Supporting Information

Auto-Redox Reaction: Tin(II)Chloride Mediated One-Step Reductive Cyclization Leading to the Synthesis of Novel Biheterocyclic 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-ones With Three Point Diversity

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General Considerations: All the solvents used for the reactions and purification were commercially available and used after distillation. All the products were characterized by ^{1}H, ^{13}C, DEPT90, DEPT135, Two-Dimensional Correlation Spectroscopy (COSY), Heteronuclear Single Quantum Coherence (HSQC), Heteronuclear Multiple Bond Correlation Spectroscopy (HMBC), FAB-MS and Elemental Analysis (C, H, N). Analytical TLC was performed using 2.5 x 5 cm plates coated with a 0.25 mm thickness of silica gel (60F-254), and visualization was accomplished with UV light and iodine. Column chromatography was performed using silica gel (100-200 and 230-400 mesh).

The NMR spectra were recorded at 298K using a 300 MHz FT-NMR spectrometer equipped with a 5 mm multinuclear inverse probehead with z-shielded gradient. Experiments were recorded in $CDCl_3$ or DMSO-d_6 at 25°C. Chemical shifts are given on the δ scale and are referenced to the TMS at 0.00 ppm for proton and 0.00 ppm for carbon. In the 1D measurement (^{1}H, ^{13}C and DEPT) 32K data points were used for the FID. The pulse programs of the following 2D experiments were taken from the Bruker software library and the parameters were as follows.

300 MHz ^{1}H-^{1}H gradient COSY spectra: relaxation delay d1 = 1s; 90° pulse, 6.85 μs for ^{1}H at –3 dB, number of scans 16; 1024 data points in t_2; spectral width 11.0 ppm in both dimensions; 256 experiments in t_1; linear prediction to 512; zero filling up to 1K.

300/75 MHz gradient HSQC spectra: relaxation delay d1 = 2s; evolution delay d2 = 3.44 ms; 90° pulse, 6.85 μs for ^{1}H, 10μs for ^{13}C hard pulses at –3.0 dB and 60μs for ^{13}C GARP decoupling with gradient ratio GPZ1:GPZ2:GPZ3 = 50:30:40.1; 1024 data points in t_2; spectral width 9.0 ppm in F2 and 160ppm in F1; number of scans 32; 256 experiments in t_1; linear prediction to 512; zero filling up to 1K and apodization with sine-bell in both dimensions prior to double Fourier transformation.

300/75 MHz gradient HMBC spectra: relaxation delay d1 = 2s; delay of the low-pass J-filter d2 = 3.44 ms; delay for evolution of long-range coupling d6 = 71 ms with gradient ratio same as HSQC; 2048 data points in t_2; spectral width 11.0 ppm in F2 and 240 ppm in F1; number of scans 52; 256 experiments in t_1; linear prediction to 512; zero filling up to 2K and apodization with 90° shifted square sine-bell in F1 dimension and sine-bell in F2 dimension prior to double Fourier transformation.

Experimental Section:

General Procedure for Synthesis of 2-Amino benzamide (2a): 2-Amino benzonitrile (1.04 g, 8.50 mmol) and potassium hydroxide (2.40 g, 42.5 mmol) were dissolved in hot ethanol (25 mL) and the reaction mixture was refluxed for two hours. The resulting yellowish brown solution was solution was allowed to cool to room temperature and the ethanol was removed in vacuo. The resulting brown solid was washed with water, saturated solution of $NaHCO_3$, and brine and extracted with ethyl acetate (50 mL x 3). The organic layer was evaporated in vacuo and the analytically pure product was obtained by recrystallization from ethanol as white solid.

2a: 0.96 g (80% yield); mp 112-114°C, ^{1}H NMR (DMSO-d_6, 300 MHz) δ = 6.45 (t, *J*= 7.8 Hz, 1H, ArH), 6.55 (brs, 2H, ArNH), 6.65 (d, *J*= 8.4 Hz, 1H, ArH), 7.05 (brs(o), 1H, -CONH), 7.11 (t(o), *J*= 8.1 Hz, 1H, ArH), 7.51 (d, *J*= 8.1 Hz, 1H, ArH): ^{13}C NMR (DMSO-d_6, 75 MHz) δ 113.7, 114.4, 116.4, 117.2, 128.8, 131.9, 150.2, 171.3; mass (FAB) *m/z* 137 (M^++1). Anal. Calcd for $C_7H_8N_2O$: C, 61.75; H, 5.92; N, 20.58. Found: C, 61.63; H, 5.94; N, 20.66.

***N*-[2-(aminocarbonyl)phenyl]-2-nitrobenzamide (3a)**: 2-Nitro benzoic acid (1.02 g, 6.1 mmol) and thionyl chloride (8 mL) were taken and the reaction mixture was refluxed for 2 hours at 80°C. The solution was allowed to cool at room temperature and thionyl chloride was evaporated in vacuo. The resulting wine red solution was taken and added dropwise to a solution of anthranilamide (**2a**) (1.04 g, 8.5 mmol) and triethylamine (3.1 mL, 17 mmol) in chloroform (25 mL) and stirred at room temperature for 2 hours. The precipitated solid was filtered, washed with ethanol to obtain compound **3a**. The analytically pure sample was obtained by recrystallization from methanol as a white colored solid.

3a: 2.2 g (86% yield); mp 195-197°C, ^{1}H NMR (DMSO-d_6, 300 MHz) δ = 7.22 (t, *J*= 8.4 Hz, 1H, ArH), 7.57 (t, *J*= 8.1 Hz, 1H, ArH), 7.75-7.90 (m, 5H, 4-ArH and 1-CONH), 8.10 (d, *J*= 8.7 Hz, 1H, ArH), 8.34 (brs, 1H, -CONH), 8.46 (brd, *J*= 8.4 Hz, 1H, ArH), 12.49 (s, 1H, ArNH): ^{13}C NMR (DMSO-d_6, 75 MHz) δ = 120.1, 120.4, 123.4, 124.6, 128.3, 128.7, 131.6, 132.0, 132.4, 134.0, 139.1, 147.1, 163.3, 171.2; mass (FAB) *m/z* 286

Anal. Calcd for $C_{14}H_{11}N_3O_4$: C, 58.95; H, 3.89; N, 14.73. Found: C, 59.12; H, 3.98; N, 14.58.

2-(2-Nitro-phenyl)-3H-quinazolin-4-one (4a): A mixture of benzamide (**3a**) (2.2 g, 7.0 mmol) in 10% aqueous KOH (50 mL) and EtOH (25 mL) was heated to reflux for 10 min. Ethanol was removed in vacuo, and the aqueous layer was extracted with ethyl acetate (50 mL x 3). The organic layer was washed with saturated solution of $NaHCO_3$, brine and dried over Na_2SO_4. Concentration of the organic layer in vacuo followed by silica gel column chromatographic purification of the residue using hexane: ethyl acetate as an elutant gave pure **4a** as light brown solid.

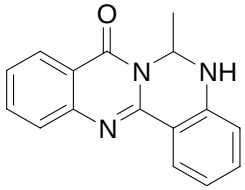
4a: 1.8 g, (95% yield); mp 210-212°C, ^{1}H NMR (DMSO-d_6, 300 MHz) δ = 7.57 (t, *J*= 7.5 Hz, 1H, ArH), 7.64 (d, *J*= 8.4, 1H, ArH), 7.80-7.94 (m, 4H, ArH), 8.19 (t, *J*= 8.1 Hz, 1H, ArH); ^{13}C NMR (DMSO-d_6, 75 MHz) δ = 121.2, 124.6, 125.9, 127.1, 127.3, 129.2, 131.5, 131.6, 134.0, 134.7, 147.5, 148.5, 151.7, 161.6; mass (FAB) *m/z* 268 (M^++1). Anal. Calcd for $C_{14}H_9N_3O_3$: C, 62.92; H, 3.39; N, 15.72. Found C, 62.78; H, 3.56, N, 15.97.

2-(3-Chloro-2-nitro-phenyl)-3H-quinazolin-4-one (4b): A mixture of benzamide (**3b**) (2.2 g, 6.9 mmol) in 10% aqueous KOH (50 mL) and EtOH (25 mL) was heated to reflux for 10 min. Ethanol was removed in vacuo, and the aqueous layer was extracted with ethyl acetate (50 mL x 3). The organic layer was washed with saturated solution of $NaHCO_3$, brine and dried over Na_2SO_4. Concentration of the organic layer in vacuo followed by silica gel column chromatographic purification of the residue using hexane: ethyl acetate as an elutant gave pure **4b** as dark brown solid.

4b: 1.8 g, (90% yield); mp 172-175°C, ^{1}H NMR ($CDCl_3$+DMSO-d_6, 300 MHz) δ = 7.53 (t, *J*= 7.5 Hz, 1H, ArH), 7.60 (d, *J*= 8.1 Hz, 1H, ArH), 7.63-7.72 (m, 1H, ArH), 7.76-7.81 (m, 2H, ArH), 7.9 (brd, *J*= 7.5 Hz, 1H, ArH), 8.22 (d, *J*= 8.1 Hz, 1H, ArH); mass (FAB) *m/z* 302 (M^++1). Anal. Calcd for $C_{14}H_8ClN_3O_3$: C, 55.74; H, 2.67; N, 13.93. Found C, 55.83; H, 2.72; N, 14.04.

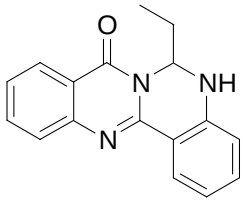
6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in ethanol and refluxed at 80°C for 2 hours. The reaction mixture was concentrated in vacuo and diluted in ethyl acetate (10 mL). The organic layer was washed with saturated $NaHCO_3$, brine

solution and dried over Na_2SO_4. The organic layer was evaporated under reduced pressure and the residue was purified by column chromatography (5% EtOAc in Hexane) to provide **5a** as a light yellow solid.



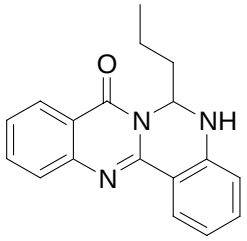
5a: 170 mg (85% yield), mp 132-133°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 1.49 (d, *J*= 5.4 Hz, 3H, -CH_3), 4.61 (brs, 1H, -NH), 6.42 (m, 1H, -CH), 6.77 (d, *J*= 7.8 Hz, 1H, ArH), 6.98 (t, *J*= 7.2 Hz, 1H, ArH), 7.32-7.44 (m(o), 2H, ArH), 7.73 (brs, 2H, ArH), 8.28 (d(o), *J*= 7.5 Hz, 1H, ArH), 8.36 (d(o), *J*= 7.5 Hz, 1H, ArH): ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 20.0, 59.5, 116.4, 116.8, 120.2, 120.6, 125.9, 126.7, 127.4, 127.6, 133.3, 134.3, 143.7, 146.7, 148.2, 160.1; mass (FAB) *m/z* 264 (M^++1). Anal. Calcd for $C_{16}H_{13}N_3O$: C, 72.99; H, 4.98; N, 15.96. Found C, 73.16; H, 5.14; N, 16.09.

6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5b): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in *n*-propanol and heated at 80°C for 2 hours. The remaining workup was similar as described for **5a**, which provided light fluorescent yellow colored solid.



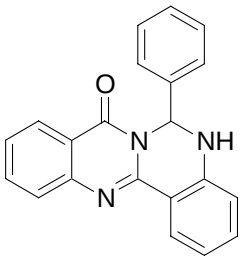
5b: 149 mg (72% yield), mp 138-140°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 0.96 (t, *J*= 7.2 Hz, 3H, -CH_3), 1.69-1.80 (m, 1H, 1-CH_2), 1.88-1.98 (m, 1H, 1-CH_2), 4.70 (brs, 1H, NH), 6.15 (brm, 1H, -CH), 6.78 (d, *J*= 7.8Hz, 1H, ArH), 6.98 (t, *J*= 7.5 Hz, 1H, ArH), 7.33-7.45 (m(o), 2H, ArH), 7.74 (brm, 2H, ArH), 8.28 (d(o), *J*= 7.8 Hz, 1H, ArH), 8.34 (d(o), *J*= 8.1 Hz, 1H, ArH): ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 9.8, 25.8, 64.0, 116.3, 117.6, 120.4, 120.6, 126.0, 126.9, 127.4, 127.7, 133.3, 134.4, 143.4, 146.9, 148.2, 160.3; mass (FAB) *m/z* 278 (M^++1). Anal. Calcd for $C_{17}H_{15}N_3O$: C, 73.63; H, 5.45; N, 15.15. Found C, 73.60; H, 5.58; N, 15.09.

6-Propyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5c): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in *n*-butanol and heated at 80°C for 2 hours. The remaining workup was similar as described for **5a**, which provided light fluorescent yellow colored solid.



5c: 140 mg (64% yield), mp 147-148°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 0.90 (t, *J*= 7.2 Hz, 3H, -CH_3), 1.40-1.52 (m, 2H, -CH_2), 1.60-1.71 (m, 1H, 1-CH_2), 1.88-1.98 (m, 1H, 1-CH_2), 4.68 (brs, 1H,-NH), 6.25 (m, 1H, -CH), 6.78 (d, *J*= 7.8 Hz, 1H, ArH), 6.99 (t, *J*= 7.5Hz, 1H, ArH), 7.33-7.45 (m(o), 2H, ArH), 7.74 (brm, 2H, ArH), 8.28 (d(o), *J*= 8.1 Hz, 1H, ArH), 8.35 (d(o), *J*= 8.1 Hz, 1H, ArH): ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 13.4, 18.6, 34.8, 62.7, 116.3, 117.6, 120.4, 120.7, 126.0, 126.8, 127.4, 127.7, 133.3, 134.3, 143.4, 146.9, 148.2, 160.2; mass (FAB) *m/z* 292 (M^++1). Anal. Calcd for $C_{18}H_{17}N_3O$: C, 74.20; H, 5.88; N, 14.42. Found C, 74.08; H, 5.98; N, 14.51.

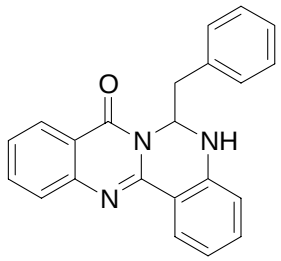
6-Phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5d): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in benzyl alcohol and heated at 90°C for 1 hour. The reaction mixture was concentrated in vacuo and diluted in ethyl acetate (10 mL). The organic layer was washed with saturated $NaHCO_3$, brine solution and the resulting benzyl alcohol was evaporated through vacuum distillation. The solid was dried in vacuo and the residue was purified by column chromatography (5% EtOAc in Hexane) to provide **5d** as a reddish brown solid.



5d: 224 mg, (92% yield), mp 188-189°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 5.14 (brs, 1H, -NH), 6.77 (d, *J*= 9 Hz, 1H, ArH), 6.94 (t, *J*= 7.8 Hz, 1H, ArH), 7.18-7.44 (m, 8H, ArH),

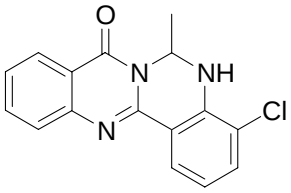
7.75 (brs, 2H, ArH), 8.31 (t, *J*= 8.4 Hz, 2H, ArH); ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 63.6, 116.3, 117.7, 120.5, 126.0, 126.1, 127.1, 127.5, 127.6, 128.5, 128.6, 133.4, 134.5, 139.2, 143.6, 147.1, 148.2, 160.7: mass (FAB) *m/z* 326 (M^++1). Anal. Calcd for $C_{21}H_{15}N_3O$: C, 74.20; H, 5.88; N, 14.42. Found C, 74.41; H, 5.90; N, 14.31.

6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in phenethyl alcohol and heated at 90°C for 1 hour. The reaction mixture was concentrated in vacuo and diluted in ethyl acetate (10 mL). The organic layer was washed with saturated $NaHCO_3$, brine solution and the resulting phenethyl alcohol was evaporated through vacuum distillation. The solid was dried in vacuo and the residue was purified by column chromatography (5% EtOAc in Hexane) to provide **5e** as a yellowish brown solid.



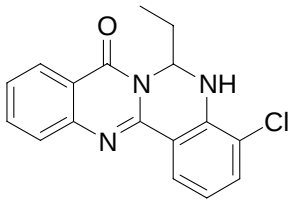
6-**Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e)**: Yellowish brown solid, 205 mg (80% yield), mp 155-156°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 2.88 (dd, *J*= 12.9 and 2.7 Hz, 1H, 1-CH_2), 3.06 (t, *J*= 12.9 Hz, 1H, 1-CH_2), 4.47 (brs, 1H, -NH), 6.19 (brd, *J*= 10.5 Hz, 1H, -CH), 6.60 (d, *J*= 8.1 Hz, 1H, ArH), 6.93 (t, *J*= 7.5 Hz, 1H, ArH), 7.12 (d(o), *J*= 7.2 Hz, 2H, ArH), 7.17-7.38 (m(o), 5H, ArH), 7.67 (brm, 2H, ArH), 8.21 (d, *J*= 8.1 Hz, 1H, ArH), 8.30 (d, *J*= 8.1 Hz, 1H, ArH); ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 38.4, 64.2, 116.1, 117.2, 120.3, 120.6, 126.1, 126.8, 127.2, 127.5, 127.8, 128.9, 129.5, 133.5, 134.4, 136.1, 143.6, 146.7, 148.2, 160.2: mass (FAB) *m/z* 340 (M^++1). Anal. Calcd for $C_{22}H_{17}N_3O$: C, 77.86; H, 5.05; N, 12.38. Found C, 77.90; H, 4.96; N, 12.57.

4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5f):



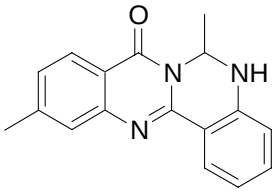
5f: 175 mg (78% yield), mp 116-118°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 1.51 (d, *J*= 6.3 Hz, 3H, -CH3), 5.10 (brs, 1H, -NH), 6.54 (m, 1H, -CH), 6.90 (t, *J*= 8.1 Hz, 1H, ArH), 7.41-7.48 (m(o), 2H, ArH), 7.73-7.74 (brm, 2H, ArH), 8.29 (d, *J*= 7.8 Hz, 2H, ArH); ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 20.5, 59.5, 117.6, 119.7, 120.2, 120.6, 126.2, 126.3, 126.8, 127.5, 132.7, 134.5, 140.3, 145.8, 147.9, 159.9: mass (FAB) *m/z* 298 (M^++1). Anal. Calcd for $C_{16}H_{12}ClN_3O$: C, 64.54; H, 4.06; N, 14.11. Found C, 64.68; H, 4.09; N, 14.20.

4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5g):



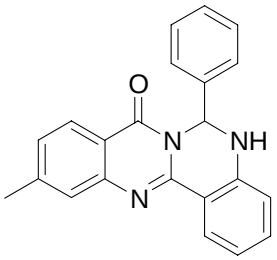
5g: 160 mg (68% yield), mp 133-134°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 0.98 (t, *J*= 7.5 Hz, 3H, -CH_3), 1.67-1.92 (m, 2H, -CH_2), 5.24 (brs, 1H, -NH), 6.23 (m, 1H, -CH), 6.91 (t, *J*= 7.8 Hz, 1H, ArH), 7.41-7.47 (m, 2H, ArH), 7.73-7.78 (brm, 2H, ArH), 8.27-8.31 (m, 2H, ArH): ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 9.7, 26.0, 64.1, 118.5, 119.9, 120.1, 120.7, 126.3, 126.4, 126.9, 127.5, 132.6, 134.5, 140.2, 146.1, 148.0, 160.1: mass (FAB) *m/z* 312 (M^++1). Anal. Calcd for $C_{17}H_{14}ClN_3O$: C, 65.49; H, 4.53; N, 13.48. Found C, 65.38; H, 4.62; N, 13.32.

6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5h):



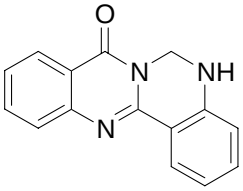
5h: 170 (82% yield), mp 128-130°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 1.49 (d, *J*= 6Hz, 3H, -CH_3), 2.50 (s, 3H, Ar-CH_3), 4.51 (brs, 1H, -NH), 6.42 (brm, 1H, -CH), 6.76 (d, *J*= 8.1 Hz, 1H, ArH), 6.98 (t, *J*= 7.5 Hz, 1H, ArH), 7.25 (d, *J*= 8.7 Hz, 1H, ArH), 7.35 (t, *J*= 7.5 Hz, 1H, ArH), 7.54 (brs, 1H, ArH), 8.16 (d, 8.1 Hz, 1H, ArH), 8.34 (d, *J*= 7.8 Hz, 1H, ArH): ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 20.1, 21.9, 59.4, 116.4, 117.1, 118.2, 120.3, 126.6, 127.1, 127.6, 133.2, 143.5, 145.3, 146.8, 148.3, 160.0: mass (FAB) *m/z* 278 (M^++1). Anal. Calcd for $C_{17}H_{15}N_3O$: C, 73.63; H, 5.45; N, 15.15. Found C, 73.55; H, 5.53, N, 15.18.

11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5i):



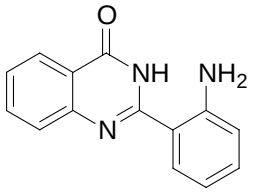
5i: 186 mg (73% yield), mp 211-212°C, ^{1}H NMR (DMSO-d_6, 300 MHz) δ = 2.47 (s, 3H, -CH_3), 6.80 (t, *J*= 9 Hz, 1H, ArH), 6.90 (d, *J*= 8.1 Hz, 1H, ArH), 7.15 (d(o), *J*= 7.2 Hz, 2H, ArH), 7.22-7.34 (m(o), 5H, ArH), 7.54 (s, 1H, ArH), 7.96 (d, *J*= 2.7 Hz, 1H, ArH), 8.08 (t, *J*= 7.5 Hz, 1H, ArH): ^{13}C NMR (DMSO-d_6, 75 MHz) δ = 21.4, 62.3, 115.7, 115.9, 117.5, 118.8, 125.7, 126.5, 126.8, 127.8, 128.1, 128.6, 133.6, 139.5, 145.1, 145.6, 147.3, 147.8, 159.6: mass (FAB) *m/z* 340 (M^++1). Anal. Calcd for $C_{22}H_{17}N_3O$: C, 77.86; H, 5.05; N, 12.38. Found C, 77.75; H, 4.98; N, 12.43.

5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5j): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in propargyl alcohol and heated at 80°C for 2 hours. The reaction mixture was concentrated in vacuo and diluted in ethyl acetate (10 mL). The organic layer was washed with saturated $NaHCO_3$, brine solution and dried over Na_2SO_4. The solid was dried in vacuo and the residue was purified by column chromatography (5% EtOAc in Hexane) to provide **5j** as a dark yellow colored solid.



5j: 142 mg (76% yield), mp 178-180°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 4.57 (brs, 1H, -NH), 5.40 (s, 2H, -CH_2), 6.82 (d, *J*= 7.8 Hz, 1H, ArH), 7.02 (t, *J*= 7.5 Hz, 1H, ArH), 7.33-7.44 (m, 2H, ArH), 7.72-7.74 (brm, 2H, ArH), 8.28 (d, *J*= 8.1 Hz, 1H, ArH), 8.35 (d, *J*= 7.8 Hz, 1H, ArH); ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 53.1, 116.1, 118.2, 120.7, 121.1, 126.1, 126.8, 127.5, 127.9, 133.1, 134.4, 146.2, 147.8, 148.2, 160.3: mass (FAB) *m/z* 250 (M^++1). Anal. Calcd for $C_{15}H_{11}N_3O$: C, 72.28; H, 4.45; N, 16.86. Found C, 72.13; H, 4.63; N, 16.61.

2-(2-Amino-phenyl)-3*H*-quinazolin-4-one (6): A solution of **4a** (200 mg, 0.75 mmol) and $SnCl_2.2H_2O$ (850 mg, 3.75 mmol), were dissolved in methanol and refluxed at 80°C for 2 hours. The reaction mixture was concentrated in vacuo and diluted in ethyl acetate (10 mL). The organic layer was washed with saturated $NaHCO_3$, brine solution and dried over Na_2SO_4. The organic layer was evaporated under reduced pressure and the residue was purified by column chromatography (15% EtOAc in Hexane) to provide **6** as a dark yellow solid.



6: 151 mg (85%yield), mp 155-156°C, ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 6.25 (brs, 2H, -NH_2), 6.81 (m, 2H, ArH), 7.27 (m, 1H, ArH), 7.46 (t, J= 7.2 Hz, 1H, ArH), 7.63-7.78 (m(o), 3H, ArH), 8.29 (d, J= 7.8 Hz, 1H, ArH), 10.25 (brs, 1H, -CONH): ^{13}C NMR ($CDCl_3$, 75 MHz) δ = 113.3, 117.3, 117.7, 120.6, 126.5, 126.6, 127.1, 132.5, 134.8, 148.6, 148.7, 152.3, 162.7: mass (FAB) *m/z* 238 (M^++1). Anal. Calcd for $C_{14}H_{11}N_3O$: C, 70.87; H, 4.67; N, 17.71. Found: C, 70.96; H, 4.63; N, 17.77.

Figure Legends:

Fig S-1: ^{1}H NMR Spectrum of 2-Amino benzamide (**2a**).

Fig S-2: ^{13}C NMR Spectrum of 2-Amino benzamide (**2a**)

Fig S-3: ^{1}H NMR Spectrum of *N*-[2-(aminocarbonyl) phenyl]-2-nitro benzamide (**3a**).

Fig S-4: ^{13}C NMR Spectrum of *N*-[2-(aminocarbonyl) phenyl]-2-nitro benzamide (**3a**).

Fig S-5: ^{1}H NMR Spectrum of 2-(2-Nitrophenyl)-3*H*-quinazolin-4-one (**4a**).

Fig S-6: ^{1}H NMR Spectrum of 2-(3-Chloro-2-nitrophenyl) quinazoline-4(3*H*)-one (**4b**).

Fig S-7: ^{1}H NMR Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-8: ^{13}C NMR Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-9: DEPT-90 NMR Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-10: COSY Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-11: HSQC Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-12: HMBC Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-13: Expanded Region of the HMBC Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).

Fig S-14: ^{1}H NMR Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5a**).

Fig S-15: ^{13}C NMR Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5a**).

Fig S-16: DEPT-135 NMR Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5a**).

Fig S-17: COSY Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5a**).

Fig S-18: HSQC Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5a**).

Fig S-19: HMBC Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5a**).

Fig S-20: ^{1}H NMR Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5b**).

Fig S-21: ^{13}C NMR Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5b**).

Fig S-22: (a). DEPT-90 and (b). DEPT-90 NMR Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5b**).

Fig S-23: COSY Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5b**).

Fig S-39: HSQC Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5e**).

Fig S-40: HMBC Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5e**).

Fig S-41: Expanded Region of the HMBC Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5e**).

Fig S-42: ^{1}H NMR Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5g**).

Fig S-43: ^{13}C NMR Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5g**).

Fig S-44: DEPT-135 NMR Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5g**).

Fig S-45: COSY Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5g**).

Fig S-46: HSQC Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5g**).

Fig S-47: ^{1}H NMR Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5h**).

Fig S-48: ^{13}C NMR Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5h**).

Fig S-49: COSY Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5h**).

Fig S-50: HSQC Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5h**).

Fig S-51: HMBC Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5h**).

Fig S-52: Extended Region of the HMBC Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5h**).

Fig S-53: ^{1}H NMR Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5i**).

Fig S-54: ^{13}C NMR Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5i**).

Fig S-55: COSY Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b]*quinazolin-8-one (**5i**).

Fig S-56: HSQC Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5i**).

Fig S-57: HMBC Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5i**).

Fig S-58: Expanded Region of the HMBC Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5i**).

Fig S-59: ^{1}H NMR Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

Fig S-60: ^{13}C NMR Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

Fig S-61: (a). DEPT-90 and (b). DEPT-135 NMR Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

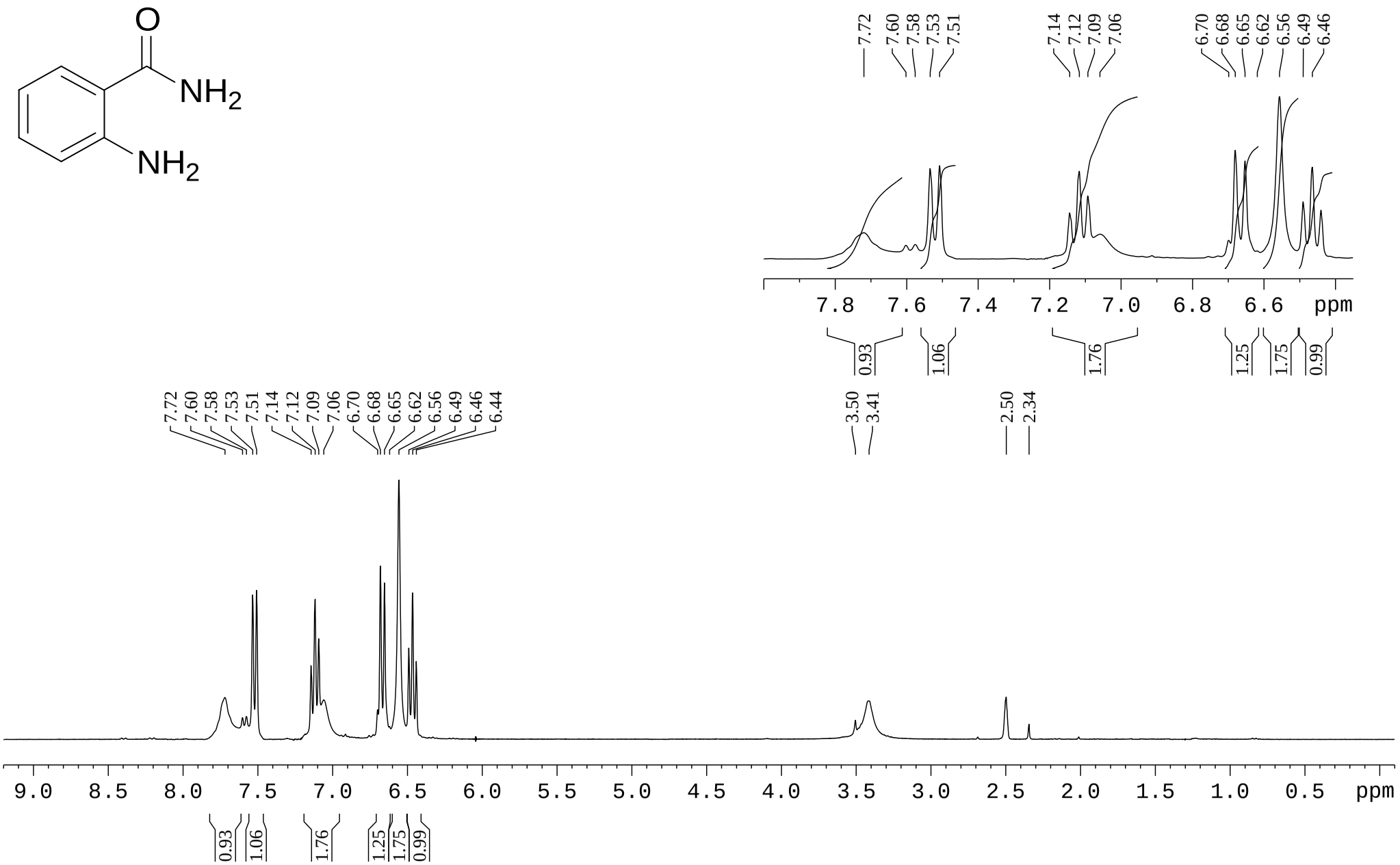
Fig S-62: COSY Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

Fig S-63: HSQC Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

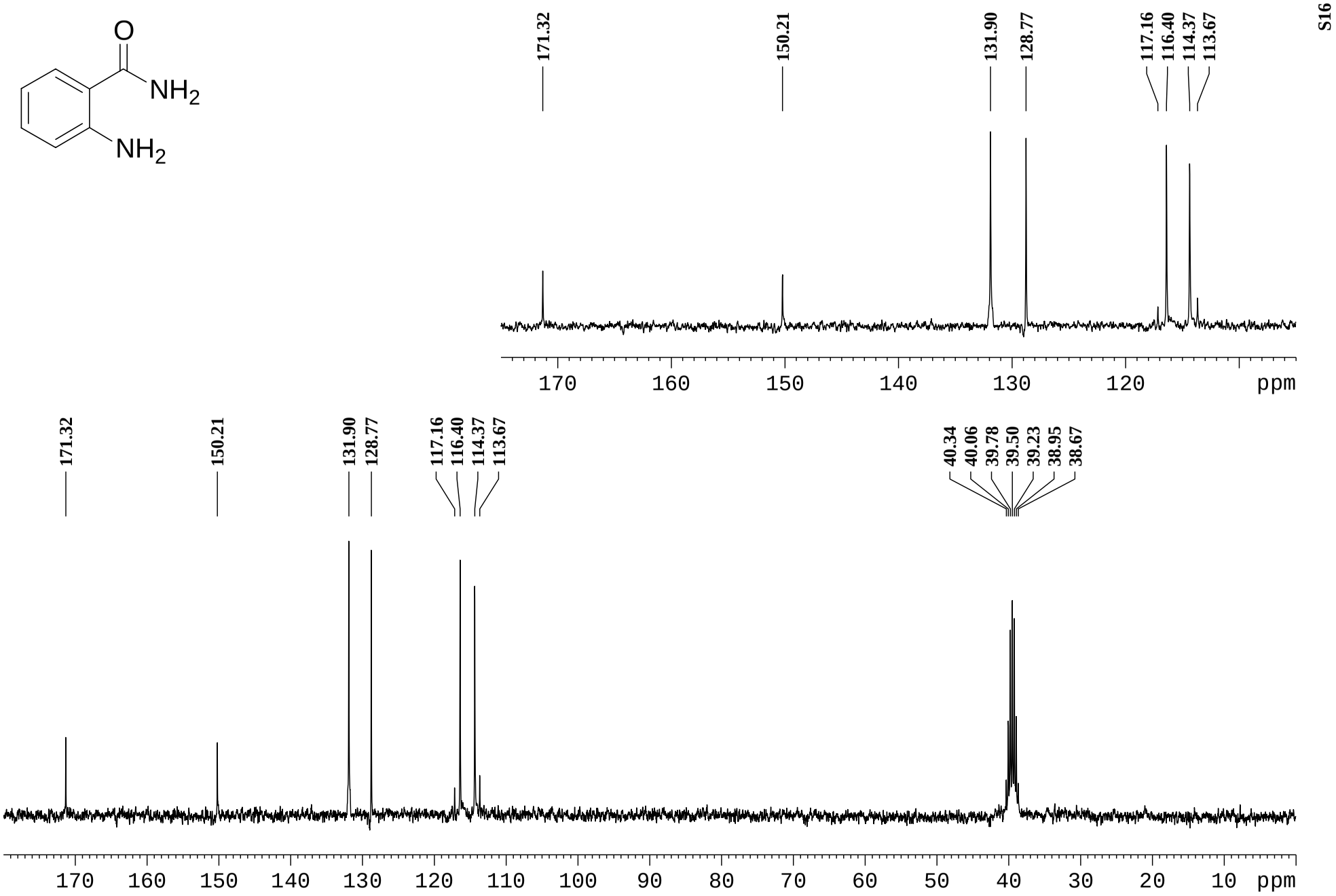
Fig S-64: HMBC Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

Fig S-65: Expanded Region of the HMBC Spectrum of 5,6-Dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5j**).

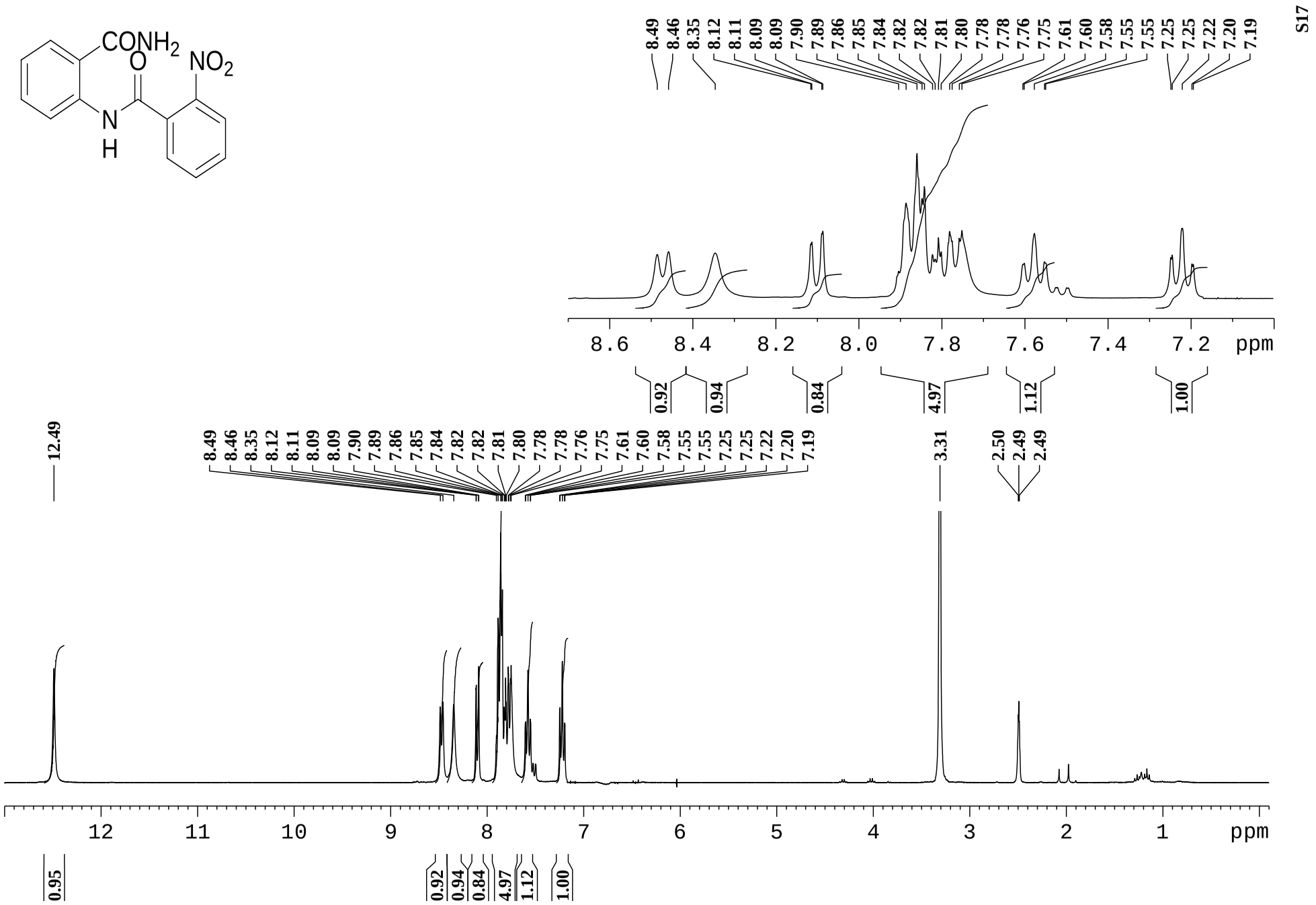
Fig S-66: 1D-NOE spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).



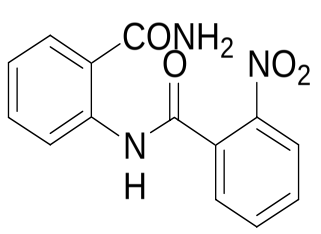
S-1: 1H NMR Spectrum of 2-Amino benzamide (2a)

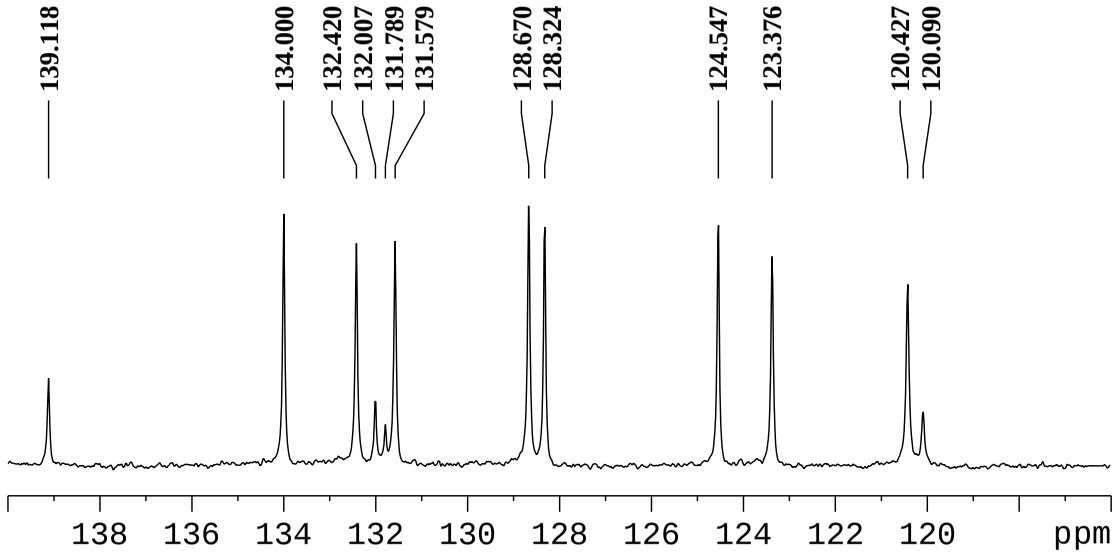


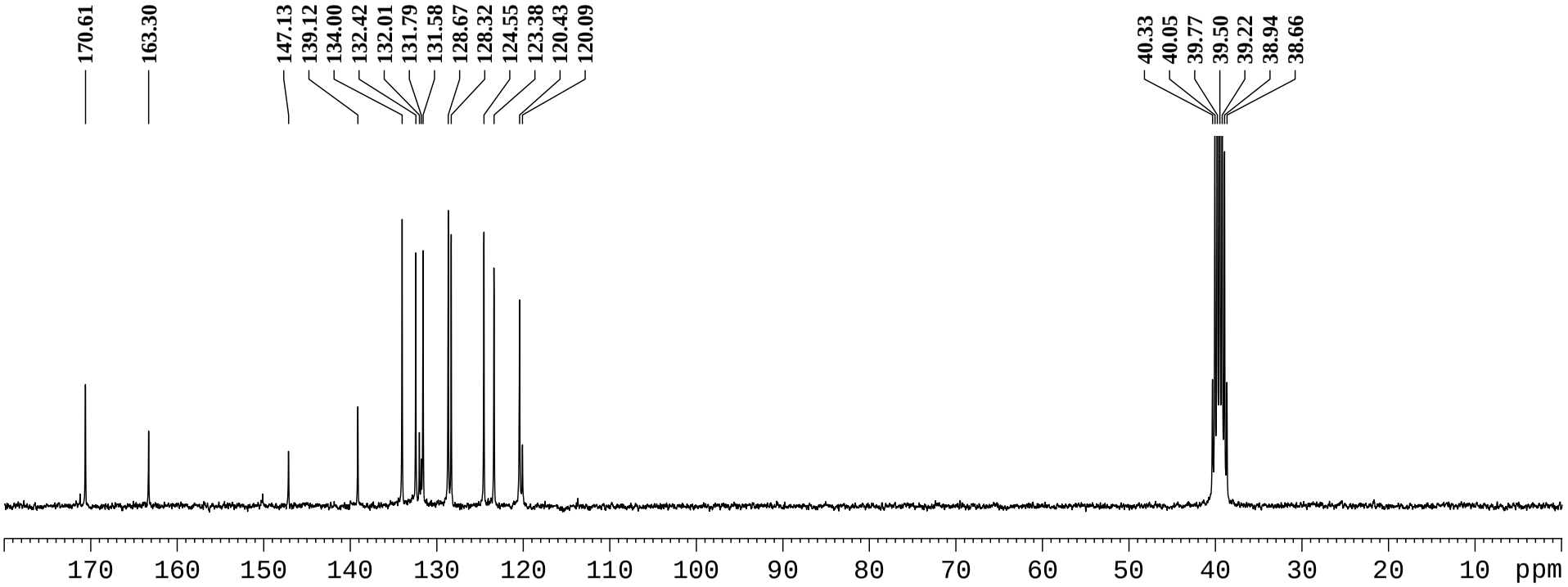
S-2: 13C NMR Spectrum of 2-Amino benzamide (2a).



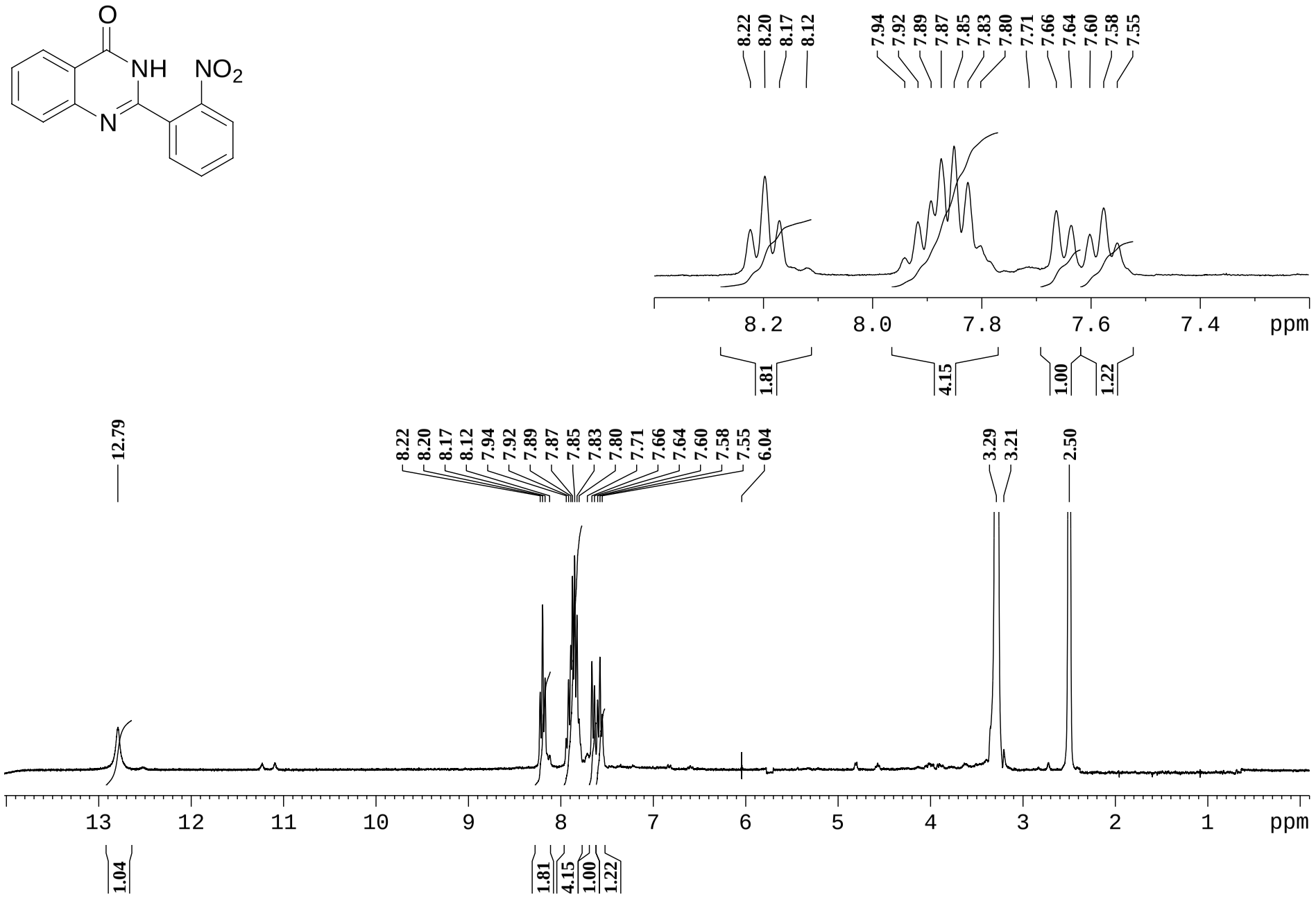
S-3: 1H NMR Spectrum of N-[2-(aminocarbonyl) phenyl]-2-nitro benzamide (3a).



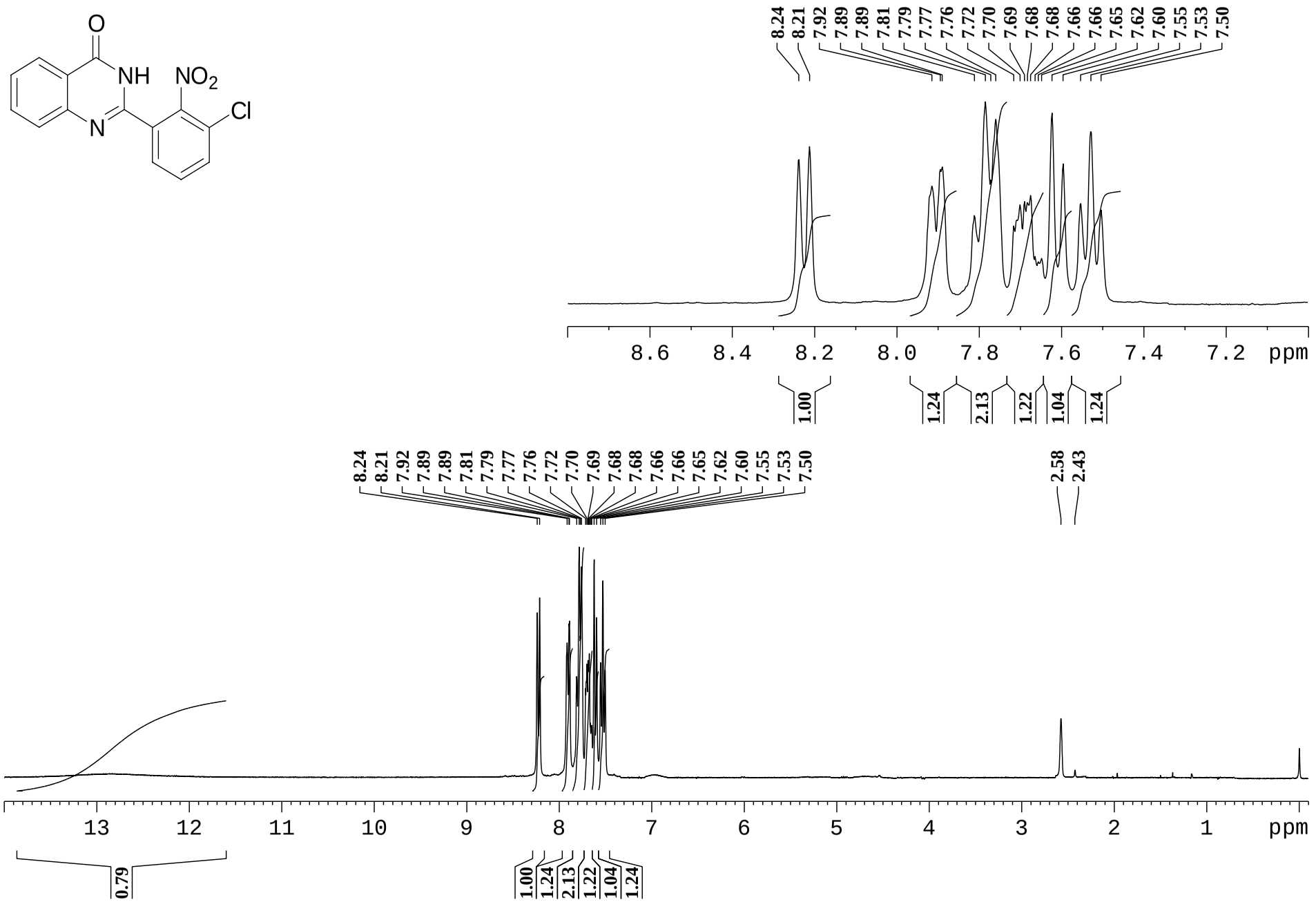




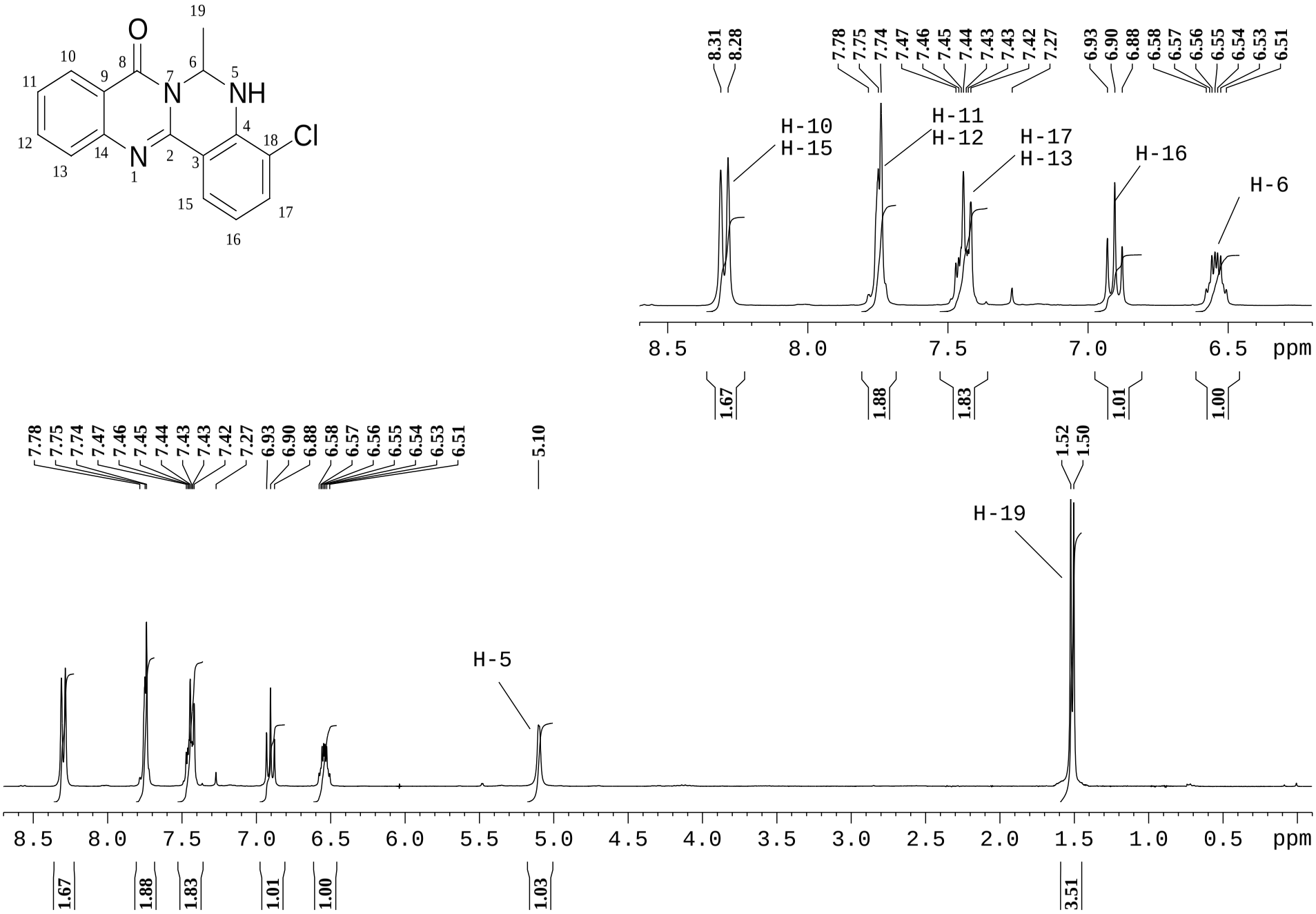
S-4: 13 C NMR Spectrum of N-[2-(aminocarbonyl)phenyl]-2-nitro benzamide (3a).



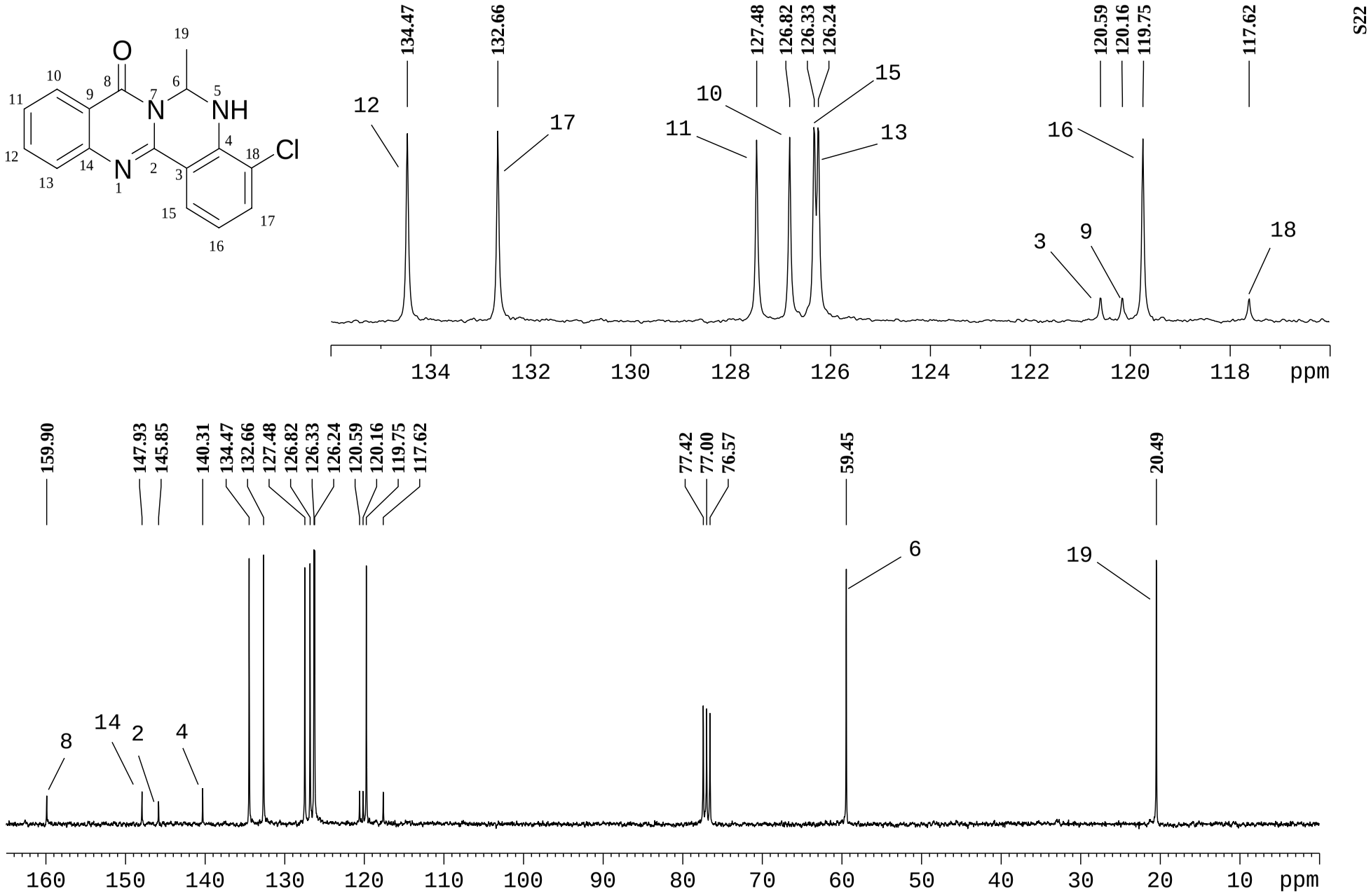
S-5: 1H NMR Spectrum of 2-(2-Nitrophenyl)-3H-quinazolin-4-one (4a).



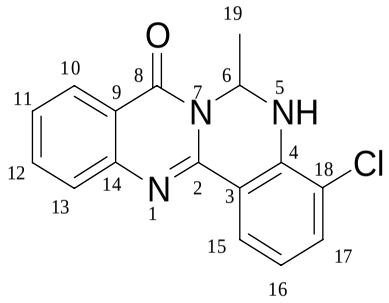
S-6: 1H NMR Spectrum of 2-(3-Chloro-2-nitrophenyl) quinazoline-4(3H)-one (4b).

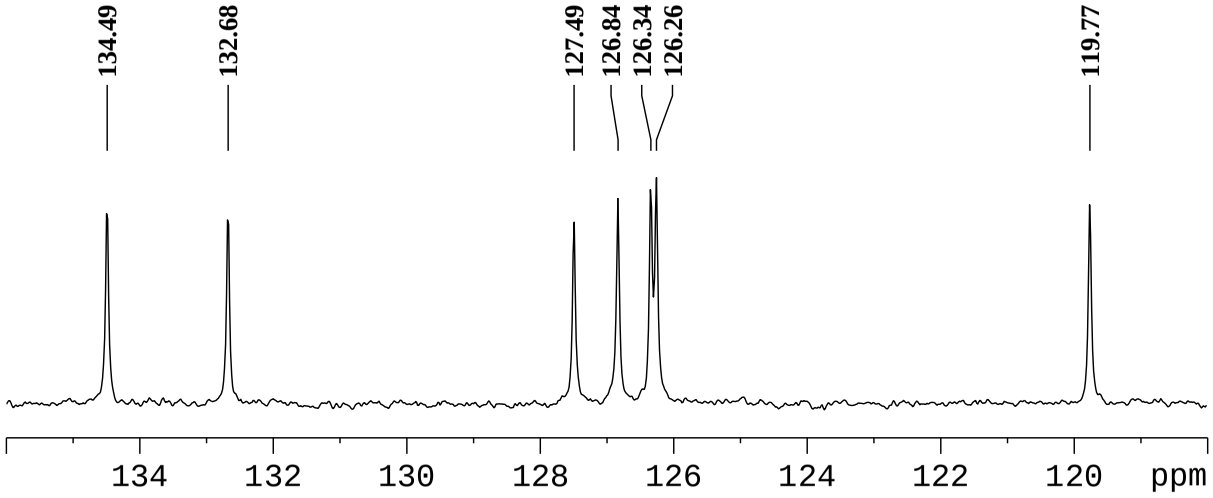


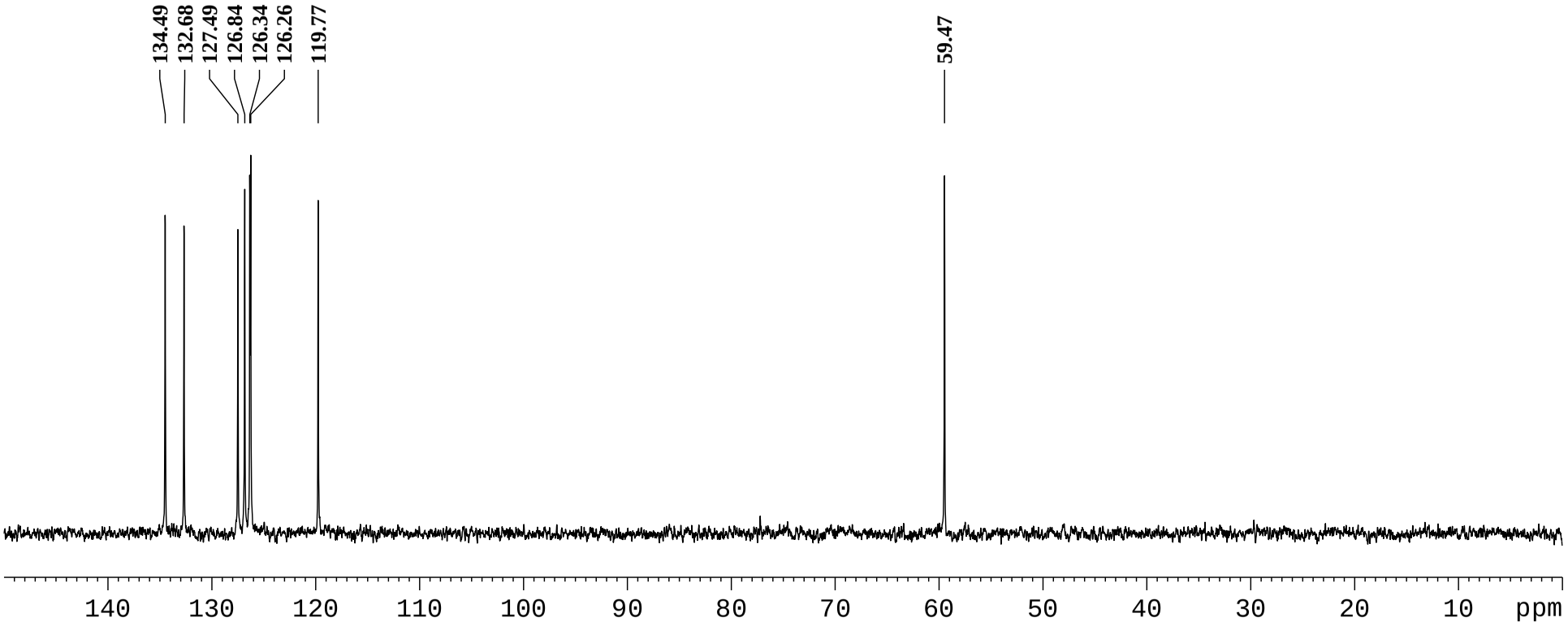
S-7: 1H NMR Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).



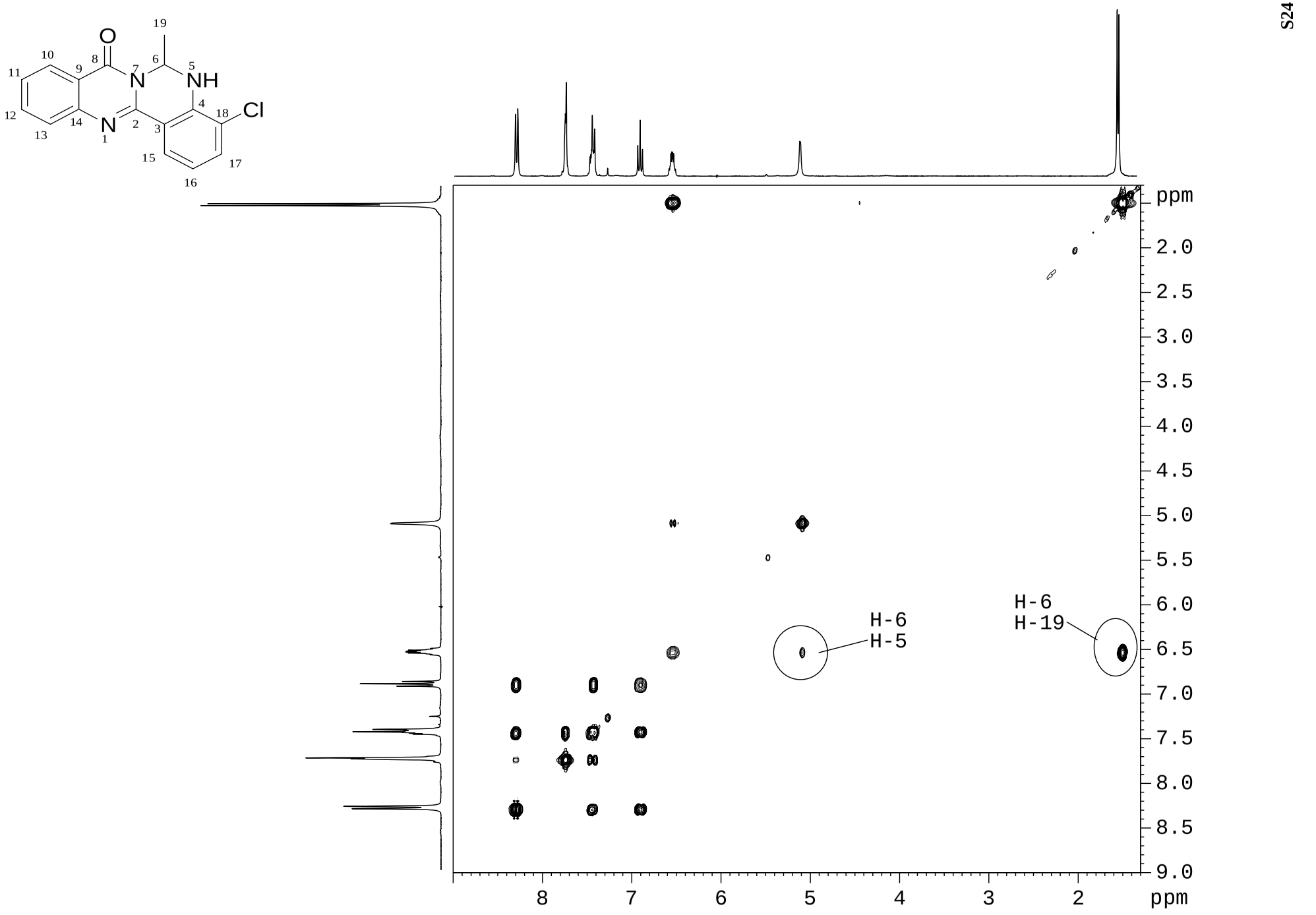
S-8: 13C NMR Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).



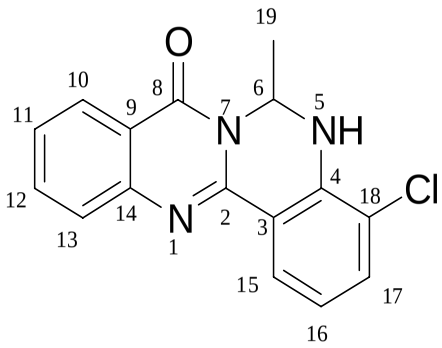


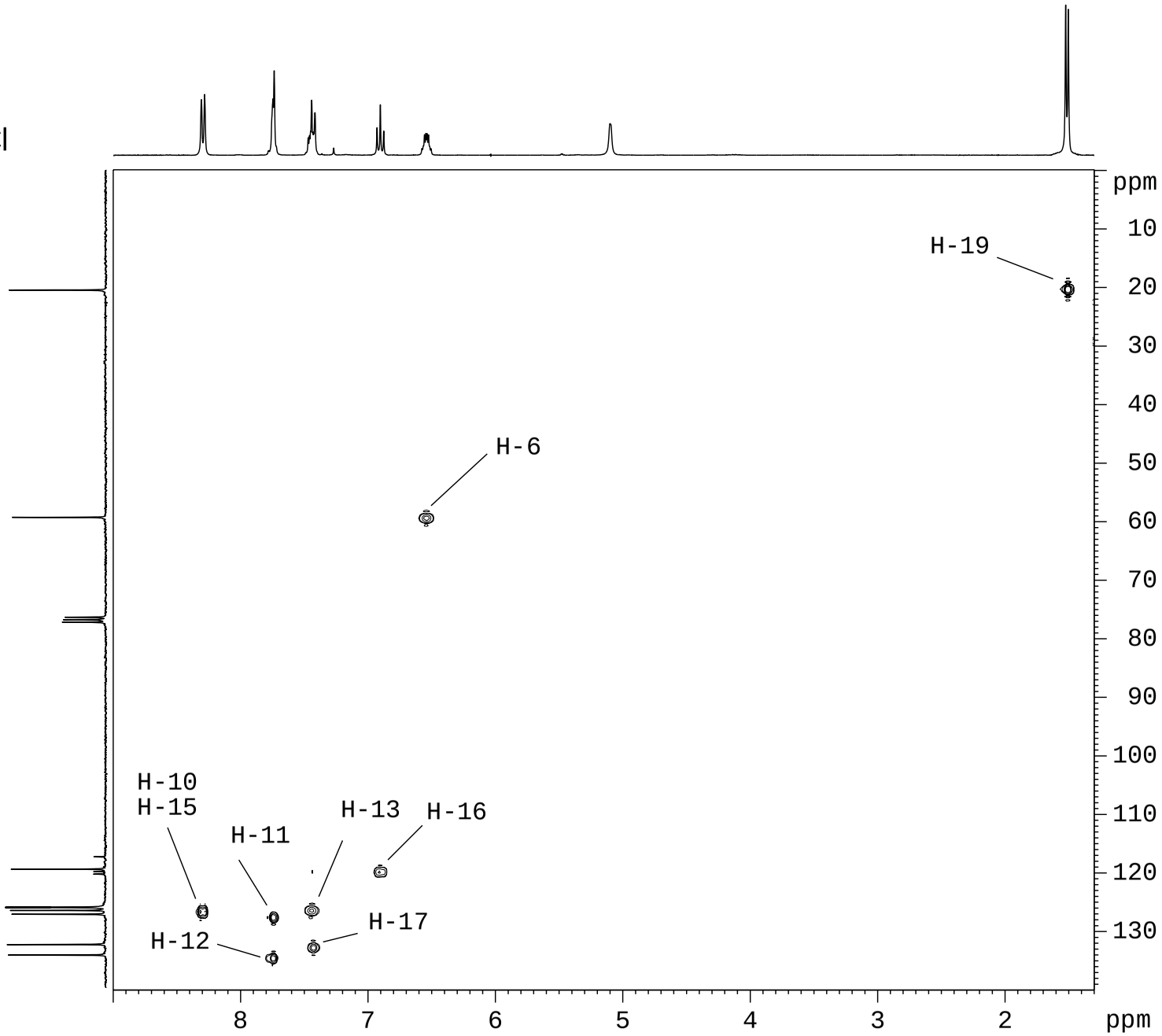


S-9: DEPT-90 NMR Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).

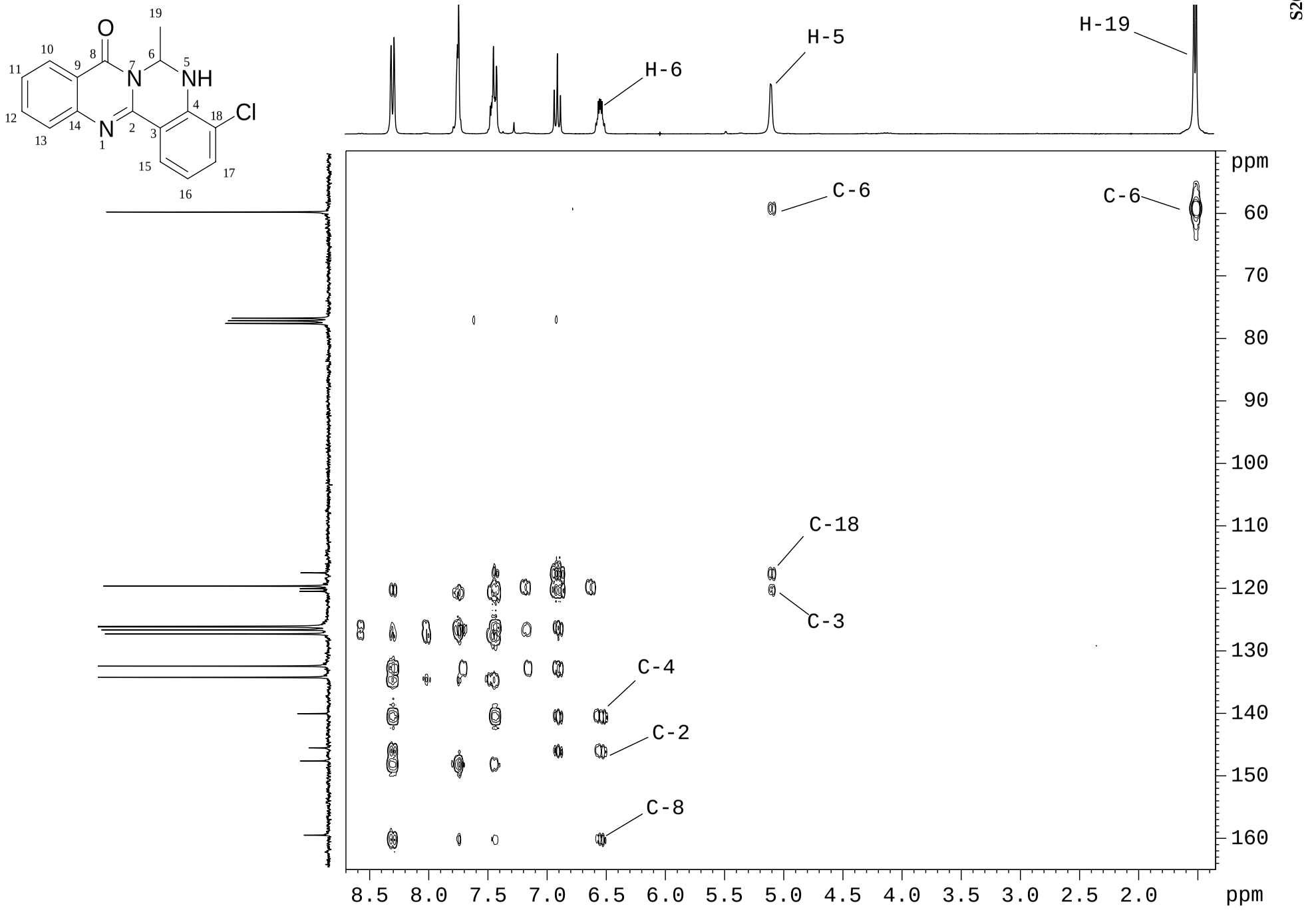


S-10: COSY Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).

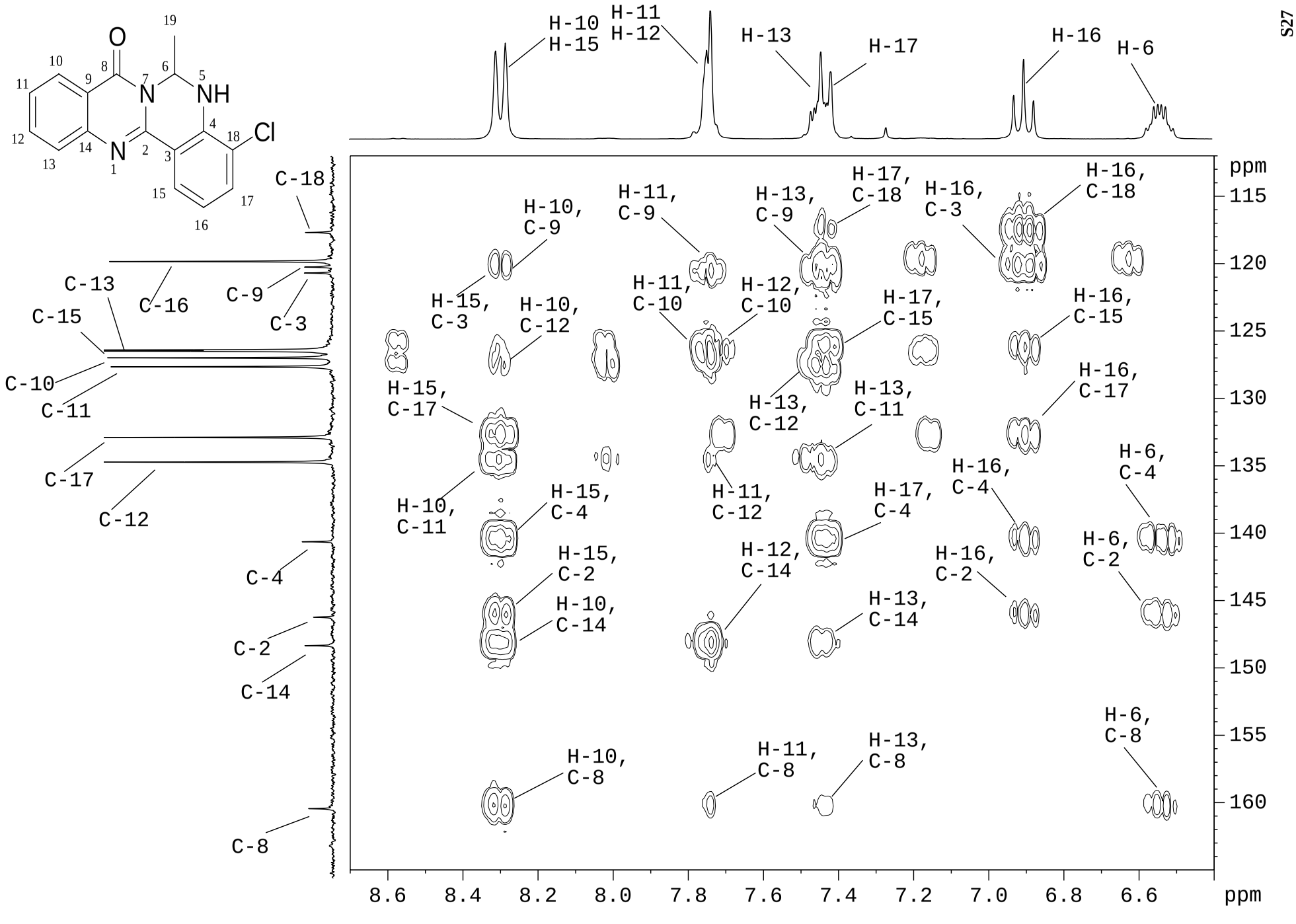




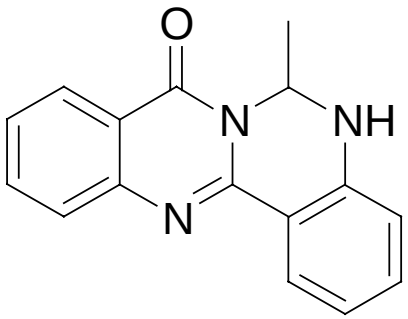
S-11: HSQC Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).

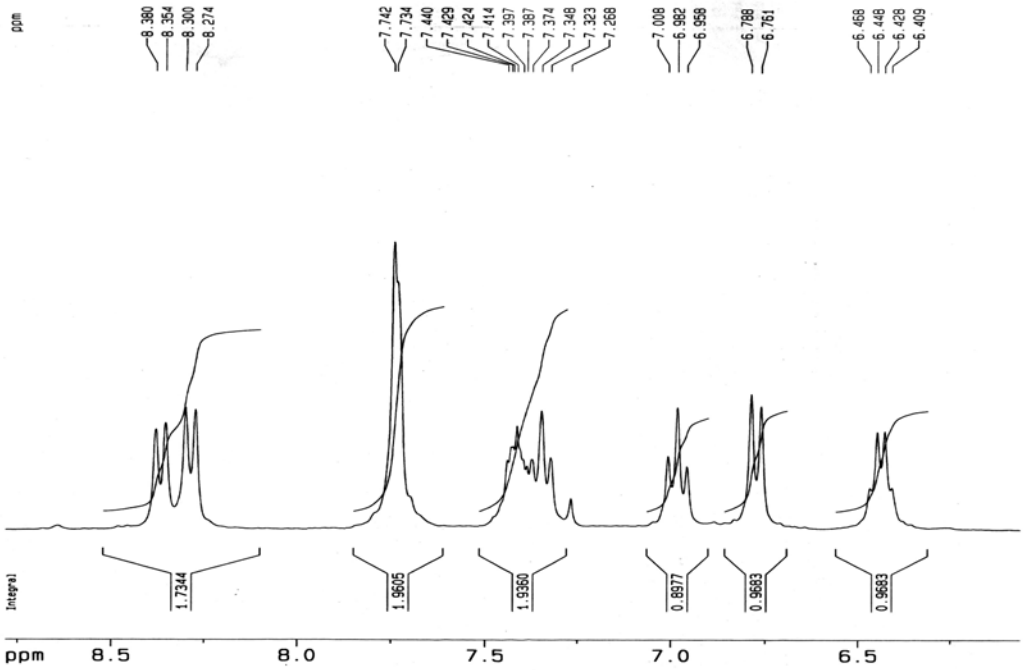


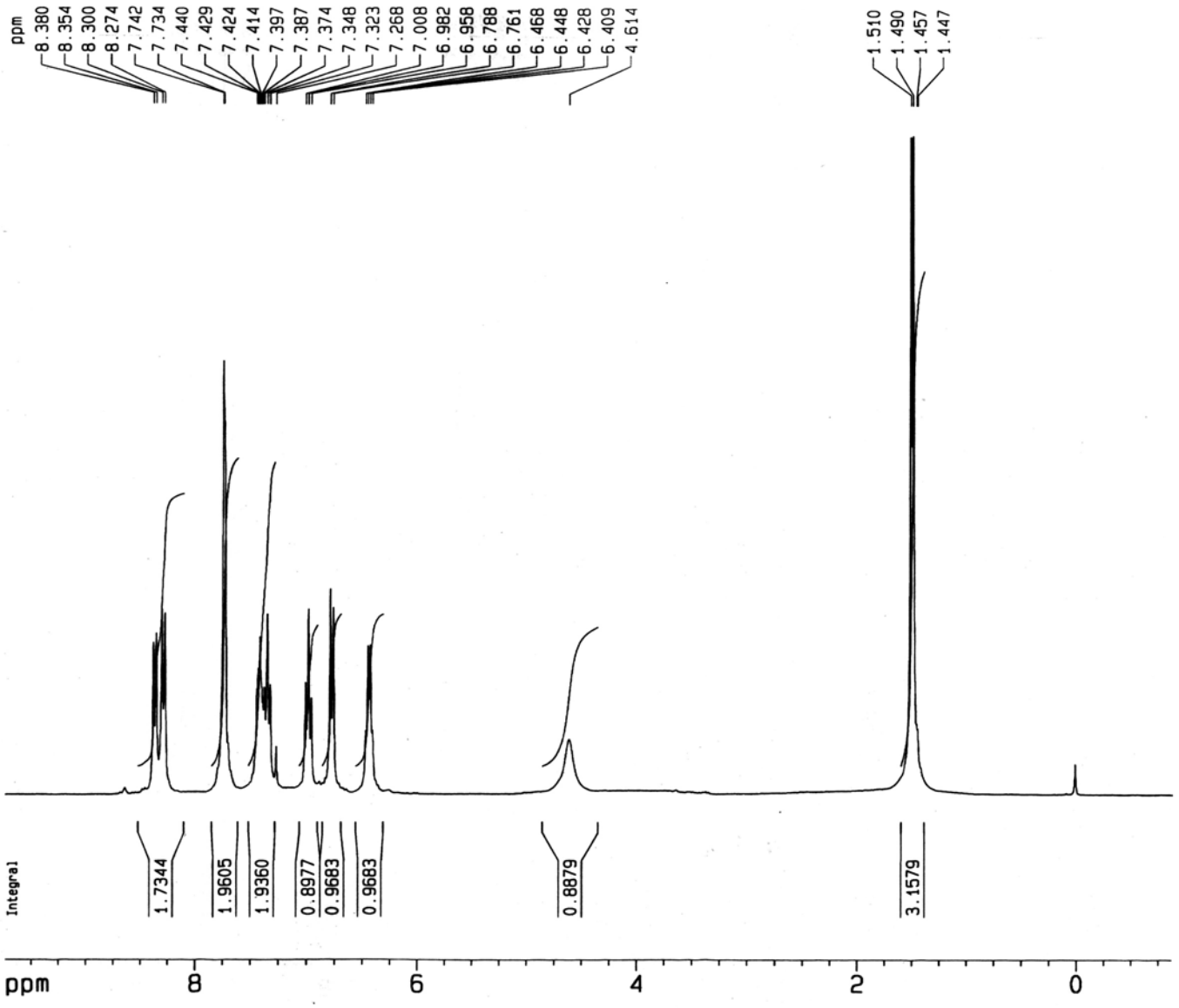
S-12: HMBC Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).



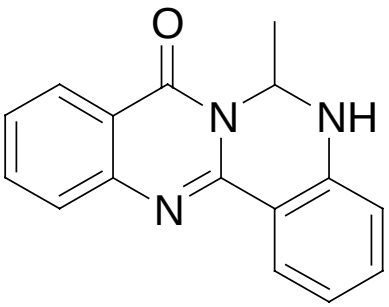
S-13: Expanded Region of the HMBC Spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5f).

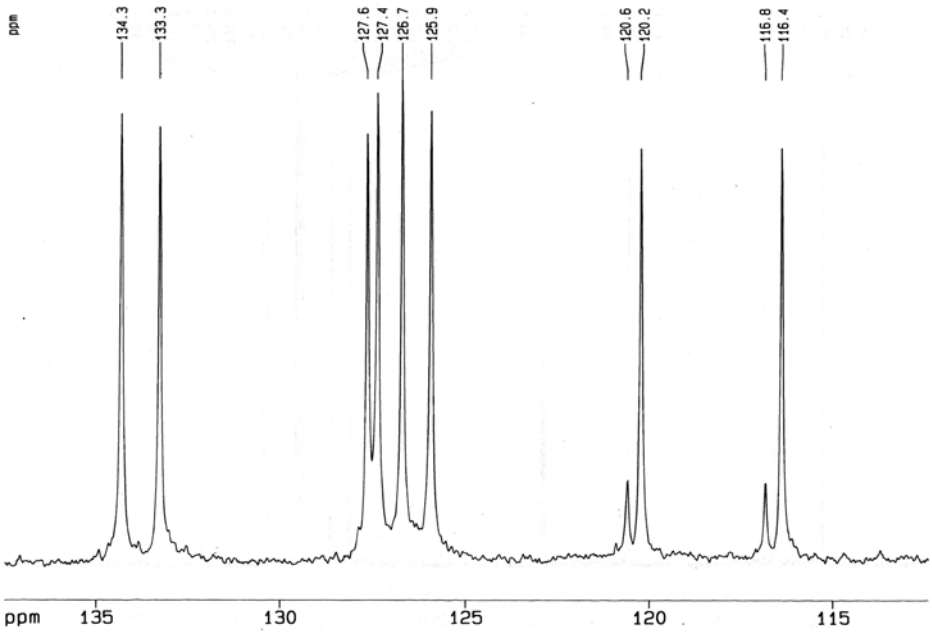


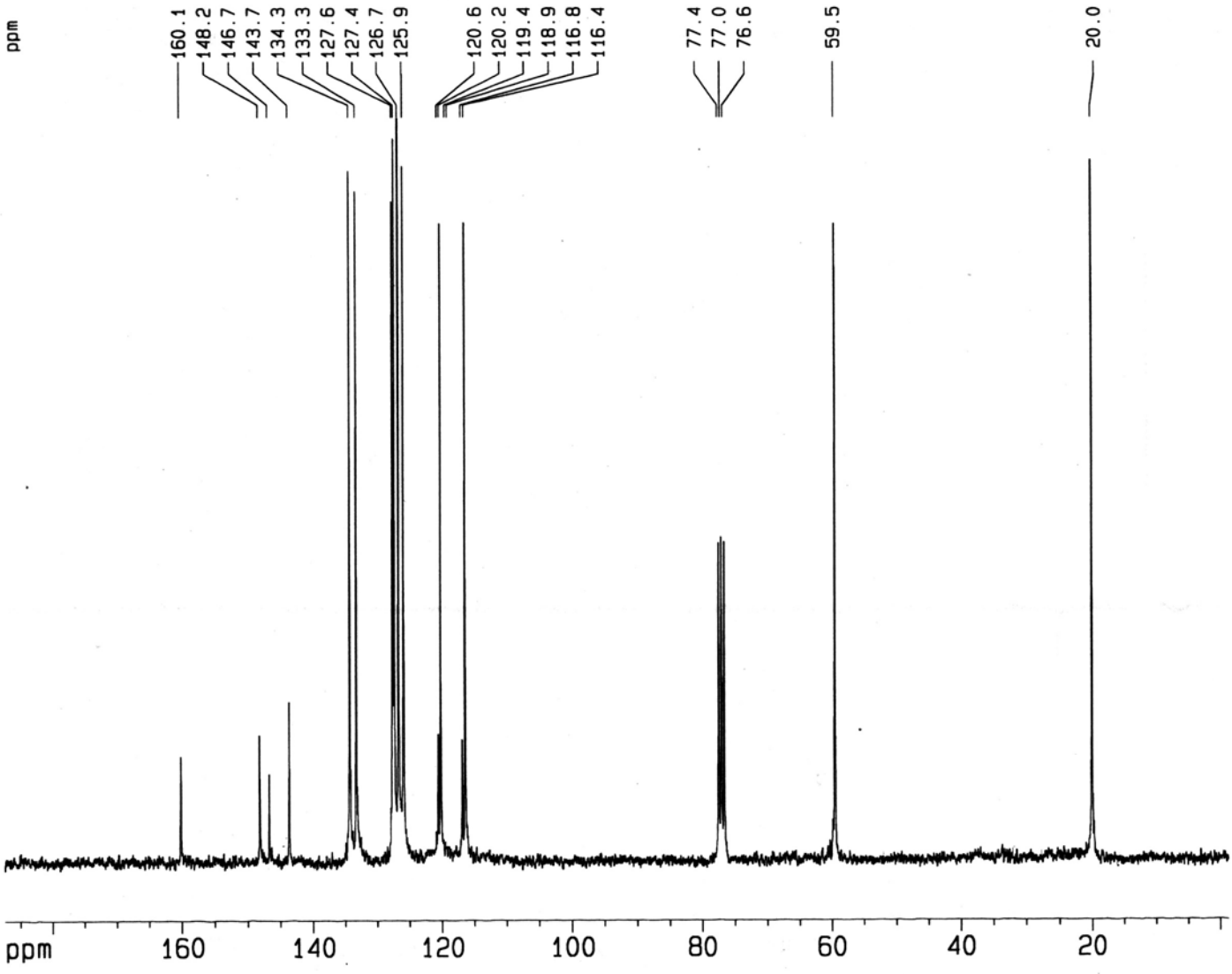




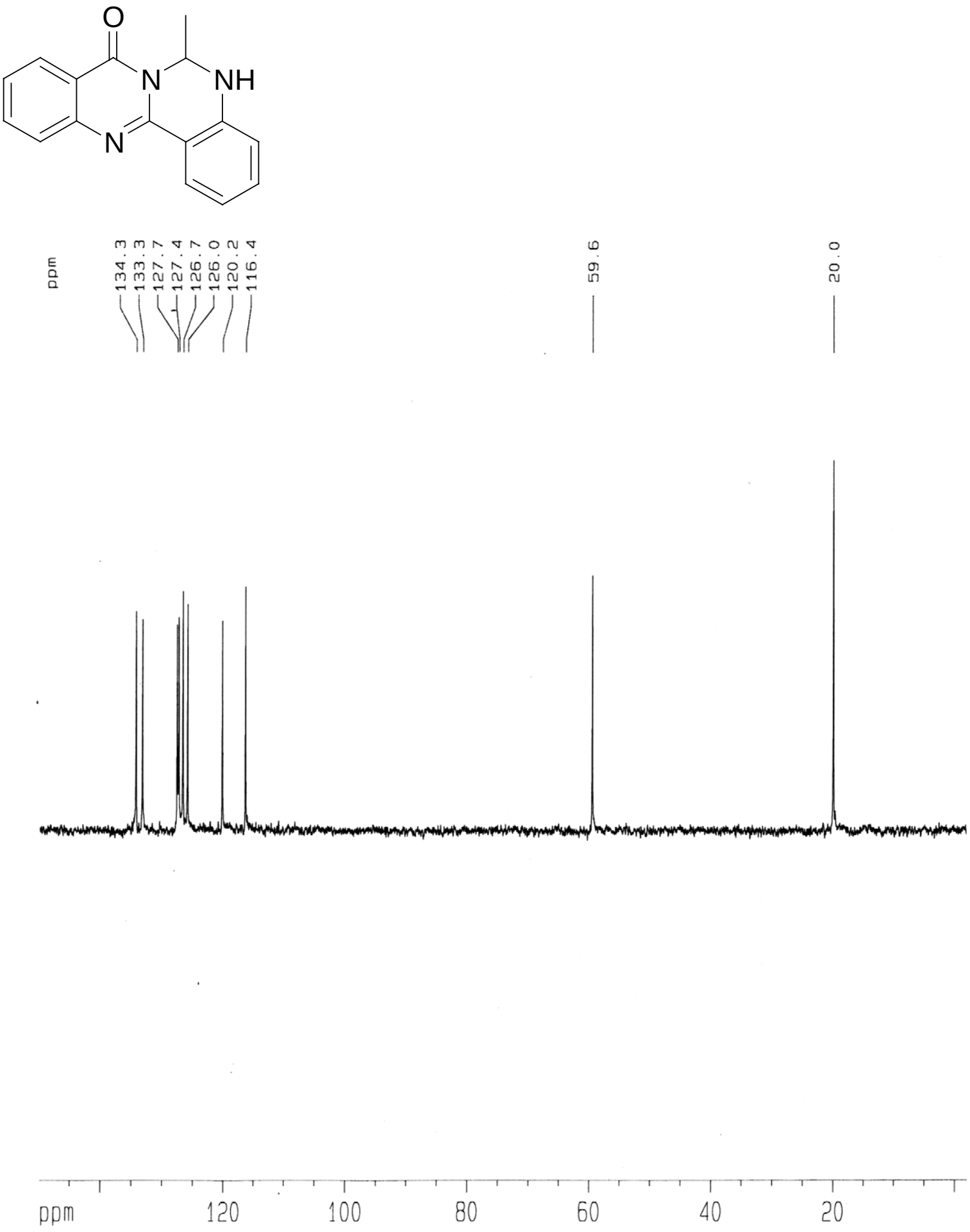
S-14: 1H NMR Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a).



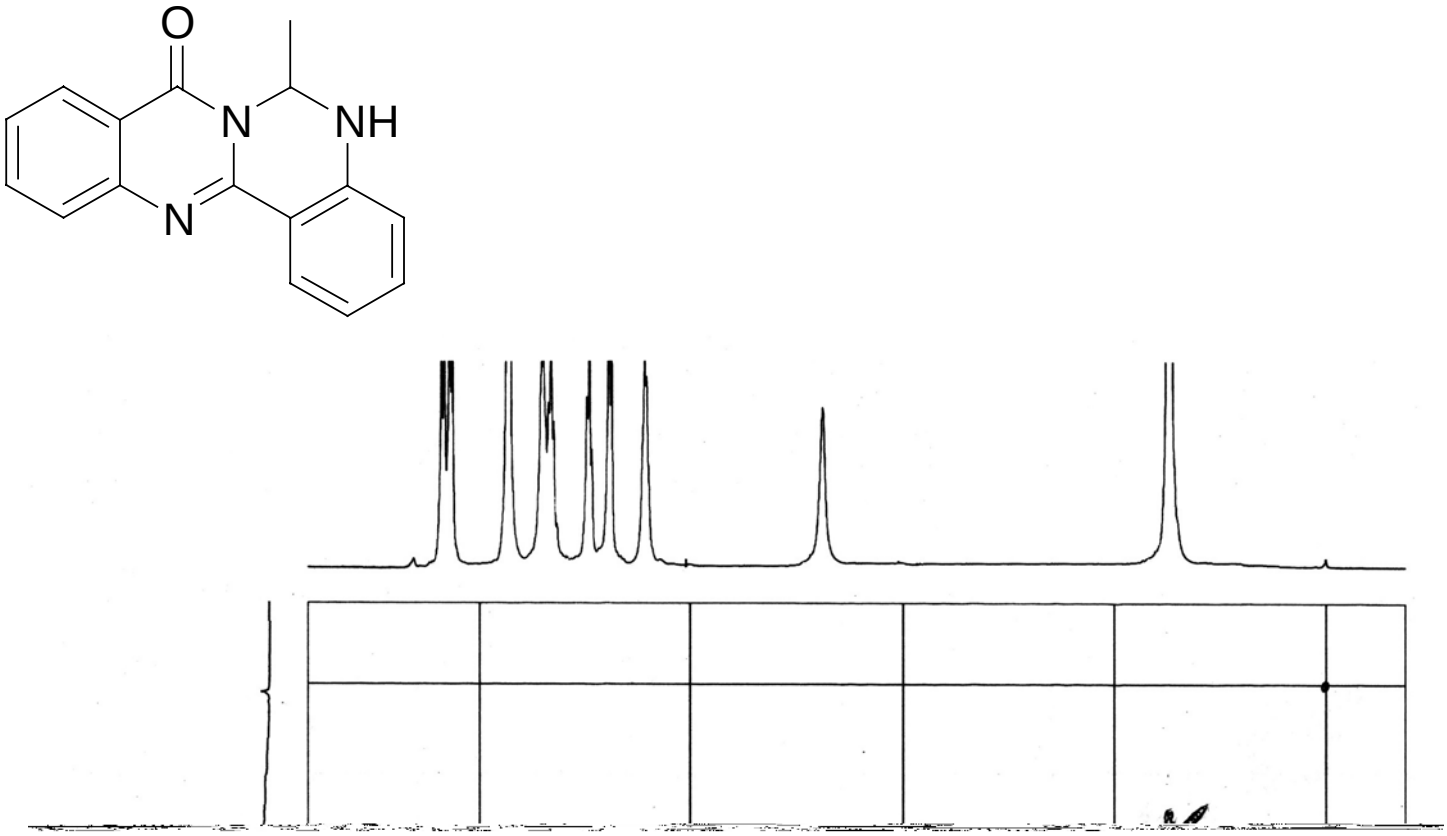




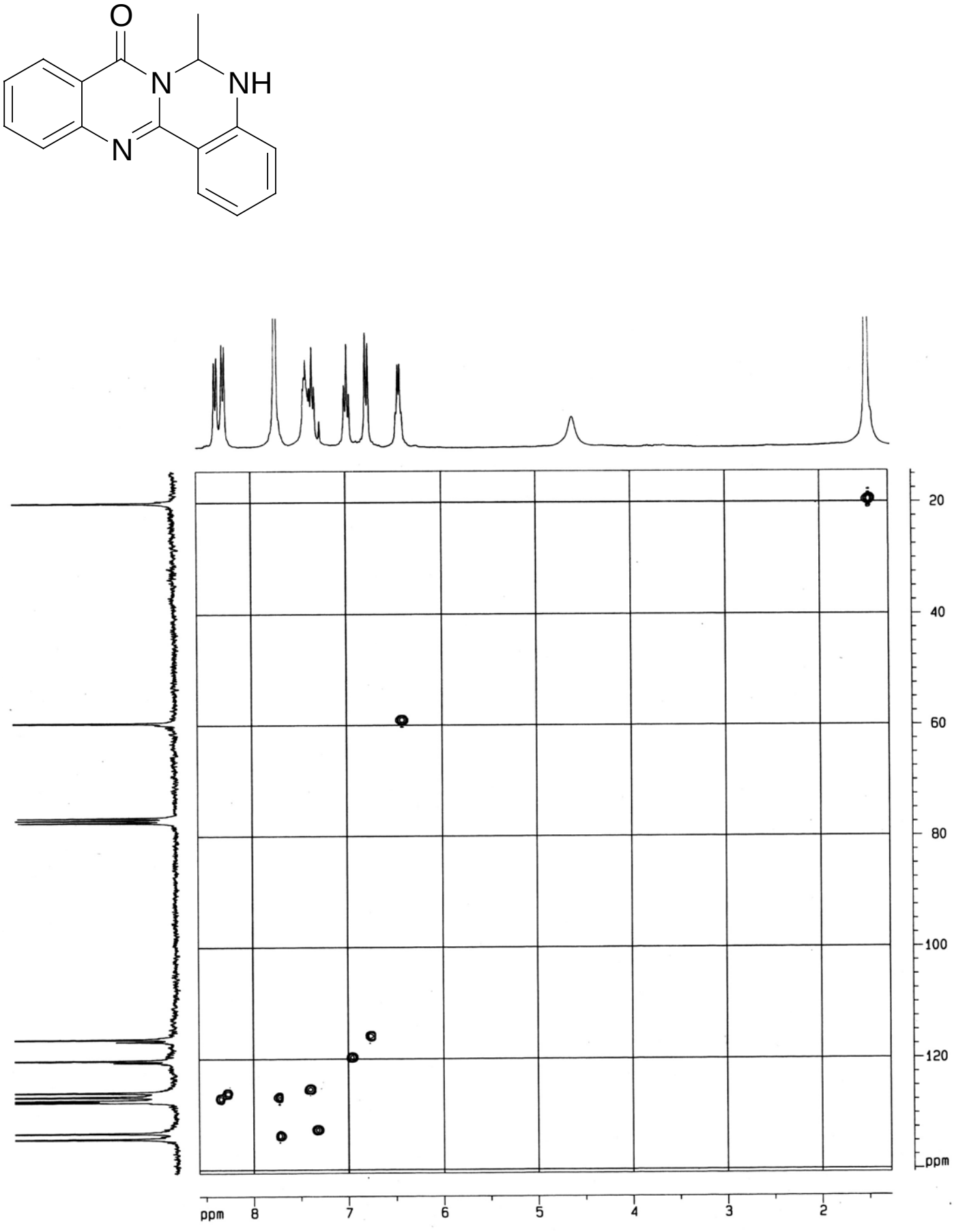
S-15: 13C NMR Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a).



S-16: DEPT-135 NMR Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a).

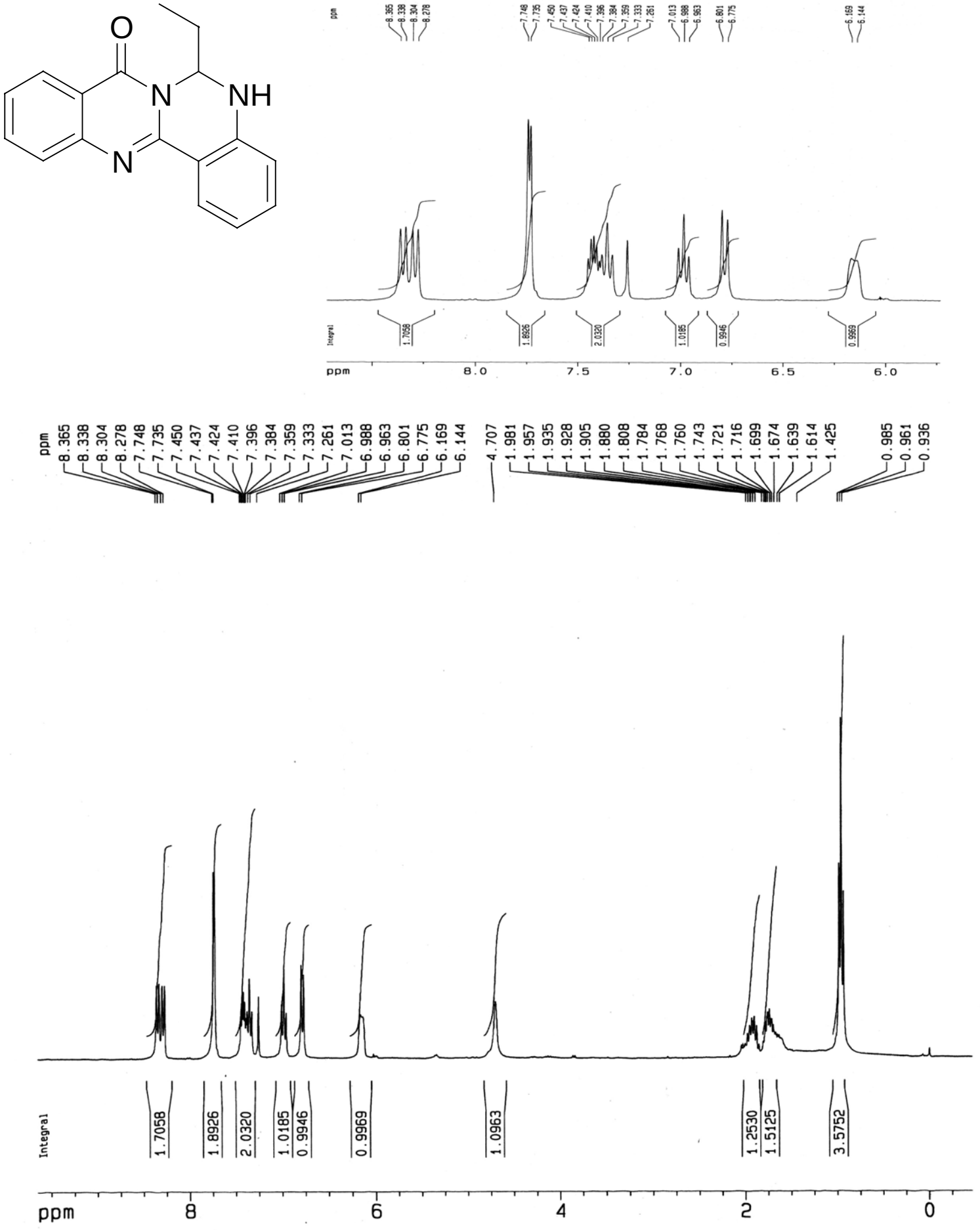


S-17: COSY Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a).

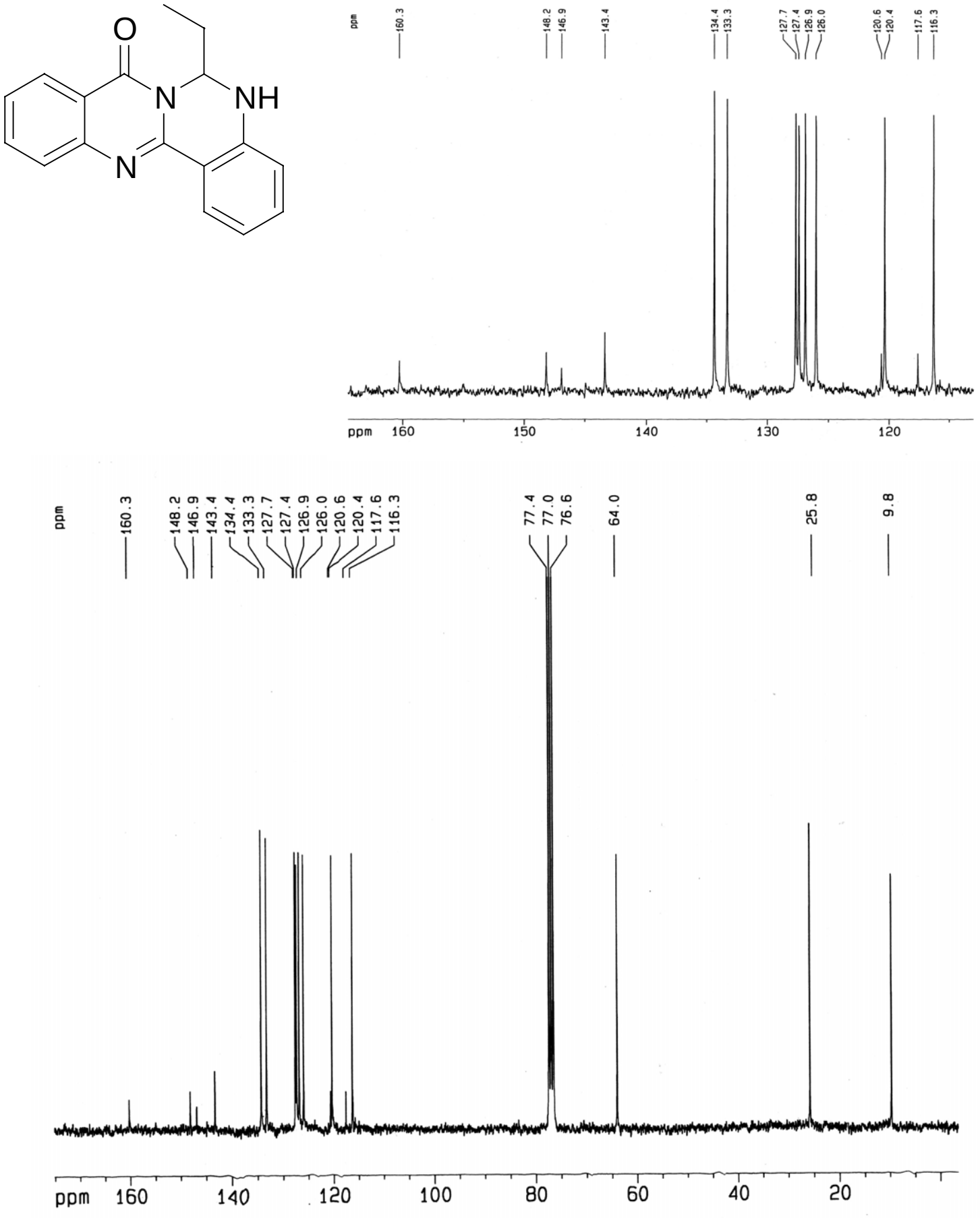


S-18: HSQC Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a).

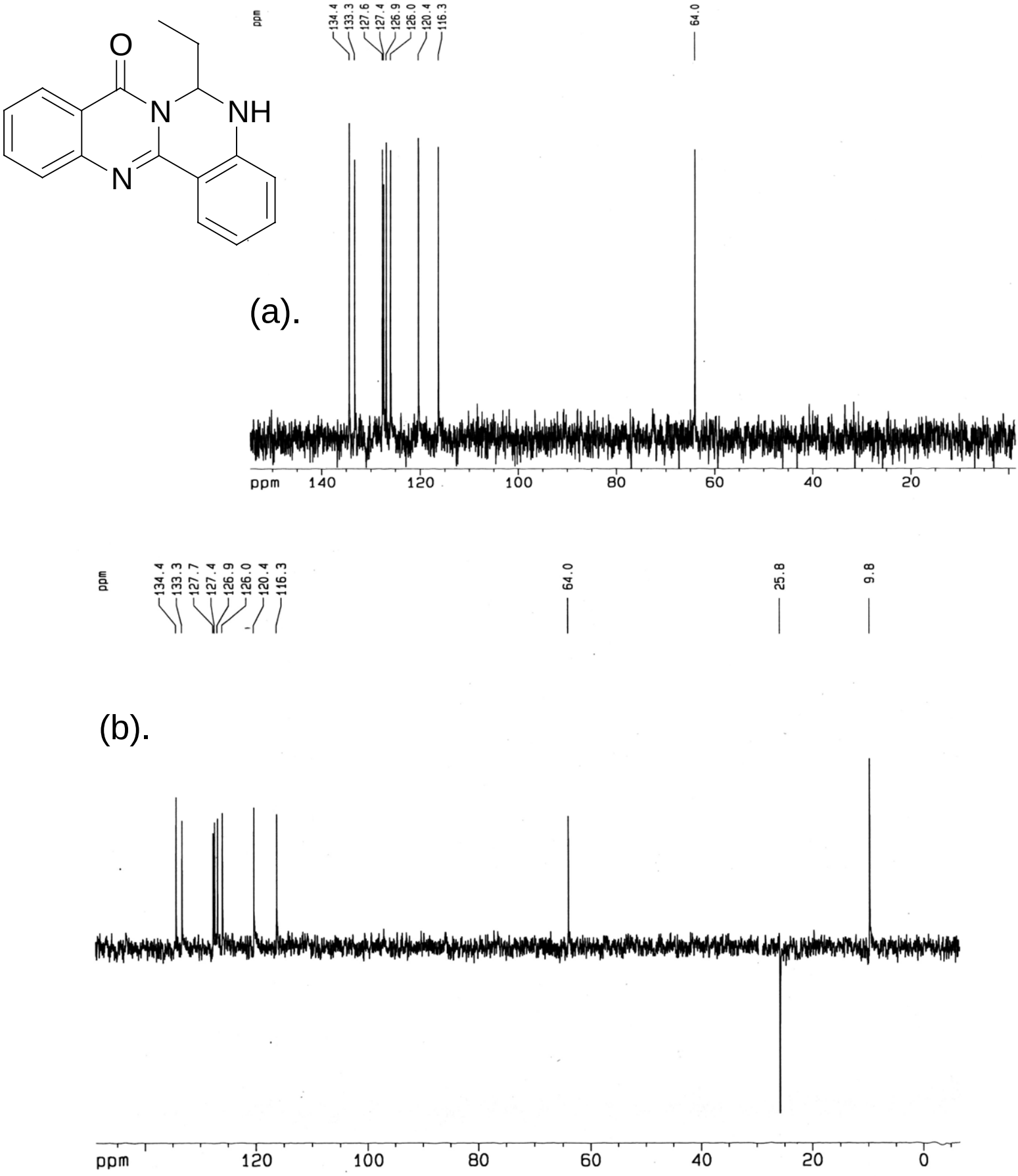
S-19: HMBC Spectrum of 6-Methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5a).



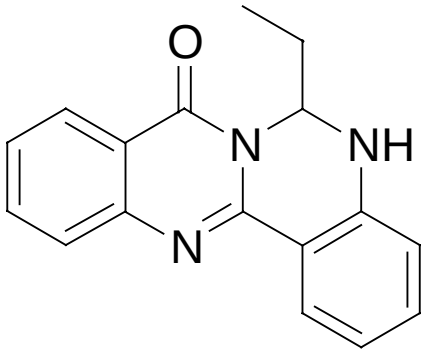
S-20: 1H NMR Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5b).

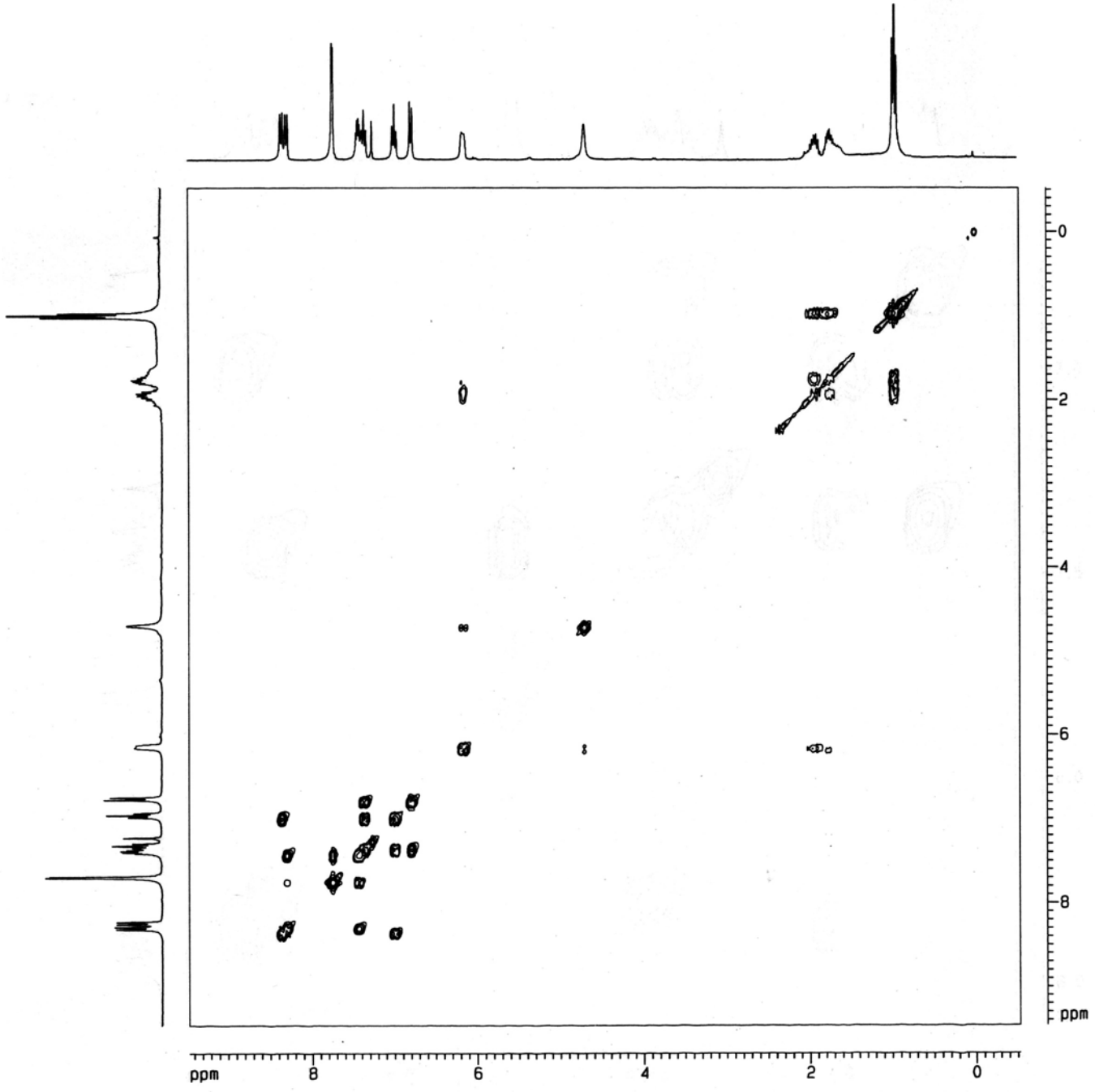


S-21: 13C NMR Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5b).

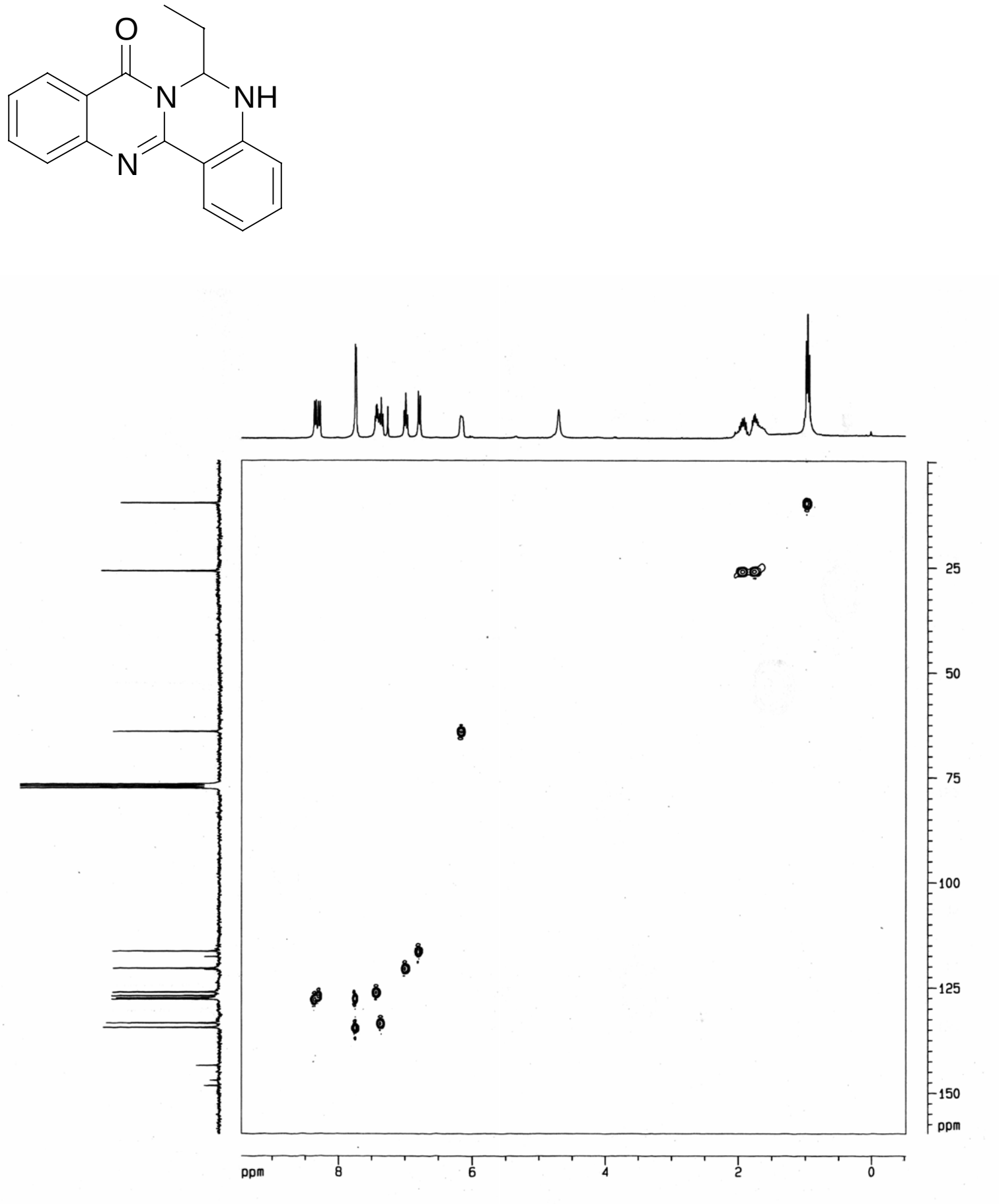


S-22: (a). DEPT-90 and (b). DEPT-90 NMR Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5b).

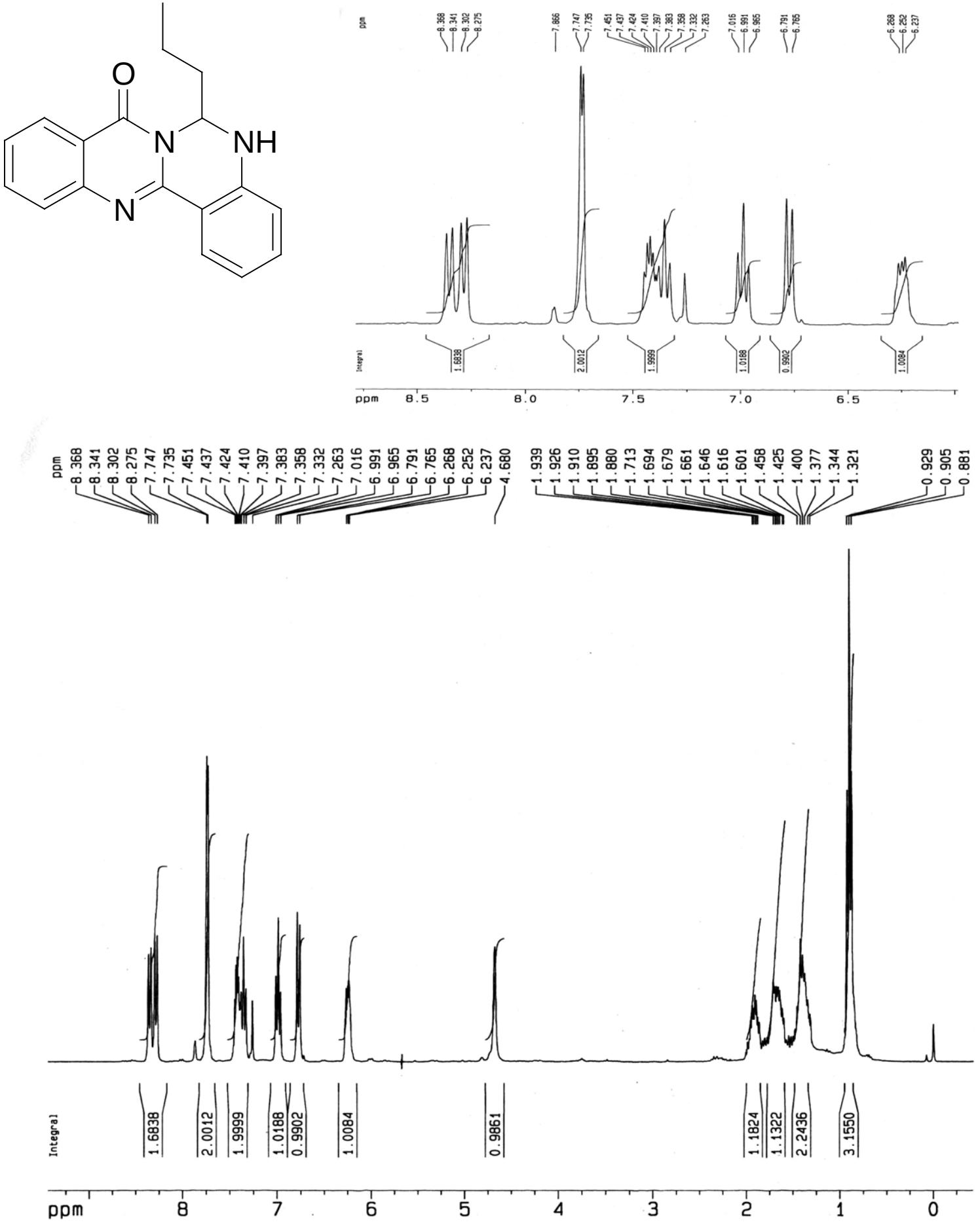




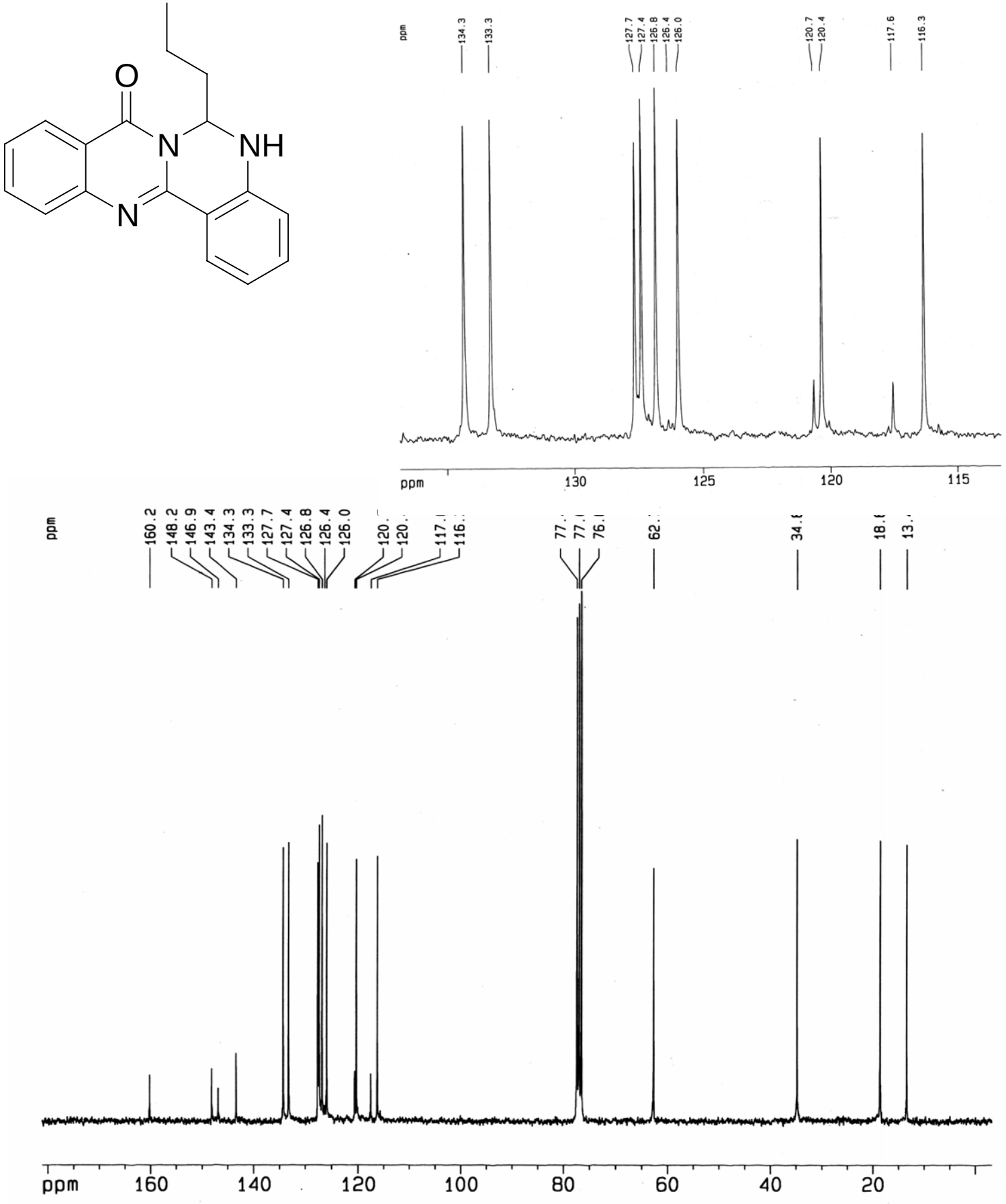
S-23: COSY Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5b).



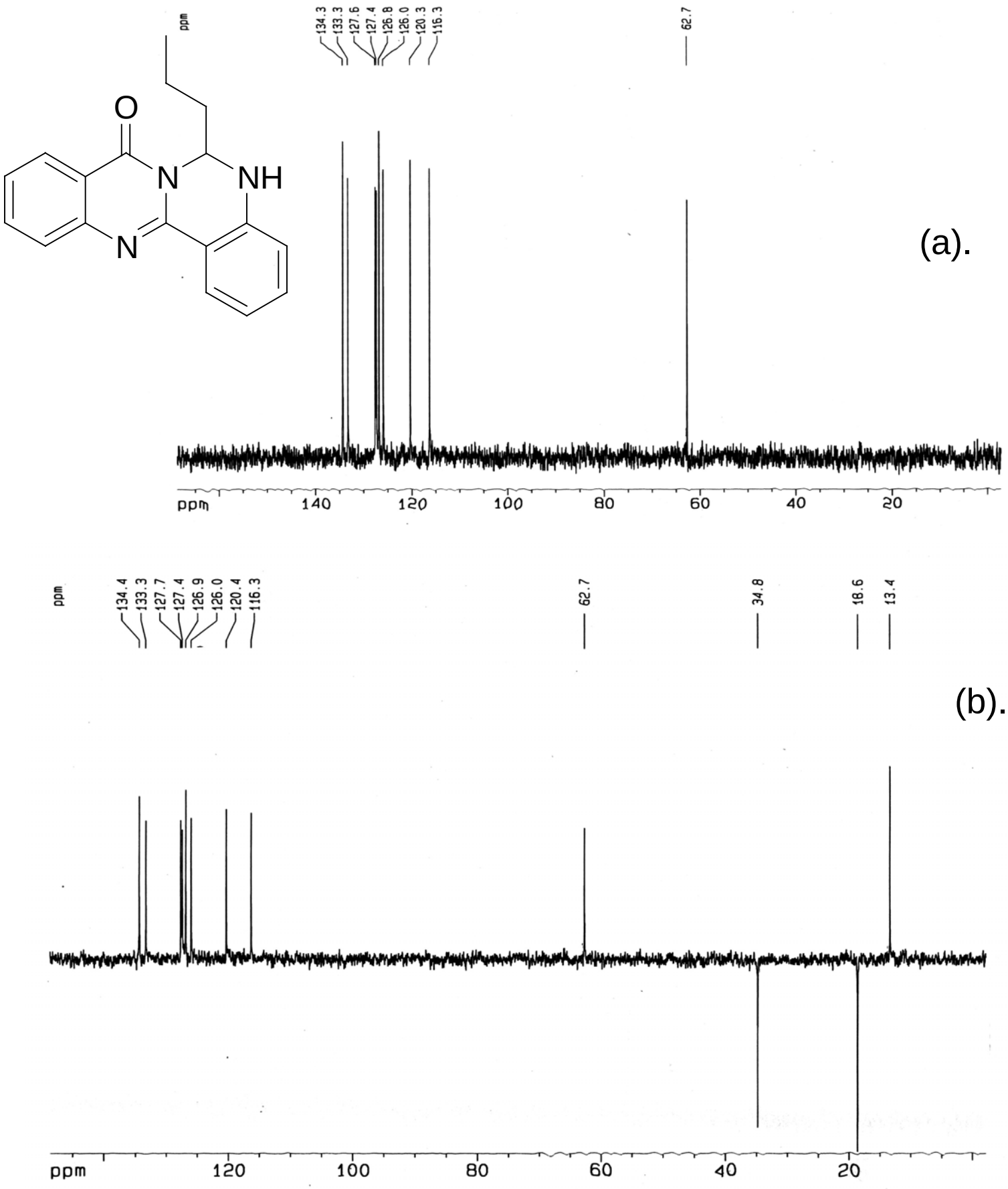
S-24: HSQC Spectrum of 6-Ethyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5b).



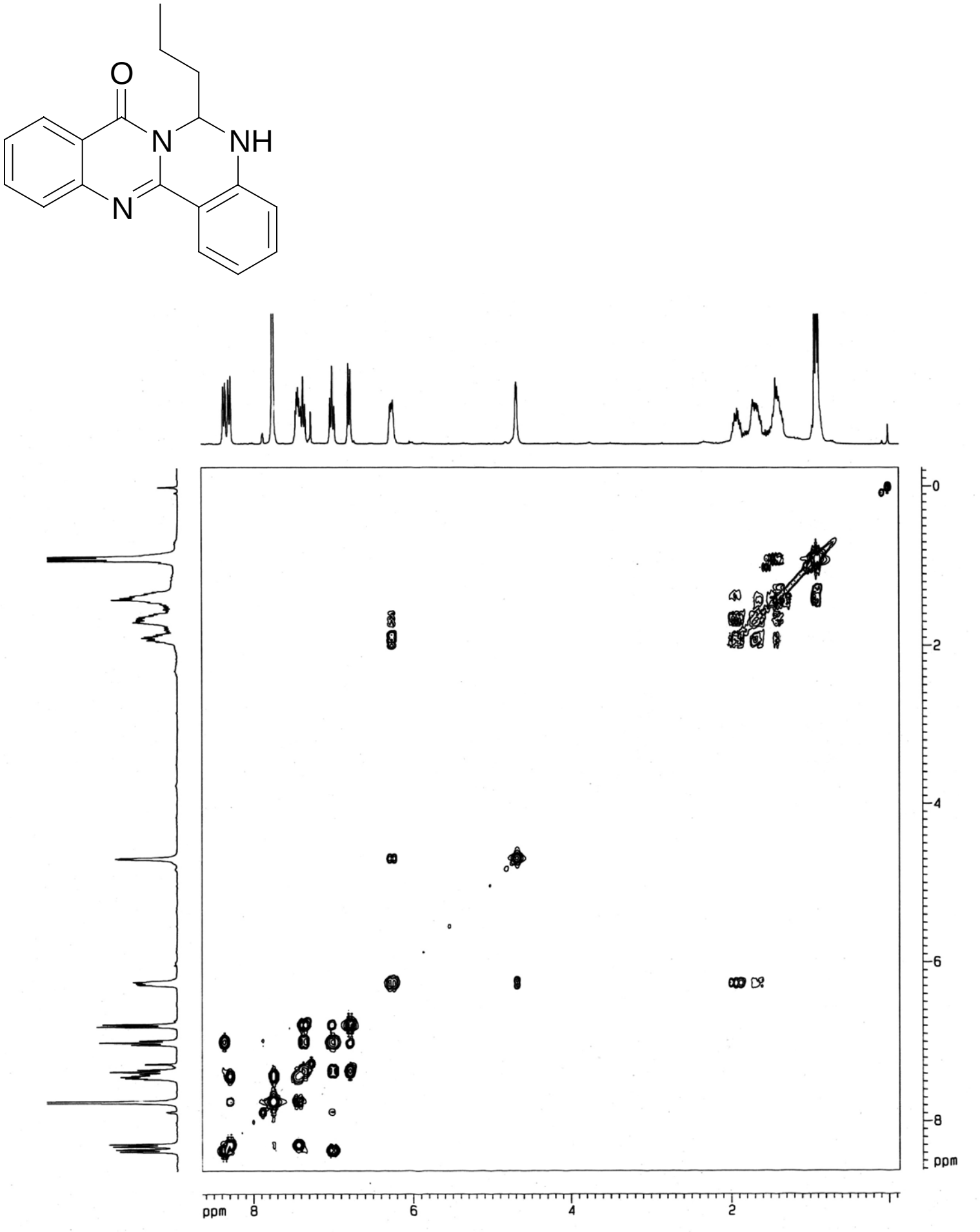
S-25: 1H NMR Spectrum of 6-Propyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5c).



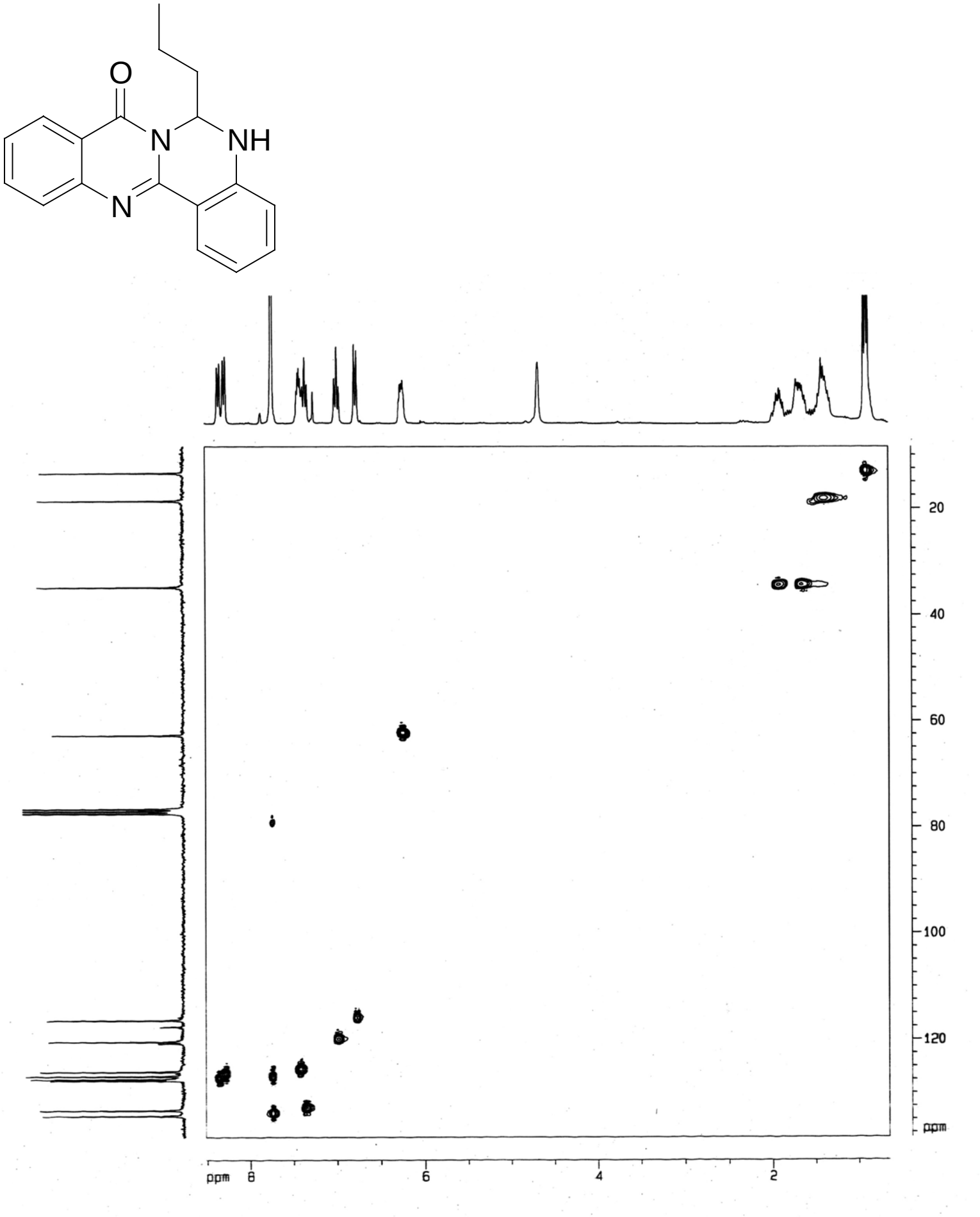
S-26: 13C NMR Spectrum of 6-Propyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5c).



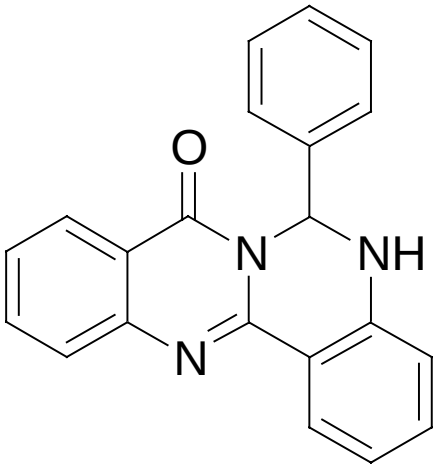
S-27: (a). DEPT-90 and (b). DEPT-135 NMR Spectrum of 6-Propyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5c).

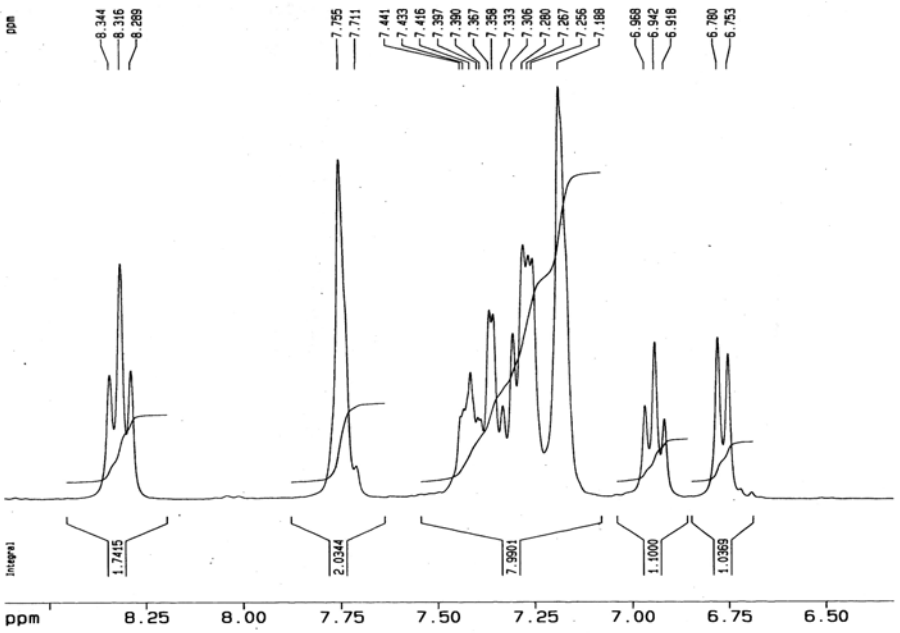


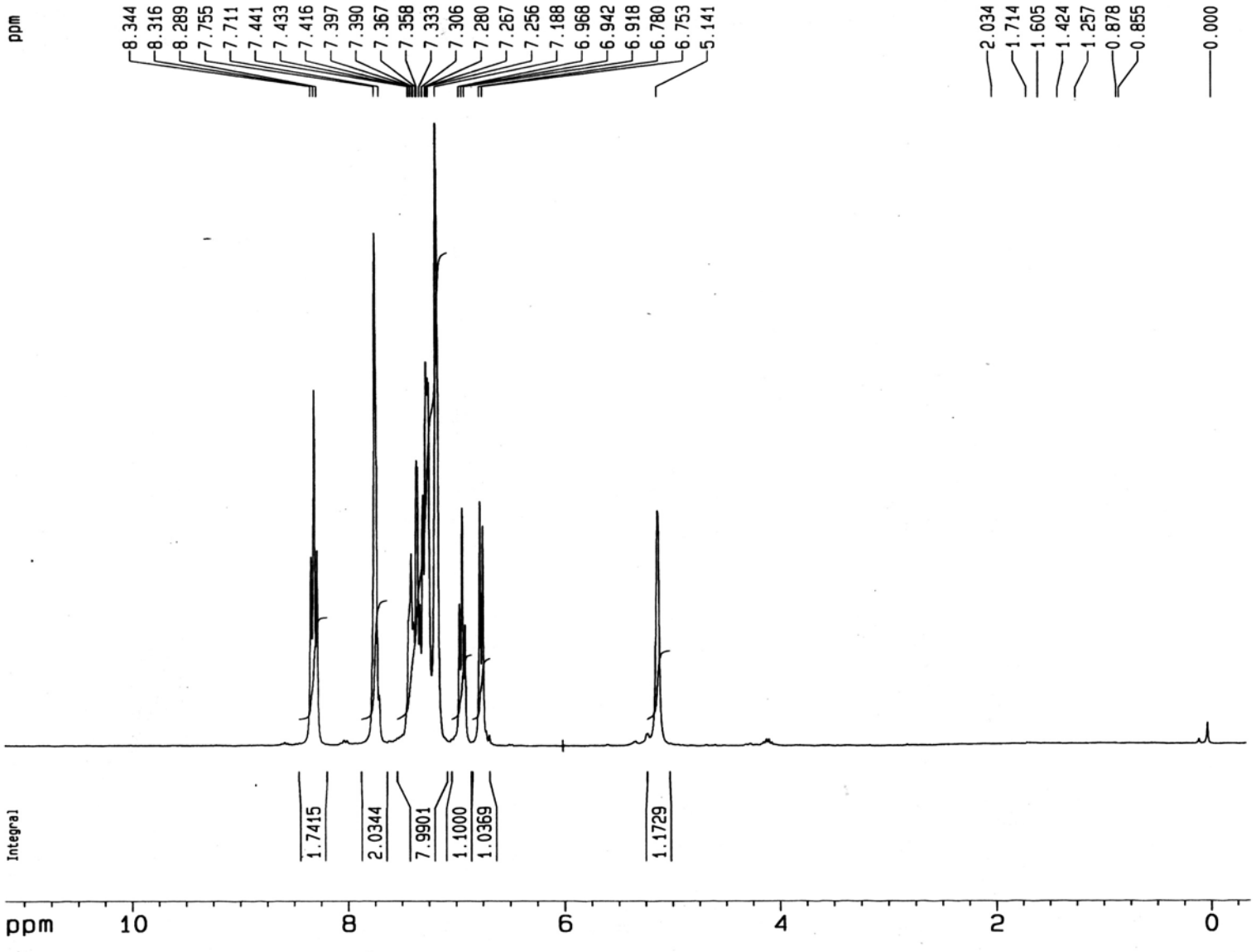
S-28: COSY Spectrum of 6-Propyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5c).



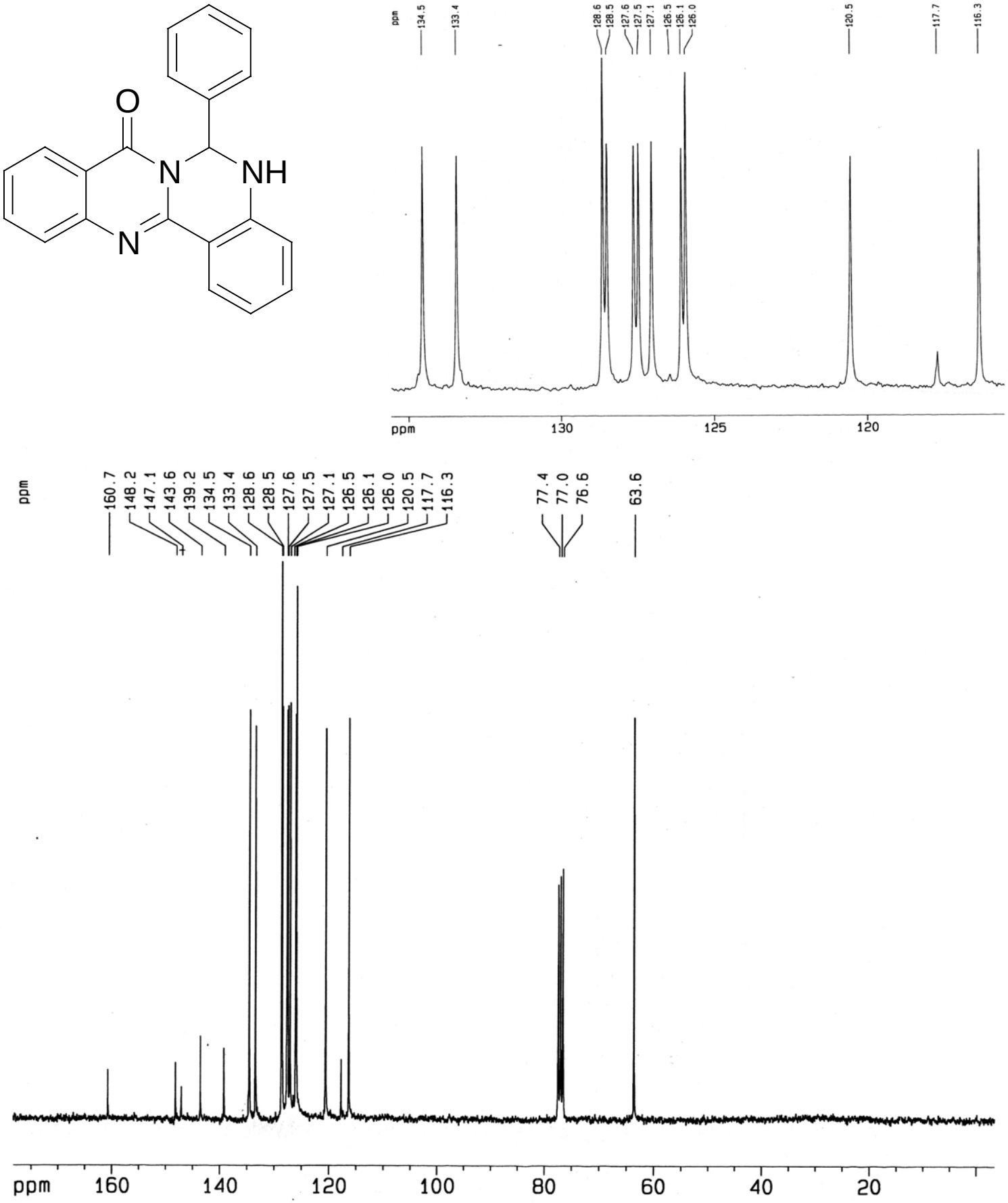
S-29: HSQC Spectrum of 6-Propyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5c).



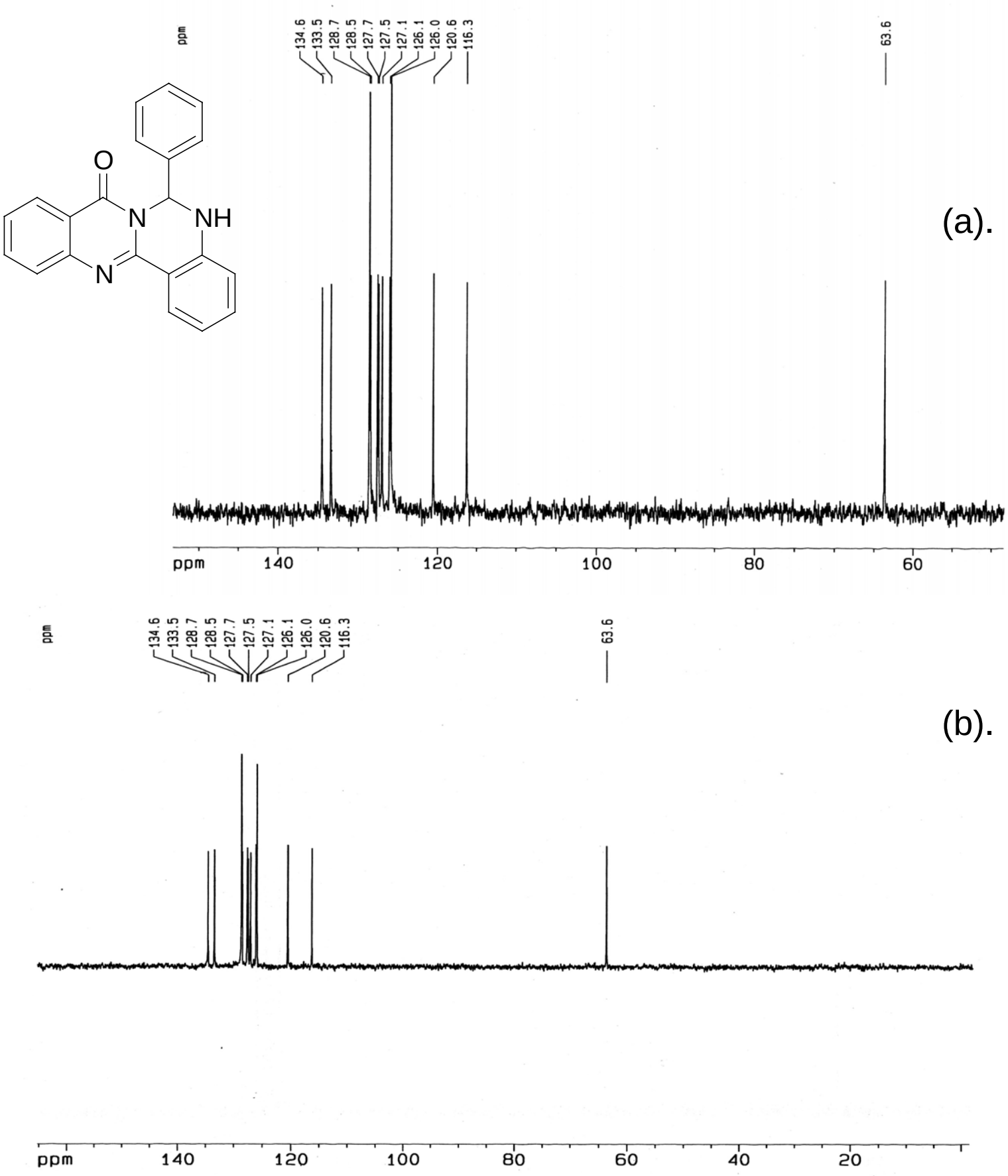




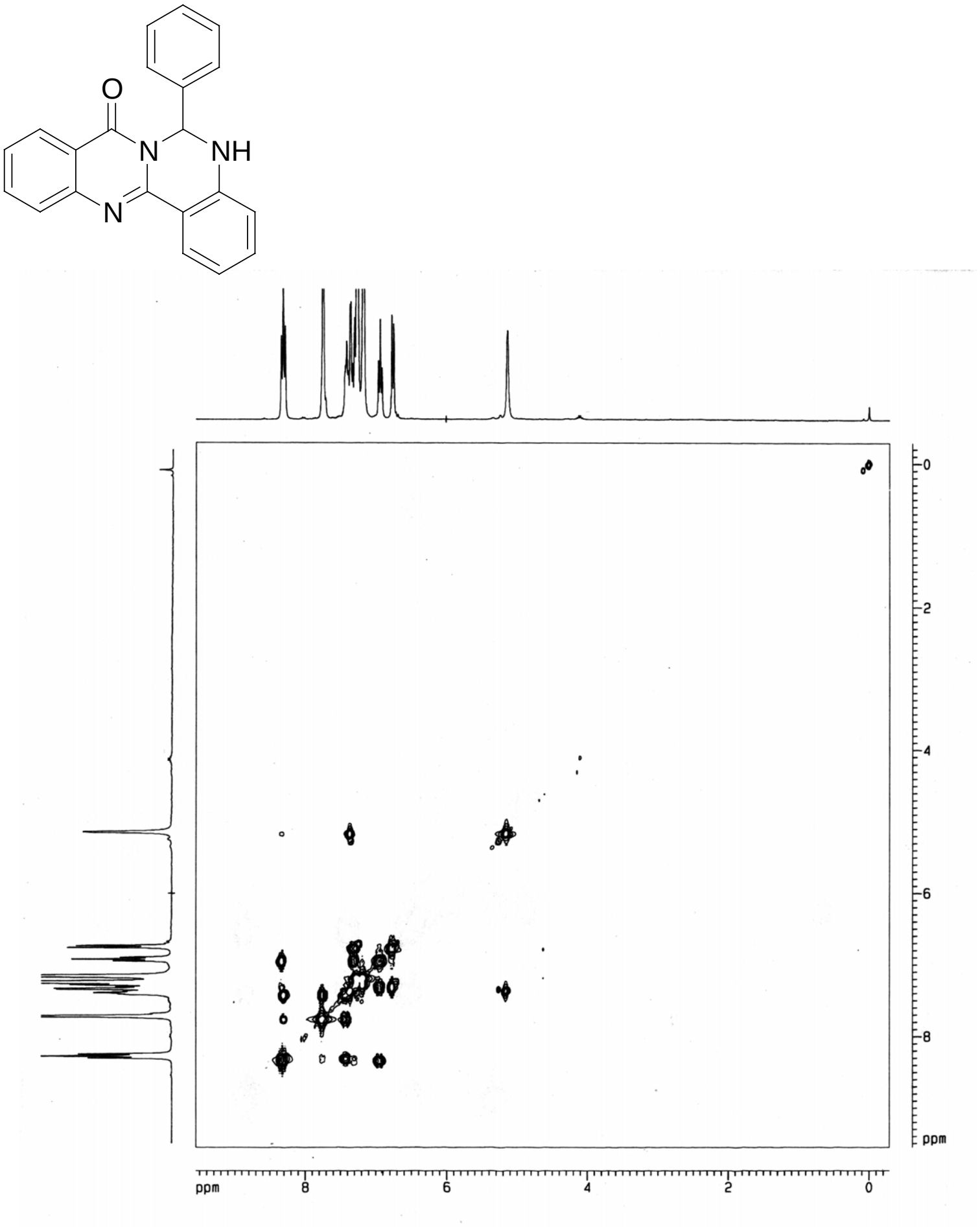
S-30: 1H NMR Spectrum of 6-Phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5d).



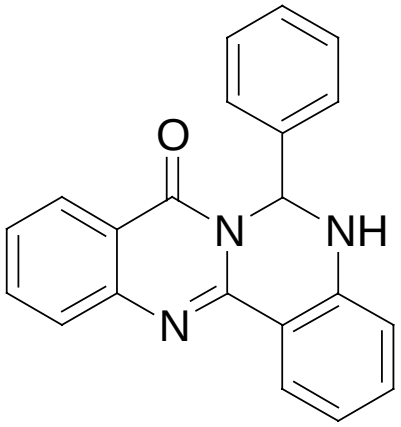
S-31: 13C NMR Spectrum of 6-Phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5d).

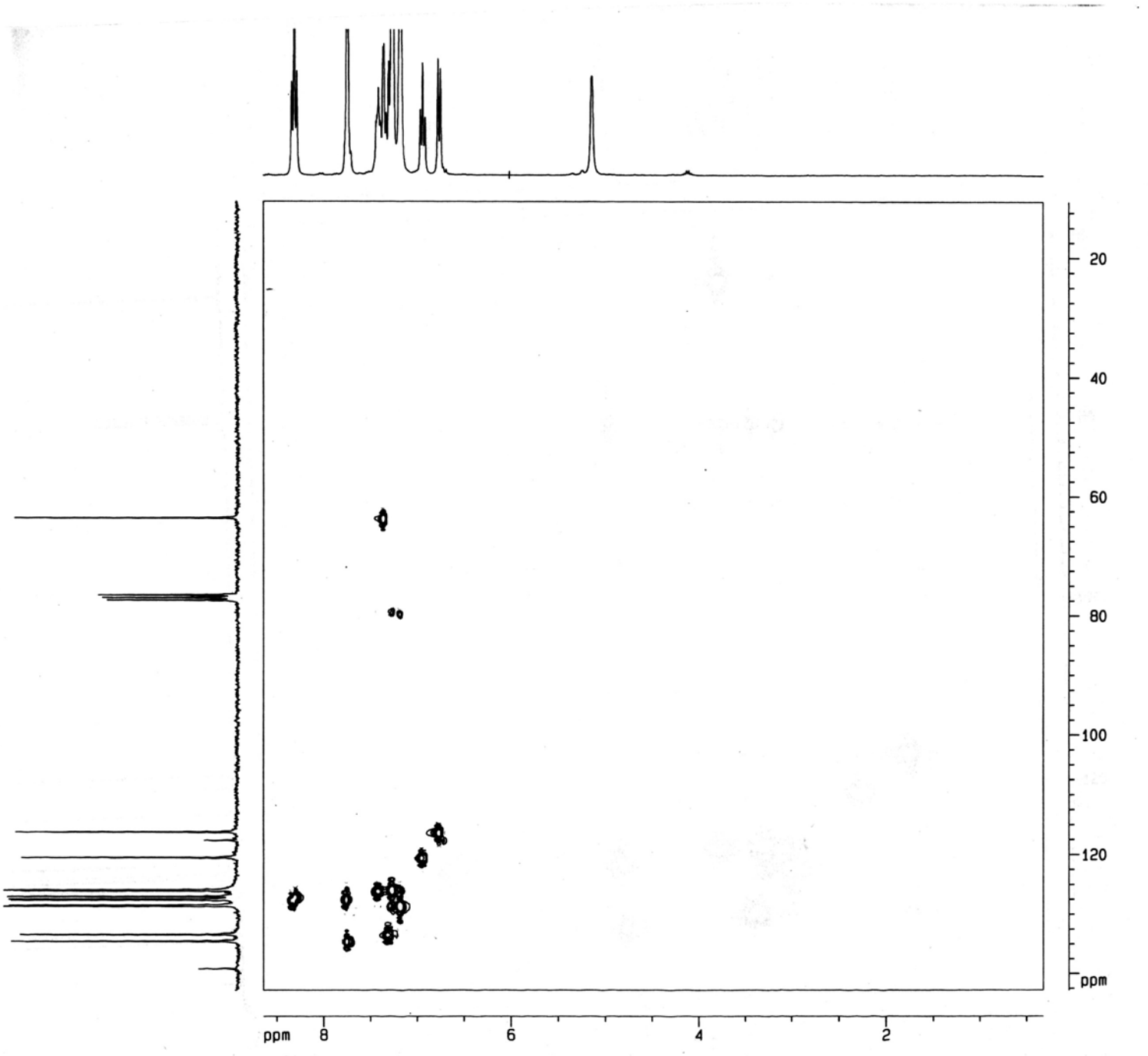


S-32: (a). DEPT-90 and (b). DEPT-135 NMR Spectrum of 6-Phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5d).

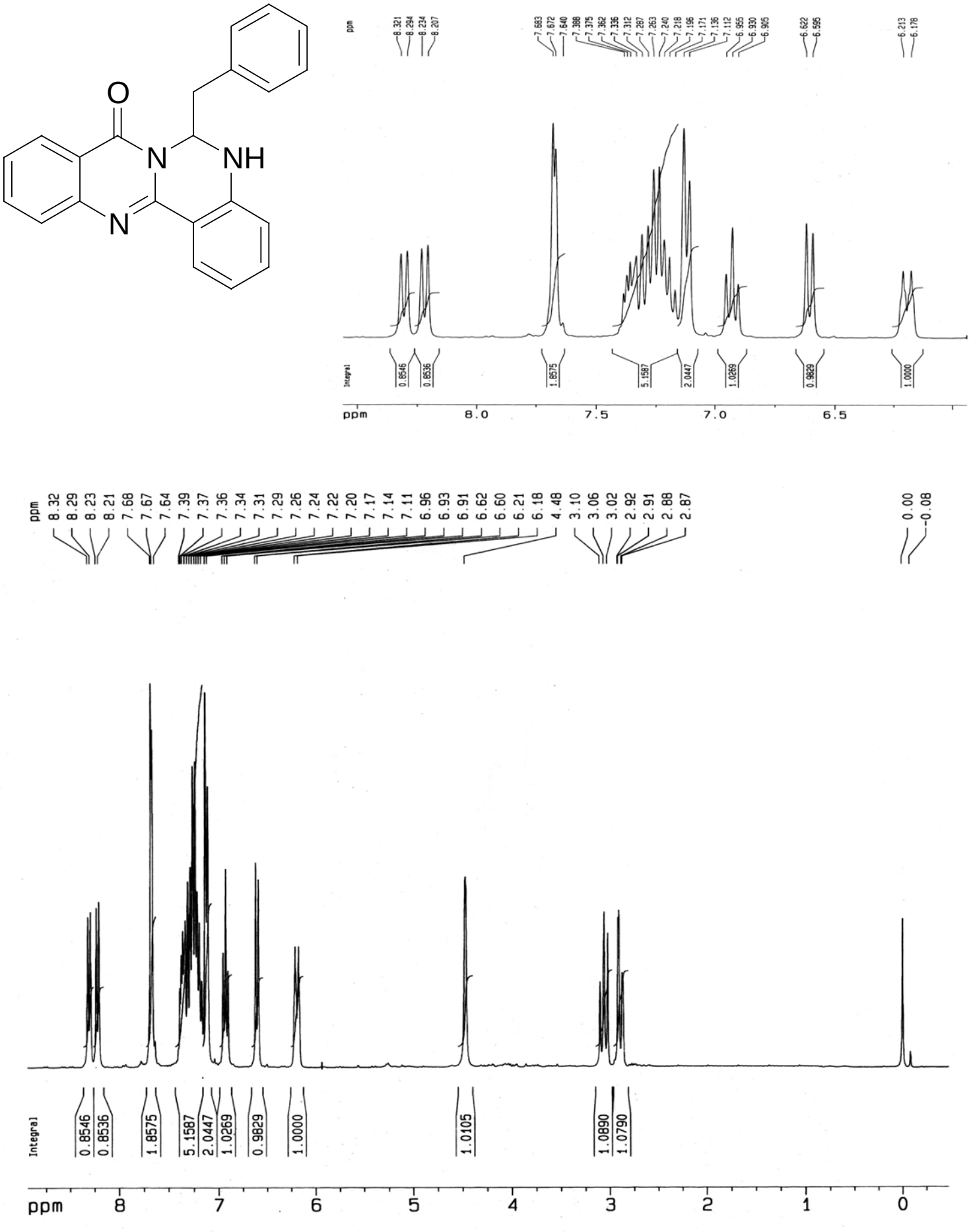


S-33: COSY Spectrum of 6-Phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5d).

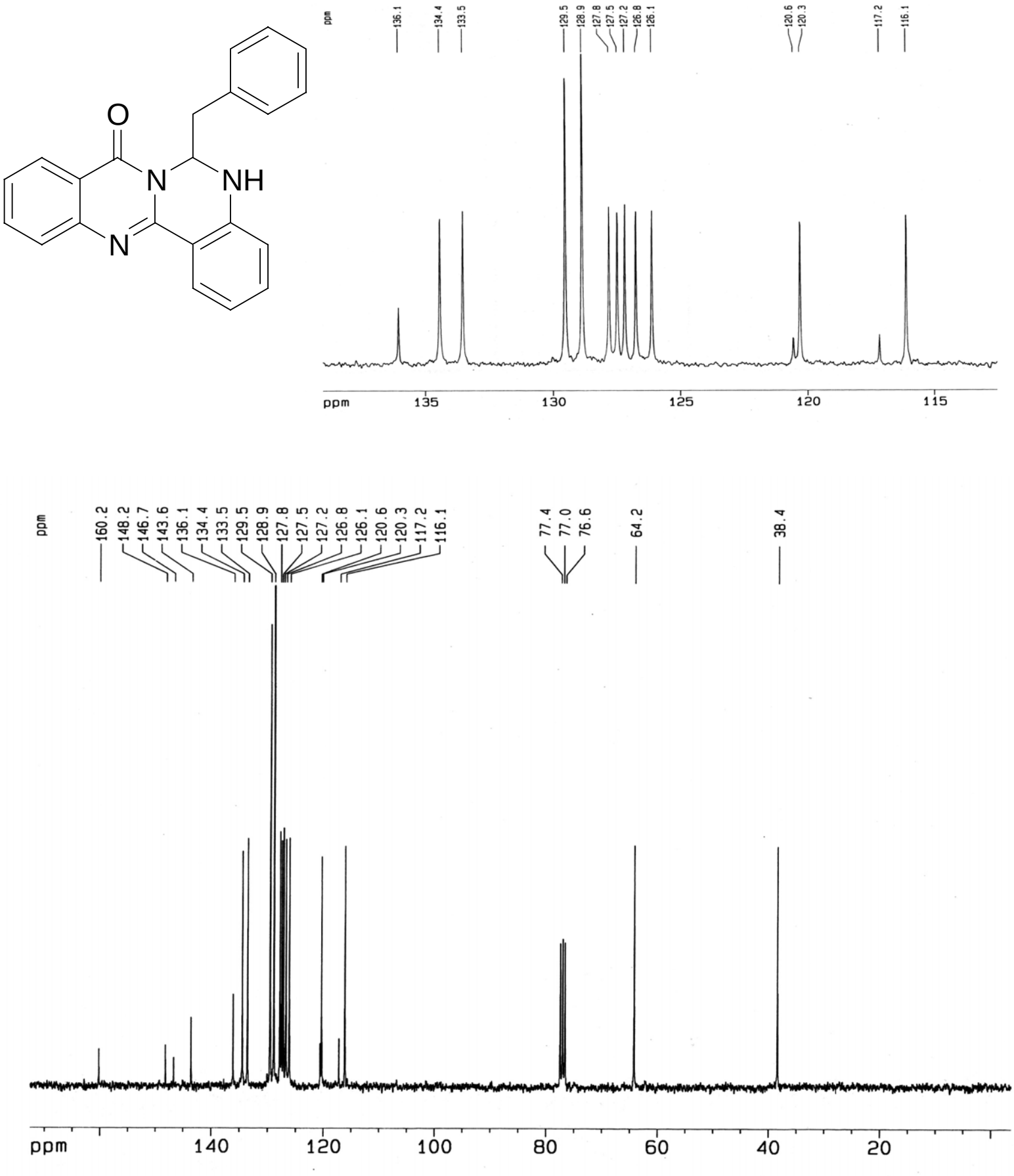




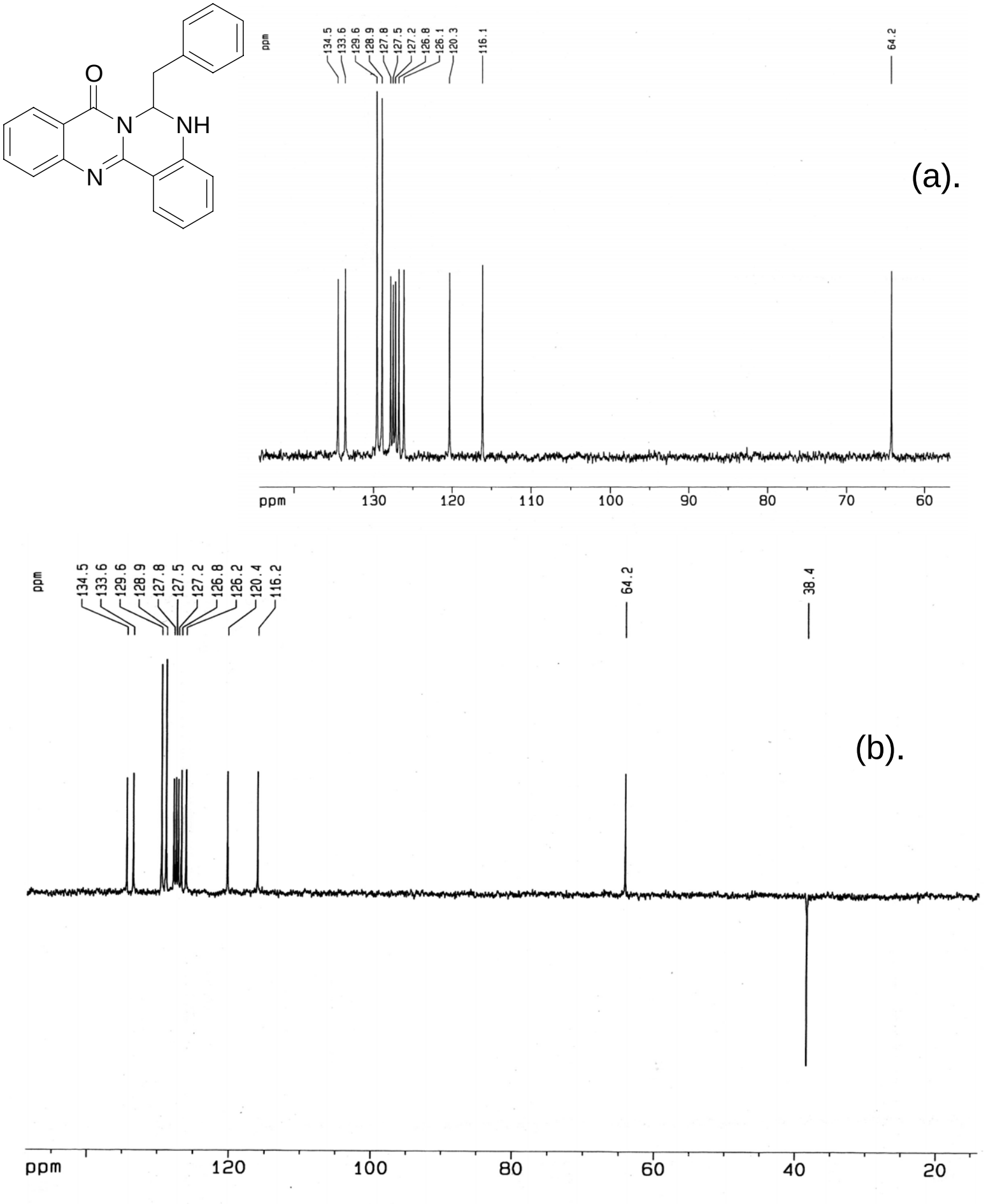
S-34: HSQC Spectrum of 6-Phenyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5d).



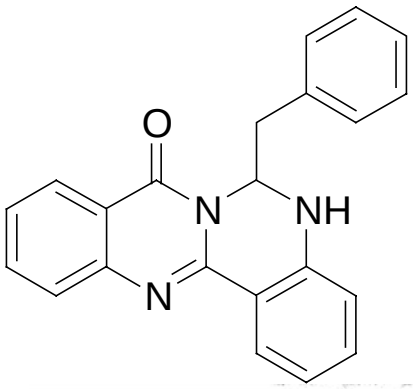
S-35: 1H NMR Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).

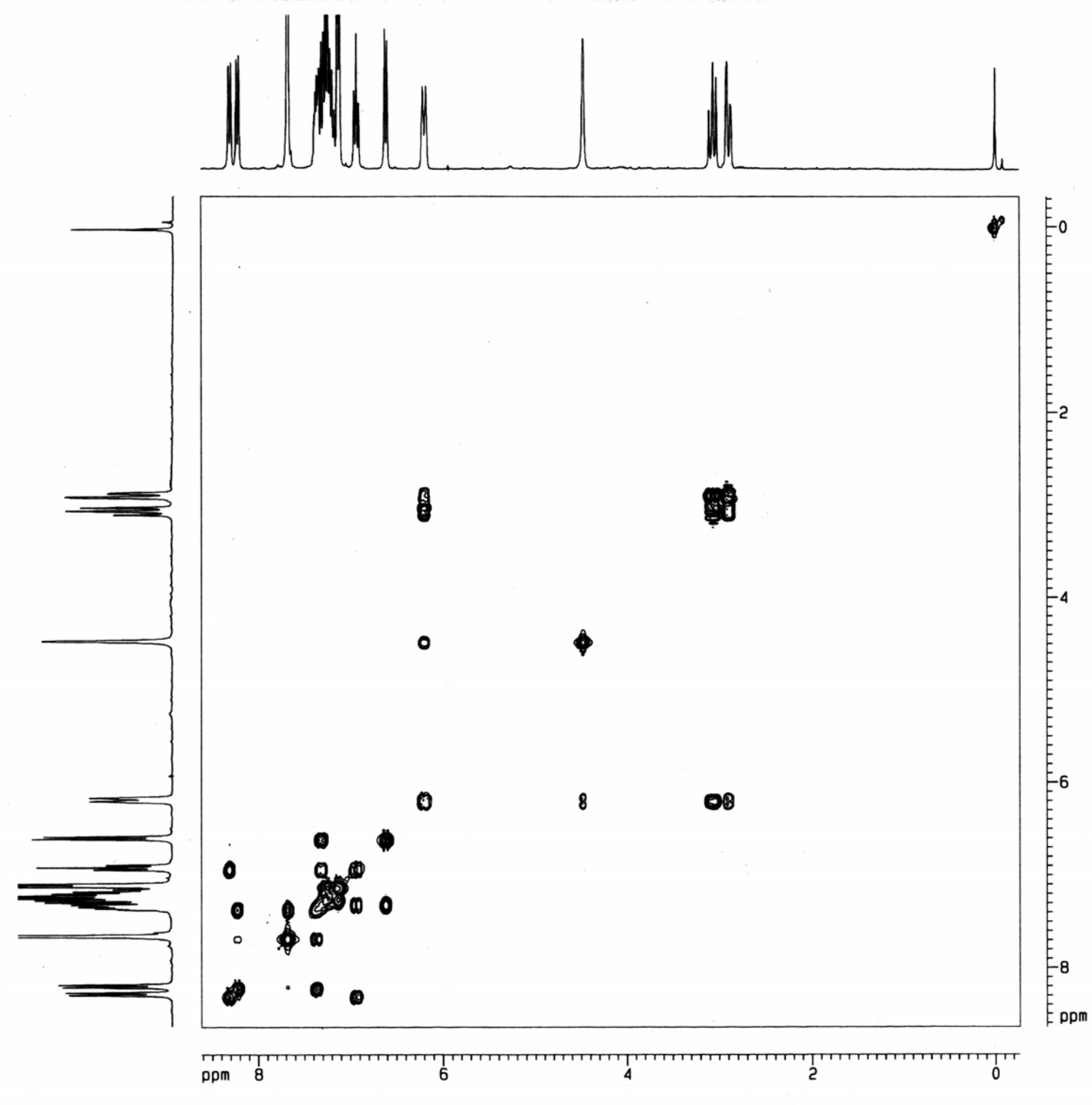


S-36: 13C NMR Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).

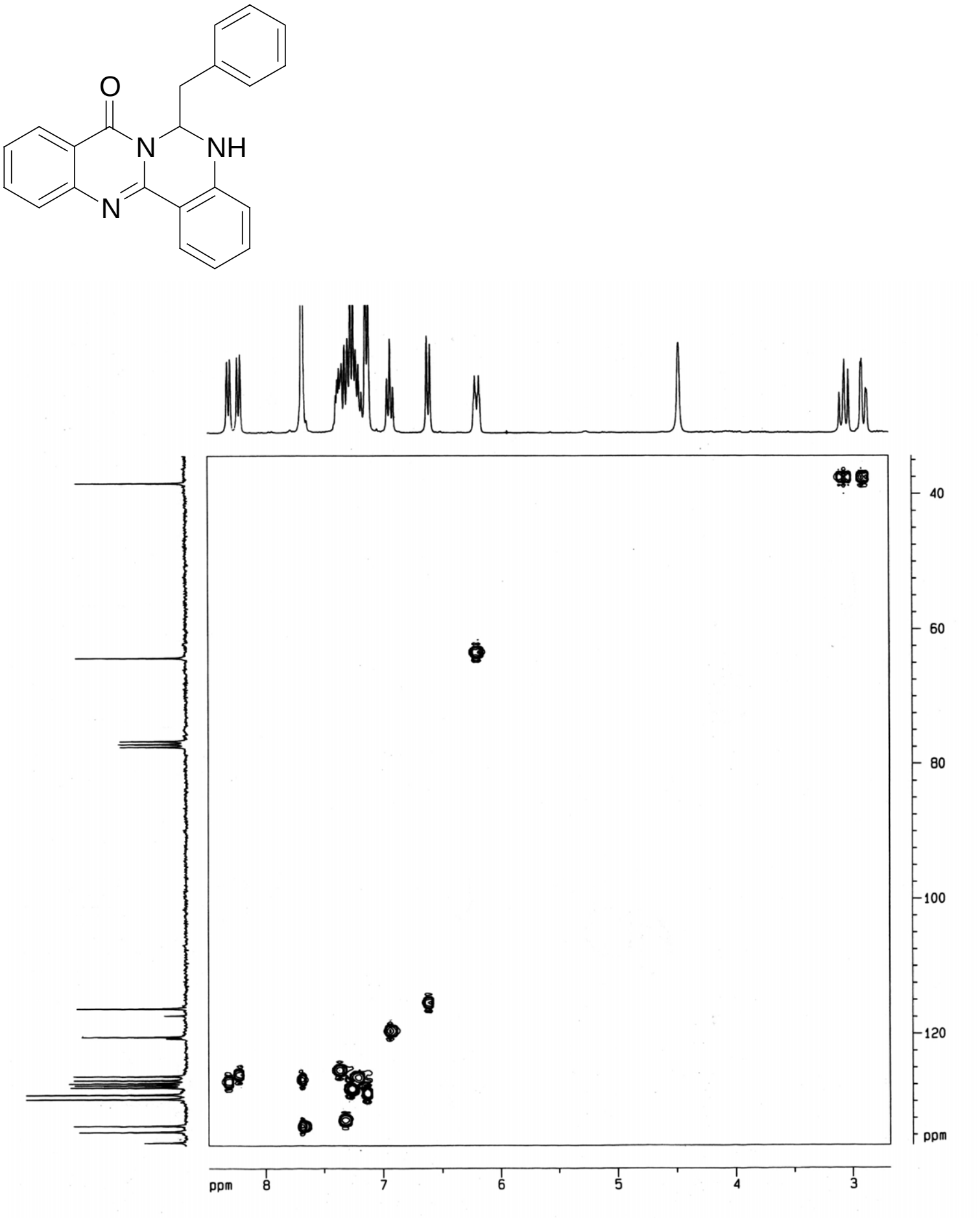


S-37: (a). DEPT-90 and (b). DEPT-135 NMR Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).

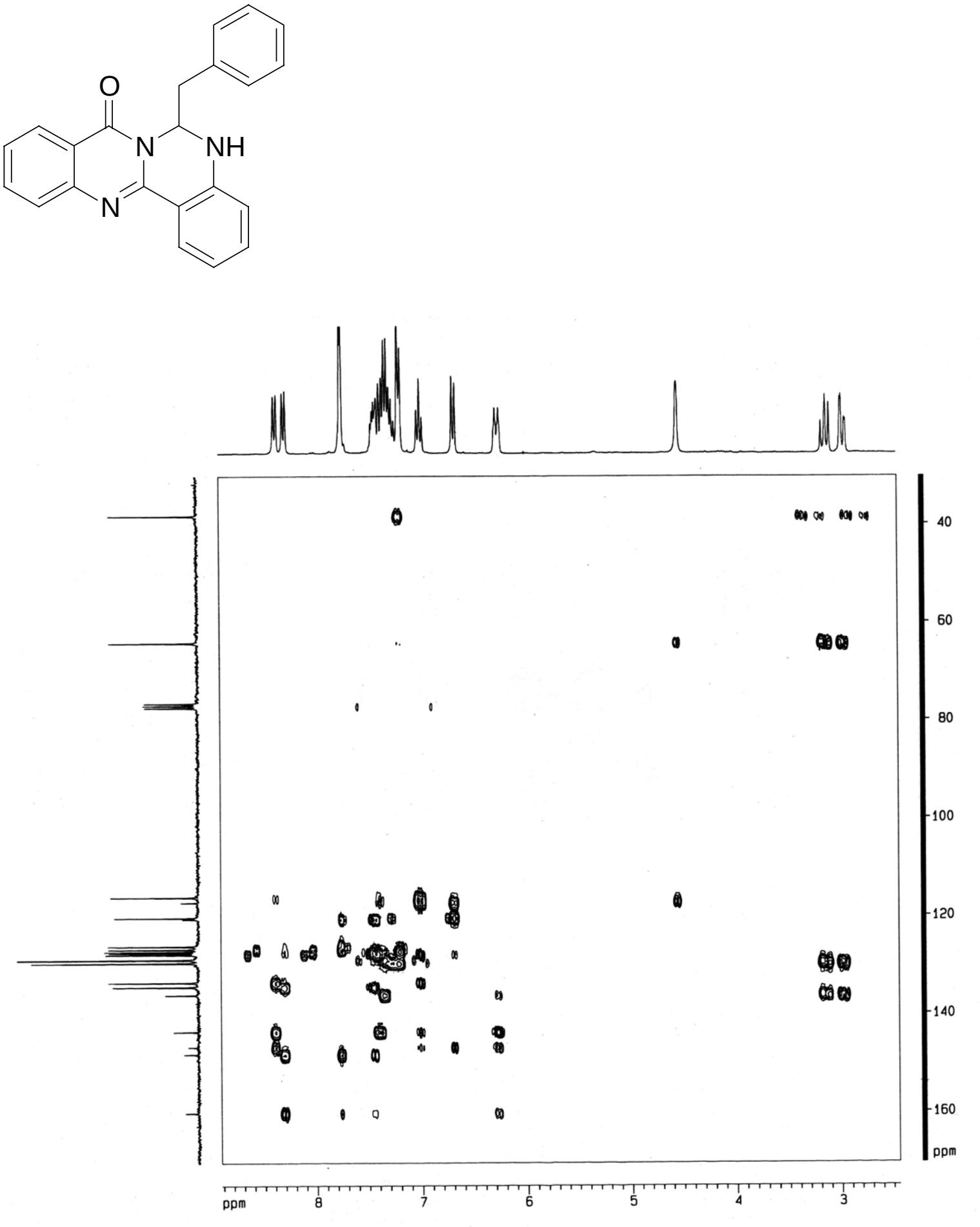




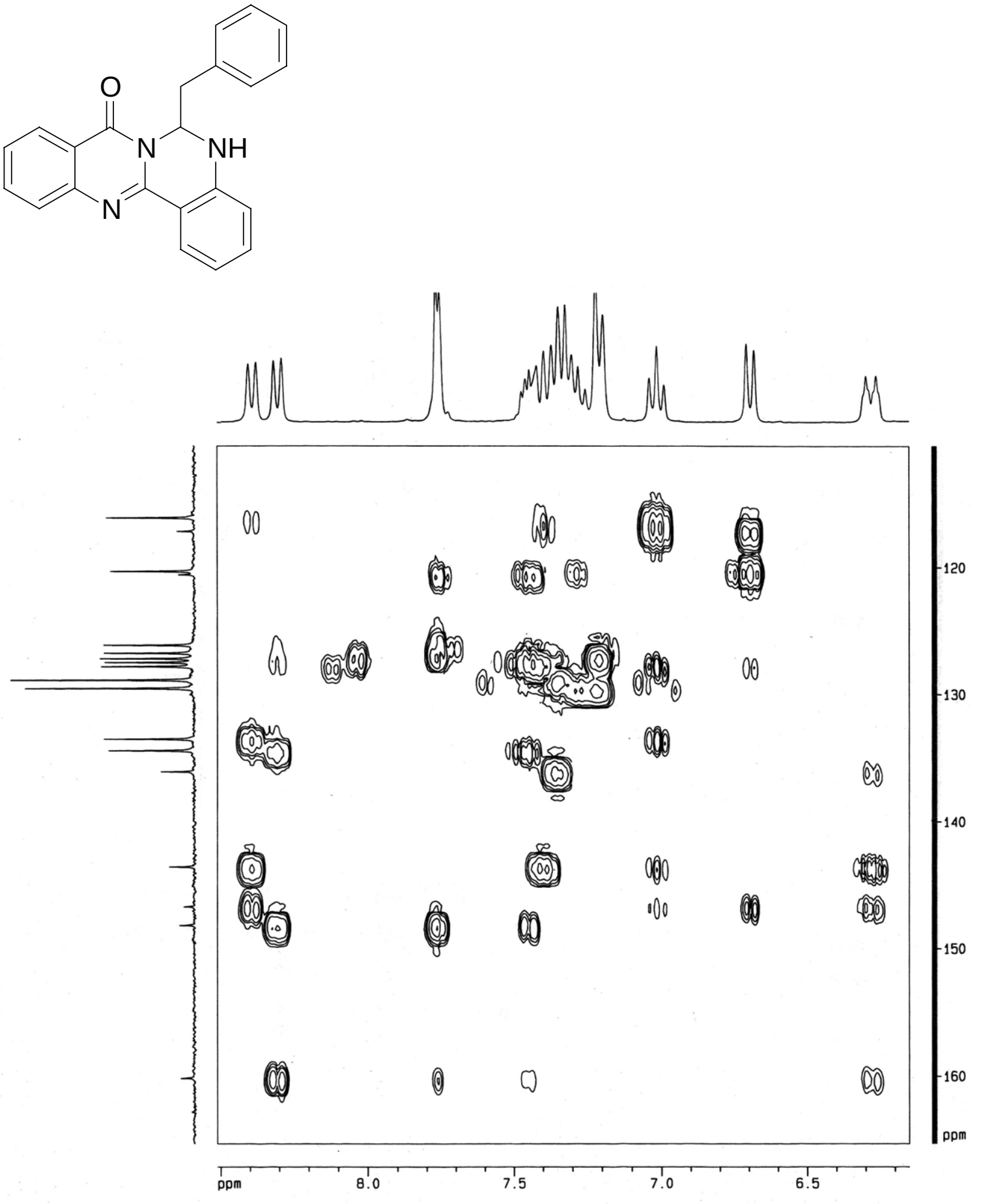
S-38: COSY Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).



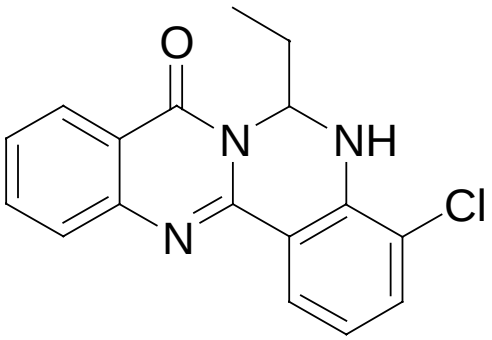
S-39: HSQC Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).

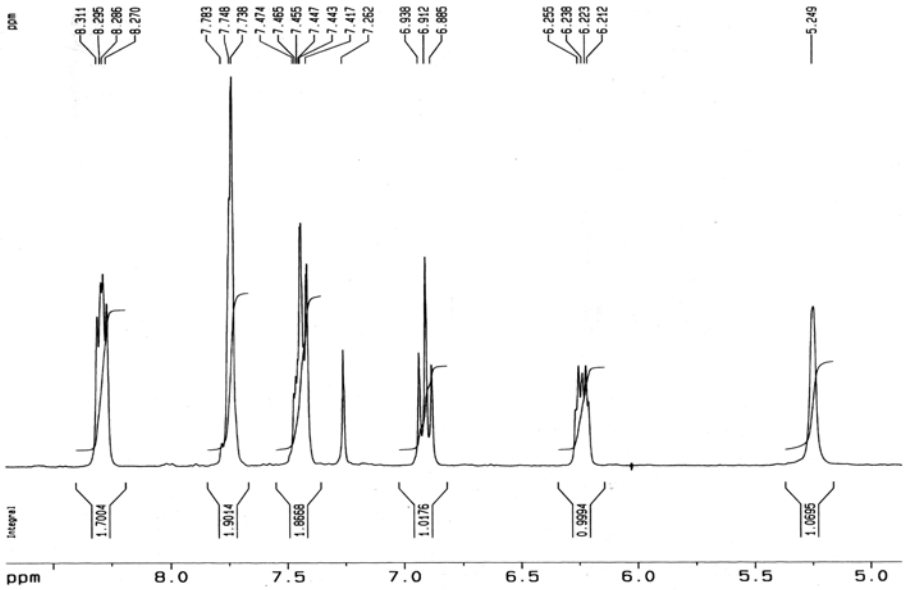


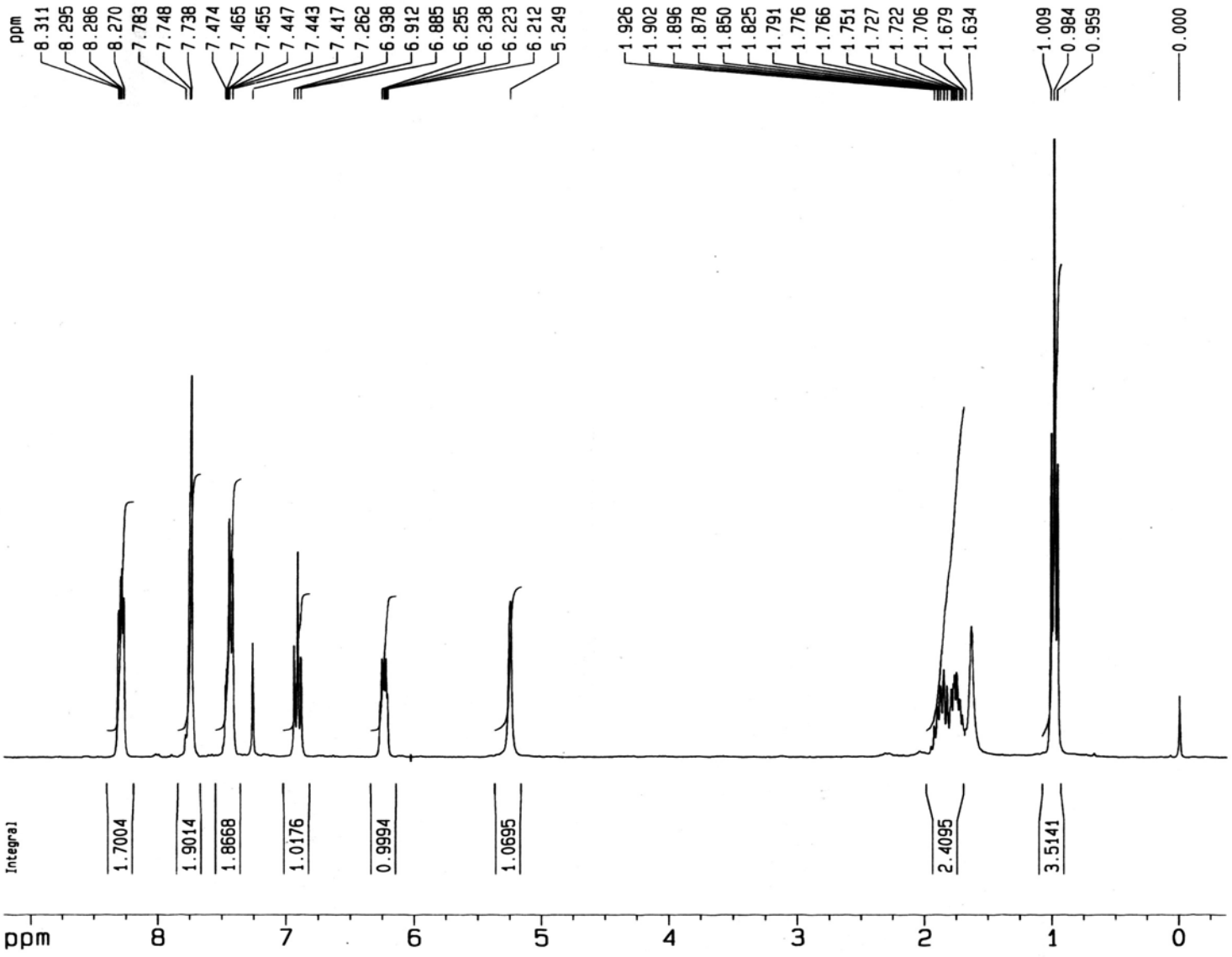
S-40: HMBC Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).



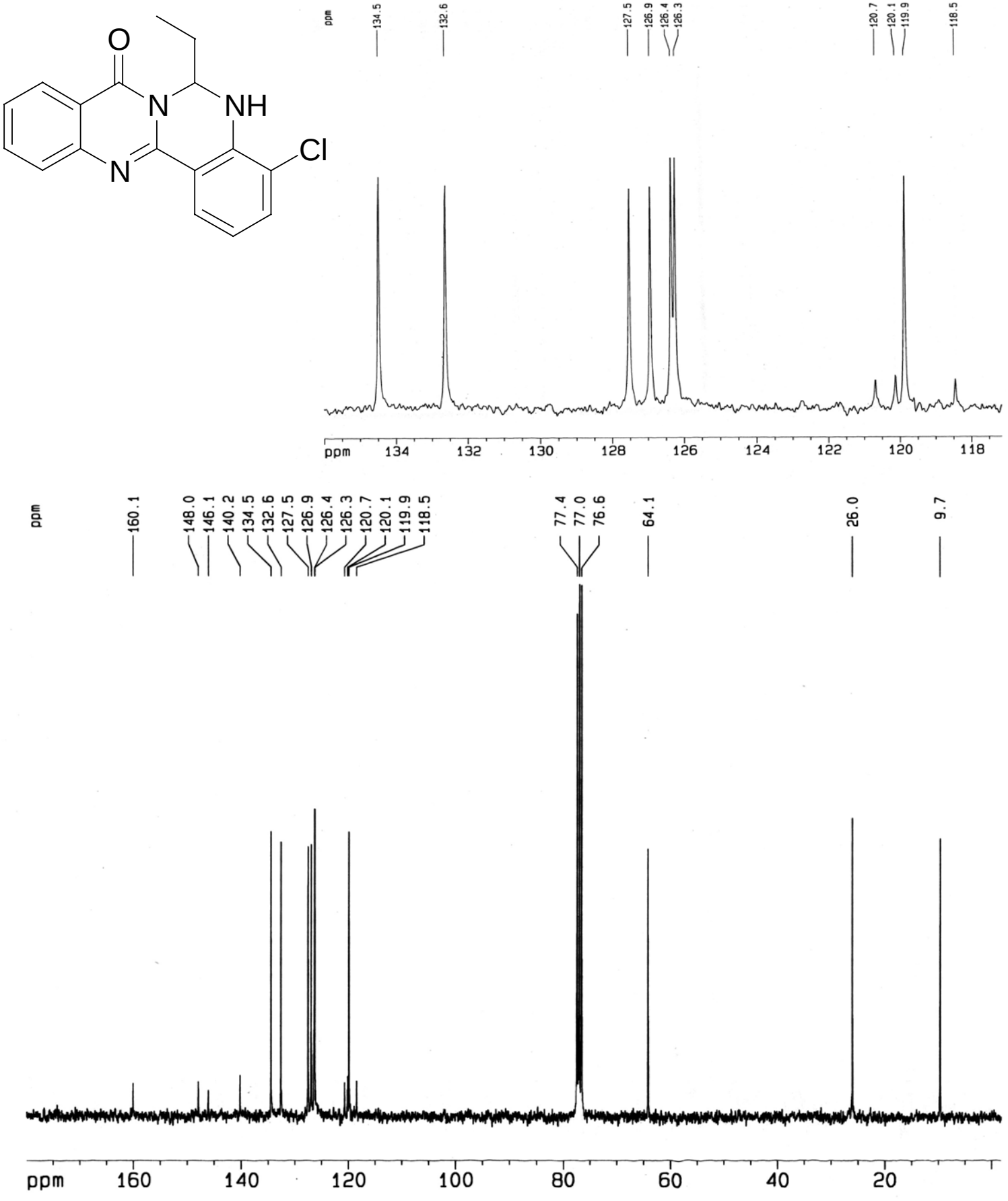
S-41: Expanded Region of the HMBC Spectrum of 6-Benzyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (5e).



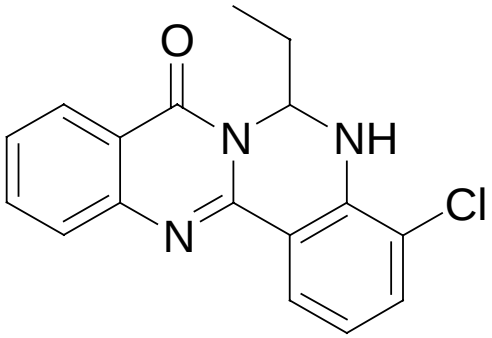


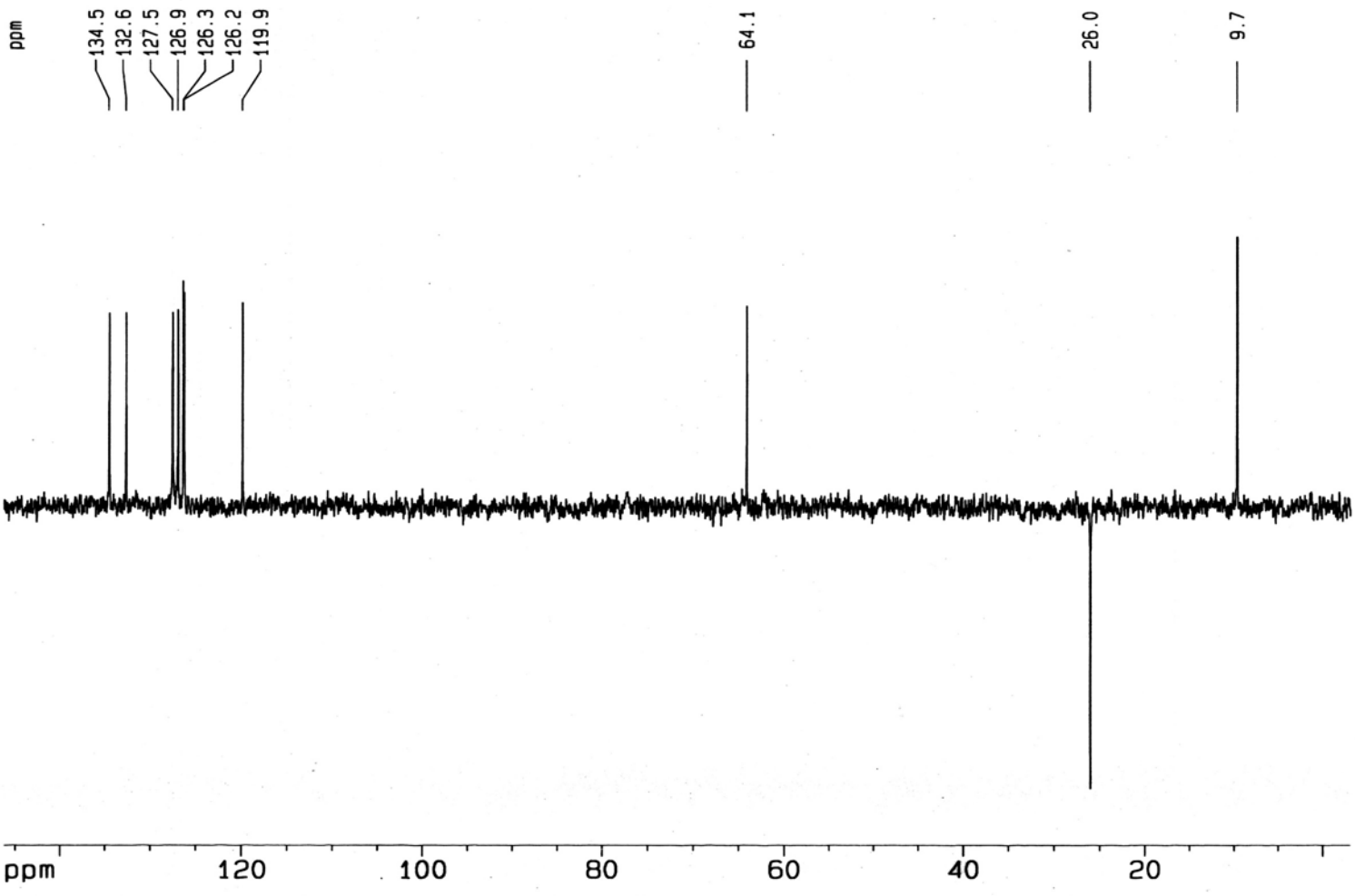


S-42: 1H NMR Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5g).

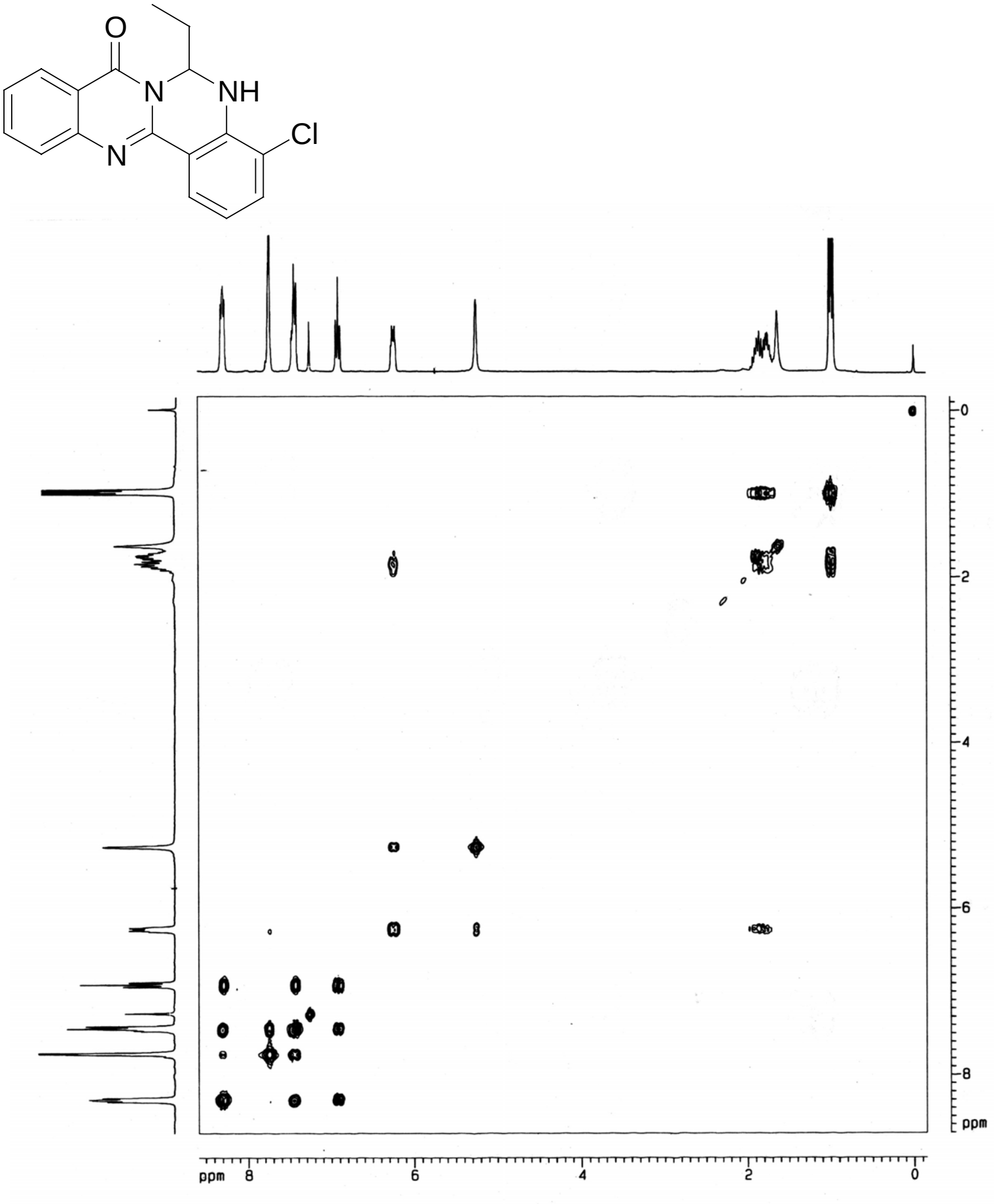


S-43: 13C NMR Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5g).

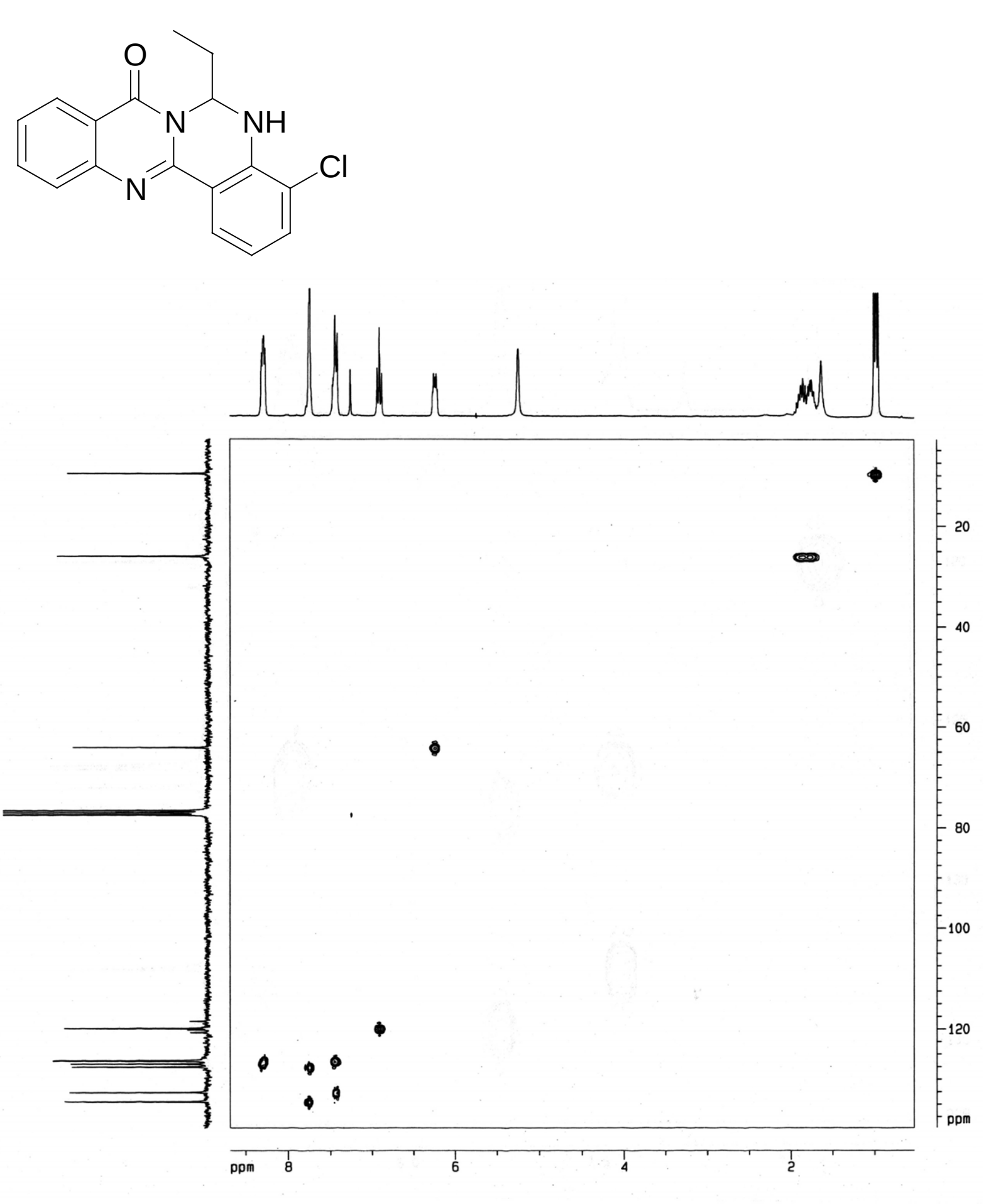




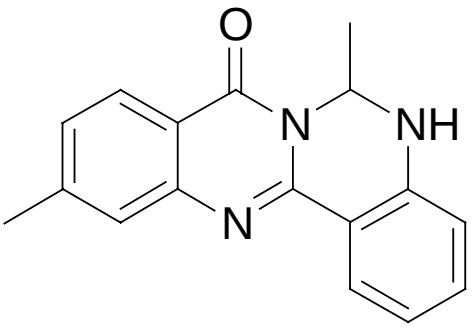
S-44: DEPT-135 NMR Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5g).

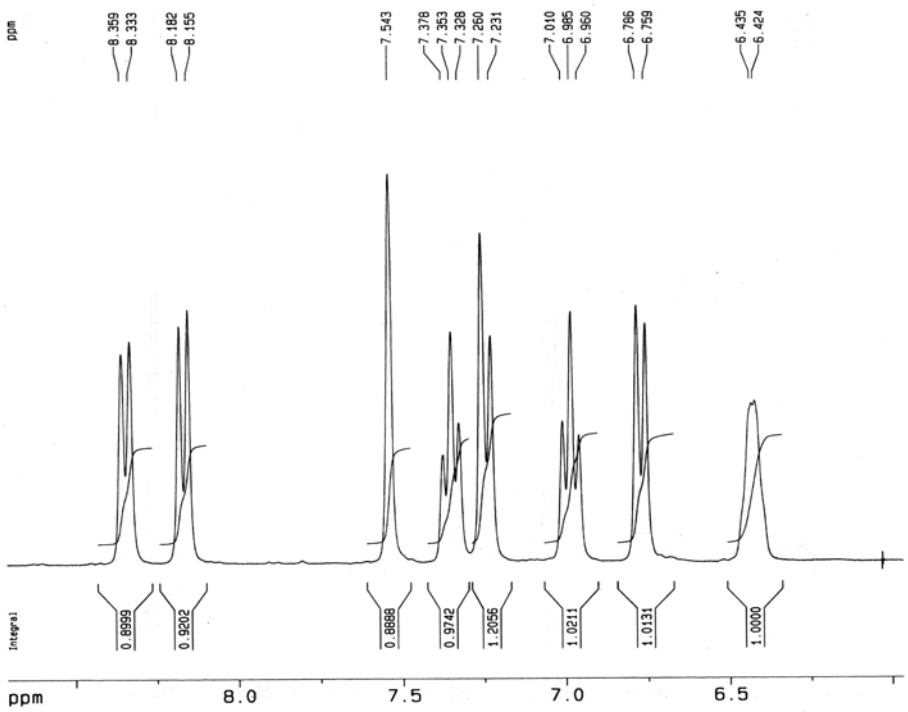


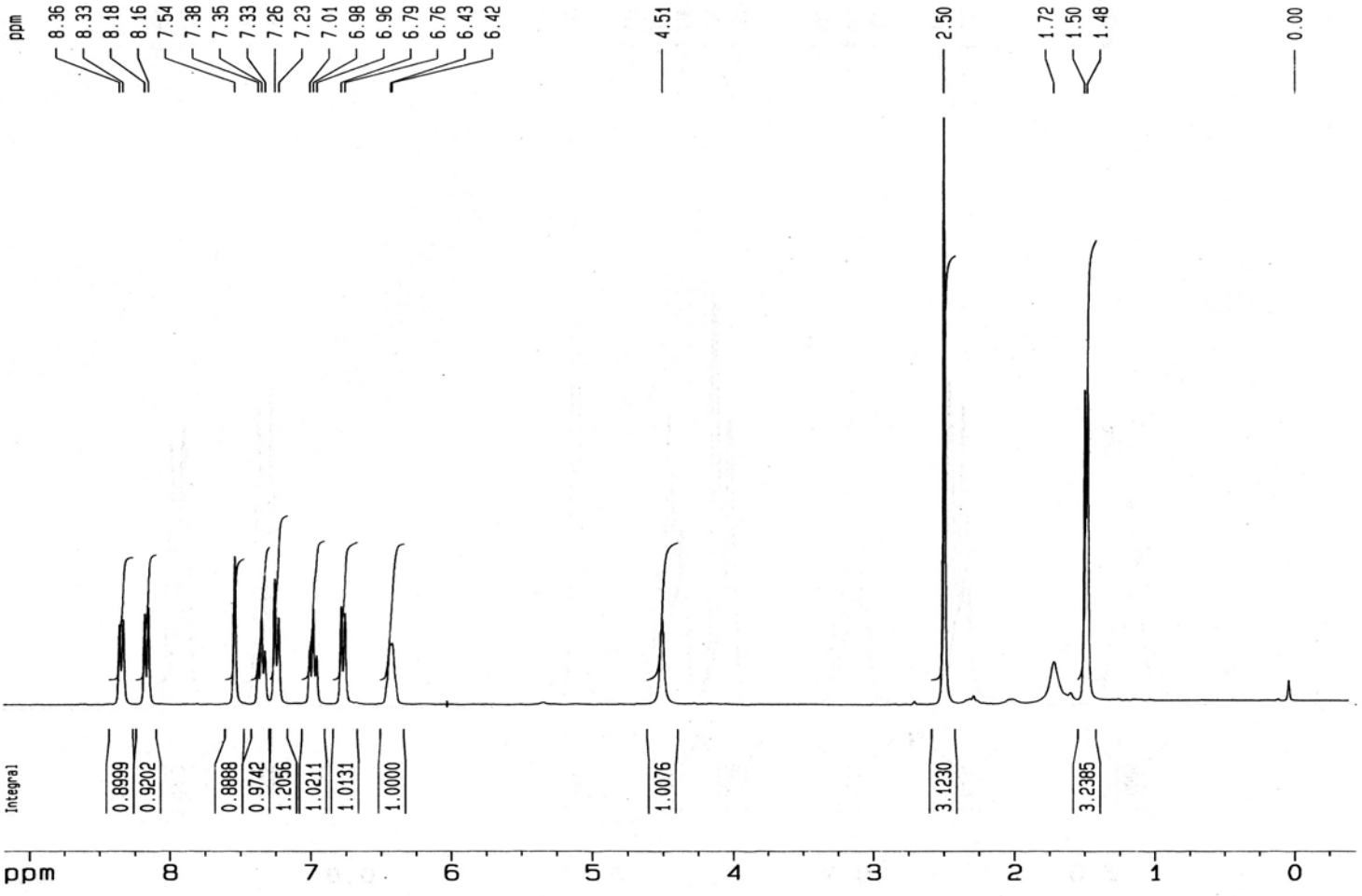
S-45: COSY Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5g).



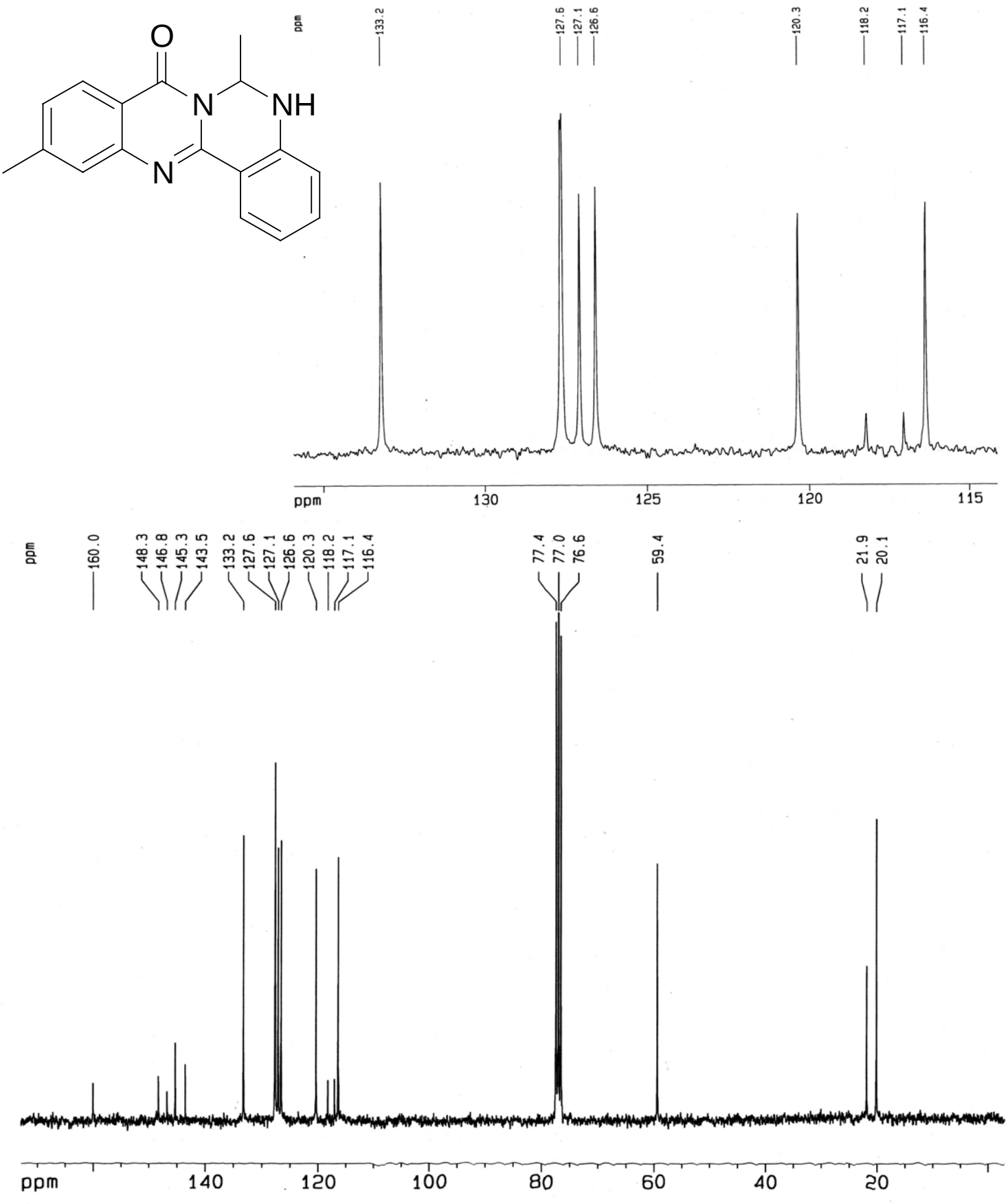
S-46: HSQC Spectrum of 4-Chloro-6-ethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5g).



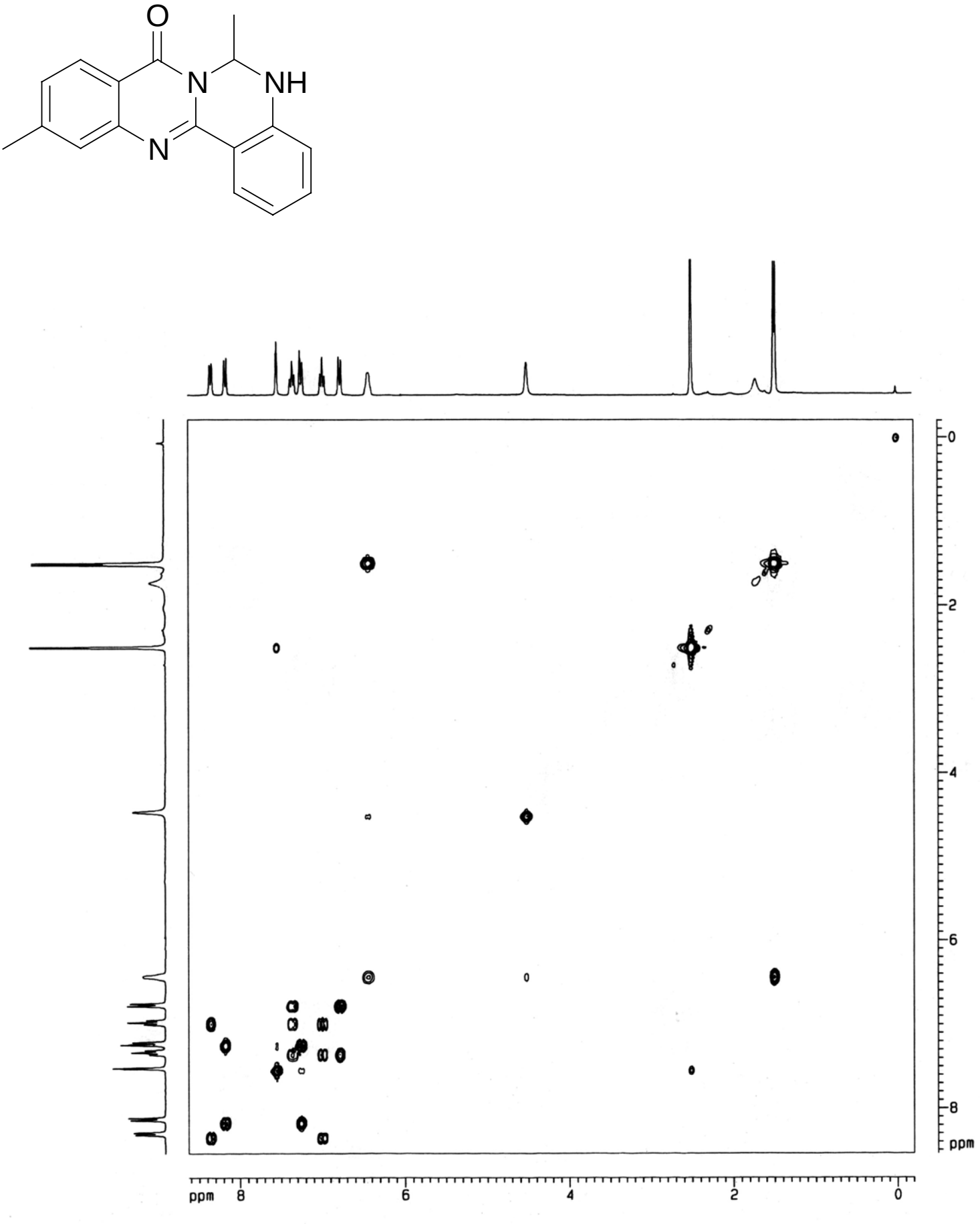




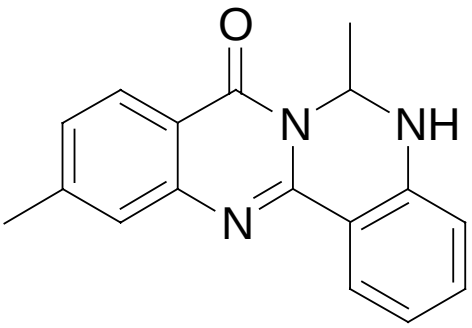
S-47: 1H NMR Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5h).

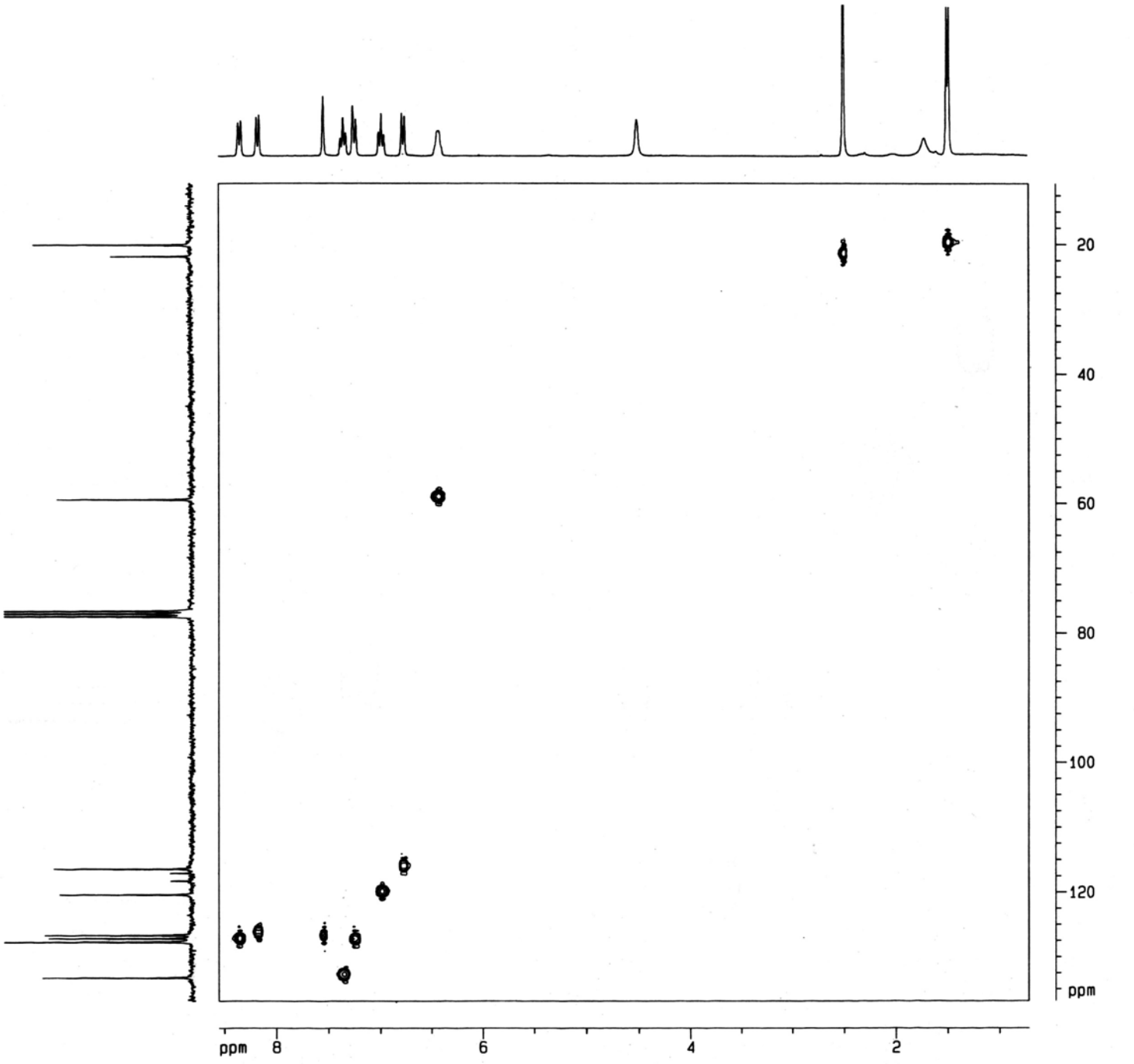


S-48: 13C NMR Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5h).

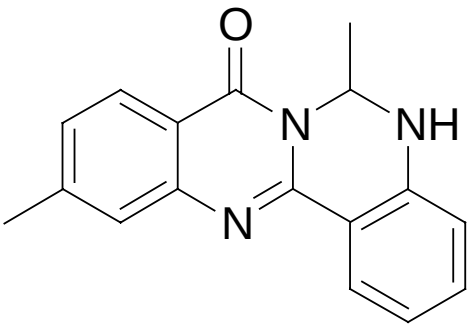


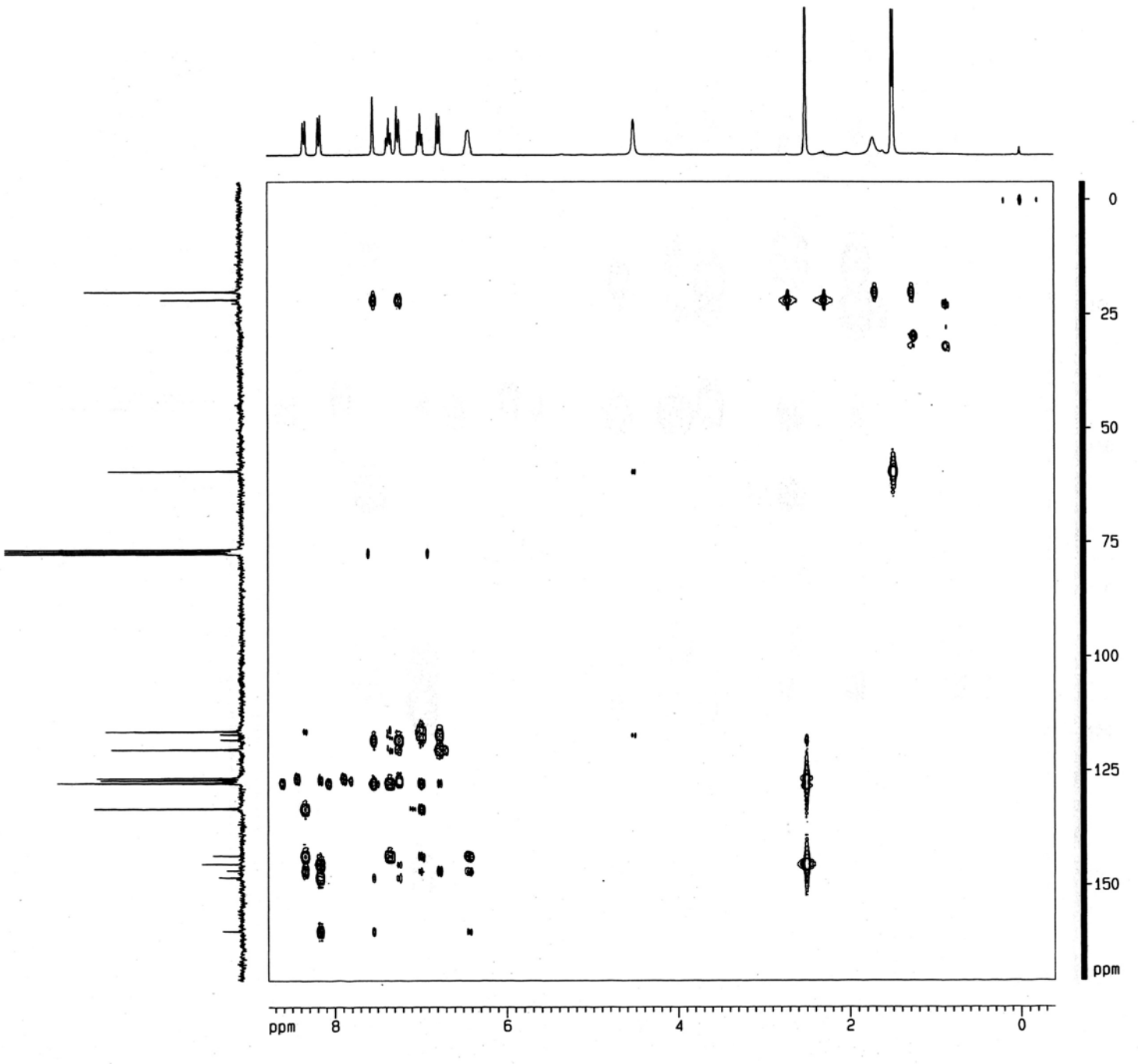
S-49: COSY Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5h).



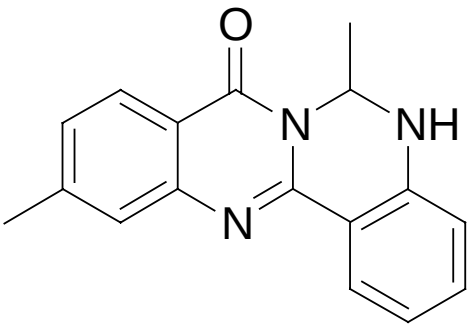


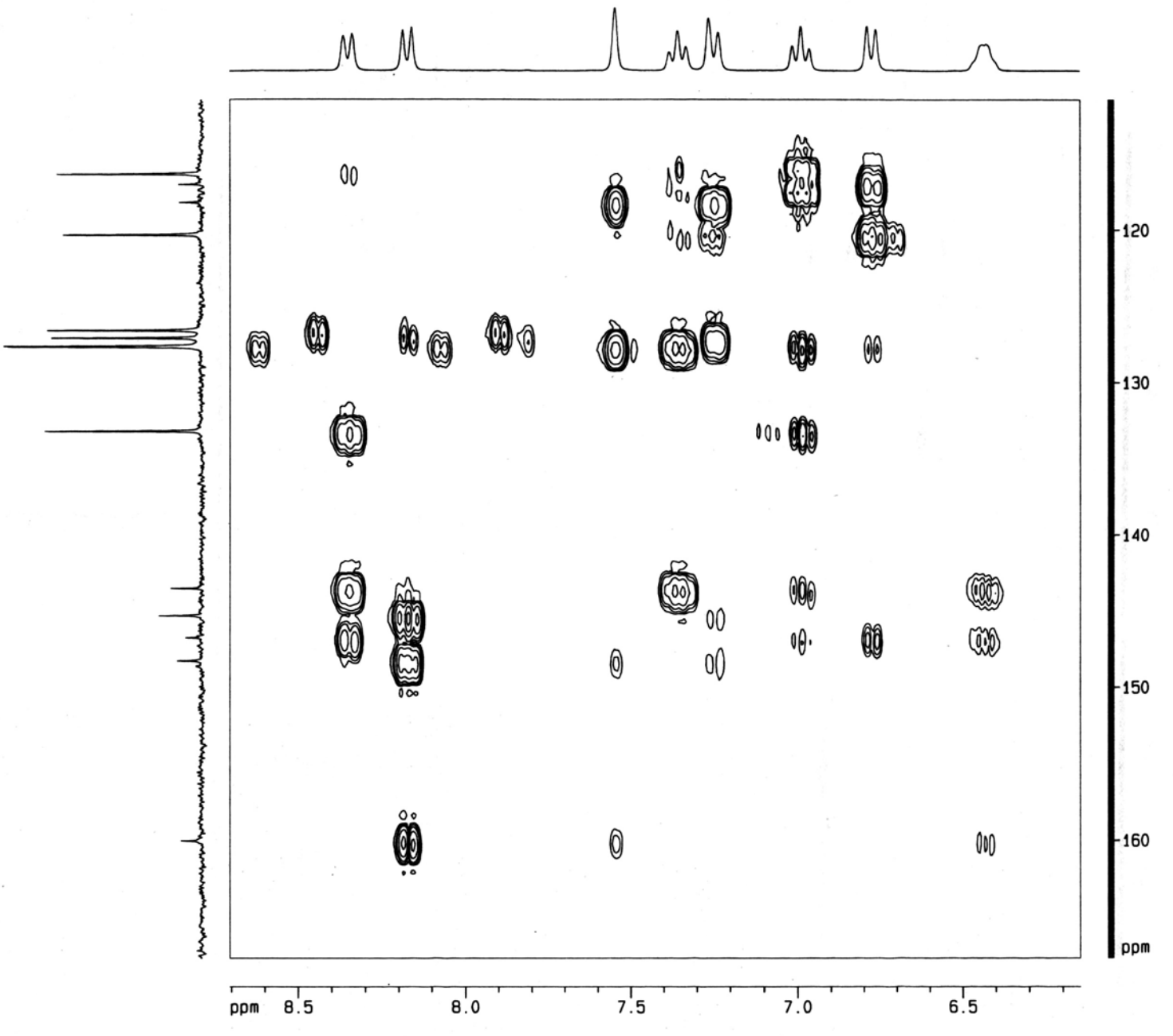
S-50: HSQC Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5h).



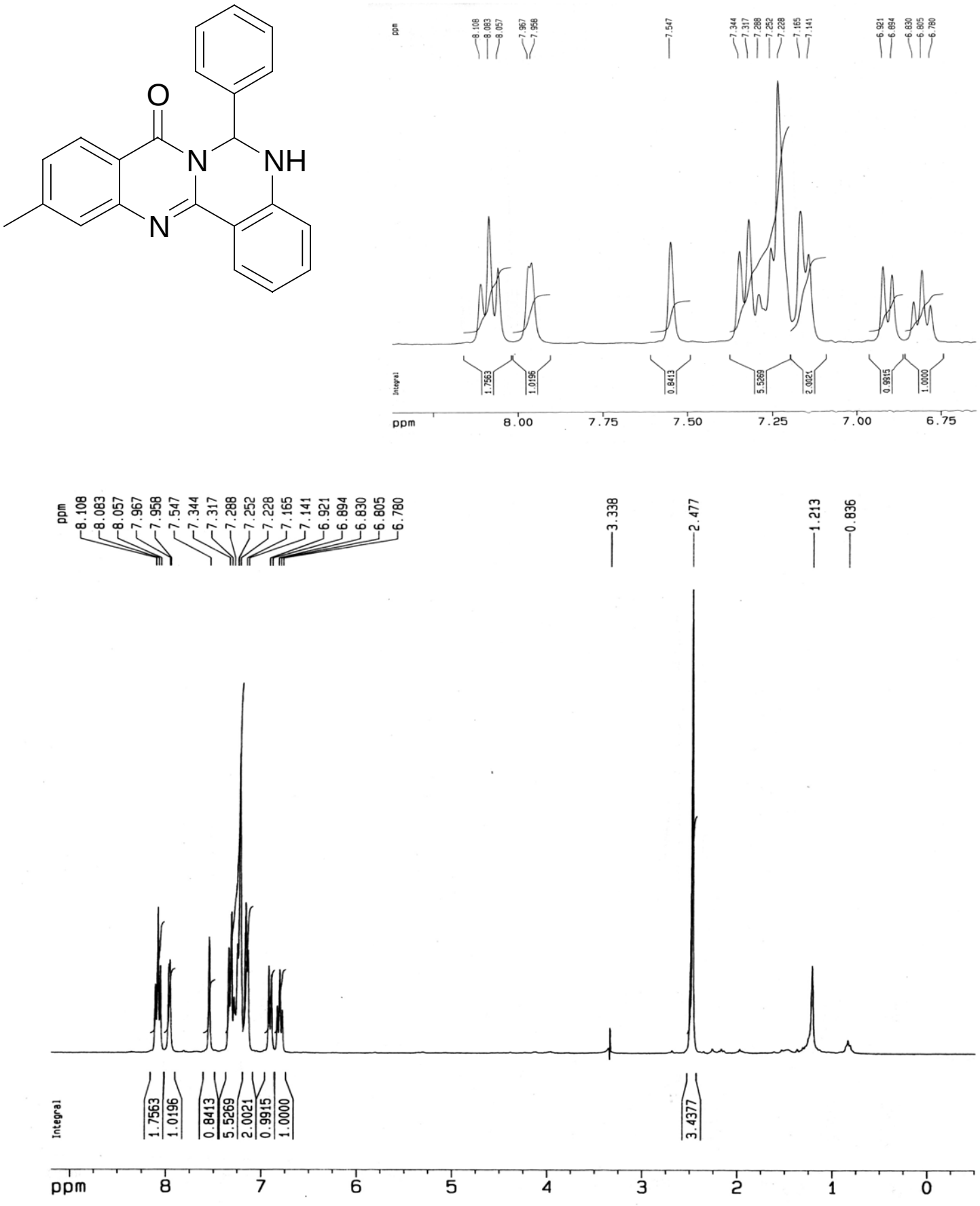


S-51: HMBC Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5h).

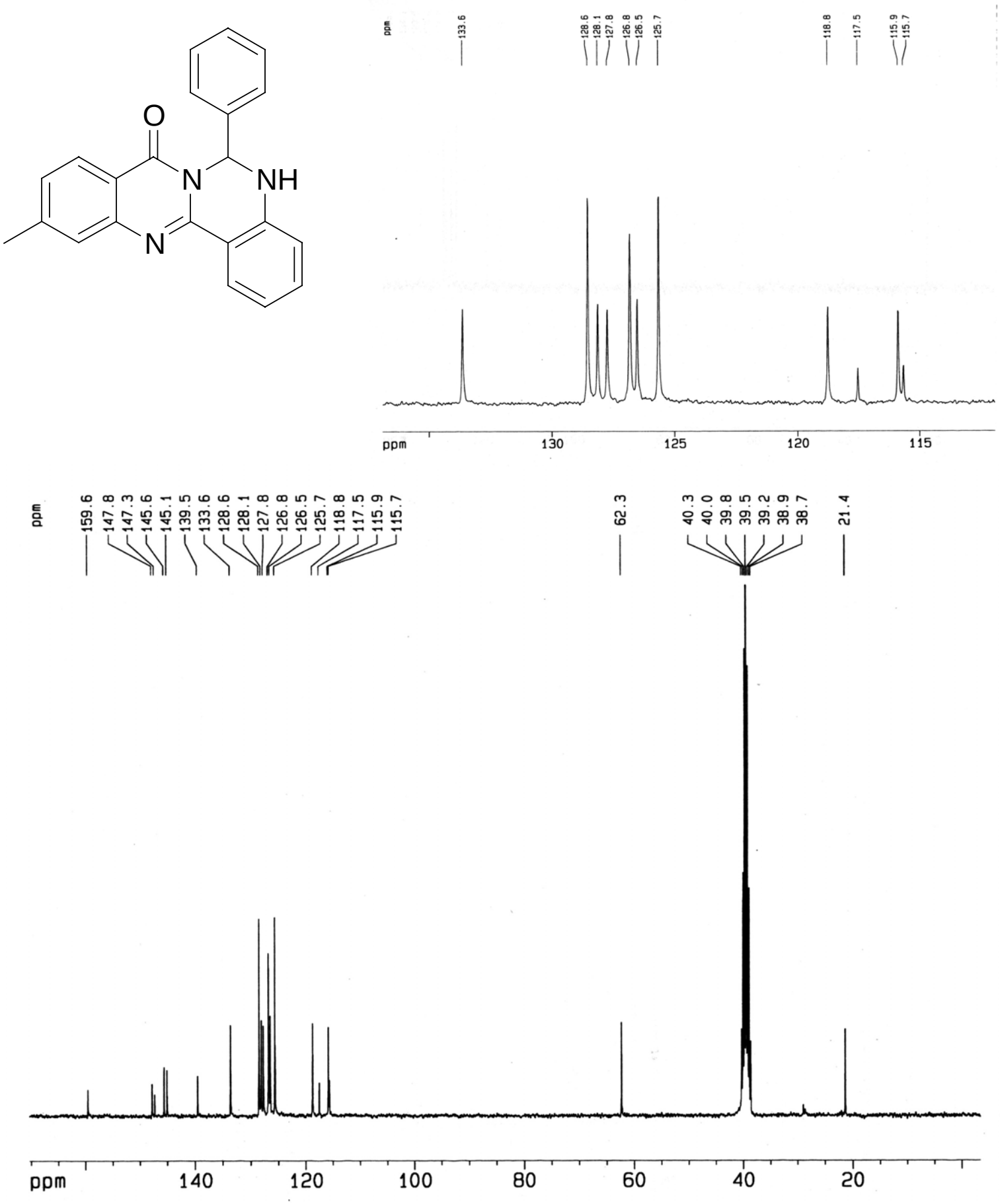




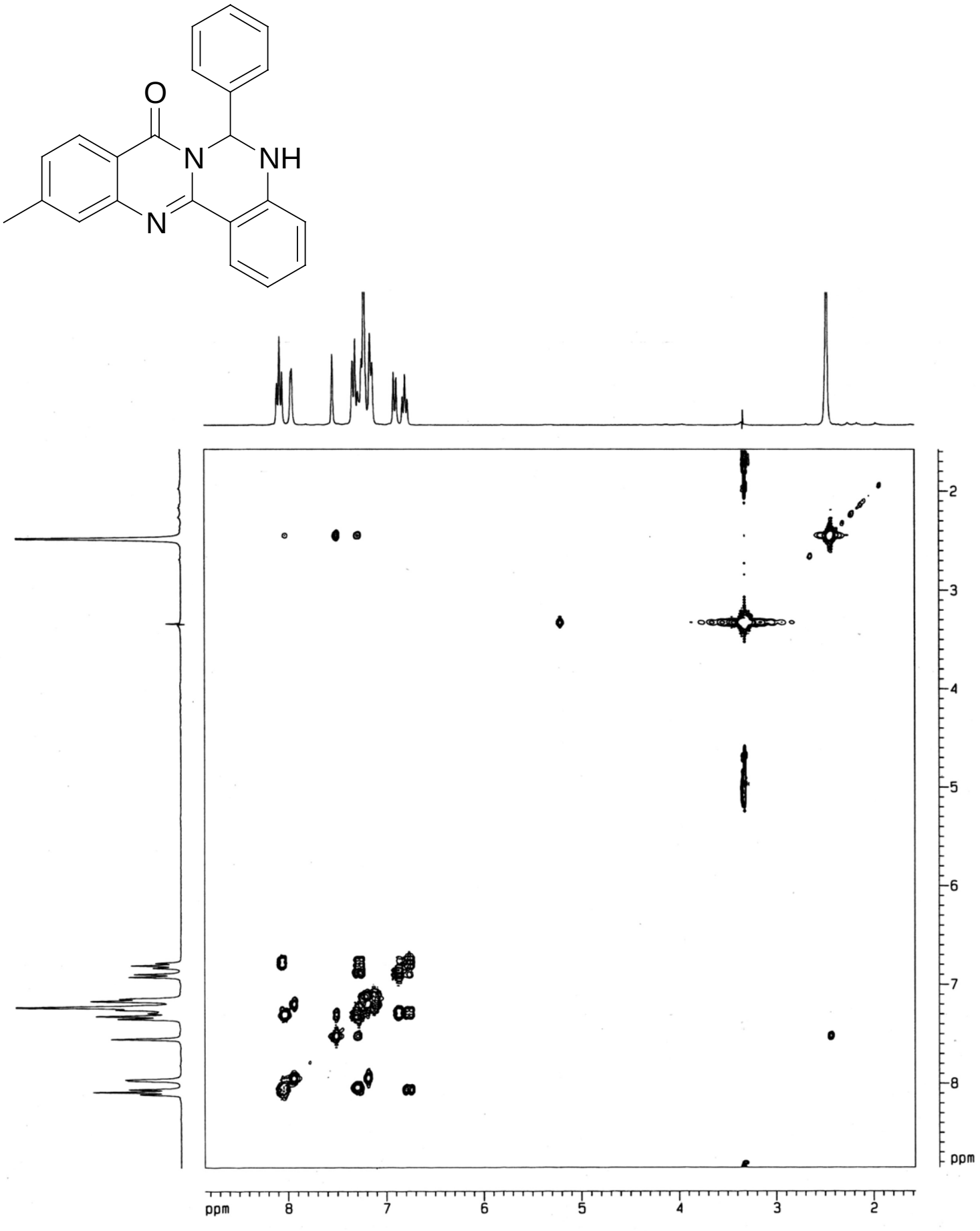
S-52: Extended Region of the HMBC Spectrum of 6,11-Dimethyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5h).



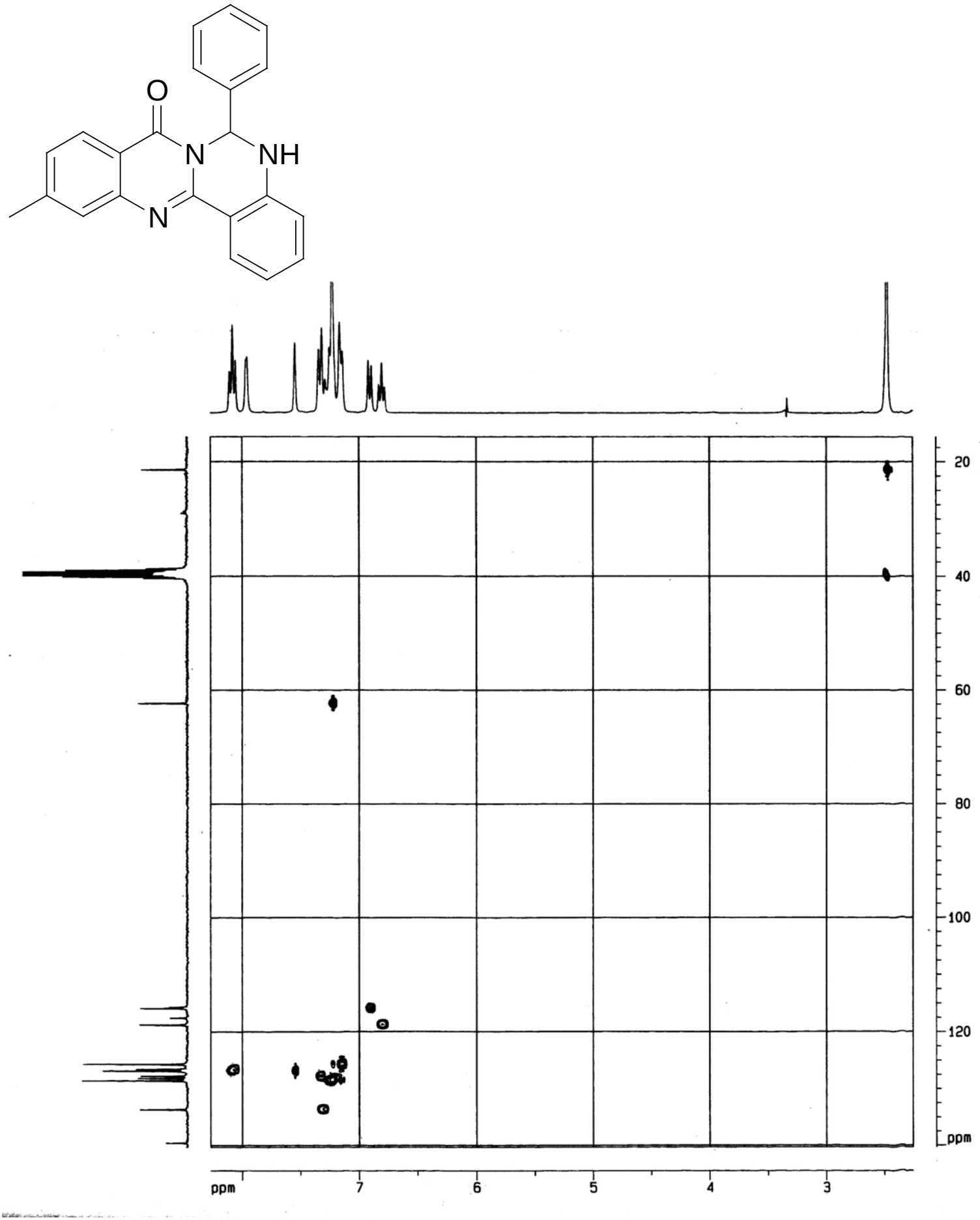
S-53: 1H NMR Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5i).



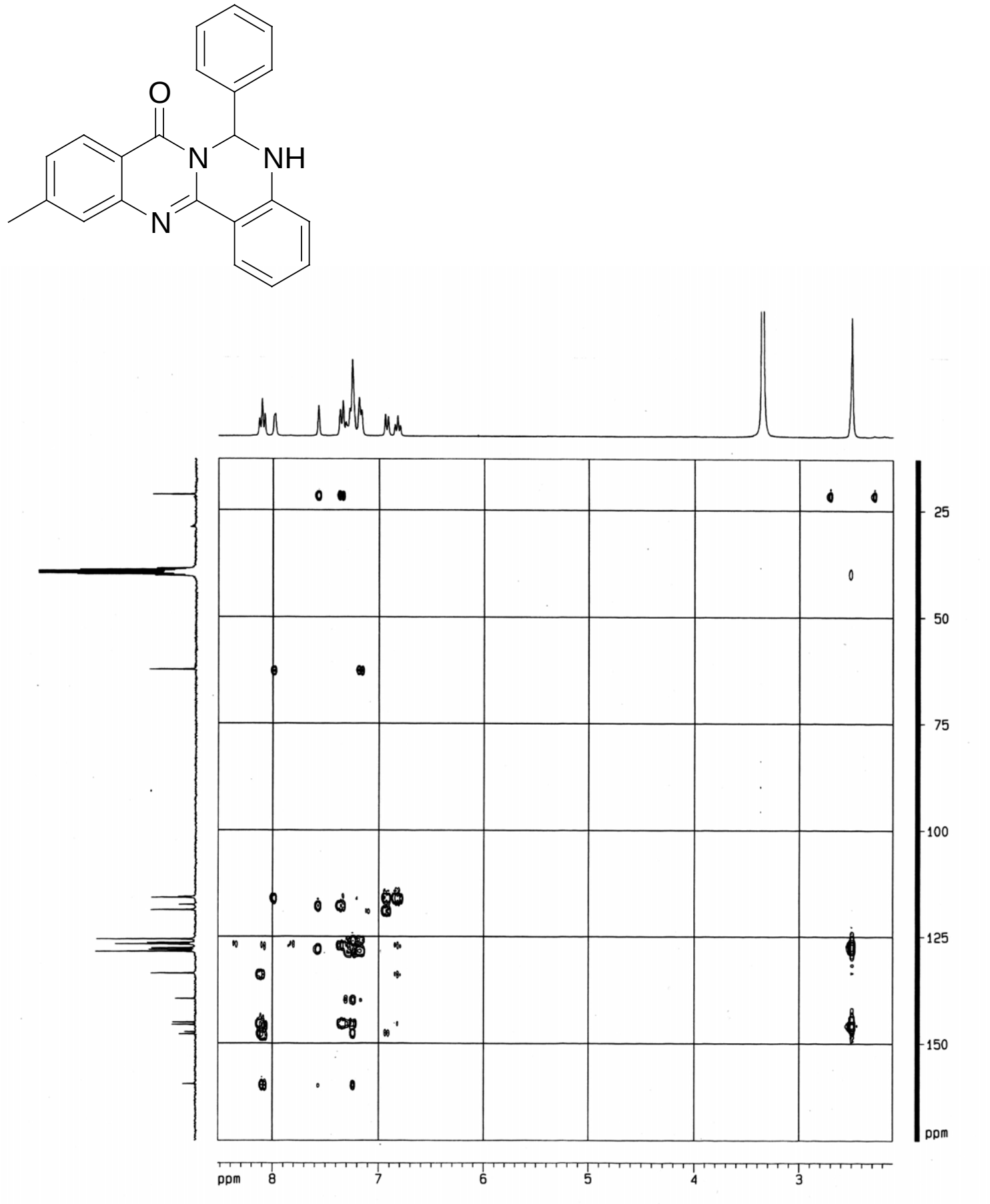
S-54: 13C NMR Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5i).



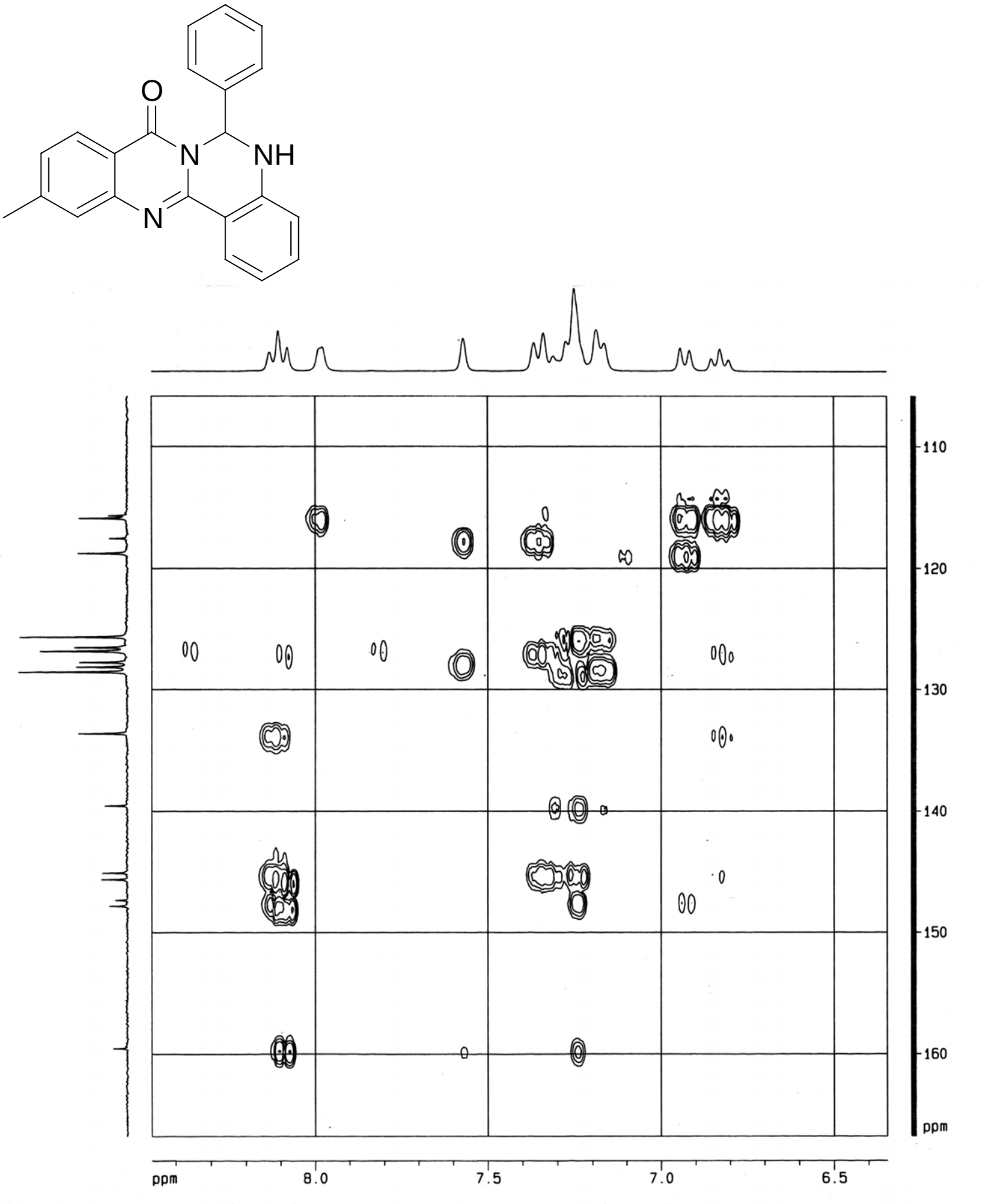
S-55: COSY Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5i).



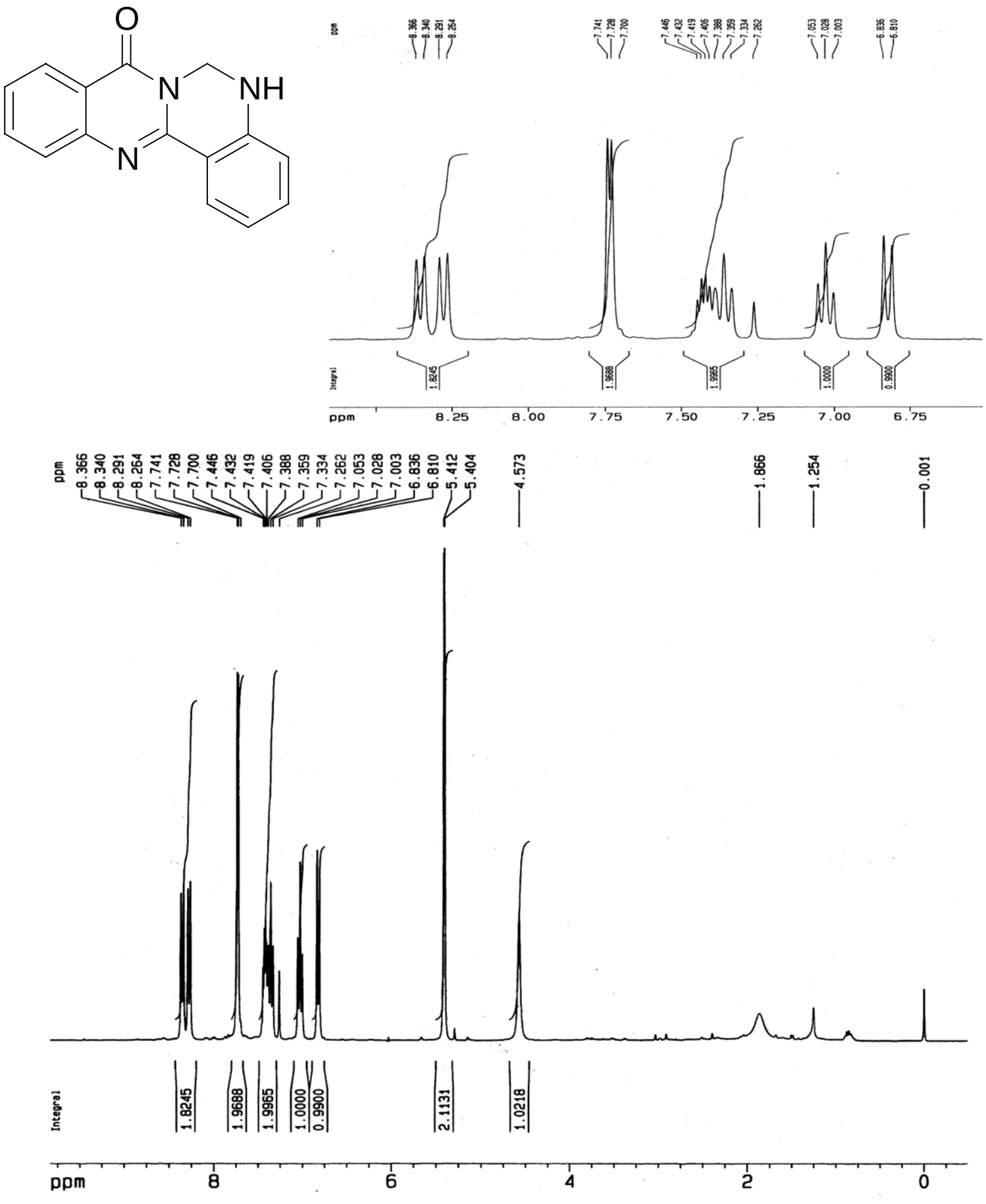
S-56: HSQC Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5i).



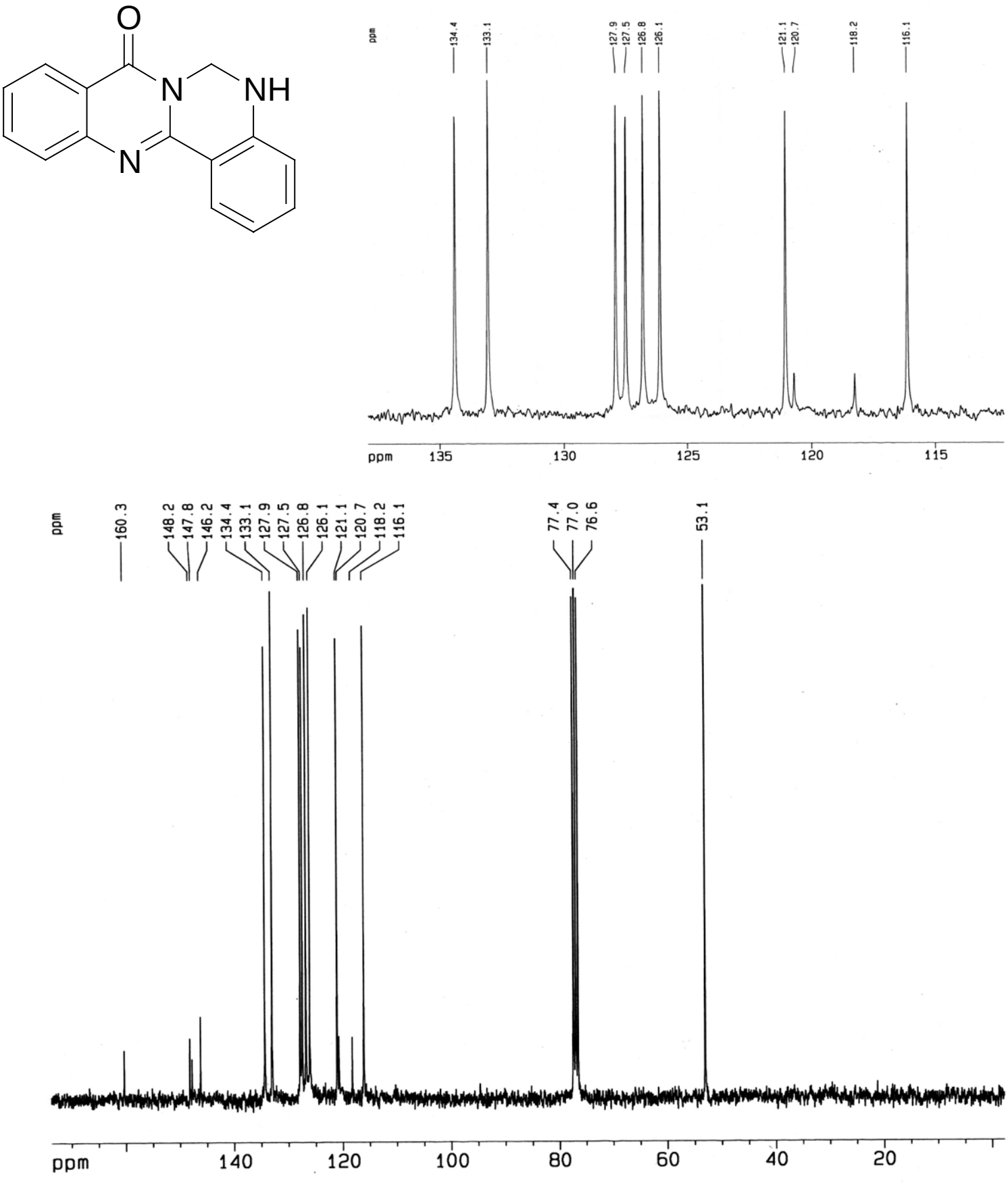
S-57: HMBC Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5i).



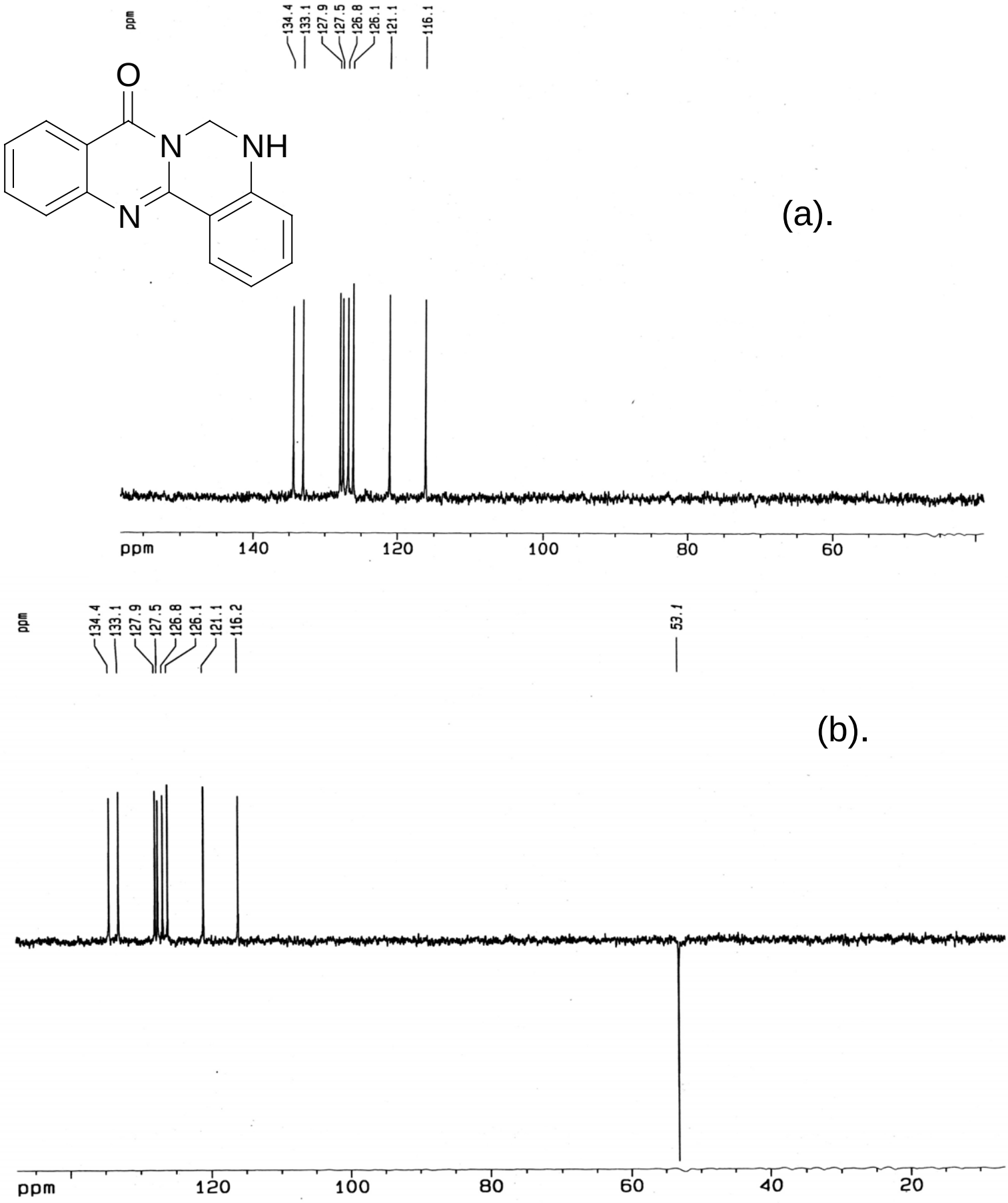
S-58: Expanded Region of the HMBC Spectrum of 11-Methyl-6-phenyl-5,6-dihydro-quinazolino[4,3-b]quinazolin-8-one (5i).



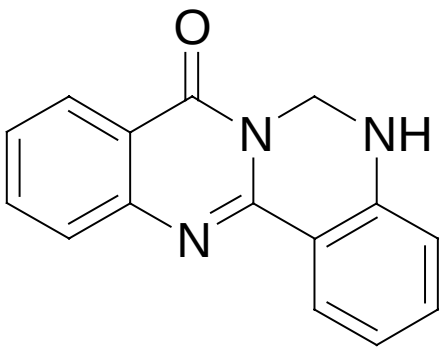
S-59: 1H NMR Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).

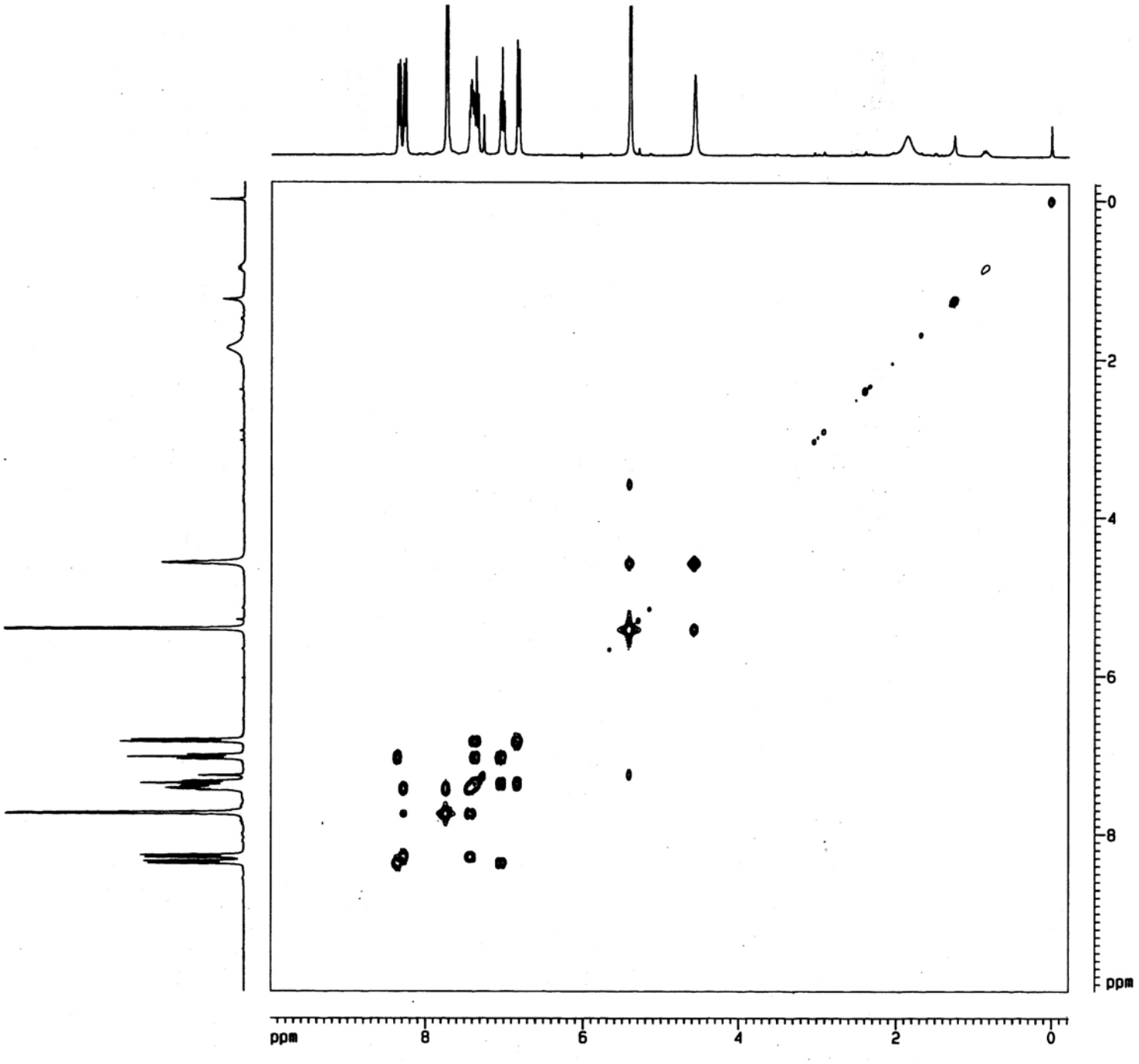


S-60: 13C NMR Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).

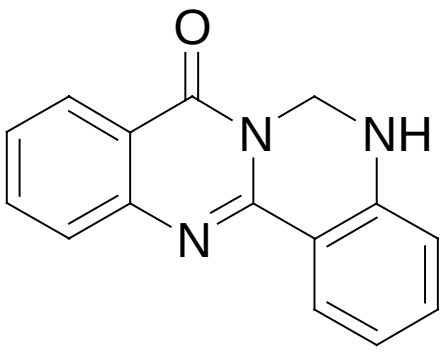


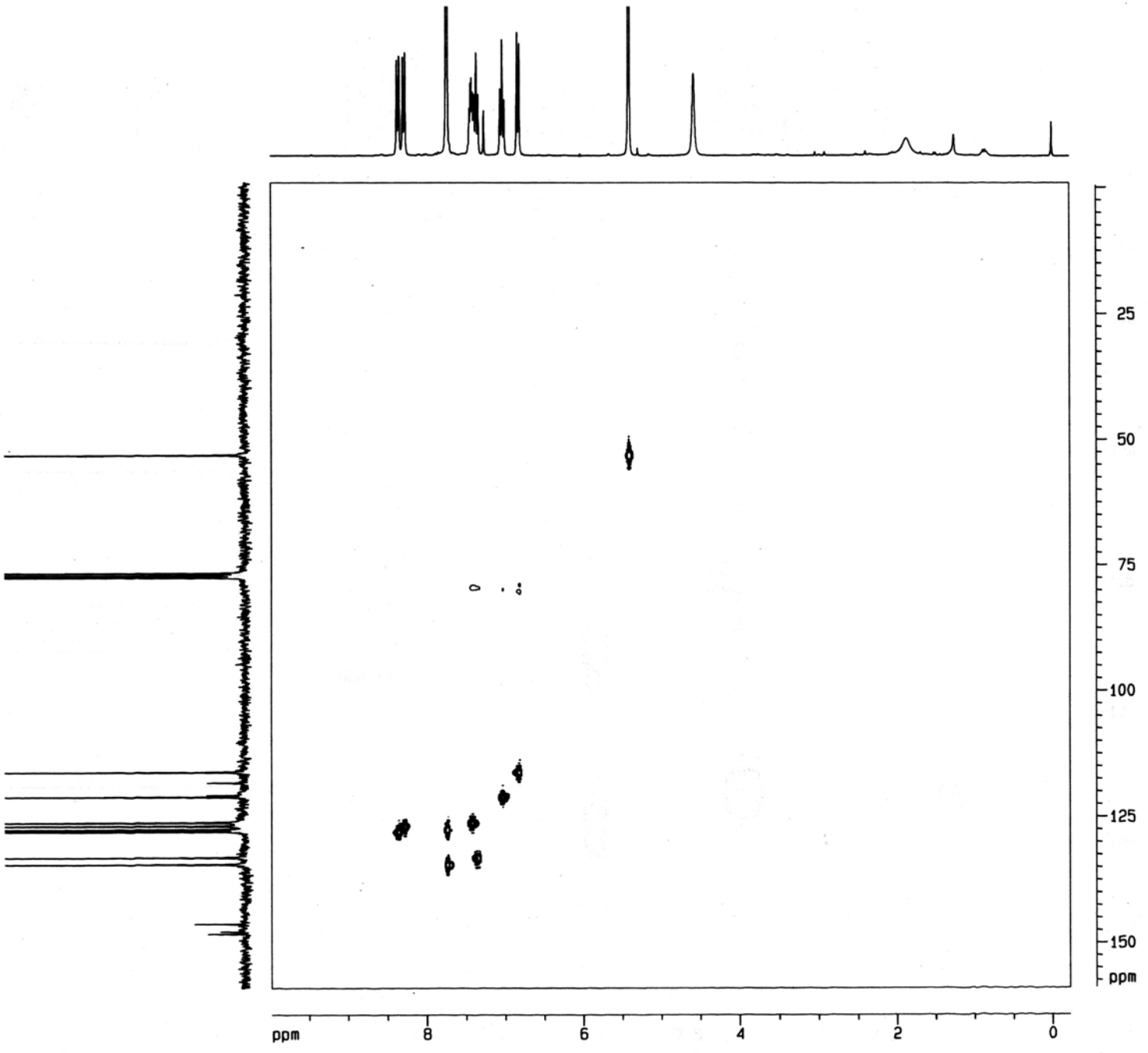
S-61: (a). DEPT-90 and (b). DEPT-135 NMR Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).



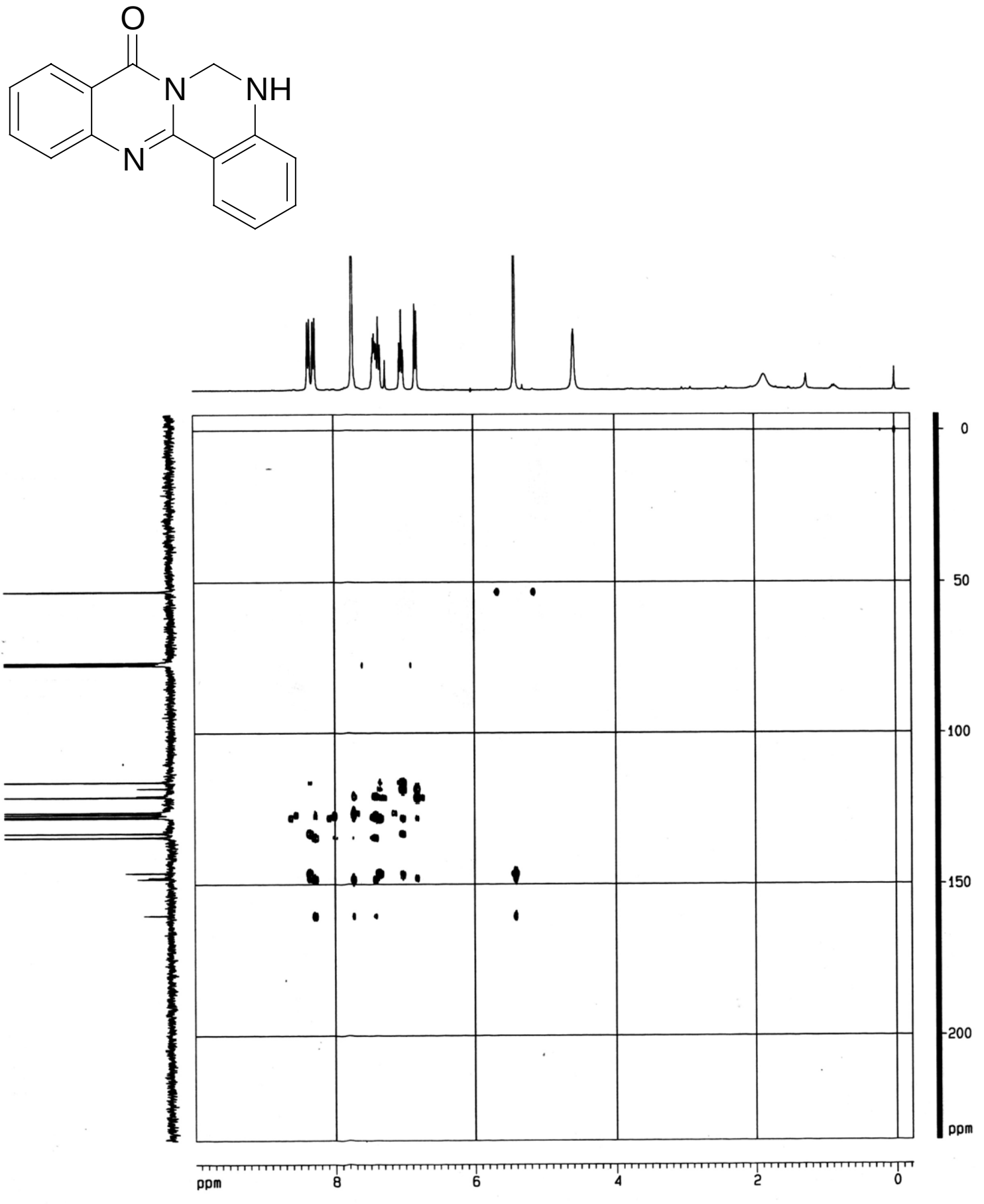


S-62:COSY Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).

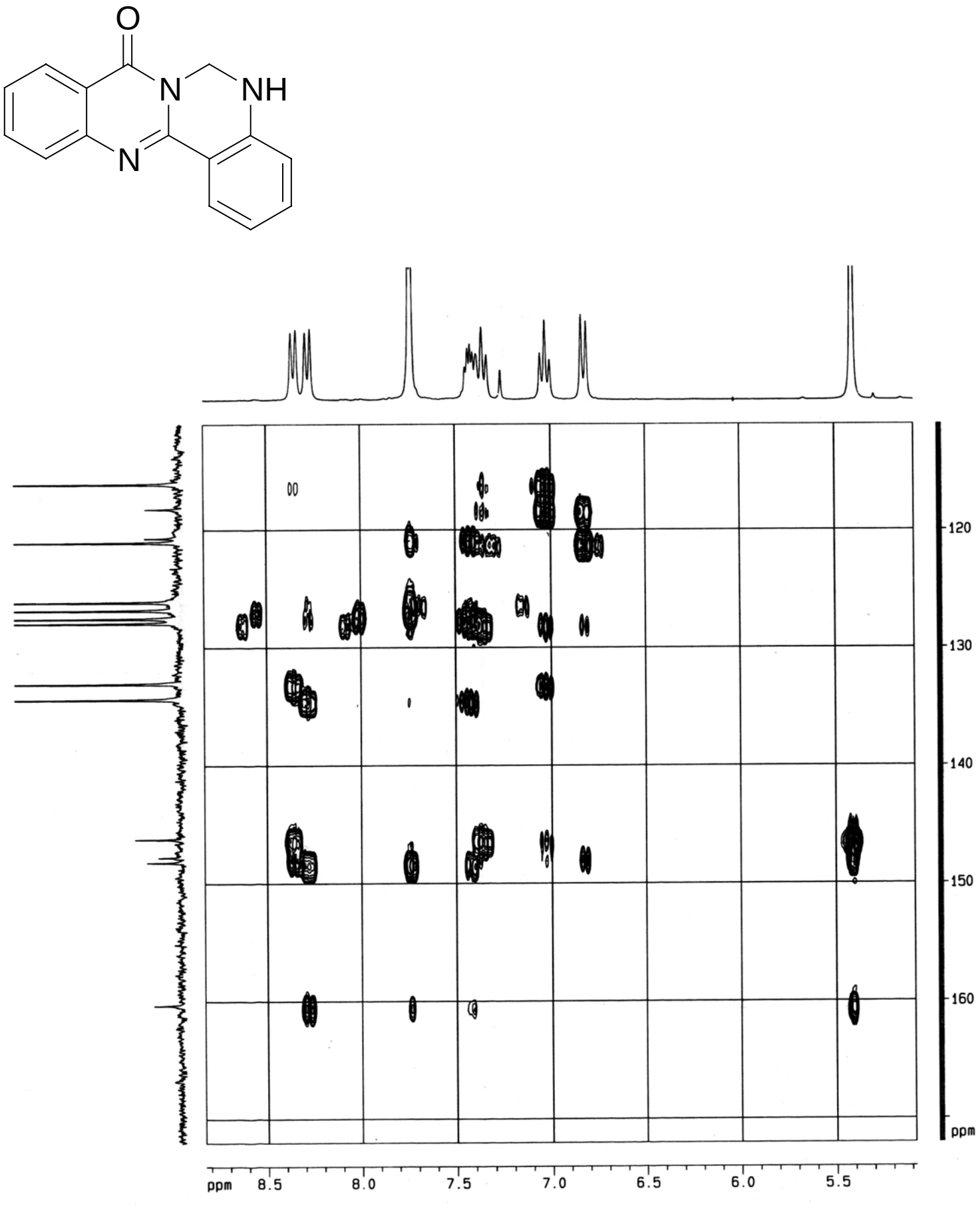




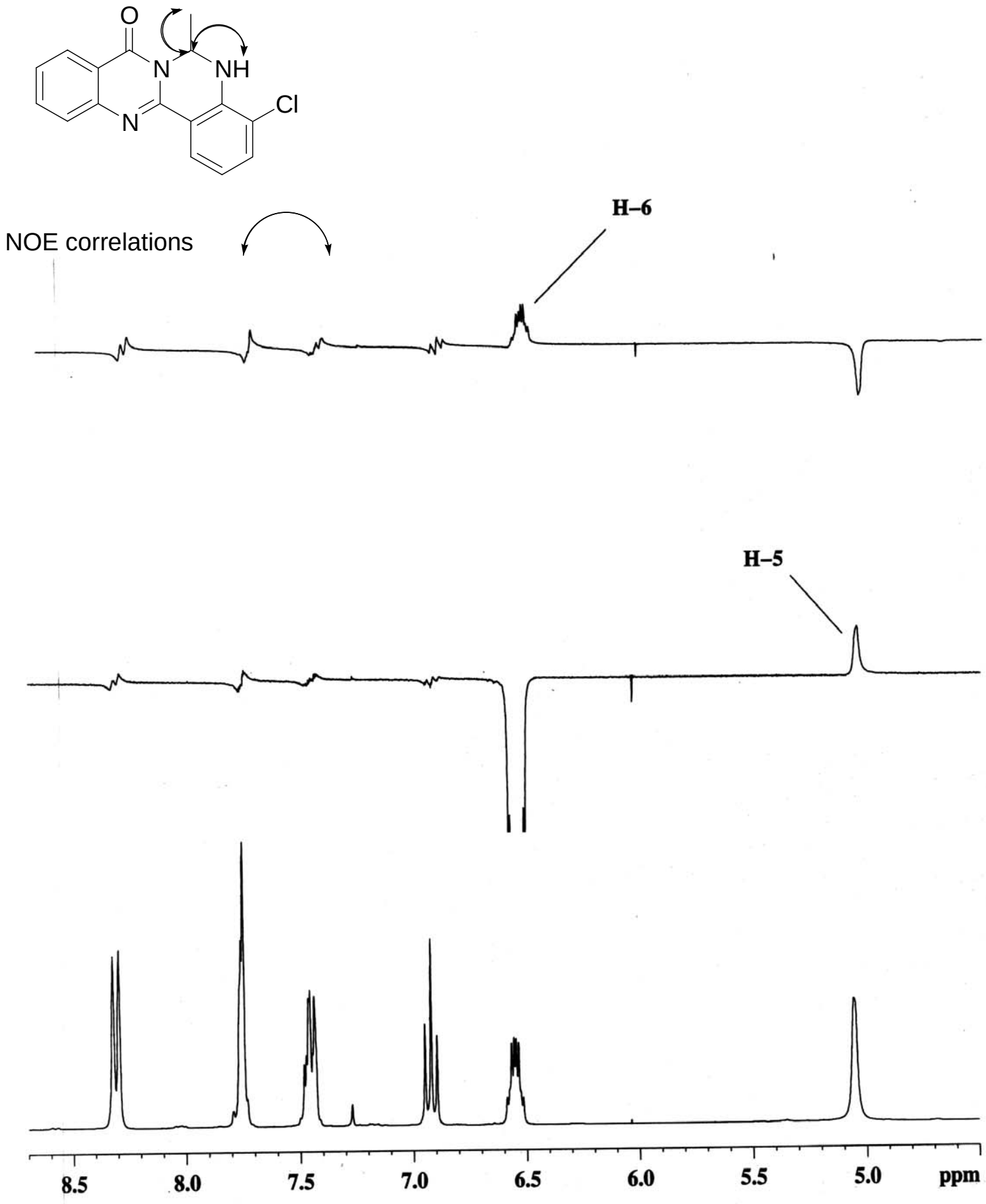
S-63: HSQC Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).



S-64: HMBC Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).



S-65: Expanded Region of the HMBC Spectrum of 5,6-Dihydro-quinazolino[4,3-b]quinazolin-8-one (5j).



S-66: 1D-NOE spectrum of 4-Chloro-6-methyl-5,6-dihydro-quinazolino[4,3-*b*]quinazolin-8-one (**5f**).