Catalyst-free cascade reaction of heterocyclic ketene aminals with *N*-substituted maleimide to synthesise bicyclic pyrrolidinone derivatives

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Supporting Information

Table of Contents

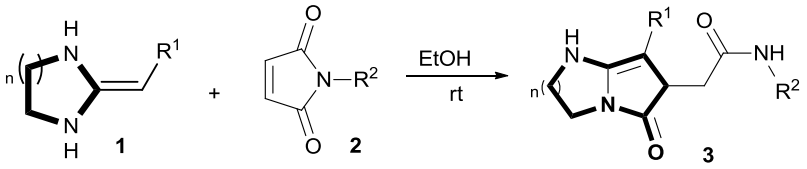
General Information

All compounds were fully characterized by spectroscopic data. The NMR spectra were recorded on a Bruker DRX500 (^{1}H: 500 MHz, ^{13}C: 125 MHz), chemical shifts (δ) are expressed in ppm, and *J* values are given in Hz, and deuterated DMSO-d_6 were used as solvent. IR spectra were recorded on a FT-IR Thermo Nicolet Avatar 360 using KBr pellet. The reactions were monitored by thin layer chromatography (TLC) using silica gel GF_{254}. 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.

The substrates **1** were synthesized according to the literature.[1] Compounds **2** was prepared according to the literature.[2]

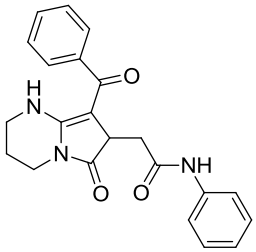
General procedure for synthesis bicyclic pyrrolidinone derivatives 3



HKA derivatives **1** (0.50 mmol), *N*-substituted maleimides **2** (0.55 mmol) and ethanol (15 ml) were placed into a 25 mL round-bottom flask and the mixture was stirred at room temperature for 20–120 min. Completion of the reaction was monitored by TLC. The reaction mixture was then filtered to obtain the pure crude product, which was further washed with 95% EtOH to give pure product **3** with a yield of 20–96%. The products were further identified by FTIR, NMR and HRMS.

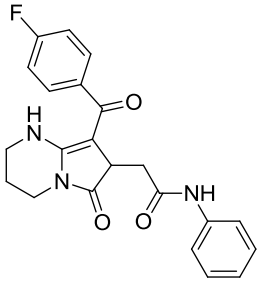
Spectroscopic data of bicyclic pyrrolidinone derivatives 3

2-(8-Benzoyl-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-phenyl acetamide (3a)



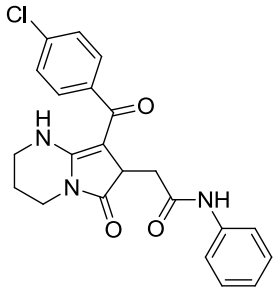
Light yellow solid: mp 191–193 °C; IR (KBr): 3424, 3244, 1740, 1632, 1526, 1434, 1158, 1082, 745 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.81 (br, 1H, NH), 9.54 (br, 1H, NH), 7.52–7.51 (m, 2H, ArH), 7.41–7.38 (m, 5H, ArH), 7.23–7.22 (m, 2H, ArH), 6.99–6.96 (m 1H, ArH), 4.03–3.95 (m, 1H, CH), 3.59–3.55 (m, 2H, NCH_2), 3.51–3.42 (m, 2H, CH_2N), 2.54–2.23 (m, 2H, CH_2CO), 1.95–1.89 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.8, 177.6, 168.4, 158.6, 142.3, 139.3, 129.6, 129.0, 128.5, 126.9, 123.3, 119.4, 88.5, 40.8, 38.7, 37.3, 36.9, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{21}N_3NaO_3$ [$(M+Na)^+$], 398.1475; found, 398.1472.

2-(8-(4-Fluorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-phenylacetamide (3b)



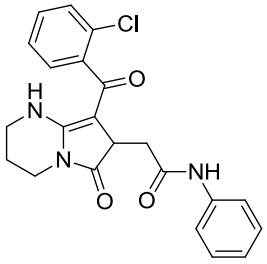
Light yellow solid: mp 192–193 °C; IR (KBr):, 3313, 2933, 1733, 1619, 1537, 1441, 1149, 853, 760 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.80 (br, 1H, NH), 9.53 (br, 1H, NH), 7.59–7.56 (m, 2H, ArH), 7.40–7.39 (m, 2H, ArH), 7.24–7.19 (m, 4H, ArH), 6.99–6.96 (m, 1H, ArH), 4.00–3.96 (m, 1H, CH), 3.58–3.55 (m, 2H, NCH_2), 3.43–3.41 (m, 2H, CH_2N), 2.57–2.27 (m, 2H, CH_2CO), 1.94–1.92 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ =181.5, 177.1, 168.3, 161.9, 158.8, 139.2, 138.7, 129.3, 128.9, 123.4, 119.4, 115.4 (d, J = 21.3 Hz), 88.4, 40.8, 38.7, 37.3, 37.0, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{20}FN_3NaO_3$ [$(M+Na)^+$], 416.1381; found, 416.1381.

2-(8-(4-Chlorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl) -*N*-phenylacetamide (3c)



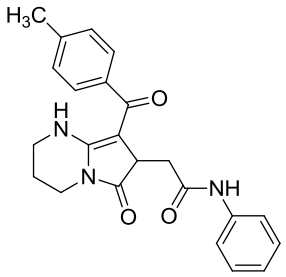
Light yellow solid: mp 211.5–212.5 °C; IR (KBr): 3369, 3228, 1735, 1635, 1525, 1368, 1269, 1160, 1086, 772, 715 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.81 (br, 1H, NH), 9.55 (br, 1H, NH), 7.55–7.53 (m, 2H, ArH), 7.43–7.38 (m, 4H, ArH), 7.24–7.21 (m, 2H, ArH), 6.99–6.96 (m, 1H, ArH), 3.96–3.96 (m, 1H, CH), 3.59–3.55 (m, 2H, NCH_2), 3.43–3.42 (m, 2H, CH_2N), 2.60–2.29 (m, 2H, CH_2CO), 1.96–1.1.91 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ =181.2, 177.1, 168.3, 159.0, 141.0, 139.2, 134.1, 128.9, 128.9, 128.6, 123.4, 119.4, 88.6, 40.7, 38.7, 37.4, 37.1, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{20}ClN_3NaO_3$ [$(M+Na)^+$], 432.1085; found, 432.1086.

2-(8-(2-Chlorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl) -*N*-phenylacetamide (3d)



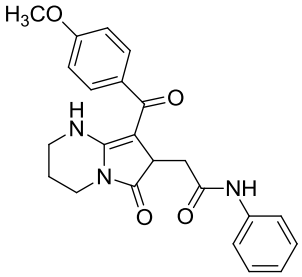
Light yellow solid: mp 190-190.6 °C; IR (KBr): 3364, 3225, 1737, 1635, 1525, 1160, 1086, 772 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.62 (br, 1H, NH), 9.52 (br, 1H, NH), 7.45–7.42 (m, 3H, ArH), 7.35–7.32(m, 2H, ArH), 7.25–7.22 (m, 3H, ArH), 7.00–6.97 (m, 1H, ArH), 3.59–3.56 (m, 2H, CH_2N), 3.55–3.53 (m, 1H, CH), 3.49–3.43 (m, 2H, NCH_2), 2.44–2.03 (m, 2H, CH_2CO), 1.96–1.91 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.7, 177.4, 168.6, 158.6, 141.8, 139.8, 130.5, 130.5, 130.4, 129.4, 128.8, 128.0, 123.8, 120.0, 89.6, 40.7, 39.3, 37.8, 37.3, 20.3; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{20}ClN_3NaO_3$ [$(M+Na)^+$], 432.1085; found, 432.1086.

2-(8-(4-Methylbenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl) -*N*-phenylacetamide (3e)



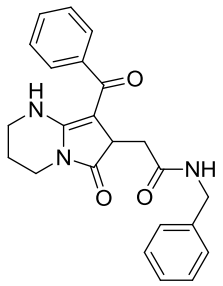
Light yellow solid: mp 215–218 °C; IR (KBr): 3256, 3120, 1738, 1683, 1631, 1525, 1439, 1160, 1094, 837, 759 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.81 (br, 1H, NH), 9.52 (br, 1H, NH), 7.44–7.40 (m, 4H, ArH), 7.24–7.22 (m, 2H, ArH), 7.21–7.18 (m, 2H, ArH), 6.99–6.96 (m, 1H, ArH), 4.03–3.99 (m, 1H, CH), 3.57–3.54 (m, 2H, CH_2N), 3.42 (m, 2H, NCH_2), 2.53–2.28 (m, 2H, CH_2CO) 2.30 (s, 3H CH_3), 2.00–1.87 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ =183.3, 177.7, 168.9, 159.0, 139.8, 139.4, 129.8, 129.4, 127.6, 123.8, 124.6, 119.8, 88.9, 41.3, 40.0, 37.8, 37.6, 21.8, 20.5; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{24}N_3O_3$ [$(M+H)^+$], 390.1812; found, 390.1816.

2-(8-(4-Methoxybenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-phenylacetamide (3f)



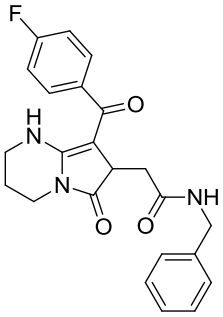
Light yellow solid: mp 207–209 °C; IR (KBr): 3334, 3244, 1731, 1638, 1526, 1438, 1161, 1090, 760 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.79 (br, 1H, NH), 9.54 (br, 1H, NH)), 7.51 (d, *J* = 8.5 Hz, 2H, ArH), 7.40 (d, *J* = 8.2 Hz, 2H, ArH), 7.23–7.20 (m, 2H, ArH), 6.99–6.96 (m, 1H, ArH), 6.91 (d, *J* = 8.6 Hz, 2H, ArH), 4.04–3.99 (m, 1H, CH), 3.76 (s, 3H, OCH_3), 3.61–3.49 (m, 4H, NCH_2CH_2N), 2.57–2.33 (m, 2H, CH_2CO), 1.96–1.89 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 187.5, 182.5, 175.1, 165.6, 163.8, 144.5, 139.8, 134.2, 134.2, 128.6, 124.6, 119.0, 95.5, 60.7, 46.2, 43.9, 42.6, 42.3, 25.2; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{24}N_3O_4$ [$(M+H)^+$], 406.1761; found, 406.1757.

2-(8-Benzoyl-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-benzyl acetamide (3g)



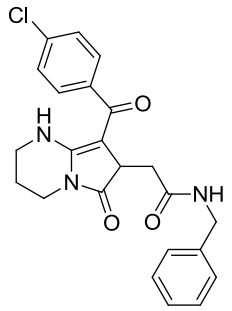
Light yellow solid: mp 209–211.5 °C; IR (KBr): 3256, 3060, 2917, 1728, 1635, 1526, 1442, 1159, 907, 742 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.83 (br, 1H, NH), 7.92 (br 1H, NH–Bn), 7.55–7.50 (m, 2H, ArH), 7.42–7.36 (m, 3H, ArH), 7.30–7.27 (m, 2H, ArH), 7.22–7.21 (m, 1H, ArH), 7.11–7.09 (m, 2H, ArH), 4.19– 4.01 (m, 2H, CH_2Ph), 3.93–3.91 (m, 1H, CH), 3.62–3.45 (m, 2H, CH_2N), 3.40–3.39 (m, 2H, NCH_2), 2.39–2.11 (m, 2H, CH_2CO), 1.98–1.85 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 183.3, 177.7, 169.8, 159.2, 142.8, 140.25, 130.0, 129.0, 127.7, 127.5, 89.0, 42.6, 41.4, 39.6, 37.8, 36.4, 20.4; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{23}N_3NaO_3$ [$(M+Na)^+$], 412.1632; found, 412.1635.

N-Benzyl-2-(8-(4-fluorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3h)



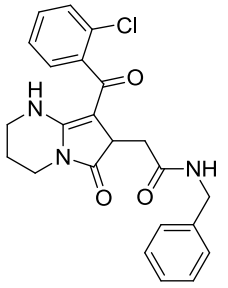
Light yellow solid: mp 218–222 °C; IR (KBr): 3236, 3052, 1734, 1637, 1526, 1446, 1099, 845, 768 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.83 (br, 1H, NH), 7.94 (br 1H, NH–Bn), 7.60–7.58 (m, 2H, ArH), 7.31–7.28 (m, 2H, ArH), 7.22–7.19 (m, 3H, ArH), 7.09–7.08 (m, 2H, ArH), 4.18–4.01 (m, 2H, CH_2Ph), 3.93–3.91 (m, 1H, CH), 3.62–3.45 (m, 4H, CH_2N), 2.44–2.15 (m, 2H, CH_2CO), 2.01–1.81 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.4, 177.2, 169.2, 163.8, 158.9, 139.7, 138.8, 129.4, 128.5, 127.2, 126.9, 115.3 (d, *J* = 21.3 Hz), 88.4, 42.1, 40.9, 38.7, 37.3, 35.9, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}FN_3NaO_3$ [$(M+Na)^+$], 430.1537; found, 430.1537.

***N*-Benzyl-2-(8-(4-chlorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3i)**



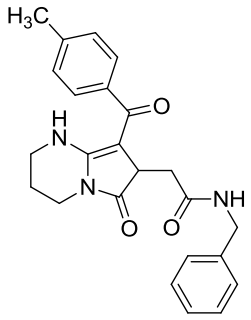
Light yellow solid: mp 232–234 °C; IR (KBr): 3366, 3227, 1736, 1635, 1525, 1086, 772 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.84 (br, 1H, NH), 7.95 (br 1H, NH–Bn), 7.56–7.54 (m, 2H, ArH), 7.44–7.42 (m, 2H, ArH) 7.31–7.28 (m, 2H, ArH), 7.21–7.19 (m, 1H, ArH)), 7.09–7.08 (m, 2H, ArH), 4.20–4.01 (m, 2H, CH_2Ph), 3.91 (m, 1H, CH), 3.63–3.49 (m, 2H, CH_2N), 3.40 (m, 2H, NCH_2), 2.55–2.16 (m, 2H, CH_2CO), 2.00–1.80 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.1, 177.1, 169.2, 159.1, 141.0, 139.7, 134.1, 129.0, 128.5, 128.5, 127.2, 126.9, 88.6, 42.1, 40.8, 38.7, 37.3, 35.9, 19.8; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}ClN_3NaO_3$ [$(M+Na)^+$], 446.1242; found, 446.1240.

_N_-Benzyl-2-(8-(2-chlorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3j)



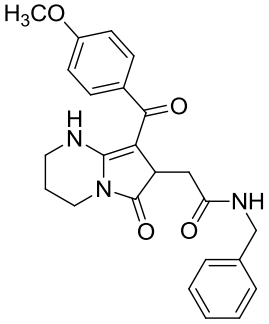
Light yellow solid: mp 199–202°C; IR (KBr): 3363, 3226, 1738, 1635, 1525, 1086, 772 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.51 (br, 1H, NH), 8.02 (br 1H, NH–Bn), 7.46–7.42 (m, 1H, ArH), 7.36–7.30 (m, 4H, ArH), 7.23–7.18 (m, 4H, ArH), 4.18–7.07 (m, 2H, CH_2Ph), 3.56–3.52 (m, 1H, CH), 3.52–3.46 (m, 4H, NCH_2CH_2N), 2.28–1.93 (m, 2H, CH_2CO), 1.93–1.89 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.7, 177.5, 169.6, 158.6, 141.9, 140.3, 130.5, 130.4, 130.4, 129.0, 129.0, 127.9, 127.5, 89.7, 42.8, 40.5, 39.2, 37.7, 36.2, 20.3; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}ClN_3NaO_3$ [$(M+Na)^+$], 446.1242; found, 446.1241.

_N_-Benzyl-2-(8-(4-methylbenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3k)



Light yellow solid: mp 179–181 °C; IR (KBr): 3559, 3420, 3227, 1724, 1633, 1521, 1450, 1099, 756 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.85 (br, 1H, NH), 7.94 (br, 1H, NH-Bn), 7.47–7.45 (d, J = 7.9 Hz, 2H, ArH), 7.30–7.27 (m, 2H, ArH), 7.21–7.18 (m, 3H, ArH), 7.11–7.10 (d, J = 7.6 Hz, 2H, ArH), 4.19 –4.02 (m, 2H, CH_2Ph), 3.95–3.94 (m, 1H, CH), 3.61–3.44 (m, 2H, CH_2N), 3.47–3.45 (m, 2H, NCH_2), 2.40–2.16 (m, 2H, CH_2CO), 2.32 (s, 3H, CH_3), 1.94–1.85 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.7, 177.2, 169.4, 158.0, 139.7, 139.5, 139.0, 129.0, 128.5, 127.2, 127.1, 126.9, 88.4, 42.1, 41.0, 38.6, 37.3, 35.9, 21.4, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{24}H_{25}N_3NaO_3$ [$(M+Na)^+$], 426.1788; found, 426.1790.

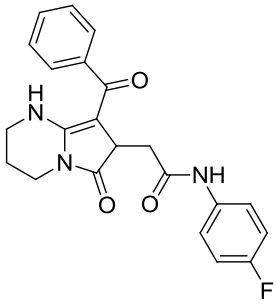
_N_-Benzyl-2-(8-(4-methoxybenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3l)



Light yellow solid: mp 216–219 °C; IR (KBr): 3285, 3199, 2917, 1735, 1632, 1518, 1433, 1360, 1258, 1161, 1198, 1023, 849, 765, 613 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.83 (br, 1H, NH), 7.91 (br 1H, NH-Bn), 7.54–7.52 (d, J = 8.6 Hz, 2H, ArH), 7.31–7.25 (m, 1H, ArH), 7.21–7.20 (m, 1H, ArH), 7.10–7.09 (m, 2H, ArH), 6.91 (d, J =8.7 Hz, 2H, ArH), 4.18–4.02 (m, 2H, CH_2Ph), 3.95 (m, 1H, CH), 3.78 (s, 3H, OCH_3), 3.60–3.47 (m, 2H, CH_2N), 3.39–3.38 (m, 2H, NCH_2), 2.45–2.21 (m, 2H, CH_2CO), 1.97–1.81 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.1, 177.2, 169.4, 160.4, 158.6, 139.7, 134.7, 128.9, 128.5, 127.2, 126.9, 113.6, 88.1, 55.5, 42.1, 41.1, 38.6, 37.3, 35.9, 20.0; HRMS (TOF ES^+): *m/z* calcd for $C_{24}H_{25}N_3NaO_4$ [$(M+Na)^+$], 442.1737; found, 442.1739.

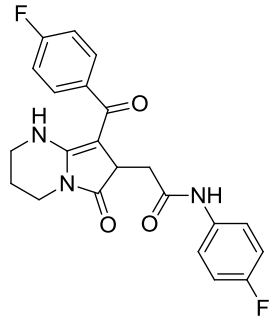
2-(8-Benzoyl-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-_N_-(4-fluo

rophenyl)acetamide (3m)



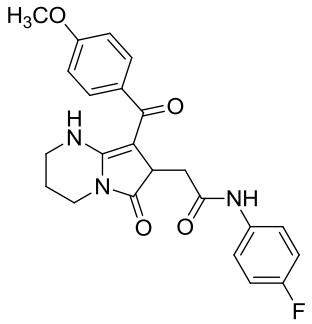
Light yellow solid: mp 190.5–191.5 °C; IR (KBr): 3264, 3068, 1728, 1631, 1519, 1088, 837 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.81 (br, 1H, NH), 9.61 (br, 1H, NH), 7.52–7.50 (m, 2H, ArH), 7.47–7.35 (m, 5H, ArH), 7.07–7.04 (m, 2H, ArH), 4.04–3.94 (m, 1H, CH), 3.64–3.48 (m, 2H, CH_2N), 3.42–3.41 (m, 2H, NCH_2), 2.54–2.21 (m, 2H, CH_2CO), 1.97–1.89 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 183.3, 177.6, 168.8, 159.1, 157.7, 142.8, 136.2, 130.1, 129.0, 127.4, 121.5, 116.0 (d, J = 21.3 Hz), 88.9, 41.2, 39.2, 37.8, 37.3, 20.4; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{20}FN_3NaO_3$ [$(M+Na)^+$], 416.1381; found, 416.1380.

2-(8-(4-Fluorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-(4-fluorophenyl)acetamide (3n)



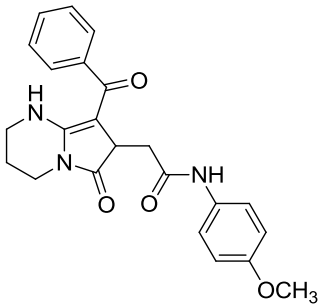
Light yellow solid: mp 129-132 °C; IR (KBr): 3267, 3076, 1733, 1636, 1515, 1440, 1267, 1224, 1158, 1082, 841, 772, 608, 510 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.80 (br, 1H, NH), 9.59(br, 1H, NH), 7.59–7.56 (m, 2H, ArH), 7.47–7.37 (m, 2H, ArH), 7.20–7.17 (m, 2H, ArH), 7.08–7.05 (m, 2H, ArH), 3.99–3.97 (m, 1H, CH), 3.62–3.50 (m, 2H, CH_2N), 3.43–3.42 (m, 2H, NCH_2), 2.57–2.25 (m, 2H, CH_2CO), 2.00–1.84 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.5, 177.1, 168.2, 162.9 (d, J = 245.0 Hz), 158.8, 158.2 (d, J = 238.8 Hz), 138.7, 135.6, 129.4, 121.1, 115.4 (d, J = 21.3 Hz), 115.3, 88.4, 40.8, 38.7, 37.3, 37.0, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{19}F_2N_3NaO_3$ [$(M+Na)^+$], 434.1287; found, 3434.1287.

***N*-(4-Fluorophenyl)-2-(8-(4-methoxybenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo [1,2-*a*]pyrimidin-7-yl)acetamide (3o)**



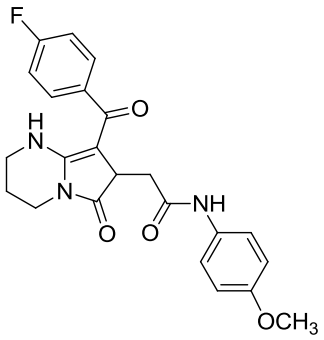
Light yellow solid: mp 182–184.5 °C; IR (KBr): 3264, 3211, 3076, 1733, 1633 1512, 1428, 1255 1160, 1090, 1012 , 845, 767, 600, 498 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.81 (br, 1H, NH), 9.60 (br, 1H, NH), 7.53 (d, J =8.4 Hz, 2H, ArH), 7.45–7.42 (m, 2H, ArH), 7.09–7.05 (m, 2H, ArH), 6.93–6.91 (d, J= 8.4, 2H, ArH), 4.04–4.02 (m, 1H, CH), 3.77 (s, 3H, OCH_3), 3.55–3.54 (m, 2H, CH_2N), 3.43–3.42 (m, 2H, NCH_2), 2.58–2.33 (m, 2H, CH_2CO), 1.98–1.86 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.2, 177.2, 168.3, 160.4, 158.5, 135.7, 134.6, 128.8, 121.1, 115.5, 115.4, 113.7, 88.1, 55.4, 41.0, 38.6, 37.3, 37.0, 20.0; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}FN_3NaO_4$ [$(M+Na)^+$], 446.1487; found, 446.1485.

2-(8-Benzoyl-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-(4-methoxyphenyl)acetamide (3p)



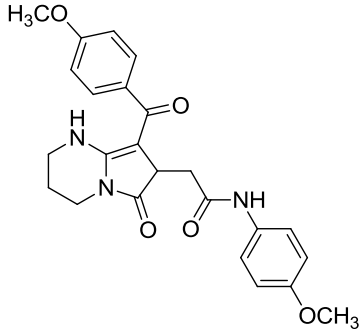
Light yellow solid: mp 184.5–185.5 °C; IR (KBr): 3260, 3010, 1732, 1634, 1521, 14363, 1088, 828 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.84 (br, 1H, NH), 9.42 (br, 1H, NH), 7.55–7.54 (m, 2H, ArH), 7.45–7.38 (m, 3H, ArH), 7.35–7.33 (d, J = 9.0 Hz, 2H, ArH), 6.85–6.82 (d, J = 9.0 Hz, 2H, ArH), 4.01–3.99 (m, 1H, CH), 3.70 (s, 3H, OCH_3), 3.58–3.56 (m, 2H, CH_2N), 3.45–3.44 (m, 2H, NCH_2), 2.53–2.22 (m, 2H, CH_2CO), 2.02–1.88 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 183.3, 177.7, 168.3, 159.1, 155.8, 142.8, 133.0, 130.1, 129.0, 127.4, 121.4, 114.5, 89.0, 56.0, 41.3, 39.2, 37.8, 37.3, 20.4; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{23}N_3NaO_4$ [$(M+Na)^+$], 428.1581; found, 428.1579

2-(8-(4-Fluorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-(4-methoxyphenyl)acetamide (3q)



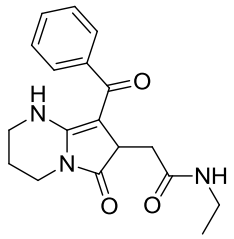
Light yellow solid: mp 225–226 °C; IR (KBr): 3260, 2937, 1732, 1655, 1521, 1088, 1033, 834 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.80 (br, 1H, NH), 9.37 (br, 1H, NH), 7.59–7.56 (m, 2H, ArH), 7.31–7.29 (d, *J*= 9.0 Hz, 2H, ArH), 7.21–7.19 (m, 2H, ArH), 6.81–6.79 (d, *J* = 9.0 Hz, 2H, ArH), 3.97–3.95 (m, 1H, CH), 3.68 (s, 3H, OCH_3), 3.57–3.53 (m, 2H, CH_2N), 3.42–3.41 (m, 2H, NCH_2), 2.54–2.23 (m, 2H, CH_2CO), 2.00–1.87 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.0, 177.7, 168.3, 163.4 (d, *J* = 245.0 Hz), 159.3, 155.9, 139.3, 133.0, 129.9, 121.4, 115.9 (d, *J* = 21.3 Hz), 114.5, 88.9, 56.0, 41.3, 39.2, 37.8, 37.4, 20.4; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}FN_3NaO_4$ [$(M+Na)^+$], 446.1487; found, 446.1484.

2-(8-(4-Methoxybenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-*N*-(4-methoxyphenyl)acetamide (3r)



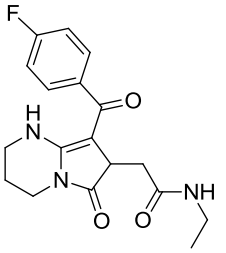
Light yellow solid: mp 165–167 °C; IR (KBr): 3432, 3338, 1727, 1632, 1517, 1250, 1168, 1106, 1031, 841, 768, 616, 535, cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.82 (br, 1H, NH), 9.41 (br, 1H, NH), 7.54–7.52 (d, *J* = 8.7 Hz, 2H, ArH), 7.33–7.31 (d, *J* = 9.0 Hz, 2H, ArH), 6.93–6.91 (d, *J* = 8.7 Hz, 2H, ArH), 6.82–6.80 (d, *J* = 9.0 Hz, 2H, ArH), 4.03–4.01 (m, 1H, CH), 3.77 (s, 3H, OCH_3), 3.68 (s, 3H, OCH_3), 3.58–3.56 (m, 2H, CH_2N), 3.45–3.41 (m, 2H, NCH_2), 2.56–2.31 (m, 2H, CH_2CO), 1.98–1.86 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.2, 177.2, 167.9, 160.4, 158.5, 155.4, 134.7, 132.5, 128.8, 120.9, 114.0, 114.0, 113.7, 88.2, 55.5, 41.0, 38.7, 37.3, 36.9, 20.0; HRMS (TOF ES^+): *m/z* calcd for $C_{24}H_{25}N_3NaO_5$ [$(M+Na)^+$], 458.1686; found, 3458.1686.

2-(8-Benzoyl-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)-N-ethylacetamide (3s)



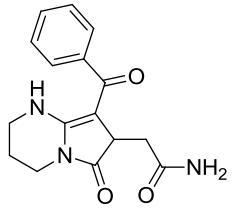
Light yellow solid: mp 143.5–144.5 °C; IR (KBr): 3340, 3236, 1734, 1637, 1528, 1445, 1364, 1269, 1162, 1088, 744, 702, 635, 523 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.79 (br, 1H, NH), 7.53–7.47 (m, 2H, ArH), 7.40–7.35 (m, 2H, ArH), 7.33 (br, 1H, NH), 3.87–3.85 (m, 1H, CH), 3.59–3.44 (m, 4H, CH_2N), 2.92–2.82 (m, 2H, CH_2Me), 2.26–1.97 (m, 2H, CH_2CO), 1.94–1.86 (m, 2H, CH_2), 0.87–0.84 (m, 3H, CH_3); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.8, 177.2, 168.9, 158.7, 142.3, 129.7, 128.4, 126.9, 88.6, 40.8, 38.7, 37.2, 36.0, 33.5, 19.9, 15.0; HRMS (TOF ES^+): *m/z* calcd for $C_{18}H_{21}N_3NaO_3$ [$(M+Na)^+$], 350.1475; found, 350.1475.

***N*-Ethyl-2-(8-(4-fluorobenzoyl)-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3t)**



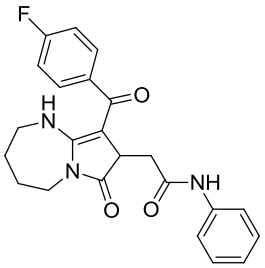
Light yellow solid: mp 199–200 °C; IR (KBr): 3324, 3072, 1729, 1631, 1538, 1446, 1372, 1102, 906, 857 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.78 (br, 1H, NH), 7.56–7.54 (m, 2H, ArH), 7.34 (br, 1H, NH), 7.20–7.16 (m, 2H, ArH), 3.85 (m, 1H, CH), 3.54–3.49 (m, 2H, CH_2N), 3.40 (m, 2H, NCH_2), 2.91–2.80 (m, 2H, CH_2Me), 2.30 –1.95 (m, 2H, CH_2CO), 1.96–1.83 (m, 2H, CH_2), 0.86–0.84 (m, 3H, CH_3); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.4, 177.2, 168.8, 162.8 (d, J = 243.8 Hz), 158.8, 138.8, 129.4, 115.3 (d, J = 21.3 Hz), 88.5, 40.8, 38.7, 37.3, 36.0, 33.4, 19.9, 15.0; HRMS (TOF ES^+): *m/z* calcd for $C_{18}H_{20}FN_3NaO_3$ [$(M+Na)^+$], 368.1381; found, 368.1379.

2-(8-Benzoyl-6-oxo-1,2,3,4,6,7-hexahydropyrrolo[1,2-*a*]pyrimidin-7-yl)acetamide (3u)



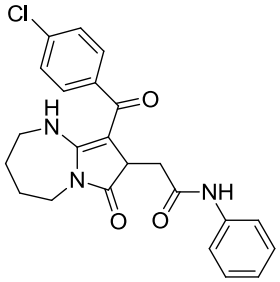
Light yellow solid: mp 223–224 °C; IR (KBr): 3352, 3154, 1730, 1629, 1525, 1082, 890 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.80 (br, 1H, NH), 7.57–7.47 (m, 2H, ArH), 7.39 (m, 3H, ArH), 6.88 (br, 1H, NH), 6.52 (br, 1H, NH), 3.86–3.84 (m, 1H, CH), 3.55–3.46 (m, 4H, NCH_2), 2.25–1.97 (m, 2H, CH_2CO), 1.90–1.87 (m, 2H, CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.7, 177.3, 171.4, 158.7, 142.3, 129.5, 128.5, 127.0, 88.6, 40.7, 38.7, 37.3, 35.6, 19.9; HRMS (TOF ES^+): *m/z* calcd for $C_{16}H_{17}N_3NaO_3$ [$(M+Na)^+$], 322.1162; found, 322.1162.

2-(9-(4-Fluorobenzoyl)-7-oxo-2,3,4,5,7,8-hexahydro-1*H*-pyrrolo[1,2-*a*][1,3]diazepin-8-yl)-*N*-phenylacetamide (3v)



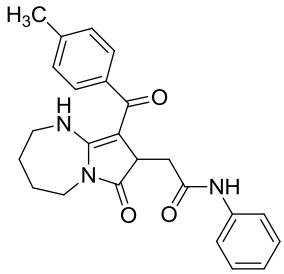
Light yellow solid: mp 178–180 °C; IR (KBr): 3444, 3314, 1734, 1682, 1619, 1537, 1442, 1090, 853, 760 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 10.62 (br, 1H, NH), 9.54 (br, 1H, NH), 7.60–7.57 (m, 2H, ArH), 7.41–7.39 (m, 2H, ArH), 7.24–7.19 (m, 4H, ArH), 7.00–6.97 (m, 1H, ArH), 4.07–3.96 (m, 1H, CH), 3.96–3.69 (m, 2H, CH_2N), 3.64–3.60 (m, 2H, NCH_2), 2.62–2.23 (m, 2H, CH_2CO), 1.99–1.82 (m,4H, CH_2CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.0, 168.2, 165.1, 163.9, 162.0, 139.2, 138.6, 129.5, 129.0, 123.4, 119.3, 115.5 (d, J = 20.0 Hz), 90.2, 41.8, 40.9, 40.6, 37.2, 27.0, 24.5; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}FN_3NaO_3$ [$(M+Na)^+$], 430.1537; found, 430.1535.

2-(9-(4-Chlorobenzoyl)-7-oxo-2,3,4,5,7,8-hexahydro-1*H*-pyrrolo[1,2-*a*][1,3]diazepin-8-yl)-*N*-phenylacetamide (3w)



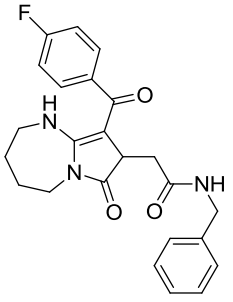
Light yellow solid: mp 194.5–195.5 °C; IR (KBr): 3368, 3226, 1736, 1634, 1525, 1438, 1086, 1008, 772 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 10.62 (br, 1H, NH), 9.56 (br, 1H, NH), 7.55–7.53 (m, 2H, ArH), 7.45–7.43 (m, 2H, ArH), 7.39–7.38 (m, 2H, ArH), 7.24–7.21 (m, 2H, ArH), 6.99–6.9 (m, 2H, ArH), 4.00–3.98 (m, 1H, CH), 3.91–3.75 (m, 2H, CH_2N), 3.61–3.60 (m, 2H, NCH_2), 2.62–2.22 (m, 2H, CH_2CO), 1.92–1.90 (m,4H, CH_2CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 181.8, 178.4, 168.2, 165.2, 140.8, 139.2, 134.3, 129.0, 129.0, 128.7, 128.7, 123.4, 119.4, 90.4, 41.8, 40.7, 37.2, 27.0, 24.5; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{22}ClN_3NaO_3$ [$(M+Na)^+$], 446.1242; found, 446.1243.

2-(9-(4-Methylbenzoyl)-7-oxo-2,3,4,5,7,8-hexahydro-1*H*-pyrrolo[1,2-*a*][1,3]diazepin-8-yl)-*N*-phenylacetamide (3x)



Light yellow solid: mp 197–198 °C; IR (KBr): 3248, 2917, 1737, 1683, 1619, 1536, 1440, 1053, 898, 756 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 10.64 (br, 1H, NH), 9.56 (br, 1H, NH), 7.45–7.40 (m, 4H, ArH), 7.24–7.19 (m, 4H, ArH), 6.99–6.96 (m, 1H, ArH), 4.05–4.03 (m, 1H, CH), 3.91–3.72 (m, 2H, CH_2N), 3.66–3.48 (m, 2H, NCH_2), 2.59–2.26 (m, 2H, CH_2CO), 2.32 (s, 3H, CH_3), 2.00–1.80 (m, 4H, CH_2CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 183.4, 178.5, 168.3, 164.9, 139.3, 129.1, 128.9, 128.9, 127.1, 123.3, 119.3, 119.3, 90.3, 41.9, 41.0, 40.7, 37.2, 27.1, 24.6, 21.4; HRMS (TOF ES^+): *m/z* calcd for $C_{24}H_{25}N_3NaO_3$ [$(M+Na)^+$], 426.1788; found, 426.1785.

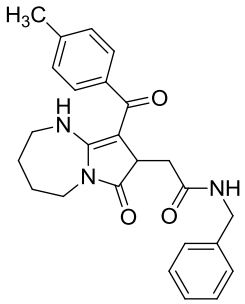
***N*-Benzyl-2-(9-(4-fluorobenzoyl)-7-oxo-2,3,4,5,7,8-hexahydro-1*H*-pyrrolo[1,2-*a*][1,3]diazepin-8-yl)acetamide (3y)**



Light yellow solid: mp 110-112 °C; IR (KBr): 3389, 3064, 1731, 1619, 1537, 1442, 1078, 837 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 10.67 (br, 1H, NH), 7.93 (br, 1H,

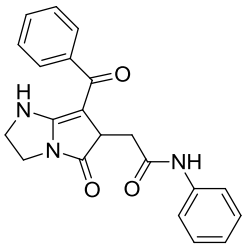
NH), 7.60–7.68 (m, 2H, ArH), 7.31–7.28 (m, 2H, ArH), 7.22–7.18 (m, 3H, ArH), 7.09–7.07 (m, 2H, ArH), 4.21–3.91 (m, 2H, CH_2Ph), 3.94–3.91 (m, 1H, CH), 3.89–3.68 (m, 2H, CH_2N), 3.58–3.55 (m, 2H, NCH_2), 2.49–2.08 (m, 2H, CH_2CO), 1.97–1.79 (m, 4H, CH_2CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 182.0, 178.5, 169.2, 165.3, 162.4 (d, J = 245.0 Hz), 139.7, 138.6, 129.5, 128.5, 127.2, 126.9, 115.4 (d, J = 21.3 Hz), 90.2, 42.1, 41.9, 41.0, 36.1, 27.0, 24.5; HRMS (TOF ES^+): *m/z* calcd for $C_{24}H_{24}FN_3NaO_3$ [$(M+Na)^+$], 444.1693; found, 444.1694.

_N_-Benzyl-2-(9-(4-methylbenzoyl)-7-oxo-2,3,4,5,7,8-hexahydro-1_H_-pyrrolo[1,2-_a_][1,3]diazepin-8-yl)acetamide (3z)



Light yellow solid: mp 110.5–112 °C; IR (KBr): 3289, 2921, 1734, 1669, 1618, 1527, 1440, 1048, 738 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 10.68 (br, 1H, NH), 7.92 (br, 1H, NH), 7.45–7.44(m, 2H, ArH), 7.31–7.27 (m, 2H, ArH), 7.22–7.18 (m, 3H, ArH), 7.12–7.09 (m, 2H, ArH), 4.21–3.99 (m, 2H, CH_2Ph), 3.92–3.62 (m, 2H, CH_2N), 3.59–3.54 (m, 1H, CH), 3.44–2.55 (m, 2H, NCH_2), 2.45–2.11 (m, 2H, CH_2CO), 2.33 (s, 3H, CH_3), 1.88–1.784 (m,4H, CH_2CH_2); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 183.3, 178.5, 169.3, 165.1, 139.7, 139.4, 139.1, 129.0, 128.5, 127.2, 126.9, 90.3, 56.4, 42.1, 41.9, 40.7, 36.1, 27.1, 24.6, 21.4; HRMS (TOF ES^+): *m/z* calcd for $C_{25}H_{27}N_3NaO_3$ [$(M+Na)^+$], 440.1945; found, 440.1943.

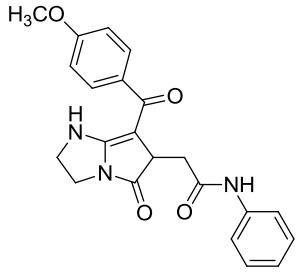
2-(7-Benzoyl-5-oxo-2,3,5,6-tetrahydro-1_H_-pyrrolo[1,2-a]imidazol-6-yl)-_N_-phenyl acetamide (3a ʾ)



Light yellow solid: mp 139–142.5 °C; IR (KBr): 3257, 3060, 1727, 1635, 1525, 1442, 1112, 742 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.55 (br, 1H, NH), 9.46 (br, 1H, NH), 7.45–7.43 (m, 2H, ArH), 7.39–7.32 (m, 3H, ArH), 7.28–7.24 (m, 4H, ArH), 7.04–7.01 (m, 1H, ArH), 3.92–3.88 (m, 1H, CH), 3.80–3.61 (m, 4H, CH_2CH_2), 2.97–2.50 (m, 2H, CH_2CO); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 189.7, 172.7, 166.4,

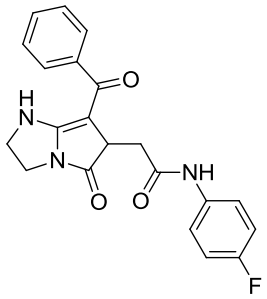
154.5, 142.4, 139.2, 129.1, 129.0, 128.5, 126.5, 123.7, 119.8, 84.7, 43.1, 41.9, 41.0, 36.5; HRMS (TOF ES^+): *m/z* calcd for $C_{21}H_{19}N_3NaO_3$ [$(M+Na)^+$], 384.1319; found, 384.1320.

2-(7-(4-Methoxybenzoyl)-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)-*N*-phenylacetamide (3b ʹ)



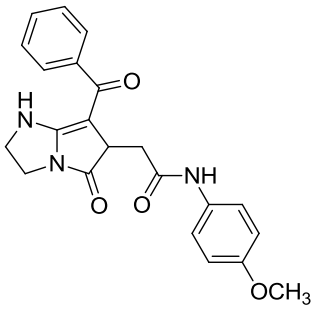
Light yellow solid: mp 122–125 °C; IR (KBr): 3416, 3305, 1690, 1614, 1496, 1443, 1170, 1023, 841 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.66 (br, 1H, NH), 9.24 (br, 1H, NH), 7.47–7.45 (m, 2H, ArH), 7.29–7.26(m, 4H, ArH), 7.04–7.02 (m, 1H, ArH), 6.91–6.90 (m, 2H, ArH), 3.89–3.86 (m, 1H, CH), 3.76 (s, 3H, OCH_3), 3.71–3.70 (m, 4H, CH_2CH_2), 2.95–2.58 (m, 2H, CH_2CO); ^{13}C NMR (125 MHz, DMSO-d_6): δ =189.3, 172.7, 167.5, 160.2, 157.2, 139.3, 134.6, 129.1, 128.6, 123.7, 119.7, 113.7, 84.7, 55.6, 43.0, 41.9, 41.1, 36.3; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{21}N_3NaO_4$ [$(M+Na)^+$], 414.1424; found, 414.1427.

2-(7-Benzoyl-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)-*N*-(4-fluorophenyl)acetamide (3c ʹ)



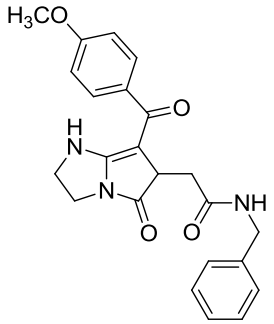
Light yellow solid: mp 131–132.5 °C; IR (KBr): 3499, 3400, 1686, 1620, 1508, 1102, 1013, 849 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 9.65 (br, 1H, NH), 9.48 (br, 1H, NH), 7.47–7.44 (m, 2H, ArH), 7.36–7.35 (m, 3H, ArH), 7.24–7.23 (m, 2H, ArH), 7.13–7.09 (m, 2H, ArH), 3.92–3.89 (m, 1H, CH), 3.77–3.57 (m, 4H, CH_2CH_2), 2.98–2.55 (m, 2H, CH_2CO); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 189.7, 172.6, 167.4, 159.4, 157.5, 142.4, 135.6, 129.1, 128.5, 126.4, 121.5, 115.6 (d, *J* = 21.3 Hz), 84.6, 43.1, 41.8, 40.9, 36.5; HRMS (TOF ES^+): *m/z* calcd for $C_{21}H_{18}FN_3NaO_3$ [$(M+Na)^+$], 402.1224; found, 402.1226.

2-(7-Benzoyl-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)-*N*-(4-methoxyphenyl)acetamide (3d΄)



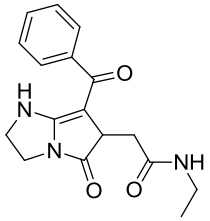
Light yellow solid: mp 181–182 °C; IR (KBr): 3264, 3072, 2958, 1689, 1624, 1504, 1025, 833, 702 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.49 (br, 1H, NH), 9.41 (br, 1H, NH), 7.36–7.33 (m, 5H, ArH), 7.25–7.22 (m, 2H, ArH), 6.85–6.83 (m, 2H, ArH), 3.92–3.85 (m, 1H, CH), 3.78–3.71 (m, 2H, CH_2N), 3.70 (s, 3H, OCH_3), 3.68–3.53 (m, 2H, NCH_2), 2.95–2.54 (m, 2H, CH_2CO); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 189.7, 172.2, 167.5, 157.5, 155.6, 142.5, 132.3, 129.1, 128.4, 126.4, 121.3, 114.1, 84.8, 55.5, 43.1, 41.8, 40.8, 36.6; HRMS (TOF ES^+): *m/z* calcd for $C_{22}H_{21}N_3NaO_4$ [$(M+Na)^+$], 414.1424; found, 414.1423.

***N*-Benzyl-2-(7-(4-methoxybenzoyl)-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)acetamide (3e΄)**



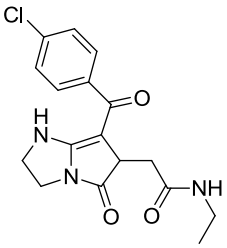
Light yellow solid: mp 191–193 °C; IR (KBr): 3309, 2925, 1691, 1625, 1498, 1450, 1021, 846, 702 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.44 (br, 1H, NH), 8.09 (br, 1H, NH-Bn), 7.32–7.31 (m, 2H, ArH), 7.25–7.23 (m, 3H, ArH), 7.21–7.20 (m, 2H, ArH), 6.82–6.80 (m, 2H, ArH), 4.26–4.11 (m, 2H, CH_2Ph), 3.92–3.82 (m, 1H, CH), 3.75 (s, 3H, OCH_3), 3.67–3.51 (m, 4H, CH_2CH_2), 2.86–2.55 (m, 2H, CH_2CO); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 189.1, 173.6, 167.8, 160.0, 139.8, 134.6, 128.6, 128.6, 127.5, 127.1, 113.5, 84.7, 55.5, 43.1, 42.6, 41.8, 40.1, 36.6; HRMS (TOF ES^+): *m/z* calcd for $C_{23}H_{23}N_3NaO_4$ [$(M+Na)^+$], 428.1581; found, 428.1579.

2-(7-Benzoyl-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)-*N*-ethylacetamide (3f΄).



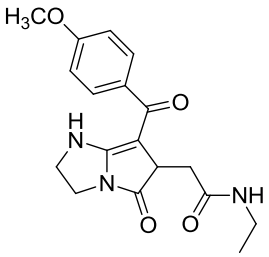
Light yellow solid: mp 230.5–231.5 °C; IR (KBr): 3110, 2952, 1695, 1590, 1529, 1458, 1123, 984, 774 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 10.18 (br, 1H, NH), 7.29–7.25 (m, 3H, ArH), 7.18–7.14 (m, 2H, ArH), 6.82 (br, 1H, NH), 3.69–3.65 (m, 1H, CH), 3.53–3.47 (m, 4H, CH_2CH_2), 3.17–3.12 (m, 2H, CH_2Me), 2.84–2.41 (m, 2H, CH_2CO), 0.93–0.86 (m, 3H, CH_3); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 189.0, 179.8, 176.4, 165.8, 144.4, 128.7, 127.2, 85.1, 43.7, 43.7, 40.0, 37.0, 33.7, 13.5; HRMS (TOF ES^+): *m/z* calcd for $C_{17}H_{20}N_3O_3$ [$(M+H)^+$], 314.1499; found, 314.1493.

2-(7-(4-Chlorobenzoyl)-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)-*N*-ethylacetamide (3g ')



Light yellow solid: mp 221–223 °C; IR (KBr): 3395, 3273, 1690, 1589, 1523, 1401, 1127, 841 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 10.12 (br, 1H, NH), 7.35–7.33 (m, 2H, ArH), 7.21–7.17 (m, 2H, ArH), 6.86 (br, 1H, NH), 3.67–3.50 (m, 4H, CH_2CH_2), 3.17–3.14 (m, 1H, CH), 2.86–2.45 (m, 2H, CH_2CO), 2.52–2.48 (m, 2H, CH_2Me), 0.90–0.86 (m,3H, CH_3); ^{13}C NMR (125 MHz, DMSO-d_6): δ =187.1, 179.1, 175.9, 165.3, 142.6, 132.7, 128.8, 128.2, 84.8, 43.1, 43.1, 40.0, 36.5, 33.2, 12.9; HRMS (TOF ES^+): *m/z* calcd for $C_{17}H_{18}ClN_3NaO_3$ [$(M+Na)^+$], 370.0929; found, 370.0928.

***N*-Ethyl-2-(7-(4-methoxybenzoyl)-5-oxo-2,3,5,6-tetrahydro-1*H*-pyrrolo[1,2-*a*]imidazol-6-yl)acetamide (3h ')**



Light yellow solid: mp 183–186 °C; IR (KBr): 3195, 2966, 1693, 1591, 1522, 1460, 1120, 1031, 825 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ = 10.19 (br, 1H, NH), 7.14

(d, *J*= 7.2 Hz, 2H, ArH), 6.84 (d, *J*= 8.1 Hz, 2H, ArH), 6.73 (br, 1H, NH), 3.85–3.81 (m, 1H, CH), 3.73 (s, 3H, OCH_3), 3.50–3.46 (m, 4H, CH_2CH_2), 3.23–3.19 (m, 2H, CH_2Me), 2.85–2.45 (m, 2H, CH_2CO), 0.94–0.89 (m, 3H, CH_3); ^{13}C NMR (125 MHz, DMSO-d_6): δ = 188.4, 179.4, 176.0, 165.2, 159.3, 136.3, 128.4, 113.5, 113.5, 84.8, 55.4, 43.1, 43.1, 36.5, 33.2, 13.0; HRMS (TOF ES^+): *m/z* calcd for $C_{18}H_{22}N_3O_4$ [$(M+H)^+$], 344.1605; found, 344.1607.

X-ray Structure and Data[3] of 3t

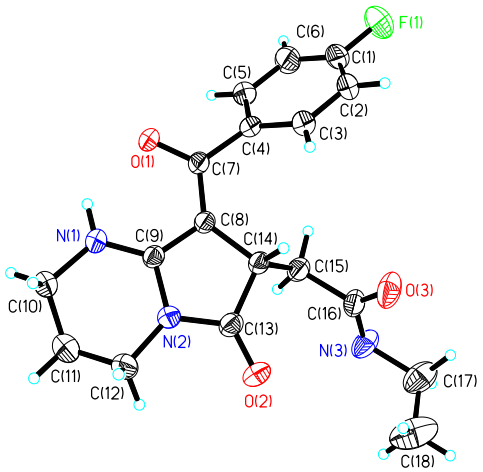


Figure S1 X-Ray crystal structure of **3t**

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

Empirical formula	$C_{18}H_{20}FN_3O_3$
Formula weight	345.15
Temperature	298(2) K
Wavelength	0.71073 A
Crystal system, space group	Triclinic, P-1
Unit cell dimensions	a = 8.9553(13) A alpha = 107.762(2) deg.
	b = 9.9570(14) A beta = 97.915(2) deg.
	c = 12.2938(17) A gamma = 98.959(2) deg.
Volume	1011.0(2) A^3
Z, Calculated density	2, 1.282 Mg/m^3
Absorption coefficient	0.096 mm^-1
F(000)	414
Crystal size	0.32 x 0.28 x 0.24 mm
Theta range for data collection	2.20 to 25.15 deg.
Limiting indices	-10<=h<=10, -11<=k<=11, -14<=l<=14
Reflections collected / unique	8074 / 3590 [R(int) = 0.0328]
Completeness to theta = 25.15	99.3 %
Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.9773 and 0.9699
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	3590 / 0 / 256
Goodness-of-fit on F^2	1.299
Final R indices [I>2sigma(I)]	R1 = 0.0723, wR2 = 0.2002
R indices (all data)	R1 = 0.1158, wR2 = 0.2346
Largest diff. peak and hole	0.557 and -0.260 e.A^-3

Table S2. Bond lengths [A] and angles [deg] for **3t**.

F(1)-C(1)	1.368(4)
N(1)-C(9)	1.320(4)
N(1)-C(10)	1.473(4)
N(1)-H(1)	0.8600
N(2)-C(13)	1.381(4)
N(2)-C(9)	1.388(4)
N(2)-C(12)	1.452(4)
N(3)-C(16)	1.337(4)
N(3)-C(17)	1.492(5)
O(1)-C(7)	1.267(3)
O(2)-C(13)	1.215(4)
O(3)-C(16)	1.235(4)
O(4)-C(20)	1.391(5)
O(4)-H(4)	0.8200
C(1)-C(6)	1.362(6)
C(1)-C(2)	1.374(6)
C(2)-C(3)	1.378(5)
C(2)-H(2)	0.9300
C(3)-C(4)	1.400(4)
C(3)-H(3)	0.9300
C(4)-C(5)	1.389(5)
C(4)-C(7)	1.499(4)
C(5)-C(6)	1.387(5)
C(5)-H(5)	0.9300
C(6)-H(6)	0.9300
C(7)-C(8)	1.416(4)
C(8)-C(9)	1.403(4)
C(8)-C(14)	1.524(4)
C(10)-C(11)	1.507(6)
C(10)-H(10A)	0.9700
C(10)-H(10B)	0.9700
C(11)-C(12)	1.501(5)
C(11)-H(11A)	0.9700
C(11)-H(11B)	0.9700
C(12)-H(12A)	0.9700
C(12)-H(12B)	0.9700
C(13)-C(14)	1.522(4)
C(14)-C(15)	1.533(4)
C(14)-H(14)	0.9800
C(15)-C(16)	1.510(4)
C(15)-H(15A)	0.9700
C(15)-H(15B)	0.9700
C(17)-C(18)	1.413(7)
C(17)-H(17A)	0.9700
C(17)-H(17B)	0.9700
C(18)-H(18A)	0.9600
C(18)-H(18B)	0.9600
C(18)-H(18C)	0.9600
C(19)-C(20)	1.455(7)
C(19)-H(19A)	0.9600
C(19)-H(19B)	0.9600
C(19)-H(19C)	0.9600
C(20)-H(20A)	0.9700
C(20)-H(20B)	0.9700

C(9)-N(1)-C(10)	123.3(3)
C(9)-N(1)-H(1)	118.4
C(10)-N(1)-H(1)	118.4
C(13)-N(2)-C(9)	110.6(3)
C(13)-N(2)-C(12)	125.2(3)
C(9)-N(2)-C(12)	124.1(3)
C(16)-N(3)-C(17)	119.6(3)
C(20)-O(4)-H(4)	109.5
C(6)-C(1)-F(1)	118.6(4)
C(6)-C(1)-C(2)	122.9(4)
F(1)-C(1)-C(2)	118.4(4)
C(1)-C(2)-C(3)	118.6(4)
C(1)-C(2)-H(2)	120.7
C(3)-C(2)-H(2)	120.7
C(2)-C(3)-C(4)	120.5(3)
C(2)-C(3)-H(3)	119.7
C(4)-C(3)-H(3)	119.7
C(5)-C(4)-C(3)	118.8(3)
C(5)-C(4)-C(7)	117.6(3)
C(3)-C(4)-C(7)	123.6(3)
C(6)-C(5)-C(4)	120.8(4)
C(6)-C(5)-H(5)	119.6
C(4)-C(5)-H(5)	119.6
C(1)-C(6)-C(5)	118.4(4)
C(1)-C(6)-H(6)	120.8
C(5)-C(6)-H(6)	120.8
O(1)-C(7)-C(8)	121.7(3)
O(1)-C(7)-C(4)	116.5(3)
C(8)-C(7)-C(4)	121.7(3)
C(9)-C(8)-C(7)	120.3(3)
C(9)-C(8)-C(14)	107.2(2)
C(7)-C(8)-C(14)	132.4(3)
N(1)-C(9)-N(2)	118.6(3)
N(1)-C(9)-C(8)	130.5(3)
N(2)-C(9)-C(8)	110.9(3)
N(1)-C(10)-C(11)	109.2(3)
N(1)-C(10)-H(10A)	109.8
C(11)-C(10)-H(10A)	109.8
N(1)-C(10)-H(10B)	109.8
C(11)-C(10)-H(10B)	109.8
H(10A)-C(10)-H(10B)	108.3
C(12)-C(11)-C(10)	111.4(4)
C(12)-C(11)-H(11A)	109.4
C(10)-C(11)-H(11A)	109.4
C(12)-C(11)-H(11B)	109.4
C(10)-C(11)-H(11B)	109.4
H(11A)-C(11)-H(11B)	108.0
N(2)-C(12)-C(11)	108.8(3)
N(2)-C(12)-H(12A)	109.9
C(11)-C(12)-H(12A)	109.9
N(2)-C(12)-H(12B)	109.9
C(11)-C(12)-H(12B)	109.9
H(12A)-C(12)-H(12B)	108.3
O(2)-C(13)-N(2)	123.9(3)
O(2)-C(13)-C(14)	127.6(3)
N(2)-C(13)-C(14)	108.5(3)
C(13)-C(14)-C(8)	102.5(2)

C(13)-C(14)-C(15)	110.1(3)
C(8)-C(14)-C(15)	117.4(2)
C(13)-C(14)-H(14)	108.8
C(8)-C(14)-H(14)	108.8
C(15)-C(14)-H(14)	108.8
C(16)-C(15)-C(14)	112.8(3)
C(16)-C(15)-H(15A)	109.0
C(14)-C(15)-H(15A)	109.0
C(16)-C(15)-H(15B)	109.0
C(14)-C(15)-H(15B)	109.0
H(15A)-C(15)-H(15B)	107.8
O(3)-C(16)-N(3)	123.6(3)
O(3)-C(16)-C(15)	121.2(3)
N(3)-C(16)-C(15)	115.1(3)
C(18)-C(17)-N(3)	110.4(5)
C(18)-C(17)-H(17A)	109.6
N(3)-C(17)-H(17A)	109.6
C(18)-C(17)-H(17B)	109.6
N(3)-C(17)-H(17B)	109.6
H(17A)-C(17)-H(17B)	108.1
C(17)-C(18)-H(18A)	109.5
C(17)-C(18)-H(18B)	109.5
H(18A)-C(18)-H(18B)	109.5
C(17)-C(18)-H(18C)	109.5
H(18A)-C(18)-H(18C)	109.5
H(18B)-C(18)-H(18C)	109.5
C(20)-C(19)-H(19A)	109.5
C(20)-C(19)-H(19B)	109.5
H(19A)-C(19)-H(19B)	109.5
C(20)-C(19)-H(19C)	109.5
H(19A)-C(19)-H(19C)	109.5
H(19B)-C(19)-H(19C)	109.5
O(4)-C(20)-C(19)	110.2(4)
O(4)-C(20)-H(20A)	109.6
C(19)-C(20)-H(20A)	109.6
O(4)-C(20)-H(20B)	109.6
C(19)-C(20)-H(20B)	109.6
H(20A)-C(20)-H(20B)	108.1

Table S3. Torsion angles [deg] for **3t**

C(6)-C(1)-C(2)-C(3)	1.0(6)
F(1)-C(1)-C(2)-C(3)	179.8(3)
C(1)-C(2)-C(3)-C(4)	-1.2(6)
C(2)-C(3)-C(4)-C(5)	0.1(5)
C(2)-C(3)-C(4)-C(7)	-176.7(3)
C(3)-C(4)-C(5)-C(6)	1.3(5)
C(7)-C(4)-C(5)-C(6)	178.3(3)
F(1)-C(1)-C(6)-C(5)	-178.4(3)
C(2)-C(1)-C(6)-C(5)	0.3(6)
C(4)-C(5)-C(6)-C(1)	-1.5(6)
C(5)-C(4)-C(7)-O(1)	-35.9(4)
C(3)-C(4)-C(7)-O(1)	140.9(3)
C(5)-C(4)-C(7)-C(8)	142.1(3)
C(3)-C(4)-C(7)-C(8)	-41.1(5)
O(1)-C(7)-C(8)-C(9)	-3.0(5)
C(4)-C(7)-C(8)-C(9)	179.1(3)
O(1)-C(7)-C(8)-C(14)	175.4(3)
C(4)-C(7)-C(8)-C(14)	-2.4(5)
C(10)-N(1)-C(9)-N(2)	0.2(5)
C(10)-N(1)-C(9)-C(8)	-179.5(3)
C(13)-N(2)-C(9)-N(1)	-178.8(3)
C(12)-N(2)-C(9)-N(1)	2.2(5)
C(13)-N(2)-C(9)-C(8)	0.9(4)
C(12)-N(2)-C(9)-C(8)	-178.0(3)
C(7)-C(8)-C(9)-N(1)	-5.2(5)
C(14)-C(8)-C(9)-N(1)	176.1(3)
C(7)-C(8)-C(9)-N(2)	175.1(3)
C(14)-C(8)-C(9)-N(2)	-3.7(3)
C(9)-N(1)-C(10)-C(11)	-28.4(5)
N(1)-C(10)-C(11)-C(12)	54.0(5)
C(13)-N(2)-C(12)-C(11)	-154.8(4)
C(9)-N(2)-C(12)-C(11)	24.0(5)
C(10)-C(11)-C(12)-N(2)	-51.6(5)
C(9)-N(2)-C(13)-O(2)	-177.1(3)
C(12)-N(2)-C(13)-O(2)	1.9(5)
C(9)-N(2)-C(13)-C(14)	2.2(4)
C(12)-N(2)-C(13)-C(14)	-178.8(3)
O(2)-C(13)-C(14)-C(8)	175.1(3)
N(2)-C(13)-C(14)-C(8)	-4.2(3)
O(2)-C(13)-C(14)-C(15)	49.4(4)
N(2)-C(13)-C(14)-C(15)	-129.9(3)
C(9)-C(8)-C(14)-C(13)	4.6(3)
C(7)-C(8)-C(14)-C(13)	-173.9(3)
C(9)-C(8)-C(14)-C(15)	125.4(3)
C(7)-C(8)-C(14)-C(15)	-53.2(5)
C(13)-C(14)-C(15)-C(16)	-70.9(3)
C(8)-C(14)-C(15)-C(16)	172.4(3)
C(17)-N(3)-C(16)-O(3)	-0.1(6)
C(17)-N(3)-C(16)-C(15)	176.5(3)
C(14)-C(15)-C(16)-O(3)	-50.8(5)
C(14)-C(15)-C(16)-N(3)	132.5(3)
C(16)-N(3)-C(17)-C(18)	94.9(5)

^{1}H NMR and ^{13}C NMR spectra for bicyclic pyrrolidinones 3

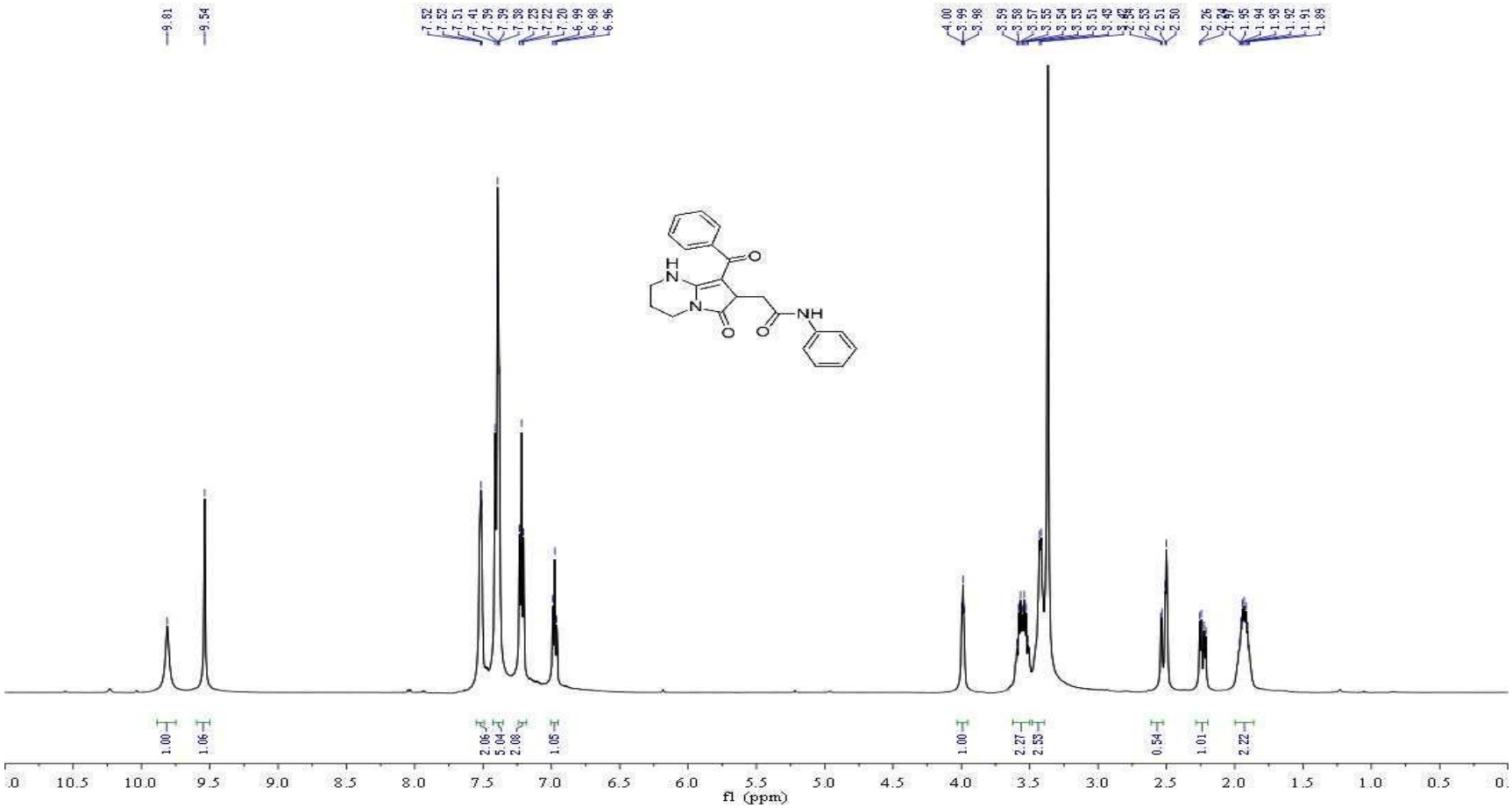


Figure 1. 1H NMR (500 MHz, DMSO-d6) spectra of compound **3a**

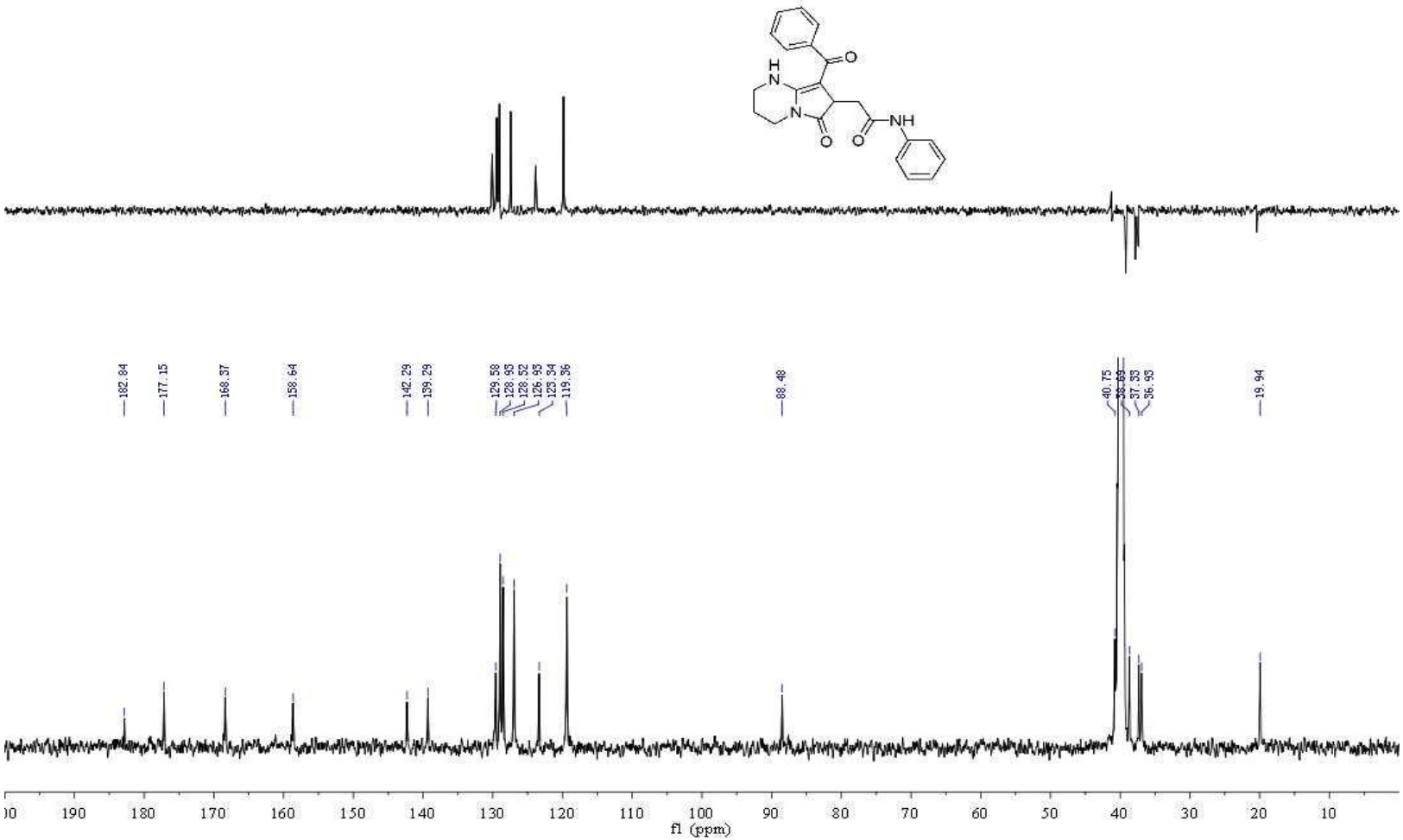


Figure 2. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3a**

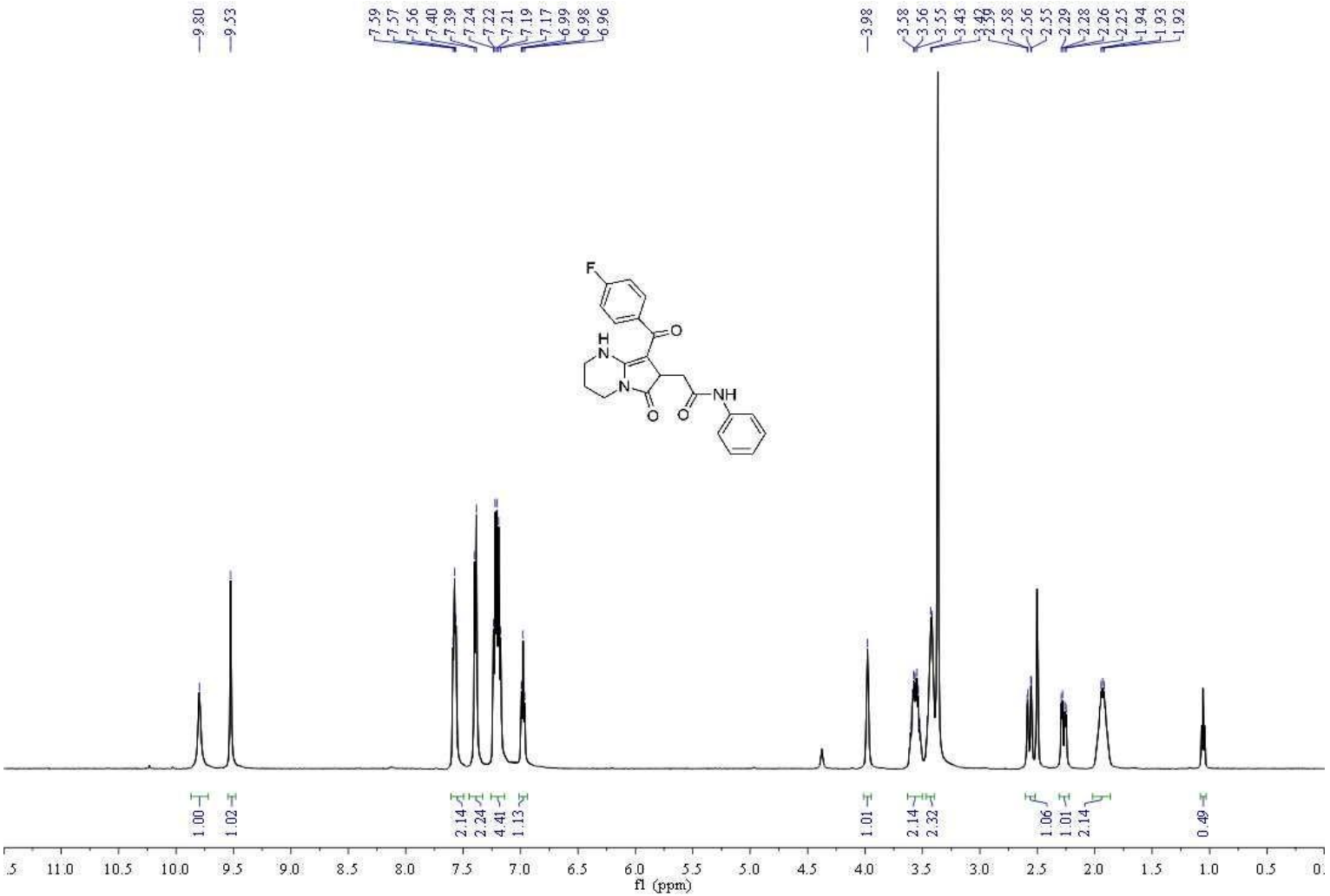


Figure 3. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3b**

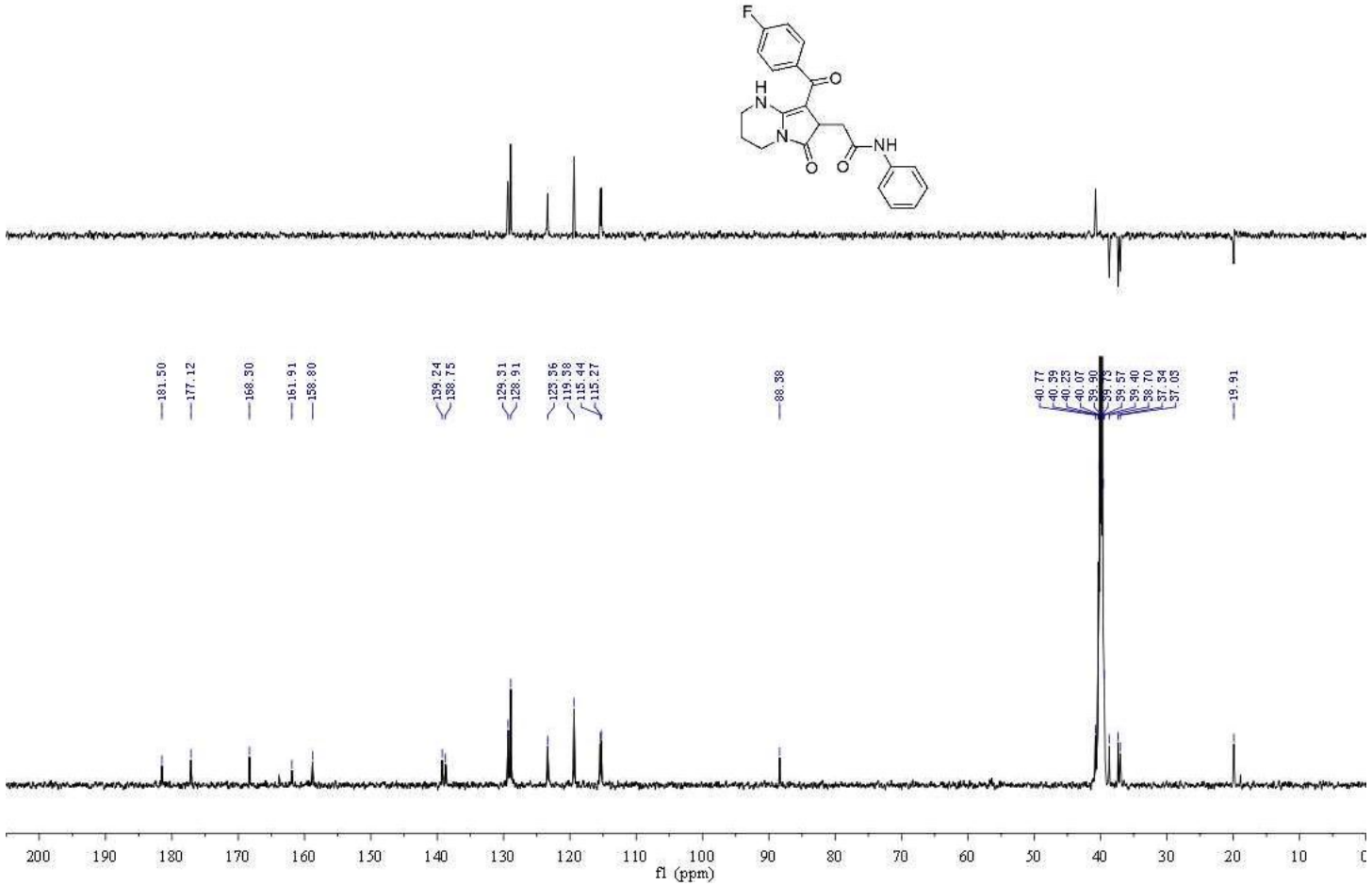


Figure 4. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3b**

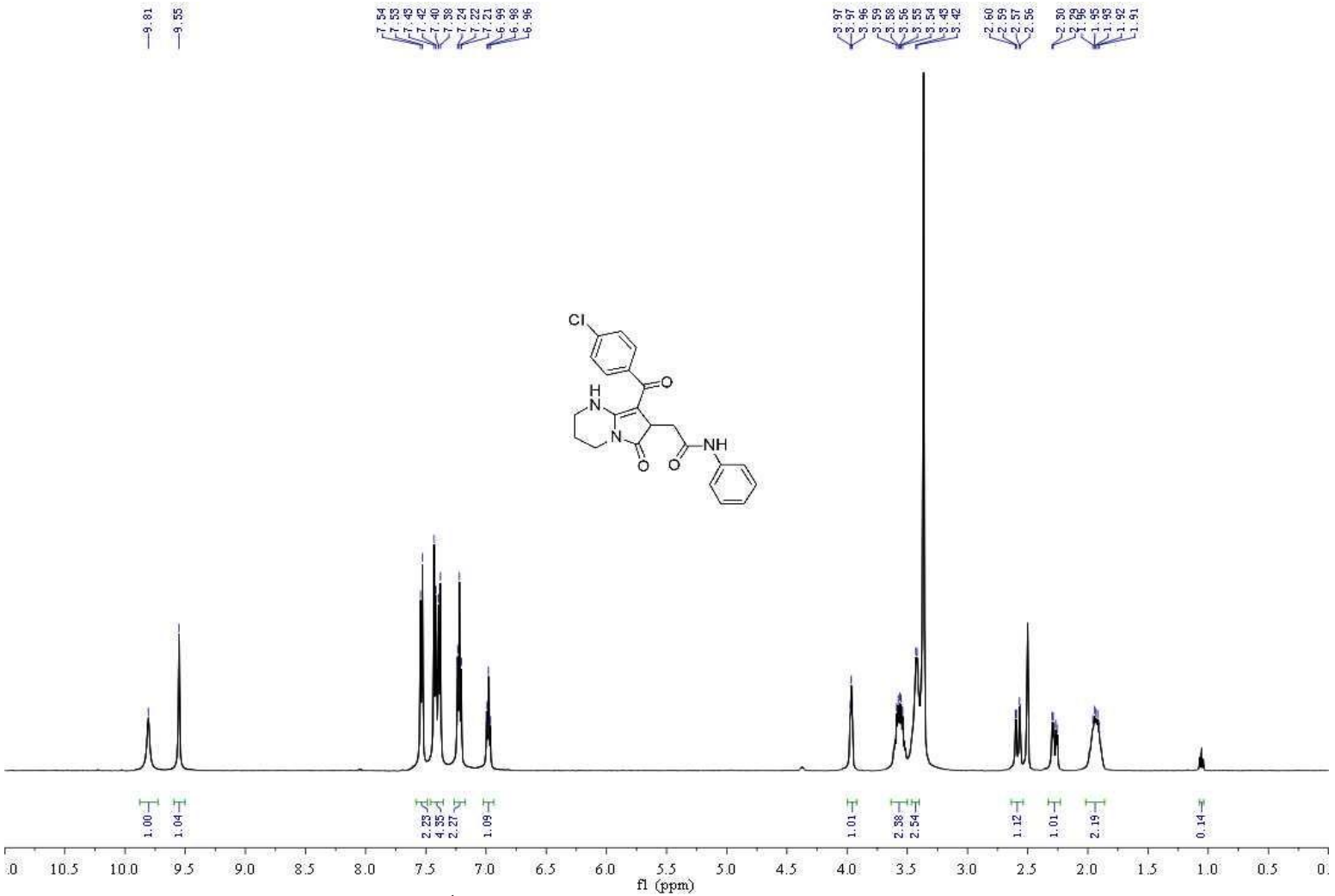


Figure 5. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3c**

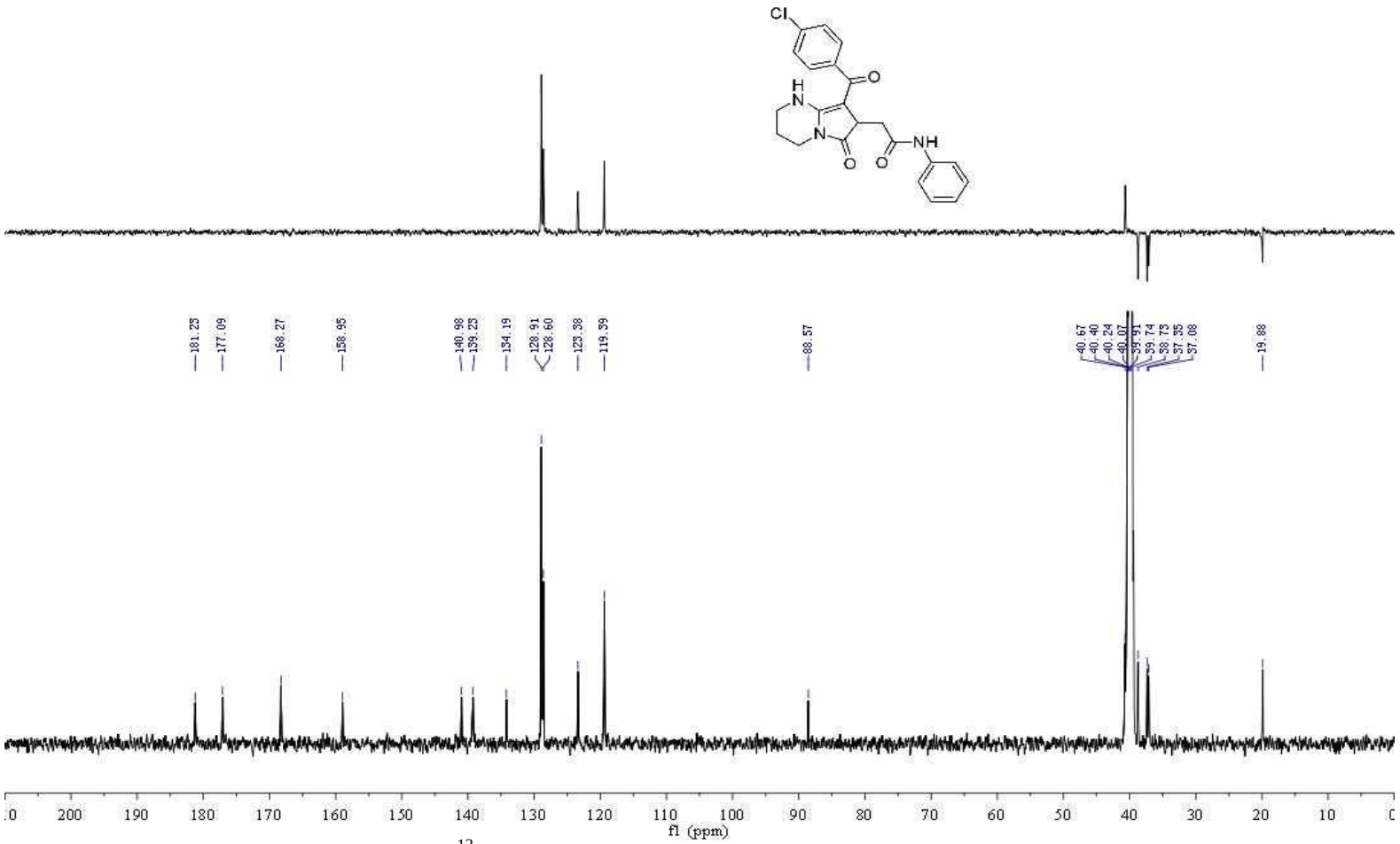


Figure 6. ^{13}C NMR (125 MHz, DMSO-d6) spectra of compound **3c**

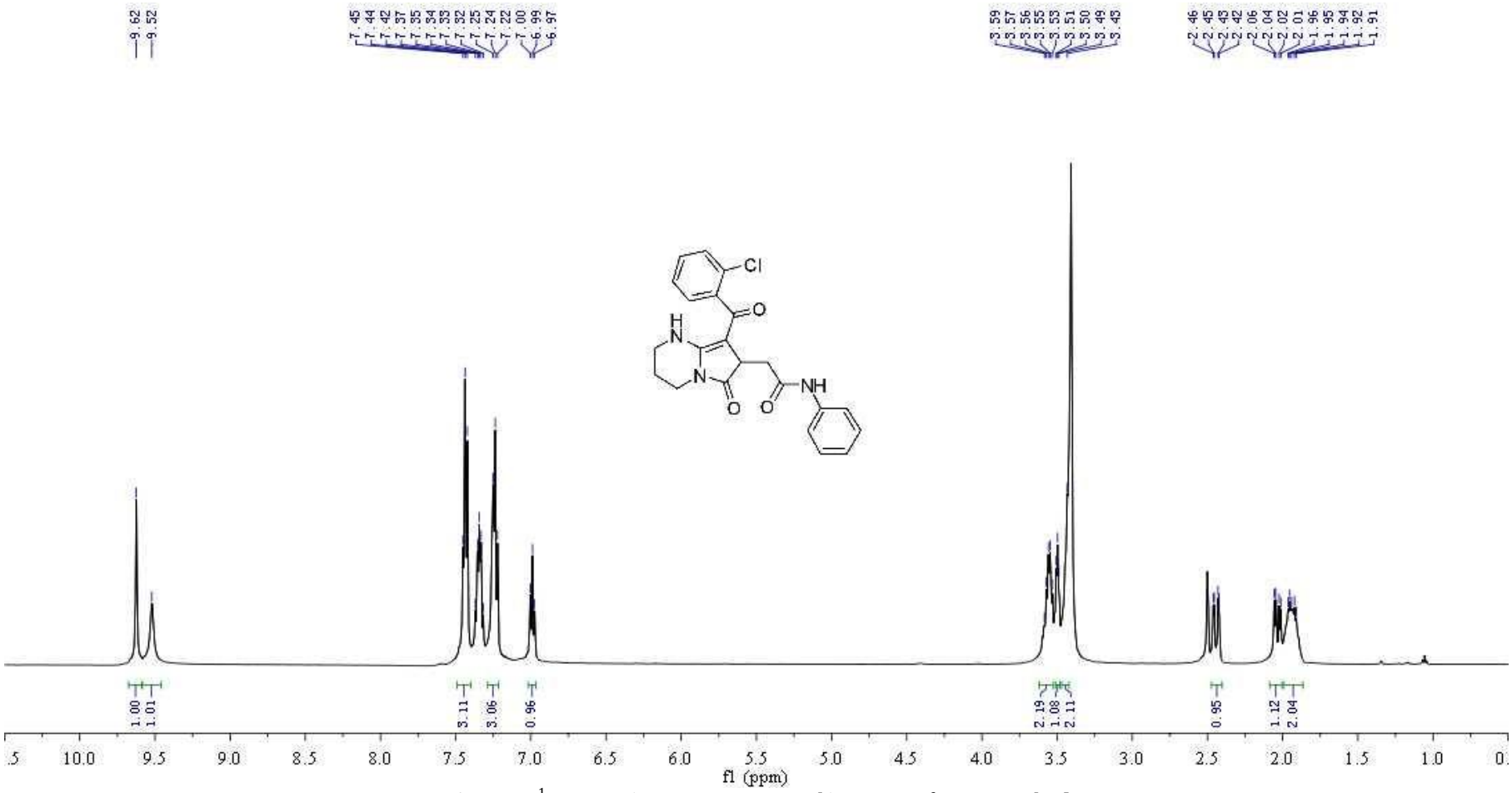


Figure 7. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3d**

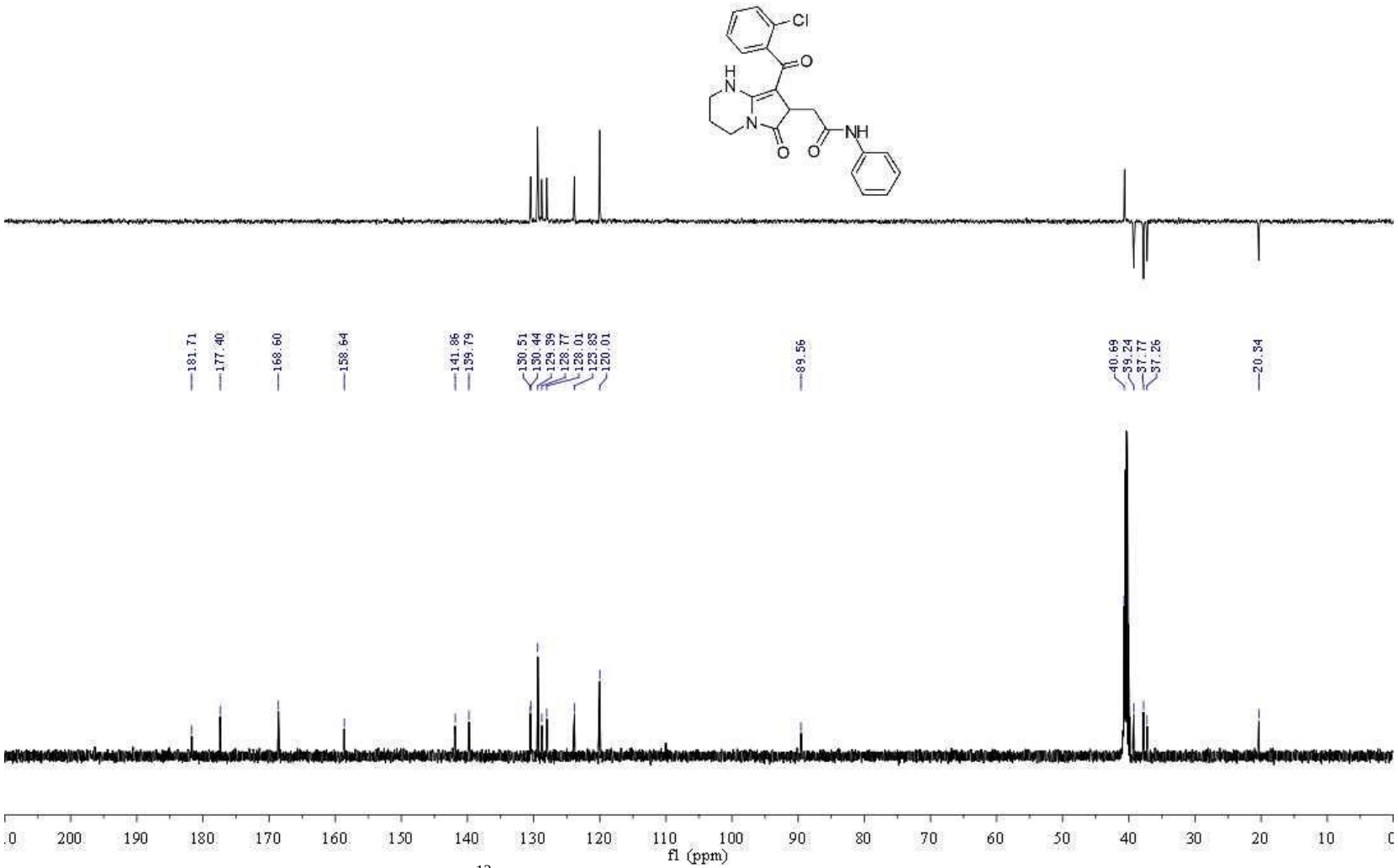


Figure 8. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3d**

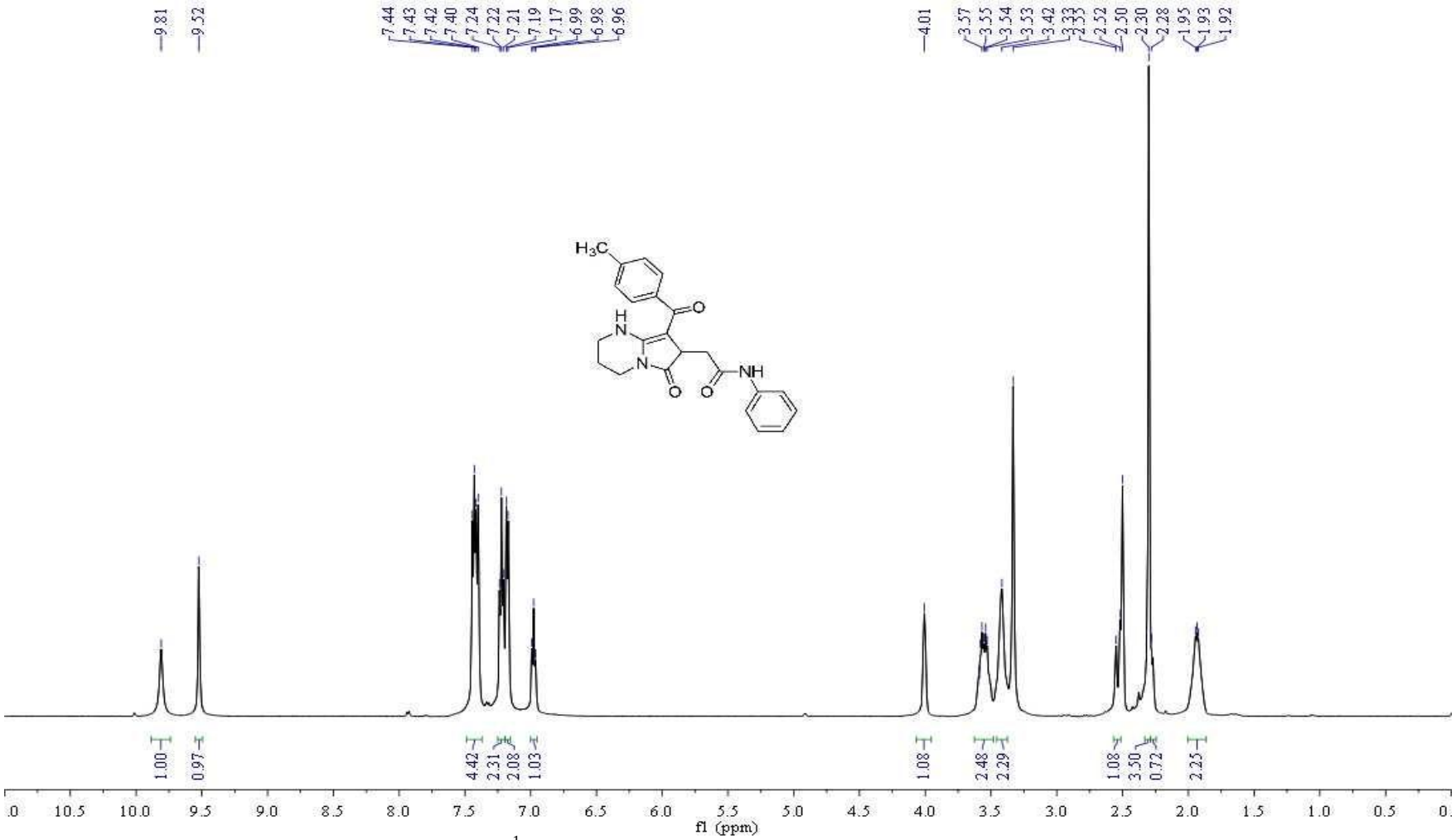


Figure 9. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3e**

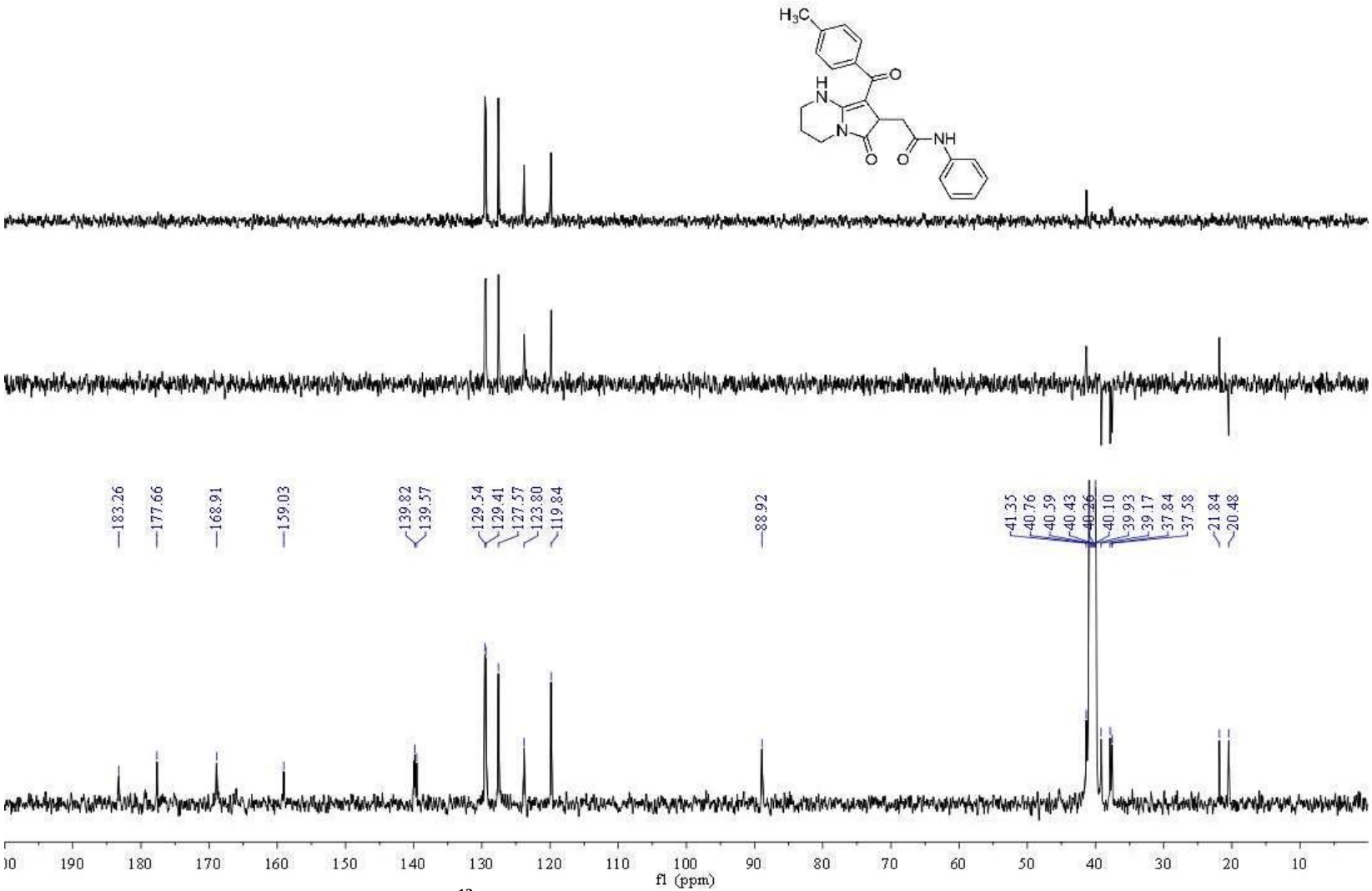


Figure 10 ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound 3e

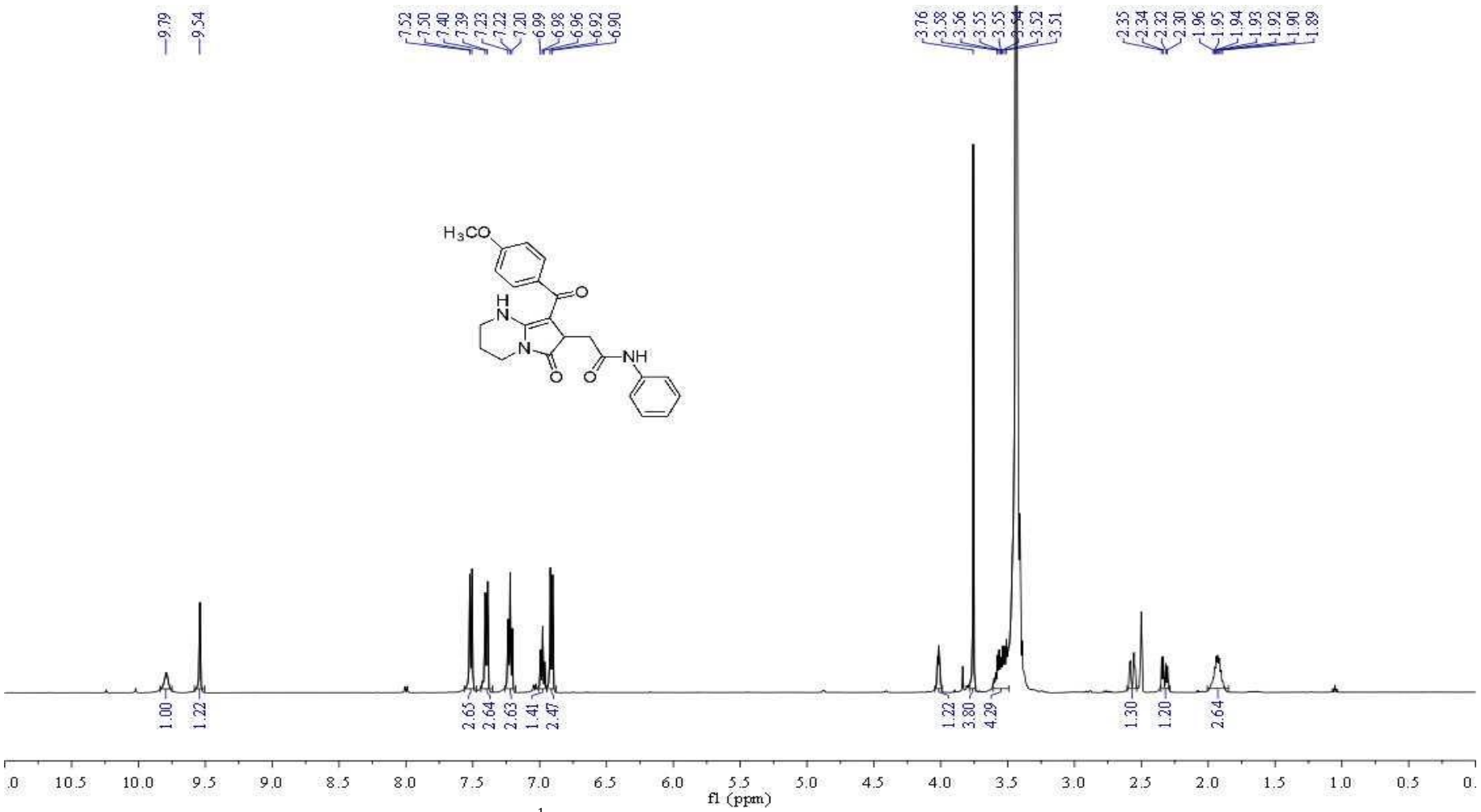


Figure 11. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3f**

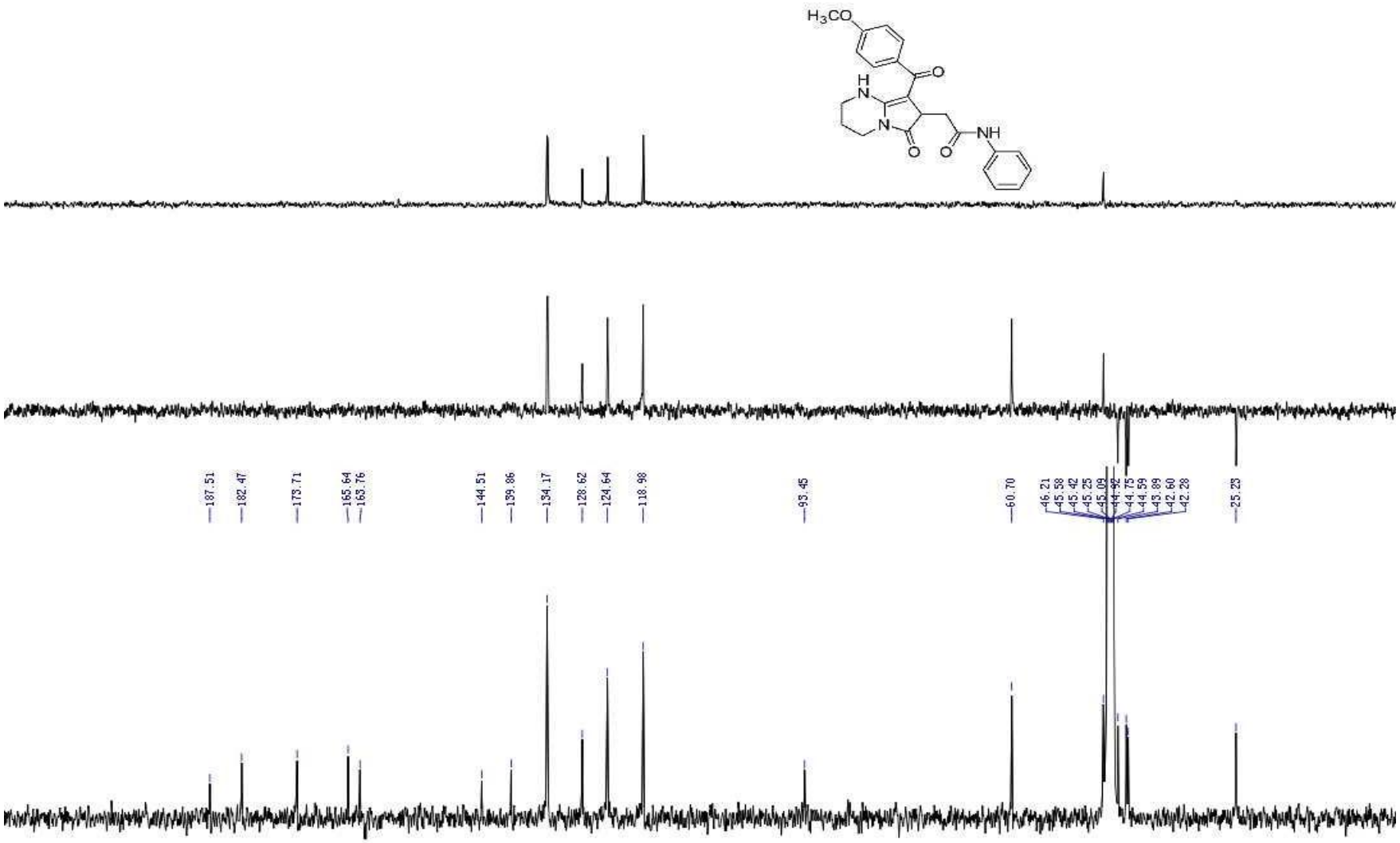


Figure 12. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3f**

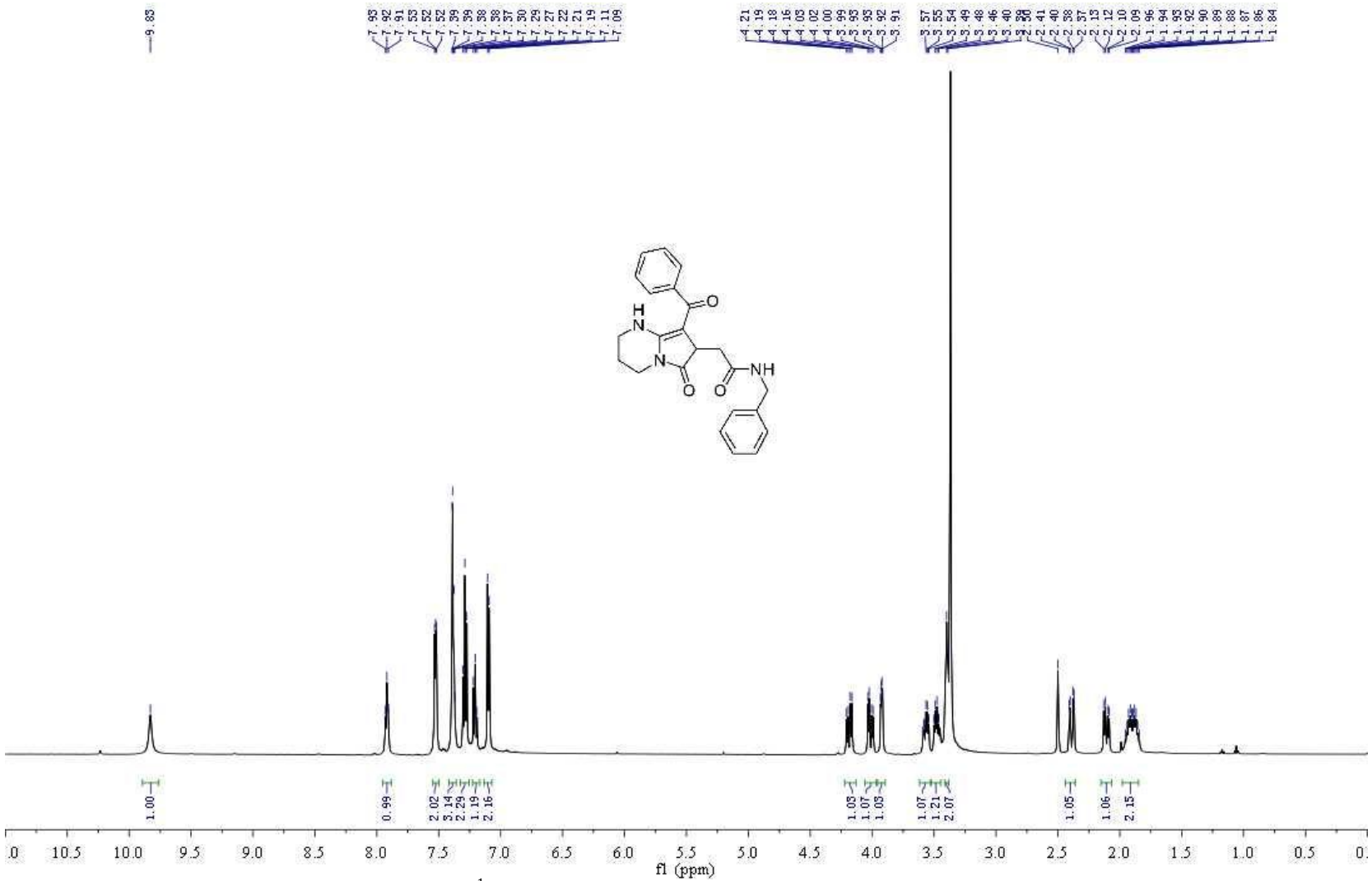


Figure 13. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3g**

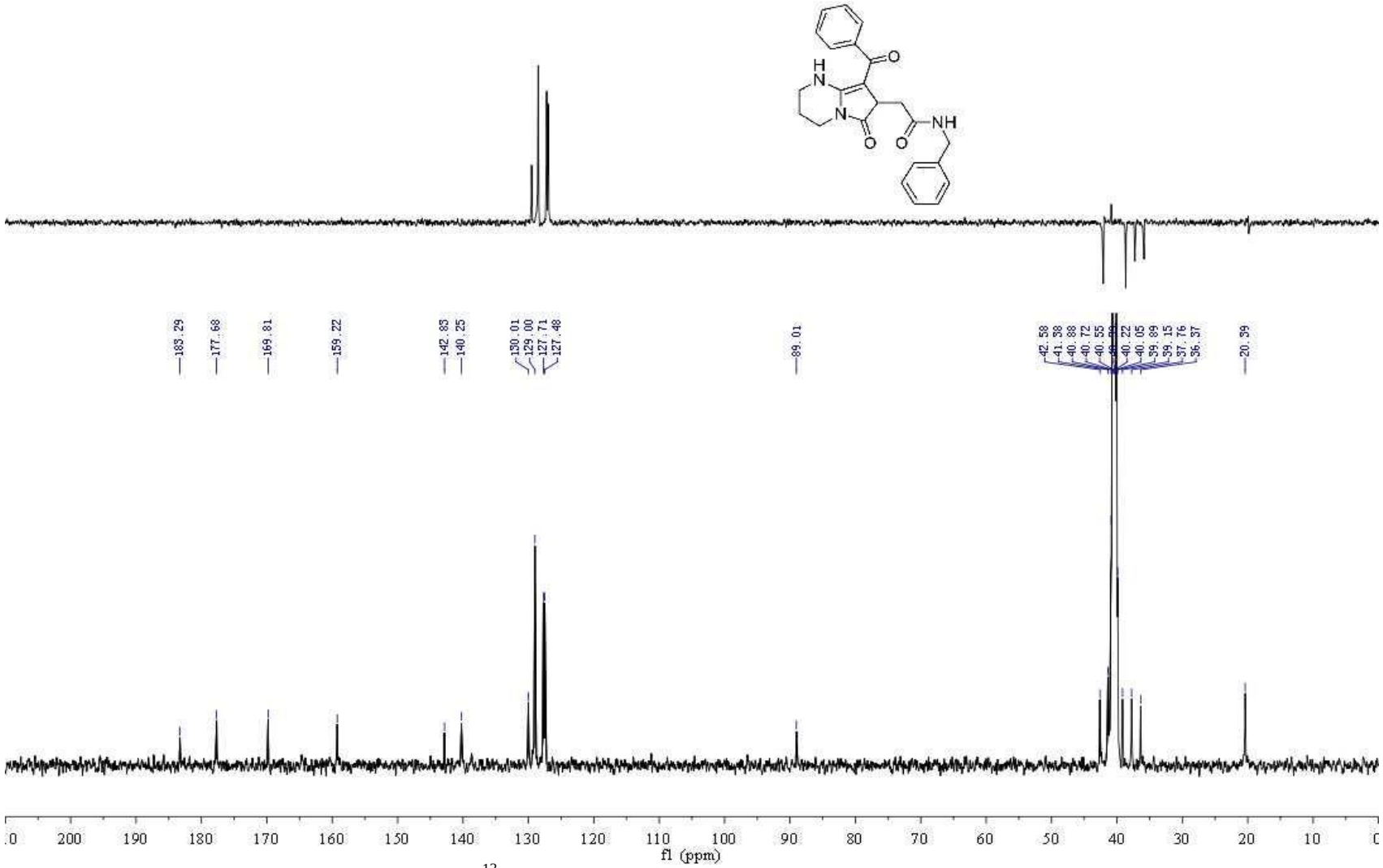


Figure 14. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3g**

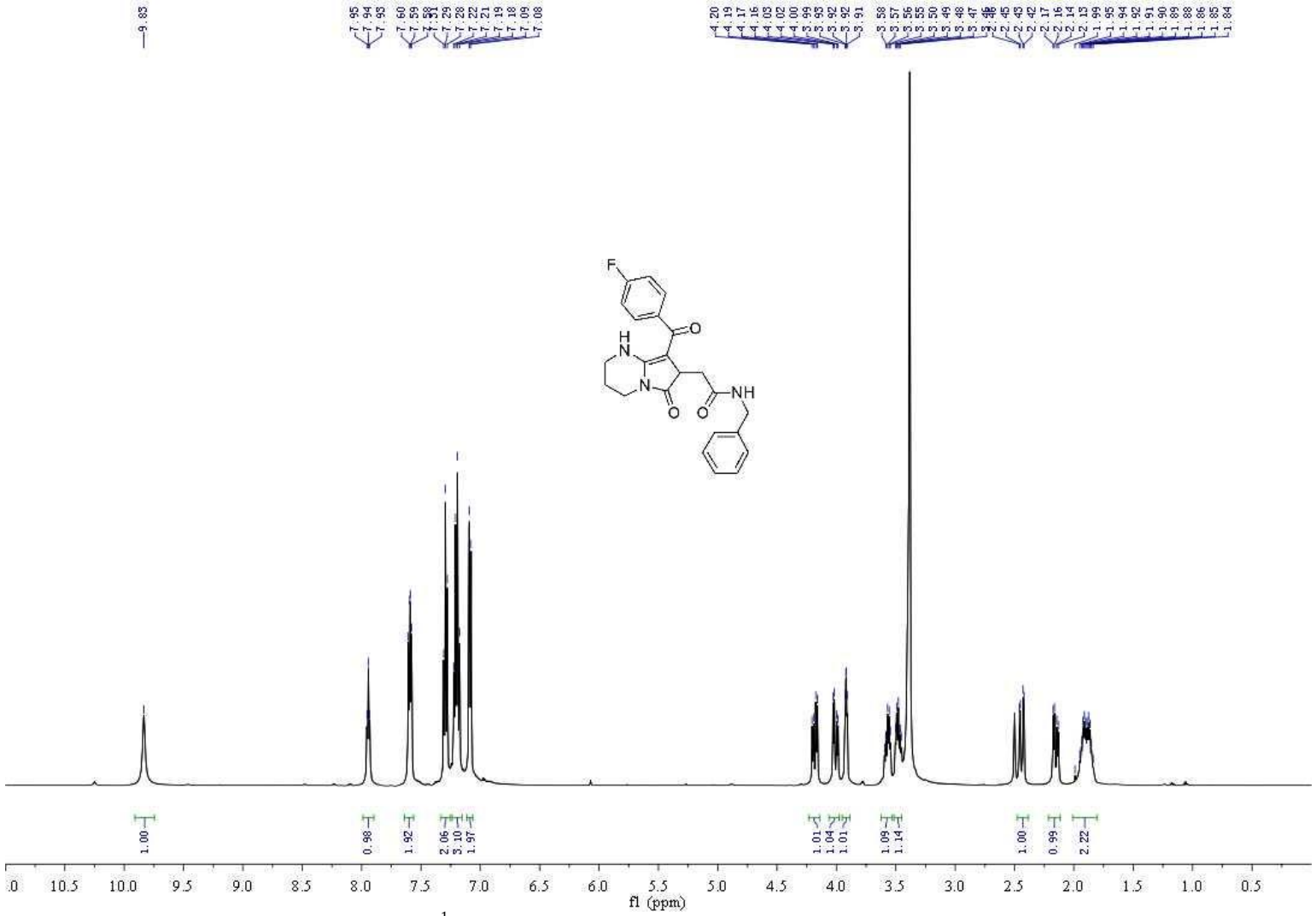


Figure 15. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3h**

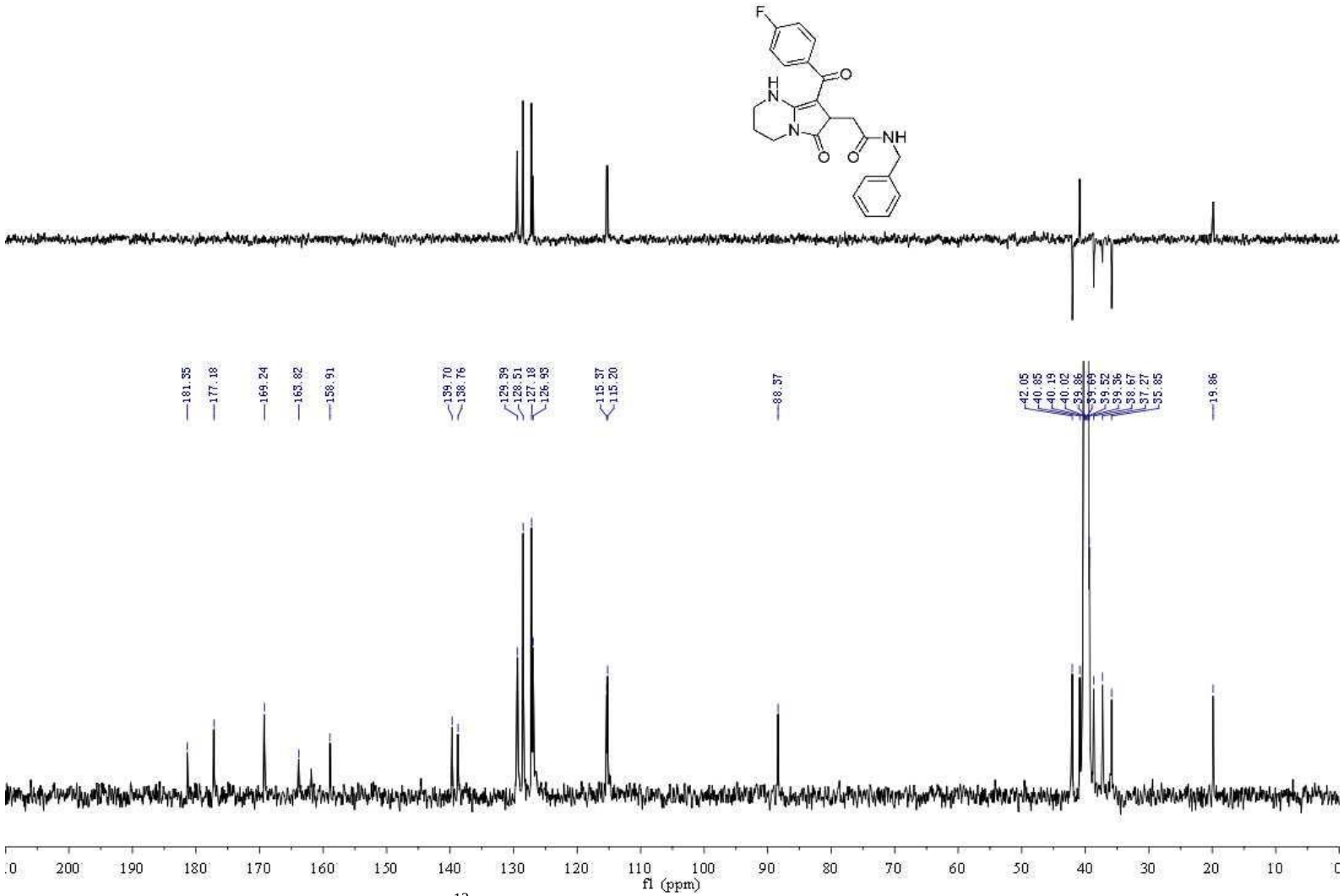


Figure 16. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3h**

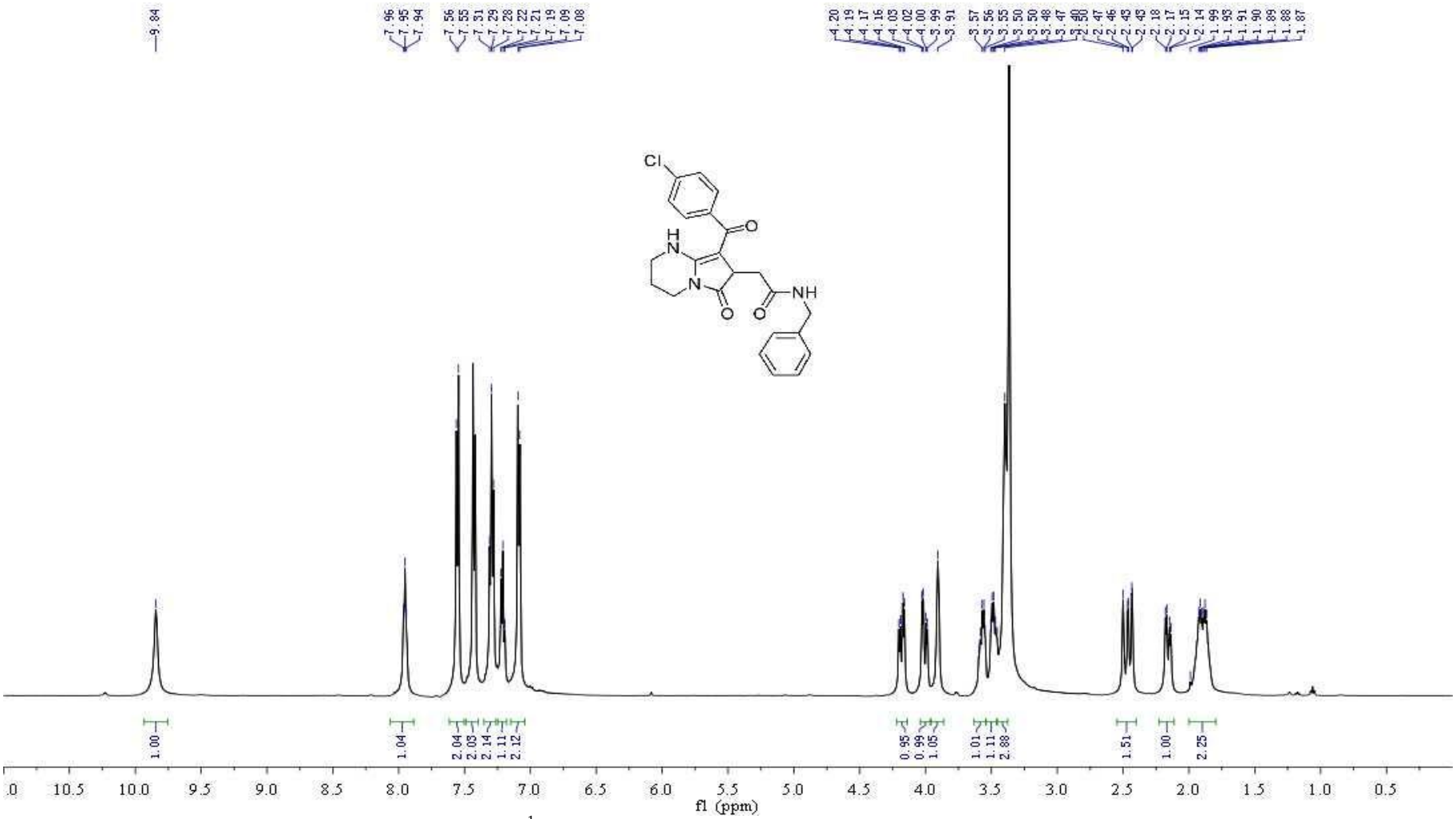


Figure 17. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3i**

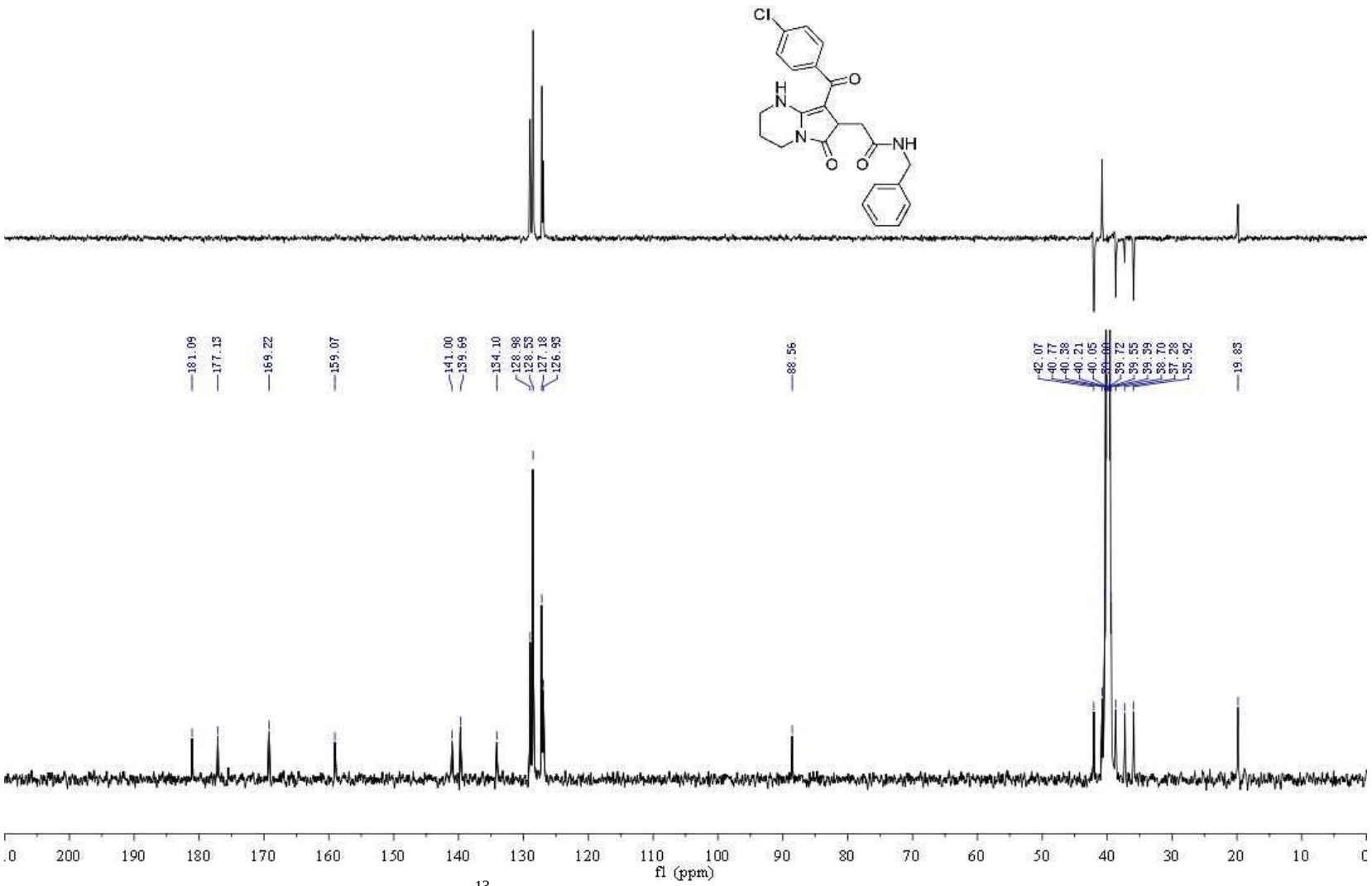


Figure 18. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3i**

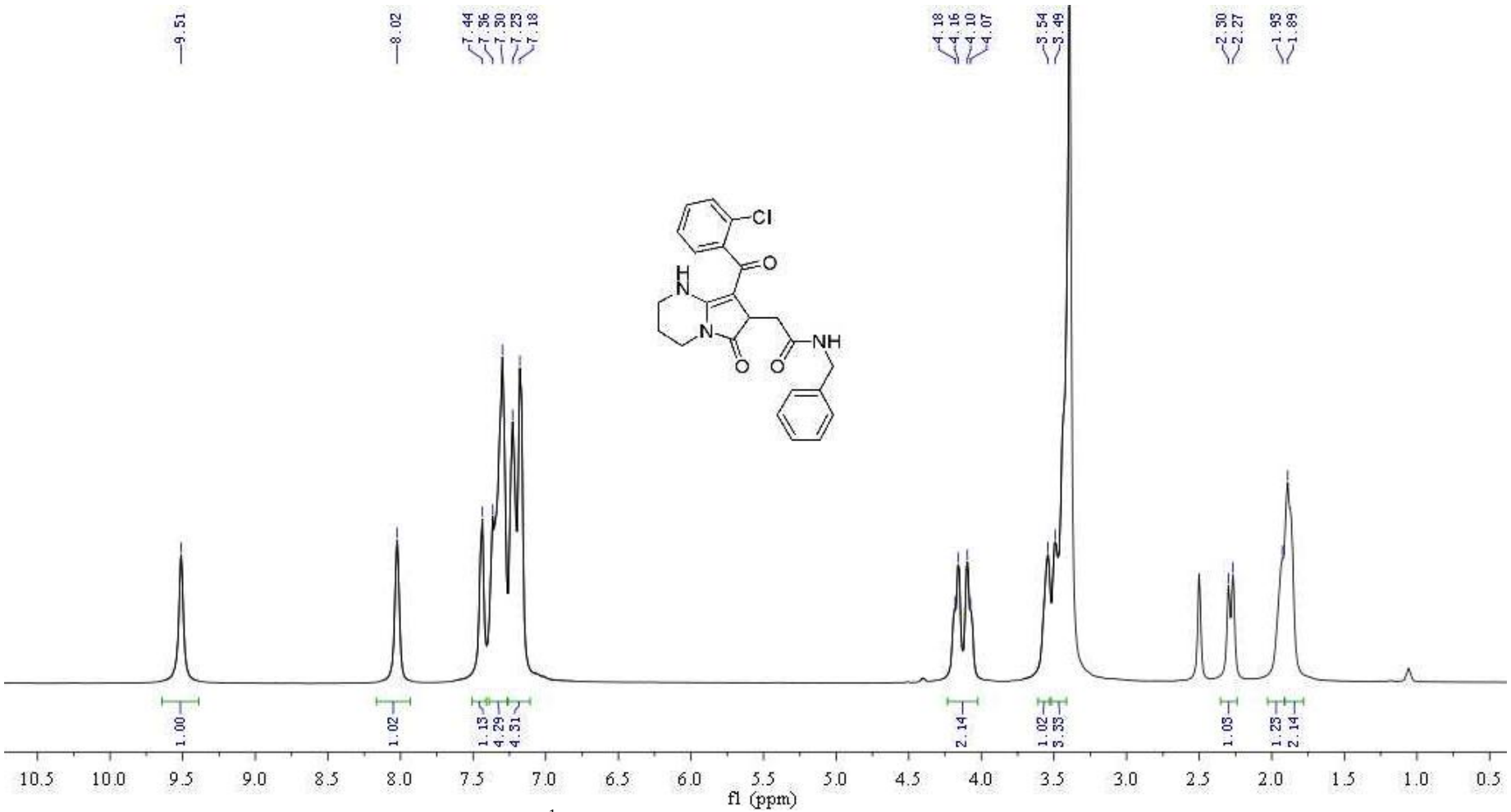


Figure 19. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3j**

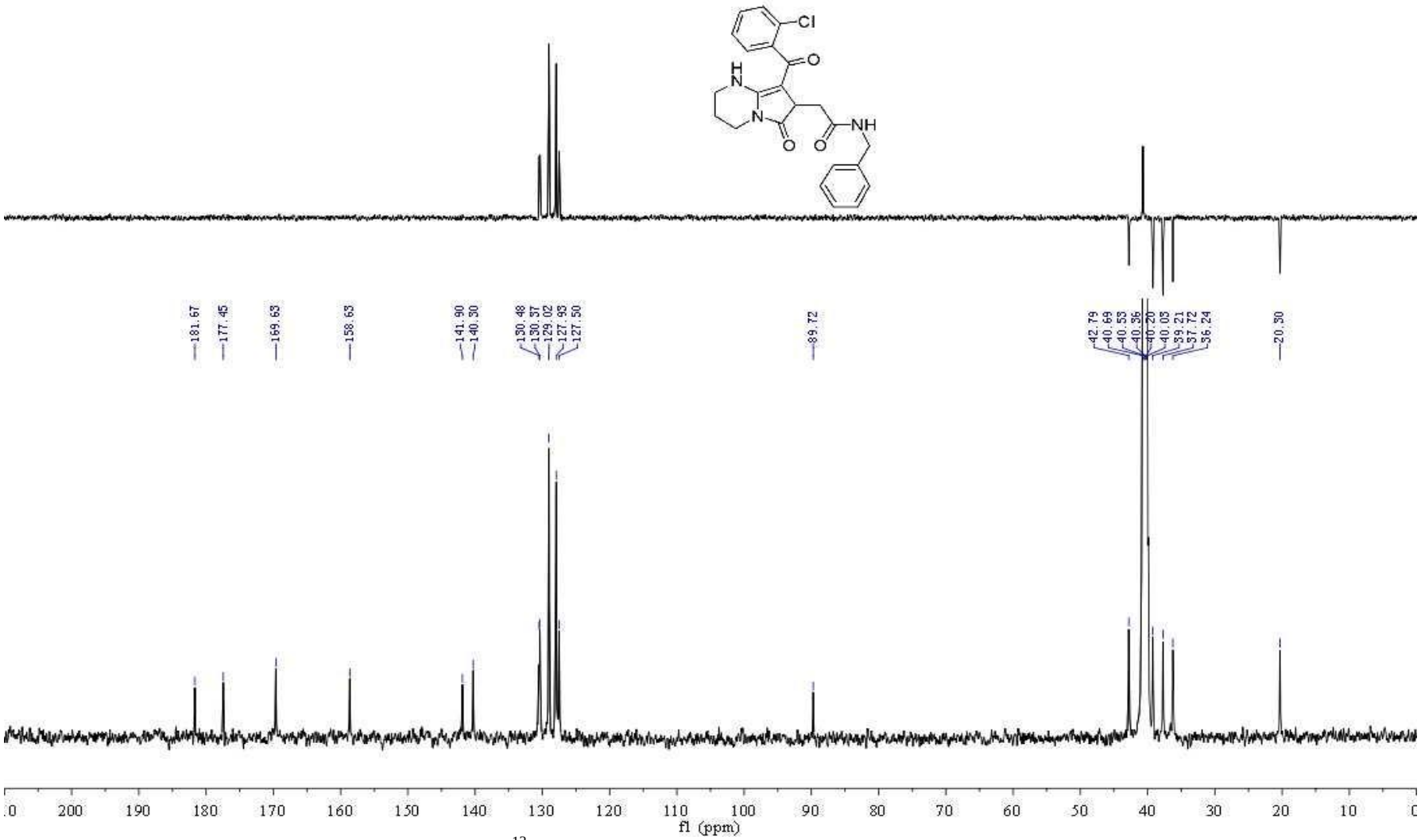


Figure 20. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3j**

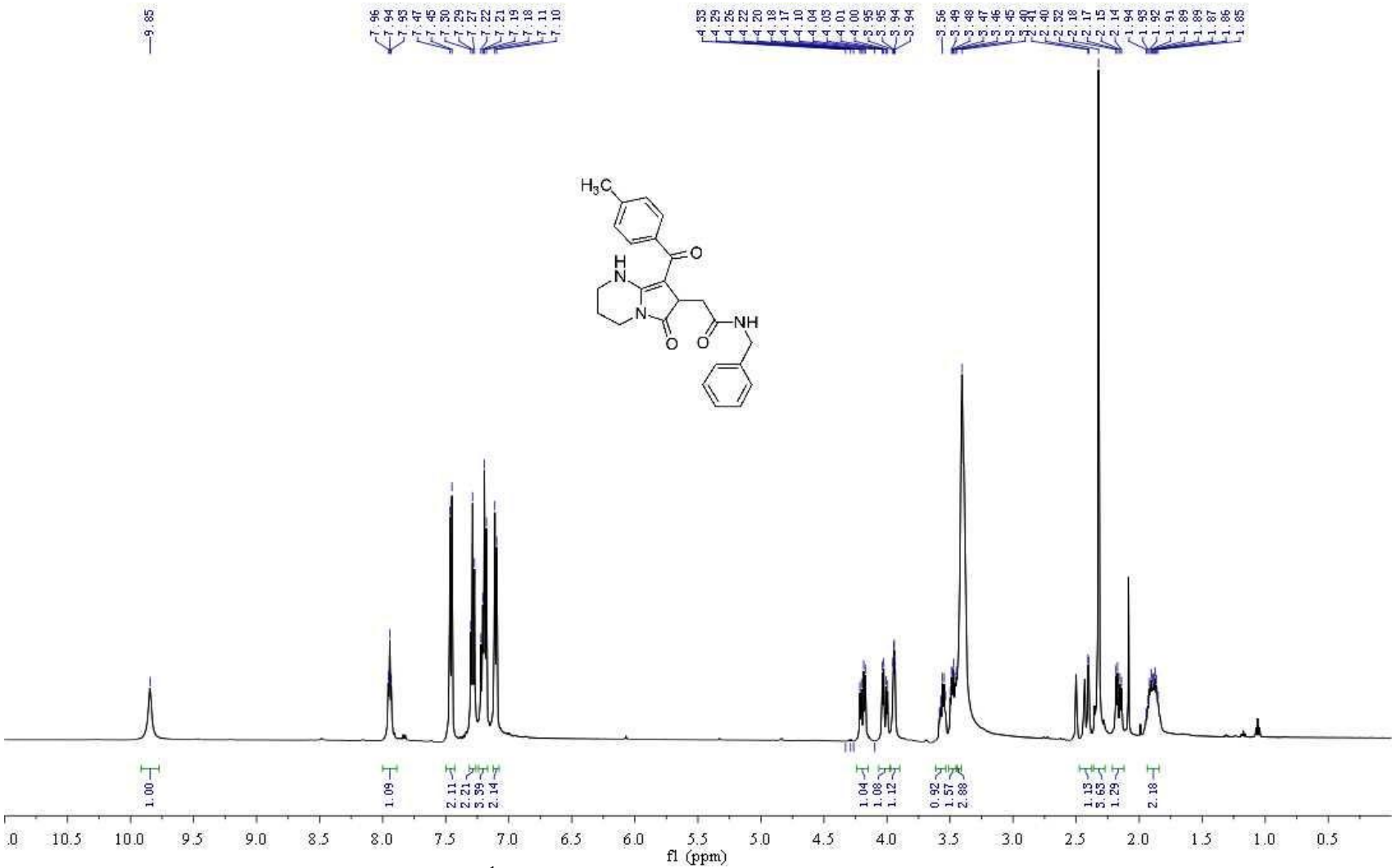


Figure 21. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3k**

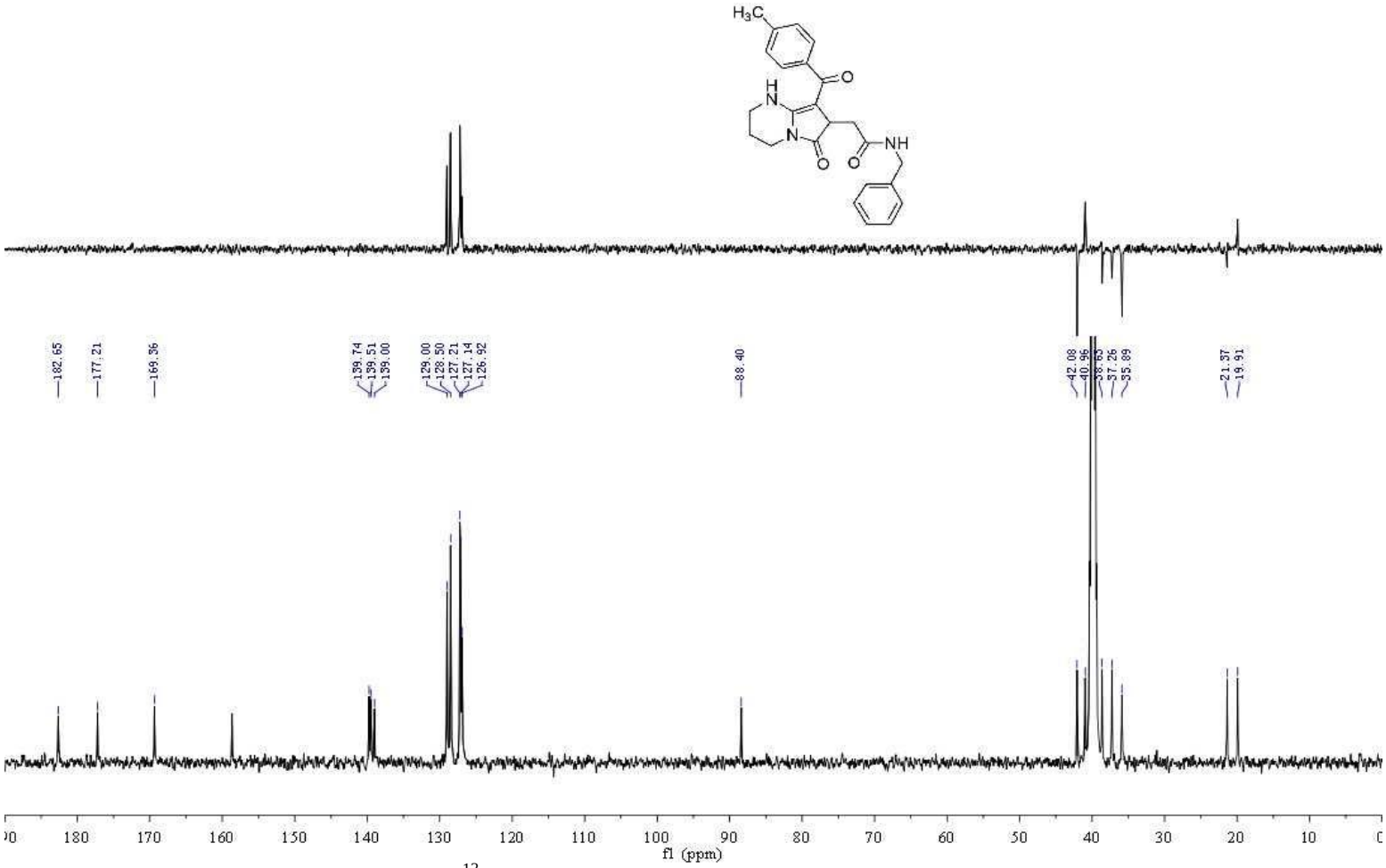


Figure 22. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3k**

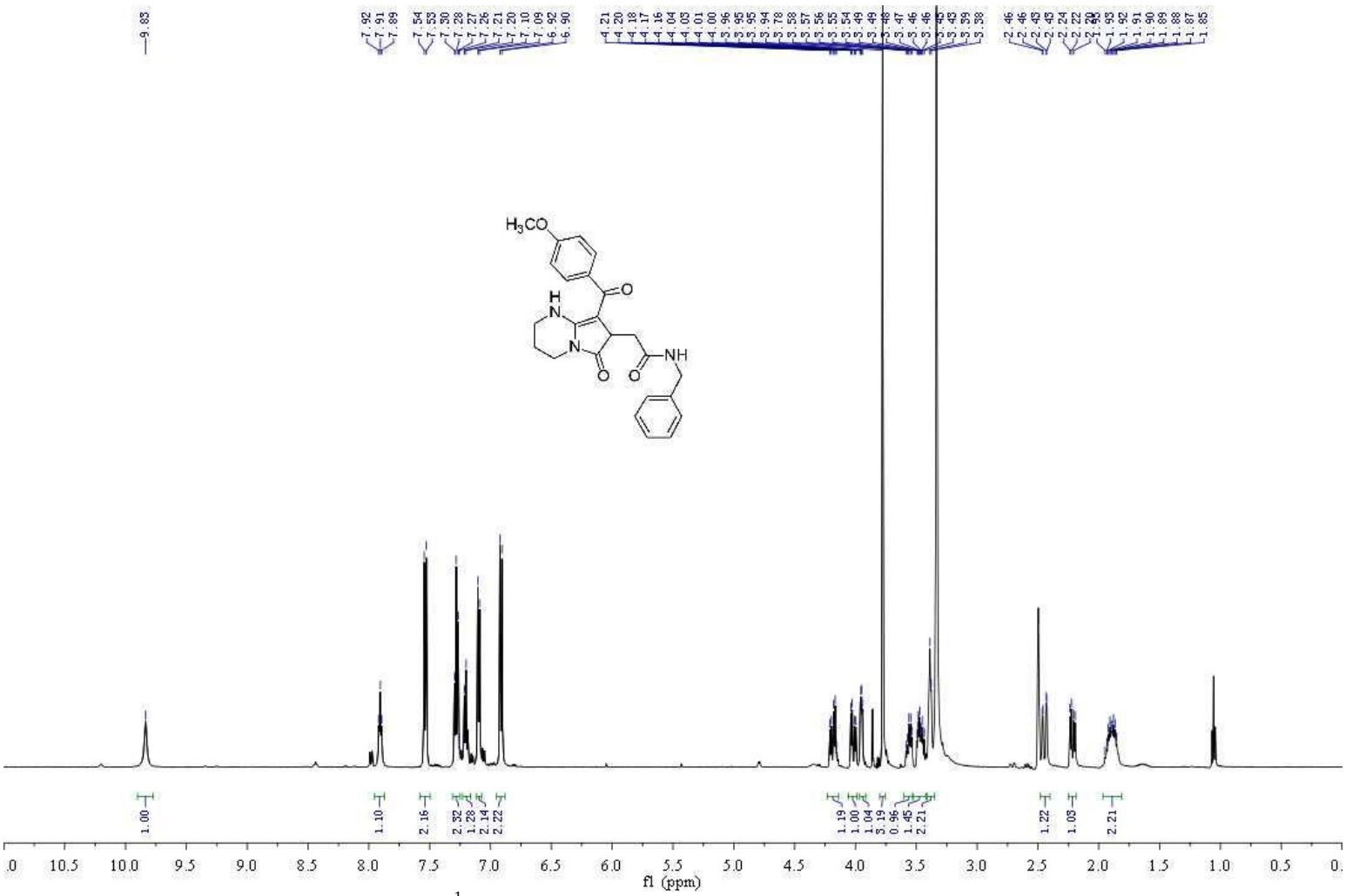


Figure 23. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3l**

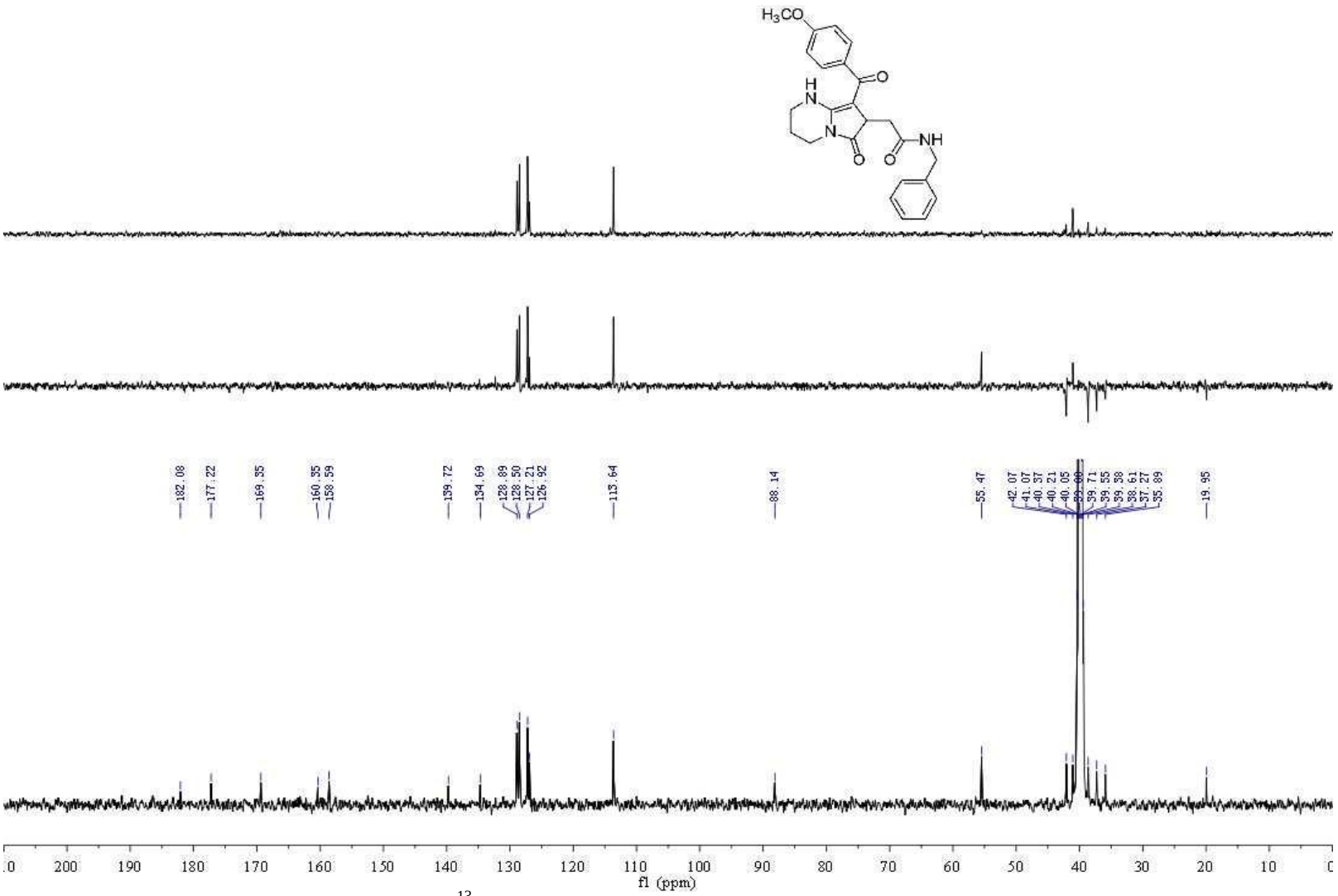


Figure 24. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3l**

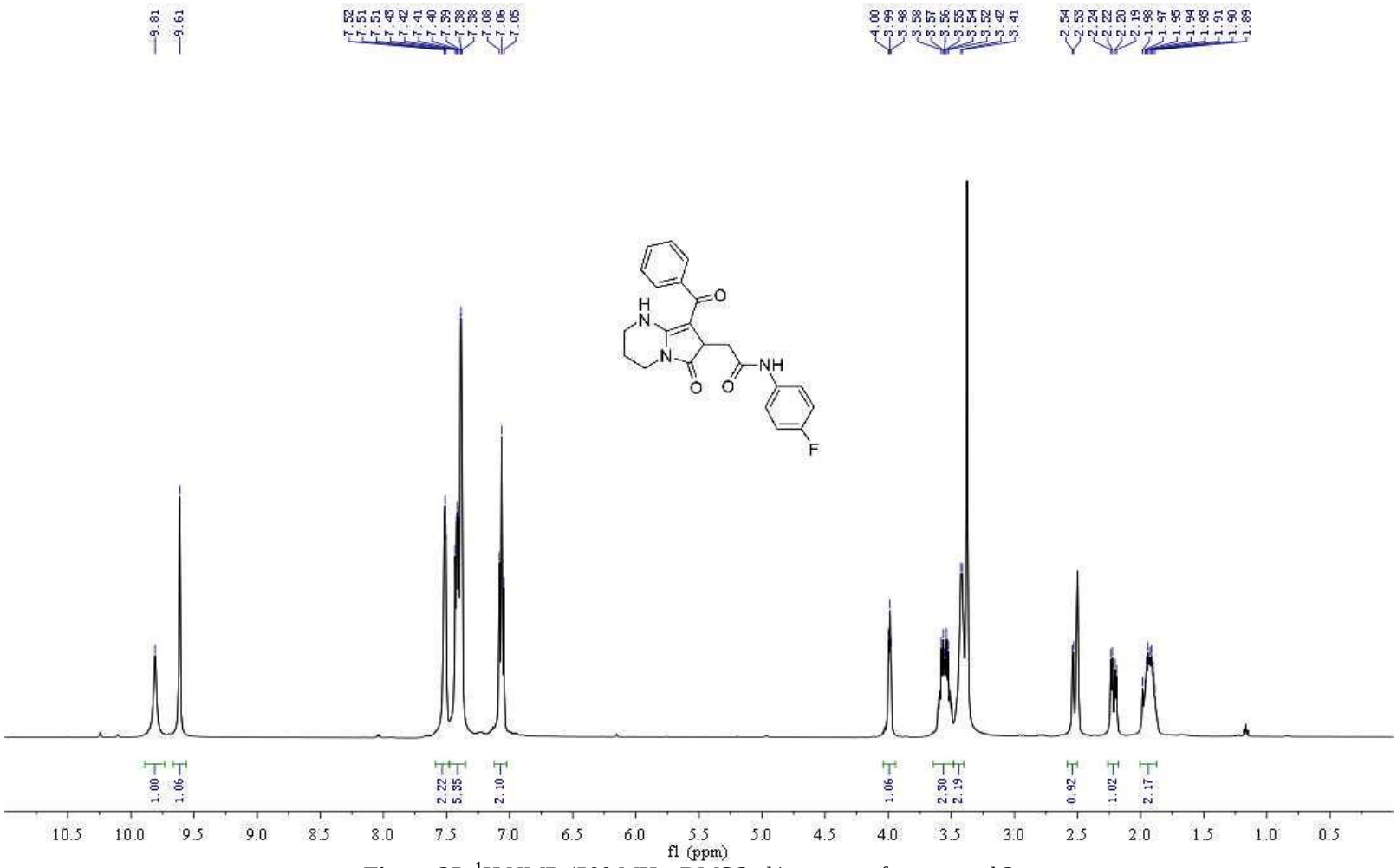


Figure 25. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3m**

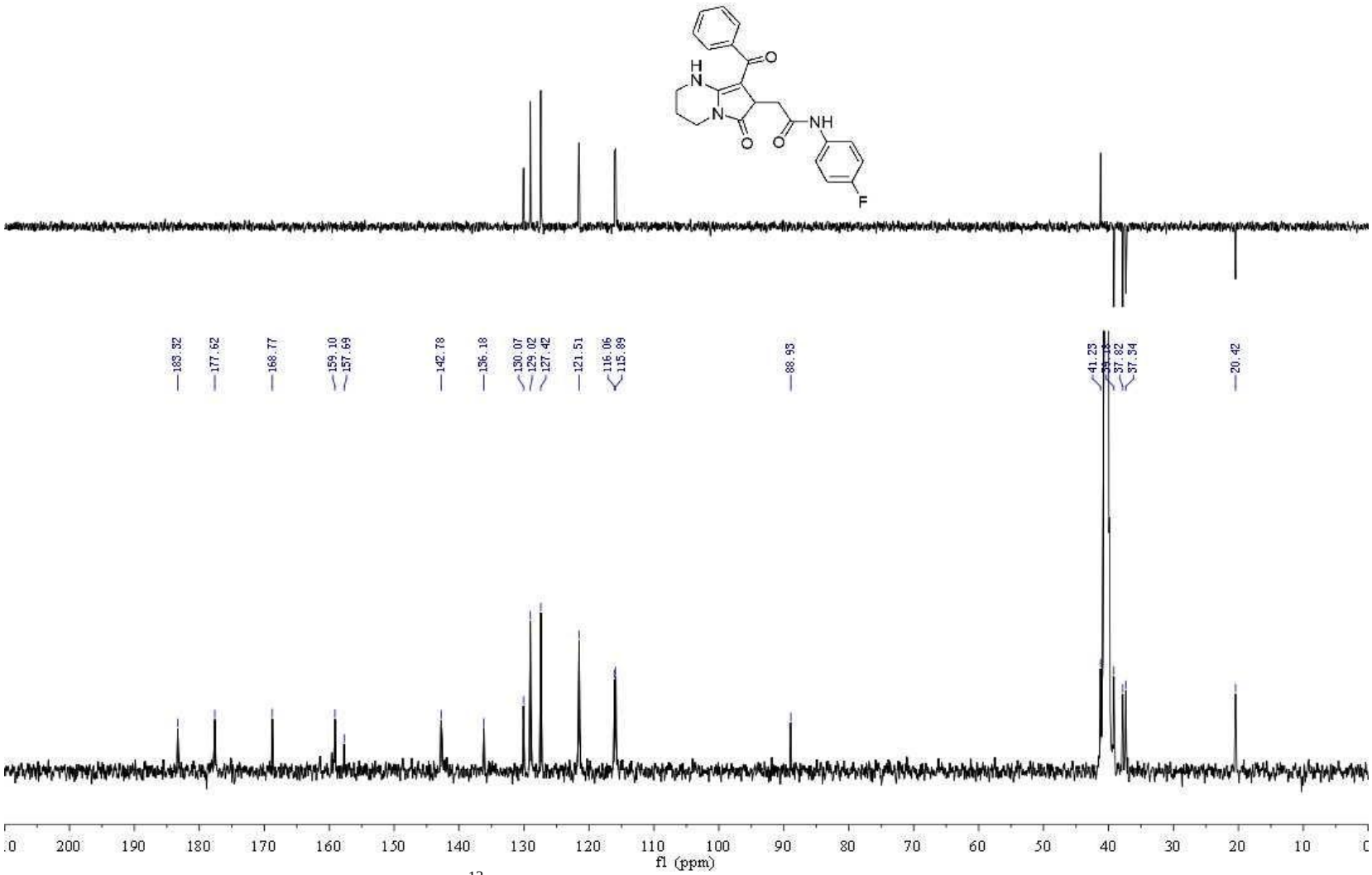


Figure 26. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3m**

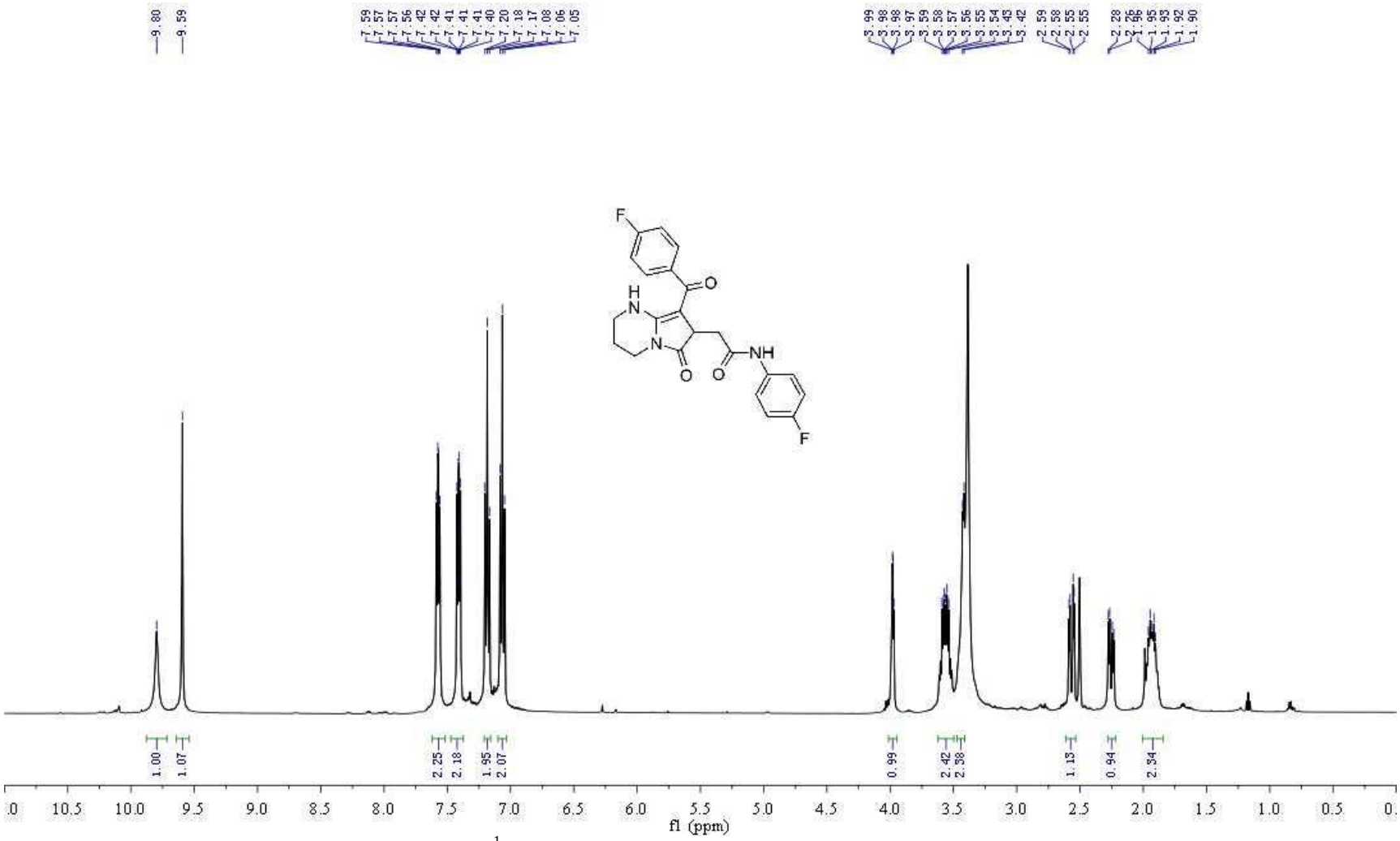


Figure 27. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3n**

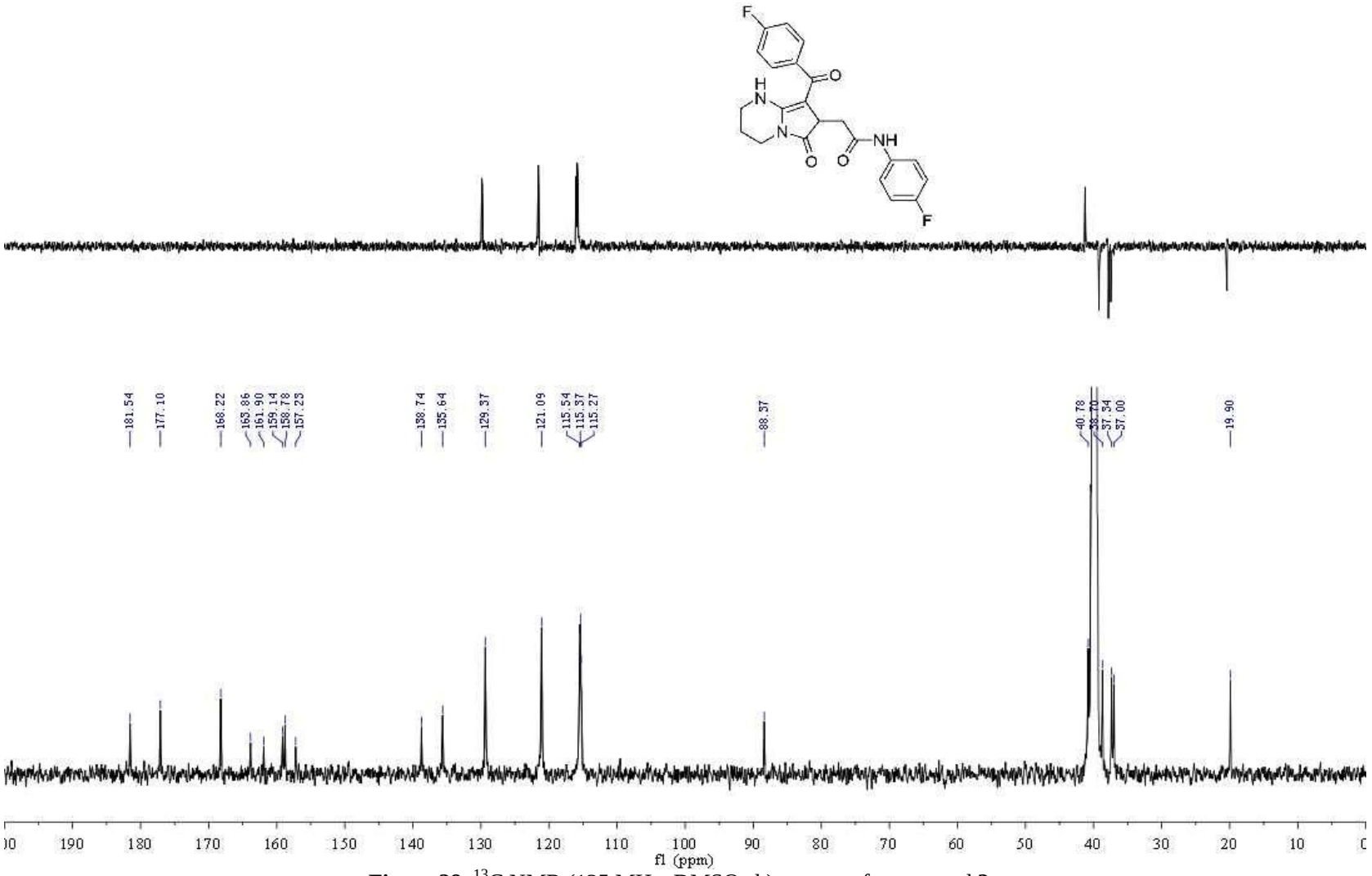


Figure 28. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3n**

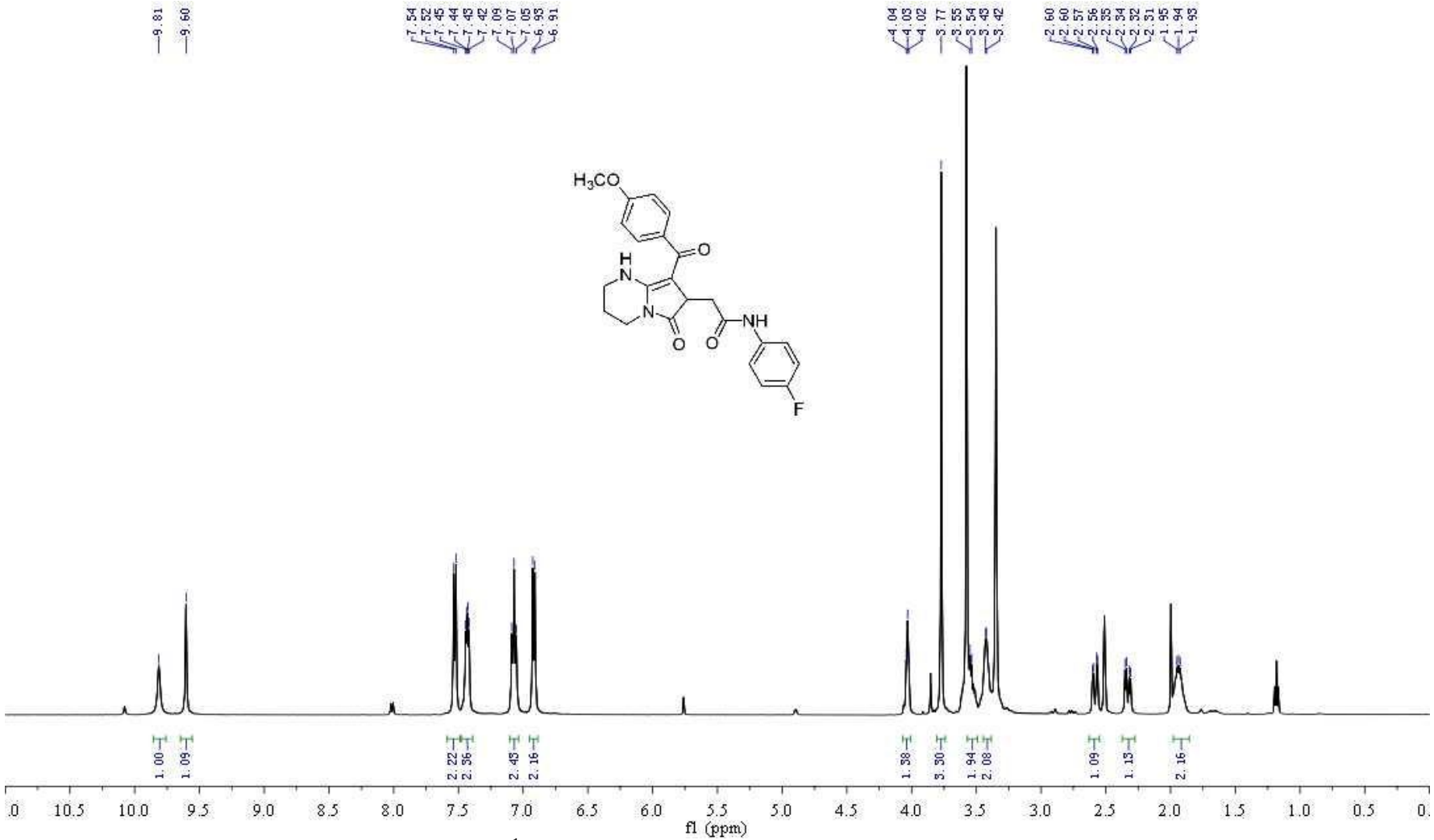


Figure 29. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3o**

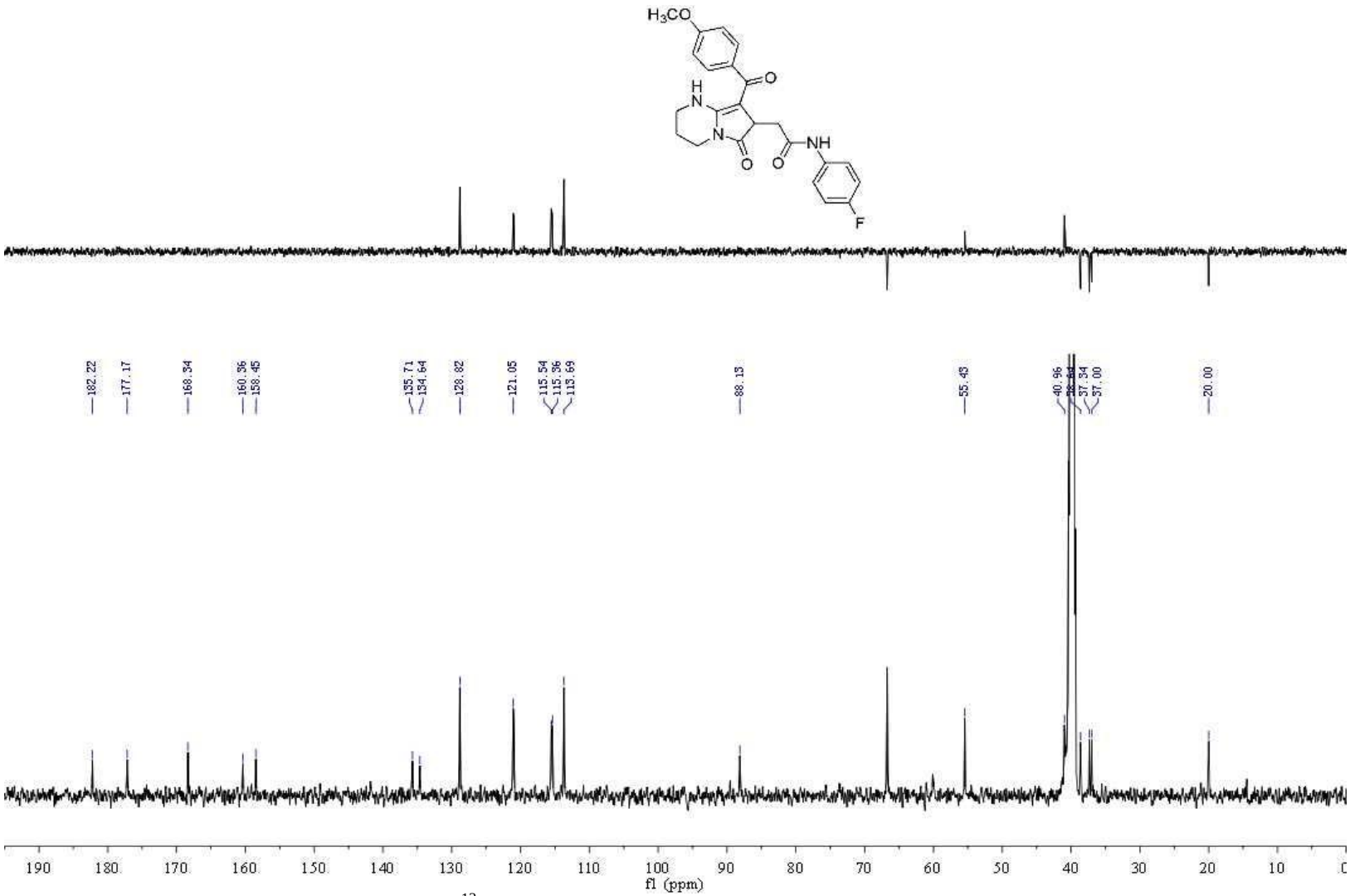


Figure 30. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3o**

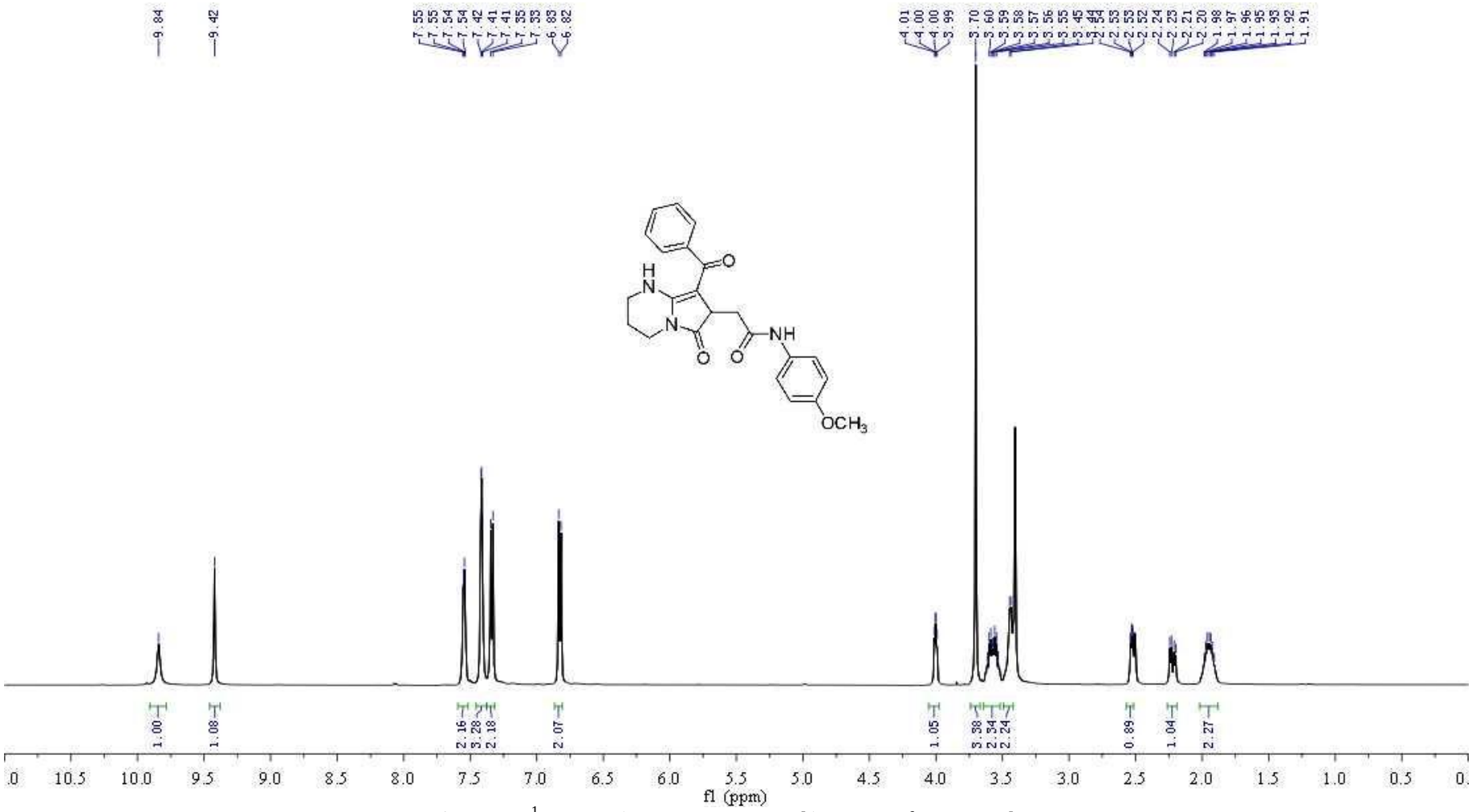


Figure 31. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3p**

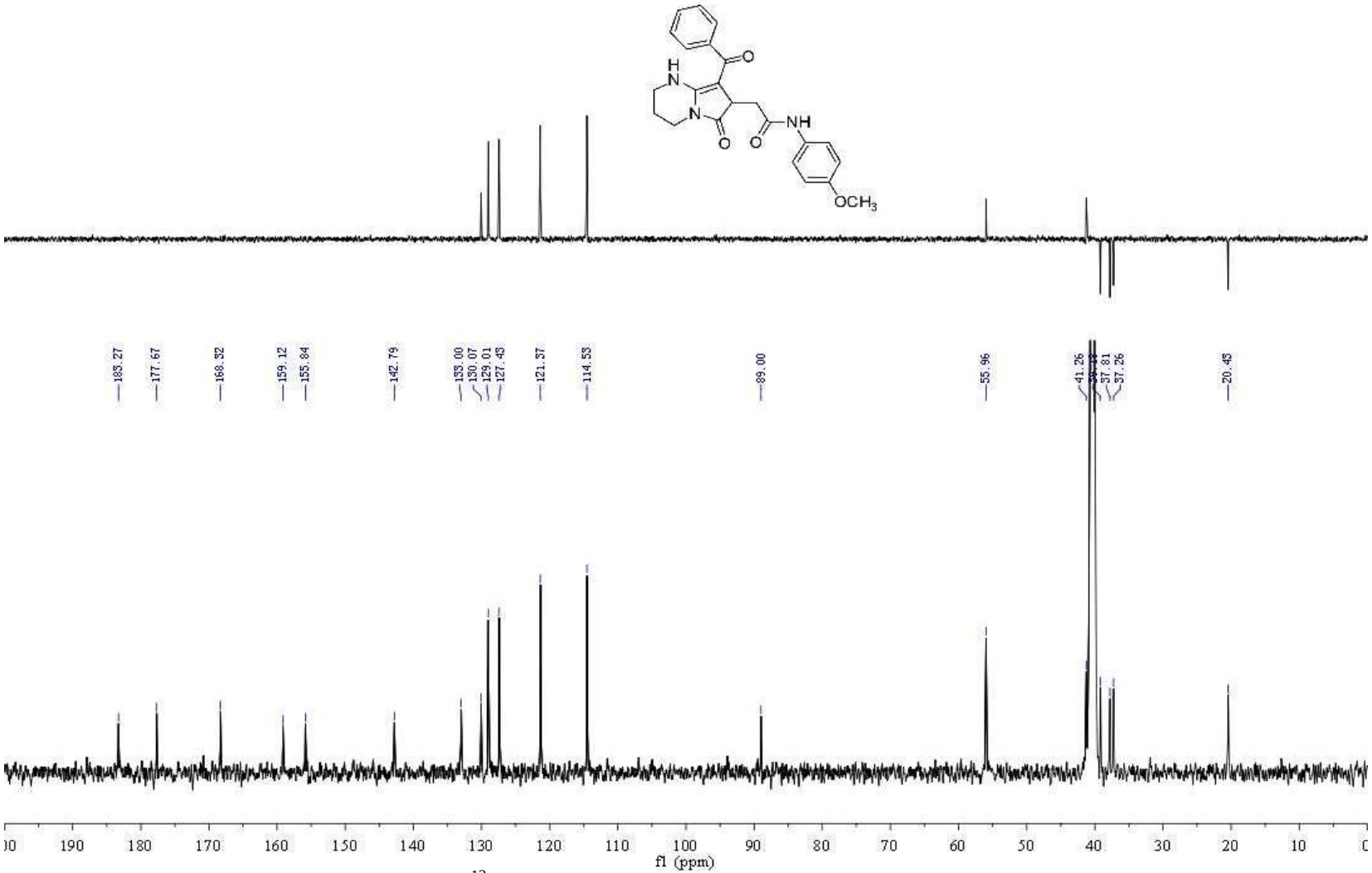


Figure 32. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3p**

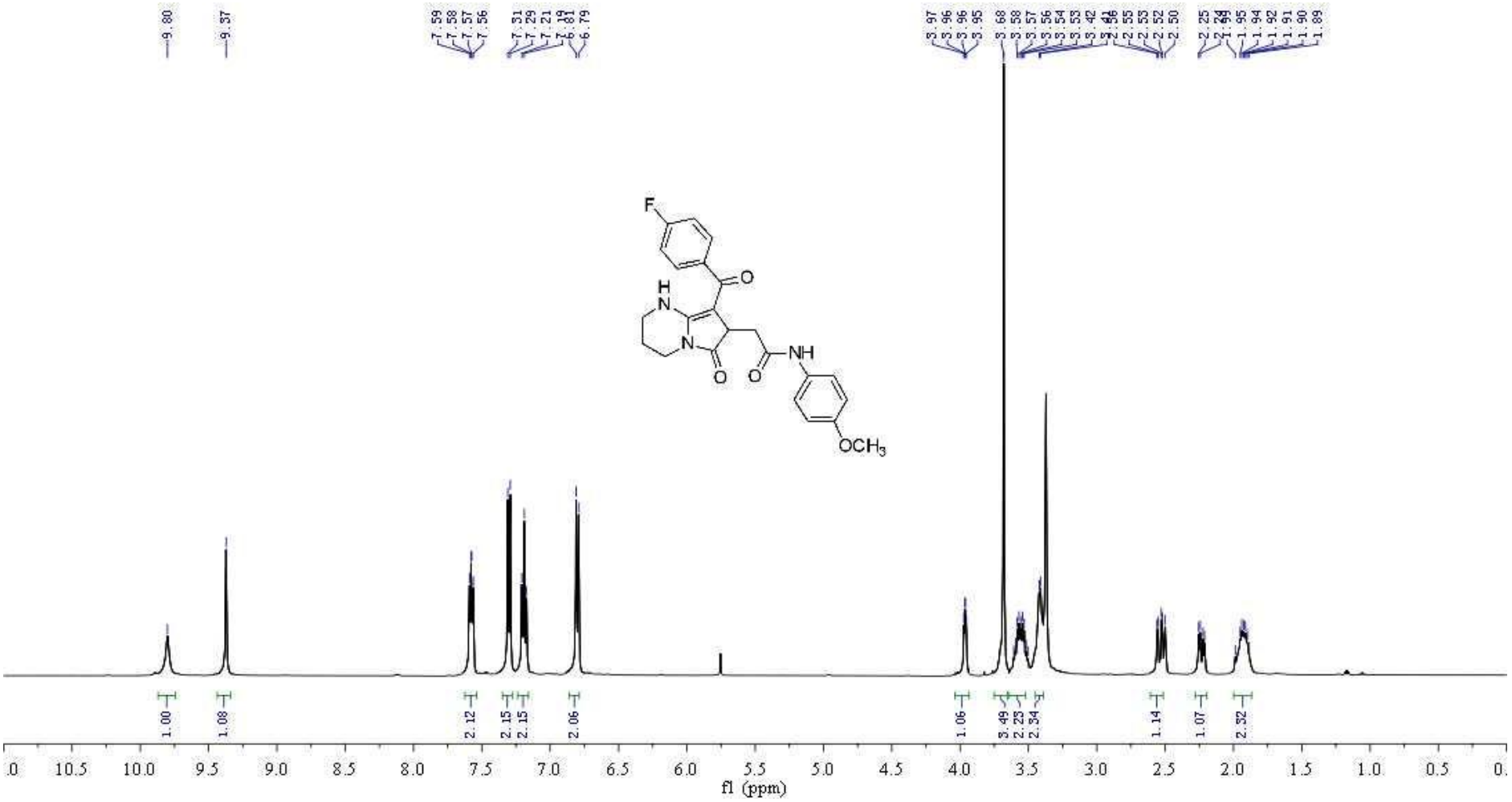


Figure 33. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3q**

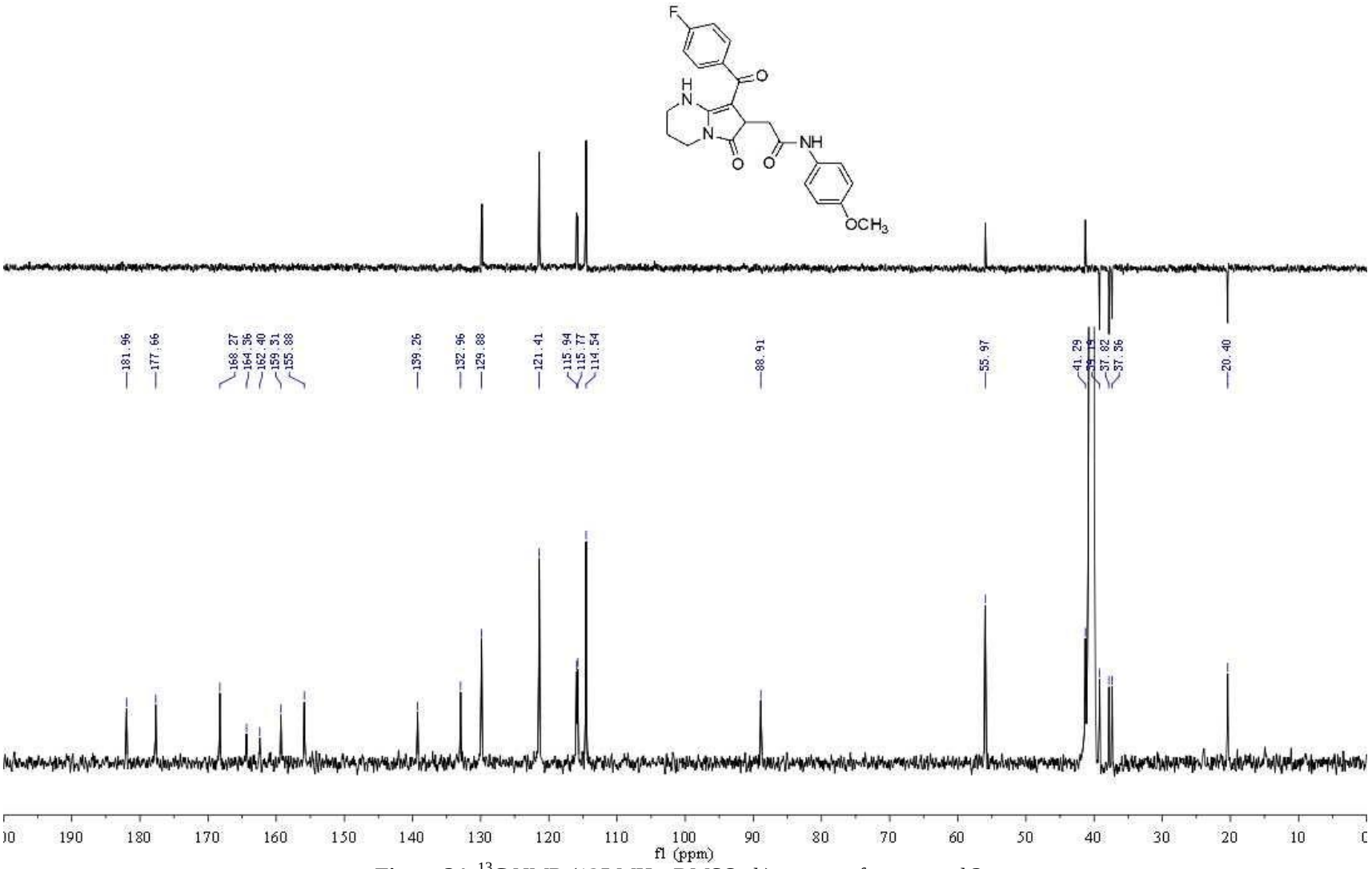


Figure 34. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3q**

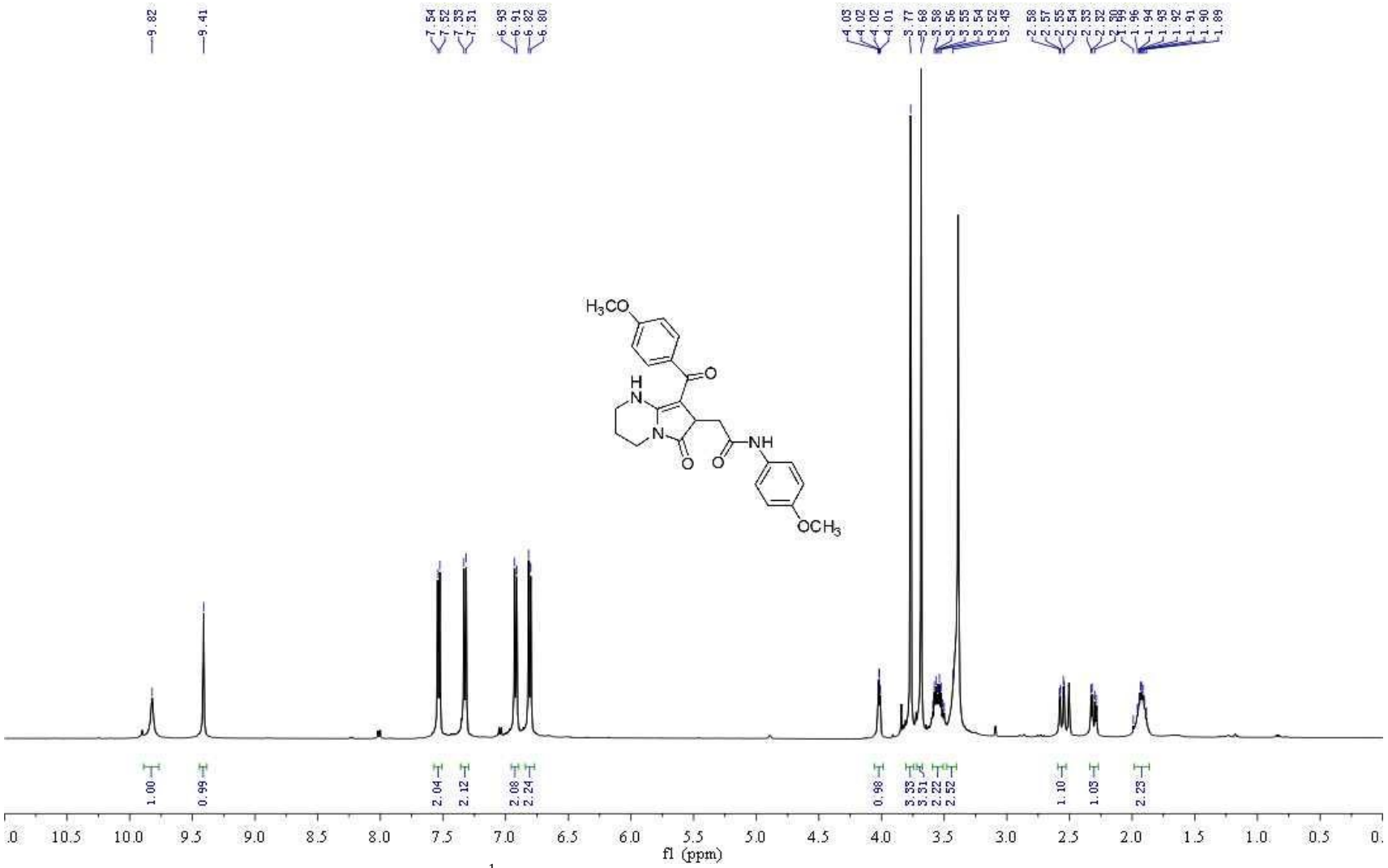


Figure 35. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3r**

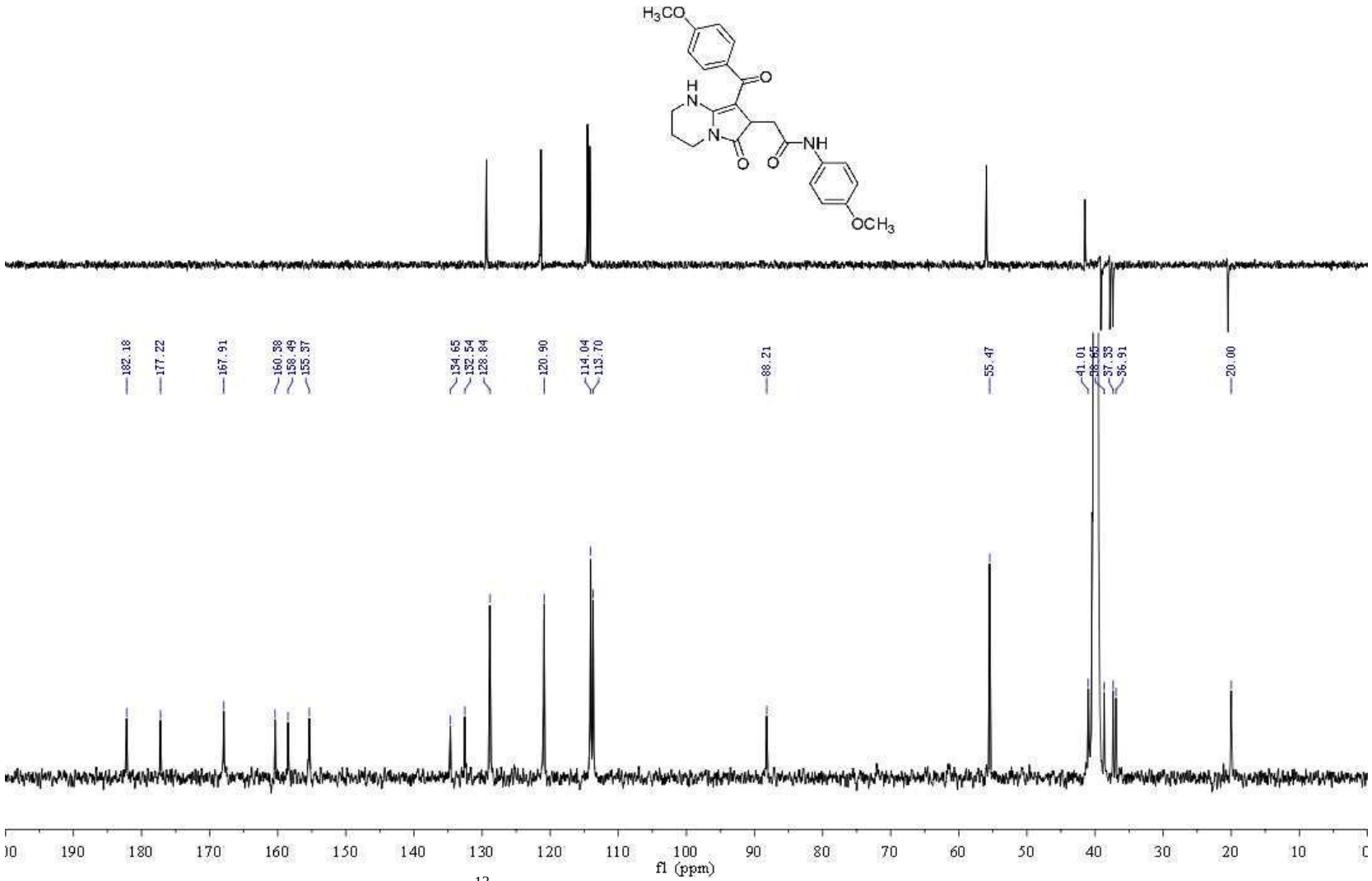


Figure 36. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3r**

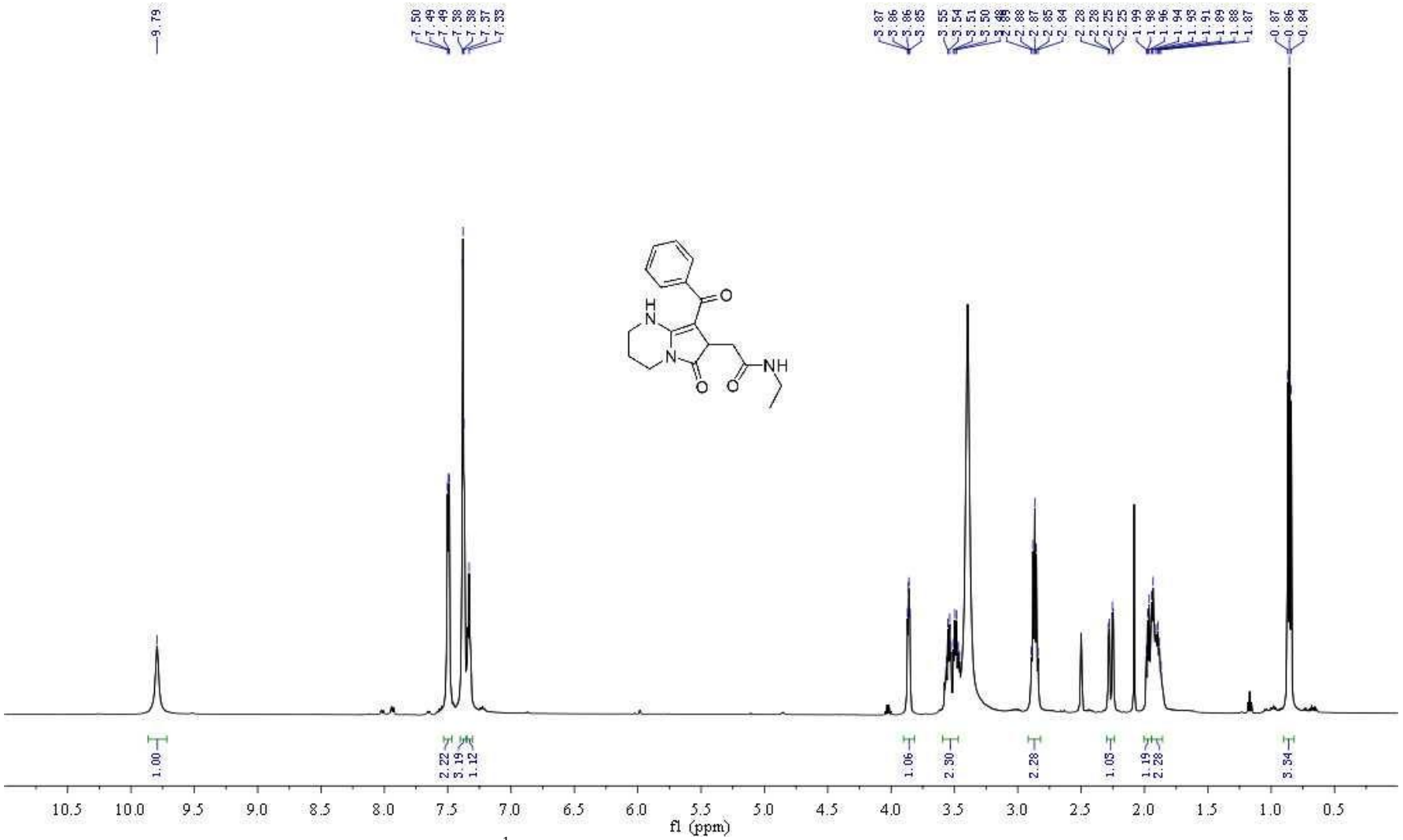


Figure 37. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3s**

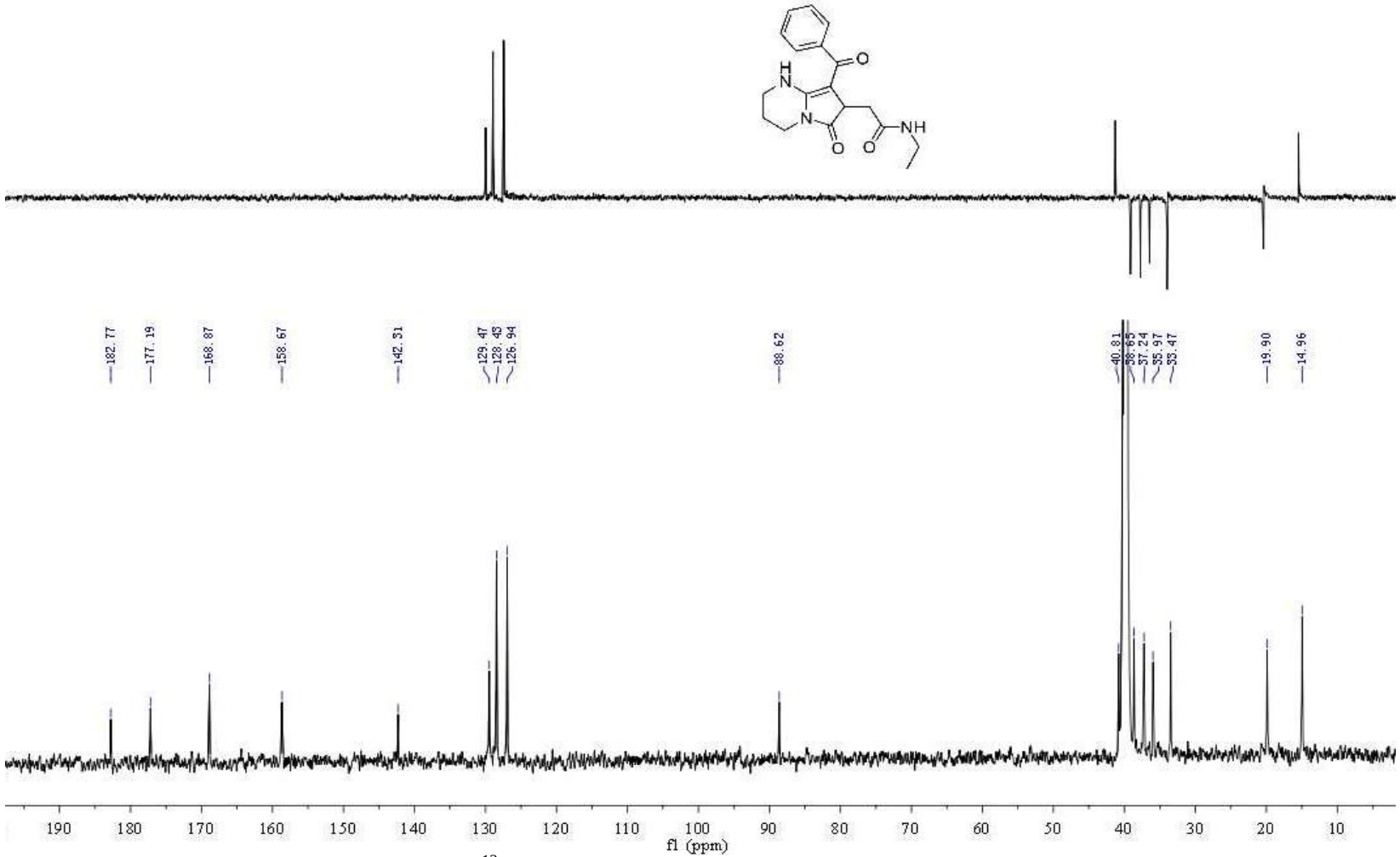


Figure 38. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3s**

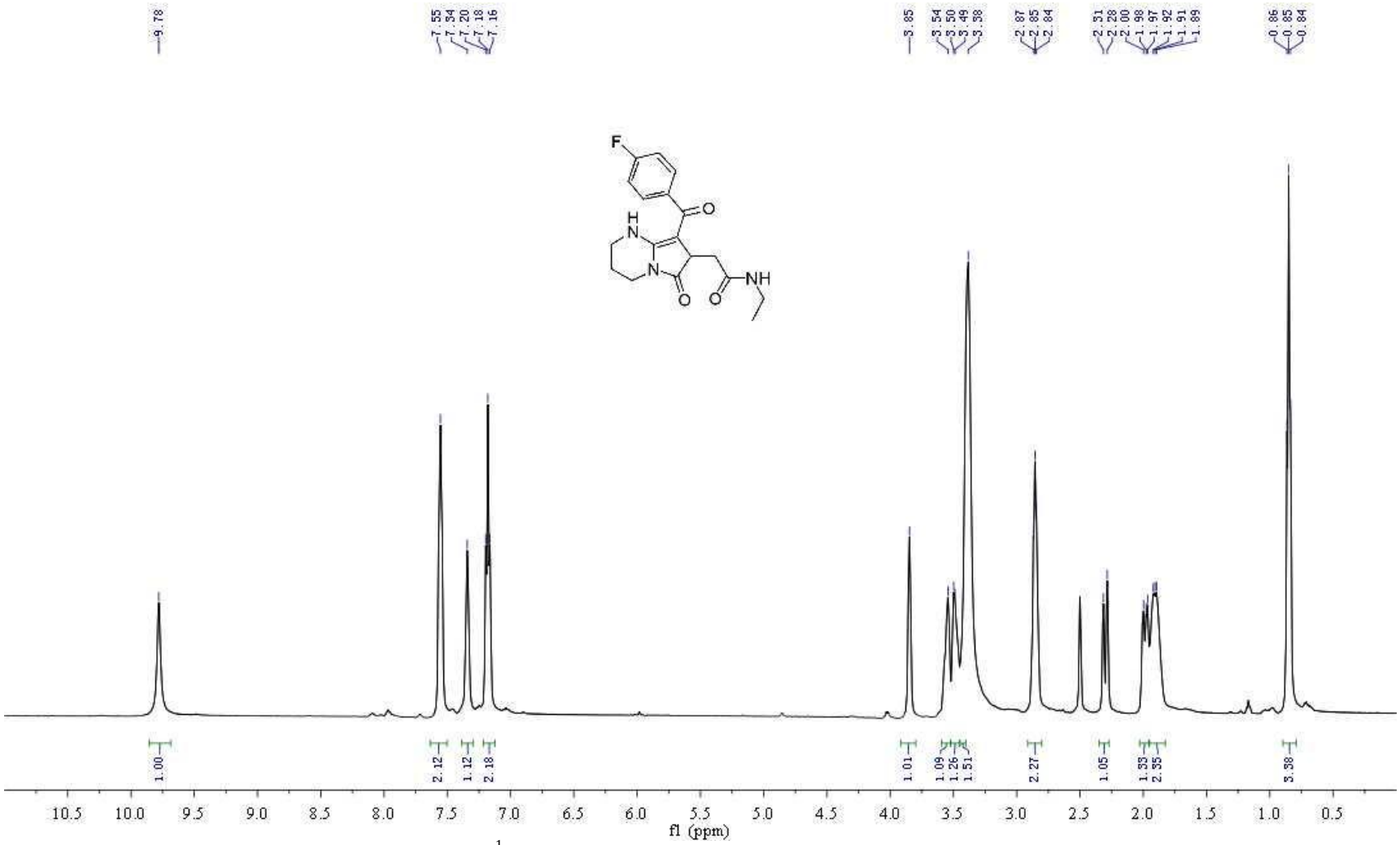


Figure 39. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3t**

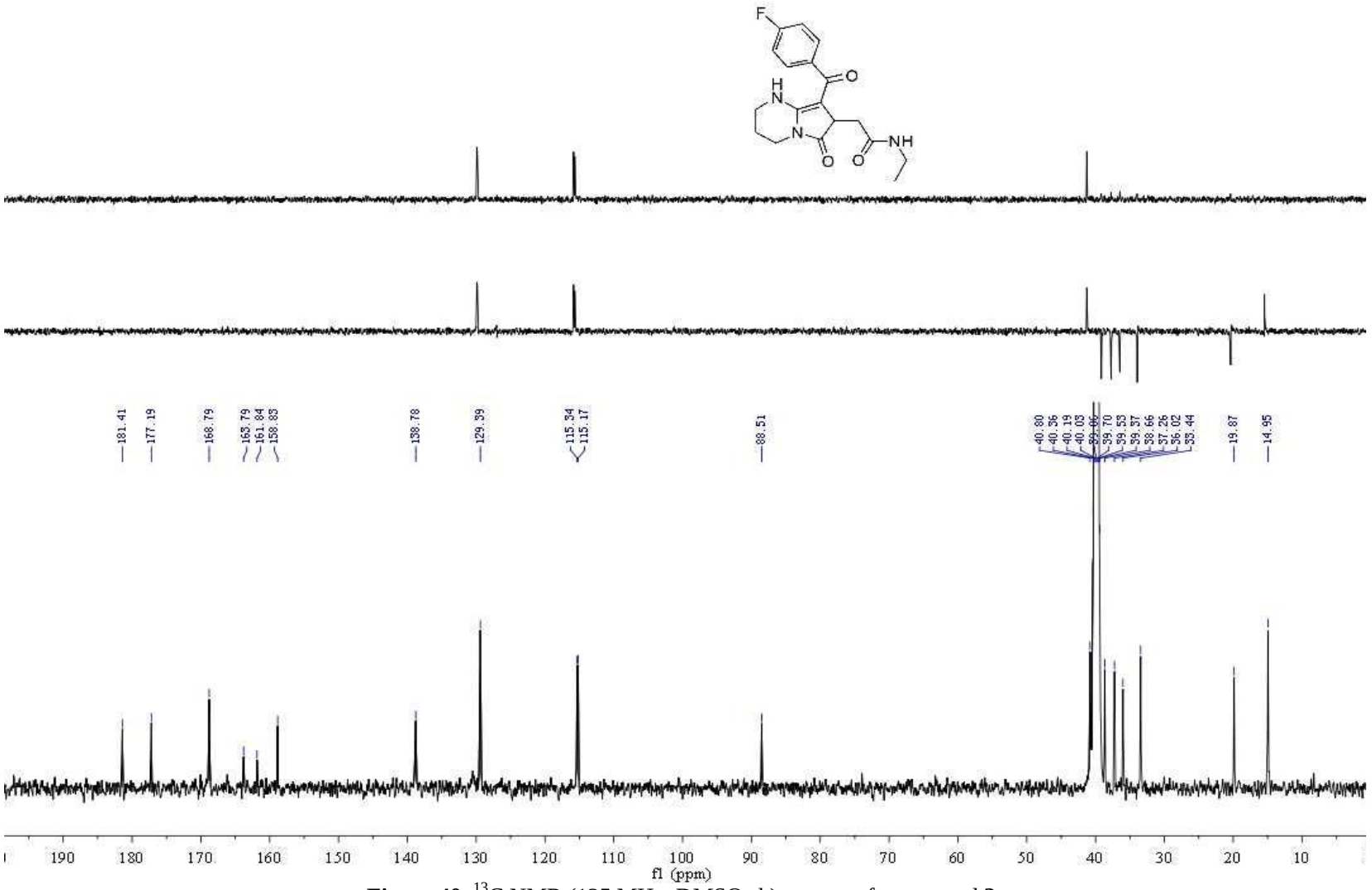


Figure 40. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3t**

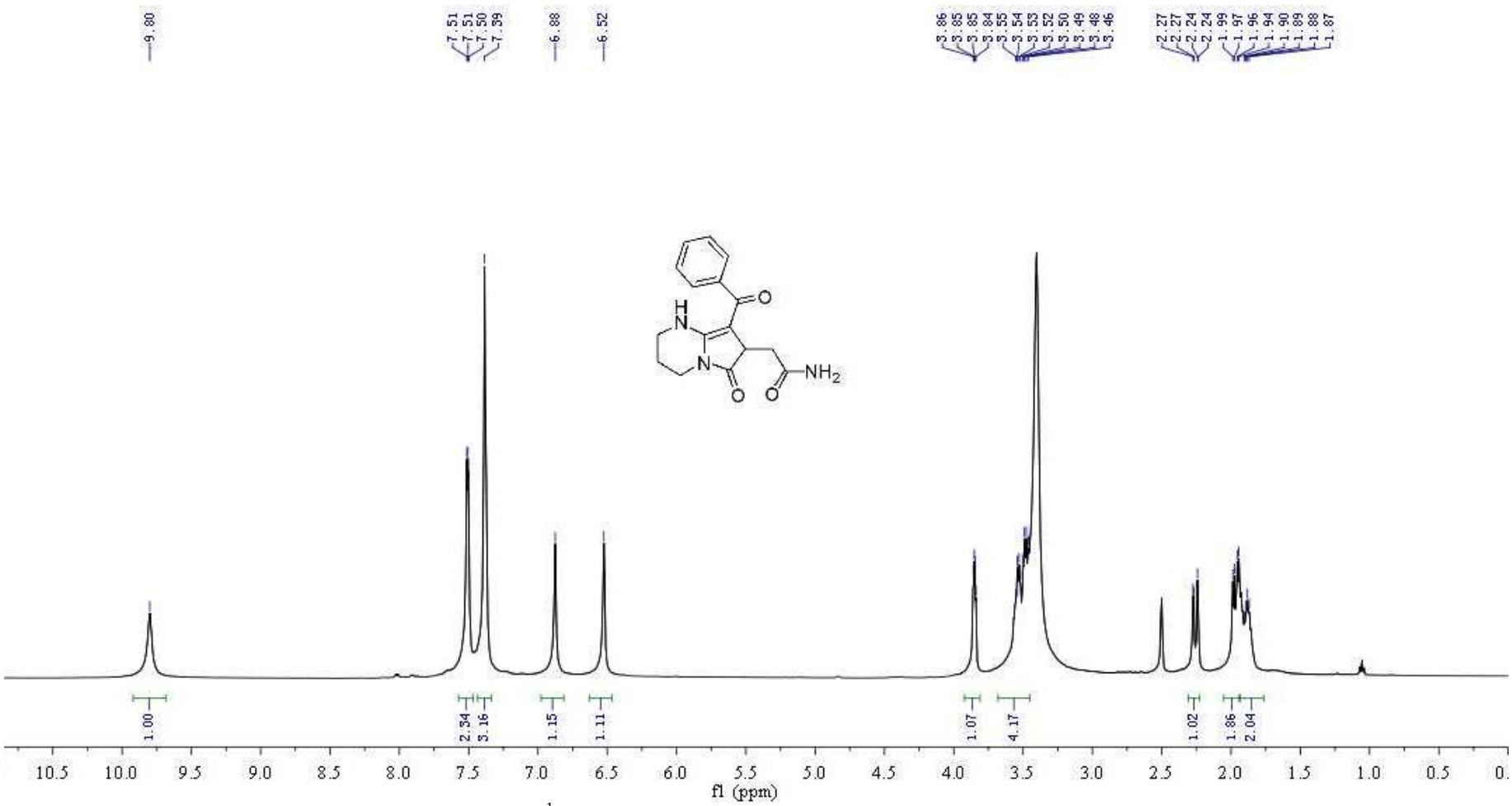


Figure 41. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3u**

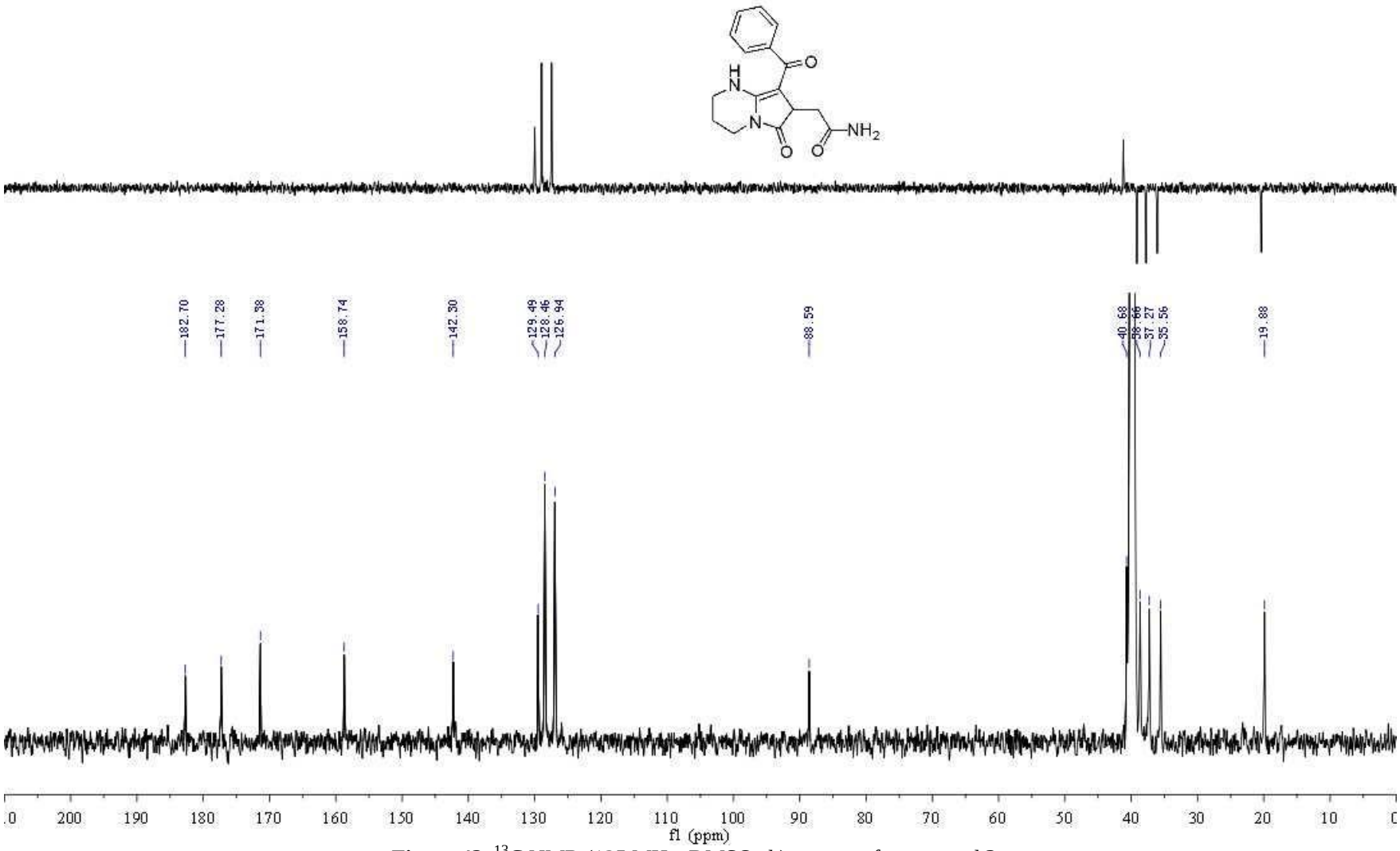


Figure 42. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3u**

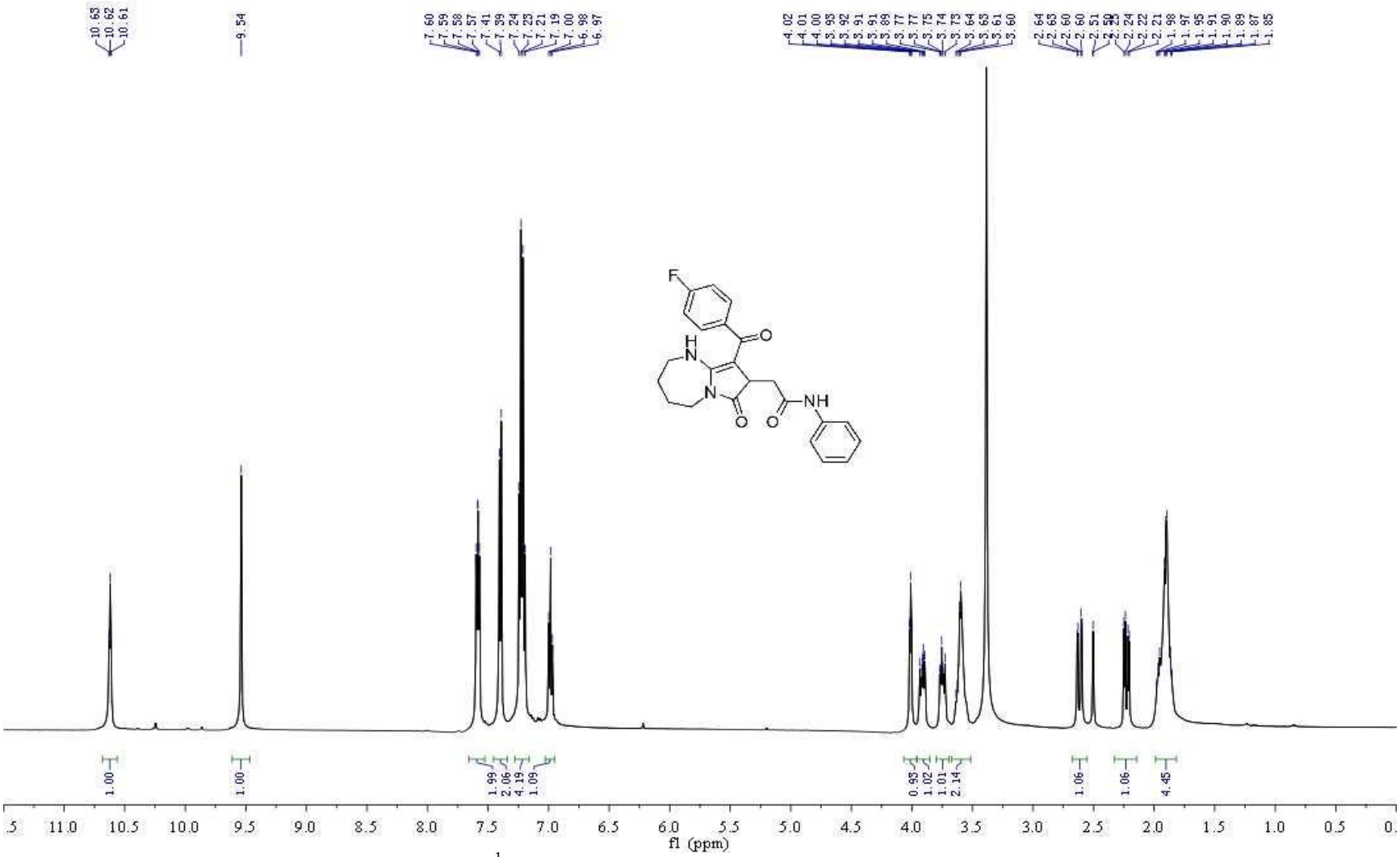


Figure 43. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3v**

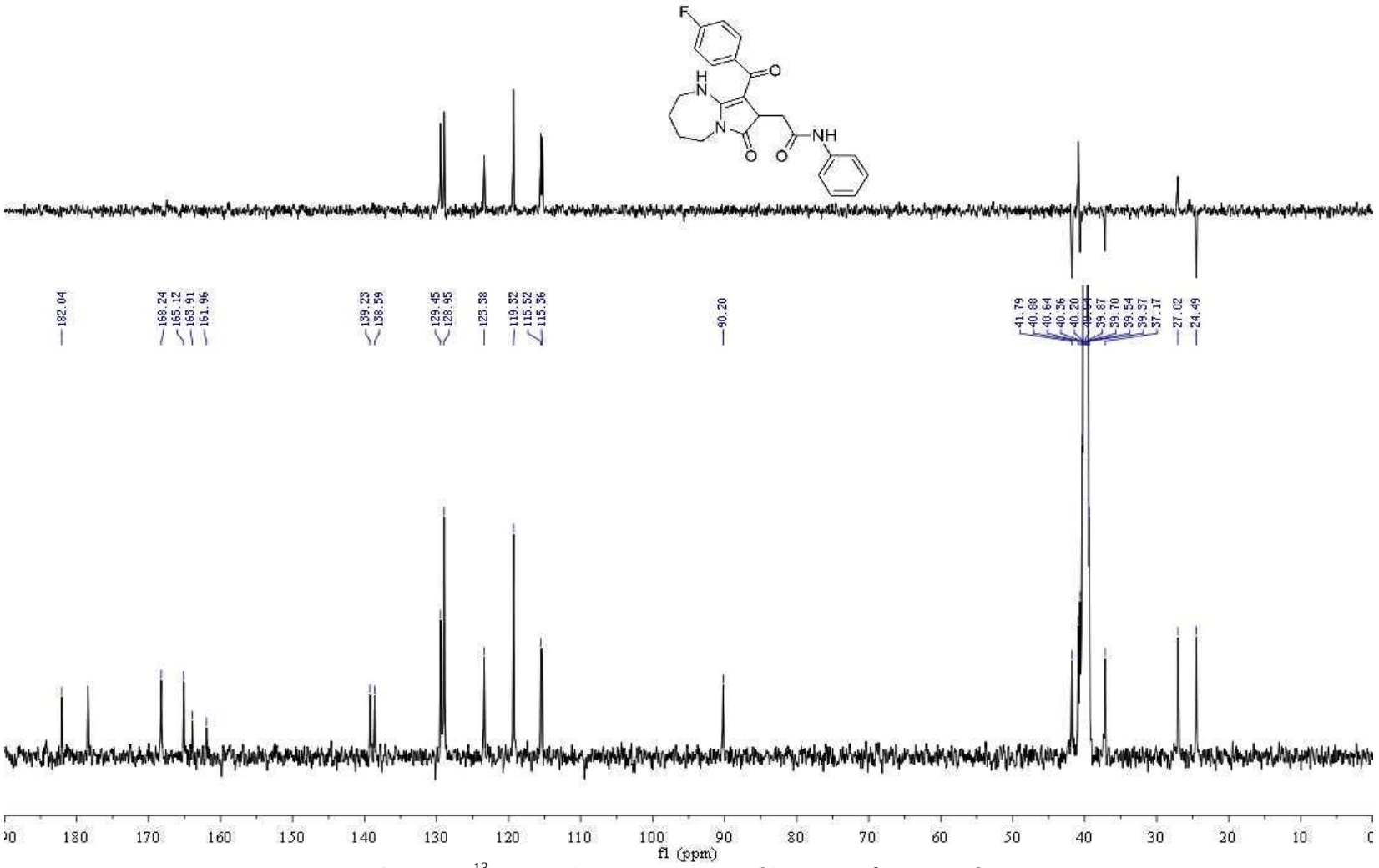


Figure 44. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3v**

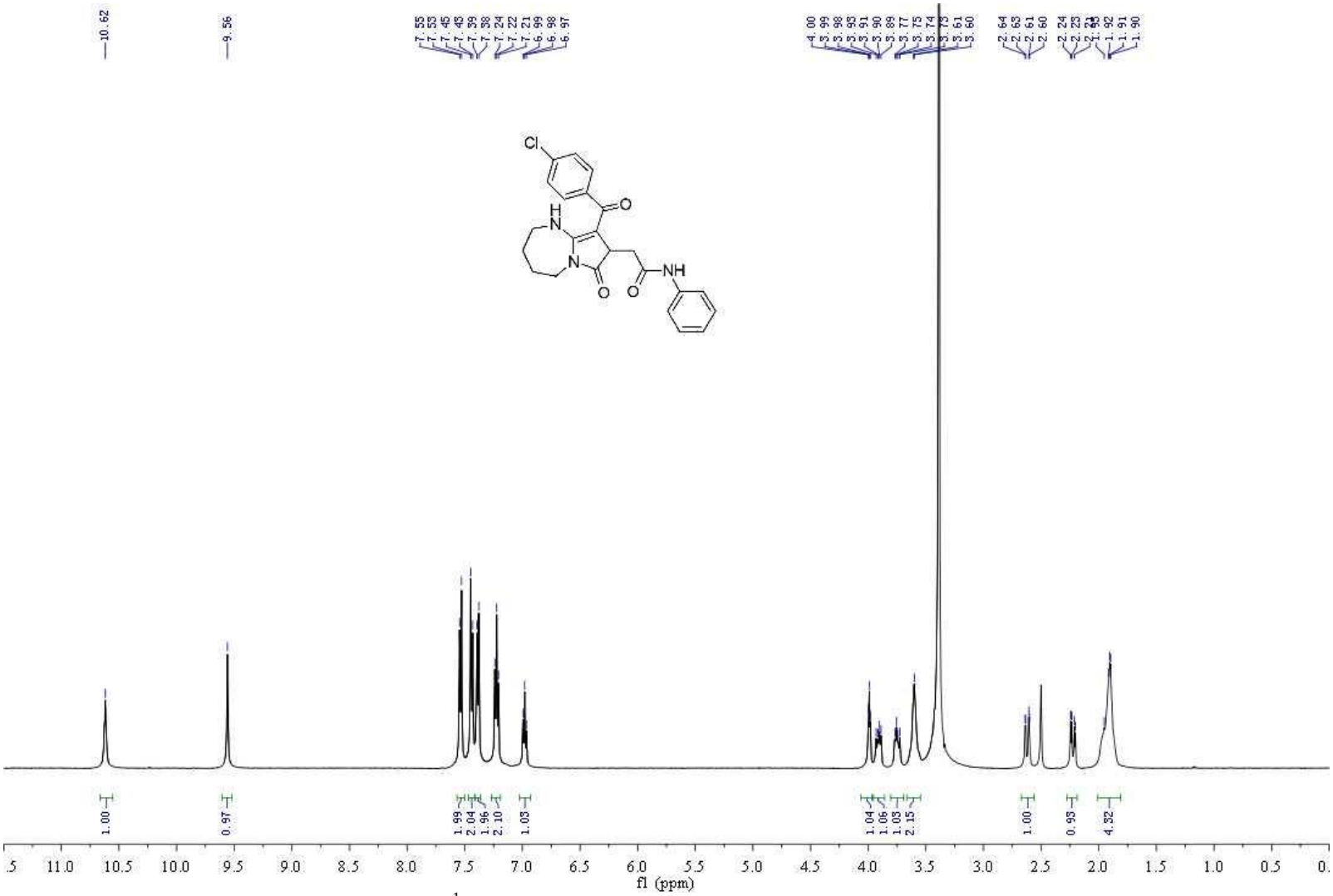


Figure 45. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3w**

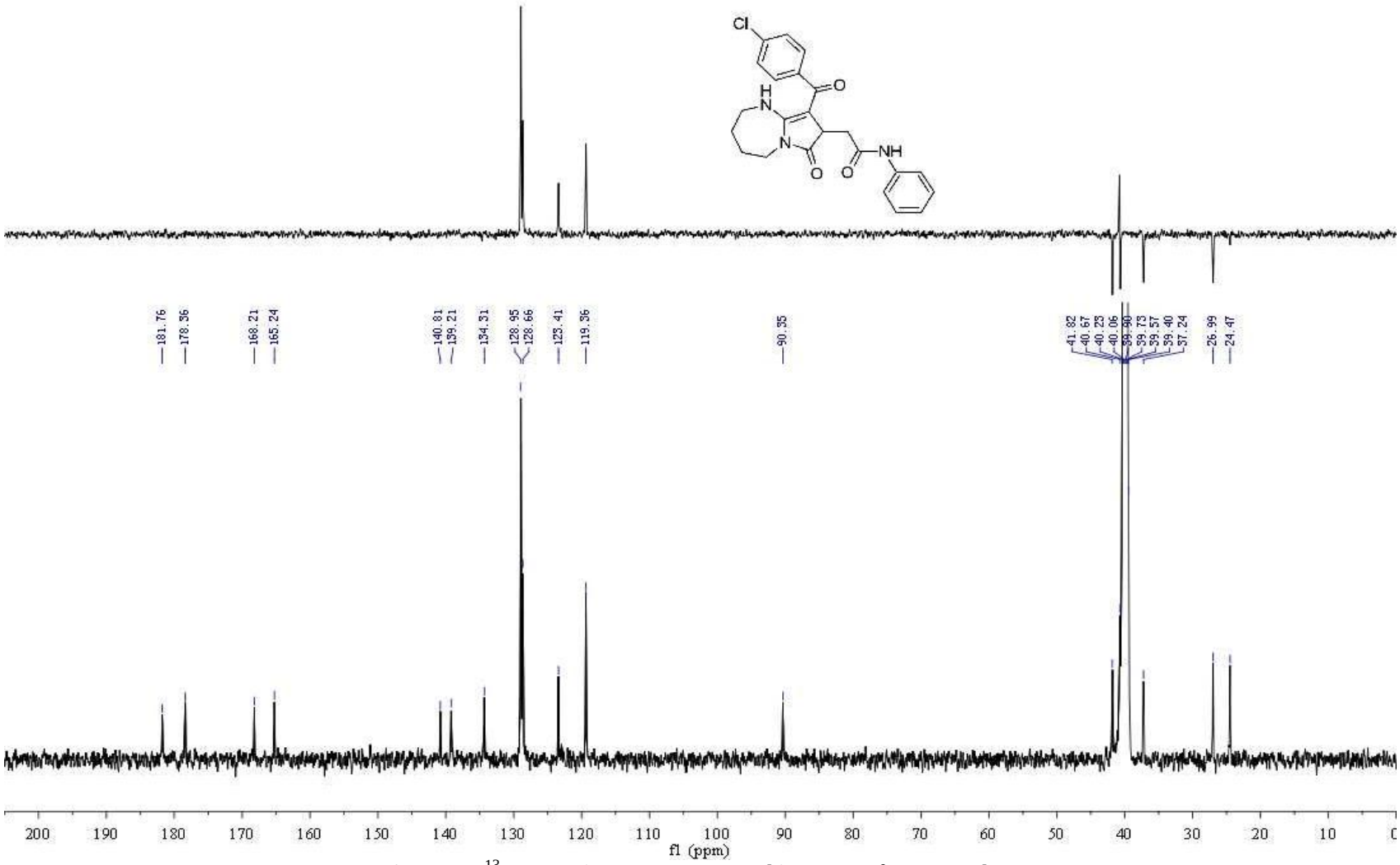


Figure 46. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3w**

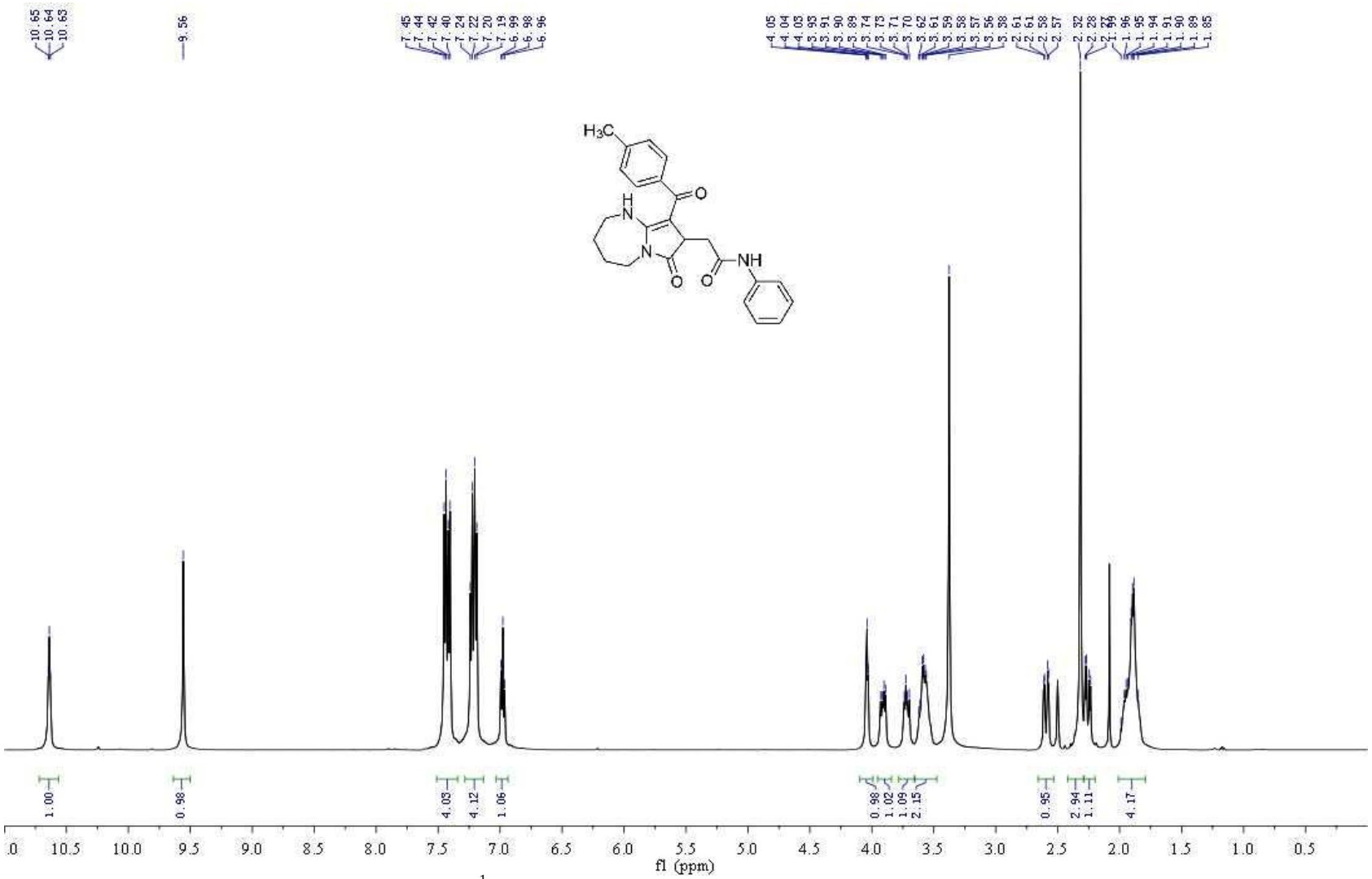


Figure 47. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3x**

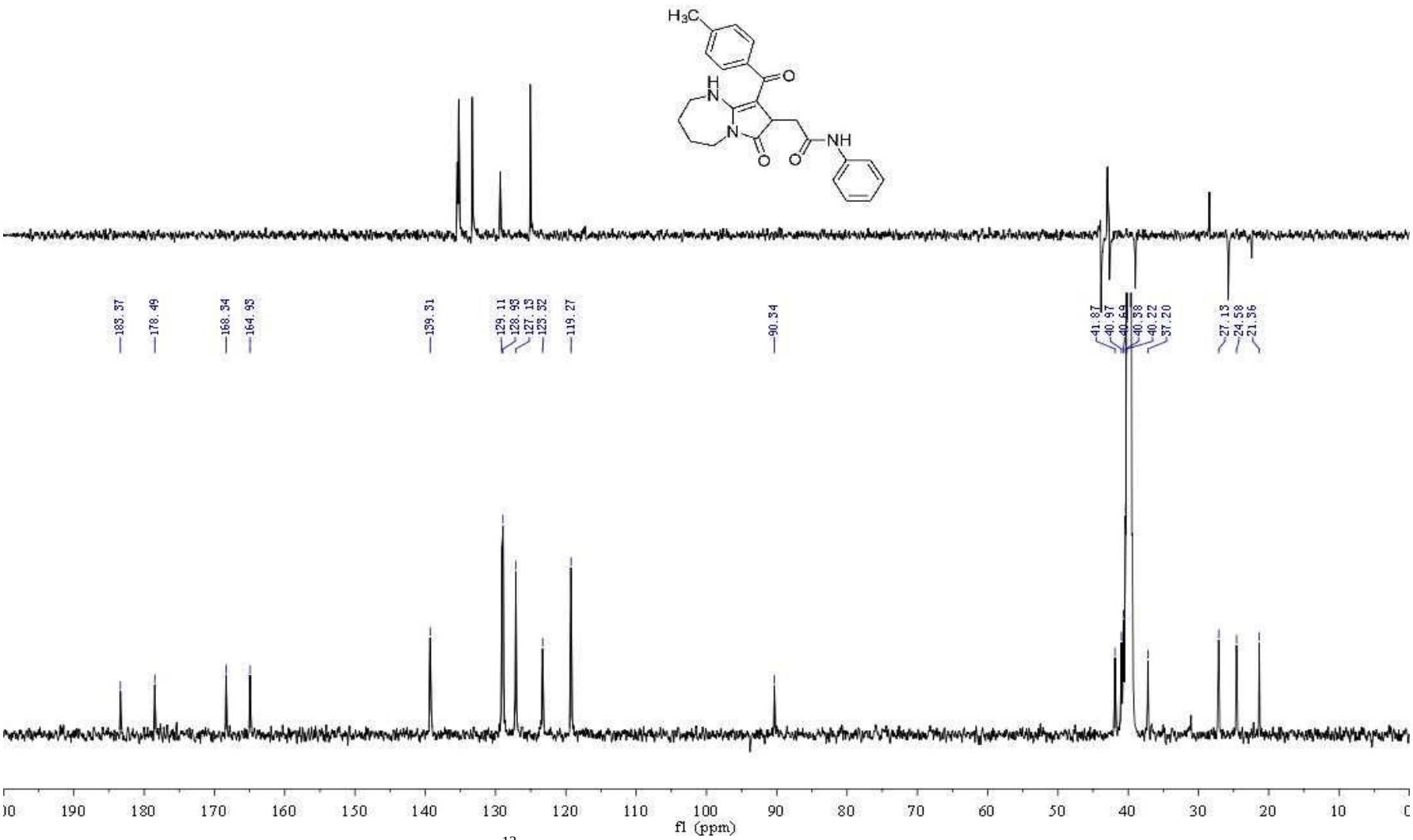


Figure 48. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3x**

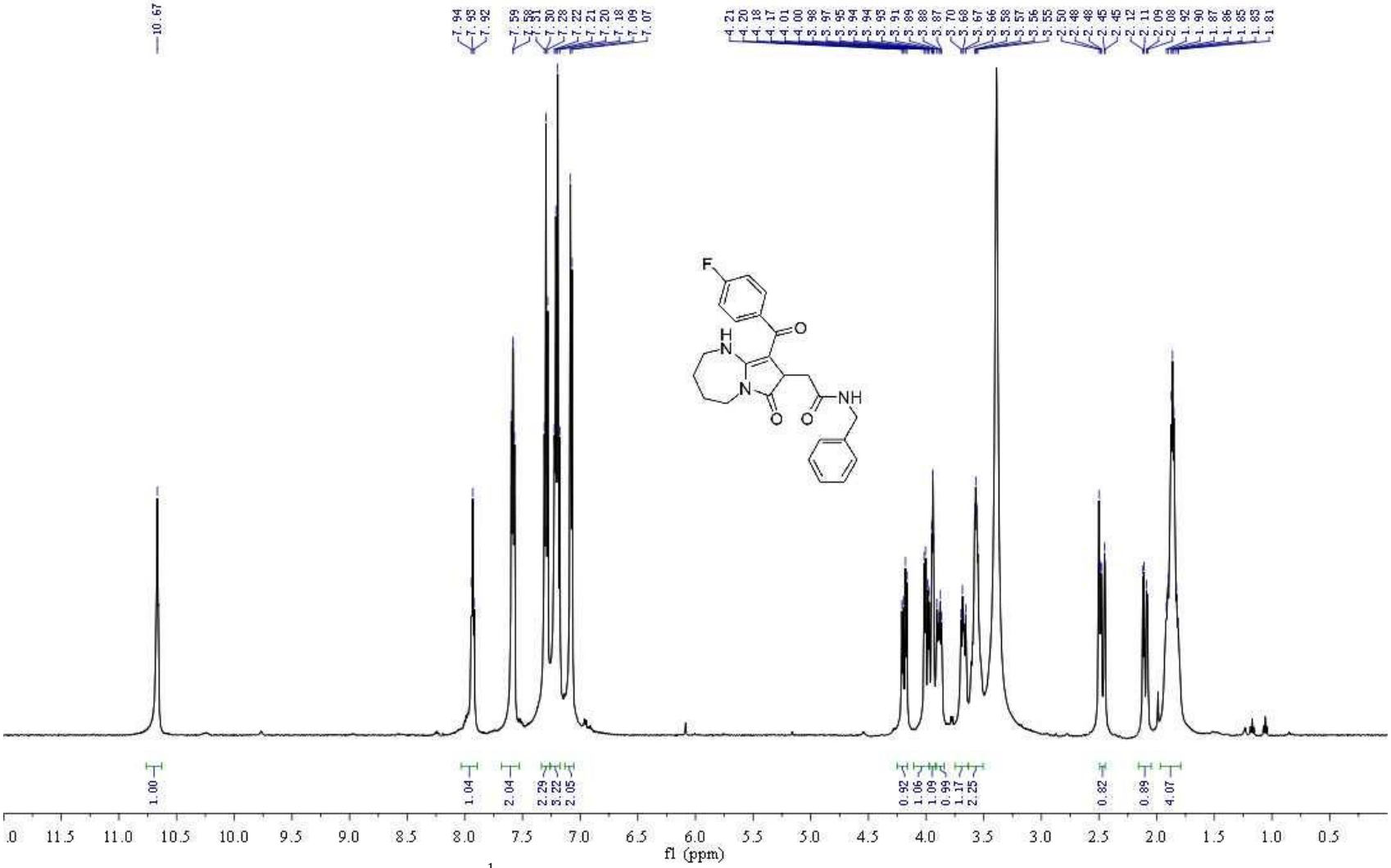


Figure 49. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3y**

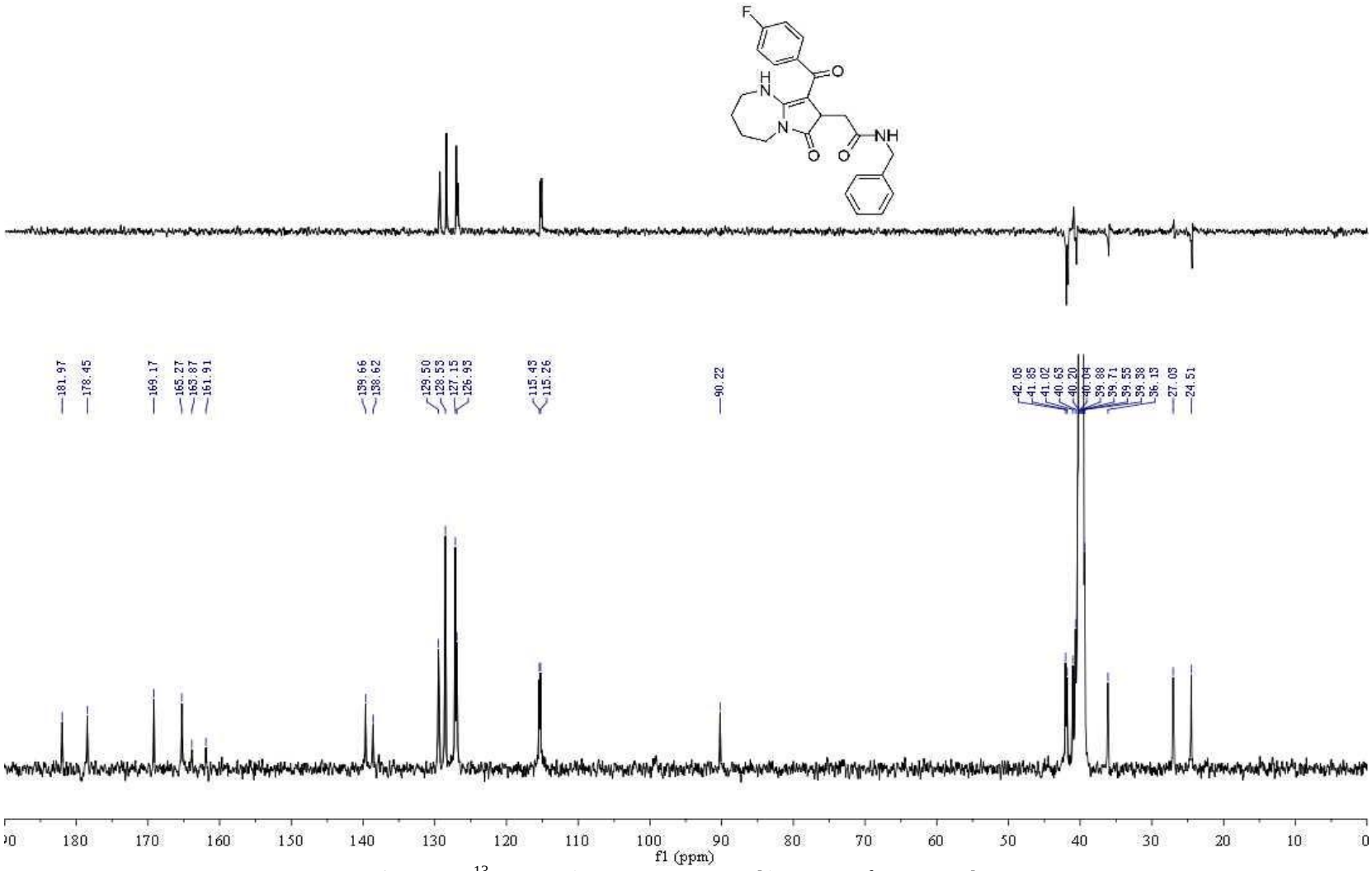


Figure 50. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3y**

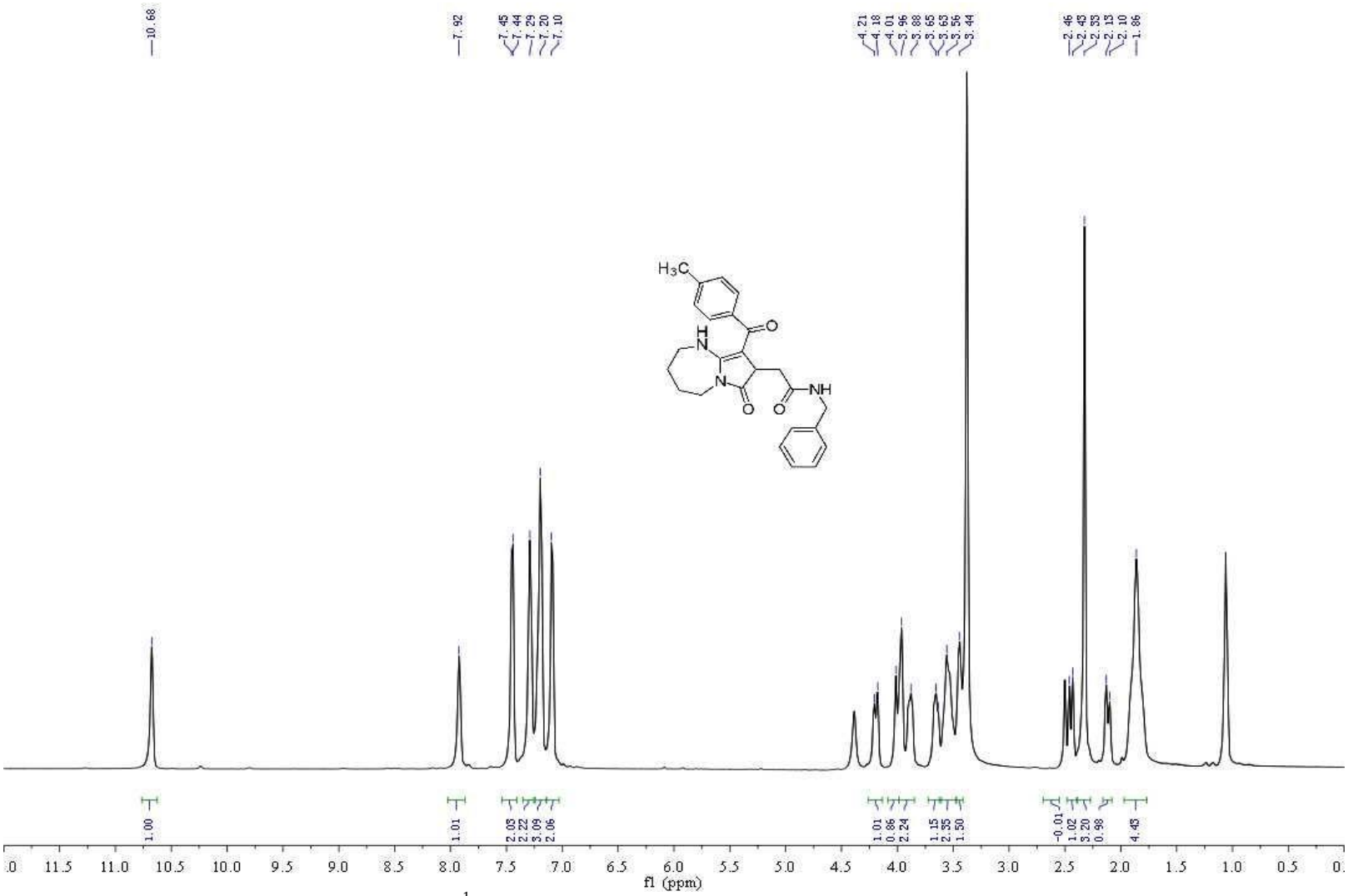


Figure 51. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3z**

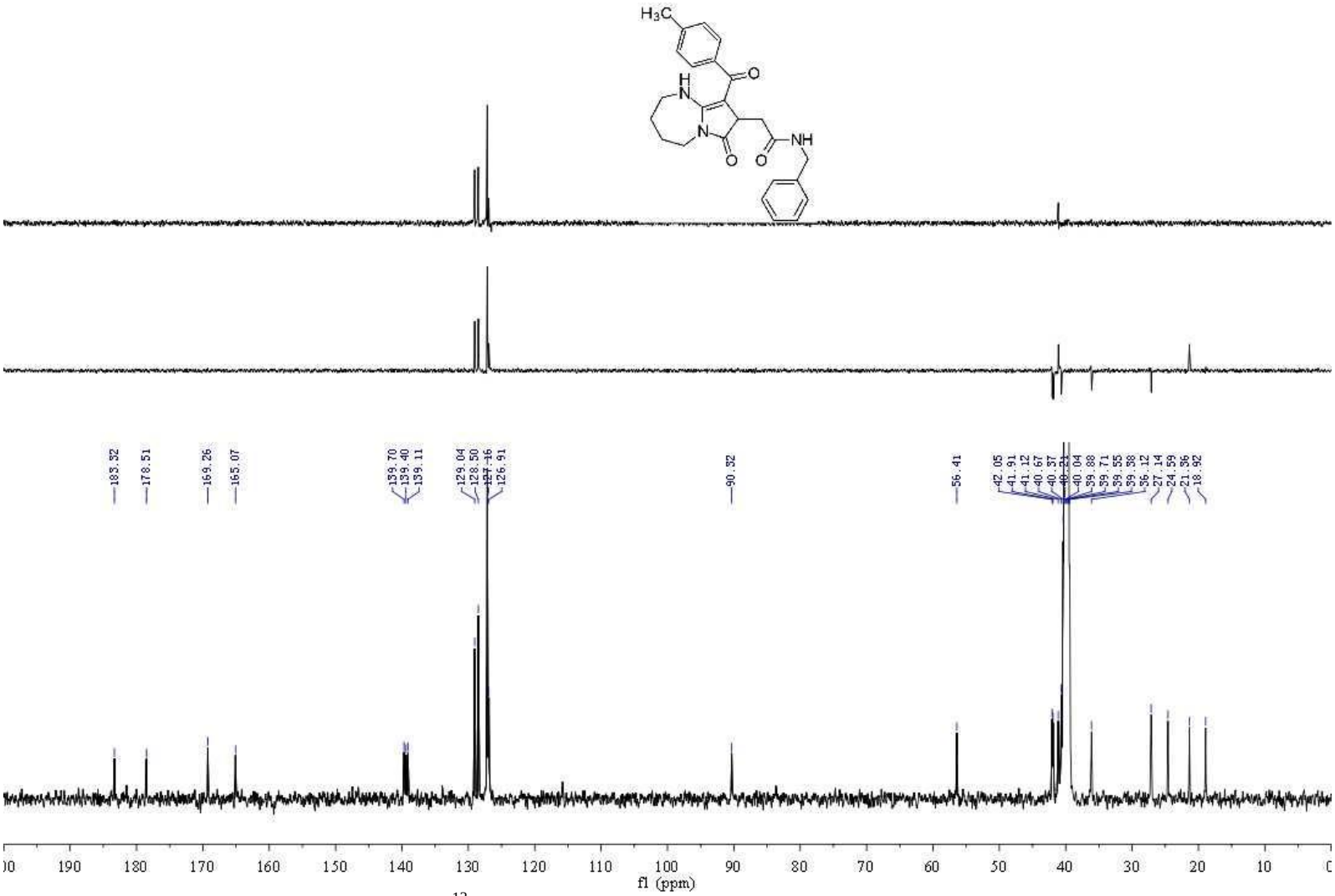


Figure 52. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3z**

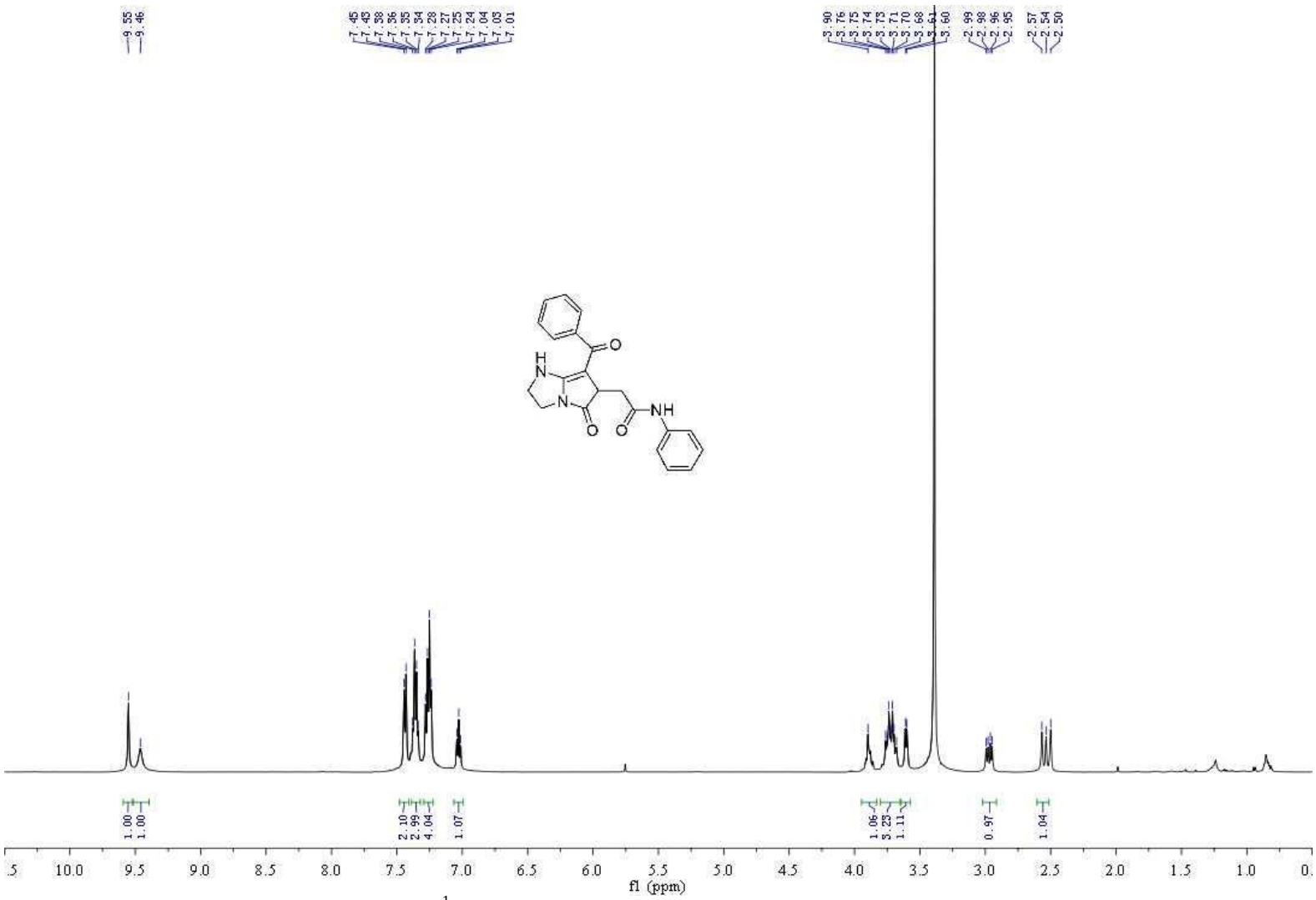


Figure 53. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3a´**

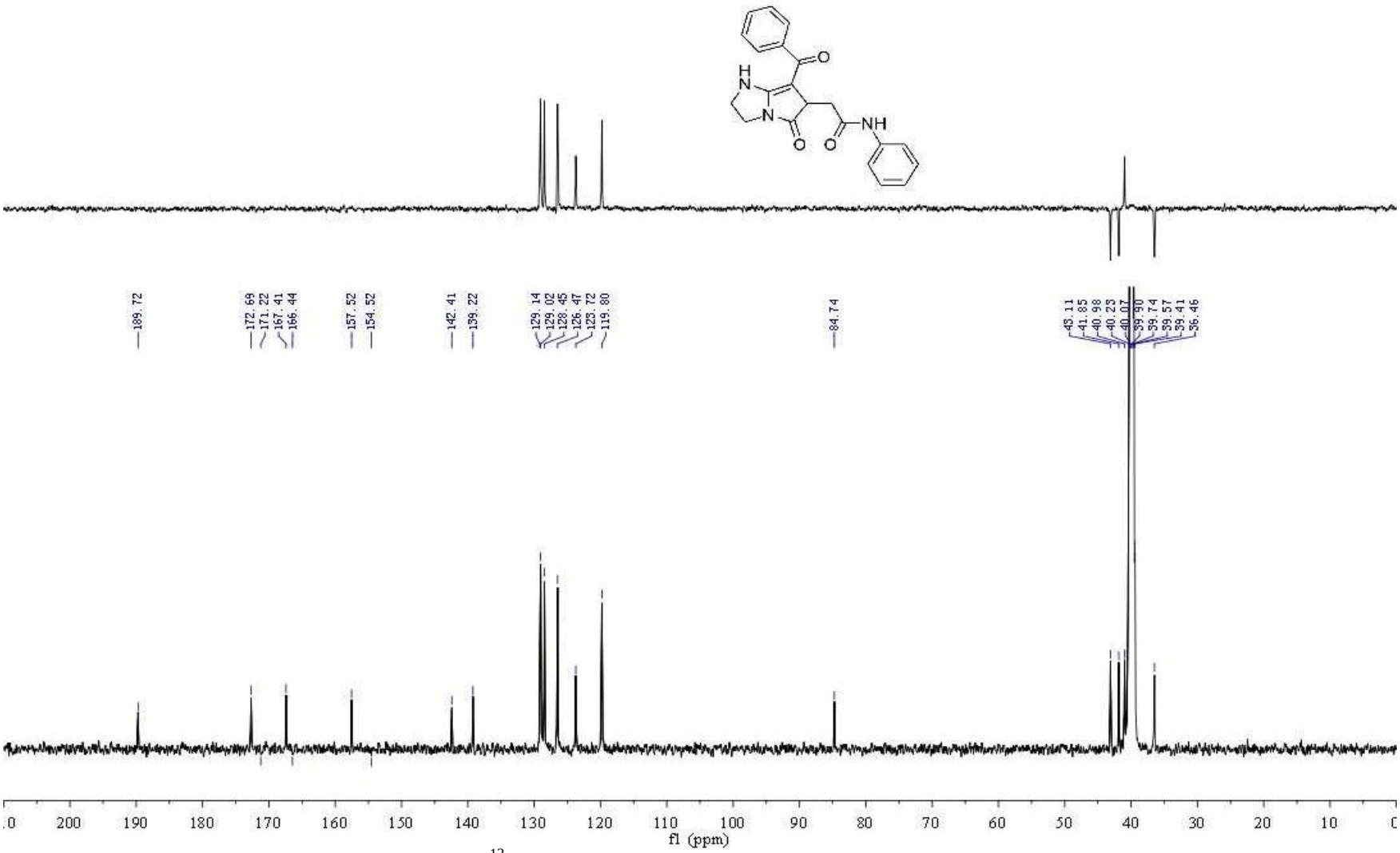


Figure 54. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3a´**

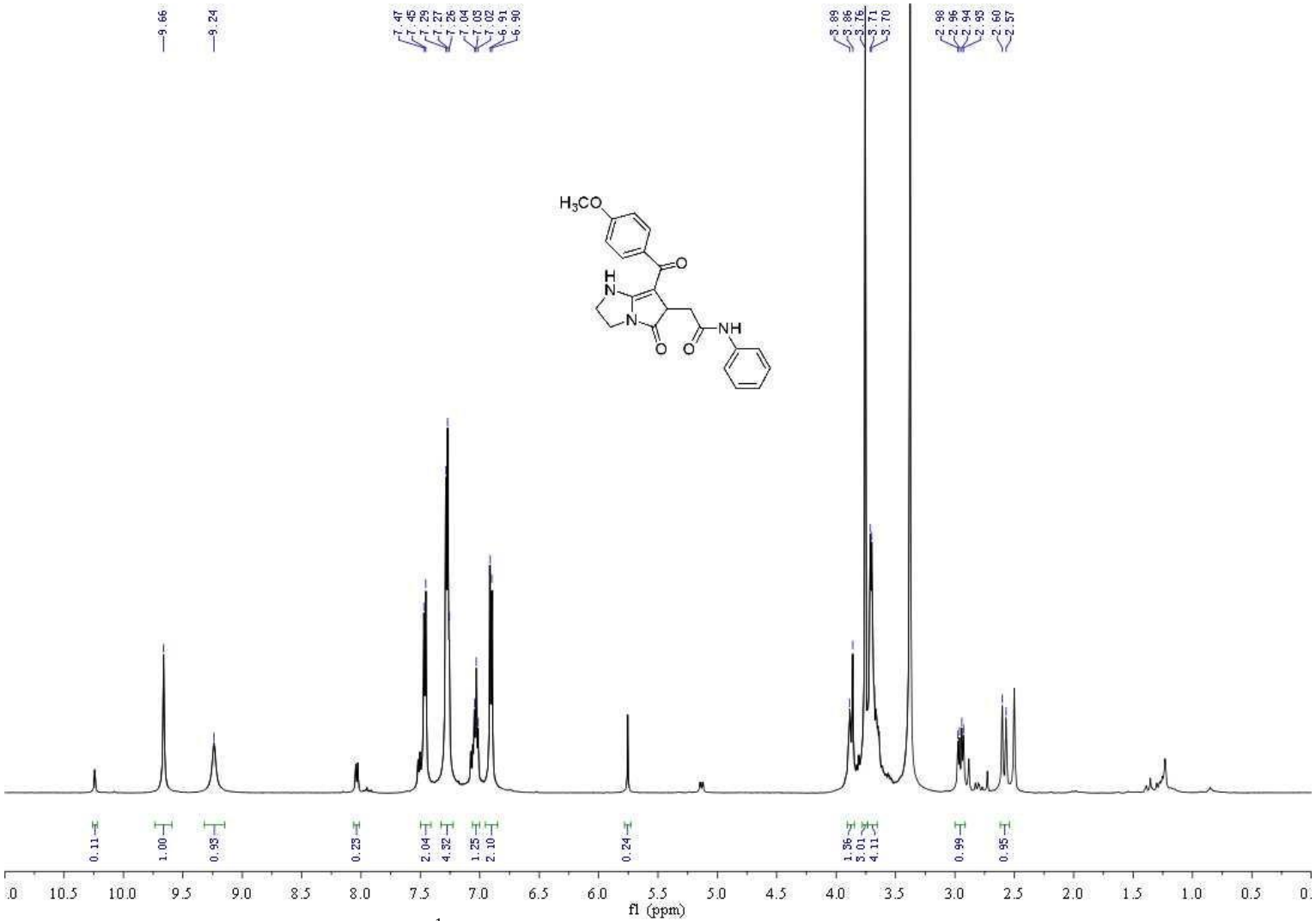


Figure 55. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3b´**

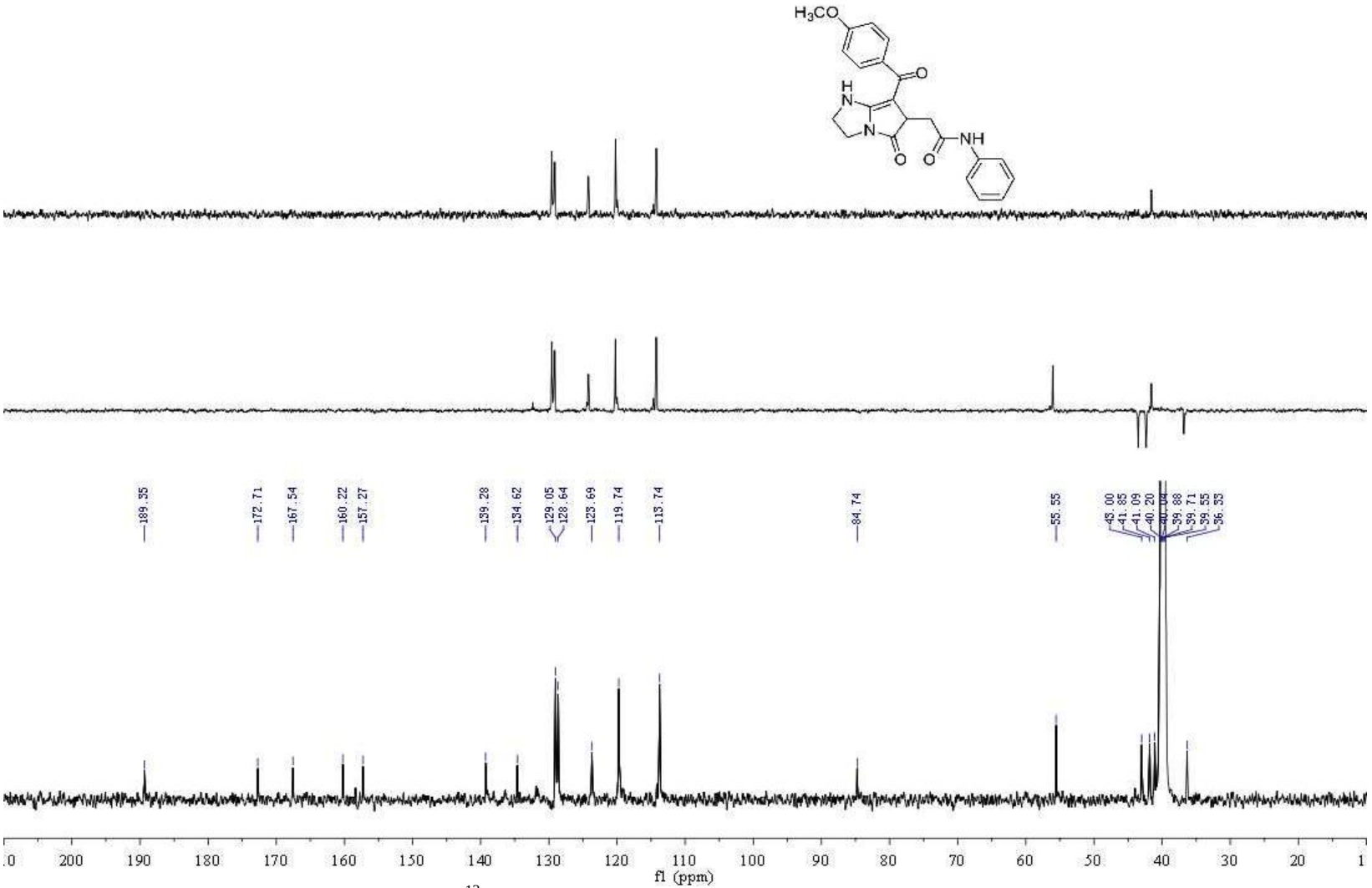


Figure 56. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3b´**

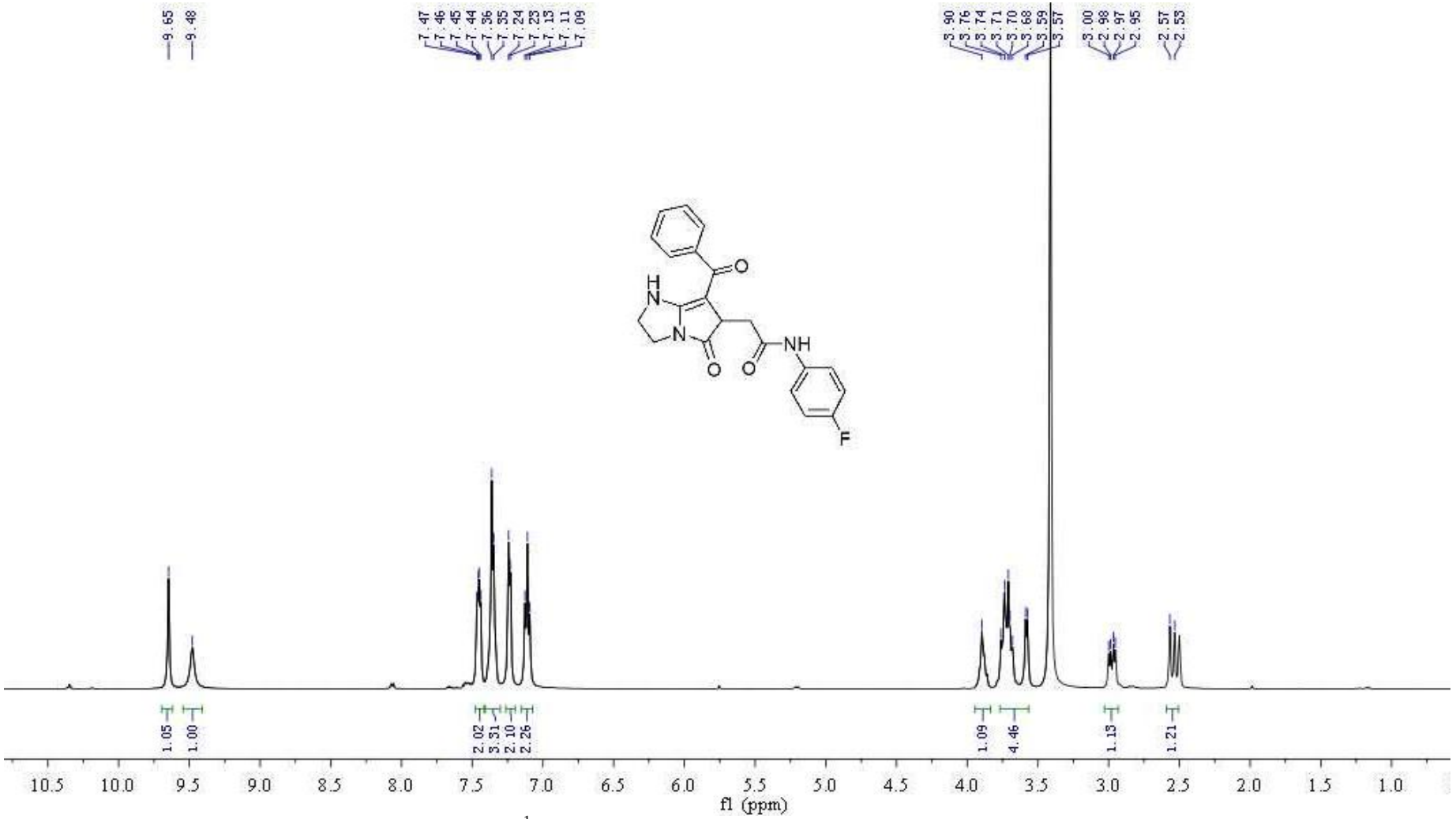


Figure 57. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3c´**

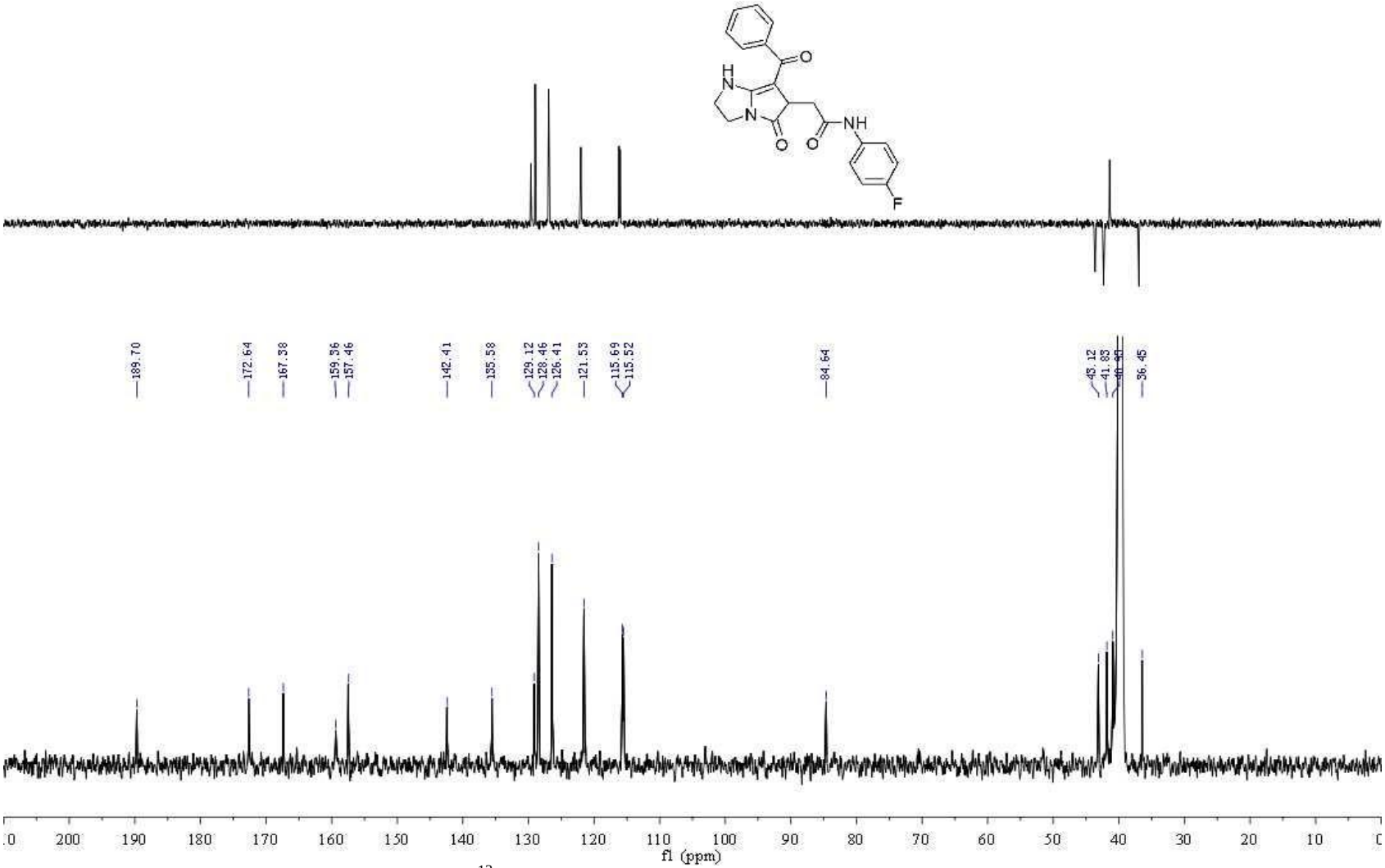


Figure 58. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3c´**

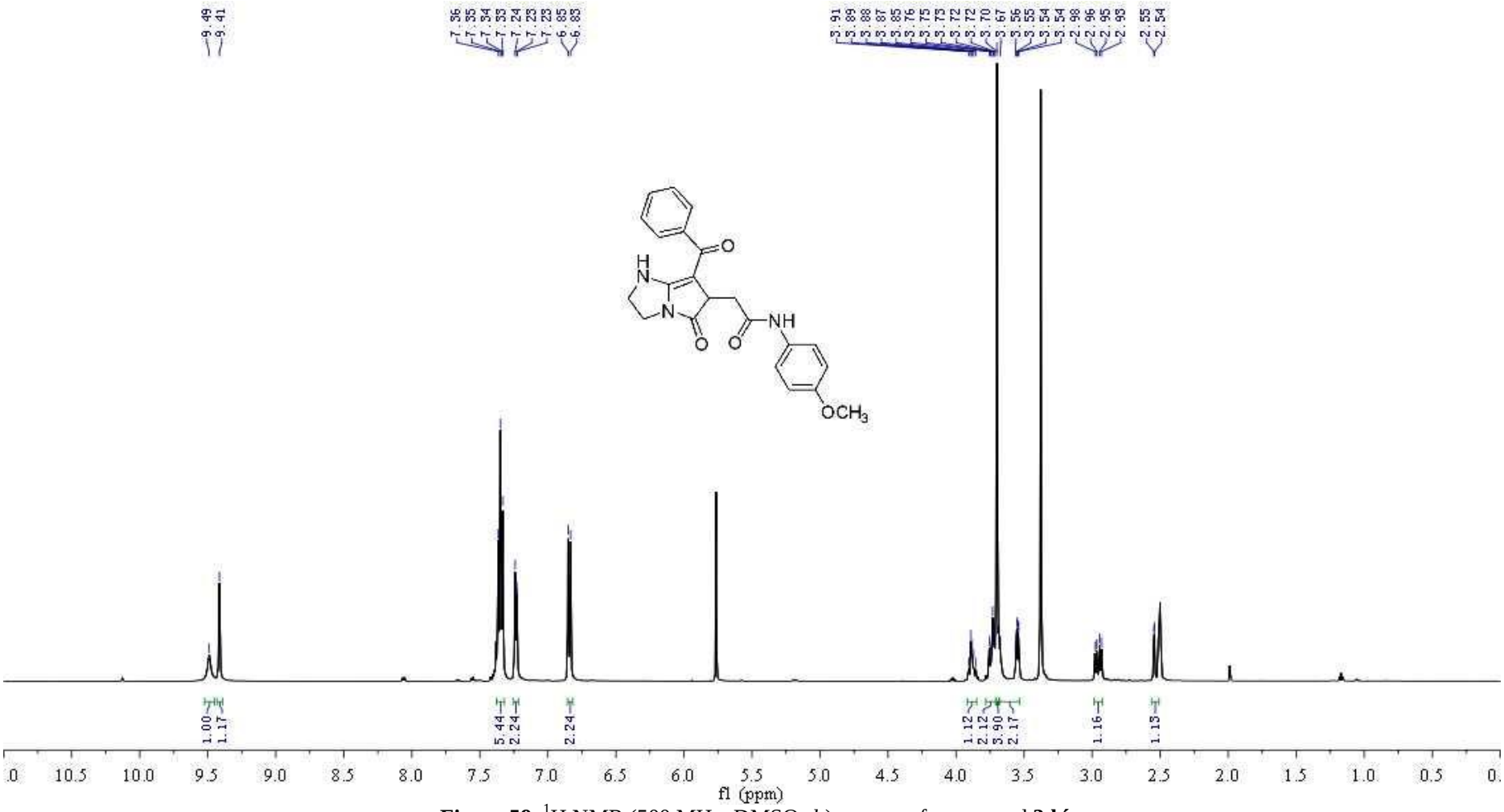


Figure 59. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3d´**

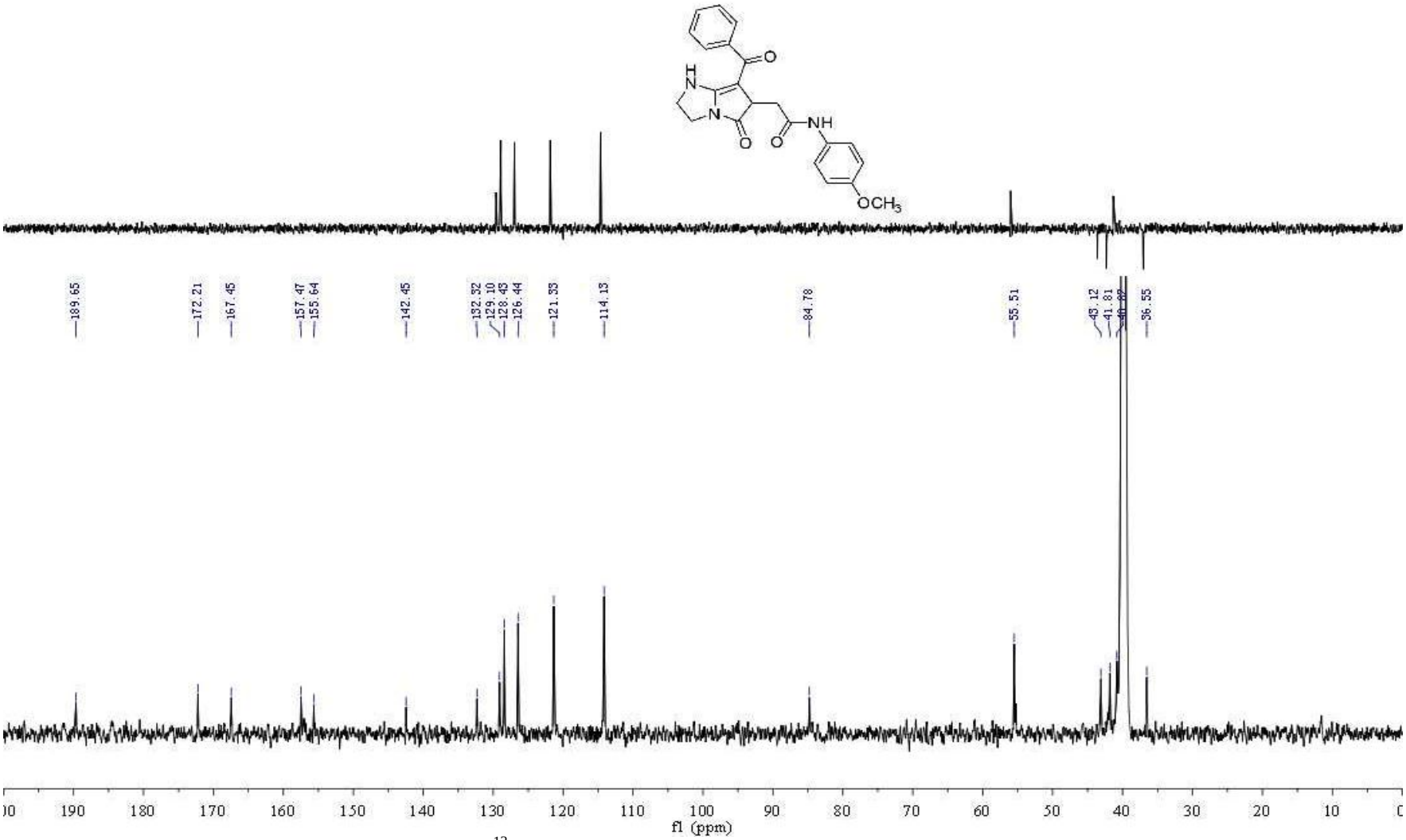


Figure 60. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3d´**

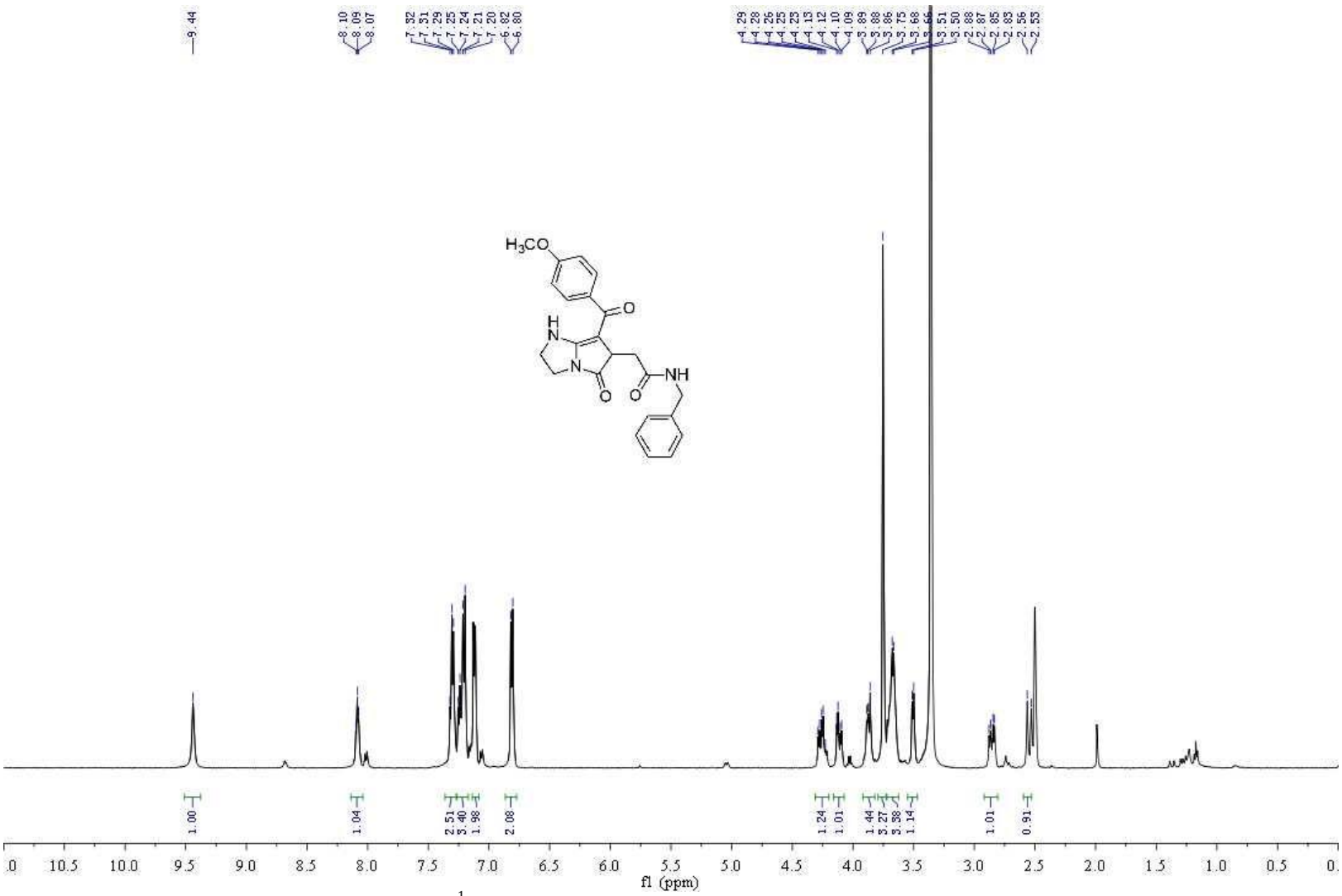


Figure 61. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3e´**

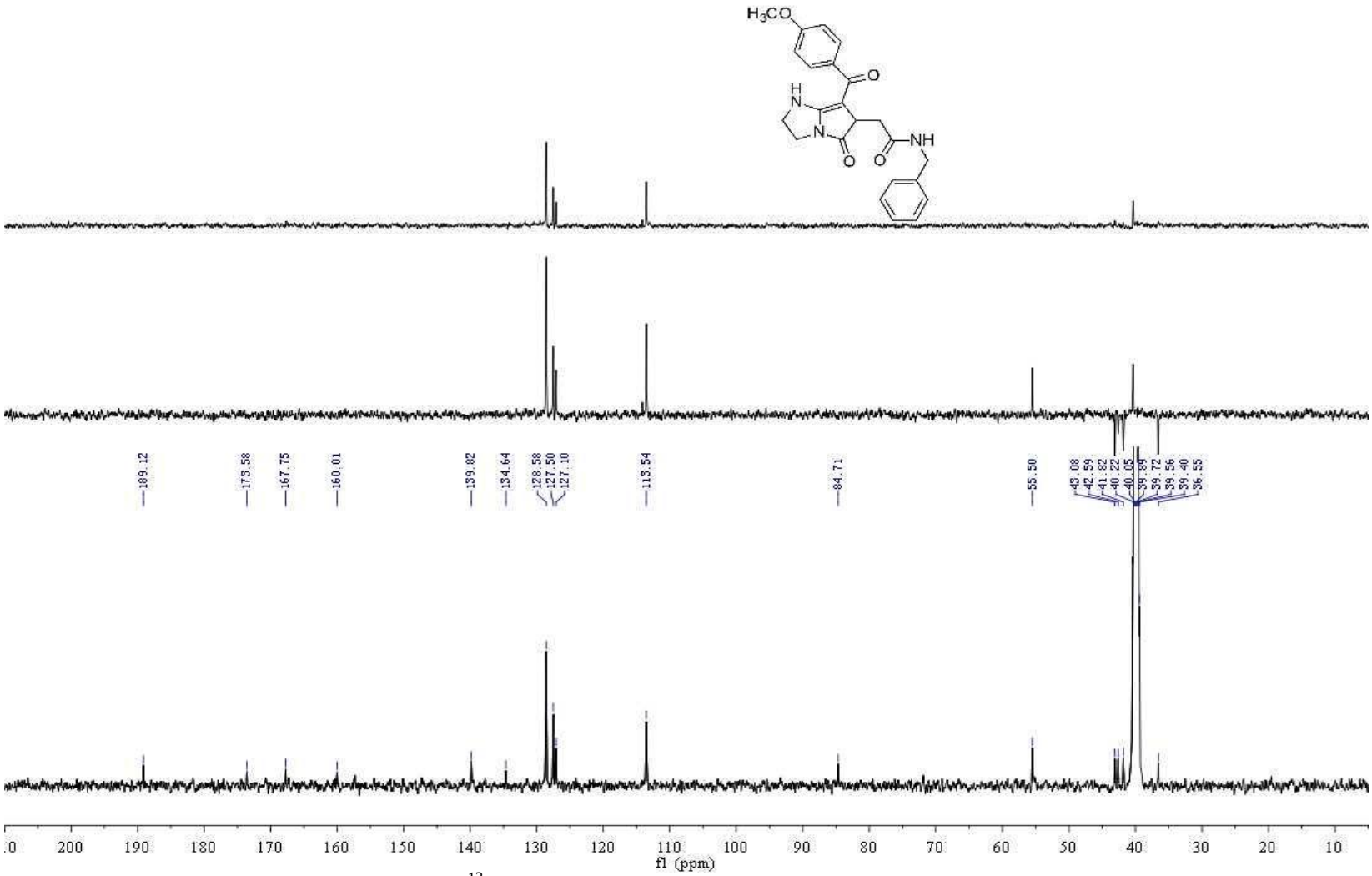


Figure 62. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3e´**

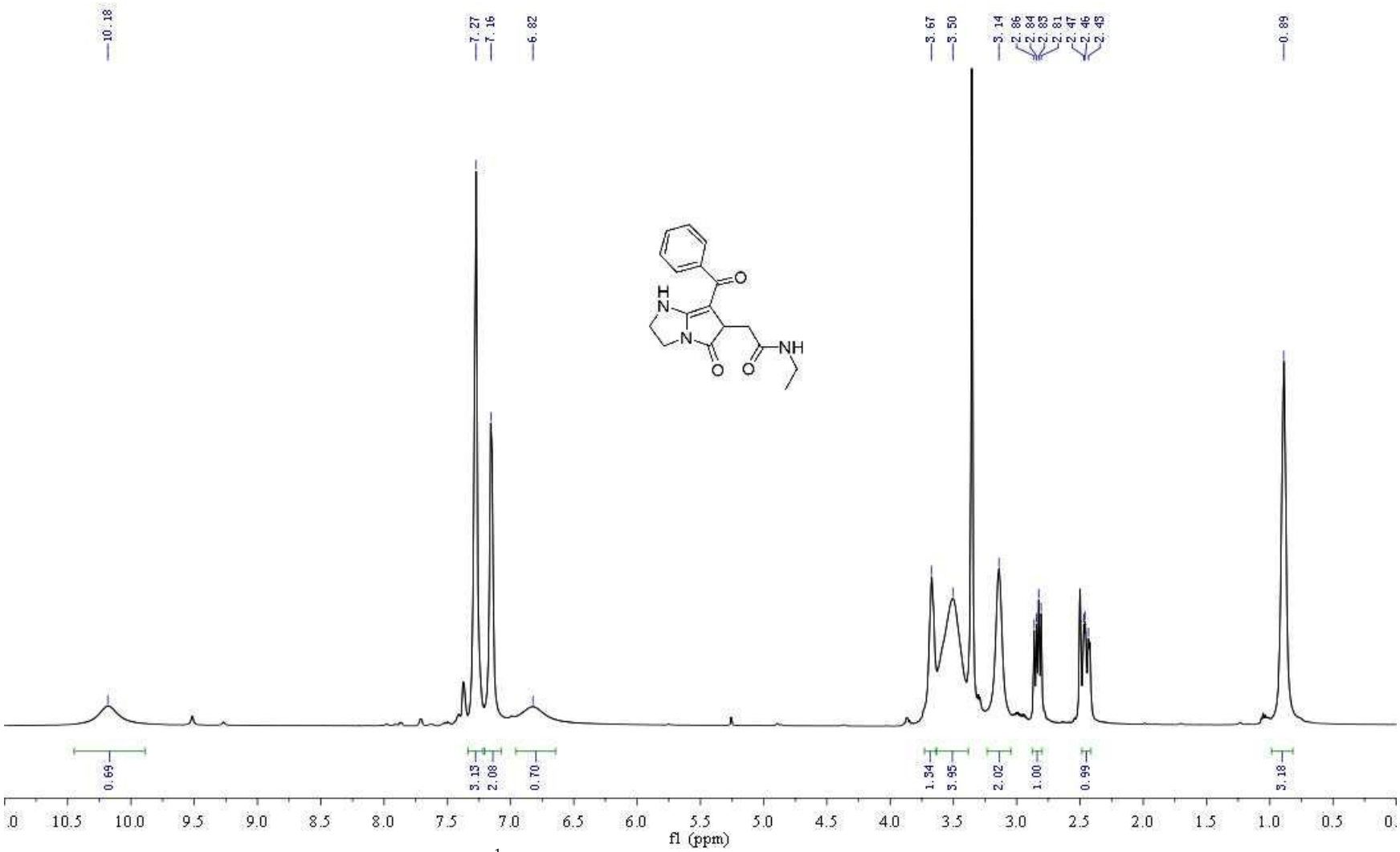


Figure 63. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3f´**

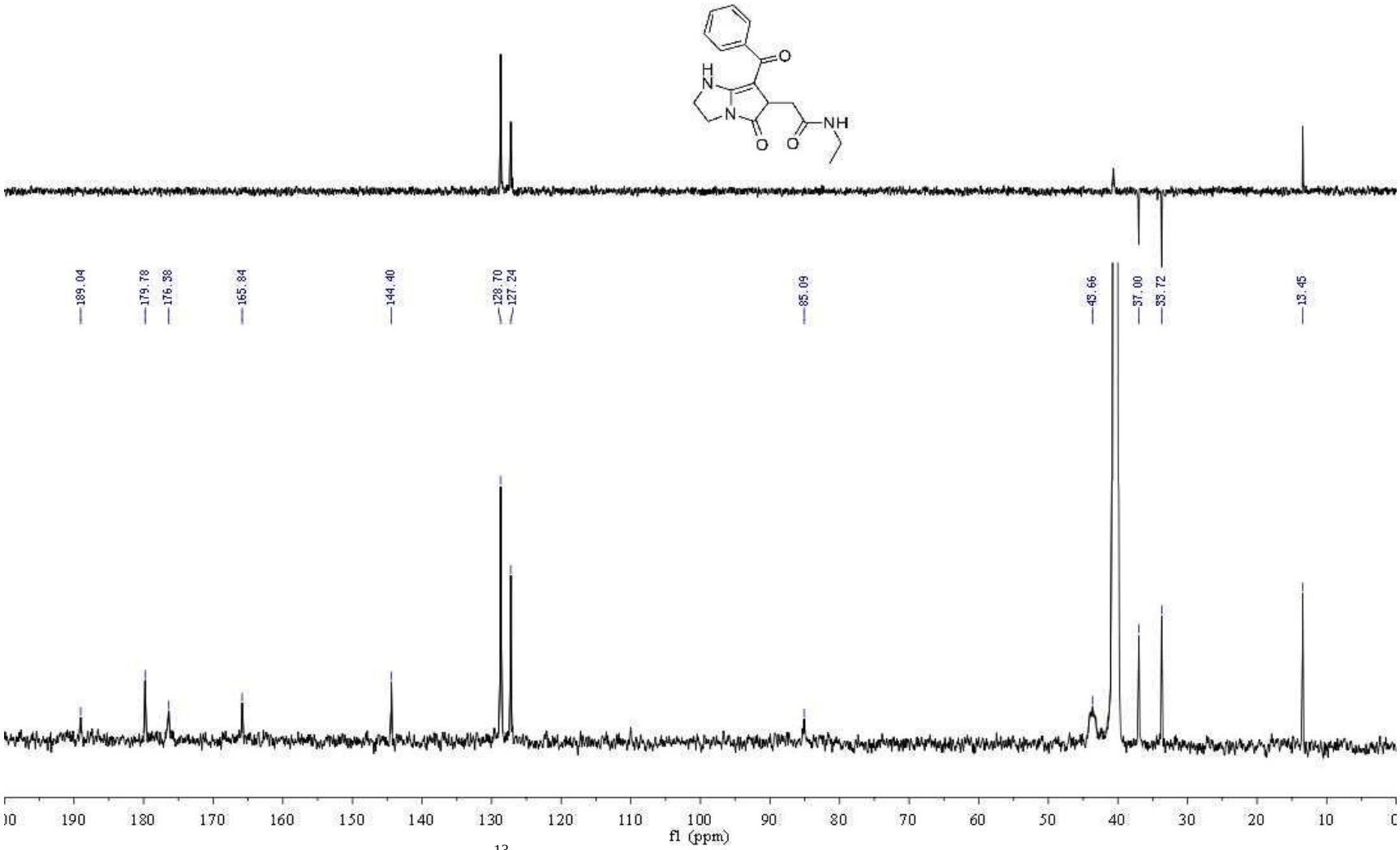


Figure 64. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3f´**

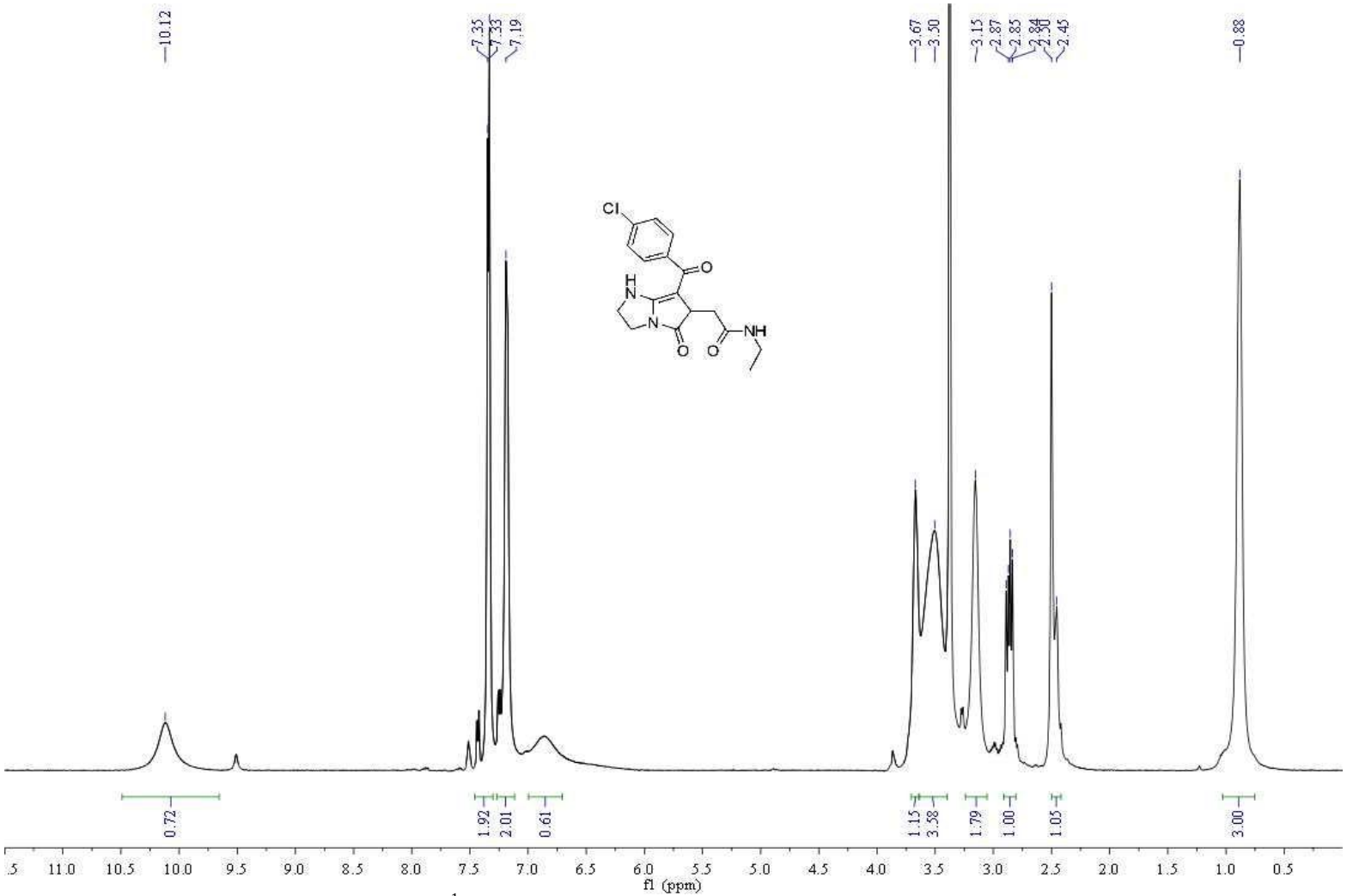


Figure 65. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3g´**

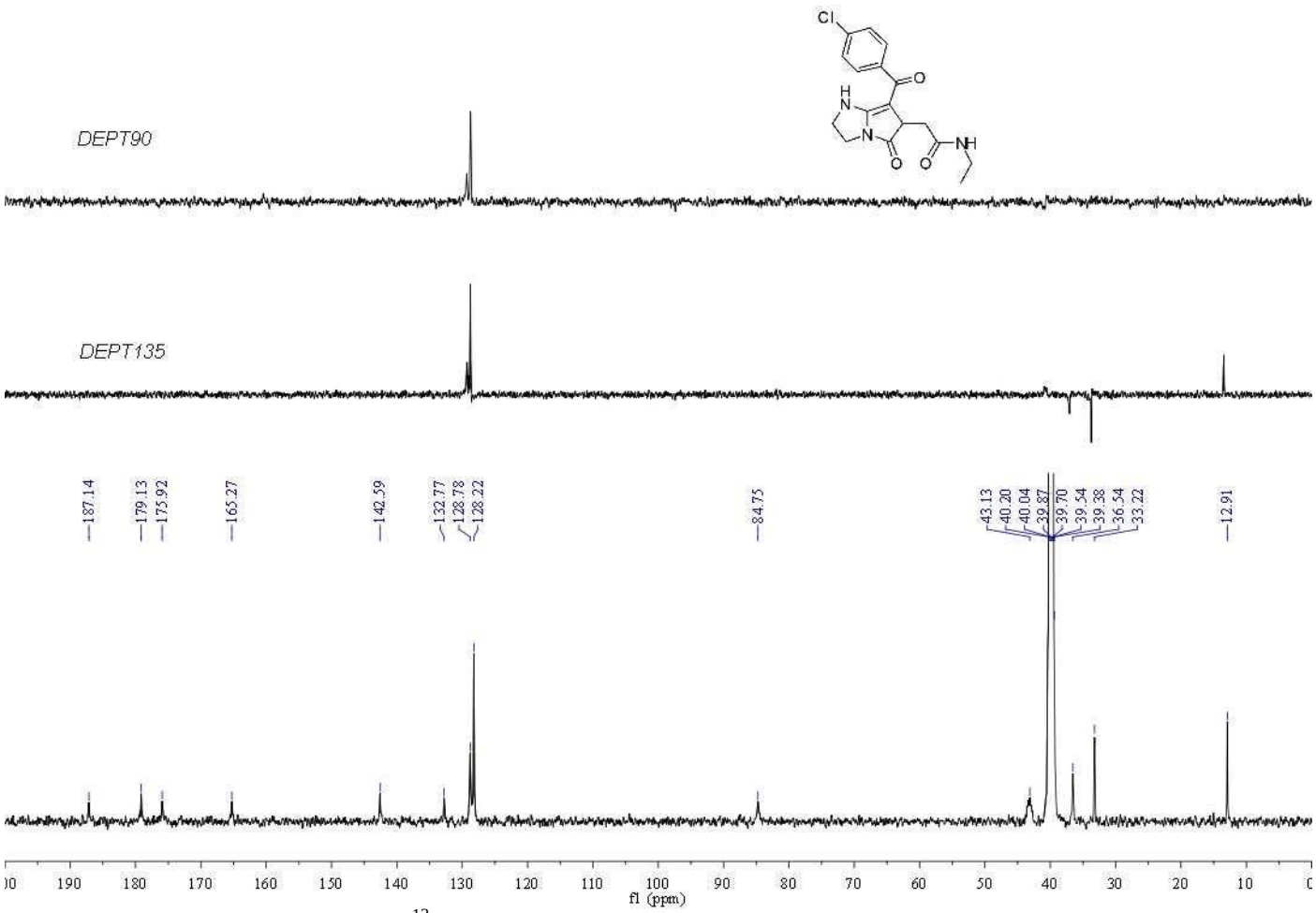


Figure 66. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3g´**

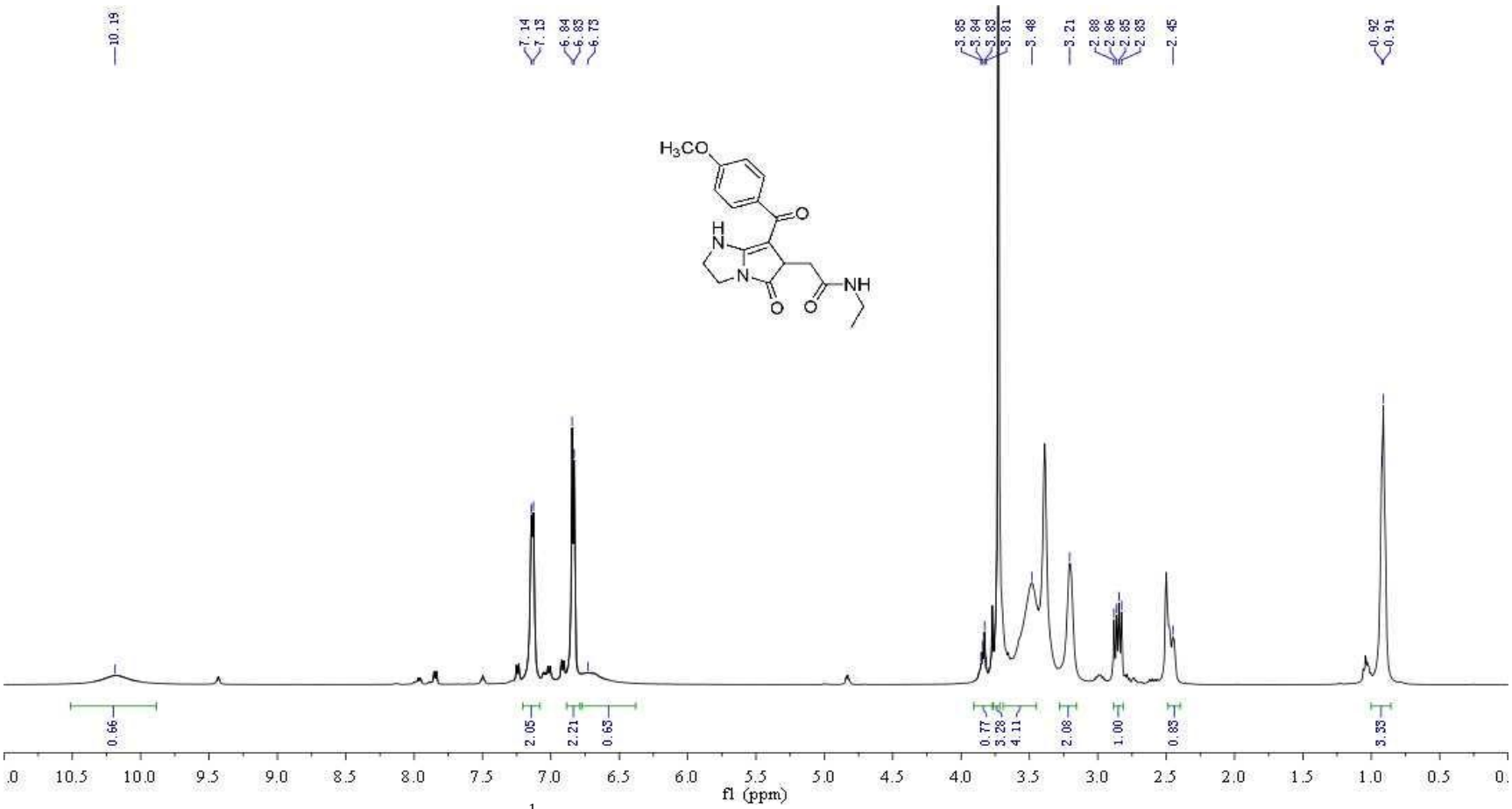


Figure 67. ^{1}H NMR (500 MHz, DMSO-d_6) spectra of compound **3h´**

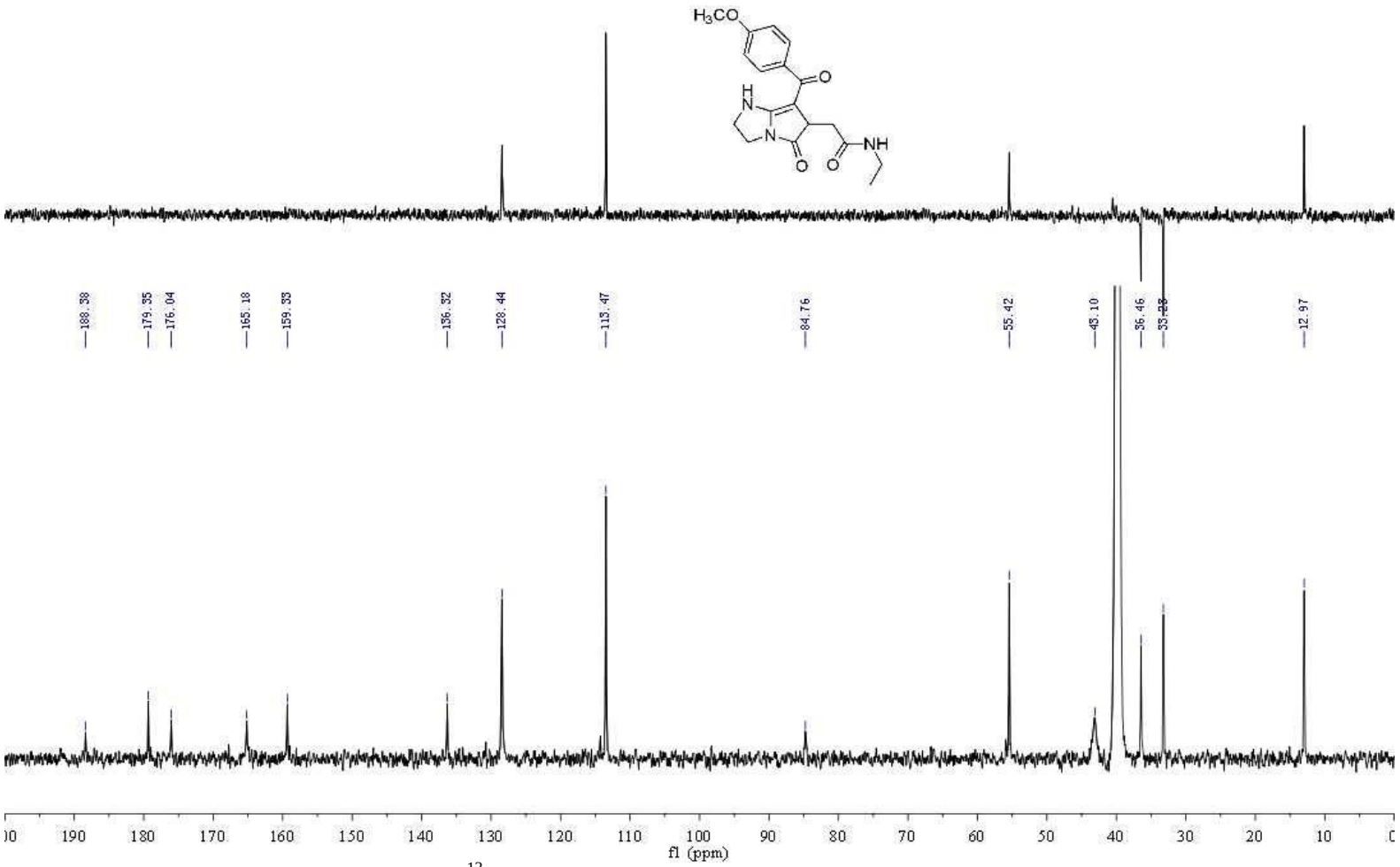


Figure 68. ^{13}C NMR (125 MHz, DMSO-d_6) spectra of compound **3h´**

References

1. Z.-T. Huang, M.-X. Wang, *Synthesis*, 1992, **12**, 1273; (*b*) Z.-J. Li, D. Charles, *Synth. Commun.*, 2001, **31**, 527.
2. Z.-C. Chen, Z.-G. Le, Z.-C. Chen, Y. Hu, Z.-G. Le, Q.-G. Zheng, *Synthesis*, 2004, **7**, 995
3. CCDC 998644 contain the supplementary crystallographic data for compound **3t**. These data can be obtained free of charge from The Cambridge Crystallographic Data Center via www.ccdc.cam.ac.uk/data_request/cif.