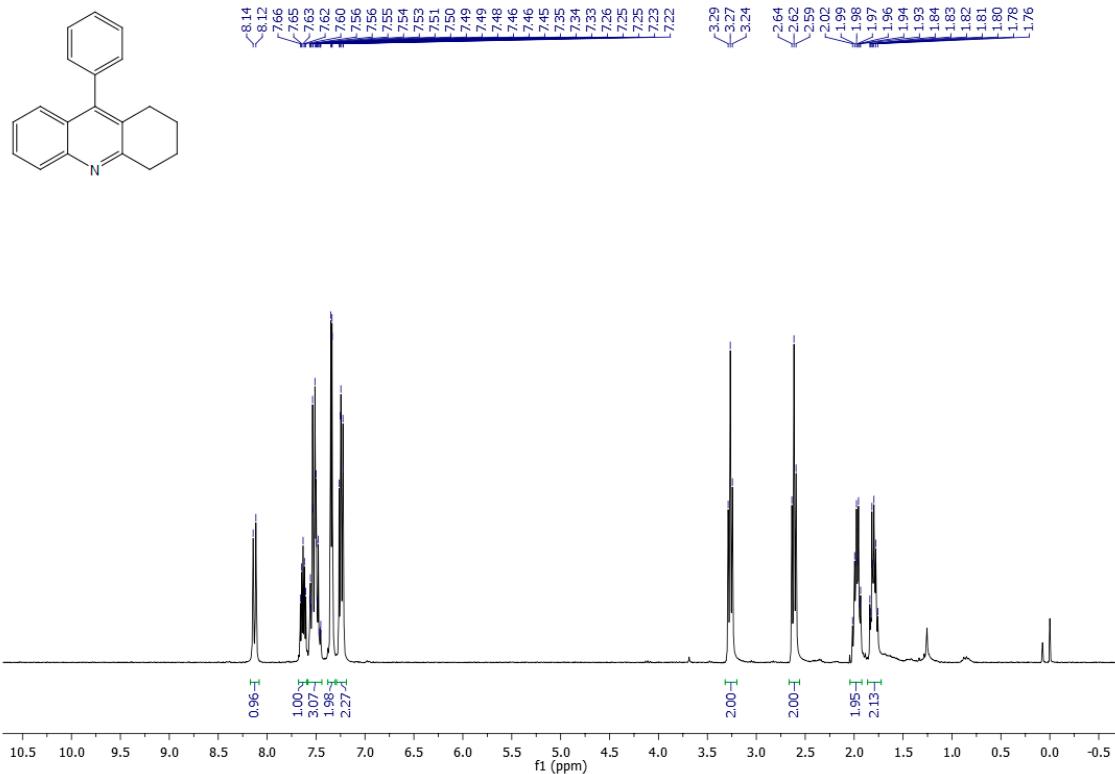
**Figure SI.6.** <sup>13</sup>C NMR spectrum of 3-(ethoxycarbonyl)-2-methyl-4-phenylquinoline (4)



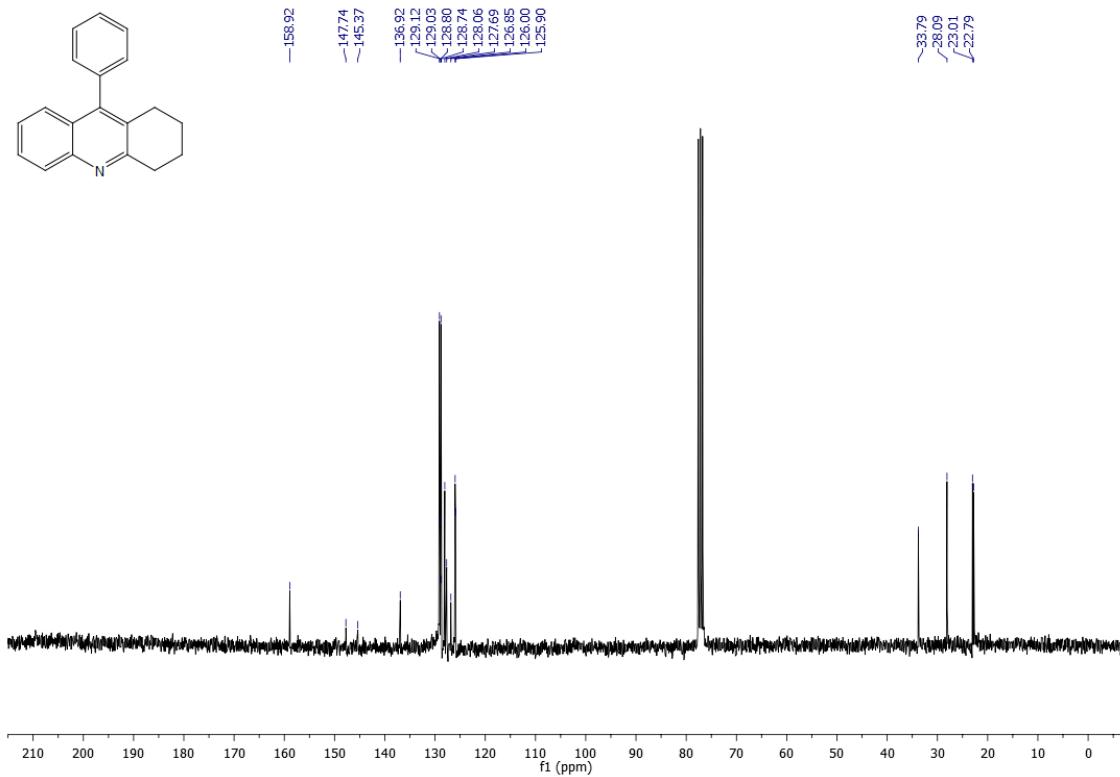
**Figure SI.7.** <sup>1</sup>H NMR spectrum of 3-acetyl-6-chloro-2-methyl-4-phenylquinoline (5)



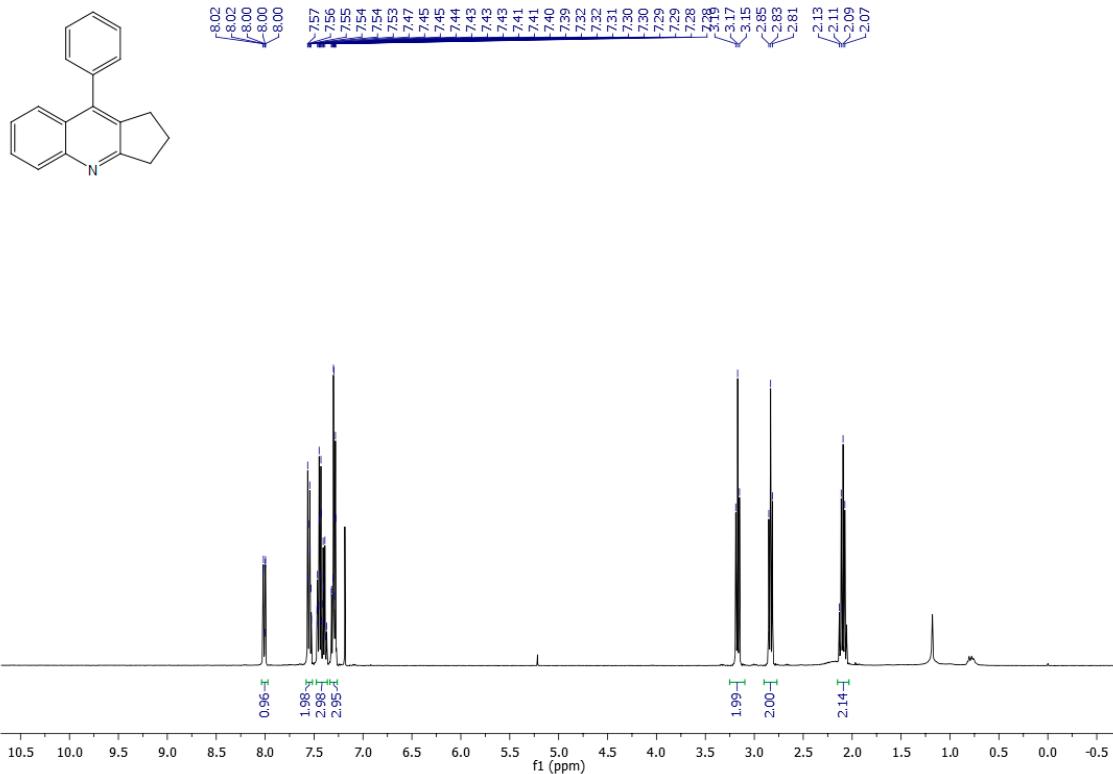
**Figure SI.8.** <sup>13</sup>C NMR spectrum of 3-acetyl-6-chloro-2-methyl-4-phenylquinoline (5)



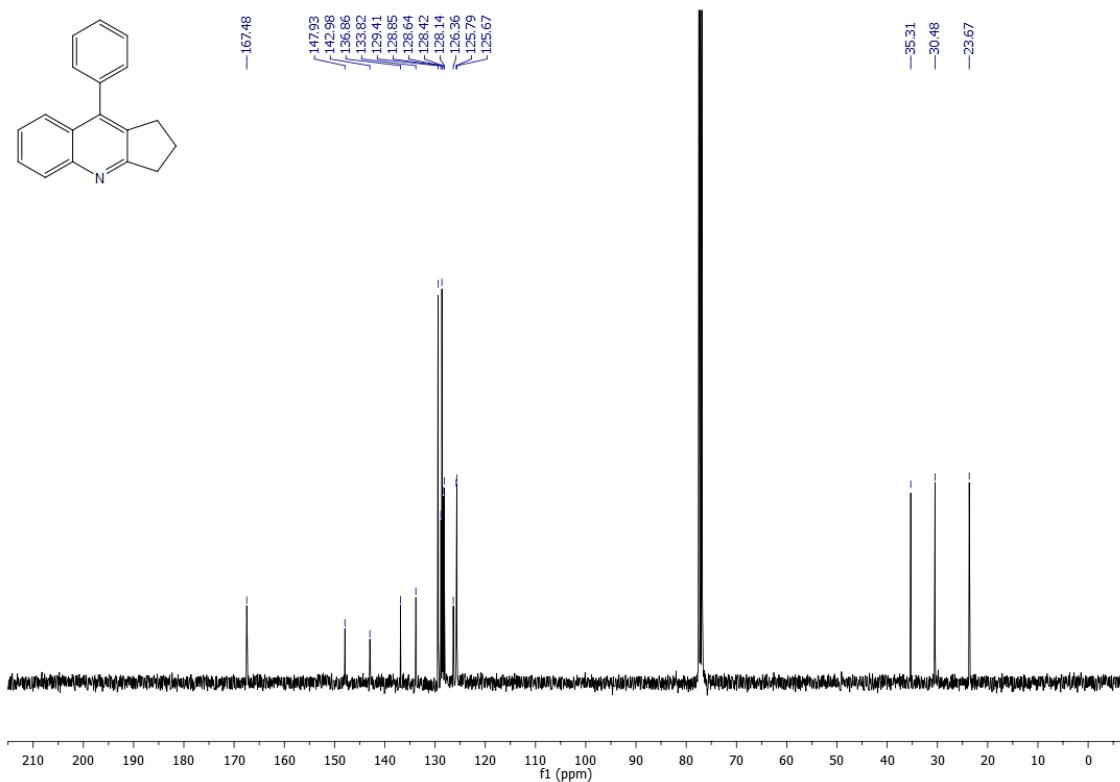
**Figure SI.9.** <sup>1</sup>H NMR spectrum of 9-phenyl-1,2,3,4-tetrahydroacridine (6)



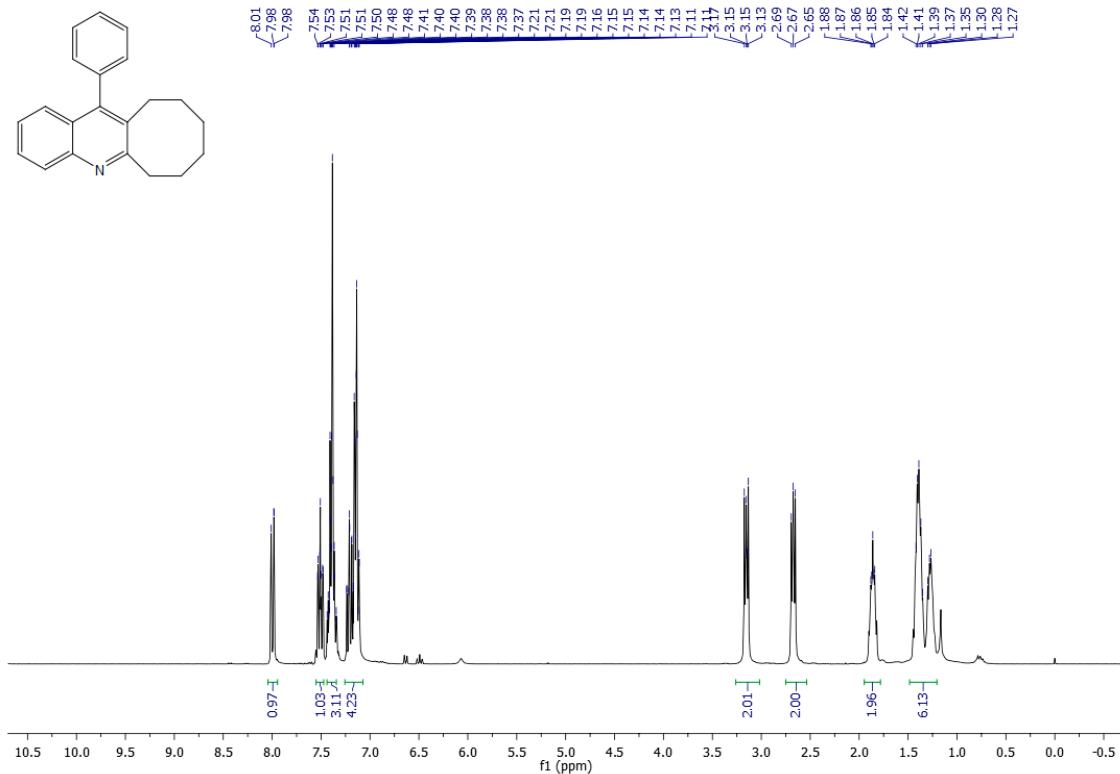
**Figure SI.10.** <sup>13</sup>C NMR spectrum of 9-phenyl-1,2,3,4-tetrahydroacridine (6)



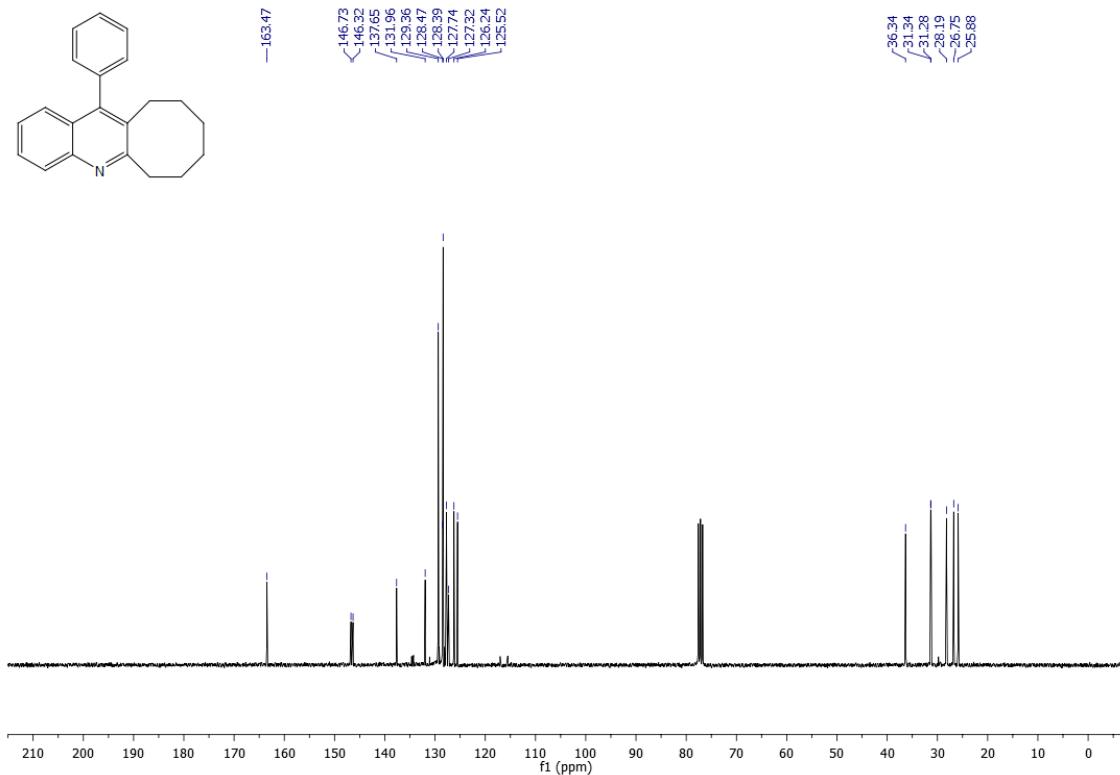
**Figure SI.11.** <sup>1</sup>H NMR spectrum of 9-phenyl-2,3-dihydro-1H-cyclopenta[*b*]quinoline (7)



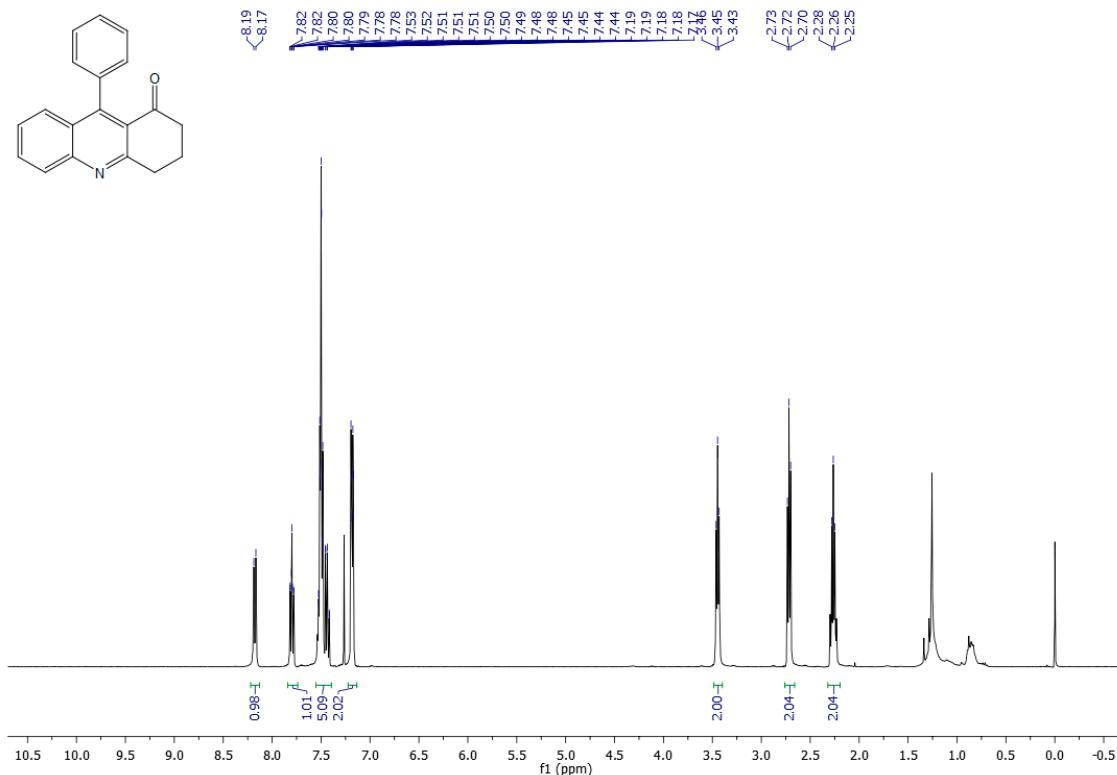
**Figure SI.12.** <sup>13</sup>C NMR spectrum of 9-phenyl-2,3-dihydro-1H-cyclopenta[*b*]quinoline (7)



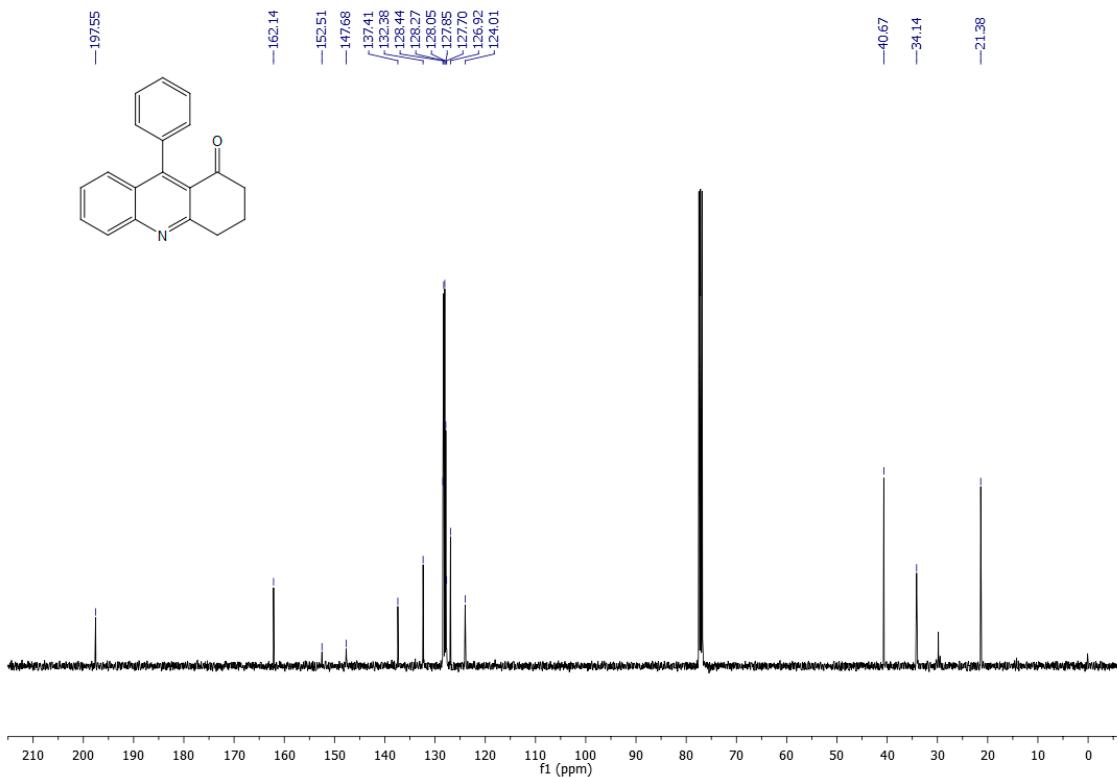
**Figure SI.13.** <sup>1</sup>H NMR spectrum of 12-phenyl-6,7,8,9,10,11-hexahydrocycloocta[b]quinoline (8)



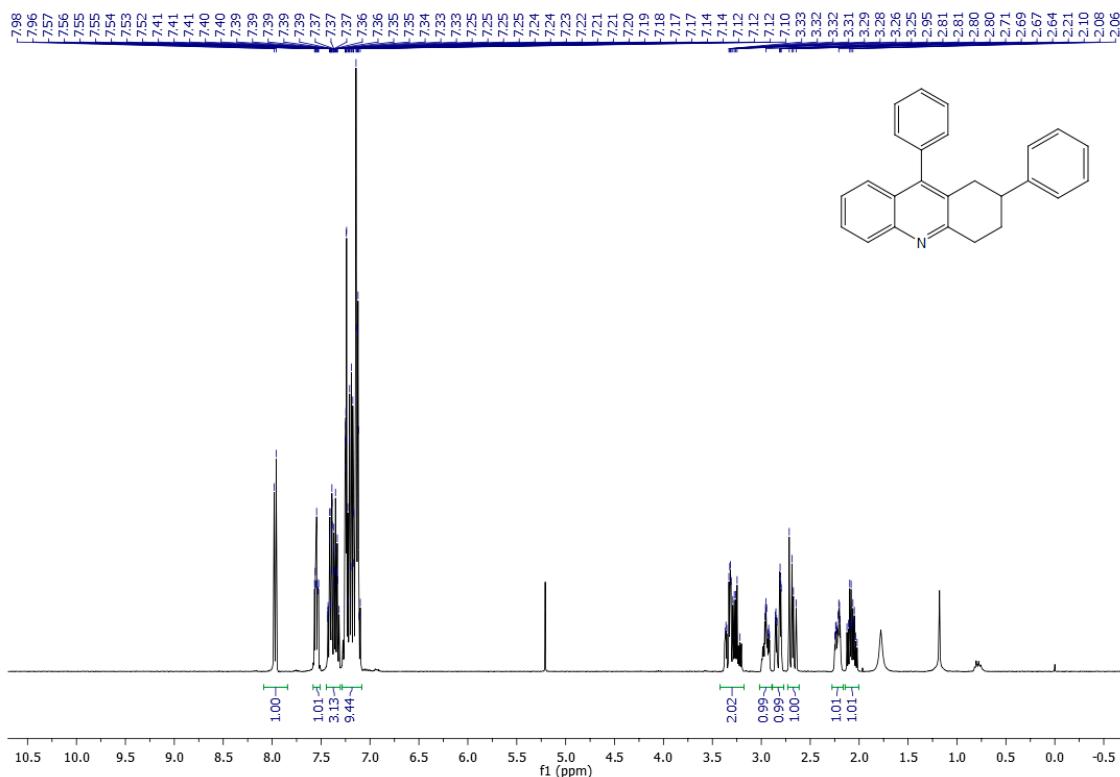
**Figure SI.14.** <sup>13</sup>C NMR spectrum of 12-phenyl-6,7,8,9,10,11-hexahydrocycloocta[b]quinoline (8)



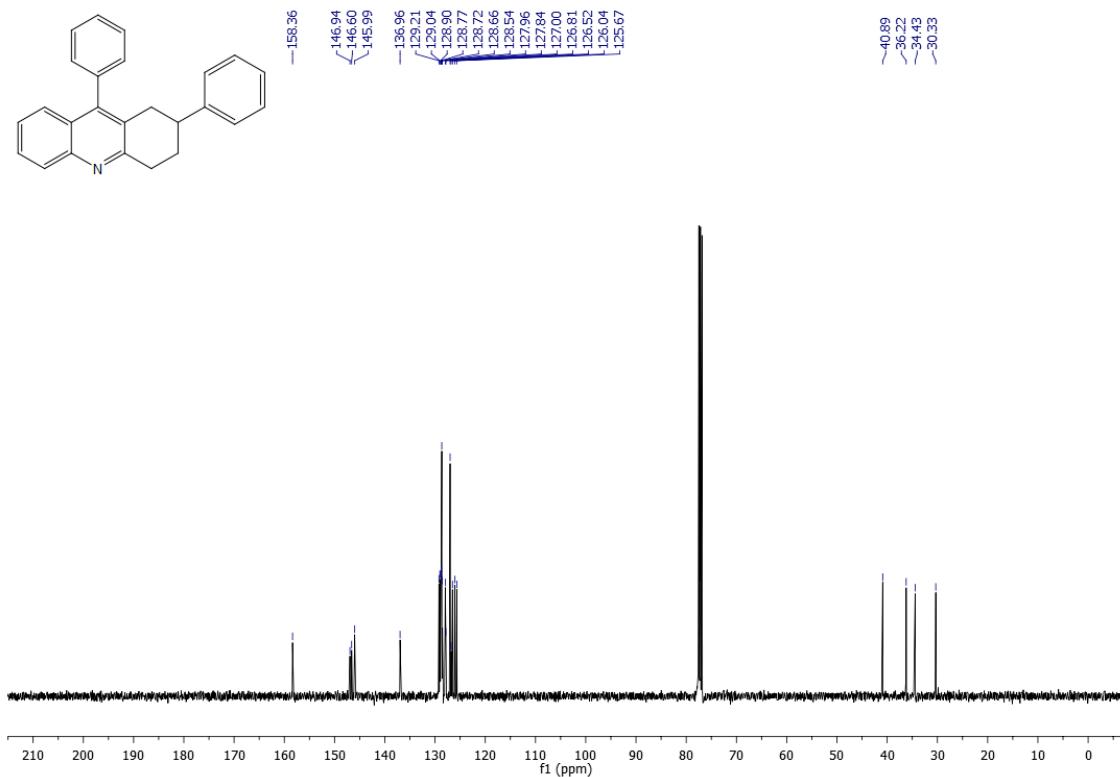
**Figure SI.15.** <sup>1</sup>H NMR spectrum of 9-phenyl-3,4-dihydroacridin-1(2H)-one (9)



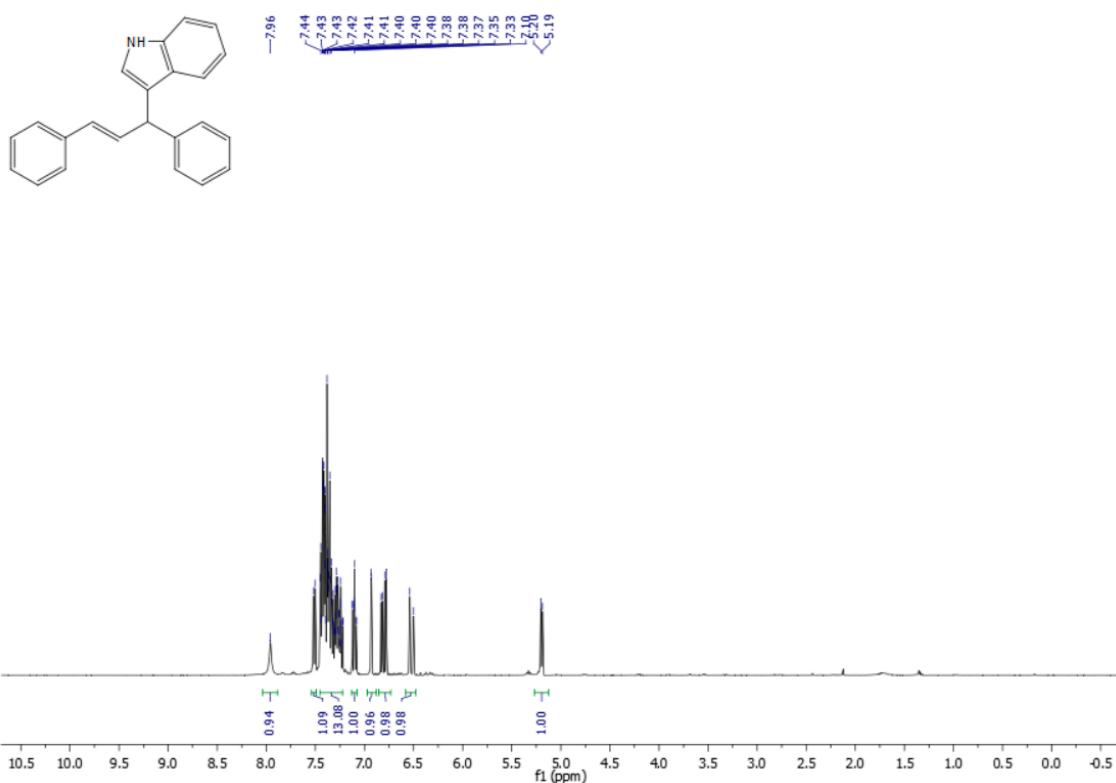
**Figure SI.16.** <sup>13</sup>C NMR spectrum of 9-phenyl-3,4-dihydroacridin-1(2H)-one (9)



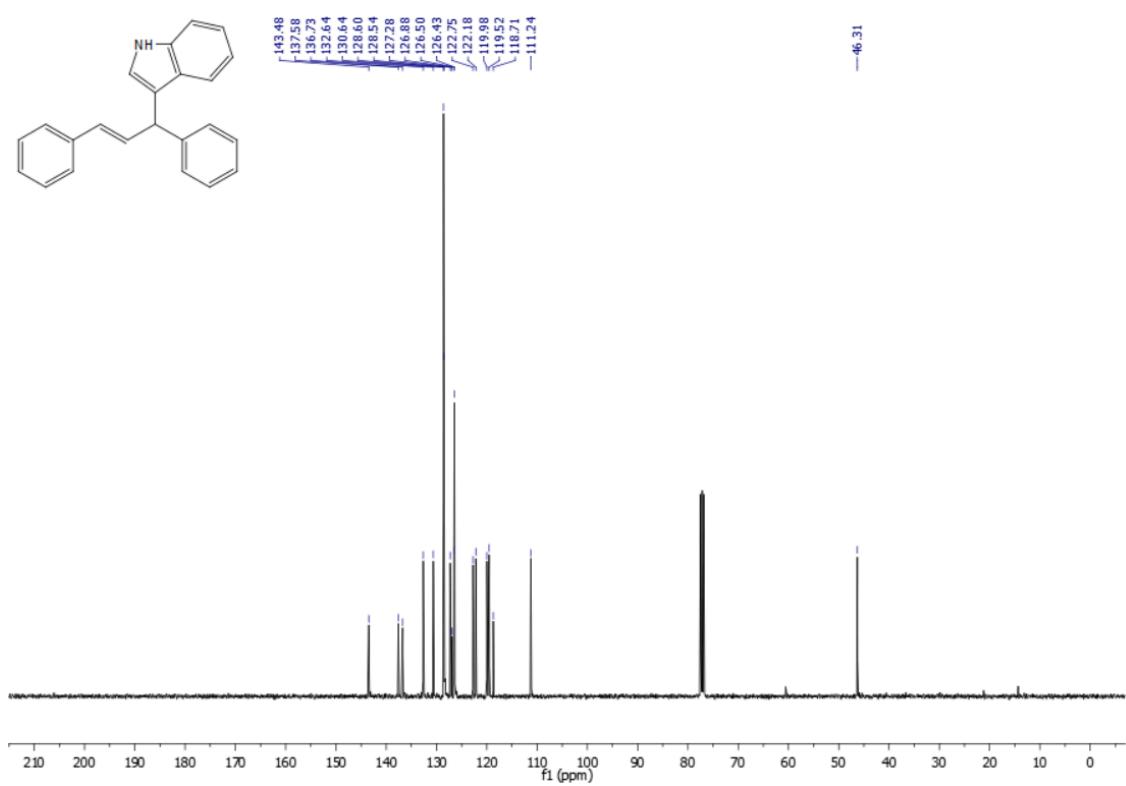
**Figure SI.17.** <sup>1</sup>H NMR spectrum of 2,9-diphenyl-1,2,3,4-tetrahydroacridine (**10**)



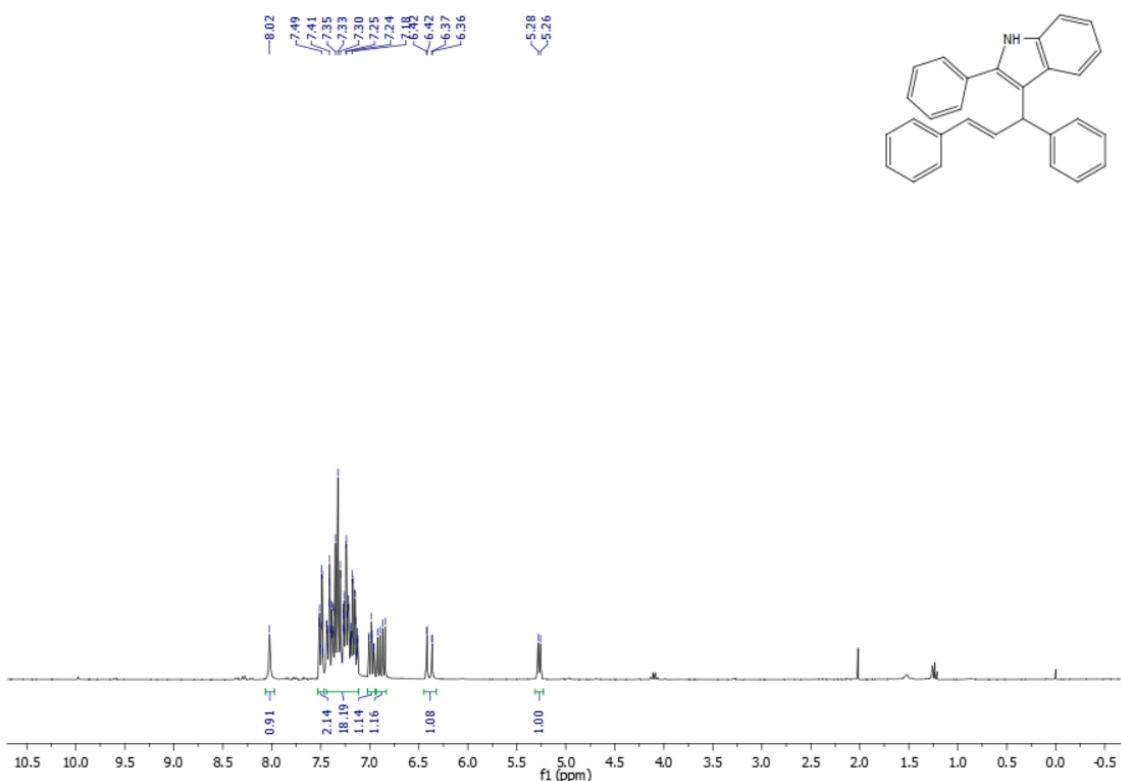
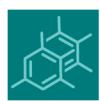
**Figure SI.18.** <sup>13</sup>C NMR spectrum of 2,9-diphenyl-1,2,3,4-tetrahydroacridine (**10**)



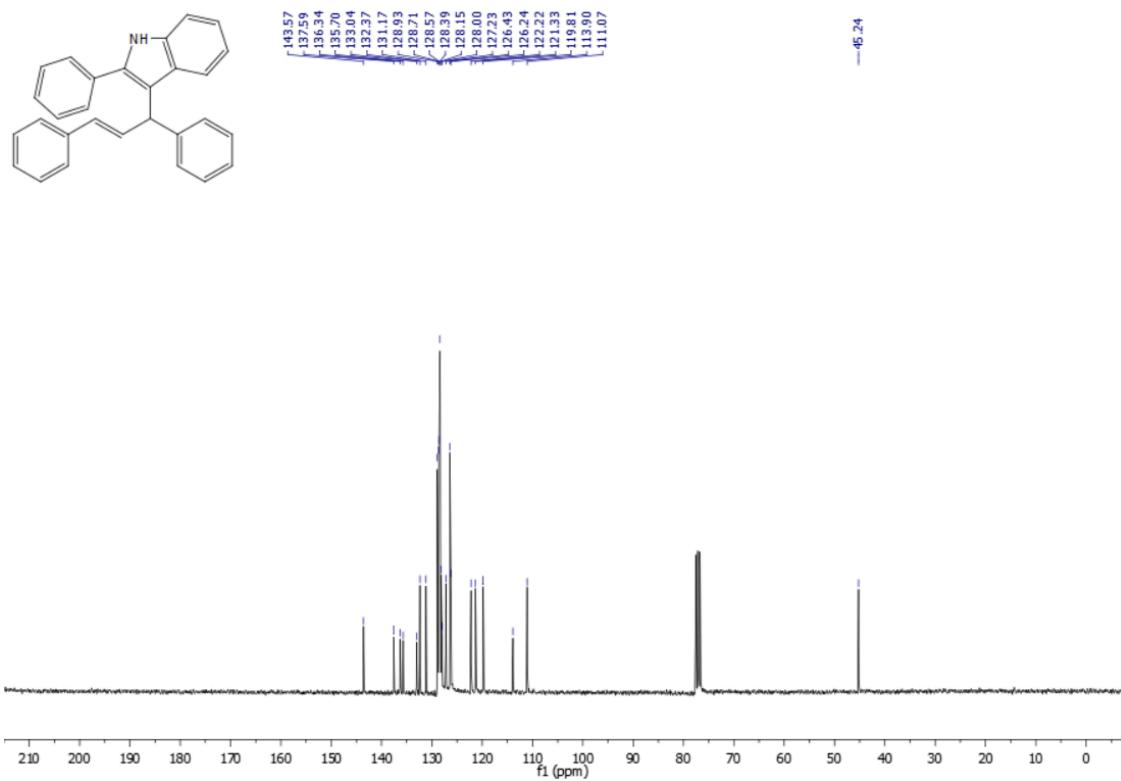
**Figure SI.19.** <sup>1</sup>H NMR spectrum of (E)-3-(1,3-diphenylallyl)-1*H*-indole (**11**)



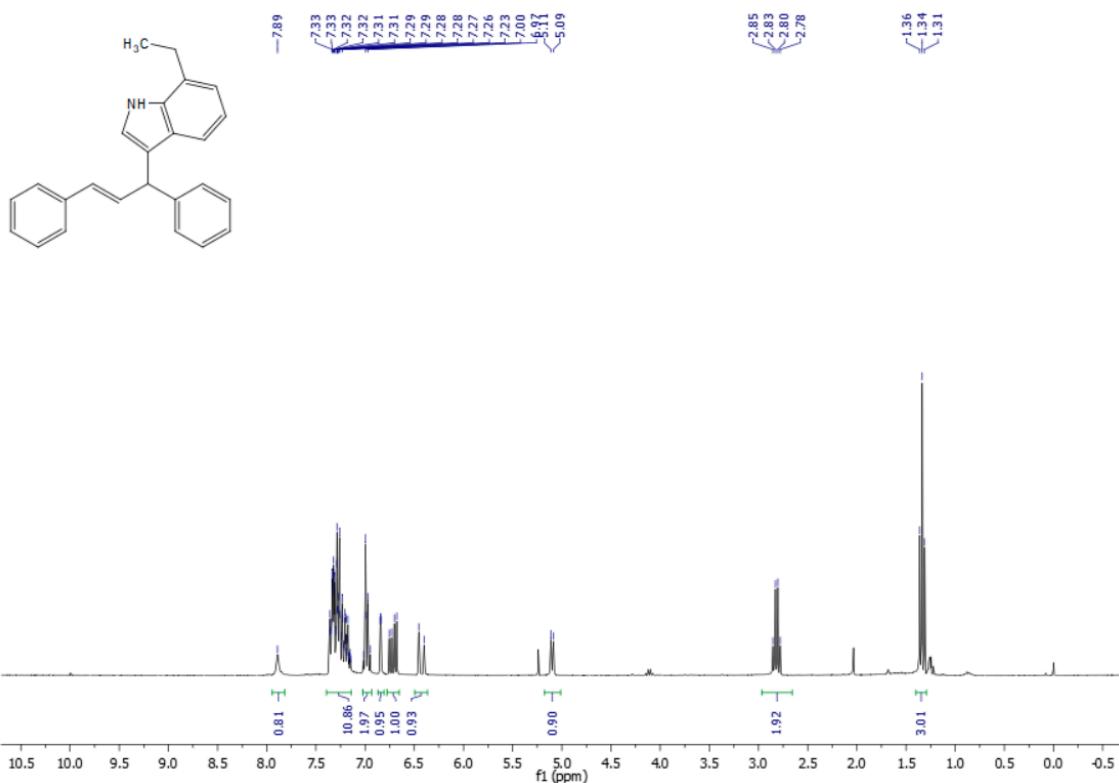
**Figure SI.20.** <sup>13</sup>C NMR spectrum of (E)-3-(1,3-diphenylallyl)-1*H*-indole (**11**)



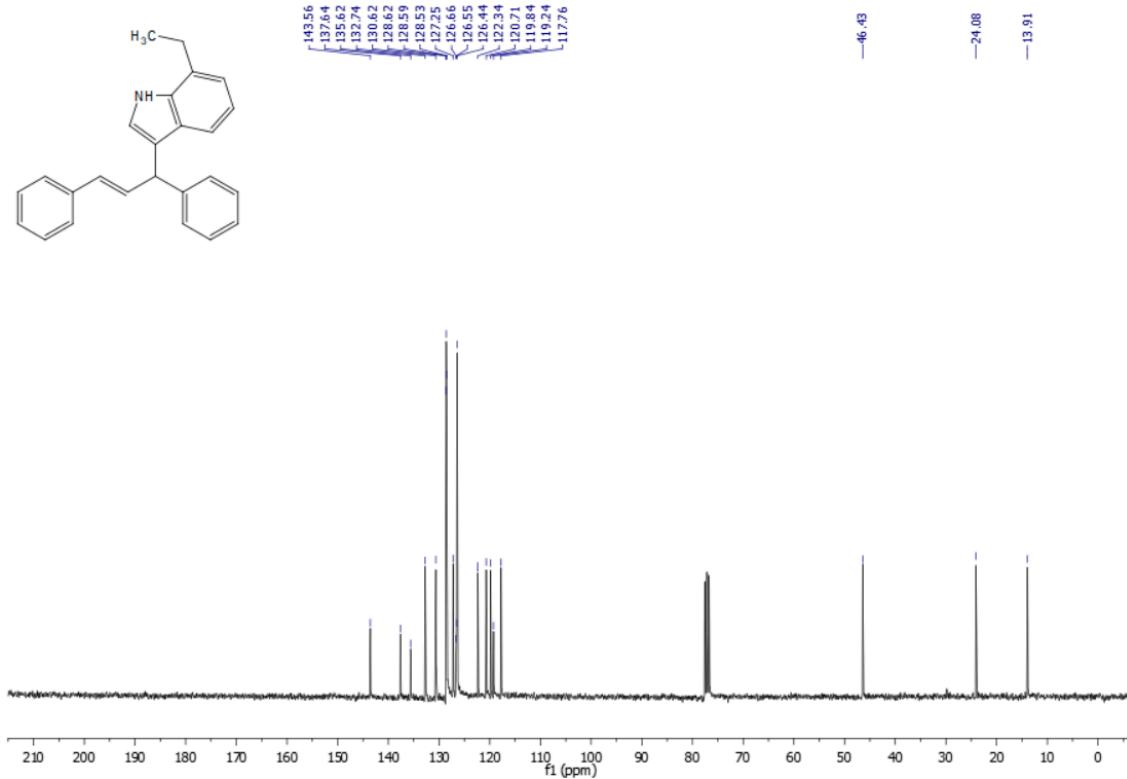
**Figure SI.21.** <sup>1</sup>H NMR spectrum of (E)-3-(1,3-diphenylallyl)-2-phenyl-1*H*-indole (**12**)



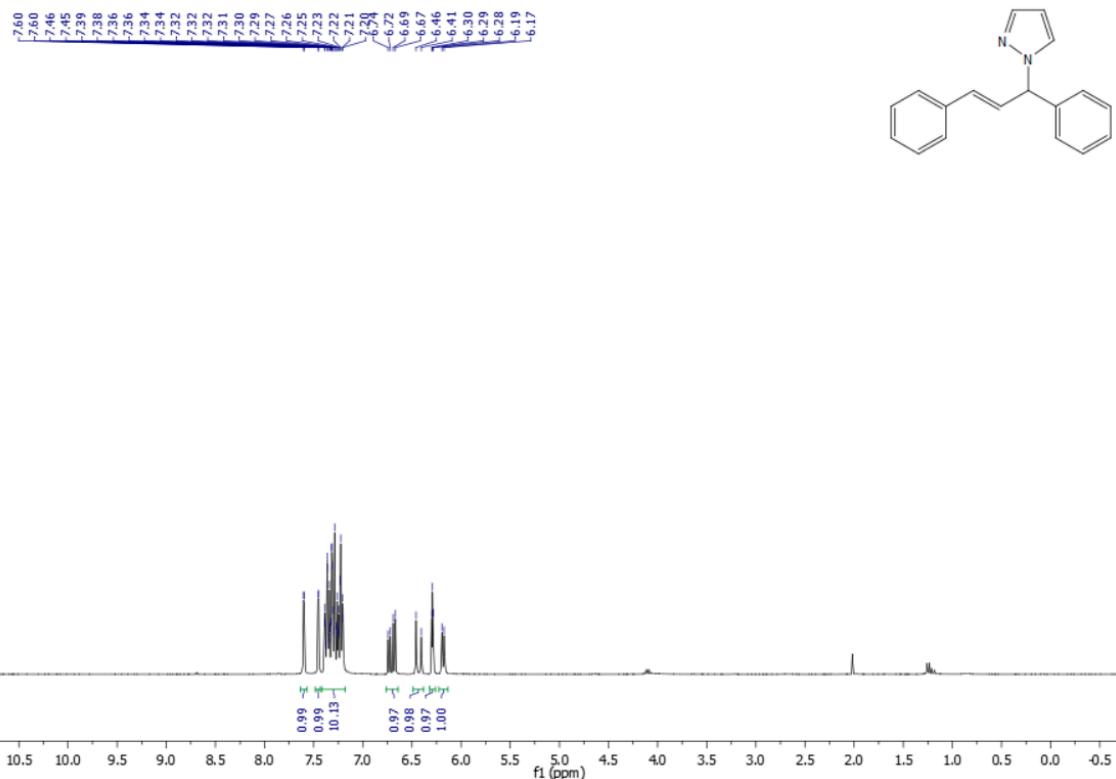
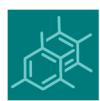
**Figure SI.22.** <sup>13</sup>C NMR spectrum of (E)-3-(1,3-diphenylallyl)-2-phenyl-1*H*-indole (**12**)



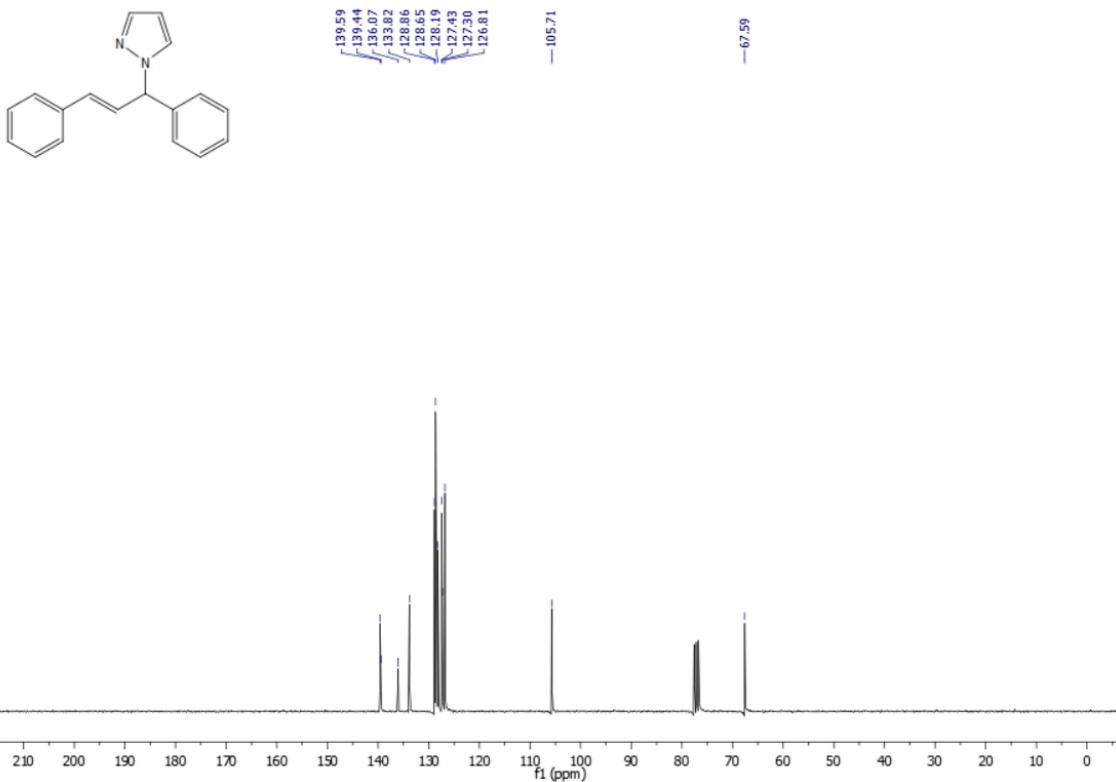
**Figure SI.23.** <sup>1</sup>H NMR spectrum of (E)-3-(1,3-diphenylallyl)-9-ethyl-1*H*-indole (**13**)



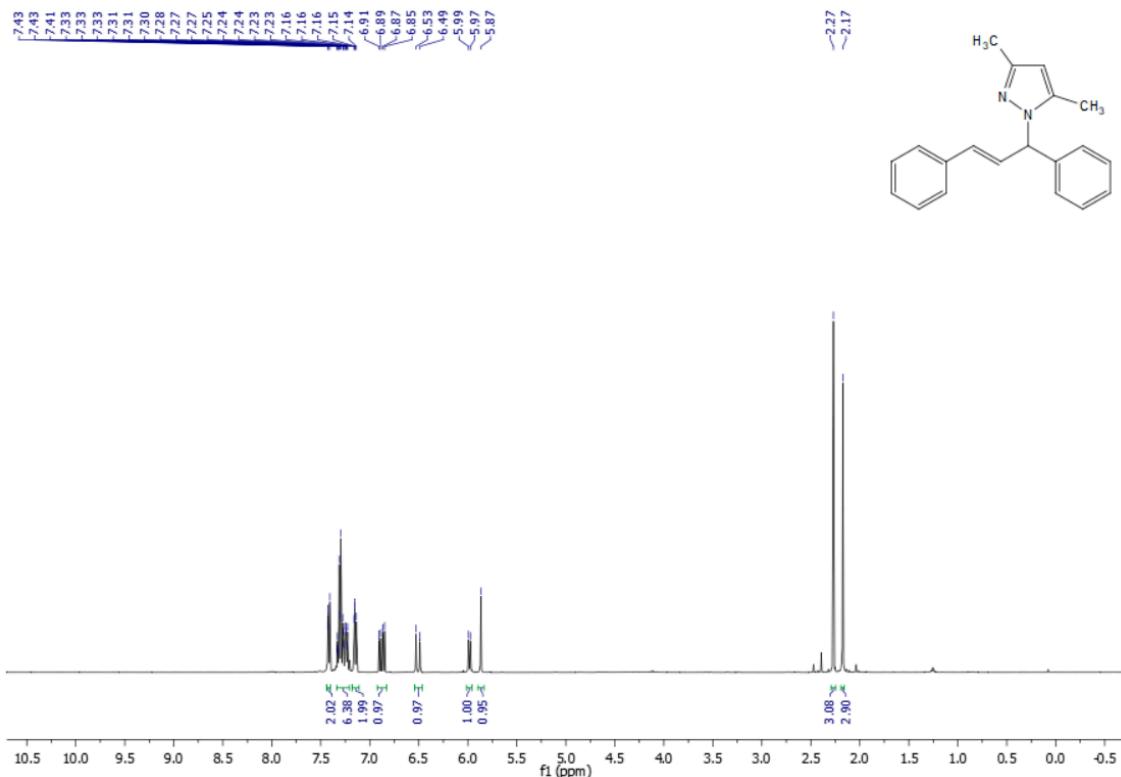
**Figure SI.24** <sup>13</sup>C NMR spectrum of (E)-3-(1,3-diphenylallyl)-9-ethyl-1*H*-indole (**13**)



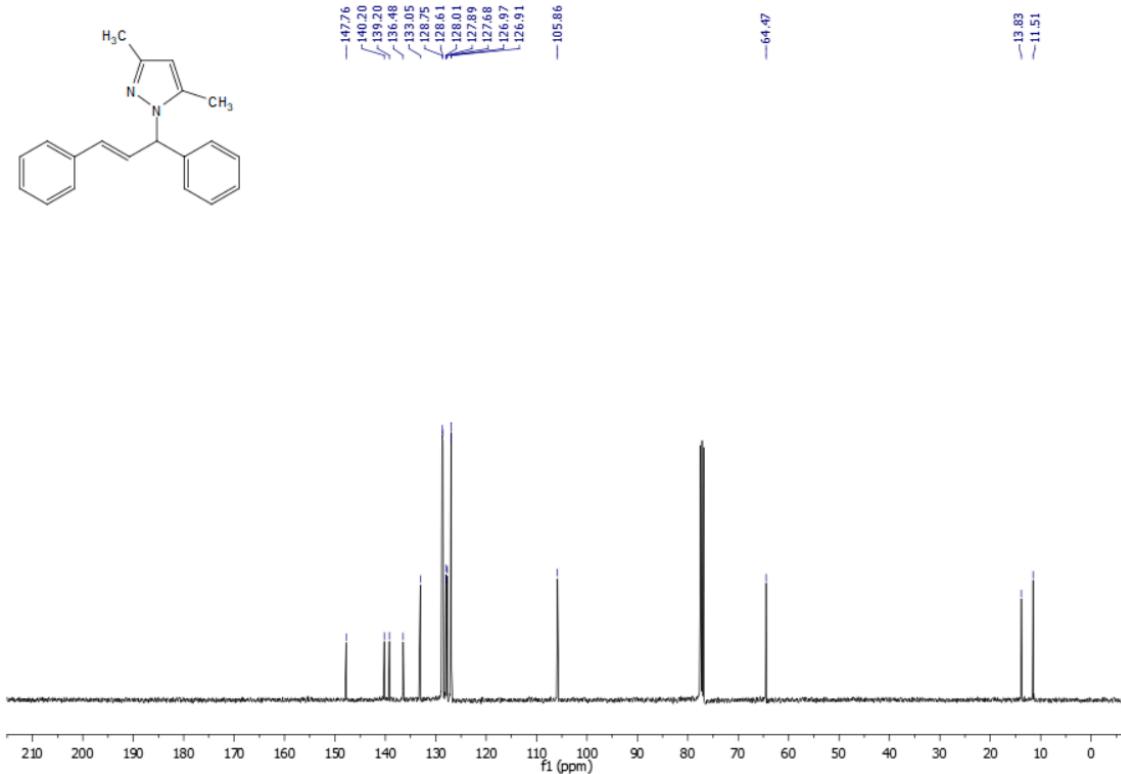
**Figure SI.25.** <sup>1</sup>H NMR spectrum of (E)-1-(1,3-diphenylallyl)-1*H*-pyrazole (**14**)



**Figure SI.26.** <sup>13</sup>C NMR spectrum of (E)-1-(1,3-diphenylallyl)-1*H*-pyrazole (**14**)



**Figure SI.27.** <sup>1</sup>H NMR spectrum of (E)-1-(1,3-diphenylallyl)-3,5-dimethyl-1*H*-pyrazole (**15**)



**Figure SI.28.** <sup>13</sup>C NMR spectrum of (E)-1-(1,3-diphenylallyl)-3,5-dimethyl-1*H*-pyrazole (**15**)

## References

1. Tanwar, B.; Kumar, A.; Yogeeswari, P.; Sriram, D.; Chakraborti, A.K. Design, development of new synthetic methodology, and biological evaluation of substituted quinolines as new anti-tubercular leads. *Bioorg. Med. Chem. Lett.* **2016**, *26*, 5960–5966. doi:10.1016/j.bmcl.2016.10.082.
2. Gisbert, P.; Albert-Soriano, M.; Pastor, I.M. Effective and sustainable access to quinolines and acridines: A heterogeneous imidazolium salt mediates C-C and C-N bond formation. *Eur. J. Org. Chem.* **2019**, *2019*, 4928–4940. <https://doi.org/10.1002/ejoc.201900880>.