Supporting Information

Iodine-promoted sequential C(sp^3)–H oxidation and cyclization of aryl methyl ketones with 2-(2-aminophenyl)quinazolin-4(3*H*)-ones

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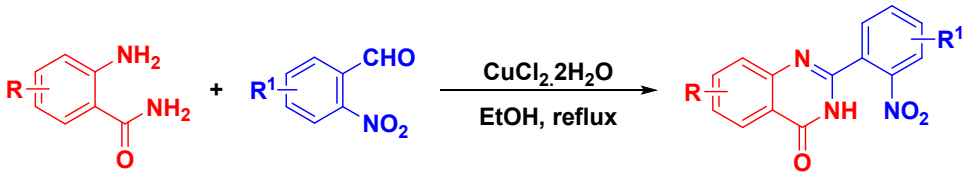
General Methods

Commercially available chemicals were purchased from Alfa Aesar, Sigma-Aldrich, TCI, Finar, Avra and used as received. All the starting materials were synthesized according to the reported procedures. Thin-layer chromatography (TLC) was performed using Merck silica gel 60 F254 precoated plates (0.25 mm) and visualized by a UV lamp for reaction monitoring. Silica gel for column chromatography (particle size 100–200 mesh) was purchased from Avra Synthesis Private Ltd India. ^{1}H and ^{13}C{^{1}H} NMR (nuclear magnetic resonance) spectra were recorded on a Bruker 400 MHz instrument. Chemical shifts were recorded in parts per million (ppm) relative to tetramethylsilane (δ 0.00), chloroform (7.26 ppm) or DMSO-d_6 (2.50 ppm). ^{1}H NMR splitting patterns are designated as s (singlet), d (doublet), t (triplet), q (quartet), dd (doublet of doublets), m (multiplet), *etc*. ^{13}C{^{1}H} NMR spectral values were reported relative to $CDCl_3$ (77.16 ppm) or DMSO-d_6 (39.52 ppm). FT-IR spectra are recorded on a Perkin Elmer spectrometer and reported in the absorption frequency (cm^{-1}). HRMS were analyzed with Agilent Q-TOF 6230.

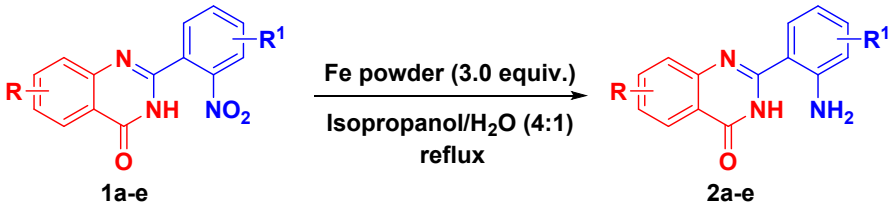
General procedure for the synthesis of 2-(2-nitrophenyl)-6,7-dihydroquinazolin-4(3*H*)-ones (1a-e)[1]



1a-e

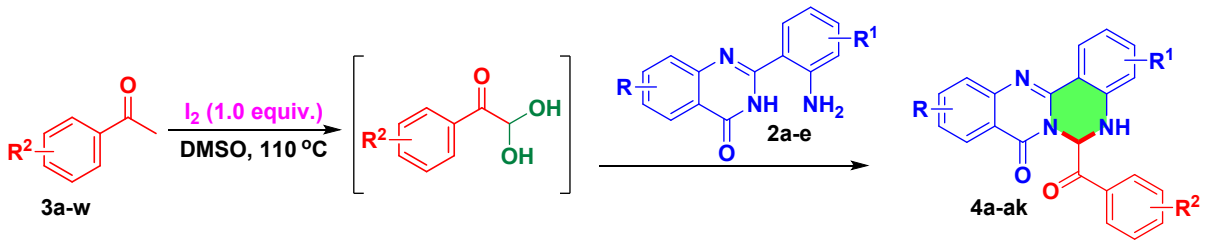
An oven-dried round bottom flask containing the solution of anthranilamide (1.0 g, 7.34 mmol, 1.0 equiv.), 2-nitrobenzaldehyde (1.2 g, 8.08 mmol, 1.1 equiv.), and $CuCl_2.2H_2O$ (2.3 g, 14.69 mmol, 2.0 equiv.) in ethanol (15 mL) was refluxed in an oil bath for 12 h, and the progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was cooled to room temperature, and distilled water was added until precipitates formed. The precipitate was filtered, washed with distilled water, and recrystallized in EtOH to get the corresponding nitro compound **1a** with a 96% yield, 1.88 g. Compounds **1b-e** were synthesized by following the same procedure.

General procedure for the synthesis of 2-(2-aminophenyl)-6,7-dihydroquinazolin-4(3*H*)-ones (2a-e)[1]



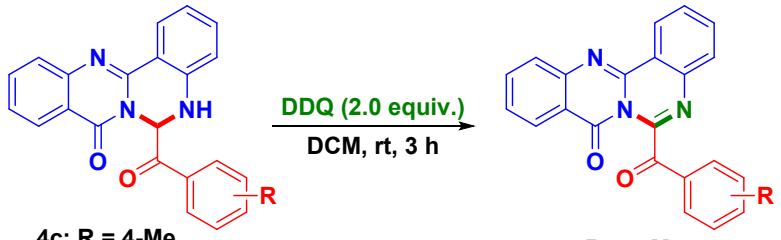
The mixture of 2-(2-nitrophenyl)-6,7-dihydroquinazolin-4(3*H*)-one **1a** (1.0 g, 3.74 mmol. 1.0 equiv.), iron powder (627.0 mg, 11.23 mmol, 3.0 equiv.) in isopropanol/H_2O (4:1) (20 mL : 5 mL) was refluxed for 4-5 h and the progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was washed with water and extracted with EtOAc three times. The combined organic layers were washed with saturated brine solution, dried with Na_2SO_4 and the solvent was evaporated under a vacuum. The residue was purified by column chromatography over silica gel with hexane/ethyl acetate to afford the corresponding amine **2a** as pale yellow solid in 79% yield, 0.70 g. Compounds **2b-e** were synthesized by following the same procedure.

General procedure for the synthesis of 6-aroyl-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-ones 4a-4ak. (4a as an example)



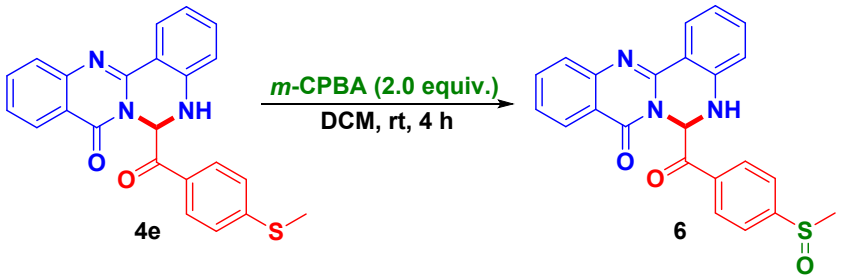
To an oven-dried reaction tube equipped with a magnetic stir bar was added acetophenone **3a** (50.1 mg, 0.42 mmol, 1.0 equiv.) and iodine (107.0 mg, 0.42 mmol, 1.0 equiv.) and 2 mL of DMSO. The resulting reaction mixture was stirred in an oil bath at 110 °C until the formation of phenylglyoxal (2 h) which was monitored by TLC. Then, 2-(2-aminophenyl)-6,7-dihydroquinazolin-4(3*H*)-one **2a** (100 mg, 0.42 mmol, 1.0 equiv.) was added to the reaction mixture. The reaction mixture was further stirred for 15 min at the same temperature and the progress of the reaction was monitored by TLC. After the completion of the reaction, the reaction mixture was washed with sodium thiosulphate to quench iodine and extracted with EtOAc (20 mL × 3). The combined organic layers were washed with brine, dried with Na_2SO_4, and the solvent was evaporated under a vacuum. The crude product was purified by column chromatography on silica gel (eluent: petroleum ether/EtOAc = 7:3, v/v) to afford pure product **4a** in 136 mg, 91% yield. All other compounds **4b-ak** were synthesized by following the same procedure.

General procedure for the synthesis of 6-benzoyl-8*H*-quinazolino[4,3-*b*]quinazolin-8-ones (5a-b)[2]



An oven-dried reaction tube containing a solution of compound **4c** (50 mg, 0.14 mmol, 1.0 equiv.) in 3 mL DCM was added DDQ (64.2 mg, 0.28 mmol, 2.0 equiv.). The reaction mixture was allowed to stir at room temperature for 3 h and the progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was washed with water and extracted with EtOAc three times. The combined organic layers were washed with saturated brine solution and dried with Na_2SO_4 and the solvent was evaporated under a vacuum. The residue was purified by column chromatography over silica gel with hexane/ethyl acetate to afford the corresponding compound **5a** as a white solid in 37 mg, 74% yield. The same procedure was followed to obtain compound **5b** from compound **4h**.

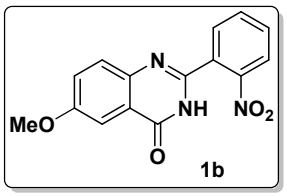
Procedure for the synthesis of 6-(4-(methylsulfinyl)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (6)[3]



To the oven-dried round bottom flask containing a solution of **4e** (100 mg, 0.25 mmol, 1.0 equiv.) in 4 mL of DCM was added *m*-chloroperoxybenzoic acid (86.4 mg, 0.50 mmol, 2.0 equiv.). The reaction mixture was stirred at room temperature and the progress of the reaction was monitored by TLC. After the completion of the reaction, the reaction mixture was washed with water and extracted with EtOAc (20 mL × 3). The combined organic layers were washed with brine, dried with anhydrous Na_2SO_4, and concentrated under a vacuum. The crude product was purified by silica gel column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v) to obtain the sulfoxide **6** as a yellow solid yield in 76 mg, 73% yield.

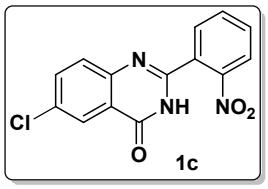
Physical and Spectroscopic data

6-Methoxy-2-(2-nitrophenyl)quinazolin-4(3*H*)-one (1b)



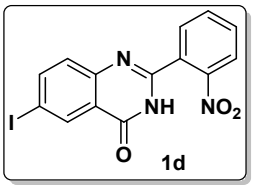
Filtered, washed with water and recrystallized in EtOH; Green solid; Yield: 1.63 g, 91%; m.p.: 217-218 ^{o}C; FT-IR (KBr) ν (cm^{-1}) 3329, 3157, 3091, 3024, 2933, 2886, 1649, 1608, 1522, 1484, 1308, 1220, 1138, 1086; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.78 (s, 1H), 8.19 (s, 1H), 8.01 – 7.74 (m, 3H), 7.71 – 7.33 (m, 3H), 3.90 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 161.1, 157.9, 149.1, 147.4, 142.8, 133.7, 131.3, 131.2, 129.0, 124.4, 123.9, 121.9, 105.8 (2C), 55.5; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{15}H_{12}N_3O_4$: 298.0828; found: 298.0824.

6-Chloro-2-(2-nitrophenyl)quinazolin-4(3*H*)-one (1c)



Filtered, washed with water and recrystallized in EtOH; White Solid; Yield: 1.6 g, 90%; m.p.: 255-256 ^{o}C; FT-IR (KBr) ν (cm^{-1}) 3330, 3028, 2931, 1674, 1601, 1527, 1458, 1350, 1315, 1149, 1071, 835, 756; ^{1}H NMR (400 MHz, DMSO-d_6) δ 13.02 (s, 1H), 8.23 (d, J = 7.7 Hz, 1H), 8.13 (s, 1H), 8.01 – 7.78 (m, 4H), 7.69 (d, J = 8.5 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 161.1, 152.7, 152.6, 147.8, 135.2, 135.1, 134.5, 132.1, 131.8, 130.2, 125.5, 125.1, 123.3, 109.4; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{14}H_9ClN_3O_3$: 302.0332; found: 302.0333.

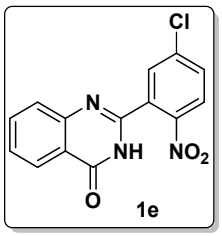
6-Iodo-2-(2-nitrophenyl)quinazolin-4(3*H*)-one (1d)



Filtered, washed with water and recrystallized in EtOH; Colourless solid; Yield: 1.2 g, 80%; m.p.: 263-264 ^{o}C; FT-IR (KBr) ν (cm^{-1}) 3335, 3080, 3030, 2936, 1680, 1595, 1526, 1454, 1402, 1346, 1315, 1146, 1085, 512; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.98 (s, 1H), 8.45 (s, 1H), 8.22 (d, J = 8.1 Hz, 1H), 8.14 (d, J = 8.4 Hz, 1H), 7.94 – 7.82 (m, 3H), 7.45 (d, J = 8.5 Hz,

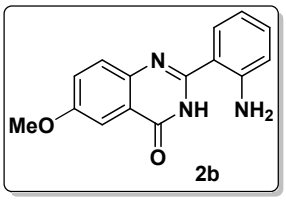
1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 160.1, 152.3, 147.8, 147.3, 143.0, 134.2, 134.0, 131.7, 131.4, 129.5, 129.0, 124.5, 122.9, 92.2; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{14}H_9IN_3O_3$: 393.9689; found: 393.9689.

2-(5-Chloro-2-nitrophenyl)quinazolin-4(3*H*)-one (1e)



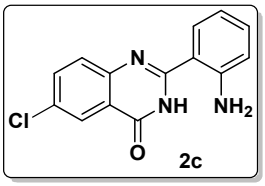
Filtered, washed with water and recrystallized in EtOH; White solid; Yield: 1.6 g, 72%; m.p.: 280-281 °C; FT-IR (KBr) ν (cm^{-1}) 3329, 3032, 2926, 1678, 1610, 1526, 1460, 1358, 1315, 1132, 1069, 830, 754; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.84 (s, 1H), 8.25 (d, J = 8.8 Hz, 1H), 8.19 (dd, J = 8.0, 1.6 Hz, 1H), 8.04 (d, J = 2.3 Hz, 1H), 7.92 (dd, J = 8.8, 2.3 Hz, 1H), 7.86 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.67 (d, J = 8.1 Hz, 1H), 7.59 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 161.4, 150.4, 148.4, 146.0, 138.6, 134.8, 131.4, 131.3, 130.8, 127.5, 127.4, 126.6, 126.0, 121.3; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{14}H_9ClN_3O_3$: 302.0332; found: 302.0333.

2-(2-Aminophenyl)-6-methoxyquinazolin-4(3*H*)-one (2b)



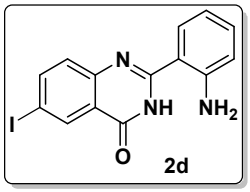
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6:4, v/v); Pale yellow solid; Yield: 678 mg, 75%; m.p.: 220-221 °C; FT-IR (KBr) ν (cm^{-1}) 3433, 3372, 3204, 3111, 3001, 2938, 2846, 1670, 1617, 1582, 1546, 1456, 1365, 1220, 1027, 825, 739; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.09 (s, 1H), 7.76 – 7.65 (m, 2H), 7.53 (d, J = 3.0 Hz, 1H), 7.42 (dd, J = 8.9, 3.0 Hz, 1H), 7.17 (t, J = 7.7 Hz, 1H), 7.00 (s, 2H), 6.82 (d, J = 8.2 Hz, 1H), 6.59 (t, J = 7.6 Hz, 1H), 3.88 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 161.9, 157.5, 151.4, 149.1, 142.5, 131.4, 128.6 (2C), 124.0, 121.2, 116.5, 115.0, 112.5, 105.8, 55.6; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{15}H_{14}N_3O_2$: 268.1086; found: 268.1084.

2-(2-Aminophenyl)-6-chloroquinazolin-4(3*H*)-one (2c)



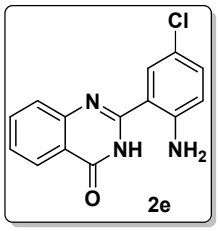
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Pale yellow solid; Yield: 650 mg, 72%; m.p.: 261-262 °C; FT-IR (KBr) ν (cm^{-1}) 3454, 3336, 3114, 3072, 2957, 16766, 1613, 1544, 1508, 1406, 1338, 1262, 1156, 1070, 734; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.38 (s, 1H), 8.06 (d, J = 2.5 Hz, 1H), 7.83 (dd, J = 8.7, 2.5 Hz, 1H), 7.77 (d, J = 8.7 Hz, 1H), 7.73 (dd, J = 8.1, 1.5 Hz, 1H), 7.21 (ddd, J = 8.4, 7.0, 1.5 Hz, 1H), 7.12 (s, 2H), 6.84 (dd, J = 8.3, 1.2 Hz, 1H), 6.60 (ddd, J = 8.2, 7.1, 1.2 Hz, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 161.1, 154.0, 149.5, 146.9, 134.6, 132.0, 130.3, 129.2, 128.9, 124.7, 121.7, 116.7, 115.0, 112.0; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{14}H_{11}ClN_3O$: 272.0591; found: 272.0590.

2-(2-Aminophenyl)-6-iodoquinazolin-4(3*H*)-one (2d)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Pale yellow solid; Yield: 650 mg, 70%; m.p.: 239-240 °C; FT-IR (KBr) ν (cm^{-1}) 3445, 3371, 3108, 3069, 2921, 1675, 1593, 1543, 1462, 1397, 1262, 1153, 1117, 827, 502; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.07 (s, 1H), 8.39 (d, J = 2.1 Hz, 1H), 8.08 (dd, J = 8.5, 2.1 Hz, 1H), 7.73 (dd, J = 8.1, 1.5 Hz, 1H), 7.54 (d, J = 8.6 Hz, 1H), 7.21 (ddd, J = 8.4, 7.0, 1.5 Hz, 1H), 7.13 (s, 2H), 6.83 (dd, J = 8.3, 1.2 Hz, 1H), 6.60 (t, J = 7.4 Hz, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 161.3, 154.6, 150.0, 147.9, 143.2, 134.5, 132.5, 129.6, 129.4, 122.8, 117.2, 115.5, 112.5, 91.3; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{14}H_{11}IN_3O$: 363.9947; found: 363.9950.

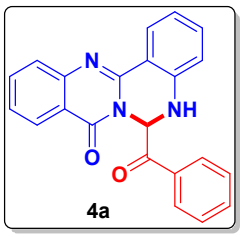
2-(2-Amino-5-chlorophenyl)quinazolin-4(3*H*)-one (2e)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Pale yellow solid; Yield: 703 mg, 78%; m.p.: 301-302 °C; FT-IR (KBr) ν (cm^{-1}) 3463, 3331, 3143, 3093,

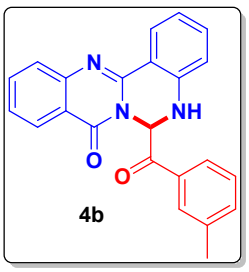
3017, 1677, 1611, 1558, 1502, 1471, 1413, 1337, 1265, 1115, 872, 768; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.22 (s, 1H), 8.14 (dd, J = 8.0, 1.5 Hz, 1H), 7.87 – 7.79 (m, 2H), 7.75 (dd, J = 8.2, 1.2 Hz, 1H), 7.51 (ddd, J = 8.1, 7.1, 1.3 Hz, 1H), 7.23 (dd, J = 8.8, 2.4 Hz, 1H), 7.19 (s, 2H), 6.86 (d, J = 8.9 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 161.9, 152.5, 148.2, 147.8, 134.6, 131.5, 128.1, 127.0, 126.5, 125.7, 120.6, 118.3, 118.2, 113.2; HRMS (ESI) (m/z) [M + H]$^{+}$ calcd. for $C_{14}H_{11}ClN_3O$: 272.0591; found: 272.0594.

6-Benzoyl-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4a)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 136 mg, 91%; m.p.: 213-214 °C; FT-IR (KBr) ν (cm^{-1}) 3391, 3061, 3005, 2956, 2852, 1743, 1663, 1591, 1553, 1467, 1382, 1272, 1227, 1154; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.19 (dd, J = 8.0, 1.6 Hz, 1H), 8.17 – 8.13 (m, 2H), 8.10 (dd, J = 7.9, 1.0 Hz, 1H), 7.87 (ddd, J = 8.6, 7.1, 1.6 Hz, 1H), 7.78 – 7.71 (m, 3H), 7.61 (t, J = 7.7 Hz, 2H), 7.58 (d, J = 5.2 Hz, 1H), 7.50 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.28 (ddd, J = 8.2, 7.2, 1.6 Hz, 1H), 6.87 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.79 (dd, J = 8.1, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.9, 160.1, 147.9, 142.9, 135.1, 134.3, 133.5, 132.8, 129.2, 129.1 (2C), 127.3, 126.8, 126.4, 126.3, 119.8, 119.5, 116.1, 115.6, 63.2; HRMS (ESI) (m/z) [M + H]$^{+}$ calcd. for $C_{22}H_{16}N_3O_2$: 354.1243; found: 354.1245.

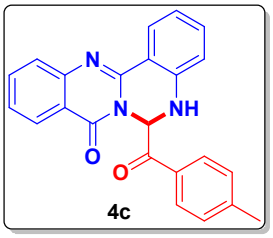
6-(3-Methylbenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4b)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); White solid; Yield: 122 mg, 78%; m.p.: 183-184 °C; FT-IR (KBr) ν (cm^{-1}) 3359, 3054, 2953, 2918, 2857, 1746, 1664, 1599, 1556, 1459, 1382, 1250, 1155; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.18 (dd, J = 8.0, 1.6 Hz, 1H), 8.10 (dd, J = 8.1, 1.5 Hz, 1H), 7.97 – 7.93 (m, 2H), 7.87 (ddd, J = 8.4, 7.1, 1.6 Hz, 1H), 7.79 – 7.71 (m, 2H), 7.57 – 7.53 (m, 2H), 7.53 – 7.47 (m, 2H), 7.28 (ddd, J = 8.4, 7.1, 1.6 Hz, 1H), 6.86 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.79 (dd, J = 8.2, 1.1 Hz, 1H),

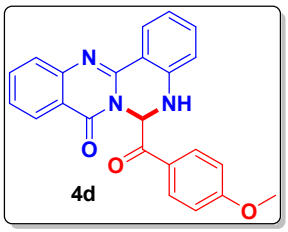
2.42 (s, 3H); $^{13}C\{^{1}H\}$NMR (100 MHz, DMSO-d_6) δ 191.9, 160.1, 147.9, 142.9, 141.3, 138.6, 135.1, 134.9, 133.4, 132.8, 129.5, 129.0, 128.5, 127.3, 126.8, 126.4, 126.3, 119.8, 119.4, 116.0, 115.6, 63.1, 20.9; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{18}N_3O_2$: 368.1399; found: 368.1397.

6-(4-Methylbenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4c)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 144 mg, 93%; m.p.: 199-200 °C; FT-IR (KBr) ν (cm^{-1}) 3372, 3057, 3009, 2956, 2845, 1743, 1664, 1598, 1555, 1441, 1390, 1276, 1182; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.18 (dd, J = 8.0, 1.6 Hz, 1H), 8.10 (dd, J = 8.0, 1.6 Hz, 1H), 8.04 (d, J = 8.1 Hz, 2H), 7.86 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.75 (d, J = 8.1 Hz, 1H), 7.71 (d, J = 5.2 Hz, 1H), 7.53 (d, J = 5.3 Hz, 1H), 7.54 – 7.45 (m, 1H), 7.41 (d, J = 7.9 Hz, 2H), 7.27 (ddd, J = 8.4, 7.2, 1.6 Hz, 1H), 6.86 (t, J = 7.5 Hz, 1H), 6.77 (d, J = 8.1 Hz, 1H), 2.42 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 191.5, 160.2, 148.0, 147.9, 145.1, 143.0, 135.2, 133.5, 130.2, 129.7, 129.4, 127.3, 126.9, 126.4, 126.3, 119.9, 119.5, 116.1, 115.7, 63.2, 21.4: HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{18}N_3O_2$: 368.1399; found: 368.1399.

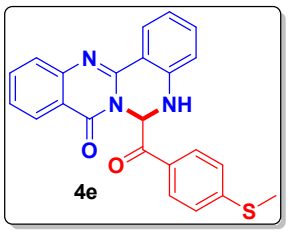
6-(4-Methoxybenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4d)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 139 mg, 86%; m.p.: 209-210 °C; FT-IR (KBr) ν (cm^{-1}) 3313, 3014, 2893, 2830, 1741, 1660, 1592, 1559, 1459, 1392, 1321, 1232, 1163, 1015; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.18 (dd, J = 8.0, 1.5 Hz, 1H), 8.13 (d, J = 8.9 Hz, 2H), 8.10 (dd, J = 8.1, 1.5 Hz, 1H), 7.86 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.75 (dd, J = 8.2, 1.2 Hz, 1H), 7.69 (d, J = 5.2 Hz, 1H), 7.54 – 7.46 (m, 2H), 7.27 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.12 (d, J = 8.9 Hz, 2H), 6.86 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H), 3.88 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 190.3, 164.0, 160.1, 148.1, 147.9, 143.0, 135.1, 133.4, 131.7, 131.4, 127.3, 126.9, 126.3,

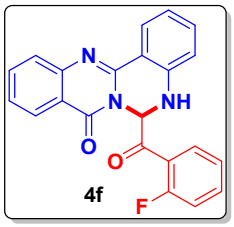
125.4, 119.9, 119.4, 116.1, 115.7, 114.4, 63.0, 55.8; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{18}N_3O_3$: 384.1348; found: 384.1352.

6-(4-(Methylthio)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4e)



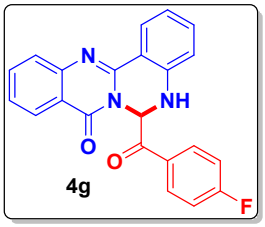
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 150 mg, 89%; m.p.: 245-246 °C; FT-IR (KBr) ν (cm^{-1}) 3356, 3012, 2921, 2844, 1661, 1585, 1554, 1440, 1319, 1233, 1156, 1092, 965; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.18 (dd, J = 8.0, 1.6 Hz, 1H), 8.10 (dd, J = 8.1, 1.5 Hz, 1H), 8.07 (d, J = 8.6 Hz, 2H), 7.87 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.75 (dd, J = 8.3, 1.2 Hz, 1H), 7.71 (d, J = 5.2 Hz, 1H), 7.55 – 7.47 (m, 2H), 7.43 (d, J = 8.7 Hz, 2H), 7.28 (ddd, J = 8.4, 7.2, 1.6 Hz, 1H), 6.86 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.79 (dd, J = 8.2, 1.2 Hz, 1H), 2.57 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 190.9, 160.1, 148.0, 147.9, 147.2, 143.0, 135.1, 133.5, 129.6, 128.7, 127.3, 126.8, 126.4, 126.3, 125.1, 119.9, 119.4, 116.1, 115.7, 63.1, 13.9; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{18}N_3O_2S$: 400.1120; found: 400.1119.

6-(2-Fluorobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4f)



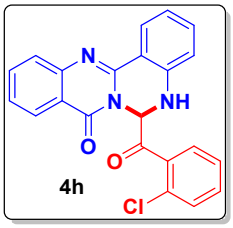
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 116 mg, 74%; m.p.: 242-243 °C; FT-IR (KBr) ν (cm^{-1}) 3373, 3060, 2959, 2922, 1713, 1670, 1600, 1562, 1442, 1323, 1181, 1155, 1013; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.16 (dd, J = 20.6, 8.0 Hz, 2H), 7.94 – 7.83 (m, 2H), 7.79 – 7.74 (m, 3H), 7.54 – 7.74 (m, 2H), 7.37 (t, J = 7.6 Hz, 1H), 7.32 – 7.23 (m, 2H), 6.90 (t, J = 7.6 Hz, 1H), 6.78 (d, J = 8.2 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.2 (d, J = 6 Hz), 161.0 (d, J = 252 Hz), 160.2, 147.9, 147.6, 142.9, 136.3 (d, J = 9 Hz), 135.2, 133.6, 130.9, 127.4, 126.9, 126.5 (d, J = 5 Hz), 125.4 (d, J = 2 Hz), 122.0 (d, J = 4 Hz), 119.8 (2C), 117.1, 116.9, 116.4, 115.9, 65.7 (d, J = 8 Hz); ^{19}F NMR (376 MHz, DMSO-d_6) δ -109.45; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{15}FN_3O_2$: 372.1148; found: 372.1148.

6-(4-Fluorobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4g)



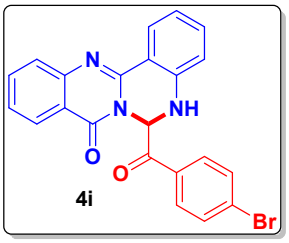
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 136 mg, 87%; m.p.: 220-221 °C; FT-IR (KBr) ν (cm^{-1}) 3275, 3060, 2999, 2930, 1697, 1656, 1602, 1565, 1479, 1379, 1228, 1160, 1006; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.28 – 8.23 (m, 2H), 8.18 (dd, J = 8.0, 1.5 Hz, 1H), 8.10 (dd, J = 8.0, 1.5 Hz, 1H), 7.87 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.78 – 7.72 (m, 2H), 7.58 (d, J = 4.9 Hz, 1H), 7.50 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.44 (t, J = 8.8 Hz, 2H), 7.28 (ddd, J = 8.5, 7.2, 1.6 Hz, 1H), 6.91 – 6.84 (m, 1H), 6.79 (d, J = 8.1 Hz, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 190.7, 165.7 (d, J = 250 Hz), 160.1, 147.9, 147.8, 142.9, 135.1, 133.5, 132.4 (d, J = 10 Hz), 129.4 (d, J = 2.4 Hz), 127.3, 126.8, 126.4, 126.3, 119.8, 119.5, 116.3, 116.1, 115.6, 63.2; ^{19}F NMR (376 MHz, DMSO-d_6) δ -103.55; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{15}FN_3O_2$: 372.1148; found: 372.1150.

6-(2-Chlorobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4h)



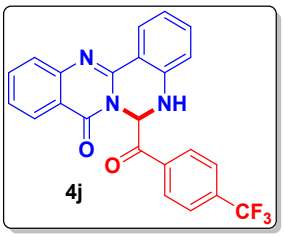
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Pale yellow solid; Yield: 111 mg, 68%; m.p.: 207-208 °C; FT-IR (KBr) ν (cm^{-1}) 3270, 3062, 2996, 2938, 1710, 1659, 1604, 1564, 1438, 1335, 1220, 1113, 1031, 806, 755; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.17 (ddd, J = 9.8, 8.0, 1.4 Hz, 2H), 7.88 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.83 (d, J = 5.0 Hz, 1H), 7.76 (dd, J = 8.2, 1.1 Hz, 1H), 7.61 (dt, J = 7.5, 1.0 Hz, 1H), 7.59 – 7.46 (m, 4H), 7.34 (ddd, J = 8.4, 7.2, 1.5 Hz, 1H), 7.28 (d, J = 4.2 Hz, 1H), 6.91 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.77 (dd, J = 8.2, 1.1 Hz, 1H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 195.8, 159.9, 147.8, 147.4, 143.5, 135.4, 135.1, 133.6, 132.9, 130.6, 130.4, 128.9, 127.4, 127.3, 126.9, 126.5, 126.4, 119.8, 119.7, 116.2, 115.7, 65.6; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{15}ClN_3O_2$: 388.0853; found: 388.0855.

6-(4-Bromobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4i)



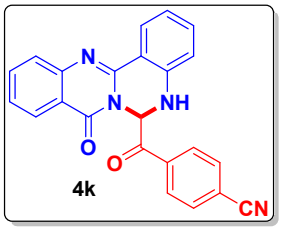
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 168 mg, 92%; m.p.: 214-215 °C; FT-IR (KBr) ν (cm^{-1}) 3386, 3054, 2959, 1745, 1663, 1579, 1558, 1468, 1389, 1319, 1227, 1071, 688, 591; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.18 (dd, J = 8.0, 1.5 Hz, 1H), 8.13 – 8.04 (m, 3H), 7.87 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.82 (d, J = 8.5 Hz, 2H), 7.78 – 7.72 (m, 2H), 7.55 (d, J = 5.1 Hz, 1H), 7.50 (t, J = 7.4 Hz, 1H), 7.28 (ddd, J = 8.4, 7.2, 1.6 Hz, 1H), 6.88 (t, J = 7.5 Hz, 1H), 6.78 (d, J = 8.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.5, 160.2, 147.9, 147.8, 142.8, 135.3, 133.6, 132.3, 131.8, 131.2, 128.7, 127.4, 126.9, 126.5, 126.4, 119.8, 119.7, 116.2, 115.7, 63.3; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{15}BrN_3O_2$: 432.0348; found: 432.0344.

6-(4-(Trifluoromethyl)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4j)



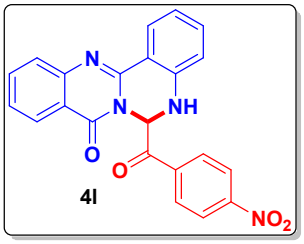
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 140 mg, 79%; m.p.: 237-238 °C; FT-IR (KBr) ν (cm^{-1}) 3385, 3062, 3028, 2962, 1746, 1662, 1595, 1557, 1435, 1385, 1315, 1227, 1134, 1065; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.34 (d, J = 7.9 Hz, 2H), 8.19 (dd, J = 8.0, 1.6 Hz, 1H), 8.12 (ddd, J = 7.9, 1.6, 0.6 Hz, 1H), 7.98 (d, J = 8.5 Hz, 2H), 7.88 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.82 – 7.73 (m, 2H), 7.64 (d, J = 5.2 Hz, 1H), 7.51 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.29 (ddd, J = 8.1, 7.2, 1.6 Hz, 1H), 6.89 (ddd, J = 8.2, 7.2, 1.1 Hz, 1H), 6.79 (dd, J = 8.3, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.7, 160.1, 147.9, 147.6, 142.7, 136.3, 135.2, 133.5, 133.2 (q, J = 67 Hz), 130.0, 127.3, 126.8, 126.5, 126.3, 126.0 (q, J = 4.0 Hz), 123.7 (q, J = 270 Hz), 119.8, 119.7, 116.2, 115.7, 63.4; ^{19}F NMR (376 MHz, DMSO-d_6) δ -62.42; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{15}F_3N_3O_2$: 422.1116; found: 422.1117.

4-(8-Oxo-5,8-dihydro-6*H*-quinazolino[4,3-*b*]quinazoline-6-carbonyl)benzonitrile (4k)



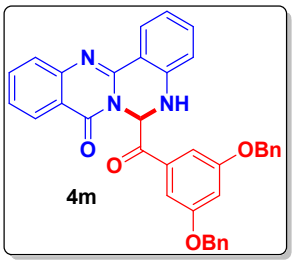
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 123 mg, 77%; m.p.: 223-224 °C; FT-IR (KBr) ν (cm^{-1}) 3398, 3052, 2964, 2232, 1746, 1663, 1594, 1558, 1435, 1381, 1225, 1118; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.30 (d, J = 8.7 Hz, 2H), 8.19 (dd, J = 8.0, 1.5 Hz, 1H), 8.13 – 8.06 (m, 3H), 7.87 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.80 (d, J = 5.3 Hz, 1H), 7.76 (ddd, J = 8.2, 1.2, 0.6 Hz, 1H), 7.64 (d, J = 5.2 Hz, 1H), 7.51 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.29 (ddd, J = 8.2, 7.2, 1.6 Hz, 1H), 6.89 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 192.1, 160.6, 148.3, 148.1, 143.1, 136.7, 135.7, 134.0, 133.5, 130.2, 127.8, 127.3, 126.9, 126.8, 120.3, 120.2, 118.5, 116.7, 116.6, 116.1, 63.9; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{15}N_4O_2$: 379.1195; found: 379.1194.

6-(4-Nitrobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4l)



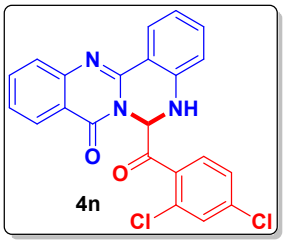
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6:4, v/v); Orange solid; Yield: 128 mg, 76%; m.p.: 214-215 °C; FT-IR (KBr) ν (cm^{-1}) 3328, 3075, 2923, 2853, 1713, 1695, 1650, 1605, 1557, 1521, 1466, 1339, 1219, 1176; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.41 – 8.36 (m, 4H), 8.19 (dd, J = 8.1, 1.6 Hz, 1H), 8.12 (dd, J = 8.0, 1.5 Hz, 1H), 7.88 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.83 (d, J = 5.3 Hz, 1H), 7.77 (dd, J = 8.2, 1.1 Hz, 1H), 7.66 (d, J = 5.2 Hz, 1H), 7.51 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.29 (ddd, J = 8.5, 7.2, 1.6 Hz, 1H), 6.90 (ddd, J = 8.2, 7.2, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.6, 160.2, 150.5, 147.9, 147.6, 142.6, 137.8, 135.3, 133.6, 130.6, 127.4, 126.9, 126.5, 126.4, 124.1, 119.9, 119.8, 116.4, 115.8, 63.6. HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{15}N_4O_4$: 399.1093; found: 399.1092.

6-(3,5-Bis(benzyloxy)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4m)



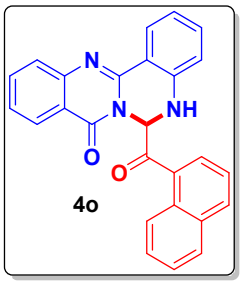
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 182 mg, 76%; m.p.: 179-180 °C; FT-IR (KBr) ν (cm^{-1}) 3323, 3024, 2944, 2835, 1744, 1668, 1593, 1555, 1371, 1212, 1155; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.18 (dd, J = 8.0, 1.5 Hz, 1H), 8.10 (ddd, J = 7.9, 1.6, 0.6 Hz, 1H), 7.87 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.76 (dd, J = 8.2, 1.3 Hz, 1H), 7.70 (d, J = 5.3 Hz, 1H), 7.55 (d, J = 5.3 Hz, 1H), 7.53 – 7.45 (m, 5H), 7.43 – 7.38 (m, 4H), 7.37 – 7.27 (m, 5H), 7.04 (t, J = 2.2 Hz, 1H), 6.87 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.82 (dd, J = 8.2, 1.1 Hz, 1H), 5.19 (d, J = 1.9 Hz, 4H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.4, 160.1, 159.9, 147.9, 142.9, 136.6, 135.2, 134.6, 133.5, 128.5, 128.0, 127.9, 127.3, 126.8, 126.4, 126.3, 119.9, 119.5, 116.2, 115.6, 108.0 (2C), 107.7, 69.8, 63.2. HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{36}H_{28}N_3O_4$: 566.2080; found: 566.2081.

6-(2,4-Dichlorobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4n)



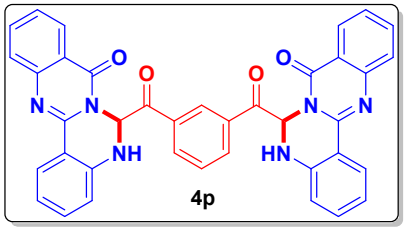
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); White solid; Yield: 111 mg, 62%; m.p.: 180-181 °C; FT-IR (KBr) ν (cm^{-1}) 3382, 3061, 2948, 1703, 1664, 1589, 1562, 1469, 1324, 1220, 1109, 1068, 825, 691; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.20 – 8.12 (m, 2H), 7.88 (ddd, J = 8.6, 7.1, 1.6 Hz, 1H), 7.81 (d, J = 4.8 Hz, 1H), 7.79 – 7.72 (m, 2H), 7.68 (d, J = 8.3 Hz, 1H), 7.61 (dd, J = 8.3, 2.0 Hz, 1H), 7.53 (ddd, J = 8.2, 7.1, 1.2 Hz, 1H), 7.34 (ddd, J = 8.3, 7.2, 1.6 Hz, 1H), 7.28 (d, J = 4.8 Hz, 1H), 6.92 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.77 (dd, J = 8.4, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 195.2, 159.9, 147.8, 147.3, 143.4, 136.9, 135.2, 134.2, 133.7, 132.0, 130.4, 130.1, 127.7, 127.4, 126.9, 126.5, 126.4, 119.8, 119.7, 116.2, 115.7, 65.6; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{14}Cl_2N_3O_2$: 422.0463; found: 422.0466.

6-(1-Naphthoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4o)



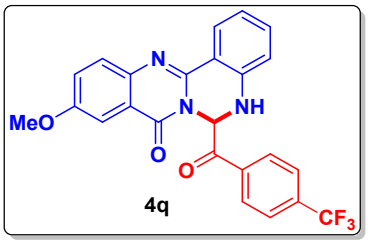
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 140 mg, 82%; m.p.: 206-207 °C; FT-IR (KBr) ν (cm^{-1}) 3387, 3062, 3012, 2959, 1746, 1666, 1592, 1556, 1469, 1385, 1240, 1180, 1101; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.28 (dd, J = 7.3, 1.2 Hz, 1H), 8.25 – 8.19 (m, 2H), 8.17 (ddd, J = 7.9, 1.6, 0.6 Hz, 1H), 8.03 (d, J = 8.1 Hz, 1H), 7.91 – 7.83 (m, 2H), 7.80 – 7.75 (m, 2H), 7.73 (dd, J = 8.3, 7.2 Hz, 1H), 7.58 – 7.52 (m, 2H), 7.51 – 7.45 (m, 2H), 7.27 (ddd, J = 8.4, 7.2, 1.6 Hz, 1H), 6.90 (ddd, J = 8.2, 7.2, 1.1 Hz, 1H), 6.68 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H}NMR (100 MHz, DMSO-d_6) δ 196.1, 160.2, 147.9, 147.8, 143.3, 135.2, 133.6, 133.4, 133.0, 132.2, 130.2, 128.7, 128.1, 127.9, 127.3, 126.9, 126.7, 126.5, 126.4, 125.0, 124.4, 119.8, 119.5, 115.8, 115.6, 65.3; HRMS (ESI) (m/z) [M + H]$^+$ calcd. for $C_{26}H_{18}N_3O_2$: 404.1399; found: 404.1398.

6,6'-Isophthaloylbis(5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one) (4p)



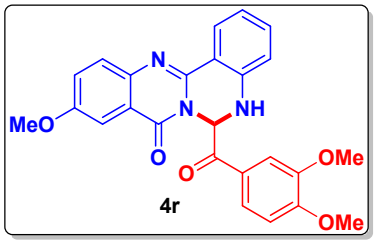
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 169 mg, 62%; m.p.: 210-211 °C; FT-IR (KBr) ν (cm^{-1}) 3352, 3068, 2925, 1737, 1677, 1598, 1563, 1464, 1325, 1233, 1160, 1012; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.84 (s, 1H), 8.44 (dd, J = 7.9, 1.7 Hz, 2H), 8.21 (dd, J = 8.0, 1.5 Hz, 2H), 8.13 (dd, J = 8.0, 1.5 Hz, 2H), 7.95 – 7.85 (m, 4H), 7.83 (t, J = 7.8 Hz, 1H), 7.79 – 7.72 (m, 4H), 7.51 (t, J = 7.4 Hz, 2H), 7.30 (td, J = 7.8, 1.6 Hz, 2H), 6.89 (t, J = 7.5 Hz, 2H), 6.80 (d, J = 8.1 Hz, 2H); ^{13}C{^{1}H}NMR (100 MHz, DMSO- d_6) δ 192.1, 160.6, 148.3, 148.2, 143.3, 135.6, 134.9, 134.0, 133.9, 130.6, 129.8, 127.8, 127.4, 126.9, 126.8, 120.3, 120.1, 116.7, 116.2, 63.9; HRMS (ESI) (m/z) [M + H]$^+$ calcd. for $C_{38}H_{25}N_6O_4$: 629.1937; found: 629.1944.

10-Methoxy-6-(4-(trifluoromethyl)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4q)



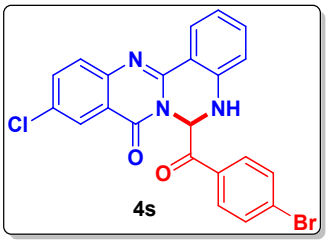
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 136 mg, 86%; m.p.: 215-216 °C; FT-IR (KBr) ν (cm^{-1}) 3327, 3108, 3047, 2943, 2838, 1655, 1614, 1567, 1489, 1364, 1323, 1224, 1066, 839; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.34 (d, J = 8.0 Hz, 2H), 8.16 (dd, J = 8.0, 1.5 Hz, 1H), 7.98 (d, J = 8.1 Hz, 2H), 7.75 – 7.69 (m, 2H), 7.63 (d, J = 5.1 Hz, 1H), 7.52 – 7.46 (m, 2H), 7.26 (ddd, J = 8.2, 7.2, 1.5 Hz, 1H), 6.88 (ddd, J = 8.2, 7.2, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H), 3.88 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 191.8, 159.9, 157.6, 145.6, 142.4, 142.3, 136.3, 133.3 (q, J = 32 Hz), 133.0, 129.9, 129.1, 126.5, 126.0 (q, J = 4 Hz), 124.7, 123.7 (q, J = 271 Hz), 120.6, 119.7, 116.2, 115.9, 106.3, 63.6, 55.7; ^{19}F NMR (376 MHz, DMSO- d_6) δ -61.75; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{24}H_{17}F_3N_3O_3$: 452.1222; found: 452.1227.

6-(3,4-Dimethoxybenzoyl)-10-methoxy-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4r)



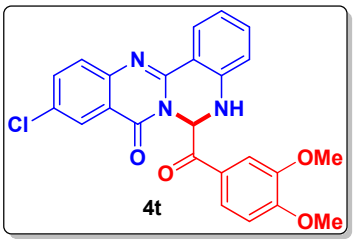
Purified by column chromatography (eluent: petroleum ether/EtOAc = 6:4, v/v); White solid; Yield: 148 mg, 89%; m.p.: 215-216 °C; FT-IR (KBr) ν (cm^{-1}) 3322, 3111, 3073, 2937, 2835, 2822, 1680, 1643, 1612, 1565, 1443, 1341, 1269, 1126, 1025; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.15 (dd, J = 8.0, 1.5 Hz, 1H), 7.93 (dd, J = 8.5, 2.1 Hz, 1H), 7.74 – 7.69 (m, 1H), 7.61 (d, J = 5.1 Hz, 1H), 7.54 (d, J = 5.1 Hz, 1H), 7.52 (d, J = 2.0 Hz, 1H), 7.50 – 7.45 (m, 2H), 7.25 (ddd, J = 8.5, 7.2, 1.6 Hz, 1H), 7.15 (d, J = 8.5 Hz, 1H), 6.85 (td, J = 7.6, 1.1 Hz, 1H), 6.80 (d, J = 8.2 Hz, 1H), 3.90 (s, 3H), 3.87 (s, 3H), 3.83 (s, 3H); $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) δ 190.3, 159.8, 157.5, 154.0, 148.9, 146.0, 142.6, 142.4, 132.9, 129.0, 126.4, 125.4, 124.6, 124.3, 120.6, 119.3, 116.0 (2C), 111.1 (2C), 106.3, 63.0, 55.9, 55.7, 55.6; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{25}H_{22}N_3O_5$: 444.1559; found: 444.1555.

6-(4-Bromobenzoyl)-10-chloro-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4s)



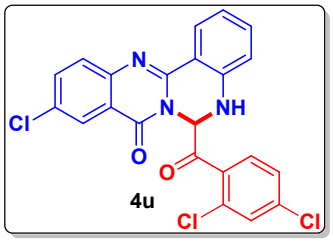
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 142 mg, 83%; m.p.: 210-211 °C; FT-IR (KBr) ν (cm^{-1}) 3341, 3040, 2926, 1670, 1564, 1510, 1460, 1392, 1327, 1229, 1115, 1008, 833, 704, 686, 524; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.16 (dd, J = 8.1, 1.5 Hz, 1H), 8.08 (d, J = 8.6 Hz, 2H), 8.04 (d, J = 2.5 Hz, 1H), 7.89 (dd, J = 8.8, 2.5 Hz, 1H), 7.82 (d, J = 8.6 Hz, 2H), 7.80 – 7.75 (m, 2H), 7.57 (d, J = 5.2 Hz, 1H), 7.29 (ddd, J = 8.4, 7.2, 1.6 Hz, 1H), 6.88 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.79 (d, J = 8.2 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.2, 159.1, 148.2, 146.6, 142.9, 135.3, 133.8, 132.2, 131.6, 131.2, 130.5, 129.5, 128.7, 126.9, 125.2, 121.0, 119.6, 116.2, 115.3, 63.4; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{14}BrClN_3O_2$: 465.9958; found: 465.9958.

10-Chloro-6-(3,4-dimethoxybenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4t)



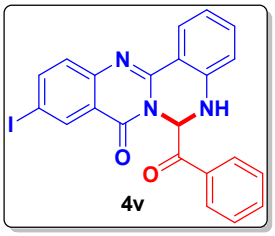
Purified by column chromatography (eluent: petroleum ether/EtOAc =6.5:3.5, v/v); White solid; Yield: 144 mg, 87%; m.p.: 214-215 °C; FT-IR (KBr) ν (cm^{-1}) 3316, 3073, 299, 2936, 2834, 2811, 1654, 1593, 1560, 1462, 1383, 1340, 1267, 1129, 1066, 758, 682; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.16 (dd, J = 8.0, 1.5 Hz, 1H), 8.03 (d, J = 2.5 Hz, 1H), 7.92 (dd, J = 8.5, 2.0 Hz, 1H), 7.88 (dd, J = 8.8, 2.6 Hz, 1H), 7.77 (d, J = 8.8 Hz, 1H), 7.73 (d, J = 5.2 Hz, 1H), 7.56 (d, J = 5.1 Hz, 1H), 7.52 (d, J = 2.0 Hz, 1H), 7.29 (ddd, J = 8.5, 7.2, 1.6 Hz, 1H), 7.15 (d, J = 8.5 Hz, 1H), 6.86 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.81 (d, J = 7.8 Hz, 1H), 3.90 (s, 3H), 3.83 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 190.1, 159.1, 154.1, 148.9, 148.5, 146.7, 143.1, 135.2, 133.7, 130.4, 129.5, 126.9, 125.2, 125.2, 124.3, 121.0, 119.3, 116.0, 115.3, 111.2, 111.1, 63.1, 55.9, 55.6; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{24}H_{19}ClN_3O_4$: 448.1064; found: 448.1065.

10-Chloro-6-(2,4-dichlorobenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4u)



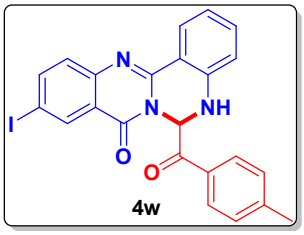
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 109 mg, 61%; m.p.: 217-218 °C; FT-IR (KBr) ν (cm^{-1}) 3284, 3092, 3063, 2924, 1710, 1659, 1576, 1468, 1372, 1355, 1219, 1112, 1010, 827, 759; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.16 (dd, J = 8.0, 1.5 Hz, 1H), 8.10 (d, J = 2.5 Hz, 1H), 7.91 (dd, J = 8.7, 2.5 Hz, 1H), 7.87 (d, J = 4.9 Hz, 1H), 7.78 (dd, J = 5.3, 3.4 Hz, 2H), 7.70 (d, J = 8.4 Hz, 1H), 7.62 (dd, J = 8.3, 2.0 Hz, 1H), 7.36 (ddd, J = 8.2, 7.2, 1.6 Hz, 1H), 7.30 (d, J = 4.8 Hz, 1H), 6.92 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 195.5, 159.4, 148.2, 147.1, 144.0, 137.4, 135.8, 134.5, 134.4, 132.4, 131.1, 130.9, 130.5, 130.1, 128.1, 127.4, 125.9, 121.4, 120.3, 116.7, 115.9, 66.2; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{13}Cl_3N_3O_2$: 456.0073; found: 456.0073.

6-Benzoyl-10-iodo-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4v)



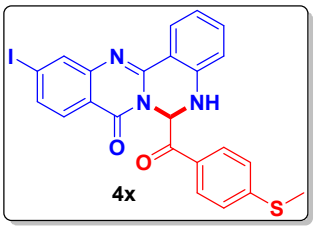
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 119 mg, 80%; m.p.: 173-174 °C; FT-IR (KBr) ν (cm^{-1}) 3316, 3065, 2954, 1702, 1681, 1601, 1580, 1457, 1393, 1320, 1244, 1070, 502; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.36 (d, J = 2.1 Hz, 1H), 8.19 – 8.11 (m, 4H), 7.79 (d, J = 5.3 Hz, 1H), 7.76 – 7.72 (m, 1H), 7.64 – 7.52 (m, 4H), 7.28 (ddd, J = 8.5, 7.2, 1.6 Hz, 1H), 6.86 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.8, 158.9, 148.5, 147.2, 143.5, 143.0, 134.5, 134.4, 133.8, 132.6, 129.5, 129.2, 129.1, 126.9, 121.7, 119.5, 116.2, 115.4, 91.1, 63.4; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{15}IN_3O_2$: 480.0209; found: 480.0207.

10-Iodo-6-(4-methylbenzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4w)



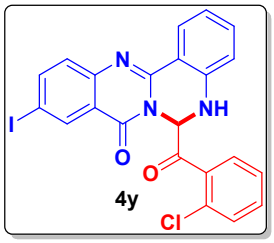
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 118 mg, 87%; m.p.: 256-257 °C; FT-IR (KBr) ν (cm^{-1}) 3306, 3115, 3063, 2964, 2835, 1679, 1646, 1605, 1556, 1458, 1334, 1235, 1161, 743, 501; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.36 (d, J = 2.1 Hz, 1H), 8.14 (ddd, J = 8.5, 6.3, 1.8 Hz, 2H), 8.04 (d, J = 8.3 Hz, 2H), 7.76 (d, J = 5.3 Hz, 1H), 7.57 – 7.50 (m, 2H), 7.41 (d, J = 8.0 Hz, 2H), 7.28 (ddd, J = 8.5, 7.2, 1.6 Hz, 1H), 6.85 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.77 (dd, J = 8.3, 1.1 Hz, 1H), 2.42 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.7, 159.3, 149.0, 147.7, 145.5, 143.9, 143.5, 135.0, 134.2, 130.5, 130.1, 129.9, 129.8, 127.4, 122.1, 119.9, 116.6, 115.8, 91.4, 63.8, 21.8; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{17}IN_3O_2$: 494.0365; found: 494.0365.

11-Iodo-6-(4-(methylthio)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4x)



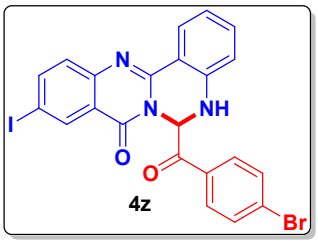
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 122 mg, 84%; m.p.: 216-217 °C; FT-IR (KBr) ν (cm^{-1}) 3356, 3142, 3012, 2921, 2844, 1668, 1585, 1554, 1443, 1324, 1239, 1158, 1064, 958, 511; ^{1}H NMR (400 MHz, DMSO-d_6) δ δ 8.36 (d, J = 2.1 Hz, 1H), 8.18 – 8.10 (m, 2H), 8.06 (d, J = 8.2 Hz, 2H), 7.75 (d, J = 5.2 Hz, 1H), 7.55 – 7.51 (m, 2H), 7.42 (d, J = 8.2 Hz, 2H), 7.28 (t, J = 7.5 Hz, 1H), 6.86 (t, J = 7.6 Hz, 1H), 6.78 (d, J = 8.2 Hz, 1H), 2.57 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.2, 159.3, 149.0, 147.8, 147.7, 143.9, 143.5, 135.0, 134.2, 130.1, 129.9, 129.0, 127.4, 125.5, 122.1, 119.9, 116.6, 115.9, 91.5, 63.7, 14.4; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{17}IN_3O_2S$: 526.0086; found: 526.0086.

6-(2-Chlorobenzoyl)-10-iodo-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4y)



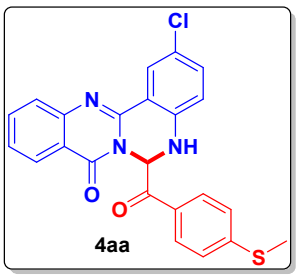
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 102 mg, 72%; m.p.: 188-189 °C; FT-IR (KBr) ν (cm^{-1}) 3328, 3017, 2928, 1665, 1584, 1558, 1442, 1374, 1263, 1212, 1155, 735, 648, 523; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.41 (d, J = 2.1 Hz, 1H), 8.17 – 8.13 (m, 2H), 7.87 (d, J = 5.0 Hz, 1H), 7.61 – 7.47 (m, 5H), 7.34 (ddd, J = 8.4, 7.1, 1.6 Hz, 1H), 7.27 (d, J = 5.0 Hz, 1H), 6.91 (ddd, J = 8.2, 7.2, 1.1 Hz, 1H), 6.76 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 195.8, 158.7, 148.0, 147.2, 143.6, 143.5, 135.3, 134.6, 133.9, 132.9, 130.6, 130.4, 129.5, 129.0, 127.4, 127.0, 121.6, 119.8, 116.2, 115.4, 91.2, 65.7; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{14}ClIN_3O_2$: 513.9819; found: 513.9818.

6-(4-Bromobenzoyl)-10-iodo-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4z)



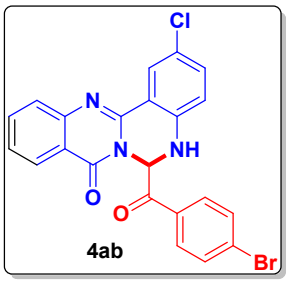
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 136 mg, 88%; m.p.: 221-222 °C; FT-IR (KBr) ν (cm^{-1}) 3338, 3023, 2919, 1667, 1583, 1567, 1453, 1380, 1267, 1228, 1155, 747, 667, 544; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.36 (d, J = 2.1 Hz, 1H), 8.17 – 8.12 (m, 2H), 8.07 (d, J = 8.6 Hz, 2H), 7.82 (d, J = 8.6 Hz, 2H), 7.79 (d, J = 5.3 Hz, 1H), 7.59 – 7.52 (m, 2H), 7.29 (ddd, J = 8.6, 7.3, 1.6 Hz, 1H), 6.87 (ddd, J = 8.1, 7.2, 1.1 Hz, 1H), 6.78 (dd, J = 8.2, 1.1 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.4, 158.9, 148.4, 147.3, 143.6, 142.9, 134.6, 133.9, 132.3, 131.7, 131.2, 129.6, 128.8, 127.0, 121.7, 119.8, 116.3, 115.5, 91.2, 63.5; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{14}BrIN_3O_2$: 557.9314; found: 557.9316.

2-Chloro-6-(4-(methylthio)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4aa)



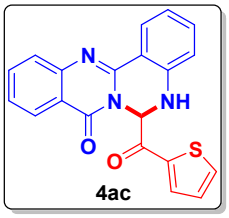
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 114 mg, 71%; m.p.: 250-251 °C; FT-IR (KBr) ν (cm^{-1}) 3365, 2962, 2924, 2844, 1664, 1589, 1561, 1496, 1418, 1321, 1238, 1178, 1095, 874, 777; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.12 – 8.09 (m, 2H), 8.07 (d, J = 8.6 Hz, 2H), 7.93 – 7.86 (m, 2H), 7.79 (d, J = 8.0 Hz, 1H), 7.57 (d, J = 5.0 Hz, 1H), 7.53 (ddd, J = 8.2, 7.0, 1.2 Hz, 1H), 7.43 (d, J = 8.6 Hz, 2H), 7.31 (dd, J = 8.6, 2.6 Hz, 1H), 6.83 (d, J = 8.7 Hz, 1H), 2.57 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 190.7, 159.9, 147.6, 147.4, 146.8, 141.9, 135.3, 133.1, 129.7, 128.5, 127.4, 126.8, 126.4, 125.6, 125.1, 123.0, 120.0, 118.0, 116.8, 63.0, 13.9; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{17}ClN_3O_2S$: 434.0730; found: 434.0730.

6-(4-Bromobenzoyl)-2-chloro-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ab)



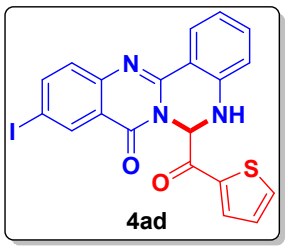
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 150 mg, 87%; m.p.: 202-203 °C; FT-IR (KBr) ν (cm^{-1}) 3377, 3079, 3063, 2959, 1668, 1587, 1563, 1494, 1421, 1323, 1276, 1234, 1152, 1010, 775, 697; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.14 – 8.06 (m, 4H), 7.95 (d, J = 5.1 Hz, 1H), 7.89 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.83 (d, J = 8.6 Hz, 2H), 7.80 (d, J = 7.6 Hz, 1H), 7.61 (d, J = 5.0 Hz, 1H), 7.53 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.32 (dd, J = 8.7, 2.6 Hz, 1H), 6.83 (d, J = 8.7 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 191.2, 159.9, 147.6, 146.6, 141.7, 135.3, 133.2, 132.2, 131.6, 131.2, 128.8, 127.5, 126.8, 126.4, 125.6, 123.2, 119.9, 118.1, 116.8, 63.1; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{14}BrClN_3O_2$: 465.9958; found: 465.9960.

6-(Thiophene-2-carbonyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ac)



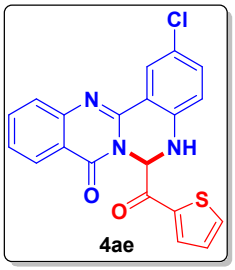
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 123 mg, 81%; m.p.: 239-240 °C; FT-IR (KBr) ν (cm^{-1}) 3370, 3090, 3011, 2972, 1746, 1659, 1597, 1557, 1469, 1390, 1274, 1181, 1067, 956; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.45 (s, 1H), 8.21 – 8.08 (m, 3H), 7.91 – 7.82 (m, 2H), 7.75 (d, J = 7.6 Hz, 1H), 7.56 – 7.46 (m, 1H) ,7.42 – 7.37 (m, 2H), 7.30 (t, J = 7.2 Hz, 1H), 6.87 (t, J = 8.4 Hz, 2H); ^{13}C{^{1}H}NMR (100 MHz, DMSO-d_6) δ 185.8, 160.0, 147.9, 147.7, 143.0, 138.9, 137.0, 135.8, 135.2, 133.5, 129.3, 127.3, 126.9, 126.4, 126.3, 119.8, 119.5, 116.1, 115.7, 64.0; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{20}H_{14}N_3O_2S$: 360.0807; found: 360.0807.

10-Iodo-6-(thiophene-2-carbonyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ad)



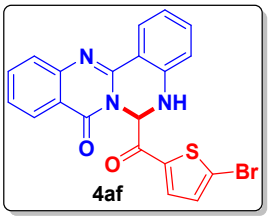
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 107 mg, 80%; m.p.: 246-247 °C; FT-IR (KBr) ν (cm^{-1}) 3325, 3084, 2949, 2923, 1656, 1597, 1566, 1458, 1379, 1236, 1106, 801, 736, 504; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.46 (dd, J = 3.9, 1.1 Hz, 1H), 8.38 (d, J = 2.1 Hz, 1H), 8.20 – 8.11 (m, 3H), 7.89 (d, J = 5.2 Hz, 1H), 7.55 (d, J = 8.6 Hz, 1H), 7.44 – 7.37 (m, 2H), 7.38 – 7.29 (m, 1H), 6.90 – 6.87 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 185.7, 158.7, 148.3, 147.2, 143.5, 143.1, 138.8, 137.1, 135.9, 134.5, 133.8, 129.5, 129.3, 126.9, 121.6, 119.6, 116.1, 115.4, 91.1, 64.2; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{20}H_{13}IN_3O_2S$: 485.9773; found: 485.9779.

2-Chloro-6-(thiophene-2-carbonyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ae)



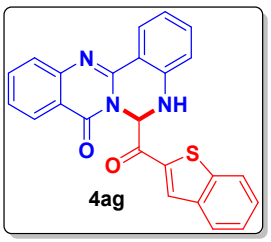
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 113 mg, 78%; m.p.: 272-273 °C; FT-IR (KBr) ν (cm^{-1}) 3307, 3094, 2992, 2923, 1675, 1642, 1595, 1560, 1470, 1406, 1324, 1234, 1151, 859, 736; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.47 (dd, J = 3.9, 1.2 Hz, 1H), 8.17 (dd, J = 5.0, 1.1 Hz, 1H), 8.11 (dd, J = 7.2, 2.0 Hz, 2H), 8.03 (d, J= 4.9 Hz, 1H), 7.88 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.79 (d, J = 8.0 Hz, 1H), 7.52 (ddd, J = 8.1, 7.1, 1.2 Hz, 1H), 7.44 (d, J = 4.9 Hz, 1H), 7.40 (dd, J = 5.0, 3.8 Hz, 1H), 7.34 (dd, J = 8.7, 2.6 Hz, 1H), 6.91 (d, J = 8.7 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 185.5, 159.9, 147.5, 146.5, 142.0, 138.8, 137.2, 136.0, 135.3, 133.2, 129.3, 127.4, 126.8, 126.4, 125.6, 123.2, 120.0, 117.9, 116.8, 63.8; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{20}H_{13}ClN_3O_2S$: 394.0417; found: 394.0419.

6-(5-Bromothiophene-2-carbonyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4af)



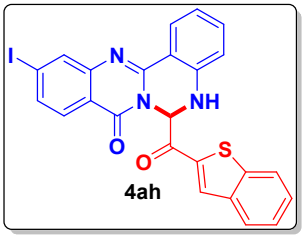
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 156 mg, 84%; m.p.: 234-235 °C; FT-IR (KBr) ν (cm^{-1}) 3360, 3074, 3022, 2970, 1652, 1598, 1557, 1488, 1409, 1384, 1233, 1154, 1080, 983, 745, 688; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.27 (d, J = 4.2 Hz, 1H), 8.18 (dd, J = 8.1, 1.5 Hz, 1H), 8.11 (dd, J = 8.0, 1.5 Hz, 1H), 7.89 – 7.81 (m, 2H), 7.75 (d, J = 8.2 Hz, 1H), 7.56 (d, J = 4.2 Hz, 1H), 7.50 (t, J = 7.5 Hz, 1H), 7.38 (d, J = 5.1 Hz, 1H), 7.33 (t, J = 7.5 Hz, 1H), 6.92 – 6.88 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 185.1, 160.0, 147.8, 147.5, 142.9, 140.3, 136.7, 135.2, 133.6, 132.8, 127.3, 126.9, 126.4, 126.4, 124.0, 119.8 (2C), 116.3, 115.8, 63.8; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{20}H_{13}BrN_3O_2S$: 437.9912; found: 437.9910.

6-(Benzo[*b*]thiophene-2-carbonyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ag)



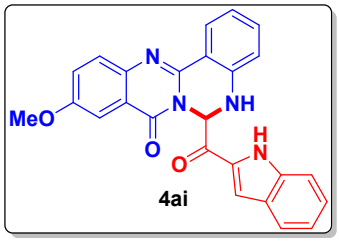
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 152 mg, 88%; m.p.: 235-236 °C; FT-IR (KBr) ν (cm^{-1}) 3392, 3017, 2970, 1746, 1651, 1593, 1555, 1470, 1321, 1248, 1067, 950; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.92 (s, 1H), 8.20 (dd, J = 7.9, 1.5 Hz, 1H), 8.16 – 8.07 (m, 3H), 7.96 (d, J = 4.8 Hz, 1H), 7.88 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.77 (dd, J = 8.2, 1.2 Hz, 1H), 7.63 – 7.48 (m, 4H), 7.31 (ddd, J = 8.6, 7.3, 1.6 Hz, 1H), 6.93 – 6.84 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 187.4, 160.1, 147.8, 147.6, 143.0, 142.0, 139.1, 138.2, 135.2, 133.6 (2C), 128.5, 127.3, 126.9, 126.7, 126.5, 126.4, 125.7, 123.3, 119.8, 119.6, 115.9, 115.5, 63.9; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{24}H_{16}N_3O_2S$: 410.0963; found: 410.0964.

6-(Benzo[*b*]thiophene-2-carbonyl)-11-iodo-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ah)



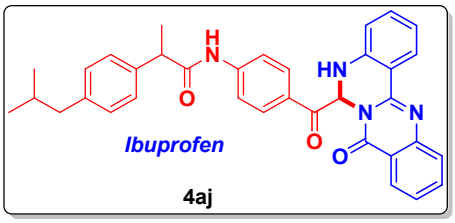
Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 115 mg, 78%; m.p.: 249-250 °C; FT-IR (KBr) ν (cm^{-1}) 3255, 3071, 2955, 2924, 1682, 1658, 1590, 1544, 1462, 1377, 1311, 1224, 1154, 1081, 968, 504; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.91 (s, 1H), 8.37 (d, J = 2.0 Hz, 1H), 8.19 – 8.11 (m, 3H), 8.09 (d, J = 8.0 Hz, 1H), 8.00 (d, J = 4.9 Hz, 1H), 7.60 (ddd, J = 8.1, 7.1, 1.5 Hz, 1H), 7.58 – 7.52 (m, 3H), 7.32 (ddd, J = 8.1, 7.3, 1.6 Hz, 1H), 6.92 – 6.84 (m, 2H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 187.3, 158.8, 148.2, 147.2, 143.6, 143.1, 142.0, 139.1, 138.1, 134.6, 133.9, 133.7, 129.5, 128.6, 127.0, 126.8, 125.7, 123.3, 121.6, 119.7, 116.0, 115.3, 91.2, 64.1; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{24}H_{15}IN_3O_2S$: 535.9930; found: 535.9932.

6-(1*H*-Indole-3-carbonyl)-10-methoxy-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (4ai)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Pale yellow solid; Yield: 136 mg, 86%; m.p.: 301-302 °C; FT-IR (KBr) ν (cm^{-1}) 3373, 3059, 2959, 2923, 2843, 1670, 1601, 1562, 1441, 1323, 1237, 1181, 1151, 1013; ^{1}H NMR (400 MHz, DMSO-d_6) δ 12.34 (s, 1H), 8.86 (s, 1H), 8.15 (dd, J = 8.2, 1.6 Hz, 1H), 7.98 (d, J = 7.9 Hz, 1H), 7.75 – 7.71 (m, 1H), 7.68 (d, J = 4.7 Hz, 1H), 7.55 – 7.46 (m, 3H), 7.32 (d, J = 4.6 Hz, 1H), 7.26 – 7.21 (m, 2H), 7.14 (ddd, J = 8.1, 7.1, 1.1 Hz, 1H), 6.85 – 6.79 (m, 2H), 3.88 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 187.6, 159.8, 157.4, 146.3, 143.4, 142.5, 136.4, 135.7, 132.8, 129.0, 126.5, 125.8, 124.5, 123.2, 122.2, 121.1, 120.7, 118.8, 115.9, 115.5, 112.3, 111.4, 106.3, 64.4, 55.7; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{25}H_{19}N_4O_3$: 423.1457; found: 423.1460.

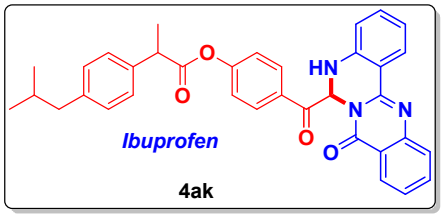
2-(4-Isobutylphenyl)-*N*-(4-(8-oxo-5,8-dihydro-6*H*-quinazolino[4,3-*b*]quinazoline-6-carbonyl)phenyl)propenamide (4aj)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7:3, v/v); Yellow solid; Yield: 199 mg, 85%; m.p.: 225-226 °C; FT-IR (KBr) ν (cm^{-1}) 3330, 3066, 2956, 2922, 2842, 2822, 1669, 1595, 1560, 1515, 1466, 1406, 1309, 1232, 1175, 1119, 891; ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.48 (s, 1H), 8.17 (dd, J = 8.0, 1.5 Hz, 1H), 8.10 (dt, J = 7.8, 2.1 Hz, 3H), 7.86 (ddd, J = 8.5, 7.1, 1.6 Hz, 1H), 7.81 (d, J = 8.8 Hz, 2H), 7.75 (d, J = 8.3 Hz, 1H), 7.68 (d, J = 5.0 Hz, 1H), 7.53 – 7.46 (m, 2H), 7.33 – 7.22 (m, 3H), 7.12 (d, J = 8.0 Hz, 2H), 6.85 (t, J = 7.4 Hz, 1H), 6.76 (d, J = 8.1 Hz, 1H), 3.85 (q, J = 6.9 Hz, 1H), 2.40 (d, J = 7.1 Hz, 2H), 1.84 – 1.74 (m, 1H), 1.42 (d, J = 6.9 Hz, 3H), 0.84 (d, J = 6.6 Hz, 6H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 190.4, 173.2, 160.1, 148.0, 147.9, 144.5, 143.0, 139.7, 138.7, 135.1, 133.4, 130.7, 129.1 (2C), 127.3, 127.2, 127.0, 126.8, 126.3, 119.9, 119.3, 118.6, 116.0, 115.7, 63.1, 45.8,

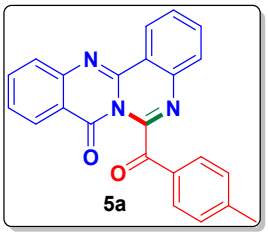
44.2, 29.6, 22.2, 18.6; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{35}H_{33}N_4O_3$: 557.2553; found: 557.2555.

4-(8-Oxo-5,8-dihydro-6*H*-quinazolino[4,3-*b*]quinazoline-6-carbonyl)phenyl 2-(4-isobutylphenyl)propanoate (4ak)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 7.5:2.5, v/v); Yellow solid; Yield: 170 mg, 72%; m.p.: 207-208 °C; FT-IR (KBr) ν (cm^{-1}) 3361, 3059, 2955, 2922, 2870, 2842, 1760, 1670, 1598, 1564, 1466, 1389, 1323, 1210, 1161, 1065; ^1H NMR (400 MHz, $CDCl_3$) δ 8.37 (dd, *J* = 8.0, 1.5 Hz, 1H), 8.21 (dd, *J* = 8.0, 1.5 Hz, 1H), 7.98 (d, *J* = 8.8 Hz, 2H), 7.81 – 7.71 (m, 2H), 7.45 – 7.37 (m, 2H), 7.29 (dd, *J* = 8.1, 1.4 Hz, 2H), 7.22 (ddd, *J* = 8.3, 7.4, 1.6 Hz, 1H), 7.15 (d, *J* = 7.9 Hz, 2H), 7.10 (d, *J* = 8.7 Hz, 2H), 6.98 (ddd, *J* = 8.2, 7.3, 1.1 Hz, 1H), 6.59 (d, *J* = 8.1 Hz, 1H), 4.97 (d, *J* = 5.0 Hz, 1H), 3.95 (q, *J* = 7.1 Hz, 1H), 2.47 (d, *J* = 7.1 Hz, 2H), 1.91 – 1.81 (m, 1H), 1.61 (d, *J* = 7.1 Hz, 3H), 0.91 (d, *J* = 6.6 Hz, 6H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6) δ 190.3, 172.9, 161.2, 155.3, 148.5, 147.6, 141.4, 141.3, 136.8, 134.9, 133.3, 131.2, 130.6, 129.8, 127.9, 127.8, 127.3, 126.9, 126.5, 122.2, 121.9, 120.5, 118.5, 117.0, 63.6, 45.4, 45.2, 30.3, 22.5, 18.5; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{35}H_{32}N_3O_4$: 558.2393; found: 558.2397.

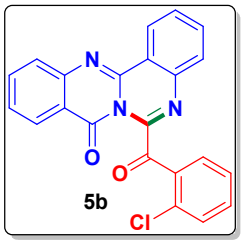
6-(4-Methylbenzoyl)-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (5a)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 9:1, v/v); White solid; Yield: 37 mg, 74%; m.p.: 304-305 °C; FT-IR (KBr) ν (cm^{-1}) 3124, 3018, 2943, 2872, 1682, 1625, 1582, 1563, 1464, 1428, 1311, 1234, 1108, 1049; ^1H NMR (400 MHz, $CDCl_3$) δ 8.90 (dd, *J* = 7.5, 1.3 Hz, 1H), 8.30 (dt, *J* = 8.0, 1.1 Hz, 1H), 7.96 (d, *J* = 8.3 Hz, 2H), 7.93 – 7.88 (m, 2H), 7.87 – 7.80 (m, 2H), 7.71 (ddd, *J* = 8.4, 6.5, 2.0 Hz, 1H), 7.52 – 7.48 (m, 1H), 7.34 (d, *J* = 7.9 Hz, 2H), 2.45 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6) δ 185.7, 159.6, 147.5, 146.8, 145.0, 144.9, 142.0, 136.3, 134.1, 132.1, 130.1, 129.7 (2C), 128.6, 127.8, 127.7, 127.1,

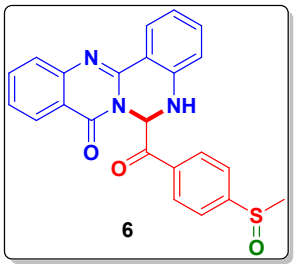
126.3, 121.6, 119.2, 22.0; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{16}N_3O_2$: 366.1243; found: 366.1243.

6-(2-Chlorobenzoyl)-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (5b)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 9:1, v/v); White solid; Yield: 35 mg, 70%; m.p.: 286-287 °C; FT-IR (KBr) ν (cm^{-1}) 3015, 2956, 2923, 1687, 1627, 1587, 1555, 1462, 1432, 1313, 1226, 1111, 1049, 740, 688; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.93 – 8.88 (m, 1H), 8.41 – 8.37 (m, 1H), 8.37 – 8.33 (m, 1H), 7.95 – 7.89 (m, 2H), 7.84 – 7.78 (m, 2H), 7.70 (ddd, *J* = 8.4, 5.6, 2.9 Hz, 1H), 7.59 – 7.50 (m, 3H), 7.46 (dd, *J* = 7.9, 1.4 Hz, 1H); ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) δ 183.1, 159.7, 147.6, 146.9, 144.9, 142.3, 136.2, 135.4, 134.5, 133.9, 132.7, 132.6, 131.3, 129.4, 128.3, 127.7, 127.6, 127.3, 126.9, 126.2, 121.3, 119.0; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{22}H_{13}ClN_3O_2$: 386.0696; found: 386.0696.

6-(4-(Methylsulfinyl)benzoyl)-5,6-dihydro-8*H*-quinazolino[4,3-*b*]quinazolin-8-one (6)



Purified by column chromatography (eluent: petroleum ether/EtOAc = 6.5:3.5, v/v); Yellow solid; Yield: 76 mg, 73%; m.p.: 247-248 °C; FT-IR (KBr) ν (cm^{-1}) 3292, 3059, 2996, 2954, 2921, 2852, 1688, 1657, 1592, 1559, 1467, 1393, 1325, 1220, 1173, 1041, 955; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.38 (d, *J* = 8.4 Hz, 2H), 8.25 (dd, *J* = 8.0, 1.5 Hz, 1H), 8.17 (dd, *J* = 7.9, 1.6 Hz, 1H), 7.98 – 7.91 (m, 3H), 7.86 – 7.80 (m, 2H), 7.67 (d, *J* = 5.2 Hz, 1H), 7.57 (td, *J* = 7.5, 1.2 Hz, 1H), 7.35 (ddd, *J* = 8.5, 7.2, 1.6 Hz, 1H), 6.95 (ddd, *J* = 8.2, 7.2, 1.1 Hz, 1H), 6.85 (d, *J* = 8.1 Hz, 1H), 2.90 (s, 3H); ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) δ 192.0, 160.6, 153.1, 148.3, 148.2, 143.2, 135.7, 134.9, 134.0, 130.3, 127.8, 127.3, 126.9, 126.8, 124.7, 120.2, 120.1, 116.7, 116.1, 63.8, 43.3; HRMS (ESI) (m/z) $[M + H]^+$ calcd. for $C_{23}H_{18}N_3O_3S$: 416.1069; found: 416.1066.

References

1. a) K. M. Khan, S. M. Saad, N. N. Shaikh,; S. Hussain, M. I. Fakhri, S. Perveen, M. Taha and M. I. Choudhary, *Bioorg. Med. Chem.,* **2014**, *22*, 3449-3454; b) S. Sahoo, M. A. Rao and S. Pal, *J. Org. Chem.,* **2023**, *88*, 10701-10710.
2. G. Pawar, S. M. Ghouse, S. V. Joshi, P. Rana, S. Kar, P. M. Sarma, S. R. Dannarm, R. Sonti and S. Nanduri, *ChemistrySelect,* **2022**, *7*, e202200500.
3. V. Rajeshkumar, C. Neelamegam and S. Anandan, *Org. Biomol. Chem.,* **2019**, *17*, 982−991.

[1]H NMR experimental studies

Experiment procedure: The reaction was carried out in the NMR tube by taking 7.0 mg of 4-methylacetophenone **3c** in 0.5 mL of DMSO-*d*$_6$ under the standard reaction condition and subsequently the ^{1}H NMR were recorded at specified time intervals.

Plot A: Indicates the presence of 4-methylacetophenone **3c** and partial conversion into phenylglyoxal and its hydrated form. The by-product DMS-*d*$_6$ was also observed at 2.05 ppm.
Plot B: Indicates the complete conversion of 4-methylacetophenone to phenylglyoxal and its hydrated form.
Plot C: Indicates the formation of product **4c** and the peaks are well aligned with ^{1}H NMR spectrum of pure product **4c (Plot D)** except -NH and -CH protons which have merged and appear as a singlet at 7.51 ppm.
Plot D: ^{1}H NMR spectrum of pure product **4c.**
Plot E: D_2O exchange experiment for the confirmation of -NH proton.

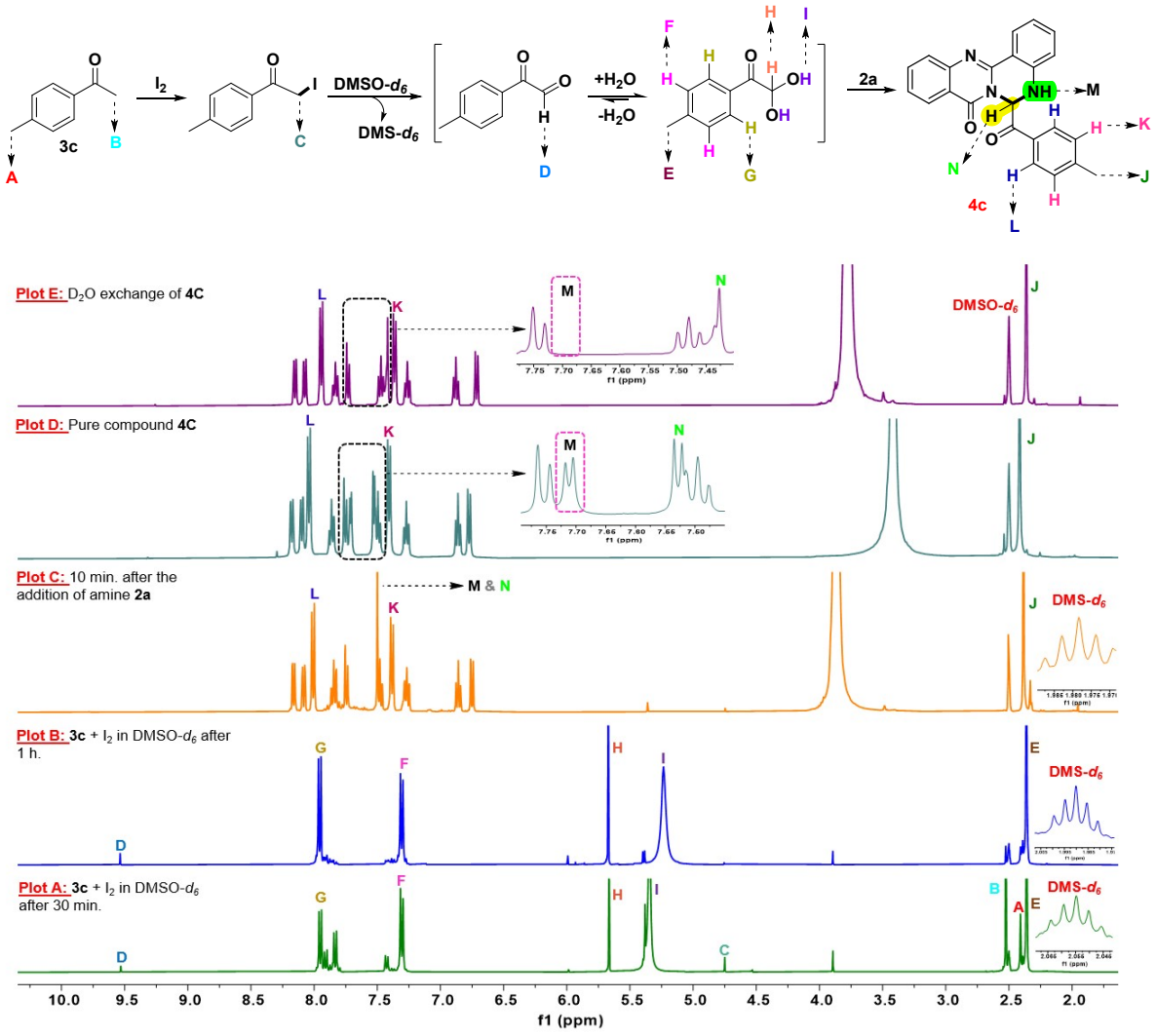


Figure S1. ^{1}H NMR experimental studies to monitor the reaction progress.

$^{13}C\{^{1}H\}$ NMR experimental studies

Experiment procedure: The reaction was carried out in the NMR tube by taking 7.0 mg of 4-methylacetophenone **3c** in 0.5 mL of DMSO-*d*$_6$ under the standard reaction condition and subsequently the $^{13}C\{^{1}H\}$ NMR were recorded at specified time intervals.

Plot A: Indicates the presence of 4-methylacetophenone **3c** and partial conversion into phenylglyoxal and its hydrated form. The by-product DMS-*d*$_6$ was also observed at 16.91 ppm.
Plot B: Indicates the complete conversion of 4-methylacetophenone **3c** to hydrated form of phenylglyoxal.
Plot C: Indicates the product **4c** formation and the peaks are well aligned with $^{13}C\{^{1}H\}$ NMR spectrum of pure product **4c** which is shown in **Plot D**.
Plot D: $^{13}C\{^{1}H\}$ NMR spectrum of pure product **4c**.

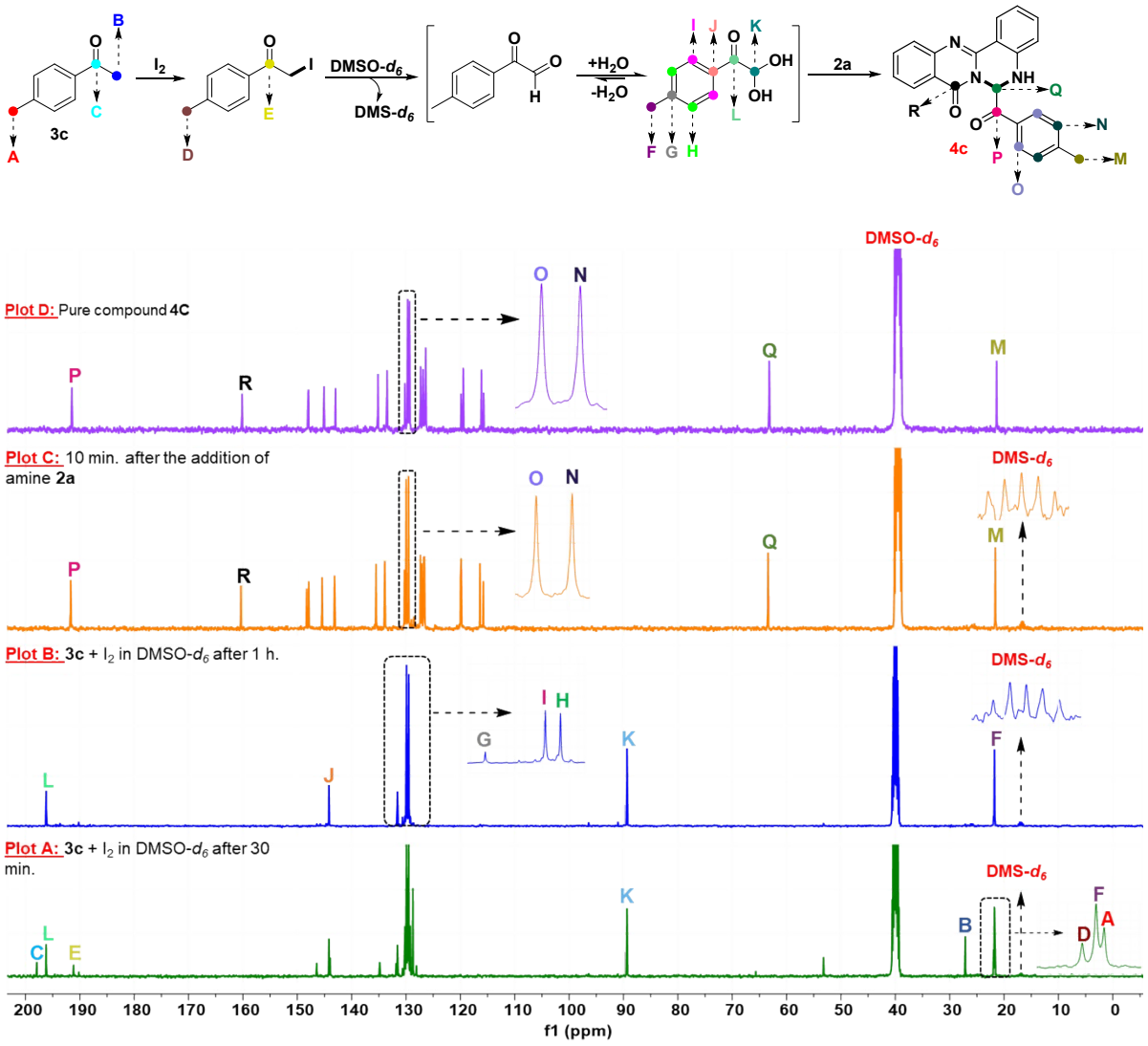


Figure S2. $^{13}C\{^{1}H\}$ NMR experimental studies to monitor the reaction progress.

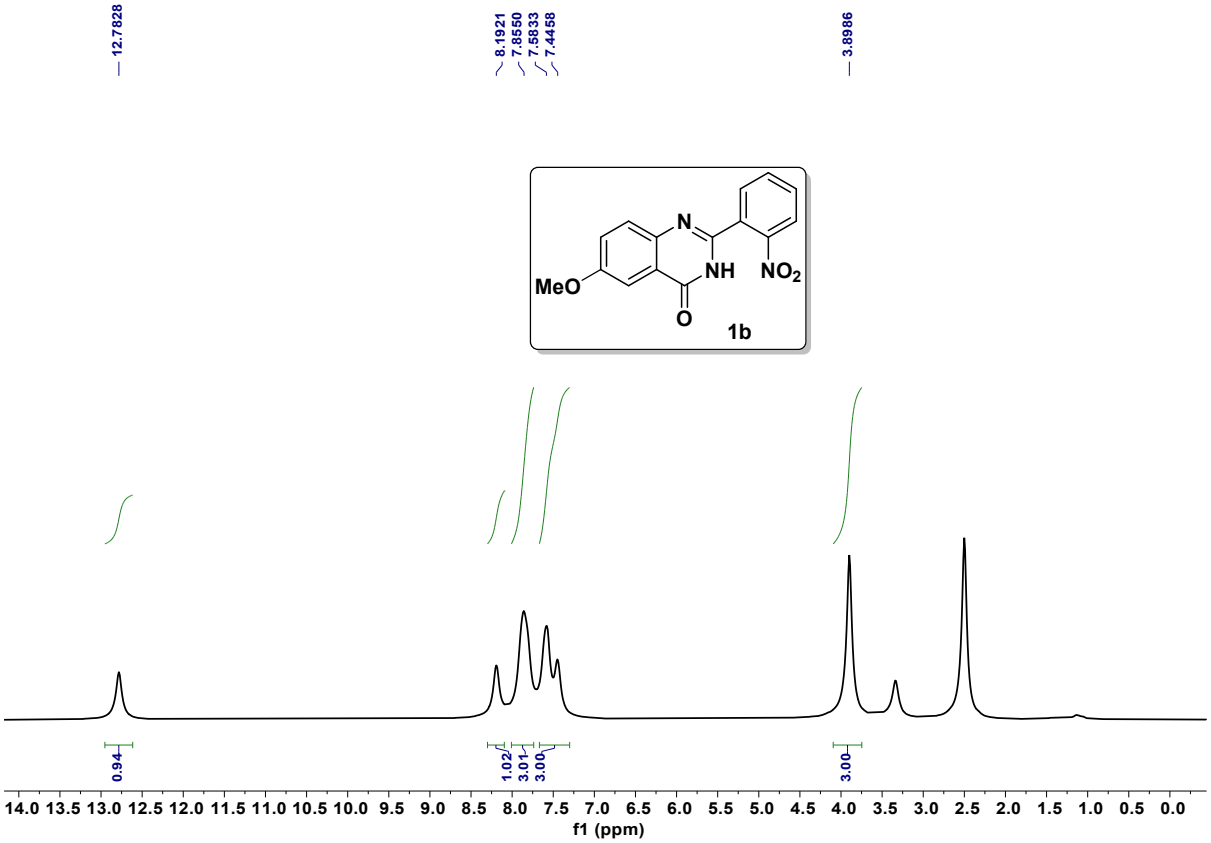


Figure 1. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **1b**

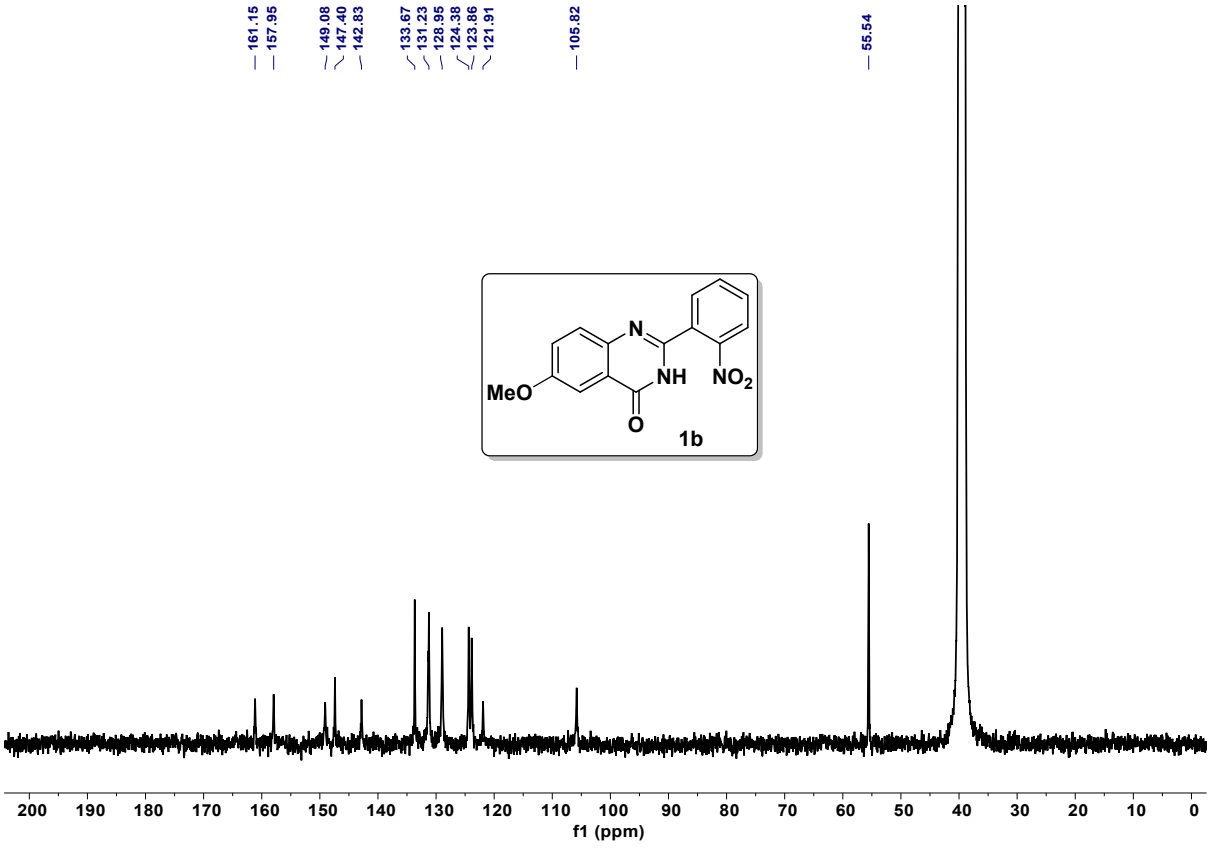


Figure 2. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **1b**

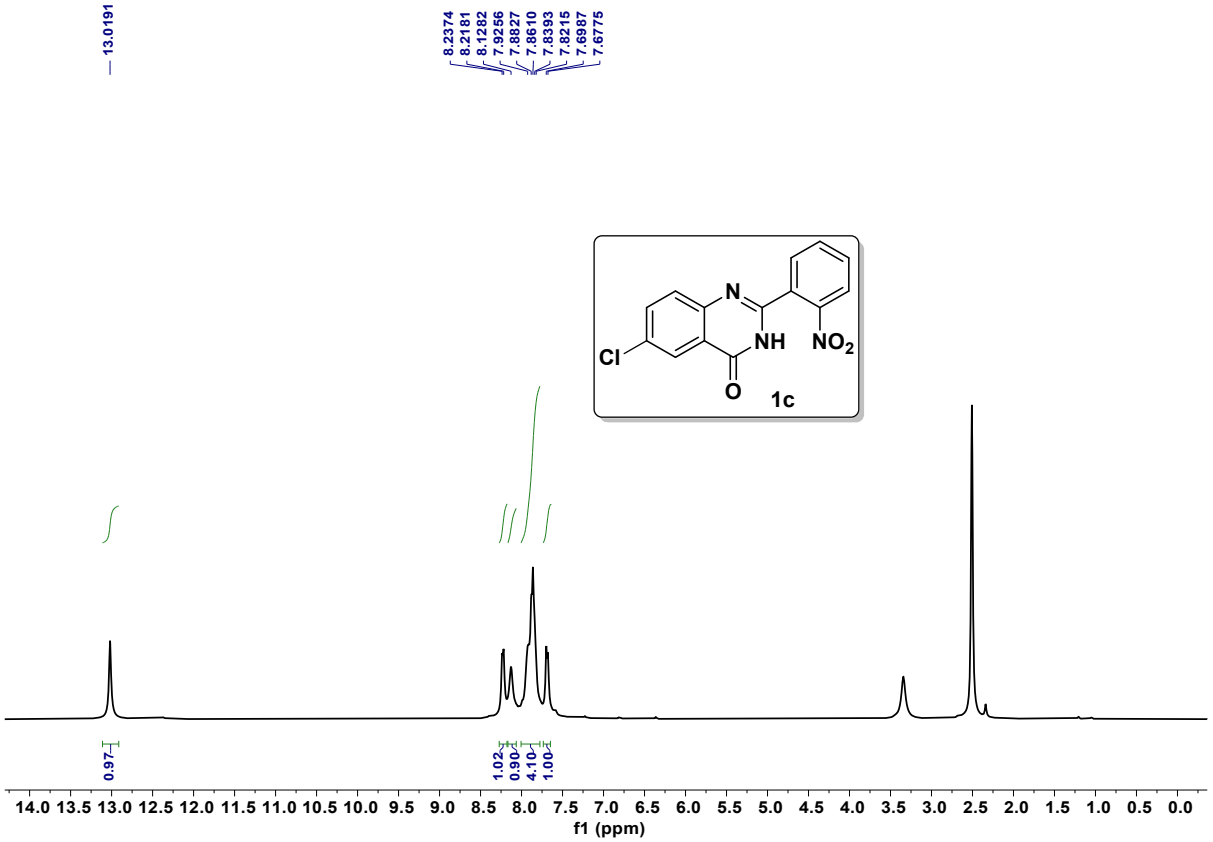


Figure 3. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **1c**

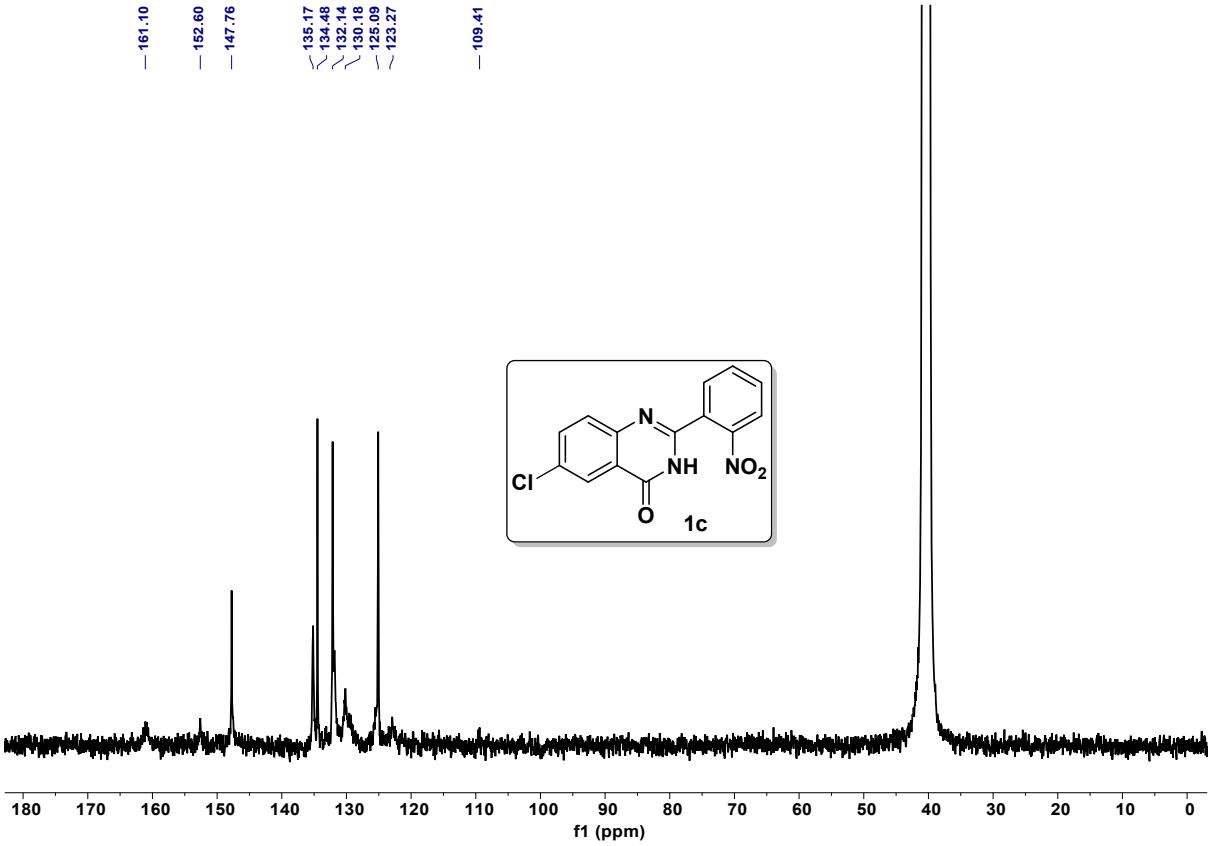


Figure 4. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **1c**

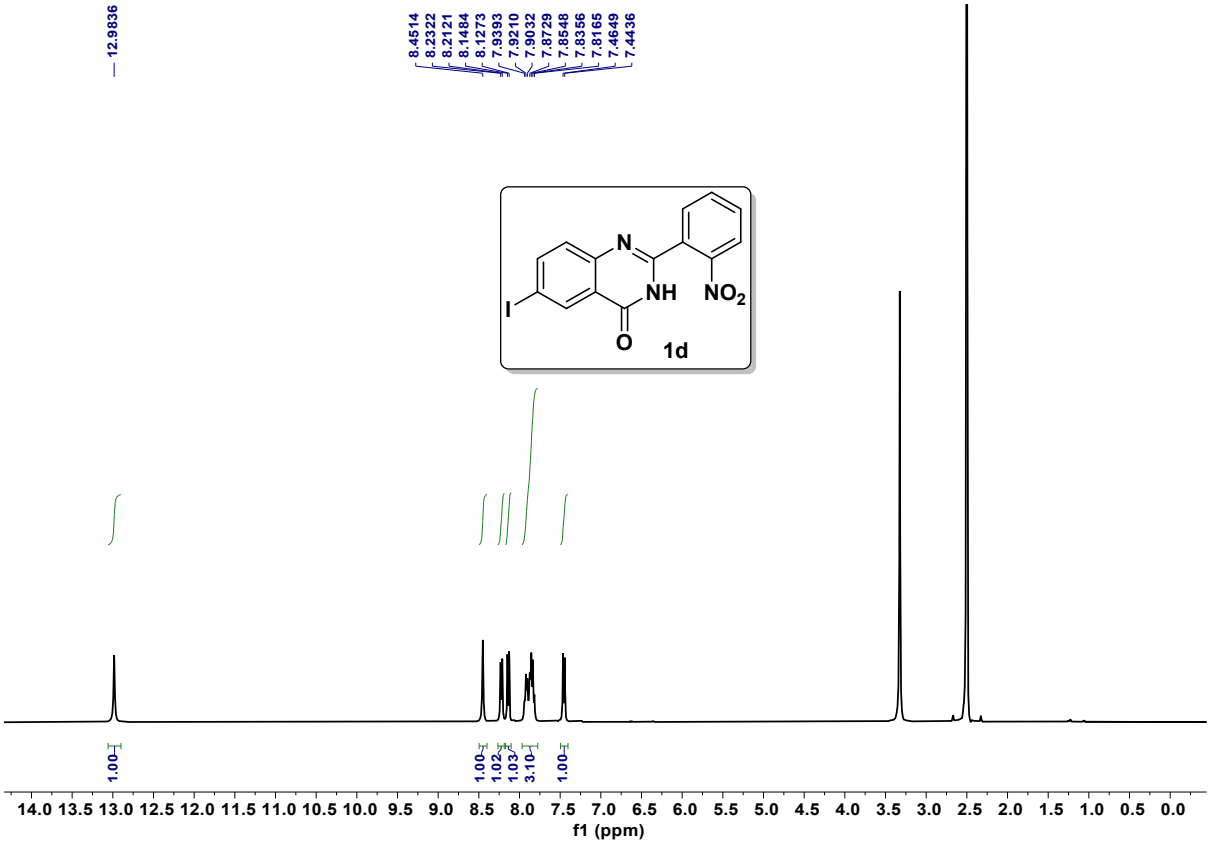


Figure 5. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **1d**

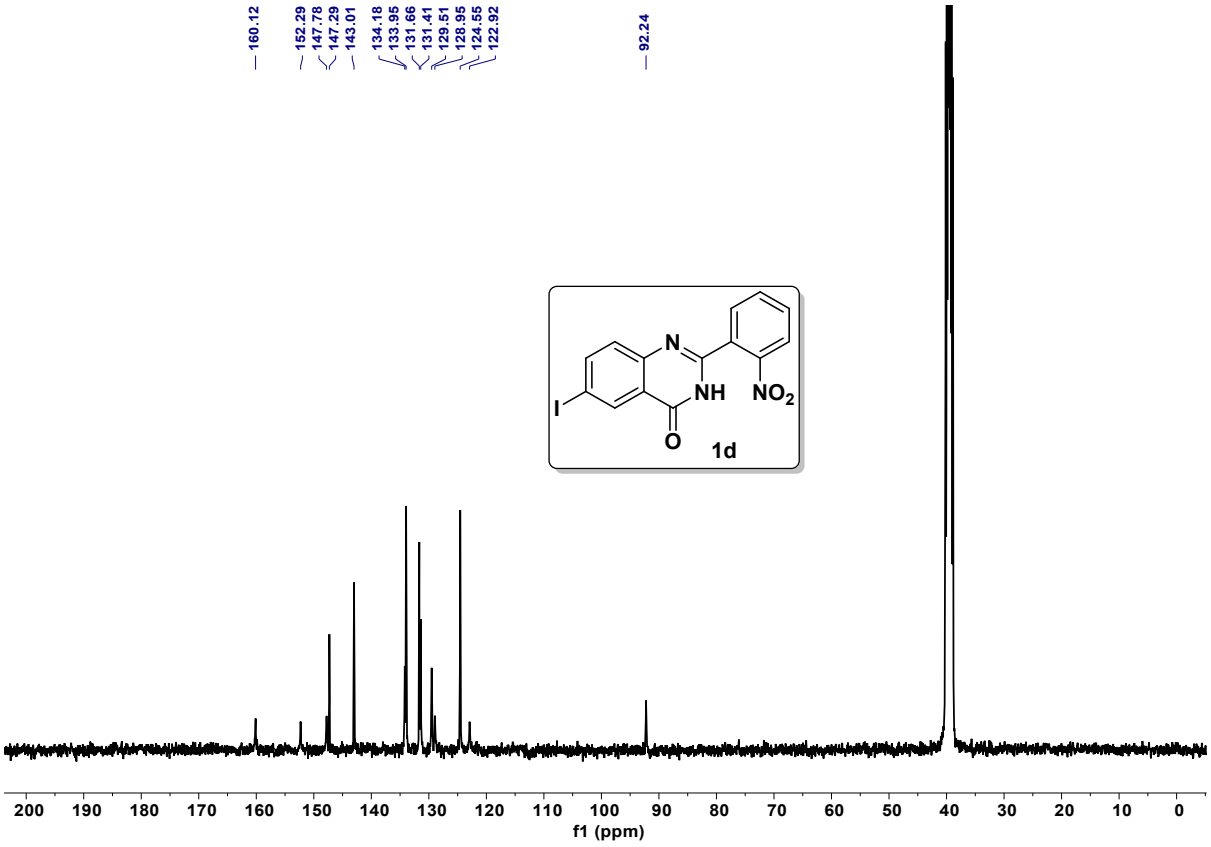


Figure 6. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **1d**

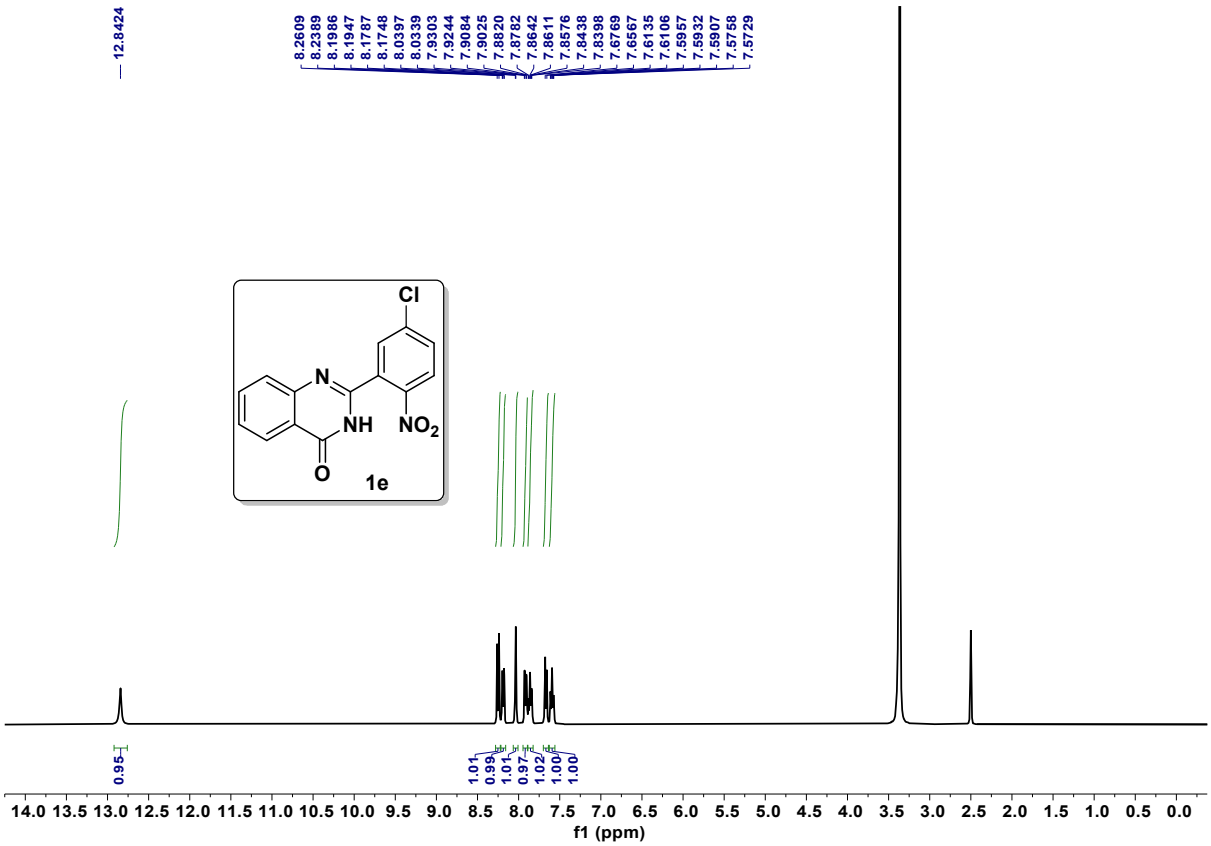


Figure 7. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **1e**

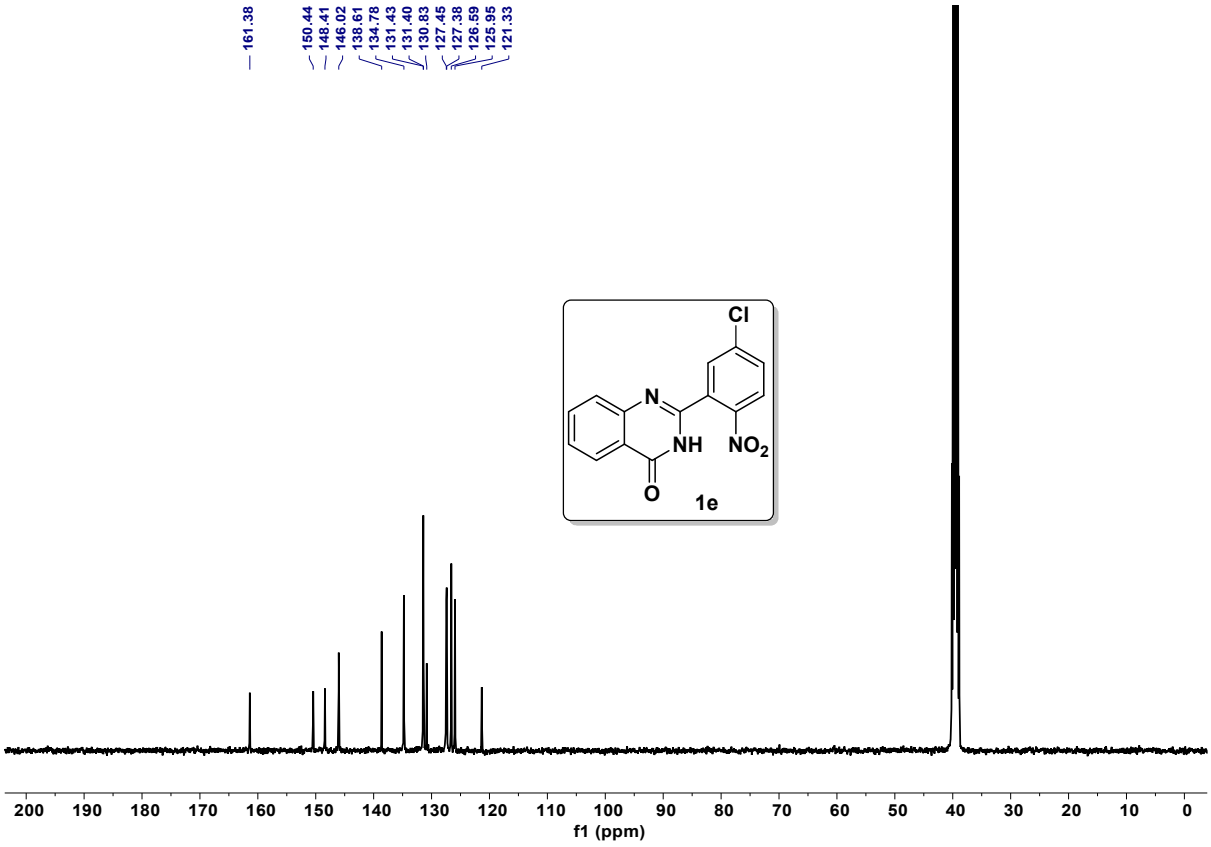


Figure 8. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **1e**

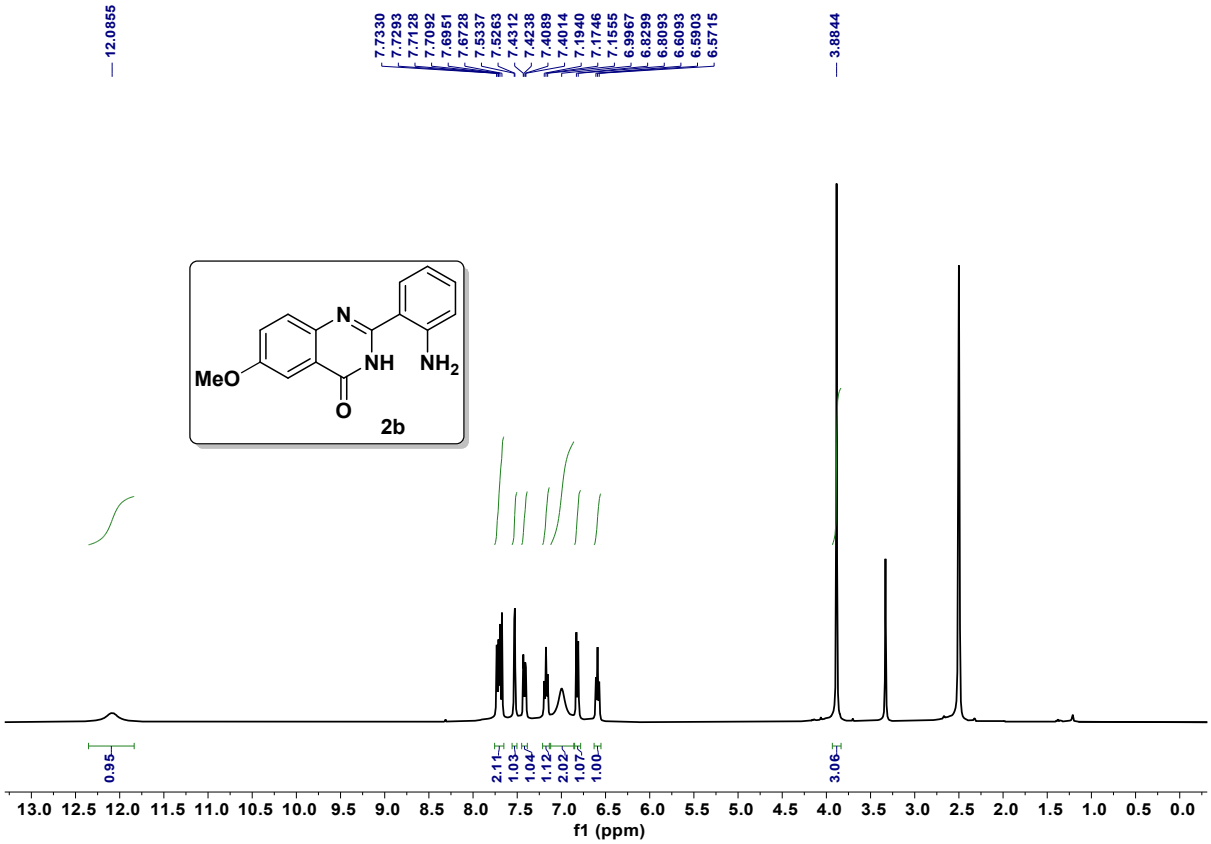


Figure 9. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **2b**

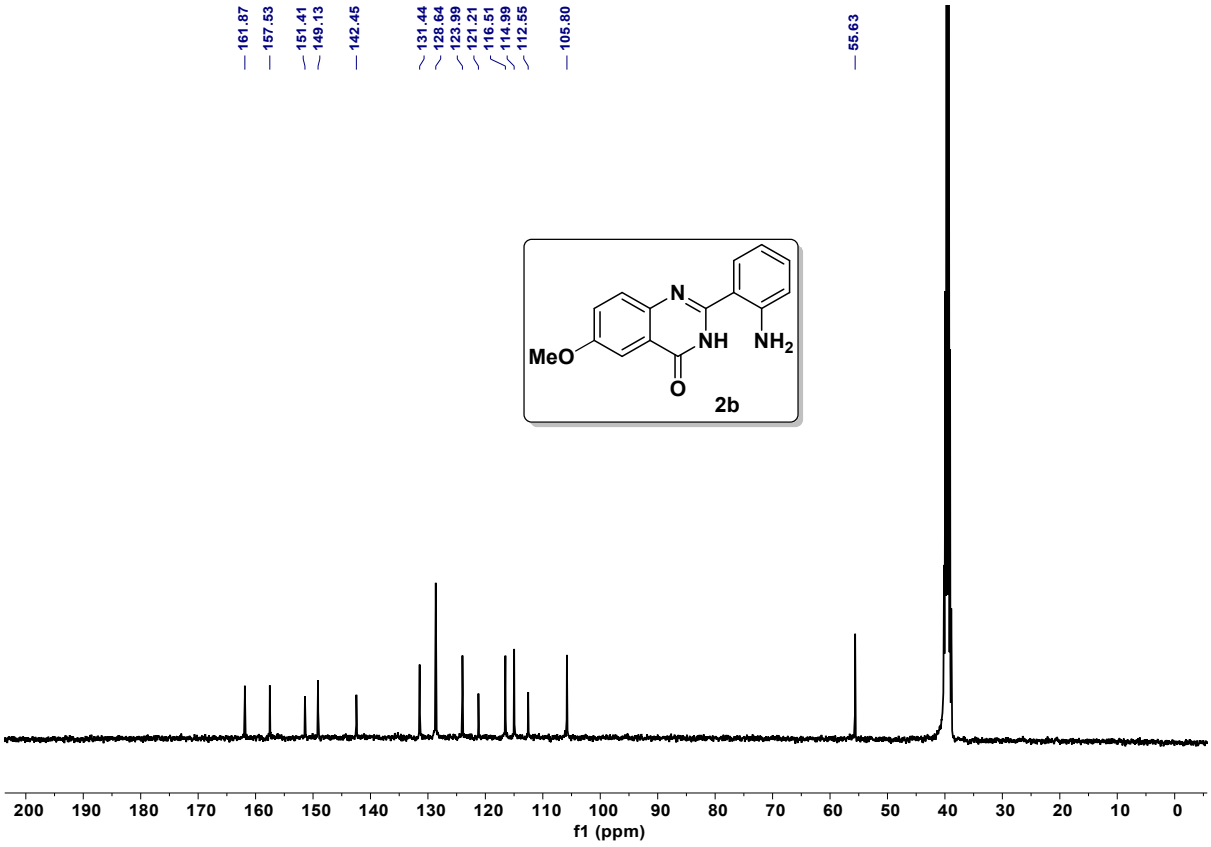


Figure 10. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **2b**

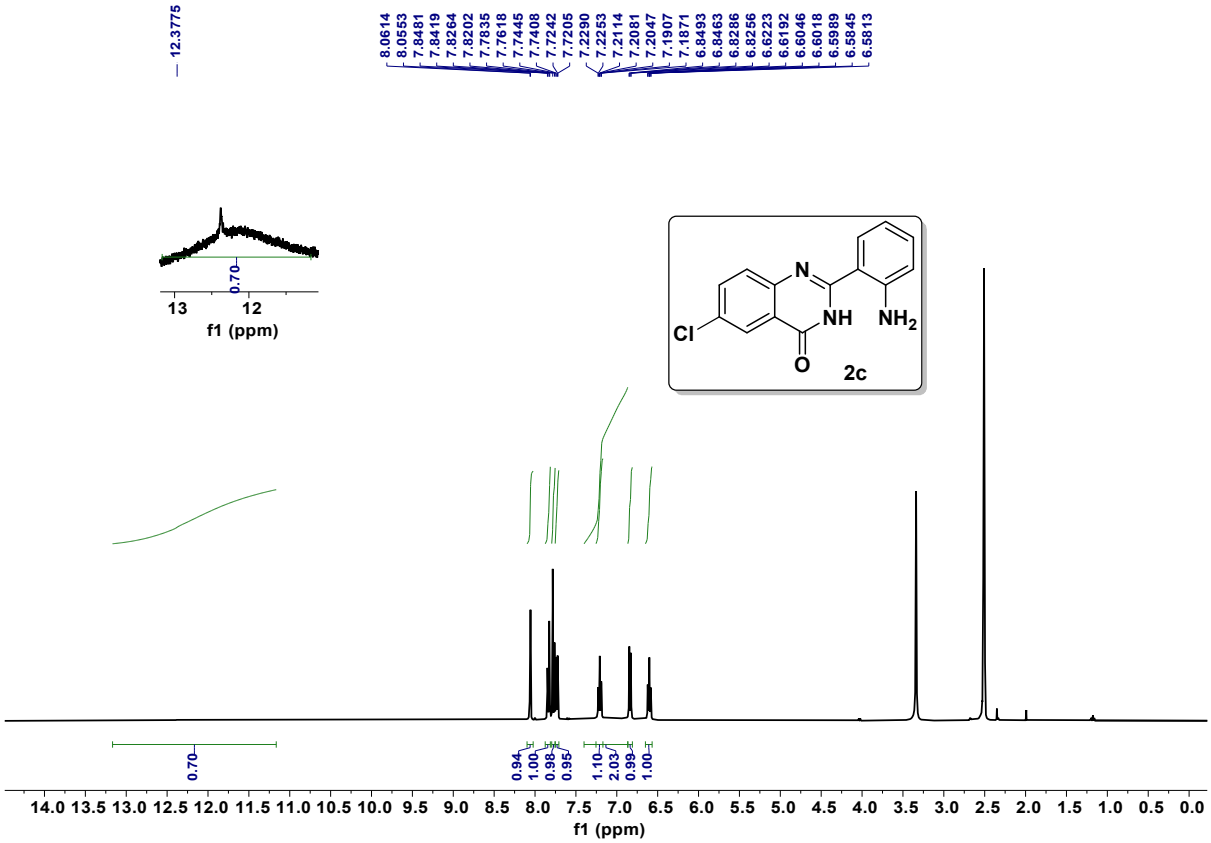


Figure 11. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **2c**

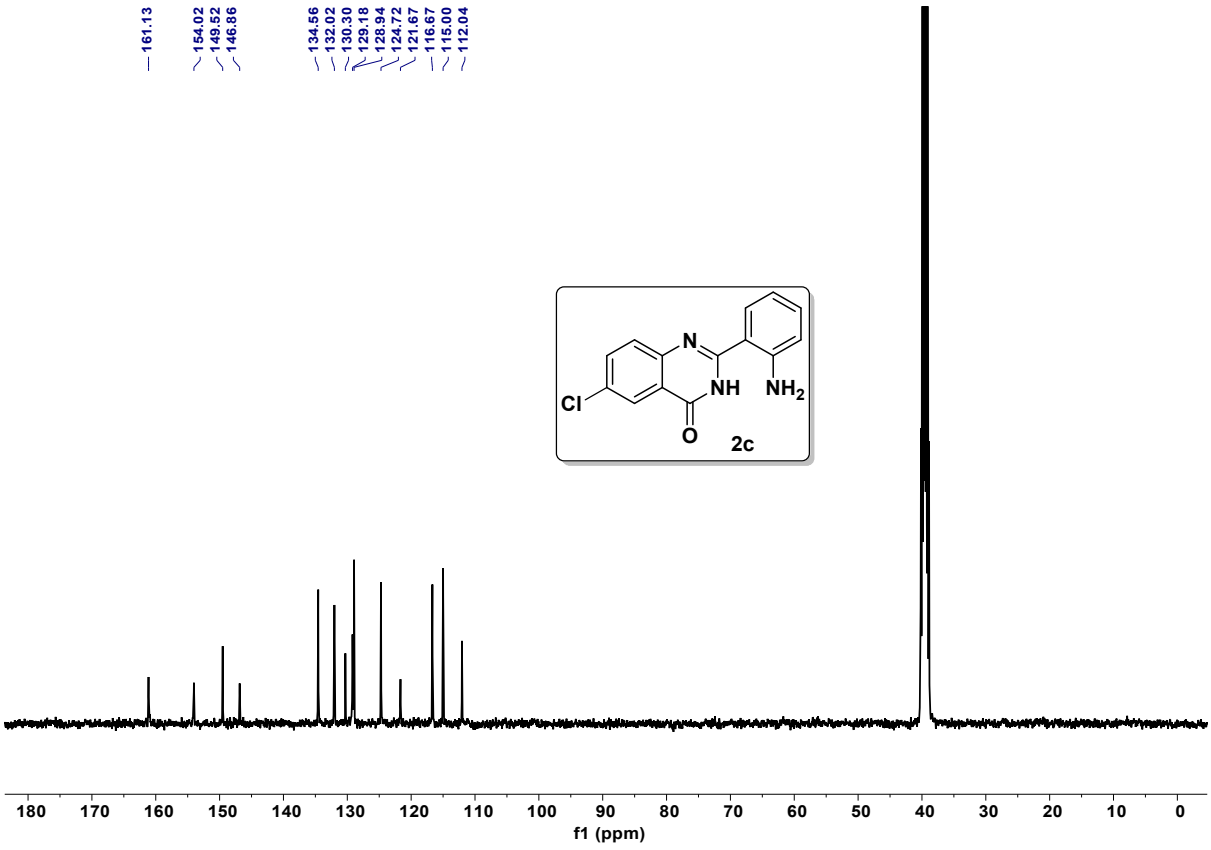


Figure 12. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **2c**

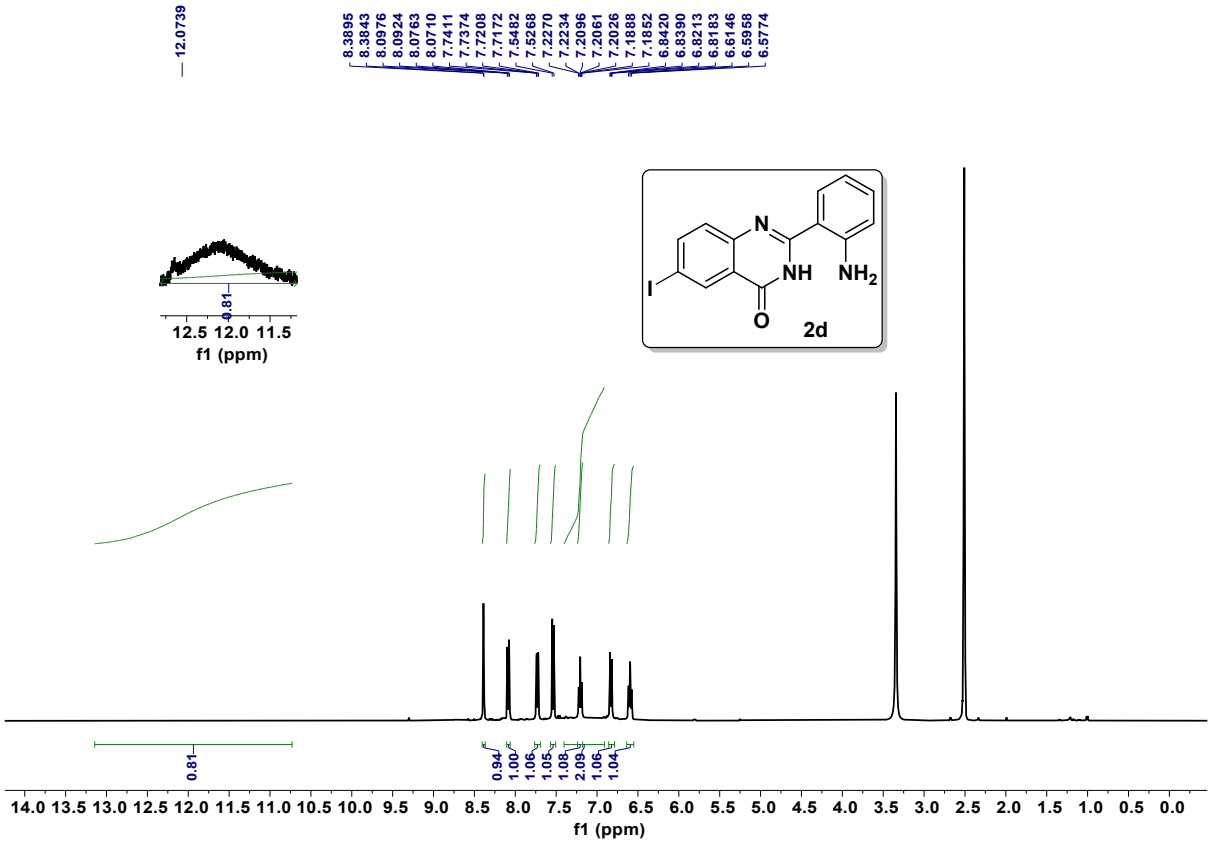


Figure 13. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **2d**

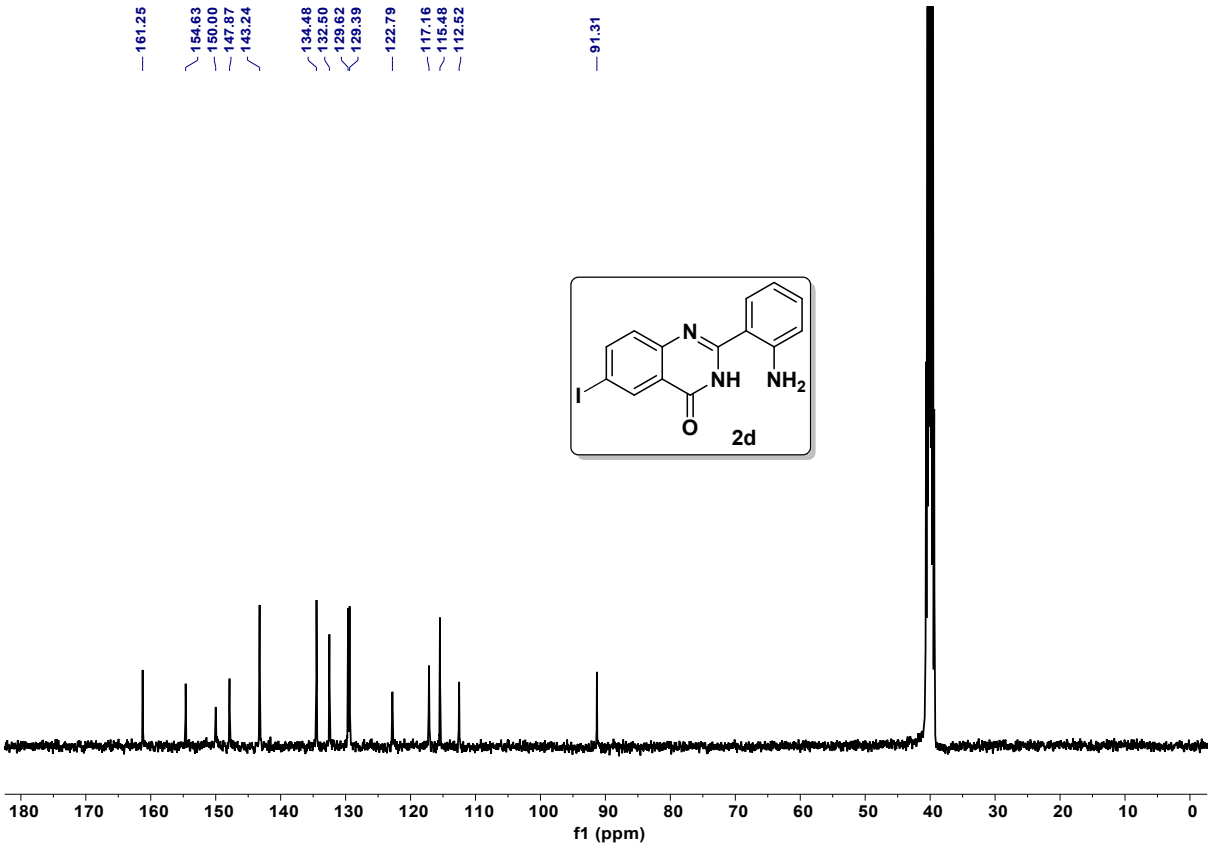


Figure 14. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **2d**

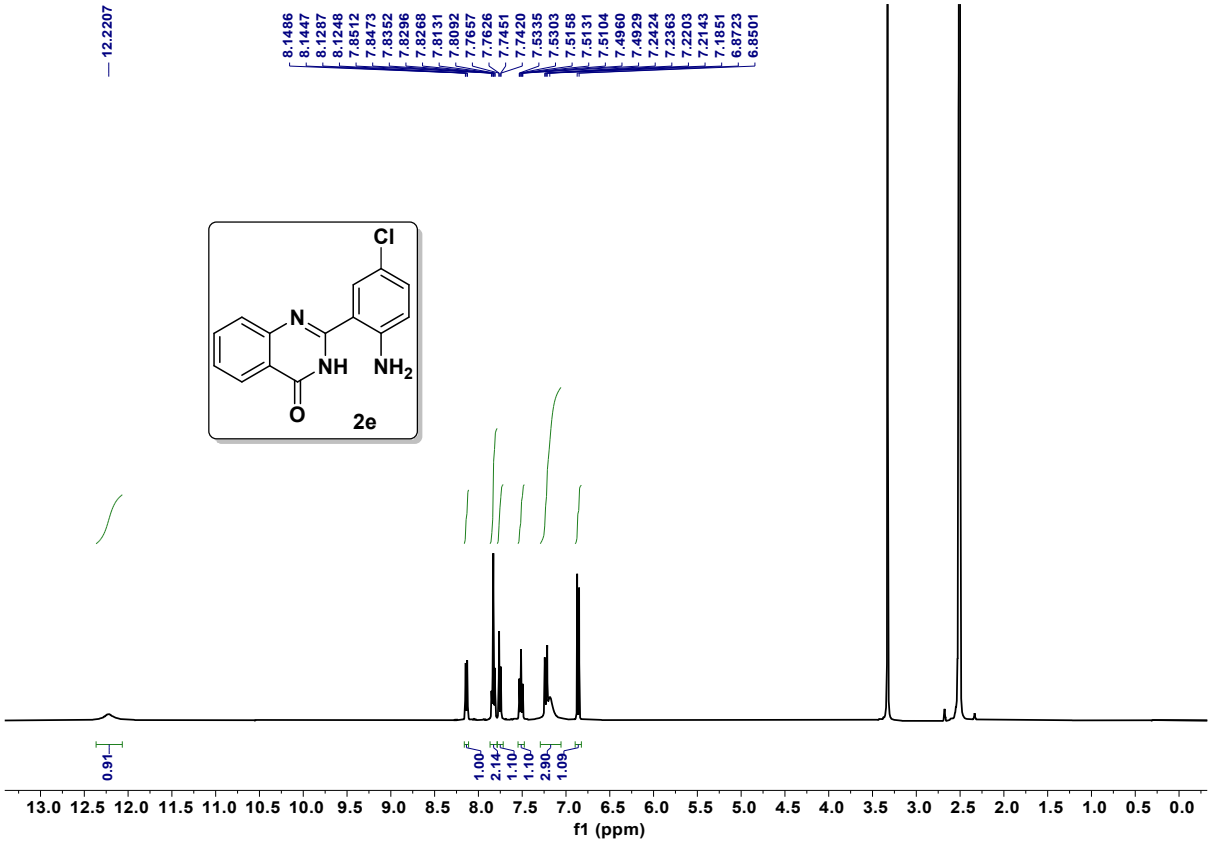


Figure 15. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **2e**

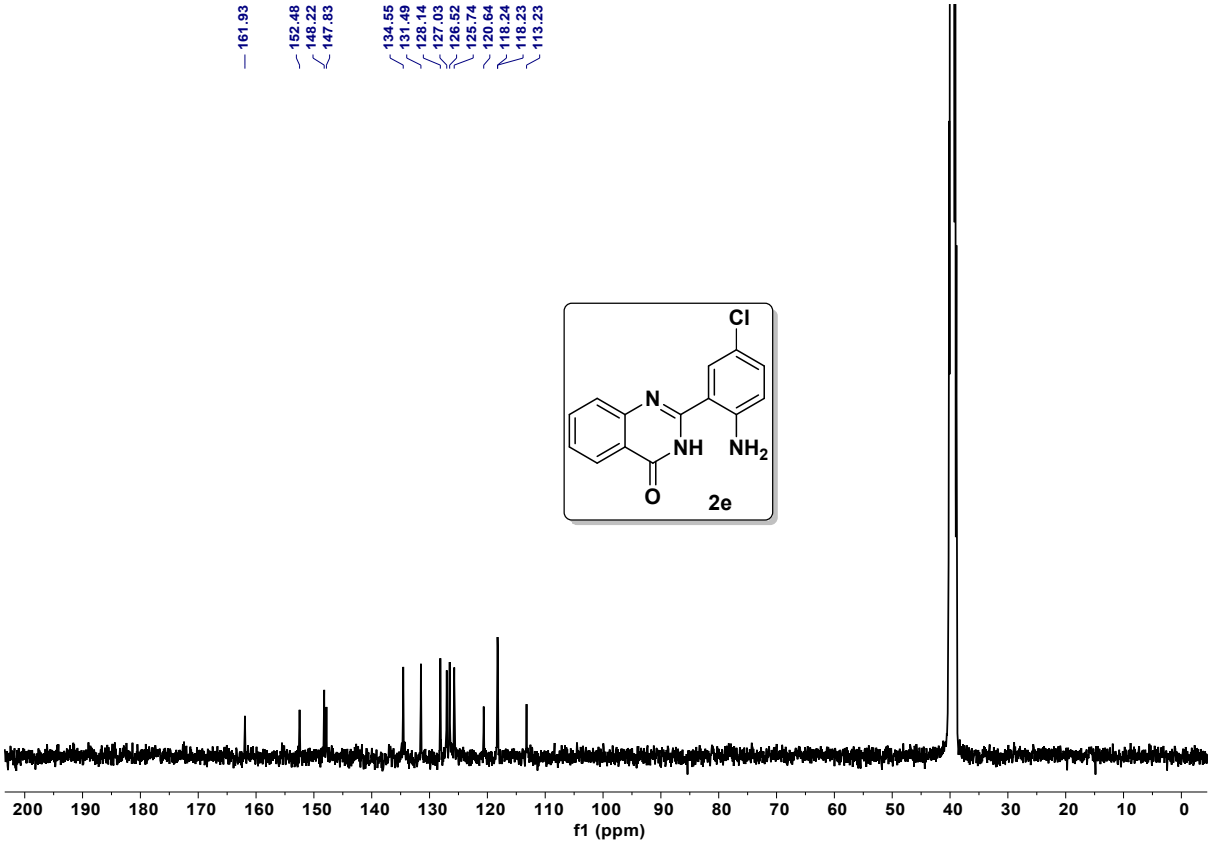


Figure 16. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **2e**

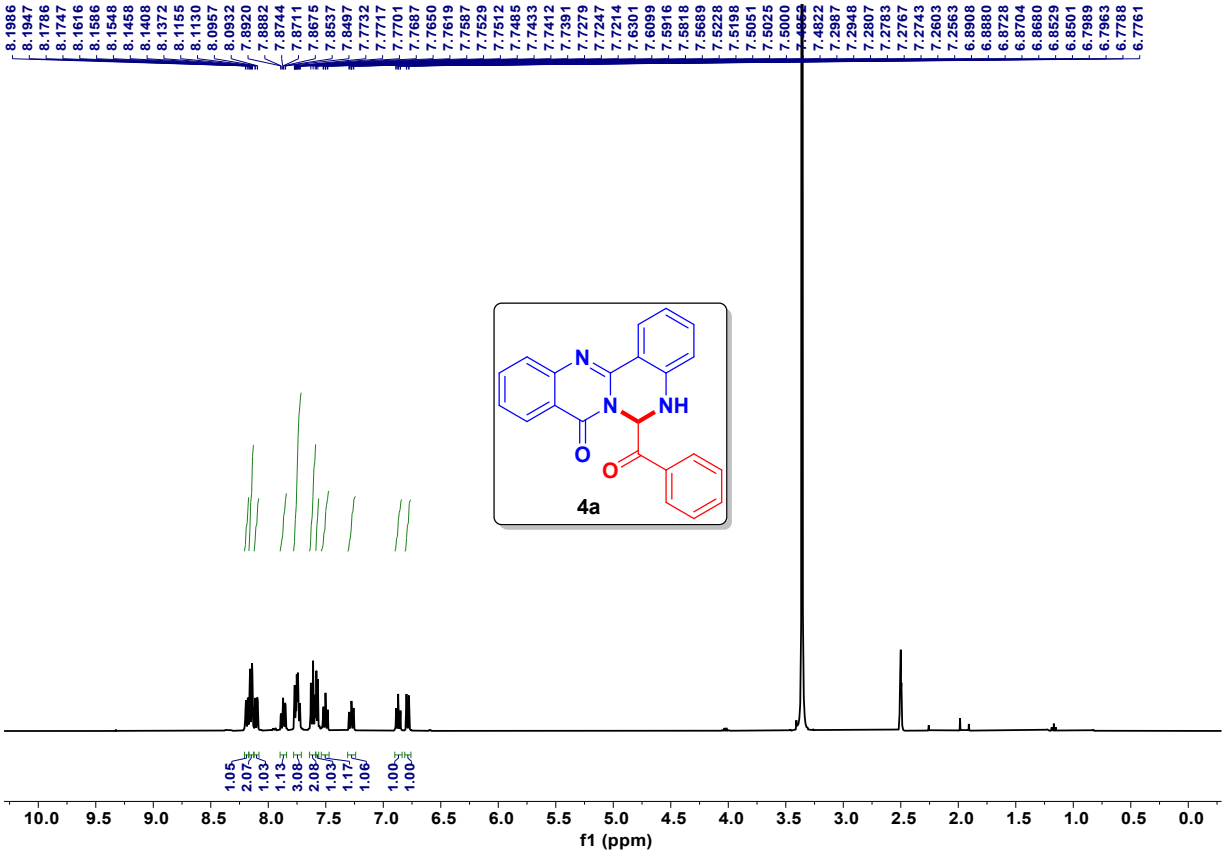


Figure 17. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4a**

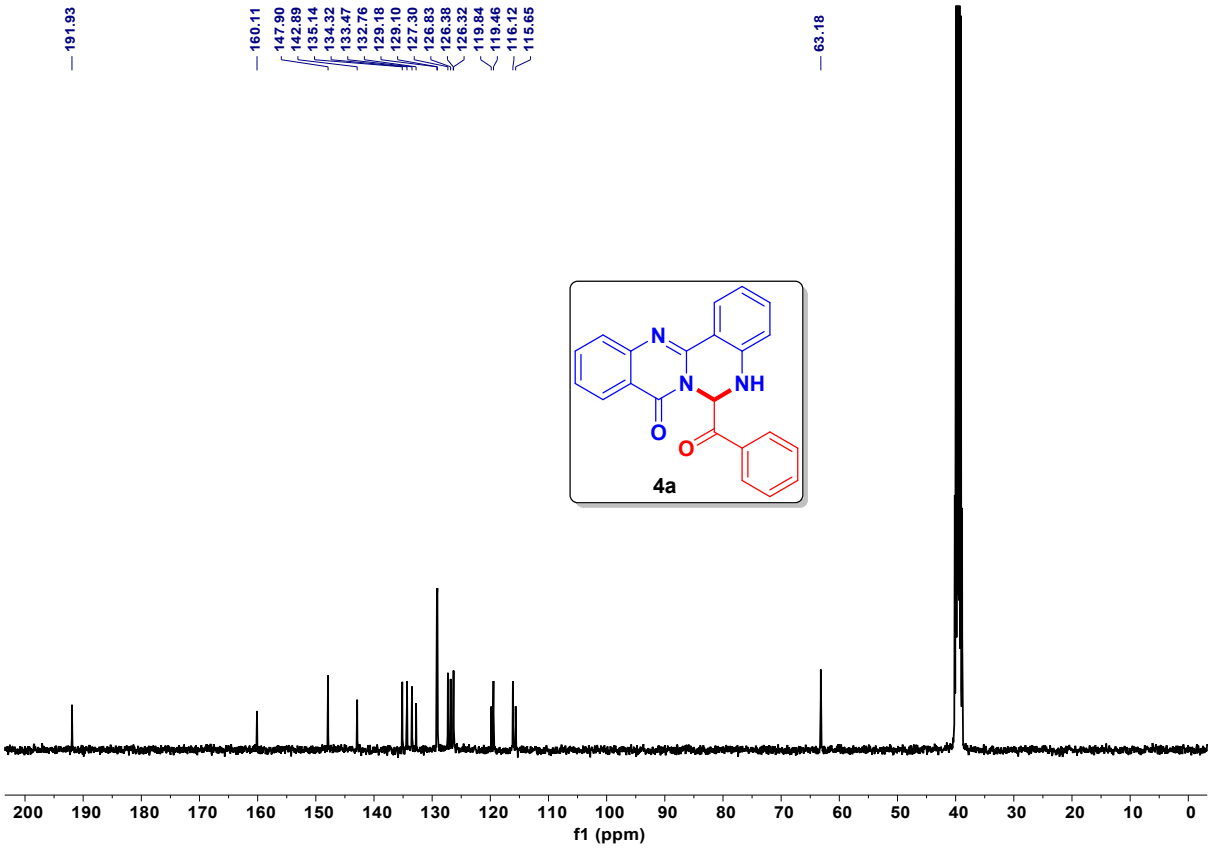


Figure 18. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4a**

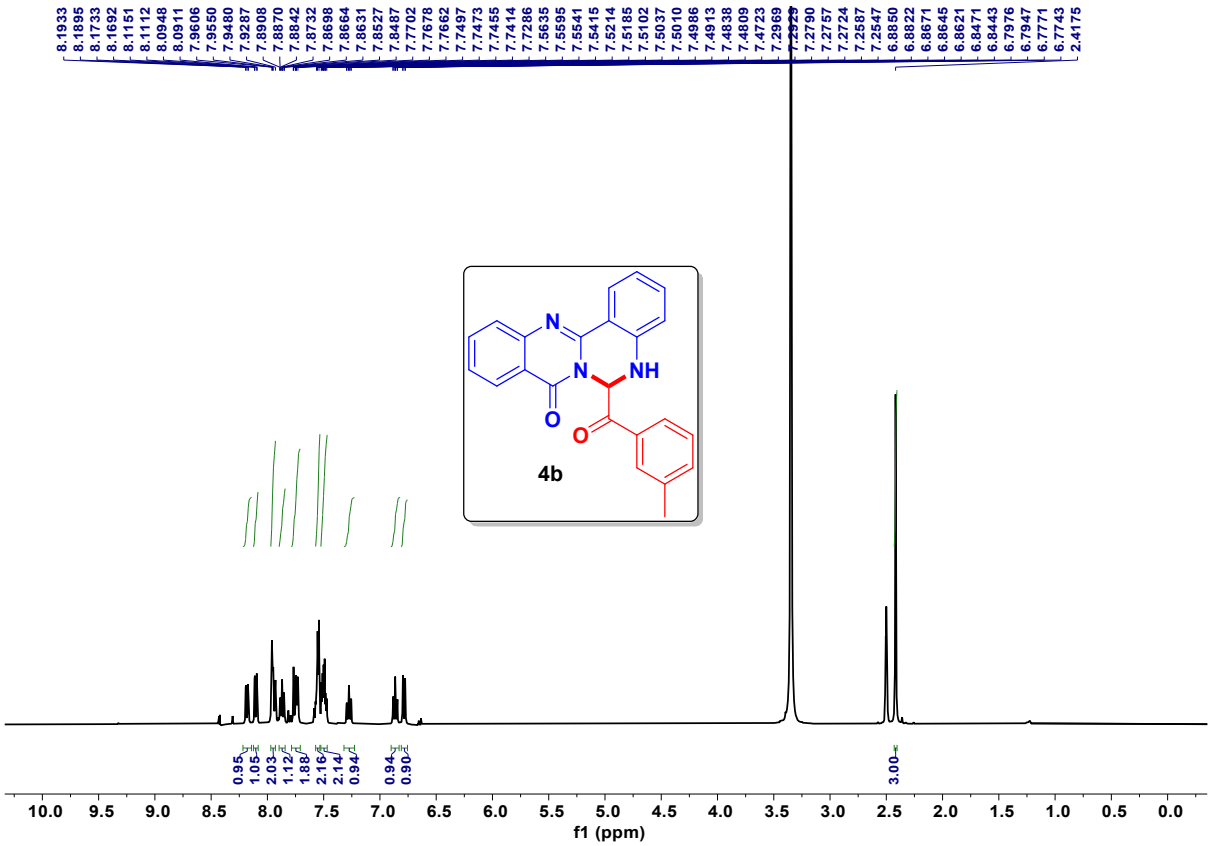


Figure 19. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4b**

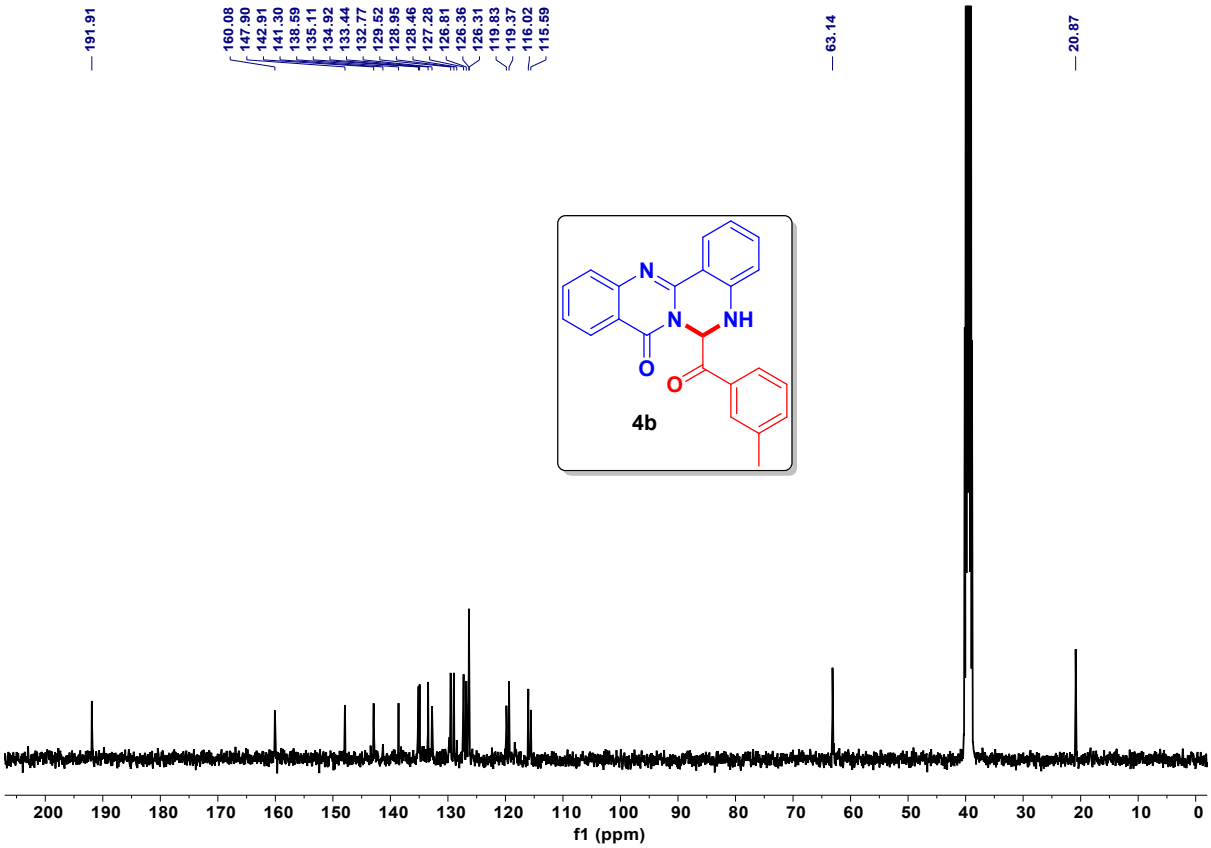


Figure 20. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4b**

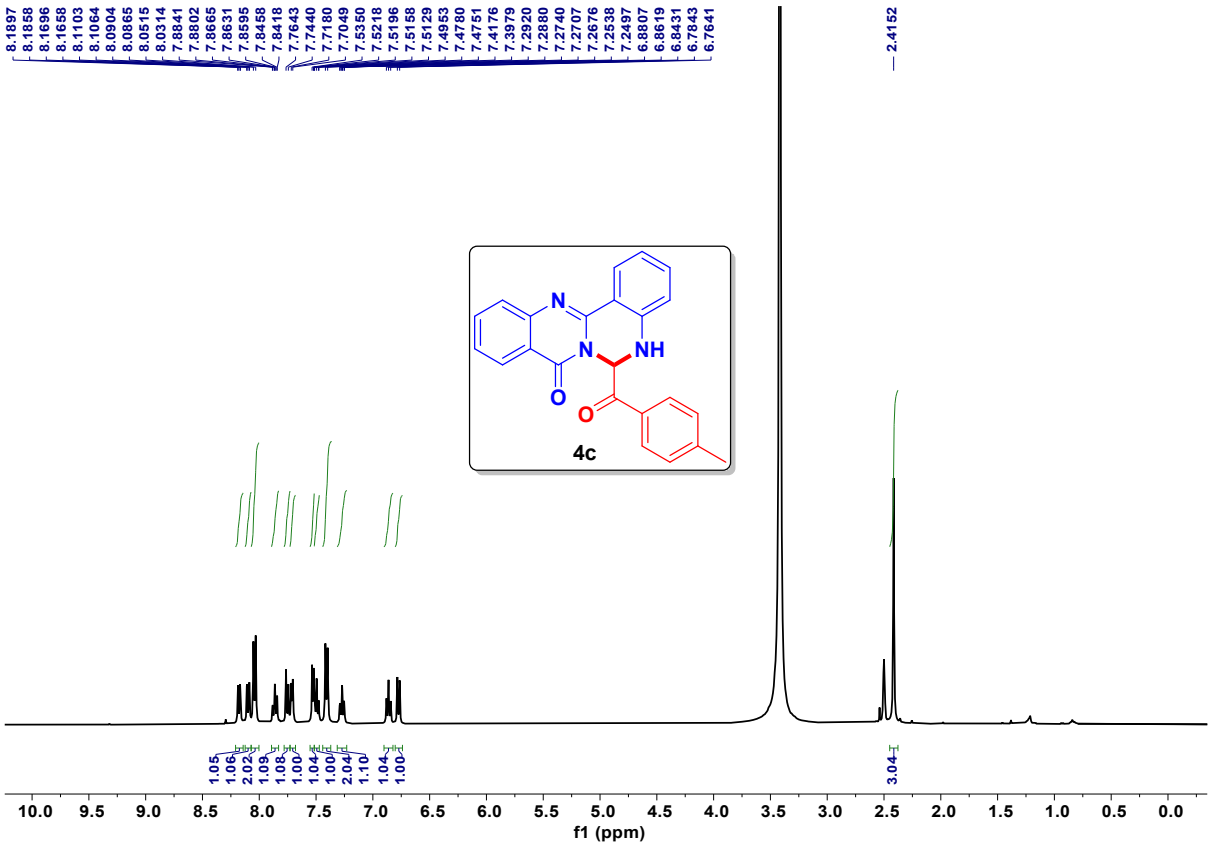


Figure 21. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4c**

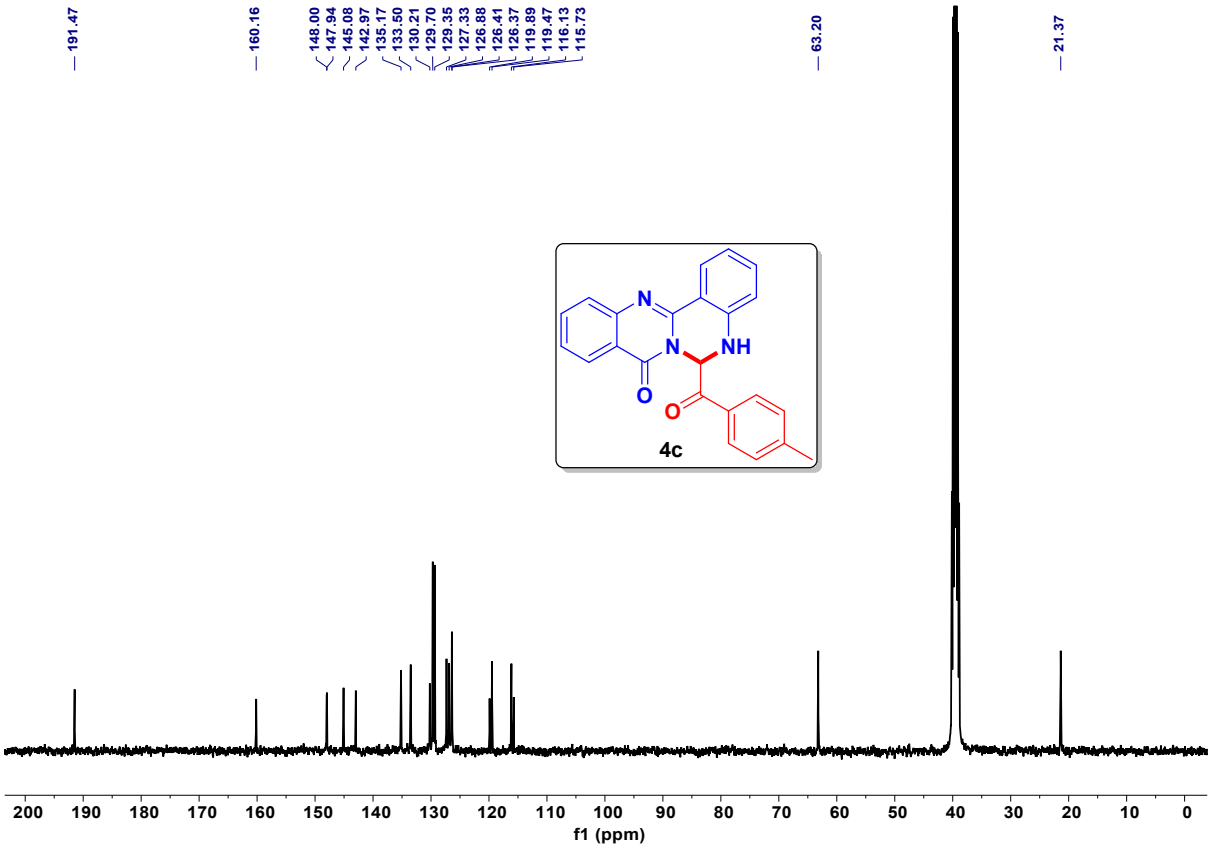


Figure 22. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4c**

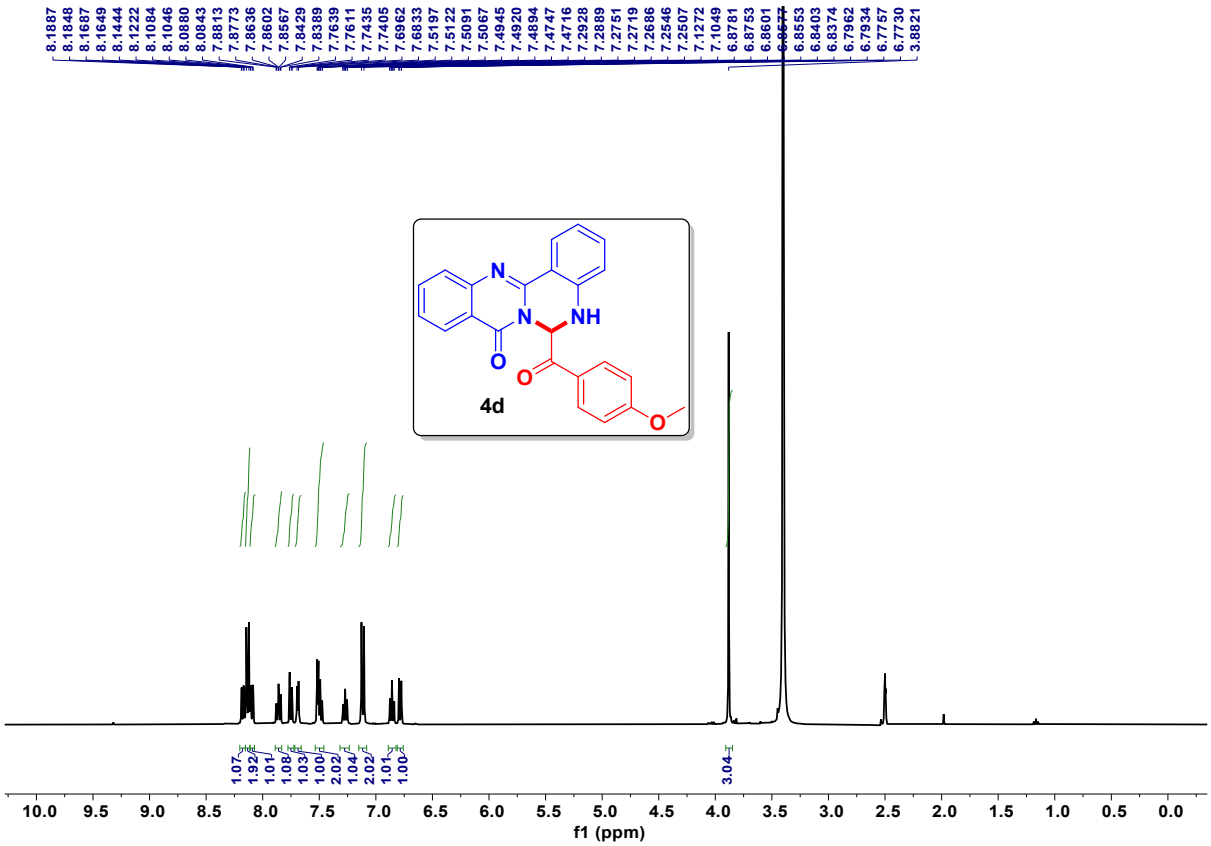


Figure 23. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4d**

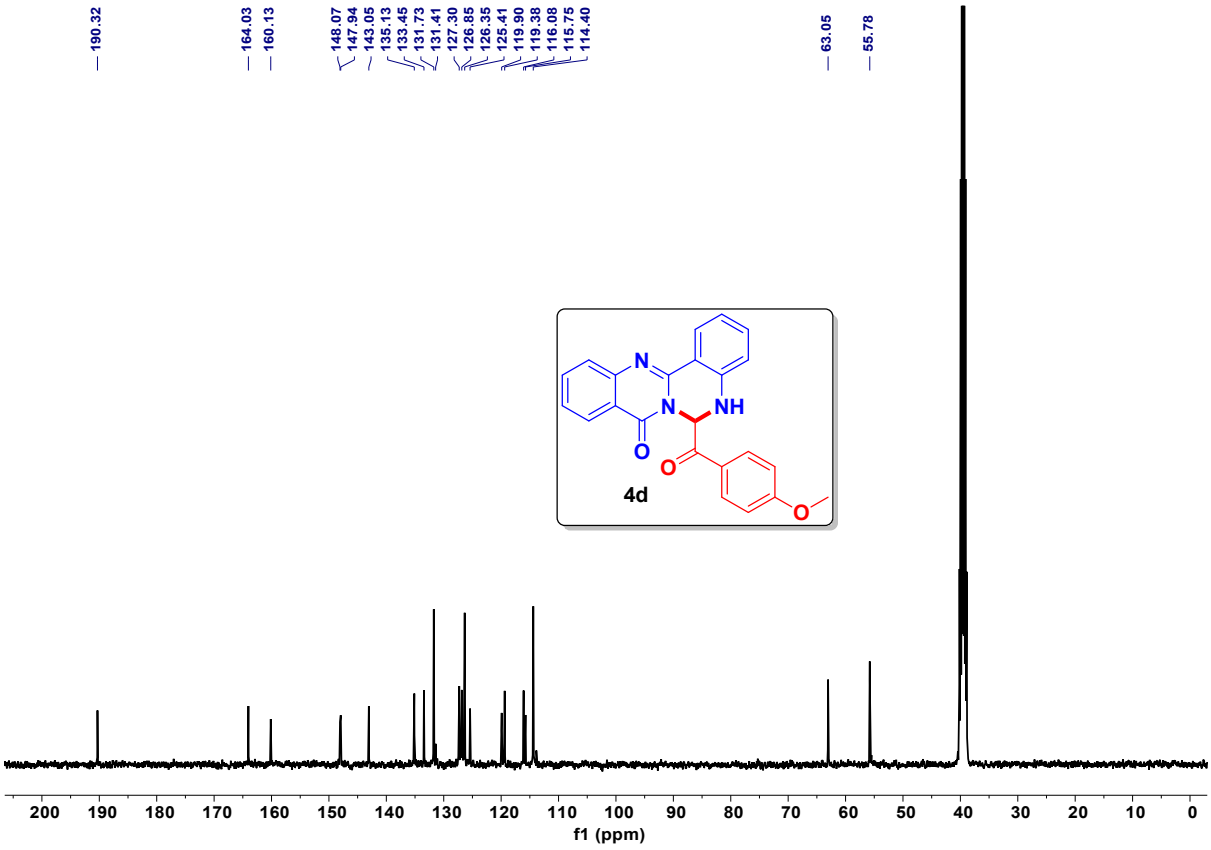


Figure 24. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4d**

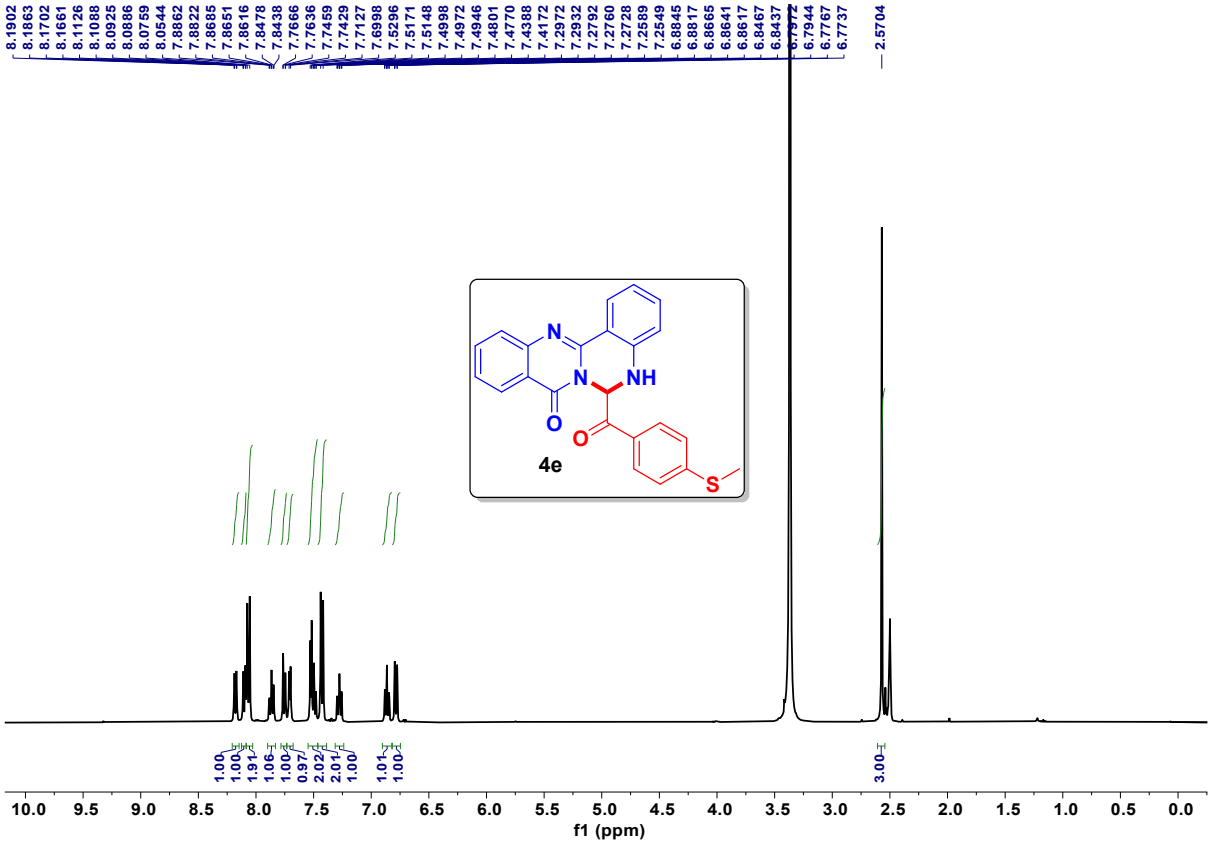


Figure 25. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4e**

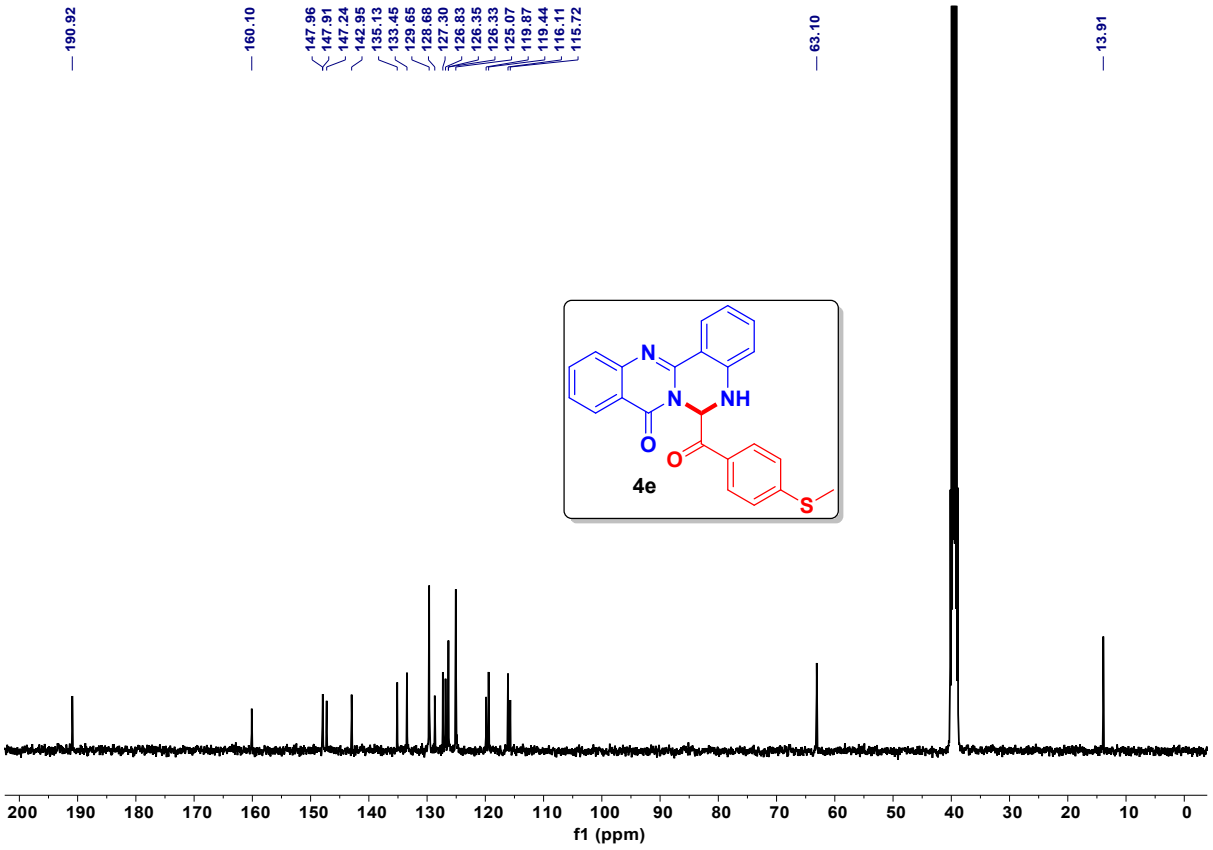


Figure 26. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4e**

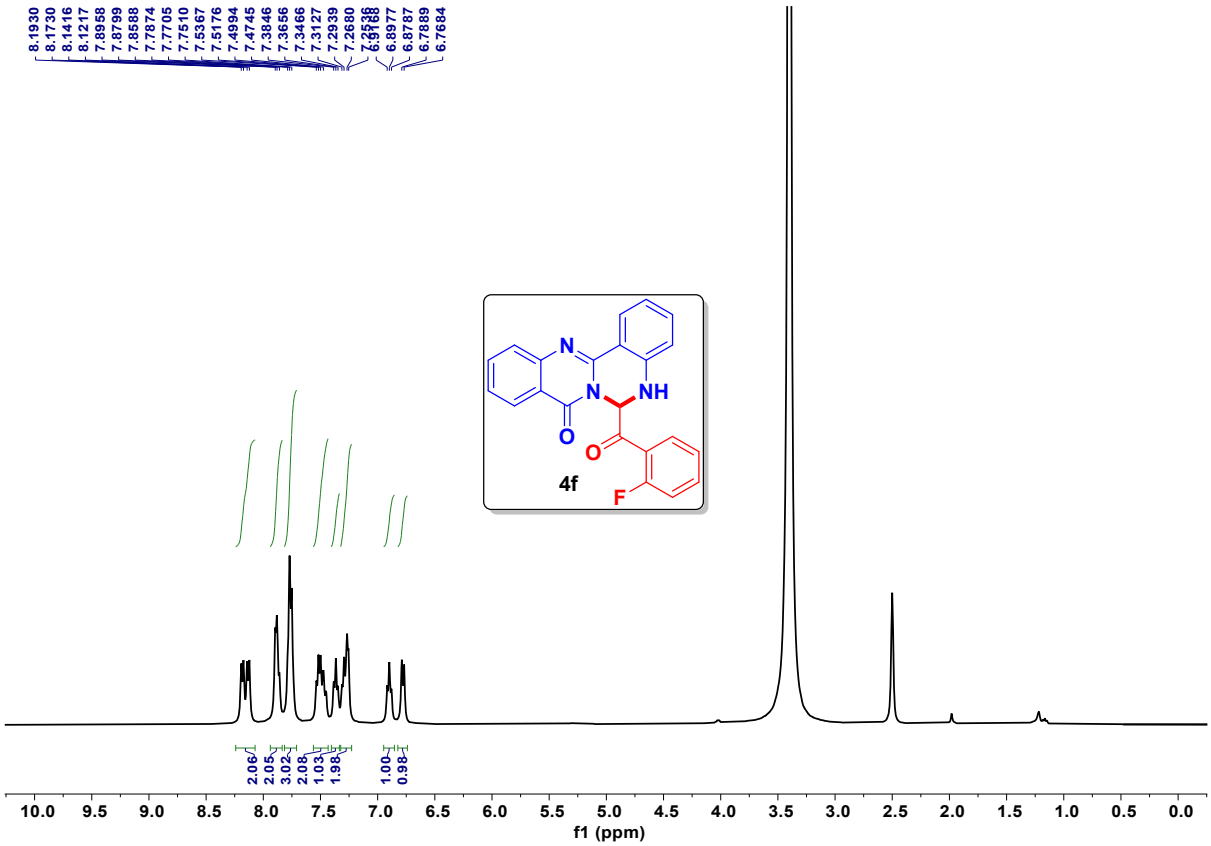


Figure 27. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4f**

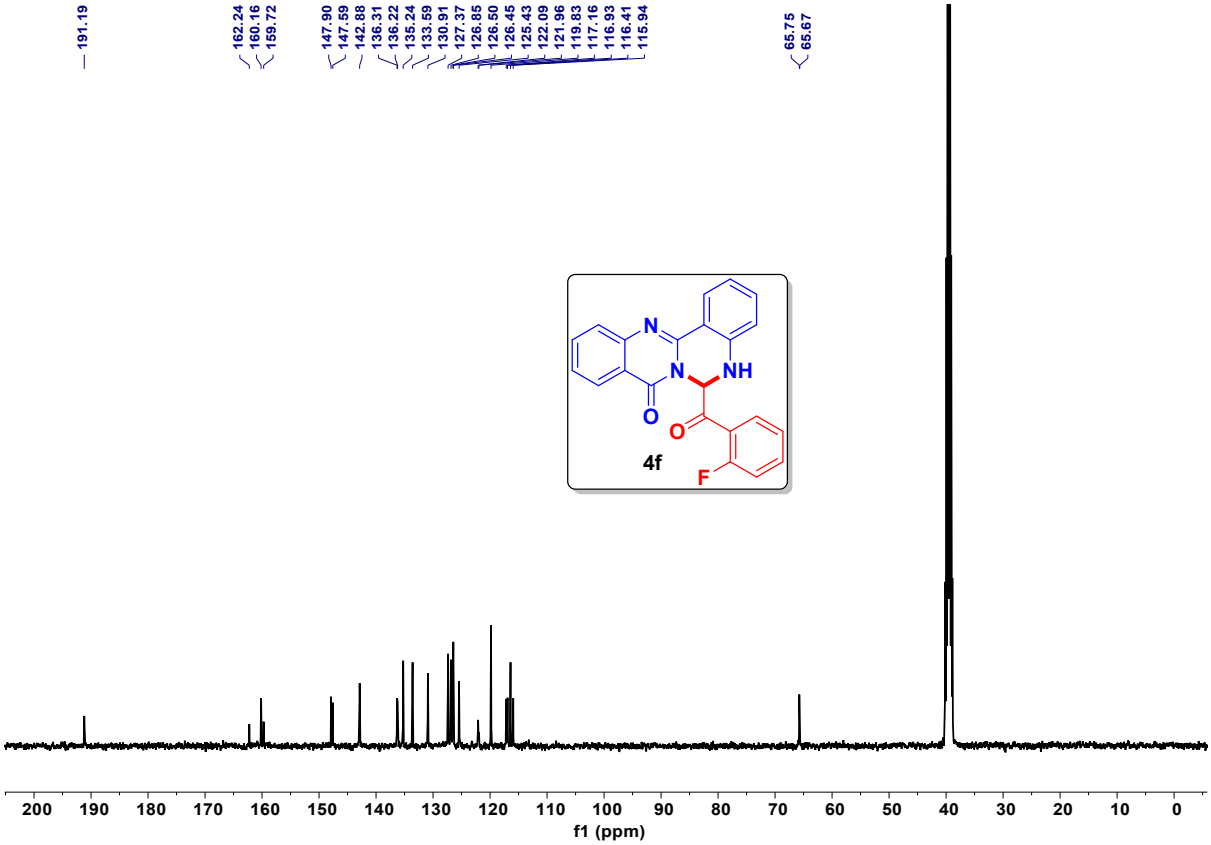


Figure 28. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4f**

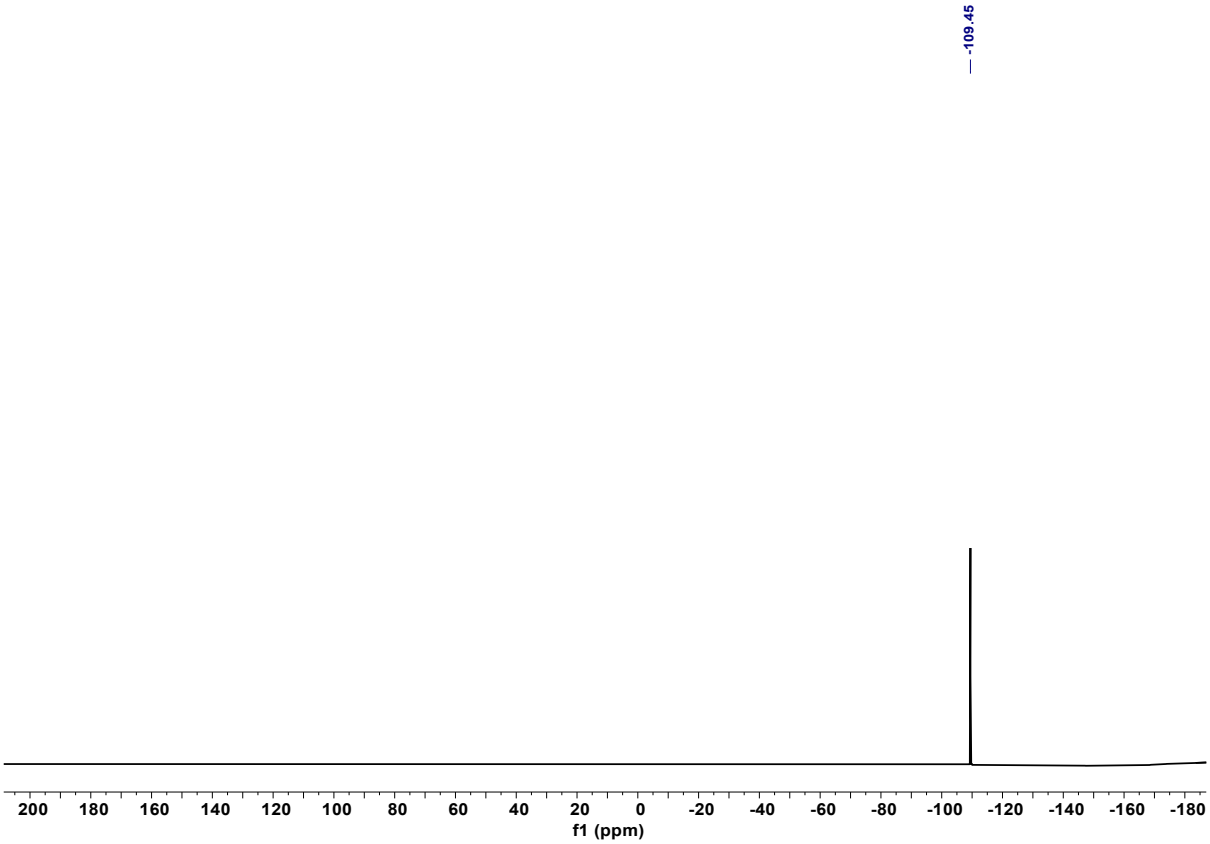


Figure 29. ^{19}F NMR (376 MHz, DMSO-d_6) of compound **4f**

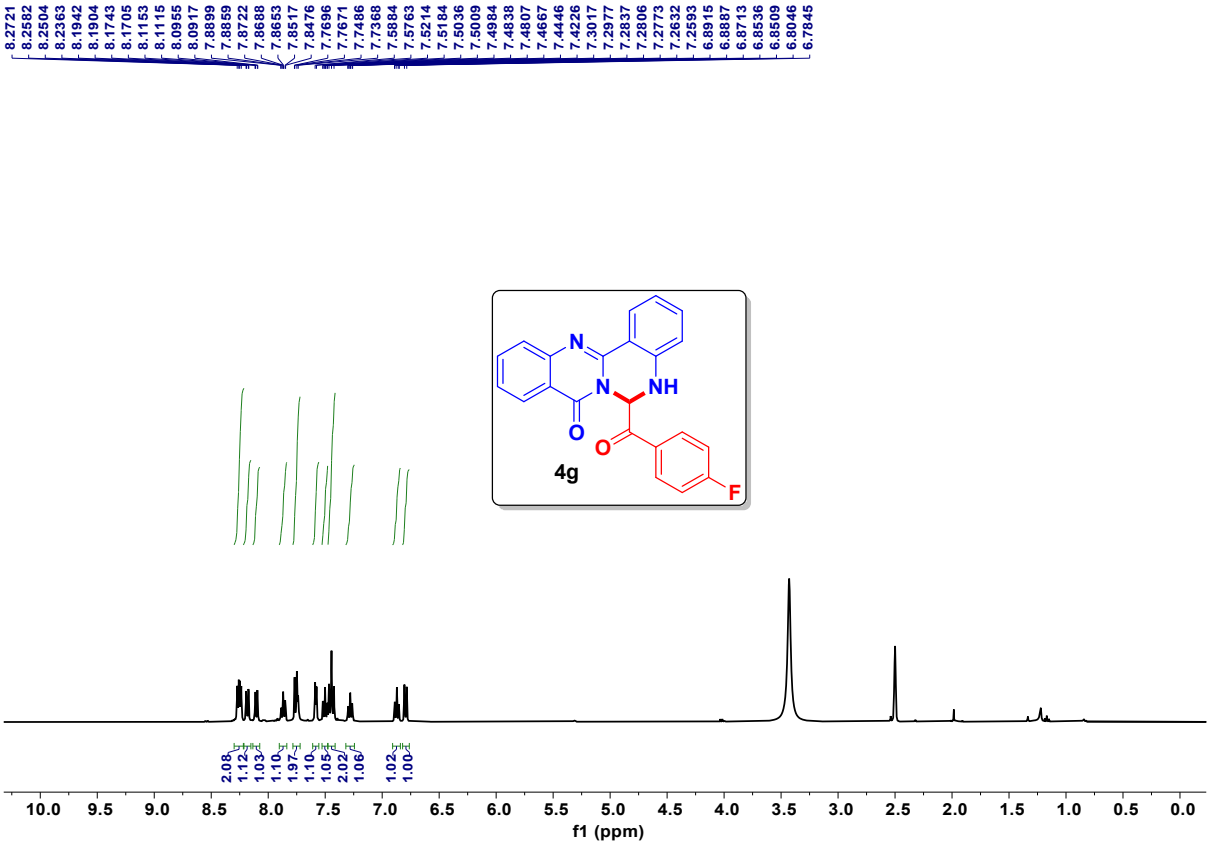


Figure 30. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4g**

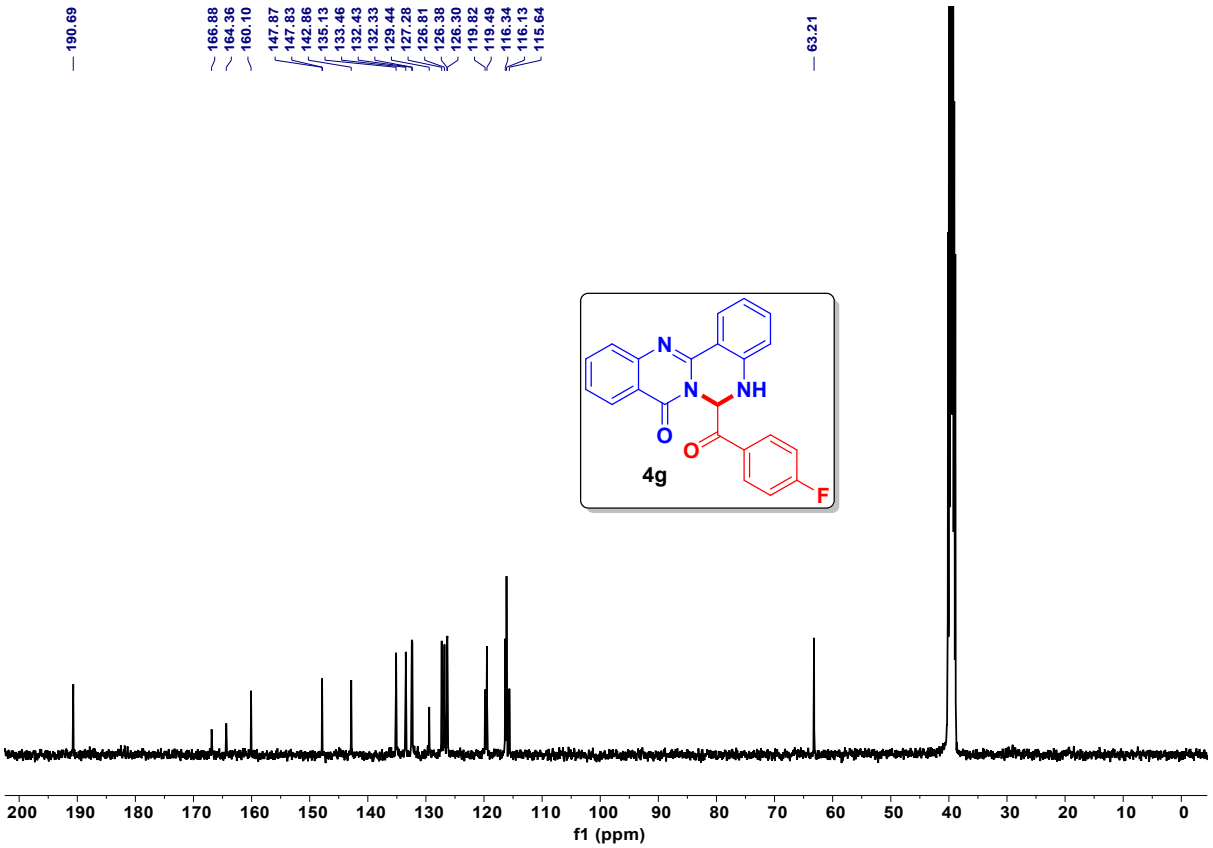


Figure 31. $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6) of compound **4g**

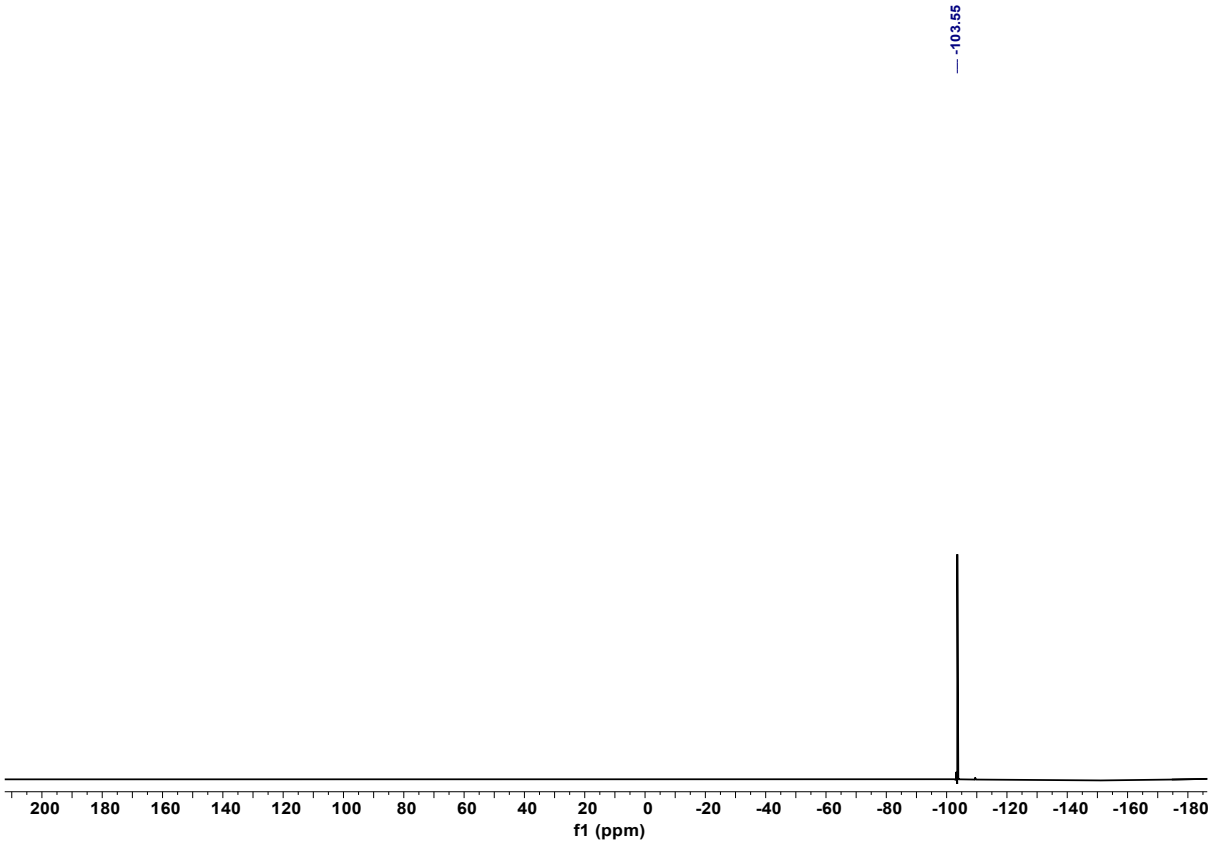


Figure 32. ^{19}F NMR (100 MHz, DMSO-d_6) of compound **4g**

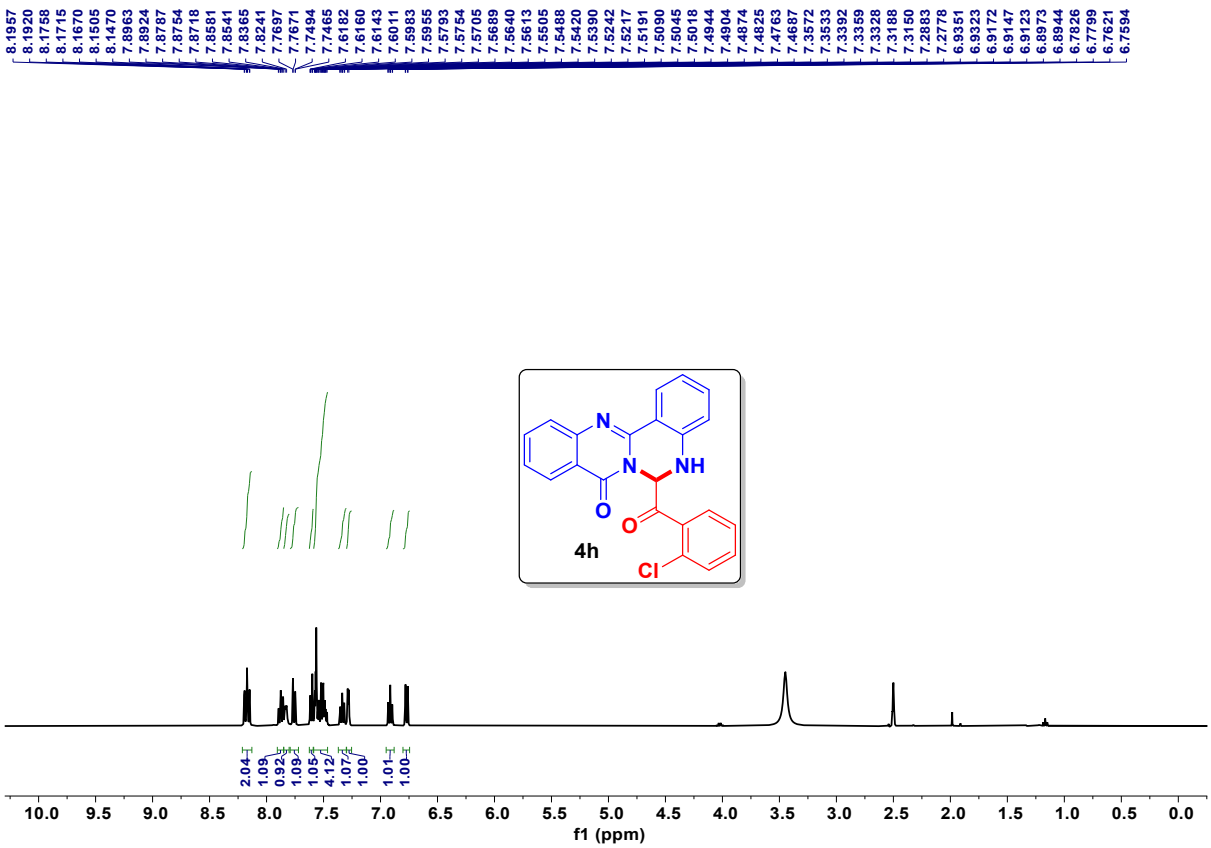


Figure 33. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4h**

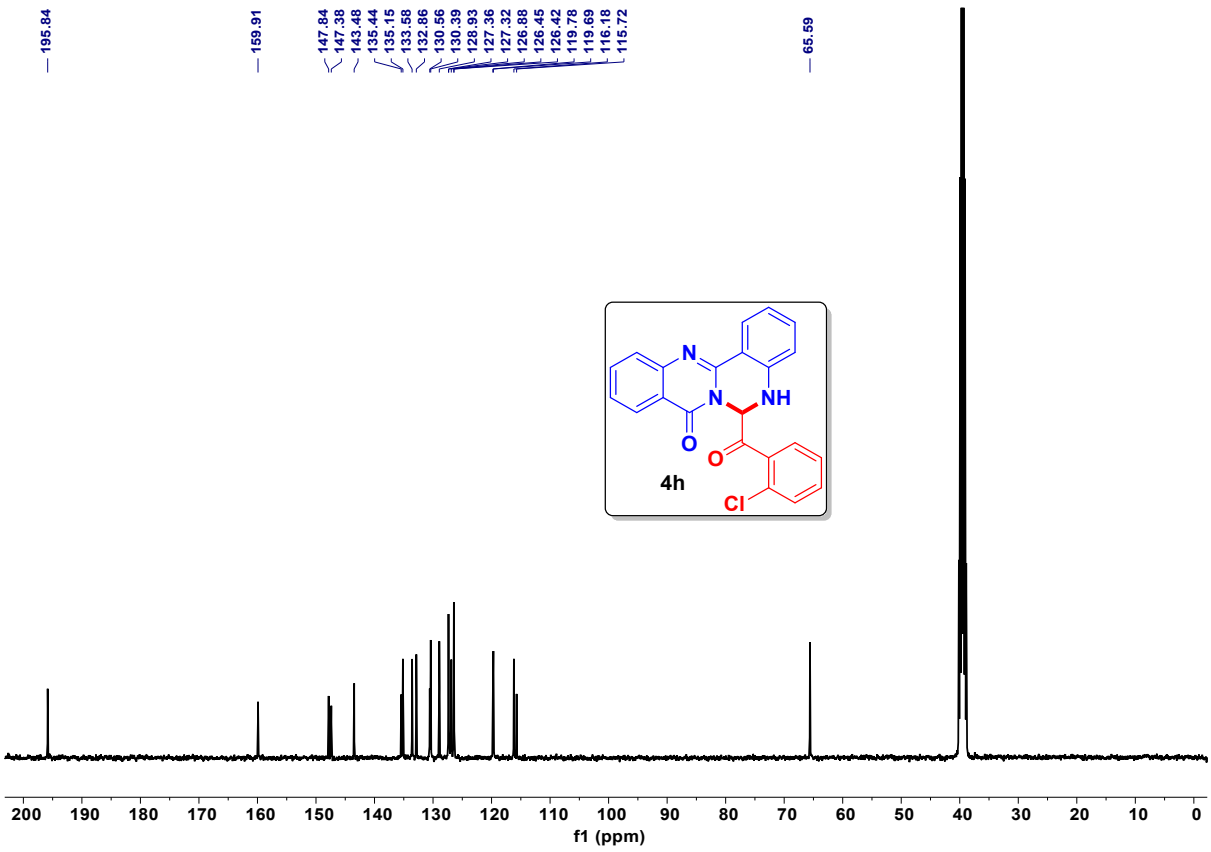


Figure 34. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4h**

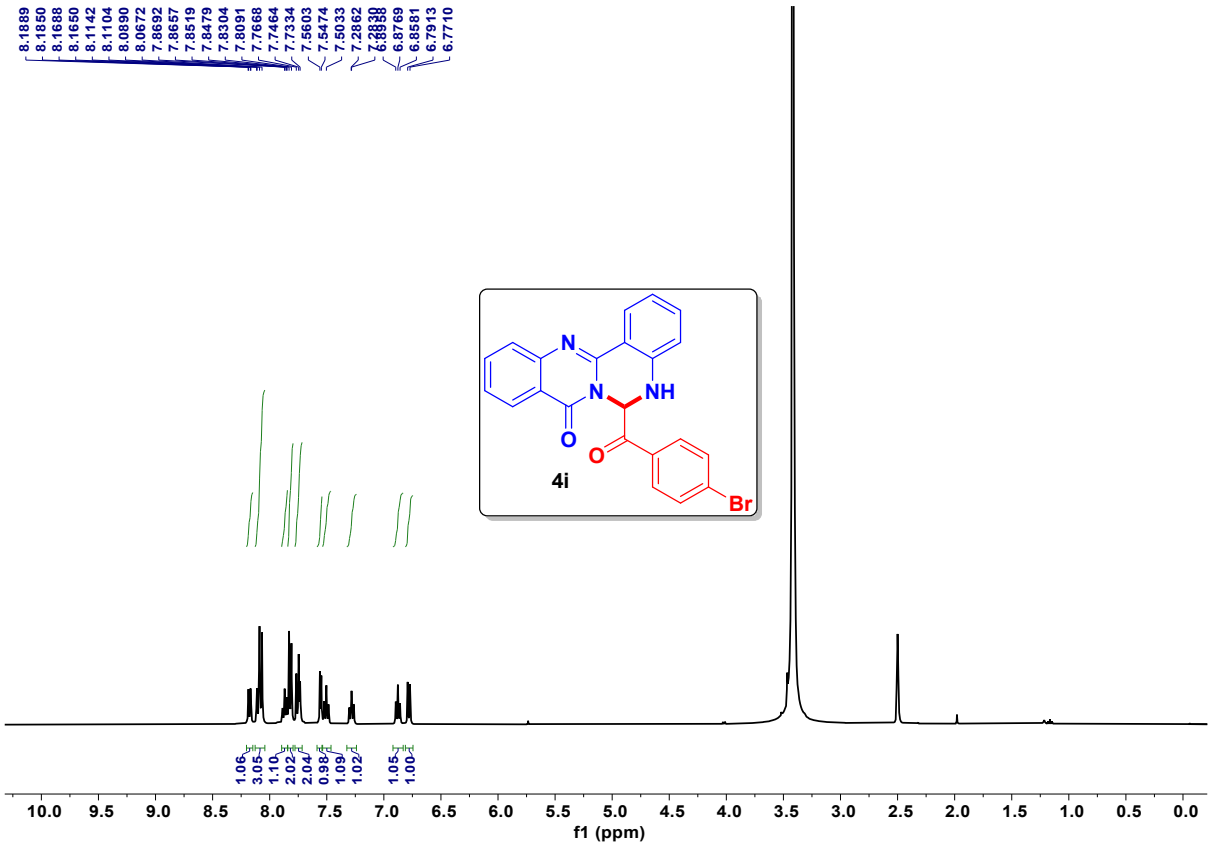


Figure 35. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4i**

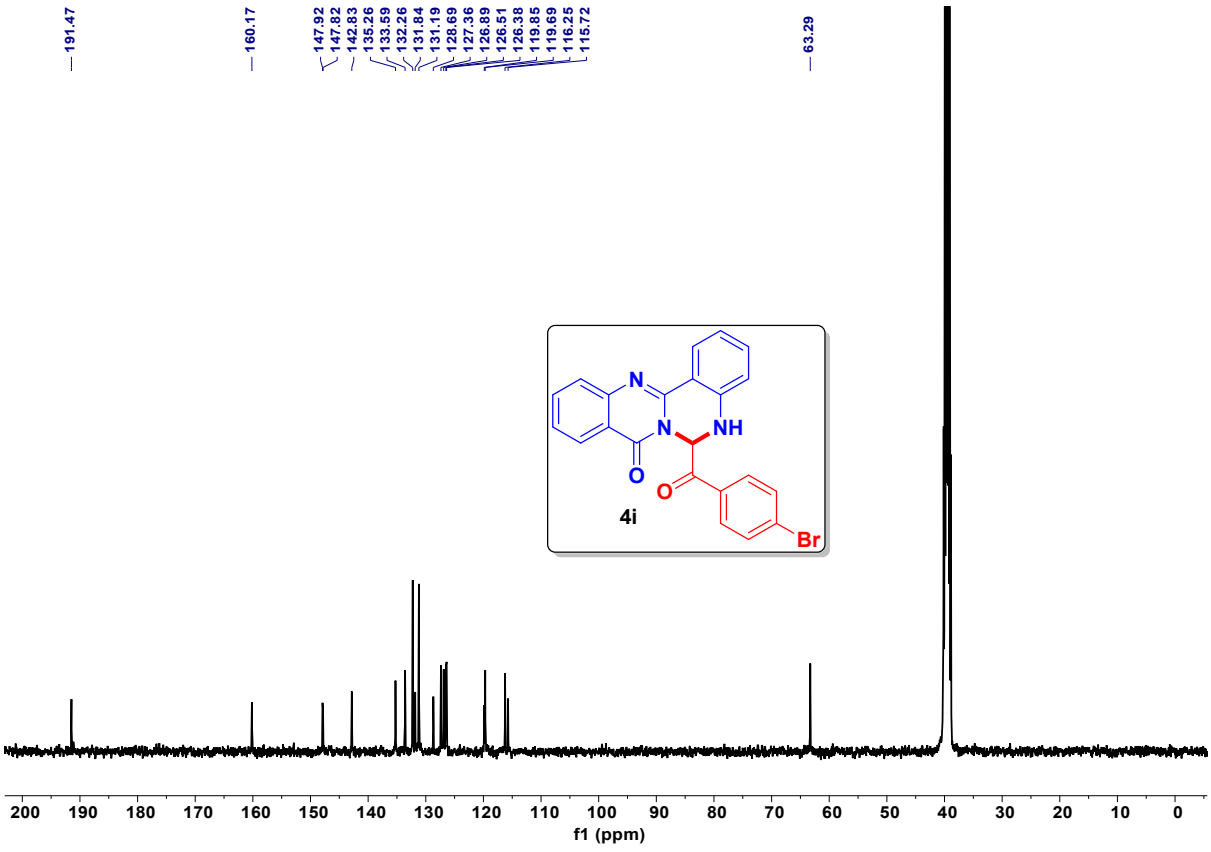


Figure 36. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4i**

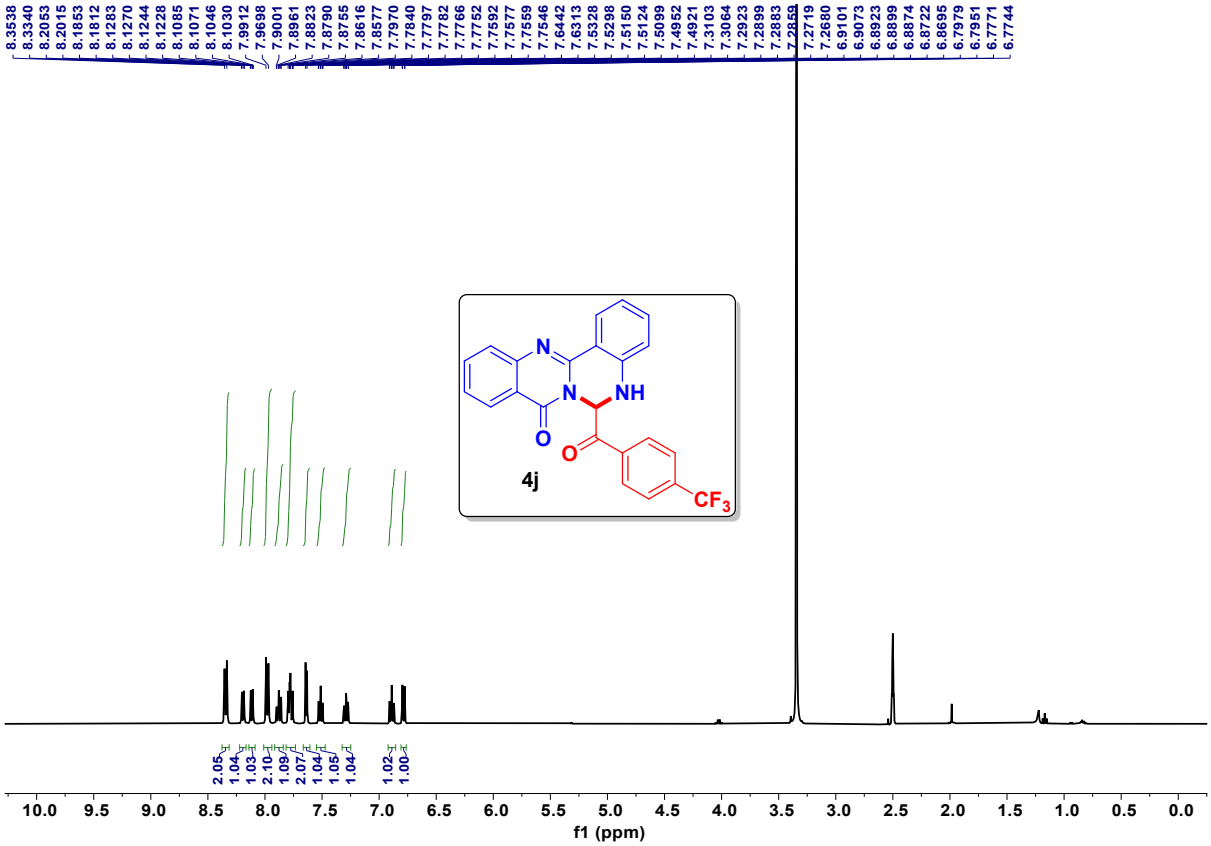


Figure 37. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4j**

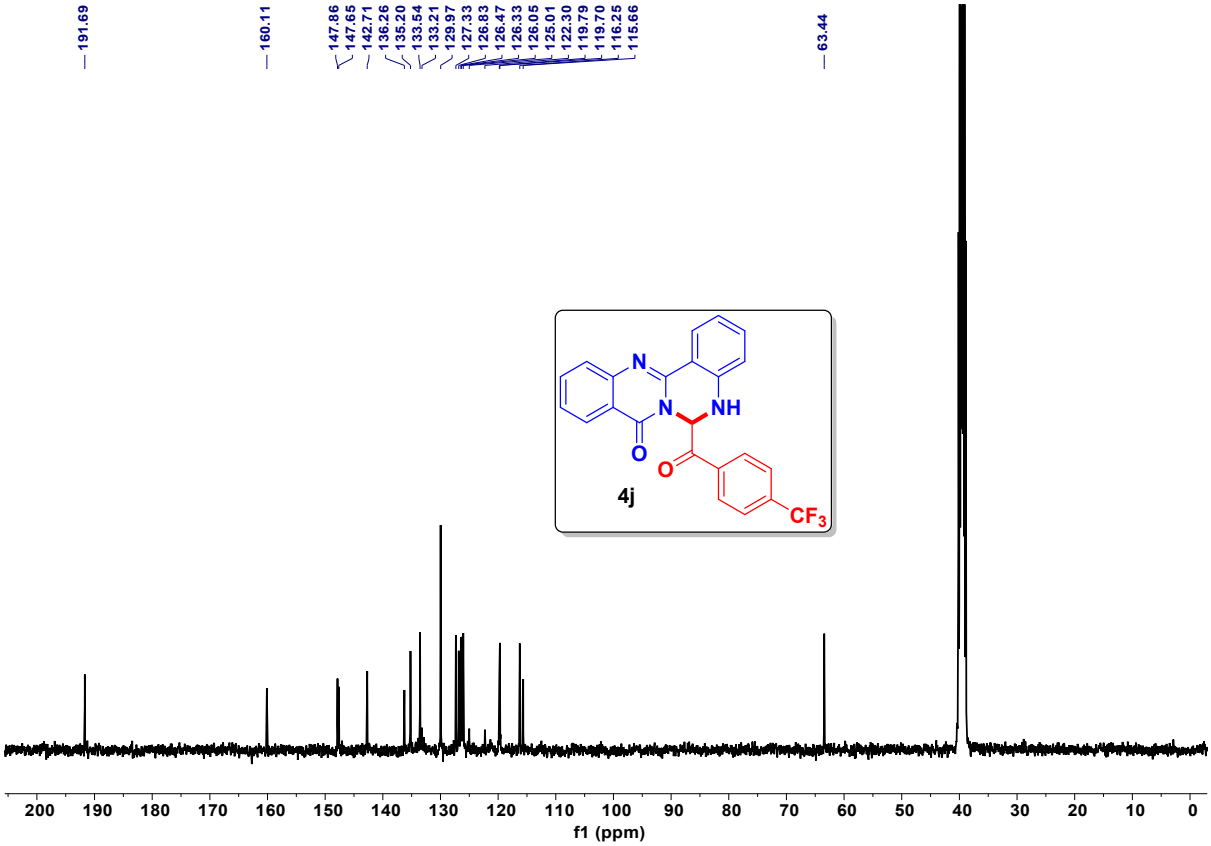


Figure 38. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4j**

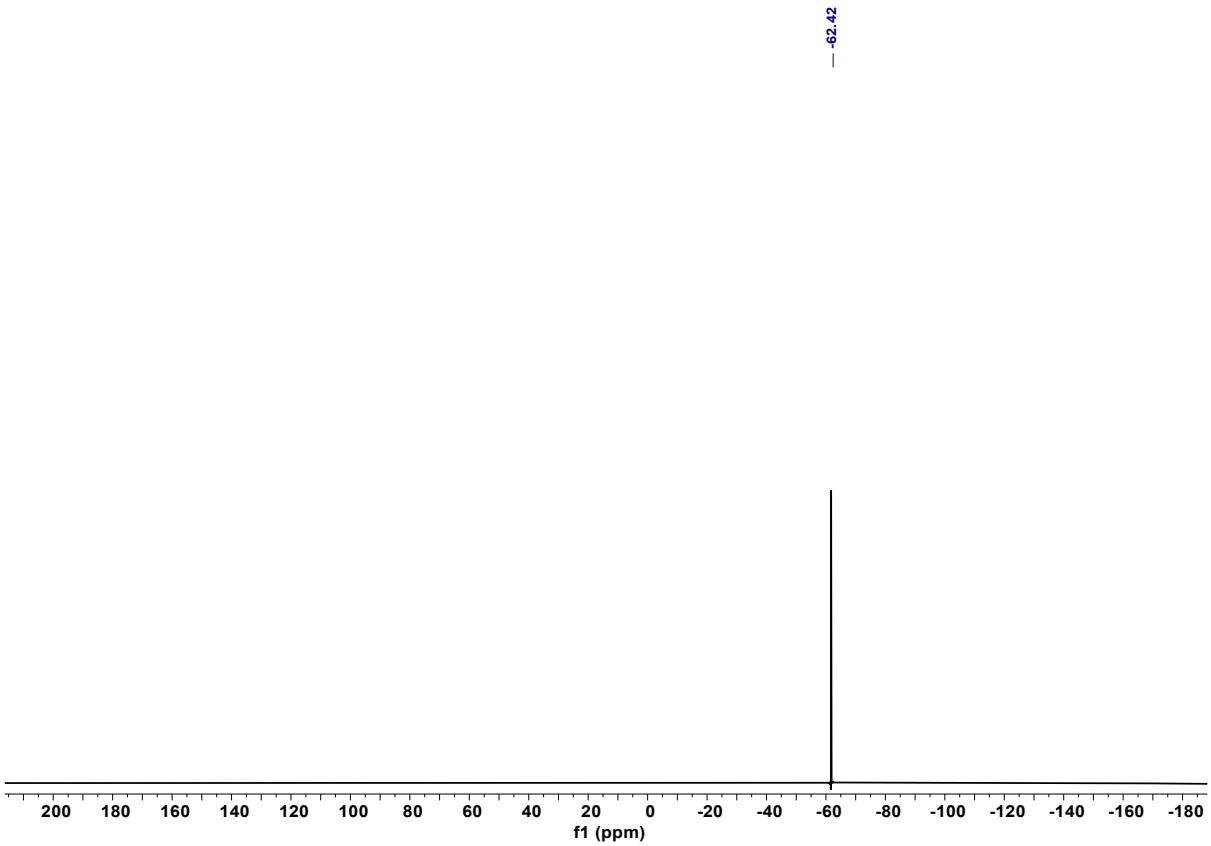


Figure 39. ^{19}F NMR (376 MHz, DMSO-d_6) of compound **4j**

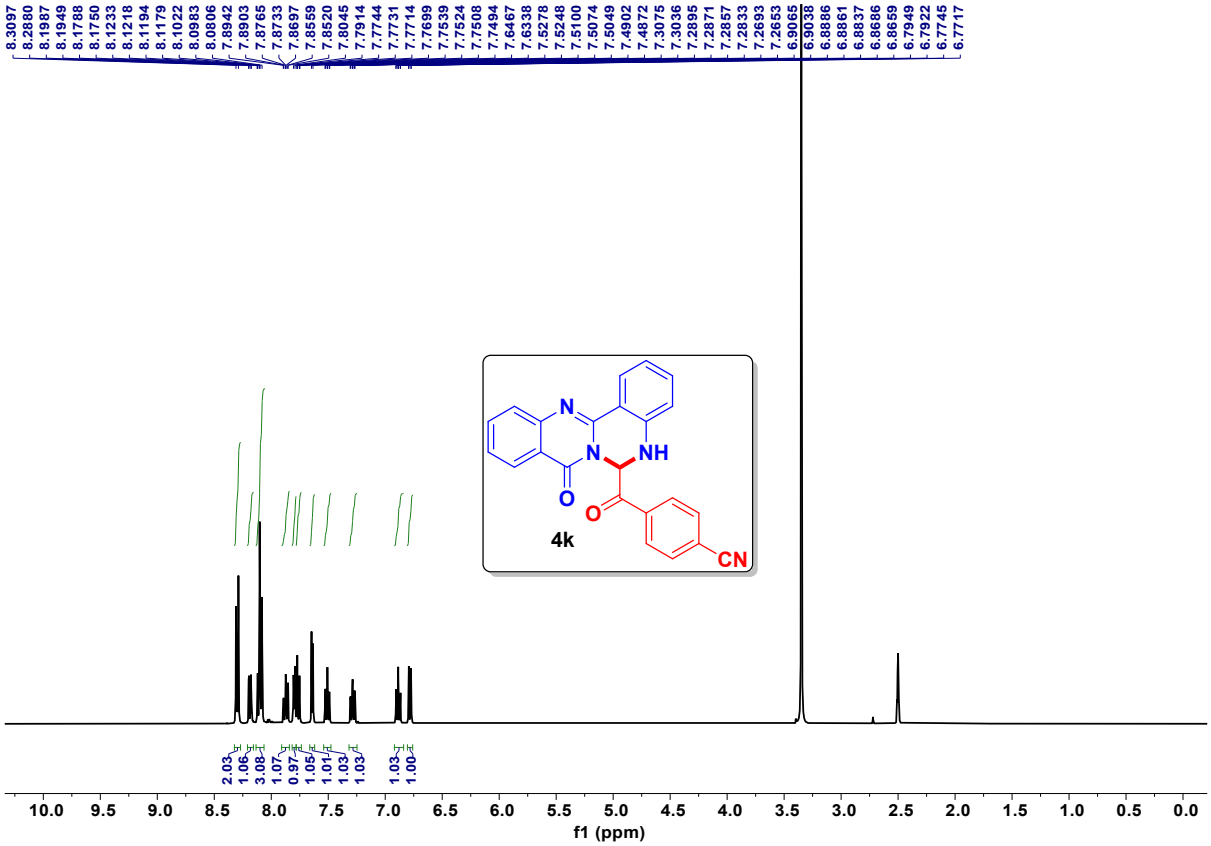


Figure 40. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4k**

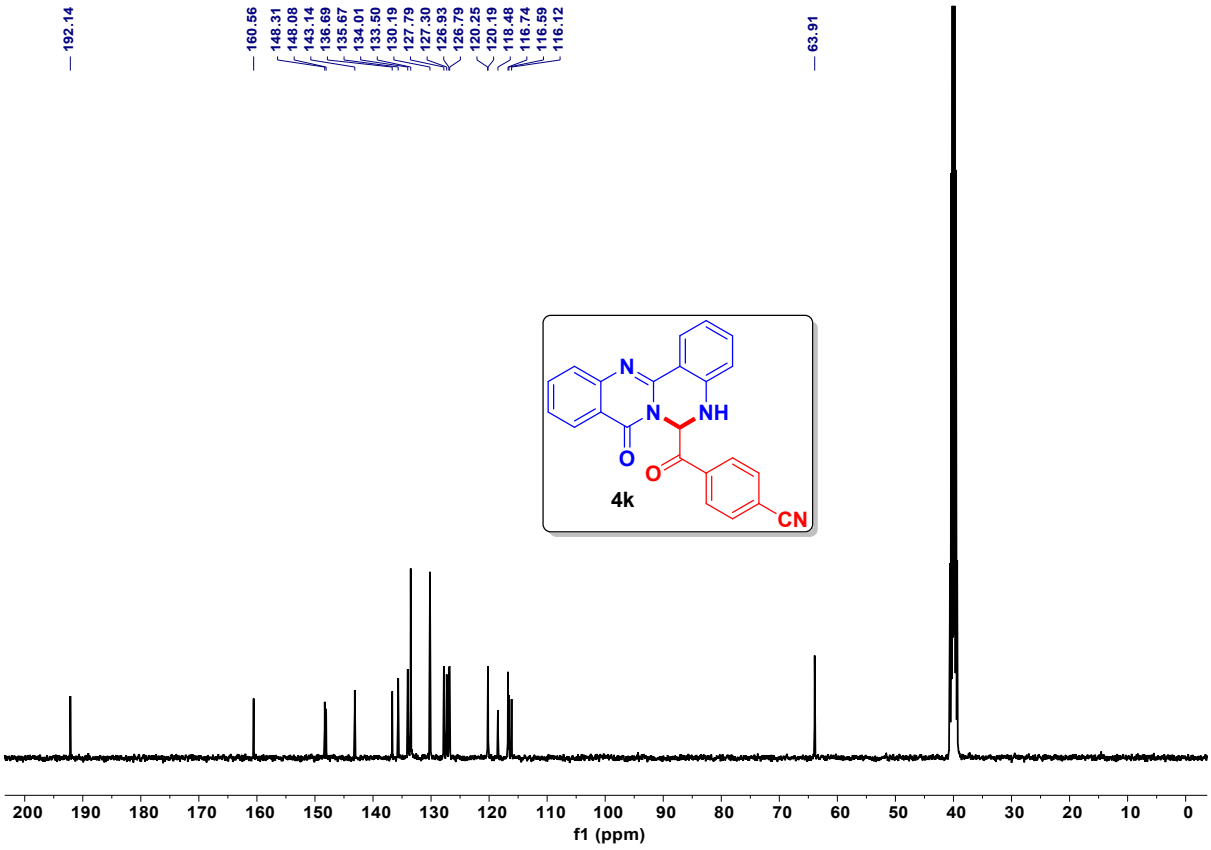


Figure 41. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4k**

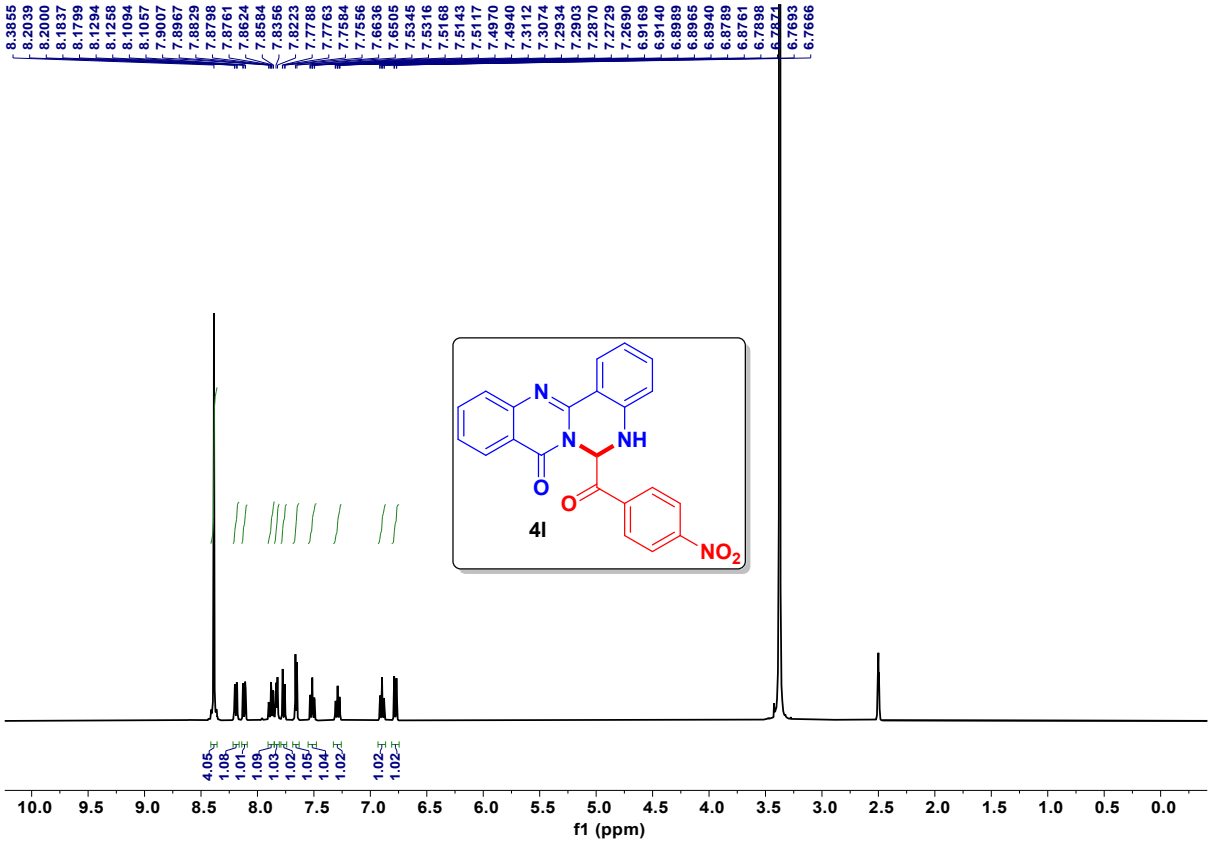


Figure 42. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4l**

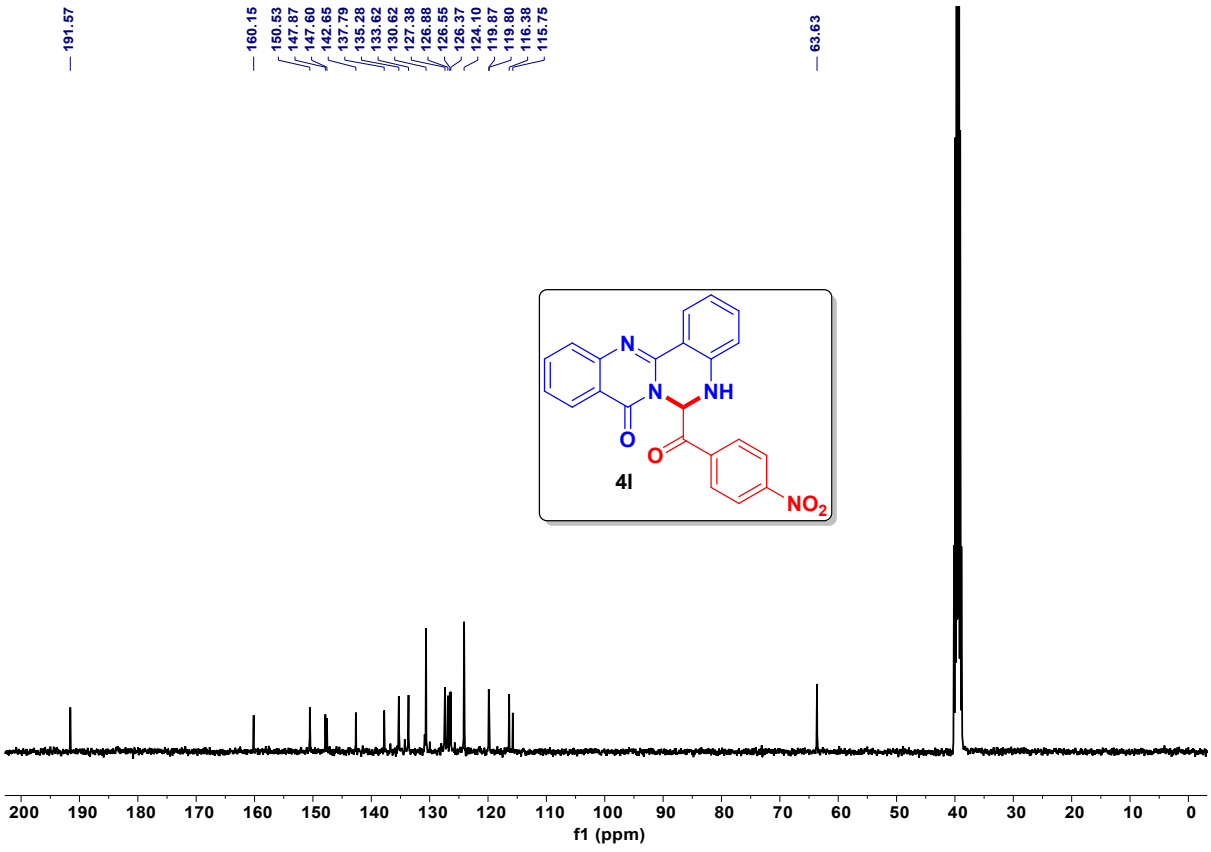


Figure 43. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4l**

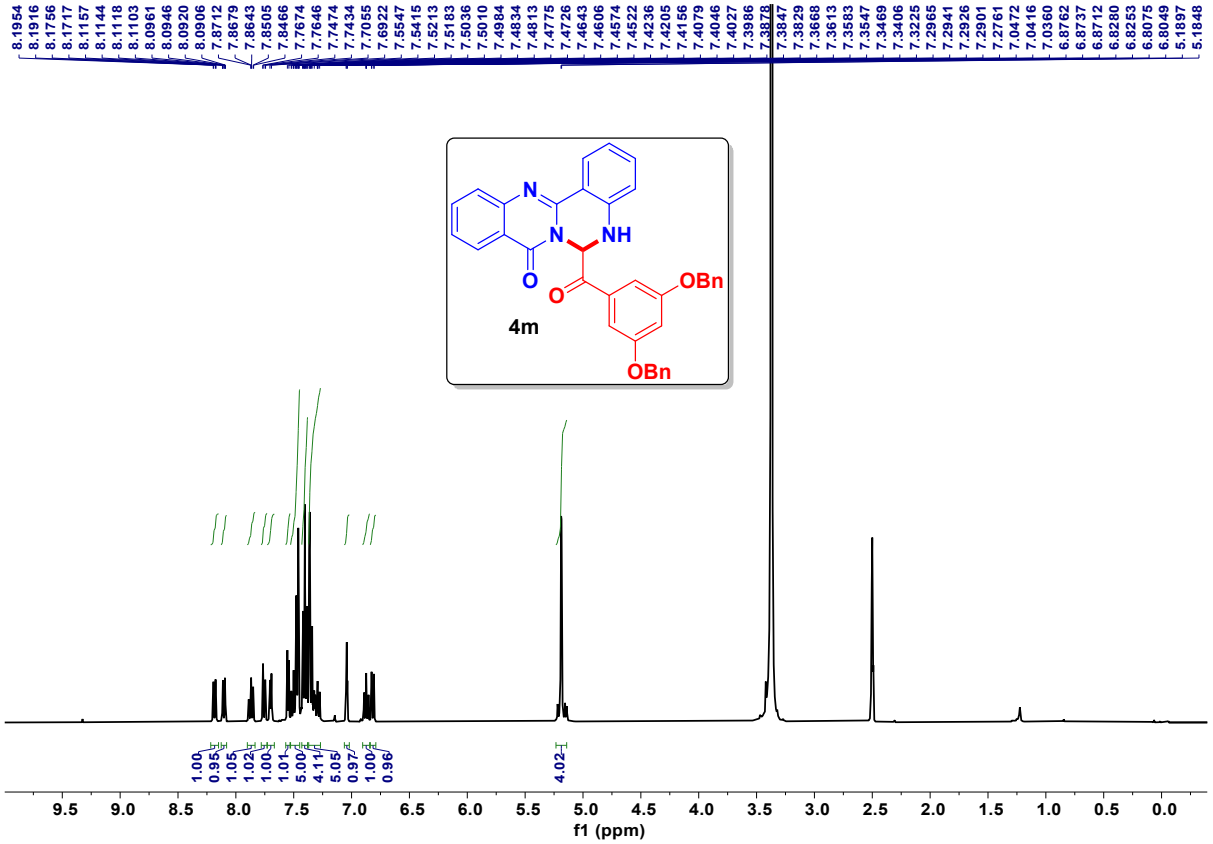


Figure 44. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4m**

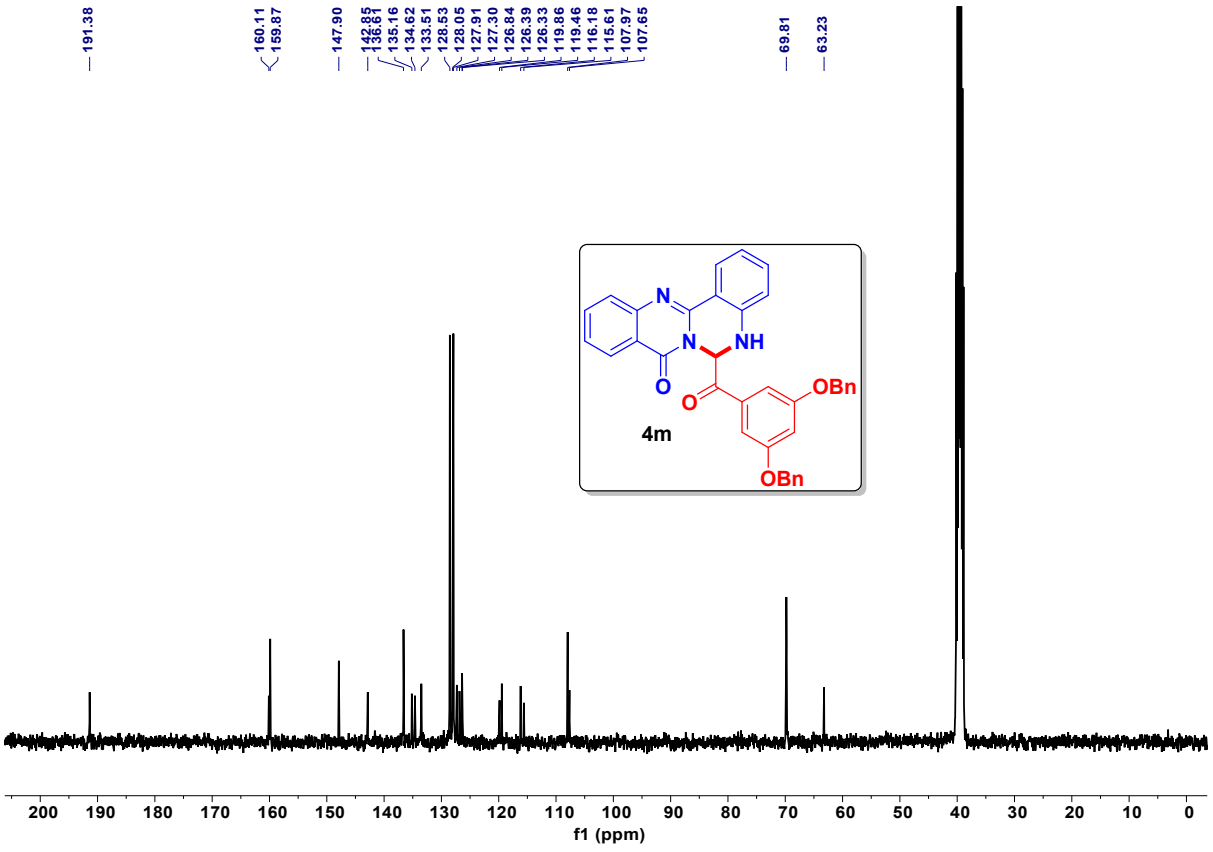


Figure 45. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4m**

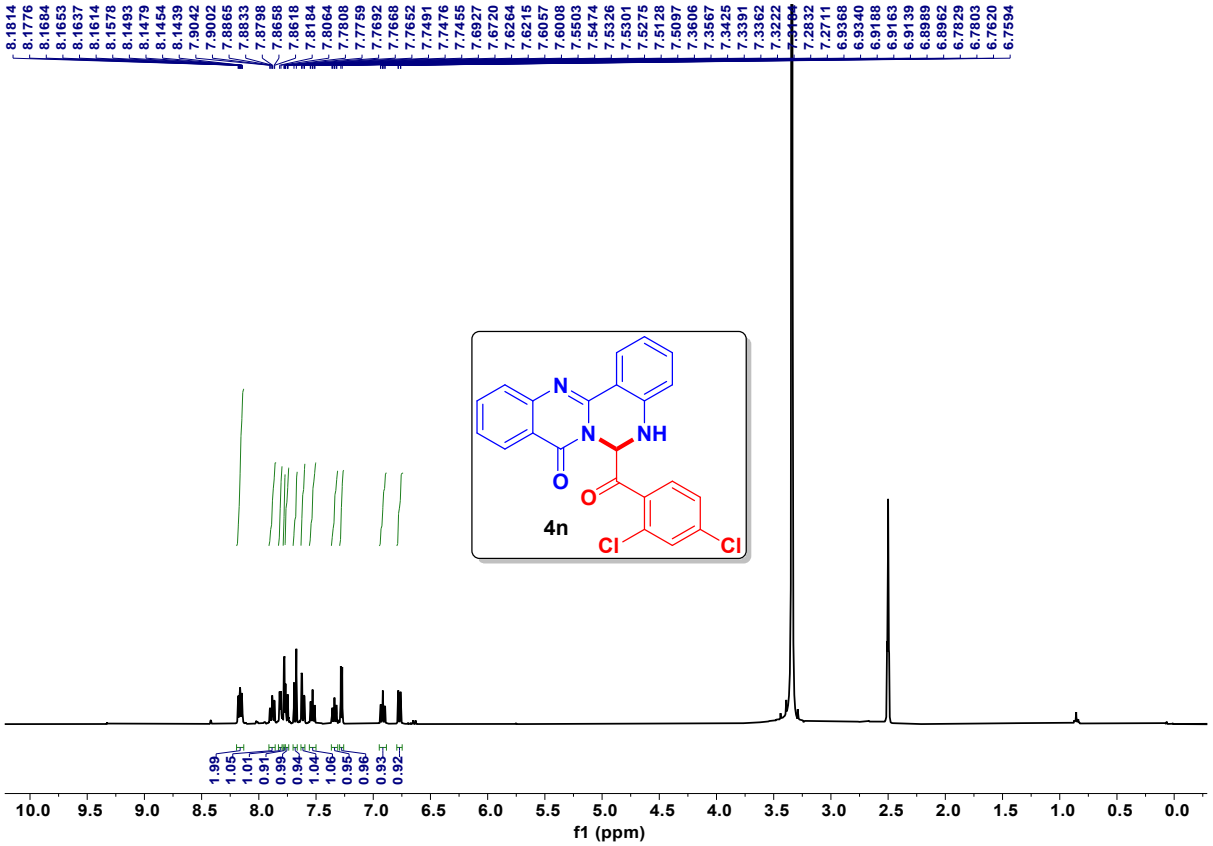


Figure 46. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4n**

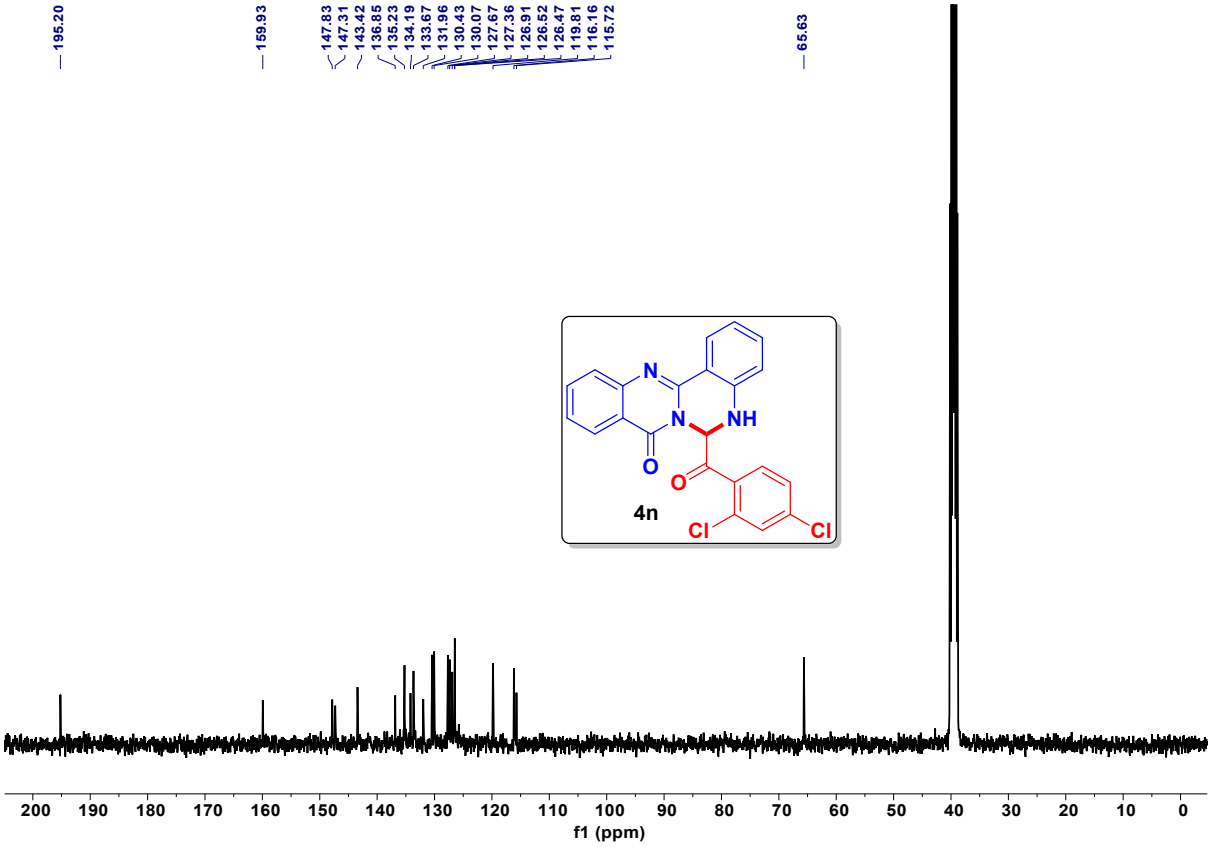


Figure 47. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4n**

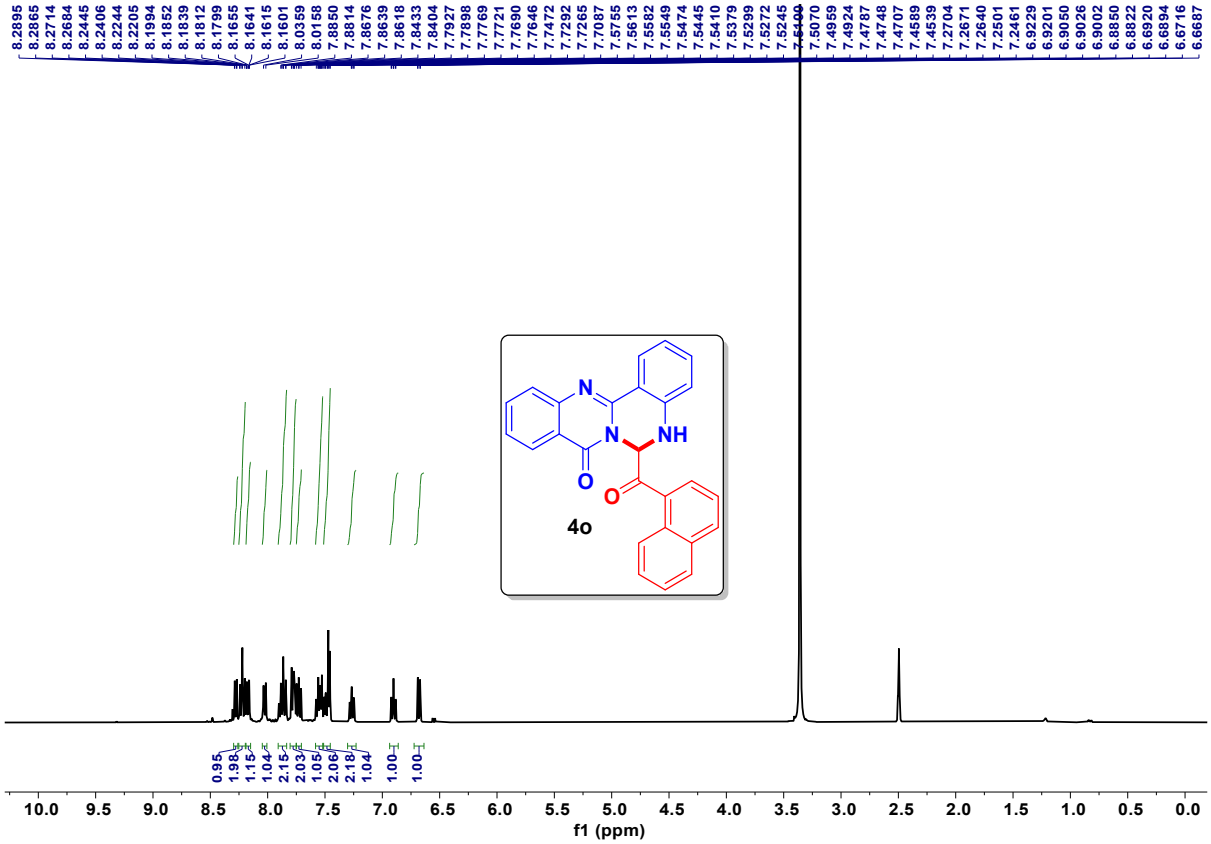


Figure 48. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4o**

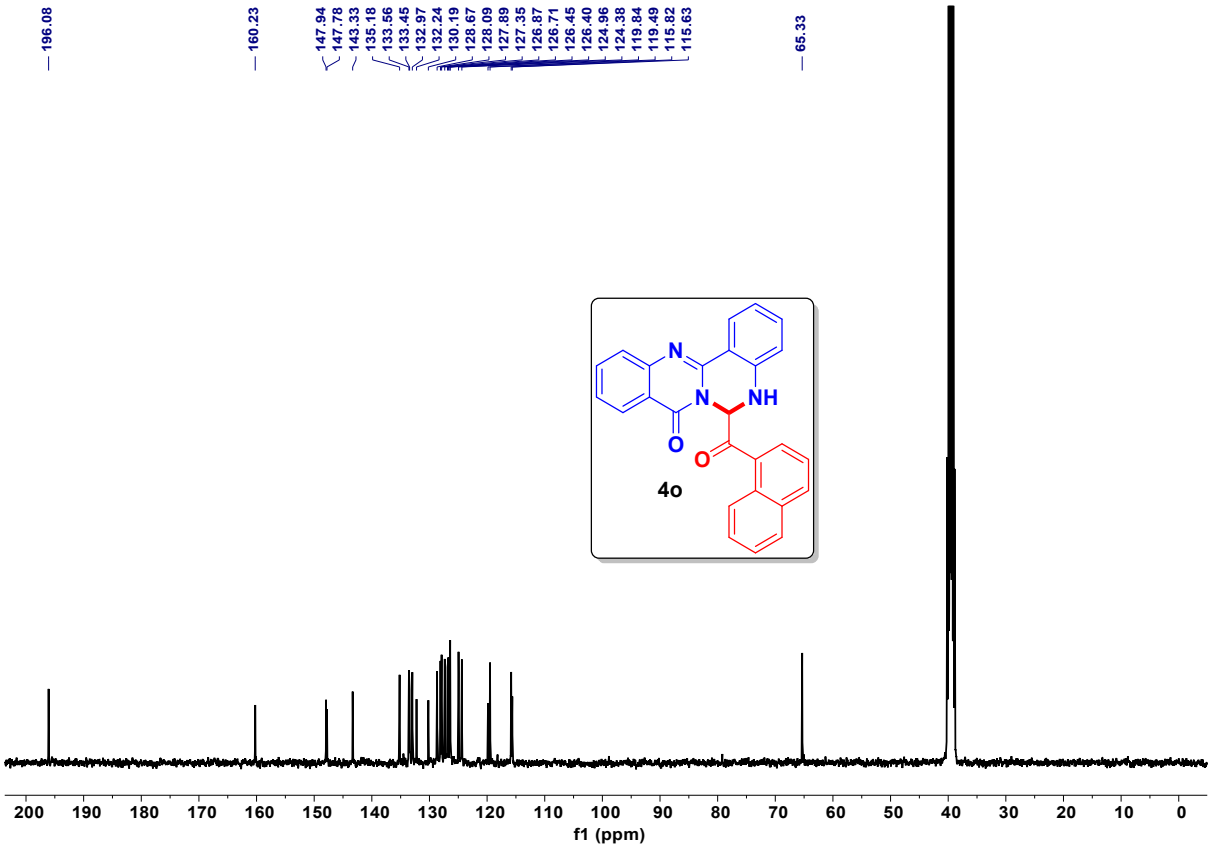


Figure 49. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4o**

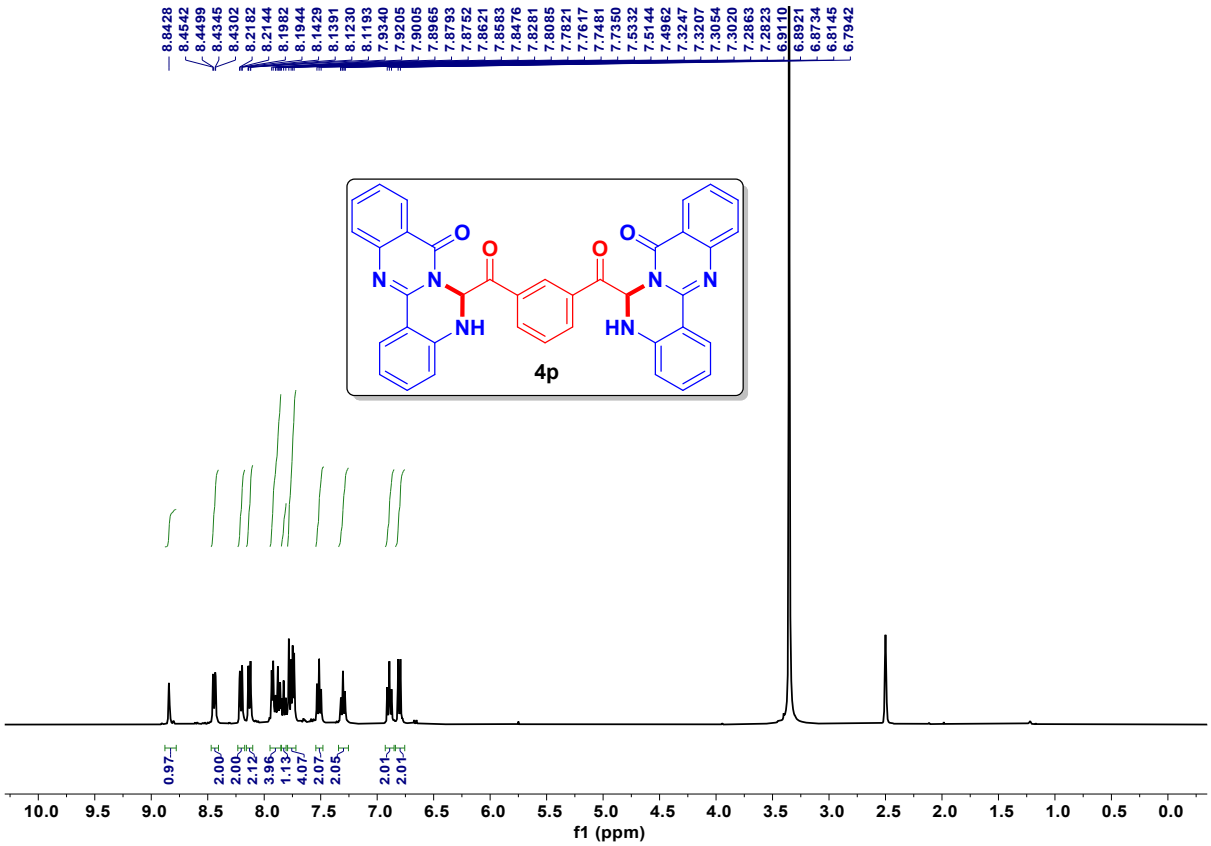


Figure 50. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4p**

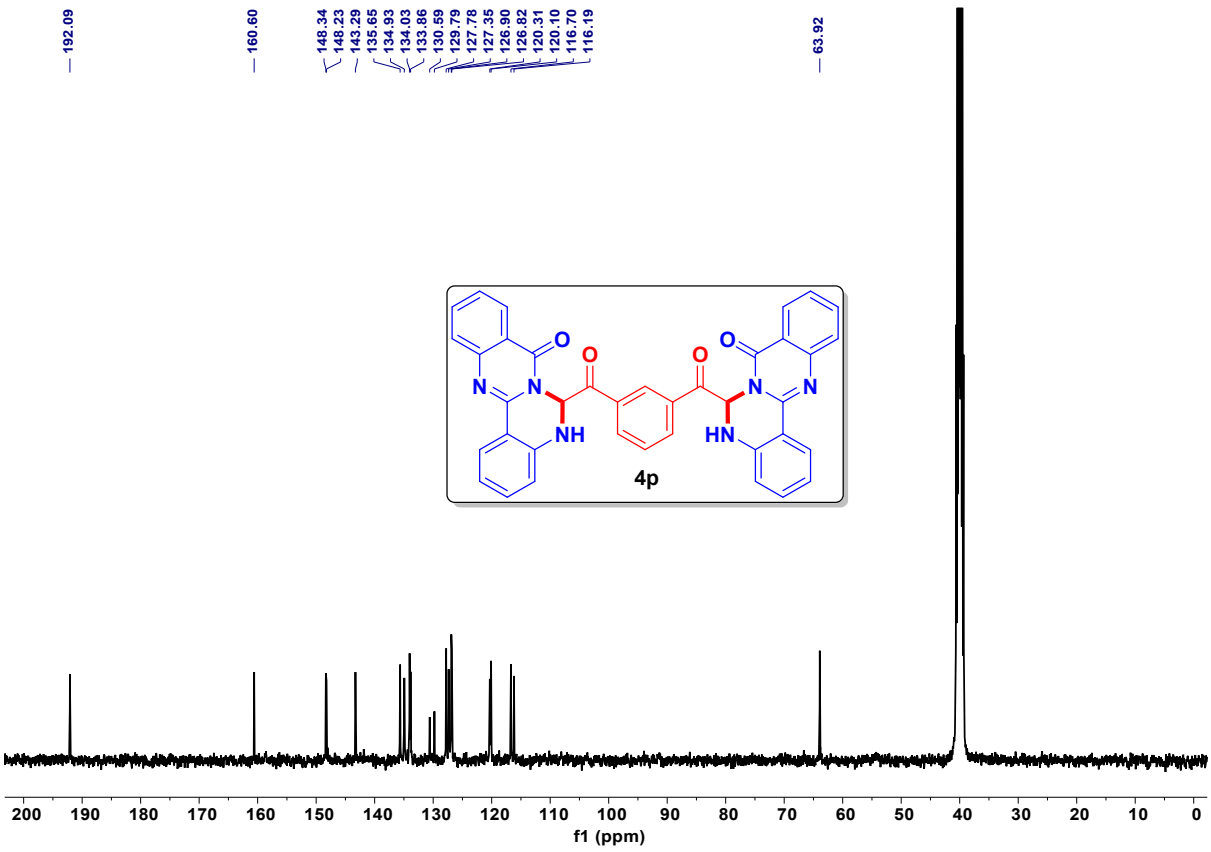


Figure 51. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4p**

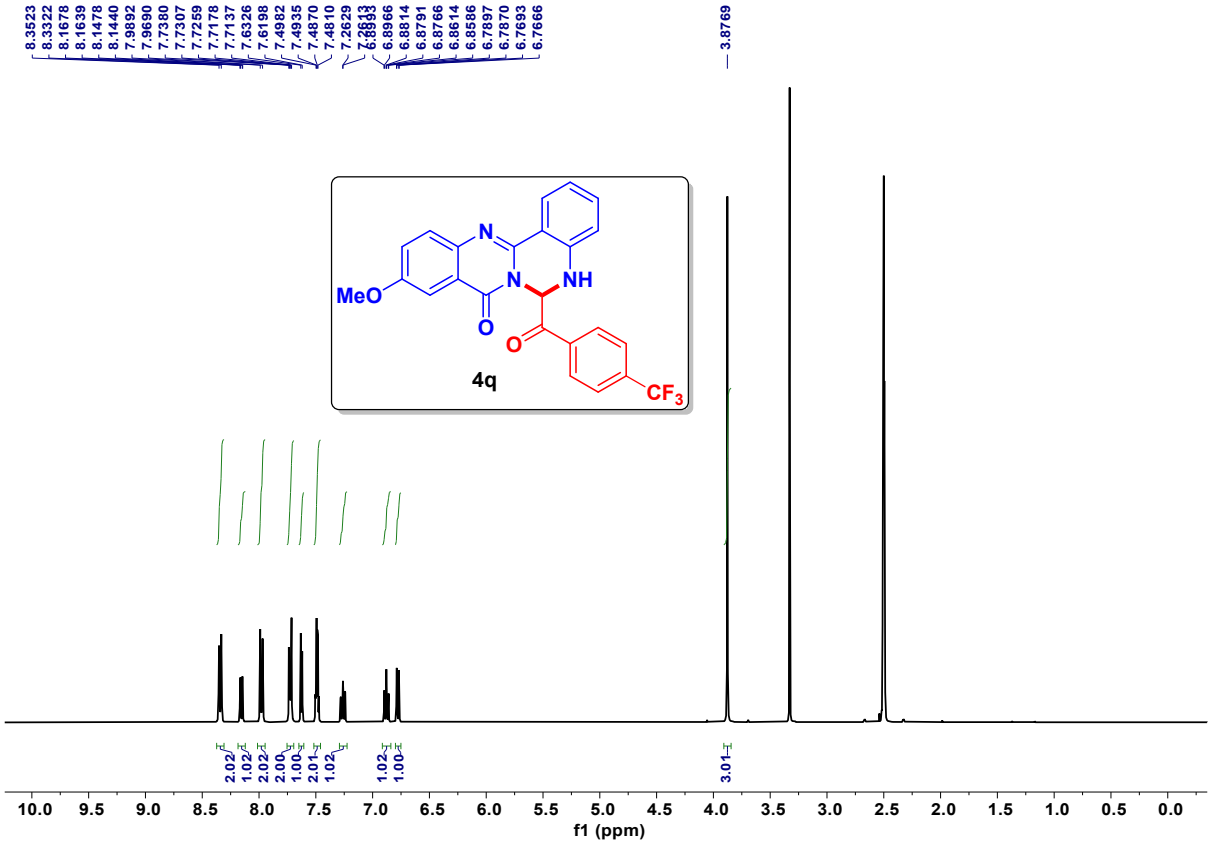


Figure 52. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4q**

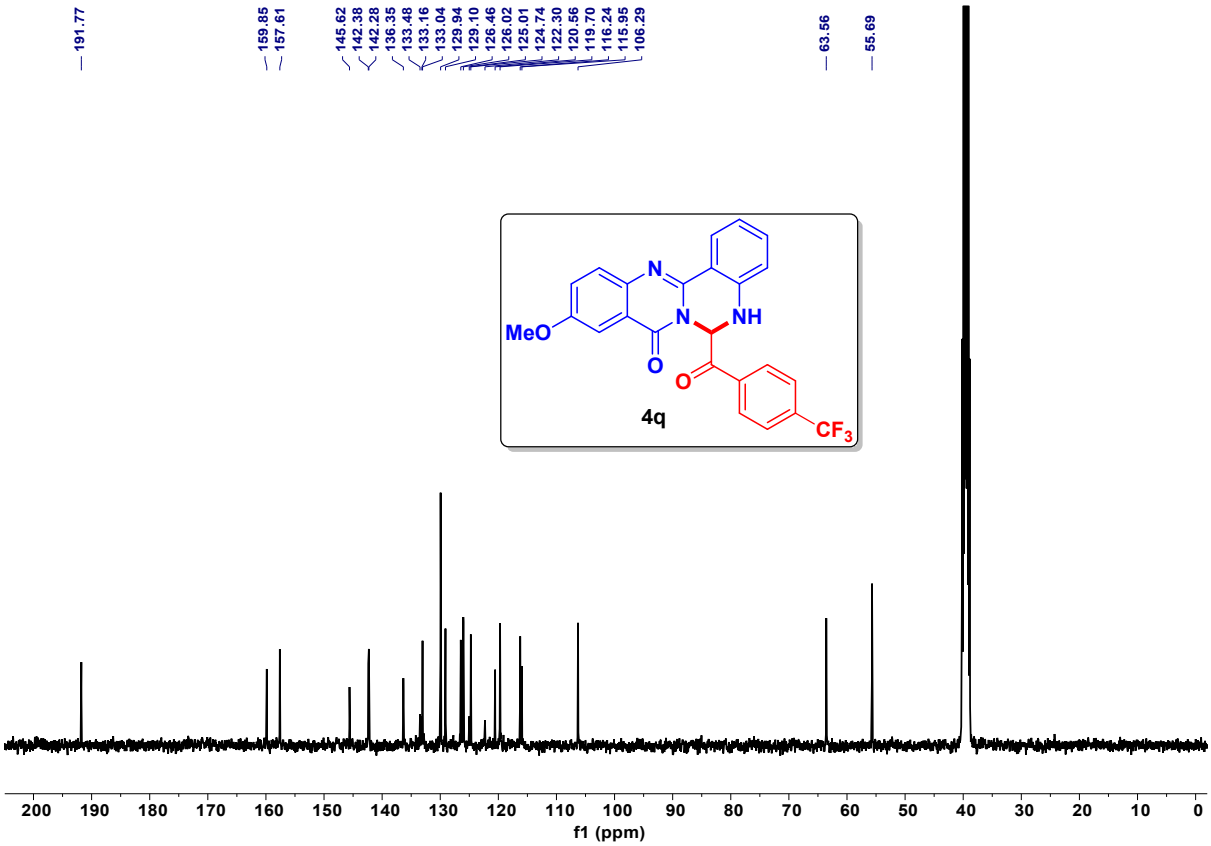


Figure 53. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4q**

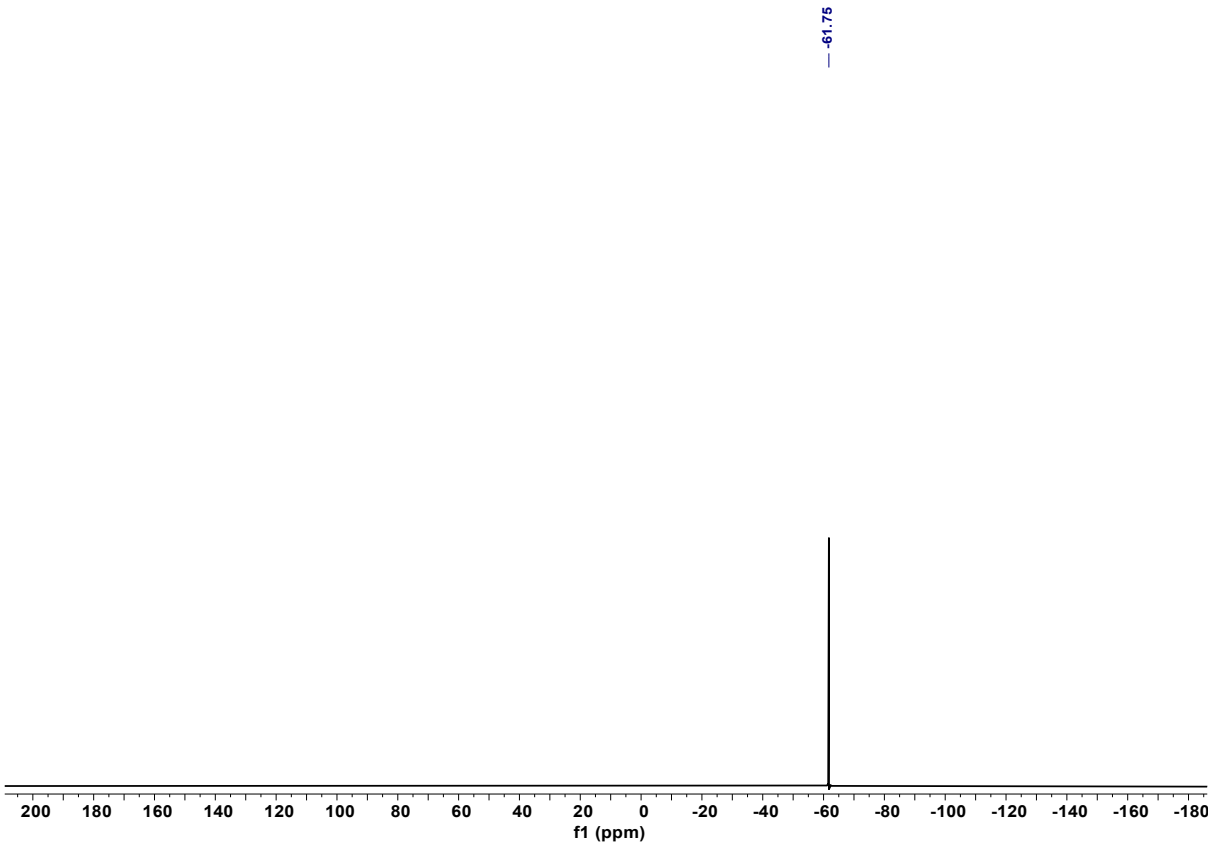


Figure 54. ^{19}F NMR (376 MHz, DMSO-d_6) of compound **4q**

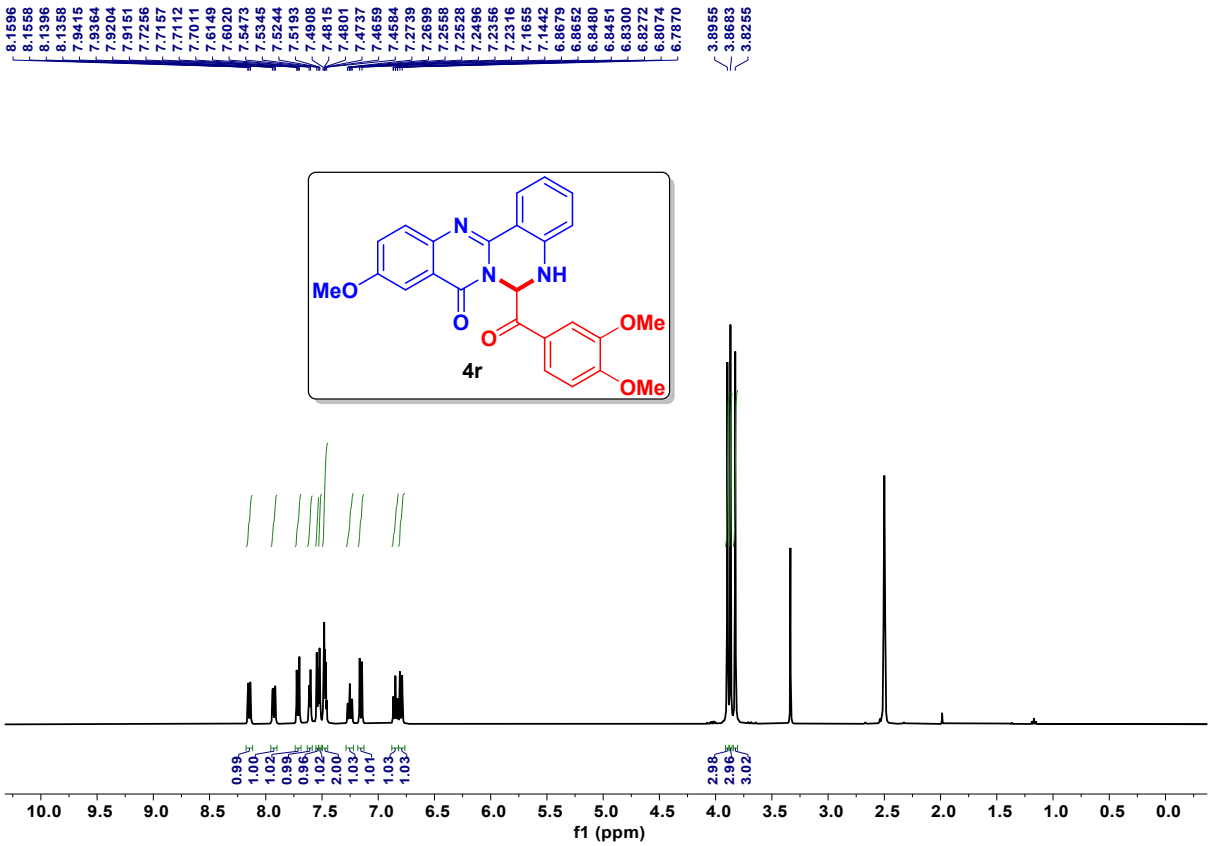


Figure 55. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4r**

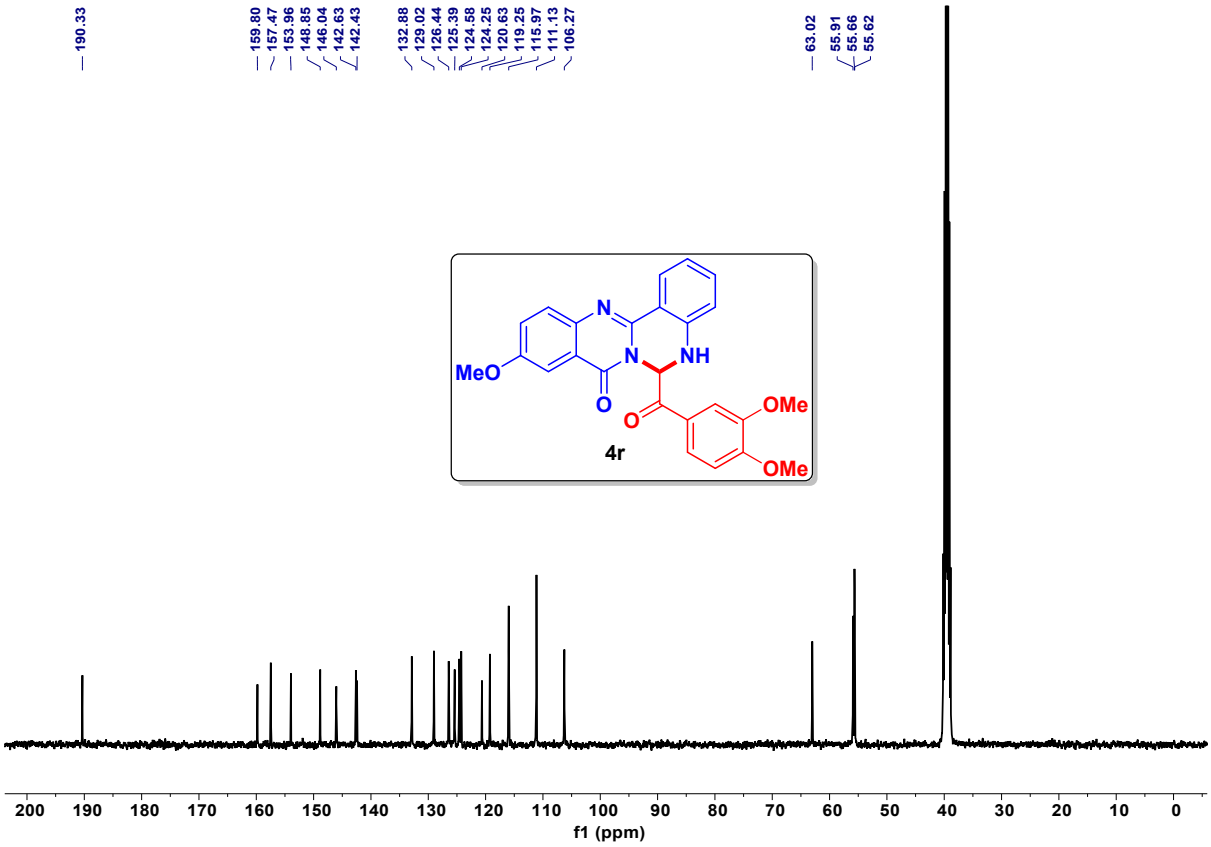


Figure 56. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4r**

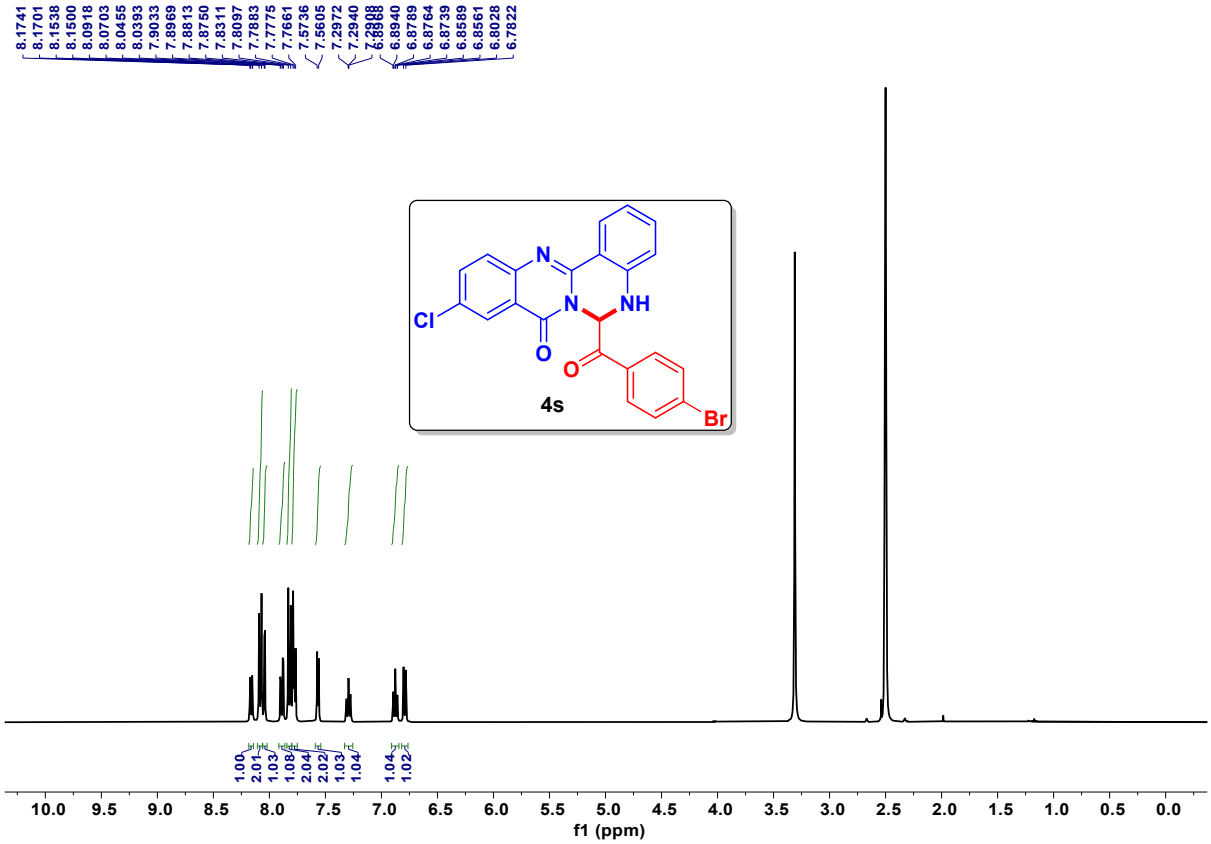


Figure 57. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4s**

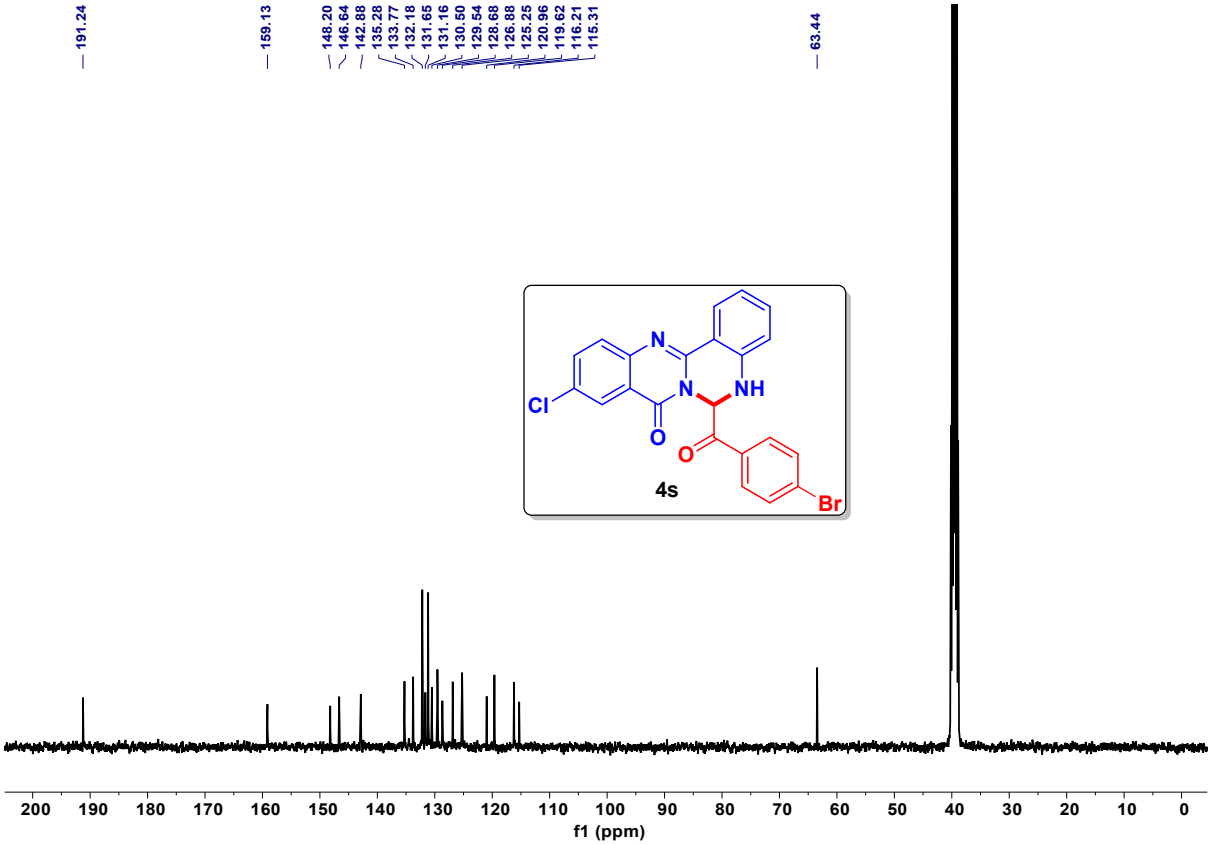


Figure 58. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4s**

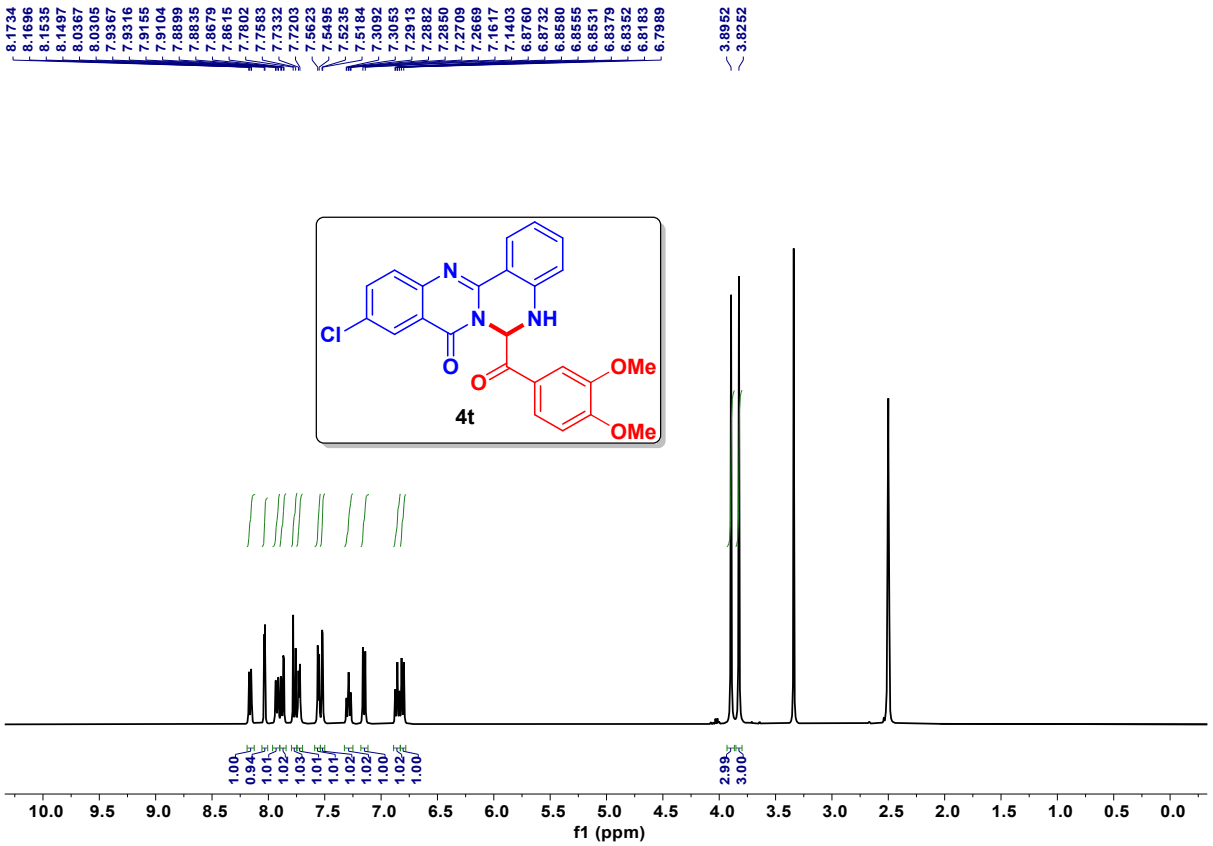


Figure 59. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4t**

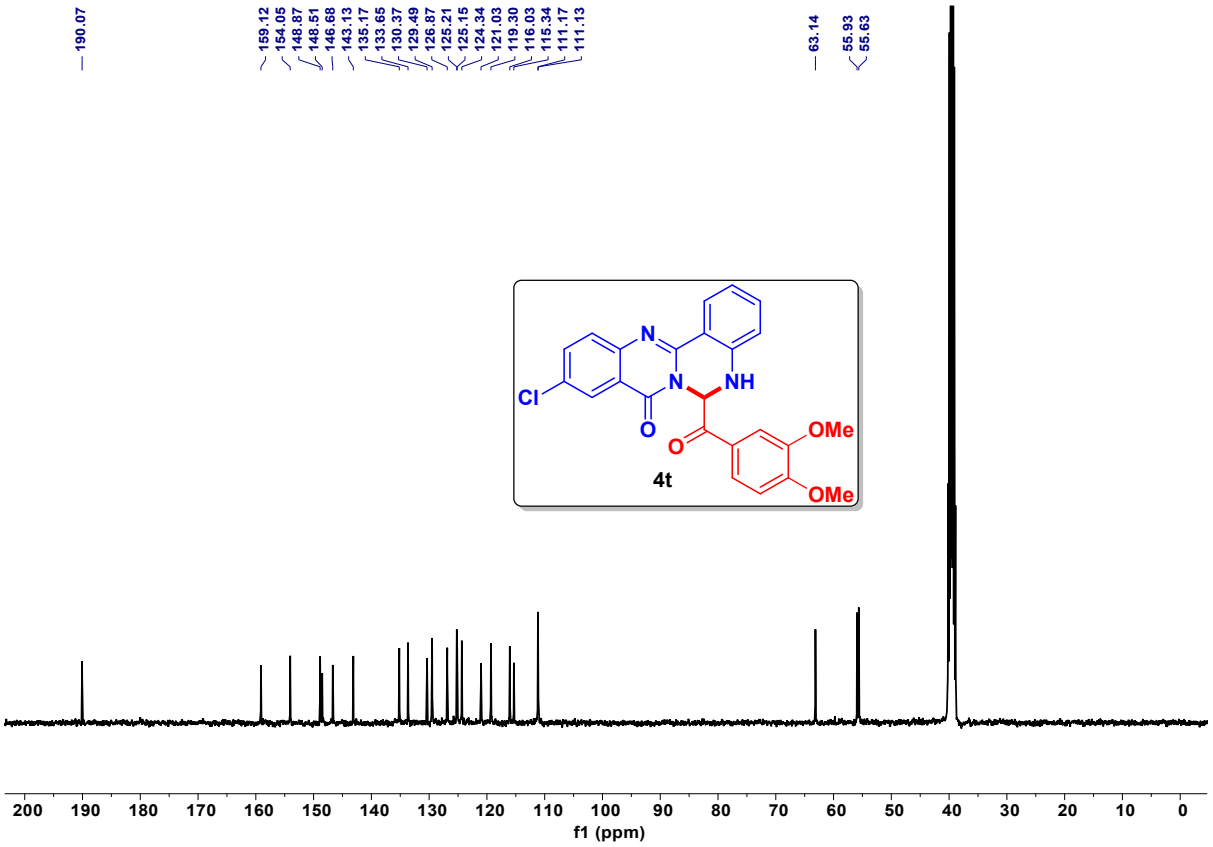


Figure 60. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4t**

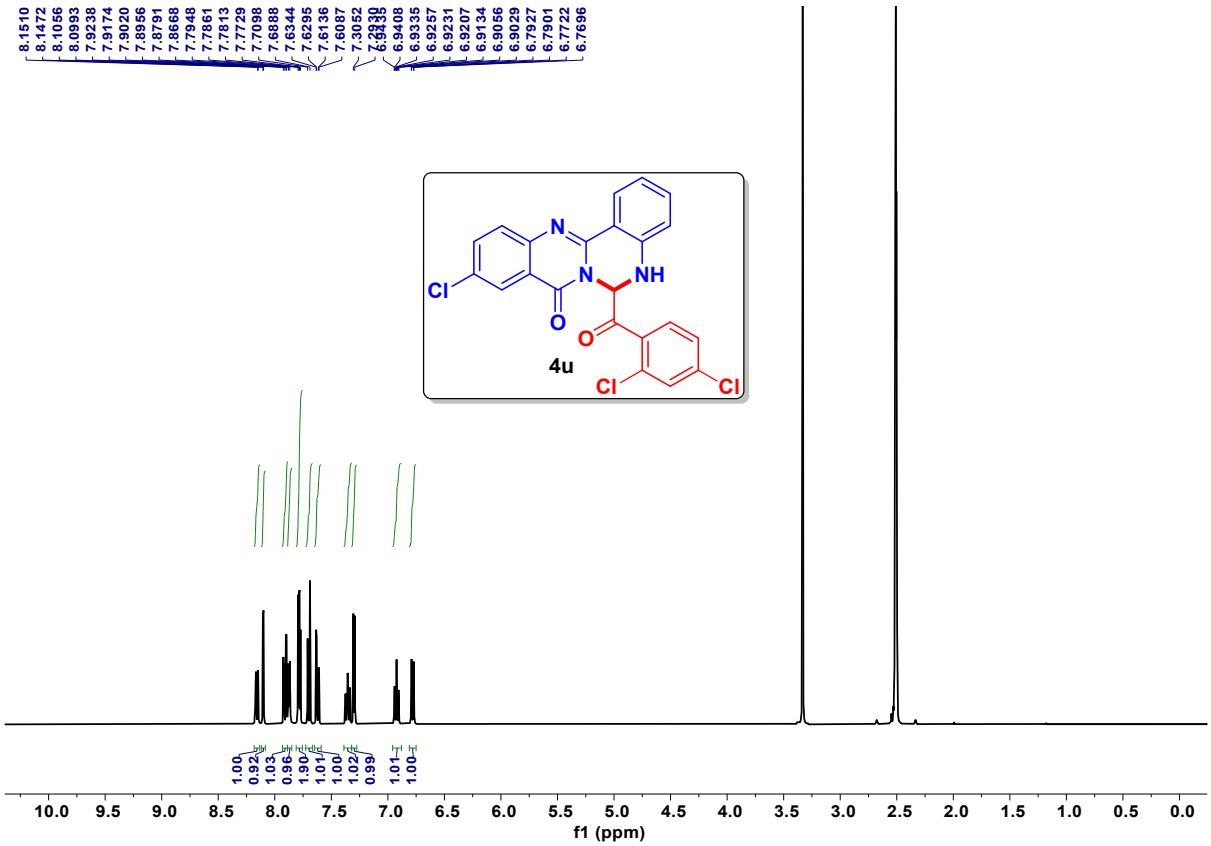


Figure 61. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4u**

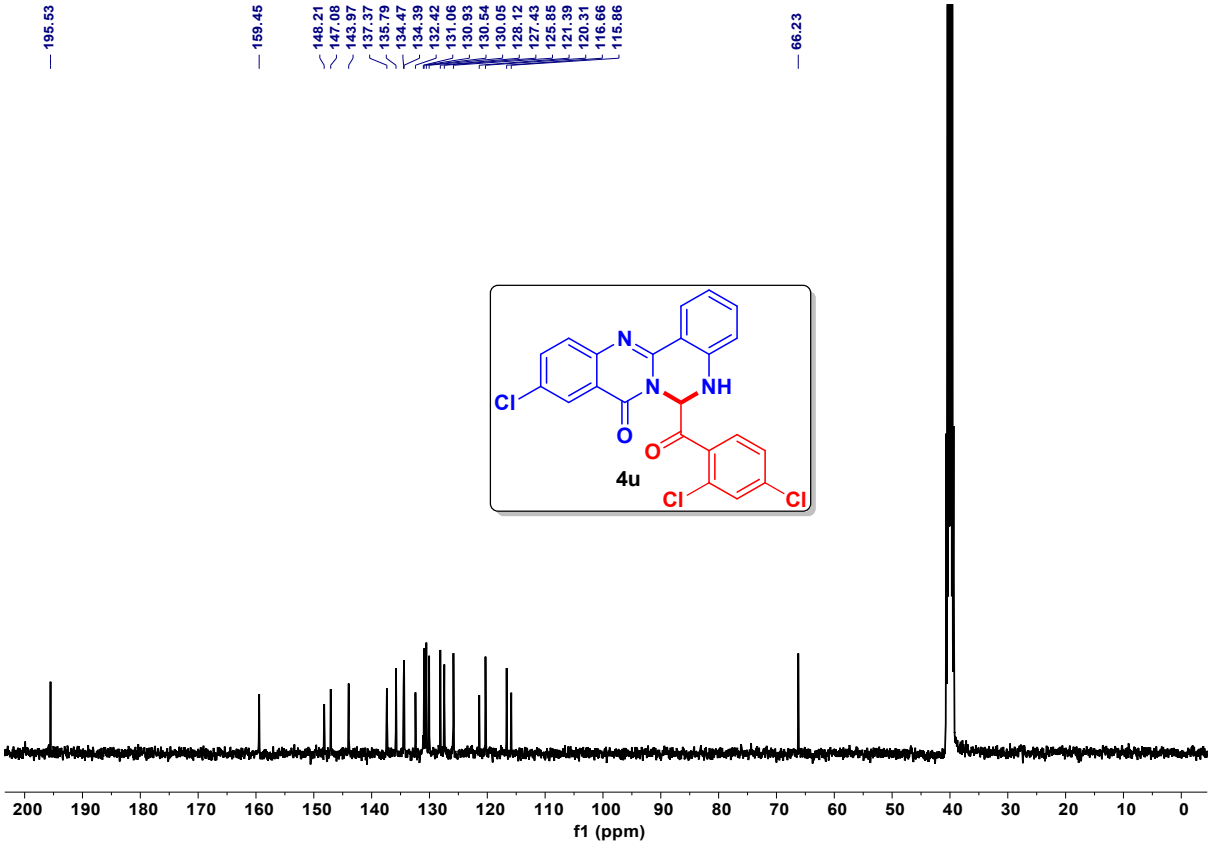


Figure 62. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4u**

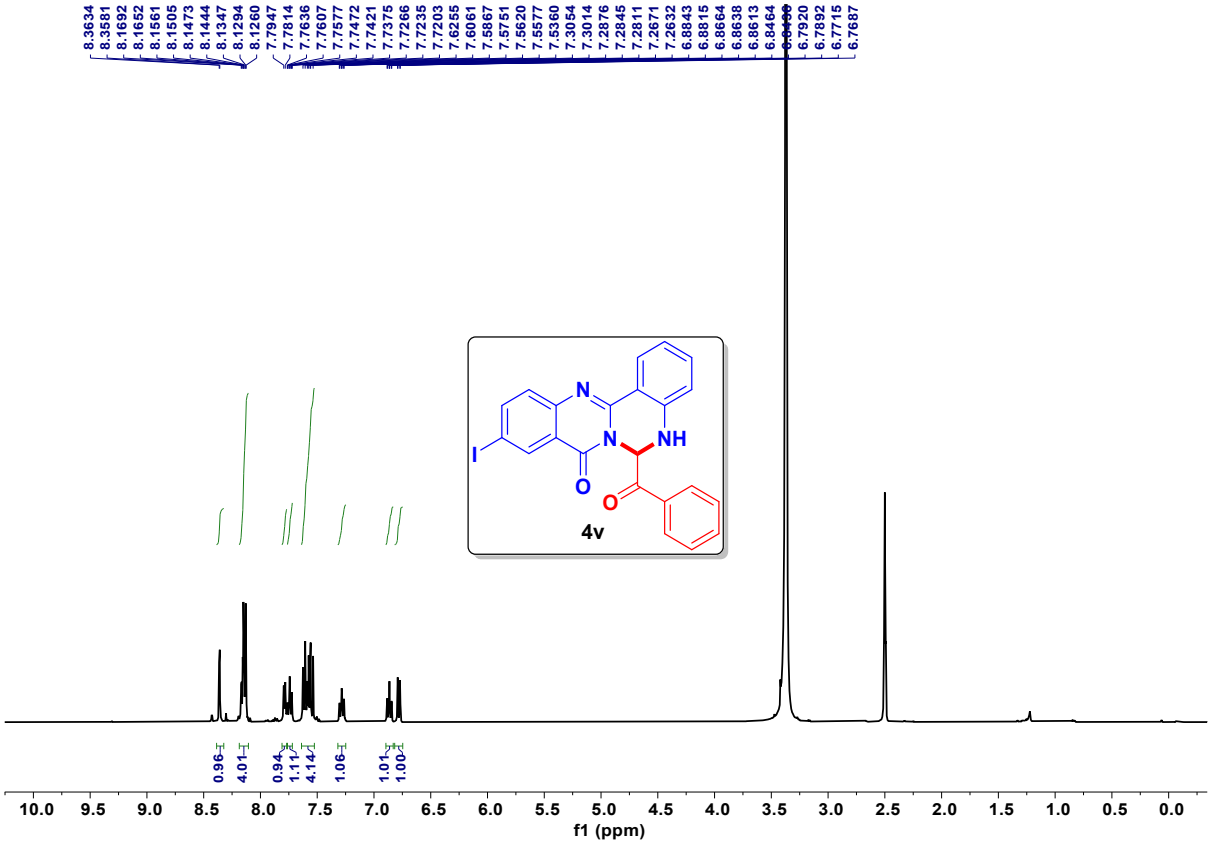


Figure 63. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4v**

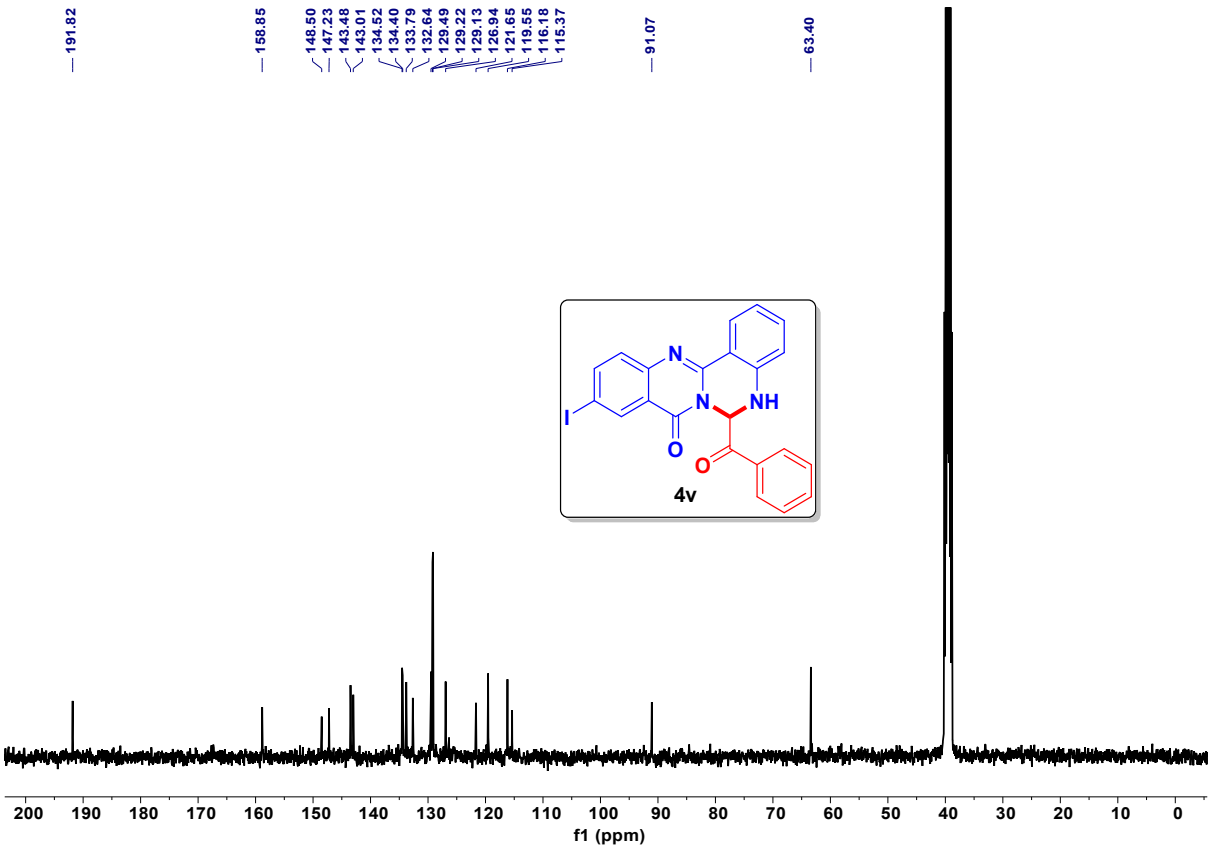


Figure 64. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4v**

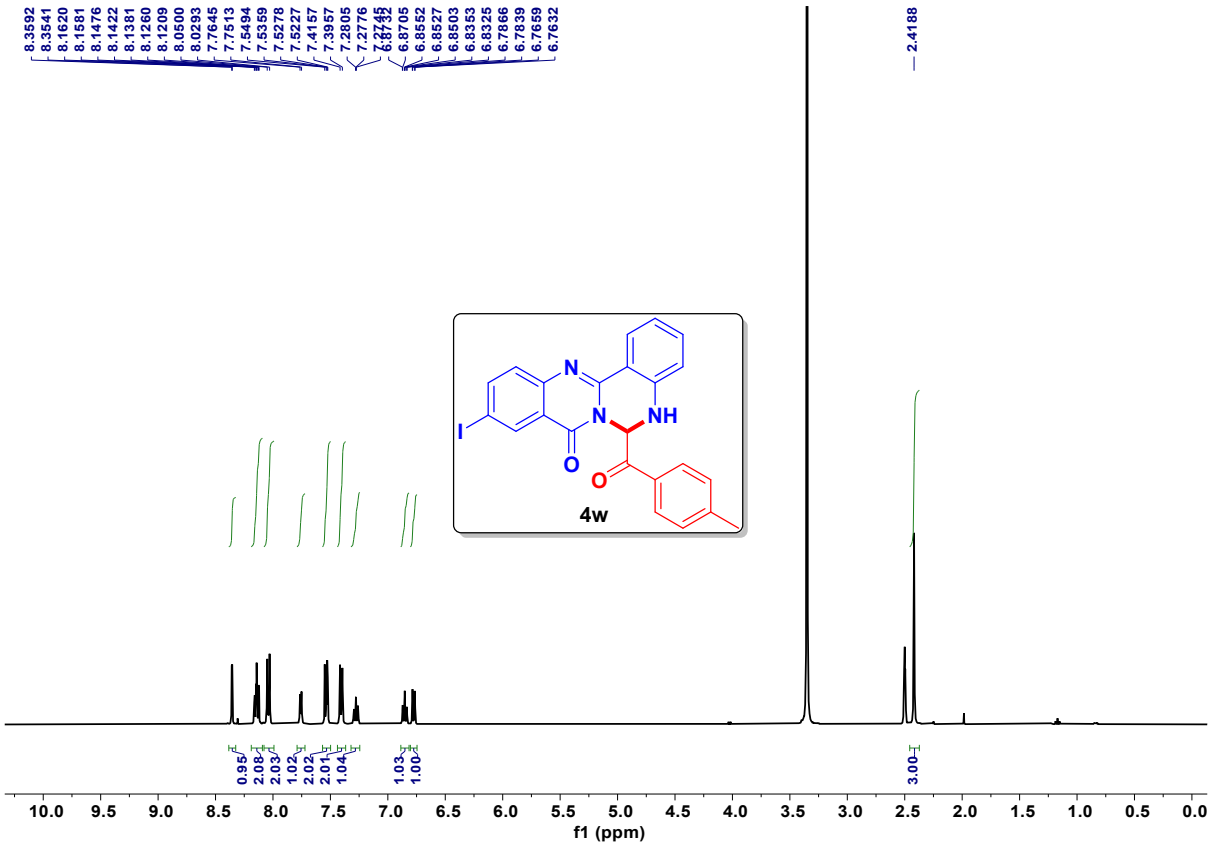


Figure 65. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4w**

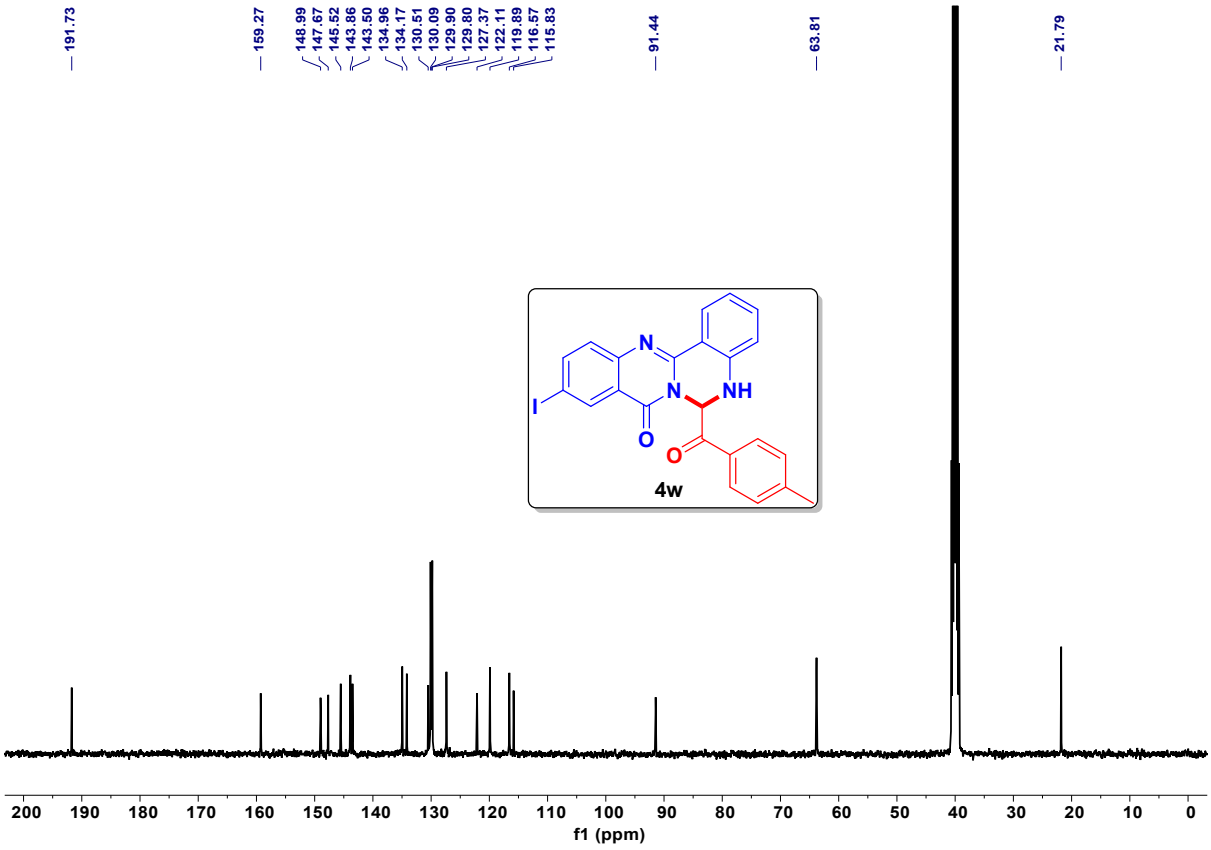


Figure 66. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4w**

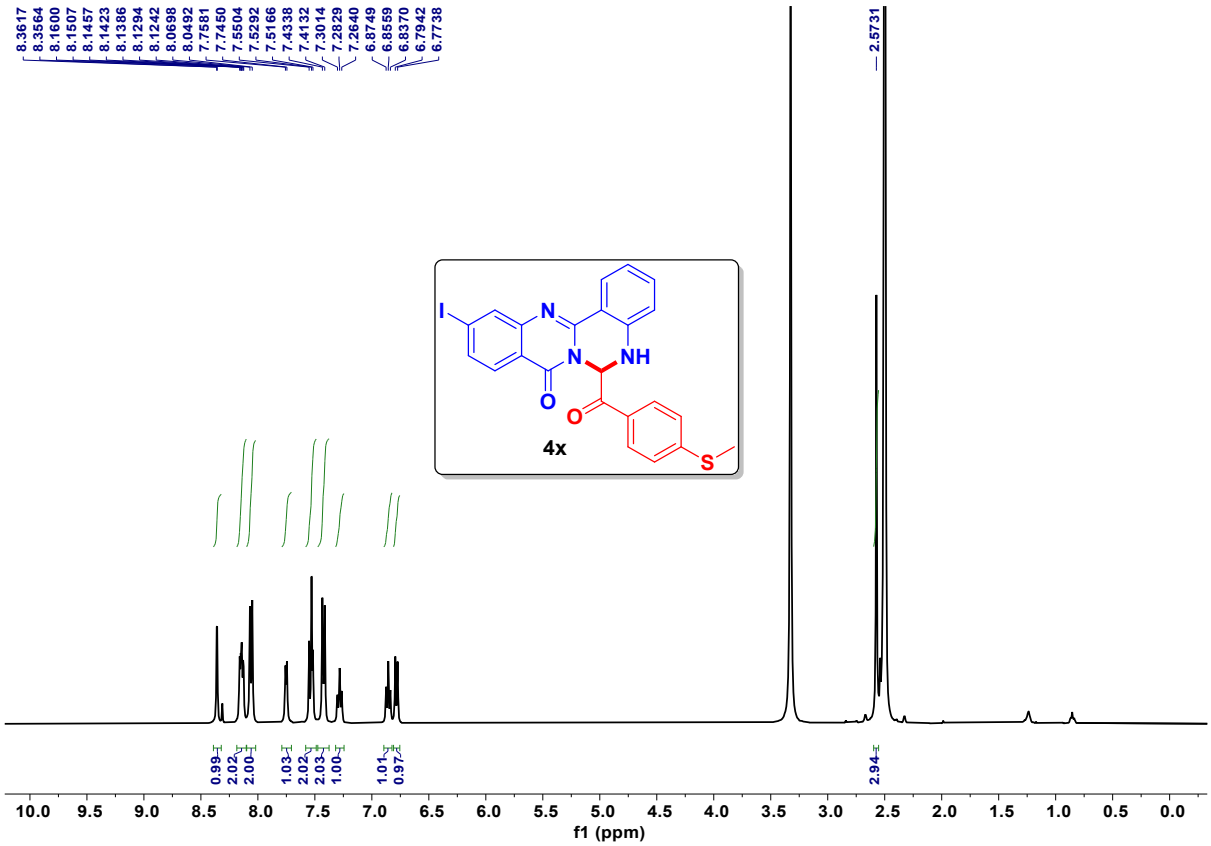


Figure 67. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4x**

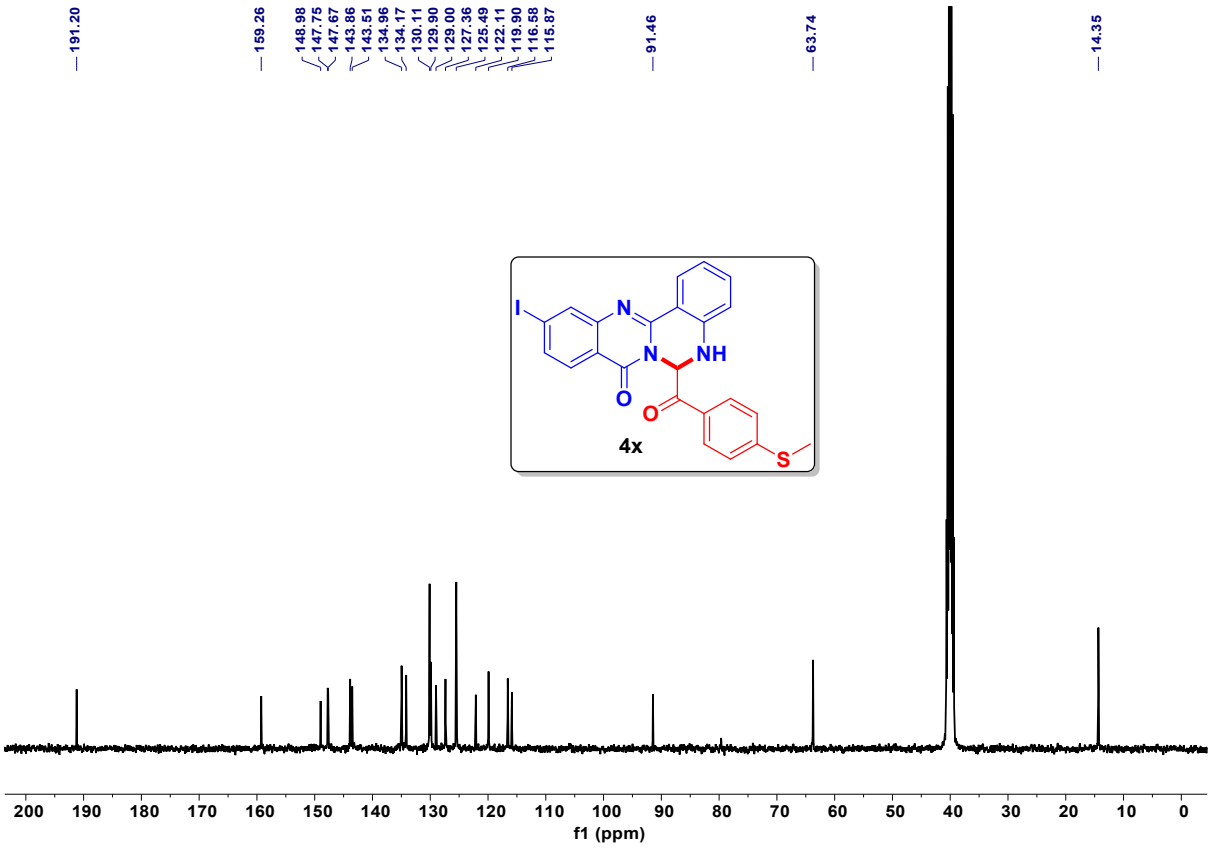


Figure 68. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4x**

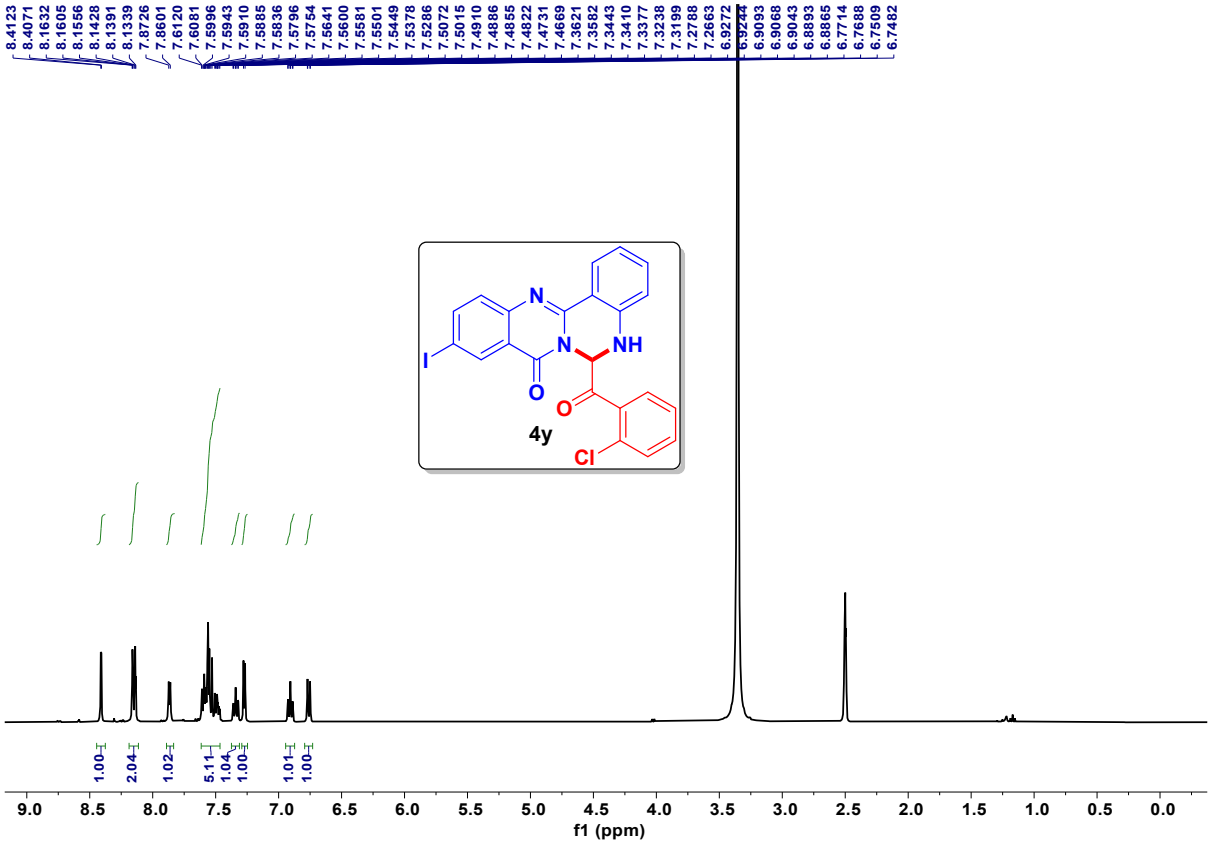


Figure 69. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4y**

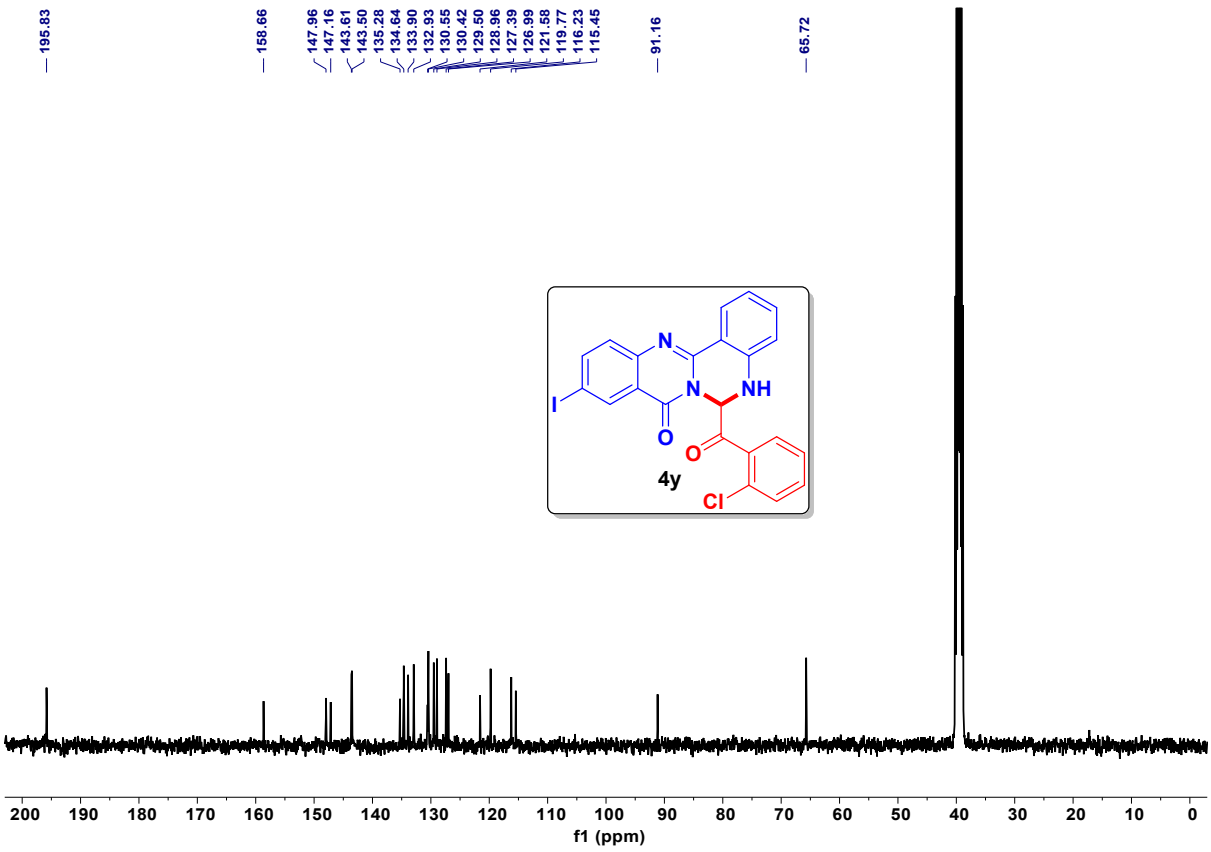


Figure 70. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4y**

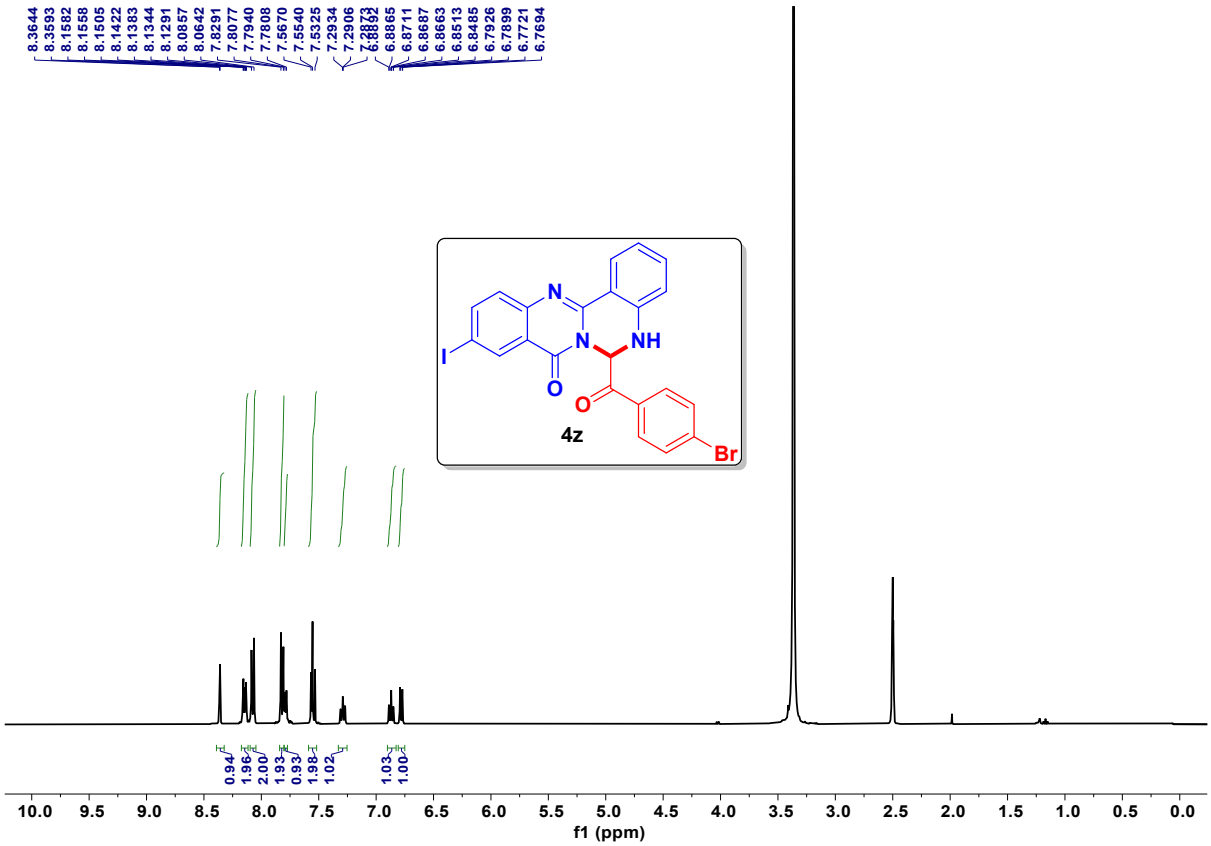


Figure 71. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4z**

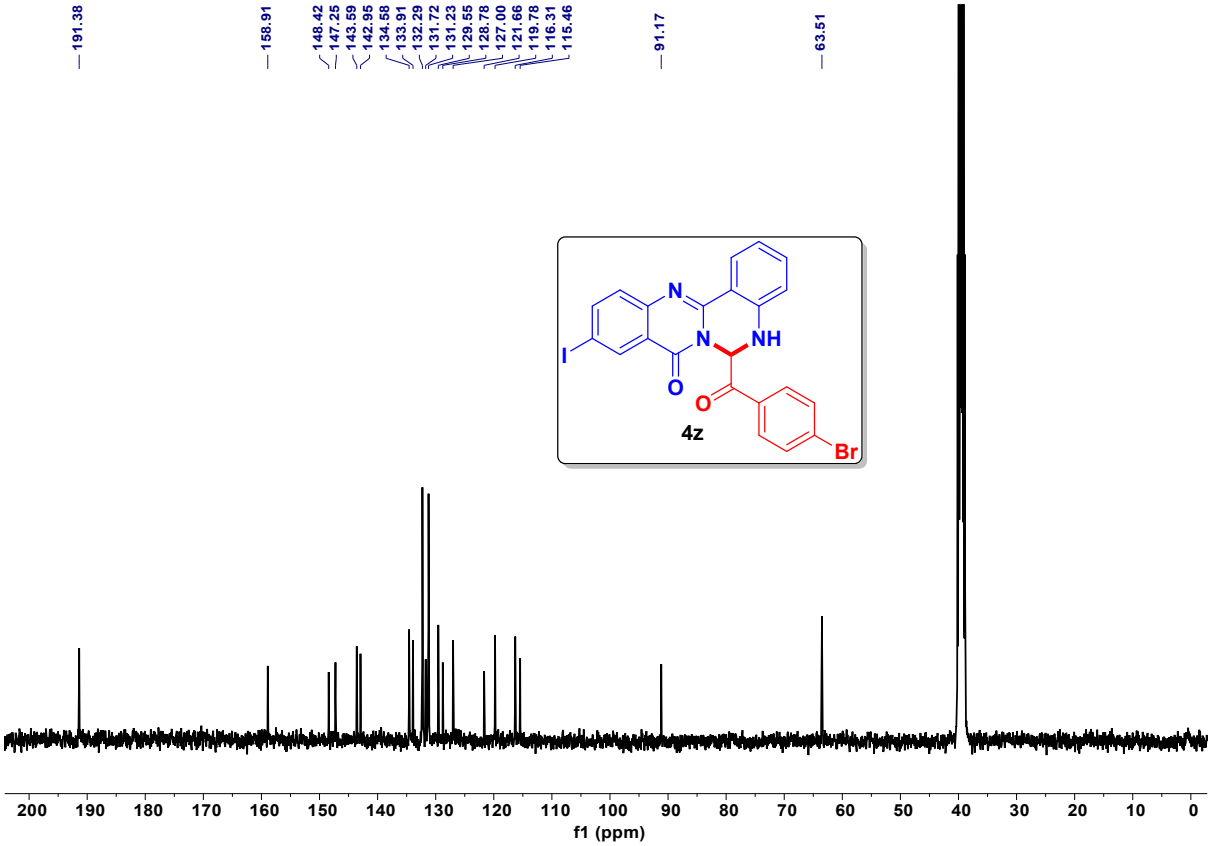


Figure 72. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4z**

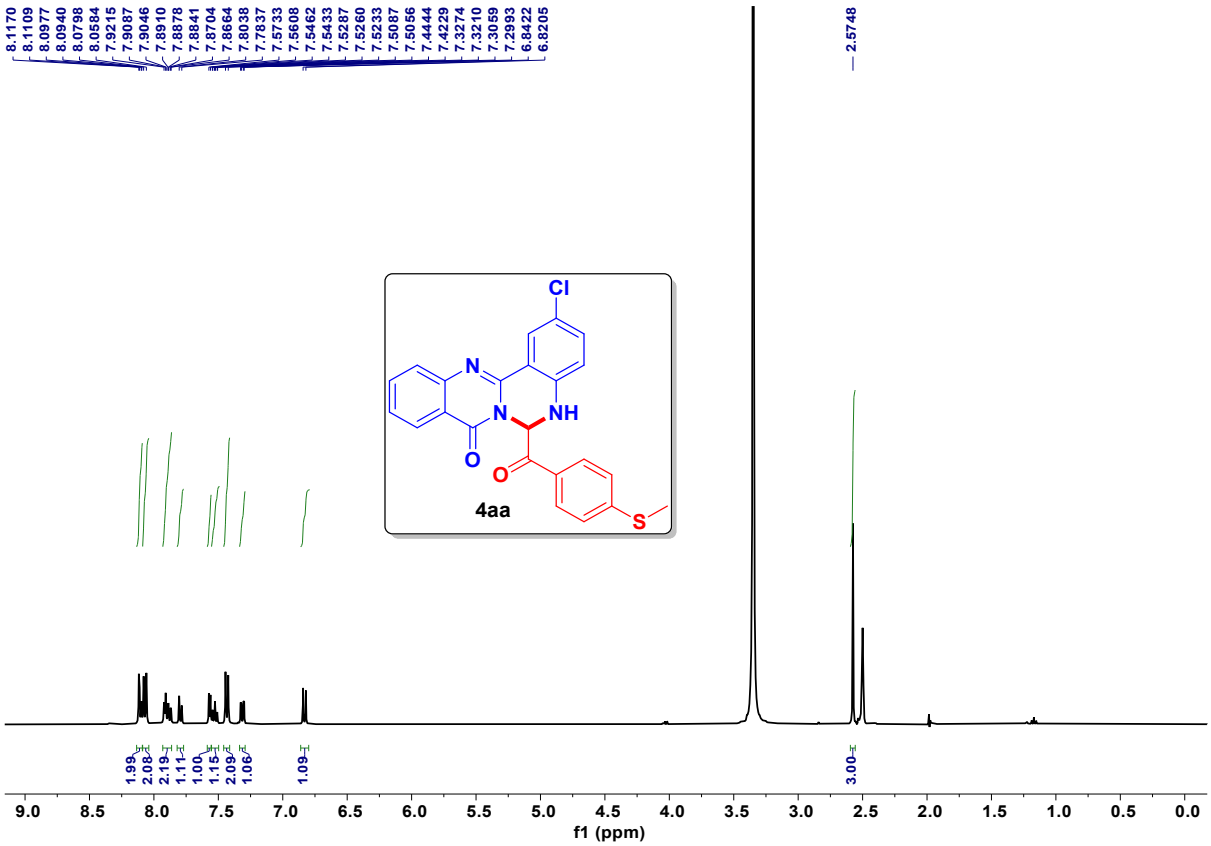


Figure 73. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4aa**

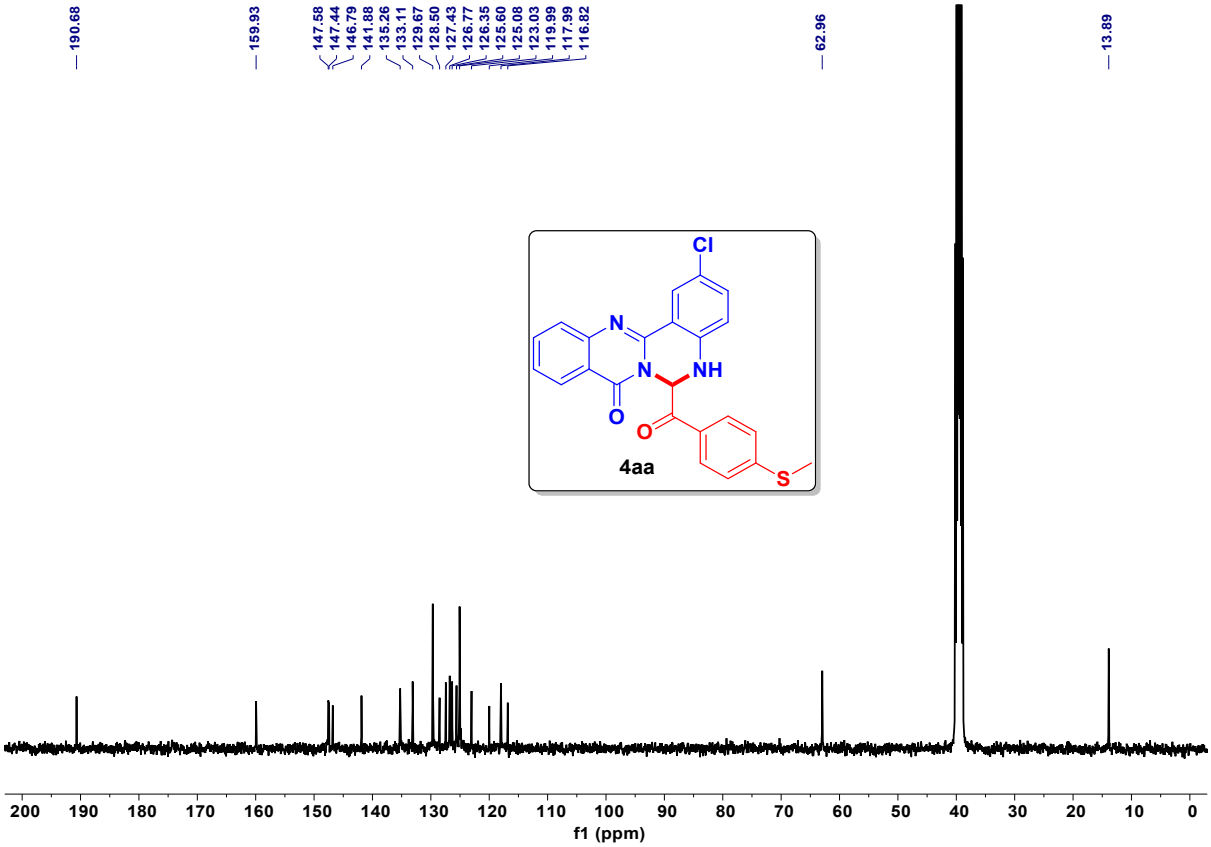


Figure 74. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4aa**

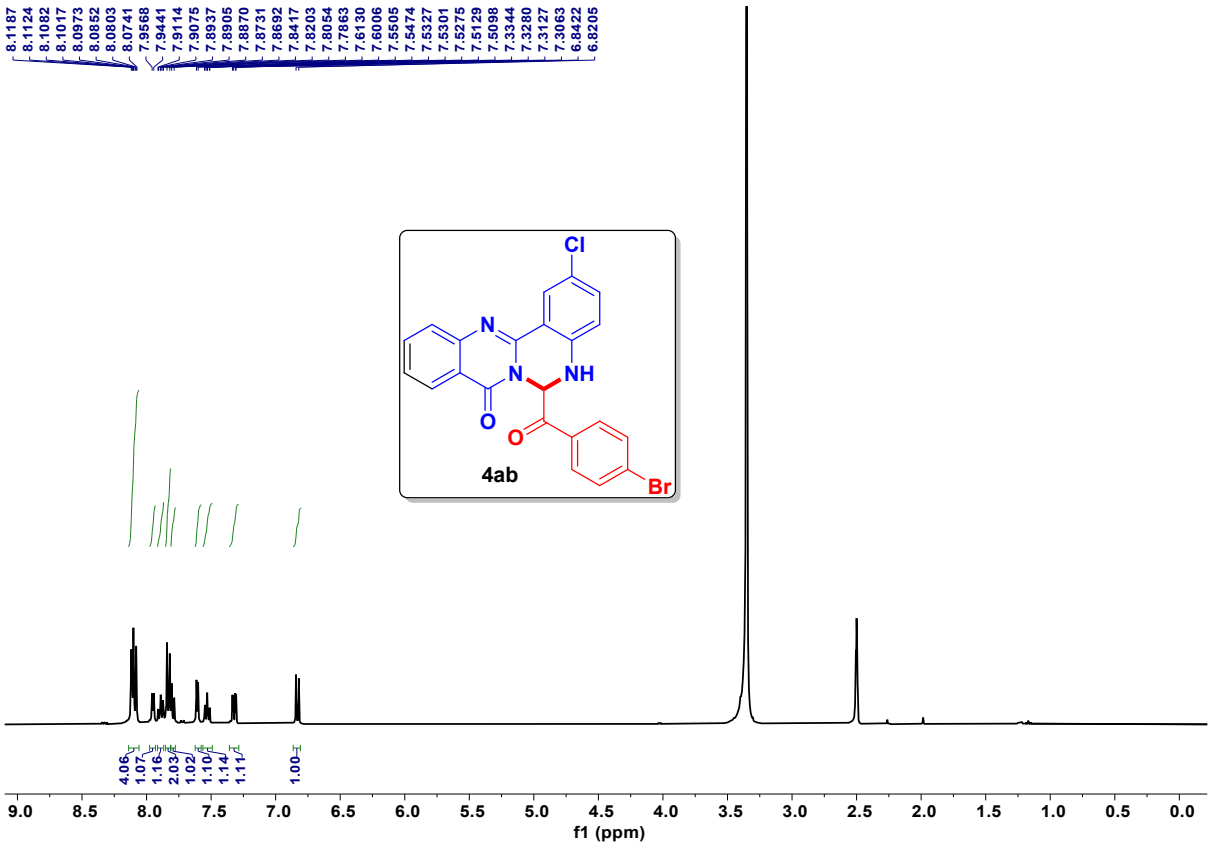


Figure 75. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ab**

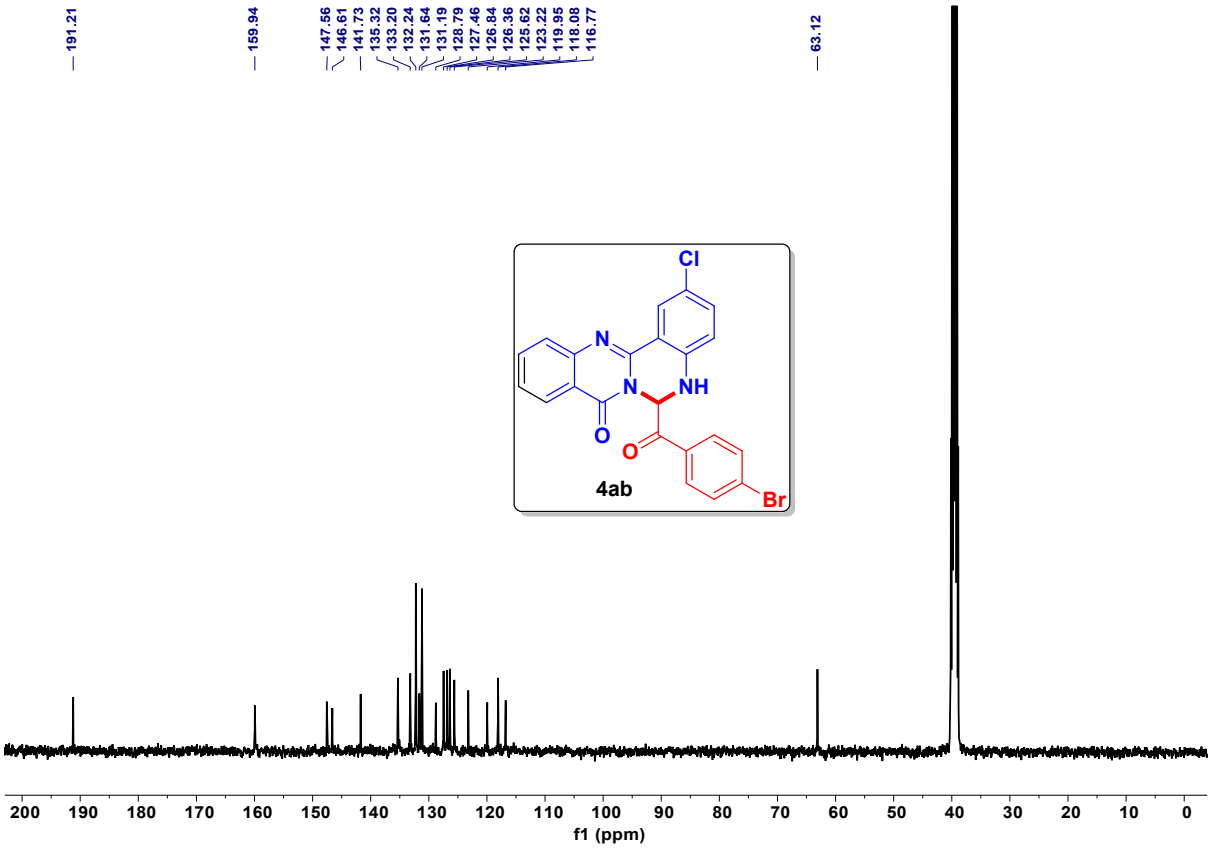


Figure 76. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ab**

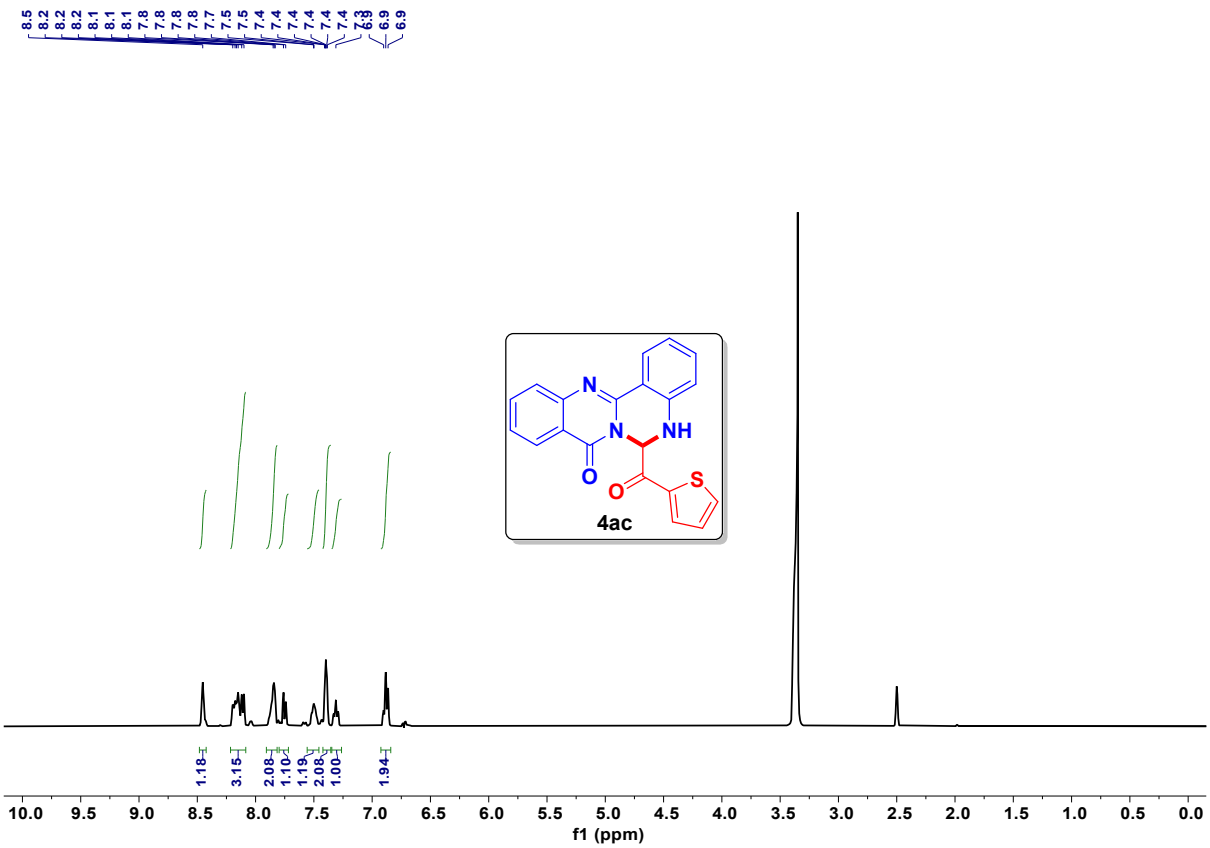


Figure 77. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ac**

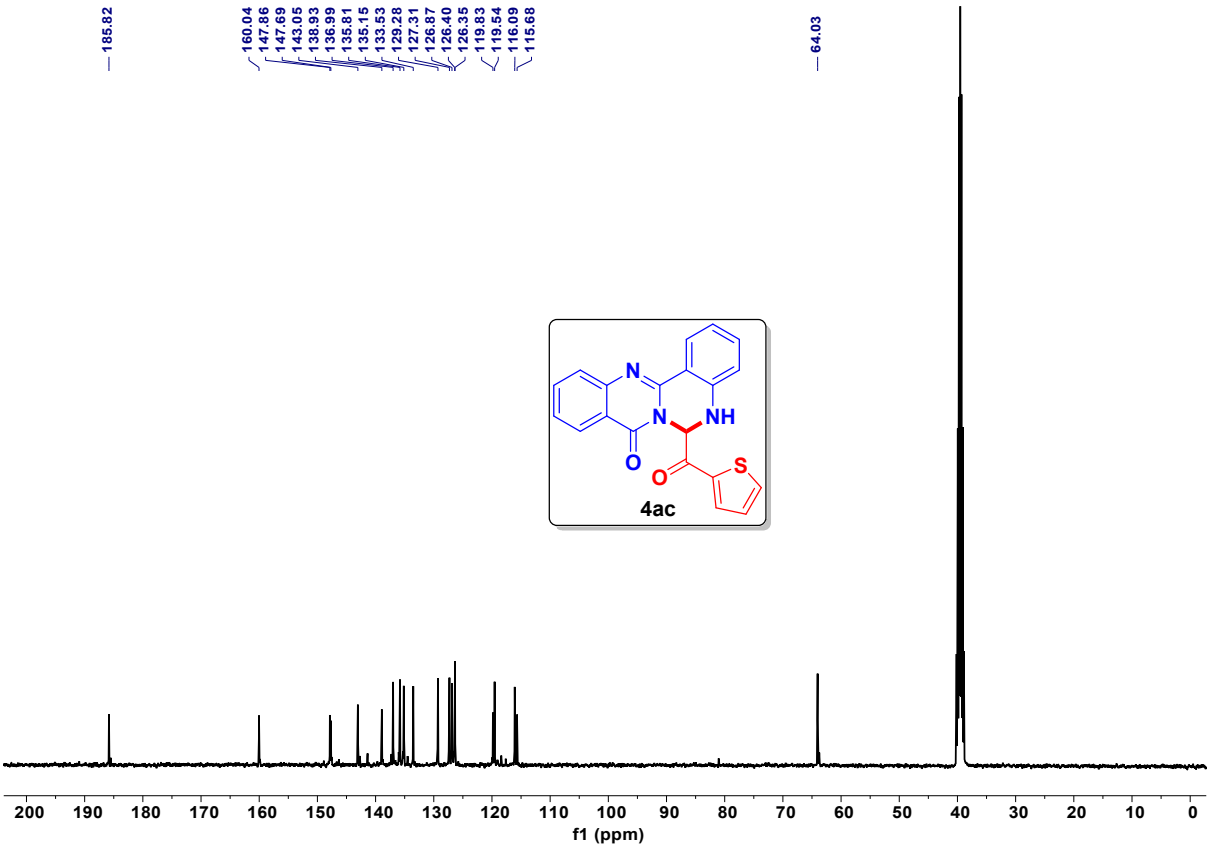


Figure 78. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ac**

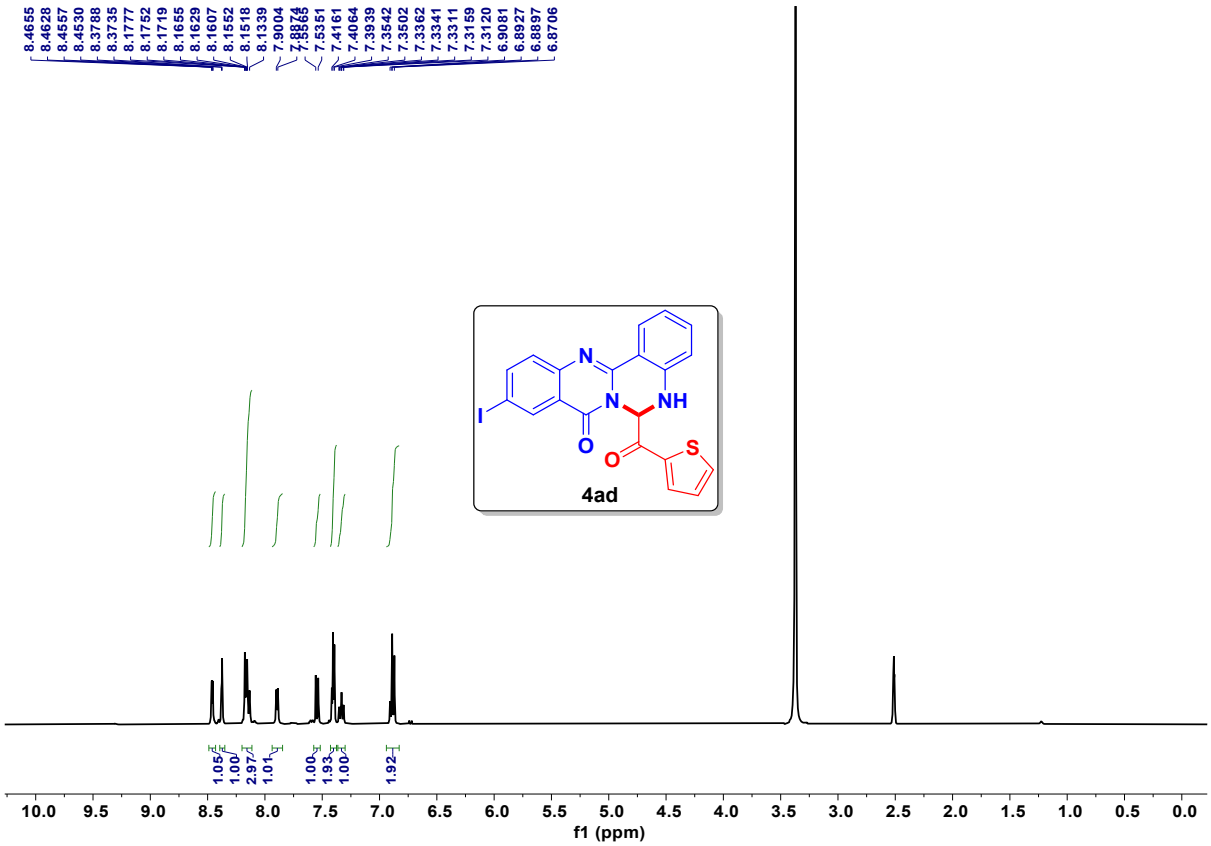


Figure 79. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ad**

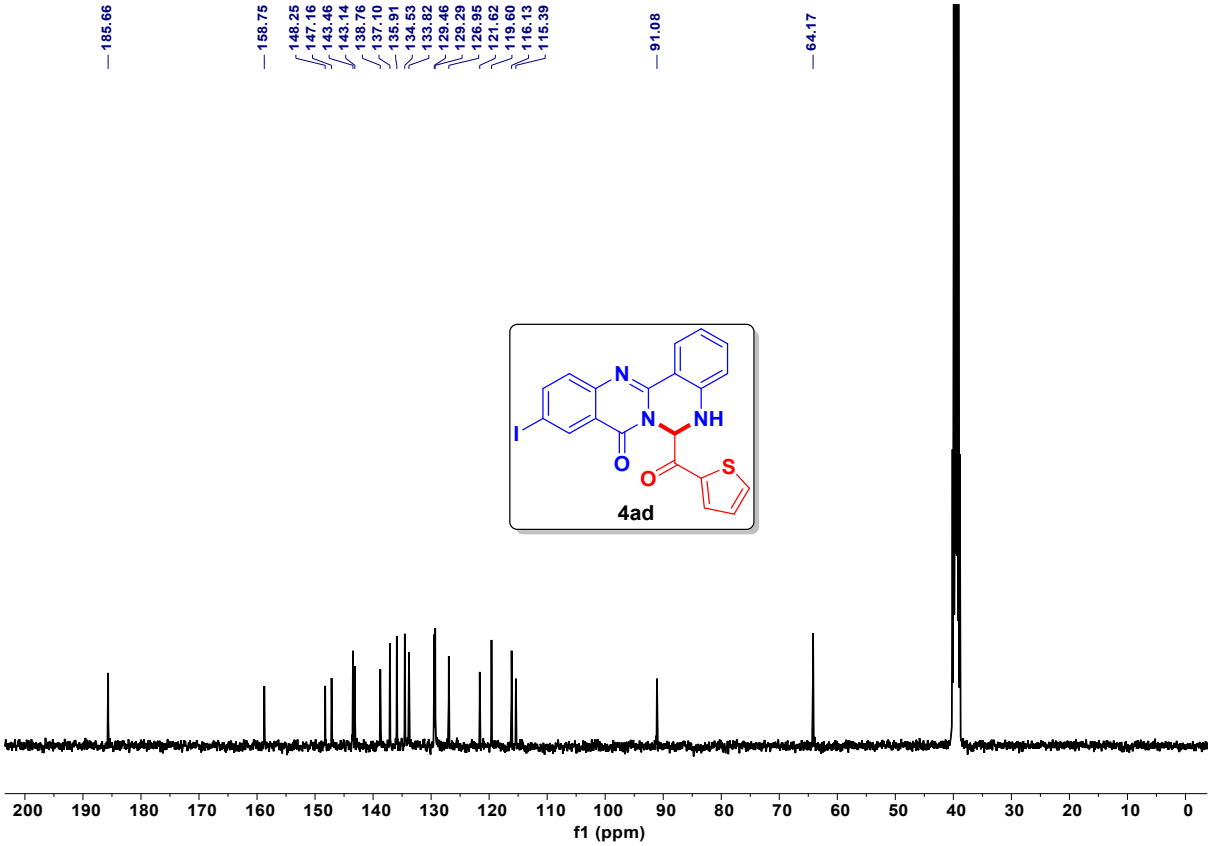


Figure 80. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ad**

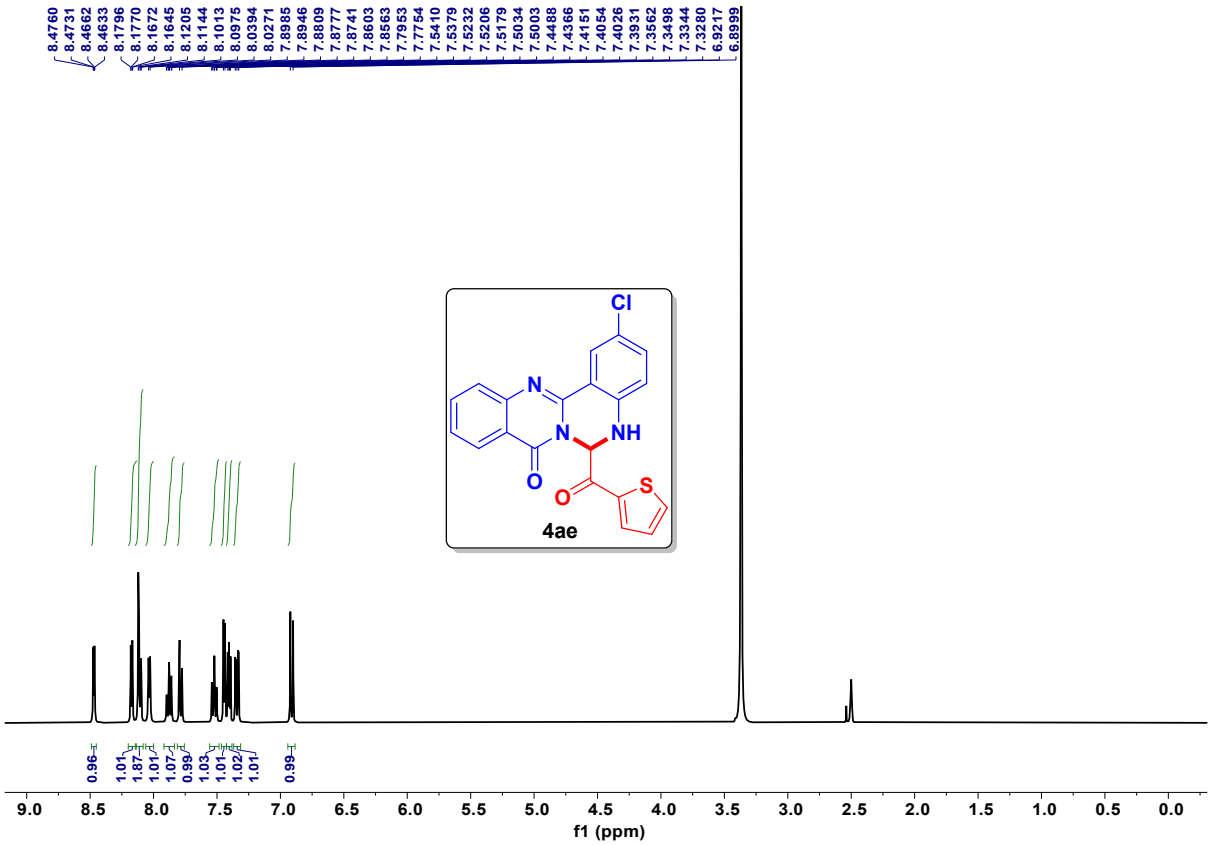


Figure 81. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ae**

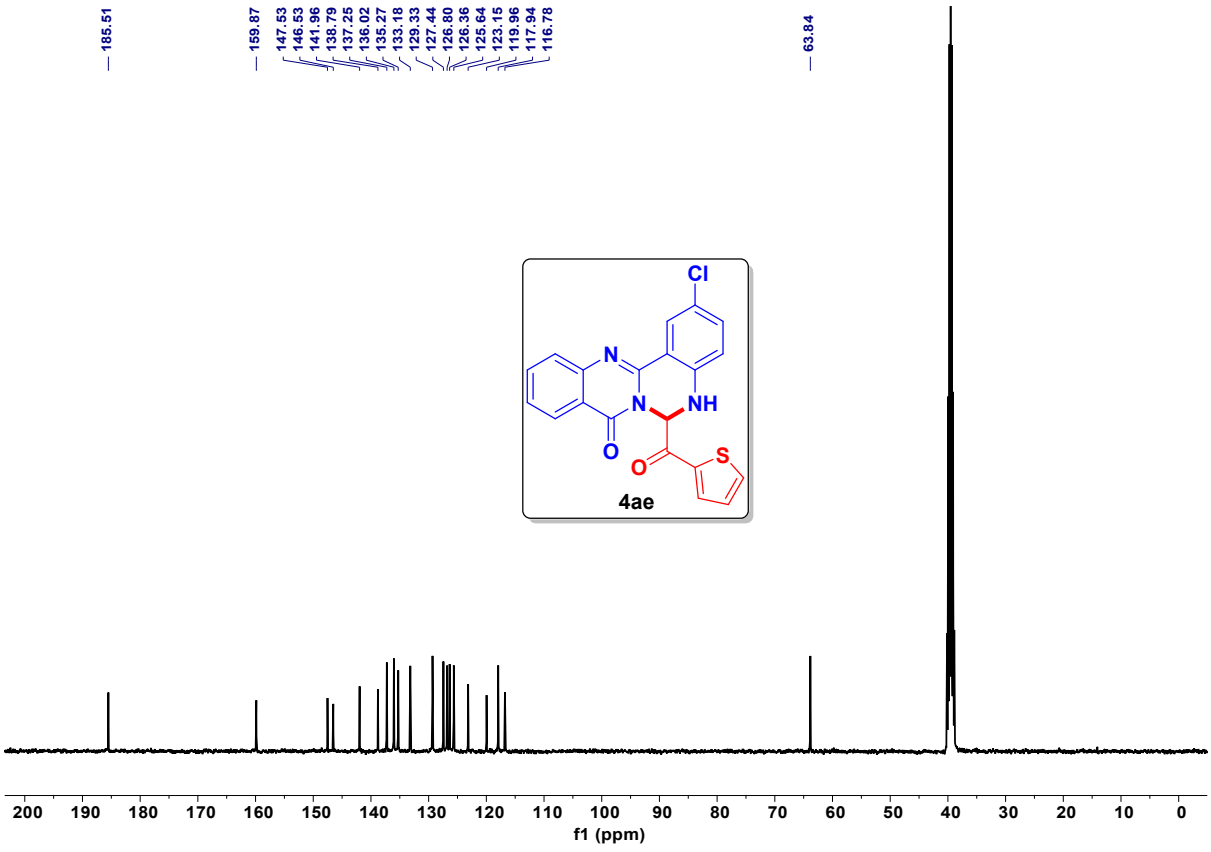


Figure 82. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ae**

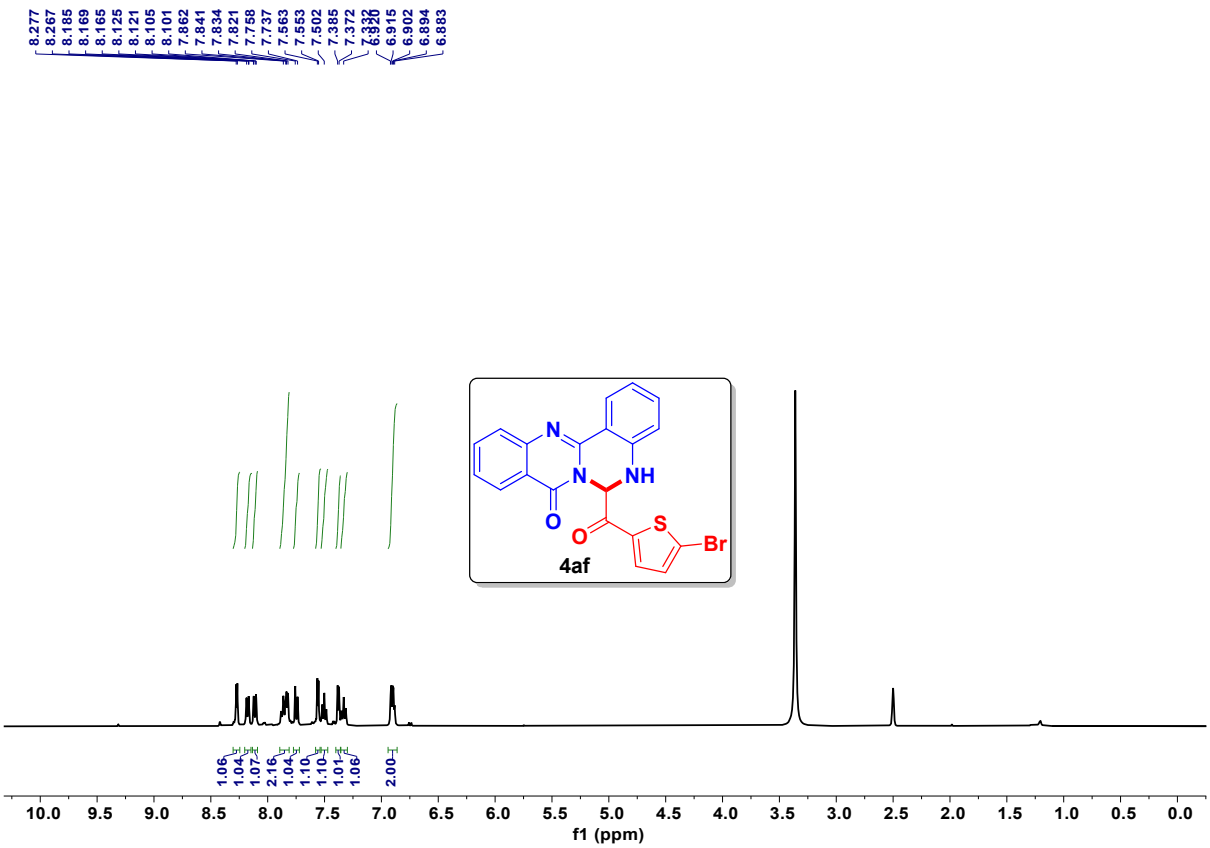


Figure 83. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4af**

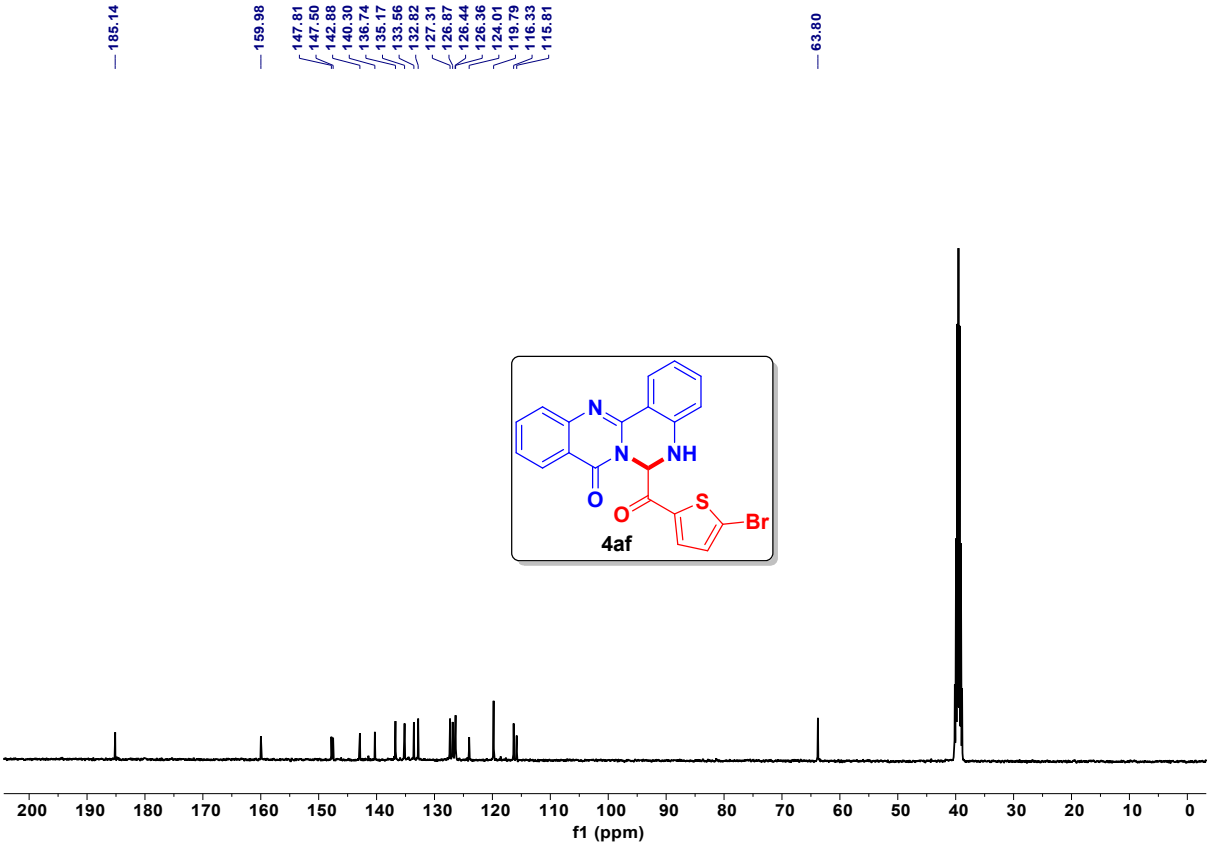


Figure 84. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4af**

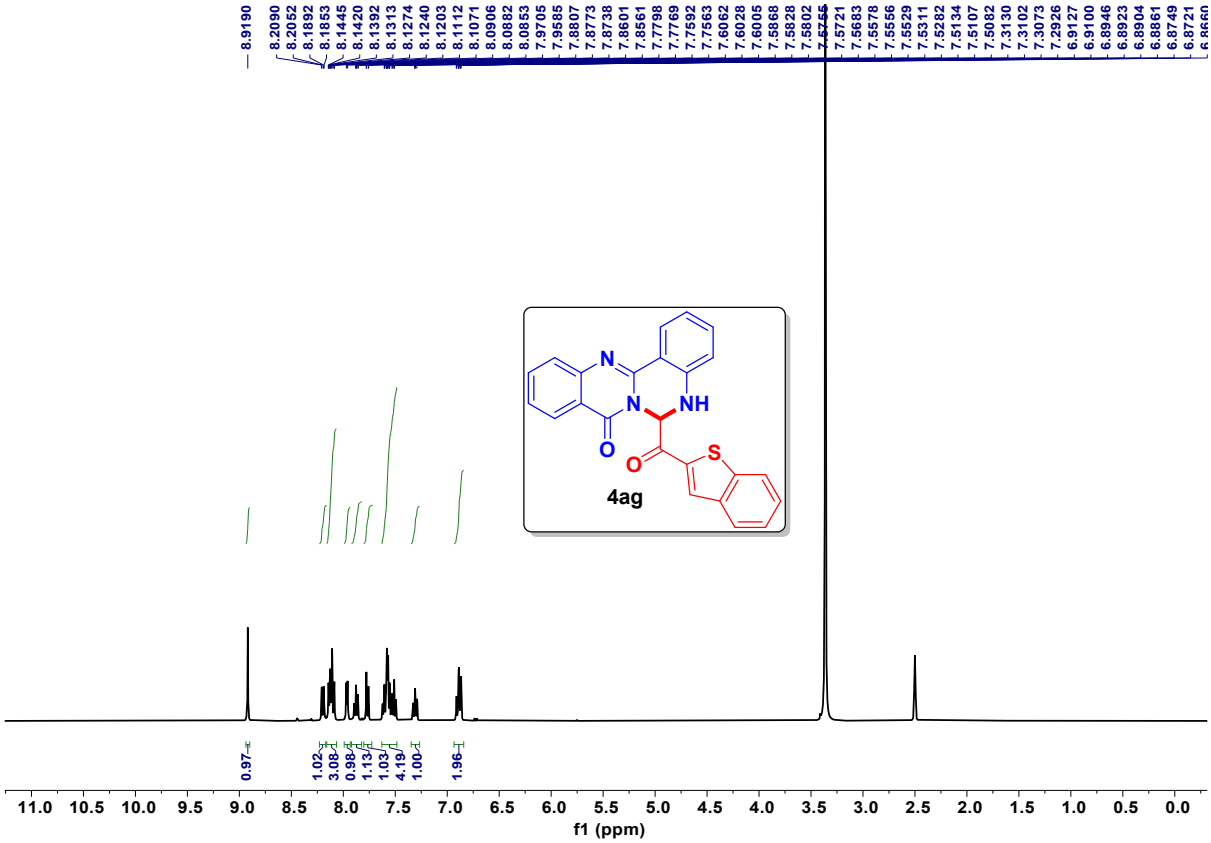


Figure 85. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ag**

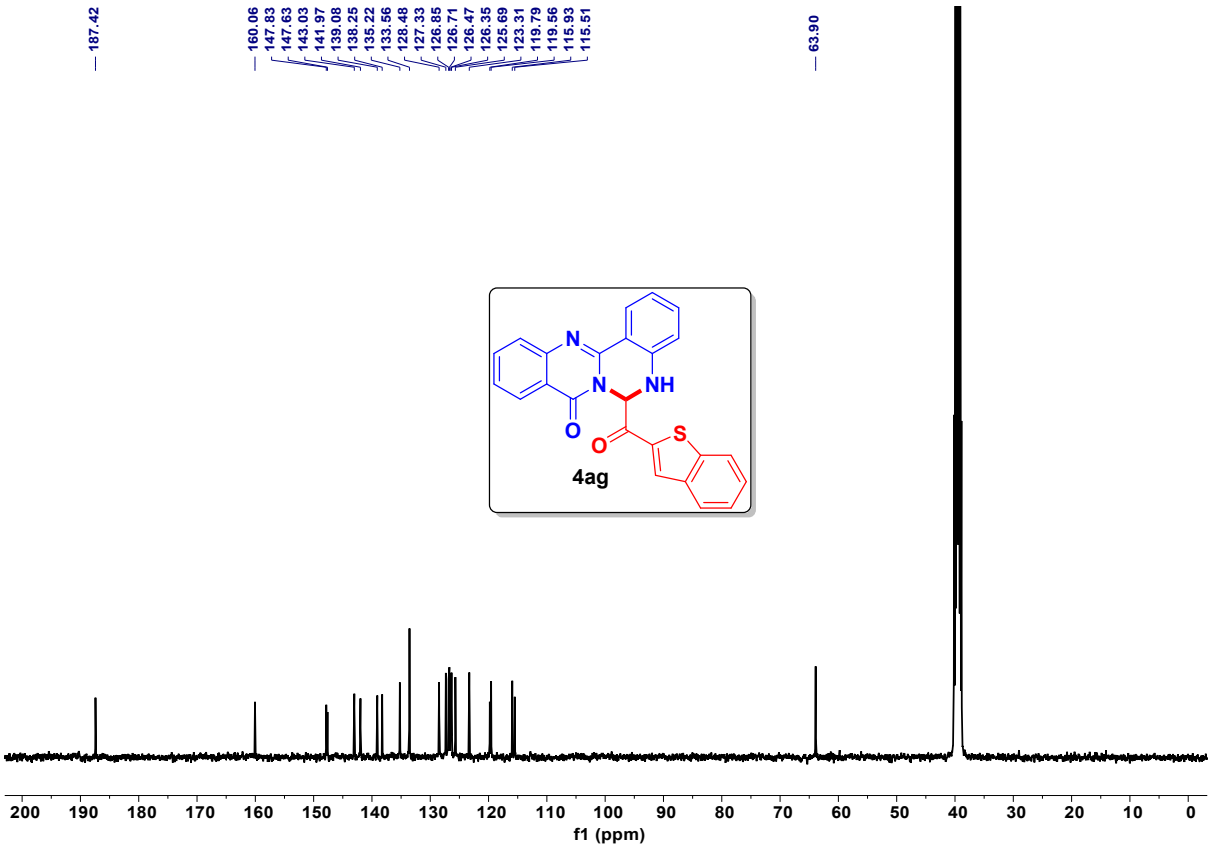


Figure 86. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ag**

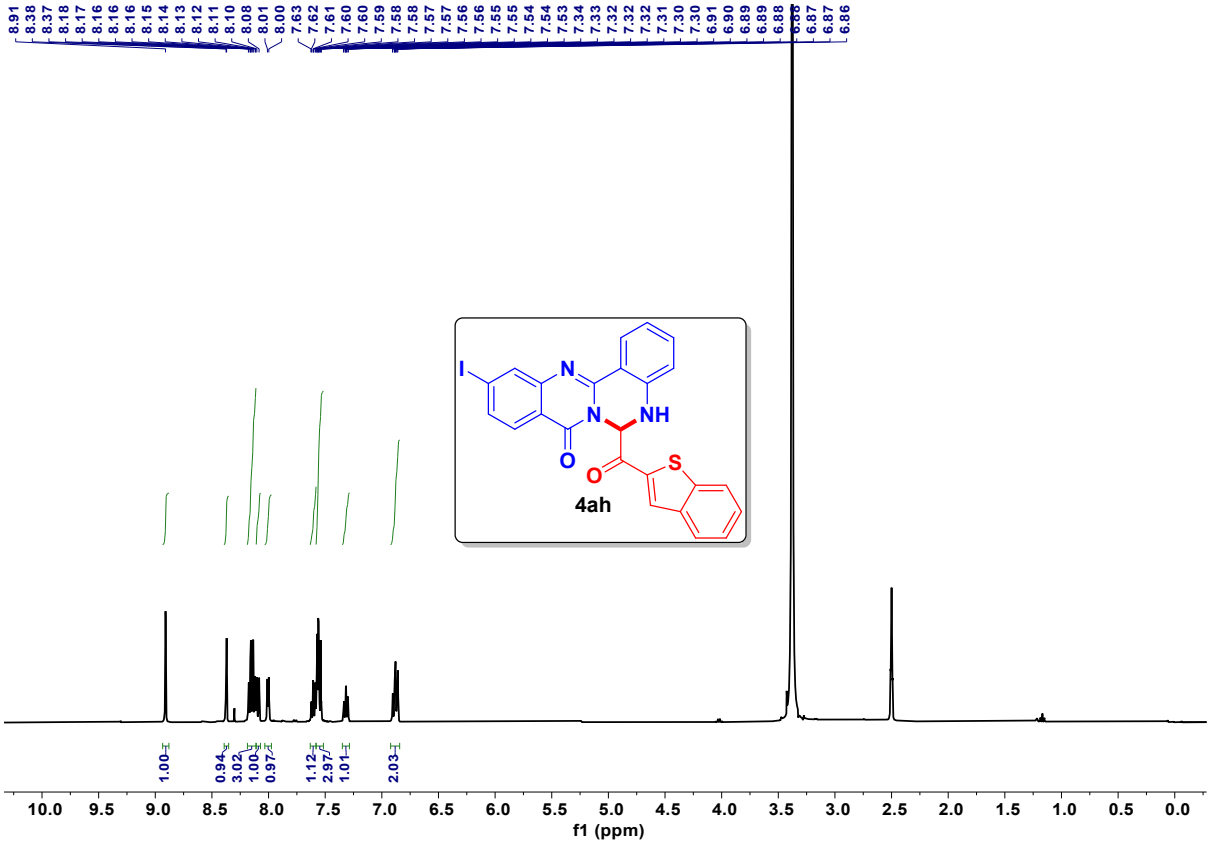


Figure 87. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ah**

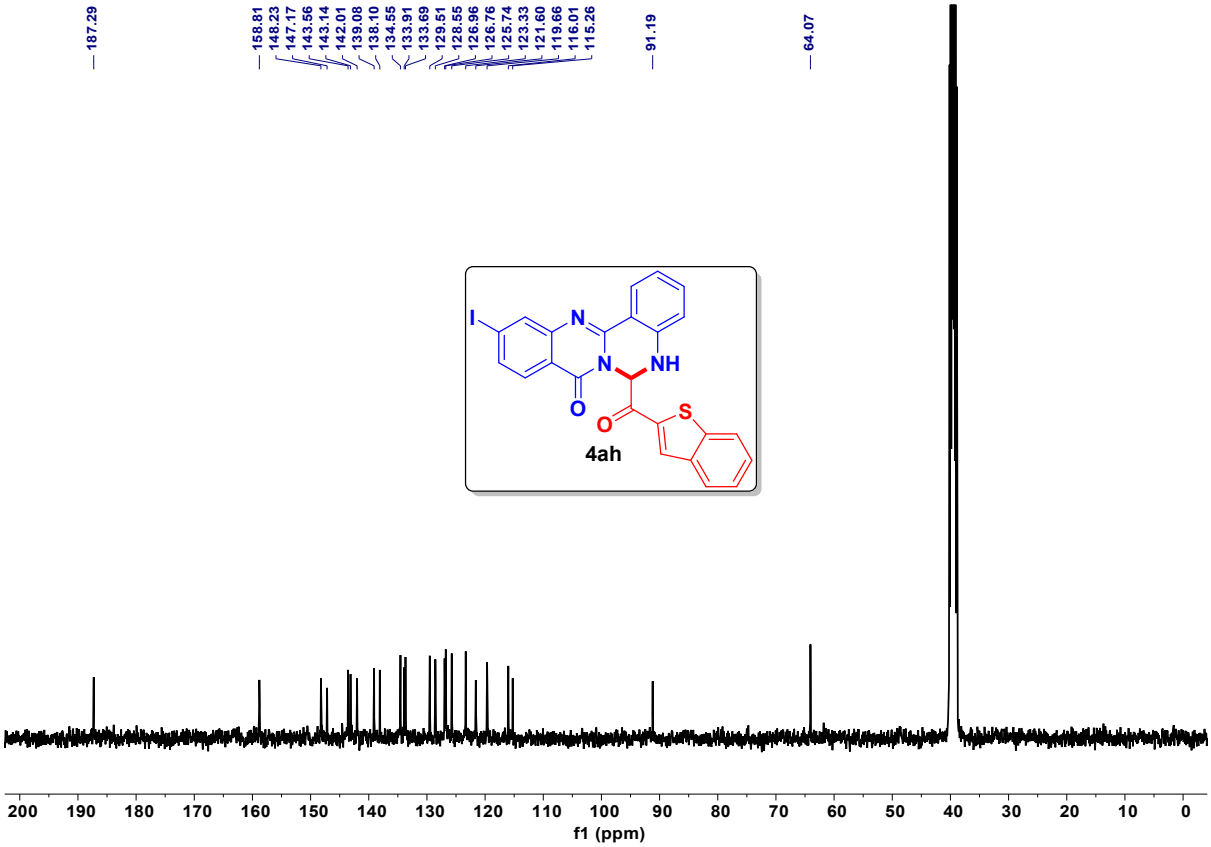


Figure 88. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ah**

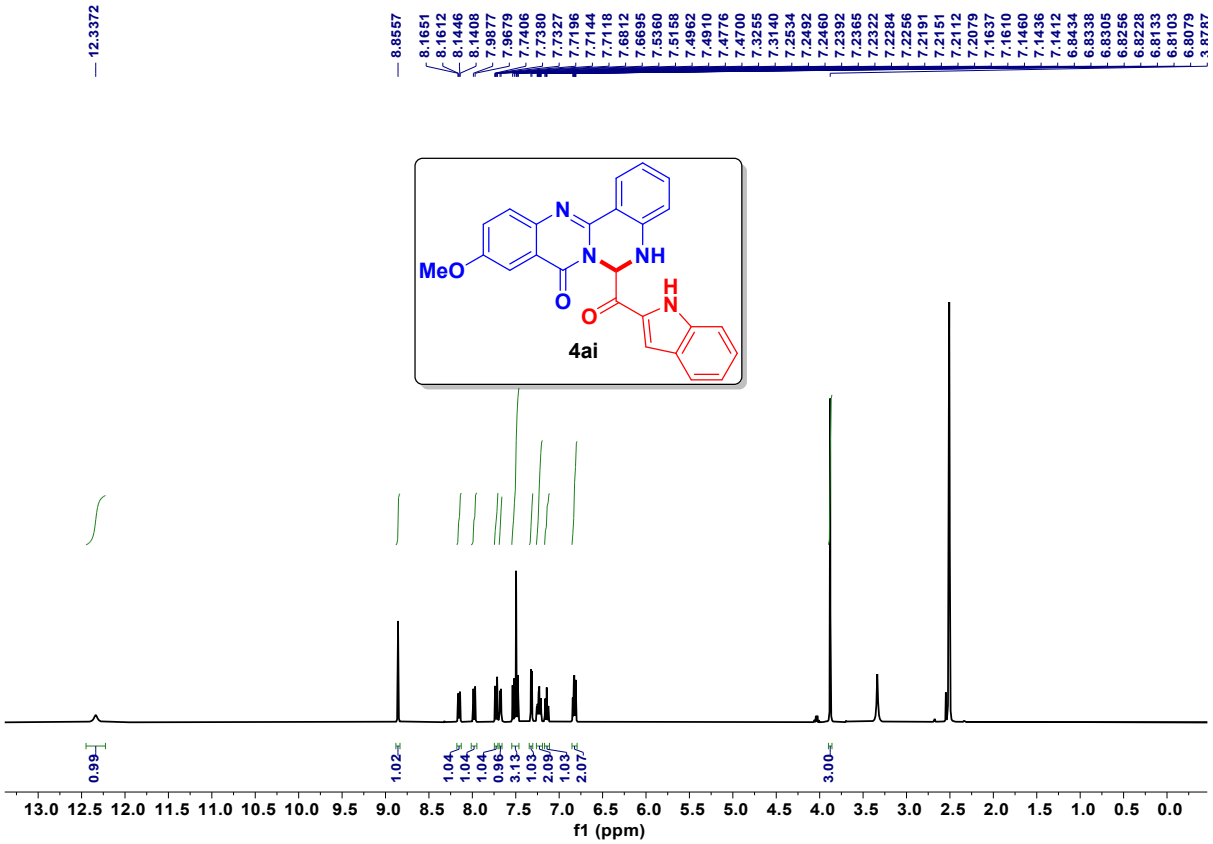


Figure 89. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4ai**

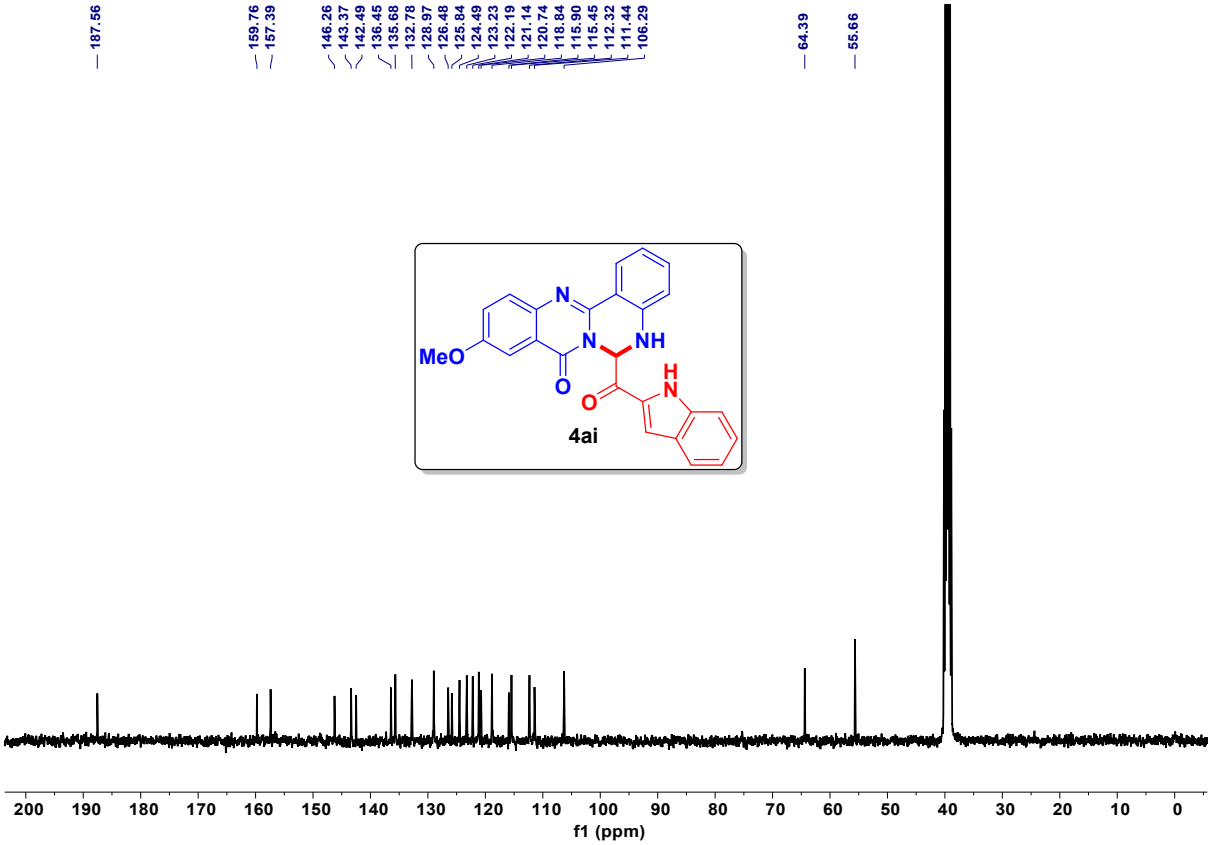


Figure 90. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4ai**

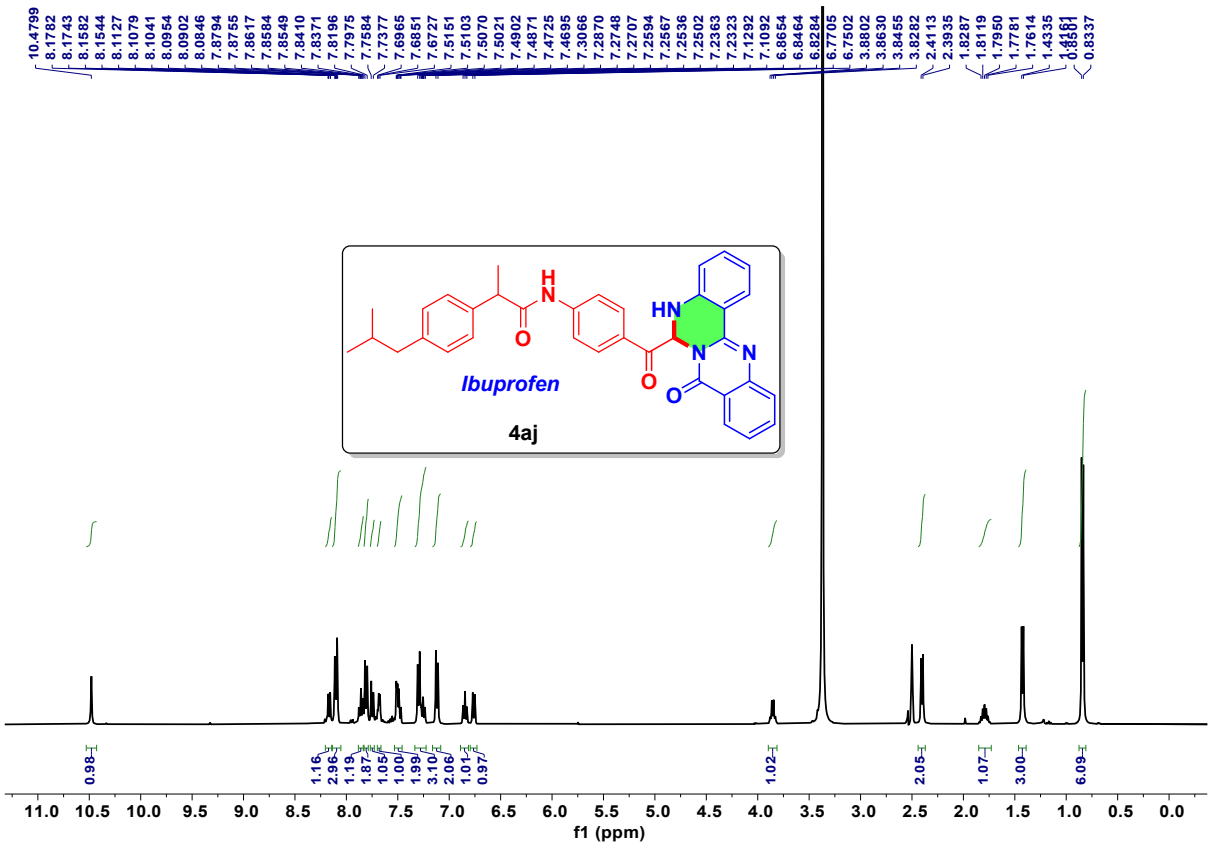


Figure 91. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **4aj**

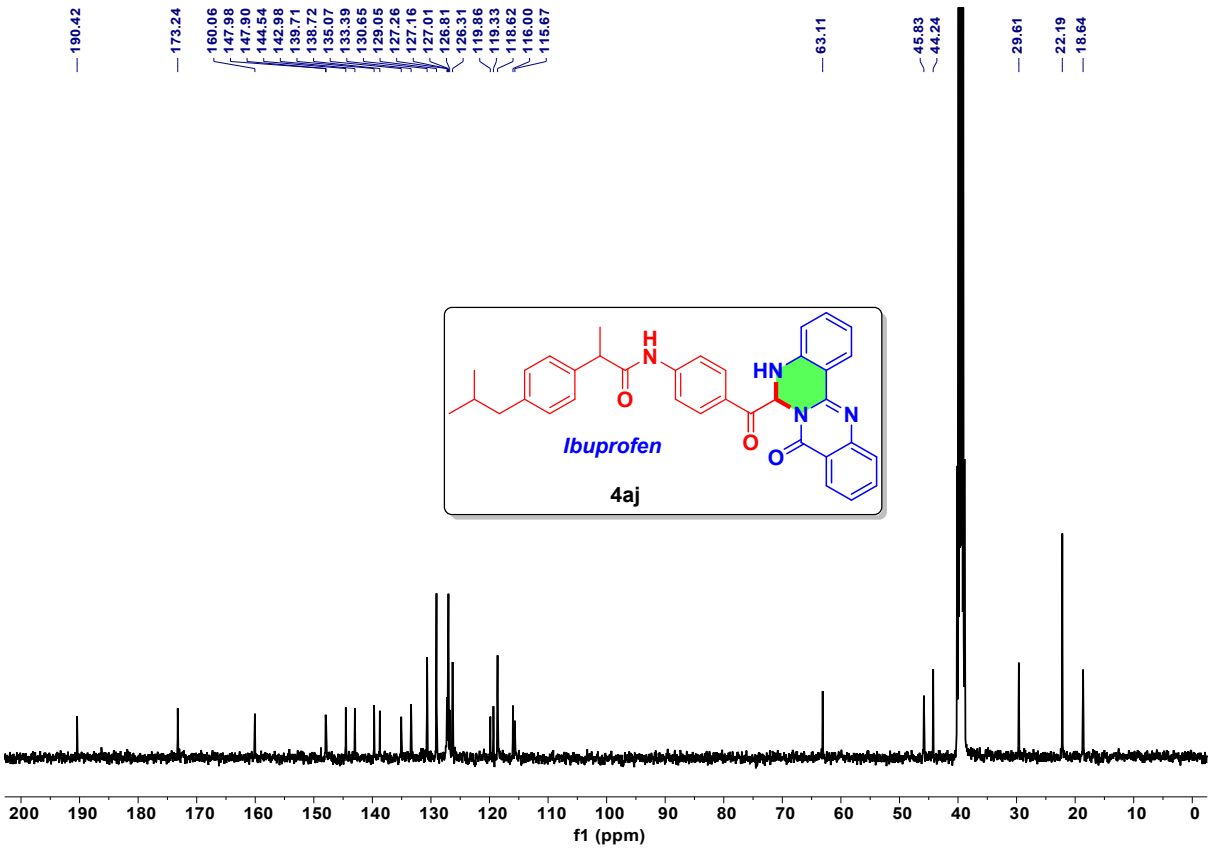


Figure 92. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **4aj**

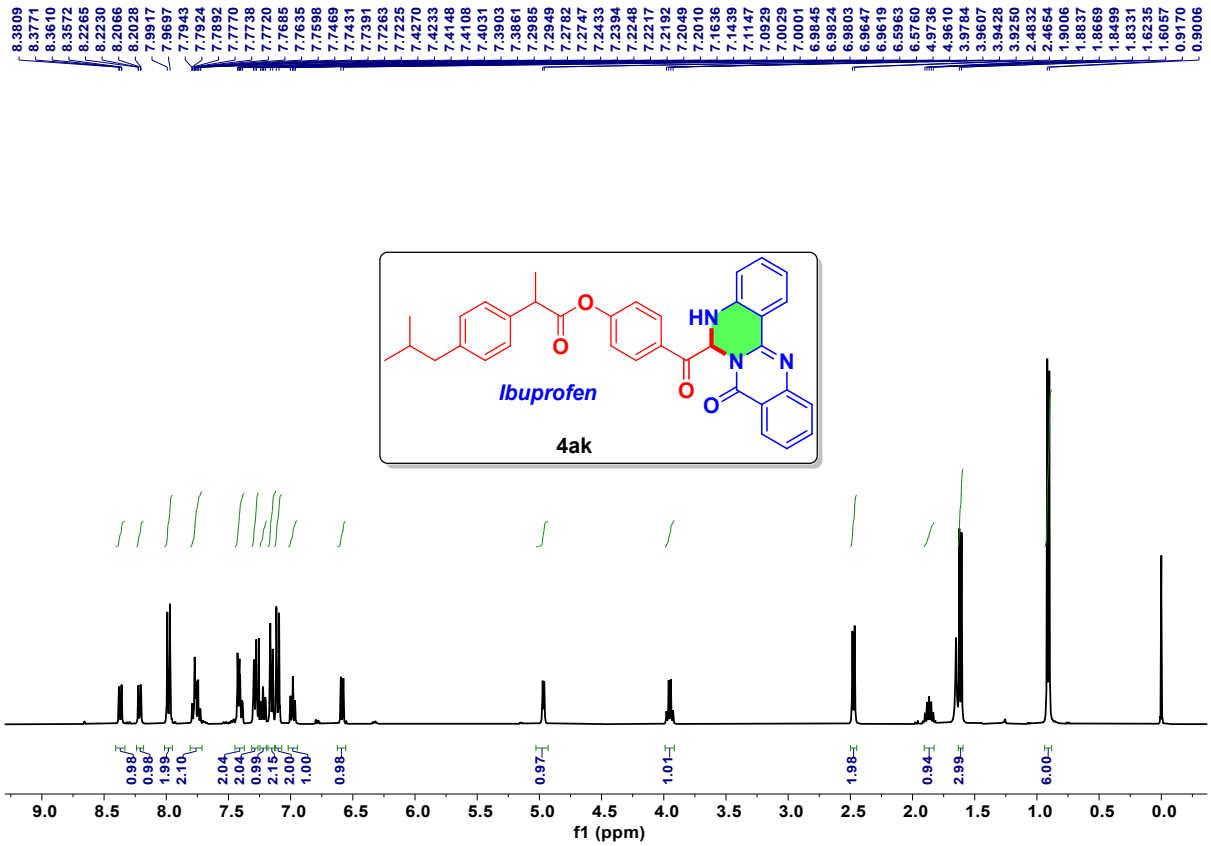


Figure 93. ^{1}H NMR (400 MHz, $CDCl_3$) of compound **4ak**

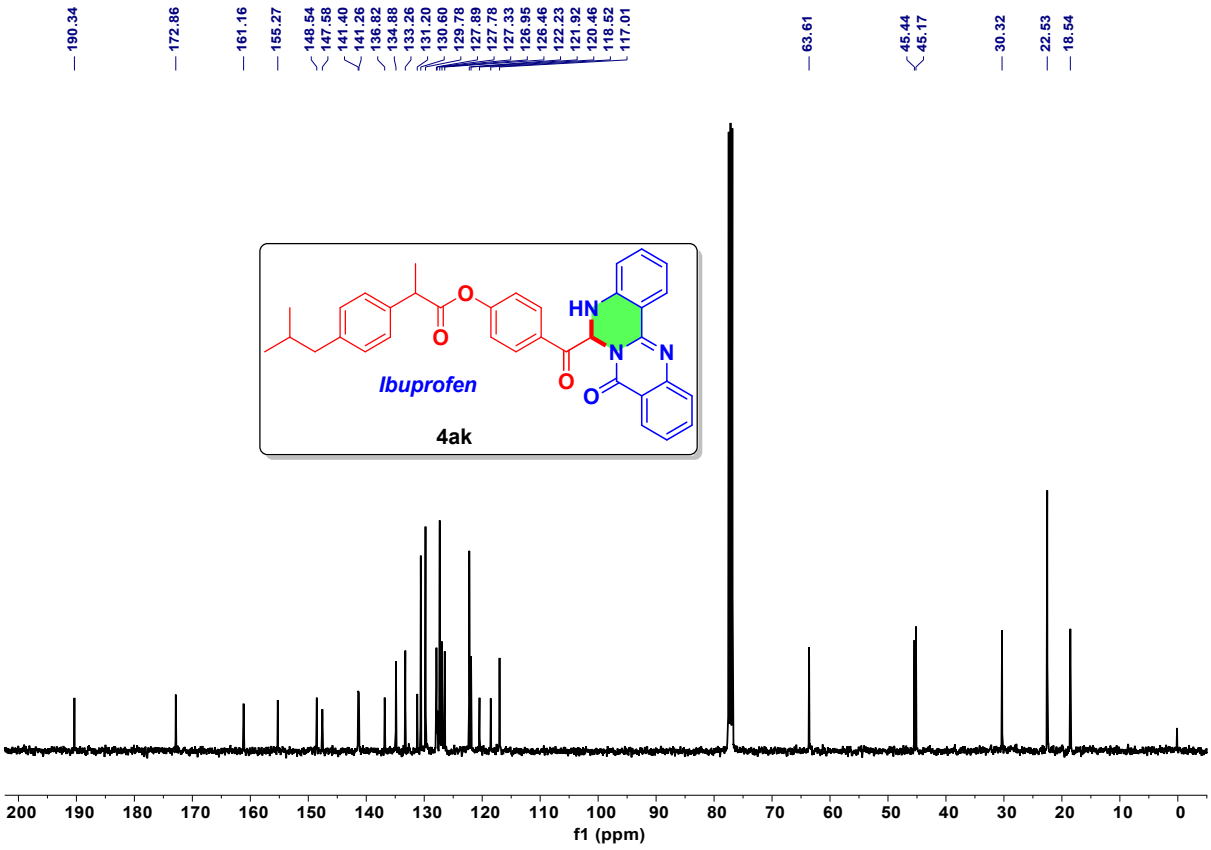


Figure 94. $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) of compound **4ak**

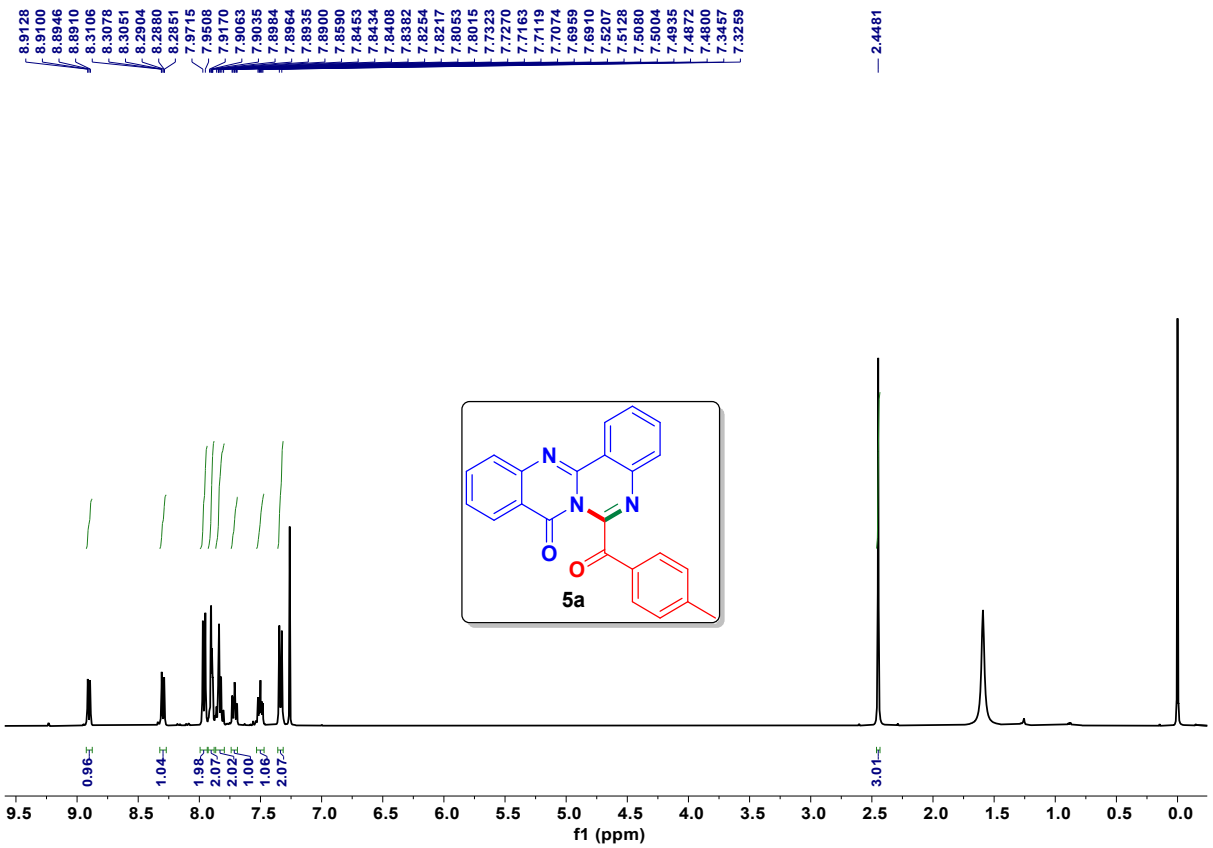


Figure 95. ^{1}H NMR (400 MHz, $CDCl_3$) of compound **5a**

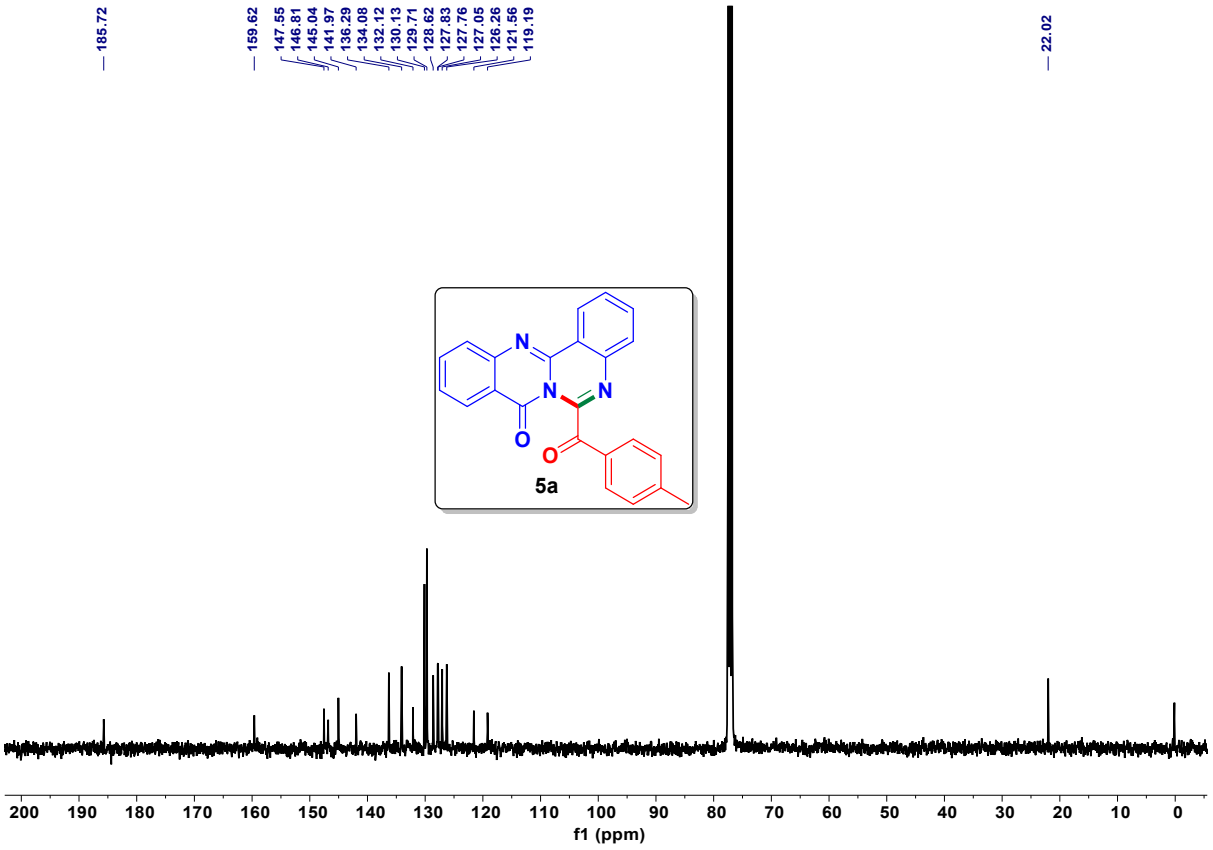


Figure 96. $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) of compound **5a**

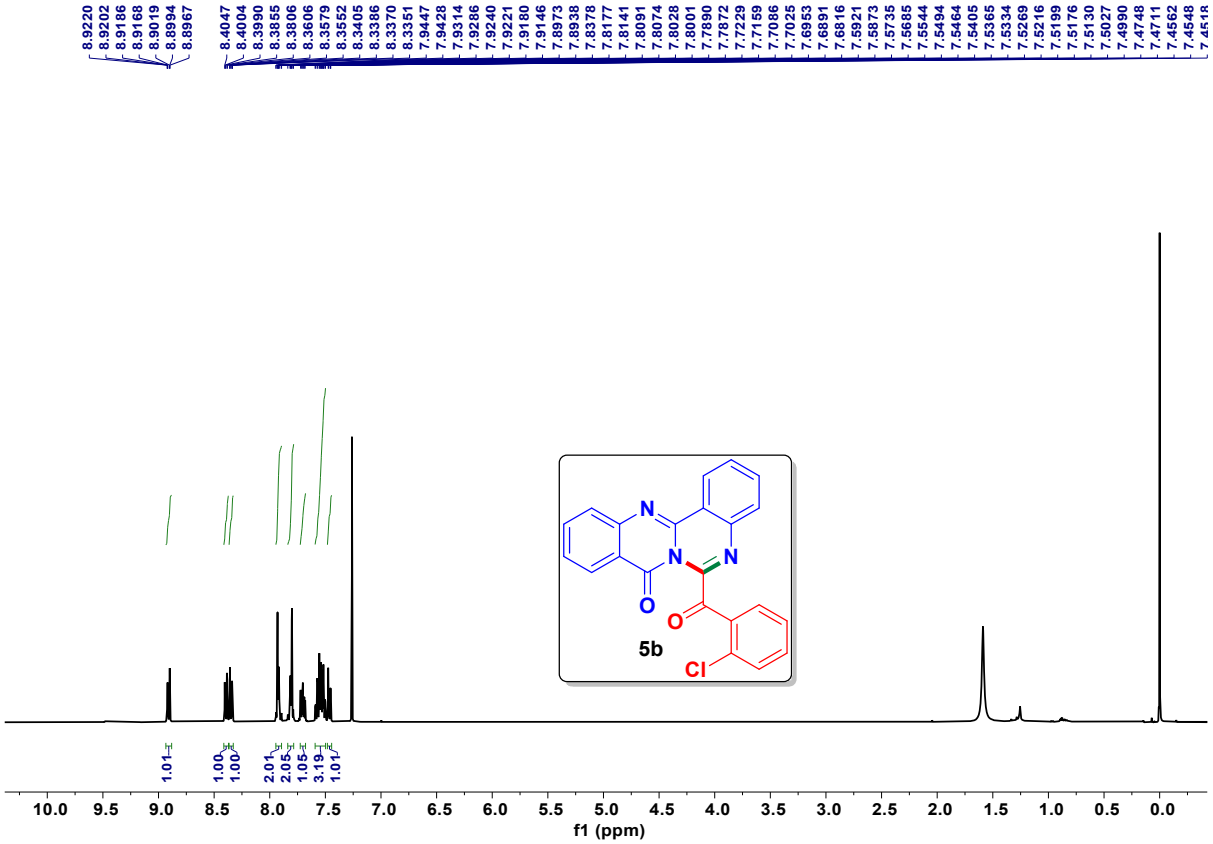


Figure 97. ^{1}H NMR (400 MHz, $CDCl_3$) of compound **5b**

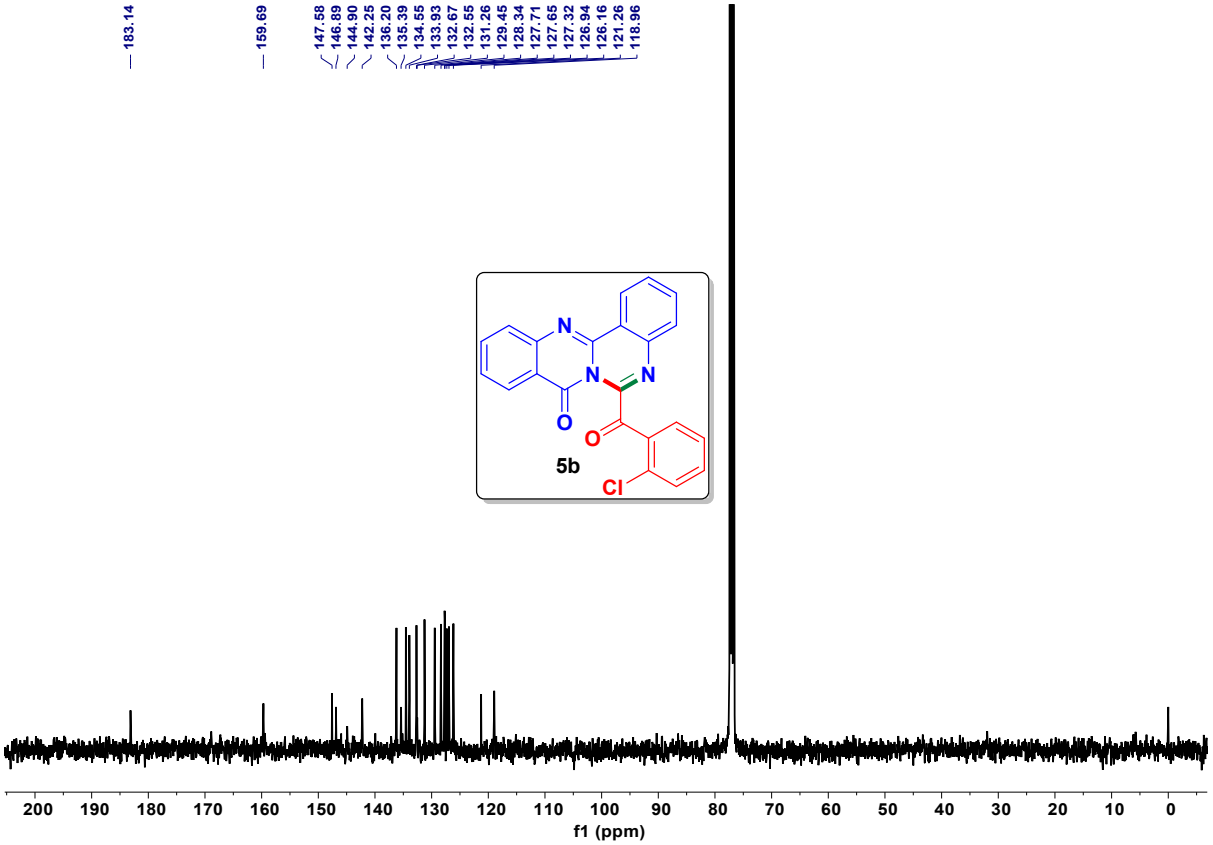


Figure 98. $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) of compound **5b**

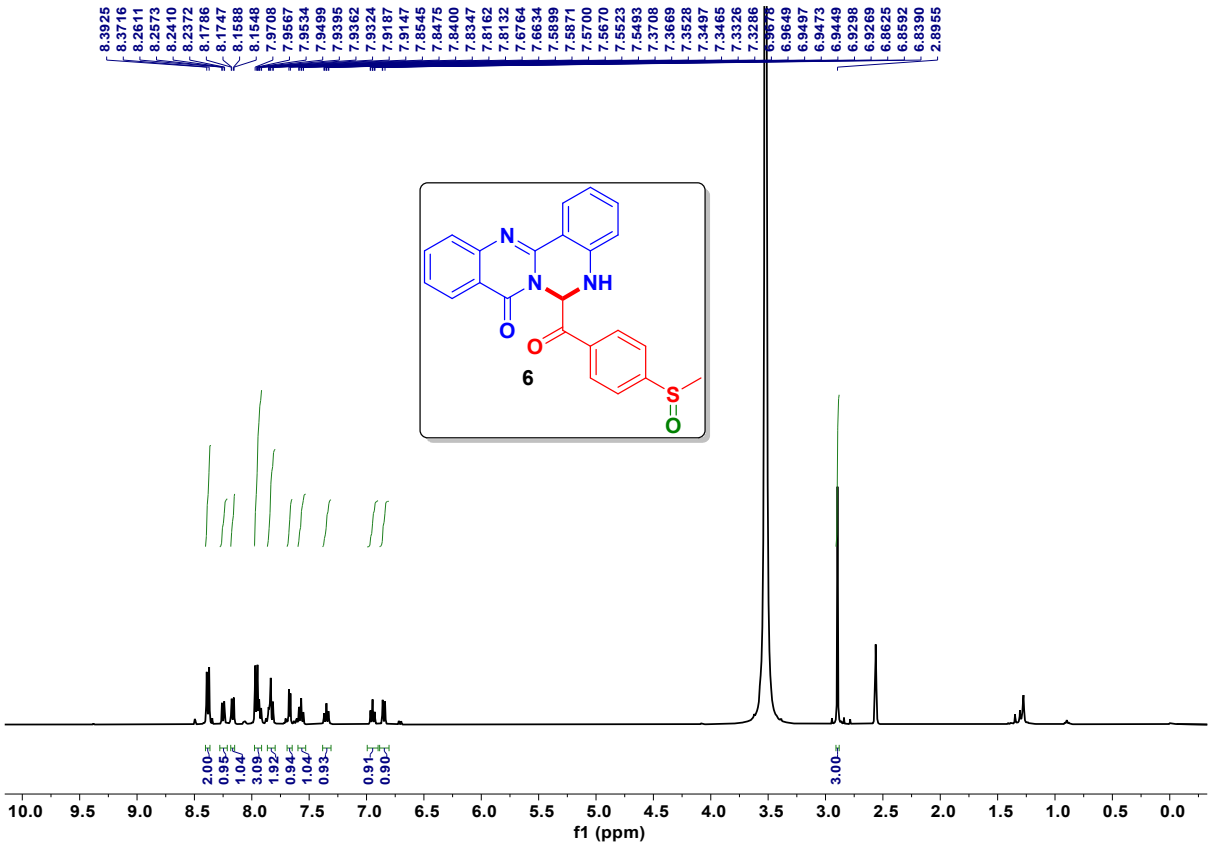


Figure 99. ^{1}H NMR (400 MHz, DMSO-d_6) of compound **6**

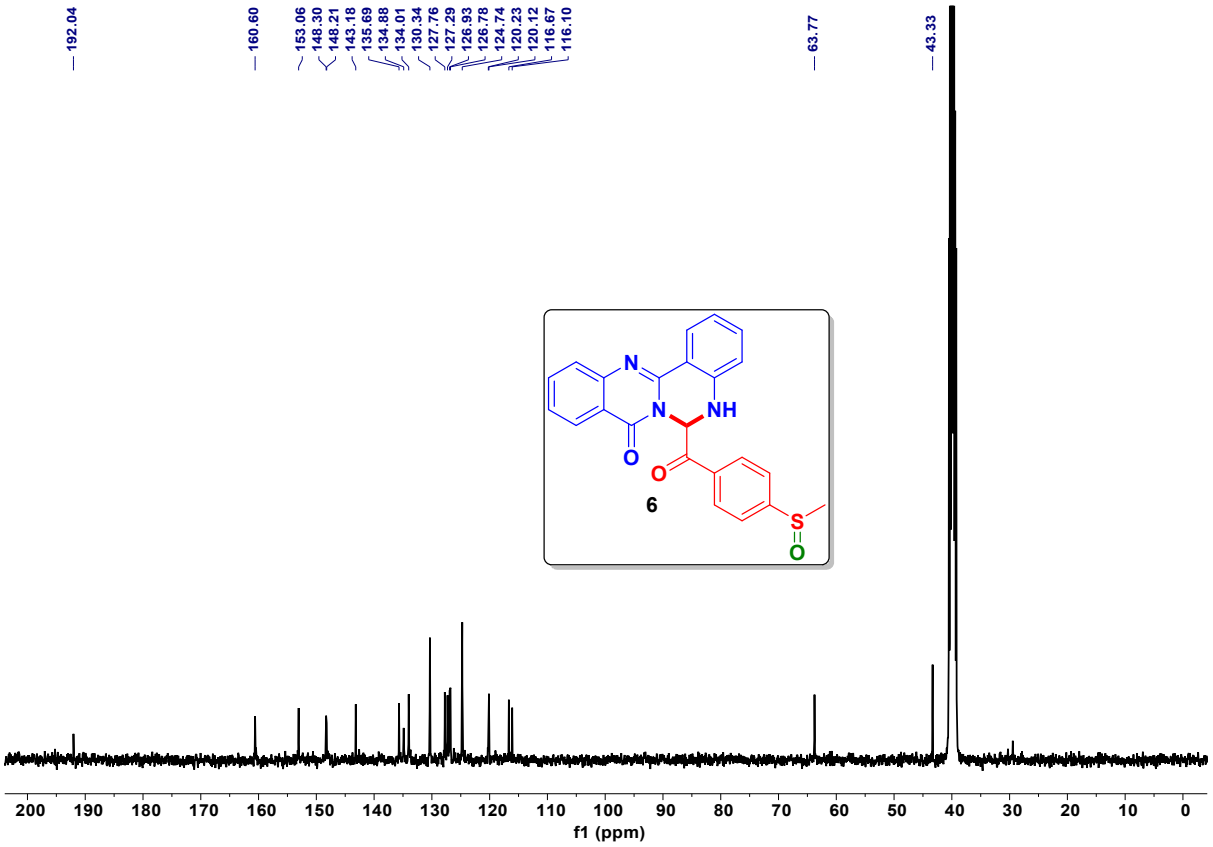


Figure 100. ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) of compound **6**