Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information for

Empowering Strategies of Electrochemical N-N Bond Forming Reactions: Direct Access to Previously Neglected 1,2,3-Triazole 1-Oxides

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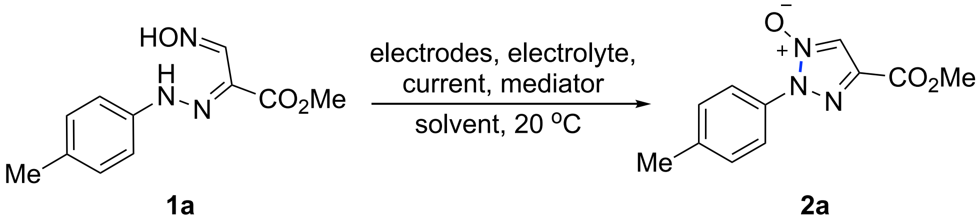
S1. Optimization Studies

Optimization of electrochemical oxidation of a model substrate 1a is presented in Table S1. The found optimal conditions which were used for the synthesis of the entire series of 1,2,3-triazole 1-oxides were as follows: C/Pt electrodes, $LiClO_4$ as an electrolyte, 5 mA current, no mediator and MeOH as a solvent (entry 1). Addition of TEMPO or *N*-hydroxyphthalimide (NHPI) as mediator resulted in a decrease of a yield of compound **2a** (entries 2,3), while utilization of ferrocene or hydroquinone completely suppressed oxidation (entries 4,5). Replacement of MeOH with MeCN slightly decreased the yield of **2a** to 89% (entry 6). At the same time, conducting electrochemical oxidation in MeOH using $^{n}Bu_4NBF_4$ as an electrolyte resulted in more substantial yield decrease (entry 7). Utilization of 1,1,1,3,3,3-hexafluoroisopropanol (HFIP) as one of the widely used reaction media for a variety of electrochemical transformations provided **2a** only in trace amounts (entry 8). Nature of electrodes was also crucial for the successful implementation of electrochemical oxidation: replacement of C/Pt electrodes with glassy carbon/Pt or C/C those resulted in a yield decrease (entries 9,12), while more harsh oxidation conditions derived from stainless steel/Pt or Pt/Pt pairs were useless (entries 10,11). Finally, variation of current from 5 mA to 3 mA or 7 mA slightly decreased the yield of **2a** (entries 13,14).

Table S1. Optimization of electrochemical oxidation of oximinohydrazone **1a**.



Entry	Solvent	Electrodes (+)/(-)	Electrolyte	Current	Mediator	Yield of **2a**,[a] %
1	MeOH	C/Pt	$LiClO_4$	5 mA	-	95
2	MeOH	C/Pt	$LiClO_4$	5 mA	TEMPO	77
3	MeOH	C/Pt	$LiClO_4$	5 mA	NHPI	81
4	MeOH	C/Pt	$LiClO_4$	5 mA	$FeCp_2$	traces[b]
5	MeOH	C/Pt	$LiClO_4$	5 mA	hydroquinone	traces[b]
			Variation of solvent and electrolyte			
6	MeCN	C/Pt	$LiClO_4$	5 mA	-	89
7	MeOH	C/Pt	$^{n}Bu_4NBF_4$	5 mA	-	74
8	HFIP	C/Pt	$^{n}Bu_4NBF_4$	5 mA	-	traces[b]
			Variation of electrodes			
9	MeOH	glassy carbon/Pt	$LiClO_4$	5 mA	-	81
10	MeOH	stainless steel/Pt	$LiClO_4$	5 mA	-	traces[b]
11	MeOH	Pt/Pt	$LiClO_4$	5 mA	-	-[c]
12	MeOH	C/C	$LiClO_4$	5 mA	-	53
			Variation of current			
13	MeOH	C/Pt	$LiClO_4$	3 mA	-	86
14	MeOH	C/Pt	$LiClO_4$	7 mA	-	88

[a] Isolated yields.

[b] Compound **2a** was detected by TLC.

[c] Decomposition of starting oximinohydrazone **1a** was observed.

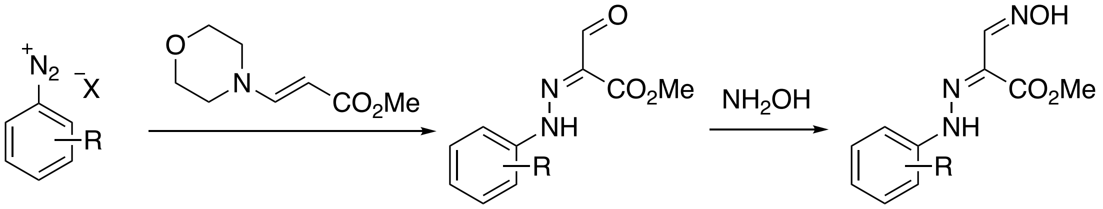
S2. Experimental Section

S2.1 General Remarks

All reactions were carried out in well-cleaned oven-dried glassware with magnetic stirring. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AM-300 (300.13 and 75.47 MHz, respectively) spectrometer and referenced to residual solvent peak. The chemical shifts are reported in ppm (δ); multiplicities are indicated by s (singlet), d (doublet), t (triplet), q (quartet), m (multiplet) and br (broad). Coupling constants, *J*, are reported in Hertz. The IR spectra were recorded on a Bruker "Alpha" spectrometer in the range 400-4000 cm^{-1} (resolution 2 cm^{-1}). Elemental analyses were performed by the CHN Analyzer Perkin-Elmer 2400. High resolution mass spectra were recorded on a Bruker microTOF spectrometer with electrospray ionization (ESI). All measurements were performed in a positive (+MS) ion mode (interface capillary voltage: 4500 V) with scan range *m/z*: 50-3000. External calibration of the mass spectrometer was performed with Electrospray Calibrant Solution (Fluka). A direct syringe injection was used for all analyzed solutions in MeCN (flow rate: 3 μL min^{-1}). Nitrogen was used as nebulizer gas (0.4 bar) and dry gas (4.0 L min^{-1}); interface temperature was set at 180 °C. All spectra were processed by using Bruker DataAnalysis 4.0 software package. The melting points were determined on Stuart SMP20 apparatus and are uncorrected. Analytical thin-layer chromatography (TLC) was carried out on Merck 25 TLC silica gel 60 F$_{254}$ aluminum sheets. The visualization of the TLC plates was accomplished with a UV light. Column chromatography was performed on silica gel 60 A (0.060-0.200 mm, Acros Organics). Thermal analysis was performed using STA 449 F3 (Netzsch) apparatus. Samples of 0.5-1.0 mg mass were poured in alumina pans covered with pierced lids and heated up to 600 °C with a constant rate of 5 K min^{-1}. All solvents were purified and dried using standard methods prior to use. All standard reagents were purchased from Aldrich or Acros Organics and used without further purification.

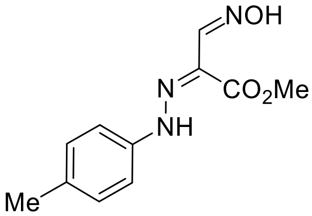
S2.2 Synthesis of Oximinohydrazones

General synthesis of starting oximes **1a-q** was accomplished according to the following scheme:



Corresponding diazonium salt (10 mmol) was added in small portions to a magnetically stirred solution of methyl 4-morpholineacrylate (10 mmol, 1.71 g) in MeCN (10 mL) at 20 °C and left stirred overnight. After the reaction was completed (TLC monitoring), the reaction mixture was diluted with water (20 mL), precipitate formed was filtered off and dried in air. The obtained crude aldehyde was used without further purification for the second step. $NH_2OH·HCl$ (15 mmol, 1.04 g) was added to a magnetically stirred suspension of the corresponding crude aldehyde in a mixture of AcONa (25 mmol, 2.05 g) in EtOH (15 mL) at 20 °C and the reaction mixture was left stirring overnight. After the reaction was completed (TLC monitoring), the reaction mixture was diluted with water (30 mL), precipitate formed was filtered off and dried in air. Crude residue was purified via recrystallization from EtOH to afford pure oxime **1a-q**.

Methyl 3-(hydroxyimino)-2-(2-(p-tolyl)hydrazinylidene)propanoate (1a).



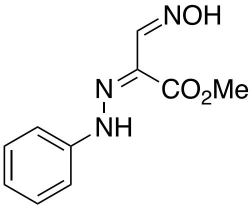
Yield 1.93 g (82%). Orange powder. M.p. 165-167°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 2.29 (s, 3H), 3.78 (s, 3H), 7.16-7.23 (m, 4H), 8.30 (s, 1H), 12.03 (s, 1H), 12.65 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 20.9, 52.4, 115.2, 122.3, 130.5, 133.4, 140.5, 145.0, 164.7.

Calcd for $C_{11}H_{13}N_3O_3$ (%): C, 56.16; H, 5.57; N, 17.86. Found (%): C, 56.03; H, 5.69; N, 17.62.

Methyl 3-(hydroxyimino)-2-(2-phenylhydrazinylidene)propanoate (1b).



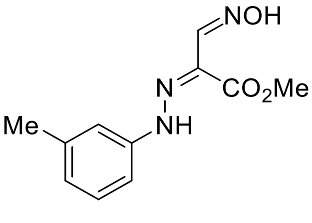
Yield 1.86 g (84%). Pale yellow powder. M.p. 164-166°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.79 (s, 3H), 7.10 (t, J = 7.2 Hz, 1H), 7.28 (d, J = 7.7 Hz, 2H), 7.41 (t, J = 7.7 Hz, 2H), 8.30 (s, 1H), 12.10 (br s, 1H), 12.65 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.5, 115.2, 122.9, 124.2, 130.1, 142.8, 145.0, 164.6.

Calcd for $C_{10}H_{11}N_3O_3$ (%): C, 54.30; H, 5.01; N, 19.00. Found (%): C, 54.47; H, 4.93; N, 18.81.

Methyl 3-(hydroxyimino)-2-(2-(3-methylphenyl)hydrazinylidene)propanoate (1c).



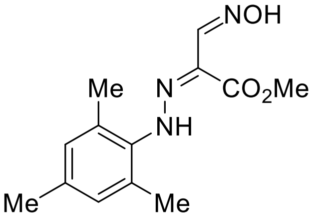
Yield 1.62 g (69%). Pale yellow powder. M.p. 183-184°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 2.33 (s, 3H), 3.79 (s, 3H), 6.92 (d, J = 7.7 Hz, 1H), 7.03-7.11 (m, 2H), 7.28 (t, J = 7.7 Hz, 1H), 8.30 (s, 1H), 12.11 (br s, 1H), 12.66 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 21.6, 52.5, 112.5, 115.6, 122.8, 125.0, 129.9, 139.6, 142.7, 145.0, 164.7.

Calcd for $C_{11}H_{13}N_3O_3$ (%): C, 56.16; H, 5.57; N, 17.86. Found (%): C, 56.37; H, 5.44; N, 17.70.

Methyl 3-(hydroxyimino)-2-(2-mesitylhydrazinylidene)propanoate (1d).



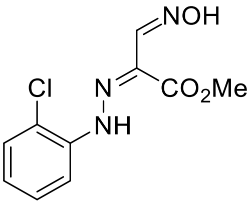
Yield 2.05 g (78%). Yellow powder. M.p. 150-152°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d_6): δ = 2.23 (s, 3H), 2.30 (s, 6H), 3.74 (s, 3H), 6.92 (s, 2H), 8.31 (s, 1H), 11.94 (br. s, 1H), 12.45 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 19.4, 20.8, 52.3, 121.5, 128.7, 130.3, 134.3, 136.8, 144.9, 165.0.

Calcd for $C_{13}H_{17}N_3O_3$ (%): C, 59.30; H, 6.51; N, 15.96. Found (%): C, 59.46; H, 6.42; N, 16.09.

Methyl 3-(hydroxyimino)-2-(2-(2-chlorophenyl)hydrazinylidene)propanoate (1e).



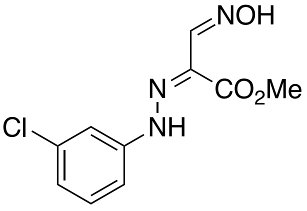
Yield 1.79 g (70%). Pale yellow powder. M.p. 156-157°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d_6): δ = 3.81 (s, 3H), 7.11 (t, *J* = 7.7 Hz, 1H), 7.43 (t, *J* = 8.0 Hz, 1H), 7.51 (d, *J* = 8.0 Hz, 1H), 7.72 (d, *J* = 8.2 Hz, 1H), 8.32 (s, 1H), 12.30 (br s, 1H), 12.97 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 52.7, 115.5, 119.6, 124.6, 125.2, 129.0, 130.0, 139.5, 144.7, 164.4.

Calcd for $C_{10}H_{10}ClN_3O_3$ (%): C, 46.98; H, 3.94; N, 16.44. Found (%): C, 47.12; H, 3.81; N, 16.20.

Methyl 3-(hydroxyimino)-2-(2-(3-chlorophenyl)hydrazinylidene)propanoate (1f).



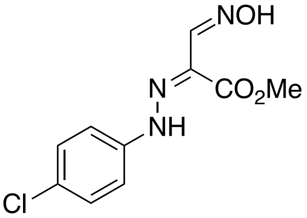
Yield 1.89 g (74%). Pale yellow powder. M.p. 144-145°C (dec.) (EtOH).

^{1}H NMR (300 MHz, DMSO-d_6): δ = 3.79 (s, 3H), 7.10-7.17 (m, 3H), 7.32 (s, 1H), 7.39 (t, *J* = 7.7 Hz, 1H), 8.27 (s, 1H), 12.16 (br s, 1H), 12.59 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 52.6, 114.0, 114.6, 123.6, 124.1, 131.7, 134.6, 144.3, 144.7, 164.4.

Calcd for $C_{10}H_{10}ClN_3O_3$ (%): C, 46.98; H, 3.94; N, 16.44. Found (%): C, 46.84; H, 4.07; N, 16.31.

Methyl 3-(hydroxyimino)-2-(2-(4-chlorophenyl)hydrazinylidene)propanoate (1g).



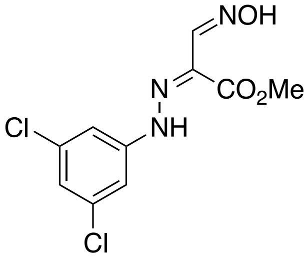
Yield 2.15 g (84%). Pale yellow powder. M.p. 138-139°C (dec.) (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.79 (s, 3H), 7.28 (d, *J* = 8.7 Hz, 2H), 7.45 (d, *J* = 8.7 Hz, 2H), 8.28 (s, 1H), 12.15 (br s, 1H), 12.61 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.6, 116.8, 123.6, 127.7, 130.0, 141.8, 144.9, 164.5.

Calcd for $C_{10}H_{10}ClN_3O_3$ (%): C, 46.98; H, 3.94; N, 16.44. Found (%): C, 46.79; H, 4.03; N, 16.62.

Methyl 3-(hydroxyimino)-2-(2-(3,5-dichlorophenyl)hydrazinylidene)propanoate (1h).



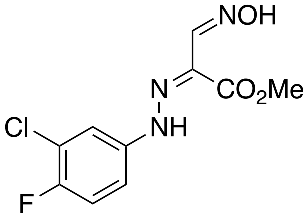
Yield 1.89 g (65%). Pale yellow powder. M.p. 182-183°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.81 (s, 3H), 7.27 (s, 3H), 8.26 (s, 1H), 12.18 (s, 1H), 12.49 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.8, 113.6, 122.7, 125.3, 135.4, 144.4, 145.3, 164.2.

Calcd for $C_{10}H_9Cl_2N_3O_3$ (%): C, 41.40; H, 3.13; N, 14.49. Found (%): C, 41.22; H, 3.27; N, 14.21.

Methyl 3-(hydroxyimino)-2-(2-(3-chloro-4-fluorophenyl)hydrazinylidene)propanoate (1i).



Yield 2.38 g (87%). Pale pink solid. M.p. 191-192°C (dec.) (EtOH).

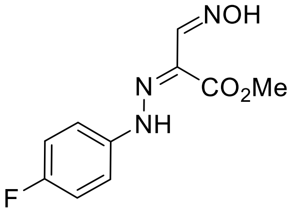
IR (KBr): 3247, 3089, 3002, 2956, 2905, 2849, 1701, 1607, 1569, 1506, 1440, 1336, 1294, 1261, 1211, 1114, 1049, 996, 902, 811, 771 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.79 (s, 3H), 7.20-7.22 (m, 1H), 7.41-7.47 (m, 3H), 8.26 (s, 1H), 12.13 (br s, 1H), 12.55 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.6, 115.5 (d, *J* = 7.2 Hz), 116.4, 118.3 (d, *J* = 22.5 Hz), 120.9 (d, *J* = 19.0 Hz), 124.0 (d, *J* = 1.7 Hz), 140.2 (d, *J* = 2.7 Hz), 144.7, 154.0 (d, *J* = 242.6 Hz), 164.4.

HRMS (ESI): *m/z* calcd for $C_{10}H_{10}{}^{35}ClFN_3O_3$: 274.0389; found: 274.0382 [M+H]$^+$.

Methyl 2-(2-(4-fluorophenyl)hydrazinylidene)-3-(hydroxyimino)propanoate (1j).



Yield 1.92 g (80%). Light yellow powder. M.p. 192-193°C (dec.) (EtOH).

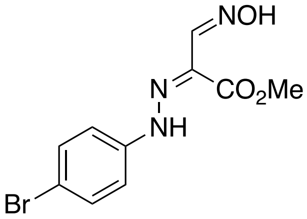
IR (KBr): 3247, 3089, 3002, 2956, 2905, 2849, 1701, 1607, 1569, 1506, 1440, 1336, 1294, 1261, 1211, 1114, 1049, 996, 956, 902, 811, 771 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.78 (s, 3H), 7.21-7.32 (m, 4H), 8.29 (s, 1H), 12.13 (br s, 1H), 12.64 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.5, 116.6 (d, *J* = 5.4 Hz), 116.8 (d, *J* = 9.4 Hz), 122.9, 139.3 (d, *J* = 2.3 Hz), 144.9, 159.1 (d, *J* = 240.1 Hz), 164.6.

Calcd for $C_{10}H_{10}FN_3O_3$ (%): C, 50.21; H, 4.21; N, 17.57. Found (%): C, 50.39; H, 4.13; N, 17.39.

Methyl 3-(hydroxyimino)-2-(2-(4-bromophenyl)hydrazinylidene)propanoate (1k).



Yield 2.35 g (78%). Yellow powder. M.p. 199-200°C (EtOH).

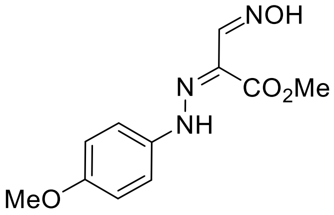
IR (KBr): 3268, 3027, 3000, 2951, 1695, 1592, 1559, 1474, 1434, 1335, 1239, 1187, 1106, 1067, 989, 938, 826, 810, 776 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.79 (s, 3H), 7.23 (d, 2H, *J* = 8.7 Hz), 7.57 (d, 2H, *J* = 8.7 Hz), 8.28 (s, 1H), 12.18 (s, 1H), 12.64 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.6, 115.7, 117.1, 123.6, 132.8, 142.2, 144.9, 164.5.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_{11}{}^{79}BrN_3O_3$: 299.9978; found: 299.9979 [M+H]$^+$.

Methyl 3-(hydroxyimino)-2-(2-(4-methoxyphenyl)hydrazinylidene)propanoate (1l).



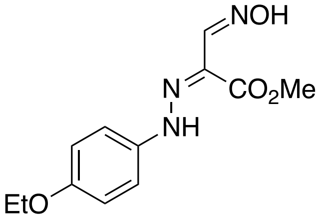
Yield 2.13 g (85%). Yellow powder. M.p. 200-201°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.76 (s, 3H), 3.77 (s, 3H), 7.00 (d, 2H, *J* = 9.0 Hz), 7.24 (d, 2H, *J* = 9.0 Hz), 8.30 (s, 1H), 11.96 (s, 1H), 12.68 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.1, 52.5, 115.7, 116.8, 121.7, 136.3, 145.0, 155.6, 164.9.

Calcd for $C_{11}H_{13}N_3O_4$ (%): C, 52.59; H, 5.22; N, 16.73. Found (%): C, 52.76; H, 5.09; N, 16.55.

Methyl 3-(hydroxyimino)-2-(2-(4-ethoxyphenyl)hydrazinylidene)propanoate (1m).



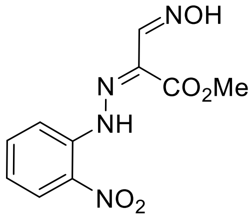
Yield 2.34 g (88%). Orange powder. M.p. 188-189°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 1.32 (t, *J* = 6.9 Hz, 3H), 3.77 (s, 3H), 4.01 (q, *J* = 6.9 Hz, 2H), 6.97 (d, *J* = 8.7 Hz, 2H), 7.21 (d, *J* = 8.7 Hz, 2H), 8.29 (s, 1H), 12.00 (s, 1H), 12.68 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 15.1, 52.3, 63.7, 115.9, 116.6, 121.6, 136.2, 145.1, 155.8, 164.8.

Calcd for $C_{12}H_{15}N_3O_4$ (%): C, 54.33; H, 5.70; N, 15.84. Found (%): C, 54.21; H, 5.79; N, 15.68.

Methyl 3-(hydroxyimino)-2-(2-(2-nitrophenyl)hydrazinylidene)propanoate (1n).



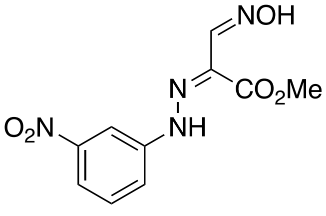
Yield 2.02 g (76%). Yellow powder. M.p. 188-189°C (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.89 (s, 3H), 7.18 (t, J = 7.8 Hz, 1H), 7.77 (t, J = 7.8 Hz, 1H), 7.96-7.98 (m, 2H), 8.21 (d, J = 8.3 Hz, 1H), 11.71 (s, 1H), 13.63 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.1, 116.4, 122.2, 126.2, 129.6, 133.7, 136.9, 139.4, 145.4, 162.1.

Calcd for $C_{10}H_{10}N_4O_5$ (%): C, 45.12; H, 3.79; N, 21.05. Found (%): C, 44.95; H, 3.92; N, 20.89.

Methyl 3-(hydroxyimino)-2-(2-(2-nitrophenyl)hydrazinylidene)propanoate (1o).



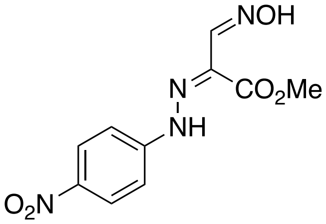
Yield 2.16 g (81%). Yellow powder. M.p. 177-178°C (dec.) (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.82 (s, 3H), 7.66 (br s, 2H), 7.90 (br s, 1H), 8.06 (s, 1H), 8.29 (s, 1H), 12.24 (s, 1H), 12.70 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.7, 109.3, 118.0, 121.3, 125.0, 131.4, 144.1, 144.5, 149.1, 164.2.

Calcd for $C_{10}H_{10}N_4O_5$ (%): C, 45.12; H, 3.79; N, 21.05. Found (%): C, 45.31; H, 3.65; N, 20.82.

Methyl 3-(hydroxyimino)-2-(2-(2-nitrophenyl)hydrazinylidene)propanoate (1p).



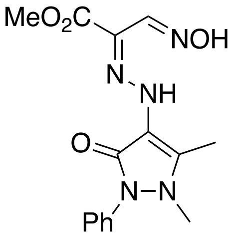
Yield 1.94 g (73%). Orange powder. M.p. 170-171°C (dec.) (EtOH).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.81 (s, 3H), 7.39 (d, *J* = 8.9 Hz, 2H), 8.24-8.27 (m, 3H), 12.37 (s, 1H), 12.68 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 52.8, 115.0, 126.3, 126.7, 142.7, 144.5, 148.3, 164.1.

Calcd for $C_{10}H_{10}N_4O_5$ (%): C, 45.12; H, 3.79; N, 21.05. Found (%): C, 45.25; H, 3.68; N, 20.94.

2-(2-(1,5-Dimethyl-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazol-4-yl)hydrazinylidene)-3-(hydroxyimino)propanoate (1q).



Yield 2.22 g (67%). Orange solid. M.p. 99-101°C.

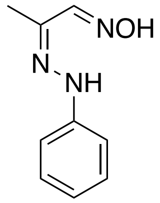
IR (KBr): 3447, 3206, 3146, 3038, 2908, 1710, 1639, 1590, 1545, 1491, 1435, 1334, 1296, 1240, 1190, 1102, 984, 906, 769 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 2.53 (s, 3H), 3.04 (s, 3H), 3.76 (s, 3H), 7.33-7.41 (m, 3H), 7.50-7.55 (m, 2H), 8.28 (s, 1H), 12.05 (s, 1H), 12.07 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 11.4, 36.7, 52.4, 114.6, 122.3, 124.1, 127.1, 129.7, 134.9, 143.4, 144.9, 159.0, 164.6.

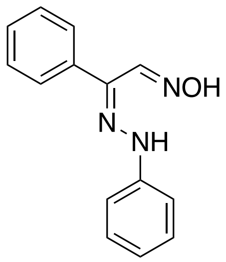
HRMS (ESI): *m*/*z* calcd for $C_{15}H_{18}N_5O_4$: 332.1353; found: 332.1360 $[M+H]^+$.

2-(2-Phenylhydrazinylidene)propanal oxime (1aa).



This compound was synthesized according to a previously reported procedure.[1] All characterization data were consistent with those reported.

2-Phenyl-2-(2-phenylhydrazinylidene)acetaldehyde oxime (1ab).



Phenylhydrazine (3 mL, 30 mmol) was added to a solution of 2-phenyl-2-(2-phenylhydrazinylidene)acetaldehyde (7.17 g, 30 mmol) in EtOH (50 mL). The reaction mixture was refluxed for 48 h, then poured into H_2O (150 mL) and the resulted mixture was left to stand for 2 weeks. The precipitate formed was filtered off, washed with water (2x30 mL) and dried in air.

Yield 1.79 g (25%). Light yellow solid. M.p. 138-140°C.

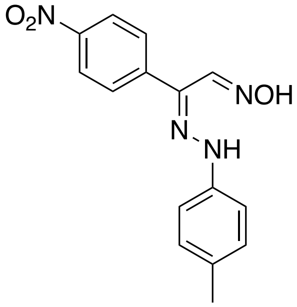
IR (KBr): 3331, 3217, 3051, 1599, 1539, 1477, 1441, 1296, 1260, 1010, 909, 892, 754 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 6.90-6.96 (m, 1H), 7.21-7.25 (m, 2H), 7.30-7.44 (m, 5H), 7.75 (d, *J* = 7.1 Hz, 2H), 8.50 (s, 1H), 11.92 (s, 1H), 12.06 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 113.8, 121.6, 126.3, 128.0, 128.9, 129.9, 131.7, 137.9, 144.6, 145.2.

HRMS (ESI): m/z calcd for $C_{14}H_{14}N_3O$: 240.1131; found: 240.1136 $[M+H]^+$.

2-(4-Nitrophenyl)-2-(2-(*p*-tolyl)hydrazinylidene)acetaldehyde oxime (1ac).



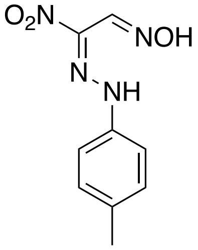
Yield 0.72 g (48%). Dark red solid. M.p. 152-154°C (dec.).

^{1}H NMR (300 MHz, DMSO-d_6): δ = 2.27 (s, 3H), 7.15-7.21 (m, 4H), 7.99-8.02 (m, 2H), 8.19-8.22 (m, 2H), 8.58 (s, 1H), 12.15 (s, 1H), 12.26 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 20.8, 114.4, 124.1, 126.5, 128.8, 130.4, 131.7, 141.6, 144.2, 144.7, 146.4.

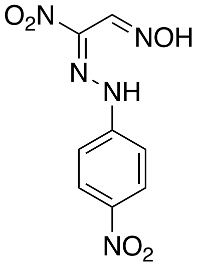
HRMS (ESI): m/z calcd for $C_{15}H_{15}N_4O_3$: 299.1139; found: 299.1145 $[M+H]^+$.

2-Nitro-2-(2-(*p*-tolyl)hydrazinylidene)acetaldehyde oxime (1ad).



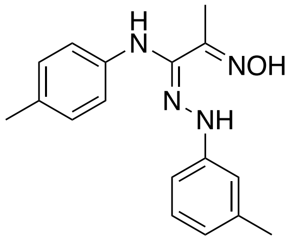
This compound was synthesized according to a previously reported procedure.[2] All characterization data were consistent with those reported.

2-Nitro-2-(2-(4-nitrophenyl)hydrazinylidene)acetaldehyde oxime (1ae).



This compound was synthesized according to a previously reported procedure.[2] All characterization data were consistent with those reported.

2-Phenyl-2-(2-phenylhydrazinylidene)acetaldehyde oxime (1af).



Et_3N (2.1 mL, 15 mmol) was added to a magnetically stirred solution of 2-oxo-*N*-(*m*-tolyl)propanehydrazonoyl chloride (1.13 g, 5 mmol) in MeOH (35 mL) at 0°C. Then *p*-toluidine (0.54 g, 5 mmol) was added in one portion. The reaction was left stirring for 1 h at 0-5°C and for 24 h at 20°C. Hydroxylamine hydrochloride (0.35 g, 5 mmol) was added to the resulted mixture and then stirred for 24 h. Then the reaction mixture was poured into H_2O (100 mL), the precipitate formed was filtered off and recrystallized from EtOH to afford pure oxime **1af**.

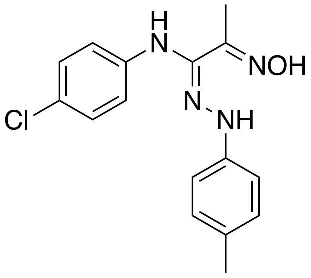
Yield 0.87 g (25%). Beige solid. M.p. 136-138°C (EtOH).

^1H NMR (300 MHz, DMSO-d_6): δ = 2.11 (s, 3H), 2.19 (s, 3H), 2.24 (s, 3H), 6.51 (d, J = 8.3 Hz, 2H), 6.58 (d, J = 7.4 Hz, 2H), 6.91-6.97 (m, 4H), 7.07 (t, J = 7.8 Hz, 1H), 7.32 (s, 1H), 8.79 (s, 1H), 11.19 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 10.9, 20.6, 21.8, 110.6, 113.7, 116.4, 120.4, 128.1, 129.2, 129.5, 136.3, 138.5, 140.8, 145.5, 151.5.

HRMS (ESI): *m*/*z* calcd for $C_{17}H_{21}N_4O$: 297.1710; found: 297.1706 $[M+H]^+$.

***N*-(4-Chlorophenyl)-2-(hydroxyimino)-*N*'-(*p*-tolyl)propanehydrazonamide (1ag).**



Et_3N (2.1 mL, 15 mmol) was added to a magnetically stirred solution of 2-oxo-*N*-(*p*-tolyl)propanehydrazonoyl chloride (1.13 g, 5 mmol) in MeOH (35 mL) at 0°C. Then *p*-chloroaniline (0.64 g, 5 mmol) was added in one portion. The reaction was left stirring for 1 h at 0-5°C and for 24 h at 20°C. Hydroxylamine hydrochloride (0.35 g, 5 mmol) was added to the resulted mixture and then stirred for 24 h. Then the reaction mixture was poured into H_2O (100 mL), the precipitate formed was filtered off and recrystallized from EtOH to afford pure oxime **1ag**.

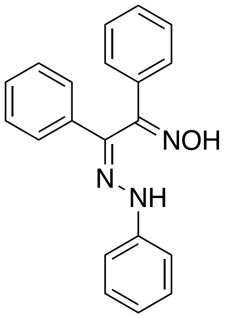
Yield 1.30 g (59%). White solid. M.p. 172-174°C (EtOH).

^1H NMR (300 MHz, DMSO-d_6): δ = 2.14 (s, 3H), 2.23 (s, 3H), 6.58 (d, J = 8.4 Hz, 2H), 7.01-7.11 (m, 4H), 7.18 (d, J = 8.4 Hz, 2H), 7.61 (s, 1H), 9.02 (s, 1H), 11.19 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 10.2, 20.2, 112.9, 116.9, 122.1, 127.8, 128.2, 129.2, 134.5, 142.3, 142.7, 151.0.

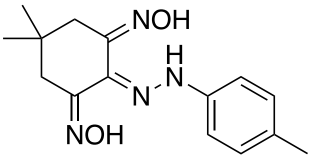
HRMS (ESI): *m*/*z* calcd for $C_{16}H_{18}{}^{35}ClN_4O$: 317.1164; found: 317.1157 $[M+H]^+$.

1,2-Diphenyl-2-(2-phenylhydrazinylidene)ethan-1-one oxime (1ah).



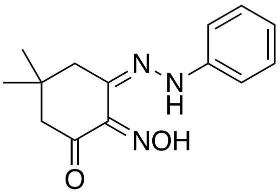
This compound was synthesized according to a previously reported procedure.[3] All characterization data were consistent with those reported.

5,5-Dimethyl-2-(2-(*p*-tolyl)hydrazinylidene)cyclohexane-1,3-dione dioxime (1ai).



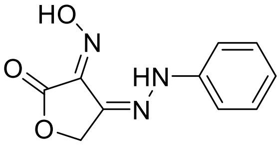
This compound was synthesized according to a previously reported procedure.[4] All characterization data were consistent with those reported.

2-(Hydroxyimino)-5,5-dimethyl-3-(2-phenylhydrazinylidene)cyclohexan-1-one (1aj).



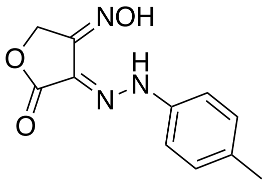
This compound was synthesized according to a previously reported procedure.[5] All characterization data were consistent with those reported.

3-(Hydroxyimino)-4-(2-phenylhydrazinylidene)dihydrofuran-2(3*H*)-one (1ak).



This compound was synthesized according to a previously reported procedure.[6] All characterization data were consistent with those reported.

4-(Hydroxyimino)-3-(2-(*p*-tolyl)hydrazinylidene)dihydrofuran-2(3*H*)-one (1al).



This compound was synthesized according to a previously reported procedure.[7]

Yield 0.91 g (78%). Light yellow solid. M.p. 183-185°C.

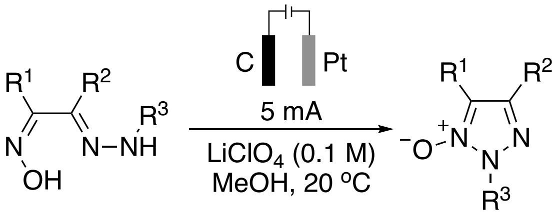
^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 2.30$ (s, 3H), 5.05 (s, 2H), 7.23 (s, 4H), 11.64 (s, 1H), 12.27 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 20.9$, 64.8, 115.4, 120.7, 130.6, 134.2, 139.9, 152.4, 166.2.

HRMS (ESI): m/z calcd for $C_{15}H_{12}N_3O_3$: 234.0873; found: 234.0871 $[M+H]^+$.

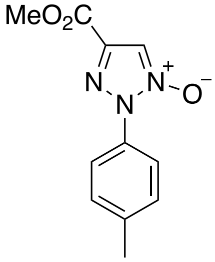
S2.3. Electrochemical Synthesis of 1,2,3-Triazole 1-Oxides

General procedure for the synthesis of 1,2,3-triazole 1-oxides **2**.



The electrolysis was performed in an undivided cell (50 mL vial) with graphite employed as anode and platinum as a cathode. Corresponding oxime (1 mmol) was dissolved in a solution of $LiClO_4$ in MeOH (0.1 M, 40 mL) and oxidized under constant current conditions (j = 2.08 mA cm^{-1}) for 2.2-3.7 F/mol (TLC monitoring). After that, the reaction mixture was concentrated under reduced pressure, the crude residue was extracted with DCM (3x30 mL) and combined organic extracts were evaporated to afford target 1,2,3-triazole 1-oxide. To regenerate $LiClO_4$, the residue obtained after extraction with DCM was stirred in an additional amount of DCM (10 mL) at 30 °C for 1 h and filtered. The precipitate was recrystallized from 95% EtOH to furnish the regeneration of $LiClO_4$, which was used in further experiments.

4-(Methoxycarbonyl)-2-(*p*-tolyl)-2*H*-1,2,3-triazole 1-oxide (2a) was synthesized according to the general procedure in 2.2 F/mol.



Yield 221 mg (95%). Pale yellow powder. M.p. 104-105°C (MeOH).

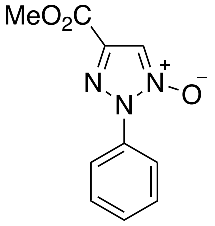
IR (KBr): 3116, 1745, 1524, 1476, 1416, 1356, 1270, 1188, 1133, 996, 929, 825, 751 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): $\delta = 2.41$ (s, 3H), 3.90 (s, 3H), 7.43 (d, $J = 8.2$ Hz, 2H), 7.71 (d, $J = 8.2$ Hz, 2H), 8.56 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 21.3$, 53.0, 118.2, 124.7, 130.2, 131.9, 136.8, 140.8, 159.8.

HRMS (ESI): m/z calcd for $C_{11}H_{12}N_3O_3$: 234.0873; found: 234.0867 $[M+H]^+$.

4-(Methoxycarbonyl)-2-phenyl-2*H*-1,2,3-triazole 1-oxide (2b) was synthesized according to the general procedure in 2.2 F/mol.



Yield 218 mg (96%). Pale yellow powder. M.p. 93-94°C (MeOH).

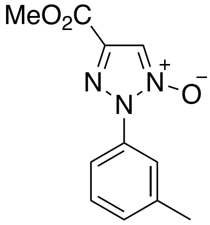
IR (KBr): 3112, 1747, 1482, 1417, 1347, 1251, 1192, 1128, 992, 815, 746 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.91 (s, 3H), 7.60-7.68 (m, 3H), 7.84-7.88 (m, 2H), 8.58 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.1, 118.3, 124.8, 129.8, 130.8, 134.3, 137.0, 159.8.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_{10}N_3O_3$: 220.0716; found: 220.0711 [M+H]$^+$.

4-(Methoxycarbonyl)-2-(3-methylphenyl)-2*H*-1,2,3-triazole 1-oxide (2c) was synthesized according to the general procedure in 2.3 F/mol.



Yield 219 mg (94%). Light brown powder. M.p. 89-90°C (MeOH).

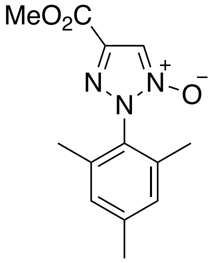
IR (KBr): 3085, 1753, 1612, 1435, 1353, 1265, 1213, 1135, 1012, 753 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 2.42 (s, 3H), 3.91 (s, 3H), 7.43 (d, *J* = 7.4 Hz, 1H), 7.52 (t, *J* = 8.1 Hz, 1H), 7.65 (s, 2H), 8.58 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 21.2, 53.1, 118.3, 121.8, 125.0, 129.5, 131.4, 134.2, 136.9, 139.6, 159.8.

HRMS (ESI): *m*/*z* [M+H]$^+$ calcd for $C_{11}H_{12}N_3O_3$: 234.0873; found: 234.0869.

2-Mesityl-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2d) was synthesized according to the general procedure in 2.3 F/mol.



Yield 240 mg (92%). Light yellow powder. M.p. 126-127°C (MeOH).

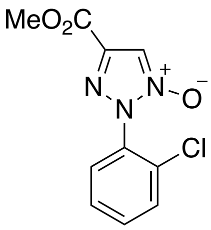
IR (KBr): 3107, 1748, 1475, 1425, 1348, 1241, 1118, 991, 814, 764 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 1.95 (s, 6H), 2.35 (s, 3H), 3.90 (s, 3H), 7.13 (s, 2H), 8.60 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 17.2, 21.2, 53.0, 117.3, 129.5, 129.8, 137.4, 137.6, 142.2, 159.8.

HRMS (ESI): *m*/*z* calcd for $C_{13}H_{16}N_3O_3$: 262.1186; found: 262.1185 [M+H]$^+$.

2-(2-Chlorophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2e) was synthesized according to the general procedure in 2.3 F/mol.



Yield 225 mg (89%). Light yellow solid. M.p. 120-121°C (MeOH).

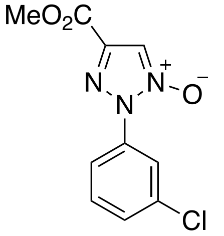
IR (KBr): 3100, 2954, 1741, 1522, 1482, 1422, 1191, 1144, 1088, 998, 931, 817, 756 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.92 (s, 3H), 7.62-7.67 (m, 1H), 7.73-7.79 (m, 1H), 7.81-7.86 (m, 1H), 8.64 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.2, 117.1, 129.1, 130.9, 131.4, 131.5, 132.6, 134.4, 138.1, 159.7.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_8{}^{35}ClN_3O_3$: 254.0327; found: 254.0325 $[M+H]^+$.

2-(3-Chlorophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2f) was synthesized according to the general procedure in 2.2 F/mol.



Yield 230 mg (91%). Light yellow solid. M.p. 112-113°C (MeOH).

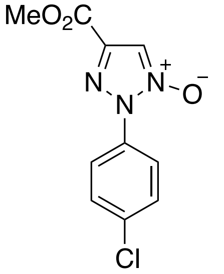
IR (KBr): 3113, 1743, 1591, 1499, 1431, 1351, 1263, 1189, 1131, 991, 928, 868, 821, 777 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.92 (s, 3H), 7.65-7.72 (m, 2H), 7.83-7.87 (m, 1H), 8.02 (br s, 1H), 8.63 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.2, 118.5, 123.2, 124.3, 130.6, 131.5, 133.8, 135.2, 137.4, 159.7.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_8{}^{35}ClN_3O_3$: 254.0327; found: 254.0328 $[M+H]^+$.

2-(4-Chlorophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2g) was synthesized according to the general procedure in 2.3 F/mol.



Yield 235 mg (93%). Pale yellow solid. M.p. 114-115°C (MeOH).

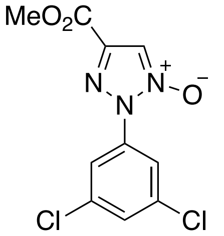
IR (KBr): 3122, 1751, 1488, 1243, 1191, 1092, 998, 830, 765 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.91 (s, 3H), 7.71 (d, *J* = 9.1 Hz, 2H), 7.91 (d, *J* = 9.1 Hz, 2H), 8.61 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.1, 118.4, 126.3, 129.8, 133.0, 135.2, 137.2, 159.7.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_8{}^{35}ClN_3O_3$: 254.0327; found: 254.0326 $[M+H]^+$.

2-(3,5-Dichlorophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2h) was synthesized according to the general procedure in 2.3 F/mol.



Yield 270 mg (94%). Pale yellow solid. M.p. 171-172°C (CH_2Cl_2).

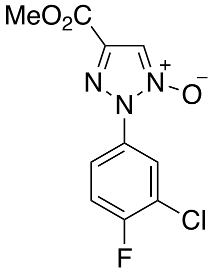
IR (KBr): 3106, 1753, 1585, 1528, 1487, 1436, 1346, 1271, 1187, 1144, 1004, 934, 858, 792, 740 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 3.92 (s, 3H), 7.93 (t, J = 1.9 Hz, 1H), 8.01 (d, J = 1.9 Hz, 2H), 8.68 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 53.2, 118.5, 123.0, 130.2, 135.0, 135.7, 137.8, 159.5.

Calcd for $C_{10}H_7Cl_2N_3O_3$ (%): C, 41.69; H, 2.45; N, 14.59. Found (%): C, 41.82; H, 2.33; N, 14.37.

2-(3-Chloro-4-fluorophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2i) was synthesized according to the general procedure in 2.3 F/mol.



Yield 253 mg (93%). Pale yellow solid. M.p. 174-175°C (MeOH).

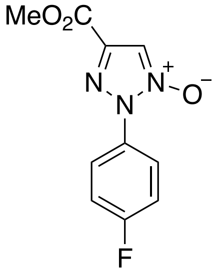
IR (KBr): 3140, 3104, 1734, 1500, 1411, 1364, 1273, 1191, 1122, 1068, 992, 928, 822, 760 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 3.92 (s, 3H), 7.72 (t, J = 9.0 Hz, 1H), 7.87-7.93 (m, 1H), 8.19 (dd, J = 4.1, 2.5 Hz, 1H), 8.64 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 52.5, 115.4 (d, J = 7.2 Hz), 116.6, 118.4 (d, J = 22.5 Hz), 120.7 (d, J = 19.0 Hz), 124.2 (d, J = 1.7 Hz), 140.3 (d, J = 2.7 Hz), 144.5, 154.3 (d, J = 242.6 Hz), 159.6.

Calcd for $C_{10}H_7ClFN_3O_3$ (%): C, 44.22; H, 2.60; N, 15.47. Found (%): C, 44.38; H, 2.47; N, 15.29.

2-(4-Fluorophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2j) was synthesized according to the general procedure in 2.3 F/mol.



Yield 218 mg (92%). Pale grey solid. M.p. 86-87°C (MeOH).

IR (KBr): 3155, 1735, 1495, 1417, 1362, 1266, 1127, 996, 935, 839, 755 cm^{-1}.

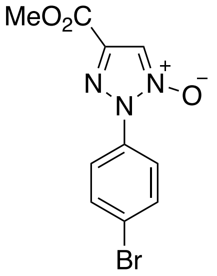
^1H NMR (300 MHz, DMSO-d_6): δ = 3.91 (s, 3H), 7.49 (t, J = 8.9 Hz, 2H), 7.88-7.93 (m, 2H), 8.59 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 53.1, 116.9 (d, J = 23.7 Hz), 118.2, 127.7 (d, J = 9.3 Hz), 130.5 (d, J = 3.3 Hz), 137.0, 159.8, 163.0 (d, J = 248.8 Hz).

^{19}F NMR (282.4 MHz, DMSO-d_6): δ = -109.7.

HRMS (ESI): m/z calcd for $C_{10}H_9FN_3O_3$: 238.0619; found: 238.0622 $[M+H]^+$.

2-(4-Bromophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2k) was synthesized according to the general procedure in 2.3 F/mol.



Yield 259 mg (87%). Pale grey solid. M.p. 114-115°C (CH_2Cl_2).

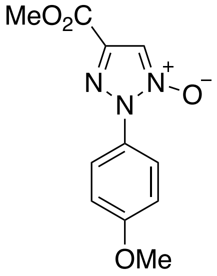
IR (KBr): 3131, 3097, 1742, 1484, 1414, 1354, 1248, 1186, 1125, 1067, 996, 931, 825, 766 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): $\delta = 3.91$ (s, 3H), 7.85 (br s, 4H), 8.60 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 53.1, 118.4, 123.8, 126.5, 132.8, 133.5, 137.2, 159.7$.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_9{}^{79}BrN_3O_3$: 297.9822; found: 297.9821; calcd for $C_{10}H_9{}^{81}BrN_3O_3$: 299.9802; found: 299.9805 $[M+H]^+$.

4-(Methoxycarbonyl)-2-(4-methoxyphenyl)-2*H*-1,2,3-triazole 1-oxide (2l) was synthesized according to the general procedure in 2.3 F/mol.



Yield 229 mg (92%). Pale grey solid. M.p. 124-125°C (CH_2Cl_2).

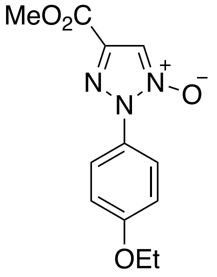
IR (KBr): 3140, 1758, 1590, 1506, 1438, 1358, 1258, 1188, 1134, 996, 935, 830, 769 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): $\delta = 3.85$ (s, 3H), 3.90 (s, 3H), 7.15 (d, $J = 9.1$ Hz, 2H), 7.73 (d, $J = 9.1$ Hz, 2H), 8.53 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 53.0, 56.2, 114.9, 117.9, 126.9, 127.1, 153.8, 159.9, 161.0$.

HRMS (ESI): *m*/*z* calcd for $C_{11}H_{12}N_3O_4$: 250.0822; found: 250.0828 $[M+H]^+$.

2-(4-Ethoxyphenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2m) was synthesized according to the general procedure in 2.3 F/mol.



Yield 246 mg (92%). Pale grey solid. M.p. 115-116°C (dec.)

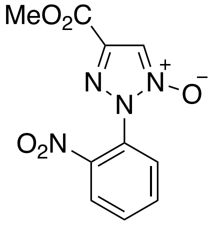
IR (KBr): 3141, 3093, 1753, 1505, 1438, 1363, 1257, 1169, 1120, 1045, 1000, 930, 840, 757 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 1.37 (t, *J* = 7.0 Hz, 3H), 3.90 (s, 3H), 4.13 (q, *J* = 7.0 Hz, 2H), 7.13 (d, *J* = 9.2 Hz, 2H), 7.71 (d, *J* = 9.2 Hz, 2H), 8.53 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 14.9, 53.0, 64.2, 115.2, 117.9, 126.8, 126.9, 136.5, 159.9, 160.2.

HRMS (ESI): *m*/*z* calcd for $C_{12}H_{14}N_3O_4$: 264.0979; found: 264.0978 $[M+H]^+$.

2-(2-Nitrophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2n) was synthesized according to the general procedure in 3.7 F/mol.



Yield 232 mg (88%). Orange powder. M.p. 145°C; T$_d$ 212°C.

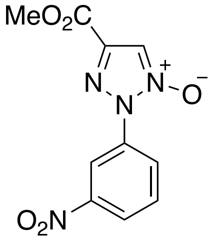
IR (KBr): 3144, 3096, 1745, 1609, 1537, 1473, 1428, 1353, 1260, 1191, 1133, 995, 930, 856, 817, 788, 740 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.93 (s, 3H), 7.95-8.01 (m, 2H), 8.06-8.11 (m, 1H), 8.34-8.37 (m, 1H), 8.70 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.3, 117.8, 125.6, 126.1, 130.3, 133.7, 135.9, 138.6, 144.4, 159.5.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_9N_4O_5$: 265.0567; found: 265.0568 $[M+H]^+$.

2-(3-Nitrophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2o) was synthesized according to the general procedure in 2.5 F/mol.



Yield 230 mg (87%). Pale grey solid. M.p. 151°C (dec.)

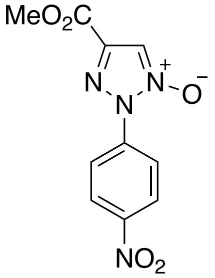
IR (KBr): 3115, 1751, 1537, 1484, 1423, 1350, 1263, 1186, 1141, 1103, 1005, 937, 892, 816, 771, 724 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 3.93 (s, 3H), 7.94 (t, *J* = 8.2 Hz, 1H), 8.35 (d, *J* = 8.2 Hz, 1H), 8.45 (d, *J* = 8.2 Hz, 1H), 8.70 (s, 1H), 8.80 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 53.2, 118.7, 119.1, 125.1, 130.3, 131.5, 134.8, 137.7, 148.2, 159.6.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_9N_4O_5$: 265.0567; found: 265.0570 $[M+H]^+$.

2-(4-Nitrophenyl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2p) was synthesized according to the general procedure in 2.5 F/mol.



Yield 240 mg (91%). Orange powder. M.p. 163°C; T_d 242°C.

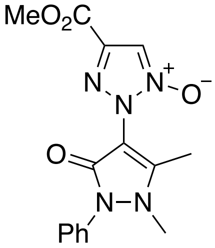
IR (KBr): 3106, 1751, 1596, 1524, 1437, 1347, 1251, 1185, 1111, 997, 853, 772, 747 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 3.93$ (s, 3H), 8.26 (d, $J = 9.2$ Hz, 2H), 8.47 (d, $J = 9.2$ Hz, 2H), 8.70 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 53.2$, 118.9, 124.3, 125.2, 138.0, 138.8, 147.6, 159.6.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_9N_4O_5$: 265.0567; found: 265.0572 $[M+H]^+$.

2-(1,5-Dimethyl-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazol-4-yl)-4-(methoxycarbonyl)-2*H*-1,2,3-triazole 1-oxide (2q) was synthesized according to the general procedure in 2.1 F/mol.



Yield 145 mg (44%). Dark orange solid. M.p. 186°C (dec.)

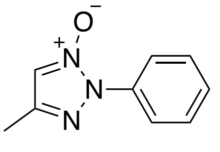
IR (KBr): 3468, 3104, 2926, 2853, 1737, 1670, 1492, 1420, 1276, 1218, 1127, 992, 929, 754 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 2.25$ (s, 3H), 3.35 (s, 3H), 3.90 (s, 3H), 7.39-7.49 (m, 3H), 7.58 (t, $J = 7.6$ Hz, 2H), 8.53 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 10.9$, 34.9, 53.1, 100.3, 116.6, 126.7, 128.7, 129.9, 134.1, 137.8, 152.9, 159.3, 159.8.

HRMS (ESI): *m*/*z* calcd for $C_{15}H_{16}N_5O_4$: 330.1197; found: 330.1196 $[M+H]^+$.

4-Methyl-2-phenyl-2*H*-1,2,3-triazole 1-oxide (2aa) was synthesized according to the general procedure in 2.1 F/mol.



Yield 140 mg (80%). M.p. 62-63°C (CH_2Cl_2).

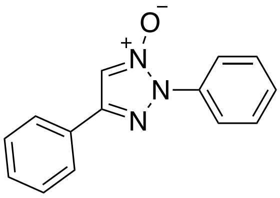
IR (KBr): 3454, 3102, 1738, 1605, 1525, 1494, 1435, 1351, 1178, 1108, 943, 854 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 2.28$ (s, 3H), 7.48-7.60 (m, 3H), 7.81-7.84 (m, 2H), 7.88 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 12.4$, 117.1, 123.6, 129.4, 129.6, 135.1, 143.2.

Calcd for $C_9H_9N_3O$ (%): C, 61.70; H, 5.18; N, 23.99. Found (%): C, 61.52; H, 5.31; N, 23.73.

2,4-Diphenyl-2*H*-1,2,3-triazole 1-oxide (2ab) was synthesized according to the general procedure in 2.1 F/mol.



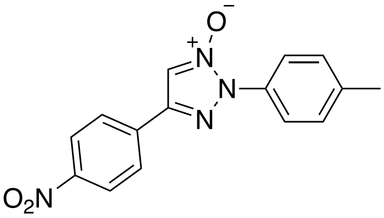
Yield 218 mg (92%). White solid. M.p. 88-89°C (CH_2Cl_2).

^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 7.48\text{-}7.65$ (m, 6H), 7.90-7.94 (m, 4H), 8.67 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 115.1$, 124.1, 125.9, 129.2, 129.5, 129.7, 129.8, 130.2, 135.0, 145.1.

HRMS (ESI): *m*/*z* calcd for $C_{14}H_{12}N_3O$: 238.0975; found: 238.0970 $[M+H]^+$.

4-(4-Nitrophenyl)-2-(p-tolyl)-2H-1,2,3-triazole 1-oxide (2ac) was synthesized according to the general procedure in 2.6 F/mol.



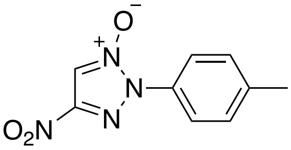
Yield 104 mg (35%). Beige solid. M.p. 142-143°C (CH_2Cl_2).

^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 2.41$ (s, 3H), 7.43 (d, $J = 8.0$ Hz, 2H), 7.79 (d, $J = 8.0$ Hz, 2H), 8.14 (d, $J = 9.1$ Hz, 2H), 8.36 (d, $J = 7.9$ Hz, 2H), 8.79 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 21.3$, 115.7, 124.2, 124.8, 126.9, 130.1, 132.3, 135.4, 140.1, 142.7, 148.2.

HRMS (ESI): *m*/*z* calcd for $C_{15}H_{13}N_4O_3$: 297.0982; found: 297.0980 $[M+H]^+$.

4-Nitro-2-(*p*-tolyl)-2*H*-1,2,3-triazole 1-oxide (2ad) was synthesized according to the general procedure in 2.6 F/mol.



Yield 158 mg (72%). Beige solid. M.p. 126-127°C (CH_2Cl_2).

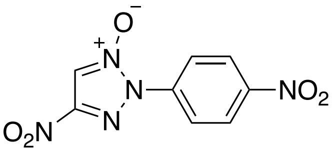
IR (KBr): 3152, 1566, 1519, 1489, 1436, 1365, 1329, 1194, 1136, 982, 831, 755 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d_6): $\delta = 7.47$ (d, $J = 8.0$ Hz, 2H), 7.73 (d, J = 8.0 Hz, 2H), 9.12 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): $\delta = 21.4$, 113.6, 125.2, 130.3, 131.1, 141.7, 149.1.

HRMS (ESI): *m*/*z* calcd for $C_9H_9N_4O_3$: 221.0669; found: 221.0677 $[M+H]^+$.

4-Nitro-2-(4-nitrophenyl)-2*H*-1,2,3-triazole 1-oxide (2ae) was synthesized according to the general procedure in 2.6 F/mol.



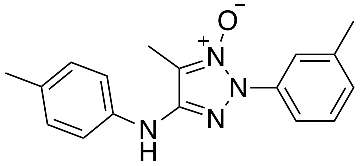
Yield 171 mg (68%). M.p. 132°C; T_d 237 °C.

^{1}H NMR (300 MHz, DMSO-d_6): δ = 8.23 (d, *J* = 9.2 Hz, 2H), 8.49 (d, *J* = 9.2 Hz, 2H), 9.26 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 114.3, 125.3, 125.4, 138.1, 148.2, 150.0.

All characterization data were consistent with those previously reported.[2]

5-Methyl-2-(*m*-tolyl)-4-(*p*-tolylamino)-2*H*-1,2,3-triazole 1-oxide (2af) was synthesized according to the general procedure in 2.1 F/mol.



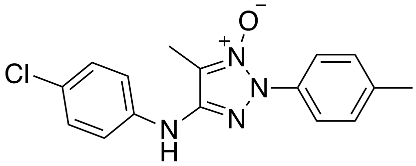
Yield 226 mg (77%). Light grey solid. M.p. 130-132°C (CH_2Cl_2).

^{1}H NMR (300 MHz, DMSO-d_6): δ = 2.24 (s, 6H), 2.39 (s, 3H), 7.10 (d, *J* = 8.2 Hz, 2H), 7.24 (d, *J* = 8.2 Hz, 1H), 7.40-7.46 (m, 3H), 7.62-7.65 (m, 2H), 8.60 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 7.8, 20.8, 21.5, 115.0, 117.2, 120.0, 123.1, 128.8, 129.3, 129.5, 129.8, 136.3, 139.0, 139.4, 145.6.

HRMS (ESI): *m/z* calcd for $C_{17}H_{19}N_4O$: 295.1553; found: 295.1554 $[M+H]^+$.

5-Methyl-2-(*m*-tolyl)-4-(*p*-tolylamino)-2*H*-1,2,3-triazole 1-oxide (2ag) was synthesized according to the general procedure in 2.1 F/mol.



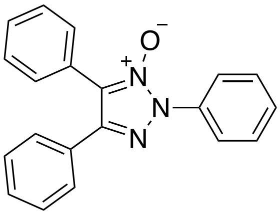
Yield 286 mg (91%). Grey solid. M.p. 100-101°C (CH_2Cl_2).

^{1}H NMR (300 MHz, DMSO-d_6): δ = 2.23 (s, 3H), 2.37 (s, 3H), 7.31-7.36 (m, 4H), 7.56 (d, *J* = 8.6 Hz, 2H), 7.70 (d, *J* = 8.1 Hz, 2H), 8.87 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d_6): δ = 7.8, 21.2, 115.0, 118.4, 123.0, 124.1, 129.2, 129.9, 133.8, 138.0, 141.0, 144.9.

HRMS (ESI): *m/z* calcd for $C_{16}H_{15}{}^{35}ClN_4O$: 315.1007; found: 315.0998 $[M+H]^+$.

2,4,5-Triphenyl-2*H*-1,2,3-triazole 1-oxide (2ah) was synthesized according to the general procedure in 2.1 F/mol.

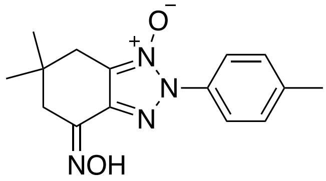


Yield 282 mg (90%). M.p. 143°C (dec.)

^{1}H NMR (300 MHz, $CDCl_3$): δ = 7.41-7.52 (m, 7H), 7.56-7.64 (m, 4H), 7.69-7.73 (m, 2H), 8.09-8.12 (m, 2H).

All characterization data were consistent with those previously reported.[3]

4-(Hydroxyimino)-6,6-dimethyl-2-(*p*-tolyl)-4,5,6,7-tetrahydro-2*H*-benzo[*d*][1,2,3]triazole 1-oxide (2ai) was synthesized according to the general procedure in 2.1 F/mol.



Yield 266 mg (93%). Beige solid. M.p. 190°C (dec.)

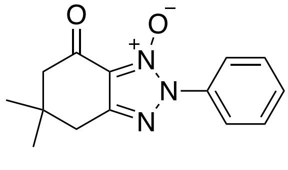
IR (KBr): 3395, 3225, 2956, 2871, 1709, 1632, 1508, 1445, 1368, 1292, 1209, 1110, 986, 945, 815 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 1.05 (s, 6H), 2.39 (s, 3H), 2.57 (s, 2H), 2.60 (s, 2H), 7.39 (d, J = 8.2 Hz, 2H), 7.74 (d, J = 8.2 Hz, 2H), 11.63 (s, 1H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 21.2, 28.3, 32.0, 36.5, 123.8, 126.1, 130.0, 132.9, 138.3, 139.6, 145.6.

HRMS (ESI): m/z calcd for $C_{15}H_{19}N_4O_2$: 287.1503; found: 287.1496 $[M+H]^+$.

5,5-Dimethyl-7-oxo-2-phenyl-4,5,6,7-tetrahydro-2H-benzo[d][1,2,3]triazole 1-oxide (2aj) was synthesized according to the general procedure in 2.1 F/mol.



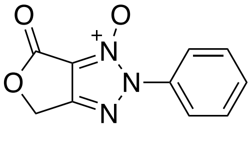
Yield 244 mg (95%). Beige solid. M.p. 130-131°C (CH_2Cl_2).

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 1.11 (s, 6H), 2.52 (s, 2H), 2.84 (s, 2H), 7.55-7.66 (m, 3H), 7.83-7.86 (m, 2H).

HRMS (ESI): m/z calcd for $C_{14}H_{16}N_3O_2$: 258.1237; found: 258.1232 $[M+H]^+$.

All characterization data were consistent with those previously reported.[8]

6-Oxo-2-phenyl-2,6-dihydro-4*H*-furo[3,4-*d*][1,2,3]triazole 1-oxide (2ak) was synthesized according to the general procedure in 2.1 F/mol.



Yield 111 mg (51%). Beige solid. M.p. 177°C (dec.)

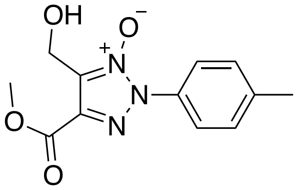
IR (KBr): 2924, 1773, 1546, 1497, 1453, 1284, 1092, 1031, 1012, 970, 764 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 5.52 (s, 2H), 7.60-7.70 (m, 3H), 7.83 (d, *J* = 8.1 Hz, 2H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 66.0, 117.4, 124.7, 130.0, 131.0, 134.6, 157.2, 157.3.

HRMS (ESI): *m*/*z* calcd for $C_{10}H_8N_3O_3$: 218.0560; found: 218.0561 $[M+H]^+$.

5-(Hydroxymethyl)-4-(methoxycarbonyl)-2-(p-tolyl)-2H-1,2,3-triazole 1-oxide (2al) was synthesized according to the general procedure in 2.1 F/mol.



Yield 150 mg (57%). Yellow solid. M.p. 194°C (dec.)

IR (KBr): 3437, 2925, 2854, 1734, 1637, 1517, 1444, 1360, 1296, 1175, 1072, 1023, 821 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d$_6$): δ = 2.41 (s, 3H), 3.93 (s, 3H), 4.67 (d, *J* = 5.7 Hz, 2H), 5.38 (t, *J* = 5.7 Hz, 1H), 7.44 (d, *J* = 8.2 Hz, 2H), 7.72 (d, *J* = 8.2 Hz, 2H).

^{13}C NMR (75.5 MHz, DMSO-d$_6$): δ = 21.3, 51.2, 53.0, 124.6, 129.5, 130.3, 131.9, 135.0, 140.8, 160.3.

Calcd for $C_{12}H_{13}N_3O_4$ (%): C, 54.75; H, 4.98; N, 15.96. Found (%): C, 54.58; H, 5.11; N, 15.68.

S2.4 NO release assay

The test molecule (0.1 mmol) was dissolved in DMSO (50 mL). 20 μL aliquot of the resulted solution was diluted with phosphate buffer solution (180 μL, pH 7.4, containing 2 μmol L-cysteine). The final concentration of the 1,2,3-triazole 1-oxide derivative was $2 \cdot 10^{-4}$ M. The mixture was incubated at 37 °C for 1 h. 50 μL aliquot of the Griess reagent (prepared by mixing sulfanilamide (4 g), *N*-naphthylethylenediamine dihydrochloride (0.2 g) and 85% H_3PO_4 (10 mL) in distilled and deionized water (final volume 100 mL)) was added and incubated for 10 min at 37 °C. UV absorbance at 540 nm was measured using a Multiskan GO Microplate Photometer and calibrated using a standard curve prepared from standard solutions of $NaNO_2$ to give the nitrite concentration. All measurements were made in triplicate. No significant NO release was measured at the absence of L-cysteine.

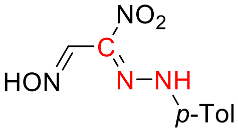
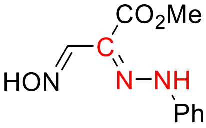
S3. Computational Details

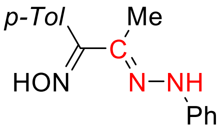
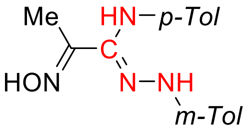
DFT calculations were performed within Gaussian09 package[9] using B3LYP/6-311++(*d*,*p*) basis set. Solvation effects were taken into account using PCM model (solvent – MeOH, ε=32.613). Stationary points were located by the Hessian matrix calculations by the absence of the imaginary frequencies.

Standard oxidation potentials were calculated according to the following equation:

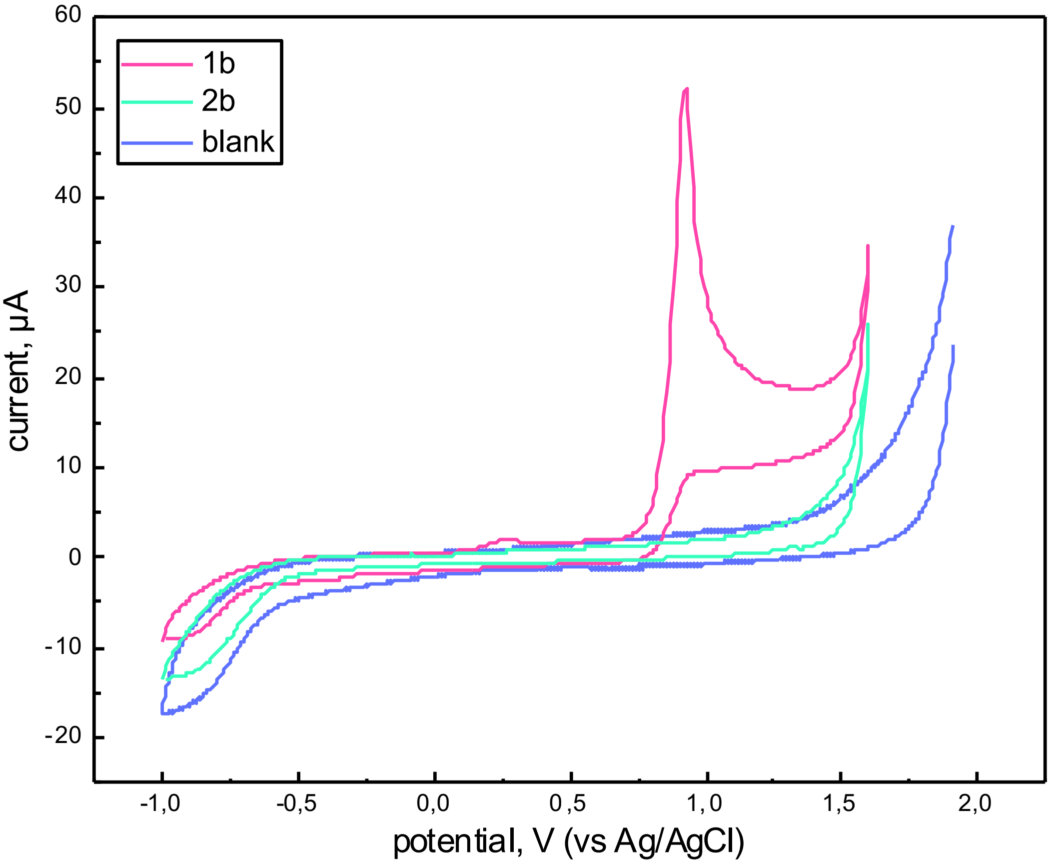
$$E^0 = -\Delta_r G^0/nF - 4.44,$$

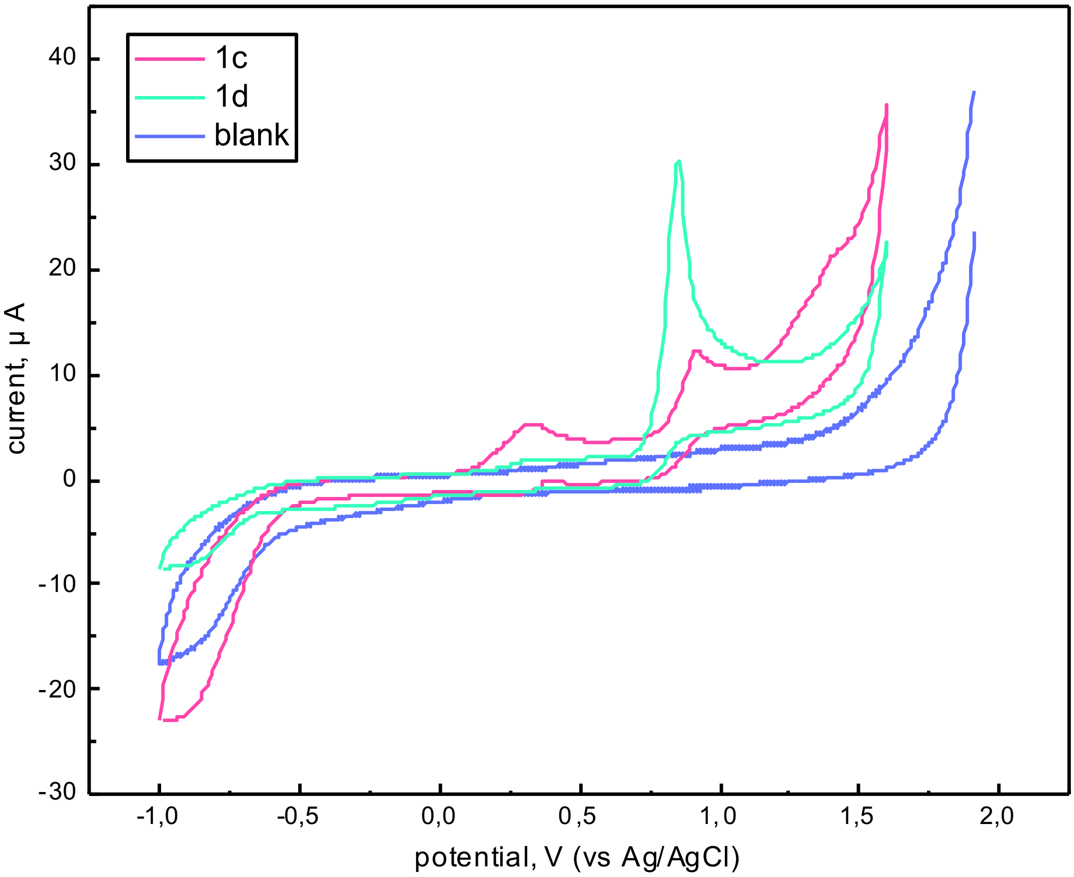
where E^0 – standard potential, $\Delta_r G^0$ – standard Gibbs free energy of reduction, n – number of electrons, F – Faraday constant, 4.44 – absolute oxidation potential of the hydrogen electrode. Hirshfeld atomic charges[10] were calculated using MultiWFN software.[11] Oxidized motif was determined by the difference between atomic charges for neutral molecule and oxidized molecule.

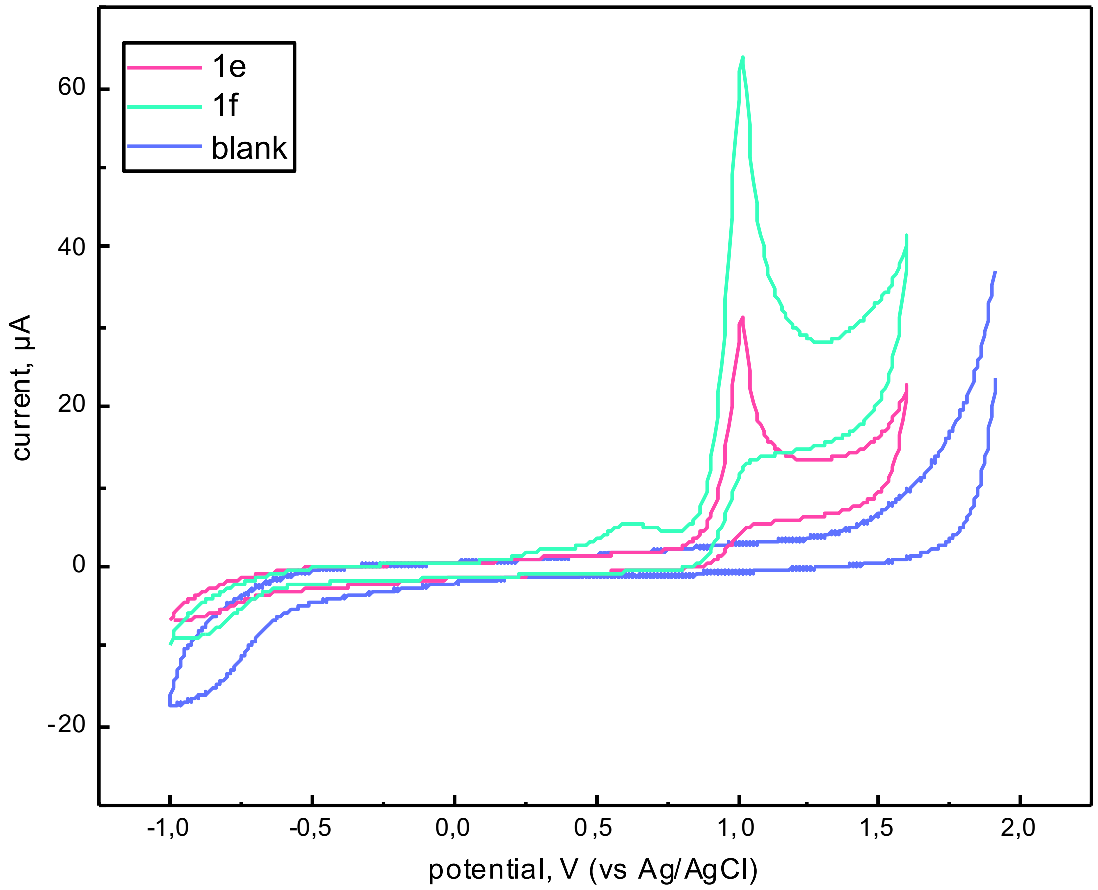
	Hirshfeld charge for neutral molecule	Hirshfeld charge for oxidized molecule	Δq
			
C	0.050217	0.118442	0.068225
N (double bonded)	-0.03963	-0.02324	0.016397
N (NH)	0.03481	0.086551	0.051741
H	0.113815	0.136797	0.022982
			
C	0.00053	0.080933	0.080402
N (double bonded)	-0.05652	-0.02721	0.029313
N (NH)	0.008537	0.093195	0.084658
H	0.106679	0.136539	0.02986

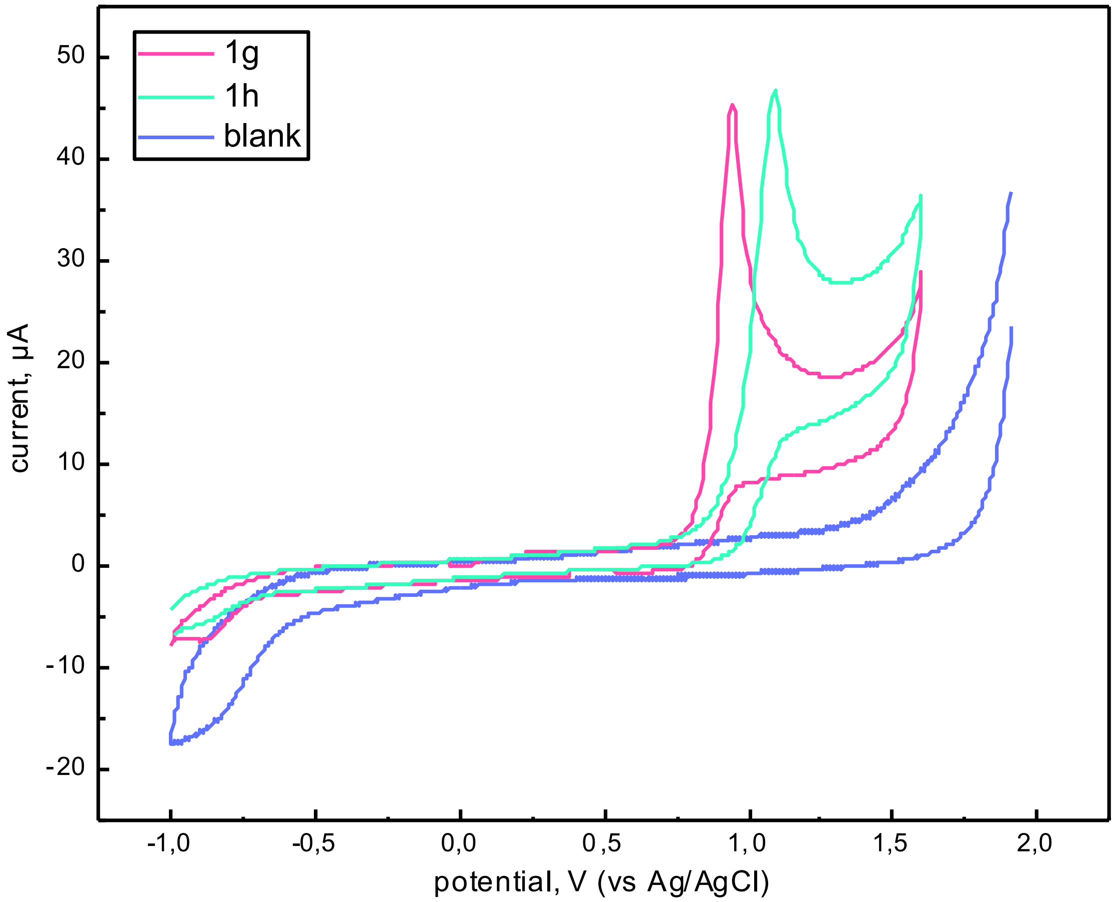
			
C	0.037661	0.109241	0.071579
N (double bonded)	-0.10479	-0.06011	0.044679
N (NH)	-0.02898	0.087697	0.116679
H	0.126254	0.164015	0.037761
			
C	0.070452	0.134284	0.063831
N (double bonded)	-0.09449	-0.07506	0.019433
N (NH, hydrazone)	-0.02648	0.066887	0.093368
H (hydrazone)	0.114228	0.160401	0.046173
N (NH, amide)	-0.10576	-0.02253	0.083229
H (amide)	0.107206	0.155722	0.048516

S4. CV curves


1b
2b
blank
current, µA
60
50
40
30
20
10
0
-10
-20
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)


1c
1d
blank
current, µ A
40
30
20
10
0
-10
-20
-30
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)


1e
1f
blank
60
40
20
0
-20
current, µA
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)


1g
1h
blank
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40
30
20
10
0
-10
-20
current, µA
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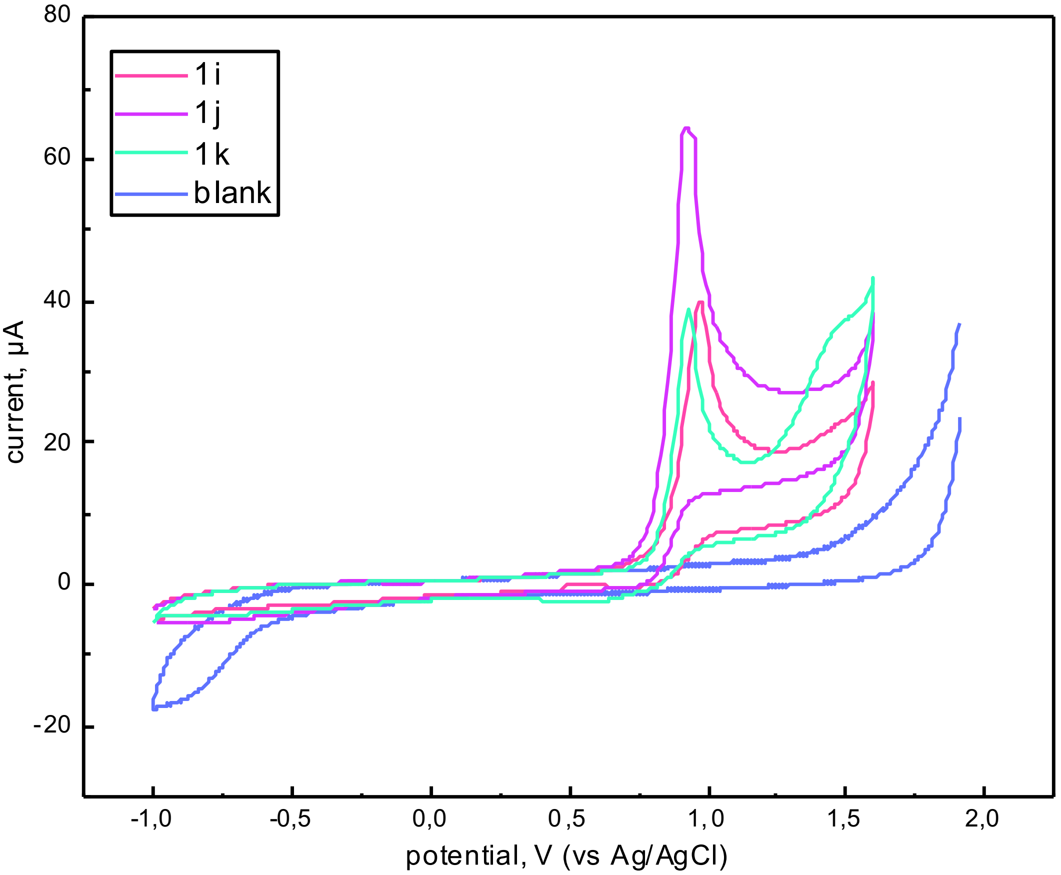
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)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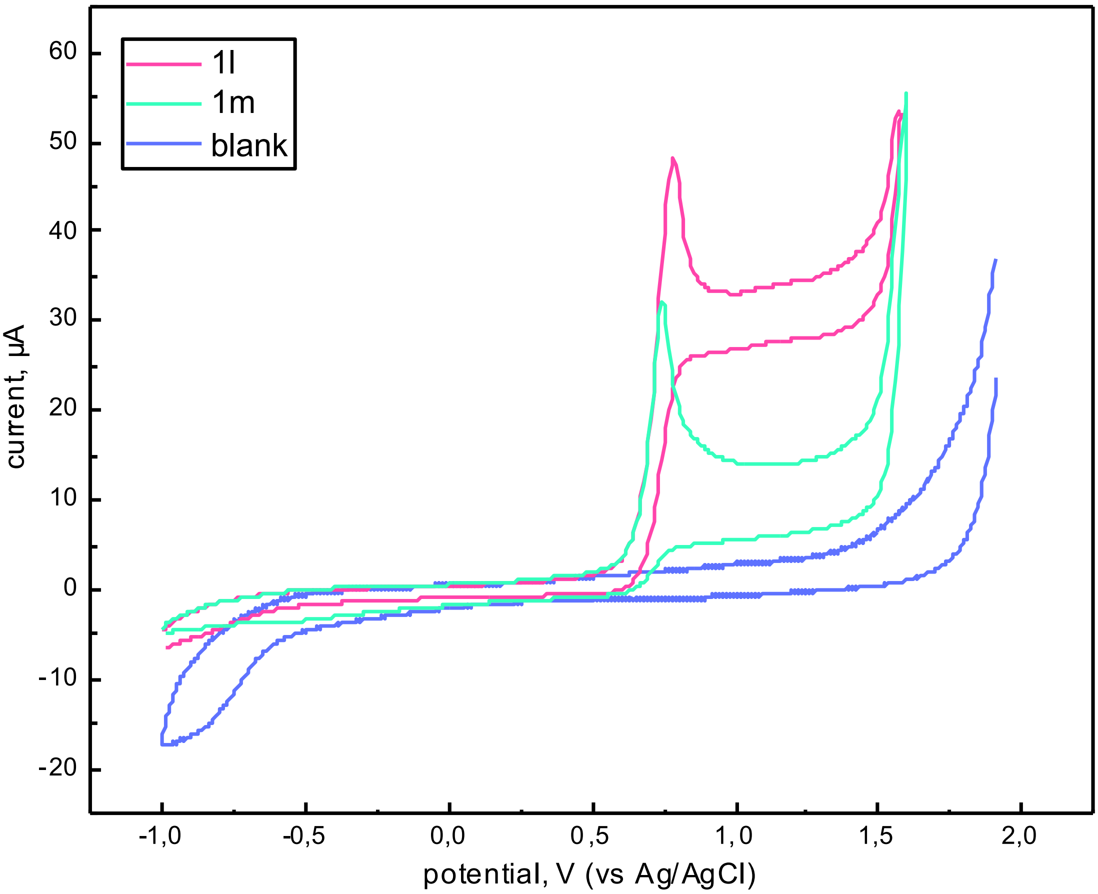
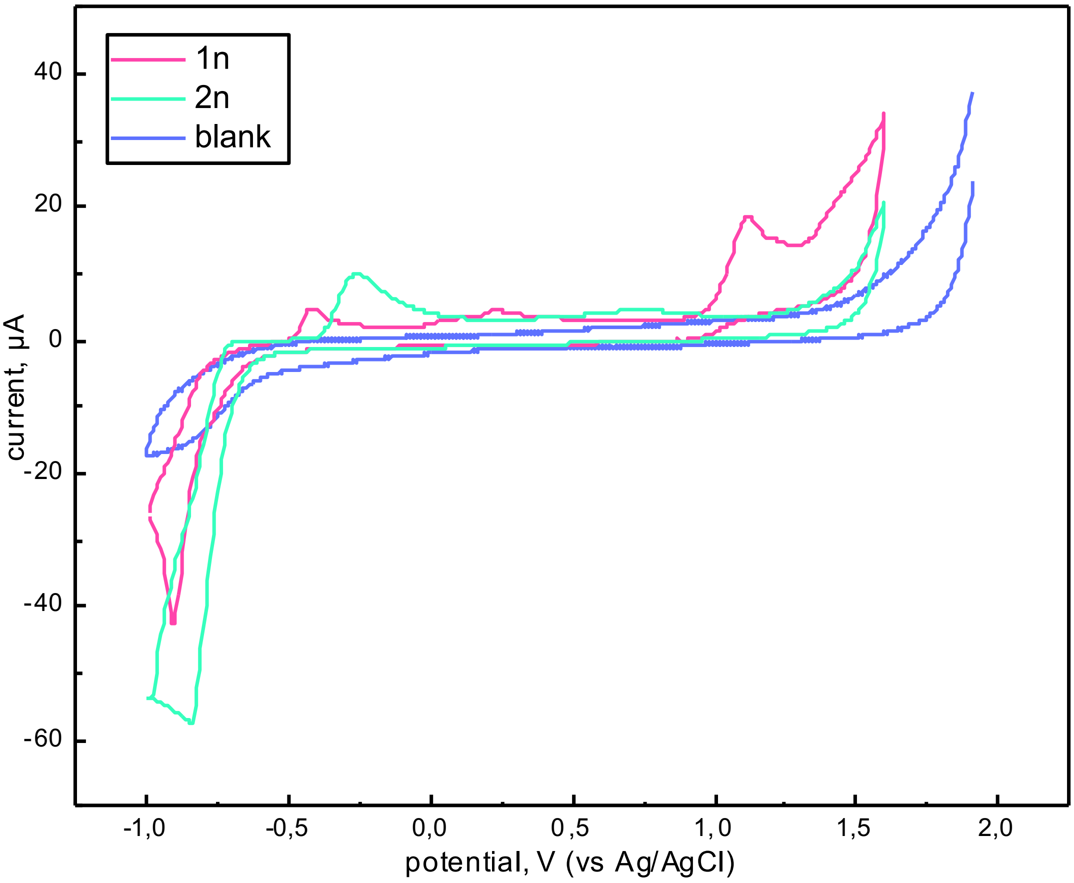
60
40
20
0
-20
current, µA
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)
1i
1j
1k
blank


60
50
40
30
20
10
0
-10
-20
current, µA
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)
1l
1m
blank


1n
2n
blank
current, µA
40
20
0
-20
-40
-60
-1,0
-0,5
0,0
0,5
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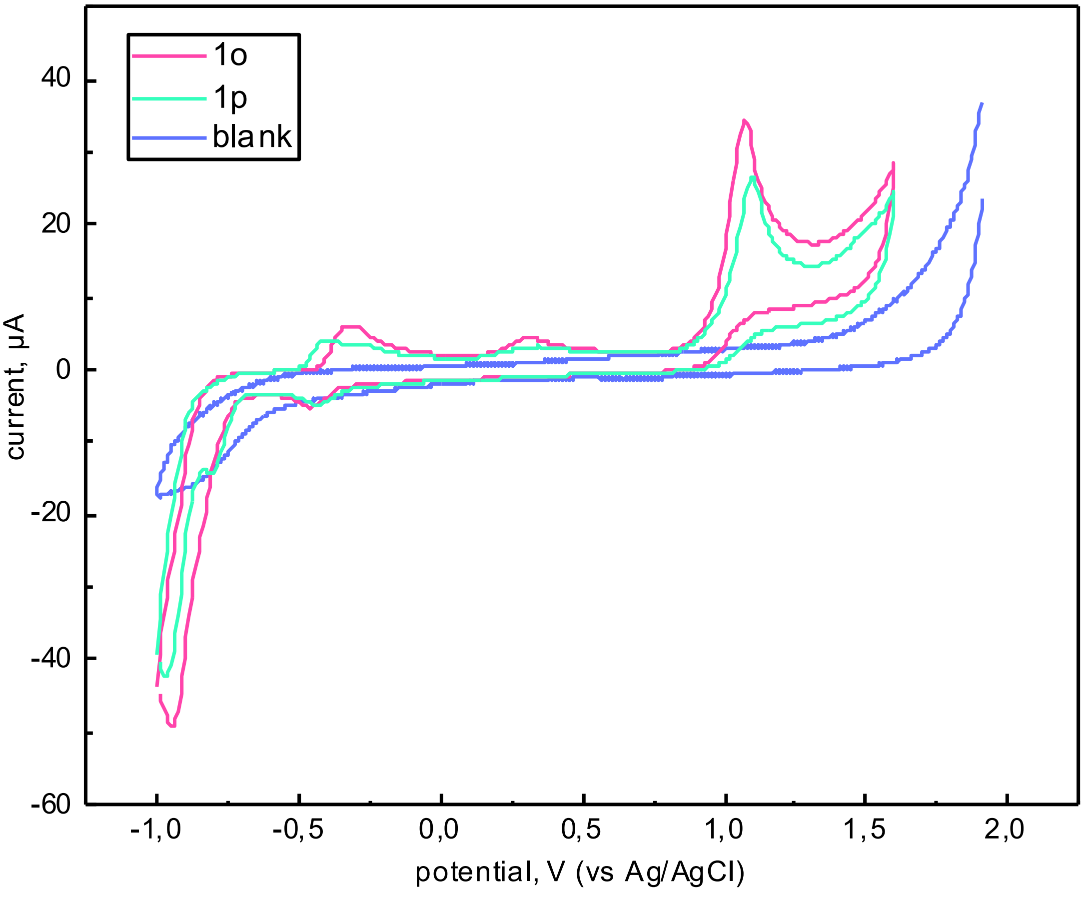
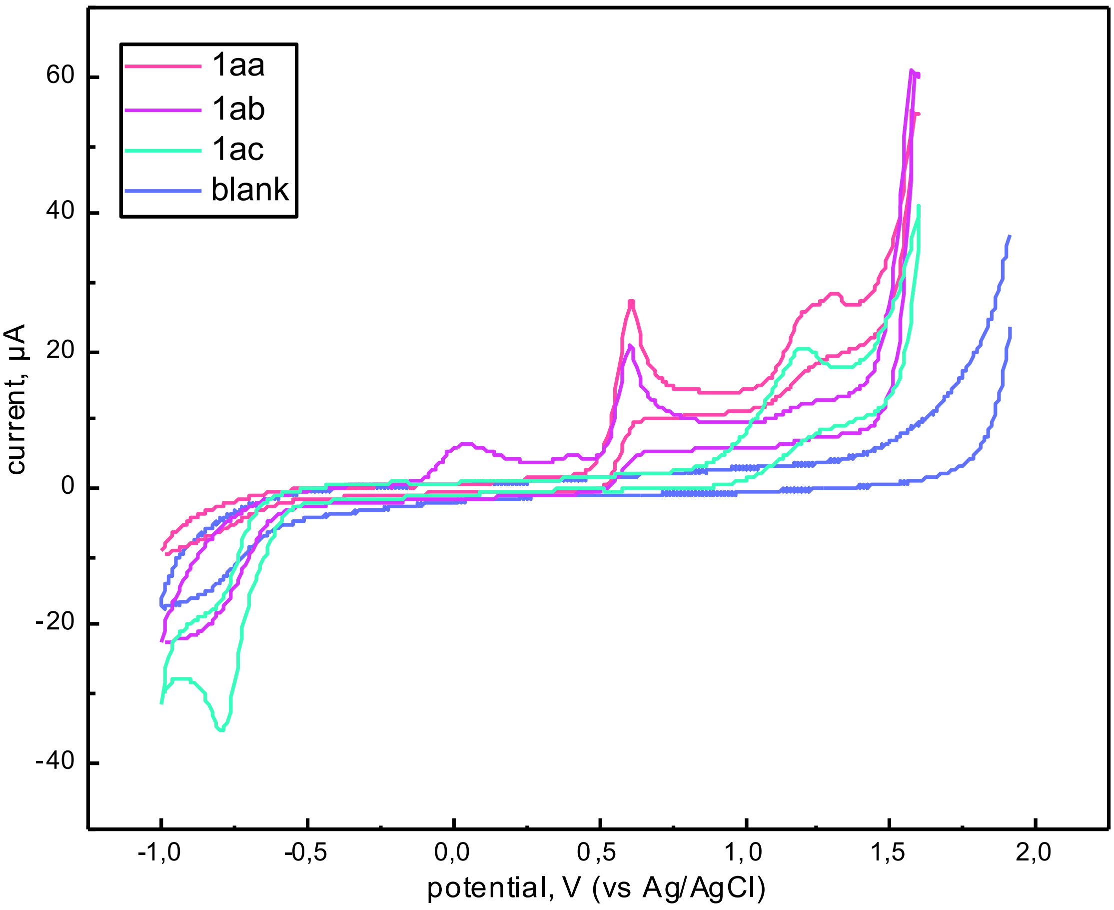
1,5
2,0
potential, V (vs Ag/AgCl)


1o
1p
blank
current, µA
40
20
0
-20
-40
-60
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)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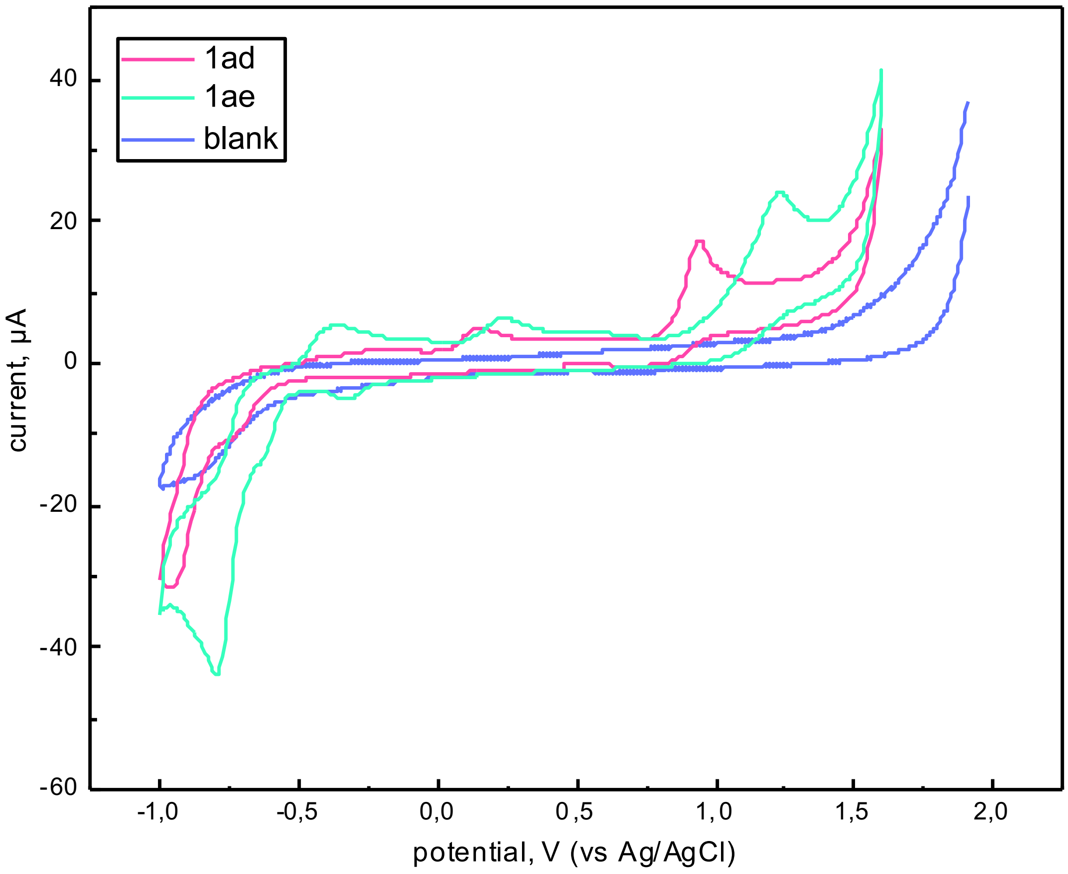
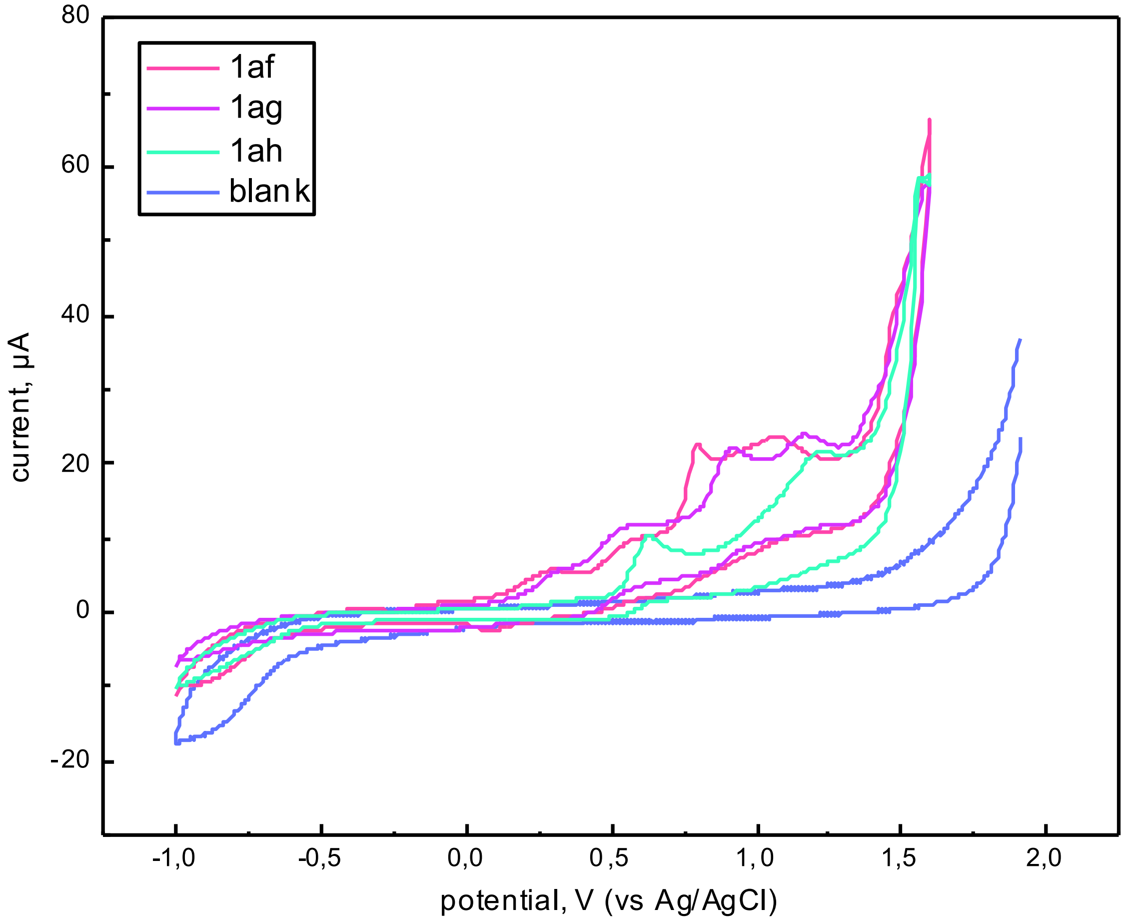
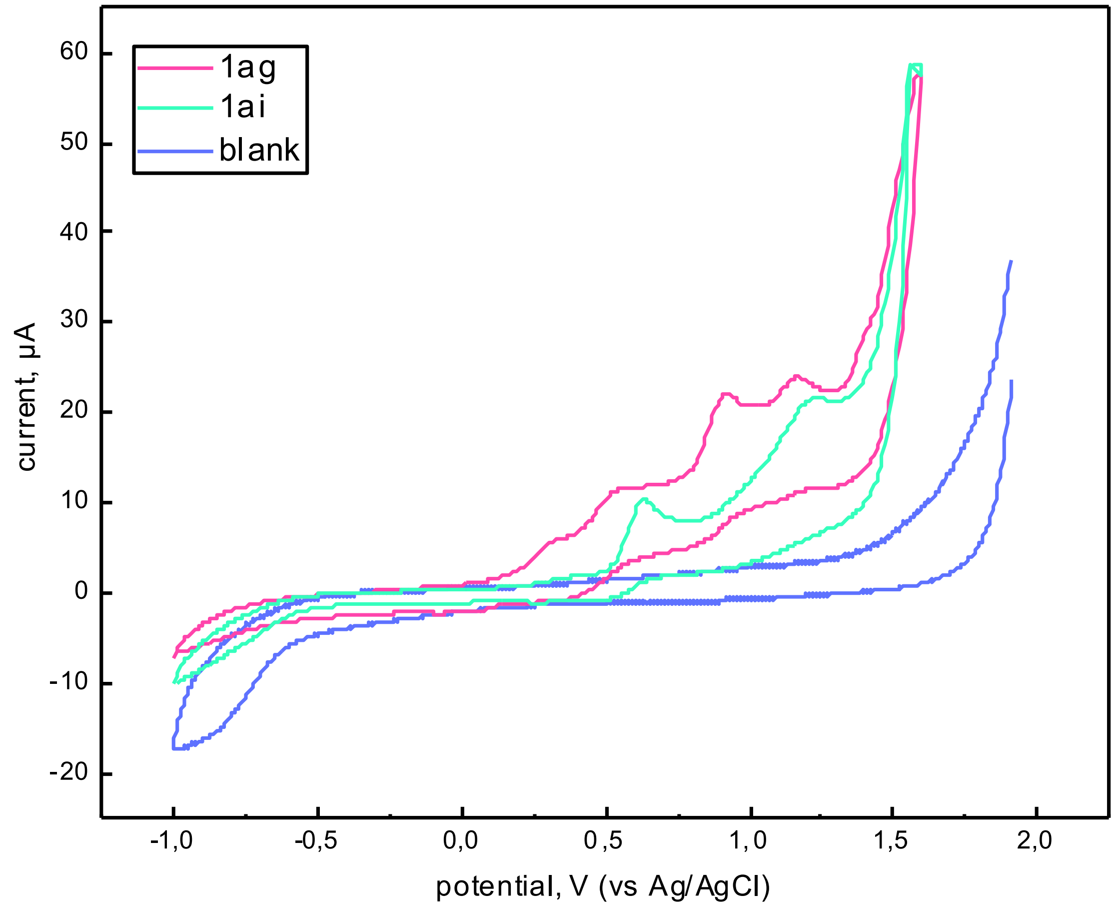
1ab
1ac
blank
60
40
20
0
-20
-40
current, µA
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)

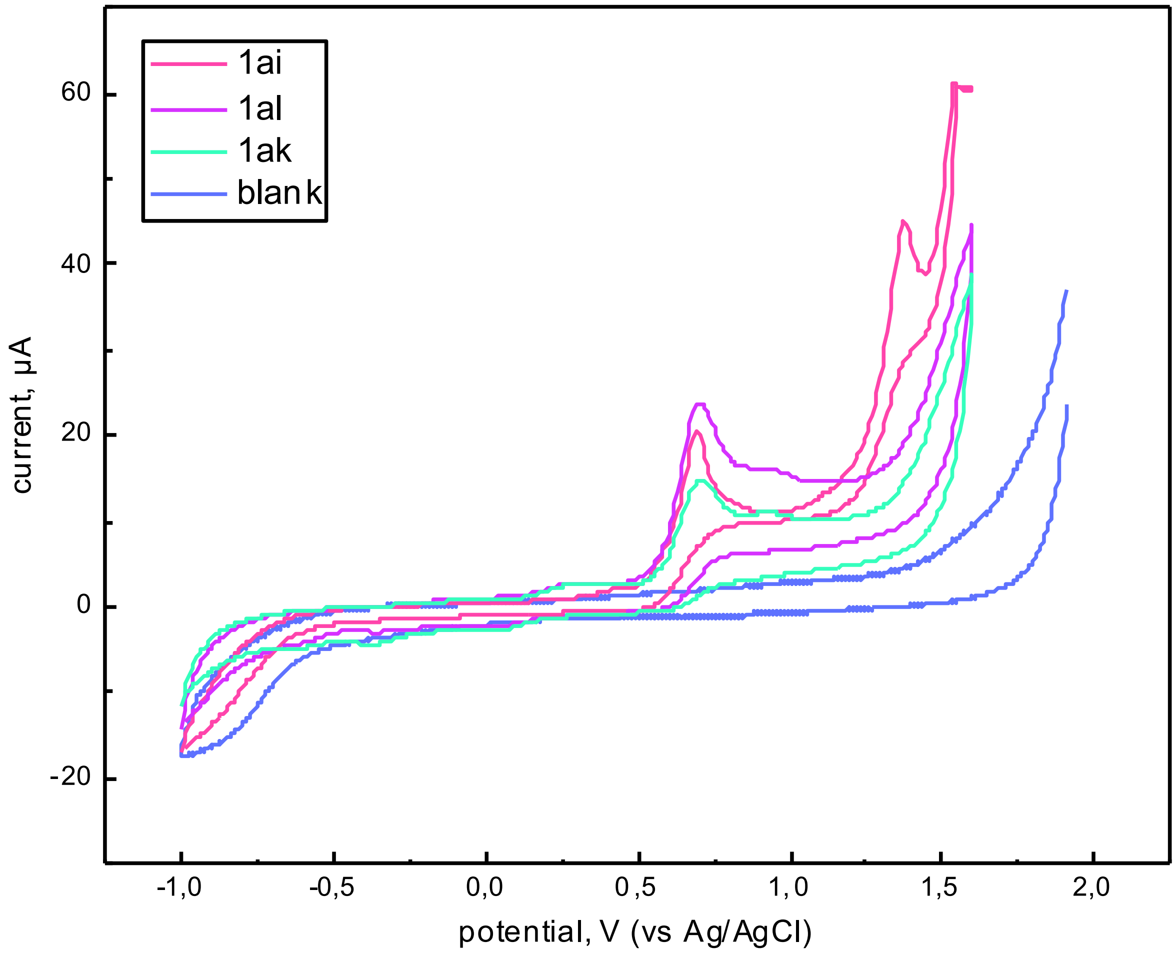

1ad
1ae
blank
40
20
0
-20
-40
-60
current, µA
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)

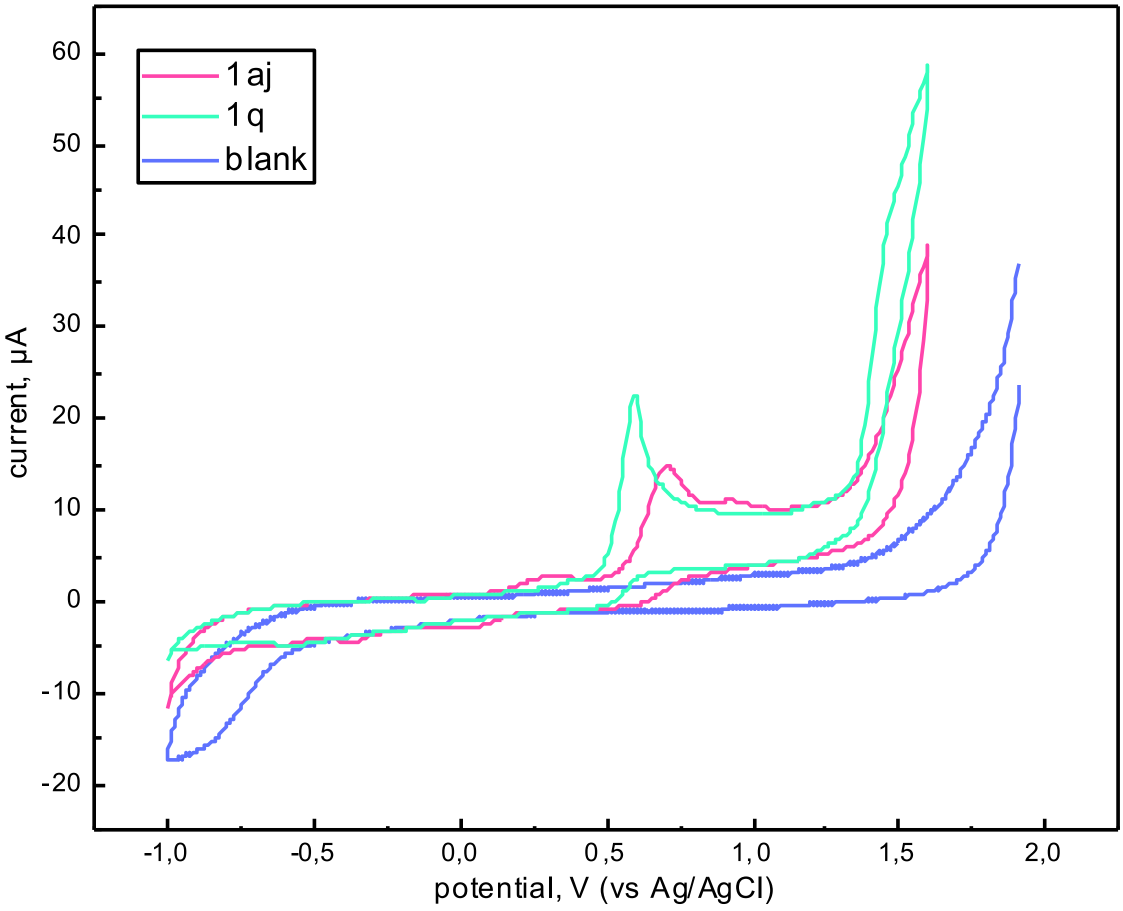


1af
1ag
1ah
blank
current, µA
80
60
40
20
0
-20
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)



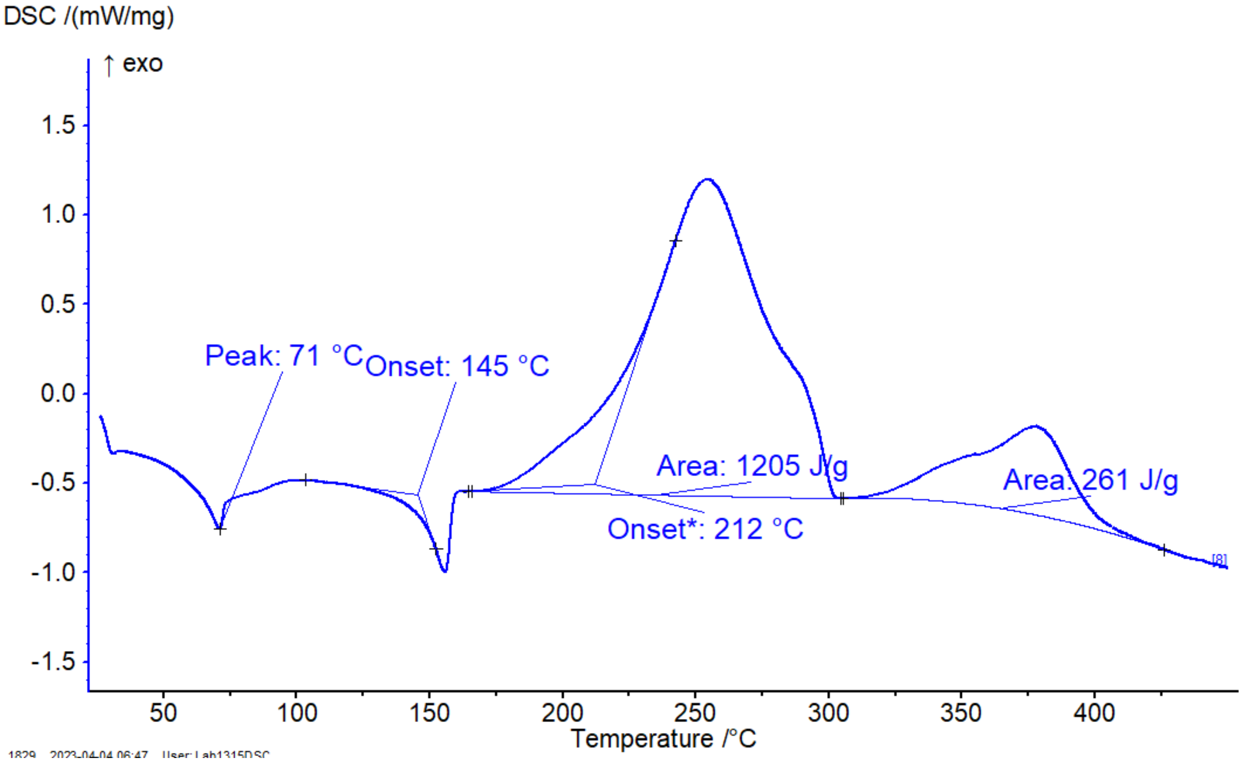
1ag
1ai
blank
current, µA
60
50
40
30
20
10
0
-10
-20
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0
potential, V (vs Ag/AgCl)


1ai
1al
1ak
blank
current, µA
potential, V (vs Ag/AgCl)
60
40
20
0
-20
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0

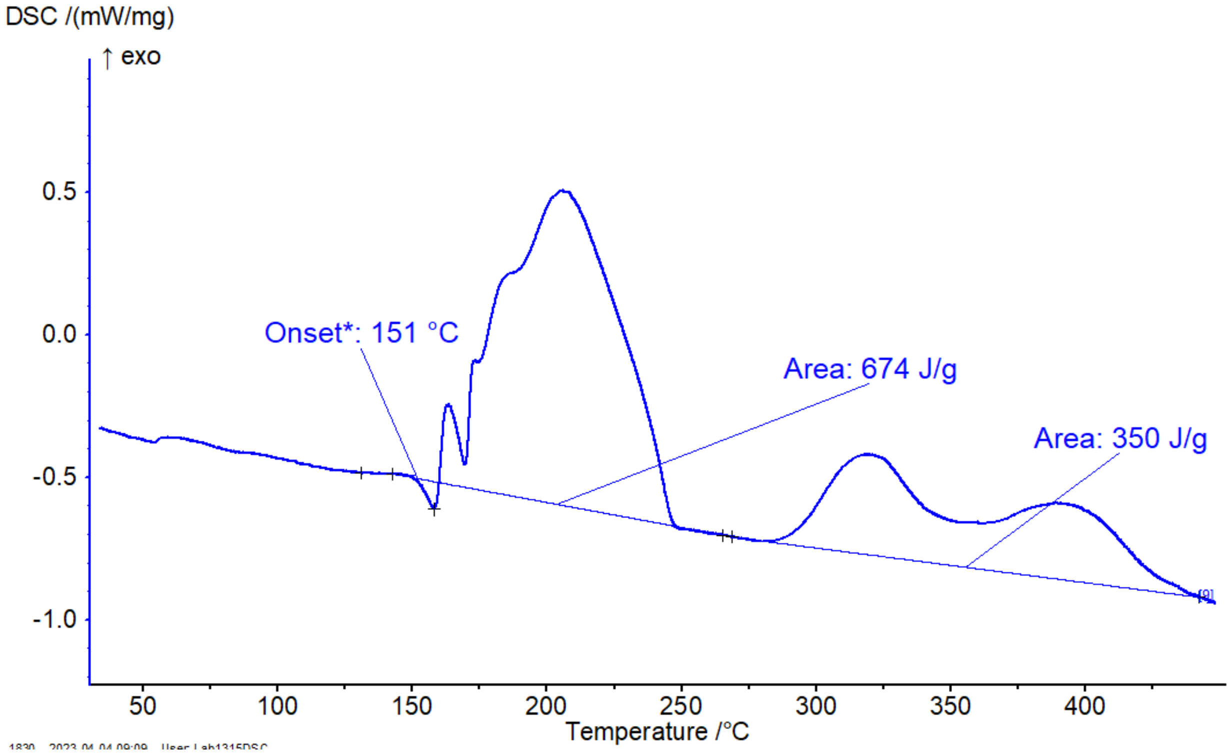

1aj
1q
blank
current, µA
potential, V (vs Ag/AgCl)
60
50
40
30
20
10
0
-10
-20
-1,0
-0,5
0,0
0,5
1,0
1,5
2,0

S5. DSC data

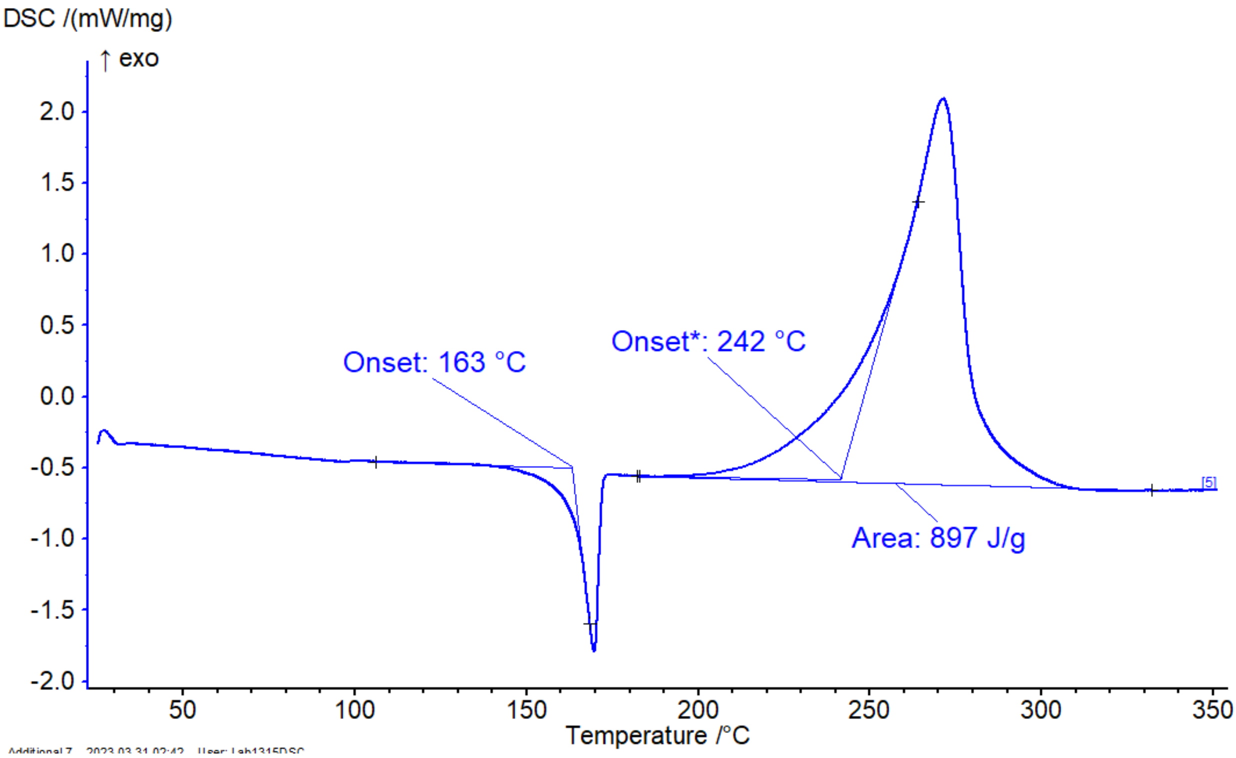
Compound **2n** reveals two endotherm prior to exothermic effect of thermolysis. The first endotherm (peak at 71°C) may correspond to the elimination of a solvent or phase transition of material. The second endotherm (onset 145°C) is apparently caused by melting of material. The latter peak is followed by the exothermic reaction of thermolysis, the overall heat effect is rather high, showing energetic potential of analyzed compound.



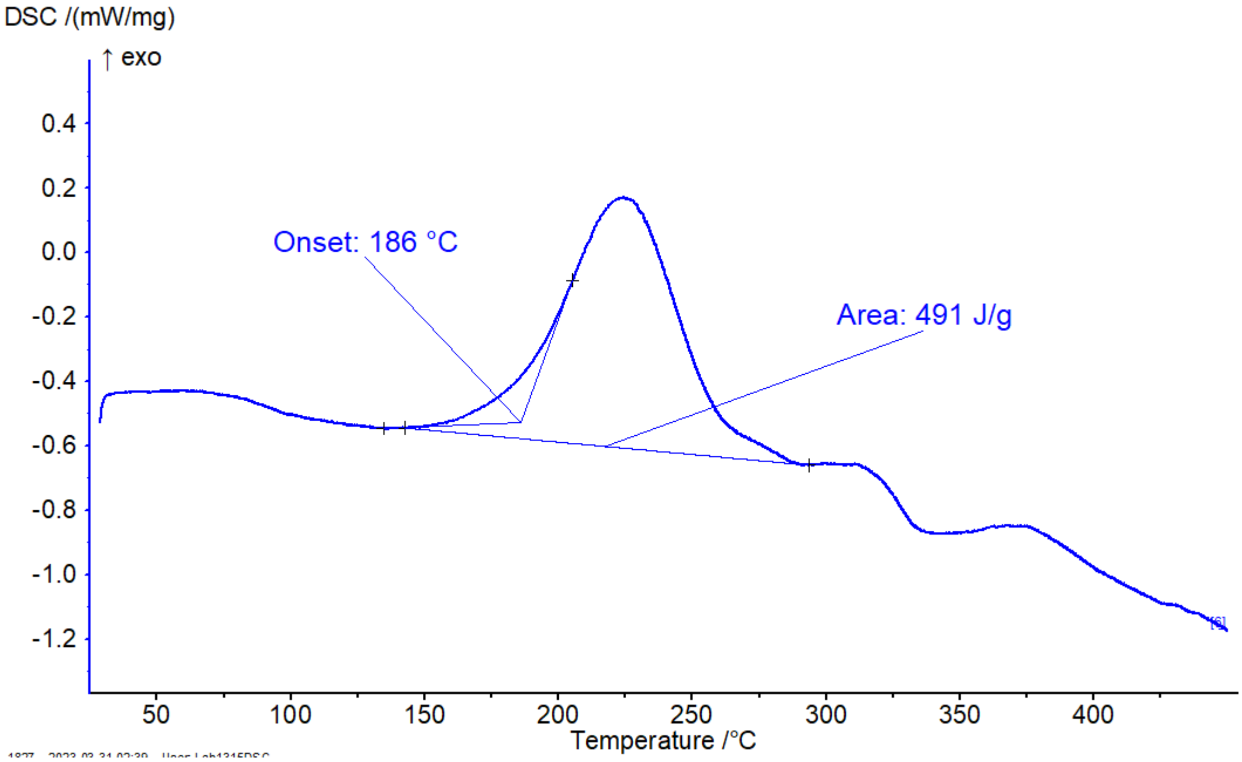
2o reveals thermal decomposition as a series of exothermic events. The initial dip of DSC curve is apparently melting of a starting compound (151°C)



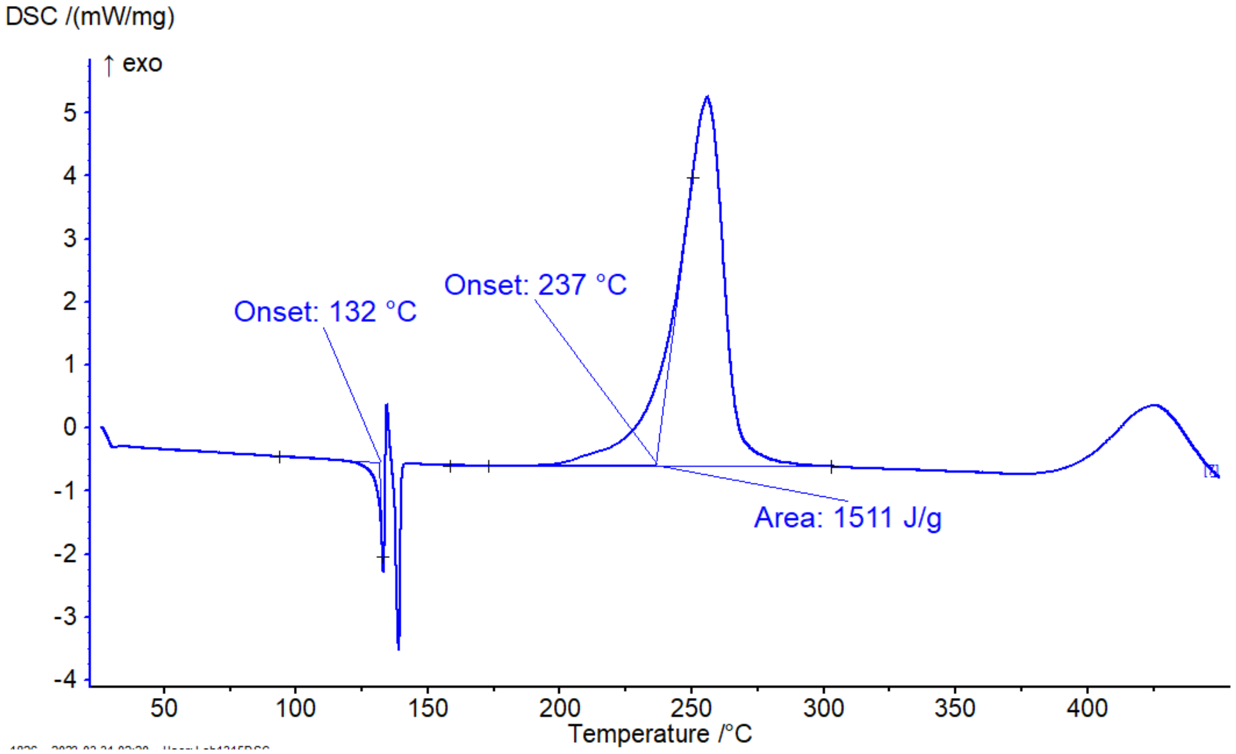
Compound **2p** melts at 163°C with subsequent decomposition (the extrapolated onset is 242°C, heat effect is 900 J g^{-1})



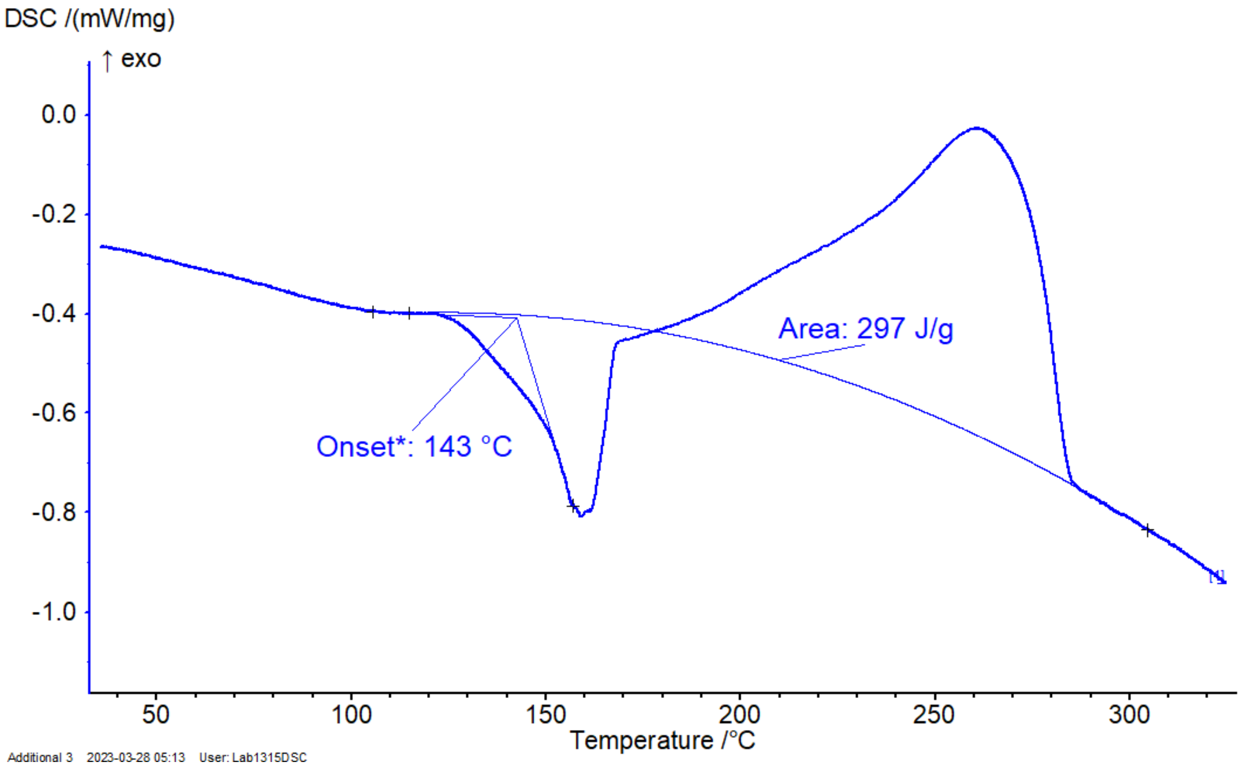
Compound **2q** reveals thermal decomposition, the extrapolated onset is 186°C at selected conditions.



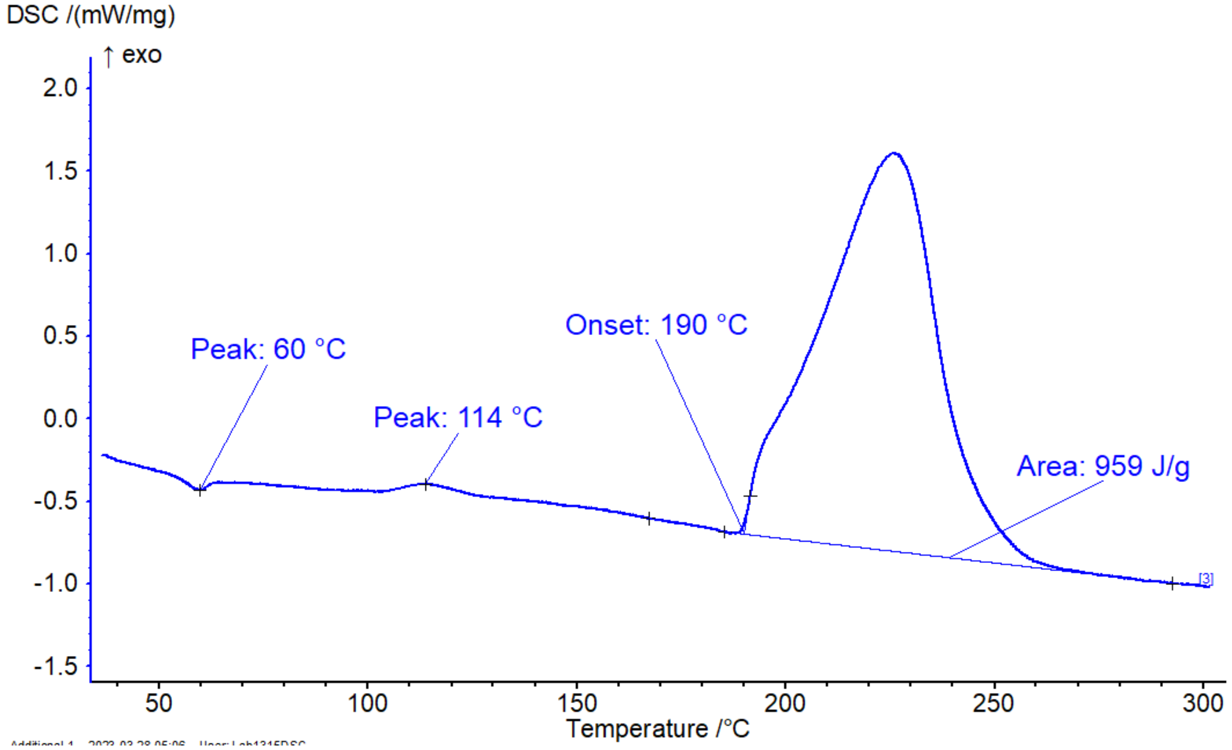
Compound **2ae** reveals a series of endothermic and exothermic events, the first of which corresponds to melting of the synthesized polymorph of the molecule. After melting, under further heating, compound decomposes with large heat release (1510 J g^{-1}).



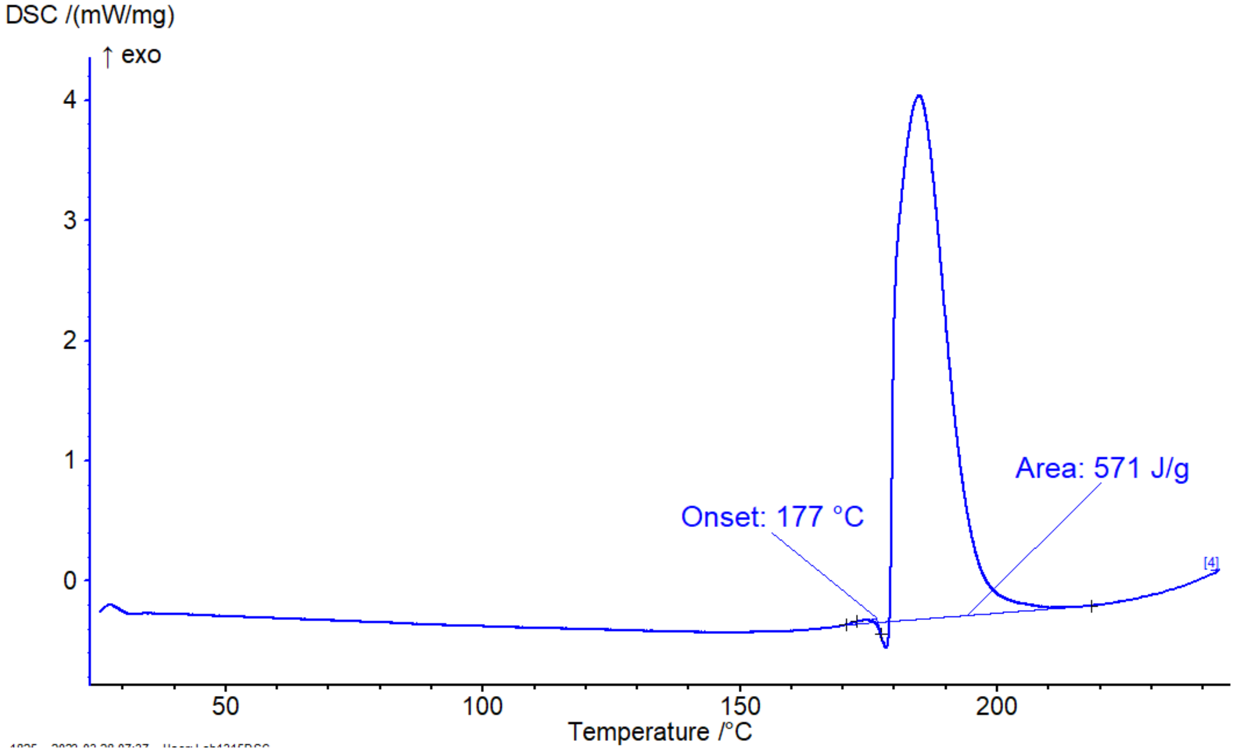
Compound **2ah** reveals first endothermic event followed by exothermic one. Based only on DSC data one cannot firmly conclude is it melting or endothermic first stage of thermolysis. The net heat release is not large.



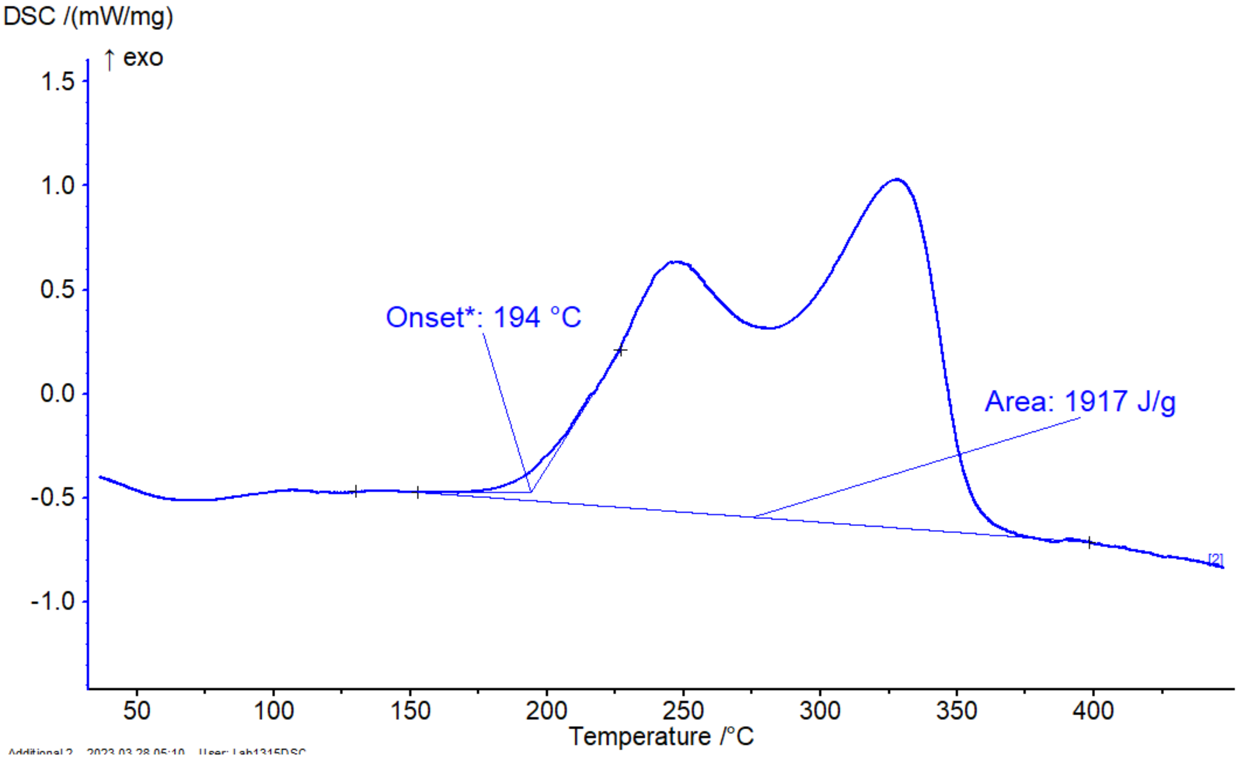
Compound **2ai** decomposes after 190°C under selected heating program, large heat release signifies the energetic character of material. Weak thermal effects around 60°C and 114°C are also observed.



Compound **2ak** melts at 177°C with subsequent thermal decomposition.



Compound **2al** shows thermal decomposition with extrapolated onset of 194°C and large heat release (1920 J g^{-1}).


DSC /(mW/mg)
↑ exo
1.5
1.0
0.5
0.0
-0.5
-1.0
Onset*: 194 °C
Area: 1917 J/g
[2]
50
100
150
200
250
300
350
400
Temperature /°C
Additional 2 2023.03.28 05:10 User: Lab1315DSC

S6. X-ray crystallographic data and refinement details

X-ray diffraction data were collected at 100K on a Bruker Quest D8 diffractometer equipped with a Photon-III area-detector (shutterless φ- and ω-scan technique), using graphite-monochromatized Mo K_{α}-radiation. The intensity data were integrated by the SAINT program[12] and were corrected for absorption and decay using SADABS.[13] The structure was solved by direct methods using SHELXT[14] and refined on F^2 using SHELXL-2018.[15] Positions of all atoms were found from the electron density-difference map. Atoms were refined with individual anisotropic (non-hydrogen atoms) or isotropic (hydrogen atoms) displacement parameters. The SHELXTL program suite[12] was used for molecular graphics. The CCDC 2258313 contains all additional supporting information.

Table S2. Crystal data and structure refinement for **2ah**.

Empirical formula	C20 H15 N3 O	
Formula weight	313.35	
Temperature	100.00 K	
Wavelength	0.71073 Å	
Crystal system	Orthorhombic	
Space group	Pbca	
Unit cell dimensions	a = 19.9852(4) Å	α= 90°.
	b = 7.55810(10) Å	β= 90°.
	c = 20.2911(4) Å	γ = 90°.
Volume	3064.97(10) Å^3	
Z	8	
Density (calculated)	1.358 g/cm^3	
Absorption coefficient	0.086 mm^{-1}	
F(000)	1312	
Crystal size	0.432 x 0.403 x 0.118 mm^3	
Theta range for data collection	2.251 to 31.498°.	
Index ranges	-29<=h<=29, -10<=k<=11, -29<=l<=29	
Reflections collected	60450	
Independent reflections	5095 [R(int) = 0.0562]	
Observed reflections	3933	
Completeness to theta = 25.242°	99.8 %	
Absorption correction	None	
Max. and min. transmission	0.7463 and 0.6962	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	5095 / 1 / 221	
Goodness-of-fit on F^2	1.026	
Final R indices [I>2sigma(I)]	R1 = 0.0497, wR2 = 0.1196	
R indices (all data)	R1 = 0.0687, wR2 = 0.1353	
Largest diff. peak and hole	0.413 and -0.282 e.Å^{-3}	

Table S3. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (Å^{2}x 10^3) for **2ah.** U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
N(1)	7607(1)	7405(1)	7258(1)	19(1)
N(2)	7600(1)	7022(2)	6598(1)	21(1)
N(3)	6974(1)	6771(1)	6376(1)	20(1)
C(4)	6576(1)	6958(2)	6902(1)	17(1)
C(5)	6961(1)	7343(2)	7468(1)	18(1)
C(6)	6803(1)	7527(2)	8170(1)	18(1)
C(7)	6336(1)	6404(2)	8465(1)	23(1)
C(8)	6209(1)	6530(2)	9137(1)	31(1)
C(9)	6545(1)	7749(2)	9522(1)	35(1)
C(10)	7012(1)	8861(2)	9232(1)	32(1)
C(11)	7140(1)	8759(2)	8561(1)	24(1)
C(12)	5846(1)	6823(2)	6822(1)	18(1)
C(13)	5580(1)	5836(2)	6301(1)	22(1)
C(14)	4892(1)	5751(2)	6206(1)	25(1)
C(15)	4461(1)	6641(2)	6630(1)	23(1)
C(16)	4722(1)	7615(2)	7151(1)	21(1)
C(17)	5410(1)	7713(2)	7247(1)	19(1)
C(18)	8180(1)	6938(2)	6192(1)	19(1)
C(19)	8770(1)	6215(2)	6436(1)	21(1)
C(20)	9324(1)	6122(2)	6022(1)	25(1)
C(21)	9284(1)	6730(2)	5377(1)	25(1)
C(22)	8688(1)	7423(2)	5140(1)	25(1)
C(23)	8131(1)	7546(2)	5548(1)	23(1)
O(1)	8142(1)	7724(2)	7569(1)	24(1)
O(1A)	6843(4)	6507(13)	5778(2)	24(1)

Table S4. Bond lengths [Å] and angles [°] for **2ah**.

N(1)-N(2)	1.3715(14)
N(1)-C(5)	1.3603(15)
N(1)-O(1)	1.2635(13)
N(2)-N(3)	1.3427(14)
N(2)-C(18)	1.4234(15)
N(3)-C(4)	1.3379(16)
N(3)-O(1A)	1.257(3)
C(4)-C(5)	1.4108(16)
C(4)-C(12)	1.4735(16)
C(5)-C(6)	1.4662(17)
C(6)-C(7)	1.3968(17)
C(6)-C(11)	1.3961(18)
C(7)-H(7)	0.9500
C(7)-C(8)	1.3901(19)
C(8)-H(8)	0.9500
C(8)-C(9)	1.383(2)
C(9)-H(9)	0.9500
C(9)-C(10)	1.386(2)
C(10)-H(10)	0.9500
C(10)-C(11)	1.3879(19)
C(11)-H(11)	0.9500
C(12)-C(13)	1.3989(17)
C(12)-C(17)	1.3967(17)
C(13)-H(13)	0.9500
C(13)-C(14)	1.3898(18)
C(14)-H(14)	0.9500
C(14)-C(15)	1.3913(19)
C(15)-H(15)	0.9500
C(15)-C(16)	1.3901(19)
C(16)-H(16)	0.9500
C(16)-C(17)	1.3917(17)
C(17)-H(17)	0.9500
C(18)-C(19)	1.3902(17)
C(18)-C(23)	1.3896(17)
C(19)-H(19)	0.9500
C(19)-C(20)	1.3911(18)
C(20)-H(20)	0.9500
C(20)-C(21)	1.389(2)
C(21)-H(21)	0.9500
C(21)-C(22)	1.3884(19)

C(22)-H(22)	0.9500
C(22)-C(23)	1.3896(18)
C(23)-H(23)	0.9500

C(5)-N(1)-N(2)	106.70(9)
O(1)-N(1)-N(2)	122.52(10)
O(1)-N(1)-C(5)	130.78(11)
N(1)-N(2)-C(18)	124.45(10)
N(3)-N(2)-N(1)	111.52(9)
N(3)-N(2)-C(18)	124.01(10)
C(4)-N(3)-N(2)	105.74(10)
O(1A)-N(3)-N(2)	122.7(4)
O(1A)-N(3)-C(4)	131.5(4)
N(3)-C(4)-C(5)	110.30(10)
N(3)-C(4)-C(12)	119.54(11)
C(5)-C(4)-C(12)	130.12(11)
N(1)-C(5)-C(4)	105.71(10)
N(1)-C(5)-C(6)	120.28(10)
C(4)-C(5)-C(6)	133.82(11)
C(7)-C(6)-C(5)	120.16(11)
C(11)-C(6)-C(5)	120.81(11)
C(11)-C(6)-C(7)	118.95(12)
C(6)-C(7)-H(7)	120.0
C(8)-C(7)-C(6)	120.05(13)
C(8)-C(7)-H(7)	120.0
C(7)-C(8)-H(8)	119.6
C(9)-C(8)-C(7)	120.74(14)
C(9)-C(8)-H(8)	119.6
C(8)-C(9)-H(9)	120.3
C(8)-C(9)-C(10)	119.40(13)
C(10)-C(9)-H(9)	120.3
C(9)-C(10)-H(10)	119.8
C(9)-C(10)-C(11)	120.47(14)
C(11)-C(10)-H(10)	119.8
C(6)-C(11)-H(11)	119.8
C(10)-C(11)-C(6)	120.38(13)
C(10)-C(11)-H(11)	119.8
C(13)-C(12)-C(4)	119.77(11)
C(17)-C(12)-C(4)	121.03(11)
C(17)-C(12)-C(13)	119.16(11)
C(12)-C(13)-H(13)	119.9
C(14)-C(13)-C(12)	120.30(12)

C(14)-C(13)-H(13)	119.9
C(13)-C(14)-H(14)	119.8
C(13)-C(14)-C(15)	120.32(12)
C(15)-C(14)-H(14)	119.8
C(14)-C(15)-H(15)	120.2
C(16)-C(15)-C(14)	119.61(11)
C(16)-C(15)-H(15)	120.2
C(15)-C(16)-H(16)	119.8
C(15)-C(16)-C(17)	120.35(12)
C(17)-C(16)-H(16)	119.8
C(12)-C(17)-H(17)	119.9
C(16)-C(17)-C(12)	120.26(12)
C(16)-C(17)-H(17)	119.9
C(19)-C(18)-N(2)	120.17(11)
C(23)-C(18)-N(2)	118.14(11)
C(23)-C(18)-C(19)	121.67(12)
C(18)-C(19)-H(19)	120.7
C(18)-C(19)-C(20)	118.66(12)
C(20)-C(19)-H(19)	120.7
C(19)-C(20)-H(20)	119.8
C(21)-C(20)-C(19)	120.49(12)
C(21)-C(20)-H(20)	119.8
C(20)-C(21)-H(21)	120.0
C(22)-C(21)-C(20)	119.92(12)
C(22)-C(21)-H(21)	120.0
C(21)-C(22)-H(22)	119.7
C(21)-C(22)-C(23)	120.50(12)
C(23)-C(22)-H(22)	119.7
C(18)-C(23)-C(22)	118.75(12)
C(18)-C(23)-H(23)	120.6
C(22)-C(23)-H(23)	120.6

Table S5. Anisotropic displacement parameters (Å^{2}x 10^3) for **2ah.** The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2}U^{11} + ... + 2 h k a^* b^* U^{12}]$

	U^{11}	U^{22}	U^{33}	U^{23}	U^{13}	U^{12}
N(1)	15(1)	22(1)	19(1)	0(1)	-2(1)	-2(1)
N(2)	14(1)	30(1)	19(1)	-1(1)	-1(1)	-1(1)
N(3)	14(1)	24(1)	22(1)	0(1)	-2(1)	-1(1)
C(4)	15(1)	17(1)	21(1)	0(1)	-1(1)	-1(1)
C(5)	15(1)	17(1)	21(1)	1(1)	0(1)	-2(1)
C(6)	16(1)	18(1)	20(1)	2(1)	-1(1)	1(1)
C(7)	19(1)	24(1)	26(1)	6(1)	0(1)	0(1)
C(8)	22(1)	42(1)	29(1)	15(1)	4(1)	6(1)
C(9)	29(1)	55(1)	20(1)	3(1)	1(1)	15(1)
C(10)	30(1)	40(1)	26(1)	-9(1)	-5(1)	7(1)
C(11)	22(1)	24(1)	25(1)	-2(1)	-2(1)	0(1)
C(12)	15(1)	16(1)	22(1)	1(1)	-1(1)	-1(1)
C(13)	18(1)	21(1)	26(1)	-3(1)	-2(1)	0(1)
C(14)	20(1)	26(1)	28(1)	-2(1)	-6(1)	-3(1)
C(15)	15(1)	26(1)	28(1)	6(1)	-2(1)	-1(1)
C(16)	17(1)	21(1)	25(1)	4(1)	2(1)	2(1)
C(17)	18(1)	18(1)	21(1)	2(1)	-1(1)	1(1)
C(18)	16(1)	20(1)	22(1)	-2(1)	2(1)	-1(1)
C(19)	18(1)	21(1)	25(1)	2(1)	-1(1)	1(1)
C(20)	18(1)	24(1)	32(1)	-1(1)	1(1)	3(1)
C(21)	21(1)	26(1)	29(1)	-5(1)	6(1)	1(1)
C(22)	26(1)	29(1)	21(1)	-1(1)	3(1)	2(1)
C(23)	20(1)	28(1)	21(1)	-1(1)	-1(1)	2(1)
O(1)	13(1)	35(1)	24(1)	-3(1)	-4(1)	-3(1)
O(1A)	13(1)	35(1)	24(1)	-3(1)	-4(1)	-3(1)

Table S6. Hydrogen coordinates (x 10^4) and isotropic displacement parameters ($Å^2$x 10 ^{3}) for **2ah**.

	x	y	z	U(eq)
H(7)	6104	5555	8206	28
H(8)	5887	5769	9333	37
H(9)	6458	7824	9981	42
H(10)	7245	9699	9495	38
H(11)	7459	9532	8367	29
H(13)	5872	5222	6010	26
H(14)	4716	5082	5850	29
H(15)	3991	6583	6563	28
H(16)	4429	8218	7443	25
H(17)	5585	8388	7602	23
H(19)	8794	5792	6876	26
H(20)	9731	5639	6181	30
H(21)	9666	6673	5098	30
H(22)	8660	7816	4696	30
H(23)	7724	8037	5389	27

Table S7. Torsion angles [°] for **2ah**.

N(1)-N(2)-N(3)-C(4)	-1.24(14)
N(1)-N(2)-N(3)-O(1A)	175.7(6)
N(1)-N(2)-C(18)-C(19)	38.48(18)
N(1)-N(2)-C(18)-C(23)	-143.43(13)
N(1)-C(5)-C(6)-C(7)	-135.94(12)
N(1)-C(5)-C(6)-C(11)	40.82(17)
N(2)-N(1)-C(5)-C(4)	-1.55(12)
N(2)-N(1)-C(5)-C(6)	174.14(10)
N(2)-N(3)-C(4)-C(5)	0.21(13)
N(2)-N(3)-C(4)-C(12)	177.86(10)
N(2)-C(18)-C(19)-C(20)	178.75(11)
N(2)-C(18)-C(23)-C(22)	-178.07(12)
N(3)-N(2)-C(18)-C(19)	-142.98(13)
N(3)-N(2)-C(18)-C(23)	35.11(18)
N(3)-C(4)-C(5)-N(1)	0.86(13)
N(3)-C(4)-C(5)-C(6)	-173.97(12)
N(3)-C(4)-C(12)-C(13)	25.82(17)
N(3)-C(4)-C(12)-C(17)	-152.02(12)
C(4)-C(5)-C(6)-C(7)	38.3(2)
C(4)-C(5)-C(6)-C(11)	-144.93(14)
C(4)-C(12)-C(13)-C(14)	-177.66(12)
C(4)-C(12)-C(17)-C(16)	177.93(11)
C(5)-N(1)-N(2)-N(3)	1.80(13)
C(5)-N(1)-N(2)-C(18)	-179.51(11)
C(5)-C(4)-C(12)-C(13)	-157.06(13)
C(5)-C(4)-C(12)-C(17)	25.10(19)
C(5)-C(6)-C(7)-C(8)	177.14(12)
C(5)-C(6)-C(11)-C(10)	-176.67(12)
C(6)-C(7)-C(8)-C(9)	-0.5(2)
C(7)-C(6)-C(11)-C(10)	0.13(19)
C(7)-C(8)-C(9)-C(10)	0.3(2)
C(8)-C(9)-C(10)-C(11)	0.2(2)
C(9)-C(10)-C(11)-C(6)	-0.4(2)
C(11)-C(6)-C(7)-C(8)	0.32(19)
C(12)-C(4)-C(5)-N(1)	-176.47(12)
C(12)-C(4)-C(5)-C(6)	8.7(2)
C(12)-C(13)-C(14)-C(15)	-0.2(2)
C(13)-C(12)-C(17)-C(16)	0.08(18)
C(13)-C(14)-C(15)-C(16)	-0.1(2)
C(14)-C(15)-C(16)-C(17)	0.44(19)

C(15)-C(16)-C(17)-C(12)	-0.41(18)
C(17)-C(12)-C(13)-C(14)	0.23(19)
C(18)-N(2)-N(3)-C(4)	-179.94(11)
C(18)-N(2)-N(3)-O(1A)	-3.0(6)
C(18)-C(19)-C(20)-C(21)	-0.45(19)
C(19)-C(18)-C(23)-C(22)	0.0(2)
C(19)-C(20)-C(21)-C(22)	-0.5(2)
C(20)-C(21)-C(22)-C(23)	1.3(2)
C(21)-C(22)-C(23)-C(18)	-1.0(2)
C(23)-C(18)-C(19)-C(20)	0.73(19)
O(1)-N(1)-N(2)-N(3)	-178.63(11)
O(1)-N(1)-N(2)-C(18)	0.07(19)
O(1)-N(1)-C(5)-C(4)	178.93(12)
O(1)-N(1)-C(5)-C(6)	-5.39(19)
O(1A)-N(3)-C(4)-C(5)	-176.4(6)
O(1A)-N(3)-C(4)-C(12)	1.3(7)

Table S8. Hydrogen bonds for **2ah** [Å and °].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
C(23)-H(23)...O(1A)	0.95	2.25	2.732(9)	110.5

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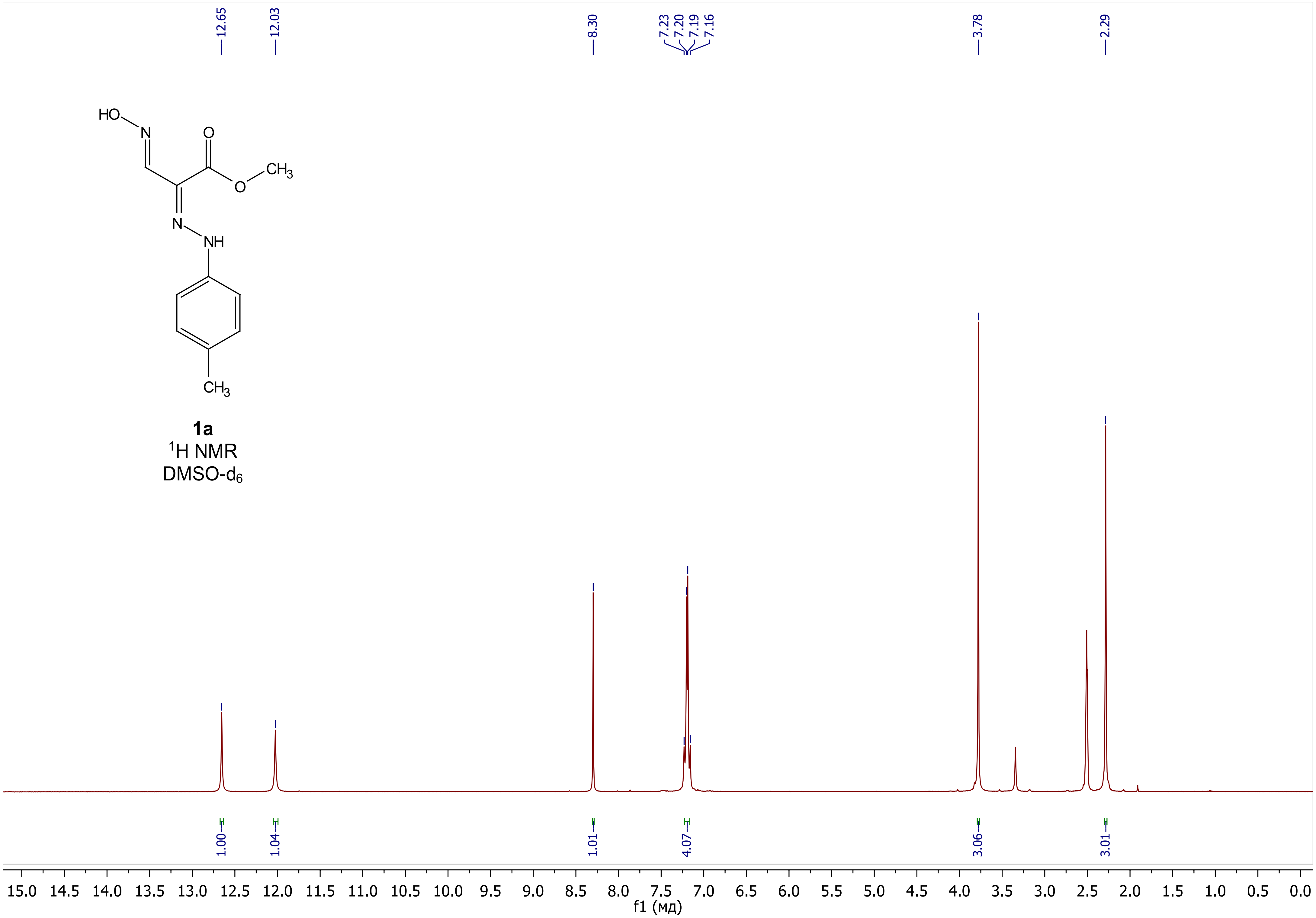
S8. Copies of NMR Spectra



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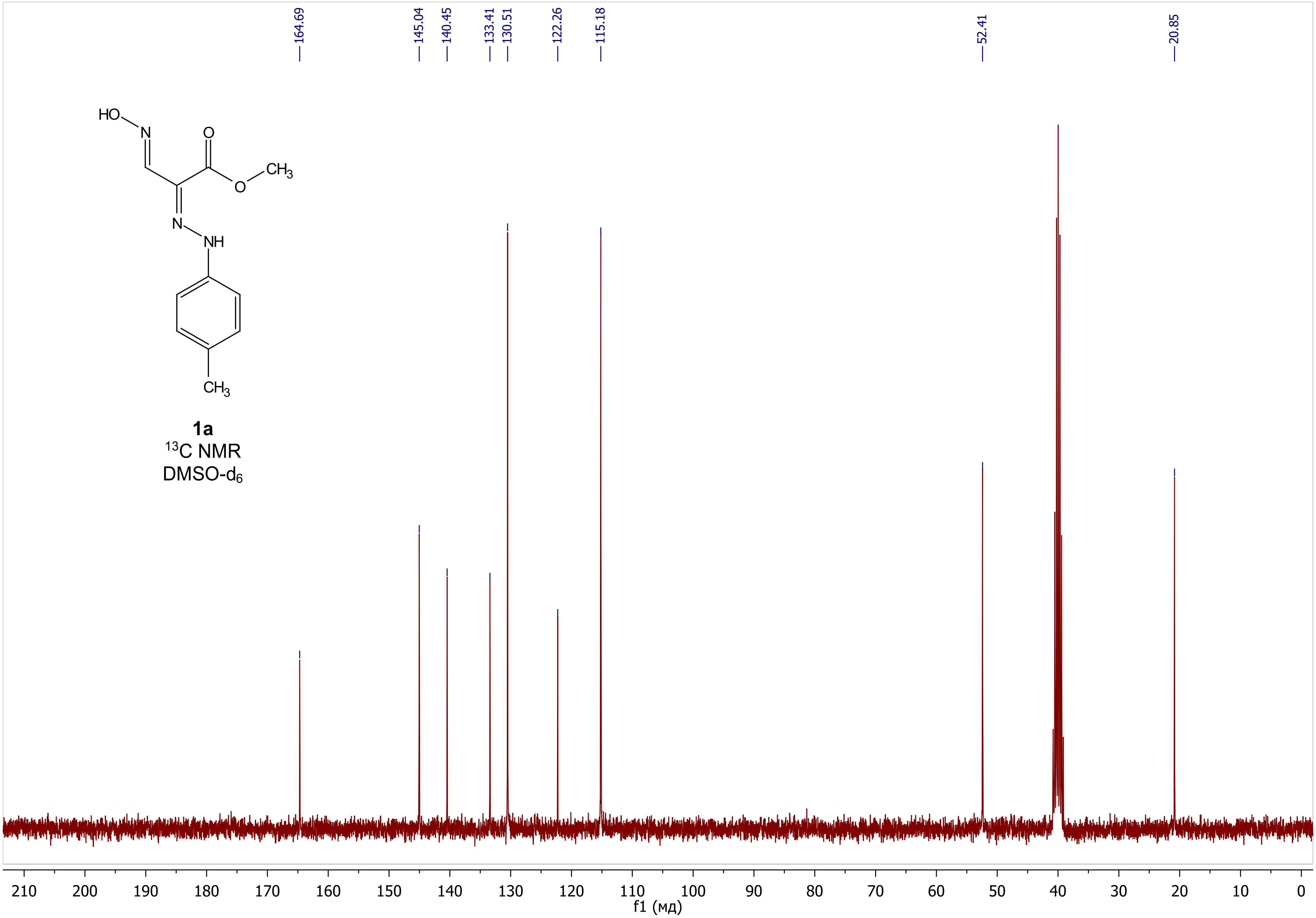
^{1}H NMR

DMSO-d_6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NH
CH3
1a
13C NMR
DMSO-d6
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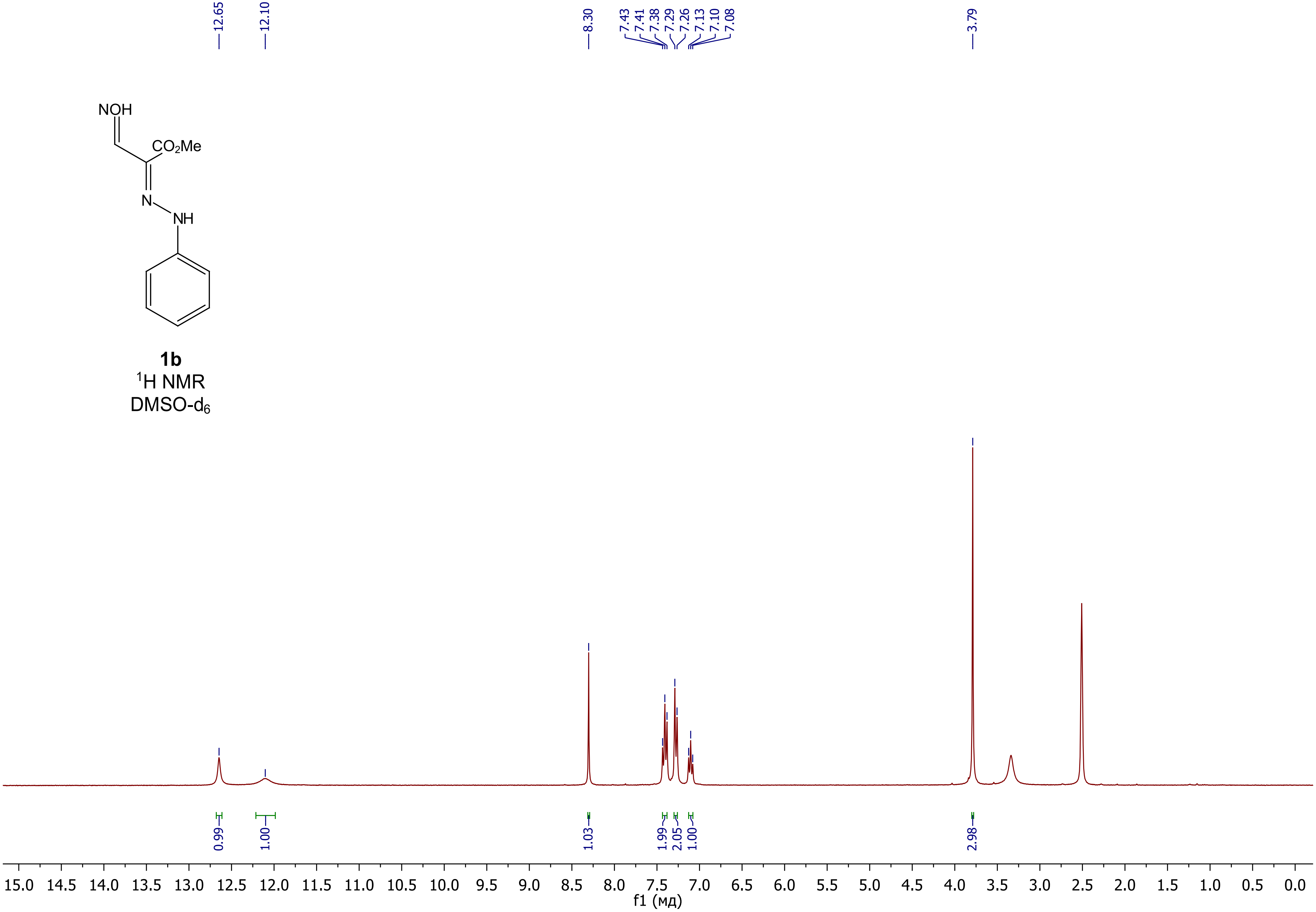
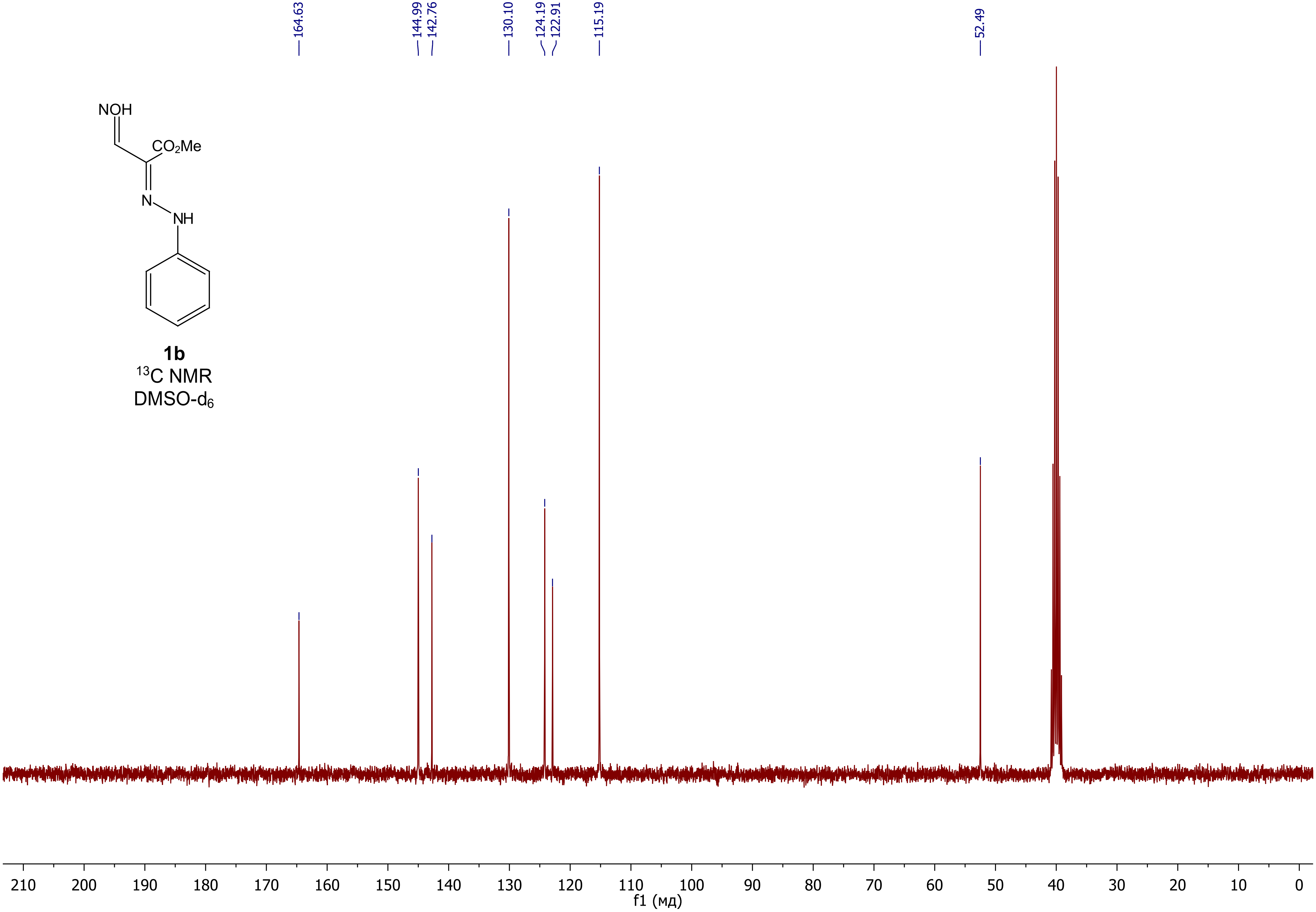


1b

^{1}H NMR

DMSO-d_6



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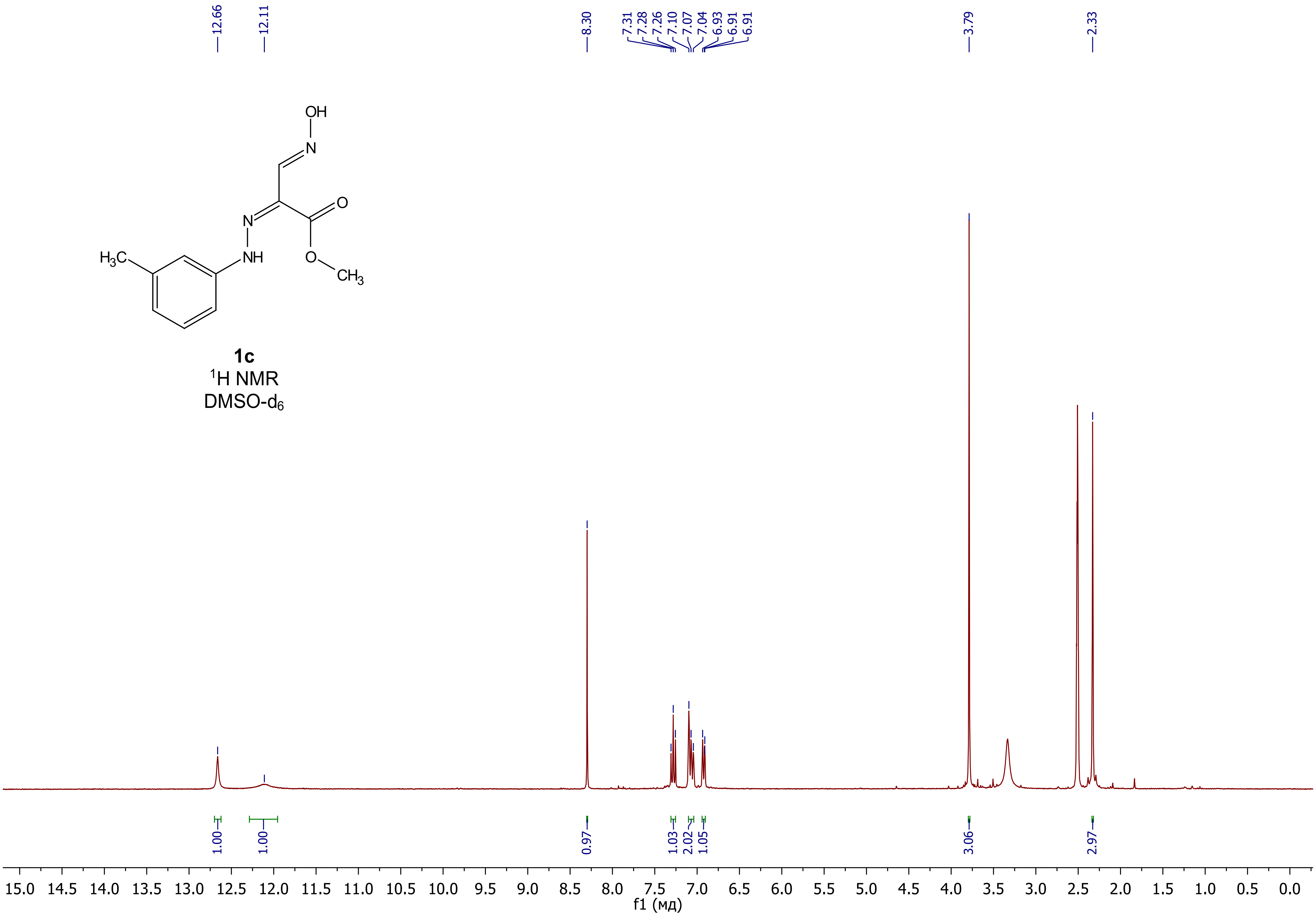

1c
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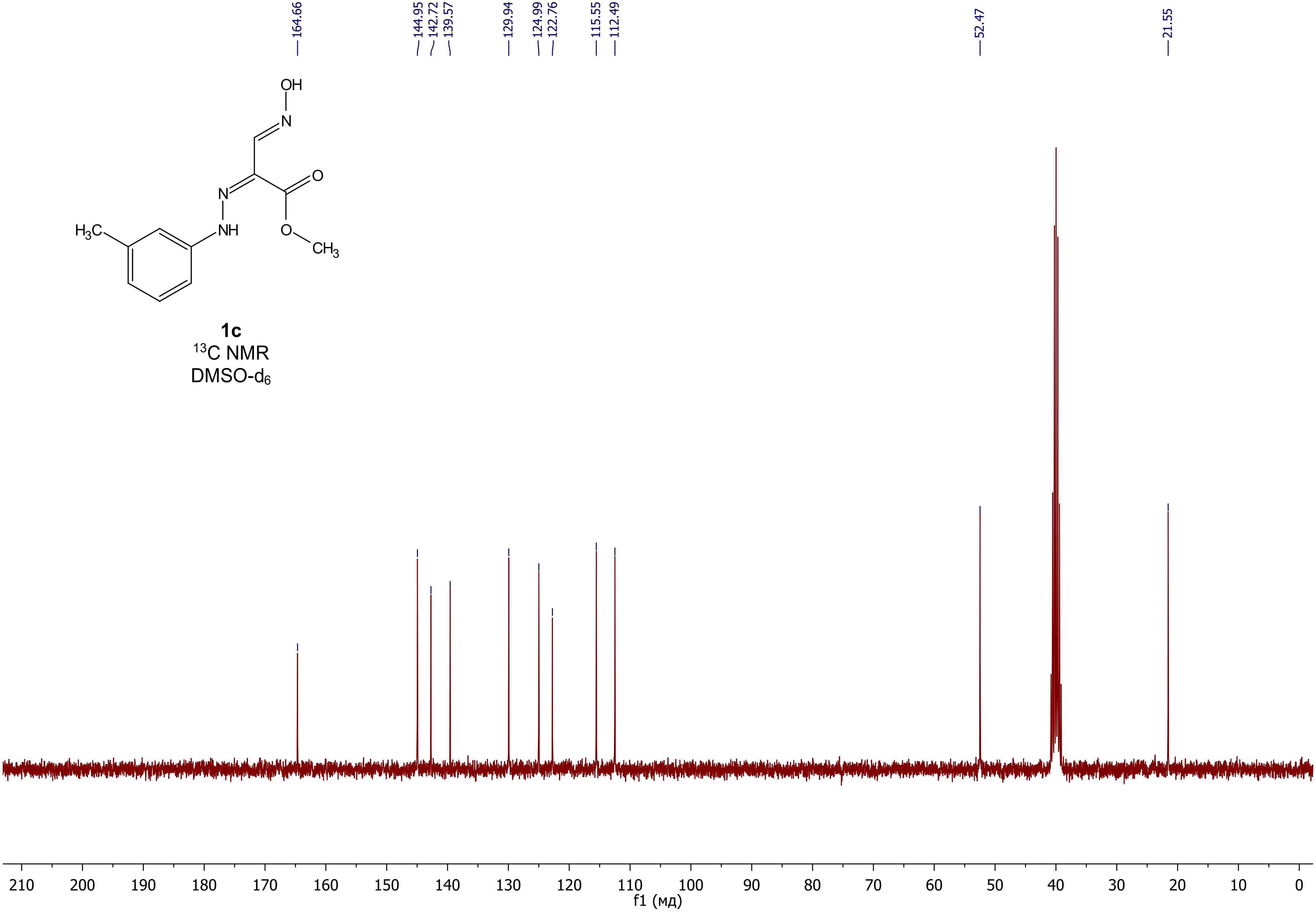

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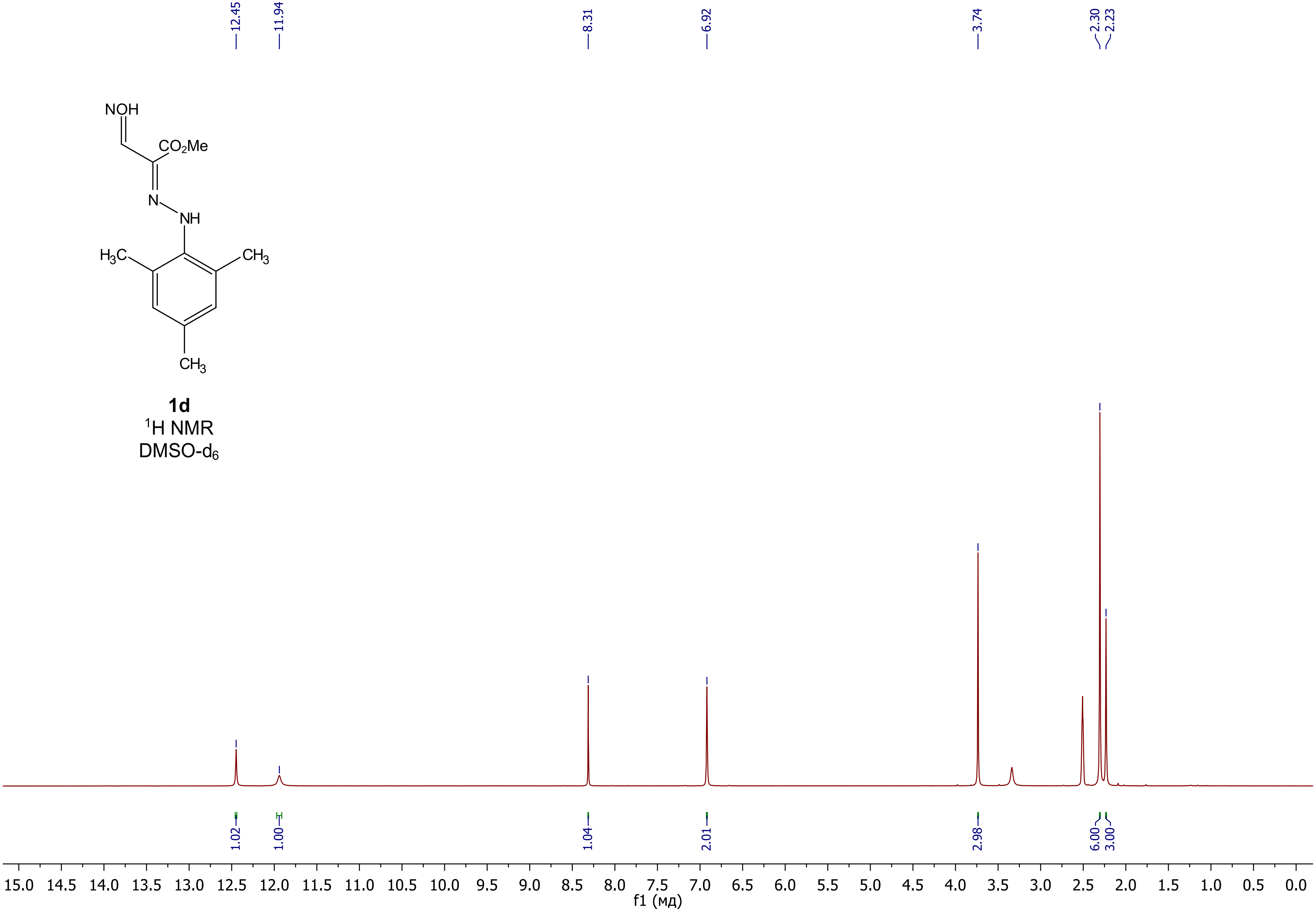

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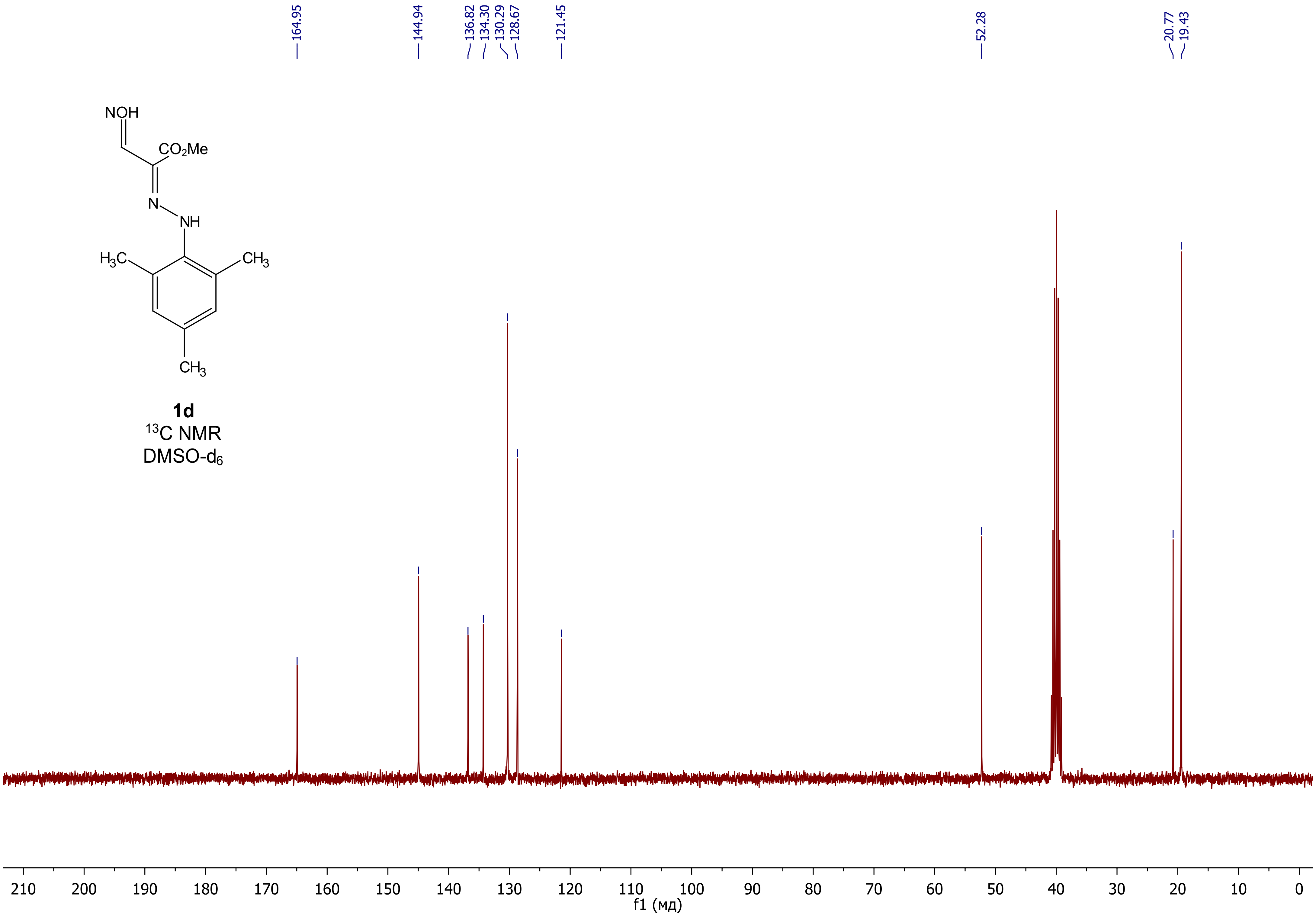

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H3C
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CH3
1d
13C NMR
DMSO-d6
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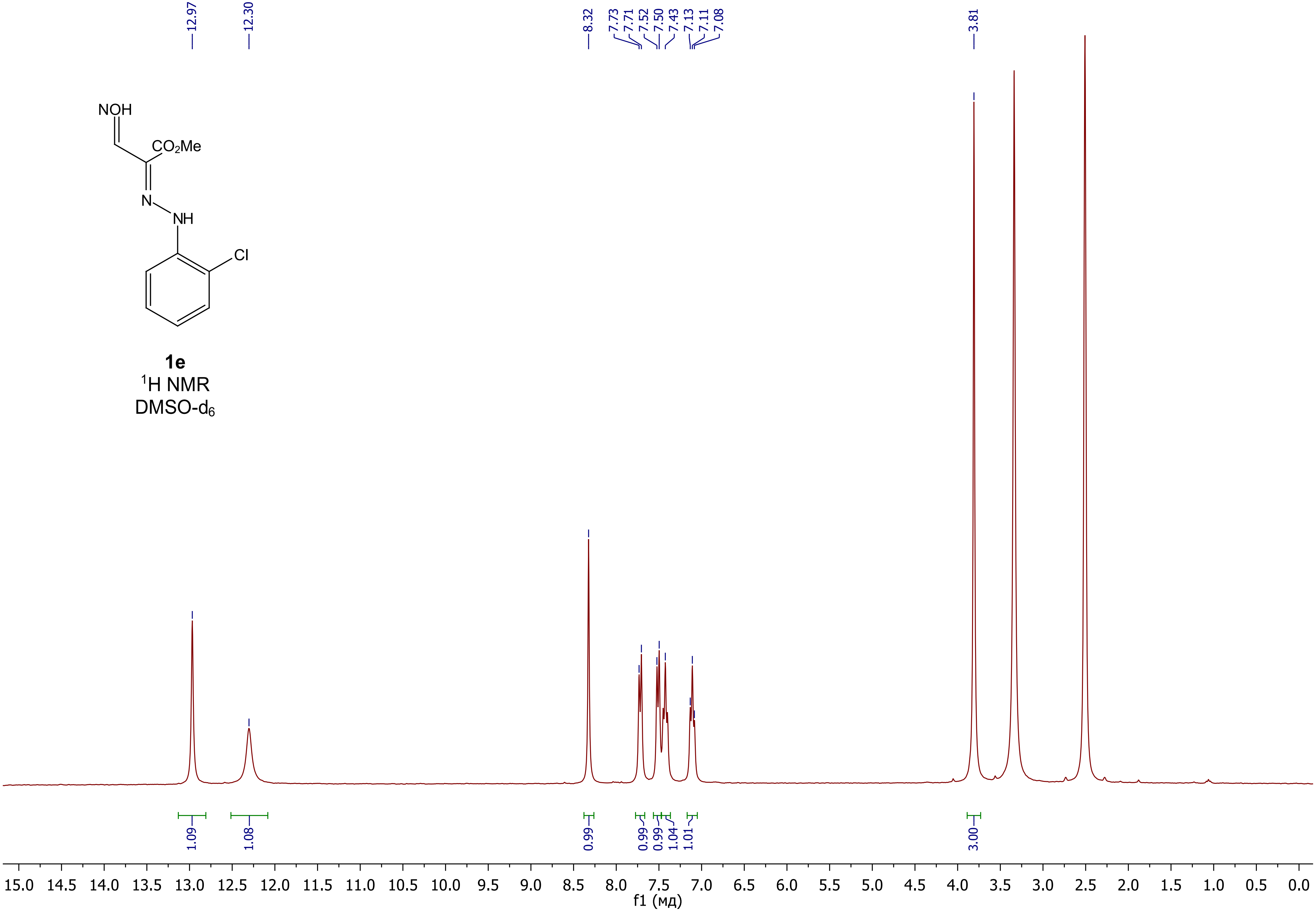

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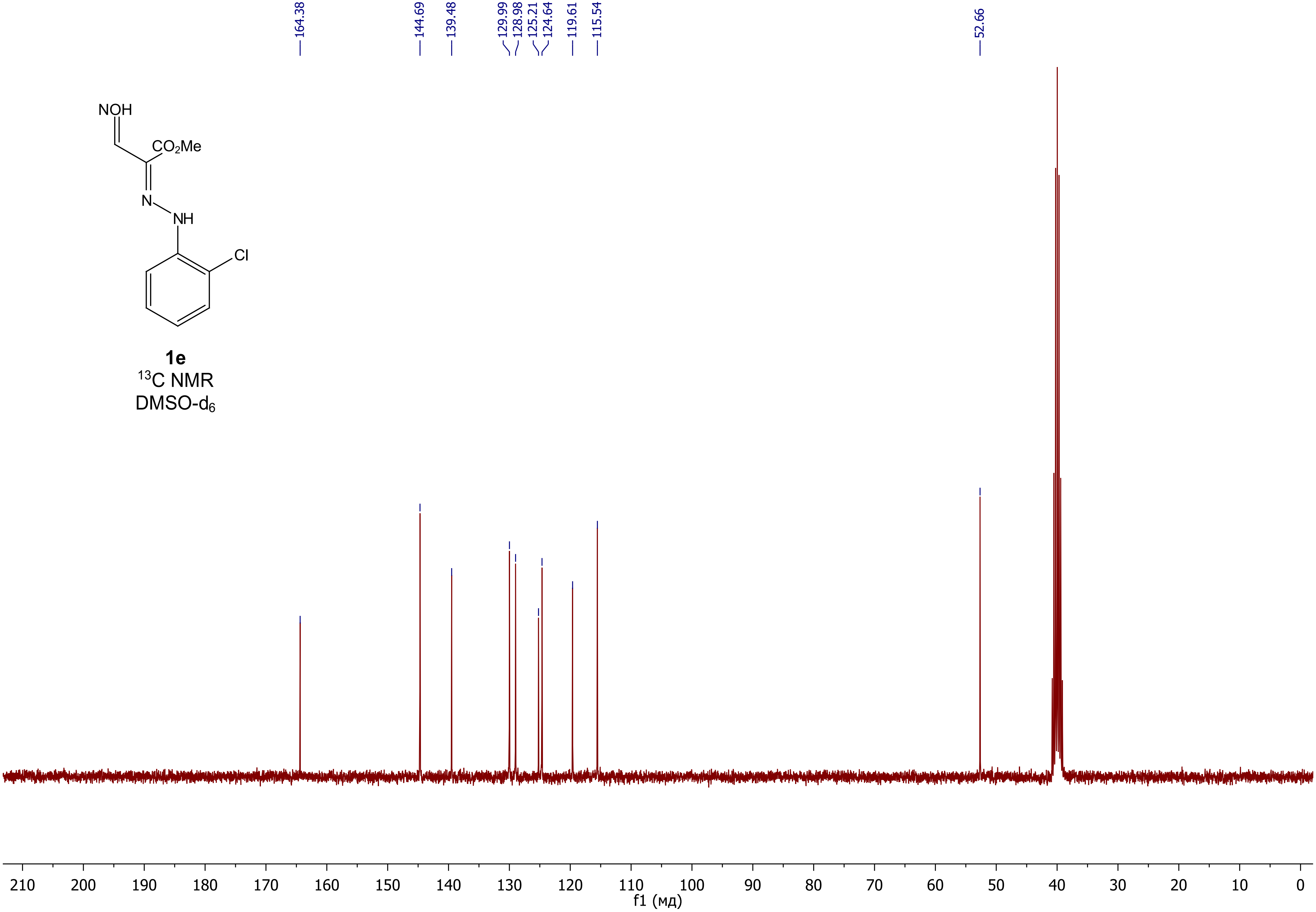
^{1}H NMR

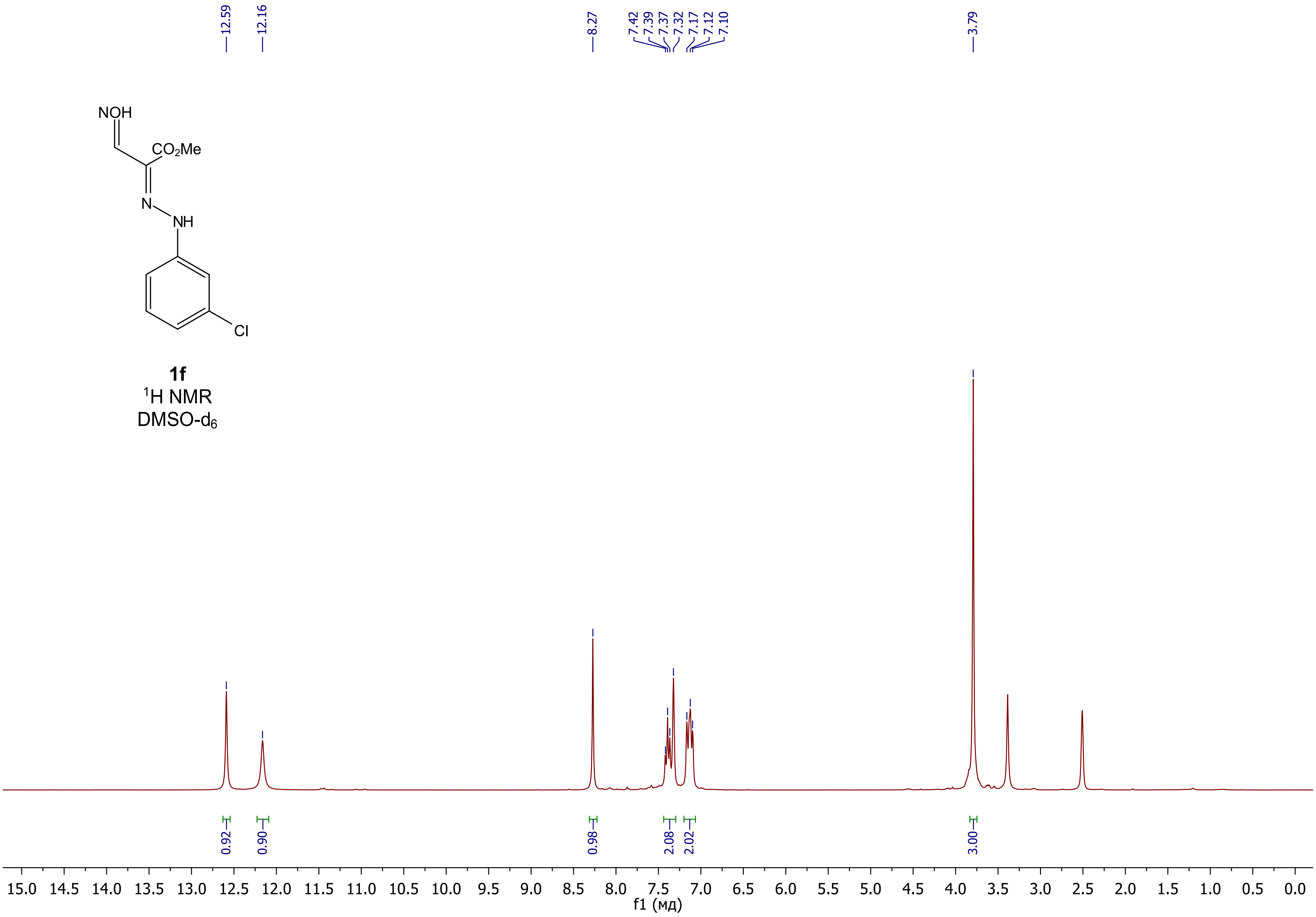
DMSO-d_6



1e

^{13}C NMR

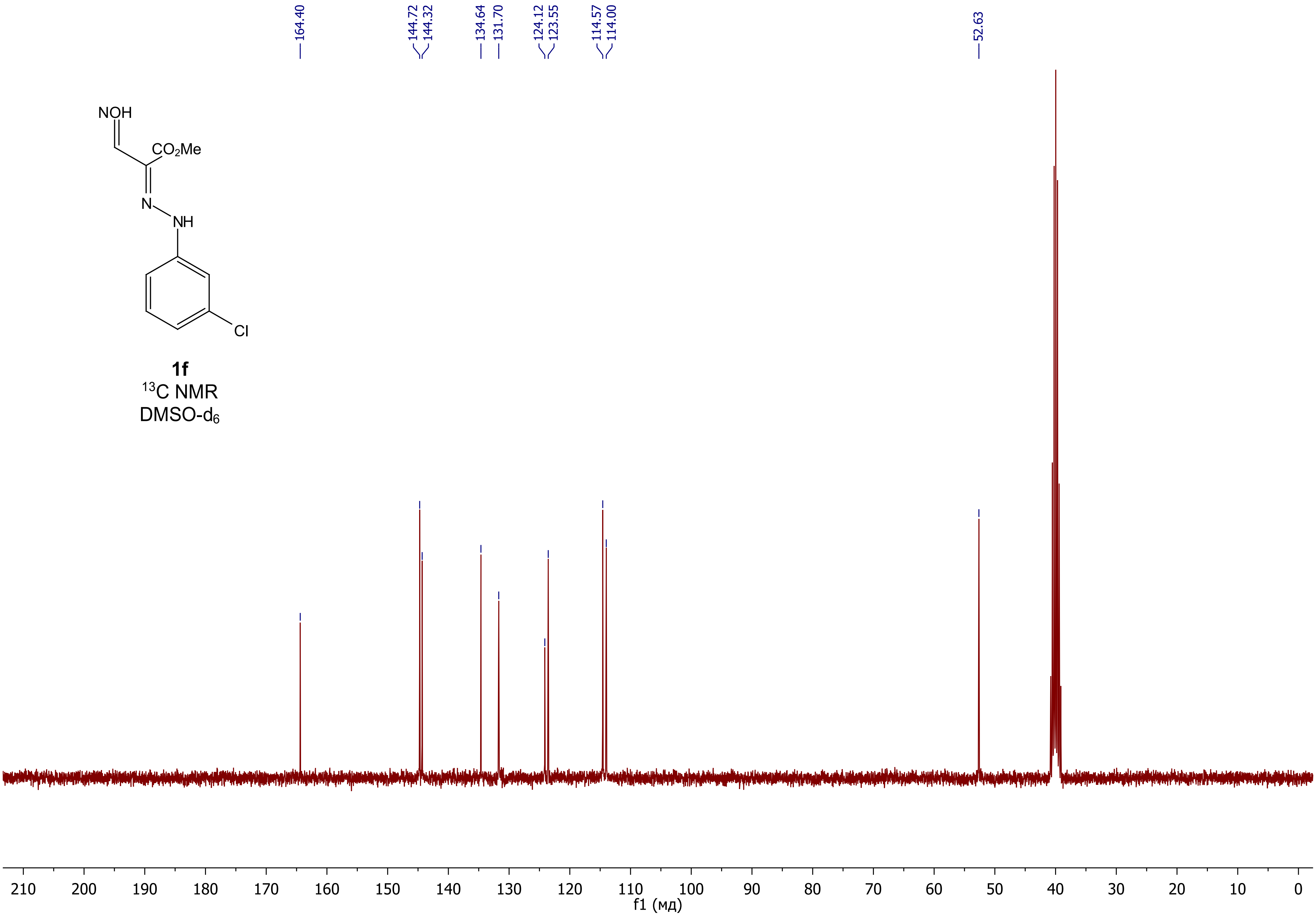
DMSO-d$_6$



1f

^{1}H NMR

DMSO-d_6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N
NH
Cl
1f
^{13}C NMR
DMSO-d$_6$
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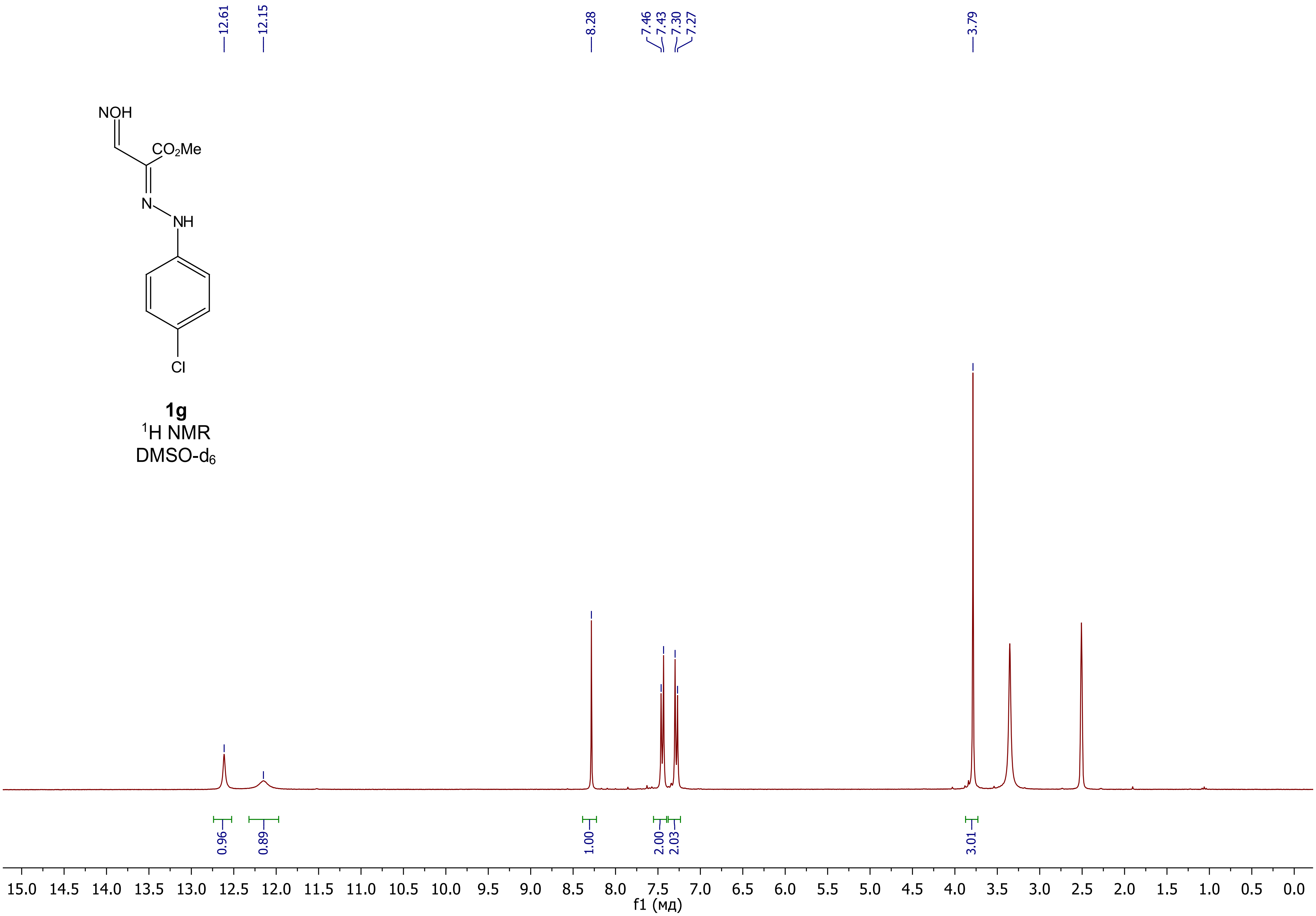
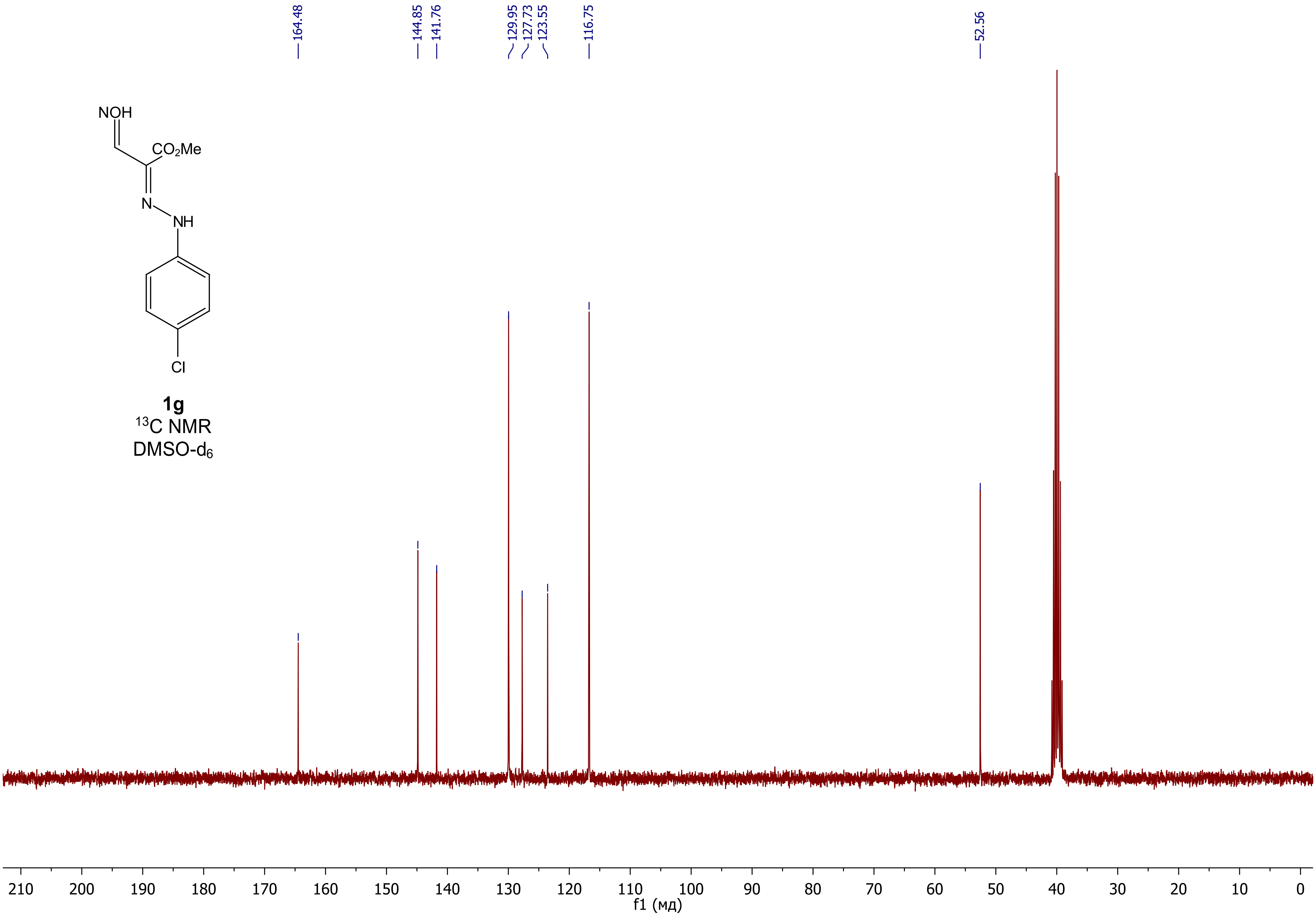
f1 (мд)



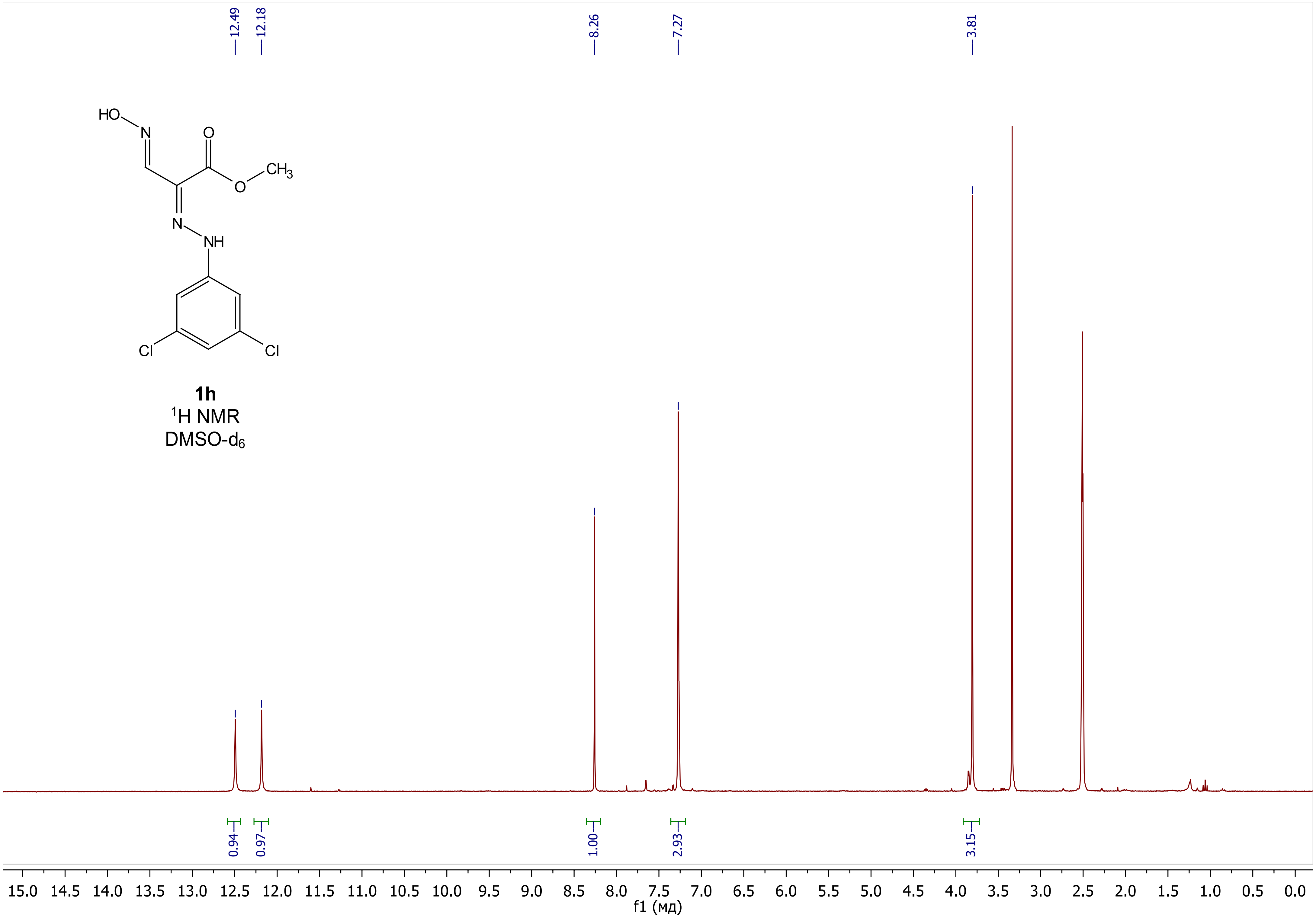
1g
^{1}H NMR
DMSO-d$_6$



1g

^{13}C NMR

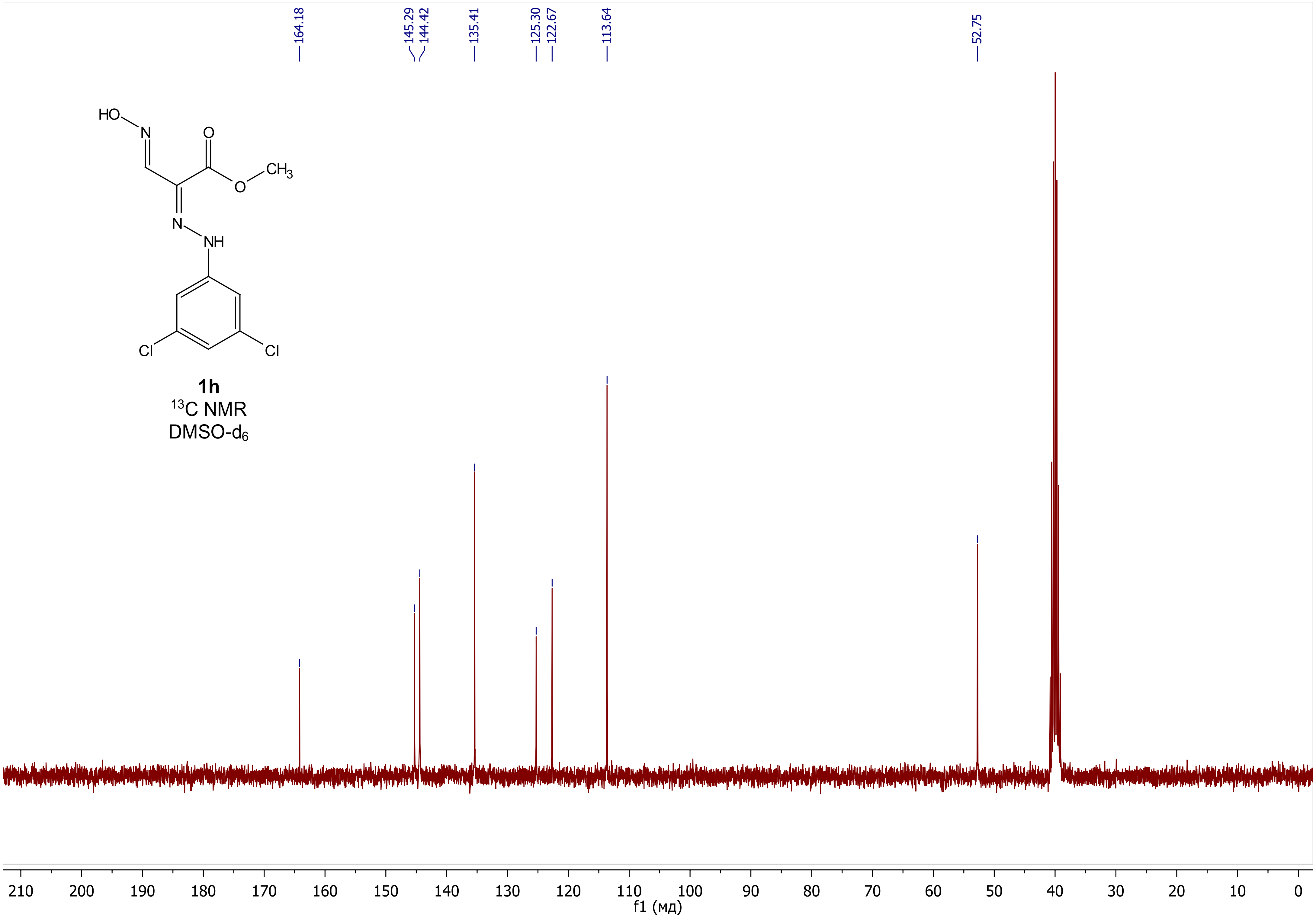
DMSO-d_6



1h

^{1}H NMR

DMSO-d_6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13C NMR
DMSO-d6
210
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180
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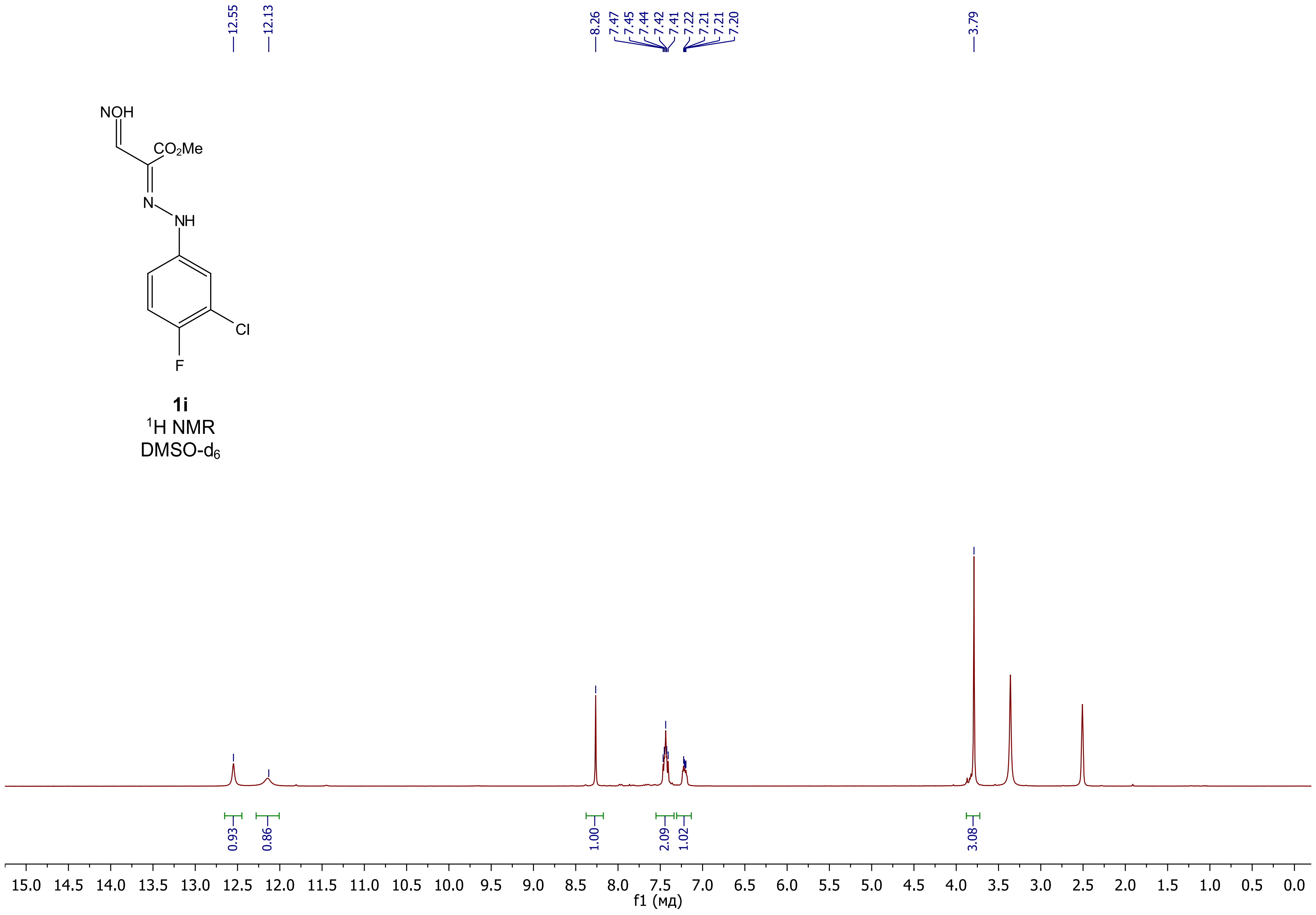
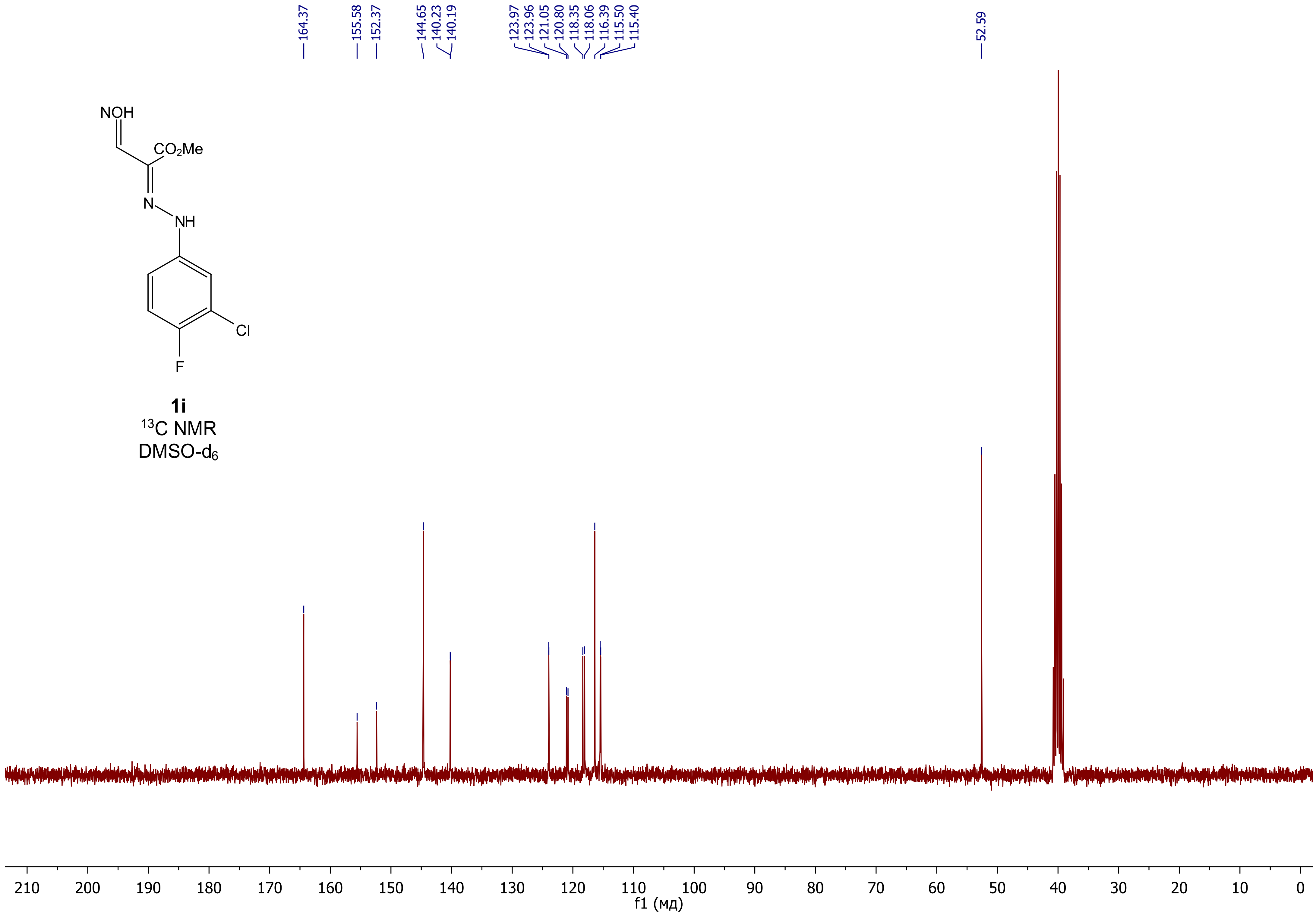
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)



1i

^{1}H NMR

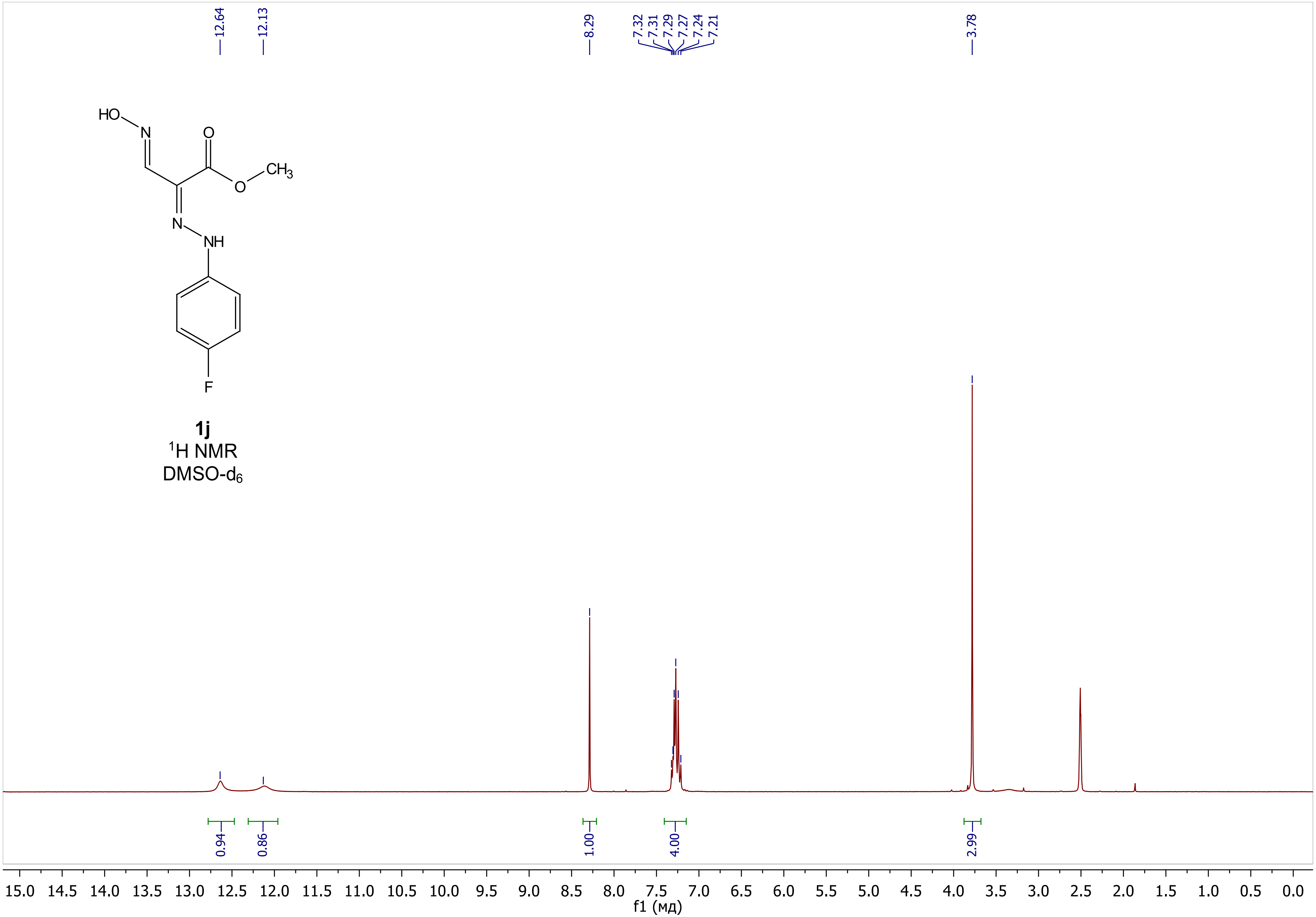
DMSO-d_6



1i

^{13}C NMR

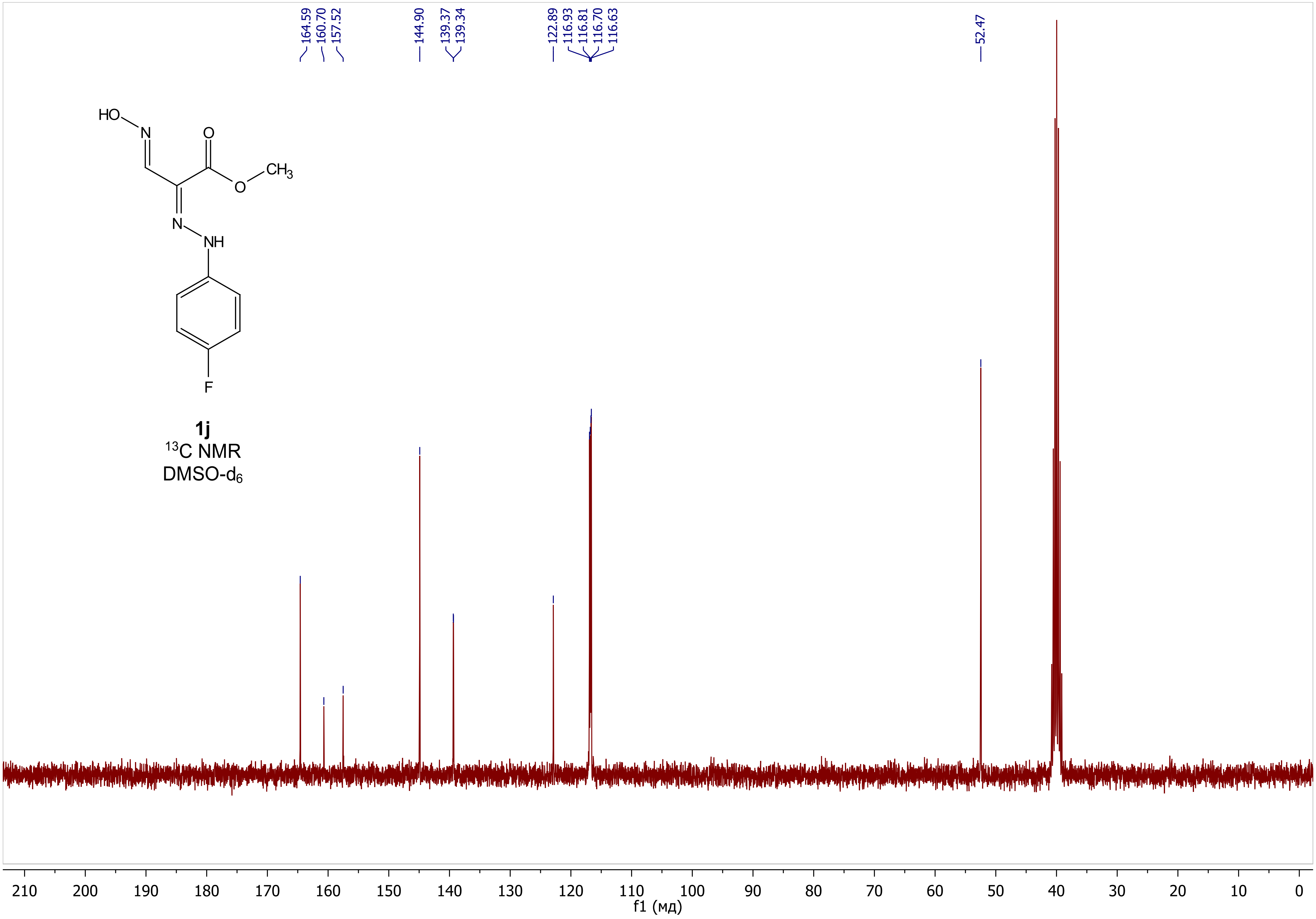
DMSO-d_6

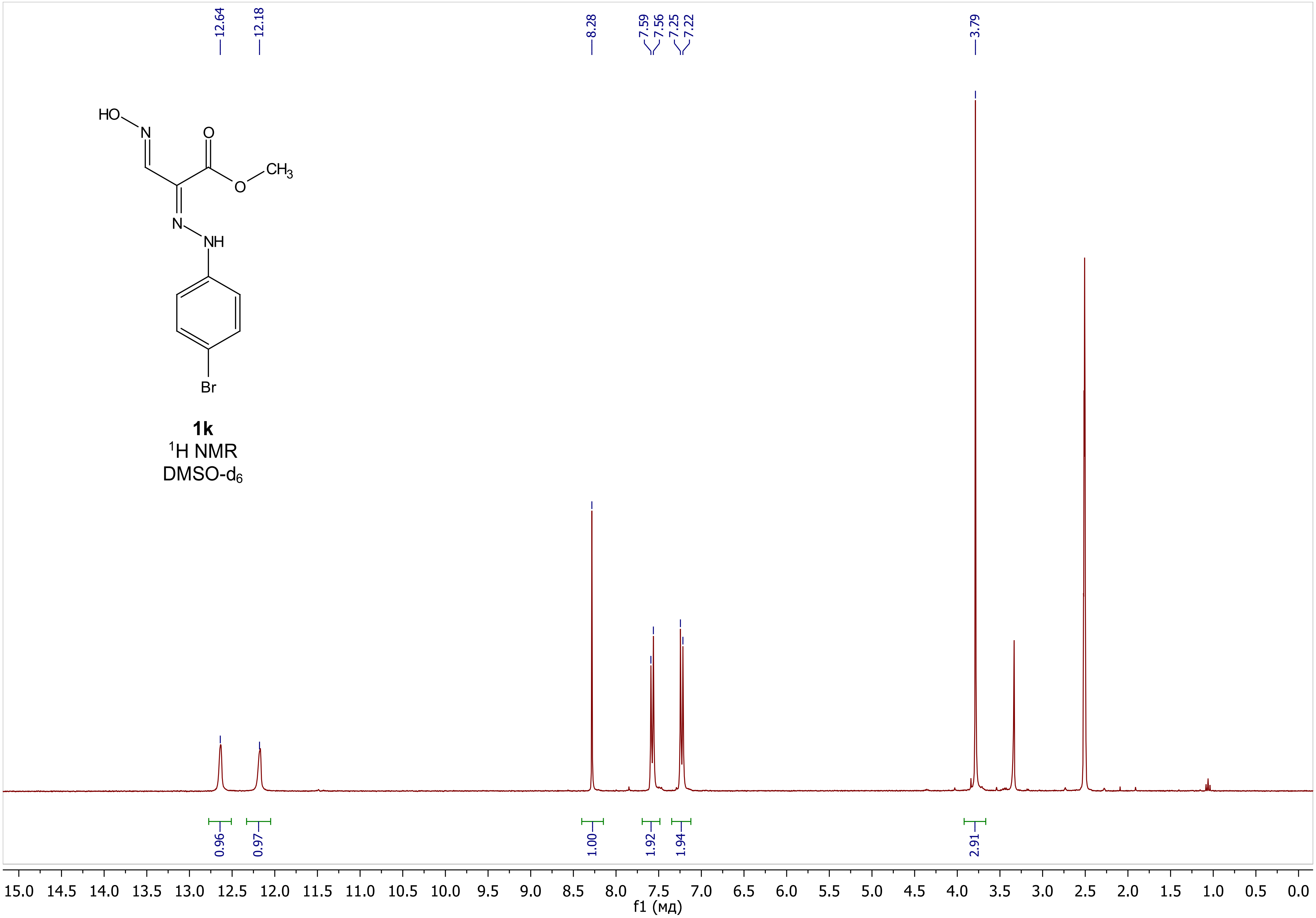


1j

^{1}H NMR

DMSO-d$_6$

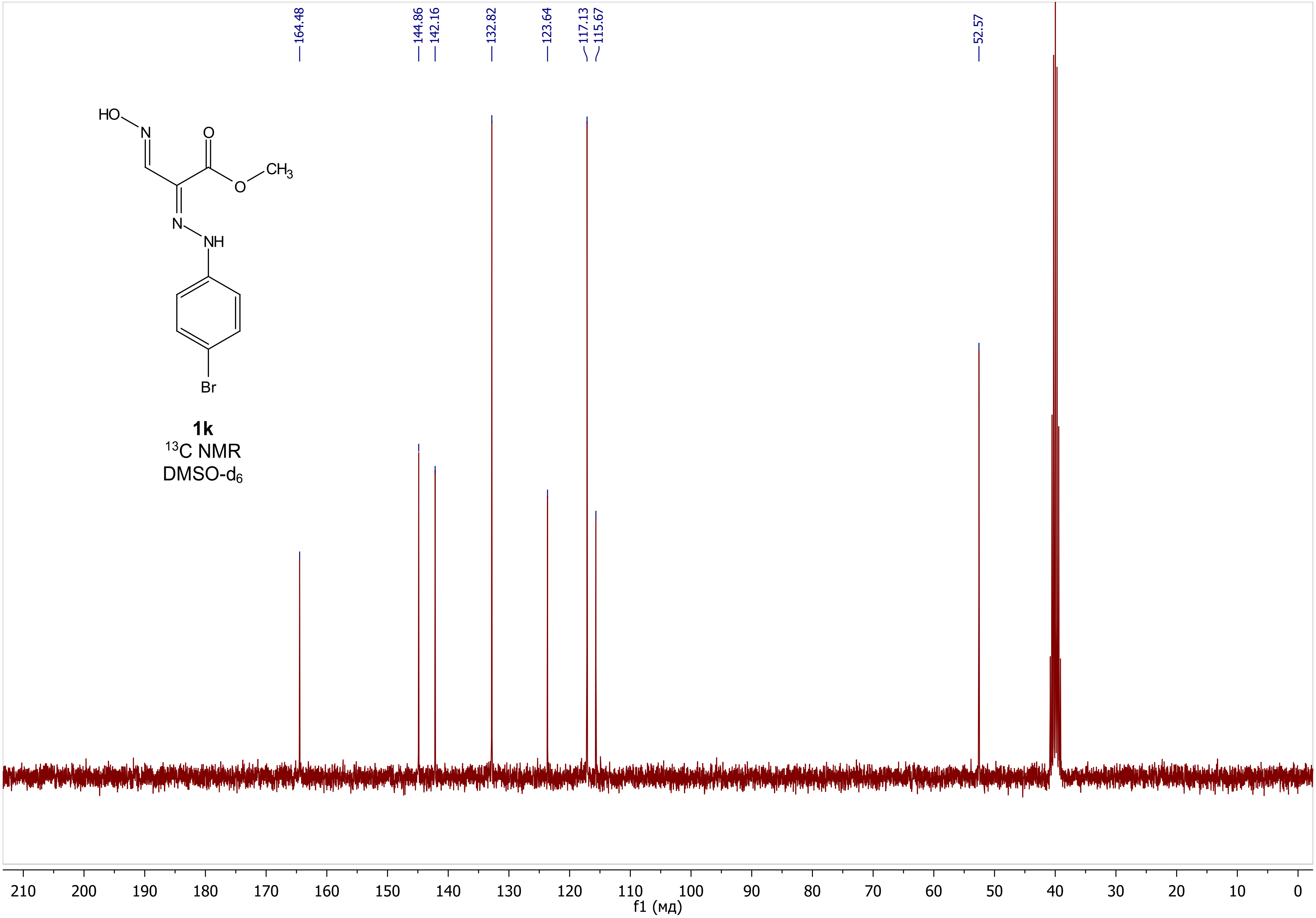

164.59
160.70
157.52
144.90
139.37
139.34
122.89
116.93
116.81
116.70
116.63
52.47
HO
N
O
CH3
O
N
NH
F
1j
13C NMR
DMSO-d6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)

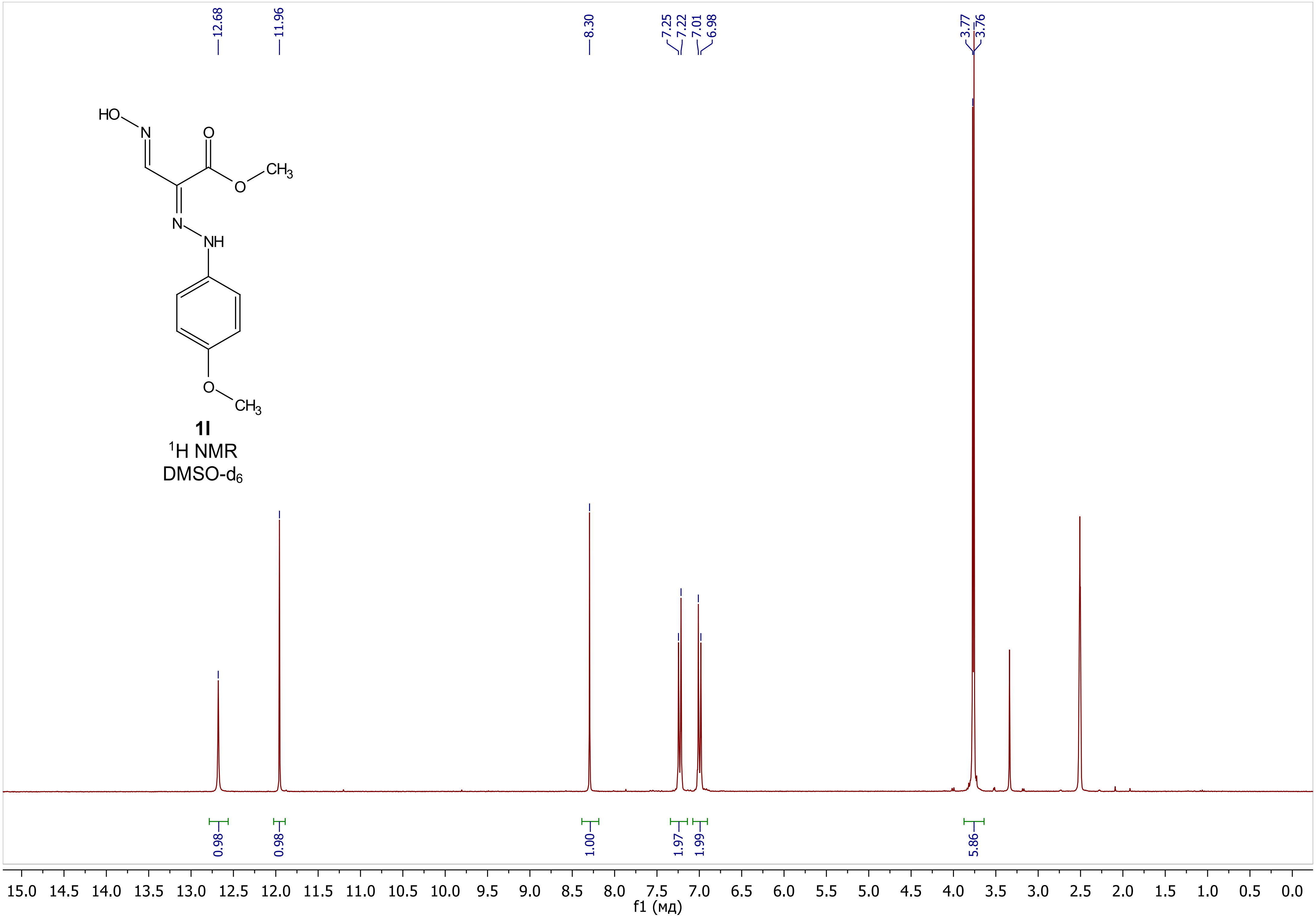


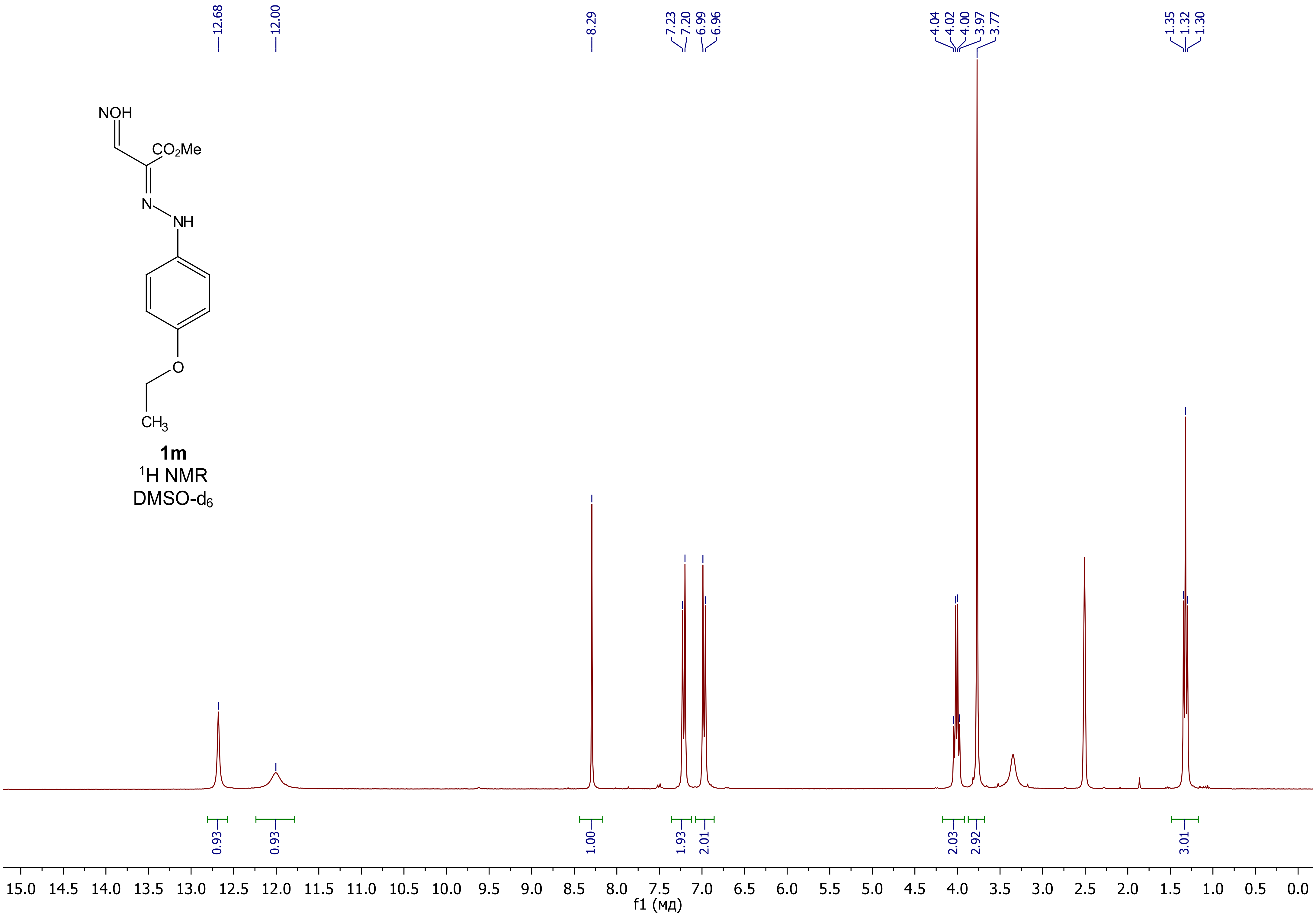
1k

^{1}H NMR

DMSO-d_6


164.48
144.86
142.16
132.82
123.64
117.13
115.67
52.57
HO
N
O
O
CH3
N
NH
Br
1k
13C NMR
DMSO-d6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)

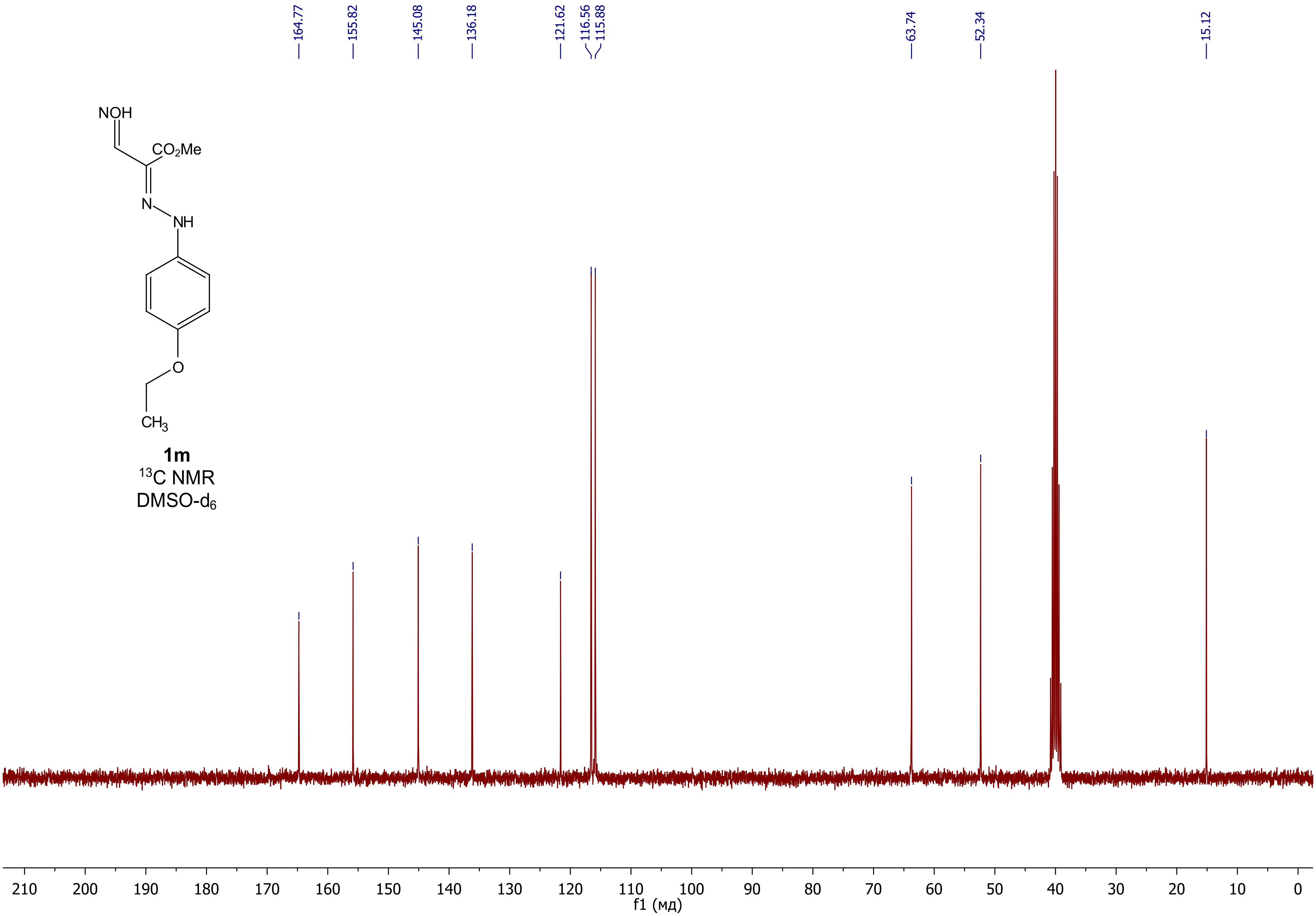

12.68
11.96
8.30
7.25
7.22
7.01
6.98
3.77
3.76
HO
N
O
CH3
O
N
NH
O
CH3
1l
¹H NMR
DMSO-d6
0.98
0.98
1.00
1.97
1.99
5.86
15.0 14.5 14.0 13.5 13.0 12.5 12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (мд)

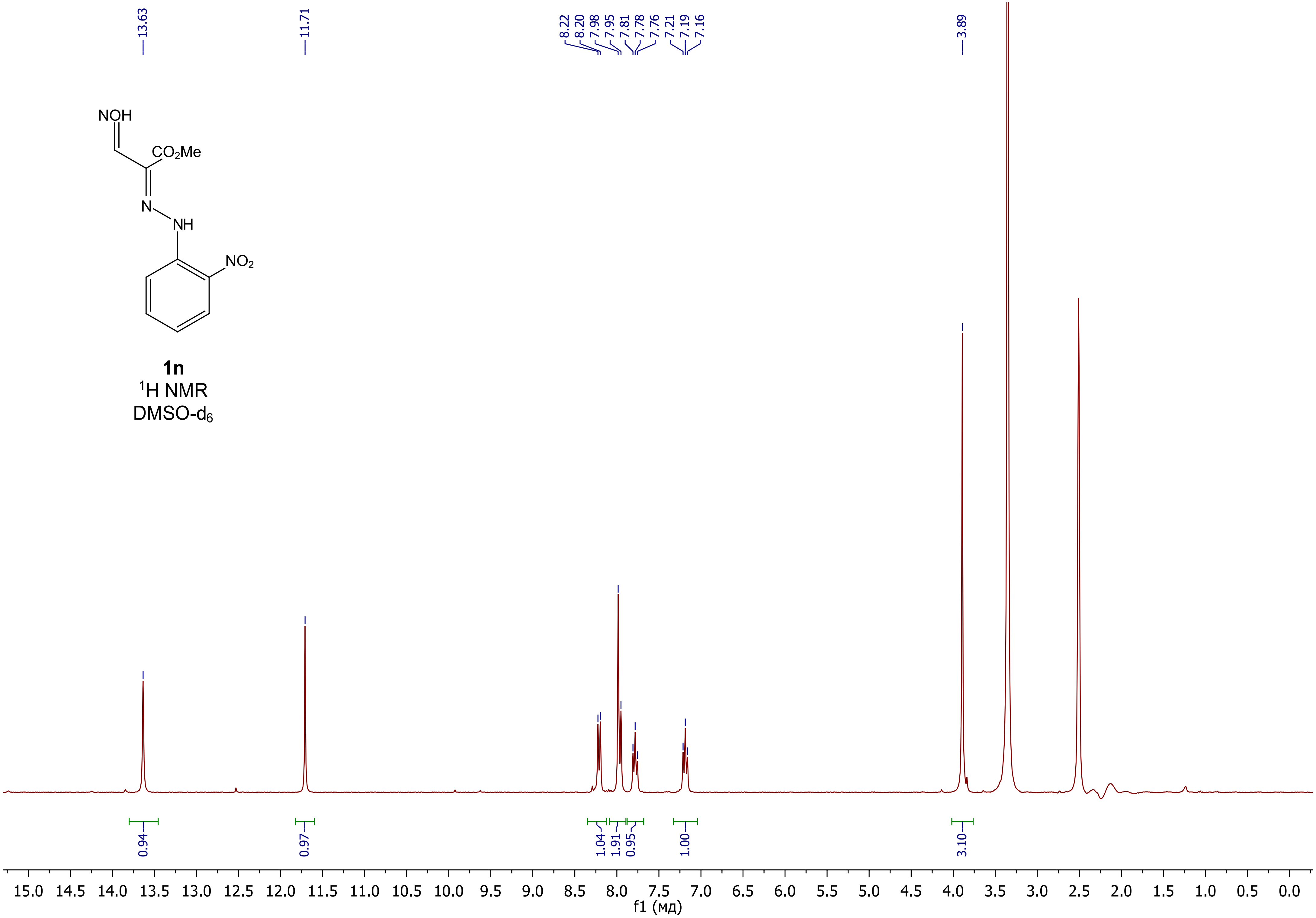


1m

^{1}H NMR

DMSO-d_6

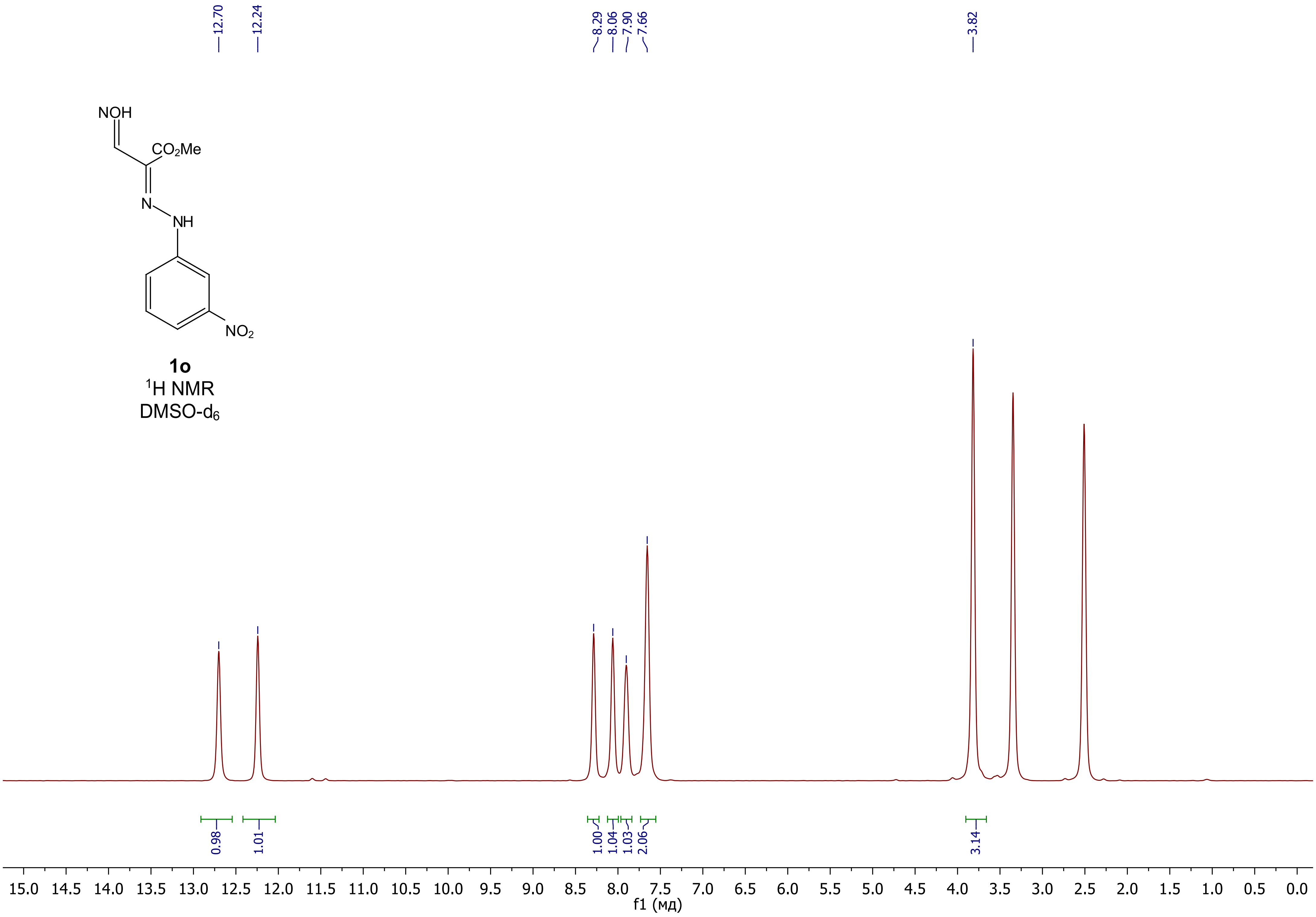

164.77
155.82
145.08
136.18
121.62
116.56
115.88
63.74
52.34
15.12
NOH
CO2Me
N
NH
O
CH3
1m
13C NMR
DMSO-d6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)



1n

^{1}H NMR

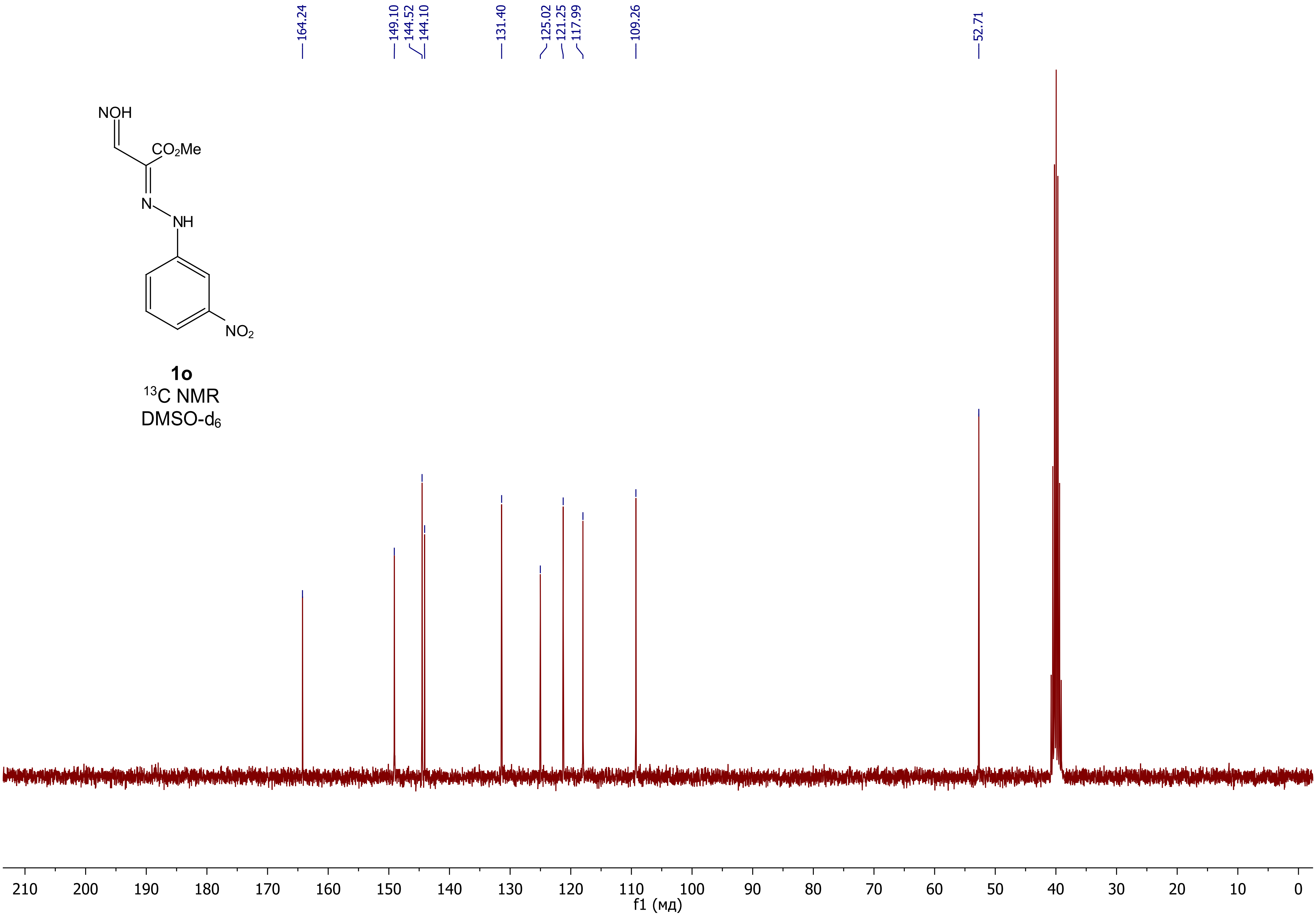
DMSO-d_6



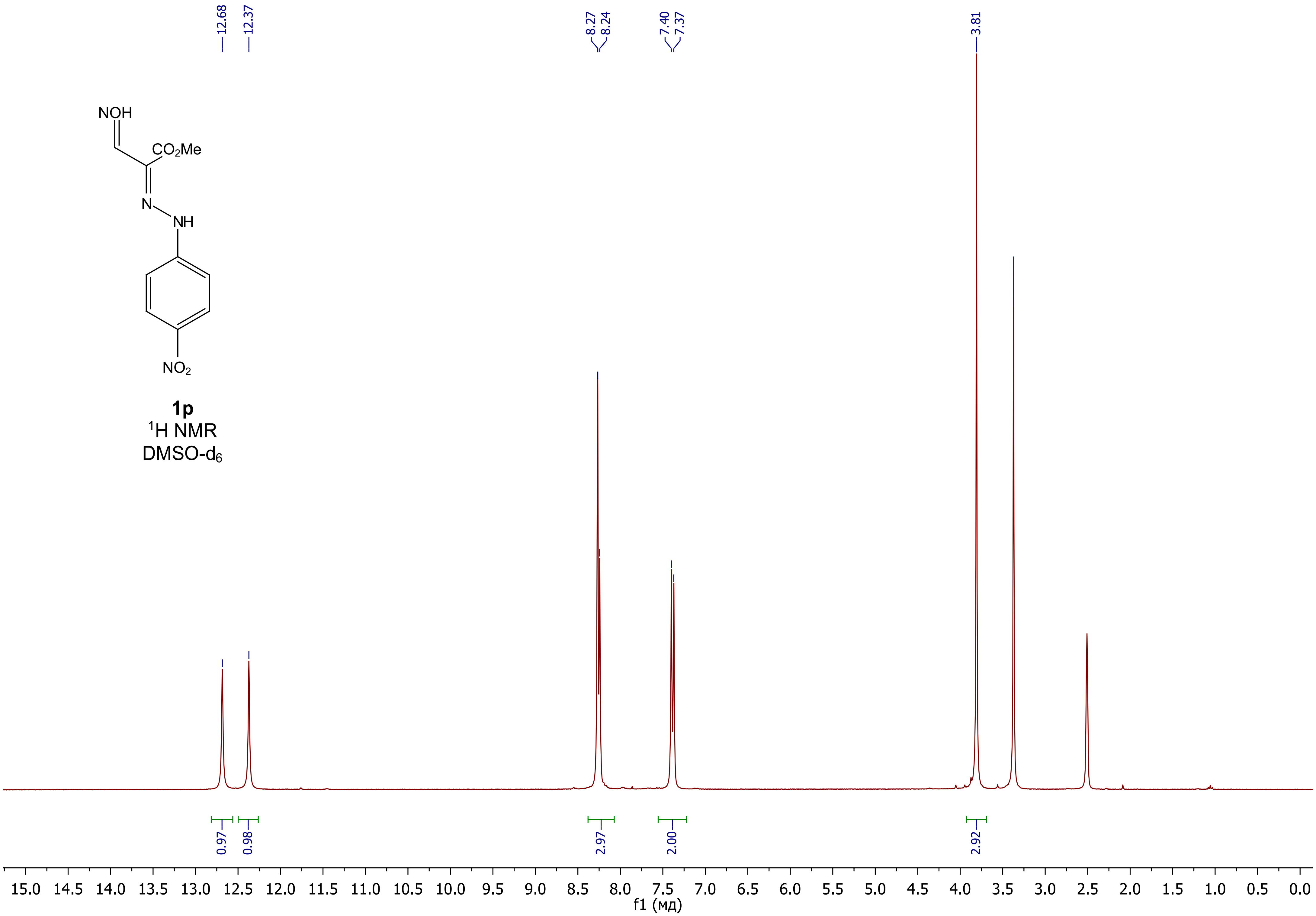
1o

^{1}H NMR

DMSO-d_6



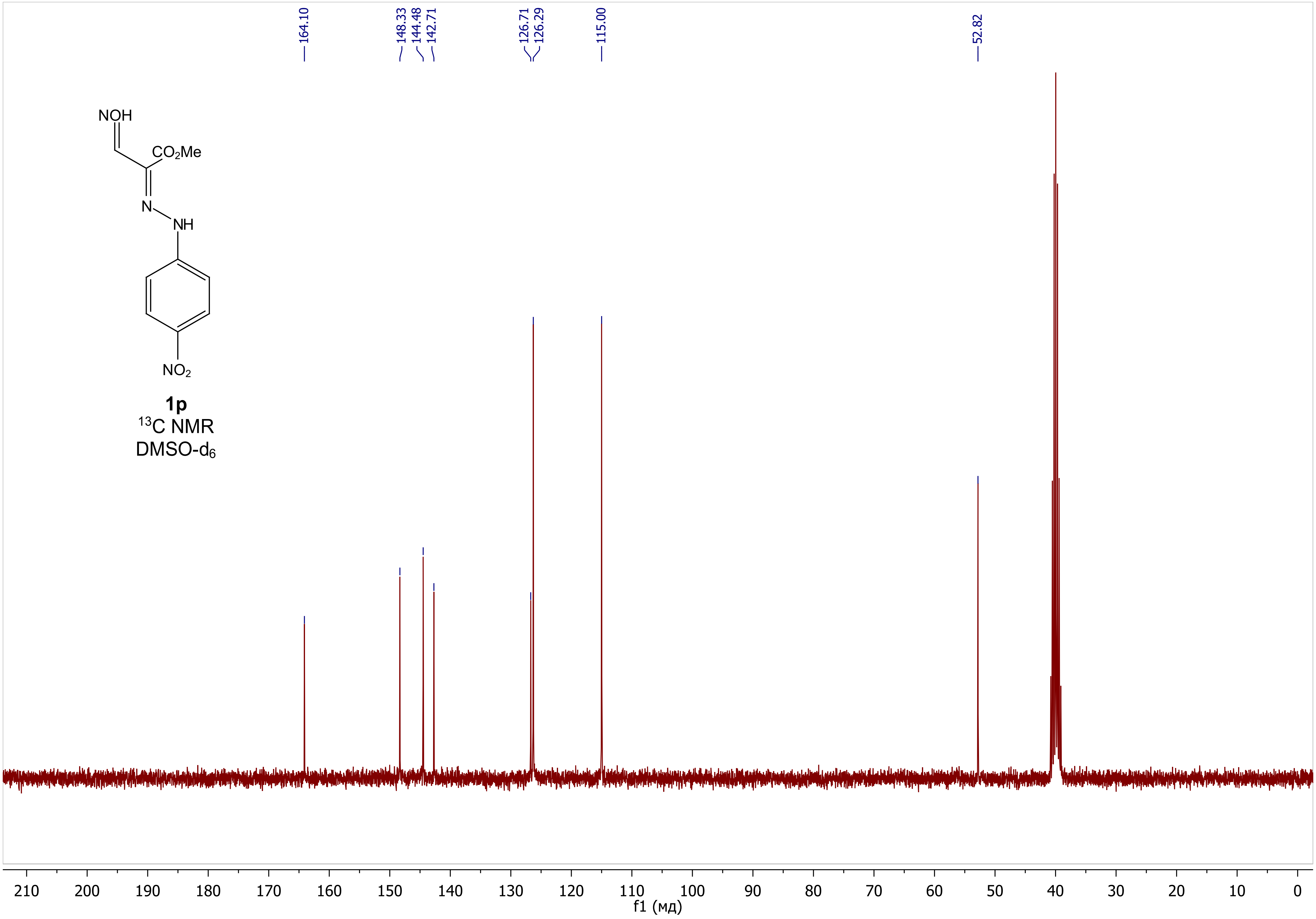
1o
^{13}C NMR
DMSO-d_6



1p

^{1}H NMR

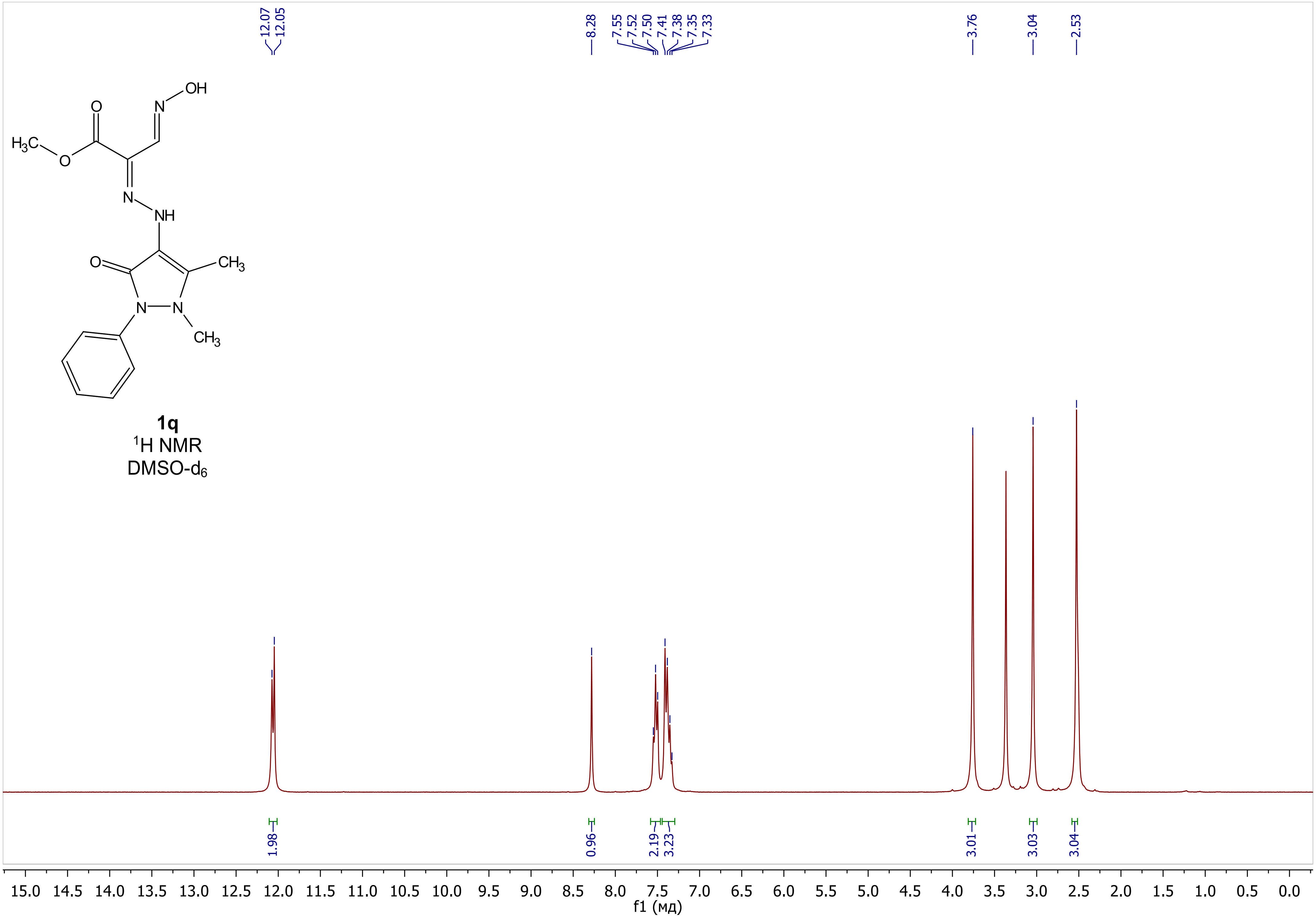
DMSO-d_6



1p

^{13}C NMR

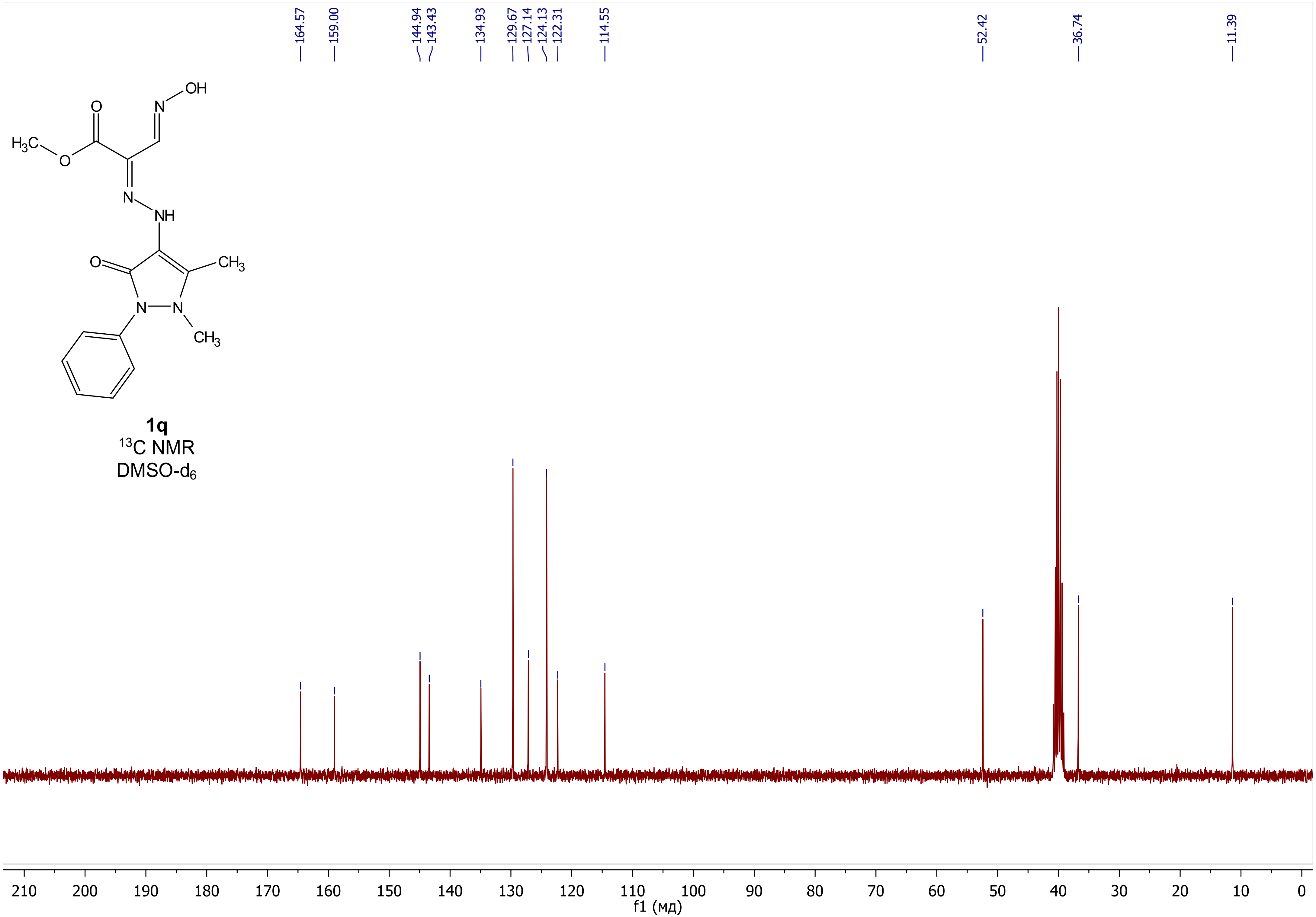
DMSO-d_6



1q

^{1}H NMR

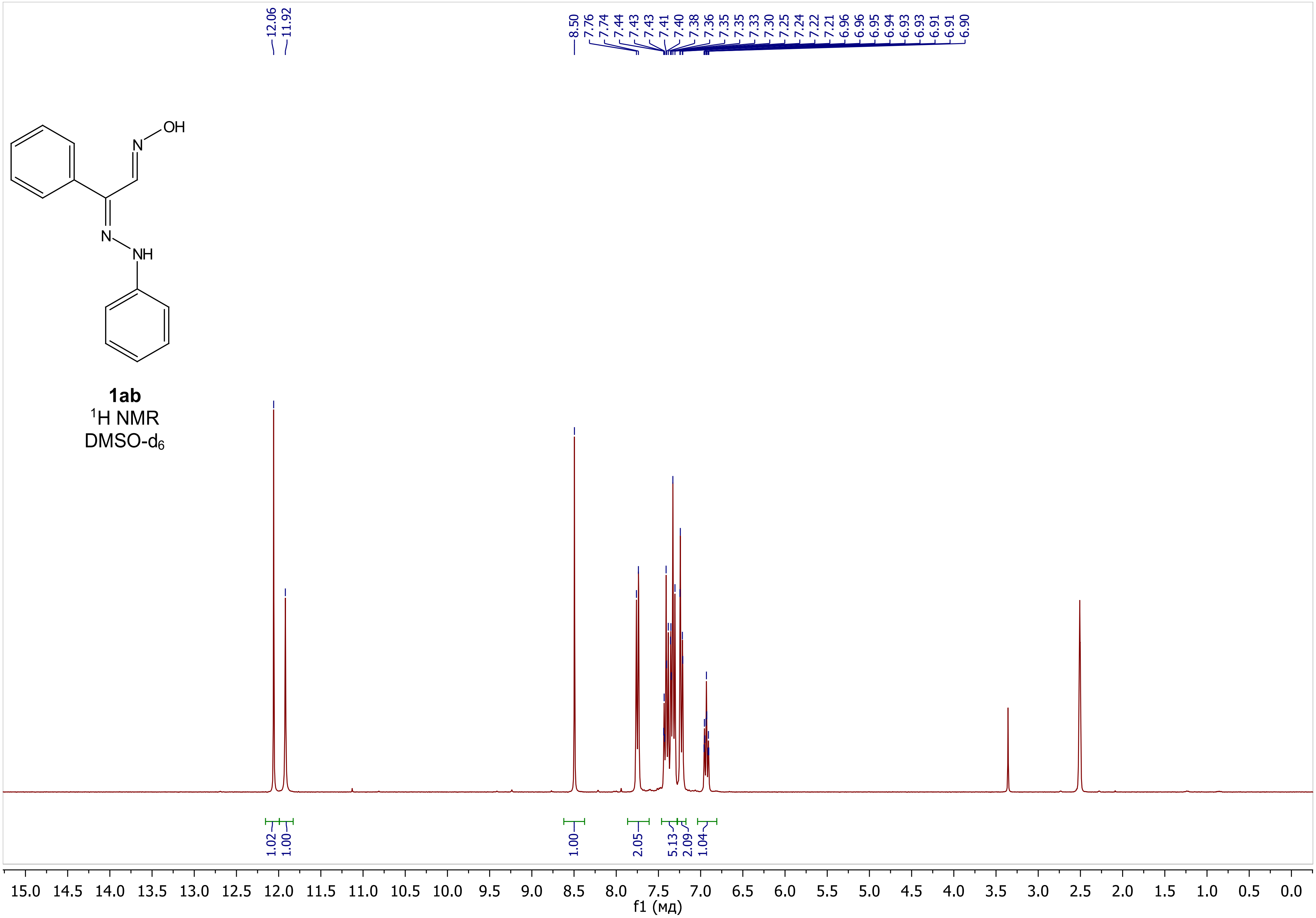
DMSO-d_6



1q

^{13}C NMR

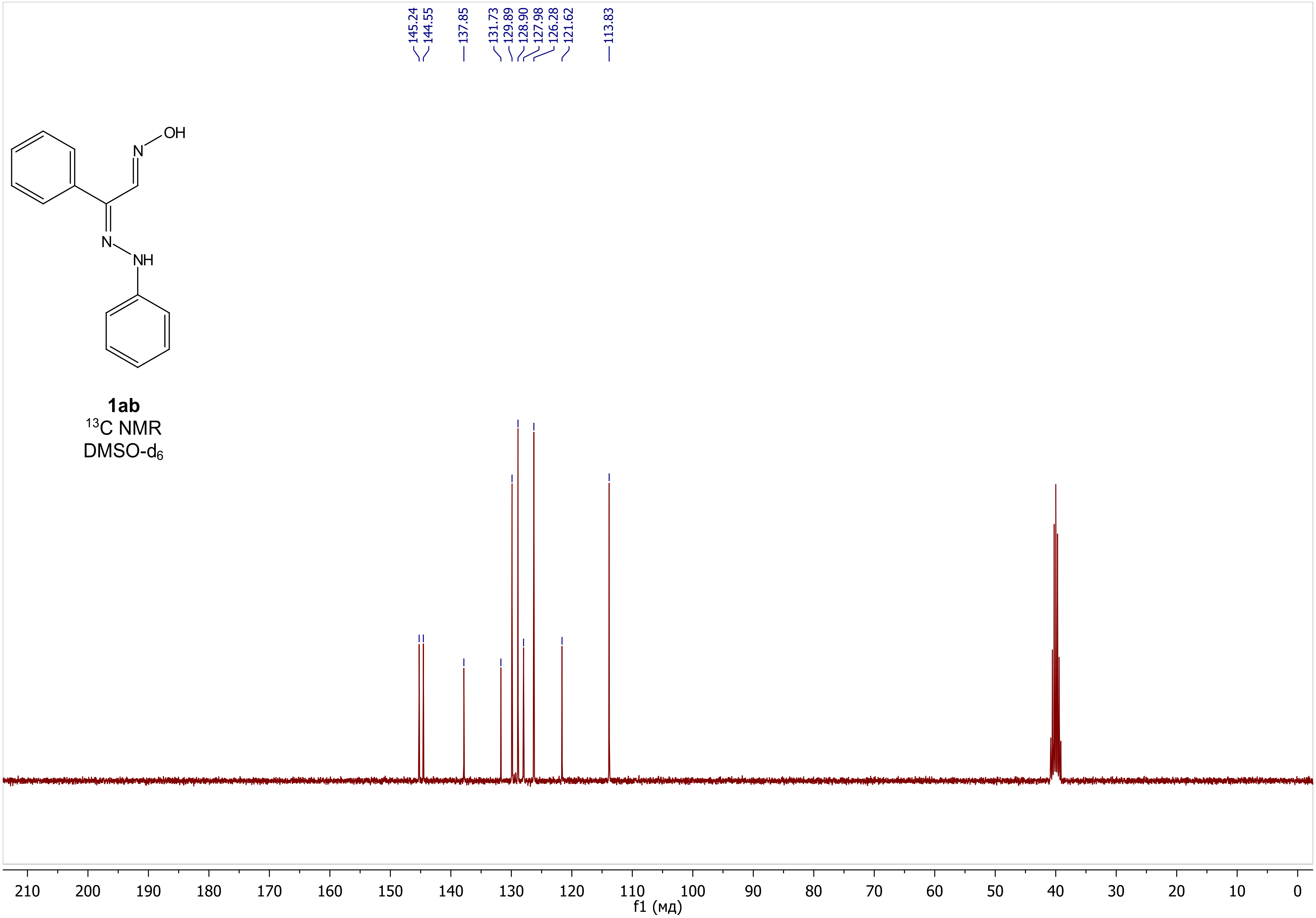
DMSO-d_6



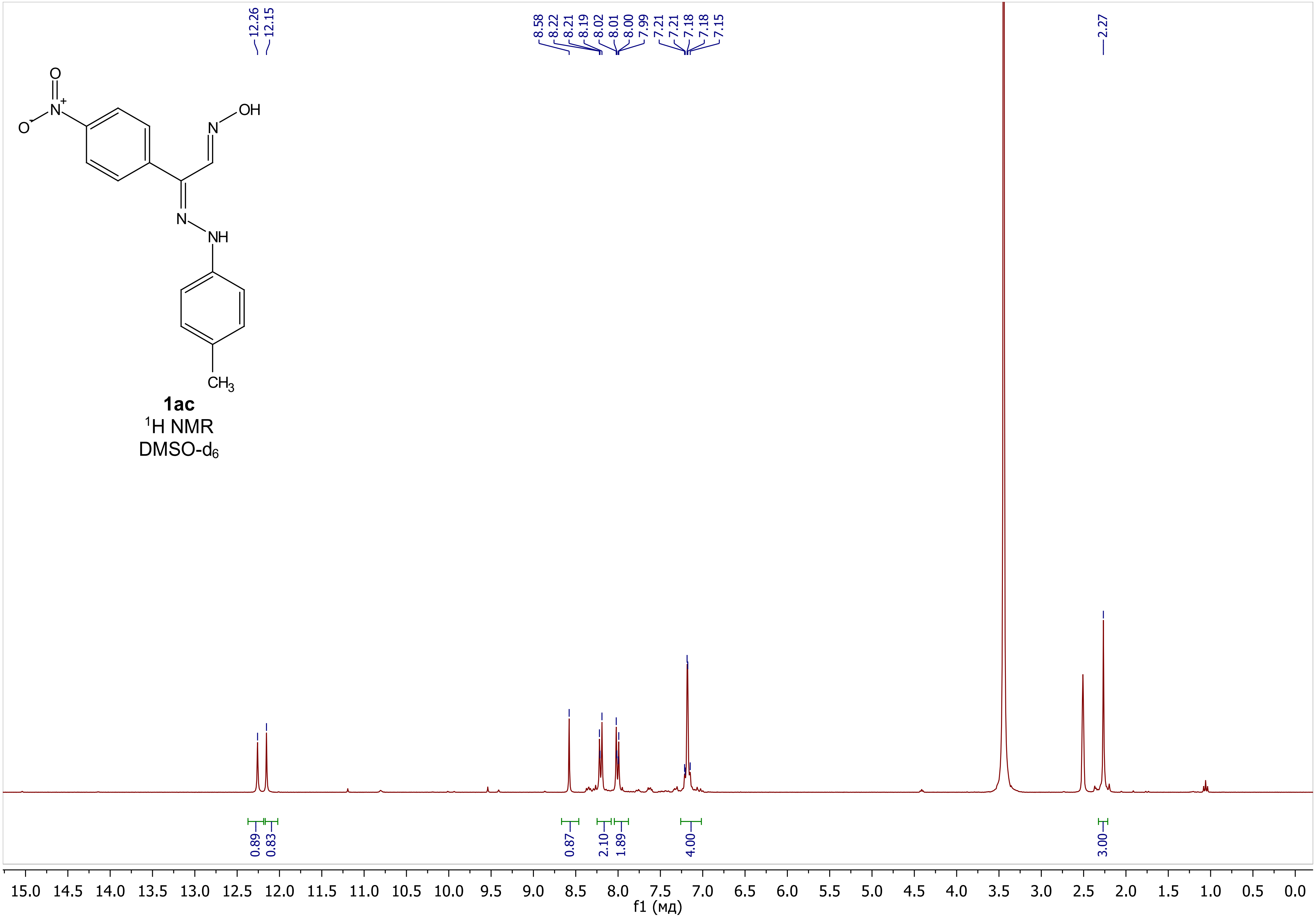
1ab

^{1}H NMR

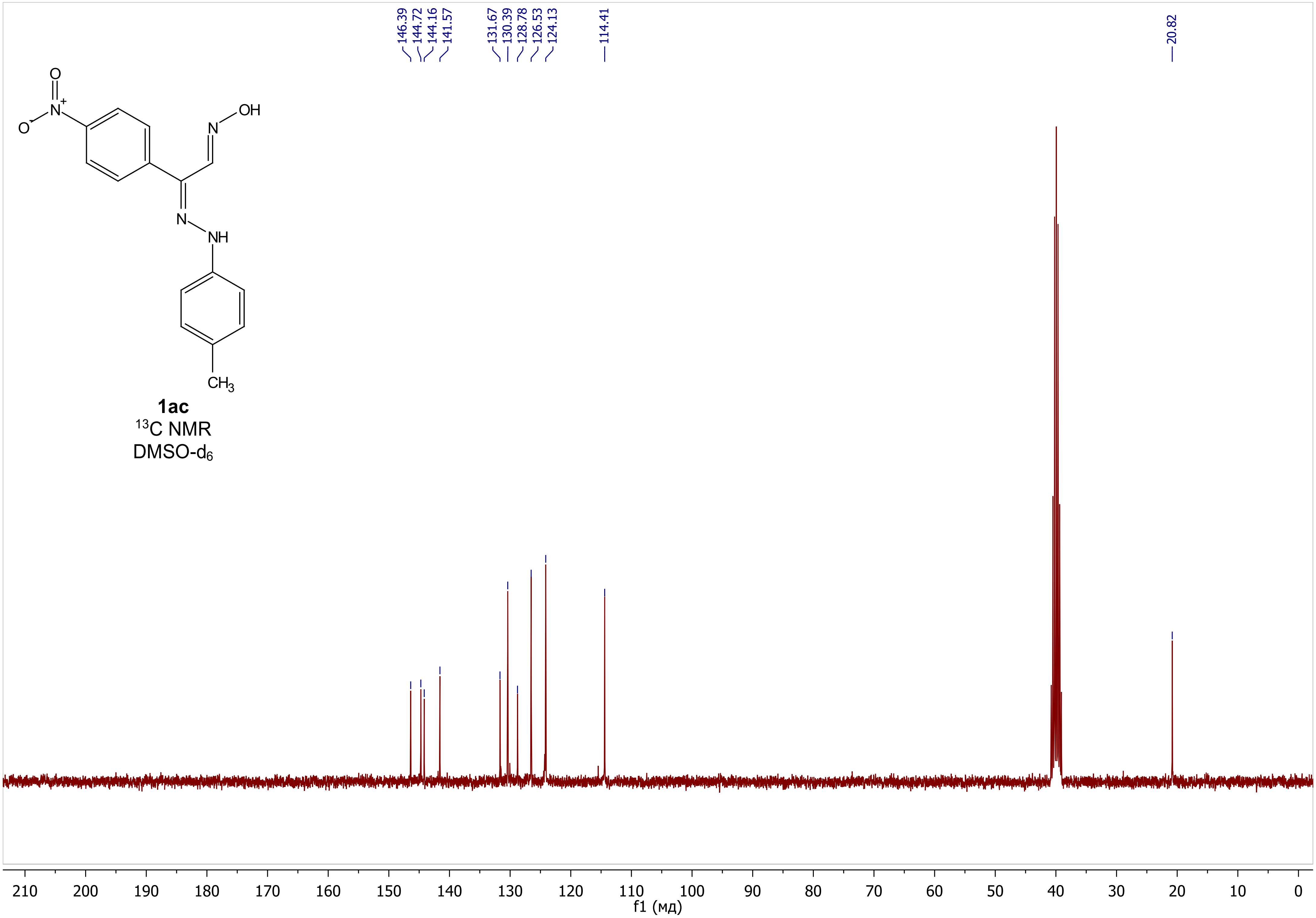
DMSO-d_6

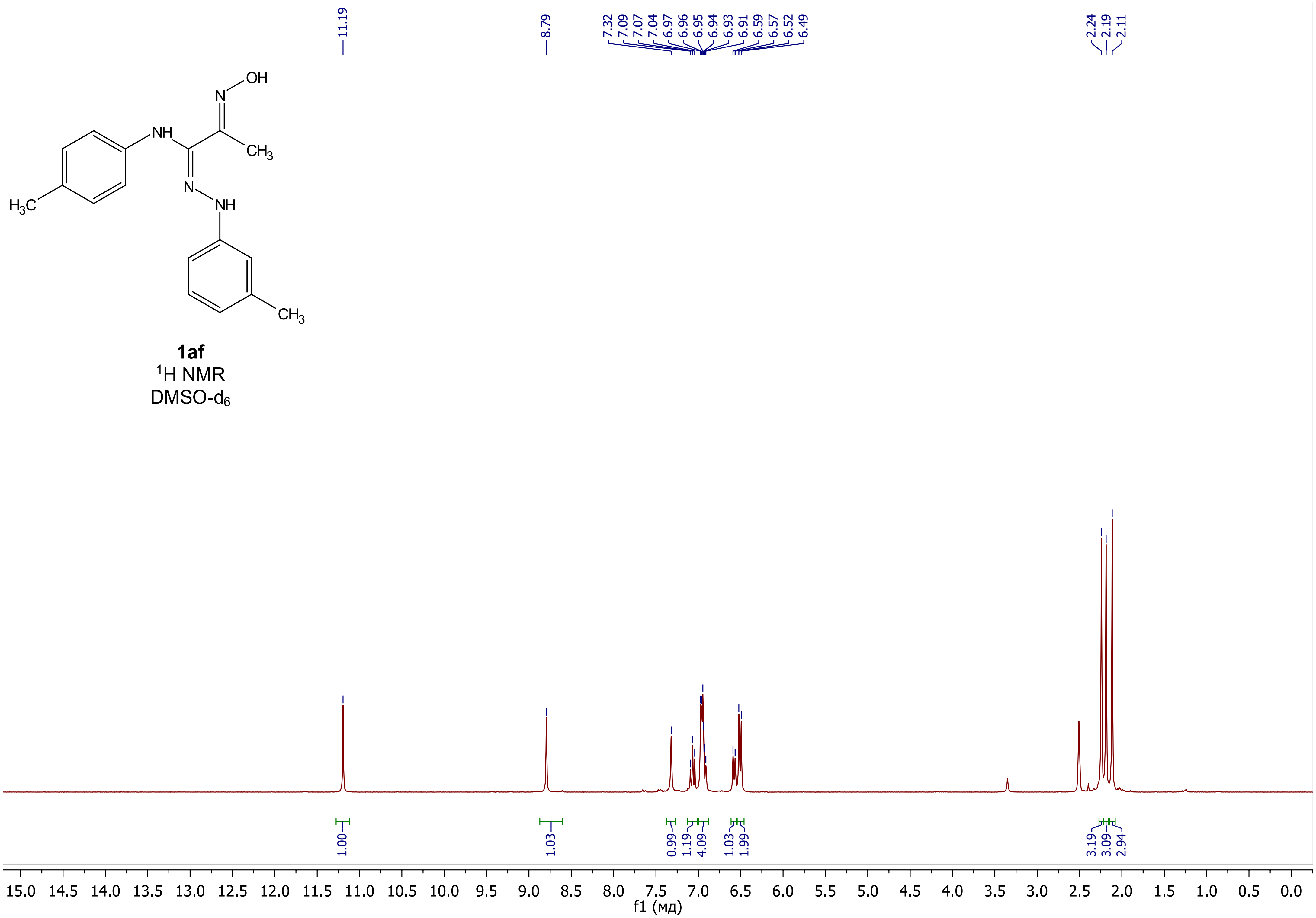


1ab
^{13}C NMR
DMSO-d_6



1ac
^{1}H NMR
DMSO-d_6

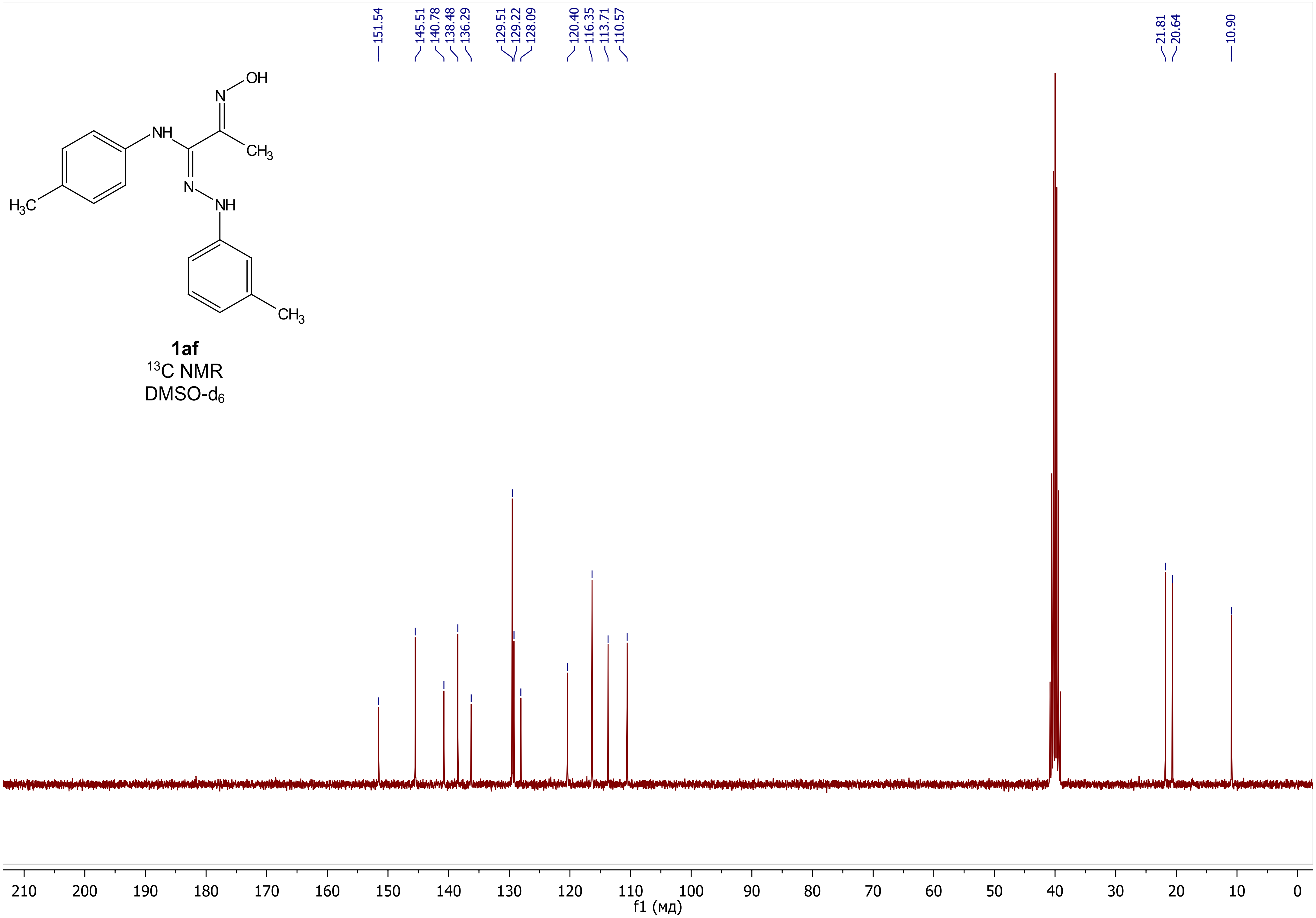

146.39
144.72
144.16
141.57
131.67
130.39
128.78
126.53
124.13
114.41
20.82
O
N
+
O
-
OH
N
N
NH
CH3
1ac
^{13}C NMR
DMSO-d$_6$
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)



1af

^{1}H NMR

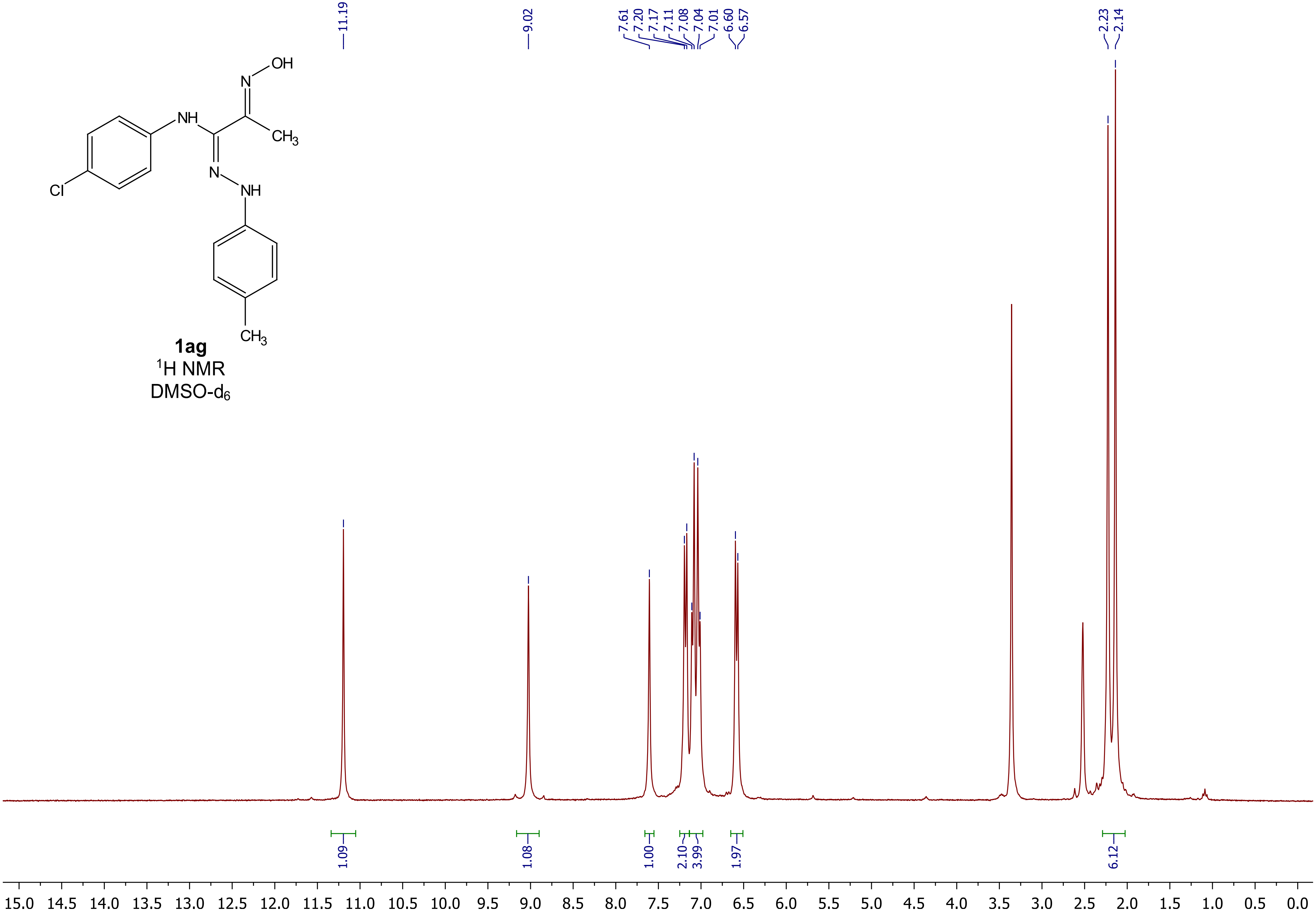
DMSO-d_6



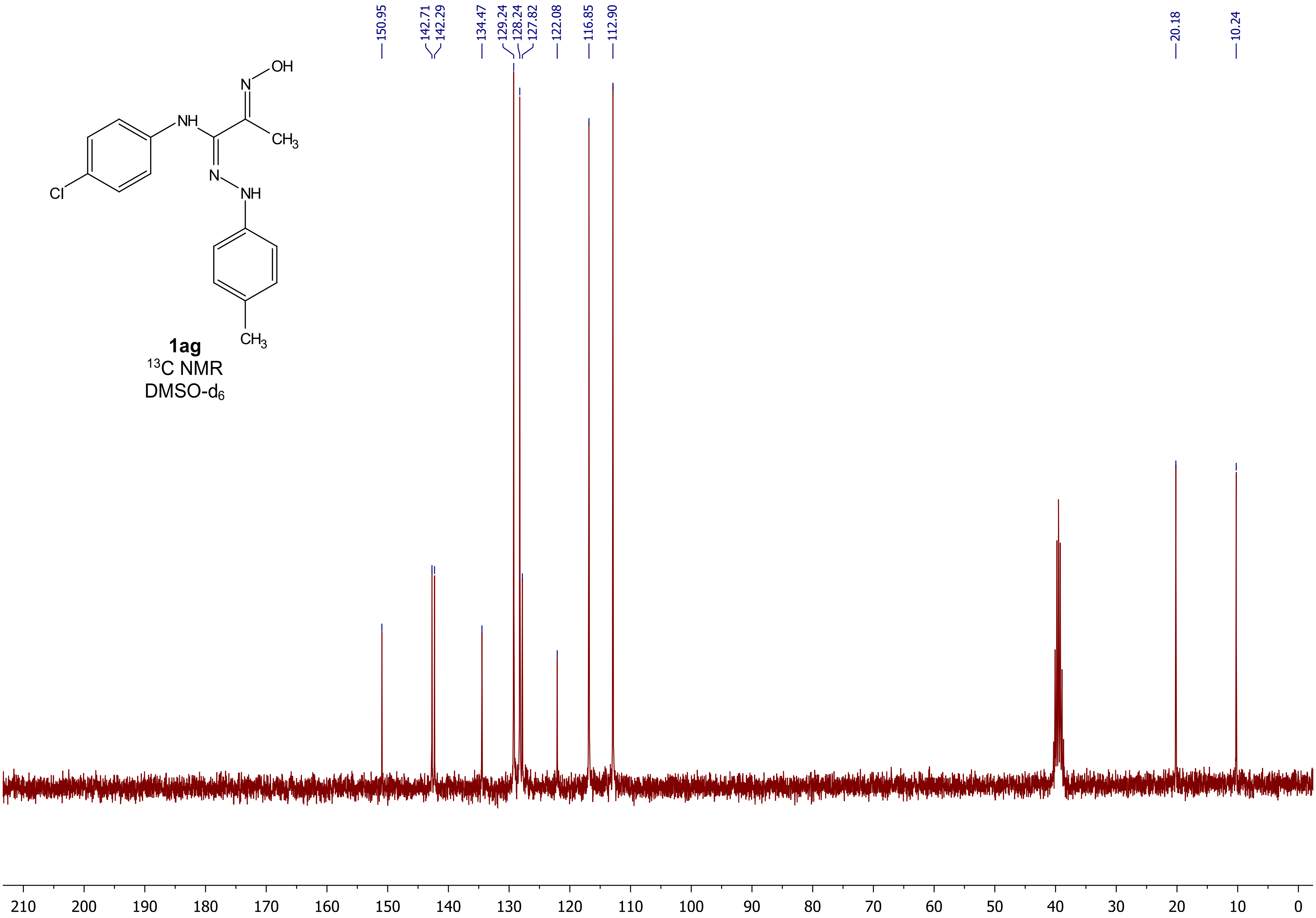
1af

^{13}C NMR

DMSO-d_6



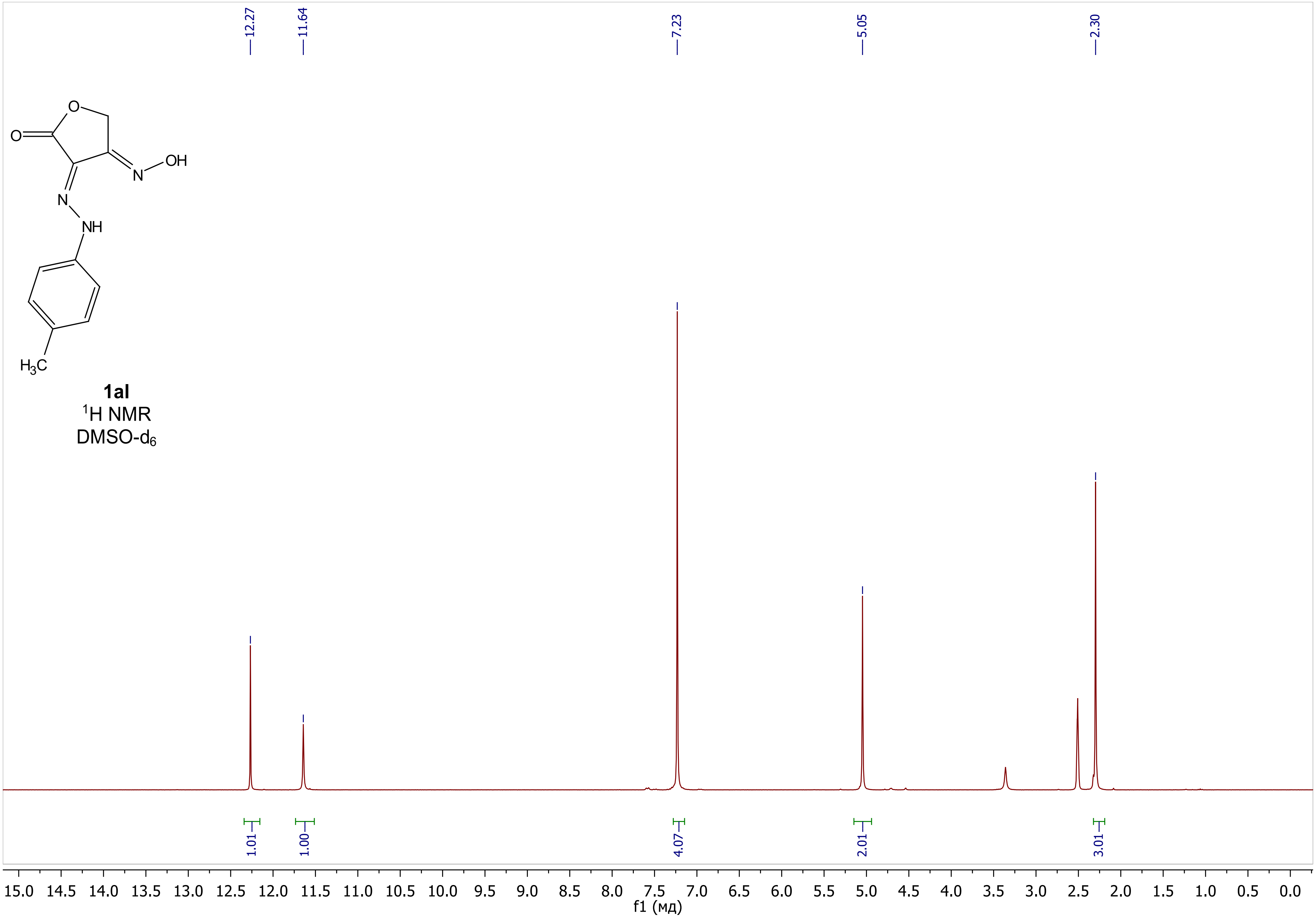
1ag
^{1}H NMR
DMSO-d_6



1ag

^{13}C NMR

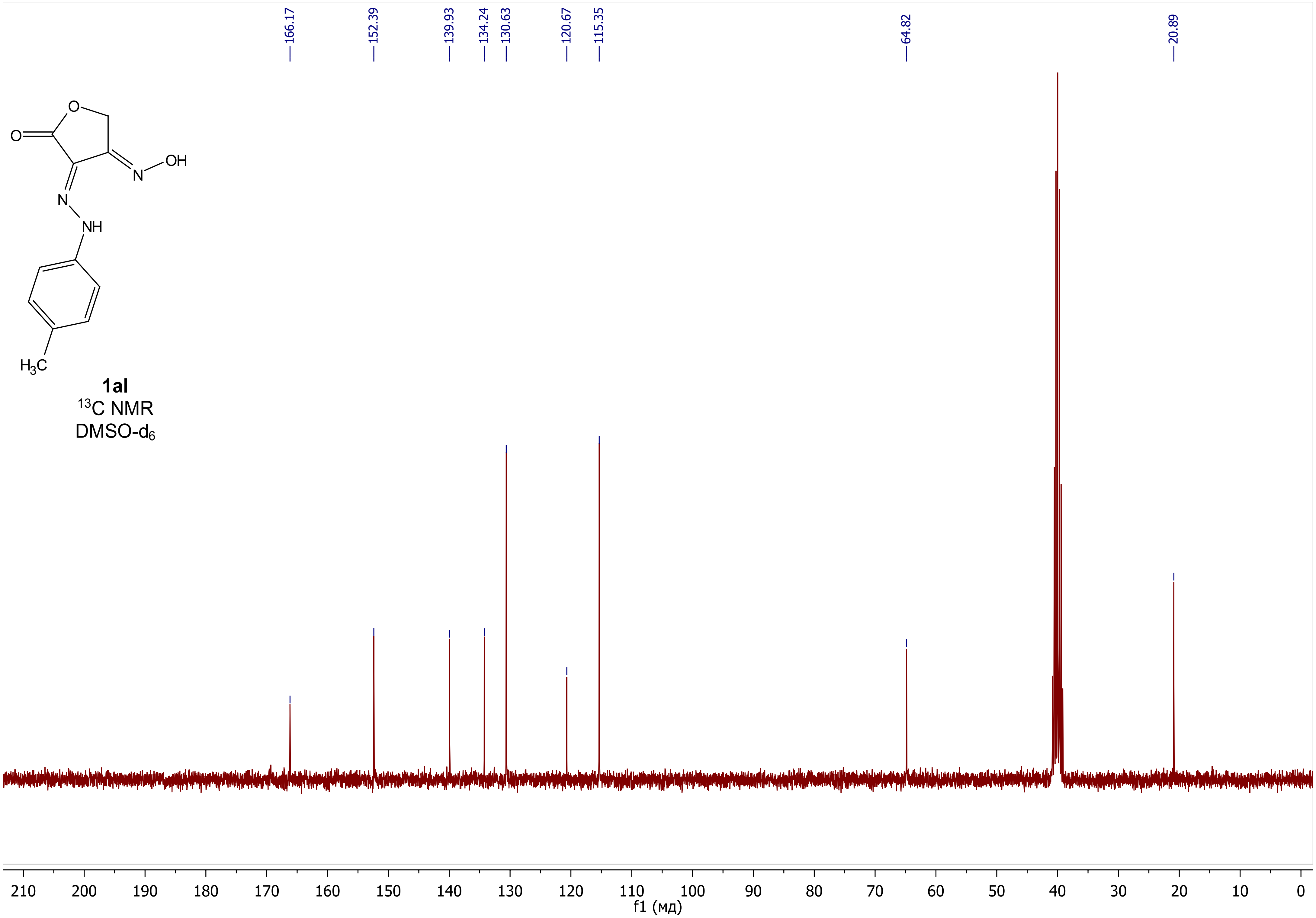
DMSO-d_6



1al

^{1}H NMR

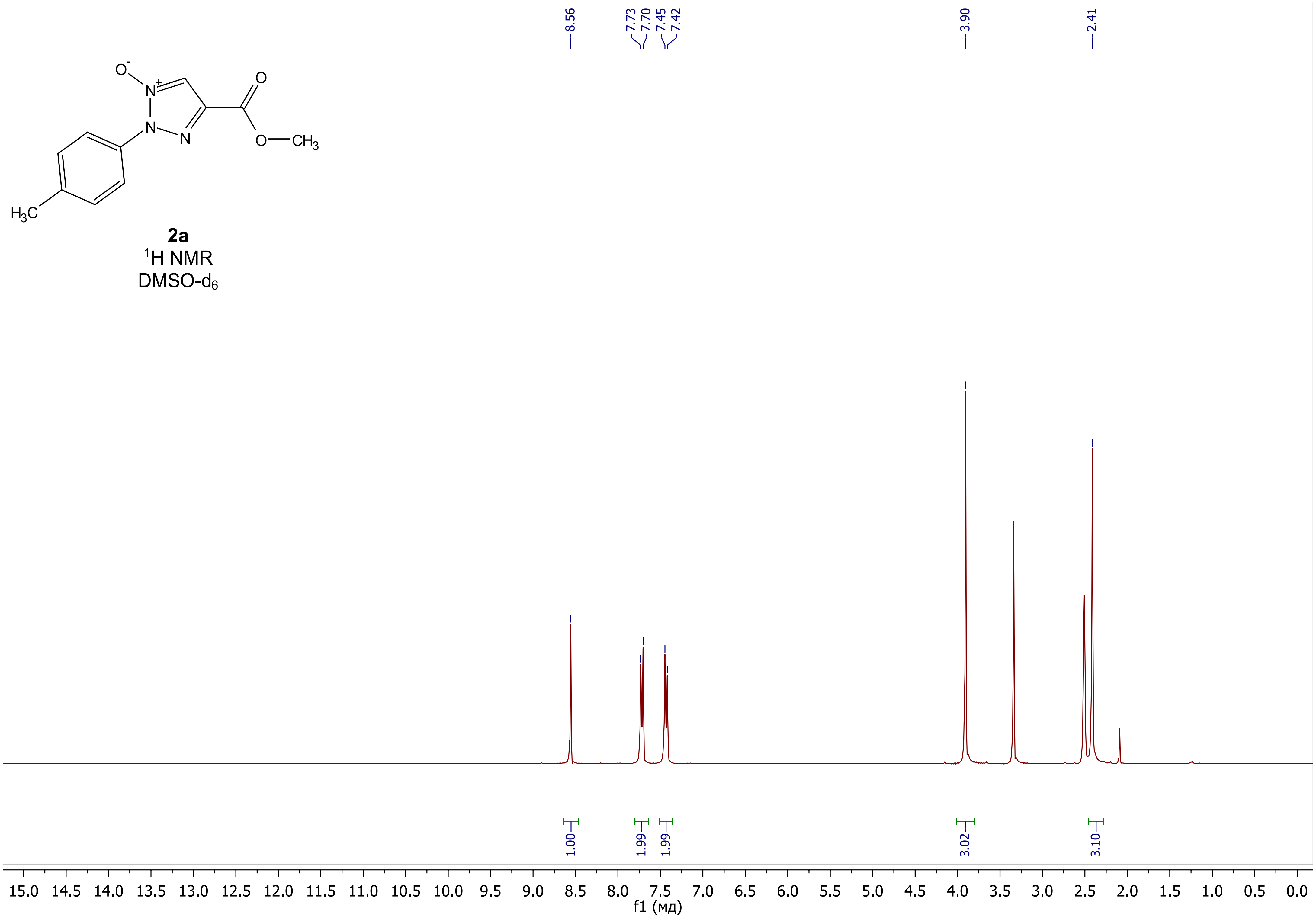
DMSO-d_6



1al

^{13}C NMR

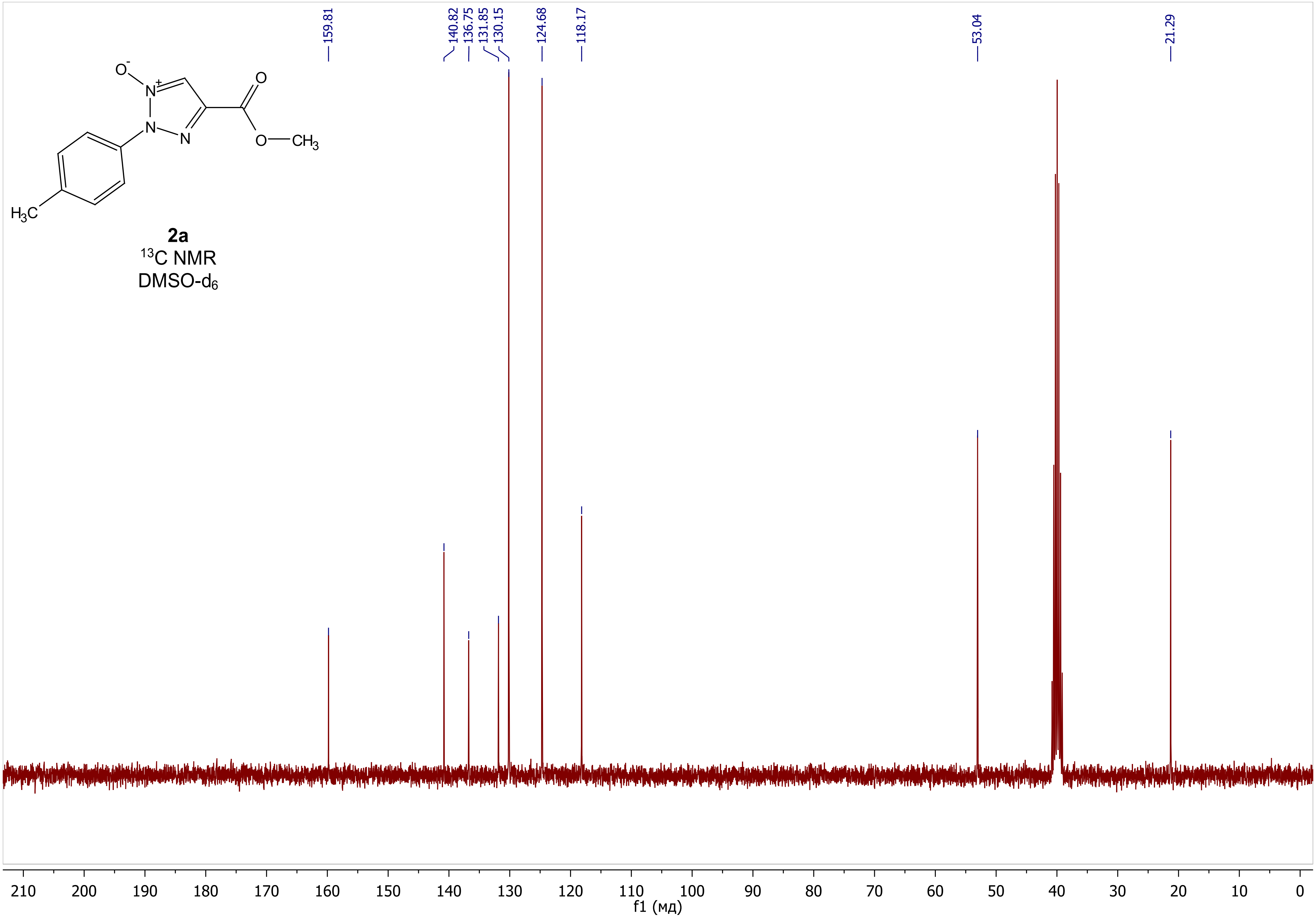
DMSO-d_6



2a

^{1}H NMR

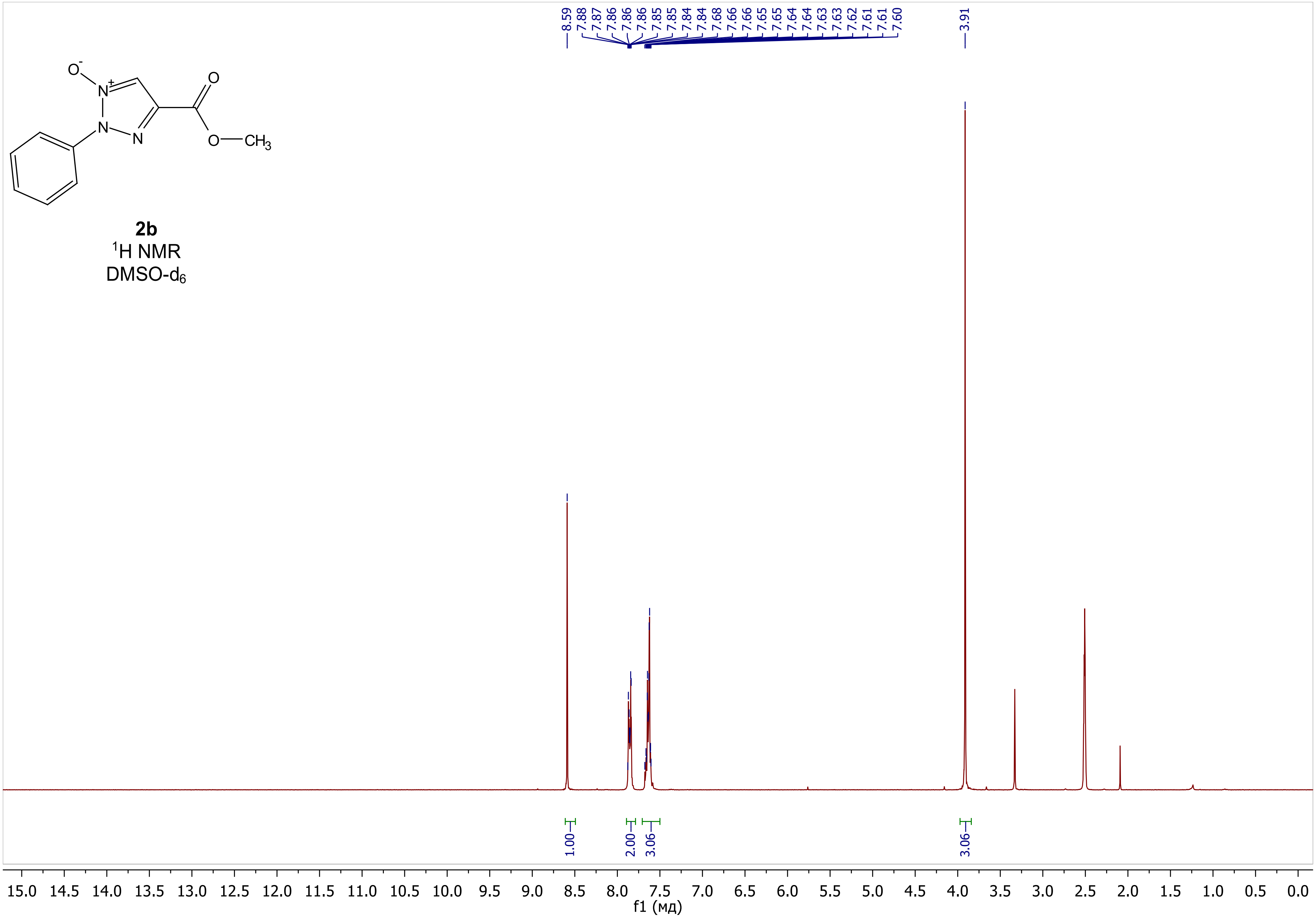
DMSO-d_6

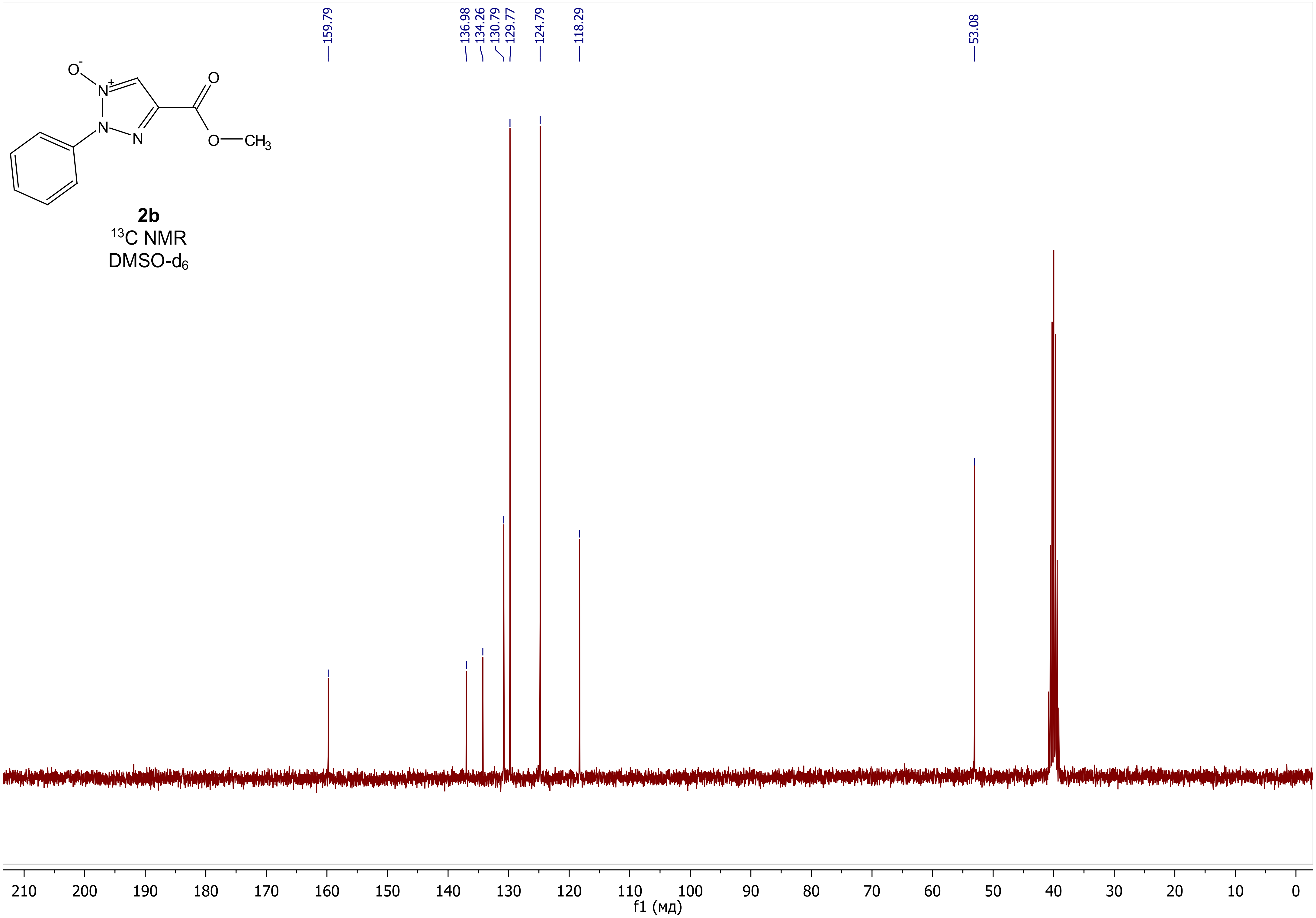


2a

^{13}C NMR

DMSO-d_6

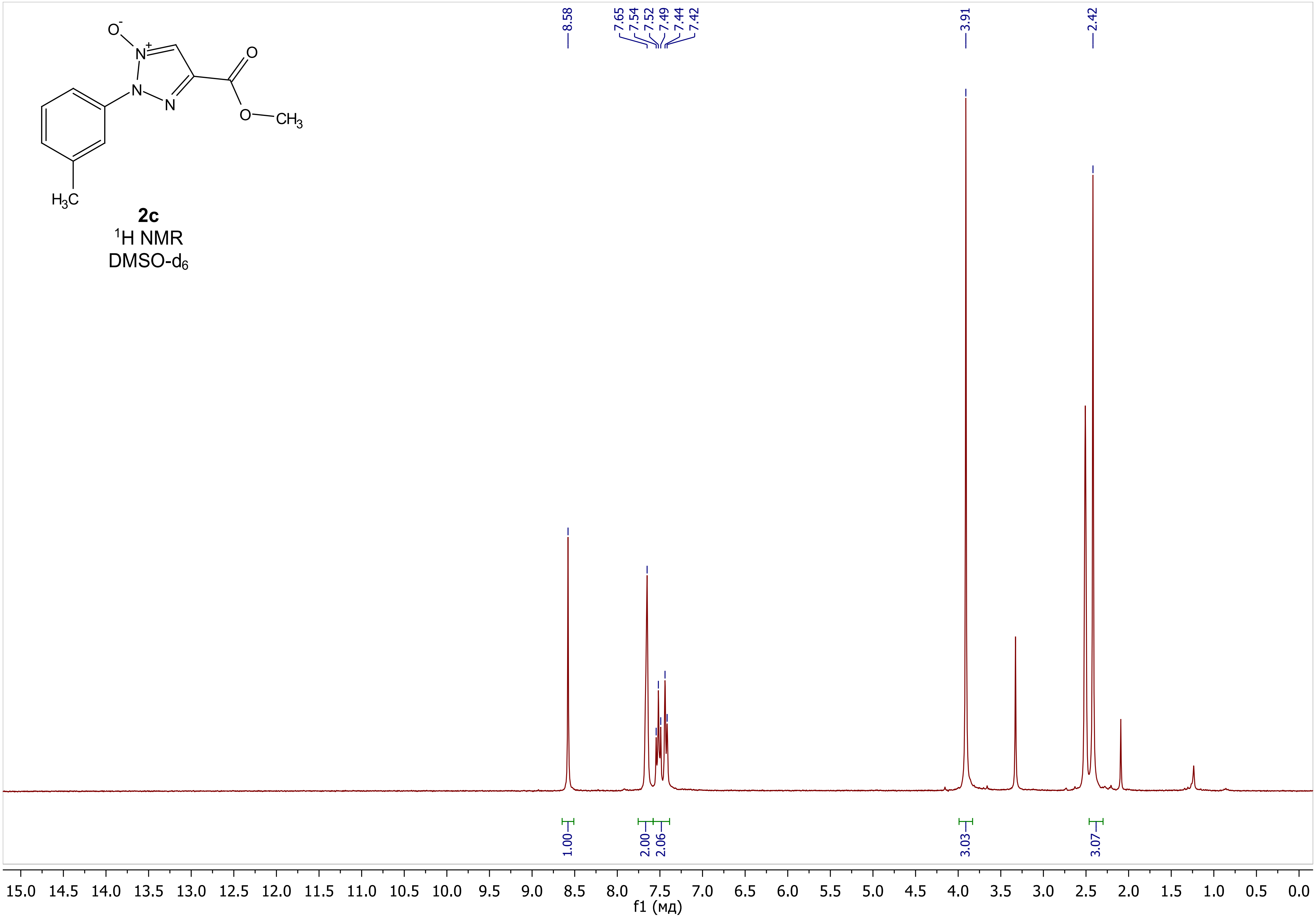

2b
1H NMR
DMSO-d6
O-
N+
N
N
O
O
CH3
8.59
7.88
7.87
7.86
7.86
7.86
7.85
7.85
7.84
7.84
7.68
7.66
7.66
7.65
7.65
7.64
7.64
7.63
7.63
7.62
7.61
7.61
7.60
3.91
1.00
2.00
3.06
3.06
15.0
14.5
14.0
13.5
13.0
12.5
12.0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (мд)



2b

^{13}C NMR

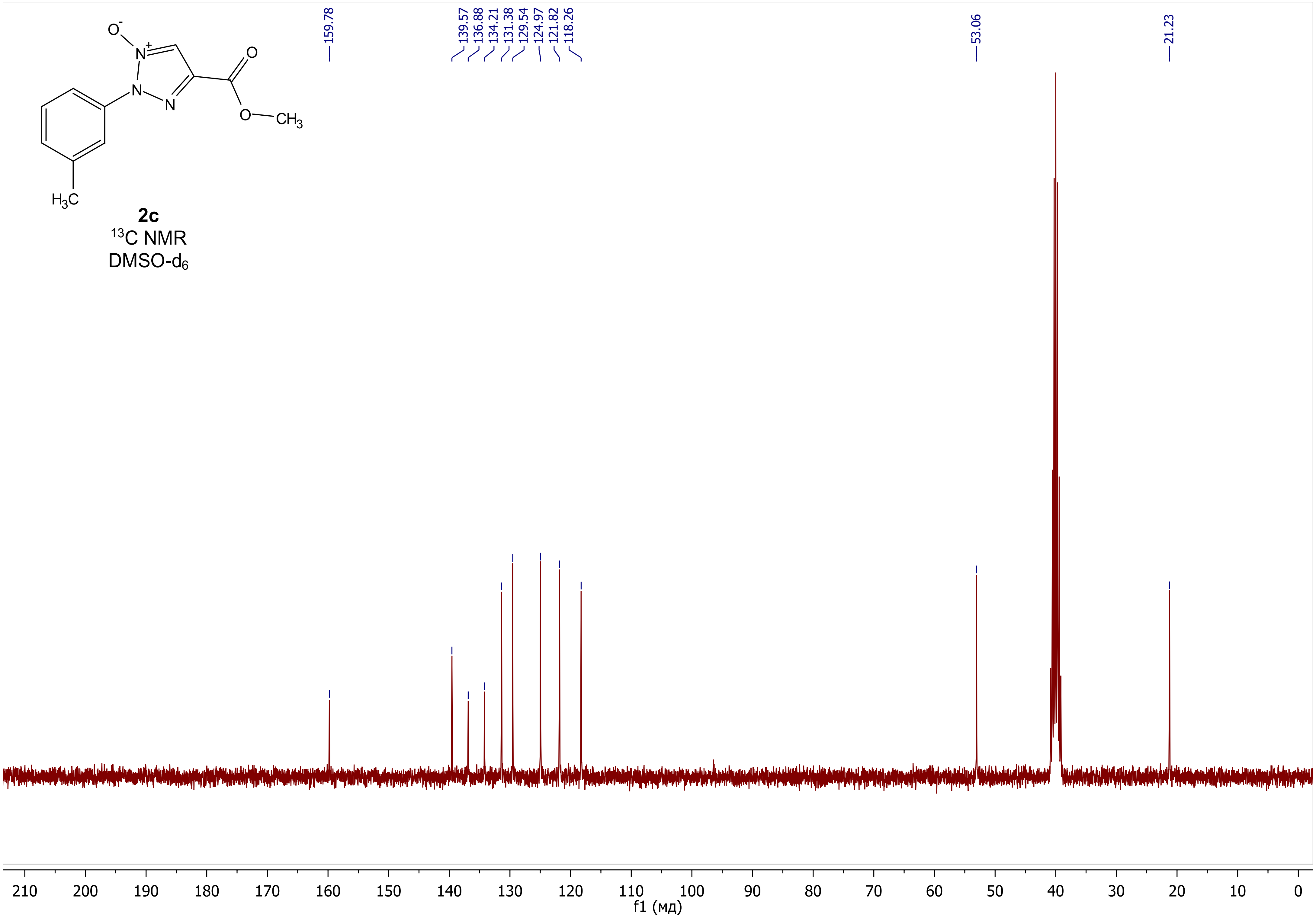
DMSO-d$_6$



2c

^{1}H NMR

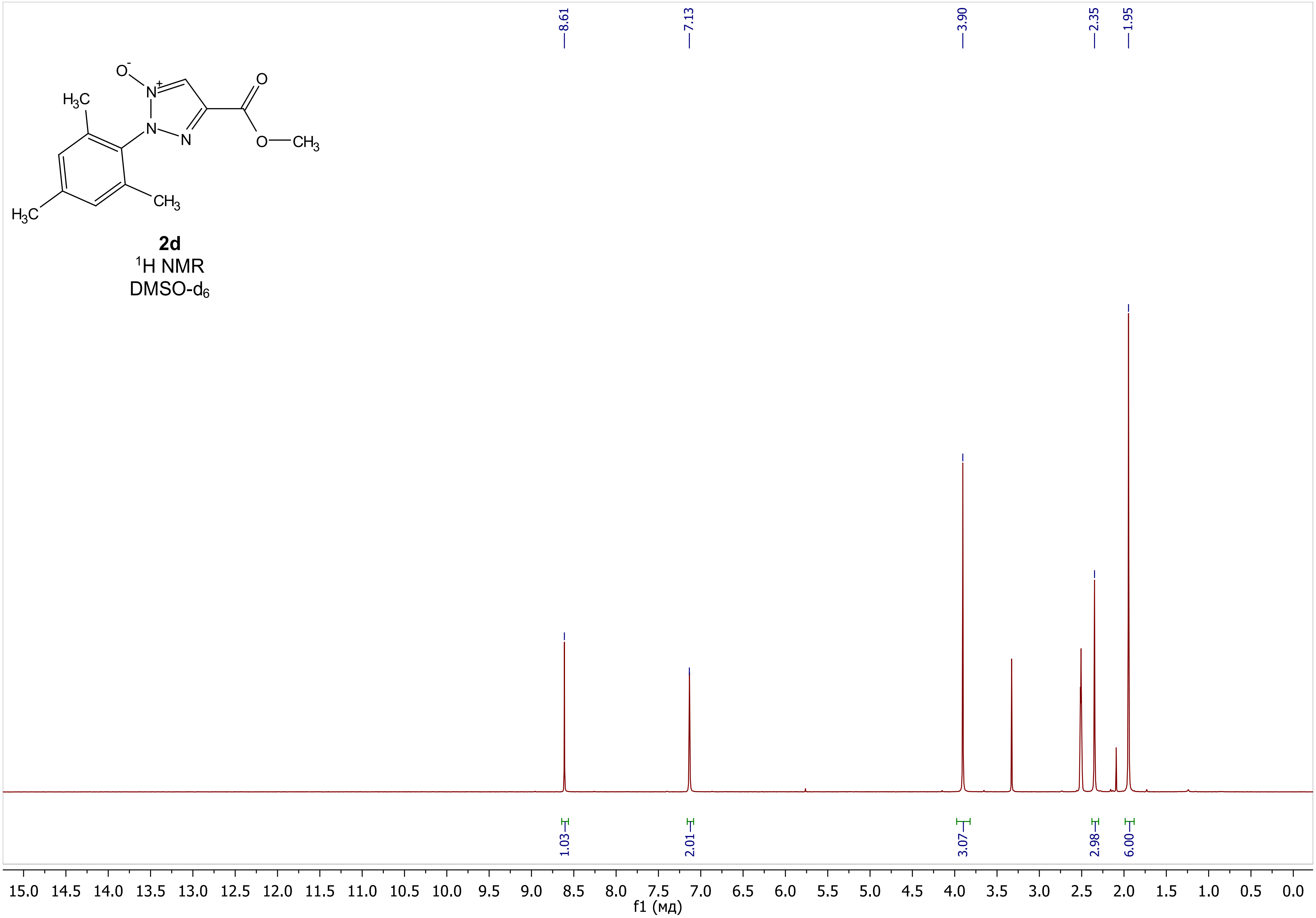
DMSO-d_6



2c

^{13}C NMR

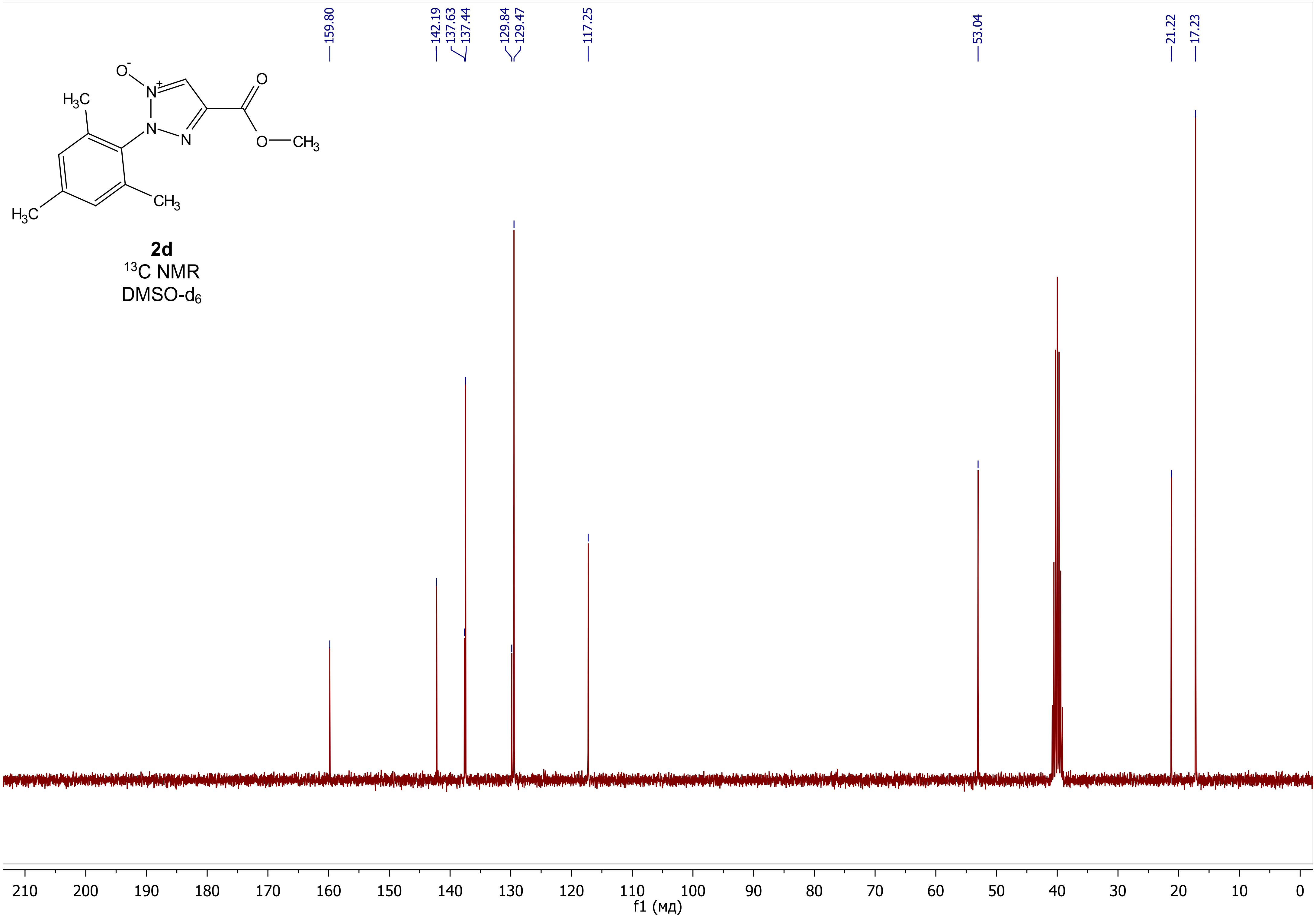
DMSO-d_6



2d

^{1}H NMR

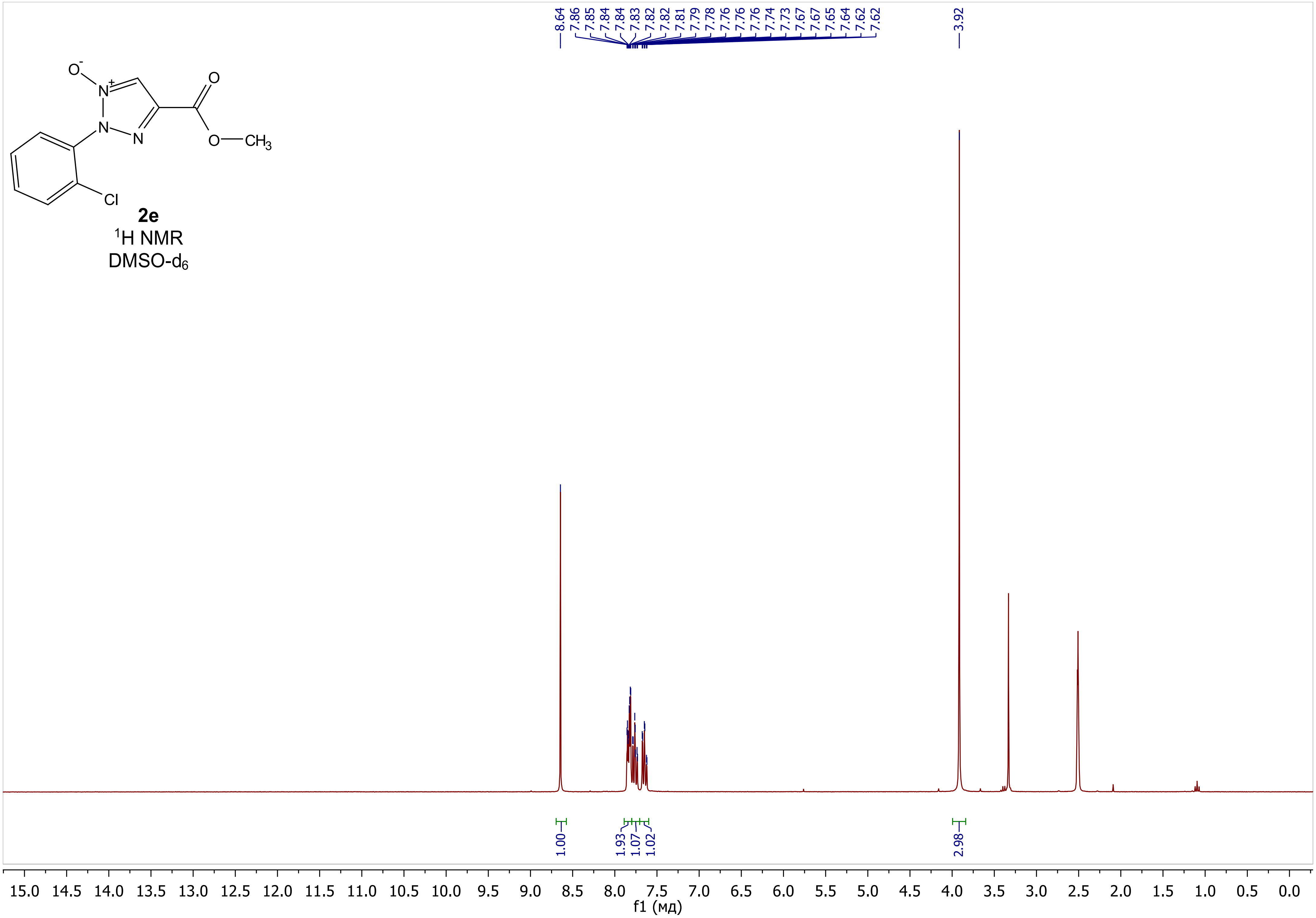
DMSO-d_6

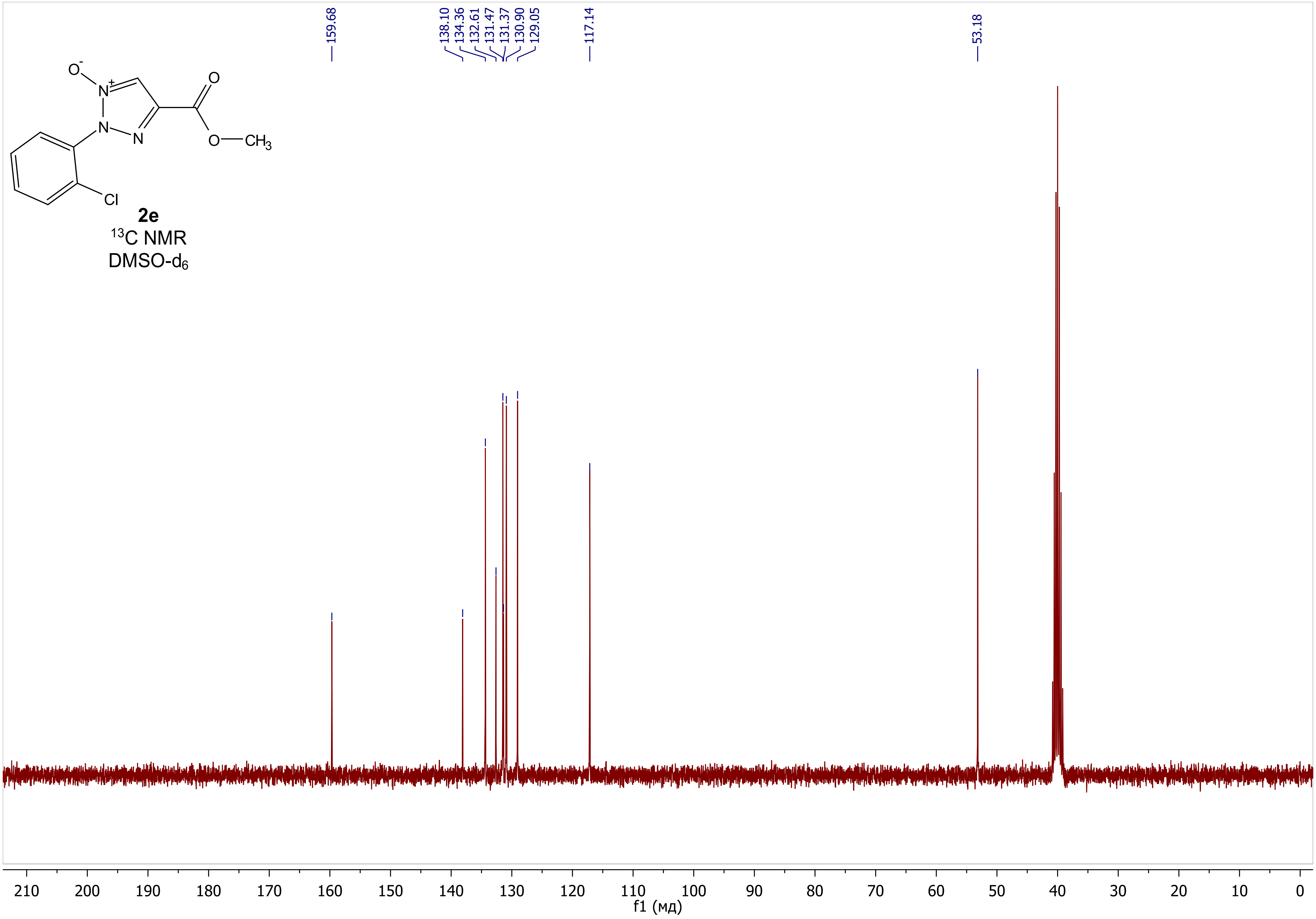


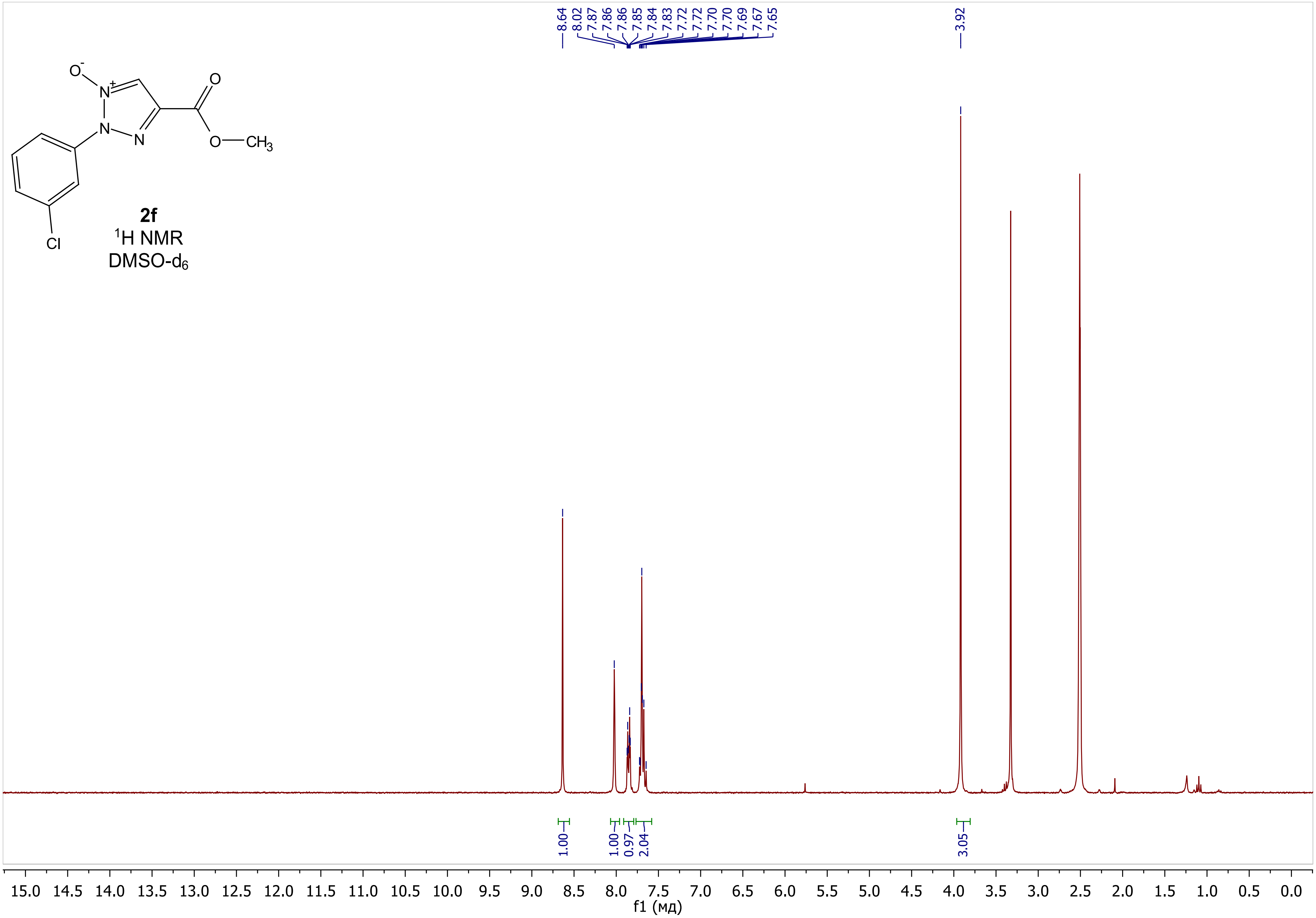
2d

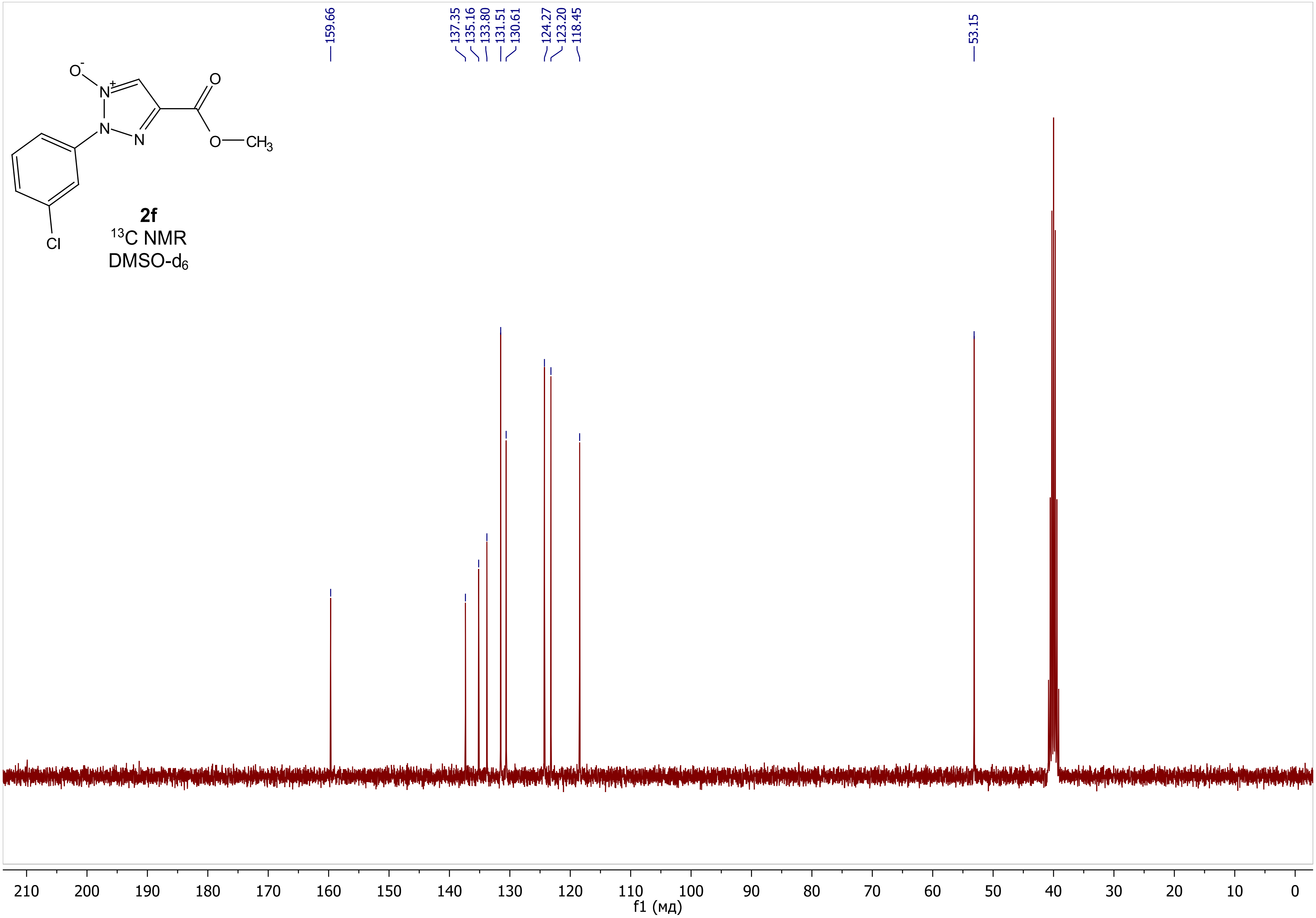
^{13}C NMR

DMSO-d$_6$


2e
^{1}H NMR
DMSO-d$_6$
O$^-$
N$^+$
N
N
O
O—CH$_3$
Cl
8.64
7.86
7.85
7.84
7.84
7.83
7.82
7.82
7.81
7.79
7.78
7.76
7.76
7.76
7.74
7.73
7.67
7.67
7.65
7.64
7.62
7.62
3.92
1.00
1.93
1.07
1.02
2.98
15.0 14.5 14.0 13.5 13.0 12.5 12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (мд)


159.68
138.10
134.36
132.61
131.47
131.37
130.90
129.05
117.14
53.18
O-
N+
N
N
O
O
CH3
Cl
2e
13C NMR
DMSO-d6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)

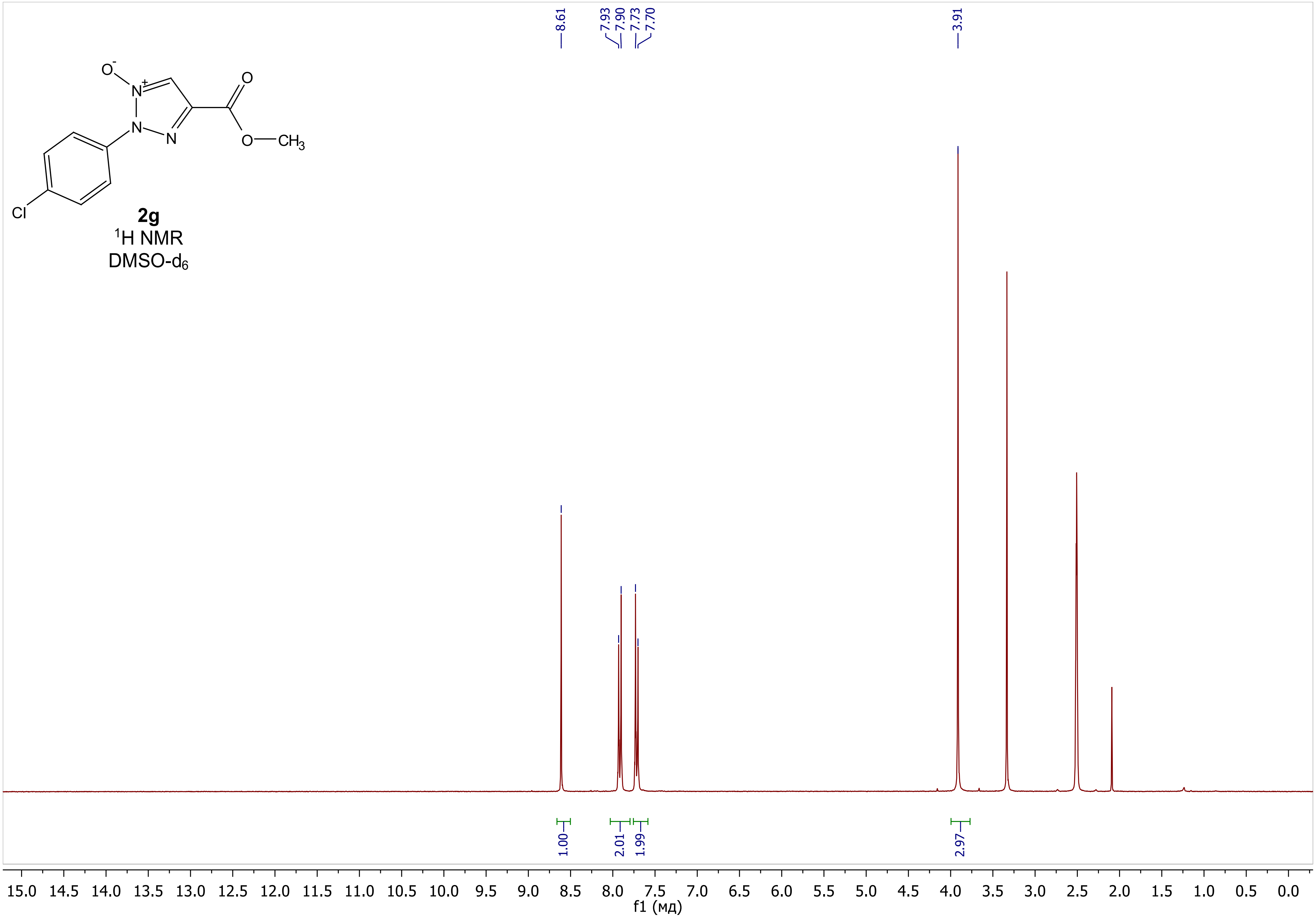

2f
^{1}H NMR
DMSO-d_6
O
N
N
N
O
O
CH$_3$
Cl
8.64
8.02
7.87
7.86
7.86
7.85
7.84
7.83
7.72
7.72
7.70
7.70
7.69
7.67
7.65
3.92
1.00
1.00
0.97
2.04
3.05
15.0 14.5 14.0 13.5 13.0 12.5 12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (мд)



2f

^{13}C NMR

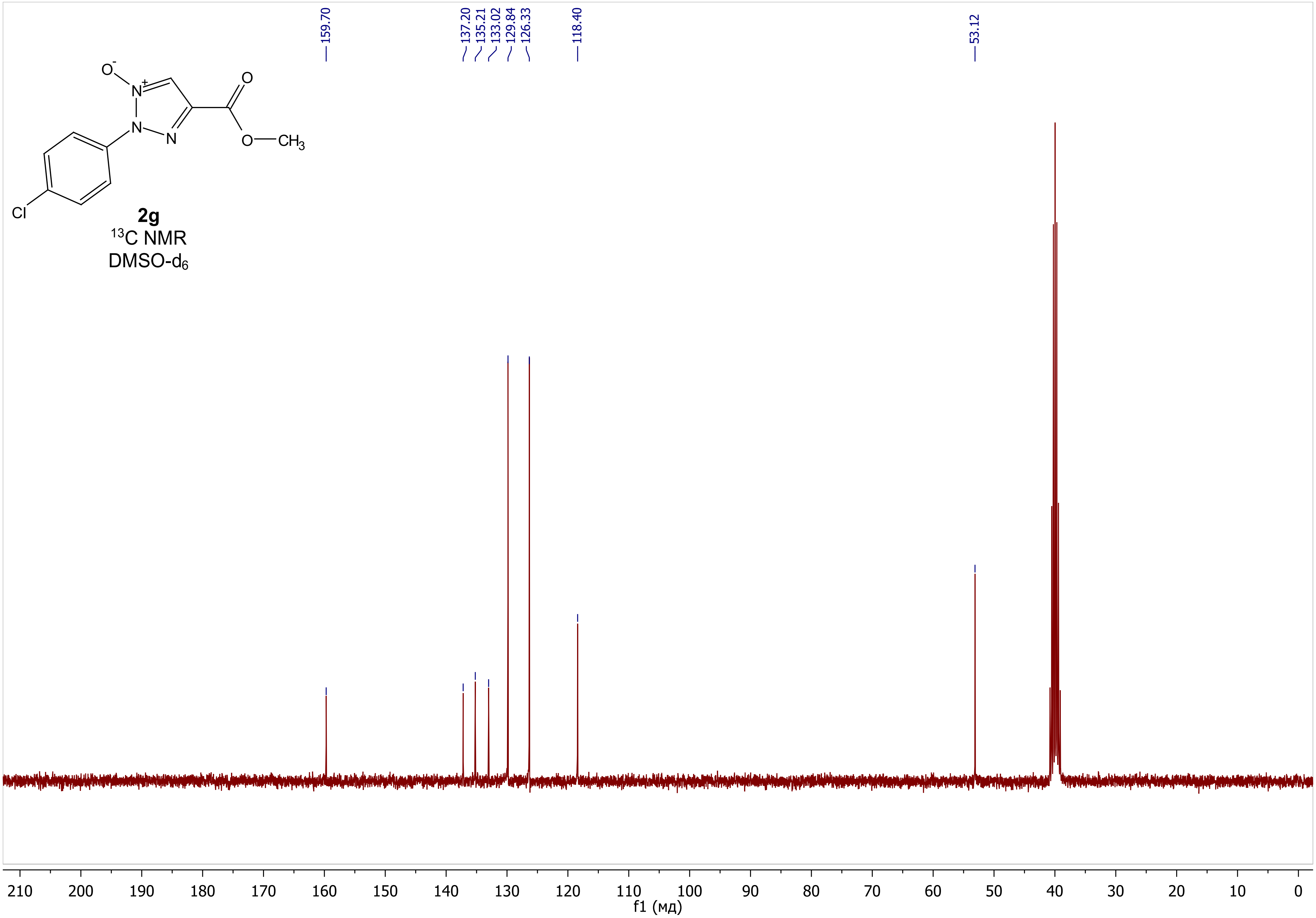
DMSO-d_6



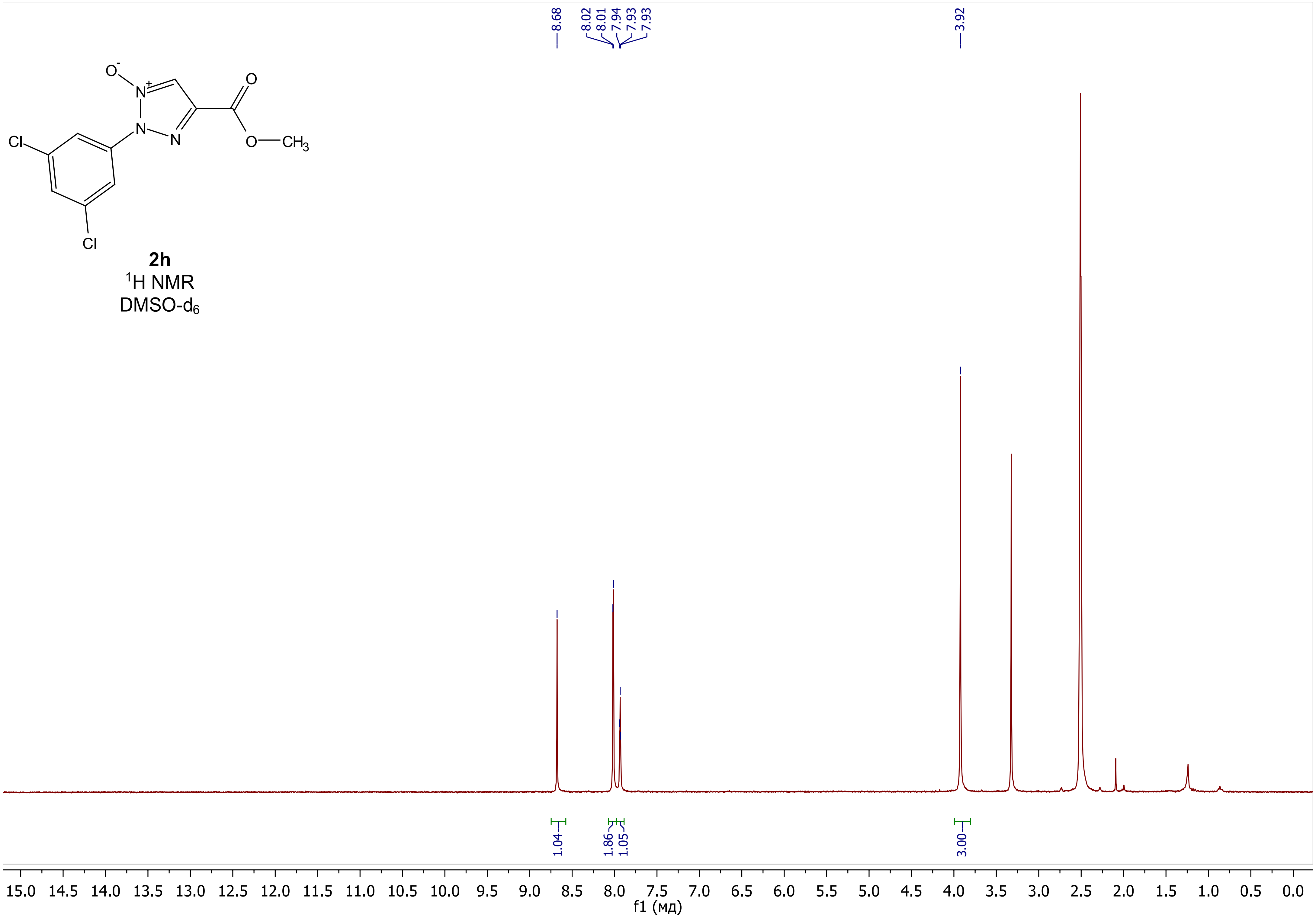
2g

^{1}H NMR

DMSO-d_6


2g

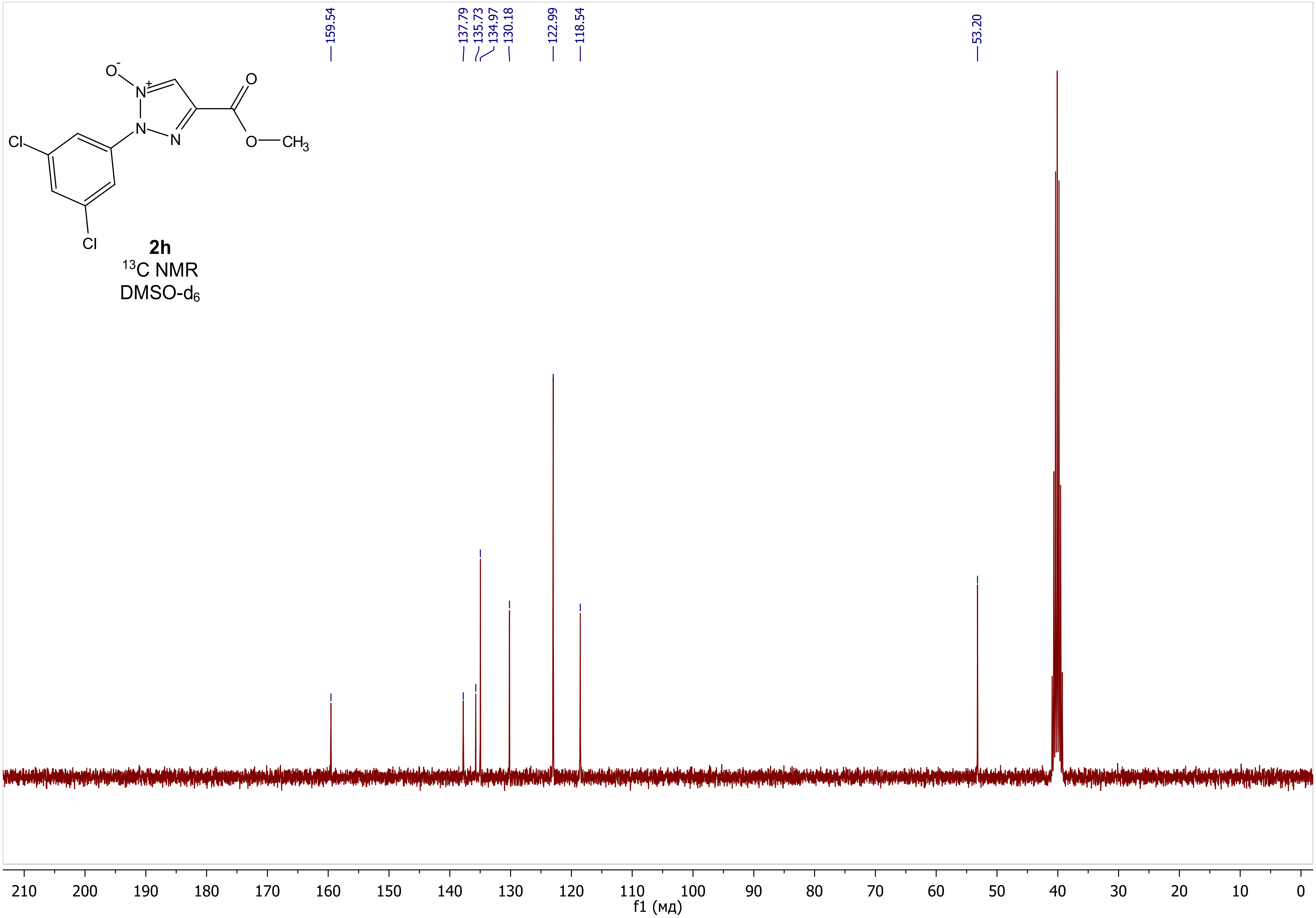
O⁻
N⁺
N
N
O
O
CH3
Cl
^{13}C NMR
DMSO-d6
159.70
137.20
135.21
133.02
129.84
126.33
118.40
53.12
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)

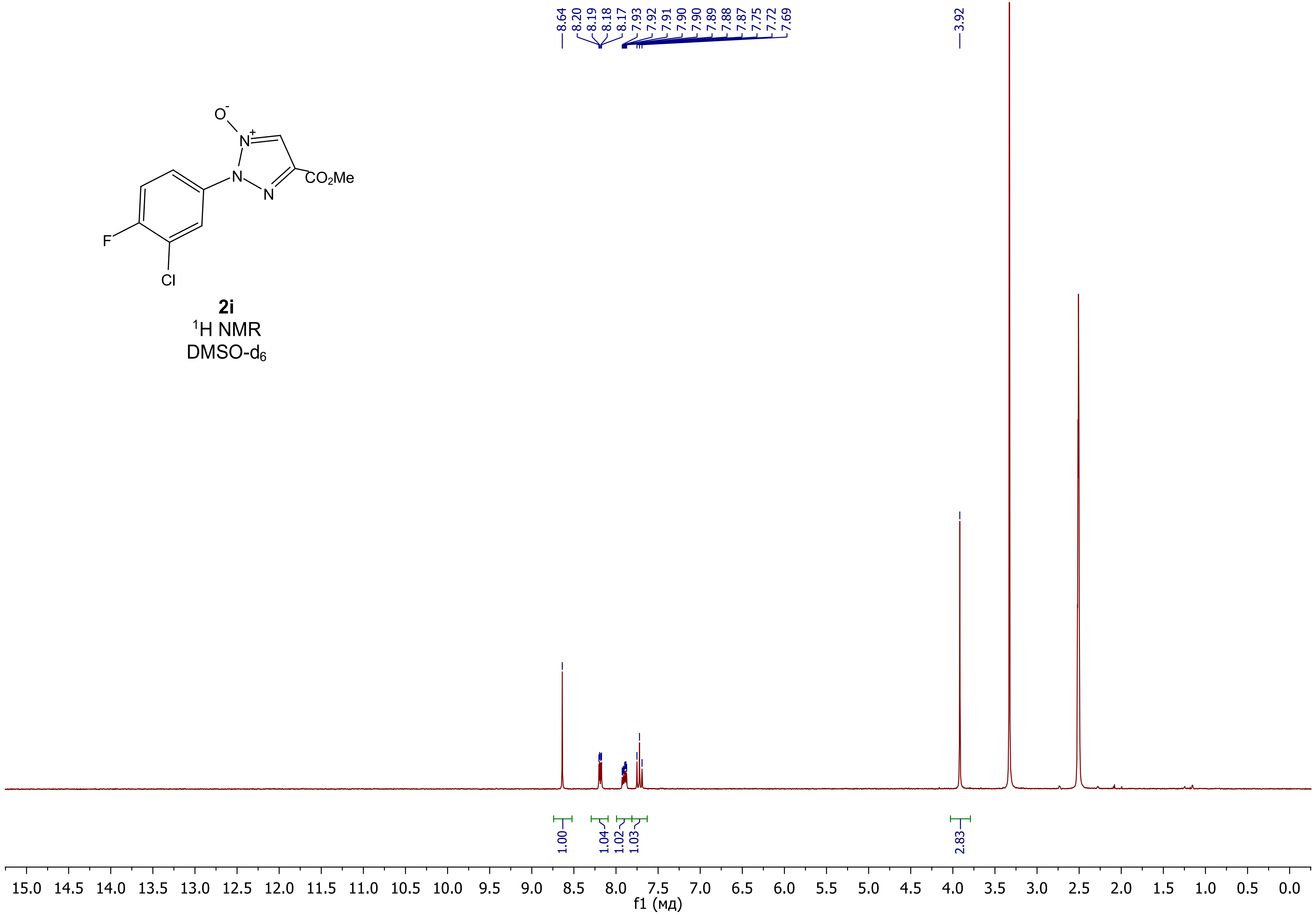


2h

^{1}H NMR

DMSO-d_6

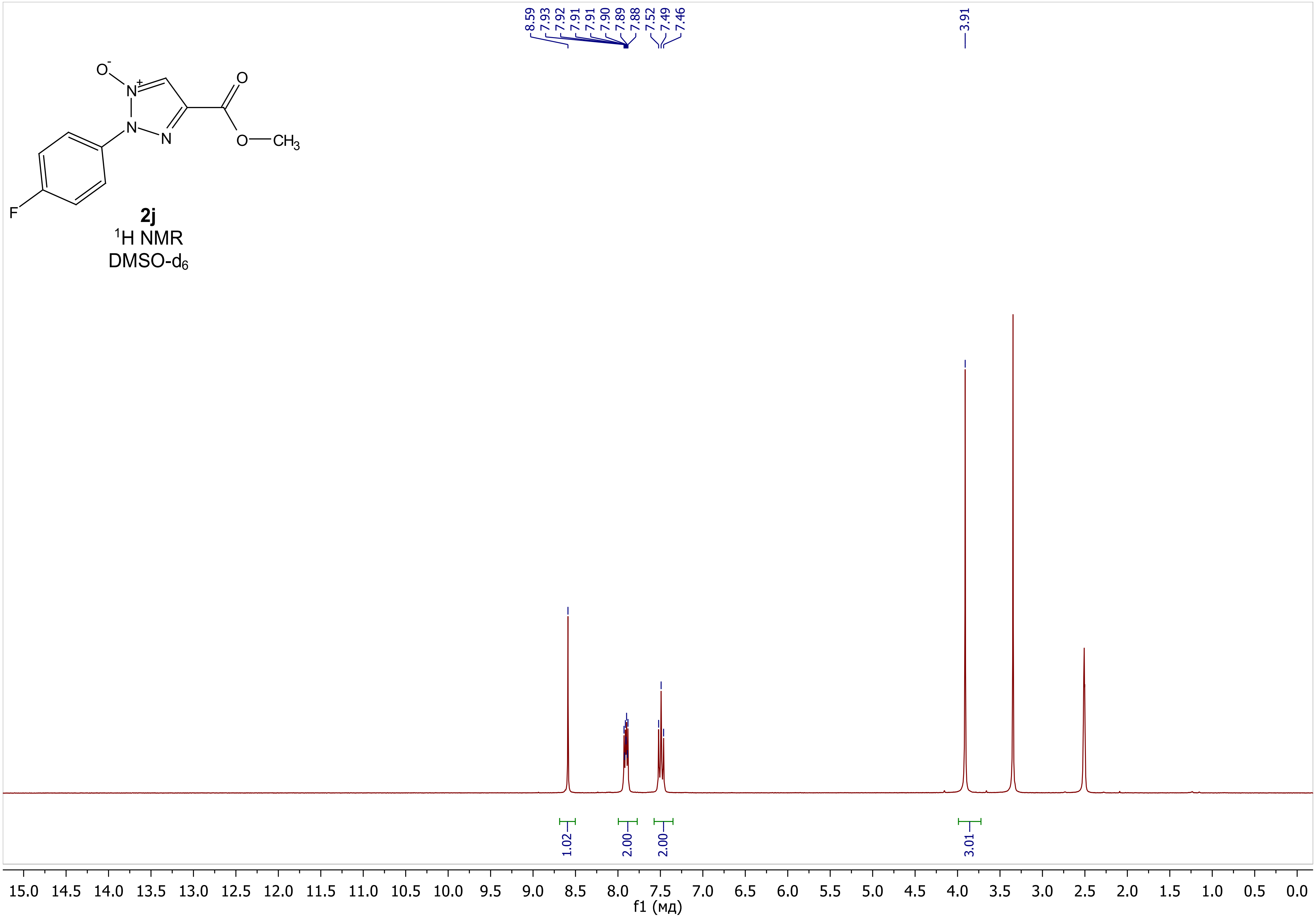

159.54
137.79
135.73
134.97
130.18
122.99
118.54
53.20
O
N
N
N
O
O
CH3
Cl
Cl
2h
13C NMR
DMSO-d6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)



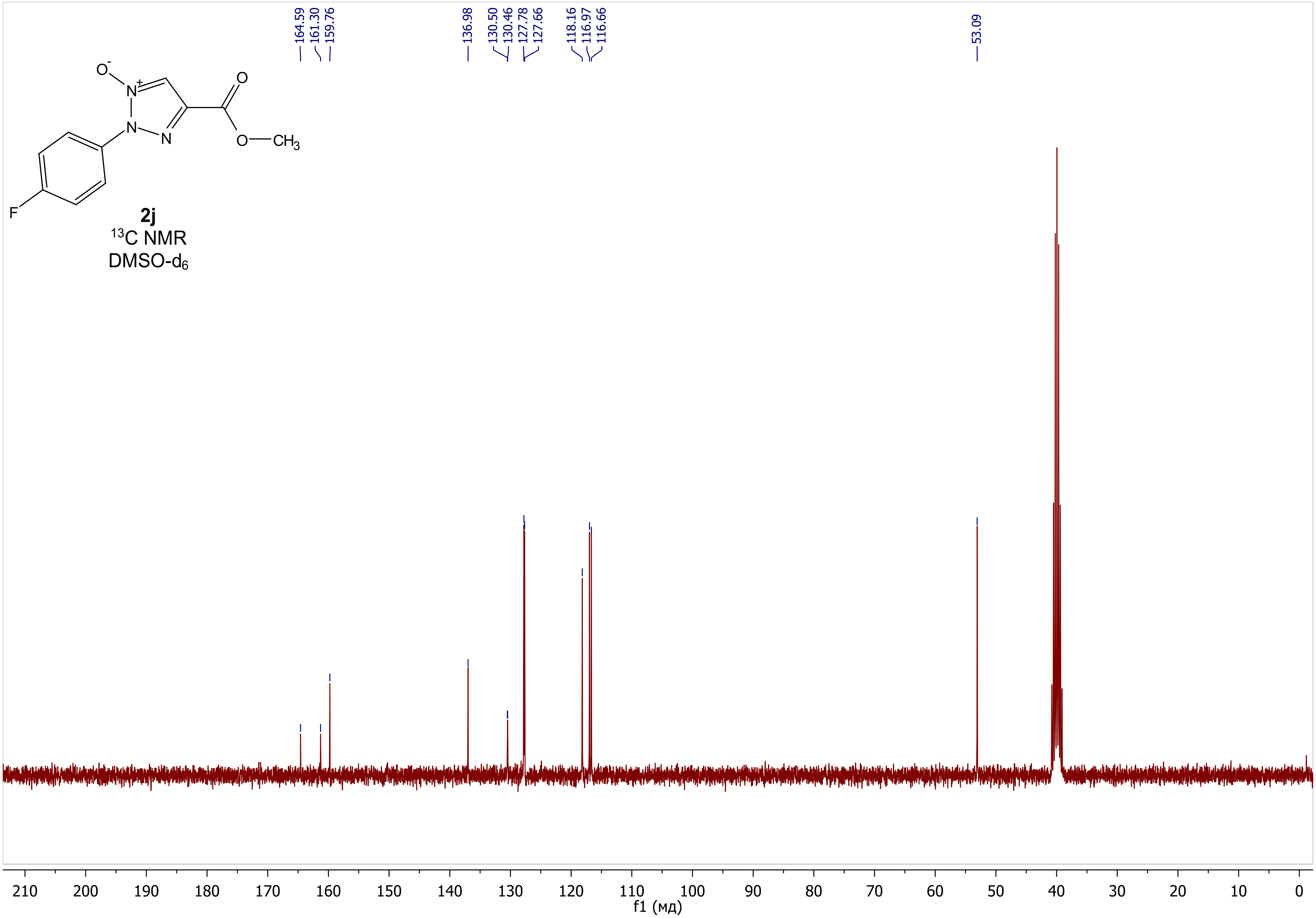
2i

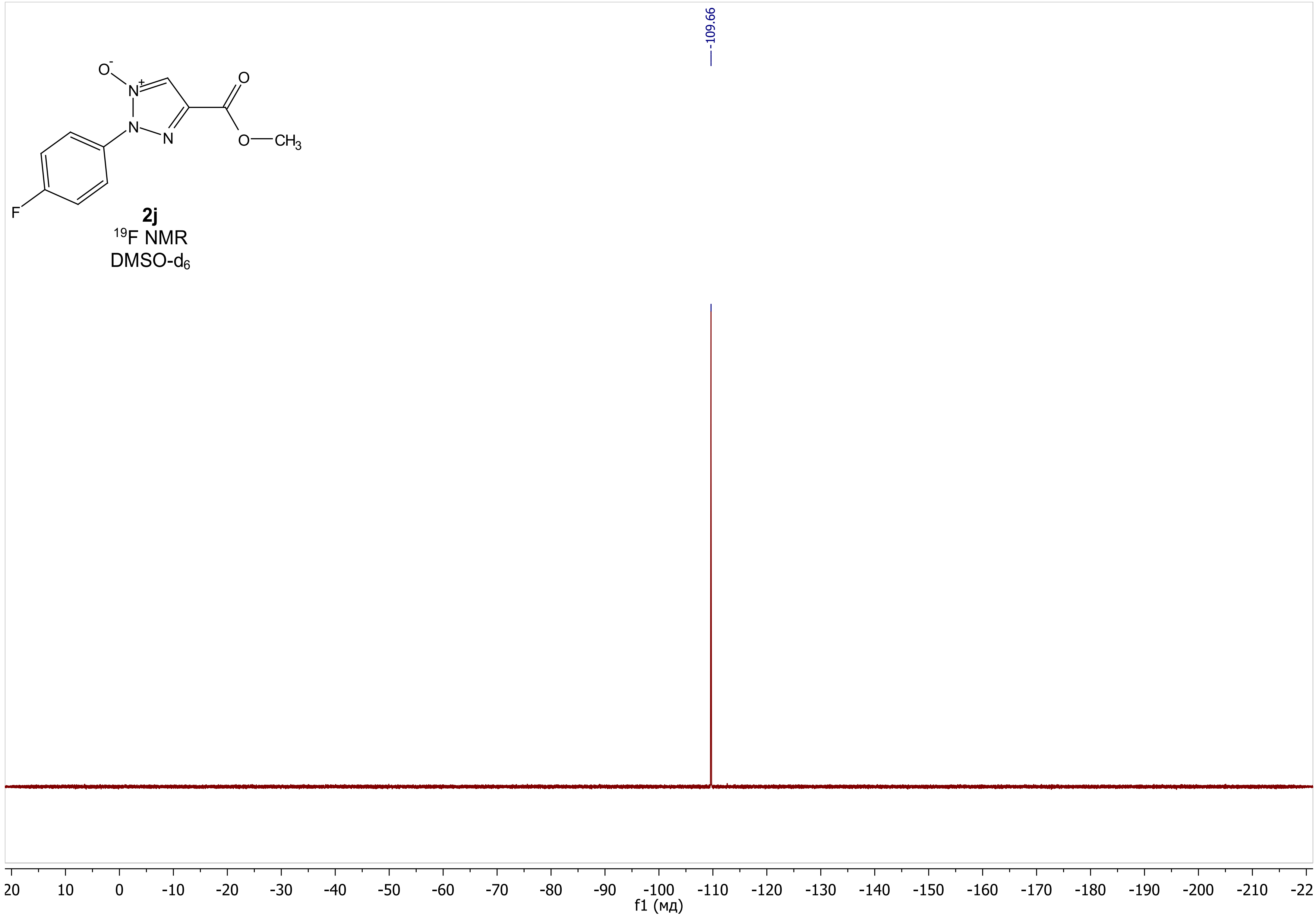
^{1}H NMR

DMSO-d_6



2j
^{1}H NMR
DMSO-d$_6$
8.59
7.93
7.92
7.91
7.91
7.90
7.89
7.88
7.52
7.49
7.46
3.91
1.02
2.00
2.00
3.01
15.0 14.5 14.0 13.5 13.0 12.5 12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (мд)

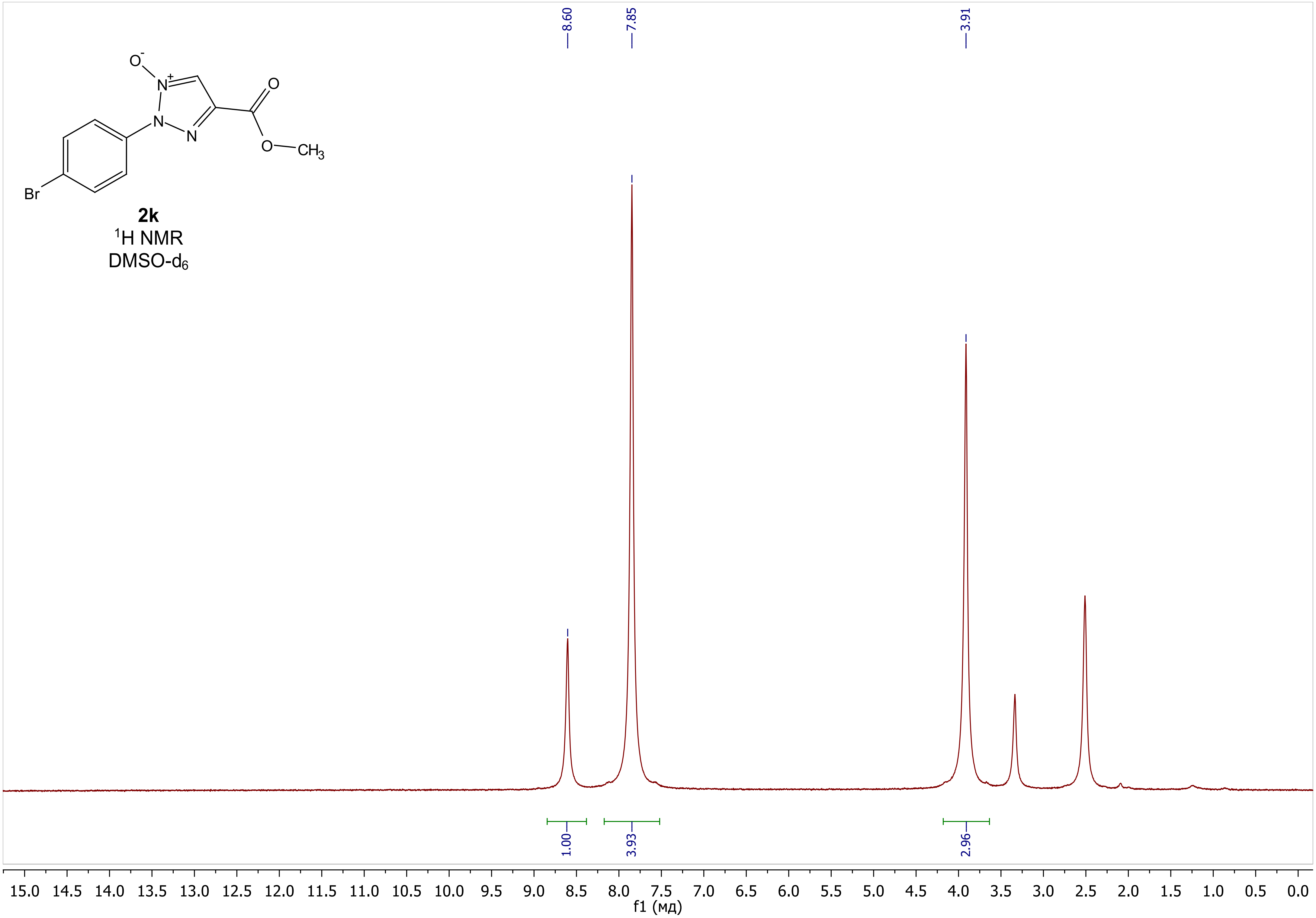

164.59
161.30
159.76
136.98
130.50
130.46
127.78
127.66
118.16
116.97
116.66
53.09
O
N
+
N
N
O
O
CH3
F
2j
13C NMR
DMSO-d6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)

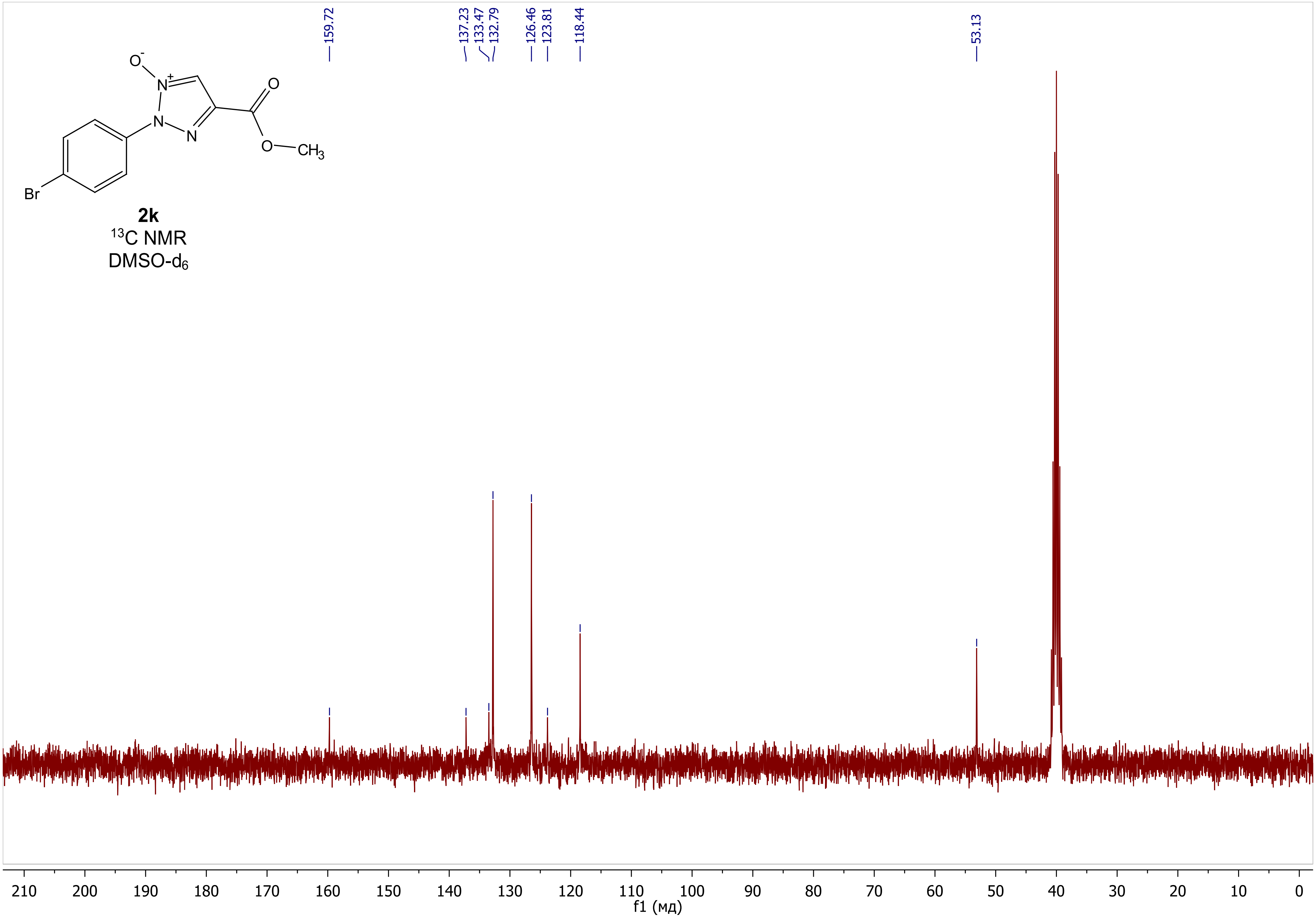


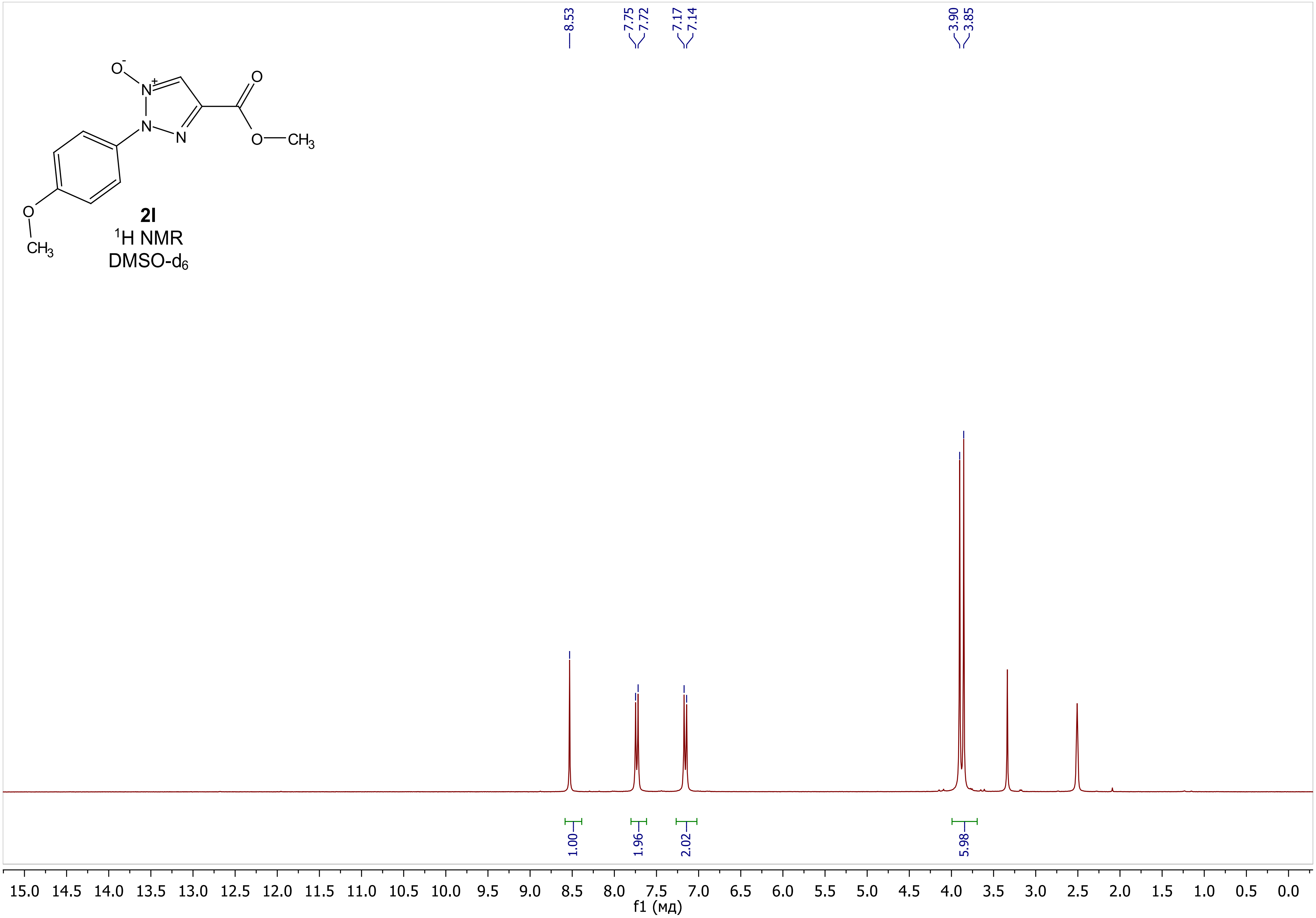
2j

^{19}F NMR

DMSO-d_6


O-
N+
N
N
O
O
CH3
Br
2k
^{1}H NMR
DMSO-d$_6$
8.60
7.85
3.91
1.00
3.93
2.96
15.0
14.5
14.0
13.5
13.0
12.5
12.0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (мд)

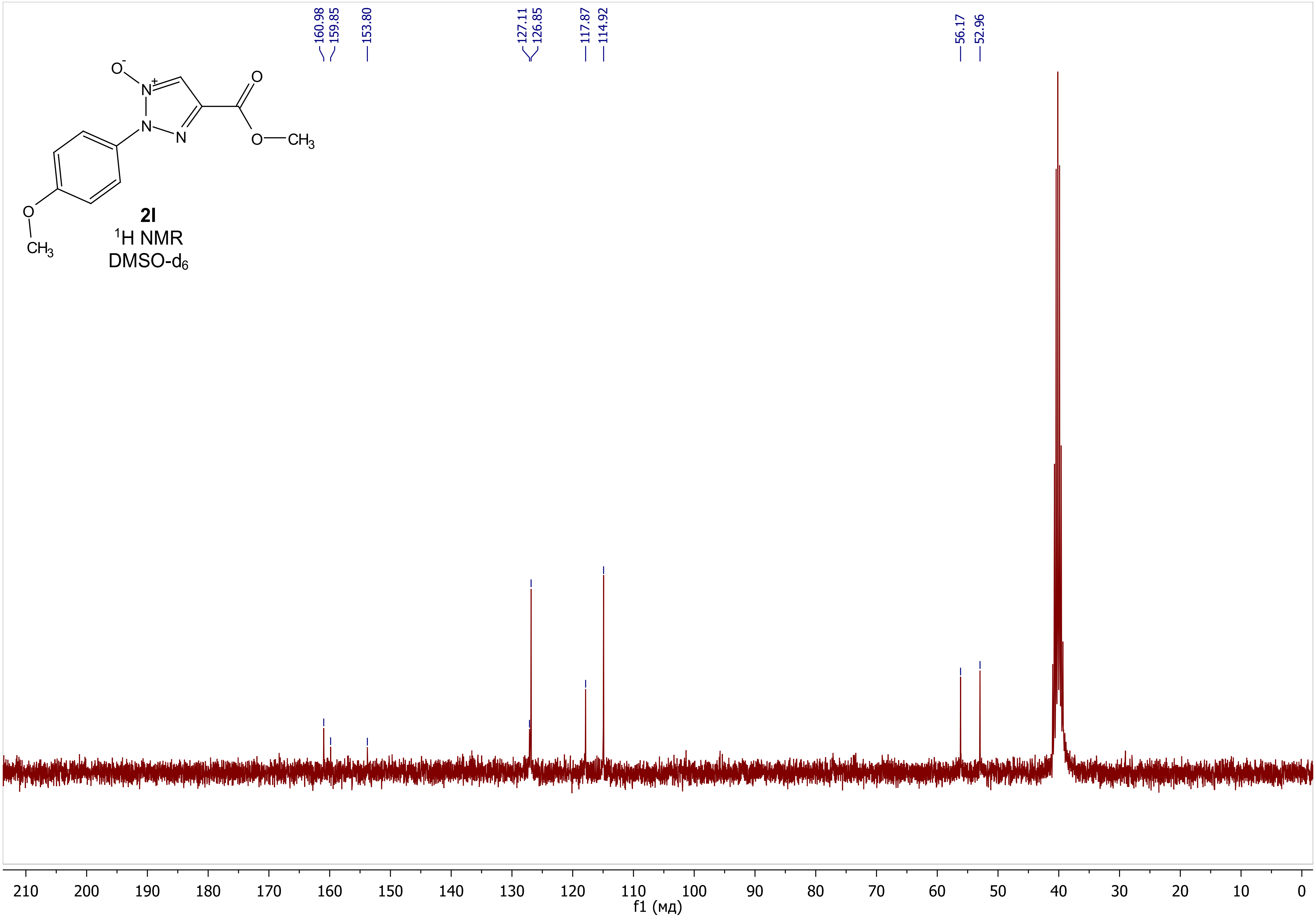

159.72
137.23
133.47
132.79
126.46
123.81
118.44
53.13
O-
N+
N
N
O
O
CH3
Br
2k
^{13}C NMR
DMSO-d$_6$
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)

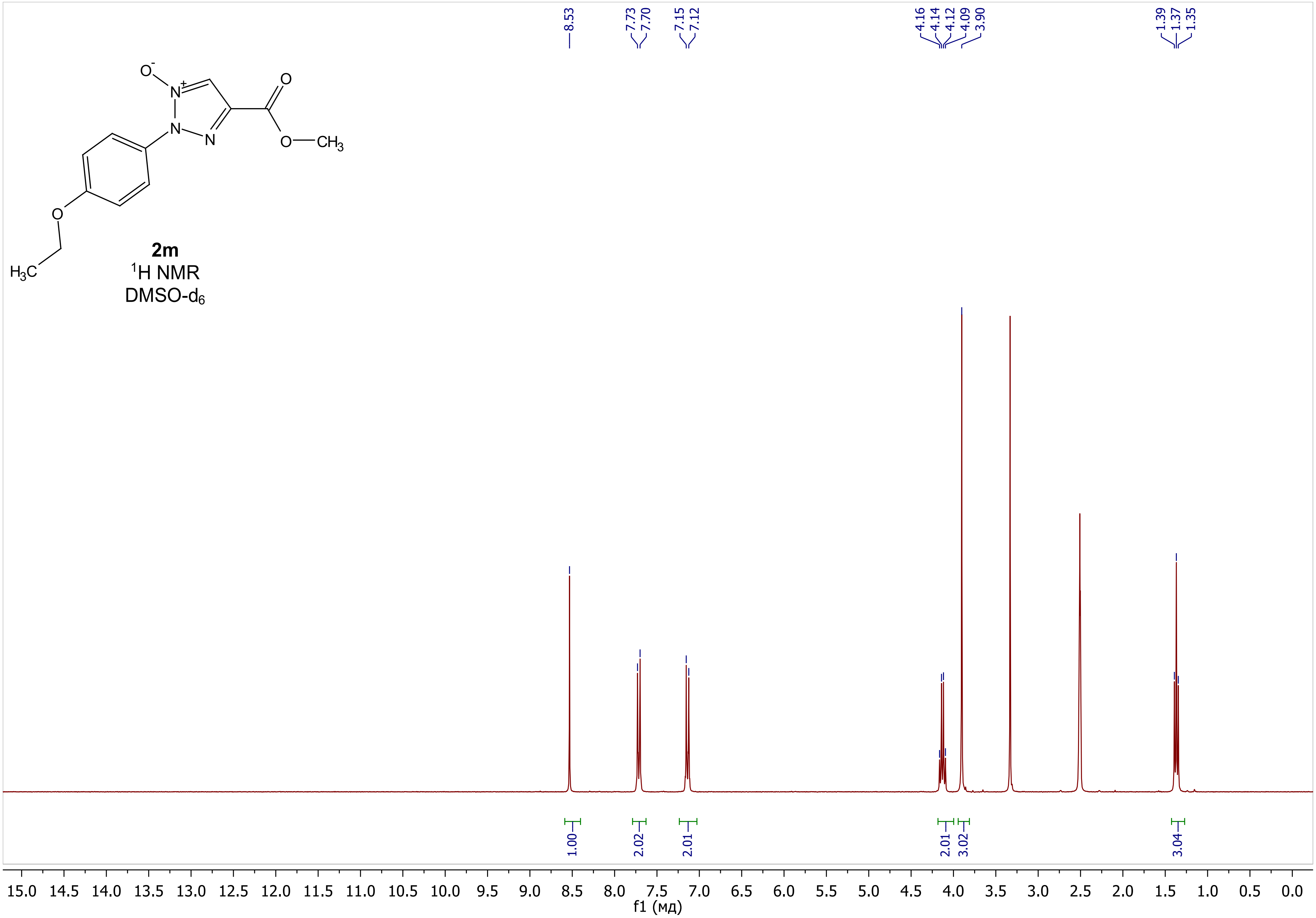


2l

^{1}H NMR

DMSO-d_6

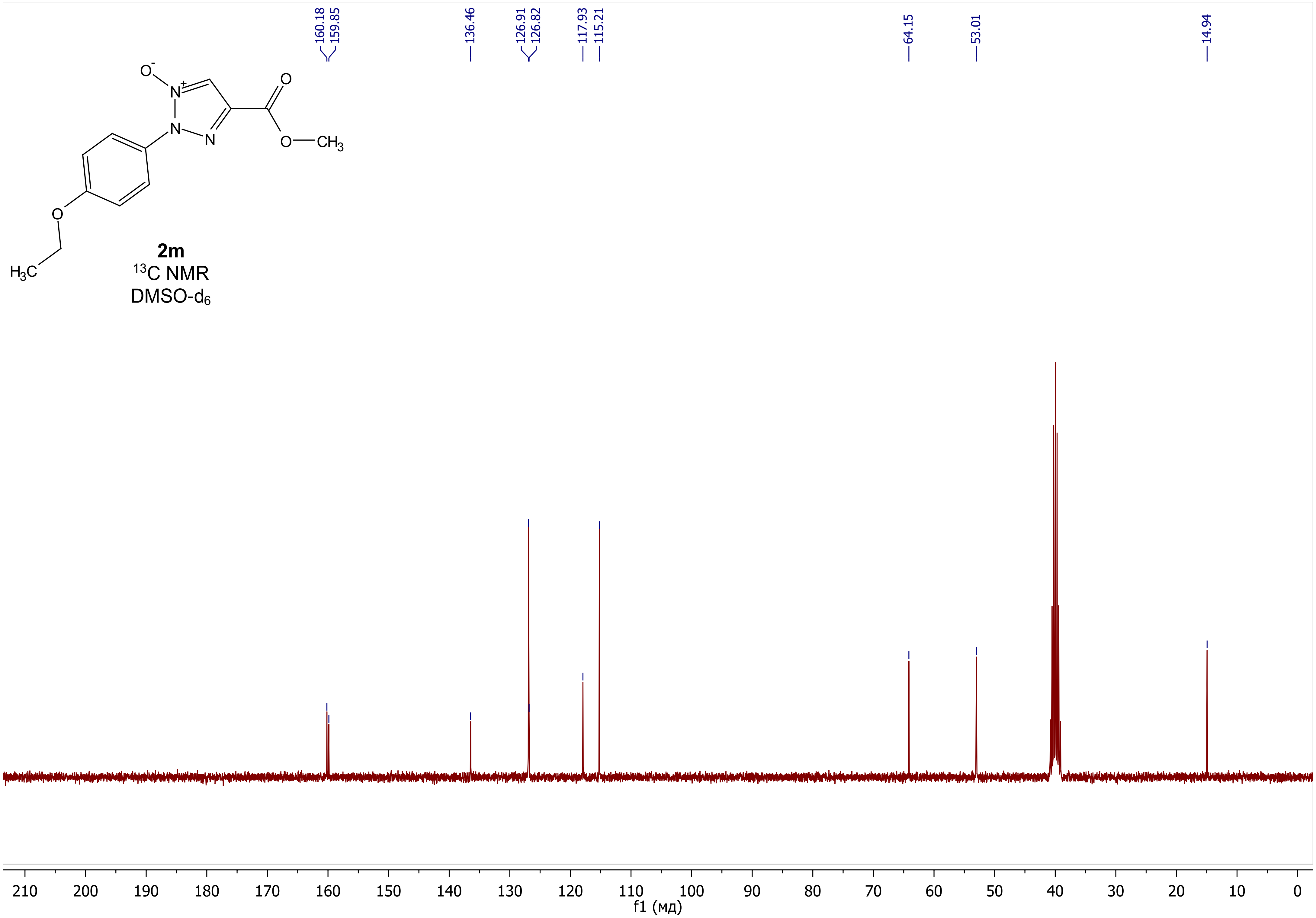

160.98
159.85
153.80
127.11
126.85
117.87
114.92
56.17
52.96
O⁻
N⁺
N
N
O
O
CH3
O
CH3
2l
^{1}H NMR
DMSO-d$_6$
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)



2m

^{1}H NMR

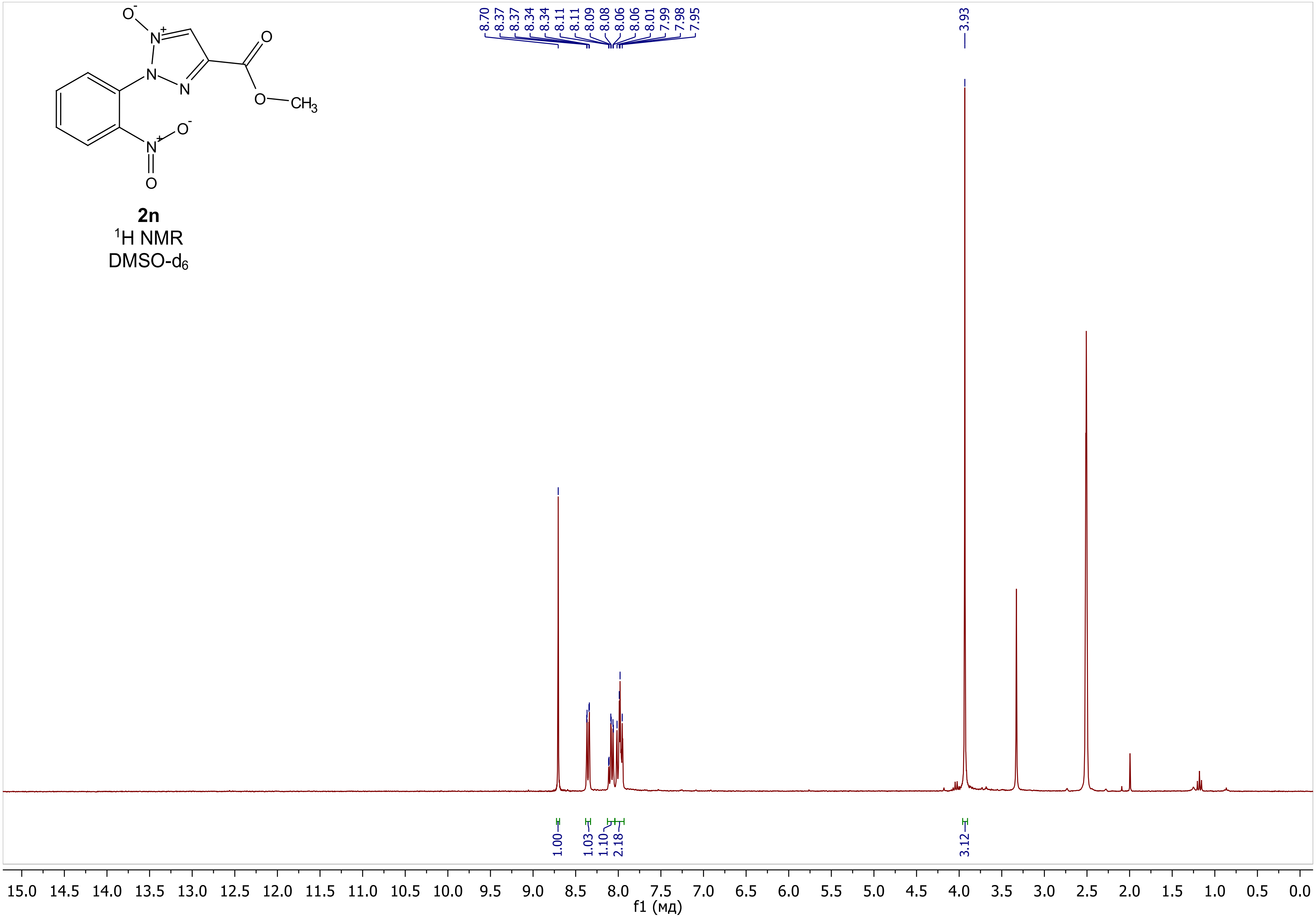
DMSO-d_6



2m

^{13}C NMR

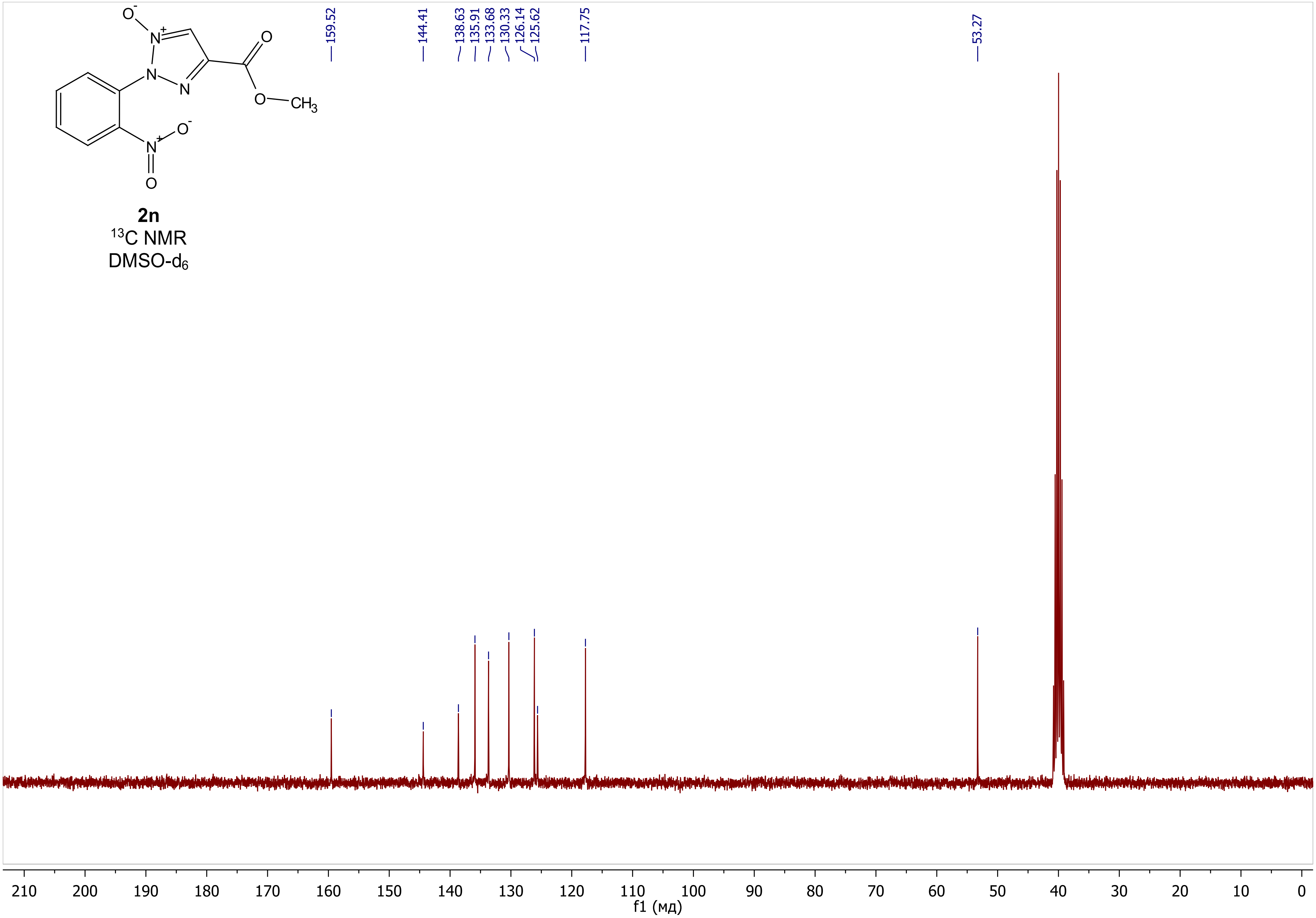
DMSO-d_6



2n

^{1}H NMR

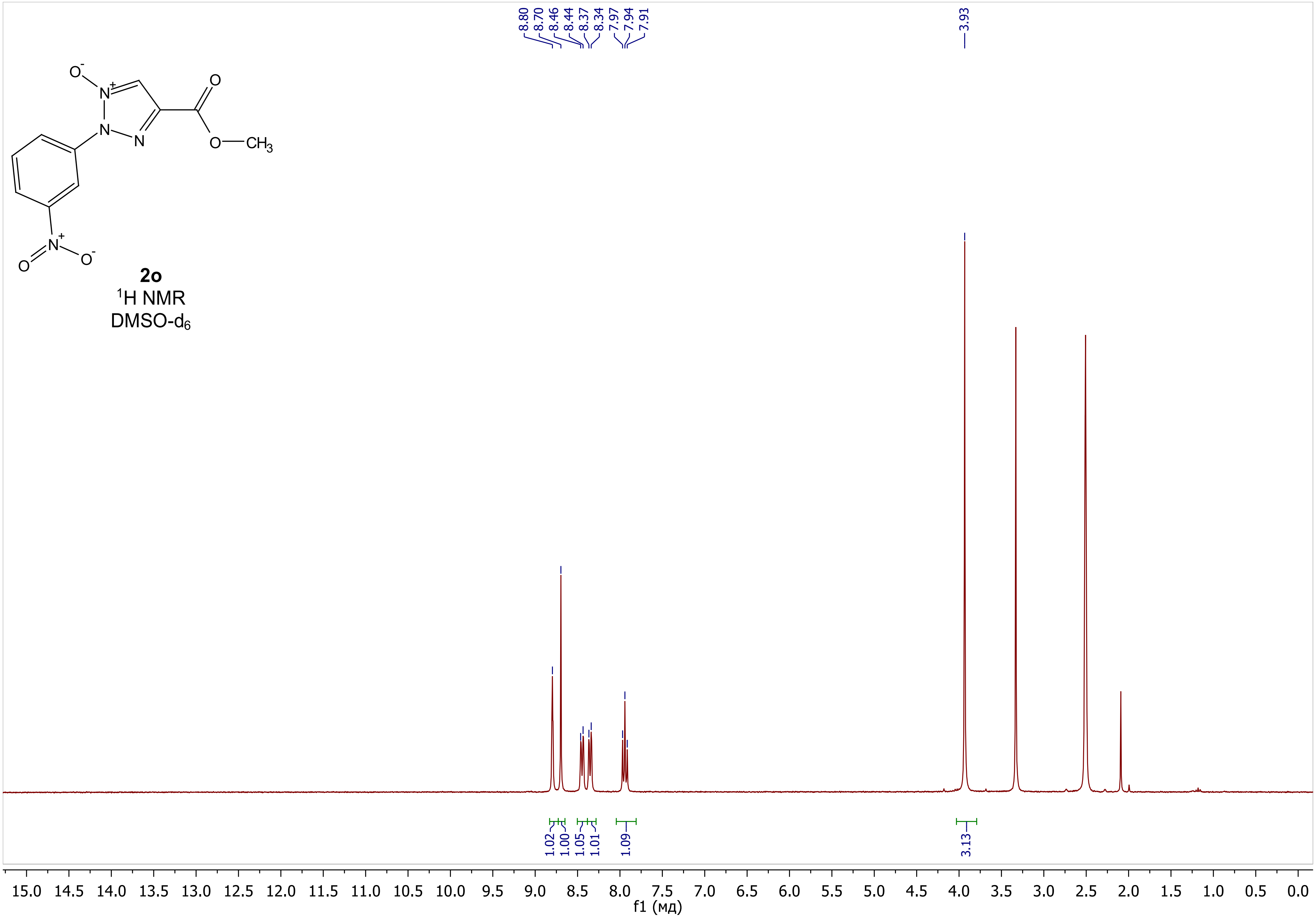
DMSO-d$_6$



2n

^{13}C NMR

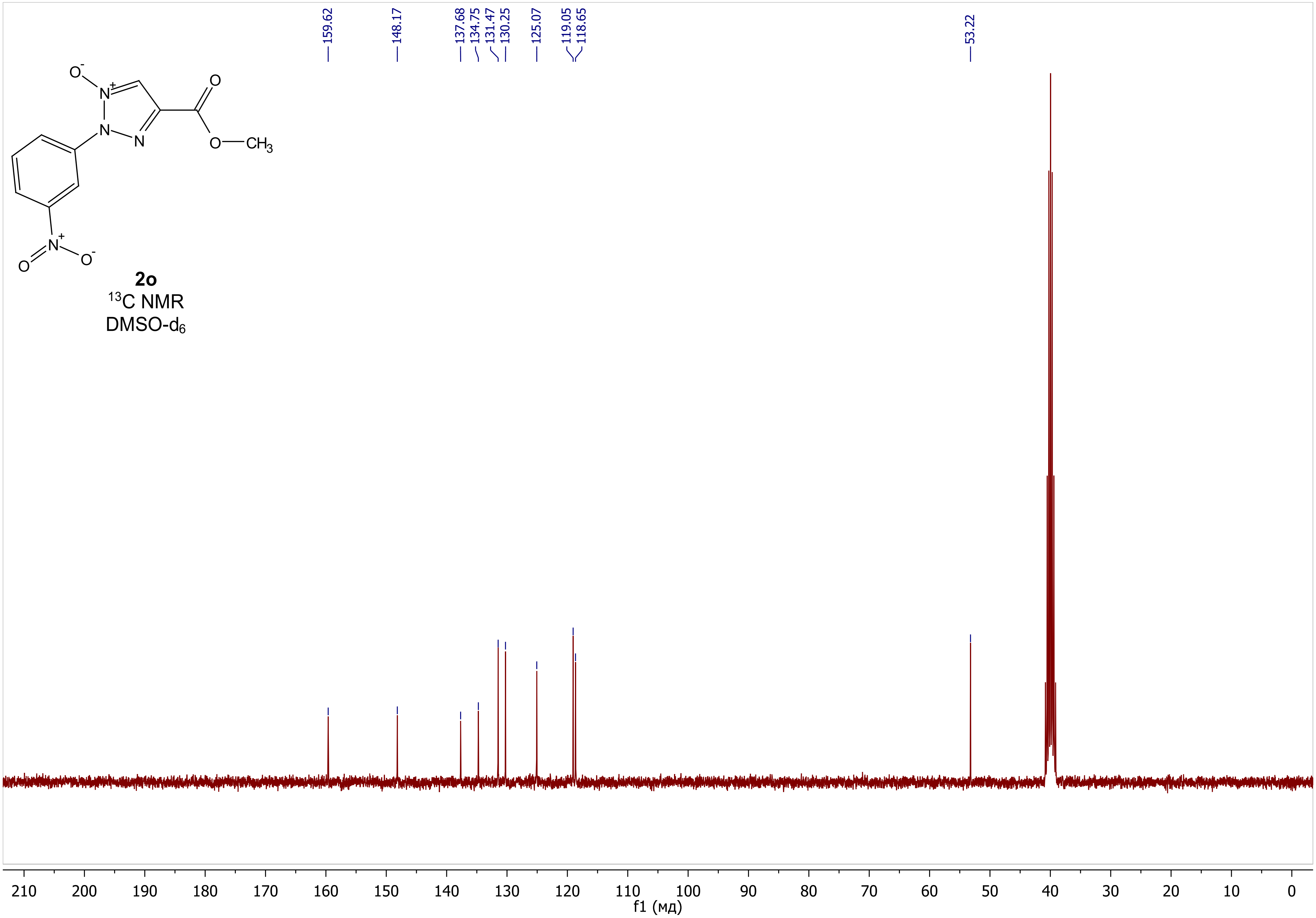
DMSO-d_6

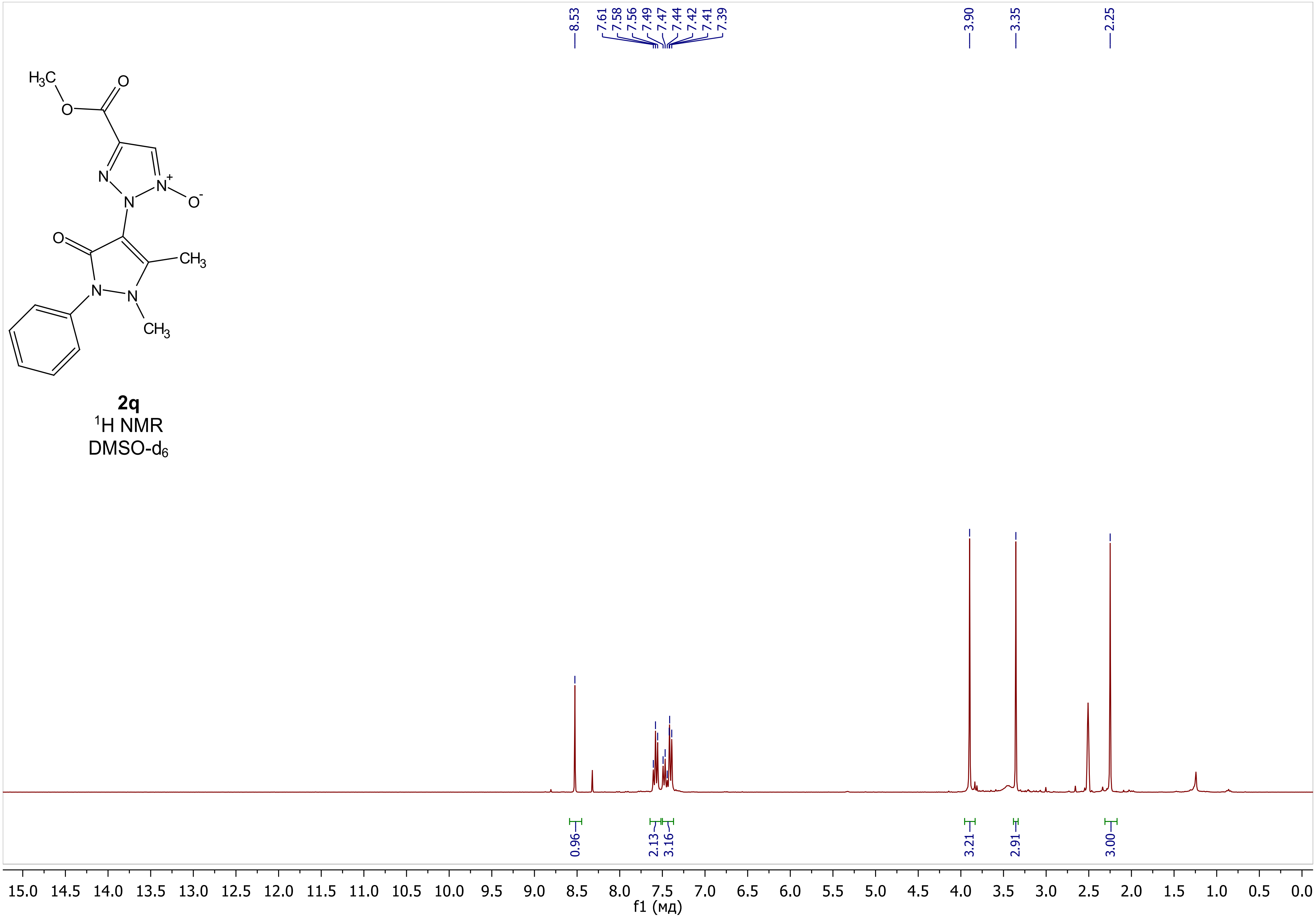


2o

^{1}H NMR

DMSO-d_6

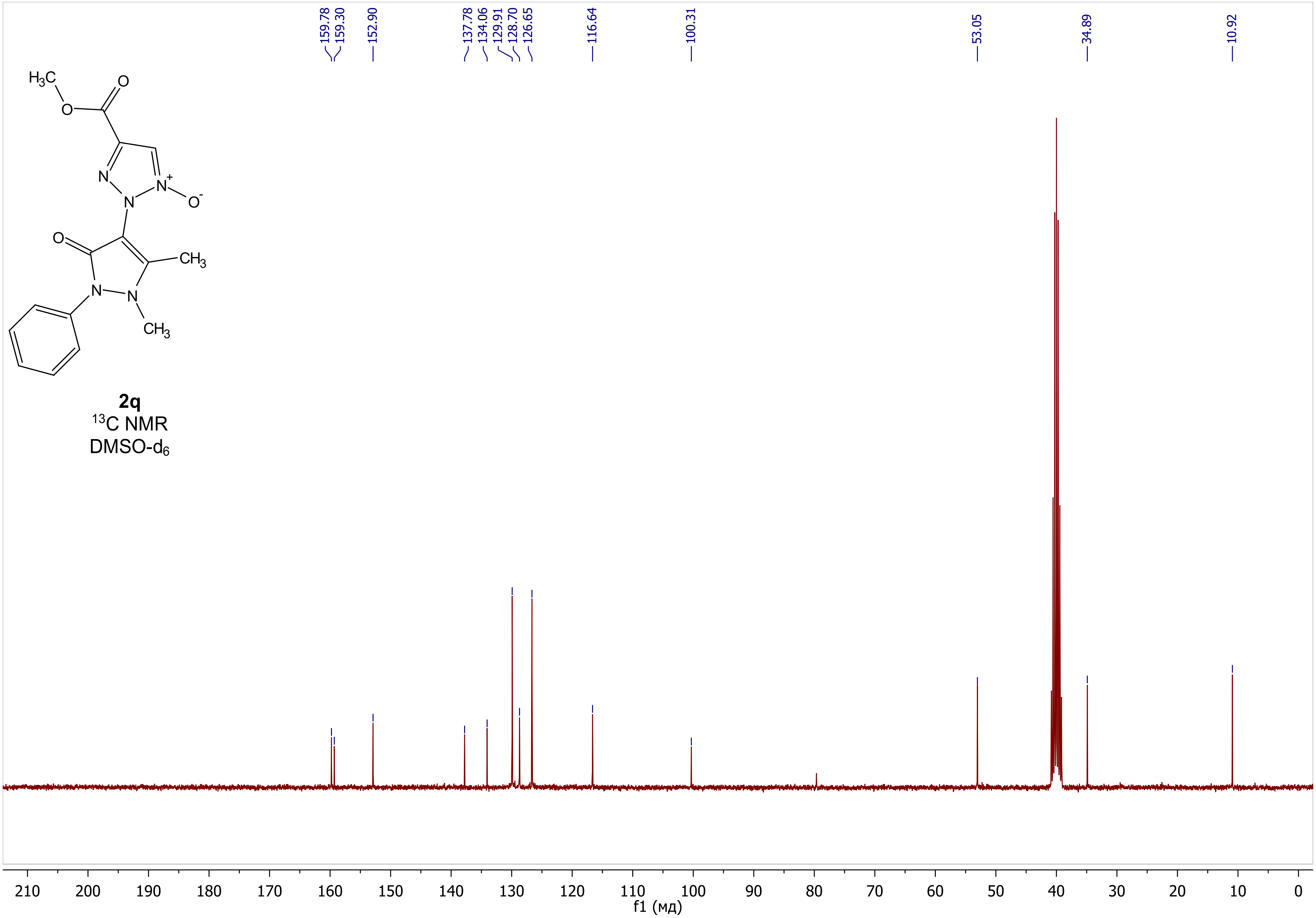

2o
13C NMR
DMSO-d6
159.62
148.17
137.68
134.75
131.47
130.25
125.07
119.05
118.65
53.22
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)
O-
N+
N
N
O
O
CH3
N+
O
O-



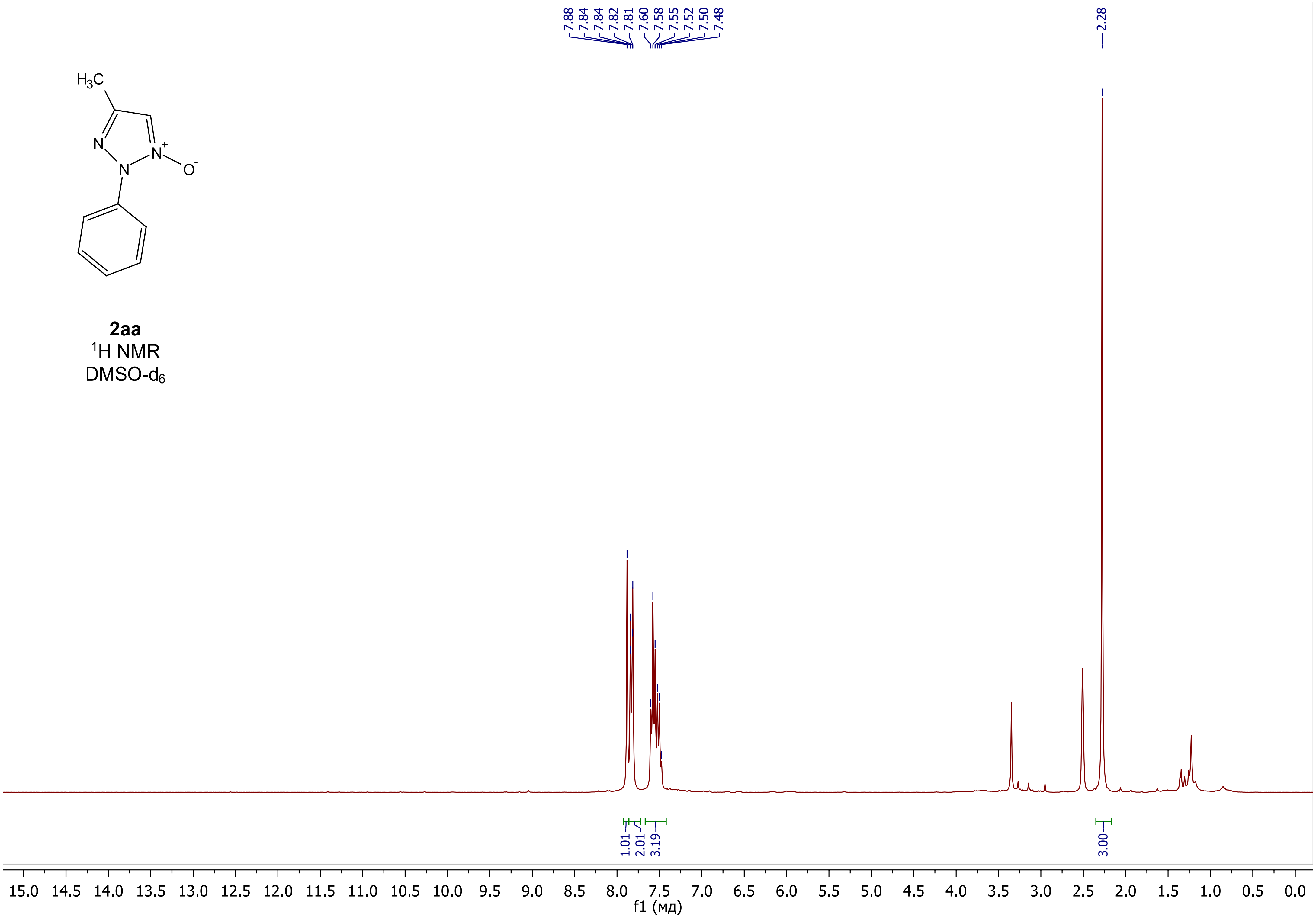
2q

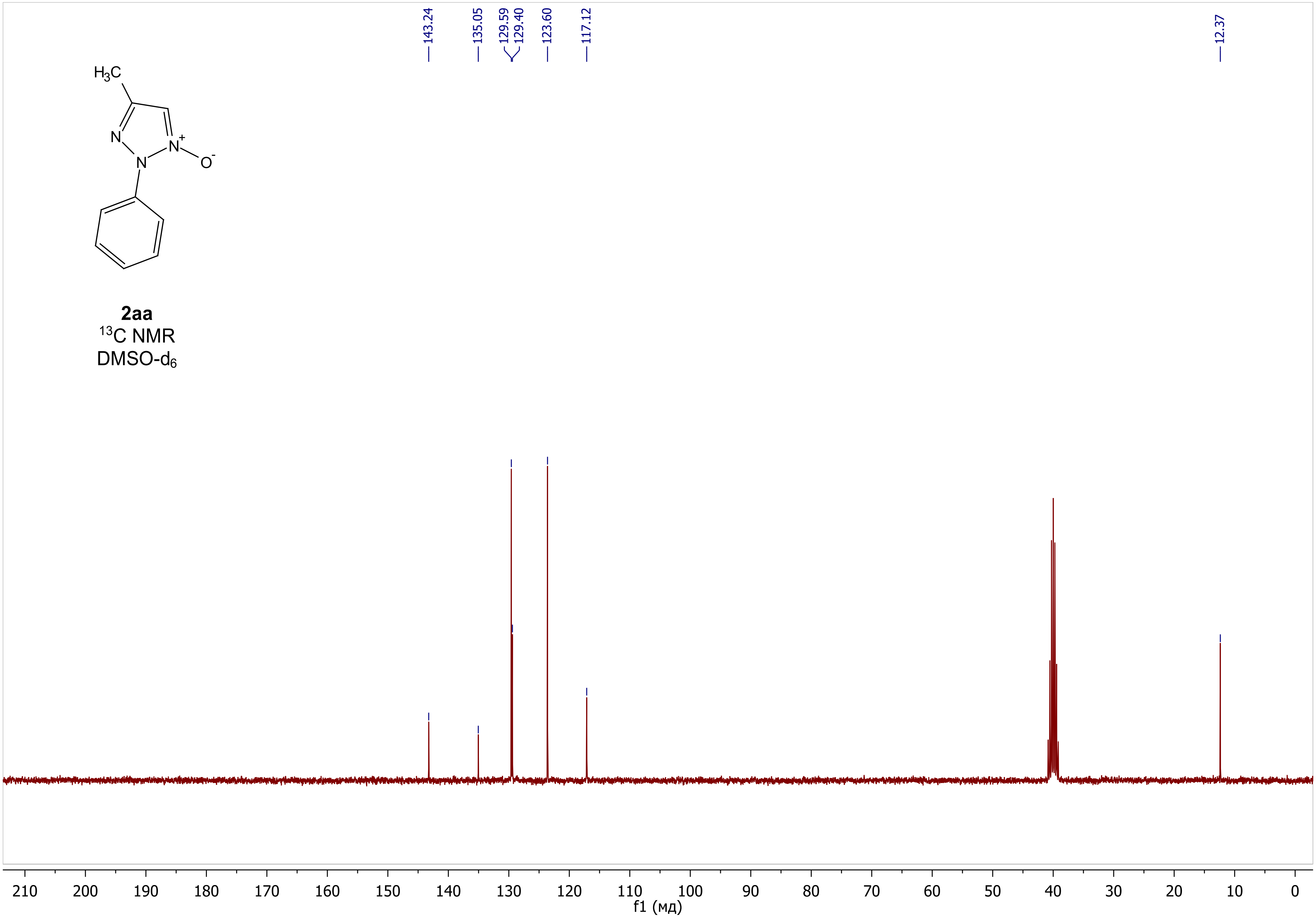
^{1}H NMR

DMSO-d_6

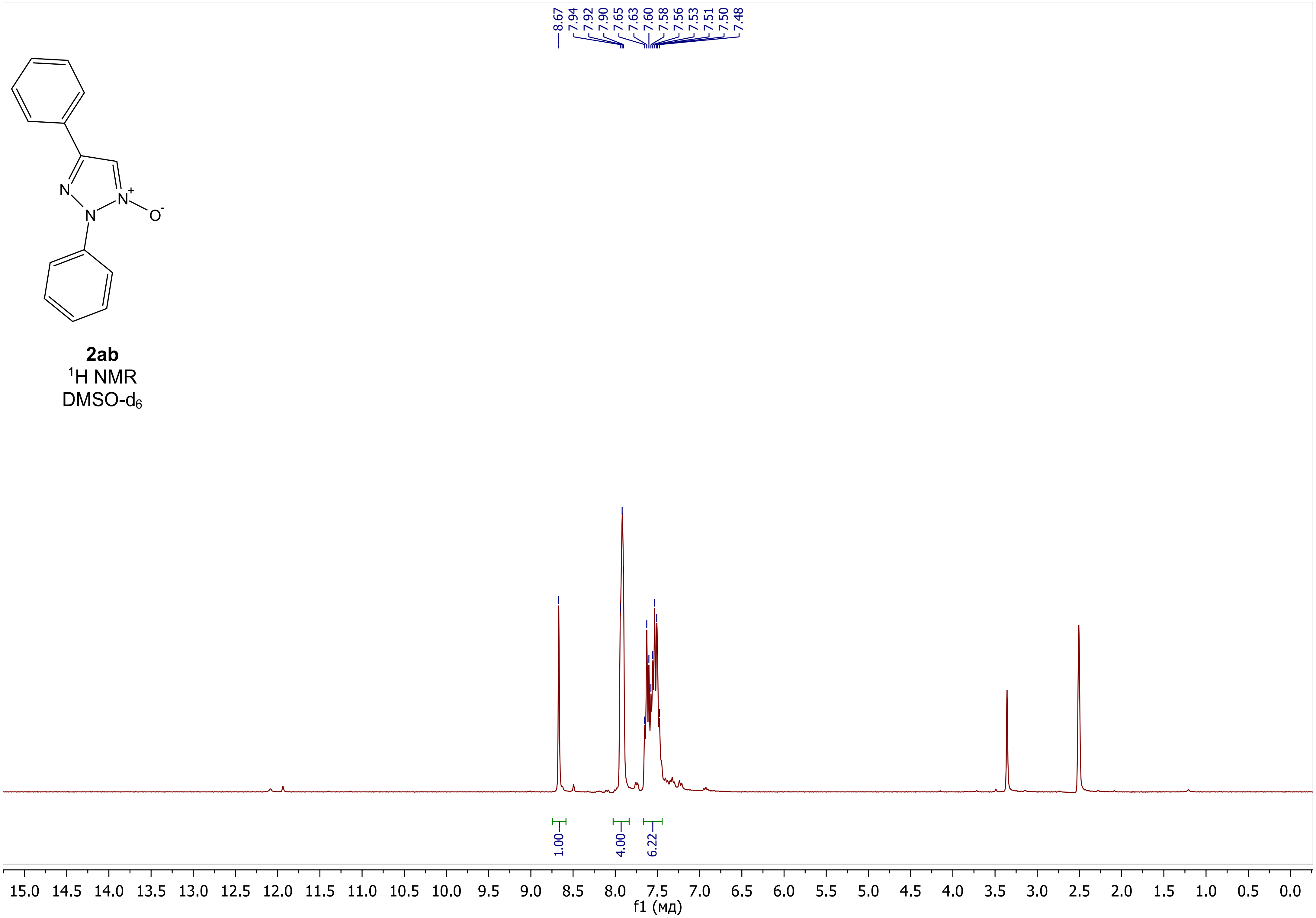


2q
^{13}C NMR
DMSO-d$_6$


H3C
N
N
N+
O-
2aa
1H NMR
DMSO-d6
7.88
7.84
7.84
7.82
7.81
7.60
7.58
7.55
7.52
7.50
7.48
2.28
1.01
2.01
3.19
3.00
15.0 14.5 14.0 13.5 13.0 12.5 12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (мд)



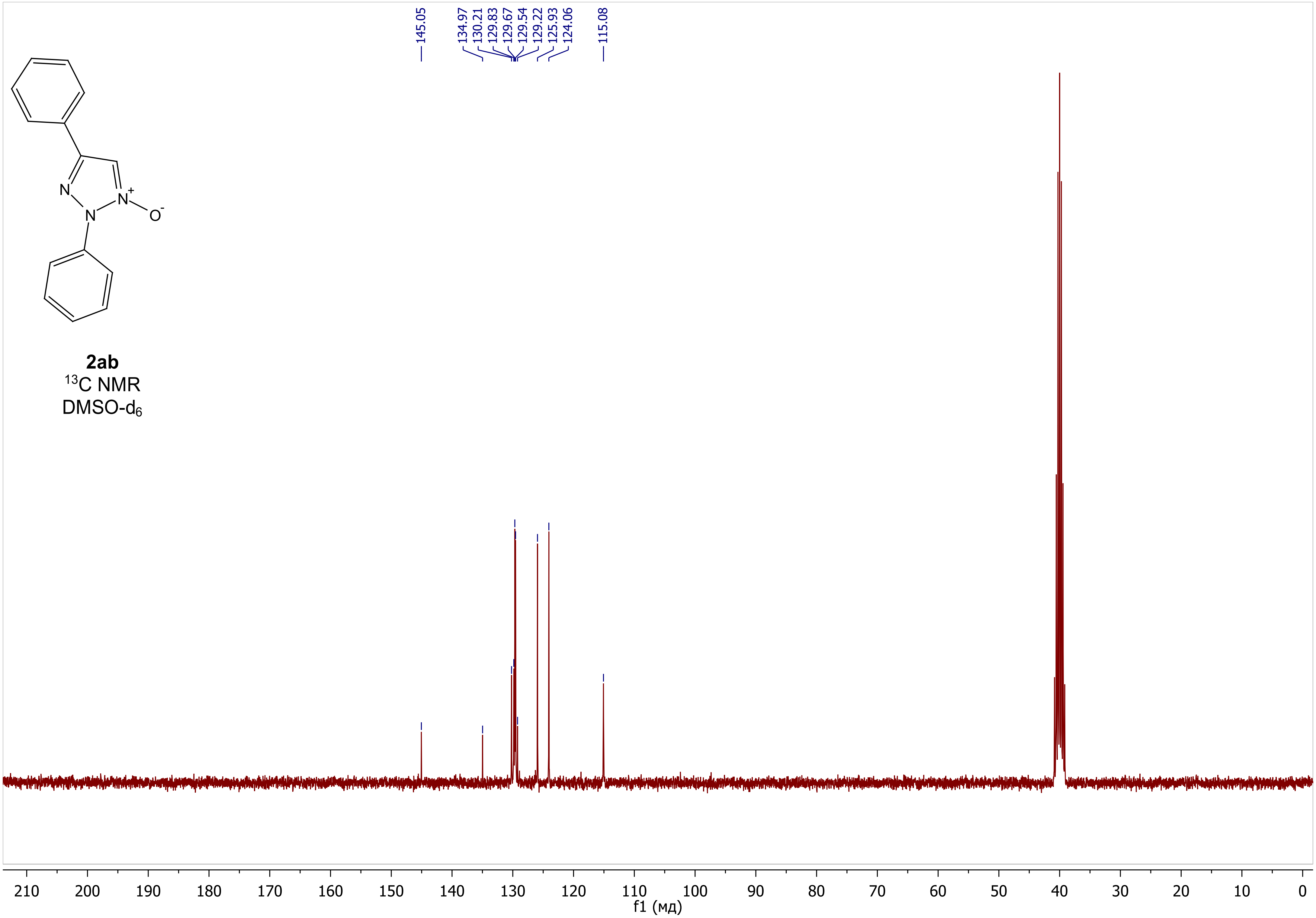
H3C
N
N
N+
O-
2aa
13C NMR
DMSO-d6
143.24
135.05
129.59
129.40
123.60
117.12
12.37
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)



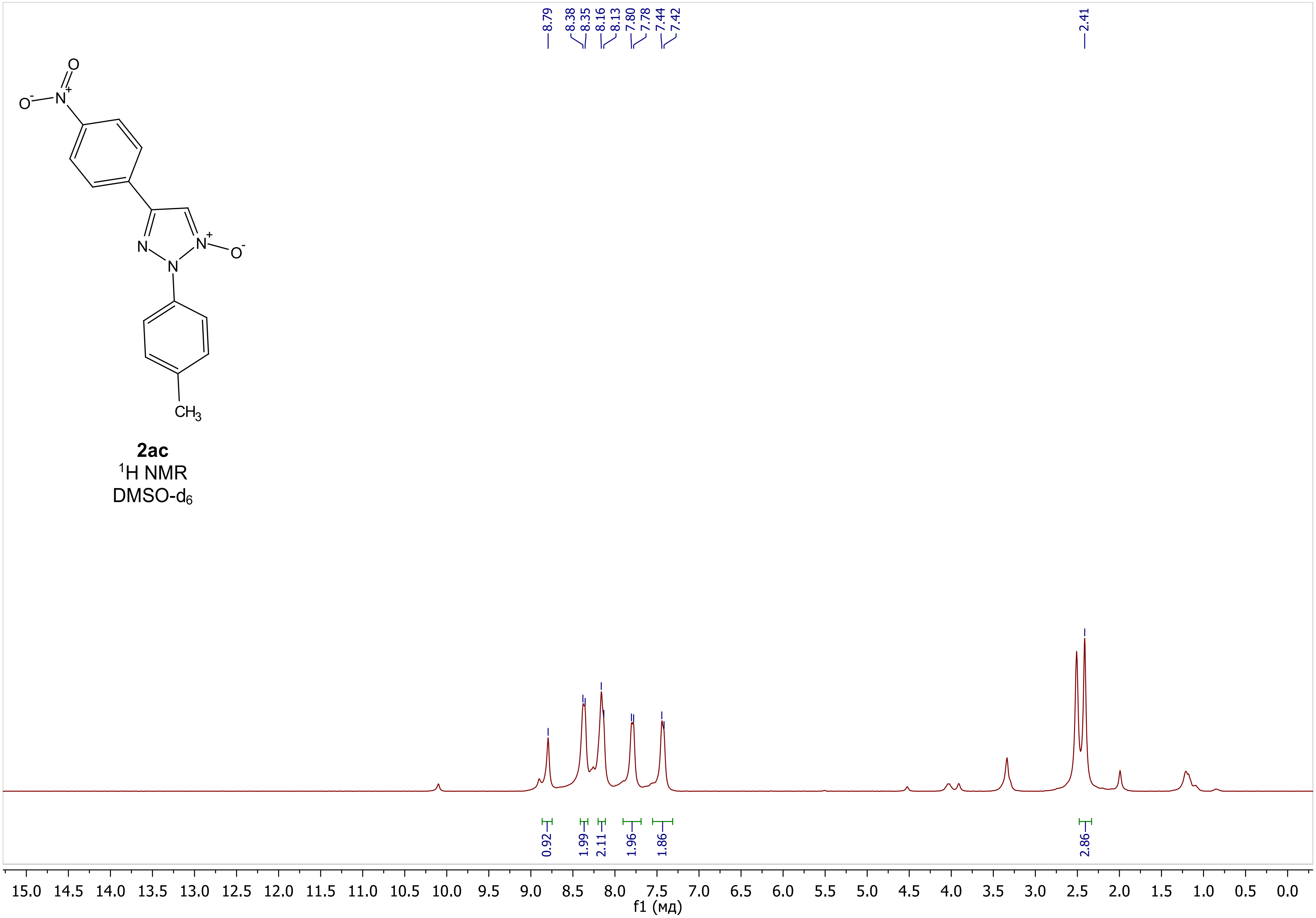
2ab

^{1}H NMR

DMSO-d$_6$



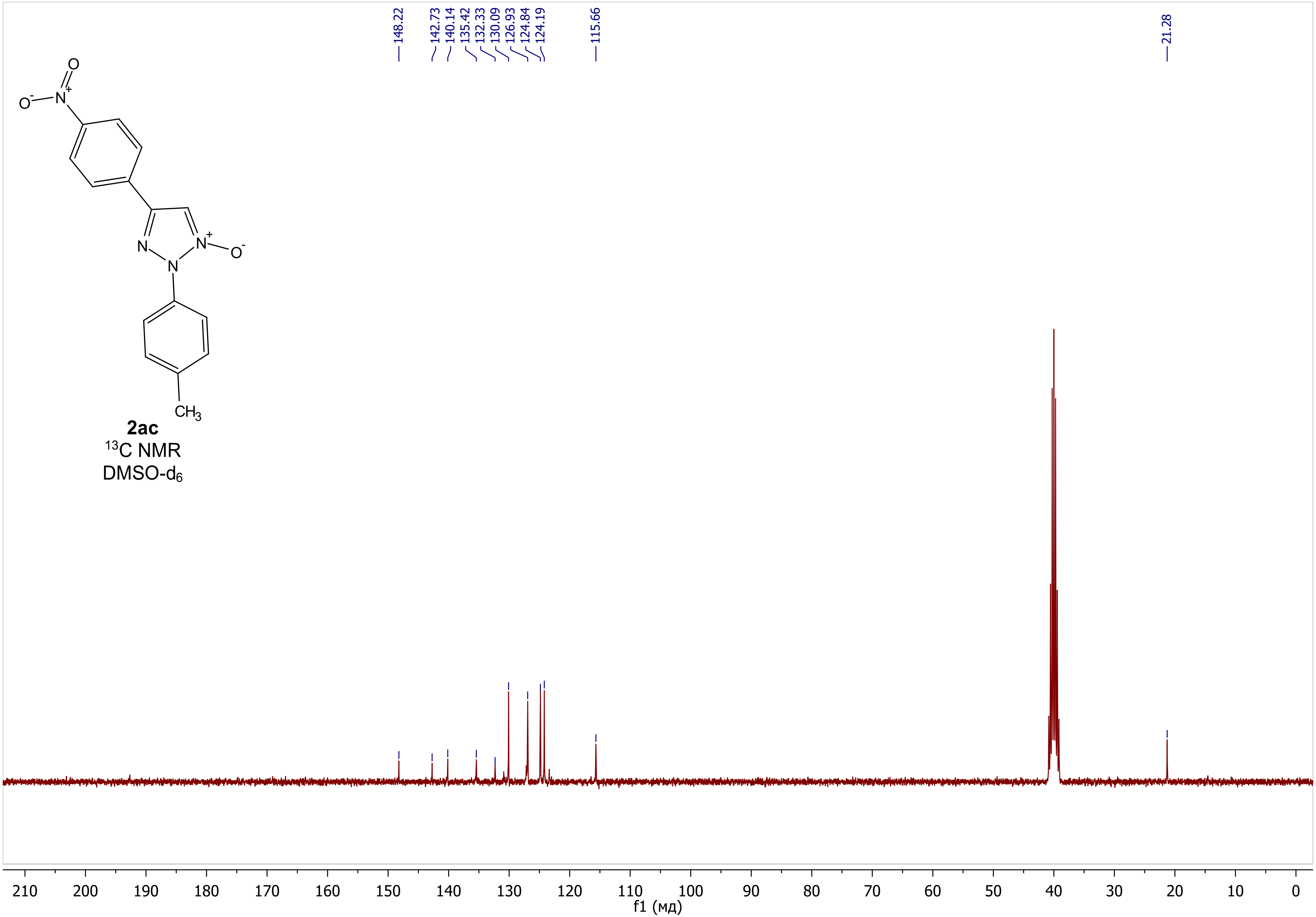
2ab
^{13}C NMR
DMSO-d_6



2ac

^{1}H NMR

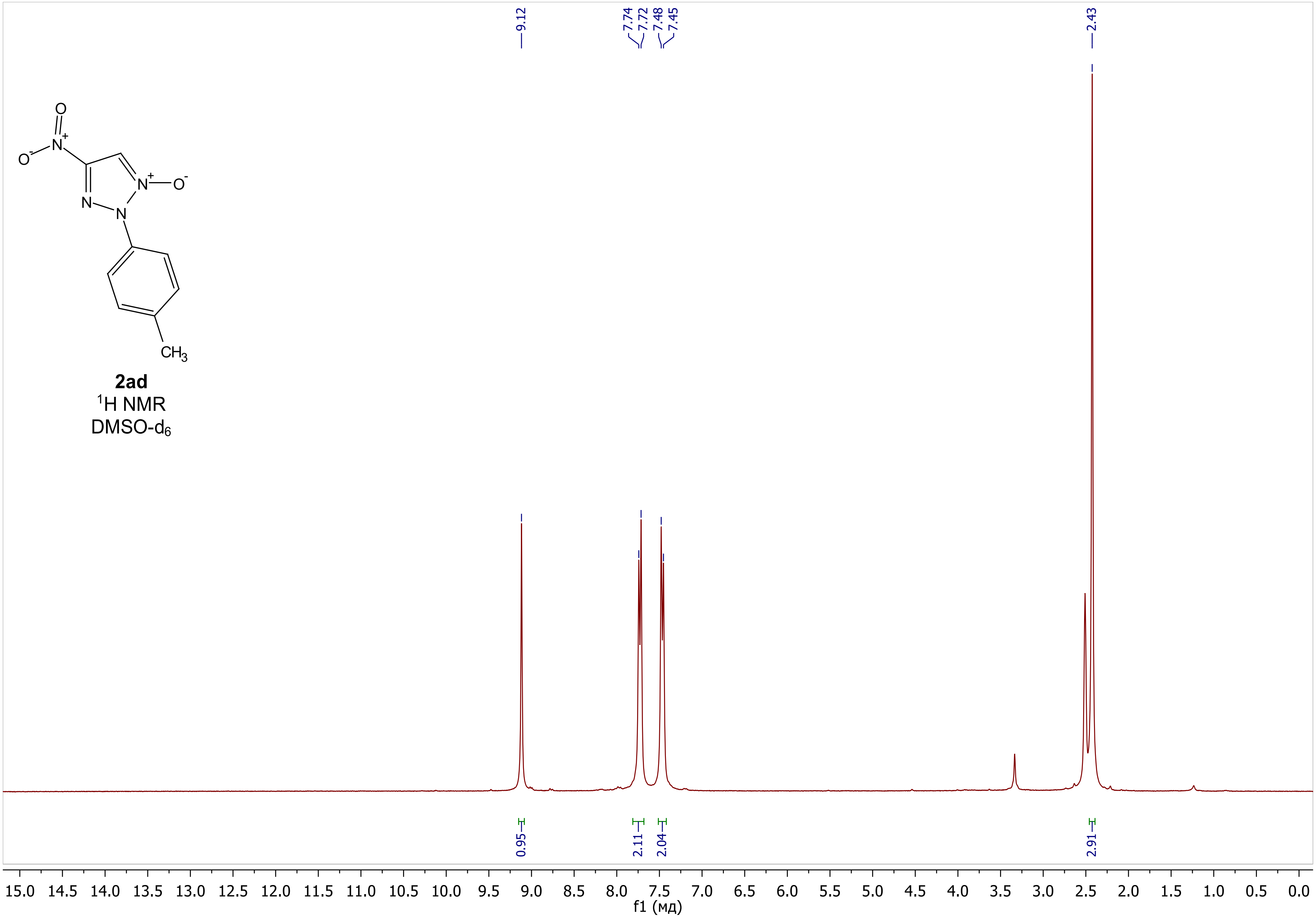
DMSO-d_6



2ac

^{13}C NMR

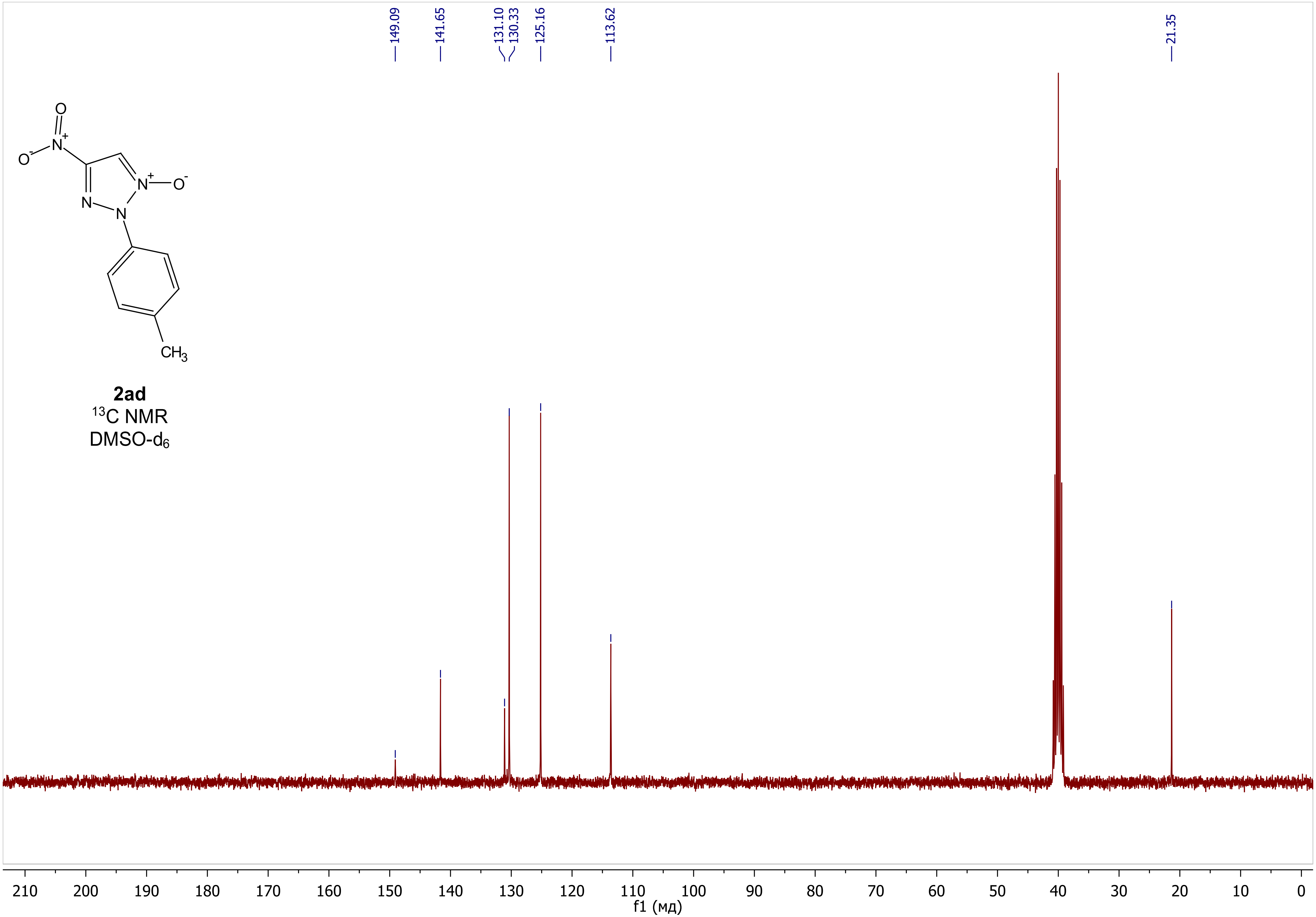
DMSO-d$_6$



2ad

^{1}H NMR

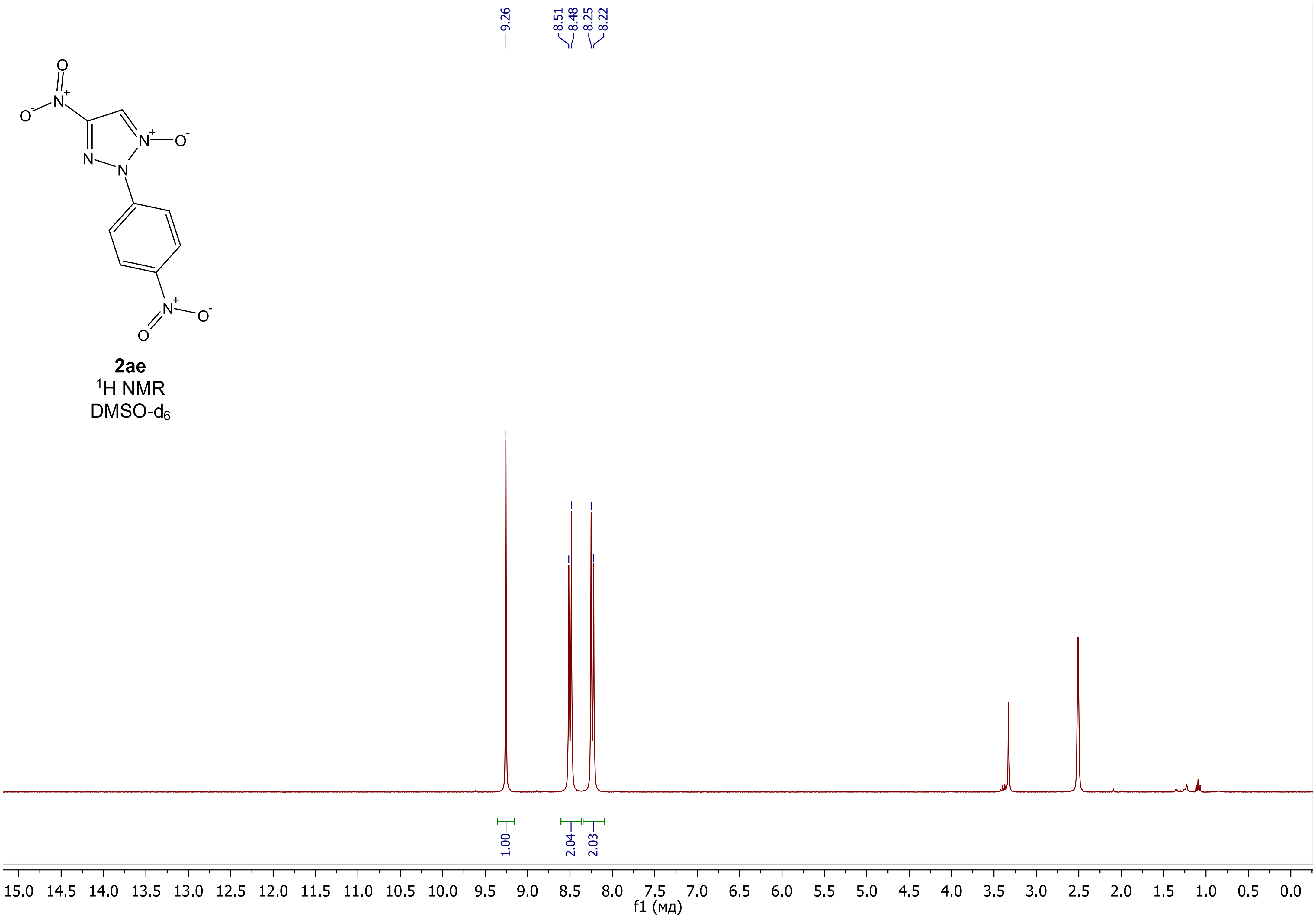
DMSO-d_6

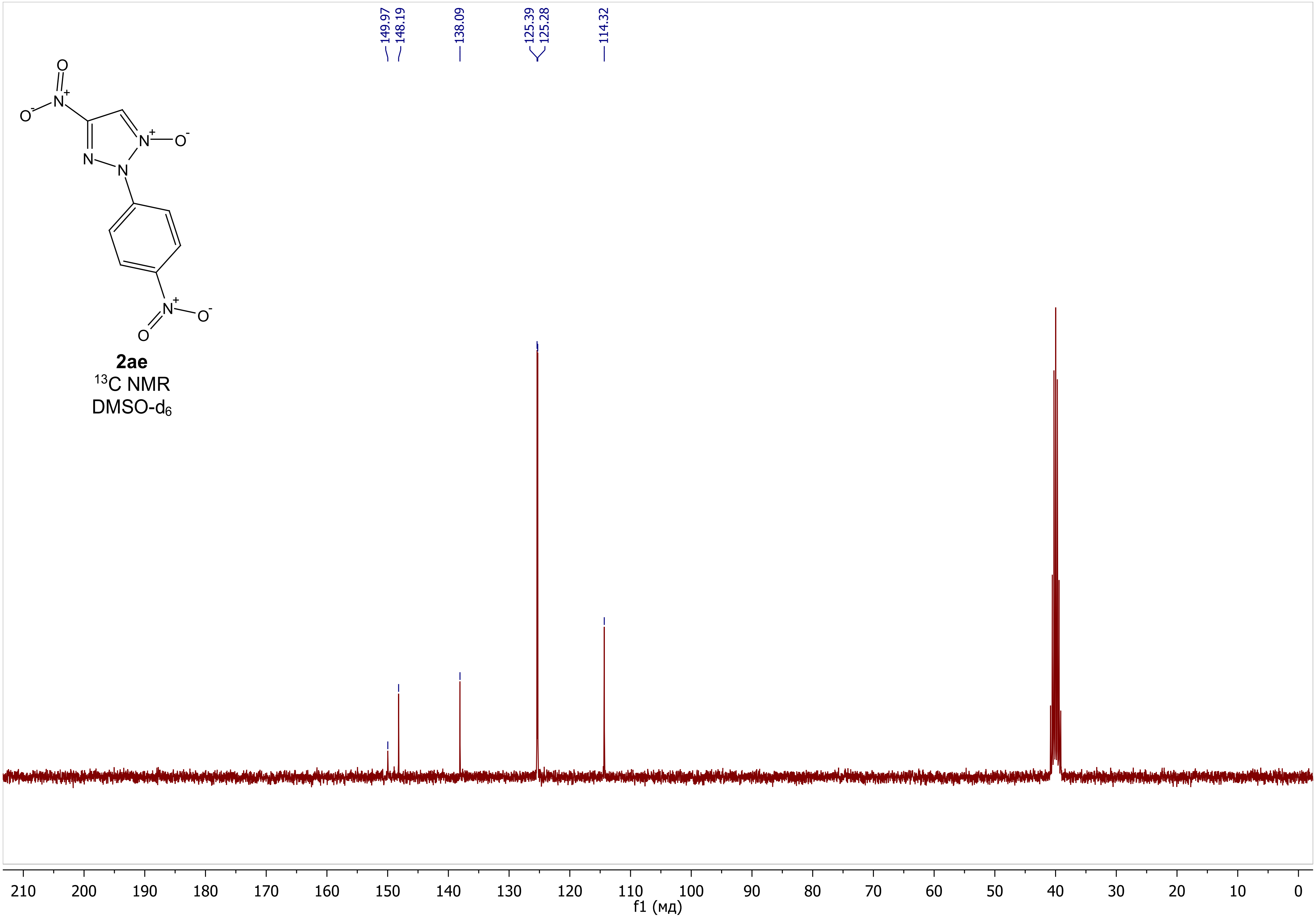


2ad

^{13}C NMR

DMSO-d_6

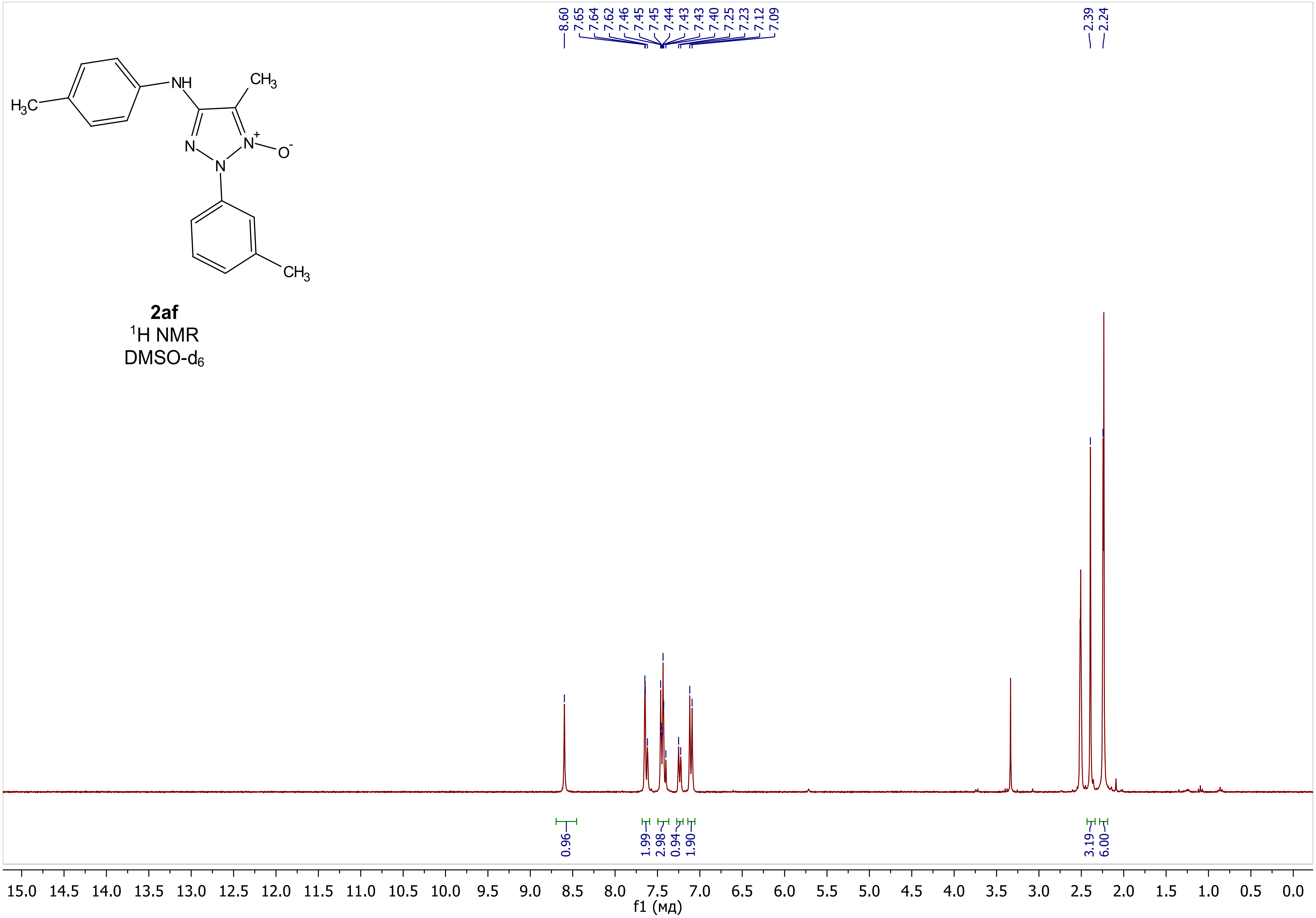

9.26
8.51
8.48
8.25
8.22
2ae
^{1}H NMR
DMSO-d_6
1.00
2.04
2.03
15.0
14.5
14.0
13.5
13.0
12.5
12.0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (мд)



2ae

^{13}C NMR

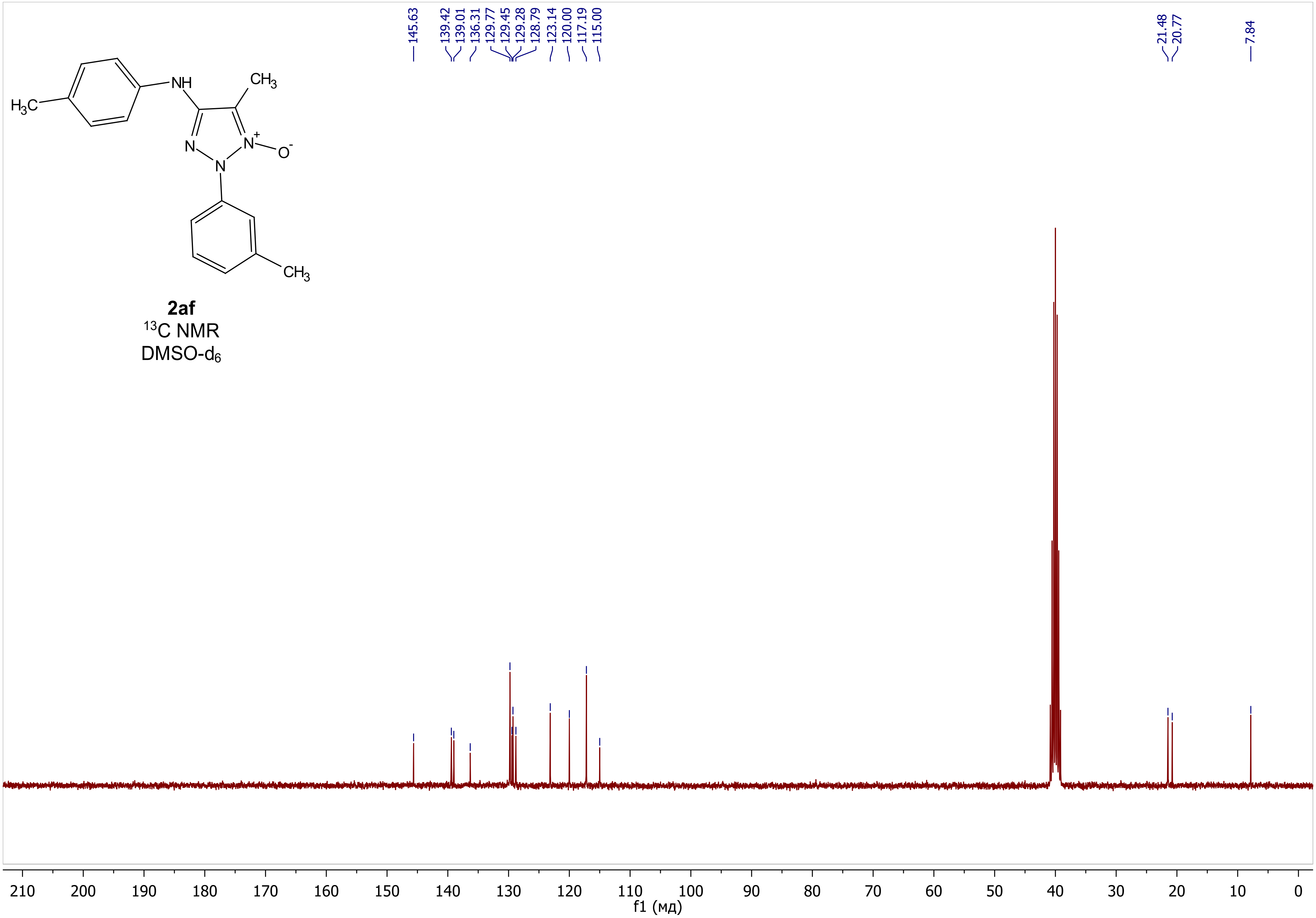
DMSO-d_6



2af

^{1}H NMR

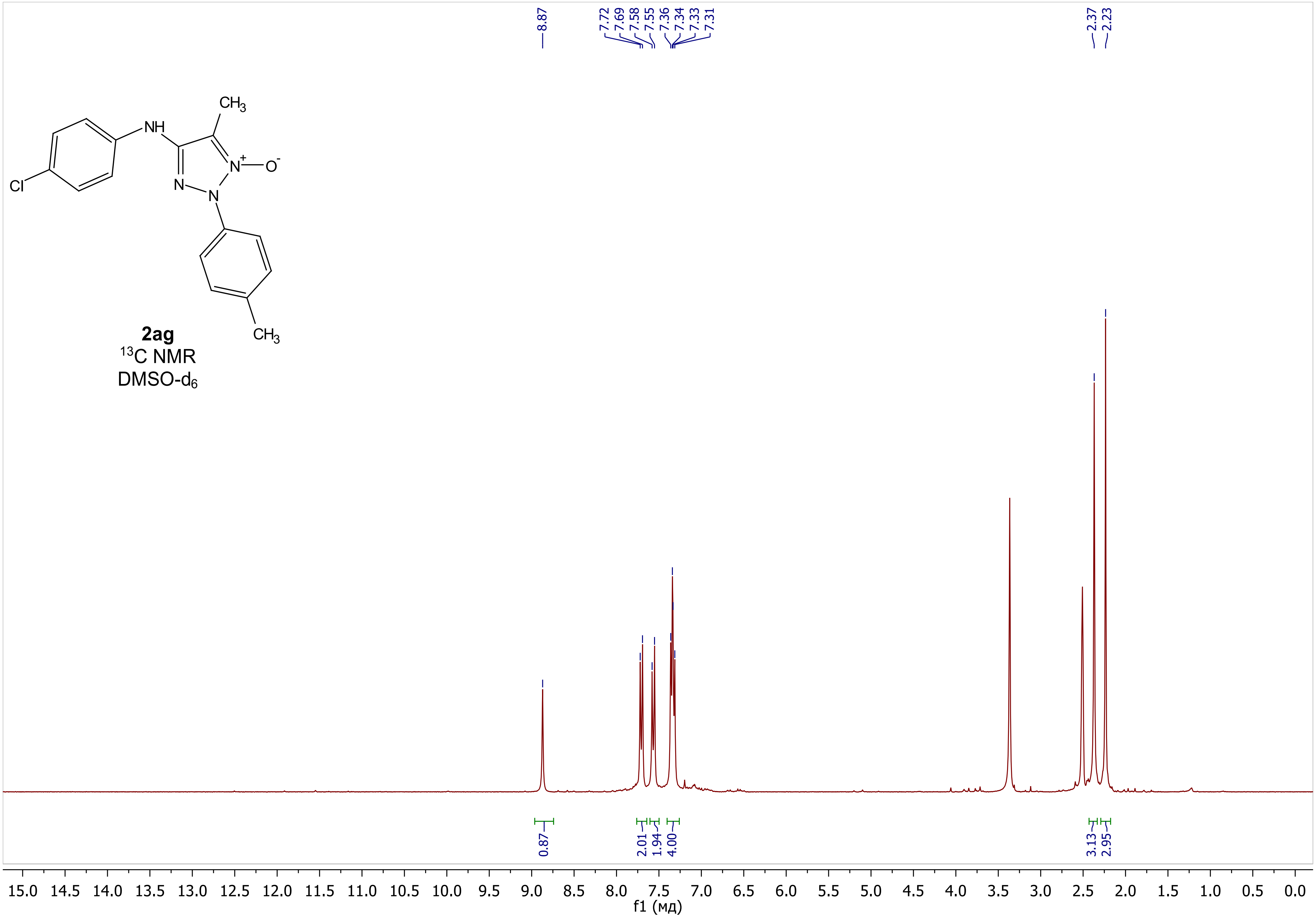
DMSO-d_6



2af

^{13}C NMR

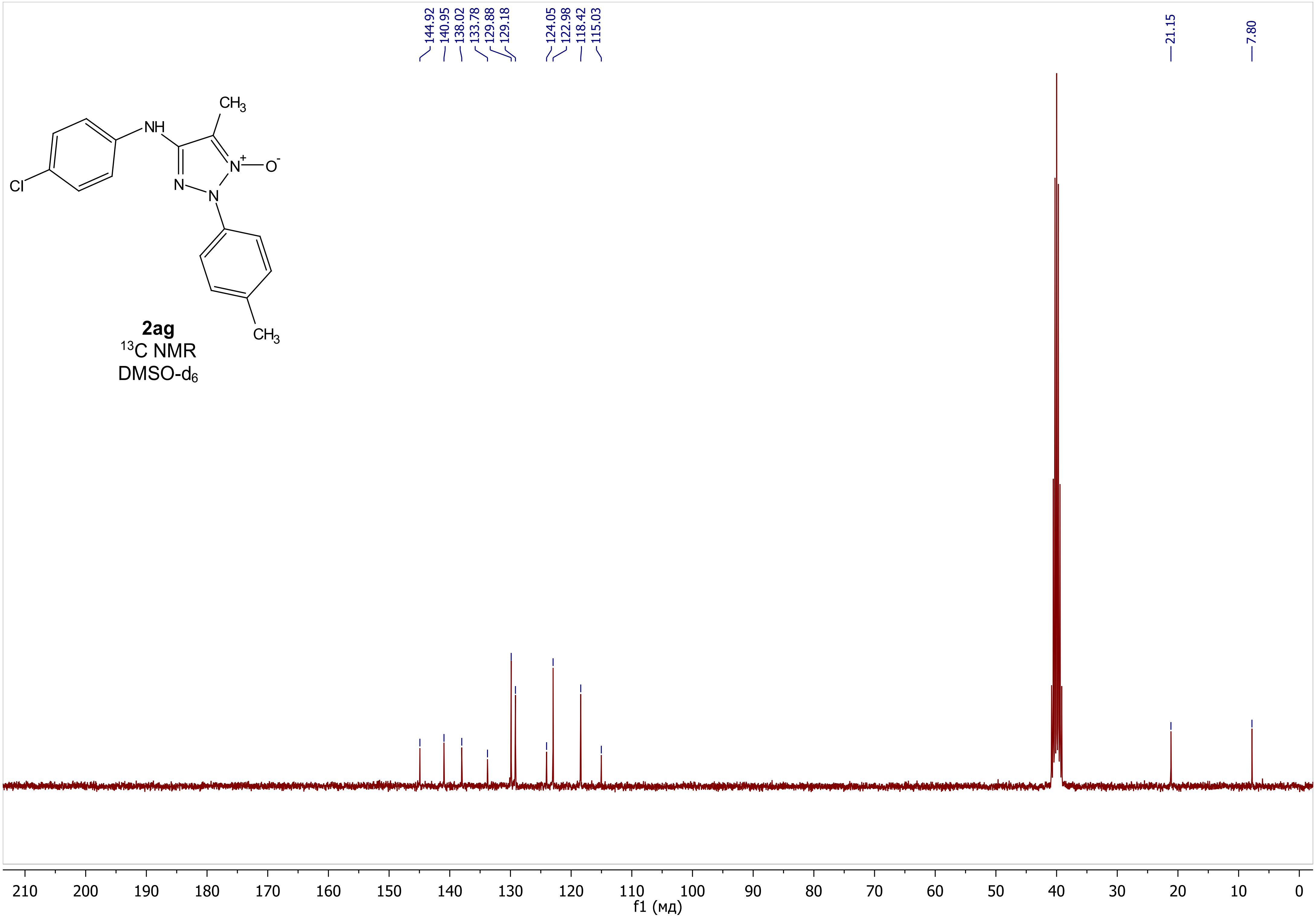
DMSO-d_6



2ag

^{13}C NMR

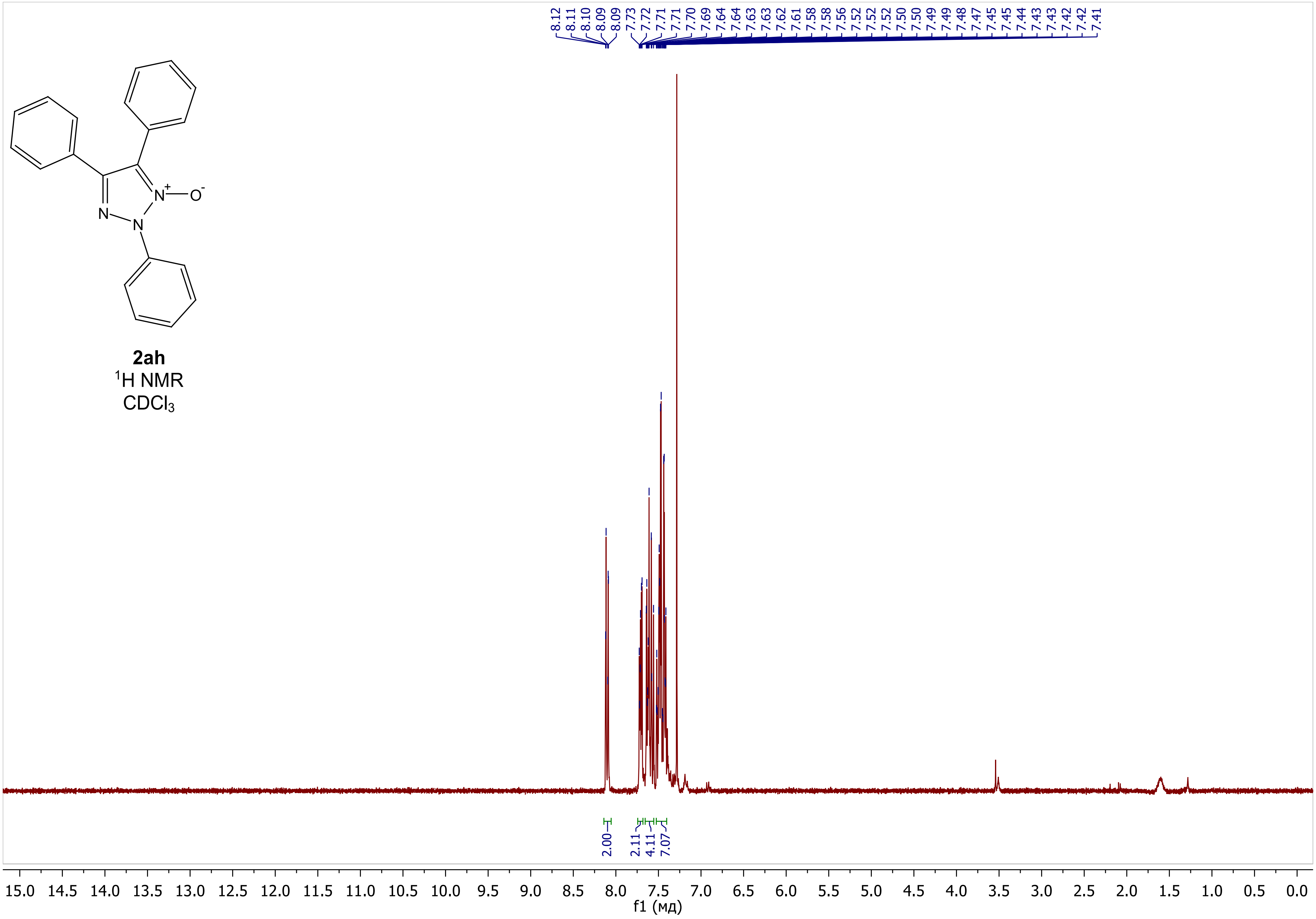
DMSO-d_6

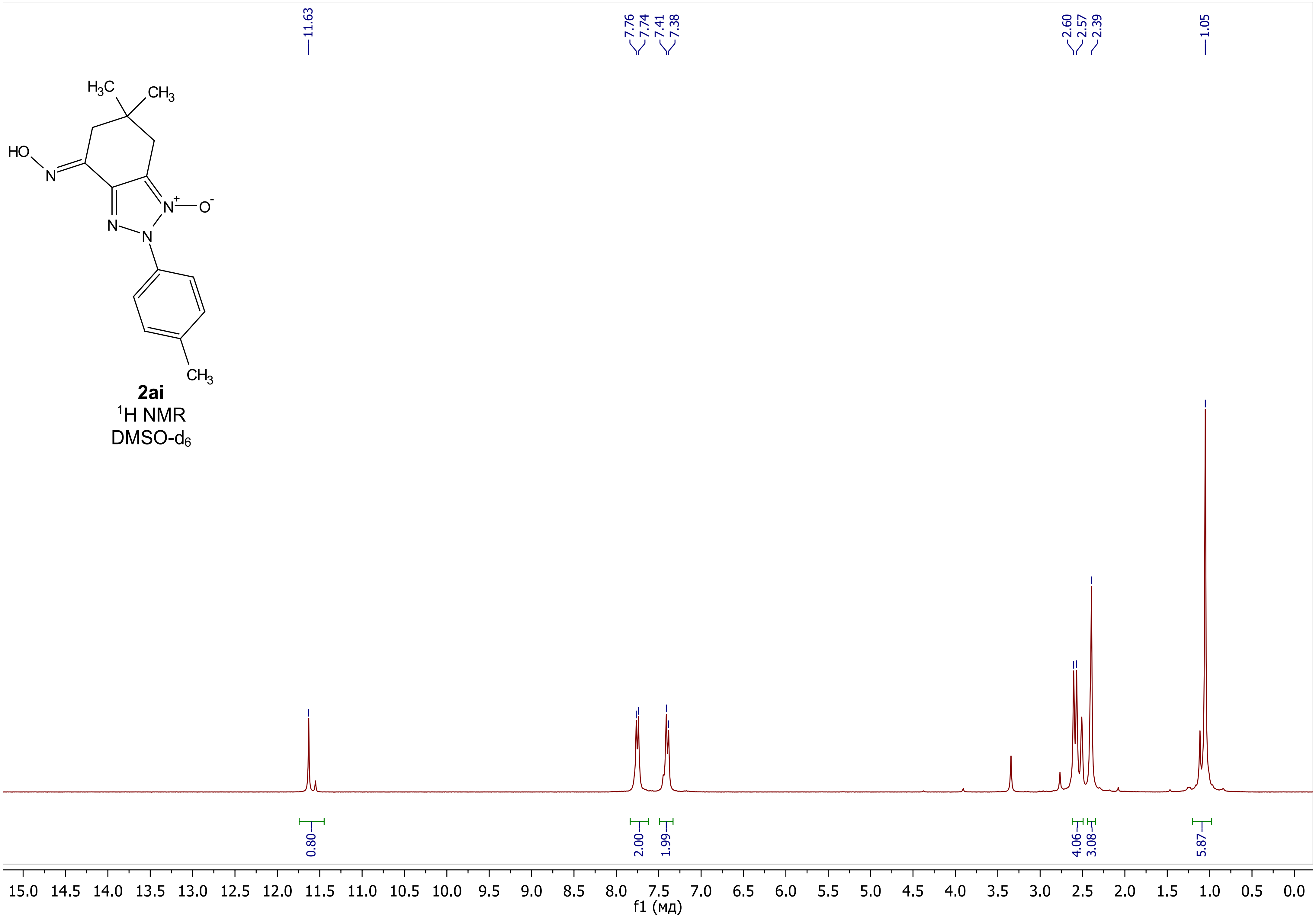


2ag

^{13}C NMR

DMSO-d_6

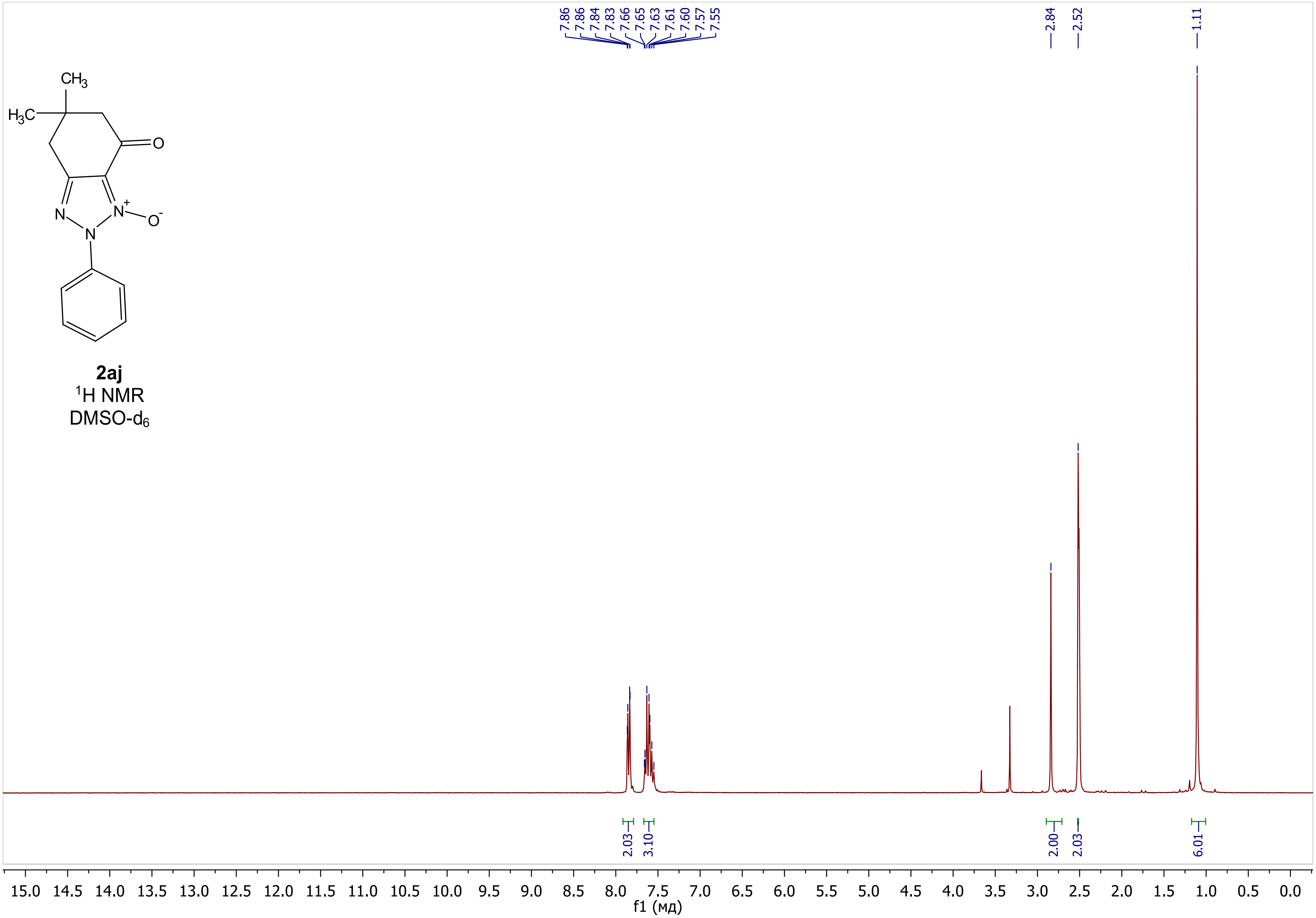

8.12
8.11
8.10
8.09
8.09
7.73
7.72
7.71
7.71
7.70
7.69
7.64
7.64
7.63
7.63
7.62
7.61
7.58
7.58
7.56
7.52
7.52
7.50
7.50
7.49
7.49
7.48
7.47
7.45
7.45
7.44
7.43
7.43
7.42
7.42
7.41
N
N
N+
O-
2ah
1H NMR
CDCl3
2.00
2.11
4.11
7.07
15.0
14.5
14.0
13.5
13.0
12.5
12.0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (мд)



2ai

^{1}H NMR

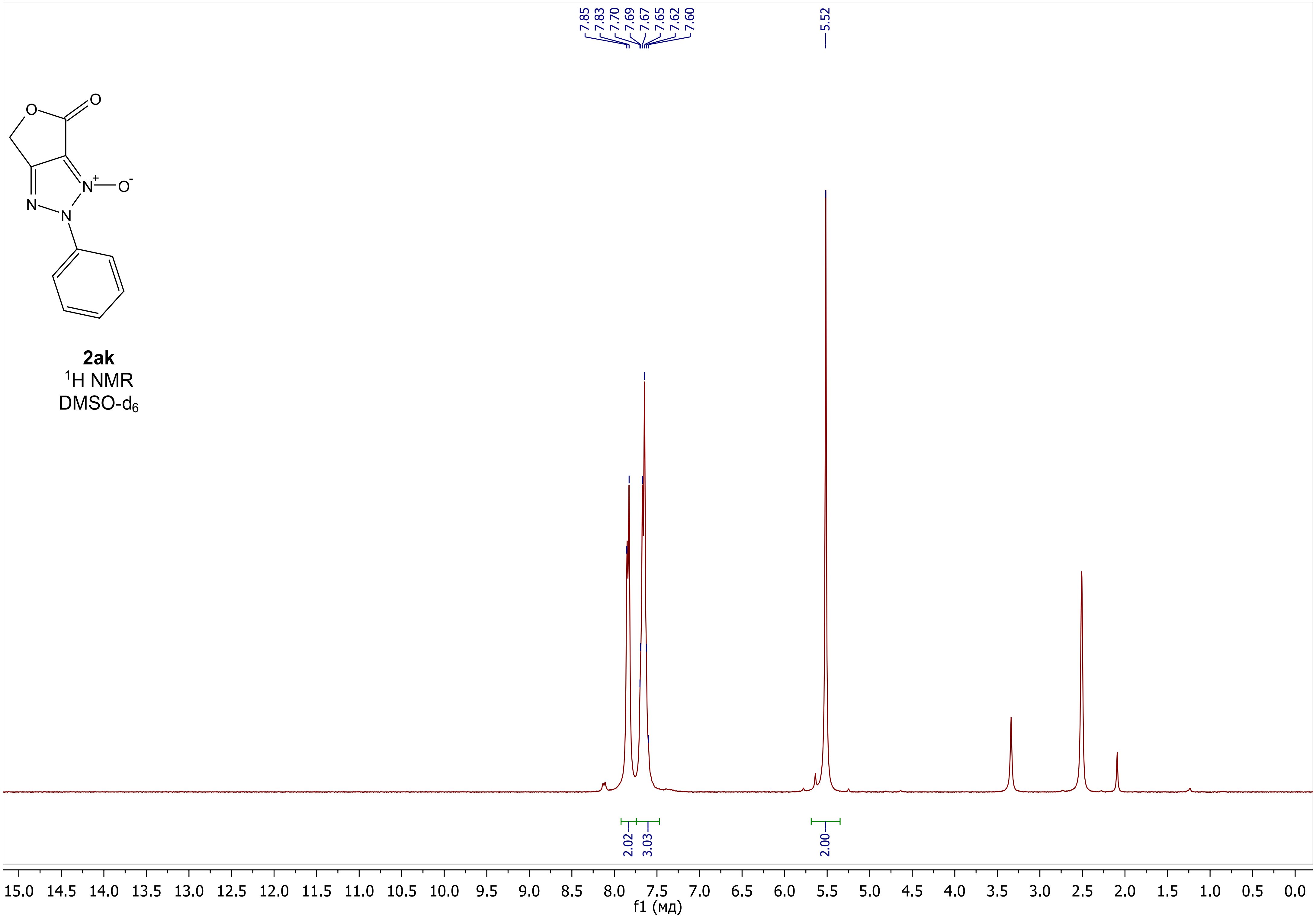
DMSO-d_6

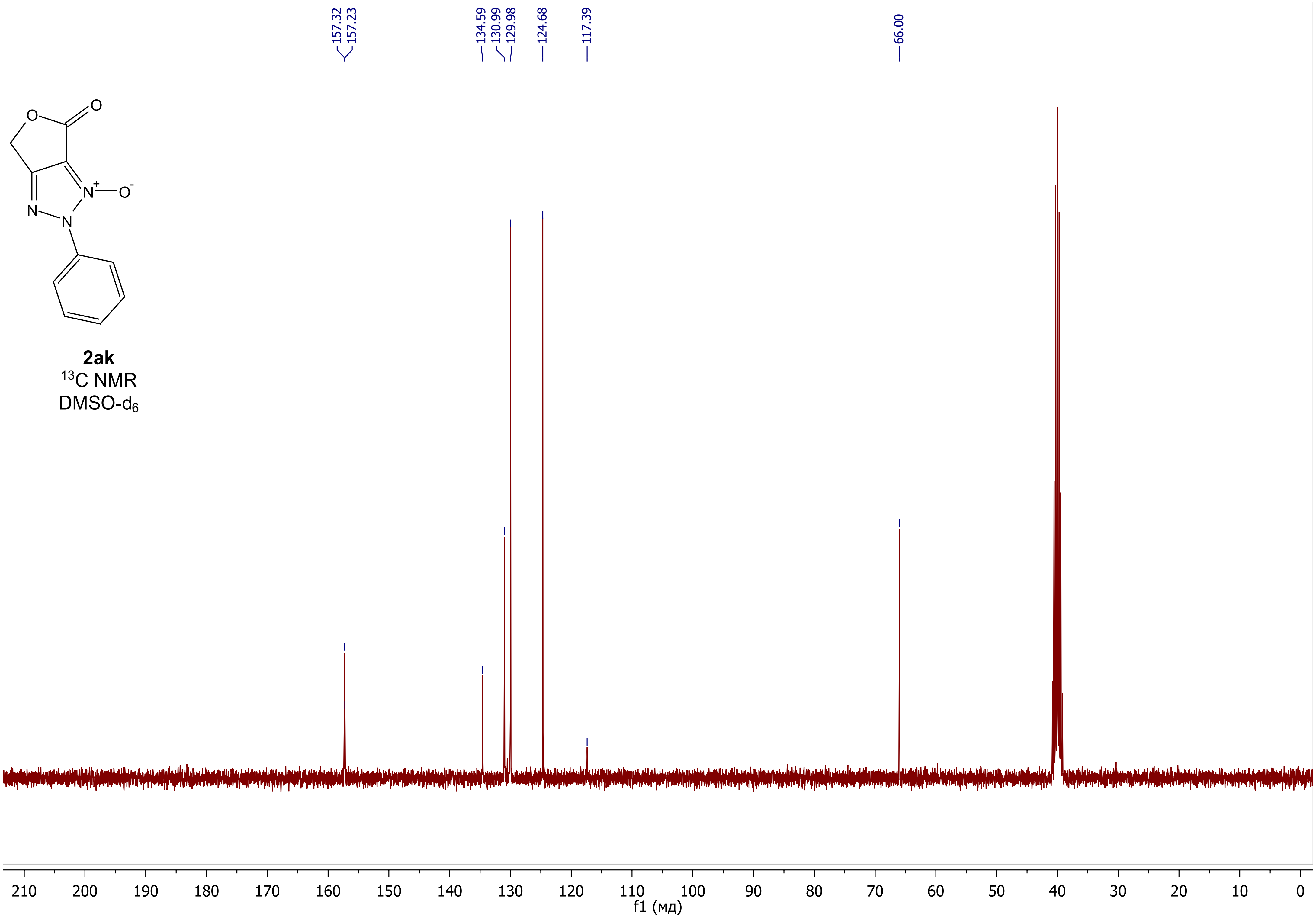


2aj

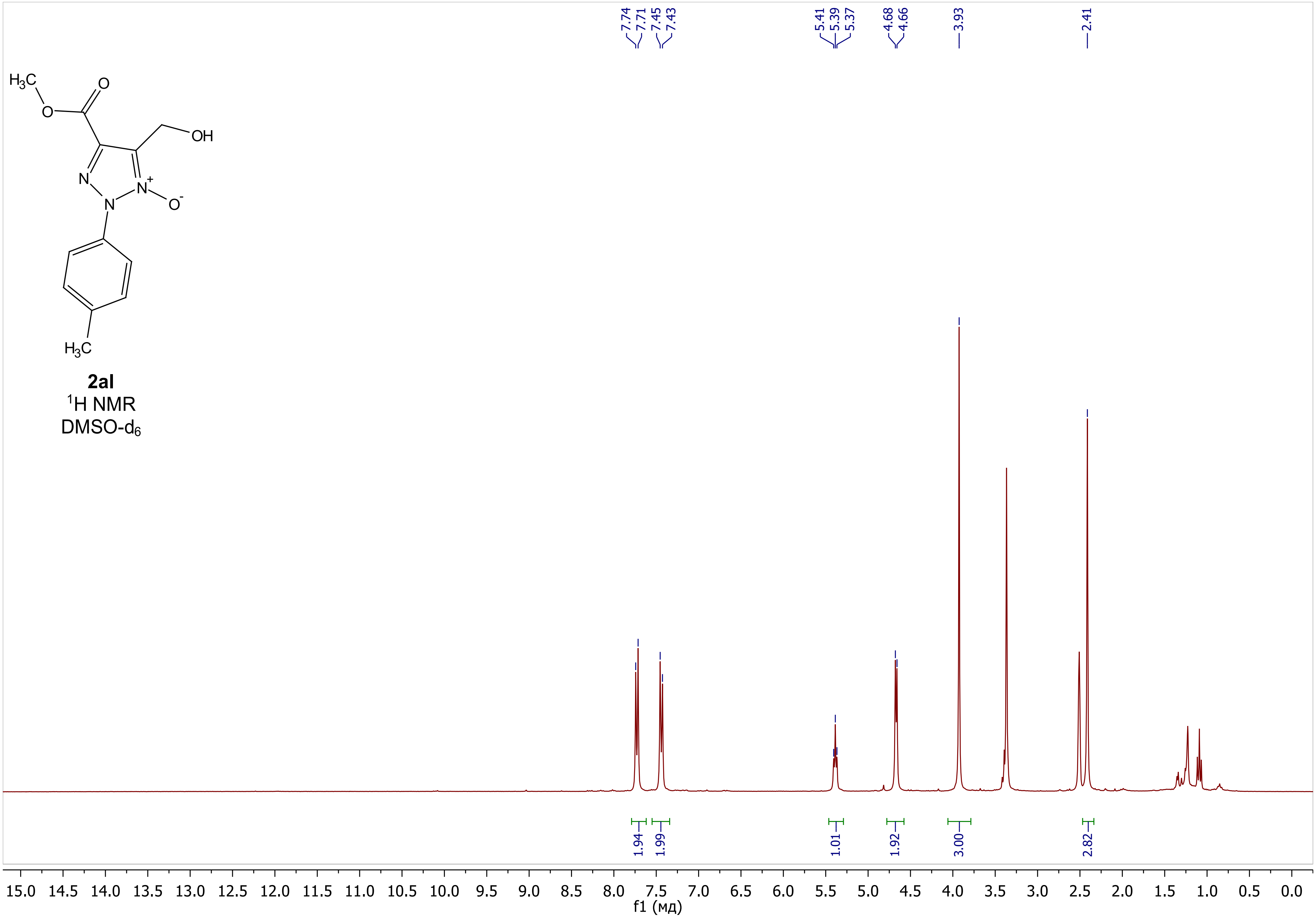
^{1}H NMR

DMSO-d_6


7.85
7.83
7.70
7.69
7.67
7.65
7.62
7.60
5.52
O
O
N
N
N
O
2ak
^{1}H NMR
DMSO-d_6
2.02
3.03
2.00
15.0
14.5
14.0
13.5
13.0
12.5
12.0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (мд)



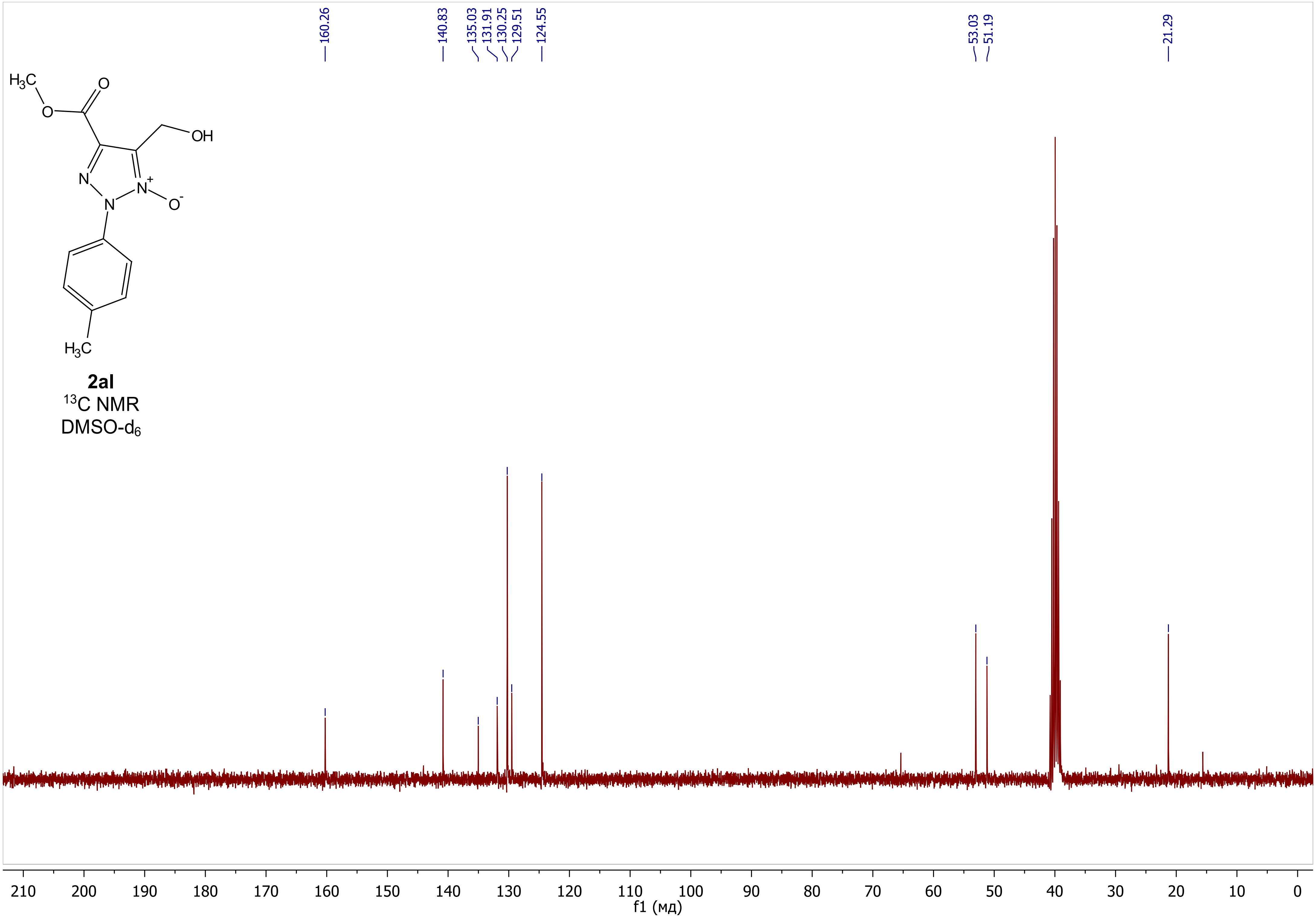
2ak
^{13}C NMR
DMSO-d_6



2al

^{1}H NMR

DMSO-d_6


160.26
140.83
135.03
131.91
130.25
129.51
124.55
53.03
51.19
21.29
H_3C
O
O
OH
N
N
N^+
O^-
H_3C
2al
^{13}C NMR
DMSO-d_6
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (мд)