Supporting information:

A highly efficient heterogeneous copper-catalyzed cascade reaction of 2-halobenzoic acids and amidines leading to quinazolinones

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The spectral data of quinazolinone derivatives 3a-3y:

2-Methylquinazolin-4(3*H*)-one 3a. White solid, mp 237-239 °C (lit.[1] 238-240 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.10 (br, 1H), 8.08 (d, *J* = 8.0 Hz, 1H), 7.76 (t, *J* = 7.6 Hz, 1H), 7.57 (d, *J* = 8.0 Hz, 1H), 7.44 (t, *J* = 7.4 Hz, 1H), 2.36 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.7, 154.2, 148.9, 134.2, 126.5, 125.8, 125.6, 120.6, 21.4 ppm.

2-Phenylquinazolin-4(3*H*)-one 3b. White solid, mp 237-238 °C (lit.[1] 236-237 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.54 (br, 1H), 8.19-8.12 (m, 3H), 7.84-7.72 (m, 2H), 7.57-7.47 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.2, 152.3, 148.7, 134.5, 132.7, 131.3, 128.6, 127.7, 127.5, 126.5, 125.8, 121.0 ppm.

2-Cyclopropylquinazolin-4(3*H*)-one 3c. White solid, mp 232-233 °C (lit.[1] 233-235 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.44 (br, 1H), 8.03 (d, *J* = 8.0 Hz, 1H), 7.70 (t, *J* = 7.6 Hz, 1H), 7.45 (d, *J* = 8.0 Hz, 1H), 7.37 (t, *J* = 7.4 Hz, 1H), 1.97-1.92 (m, 1H), 1.10-1.00 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.6, 159.0, 149.1, 134.2, 126.4, 125.7, 125.2, 120.6, 13.4, 9.0 ppm.

2-(*tert*-Butyl)quinazolin-4(3*H*)-one 3d. White solid, mp 207-209 °C (lit.[2] 206-208 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.95 (br, 1H), 8.13 (d, *J* = 7.6 Hz, 1H), 7.79 (t, *J* = 7.2 Hz, 1H), 7.65 (d, *J* = 8.0 Hz, 1H), 7.50 (t, *J* = 7.6 Hz, 1H), 1.38 (s, 9H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.6, 162.3, 148.3, 134.2, 127.3, 126.1, 125.5, 120.6, 37.2, 27.8 ppm.

2-(*p*-Tolyl)quinazolin-4(3*H*)-one 3e. White solid, mp 260-262 °C (lit.[2] 261-263 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.49 (br, 1H), 8.15 (d, *J* = 8.0 Hz, 1H), 8.10 (d, *J* = 7.6 Hz, 2H), 7.82 (t, *J* = 7.4 Hz, 1H), 7.72 (d, *J* = 7.6 Hz, 1H), 7.50 (t, *J* = 7.0 Hz, 1H), 7.34 (d, *J* = 7.2 Hz, 2H), 2.38 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.2, 152.2, 148.8, 141.4, 134.5, 129.9, 129.1, 127.6, 127.4, 126.3, 125.8, 120.9, 20.9 ppm.

6-Chloro-2-methylquinazolin-4(3*H*)-one 3f. White solid, mp 295-297 °C (lit.[1] 294-296 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.37 (br, 1H), 7.95 (d, *J* = 2.0 Hz, 1H), 7.74 (t, *J* = 8.4 Hz, 1H), 7.55 (d, *J* = 8.4 Hz, 1H), 2.32 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.7, 154.9, 147.6, 134.3, 130.0, 128.8, 124.6, 121.8, 21.4 ppm.

6-Chloro-2-phenylquinazolin-4(3*H*)-one 3g. White solid, mp 287-289 °C (lit.[1] 286-

288 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.72 (br, 1H), 8.18 (d, *J* = 7.2 Hz, 2H), 8.09 (d, *J* = 2.0 Hz, 1H), 7.88-7.84 (m, 1H), 7.77 (d, *J* = 8.8 Hz, 1H), 7.61-7.54 (m, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.3, 152.8, 147.5, 134.7, 132.4, 131.6, 130.7, 129.7, 128.6, 127.8, 126.0, 122.2 ppm.

6-Chloro-2-cyclopropylquinazolin-4(3*H*)-one 3h. White solid, mp 292-294 °C (lit.[1] 294-296 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.61 (br, 1H), 7.97 (d, *J* = 2.4 Hz, 1H), 7.73 (d, *J* = 8.8 Hz, 1H), 7.49 (d, *J* = 8.8 Hz, 1H), 1.99-1.95 (m, 1H), 1.10-1.04 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.6, 159.7, 147.8, 134.3, 129.4, 128.7, 124.7, 121.8, 13.5, 9.6 ppm.

2-(*tert*-Butyl)-6-chloroquinazolin-4(3*H*)-one 3i. White solid, mp 256-258 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.14 (br, 1H), 8.06 (d, *J* = 2.0 Hz, 1H), 7.84 (dd, *J* = 8.8, 2.4 Hz, 1H), 7.67 (d, *J* = 8.8 Hz, 1H), 1.38 (s, 9H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 163.3, 161.3, 147.0, 134.4, 130.4, 129.5, 124.6, 121.9, 37.3, 27.7 ppm. Anal. Calcd. for $C_{12}H_{13}N_2OCl$: C, 60.90; H, 5.54; N, 11.83. Found: C, 60.68; H, 5.38; N, 11.65.

6-Chloro-2-(*p*-tolyl)quinazolin-4(3*H*)-one 3j. White solid, mp 300-302 °C (lit.[3] 302-304 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.61 (br, 1H), 8.08 (d, *J* = 8.0 Hz, 2H), 8.06 (d, *J* = 2.0 Hz, 1H), 7.85-7.82 (m, 1H), 7.74 (d, *J* = 8.8 Hz, 1H), 7.35 (d, *J* = 8.0 Hz, 2H), 2.39 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.3, 152.8, 147.6, 141.7, 134.6, 130.6, 129.6, 129.2, 127.7, 125.9, 124.8, 122.1, 20.9 ppm.

2-Methyl-6-nitroquinazolin-4(3*H*)-one 3k. Light yellow solid, mp 291-293 °C (lit.[1] 293-295 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.69 (br, 1H), 8.68 (s, 1H), 8.43

(d, *J* = 8.8 Hz, 1H), 7.67 (d, *J* = 9.2 Hz, 1H), 2.37 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 160.8, 158.5, 153.1, 144.3, 128.3, 128.1, 121.8, 120.6, 21.7 ppm.

6-Nitro-2-phenylquinazolin-4(3*H*)-one 3l. Light yellow solid, mp 295-297 °C (lit.[1] 297-299 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 13.01 (br, 1H), 8.82 (s, 1H), 8.55 (d, *J* = 8.8 Hz, 1H), 8.22 (d, *J* = 7.6 Hz, 2H), 7.90 (d, *J* = 8.8 Hz, 1H), 7.67-7.57 (m, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.1, 156.2, 153.5, 145.2, 132.7, 132.5, 129.7, 129.2, 129.0, 128.7, 122.5, 121.5 ppm.

2-Cyclopropyl-6-nitroquinazolin-4(3*H*)-one 3m. Light yellow solid, mp 291-293 °C (lit.[1] 290-292 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.89 (br, 1H), 8.68 (d, *J* = 2.8 Hz, 1H), 8.38 (dd, *J* = 9.2, 2.8 Hz, 1H), 7.57 (d, *J* = 9.2 Hz, 1H), 2.01-1.95 (m, 1H), 1.16-1.07 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 163.4, 160.7, 153.5, 143.7, 128.2, 128.0, 122.0, 120.4, 13.9, 10.5 ppm.

2-(*tert*-Butyl)-6-nitroquinazolin-4(3*H*)-one 3n. Light yellow solid, mp 263-265 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.31 (br, 1H), 8.68 (d, *J* = 2.4 Hz, 1H), 8.41 (dd, *J* = 9.2, 2.8 Hz, 1H), 7.71 (d, *J* = 9.2 Hz, 1H), 1.36 (s, 9H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 166.6, 161.5, 152.5, 144.5, 129.0, 128.1, 121.7, 120.6, 37.7, 27.6 ppm. Anal. Calcd. for $C_{12}H_{13}N_3O_3$: C, 58.30; H, 5.30; N, 16.99. Found: C, 58.49; H, 5.12; N, 16.78.

6-Nitro-2-(*p*-tolyl)quinazolin-4(3*H*)-one 3o. Light yellow solid, mp 301-303 °C (lit.[4] 304-305 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.92 (br, 1H), 8.80 (s, 1H), 8.52 (d, *J* = 8.8 Hz, 1H), 8.11 (d, *J* = 7.6 Hz, 2H), 7.89 (d, *J* = 8.8 Hz, 1H), 7.36 (d, *J* = 7.6 Hz, 2H), 2.38 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.0, 156.0, 153.6,

145.1, 143.1, 129.8, 129.7, 129.6, 128.9, 128.7, 122.5, 121.4, 21.5 ppm.

6-Fluoro-2-phenylquinazolin-4(3*H*)-one 3p. White solid, mp 282-284 °C (lit.[5] 280-282 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.60 (br, 1H), 8.22-8.16 (m, 3H), 7.61-7.33 (m, 5H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 165.8 (d, $^{1}J_{C\text{-}F}$ = 249.4 Hz), 161.5, 153.7, 150.8 (d, $^{3}J_{C\text{-}F}$ = 14.2 Hz), 132.4, 131.6, 128.9 (d, $^{3}J_{C\text{-}F}$ = 10.9 Hz), 128.5, 127.8, 118.0, 115.0 (d, $^{2}J_{C\text{-}F}$ = 23.4 Hz), 122.4 (d, $^{2}J_{C\text{-}F}$ = 20.9 Hz) ppm.

2-Cyclopropyl-6-fluoroquinazolin-4(3*H*)-one 3q. White solid, mp 252-253 °C (lit.[6] 253-254 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.51 (br, 1H), 8.13-8.09 (m, 1H), 7.27-7.21 (m, 2H), 2.01-1.95 (m, 1H), 1.14-1.05 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 165.7 (d, $^{1}J_{C\text{-}F}$ = 248.7 Hz), 160.8, 160.6, 151.3 (d, $^{3}J_{C\text{-}F}$ = 13.3 Hz), 128.8 (d, $^{3}J_{C\text{-}F}$ = 10.9 Hz), 117.6, 113.7 (d, $^{2}J_{C\text{-}F}$ = 23.4 Hz), 113.3 (d, $^{2}J_{C\text{-}F}$ = 21.4 Hz), 13.4, 9.7 ppm.

2-(*tert*-Butyl)-6-fluoroquinazolin-4(3*H*)-one 3r. White solid, mp 222-224 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.96 (br, 1H), 8.15-8.12 (m, 1H), 7.37-7.31 (m, 2H), 1.34 (s, 9H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 166.2 (d, $^{1}J_{C\text{-}F}$ = 249.2 Hz), 164.7, 162.0, 151.0 (d, $^{3}J_{C\text{-}F}$ = 13.1 Hz), 129.1 (d, $^{3}J_{C\text{-}F}$ = 10.9 Hz), 118.1, 115.2 (d, $^{2}J_{C\text{-}F}$ = 23.4 Hz), 112.7 (d, $^{2}J_{C\text{-}F}$ = 21.3 Hz), 37.8, 28.2 ppm. Anal. Calcd. for $C_{12}H_{13}N_2OF$: C, 65.44; H, 5.95; N, 12.71. Found: C, 65.18; H, 5.73; N, 12.58.

6-Fluoro-2-(*p*-tolyl)quinazolin-4(3*H*)-one 3s. White solid, mp 301-303 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.53 (br, 1H), 8.22-8.17 (m, 1H), 8.09 (d, *J* = 8.0 Hz, 2H), 7.49-7.34 (m, 4H), 2.40 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 165.8 (d, $^{1}J_{C\text{-}F}$ = 249.5 Hz), 161.5, 153.6, 150.9 (d, $^{3}J_{C\text{-}F}$ = 13.0 Hz), 131.7, 131.4, 129.2,

128.9 (d, $^{3}J_{C\text{-}F}$ = 10.6 Hz), 127.8, 117.9, 114.8 (d, $^{2}J_{C\text{-}F}$ = 23.3 Hz), 112.2 (d, $^{2}J_{C\text{-}F}$ = 23.6 Hz), 20.9 ppm. Anal. Calcd. for $C_{15}H_{11}N_2OF$: C, 70.86; H, 4.36; N, 11.01. Found: C, 70.59; H, 4.55; N, 10.87.

6-Methyl-2-phenylquinazolin-4(3*H*)-one 3t. White solid, mp 236-238 °C (lit.[2] 238-240 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.46 (br, 1H), 8.16 (d, *J* = 7.2 Hz, 2H), 7.94 (s, 1H), 7.63-7.52 (m, 5H), 2.44 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.1, 151.4, 146.7, 136.3, 135.8, 132.8, 131.2, 128.5, 127.6, 127.3, 125.2, 120.7, 20.8 ppm.

2-(*tert*-Butyl)-6-methylquinazolin-4(3*H*)-one 3u. White solid, mp 205-207 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.79 (br, 1H), 7.88 (s, 1H), 7.58 (dd, *J* = 8.4, 2.0 Hz, 1H), 7.51 (d, *J* = 8.0 Hz, 1H), 2.42 (s, 3H), 1.35 (s, 9H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 167.4, 167.0, 151.6, 140.9, 140.7, 132.4, 130.2, 125.6, 42.3, 33.0, 26.0 ppm. Anal. Calcd. for $C_{13}H_{16}N_2O$: C, 72.20; H, 7.46; N, 12.95. Found: C, 72.38; H, 7.27; N, 12.74.

6-Methyl-2-(*p*-tolyl)quinazolin-4(3*H*)-one 3v. White solid, mp 269-271 °C (lit.[7] 271-272 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.37 (br, 1H), 8.07 (d, *J* = 7.2 Hz, 2H), 7.93 (s, 1H), 7.63-7.61 (m, 2H), 7.33 (d, *J* = 7.2 Hz, 2H), 2.44 (s, 3H), 2.37 (s, 3H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.1, 151.4, 146.8, 135.8, 131.7, 131.5, 129.1, 128.6, 127.5, 127.3, 125.2, 120.6, 20.9, 20.8 ppm.

7-Methyl-2-phenylquinazolin-4(3*H*)-one 3w. White solid, mp 238-240 °C (lit.[4] 240-241 °C). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.41 (br, 1H), 8.17 (d, *J* = 7.6 Hz, 2H), 8.03 (d, *J* = 8.0 Hz, 1H), 7.57-7.51 (m, 4H), 7.32 (d, *J* = 8.0 Hz, 1H), 2.46 (s, 3H)

ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 162.1, 152.3, 148.8, 145.0, 132.8, 131.3, 128.6, 128.0, 127.7, 127.1, 125.7, 118.5, 21.3 ppm.

2-Cyclopropyl-7-methyquinazolin-4(3*H*)-one 3x. White solid, mp 235-237 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 12.29 (br, 1H), 7.92 (d, *J* = 8.0 Hz, 1H), 7.27 (s, 1H), 7.19 (d, *J* = 8.0 Hz, 1H), 2.39 (s, 3H), 1.99-1.92 (m, 1H), 1.08-0.99 (m, 4H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 161.5, 158.9, 149.2, 144.6, 126.6, 126.1, 125.6, 118.2, 21.2, 13.3, 9.3 ppm. Anal. Calcd. for $C_{12}H_{12}N_2O$: C, 71.98; H, 6.04; N, 13.98. Found: C, 71.74; H, 5.87; N, 13.79.

2-(*tert*-Butyl)-7-methylquinazolin-4(3*H*)-one 3y. White solid, mp 208-210 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 11.75 (br, 1H), 7.96 (d, *J* = 7.6 Hz, 1H), 7.42 (s, 1H), 7.27 (d, *J* = 8.0 Hz, 1H), 2.42 (s, 3H), 1.33 (s, 9H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6): δ = 167.4, 166.9, 153.2, 149.4, 132.3, 131.7, 130.2, 123.0, 41.9, 32.5, 26.0 ppm. Anal. Calcd. for $C_{13}H_{16}N_2O$: C, 72.20; H, 7.46; N, 12.95. Found: C, 71.97; H, 7.21; N, 12.79.

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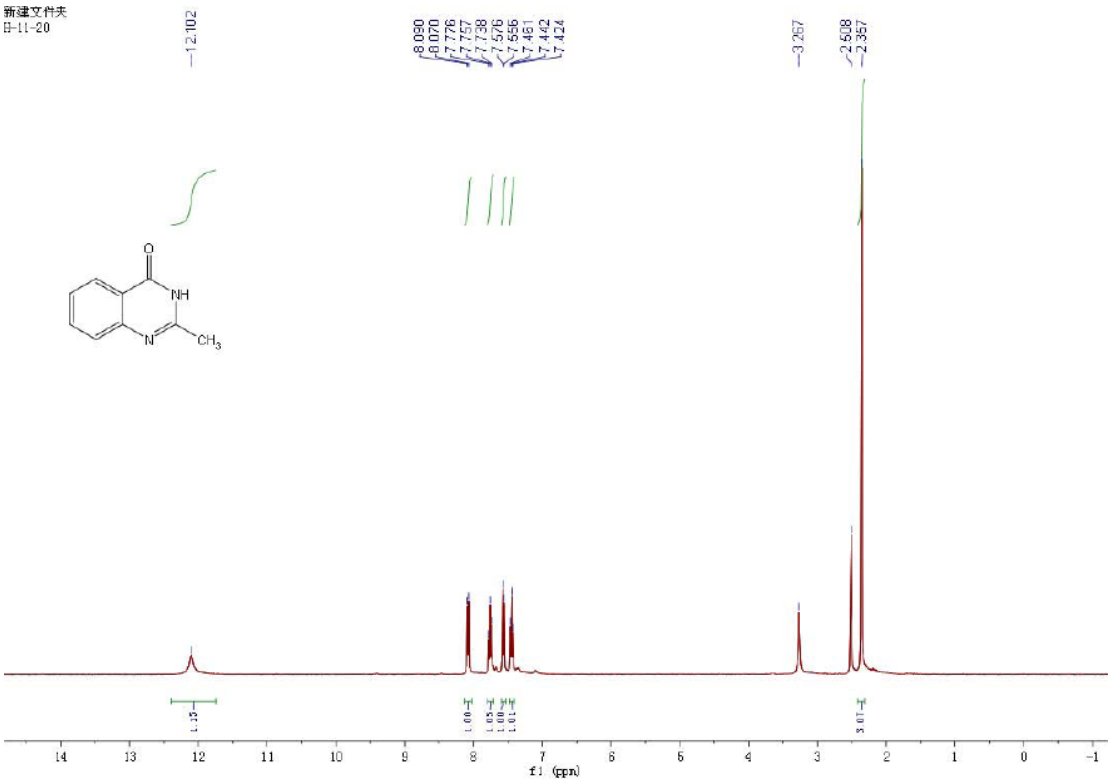
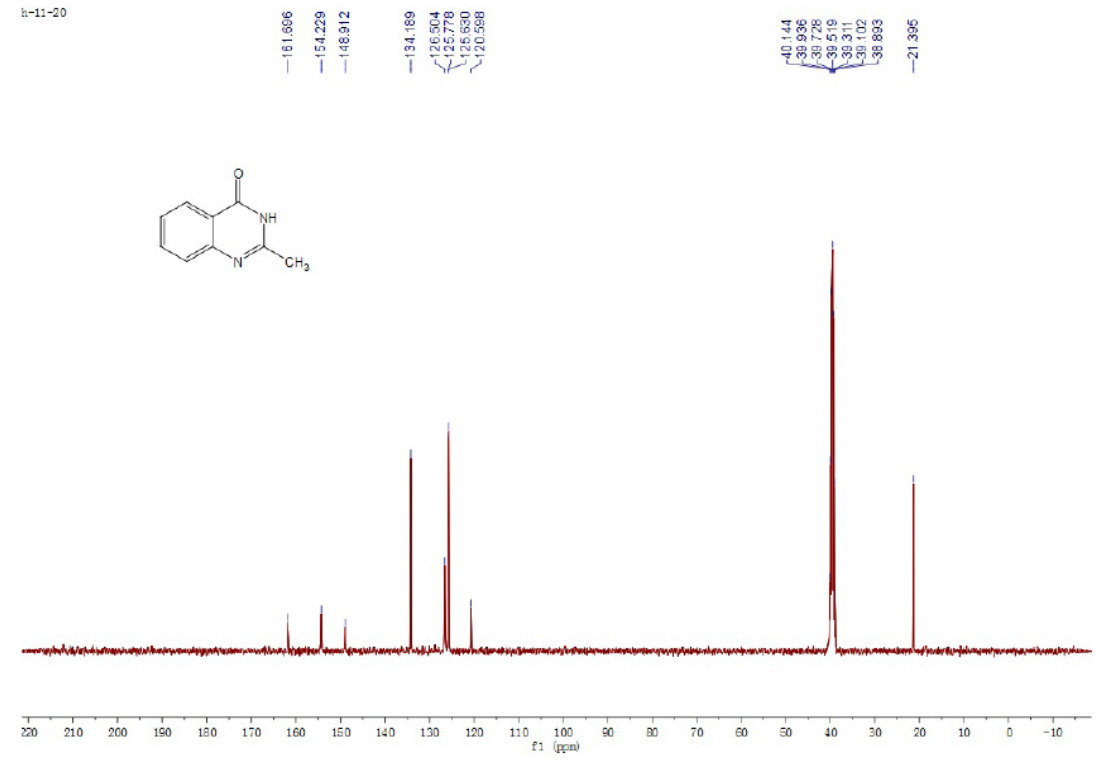
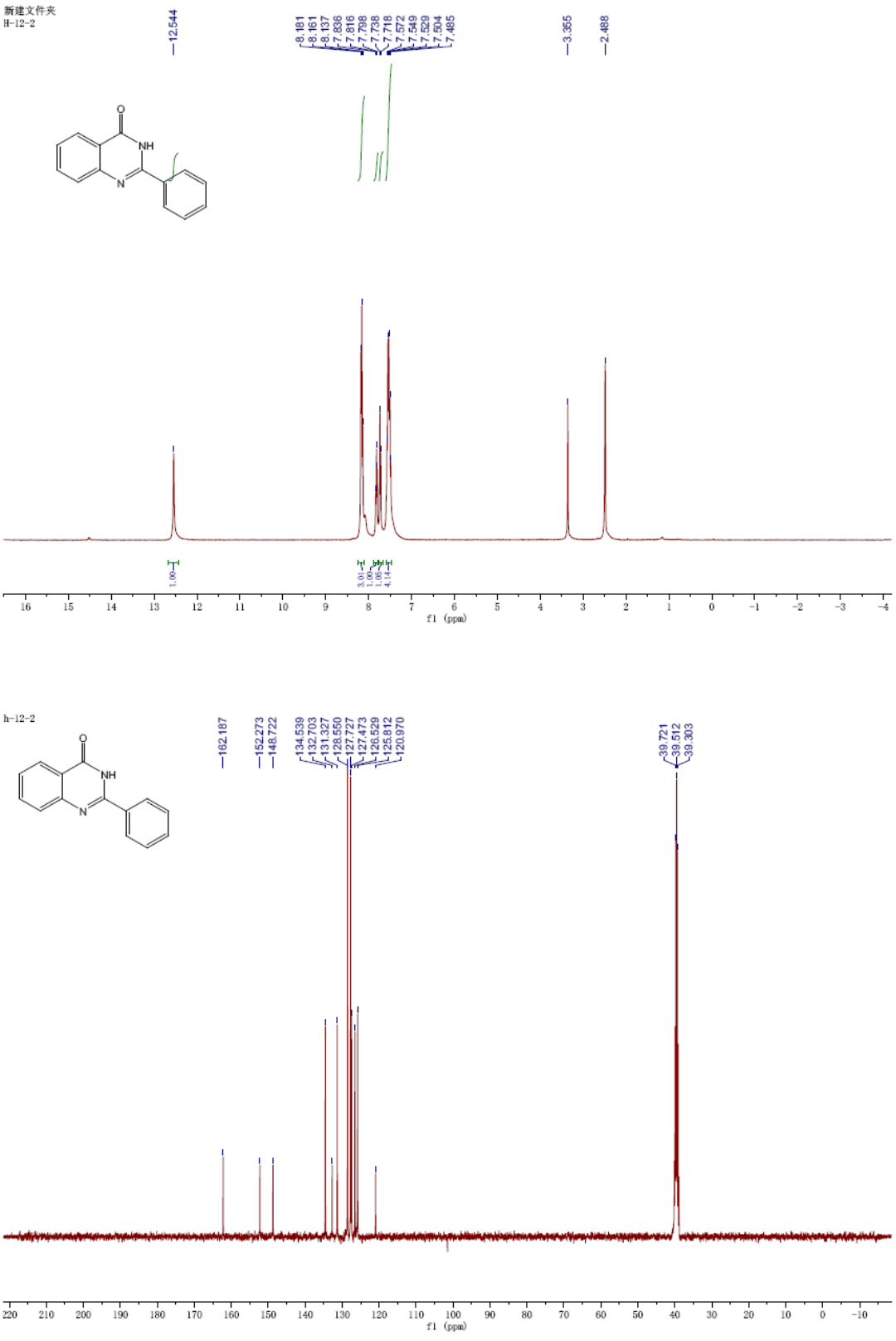
^{1}H NMR and ^{13}C NMR spectra of compounds 3a-3y.

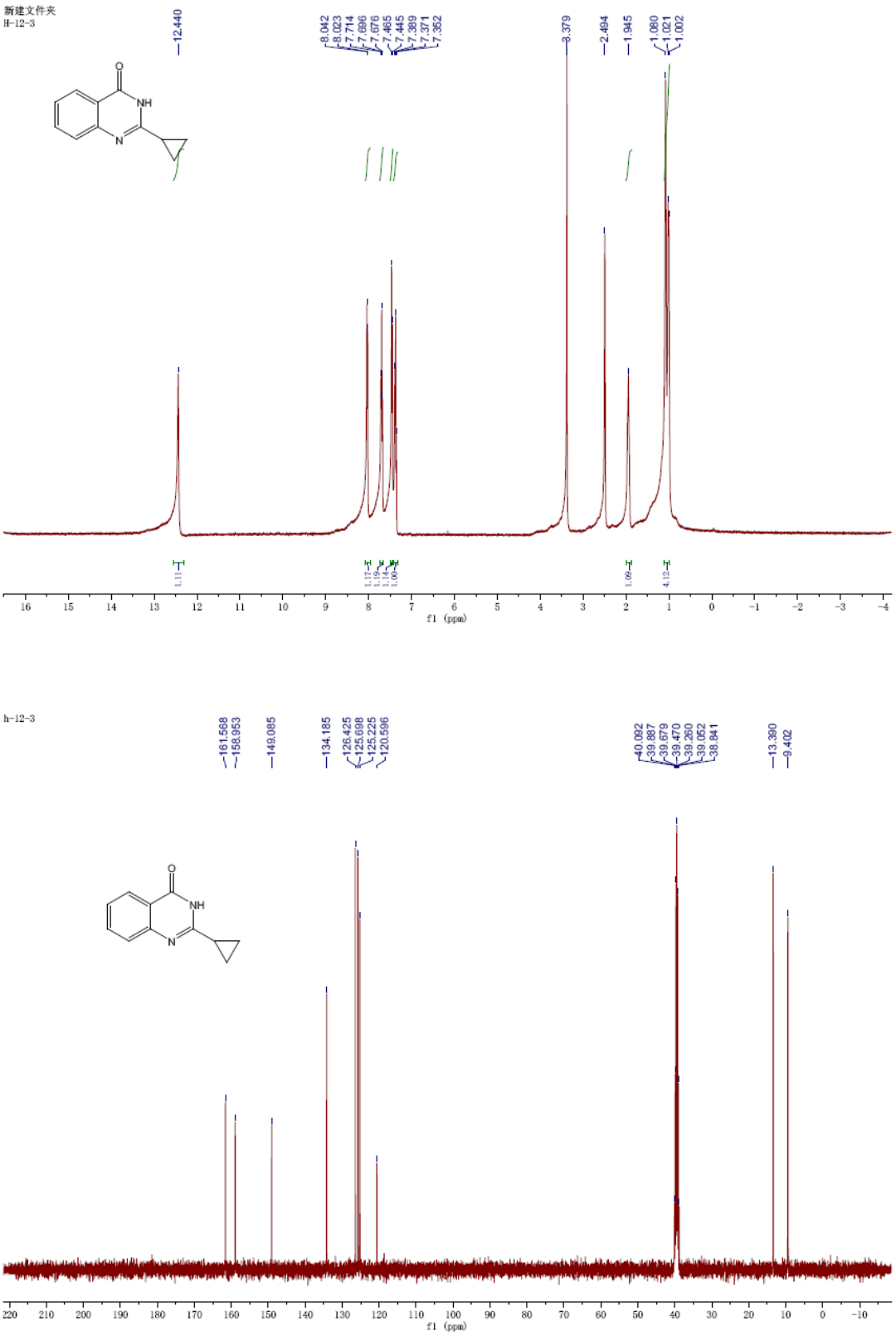




^{1}H NMR and ^{13}C NMR spectra of compound **3a**



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



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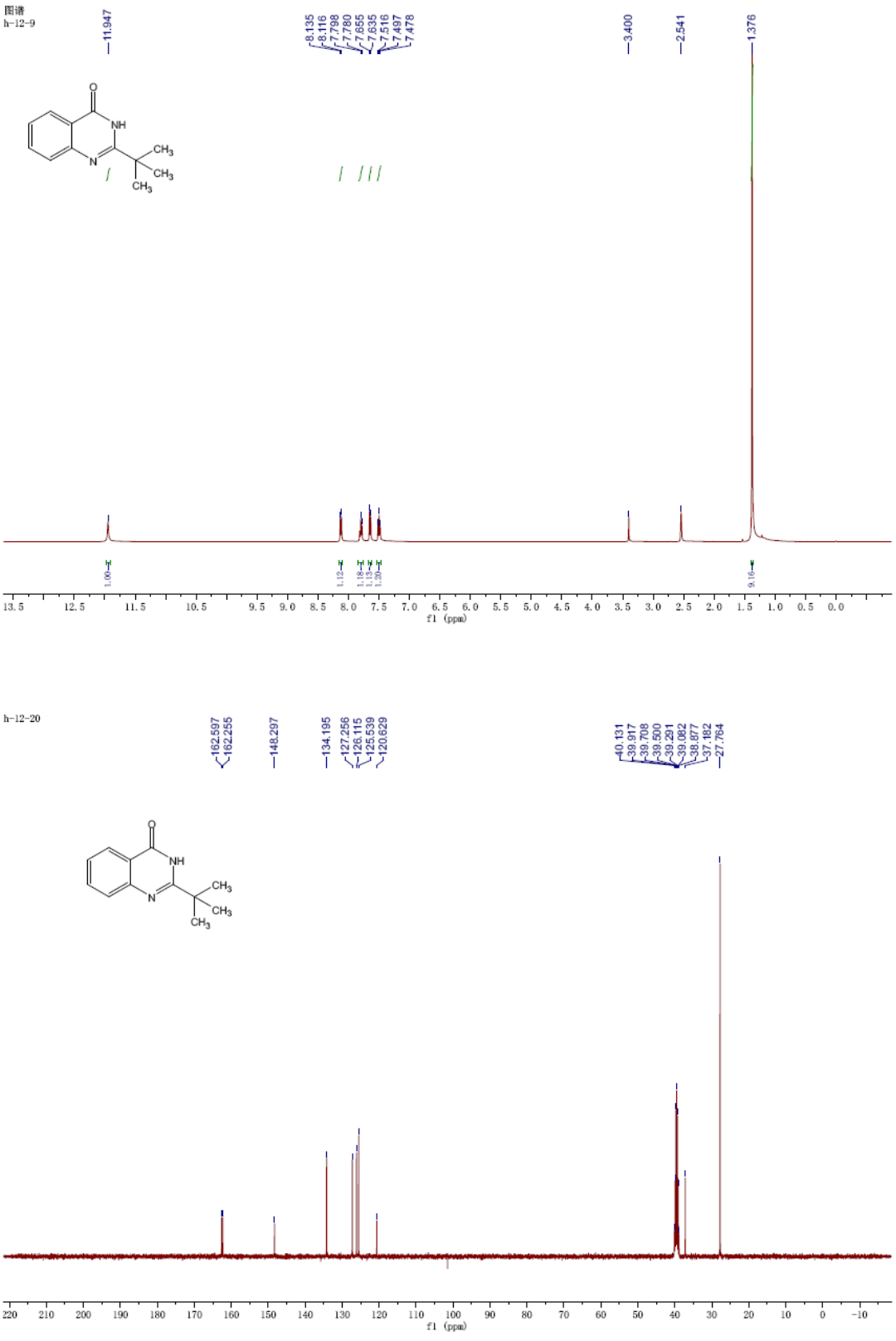

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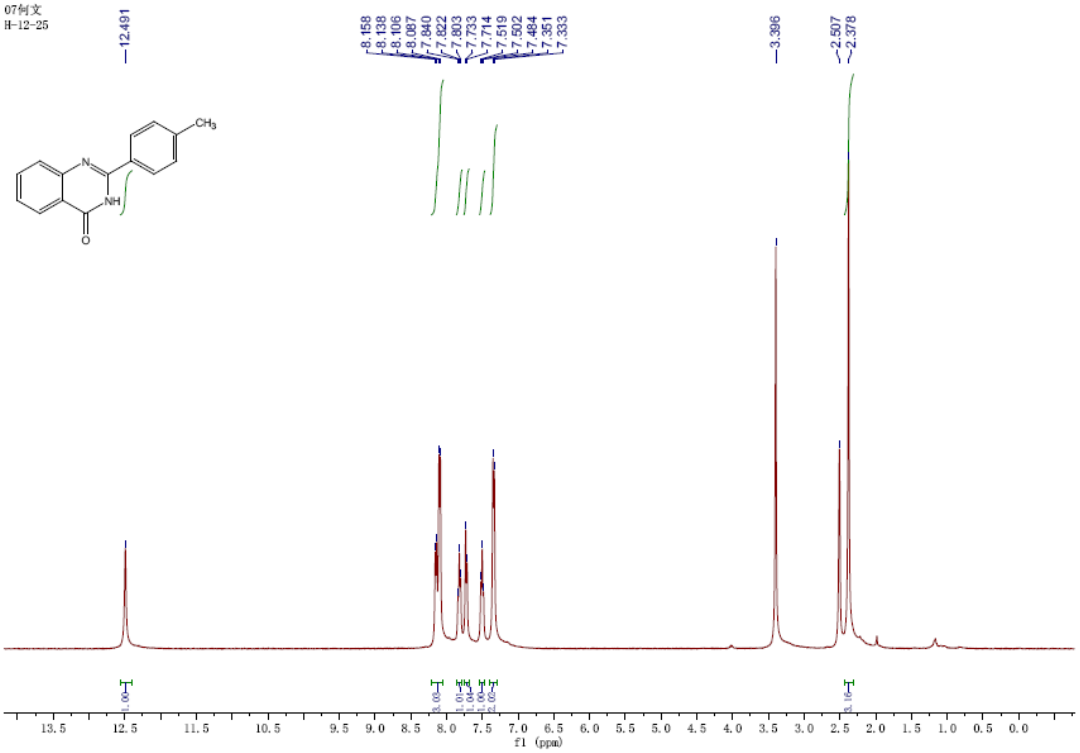
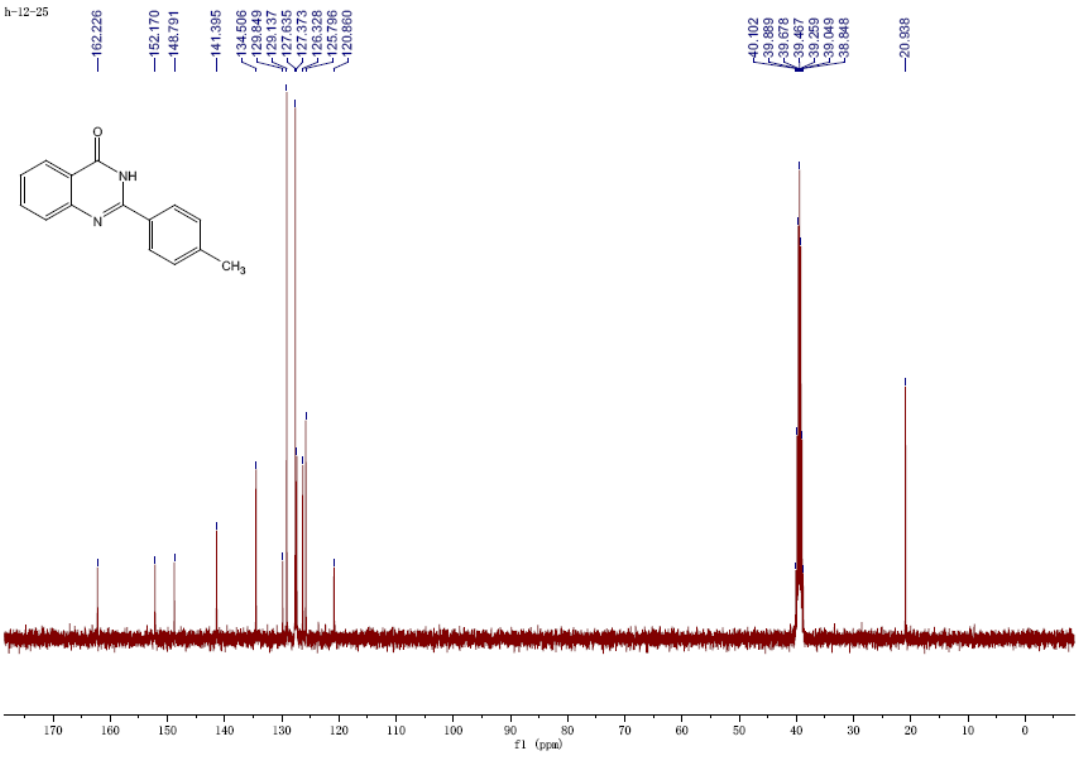
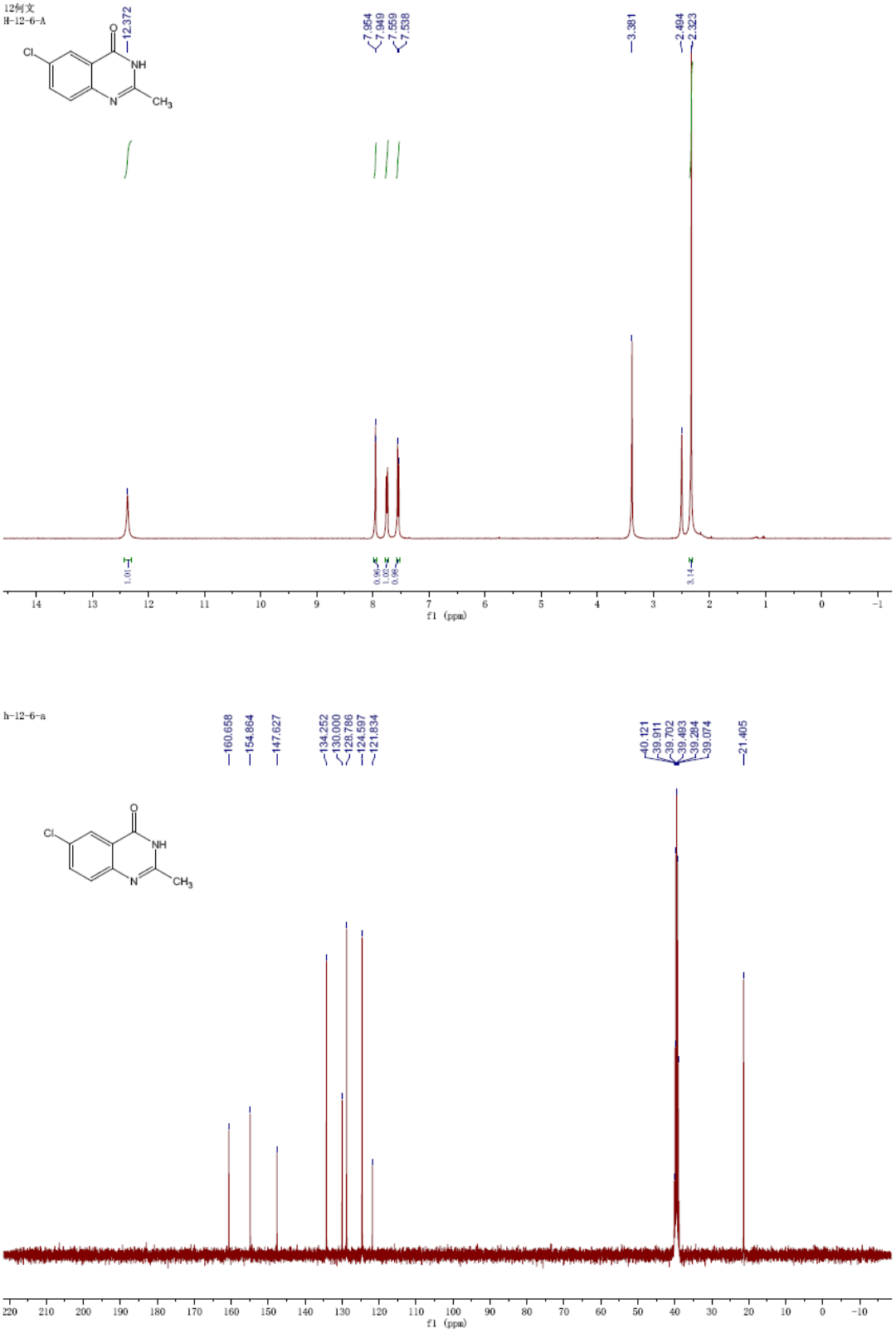
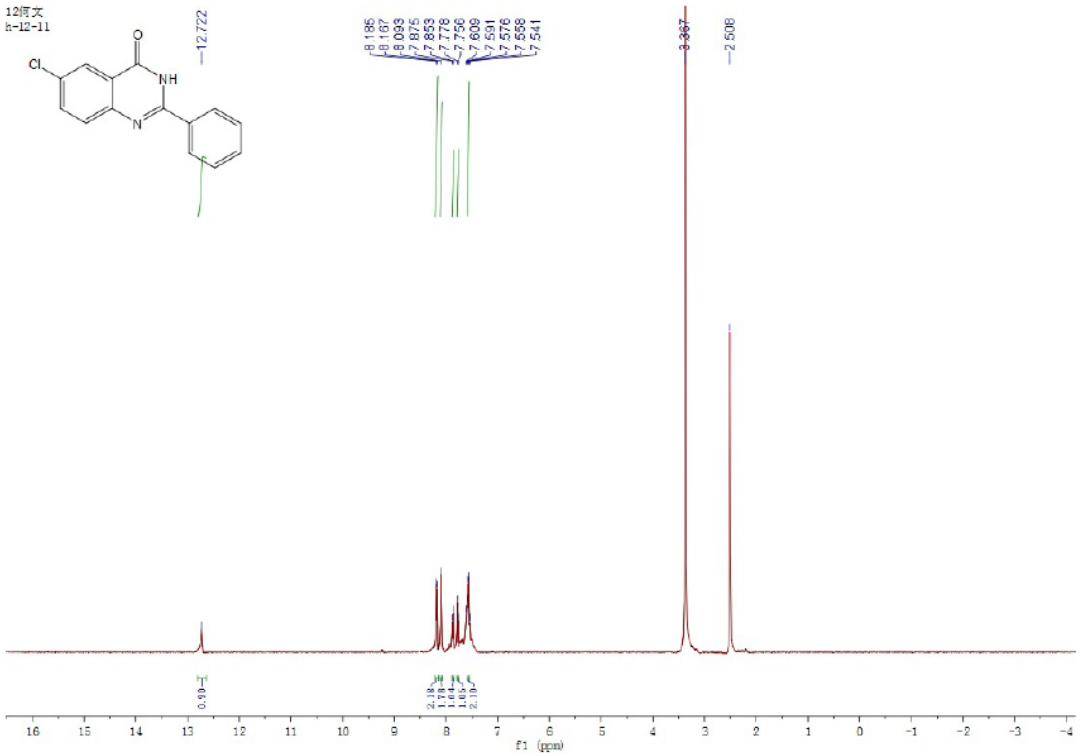


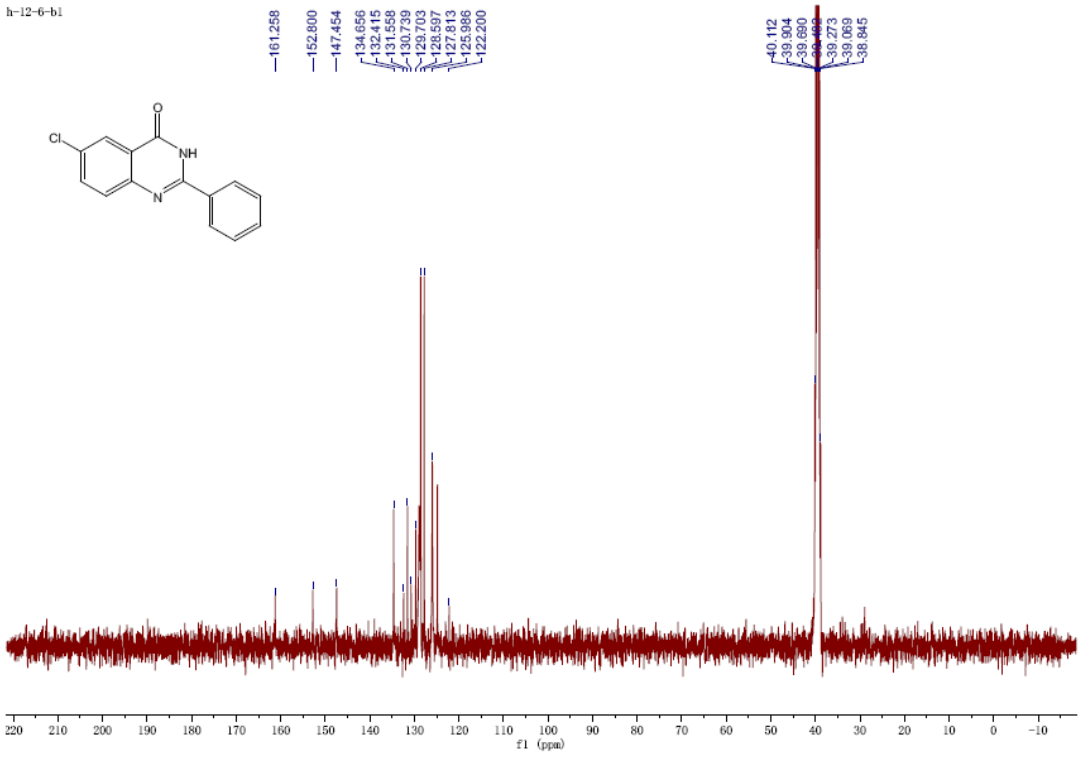


^{1}H NMR and ^{13}C NMR spectra of compound **3e**

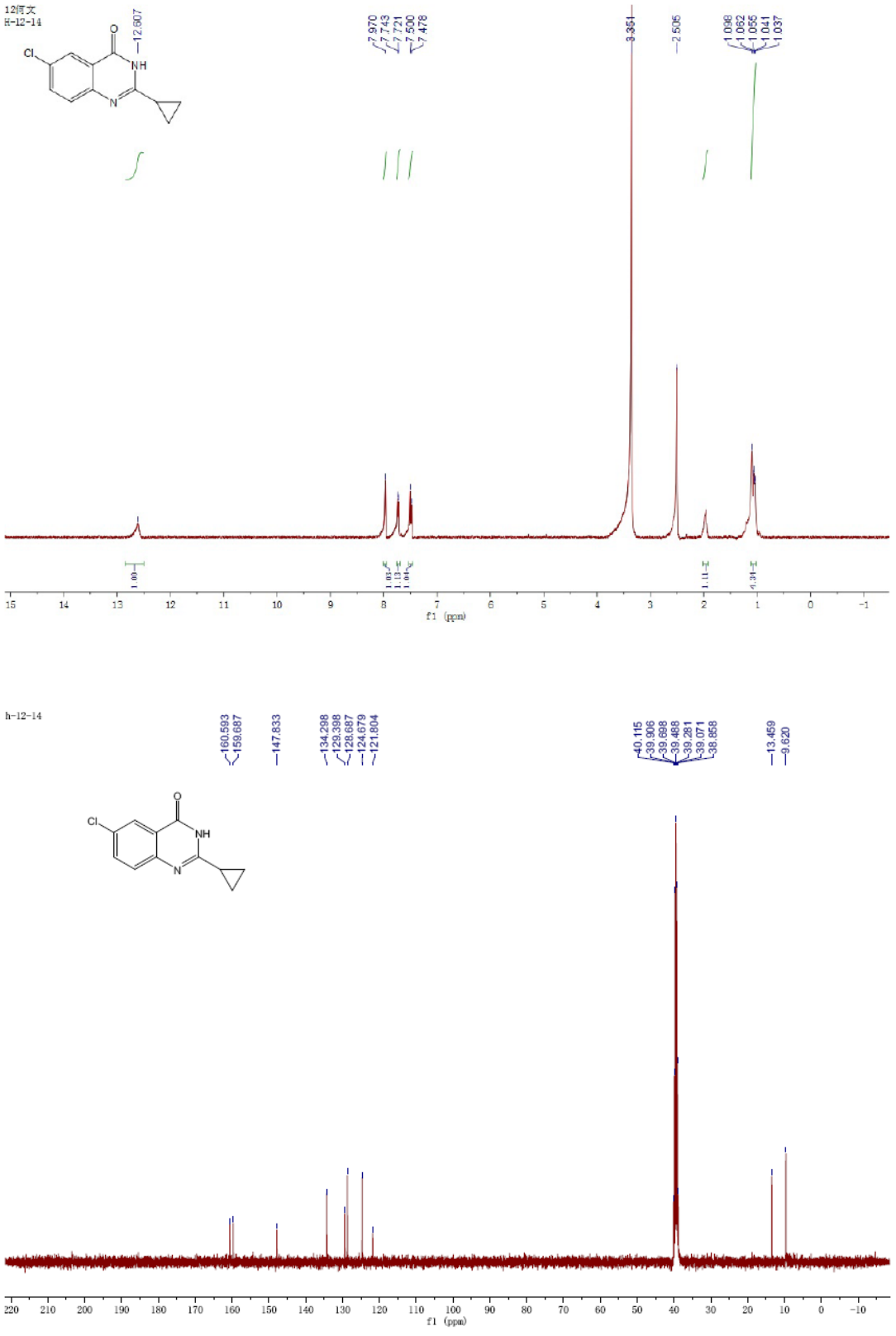


^{1}H NMR and ^{13}C NMR spectra of compound **3f**

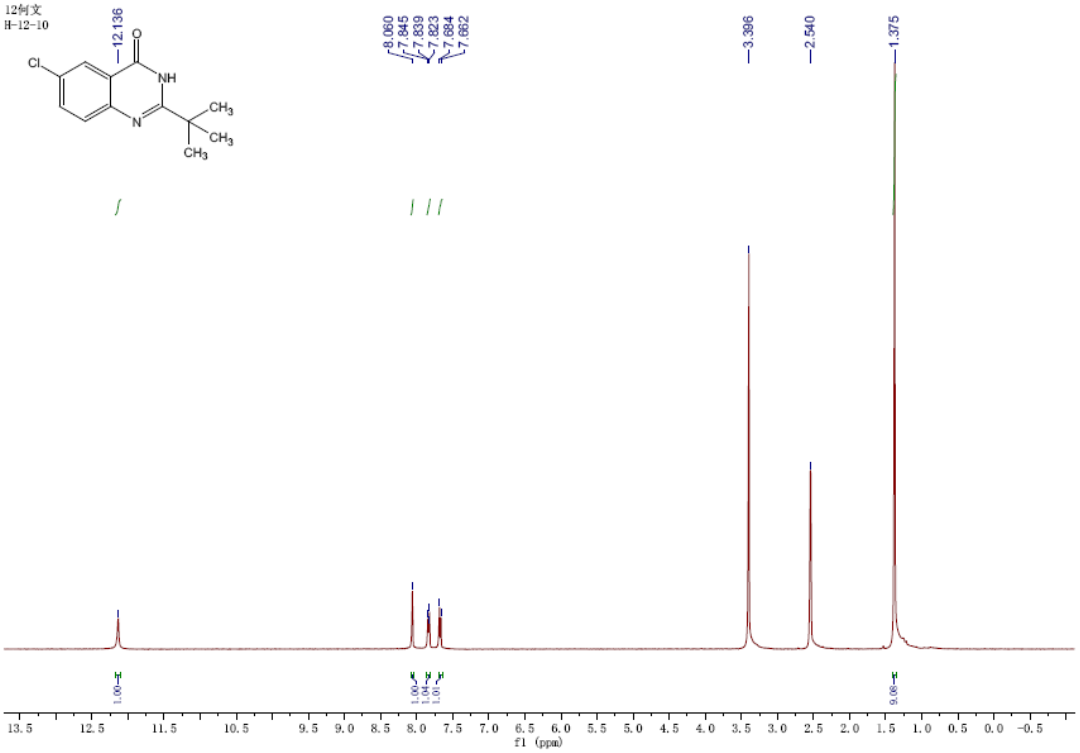


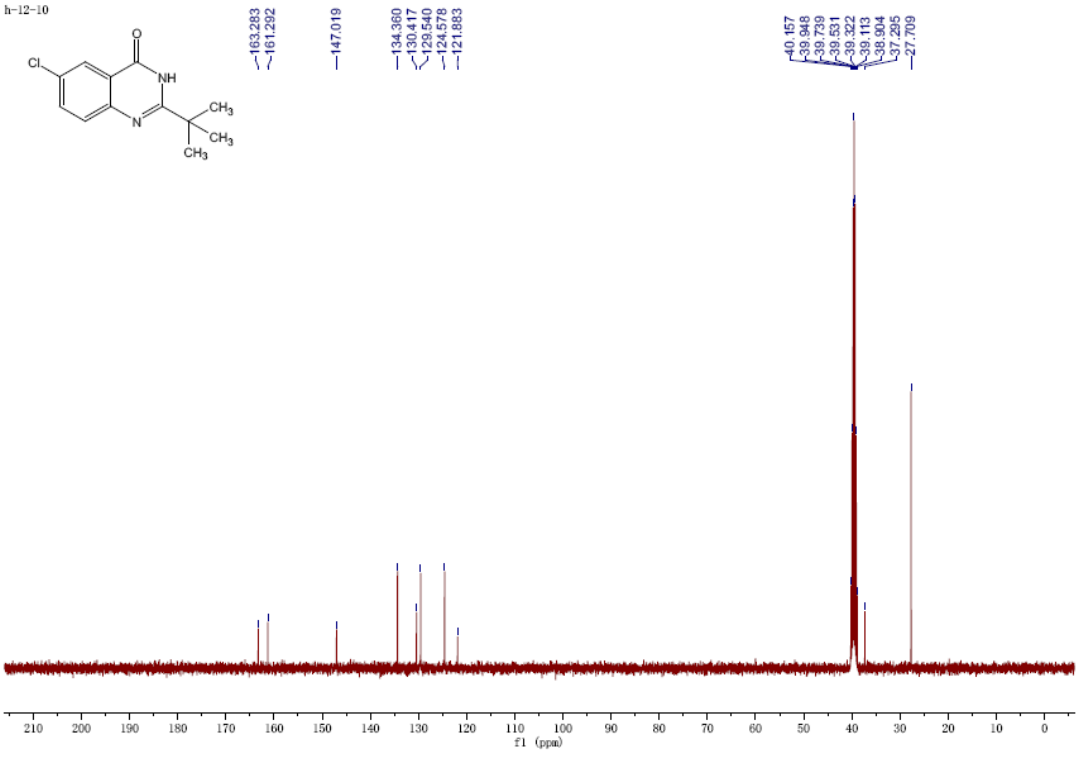


^{1}H NMR and ^{13}C NMR spectra of compound **3g**

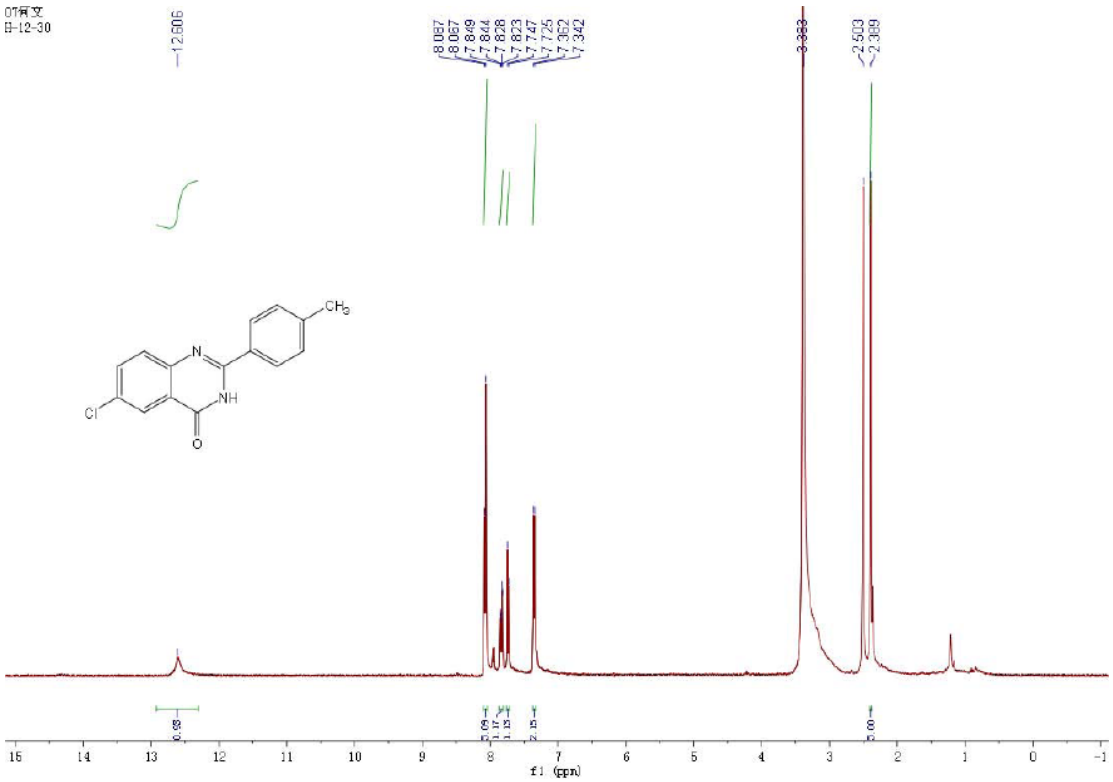


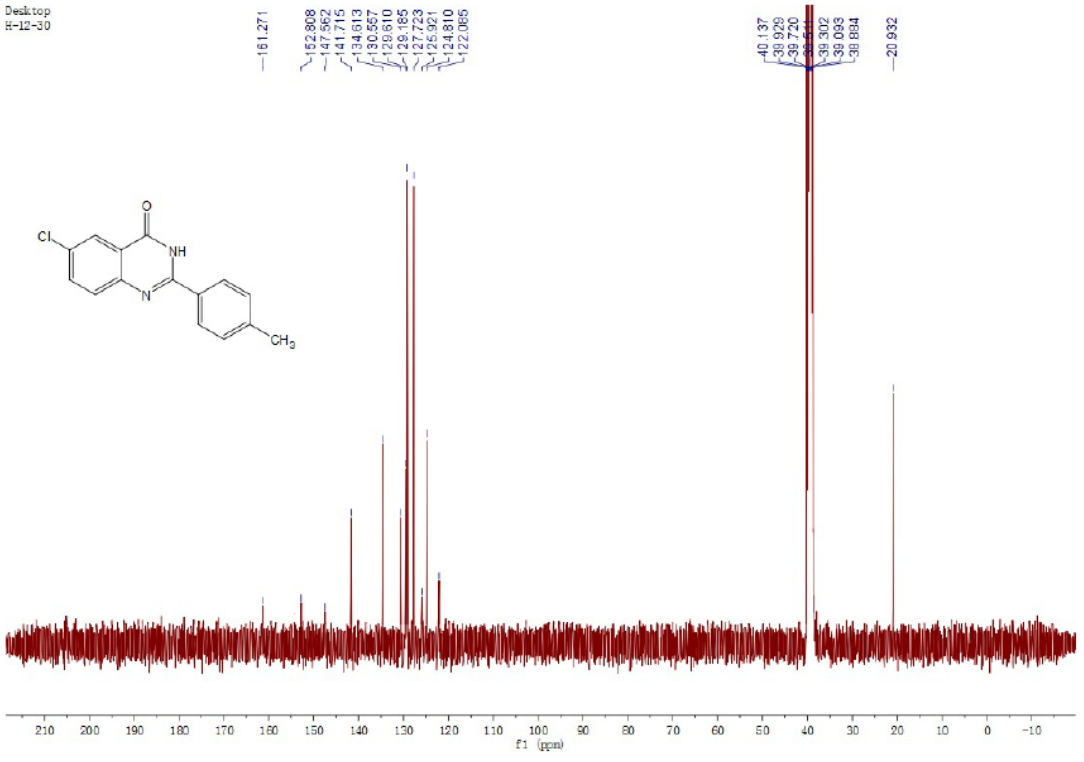
^{1}H NMR and ^{13}C NMR spectra of compound **3h**



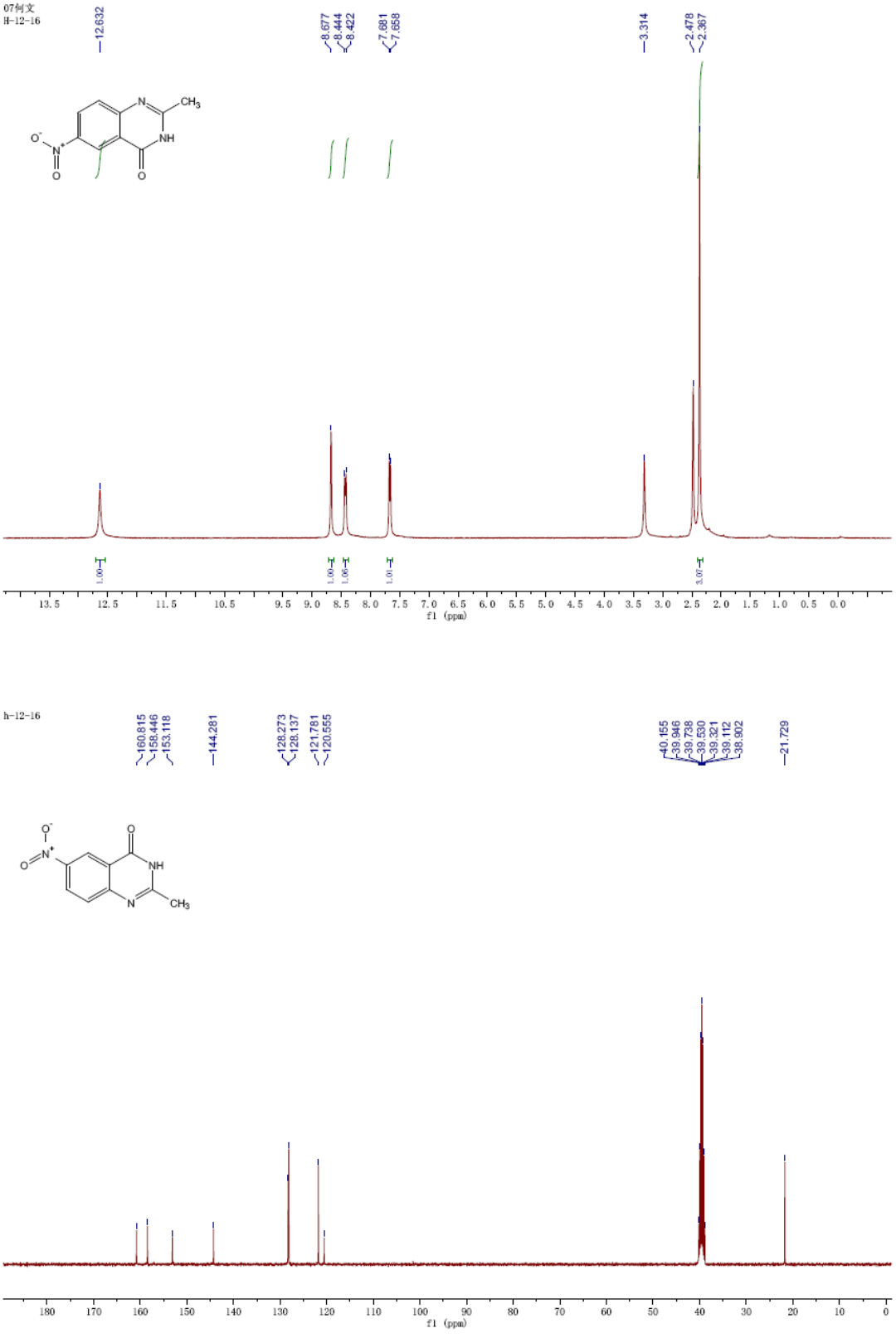


^{1}H NMR and ^{13}C NMR spectra of compound **3i**

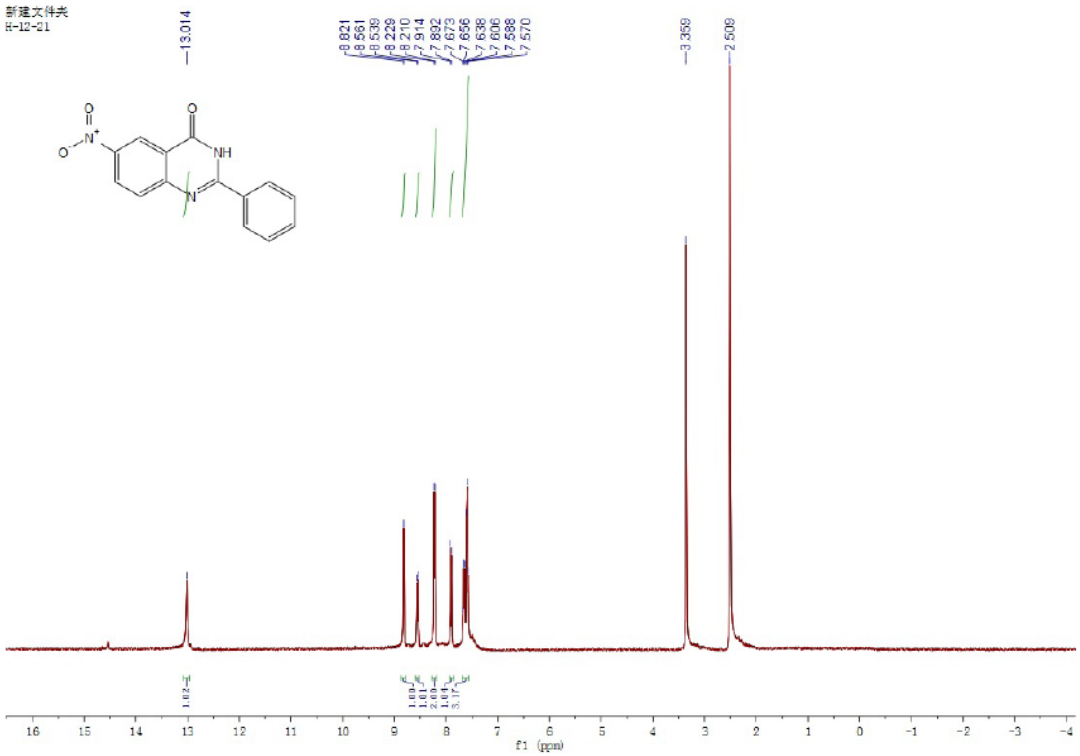


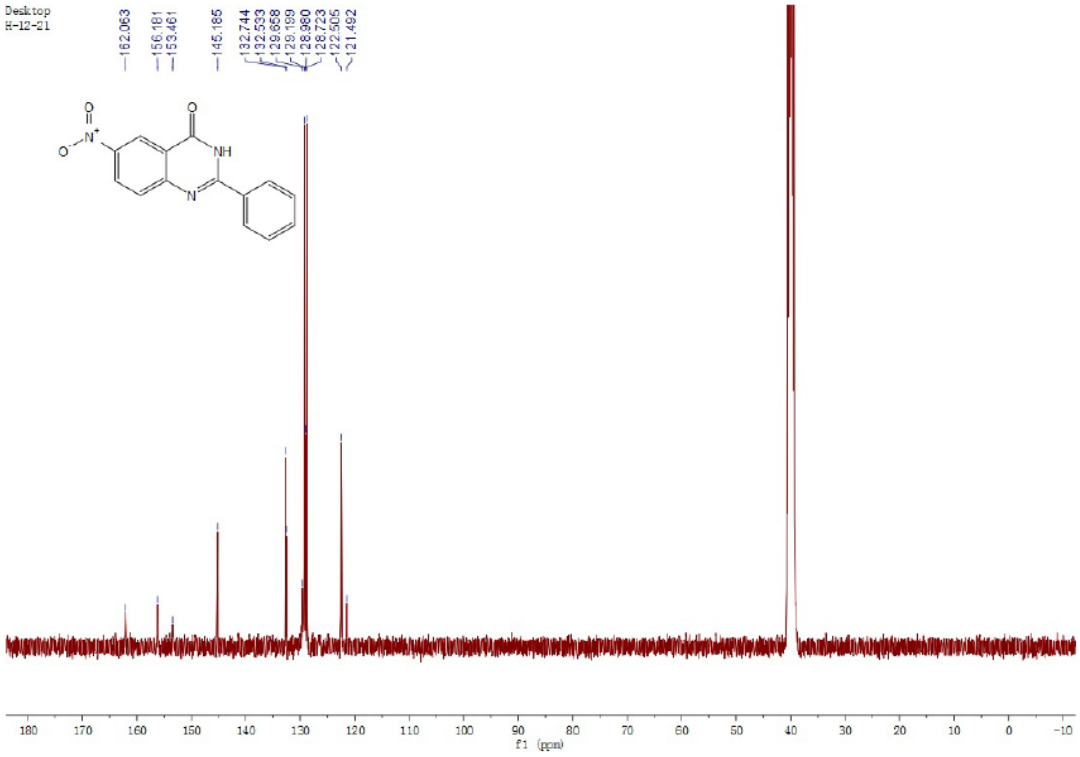


^{1}H NMR and ^{13}C NMR spectra of compound **3j**

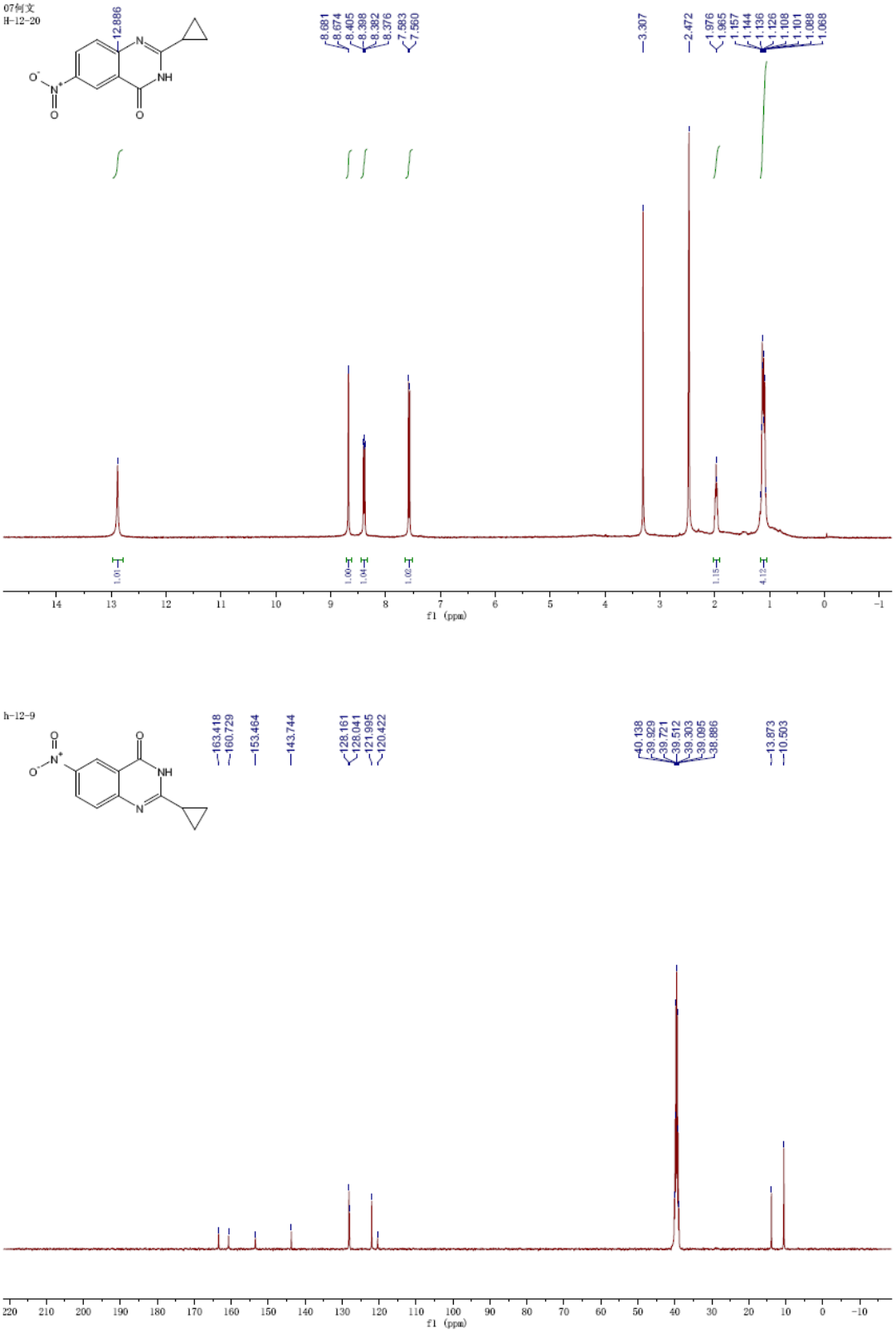


^{1}H NMR and ^{13}C NMR spectra of compound **3k**

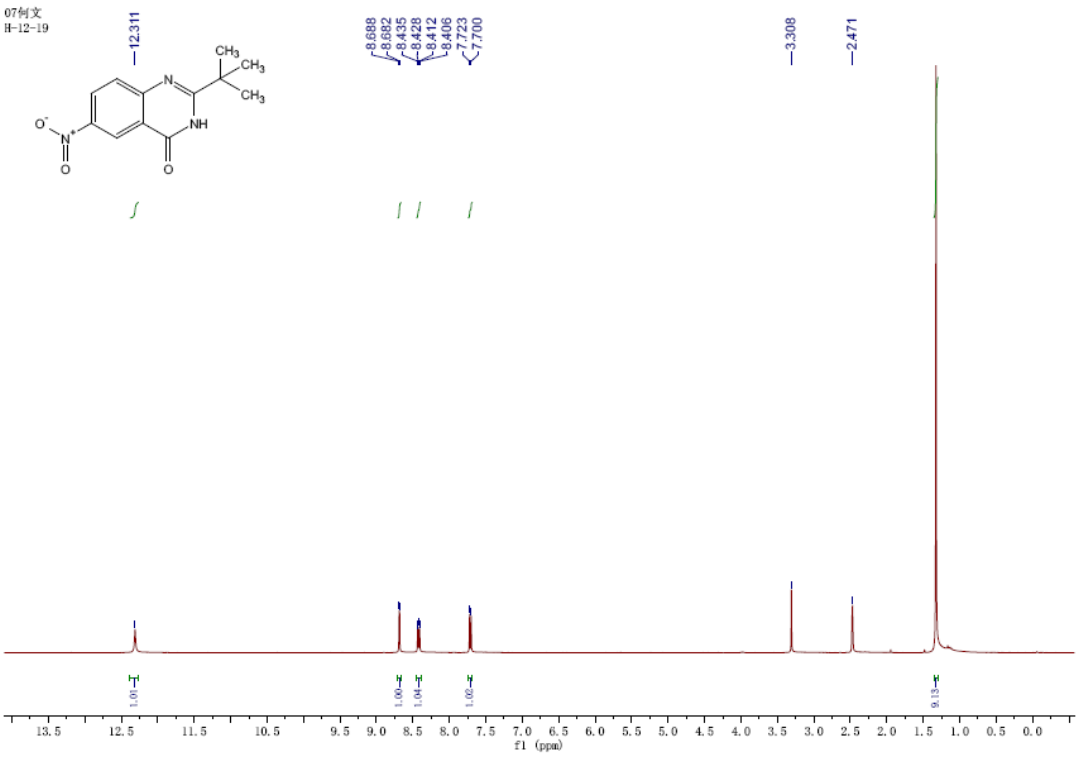


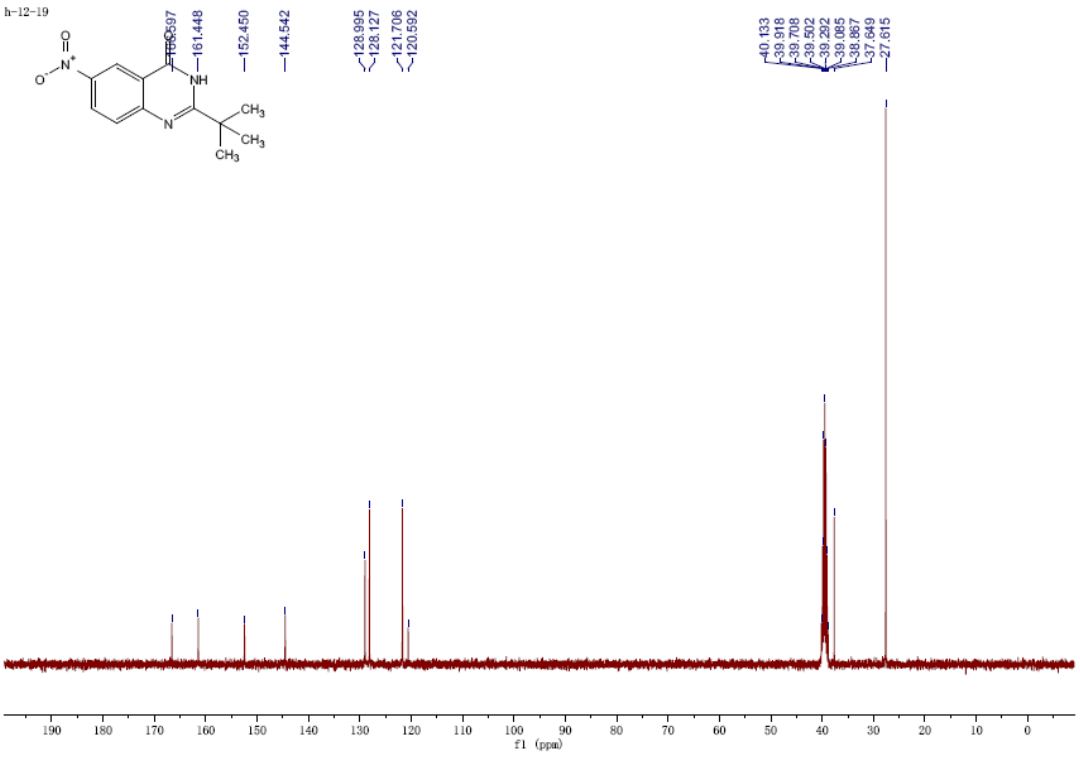


^{1}H NMR and ^{13}C NMR spectra of compound **3l**

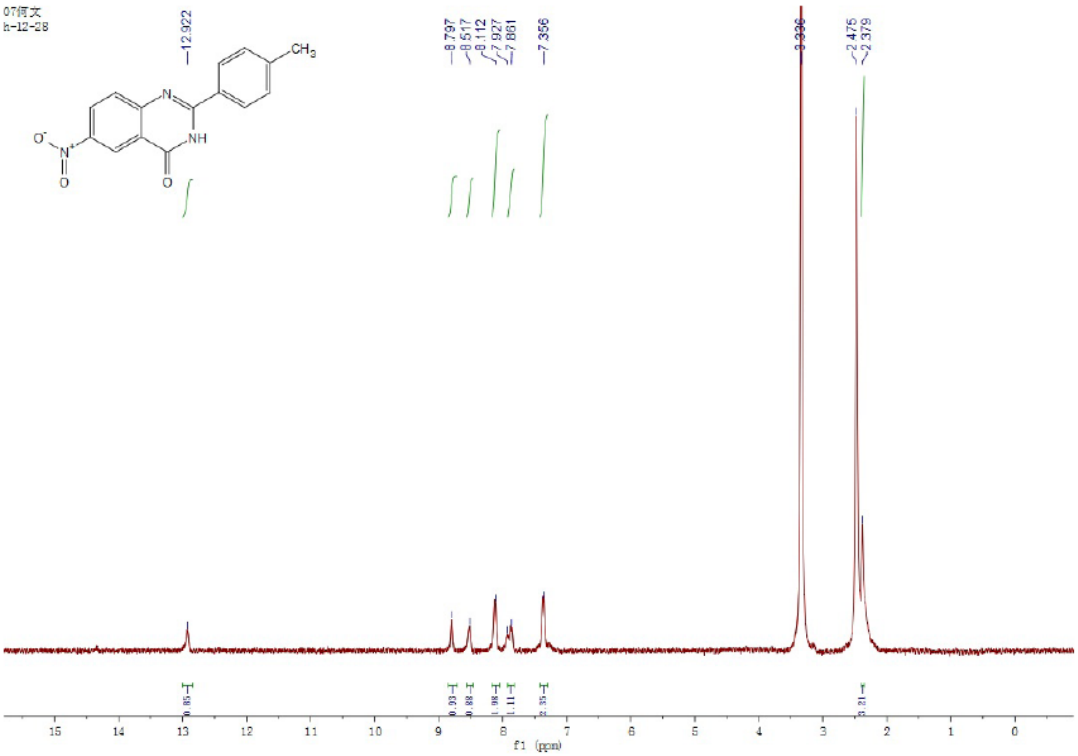


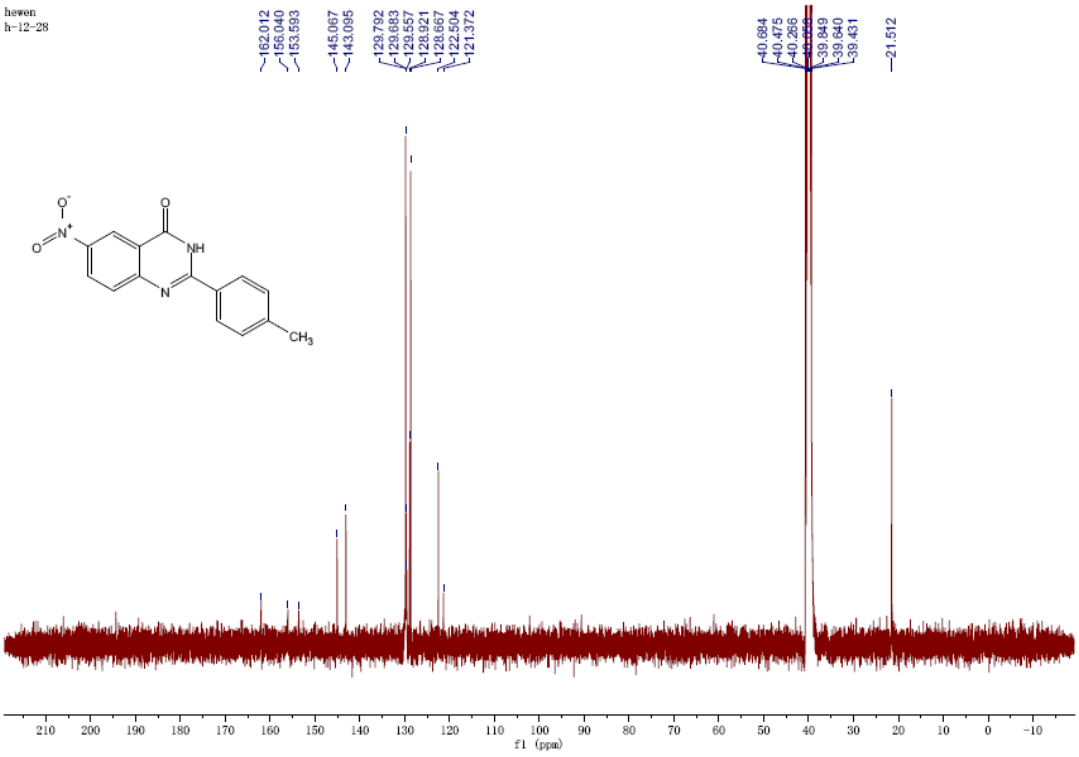
^{1}H NMR and ^{13}C NMR spectra of compound **3m**



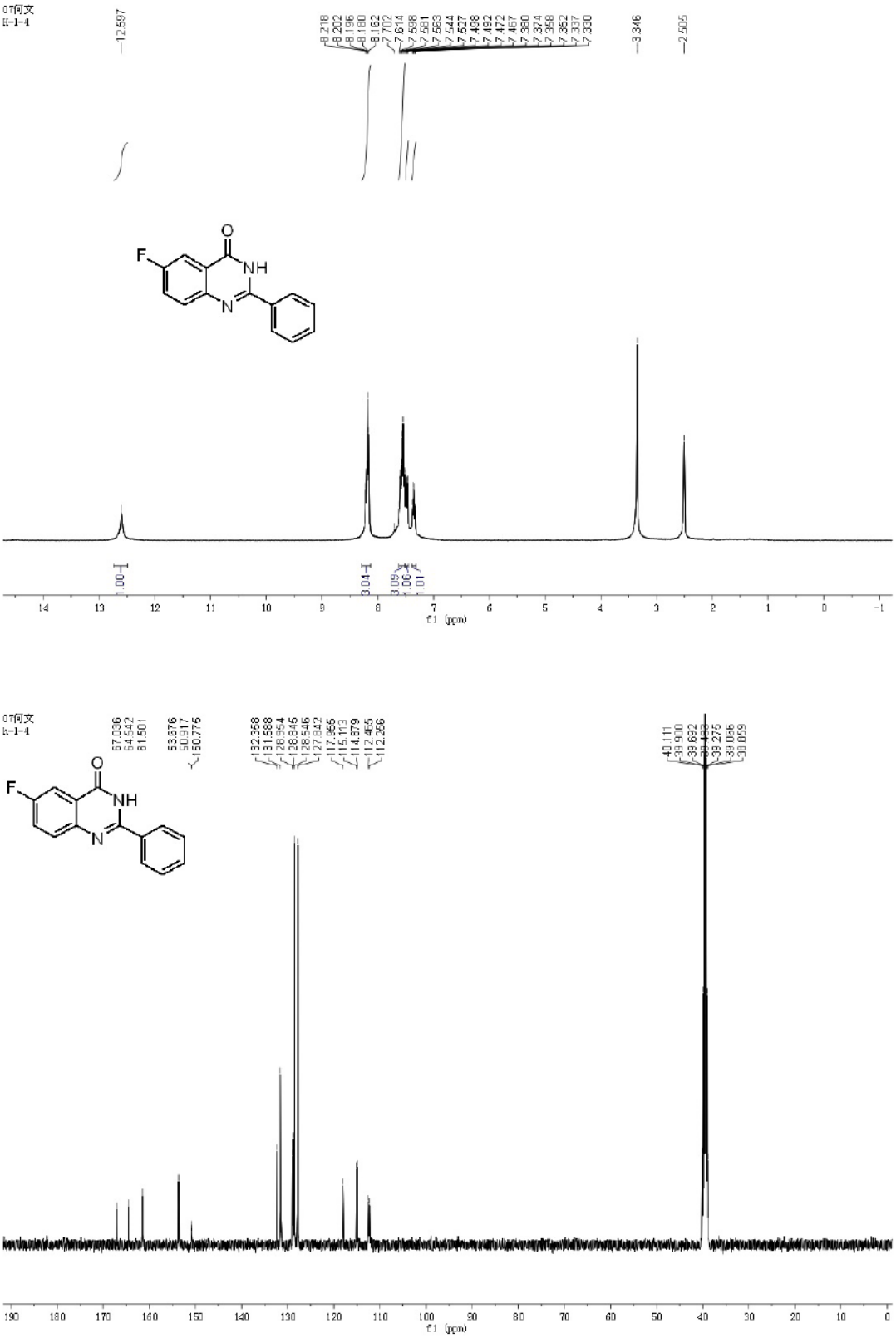


^{1}H NMR and ^{13}C NMR spectra of compound **3n**

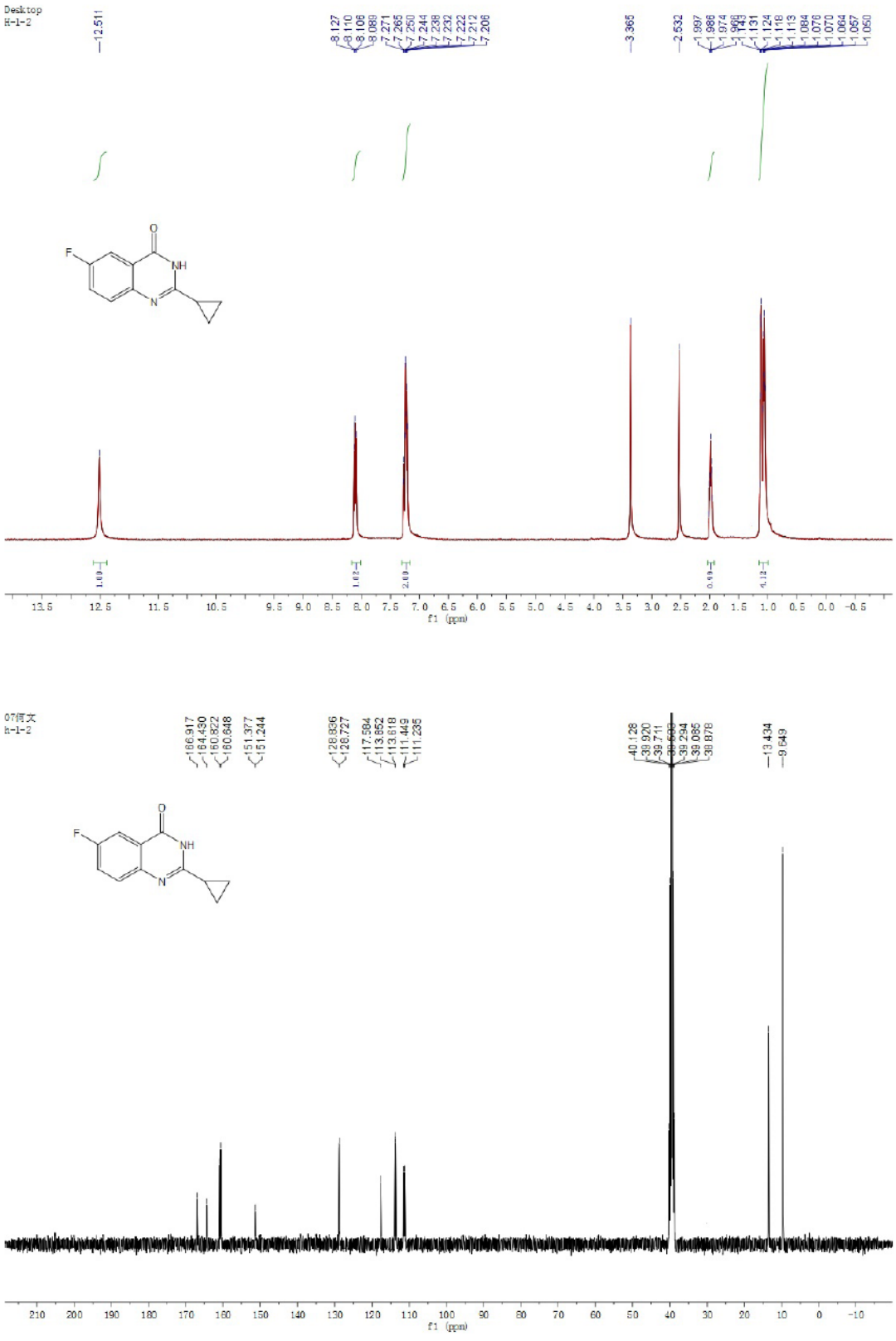




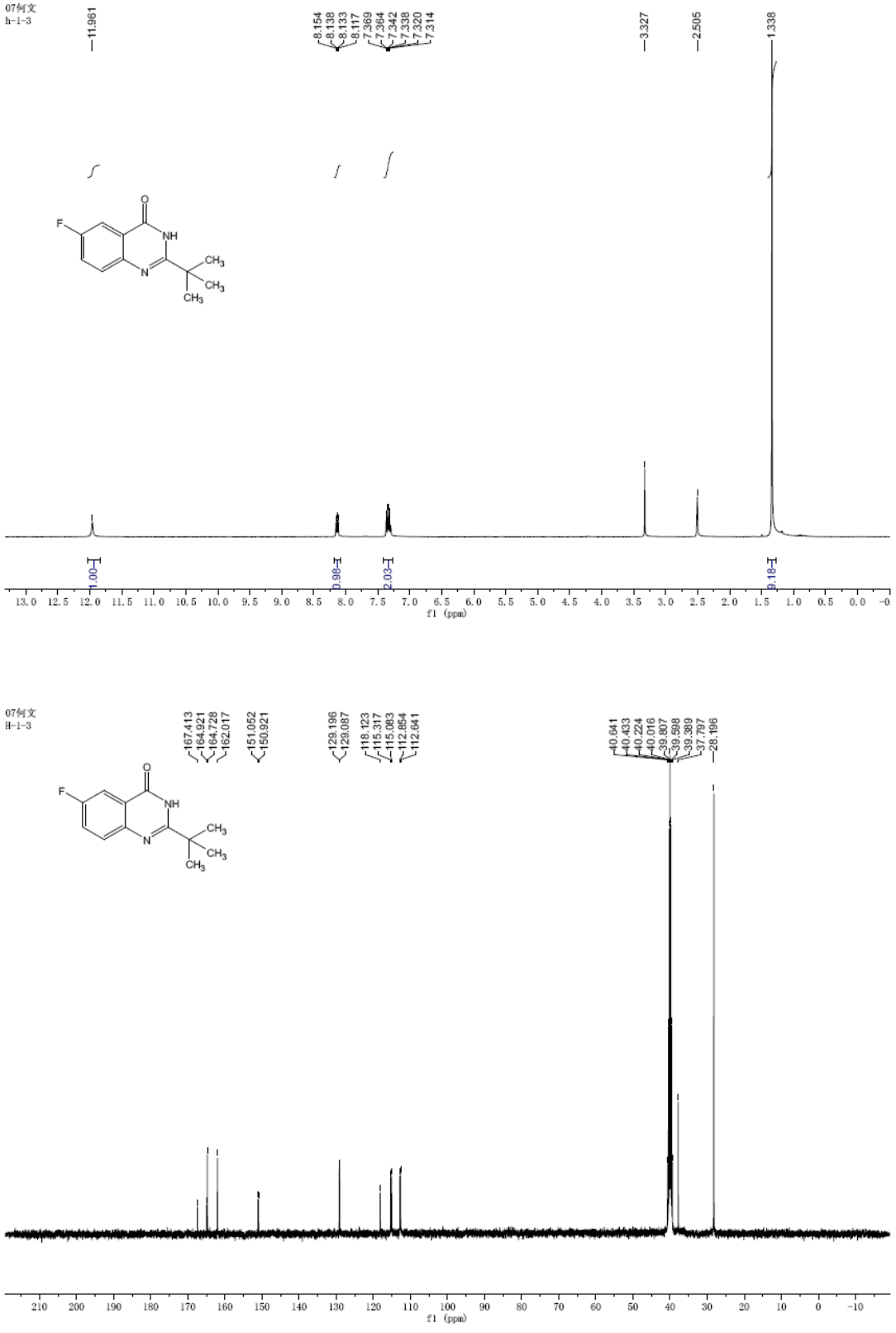
^{1}H NMR and ^{13}C NMR spectra of compound **3o**



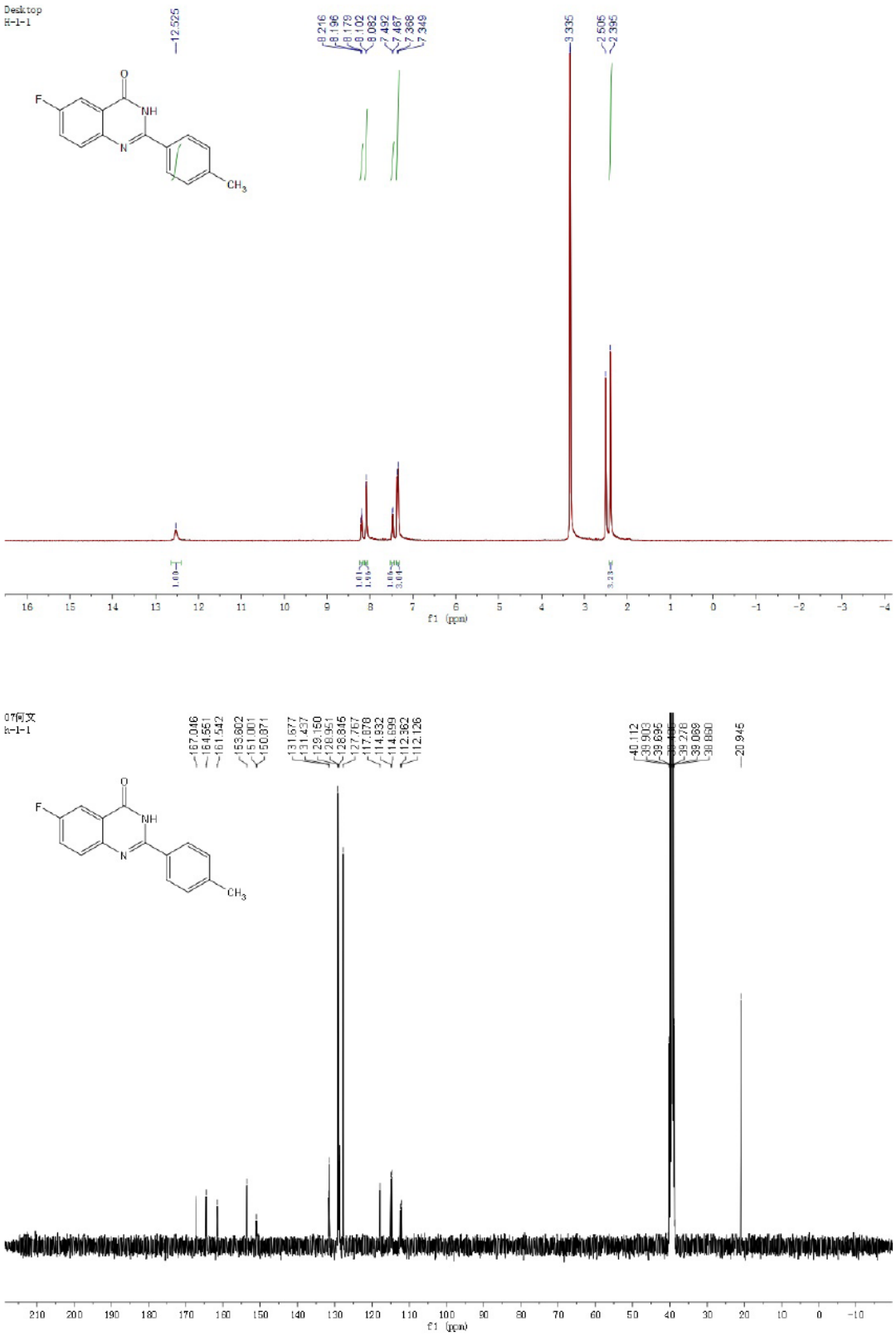
^{1}H NMR and ^{13}C NMR spectra of compound **3p**



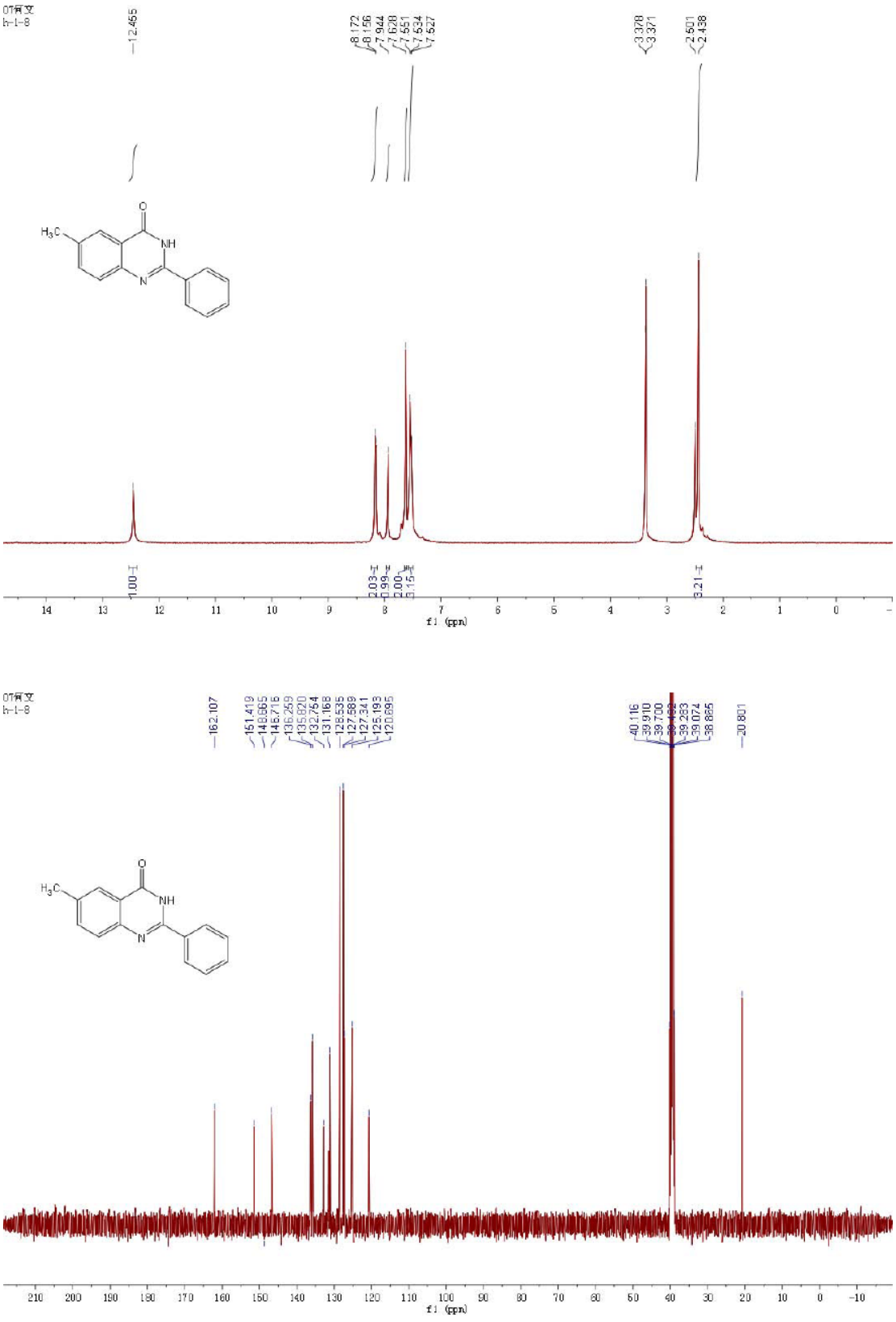
^{1}H NMR and ^{13}C NMR spectra of compound **3q**



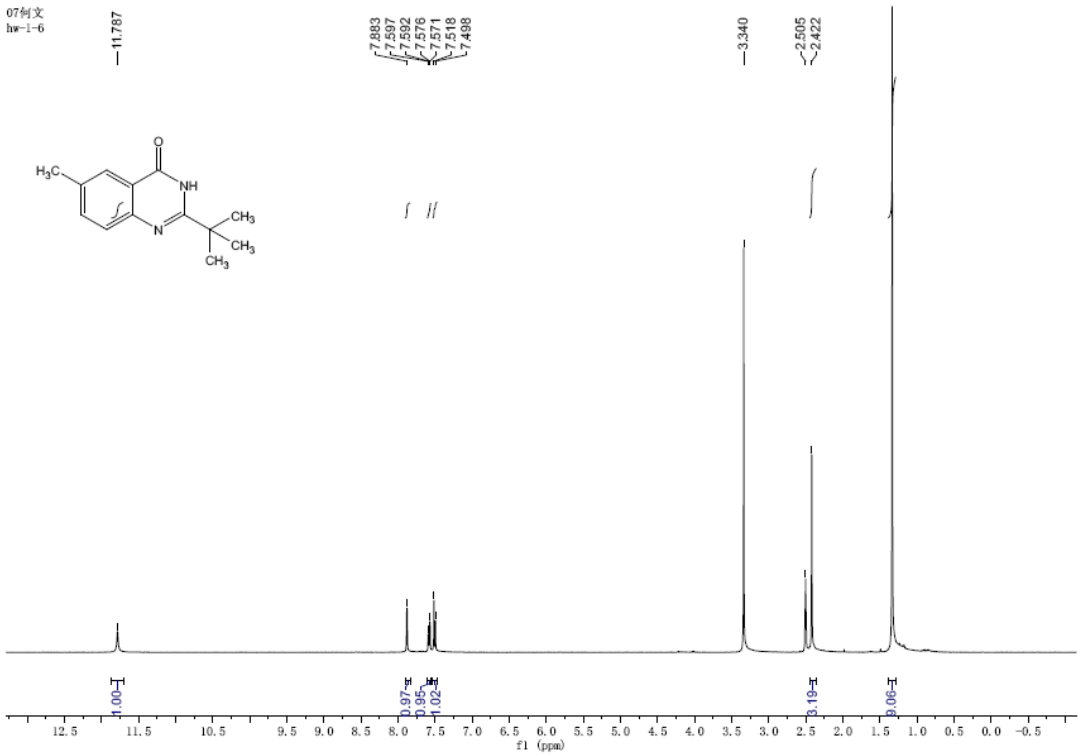
^{1}H NMR and ^{13}C NMR spectra of compound **3r**

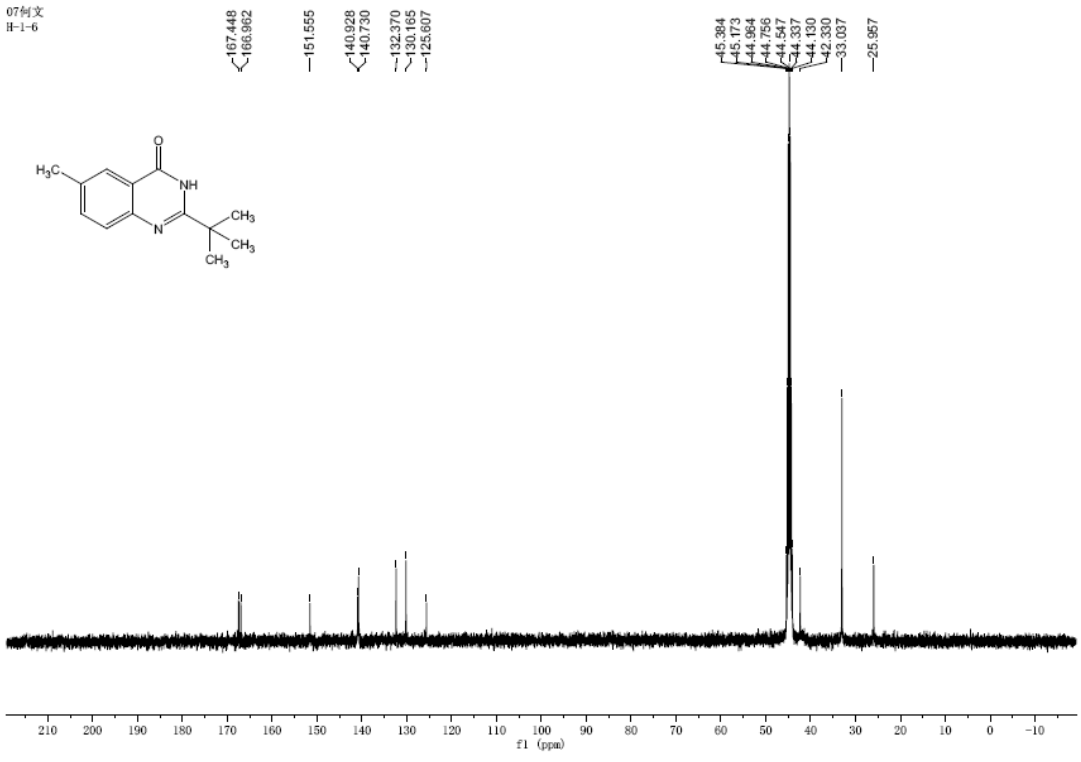


^{1}H NMR and ^{13}C NMR spectra of compound **3s**

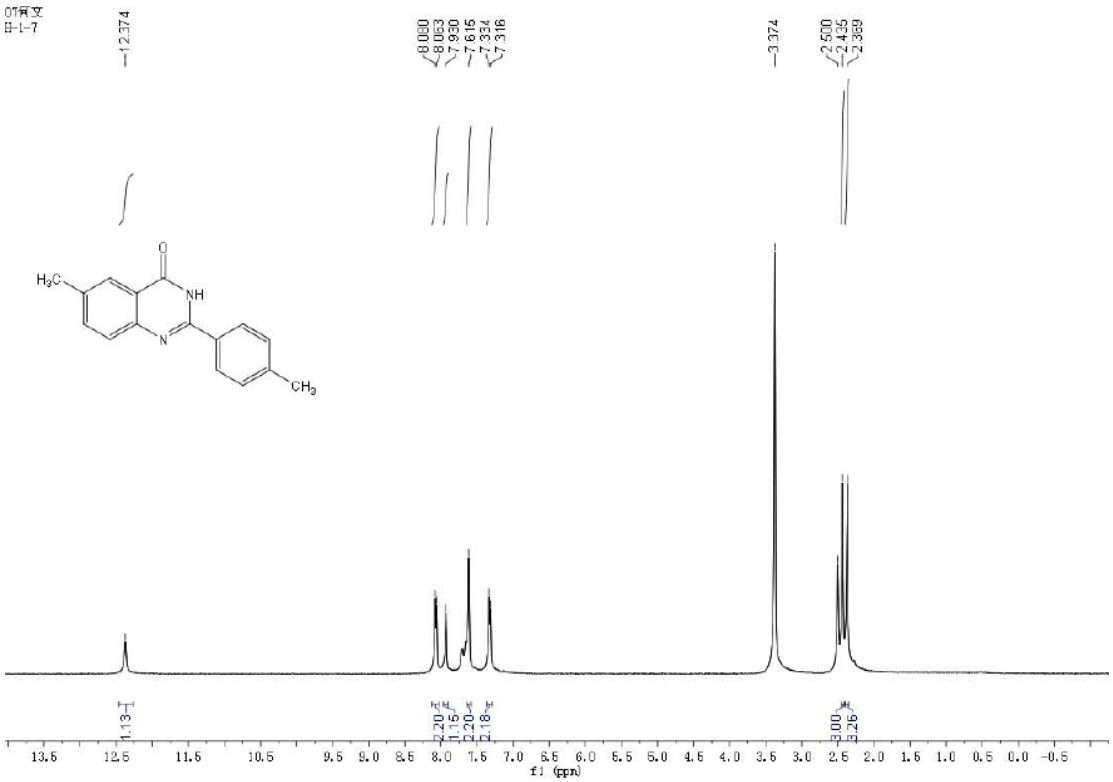


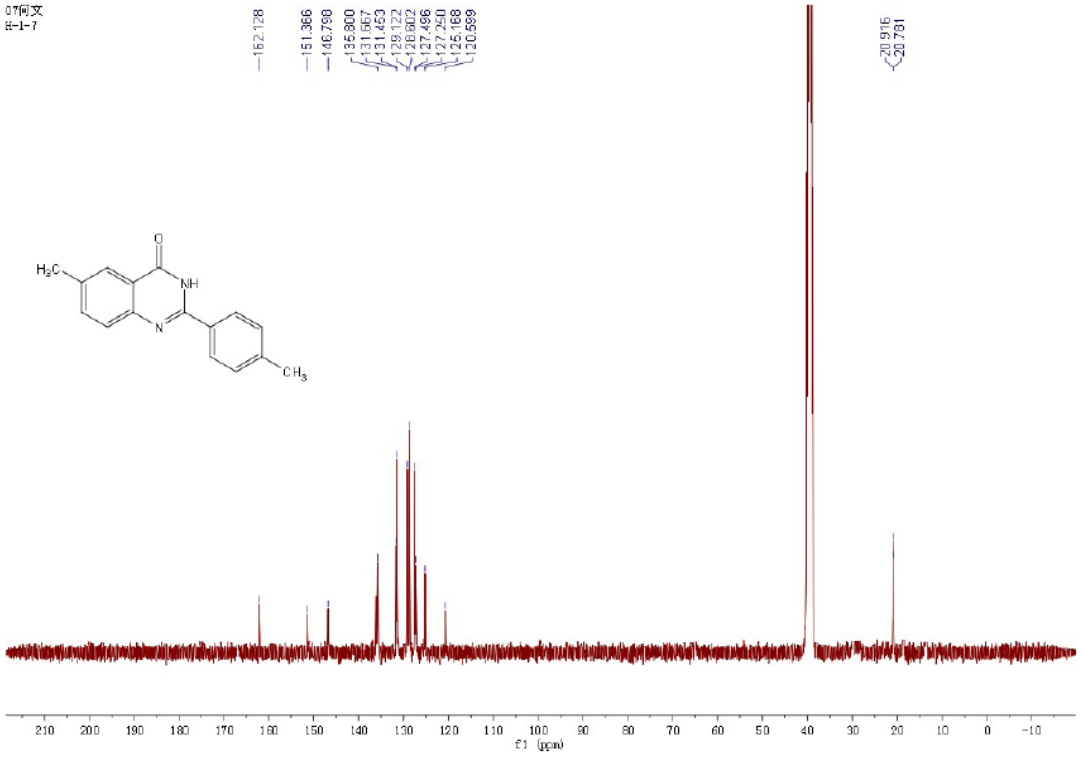
^{1}H NMR and ^{13}C NMR spectra of compound **3t**



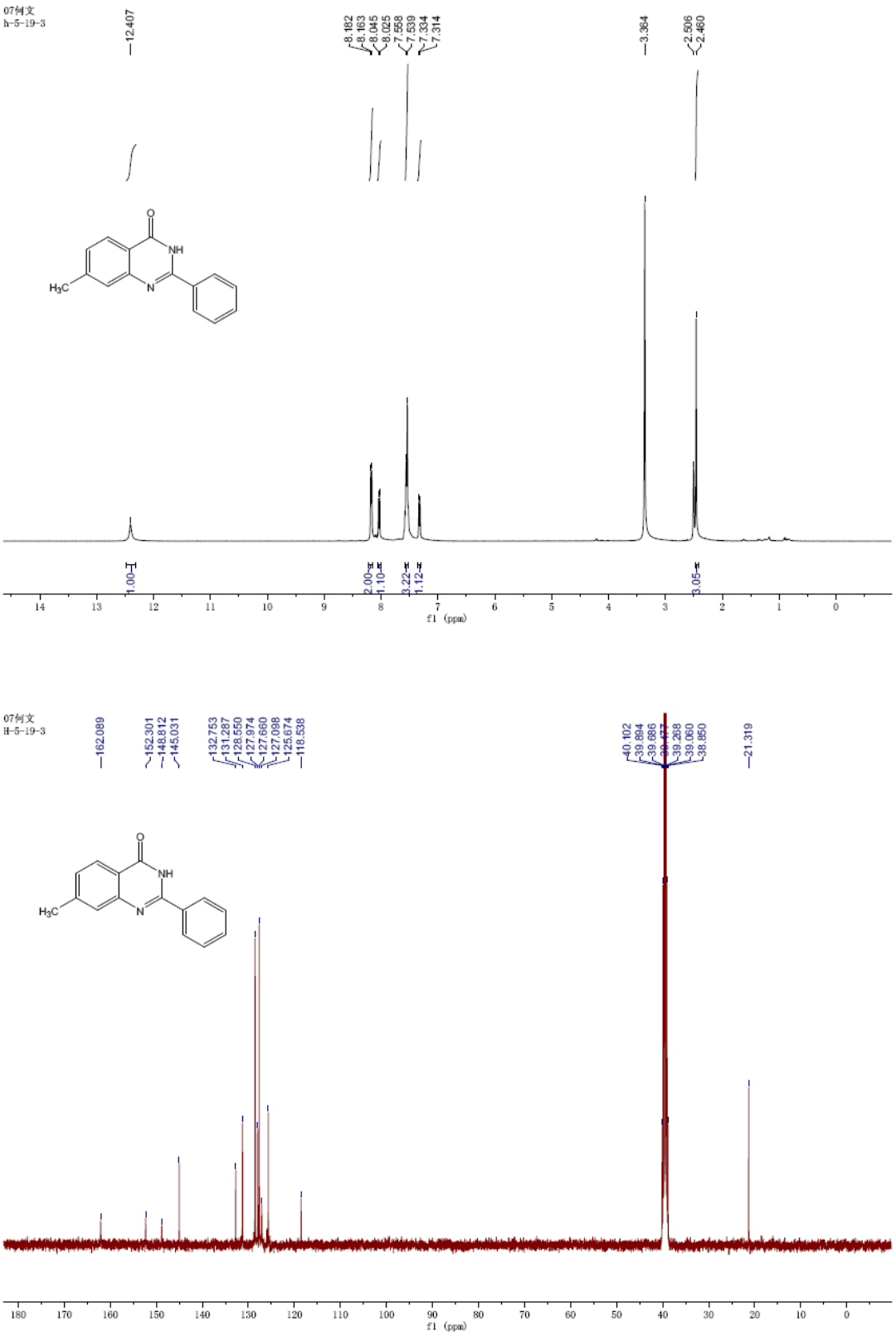


^{1}H NMR and ^{13}C NMR spectra of compound **3u**

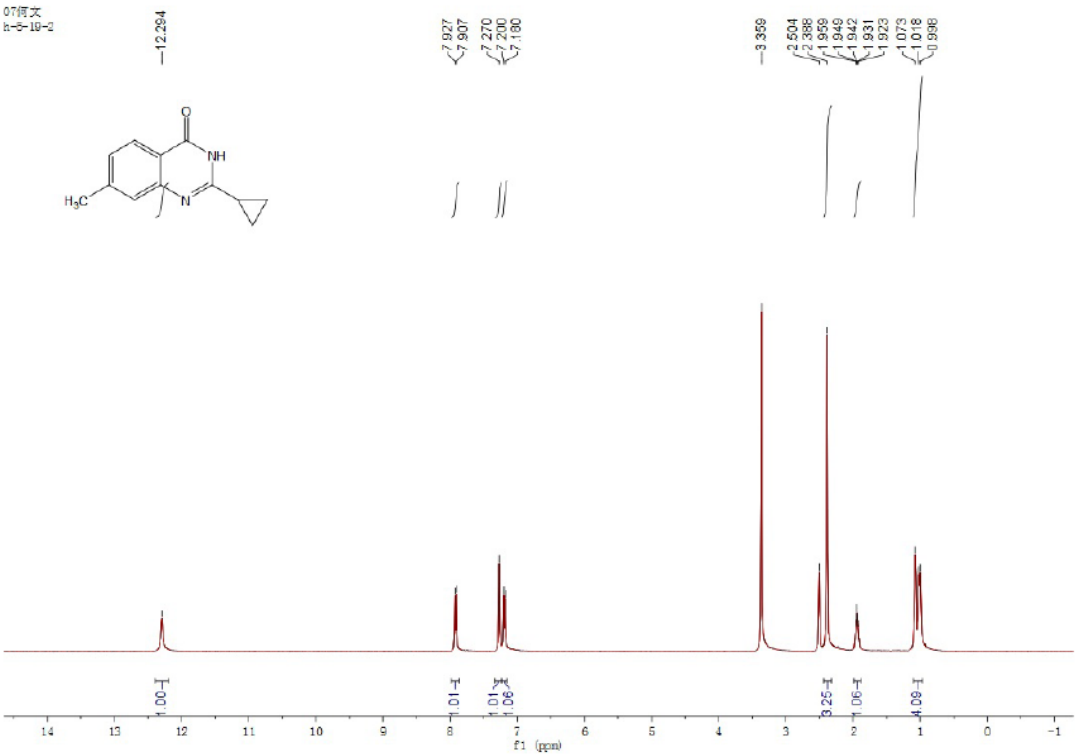


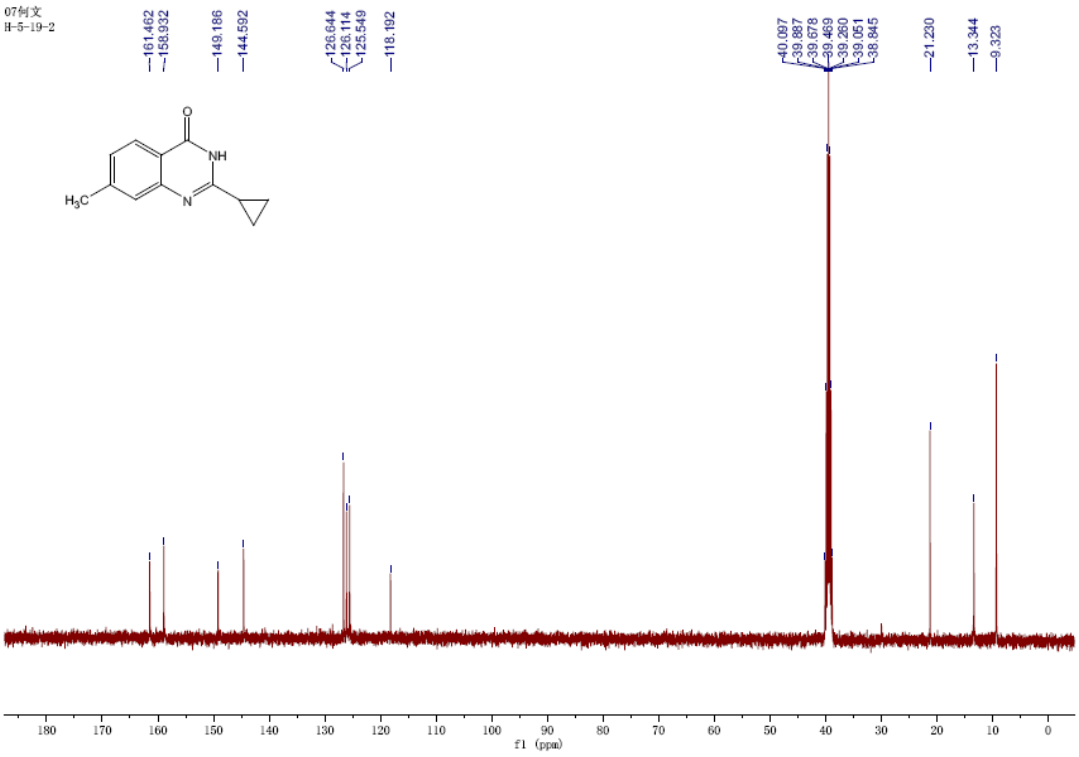


^{1}H NMR and ^{13}C NMR spectra of compound **3v**

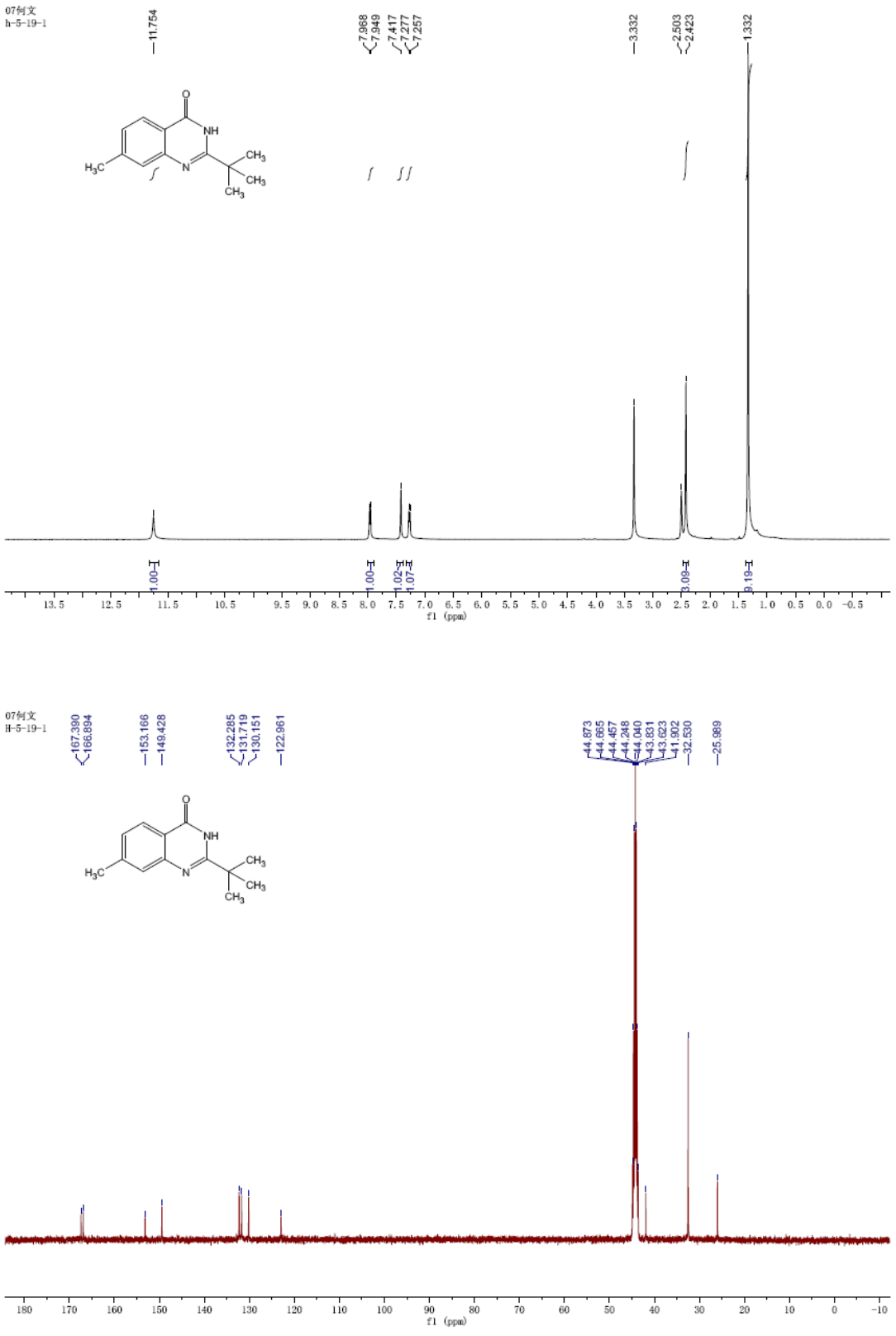


^{1}H NMR and ^{13}C NMR spectra of compound **3w**





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



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