

Supporting Information

Construction of C(sp²)-S and C(sp²)-Se Bonds *via* Silver(I)-Mediated Coupling Reaction of Heterocyclic Ketene Aminals with Diaryl Dichalcogenides

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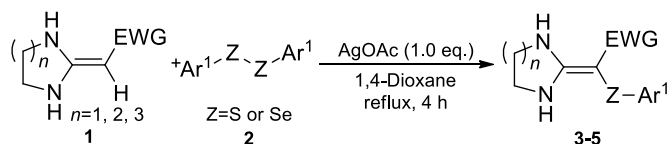
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General Information

All compounds were fully characterized by spectroscopic data. The NMR spectra were recorded on a Bruker DRX500 or DRX400, chemical shifts (δ) are expressed in ppm, and J values are given in Hz, and deuterated CDCl_3 was used as solvent. The reactions were monitored by thin layer chromatography (TLC) using silica gel GF₂₅₄. The melting points were determined on XT-4A melting point apparatus and are uncorrected. HRMs were performed on a Agilent LC/Msd TOF instrument.

All chemicals and solvents were used as received without further purification unless otherwise stated. Compounds **2** were prepared according to the literature.¹

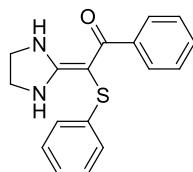
General Procedure for the Preparation of Compounds 3–5



To a 25 ml round-bottom flask, HKAs **1** (1 mmol), 1,2-diphenyldiselenane **2** (1 mmol), AgOAc (1 mmol) in dioxane (15 mL) were added. The resulting mixture was heated to reflux and stirred until all starting diphenyldiselenane was consumed, as evidenced by TLC. After cooling to room temperature, the solid AgOAc was removed by filtration. The organic layer was then concentrated under reduced pressure to yield crude product. The crude compound was then purified by recrystallization with ethyl acetate and petroleum to afford compounds **3-5** as a white solid.

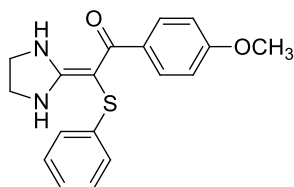
Analytic Data for the Products

2-(Imidazolidin-2-ylidene)-1-phenyl-2-(phenylthio)ethanone (**3a**)



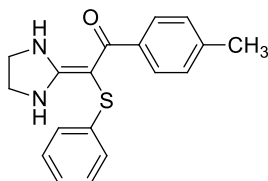
White solid, yield 94%; Mp 202–204 °C; IR (KBr): 3194, 1573, 1482, 1373, 1299, 735 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.53 (t, J = 8.8 Hz, 2H, NCH_2), 3.88 (t, J = 8.8 Hz, 2H, NCH_2), 5.78 (br, 1H, NH), 7.06–7.39 (m, ArH, 10H), 9.91 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.6, 45.8, 76.1, 124.7, 125.0, 127.4, 127.7, 129.1, 129.3, 141.0, 142.6, 168.7, 194.1; HRMS (TOF ES^+): m/z calcd for $\text{C}_{17}\text{H}_{16}\text{N}_2\text{NaOS}$ [(M+Na $^+$)], 319.0876; found, 319.0874.

2-(Imidazolidin-2-ylidene)-1-(4-methoxyphenyl)-2-(phenylthio)ethanone (**3b**)



White solid, yield 97%; mp 176–177 °C; IR (KBr): 3366, 3269, 1576, 1369, 1241, 732 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.57 (t, J = 8.9 Hz, 2H, NCH_2), 3.75 (s, 3H, CH_3), 3.90 (t, J = 8.9 Hz, 2H, NCH_2), 5.72 (br, 1H, NH), 6.74 (t, J = 8.6 Hz, 2H, ArH), 7.06–7.26 (m, 5H, ArH), 7.45 (t, J = 8.6 Hz, 2H, ArH), 9.95 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.6, 45.8, 55.6, 75.6, 112.9, 124.6, 125.0, 129.3, 129.6, 134.9, 141.0, 160.6, 168.9, 193.2; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{18}\text{N}_2\text{NaO}_2\text{S}$ [(M+Na $^+$)], 349.0981; found, 349.0971.

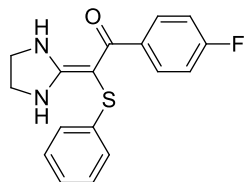
2-(Imidazolidin-2-ylidene)-2-(phenylthio)-1-(*p*-tolyl)ethanone (**3c**)



White solid, yield 94%; mp 213–214 °C; IR (KBr): 3416, 3285, 1579, 1531, 1365, 1298, 744 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 2.28 (s, 3H, CH_3), 3.57 (t, J = 8.8 Hz, 2H, NCH_2), 3.91 (t, J = 8.8 Hz, 2H, NCH_2), 5.72 (br, 1H, NH), 7.02 (d, J = 7.8 Hz, 2H, ArH), 7.06–7.33 (m, ArH, 7H), 9.94 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 21.8, 42.6,

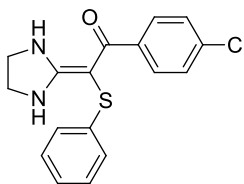
45.8, 75.9, 124.7, 124.9, 127.5, 128.4, 129.3, 139.1, 139.7, 141.0, 168.7, 194.1; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₈N₂NaOS [(M+Na⁺)], 333.1032; found, 333.1027.

1-(4-Fluorophenyl)-2-(imidazolidin-2-ylidene)-2-(phenylthio)ethanone (**3d**)



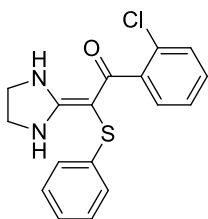
White solid, yield 93%; mp 182–183 °C; IR (KBr): 3305, 1580, 1532, 1370, 1303, 743 cm⁻¹; ¹H NMR (500MHz,CDCl₃): δ = 3.50–3.57 (m, 2H, NCH₂), 3.85–3.89 (m, 2H, NCH₂), 5.80 (br, 1H, NH), 6.84–7.41 (m, 9H, ArH), 9.86 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃) : δ = 42.6, 45.8, 75.9, 114.5 (d, J = 20.0 Hz), 124.6, 125.1, 129.4, 129.7, 138.6, 140.7, 163.4 (d, J = 245.0 Hz), 168.7, 192.7; HRMS (TOF ES⁺): m/z calcd for C₁₇H₁₅FN₂NaOS [(M+Na⁺)], 337.0781; found, 337.0783.

1-(4-Chlorophenyl)-2-(imidazolidin-2-ylidene)-2-(phenylthio)ethanone (**3e**)



White solid, yield 89%; mp 200–202 °C; IR (KBr): 3413, 3267, 1579, 1532, 1370, 1306, 742 cm⁻¹; ¹H NMR (500 MHz,CDCl₃): δ = 3.56 (t, J = 8.9 Hz, 2H, NCH₂), 3.90 (t, J = 8.9 Hz, 2H, NCH₂), 5.80 (br, 1H, NH), 7.06–7.34 (m, 9H, ArH), 9.85 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 42.6, 45.8, 76.0, 124.6, 125.2, 127.9, 129.0, 129.4, 134.9, 140.6, 140.9, 168.7, 192.6; HRMS (TOF ES⁺): m/z calcd for C₁₇H₁₅ClN₂NaOS [(M+Na⁺)], 353.0486; found, 353.0486.

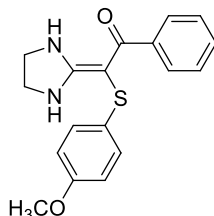
1-(2-Chlorophenyl)-2-(imidazolidin-2-ylidene)-2-(phenylthio)ethanone (**3f**)



White solid, yield 85%; mp 194–195 °C; IR (KBr): 3193, 1582, 1540, 1482, 1381, 1299, 747 cm⁻¹; ¹H NMR (500 MHz,CDCl₃): δ = 3.58 (t, J = 8.9 Hz, 2H, NCH₂), 3.92 (t, J = 8.9 Hz, 2H, NCH₂), 5.80 (br, 1H, NH), 7.05–7.30 (m, 9H, ArH), 9.74 (br, 1H, NH); ¹³C

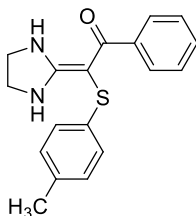
NMR (125 MHz, CDCl₃): δ = 42.5, 45.7, 125.1, 126.3, 127.6, 129.0, 129.5, 130.6, 140.2, 142.2, 168.0, 192.1; HRMS (TOF ES⁺): m/z calcd for C₁₇H₁₅ClN₂NaOS [(M+Na⁺)], 353.0486; found, 353.0485.

2-(Imidazolidin-2-ylidene)-2-((4-methoxyphenyl)selanyl)-1-phenylethanone (**3g**)



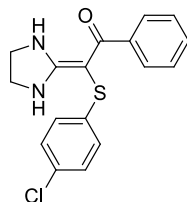
White solid, yield 94%, mp 218–220 °C; IR (KBr): 3403, 3304, 1589, 1533, 1482, 1367, 1298, 700 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 3.55 (t, J = 8.9 Hz, 2H, NCH₂), 3.70 (s, 3H, CH₃), 3.88 (t, J = 8.9 Hz, 2H, NCH₂), 5.82 (br, 1H, NH), 6.78–7.40 (m, 9H, ArH), 9.87 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 42.6, 45.8, 55.8, 115.1, 126.4, 127.1, 127.4, 127.7, 129.1, 131.6, 142.7, 157.9, 168.8, 194.1; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₉N₂O₂S [(M+H⁺)], 327.1162; found, 327.1168.

2-(Imidazolidin-2-ylidene)-1-phenyl-2-(*p*-tolylselanyl)ethanone (**3h**)



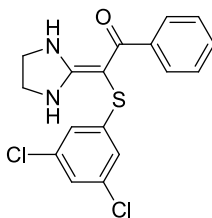
White solid, yield 95%; mp 196–197 °C; IR (KBr): 3424, 3318, 1578, 1359, 1298, 702 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 2.29 (s, 3H, ArCH₃), 3.49–3.53 (m, 2H, NCH₂), 3.85–3.88 (m, 2H, NCH₂), 5.77 (br, 1H, NH), 7.01–7.41 (m, 9H, ArH), 9.91 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 21.3, 42.6, 45.8, 76.4, 124.8, 127.4, 127.7, 129.1, 130.1, 134.7, 137.4, 142.7, 168.7, 194.0; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₉N₂OS [(M+H⁺)], 311.1213; found, 311.1216.

2-((4-Chlorophenyl)selanyl)-2-(imidazolidin-2-ylidene)-1-phenylethanone (**3i**)



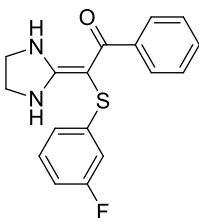
White solid, yield 90%; mp 211–213 °C; IR (KBr): 3200, 1581, 1377, 1299, 705 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 3.58 (t, *J* = 8.7 Hz, 2H, NCH₂), 3.91 (t, *J* = 8.7 Hz, 2H, NCH₂), 5.73 (br, 1H, NH), 7.03–7.37 (m, 9H, ArH), 9.90 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 42.6, 45.8, 75.8, 126.0, 127.3, 127.7, 129.2, 129.3, 130.7, 139.6, 142.4, 168.6, 194.2; HRMS (TOF ES⁺): *m/z* calcd for C₁₇H₁₆ClN₂OS [(M+H⁺)], 331.0666; found, 331.0663.

2-((3,5-Dichlorophenyl)selanyl)-2-(imidazolidin-2-ylidene)-1-phenylethanone (**3j**)



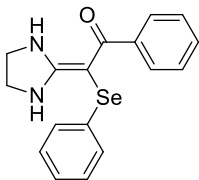
White solid, yield 84%; mp 213–214 °C; IR (KBr): 3208, 1575, 1377, 1300, 793 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 3.55–3.61 (m, 2 H, NCH₂), 3.89–3.94 (m, 2 H, NCH₂), 5.76 (br, 1 H, NH), 6.95–7.34 (m, 8H, ArH), 9.89 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 42.6, 45.8, 74.7, 122.7, 125.2, 127.2, 127.8, 129.3, 135.8, 142.2, 145.3, 168.3, 194.1; HRMS (TOF ES⁺): *m/z* calcd for C₁₇H₁₅Cl₂N₂OS [(M+H⁺)], 365.0277; found, 365.0258.

2-((3-Fluorophenyl)selanyl)-2-(imidazolidin-2-ylidene)-1-phenylethanone (**3k**)



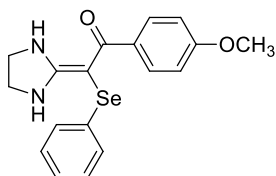
White solid, yield 88%; mp 204–205 °C; IR (KBr): 3423, 3305, 1581, 1367, 1299, 733 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 3.53 (t, *J* = 8.8 Hz, 2 H, NCH₂), 3.88 (t, *J* = 8.8 Hz, 2 H, NCH₂), 5.79 (br, 1 H, NH), 6.73–7.36 (m, 9H, ArH), 9.89 (br, 1 H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 42.6, 45.8, 75.5, 111.5 (d, *J* = 23.7 Hz), 111.9 (d, *J* = 21.3 Hz), 112.0, 120.3, 127.3, 127.8, 129.2, 130.6, 144.0, 163.2 (d, *J* = 245.0 Hz), 168.5, 194.1; HRMS (TOF ES⁺): *m/z* calcd for C₁₇H₁₆FN₂OS [(M+H⁺)], 315.0962; found, 315.0965.

2-(Imidazolidin-2-ylidene)-1-phenyl-2-(phenylselanyl)ethanone (**3l**)



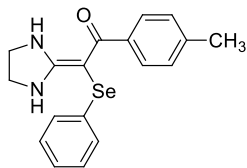
White solid, yield 91%; mp 161–163 °C; IR (KBr): 3409, 3310, 1573, 1368, 1297, 728 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.48–3.52 (m, 2H, NCH_2), 3.86–3.90 (m, 2H, NCH_2), 5.78 (br, 1H, NH), 7.11–7.34 (m, 10H, ArH), 9.98 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.5, 46.0, 73.9, 125.8, 127.3, 127.5, 127.6, 128.9, 129.5, 135.7, 143.7, 168.5, 194.2; HRMS (TOF ES^+): m/z calcd for $\text{C}_{17}\text{H}_{16}\text{N}_2\text{NaOSe}$ [$\text{M}+\text{Na}^+$], 367.0320; found, 367.0315.

2-(Imidazolidin-2-ylidene)-1-(4-methoxyphenyl)-2-(phenylselanyl)ethanone (**3m**)



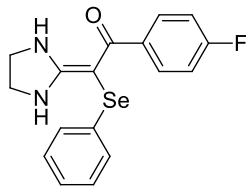
White solid, yield 93%; mp 144–146 °C; IR (KBr): 3309, 1603, 1347, 818, 737 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.57–3.61 (m, 2H, NCH_2), 3.78 (s, 3H, OCH_3), 3.93–3.96 (m, 2H, NCH_2), 5.77 (br, 1H, NH), 6.75–7.43 (m, 9H, ArH), 10.04 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.6, 46.0, 55.6, 73.5, 112.9, 125.8, 127.4, 129.5, 129.6, 135.8, 135.9, 160.5, 168.7, 193.5; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{19}\text{N}_2\text{O}_2\text{Se}$ [$\text{M}+\text{H}^+$] 375.0606; found, 375.0601.

2-(Imidazolidin-2-ylidene)-2-(phenylselanyl)-1-(*p*-tolyl)ethanone (**3n**)



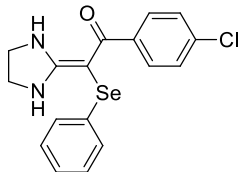
White solid, yield 91%; mp: 205–207 °C; IR (KBr): 3391, 3252, 1572, 1529, 1366, 1300, 733 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 2.12 (s, ArCH_3), 3.55–3.66 (m, 2H, NCH_2), 3.88–3.95 (m, 2H, NCH_2), 5.74 (br, 1H, NH), 6.74 (d, J = 8.6 Hz, 2H, ArH), 7.12–7.27 (m, 5H, ArH), 7.39 (d, J = 8.6 Hz, 2H, ArH), 10.02 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.6, 46.0, 55.6, 67.5, 112.8, 125.8, 127.4, 129.5, 129.6, 135.8, 135.9, 160.5, 168.7, 193.5; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{19}\text{Na}_2\text{OSe}$ [$\text{M}+\text{H}^+$], 359.0657; found, 359.0661.

1-(4-Fluorophenyl)-2-(imidazolidin-2-ylidene)-2-(phenylselanyl)ethanone (**3o**)



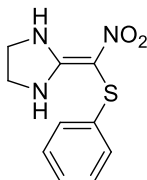
White solid, yield 87%; mp 167–169 °C; IR (KBr): 3412, 3312, 1575, 1525, 1367, 1298, 726 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.53–3.58 (m, 2H, NCH_2), 3.90–3.94 (m, 2H, NCH_2), 5.81 (br, 1H, NH), 6.85–6.90 (m, 2H, ArH), 7.14–7.15 (m, 1H, ArH), 7.22–7.37 (m, 6H, ArH), 9.96 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.5, 46.0, 73.7, 114.5 (d, J = 20.0 Hz), 126.0, 127.4, 129.6, 129.6, 135.5, 139.6, 163.2 (d, J = 246.3 Hz), 168.6, 192.9; HRMS (TOF ES^+): m/z calcd for $\text{C}_{17}\text{H}_{16}\text{FN}_2\text{OSe}$ [($\text{M}+\text{H}^+$)], 363.0406; found, 363.0412.

1-(4-Chlorophenyl)-2-(imidazolidin-2-ylidene)-2-(phenylselanyl)ethanone (**3p**)



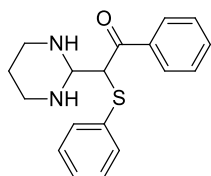
White solid, yield 88%; mp 184–186 °C; IR (KBr): 3405, 3263, 1576, 1528, 1369, 1305, 736 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.56–3.60 (m, 2H, NCH_2), 3.91–3.96 (m, 2H, NCH_2), 5.77 (br, 1H, NH), 7.13–7.28 (m, 9H, ArH), 9.95 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 42.5, 46.0, 73.7, 126.0, 127.3, 127.8, 128.9, 129.6, 134.7, 135.4, 141.9, 168.6, 192.8; HRMS (TOF ES^+): m/z calcd for $\text{C}_{17}\text{H}_{16}\text{ClN}_2\text{OSe}$ [($\text{M}+\text{H}^+$)], 379.0111; found, 379.0103.

2-(nitro(phenylthio)methylene)imidazolidine (**3q**)



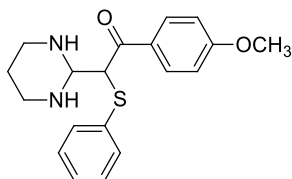
White solid, yield 88%; mp 163.5–164 °C; IR (KBr): 3350, 3256, 1576, 1392, 1335, 738 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 3.75–3.79 (m, 2H, NCH_2), 3.95–4.00 (m, 2H, NCH_2), 5.83 (br, 1H, NH), 7.13–7.17 (m, 3H, ArH), 7.24–7.28 (m, 2H, ArH), 8.83 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 43.1, 45.6, 98.4, 125.6, 125.9, 129.1, 136.1, 163.3; HRMS (TOF ES^+): m/z calcd for $\text{C}_{10}\text{H}_{11}\text{N}_3\text{NaO}_2\text{S}$ [($\text{M}+\text{Na}^+$)], 260.0464; found, 260.0462.

1-Phenyl-2-(phenylthio)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4a**)



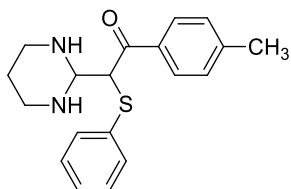
White solid, yield 95%; mp 167–1689 °C; IR (KBr): 3352, 3279, 1586, 1344, 1205, 742 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.79–1.82 (m, 2H, NCH_2), 3.27–3.43 (m, 4H, NCH_2), 6.49 (br, 1H, NH), 7.04–7.08 (m, 3H, ArH), 7.15–7.18 (m, 4H, ArH), 7.24–7.28 (m, 2H, ArH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 38.8, 39.5, 78.4, 124.7, 125.0, 126.8, 127.7, 128.5, 129.2, 141.0, 143.9, 160.8, 193.0; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{19}\text{N}_2\text{OS}$ [($\text{M}+\text{H}^+$)], 311.1213; found, 211.1209.

1-(4-Methoxyphenyl)-2-(phenylthio)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4b**)



White solid, yield 98%; mp 177–178 °C; IR (KBr): 3364, 3050, 1585, 1344, 1241, 735 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.93–1.96 (m, 2H, CH_2), 3.30–3.35 (m, 2H, NCH_2), 3.43–3.49 (m, 2H, NCH_2), 3.74 (s, 3H, OCH_3), 6.49 (br, 1H, NH), 6.71 (d, J = 7.3 Hz, 2H, ArH), 7.08–7.26 (m, 5H, ArH), 7.33 (d, J = 7.3 Hz, 2H, ArH), 12.05 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 38.8, 39.5, 55.5, 112.9, 124.6, 125.0, 128.9, 129.3, 136.3, 141.1, 160.0, 160.9, 192.4; HRMS (TOF ES^+): m/z calcd for $\text{C}_{19}\text{H}_{21}\text{N}_2\text{O}_2\text{S}$ [($\text{M}+\text{H}^+$)], 341.1318; found, 341.1316.

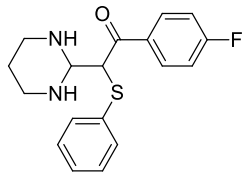
2-(Phenylthio)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)-1-(*p*-tolyl)ethanone (**4c**)



White solid, yield 96%; mp 175–177 °C; IR (KBr): 3330, 3052, 1584, 1339, 1164, 746 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.88–1.92 (m, 2H, NCH_2), 2.26 (s, 3H, ArCH_3), 3.25–3.29 (m, 2H, NCH_2), 3.40–3.47 (m, 2H, NCH_2), 6.48 (br, 1H, NH), 6.98 (d, J = 7.6 Hz, 2H, ArH), 6.97–7.12 (m, 3H, ArH), 7.22–7.25 (m, 4H, ArH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 21.8, 38.8, 39.5, 78.2, 124.6, 125.0, 127.0, 128.3, 129.2, 138.3, 141.1,

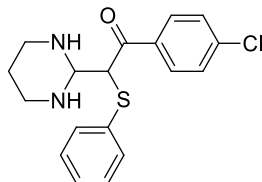
160.8, 193.0; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₁N₂OS, [(M+H⁺)], 325.1369; found, 325.1365.

1-(4-Fluorophenyl)-2-(phenylthio)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4d**)



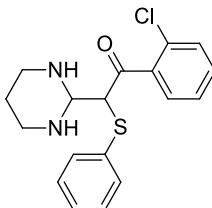
White solid, yield 92%; mp 122–124 °C; IR (KBr): 3322, 3060, 1586, 1343, 1210, 746 cm⁻¹; ¹H NMR (400 MHz, CDCl₃): δ = 1.93–1.99 (m, 2H, CH₂), 3.39–3.43 (m, 4H, NCH₂), 6.51 (br, 1H, NH), 6.83–7.32 (m, 9H, ArH), 11.92 (br, 1H, NH); ¹³C NMR (100 MHz, CDCl₃): δ = 20.0, 39.0, 39.0, 77.9, 114.1 (d, *J* = 26.3 Hz), 124.2, 124.7, 128.6 (d, *J* = 8.0 Hz), 128.9, 139.4, 140.3, 160.4, 162.5 (d, *J* = 245.0 Hz), 191.3; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₇FN₂NaOS [(M+Na⁺)], 351.0938; found, 351.0637.

1-(4-Chlorophenyl)-2-(phenylthio)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4e**)



White solid, yield 90%; mp 180–181 °C; IR (KBr): 330, 3052, 1584, 1339, 1164, 746 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.95–1.98 (m, 2H, CH₂), 3.39–3.44 (m, 4H, NCH₂), 6.52 (br, 1H, NH), 7.07–7.48 (m, 9H, ArH), 11.85 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 20.4, 39.1, 39.1, 78.5, 124.6, 125.2, 127.9, 128.5, 129.3, 134.2, 140.6, 142.2, 160.8, 191.6; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₇ClN₂NaOS [(M+Na⁺)], 367.0642; found, 367.0637.

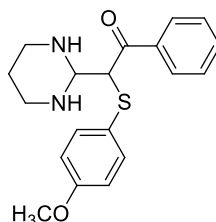
1-(2-Chlorophenyl)-2-(phenylthio)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4f**)



White solid, yield 87%; mp 163–165 °C; IR (KBr): 3261, 1593, 1341, 1210, 747 cm⁻¹; ¹H NMR (400 MHz, CDCl₃): δ = 1.95–1.99 (m, 2H, CH₂), 3.38–3.44 (m, 4H, NCH₂), 7.00–7.28 (m, 9H, ArH); ¹³C NMR (100 MHz, CDCl₃): δ = 19.9, 38.7, 38.7, 79.5, 124.5,

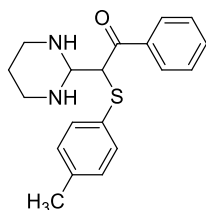
124.7, 125.9, 126.9, 128.4, 128.6, 129.1, 130.1, 139.7, 142.4, 160.0, 189.9; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₇ClN₂NaOS [(M+Na⁺)], 367.0642; found, 367.0635.

2-((4-Methoxyphenyl)selanyl)-1-phenyl-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4g**)



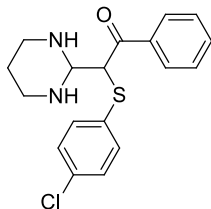
White solid, yield 95%; mp 174–176 °C; IR (KBr): 3368, 3187, 1592, 1354, 1232, 743 cm⁻¹; ¹H NMR (400 MHz, CDCl₃): δ = 1.93–1.96 (m, 2H, CH₂), 3.35–3.45 (m, 4H, NCH₂), 3.76 (s, 3H, OCH₃), 6.56 (br, 1H, NH), 6.78–7.31 (m, 9H, ArH), 11.93 (br, 1H, NH); ¹³C NMR (100 MHz, CDCl₃): δ = 20.1, 38.9, 38.9, 55.5, 79.2, 114.7, 125.8, 126.6, 127.3, 128.1, 131.4, 143.6, 157.5, 160.5, 192.6; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₁N₂O₂S [(M+H⁺)], 341.1318; found, 341.1319.

1-Phenyl-2-(tetrahydropyrimidin-2(1*H*)-ylidene)-2-(*p*-tolylselanyl)ethanone (**4h**)



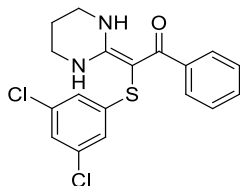
White solid, yield 96%; mp 213–214 °C; IR (KBr): 3369, 1593, 1342, 1208, 800 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.94–1.97 (m, 2H, CH₂), 2.29 (s, 3H, ArCH₃), 3.31–3.34 (m, 2H, NCH₂), 3.45–3.49 (m, 2H, NCH₂), 6.51 (br, 1H, NH), 6.99–7.32 (m, 9H, ArH), 11.97 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 20.4, 21.3, 38.8, 39.5, 78.7, 124.7, 126.9, 127.6, 128.4, 130.0, 134.7, 137.4, 143.9, 160.8, 193.0; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₁N₂O S [(M+H⁺)], 325.1369; found, 325.1364.

2-((4-Chlorophenyl)selanyl)-1-phenyl-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4i**)



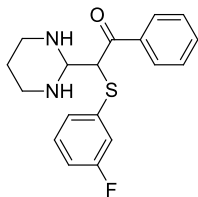
White solid, yield 93%; mp 210–211 °C; IR (KBr): 3360, 1588, 1344, 1210, 809 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.93–1.96 (m, 2H, CH_2), 3.33–3.36 (m, 2H, NCH_2), 3.44–3.47 (m, 2H, NCH_2), 6.43 (br, 1H, NH), 7.00–7.27 (m, 9H, ArH), 11.89 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 38.8, 39.5, 78.1, 125.9, 126.7, 127.7, 128.6, 129.3, 130.7, 139.6, 143.7, 160.7, 193.1; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{18}\text{ClN}_2\text{OS}$ [($\text{M}+\text{H}^+$)], 345.0823, found, 345.0829.

2-((3,5-Dichlorophenyl)selanyl)-1-phenyl-2-(tetrahydropyrimidin-2(1H)-ylidene)ethanone (**4j**)



White solid, yield 85%; mp 223–225 °C; IR (KBr): 3276, 3067, 1596, 1334, 1214, 786 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.96–2.01 (m, 2H, CH_2), 3.34–3.38 (m, 2H, NCH_2), 3.46–3.50 (m, 2H, NCH_2), 6.35 (br, 1H, NH), 6.94–7.27 (m, 8H, ArH), 11.89 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.3, 38.8, 39.6, 122.7, 125.2, 126.7, 127.8, 128.7, 135.8, 143.5, 145.3, 160.5, 193.4; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{17}\text{Cl}_2\text{N}_2\text{OS}$ [($\text{M}+\text{H}^+$)], 379.0433; found, 379.0437.

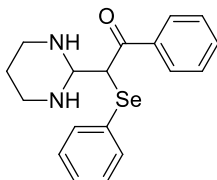
2-((4-Fluorophenyl)selanyl)-1-phenyl-2-(tetrahydropyrimidin-2(1H)-ylidene)ethanone (**4k**)



White solid, yield 92%; mp 181–183 °C; IR (KBr): 3283, 1590, 1342, 1210, 780 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.95–1.98 (m, 2H, CH_2), 3.32–3.36 (m, 2H, NCH_2), 3.67–3.72 (m, 2H, NCH_2), 6.43 (br, 1H, NH), 6.73–7.29 (m, 9H, ArH), 11.93 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 38.8, 39.5, 111.5 (d, J = 23.8 Hz), 112.0 (d,

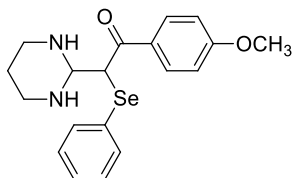
$J = 21.3$ Hz), 120.3, 123.1, 126.8, 127.7, 128.6, 130.5, 143.9, 160.7, 163.6 (d, $J = 246.3$ Hz), 164.8, 193.2; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₈FN₂OS [(M+H⁺)], 329.1118; found, 329.1118.

1-Phenyl-2-(phenylselanyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4l**)



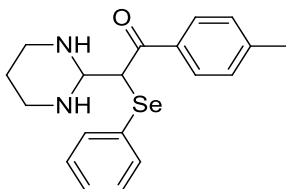
White solid, yield 92%; mp 162–164 °C; IR (KBr): 3353, 1587, 1343, 741 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): $\delta = 1.94$ – 1.98 (m, 2H, CH₂), 3.32–3.37 (m, 2H, NCH₂), 3.46–3.50 (m, 2H, NCH₂), 6.57 (br, ¹H, NH), 7.15–7.31 (m, 10H, ArH), 12.12 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): $\delta = 20.0$, 38.5, 39.2, 77.8, 125.3, 126.3, 126.8, 127.1, 127.7, 129.0, 135.3, 144.6, 160.1, 192.6; HRMS (TOF ES⁺): m/z calcd for C₁₈H₁₉N₂OSe [(M+H⁺)], 359.0657, found, 359.0668.

1-(4-Methoxyphenyl)-2-(phenylselanyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone (**4m**)



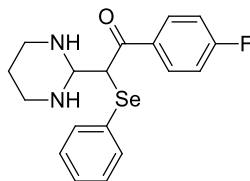
White solid, yield 95%; mp 154–156 °C; IR (KBr): 3353, 1581, 1344, 1240, 799 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): $\delta = 1.90$ – 1.95 (m, 2H, CH₂), 3.33–3.45 (m, 4H, NCH₂), 3.73 (s, 3H, OCH₃), 6.53 (br, 1H, NH), 6.69–7.29 (m, 9H, ArH); ¹³C NMR (125 MHz, CDCl₃): $\delta = 20.5$, 39.2, 39.8, 55.5, 112.8, 125.8, 127.3, 128.8, 129.5, 135.9, 137.6, 159.9, 160.8, 192.5; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₁N₂O₂Se [(M+H⁺)], 389.0763; found, 389.0768.

2-(Phenylselanyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)-1-(*p*-tolyl)ethanone (**4n**)



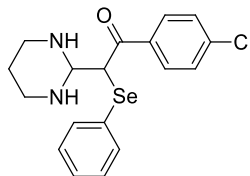
White solid, yield 94%; mp 157–159 °C; IR (KBr): 3363, 3325, 1582, 1341, 1207, 742 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.94–1.97 (m, 2H, CH_2), 2.31 (s, 3H, ArCH_3), 3.34–3.47 (m, 4H, NCH_2), 6.56 (br, 1H, NH), 7.01–7.30 (m, 9H, ArH), 12.17 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.5, 21.8, 39.0, 39.7, 78.1, 125.8, 126.9, 127.3, 128.3, 129.5, 135.9, 138.0, 142.2, 160.7, 193.2; HRMS (TOF ES^+): m/z calcd for $\text{C}_{19}\text{H}_{21}\text{N}_2\text{OSe}$ [($\text{M}+\text{H}^+$)], 373.0814; found, 373.0817.

1-(4-Fluorophenyl)-2-(phenylselanyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone
(4o)



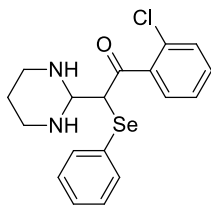
White solid, yield 90%; mp 139–140 °C; IR (KBr): 3322, 1582, 1346, 1210, 740 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.93–1.98 (m, 2H, CH_2), 3.40–3.47 (m, 4H, NCH_2), 6.59 (br, 1H, NH), 6.85–7.30 (m, 9H, ArH), 12.07 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 39.6, 40.9, 78.5, 114.4 (d, J = 21.3 Hz), 125.9, 127.2, 128.9, 129.6, 135.6, 141.1, 160.7, 163.8, 191.8; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{18}\text{FN}_2\text{OSe}$ [($\text{M}+\text{H}^+$)], 377.0563; found, 377.0565.

1-(4-Chlorophenyl)-2-(phenylselanyl)-2-(tetrahydropyrimidin-2(1*H*)-ylidene)ethanone
(4p)



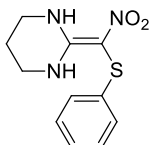
White solid, yield 86%; mp 176–177 °C; IR (KBr): 3319, 3057, 1581, 1345, 1205, 740 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.91–1.94 (m, 2H, CH_2), 3.36–3.41 (m, 4H, NCH_2), 6.58 (br, 1H, NH), 7.12–7.25 (m, 9H, ArH), 11.98 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 39.4, 39.4, 78.3, 126.0, 127.2, 127.8, 128.4, 129.6, 133.9, 135.5, 143.4, 160.6, 191.6; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{18}\text{ClN}_2\text{OSe}$ [($\text{M}+\text{H}^+$)], 393.0265; found, 393.0267.

1-(2-Chlorophenyl)-2-(imidazolidin-2-ylidene)-2-(phenylselanyl)ethanone **(4q)**



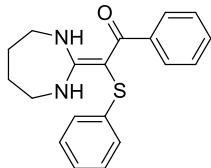
White solid, yield 83%; mp 151–153 °C; IR (KBr): 3350, 1589, 1350, 1211, 747 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.89–1.93 (m, 2H, CH_2), 3.28–3.32 (m, 2H, NCH_2), 3.41–3.45 (m, 2H, NCH_2), 6.49 (br, 1H, NH), 6.99–7.24 (m, 9H, ArH), 11.78 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 20.4, 39.0, 39.6, 79.8, 125.9, 126.4, 127.7, 128.7, 129.4, 130.5, 134.9, 144.0, 160.2, 190.1; HRMS (TOF ES^+): m/z calcd for $\text{C}_{18}\text{H}_{17}\text{ClN}_2\text{NaOSe}$ [($\text{M}+\text{Na}^+$)], 415.0087, found, 415.0083.

2-(nitro(phenylthio)methylene)hexahydropyrimidine (**4r**)



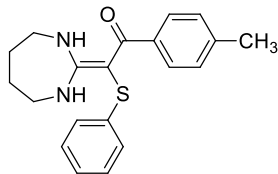
White solid, yield 90%; mp 152.5–153 °C; IR (KBr): 3284, 1585, 1356, 1200, 1127, 738 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.93–1.98 (m, 2H, CH_2), 3.42–3.47 (m, 4H, NCH_2), 7.11–7.15 (m, 3H, ArH), 7.22–7.26 (m, 2H, ArH); ^{13}C NMR (125 MHz, CDCl_3): δ = 19.0, 39.0, 39.0, 100.8, 125.5, 125.9, 129.1, 135.7, 155.7; HRMS (TOF ES^+): m/z calcd for $\text{C}_{11}\text{H}_{13}\text{N}_3\text{NaO}_2\text{S}$ [($\text{M}+\text{Na}^+$)], 274.0621; found, 274.0619.

2-(1,3-Diazepan-2-ylidene)-1-phenyl-2-(phenylthio)ethanone (**5a**)



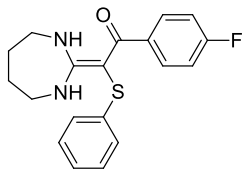
White solid, yield 92%; mp 177–178 °C; IR (KBr): 3363, 1594, 1343, 1202, 742 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.75–1.81 (m, 4H, CH_2CH_2), 3.20–3.23 (m, 2H, NCH_2), 3.45–3.48 (m, 2H, NCH_2), 6.55 (br, 1H, NH), 7.08–7.31 (m, 10H, ArH), 12.14 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 28.0, 28.3, 45.5, 46.3, 81.6, 124.8, 125.1, 126.9, 127.6, 128.7, 129.3, 140.9, 143.9, 170.7, 194.2; HRMS (TOF ES^+): m/z calcd for $\text{C}_{19}\text{H}_{21}\text{N}_2\text{OS}$ [($\text{M}+\text{H}^+$)], 325.1369; found, 325.1368.

2-(1,3-Diazepan-2-ylidene)-2-(phenylselanyl)-1-(*p*-tolyl)ethanone (**5b**)



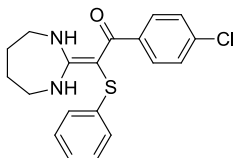
White solid, yield 93%; mp 157–158 °C; IR (KBr): 3317, 2932, 1605, 1349, 1200, 741 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.73–1.80 (m, 4H, CH_2CH_2), 2.29 (s, 3H, ArCH_3), 3.18–3.20 (m, 2H, NCH_2), 3.43–3.47 (m, 2H, NCH_2), 6.54 (br, 1H, NH), 7.01 (d, J = 7.7 Hz, 2H), 7.00–7.13 (m, 3H, ArH), 7.24–7.27 (m, 3H, ArH), 12.20 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 21.8, 28.1, 28.3, 45.5, 46.3, 81.4, 124.8, 125.1, 127.1, 128.3, 128.3, 129.3, 138.6, 141.0, 170.8, 194.2; HRMS (TOF ES^+): m/z calcd for $\text{C}_{20}\text{H}_{23}\text{N}_2\text{OS}$ [($\text{M}+\text{H}^+$)], 339.1526, found, 339.1518.

2-(1,3-Diazepan-2-ylidene)-1-(4-fluorophenyl)-2-(phenylthio)ethanone (**5c**)



White solid, yield 90%; mp 124–125 °C; IR (KBr): 3331, 3060, 1591, 1348, 1207, 847, 740 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.76–1.84 (m, 4H, CH_2CH_2), 3.22–3.26 (m, 2H, NCH_2), 3.46–3.50 (m, 2H, NCH_2), 6.59 (br, 1H, NH), 6.85–6.91 (m, 2H, ArH), 7.09–7.14 (m, 3H, ArH), 7.25–7.29 (m, ArH, 2H), 7.33–7.36 (m, ArH, 2H), 12.12 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 28.0, 28.2, 45.5, 46.3, 81.5, 114.4 (d, J = 21.3 Hz), 124.7, 125.2, 129.2, 129.4, 139.9, 140.6, 163.0 (d, J = 245.0 Hz), 170.7, 192.80; HRMS (TOF ES^+): m/z calcd for $\text{C}_{19}\text{H}_{19}\text{FN}_2\text{NaOS}$ [($\text{M}+\text{Na}^+$)], 365.1094; found, 365.1087.

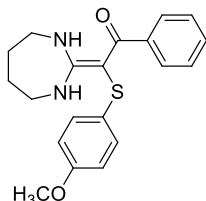
1-(4-Chlorophenyl)-2-(1,3-diazepan-2-ylidene)-2-(phenylthio)ethanone (**5d**)



White solid, yield 87%; mp 156–157 °C; IR (KBr): 3325, 3053, 1594, 1349, 1202, 833, 744 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.68–1.81 (m, 4H, CH_2CH_2), 3.20–3.3.24 (m, 2H, NCH_2), 3.67–3.70 (m, 2H, NCH_2), 6.56 (br, 1H, NH), 7.05–7.25 (m, 9H, ArH), 12.06 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 27.9, 28.2, 45.5, 46.3, 81.5, 124.7, 125.3,

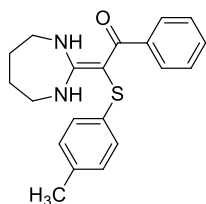
127.8, 128.5, 129.4, 134.4, 140.5, 142.2, 170.6, 192.7; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₀ClN₂OS [(M+H⁺)], 359.0979; found, 359.0975.

2-(1,3-Diazepan-2-ylidene)-2-((4-methoxyphenyl)thio)-1-phenylethanone (**5e**)



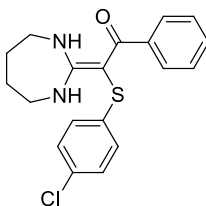
White solid, yield 96%; mp 171–172 °C; IR (KBr): 3316, 2930, 1600, 1342, 1238, 816 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.75–1.79 (m, 4H, CH₂CH₂), 3.19–3.23 (m, 2H, NCH₂), 3.42–3.47 (m, 2H, NCH₂), 3.76 (s, 3H, OCH₃), 6.64 (br, 1H, NH), 6.80 (d, *J* = 8.5 Hz), 6.98 (d, *J* = 8.5 Hz), 7.18–7.26 (m, 3H, ArH), 7.28–7.32 (m, 2H, ArH), 12.11 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 28.0, 28.3, 45.5, 46.3, 55.8, 82.8, 115.1, 126.3, 126.9, 127.6, 128.6, 131.5, 144.0, 157.9, 170.8, 194.1; HRMS (TOF ES⁺): m/z calcd for C₂₀H₂₃N₂O₂S [(M+H⁺)], 355.1475; found, 355.147.

2-(1,3-Diazepan-2-ylidene)-1-phenyl-2-(*p*-tolylthio)ethanone (**5f**)



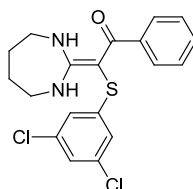
White solid, yield 95%; mp 175–177 °C; IR (KBr): 3334, 3046, 1600, 1345, 1200, 800 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.75–1.81 (m, 4H, CH₂CH₂), 2.30 (s, 3H, ArCH₃), 3.19–3.23 (m, 2H, NCH₂), 3.42–3.48 (m, 2H, NCH₂), 6.58 (br, 1H, NH), 6.98 (d, *J* = 8.2 Hz, 2H, ArH), 7.06 (d, *J* = 8.2 Hz, 2H, ArH), 7.17–7.32 (m, 5H, ArH), 12.14 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 21.3, 28.0, 28.3, 45.5, 46.3, 81.9, 124.8, 126.9, 127.6, 128.6, 130.1, 134.8, 137.2, 143.9, 170.7, 194.1; HRMS (TOF ES⁺): m/z calcd for C₂₀H₂₃N₂OS [(M+H⁺)], 339.1526; found, 339.1527.

2-((4-Chlorophenyl)thio)-2-(1,3-diazepan-2-ylidene)-1-phenylethanone (**5g**)



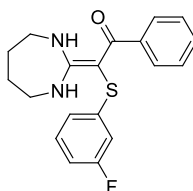
White solid, yield 91%; mp 200–202 °C; IR (KBr): 3318, 1603, 1346, 1199, 803 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.74–1.82 (m, 4H, CH₂CH₂), 3.21–3.23 (m, 2H, NCH₂), 3.45–3.48 (m, 2H, NCH₂), 6.48 (br, 1H, NH), 7.00 (d, *J* = 8.6 Hz, 2H, ArH), 7.17–7.28 (m, 7H, ArH), 12.1 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 27.9, 28.2, 45.5, 46.3, 81.2, 126.0, 126.7, 127.7, 128.8, 129.4, 130.8, 139.5, 143.7, 170.5, 194.2; HRMS (TOF ES⁺): *m/z* calcd for C₁₉H₂₀ClN₂OS [(M+H⁺)], 359.0979; found, 359.0982.

2-(1,3-Diazepan-2-ylidene)-2-((3,5-dichlorophenyl)thio)-1-phenylethanone (**5h**)



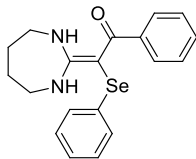
White solid, yield 83%; mp 139–141 °C; IR (KBr): 3308, 1561, 1348, 1206, 791 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.79–1.84 (m, 4H, CH₂CH₂), 3.24–3.28 (m, 2H, NCH₂), 3.47–3.50 (m, 2H, NCH₂), 6.38 (br, 1H, NH), 6.91 (s, 2H, ArH), 7.04 (s, 1H, ArH), 7.20–7.27 (m, 5H, ArH), 12.07 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 27.8, 28.1, 45.4, 46.3, 80.0, 122.7, 125.3, 126.6, 127.8, 128.9, 135.8, 143.5, 145.1, 170.1, 194.3; HRMS (TOF ES⁺): *m/z* calcd for C₁₉H₁₉Cl₂N₂OS [(M+H⁺)], 393.0590; found, 393.0596.

2-(1,3-Diazepan-2-ylidene)-2-((4-fluorophenyl)thio)-1-phenylethanone (**5i**)



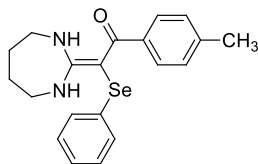
White solid, yield 88%, mp 188–189 °C; IR (KBr): 3255, 1556, 1354, 1210, 776 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): δ = 1.75–1.83 (m, 4H, CH₂CH₂), 3.19–3.23 (m, 2H, NCH₂), 3.42–3.47 (m, 2H, NCH₂), 6.48 (br, 1H, NH), 6.74–6.86 (m, 3H, ArH), 7.15–7.83 (m, 6H, ArH), 12.09 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): δ = 27.9, 28.2, 45.5, 46.3, 111.5 (d, *J* = 23.8 Hz), 112.0 (d, *J* = 21.3 Hz), 120.4, 126.8, 127.3, 127.7, 128.7, 128.8, 130.6, 143.7, 163.8 (d, *J* = 246.3 Hz), 170.4, 194.2; HRMS (TOF ES⁺): *m/z* calcd for C₁₉H₂₀FN₂OS [(M+H⁺)], 343.1275; found, 343.1271.

2-(1,3-Diazepan-2-ylidene)-1-phenyl-2-(phenylselanyl)ethanone (**5j**)



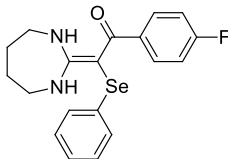
White solid, yield 90%; mp 154–156 °C; IR (KBr): 3347, 1593, 1342, 1202, 737 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.72–1.77 (m, 4H, CH_2CH_2), 3.15–3.19 (m, 2H, CH_2), 3.42–3.46 (m, 2H, CH_2), 6.53 (br, 1H, NH), 7.12–7.28 (m, 10H, ArH), 12.17 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 28.2, 28.2, 45.7, 46.4, 81.6, 125.9, 126.8, 127.5, 127.6, 128.5, 129.5, 135.7, 145.0, 170.7, 194.4; HRMS (TOF ES^+): m/z calcd for $\text{C}_{19}\text{H}_{21}\text{N}_2\text{OSe}$ [(M+ H^+)], 373.0814; found, 373.0816.

2-(1,3-Diazepan-2-ylidene)-2-(phenylselanyl)-1-(*p*-tolyl)ethanone (**5k**)



White solid, yield 93%, mp 125–127 °C; IR (KBr): 3308, 1603, 1346, 1204, 818, 736 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.72–1.77 (m, 4H, CH_2CH_2), 3.15–3.18 (m, 2H, NCH_2), 3.41–3.45 (m, 2H, NCH_2), 6.50 (br, 1H, NH), 7.00 (d, J = 7.4 Hz, 2H, ArH), 7.13–7.26 (m, 7H, ArH), 12.19 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 21.8, 28.1, 28.2, 45.7, 46.5, 81.5, 125.9, 127.0, 127.4, 128.3, 129.5, 135.8, 138.5, 142.1, 170.8, 194.5; HRMS (TOF ES^+): m/z calcd for $\text{C}_{20}\text{H}_{23}\text{N}_2\text{OSe}$ [(M+ H^+)], 387.0970; found, 387.0970.

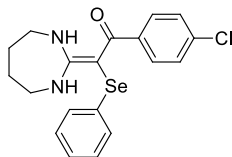
2-(1,3-Diazepan-2-ylidene)-1-(4-fluorophenyl)-2-(phenylselanyl)ethanone (**5l**)



White solid, yield 86%; mp 137–138 °C; IR (KBr): 3319, 3053, 1598, 1349, 1207, 841, 737 cm^{-1} ; ^1H NMR (500 MHz, CDCl_3): δ = 1.73–1.80 (m, 4H, CH_2CH_2), 2.17 (s, 3H, ArH), 3.18–3.22 (m, 2H, NCH_2), 3.43–3.48 (m, 2H, NCH_2), 6.54 (br, 1H, NH), 6.86 (t, J = 8.5 Hz, 2H, ArH), 7.17–7.30 (m, 7H, ArH), 12.14 (br, 1H, NH); ^{13}C NMR (125 MHz, CDCl_3): δ = 28.0, 28.2, 45.6, 46.5, 81.5, 114.4 (d, J = 21.3 Hz), 126.0, 127.4, 129.0,

129.6, 135.5, 141.0, 163.0 (d, $J = 245.0$ Hz), 170.7, 193.1; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₀FN₂OSe [(M+H⁺)], 391.0719; found, 391.0718.

1-(4-Chlorophenyl)-2-(1,3-diazepan-2-ylidene)-2-(phenylselanyl)ethanone (**5m**)



White solid, yield 85%; mp 141–143 °C; IR (KBr): 3315, 3064, 1590, 1346, 1203, 739 cm⁻¹; ¹H NMR (500 MHz, CDCl₃): $\delta = 1.77$ – 1.83 (m, 4H, CH₂CH₂), 3.22–3.25 (m, 2H, NCH₂), 3.46–3.50 (m, 2H, NCH₂), 6.58 (br, 1H, NH), 7.14–7.33 (m, 9H, ArH), 12.15 (br, 1H, NH); ¹³C NMR (125 MHz, CDCl₃): $\delta = 28.0$, 28.0, 45.6, 46.4, 81.5, 126.1, 127.4, 127.8, 128.4, 129.6, 134.3, 135.4, 143.4, 170.6, 192.9; HRMS (TOF ES⁺): m/z calcd for C₁₉H₂₀ClN₂OSe [(M+H⁺)], 407.0424; found, 407.0416.

Crystal X-ray structure and packing diagrams of compound **3g**²

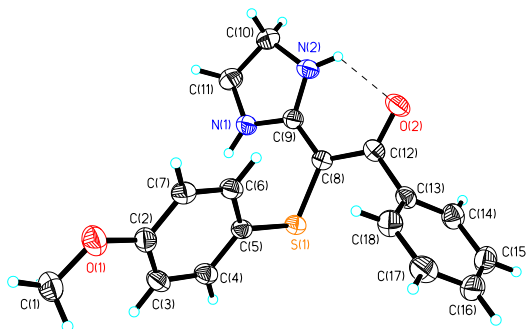


Fig. A ORTEP view of the molecular structure of **3g**, thermal ellipsoids are drawn at 30% probability.

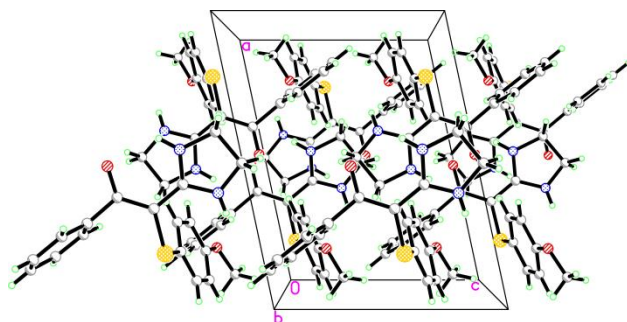


Fig. B Packing in the crystal structure of **3g**, viewed along the *b* axis.

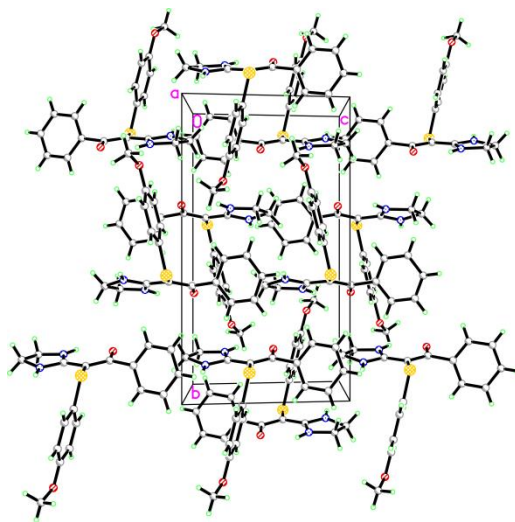


Fig. C Packing in the crystal structure of **3g**, viewed along the *a* axis.

Table S1. Crystal data and structure refinement for **3g**

Empirical formula	C ₁₈ H ₁₈ N ₂ O ₂ S	
Formula weight	326.40	
Temperature	298(2) K	
Wavelength	0.71073 Å	
Crystal system, space group	Monoclinic, P2(1)/c	
Unit cell dimensions	a = 11.5631(18) Å	alpha = 90 deg.
	b = 15.924(3) Å	beta = 102.048(2) deg.
	c = 8.8842(14) Å	gamma = 90 deg.
Volume	1599.8(4) Å ³	
Z, Calculated density	4, 1.355 Mg/m ³	
Absorption coefficient	0.214 mm ⁻¹	
F(000)	688	
Crystal size	0.23 x 0.16 x 0.14 mm	
Theta range for data collection	1.80 to 28.19 deg.	
Limiting indices	-15<=h<=15, -20<=k<=20, -8<=l<=11	
Reflections collected / unique	10822 / 3761 [R(int) = 0.0468]	
Completeness to theta = 28.19	95.5 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.9707 and 0.9525	
Refinement method	Full-matrix least-squares on F ²	
Data / restraints / parameters	3761 / 0 / 209	
Goodness-of-fit on F ²	0.926	
Final R indices [I>2sigma(I)]	R1 = 0.0476, wR2 = 0.1024	
R indices (all data)	R1 = 0.0963, wR2 = 0.1267	
Largest diff. peak and hole	0.178 and -0.268 e.Å ⁻³	

Table S2. Geometric parameters of compound **3g**

<i>Bond lengths (Å)</i>			
N(1)-C(9)	1.314(3)	C(6)-H(6)	0.9300
N(1)-C(11)	1.457(3)	C(7)-H(7)	0.9300
N(1)-H(1)	0.8600	C(8)-C(12)	1.409(3)
N(2)-C(9)	1.334(3)	C(8)-C(9)	1.413(3)
N(2)-C(10)	1.459(3)	C(10)-C(11)	1.509(3)
N(2)-H(2)	0.8600	C(10)-H(10A)	0.9700
O(1)-C(2)	1.378(3)	C(10)-H(10B)	0.9700
O(1)-C(1)	1.417(3)	C(11)-H(11A)	0.9700
O(2)-C(12)	1.254(3)	C(11)-H(11B)	0.9700
S(1)-C(8)	1.755(2)	C(12)-C(13)	1.509(3)

S(1)-C(5)	1.778(2)	C(13)-C(14)	1.379(3)
C(1)-H(1A)	0.9600	C(13)-C(18)	1.386(3)
C(1)-H(1B)	0.9600	C(14)-C(15)	1.380(3)
C(1)-H(1C)	0.9600	C(14)-H(14)	0.9300
C(2)-C(7)	1.381(3)	C(15)-C(16)	1.366(3)
C(2)-C(3)	1.387(3)	C(15)-H(15)	0.9300
C(3)-C(4)	1.383(3)	C(16)-C(17)	1.379(4)
C(3)-H(3)	0.9300	C(16)-H(16)	0.9300
C(4)-C(5)	1.384(3)	C(17)-C(18)	1.381(3)
C(4)-H(4)	0.9300	C(17)-H(17)	0.9300
C(5)-C(6)	1.393(3)	C(18)-H(18)	0.9300
C(6)-C(7)	1.368(3)		
<i>Bond angles (°)</i>			
C(9)-N(1)-C(11)	111.79(19)	N(2)-C(9)-C(8)	126.3(2)
C(9)-N(1)-H(1)	124.1	N(1)-C(9)-C(8)	124.6(2)
C(11)-N(1)-H(1)	124.1	N(2)-C(10)-C(11)	103.20(19)
C(9)-N(2)-C(10)	111.6(2)	N(2)-C(10)-H(10A)	111.1
C(9)-N(2)-H(2)	124.2	C(11)-C(10)-H(10A)	111.1
C(10)-N(2)-H(2)	124.2	N(2)-C(10)-H(10B)	111.1
C(2)-O(1)-C(1)	117.4(2)	C(11)-C(10)-H(10B)	111.1
C(8)-S(1)-C(5)	104.70(10)	H(10A)-C(10)-H(10B)	109.1
O(1)-C(1)-H(1A)	109.5	N(1)-C(11)-C(10)	102.5(2)
O(1)-C(1)-H(1B)	109.5	N(1)-C(11)-H(11A)	111.3
H(1A)-C(1)-H(1B)	109.5	C(10)-C(11)-H(11A)	111.3
O(1)-C(1)-H(1C)	109.5	N(1)-C(11)-H(11B)	111.3
H(1A)-C(1)-H(1C)	109.5	C(10)-C(11)-H(11B)	111.3
H(1B)-C(1)-H(1C)	109.5	H(11A)-C(11)-H(11B)	109.2
O(1)-C(2)-C(7)	115.6(2)	O(2)-C(12)-C(8)	123.6(2)
O(1)-C(2)-C(3)	124.8(2)	O(2)-C(12)-C(13)	117.0(2)
C(7)-C(2)-C(3)	119.5(2)	C(8)-C(12)-C(13)	119.33(19)
C(4)-C(3)-C(2)	119.5(2)	C(14)-C(13)-C(18)	118.1(2)
C(4)-C(3)-H(3)	120.3	C(14)-C(13)-C(12)	120.0(2)
C(2)-C(3)-H(3)	120.3	C(18)-C(13)-C(12)	121.9(2)
C(3)-C(4)-C(5)	121.5(2)	C(13)-C(14)-C(15)	121.3(2)
C(3)-C(4)-H(4)	119.2	C(13)-C(14)-H(14)	119.3
C(5)-C(4)-H(4)	119.2	C(15)-C(14)-H(14)	119.3
C(4)-C(5)-C(6)	117.9(2)	C(16)-C(15)-C(14)	120.0(2)
C(4)-C(5)-S(1)	118.36(17)	C(16)-C(15)-H(15)	120.0
C(6)-C(5)-S(1)	123.73(17)	C(14)-C(15)-H(15)	120.0

C(7)-C(6)-C(5)	121.1(2)	C(15)-C(16)-C(17)	119.8(2)
C(7)-C(6)-H(6)	119.5	C(15)-C(16)-H(16)	120.1
C(5)-C(6)-H(6)	119.5	C(17)-C(16)-H(16)	120.1
C(6)-C(7)-C(2)	120.5(2)	C(16)-C(17)-C(18)	120.1(2)
C(6)-C(7)-H(7)	119.7	C(16)-C(17)-H(17)	120.0
C(2)-C(7)-H(7)	119.7	C(18)-C(17)-H(17)	120.0
C(12)-C(8)-C(9)	120.69(19)	C(17)-C(18)-C(13)	120.7(2)
C(12)-C(8)-S(1)	121.79(16)	C(17)-C(18)-H(18)	119.6
C(9)-C(8)-S(1)	117.33(17)	C(13)-C(18)-H(18)	119.6
N(2)-C(9)-N(1)	109.1(2)		

Torsion angles (°)

C(1)-O(1)-C(2)-C(7)	174.9(2)	C(12)-C(8)-C(9)-N(1)	-179.3(2)
C(1)-O(1)-C(2)-C(3)	-5.1(3)	S(1)-C(8)-C(9)-N(1)	5.6(3)
O(1)-C(2)-C(3)-C(4)	179.1(2)	C(9)-N(2)-C(10)-C(11)	-9.7(3)
C(7)-C(2)-C(3)-C(4)	-0.9(3)	C(9)-N(1)-C(11)-C(10)	-12.1(3)
C(2)-C(3)-C(4)-C(5)	-0.3(3)	N(2)-C(10)-C(11)-N(1)	12.3(3)
C(3)-C(4)-C(5)-C(6)	1.4(3)	C(9)-C(8)-C(12)-O(2)	-3.7(3)
C(3)-C(4)-C(5)-S(1)	-178.29(17)	S(1)-C(8)-C(12)-O(2)	171.19(18)
C(8)-S(1)-C(5)-C(4)	169.52(17)	C(9)-C(8)-C(12)-C(13)	177.3(2)
C(8)-S(1)-C(5)-C(6)	-10.1(2)	S(1)-C(8)-C(12)-C(13)	-7.8(3)
C(4)-C(5)-C(6)-C(7)	-1.3(3)	O(2)-C(12)-C(13)-C(14)	-64.5(3)
S(1)-C(5)-C(6)-C(7)	178.36(17)	C(8)-C(12)-C(13)-C(14)	114.5(2)
C(5)-C(6)-C(7)-C(2)	0.1(4)	O(2)-C(12)-C(13)-C(18)	113.4(3)
O(1)-C(2)-C(7)-C(6)	-179.0(2)	C(8)-C(12)-C(13)-C(18)	-67.6(3)
C(3)-C(2)-C(7)-C(6)	1.0(4)	C(18)-C(13)-C(14)-C(15)	0.5(4)
C(5)-S(1)-C(8)-C(12)	107.30(19)	C(12)-C(13)-C(14)-C(15)	178.5(2)
C(5)-S(1)-C(8)-C(9)	-77.62(18)	C(13)-C(14)-C(15)-C(16)	-0.6(4)
C(10)-N(2)-C(9)-N(1)	2.4(3)	C(14)-C(15)-C(16)-C(17)	-0.2(4)
C(10)-N(2)-C(9)-C(8)	-176.5(2)	C(15)-C(16)-C(17)-C(18)	1.1(4)
C(11)-N(1)-C(9)-N(2)	6.5(3)	C(16)-C(17)-C(18)-C(13)	-1.2(4)
C(11)-N(1)-C(9)-C(8)	-174.6(2)	C(14)-C(13)-C(18)-C(17)	0.4(4)
C(12)-C(8)-C(9)-N(2)	-0.5(3)	C(12)-C(13)-C(18)-C(17)	-177.6(2)
S(1)-C(8)-C(9)-N(2)	-175.64(18)		

Spectra of Target Compounds 3-5

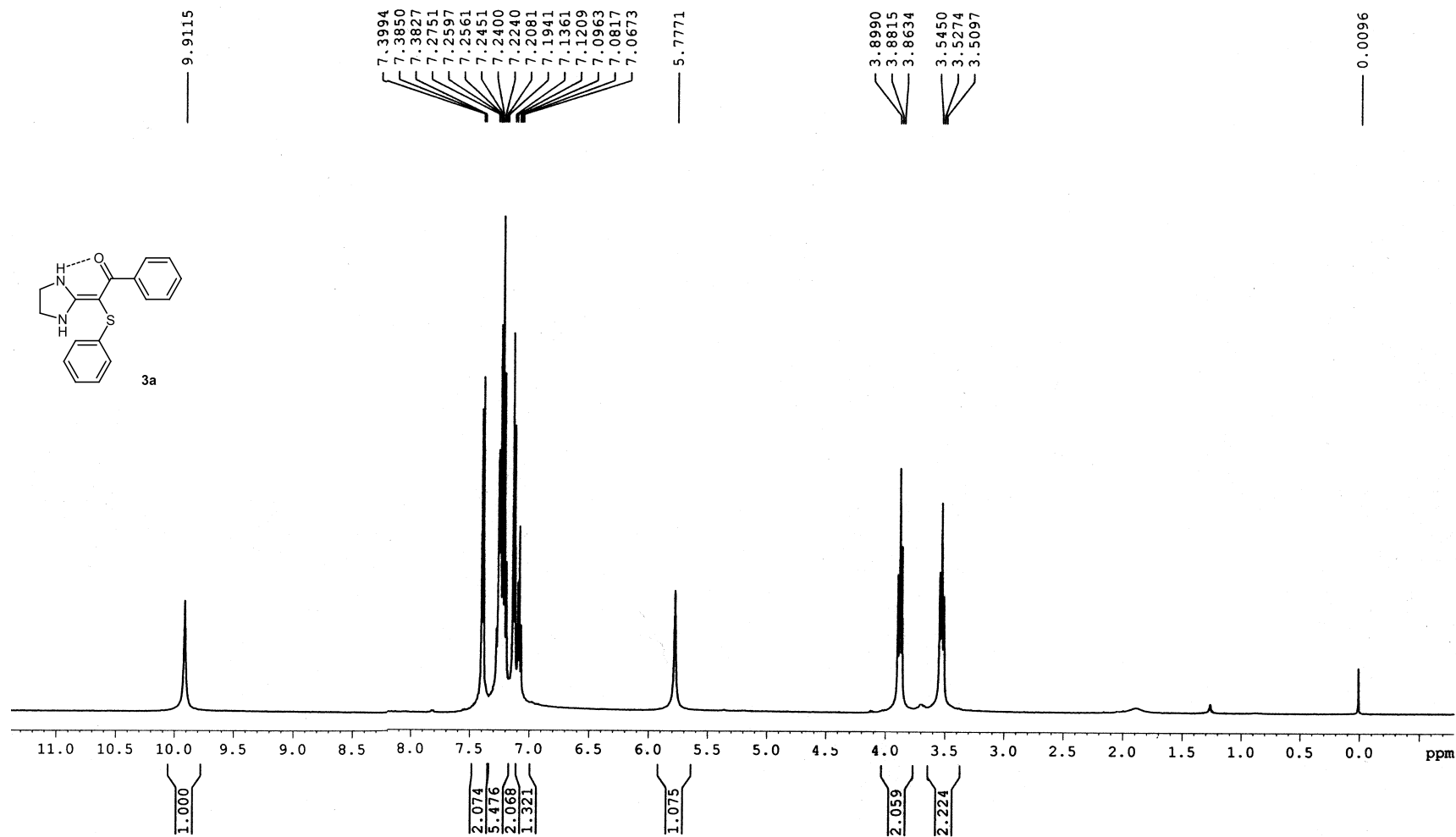


Figure S1 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3a**

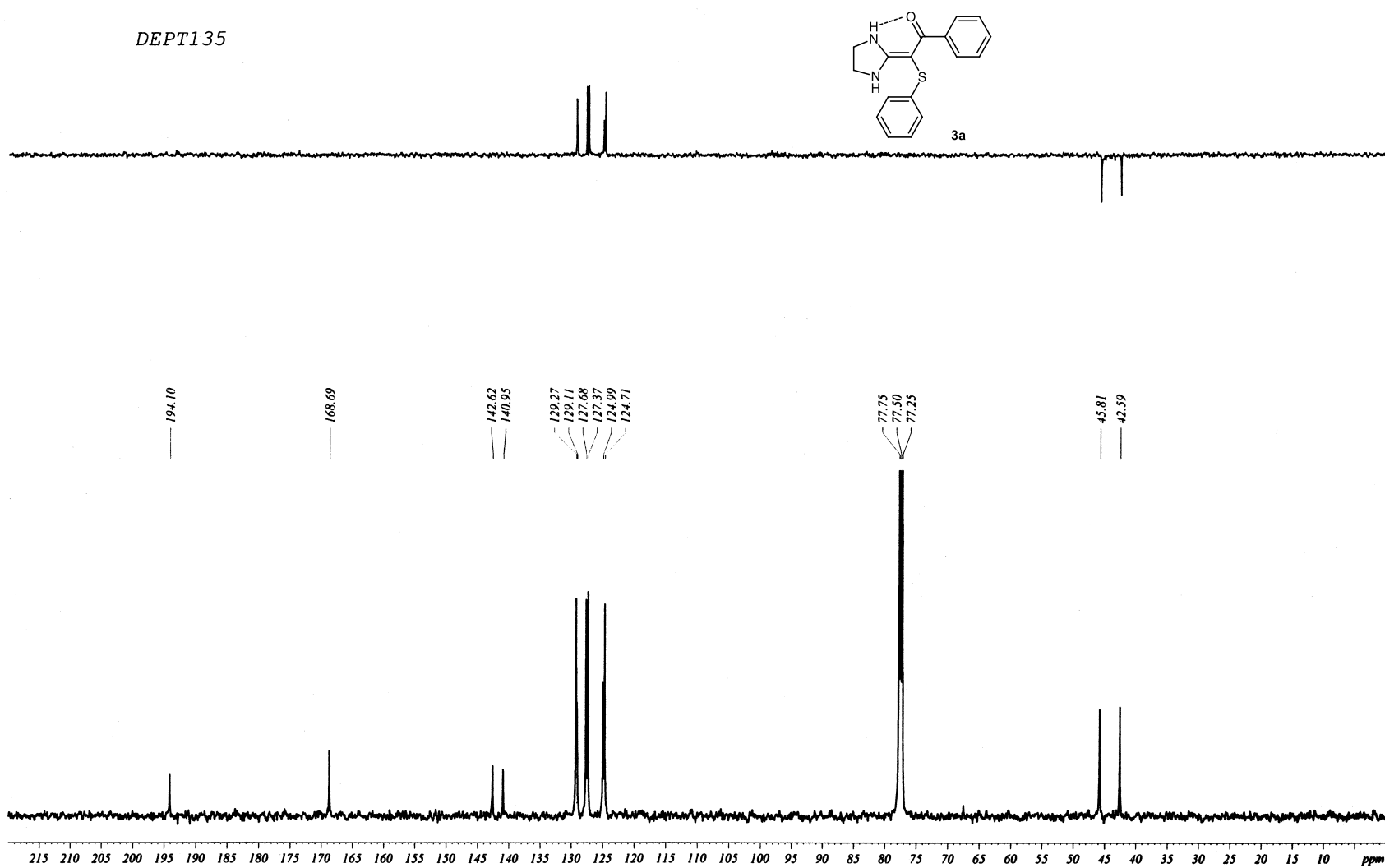


Figure S2 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound 3a

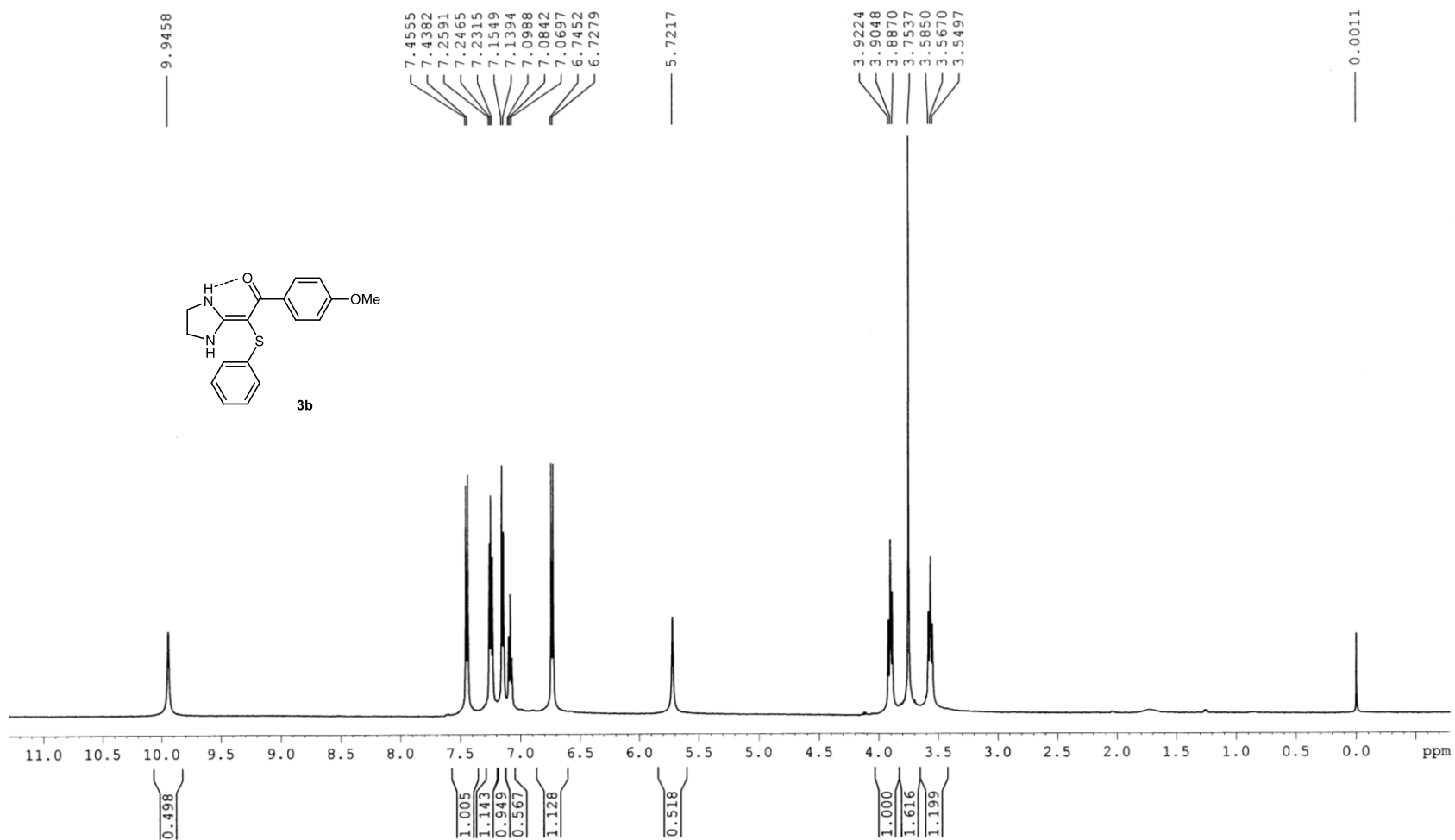


Figure S3 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3b**

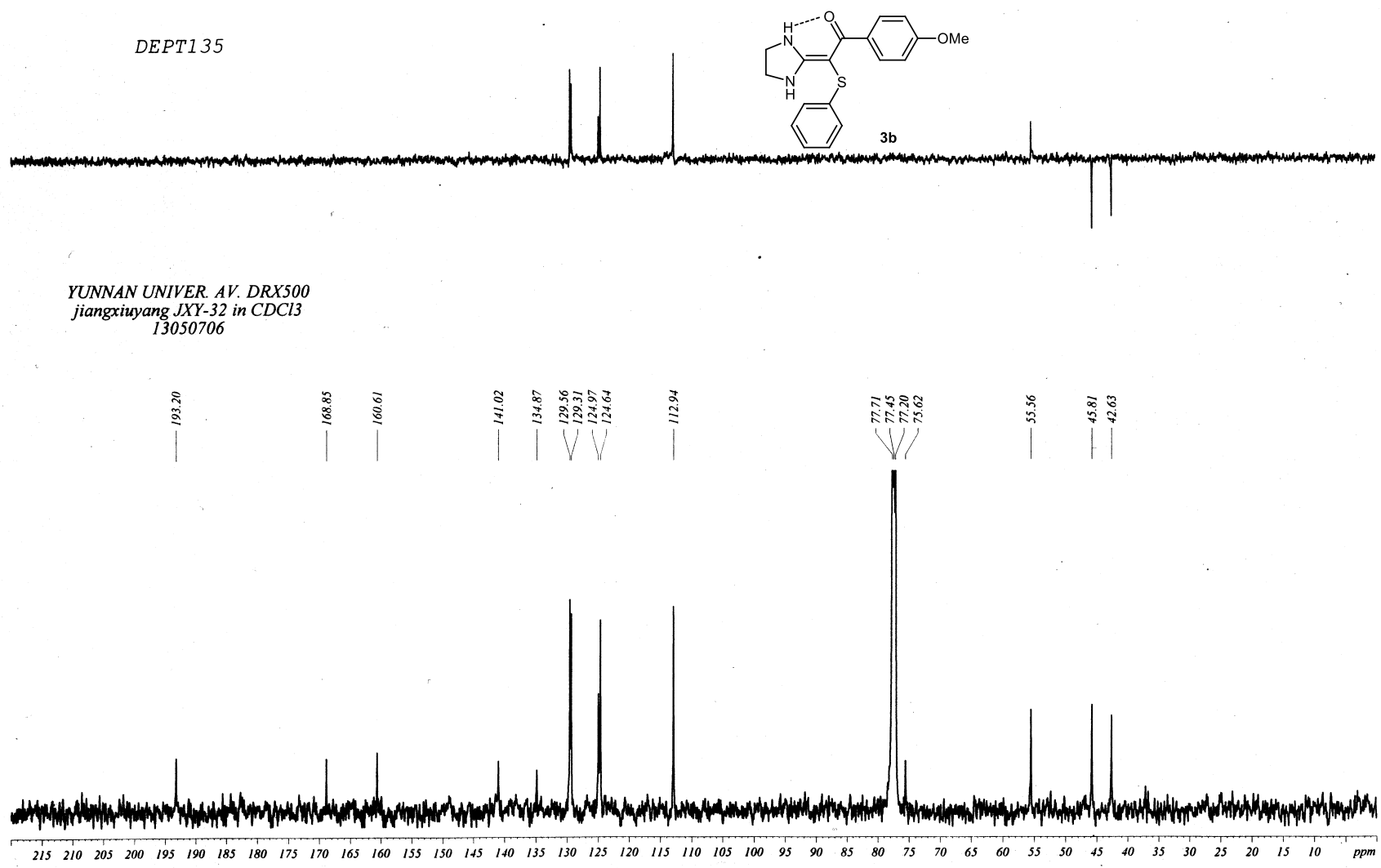


Figure S4 ¹³C NMR spectrum (125 MHz, CDCl₃) of compound 3b

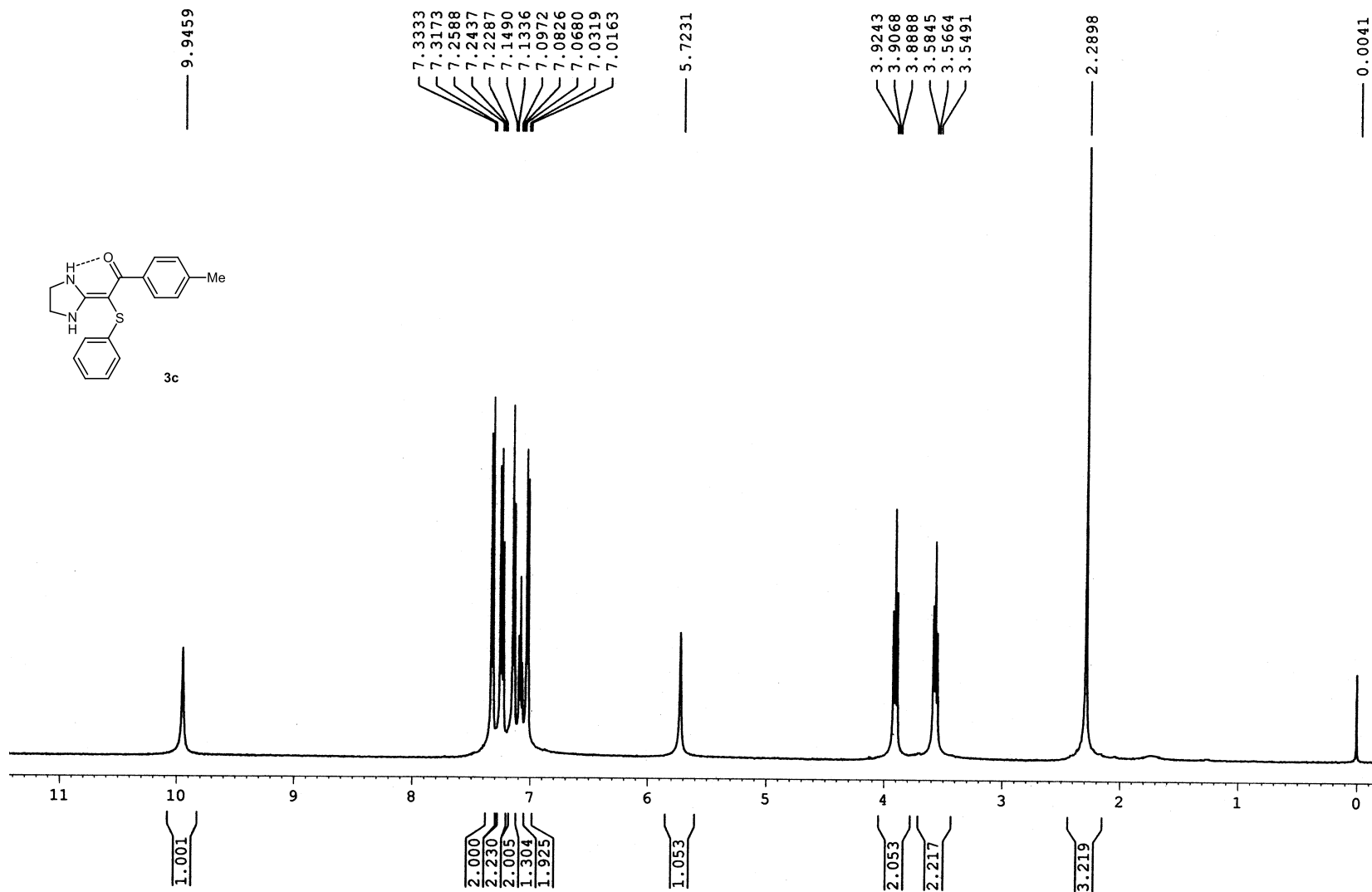


Figure S5 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3c**

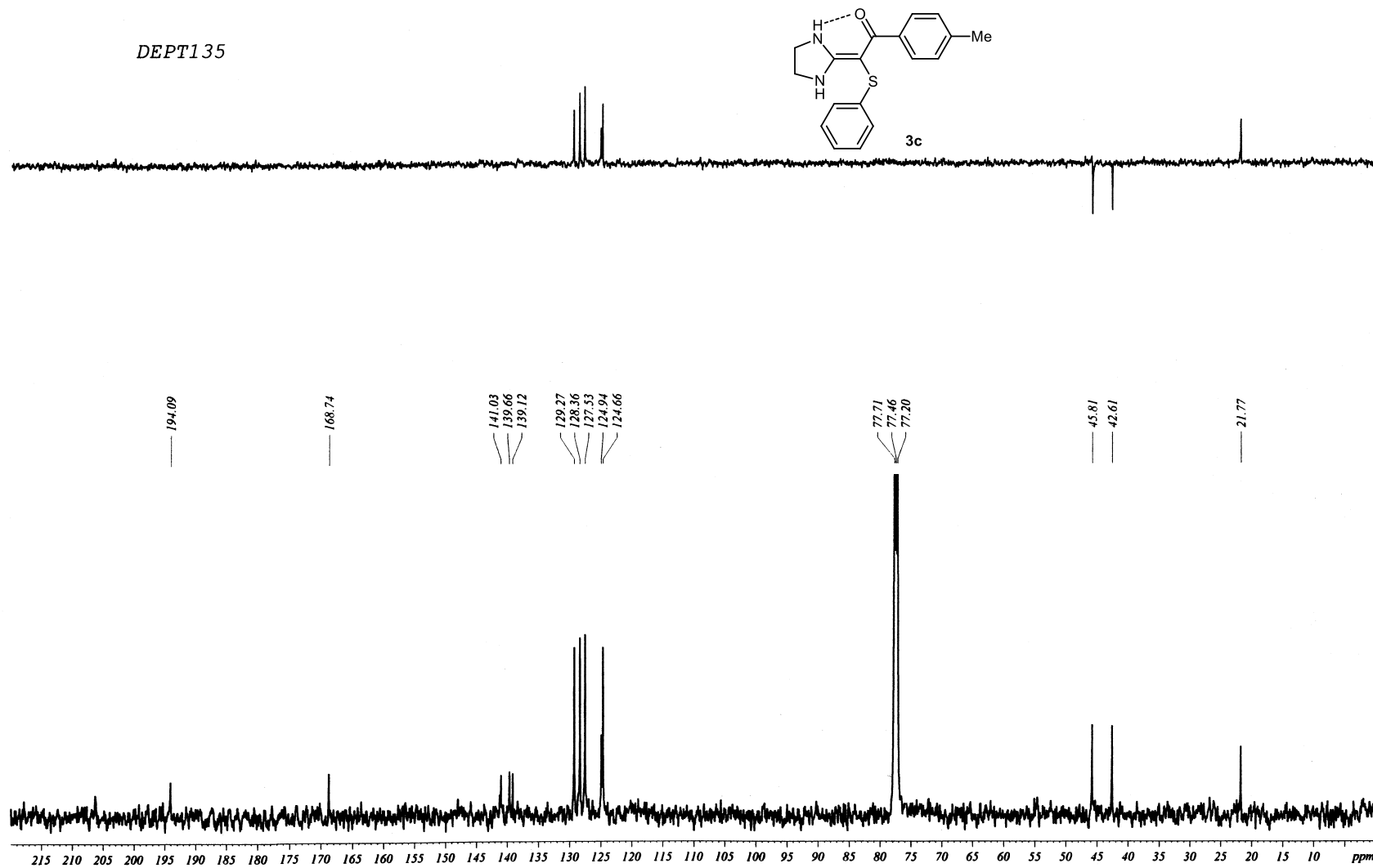


Figure S6 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound 3c

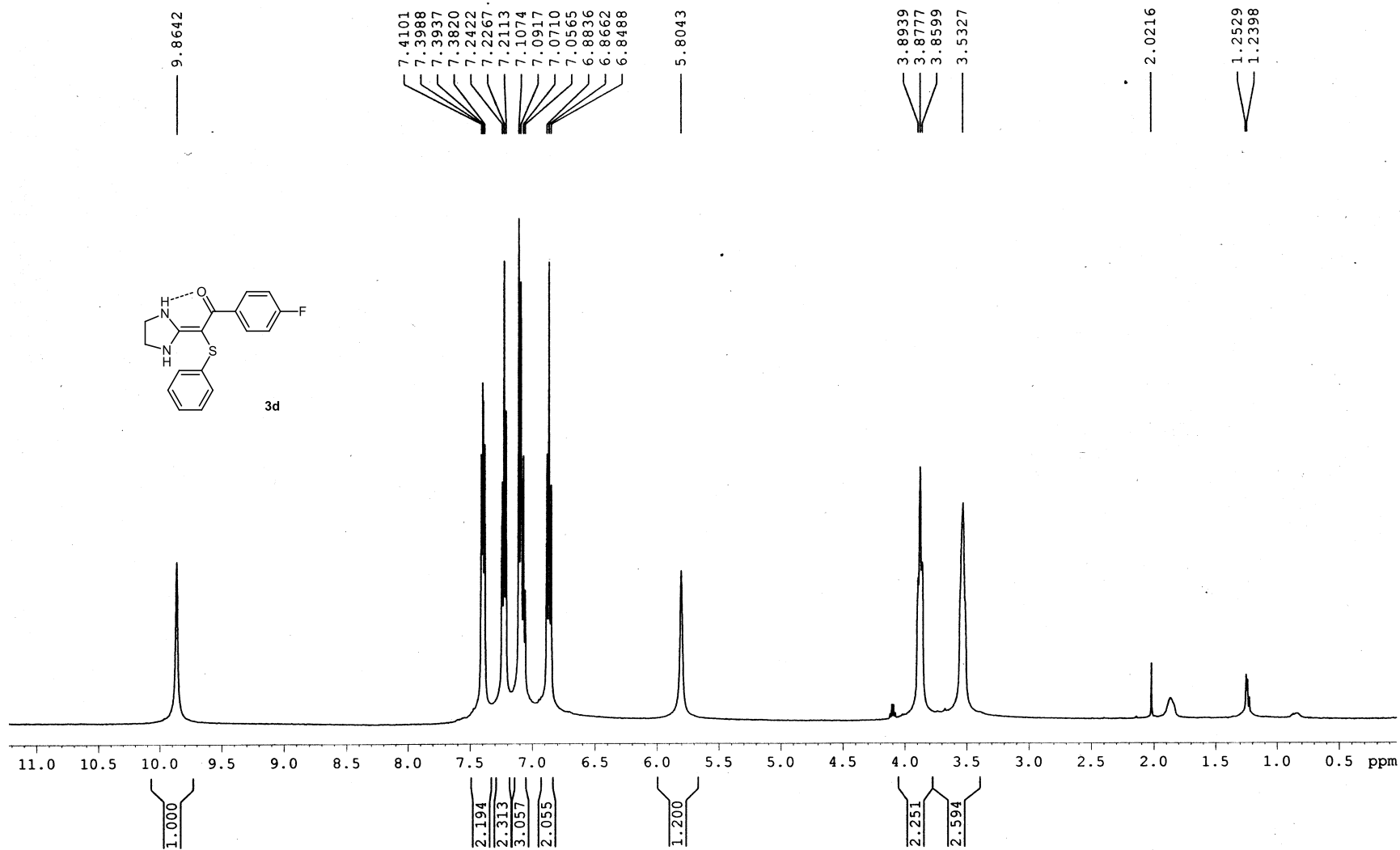


Figure S7 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3d**

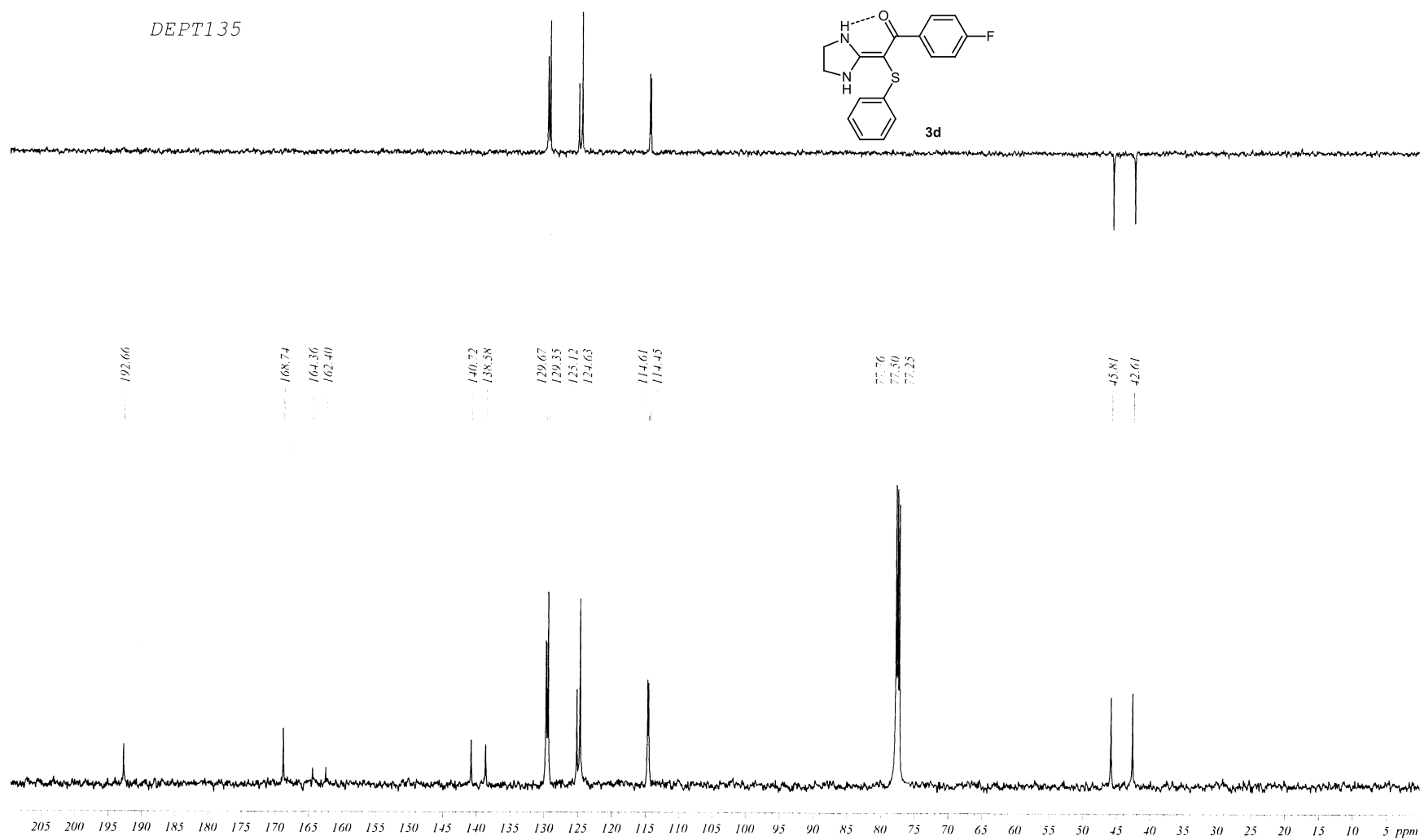


Figure S8 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3d**

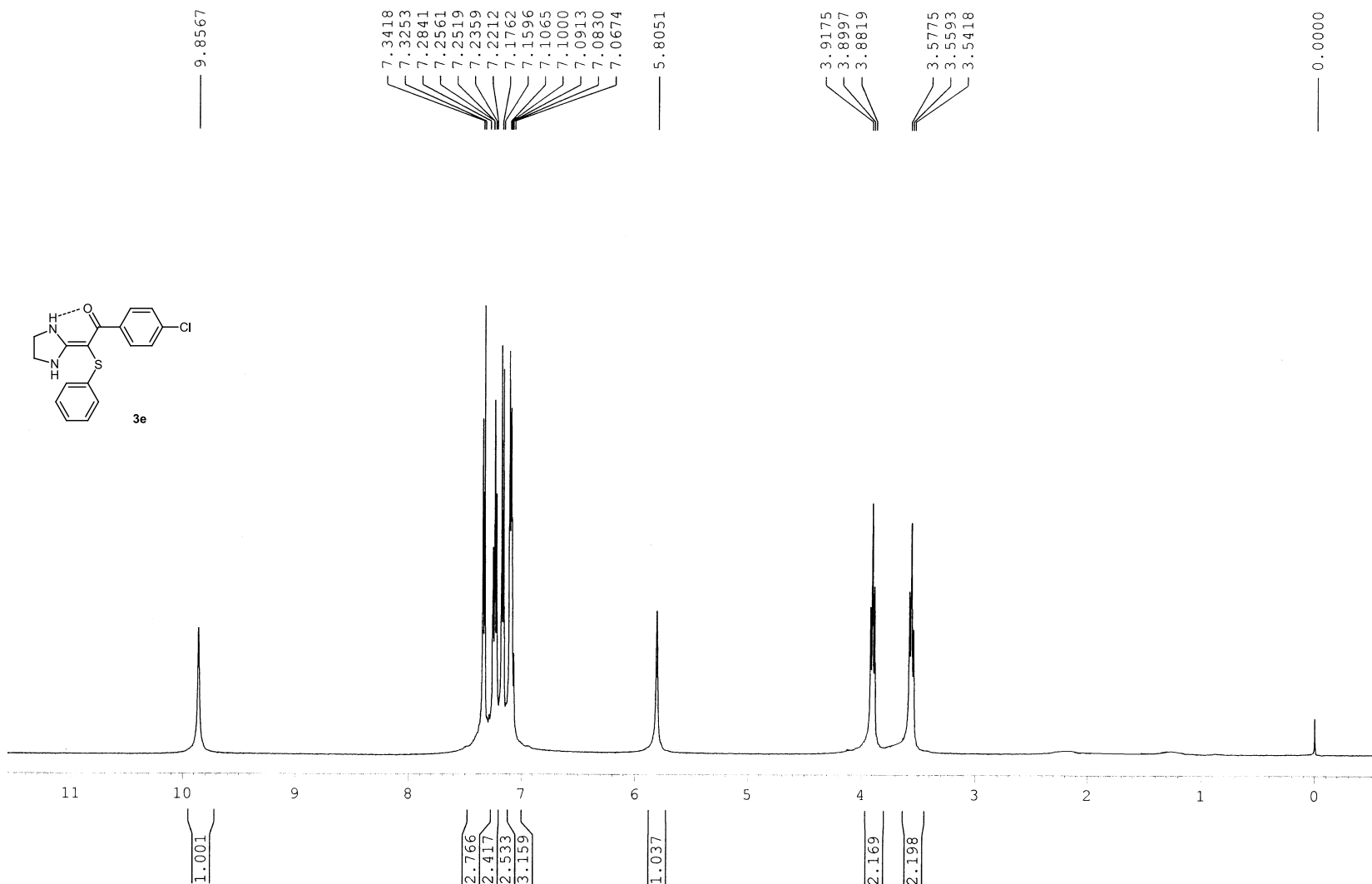


Figure S9 $^1\text{H NMR}$ spectrum (500 MHz, CDCl_3) of compound **3e**

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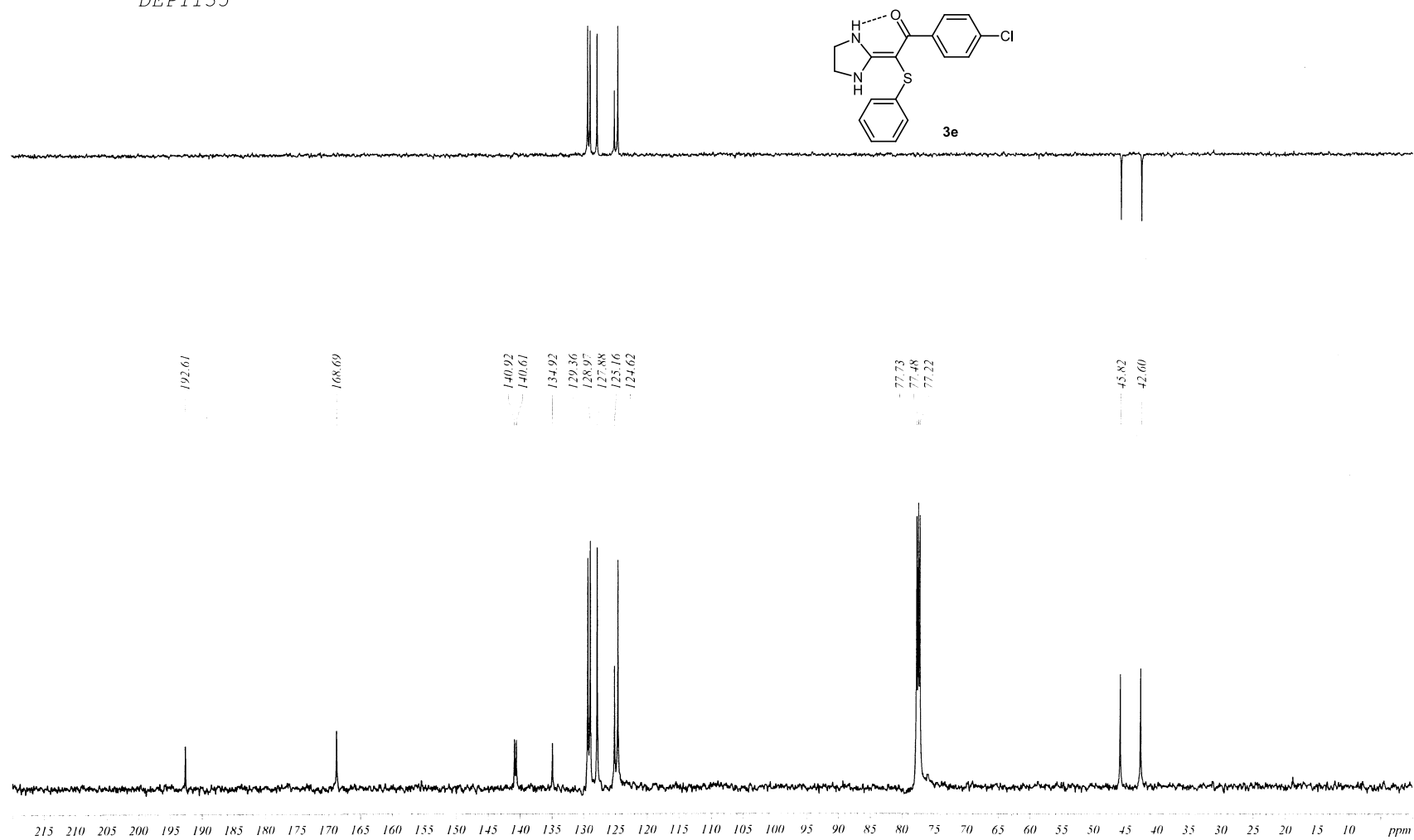


Figure S10 ¹³C NMR spectrum (125 MHz, CDCl₃) of compound **3e**

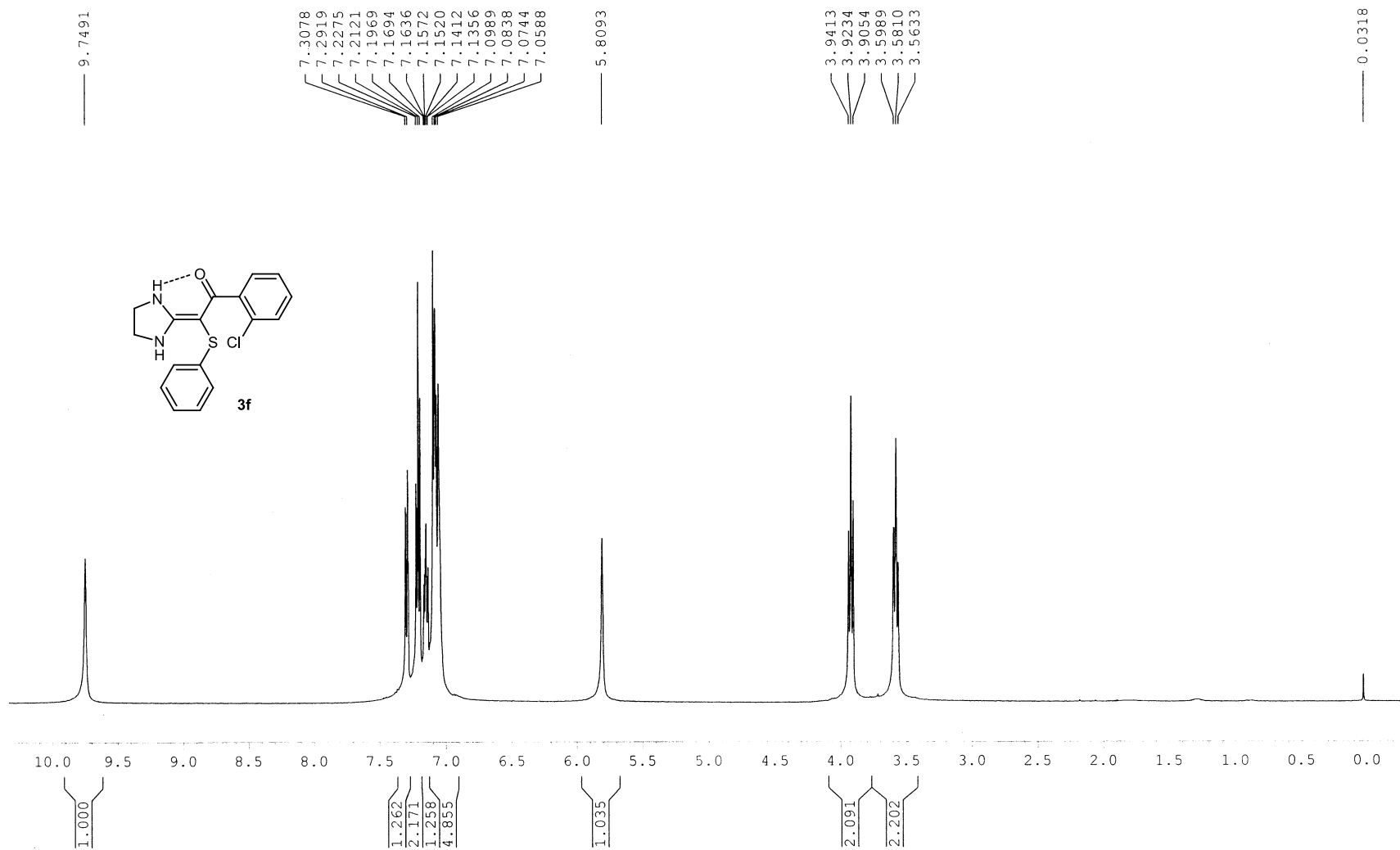


Figure S11 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3f**

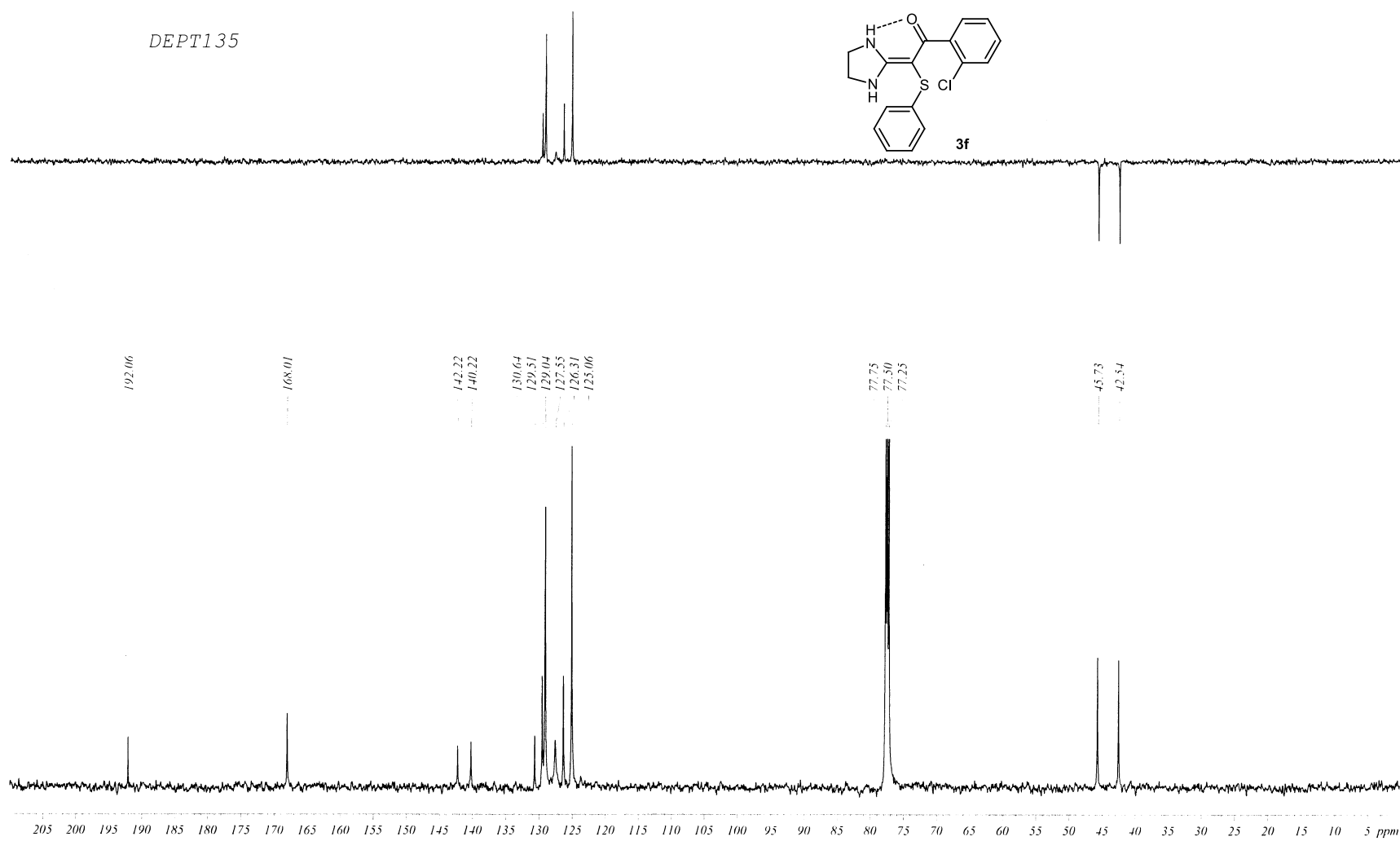


Figure S12 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3f**

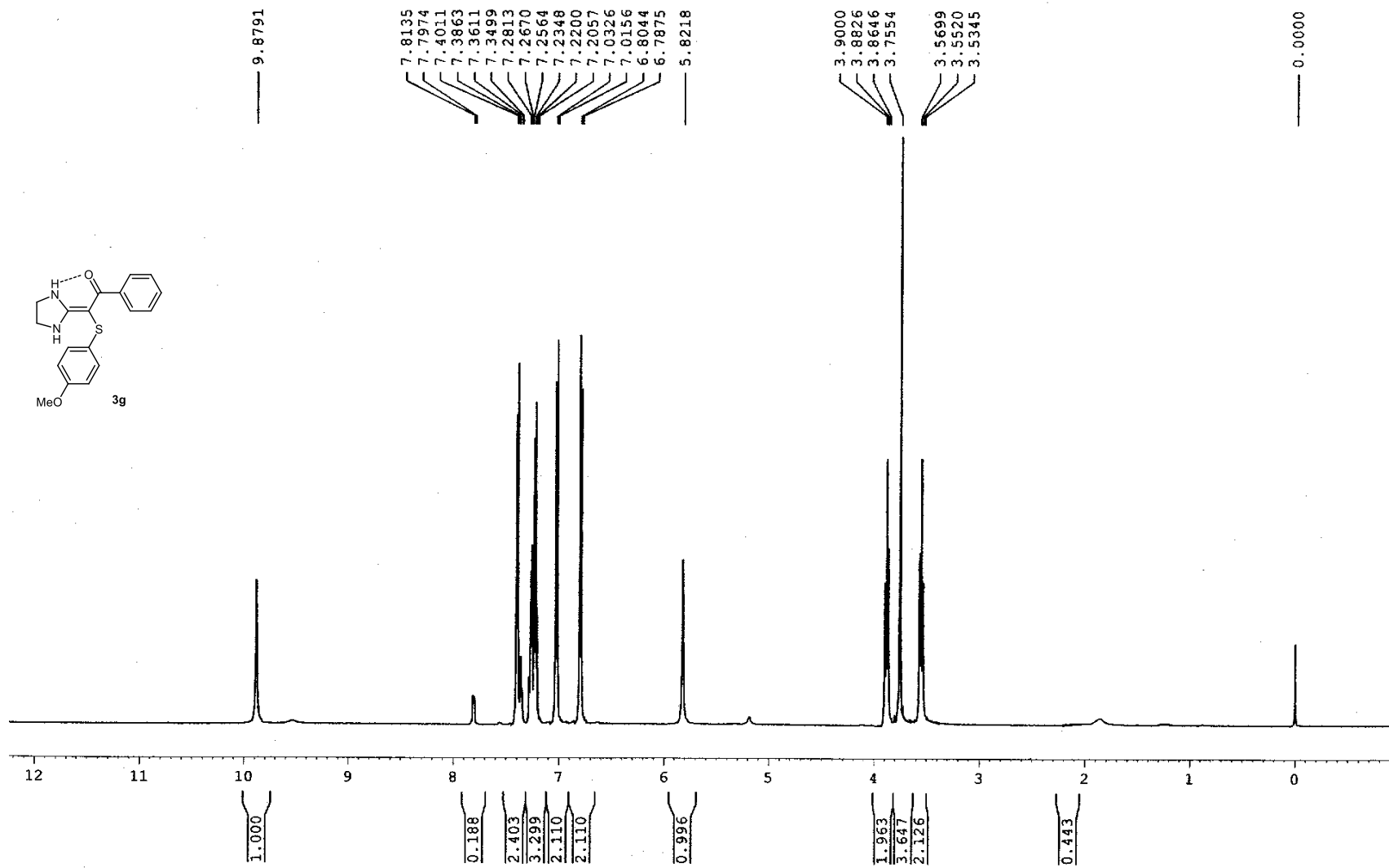


Figure S13 ¹H NMR spectrum (500 MHz, CDCl₃) of compound **3g**

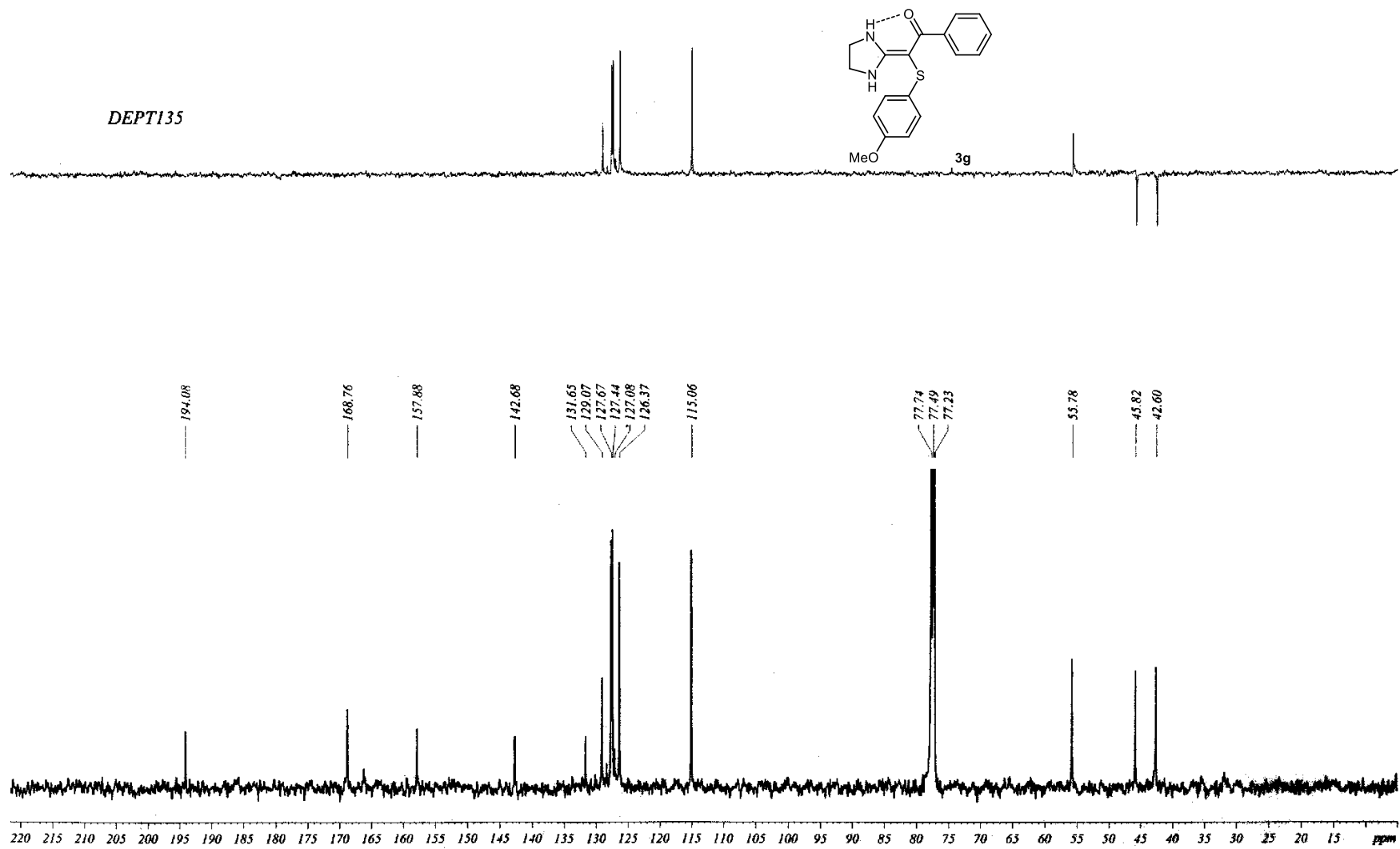


Figure S14 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3g**

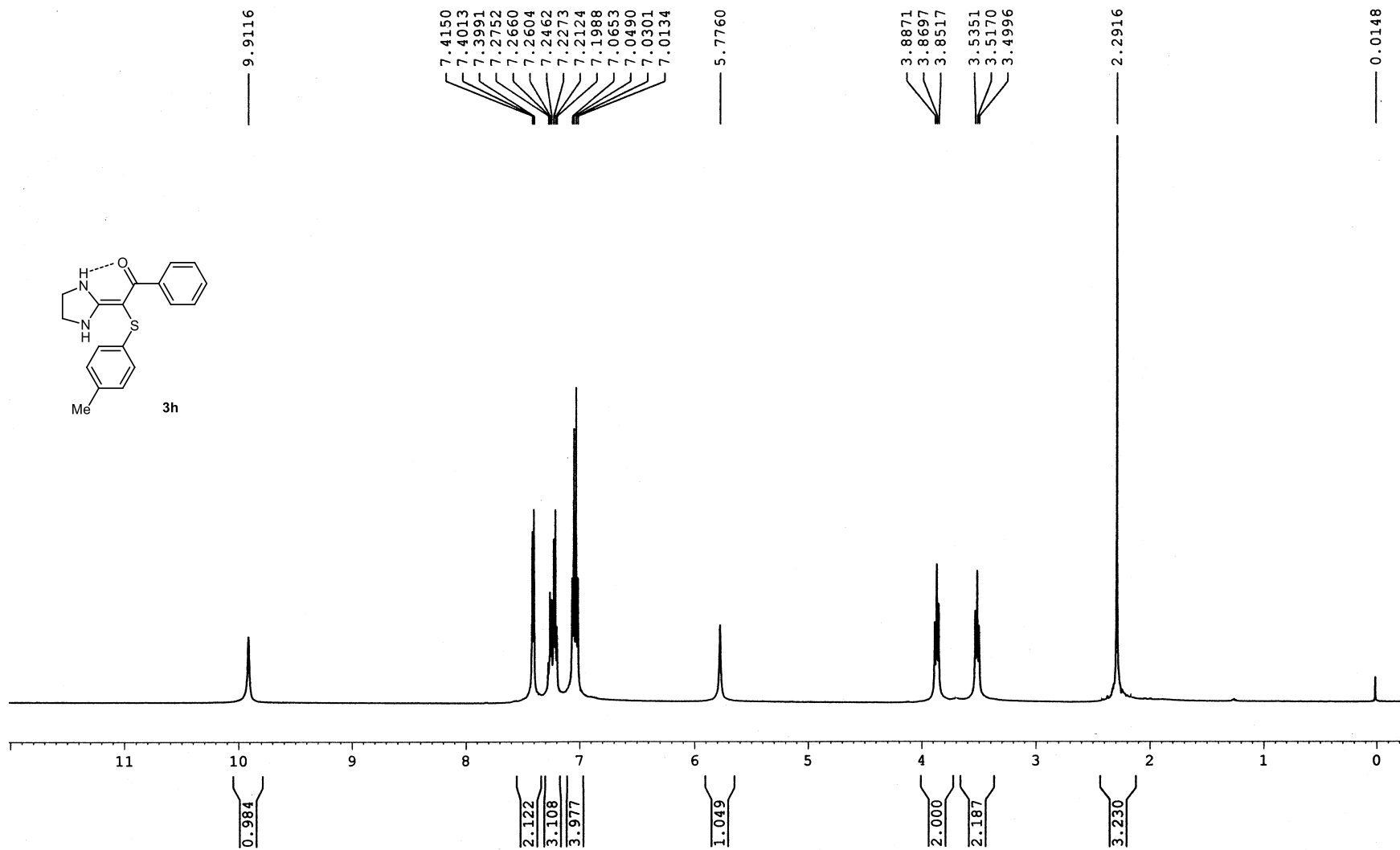


Figure S15 ¹H NMR spectrum (500 MHz, CDCl₃) of compound **3h**

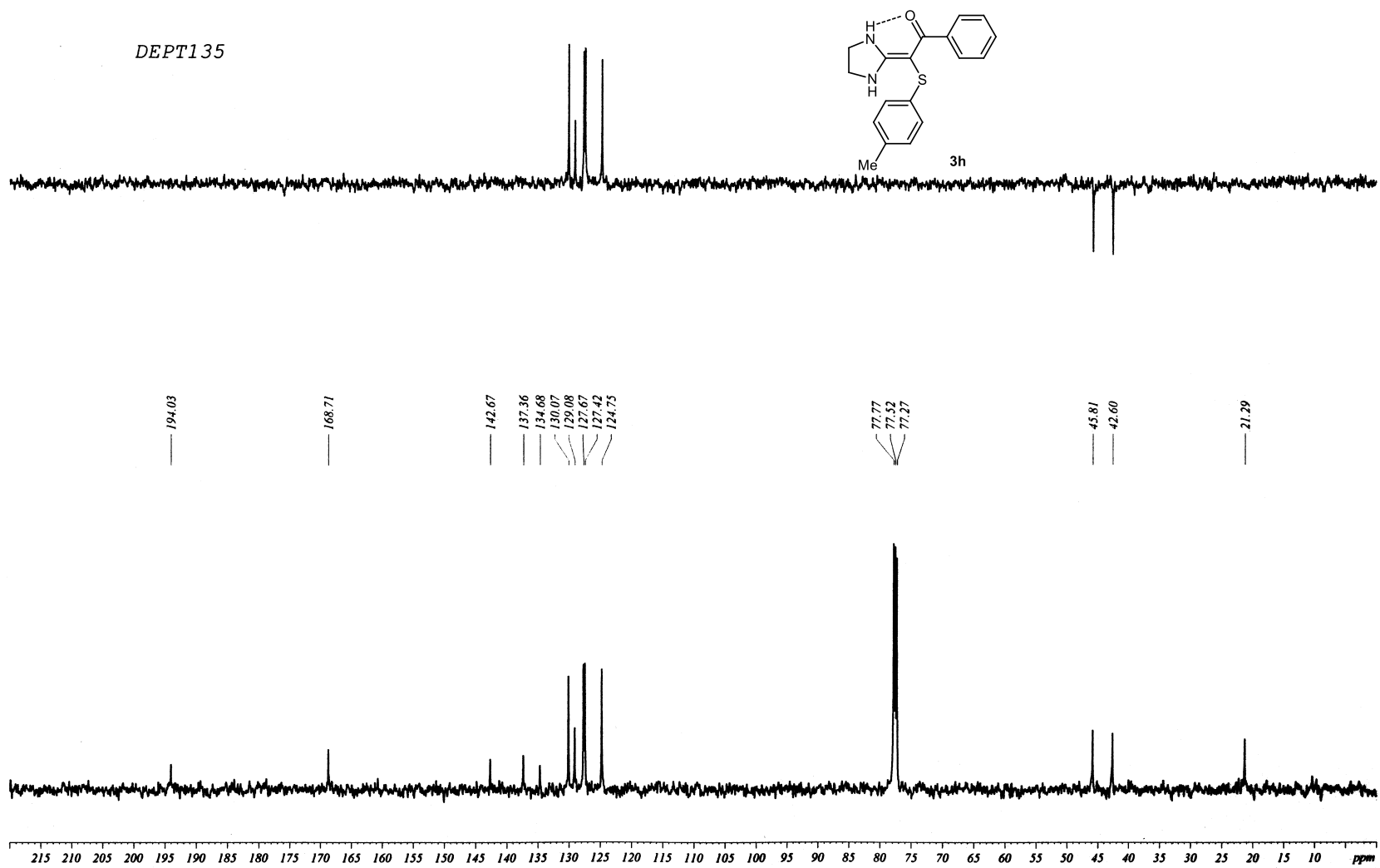


Figure S16 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3h**

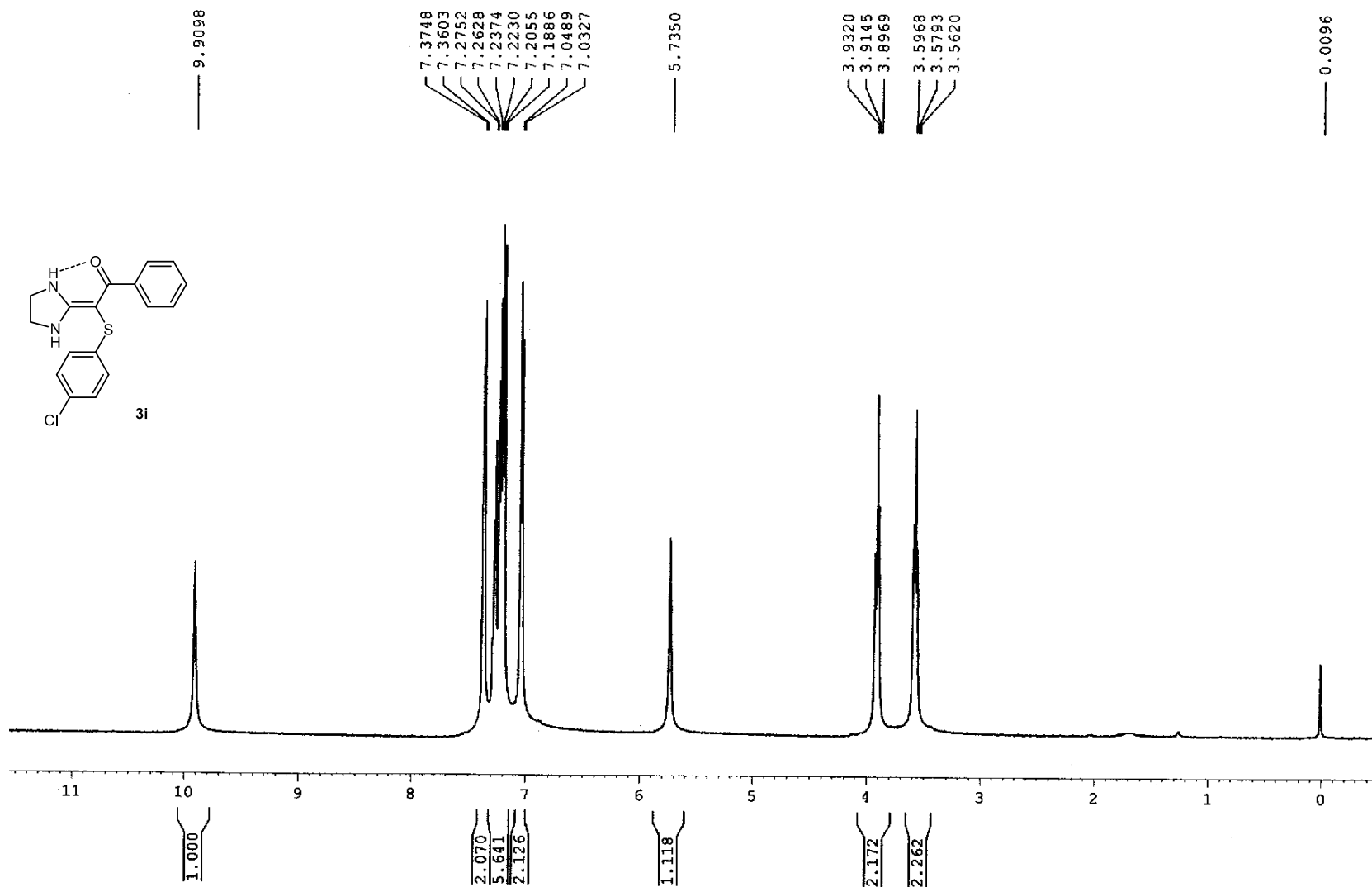


Figure S17 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3i**

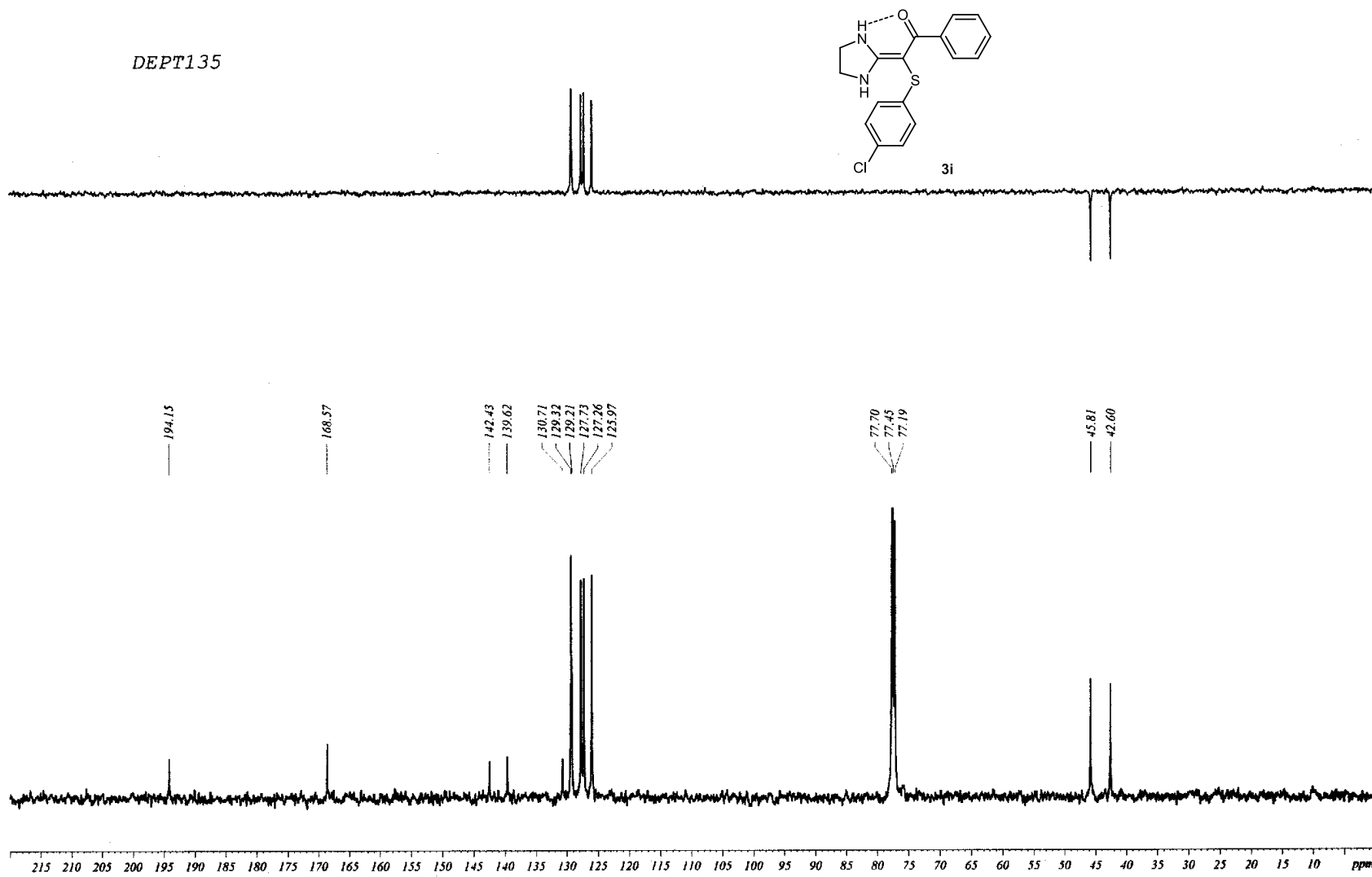


Figure S18 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3i**

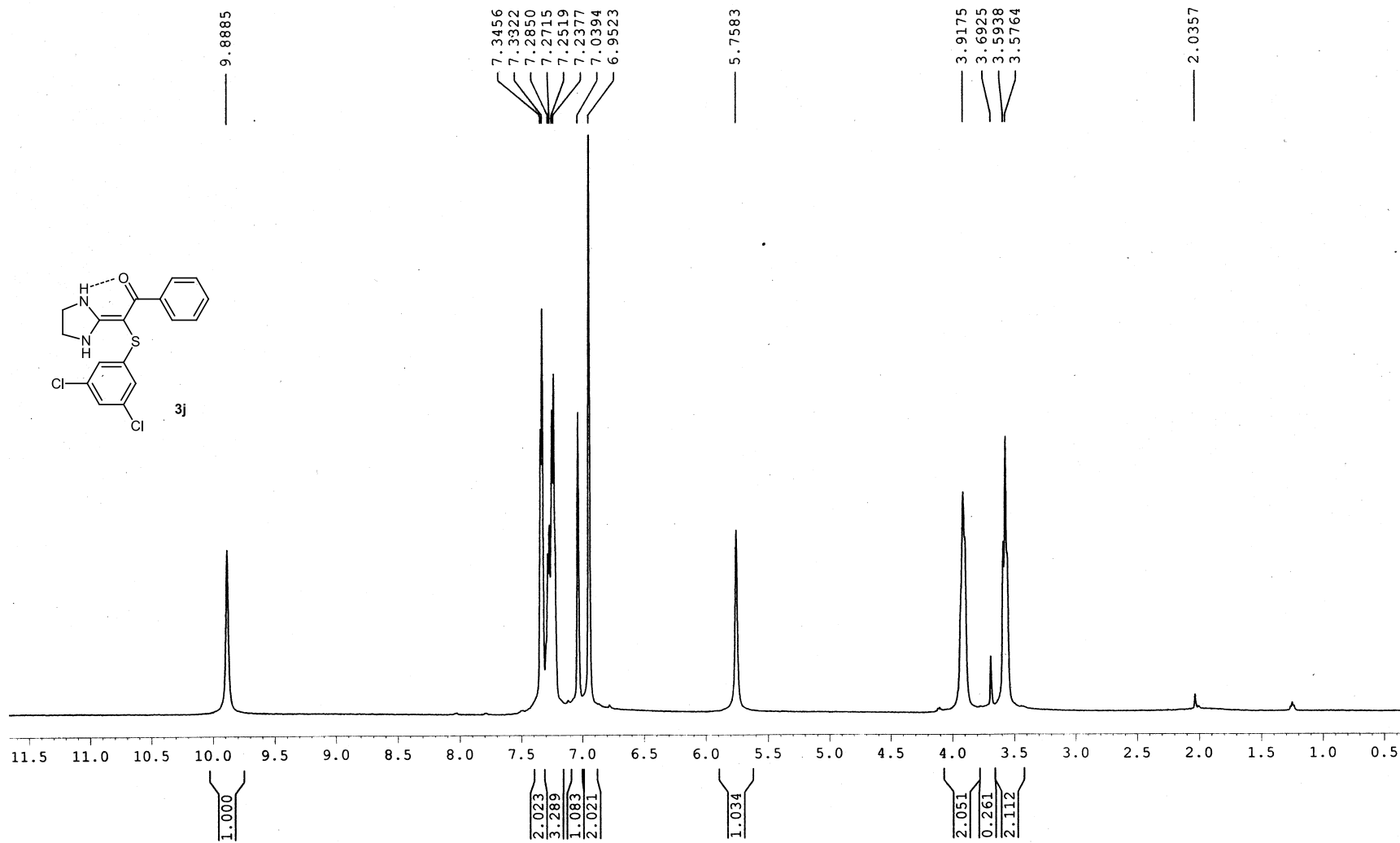


Figure S19 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3j**

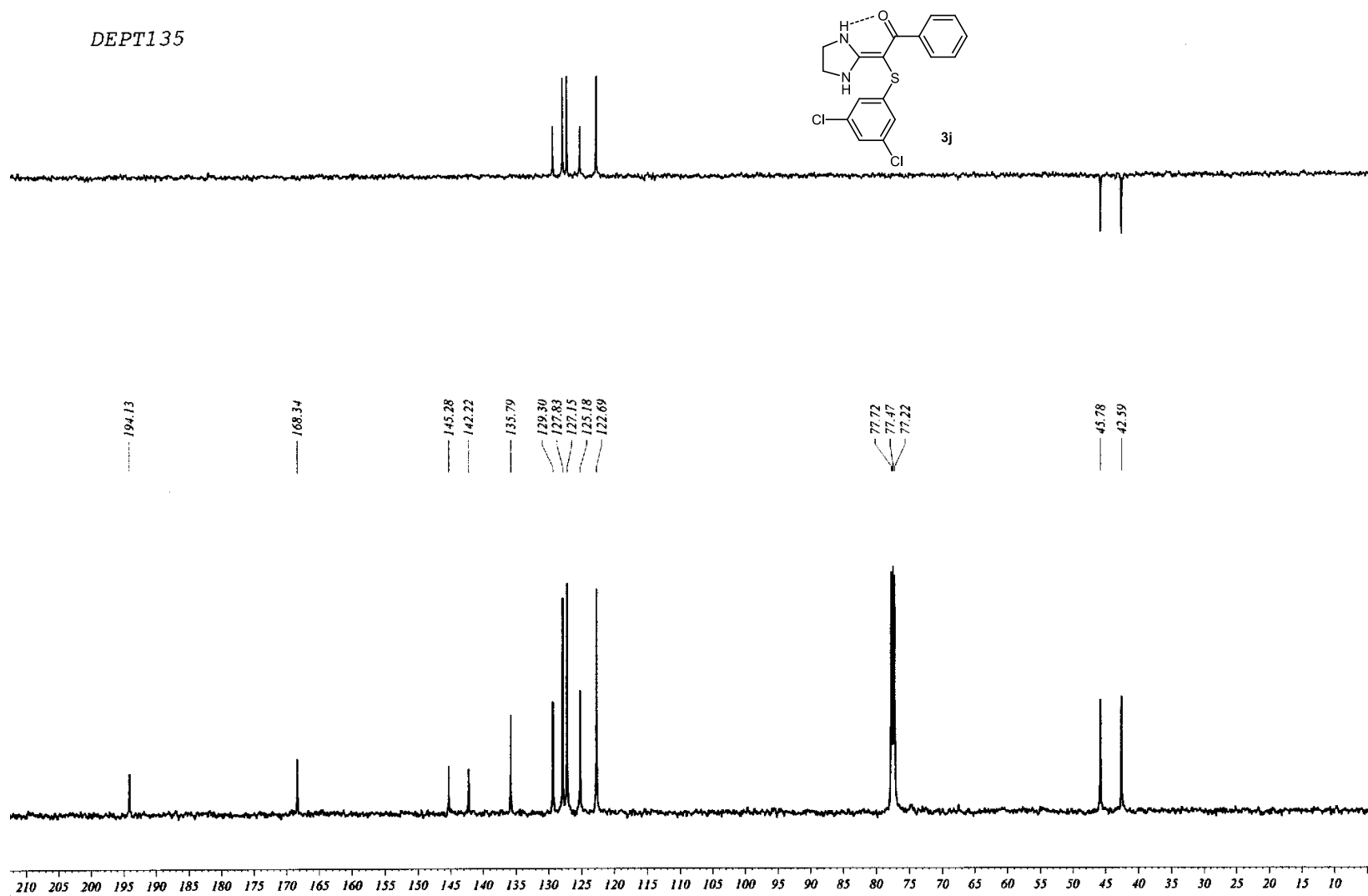


Figure S20 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3j**

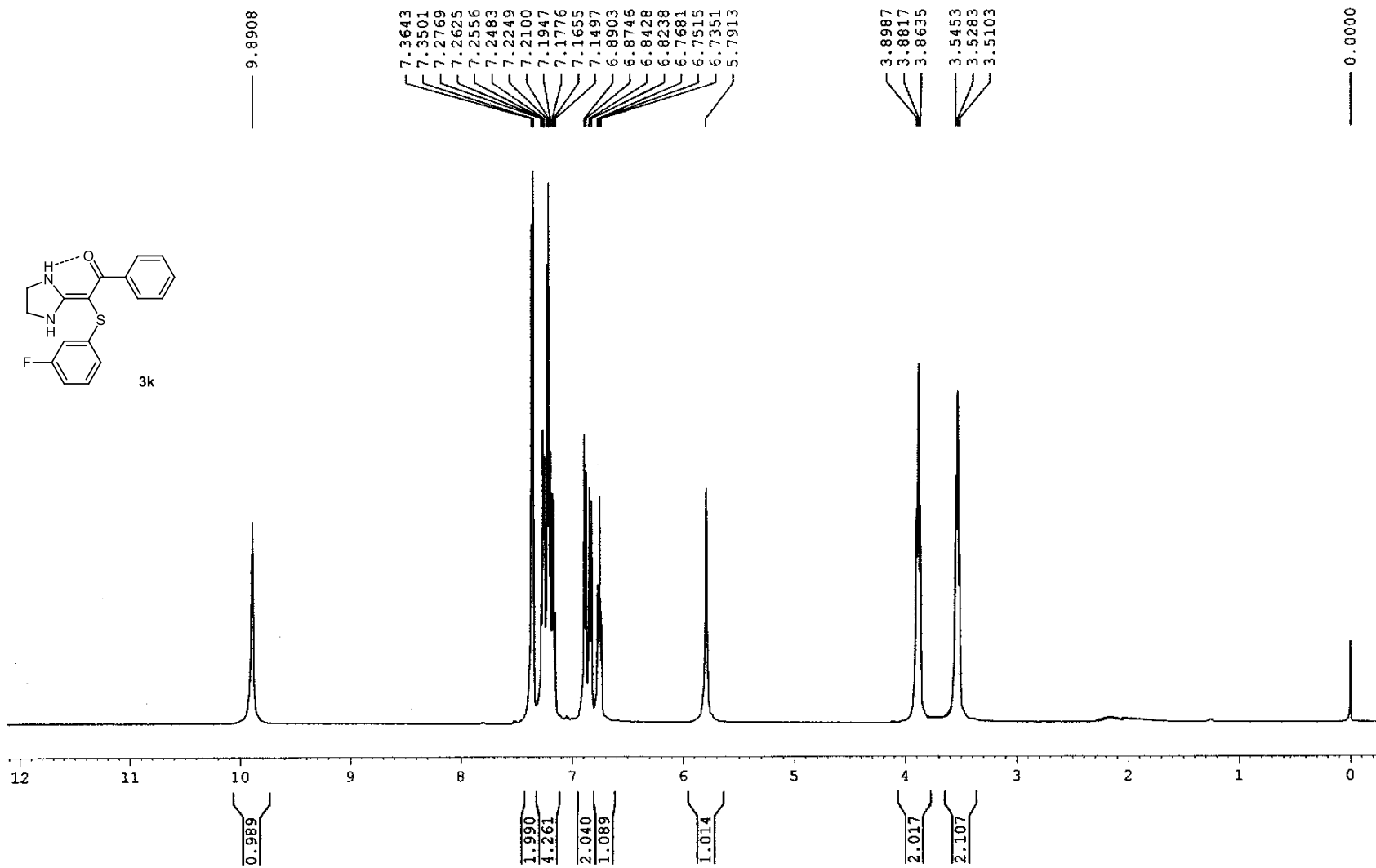


Figure S21 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3k**

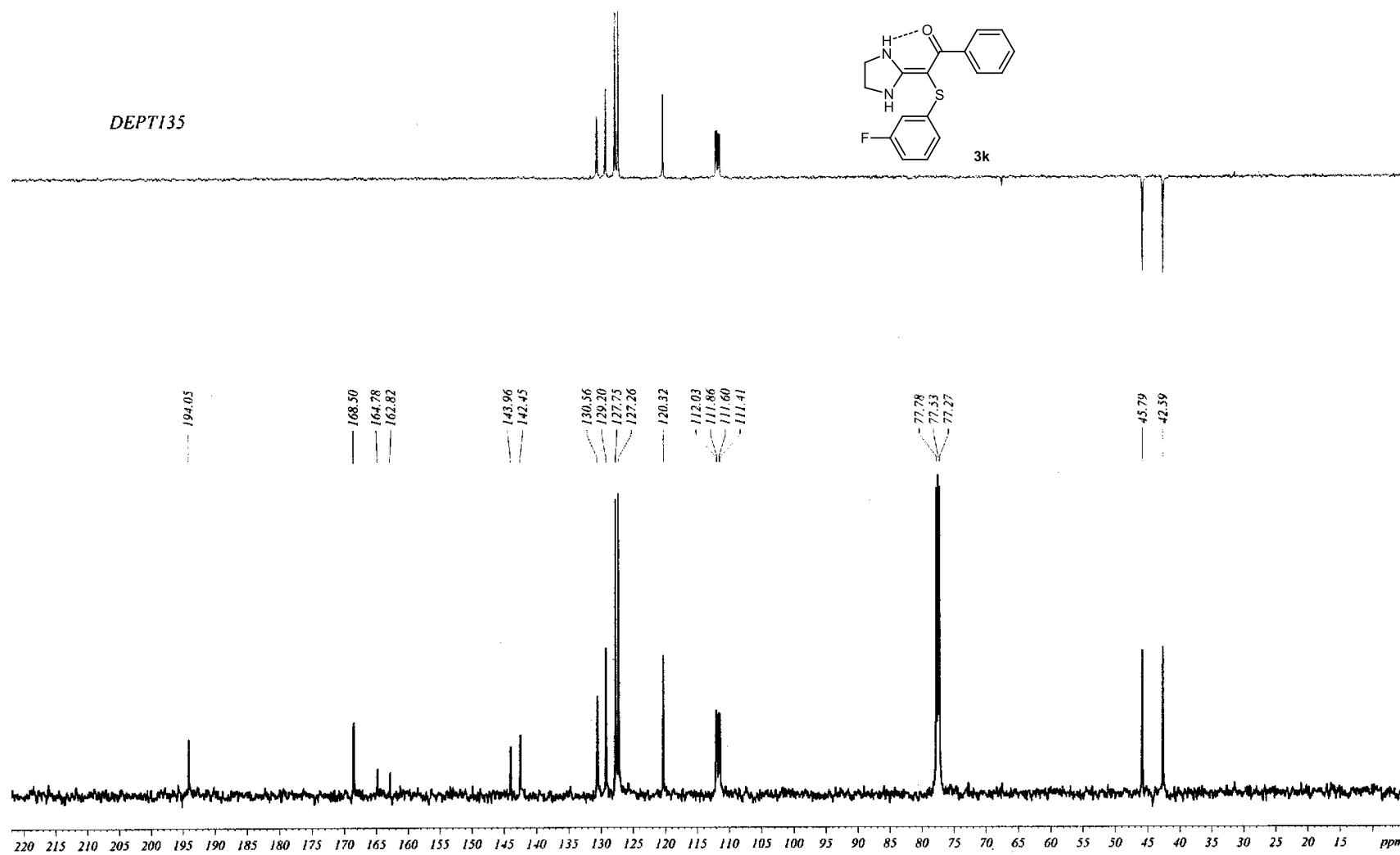


Figure S22 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3k**

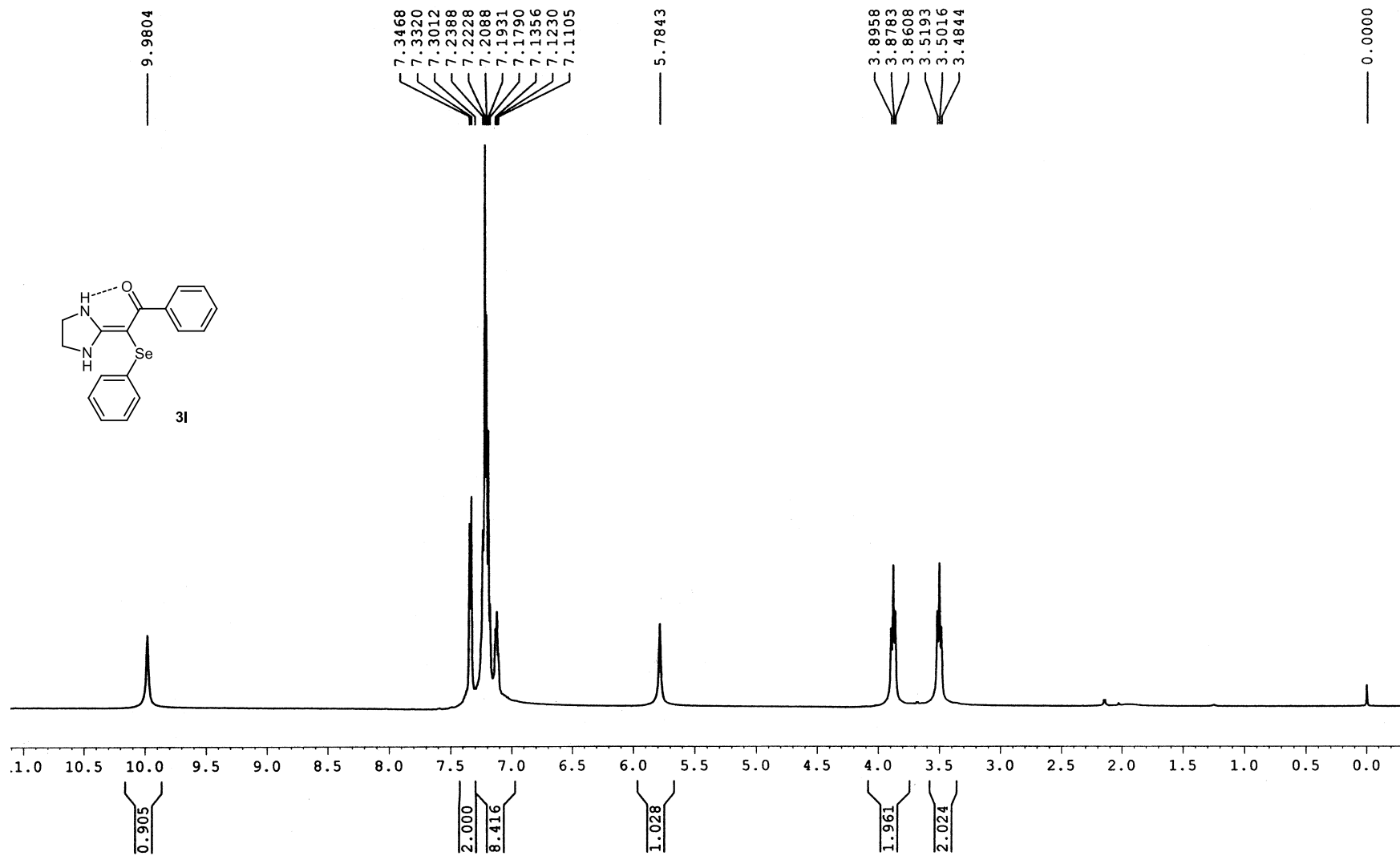


Figure S23 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **31**

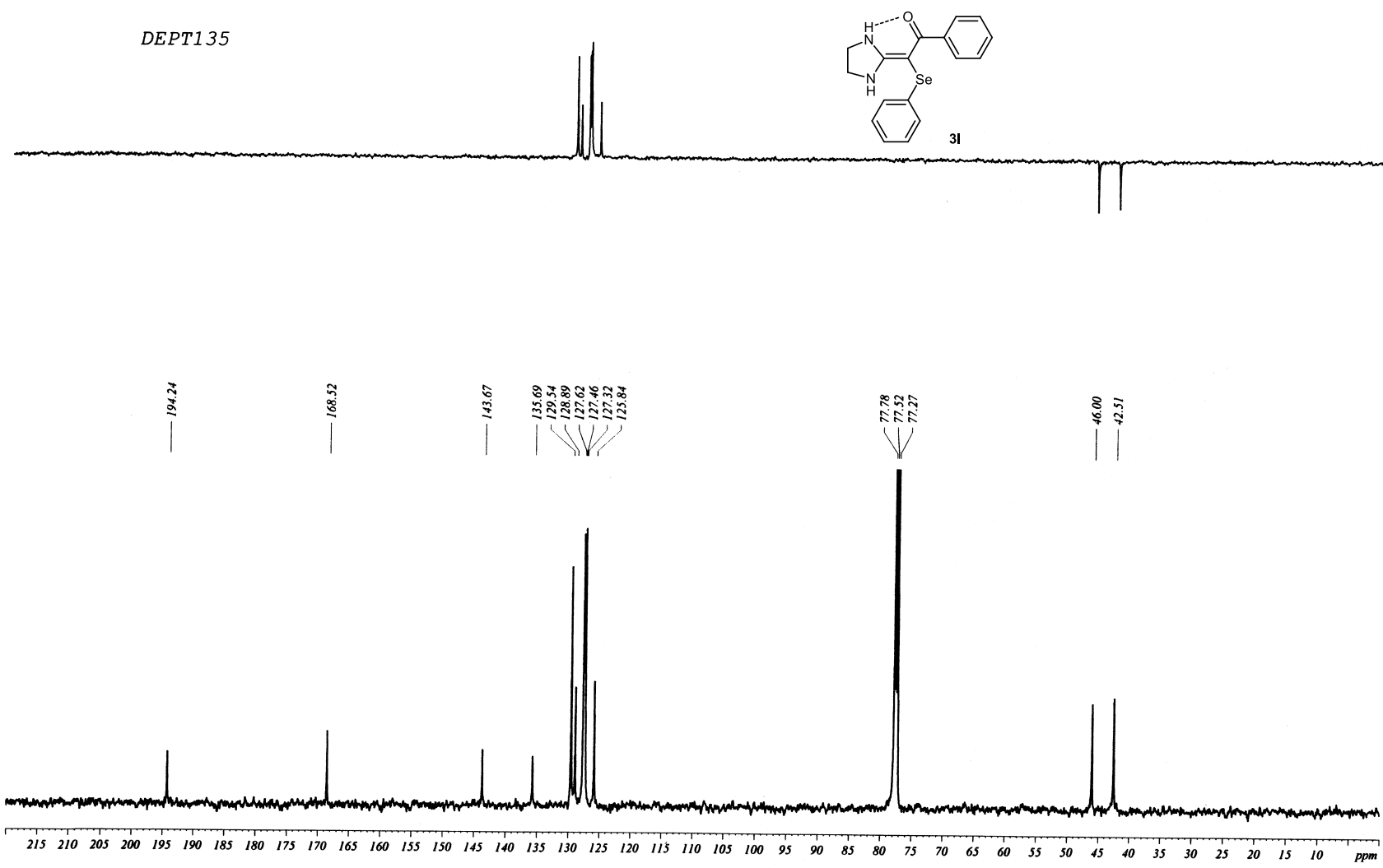


Figure S24 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **31**

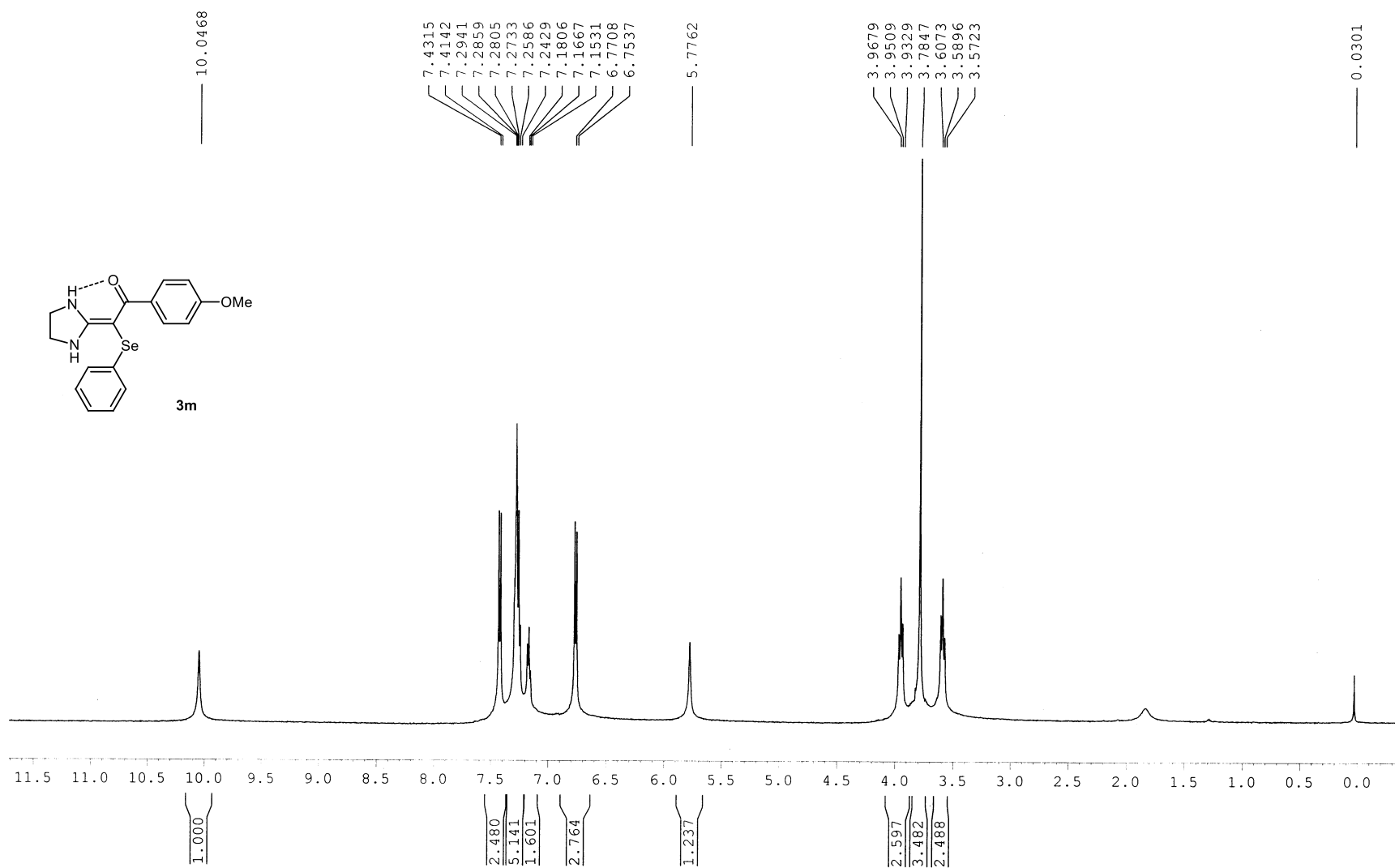


Figure S25 $^1\text{H NMR}$ spectrum (500 MHz, CDCl_3) of compound **3m**

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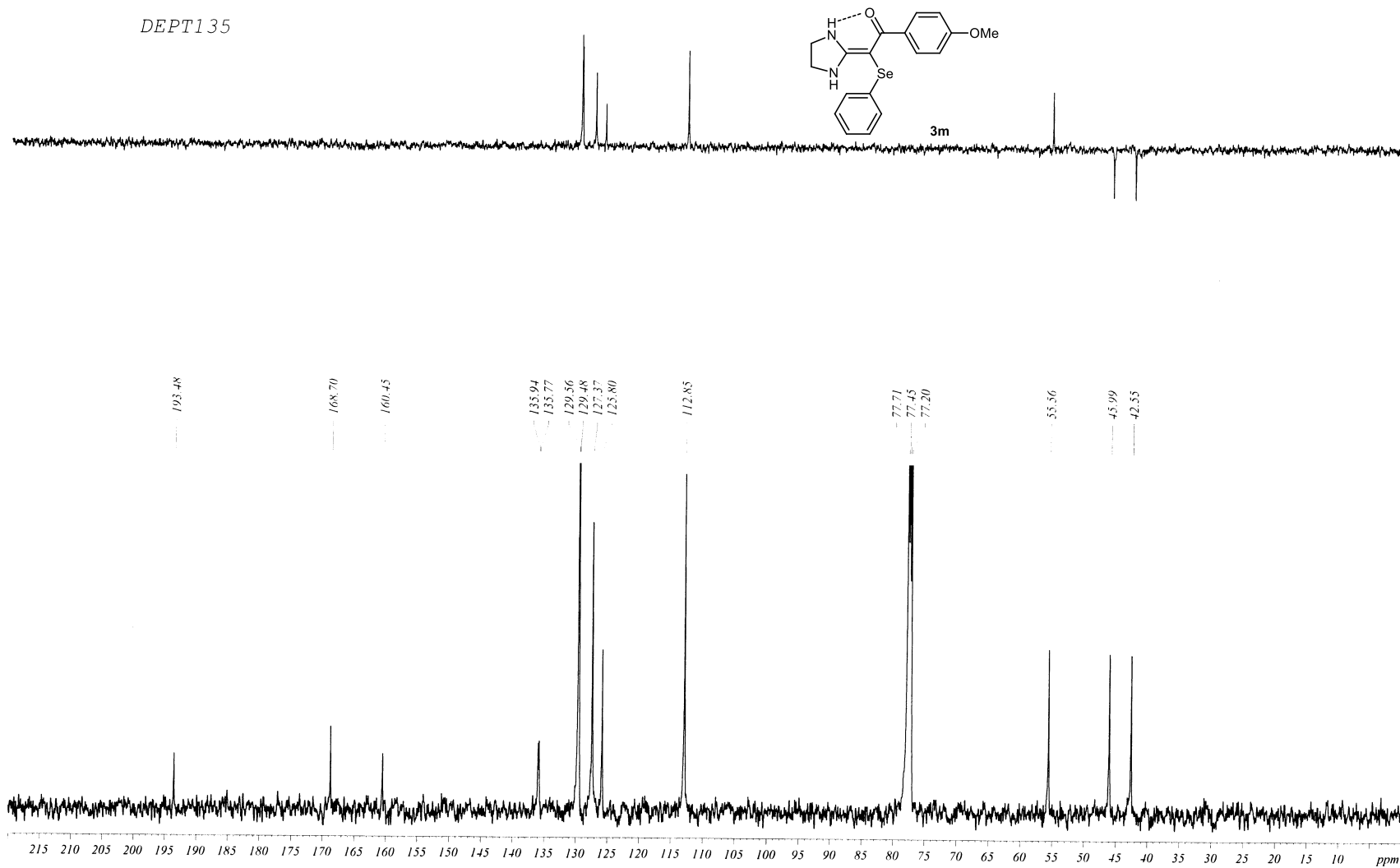


Figure S26 ¹³C NMR spectrum (125 MHz, CDCl₃) of compound **3m**

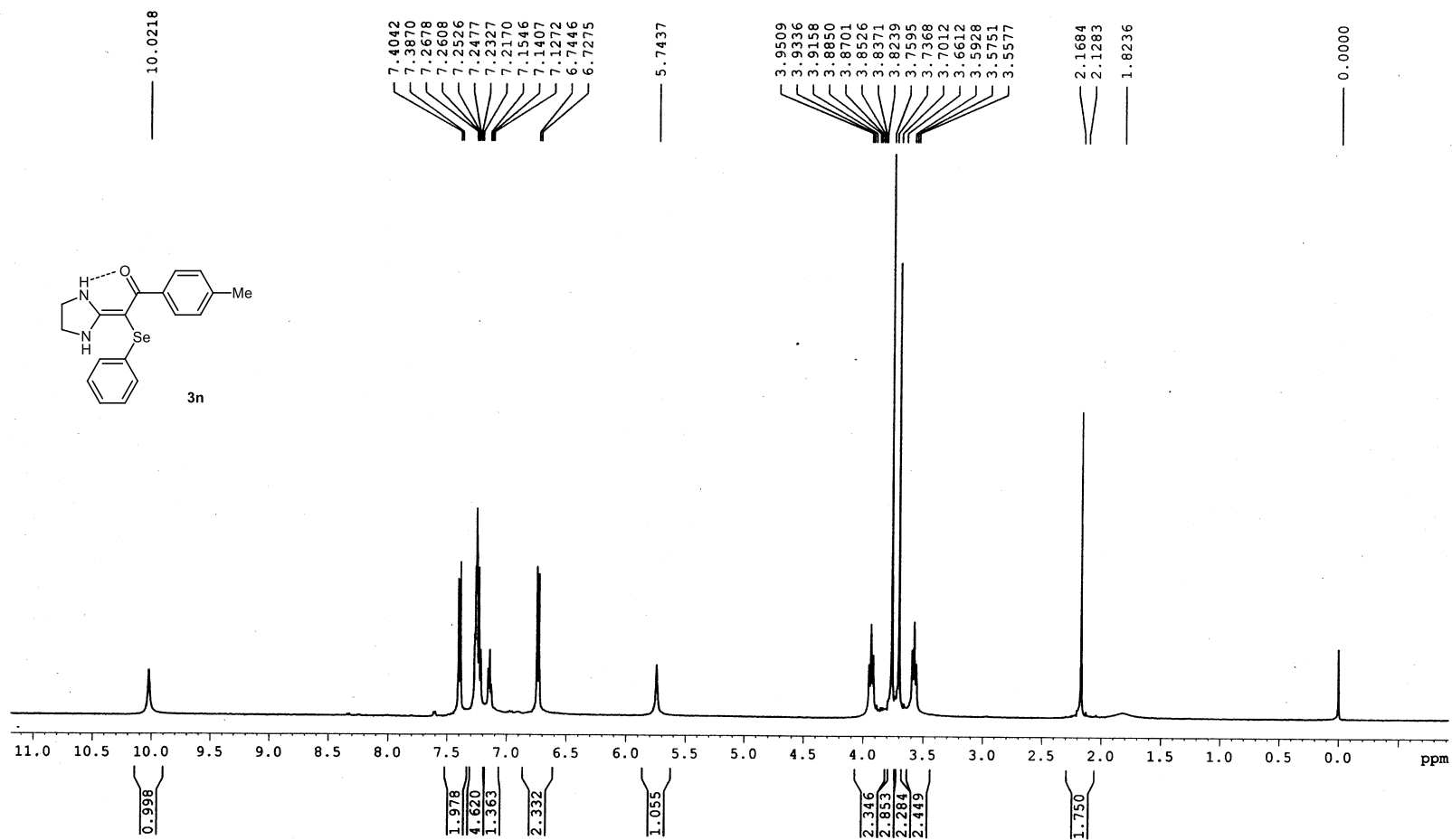
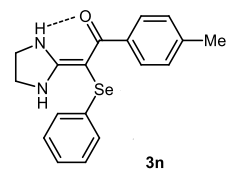


Figure S27 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3n**

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jiangxiuyang JXY-15 in CDCl3
13050604

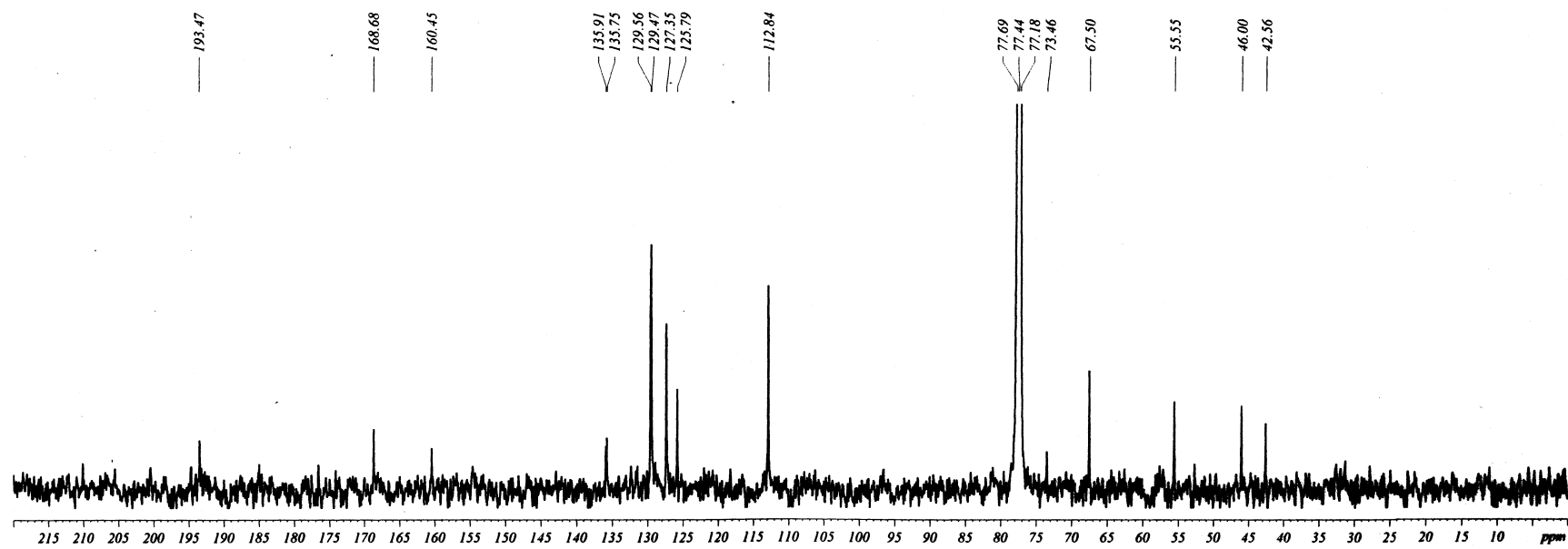


Figure S28 ¹³C NMR spectrum (125 MHz, DMSO-d₆) of compound 3n

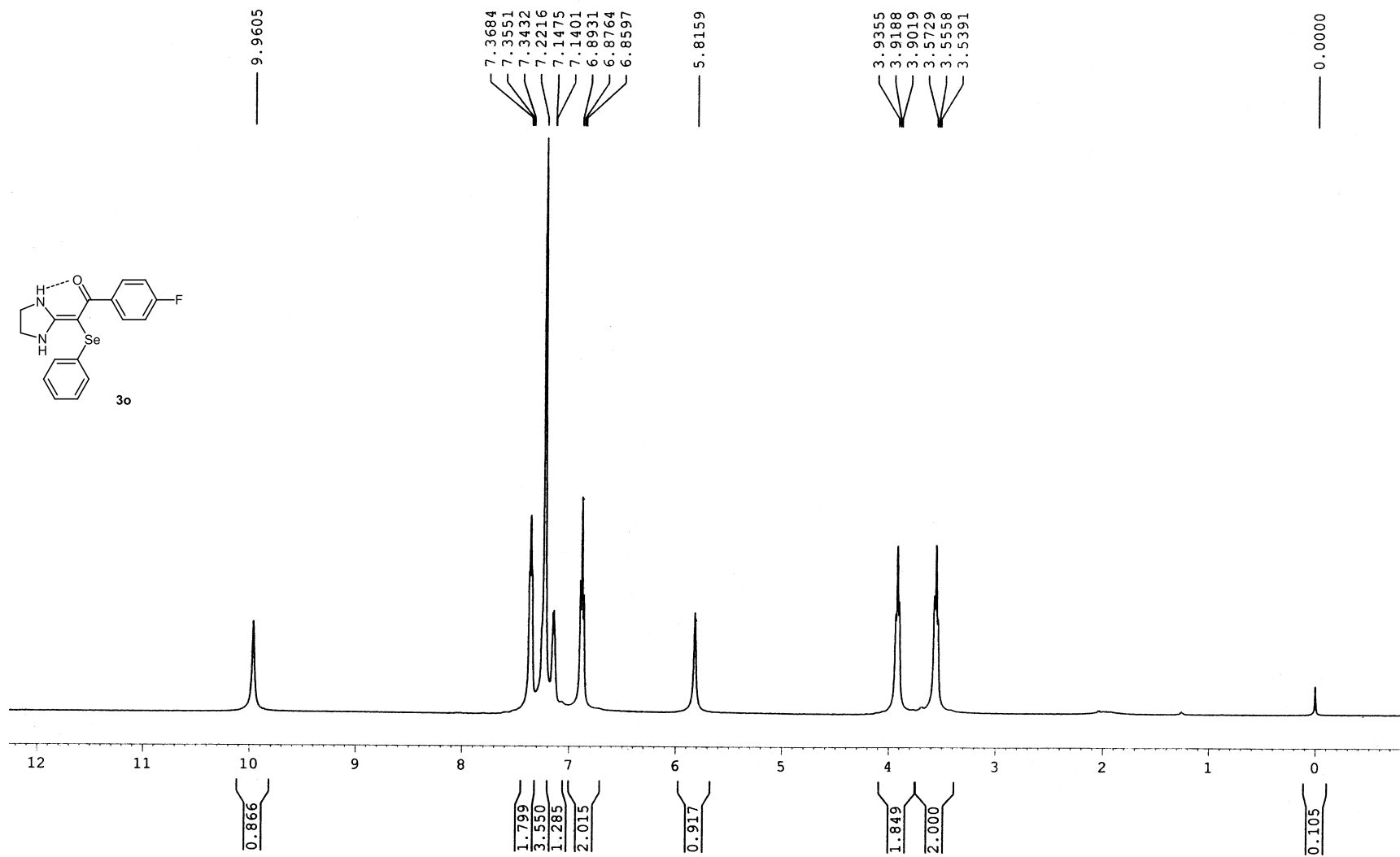


Figure S29 $^1\text{H NMR}$ spectrum (500 MHz, CDCl_3) of compound **3o**

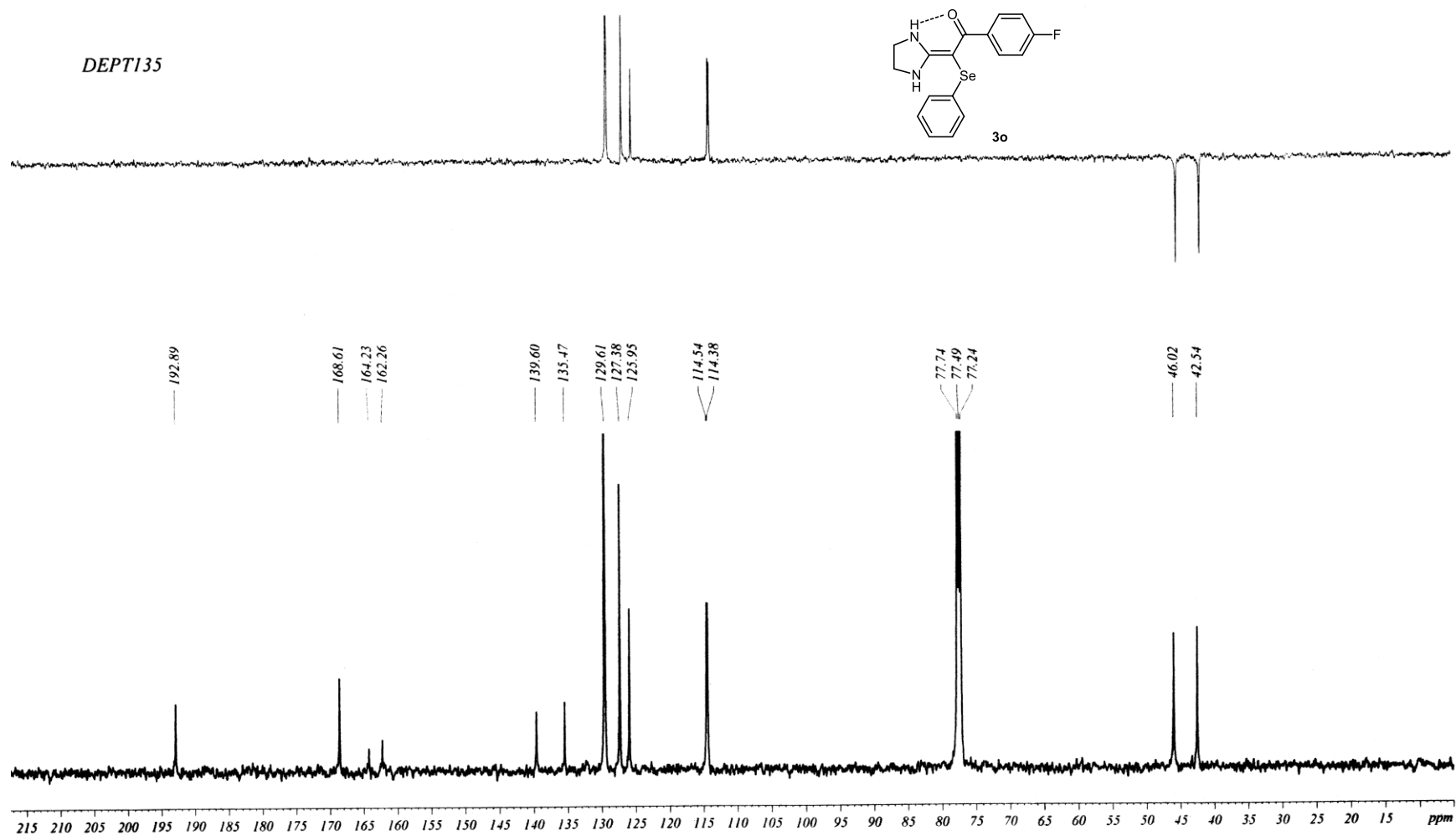


Figure S30 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3o**

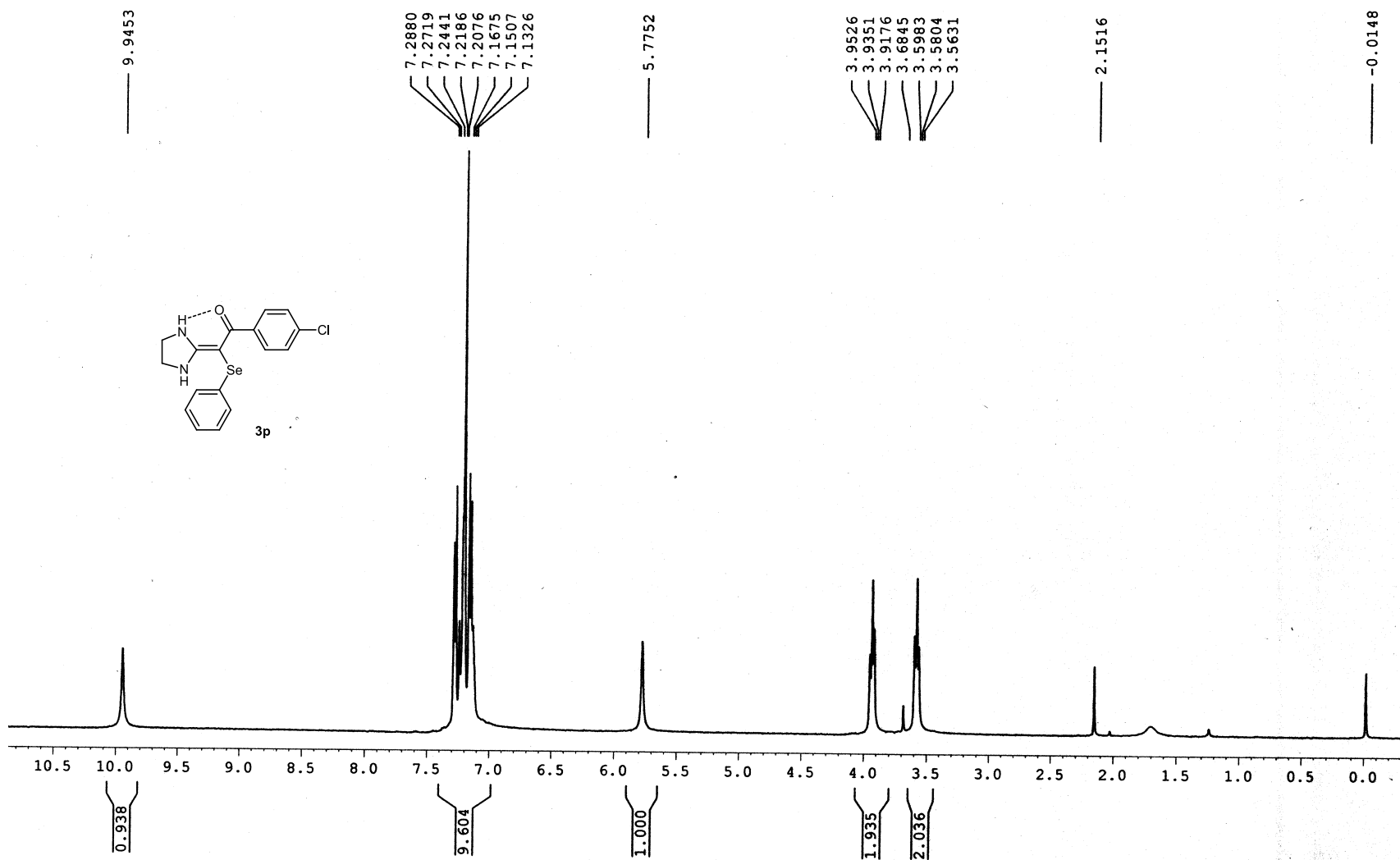


Figure S31 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **3p**

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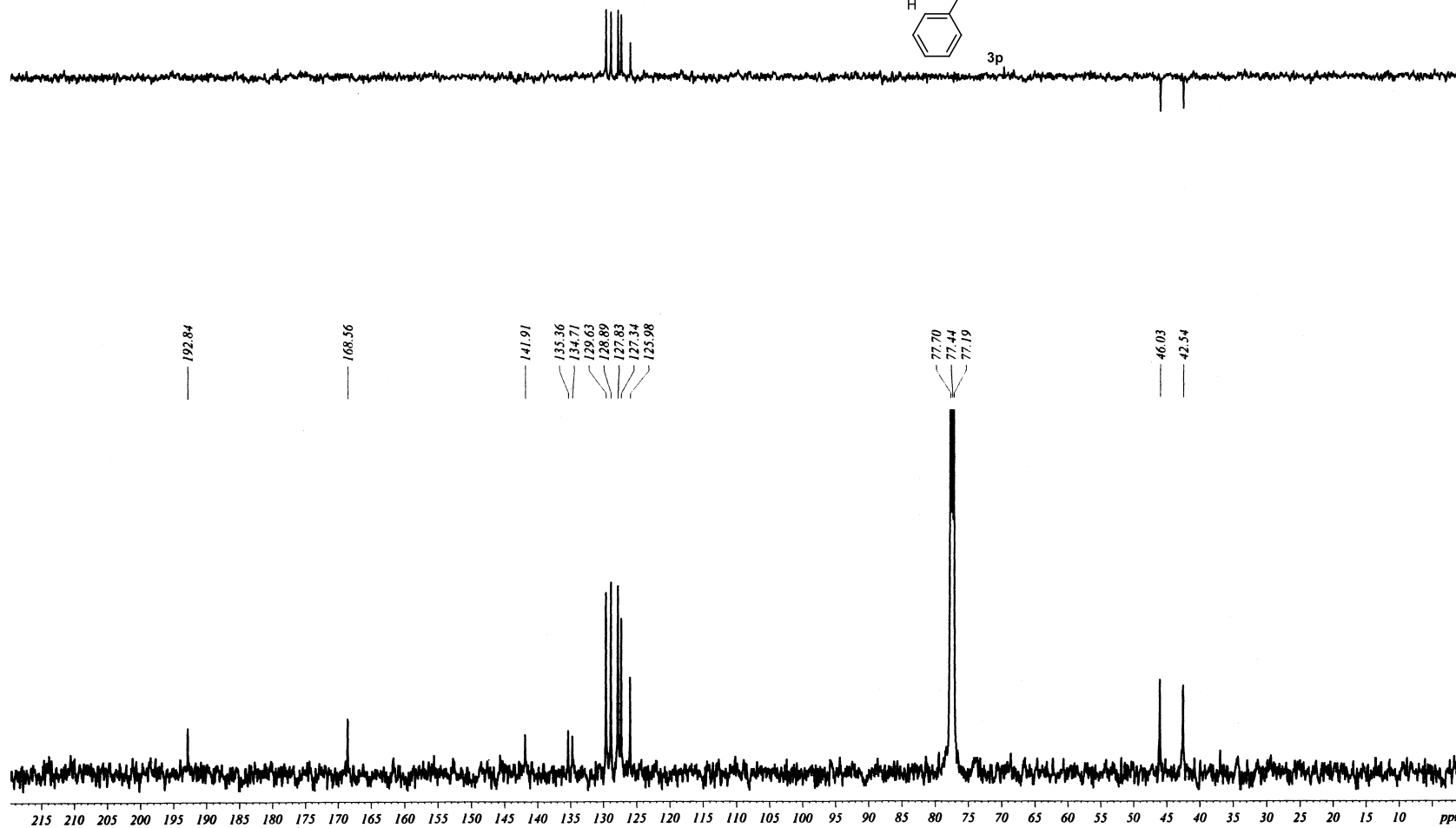
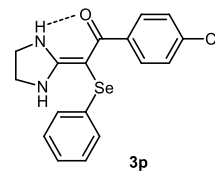


Figure S32 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3p**

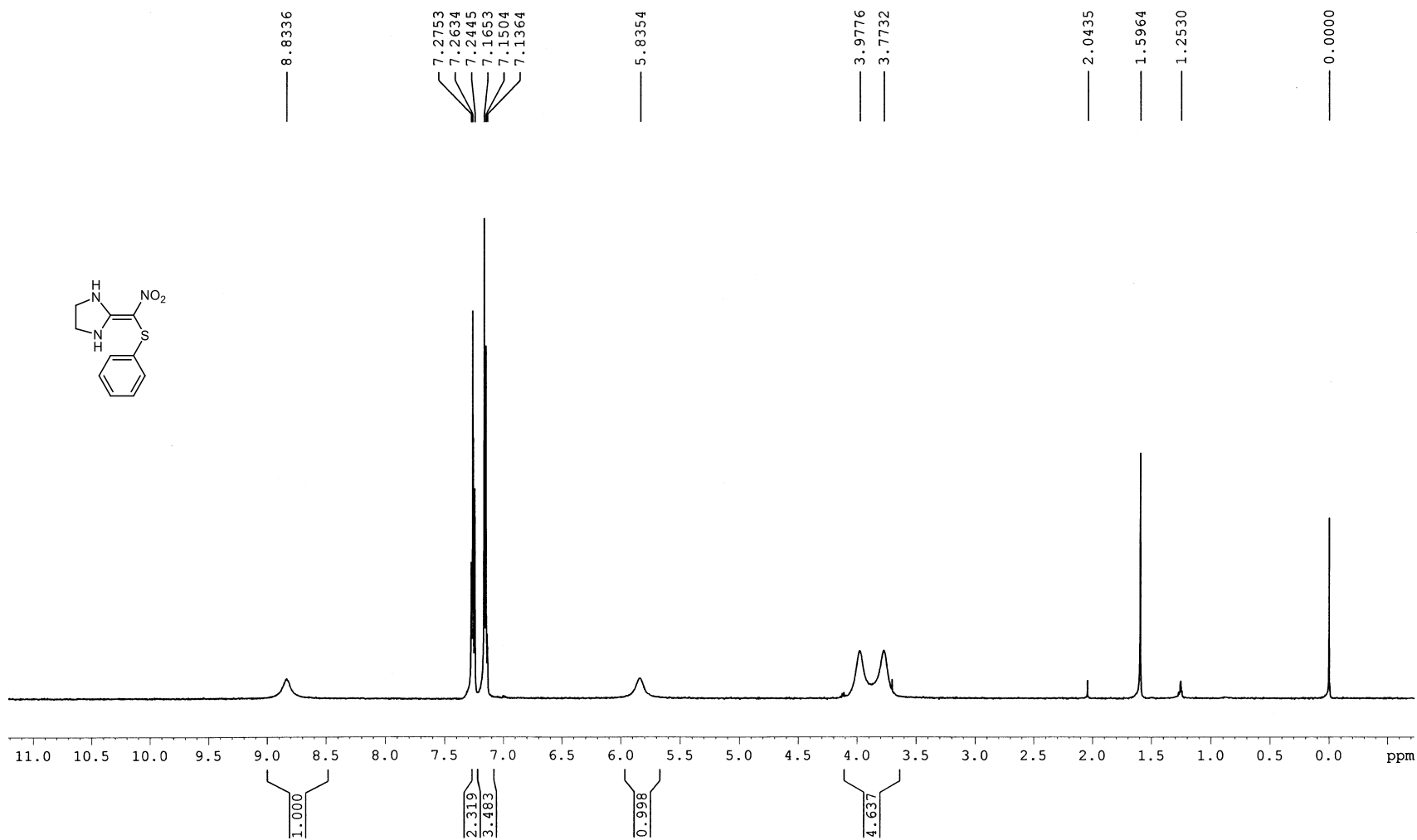


Figure S33 ¹H NMR spectrum (500 MHz, CDCl₃) of compound **3q**

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YUNNAN UNIVER. AV. DRX 500
jiangxiuyang JXY-N5 in CDCL3
14051301

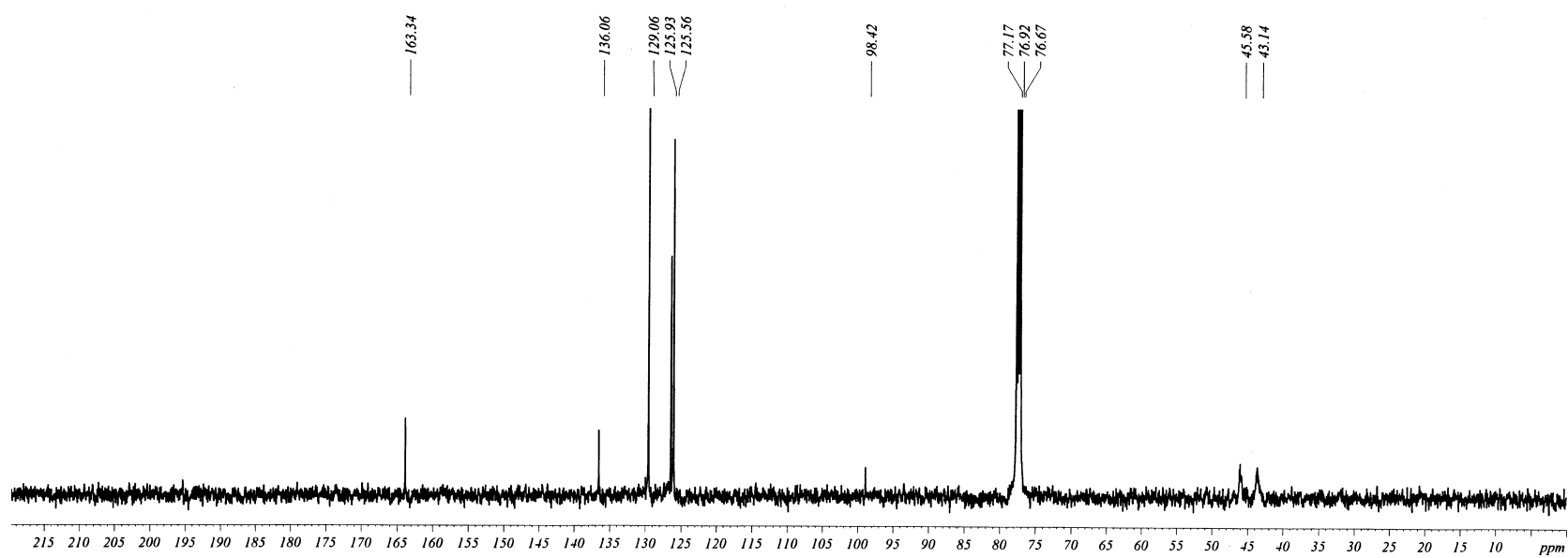


Figure S34 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **3q**

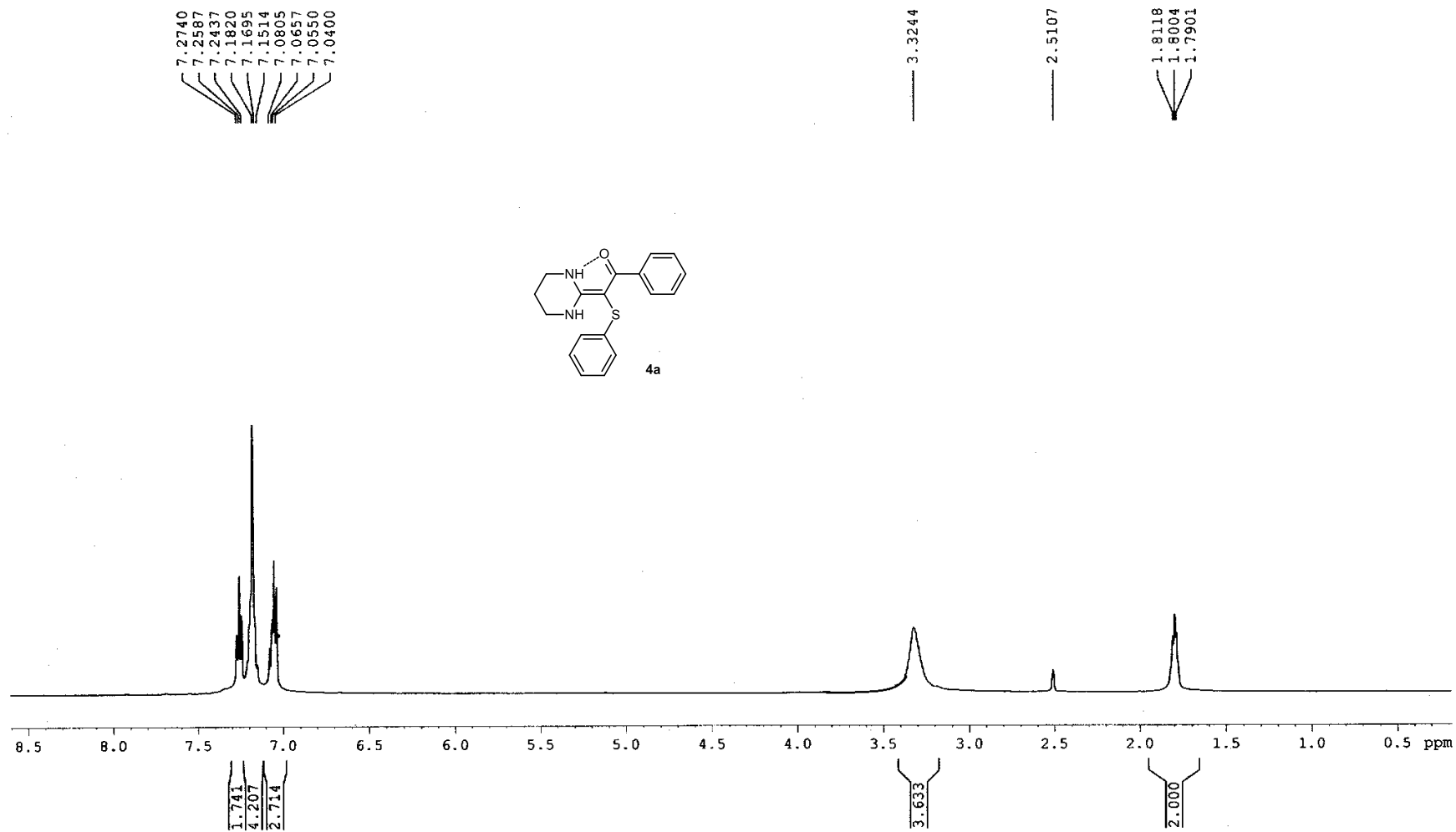


Figure S35 ¹H NMR spectrum (500 MHz, DMSO-*d*₆) of compound **4a**

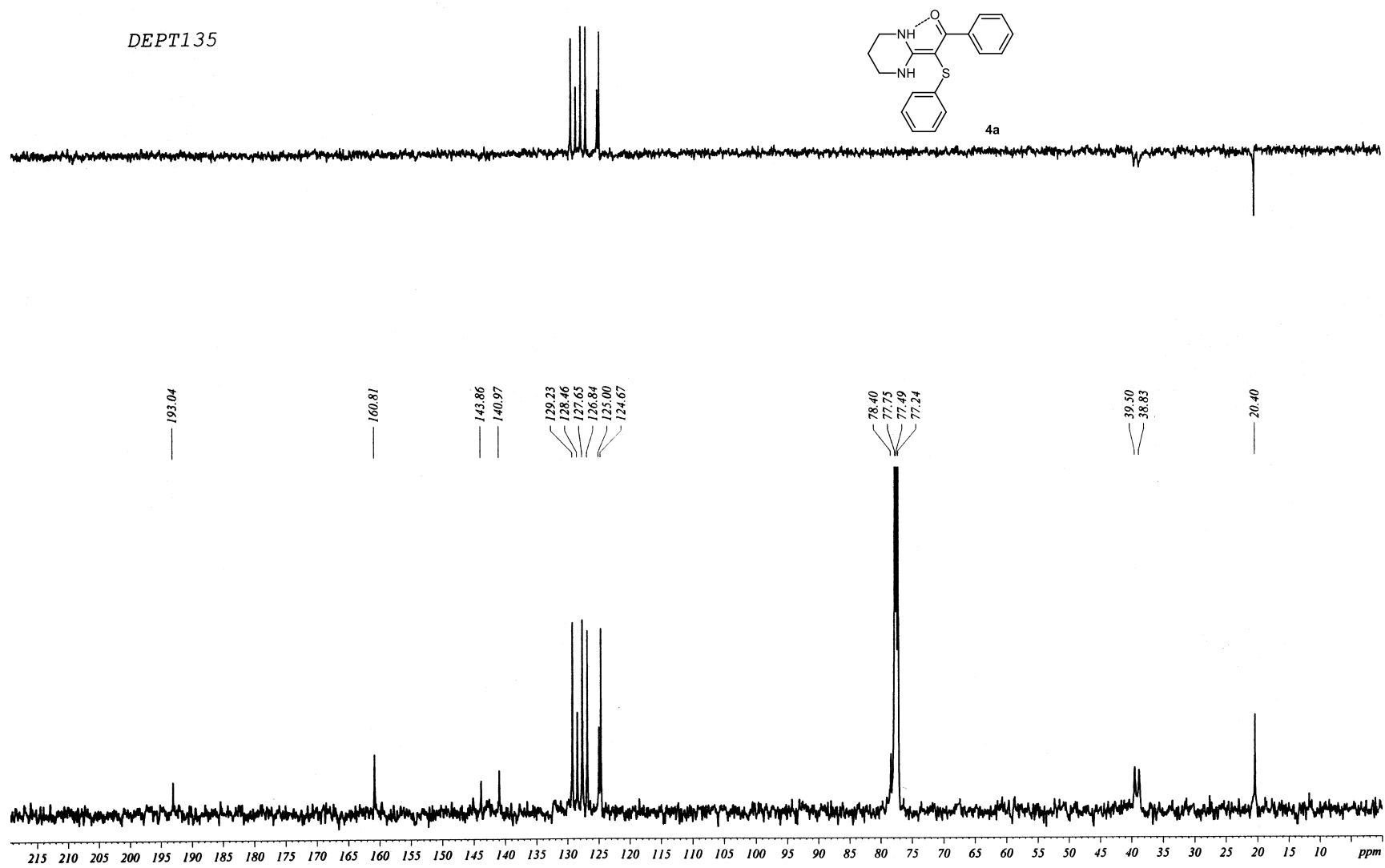


Figure S36 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound 4a

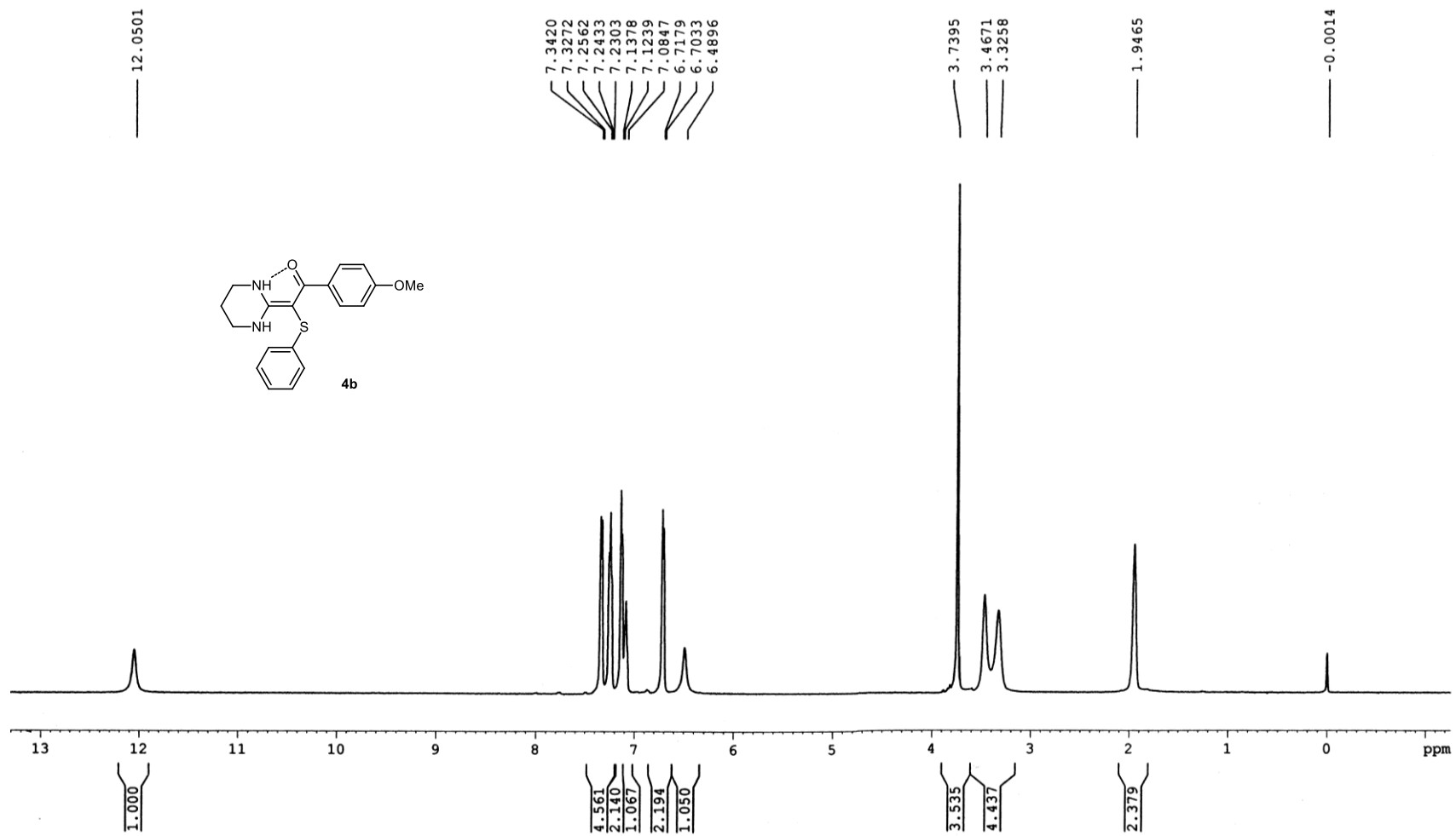


Figure S37 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **4b**

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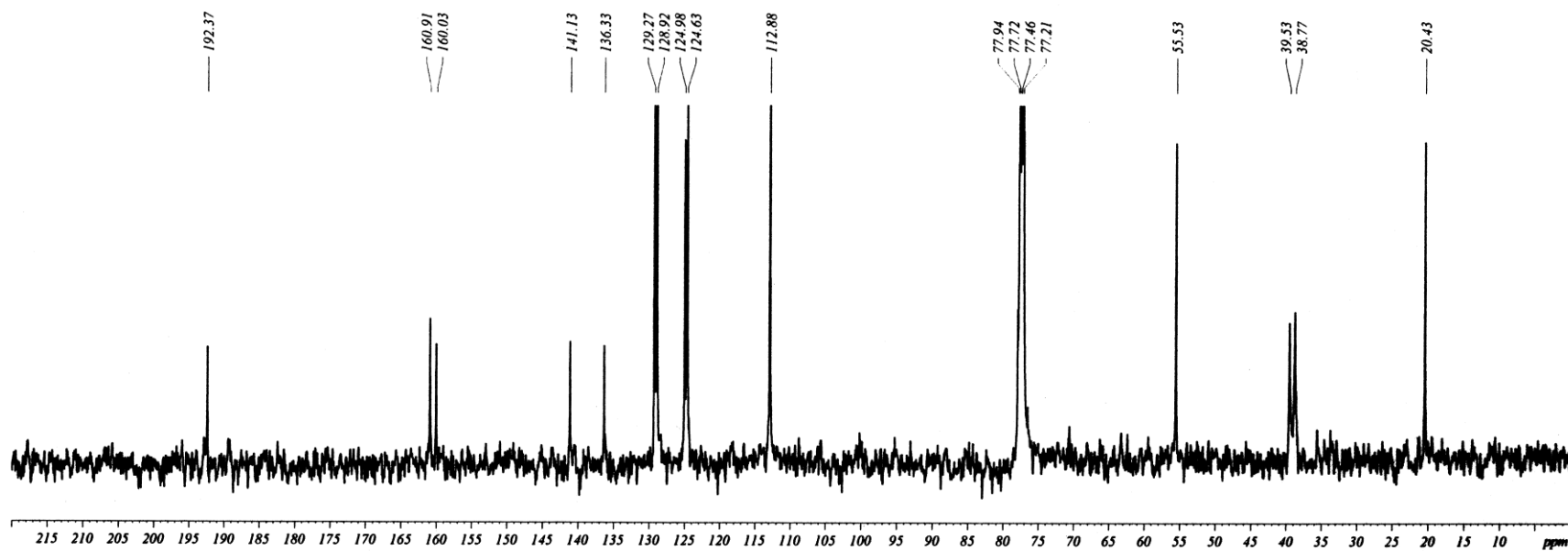
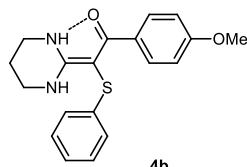


Figure S38 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **4b**

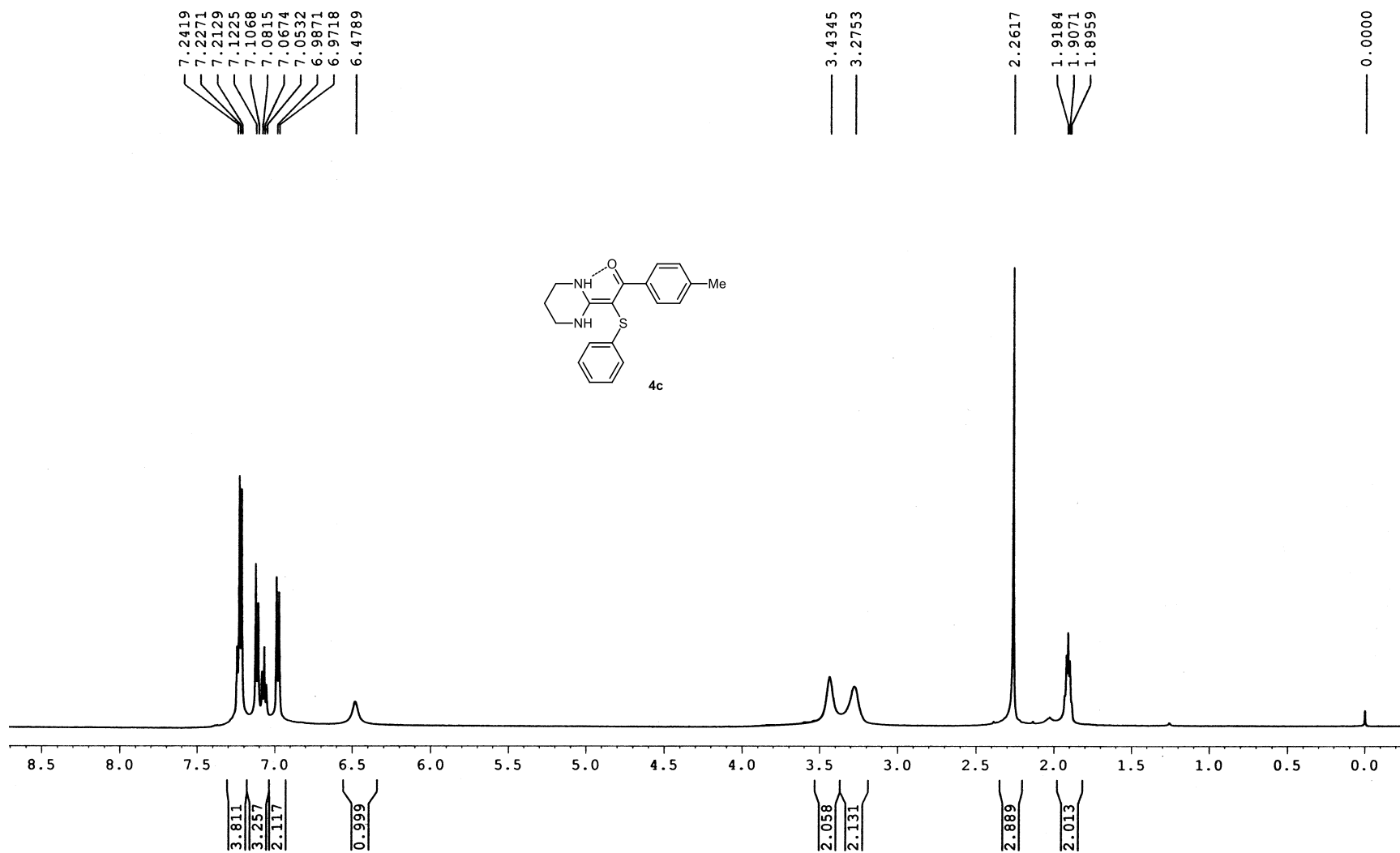


Figure S39 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **4c**

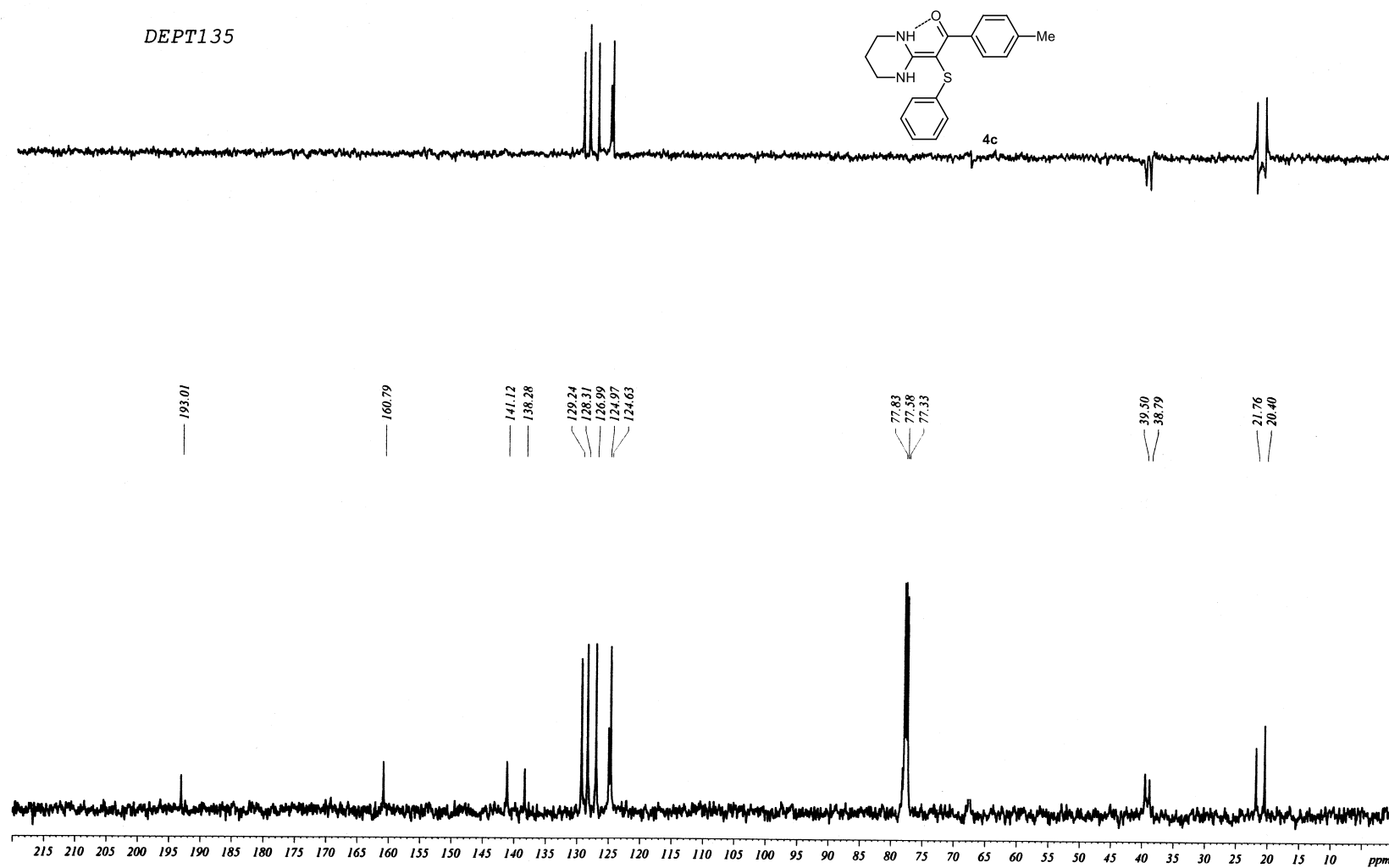


Figure S40 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **4c**

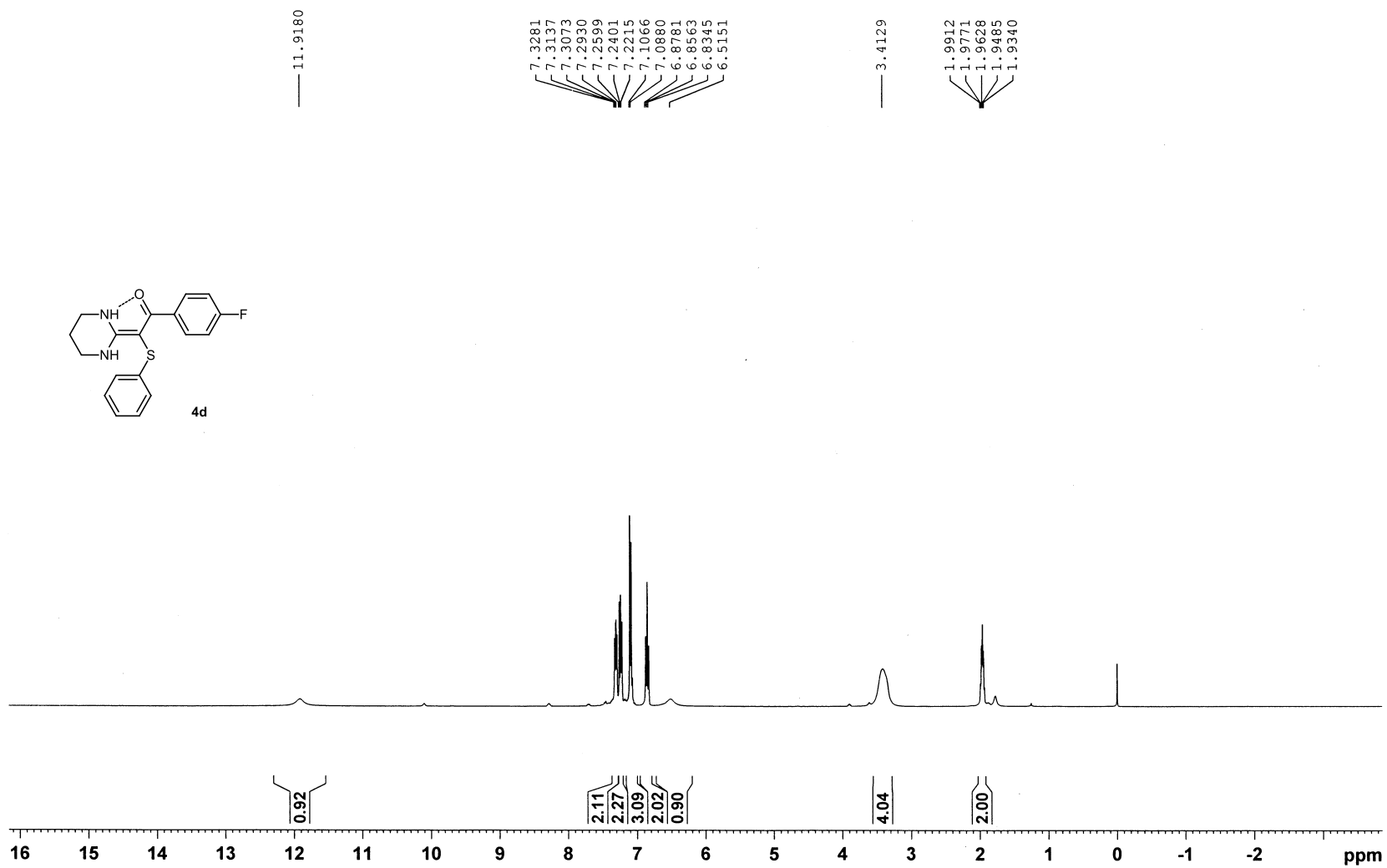


Figure S41 ^1H NMR spectrum (400 MHz,) of compound **4d**

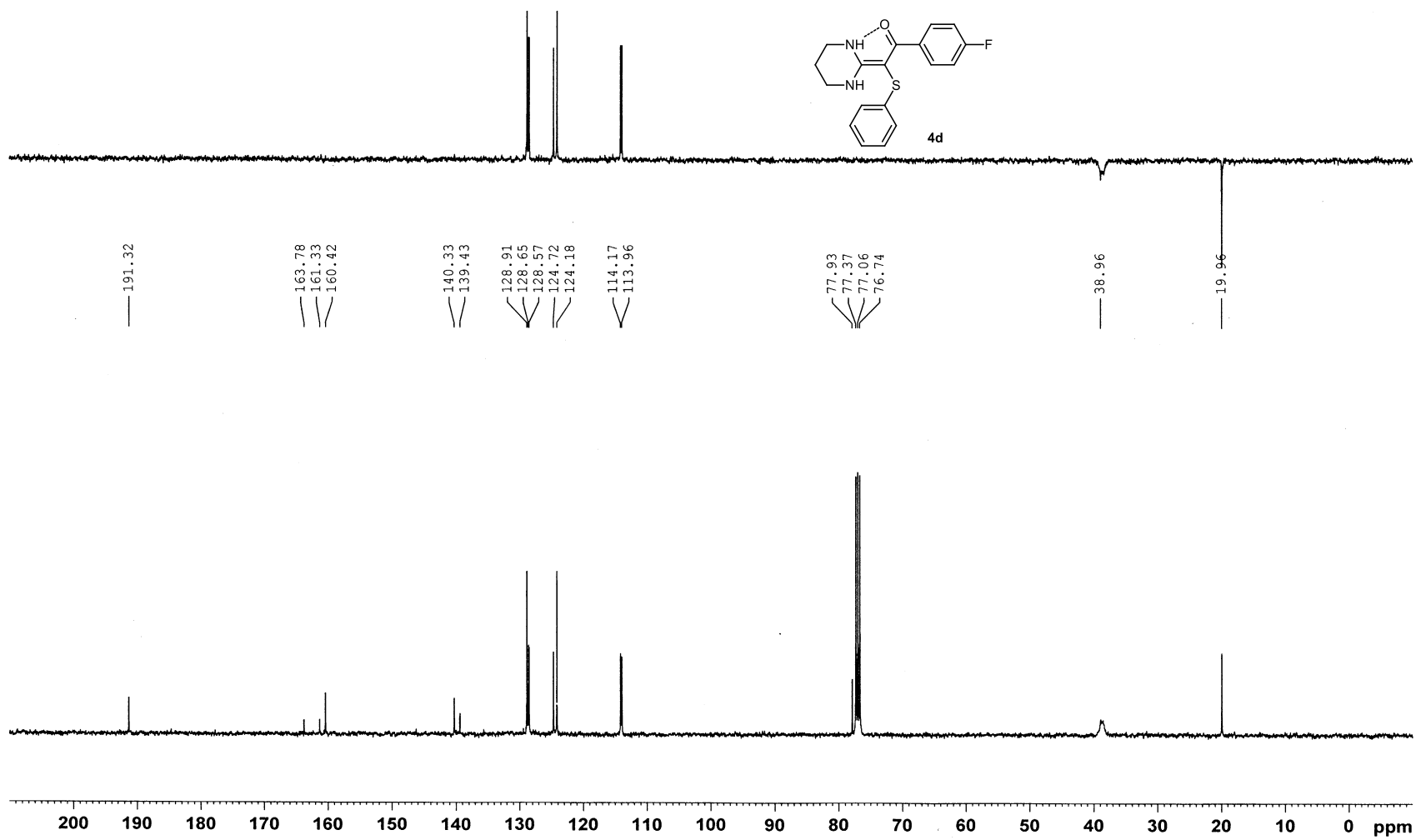


Figure S42 ¹³C NMR spectrum (100 MHz,) of compound 4d

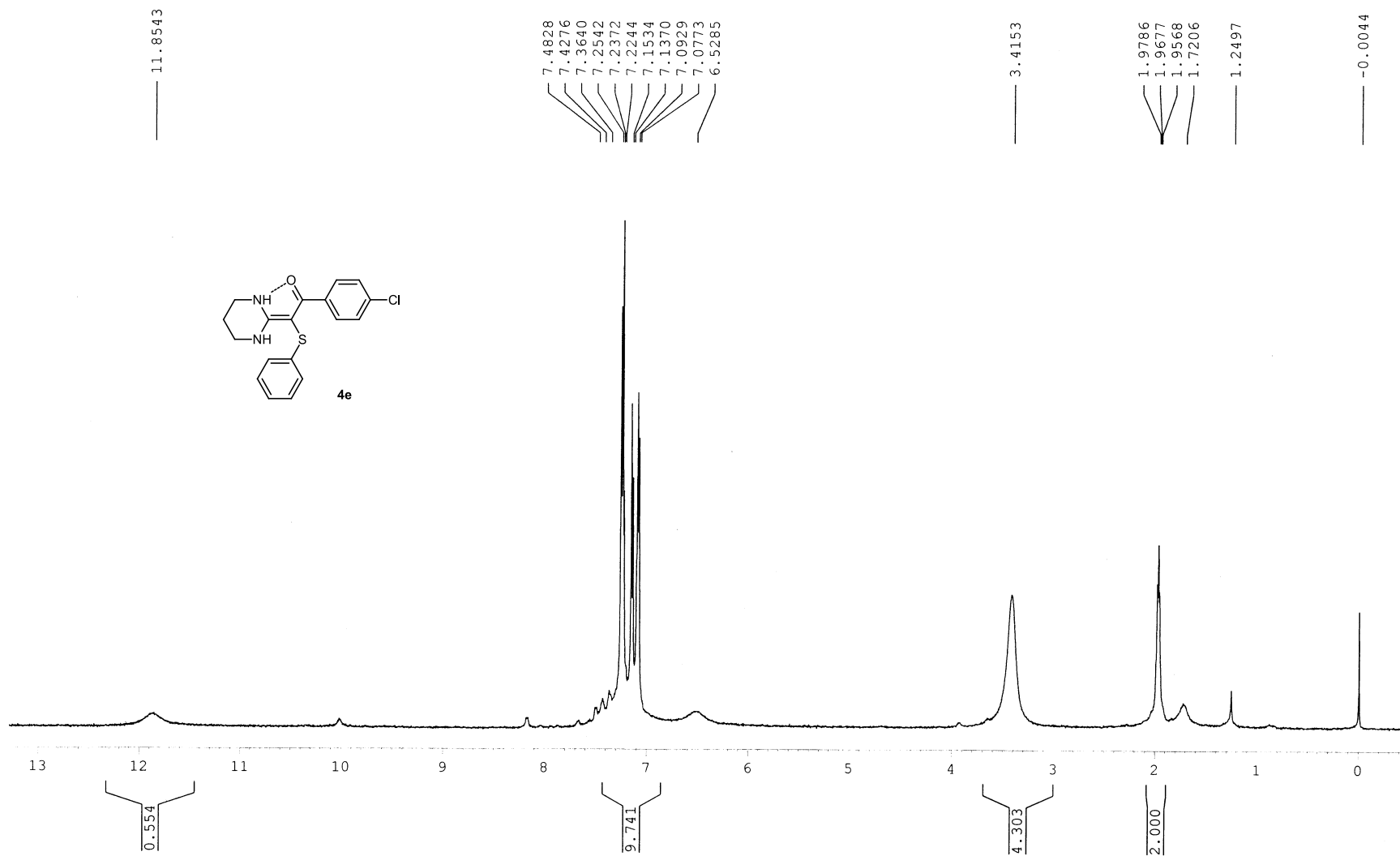


Figure S43 ¹H NMR spectrum (500 MHz, CDCl₃) of compound **4e**

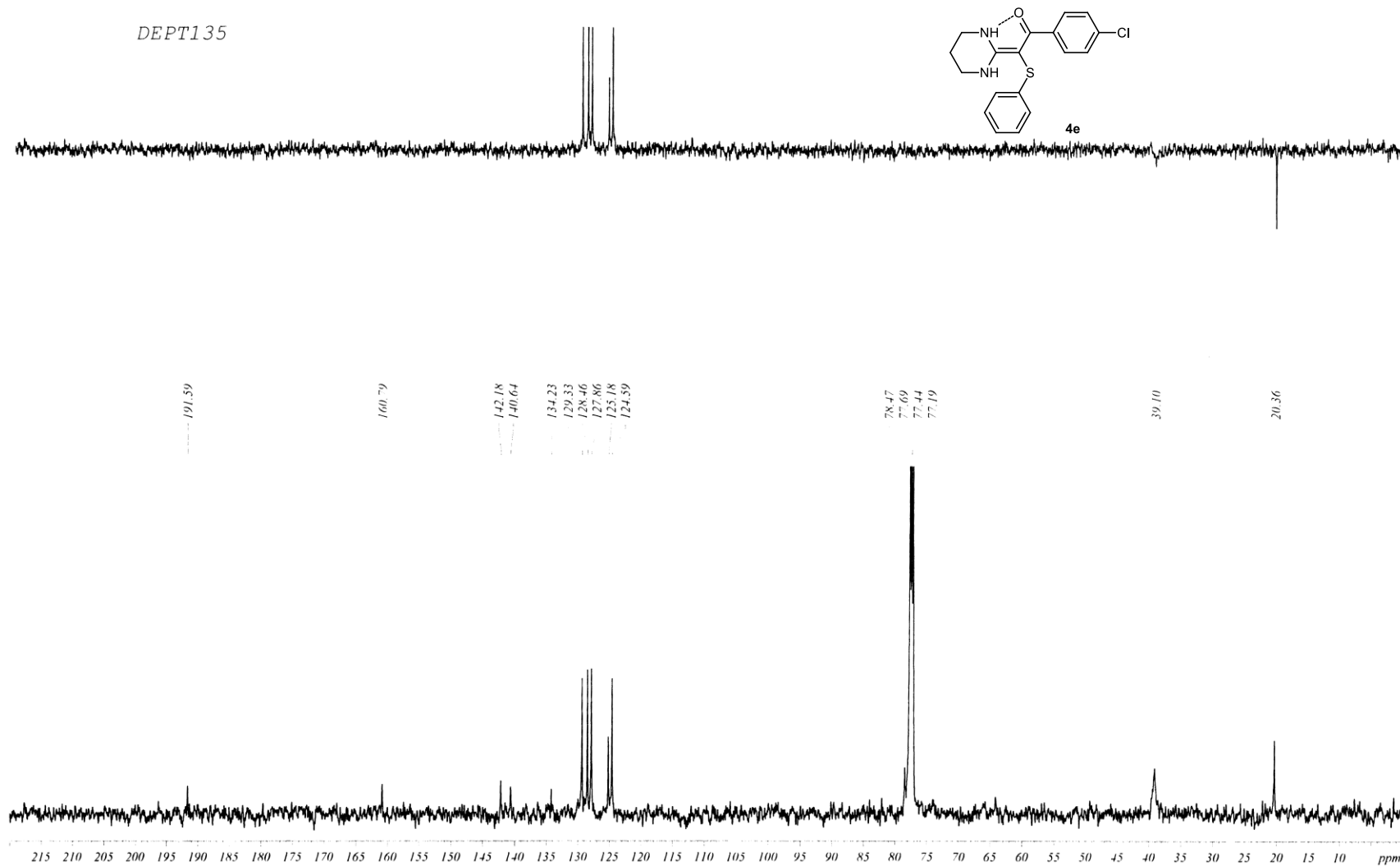
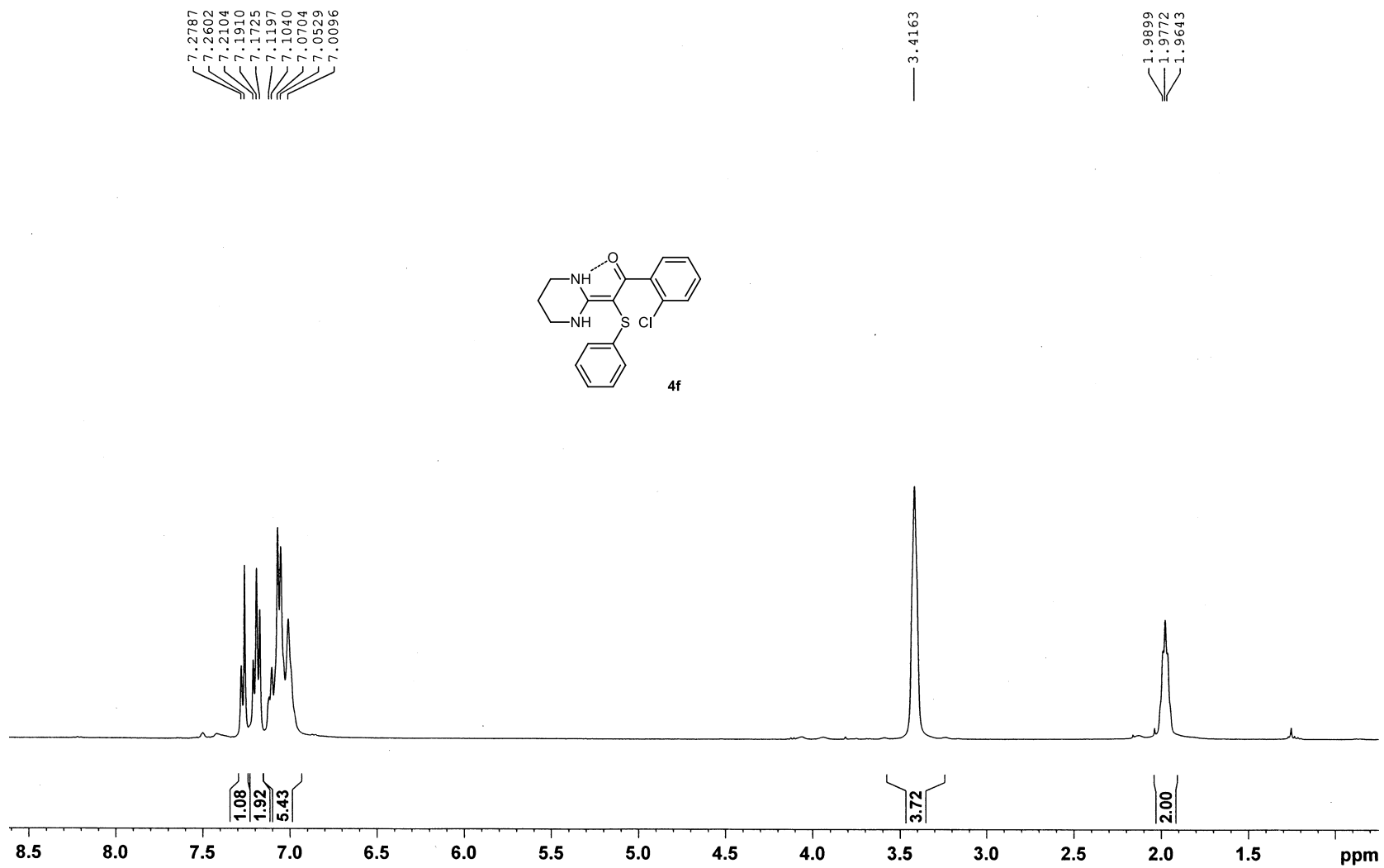


Figure S44 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **4e**



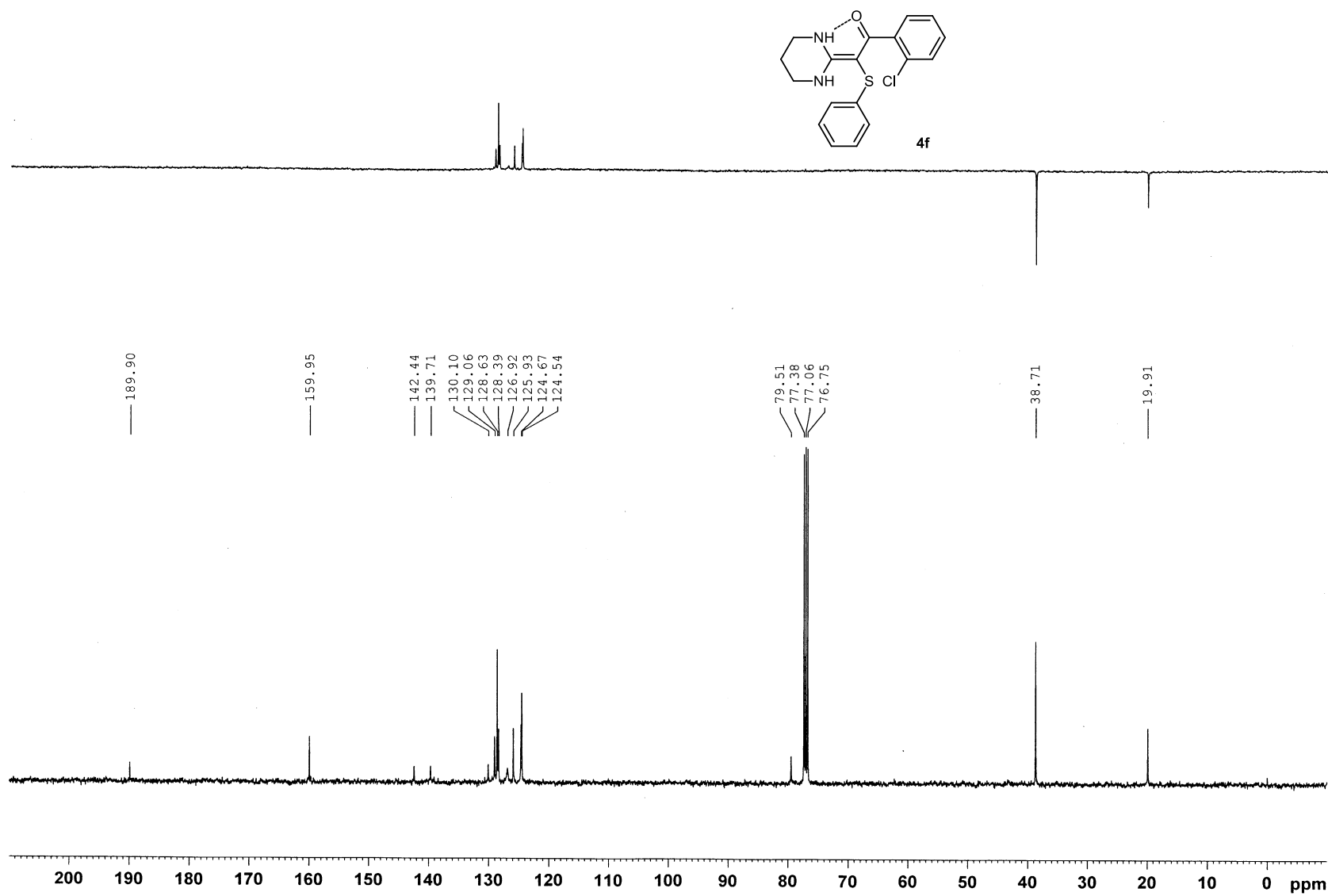


Figure S46 ^{13}C NMR spectrum (100MHz, CDCl_3) of compound **4f**

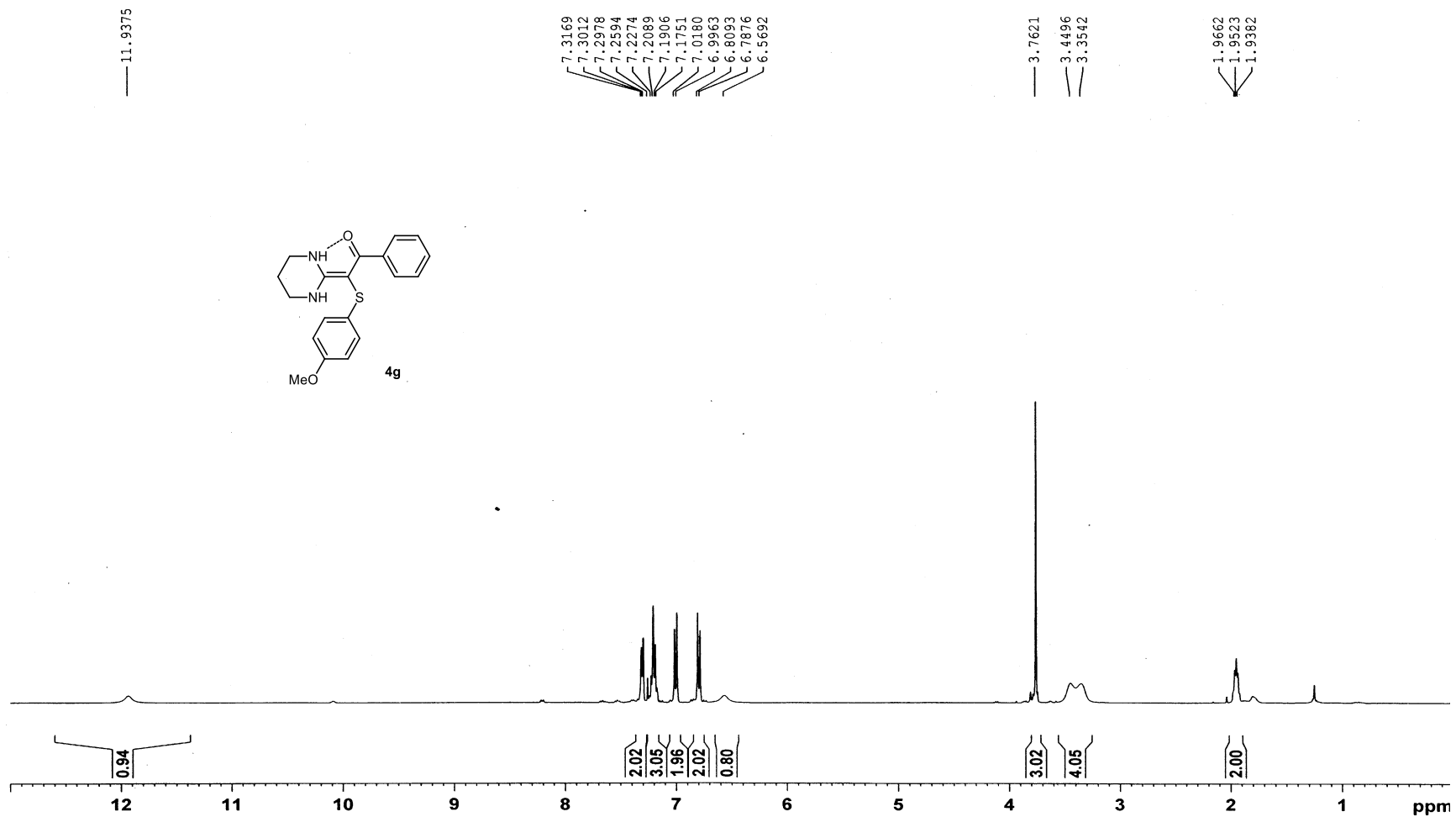


Figure S47 ¹H NMR spectrum (400 MHz, CDCl₃) of compound **4g**

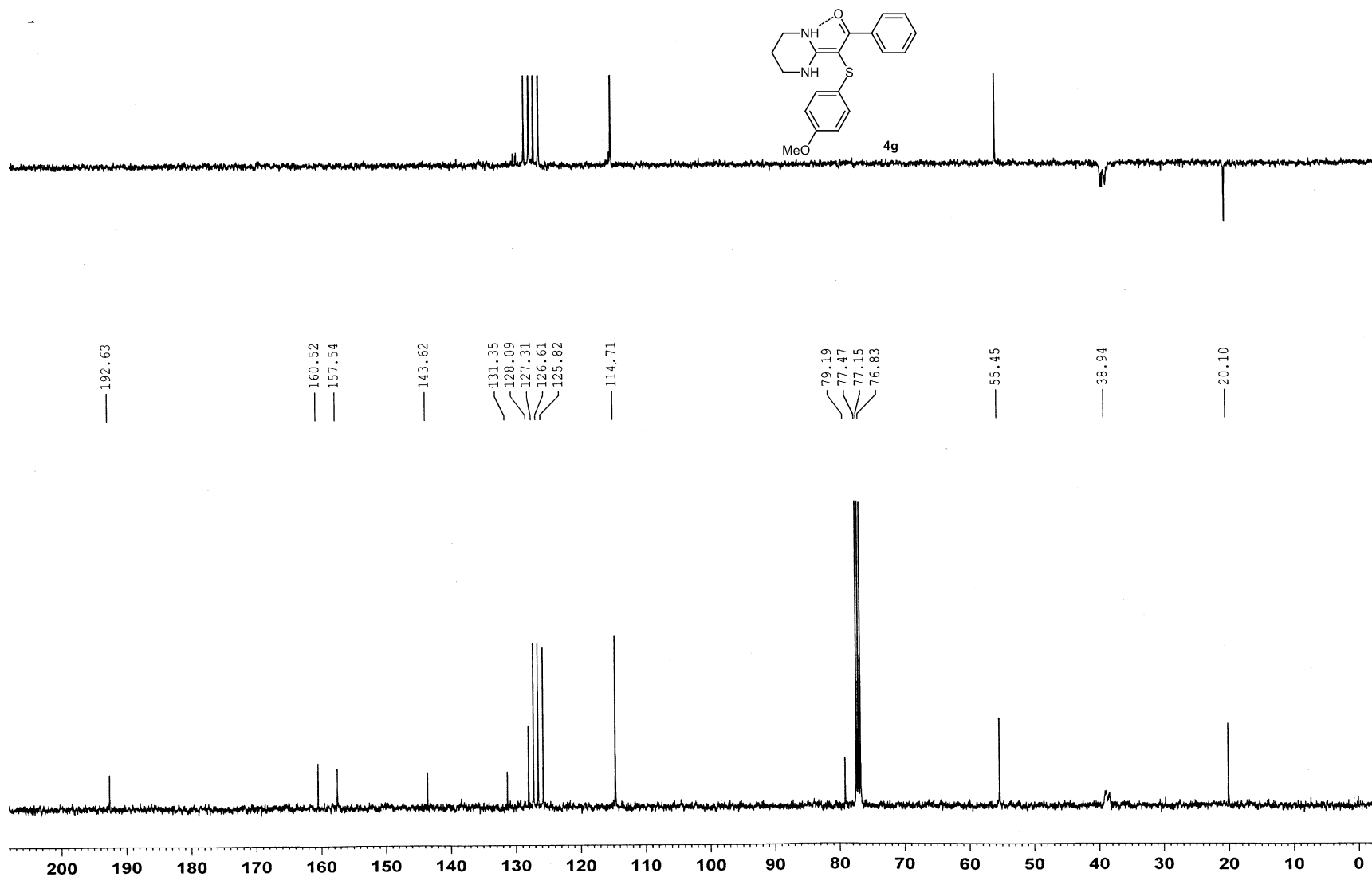


Figure S48 ¹³C NMR spectrum (100 MHz, CDCl₃) of compound 4g

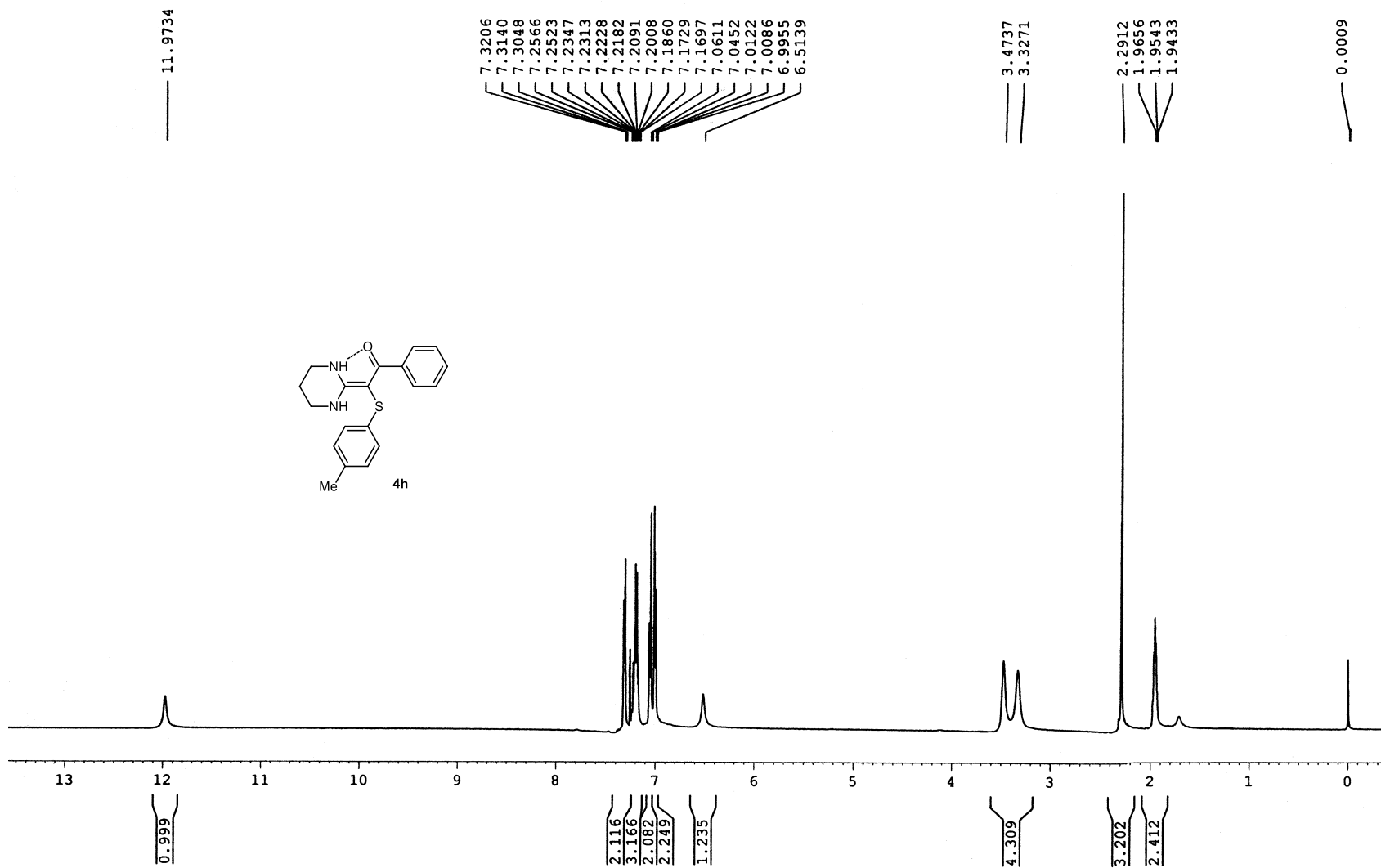


Figure S49 ^1H NMR spectrum (500 MHz, CDCl_3) of compound **4h**

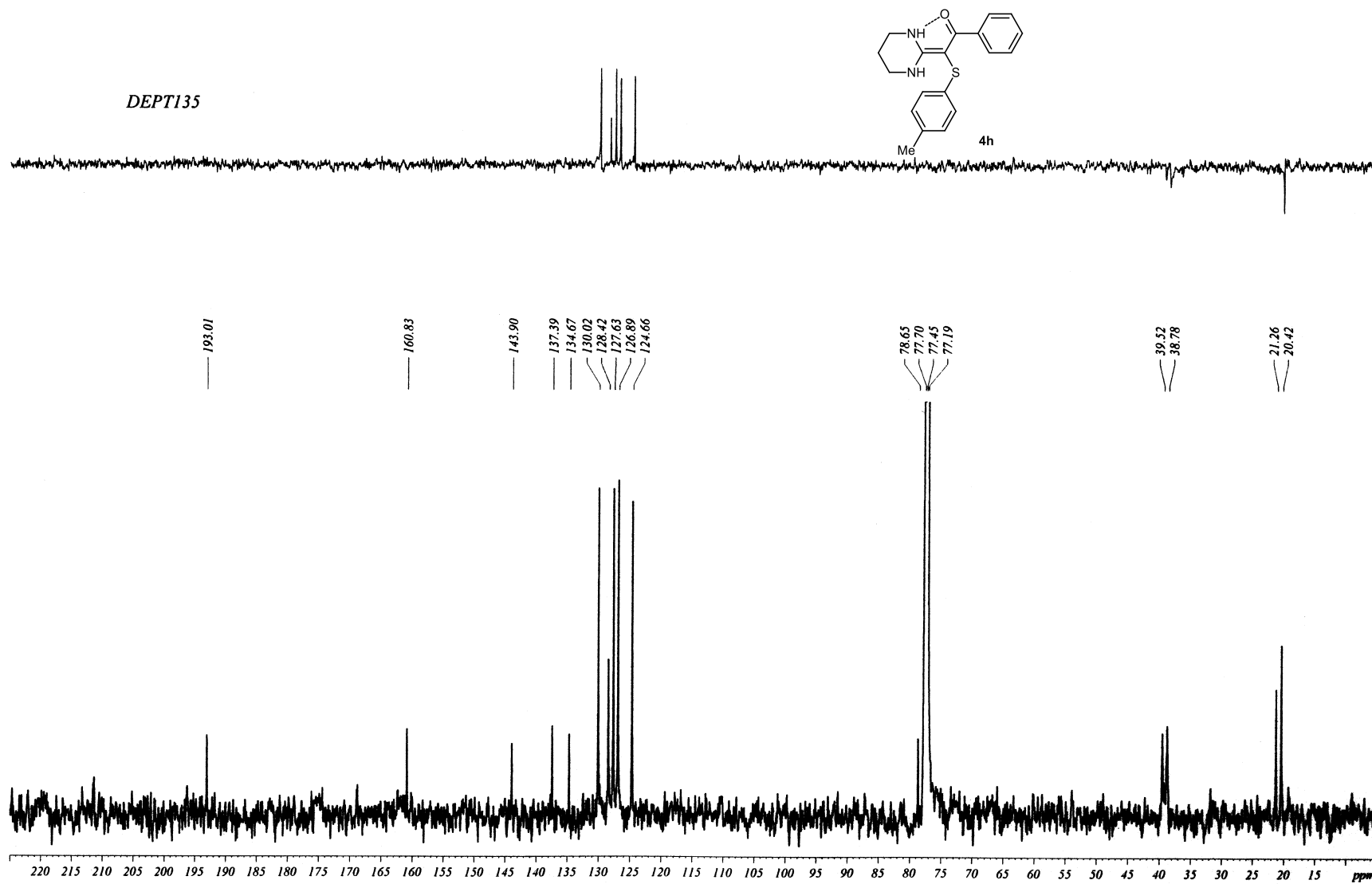


Figure S50 ^{13}C NMR spectrum (125 MHz, CDCl_3) of compound **4h**

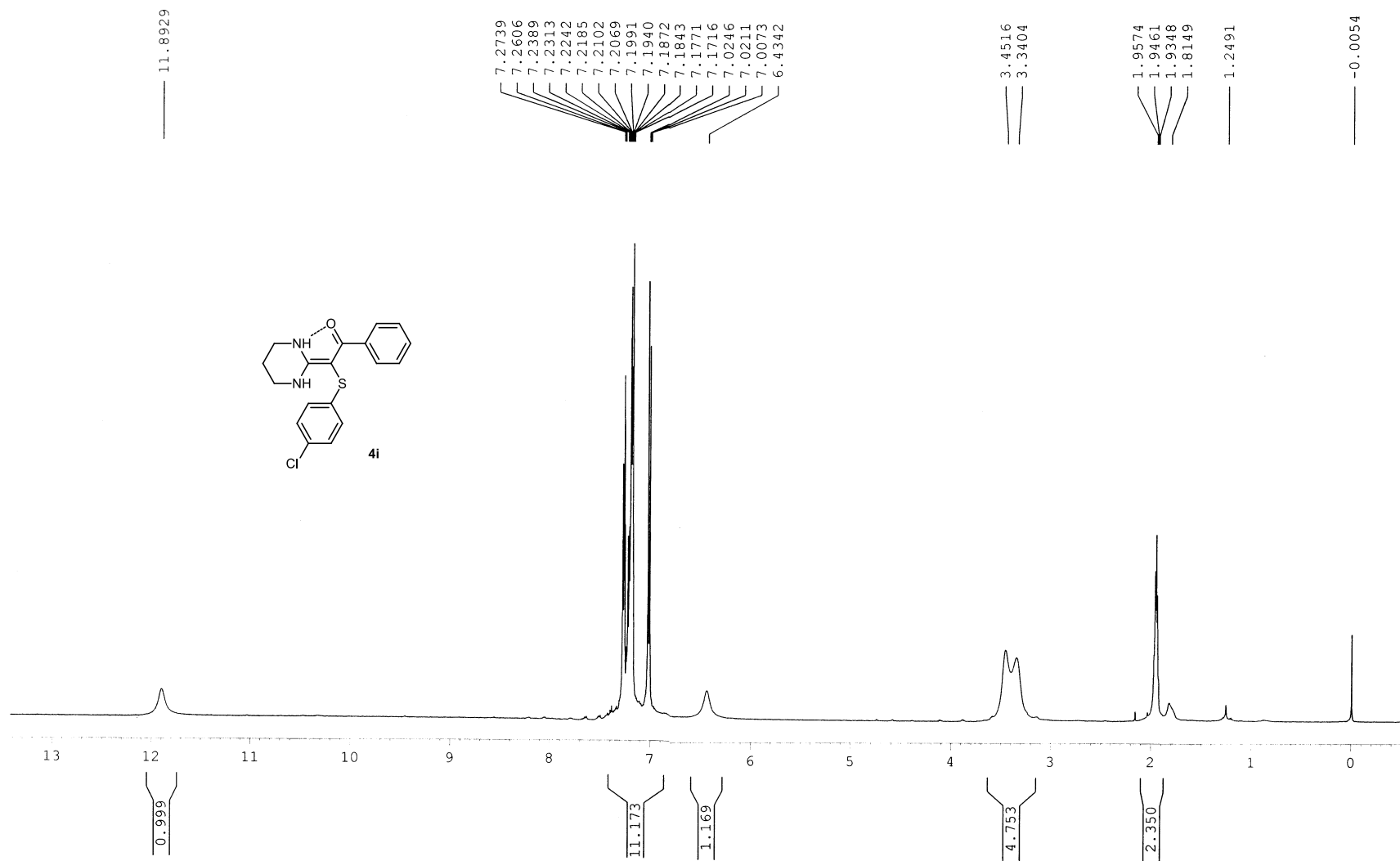


Figure S51. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4i**

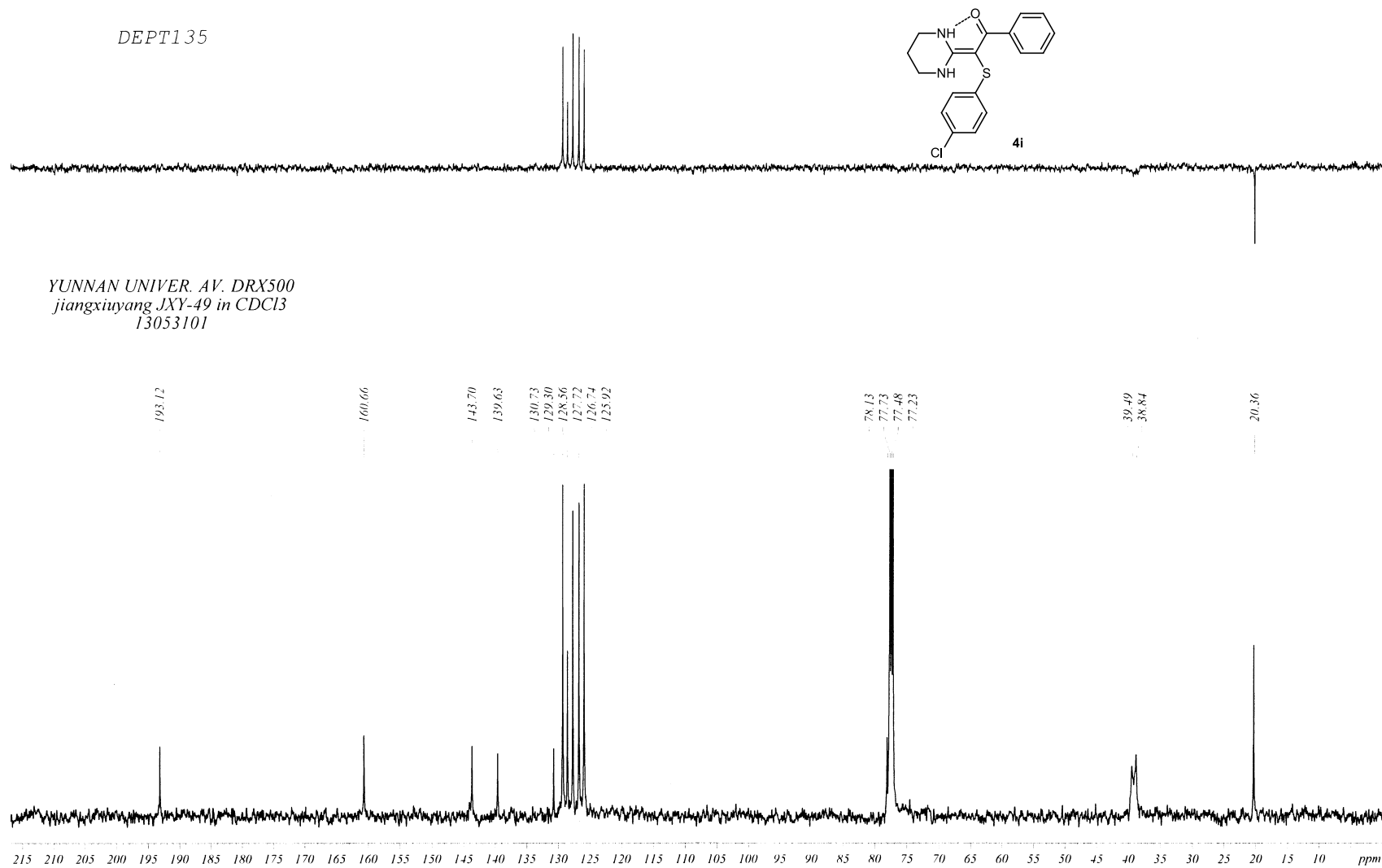


Figure S52. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **4i**

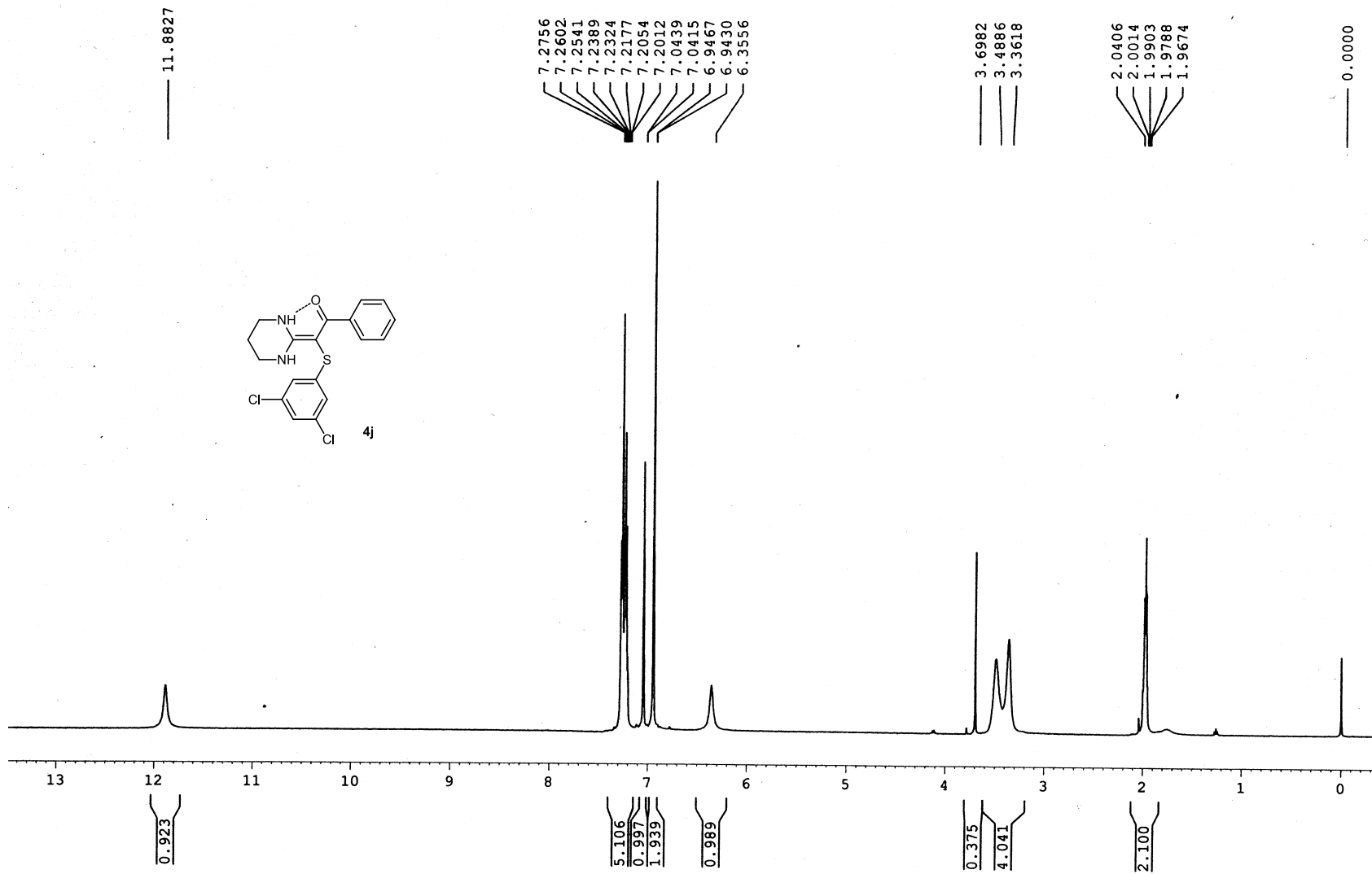


Figure S53. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4j**

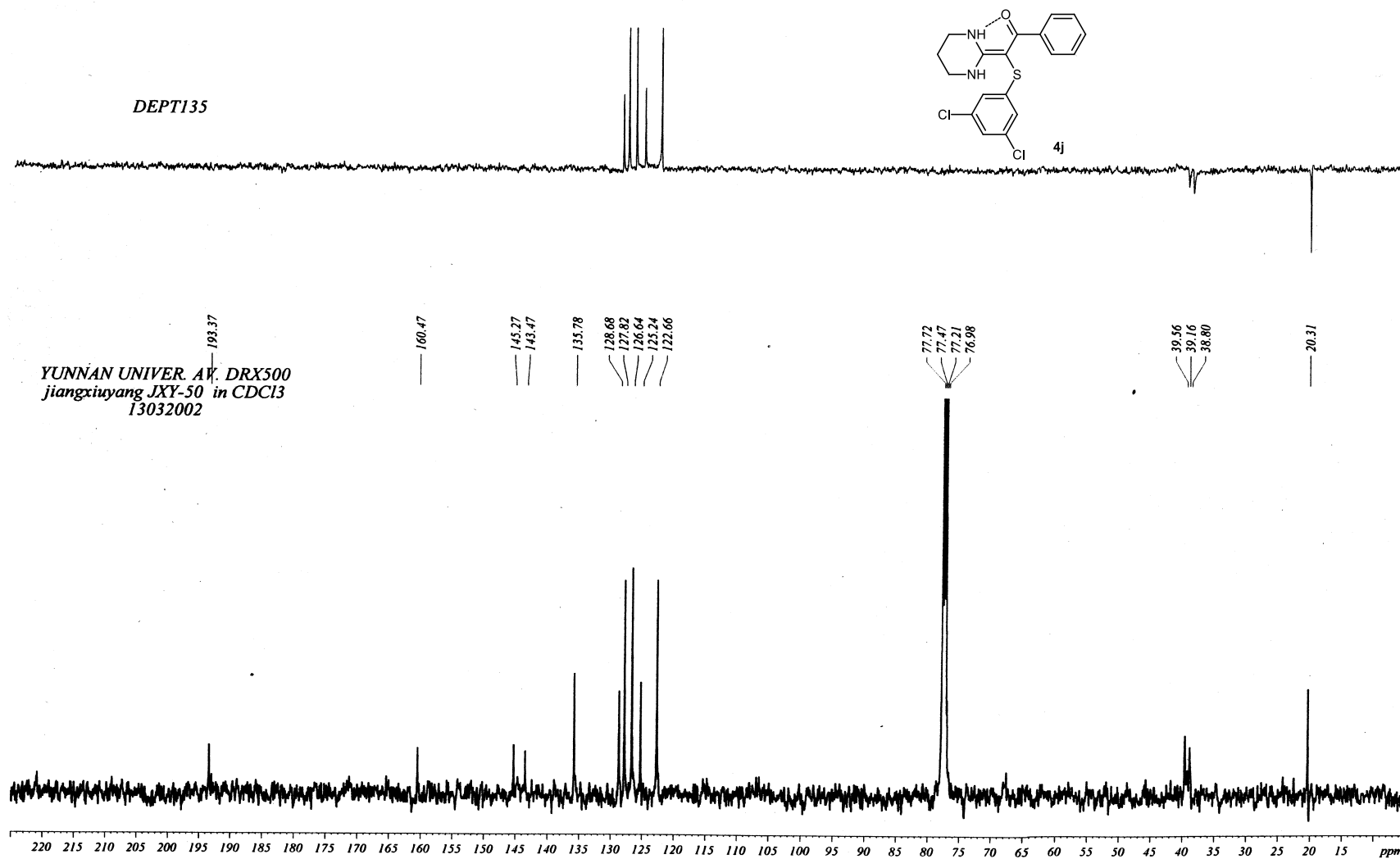


Figure S54. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 4j

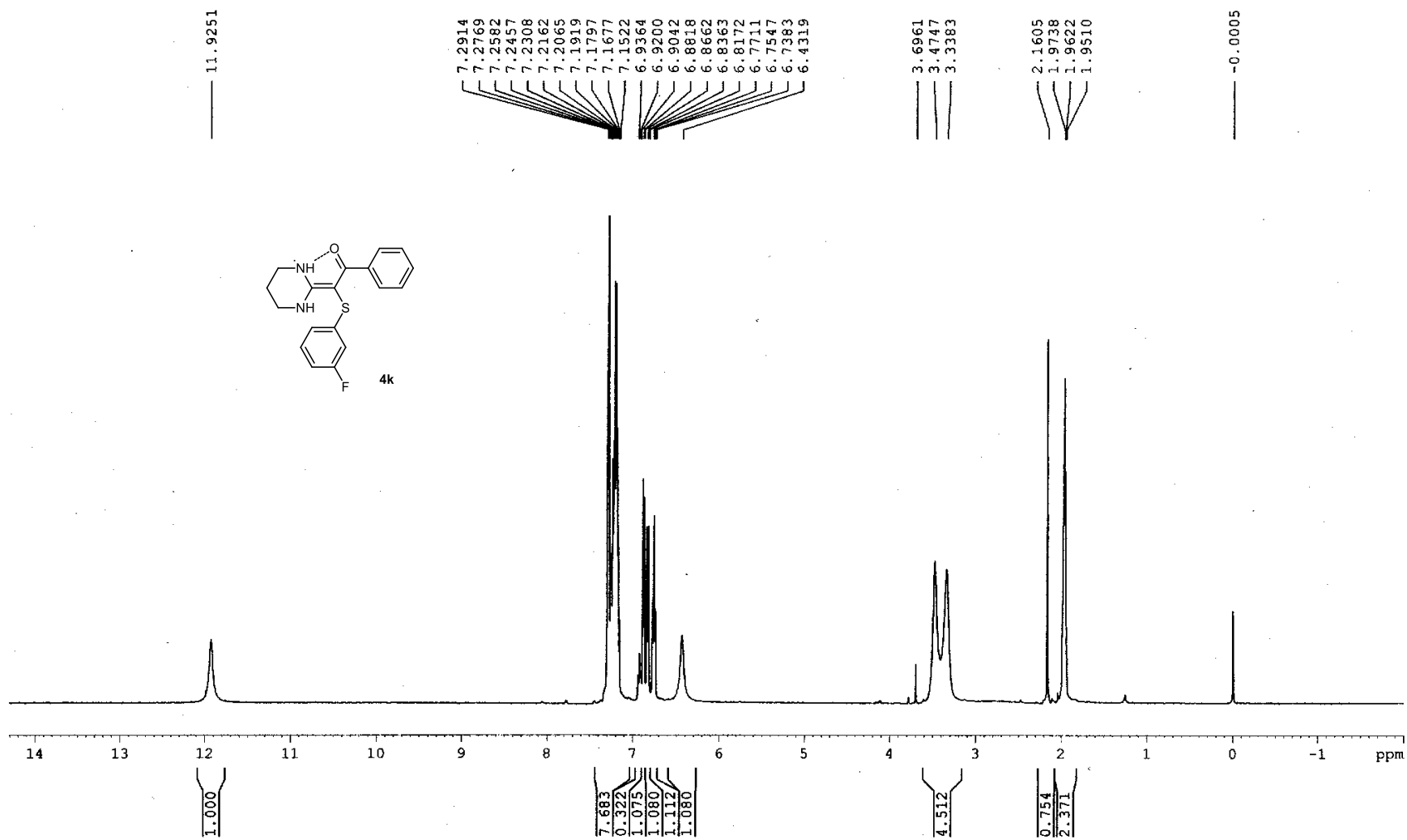


Figure S55. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4k**

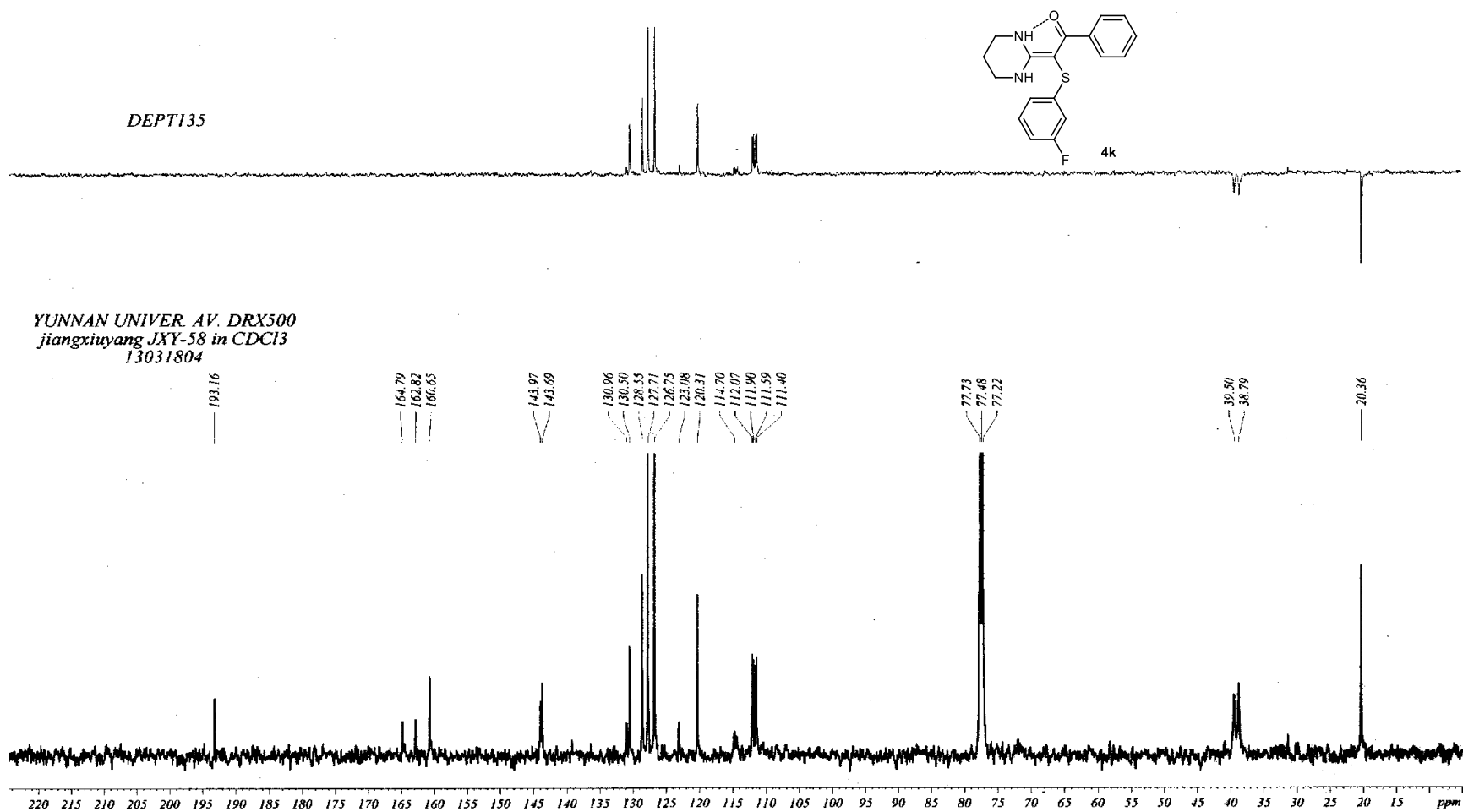


Figure S56. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **4k**

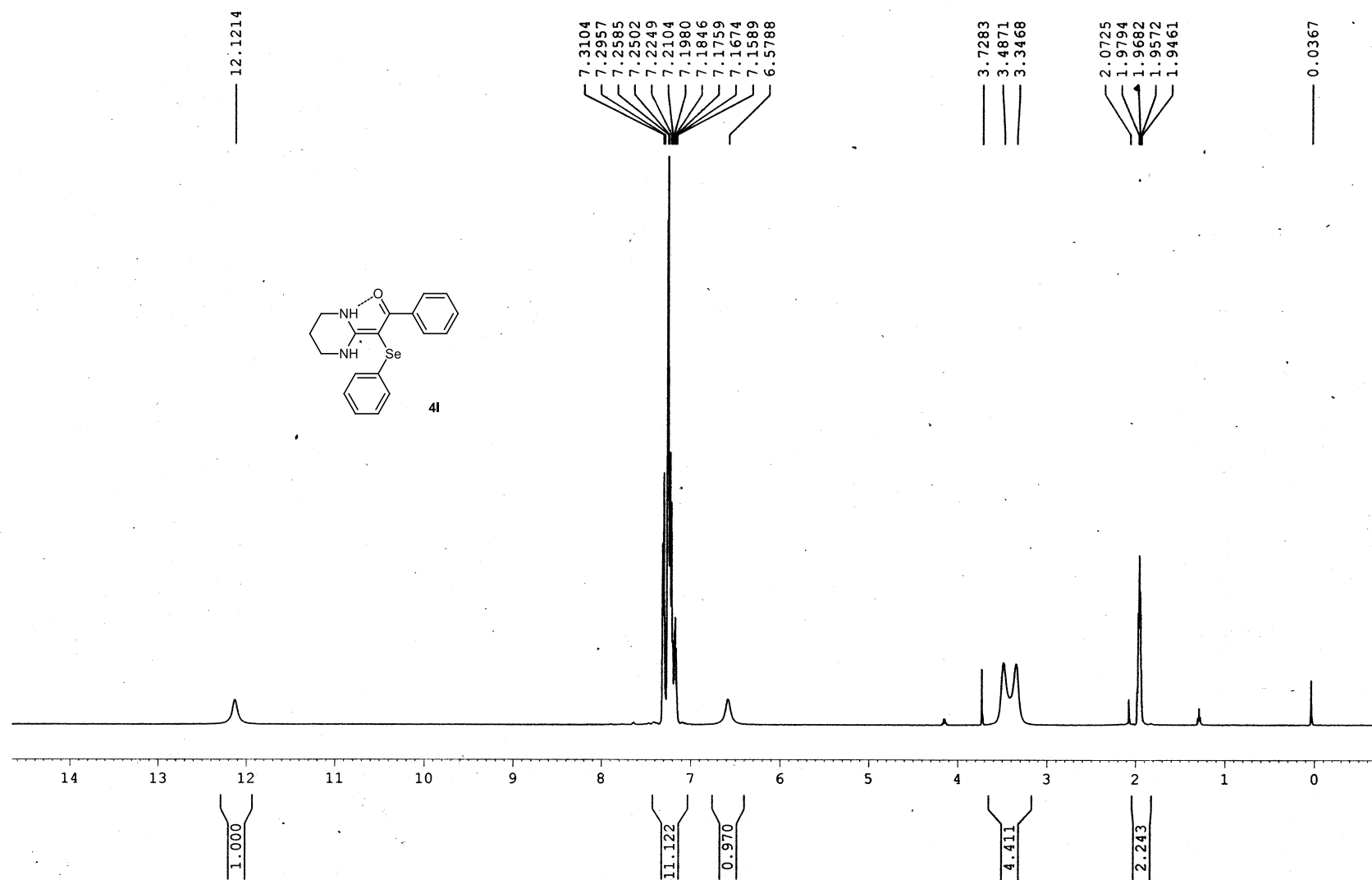


Figure S57. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4l**

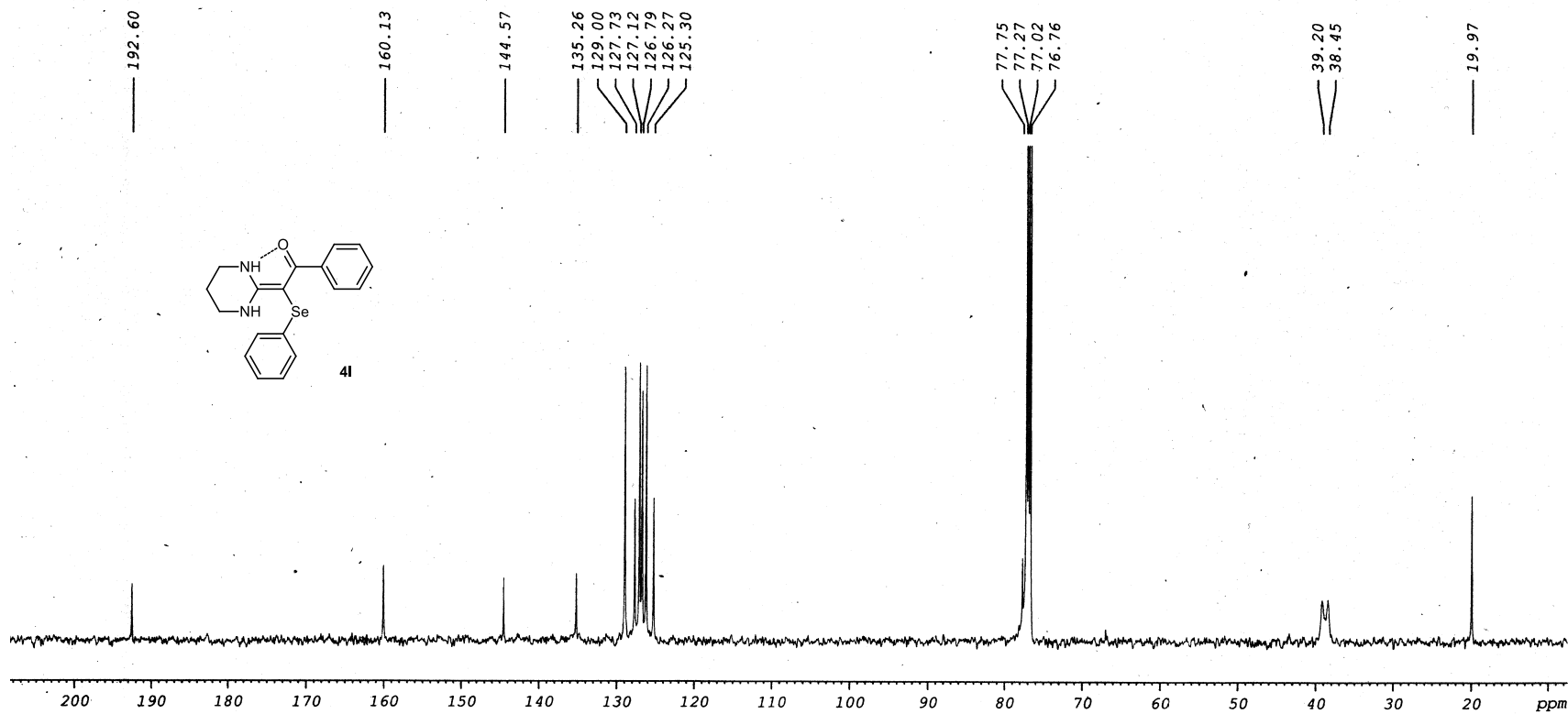


Figure S58. ^{13}C NMR (125 MHz, CDCl_3) spectra of compound **4I**

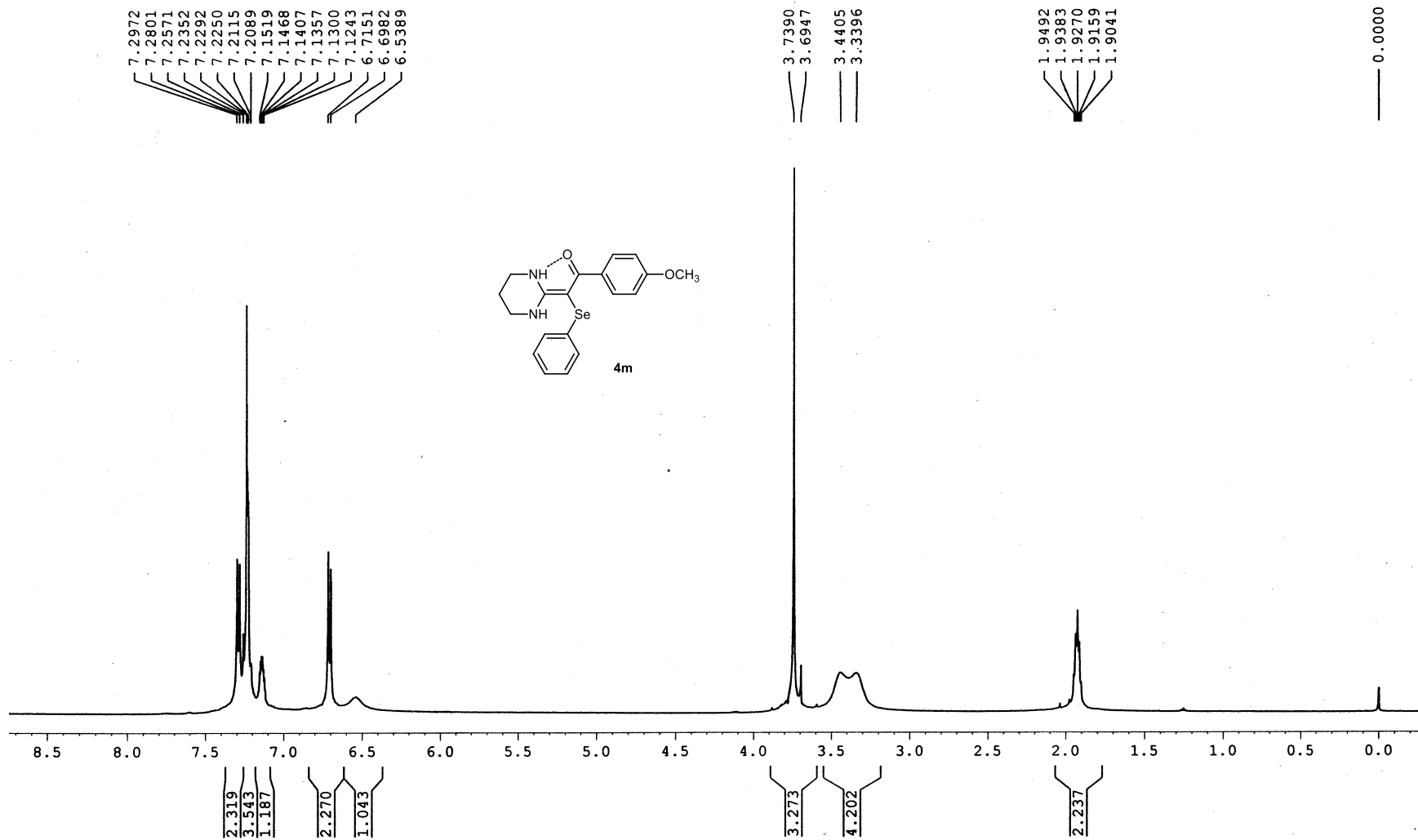
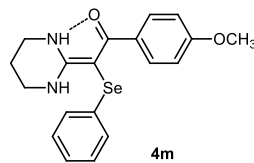


Figure S59. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4m**

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YUNNAN UNIVER. AV. DRX500
jiangxiuyang JXY-20 in CDCl₃
13050702

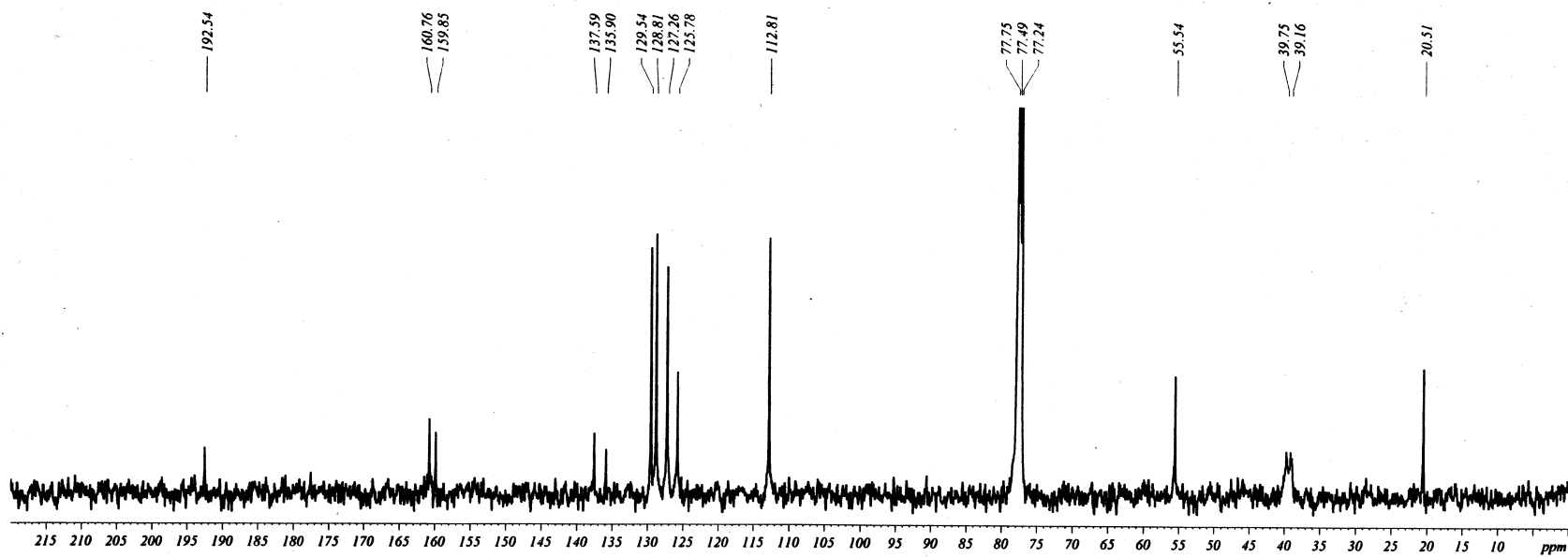
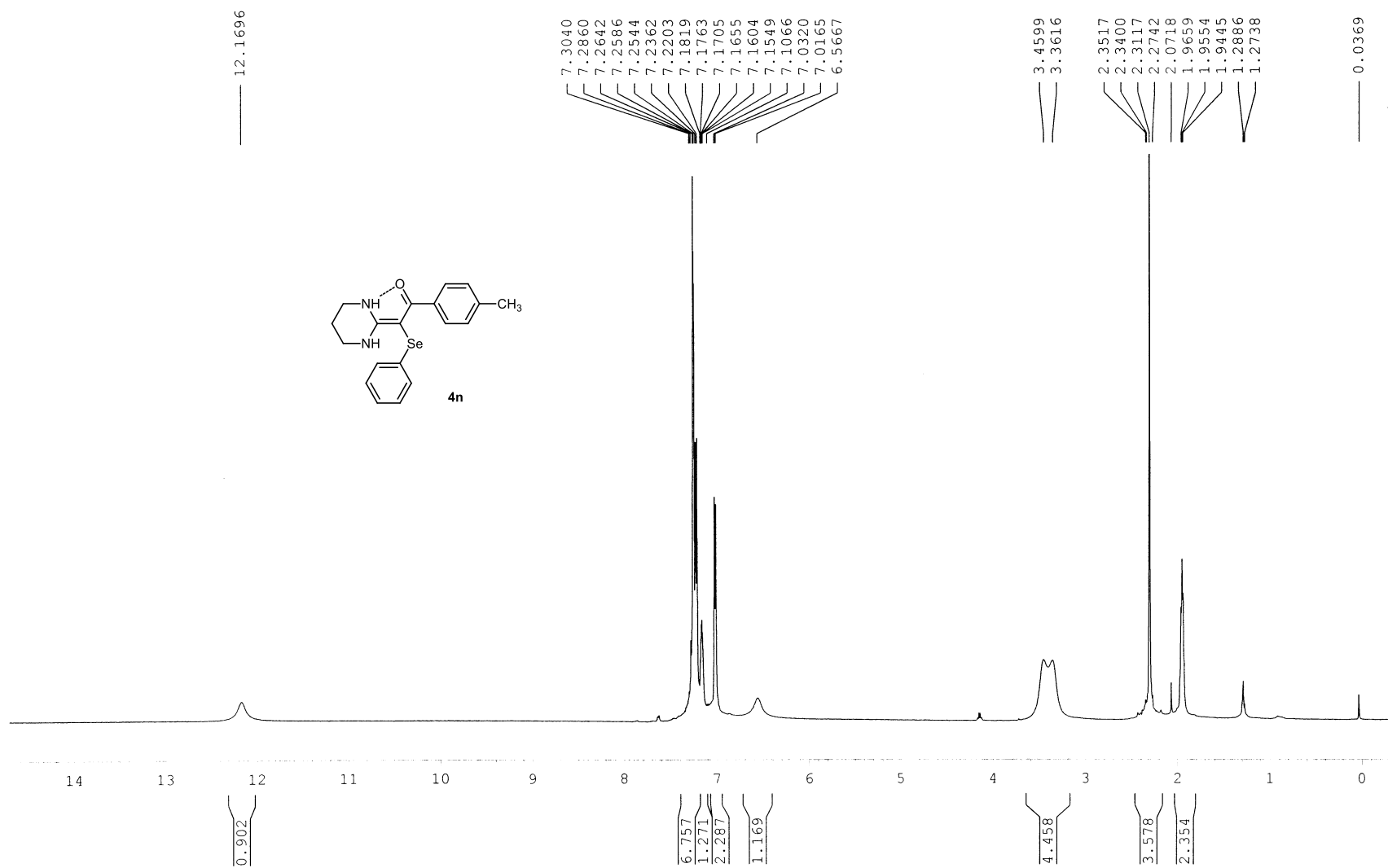


Figure S60. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 4m



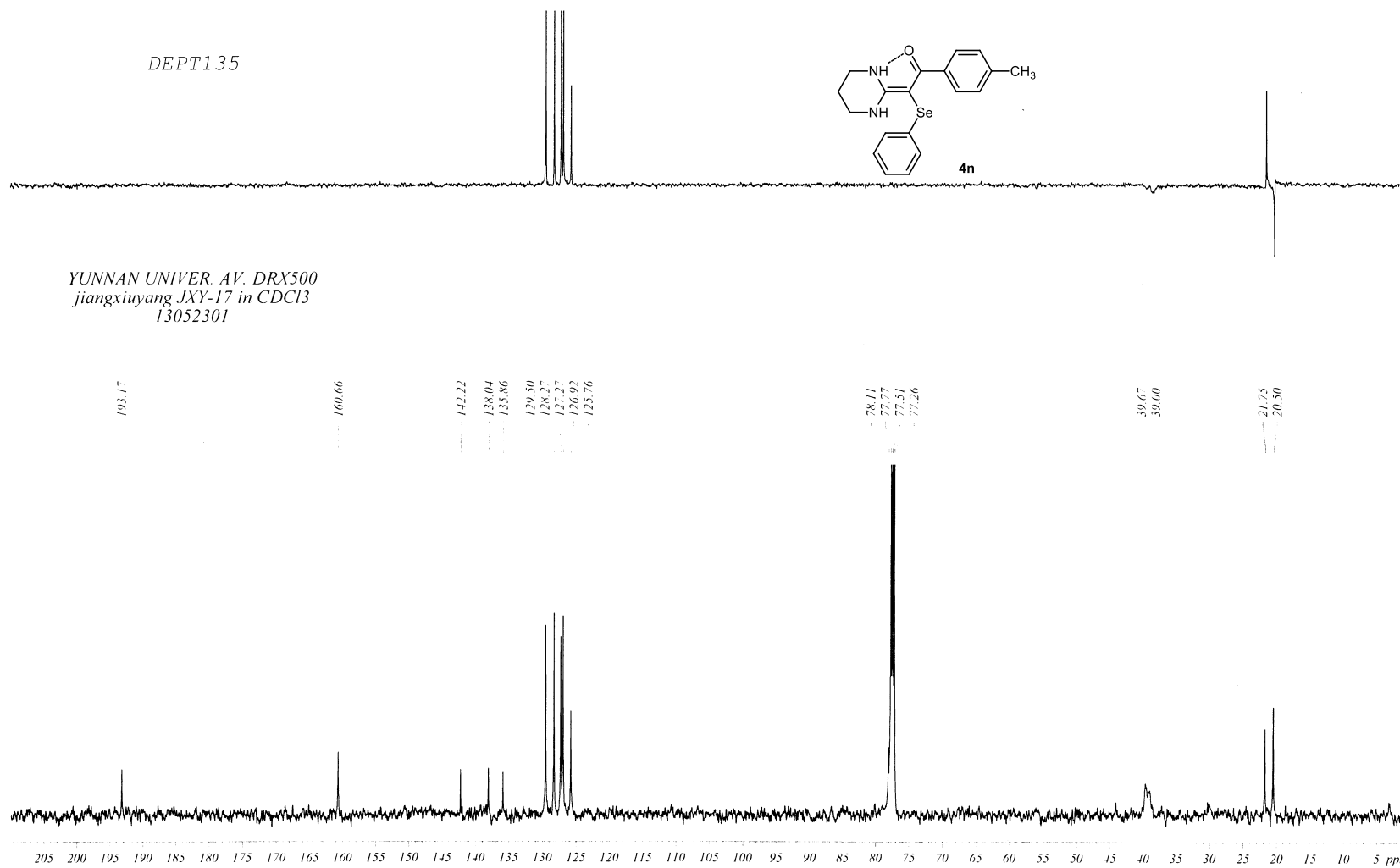


Figure S62. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **4n**

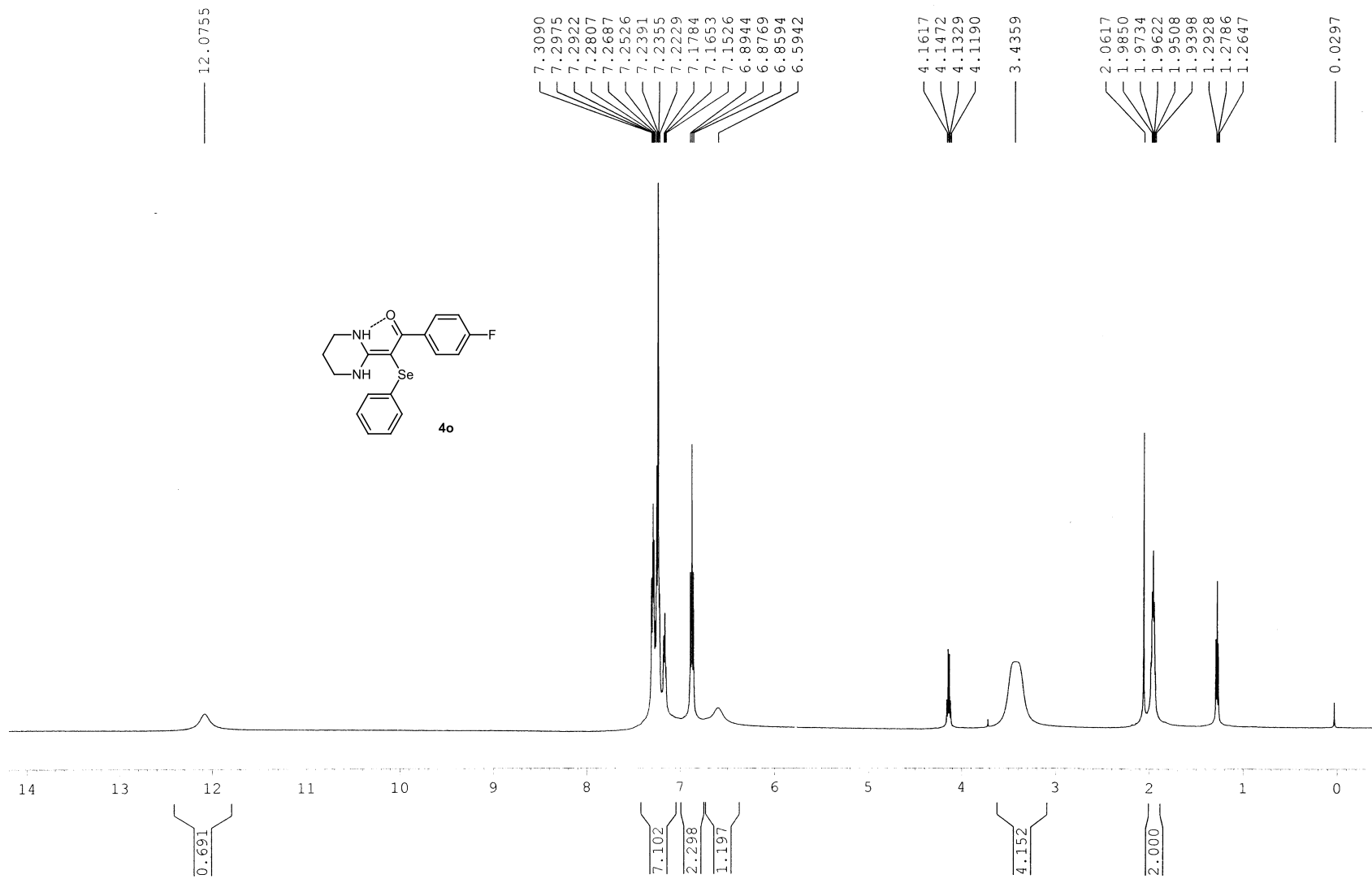


Figure S63. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4o**

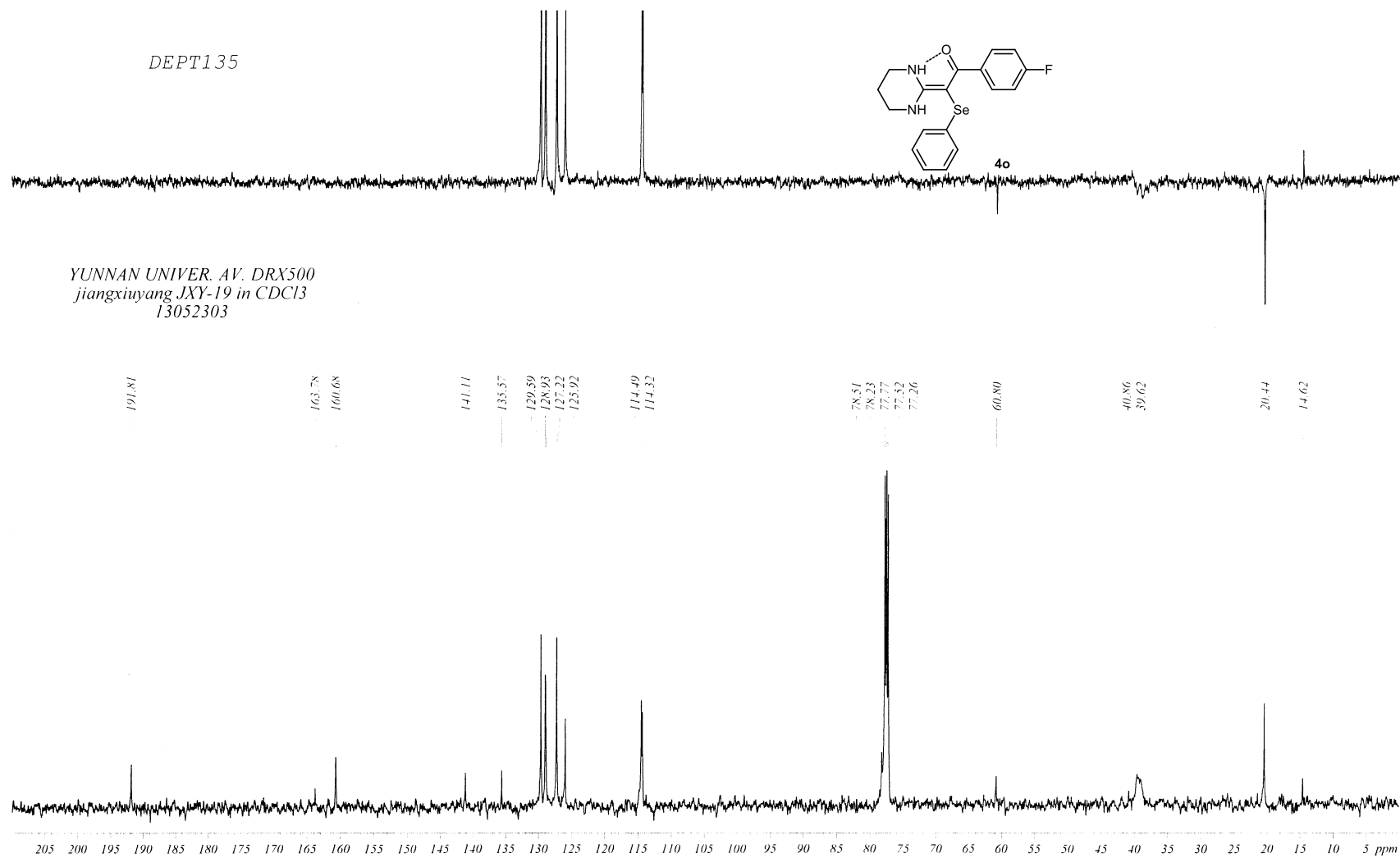


Figure S64. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **4o**

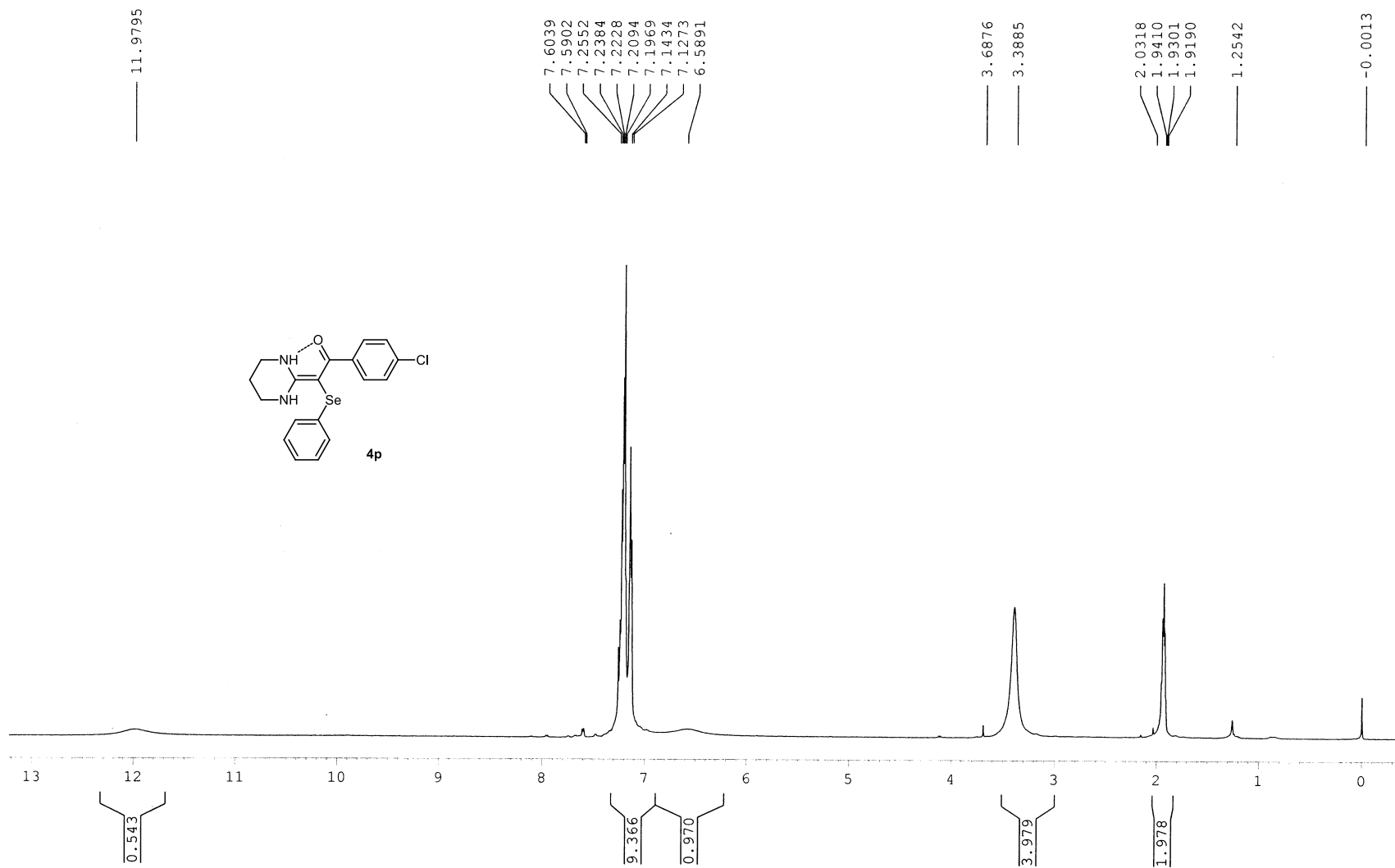


Figure S65. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4p**

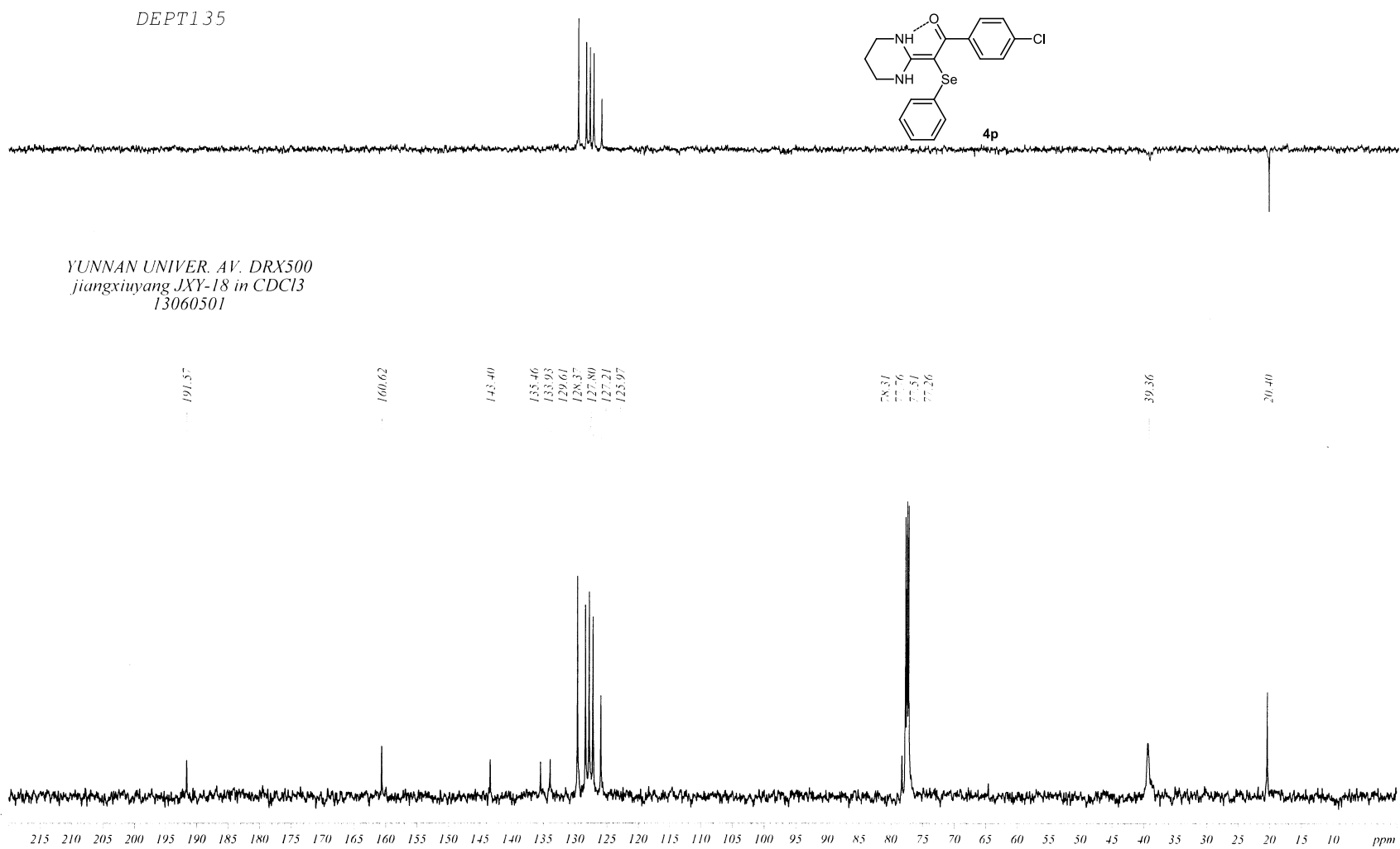


Figure S66. ^{13}C NMR (125 MHz, CDCl_3) spectra of compound **4p**

YUNNAN UNIVER. AV. DRX500
jiangxiuyang JXY-A-8 in CDCl₃
13050901

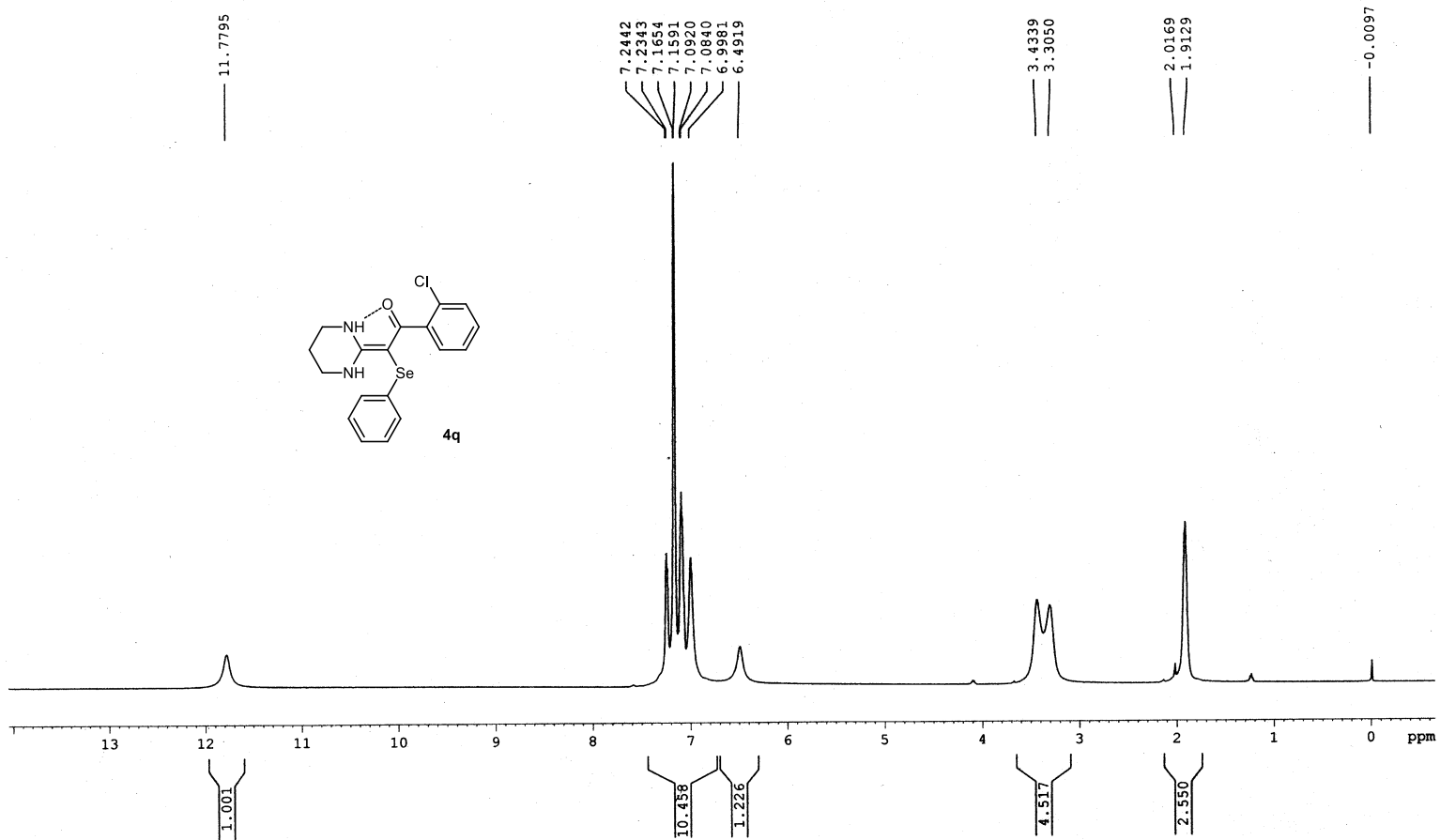


Figure S67. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4q**

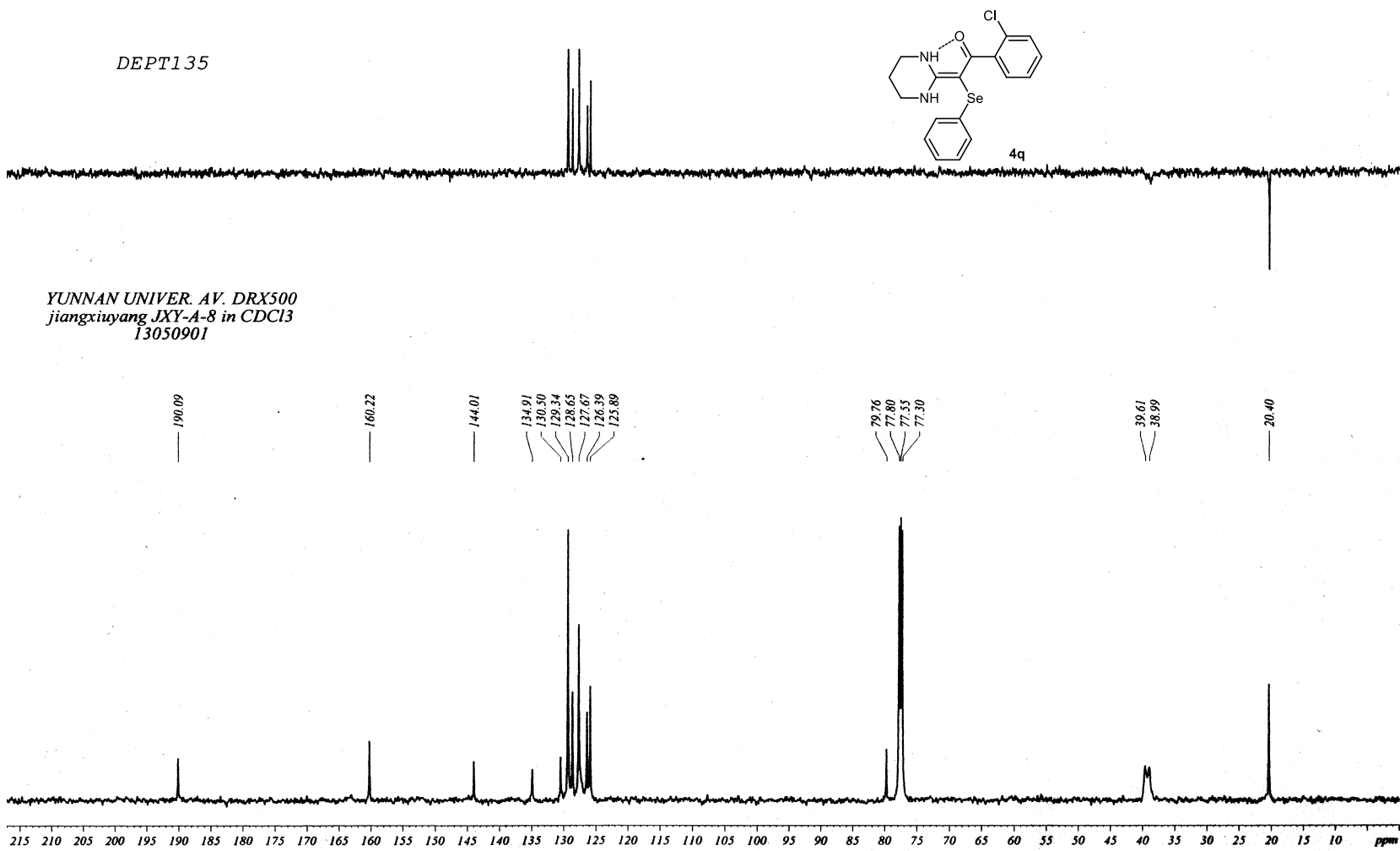


Figure S68. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 4q

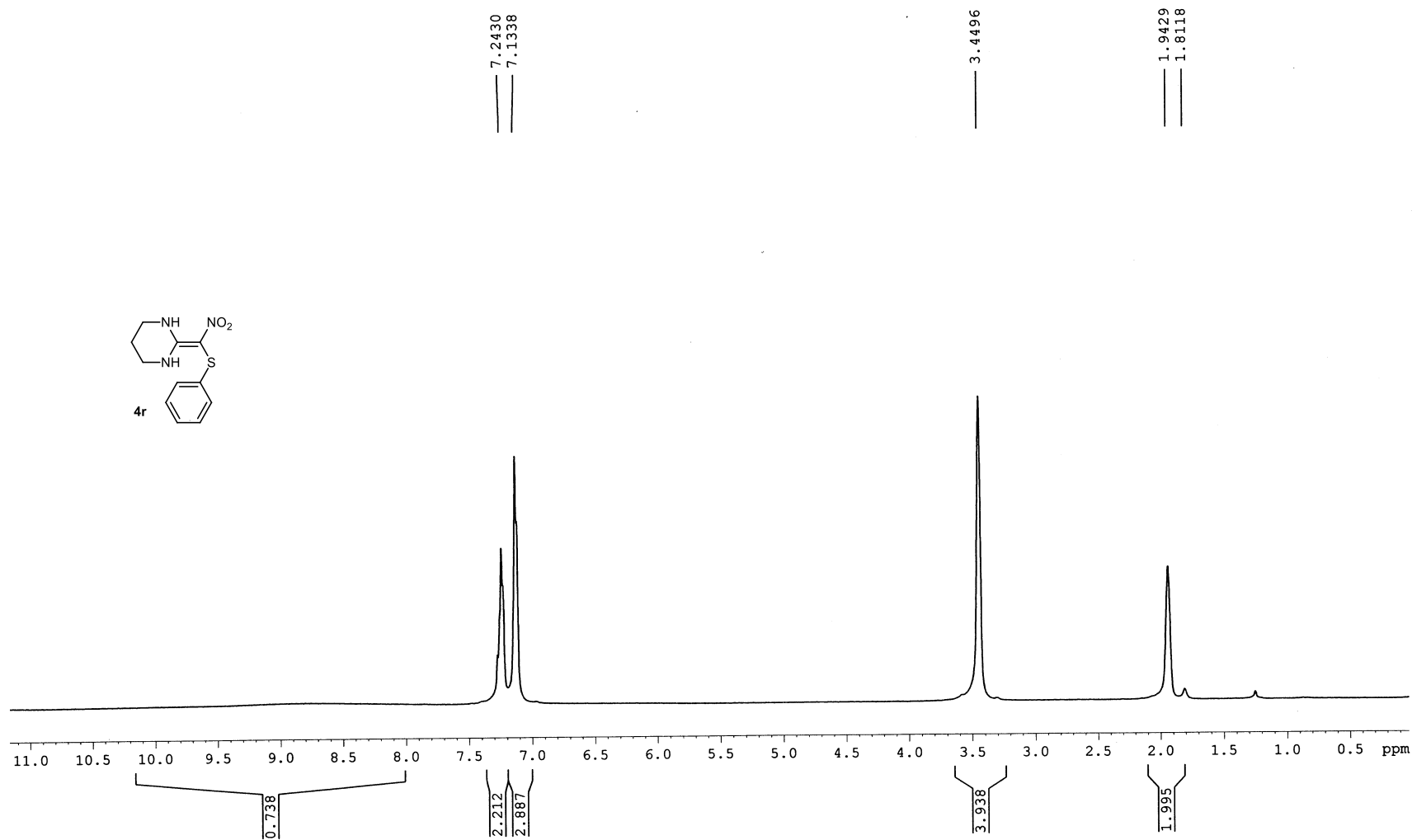


Figure S69. ¹H NMR (500 MHz, CDCl₃) spectra of compound **4r**

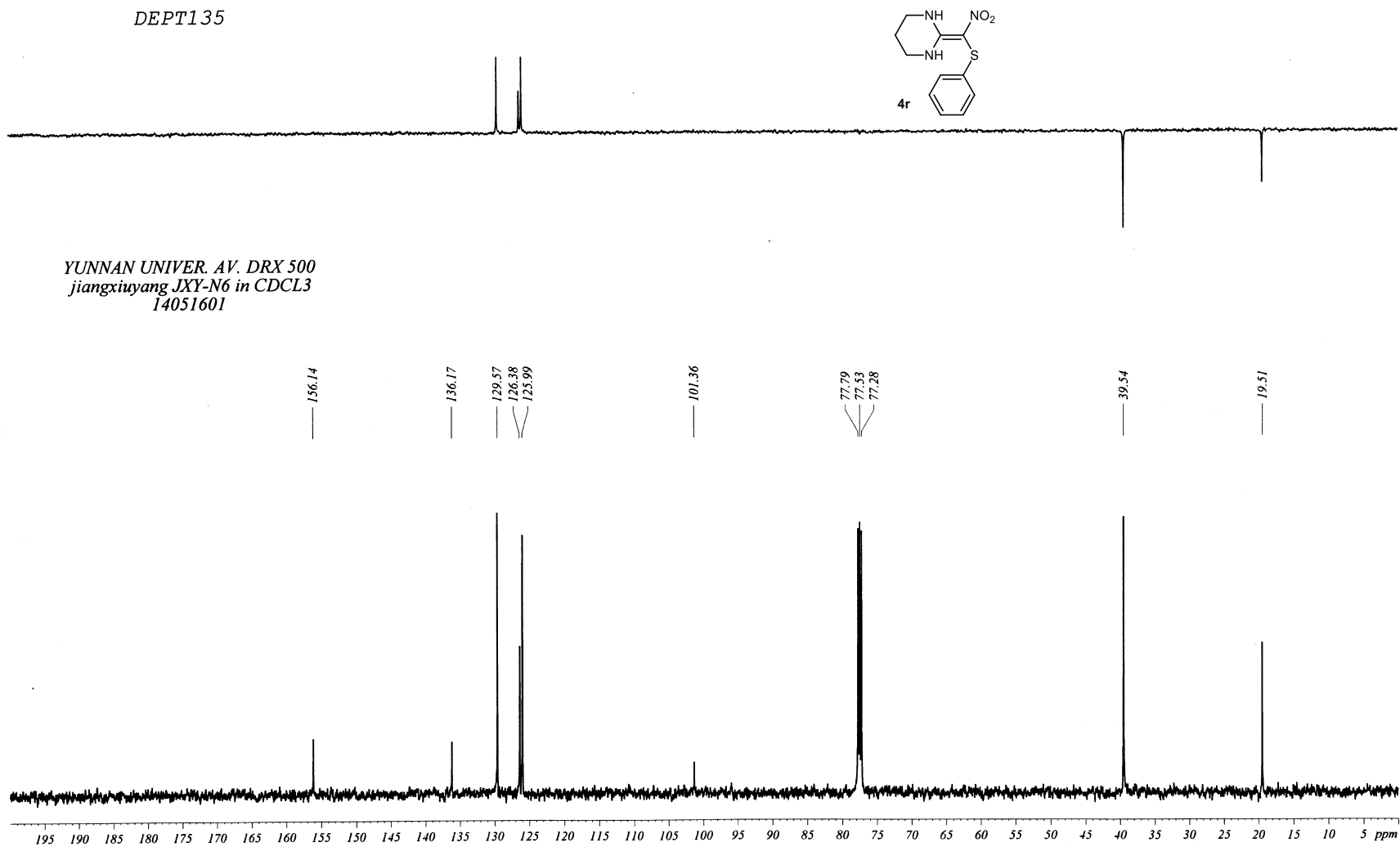


Figure S70. ^{13}C NMR (125 MHz, CDCl_3) spectra of compound **4r**

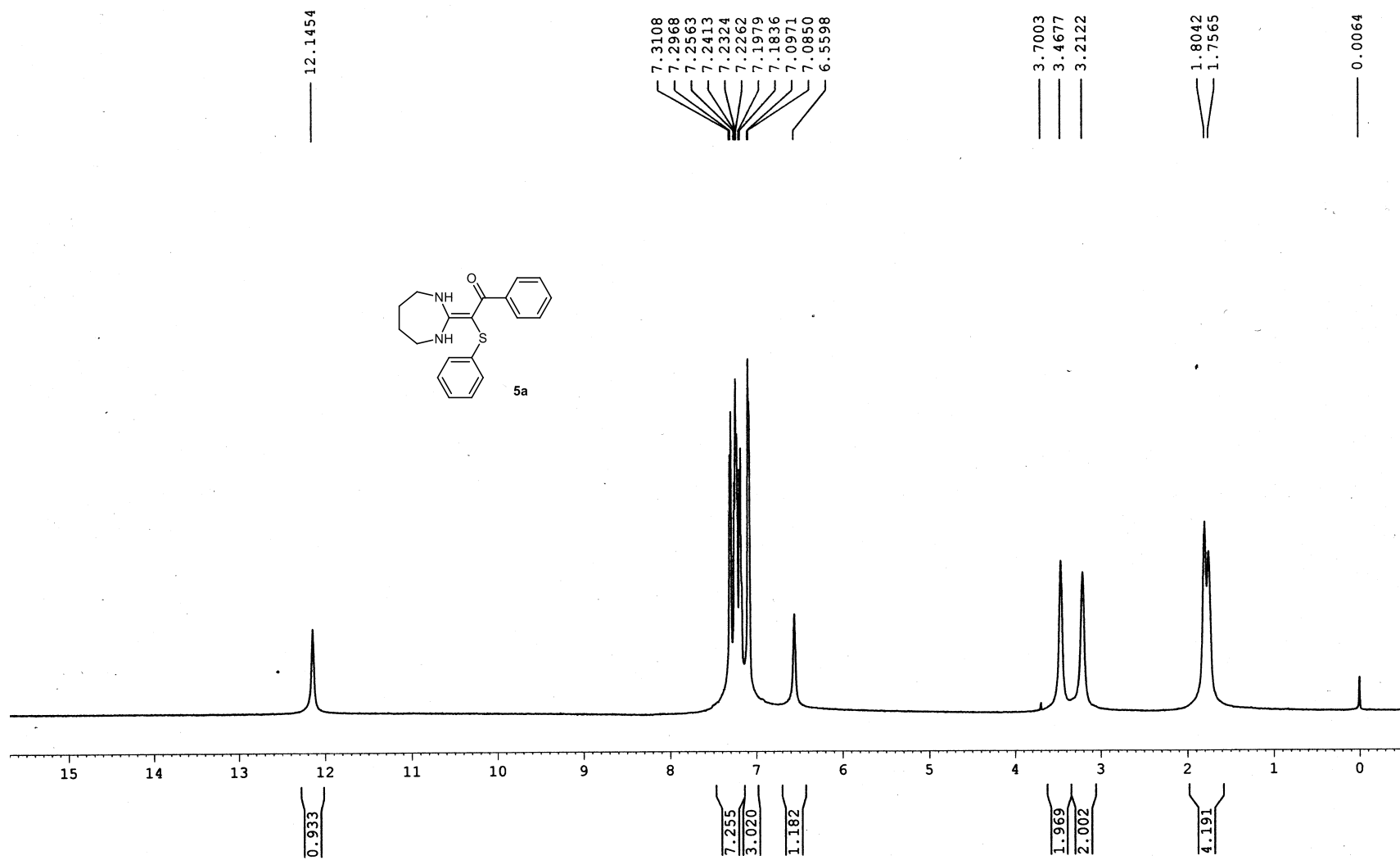


Figure S71. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5a**

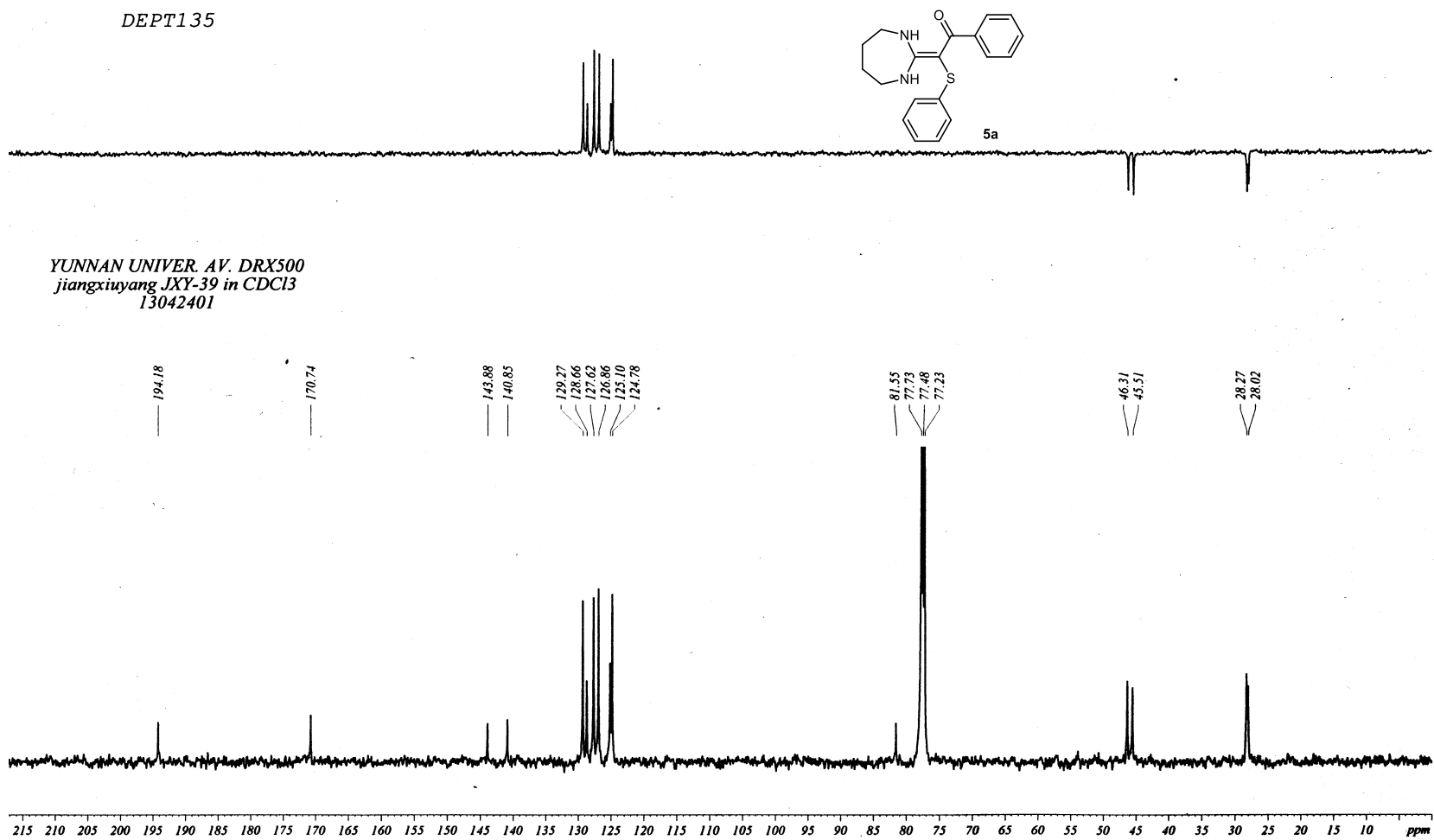
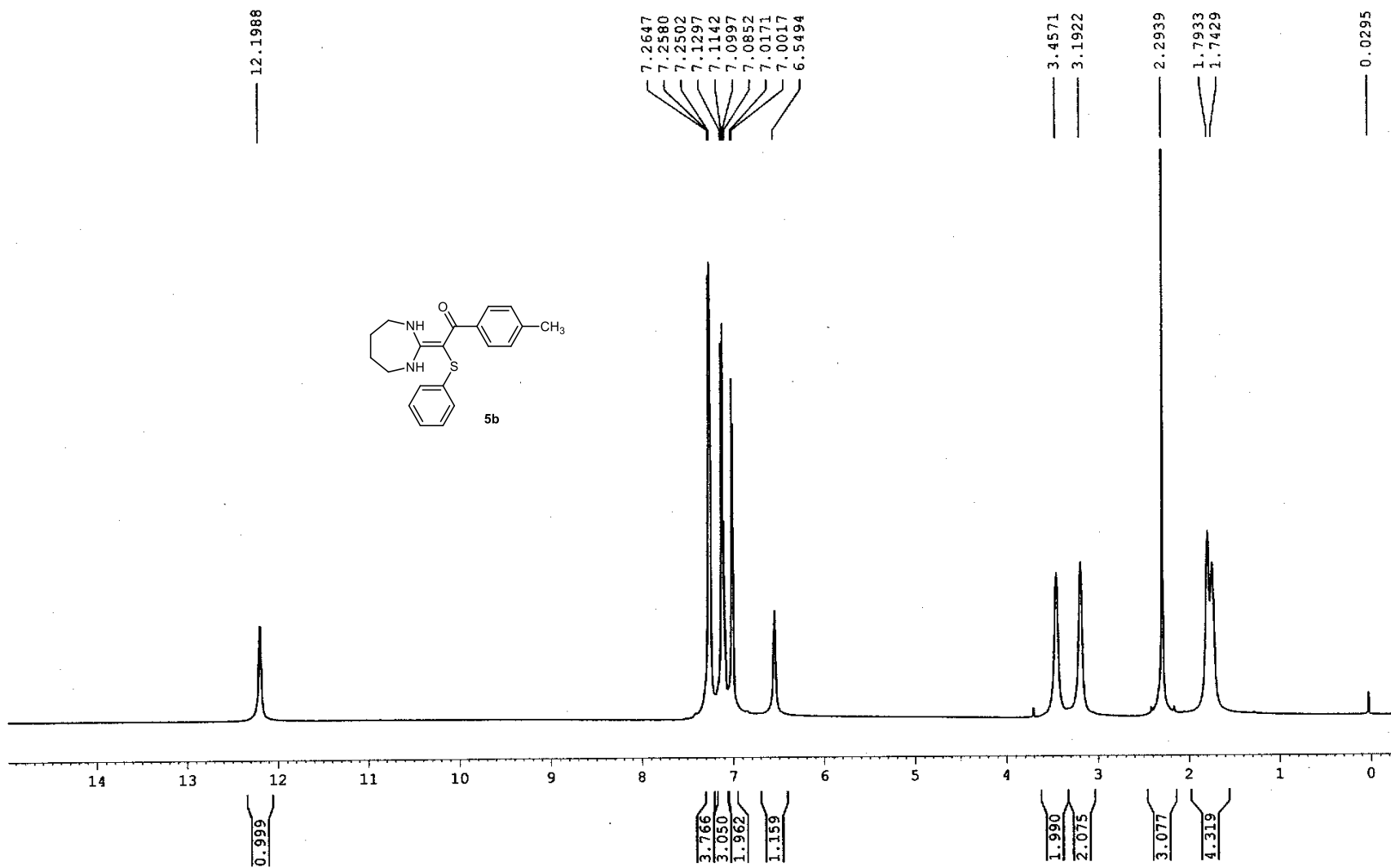


Figure S72. ^{13}C NMR (125 MHz, CDCl_3) spectra of compound **5a**



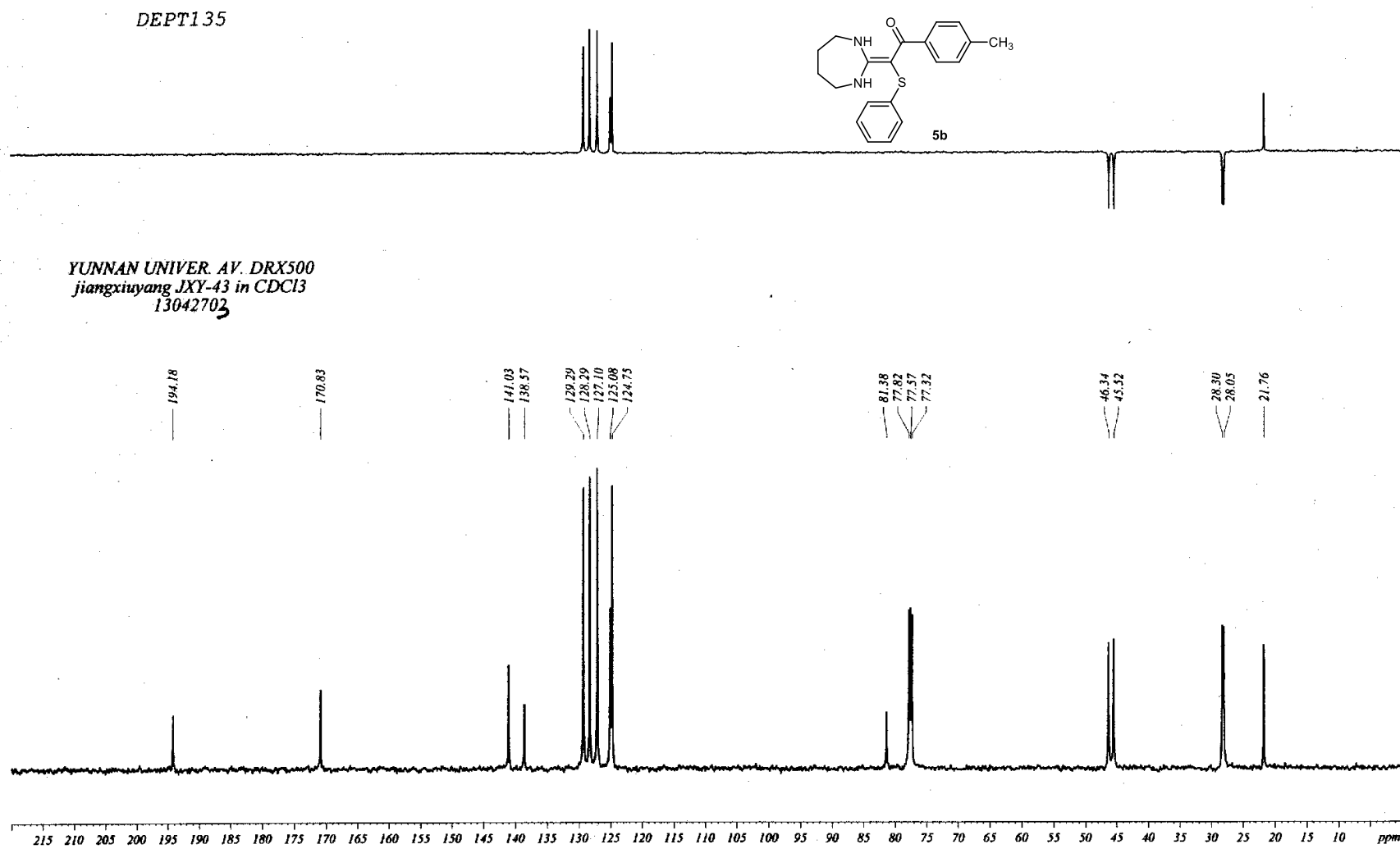


Figure S74. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5b

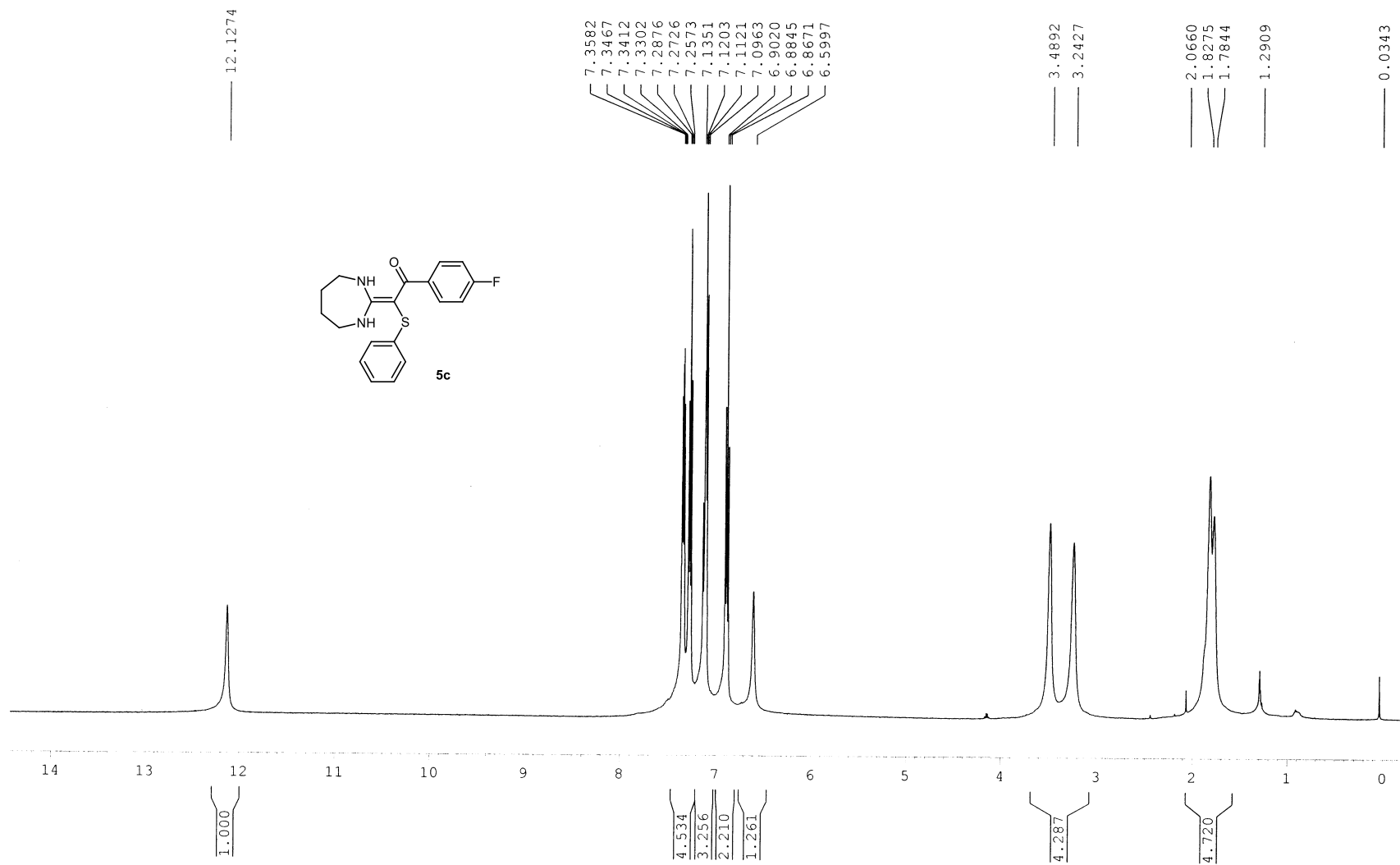
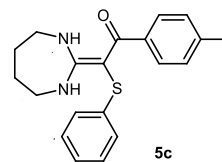


Figure S75. ^1H NMR (500 MHz, CDCl_3) spectra of compound **5c**

DEPT135



YUNNAN UNIVER. AV. DRX500
jiangxiuyang JXY-42 in CDCl₃
13042403

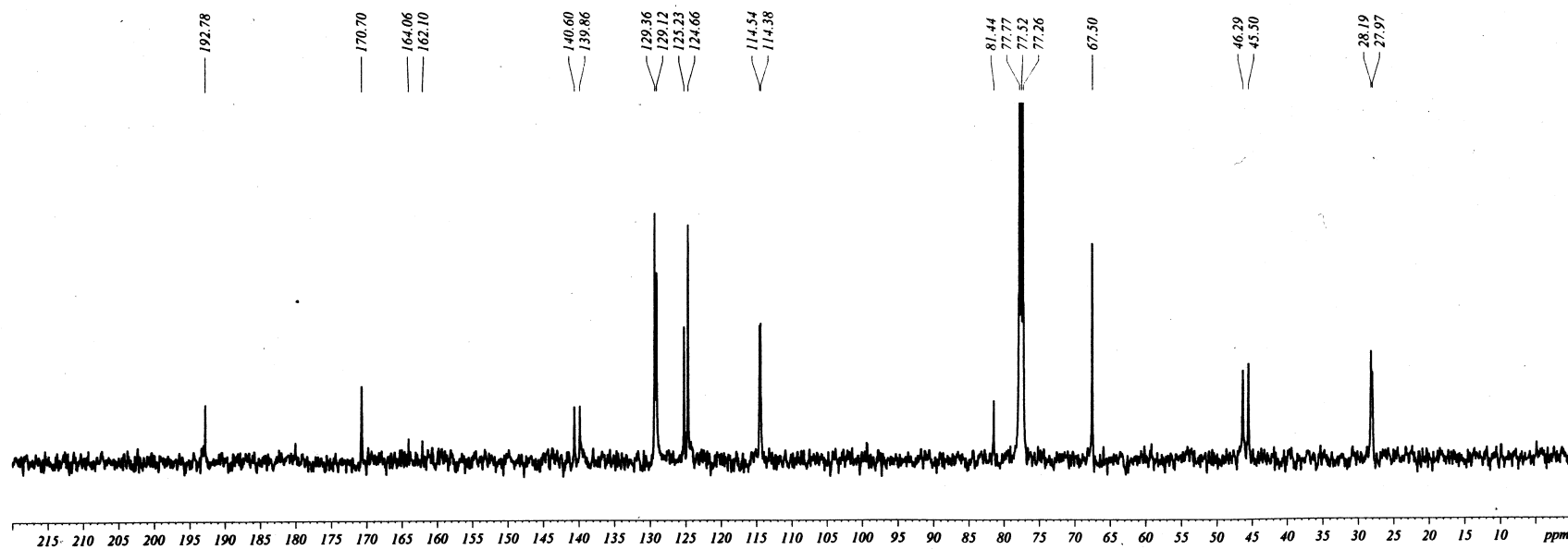


Figure S76. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5c

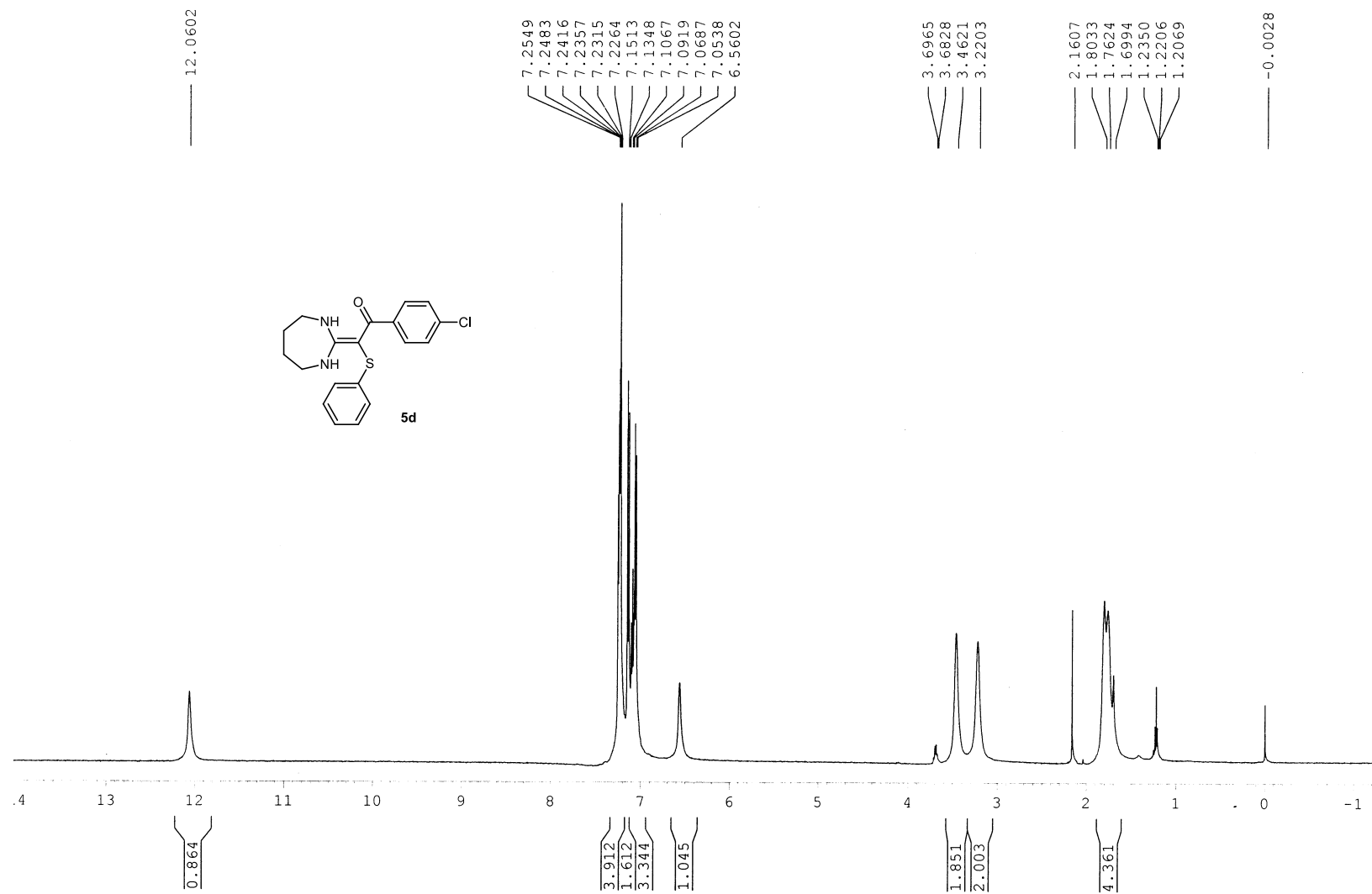
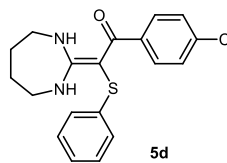


Figure S77. ^1H NMR (500 MHz, CDCl_3) spectra of compound **5d**

DEPT135



YUNNAN UNIVER. AV. DRX500
jiangxiyang JXY-41 in CDCl₃
13052902

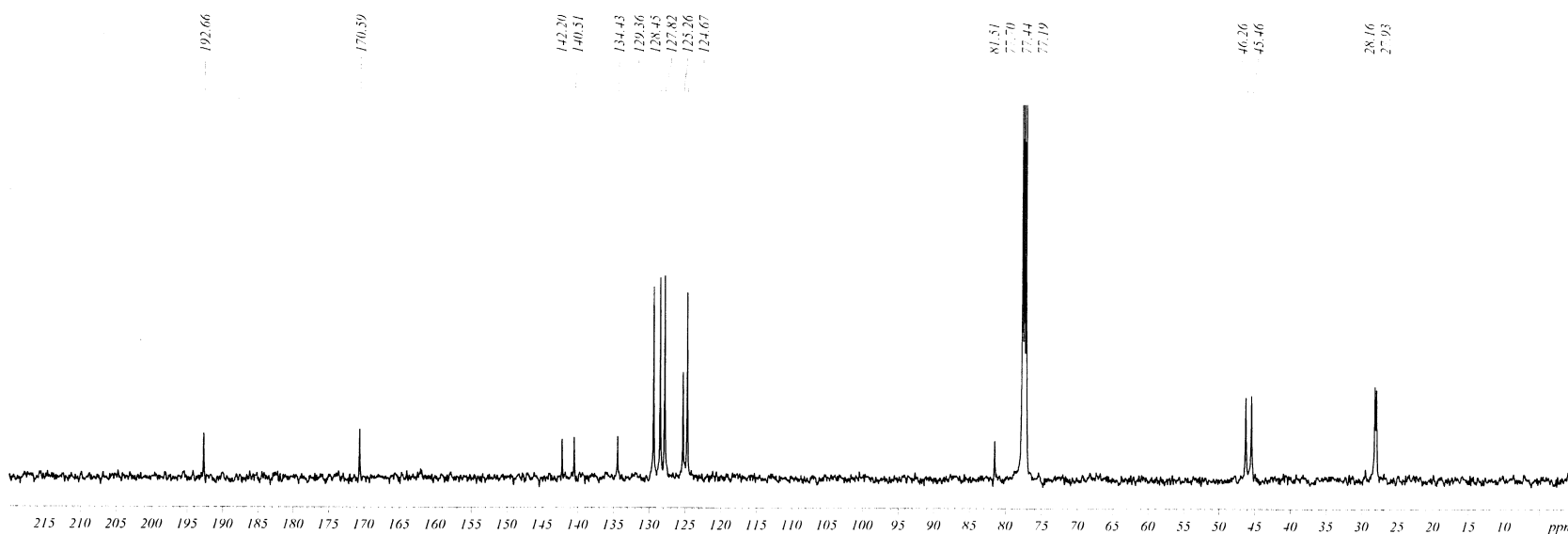
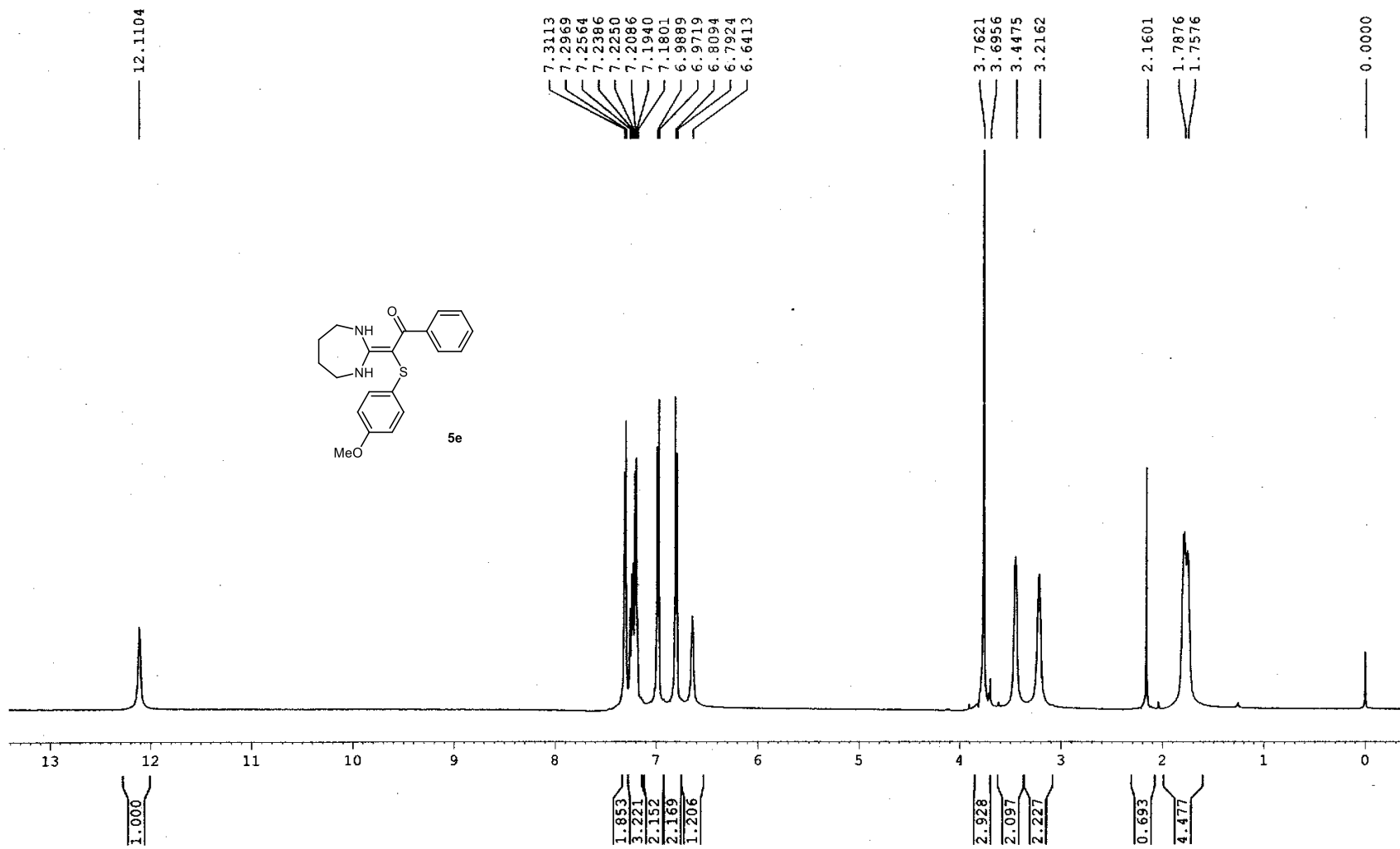


Figure S78. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5d



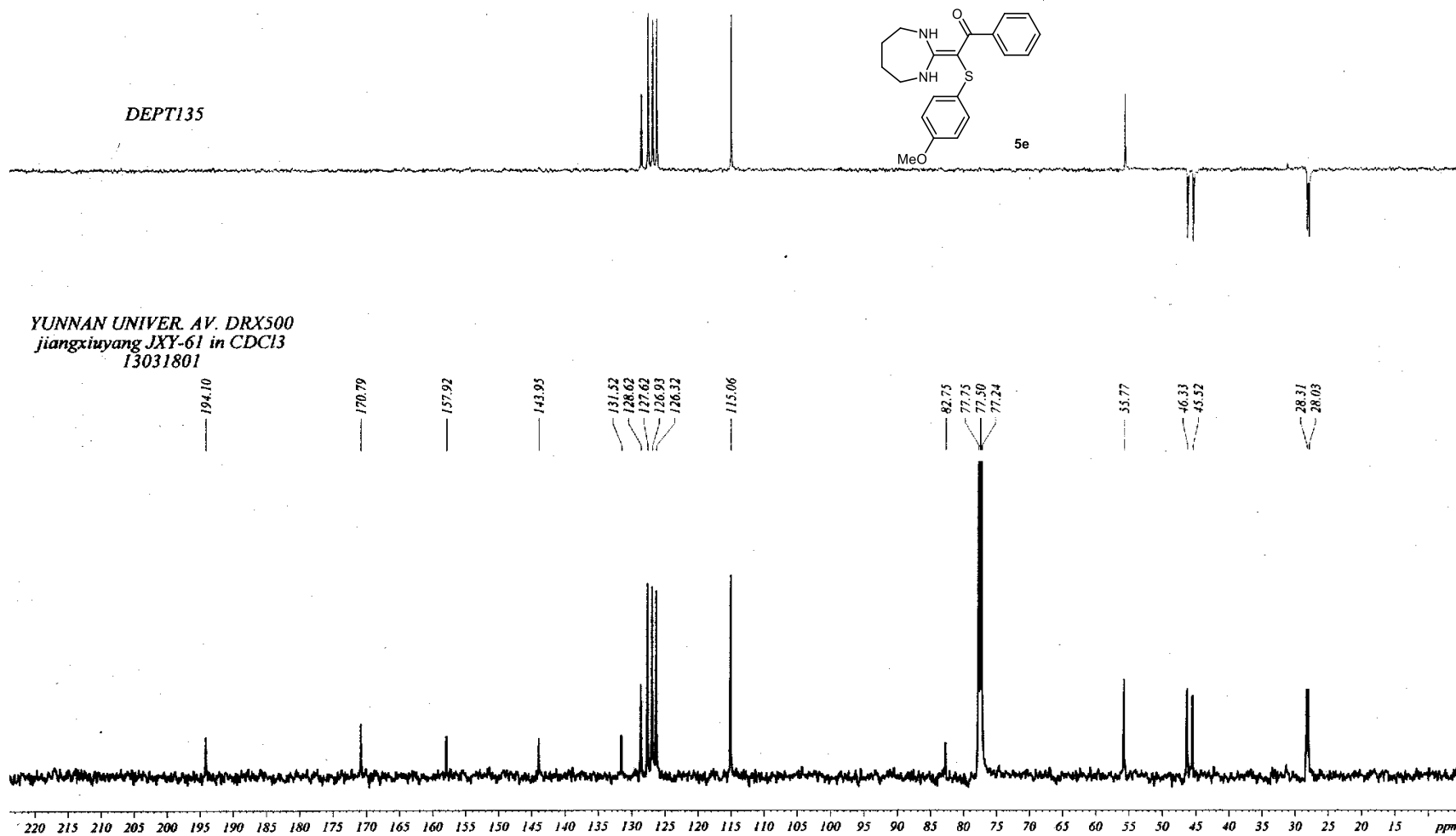


Figure S80. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5e

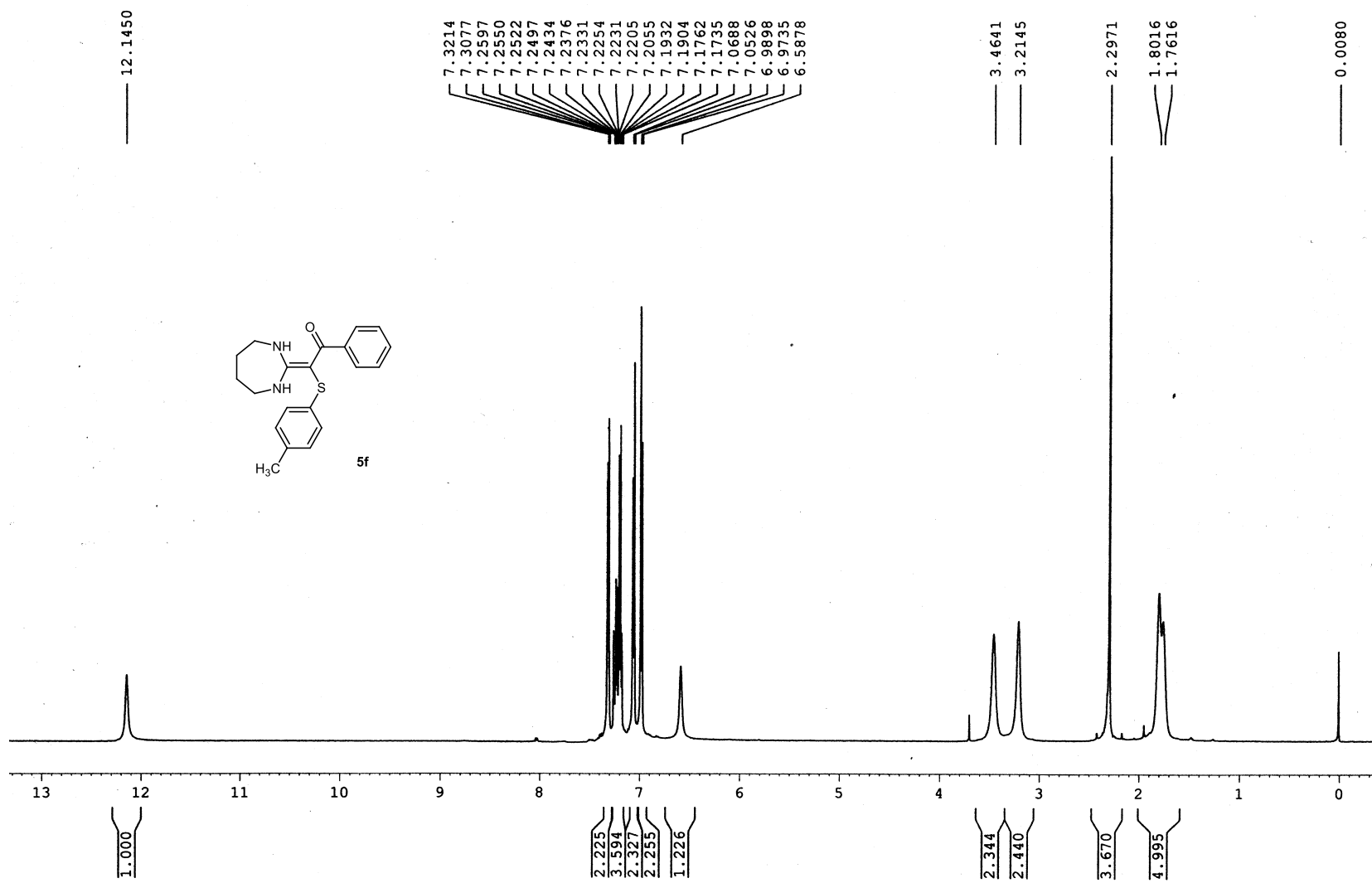


Figure S81. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5f**

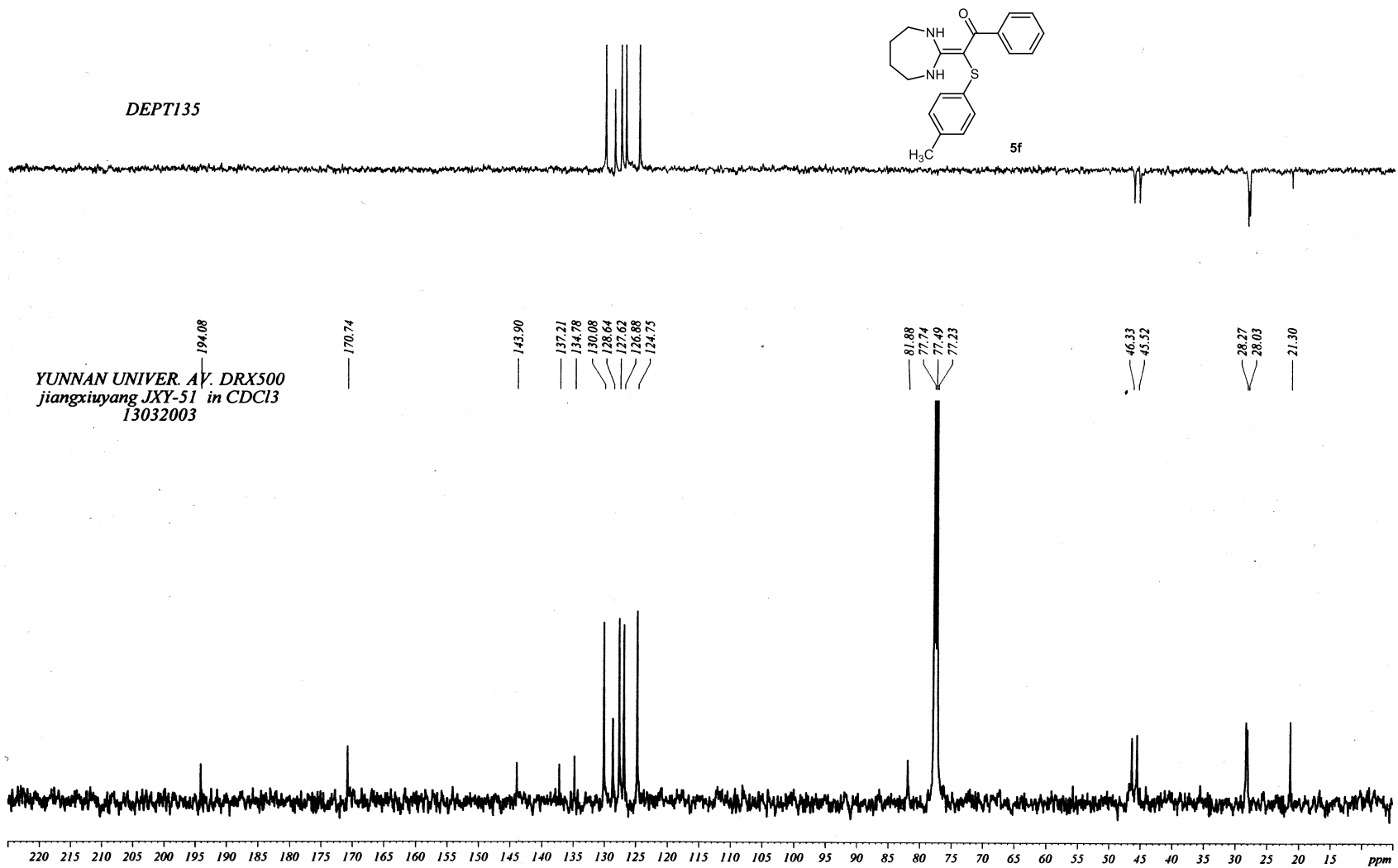


Figure S82. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5f

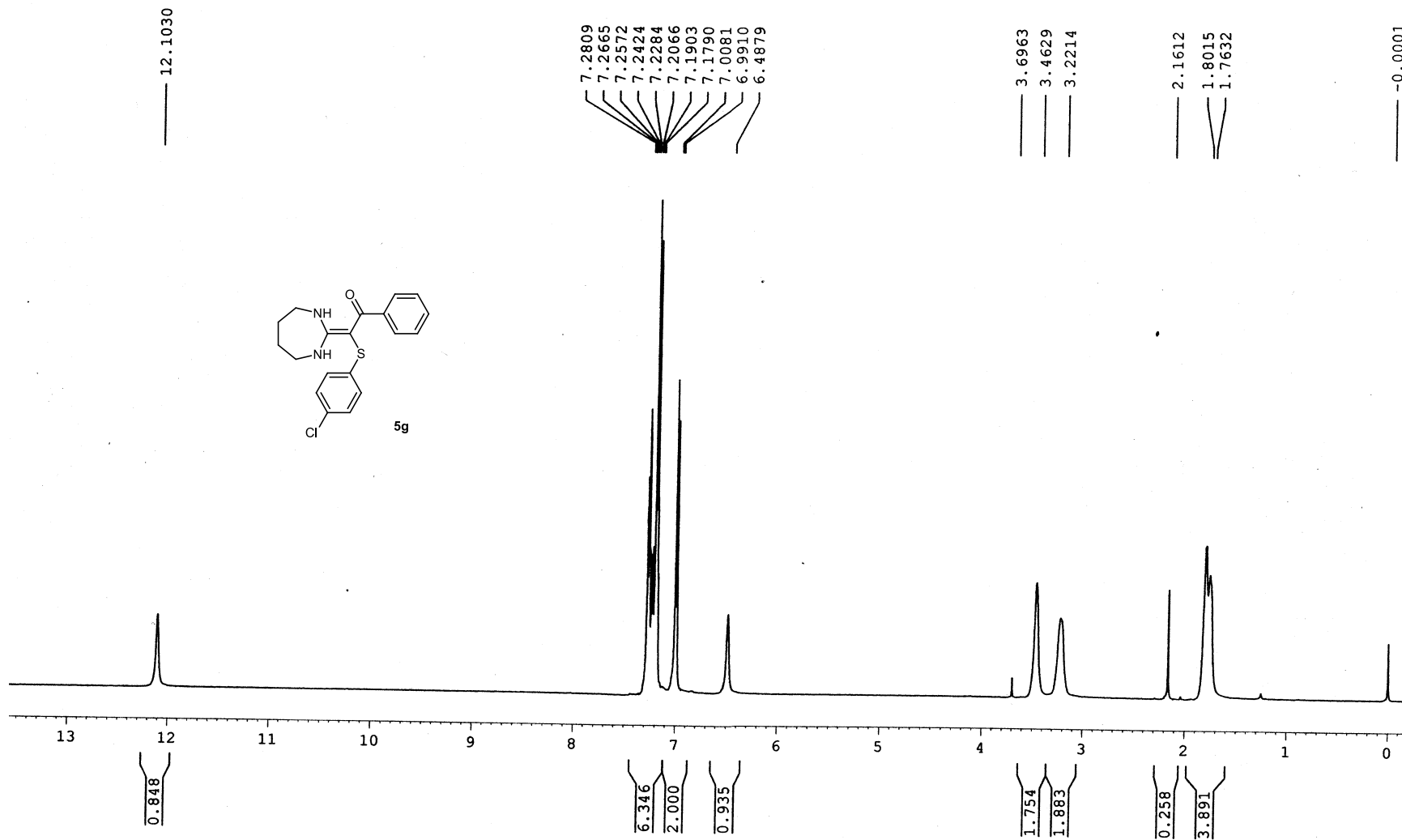


Figure S83. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5g**

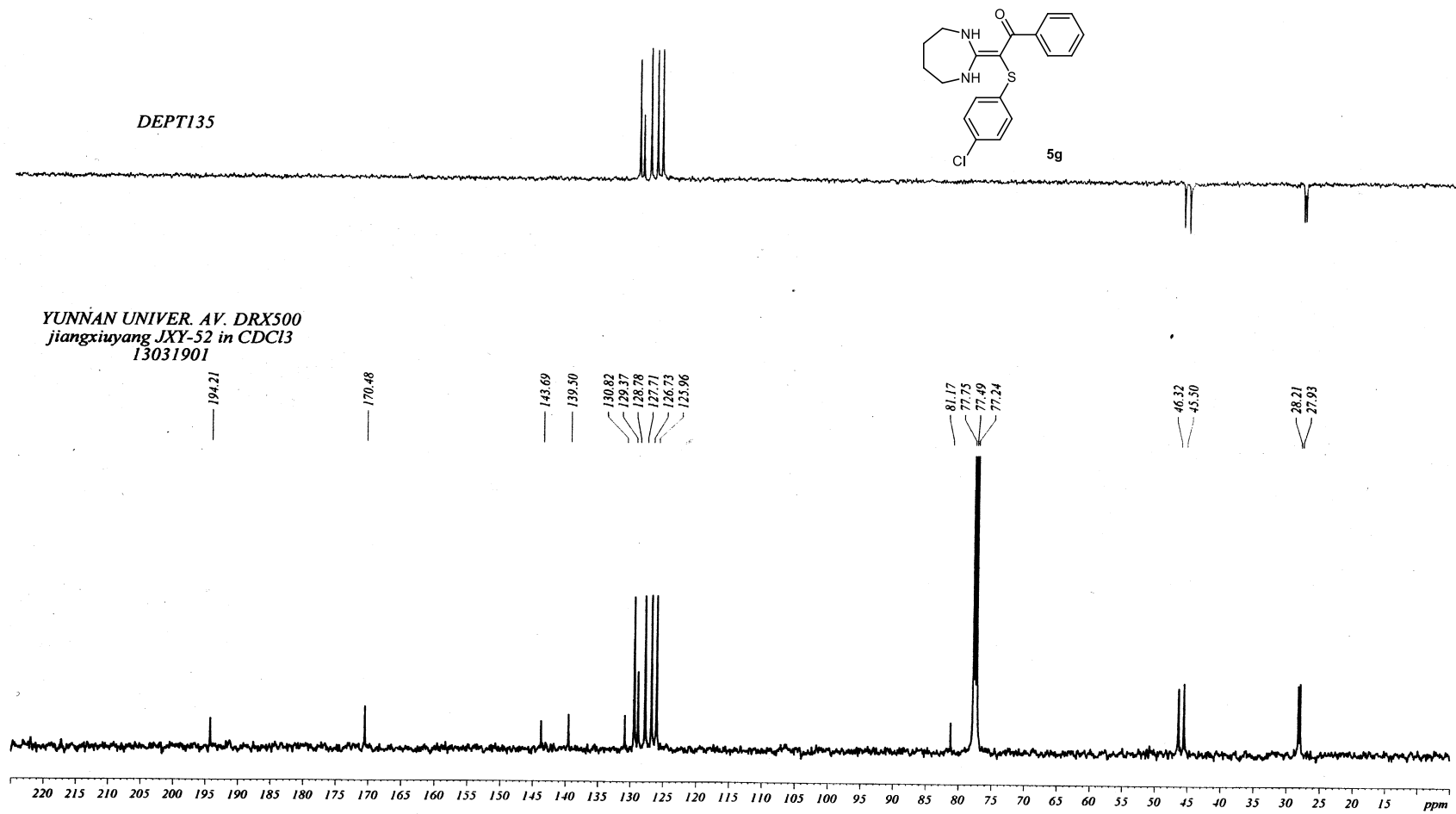


Figure S84. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **5g**

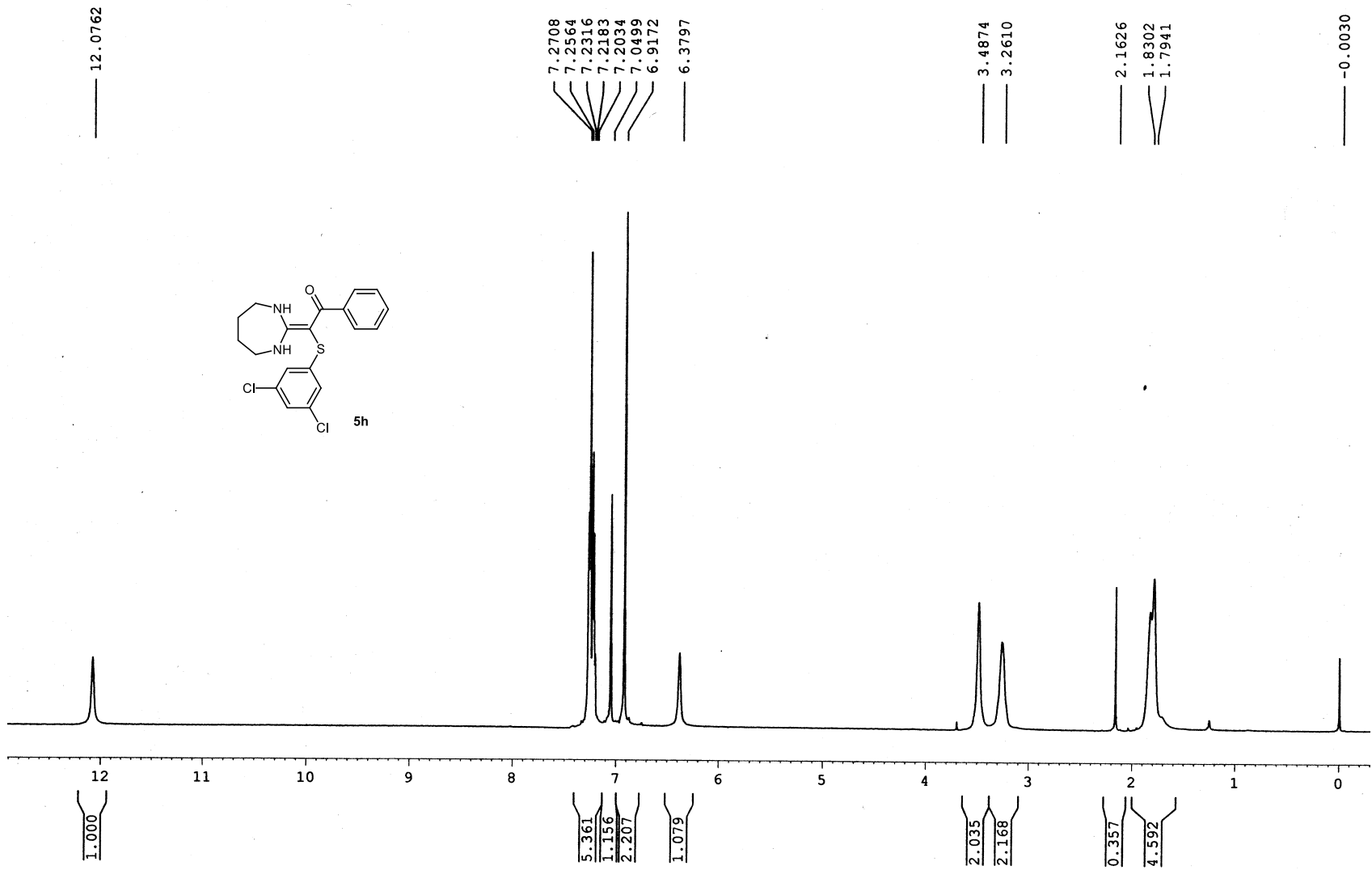


Figure S85. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5h**

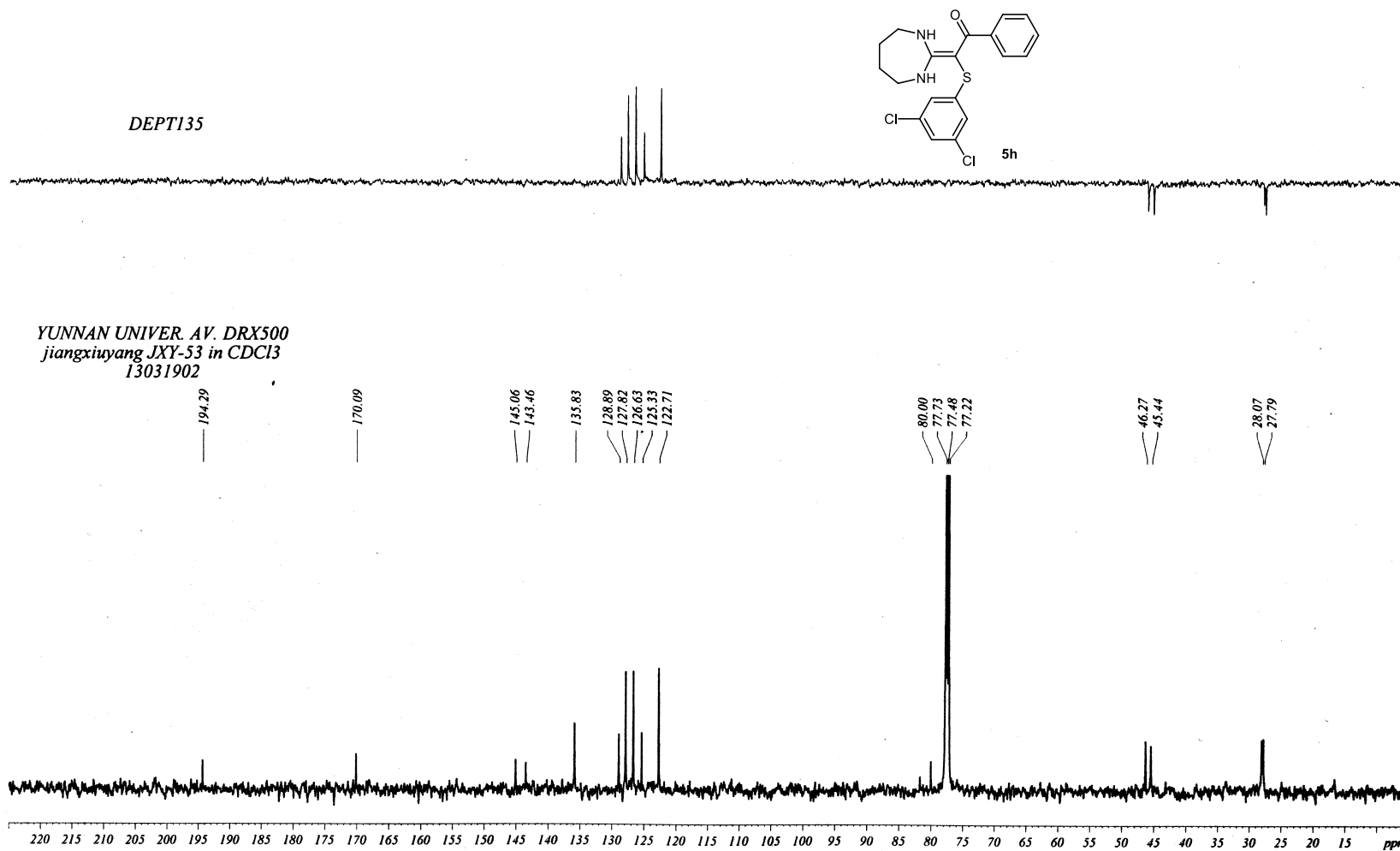


Figure S86. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **5h**

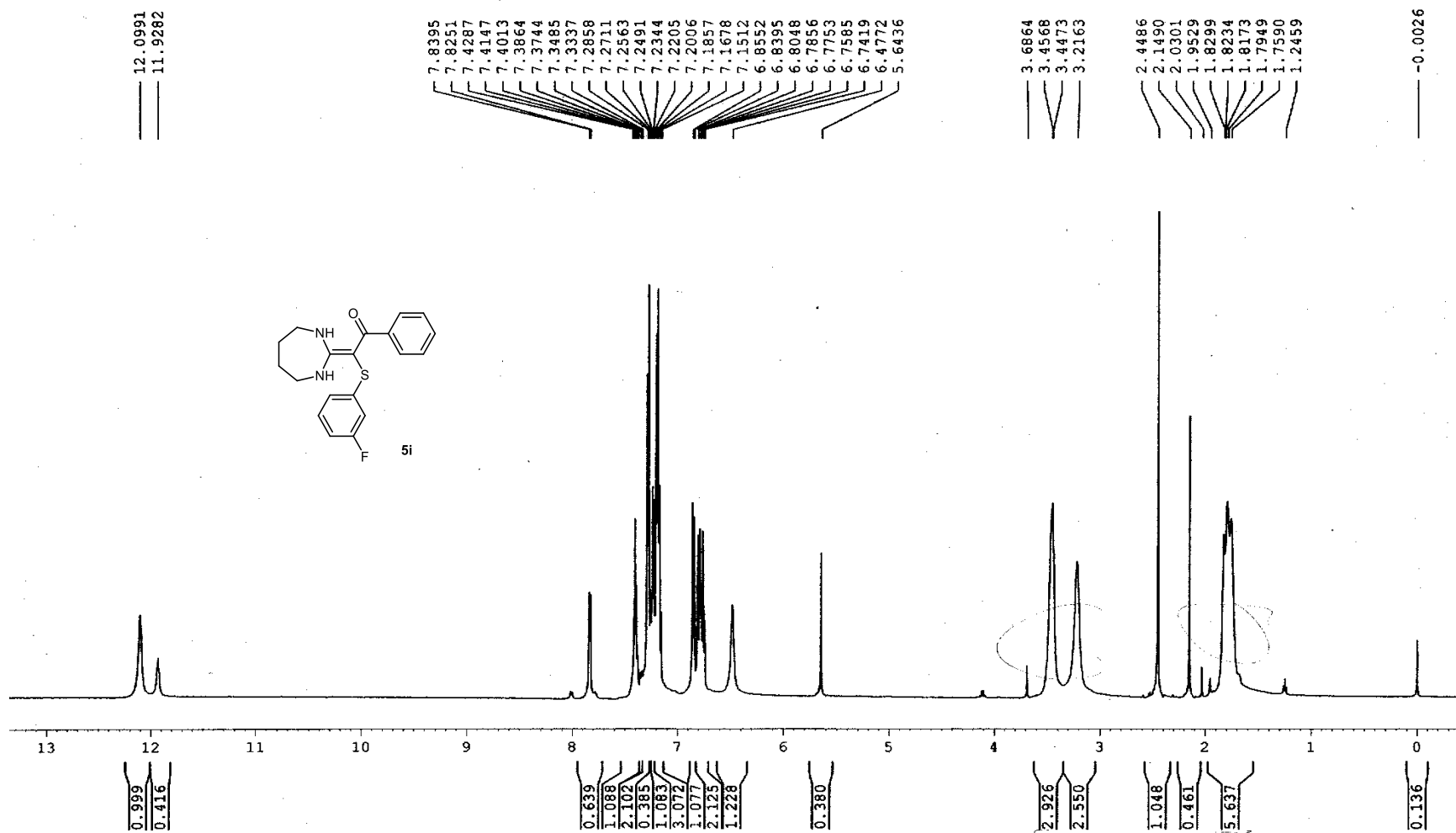


Figure S87. ^1H NMR (500 MHz, CDCl_3) spectra of compound **5i**

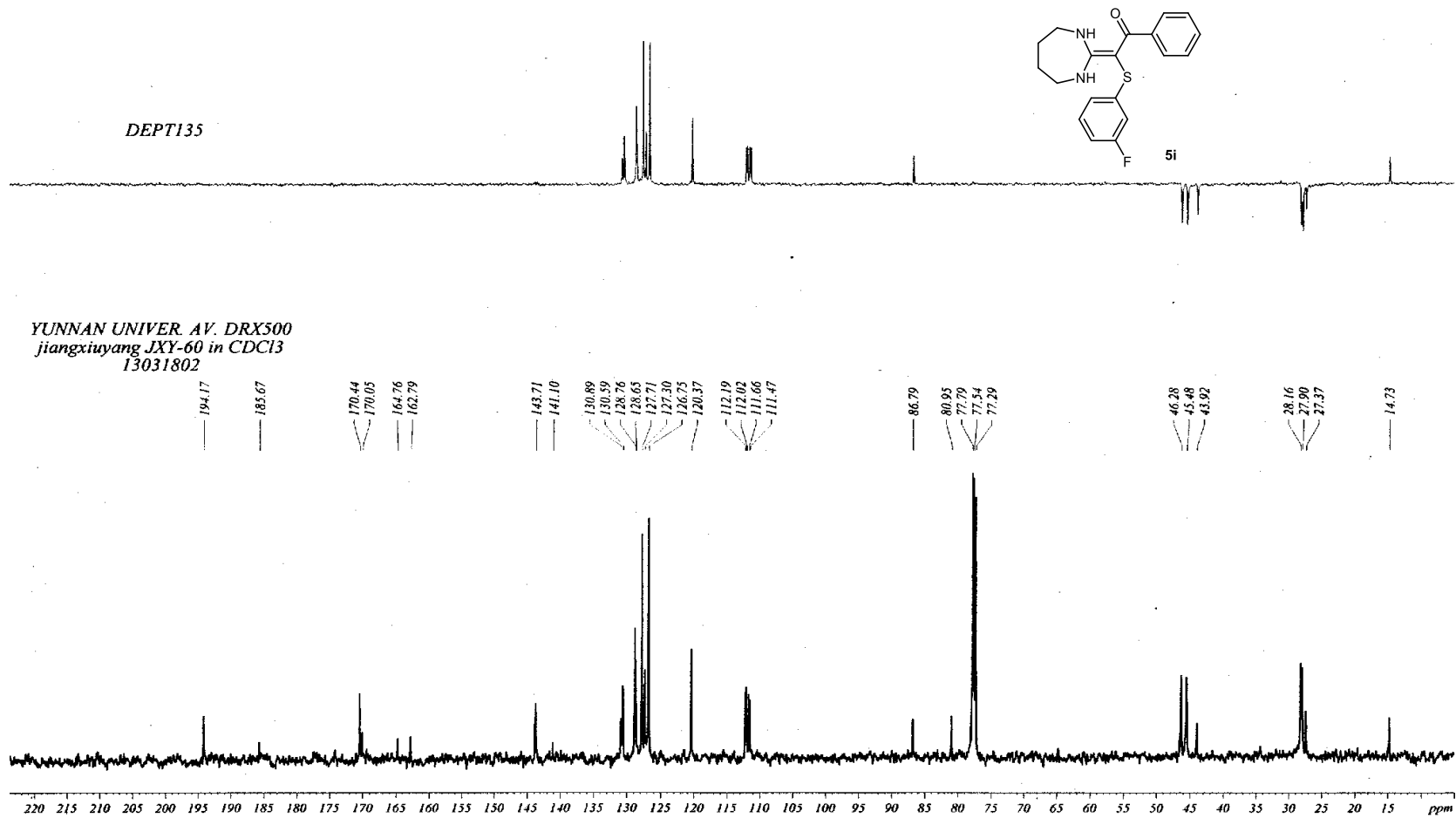


Figure S88. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **5i**

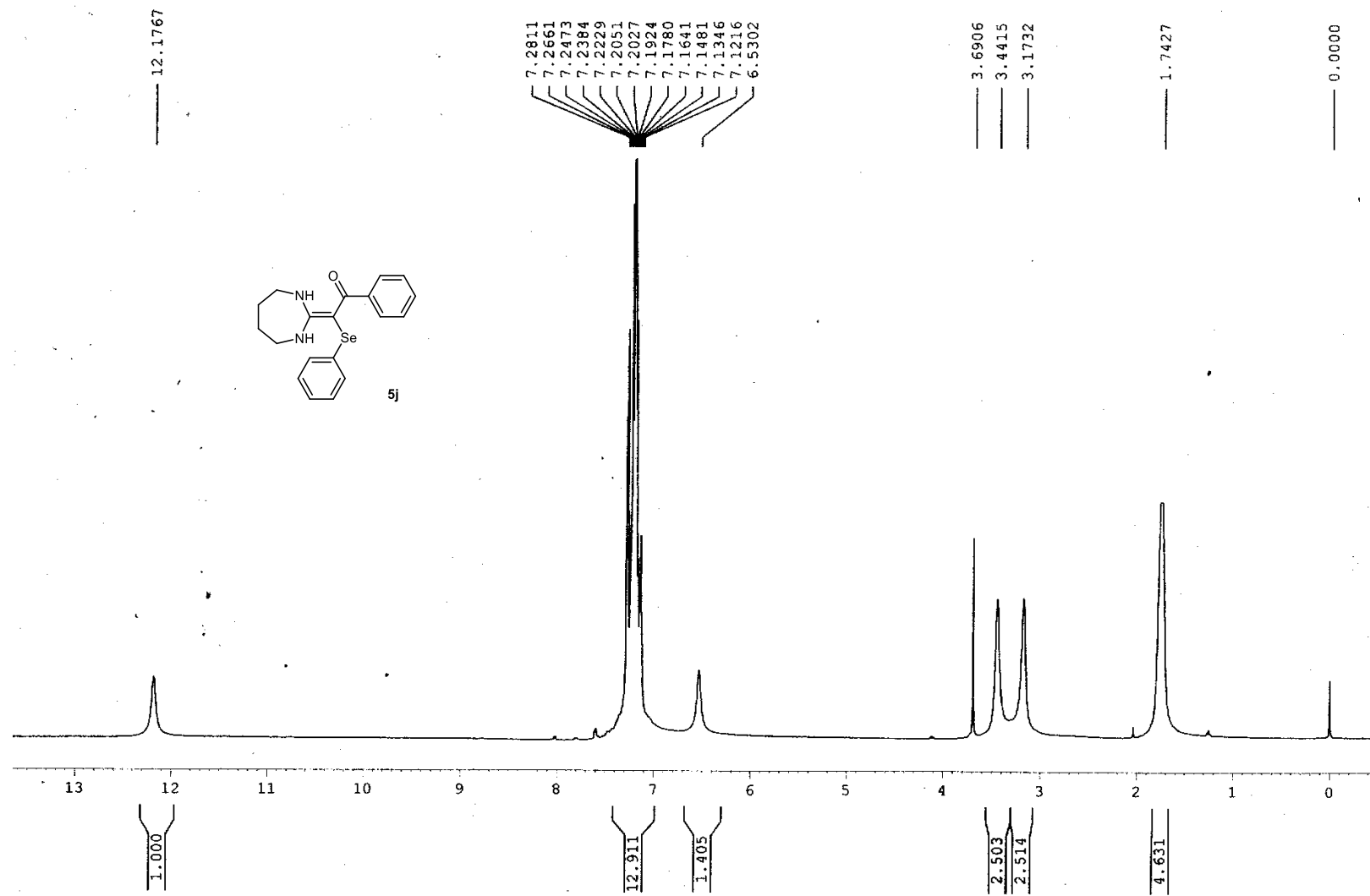


Figure S89. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5j**

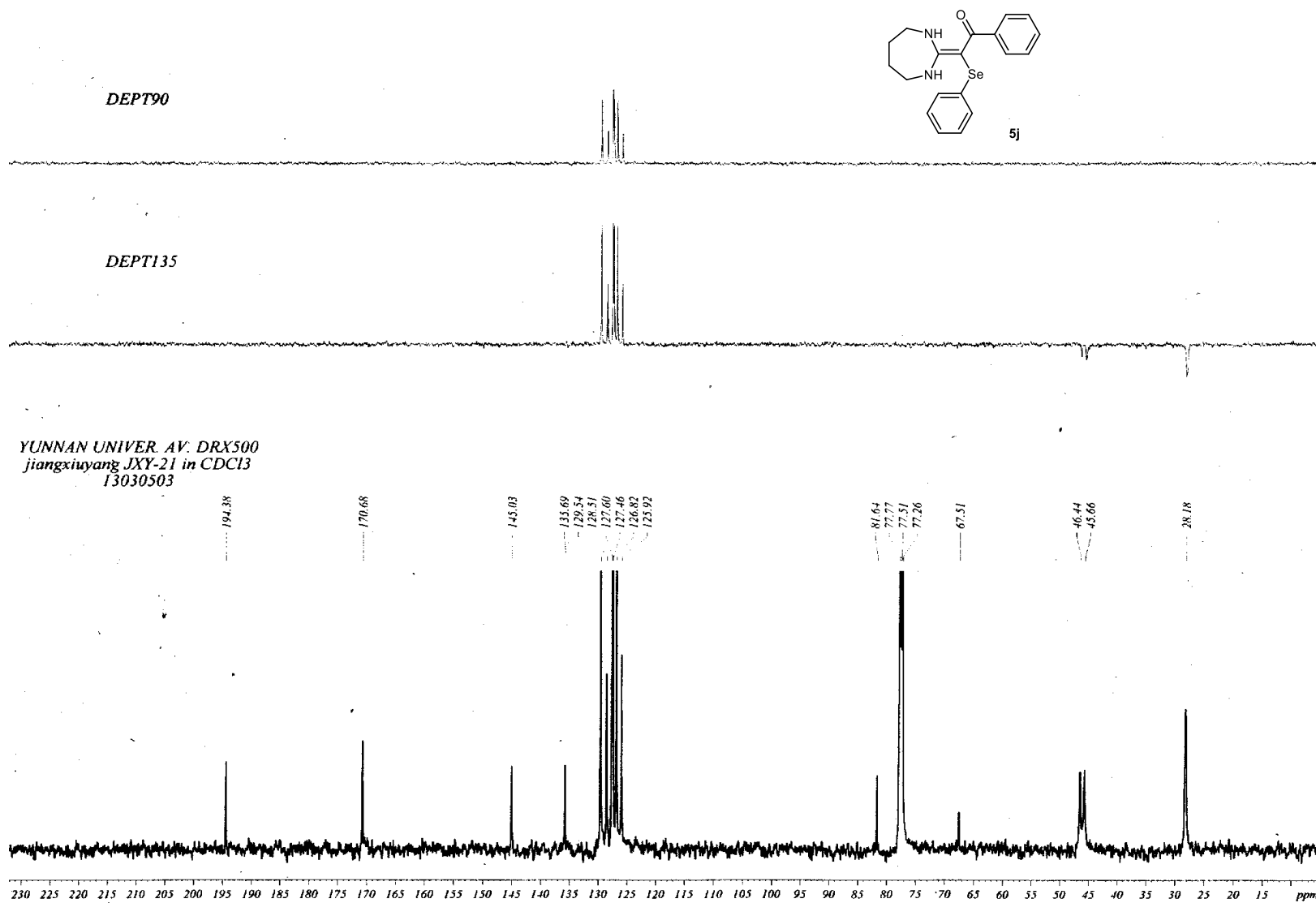


Figure S90. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5j

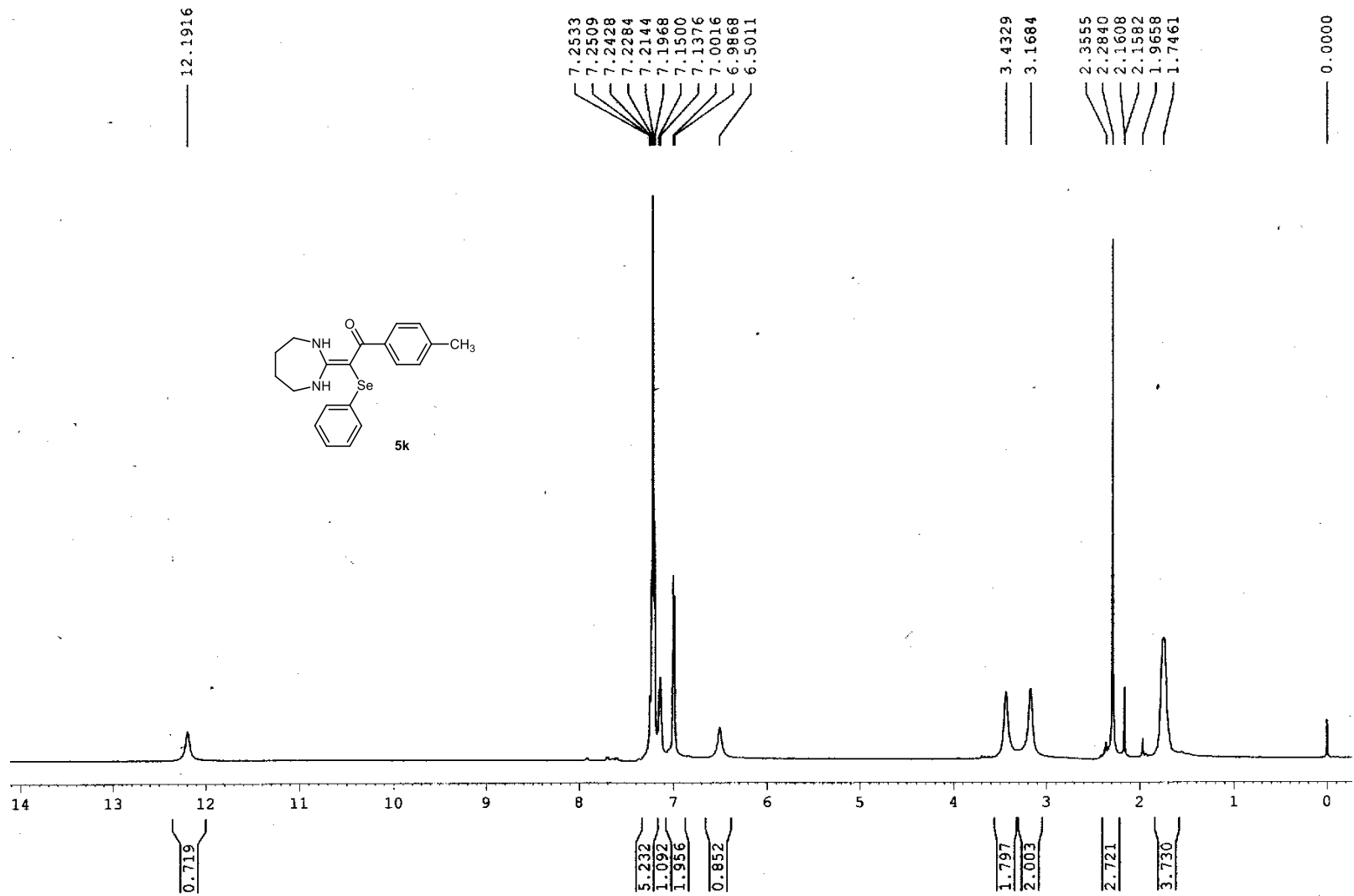


Figure S91. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5k**

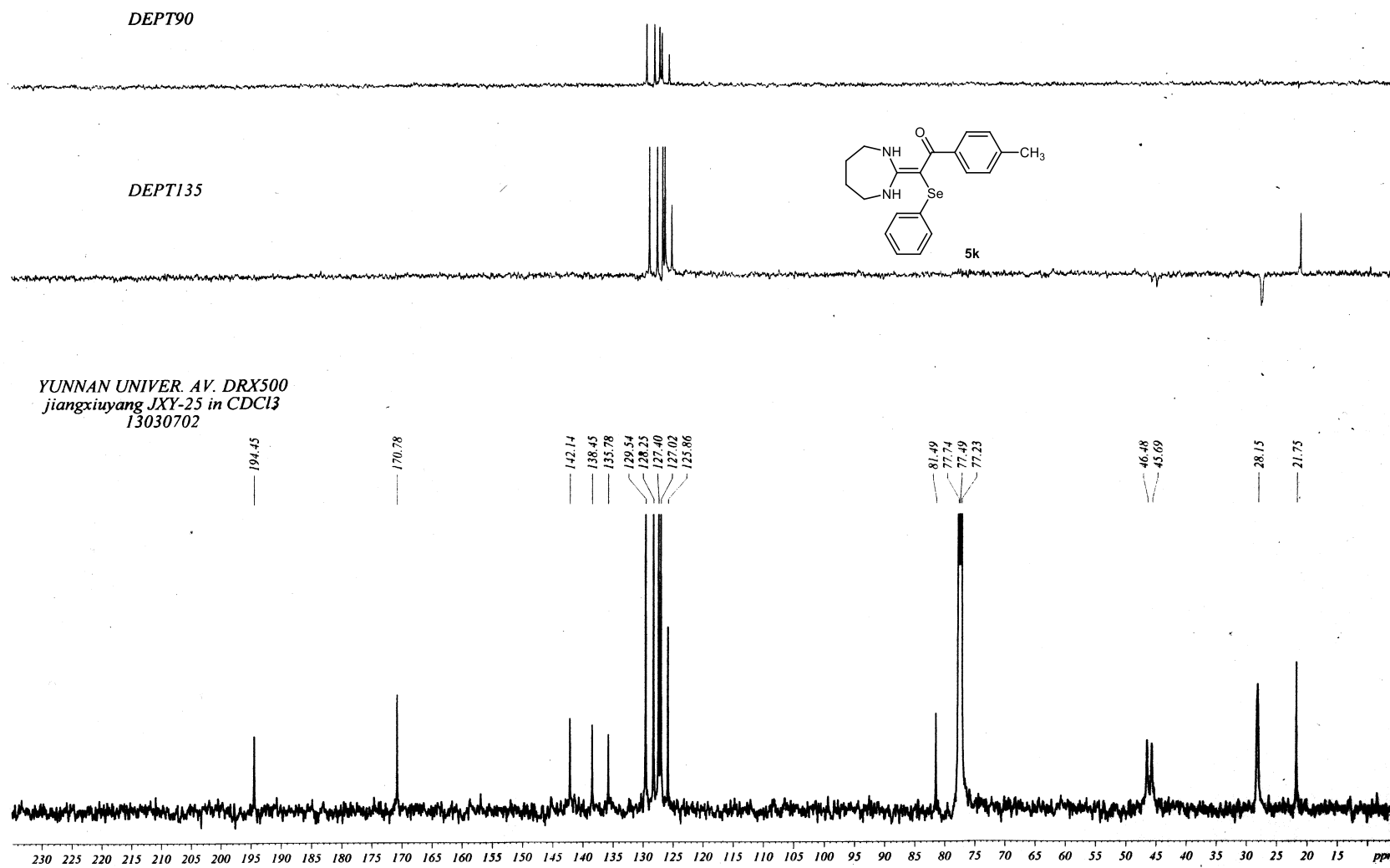


Figure S92. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 5k

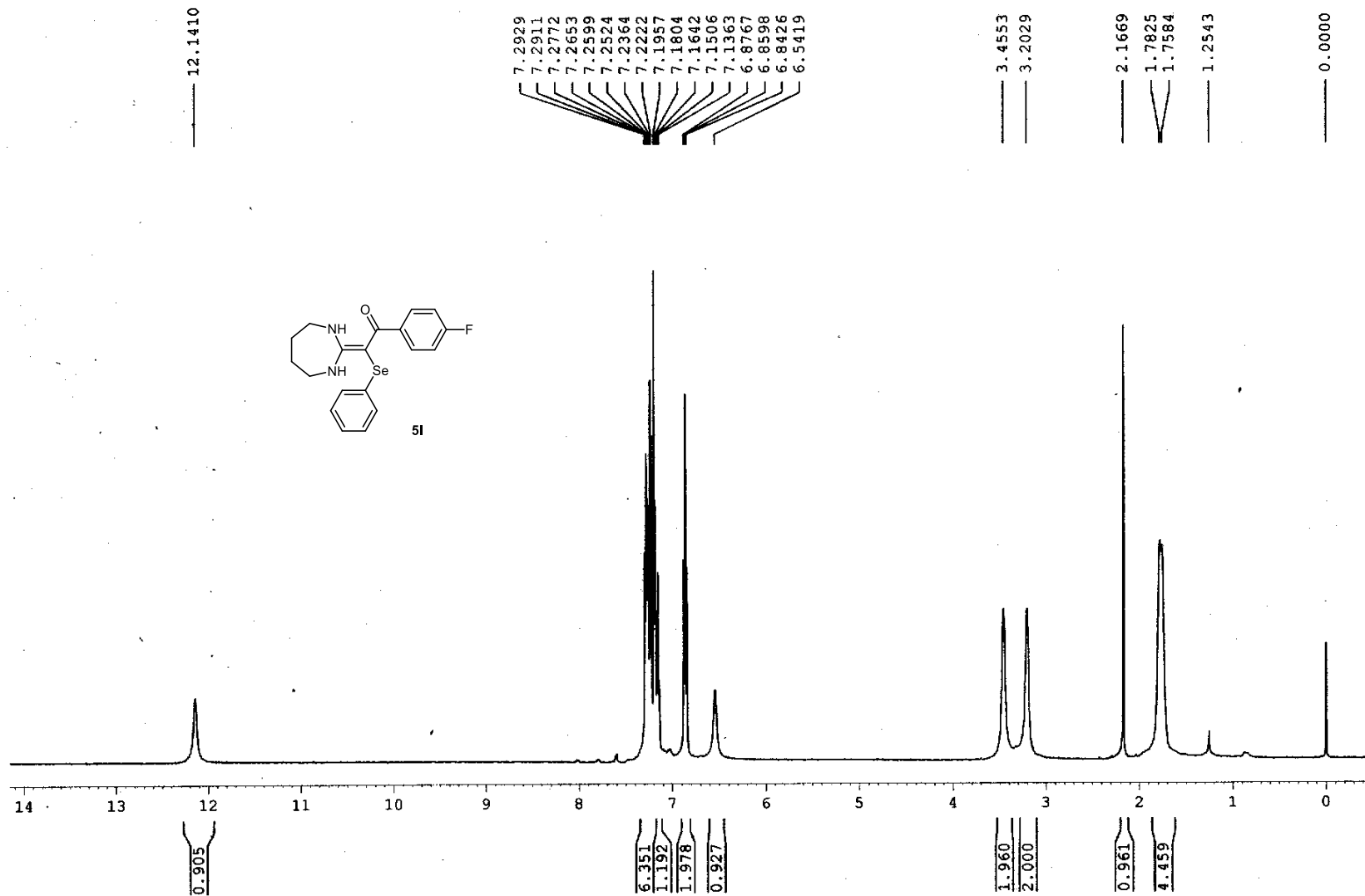


Figure S93. ^1H NMR (500 MHz, CDCl_3) spectra of compound **5l**

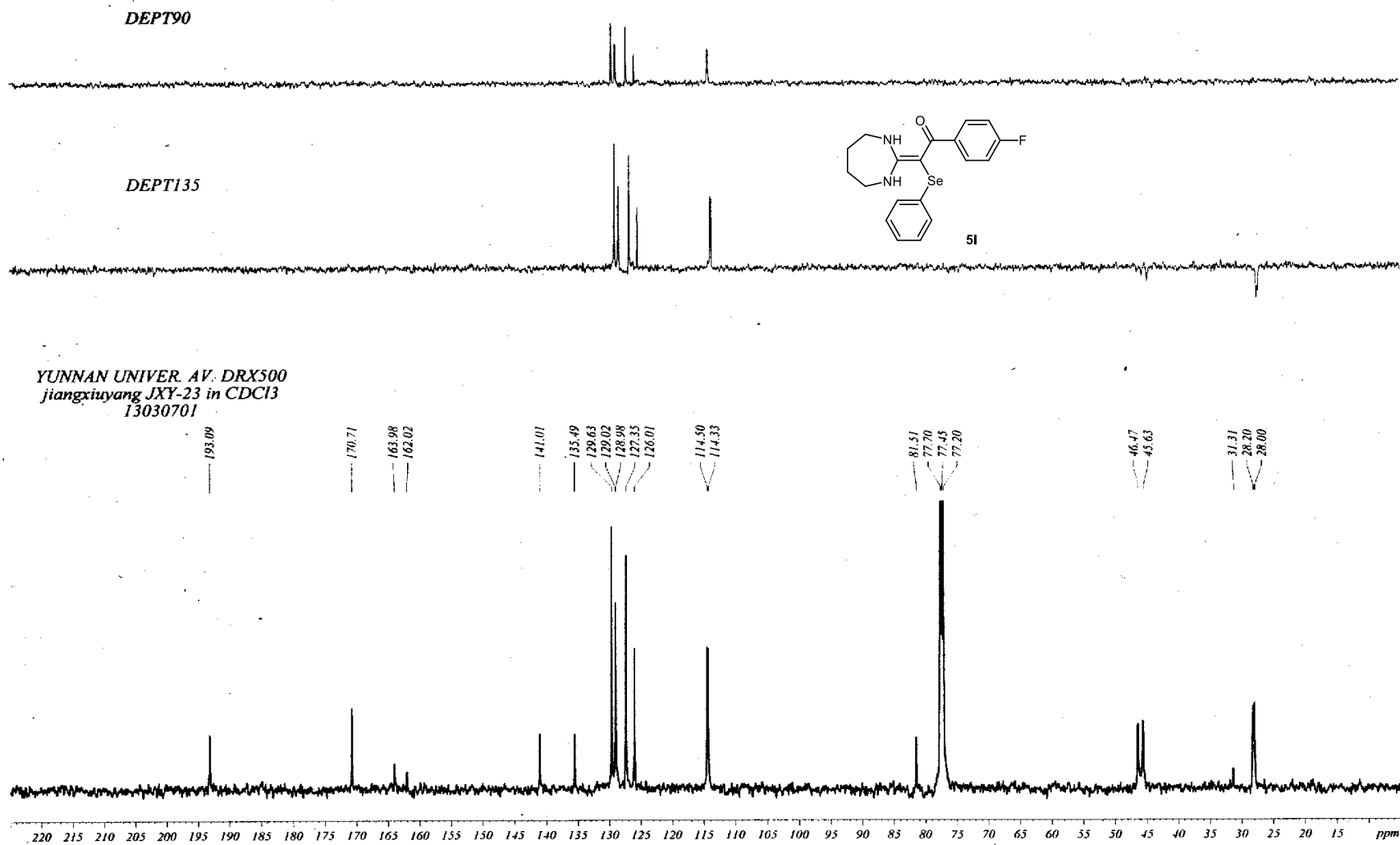


Figure S94. ¹³C NMR (125 MHz, CDCl₃) spectra of compound 51

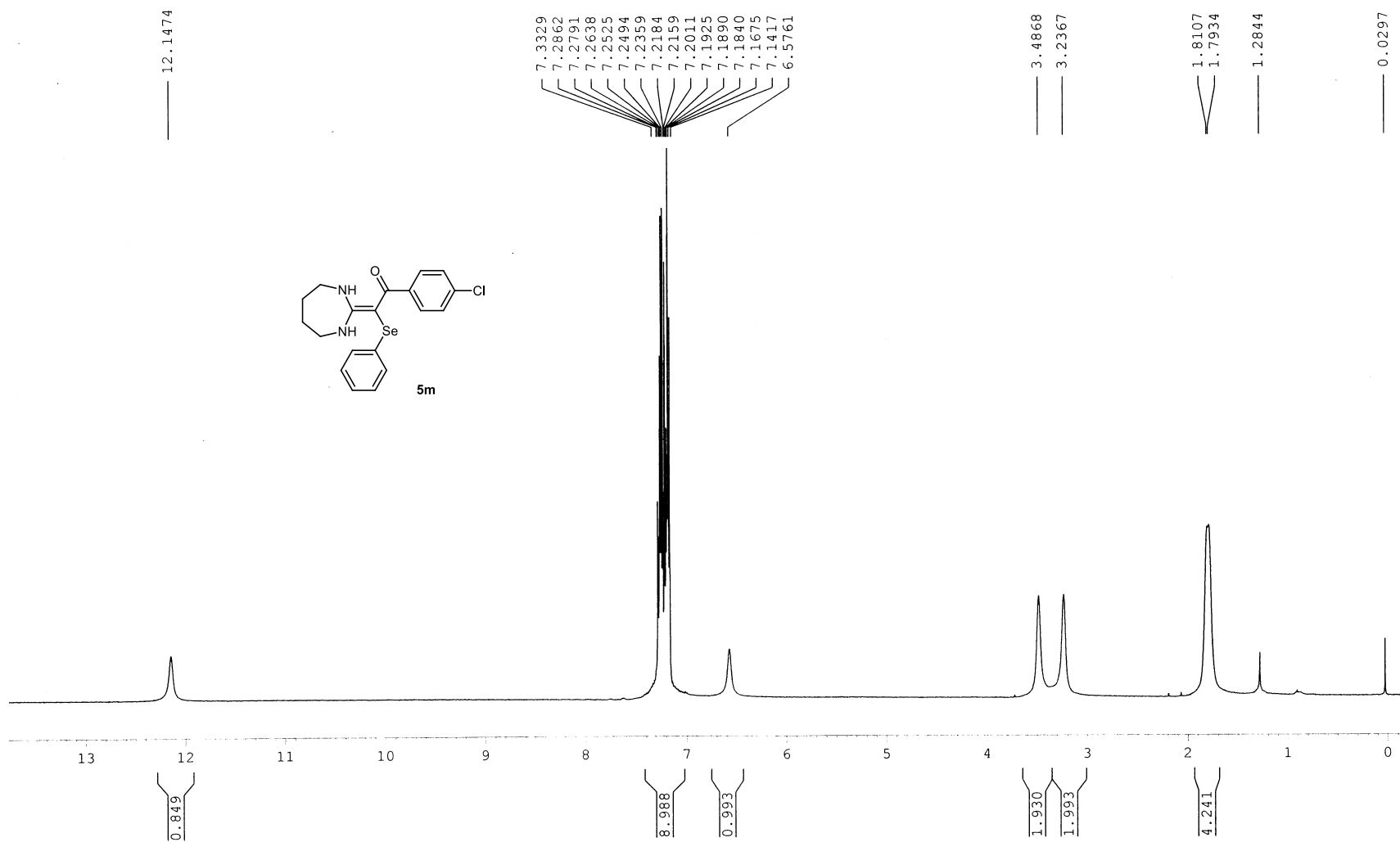


Figure S95. ¹H NMR (500 MHz, CDCl₃) spectra of compound **5m**

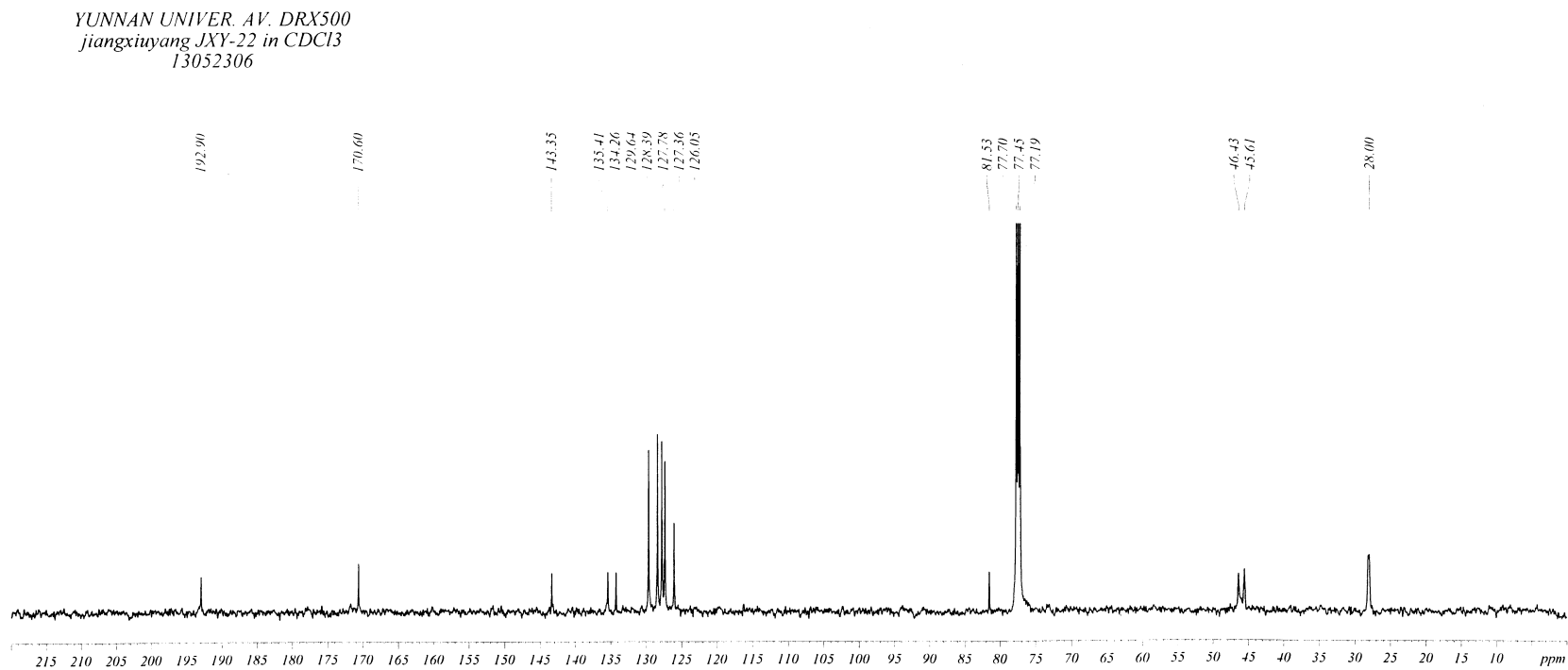
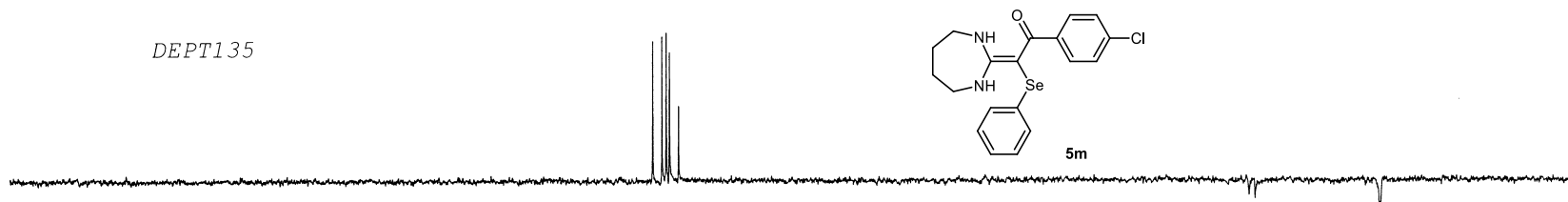


Figure S96. ¹³C NMR (125 MHz, CDCl₃) spectra of compound **5m**

Notes and References

1. (a) Huang, Z.-T.; Wang, M.-X. *Synthesis* **1992**, *12*, 1273–1276. (b) Li, Z.-J.; Charles, D. *Synth. Commun.* **2001**, *31*, 527–533. (c) X.-B. Chen, X.-M. Liu, R. Huang, S.-J. Yan, J. Lin, *Eur, J. Org. Chem.* **2013**, 4607.
2. CCDC 949284 which containing in the electronic supplementary information (ESI) for crystallographic data of compound **3g**. This material is available free of charge from The Cambridge Crystallographic Data Center *via* the Internet at www.ccdc.cam.ac.uk/data_request/cif.