Supporting Information

Visible-light-induced alkoxycarbonylation/cyclization of 1,7-enynes: synthesis of dihydropyranones containing all-carbon quaternary centers

Jian-Qiang Chen,[a] Qi Chen,[a] Baofu Chen*[a] and Jie Wu*[a,b,c]

[a] Taizhou Central Hospital (Taizhou University Hospital) & School of Pharmaceutical and Materials Engineering, Taizhou University, 999 Donghai Avenue, Taizhou, 318000, China.
[b] State Key Laboratory of Organometallic Chemistry, Shanghai Institute of Organic Chemistry, Chinese Academy of Sciences, Shanghai 200032, China.
[c] School of Chemistry and Chemical Engineering, Henan Normal University, Xinxiang 453007, China.
Corresponding author: chenbf@tzc.edu.cn; jie_wu@fudan.edu.cn

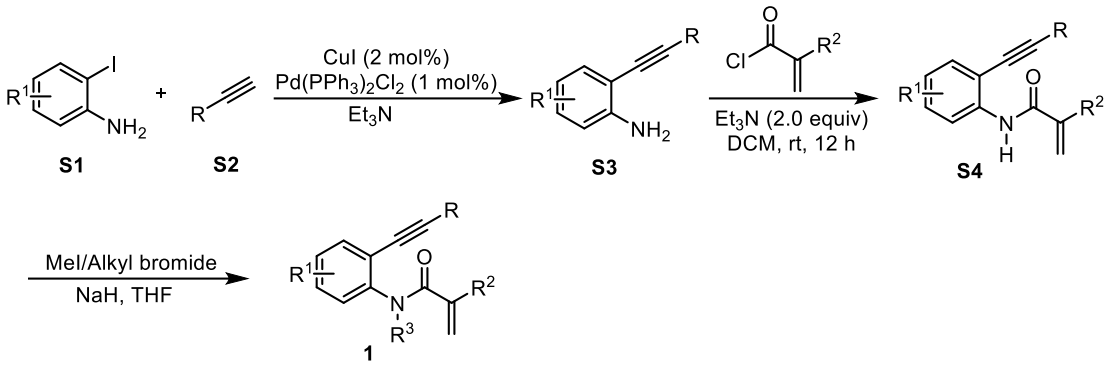
Contents

1. General information

All glassware was thoroughly oven-dried. Chemicals and solvents were either purchased from commercial suppliers or purified by standard techniques. Thin-layer chromatography plates were visualized by exposure to ultraviolet light and/or staining with phosphomolybdic acid followed by heating on a hot plate. Flash chromatography was carried out using silica gel (200−300 mesh). ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker AM-400 (400 MHz). The spectra were recorded in deuterochloroform ($CDCl_3$) as solvent at room temperature, ^{1}H and ^{13}C NMR chemical shifts are reported in ppm relative to the residual solvent peak. The residual solvent signals were used as references and the chemical shifts were converted to the TMS scale ($CDCl_3$: δ_H = 7.26 ppm, δ_C = 77.0 ppm). Data for ^{1}H NMR are reported as follows: chemical shift (δ ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet, dd = doublet, br = broad), integration, coupling constant (Hz) and assignment. Data for ^{13}C NMR are reported as chemical shift. Electrospray–ionisation HRMS data were acquired on a Q–TOF mass spectrometer (Waters SYNAPT G2-Si) LC-MS TOF.

2. General experimental procedure

1. General procedure for the synthesis of substrates 1a-1r and 1t-1w.[1]



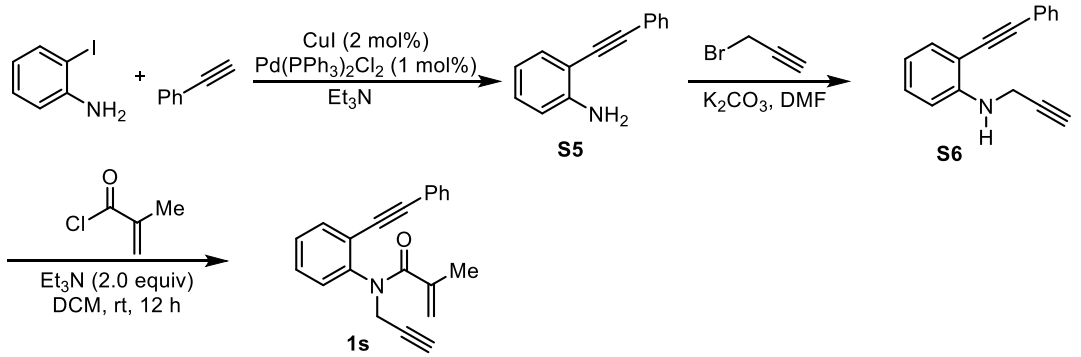
Preparation of the compound (**S3**): Nitrogen was bubbled through neat triethylamine (50 mL) for 20 min. Aryl iodide **S1** (20 mmol), $Pd(PPh_3)_2Cl_2$ (280.8 mg, 0.4 mmol, 2 mol%), CuI (152.4 mg, 0.8 mmol, 4 mol%) and terminal acetyle **S2** (24 mmol) were solubilized in triethylamine. The resulting mixture was stirred at room temperature for 2 h under N_2 atmosphere. Then the crude mixture was filtered through a shot pad of celite and washed with DCM (20 mL) for three times, and the combined organic layer was concentrated under reduced pressure. The resulting crude mixture was purified by flash chromatography using ethyl acetate and petroleum ether as eluent to afford the desired product **S3.**

Preparation of the compound (**S4**): In a 100 mL flask with a stir-bar was charged with **S3** (10 mmol, 1.0 equiv) in DCM (0.5 M). The solution was stirred at 0 °C, triethylamine (2.0 g, 20 mmol, 2.0 equiv) and acryloyl chloride (15 mmol, 1.5 equiv) was added. The solution was warmed up to room temperature and stirred at room temperature for 12 h. The mixture was diluted with DCM (50 mL) and saturated NH_4Cl solution (50 mL). The organic and aqueous layers were separated. The aqueous layer was extracted with DCM (50 mL x 3). The combined organic layer was washed with brine, dried over Na_2SO_4, filtered and concentrated in vacuo to give a residue, which was purified by flash chromatography and then recrystallized from PE/EtOAc to afford the products **S4**.

Preparation of the compound (**1**): Compound **S4** (5 mmol) was solubilized in THF

(20 mL) under N_2 atmosphere. NaH 60% (300 mg, 7.5 mmol) was added portionwise at 0 °C. MeI (778 μL, 12,5 mmol), EtI (1.0 mL, 12.5 mmol), allyl bromide (1.08 mL, 12.5 mmol) or BrBn (1.48 mL, 12.5 mmol) were added dropwise. The resulting mixture was stirred at room temperature for 3 h under N_2 atmosphere. The reaction mixture was quenched with H_2O (40 mL). THF was removed under reduced pressure. The aqueous layer was extracted with EtOAc (3 x 30 mL). The combined organic layers were washed with brine (30 mL) and dried over $MgSO_4$. The crude material was concentrated under reduced pressure and purified by flash column chromatography on silica gel (1:4, EtOAc/hexane).

2. General procedure for the synthesis of substrates 1s.[2]



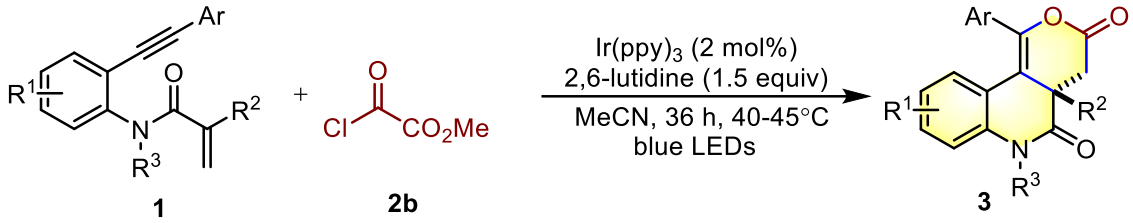
Preparation of the compound (**S5**): Nitrogen was bubbled through neat triethylamine (50 mL) for 20 min. 2-iodoaniline (10 mmol), $Pd(PPh_3)_2Cl_2$ (140.4 mg, 0.2 mmol, 2 mol%), CuI (76.2 mg, 0.4 mmol, 4 mol%) and phenylacetylene (12 mmol) were solubilized in triethylamine. The resulting mixture was stirred at room temperature for 2 h under N_2 atmosphere. Then the crude mixture was filtered through a shot pad of celite and washed with DCM (10 mL) for three times, and the combined organic layer was concentrated under reduced pressure. The resulting crude mixture was purified by flash chromatography using ethyl acetate and petroleum ether as eluent to afford the desired product **S5**.

Preparation of the compound (**S6**): To a mixture of S5 (963 mg, 5 mmol) and potassium carbonate (1.38 g, 10 mmol) in DMF (12 mL) was added propargyl bromide (1.19 g, 10 mmol) under argon atmosphere at room temperature. The resulting mixture was stirred at ambient temperature overnight. Water (12 mL) was added and the mixture was extracted by dichloromethane for 3 times (20 mL×3). The collected organic phases

were dried over magnesium sulfate, then concentrated and purified by silica gel column chromatography (eluent: petrol ether/ ethyl acetate = 75:1) to afford product **S6**.

Preparation of the compound (**1s**): In a 100 mL flask with a stir-bar was charged with **S6** (2 mmol, 1.0 equiv) in DCM (0.5 M). The solution was stirred at 0 °C, triethylamine (405 mg, 4 mmol, 2.0 equiv) and methacryloyl chloride (3 mmol, 1.5 equiv) was added. The solution was warmed up to room temperature and stirred at room temperature for 12 h. The mixture was diluted with DCM (10 mL) and saturated NH_4Cl solution (10 mL). The organic and aqueous layers were separated. The aqueous layer was extracted with DCM (10 mL x 3). The combined organic layer was washed with brine, dried over Na_2SO_4, filtered and concentrated in vacuo to give a residue, which was purified by flash chromatography and then recrystallized from PE/EtOAc to afford the products **1s**.

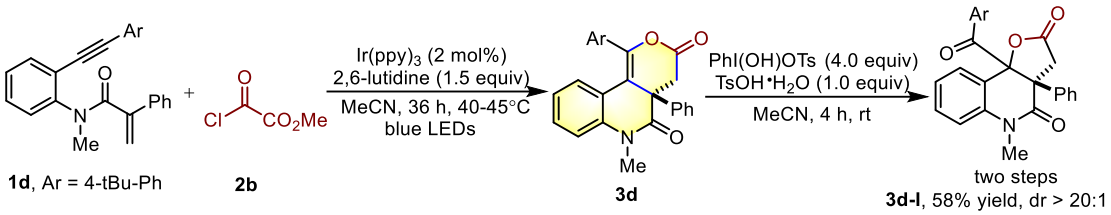
3. General procedure for dihydropyranone



All optimization reactions were set up in a glove box under N_2 atmosphere. Substrate **1** (0.2 mmol, 1.0 equiv), methyl chlorooxoacetate **2b** (0.6 mmol, 3.0 equiv) and 2,6-lutidine (0.3 mmol, 1.5 equiv) were added to a solution of photocatalyst $Ir(ppy)_3$ (2 mol %) in dry MeCN (4 mL) at room temperature. The heterogenous mixture was placed in the irradiation apparatus equipped with blue LEDs. The resulting mixture was stirred for 36 h. Upon completion of the reaction, the mixture was diluted with ethyl acetate (30 mL), washed with brine (10 ml x 2), dried with Na_2SO_4 and the solvent was evaporated. The crude product was purified by column chromatography on silica gel to afford the desired product **3**.

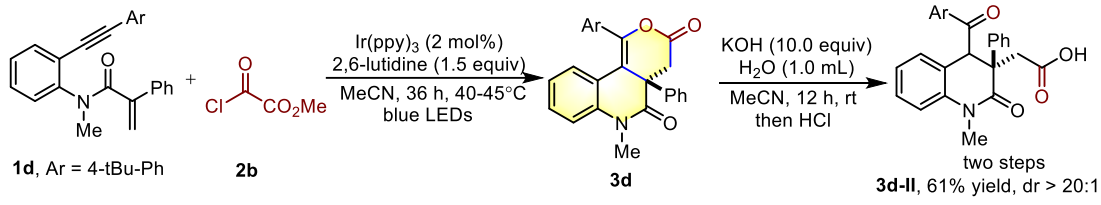
4. Synthetic applications

1. General procedure for the synthesis of 3d-I



All optimization reactions were set up in a glove box under N_2 atmosphere. Substrate **1d** (0.2 mmol, 1.0 equiv), methyl chlorooxoacetate **2b** (0.6 mmol, 3.0 equiv) and 2,6-lutidine (0.3 mmol, 1.5 equiv) were added to a solution of photocatalyst $Ir(ppy)_3$ (2 mol %) in dry MeCN (4 mL) at room temperature. The heterogenous mixture was placed in the irradiation apparatus equipped with blue LEDs. The resulting mixture was stirred for 36 h. Upon completion of the reaction, the mixture was diluted with methyl acetate (30 mL), washed with brine (10 x 3 mL), dried with Na_2SO_4. After evaporation of the solvent, the crude product was used in the following step without further purification. To a solution of crude product in MeCN (4.0 mL) was added *p*-TsOH·H_2O (0.2 mmol, 1.0 equiv) and [(hydroxy)-tosyloxyiodo]benzene (0.8 mmol, 4.0 equiv) in one portion. The reaction mixture was stirred at rt for 4 h, and then the reaction was quenched with the addition of brine (15 mL) and diluted with EtOAc (30 mL). The combined organic layers were washed with brine (3 × 10 mL), dried over Na_2SO_4, filtered and concentrated under reduced pressure. The resulting residue was purified by flash column chromatography with gradient eluents (petroleum ether / ethyl acetate = 3/1) to provide compound **3d-I**.

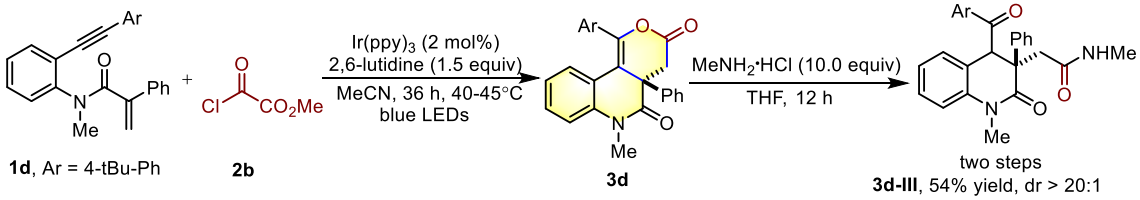
2. General procedure for the synthesis of 3d-II



All optimization reactions were set up in a glove box under N_2 atmosphere. Substrate **1d** (0.2 mmol, 1.0 equiv), methyl chlorooxoacetate **2b** (0.6 mmol, 3.0 equiv) and 2,6-

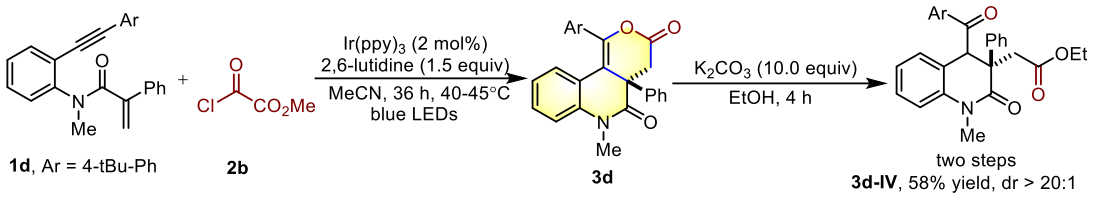
lutidine (0.3 mmol, 1.5 equiv) were added to a solution of photocatalyst $Ir(ppy)_3$ (2 mol %) in dry MeCN (4 mL) at room temperature. The heterogenous mixture was placed in the irradiation apparatus equipped with blue LEDs. The resulting mixture was stirred for 36 h. Upon completion of the reaction, KOH (2.0 mmol, 10.0 equiv) and H_2O (1 mL) were added. The reaction mixture was stirred at rt for 12 h, and then the reaction was quenched with 6 M HCl (2.0 mL) and diluted with EtOAc (30 mL). The combined organic layers were washed with brine (3 × 10 mL), dried over Na_2SO_4, filtered and concentrated under reduced pressure. The resulting residue was purified by flash column chromatography with gradient eluents (petroleum ether / ethyl acetate = 1/1) to provide compound **3d-II**.

3. General procedure for the synthesis of 3d-III



All optimization reactions were set up in a glove box under N_2 atmosphere. Substrate **1d** (0.2 mmol, 1.0 equiv), methyl chlorooxoacetate **2b** (0.6 mmol, 3.0 equiv) and 2,6-lutidine (0.3 mmol, 1.5 equiv) were added to a solution of photocatalyst $Ir(ppy)_3$ (2 mol %) in dry MeCN (4 mL) at room temperature. The heterogenous mixture was placed in the irradiation apparatus equipped with blue LEDs. The resulting mixture was stirred for 36 h. Upon completion of the reaction, the mixture was diluted with methyl acetate (30 mL), washed with brine (10 x 3 mL), dried with Na_2SO_4. After evaporation of the solvent, the crude product was used in the following step without further purification. To a solution of crude product in THF (4.0 mL) was added methylamine hydrochloride (2.0 mmol, 10.0 equiv) in one portion. The reaction mixture was stirred at rt for 12 h, and then the reaction was quenched with the addition of brine (15 mL) and diluted with EtOAc (30 mL). The combined organic layers were washed with brine (3 × 10 mL), dried over Na_2SO_4, filtered and concentrated under reduced pressure. The resulting residue was purified by flash column chromatography with gradient eluents (petroleum ether / ethyl acetate = 3/2) to provide compound **3d-III**.

4. General procedure for the synthesis of 3d-IV



All optimization reactions were set up in a glove box under N_2 atmosphere. Substrate **1d** (0.2 mmol, 1.0 equiv), methyl chlorooxoacetate **2b** (0.6 mmol, 3.0 equiv) and 2,6-lutidine (0.3 mmol, 1.5 equiv) were added to a solution of photocatalyst $Ir(ppy)_3$ (2 mol %) in dry MeCN (4 mL) at room temperature. The heterogenous mixture was placed in the irradiation apparatus equipped with blue LEDs. The resulting mixture was stirred for 36 h. Upon completion of the reaction, the mixture was diluted with methyl acetate (30 mL), washed with brine (10 x 3 mL), dried with Na_2SO_4. After evaporation of the solvent, the crude product was used in the following step without further purification. To a solution of crude product in EtOH (4.0 mL) was added K_2CO_3 (2.0 mmol, 10.0 equiv) in one portion. The reaction mixture was stirred at rt for 4 h, and then the reaction was quenched with the addition of brine (15 mL) and diluted with EtOAc (30 mL). The combined organic layers were washed with brine (3 × 10 mL), dried over Na_2SO_4, filtered and concentrated under reduced pressure. The resulting residue was purified by flash column chromatography with gradient eluents (petroleum ether / ethyl acetate = 4/1) to provide compound **3d-IV**.

5. Devices for the photocatalytic reactions

Irradiation of visible light was performed with a 36 W Blue LED strip. All photocatalyzed alkoxycarbonylation/cyclization reactions were carried out at room temperature (r.t.) with fan-assisted cooling to maintain a temperature of approximately 40-45 °C. The distance between tube and lamp was approximately 3 cm.

Manufacture of the light source: LED strip

Manufacturer: Greethink

Model: GT-5050-Blue

Wavelength of peak intensity: 460-470 nm

Material of the irradiation vessel: borosilicate glass

Distance of the irradiation vessel from the light source: approximately 3 cm.

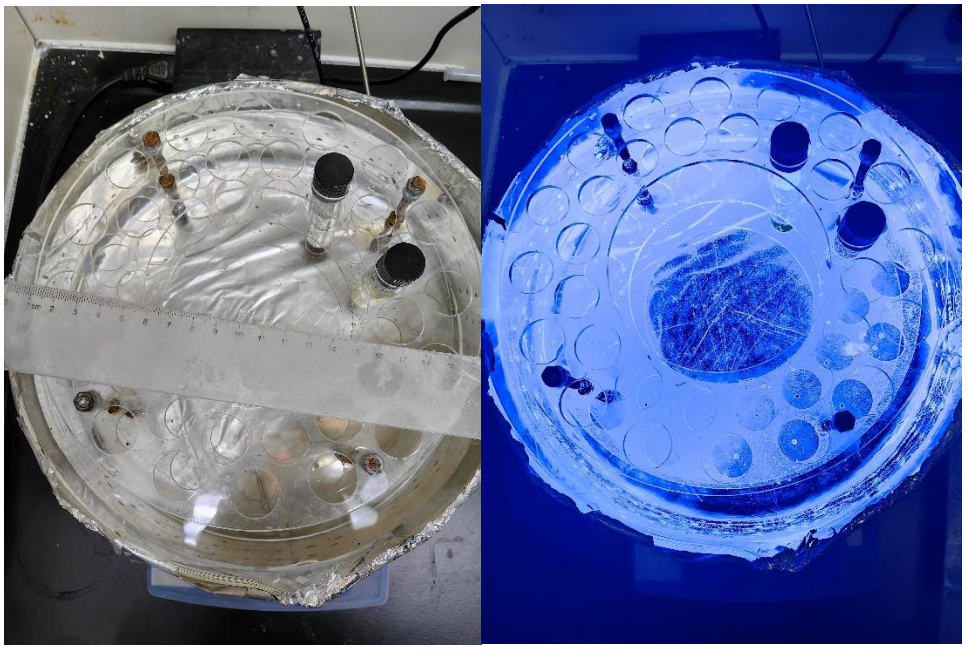
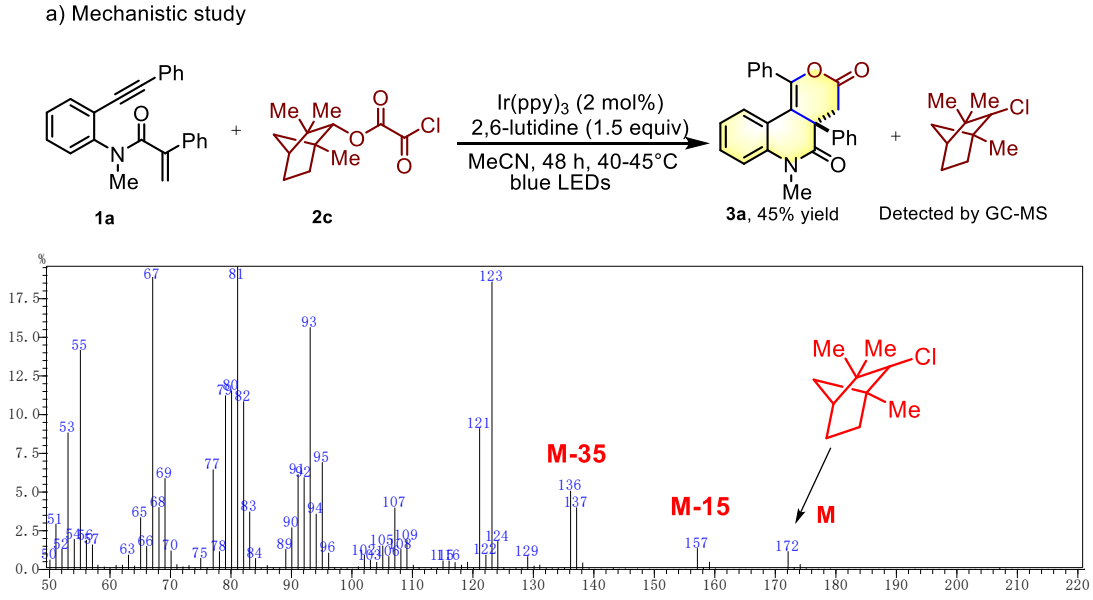
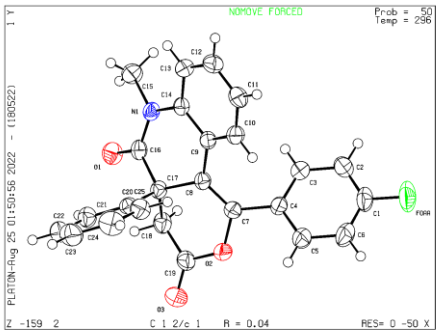


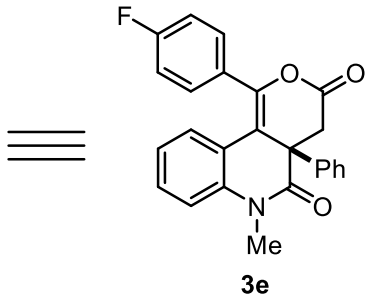
Figure S1. The reaction set-up for the photocatalytic reactions

6. Mechanistic study



7. X-ray data for compound 3e (CCDC 2203307)





3e

Bond precision: C-C = 0.0022 A Wavelength=0.71073

Cell: a=23.665(3) b=9.9238(11) c=18.980(2)
alpha=90 beta=117.870(2) gamma=90

Temperature: 296 K

	Calculated	Reported
Volume	3940.4(8)	3940.4(8)
Space group	C 2/c	C 1 2/c 1
Hall group	-C 2yc	-C 2yc
Moiety formula	C25 H18 F N O3	C25 H18 F N O3
Sum formula	C25 H18 F N O3	C25 H18 F N O3
Mr	399.40	399.40
Dx,g cm-3	1.347	1.347
Z	8	8
Mu (mm-1)	0.095	0.095
F000	1664.0	1664.0
F000'	1664.87	
h,k,lmax	28,11,22	28,11,22
Nref	3472	3462
Tmin,Tmax	0.976,0.979	
Tmin'	0.976	

Correction method= Not given

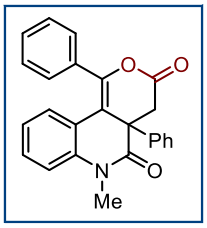
Data completeness= 0.997 Theta(max)= 24.997

R(reflections)= 0.0378(2768) wR2(reflections)= 0.1155(3462)

S = 1.041 Npar = 272

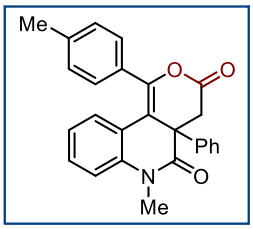
8. Characterization of new substrates and all products

6-methyl-1,4a-diphenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3a)



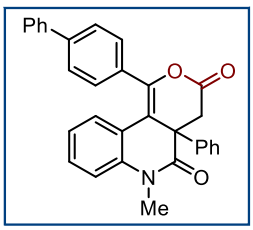
Purification by flash chromatography (DCM). Colorless oil; 64% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.62−7.59 (m, 2H), 7.44−7.35 (m, 3H), 7.31 (d, *J* = 7.1 Hz, 1H), 7.26−7.17 (m, 3H), 7.10 (td, *J* = 7.8, 1.3 Hz, 1H), 6.96 (dd, *J* = 7.8, 1.0 Hz, 1H), 6.89 (d, *J* = 8.0 Hz, 1H), 6.71 (td, *J* = 7.6, 0.7 Hz, 1H), 3.49−3.44 (m, 4H), 3.33 (d, *J* = 15.8 Hz, 1H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 168.3, 166.2, 148.9, 138.5, 137.3, 132.6, 130.1, 129.9, 129.7, 129.2, 128.7, 128.6, 128.2, 126.1, 123.0, 120.6, 115.4, 110.4, 50.4, 40.2, 30.8; HRMS (ESI) for $C_{25}H_{20}NO_3$ $[M+H]^+$ calcd. 382.1438, found 382.1440.

6-methyl-4a-phenyl-1-(p-tolyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3b)



Purification by flash chromatography (DCM). Colorless oil; 52% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.49 (d, *J* = 8.0 Hz, 2H), 7.30 (d, *J* = 7.4 Hz, 2H), 7.25−7.15 (m, 5H), 7.10 (t, *J* = 7.8 Hz, 1H), 7.01 (d, *J* = 8.0 Hz, 1H), 6.88 (d, *J* = 8.2 Hz, 1H), 6.73 (t, *J* = 7.5 Hz, 1H), 3.47−3.42 (m, 4H), 3.32 (d, *J* = 15.8 Hz, 1H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 168.3, 166.4, 149.0, 140.4, 138.4, 137.3, 129.8, 129.6, 129.6, 129.3, 129.2, 128.6, 128.2, 126.1, 123.0, 120.9, 115.3, 109.7, 50.4, 40.2, 30.8, 21.5; HRMS (ESI) for $C_{26}H_{22}NO_3$ $[M+H]^+$ calcd. 396.1594, found 396.1605.

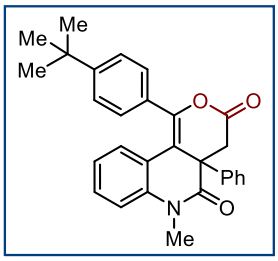
1-([1,1'-biphenyl]-4-yl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3c)



Purification by flash chromatography (DCM). White solid; mp 241-243°C; 53% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.68 (d, *J* = 8.1 Hz, 2H), 7.65−7.59 (m, 4H), 7.47 (t, *J* = 7.5 Hz, 2H), 7.40 (d, *J* = 7.2 Hz, 1H), 7.33 (d, *J* = 7.6 Hz, 2H), 7.27−7.19 (m, 3H), 7.10 (t, *J* = 7.9 Hz, 2H), 6.91 (d, *J* = 8.1 Hz, 1H), 6.75 (t, *J* = 7.6 Hz, 1H),

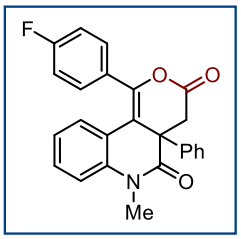
3.49−3.45 (m, 4H), 3.35 (d, J = 15.9 Hz, 1H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 168.3, 166.2, 148.5, 142.7, 139.9, 138.5, 137.3, 131.4, 130.2, 129.9, 129.2, 128.9, 128.8, 128.3, 127.9, 127.1, 127.0, 126.1, 123.1, 120.7, 115.4, 110.5, 50.5, 40.2, 30.8; HRMS (ESI) for $C_{31}H_{24}NO_3$ $[M+H]^+$ calcd. 458.1751, found 458.1762.

1-(4-(tert-butyl)phenyl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3d)



Purification by flash chromatography (DCM). Colorless oil; 65% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.53 (d, J = 8.3 Hz, 2H), 7.37 (d, J = 8.4 Hz, 2H), 7.31 (d, J = 7.5 Hz, 2H), 7.23−7.16 (m, 3H), 7.09−7.03 (m, 2H), 6.87 (d, J = 8.2 Hz, 1H), 6.73 (t, J = 7.6 Hz, 1H), 3.47−3.42 (m, 4H), 3.31 (d, J = 15.8 Hz, 1H), 1.33 (s, 9H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 168.3, 166.3, 153.5, 148.9, 138.4, 137.3, 129.8, 129.6, 129.4, 129.1, 128.6, 128.2, 126.1, 125.5, 123.0, 120.8, 115.3, 109.8, 50.4, 40.2, 34.8, 31.1, 30.7; HRMS (ESI) for $C_{29}H_{28}NO_3$ $[M+H]^+$ calcd. 438.2064, found 438.2075.

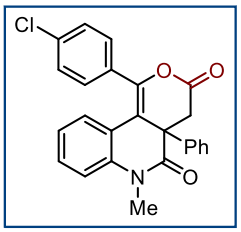
1-(4-fluorophenyl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3e)



Purification by flash chromatography (DCM). White solid; mp 218-220℃; 59% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.62−7.57 (m, 2H), 7.29 (d, J = 7.4 Hz, 2H), 7.26−7.18 (m, 3H), 7.14−7.04 (m, 3H), 6.96 (d, J = 7.3 Hz, 1H), 6.90 (d, J = 8.2 Hz, 1H), 6.75 (t, J = 7.5 Hz, 1H), 3.48−3.43 (m, 4H), 3.33 (d, J = 15.8 Hz, 1H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 168.2, 166.2, 163.5 (d, J = 249.7 Hz), 147.8, 138.6, 137.2, 131.8 (d, J = 8.3 Hz), 129.7, 129.2, 128.9, 128.7 (d, J = 3.4 Hz), 128.3, 126.1, 123.1, 120.4, 115.8 (d, J = 21.8 Hz), 115.5, 110.5, 50.4, 40.2, 30.8;HRMS (ESI) for $C_{25}H_{19}FNO_3$ $[M+H]^+$ calcd. 400.1343, found 400.1350.

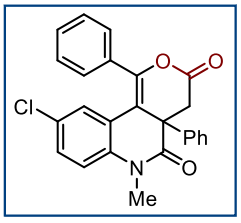
1-(4-chlorophenyl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-

dione (3f)



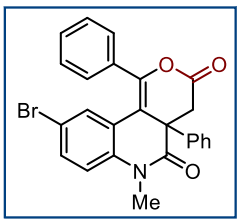
Purification by flash chromatography (DCM). White solid; mp 174-176°C; 30% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.55 (d, *J* = 8.5 Hz, 2H), 7.34 (d, *J* = 8.5 Hz, 2H), 7.30−7.19 (m, 5H), 7.15−7.10 (m, 1H), 6.97 (d, *J* = 7.7 Hz, 1H), 6.90 (d, *J* = 8.2 Hz, 1H), 6.76 (t, *J* = 7.4 Hz, 1H), 3.48−3.43 (m, 4H), 3.32 (d, *J* = 15.8 Hz, 1H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 168.2, 166.0, 147.6, 138.6, 137.1, 136.1, 131.1, 129.8, 129.2, 129.1, 128.9, 128.3, 126.1, 123.2, 120.3, 115.5, 111.1, 50.5, 40.2, 30.8; HRMS (ESI) for $C_{25}H_{19}ClNO_3$ $[M+H]^+$ calcd. 416.1048, found 416.1057.

9-chloro-6-methyl-1,4a-diphenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3g)



Purification by flash chromatography (DCM). Colorless oil; 55% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.59 (d, *J* = 7.2 Hz, 2H), 7.50−7.39 (m, 3H), 7.29−7.21 (m, 5H), 7.06 (dd, *J* = 8.8, 2.2 Hz, 1H), 6.86 (d, *J* = 2.2 Hz, 1H), 6.82 (d, *J* = 8.8 Hz, 1H), 3.48−3.43 (m, 4H), 3.33 (d, *J* = 15.9 Hz, 1H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 168.0, 165.8, 150.1, 137.1, 136.9, 131.9, 130.6, 129.6, 129.4, 129.3, 128.8, 128.5, 128.5, 128.3, 126.0, 122.3, 116.6, 109.2, 50.2, 40.1, 30.9; HRMS (ESI) for $C_{25}H_{19}ClNO_3$ $[M+H]^+$ calcd. 416.1048, found 416.1057.

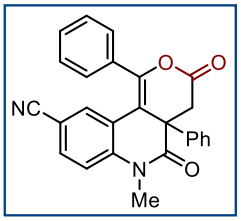
9-bromo-6-methyl-1,4a-diphenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3h)



Purification by flash chromatography (DCM). Colorless oil; 51% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.59 (d, *J* = 7.1 Hz, 2H), 7.50−7.40 (m, 3H), 7.29−7.21 (m, 5H), 7.20 (dd, *J* = 8.8, 2.2 Hz, 1H), 7.00 (d, *J* = 2.2 Hz, 1H), 6.76 (d, *J* = 8.8 Hz, 1H), 3.48−3.43 (m, 4H), 3.32 (d, *J* = 15.9 Hz, 1H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 168.0, 165.8, 150.1, 137.6, 136.9, 132.2, 131.9, 131.4, 130.6, 129.6, 129.4, 128.8, 128.5, 126.0, 122.6, 116.9, 115.7, 109.1, 50.2, 40.1, 30.9; HRMS (ESI) for $C_{25}H_{19}BrNO_3$

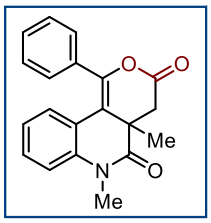
$[M+H]^+$ calcd. 460.0543, found 460.0548.

6-methyl-3,5-dioxo-1,4a-diphenyl-4,4a,5,6-tetrahydro-3H-pyrano[4,3-c]quinoline-9-carbonitrile (3i)



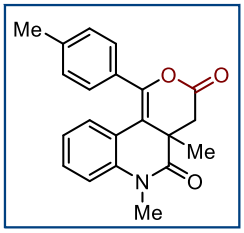
Purification by flash chromatography (DCM). White solid; mp 195-197℃; 57% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.56 (d, *J* = 7.5 Hz, 2H), 7.50 (t, *J* = 7.3 Hz, 1H), 7.44−7.35 (m, 3H), 7.30−7.21 (m, 5H), 7.19 (s, 1H), 6.97 (d, *J* = 8.6 Hz, 1H), 3.49 (s, 3H), 3.44 (d, *J* = 15.8 Hz, 1H), 3.35 (d, *J* = 15.8 Hz, 1H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 168.3, 165.2, 151.0, 141.8, 136.4, 133.4, 132.2, 131.4, 131.1, 129.6, 129.5, 129.1, 128.7, 125.8, 121.9, 118.0, 116.0, 107.9, 106.4, 50.2, 40.0, 30.9; HRMS (ESI) for $C_{26}H_{19}N_2O_3$ $[M+H]^+$ calcd. 407.1390, found 407.1400.

4a,6-dimethyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3j)



Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). White solid; mp 103-105℃; 60% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.49 (d, *J* = 7.0 Hz, 2H), 7.38−7.30 (m, 3H), 7.22 (td, *J* = 7.8, 1.1 Hz, 1H), 7.04 (d, *J* = 8.2 Hz, 1H), 6.87 (dd, *J* = 7.8, 0.9 Hz, 1H), 6.75 (t, *J* = 7.4 Hz, 1H), 3.45 (s, 3H), 3.23 (d, *J* = 15.9 Hz, 1H), 2.98 (d, *J* = 15.9 Hz, 1H), 1.29 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.6, 167.1, 146.5, 138.6, 132.8, 130.0, 129.8, 129.6, 128.9, 128.5, 122.9, 119.6, 115.1, 112.5, 42.1, 37.4, 30.4, 23.1; HRMS (ESI) for $C_{20}H_{17}NO_3Na$ $[M+Na]^+$ calcd. 342.1101, found 342.1110.

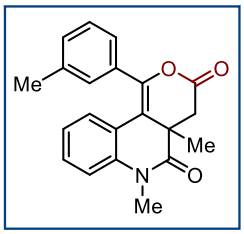
4a,6-dimethyl-1-(p-tolyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3k)



Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). Colorless oil; 57% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.37 (d, *J* = 8.1 Hz, 2H), 7.24−7.19 (m, 1H), 7.12 (d, *J* = 8.1 Hz, 2H), 7.03 (d, *J* = 8.2 Hz, 1H), 6.92 (dd, *J* = 7.8, 1.2 Hz, 1H), 6.76 (t, *J* = 7.4 Hz, 1H), 3.44 (s, 3H), 3.21 (d, *J* = 15.9 Hz, 1H), 2.96 (d, *J* = 15.9 Hz, 1H), 2.37 (s, 3H), 1.27 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.7,

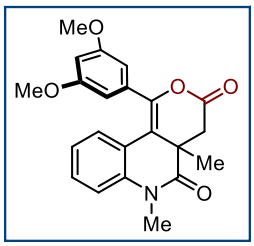
167.2, 146.6, 140.1, 138.6, 130.0, 129.8, 129.5, 129.2, 128.7, 122.9, 119.9, 115.1, 111.9, 42.1, 37.4, 30.4, 23.1, 21.4; HRMS (ESI) for $C_{21}H_{19}NO_3Na$ $[M+Na]^+$ calcd. 356.1257, found 356.1266.

4a,6-dimethyl-1-(m-tolyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3l)



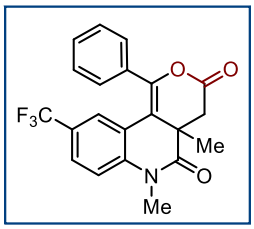
Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). Colorless oil; 57% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.35 (s, 1H), 7.25−7.17 (m, 4H), 7.03 (d, *J* = 8.2 Hz, 1H), 6.89 (dd, *J* = 8.0, 1.3 Hz, 1H), 6.75 (t, *J* = 7.4 Hz, 1H), 3.44 (s, 3H), 3.22 (d, *J* = 15.9 Hz, 1H), 2.97 (d, *J* = 15.9 Hz, 1H), 2.32 (s, 3H), 1.28 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.7, 167.1, 146.7, 138.5, 138.4, 132.7, 130.6, 130.0, 130.0, 128.8, 128.3, 126.9, 122.8, 119.7, 115.0, 112.3, 42.1, 37.4, 30.4, 23.1, 21.3; HRMS (ESI) for $C_{21}H_{19}NO_3Na$ $[M+Na]^+$ calcd. 356.1257, found 356.1264.

1-(3,5-dimethoxyphenyl)-4a,6-dimethyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3m)



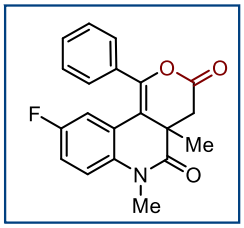
Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). Colorless oil; 53% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.24−7.19 (m, 1H), 7.02 (d, *J* = 8.1 Hz, 1H), 6.95 (dd, *J* = 7.8, 1.2 Hz, 1H), 6.79 (t, *J* = 7.6 Hz, 1H), 6.61 (d, *J* = 2.2 Hz, 2H), 6.47 (t, *J* = 2.2 Hz, 1H), 3.71 (s, 6H), 3.44 (s, 3H), 3.22 (d, *J* = 15.9 Hz, 1H), 2.97 (d, *J* = 15.9 Hz, 1H), 1.28 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.6, 167.0, 160.7, 146.2, 138.5, 134.5, 130.1, 129.0, 122.9, 119.4, 115.0, 112.9, 107.5, 102.5, 55.5, 42.1, 37.4, 30.4, 23.1; HRMS (ESI) for $C_{22}H_{21}NO_5Na$ $[M+Na]^+$ calcd. 402.1312, found 402.1320.

4a,6-dimethyl-1-phenyl-9-(trifluoromethyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3n)



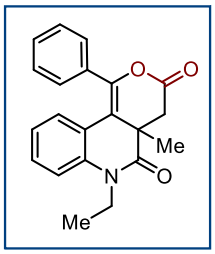
Purification by flash chromatography (petroleum ether/ethyl acetate = 4/1). White solid; mp 115-117°C; 66% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.46−7.40 (m, 4H), 7.37−7.33 (m, 2H), 7.11 (d, *J* = 8.6 Hz, 1H), 7.05 (d, *J* = 1.3 Hz, 1H), 3.47 (s, 3H), 3.25 (d, *J* = 16.0 Hz, 1H), 3.01 (d, *J* = 16.0 Hz, 1H), 1.31 (s, 3H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 170.7, 166.4, 148.2, 141.1, 131.9, 130.5, 129.4, 128.8, 127.2 (q, *J* = 3.8 Hz), 125.5 (q, *J* = 3.7 Hz), 125.0 (q, *J* = 33.0 Hz), 123.4 (q, *J* = 270.2 Hz), 120.0, 115.1, 111.0, 41.9, 37.3, 30.6, 23.3; HRMS (ESI) for $C_{21}H_{16}F_3NO_3Na$ $[M+Na]^+$ calcd. 410.0974, found 410.0979.

9-fluoro-4a,6-dimethyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3o)



Purification by flash chromatography (petroleum ether/ethyl acetate = 4/1). White solid; mp 158-160°C; 55% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.50 (d, *J* = 7.1 Hz, 2H), 7.46−7.36 (m, 3H), 7.02−6.91 (m, 2H), 6.55 (dd, *J* = 9.7, 2.7 Hz, 1H), 3.45 (s, 3H), 3.24 (d, *J* = 16.0 Hz, 1H), 3.00 (d, *J* = 16.0 Hz, 1H), 1.31 (s, 3H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 170.3, 166.6, 158.2 (d, *J* = 241.1 Hz), 147.7, 135.0 (d, *J* = 2.4 Hz), 132.2, 130.4, 129.6, 128.8, 121.5 (d, *J* = 8.3 Hz), 116.4 (d, *J* = 12.4 Hz), 116.3 (d, *J* = 3.9 Hz), 115.6 (d, *J* = 22.8 Hz), 111.7 (d, *J* = 2.1 Hz), 41.9, 37.3, 30.7, 23.1; HRMS (ESI) for $C_{20}H_{16}FNO_3Na$ $[M+Na]^+$ calcd. 360.1006, found 360.1013.

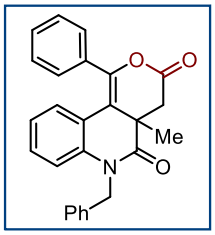
6-ethyl-4a-methyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3p)



Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). White solid; mp 142-144°C; 57% yield; **^{1}H NMR (400 MHz, CDCl$_3$)** δ (ppm) 7.49−7.46 (m, 2H), 7.39−7.29 (m, 3H), 7.20 (td, *J* = 7.8, 1.5 Hz, 1H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.88 (dd, *J* = 7.9, 1.4 Hz, 1H), 6.73 (td, *J* = 7.6, 0.8 Hz, 1H), 4.23−4.13 (m, 1H), 3.96−3.86 (m, 1H), 3.19 (d, *J* = 16.0 Hz, 1H), 3.01 (d, *J* = 16.0 Hz, 1H), 1.32 (t, *J* = 7.1 Hz, 3H), 1.28 (s, 3H); **^{13}C NMR (100 MHz, CDCl$_3$)** δ (ppm) 170.1, 167.2, 146.3, 137.6, 132.9, 130.4, 129.8, 129.7, 128.9, 128.5, 122.7, 119.8, 114.9, 112.5, 42.0, 38.4, 37.3, 23.0,

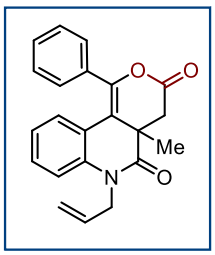
12.5; HRMS (ESI) for $C_{21}H_{19}NO_3Na$ $[M+Na]^+$ calcd. 356.1257, found 356.1267.

6-benzyl-4a-methyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3q)



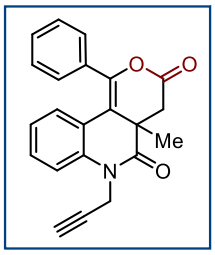
Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). Colorless oil; 53% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.50 (d, *J* = 7.0 Hz, 2H), 7.40−7.28 (m, 6H), 7.22 (d, *J* = 7.4 Hz, 2H), 7.06 (td, *J* = 7.8, 0.9 Hz, 1H), 6.93−6.87 (m, 2H), 6.70 (t, *J* = 7.6 Hz, 1H), 5.54 (d, *J* = 16.2 Hz, 1H), 4.91 (d, *J* = 16.2 Hz, 1H), 3.25 (d, *J* = 16.0 Hz, 1H), 3.09 (d, *J* = 16.0 Hz, 1H), 1.42 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.9, 167.0, 146.6, 138.0, 136.4, 132.8, 130.1, 129.9, 129.7, 128.9, 128.9, 128.6, 127.3, 126.0, 123.0, 119.8, 116.0, 112.2, 47.0, 42.3, 37.4, 23.3; HRMS (ESI) for $C_{26}H_{21}NO_3Na$ $[M+Na]^+$ calcd. 418.1414, found 418.1422.

6-allyl-4a-methyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3r)



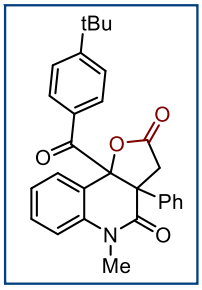
Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). Colorless oil; 48% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.51−7.48 (m, 2H), 7.40−7.30 (m, 3H), 7.17 (td, *J* = 7.8, 1.4 Hz, 1H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.88 (dd, *J* = 7.8, 1.2 Hz, 1H), 6.73 (td, *J* = 7.6, 0.8 Hz, 1H), 5.99−5.89 (m, 1H), 5.27−5.23 (m, 1H), 5.16 (d, *J* = 17.8 Hz, 1H), 4.93−4.87 (m, 1H), 4.35−4.29 (m, 1H), 3.21 (d, *J* = 16.0 Hz, 1H), 3.02 (d, *J* = 16.0 Hz, 1H), 1.33 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.4, 167.1, 146.5, 137.9, 132.8, 131.8, 130.1, 129.9, 129.7, 128.8, 128.6, 122.9, 119.7, 116.5, 115.8, 112.3, 45.8, 42.2, 37.3, 23.2; HRMS (ESI) for $C_{22}H_{19}NO_3Na$ $[M+Na]^+$ calcd. 368.1257, found 368.1270.

4a-methyl-1-phenyl-6-(prop-2-yn-1-yl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3s)



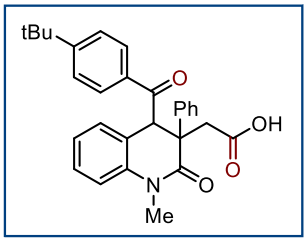
Purification by flash chromatography (petroleum ether/ethyl acetate = 6/1). White solid; mp 156-158℃; 62% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.50−7.47 (m, 2H), 7.39−7.30 (m, 3H), 7.25 (d, *J* = 3.8 Hz, 2H), 6.89 (d, *J* = 7.8 Hz, 2H), 6.80−6.75 (m, 1H), 5.00−4.94 (m, 1H), 4.58−4.52 (m, 1H), 3.22 (d, *J* = 16.0 Hz, 1H), 3.01 (d, *J* = 16.0 Hz, 1H), 2.29 (t, *J* = 2.4 Hz, 1H), 1.31 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 170.1, 166.8, 146.8, 137.0, 132.7, 130.2, 129.9, 129.6, 128.9, 128.6, 127.9, 123.3, 119.8, 115.5, 112.0, 78.0, 72.4, 42.2, 37.2, 32.8, 22.9; HRMS (ESI) for $C_{22}H_{17}NO_3Na$ $[M+Na]^+$ calcd. 366.1101, found 366.1111.

9b-(4-(tert-butyl)benzoyl)-5-methyl-3a-phenyl-3,3a,5,9b-tetrahydrofuro[3,2-c]quinoline-2,4-dione (3d-I)



Purification by flash chromatography (petroleum ether/ethyl acetate = 3/1). Colorless oil; 58% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.37 (dd, *J* = 8.8, 2.3 Hz, 1H), 7.27 (d, *J* = 8.4 Hz, 2H), 7.19−7.06 (m, 10H), 3.51 (s, 3H), 3.25 (d, *J* = 17.4 Hz, 1H), 3.19 (d, *J* = 17.4 Hz, 1H), 1.18 (s, 9H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 196.3, 172.3, 167.4, 157.1, 137.1, 134.7, 132.4, 131.1, 129.3, 129.2, 128.9, 128.7, 128.6, 128.4, 125.0, 116.6, 92.0, 57.6, 38.8, 35.0, 30.8, 30.7; HRMS (ESI) for $C_{29}H_{27}NO_4Na$ $[M+Na]^+$ calcd. 476.1832, found 476.1839.

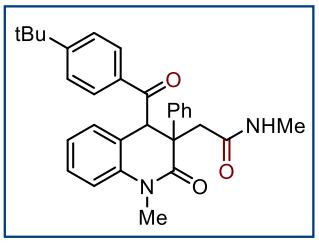
2-(4-(4-(tert-butyl)benzoyl)-1-methyl-2-oxo-3-phenyl-1,2,3,4-tetrahydroquinolin-3-yl)acetic acid (3d-II)



Purification by flash chromatography (petroleum ether/ethyl acetate = 1/1). White solid; mp 208-210℃; 61% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 8.04 (d, *J* = 8.4 Hz, 2H), 7.53 (d, *J* = 8.4 Hz, 2H), 7.27−7.19 (m, 4H), 7.17−7.08 (m, 3H), 6.94 (d, *J* = 8.1 Hz, 1H), 6.83 (t, *J* = 7.5 Hz, 1H), 5.40 (s, 1H), 3.58 (s, 3H), 3.54 (d, *J* = 14.8 Hz, 1H), 2.86 (d, *J* = 14.8 Hz, 1H), 1.35 (s, 9H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 196.5, 174.2, 171.4, 158.1, 139.1, 136.2, 132.8,

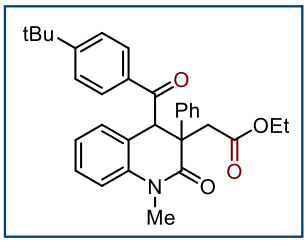
129.3, 128.9, 128.8, 128.1, 126.1, 126.0, 123.9, 122.2, 115.8, 54.4, 50.4, 44.1, 35.2, 31.0, 30.8; HRMS (ESI) for $C_{29}H_{29}NO_4Na$ $[M+H]^+$ calcd. 478.1989, found 478.2004.

2-(4-(4-(tert-butyl)benzoyl)-1-methyl-2-oxo-3-phenyl-1,2,3,4-tetrahydroquinolin-3-yl)-N-methylacetamide (3d-III)



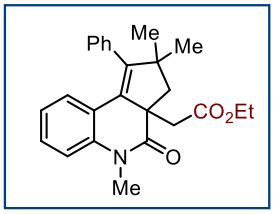
Purification by flash chromatography (petroleum ether/ethyl acetate = 3/2). White solid; mp 168-170°C; 54% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.78 (d, *J* = 8.4 Hz, 2H), 7.49 (d, *J* = 7.7 Hz, 2H), 7.38 (d, *J* = 8.4 Hz, 2H), 7.27−7.22 (m, 1H), 7.20−7.12 (m, 3H), 7.09 (t, *J* = 7.3 Hz, 1H), 7.00 (d, *J* = 8.1 Hz, 1H), 6.94 (t, *J* = 7.4 Hz, 1H), 6.18 (q, *J* = 4.2 Hz, 1H), 5.51 (s, 1H), 3.51 (s, 3H), 3.11 (d, *J* = 16.0 Hz, 1H), 2.89 (d, *J* = 16.0 Hz, 1H), 2.49 (d, *J* = 4.7 Hz, 3H), 1.30 (s, 9H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 196.9, 171.8, 170.0, 157.0, 139.5, 137.8, 133.6, 129.0, 128.6, 128.6, 128.5, 127.8, 126.9, 125.6, 123.3, 123.1, 115.2, 53.7, 49.6, 43.2, 35.0, 31.0, 30.7, 26.2; HRMS (ESI) for $C_{30}H_{32}N_2O_3Na$ $[M+Na]^+$ calcd. 491.2305, found 491.2316.

ethyl 2-(4-(4-(tert-butyl)benzoyl)-1-methyl-2-oxo-3-phenyl-1,2,3,4-tetrahydroquinolin-3-yl)acetate (3d-IV)



Purification by flash chromatography (petroleum ether/ethyl acetate = 3/1). Colorless oil; 58% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 8.15 (d, *J* = 8.4 Hz, 2H), 7.53 (d, *J* = 8.5 Hz, 2H), 7.38 (d, *J* = 7.7 Hz, 2H), 7.25 (d, *J* = 6.8 Hz, 2H), 7.21−7.16 (m, 2H), 7.12 (d, *J* = 7.2 Hz, 1H), 7.07−7.02 (m, 1H), 6.83 (d, *J* = 7.4 Hz, 1H), 6.74 (d, *J* = 8.1 Hz, 1H), 6.19 (s, 1H), 3.88−3.74 (m, 2H), 3.47 (d, *J* = 15.7 Hz, 1H), 3.40 (s, 3H), 2.95 (d, *J* = 15.7 Hz, 1H), 1.35 (s, 9H), 0.89 (t, *J* = 7.1 Hz, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 197.2, 171.7, 170.7, 157.2, 139.8, 137.5, 133.4, 129.6, 129.2, 128.3, 128.2, 127.2, 126.9, 125.7, 122.7, 122.2, 115.3, 60.1, 48.9, 48.7, 41.0, 35.1, 31.0, 30.5, 13.8; HRMS (ESI) for $C_{31}H_{33}NO_4Na$ $[M+Na]^+$ calcd. 506.2302, found 506.2309.

ethyl 2-(2,2,5-trimethyl-4-oxo-1-phenyl-2,3,4,5-tetrahydro-3aH-cyclopenta[c]quinolin-3a-yl)acetate (3y')



Purification by flash chromatography (petroleum ether/ethyl acetate = 8/1). Colorless oil; 58% yield; **^{1}H NMR (400 MHz, $CDCl_3$)** δ (ppm) 7.37−7.29 (m, 3H), 7.19−7.14 (m, 1H), 7.11−7.09 (m, 2H), 7.01 (d, J = 8.2 Hz, 1H), 6.71 (d, J = 4.0 Hz, 2H), 4.03−3.90 (m, 2H), 3.42 (s, 3H), 2.64−2.40 (m, 4H), 1.38 (s, 3H), 1.19 (t, J = 7.1 Hz, 3H), 0.99 (s, 3H); **^{13}C NMR (100 MHz, $CDCl_3$)** δ (ppm) 172.7, 170.2, 150.2, 139.6, 136.2, 130.8, 129.0, 128.3, 128.1, 127.5, 127.2, 122.3, 121.4, 114.7, 60.6, 54.3, 48.0, 46.1, 43.3, 30.1, 29.4, 29.0, 14.1; HRMS (ESI) for $C_{25}H_{28}NO_3$ $[M+H]^+$ calcd. 390.2064, found 390.2072.

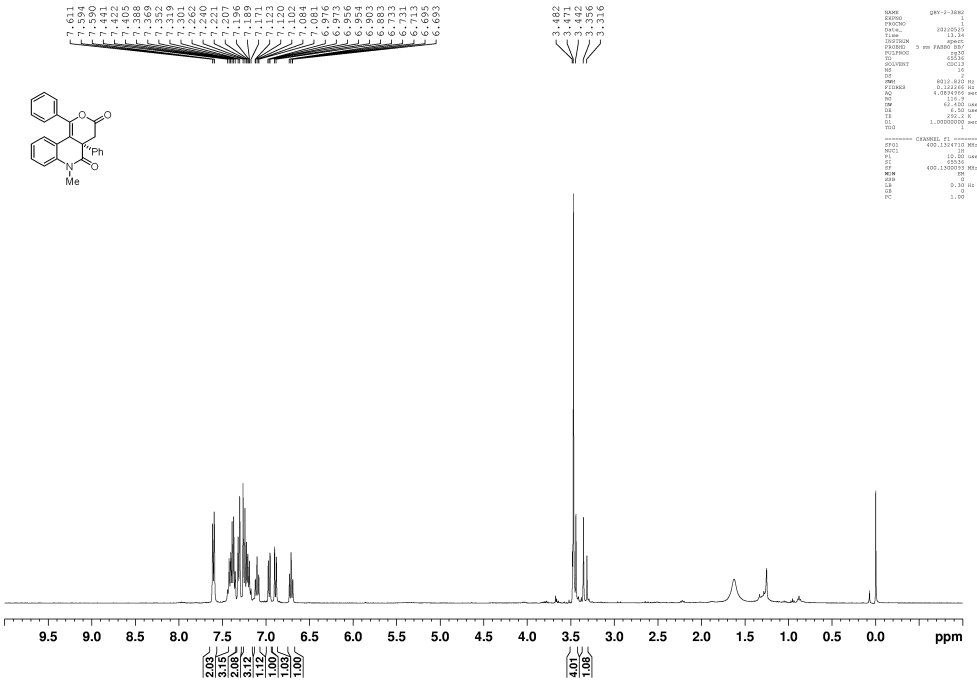
9. References

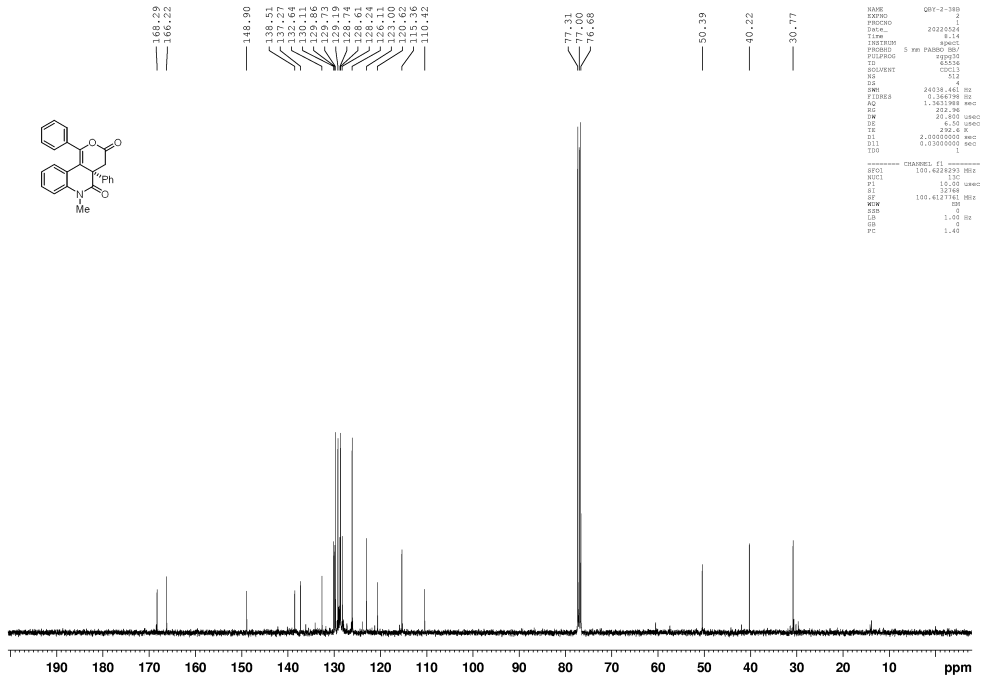
1 B. A. L. Sacchelli, B. C. Rocha, L. H. Andrade, Cascade reactions assisted by microwave irradiation: ultrafast construction of 2-quinolinone-fused γ-lactones from *N*-(*o*-ethynylaryl)acrylamides and formamide, *Org. Lett.*, 2021, **23**, 5071–5075.

2 Z. Cao, D. M. Bassani and B. Bibal, Photoreduction of thioether gold(III) complexes: mechanistic insight and homogeneous catalysis, *Chem. Eur. J.* 2018, **24**, 18779–18787.

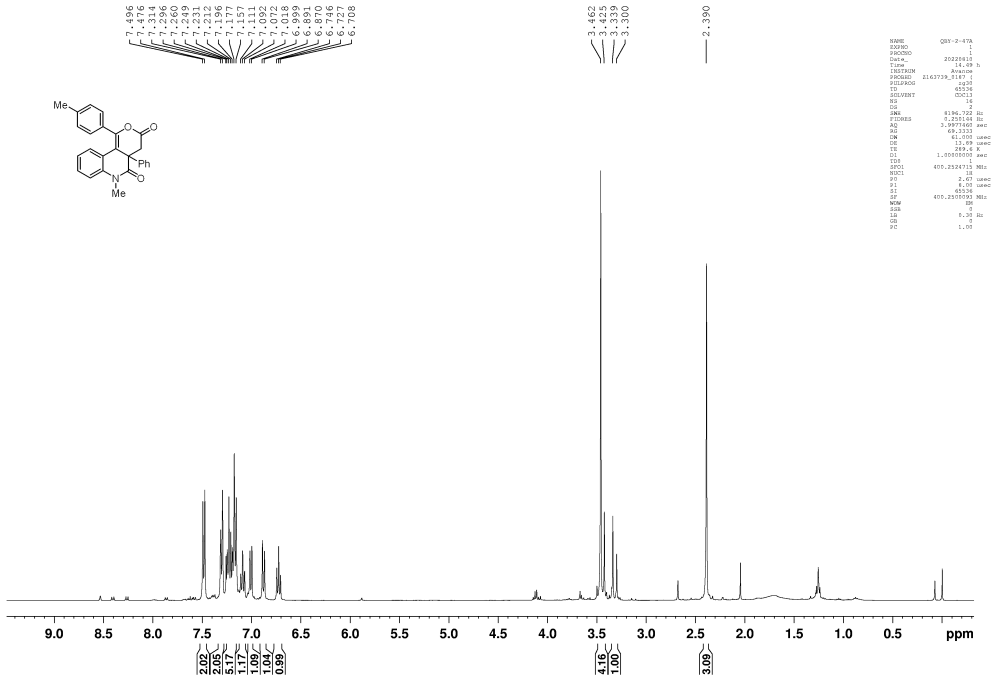
10. NMR spectra of compounds

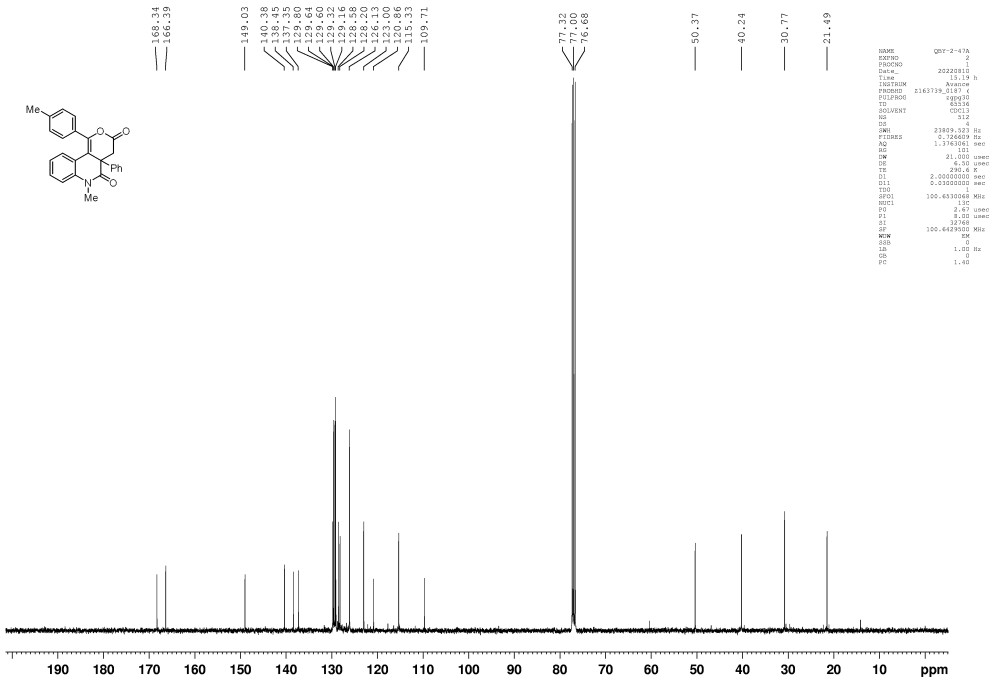
6-methyl-1,4a-diphenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3a)



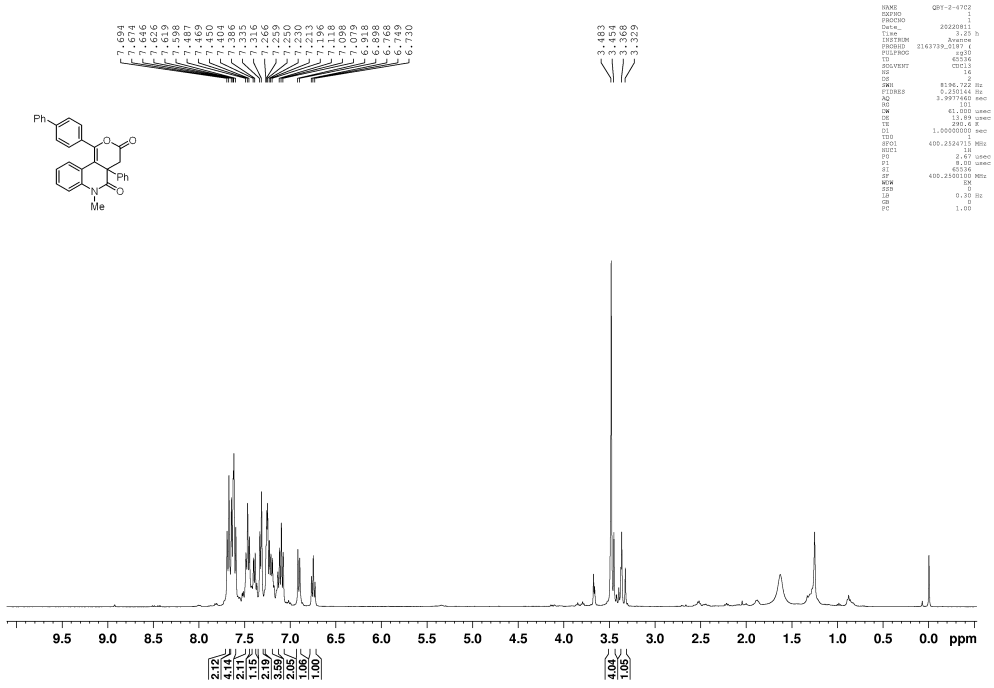


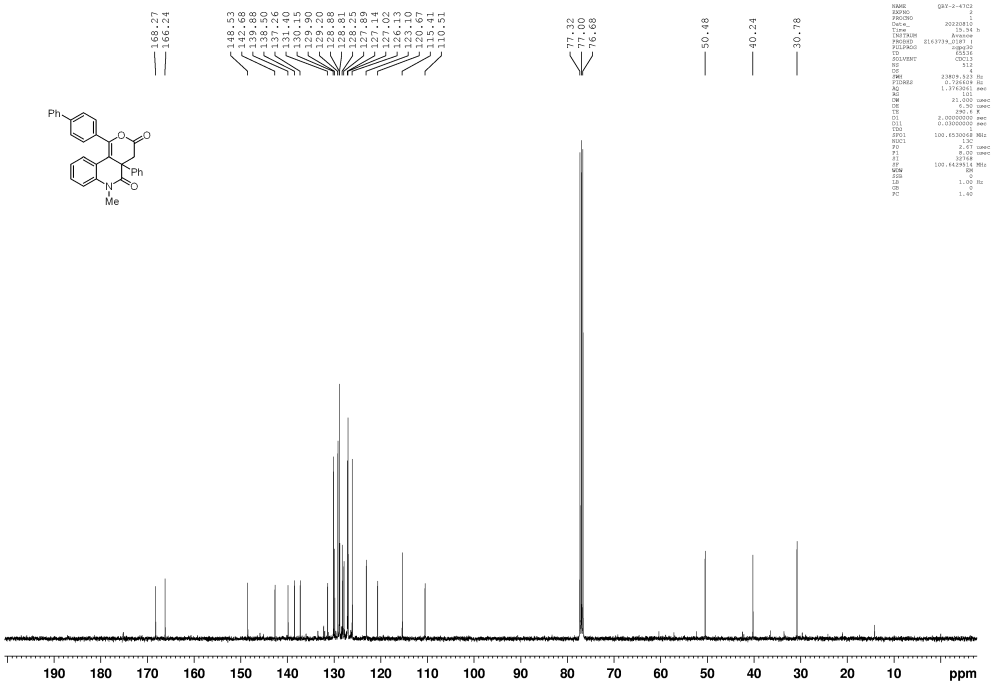
6-methyl-4a-phenyl-1-(p-tolyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3b)



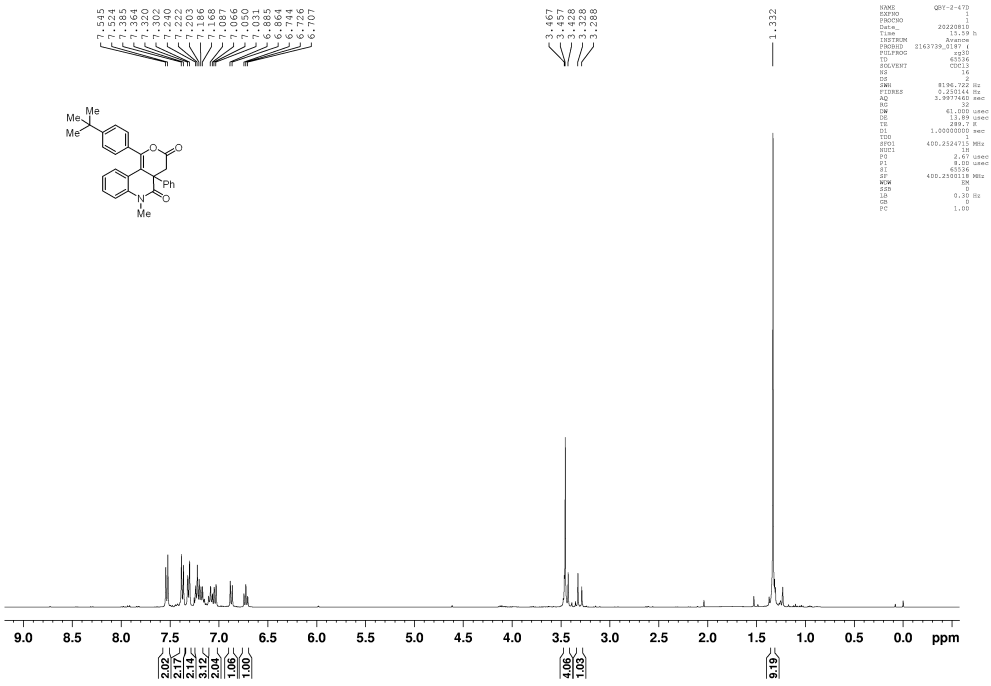


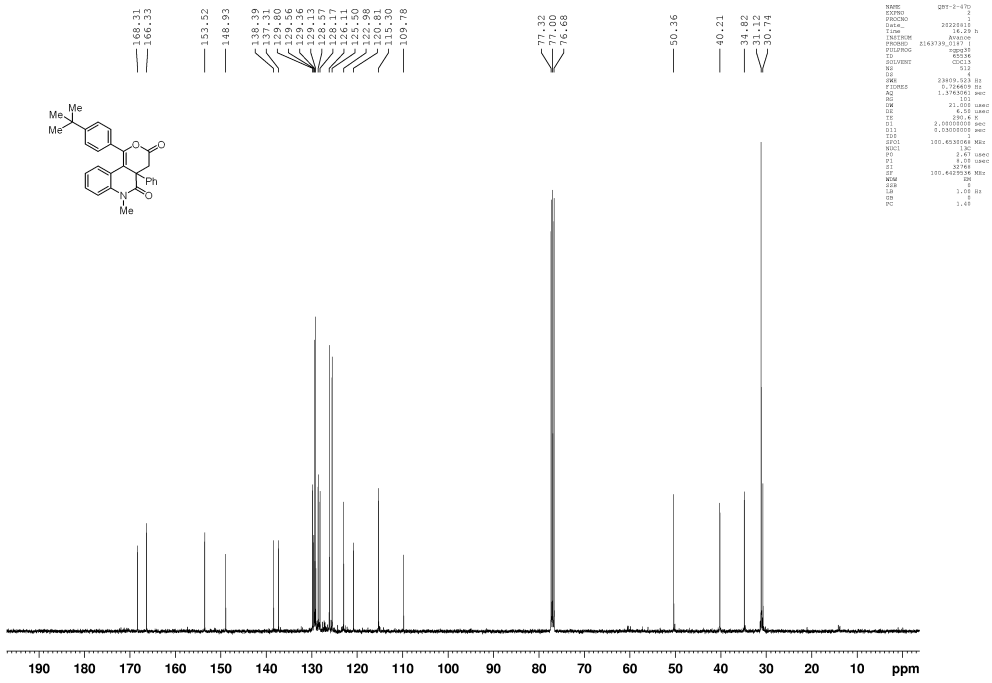
1-([1,1'-biphenyl]-4-yl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3c)



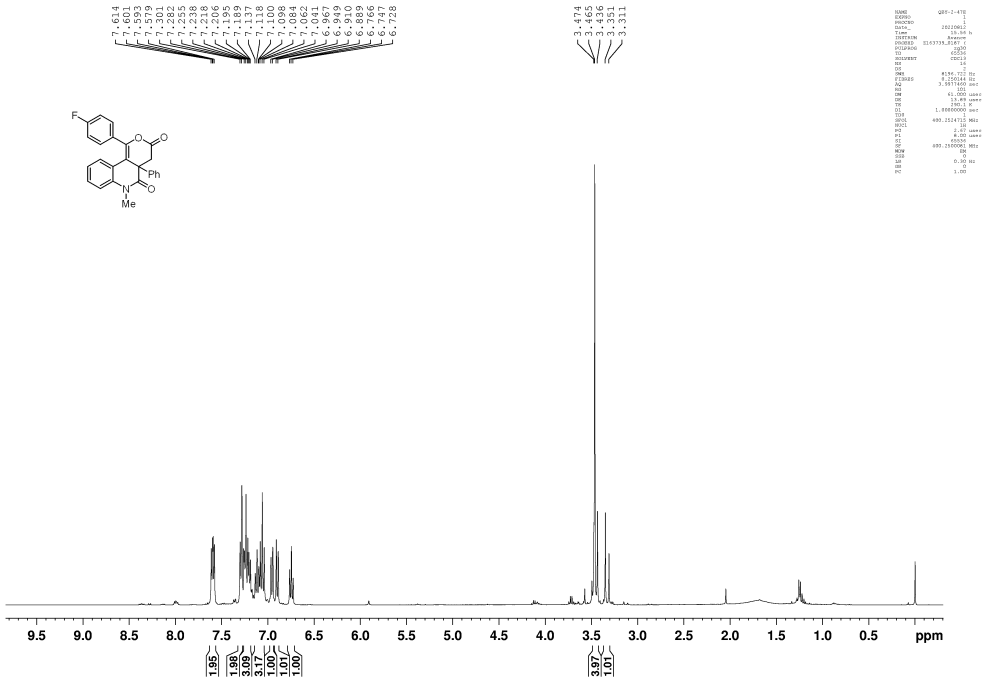


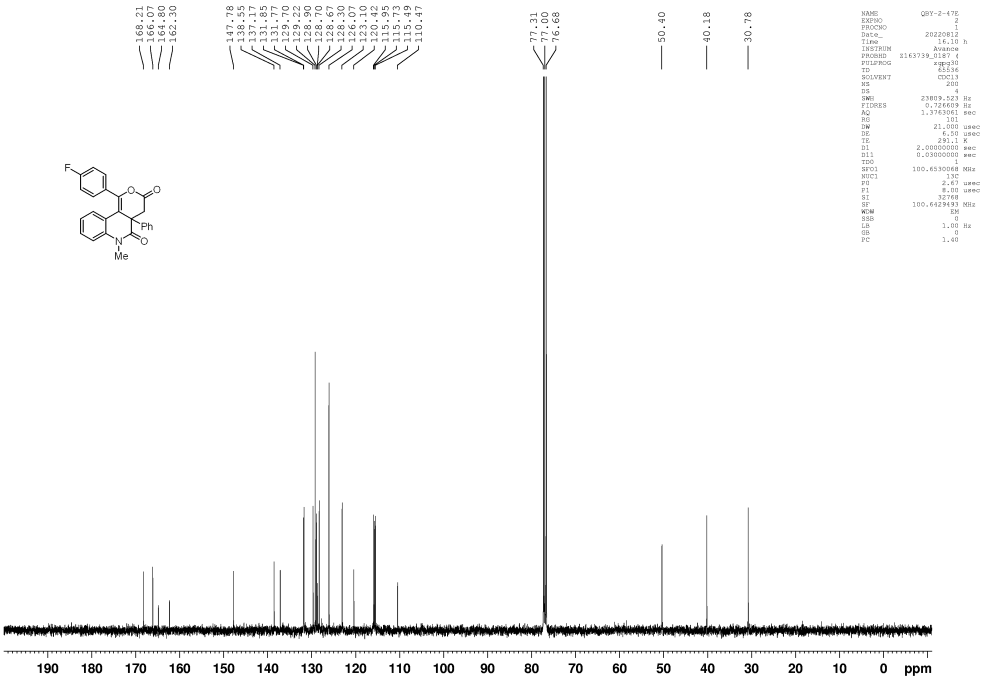
1-(4-(tert-butyl)phenyl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3d)



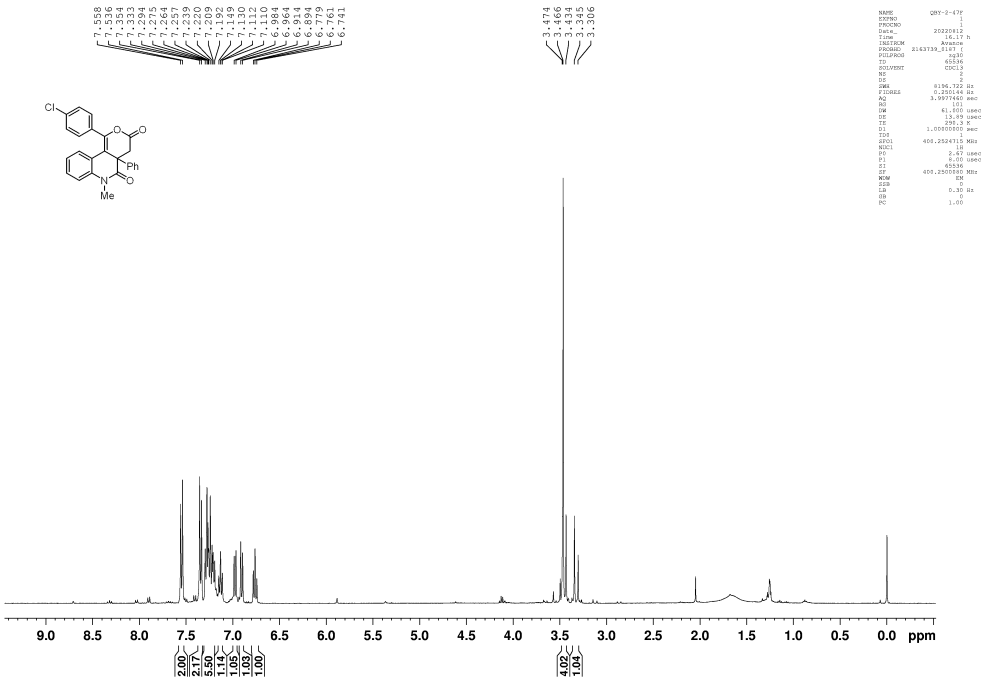


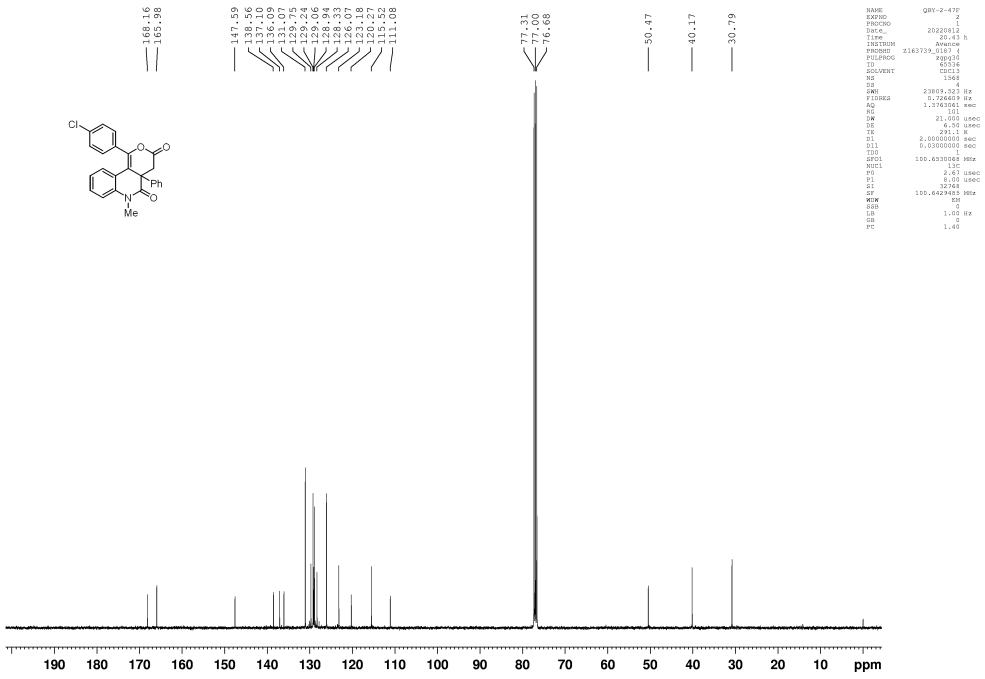
1-(4-fluorophenyl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3e)



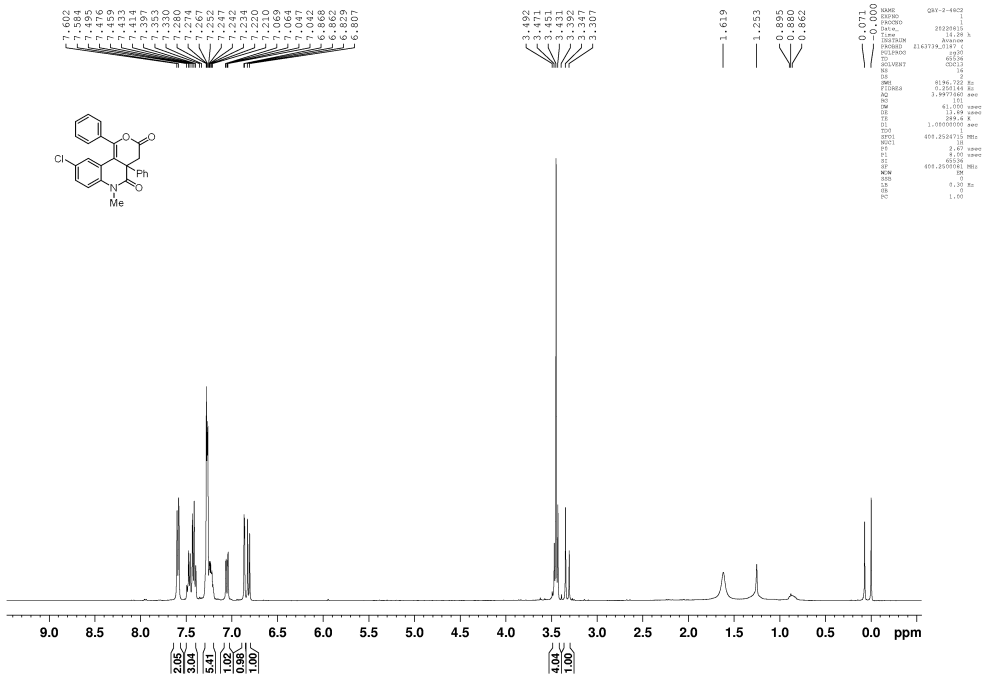


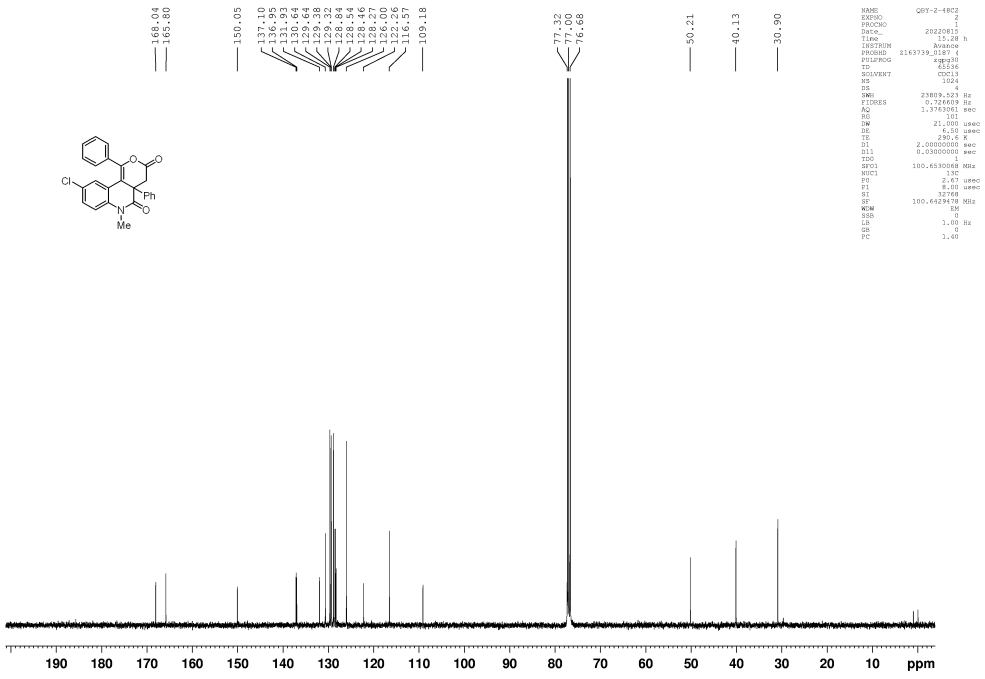
1-(4-chlorophenyl)-6-methyl-4a-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3f)



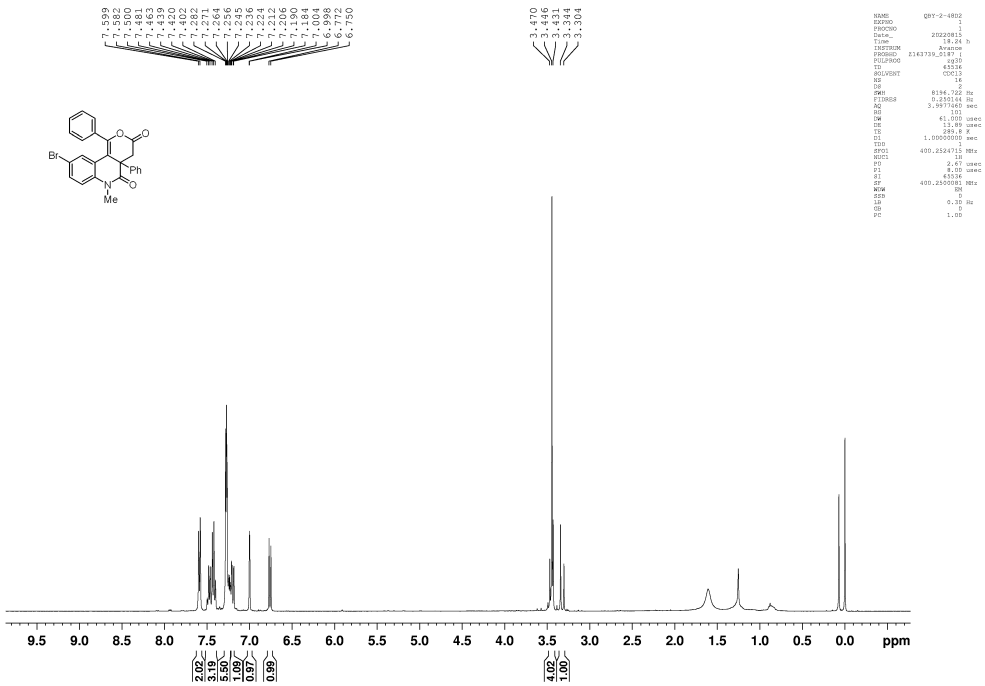


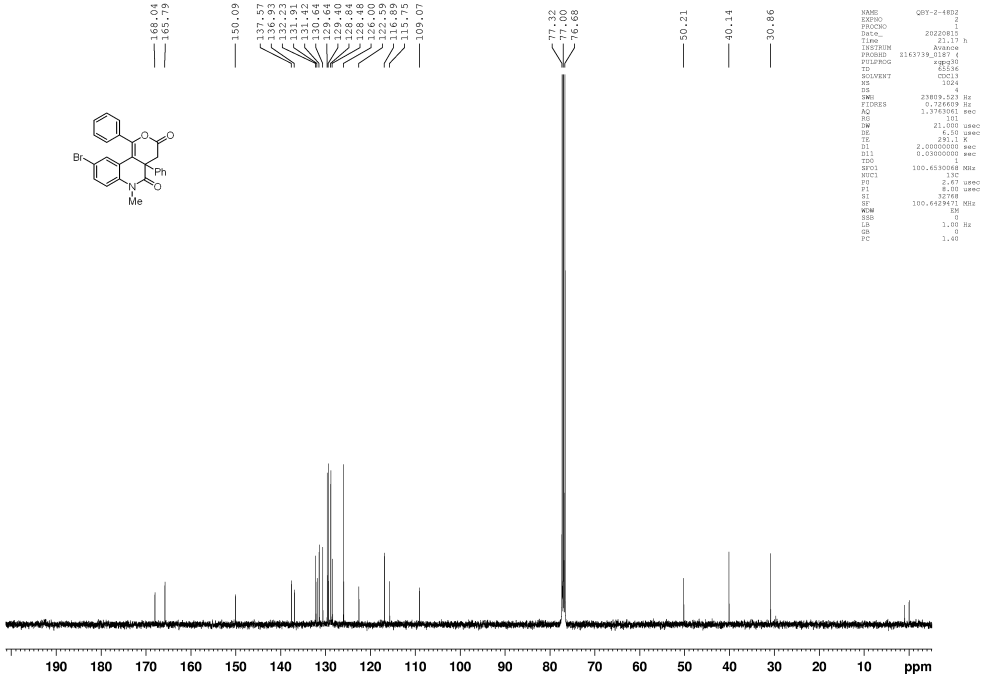
9-chloro-6-methyl-1,4a-diphenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3g)



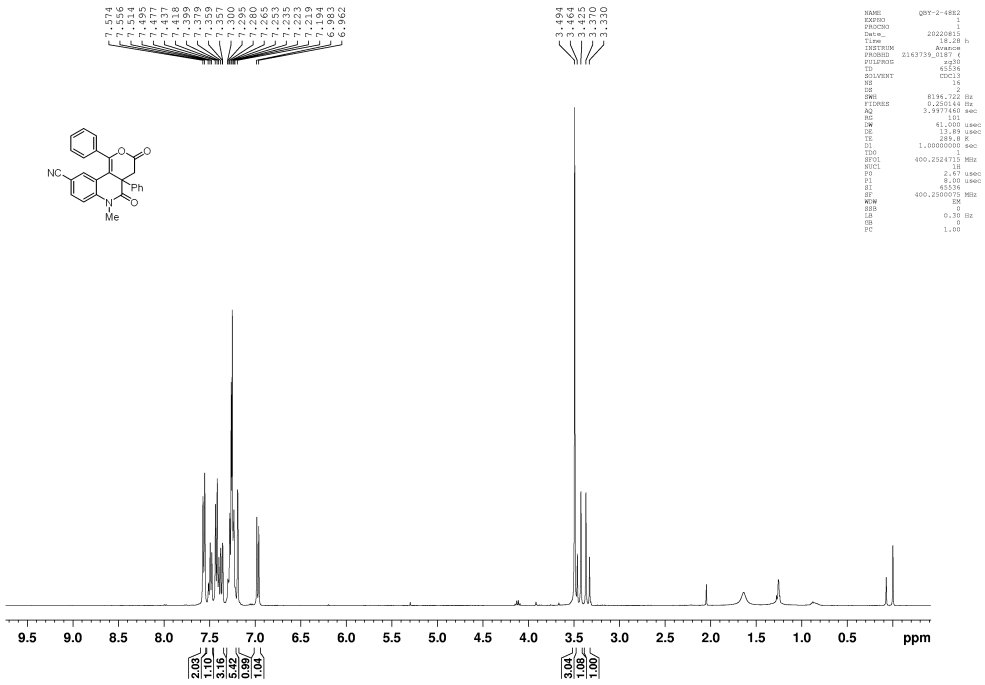


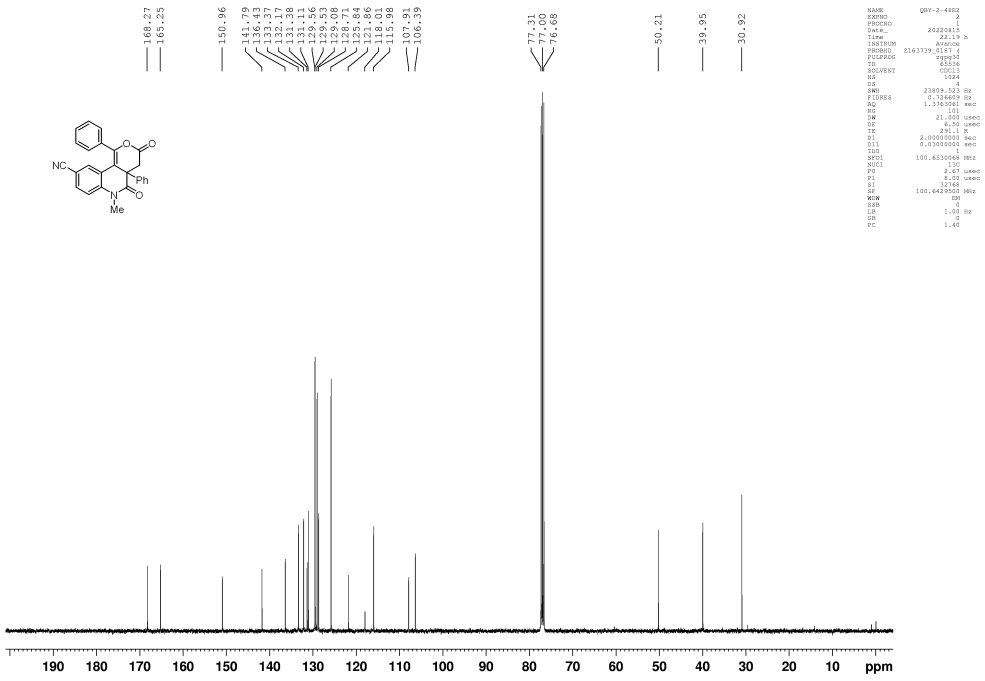
9-bromo-6-methyl-1,4a-diphenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3h)



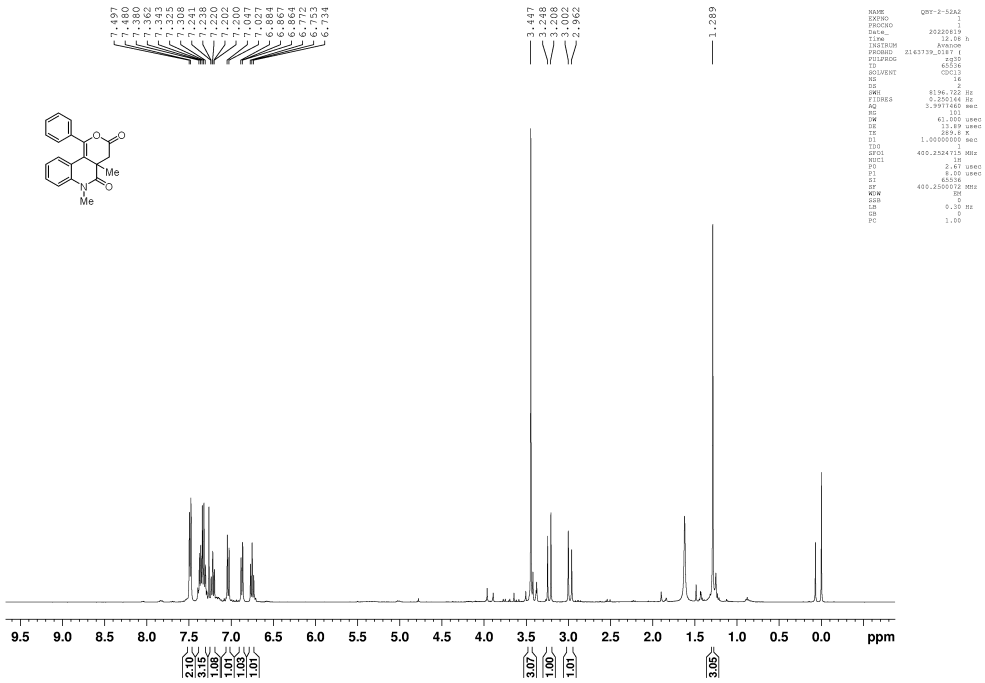


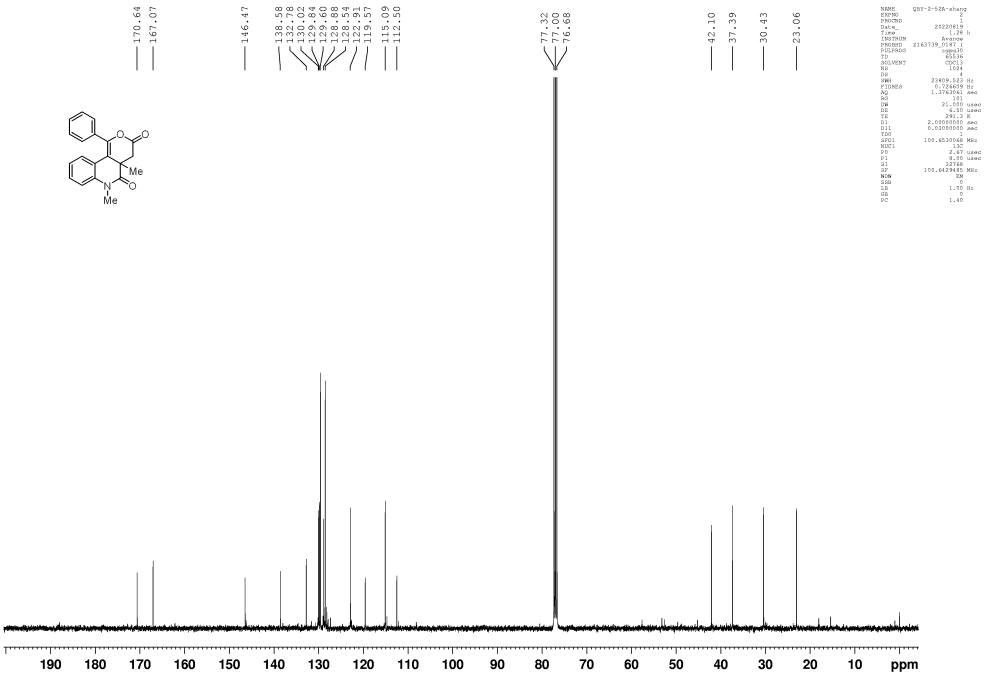
6-methyl-3,5-dioxo-1,4a-diphenyl-4,4a,5,6-tetrahydro-3H-pyrano[4,3-c]quinoline-9-carbonitrile (3i)



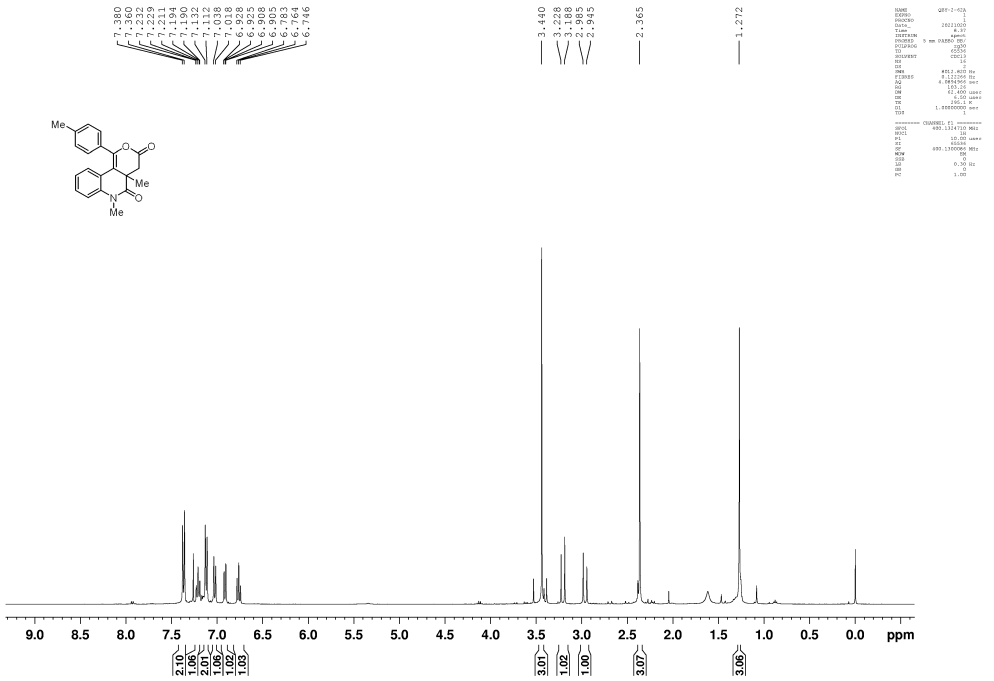


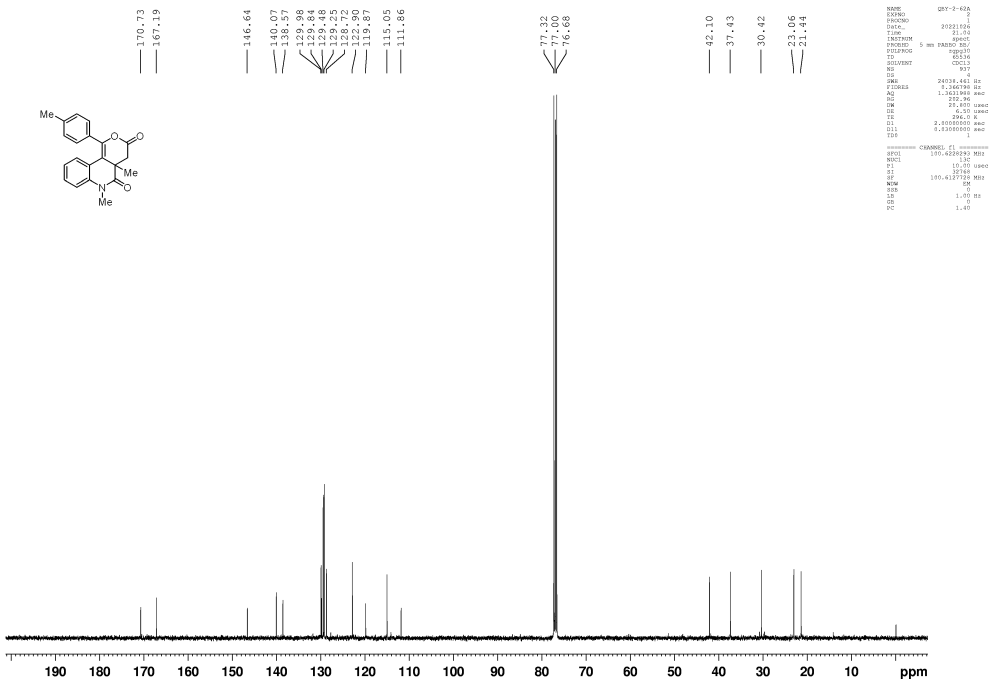
4a,6-dimethyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3j)



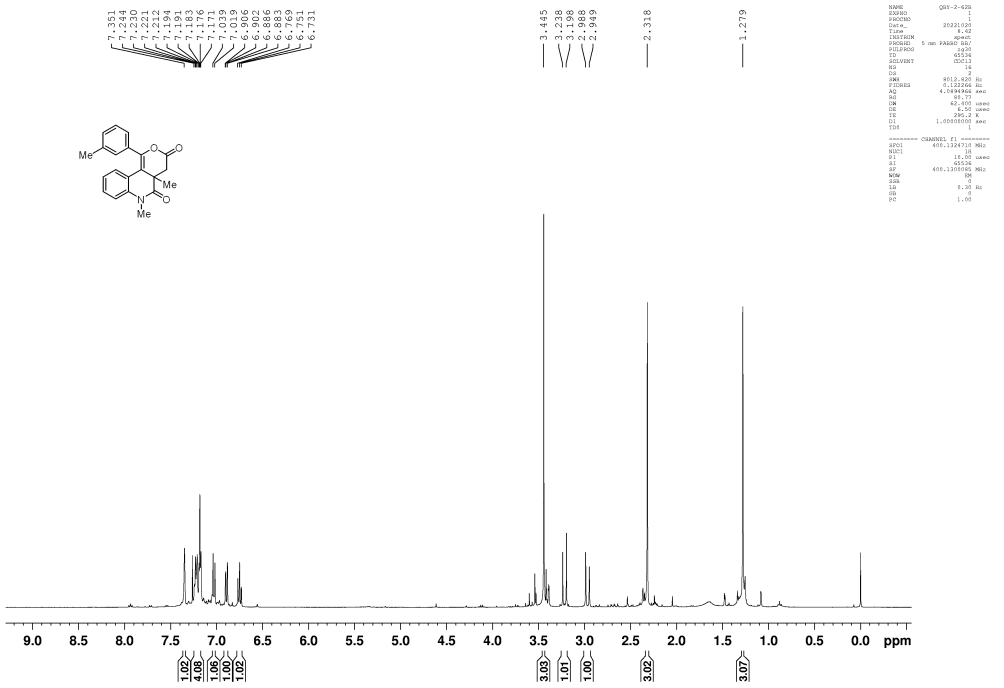


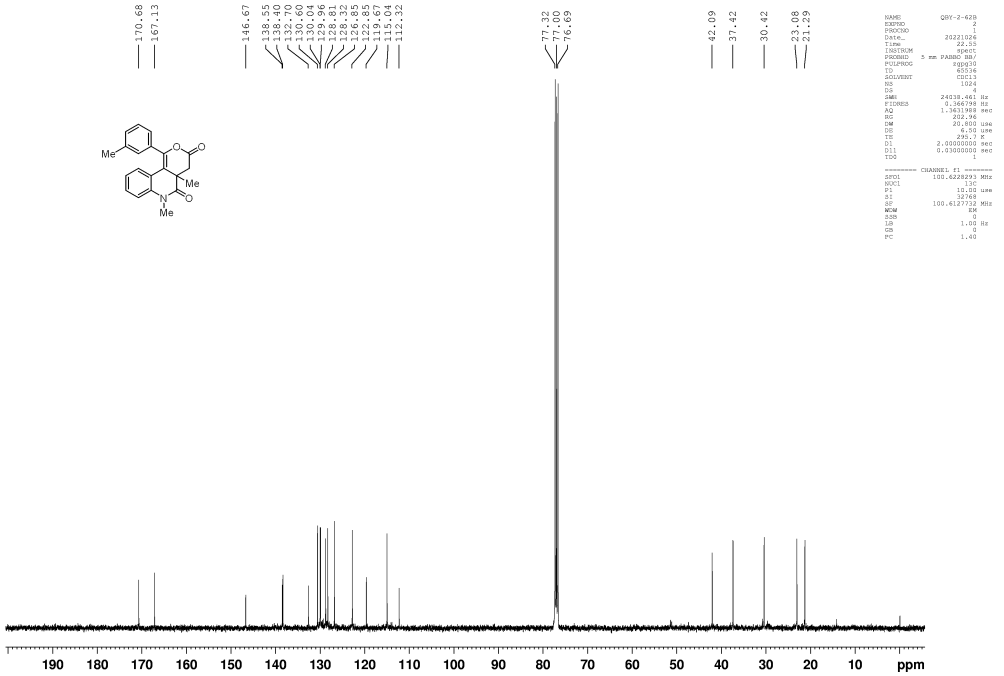
4a,6-dimethyl-1-(p-tolyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3k)



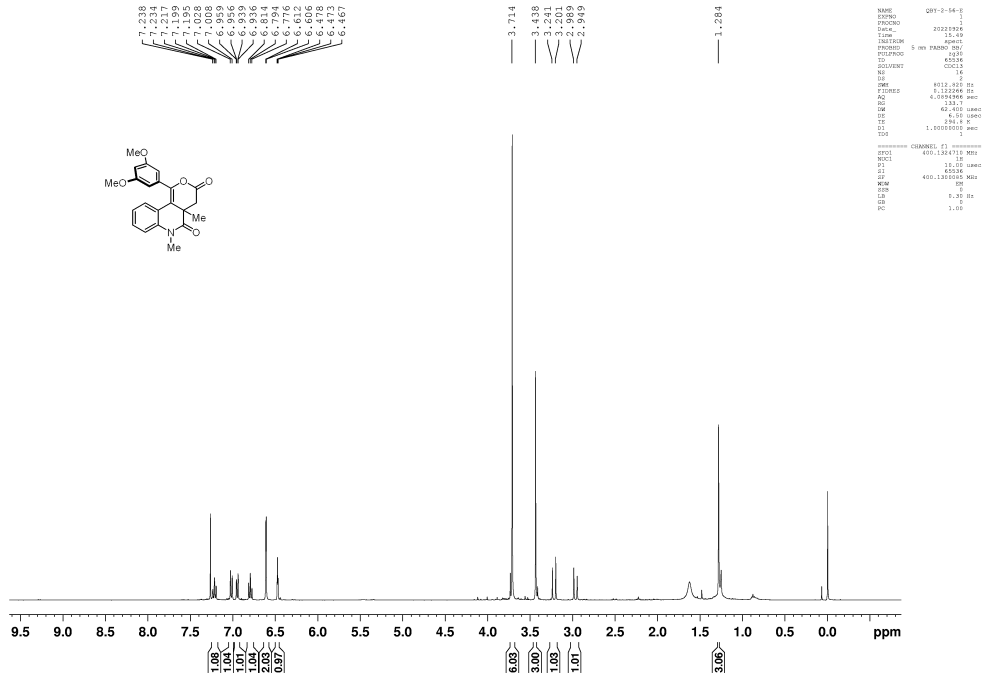


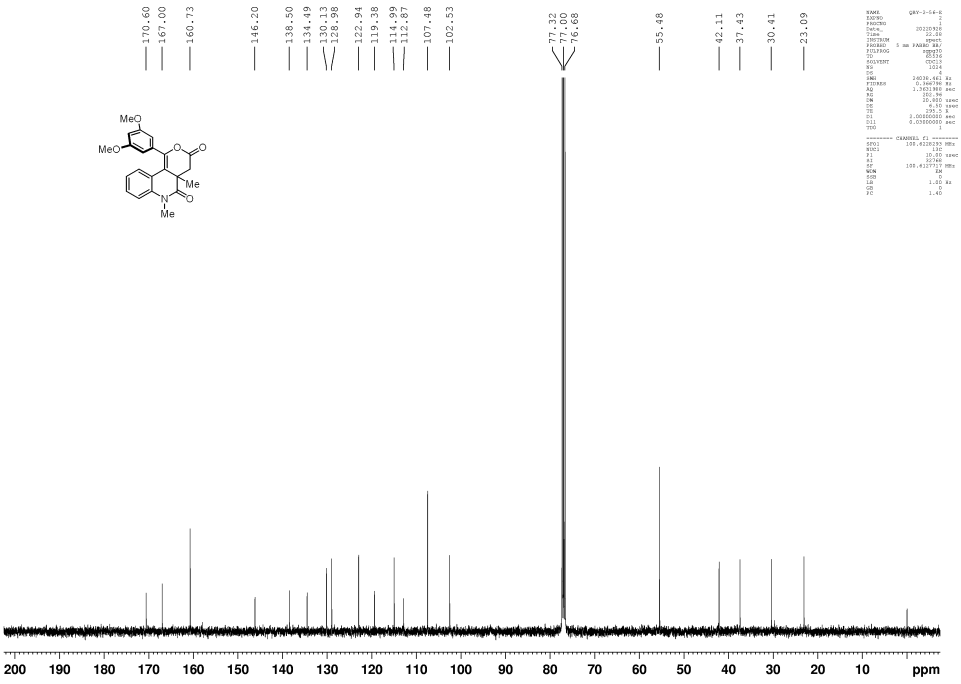
4a,6-dimethyl-1-(m-tolyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3l)



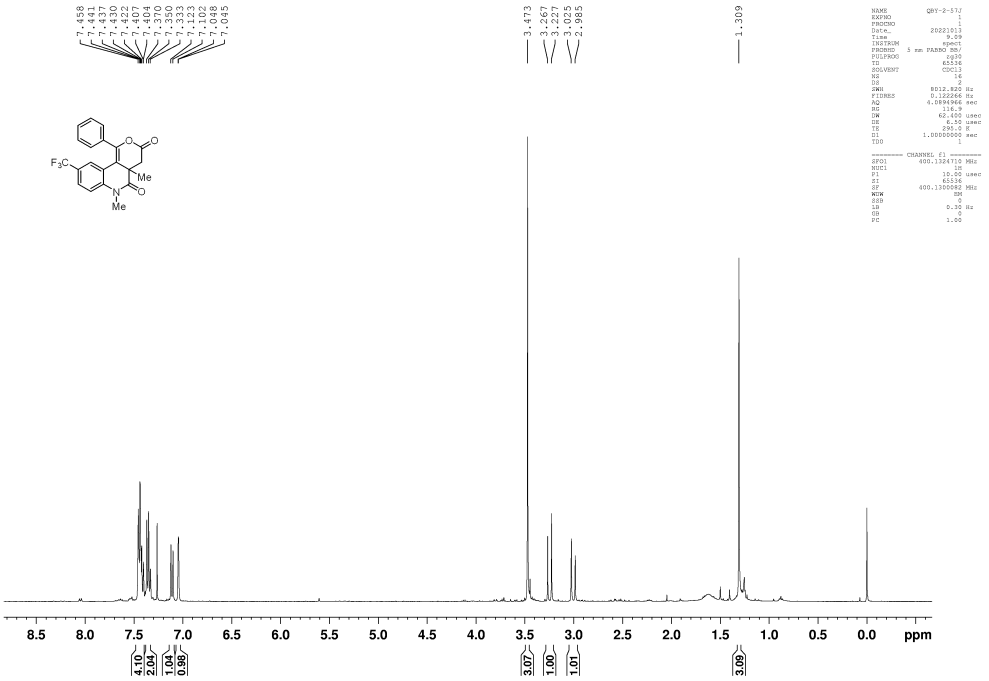


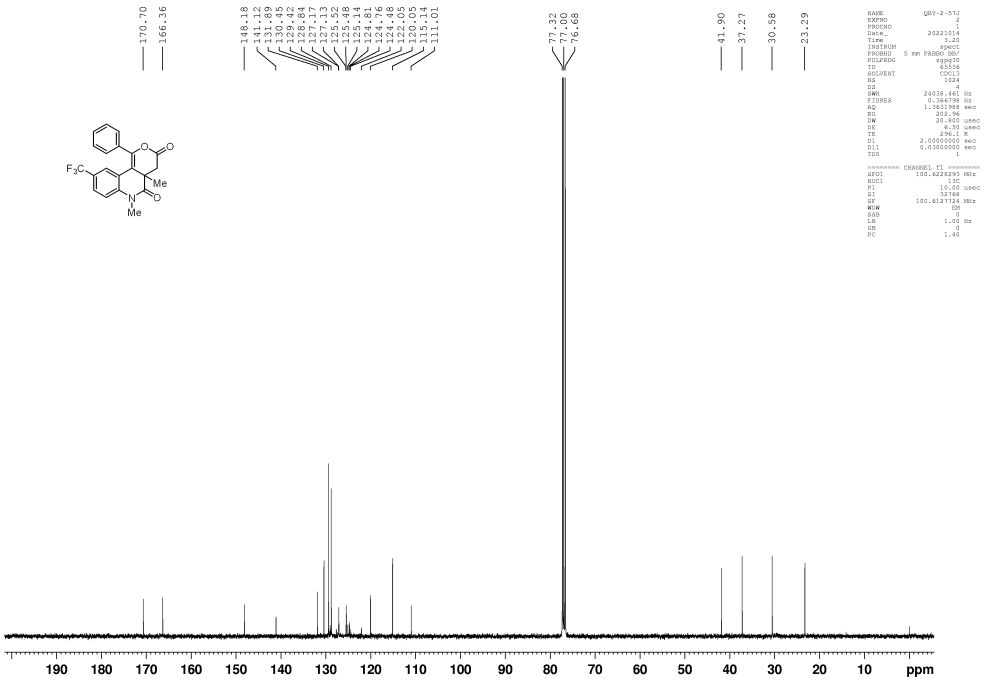
1-(3,5-dimethoxyphenyl)-4a,6-dimethyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3m)



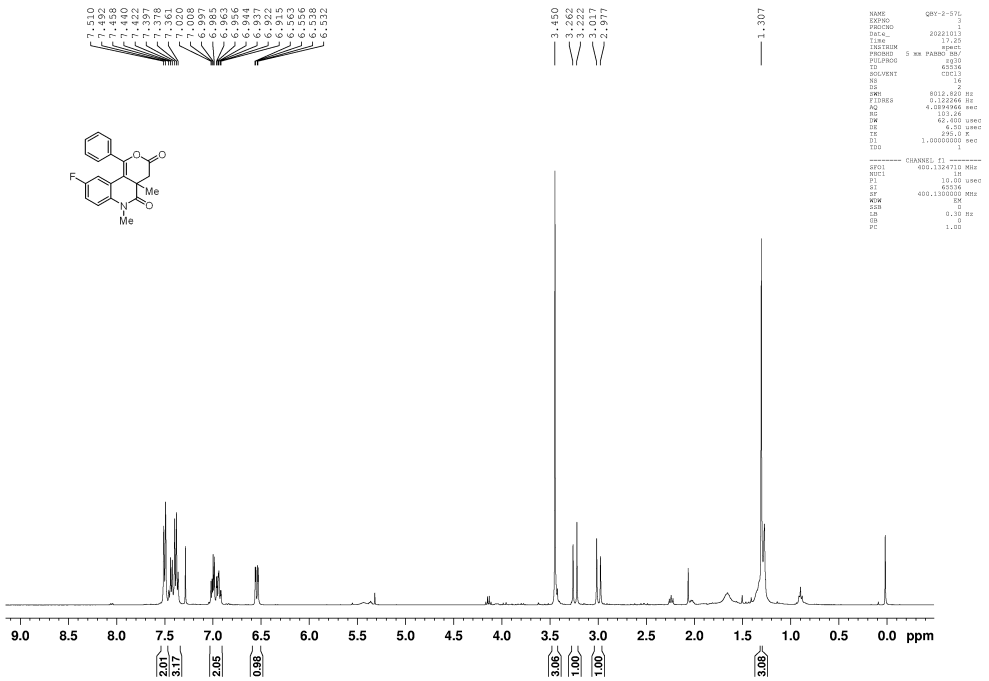


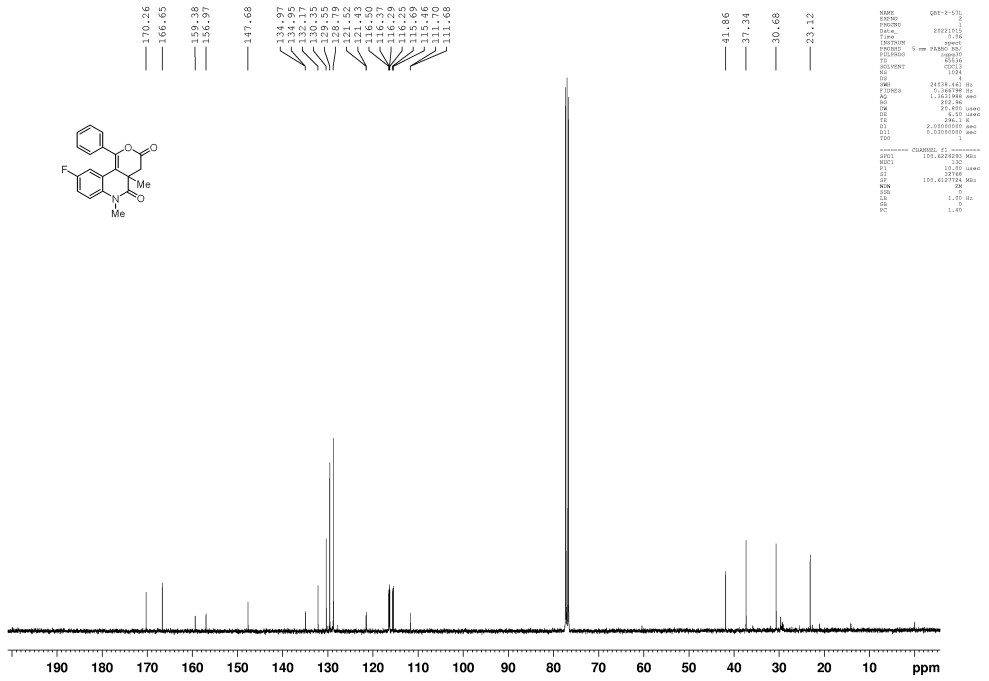
4a,6-dimethyl-1-phenyl-9-(trifluoromethyl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3n)



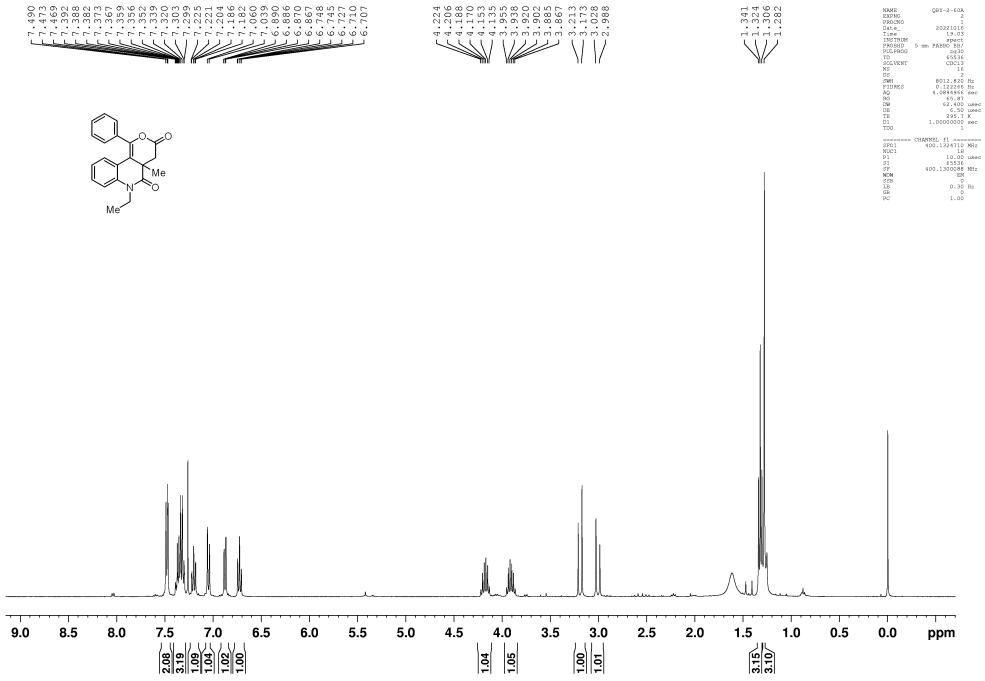


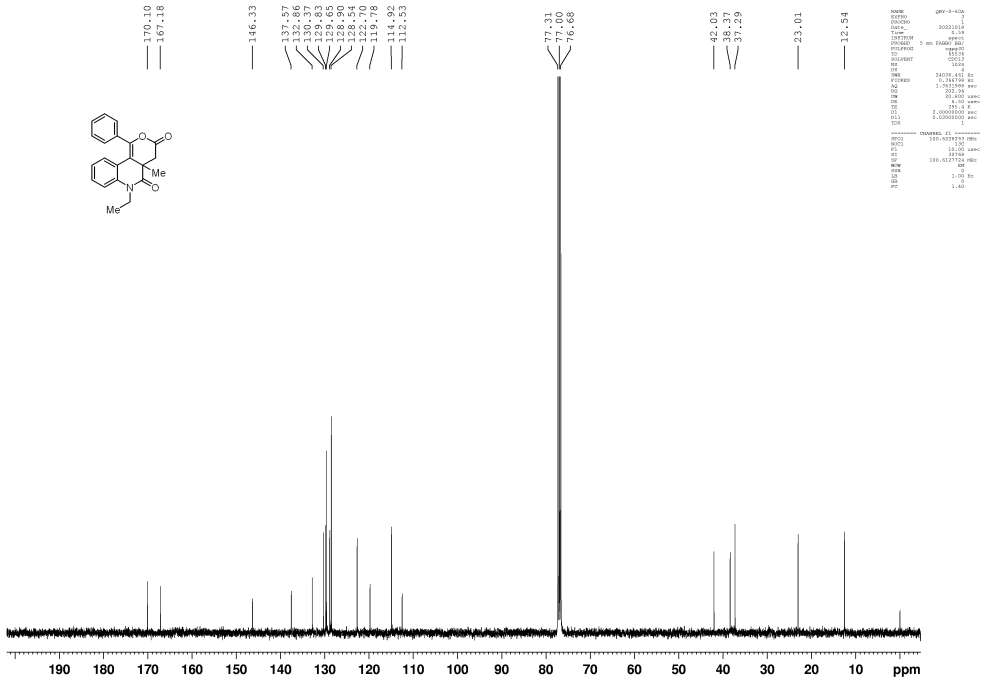
9-fluoro-4a,6-dimethyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3o)



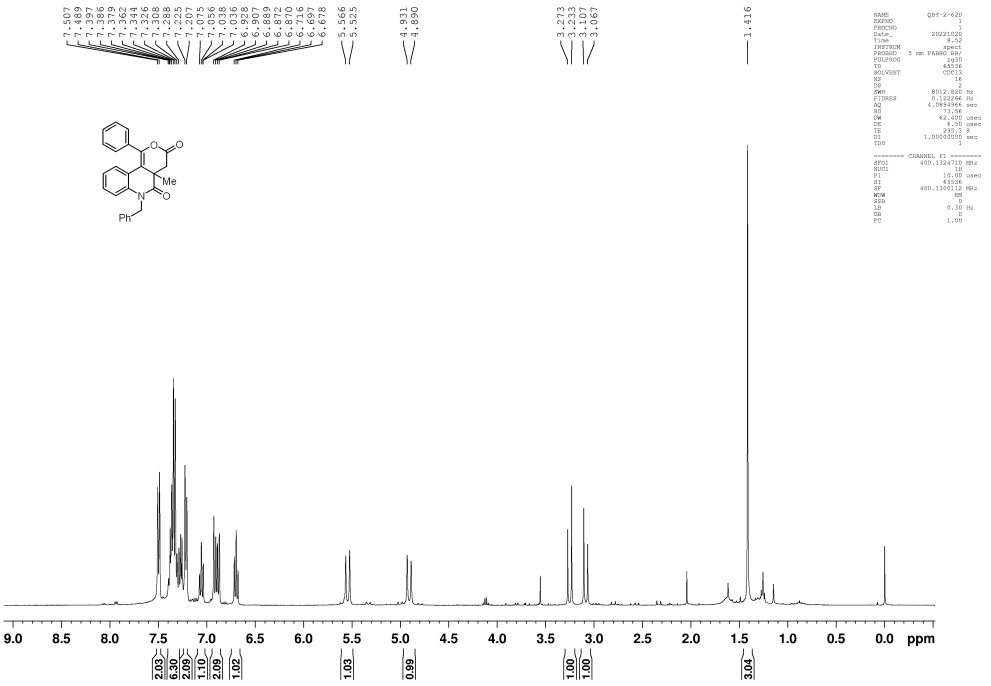


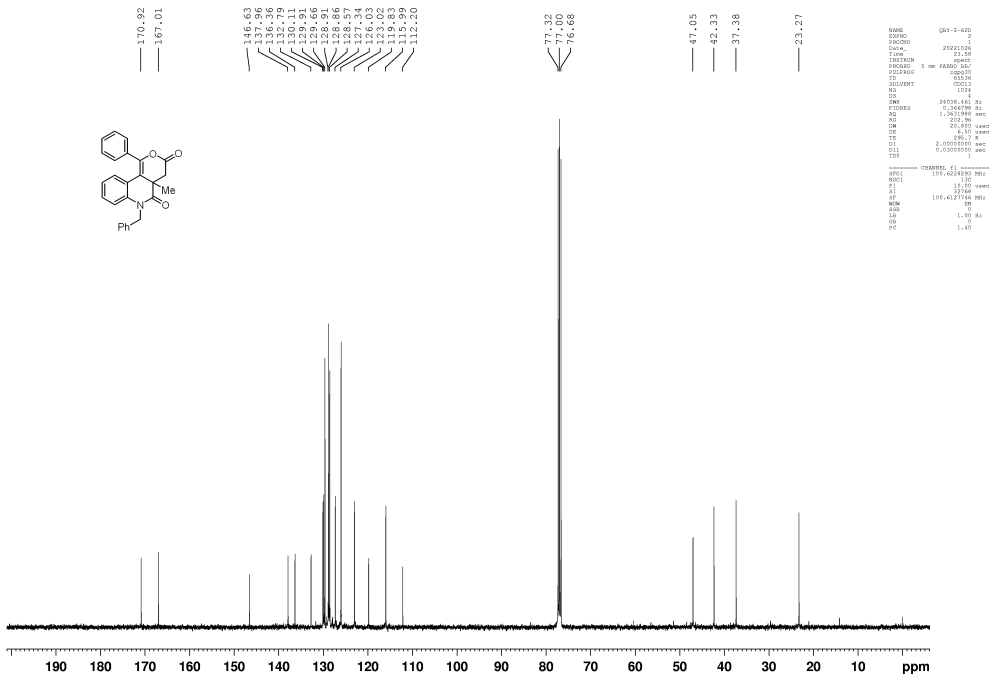
6-ethyl-4a-methyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3p)



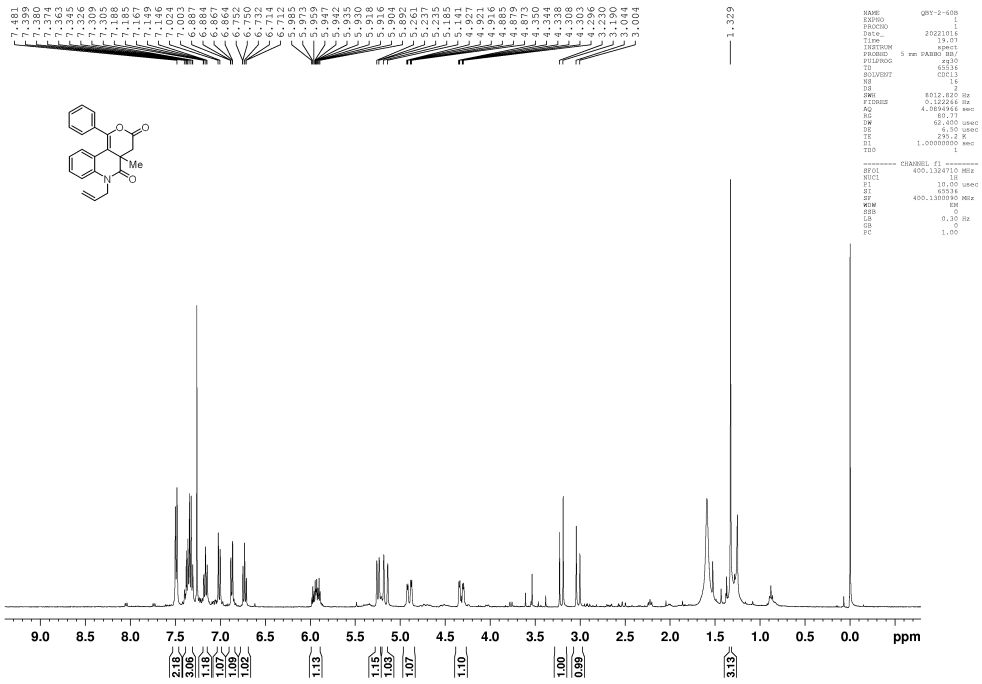


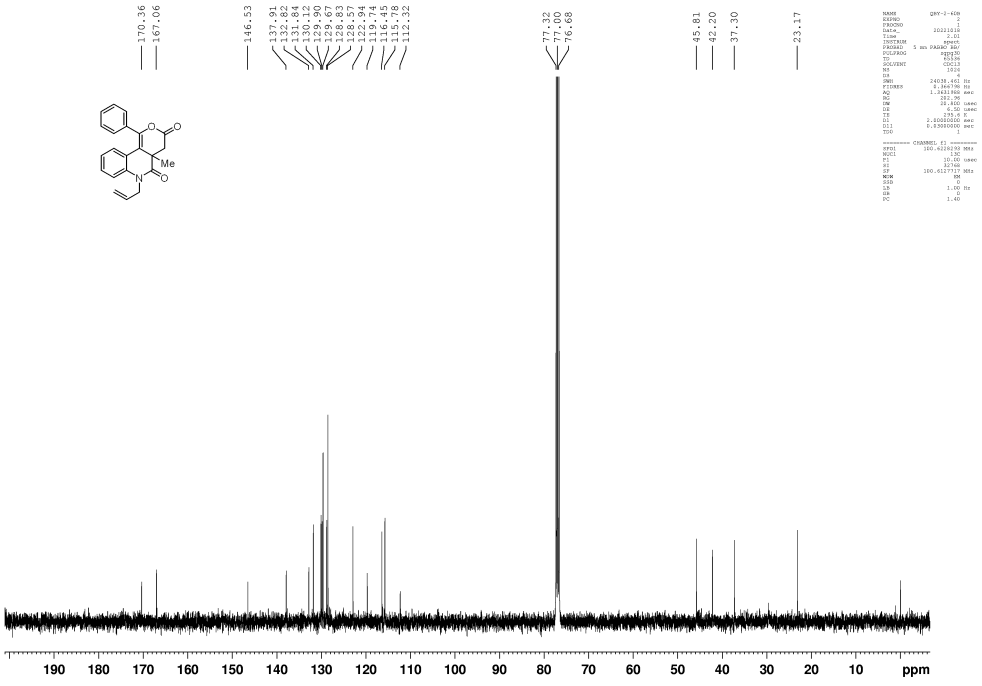
6-benzyl-4a-methyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3q)



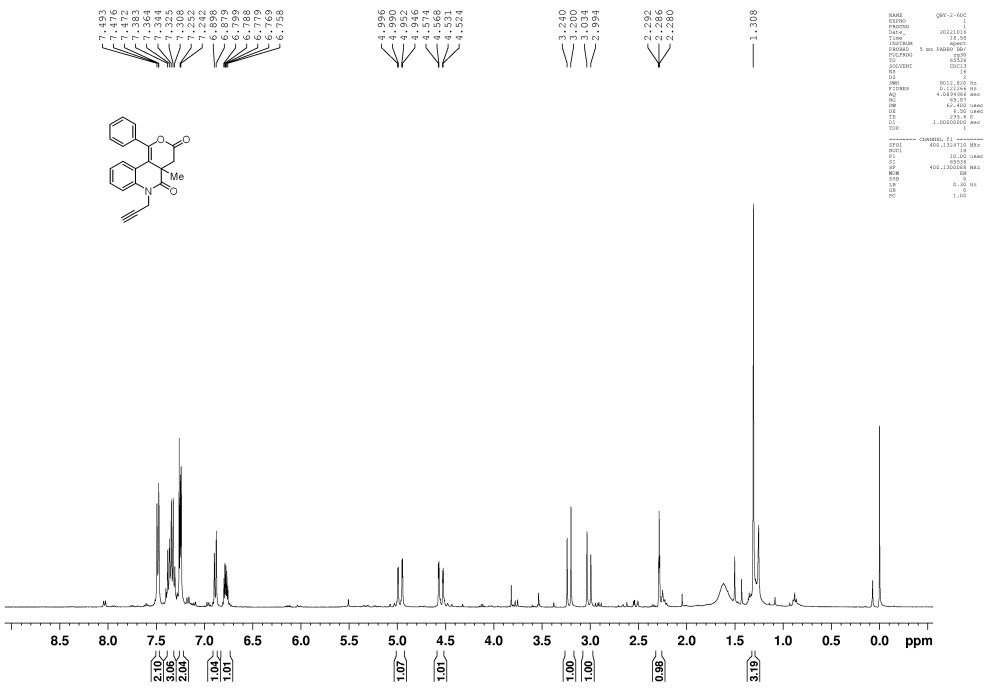


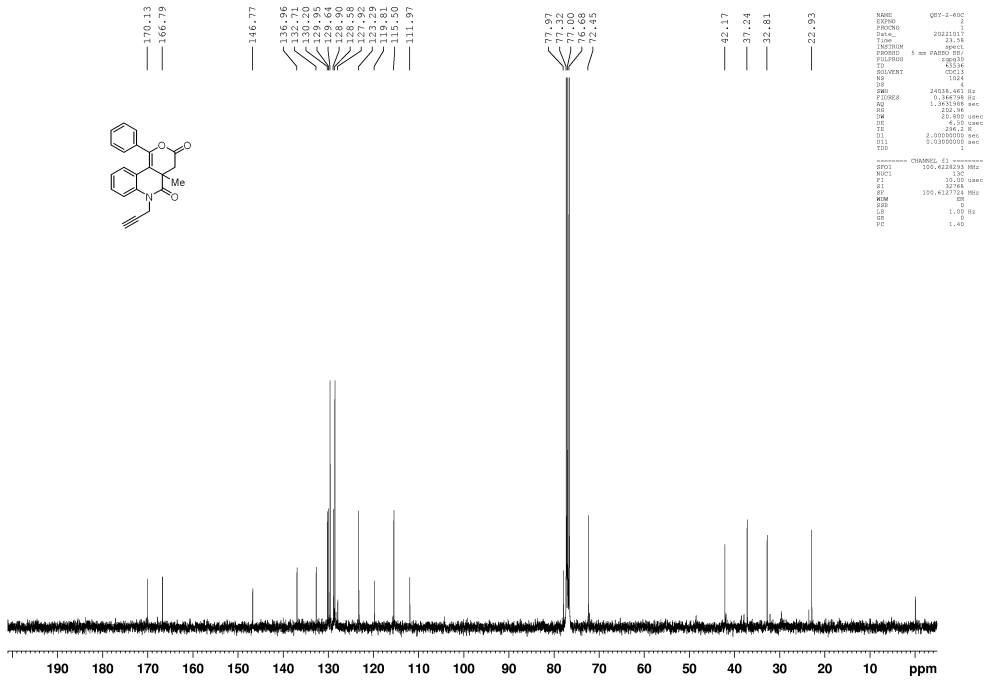
6-allyl-4a-methyl-1-phenyl-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3r)



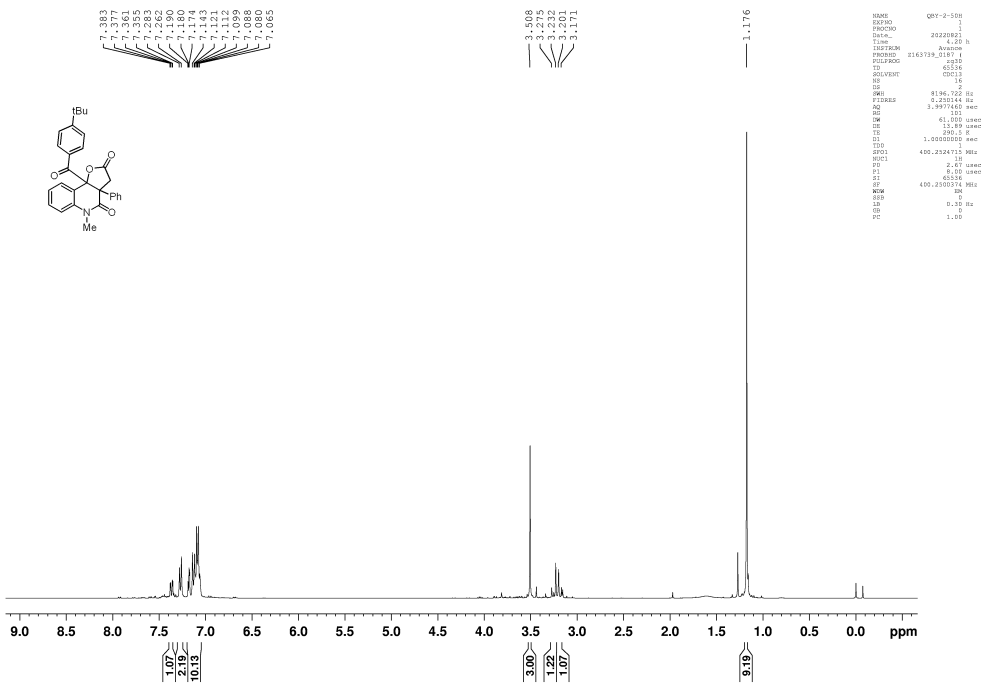


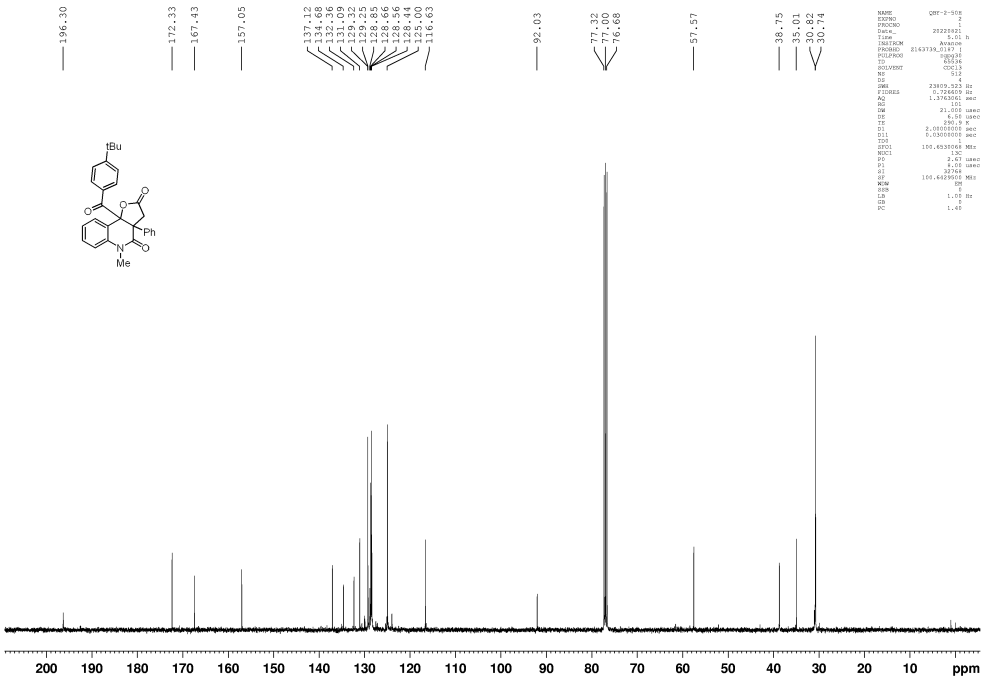
4a-methyl-1-phenyl-6-(prop-2-yn-1-yl)-4,4a-dihydro-3H-pyrano[4,3-c]quinoline-3,5(6H)-dione (3s)



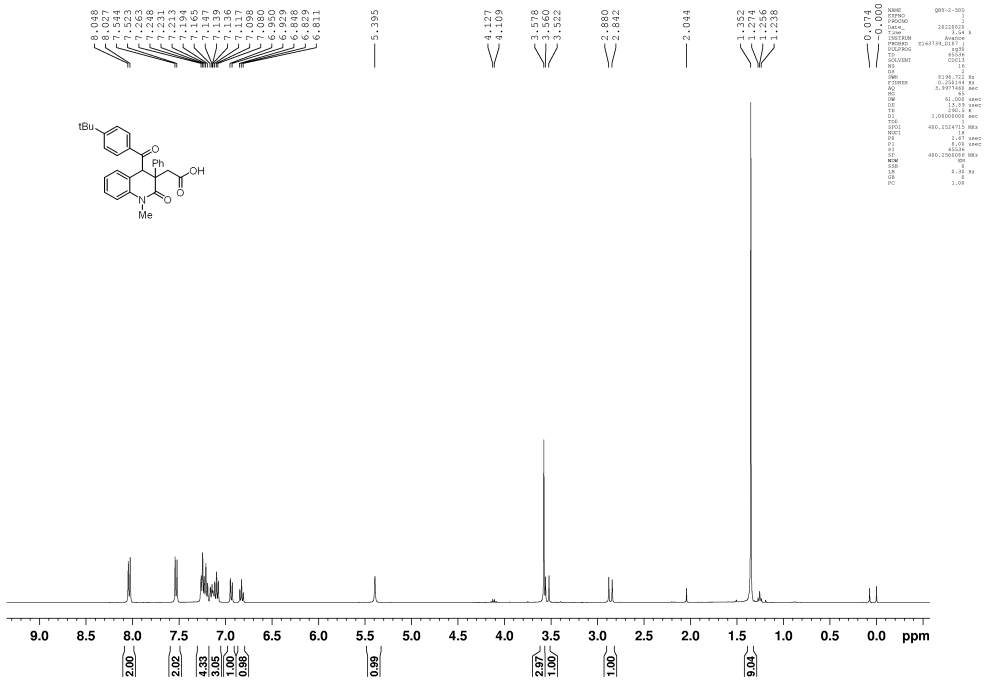


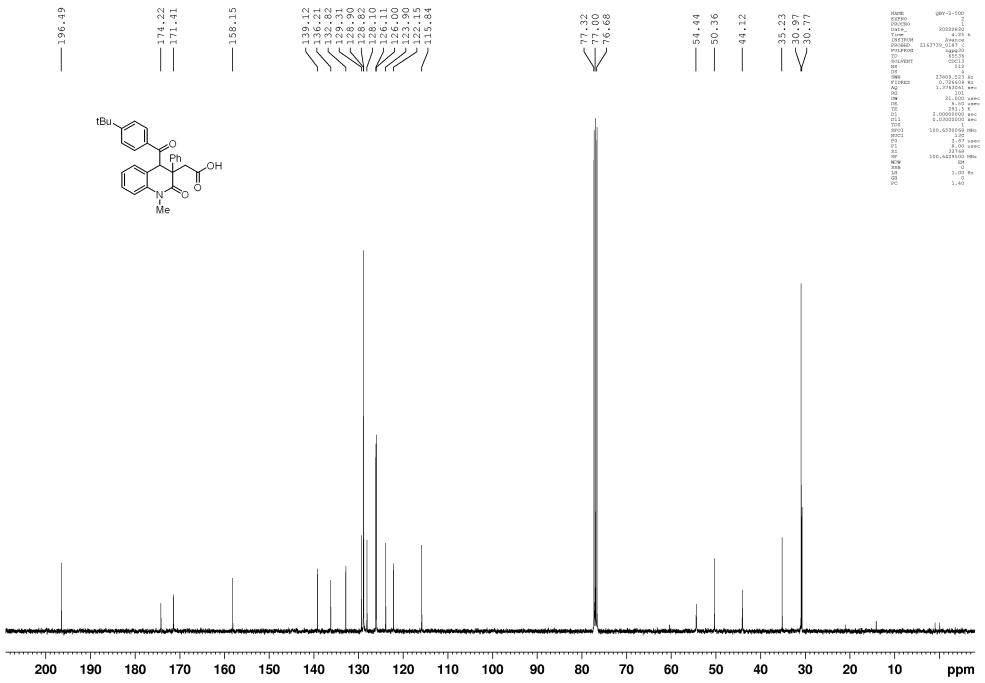
9b-(4-(tert-butyl)benzoyl)-5-methyl-3a-phenyl-3,3a,5,9b-tetrahydrofuro[3,2-c]quinoline-2,4-dione (3d-I)



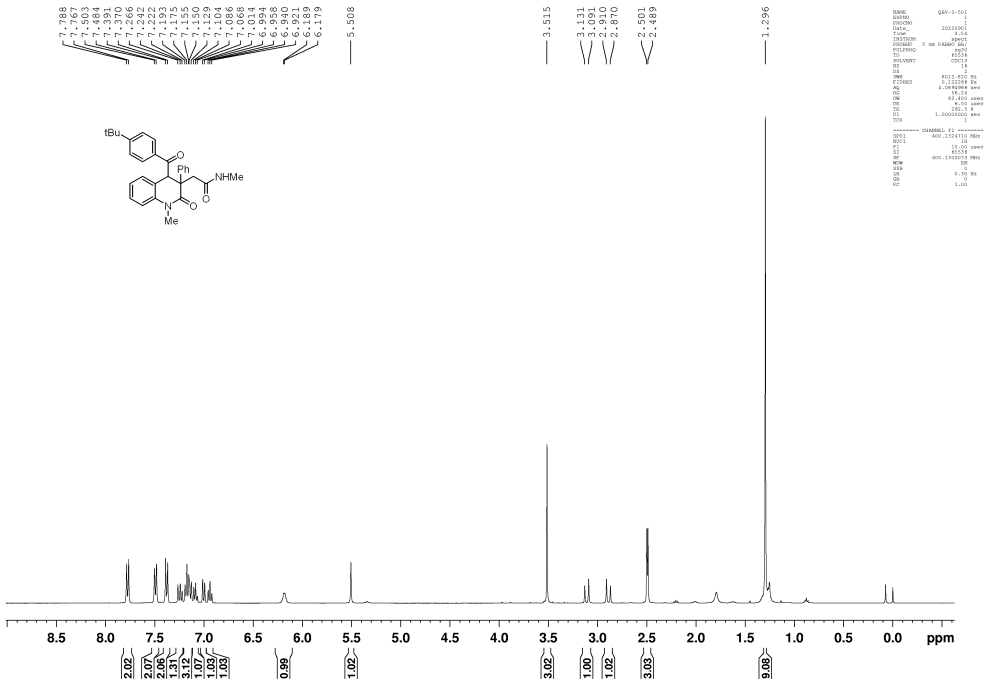


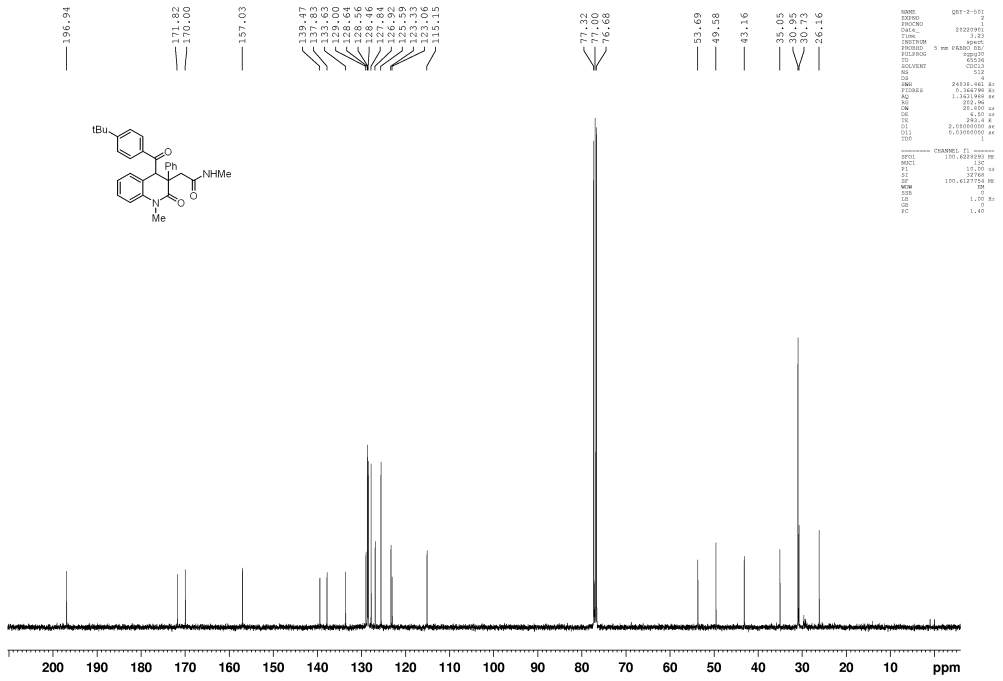
2-(4-(4-(tert-butyl)benzoyl)-1-methyl-2-oxo-3-phenyl-1,2,3,4-tetrahydroquinolin-3-yl)acetic acid (3d-II)



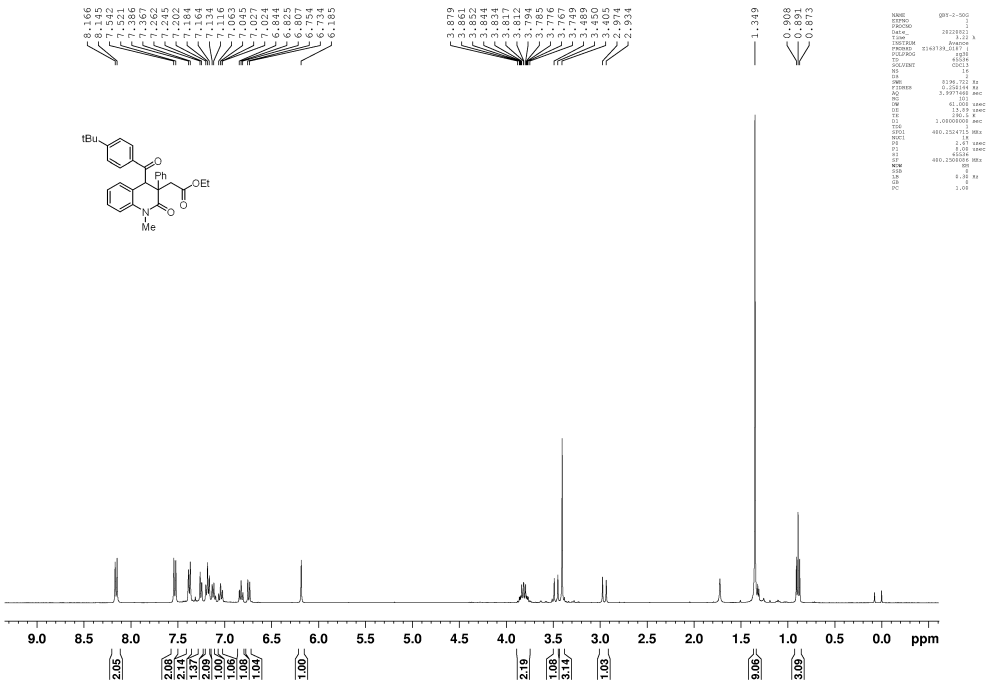


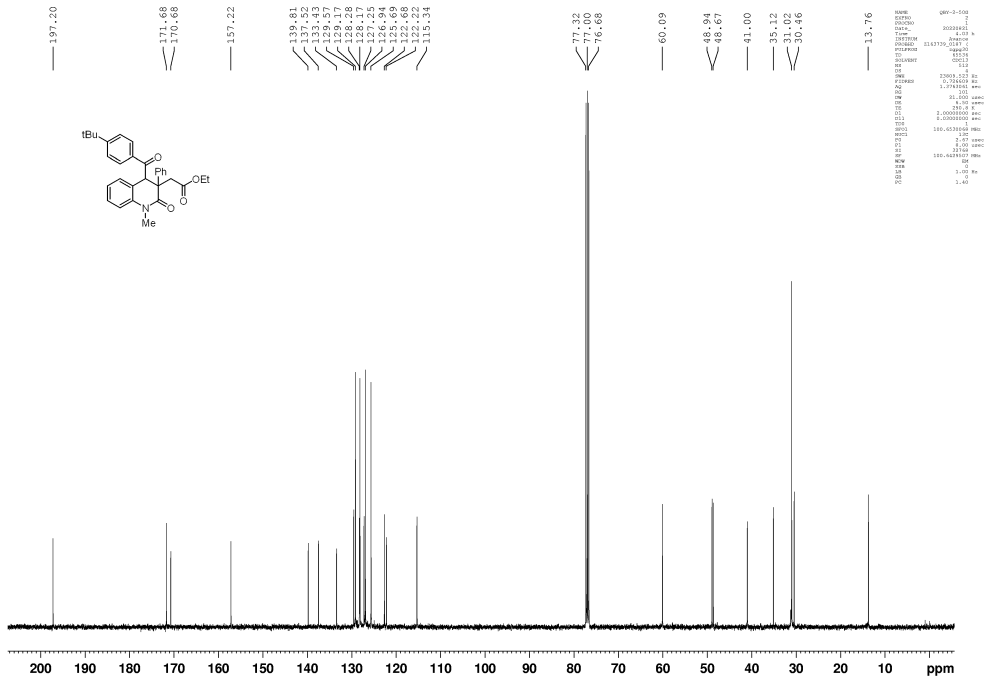
2-(4-(4-(tert-butyl)benzoyl)-1-methyl-2-oxo-3-phenyl-1,2,3,4-tetrahydroquinolin-3-yl)-N-methylacetamide (3d-III)





ethyl 2-(4-(4-(tert-butyl)benzoyl)-1-methyl-2-oxo-3-phenyl-1,2,3,4-tetrahydroquinolin-3-yl)acetate (3d-IV)





ethyl 2-(2,2,5-trimethyl-4-oxo-1-phenyl-2,3,4,5-tetrahydro-3aH-cyclopenta[c]quinolin-3a-yl)acetate (3y')