Supporting Information

Synthesis of 1,2-Dihydroquinolines by Co(III)-Catalyzed [3 + 3] Annulation of Anilides with Benzylallenes

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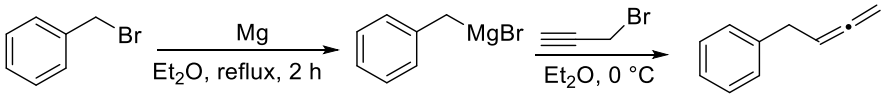
General information

Unless otherwise mentioned, all reactions were carried out under an atmosphere of nitrogen in flame dried glassware. The dry solvents used were purified by distillation over the drying agents indicated in parentheses and were transferred under nitrogen: THF (Na-benzophenone), toluene (Na), CH_2Cl_2 (CaH_2) and CH_3CN (CaH_2). Anhydrous PhCl, DCE, CH_3NO_2, $CH_3CH_2NO_2$, DMF, TFE, *o*-xylene and benzene were purchased from Acros Organics, Alfa Aesar and used as such from the commercial sources. Commercially available chemicals were obtained from Acros Organics, Sigma Aldrich Chemical Co., Alfa Aesar and used as such from the commercial sources. All the reactions were monitored by analytical thin layer chromatography (TLC) using commercial aluminum sheets precoated with silica gel (TLC Silica Gel 60 F_{254}). TLC plates were visualized by exposure to short wave ultraviolet light (254 nm). Flash chromatography was performed on siliaflash G60 (70-230 mesh) by standard techniques using appropriate mixtures of *n*-hexane and ethyl acetate. ^{1}H and ^{13}C NMR spectra were recorded on Mercury 400 MHz (**^{1}H** was recorded at rt) and Bruker DMX 600 MHz (**^{1}H** and **^{13}C NMR** were recorded at ≈ -20 to -40 °C) in solvents as indicated. Chemical shifts (δ) for ^{1}H and ^{13}C NMR spectra are given in ppm relative to TMS. The residual solvent signals were used as references for ^{1}H and ^{13}C NMR spectra and the chemical shifts converted to the TMS scale (TMS: δH = 0.00 ppm, $CDCl_3$: δH = 7.24 ppm, δC = 77.00 ppm). EI mass spectra were recorded on JEOL JMS-700. IR spectra were recorded neat on a Horiba Fourier Transform Infrared Spectroscopy FT-720. The wave numbers (υ) of recorded IR-signals are quoted in cm^{-1}. Melting points were recorded on a Fargo MP-2D apparatus. Anilides[2], *d_5*-acetanilide[2], and allenes[3] were prepared according to the previously reported procedure.

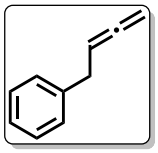
General procedure for preparation of allenes[3]



To a 100-mL round-bottomed flask containing dry magnesium turnings (2.9 g, 0.12 mol, 1.2 equiv), diethyl ether (40 mL) and 1,2-Dibromoethane (0.5 mL) were added under N_2. Then benzyl bromide (0.1 mol) was added to the reaction mixture slowly. After the addition, the mixture is heated under reflux for 2 h. Another 250-mL round-bottomed flask containing propargyl bromide (17.8 mL of an 80 wt. % solution in toluene, 0.12 mol, 1.2 equiv) and diethyl ether (20 mL) was cooled to 0 °C. The prepared Grignard reagent was added to the propargyl bromide slowly. After

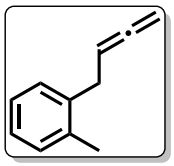
the addition, the resulting mixture is stirred for an additional 2 h at 0 °C. Then the mixture is quenched with aqueous ammonium chloride solution. The aqueous phase is extracted with diethyl ether. The combined organic phases were dried over anhydrous $MgSO_4$, and concentrated by rotary evaporation. Purification is performed by vacuum distillation to afford corresponding allene.

Buta-2,3-dien-1-ylbenzene (2a)



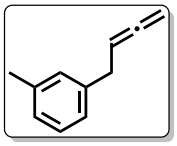
Colorless oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.37–7.32 (m, 2 H), 7.29–7.24 (m, 3 H), 5.36-5.29 (m, 1 H), 4.78–4.75 (m, 2 H), 3.42-3.39 (m, 2 H).

1-(Buta-2,3-dien-1-yl)-2-methylbenzene (2b)



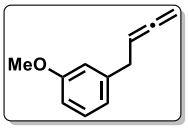
Pale yellow oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.19–7.11 (m, 4 H), 5.26–5.19 (m, 1 H), 4.69–4.66 (m, 2 H), 3.35–3.31 (m, 2 H), 2.30 (s, 3 H).

1-(Buta-2,3-dien-1-yl)-3-methylbenzene (2c)



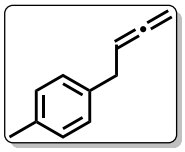
Colorless oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.19 (t, *J* = 7.6 Hz, 1 H), 7.05–7.02 (m, 3 H), 5.30–5.23 (m, 1 H), 4.74–4.71 (m, 2H), 3.35-3.31 (m, 2 H), 2.34 (S, 3 H)

1-(Buta-2,3-dien-1-yl)-3-methoxybenzene (2d)



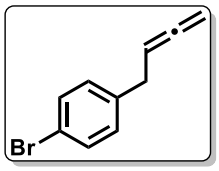
Pale yellow oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.20 (t, *J* = 8.0 Hz, 1 H), 6.81 (d, *J* = 7.2 Hz, 1 H), 6.77–6.73 (m, 2 H), 5.29–5.22 (m, 1 H), 4.73–4.70 (m, 2 H), 3.79 (S, 3 H), 3.47-3.31 (m, 2 H).

1-(Buta-2,3-dien-1-yl)-4-methylbenzene (2e)



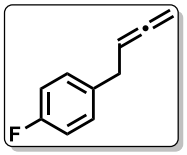
Pale yellow oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.26–7.19 (m, 4 H), 5.42–5.35 (m, 1 H), 4.85–4.82 (m, 2 H), 3.46–3.42 (m, 2 H), 2.45 (s, 3 H).

1-Bromo-4-(buta-2,3-dien-1-yl)benzene (2f)



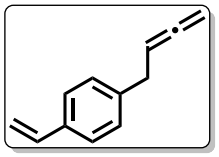
Pale yellow oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.39 (d, *J* = 8.4 Hz, 2 H), 7.08 (d, *J* = 8.4 Hz, 2 H), 5.25–5.18 (m, 1 H), 4.72–4.69 (m, 2 H), 3.30–3.26 (m, 2 H).

1-(Buta-2,3-dien-1-yl)-4-fluorobenzene (2g)



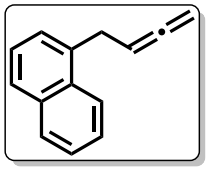
Pale yellow oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.20–7.16 (m, 2 H), 7.01–6.95 (m, 2 H), 5.29–5.22 (m, 1 H), 4.74–4.71 (m, 2 H), 3.34–3.31 (m, 2 H).

1-(Buta-2,3-dien-1-yl)-4-vinylbenzene (2h)



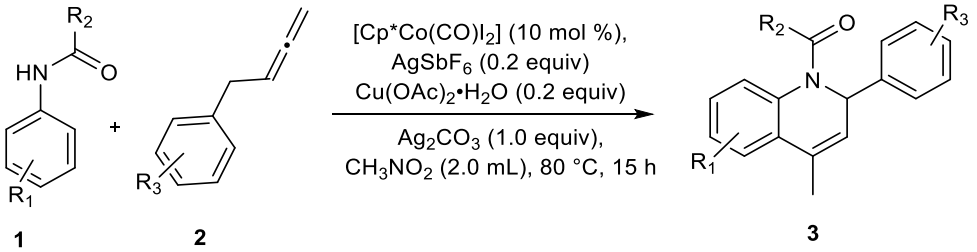
Colorless oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 7.33 (d, *J* = 8.0 Hz, 2 H), 7.17 (d, *J* = 8.0 Hz, 2 H), 6.68 (q, *J* = 10.8 Hz, 1 H), 5.69 (d, *J* = 17.6 Hz, 1 H), 5.28–5.21 (m, 1 H), 5.18 (d, *J* = 10.8 Hz, 1 H), 4.71–4.68 (m, 2 H), 3.33–3.31 (m, 2 H).

1-(Buta-2,3-dien-1-yl)naphthalene (2i)



Pale yellow oil; **^{1}H NMR** (400 MHz, $CDCl_3$): δ 8.09 (d, *J* = 8.0 Hz, 1 H), 7.89 (d, *J* = 7.6 Hz, 1 H), 7.77 (d, *J* = 8.0 Hz, 1 H), 7.57–7.51 (m, 2 H), 7.46–7.40 (m, 2 H), 5.46–5.39 (m, 1 H), 4.75–4.72 (m, 2 H), 3.85–3.82 (m, 2 H).

General procedure for Co^{III}-catalyzed [3+3] annulation of anilides with allenes



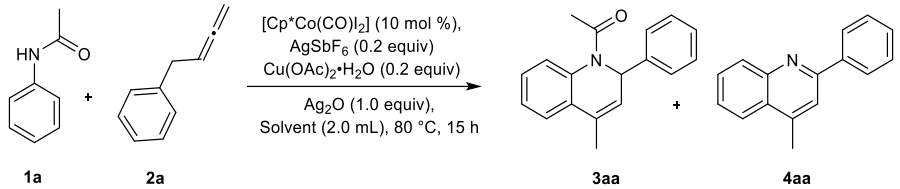
1 **2** **3**

To a sealed tube, anilides **1** (0.33 mmol), allenes **2** (0.50 mmol), $[CoCp^*(CO)I_2]$ (0.030 mmol), Ag_2CO_3 (0.33 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.070 mmol) and $AgSbF_6$ (0.070 mmol) were added inside

the glove box sealed with a rubber septum. Nitromethane (2.0 mL) was added to the sealed tube via syringe and the reaction mixture was allowed to stir at 80 °C for 15 h. Then, the mixture was cooled and diluted with CH_2Cl_2 (10 mL). The reaction mixture was filtered through a celite pad and the celite pad washed with CH_2Cl_2 (3 × 10 mL). The filtrate was concentrated under reduced pressure and the residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure product **3**.

Optimization Studies:

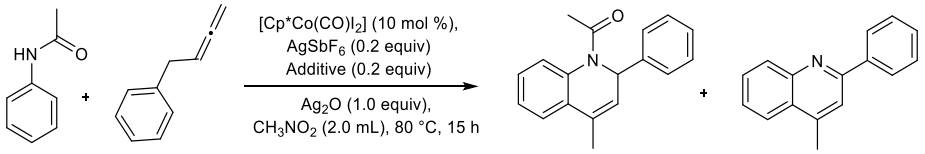
i) Table S1. Screening of Solvents[a,b]



Entry	Solvent	Yield of **3aa** (%)	Yield of **4aa** (%)
1	DCE	60	trace
2	PhCl	58	trace
3	toluene	13	-
4	CH_3NO_2	90 (86)	-
5	dioxane	-	-
6	TFE	-	-
7	ACN	-	-
8	DMF	-	-
9	o-xylene	52	-
10	$CH_3CH_2NO_2$	73	-
11[c]	CH_3NO_2	48	-
12[d]	CH_3NO_2	-	-

[a] Unless otherwise mentioned, all reactions were carried out using **1a (**0.20 mmol), **2a** (0.30 mmol), [CoCp*(CO)I_2] (10 mol %), $AgSbF_6$ (0.040 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.040 mmol), Ag_2O (0.20 mmol) and solvent (2.0 mL) at 80 °C for 15 h. [b] Yields were determined by the ^1H NMR integration method; the value in the parenthesis was isolated yield. [c] No $Cu(OAc)_2 \cdot H_2O$. [d] No $AgSbF_6$.

ii) Table S2. Screening of Acetate Additives[a,b]

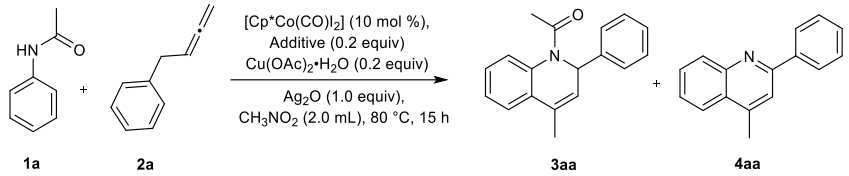


1a 2a 3aa 4aa

Entry	Additive 1	Yield of **3aa** (%)	Yield of **4aa** (%)
1	$Cu(OAc)_2 \cdot H_2O$	90 (86)	-
2	$Mn(OAc)_3 \cdot 2H_2O$	54	-
3	$Fe(OAc)_2$	14	-
4	$Cu(OAc)_2$	60	-
5	$Cu(OPiv)_2$	20	-
6	$Mn(OAc)_2$	14	-
7	CH_3COOH	48	-
8	PivOH	10	-
9	NaOAc	-	-
10	KOAc	-	-
11	CsOAc	-	-

[a] Unless otherwise mentioned, all reactions were carried out using **1a (**0.20 mmol), **2a** (0.30 mmol), $[CoCp^*(CO)I_2]$ (10 mol %), $AgSbF_6$ (0.040 mmol), Additive 1 (0.040 mmol), Ag_2O (0.20 mmol) and solvent (2.0 mL) at 80 °C for 15 h. [b] Yields were determined by the ^1H NMR integration method; the value in the parenthesis was isolated yield.

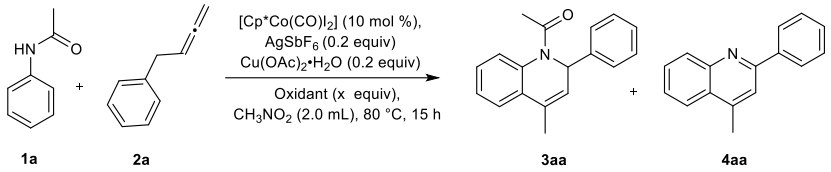
iii) Table S3. Screening of Silver Additives[a,b]



Entry	Additive 2	Yield of **3aa** (%)	Yield of **4aa** (%)
1	$AgSbF_6$	90 (86)	-
2	$AgPF_6$	82	-
3	AgOTf	74	-
4	AgBF4	66	-
5	$NaSbF_6$	16	-

[a] Unless otherwise mentioned, all reactions were carried out using **1a (**0.20 mmol), **2a** (0.30 mmol), [CoCp*(CO)I_2] (10 mol %), Additive (0.04 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.04 mmol), Ag_2O (0.20 mmol) and solvent (2.0 mL) at 80 ˚C for 15 h. [b] Yields were determined by the ^1H NMR integration method; the value in the parenthesis was isolated yield.

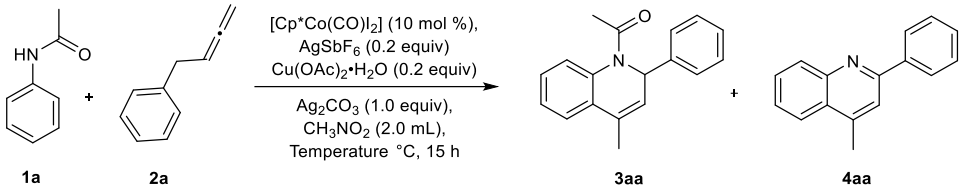
iv) Table S4. Screening of Oxidants[a,b]



Entry	Oxidant (equiv)	Yield of **3aa** (%)	Yield of **4aa** (%)
1	Ag_2O (1)	90 (86)	-
2	***Ag_2CO_3 (1)***	***95 (91)***	**-**
3[c]	Ag_2CO_3 (1)	55	-
4	$Cu(OAc)_2 \cdot H_2O$ (2)	62	-
5	AgOAc (2)	82	-
6	$Mn(OAc)_2$ (2)	47	-
7	O_2	24	14
8	$AgBF_4$ (2)	22	26
9	$AgPF_6$ (2)	-	-
10	AgOTf (2)	18	-
11	$Cu(OAc)_2$ (2)	60	-
12	Ag_2CO_3 (1) / Open air	47	15
13	Ag_2CO_3 (0.03) / Open air	30	20
14	Open air	24	37

[a] Unless otherwise mentioned, all reactions were carried out using **1a (**0.20 mmol), **2a** (0.30 mmol), [CoCp*(CO)I_2] (10 mol %), $AgSbF_6$ (0.04 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.04 mmol), Oxidant (0.20 mmol) and solvent (2.0 mL) at 80 ˚C for 15 h. [b] Yields were determined by the ^1H NMR integration method; the value in the parenthesis was isolated yield. [c] No $Cu(OAc)_2 \cdot H_2O$.

v) Table S5. Screening of Temperatures[a,b]

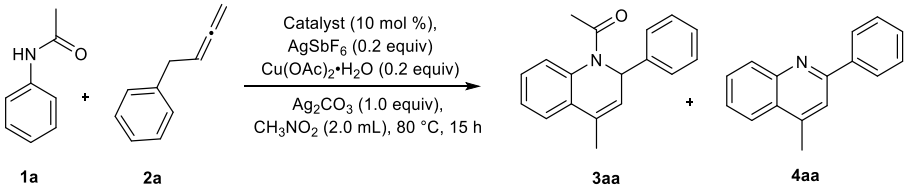


1a 2a 3aa 4aa

Entry	Temperature °C	Yield of **3aa** (%)	Yield of **4aa** (%)
1	60	76	-
2	***80***	***95 (91)***	**-**
3	100	78	9
4	120	75	10

[a] Unless otherwise mentioned, all reactions were carried out using **1a (**0.20 mmol), **2a** (0.30 mmol), [CoCp*(CO)I_2] (10 mol %), $AgSbF_6$ (0.040 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.04 mmol), Oxidant (0.20 mmol) and solvent (2.0 mL) at temperature ˚C for 15 h. [b] Yields were determined by the ^1H NMR integration method; the value in the parenthesis was isolated yield.

vi) Table S6. Screening of Catalysts[a,b]



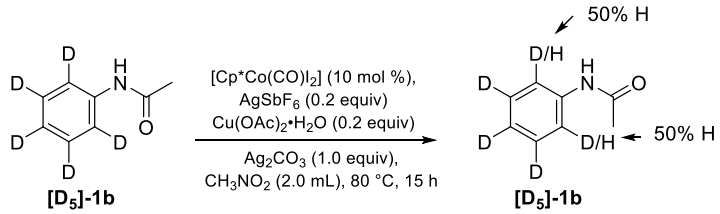
1a 2a 3aa 4aa

Entry	Catalyst	Yield of **3aa** (%)	Yield of **4aa** (%)
1	***[CoCp*(CO)I_2]***	***95 (91)***	**-**
2	$Co(OAc)_2 \cdot 4H_2O$	-	-
3	$Co(OAc)_2$	-	-
4	$Co(acac)_3$	-	-
5	CoI_2	-	-
6	$CoBr_2$	-	-
7	$[RhCp^*Cl_2]_2$	35	-
8	$[IrCp^*Cl_2]_2$	-	-
9[c]	[CoCp*(CO)I_2]	56	-
10	$[RuCl_2(p\text{-cymene})]_2$	-	-
11	$Pd(OAc)_2$	-	-

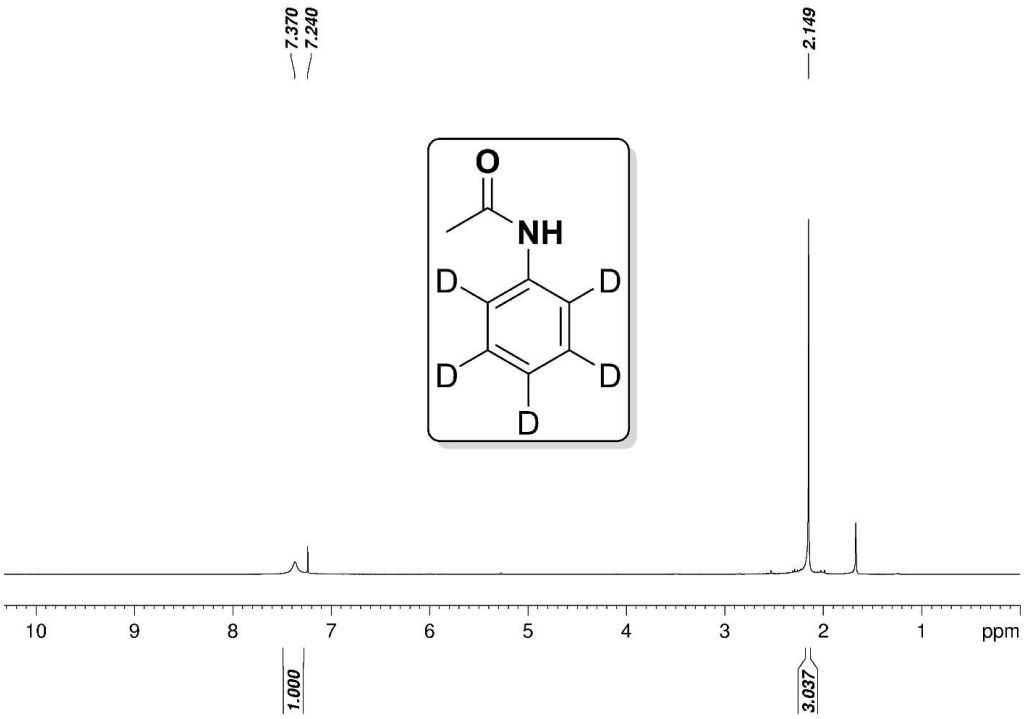
[a] Unless otherwise mentioned, all reactions were carried out using **1a (**0.20 mmol), **2a** (0.30 mmol), Catalyst (10 mol %), $AgSbF_6$ (0.040 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.040 mmol), Ag_2O (0.20 mmol) and solvent (2.0 mL) at 80 ˚C for 15 h. [b] Yields were determined by the ^1H NMR integration method; the value in the parenthesis was isolated yield. [c] [CoCp*(CO)I_2] (5 mol %).

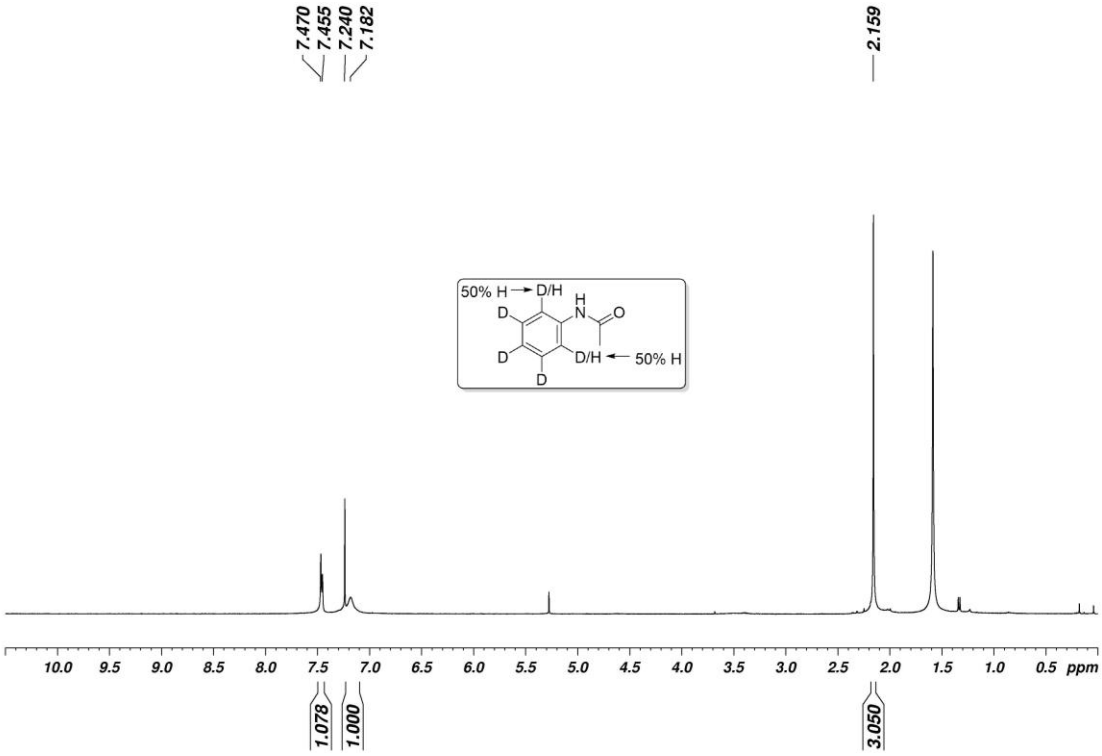
Mechanistic studies:

i) Reaction of D_5-1b without Allene

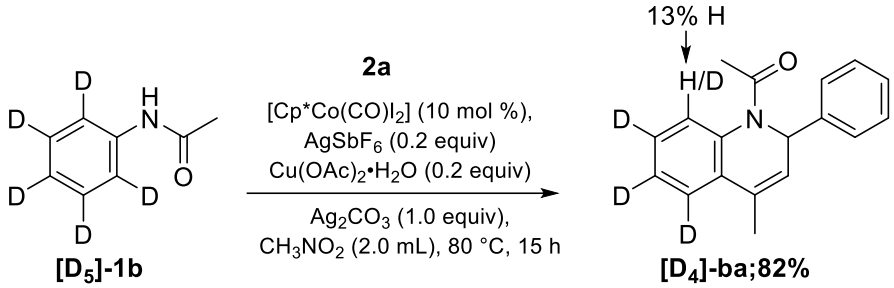


To a sealed tube, **D_5-1b** (0.33 mmol), [CoCp*(CO)I_2] (0.03 mmol), Ag_2CO_3 (0.33 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.070 mmol) and $AgSbF_6$ (0.070 mmol) were added inside the glove box and sealed with a rubber septum. CH_3NO_2 (2.0 mL) was added to the seal tube via syringe and the reaction mixture was allowed to stir at 80 ˚C for 15 h. Then, the mixture was cooled and diluted with CH_2Cl_2 (10 mL). The mixture was filtered through a celite pad and the celite pad was washed with CH_2Cl_2 (3 × 10 mL). The filtrate was concentrated under reduced pressure. The H^1NMR analysis indicates that there is a 50% ortho H/D exchange.



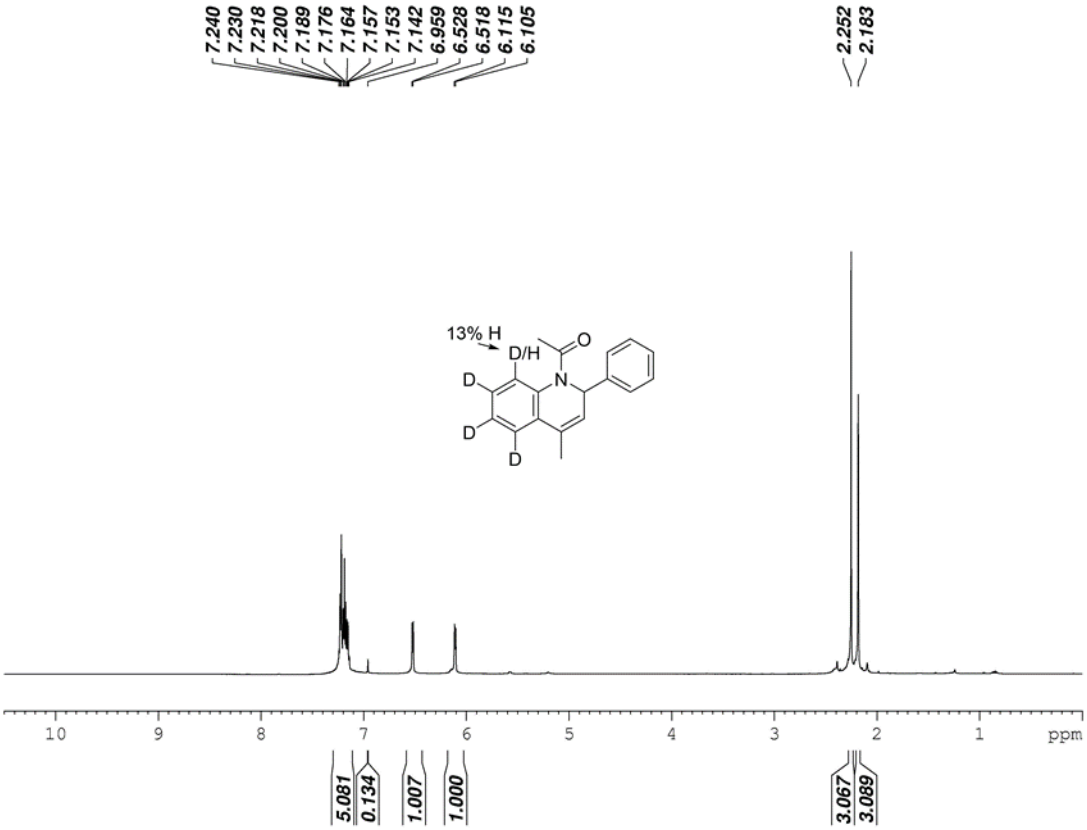


ii) Reaction of D_5-1b with Allene



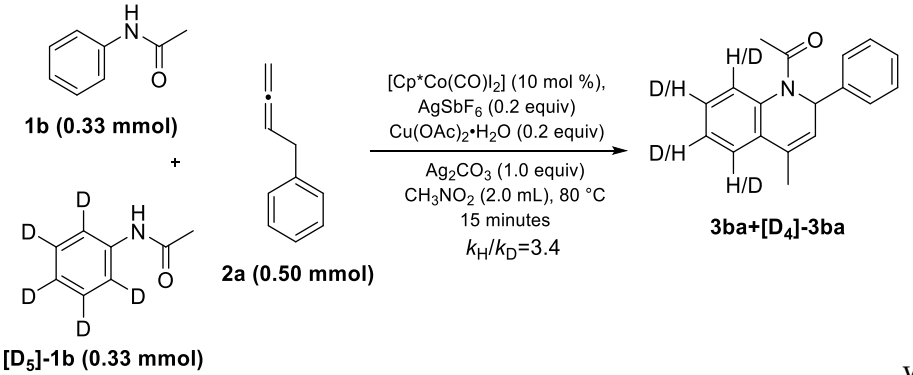
To a sealed tube, [CoCp*(CO)I_2] (0.03 mmol), Ag_2CO_3 (0.33 mmol), **D_5-1b** (0.33 mmol), **2a** (0.50 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.070 mmol) and $AgSbF_6$ (0.070 mmol) were added inside a glove box and sealed with a rubber septum. CH_3NO_2 (2.0 mL) was added to the sealed tube via syringe and the reaction mixture was allowed to stir at 80 °C for 15 h. Then, the mixture was cooled and diluted with CH_2Cl_2 (10 mL). The mixture was filtered through a celite pad and the celite pad was washed with CH_2Cl_2 (3 × 10 mL). The filtrate was concentrated under reduced pressure and the

residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure product [**D_4**]-**ba** in 82% yield. The H^1NMR analysis indicates that there is a 13% ortho H/D exchange.



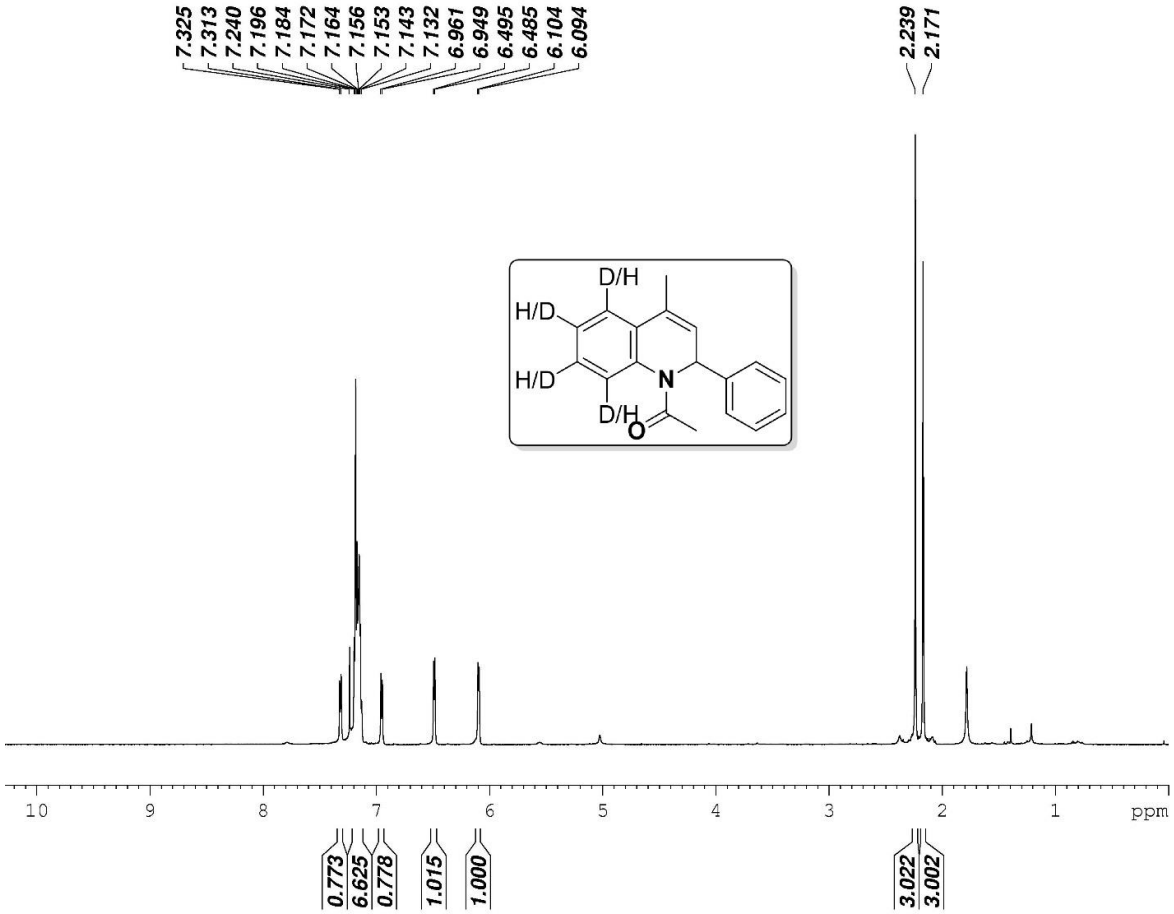
iii) Kinetic Isotope Experiments

a) Competition experiment

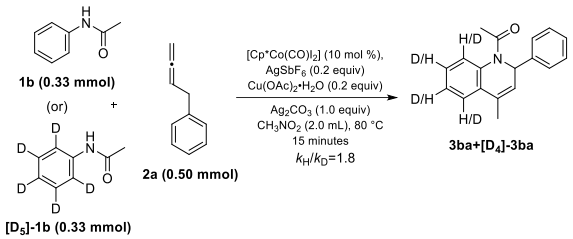


we

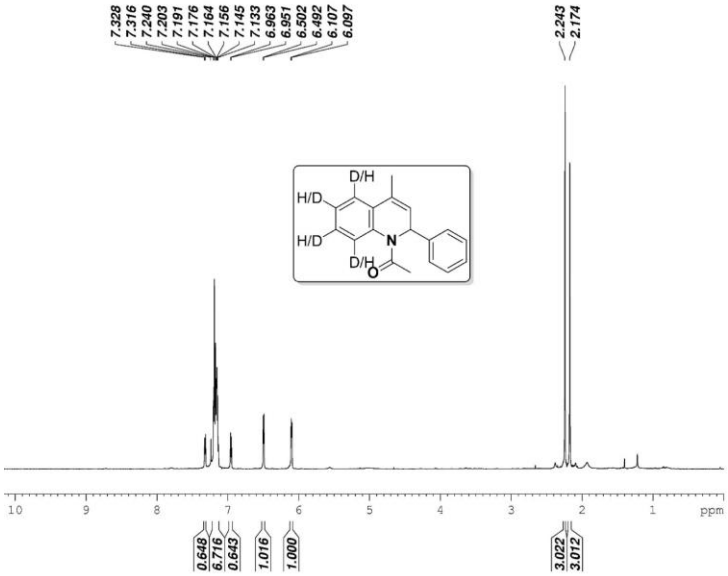
To a sealed tube, [CoCp*(CO)I_2] (0.03 mmol), Ag_2CO_3 (0.33 mmol), **1b** (0.33 mmol), **D_5-1b** (0.33 mmol), allenes **2** (0.50 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.07 mmol) and $AgSbF_6$ (0.07 mmol) were added inside the glove box and sealed with a rubber septum. CH_3NO_2 (2.0 mL) was added to the sealed tube via syringe and the reaction mixture was allowed to stir at 80 ˚C for 15 minutes. Then, the mixture was cooled and diluted with CH_2Cl_2 (10 mL). The mixture was filtered through a Celite pad and the Celite pad was washed with CH_2Cl_2 (3 × 10 mL). The filtrate was concentrated under reduced pressure and the residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure products **3ba**+**[D_4]-3ba** in 16% yield. An intermolecular kinetic isotopic effect KIE ≈ 3.4 was determined by ^{1}H NMR.



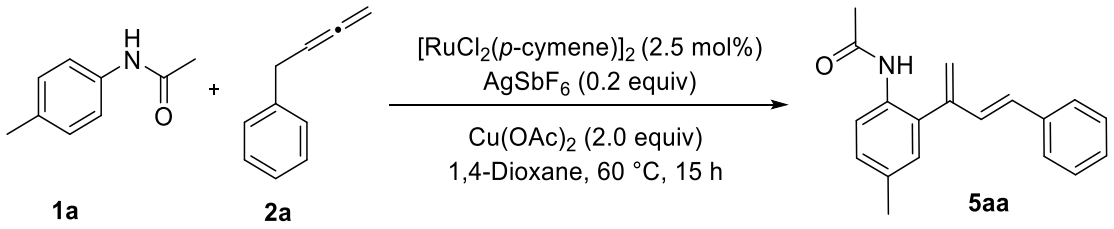
b) Parallel experiment



To two separate sealed tubes, [CoCp*(CO)I_2] (0.030 mmol), Ag_2CO_3 (0.33 mmol), **1b** or **D_5-1b** (0.33 mmol), allenes **2** (0.50 mmol), $Cu(OAc)_2 \cdot H_2O$ (0.070 mmol) and $AgSbF_6$ (0.070 mmol) were added inside the glove box and sealed with a rubber septum. CH_3NO_2 (2.0 mL) was added to the sealed tube via syringe and the reaction mixture was allowed to stir at 80 °C for 15 minutes. Then, the mixture was cooled and diluted with CH_2Cl_2 (10 mL). Both the mixtures were mixed and filtered through a celite pad and the celite pad was washed with CH_2Cl_2 (3 × 10 mL). The filtrate was concentrated under reduced pressure and the residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure products **3ba**+**[D_4]-3ba** in 13% yield. An intermolecular kinetic isotopic effect KIE ≈ 1.8 was determined by ^{1}H NMR..

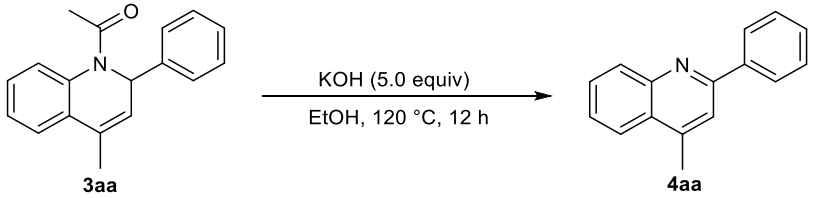


iv) Procedure for synthesis of (*E*)-*N*-(4-Methyl-2-(4-phenylbuta-1,3-dien-2-yl)phenyl)acetamide (5aa)



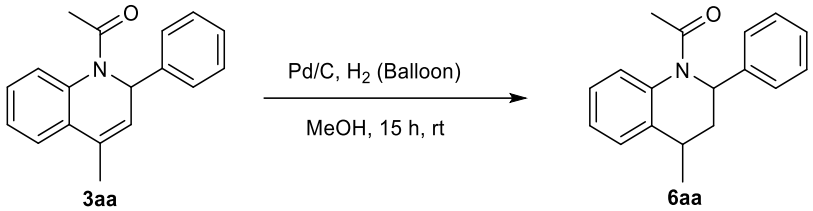
To a sealed tube, $[RuCl_2(p\text{-cymene})]_2$ (2.5 mol%), 4-methyl acetanilide **1a** (1.34 mmol), allenes **2a** (2.00 mmol), $Cu(OAc)_2$ (2.68 mmol) and $AgSbF_6$ (0.27 mmol) were added inside the glove box and closed with a rubber septum. 1,4-Dioxane (10 mL) was added to the sealed tube via syringe and the reaction mixture was allowed to stir at 60 °C for 15 h. Then, the mixture was cooled and diluted with CH_2Cl_2 (20 mL). The mixture was filtered through a celite pad and the celite pad was washed with CH_2Cl_2 (3 × 10 mL). The filtrate was concentrated under reduced pressure and the residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure product **5aa** in 33% yield.

Procedure for synthesis of 4aa



To a solution of 1-(4-Methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one **3aa** (0.38 mmol) in EtOH (10 mL) was added potassium hydroxide (1.90 mmol) under nitrogen atmosphere. The reaction mixture was allowed to stir at 120 °C for 12 h under N_2 atmosphere. Then, the reaction mixture was diluted with water and extracted with CH_2Cl_2 (2 × 10 mL). The combined organic layer was washed with brine (2 × 10 mL), dried over $MgSO_4$ and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure product **4aa** in 87%.

Procedure for the synthesis of 6aa



3aa 6aa

To a solution of 1-(4-methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one **3aa** (0.38 mmol) in MeOH (10 mL) was added Pd/C (10 mol %) under a nitrogen atmosphere. The reaction mixture was allowed to stir at room temperature for 15 h under H_2 atmosphere (1 atm). Then, the reaction mixture was diluted with CH_2Cl_2 (10 mL) and the mixture was filtered through a celite pad. The filtrate was concentrated under reduced pressure and the residue was purified by silica gel column chromatography using hexane/ethyl acetate as eluent to afford the desired pure product **6aa** in 93%.

References

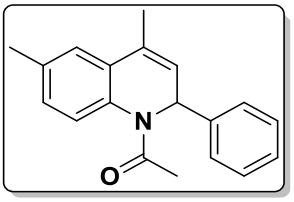
(1) Perrin, D. D.; Armarego, W. L. F. *In Purification of Laboratory Chemicals*, 3rd ed.; Pergamon Press: Oxford, 1988.

(2) Stuart, D. R.; Bertrand-Laperle, M.; Burgess, K. M. N.; Fagnou, K. *J. Am. Chem. Soc*., **2008**, *130*, 16474–16475.

(3) Ni, S.; Zhu, C.; Chen, J.; Ma, S. *Org. Synth.* **2013,** *90*, 327–337.

Spectroscopic Data

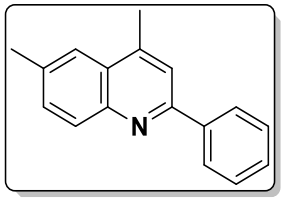
1-(4,6-Dimethyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3aa)



White solid: m.p. 109-112 ˚C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.21–7.12 (m, 6 H), 6.95 (dd, *J* = 7.8 Hz, *J* = 1.2 Hz, 1 H), 6.85 (d, *J* = 8.4 Hz, 1 H), 6.49 (d, *J* = 6.0 Hz, 1 H), 6.09 (dd, *J* = 6.0 Hz, *J* = 1.2 Hz, 1 H), 2.32 (s, 3 H), 2.23 (s, 3 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 138.8, 134.9, 132.2, 130.7, 129.6, 128.1, 127.6, 127.3, 126.3, 124.4, 123.9, 52.3, 22.6, 21.1, 18.3;

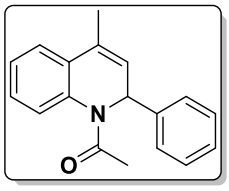
HRMS (EI^+): calcd for $C_{19}H_{19}NO$ 277.1467, found 277.1464; **IR** (KBr): 2915, 1820, 1658, 1493 and 1033 cm^{-1}.

4,6-Dimethyl-2-phenylquinoline (4aa)



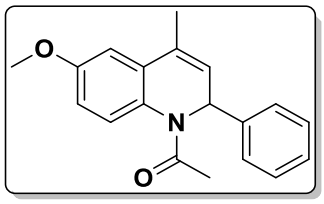
Yellow solid: m.p. 88-91 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 8.12–8.10 (m, 2 H), 8.05 (d, *J*=8.4 Hz, 1 H), 7.73 (s, 1 H), 7.66 (d, *J*=0.6 Hz, 1 H), 7.53 (dd, *J*=8.4 Hz, *J*=1.8 Hz, 1 H), 7.50–7.48 (m, 2 H), 7.43–7.40 (m, 1 H), 2.72 (s, 3 H), 2.55 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 156.2, 146.5, 144.0, 139.8, 135.8, 131.5, 129.9, 129.0, 128.7, 127.4, 127.1, 122.6, 119.7, 21.8, 19.0; **HRMS** (EI^+): calcd for $C_{17}H_{15}N$ 233.1204 found 233.1202; **IR** (KBr): 2923, 1596, 1349, 817 and 701 cm^{-1}.

1-(4-Methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3ba)



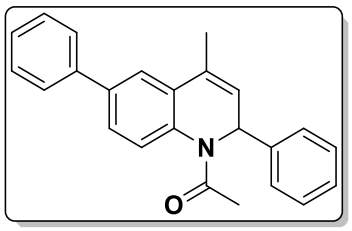
Orange solid: m.p. 108-110 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.33–7.32 (m, 1 H), 7.22–7.13 (m, 7 H), 6.96–6.95 (m, 1 H), 6.52 (d, *J* = 6.0 Hz, 1 H), 6.11 (d, *J* = 6.0 Hz, 1 H) 2.24 (s, 3 H), 2.18 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.0, 138.7, 134.8, 130.6, 129.8, 128.1, 127.3, 127.0, 126.5, 125.3, 124.6, 123.3, 52.4, 22.6, 18.2; **HRMS** (EI^+): calcd for $C_{18}H_{17}NO$ 263.1310, found 263.1304; **IR** (KBr): 2946, 1658, 1488, 1373 and 817 cm^{-1}.

1-(6-Methoxy-4-methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3ca)



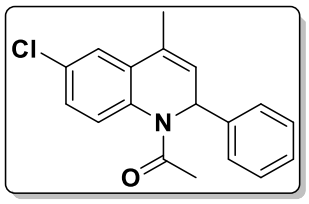
White solid: m.p. 105-108 ˚C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.19–7.13 (m, 5 H), 6.87 (d, *J* = 9.0 Hz, 1 H), 6.84 (d, *J* = 3.0 Hz, 1 H), 6.67 (dd, *J* = 9.0 Hz, *J*=3.0 Hz, 1 H), 6.49 (d, *J* = 6.0 Hz, 1 H), 6.11 (dd, *J* = 6.6 Hz, *J* = 1.2 Hz, 1 H), 3.78 (s, 3 H), 2.20 (s, 3 H), 2.15 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.0, 156.8, 138.5, 131.1, 130.7, 128.1, 127.8, 127.3, 127.3, 127.1, 125.5, 55.2, 52.2, 22.5, 18.2; **HRMS** (EI^+): calcd for $C_{19}H_{19}NO_2$ 293.1416, found 293.1417; **IR** (KBr): 2946, 1743, 1656, 1033 and 817 cm^{-1}.

1-(4-Methyl-2,6-diphenylquinolin-1(2*H*)-yl)ethan-1-one (3da)



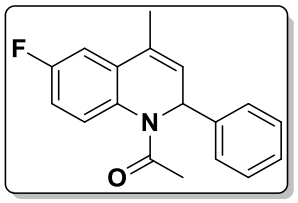
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.59–7.58 (m, 2 H), 7.53 (d, *J* = 1.8 Hz, 1 H), 7.46 (t, *J* = 7.8 Hz, 2 H), 7.38–7.36 (m, 2 H), 7.27 (d, *J* = 7.2 Hz, 2 H), 7.21 (t, *J* = 7.2 Hz, 2 H), 7.18 (d, *J* = 7.2 Hz, 1 H), 7.04 (d, *J* = 8.4 Hz, 1 H), 6.55 (d, *J* = 6.0 Hz, 1 H), 6.16 (d, *J* = 6.0 Hz, 1 H), 2.31 (s, 3 H), 2.24 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 140.0, 138.7, 138.0, 134.0, 130.6, 130.1, 128.7, 128.2, 127.4, 127.3, 127.3, 126.7, 125.7, 124.8, 122.0, 52.5, 22.8, 18.3; **HRMS** (EI^+): calcd for $C_{24}H_{21}NO$ 339.1623, found 339.1627; **IR** (KBr): 2915, 1774, 1481, 1373 and 701 cm^{-1}.

1-(6-Chloro-4-methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3ea)



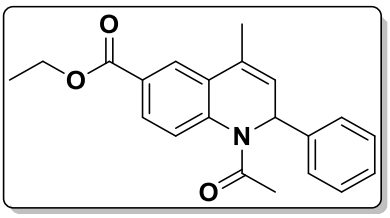
White solid: m.p. 100-103 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.29 (d, *J* = 1.8 Hz, 1 H), 7.20–7.15 (m, 5 H), 7.11 (dd, *J* = 8.4 Hz, *J* = 1.8 Hz, 1 H), 6.88 (d, *J* = 8.4 Hz, 1 H), 6.49 (d, *J* = 6.0 Hz, 1 H), 6.14 (d, *J* = 6.0 Hz, 1 H), 2.22 (s, 3 H), 2.15 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 169.9, 138.1, 133.2, 131.4, 130.6, 129.9, 128.2, 127.8, 127.6, 127.3, 126.8, 125.6, 123.4, 52.3, 22.6, 18.1; **HRMS** (EI^+): calcd for $C_{18}H_{16}ClNO$ 297.0920, found 297.0917; **IR** (KBr): 2915, 1743, 1658, 1103 and 601 cm^{-1}.

1-(6-Fluoro-4-methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3fa)



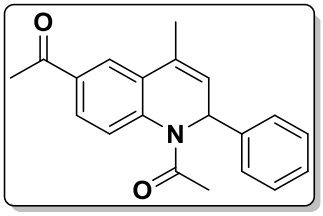
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.7.21–7.15 (m, 5 H), 7.02 (dd, *J* = 9.0 Hz, *J* = 2.4 Hz, 1 H), 6.91 (dd, *J* = 9.0 Hz, *J* = 5.4 Hz, 1 H), 6.85 (td, *J* = 8.4 Hz, *J* = 3.0 Hz, 1 H), 6.52 (d, *J* = 6.0 Hz, 1 H), 6.16 (d, *J* = 6.0 Hz, 1 H), 2.21 (s, 3 H), 2.15 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.0, 160.0 (d, *J* = 243 Hz), 138.2, 131.8, 131.7, 130.6, 130.2, 128.2, 127.8, 127.5, 127.3, 125.8, 125.8, 113.5 (d, *J* = 22.5 Hz), 110.3 (d, *J* = 22.5 Hz), 52.2, 22.5, 18.1; **HRMS** (EI^+): calcd for $C_{18}H_{16}FNO$ 281.1216, found 281.1211; **IR** (KBr): 2946, 1743, 1650, 1272 and 1033 cm^{-1}.

Ethyl 1-acetyl-4-methyl-2-phenyl-1,2-dihydroquinoline-6-carboxylate (3ga)



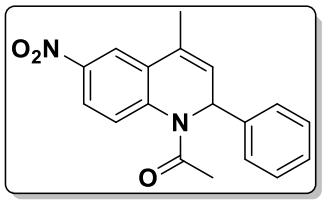
White solid: m.p. 155-158 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.99 (s, 1 H), 7.84 (d, *J* = 7.2 Hz, 1 H), 7.17–7.15 (m, 5 H), 7.00 (d, *J* = 8.4 Hz, 1 H), 6.47 (d, *J* = 6.0 Hz, 1 H), 6.12 (d, *J* = 6.0 Hz, 1 H), 4.35–4.34 (m, 2 H), 2.26 (s, 3 H), 2.23 (s, 3 H), 1.36 (t, *J*=7.2 Hz, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 169.9, 166.0, 138.8, 138.2, 130.3, 129.6, 128.4, 128.2, 127.6, 127.3, 127.0, 126.8, 124.7, 124.1, 61.1, 52.7, 22.9, 18.3, 14.2; **HRMS** (EI^+): calcd for $C_{21}H_{21}NO_3$ 335.1521, found 335.1529; **IR** (KBr): 2977, 1712, 1666, 1025 and 825 cm^{-1}.

1,1'-(4-Methyl-2-phenylquinoline-1,6(2*H*)-diyl)bis(ethan-1-one) (3ha)



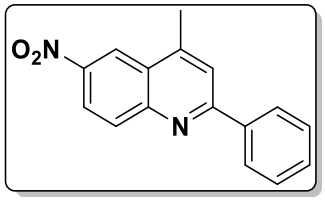
White solid: m.p. 134-137 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.93 (s, 1 H), 7.75 (d, *J* = 7.2 Hz, 1 H), 7.17 (s, 5 H), 7.02 (d, *J* = 8.4 Hz, 1 H), 6.46 (d, *J* = 6.0 Hz, 1 H), 6.13 (d, *J* = 5.4 Hz, 1 H), 2.59 (s, 3 H), 2.26 (s, 3 H), 2.23 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 197.4, 169.9, 139.1, 138.2, 133.4, 130.2, 129.7, 128.2, 127.7, 127.6, 127.3, 127.2, 124.1, 123.2, 52.7, 26.7, 22.9, 18.3; **HRMS** (EI^+): calcd for $C_{20}H_{19}NO_2$ 305.1416, found 305.1413; **IR** (KBr): 2946, 1673, 1558, 917 and 817 cm^{-1}.

1-(4-Methyl-6-nitro-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3ia)



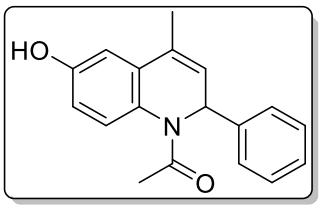
Brown solid: m.p. 157-160 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 8.21 (d, *J* = 1.8 Hz, 1 H), 8.06 (dd, *J* = 8.4 Hz, *J* = 1.8 Hz, 1 H), 7.19–7.17 (m, 5 H), 7.08 (d, *J* = 9.0 Hz, 1 H), 6.49 (d, *J* = 6.0 Hz, 1 H), 6.22 (d, *J* = 6.0 Hz, 1 H), 2.30 (s, 3 H), 2.26 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 169.9, 144.1, 140.5, 137.6, 130.3, 129.5, 128.4, 127.9, 127.2, 124.3, 122.5, 118.8, 52.9, 23.0, 18.2; **HRMS** (EI^+): calcd for $C_{18}H_{16}N2O_3$ 308.1161, found 308.1159; **IR** (KBr): 2923, 1812, 1673, 1342 and 817 cm^{-1}.

4-Methyl-6-nitro-2-phenylquinoline (4ia)



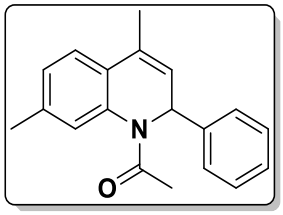
Brown solid: m.p. 177-180 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 8.94 (d, *J* = 2.4 Hz, 1 H), 8.45 (dd, *J* = 9.0 Hz, *J* = 2.4 Hz, 1 H), 8.24 (d, *J* = 9.0 Hz, 1 H), 8.18–8.17 (m, 2 H), 7.85 (s, 1 H), 7.55–7.48 (m, 3 H), 2.84 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 160.3, 150.5, 147.1, 144.9, 138.5, 131.8, 130.3, 129.0, 127.7, 126.2, 122.9, 121.2, 120.8, 19.1; **HRMS** (EI^+): calcd for $C_{16}H_{12}N_2O_2$ 264.0899, found 264.0895; **IR** (KBr): 2946, 1820, 1550, 1342 and 709 cm^{-1}.

1-(6-Hydroxy-4-methyl-2-phenylquinolin-1(2H)-yl)ethan-1-one (3ja)



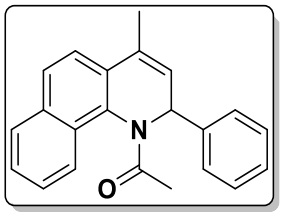
Yellow solid: m.p. 155-157 ˚C; **^{1}H NMR** (600 MHz, CD_3OD): δ 7.20-7.15 (m, 5 H), 6.95 (d, *J* = 9.0 Hz, 1 H), 6.81 (d, *J* = 3.0 Hz, 1 H), 6.61 (dd, *J* = 2.0 Hz, *J* = 8.4 Hz, 1 H), 6.43 (d, *J* = 6.0 Hz, 1 H), 6.18 (d, *J* = 6.6 Hz, 1 H), 2.19 (s, 3 H), 2.15(s, 3 H); **^{13}C NMR** (150 MHz, CD_3OD): δ 172.4, 157.2, 139.8, 132.7, 132.6, 129.3, 128.6, 128.5, 127.5, 127.3, 127.2, 114.7, 111.1, 53.6, 22.5, 18.4; **HRMS** (EI^+): calcd for $C_{18}H_{17}NO_2$ 279.1259, found 323.1263; **IR** (KBr): 3665, 2340, 1677, 1481, 709 and 517 cm^{-1}.

1-(4,7-Dimethyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3ka)



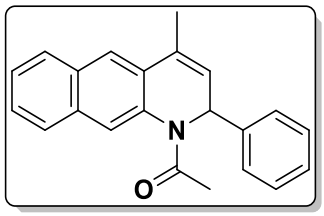
Colourless oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.22–7.14 (m, 6 H), 6.97 (d, *J* = 7.8 Hz, 1 H), 6.77 (s, 1 H), 6.50 (d, *J* = 6.0 Hz, 1 H), 6.03 (dd, *J* = 6.6 Hz, *J* = 1.2 Hz, 1 H), 2.26 (s, 6 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 138.8, 136.9, 134.5, 130.6, 128.0, 127.3, 127.2, 127.1, 125.9, 125.1, 125.1, 123.0, 52.2, 22.7, 21.2, 18.2; **HRMS** (EI^+): calcd for $C_{19}H_{19}NO$ 277.1467, found 277.1461; **IR** (KBr): 2923, 1658, 1504, 1373 and 856 cm^{-1}.

1-(4-Methyl-2-phenylbenzo[*h*]quinolin-1(2*H*)-yl)ethan-1-one (3la)



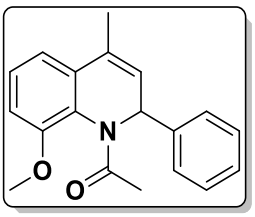
Brown oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.73 (d, *J* = 8.4 Hz, 1 H), 7.68 (d, *J* = 8.4 Hz, 2 H), 7.52 (d, *J* = 9.0 Hz, 1 H), 7.41 (t, *J* = 7.2 Hz, 1 H), 7.35 (t, *J* = 7.8 Hz, 1 H), 7.27 (d, *J* = 7.8 Hz, 2 H), 7.11 (t, *J* = 7.2 Hz, 2 H), 7.05 (t, *J* = 7.2 Hz, 1 H), 6.75 (d, *J* = 6.0 Hz, 1 H), 6.31 (d, *J* = 6.0 Hz, 1 H), 2.30 (s, 3 H), 1.82 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 172.4, 138.5, 132.9, 131.5, 130.4, 129.1, 128.0, 127.9, 127.7, 127.2, 127.0, 126.5, 126.1, 125.5, 123.4, 121.0, 52.8, 23.2, 18.4; **HRMS** (EI^+): calcd for $C_{22}H_{19}NO$ 313.1467, found 313.1463; **IR** (KBr): 2946, 1658, 1465, 1373 and 771 cm^{-1}.

1-(4-Methyl-2-phenylbenzo[*g*]quinolin-1(2*H*)-yl)ethan-1-one (3ma)



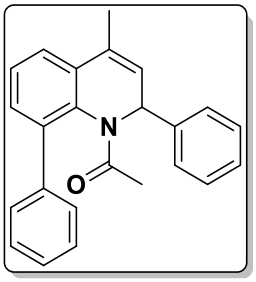
Brown oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.86–7.84 (m,1 H), 7.79 (s, 1 H), 7.68–7.67 (m, 1 H), 7.47–7.43 (m, 2 H), 7.33 (s, 1 H), 7.28–7.26 (m, 2 H), 7.20–7.14 (m, 3 H) 6.61 (d, *J* = 6.6 Hz, 1 H), 6.23 (dd, *J* = 6.0 Hz, *J* = 1.2 Hz, 1 H), 2.32 (s, 6 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 169.9, 138.7, 133.3, 132.0, 131.0, 130.7, 128.9, 128.2, 128.0, 127.7, 127.5, 127.4, 126.8, 126.3, 125.8, 122.3, 122.2, 53.0, 22.7, 18.4; **HRMS** (EI^+): calcd for $C_{22}H_{19}NO$ 313.1467 found 313.1469; **IR** (KBr): 2946, 1658, 1465, 1373 and 887 cm^{-1}.

1-(8-Methoxy-4-methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3na)



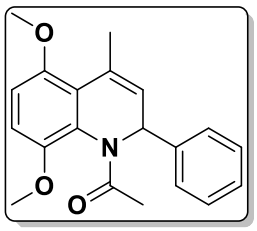
Colourless oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.23 (d, *J* = 7.8 Hz, 2 H), 7.18–7.15 (m, 2 H), 7.12–7.10 (m, 2 H), 6.92 (dd, *J* = 7.8 Hz, *J* = 1.2 Hz, 1 H), 6.76 (d, *J* = 8.4 Hz, 1 H), 6.48 (d, *J* = 6.0 Hz, 1 H), 6.25 (dd, *J* = 6.6 Hz, *J*=1.2 Hz, 1 H), 3.70 (s, 3 H), 2.15 (s, 3 H), 2.05 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 172.1, 152.5, 139.0, 131.5, 130.8, 127.8, 127.7, 126.8, 126.6, 126.1, 123.9, 115.6, 111.1, 55.5, 52.1, 21.5, 18.4; **HRMS** (EI^+): calcd for $C_{19}H_{19}NO_2$ 293.1416, found 293.1412; **IR** (KBr): 2946, 1658, 1465, 1365 and 817 cm^{-1}.

1-(4-Methyl-2,8-diphenylquinolin-1(2*H*)-yl)ethan-1-one (3oa)



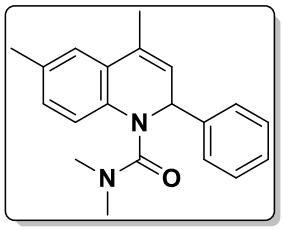
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.41–7.39 (m, 1 H), 7.33–7.28 (m, 6 H), 7.19–7.12 (m, 4 H), 6.67–6.66 (m, 1 H), 6.61 (d, *J*=6.6 Hz, 2 H), 6.10 (dd, *J* = 6.0 Hz, *J* = 1.2 Hz, 1 H), 2.26 (s, 3 H), 1.50 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.4, 139.0, 137.8, 136.2, 131.9, 131.4, 130.1, 128.8, 128.4, 128.2, 127.6, 127.1, 126.3, 122.3, 53.2, 22.1, 18.5; **HRMS** (EI^+): calcd for $C_{24}H_{21}NO$ 339.1623, found 339.1622; **IR** (KBr): 2946, 1658, 1450, 1365 and 856 cm^{-1}.

1-(5,8-Dimethoxy-4-methyl-2-phenylquinolin-1(2*H*)-yl)ethan-1-one (3pa)



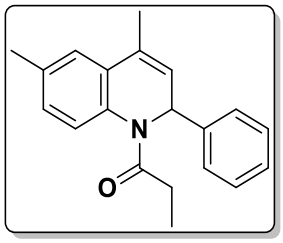
Yellow solid: m.p. 100-103 ˚C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.23–7.21 (m, 2 H), 7.13 (t, *J* = 7.2 Hz, 2 H), 7.08 (t, *J* = 7.2 Hz, 1 H), 6.65 (d, *J* = 9.0 Hz, 1 H), 6.59 (d, *J* = 9.0 Hz, 1 H), 6.39 (d, *J* = 6.6 Hz, 1 H), 6.18 (dd, *J* = 6.6 Hz, *J* = 1.2 Hz, 1 H), 3.70 (s, 3 H), 3.59 (s, 3 H), 2.30 (s, 3 H), 2.04 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 172.2, 150.4, 147.0, 138.7, 131.8, 128.2, 127.7, 126.7, 125.7, 120.3, 111.1. 108.8, 56.0, 55.5, 51.5, 22.4, 21.6; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO_3$ 323.1521, found 323.1525; **IR** (KBr): 2931, 1658, 1442, 1365 and 856 cm^{-1}.

N,*N*,4,6-Tetramethyl-2-phenylquinoline-1(2*H*)-carboxamide (3qa)



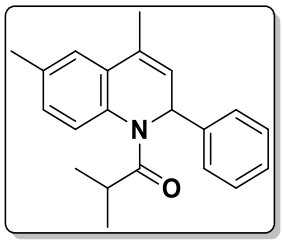
Green oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.31–7.30 (m, 2 H), 7.17–7.14 (m, 2 H), 7.12–7.10 (m, 1 H), 7.03 (d, *J* = 1.8 Hz, 1 H), 6.93 (dd, *J* = 7.8 Hz, *J* = 1.2 Hz, 1 H), 6.79 (d, *J* = 8.4 Hz, 1 H), 6.01 (dd, *J* = 6.0 Hz, *J* = 1.2 Hz, 1 H), 5.52 (d, *J* = 6.6 Hz, 1 H), 2.69 (s, 6 H), 2.28 (s, 3 H), 2.10 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 160.0, 141.4, 135.4, 131.6, 130.1, 128.7, 128.0, 127.4, 127.3, 127.0, 126.3, 124.3, 120.5, 57.1, 38.1, 20.9, 18.3; **HRMS** (EI^+): calcd for $C_{20}H_{22}N_2O$ 306.1732, found 306.1728; **IR** (KBr): 2923, 1650, 1488, 1380 and 833 cm^{-1}.

1-(4,6-Dimethyl-2-phenylquinolin-1(2*H*)-yl)propan-1-one (3ra)



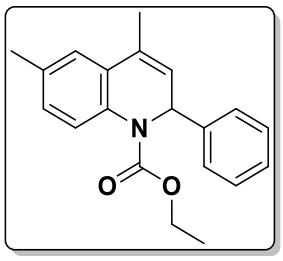
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.23–7.14 (m, 6 H), 6.95 (d, *J* = 7.2 Hz, 1 H), 6.86 (d, *J* = 8.4 Hz, 1 H), 6.52 (d, *J* = 6.0 Hz, 1 H), 6.10 (d, *J* = 5.4 Hz, 1 H), 2.64–2.59 (m, 1 H), 2.47–2.42 (m, 1 H), 2.33 (s, 3 H), 2.17 (s, 3 H), 1.17 (t, *J*=7.2 Hz, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 173.8, 138.8, 134.8, 131.8, 130.7, 129.7, 128.0, 127.5, 127.4, 127.2, 126.5, 124.3, 124.0, 52.4, 27.4, 21.0, 18.2, 10.2; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO$ 291.1623 ,found 291.1624; **IR** (KBr): 2938, 1658, 1496, 1380 and 848 cm^{-1}.

1-(4,6-Dimethyl-2-phenylquinolin-1(2*H*)-yl)-2-methylpropan-1-one (3sa)



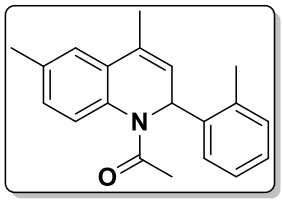
Brown solid: m.p. 87-90 °C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.23–7.15 (m, 6 H), 6.95 (d, *J* = 7.8 Hz, 1 H), 6.84 (d, *J* = 7.8 Hz, 1 H), 6.44 (d, *J* = 6.0 Hz, 1 H), 6.09 (d, *J* = 6.6 Hz, 1 H), 3.23–3.19 (m, 1 H), 2.34 (s, 3 H), 2.18 (s, 3 H), 1.29 (d, *J* = 6.6 Hz, 3 H), 0.98 (d, *J* = 6.6 Hz, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 177.8, 138.7, 134.8, 131.9, 130.7, 129.8, 128.0, 127.5, 127.4, 127.2, 126.5, 124.0, 124.0, 52.6, 30.3, 21.1, 20.5, 19.7, 18.2; **HRMS** (EI^+): calcd for $C_{21}H_{23}NO$ 305.1780, found 305.1782; **IR** (KBr): 2969, 1658, 1488, 1388 and 825 cm^{-1}.

Ethyl 4,6-dimethyl-2-phenylquinoline-1(2*H*)-carboxylate (3ta)



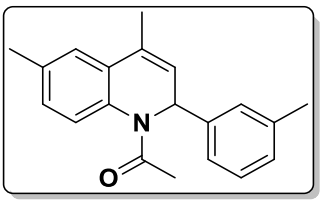
Brown oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.7.23–7.15 (m, 6 H), 7.07 (s, 1 H), 6.99 (d, *J* = 8.4 Hz, 1 H), 6.08 (s, 1 H), 6.01 (d, *J* = 6.0 Hz, 1 H), 4.35–4.20 (m, 2 H), 2.31 (s, 3 H), 2.13 (s, 3 H), 1.31 (t, *J*=7.2 Hz, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 154.6, 140.1, 133.3, 132.1, 130.5, 128.6, 128.2, 128.0, 127.4, 127.0, 124.8, 124.4, 123.7, 62.1, 55.0, 21.0, 18.5, 14.4; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO_2$ 307.1572, found 307.1571; **IR** (KBr): 2946, 1697, 1589, 1496 and 825 cm^{-1}.

1-(4,6-Dimethyl-2-(*o*-tolyl)quinolin-1(2*H*)-yl)ethan-1-one (3ab)



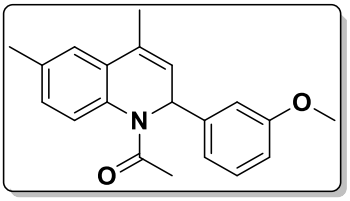
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.15–7.12 (m, 2 H), 7.07 (t, *J* = 7.2 Hz, 1 H), 7.00 (dd, *J* = 7.8 Hz, *J* = 1.2Hz, 1 H), 6.92–6.88 (m, 2 H), 6.85 (d, *J* = 7.2 Hz, 1 H), 6.64 (d, *J* = 6.0 Hz, 1 H), 6.02 (d, *J* = 6.0 Hz, 1 H), 2.57 (s, 3 H), 2.37 (s, 3 H), 2.19 (s, 3 H), 2.13 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.2, 136.6, 135.8, 135.1, 133.0, 130.5, 130.0, 129.9, 127.7, 127.5, 127.3, 127.1, 125.5, 124.8, 123.9, 50.3, 22.8, 21.1, 19.9, 18.2; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO$ 291.1623, found 291.1626; **IR** (KBr): 2923, 1658, 1488, 1373 and 817 cm^{-1}.

1-(4,6-Dimethyl-2-(*m*-tolyl)quinolin-1(2*H*)-yl)ethan-1-one (3ac)



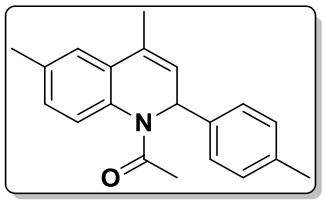
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.12 (s, 1 H), 7.06 (t, *J* = 7.8 Hz, 2 H), 6.97–6.93 (m, 3 H), 6.86 (d, *J* = 7.8 Hz, 1 H), 6.46 (d, *J* = 6.0 Hz, 1 H), 6.08 (d, *J* = 6.0 Hz, 1 H), 2.33 (s, 3 H), 2.24 (s, 6 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 138.8, 137.7, 134.8, 132.3, 130.5, 129.6, 128.1, 128.1, 127.9, 127.6, 126.5, 124.4, 124.1, 123.9, 52.3, 22.7, 21.3, 21.1, 18.3; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO$ 291.1623, found 291.1620; **IR** (KBr): 2923, 1658, 1496, 1373 and 840 cm^{-1}.

1-(2-(3-Methoxyphenyl)-4,6-dimethylquinolin-1(2*H*)-yl)ethan-1-one (3ad)



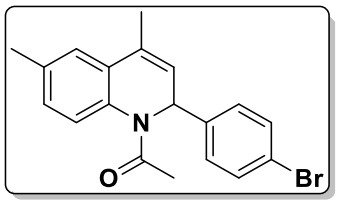
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.11-7.08 (m, 2H), 6.94 (d, *J* = 7.8 Hz, 1H), 6.85 (d, *J* = 7.8 Hz,1H), 6.79-6.78 (m, 1H), 6.74 (s, 1H), 6.68-6.66 (m, 1H), 6.44 (d, *J* = 6.6 Hz, 1H), 6.06-6.05 (m, 1H), 3.68 (s, 3H), 2.31 (s, 3H), 2.22 (s, 3H), 2.14(s, 3H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.2, 159.0, 140.5, 134.9, 132.2, 130.8, 129.5, 129.1, 127.6, 126.1, 124.4, 124.0, 119.6, 113.1, 112.1, 54.9, 52.1, 22.7, 21.1, 18.3; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO_2$ 307.1572 found 307.1576; **IR** (KBr): 2915, 2360, 1658, 1041 and 794 cm^{-1}.

1-(4,6-Dimethyl-2-(*p*-tolyl)quinolin-1(2*H*)-yl)ethan-1-one (3ae)



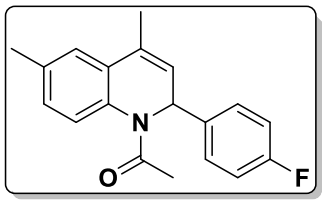
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.12 (s, 1 H), 7.09 (d, *J* = 7.8 Hz, 2 H), 6.99 (t, *J* = 8.4 Hz, 2 H), 6.95 (d, *J* = 7.8 Hz, 1 H), 6.84 (d, *J* = 7.8 Hz, 1 H),6.46 (d, *J* = 6.0 Hz, 1 H), 6.07 (d, *J* = 6.0 Hz, 1 H), 2.32 (s, 3 H), 2.22 (s, 3 H), 2.22 (s, 3 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 169.9, 136.9, 135.8, 134.8, 132.4, 130.5, 129.7, 128.8, 127.5, 127.3, 126.7, 124.4, 123.9, 52.2, 22.6, 21.0, 20.9, 18.2; **HRMS** (EI^+): calcd for $C_{20}H_{21}NO$ 291.1623, found 291.1620; **IR** (KBr): 2923, 1658, 1493, 1380 and 809 cm^{-1}.

1-(2-(4-Bromophenyl)-4,6-dimethylquinolin-1(2*H*)-yl)ethan-1-one (3af)



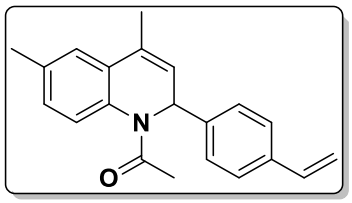
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.26 (d, *J* = 8.4 Hz, 2 H), 7.11 (s, 1 H), 7.06–7.05 (m, 2 H), 6.94 (d, *J* = 7.2 Hz, 1 H), 6.82 (d, *J* = 8.4 Hz, 1 H), 6.42 (d, *J* = 6.0 Hz, 1 H), 6.03 (dd, *J* = 6.6 Hz, *J* = 1.2 Hz, 1 H), 2.30 (s, 3 H), 2.21 (s, 3 H), 2.15 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 137.9, 135.1, 132.0, 131.3, 131.1, 129.5, 129.2, 127.8, 125.6, 124.3, 124.1, 121.2, 51.6, 22.5, 21.0, 18.2; **HRMS** (EI^+): calcd for $C_{19}H_{18}BrNO$ 355.0572, found 355.0568; **IR** (KBr): 2946, 1658, 1488, 1373 and 694 cm^{-1}.

1-(2-(4-Fluorophenyl)-4,6-dimethylquinolin-1(2*H*)-yl)ethan-1-one (3ag)



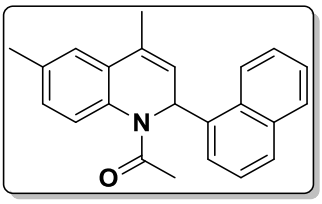
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.16-7.14 (m, 2 H), 7.11 (s, 1 H), 6.94 (d, *J* = 8.4 Hz, 1 H), 6.85-6.81 (m, 3 H), 6.44(d, *J* = 6.0 Hz, 1 H), 6.03 (d, *J* = 5.4 Hz, 1 H), 2.32 (s, 3 H), 2.21 (s, 3 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 161.9 (d, *J* = 245 Hz), 135.0, 134.5, 132.0, 131.0, 129.5, 129.2 (d, *J* = 9 Hz), 127.8, 126.1, 124.4, 124.0, 115.0 (d, *J* = 21 Hz), 51.6, 22.7, 21.1, 18.3; **HRMS** (EI^+): calcd for $C_{19}H_{18}FNO$ 295.1372, found 295.1368; **IR** (KBr): 3039, 1360, 1658, 817 and 671 cm^{-1}.

1-(4,6-Dimethyl-2-(4-vinylphenyl)quinolin-1(2*H*)-yl)ethan-1-one (3ah)



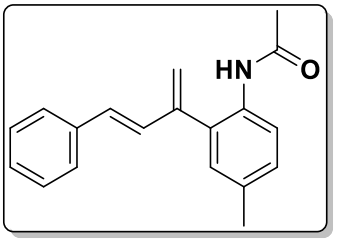
Yellow oil: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.22 (d, J = 7.8 Hz, 2 H), 7.15 (d, *J* = 7.8 Hz, 2 H), 7.12 (s, 1 H), 6.95 (d, *J* = 7.8 Hz, 1 H), 6.84 (d, *J* = 7.8 Hz, 1 H),6.60-6.55 (m, 1 H), 6.47 (d, *J* = 6.0 Hz, 1 H), 6.07 (d, *J* = 6.0 Hz, 1 H), 5.64 (d, *J* = 17.4 Hz, 1 H), 5.15 (d, *J* = 10.8 Hz, 1 H), 2.32 (s, 3 H), 2.23 (s, 3 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.1, 138.3, 136.2, 136.0, 134.9, 132.1, 130.8, 129.5, 127.6, 127.5, 126.1, 125.9, 124.3, 124.0, 113.7, 51.9, 22.7, 21.1, 18.3; **HRMS** (EI^+): calcd for $C_{21}H_{21}NO$ 303.1623, found 303.1617; **IR** (KBr): 2915, 1658, 1496, 1018 and 794 cm^{-1}.

1-(4,6-Dimethyl-2-(naphthalen-1-yl)quinolin-1(2*H*)-yl)ethan-1-one (3ai)



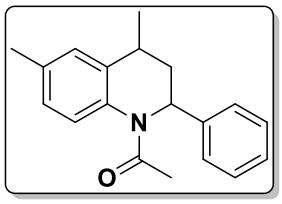
Brown solid: m.p. 167-170 ˚C; **^{1}H NMR** (600 MHz, $CDCl_3$): δ 8.77 (d, *J* = 8.4 Hz, 1 H), 7.79 (d, *J* = 7.8 Hz, 1 H), 7.67–7.60 (m, 2 H), 7.48 (t, *J* = 7.2 Hz, 1 H), 7.29 (d, *J* = 5.4 Hz, 1 H), 7.18–7.09 (m, 3 H), 6.88 (d, *J* = 7.2 Hz, 1 H), 6.74 (d, *J* = 7.8 Hz, 1 H), 6.15 (d, *J* = 4.8 Hz, 1 H), 2.34 (s, 3 H), 2.22 (s, 3 H), 2.16 (s, 3 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.3, 135.5, 133.7, 133.1, 132.9, 131.6, 131.1 130.6, 128.4, 128.4, 127.6, 127.5, 126.4, 125.9, 125.5, 125.4, 124.7, 124.4, 123.8, 49.5, 22.8, 21.1, 18.2; **HRMS** (EI^+): calcd for $C_{23}H_{21}NO$ 327.1623, found 327.1621; **IR** (KBr): 2946, 1658, 1488, 1373 and 894 cm^{-1}.

(*E*)-N-(4-Methyl-2-(4-phenylbuta-1,3-dien-2-yl)phenyl)acetamide (5aa)



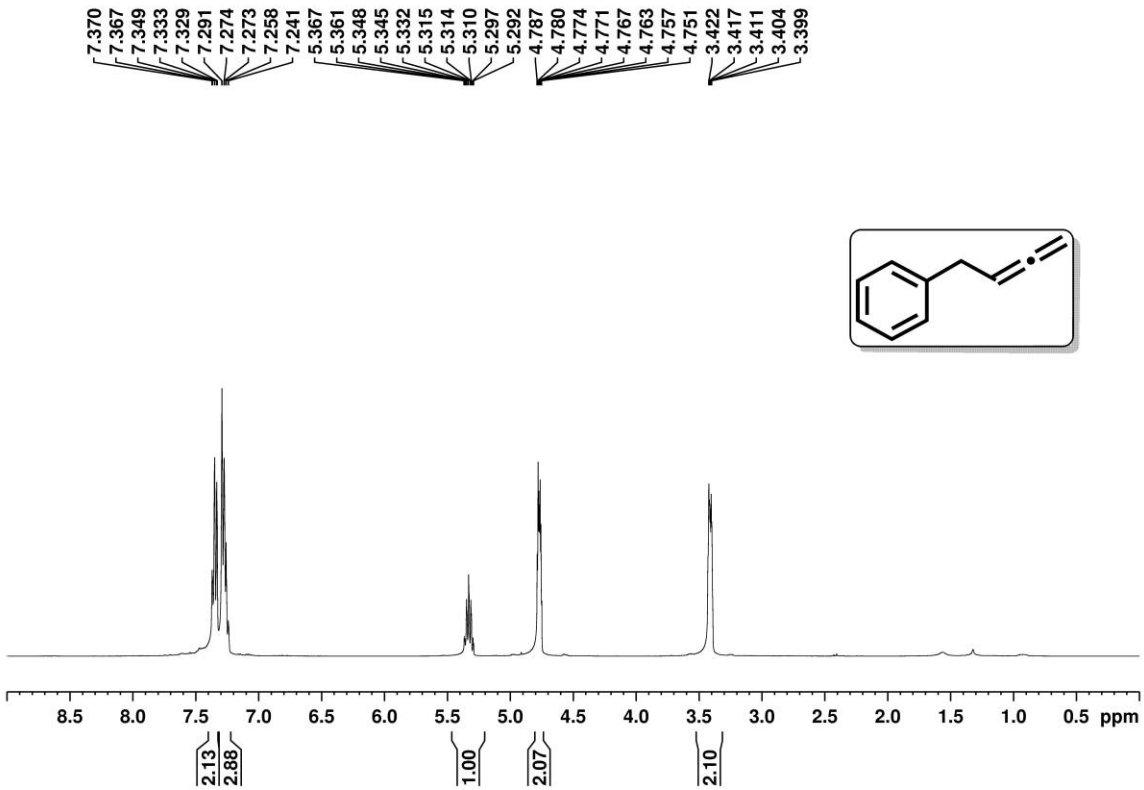
Yellow oil: **^{1}H NMR** (400 MHz, $CDCl_3$): δ 8.12 (d, *J* = 8.4 Hz, 1 H), 7.36 (d, *J* = 7.6 Hz, 2 H), 7.30 (t, *J* = 7.2 Hz, 2 H), 7.25–7.14 (m, 3 H), 7.03 (d, *J* = 16.0 Hz, 2 H), 6.18 (d, *J* = 16.0 Hz, 1 H), 5.61 (s, 1 H), 5.23 (s, 1 H), 2.34 (s, 3 H), 2.02 (s, 3 H); **^{13}C NMR** (100 MHz, $CDCl_3$): δ 168.1, 145.0, 136.4, 133.6, 132.9, 132.4, 130.2, 129.7, 129.2, 129.1, 128.6, 128.0, 126.7, 121.2, 120.4, 24.6, 20.8.

1-(4,6-Dimethyl-2-phenyl-3,4-dihydroquinolin-1(2*H*)-yl)ethan-1-one (6aa)

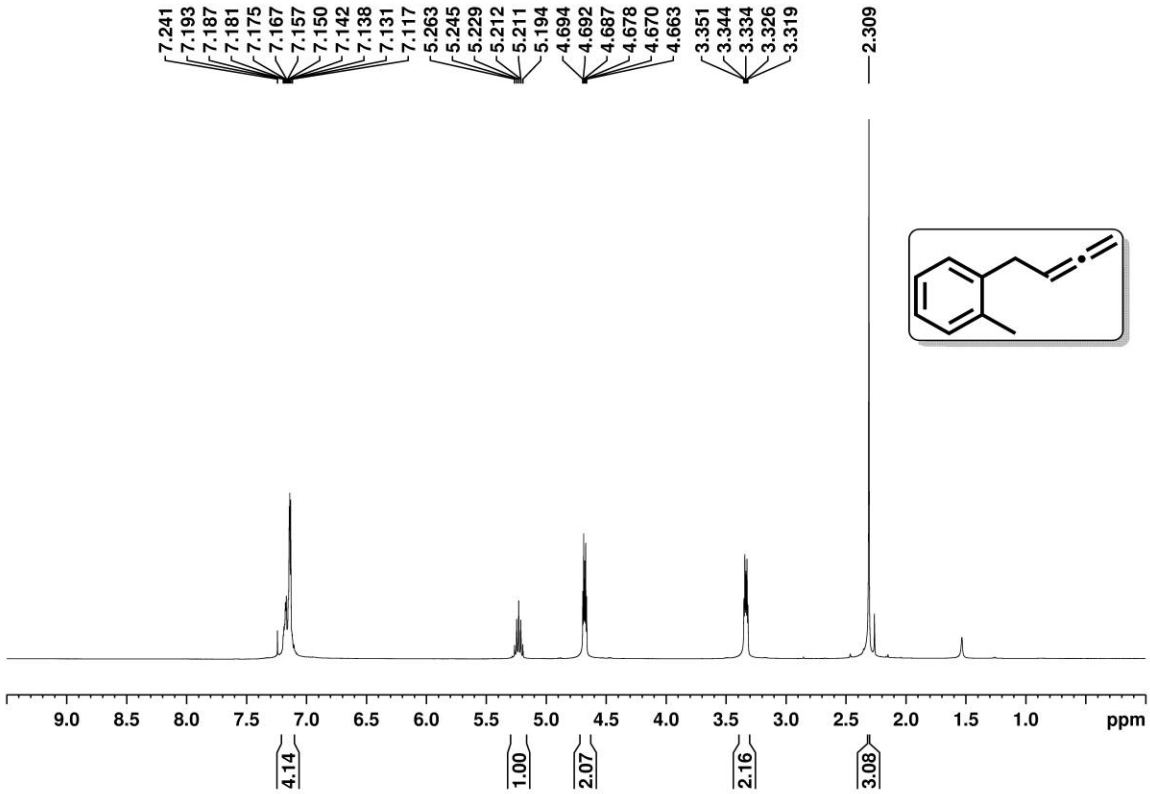


white solid: **^{1}H NMR** (600 MHz, $CDCl_3$): δ 7.25-7.22 (m, 2 H), 7.17-7.15 (m, 3 H), 7.11-7.07 (m, 3 H), 5.67 (t, *J* = 9.6 Hz, 1 H), 2.68-2.65 (m, 1 H), 2.85-2.54 (m, 1 H), 2.39 (s, 3 H), 2.13 (s, 3 H), 1.35-1.34 (m, 4 H); **^{13}C NMR** (150 MHz, $CDCl_3$): δ 170.0, 143.6, 140.3, 135.5, 135.5, 128.2, 126.8, 126.6, 126.1, 125.2, 124.0, 56.6, 43.5, 30.1, 22.9, 21.2, 16.4; **HRMS** (EI^+): calcd for $C_{19}H_{21}NO$ 279.1623, found 279.1620; **IR** (KBr): 3432, 2360, 1658, 1018 and 709 cm^{-1}.

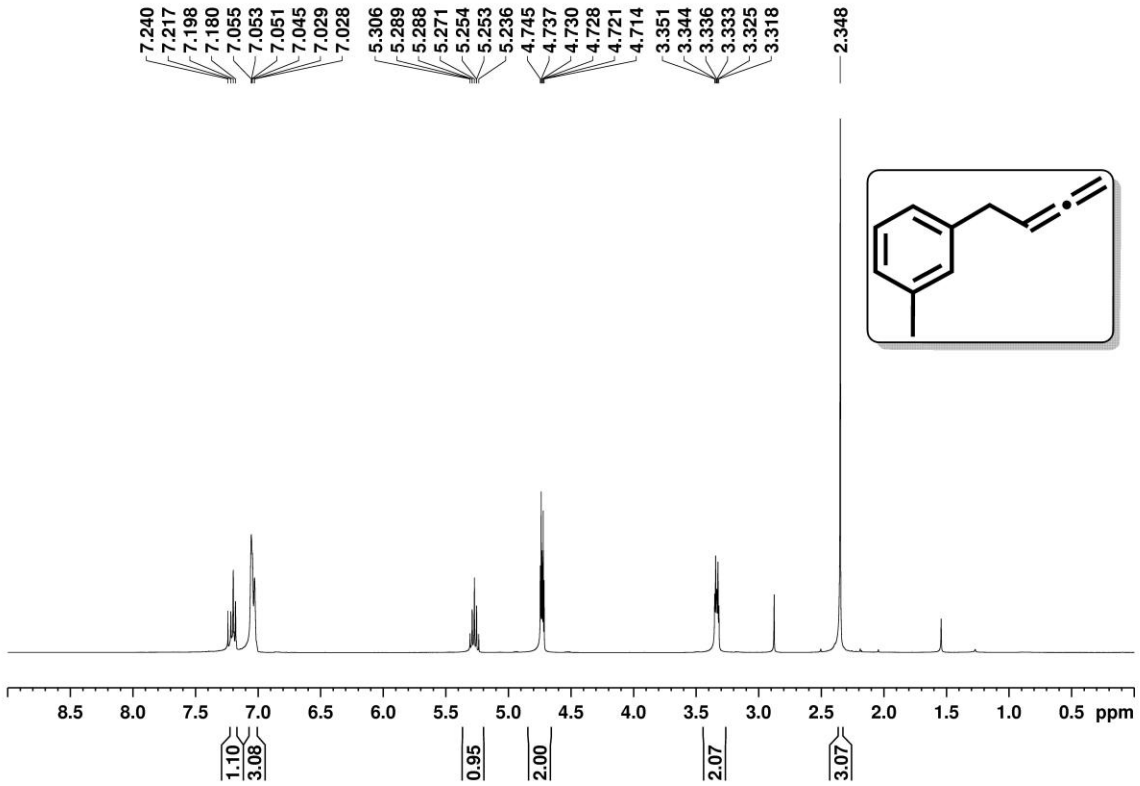
^{1}H NMR spectra of compound **2a**



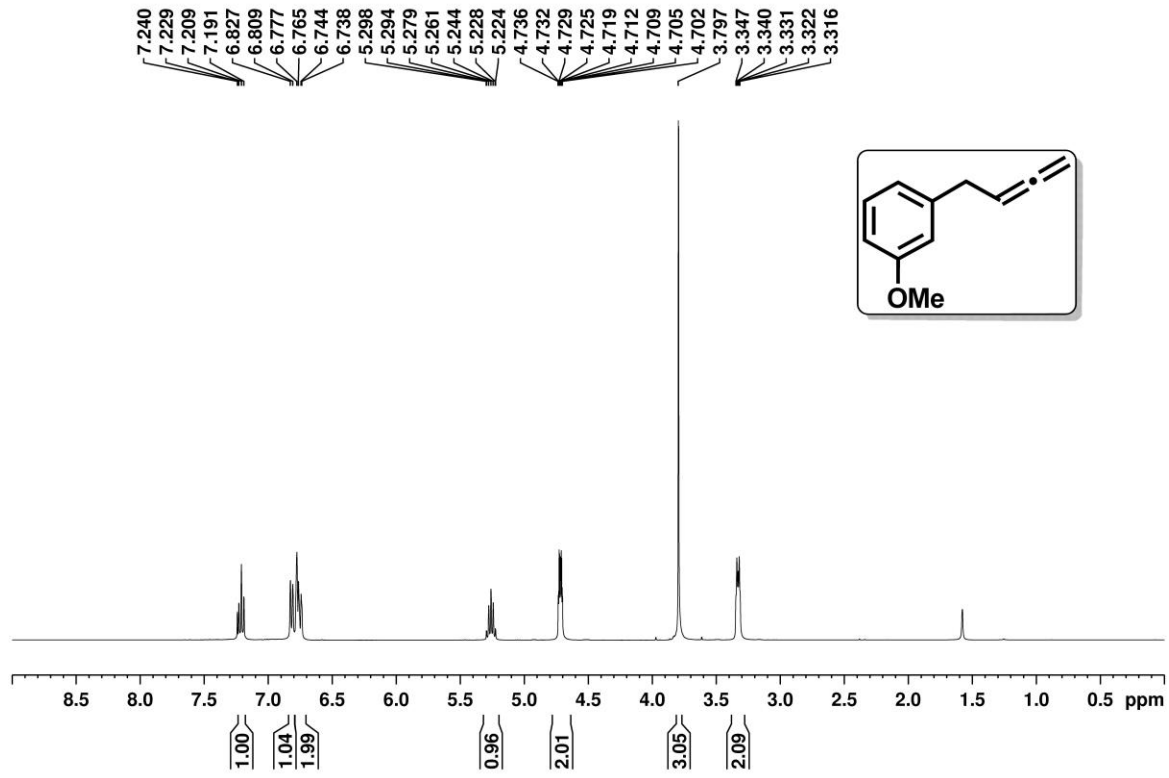
^{1}H NMR spectra of compound **2b**



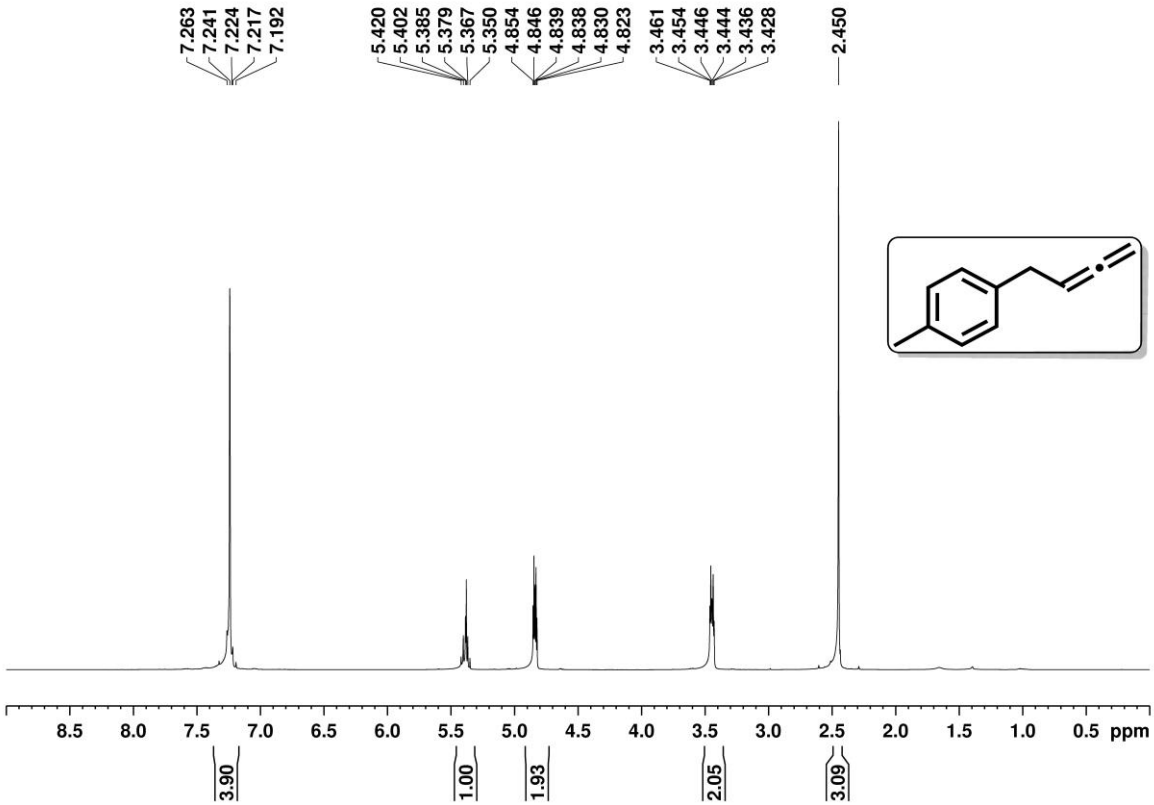
^{1}H NMR spectra of compound **2c**



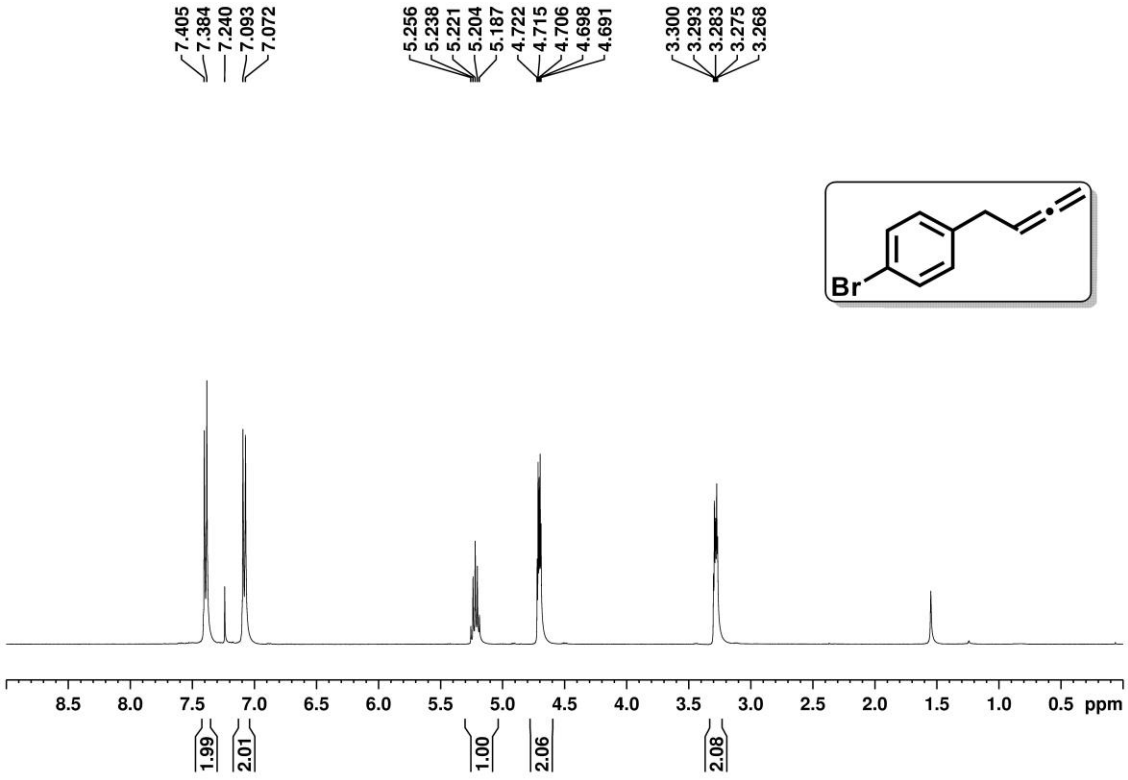
^{1}H NMR spectra of compound **2d**



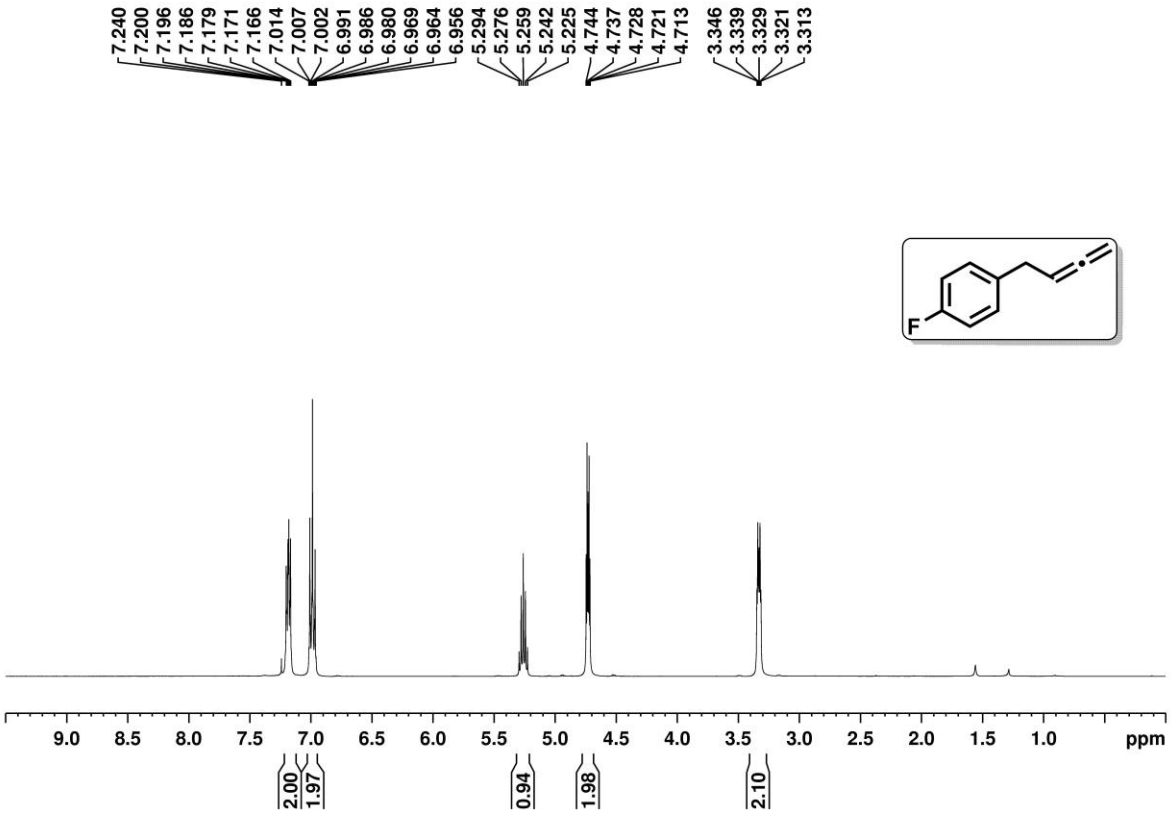
^{1}H NMR spectra of compound **2e**



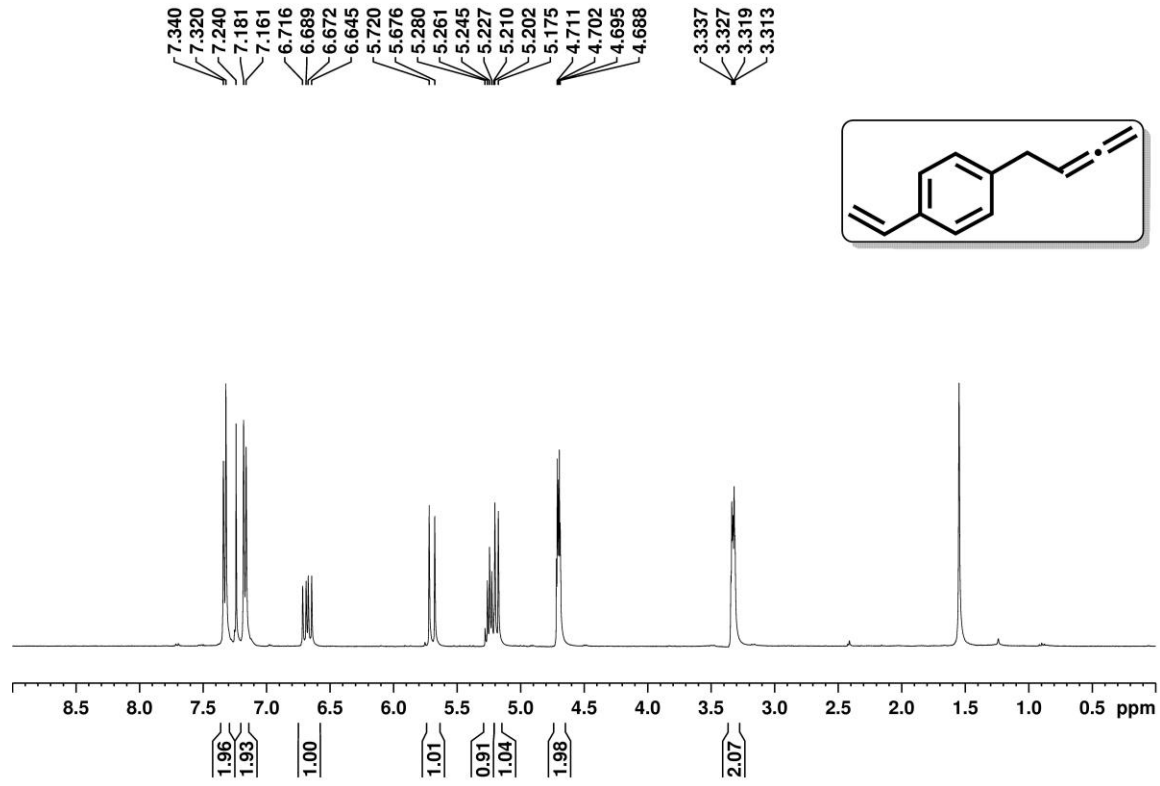
^{1}H NMR spectra of compound **2f**



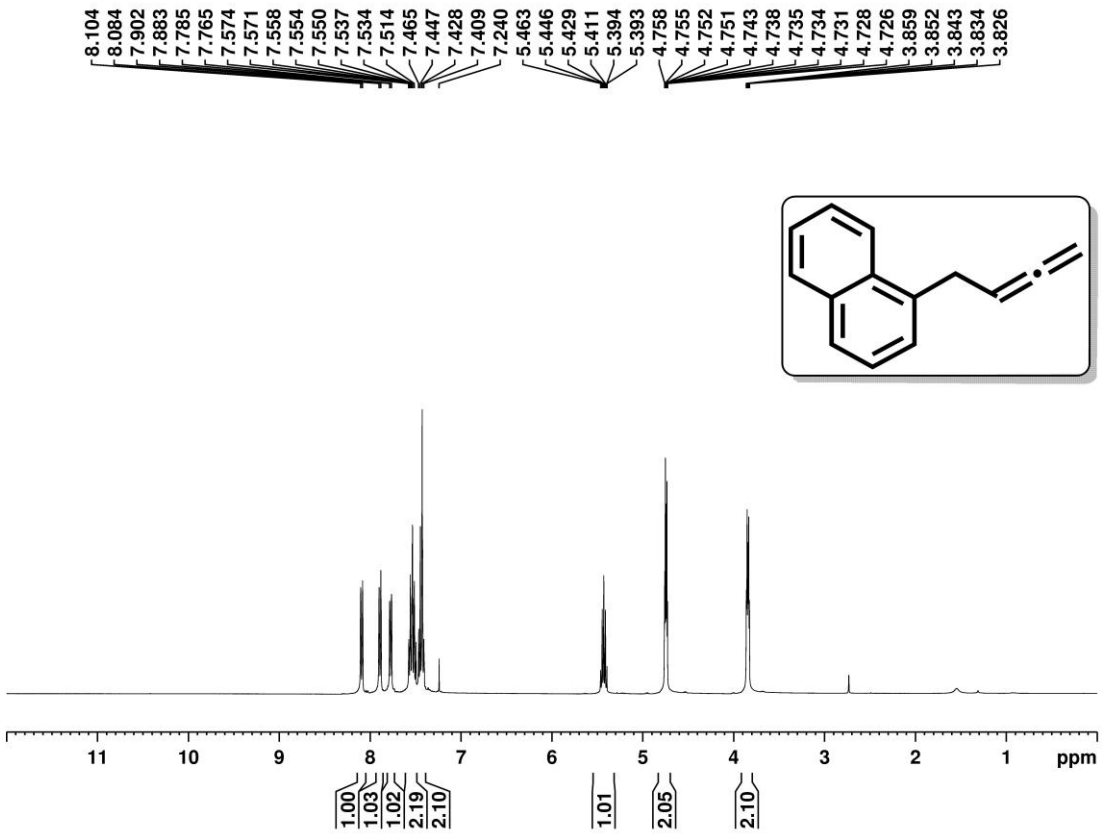
^{1}H NMR spectra of compound **2g**



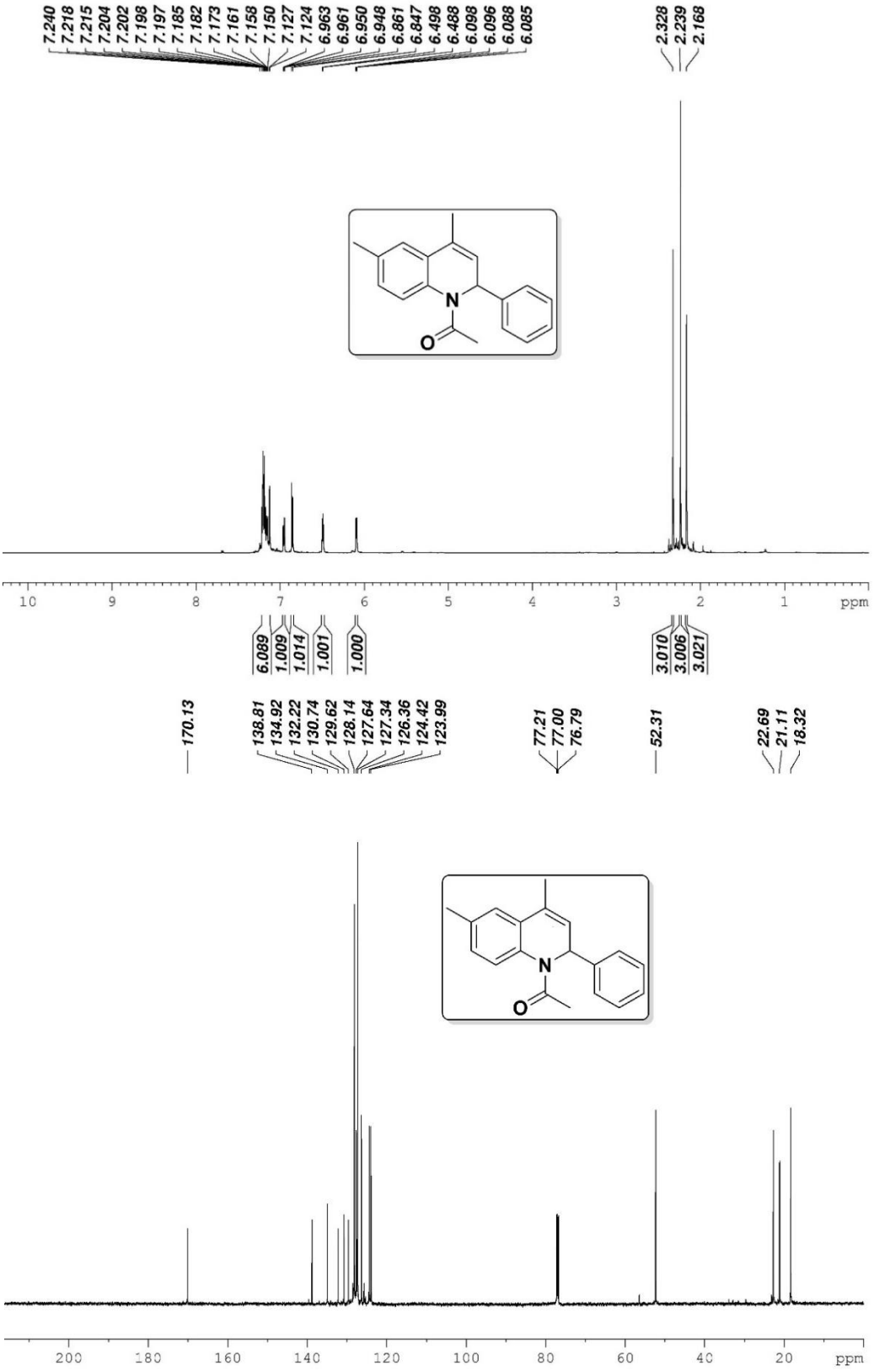
^{1}H NMR spectra of compound **2h**



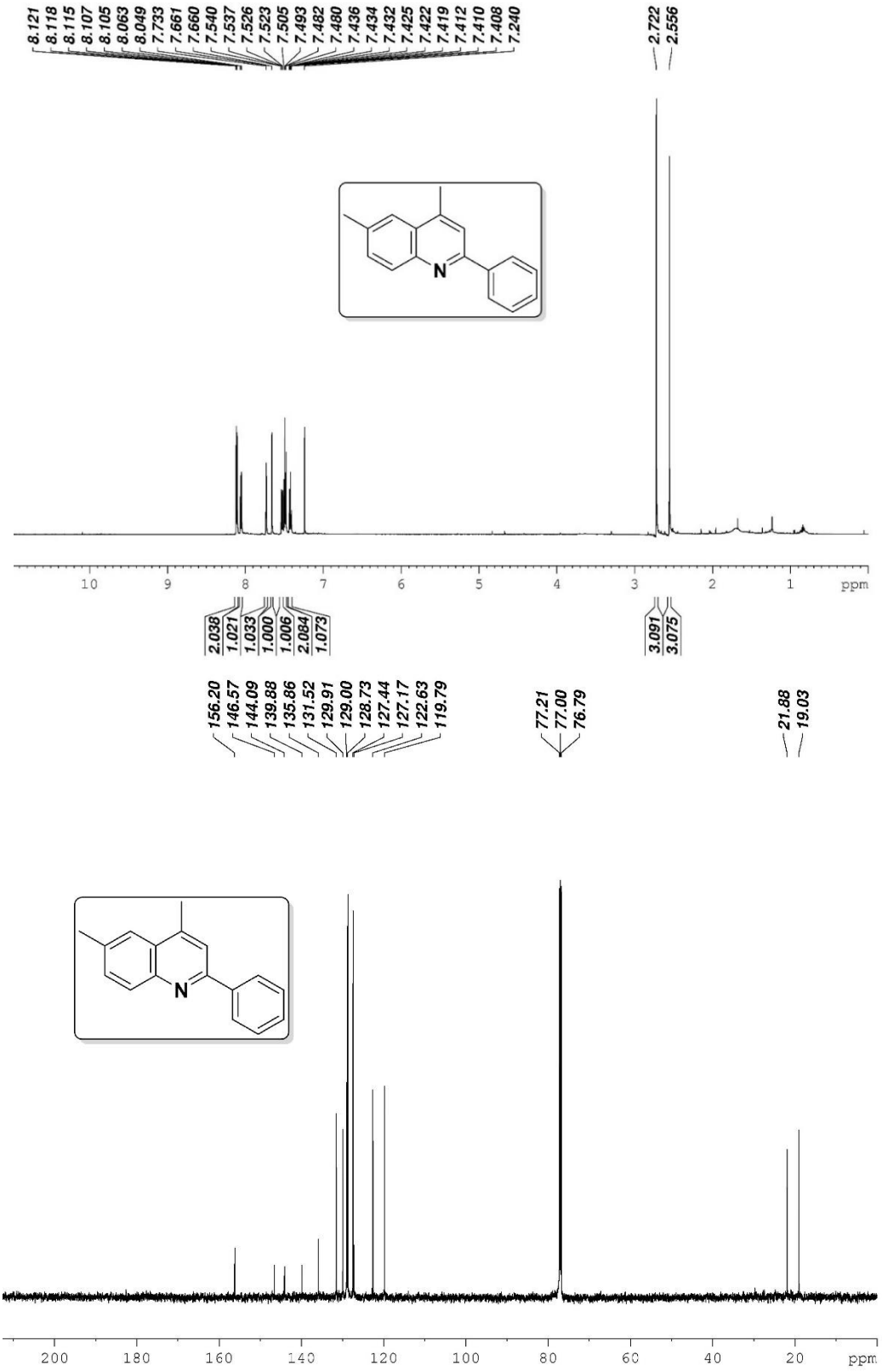
^{1}H NMR spectra of compound **2i**



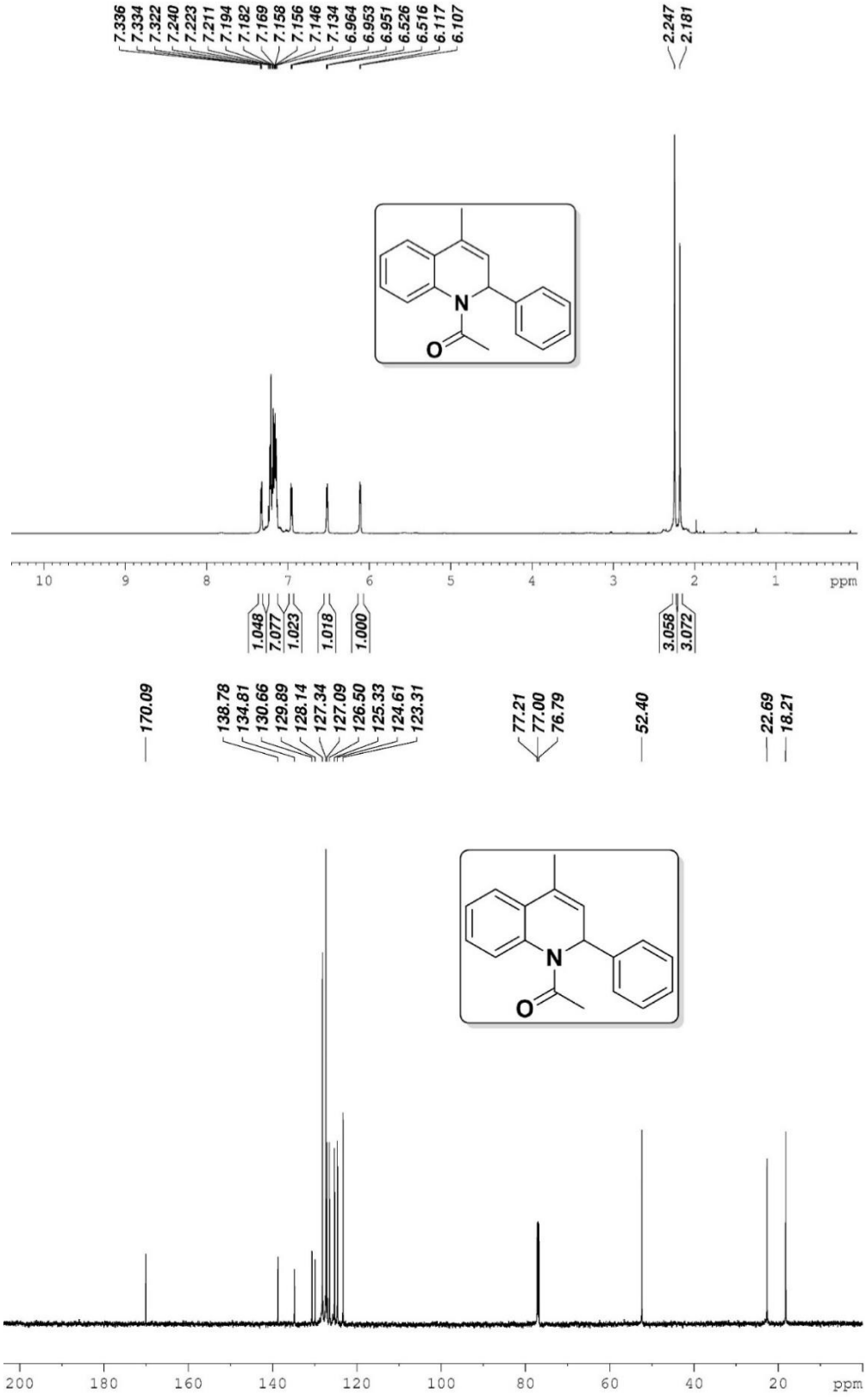
^{1}H and ^{13}C NMR spectra of compound **3aa**



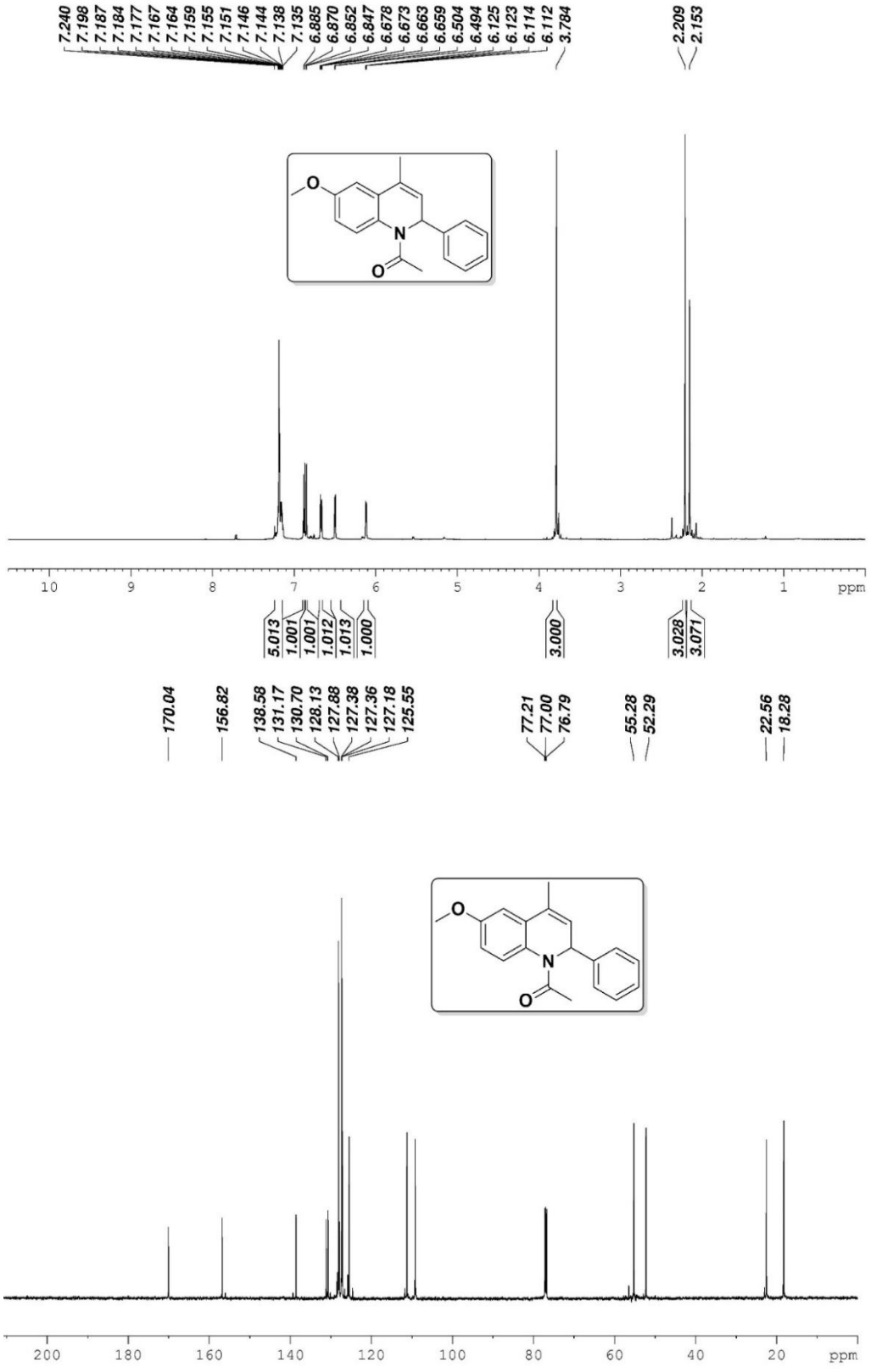
^{1}H and ^{13}C NMR spectra of compound **4aa**



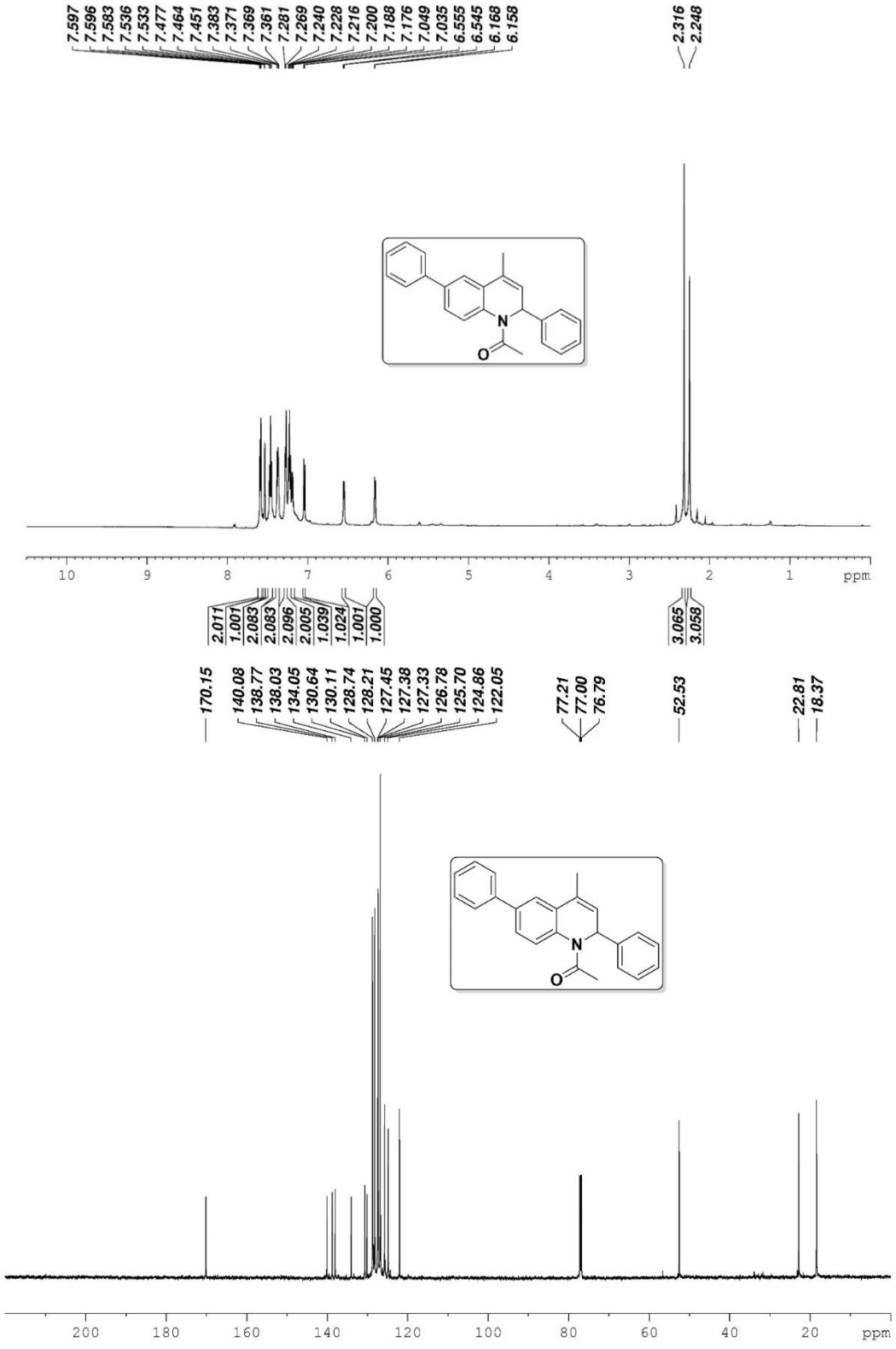
^{1}H and ^{13}C NMR spectra of compound **3ba**



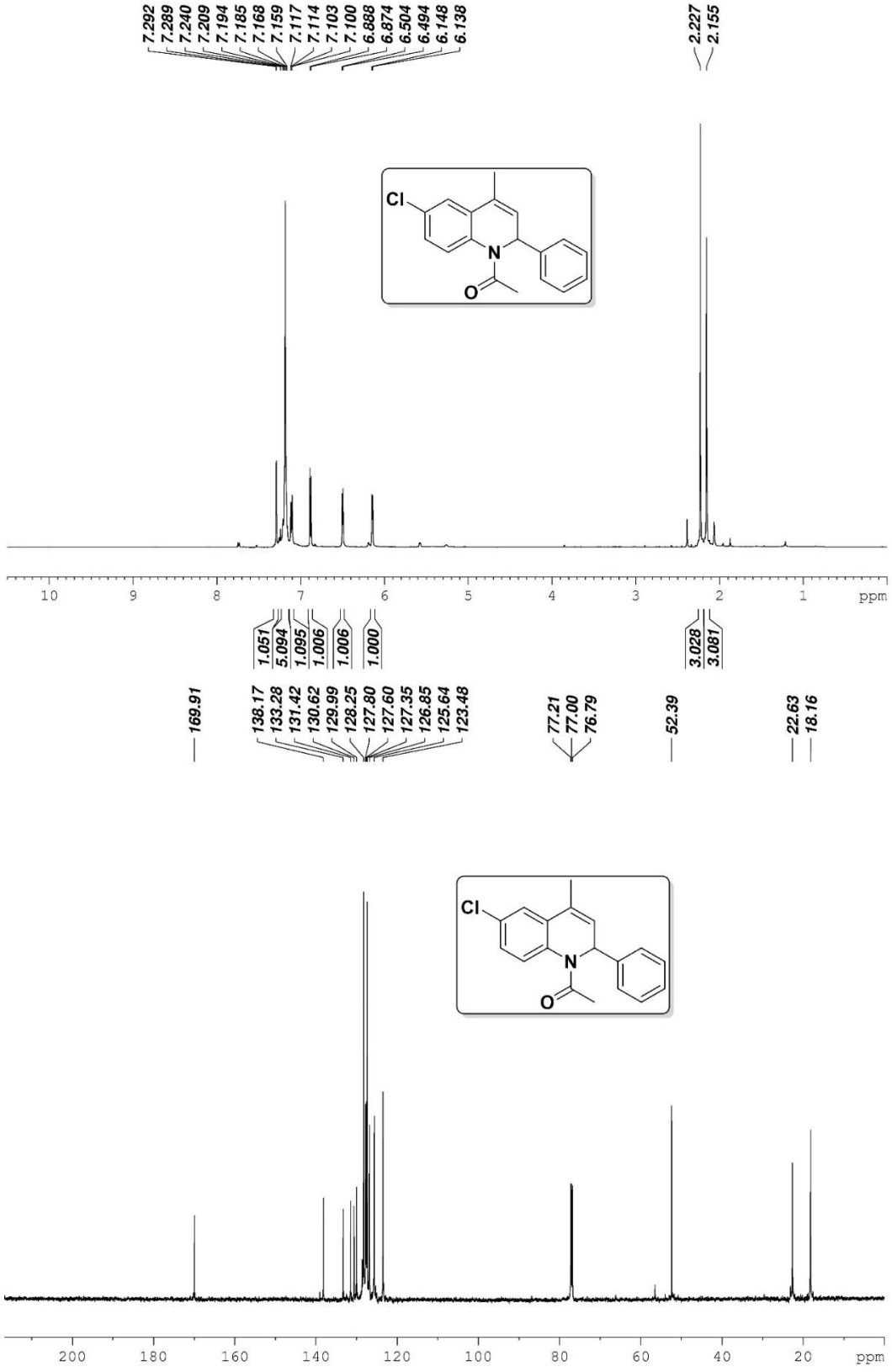
^{1}H and ^{13}C NMR spectra of compound **3ca**



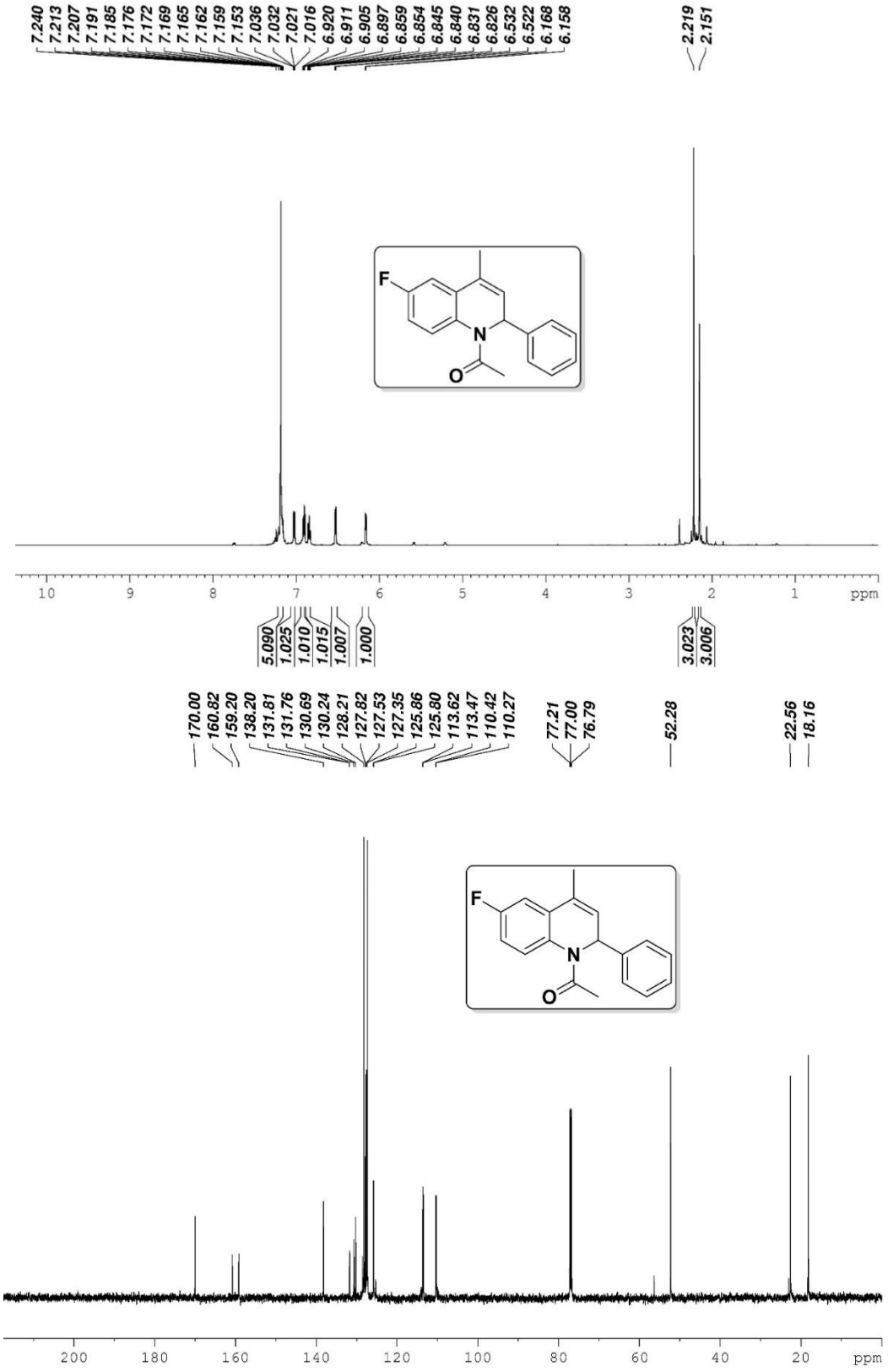
^{1}H and ^{13}C NMR spectra of compound **3da**



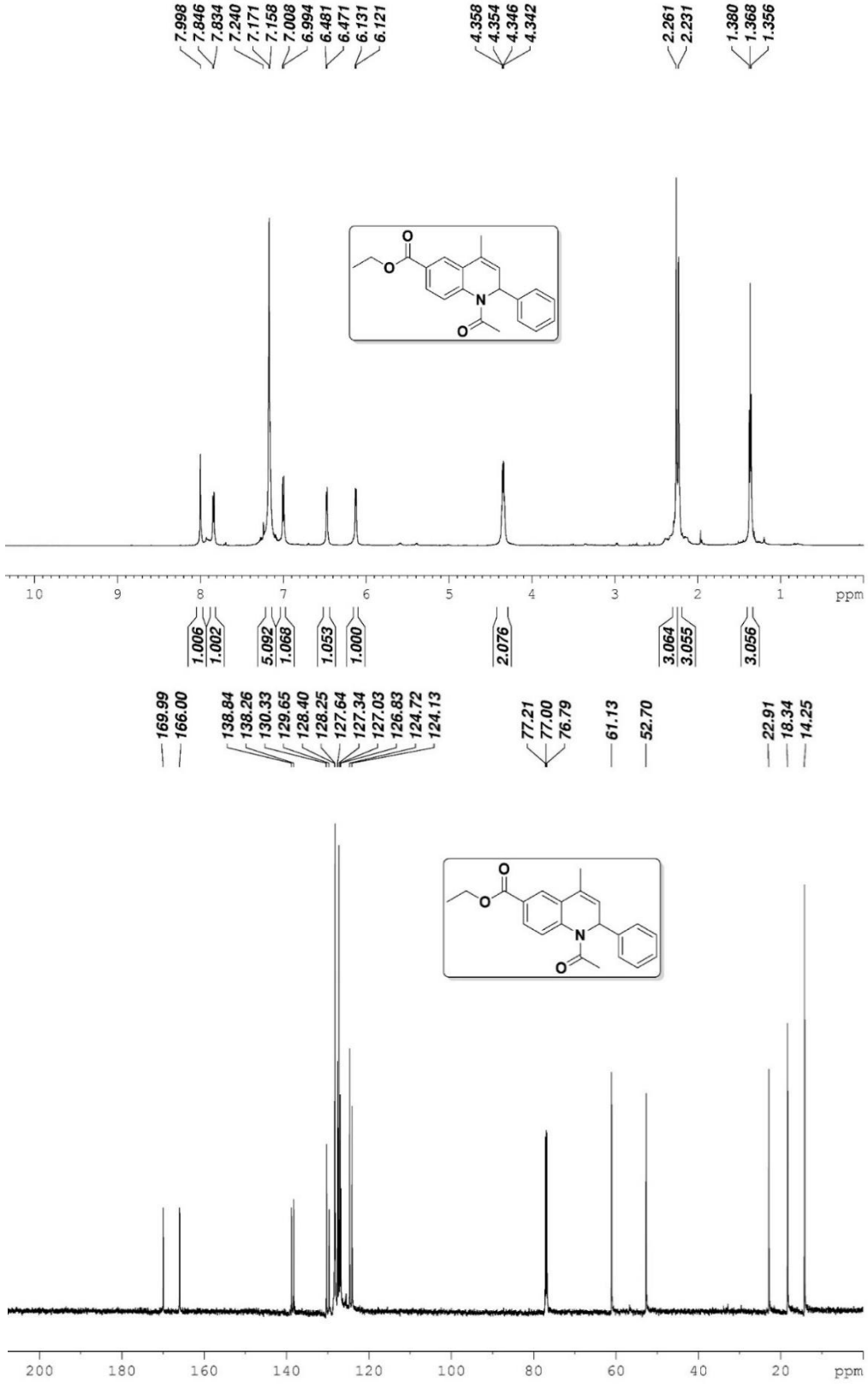
^{1}H and ^{13}C NMR spectra of compound **3ea**



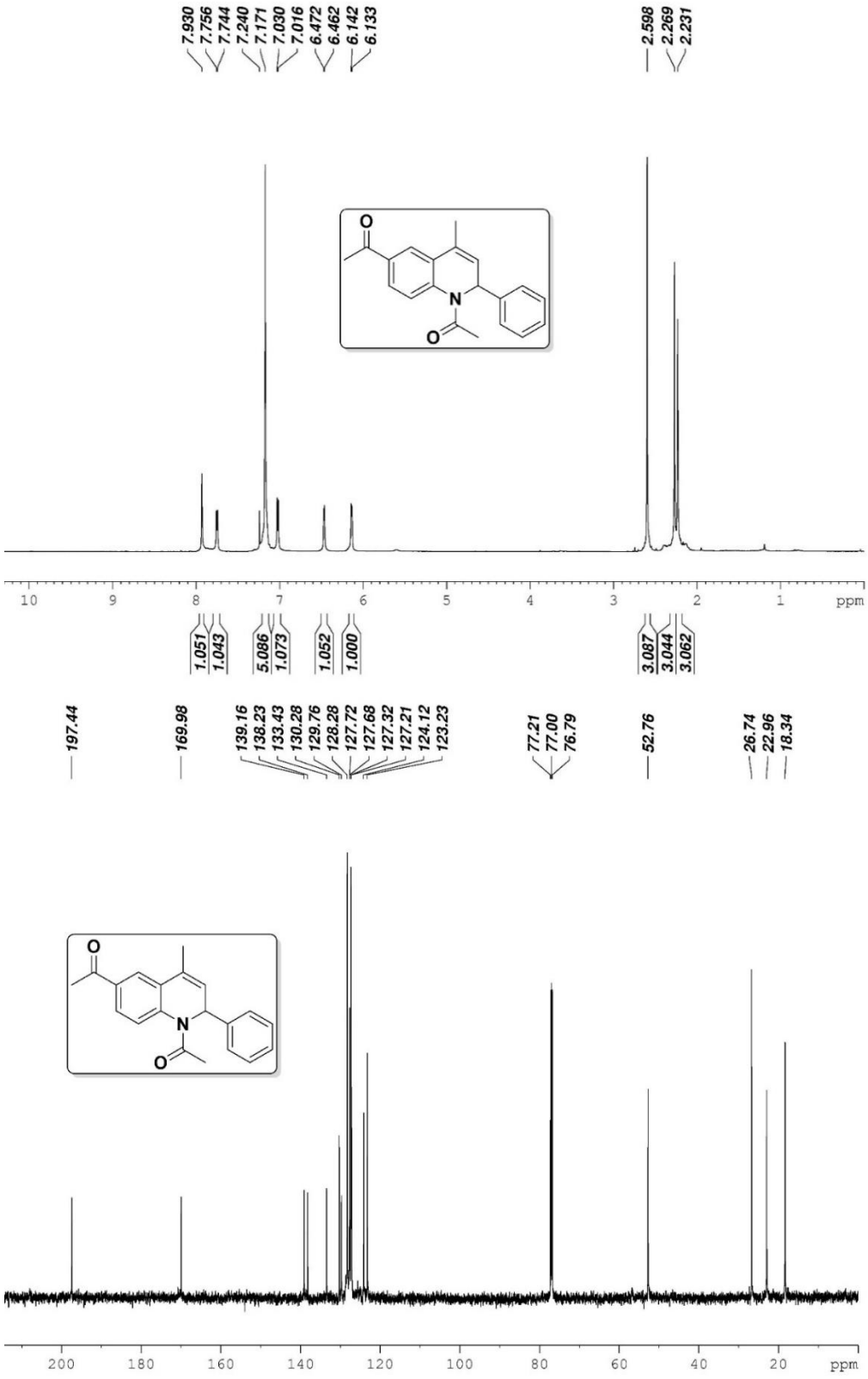
^{1}H and ^{13}C NMR spectra of compound **3fa**



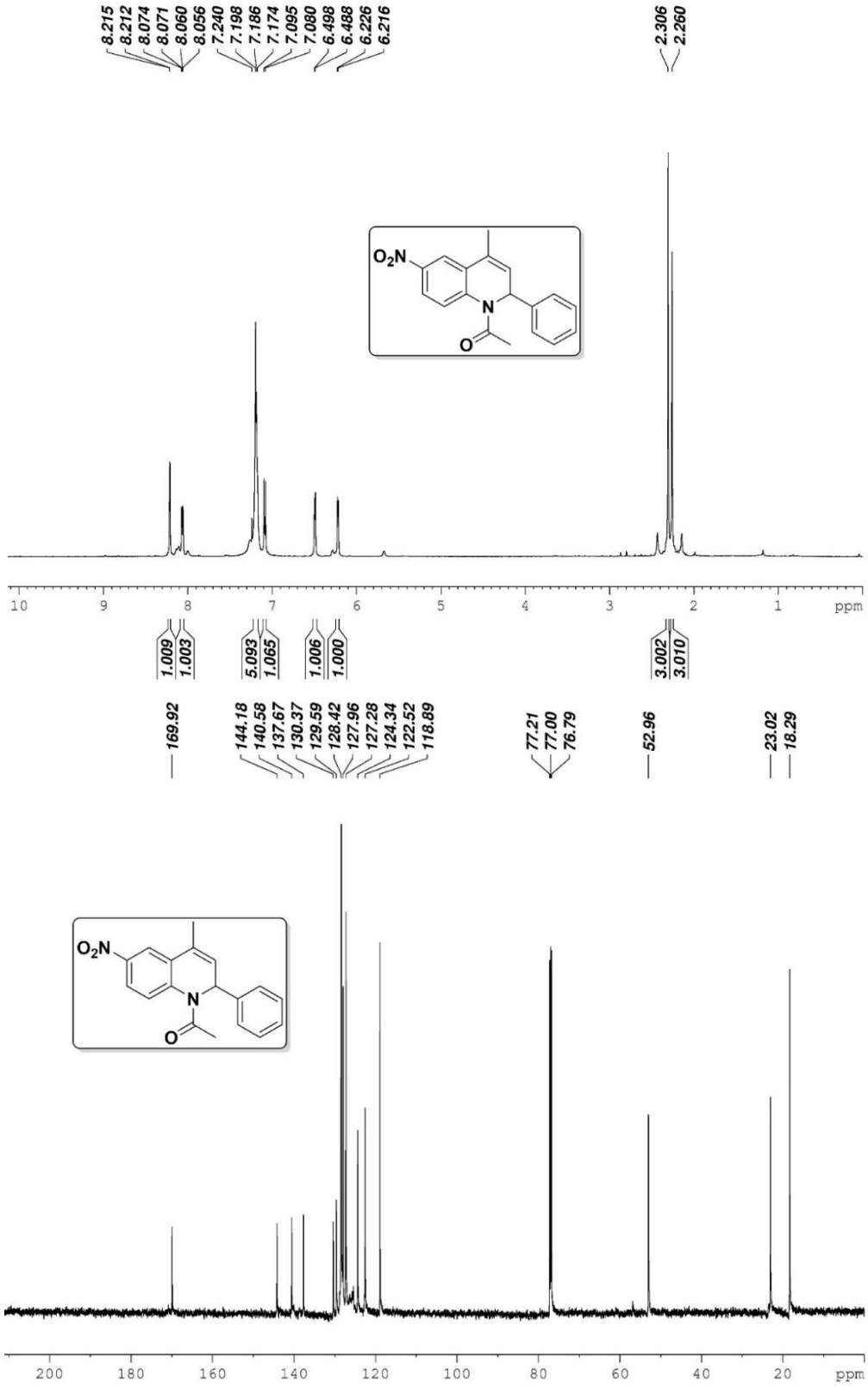
^{1}H and ^{13}C NMR spectra of compound **3ga**



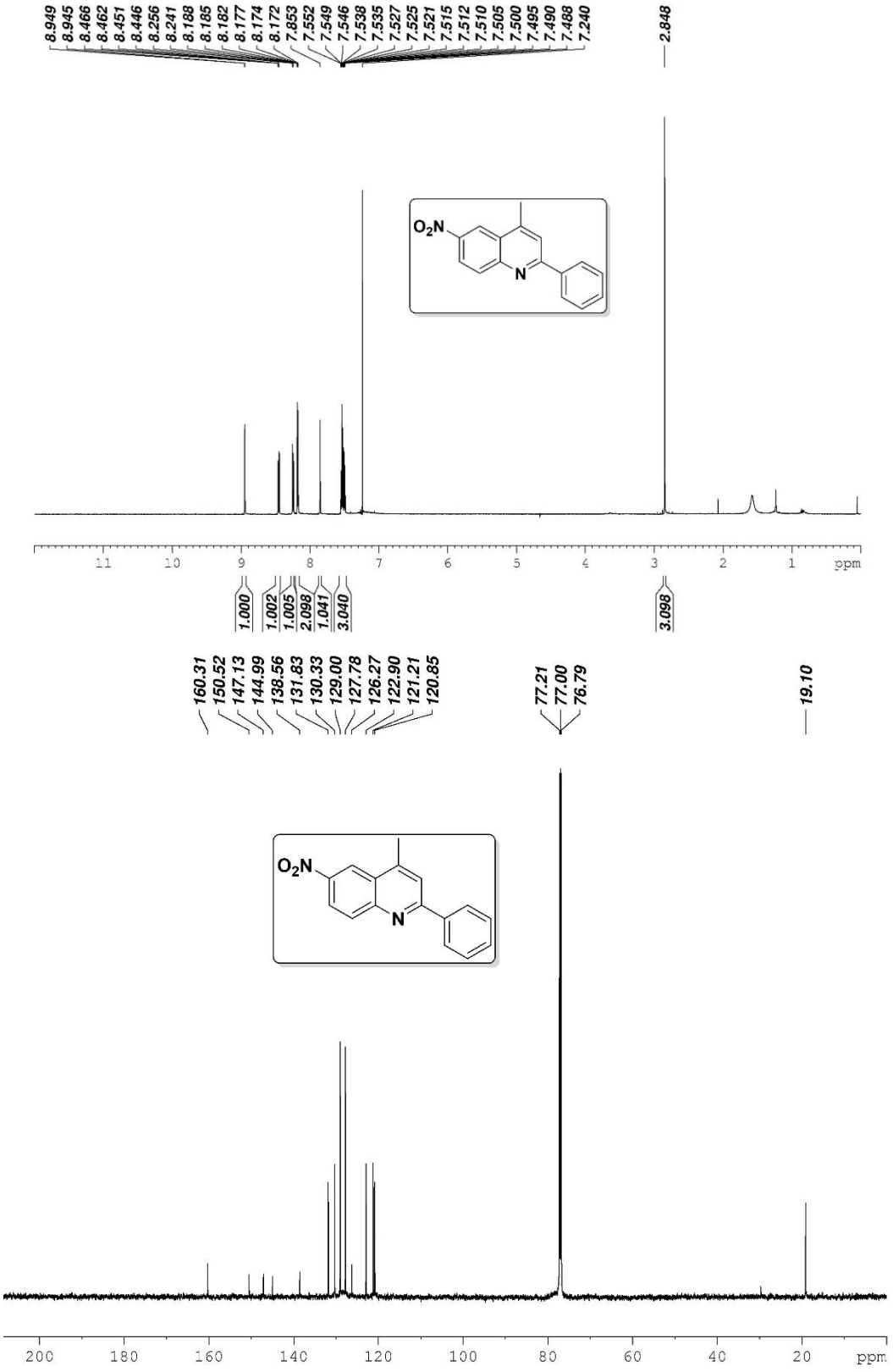
^{1}H and ^{13}C NMR spectra of compound **3ha**



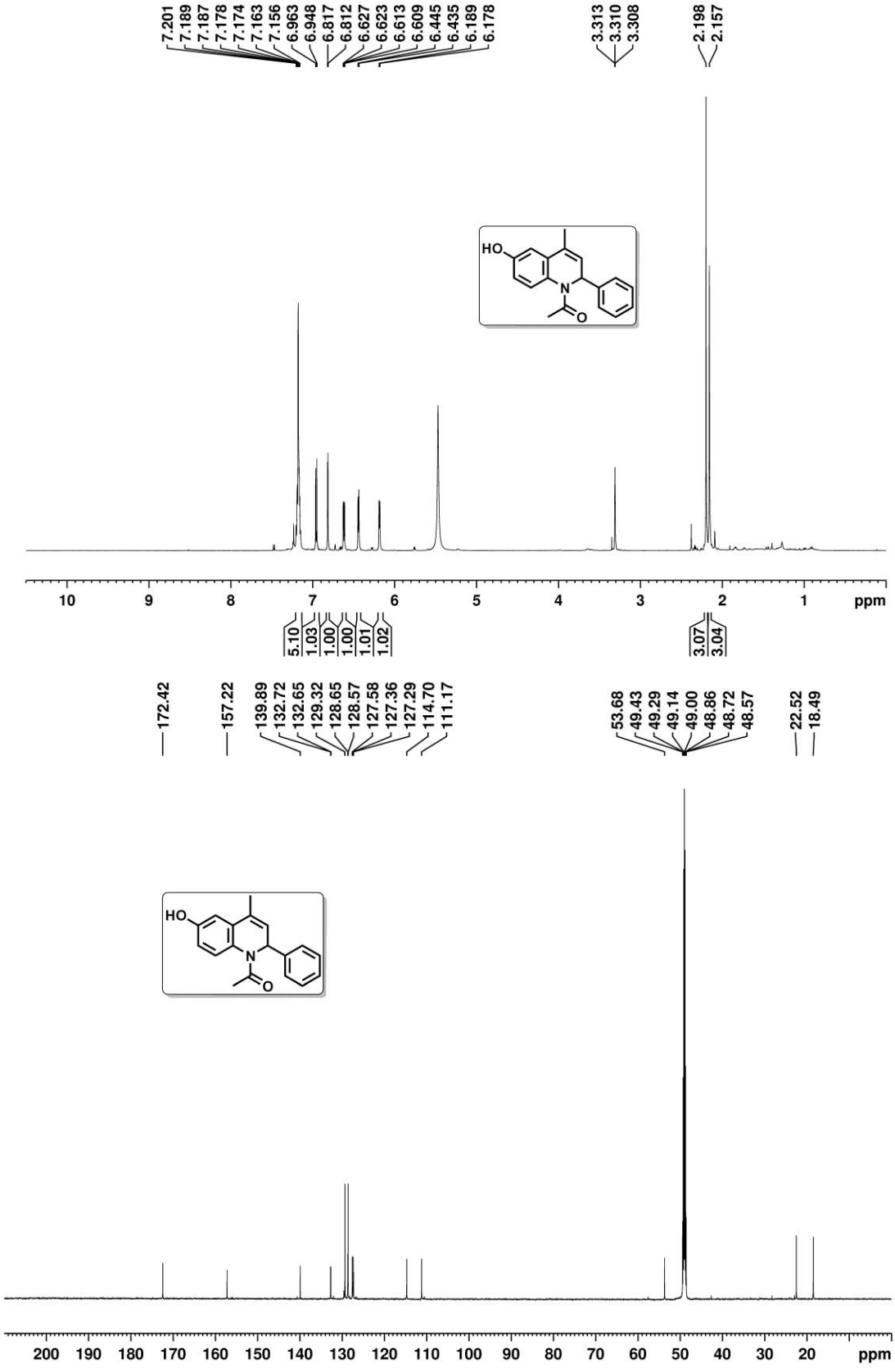
^{1}H and ^{13}C NMR spectra of compound **3ia**



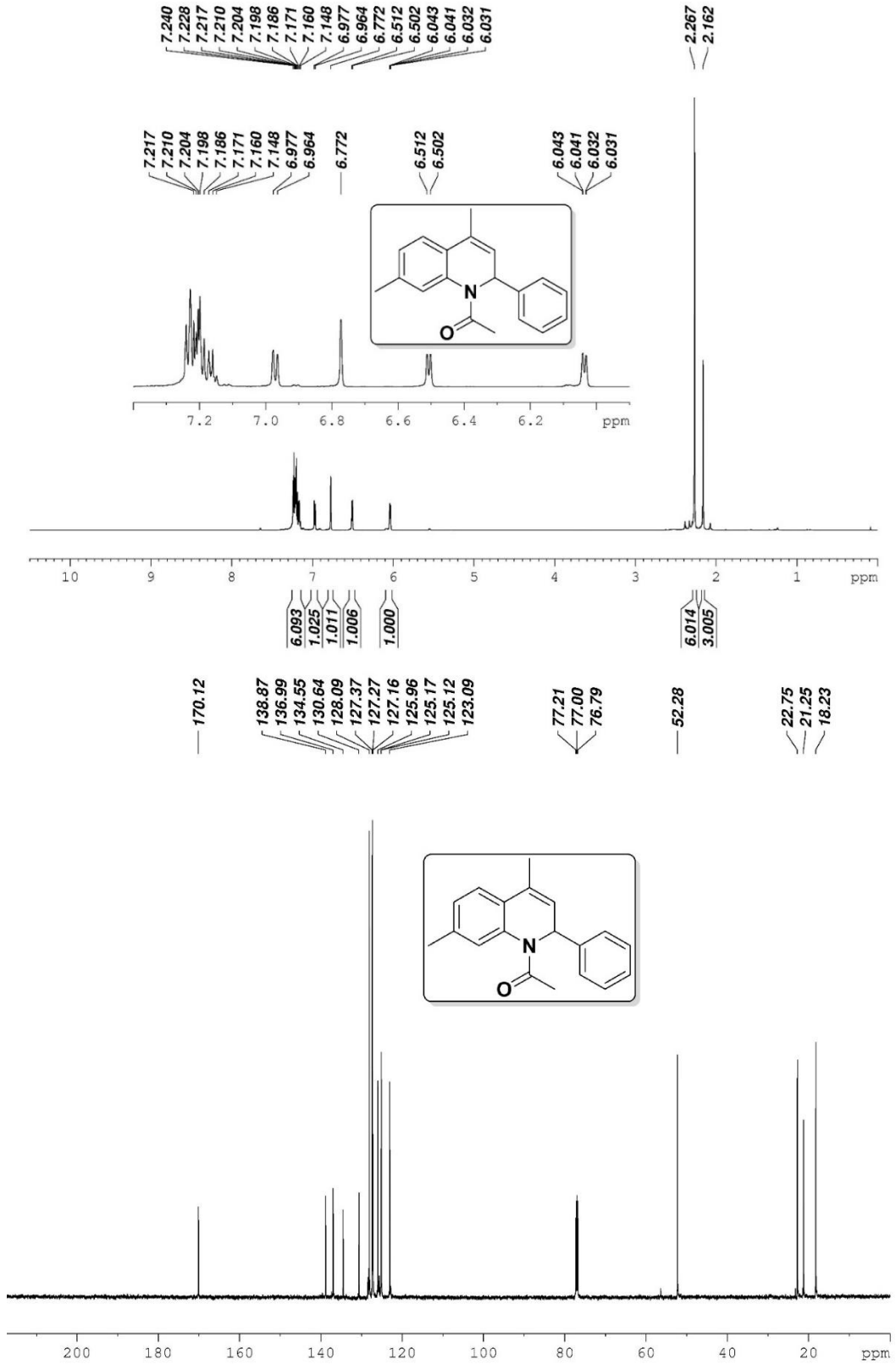
^{1}H and ^{13}C NMR spectra of compound **4ia**



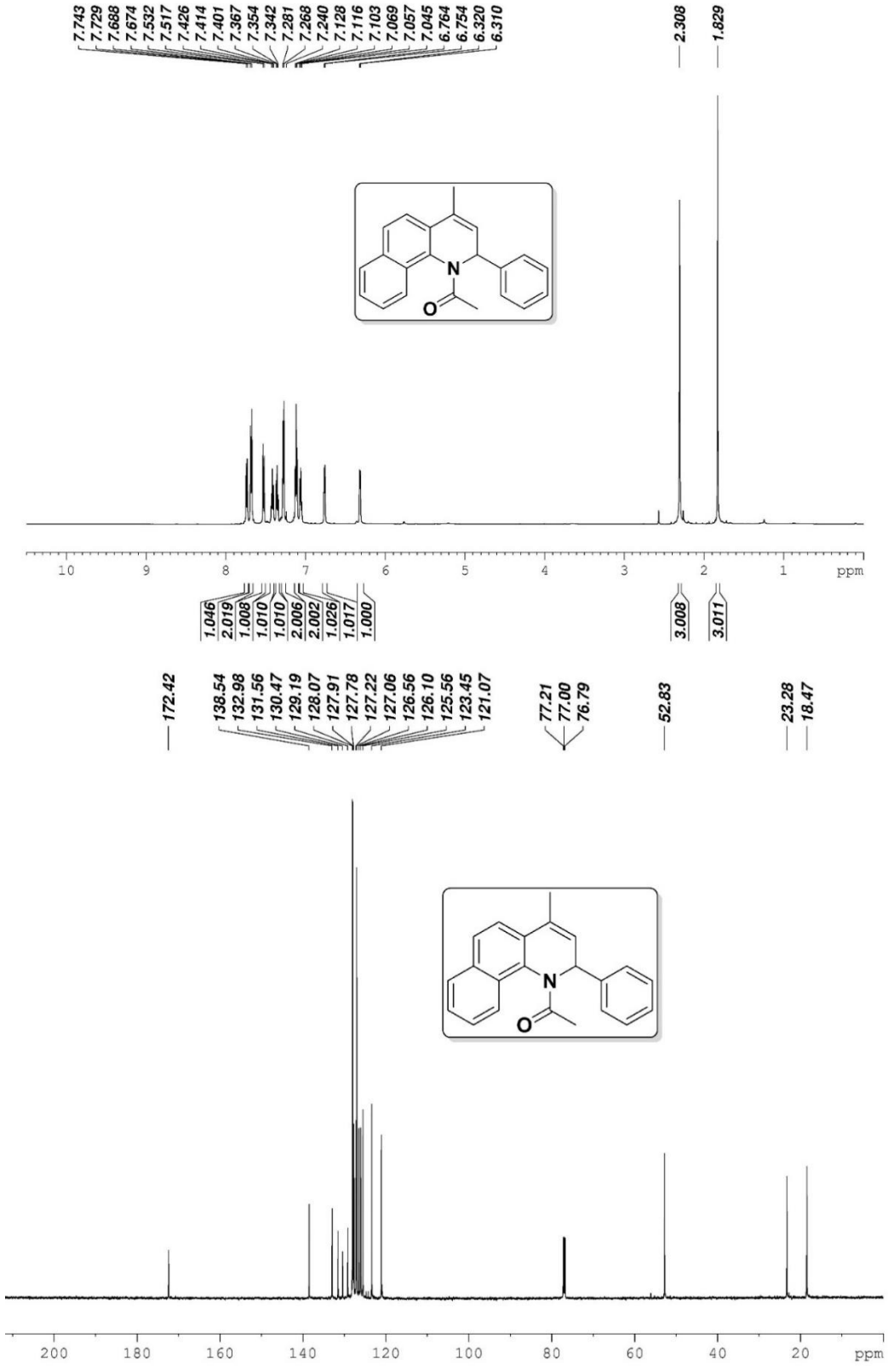
^{1}H and ^{13}C NMR spectra of compound **3ja**



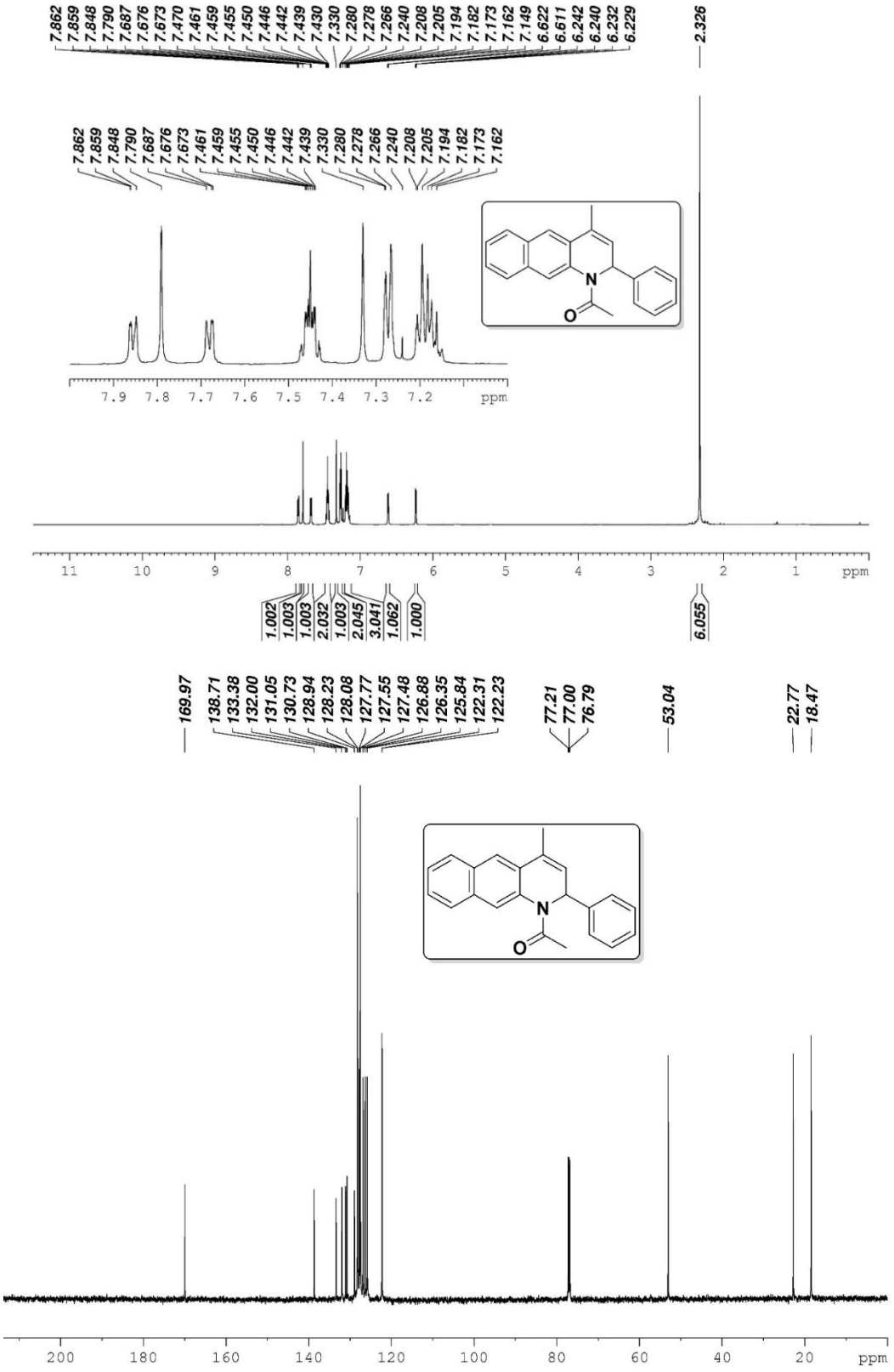
^{1}H and ^{13}C NMR spectra of compound **3ka**



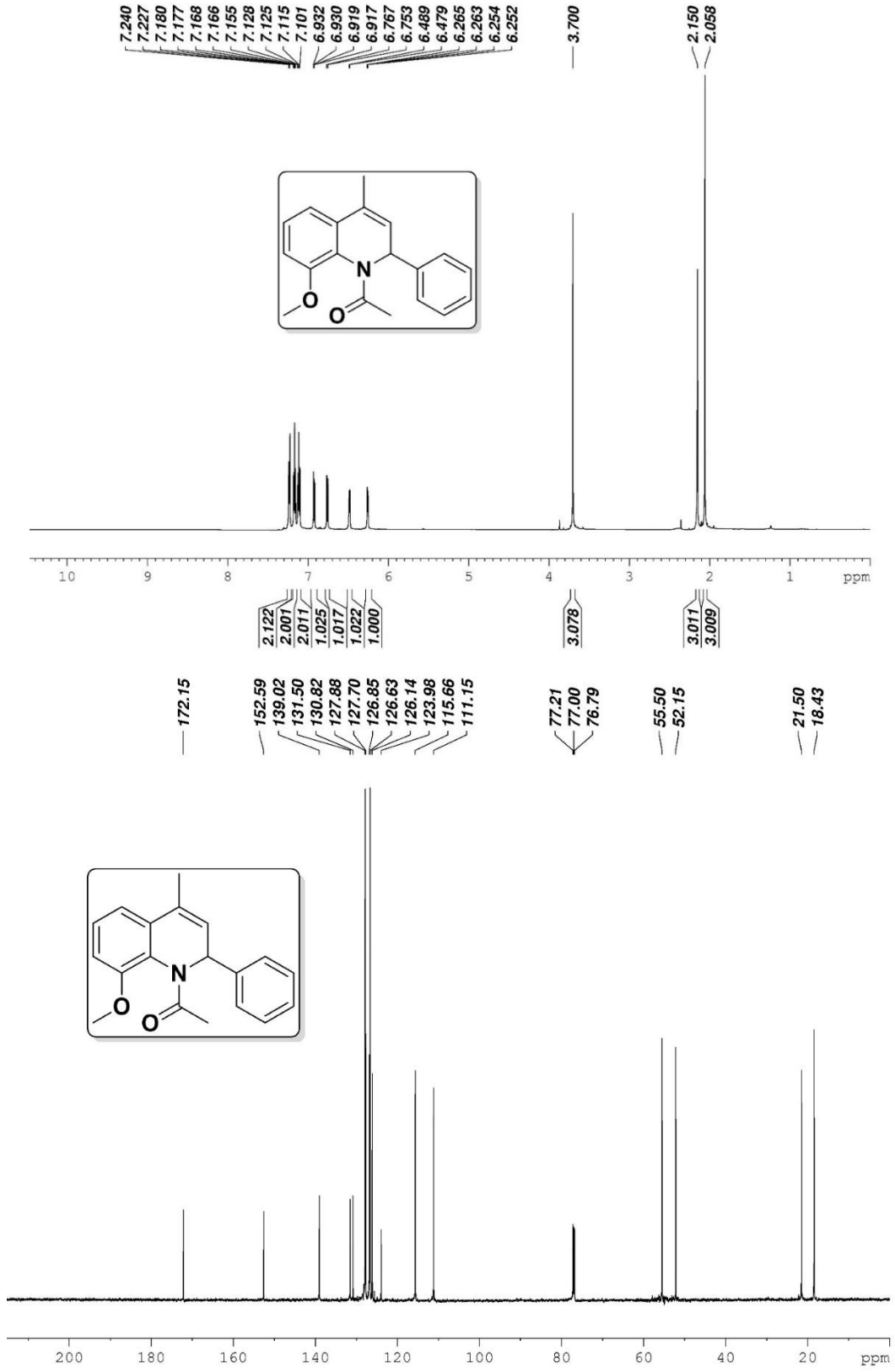
^{1}H and ^{13}C NMR spectra of compound **3la**



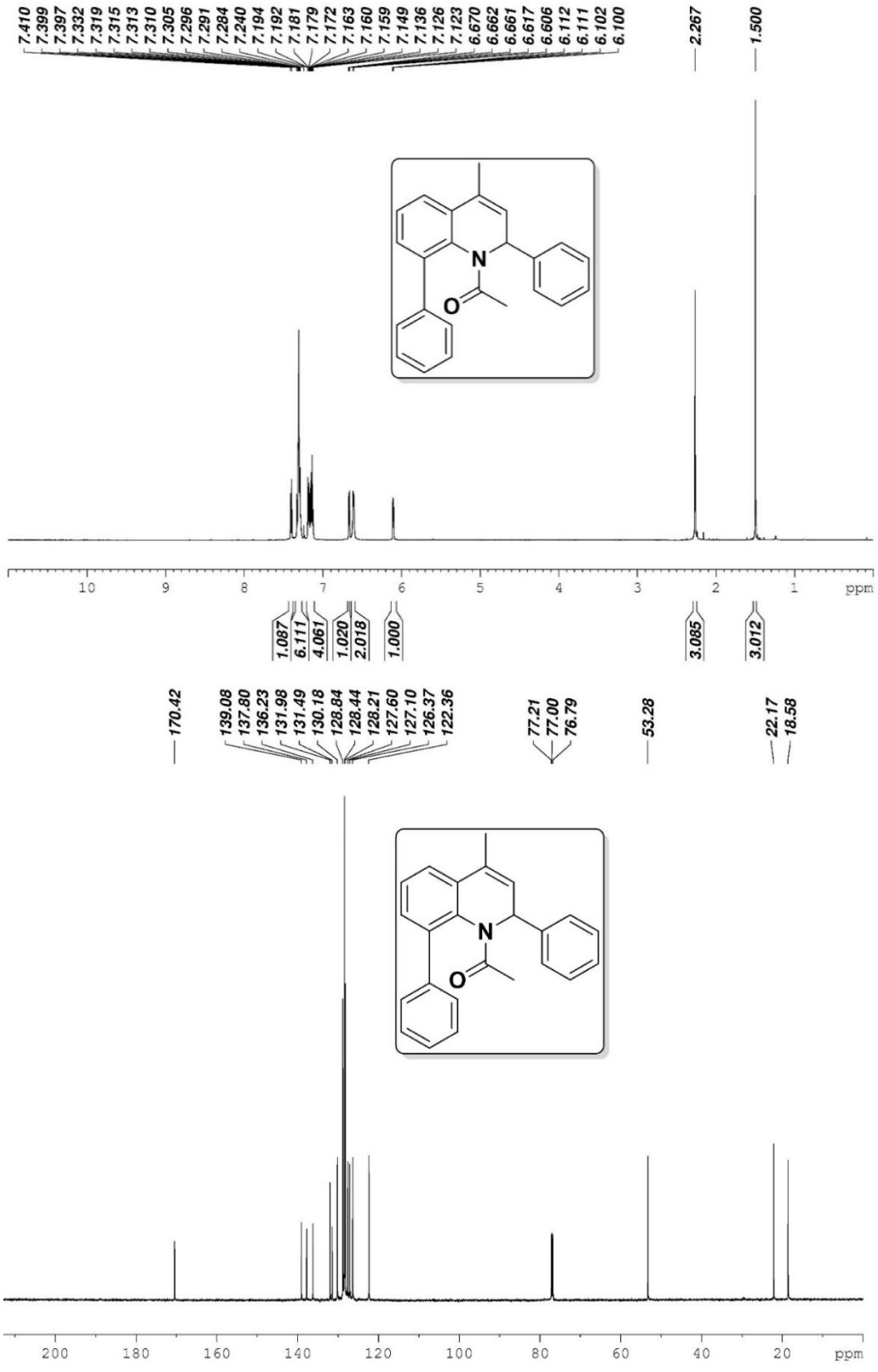
^{1}H and ^{13}C NMR spectra of compound **3ma**



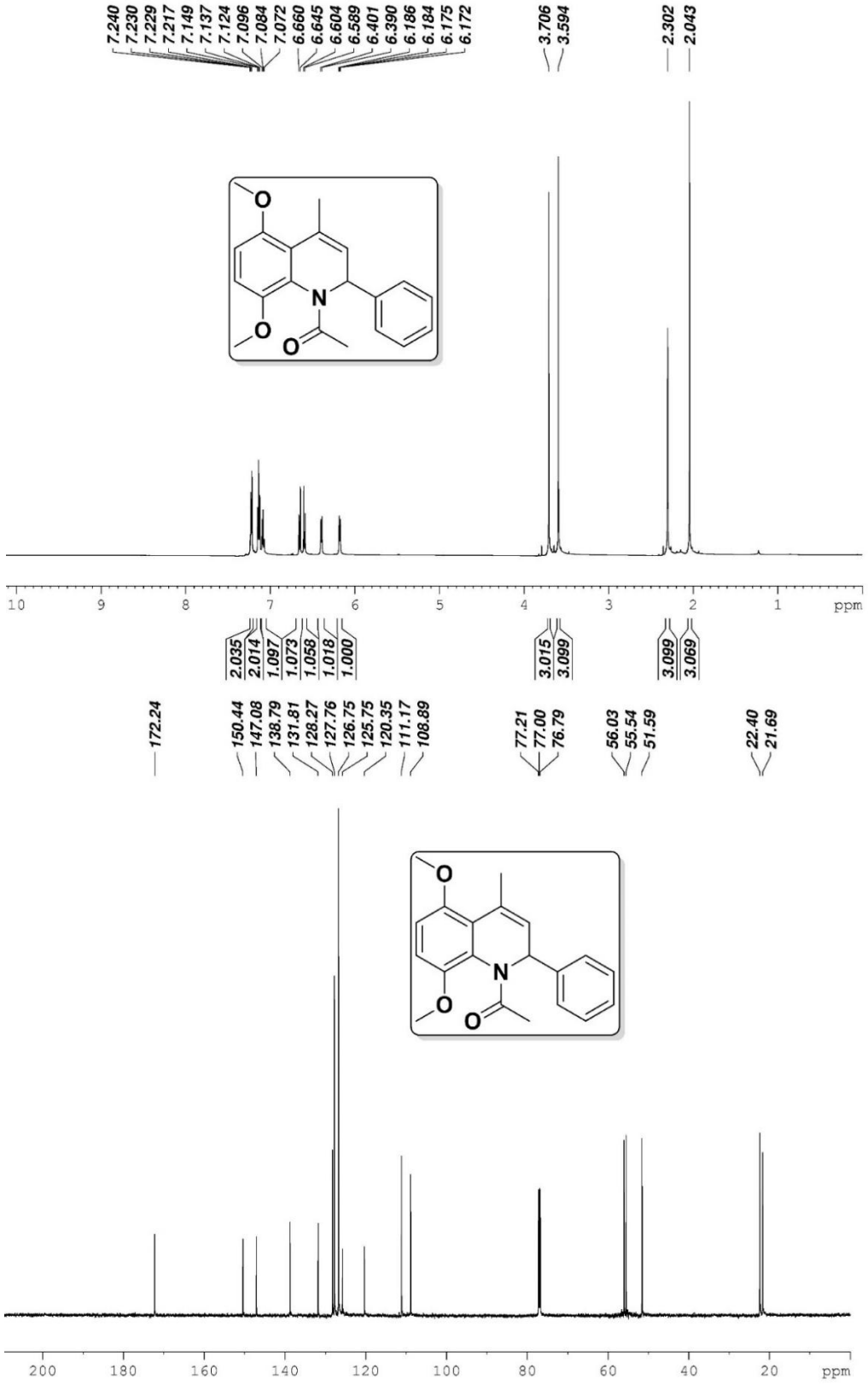
^{1}H and ^{13}C NMR spectra of compound **3na**



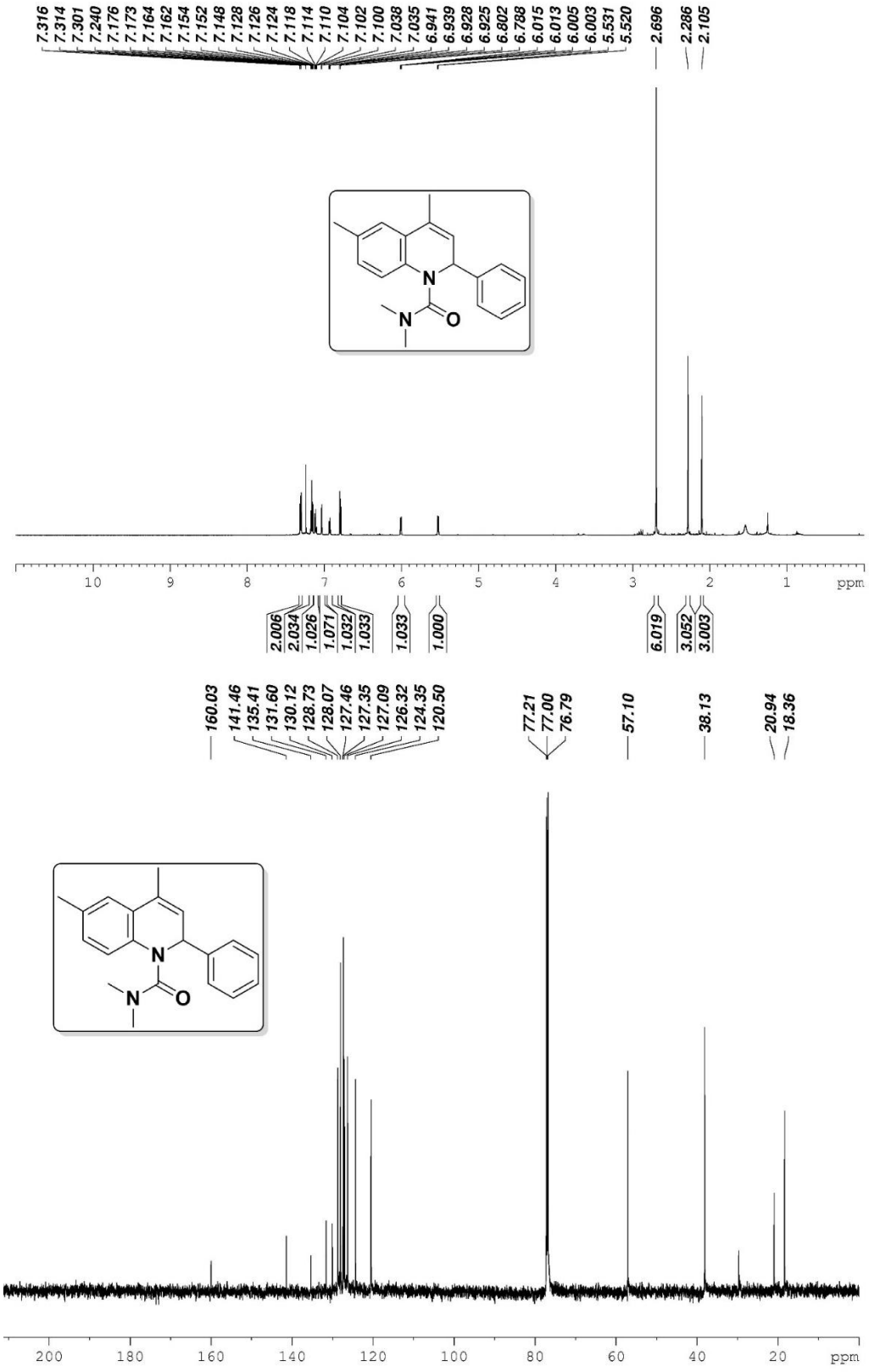
^{1}H and ^{13}C NMR spectra of compound **3oa**



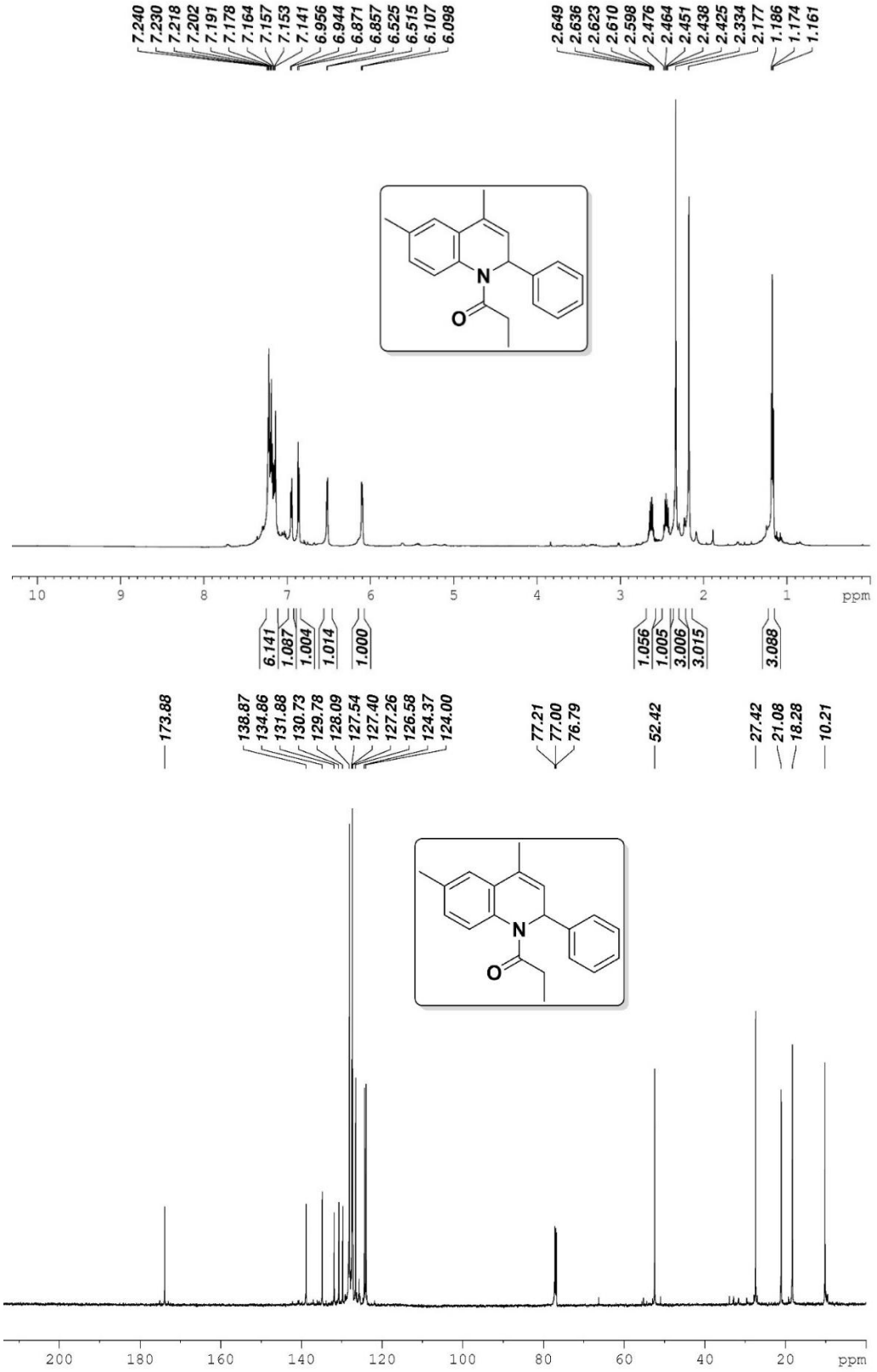
^{1}H and ^{13}C NMR spectra of compound **3pa**



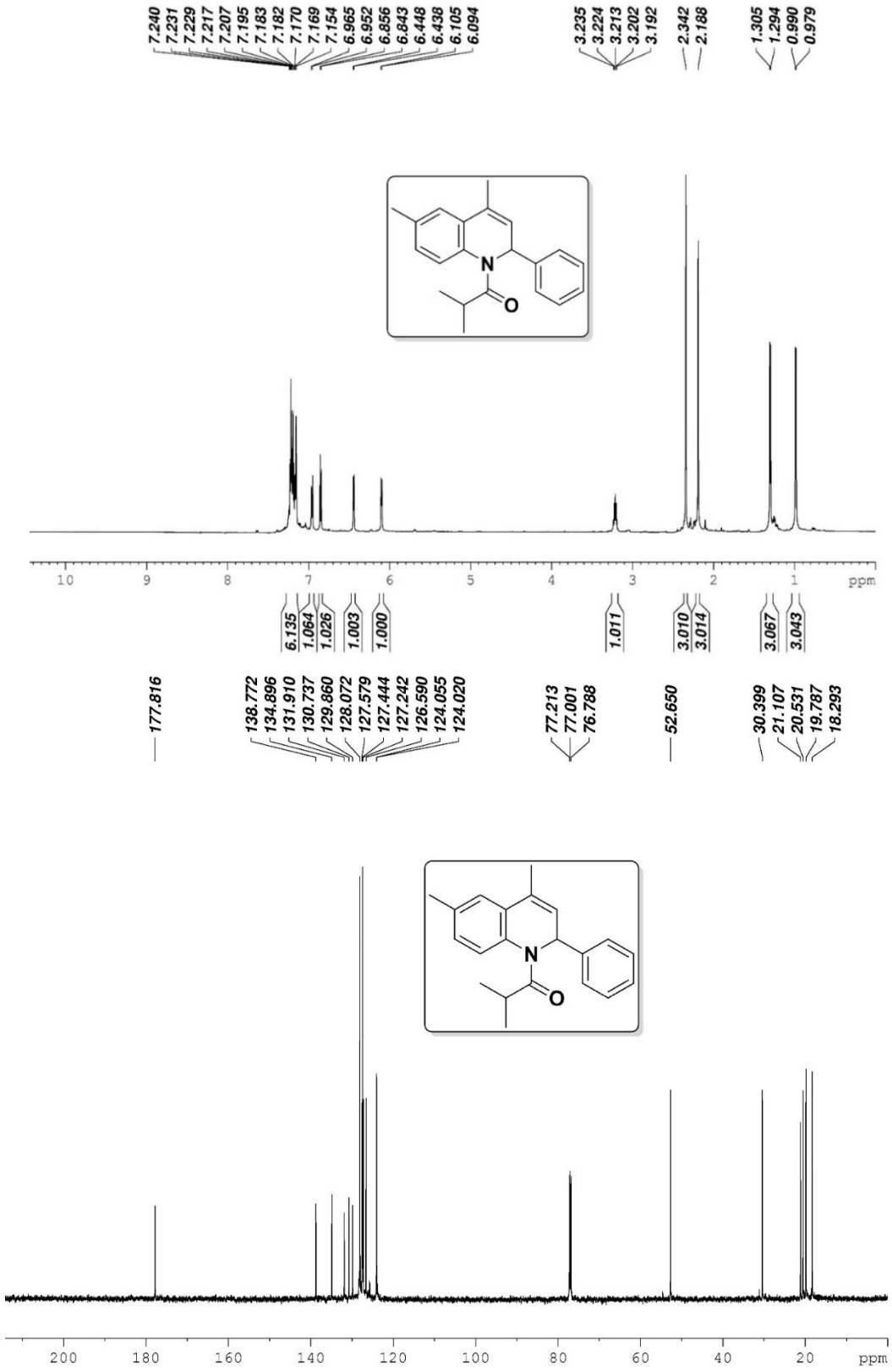
^{1}H and ^{13}C NMR spectra of compound **3qa**



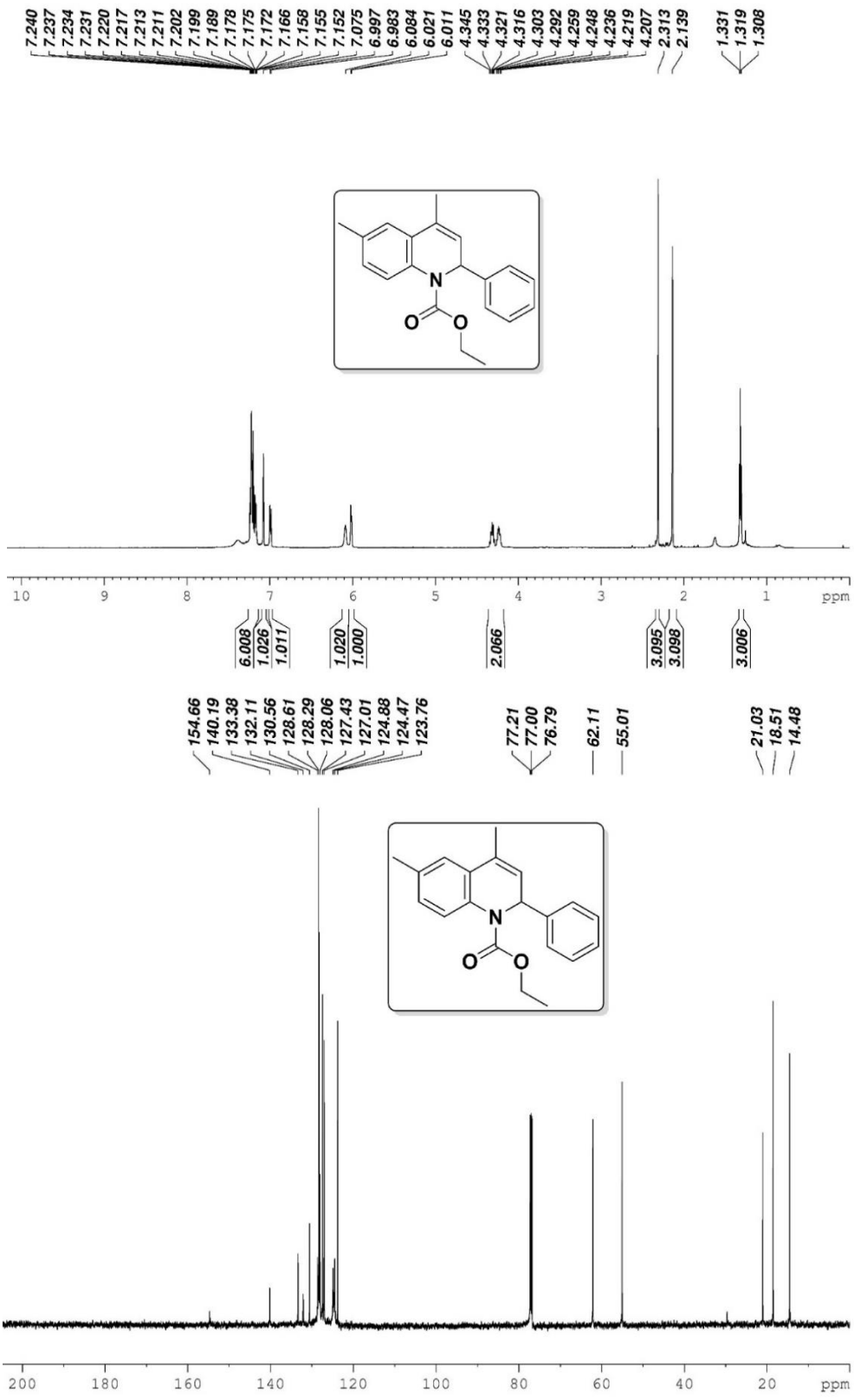
^{1}H and ^{13}C NMR spectra of compound **3ra**



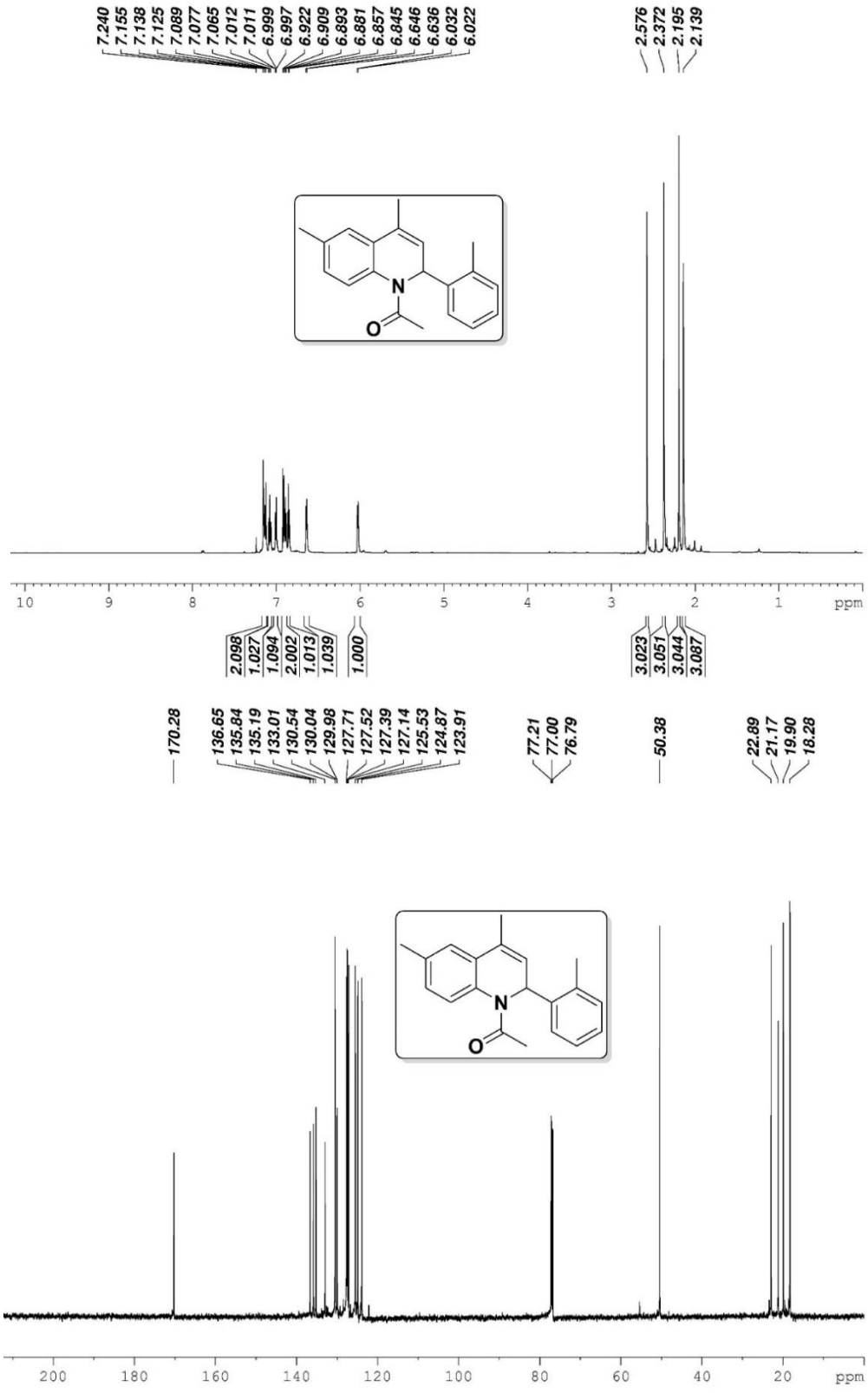
^{1}H and ^{13}C NMR spectra of compound **3sa**



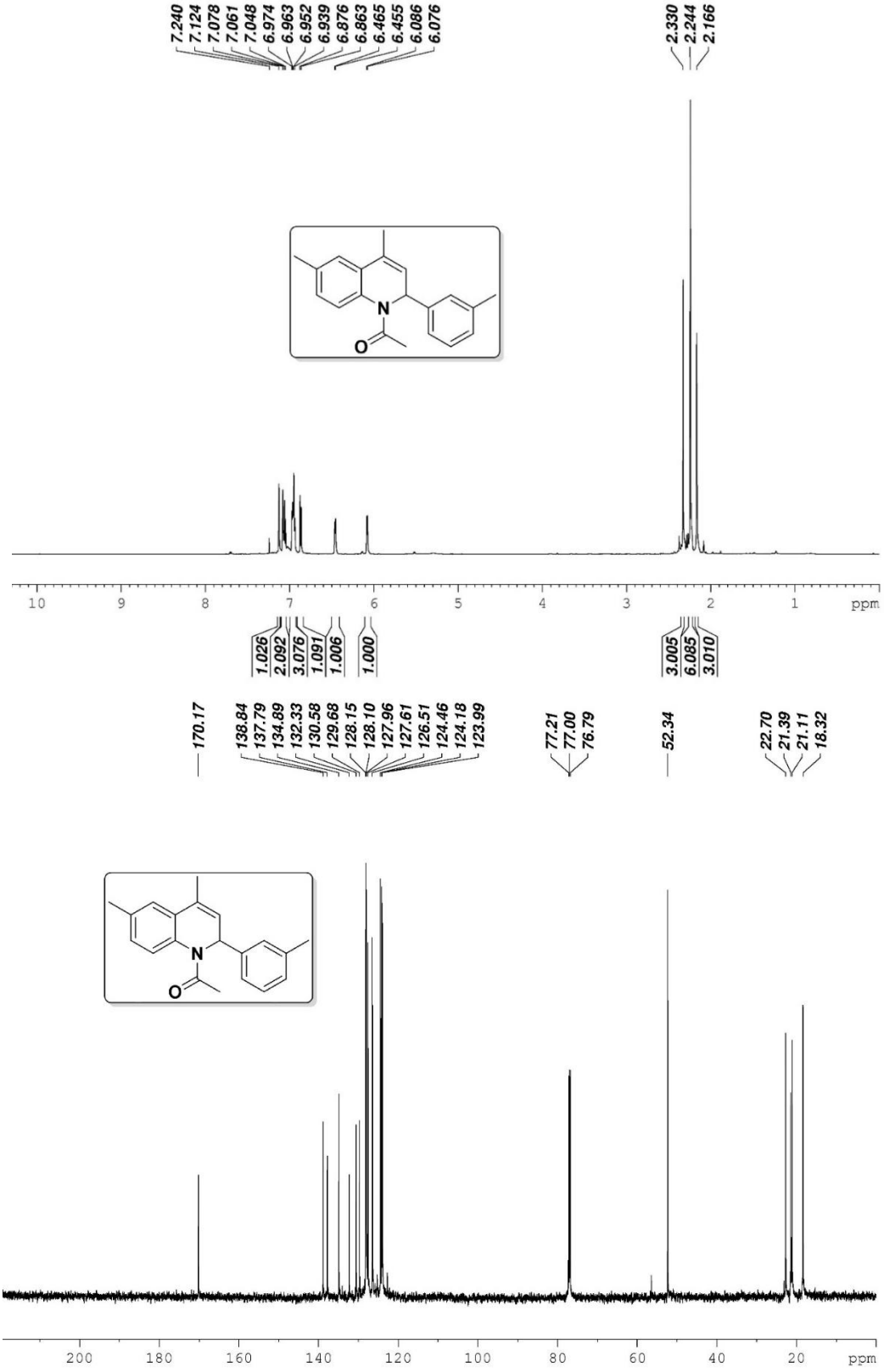
^{1}H and ^{13}C NMR spectra of compound **3ta**



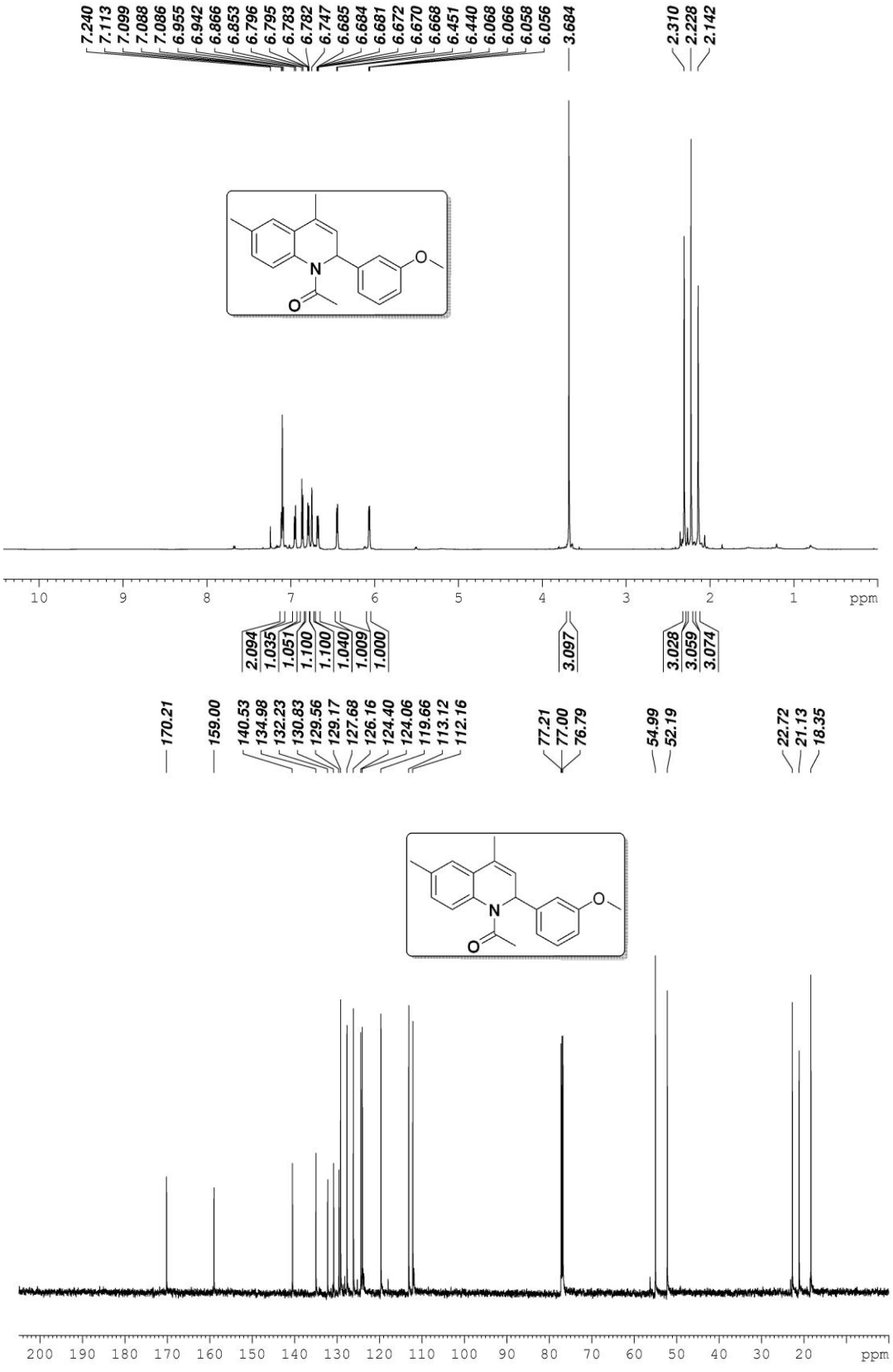
^{1}H and ^{13}C NMR spectra of compound **3ab**



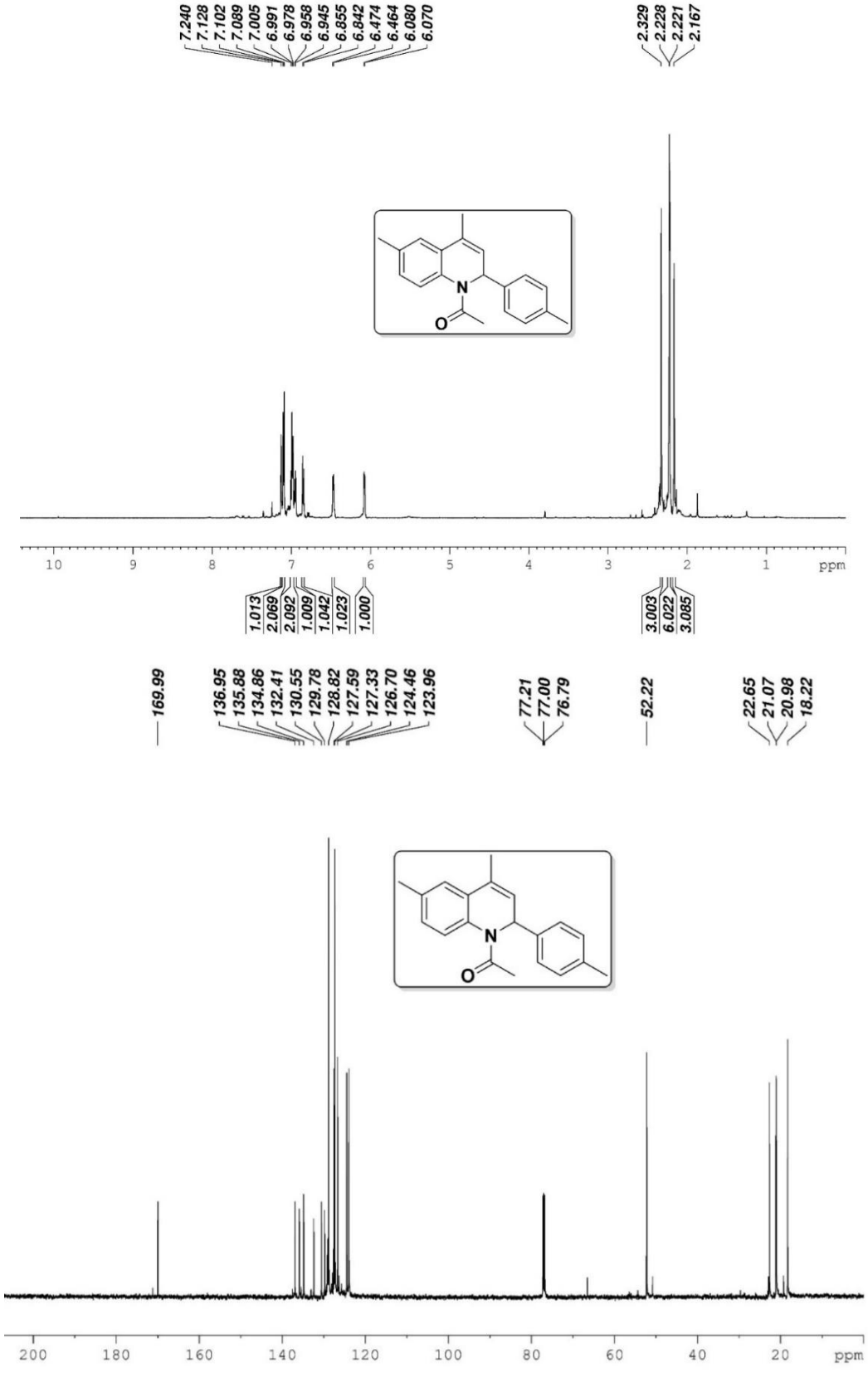
^{1}H and ^{13}C NMR spectra of compound **3ac**



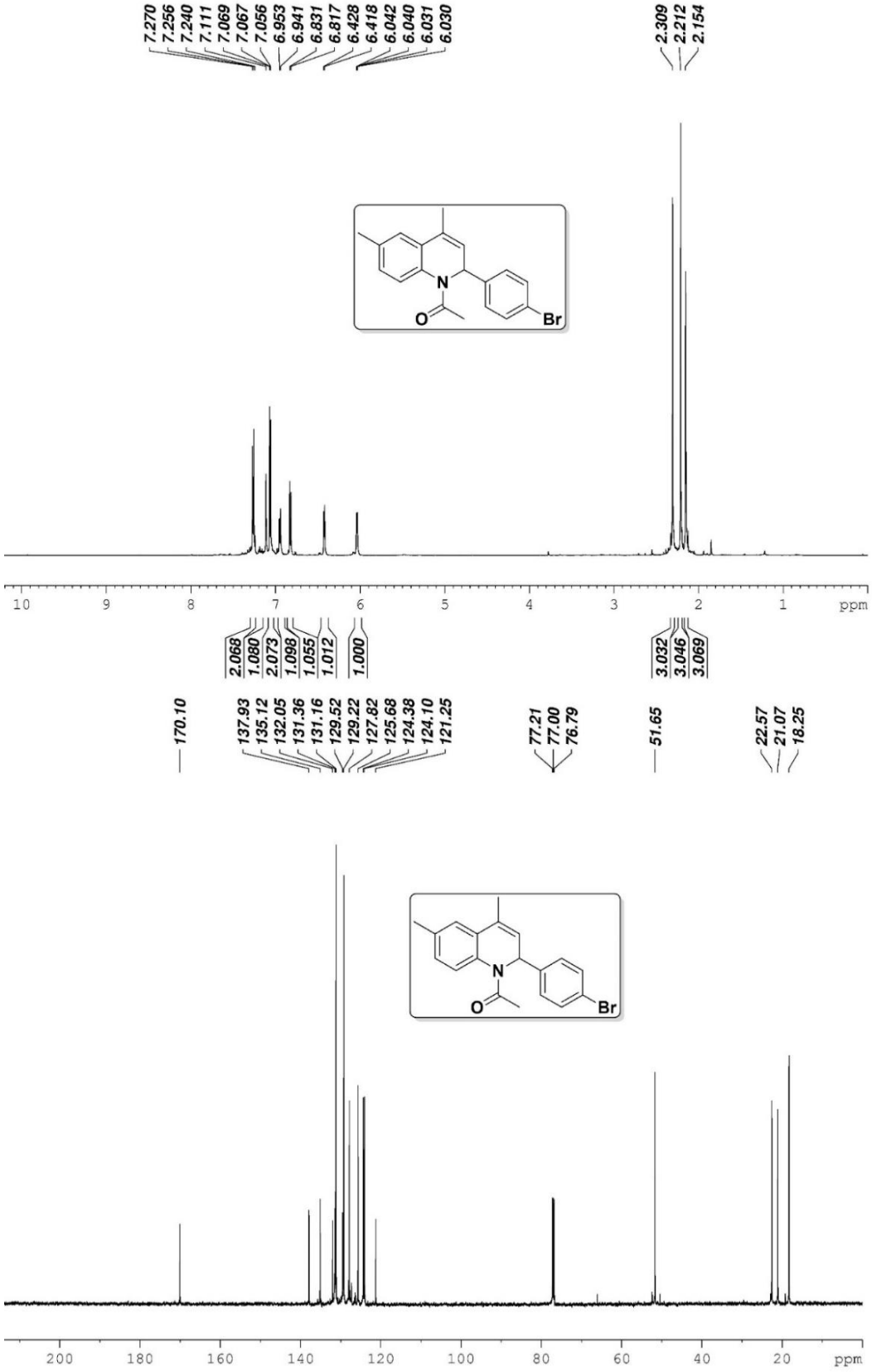
^{1}H and ^{13}C NMR spectra of compound **3ad**



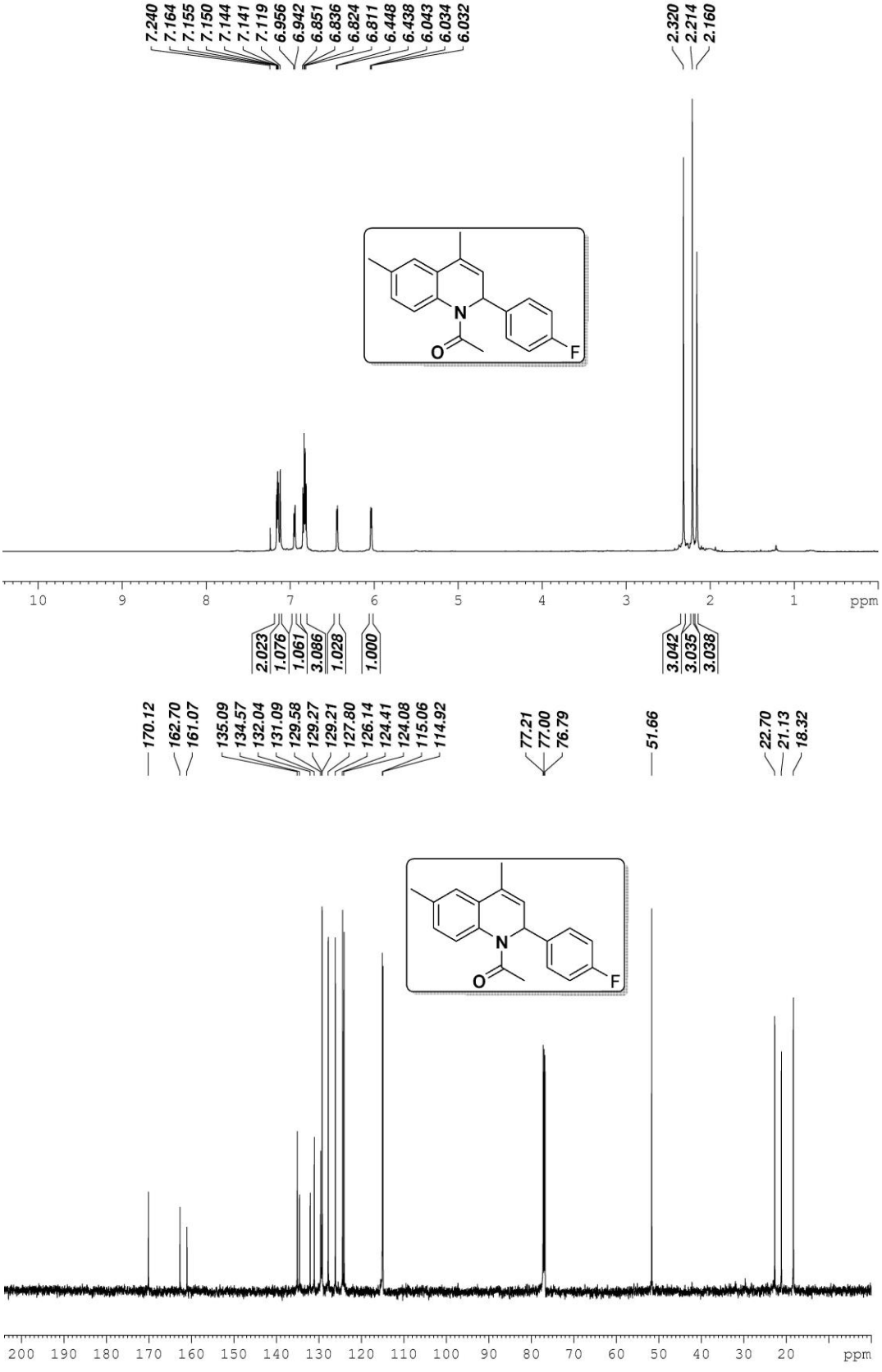
^{1}H and ^{13}C NMR spectra of compound **3ae**



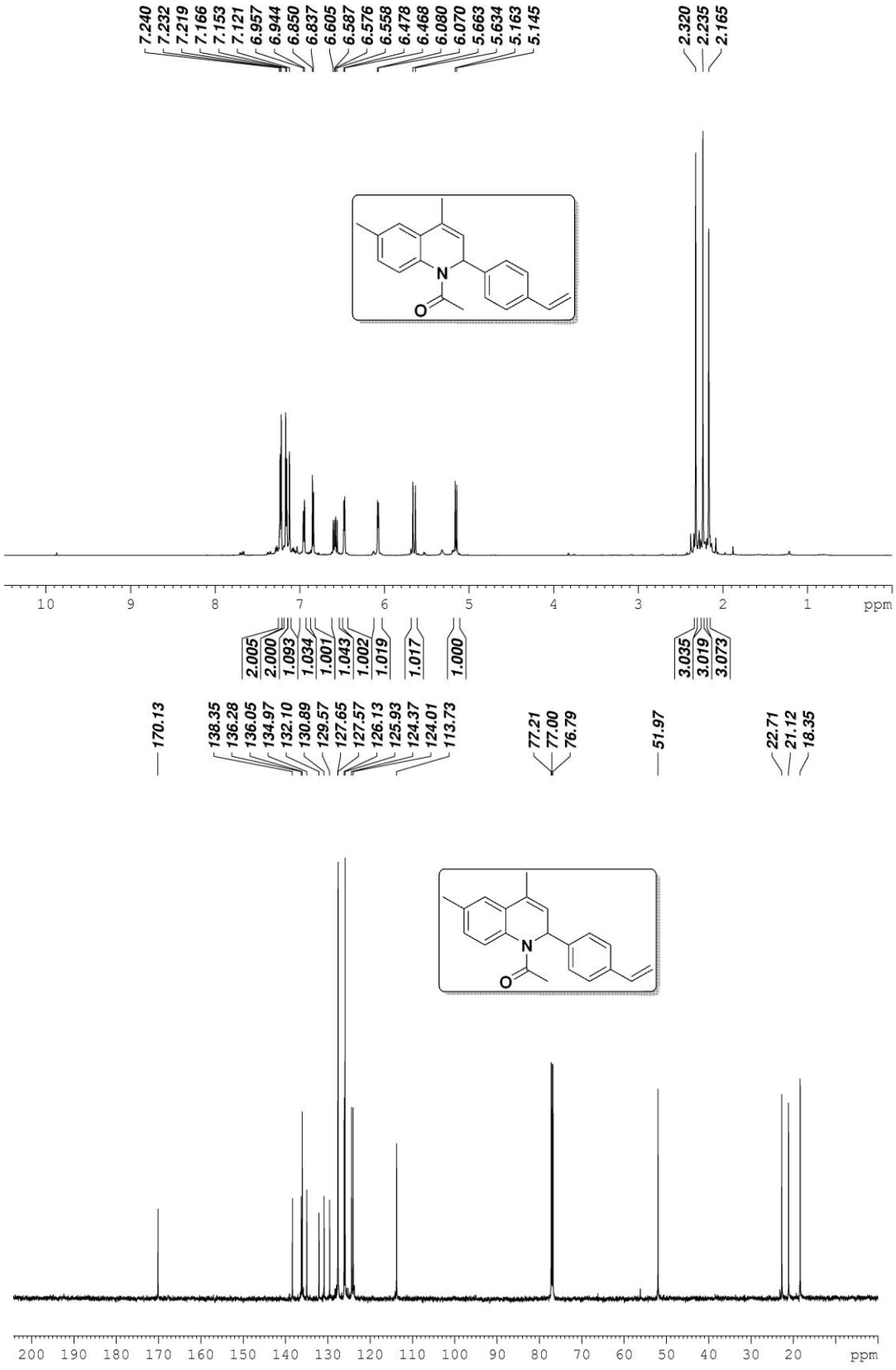
^{1}H and ^{13}C NMR spectra of compound **3af**



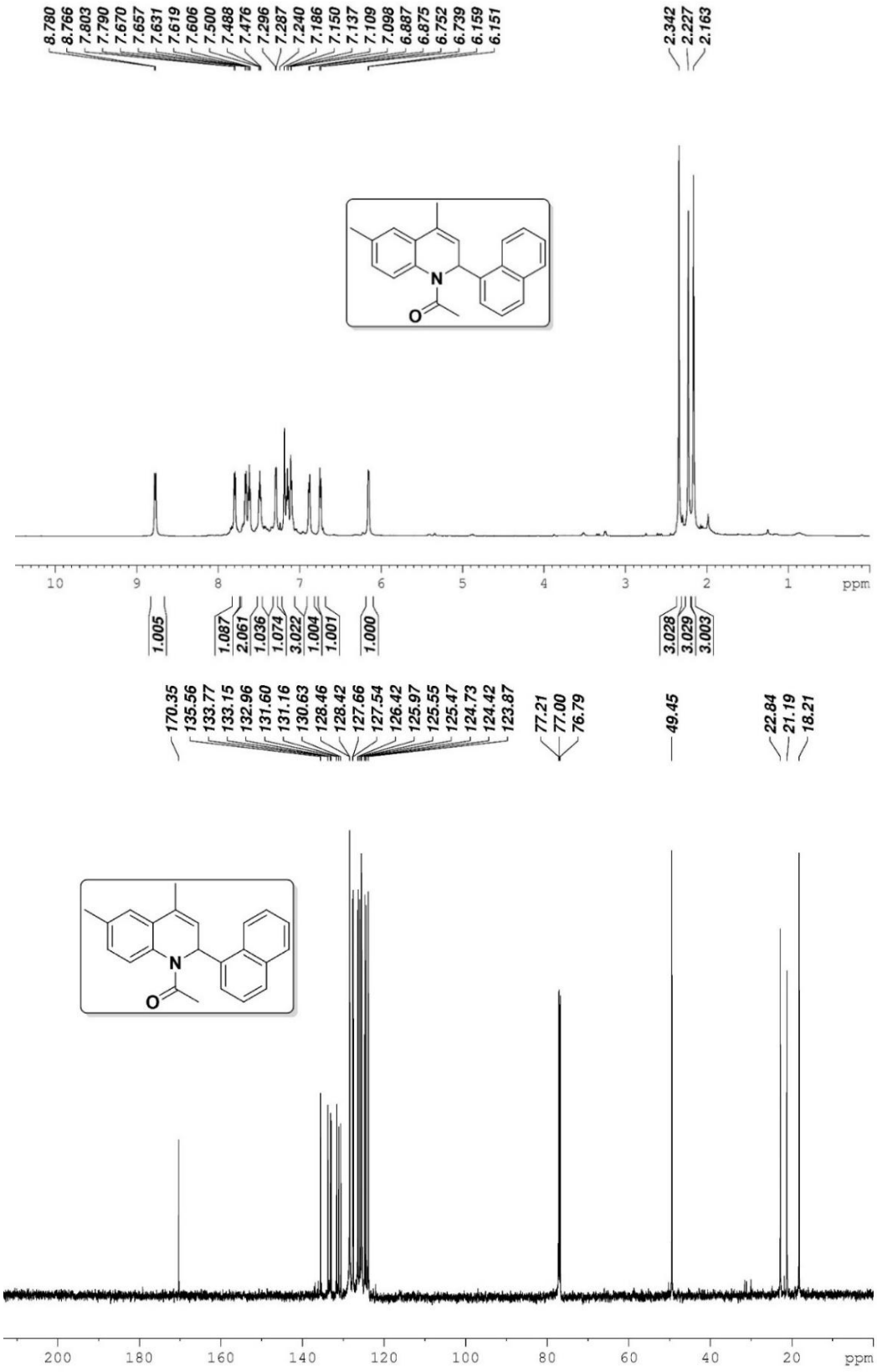
^{1}H and ^{13}C NMR spectra of compound **3ag**



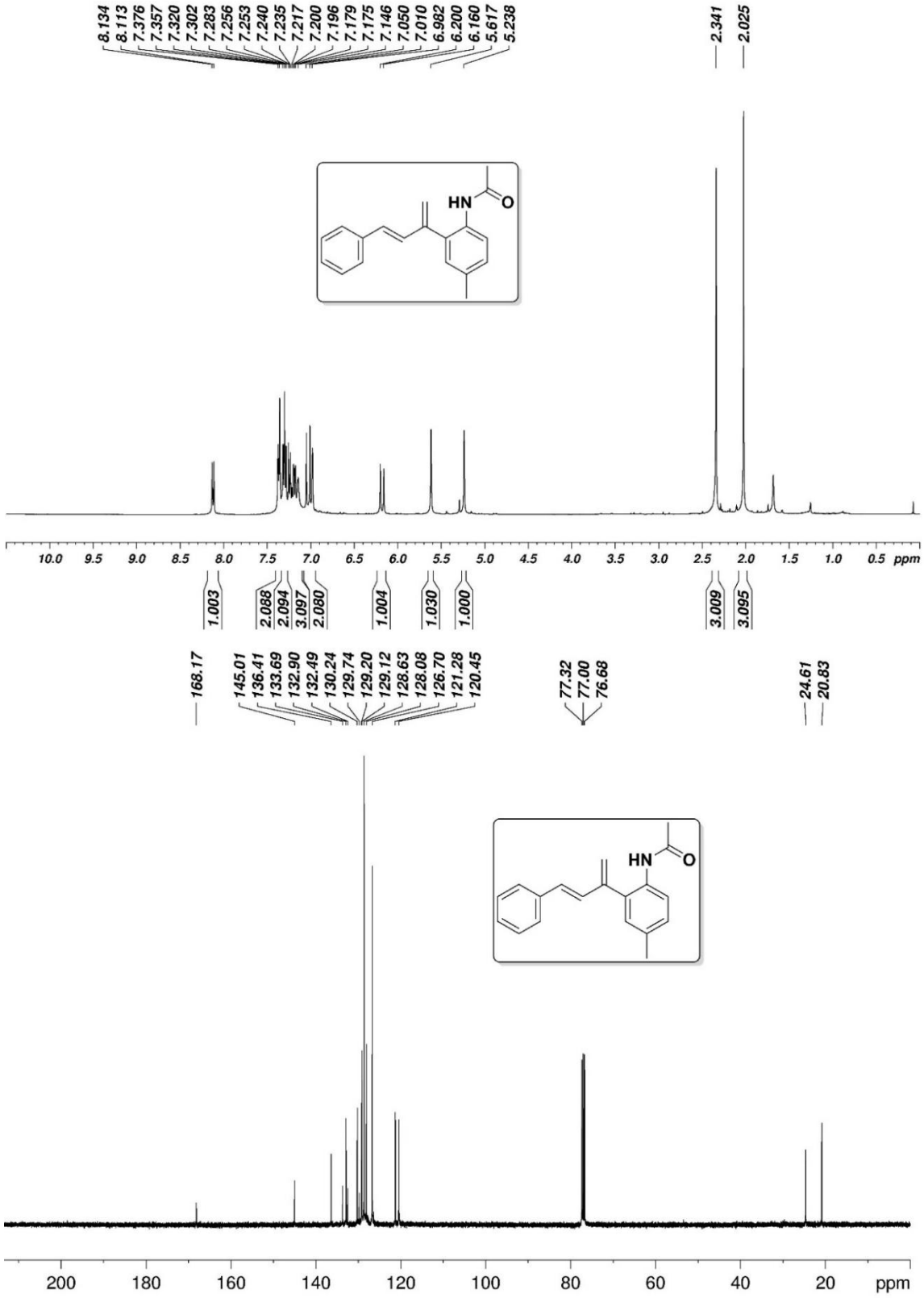
^{1}H and ^{13}C NMR spectra of compound **3ah**



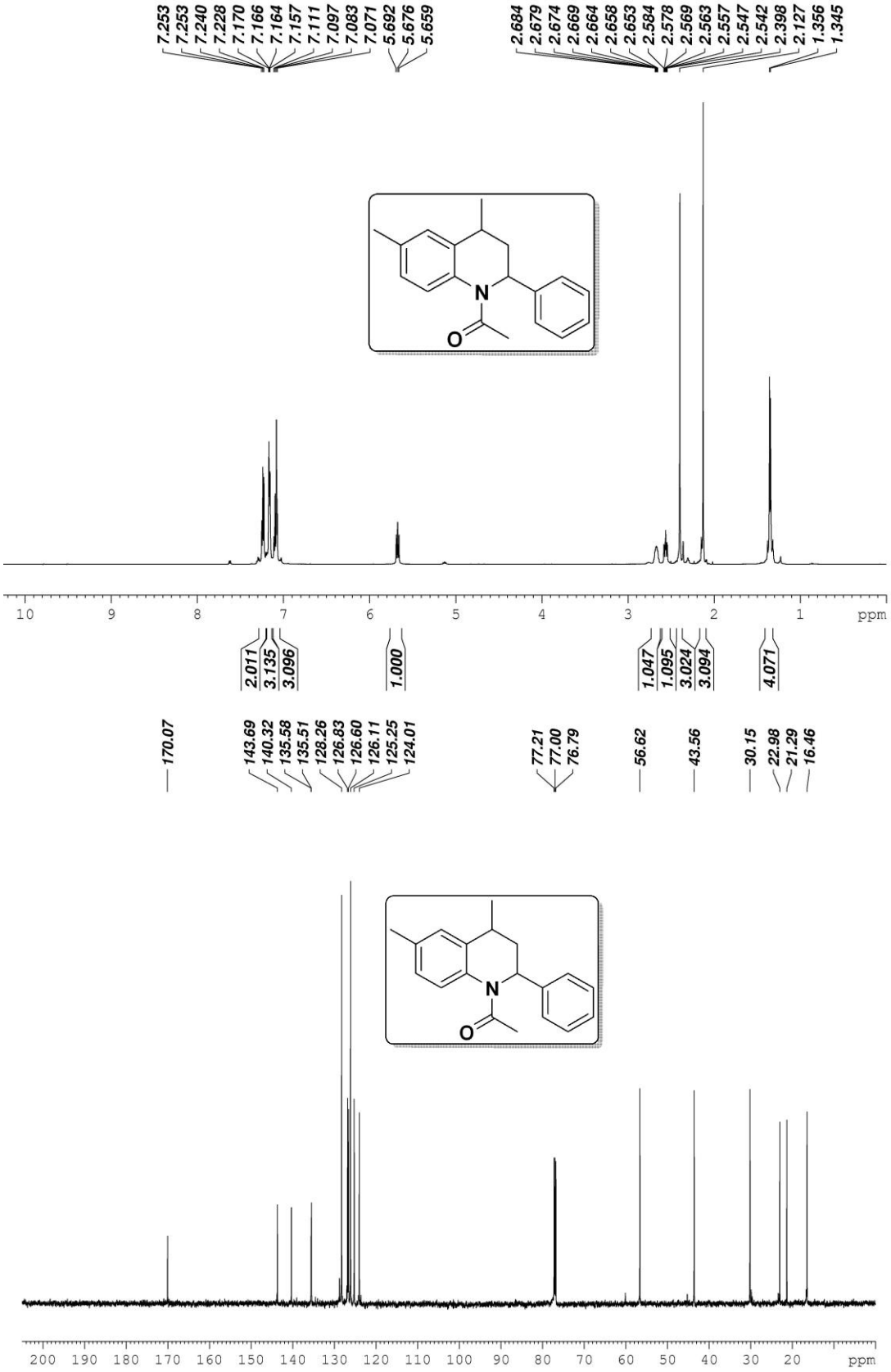
^{1}H and ^{13}C NMR spectra of compound **3ai**



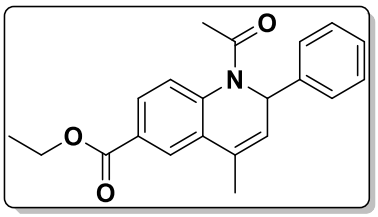
^{1}H and ^{13}C NMR spectra of compound **5aa**



^{1}H and ^{13}C NMR spectra of compound **6aa**



ORTEP Diagram and X-ray Data of 3ga



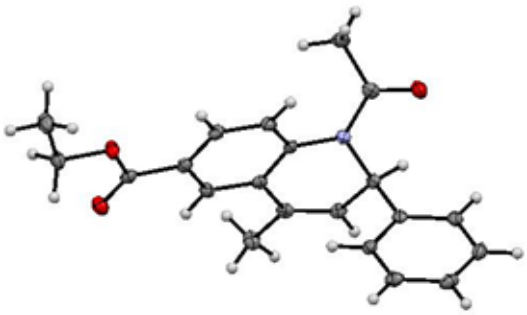


Table S7. Crystal data and structure refinement for 170626LT2.

Identification code	170626LT2	
Empirical formula	C21 H21 N O3	
Formula weight	335.39	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	P 21/c	
Unit cell dimensions	a = 13.6373(5) Å	α= 90°.
	b = 15.8546(6) Å	β= 104.499(2)°.
	c = 8.1178(2) Å	γ = 90°.
Volume	1699.28(10) $Å^3$	
Z	4	
Density (calculated)	1.311 Mg/m^3	
Absorption coefficient	0.087 mm^{-1}	
F(000)	712	
Crystal size	0.20 x 0.15 x 0.15 mm^3	
Theta range for data collection	1.542 to 26.461°.	
Index ranges	-17<=h<=17, -19<=k<=19, -7<=l<=10	
Reflections collected	14623	
Independent reflections	3499 [R(int) = 0.0383]	
Completeness to theta = 25.242°	100.0 %	

Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.9485 and 0.8901
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	3499 / 0 / 229
Goodness-of-fit on F^2	1.081
Final R indices [I>2sigma(I)]	R1 = 0.0409, wR2 = 0.0984
R indices (all data)	R1 = 0.0514, wR2 = 0.1120
Extinction coefficient	n/a
Largest diff. peak and hole	0.299 and -0.342 e.$Å^{-3}$