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SUPPLEMENTARY INFORMATION

Acylmethylidenepyrrolo[1,2-*c*]oxazoles via [3+2]-cyclization of 2-acylethynylpyrroles with carbonyl compounds

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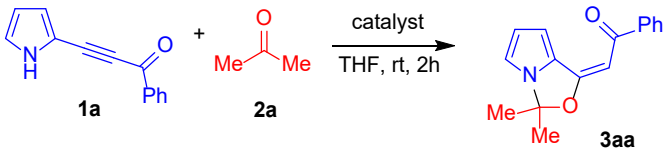
Contents

General Information

IR spectra were obtained on a "Bruker IFS-25" spectrometer (KBr pellets or films in 400-4000 cm^{-1} region). ^{1}H (400.13 MHz), ^{13}C (100.6 MHz) NMR spectra were recorded on a "Bruker Avance 400" instrument in $CDCl_3$. The assignment of signals in the ^{1}H NMR spectra was made using COSY and NOESY experiments. Resonance signals of carbon atoms were assigned based on ^{1}H-^{13}C HSQC and ^{1}H-^{13}C HMBC experiments. The ^{1}H chemical shifts (δ) were referenced to the residual solvent protons (7.26 ppm, $CDCl_3$), the ^{13}C chemical shifts were expressed with respect to the deuterated solvent (77.1 ppm). Coupling constants in hertz (Hz) were measured from one-dimensional spectra and multiplicities were abbreviated as following: br (broad), s (singlet), d (doublet), t (triplet), m (multiplet). The chemical shifts were recorded in ppm. The (C, H, N) microanalyses were performed on a Flash EA 1112 CHNS-O/MAS (CHN Analyzer) instrument. Sulfur was determined by complexometric titration with Chlorasenazo III. Melting points (uncorrected) were determined with SMP50 Stuart Automatic melting point (Stuart Scientific).

Table S1. Reaction of 1a with acetone. Screening Experiments.

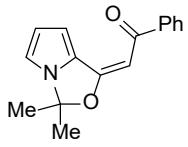
Acylethynylpyrrole **1a** (1 mmol) and acetone (1 mmol) were dissolved in THF (5 ml), then base was added. Reaction mixture was stirred at room temperature for 2 hours (TLC silica gel control, *n*-hexane-diethyl ether, 1:1). Then reaction mixture was diluted with cold water (12 ml) and extracted by diethyl ether (3×10 ml). Combined organic extracts were washed with water, dried over $CaCl_2$, solvent was removed and the residue was analyzed by ^{1}H NMR.



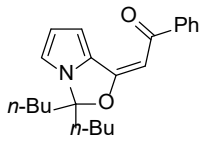
Entry	Base	Base amount, eq.	Composition of reaction mixture (^{1}H NMR data), %	
			Acylethynylpyrrole **1a**	Pyrrolooxazole **3aa** (isolated yield, %)
1.	LiOH	1	30	70
2.	LiOH	1.5	24	76
3.	NaOH	1	5	95
4.	KOH	0.5	7	93
5.	KOH	1	0	100 (70)
6.	(*n*-Bu)$_4$NOH (40% aq. soln.)	1	0	100 (68)
7.	(*n*-Bu)$_4$NF	1	~100	0
8.	K_2CO_3	1	~100	0
9.	TMEDA	1	~100	0
10.	*i*-Pr$_2$NEt	1	~100	0
11.	DABCO	1	~100	0

Synthesis of pyrrolo[1,2-*c*]oxazoles 3. General procedure.

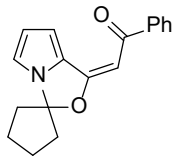
Acylethynylpyrrole **1** (1 mmol) and ketone or aldehyde **2** (1 mmol) were dissolved in THF (4-6 ml), then KOH (1 mmol) was added. Reaction mixture was stirred at room temperature for 2 hours. Then reaction mixture was diluted with cold water (12 ml) and extracted by diethyl ether (3×10 ml). Combined organic extracts were washed with water, dried over $CaCl_2$, solvent was removed to give the mixture containing *E*- and *Z*-isomers of pyrrole[1,2-*c*]oxazoles **3** (^{1}H NMR data). This mixture was fractionated by flash chromatography (SiO_2, *n*-hexane/diethyl ether, 5-10%) to afford pure *E*-isomer of pyrrole[1,2-*c*]oxazole **3**.



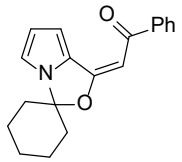
(*E*)-2-(3,3-Dimethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3aa). Yield: 0.177 g (70%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, Ph), 7.67-7.66 (m, 1H, H-3, pyrrole), 7.50-7.43 (m, 3H, $H_{m,p}$, Ph), 6.92 (m, 1H, H-5, pyrrole), 6.54 (m, 1H, H-4, pyrrole), 6.49 (s, 1H, HC=), 1.78 (s, 6H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.8, 160.0, 140.0, 131.6, 128.4 (2C), 128.0, 127.6 (2C), 116.3, 116.0, 112.5. 96.0, 92.9, 28.0 (2C); IR (film) 3120, 3103, 3060, 2989, 2932, 1656, 1560, 1537, 1457, 1389, 1354, 1312, 1253, 1221, 1178, 1127, 1051, 1038, 1006, 945, 888, 815, 776, 734, 699, 642, 484 cm^{-1}; Anal. Calcd for $C_{16}H_{15}NO_2$: C, 75.87; H, 5.97; N, 5.53; O, 12.63%. Found: C, 75.65; H, 6.32; N, 5.23%.



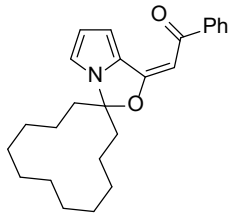
(*E*)-2-(3,3-Dibutyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3ab). Yield: 0.256 g (76%), yellow crystals, mp 103-104 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.99 (m, 2H, H_o, Ph), 7.67-7.66 (m, 1H, H-3, pyrrole), 7.50-7.45 (m, 3H, $H_{m,p}$, Ph), 6.86 (m, 1H, H-5, pyrrole), 6.55 (m, 1H, H-4, pyrrole), 6.50 (s, 1H, HC=), 2.14-2.06 (m, 2H, *n*-Bu), 1.98-1.90 (m, 2H, *n*-Bu), 1.26 (m, 6H, *n*-Bu), 0.85-0.81 (m, 8H, *n*-Bu); ^{13}C NMR (100.6 MHz, $CDCl_3$,): δ 188.8, 161.4, 140.2, 131.5, 129.7, 128.4 (2C), 127.7 (2C), 116.3, 116.2, 112.0, 99.9, 92.1, 39.7 (2C), 24.0 (2C), 22.4 (2C), 13.9 (2C); IR (film) 2956, 2930, 2869, 1653, 1554, 1455, 1357, 1249, 1215, 1164, 1132, 1037, 995, 961, 903, 850, 812, 772, 741, 699, 640 cm^{-1}; Anal. Calcd for $C_{22}H_{27}NO_2$: C, 78.30; H, 8.06; N, 4.15; O, 9.48%. Found: C, 78.01; H, 7.82; N, 4.01%.



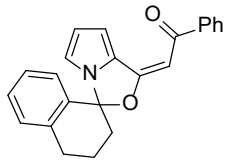
(*E*)-1-Phenyl-2-(1'*H*-spiro[cyclopentane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)ethan-1-one (3ac). Yield: 0.195 g (70%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, Ph), 7.67 (m, 1H, H-3, pyrrole), 7.50-7.43 (m, 3H, $H_{m,p}$, Ph), 6.93 (m, 1H, H-5, pyrrole); 6.55 (m, 1H, H-4, pyrrole); 6.50 (s, 1H, HC=); 2.25 (m, 2H, CH_2), 2.13-2.00 (m, 6H, CH_2); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.8, 160.1, 140.2, 131.6, 128.8, 128.5 (2C), 127.7 (2C), 116.5, 115.8, 112.2, 105.4, 92.9, 39.6 (2C), 23.9 (2C); IR (film): 3102, 2968, 1658, 1583, 1561, 1456, 1421, 1385, 1358, 1285, 1253, 1220, 1143, 1047, 980, 886, 814, 776, 732, 698, 641 cm^{-1}; Anal. Calcd for $C_{18}H_{17}NO_2$: C, 77.40; H, 6.13; N, 5.01; O, 11.45%. Found: C, 77.19; H, 6.35; N, 5.23%.



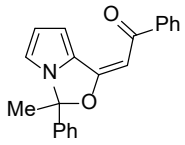
(*E*)-1-Phenyl-2-(1'*H*-spiro[cyclohexane-1,3'-pyrrolo[1,2-c]oxazol]-1'-ylidene)ethan-1-one (3ad). Yield: 0.226 g (77%), yellow crystals, mp 110 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.99 (m, 2H, H_o, Ph), 7.69-7.68 (m, 1H, H-3, pyrrole), 7.50-7.43 (m, 3H, $H_{m,p}$, Ph), 6.93 (m, 1H, H-5, pyrrole), 6.52-6.51 (m, 2H, H-4, pyrrole, HC=), 1.94-1.89 (m, 10H, cyclohexane); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.8, 160.4, 140.1, 131.6, 128.4 (2C), 128.0, 127.7 (2C), 116.5, 115.8, 112.3, 97.0, 92.9, 37.7 (2C), 24.4, 22.8 (2C); IR (film) 2940, 2862, 1705, 1654, 1555, 1535, 1455, 1386, 1354, 1294, 1275, 1245, 1219, 1164, 1126, 1049, 999, 951, 913, 874, 840, 814, 775, 733, 699, 639, 489 cm^{-1}; Anal. Calcd for $C_{19}H_{19}NO_2$: C, 77.79; H, 6.53; N, 4.77; O, 10.91%. Found: C, 77.44; H, 6.31; N, 4.65%.



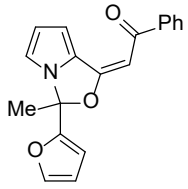
(*E*)-1-Phenyl-2-(1'*H*-spiro[cyclododecane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)ethan-1-one (3ae). Yield: 275 g (73%), white crystals, mp 184-186 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): 7.99-7.97 (m, 2H, H_o, Ph), 7.70-7.69 (m, 1H, H-3, pyrrole), 7.49-7.43 (m, 3H, $H_{m,p}$, Ph), 6.94 (m, 1H, H-5, pyrrole), 6.48 (m, 2H, H-4, pyrrole, HC=), 1.99-1.96 (m, 4H, CH_2), 1.68 (m, 4H, CH_2), 1.45 (m, 14H, CH_2); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.7, 160.4, 140.3, 131.5, 128.4, 128.3 (2C), 127.7 (2C), 117.5, 115.6, 112.3, 100.8, 92.7, 34.5 (2C), 26.0 (2C), 25.7, 22.8 (2C), 22.3 (2C), 19.6 (2C); IR (film): 3437, 3108, 2934, 2855, 1649, 1581, 1546, 1456, 1351, 1250, 1140, 1049, 1035, 1016, 987, 960, 903, 888, 875, 849, 807, 770, 748, 695, 636 cm^{-1}; Anal. Calcd for $C_{25}H_{31}NO_2$: C, 79.54; H, 8.28; N, 3.71; O, 8.48%. Found: C, 79.28; H, 8.51; N, 3.52%.



(*E*)-2-(3,4-Dihydro-1'*H*,2*H*-spiro[naphthalene-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-phenylethan-1-one (3af). Yield: 233 g (68%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, Ph); 7.78-7.77 (m, 1H, H-3, pyrrole), 7.52-7.42 (m, 3H, $H_{m,p}$, Ph), 7.33-7.29 (m, 1H, CH, tetralone), 7.23-7.21 (m, 1H, CH, tetralone), 7.15-7.11 (m, 1H, CH, tetralone), 6.79 (m, 1H, H-5, pyrrole), 6.64-6.62 (m, 1H, CH, tetralone), 6.58 (m, 1H, H-4, pyrrole), 6.52 (s, 1H, HC=), 3.01-2.97 (m, 2H, CH_2), 2.41-2.29 (m, 3H, CH_2), 2.16-2.13 (m, 1H, CH_2); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 189.0, 159.8, 140.1, 138.4, 133.9, 131.6, 130.1, 129.6, 129.2, 128.4 (2C), 127.7 (2C), 127.3, 126.4, 117.3, 116.6, 112.3, 95.5, 93.0, 37.2, 28.9, 20.1; IR (film): 3298, 3061, 2943, 1658, 1561, 1491, 1454, 1354, 1309, 1286, 1249, 1218, 1136, 1095, 1043, 958, 921, 874, 815, 763, 731, 698, 636 cm^{-1}; Anal. Calcd for $C_{23}H_{19}NO_2$: C, 80.92; H, 5.61; N, 4.10; O, 9.37%. Found: C, 80.69; H, 5.32; N, 4.38%.

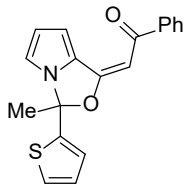


(*E*)-2-(3-Methyl-3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3ag). Yield: 0.192 g (61%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, COPh), 7.75-7.74 (m, 1H, H-3, pyrrole), 7.52-7.48 (m, 1H, H_p, Ph), 7.47-7.43 (m, 2H, H_m, Ph), 7.40-7.39 (m, 3H, $H_{m,p}$, COPh), 7.34-7.31 (m, 2H, H_o, Ph), 6.94-6.93 (m, 1H, H-5, pyrrole), 6.58-6.57 (m, 1H, H-4, pyrrole), 6.56 (s, 1H, HC=), 2.17 (s, 3H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.9, 159.9, 140.0, 139.4, 131.7, 129.8, 129.0 (2C), 128.8, 128.5 (2C), 127.7 (2C), 125.1 (2C), 117.3, 116.6, 112.6, 96.9, 93.2, 26.8; IR (film) 3062, 2928, 1657, 1583, 1560, 1495, 1452, 1385, 1352, 1284, 1258, 1218, 1152, 1117, 1098, 1042, 1001, 908, 815, 767, 734, 697, 480 cm^{-1}; Anal. Calcd for $C_{21}H_{17}NO_2$: C, 79.98; H, 5.43; N, 4.44; O, 10.15%. Found: C, 79.69; H, 5.19; N, 4.63%.

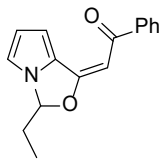


(*E*)-2-(3-(Furan-2-yl)-3-methyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3ah). Yield: 0.195 g (64%), red oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, Ph), 7.73-7.72 (m, 1H, H-3, pyrrole), 7.52-7.43 (m, 4H, $H_{m,p}$ Ph, H-3, furan), 6.92 (m, 1H, H-5, pyrrole), 6.58-6.56 (m, 1H, H-4, pyrrole), 6.56 (s, 1H, HC=), 6.37-6.36 (m, 1H, H-5, furan), 6.30-6.29 (m, 1H, H-4, furan), 2.15 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.9, 159.5, 151.1, 144.1, 140.0, 131.8, 128.8, 128.5 (2C), 127.7 (2C), 117.2, 116.7, 112.7, 110.8, 108.8, 93.5, 92.1, 25.3; IR (film): 3123, 2923, 1660, 1584, 1565, 1453,

1418, 1386, 1352, 1311, 1269, 1249, 1218, 1151, 1101, 1042, 900, 848, 817, 776, 734, 698, 640, 596 cm^{-1}; Anal. Calcd for $C_{19}H_{15}NO_3$: C, 74.74; H, 4.95; N, 4.59; O, 15.72%. Found: C, 74.47; H, 4.79; N, 4.90%.

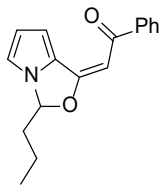


(*E*)-2-(3-Methyl-3-(thiophen-2-yl)-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3ai). Yield: 0.199 g (62%), yellow oil, ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, Ph), 7.73-7.72 (m, 1H, H-3, pyrrole), 7.50 (m, 1H, H_p, Ph), 7.45 (m, 2H, H_m, Ph), 7.36-7.35 (m, 1H, H-3, furan), 7.03 (m, 1H, H-5, thiophene), 6.99 (m, 1H, H-4, thiophene), 6.95 (m, 1H, H-5, pyrrole), 6.58 (m, 1H, H-4, pyrrole), 6.56 (s, 1H, HC=), 2.22 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.8, 159.1, 143.2, 139.8, 131.7, 128.4, 128.4 (2C), 127.6 (2C), 127.4, 127.2, 126.0, 117.3, 116.7, 112.6, 94.3, 93.4, 27.8; IR (film) 3104, 2993, 2932, 1659, 1583, 1561, 1451, 1419, 1384, 1286, 1250, 1217, 1149, 1097, 1043, 900, 852, 816, 776, 710, 637, 608, 540 cm^{-1}; Anal. Calcd for $C_{19}H_{15}NO_2S$: C, 71.01; H, 4.70; N, 4.36; O, 9.96; S, 9.98%. Found: C, 70.88; H, 4.42; N, 4.71; S, 10.12%



(*E*)-2-(3-Ethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3ak).

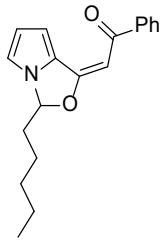
Yield: 0.172 g (68%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.98 (m, 2H, H_o, Ph), 7.70-7.69 (m, 1H, H-3, pyrrole), 7.51-7.44 (m, 3H, $H_{m,p}$, Ph), 6.95 (m, 1H, H-5, pyrrole), 6.55 (m, 2H, H-4, pyrrole, HC=), 6.06 (m, 1H, CH), 2.17-2.14 (m, 1H, CH_2), 1.99-1.95 (m, 1H, CH_2), 0.93 (m, 3H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.9, 161.0, 140.1, 131.7, 128.5 (2C), 127.8 (2C), 116.8, 116.4, 112.3, 92.9, 91.5, 77.4, 29.1, 6.4; IR (film): 3103, 3060, 2973, 2935, 1658, 1583, 1562, 1461, 1358, 1311, 1286, 1243, 1215, 1179, 1139, 1115, 1041, 959, 852, 814, 775, 733, 697, 642, 603 cm^{-1}; Anal. Calcd for $C_{16}H_{15}NO_2$: C, 75.87; H, 5.97; N, 5.53; O, 12.63%; Found: C, 75.59; H, 5.71; N, 5.70%;



(*E*)-1-Phenyl-2-(3-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (3al).

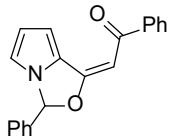
Yield: 0.192 g (72%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.98 (m, 2H, H_o, Ph), 7.69-7.68 (m, 1H, H-3, pyrrole), 7.51-7.45 (m, 3H, $H_{m,p}$, Ph), 6.95 (m, 1H, H-5, pyrrole), 6.54 (m, 2H, H-4, pyrrole, HC=), 6.07 (m, 1H, CH), 2.12-2.03 (m, 1H, CH_2), 1.96-1.87 (m, 1H, CH_2), 1.56-1.47 (m, 1H, CH_2), 1.40-1.31 (m, 1H, CH_2), 0.99 (m, 3H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.8, 160.9, 140.0, 131.7, 129.0, 128.5 (2C), 127.7 (2C), 116.9, 116.3, 112.3, 92.8, 90.7, 38.0, 15.9, 13.7; IR (film): 3003, 2961, 2874,

1655, 1580, 1550, 1460, 1431, 1380, 1355, 1310, 1283, 1245, 1214, 1137, 1110, 1021, 949, 873, 813, 728, 692, 640 cm^{-1}; Anal. Calcd for $C_{17}H_{17}NO_2$: C, 76.38; H, 6.41; N, 5.24; O, 11.97%; Found: C, 76.19; H, 6.69; N, 5.01%.



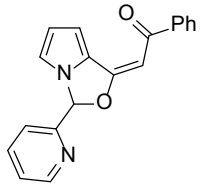
(*E*)-2-(3-Pentyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3am).

Yield: 0.209 g (71%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.98 (m, 2H, H_o, Ph), 7.70-7.69 (m, 1H, H-3, pyrrole), 7.52-7.43 (m, 3H, $H_{m,p}$, Ph), 6.95 (m, 1H, H-5, pyrrole), 6.54 (m, 2H, H-4, pyrrole, HC=), 6.07-6.04 (m, 1H, CH), 2.12-2.05 (m, 1H, CH_2), 1.97-1.88 (m, 1H, CH_2), 1.50-1.47 (m, 1H, CH_2), 1.33 (m, 5H, CH_2), 0.89 (m, 3H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.9, 160.9, 140.1, 131.7, 129.0, 128.5 (2C), 127.7 (2C), 116.9, 116.3, 112.3, 92.9, 90.9, 36.0, 31.4, 22.5, 22.1, 14.0; IR (film): 2955, 2930, 2862, 1659, 1580, 1563, 1461, 1358, 1310, 1286, 1245, 1215, 1138, 1110, 1043, 1020, 923, 855, 814, 774, 734, 697, 643 cm^{-1}; Anal. Calcd for $C_{19}H_{21}NO_2$: C, 77.26; H, 7.17; N, 4.74; O, 10.83%; Found: C, 77.02; H, 7.30; N, 4.45%.



(*E*)-1-Phenyl-2-(3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (3an)

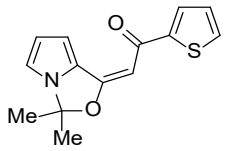
Yield: 0.193 g (64%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.98 (m, 2H, H_o, Ph), 7.78-7.77 (m, 1H, H-3, pyrrole), 7.51-7.45 (m, 6H, Ph), 7.31-7.29 (m, 2H, Ph), 6.87 (m, 1H, H-5, pyrrole), 6.84 (m, 1H, H-4, pyrrole), 6.60 (s, 1H, HC=), 6.59-6.57 (m, 1H, CH); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 189.0, 160.3, 140.0, 135.7, 131.8, 131.0, 129.4 (2C), 129.3, 128.5 (2C), 127.8 (2C), 127.0 (2C), 117.8, 116.9, 112.5, 93.3, 91.3; IR (film): 2922, 2853, 1658, 1580, 1563, 1541, 1454, 1351, 1309, 1284, 1243, 1214, 1132, 1101, 1044, 1012, 970, 920, 850, 810, 756, 733, 694, 624 cm^{-1}; Anal. Calcd for $C_{20}H_{15}NO_2$: C, 79.72; H, 5.02; N, 4.65; O, 10.62%; Found: C, 79.48; H, 5.30; N, 4.81%.



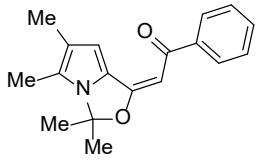
(*E*)-1-Phenyl-2-(3-(pyridin-2-yl)-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (3ao)

Yield: 0.208 g (69%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.69-8.68 (m, 1H, H-6, pyridine), 8.01-8.00 (m, 2H, H_o, Ph), 7.75 (m, 2H, H-3, pyrrole, H-3, pyridine), 7.52-7.45 (m, 3H, $H_{m,p}$, Ph), 7.39-7.36 (m, 1H, H-4, pyridine), 7.29-7.27 (m, 1H, H-5, pyridine), 7.07 (m, 1H, H-5, pyrrole), 6.97 (m, 1H, H-4,

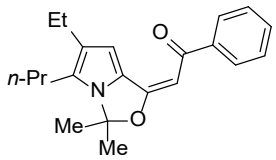
pyrrole), 6.69 (s, 1H, HC=), 6.55 (m, 1H, CH); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.9, 160.3, 155.2, 149.9, 139.9, 137.8, 131.9, 128.6 (2C), 128.5, 127.8 (2C), 125.1, 120.3, 118.2, 116.8, 112.7, 93.5, 90.7; IR (film): 3102, 3059, 3008, 1658, 1582, 1553, 1536, 1455, 1440, 1385, 1350, 1303, 1286, 1242, 1214, 1133, 1105, 1045, 1015, 990, 850, 816, 774, 748, 695, 639 cm^{-1}; Anal. Calcd for $C_{19}H_{14}N_2O_2$: C, 75.48; H, 4.67; N, 9.27; O, 10.58%; Found: C, 75.24; H, 4.67; N, 9.27%.



(*E*)-2-(3,3-Dimethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3ba). Yield: 0.189 g (73%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.69-7.68 (m, 1H, H-3, pyrrole), 7.62-7.61 (m, 1H, H-3, thiophene), 7.55-7.54 (m, 1H, H-5, thiophene), 7.11 (m, 1H, H-4, thiophene), 6.90 (m, H-5, pyrrole), 6.52 (m, 1H, H-4, pyrrole), 6.36 (s, 1H, HC=), 1.77 (s, 6H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 181.2, 159.7, 147.4, 131.6, 129.4, 128.0, 127.9, 116.3, 116.1, 112.5, 96.1, 92.7, 28.0 (2C); IR (film) 3102, 2989, 1644, 1560, 1517, 1457, 1415, 1353, 1304, 1256, 1222, 1126, 1088, 1055, 1002, 942, 910, 877, 854, 809, 728 cm^{-1}; Anal. Calcd for $C_{14}H_{13}NO_2S$: C, 64.84; H, 5.05; N, 5.40; O, 12.34; S, 12.36%. Found: C, 64.06; H, 37; N, 5.13; S, 12.48%.

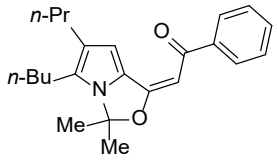


(*E*)-1-Phenyl-2-(3,3,5,6-tetramethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (3ca). Yield: 0.202 g (72%), yellow crystals, mp 103 °C, ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.17-8.15 (m, 2H, H_o, Ph), 7.54 (s, 1H, H-3, pyrrole), 7.45-7.43 (m, 3H, $H_{m,p}$, Ph), 6.37 (s, 1H, HC=), 2.30 (s, 3H, CH_3), 2.10 (s, 3H, CH_3), 2.79 (s, 6H, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.4, 160.0, 140.4, 131.3, 128.3 (2C), 127.6 (2C), 125.7, 125.6, 124.1, 113.6, 96.1, 91.3, 26.8 (2C), 11.5, 9.7; IR (film) 2921, 2862, 1655, 1582, 1546, 1501, 1455, 1386, 1343, 1247, 1169, 1099, 1042, 1003, 946, 893, 832, 773, 699, 637 cm^{-1}; Anal. Calcd for $C_{18}H_{19}NO_2$: C, 76.84; H, 6.81; N, 4.98; O, 11.37%. Found: C, 76.61; H, 6.52; N, 5.20%.

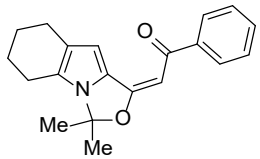


(*E*)-2-(6-Ethyl-3,3-dimethyl-5-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3da). Yield: 0.191 g (59%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.98-7.96 (m, 2H, H_o, Ph), 7.64 (s, 1H, H-3, pyrrole), 7.48-7.43 (m, 3H, $H_{m,p}$, Ph), 6.37 (s, 1H, HC=), 2.61 (m, 2H, CH_2), 2.50 (m, 2H, CH_2), 1.79 (s, 6H, CH_3), 1.61-1.59 (m, 2H, CH_2), 1.28-1.26 (m, 3H, CH_3), 1.02-1.00 (m, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.6, 160.0, 140.5, 131.3, 131.2, 130.3, 128.3 (2C), 127.6 (2C), 125.8, 112.3, 96.2, 91.4, 27.7 (2C), 27.1, 24.3, 19.6, 15.2, 14.2; IR (film) 2962, 2871, 1655, 1582, 1549, 1494, 1456,

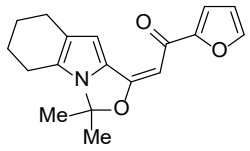
1385, 1356, 1311, 1244, 1158, 1104, 1056, 1016, 947, 893, 836, 772, 699 cm^{-1}; Anal. Calcd for $C_{21}H_{25}NO_2$: C, 77.98; H, 7.79; N, 4.33; O, 9.89%. Found: C, 77.61; H, 7.45; N, 4.18%.



(*E*)-2-(5-Butyl-3,3-dimethyl-6-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3ea). Yield 0.235 g (67%), dark red oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.98-7.96 (m, 2H, H_o, COPh), 7.60 (m, 1H, H-3, pyrrole), 7.45-7.43 (m, 3H, $H_{m,p}$, COPh), 6.37 (s, 1H, HC=), 2.62 (m, 2H, CH_2), 2.42 (m, 2H, CH_2), 1.79 (s, 6H, CH_3), 1.69-1.67 (m, 2H, CH_2), 1.54 (m, 2H, CH_2), 1.44-1.42 (m, 2H, CH_2), 0.99-0.97 (m, 6H, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.5, 160.0, 140.4, 131.2, 130.8, 129.4, 128.3 (2C), 127.5 (2C), 125.7, 112.8, 96.2, 91.3, 33.2, 28.6, 27.6 (2C), 24.8, 23.9, 22.9, 14.2, 13.8; IR (film) 2957, 2870, 1655, 1555, 1463, 1386, 1357, 1246, 1160, 1105, 1058, 1018, 948, 894, 835, 773, 700, 639 cm^{-1}; Anal. Calcd for $C_{23}H_{29}NO_2$: C, 78.59; H, 8.32; N, 3.99; O, 9.10%. Found: C, 78.29; H, 8.11; N, 4.23%

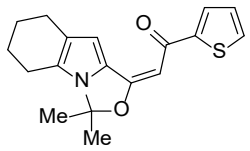


(*E*)-2-(3,3-Dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (3fa). Yield: 0.209 g (68%), yellow crystals; mp 131 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.98-7.00 (m, 2H, H_o, Ph), 7.50-7.42 (m, 4H, $H_{m,p}$, Ph, H-3, pyrrole), 6.39 (s, 1H, HC=), 2.72-2.70 (m, 2H, CH_2-7), 2.64-2.62 (m, 2H, CH_2-4), 1.88-1.84 (m, 4H, CH_2-5,6), 1.77 (s, 6H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.3, 160.3, 140.4, 131.3, 128.3 (2C), 128.2, 127.6, 127.5 (2C), 126.3, 126.1, 95.8, 91.5, 26.7 (2C), 23.5, 23.1, 22.8, 21.8; IR (film) 2934, 2852, 1654, 1598, 1541, 1503, 1444, 1383, 1306, 1241, 1175, 1151, 1099, 1062, 1024, 950, 909, 893, 835, 815, 774, 732, 698, 643, 487 cm^{-1}; Anal. Calcd for $C_{20}H_{21}NO_2$: C, 78.15; H, 6.89; N, 4.56; O, 10.41%. Found: C, 78.37; H, 6.58; N, 4.33%.

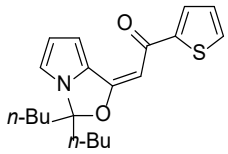


(*E*)-2-(3,3-Dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(furan-2-yl)ethan-1-one (3ga). Yield: 0.205 g (69%), orange crystals, mp 118 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.52 (s, 1H, H-3, pyrrole), 7.45 (m, 1H, H-5, furan), 7.18-7.17 (m, 1H, H-3, furan), 6.51-6.50 (m, 1H, H-4, furan), 6.29 (s, 1H, HC=), 2.70 (m, 2H, CH_2-7), 2.62 (m, 2H, CH_2-4), 1.89-1.87 (m, 2H, CH_2-6), 1.76 (m, 8H, CH_3, CH_2-5); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 177.2, 160.2, 154.9, 144.8, 128.4, 126.4, 113.9, 112.1, 111.5, 96.0, 91.1, 29.4, 26.8 (2C), 23.5, 23.1, 22.9, 21.9; IR (Film) 3116, 2924, 2852, 1783, 1702, 1660, 1648, 1583, 1502, 1467, 1443, 1383, 1306, 1251, 1221, 1179, 1150, 1126, 1085, 1030, 1011, 952, 931, 872,

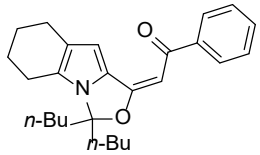
834, 802, 756, 699 cm^{-1}; Anal. Calcd for $C_{18}H_{19}NO_3$: C,72.71; H, 6.44; N, 4.71; O, 16.14%. Found: C,72.42; H, 6.21; N, 4.56;



(*E*)-2-(3,3-Dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3ha). Yield 0.213 g (68%), yellow crystals, mp 104 °C, ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.66-7.65 (m, 1H, H-5, thiophene), 7.51-7.50 (m, 1H, H-3, thiophene), 7.42 (s, 1H, H-3, pyrrole), 7.07 (m, 1H, H-4, thiophene), 6.25 (s, 1H, HC=), 2.69 (m, 2H, CH_2-7), 2.61 (m, 2H, CH_2-4), 1.86 (m, 2H, CH_2-6), 1.75 (m, 8H, CH_2-5, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 180.9, 160.0, 147.9, 131.1, 128.9, 128.2, 127.9, 126.3, 126.2, 111.5, 96.0, 91.3, 26.8 (2C), 23.5, 23.1, 22.8, 21.8; IR (film): 2933, 2851, 1648, 1545, 1417, 1371, 1246, 1172, 1150, 1081, 947, 910, 878, 853, 817, 785, 729, 648 cm^{-1}; Anal. Calcd for $C_{18}H_{19}NO_2S$: C, 68.98; H, 6.11; N, 4.47; O, 10.21; S, 10.23%. Found: C, 68.69; H, 6.32; N, 4.30; S, 10.01%.

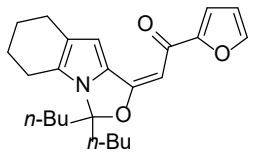


(*E*)-2-(3,3-Dibutyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3bb). Yield: 0.226 (66%), yellow crystals, mp 108 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.69-7.68 (m, 1H, H-3, pyrrole), 7.62-7.61 (m, 1H, H-5, thiophene), 7.55-7.54 (m, 1H, H-3, thiophene), 7.11 (m, 1H, H-4, thiophene), 6.84 (m, 1H, H-5, pyrrole), 6.53 (m, 1H, H-4, pyrrole), 6.37 (s, 1H, HC=), 2.13-2.05 (m, 2H, CH_2, *n*-Bu), 1.97-1.90 (m, 2H, CH_2, *n*-Bu), 1.28-1.24 (m, 6H, CH_2, *n*-Bu), 0.85-0.81 (m, 8H, CH_2, CH_3, *n*-Bu); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 181.2, 161.0, 147.6, 131.5, 129.6, 129.2, 127.9, 116.3, 116.2, 112.1, 100.0, 91.9, 39.7 (2C), 24.0 (2C), 22.3 (2C), 13.9 (2C); IR (film) 2957, 2869, 1648, 1556, 1455, 1416, 1381, 1356, 1287, 1250, 1156, 1129, 1083, 1059, 1035, 994, 909, 885, 858, 823, 796, 730, 648 cm^{-1}; Anal. Calcd for $C_{20}H_{25}NO_2S$: C, 69.94; H, 7.34; N, 4.08; O, 9.32; S, 9.33%. Found: C, 70.29; H, 7.52; N, 4.23; S, 9.12%.

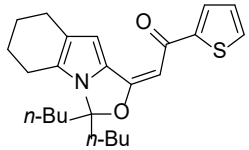


(*E*)-2-(3,3-Dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (3fb). Yield: 0.242 g (62%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.99 (m, 2H, H_o, Ph), 7.49-7.43 (m, 4H, $H_{m,p}$, Ph, H-3, pyrrole), 6.39 (s, 1H, HC=), 2.66-2.62 (m, 4H, CH_2-4,7), 2.11 (m, 2H, CH_2, *n*-Bu), 2.00 (m, 2H, CH_2, *n*-Bu), 1.86 (m, 2H, CH_2-6), 1.77 (m, 2H, CH_2-5), 1.26 (m, 4H, CH_2, *n*-Bu), 0.84 (m, 10H, CH_2, CH_3, *n*-Bu); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.4, 161.9, 140.5, 131.2, 128.3 (2C), 128.2, 128.1, 127.6 (2C), 126.1, 110.9, 99.9, 90.7, 38.5 (2C), 24.1 (2C), 23.6, 23.3, 23.0, 22.3 (2C), 22.2, 14.0 (2C) cm^{-1}; IR (film) 3060, 2923, 2859, 2360, 2162, 1654, 1541, 1502, 1444, 1383, 1325, 1305, 1235, 1152, 1101,

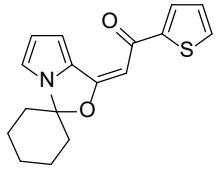
1051, 1026, 963, 905, 835, 815, 772, 699, 641 cm^{-1}; Anal. Calcd for $C_{26}H_{33}NO_2$: C, 79.76; H, 8.50; N, 5.58; O, 8.17%. Found: C, 79.52; H, 8.29; N, 5.71%.



(*E*)-2-(3,3-Dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(furan-2-yl)ethan-1-one (3gb). Yield: 0.297 g (78%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.51 (s, 1H, H-3, pyrrole), 7.43 (m, 1H, H-5, furan), 7.11-7.10 (m, 1H, H-3, furan), 6.50-6.49 (m, 1H, H-4, furan), 6.31 (s, 1H, HC=), 2.63-2.61 (m, 4H, CH_2-4,7), 2.05 (m, 2H, CH_2, *n*-Bu), 1.98 (m, 2H, CH_2, *n*-Bu), 1.83 (m, 2H, CH_2-6), 1.78 (m, 2H, CH_2-5), 1.25 (m, 6H, CH_2, *n*-Bu), 0.83 (m, 8H, CH_2, CH_3, *n*-Bu); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 177.5, 161.7, 155.2, 144.6, 128.3, 128.1, 126.2, 113.6, 112.1, 111.0, 100.1, 90.3, 38.5 (2C), 24.1 (2C), 23.6, 23.3, 23.0, 22.4 (2C), 22.2, 14.0 (2C); IR (film) 2930, 2860, 1653, 1575, 1550, 1502, 1468, 1443, 1381, 1333, 1321, 1304, 1263, 1239, 1224, 1177, 1151, 1132, 1105, 1083, 1060, 1009, 962, 937, 909, 882, 834, 802, 752, 733 cm^{-1}; Anal. Calcd for $C_{24}H_{31}NO_3$: C,75.56; H, 8.19; N, 3.67; O, 12.58%. Found: C,75.21; H, 8.02; N, 3.90%.

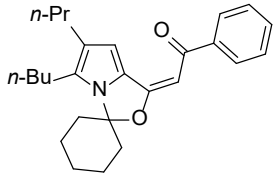


(*E*)-2-(3,3-Dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3hb). Yield 0.290 g (73%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.65-7.64 (m, 1H, H-5, thiophene), 7.50-7.49 (m, 1H, H-3, thiophene), 7.42 (s, 1H, H-3, pyrrole), 7.09 (m, 1H, H-4, thiophene), 6.27 (s, 1H, HC=), 2.61 (m, 4H, CH_2-4,7), 2.09 (m, 2H, CH_2, *n*-Bu), 1.98 (m, 2H, CH_2, *n*-Bu), 1.86-1.85 (m, 2H, CH_2-6), 1.77 (m, 2H, CH_2-5), 1.25 (m, 6H, CH_2, *n*-Bu), 0.83 (m, 8H, CH_2, CH_3, *n*-Bu); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 180.9, 161.5, 148.1, 131.0, 128.7, 128.2, 128.0, 127.8, 126.1, 111.0, 100.0, 90.5, 38.5 (2C), 24.1 (2C), 23.5, 23.2, 23.0, 22.3 (2C), 22.1, 14.0 (2C); IR (film): 2926, 2860, 1644, 1548, 1418, 1384, 1246, 1152, 1064, 909, 829, 788, 731, 648 cm^{-1}; Anal. Calcd for $C_{24}H_{31}NO_2S$: C, 72.51; H, 7.86; N, 3.52; O, 8.05; S, 8.06%. Found: C, 72.20; H, 7.59; N, 3.29; S, 8.20%.

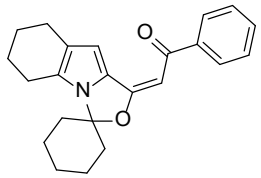


(*E*)-2-(1'H-Spiro[cyclohexane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-(thiophen-2-yl)ethan-1-one (3bd). Yield: 0.206 g (69%), yellow crystals; mp 127 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.70-7.69 (m, 1H, H-3, pyrrole), 7.63-7.62 (m, 1H, H-5, thiophene), 7.54-7.53 (m, 1H, H-3, thiophene), 7.12-7.10 (m, H-4, thiophene), 6.92 (m, H-5, pyrrole), 6.50-6.49 (m, 1H, H-4, pyrrole), 6.39 (s, 1H, HC=), 1.93-1.86 (m, 10H, CH_2, cyclohexyl); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 181.0, 159.9, 147.4, 131.4, 129.2, 127.9, 127.8, 116.5,

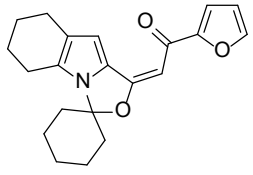
115.7, 112.2, 97.1, 92.5, 37.5 (2C), 24.2, 22.6 (2C); IR (film) 2940, 2862, 1648, 1637, 1566, 1540, 1517, 1454, 1415, 1384, 1353, 1295, 1277, 1246, 1231, 1160, 1144, 1124, 1083, 1058, 1032, 947, 911, 849, 835, 809, 731, 644, 605 cm^{-1}; Anal. Calcd for $C_{17}H_{17}NO_2S$: C, 68.20; H, 5.72; N, 4.68; O, 10.69; S, 10.71%. Found: C, 68.03; H, 5.51; N, 4.54; S, 10.90%.



(*E*)-2-(5'-Butyl-6'-propyl-1'*H*-spiro[cyclohexane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-phenylethan-1-one (3cd). Yield 0.266 g (68%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.99-7.97 (m, 2H, H_o, Ph), 7.63 (s, 1H, H-3, pyrrole), 7.46-7.42 (m, 3H, $H_{m,p}$, Ph), 6.41 (s, 1H, HC=), 2.63 (m, 2H, CH_2), 2.42 (m, 2H, CH_2), 2.02-1.97 (m, 2H, CH_2, cyclohexyl), 1.91-1.85 (m, 8H, CH_2, cyclohexyl), 1.71-1.65 (m, 2H, CH_2), 1.55-1.51 (m, 2H, CH_2), 1.45-1.40 (m, 2H, CH_2), 1.00-0.95 (m, 6H, CH_3); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.5, 160.3, 140.6, 131.2, 129.5, 128.3 (2C), 127.6 (2C), 125.9, 112.9, 97.6, 91.4, 37.1 (2C), 33.3, 31.7, 28.6, 25.0, 24.7, 23.4, 23.2 (2C), 23.0, 14.3, 13.9; IR (film) 2930, 2868, 1653, 1547, 1386, 1355, 1260, 1237, 1168, 1106, 1053, 955, 908, 876, 838, 772, 732, 699, 634 cm^{-1}; Anal. Calcd for $C_{26}H_{33}NO_2$: C, 79.76; H, 8.50; N, 3.58; O, 8.17%. Found: C, 79.44; H, 8.62; N, 3.26%.

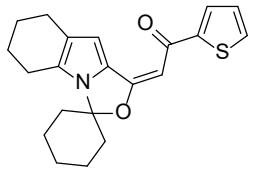


(*E*)-1-Phenyl-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)ethan-1-one (3fd). Yield: 0.285 g (82%), yellow crystals, mp 177 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.98 (m, 2H, H_o, Ph), 7.51-7.43 (m, 4H, $H_{m,p}$, Ph, H-3, pyrrole), 6.39 (s, 1H, HC=), 2.72 (m, 2H, CH_2-7), 2.63 (m, 2H, m, 2H, CH_2-4), 2.02 (m, 2H, CH_2-6), 1.87-1.78 (m, 12H, CH_2, cyclohexyl, CH_2-5); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.3, 160.7, 140.4, 131.3, 128.4, 128.3 (2C), 127.6 (2C), 126.6, 126.2, 111.4, 97.1, 91.6, 36.2 (2C), 24.6, 23.6, 23.2, 23.0 (2C), 23.0, 22.3; IR (film) 3437, 2936, 2856, 1653, 1580, 1542, 1445, 1382, 1333, 1306, 1259, 1230, 1202, 1150, 1098, 1049, 955, 905, 875, 836, 815, 773, 696, 633 cm^{-1}; Anal. Calcd for $C_{23}H_{25}NO_2$: C, 79.51; H, 7.25; N, 4.03; O, 9.21%. Found: C, 79.22; H, 7.03; N, 4.31%.

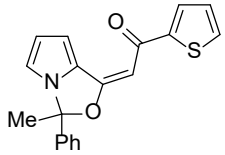


(*E*)-1-(Furan-2-yl)-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)ethan-1-one (3gd). Yield: 0.212 g (63%), yellow crystals, mp 196 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.54 (m, 1H, H-5, furan), 7.49 (s, 1H, H-3, pyrrole), 7.41-7.40 (m, 1H, H-3, furan), 6.53-6.52 (m,

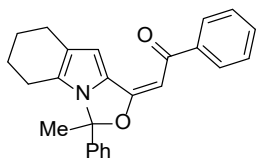
1H, H-4, furan), 6.32 (s, 1H, HC=), 2.74-2.71 (m, 2H, CH_2-7), 2.64-2.61 (m, 2H, CH_2-4), 2.04-2.02 (m, 2H, CH_2-6), 1.85-1.77 (m, 10H, CH_2, cyclohexyl), 1.77-1.75 (m, 2H, CH_2-5); ^{13}C NMR ($CDCl_3$, 100.6 MHz) δ 177.2, 160.4, 154.9, 144.6, 128.5, 126.5, 126.2, 113.7, 112.1, 111.4, 97.2, 91.0, 36.1 (2C), 24.5, 23.5, 23.2, 22.9 (2C), 22.9, 22.2; IR (film) 2937, 2856, 1646, 1573, 1501, 1470, 1382, 1305, 1279, 1264, 1242, 1202, 1178, 1151, 1065, 1010, 959, 905, 858, 836, 816, 732 cm^{-1}; Anal. Calcd for $C_{21}H_{23}NO_3$: C, 74.75; H, 6.87; N, 4.15; O, 14.22 %. Found: C, 74.92; H, 6.58; N, 4.06%.



(*E*)-2-(5',6',7',8'-Tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)-1-(thiophen-2-yl)ethan-1-one (3hd). Yield: 0.226 g (64%), yellow crystals, mp 175 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.67-7.66 (m, 1H, H-5, thiophene), 7.51-7.50 (m, 1H, H-3, thiophene), 7.45 (s, 1H, H-3, pyrrole), 7.09 (m, 1H, H-4, thiophene), 6.28 (s, 1H, HC=), 2.73-2.71 (m, 2H, CH_2-7), 2.62-2.60 (m, 2H, CH_2-4), 2.02-2.00 (m, 2H, CH_2-6), 1.89-1.85 (m, 10H, CH_2, cyclohexyl), 1.77-1.75 (m, 2H, CH_2-5); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 180.8, 160.3, 147.9, 131.0, 128.9, 128.4, 127.9, 126.5, 126.2, 111.5, 97.2, 91.4, 36.1 (2C), 24.6, 23.5, 23.2, 23.0 (2C), 23.0, 22.2; IR (film): 2935, 2854, 1645, 1542, 1418, 1385, 1260, 1236, 1151, 1067, 954, 907, 836, 787, 731 cm^{-1}; Anal. Calcd for $C_{21}H_{23}NO_2S$: C, 71.36; H, 6.56; N, 3.96; O, 9.05; S, 9.07%. Found: C, 71.07; H, 6.33; N, 4.18; S, 9.33%

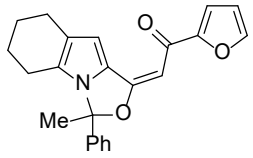


(*E*)-2-(3-Methyl-3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3bg). Yield: 0.193 g (60%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.70-7.67 (m, 2H, H-5, thiophene, H_p, Ph), 7.55-7.54 (m, 1H, H-3, pyrrole), 7.40-7.38 (m, 3H, H-3, thiophene, H_m, Ph), 7.33-7.30 (m, 2H, H_o, Ph), 7.12-7.10 (m, 1H, H-4, thiophene), 6.91 (m, 1H, H-5, pyrrole), 6.57-6.55 (m, 1H, H-4, pyrrole), 6.42 (s, 1H, HC=), 2.17 (s, 3H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 181.3, 159.5, 147.4, 139.3, 131.8, 129.8, 129.5, 129.1 (2C), 128.8, 128.1, 125.1 (2C), 117.3, 116.7, 112.7, 97.0, 93.1, 26.8; IR (film) 1646, 1562, 1539, 1415, 1348, 1351, 1289, 1258, 1218, 1147, 1098, 1051, 892, 858, 808, 764, 728, 696 cm^{-1}; Anal. Calcd for $C_{19}H_{15}NO_2S$: C, 71.01; H, 4.70; N, 4.36; O, 9.96; S, 9.98%. Found: C, 70.72; H, 4.39; N, 4.16; S, 9.77%.

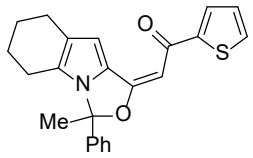


(*E*)-2-(3-Methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (3fg). Yield: 0.214 g (58%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.98-7.96

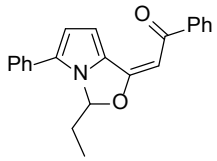
(m, 2H, H_o, COPh), 7.60 (s, 1H, H-3, pyrrole), 7.48-7.39 (m, 6H, $H_{m,p}$, Ph, COPh), 7.24-7.23 (m, 2H, H_o, Ph), 6.41 (s, 1H, HC=), 2.65 (m, 2H, CH_2-7), 2.54 (m, 1H, CH_2-4), 2.20 (m, 4H, CH_3, CH_2-4), 1.72 (m, 4H, CH_2-5,6); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 188.5, 160.2, 140.3, 138.5, 131.4, 129.8, 129.3, 129.0 (2C), 128.4 (2C), 127.6 (2C), 127.2, 126.5, 125.7 (2C), 111.5, 96.3, 91.8, 24.5, 23.5, 23.1, 22.7, 22.0; IR (film) 2930, 2852, 1654, 1545, 1501, 1448, 1381, 1334, 1305, 1238, 1160, 1051, 1028, 1010, 906, 840, 814, 768, 732, 696, 647, 615 cm^{-1}; Anal. Calcd for $C_{25}H_{23}NO_2$: C, 81.27; H, 6.28; N, 3.79; O, 8.66%. Found: C, 81.50; H, 6.44; N, 3.55%.



(*E*)-1-(Furan-2-yl)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)ethan-1-one (3gg). Yield 0.230 g (64%), yellow crystals, mp 132-133 °C, ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.55 (s, 1H, H-3, pyrrole), 7.51 (m, 1H, H-5, furan), 7.40-7.39 (m, 3H, $H_{m,p}$, Ph), 7.24-7.23 (m, 2H_o, Ph), 7.11-7.10 (m, 1H, H-3, furan), 6.50-6.49 (m, 1H, H-4, furan), 6.33 (s, 1H, HC=), 2.66 (m, 2H, CH_2-7), 2.54-2.50 (m, 1H, CH_2-4), 2.21-2.19 (m, 4H, CH_3, CH_2-4), 1.75-1.70 (m, 4H, CH_2-5,6); ^{13}C NMR ($CDCl_3$, 100.6 MHz) δ 177.6, 160.0, 155.0, 144.8, 138.5, 129.8, 129.4, 129.0 (2C), 127.2, 126.7, 125.8 (2C), 113.9, 112.1, 111.6, 96.5, 91.4, 24.5, 23.6, 23.2, 22.8, 22.0; IR (film) 2934, 2851, 1651, 1574, 1547, 1468, 1376, 1304, 1263, 1176, 1151, 1055, 1009, 884, 824, 763, 732, 697 cm^{-1}; Anal. Calcd for $C_{23}H_{21}NO_3$: C,76.86; H, 5.89; N, 3.90; O, 13.35%. Found: C,76.52; H, 6.16; N, 4.11%.

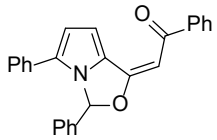


(*E*)-2-(3-Methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (3hg). Yield: 0.259 g (69%), yellow crystals, mp 125 °C, ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 7.63-7.62 (m, 1H, H-5, thiophene), 7.54 (s, 1H, H-3, pyrrole), 7.51-7.52 (m, 1H, H-3, thiophene), 7.39-7.38 (m, 3H, $H_{m,p}$, Ph), 7.23-7.21 (m, 2H, H_o, Ph), 7.08 (m, 1H, H-4, thiophene), 6.28 (s, 1H, HC=), 2.63 (m, 2H, CH_2-7), 2.48 (m, 1H, CH_2-4), 2.19 (m, 4H, CH_3, CH_2-4), 1.75-1.70 (m, 4H, CH_2-5,6); ^{13}C NMR ($CDCl_3$, 100.6 MHz): δ 181.1, 159.9, 147.9, 138.4, 131.3, 129.8, 129.4, 129.0 (2C), 127.9, 126.6, 125.7 (2C), 111.7, 96.5, 91.6, 24.5, 23.5, 23.1, 22.7, 22.0; IR (film): 2933, 2851, 1643, 1546, 1417, 1372, 1243, 1153, 1058, 893, 817, 731, 648 cm^{-1}; Anal. Calcd for $C_{21}H_{23}NO_2S$: C, 73.57; H, 5.64; N, 3.73; O, 8.52; S, 8.54%. Found: C, 73.70; H, 5.86; N, 3.31; S, 8.80%



(*E*)-2-(3-Ethyl-5-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3ik)

Yield: 0.243 g (74%), yellow oil; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.01-8.00 (m, 2H, Ph), 7.82 (d, *J* = 3.9 Hz, 1H, H-3, pyrrole), 7.51-7.42 (m, 7H, Ph), 7.35-7.32 (m, 1H, Ph), 6.70 (d, *J* = 3.9 Hz, 1H, H-4, pyrrole), 6.59 (s, 1H, HC=), 6.54-6.53 (m, 1H, CH), 2.03-1.96 (m, 1H, CH_2), 1.77-1.70 (m, 1H, CH_2), 0.69-0.66 (m, 3H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.8, 160.7, 140.2, 133.2, 131.7, 131.2, 130.7, 129.2 (2C), 128.5 (2C), 128.0, 127.8 (2C), 126.3 (2C), 115.6, 113.4, 92.8, 92.1, 27.5, 6.0; IR (film): 3062, 2973, 2936, 1655, 1581, 1553, 1516, 1455, 1364, 1327, 1302, 1249, 1223, 1176, 1115, 1047, 963, 851, 812, 758, 695, 671 cm^{-1}; Anal. Calcd for $C_{22}H_{19}NO_2$: C, 80.22; H, 5.81; N, 4.25; O, 9.71%; Found: C, 79.94; H, 6.03; N, 4.54%.

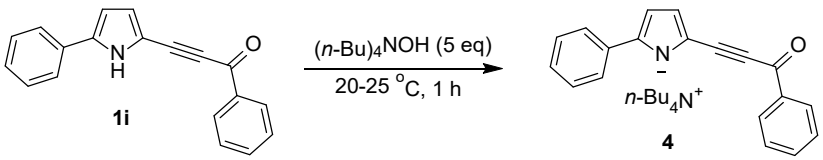


(*E*)-2-(3,5-Diphenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (3in)

Yield: 0.162 g (43%), yellow crystals, mp 185 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.00-7.98 (m, 2H, H_o, Ph), 7.95-7.94 (m, 1H, H-3, pyrrole), 7.51-7.43 (m, 3H, Ph), 7.30-7.26 (m, 5H, Ph), 7.24-7.20 (m, 3H, CH), 7.18-7.16 (m, 1H, Ph), 7.10-7.08 (m, 2H, Ph), 6.81-6.80 (m, 1H, H-4, pyrrole), 6.57 (s, 1H, HC=); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 188.9, 159.5, 140.1, 135.5, 134.1, 131.8, 131.0, 130.7, 130.6, 129.2 (2C), 128.7 (2C), 128.5 (2C), 127.8 (3C), 126.9 (2C), 126.4 (2C), 115.8, 113.8, 93.4, 92.1; IR (film): 3064, 3035, 1655, 1581, 1555, 1455, 1355, 1314, 1287, 1249, 1222, 1173, 1111, 1045, 1019, 976, 852, 758, 694 672 cm^{-1}; Anal. Calcd for $C_{26}H_{19}NO_2$: C, 82.74; H, 5.07; N, 3.71; O, 8.48%; Found: C, 82.51; H, 5.29; N, 3.67%.

Synthesis of tetrabutylammonium salt 4

1-Phenyl-3-(5-phenyl-1*H*-pyrrol-2-yl)prop-2-yn-1-one **1i** (0.271 g, 1 mmol) was dissolved in THF (4-6 ml), then *n*-Bu_4NOH (5 mmol, 40% aq. soln.) was added. Reaction mixture was stirred at room temperature for 1 hours. Then reaction mixture was diluted with *n*-hexane and the precipitate formed was filtered off to give a pure tetrabutylammonium salt **4**.

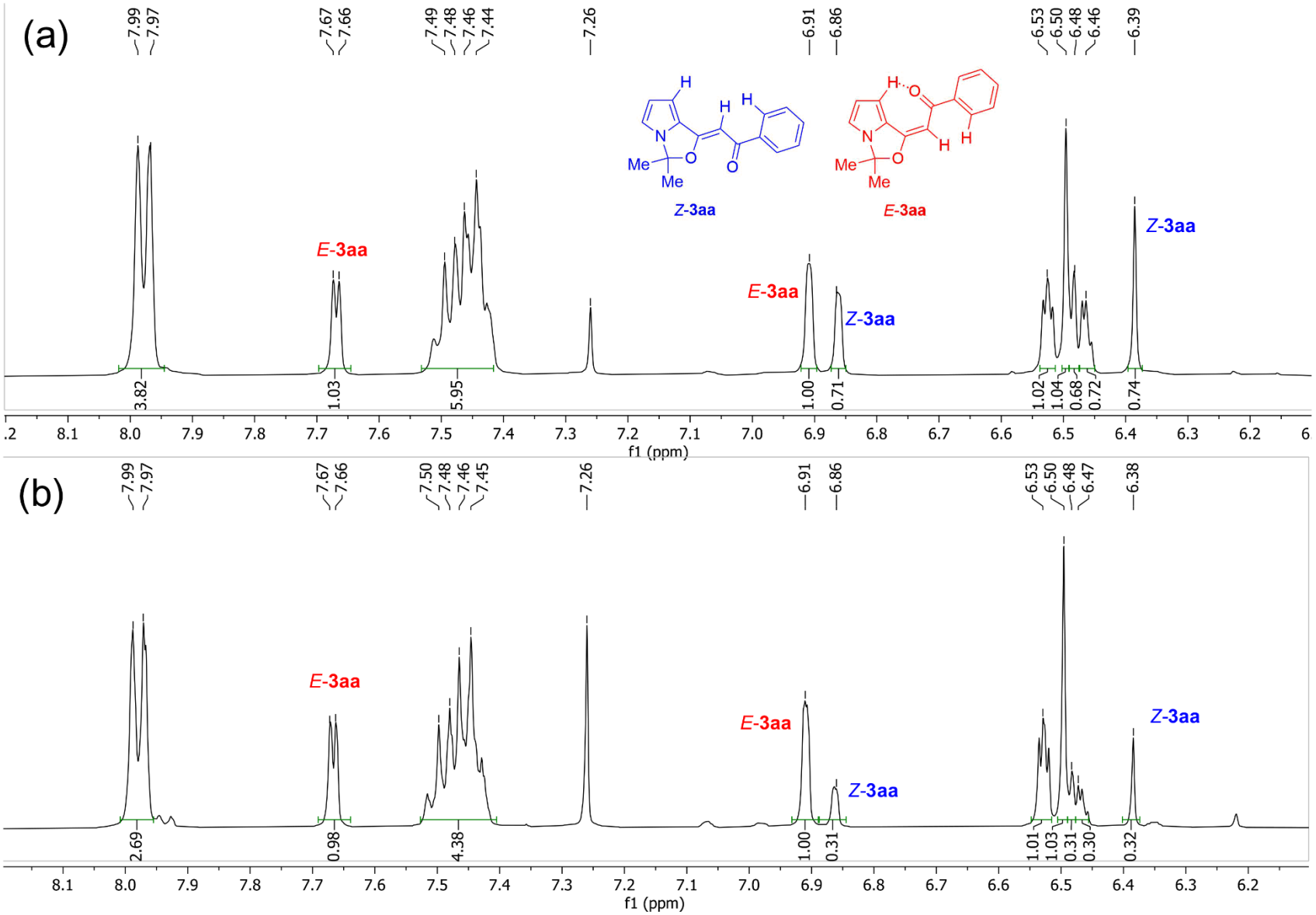


1i **4**

Tetrabutylammonium 2-(3-oxo-3-phenylprop-1-yn-1-yl)-5-phenylpyrrol-1-ide (4)

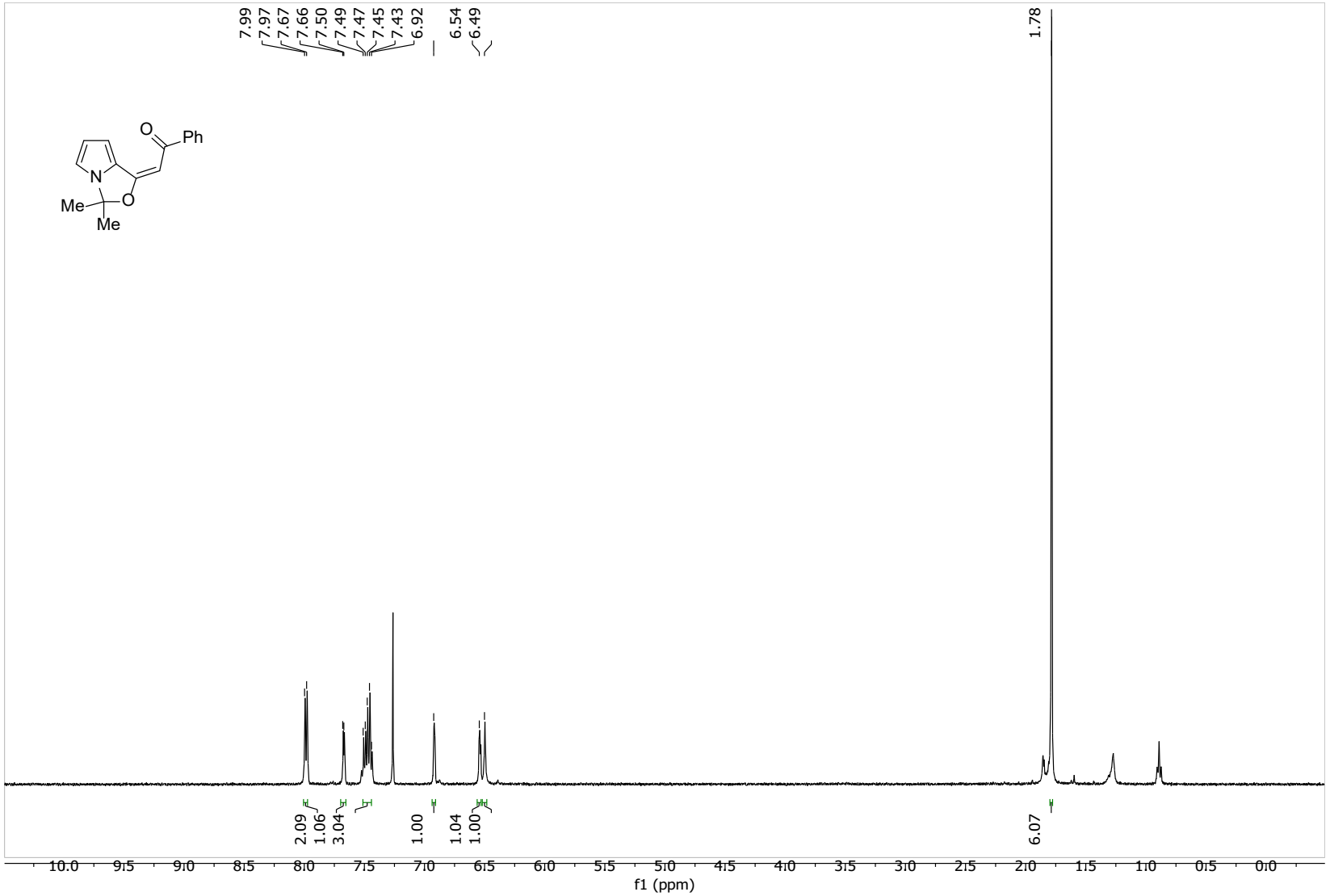
Yield: 0.496 g (97%), orange crystals, mp 155-156 °C; ^{1}H NMR (400.13 MHz, $CDCl_3$): δ 8.24-8.22 (m, 2H, Ph), 7.84-7.82 (m, 2H, Ph), 7.52-7.48 (m, 1H, Ph), 7.48-7.41 (m, 2H, Ph), 7.26-7.22 (m, 2H, Ph), 7.06-7.03 (m, 1H, Ph), 6.97 (d, *J* = 3.4 Hz, 1H, H-3), 6.62 (d, *J* = 3.4 Hz, 1H, H-4), 2.94-2.90 (m, 8H, CH_2), 1.40-1.32 (m, 8H, CH_2), 1.29-1.20 (m, 8H, CH_2), 0.85 (t, *J* = 7.1 Hz, 12H, CH_3); ^{13}C NMR (100.6 MHz, $CDCl_3$): δ 176.2, 147.1, 145.6, 138.5, 138.2, 132.3, 129.0 (2C), 128.3 (2C), 128.2 (2C), 124.8, 124.6 (2C), 124.5, 108.6, 94.7, 77.4, 58.1 (4C), 23.8 (4C), 19.6 (4C), 13.7 (4C); IR (film) 3059, 2962, 2874, 2364, 2113, 1596, 1566, 1467, 1444, 1381, 1313, 1238, 1211, 1170, 1019, 953, 882, 773, 739, 701, 651 cm^{-1}; Anal. Calcd for $C_{35}H_{48}N_2O$: C, 81.98; H, 9.44; N, 5.46; O, 3.12%. Found: C, 81.61; H, 9.67; N, 5.21%.

Figure S1: *E/Z*-ratios of products in the reaction of acylethynylpyrrole 1a with acetone in THF after 2h (a) and 4h (b).

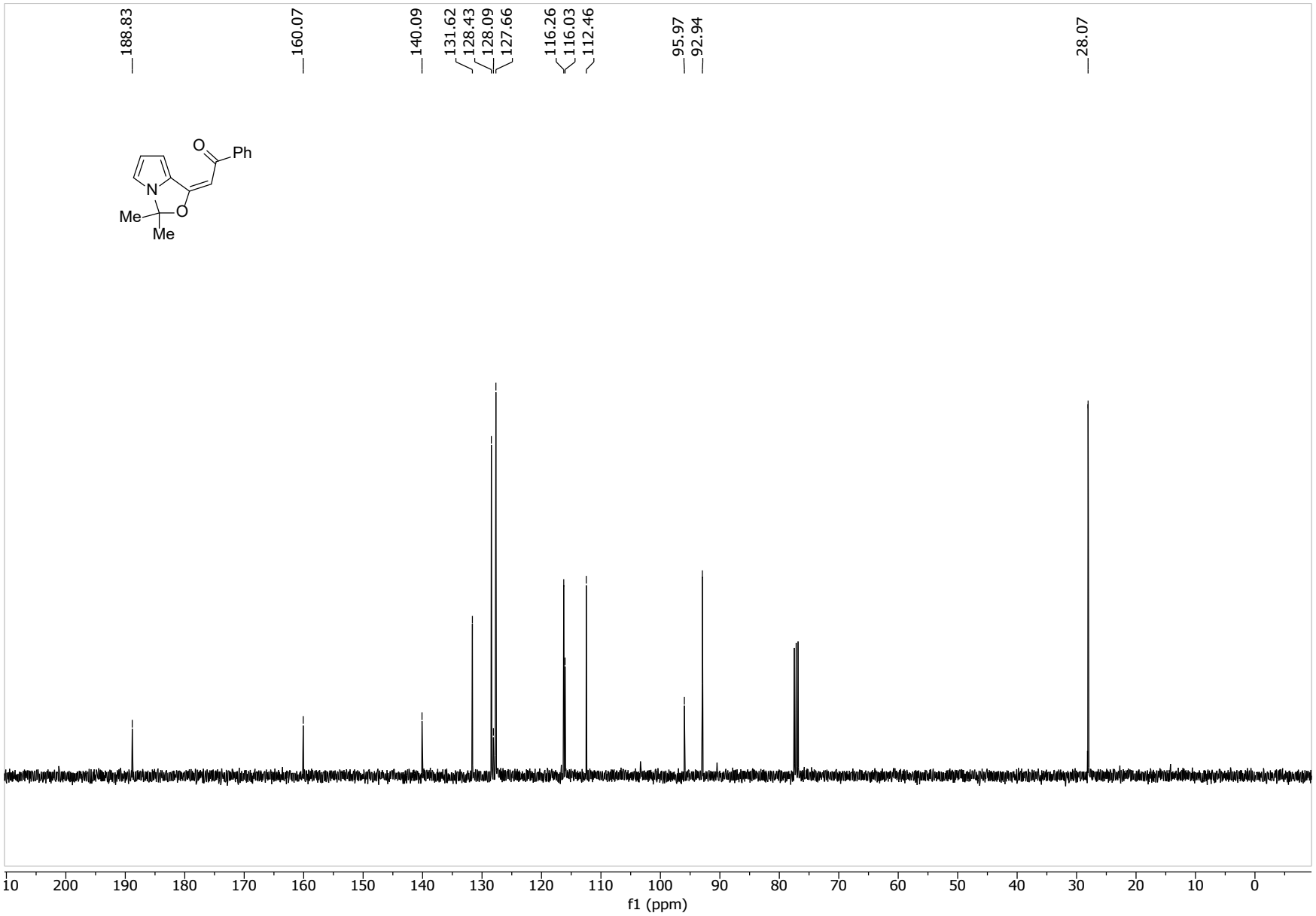


NMR Spectra of synthesized compounds

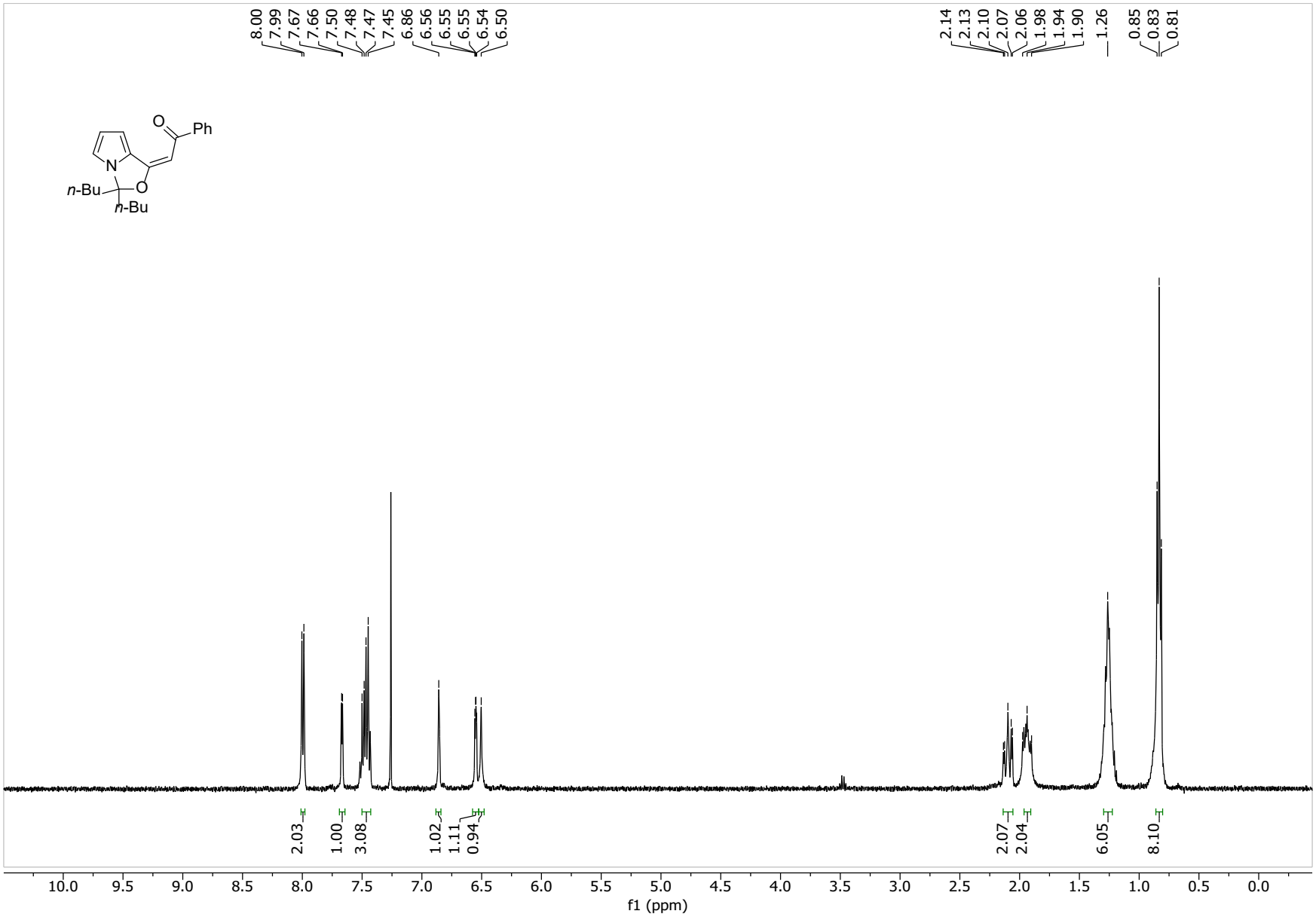
^{1}H NMR spectrum of (*E*)-2-(3,3-dimethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3aa**) in $CDCl_3$



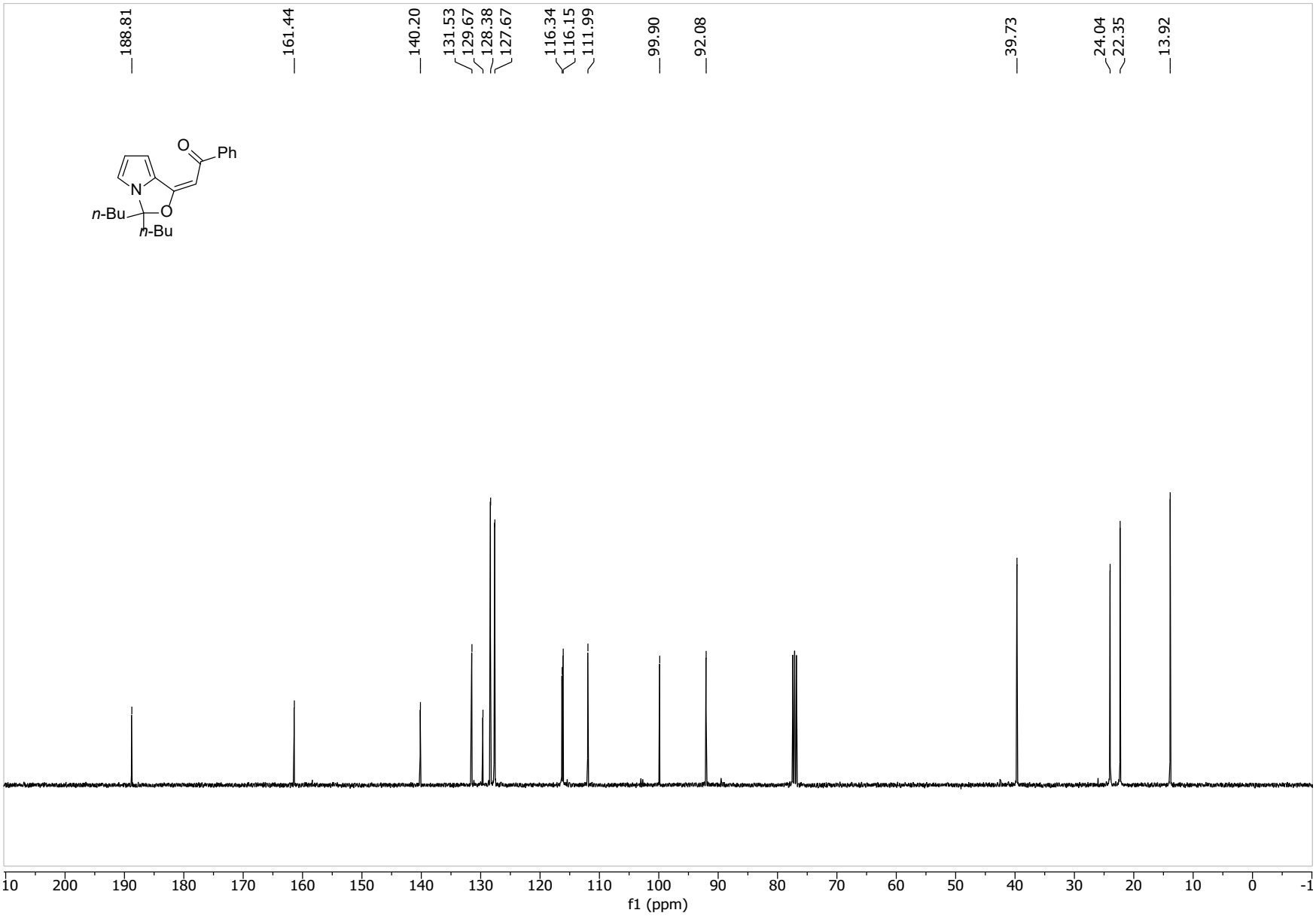
^{13}C NMR spectrum of (*E*)-2-(3,3-dimethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3aa**) in $CDCl_3$



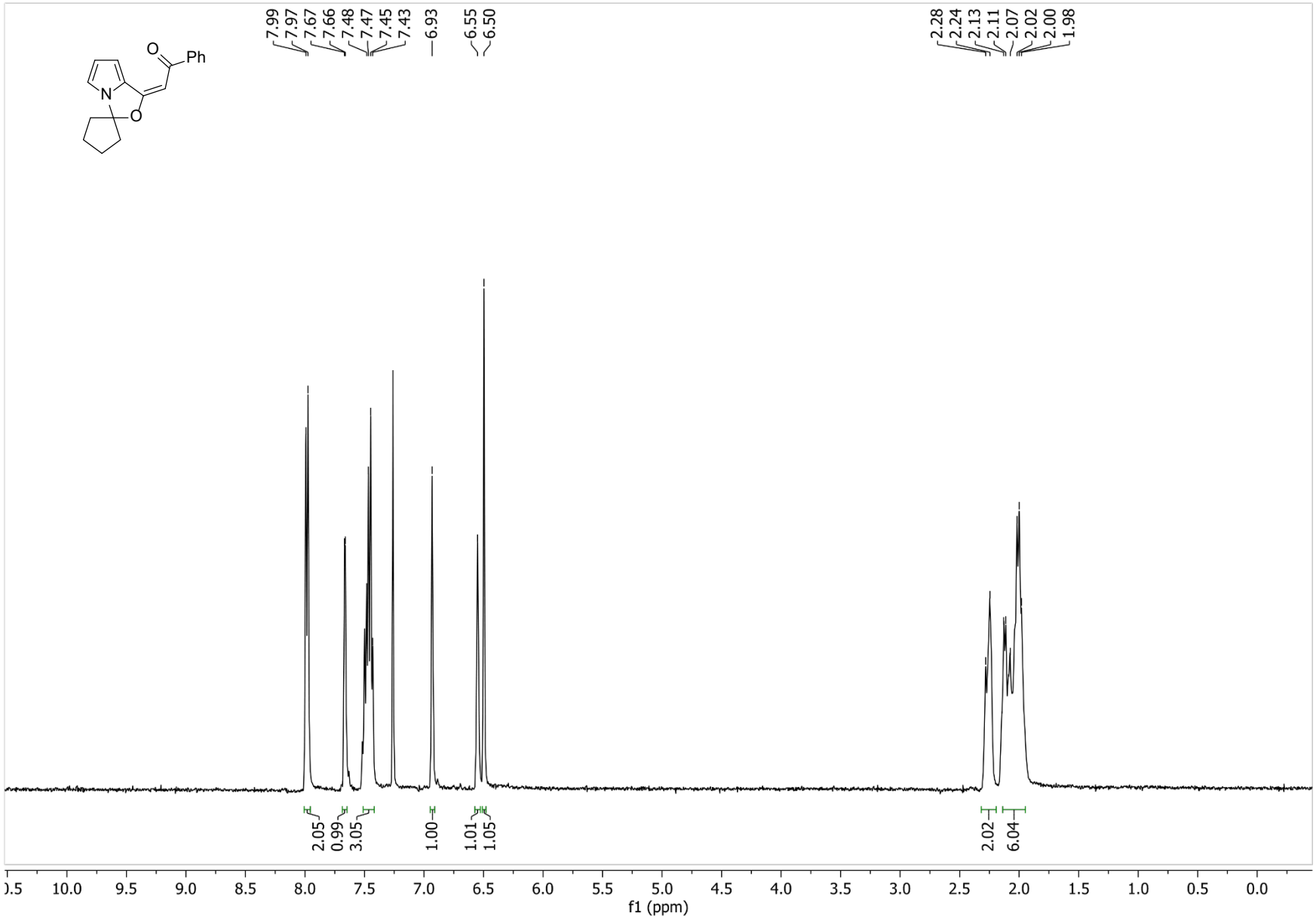
^{1}H NMR spectrum of (*E*)-2-(3,3-dibutyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ab**) in $CDCl_3$



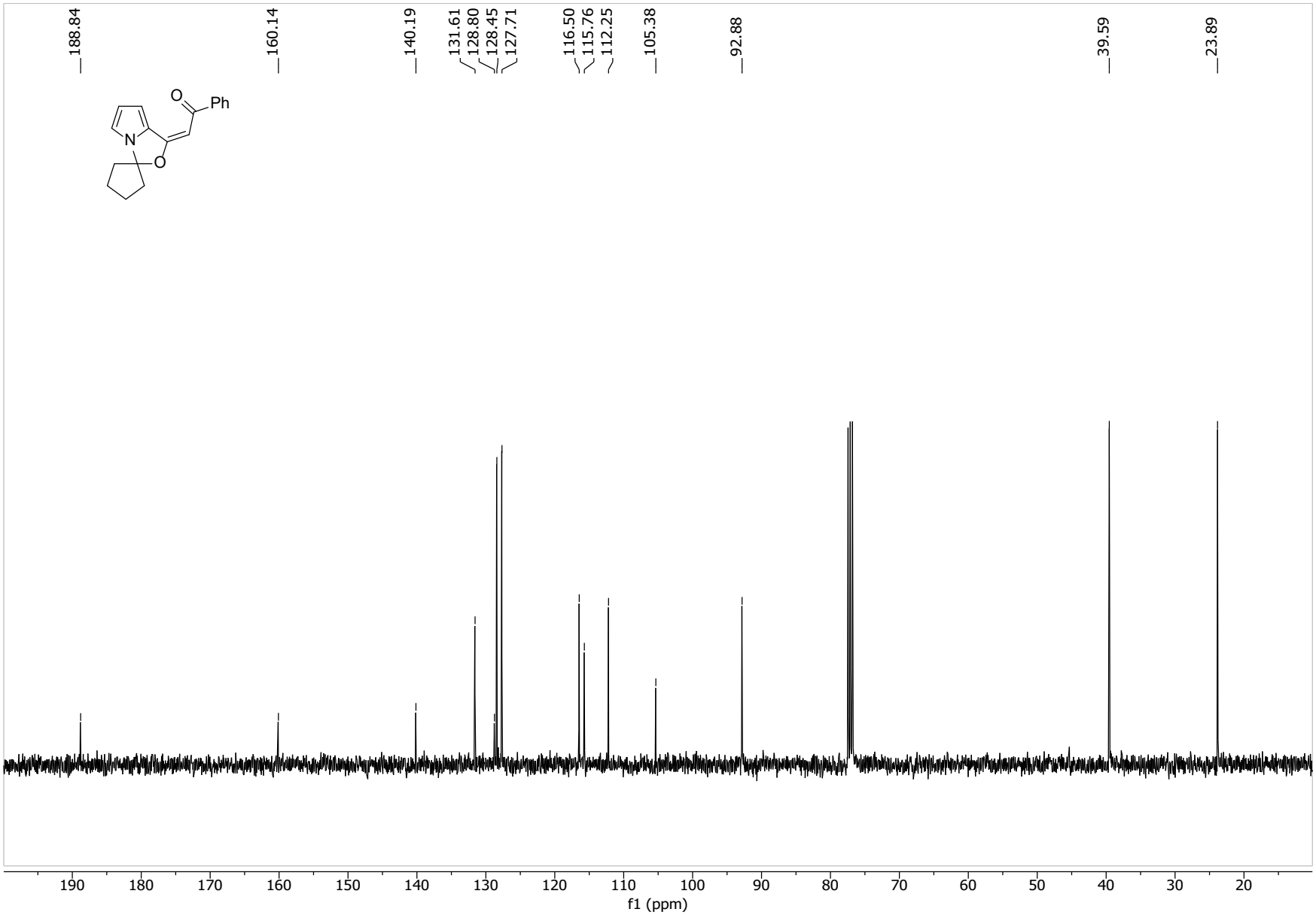
^{13}C NMR spectrum of (*E*)-2-(3,3-dibutyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ab**) in $CDCl_3$



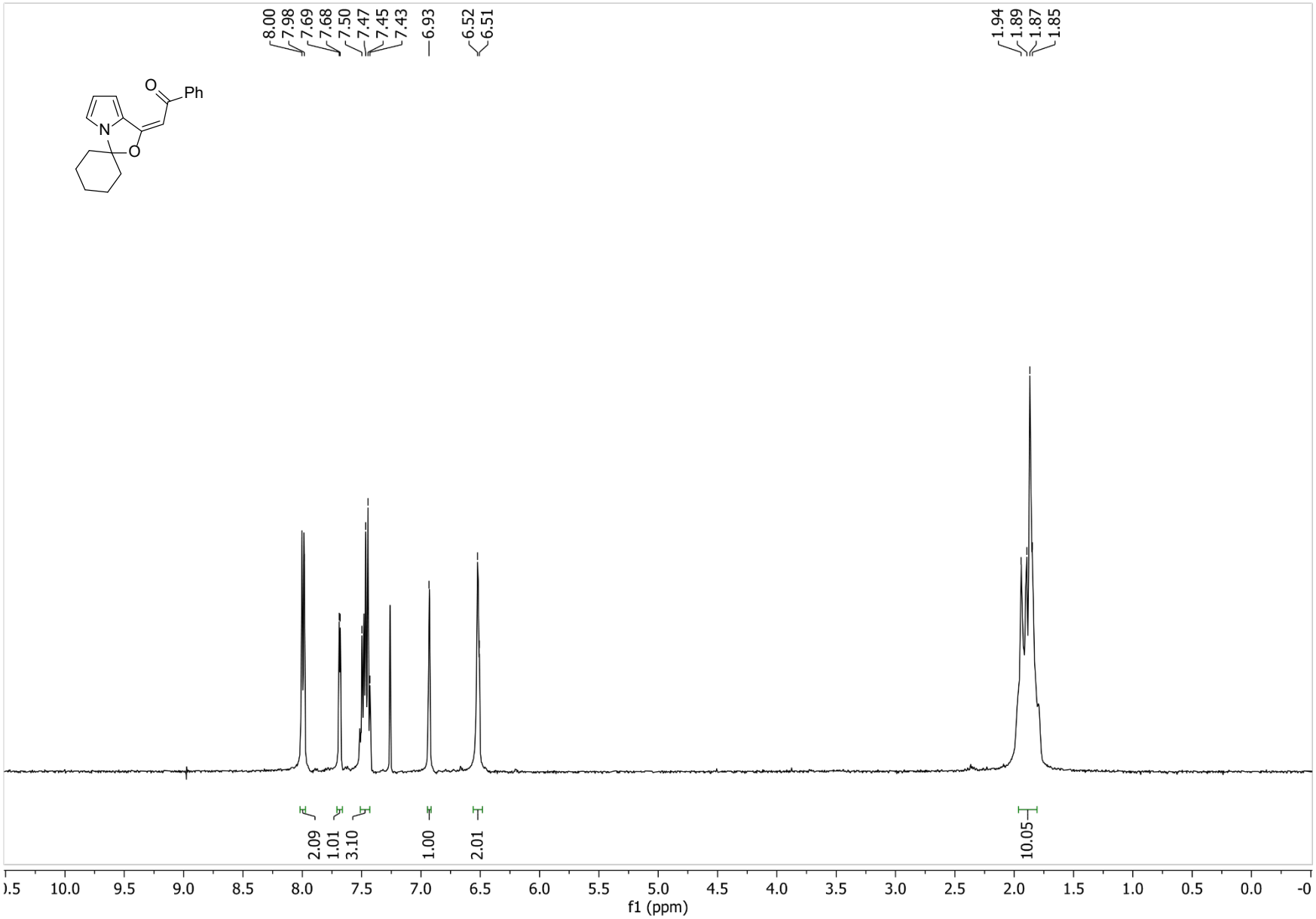
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(1'*H*-spiro[cyclopentane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)ethan-1-one (**3ac**) in $CDCl_3$



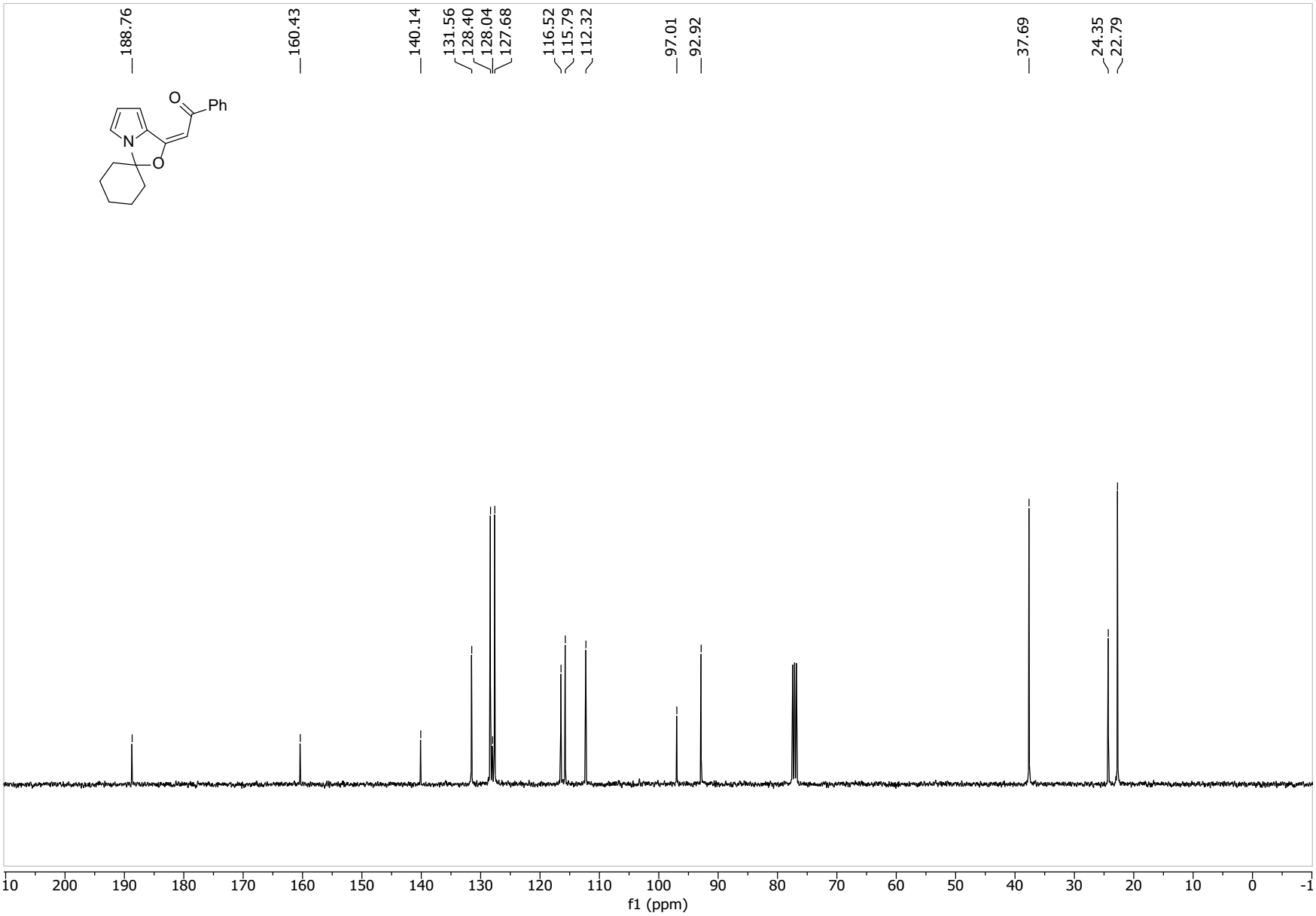
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(1'*H*-spiro[cyclopentane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)ethan-1-one (**3ac**) in $CDCl_3$



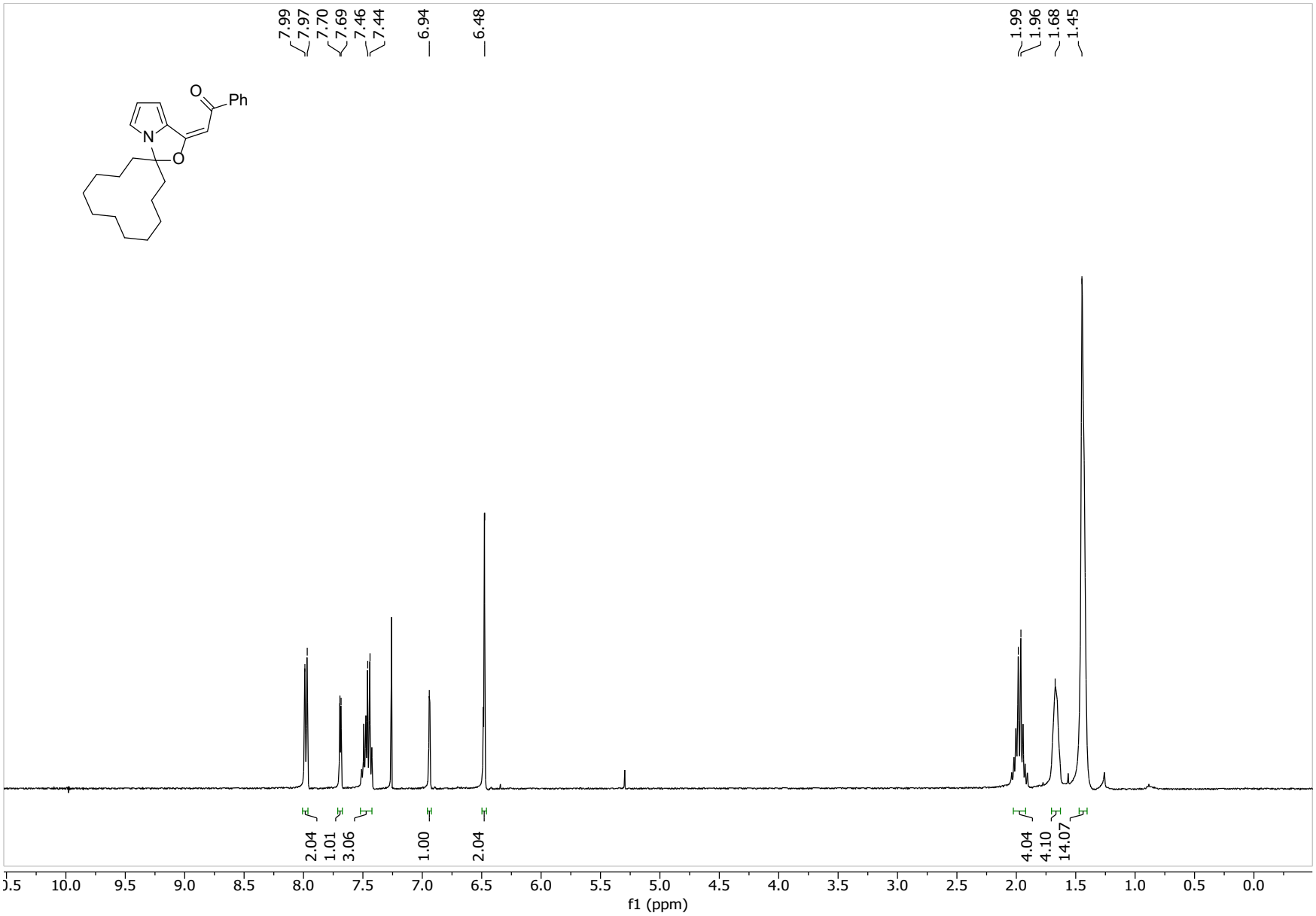
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(1'*H*-spiro[cyclohexane-1,3'-pyrrolo[1,2-c]oxazol]-1'-ylidene)ethan-1-one (**3ad**) in $CDCl_3$



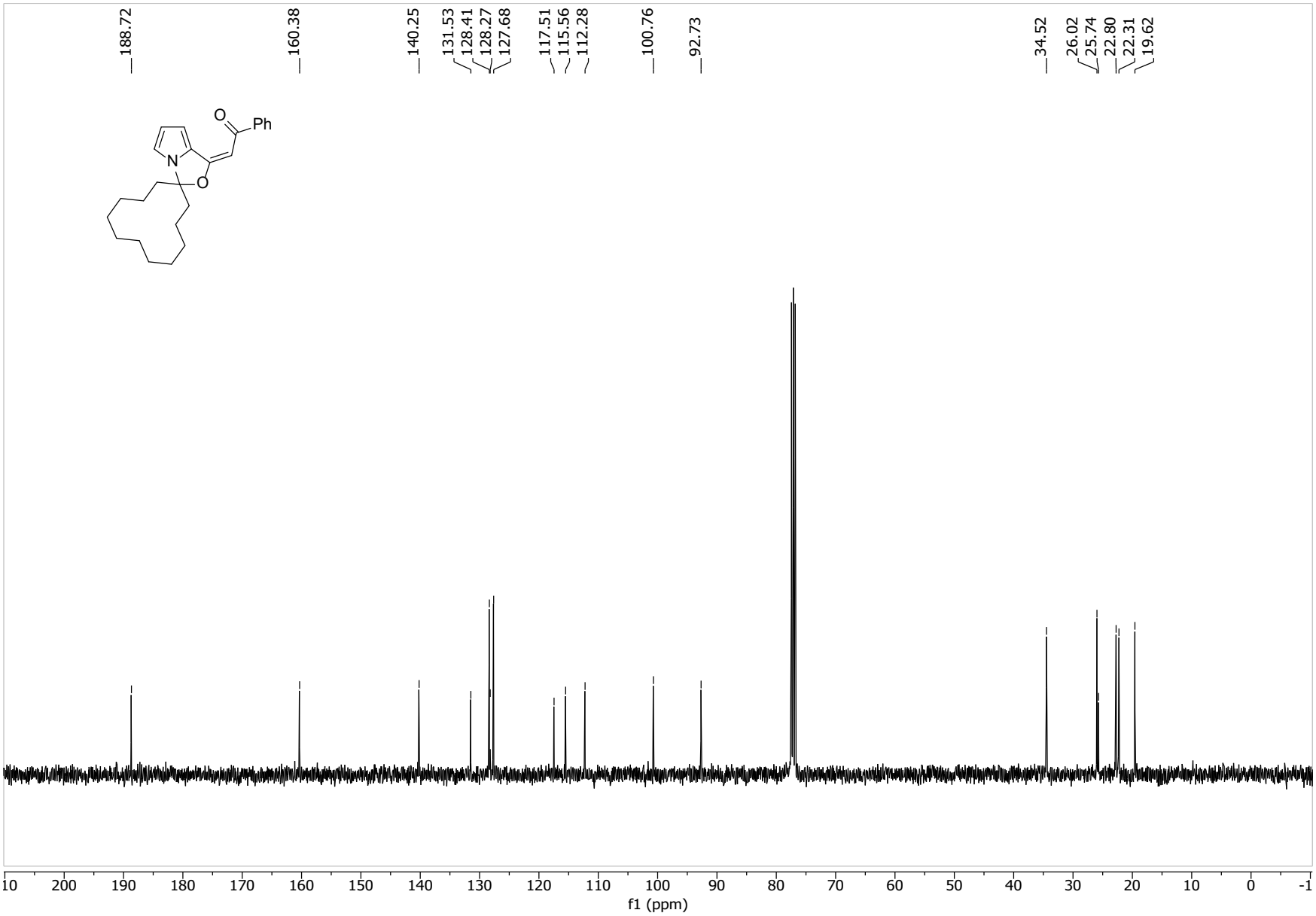
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(1'*H*-spiro[cyclohexane-1,3'-pyrrolo[1,2-c]oxazol]-1'-ylidene)ethan-1-one (**3ad**) in $CDCl_3$



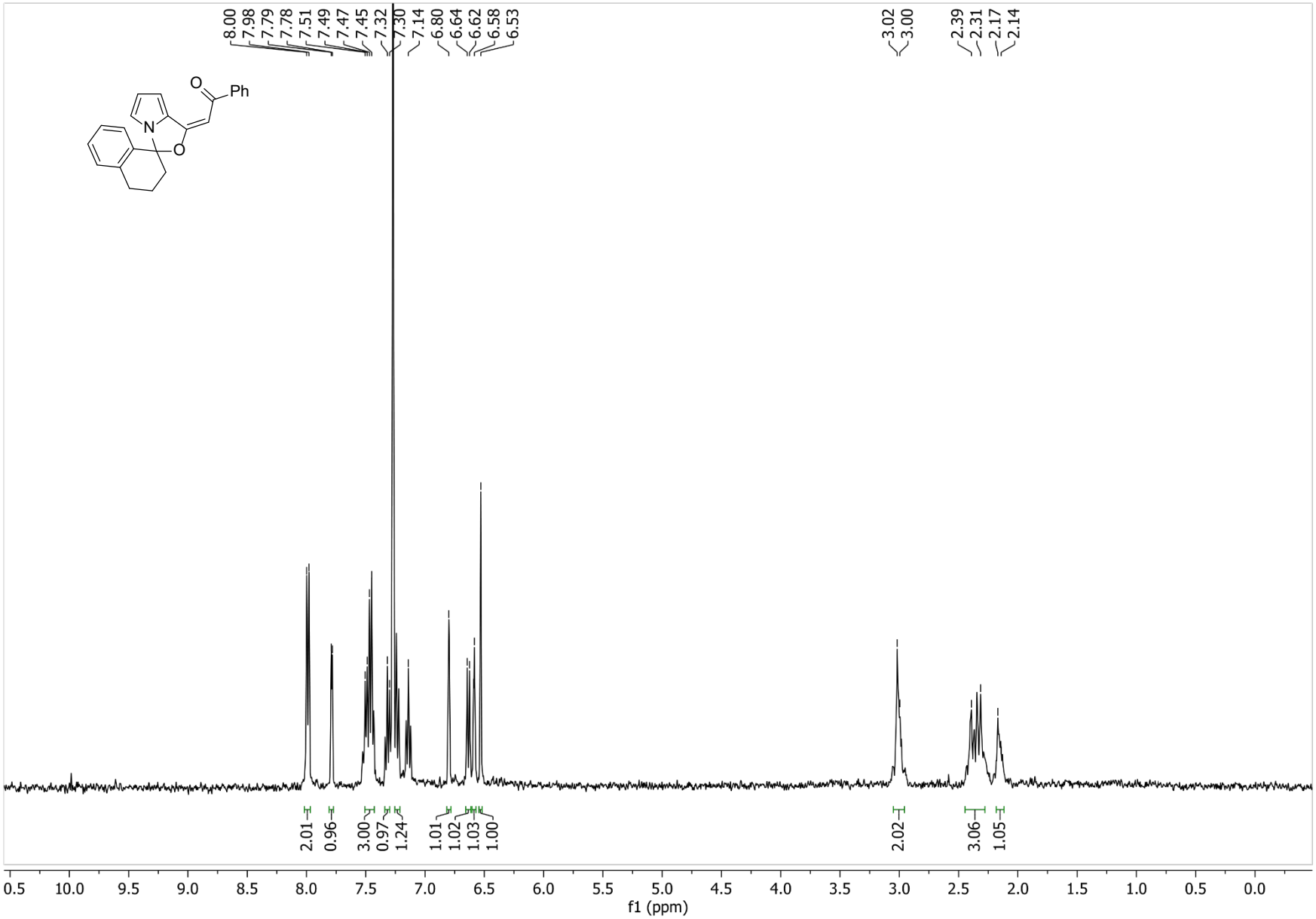
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(1'*H*-spiro[cyclododecane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)ethan-1-one (**3ae**) in $CDCl_3$



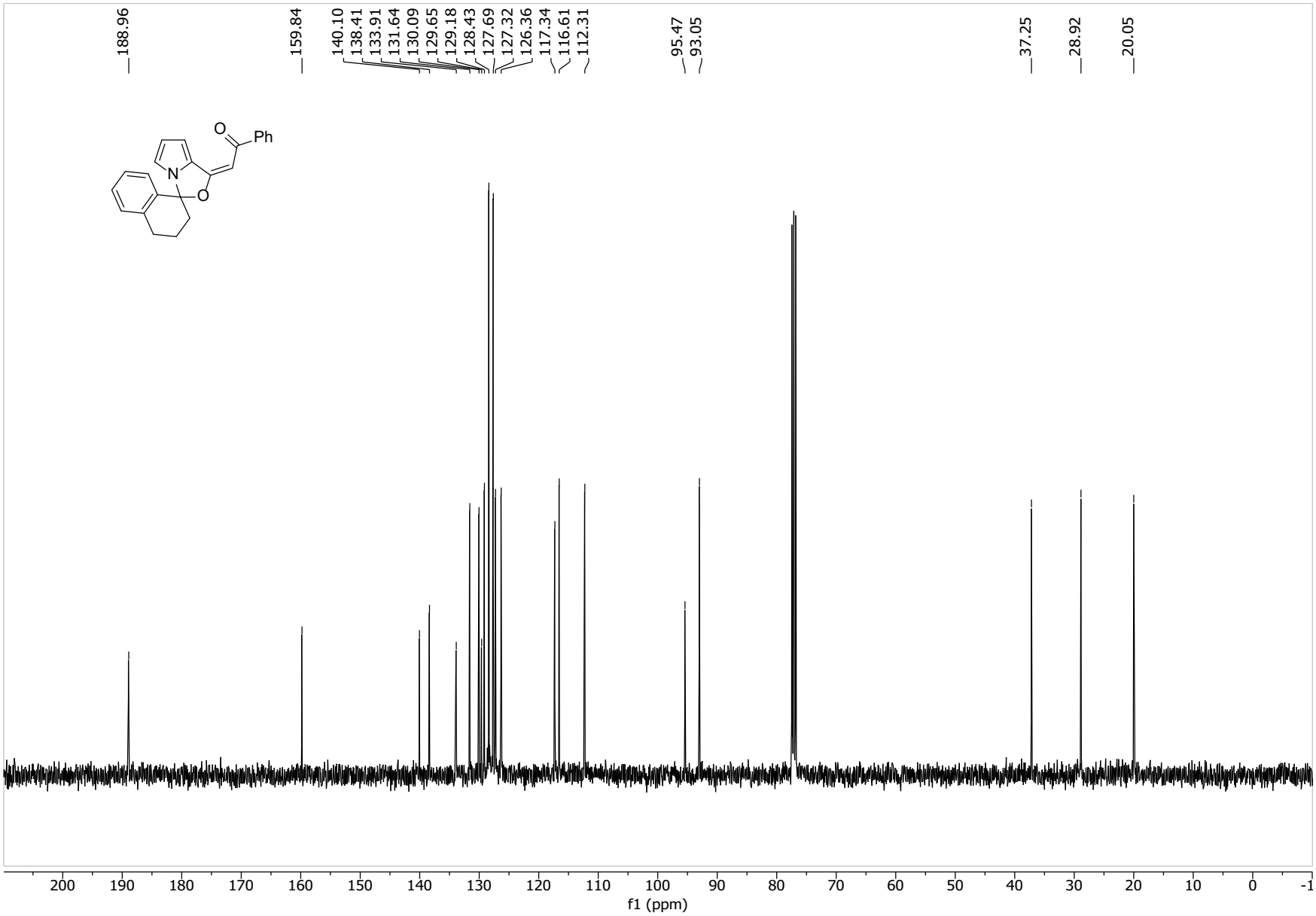
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(1'*H*-spiro[cyclododecane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)ethan-1-one (**3ae**) in $CDCl_3$



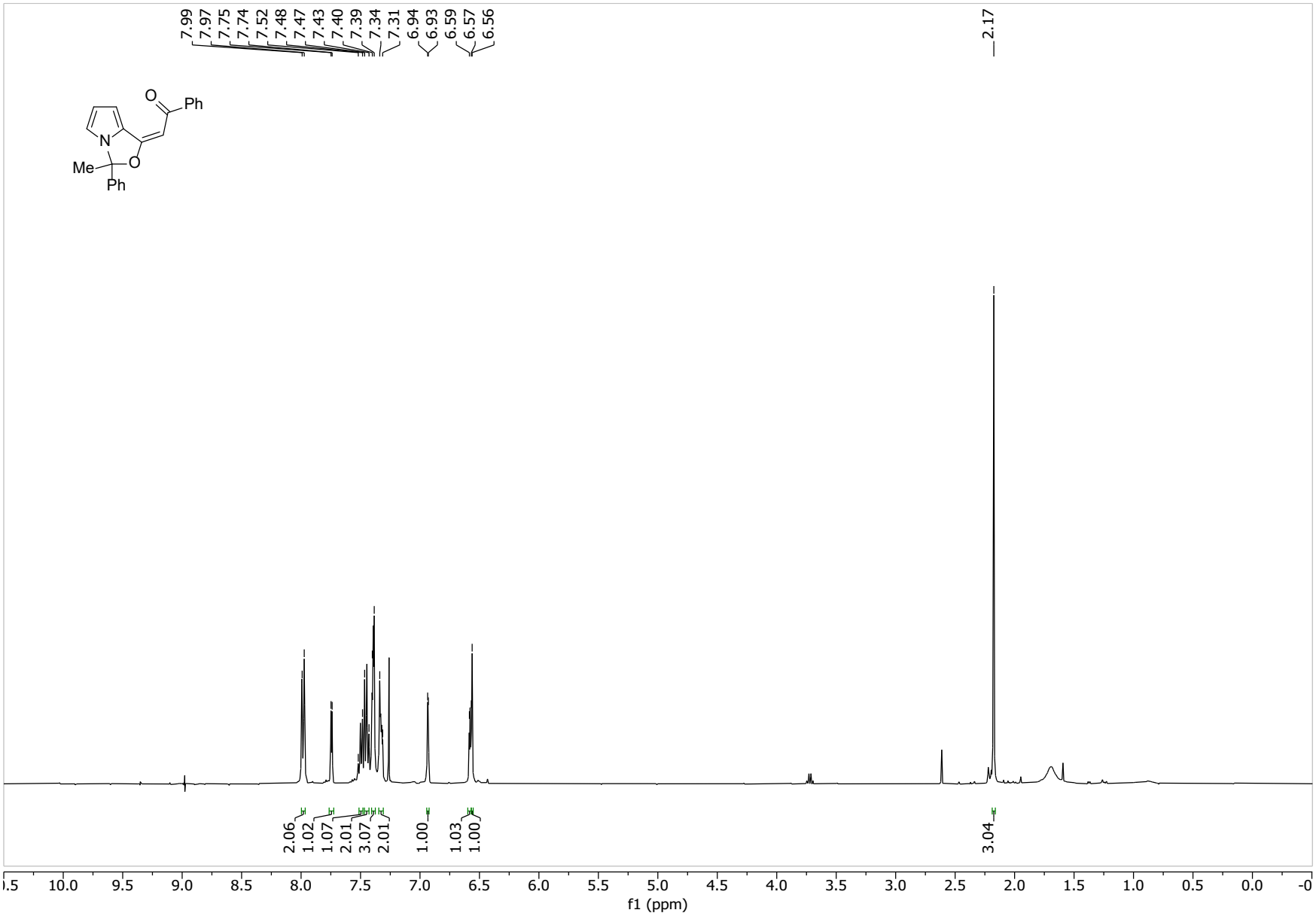
^{1}H NMR spectrum of (*E*)-2-(3,4-dihydro-1'*H*,2*H*-spiro[naphthalene-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-phenylethan-1-one (**3af**) in $CDCl_3$



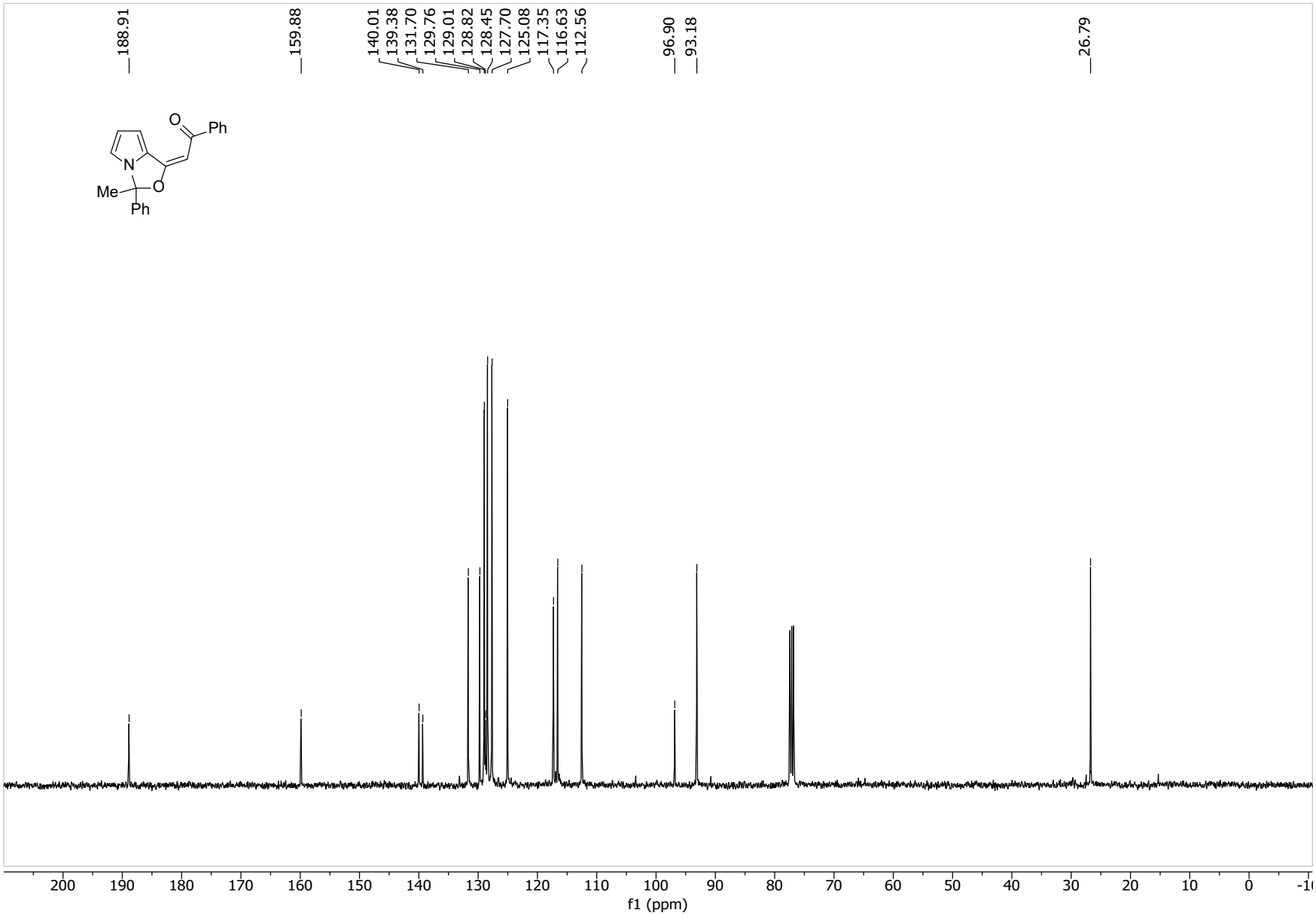
^{13}C NMR spectrum of (*E*)-2-(3,4-dihydro-1'*H*,2*H*-spiro[naphthalene-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-phenylethan-1-one (**3af**) in $CDCl_3$



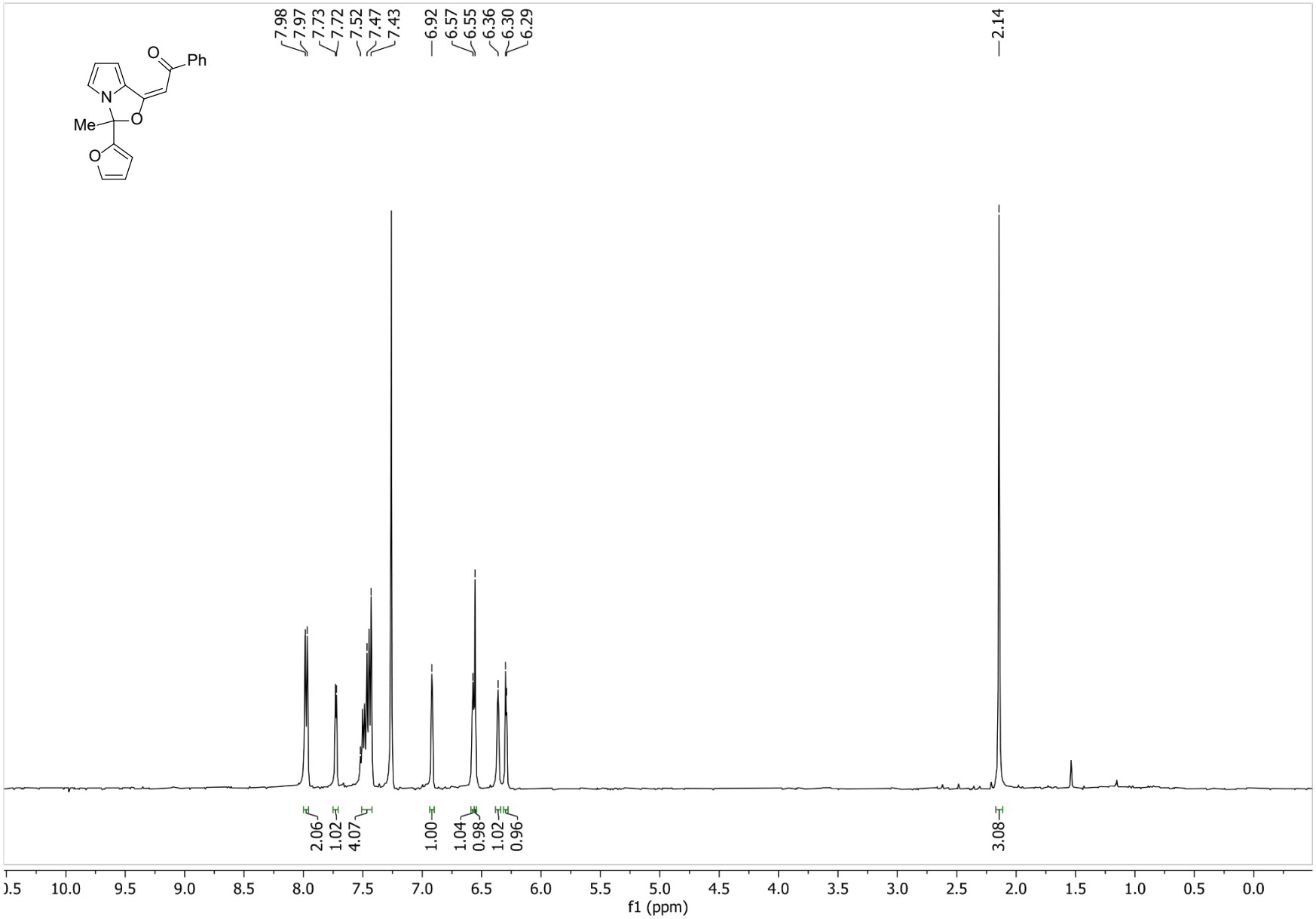
^{1}H NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ag**) in $CDCl_3$



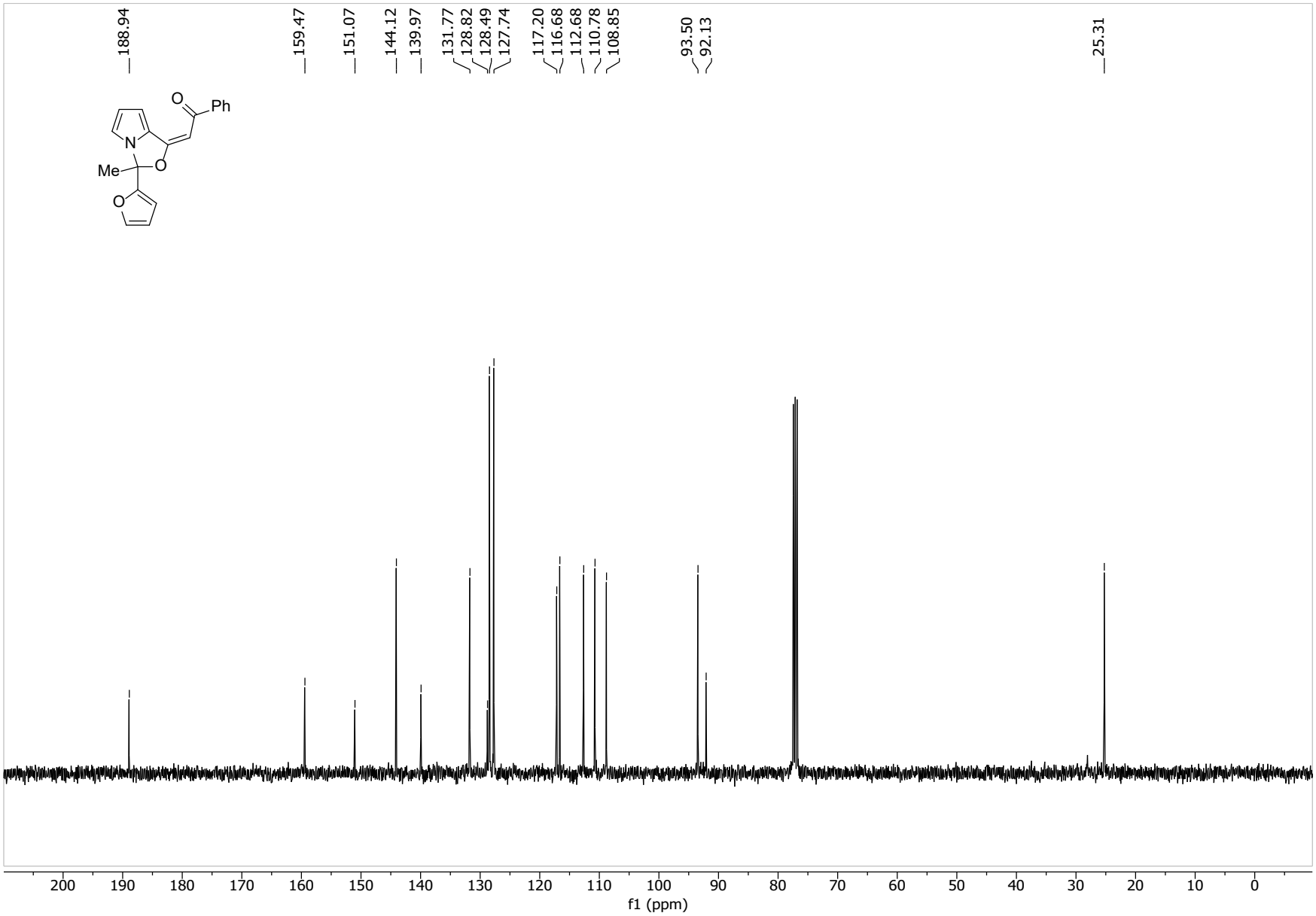
^{13}C NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ag**) in $CDCl_3$



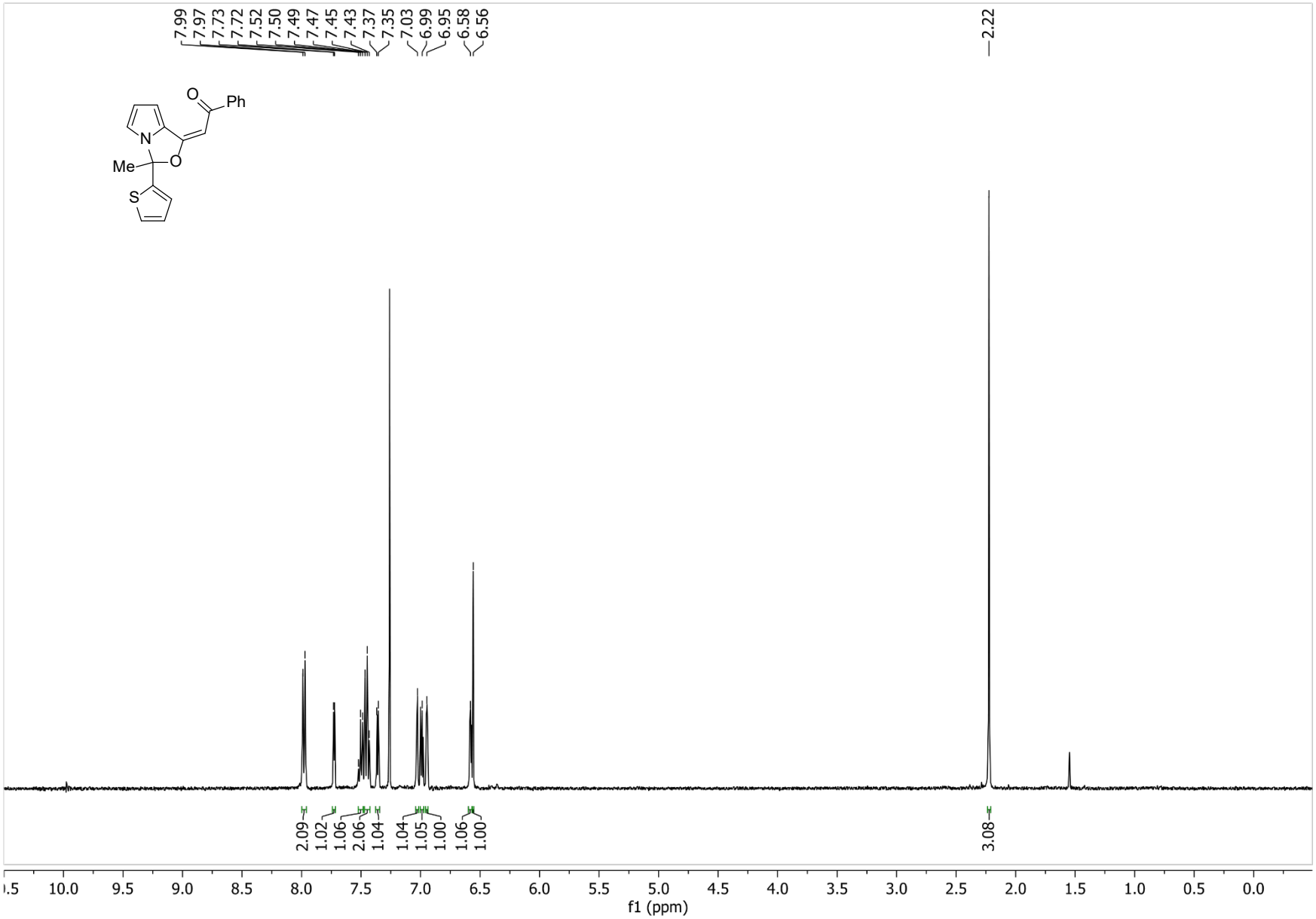
^{1}H NMR spectrum of (*E*)-2-(3-(furan-2-yl)-3-methyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ah**) in $CDCl_3$



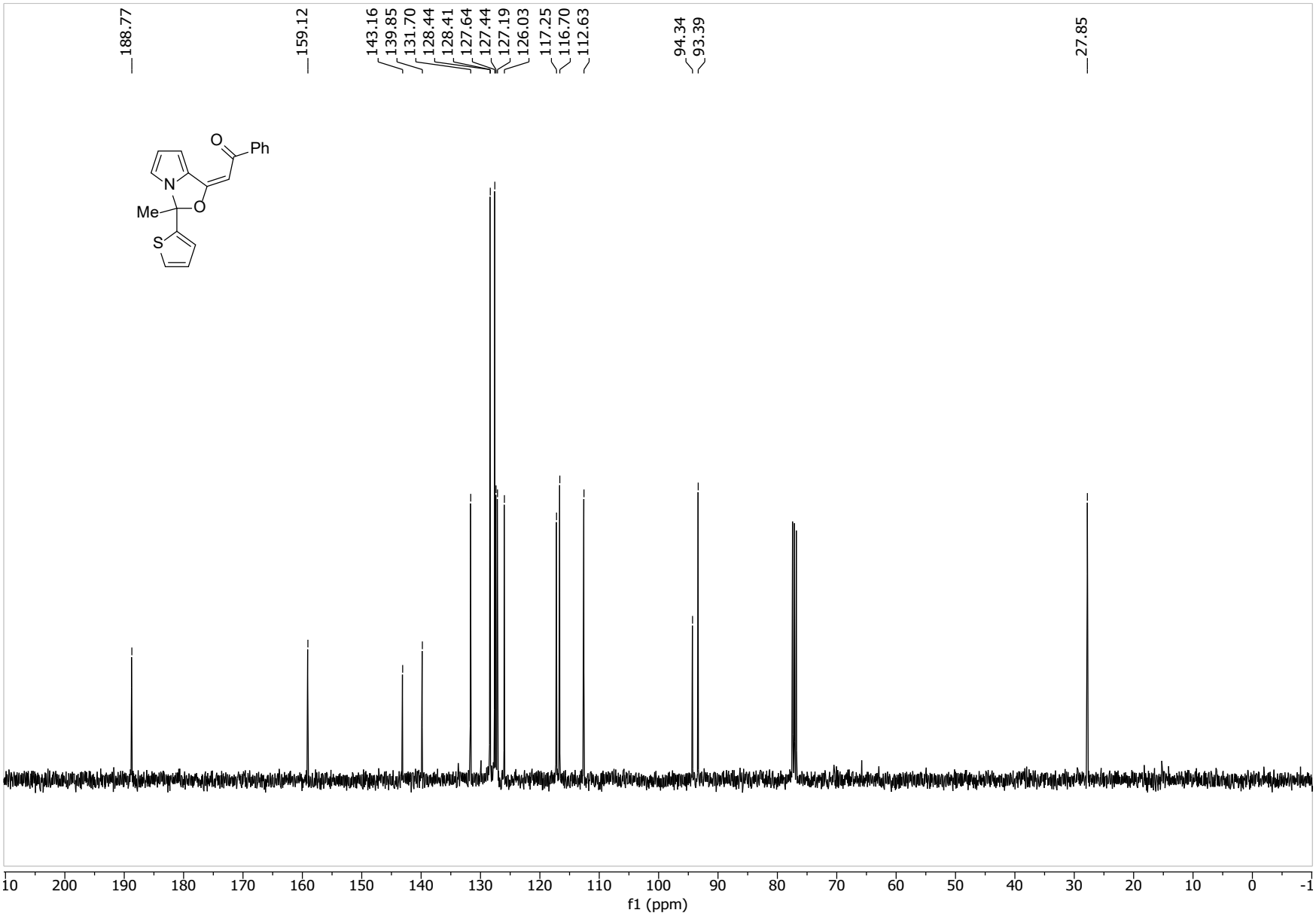
^{13}C NMR spectrum of (*E*)-2-(3-(furan-2-yl)-3-methyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ah**) in $CDCl_3$



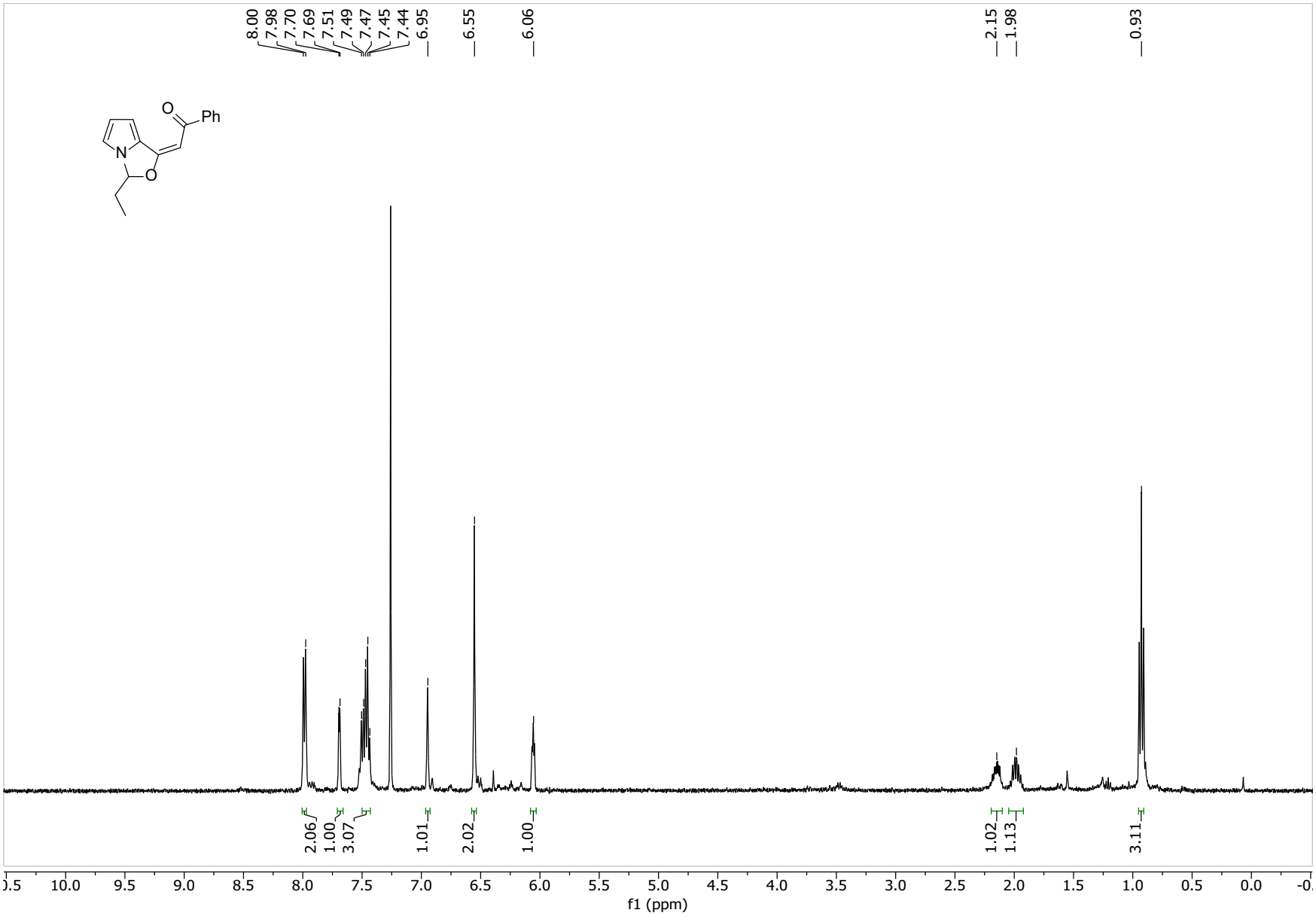
^{1}H NMR spectrum of (*E*)-2-(3-methyl-3-(thiophen-2-yl)-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ai**) in $CDCl_3$



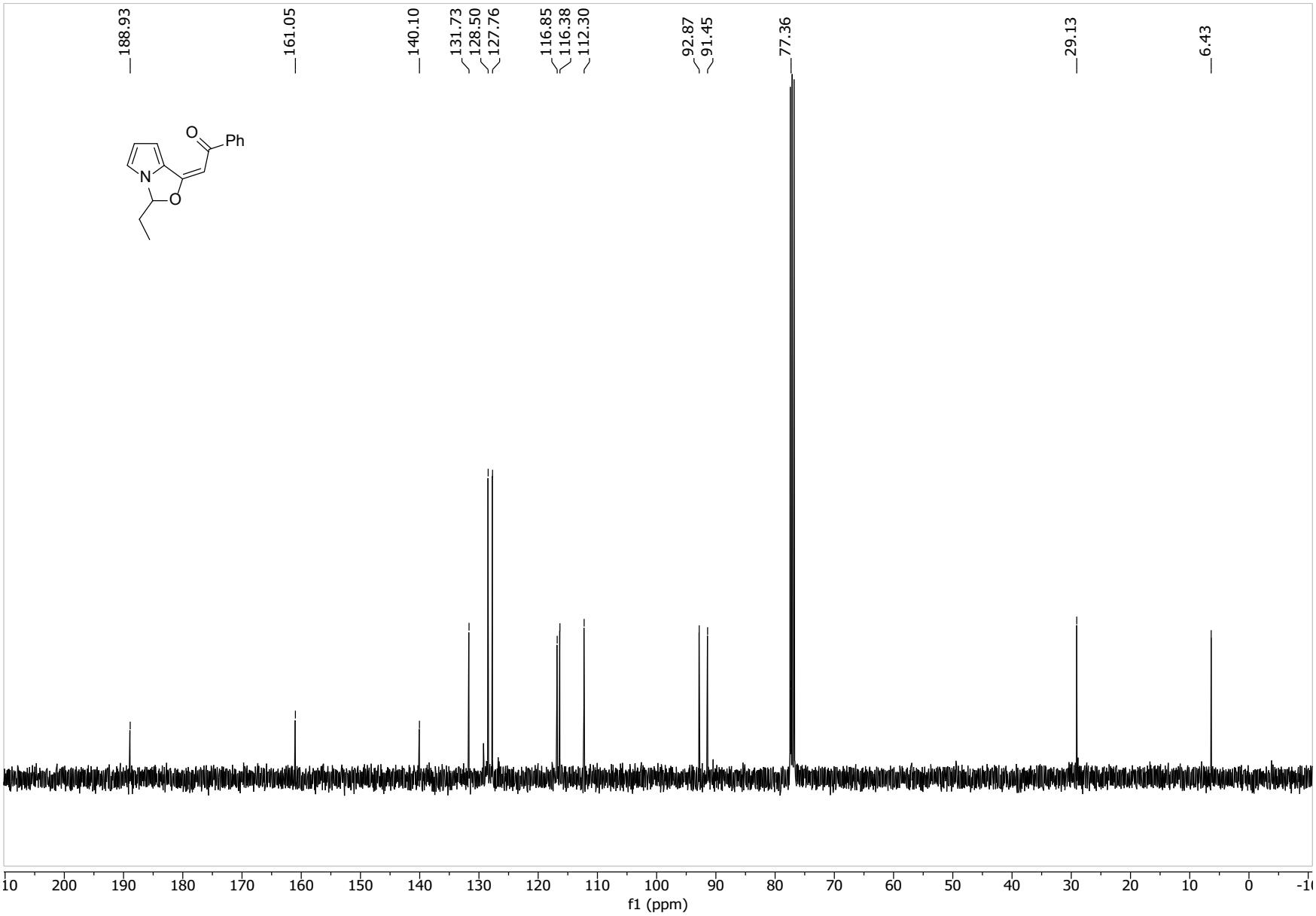
^{12}C NMR spectrum of (*E*)-2-(3-methyl-3-(thiophen-2-yl)-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ai**) in $CDCl_3$



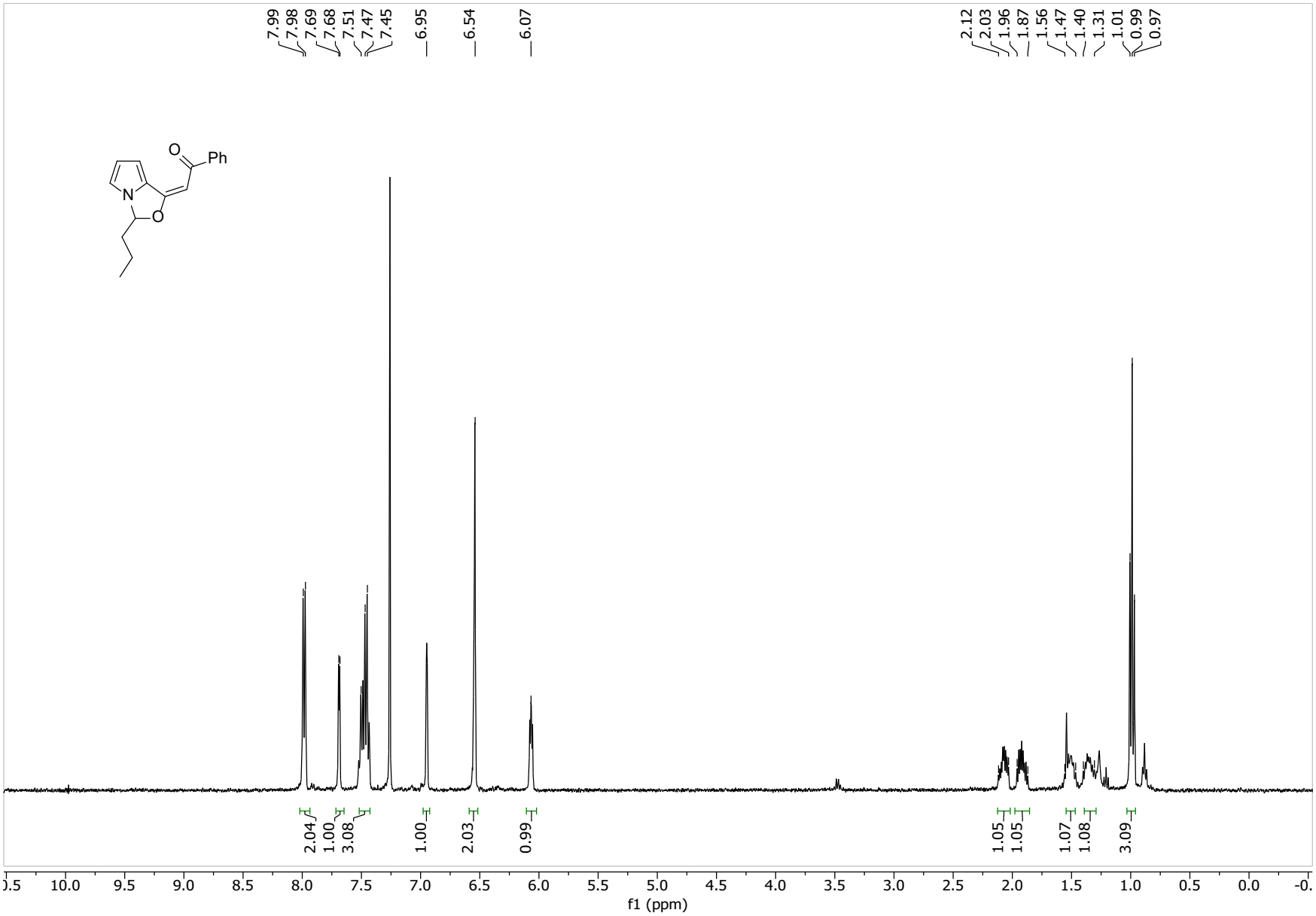
^{1}H NMR spectrum of (*E*)-2-(3-ethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ak**) in $CDCl_3$



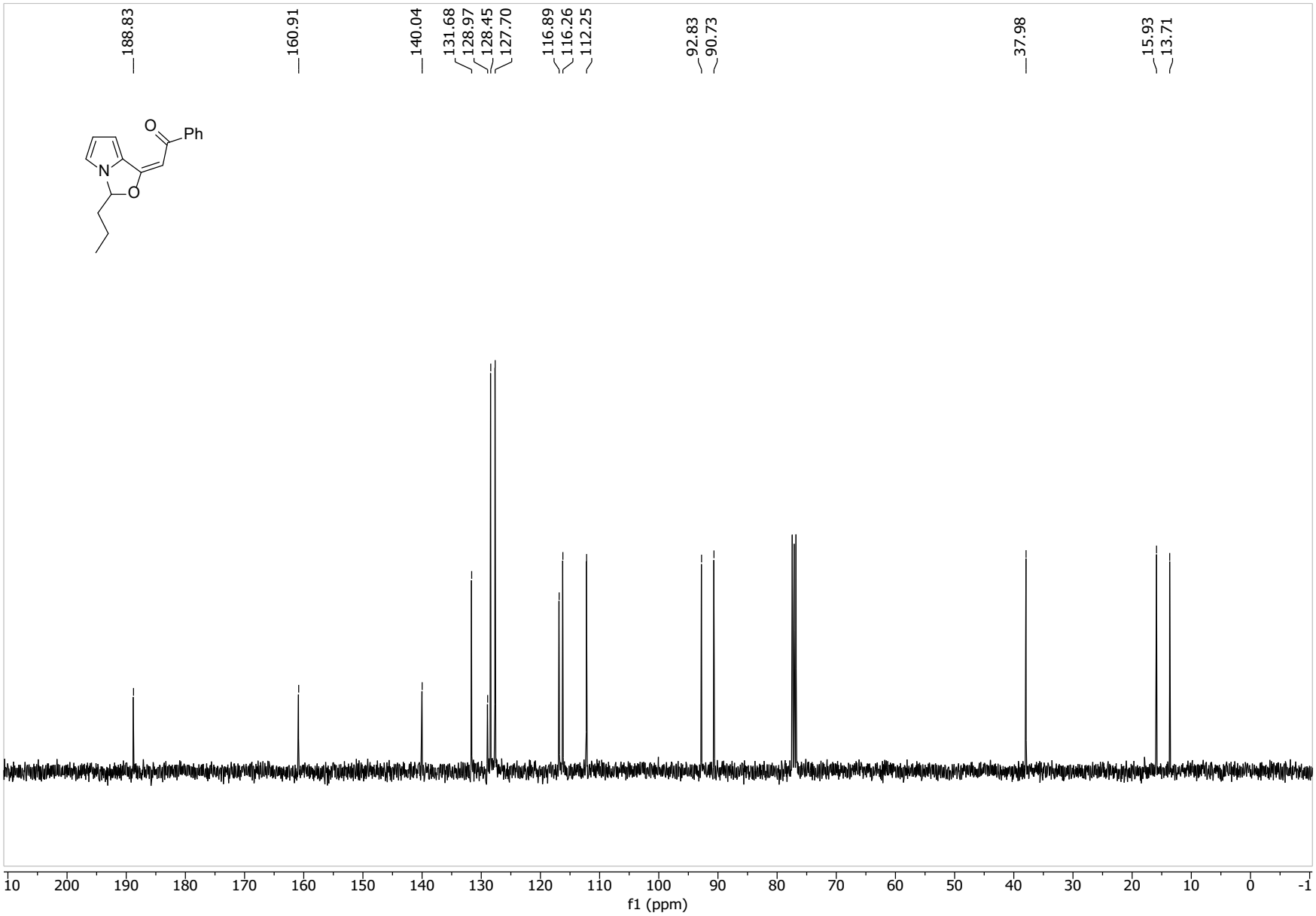
^{13}C NMR spectrum of (*E*)-2-(3-ethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ak**) in $CDCl_3$



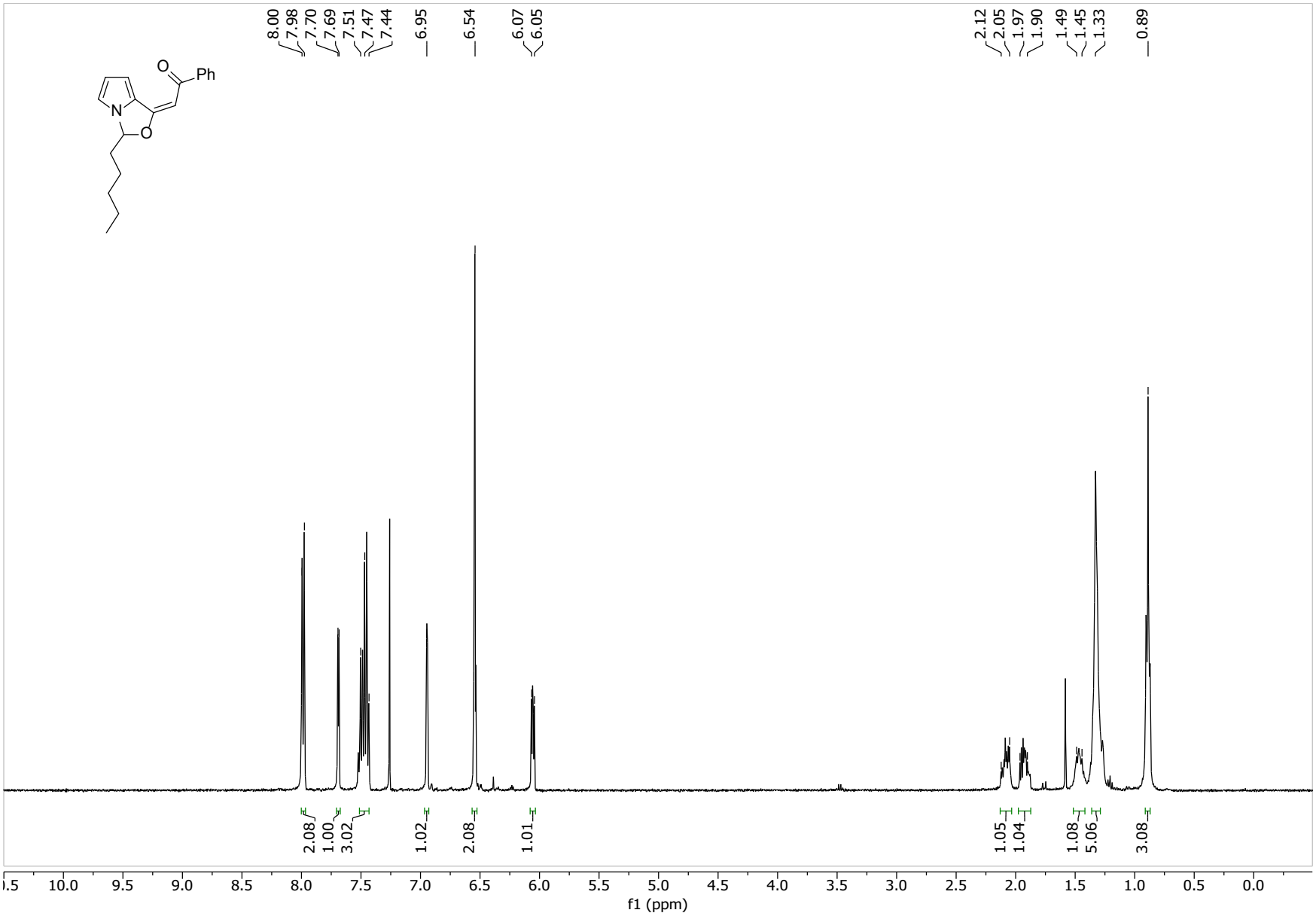
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(3-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3al**) in $CDCl_3$



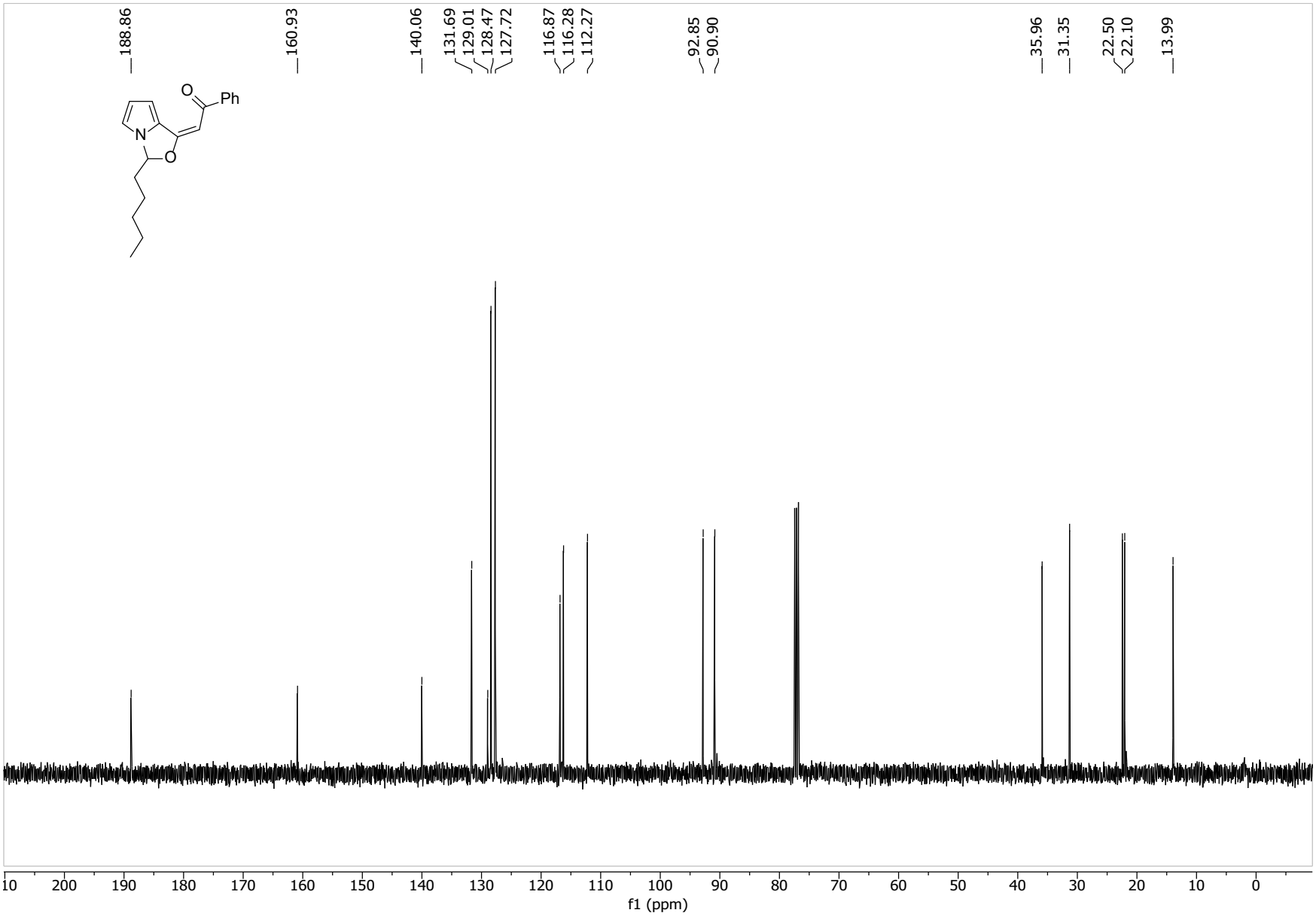
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(3-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3al**) in $CDCl_3$



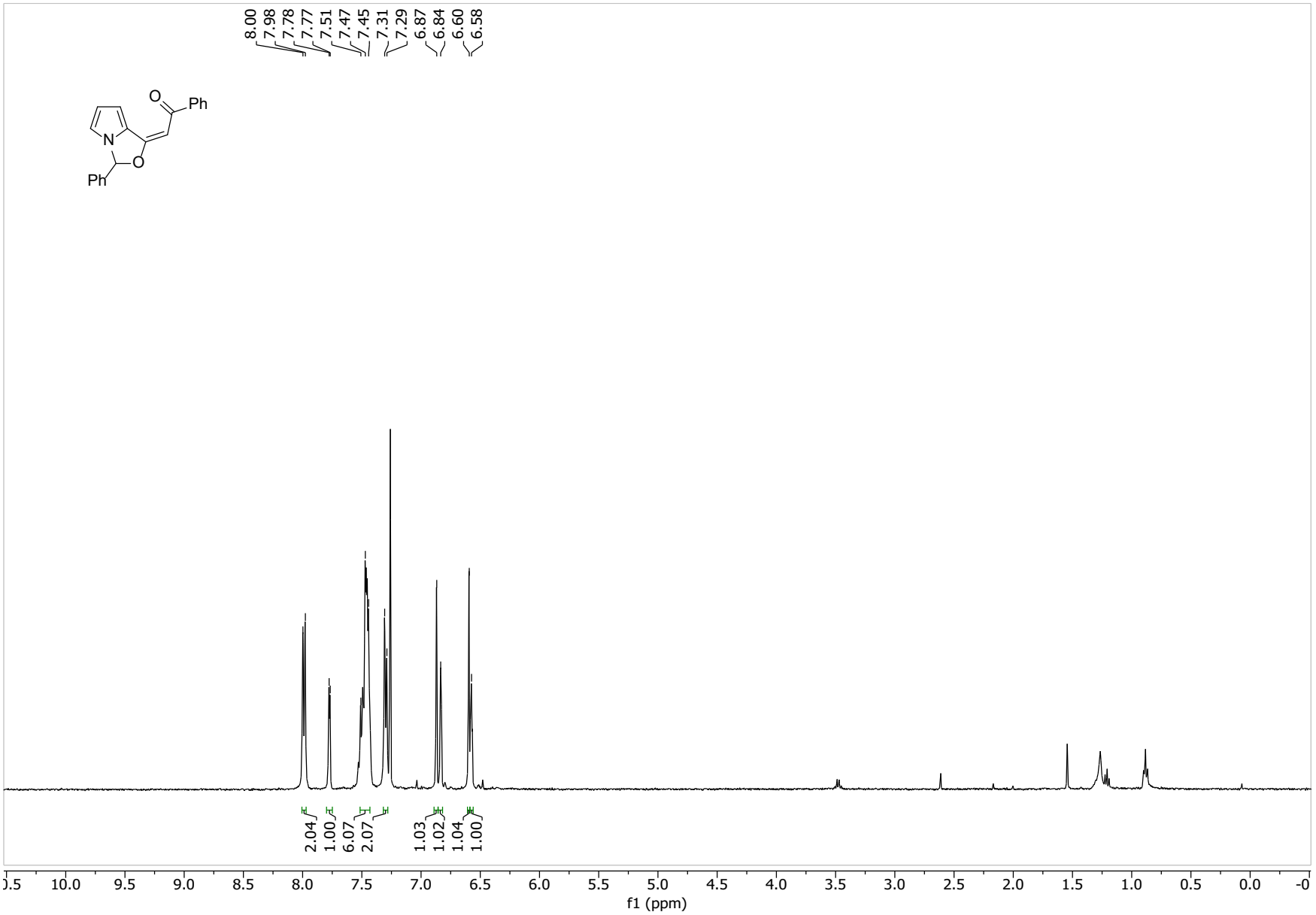
^{1}H NMR spectrum of (*E*)- 2-(3-pentyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3am**) in $CDCl_3$



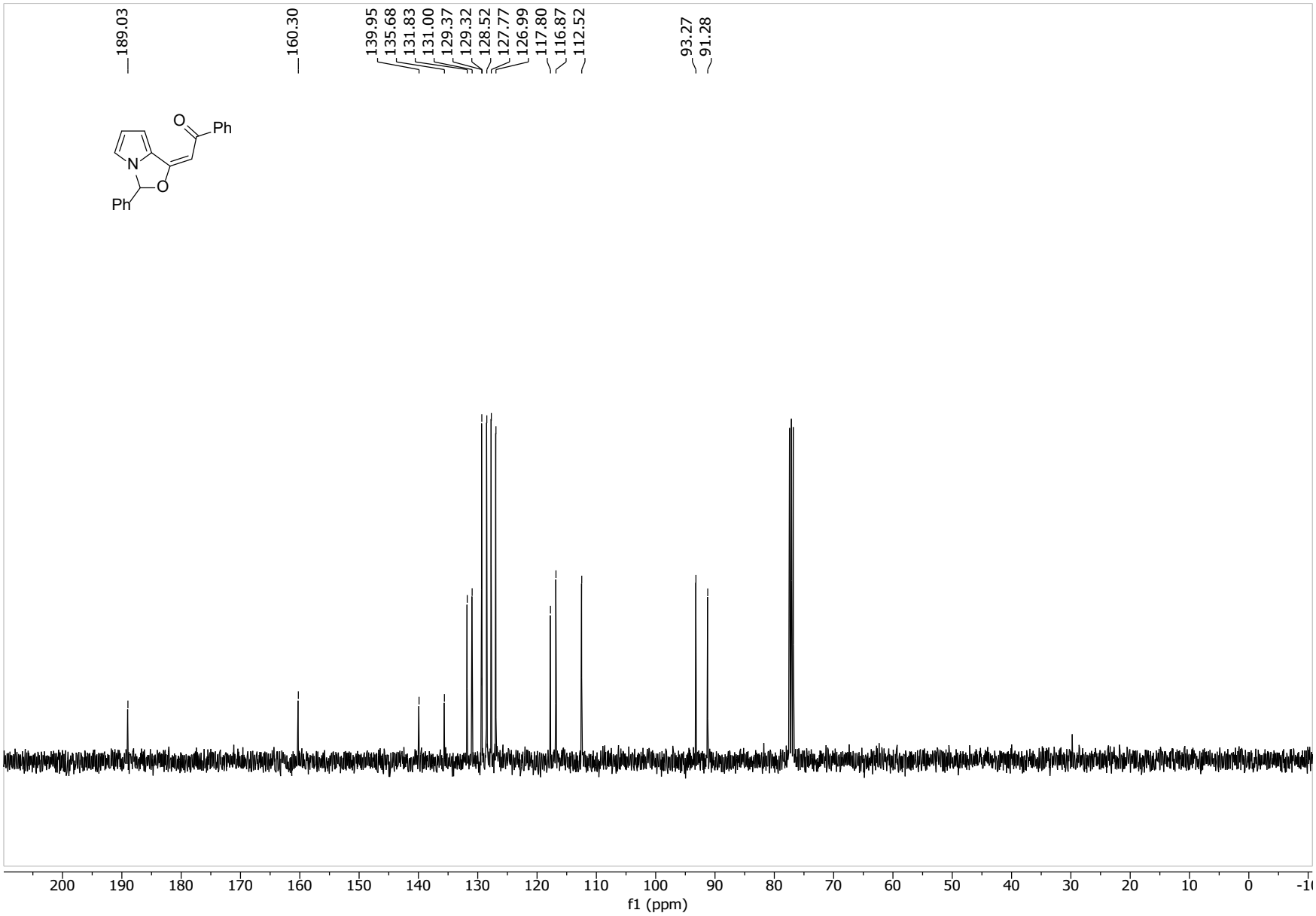
^{13}C NMR spectrum of (*E*)-2-(3-pentyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3am**) in $CDCl_3$



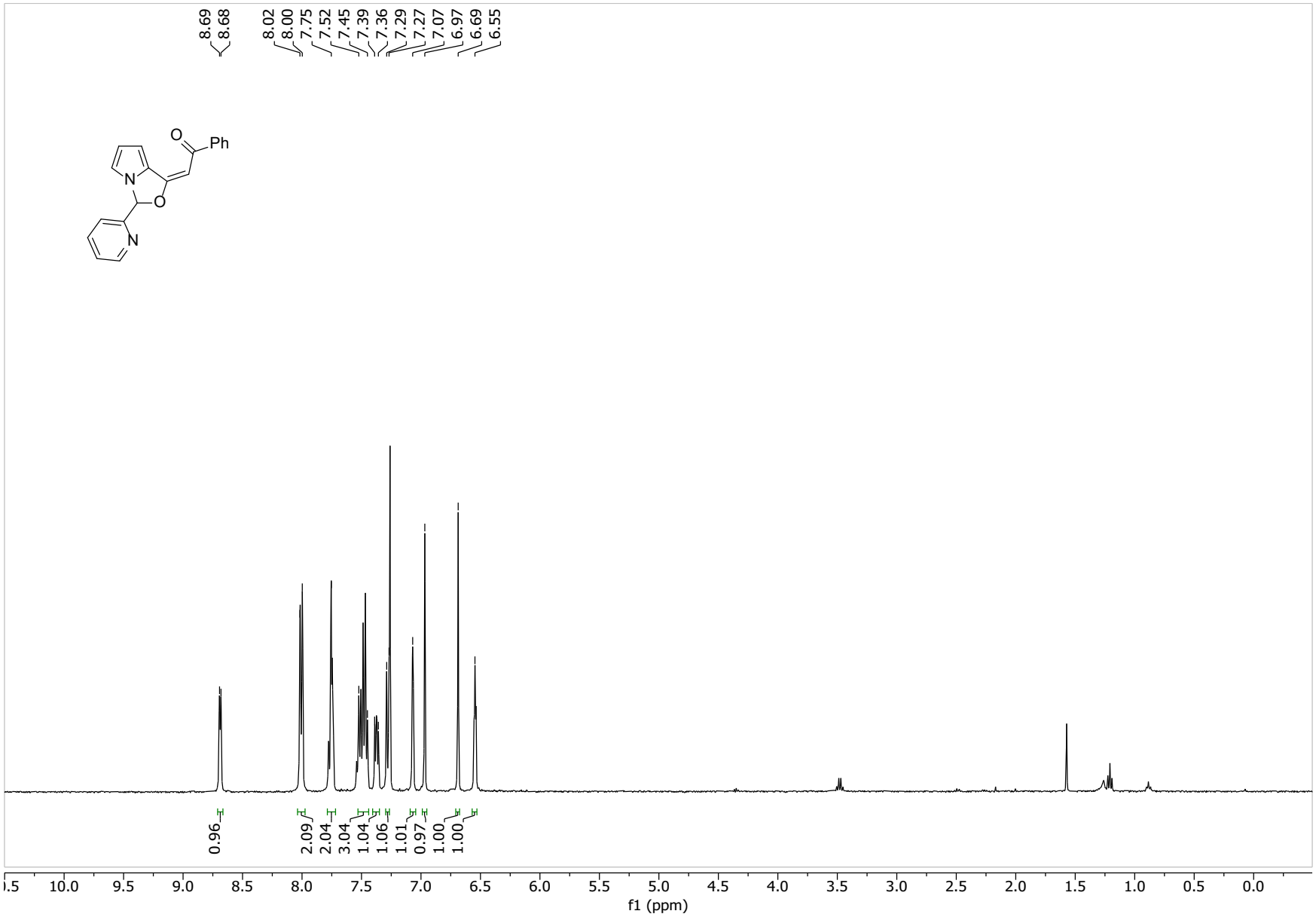
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3an**) in $CDCl_3$



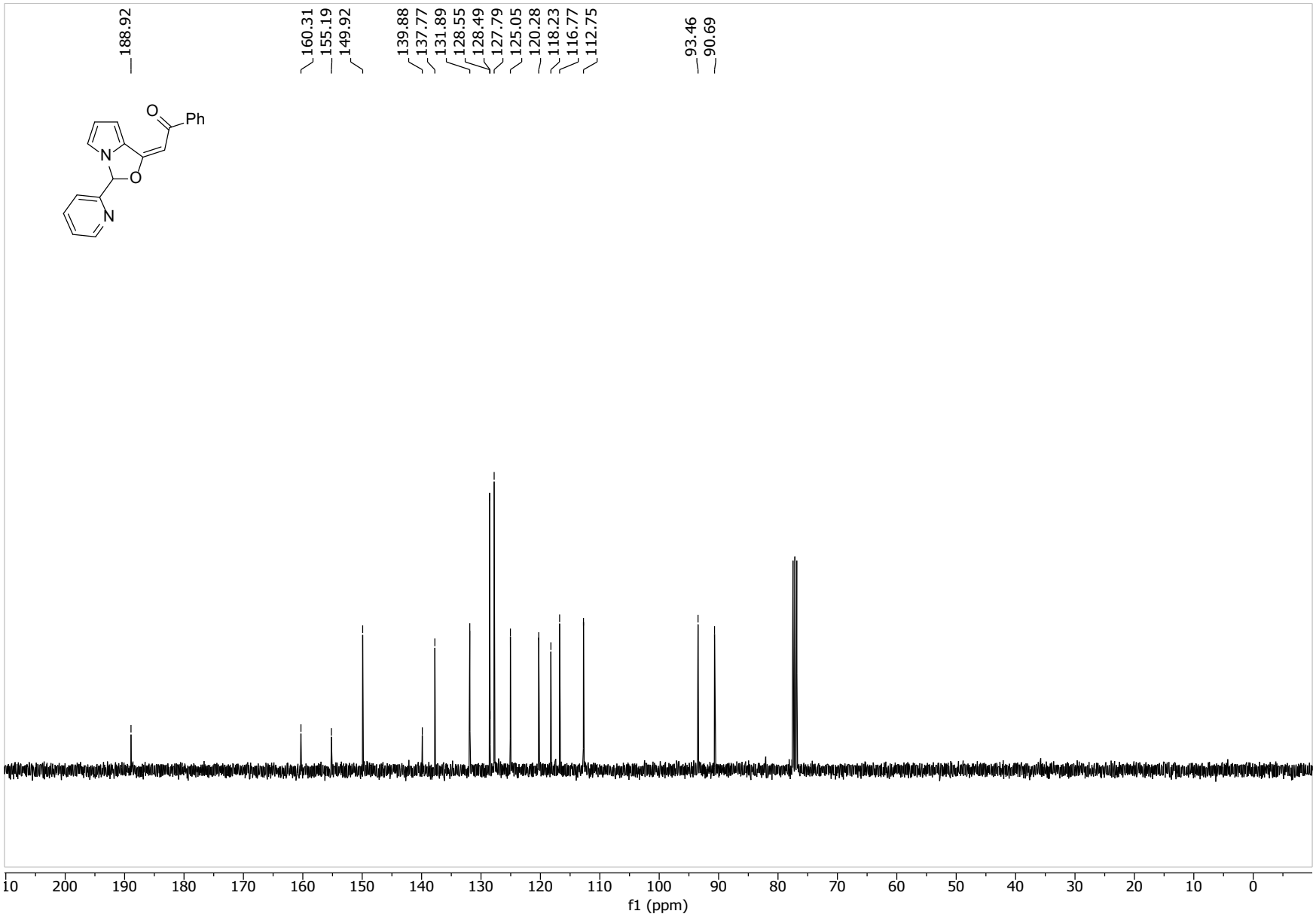
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3an**) in $CDCl_3$



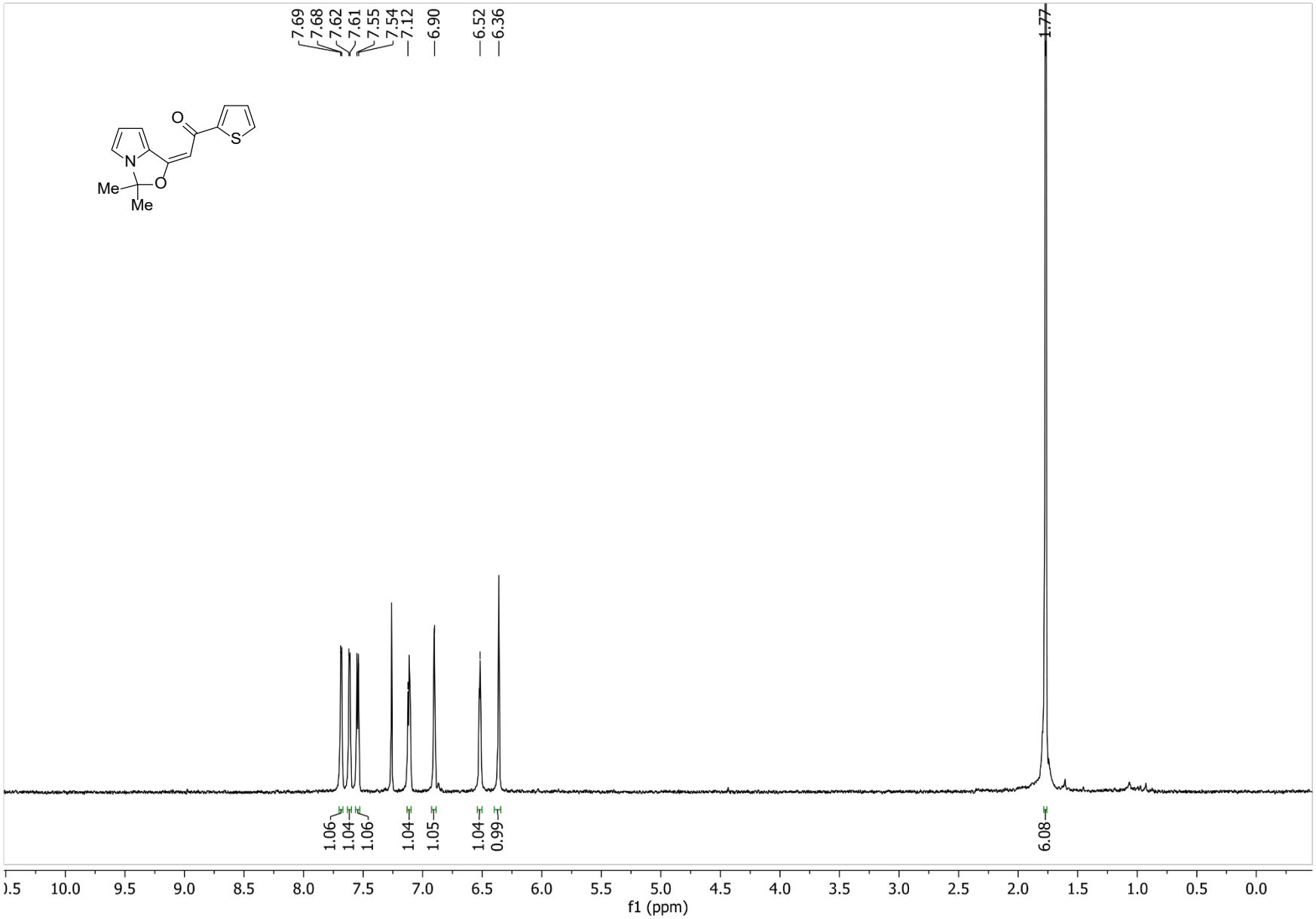
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(3-(pyridin-2-yl)-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3ao**) in $CDCl_3$



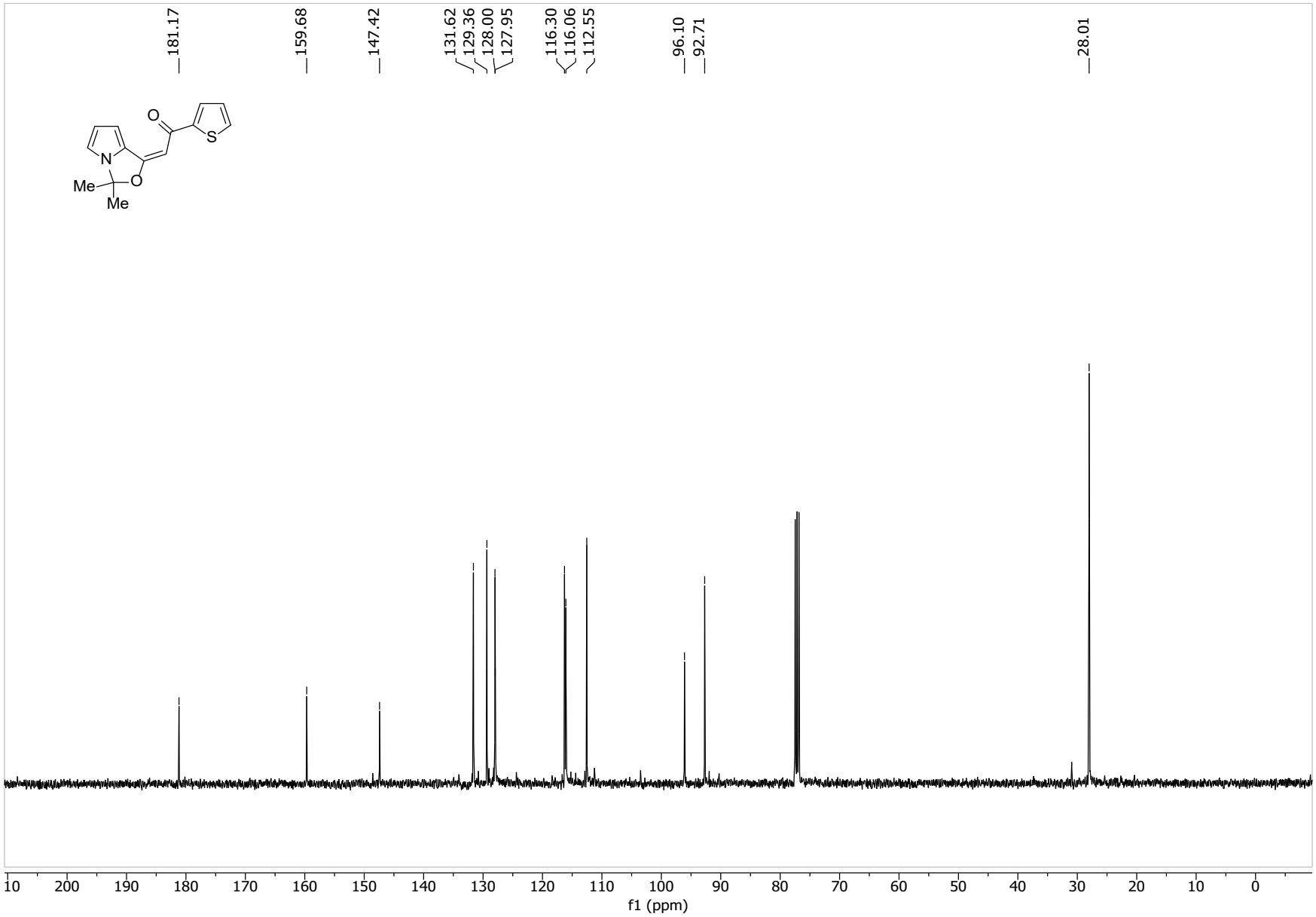
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(3-(pyridin-2-yl)-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3ao**) in $CDCl_3$



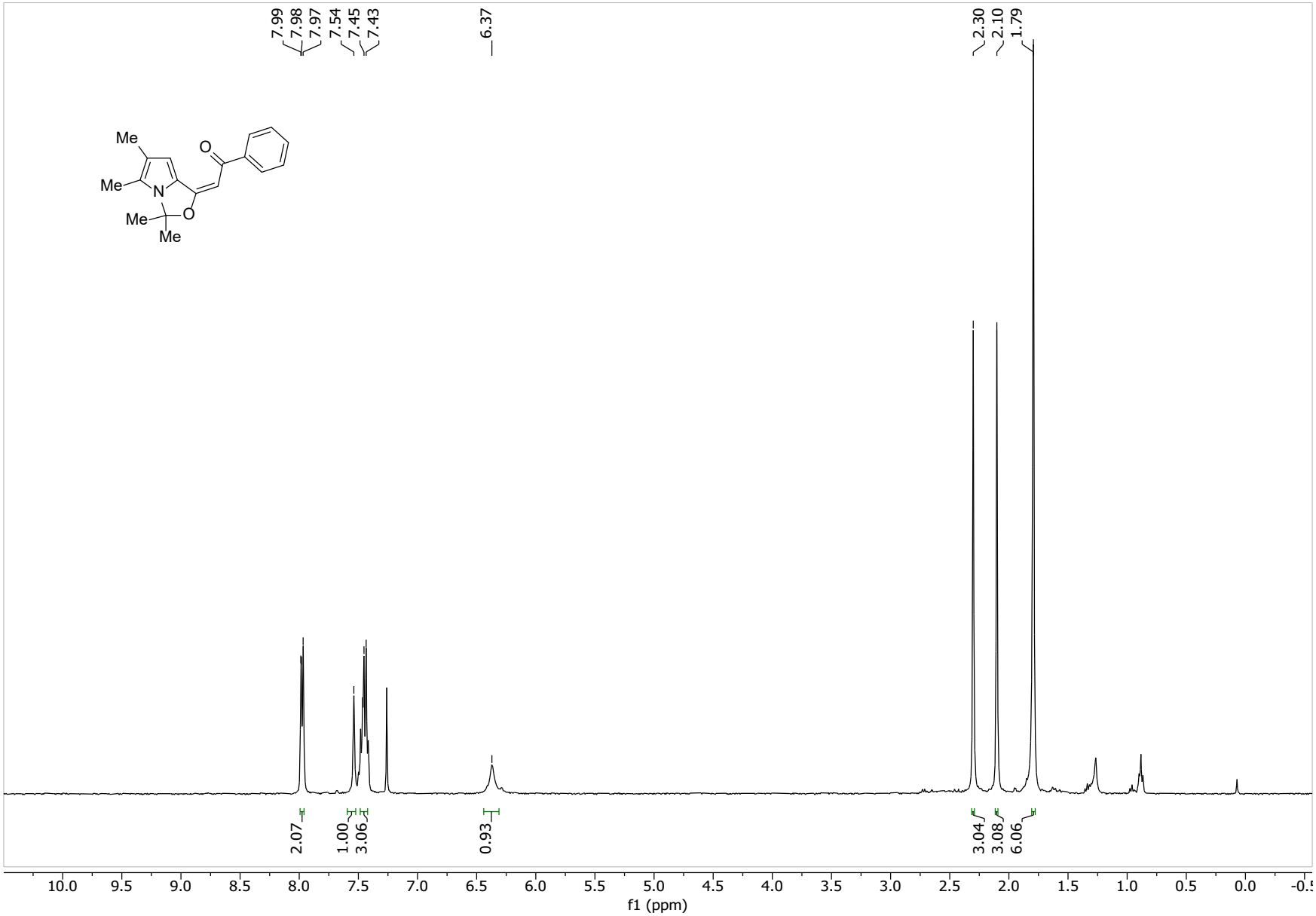
^{1}H NMR spectrum of (*E*)-2-(3,3-dimethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3ba**) in $CDCl_3$



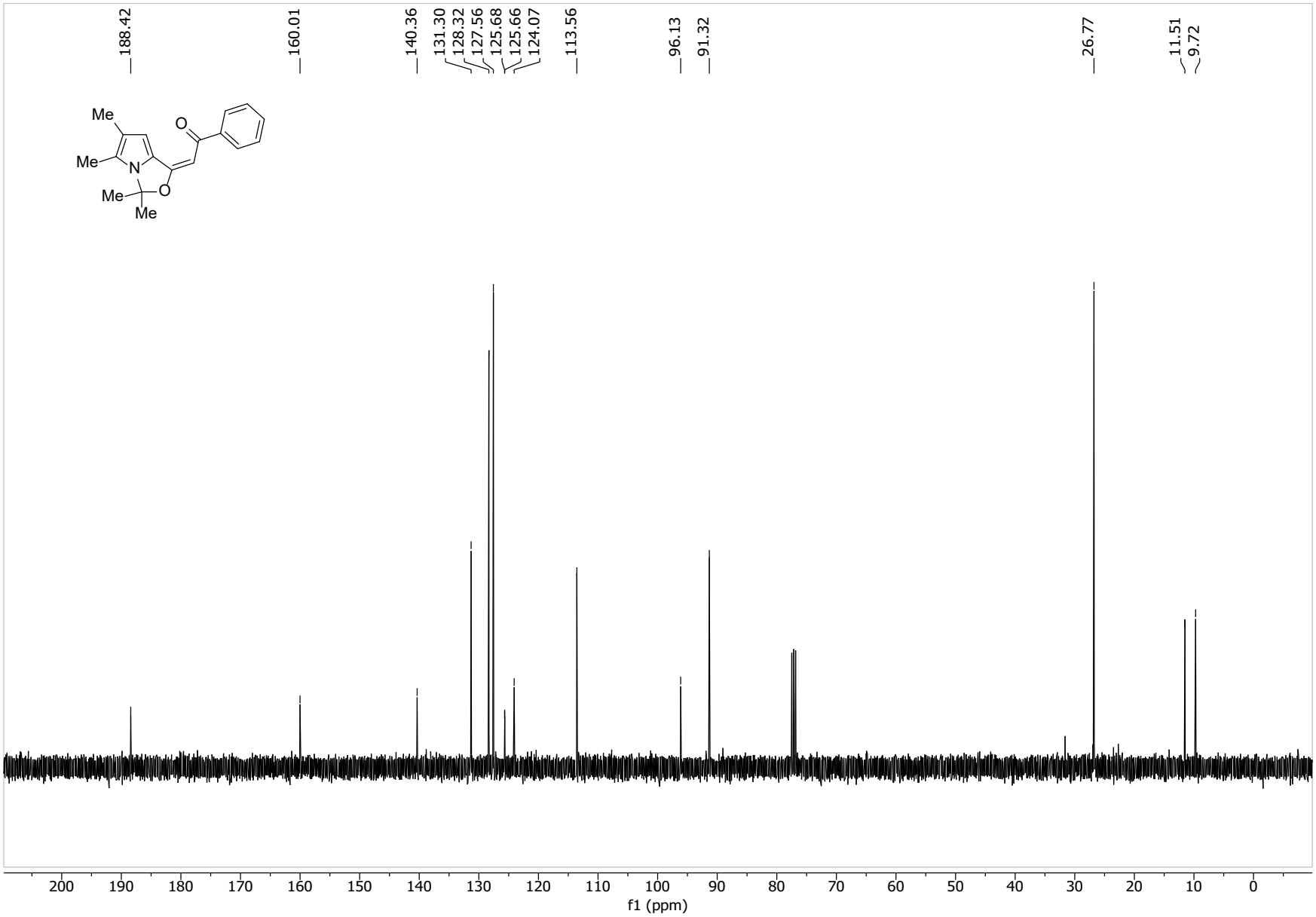
^{1}H NMR spectrum of (*E*)-2-(3,3-dimethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3ba**) in $CDCl_3$



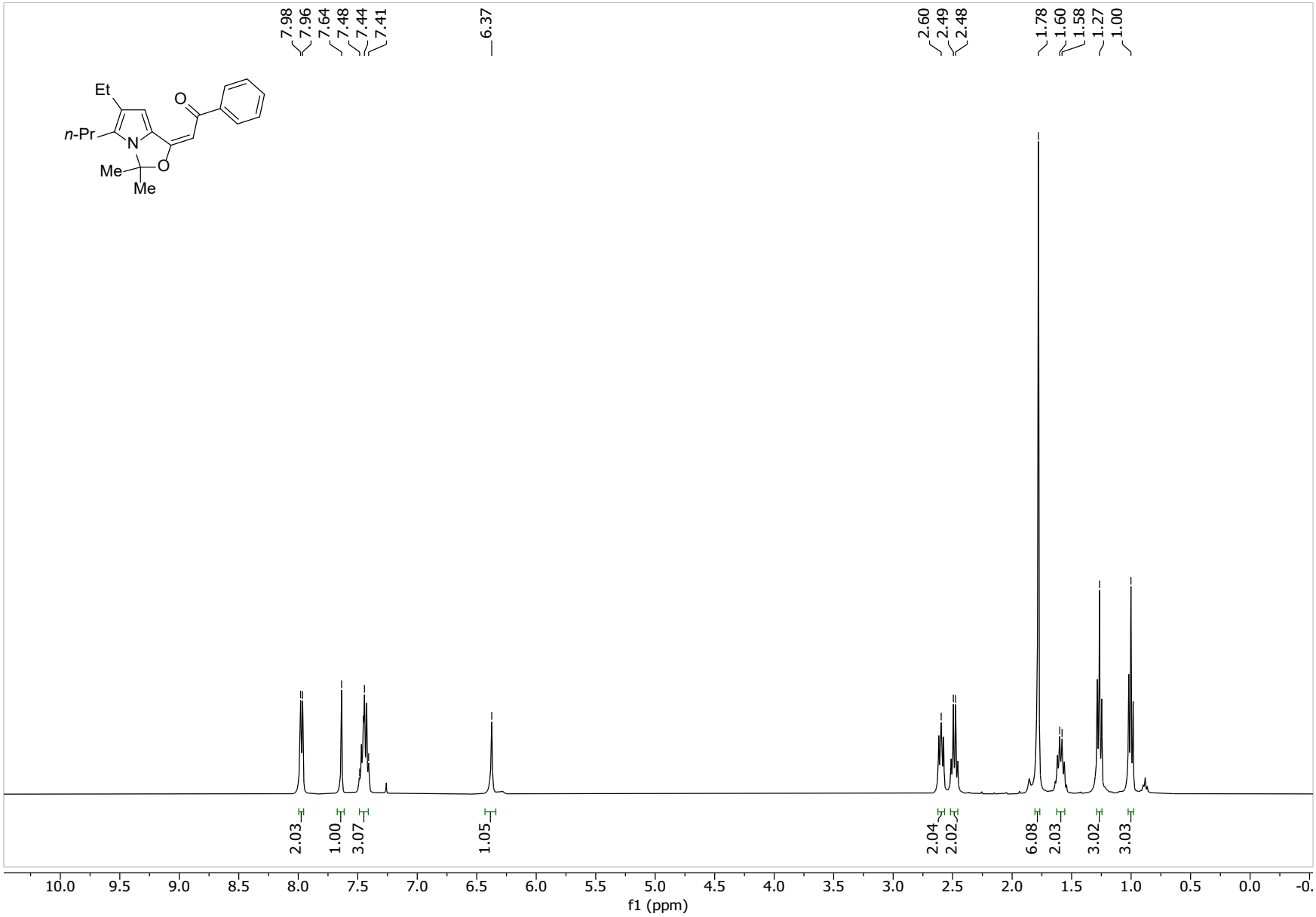
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(3,3,5,6-tetramethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3ca**) in $CDCl_3$



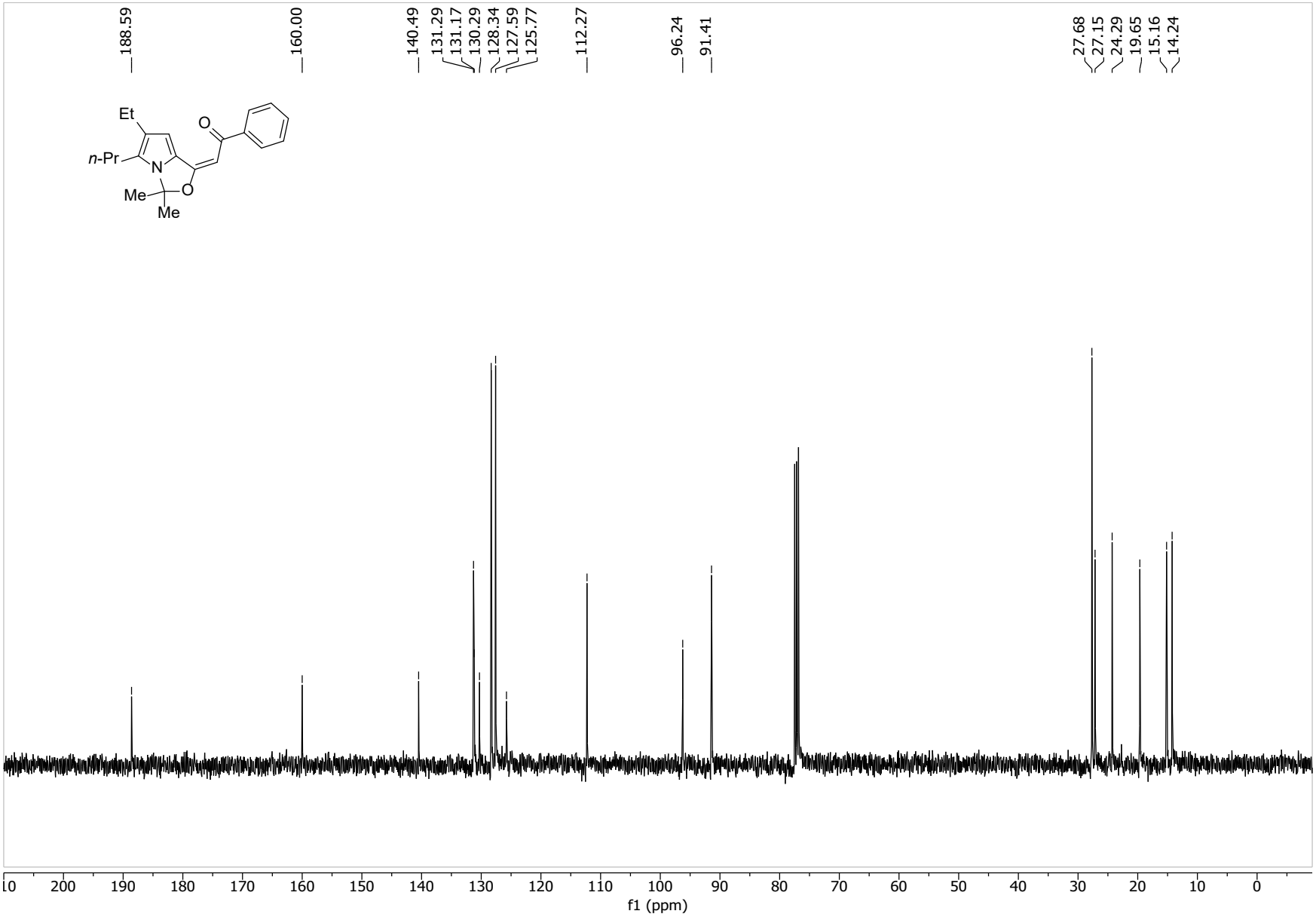
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(3,3,5,6-tetramethyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)ethan-1-one (**3ca**) in $CDCl_3$



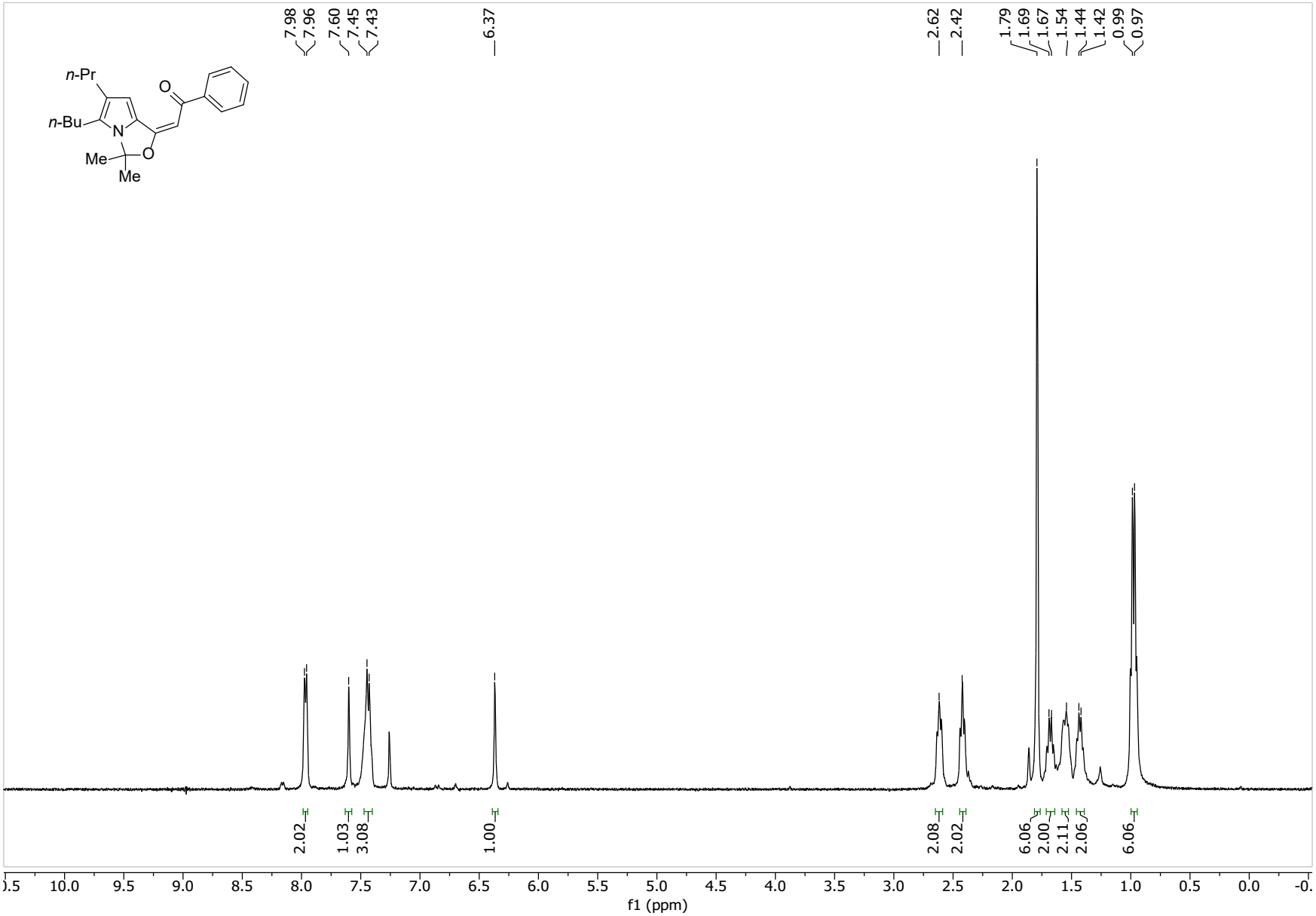
^{1}H NMR spectrum of (*E*)-2-(6-ethyl-3,3-dimethyl-5-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3da**) in $CDCl_3$



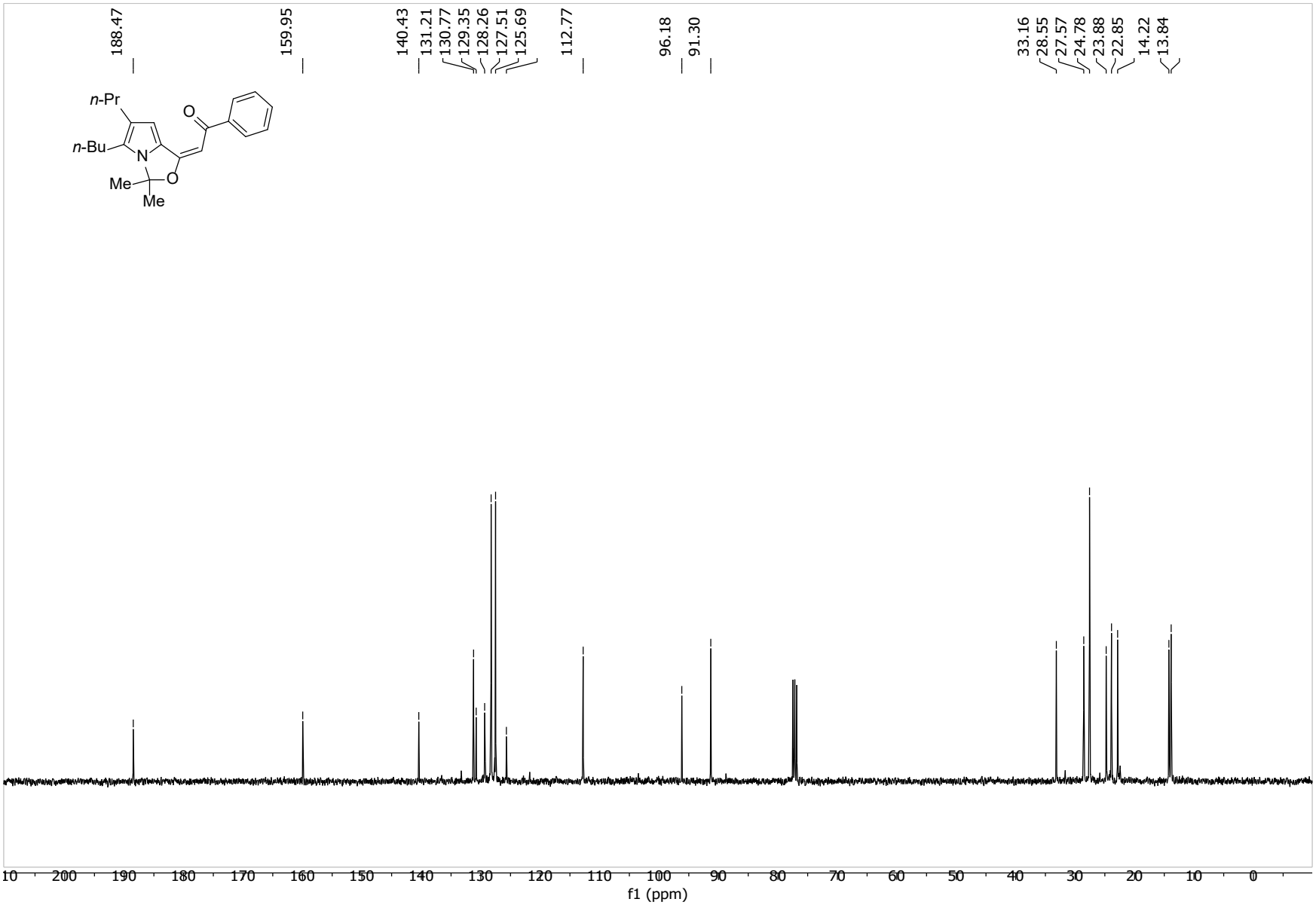
^{13}C NMR spectrum of (*E*)-2-(6-ethyl-3,3-dimethyl-5-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3da**) in $CDCl_3$



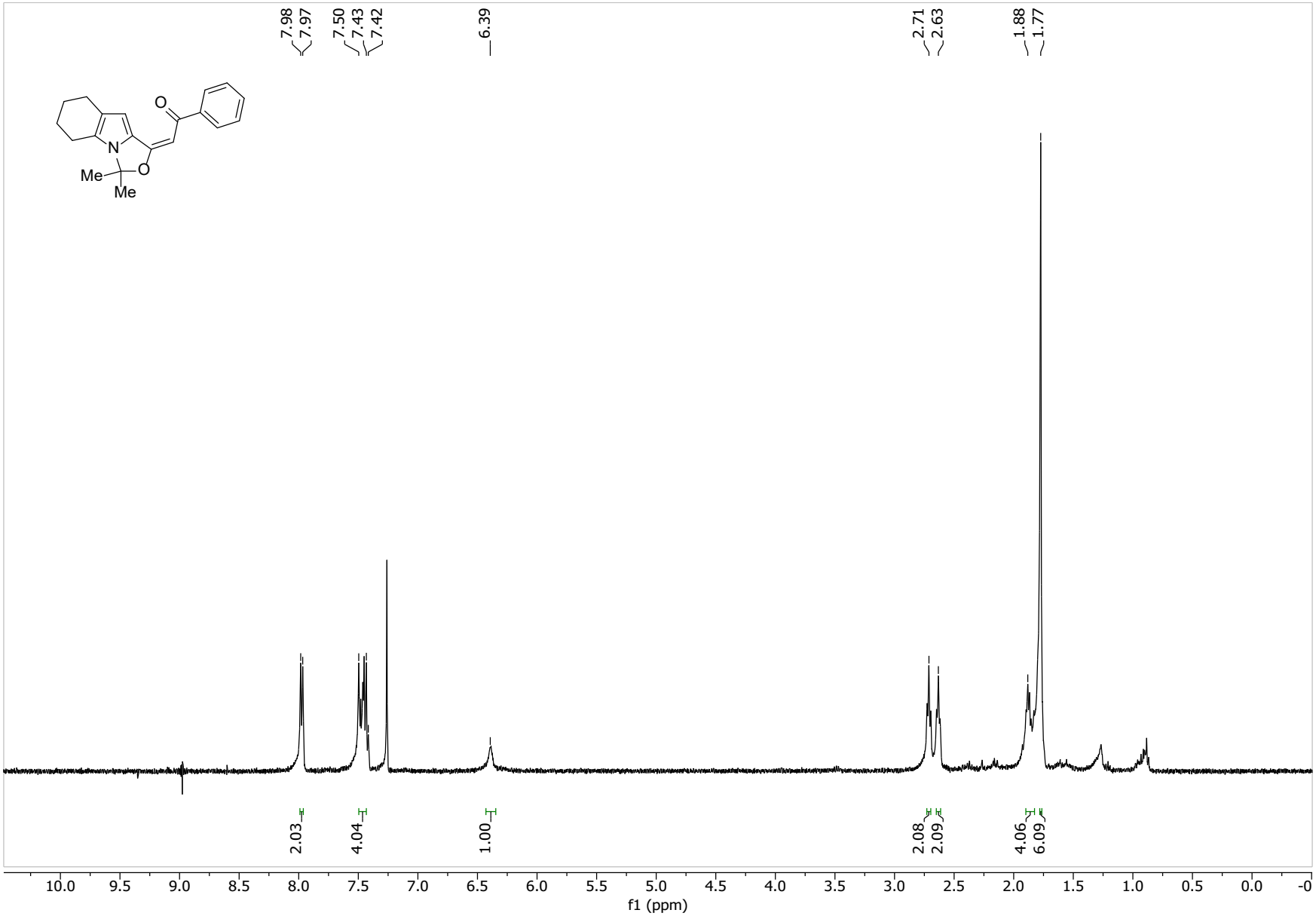
^{1}H NMR spectrum of (*E*)-2-(5-butyl-3,3-dimethyl-6-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3ea**) in $CDCl_3$



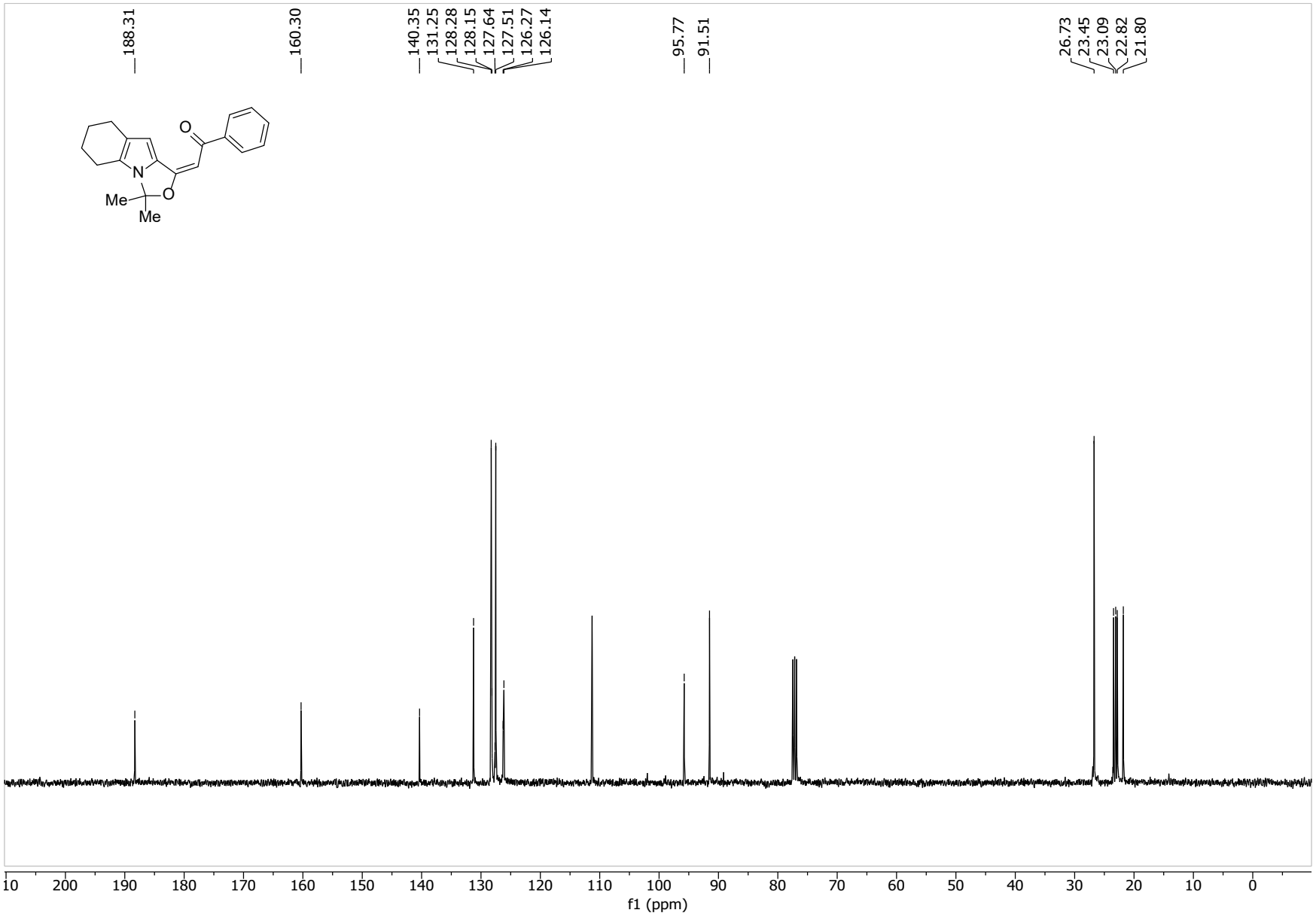
^{1}H NMR spectrum of (*E*)-2-(5-butyl-3,3-dimethyl-6-propyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3ea**) in $CDCl_3$



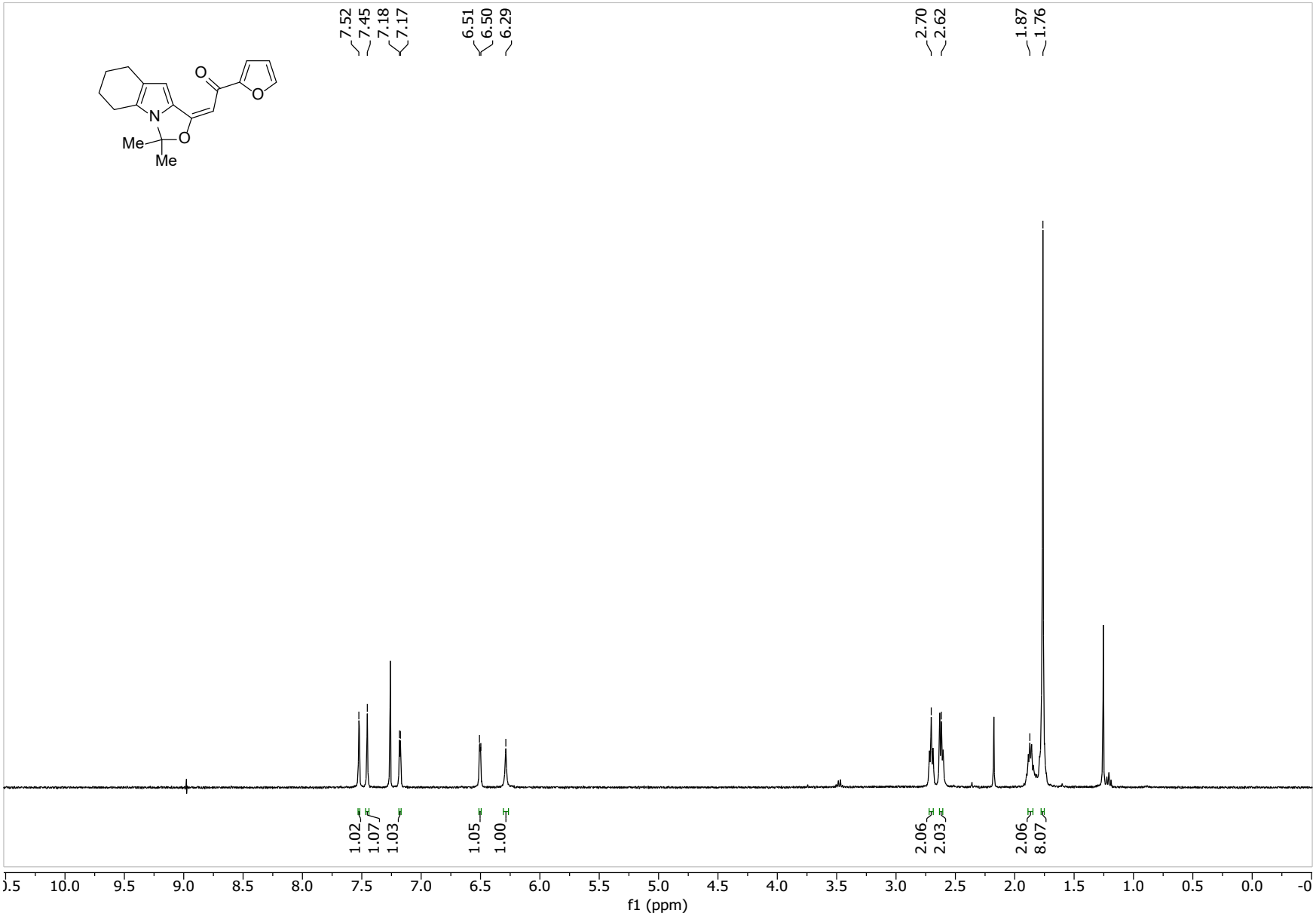
^{1}H NMR spectrum of (*E*)-2-(3,3-dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (**3fa**) in $CDCl_3$



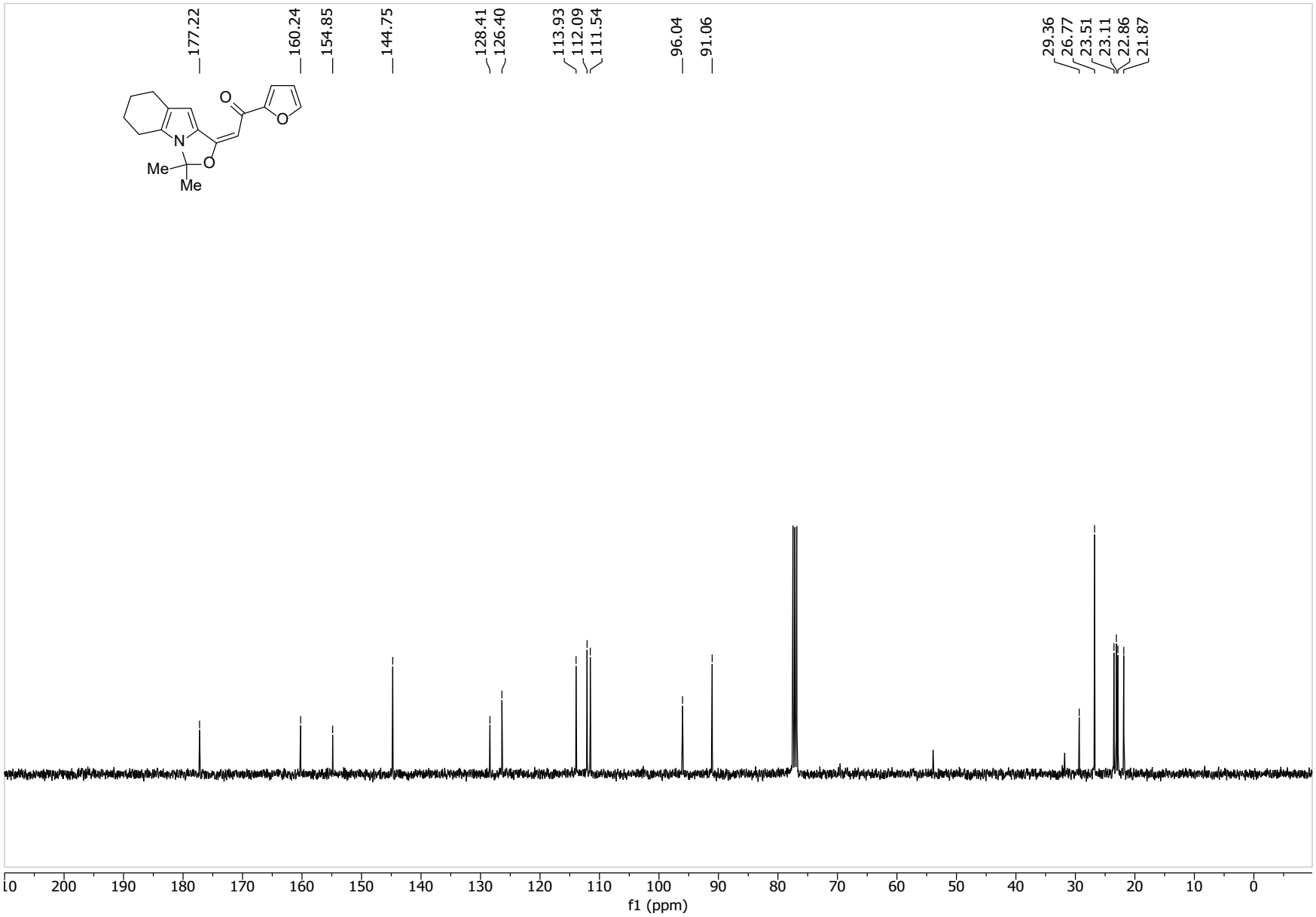
^{13}C NMR spectrum of (*E*)-2-(3,3-dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (**3fa**) in $CDCl_3$



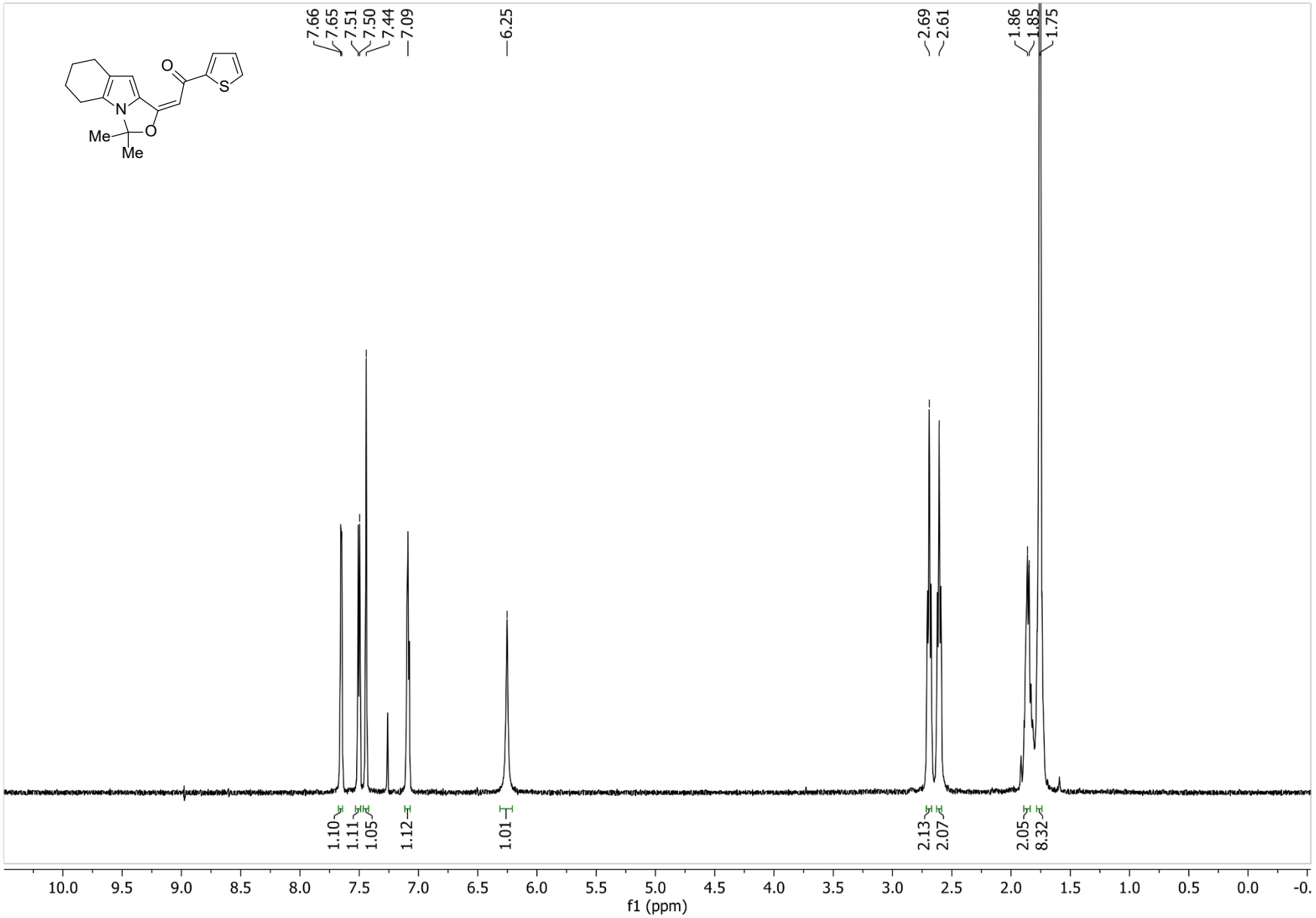
^{1}H NMR spectrum of (*E*)-2-(3,3-dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(furan-2-yl)ethan-1-one (**3ga**) in $CDCl_3$



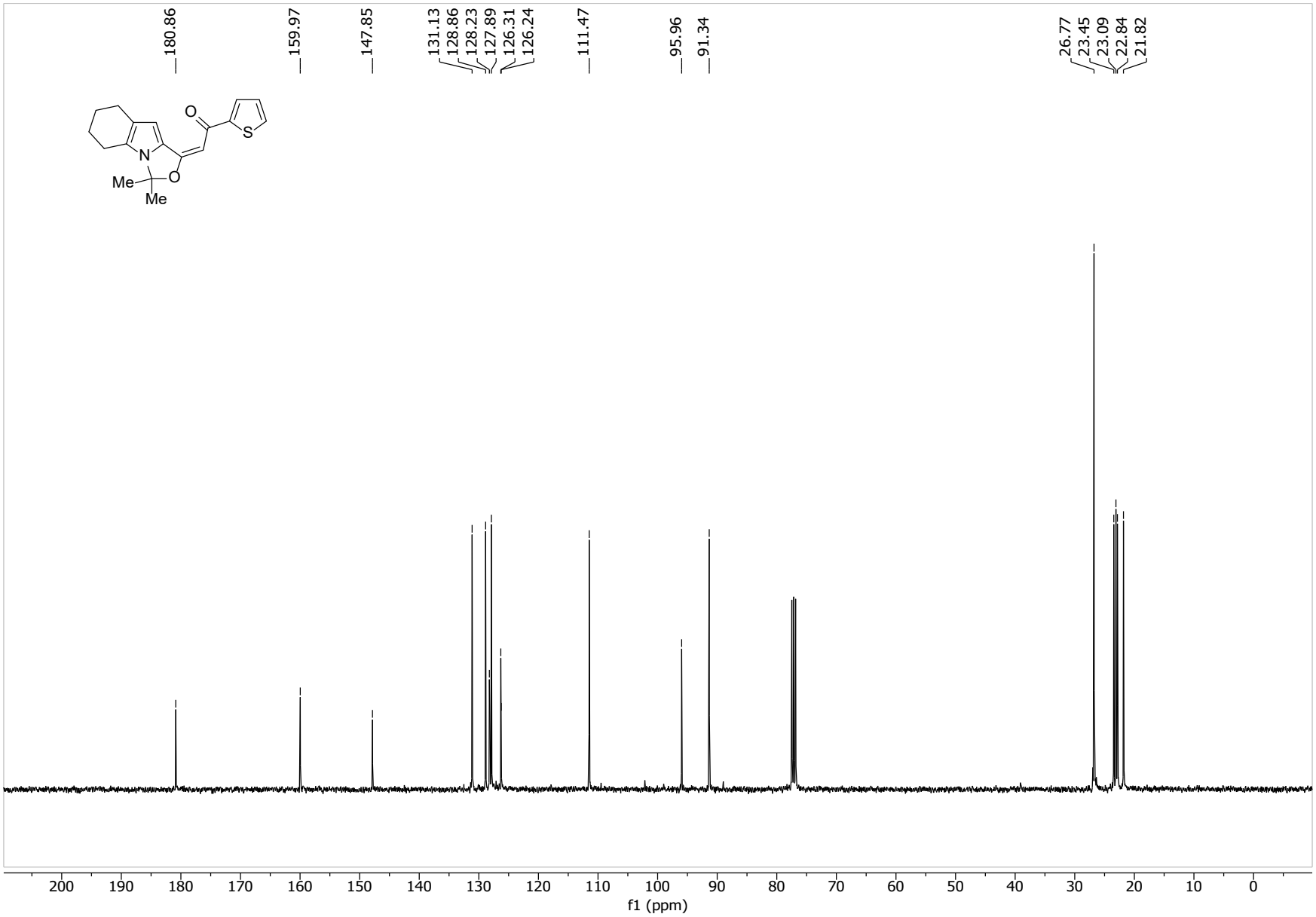
^{13}C NMR spectrum of (*E*)-2-(3,3-dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(furan-2-yl)ethan-1-one (**3ga**) in $CDCl_3$



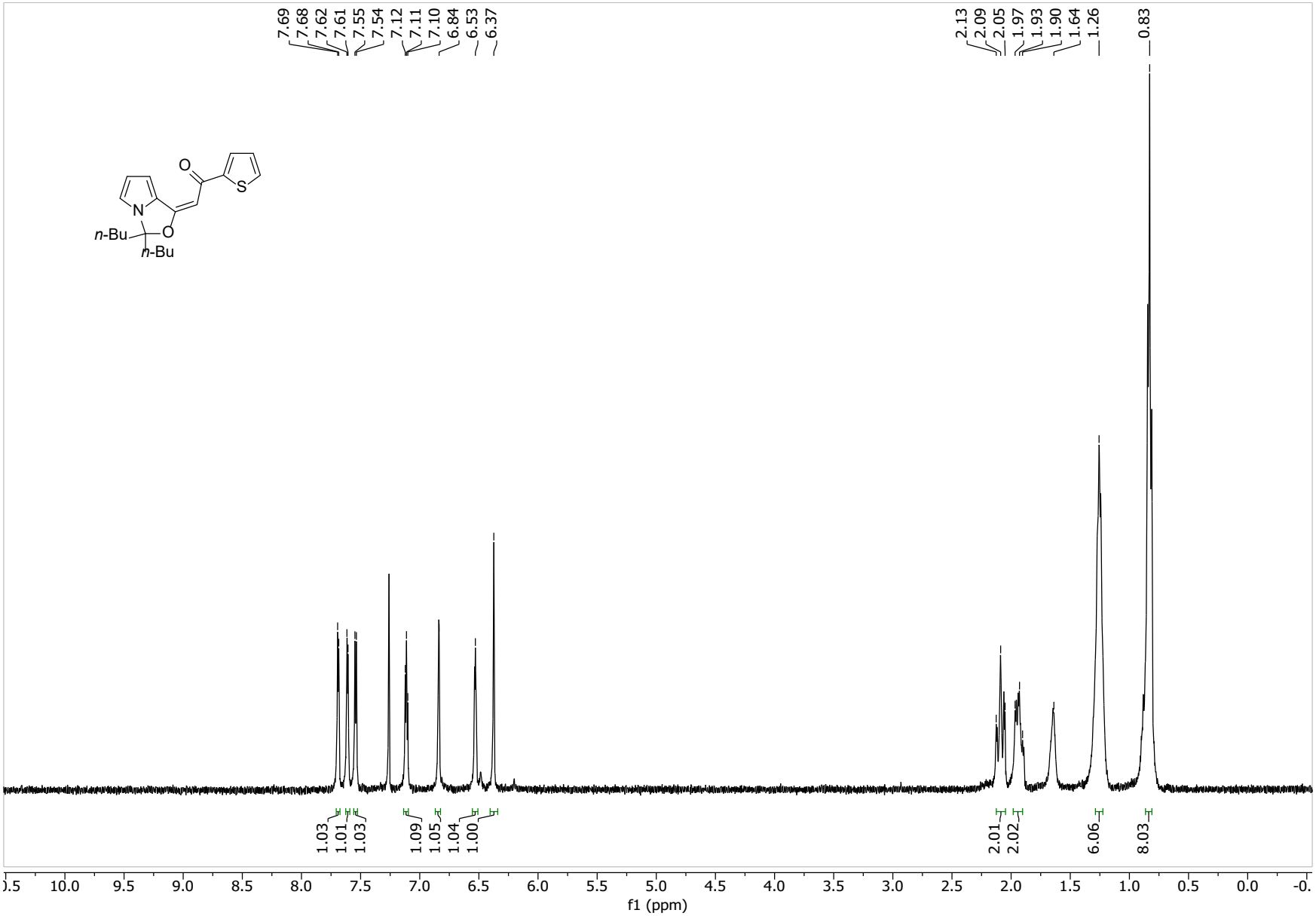
^{1}H NMR spectrum of (*E*)-2-(3,3-dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3ha**) in $CDCl_3$



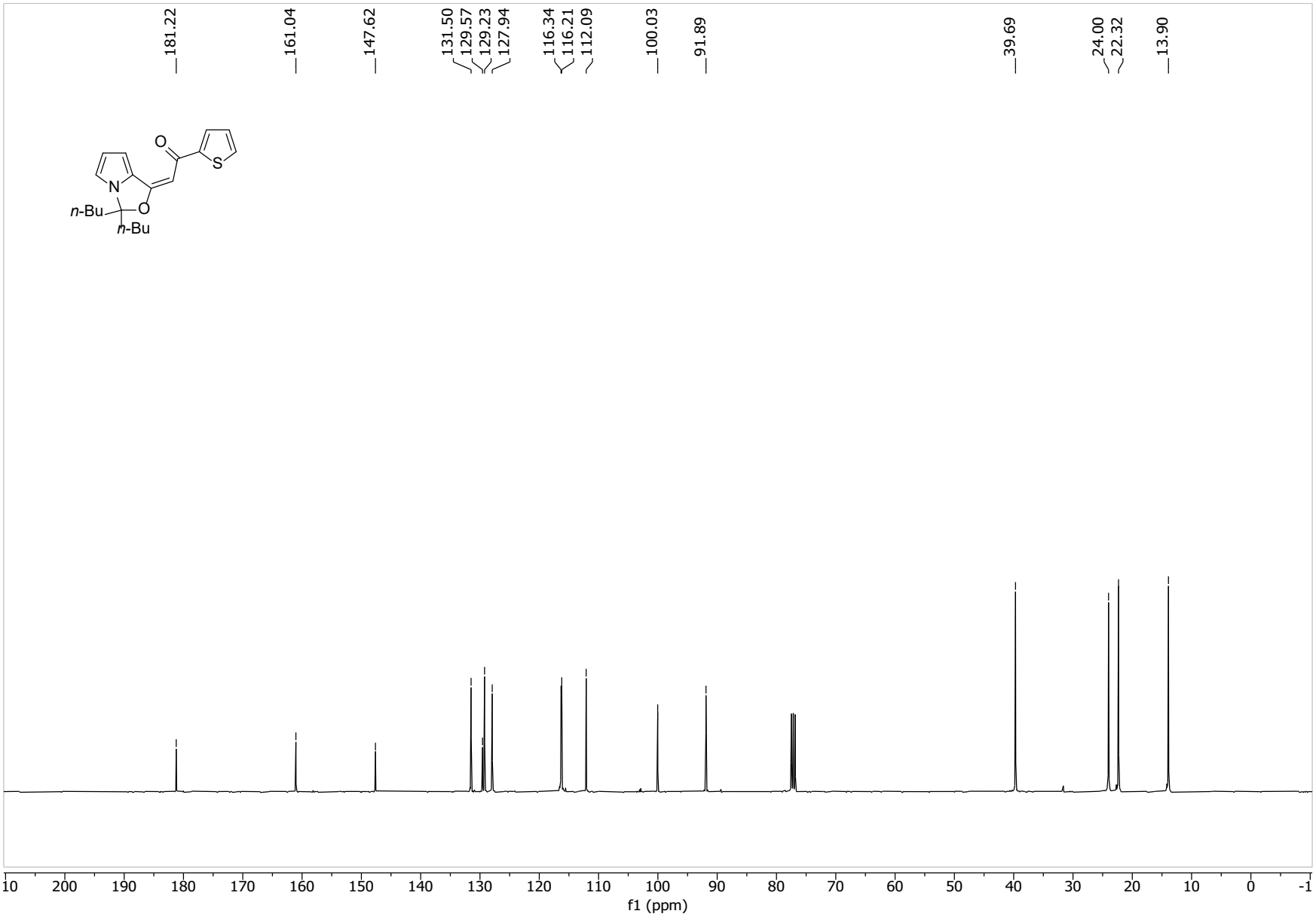
^{13}C NMR spectrum of (*E*)-2-(3,3-dimethyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3ha**) in $CDCl_3$



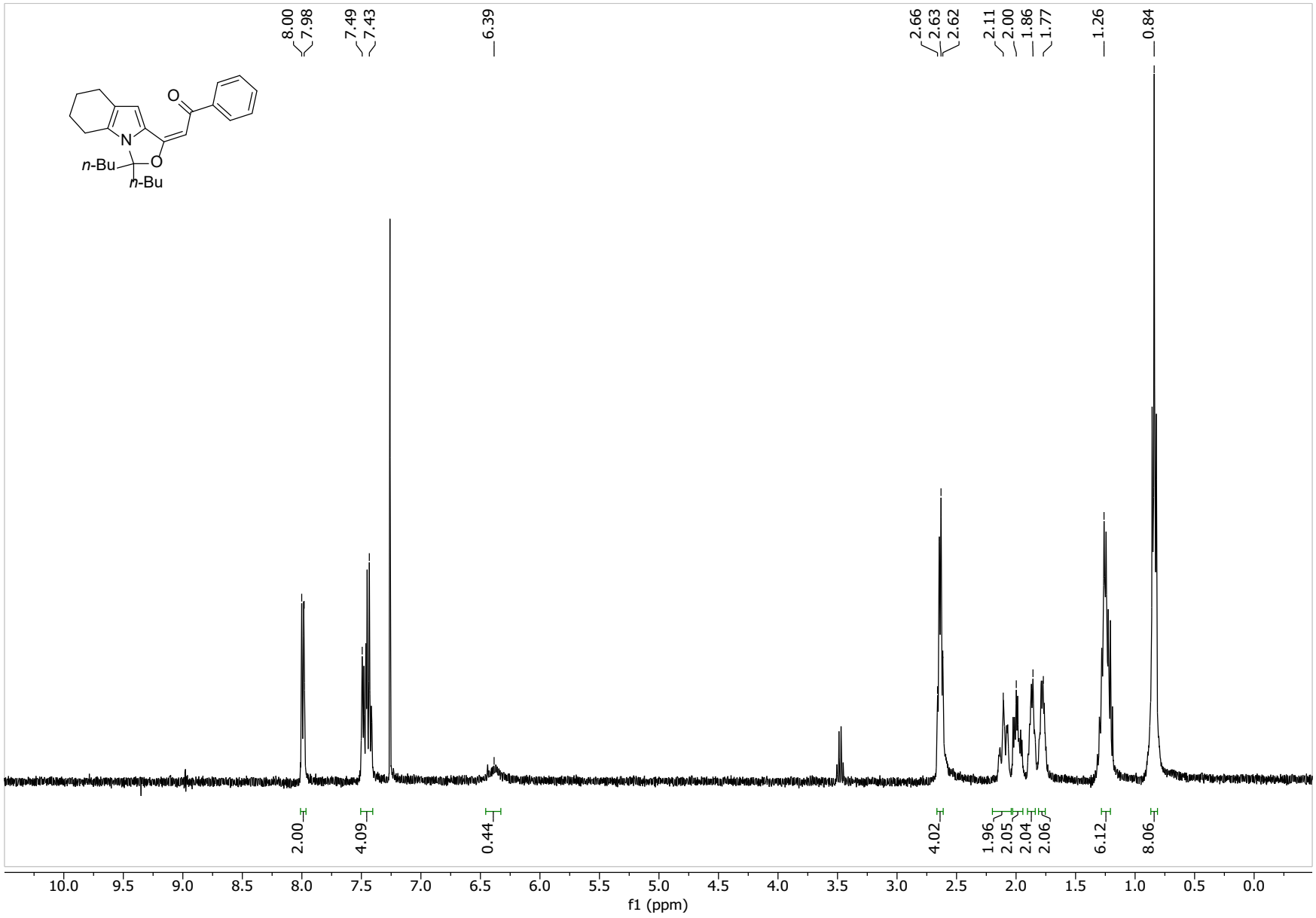
^{1}H NMR spectrum of (*E*)-2-(3,3-dibutyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3bb**) in $CDCl_3$



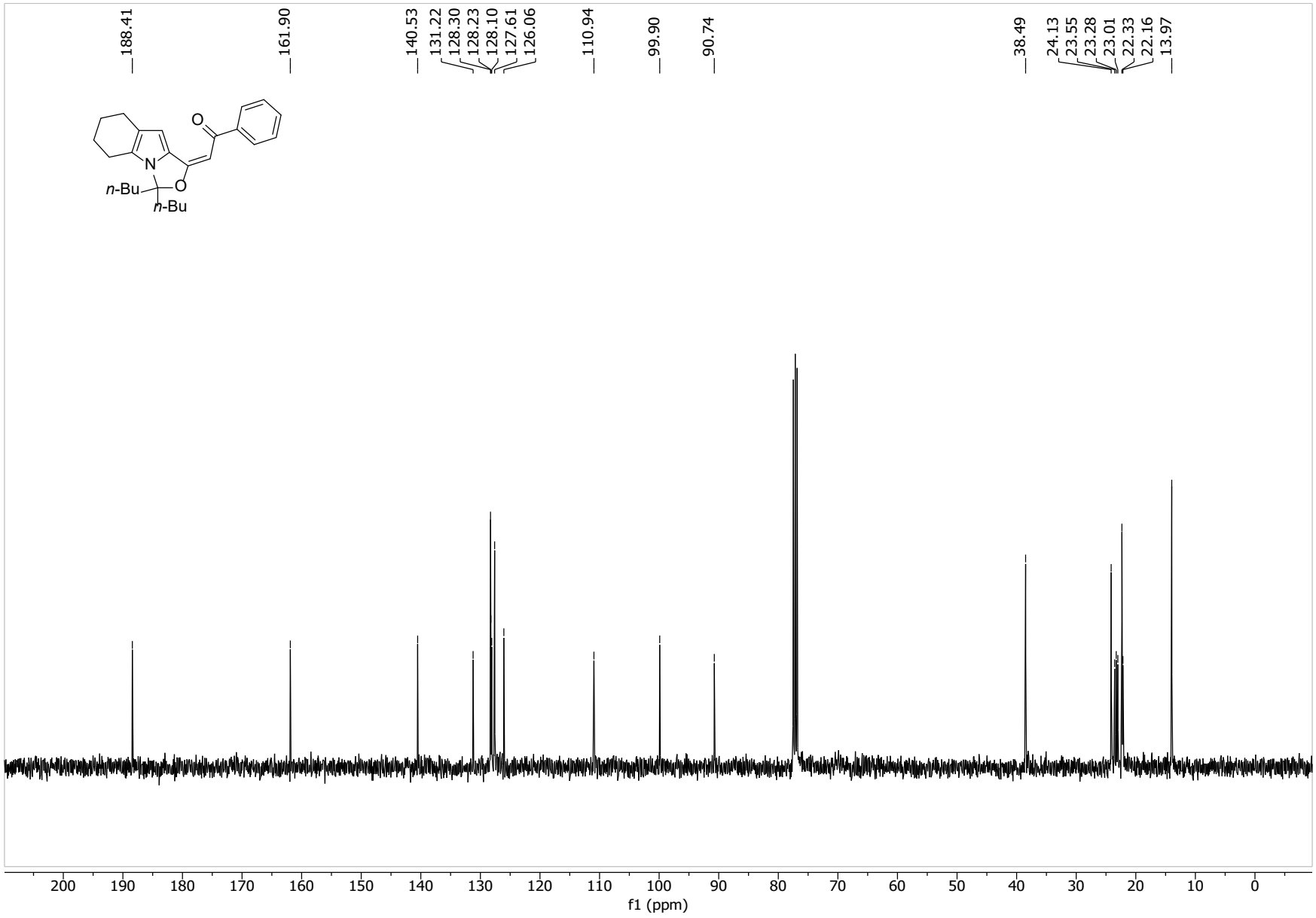
^{13}C NMR spectrum of (*E*)-2-(3,3-dibutyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3bb**) in $CDCl_3$



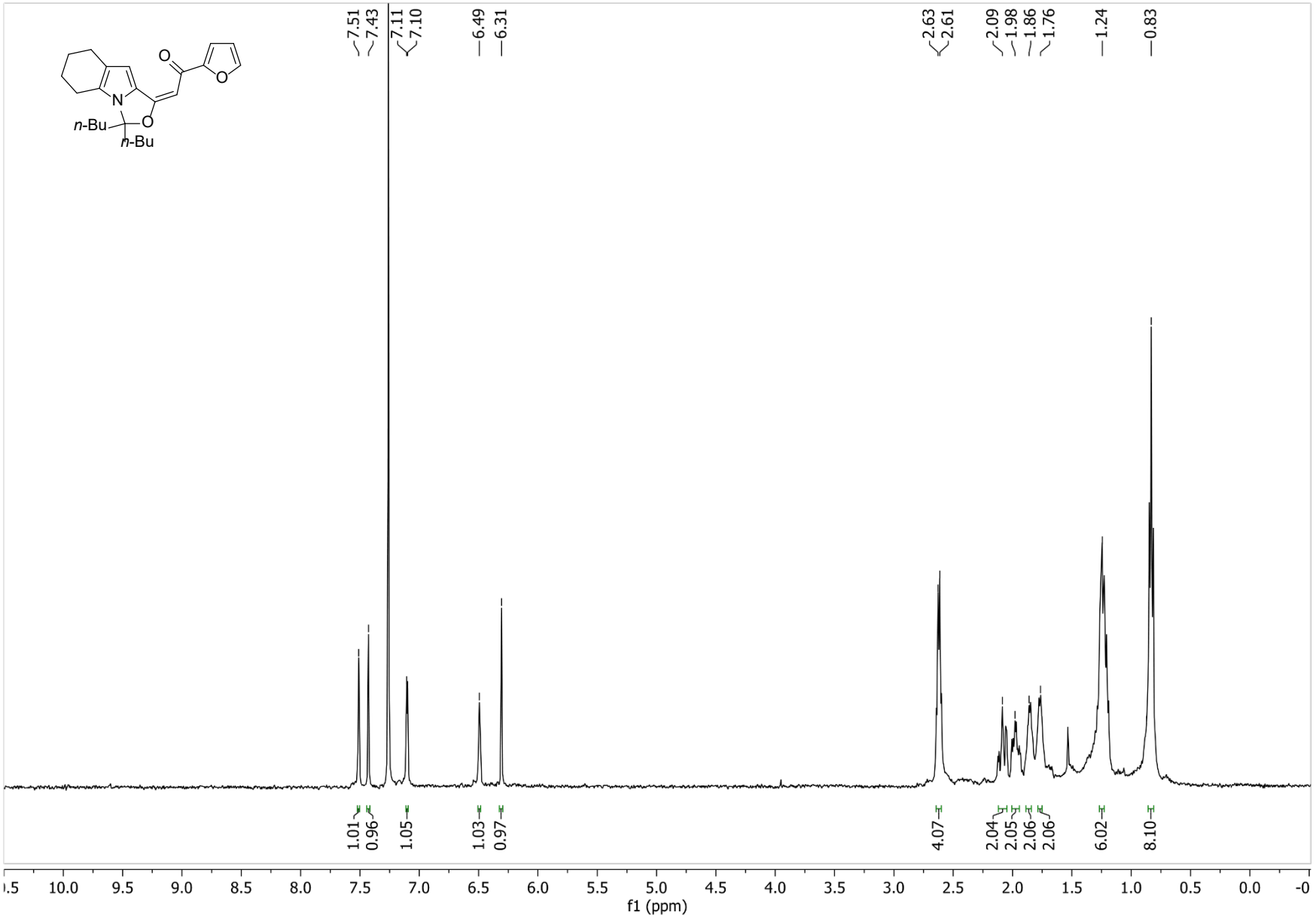
^{1}H NMR spectrum of (*E*)-2-(3,3-dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (**3fb**) in $CDCl_3$



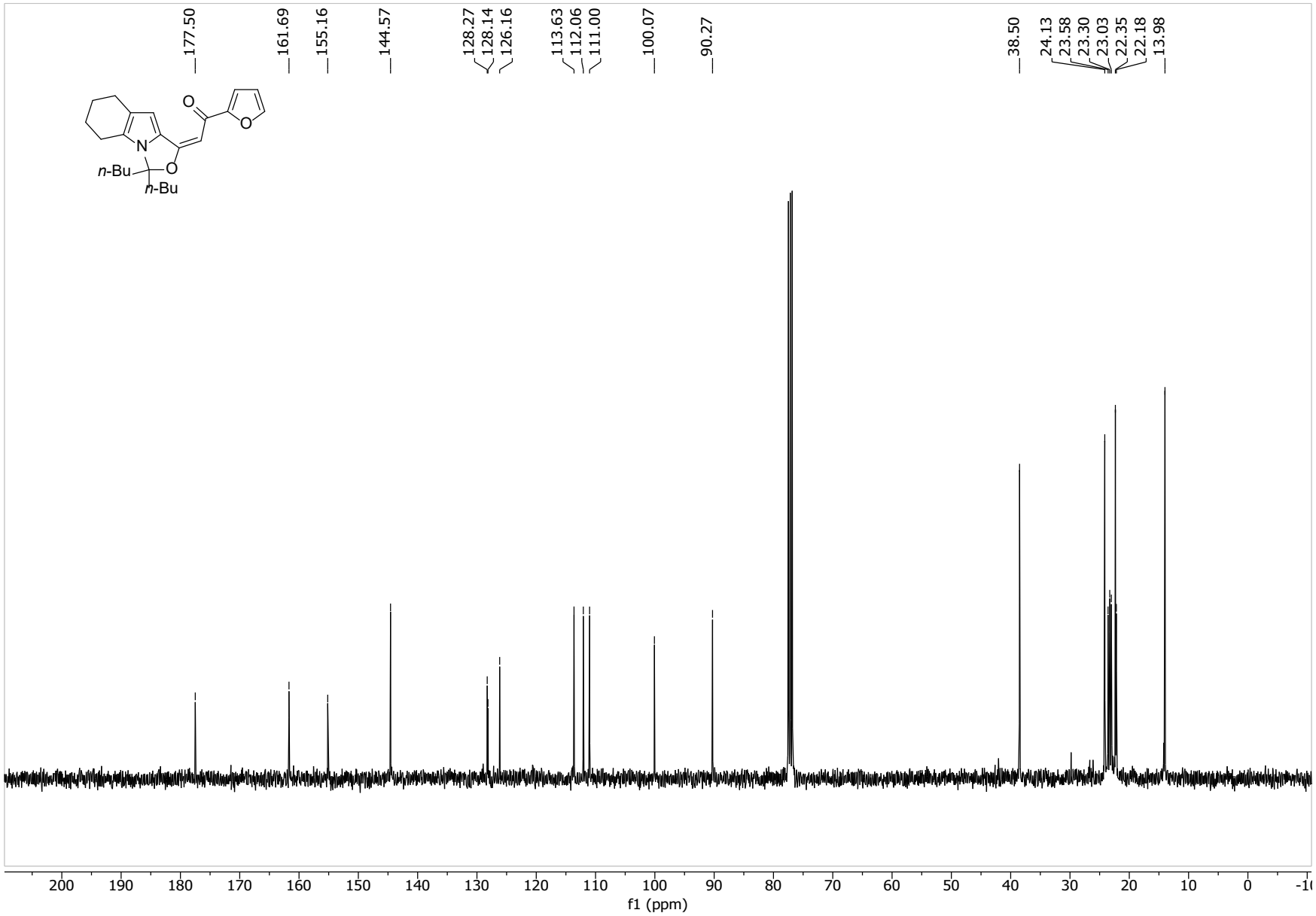
^{13}C NMR spectrum of (*E*)-2-(3,3-dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (**3fb**) in $CDCl_3$



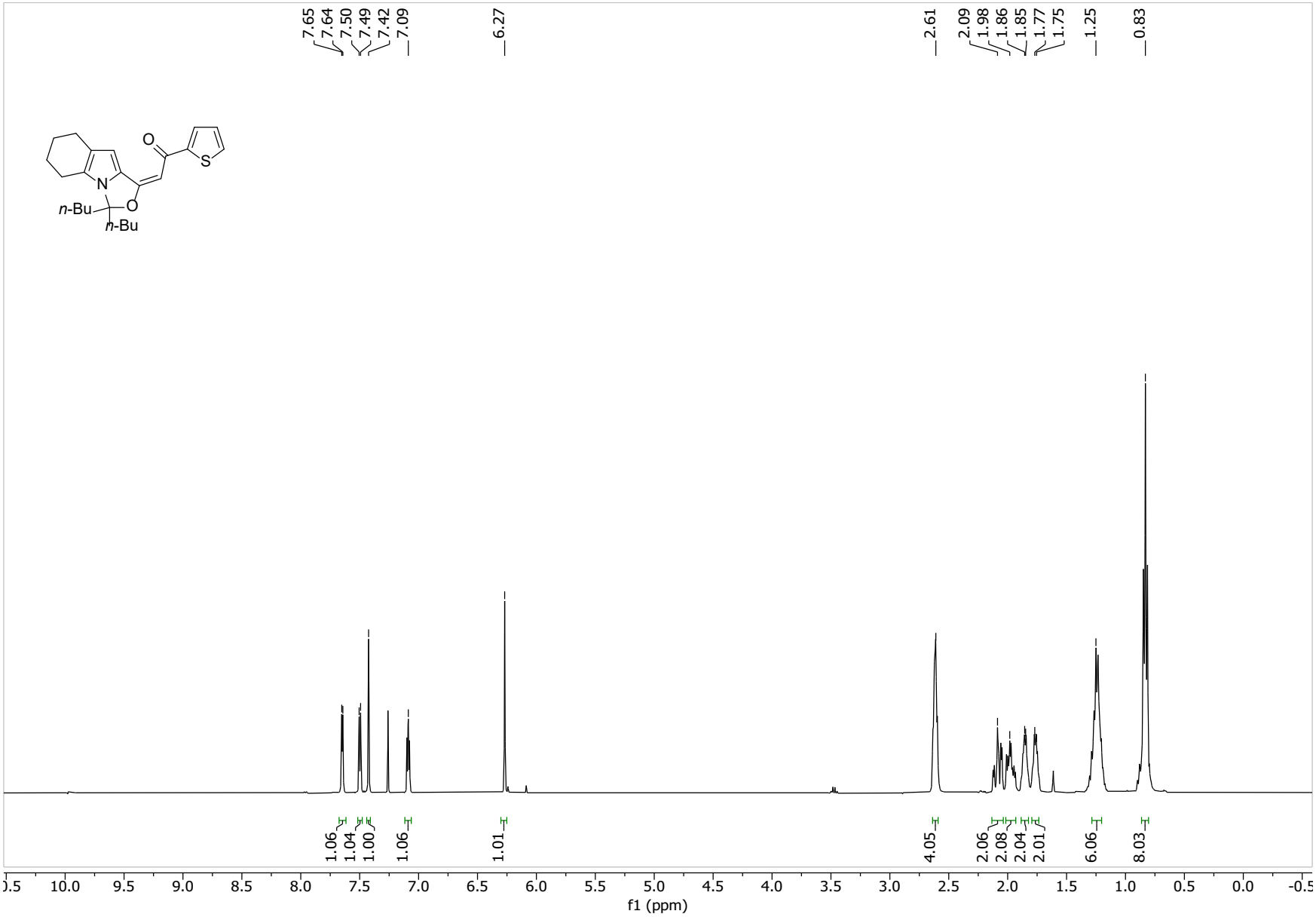
^{1}H NMR spectrum of (*E*)-2-(3,3-dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(furan-2-yl)ethan-1-one (**3gb**) in $CDCl_3$



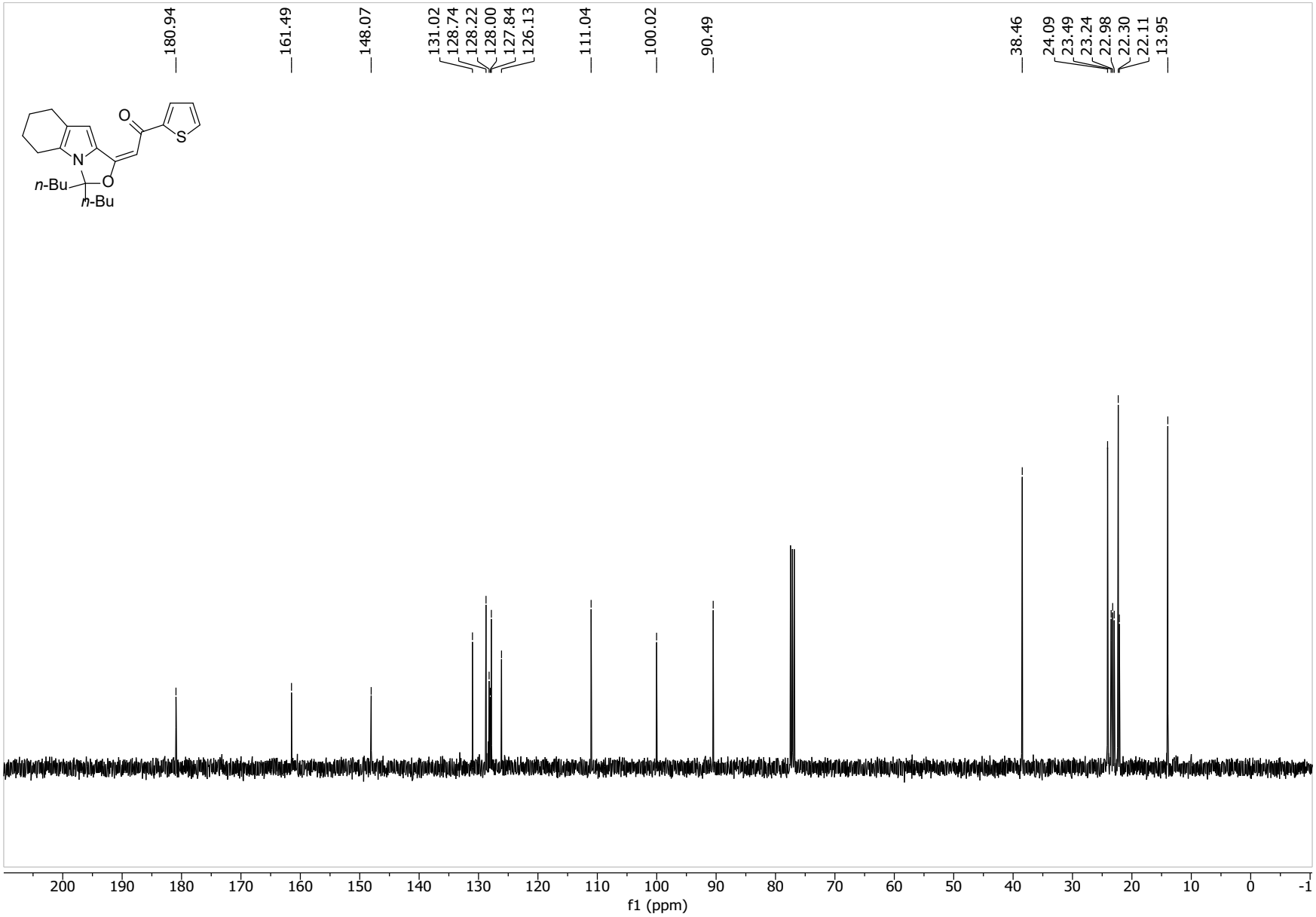
^{13}C NMR spectrum of (*E*)-2-(3,3-dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(furan-2-yl)ethan-1-one (**3gb**) in $CDCl_3$



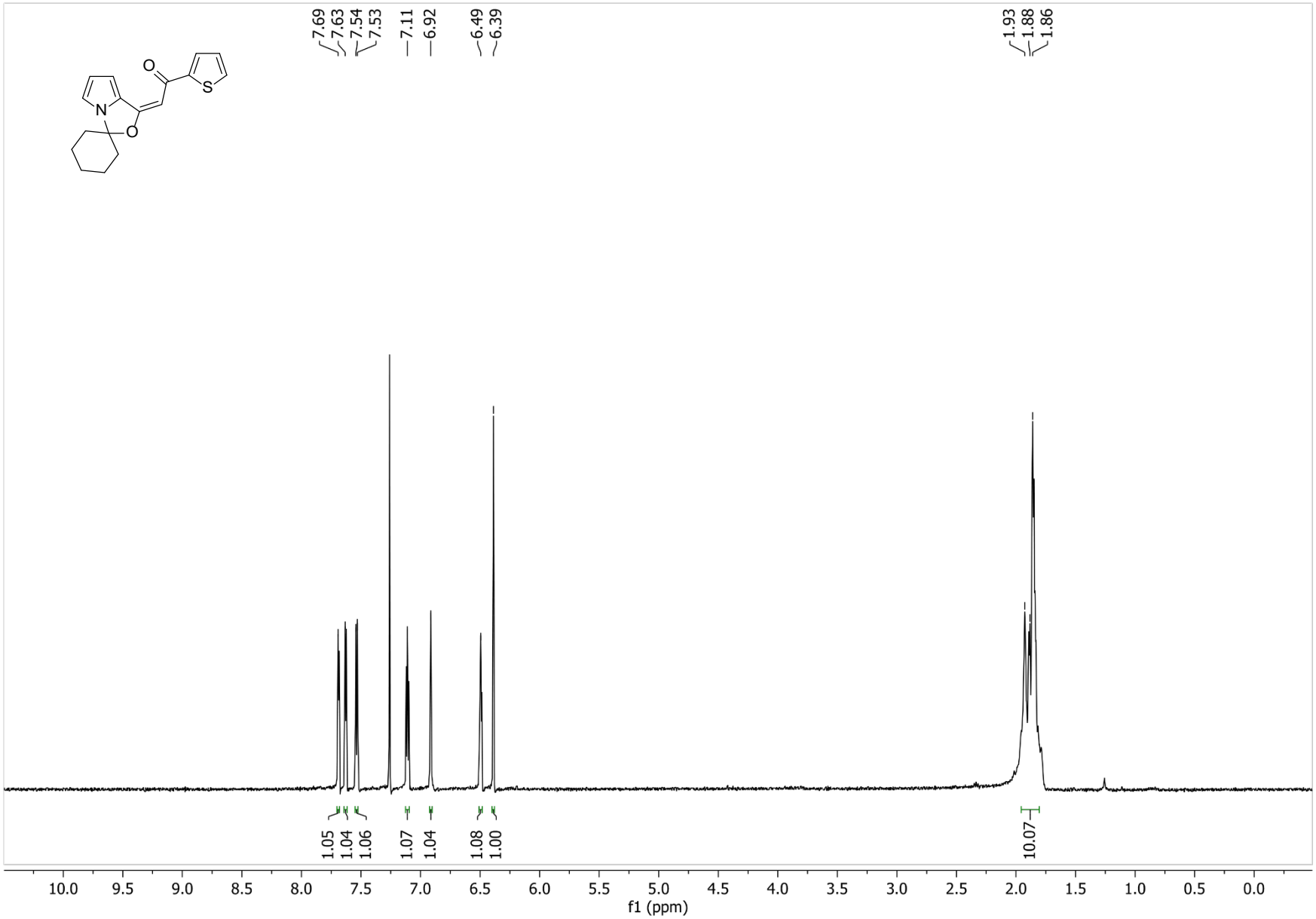
^{1}H NMR spectrum of (*E*)-2-(3,3-dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3hb**) in $CDCl_3$



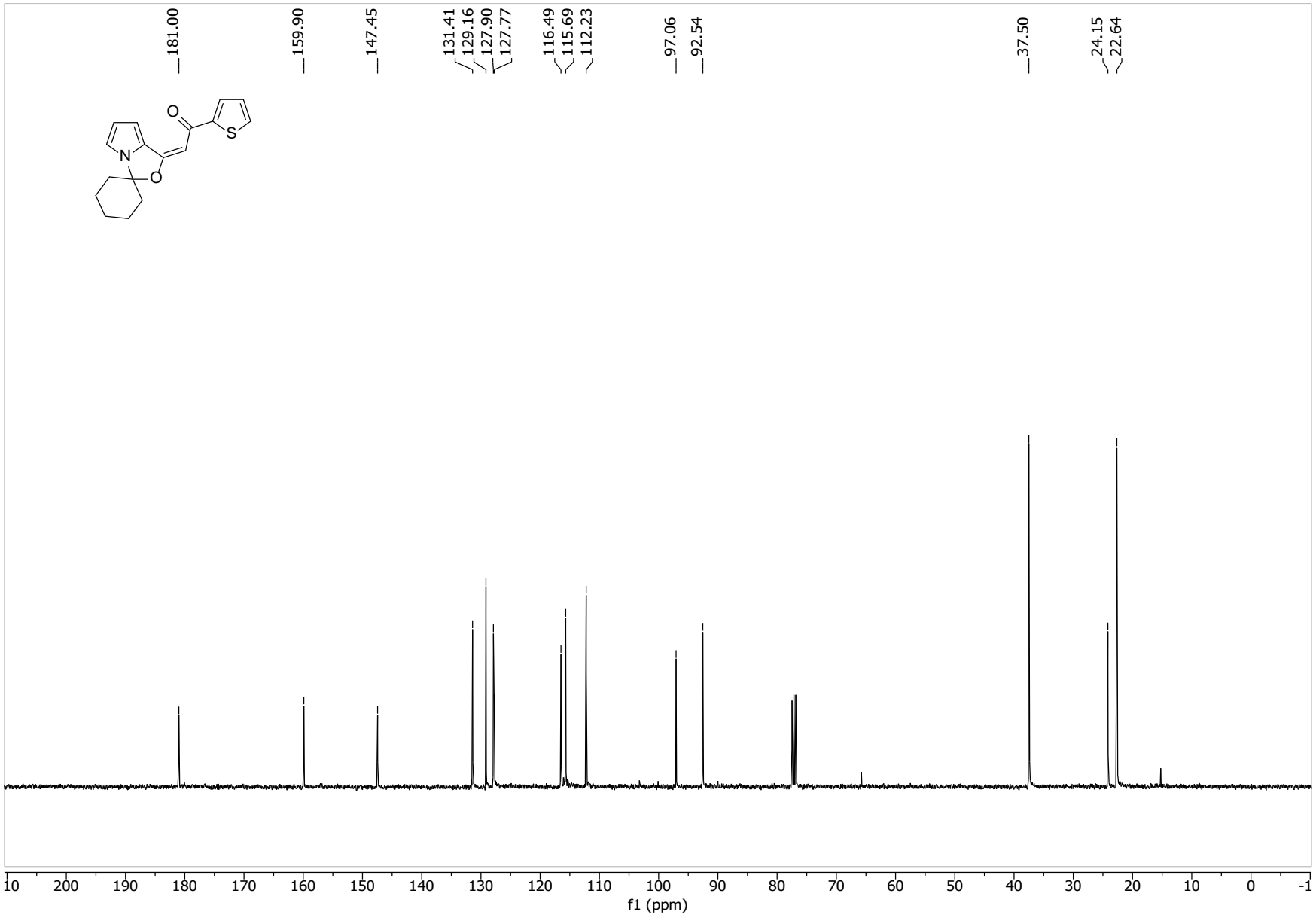
^{13}C NMR spectrum of (*E*)-2-(3,3-dibutyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3hb**) in $CDCl_3$



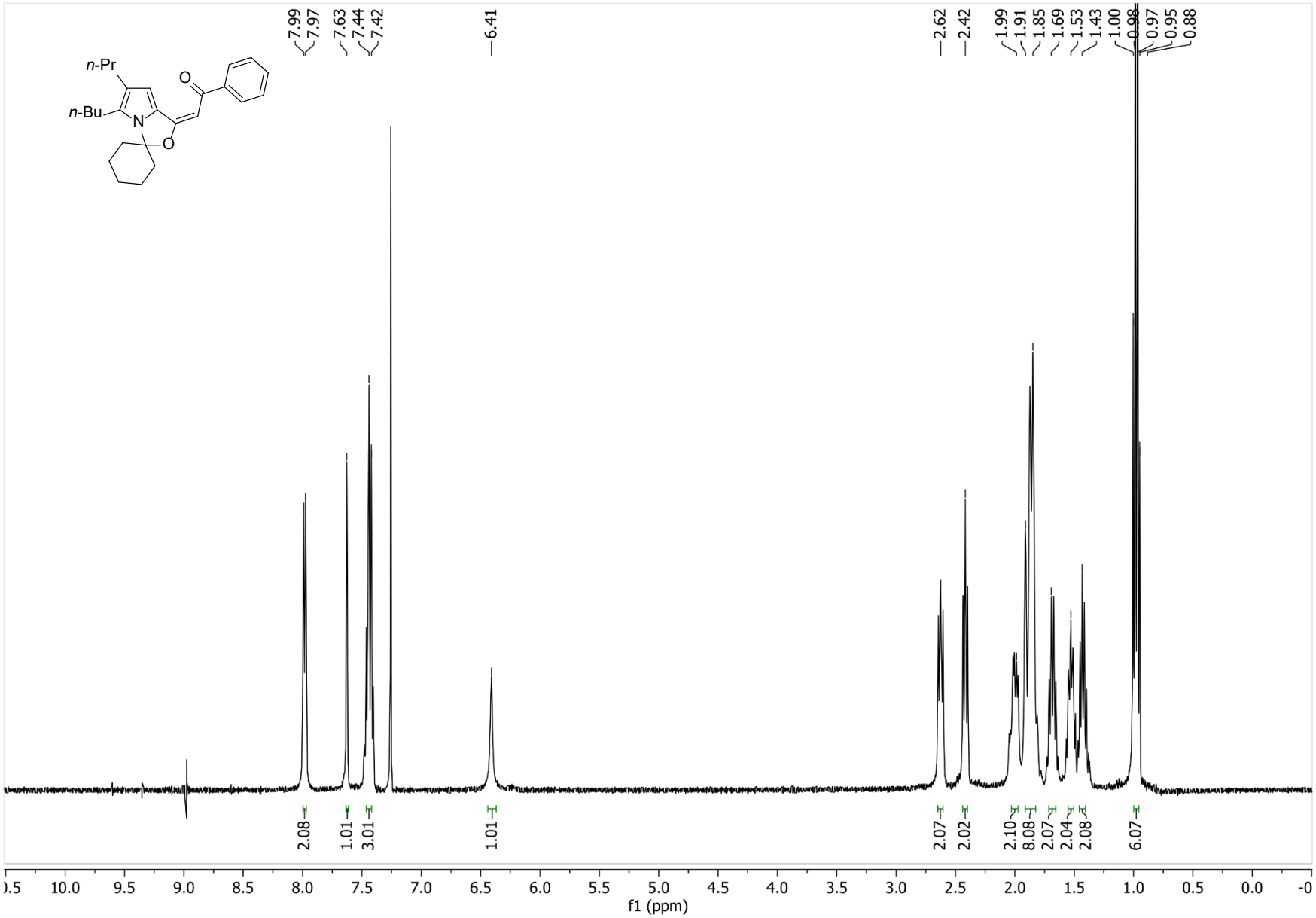
^{1}H NMR spectrum of (*E*)-2-(1'H-spiro[cyclohexane-1, 3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3bd**) in $CDCl_3$



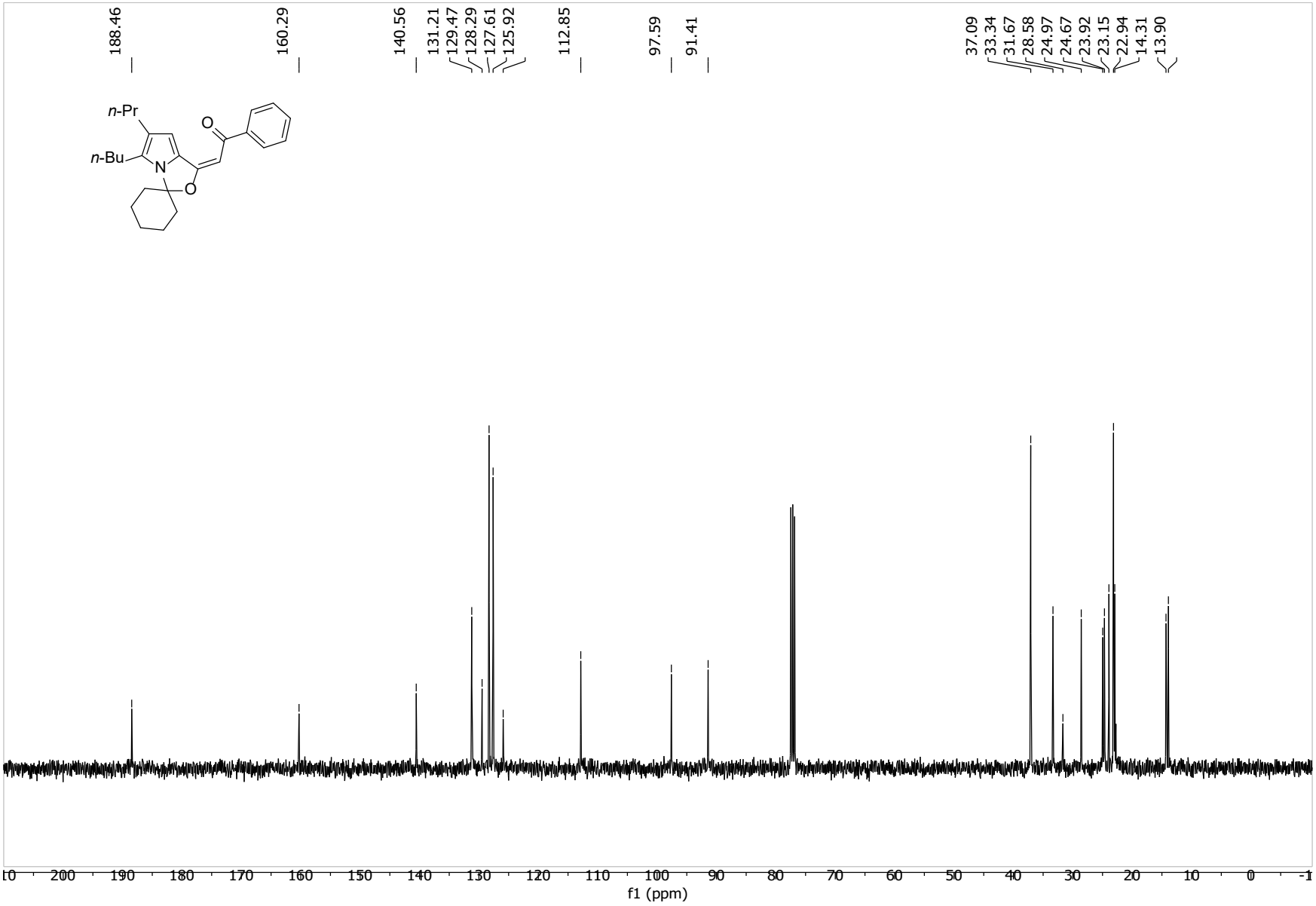
^{13}C NMR spectrum of (*E*)-2-(1'H-spiro[cyclohexane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3bd**) in $CDCl_3$



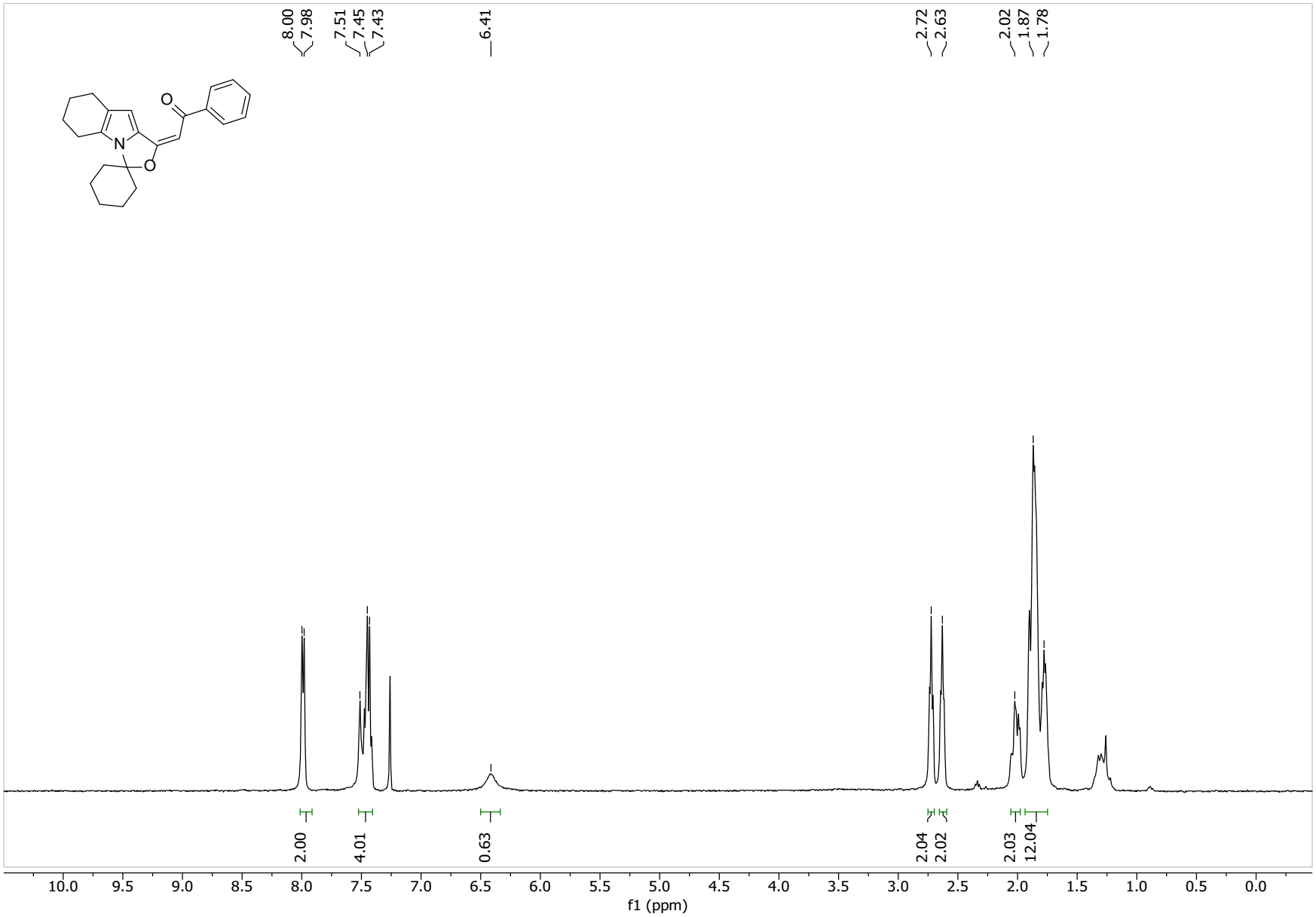
^{1}H NMR spectrum of (*E*)-2-(5'-butyl-6'-propyl-1'*H*-spiro[cyclohexane-1,3'-pyrrolo [1,2-*c*]oxazol]-1'-ylidene)-1-phenylethan-1-one (**3cd**) in $CDCl_3$



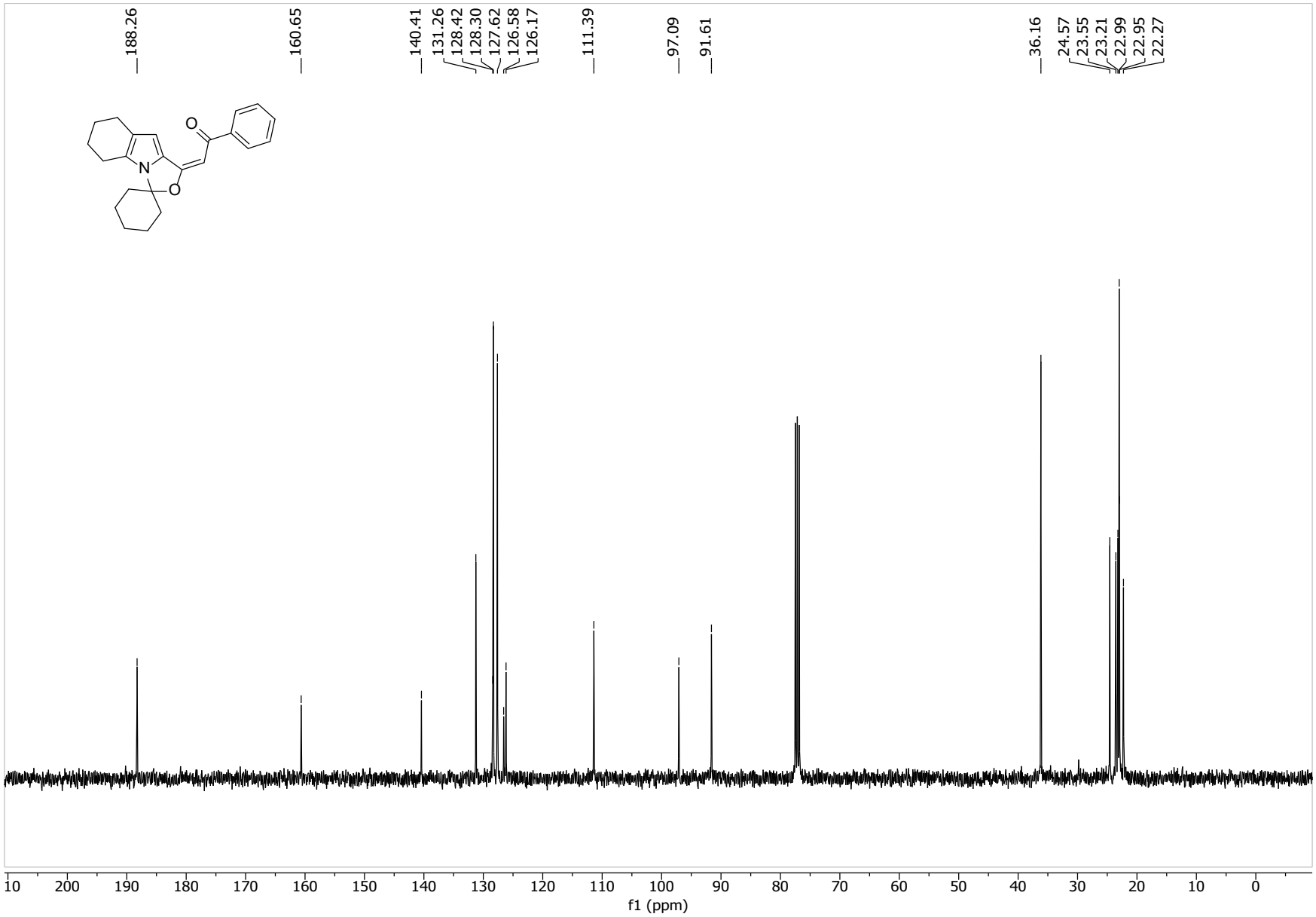
^{13}C NMR spectrum of (*E*)-2-(5'-butyl-6'-propyl-1'*H*-spiro[cyclohexane-1,3'-pyrrolo[1,2-*c*]oxazol]-1'-ylidene)-1-phenylethan-1-one (**3cd**) in $CDCl_3$



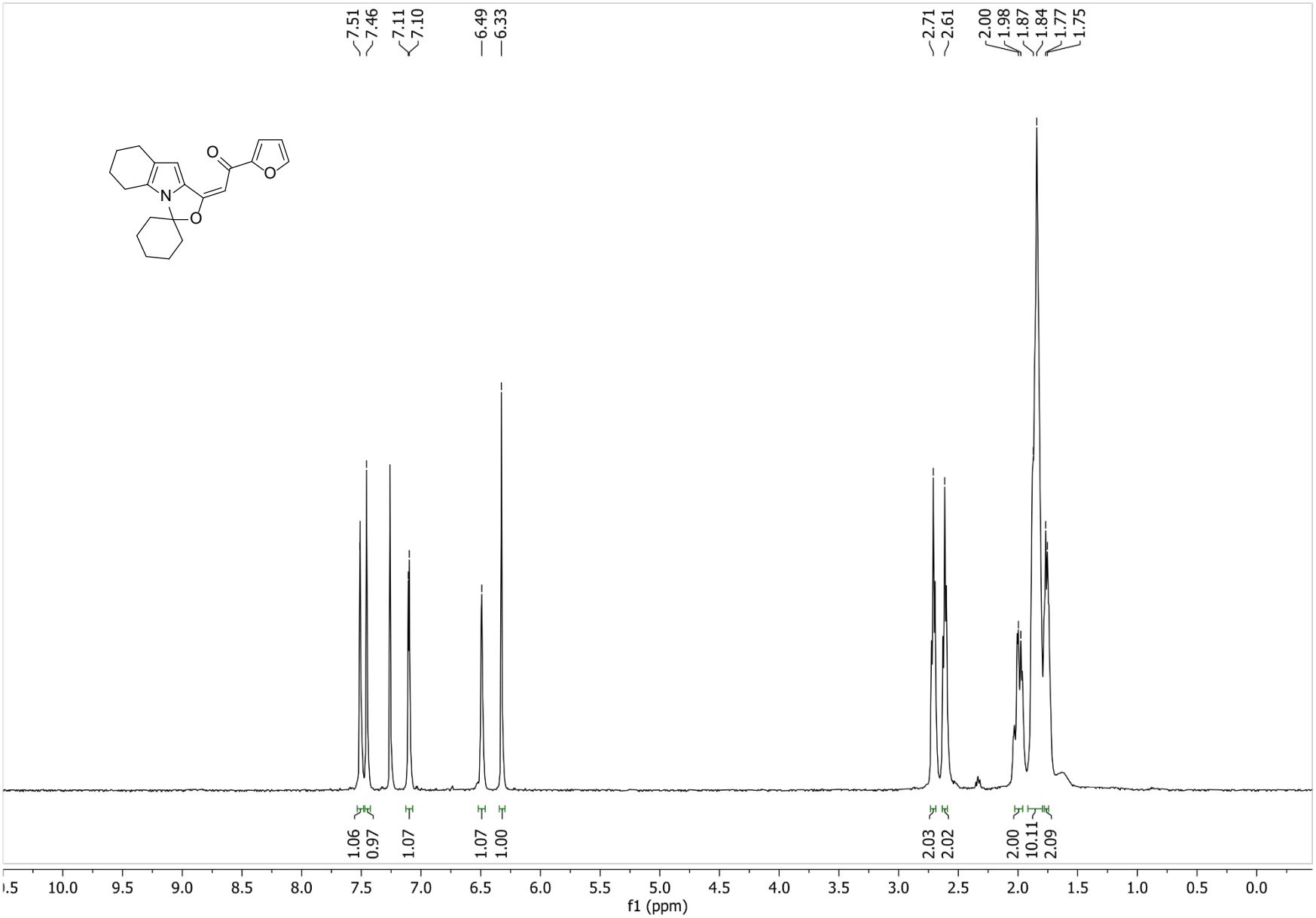
^{1}H NMR spectrum of (*E*)-1-phenyl-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)ethan-1-one (**3fd**) in $CDCl_3$



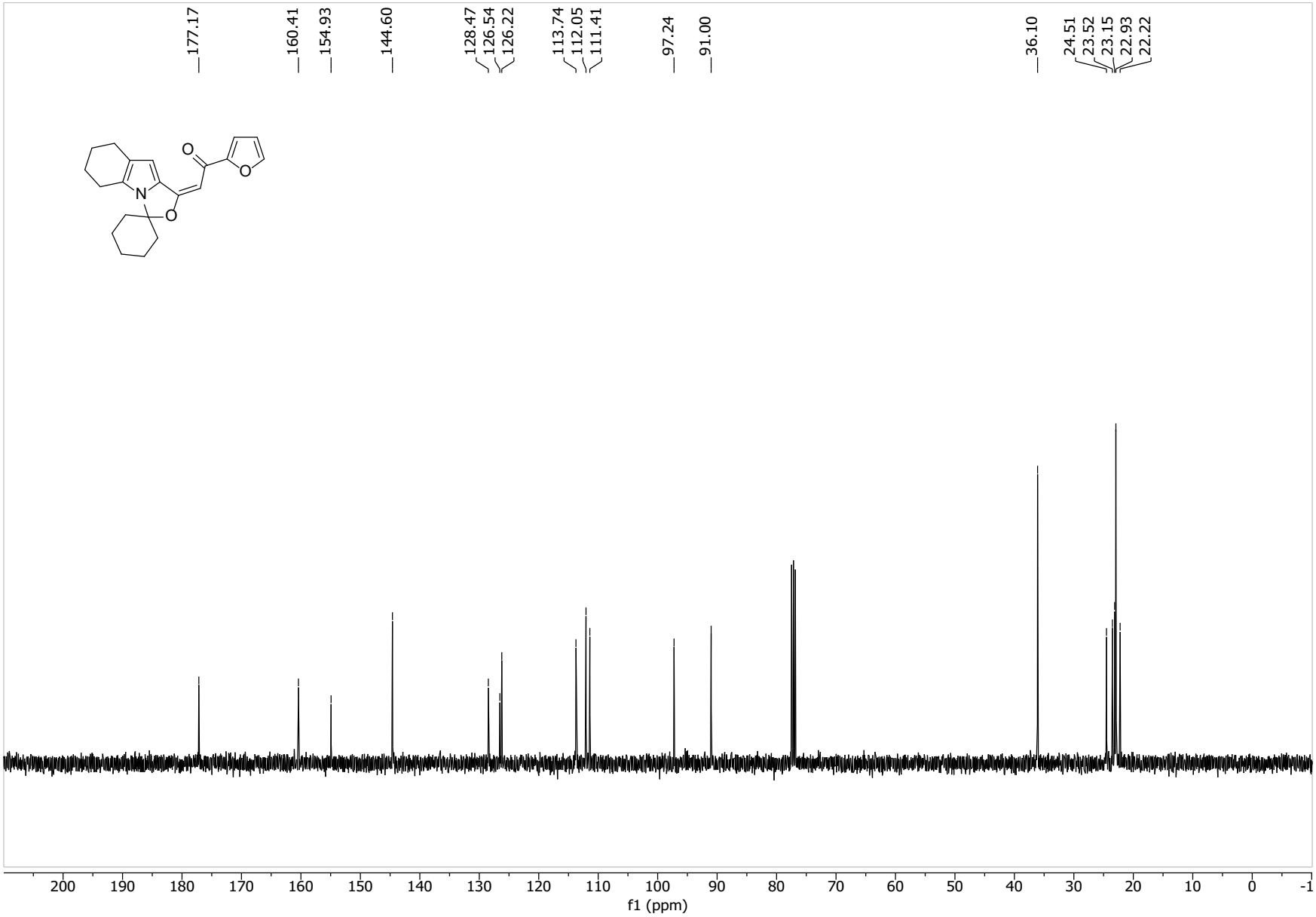
^{13}C NMR spectrum of (*E*)-1-phenyl-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)ethan-1-one (**3fd**) in $CDCl_3$



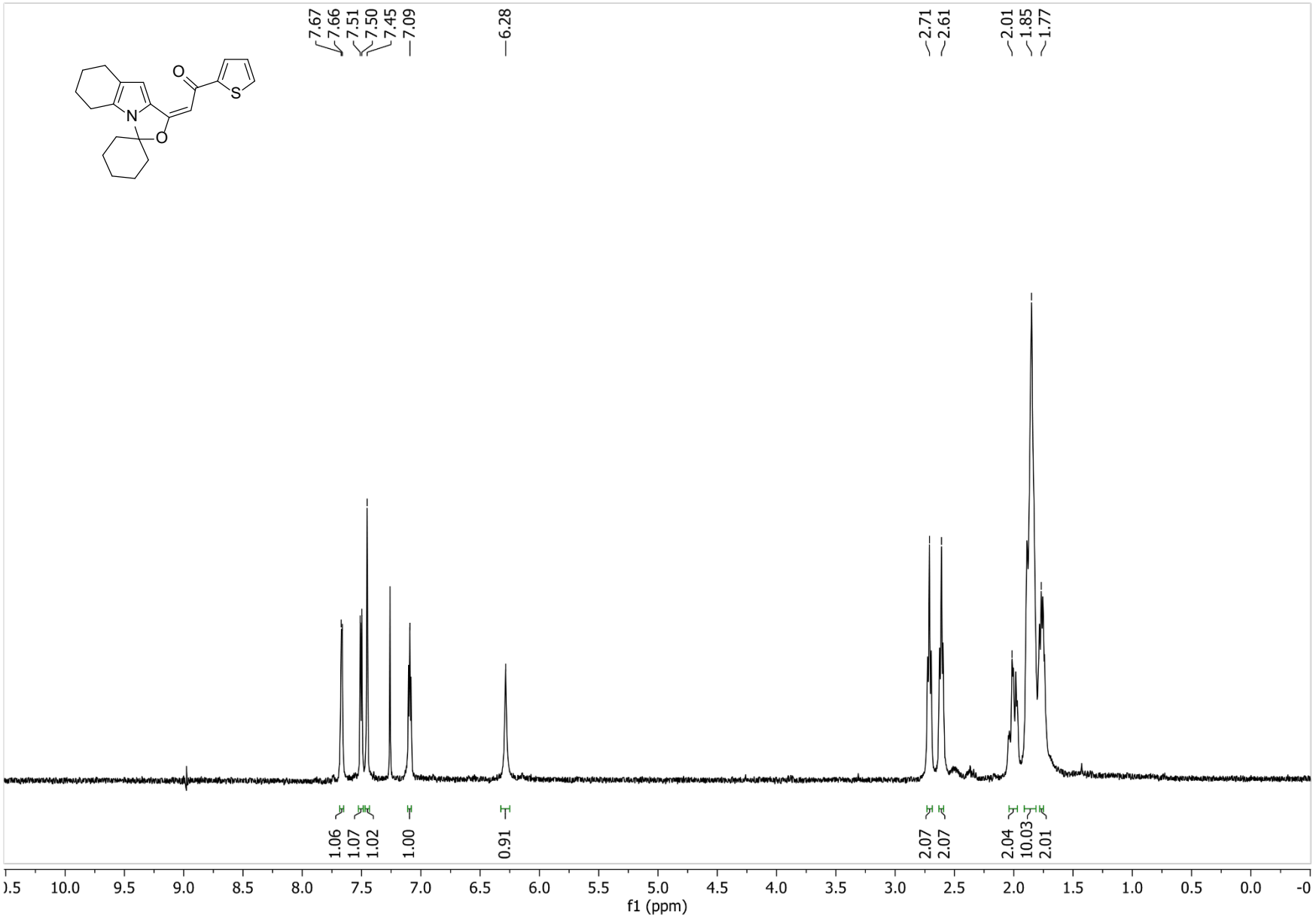
^{1}H NMR spectrum of (*E*)-1-(furan-2-yl)-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)ethan-1-one (**3gd**) in $CDCl_3$



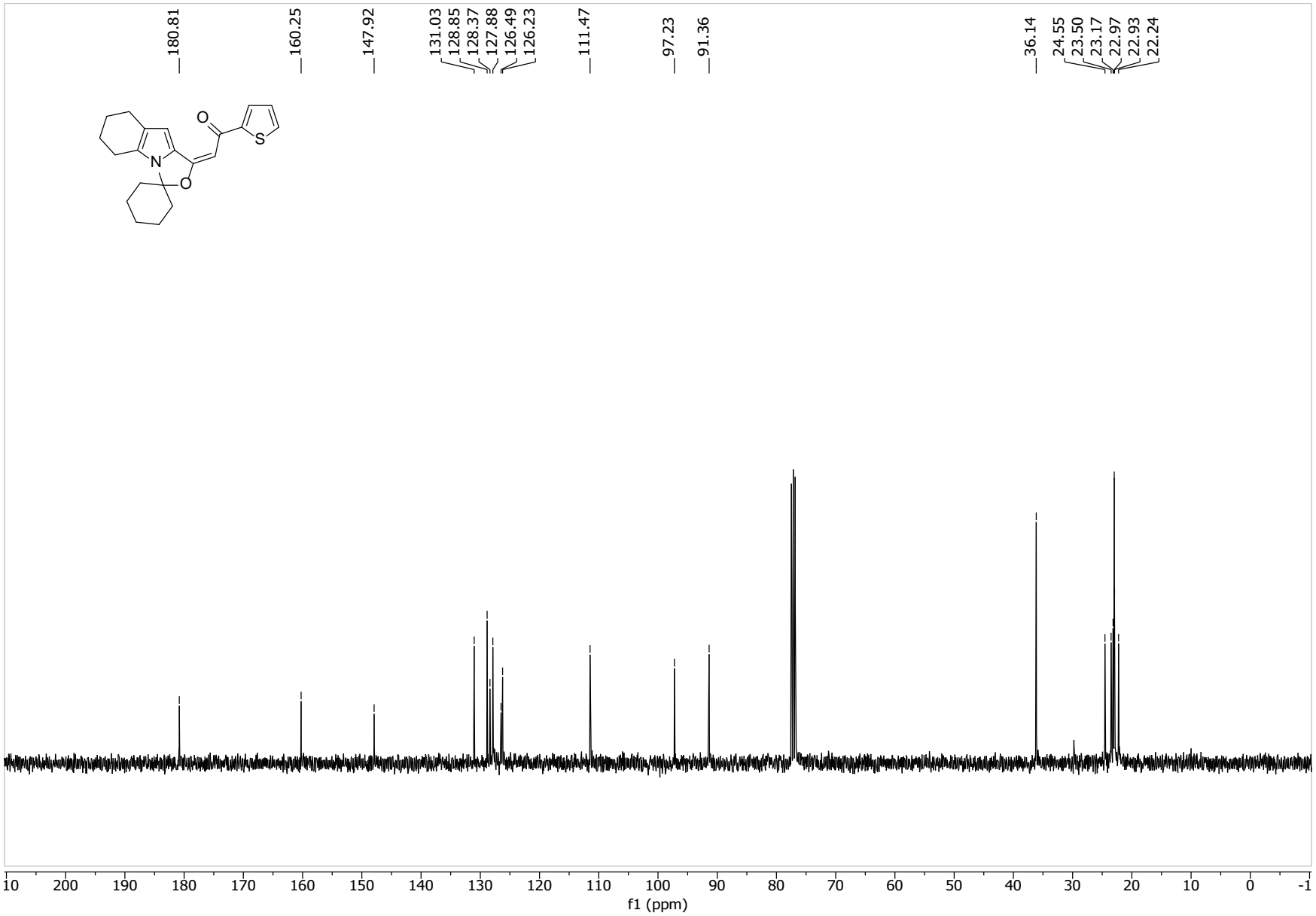
^{13}C NMR spectrum of (*E*)-1-(furan-2-yl)-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1, 3'-oxazolo[3,4-*a*]indol]-1'-ylidene)ethan-1-one (**3gd**) in $CDCl_3$



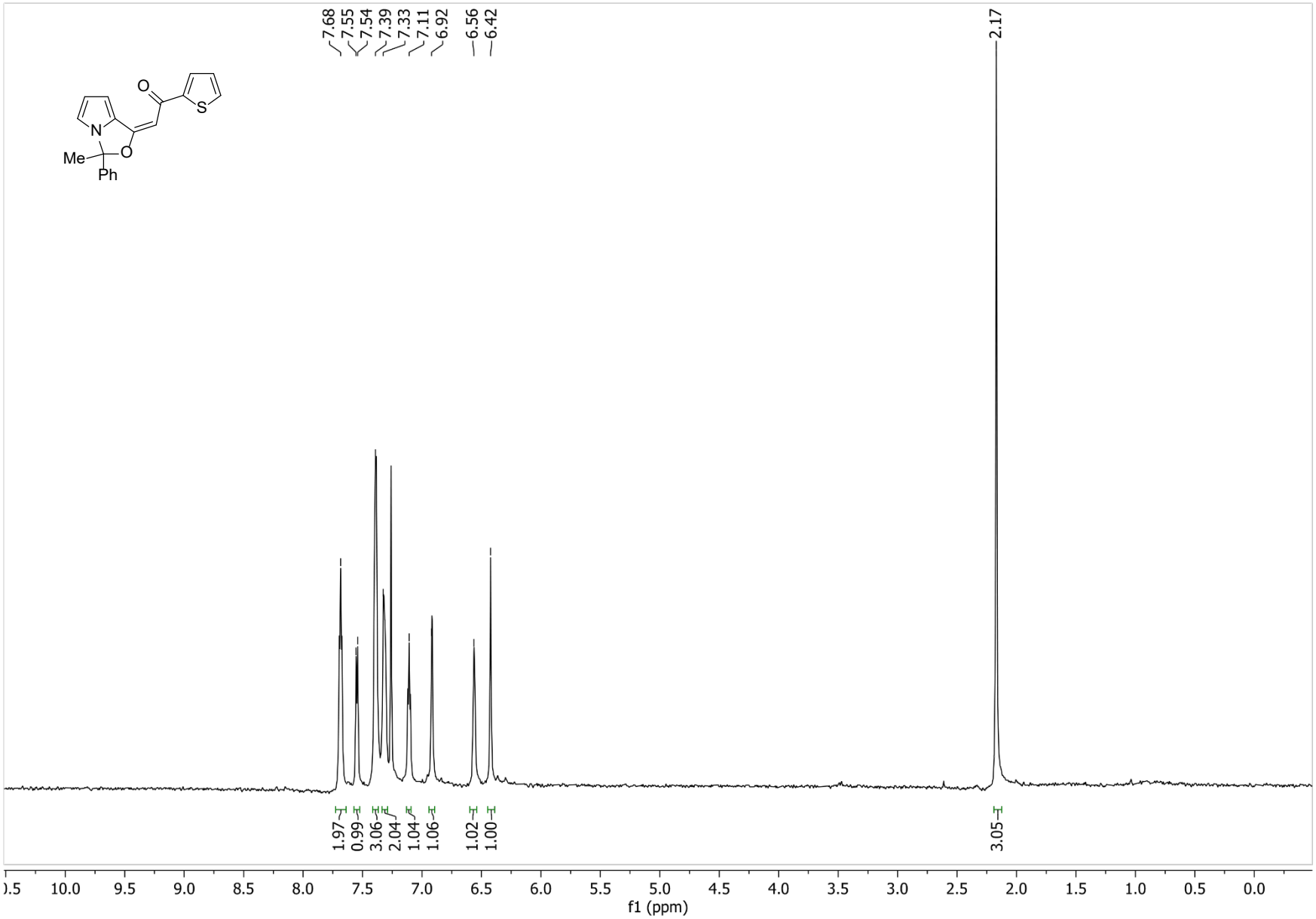
^{1}H NMR spectrum of (*E*)- 2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3hd**) in $CDCl_3$



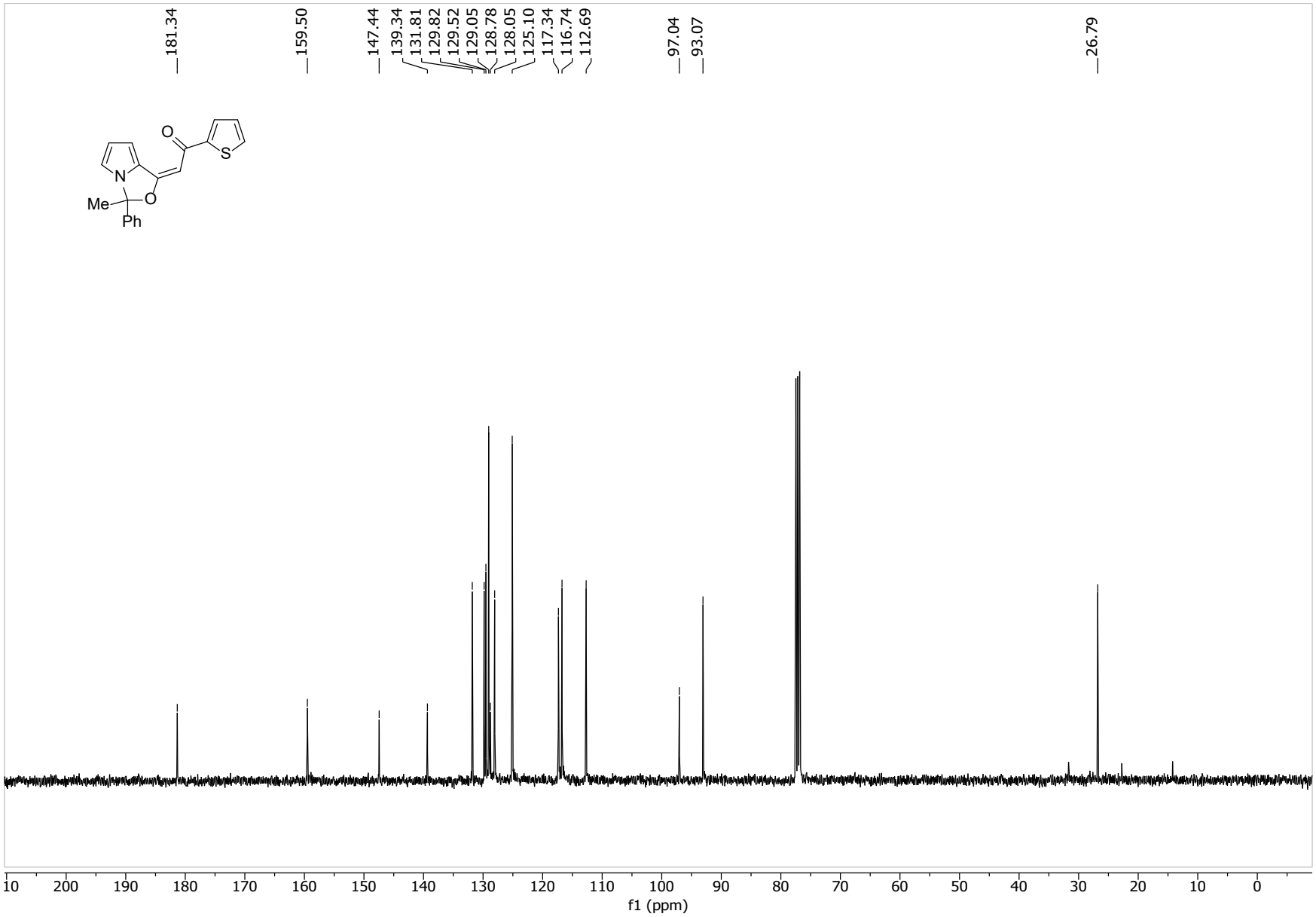
^{13}C NMR spectrum of (*E*)-2-(5',6',7',8'-tetrahydro-1'*H*-spiro[cyclohexane-1,3'-oxazolo[3,4-*a*]indol]-1'-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3hd**) in $CDCl_3$



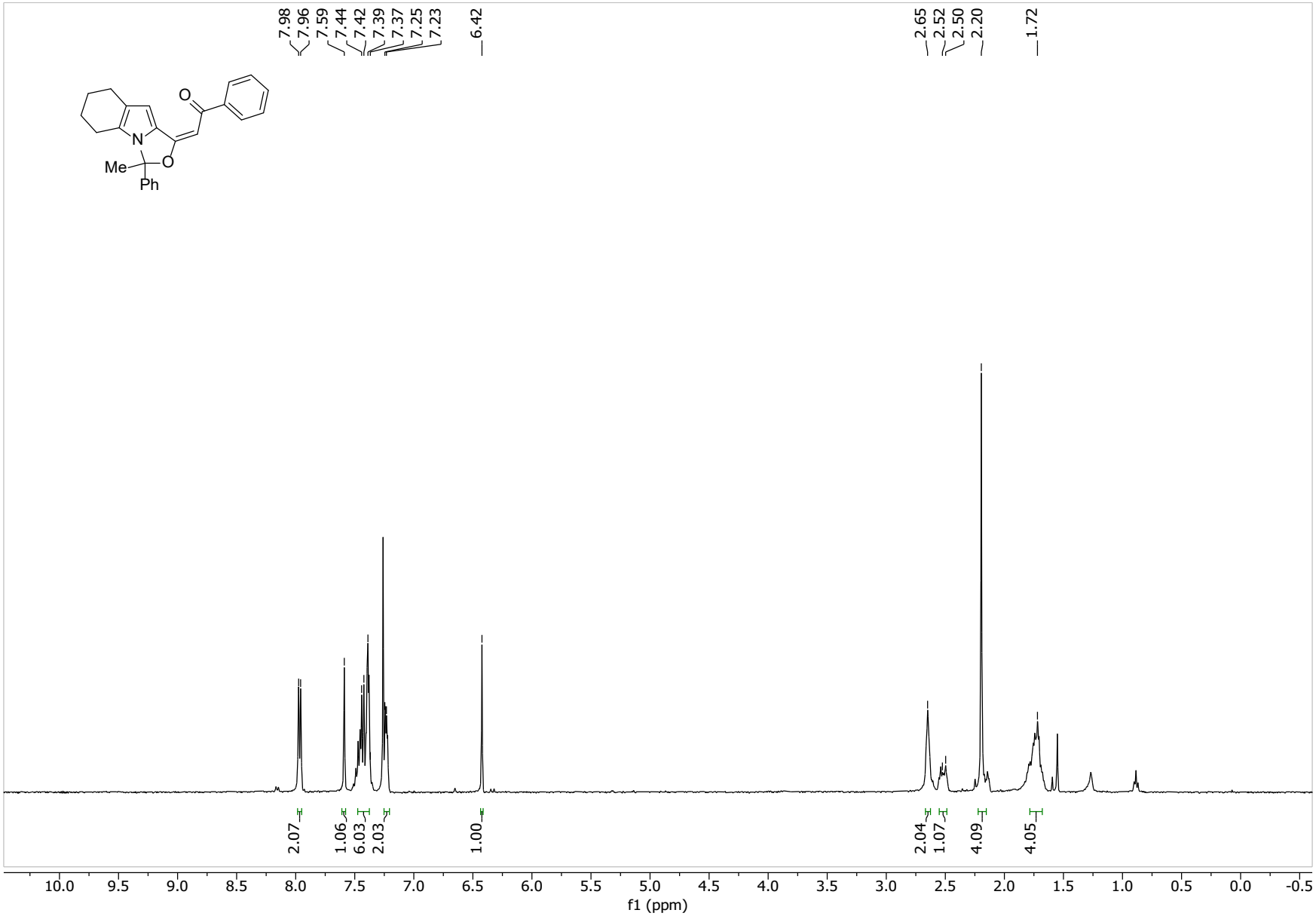
^{1}H NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3bg**) in $CDCl_3$



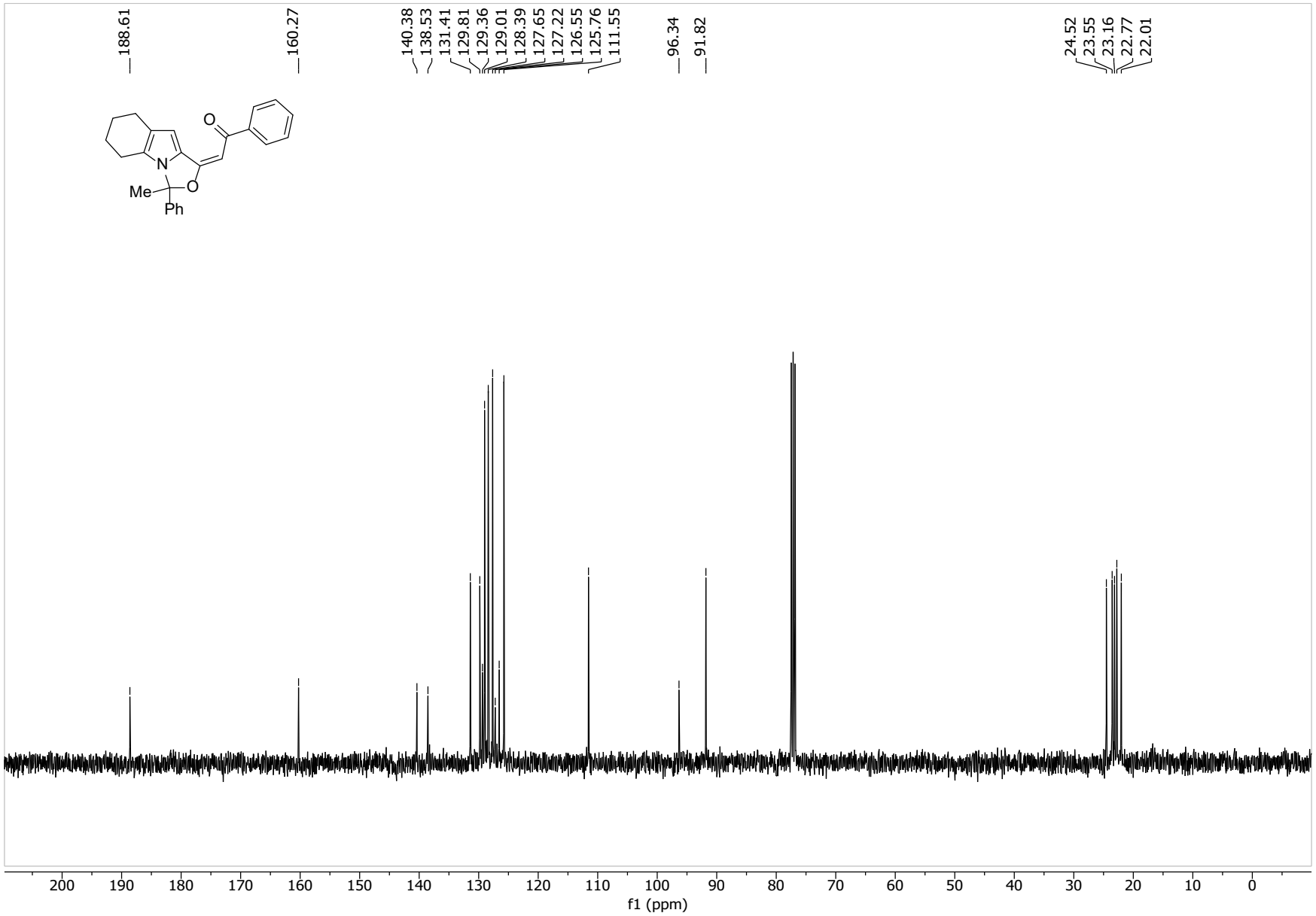
^{13}C NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3bg**) in $CDCl_3$



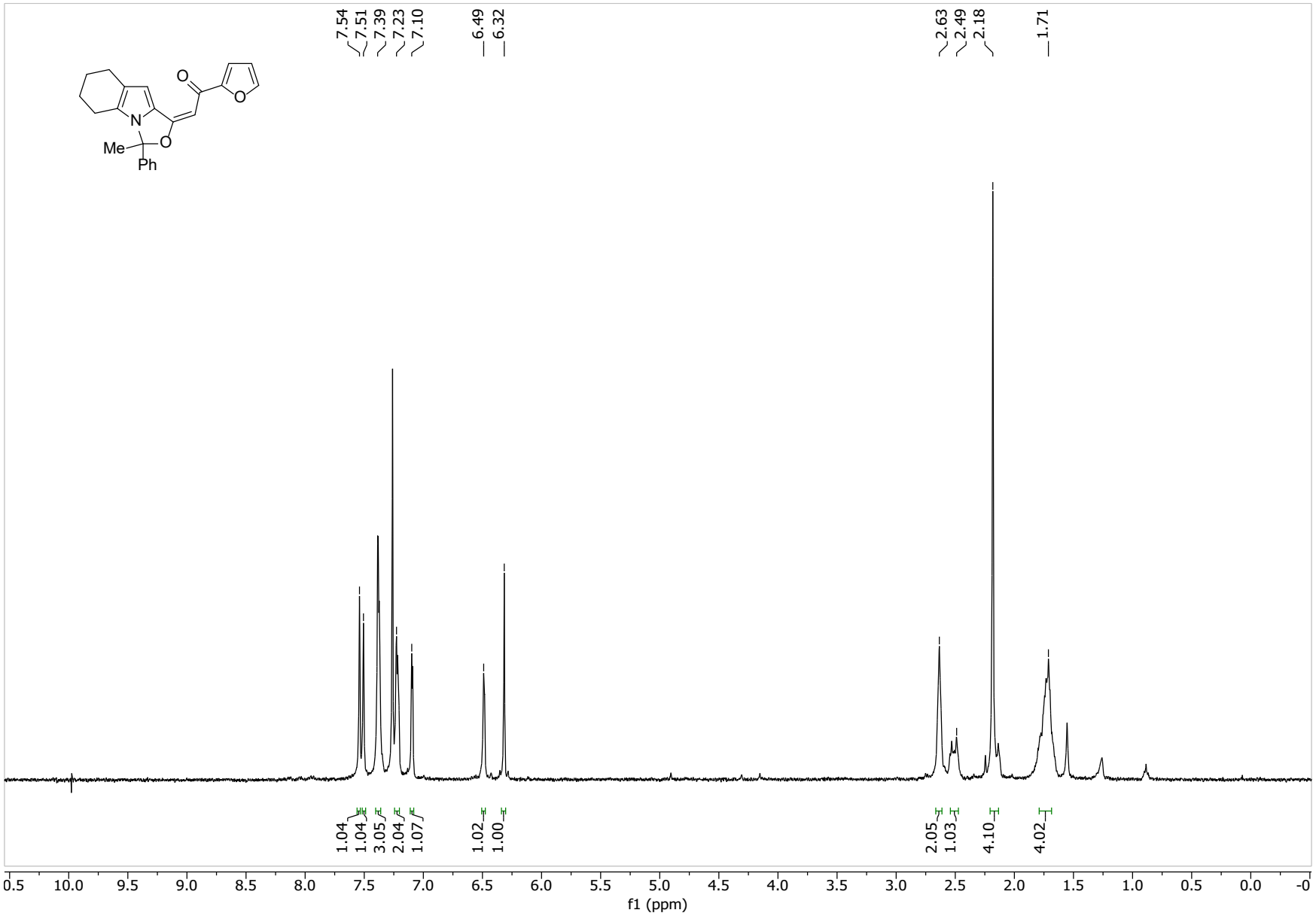
^{1}H NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (**3fg**) in $CDCl_3$



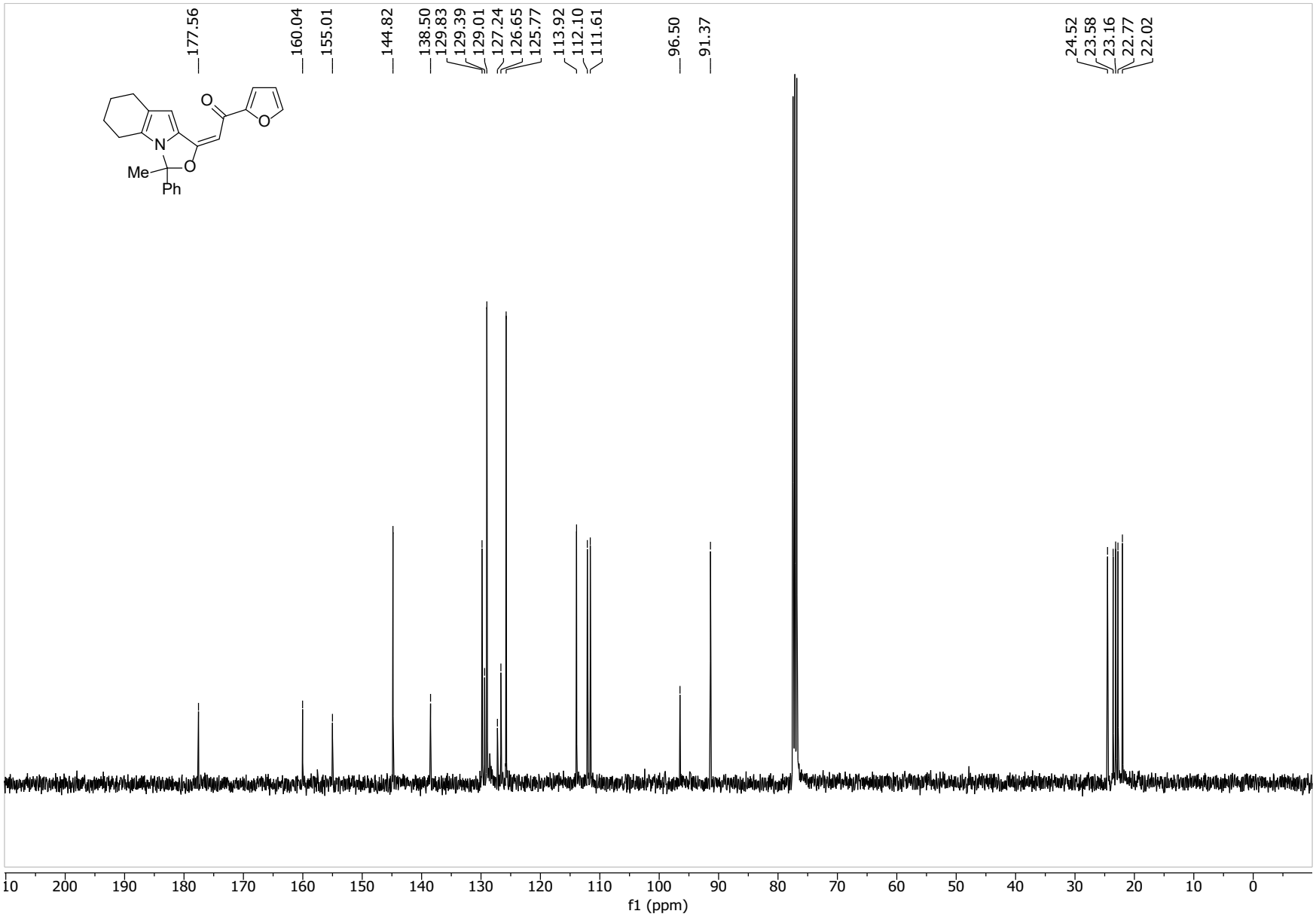
^{13}C NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-phenylethan-1-one (**3fg**) in $CDCl_3$



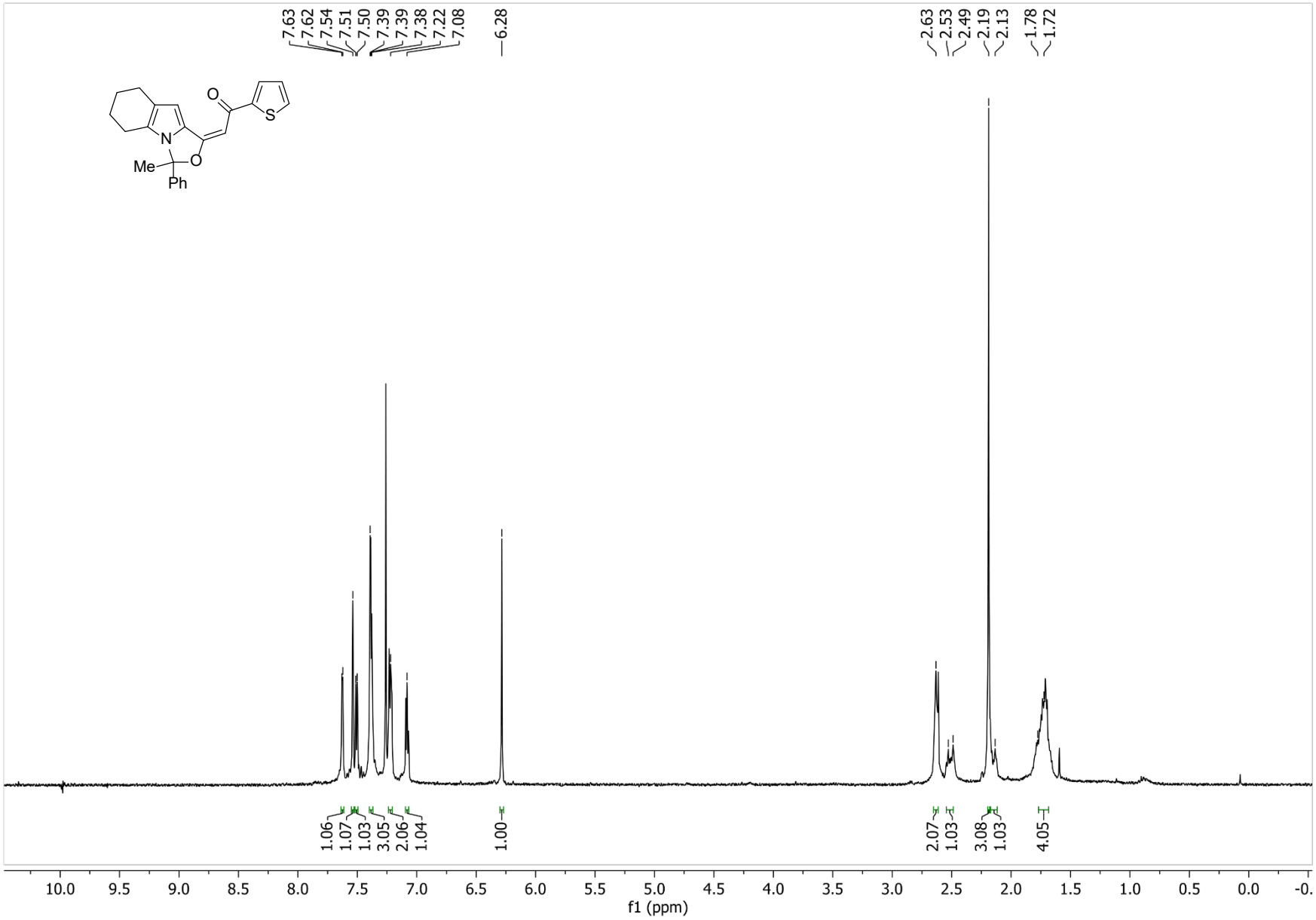
^{1}H NMR spectrum of (*E*)-1-(furan-2-yl)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)ethan-1-one (**3gg**) in $CDCl_3$



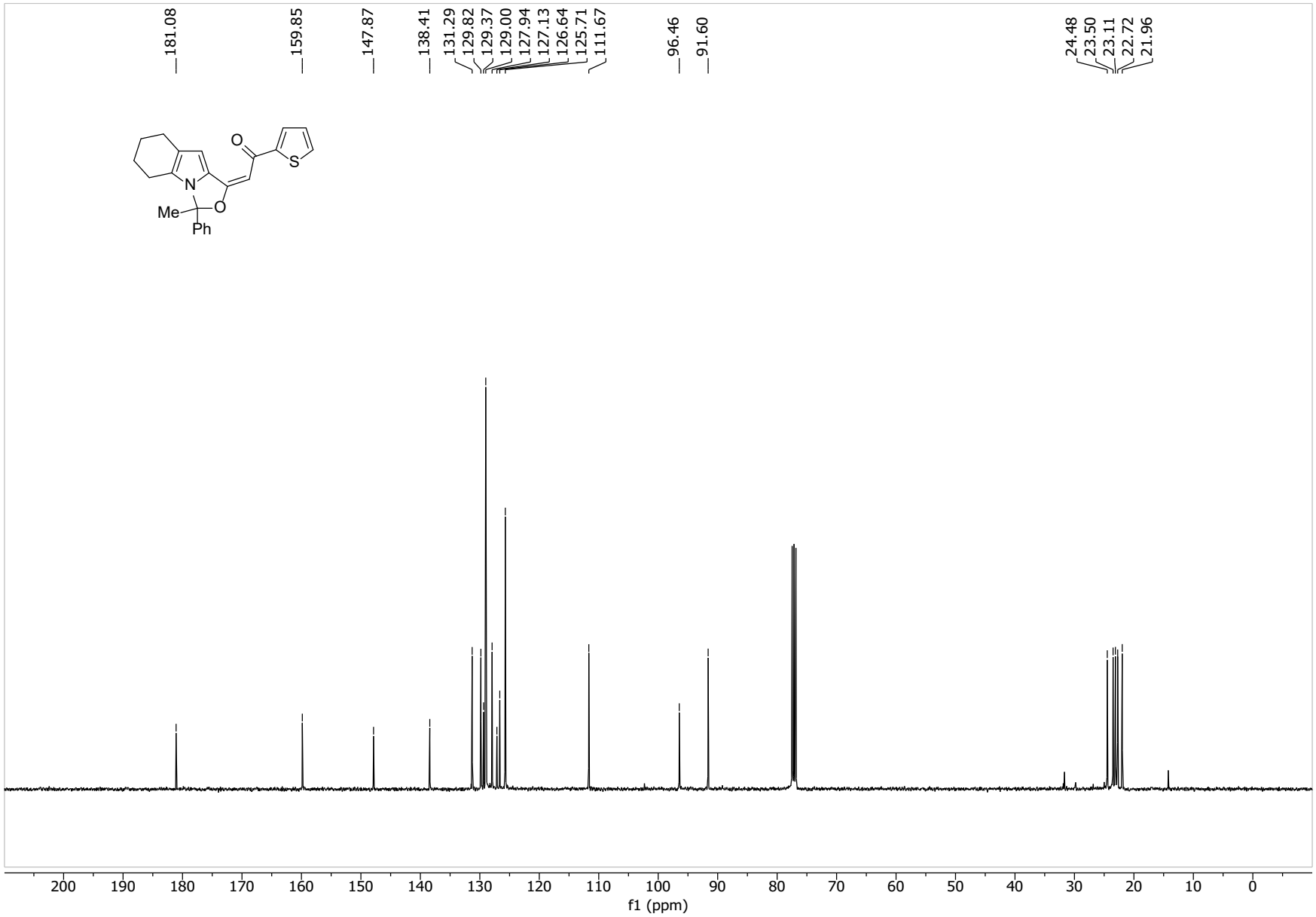
^{13}C NMR spectrum of (*E*)-1-(furan-2-yl)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)ethan-1-one (**3gg**) in $CDCl_3$



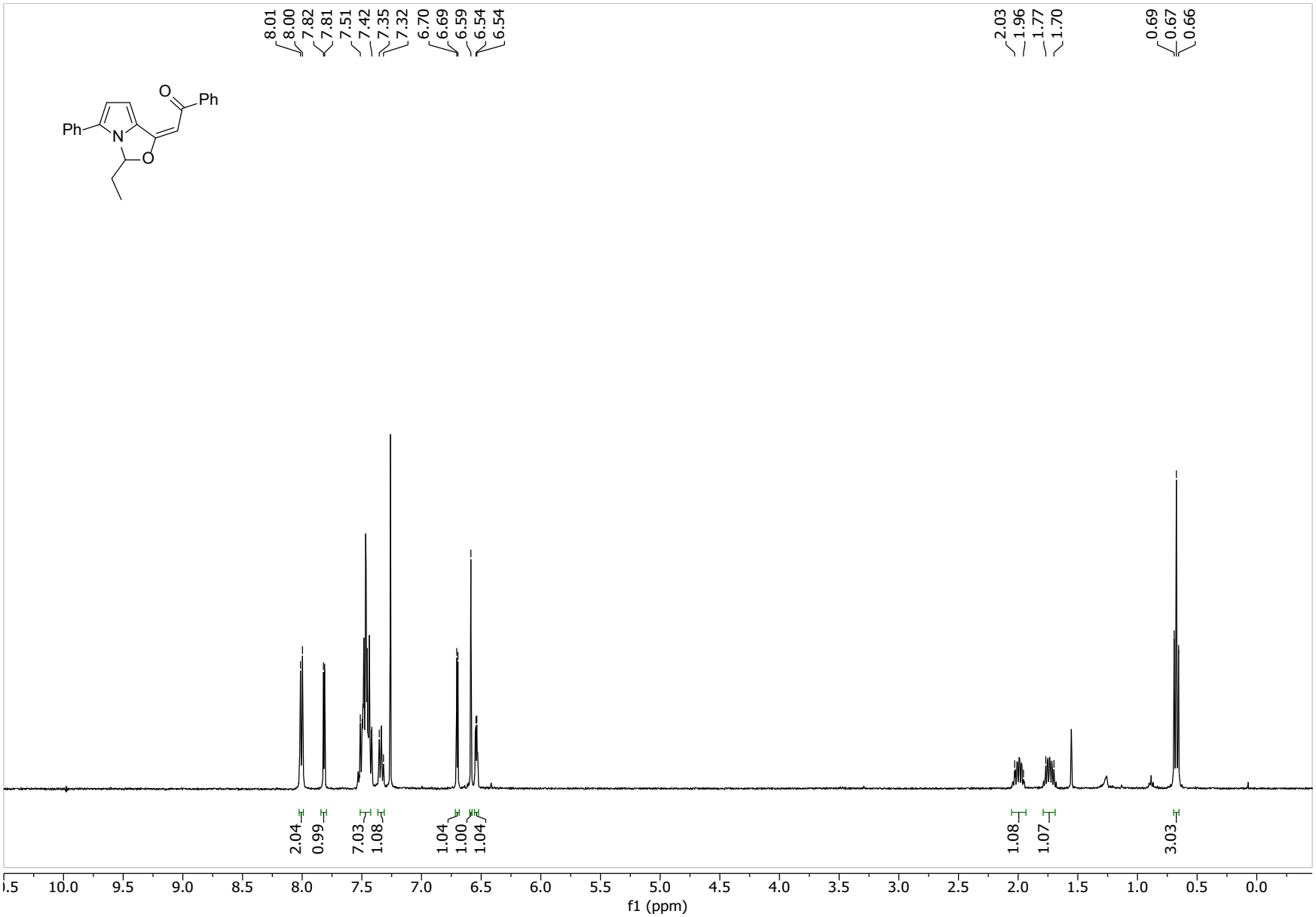
^{1}H NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3hg**) in $CDCl_3$



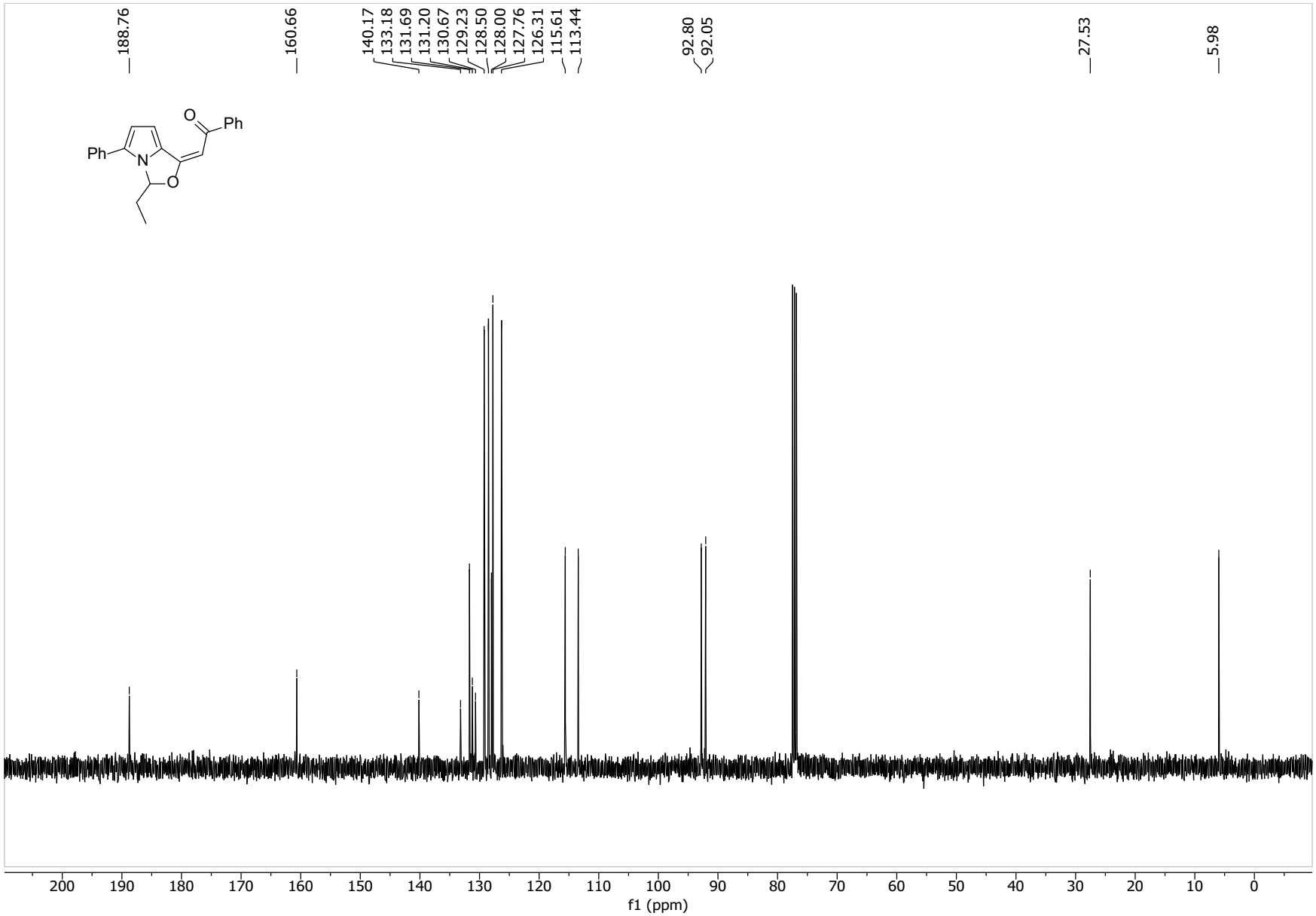
^{13}C NMR spectrum of (*E*)-2-(3-methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*,3*H*-oxazolo[3,4-*a*]indol-1-ylidene)-1-(thiophen-2-yl)ethan-1-one (**3hg**) in $CDCl_3$



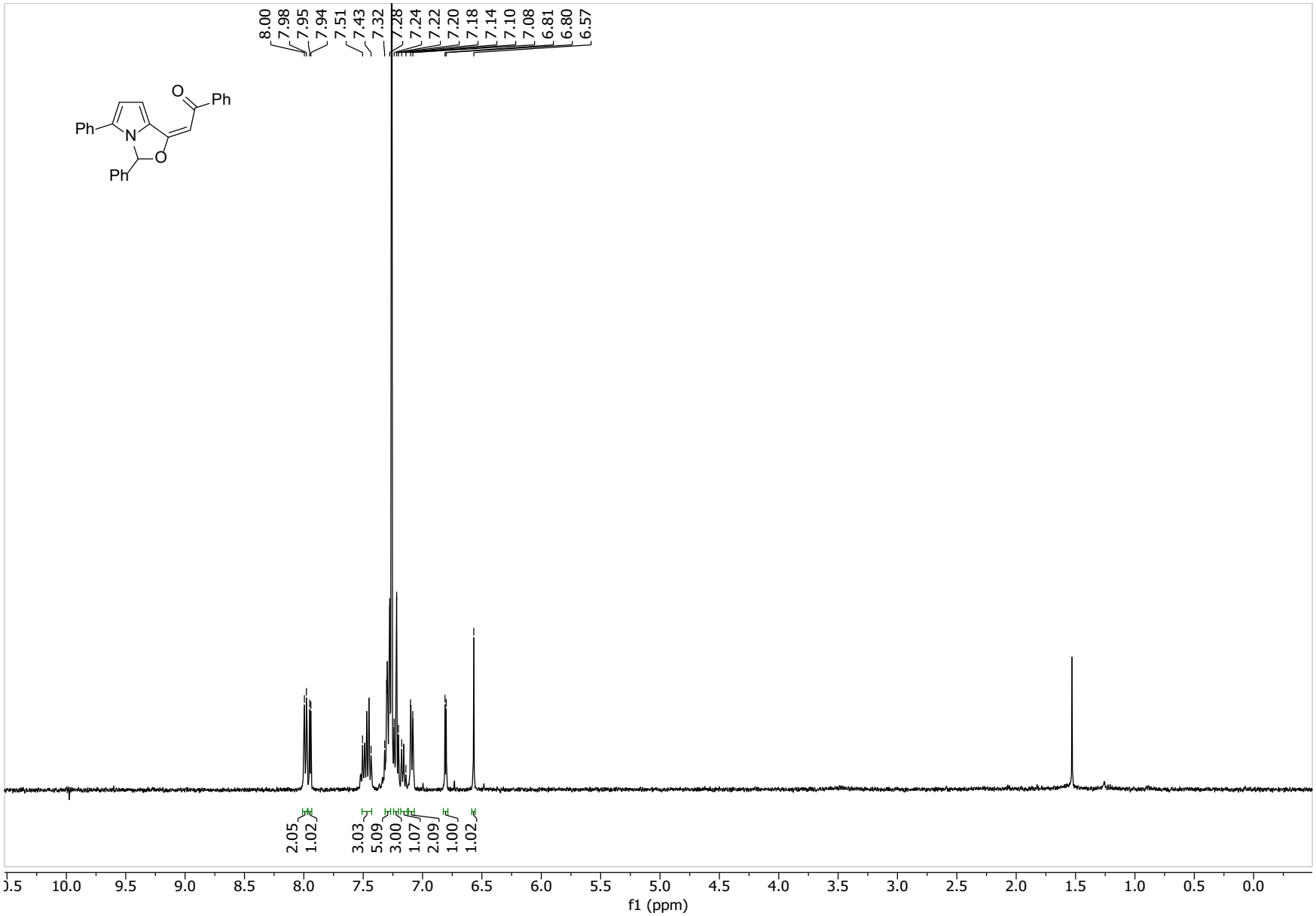
^{1}H NMR spectrum of (*E*)-2-(3-ethyl-5-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ik**) in $CDCl_3$



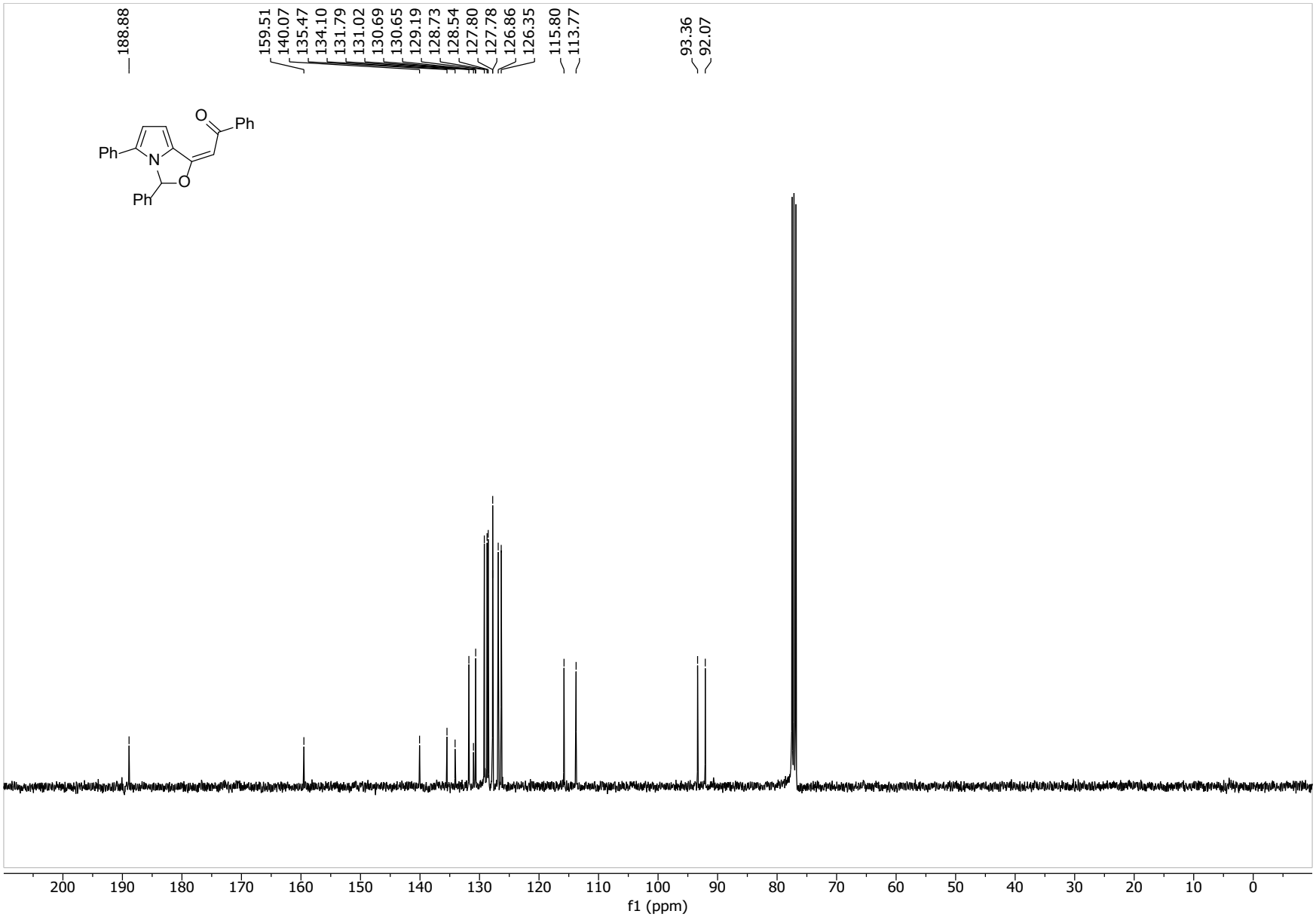
^{13}C NMR spectrum of (*E*)-2-(3-ethyl-5-phenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3ik**) in $CDCl_3$



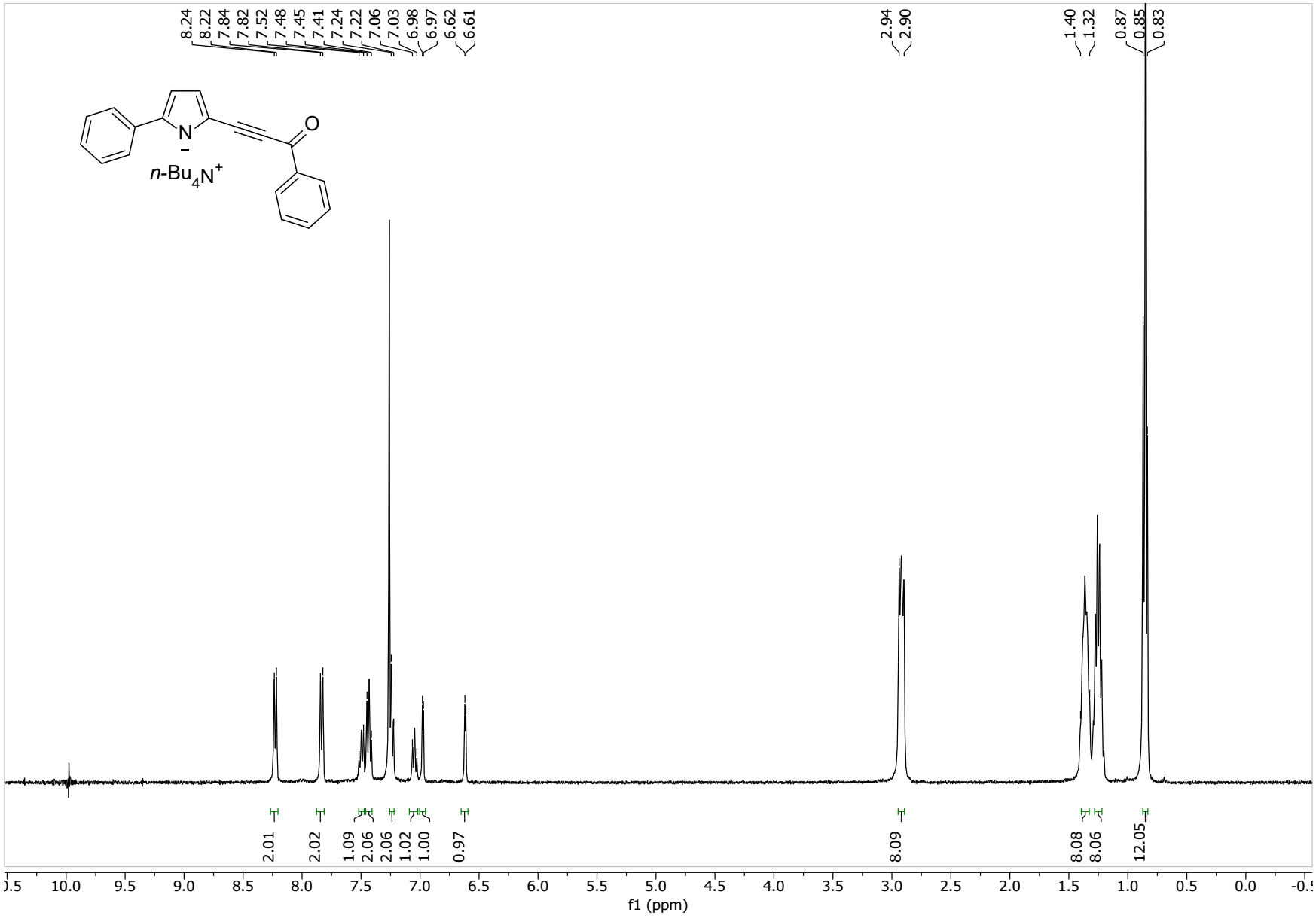
^{1}H NMR spectrum of (*E*)-2-(3,5-diphenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3in**) in $CDCl_3$



^{13}C NMR spectrum of (*E*)-2-(3,5-diphenyl-1*H*,3*H*-pyrrolo[1,2-*c*]oxazol-1-ylidene)-1-phenylethan-1-one (**3in**) in $CDCl_3$



^{1}H NMR spectrum of tetrabutylammonium 2-(3-oxo-3-phenylprop-1-yn-1-yl)-5-phenylpyrrol-1-ide (**4**) in $CDCl_3$



^{13}C NMR spectrum of tetrabutylammonium 2-(3-oxo-3-phenylprop-1-yn-1-yl)-5-phenylpyrrol-1-ide (**4**) in $CDCl_3$