

Supporting Information

Synthesis, structure investigation and evaluation of anti-orthopoxvirus activity of 2-(2-arylethenyl)imidazoles

Vladislav O. Golfarb-Abramov,^a Anastasia V. Kazennova,^a Elizaveta I. Basanova,^a Olga A. Serova,^b Nikolai I. Bormotov,^b Larisa N. Shishkina,^b Vyacheslav I. Krasnov,^c Dmitriy N. Polovyanenko,^c Ivan A. Moskalev,^d Alexey Yu. Fedorov,^d Sergey G. Arkhipov,^d Margarita G. Ilyina,^d Sophia S. Borisevich,^d Olga I. Yarovaya,^c and Polina A. Nikitina*^a

^a Department of Fine Organic Synthesis and Chemistry of Dyes, D.I. Mendeleev University of Chemical Technology of Russia, Miusskaya sq., 9, 125047, Moscow, Russia. E-mail: polinandrevna@yandex.ru

^b State Research Center of Virology and Biotechnology VECTOR, Rospotrebnadzor, 630559, Koltsovo, Russia.

^c N.N. Vorozhtsov Institute of Organic Chemistry SB RAS, Lavrentyev Ave., 9, 630090, Novosibirsk, Russia.

^d SRF "SKIF", 630559, Koltsovo, Russia.

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SI 1. Comparison of NMR spectroscopy data

Table S1. ¹H chemical shift values (δ, ppm) and spin-spin coupling constants (*J*, Hz) of the doublets of the ethylene bond protons of the compounds **3a-h**, **4a-h**, **6a-d** in ¹H NMR spectra in DMSO-d₆.

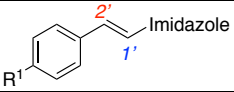
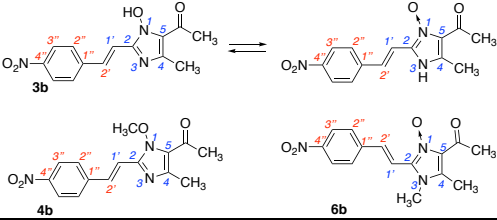
							
No	R ¹	R ²	R ³	H1'		H2'	
				δ, ppm	<i>J</i> , Hz	δ, ppm	<i>J</i> , Hz
3a	NO ₂	CH ₂ C(CH ₃) ₃ CH ₂ CO		7.31	16.2	7.70	16.2
3b	NO ₂	CH ₃	COCH ₃	7.34	16.3	7.68	16.3
3c	NO ₂	CH ₃	COOC ₂ H ₅	7.31	16.3	7.65	16.3
3d	NO ₂	CH ₃	CH ₃	7.22	16.4	7.46	16.4
3e	H	CH ₂ C(CH ₃) ₃ CH ₂ CO		7.09	16.3	7.62	16.3
3f	H	CH ₃	COCH ₃	7.11	16.3	7.59	16.3
3g	H	CH ₃	COOC ₂ H ₅	7.07	16.3	7.55	16.3
3h	H	CH ₃	CH ₃	6.95	16.5	7.32	16.5
4a	NO ₂	CH ₂ C(CH ₃) ₃ CH ₂ CO		7.40	16.2	7.78	16.2
4b	NO ₂	CH ₃	COCH ₃	7.36	16.2	7.73	16.2
4c	NO ₂	CH ₃	COOC ₂ H ₅	7.36	16.2	7.71	16.2
4d	NO ₂	CH ₃	CH ₃	7.27	16.2	7.42	16.2
4e	H	CH ₂ C(CH ₃) ₃ CH ₂ CO		7.16	16.2	7.69	16.2
4f	H	CH ₃	COCH ₃	7.13	16.2	7.64	16.2
4g	H	CH ₃	COOC ₂ H ₅	7.12	16.2	7.62	16.2
4h	H	CH ₃	CH ₃	7.01	16.3	7.32	16.3
6a	NO ₂	CH ₂ C(CH ₃) ₃ CH ₂ CO		7.36	15.8	8.85	15.8
6b	NO ₂	CH ₃	COCH ₃	7.35	15.8	8.88	15.8
6c	NO ₂	CH ₃	CH ₃	7.34	15.7	8.73	15.7
6d	H	CH ₃	CH ₃	7.10	15.8	8.67	15.8

Table S2. ^{13}C NMR chemical shift values (δ , ppm) for 5-acetylsubstituted imidazole derivatives **3b**, **4b**, **6b**.

						
δ_{C} , ppm						
	3b		4b		6b	
	DMSO- d_6	acetone- d_6	DMSO- d_6	acetone- d_6	DMSO- d_6	acetone- d_6
C2 (Im)	141.3	140.3	141.1	142.3	132.0	133.4
C4 (Im)	143.4	145.1	144.0	145.2	133.2	134.0
C5 (Im)	126.0	124.5	124.2	125.5	128.9	130.5
4-CH₃	16.3	16.9	16.8	17.4	10.1	10.4
C=O (5-Ac)	187.3	191.3	186.1	186.7	190.1	191.0
CH ₃ (5-Ac)	29.7	28.9	29.5	29.8	30.4	30.9
C1'	116.3	116.5	115.6	116.7	113.1	113.7
C2'	131.3	133.1	132.9	133.8	128.6	129.8
C1'' (Ph)	142.6	143.6	142.2	143.6	144.1	145.5
C2'' (Ph)	128.0	129.0	128.4	129.1	127.8	128.5
C3'' (Ph)	124.1	125.0	123.9	124.9	123.9	124.9
C4'' (Ph)	146.8	148.5	147.0	148.6	146.5	148.0
OCH ₃ /NCH ₃	--	--	68.2	68.8	30.5	31.0

Note: For the convenience, the numeration of the atoms in N-oxide cycle is the same as in N-methoxy- and N-hydroxy- derivatives.

Table S3. ^{13}C and ^{15}N NMR chemical shift values (δ , ppm) for 4,5-dimethylsubstituted imidazole derivatives **3h**, **4h**, **6d**.

	3h		4h		6d	
	DMSO- d_6	acetone- d_6	DMSO- d_6	acetone- d_6	DMSO- d_6	acetone- d_6
δ_c , ppm						
C2 (Im)	135.5 (br.)	136.7 (br.)	136.9	138.1	131.0	132.5
C4 (Im)	125.6 (br.)	125.6 (br.)	129.8	131.2	121.5	122.1
C5 (Im)	123.0	124.9	120.0	120.7	125.4	126.9
4-CH₃	11.7	11.4	13.0	13.4	8.7	9.0
5-CH ₃	7.3	7.6	7.2	7.6	7.2	7.4
C1'	111.9	112.2	112.9	114.1	109.8	110.3
C2'	128.3	130.7	129.4	130.4	129.1	130.7
C1'' (Ph)	136.5	137.7	136.5	138.0	137.5	139.1
C2'' (Ph)	126.3	127.4	128.0	127.5	126.6	127.5
C3'' (Ph)	128.9	129.7	128.8	129.6	128.8	129.6
C4'' (Ph)	127.9	128.9	126.7	128.8	127.8	128.6
OCH ₃ / NCH ₃	--	--	67.1	67.4	30.2	30.6
δ_N , ppm						
N1		226.7		221.2		261.6
N3		190.7		250.2		132.5

Notes: (br.) – broadened signal. For the convenience, the numeration of the atoms in N-oxide cycle is the same as in N-methoxy- and N-hydroxy- derivatives.

SI 2. Single crystal X-ray analysis

Table S4. Crystallographic characteristics, details of the experiment and structure refinement for compounds **3a,c**.

Parameter	3a	3c
Chemical formula	C ₁₇ H ₁₇ N ₃ O ₄ ·C ₃ H ₇ NO	C ₁₅ H ₁₅ N ₃ O ₅ ·C ₁₅ H ₁₄ N ₃ O ₅
Mr	400.43	633.59
Crystal system, space group	Monoclinic, P21/c	Triclinic, P-1
Temperature (K)	293(2)	293(2)
a, b, c (Å)	7.4075 (5), 13.1203 (11), 21.0062 (15)	7.3738 (5), 14.1463 (10), 15.4463 (10)
α, β, γ (°)	90, 92.715 (6), 90	66.399 (6), 88.379 (5), 86.836 (5)
V (Å ³)	2039.3 (3)	1474.18 (18)
Z	4	2
Radiation type	Mo Kα	Mo Kα
μ (mm ⁻¹)	0.10	0.11
Crystal size (mm)	0.48 × 0.4 × 0.08 × 0.40	0.96 × 0.17 × 0.06
Data collection		
Diffractometer	TD-5000	TD-5000
Absorption correction	For a cylinder mounted on the φ axis CrysAlis PRO 1.171.43.143a (Rigaku Oxford Diffraction, 2024) Empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.	Multi-scan CrysAlis PRO 1.171.42.49 (Rigaku Oxford Diffraction, 2022) Empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.
T _{min} , T _{max}	0.630, 1.000	0.697, 1.000
No. of measured, independent and observed [I > 2σ(I)] reflections	12599, 5367, 3387	29468, 9515, 4312
R _{int}	0.082	0.107
(sin θ/λ) _{max} (Å ⁻¹)	0.694	0.781
Refinement		
R[F ² > 2σ(F ²)], wR(F ²), S	0.059, 0.187, 1.04	0.088, 0.294, 0.98
No. of reflections	5367	9515
No. of parameters	274	420
No. of restraints	7	6
H-atom treatment	H atoms treated by a mixture of independent and constrained refinement	H-atom parameters constrained
Δρ _{max} , Δρ _{min} (e Å ⁻³)	0.17, -0.26	0.36, -0.27
CCDC deposition number	2469646	2469647

Table S5. Crystallographic characteristics, details of the experiment and structure refinement for compounds **4c,f**.

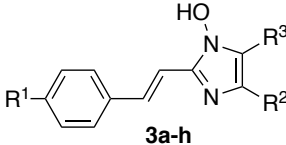
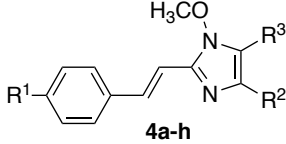
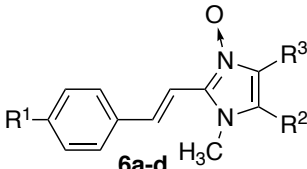
Parameter	4c	4f
Chemical formula	C ₁₆ H ₁₈ N ₃ O ₅	C ₁₅ H ₁₆ N ₂ O ₂
Mr	332.33	256.30
Crystal system, space group	Monoclinic, P21/c	Triclinic, P-1
Temperature (K)	293(2)	293(2)
a, b, c (Å)	4.7308 (9), 38.246 (3), 9.089 (2)	7.5492 (8), 7.7057 (9), 13.4133 (14)
α, β, γ (°)	90, 96.796 (17), 90	96.792 (9), 99.738 (9), 114.016 (11)
V (Å ³)	1632.9 (5)	687.12 (14)
Z	4	4
Radiation type	Mo Kα	Mo Kα
μ (mm ⁻¹)	0.10	0.08
Crystal size (mm)	0.84 × 0.09 × 0.06	0.9 × 0.64 × 0.04
Data collection		
Diffractometer	TD-5000	TD-5000
Absorption correction	For a sphere CrysAlis PRO 1.171.43.143a (Rigaku Oxford Diffraction, 2024) Spherical absorption correction using equivalent radius and absorption coefficient. Empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.	Multi-scan CrysAlis PRO 1.171.42.49 (Rigaku Oxford Diffraction, 2022) Empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.
T _{min} , T _{max}	0.937, 0.937	0.413, 1.000
No. of measured, independent and observed [I > 2σ(I)] reflections	11371, 2799, 1581	7588, 2317, 1846
R _{int}	0.117	0.113
(sin θ/λ) _{max} (Å ⁻¹)	0.595	0.595
Refinement		
R[F ² > 2σ(F ²)], wR(F ²), S	0.080, 0.274, 1.01	0.077, 0.224, 1.03
No. of reflections	2799	2317
No. of parameters	220	175
No. of restraints	2	-
H-atom treatment	H-atom parameters constrained	H-atom parameters constrained
Δρ _{max} , Δρ _{min} (e Å ⁻³)	0.37, -0.28	0.23, -0.22
CCDC deposition number	2469648	2469649

Table S6. Crystallographic characteristics, details of the experiment and structure refinement for compounds **6b,d**.

Parameter	6d	6b
Chemical formula	C ₁₄ H ₁₆ N ₂ O·H ₂ O	C ₁₄ H ₁₅ N ₃ O ₃ ·H ₂ O
Mr	246.30	291.30
Crystal system, space group	Monoclinic, C2/c	Monoclinic, P21/n
Temperature (K)	293(2)	293(2)
a, b, c (Å)	13.398 (2), 9.5481 (13), 20.887 (4)	12.9411 (8), 8.0804 (6), 14.2750 (11)
α, β, γ (°)	90, 97.339 (17), 90	90, 105.129 (8), 90
V (Å ³)	2650.1 (8)	1440.99 (19)
Z	8	4
Radiation type	Mo Kα	Mo Kα
μ (mm ⁻¹)	0.08	0.10
Crystal size (mm)	0.5 × 0.48 × 0.08 × 0.40 (radius)	0.7 × 0.12 × 0.12 × 0.40 (radius)
Data collection		
Diffractometer	TD-5000	TD-5000
Absorption correction	For a sphere CrysAlis PRO 1.171.43.143a (Rigaku Oxford Diffraction, 2024) Spherical absorption correction using equivalent radius and absorption coefficient. Empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.	For a sphere CrysAlis PRO 1.171.43.143a (Rigaku Oxford Diffraction, 2024) Spherical absorption correction using equivalent radius and absorption coefficient. Empirical absorption correction using spherical harmonics, implemented in SCALE3 ABSPACK scaling algorithm.
T _{min} , T _{max}	0.993, 0.993	0.940, 0.940
No. of measured, independent and observed [I > 2σ(I)] reflections	8198, 2291, 1491	10865, 2482, 1968
R _{int}	0.071	0.072
(sin θ/λ) _{max} (Å ⁻¹)	0.595	0.595
Refinement		
R[F ² > 2σ(F ²)], wR(F ²), S	0.093, 0.294, 1.10	0.050, 0.149, 1.07
No. of reflections	2291	2482
No. of parameters	139	223
No. of restraints	1	-
H-atom treatment	H-atom parameters constrained	H atoms treated by a mixture of independent and constrained refinement
Δρ _{max} , Δρ _{min} (e Å ⁻³)	0.65, -0.23	0.20, -0.16
CCDC deposition number	2469650	2469651

SI 3.1. Evaluation of anti-orthopoxvirus activity

Table S7. Cytotoxicity and antiviral activity of compounds **3a-h**, **4a-h**, **6a-d** against Vaccinia virus (VACV, Copenhagen strain) in Vero cell culture.

No	R ¹	R ²	R ³	CC ₅₀ , μg/mL	IC ₅₀ , μg/mL	SI
 3a-h						
3a	NO ₂		CH ₂ C(CH ₃) ₃ CH ₂ CO	8.8	2.1	4*
3b	NO ₂	CH ₃	COCH ₃	6.3	2.4	3*
3c	NO ₂	CH ₃	COOC ₂ H ₅	4.2	N/A	--
3d	NO ₂	CH ₃	CH ₃	4.6	N/A	--
3e	H		CH ₂ C(CH ₃) ₃ CH ₂ CO	47.5±13.1	3.27±1.96	15
3f	H	CH ₃	COCH ₃	19.4±4.7	1.16±0.71	17
3g	H	CH ₃	COOC ₂ H ₅	23.3±5.6	2.85±1.70	8
3h	H	CH ₃	CH ₃	0.85	N/A	--
 4a-h						
4a	NO ₂		CH ₂ C(CH ₃) ₃ CH ₂ CO	>100	N/A	--
4b	NO ₂	CH ₃	COCH ₃	238.7±58.4	0.77±0.40	305
4c	NO ₂	CH ₃	COOC ₂ H ₅	181.0±42.2	0.95±0.22	188
4d	NO ₂	CH ₃	CH ₃	115.7±9.45	0.57±0.16	202
4e	H		CH ₂ C(CH ₃) ₃ CH ₂ CO	>100	N/A	--
4f	H	CH ₃	COCH ₃	27.47±6.8	0.67±0.32	41
4g	H	CH ₃	COOC ₂ H ₅	12.96±0.25	0.17±0.03	76
4h	H	CH ₃	CH ₃	0.80	N/A	--
 6a-d						
6a	NO ₂		CH ₂ C(CH ₃) ₃ CH ₂ CO	>100	32.437	>3*
6b	NO ₂	CH ₃	COCH ₃	>100	33.4	3*
6c	NO ₂	CH ₃	CH ₃	2.393	N/A	--
6d	H	CH ₃	CH ₃	1.248	0.41	3*
Reference substances						
Cidofovir				290.0±65.6	8.97±1.99	32
NIOCH-14				455.2±82.4	0.003±0.002	151882

Note: CC₅₀ – 50% cytotoxicity concentration at which 50% of cells in uninfected monolayers are destroyed; IC₅₀ – 50% virus inhibiting concentration at which 50% of cells in infected monolayers are preserved; for certain compounds expanded testing was performed in 3 repeats; M±SD; M – mean value; SD – standard deviation; n=3 – the number of repeats of measurement of CC₅₀ and IC₅₀; SI – selectivity index of the compound, ratio CC₅₀/IC₅₀. * - chemical compounds with SI<8 are considered as non-perspective for further investigation as antiviral chemicals. N/A – not active.

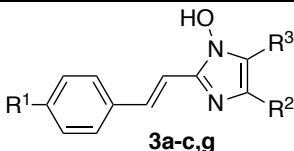
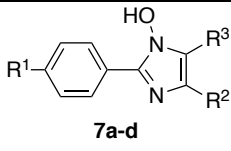
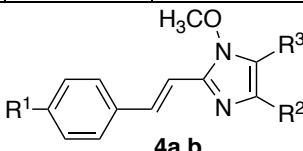
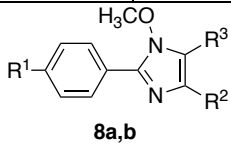
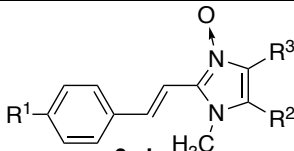
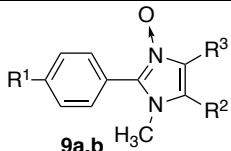
Table S8. Cytotoxicity and antiviral activity of 2-(2-arylethenyl)-1-methoxyimidazoles **4b-d,f,g** against Cowpox virus (CPXV, Grishak strain) and Ectromelia virus (ECTV, K-1 strain) in Vero cell culture.

Nº	CC ₅₀ , µg/mL	IC ₅₀ (CPXV), µg/mL	SI (CPXV)	IC ₅₀ (ECTV), µg/mL	SI(ECTV)
4b	238.7±58.4	6.5±0.5	37	3.2±0.4	74
4c	181.0±42.2	76.0±20.7	2*	64.7±5.7	3*
4d	115.7±9.45	10.4±1.1	11	2.3±0.2	50
4f	27.47±6.8	4.12±0.17	7*	4.13±0.13	7*
4g	12.96±0.25	1.7±0.1	8	1.4±0.1	9
Cidofovir	290.0±65.6	10.88±2.64	27	10.37±1.87	28
NIOCH-14	455.2±82.4	0.036±0.006	12685	0.006±0.003	76953

Note: CC₅₀ – 50% cytotoxicity concentration at which 50% of cells in uninfected monolayers are destroyed; IC₅₀ – 50% virus inhibiting concentration at which 50% of cells in infected monolayers are preserved; M±SD; M – mean value; SD – standard deviation; n=3 – the number of repeats of measurement of CC₅₀ and IC₅₀; SI – selectivity index of the compound, ratio CC₅₀/IC₅₀. * - chemical compounds with SI<8 are considered as non-perspective for further investigation as antiviral chemicals.

SI 3.2. Comparison of anti-orthopoxvirus activities of novel 2-(2-arylethenyl)imidazoles with previously described [10.1016/j.bmcl.2022.129080; 10.1039/d4md00181h] 2-arylimidazoles similarly substituted at positions 1,4,5 of heterocycle and position 4 of aryl moiety

Table S9. Cytotoxicity and antiviral activity of compounds **3a-c,g**, **4a,b**, **6a,b**, **7a-d**, **8a,b**, **9a,b** against Vaccinia virus (VACV, Copenhagen strain) in Vero cell culture.

№	R ¹	R ²	R ³	CC ₅₀ , μM	IC ₅₀ , μM	SI	№	CC ₅₀ , μM	IC ₅₀ , μM	SI
 3a-c,g							 7a-d			
3a	NO ₂	CH ₂ C(CH ₃) ₂ CH ₂ CO		26.8	6.4	4*	7a	142.38±48.45	0.133±0.03	1072
3b	NO ₂	CH ₃	COCH ₃	22.0	8.4	3*	7b	28.33±6.89	0.651±0.19	44
3c	NO ₂	CH ₃	COOC ₂ H ₅	13.4	N/A	--	7c	45.70±8.93	0.45±0.03	102
3g	H	CH ₃	COOC ₂ H ₅	85.6±20.6	10.5±6.2	8	7d	171.54±55.28	N/A	
 4a,b							 8a,b			
4a	NO ₂	CH ₂ C(CH ₃) ₂ CH ₂ CO		>292.9	N/A	--	8a	1154.34±272.41	3.96±0.22	291
4b	NO ₂	CH ₃	COCH ₃	792.2±193.8	2.6±1.3	305	8b	443.58±102.02	4.76±0.11	93
 6a,b							 9a,b			
6a	NO ₂	CH ₂ C(CH ₃) ₂ CH ₂ CO		>292.9	95.0	>3*	9a	2440.93±610.23	183.4±42.2	13
6b	NO ₂	CH ₃	COCH ₃	>331.9	110.9	>3*	9b	2979.00±655.38	N/A	

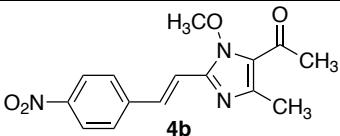
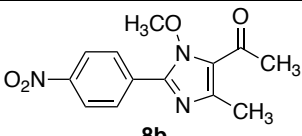
Note: CC₅₀ – 50% cytotoxicity concentration at which 50% of cells in uninfected monolayers are destroyed; IC₅₀ – 50% virus inhibiting concentration at which 50% of cells in infected monolayers are preserved; for certain compounds expanded testing was performed in 3 repeats; M±SD; M – mean value; SD – standard deviation; n=3 – the number of repeats of measurement of CC₅₀ and IC₅₀; SI – selectivity index of the compound, ratio CC₅₀/IC₅₀. * - chemical compounds with SI<8 are considered as non-perspective for further investigation as antiviral chemicals. N/A – not active. Data for compounds **7a,b**, **8a,b**, **9a,b** were obtained from [10.1039/d4md00181h]; data for compounds **7c,d** – from [10.1016/j.bmcl.2022.129080].

Commentary. For now, the influence of the presence of ethenyl linker between heterocyclic and aryl moieties of imidazole derivatives under study is not evident enough. Nevertheless, it is clear that in most cases methylation of 1-hydroxyimidazoles led to less toxic

compounds. Obviously, 2-(2-arylethenyl)-1-hydroxyimidazoles **3a-c,g** exhibited poorer virus-inhibiting activities than the ones of similarly substituted 2-aryl-1-hydroxyimidazoles **7a-d**. Strangely enough, while methylation of 2-aryl-1-hydroxyimidazoles **7a,b** resulted in the decrease of virus-inhibiting activity in case of 2-aryl-1-methoxyimidazoles **8a,b**; 2-(2-arylethenyl)-1-methoxyimidazoles could demonstrate more promising virus-inhibiting concentrations as in the case of compound **4b**. The activities of 1-methylimidazole 3-oxides **6a,b**, **9a,b** were poor regardless of the character of substituent in position 2 of imidazole.

It may also be of interest to compare the activities against various orthopoxviruses of the lead compound of 2-(arylethenyl)imidazole series (**4b**) and of similarly substituted at positions 1,4,5 of heterocycle 2-(4-nitrophenyl)imidazole **8b** [[10.1039/d4md00181h](#)] (Table S10).

Table S10. Cytotoxicity and antiviral activity of compounds **4b** and **8b** against Vaccinia virus (VACV, Copenhagen strain), Cowpox virus (CPXV, Grishak strain) and Ectromelia virus (ECTV, K-1 strain) in Vero cell culture.

 4b				 8b			
CC ₅₀ , μM	Virus	IC ₅₀ , μM	SI	CC ₅₀ , μM	Virus	IC ₅₀ , μM	SI
792.2±193.8	VACV	2.6±1.3	305	443.58±102.02	VACV	4.76±0.11	93
	CPXV	21.6±1.6	37		CPXV	22.27±4.89	20
	ECTV	10.7±1.4	74		ECTV	13.88±1.13	32

Note: CC₅₀ – 50% cytotoxicity concentration at which 50% of cells in uninfected monolayers are destroyed; IC₅₀ – 50% virus inhibiting concentration at which 50% of cells in infected monolayers are preserved; M±SD; M – mean value; SD – standard deviation; n=3 – the number of repeats of measurement of CC₅₀ and IC₅₀; SI – selectivity index of the compound, ratio CC₅₀/IC₅₀.

Strictly speaking, the virus-inhibiting concentrations of compounds **4b** and **8b** against all the three orthopoxviruses are comparable. Due to its lower cytotoxicity, 1-methoxy-2-(2-(4-nitrophenyl)ethenyl)imidazole **4b** obtained higher selectivity indices than the ones of 1-methoxy-2-(4-nitrophenyl)imidazole **8b**.

SI 4. Copies of NMR spectra of compounds 3a-h, 4a-h, 5, 6a-d.

S4.1. 3-Hydroxy-6,6-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (**3a**).

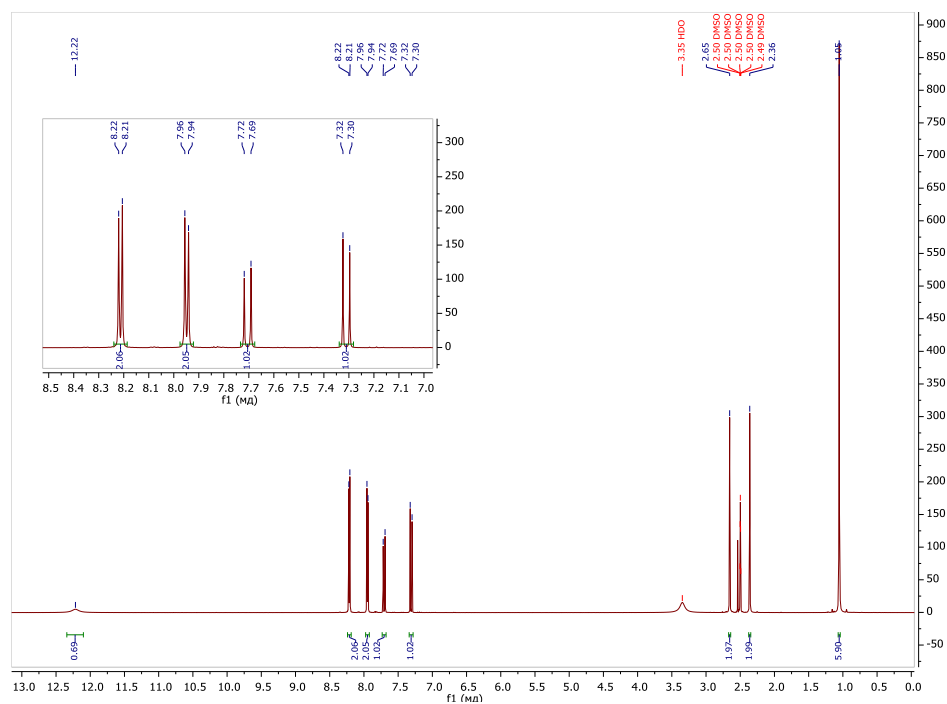
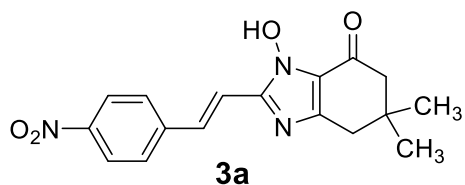


Fig. S 1. ¹H NMR spectrum of compound 3a (600 MHz, DMSO-*d*₆).

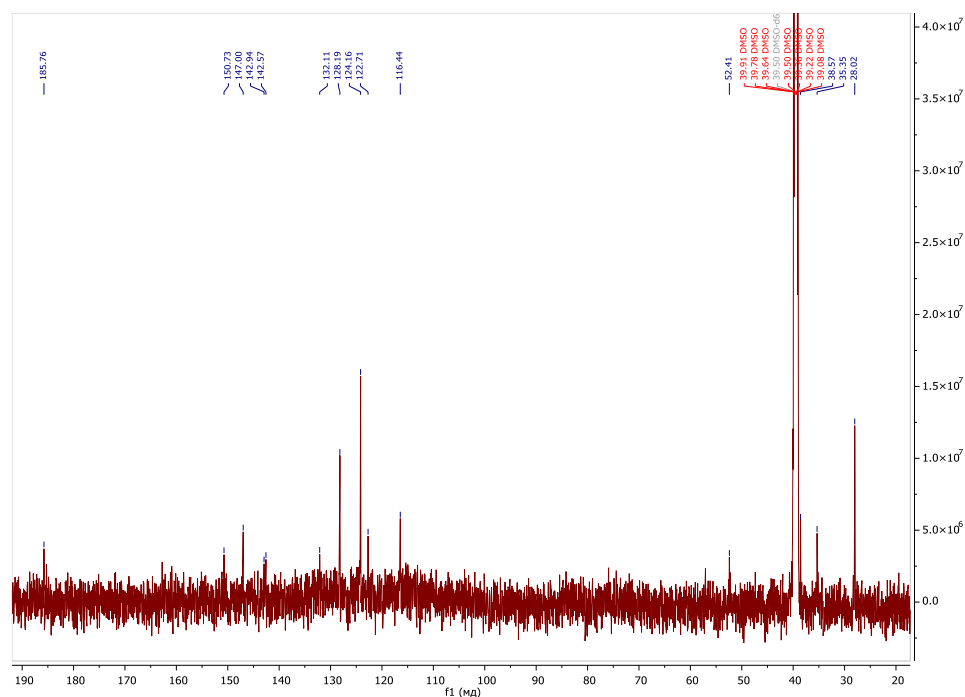


Fig. S 2. ¹³C NMR spectrum of compound 3a (151 MHz, DMSO-*d*₆).

S4.2. 1-(1-Hydroxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazol-5-yl)ethanone (**3b**).

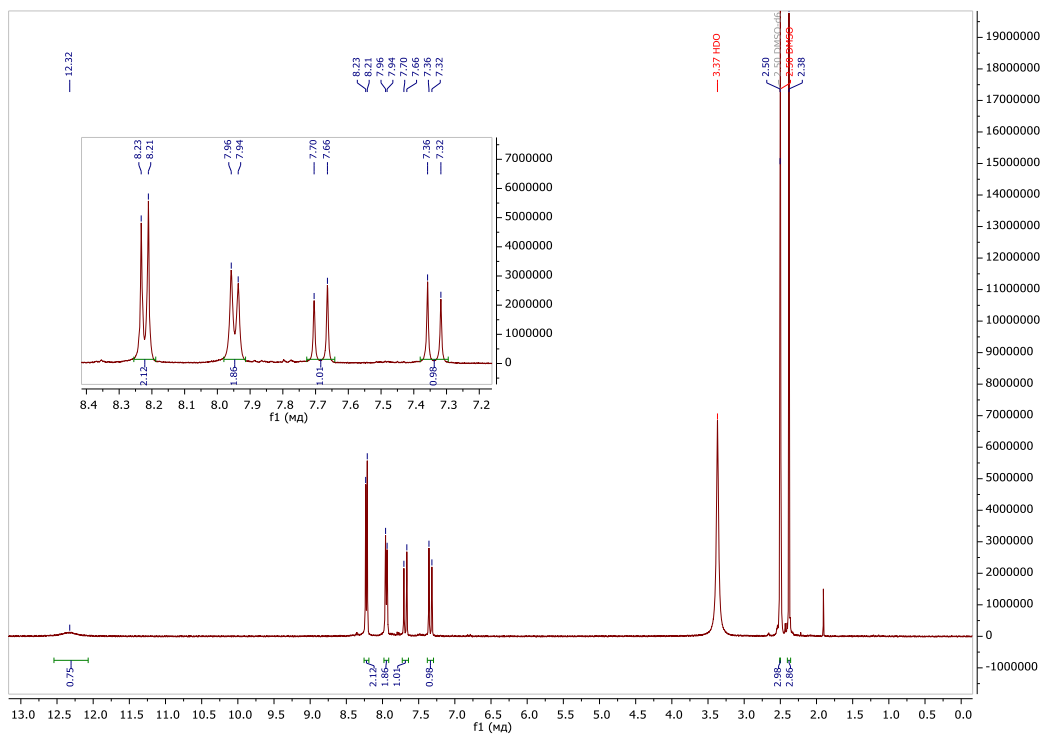
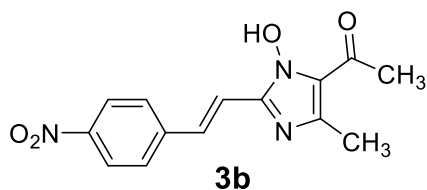


Fig. S 3. ¹H NMR spectrum of compound **3b** (400 MHz, DMSO-*d*₆).

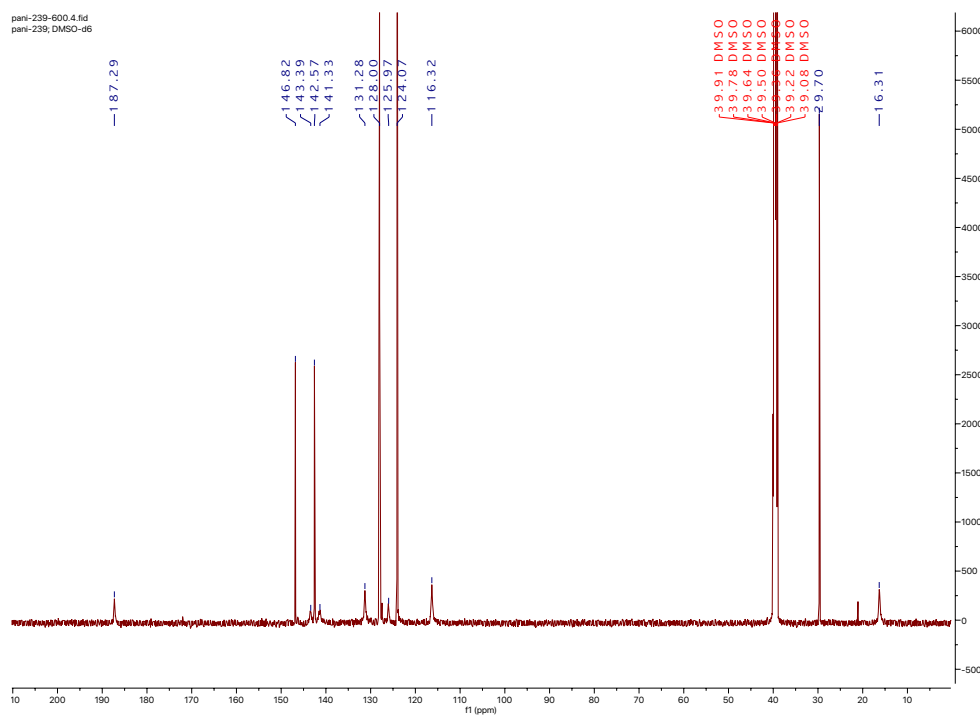


Fig. S 4. ¹³C NMR spectrum of compound **3b** (151 MHz, DMSO-*d*₆).

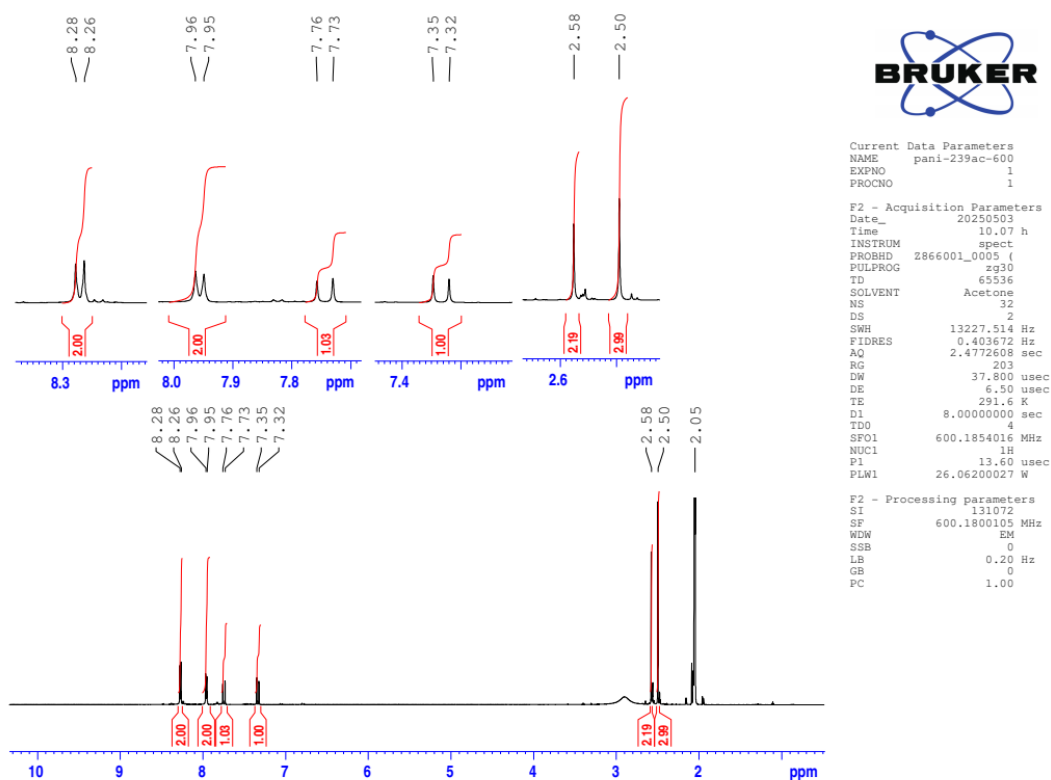


Fig. S 5. ^1H NMR spectrum of compound 3b (600 MHz, acetone- d_6). The integral intensity of 2.58 ppm signal of methyl group is lowered due to the partial exchange of hydrogen for deuterium from the solvent.

^{13}C

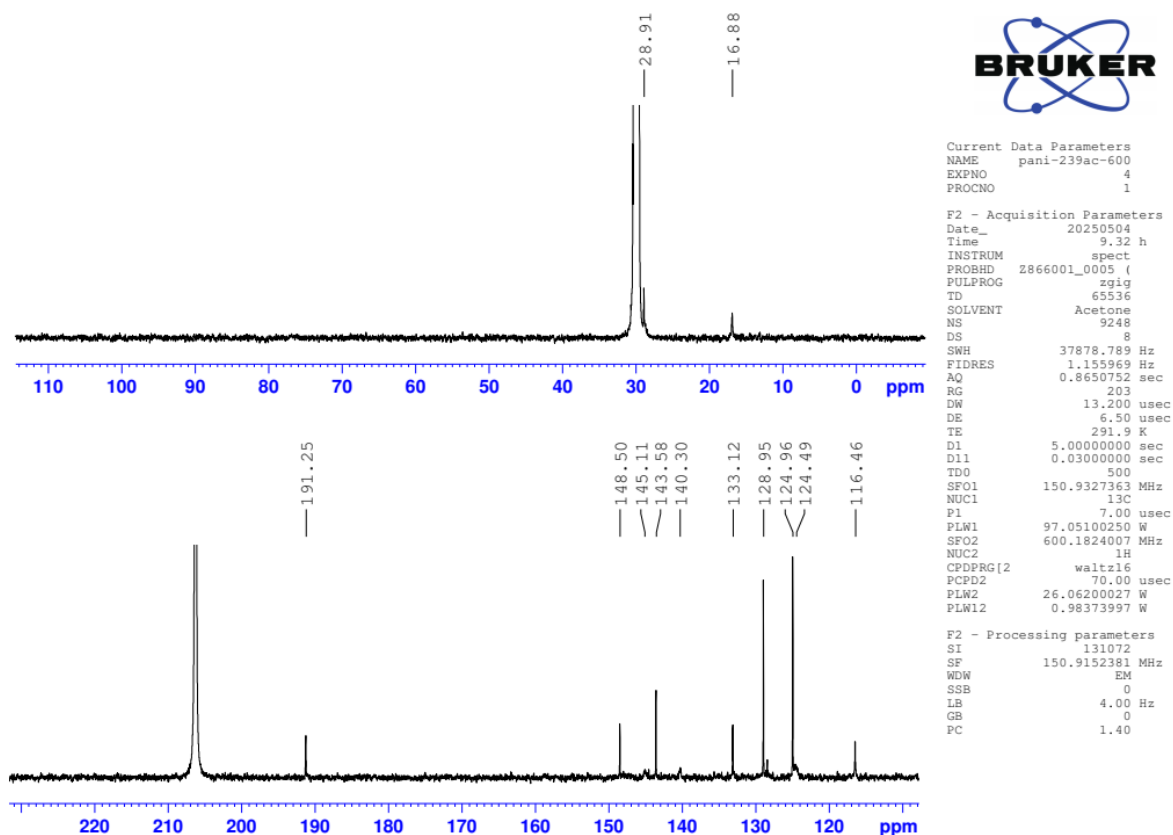


Fig. S 6. ^{13}C NMR spectrum of compound 3b (151 MHz, acetone- d_6).

¹H-¹³C HSQC

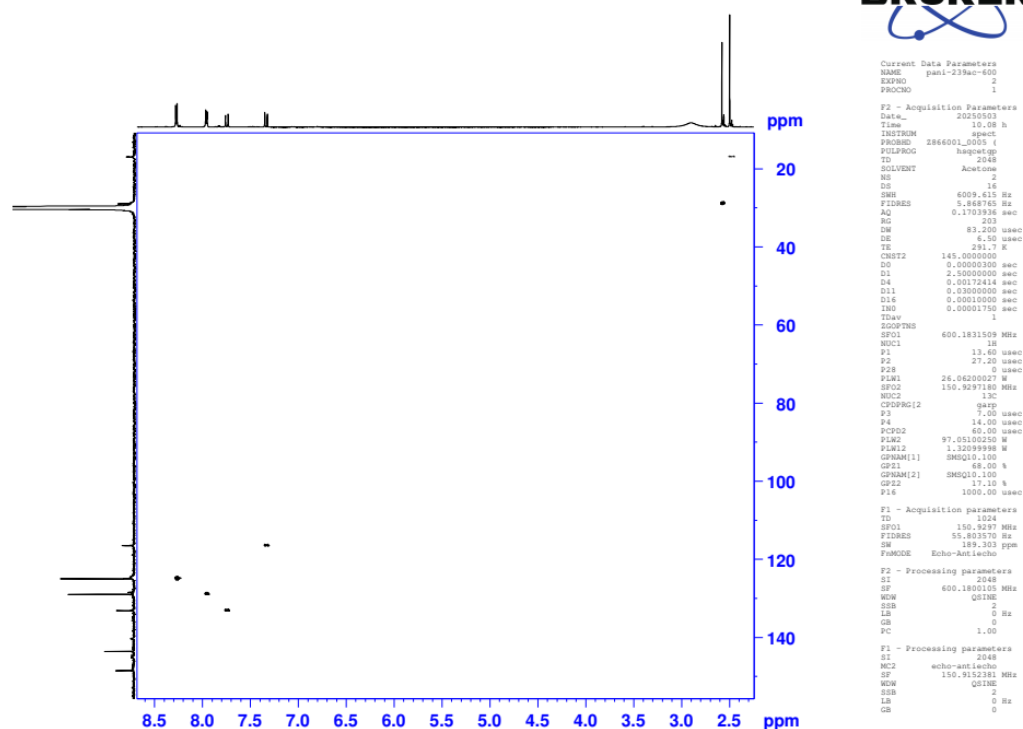


Fig. S 7. ¹H-¹³C HSQC spectrum of compound 3b (600 MHz – ¹H, 151 MHz – ¹³C, acetone-*d*₆).

¹H-¹³C HMBC

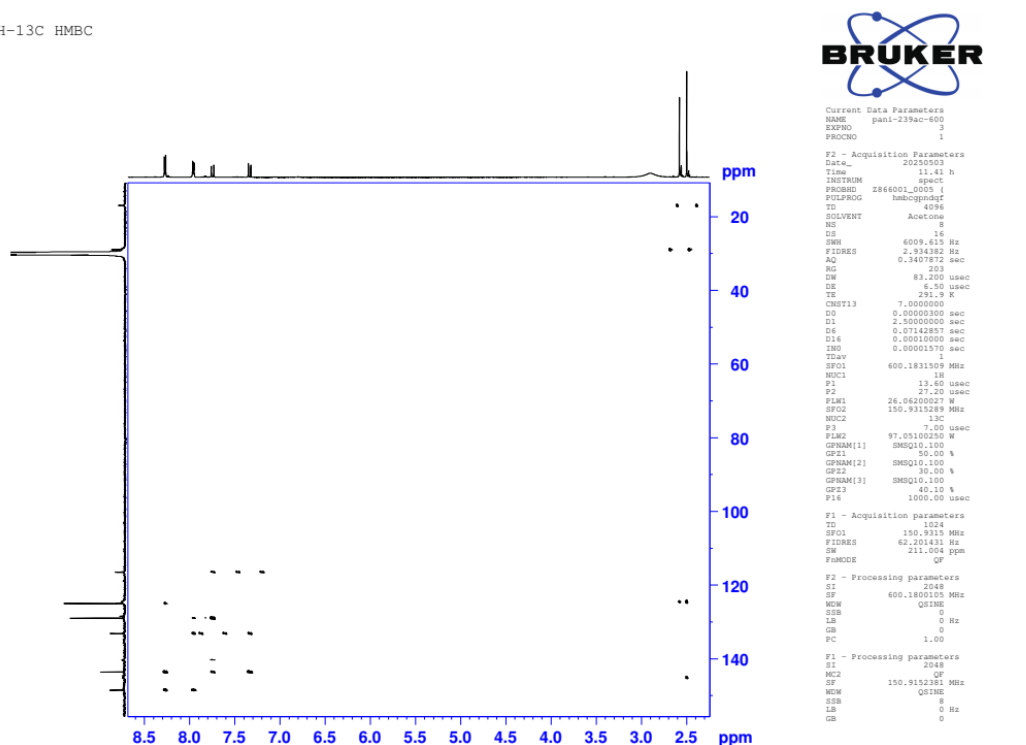


Fig. S 8. ¹H-¹³C HMBC spectrum of compound 3b (600 MHz – ¹H, 151 MHz – ¹³C, acetone-*d*₆).

^1H - ^{13}C HMBC

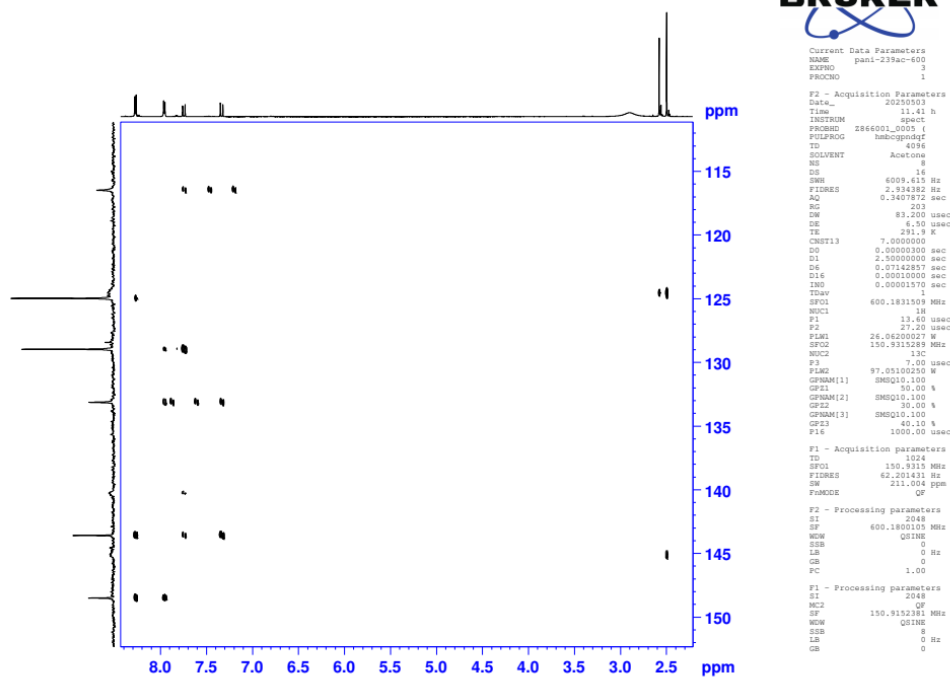


Fig. S 9. Part of ^1H - ^{13}C HMBC spectrum of compound 3b (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

^1H - ^1H NOESY

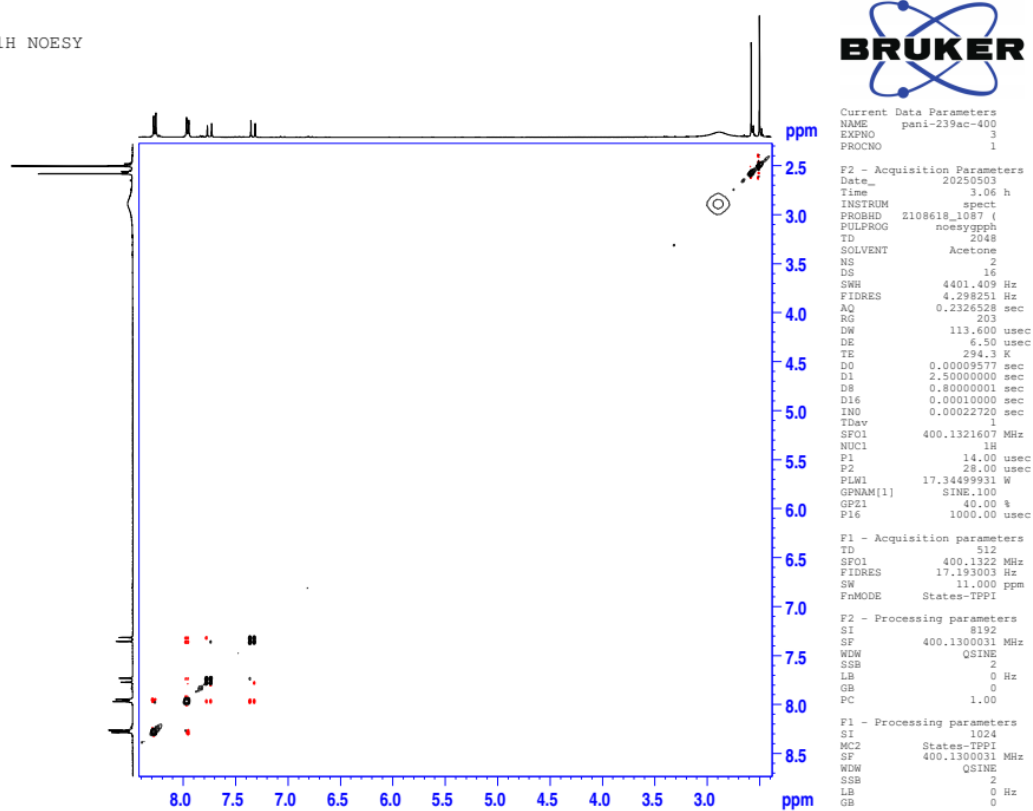


Fig. S 10. ^1H - ^1H NOESY spectrum of compound 3b (400 MHz, acetone- d_6).

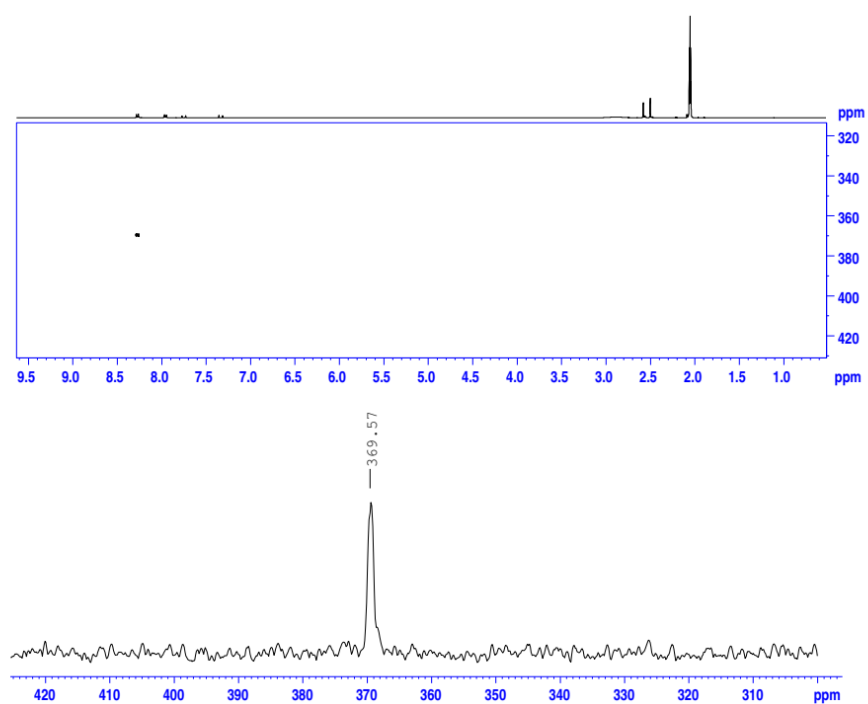


Fig. S 11. ^1H - ^{15}N HMBC spectrum of compound 3b (400 MHz – ^1H , 41 MHz – ^{15}N , acetone- d_6).

S4.3. Ethyl 1-hydroxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole-5-carboxylate (3c**).**

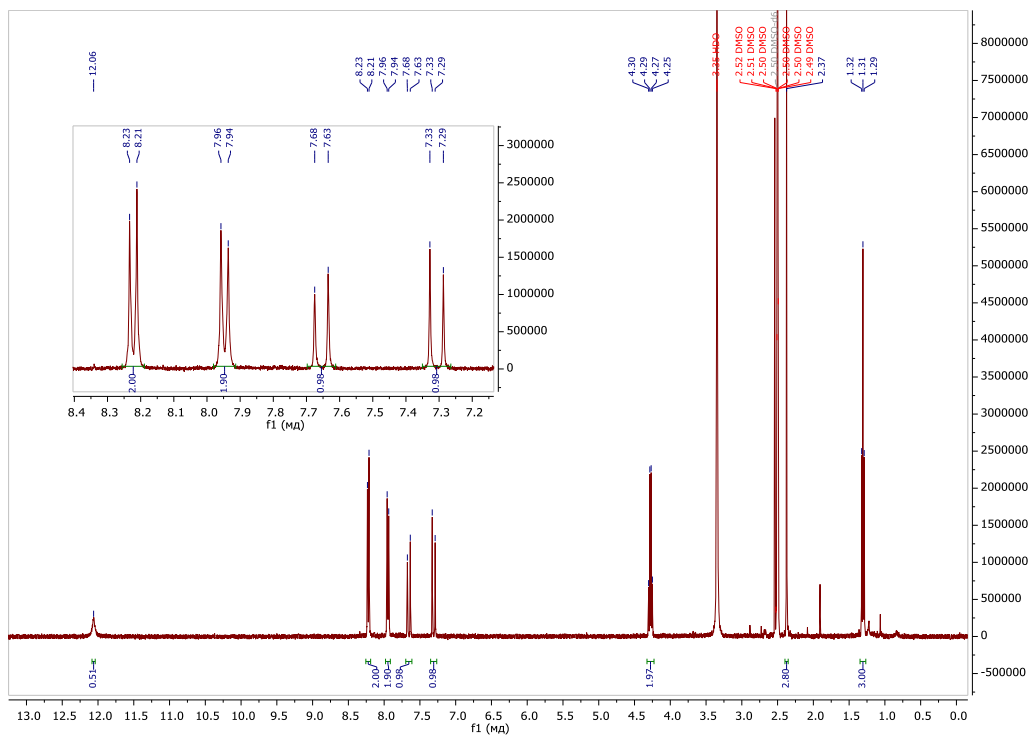
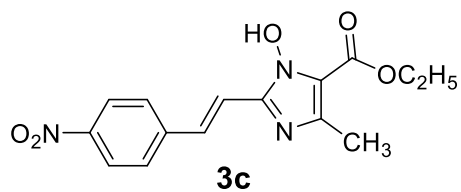


Fig. S 12. ¹H NMR spectrum of compound 3c (400 MHz, DMSO-*d*₆).

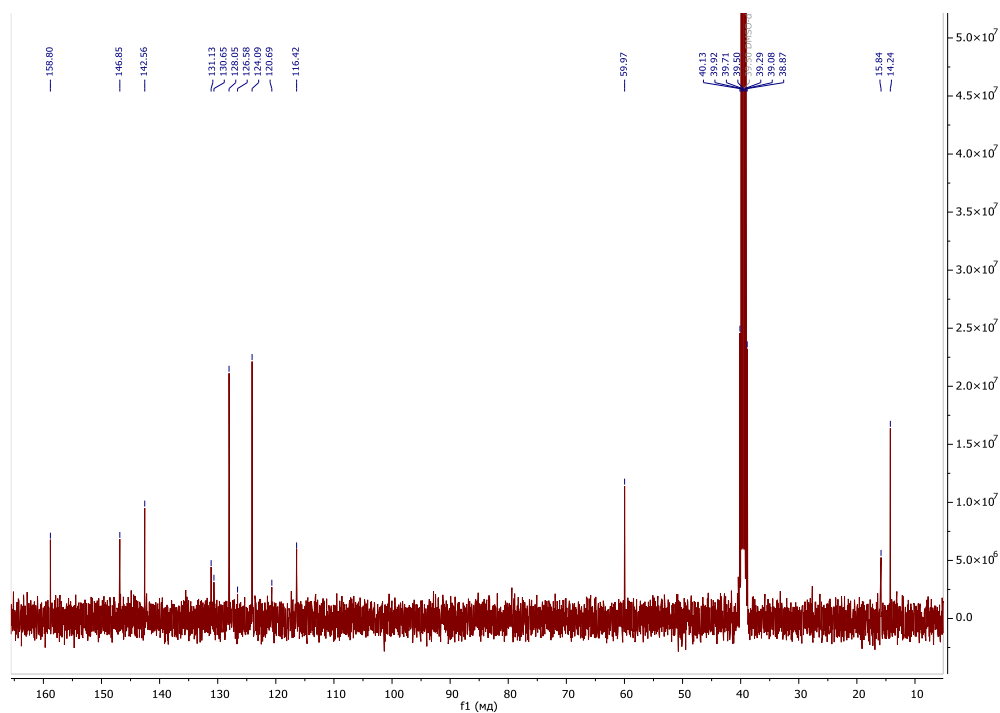
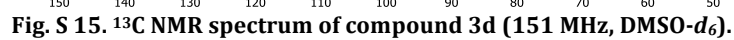


Fig. S 13. ¹³C NMR spectrum of compound 3c (101 MHz, DMSO-*d*₆).

3d



S4.5. 3-Hydroxy-6,6-dimethyl-2-((*E*)-2-phenylethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (**3e**).

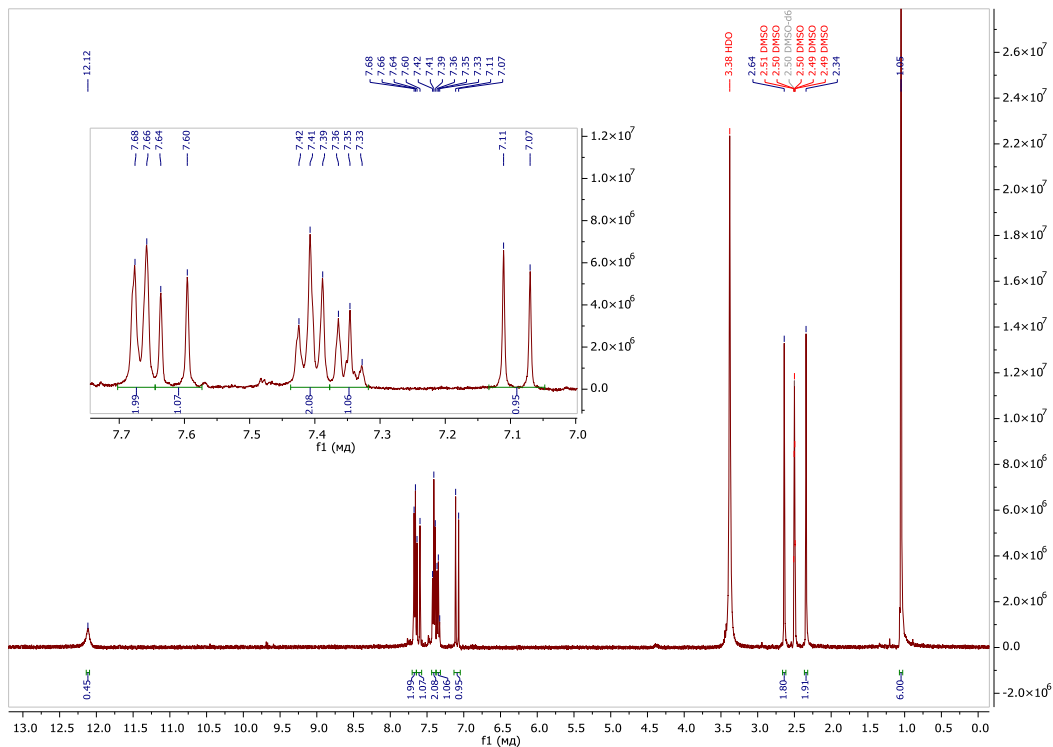
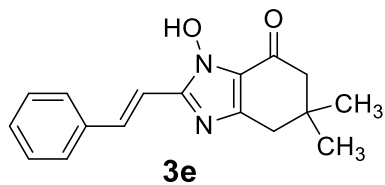


Fig. S 16. ¹H NMR spectrum of compound **3e** (400 MHz, DMSO-*d*₆).

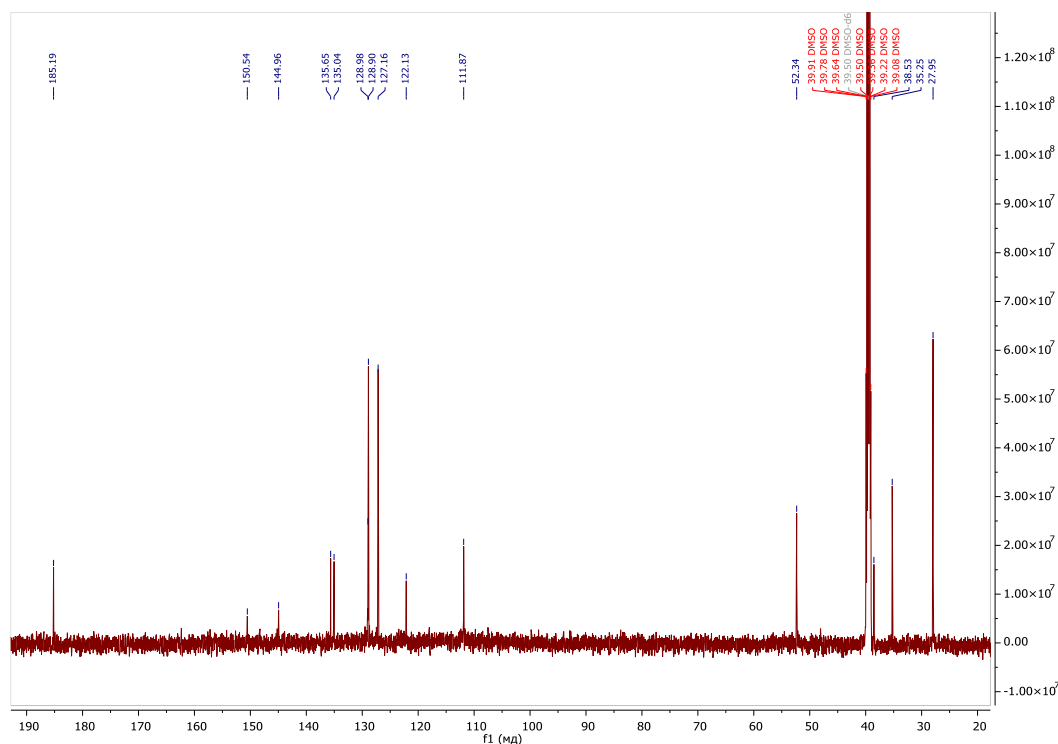


Fig. S 17. ¹³C NMR spectrum of compound **3e** (151 MHz, DMSO-*d*₆).

3.6. 1-(1-Hydroxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazol-5-yl)ethanone (**3f**).

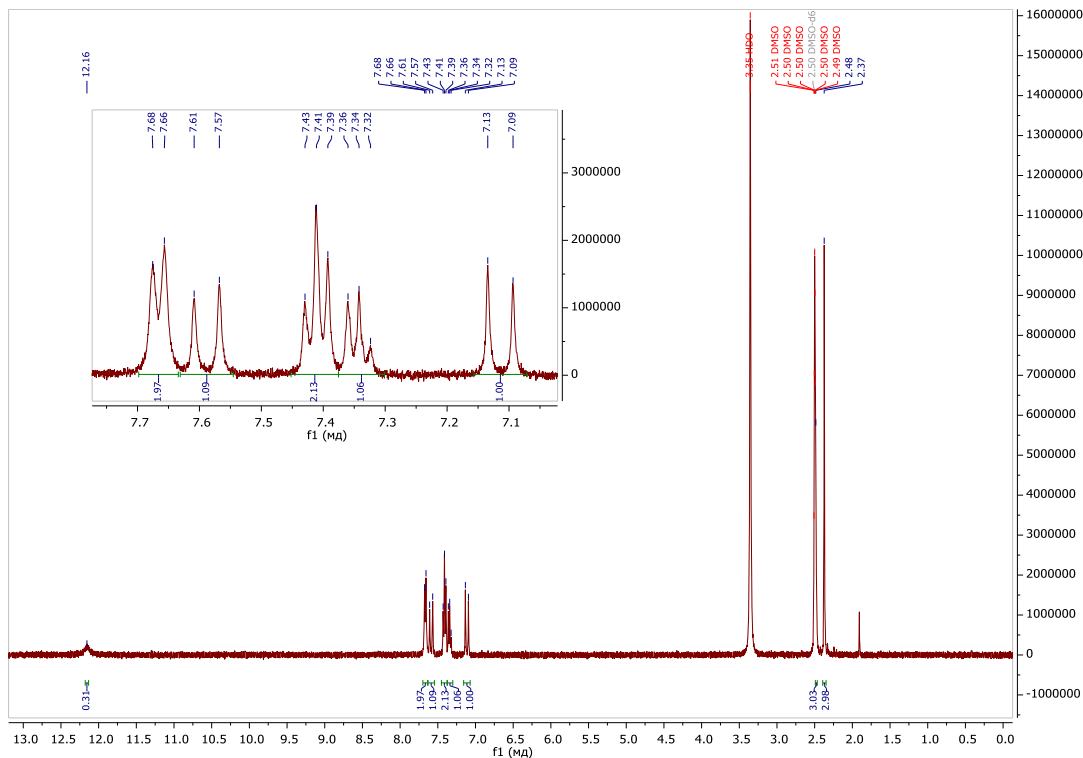
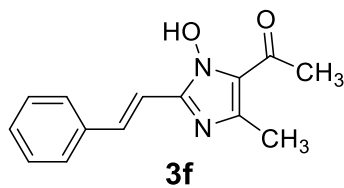


Fig. S 18. ¹H NMR spectrum of compound **3f** (400 MHz, DMSO-*d*₆).

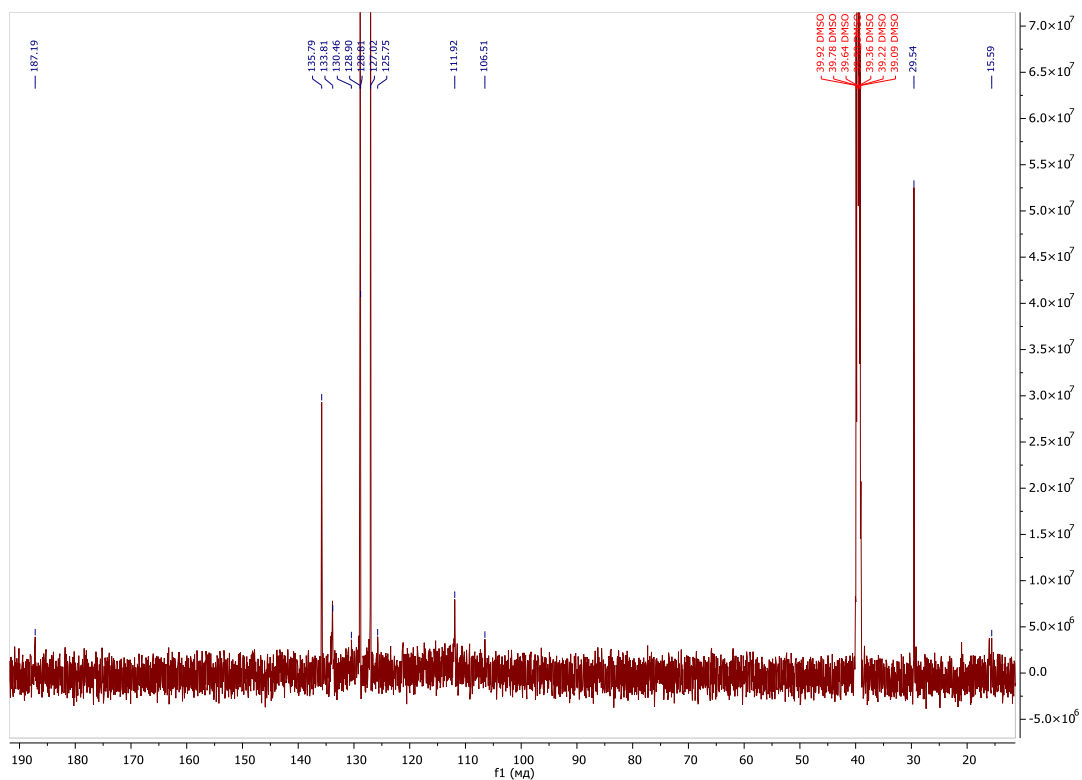


Fig. S 19. ¹³C NMR spectrum of compound **3f** (151 MHz, DMSO-*d*₆).

S4.7. Ethyl 1-hydroxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole-5-carboxylate (**3g**).

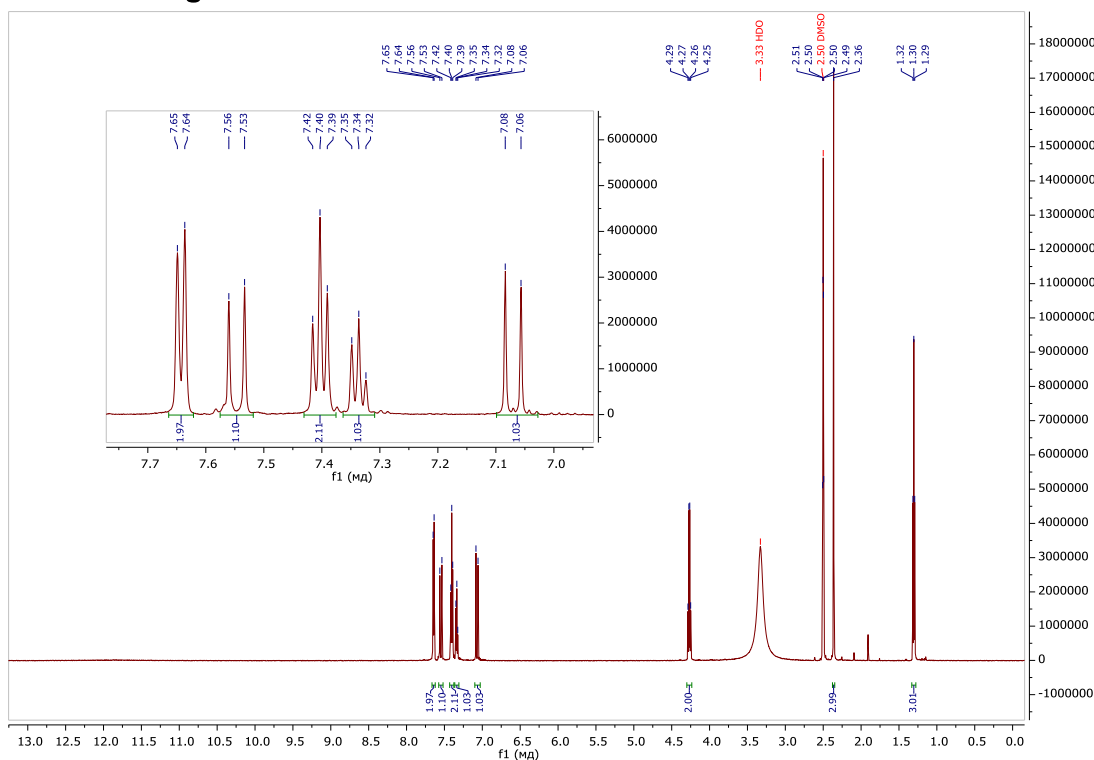
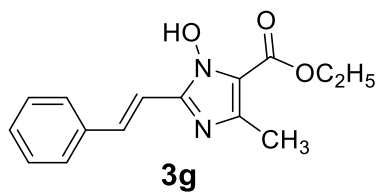


Fig. S 20. ¹H NMR spectrum of compound **3g** (600 MHz, DMSO-*d*₆).

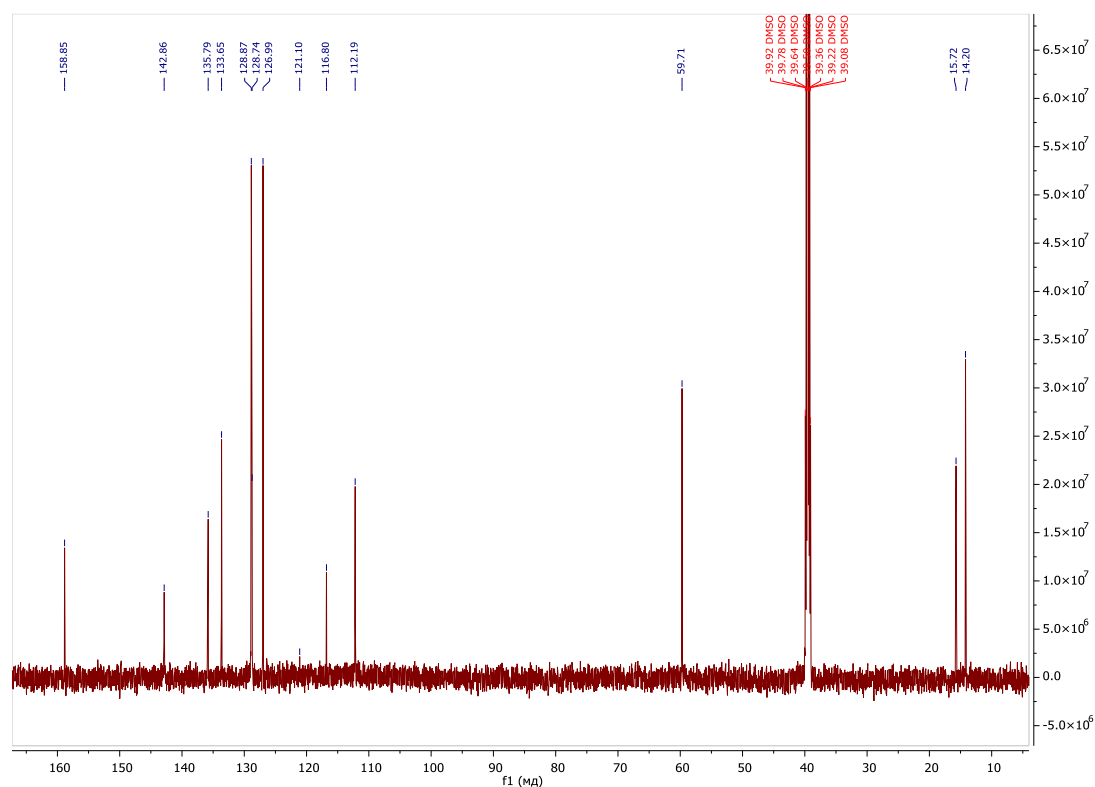


Fig. S 21. ¹³C NMR spectrum of compound **3g** (151 MHz, DMSO-*d*₆).

S4.8. 4,5-Dimethyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazol-1-ol (**3h**).

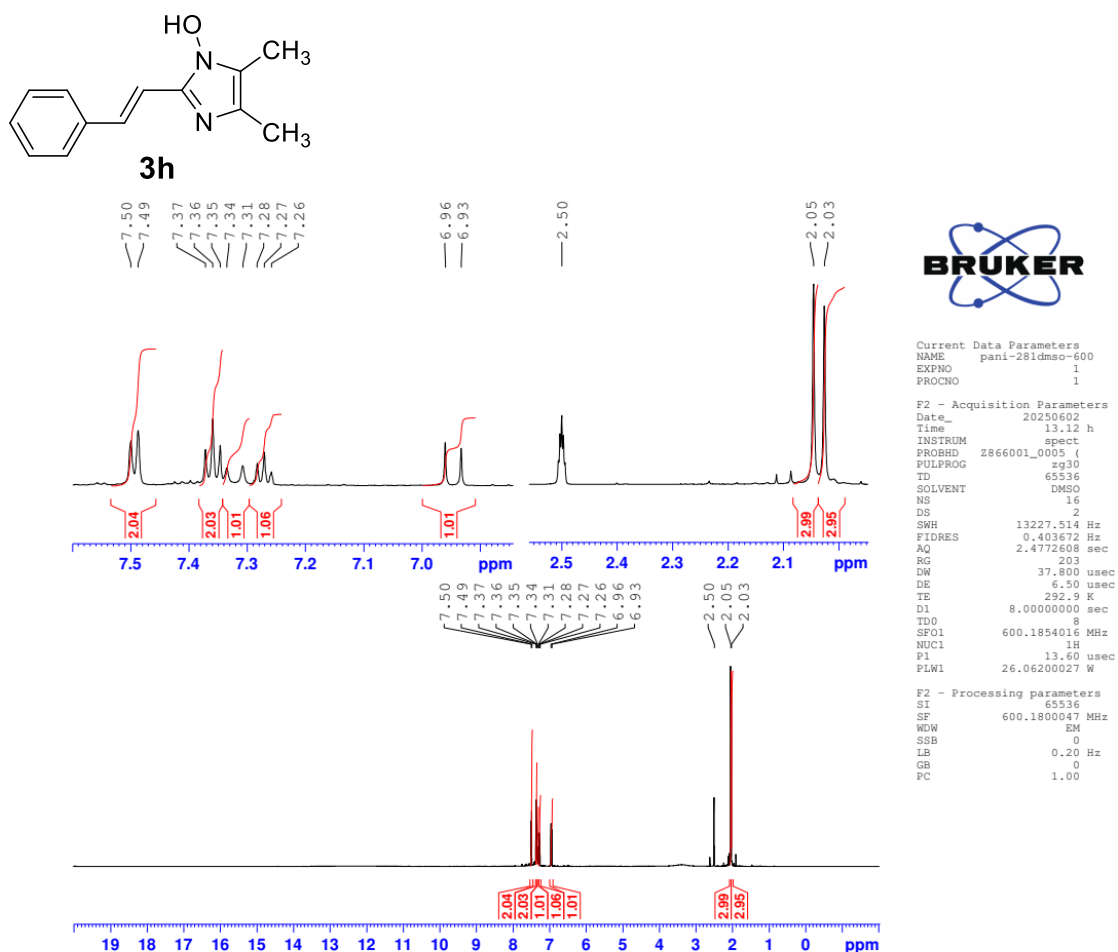


Fig. S 22. ¹H NMR spectrum of compound 3h (600 MHz, DMSO-*d*₆, 23.4°C).

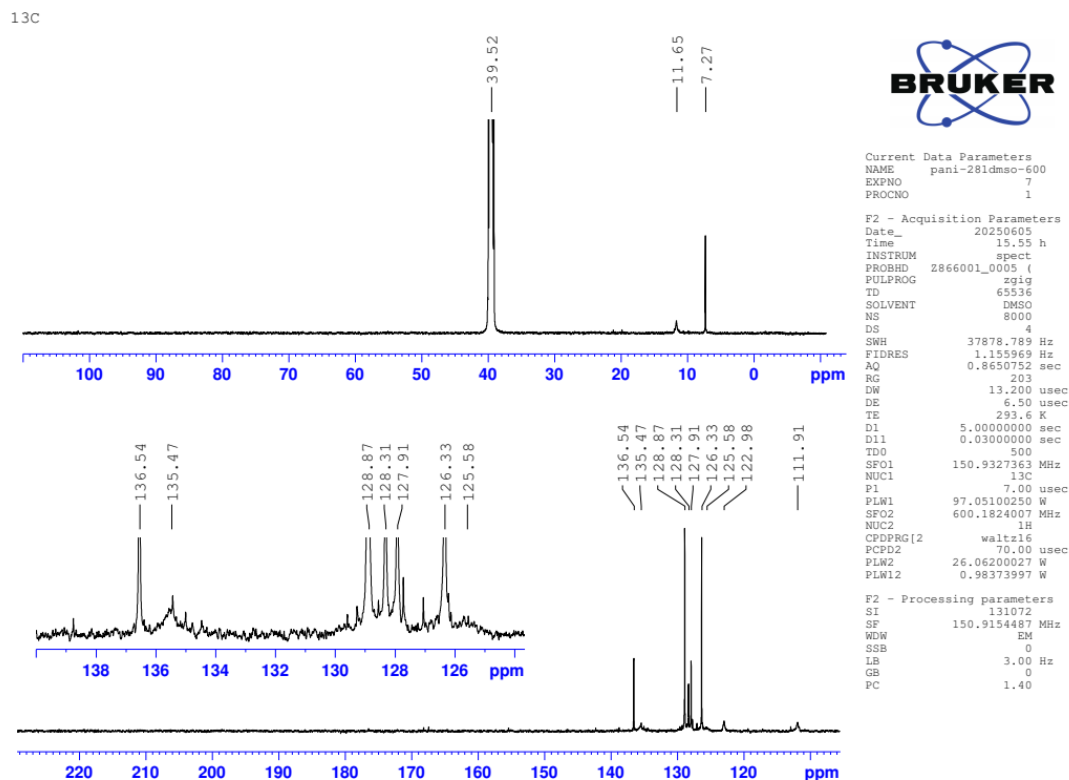


Fig. S 23. ¹³C NMR spectrum of compound 3h (151 MHz, DMSO-*d*₆, 23.4°C).

PANI-281; acetone-d₆

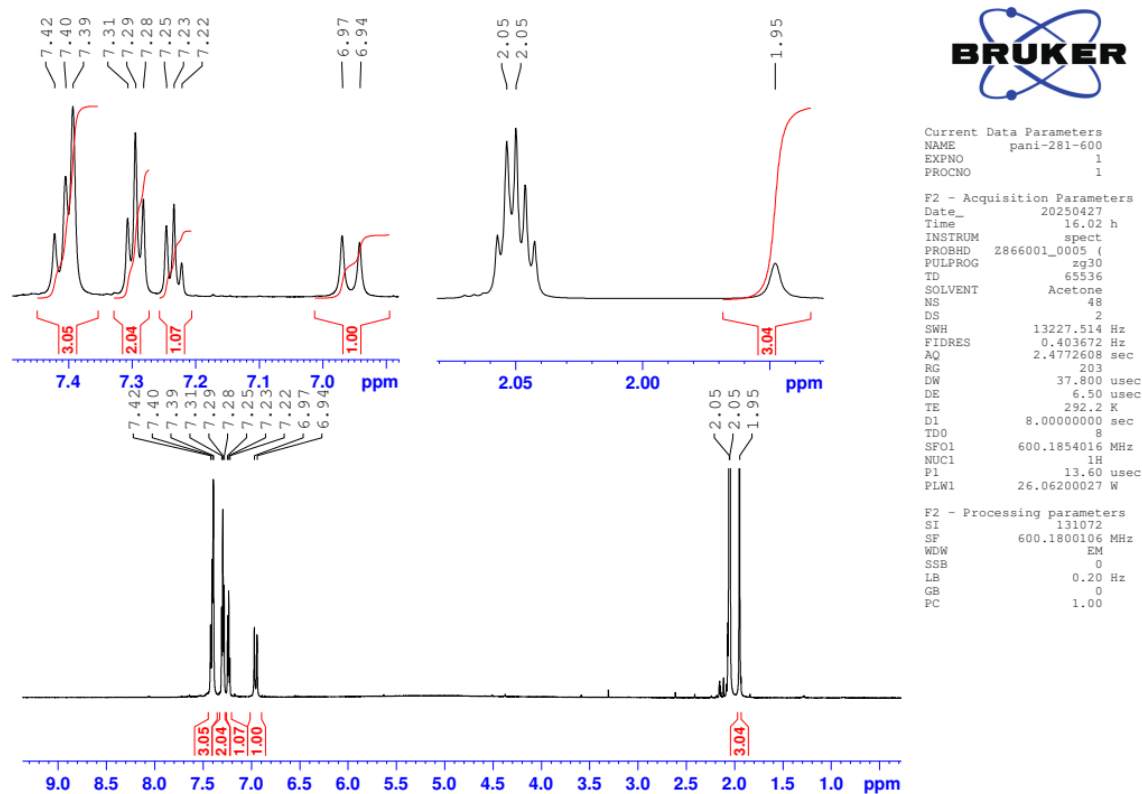


Fig. S 24. ¹H NMR spectrum of compound 3h (600 MHz, acetone-d₆).

¹H-¹H COSY

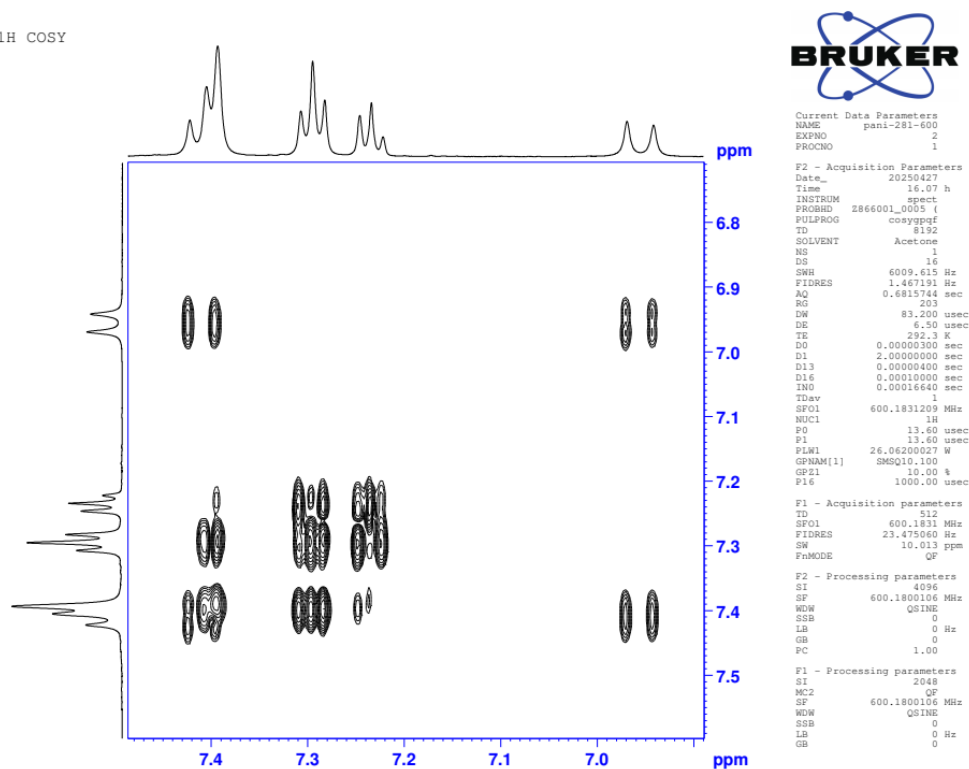


Fig. S 25. ¹H-¹H COSY spectrum of compound 3h (600 MHz, acetone-d₆).

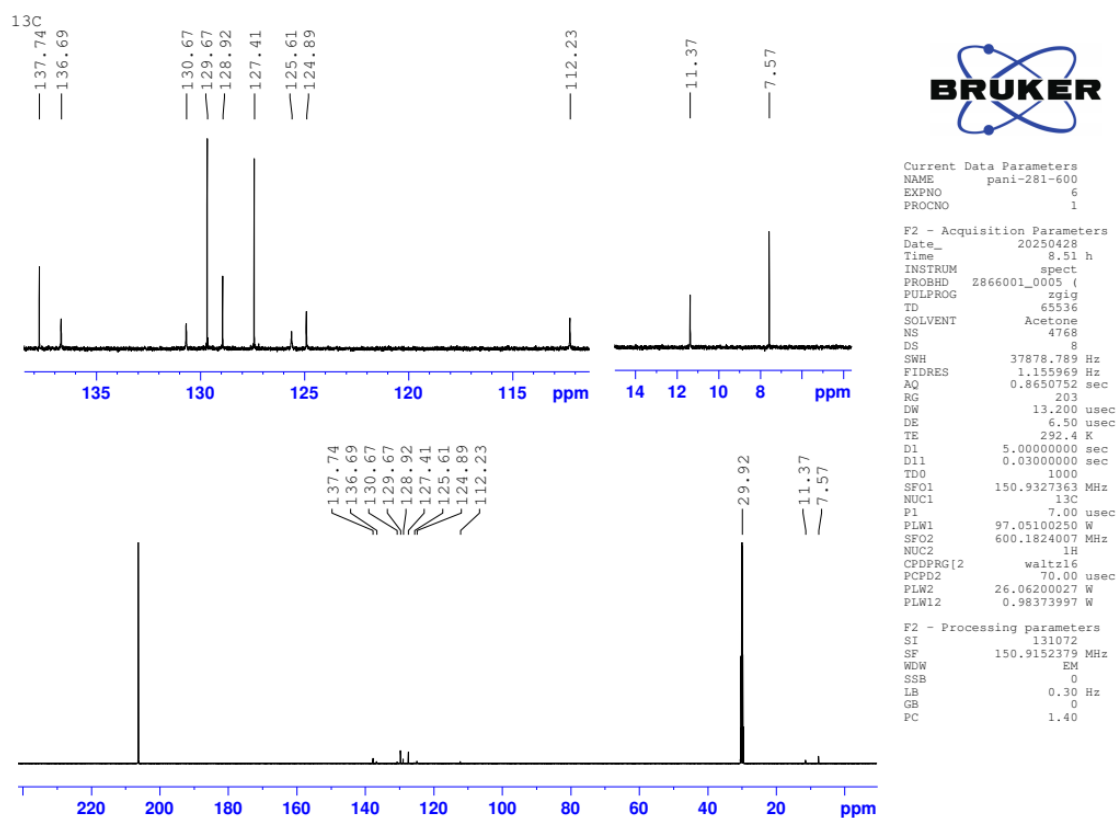


Fig. S 26. ^{13}C NMR spectrum of compound 3h (151 MHz, acetone- d_6).

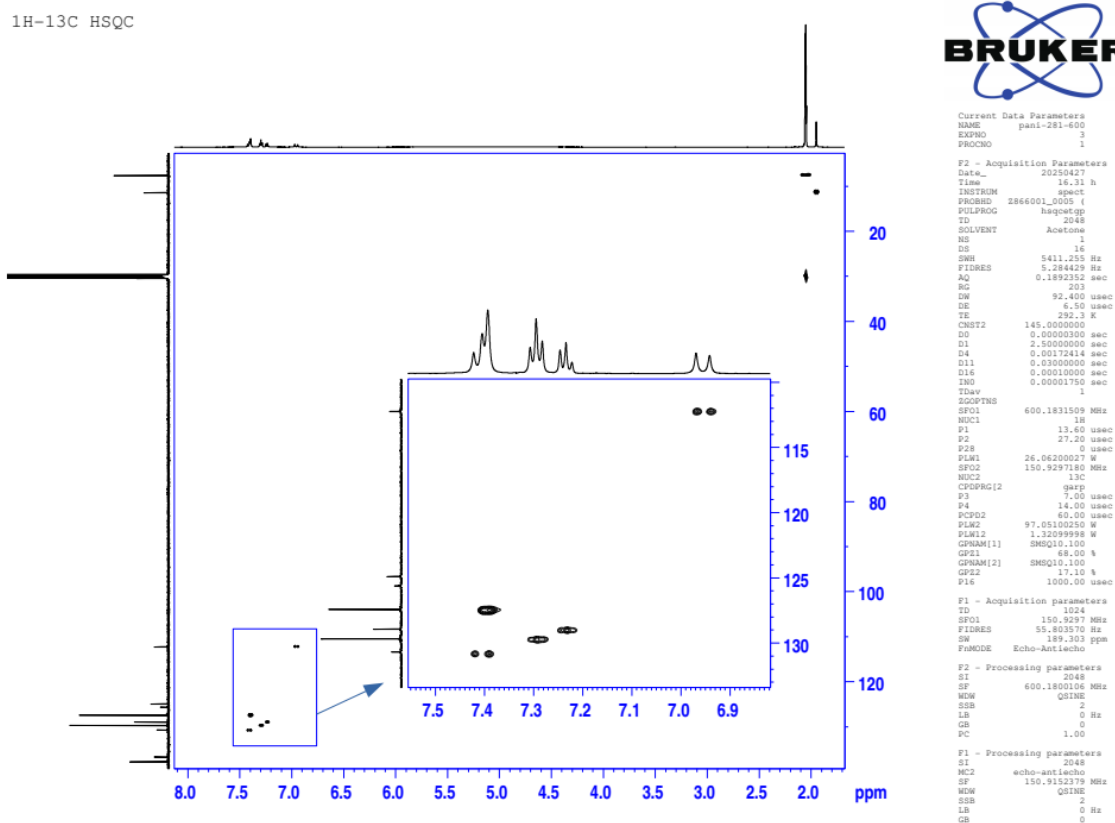


Fig. S 27. ^1H - ^{13}C HSQC spectrum of compound 3h (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

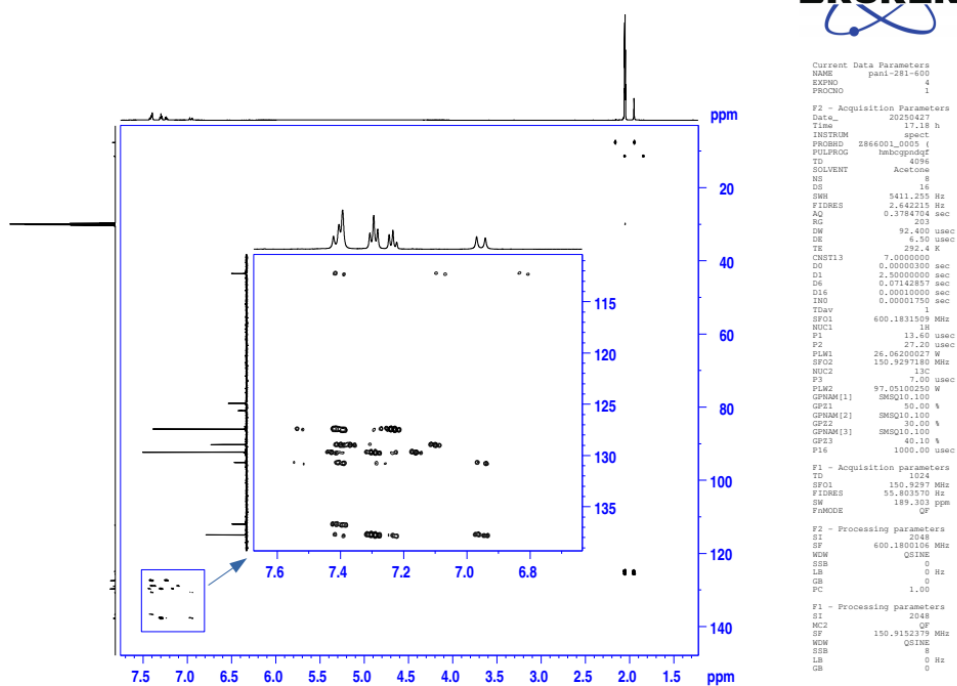


Fig. S 28. ^1H - ^{13}C HMBC spectrum of compound 3h (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

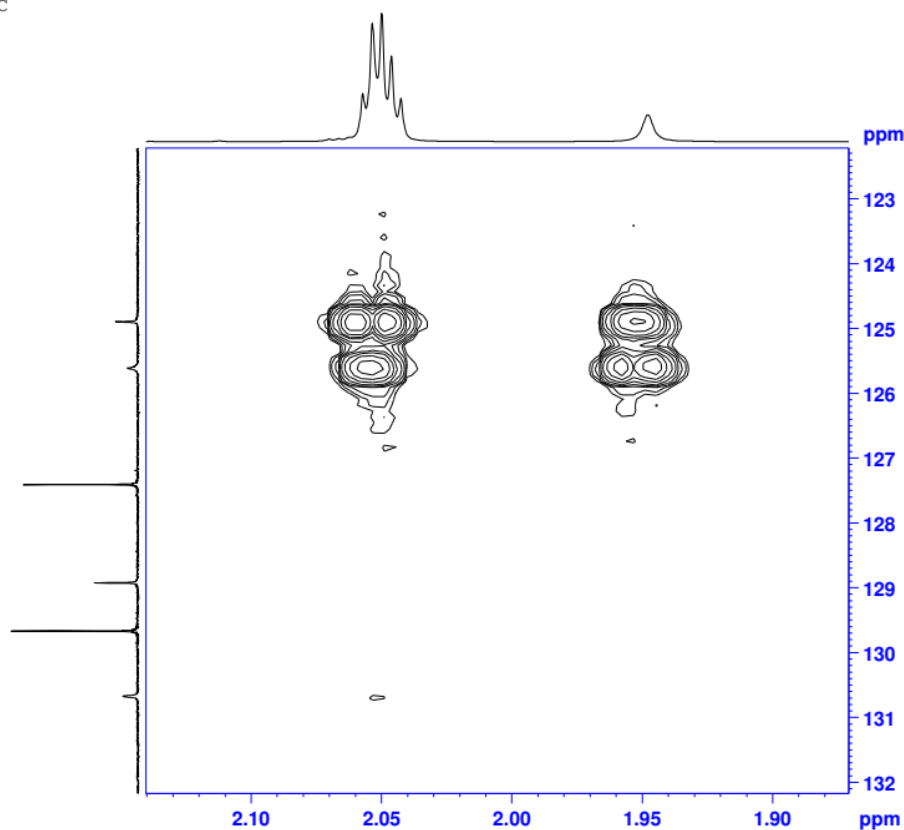


Fig. S 29. Part of ^1H - ^{13}C HMBC spectrum of compound 3h (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

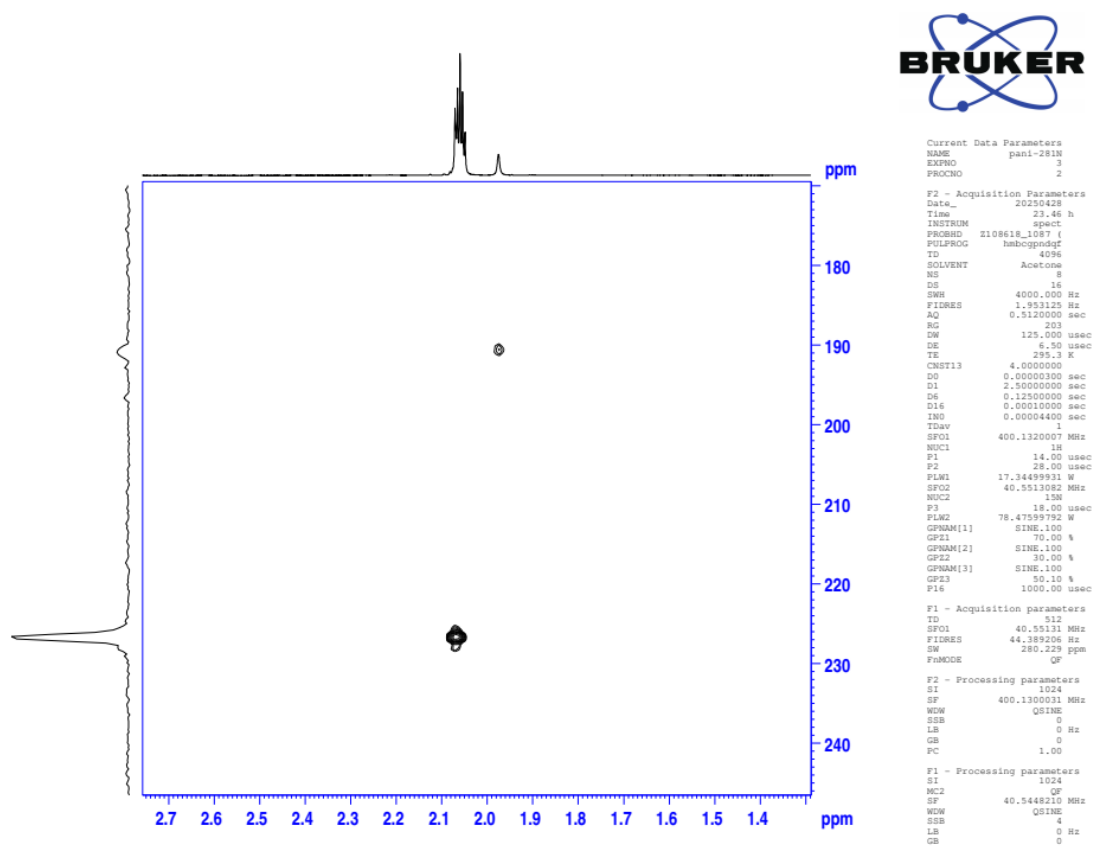


Fig. S 30. ^1H - ^{15}N HMBC spectrum of compound 3h (400 MHz - ^1H , 41 MHz - ^{15}N , acetone- d_6).

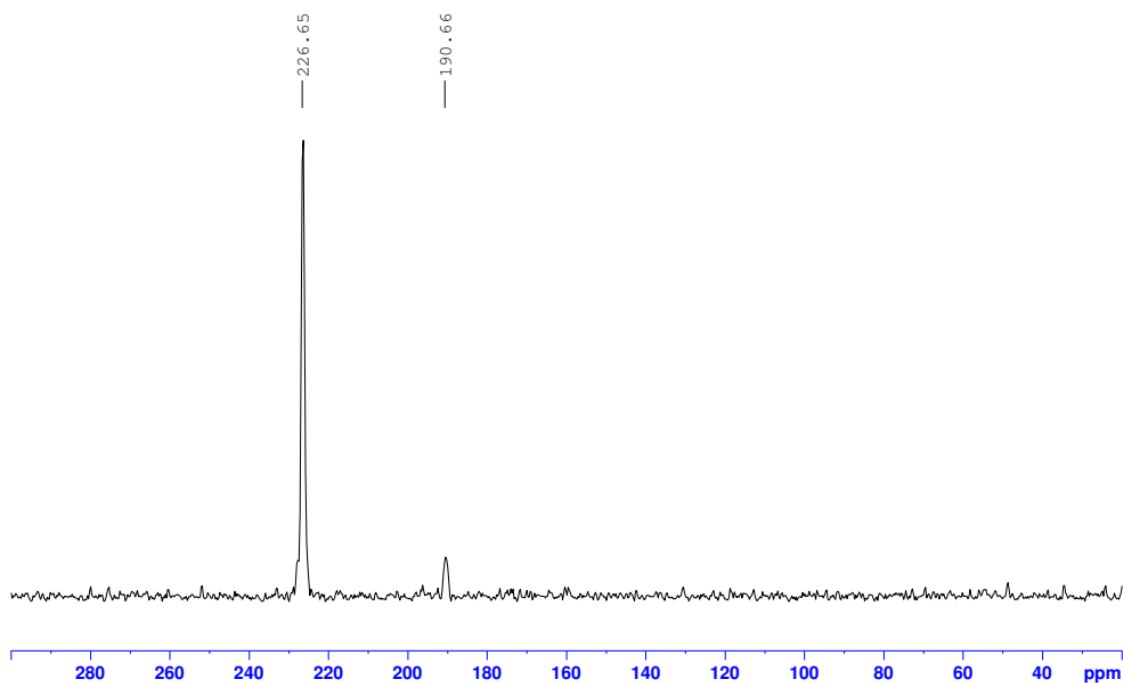


Fig. S 31. ^{15}N NMR spectrum of compound 3h (41 MHz, acetone- d_6), obtained by projection from ^1H - ^{15}N HMBC spectrum.

¹H-¹³C HSQC

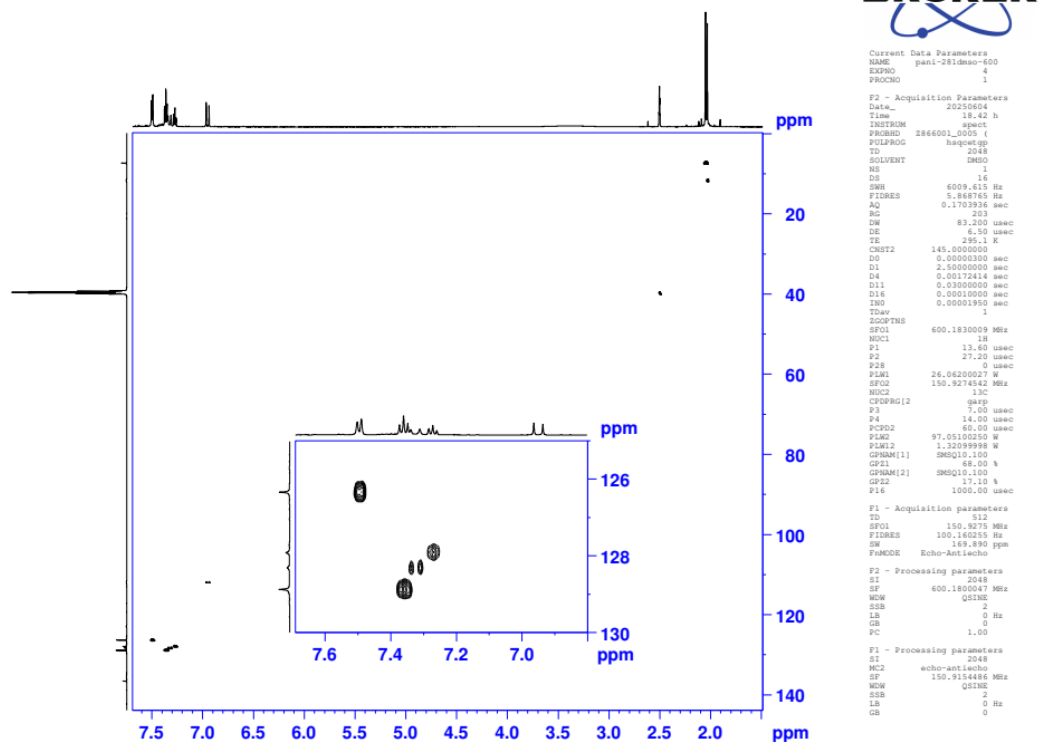


Fig. S 32. ¹H-¹³C HSQC spectrum of compound 3h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆, 23.4°C).

¹H-¹³C HMBC

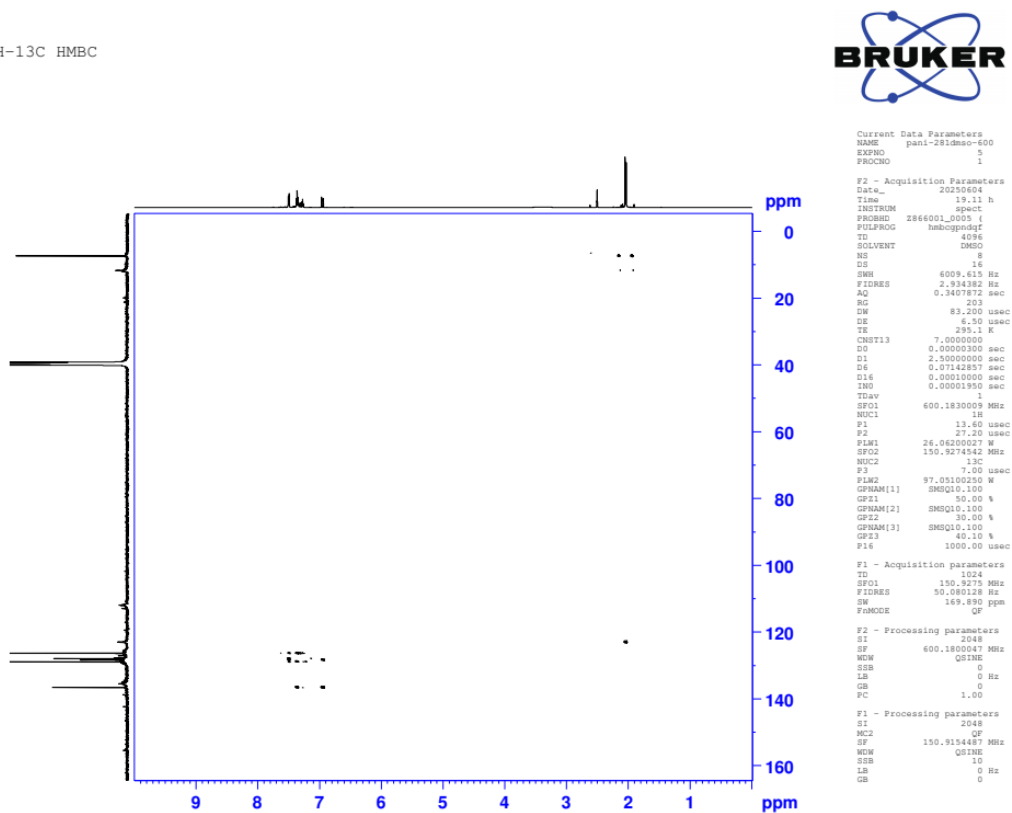


Fig. S 33. ¹H-¹³C HMBC spectrum of compound 3h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆, 23.4°C).

¹H-¹³C HMBC

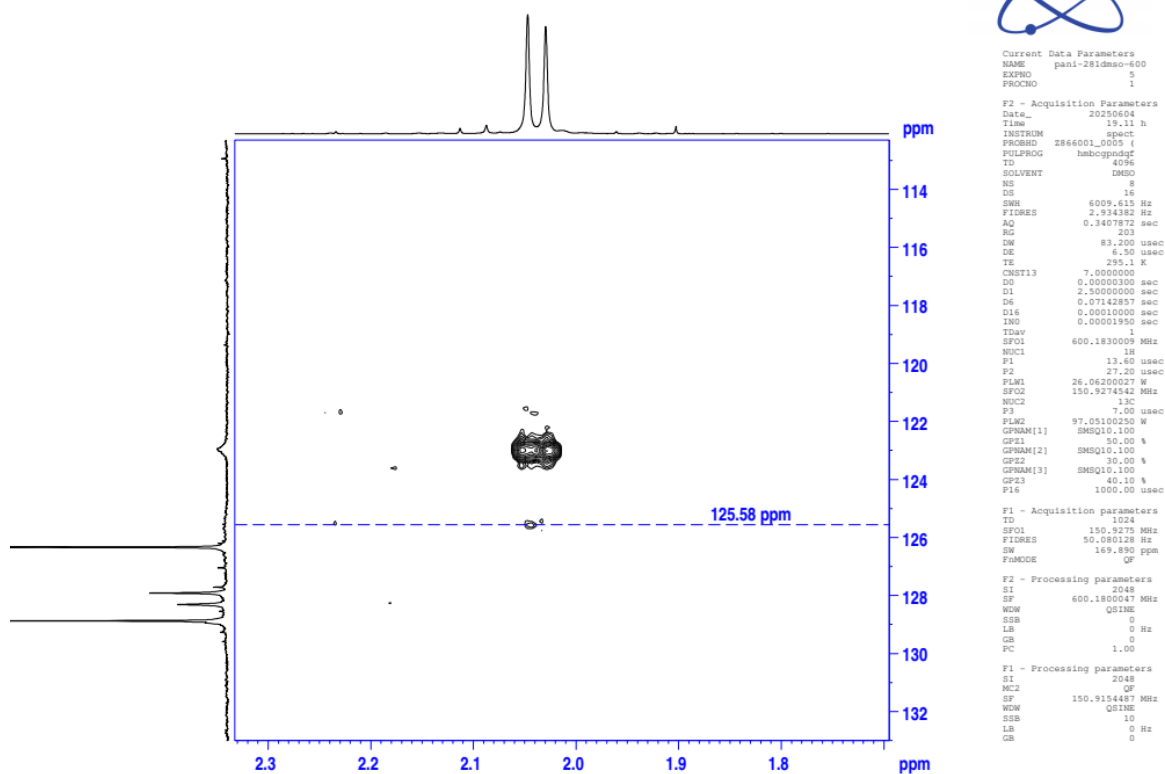


Fig. S 34. Part of ¹H-¹³C HMBC spectrum of compound 3h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆, 23.4°C).

¹H-¹³C HMBC

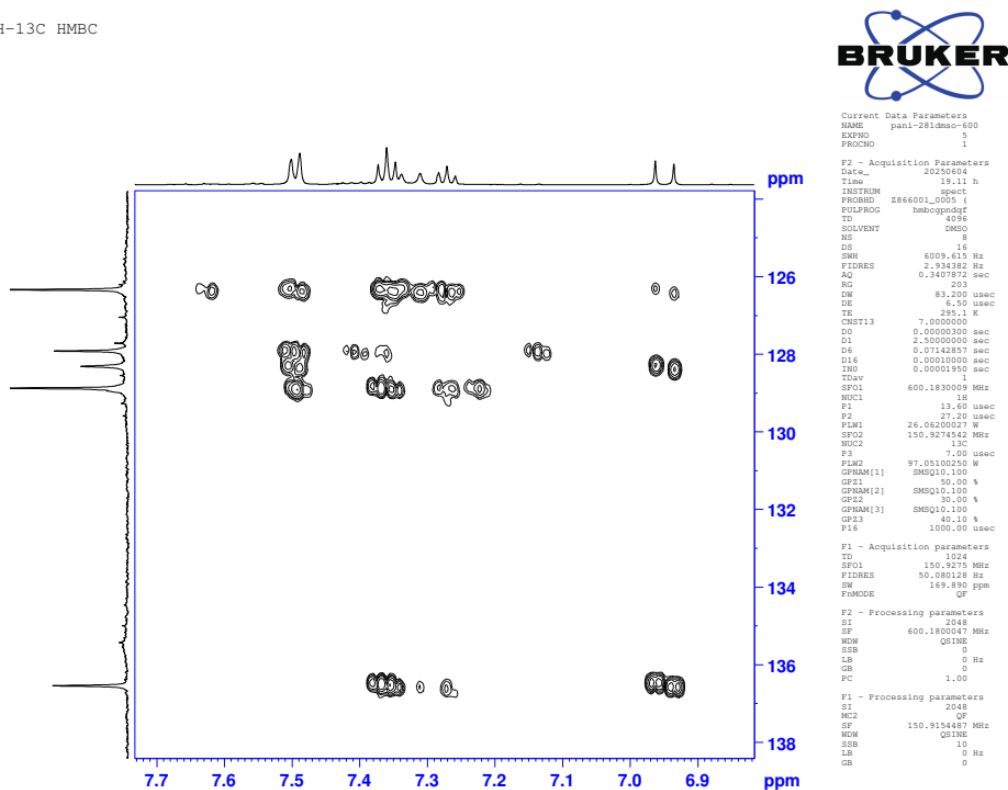
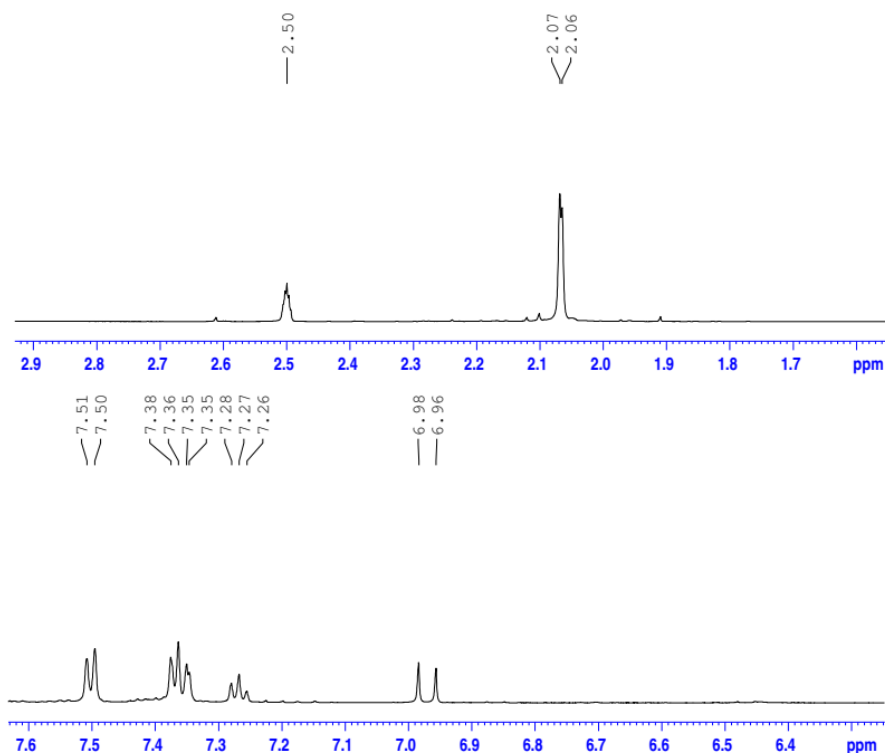


Fig. S 32. Part of ¹H-¹³C HMBC spectrum of compound 3h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆, 23.4°C).

DMSO-d₆, T(true)=49.8C or 322.9K [T(mes)=320.0K]



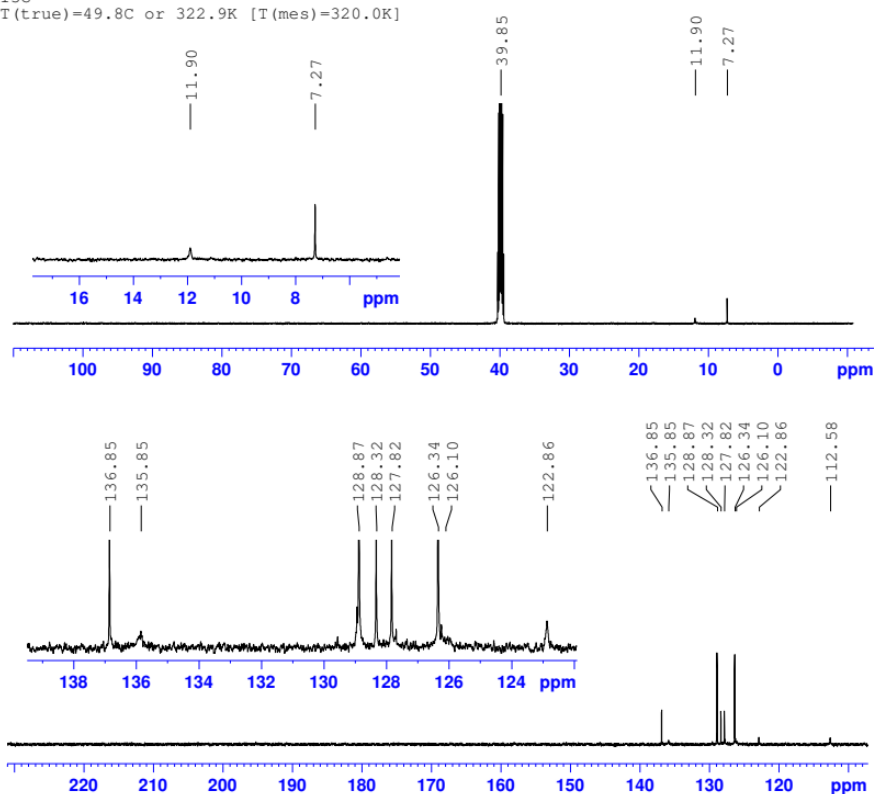
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PROCNO 1

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TD 65536
SOLVENT DMSO
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DS 2
SWH 13227.514 Hz
FIDRES 0.403672 Hz
AQ 2.4772608 sec
RG 203
DW 37.800 usec
DE 6.50 usec
TE 320.0 K
D1 8.00000000 sec
TD0 4
SFO1 600.1854016 MHz
NUC1 1H
P1 13.60 usec
PLW1 26.0620027 W

F2 - Processing parameters
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SF 600.1800048 MHz
WDW EM
SSB 0
LB 0.20 Hz
GB 0
PC 1.00

Fig. S 33. ¹H NMR spectrum of compound 3h (600 MHz, DMSO-d₆, 49.8°C).

¹³C
T(true)=49.8C or 322.9K [T(mes)=320.0K]



Current Data Parameters
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PROCNO 1

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TD 65536
SOLVENT DMSO
NS 1056
DS 0
SWH 37878.789 Hz
FIDRES 1.155969 Hz
AQ 0.8650752 sec
RG 203
DW 13.200 usec
DE 6.50 usec
TE 320.0 K
D1 5.00000000 sec
D11 0.03000000 sec
TD0 100
SFO1 150.9327363 MHz
NUC1 13C
P1 7.00 usec
PLW1 97.05100250 W
SFO2 600.1824007 MHz
NUC2 1H
CPDPRG2 waltz16
PCPD2 70.00 usec
PLW2 26.0620027 W
PLW12 0.98373997 W

F2 - Processing parameters
SI 131072
SF 150.9154396 MHz
WDW EM
SSB 0
LB 2.00 Hz
GB 0
PC 1.40

Fig. S 34. ¹³C NMR spectrum of compound 3h (151 MHz, DMSO-d₆, 49.8°C) calibrated by 3'' signal of phenyl moiety, its chemical shift taken equal to 128.97 ppm as at 22.4°C.

¹H-¹³C HMBC
T(true)=49.8C or 322.9K [T(mes)=320.0K]

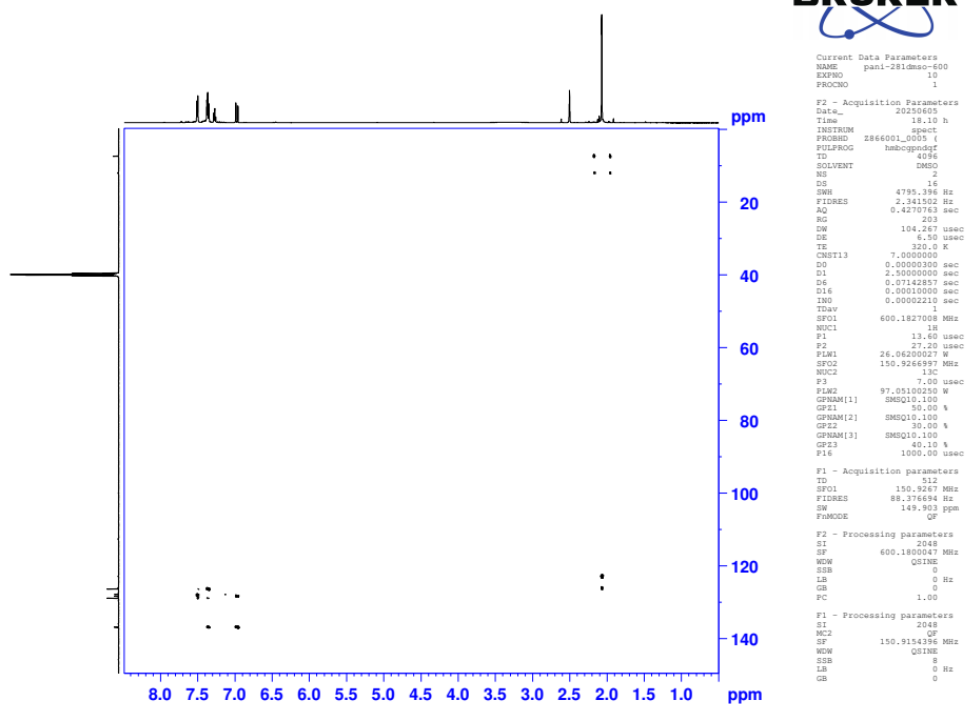


Fig. S 35. ¹H-¹³C HMBC spectrum of compound 3h (600 MHz – ¹H, 151 MHz – ¹³C, DMSO-*d*₆, 49.8°C).

¹H-¹³C HMBC
T(true)=49.8C or 322.9K [T(mes)=320.0K]

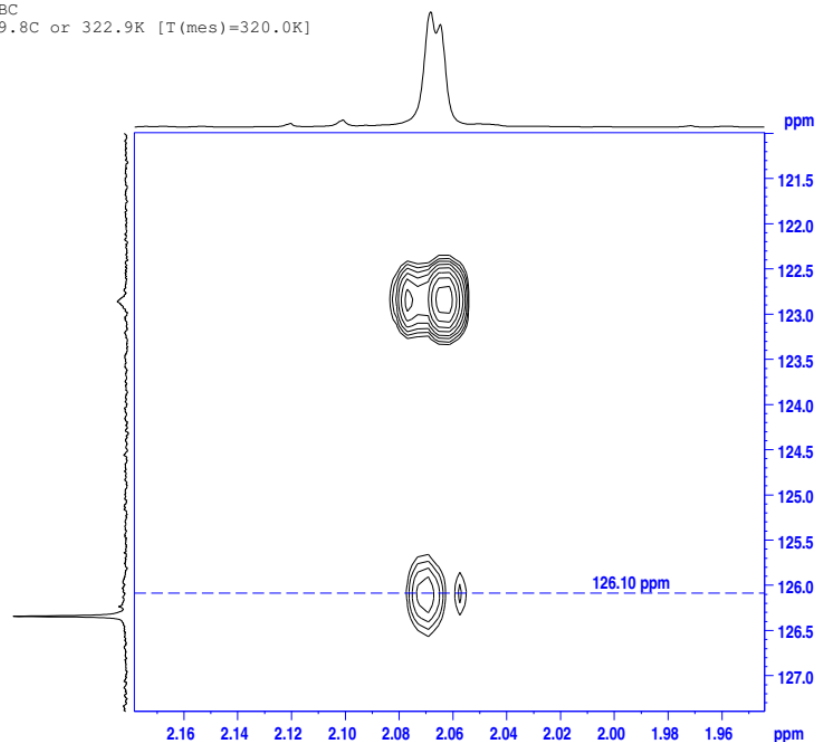


Fig. S 36. Part of ¹H-¹³C HMBC spectrum of compound 3h (600 MHz – ¹H, 151 MHz – ¹³C, DMSO-*d*₆, 49.8°C).

^1H - ^{13}C HMBC
T(true)=49.8C or 322.9K [T(mes)=320.0K]

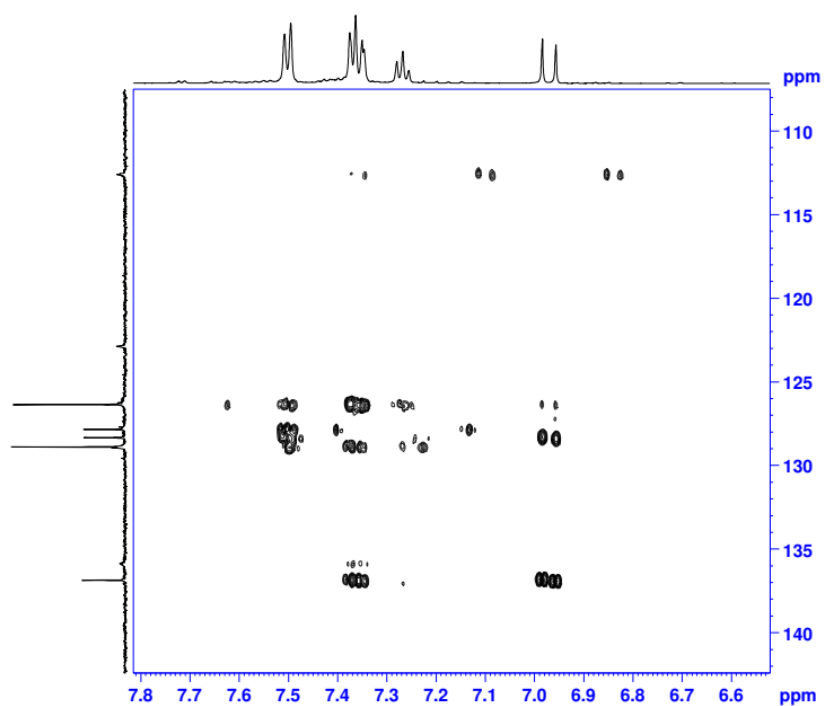


Fig. S 37. Part of ^1H - ^{13}C HMBC spectrum of compound 3h (600 MHz - ^1H , 151 MHz - ^{13}C , $\text{DMSO}-d_6$, 49.8°C).

S4.9. 3-Methoxy-6,6-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-3,5,6,7-tetrahydro-4H-benzimidazol-4-one (**4a**).

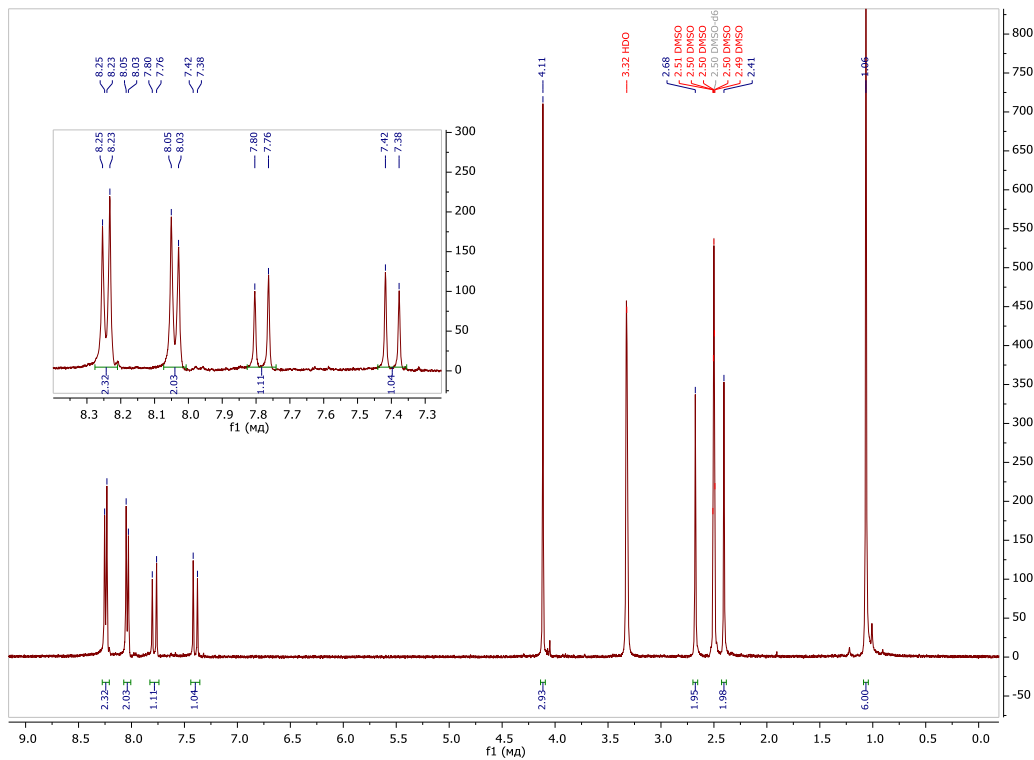
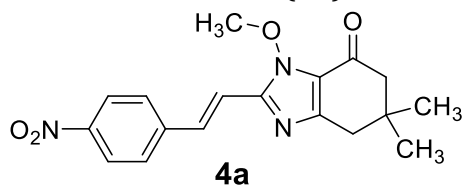


Fig. S 38. ¹H NMR spectrum of compound **4a** (400 MHz, DMSO-*d*₆).

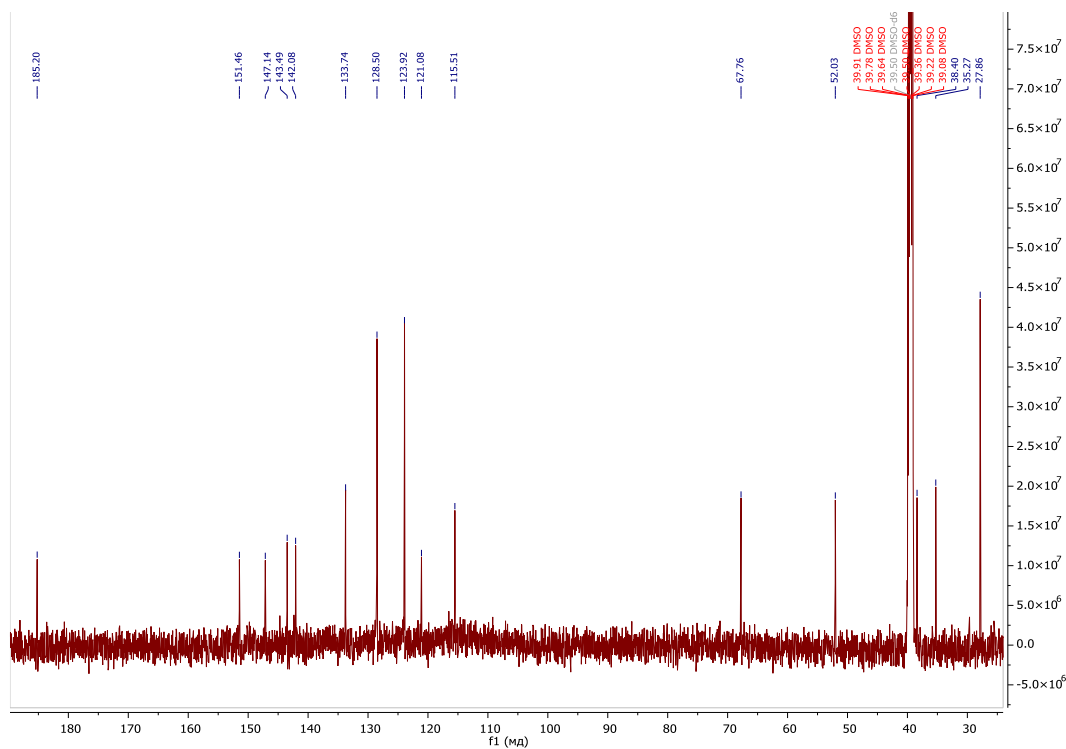


Fig. S 39. ¹³C NMR spectrum of compound **4a** (151 MHz, DMSO-*d*₆).

S4.10. 1-(1-Methoxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazol-5-yl)ethanone (**4b**).

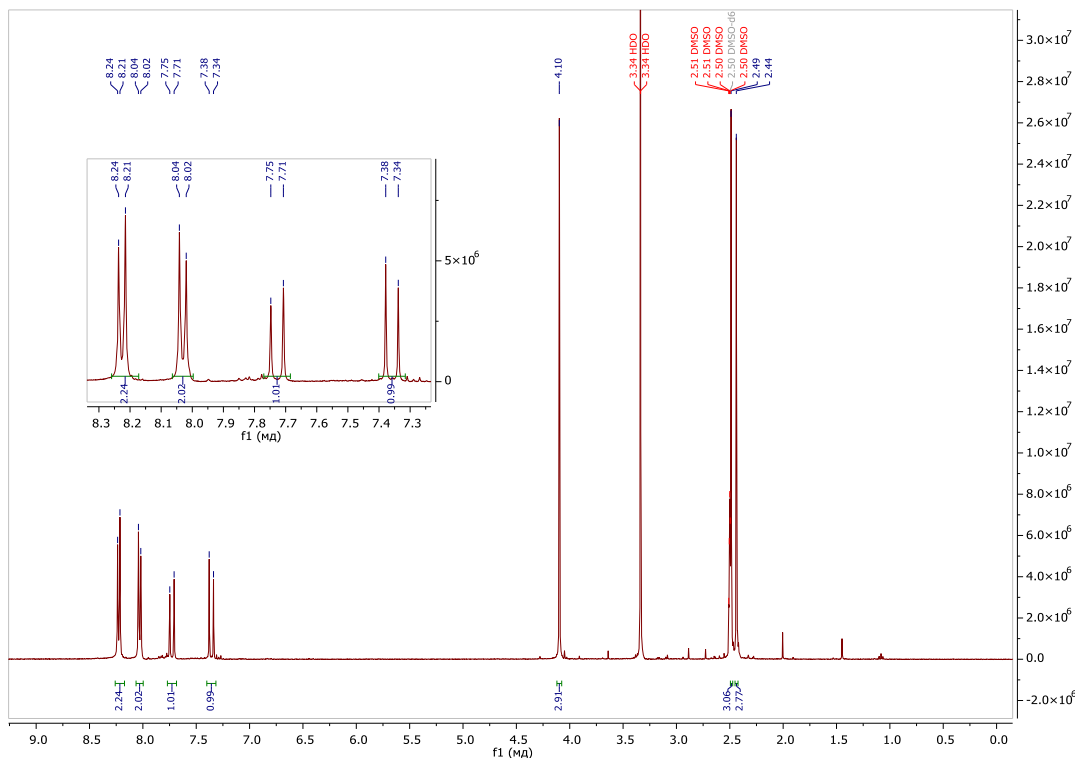
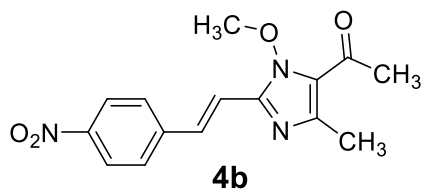


Fig. S 40. ¹H NMR spectrum of compound **4b** (400 MHz, DMSO-*d*₆).

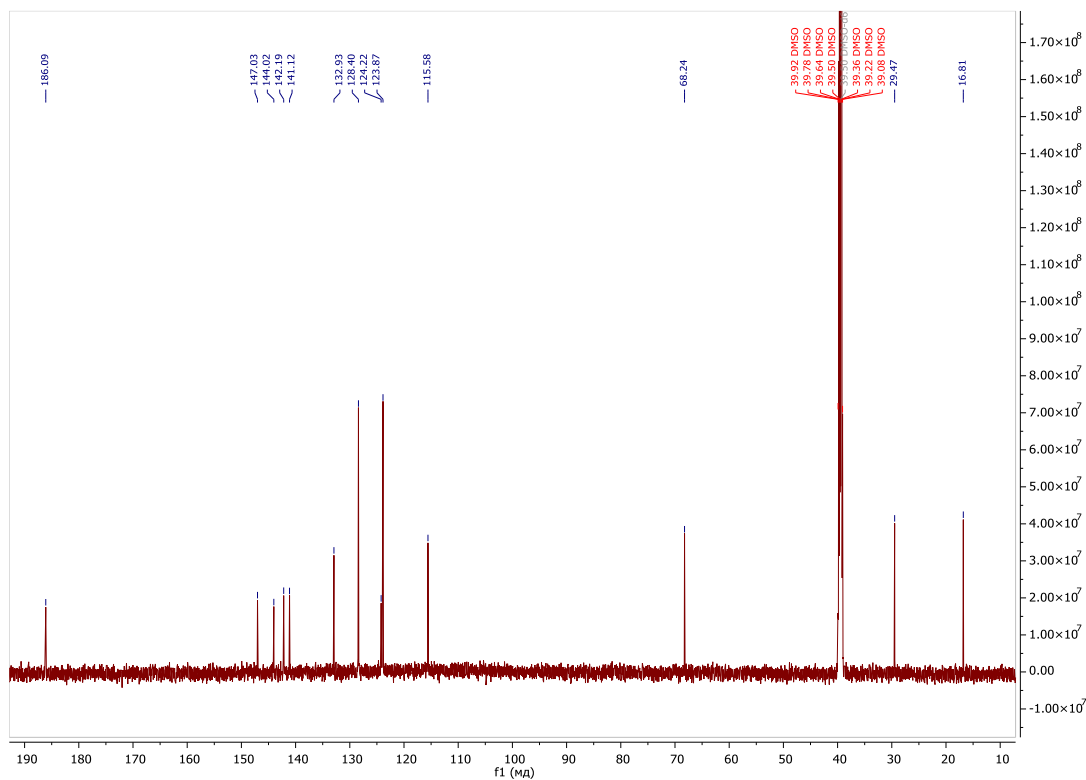


Fig. S 41. ¹³C NMR spectrum of compound **4b** (151 MHz, DMSO-*d*₆).

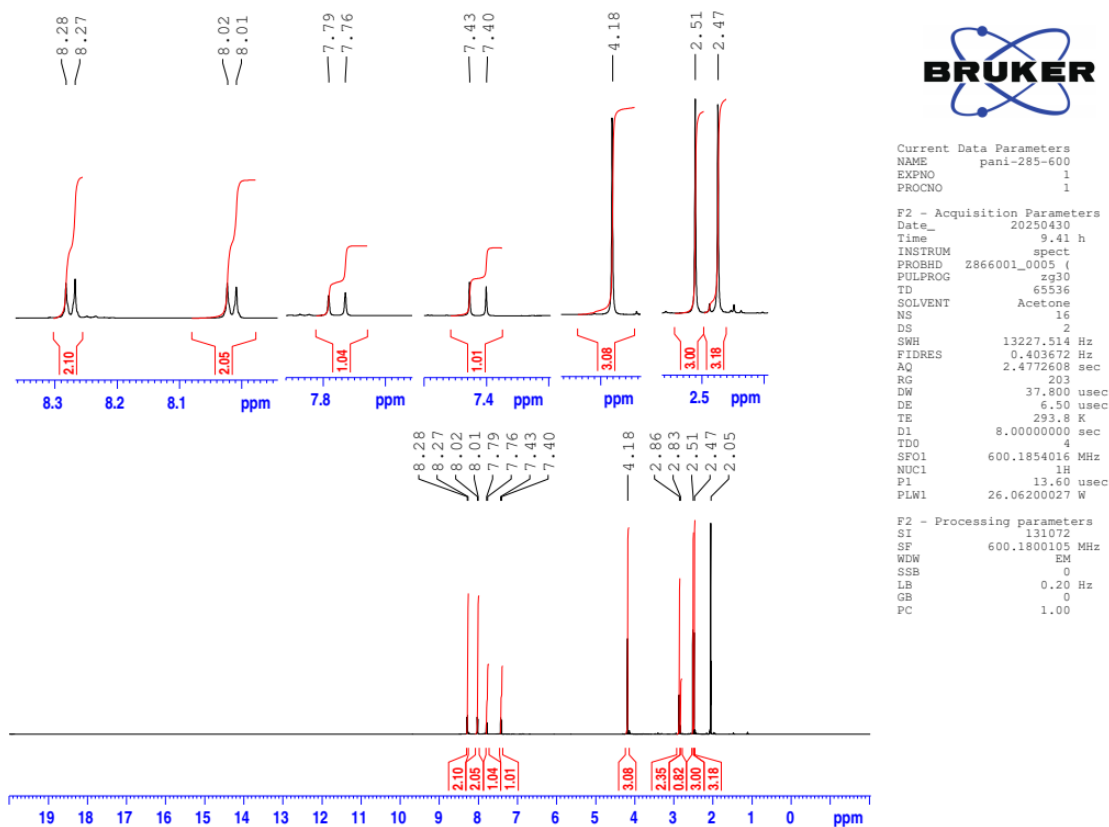


Fig. S 42. ^1H NMR spectrum of compound 4b (600 MHz, acetone- d_6).

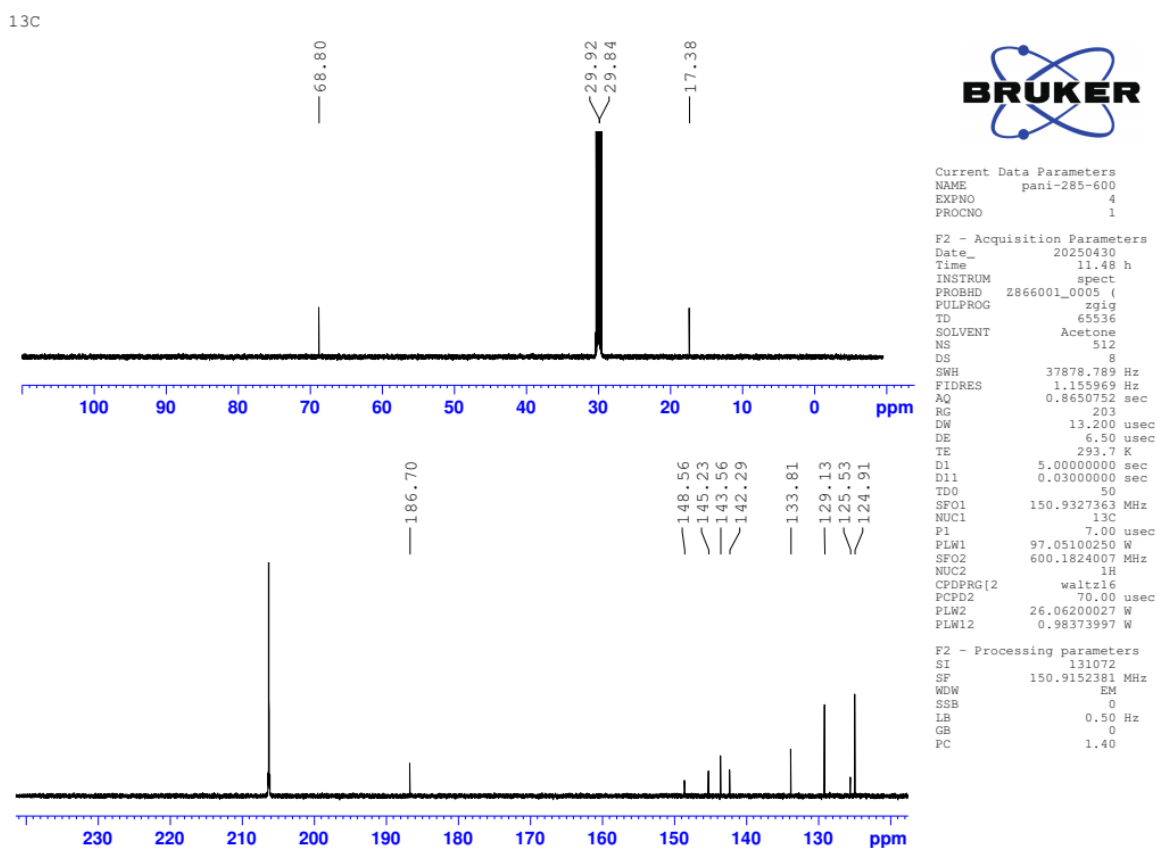


Fig. S 43. ^{13}C NMR spectrum of compound 4b (151 MHz, acetone- d_6).

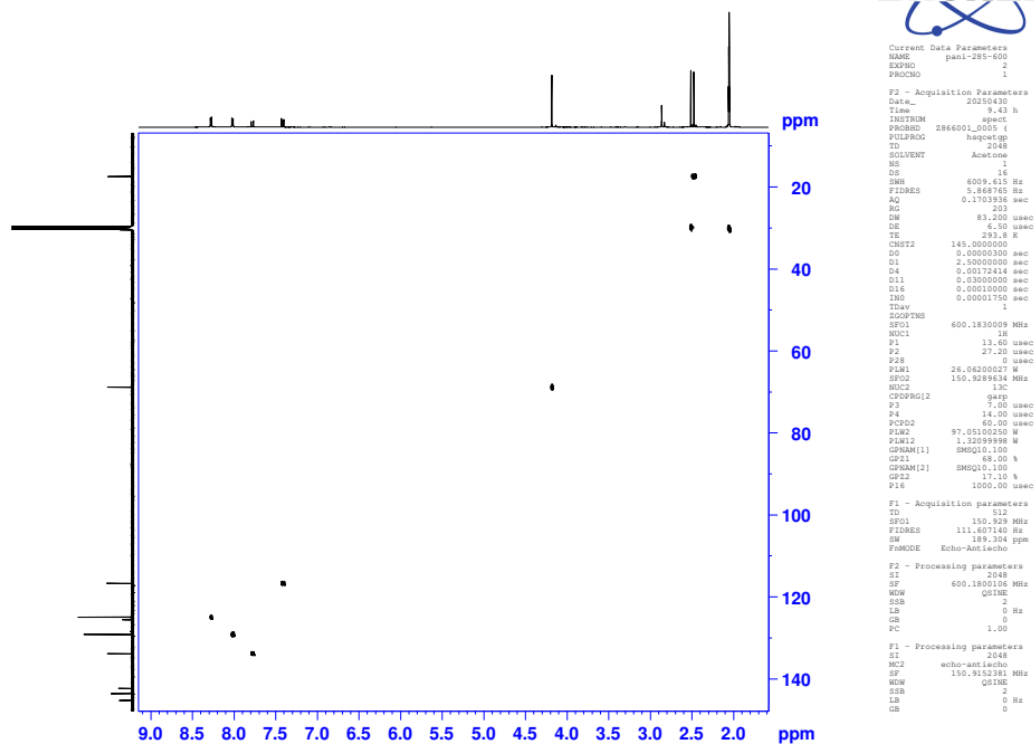


Fig. S 44. ^1H - ^{13}C HSQC spectrum of compound 4b (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

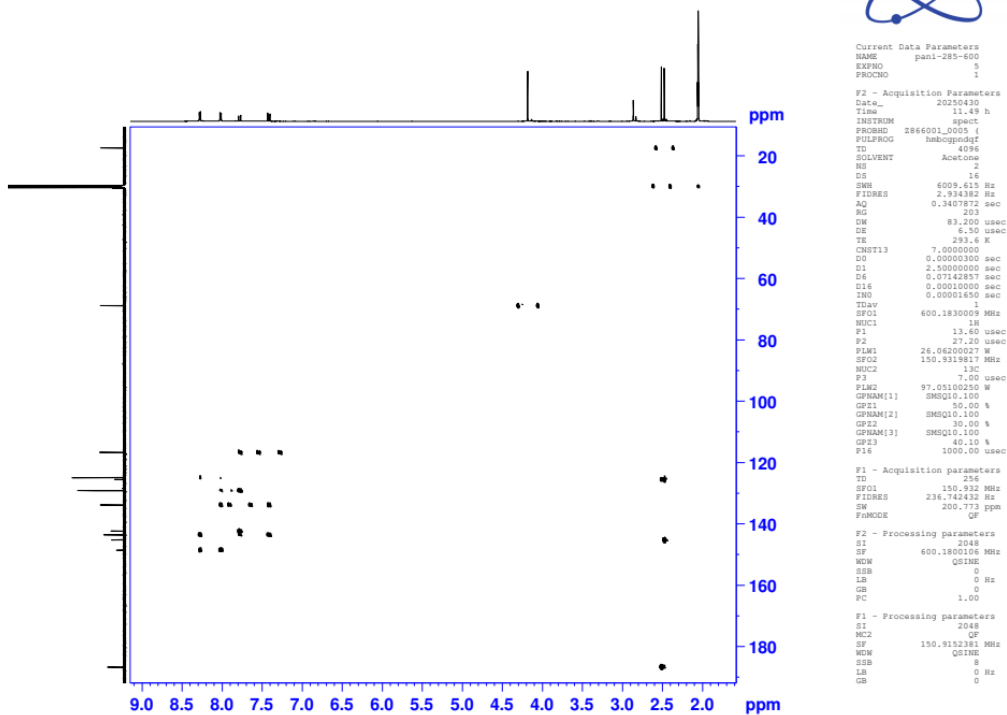


Fig. S 48. ^1H - ^{13}C HMBC spectrum of compound 4b (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

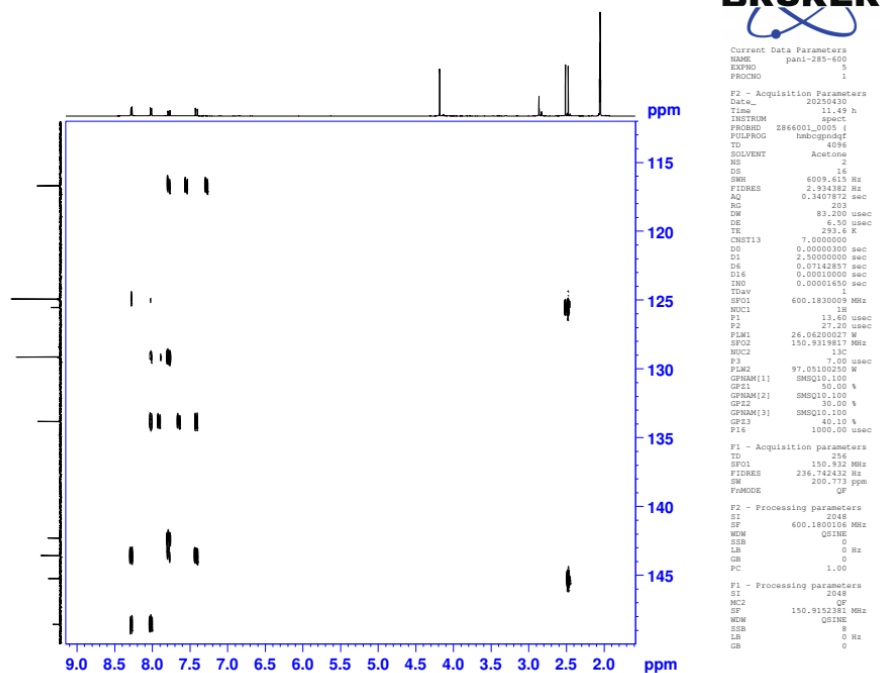


Fig. S 45. Part of ¹H-¹³C HMBC spectrum of compound 4b (600 MHz - ¹H, 151 MHz - ¹³C, acetone-*d*₆).

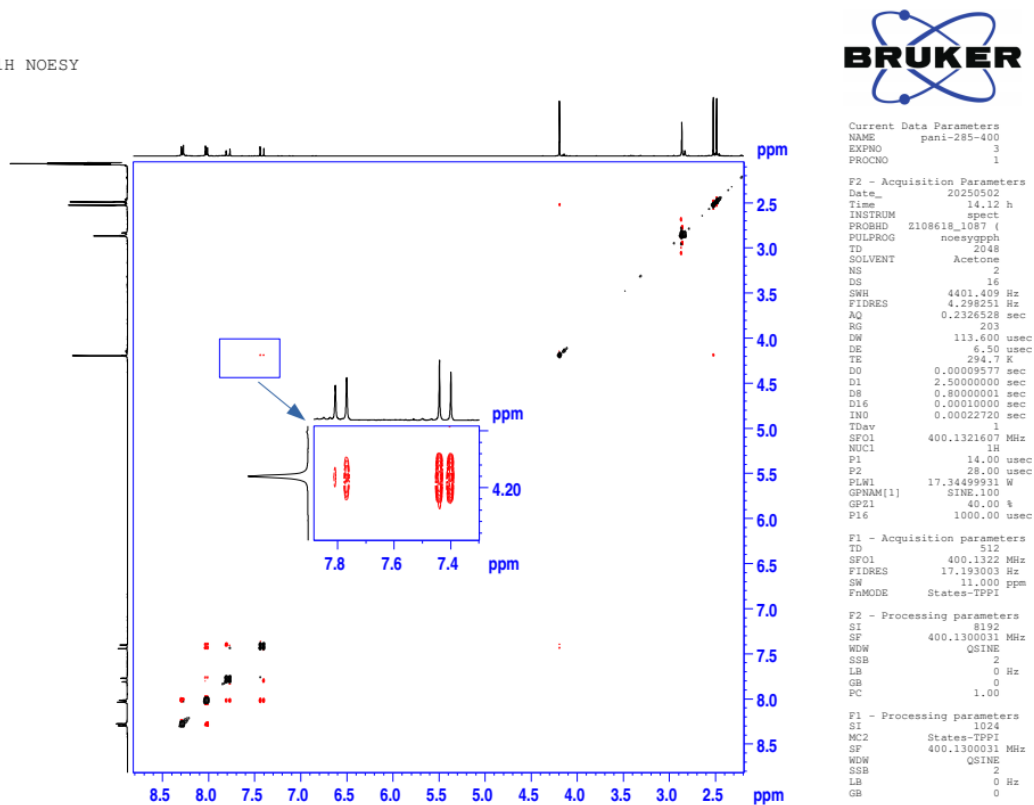


Fig. S 46. ¹H-¹H NOESY spectrum of compound 4b (400 MHz, acetone-*d*₆).

¹H-¹⁵N HMBC



Current Data Parameters
NAME pani-281-400
EXPNO 2
PROCNO 1

F2 - Acquisition Parameters
Date_ 20030502
Time 10.32 h
INSTRUM spect
PROBHD Z108618_1087 (hmbcgpndgt)
PULPROG hmbcgpndgt
TD 65536
SOLVENT Acetone
NS 8
DS 16
SWH 4000.000 Hz
FIDRES 1.931125 Hz
AQ 0.5120000 sec
RG 200
DM 125.000 usec
DE 6.50 usec
TE 295.1 K
CNS113 3.0000000
DO 0.0000000 sec
D1 2.50000000 sec
D6 0.16666670 sec
d16 0.00010000 sec
INO 0.00004400 sec
TDAY
SFO1 400.1320007 MHz
NUC1 15
P1 14.00 usec
P2 28.00 usec
PLM1 17.34499931 W
SFO2 40.5513982 MHz
NUC2 15N
P3 18.00 usec
PLM2 78.47599792 W
GPAM[1] SINE,100
GP1 70.00 %
GPAM[2] SINE,100
GP2 30.00 %
GPAM[3] SINE,100
GP3 50.10 %
P16 1000.00 usec

F1 - Acquisition parameters
TD 512
SFO1 40.55131 MHz
FIDRES 44.389206 Hz
SW 280.229 ppm
FIRMODE QF

F2 - Processing parameters
SI 1024
SF 400.1300031 MHz
WDW Q5INE
SSB 0
LB 0 Hz
GB 0
PC 1.00

F1 - Processing parameters
SI 1024
NUC2 QF
SF 40.5448210 MHz
WDW Q5INE
SSB 0
LB 0 Hz
GB 0

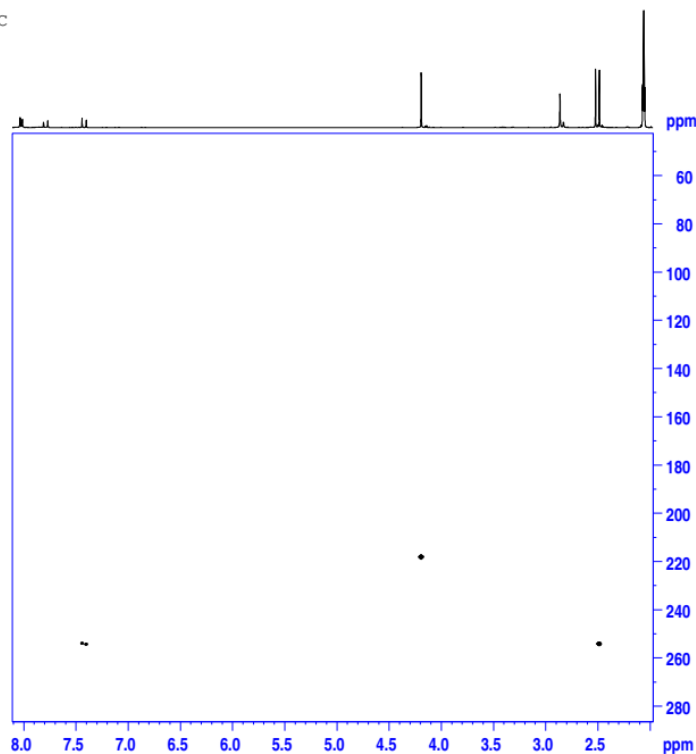


Fig. S 47. ¹H-¹⁵N HMBC spectrum of compound 4b (400 MHz – ¹H, 41 MHz – ¹⁵N, acetone-*d*₆).

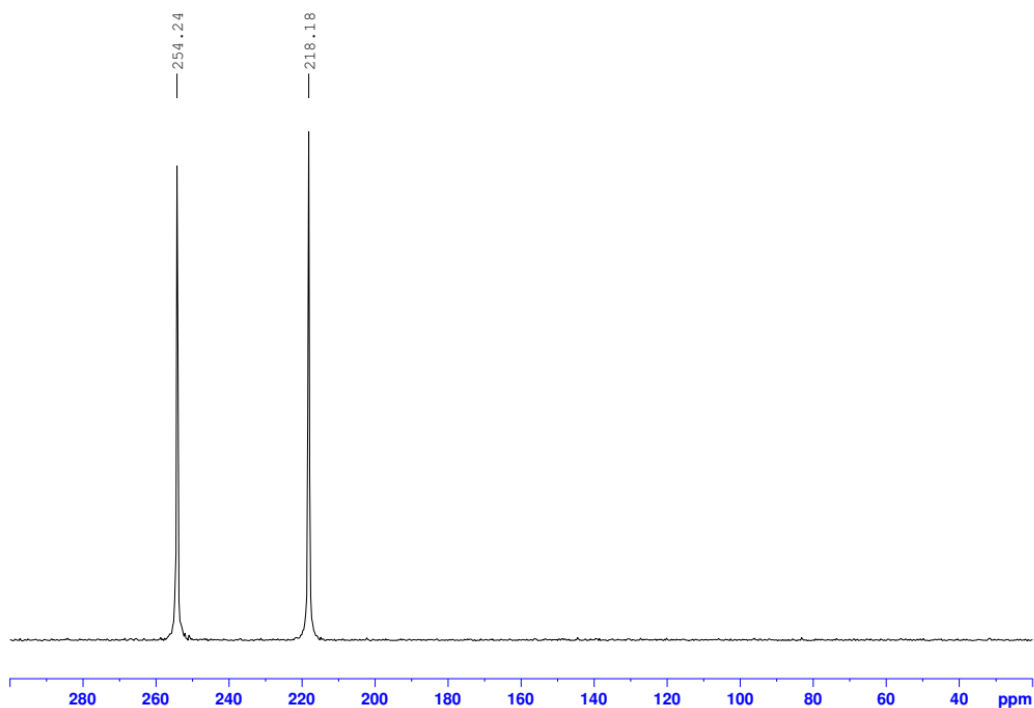


Fig. S 48. ¹⁵N NMR spectrum of compound 4b (41 MHz, acetone-*d*₆), obtained by projection from ¹H-¹⁵N HMBC spectrum.

S4.11. Ethyl 1-methoxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole-5-carboxylate (**4c**).

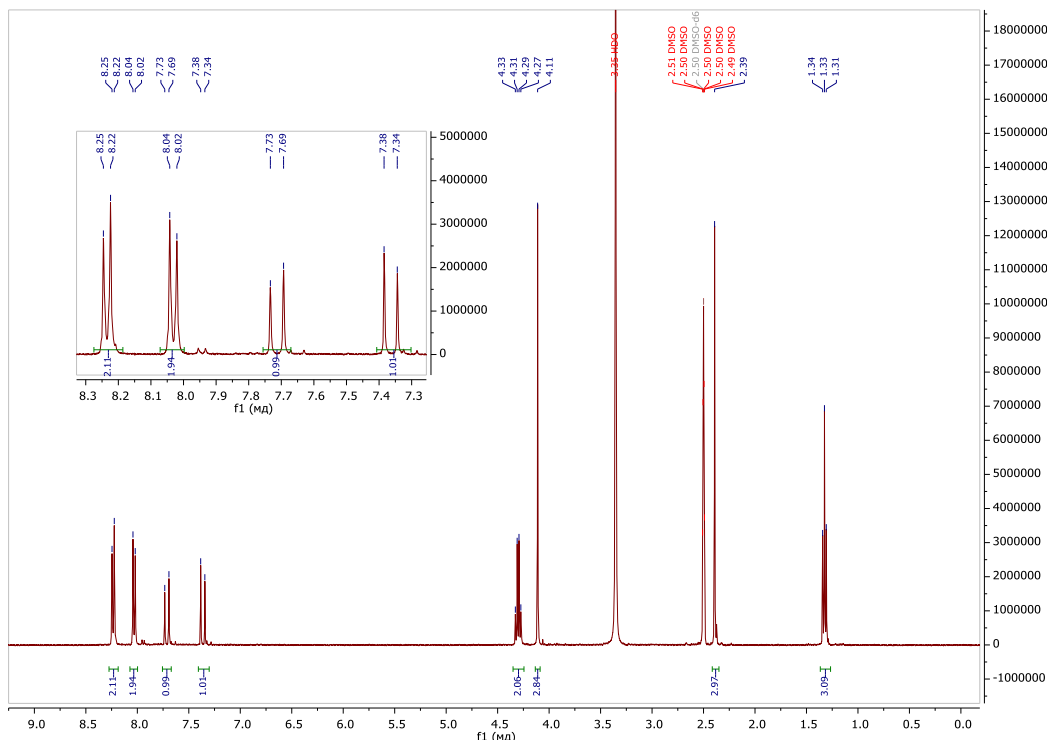
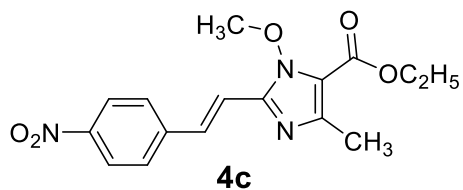


Fig. S 49. ¹H NMR spectrum of compound **4c** (400 MHz, DMSO-*d*₆).

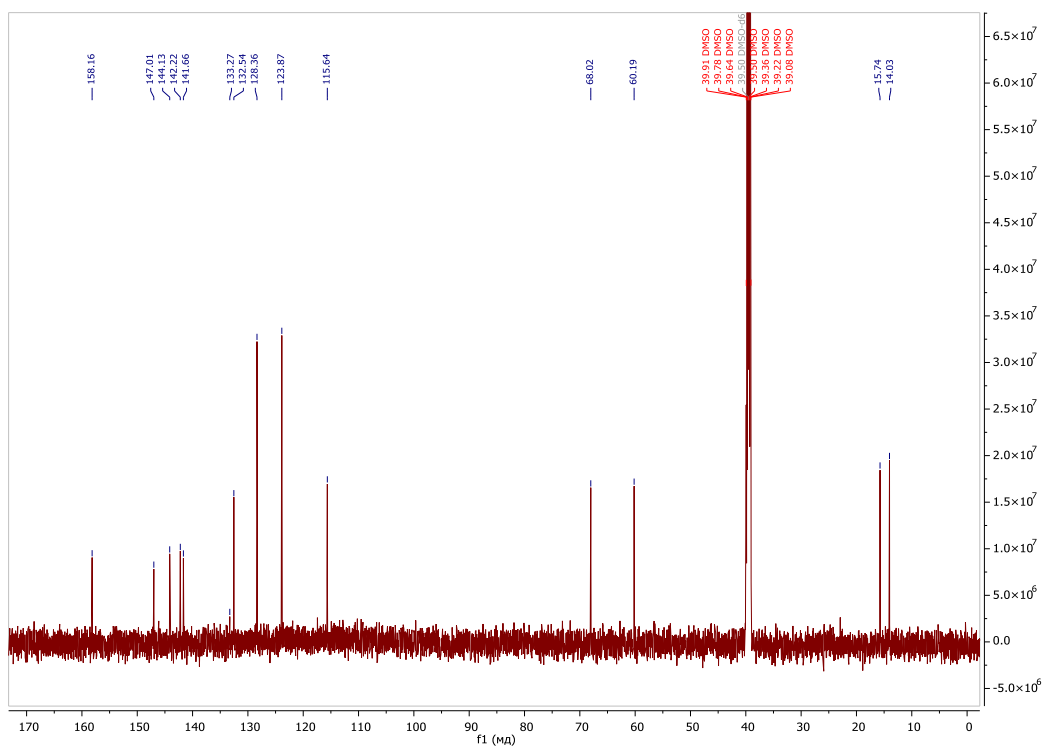


Fig. S 50. ¹³C NMR spectrum of compound **4c** (151 MHz, DMSO-*d*₆).

S4.12. 1-Methoxy-4,5-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole (**4d**).

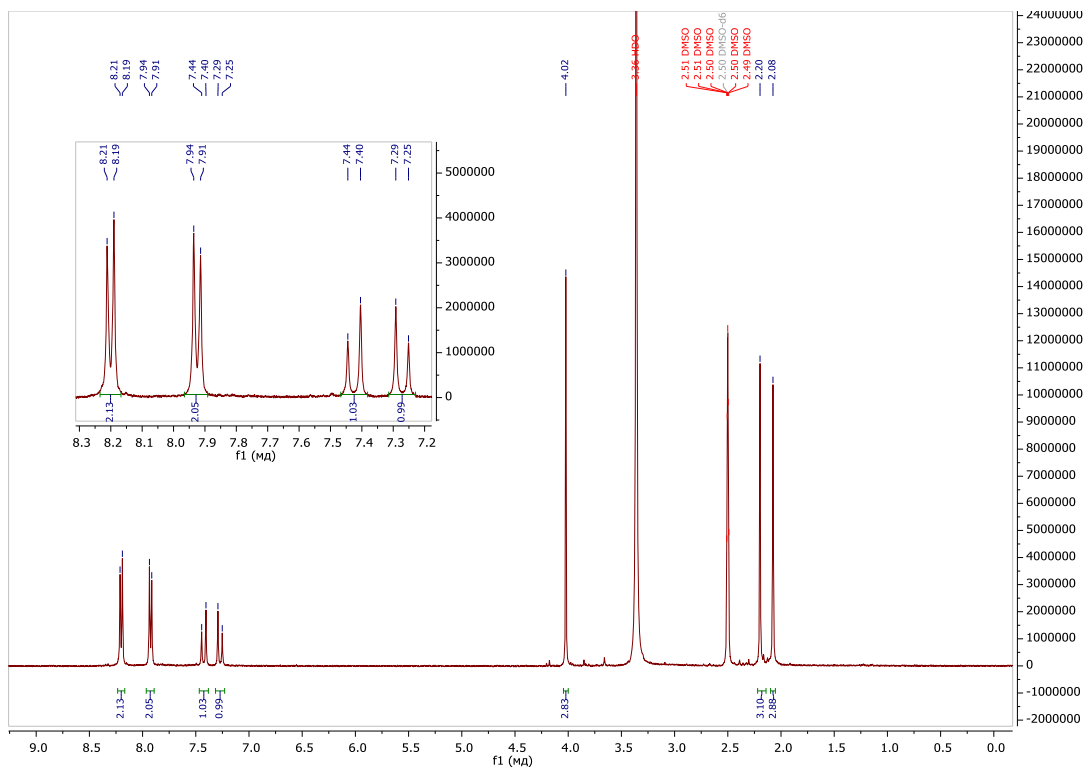
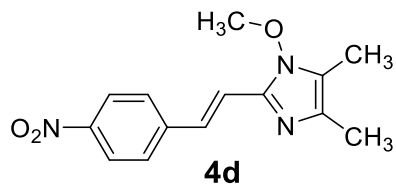


Fig. S 51. ¹H NMR spectrum of compound 4d (400 MHz, DMSO-*d*₆).

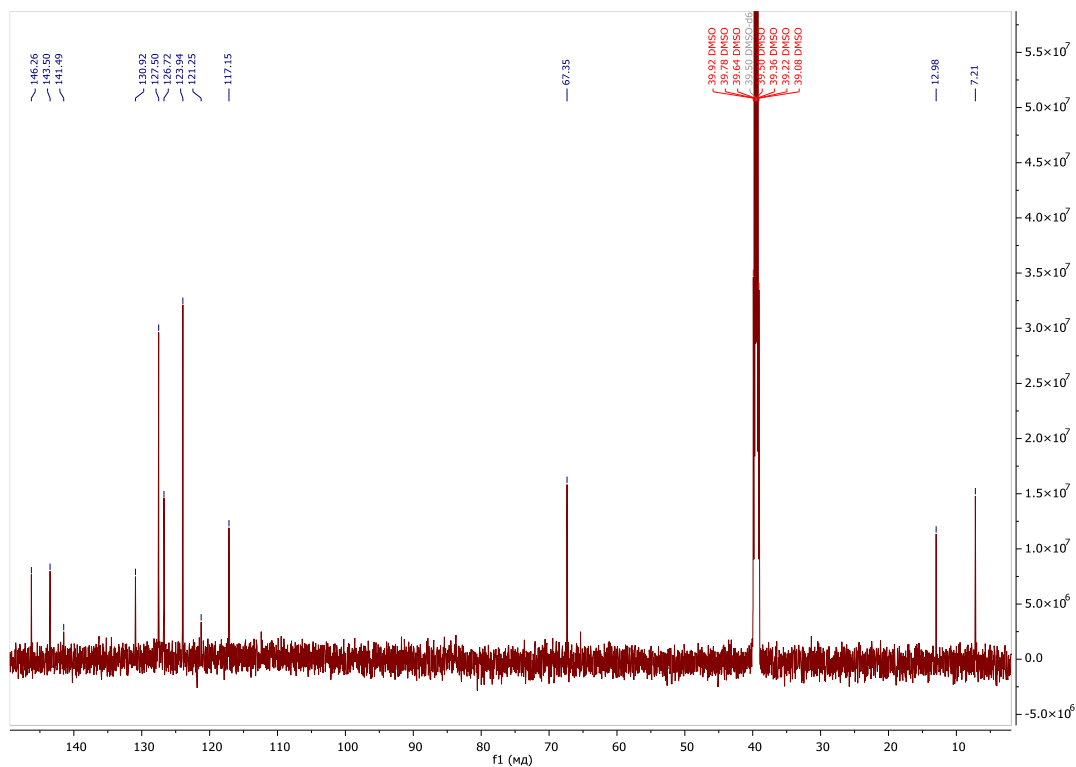


Fig. S 52. ¹³C NMR spectrum of compound 4d (151 MHz, DMSO-*d*₆).

S4.13. 3-Methoxy-6,6-dimethyl-2-((*E*)-2-phenylethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (**4e**).

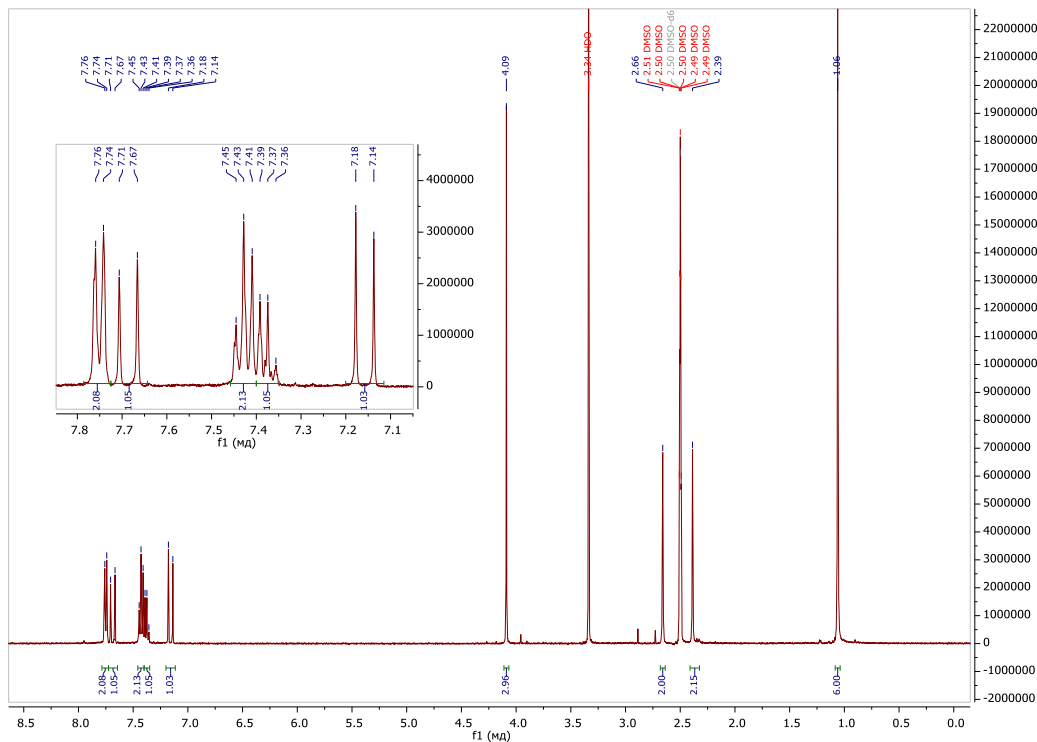
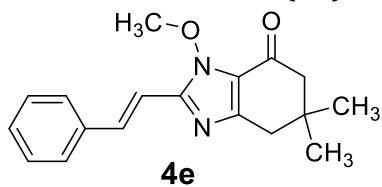


Fig. S 53. ^1H NMR spectrum of compound **4e** (400 MHz, $\text{DMSO}-d_6$).

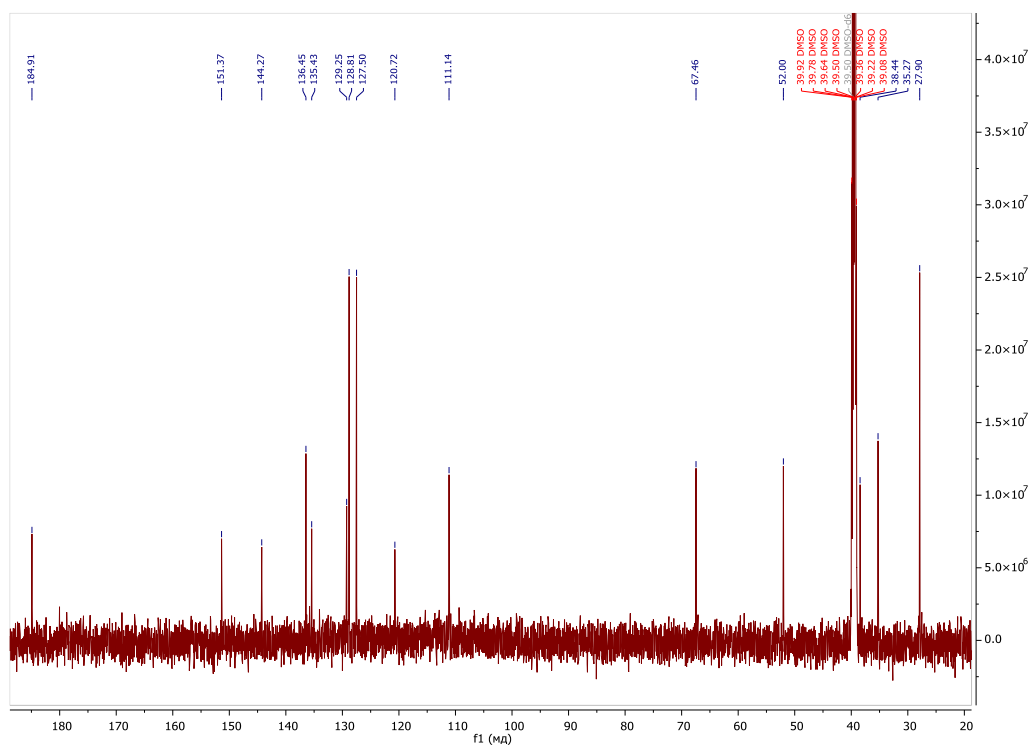


Fig. S 58. ^{13}C NMR spectrum of compound **4e** (151 MHz, $\text{DMSO}-d_6$).

S4.14. 1-(1-Methoxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazol-5-yl)ethanone (**4f**).

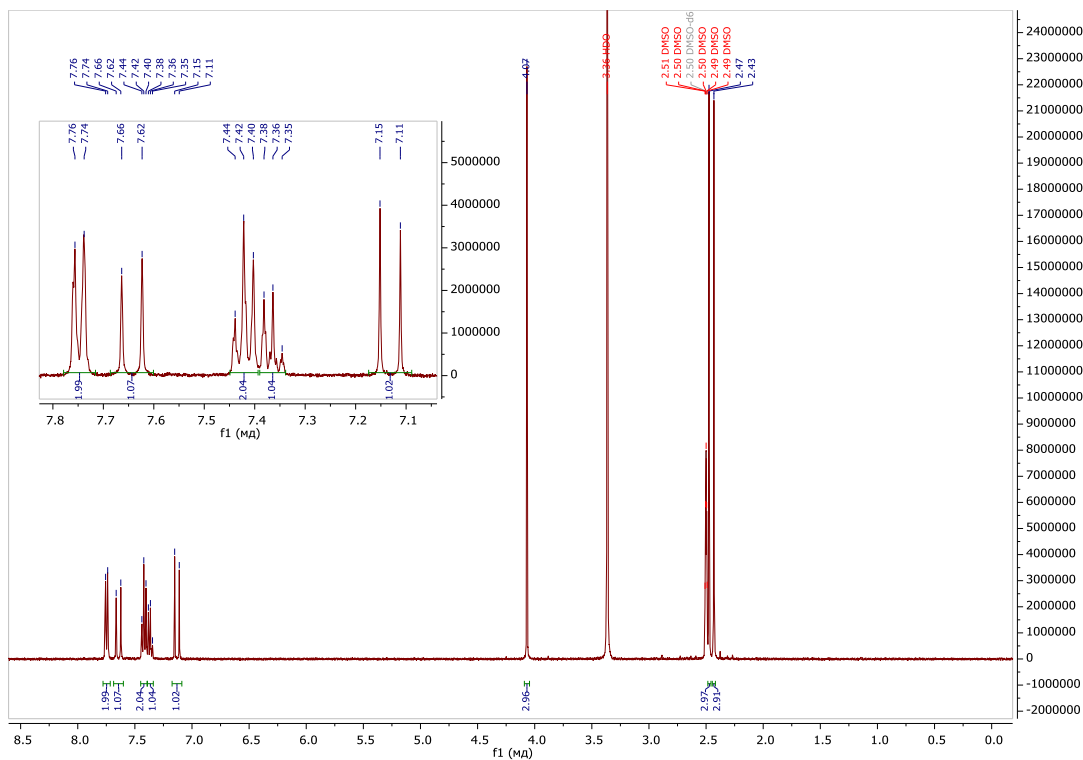
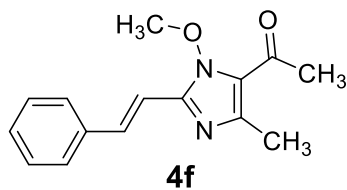


Fig. S 59. ¹H NMR spectrum of compound **4f** (400 MHz, DMSO-*d*₆).

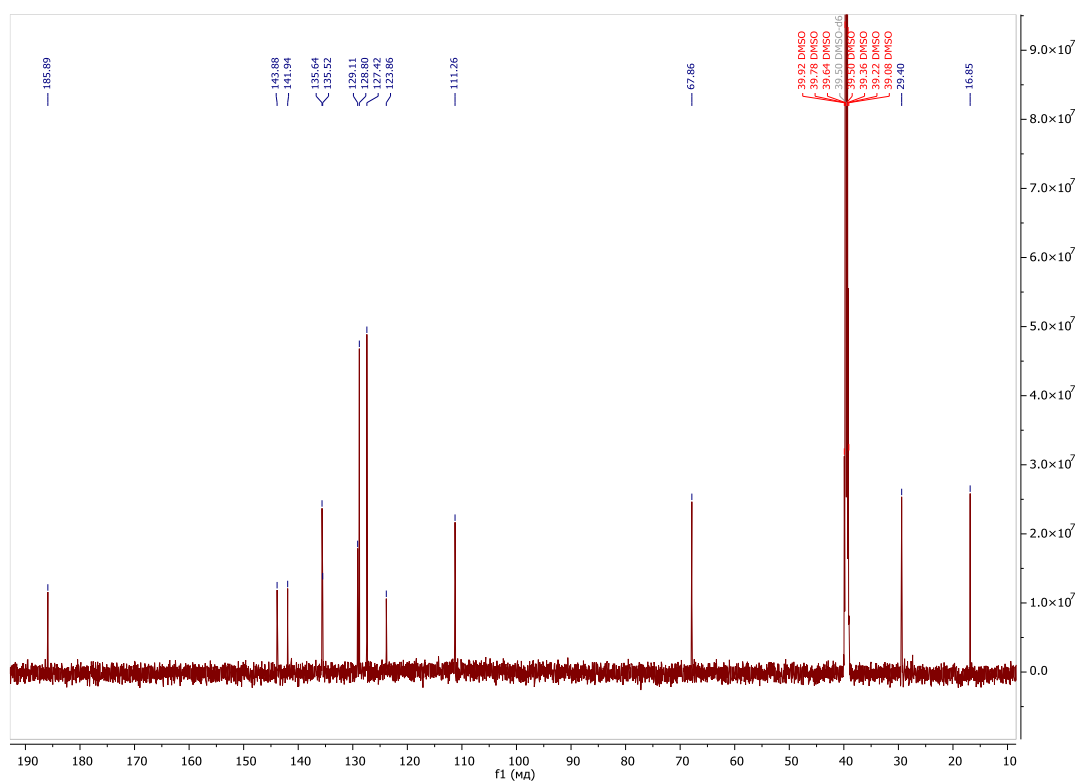


Fig. S 54. ¹³C NMR spectrum of compound **4f** (151 MHz, DMSO-*d*₆).

S4.15. Ethyl 1-methoxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole-5-carboxylate (**4g**).

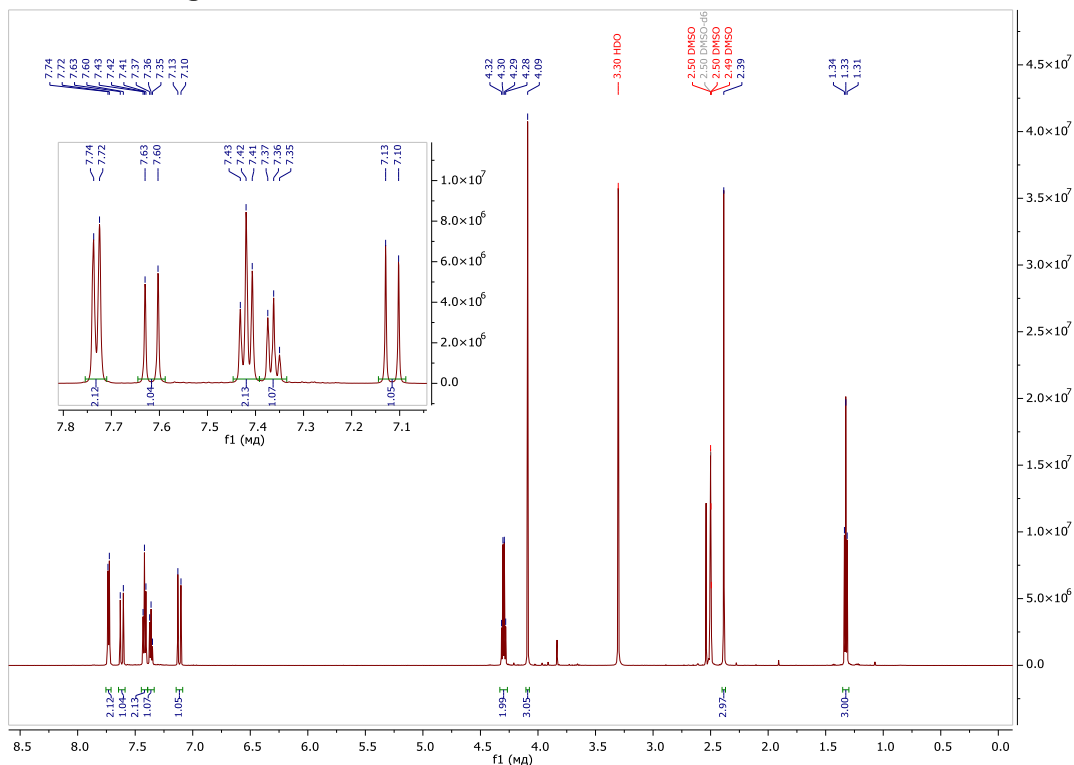
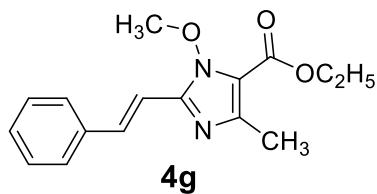


Fig. S 55. ¹H NMR spectrum of compound **4g** (600 MHz, DMSO-*d*₆).

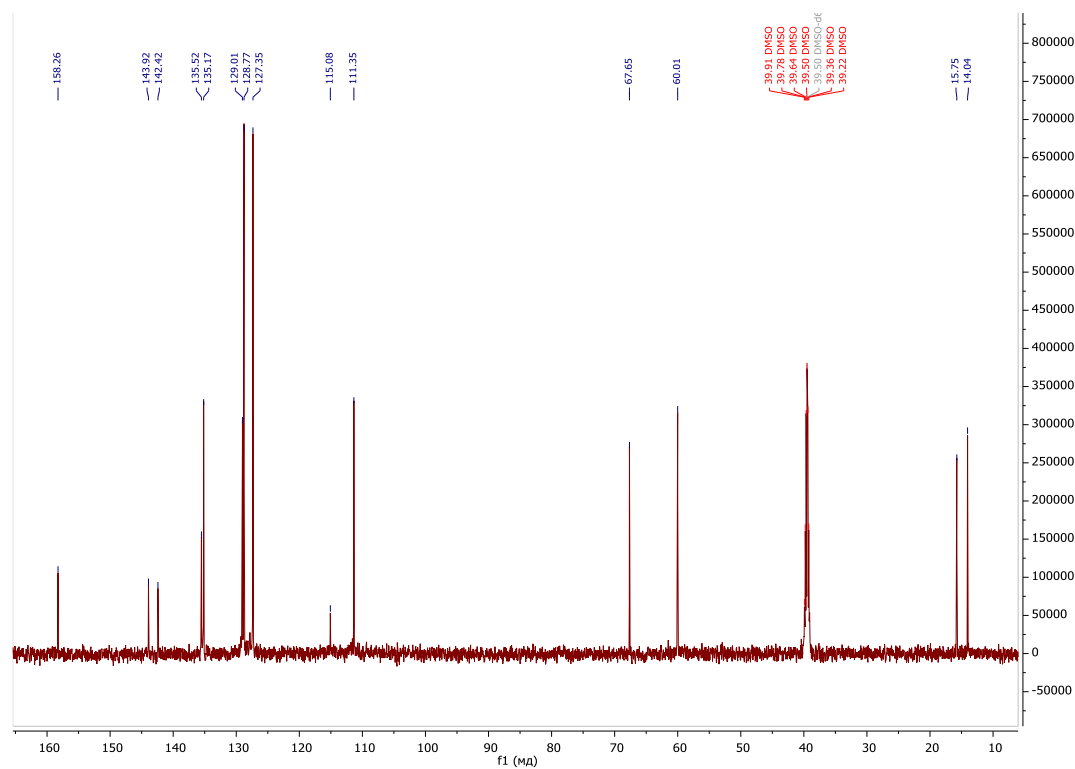
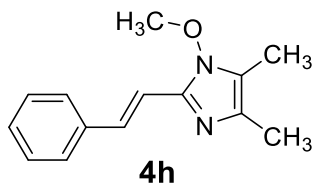


Fig. S 56. ¹³C NMR spectrum of compound **4g** (151 MHz, DMSO-*d*₆).

S4.16. 1-Methoxy-4,5-dimethyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole (**4h**).



PANI-288; DMSO-*d*₆

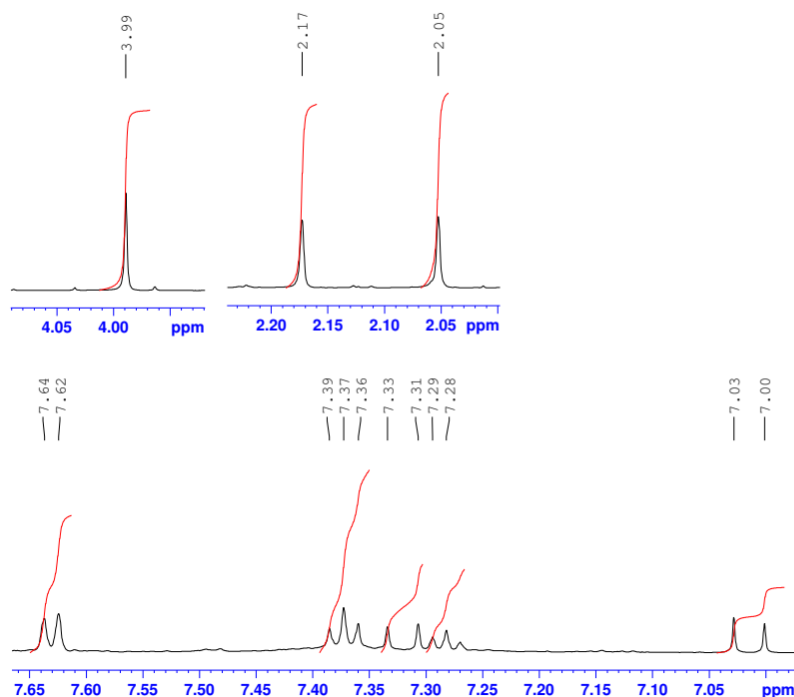


Fig. S 63. ¹H NMR spectrum of compound 4h (600 MHz, DMSO-*d*₆).

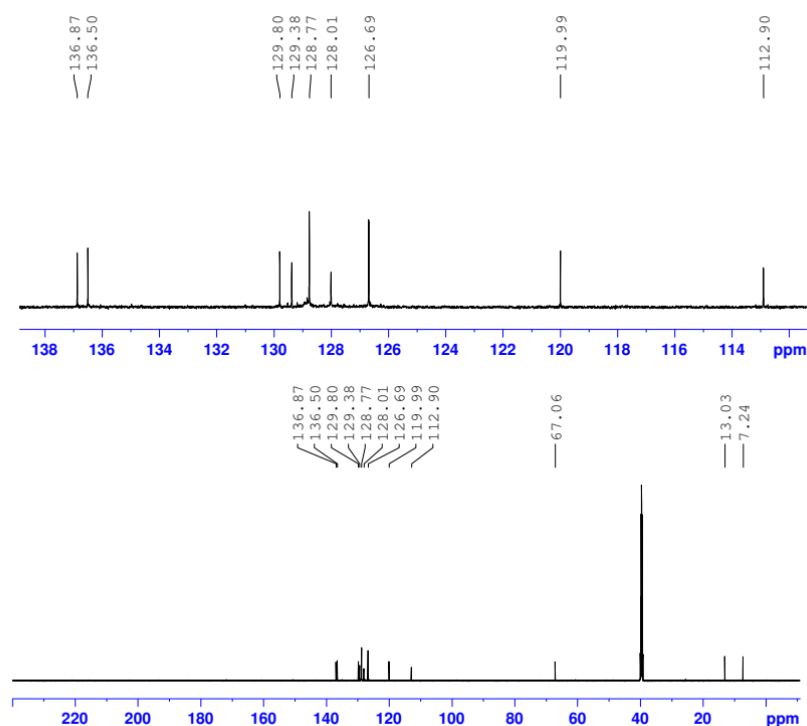


Fig. S 64. ¹³C NMR spectrum of compound 4h (151 MHz, DMSO-*d*₆).

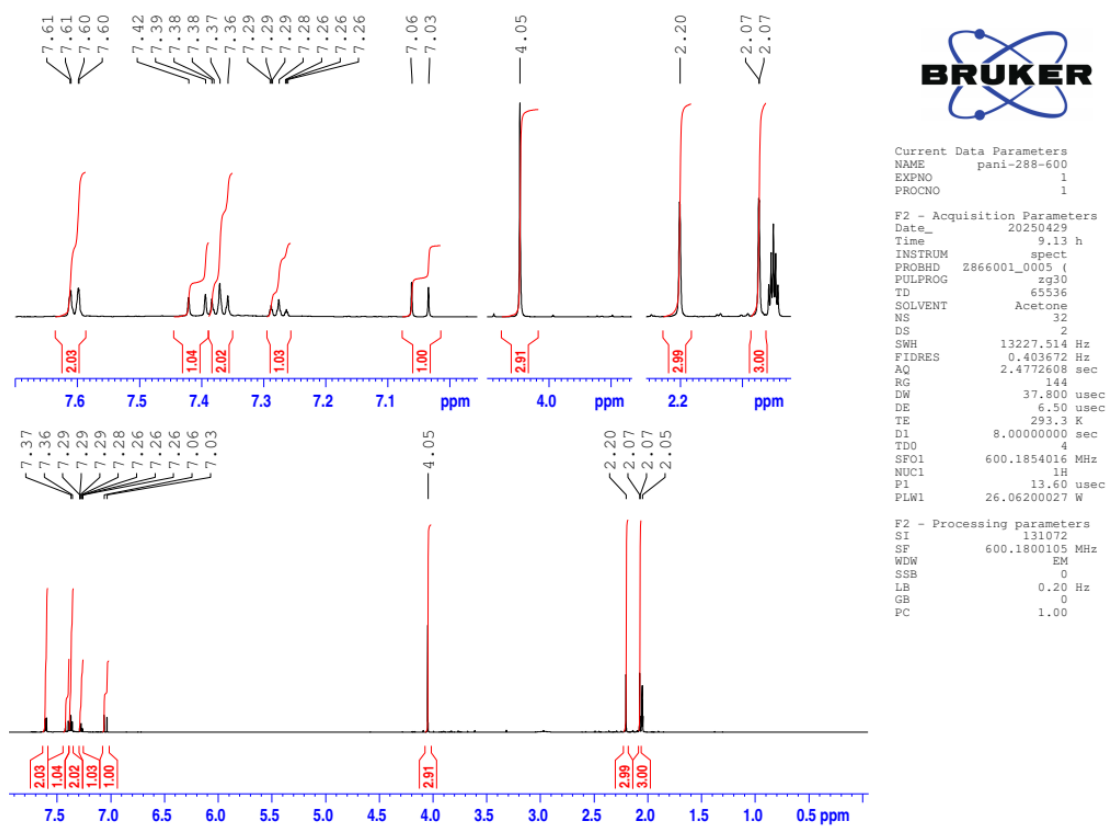


Fig. S 57. ^1H NMR spectrum of compound 4h (600 MHz, acetone- d_6).

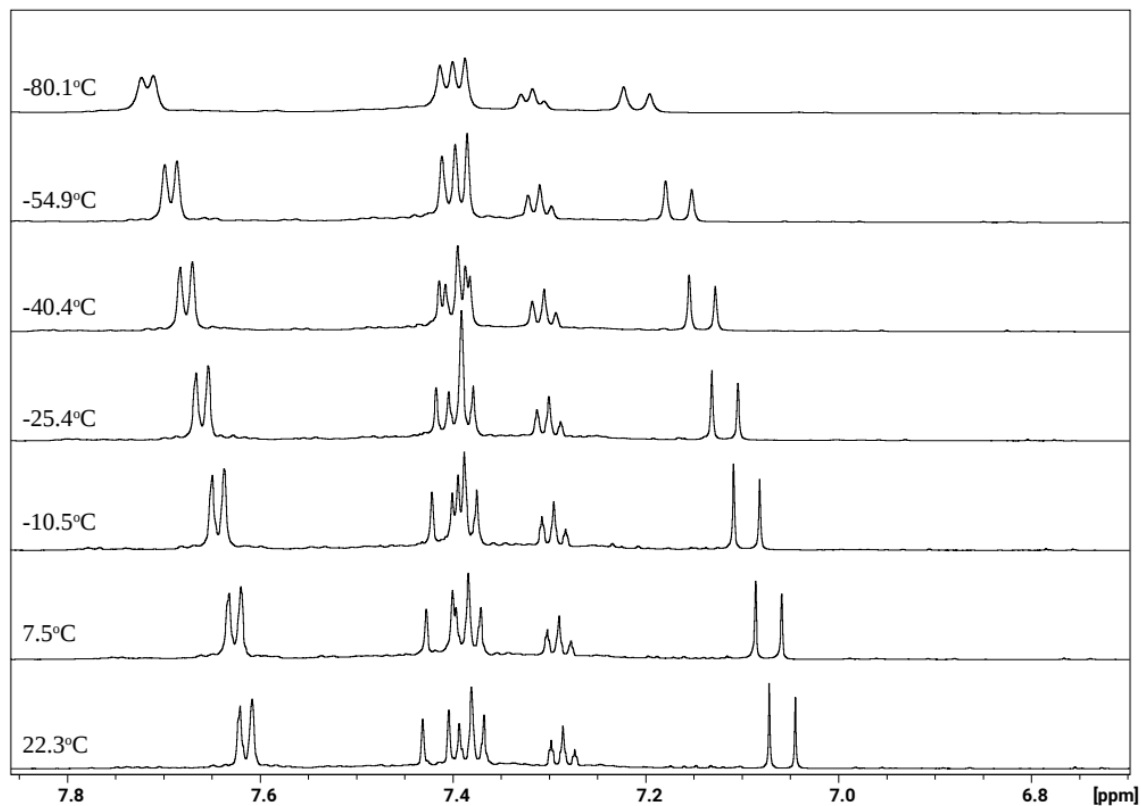


Fig. S 58. ^1H NMR spectra of compound 4h registered at different temperatures (600 MHz, acetone- d_6 , range 7.0-9.0 ppm).

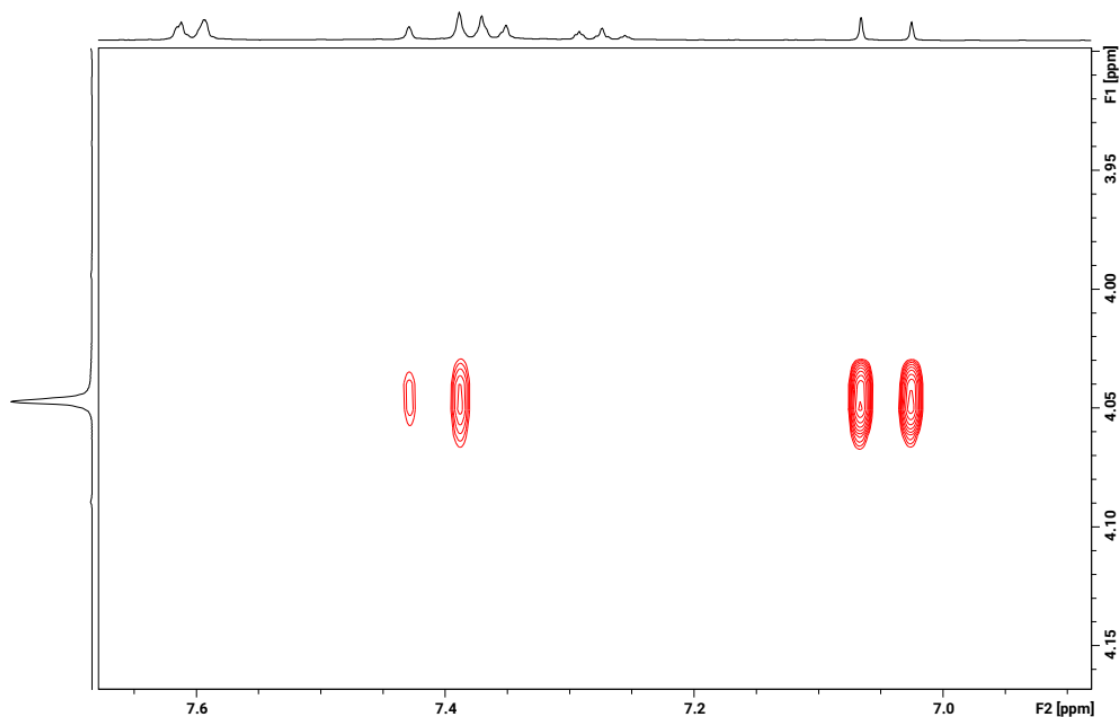


Fig. S 59. Part of ^1H - ^1H NOESY spectrum of compound 4h (400 MHz, acetone- d_6 , mixing time – 0.8 s).

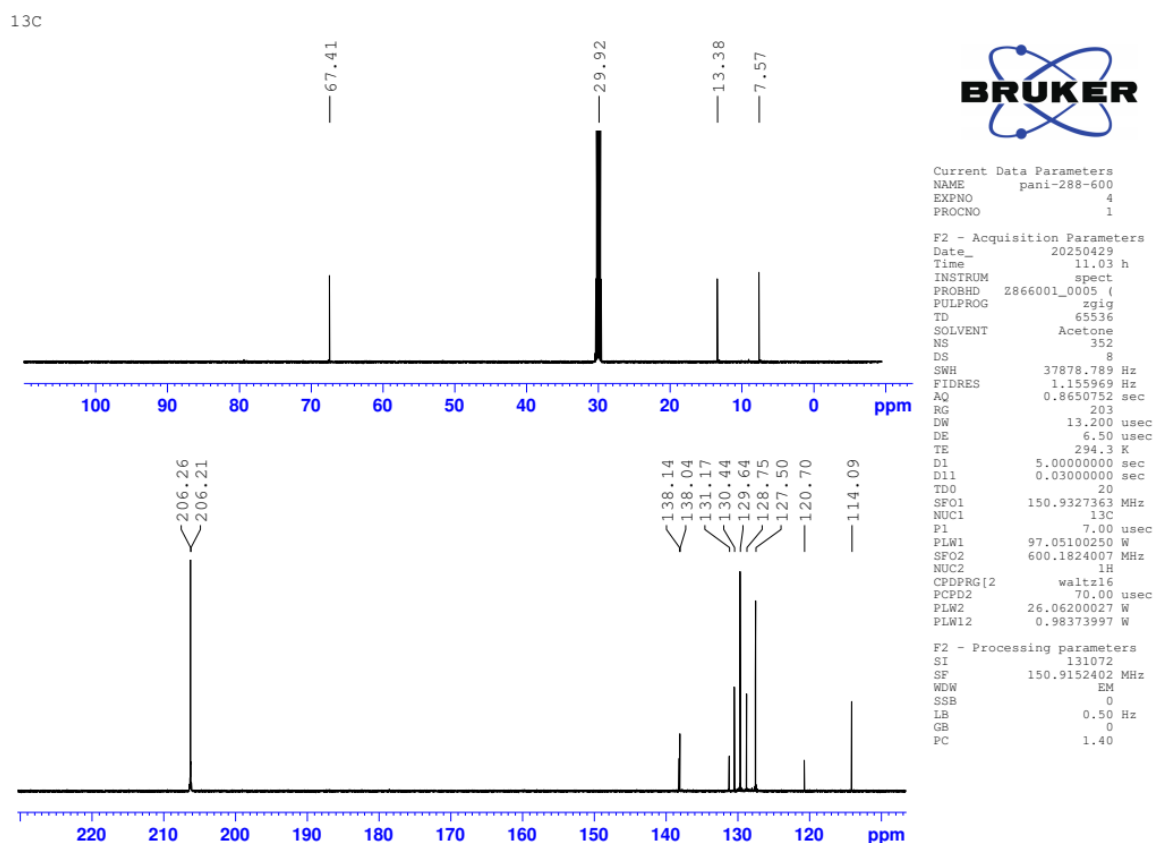


Fig. S 68. ^{13}C NMR spectrum of compound 4h (151 MHz, acetone- d_6).

^1H - ^{13}C HSQC

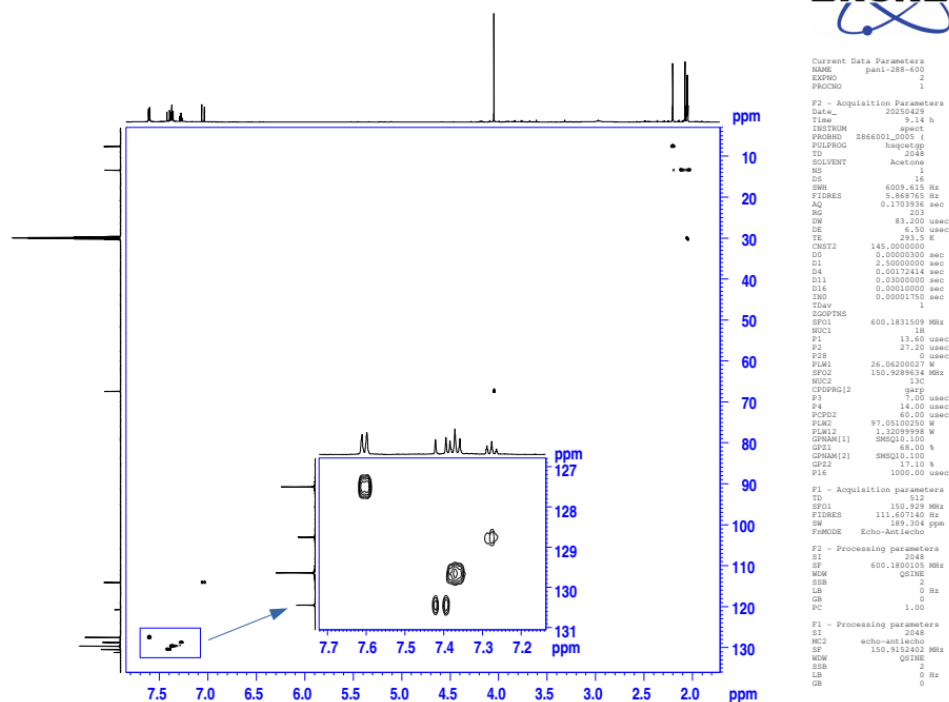


Fig. S 69. ^1H - ^{13}C HSQC spectrum of compound 4h (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

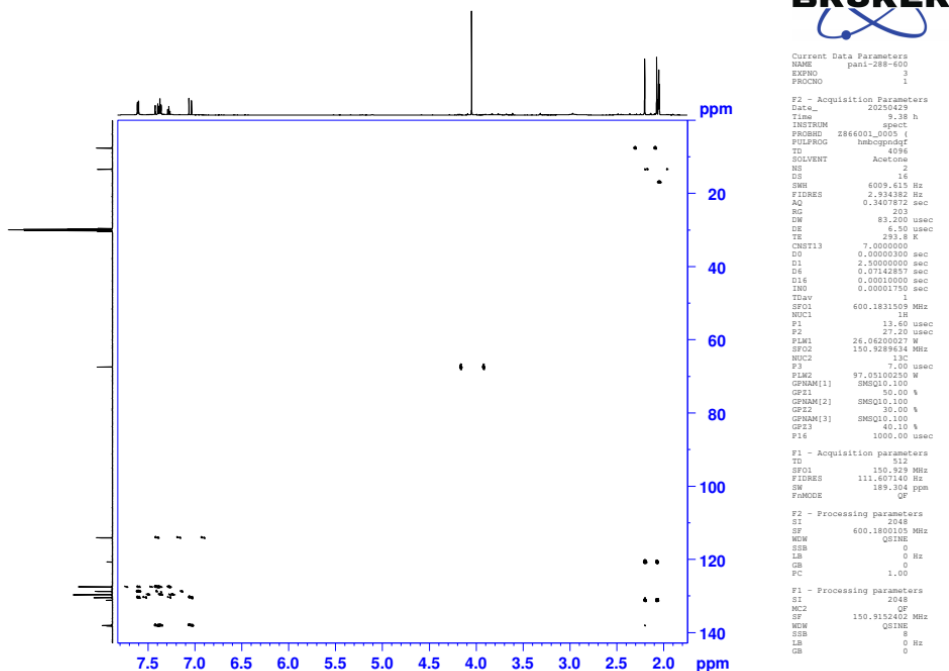


Fig. S 60. ^1H - ^{13}C HMBC spectrum of compound 4h (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

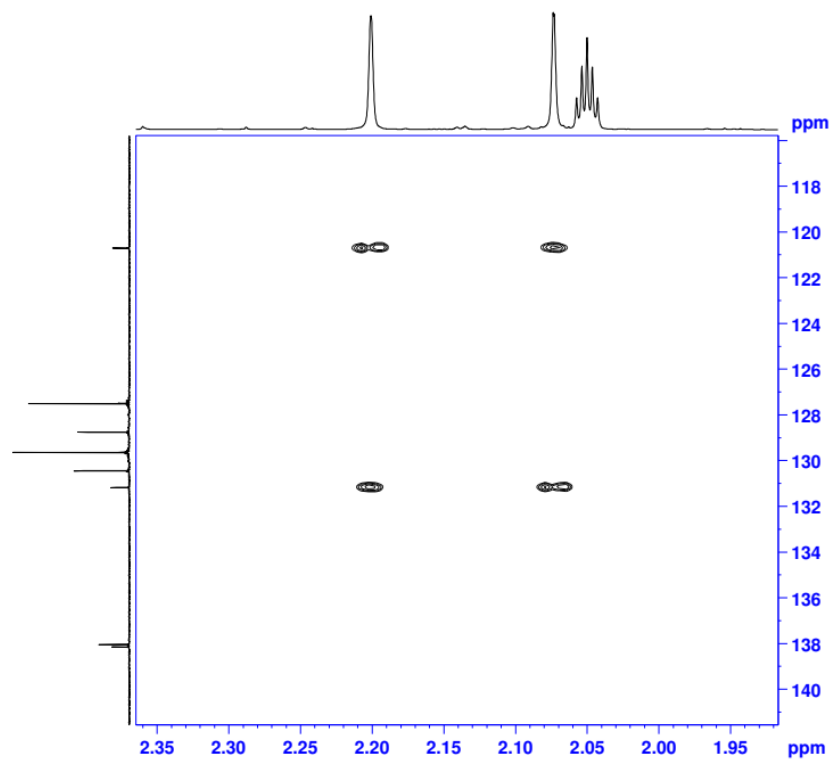


Fig. S 61. Part of ^1H - ^{13}C HMBC spectrum of compound 4h (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

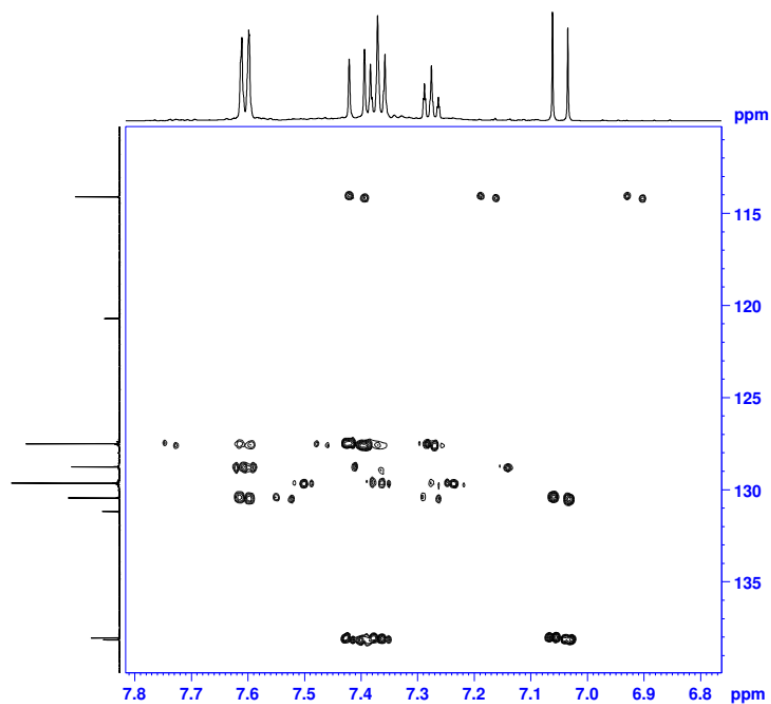
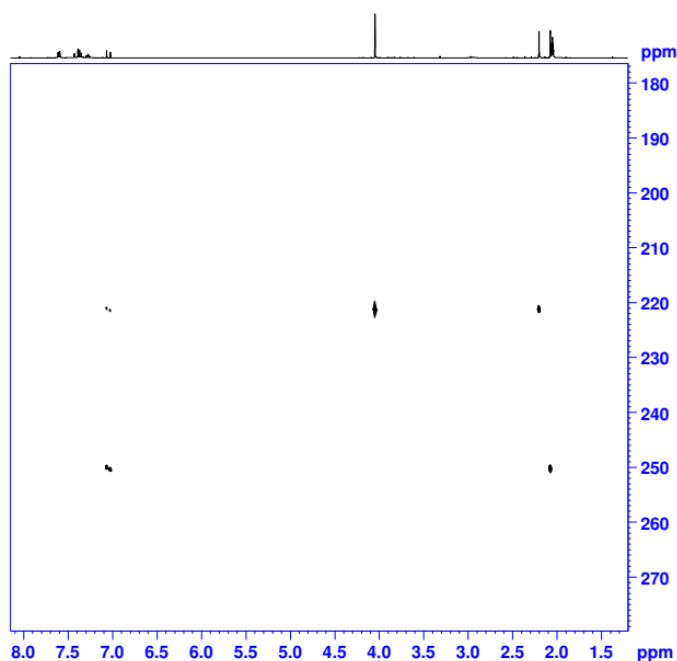


Fig. S 62. Part of ^1H - ^{13}C HMBC spectrum of compound 4h (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).



Current Data Parameters
NAME pant-288-405
EXPNO 2
PROCNO 2

F2 - Acquisition Parameters
Date_ 20230501
Time 14.18 h
INSTRUM spect
PROBHD Z108618_1087 (I
PULPROG hmcgpgdgt
TD 4096
SOLVENT Acetone
NS 8
DS 16
SWH 4000.000 Hz
FIDRES 1.953125 Hz
AQ 0.5120000 sec
RG 323
DW 125.000 usec
DE 6.50 usec
TE 295.4 K
CST13 1.0000000
D0 0.0000000 sec
D1 2.5000000 sec
D6 0.14466870 sec
D16 0.0001000 sec
TMO 0.00004400 sec
TDav 1
SFO1 400.132007 MHz
NUC1 1H
P1 14.00 usec
P2 28.00 usec
PLM1 17.34499931 W
SFO2 40.5513082 MHz
NUC2 15N
P3 18.00 usec
PLM2 78.47899792 W
GPRAM(1) SINE,100
GP21 70.00 %
GPRAM(2) SINE,100
GP22 30.00 %
GPRAM(3) SINE,100
GP23 50.10 %
P16 1000.00 usec

F1 - Acquisition parameters
SI 112
SFO1 40.55131 MHz
FIDRES 44.389306 Hz
SW 280.229 ppm
F1MODE QF

F2 - Processing parameters
SI 2048
SF 400.1300059 MHz
WDW CM
SSB 0
LB -2.00 Hz
GB 0.2
PC 1.00

F1 - Processing parameters
SI 2048
MC2 QF
SF 40.5448210 MHz
WDW GETHS
SSB 4
LB 0 Hz
GB 0

Fig. S 63. ^1H - ^{15}N HMBC spectrum of compound 4h (400 MHz – ^1H , 41 MHz – ^{15}N , acetone- d_6).

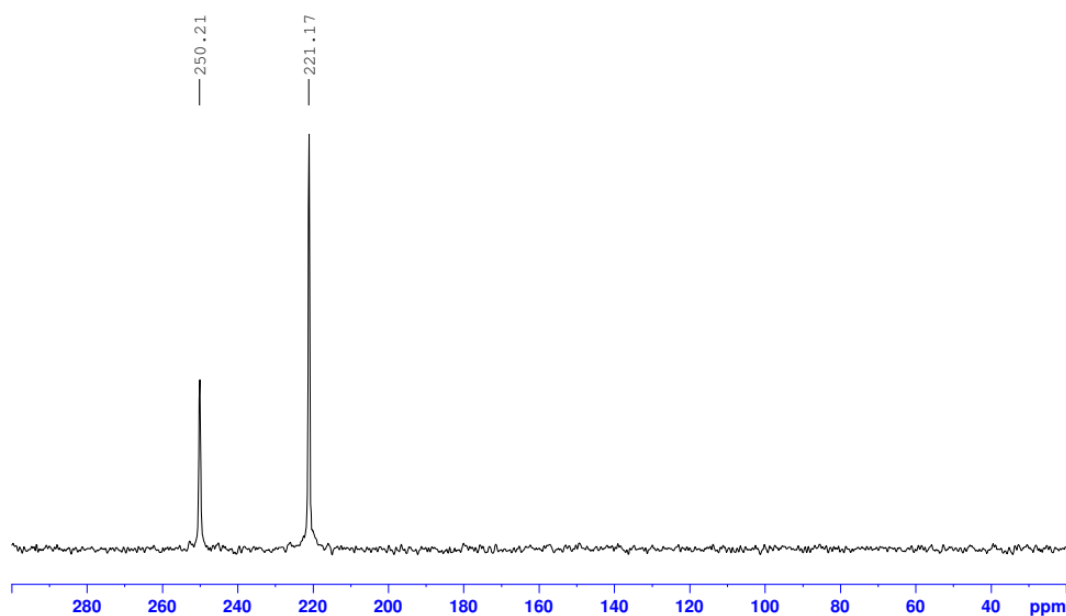
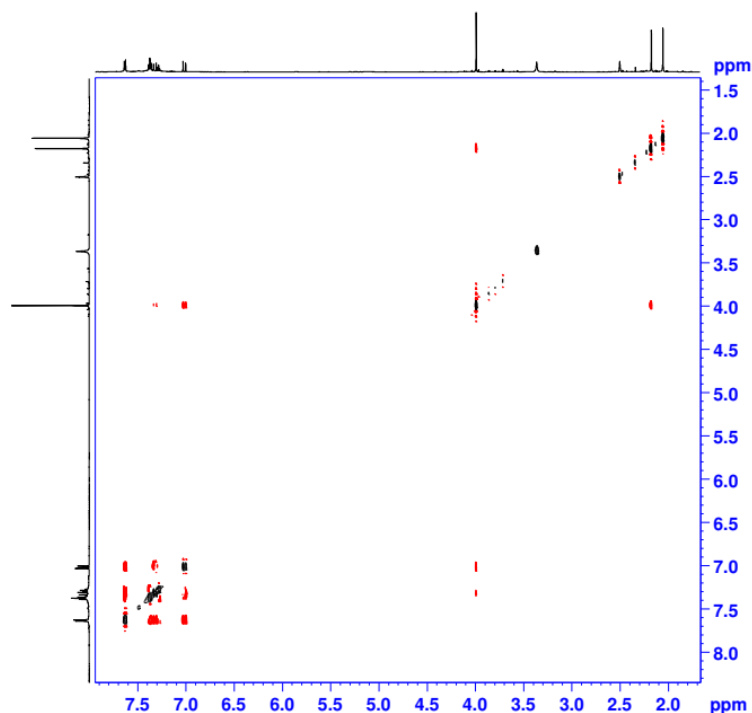


Fig. S 64. ^{15}N NMR spectrum of compound 4h (41 MHz, acetone- d_6), obtained by projection from ^1H - ^{15}N HMBC spectrum.

^1H - ^1H NOESY



Current Data Parameters
NAME pani-288dms
EXPNO 5
PROCNO 1

F2 - Acquisition Parameters
Date_ 20250604
Time 13.19 h
INSTRUM spect
PROBHD Z866001_0005 ()
PULPROG noesygpgppp
TD 4096
SOLVENT DMSO
NS 2
DS 16
SWH 9014.423 Hz
FIDRES 4.401574 Hz
AQ 0.2271915 sec
RG 144
DW 55.467 usec
DE 6.50 usec
TE 294.7 K
D0 0.0003818 sec
D1 2.50000000 sec
D8 0.50000000 sec
D11 0.03000000 sec
D12 0.00002000 sec
D16 0.00010000 sec
IND 0.0001100 sec
TDav 1
SFO1 600.1836011 MHz
NUC1 1H
P1 13.60 usec
P2 27.20 usec
P17 2500.00 usec
PLW1 26.06200027 W
PLW10 7.71250010 W
GPM1[1] SMO10.100
GPZ1 40.00 %
P16 1000.00 usec

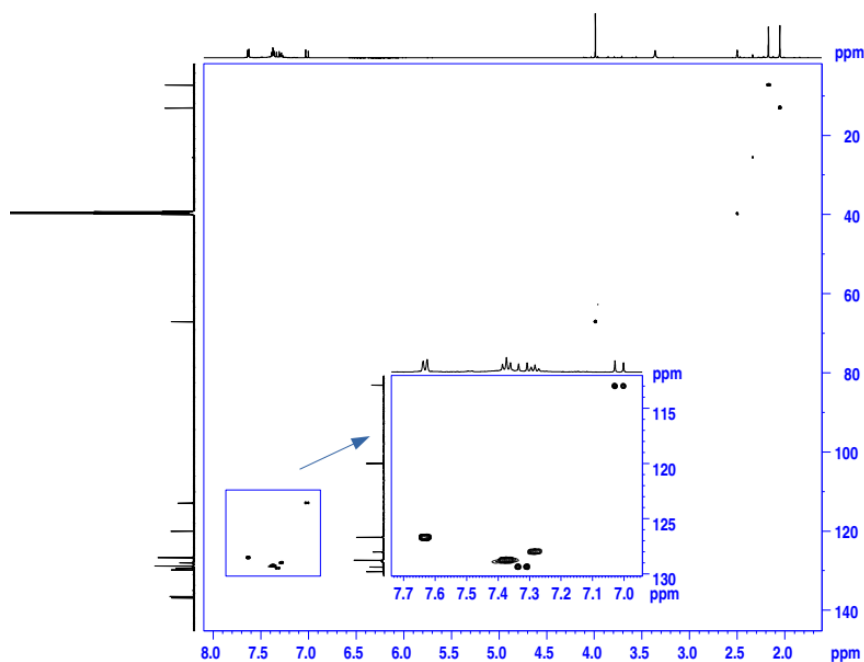
F1 - Acquisition parameters
TD 512
SFO1 600.1836 MHz
FIDRES 35.191441 Hz
SW 15.010 ppm
F2MODE States-TFPI

F2 - Processing parameters
SI 2048
SF 600.1800049 MHz
WDW QSI
SSB 2
LB 0 Hz
GB 0
PC 1.00

F1 - Processing parameters
SI 2048
NC2 States-TFPI
SF 600.1800049 MHz
WDW QSI
SSB 2
LB 0 Hz
GB 0

Fig. S 65. ^1H - ^1H NOESY spectrum of compound 4h (600 MHz, $\text{DMSO}-d_6$, mixing time - 0.5 s).

^1H - ^{13}C HSQC



Current Data Parameters
NAME pani-288dms
EXPNO 3
PROCNO 1

F2 - Acquisition Parameters
Date_ 20250604
Time 11.14 h
INSTRUM spect
PROBHD Z866001_0005 ()
PULPROG hsqc
TD 2048
SOLVENT DMSO
NS 1
DS 14
SWH 6009.615 Hz
FIDRES 5.868765 Hz
AQ 0.1703526 sec
RG 203
DW 83.200 usec
DE 6.50 usec
TE 294.4 K
CHST2 145.0000000
D0 0.00000000 sec
D1 2.50000000 sec
D4 0.05172414 sec
D11 0.03000000 sec
D16 0.00010000 sec
IND 0.00001850 sec
TDav 1
SCOPFNS
SFO1 600.1833010 MHz
NUC1 1H
P1 13.60 usec
P2 27.20 usec
P28 0 usec
PLW1 26.06200027 W
SFO2 150.9274542 MHz
NUC2 ^{13}C
CHST2[2] gmc
P1 7.00 usec
P4 14.00 usec
PCPD2 60.00 usec
PLW2 97.05100050 W
PLW12 1.32099998 W
GPM1[1] SMO10.100
GPZ1 68.00 %
GPM1[2] SMO10.100
GPZ2 17.10 %
P16 1000.00 usec

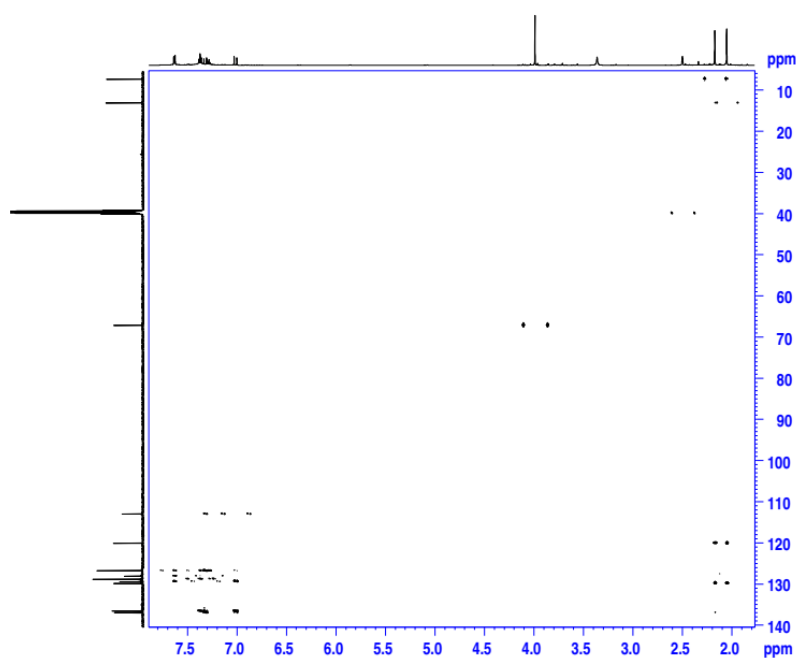
F1 - Acquisition parameters
TD 512
SFO1 150.9275 MHz
FIDRES 100.140255 Hz
SW 169.890 ppm
F2MODE Echo-Antiecho

F2 - Processing parameters
SI 2048
SF 600.1800047 MHz
WDW QSI
SSB 2
LB 0 Hz
GB 0
PC 1.00

F1 - Processing parameters
SI 2048
NC2 echo-antiecho
SF 150.9154486 MHz
WDW QSI
SSB 2
LB 0 Hz
GB 0

Fig. S 76. ^1H - ^{13}C HSQC spectrum of compound 4h (600 MHz - ^1H , 151 MHz - ^{13}C , $\text{DMSO}-d_6$).

¹H-¹³C HMBC



```

NAME      1
PROCNO    1

F2 - Acquisition Parameters
Date_     20250604
Time      11:38 h
INSTRUM   spect
PROBHD    Z866001_0005 (
PULPROG   hmcgpgmdg2
TD         4096
SOLVENT   DMSO
NS         2
DS         16
SWH        6009.615 Hz
FIDRES     2.934392 Hz
AQ         0.3407872 sec
RG         203
DW         83.200 usec
DE         6.50 usec
TE         294.2 K
CNS113     7.0000000
D0         0.0000000 sec
D1         2.5000000 sec
D6         0.07142857 sec
D16        0.00010000 sec
IND        0.00001950 sec
TD0AV      1
SF01       600.1833010 MHz
NUC1       1H
P1         13.60 usec
P2         27.20 usec
PL1        26.0620027 W
SF02       150.9274542 MHz
NUC2       13C
P3         7.00 usec
PL12       97.05100250 W
GPHAM[1]   SMSG10.100
GP1        50.00 %
GPHAM[2]   SMSG10.100
GP2        30.00 %
GPHAM[3]   SMSG10.100
GP3        40.10 %
P14        1000.00 usec

F1 - Acquisition parameters
TD         1024
SF01       150.9275 MHz
FIDRES     50.080128 Hz
SW         169.890 ppm
F0         0
PC         1.00

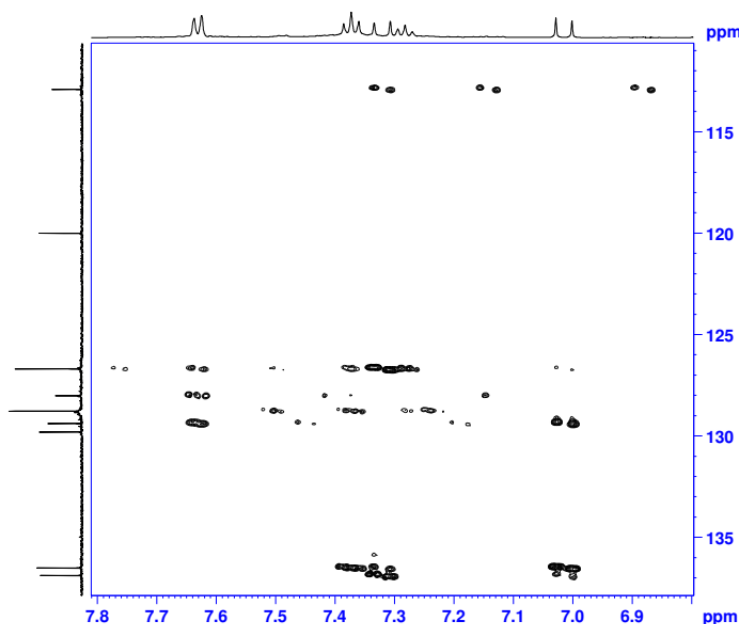
F2 - Processing parameters
SI         2048
SF         600.1800047 MHz
WDW        QSI
SSB         0
LB          0 Hz
GB          0
PC         1.00

F1 - Processing parameters
SI         2048
MC2        QF
SF         150.9154486 MHz
WDW        QSI
SSB         8
LB          0 Hz
GB          0

```

Fig. S 77. ¹H-¹³C HMBC spectrum of compound 4h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆).

¹H-¹³C HMBC



```

Current Data Parameters
NAME      pani-288dms
EXPNO     4
PROCNO    1

F2 - Acquisition Parameters
Date_     20250604
Time      11:38 h
INSTRUM   spect
PROBHD    Z866001_0005 (
PULPROG   hmcgpgmdg2
TD         4096
SOLVENT   DMSO
NS         2
DS         16
SWH        6009.615 Hz
FIDRES     2.934392 Hz
AQ         0.3407872 sec
RG         203
DW         83.200 usec
DE         6.50 usec
TE         294.2 K
CNS113     7.0000000
D0         0.0000000 sec
D1         2.5000000 sec
D6         0.07142857 sec
D16        0.00010000 sec
IND        0.00001950 sec
TD0AV      1
SF01       600.1833010 MHz
NUC1       1H
P1         13.60 usec
P2         27.20 usec
PL1        26.0620027 W
SF02       150.9274542 MHz
NUC2       13C
P3         7.00 usec
PL12       97.05100250 W
GPHAM[1]   SMSG10.100
GP1        50.00 %
GPHAM[2]   SMSG10.100
GP2        30.00 %
GPHAM[3]   SMSG10.100
GP3        40.10 %
P14        1000.00 usec

F1 - Acquisition parameters
TD         1024
SF01       150.9275 MHz
FIDRES     50.080128 Hz
SW         169.890 ppm
F0         0
PC         1.00

F2 - Processing parameters
SI         2048
SF         600.1800047 MHz
WDW        QSI
SSB         0
LB          0 Hz
GB          0
PC         1.00

F1 - Processing parameters
SI         2048
MC2        QF
SF         150.9154486 MHz
WDW        QSI
SSB         8
LB          0 Hz
GB          0

```

Fig. S 78. Part of ¹H-¹³C HMBC spectrum of compound 4h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆).

¹H-¹³C HMBC

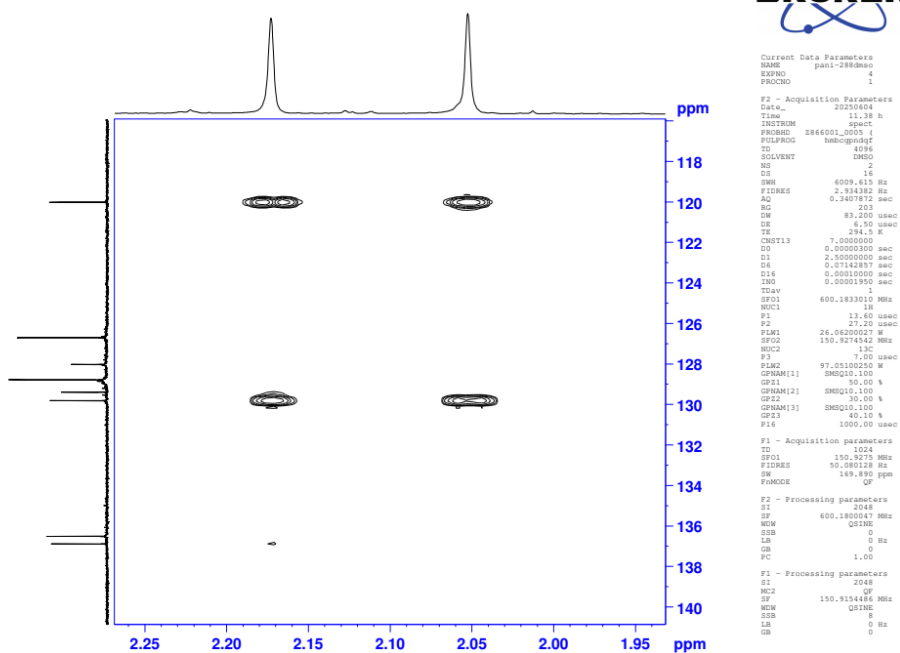


Fig. S 79. Part of ¹H-¹³C HMBC spectrum of compound 4h (600 MHz - ¹H, 151 MHz - ¹³C, DMSO-*d*₆).

S4.17. 1,6,6-Trimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-4-oxo-4,5,6,7-tetrahydro-1*H*-benzimidazole 3-oxide (**6a**).

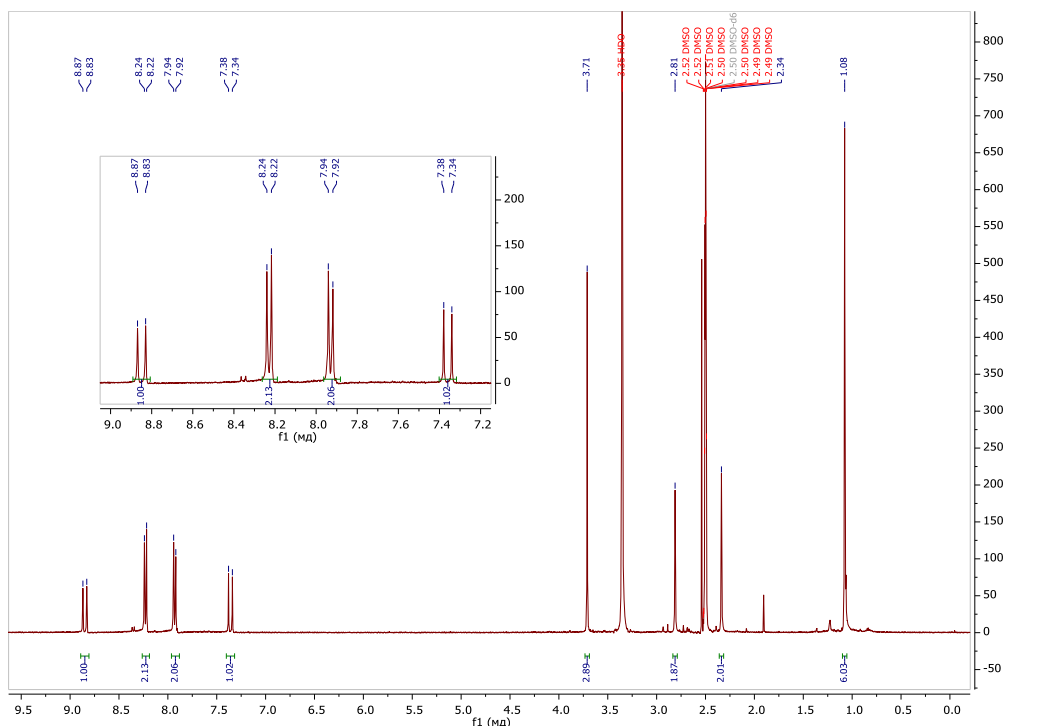
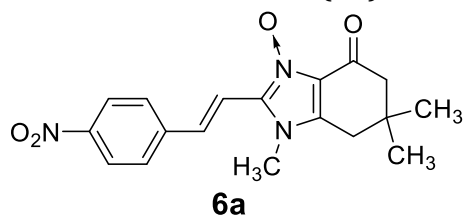


Fig. S 66. ^1H NMR spectrum of compound **6a** (400 MHz, $\text{DMSO}-d_6$).

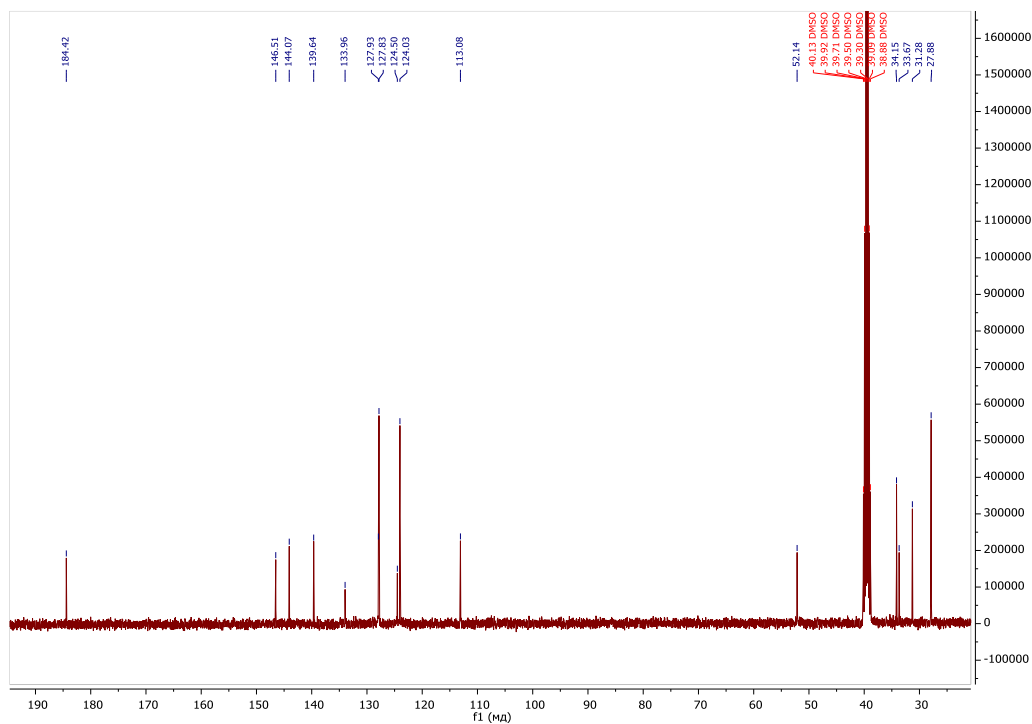


Fig. S 67. ^{13}C NMR spectrum of compound **6a** (101 MHz, $\text{DMSO}-d_6$).

S4.18. 4-Acetyl-1,5-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole 3-oxide (**6b**).

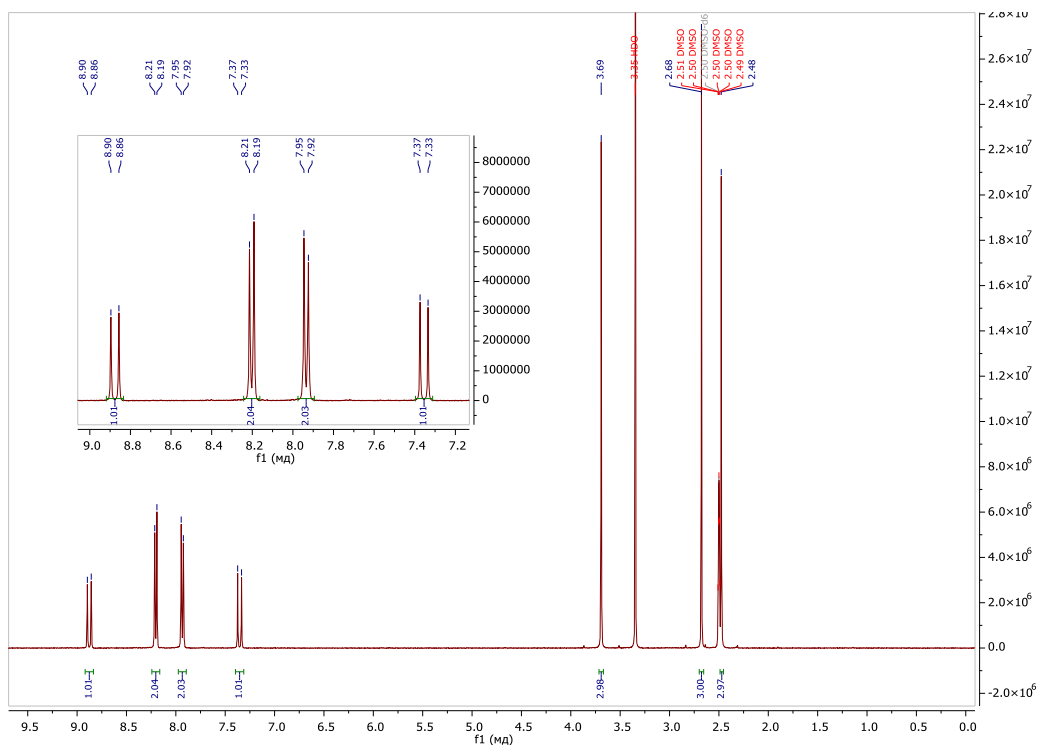
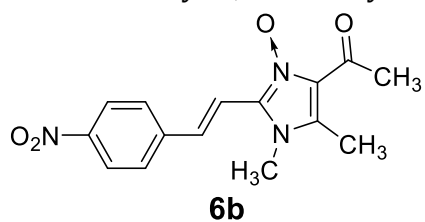


Fig. S 68. ¹H NMR spectrum of compound **6b** (400 MHz, DMSO-*d*₆).

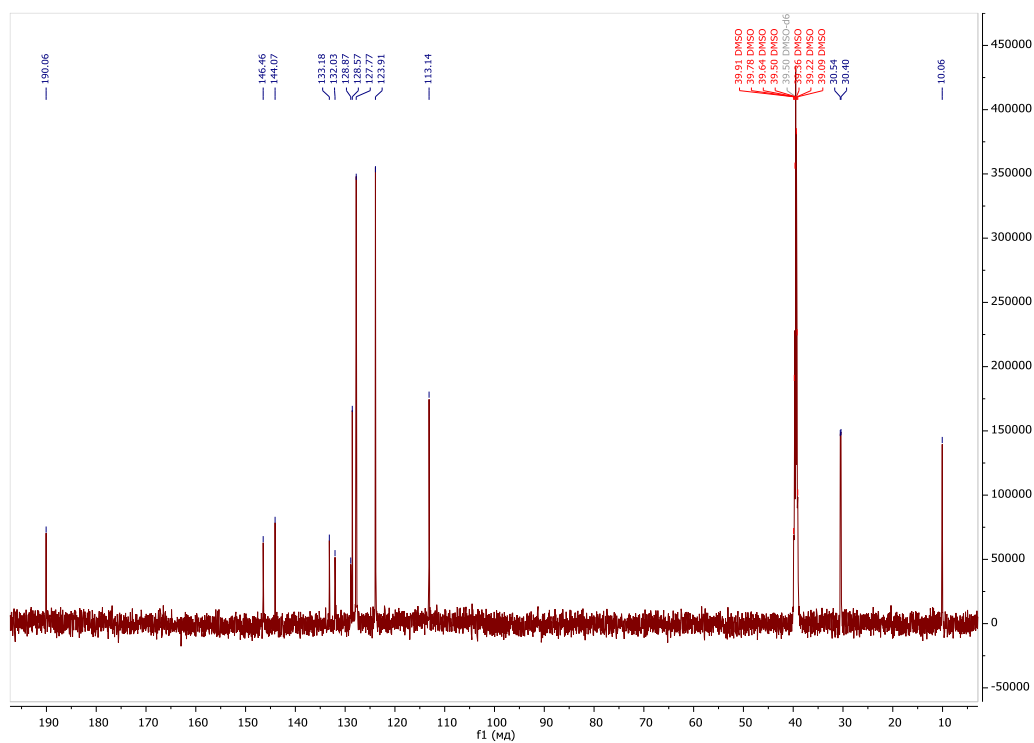


Fig. S 69. ¹³C NMR spectrum of compounds **6b** (151 MHz, DMSO-*d*₆).

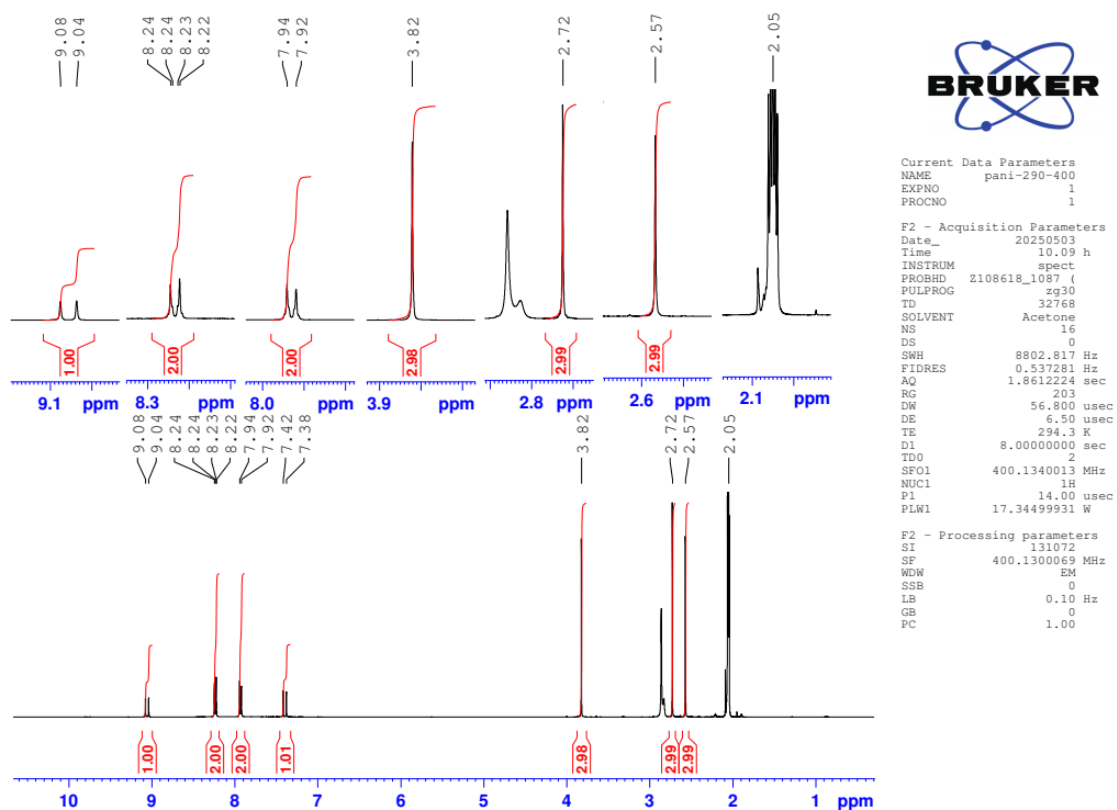


Fig. S 70. ^1H NMR spectrum of compound 6b (400 MHz, acetone- d_6).

13C

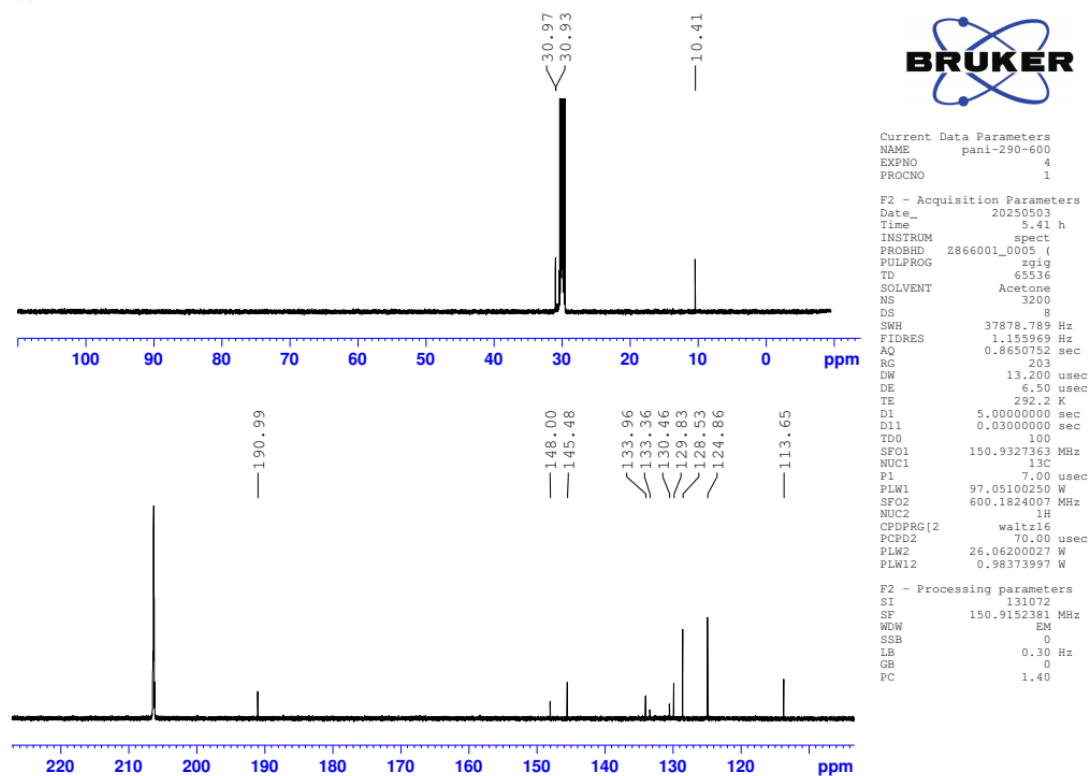


Fig. S 85. ^{13}C NMR spectrum of compound 6b (151 MHz, acetone- d_6).

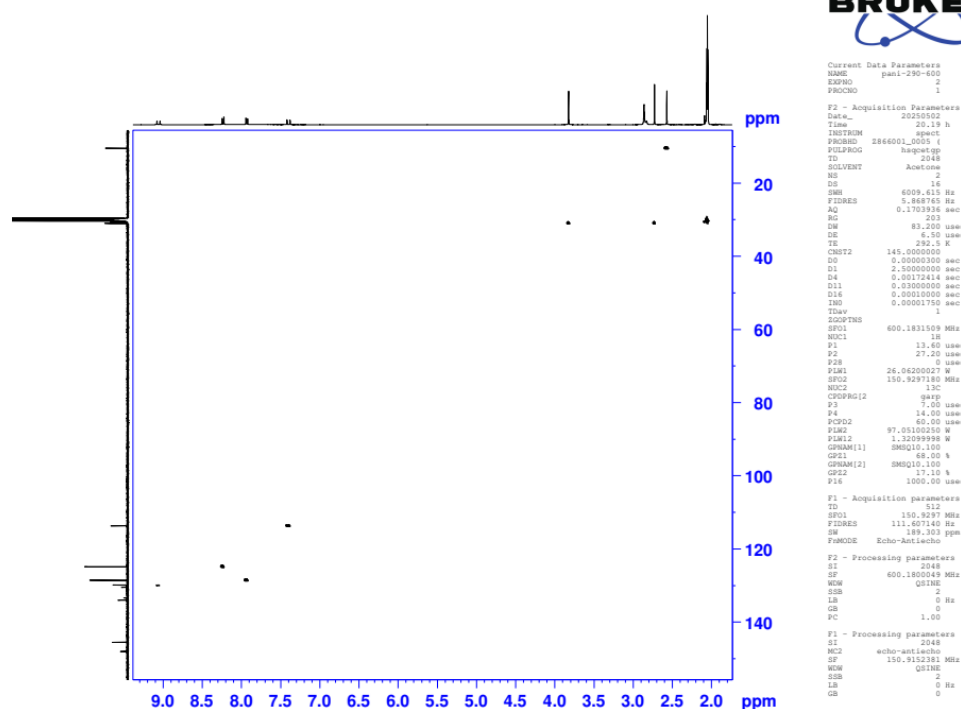


Fig. S 86. ^1H - ^{13}C HSQC spectrum of compound 6b (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

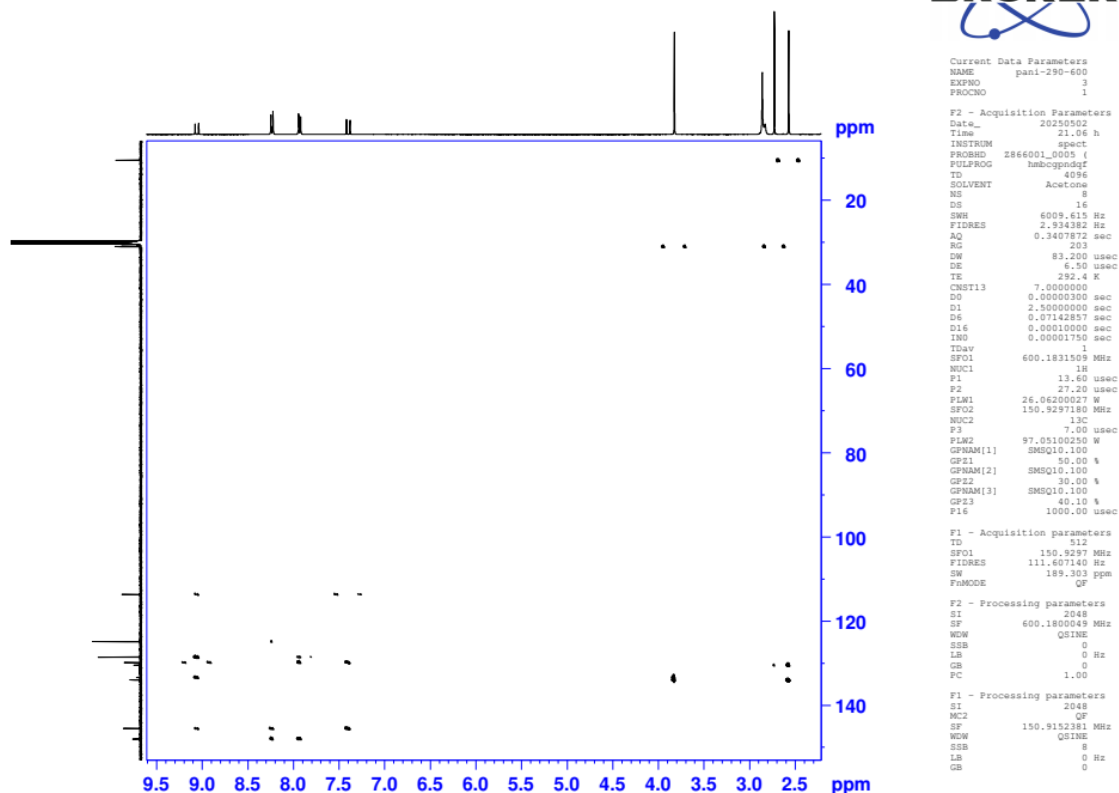


Fig. S 87. ^1H - ^{13}C HMBC spectrum of compound 6b (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

¹H-¹H NOESY

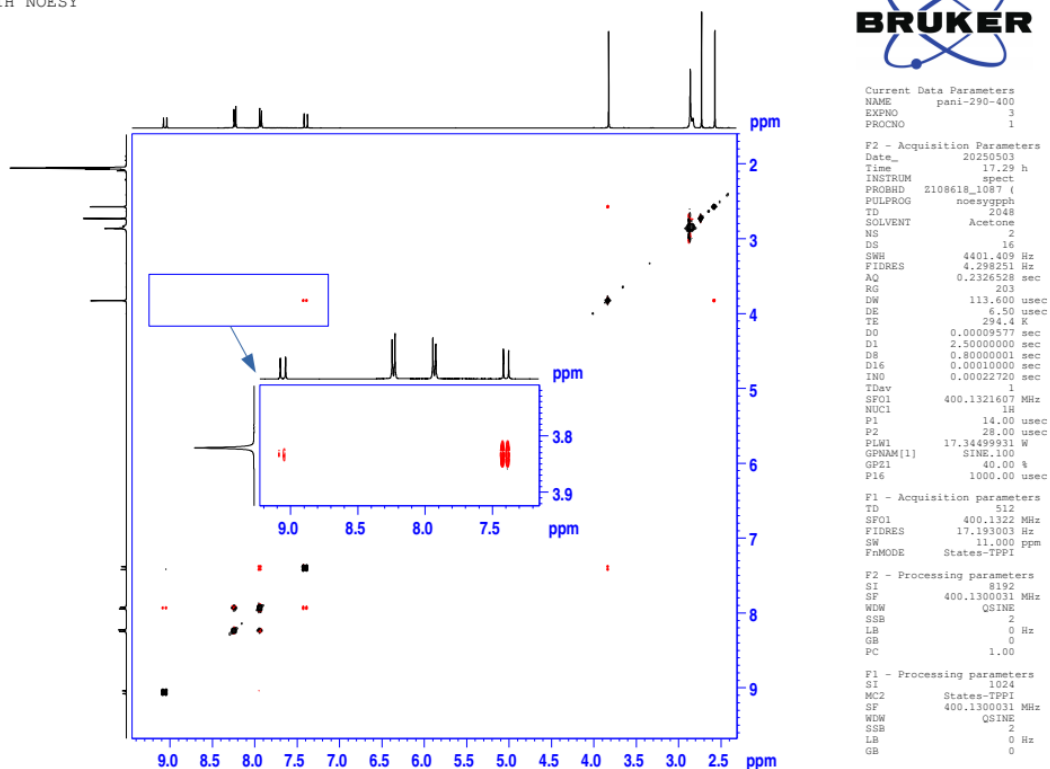


Fig. S 71. ¹H-¹H NOESY spectrum of compound 6b (400 MHz, acetone-*d*₆).

pani-290
acetone-*d*₆
¹H-¹⁵N HMBC

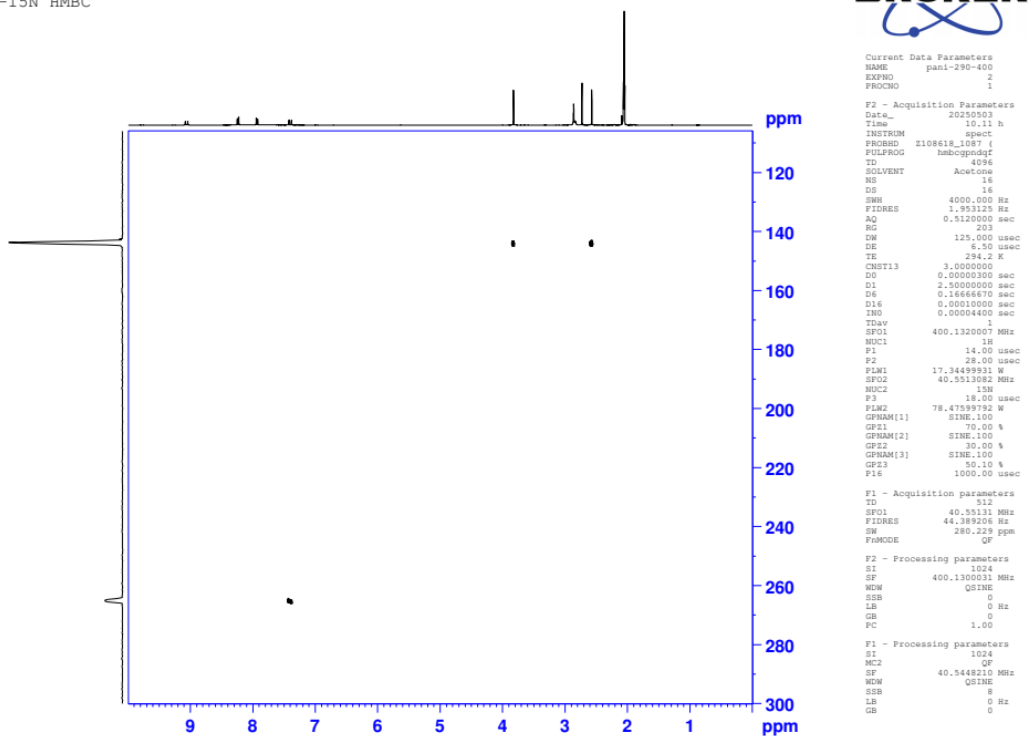


Fig. S 72. ¹H-¹⁵N HMBC spectrum of compound 6b (400 MHz -¹H, 41 MHz -¹⁵N, acetone-*d*₆).

S4.19. 1,4,5-Trimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole 3-oxide (**6c**).

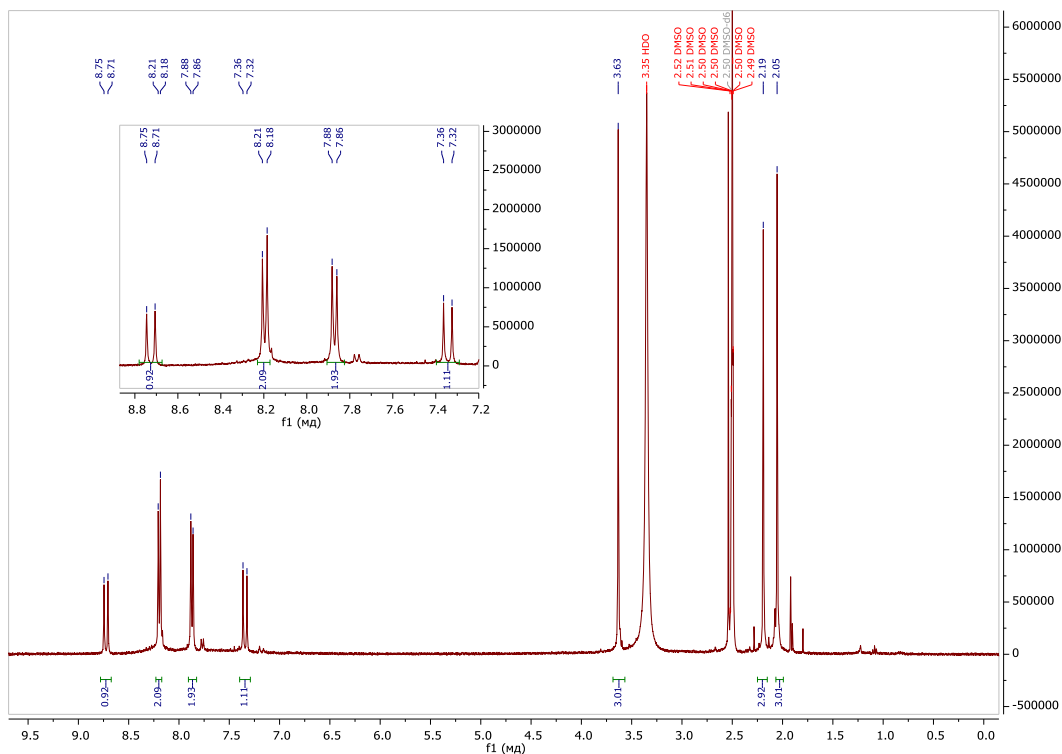
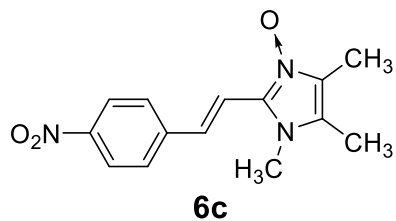


Fig. S 73. ¹H NMR spectrum of compound 6c (400 MHz, DMSO-*d*₆).

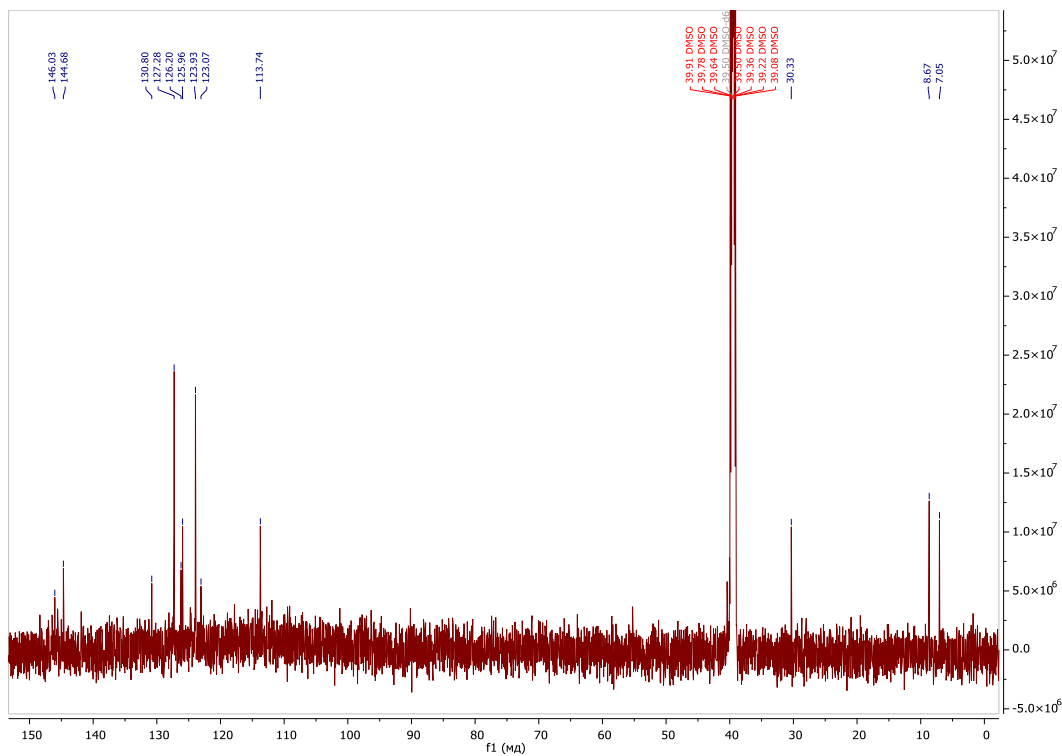


Fig. S 74. ¹³C NMR spectrum of compound 6c (151 MHz, DMSO-*d*₆).

S4.20. 1,4,5-Trimethyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole 3-oxide (**6d**).

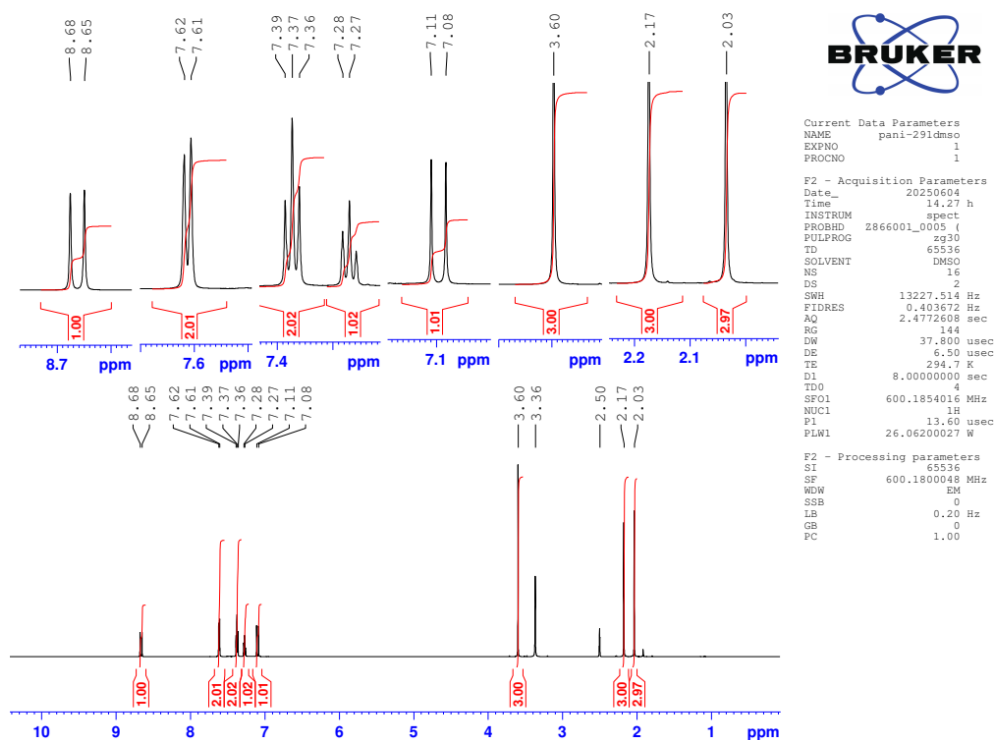
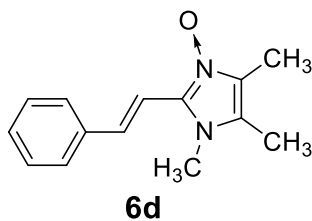


Fig. S 75. ¹H NMR spectrum of compound **6d** (600 MHz, DMSO-*d*₆).

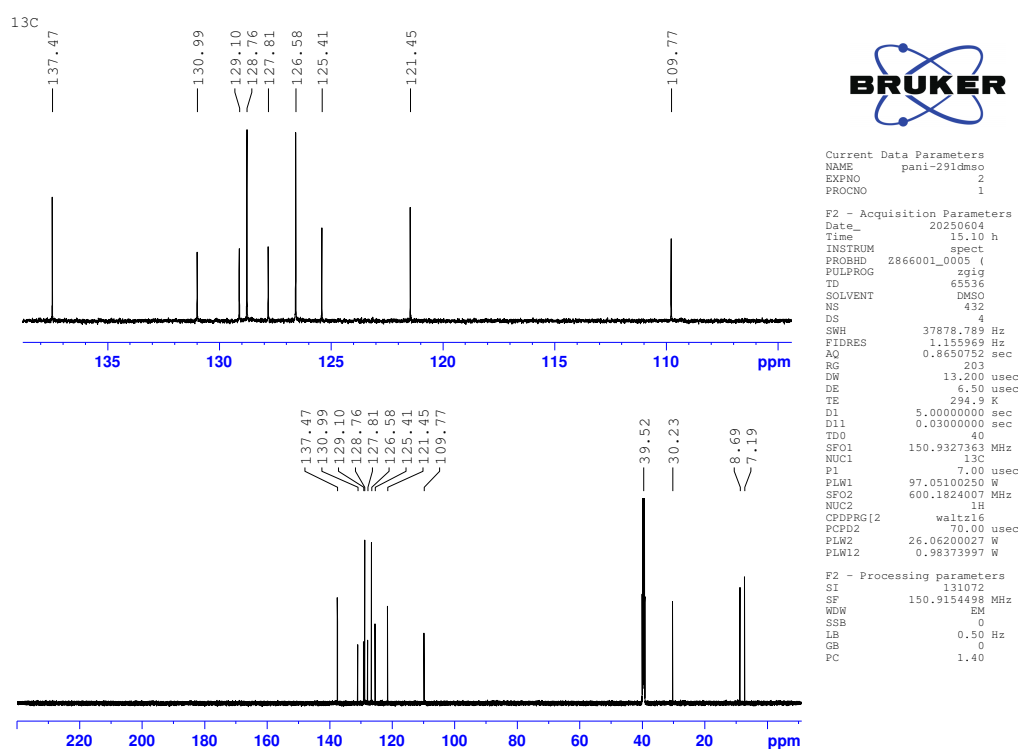


Fig. S 76. ¹³C NMR spectrum of compound **6d** (151 MHz, DMSO-*d*₆).

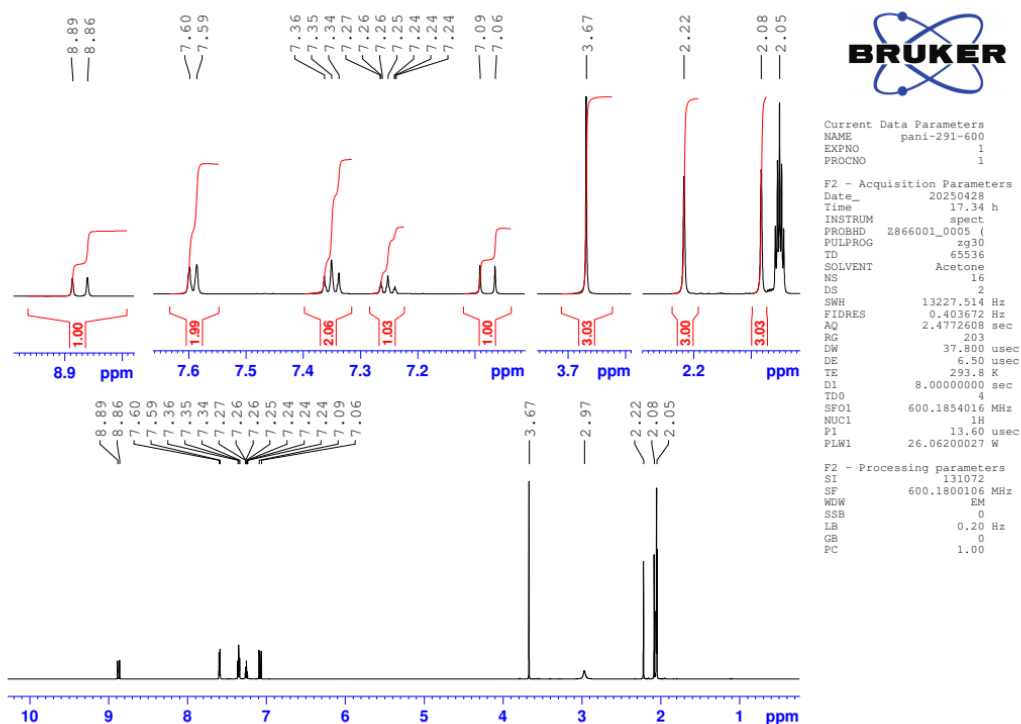


Fig. S 77. ^1H NMR spectrum of compounds 6d (600 MHz, acetone- d_6).

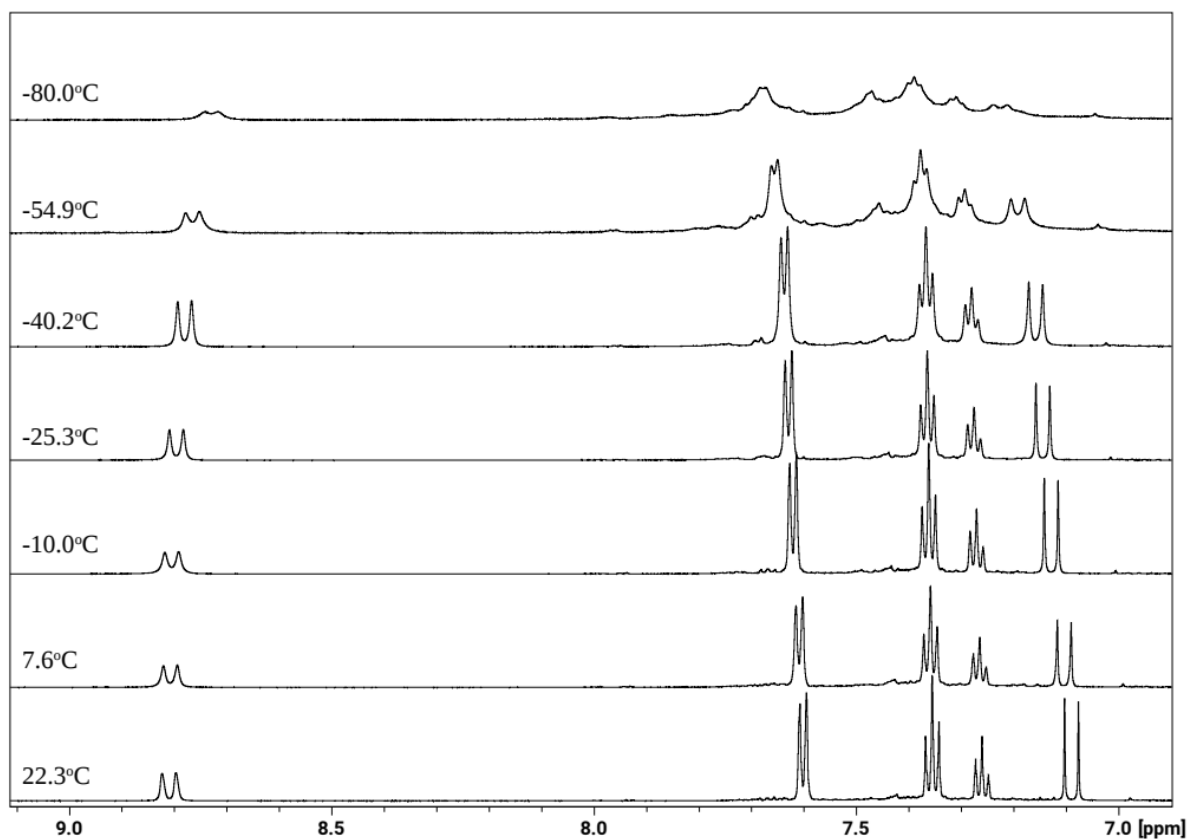


Fig. S 78. ^1H NMR spectra of compound 6d at different temperatures (600 MHz, acetone- d_6 , range 7.0-9.0 ppm).

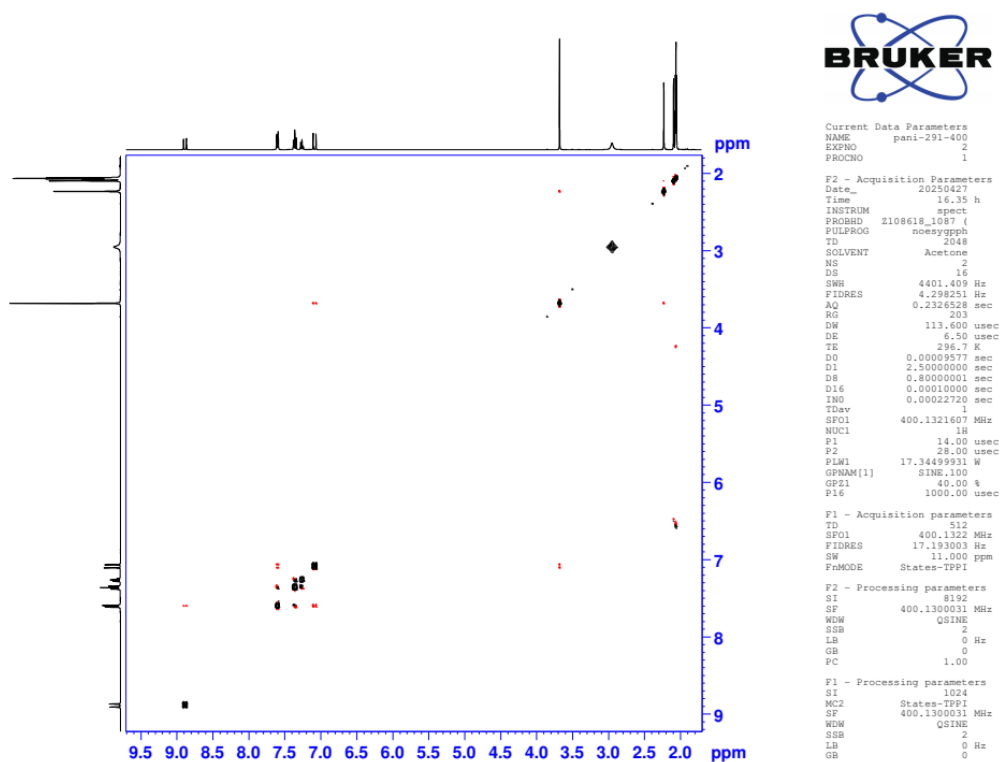


Fig. S 79. ^1H - ^1H NOESY spectrum of compound 6d (400 MHz, acetone- d_6 , mixing time – 0.8 s).

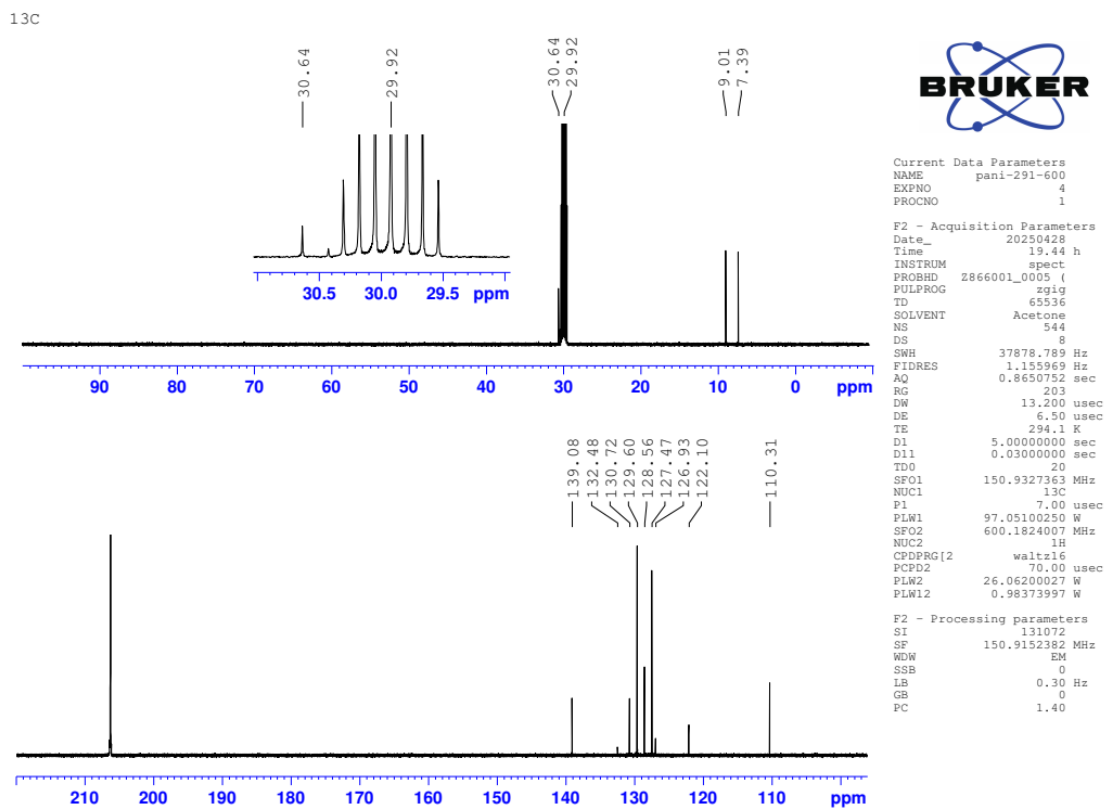


Fig. S 80. ^{13}C NMR spectrum of compound 6d (151 MHz, acetone- d_6).

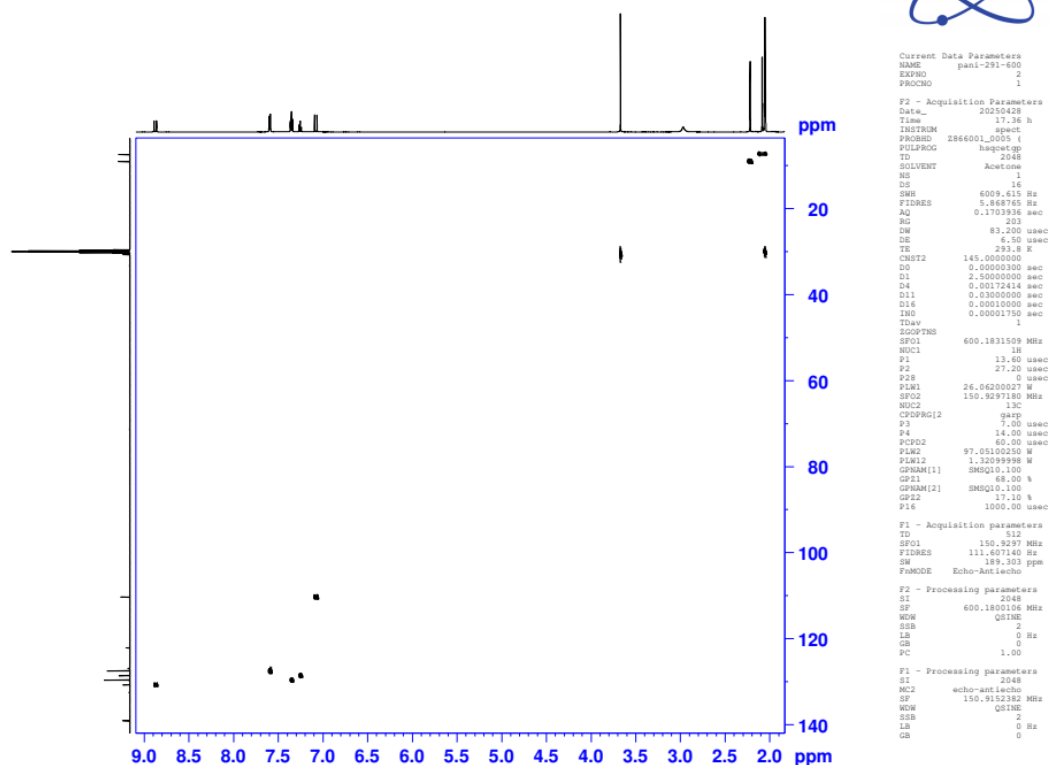


Fig. S 818. ^1H - ^{13}C HSQC spectrum of compound 6d (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

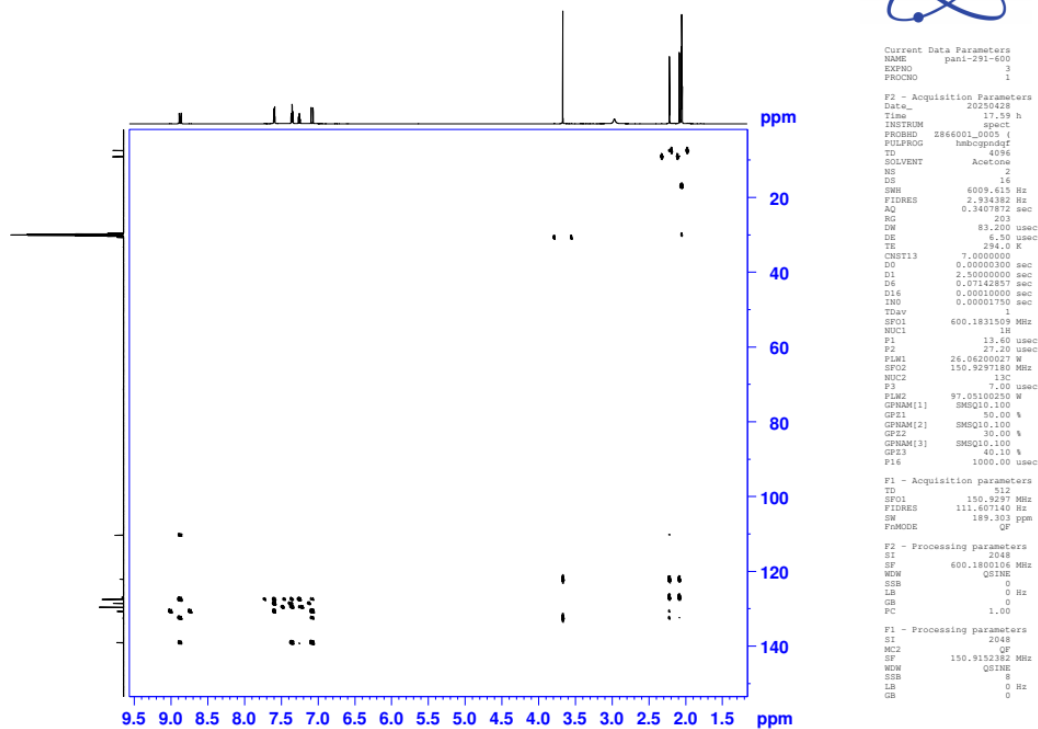
¹H-¹³C HMBC

Fig. S 82. ^1H - ^{13}C HMBC spectrum of compound 6d (600 MHz – ^1H , 151 MHz – ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

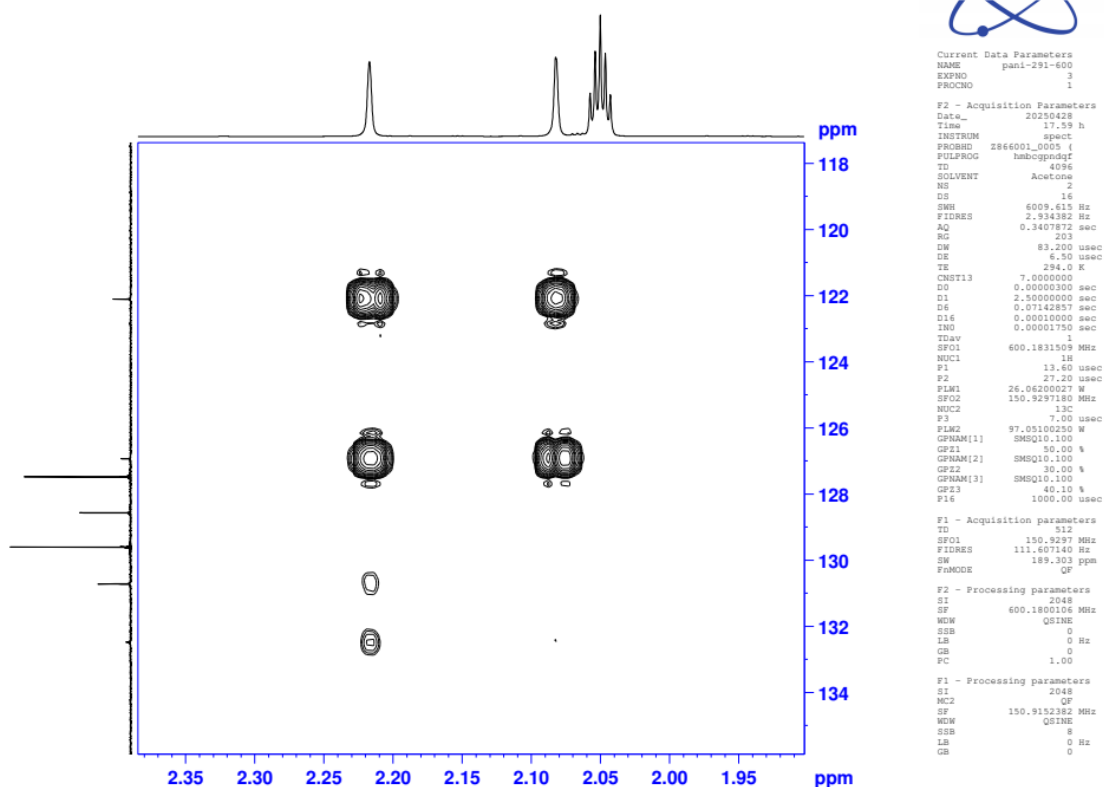


Fig. S 83. Part of ^1H - ^{13}C HMBC spectrum of compound 6d (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

^1H - ^{13}C HMBC

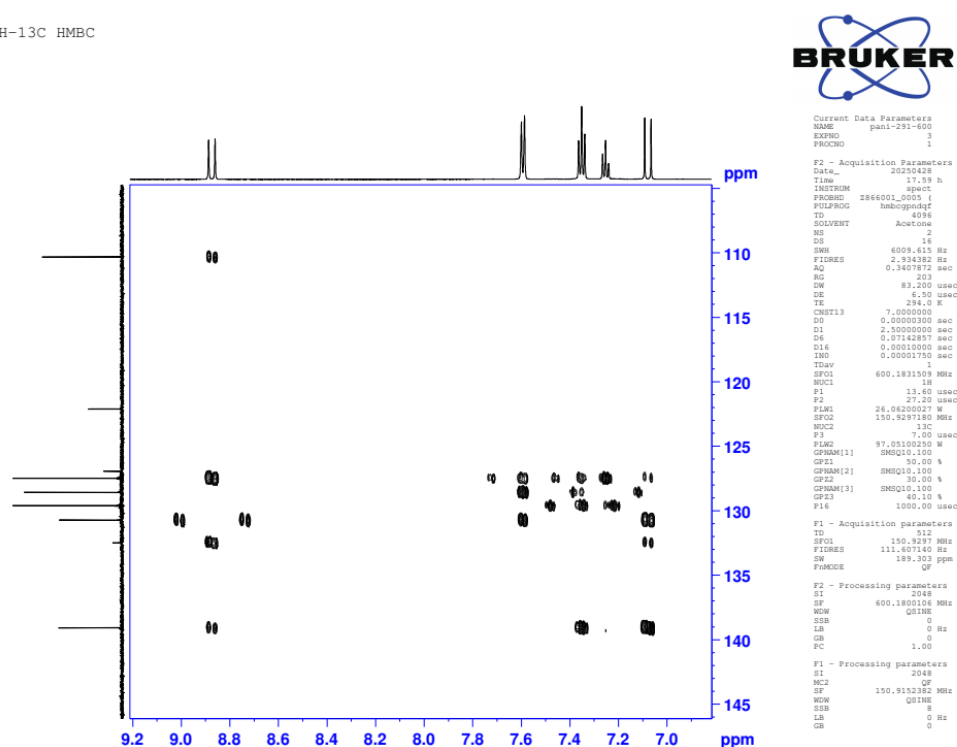


Fig. S 84. Part of ^1H - ^{13}C HMBC spectrum of compound 6d (600 MHz - ^1H , 151 MHz - ^{13}C , acetone- d_6).

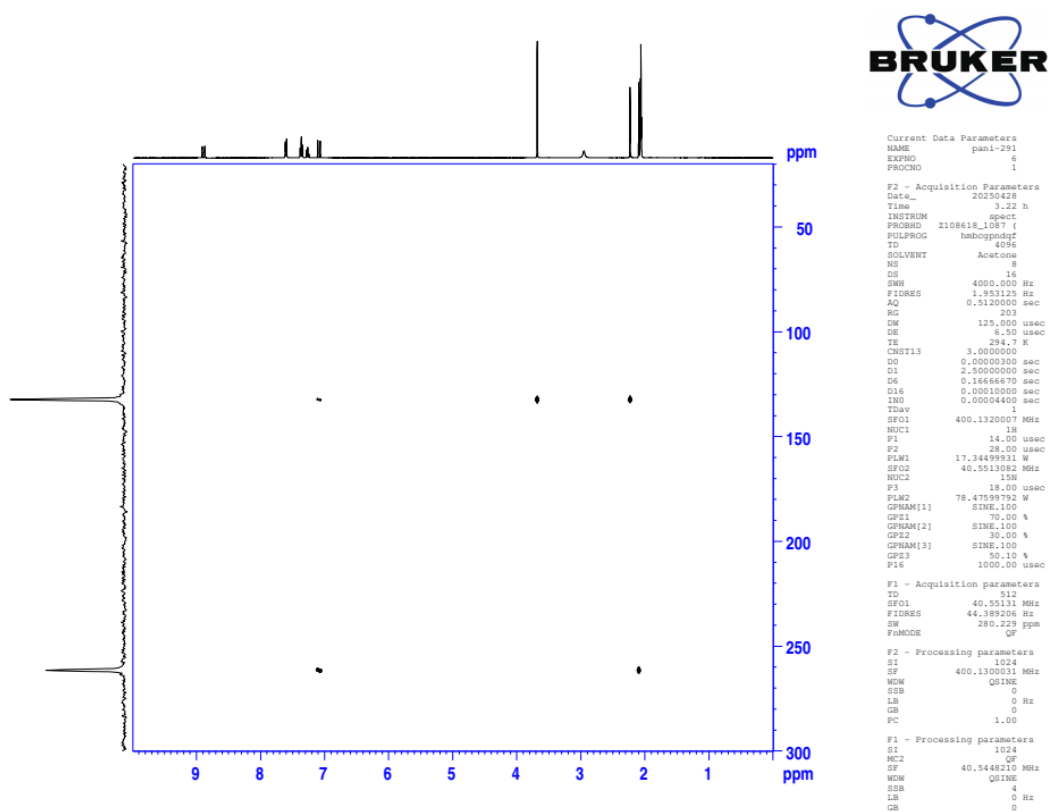


Fig. S 85. ^1H - ^{15}N HMBC spectrum of compound 6d (400 MHz – ^1H , 41 MHz – ^{15}N , acetone- d_6).

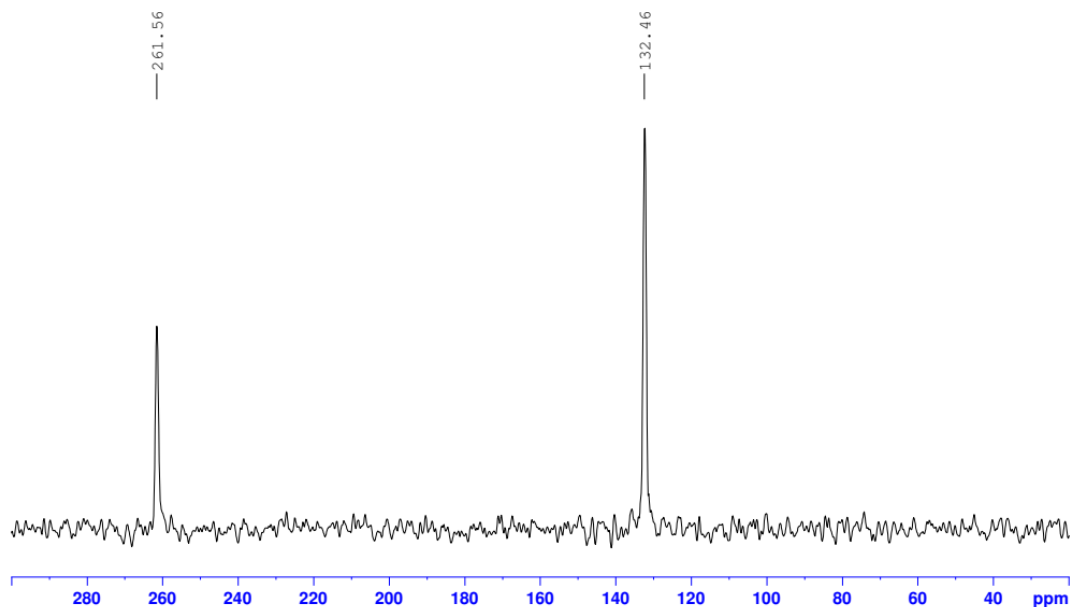


Fig. S 86. ^{15}N NMR spectrum of compound 6d (41 MHz, acetone- d_6), obtained from projection of ^1H - ^{15}N HMBC spectrum.

^1H - ^1H NOESY

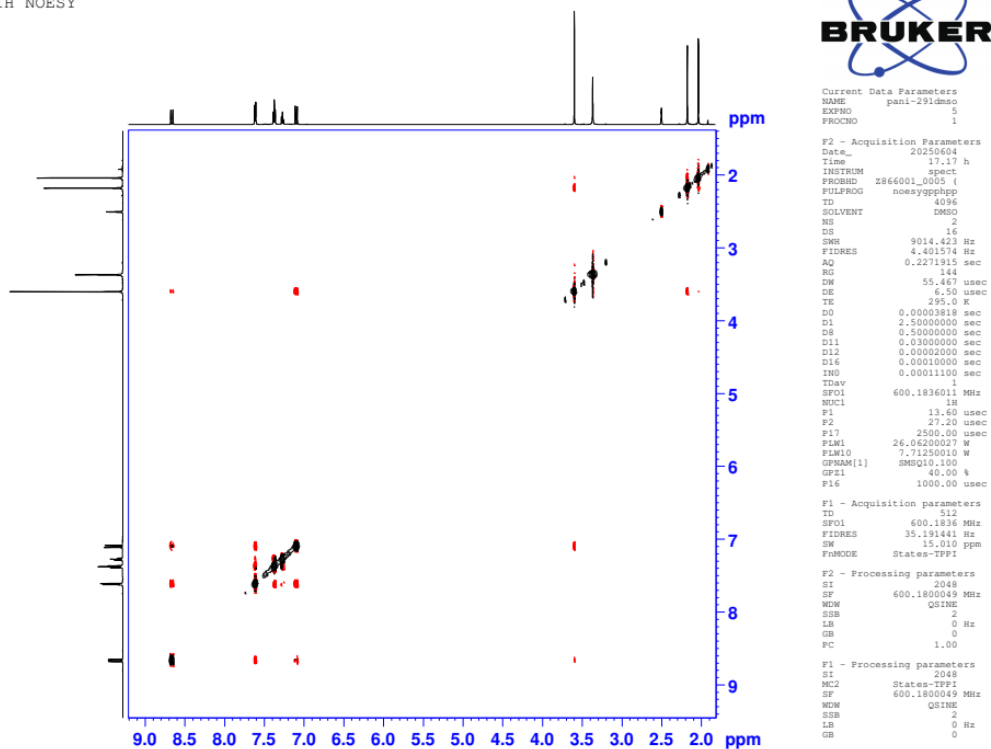


Fig. S 87. ^1H - ^1H NOESY spectrum of compound 6d (600 MHz, DMSO- d_6 , mixing time – 0.5 s).

^1H - ^{13}C HSQC

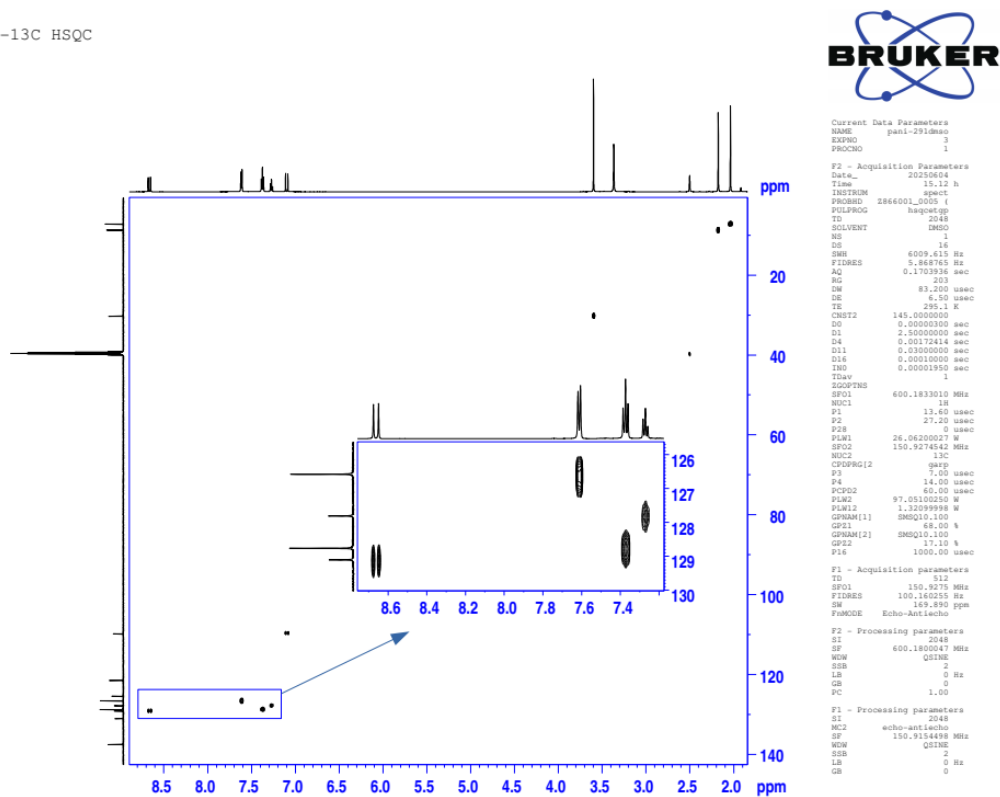


Fig. S 88. ^1H - ^{13}C HSQC spectrum of compound 6d (600 MHz – ^1H , 151 MHz – ^{13}C , DMSO- d_6).

¹H-¹³C HMBC

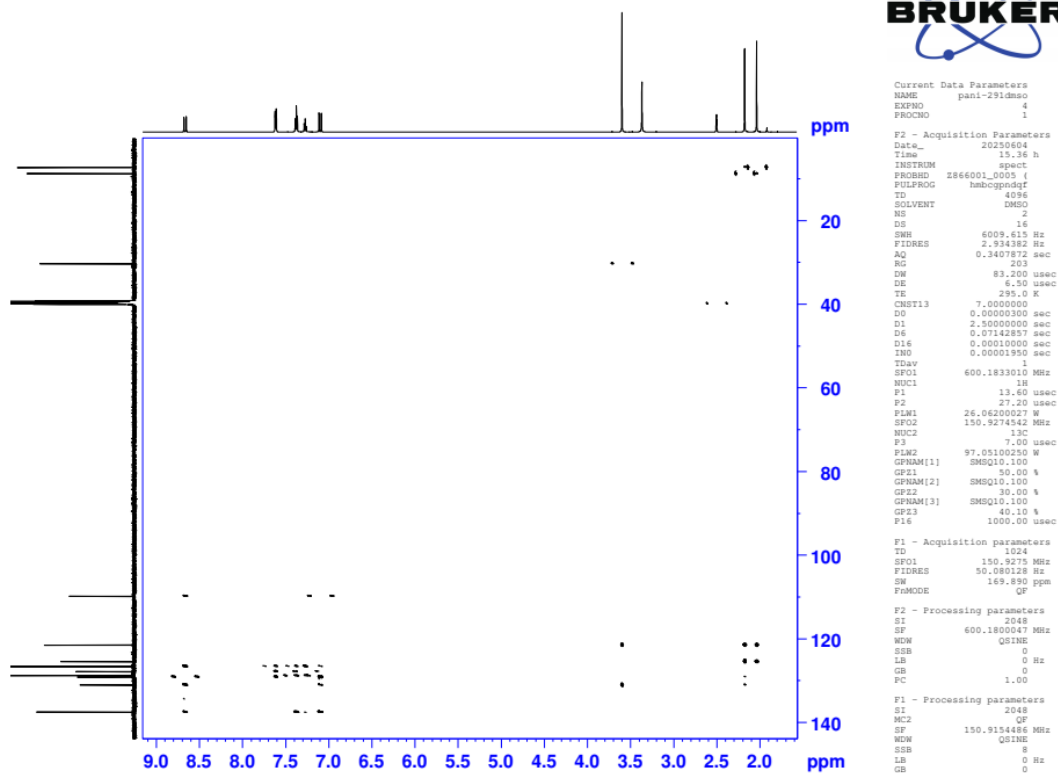


Fig. S 89. ¹H-¹³C HMBC spectrum of compound 6d (600 MHz – ¹H, 151 MHz – ¹³C, DMSO-*d*₆).

¹H-¹³C HMBC

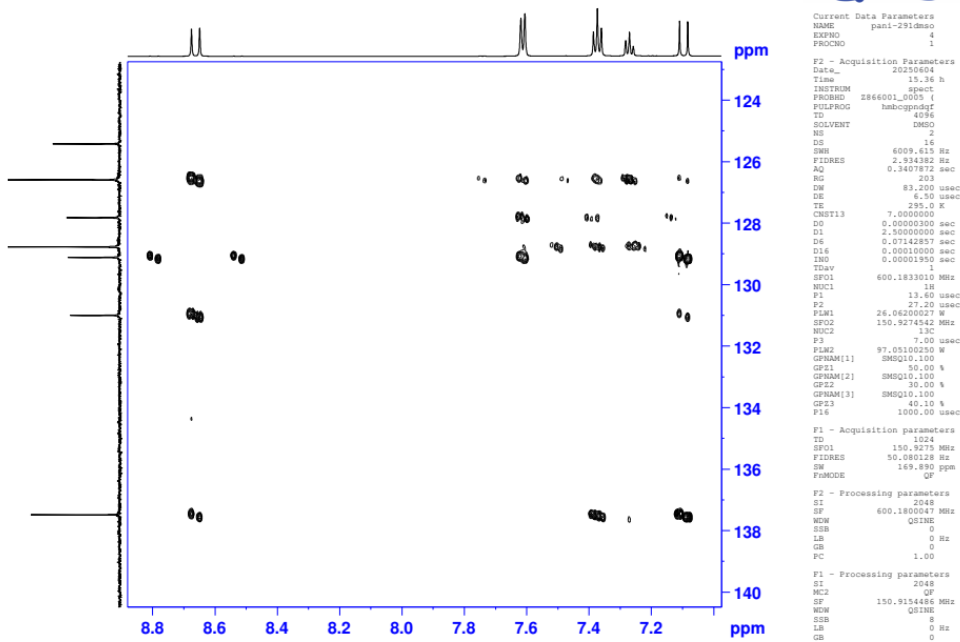
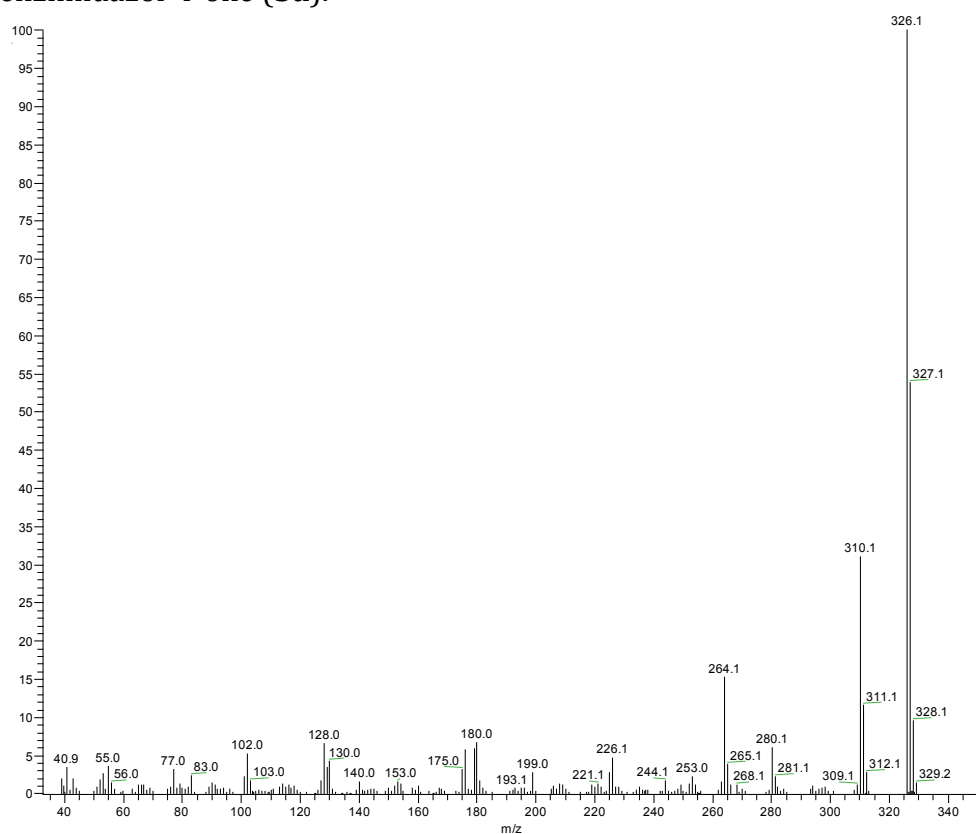


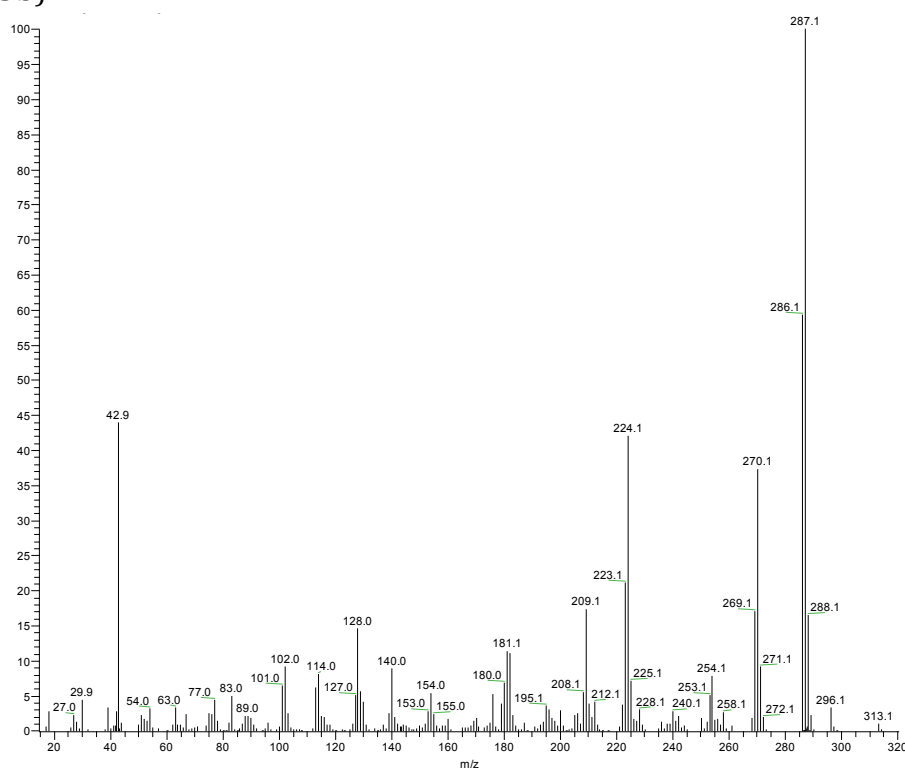
Fig. S 90. Part of ¹H-¹³C HMBC spectrum of compound 4d (600 MHz – ¹H, 151 MHz – ¹³C, DMSO-*d*₆).

SI 5. Copies of HRMS spectra of compounds 3a-h, 4a-h, 5, 6a-d.

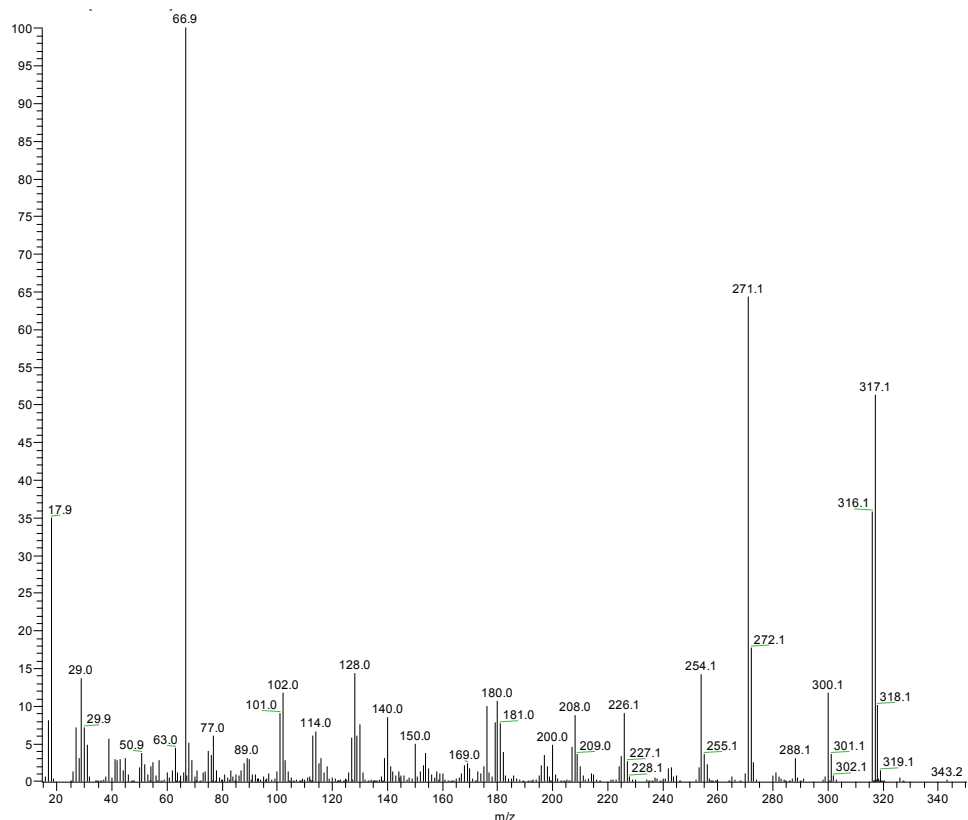
S5.1. 3-Hydroxy-6,6-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (3a).



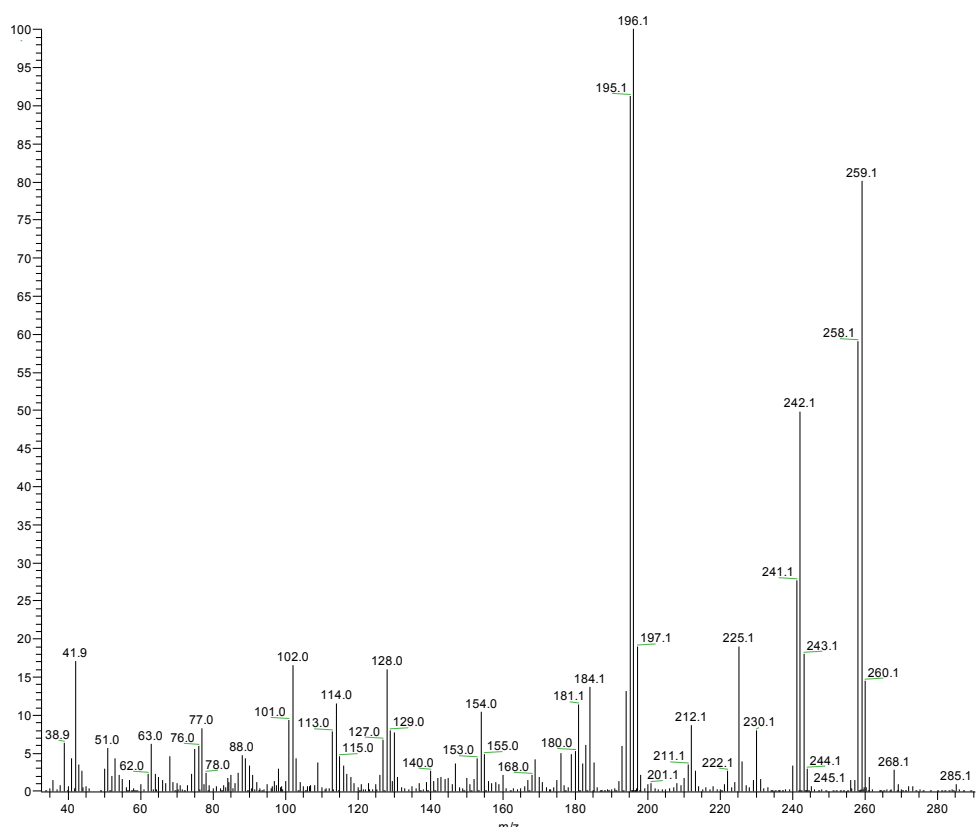
S5.2. 1-(1-Hydroxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazol-5-yl)ethanone (3b).



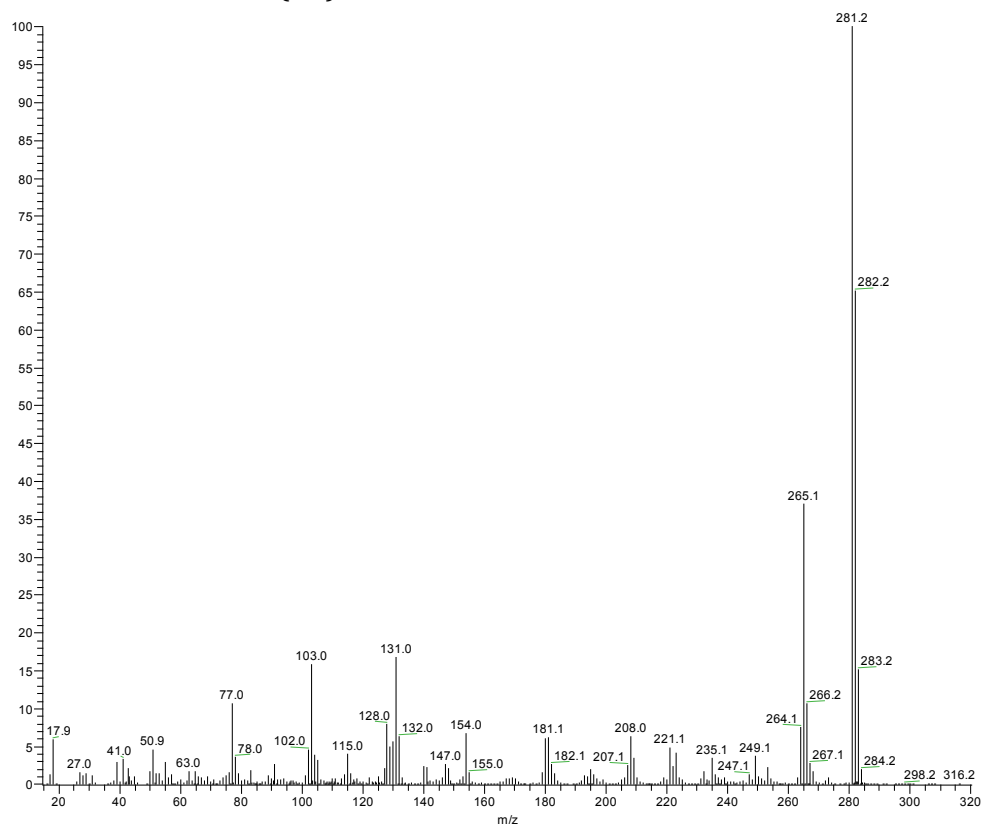
S5.3. Ethyl 1-hydroxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole-5-carboxylate (**3c**).



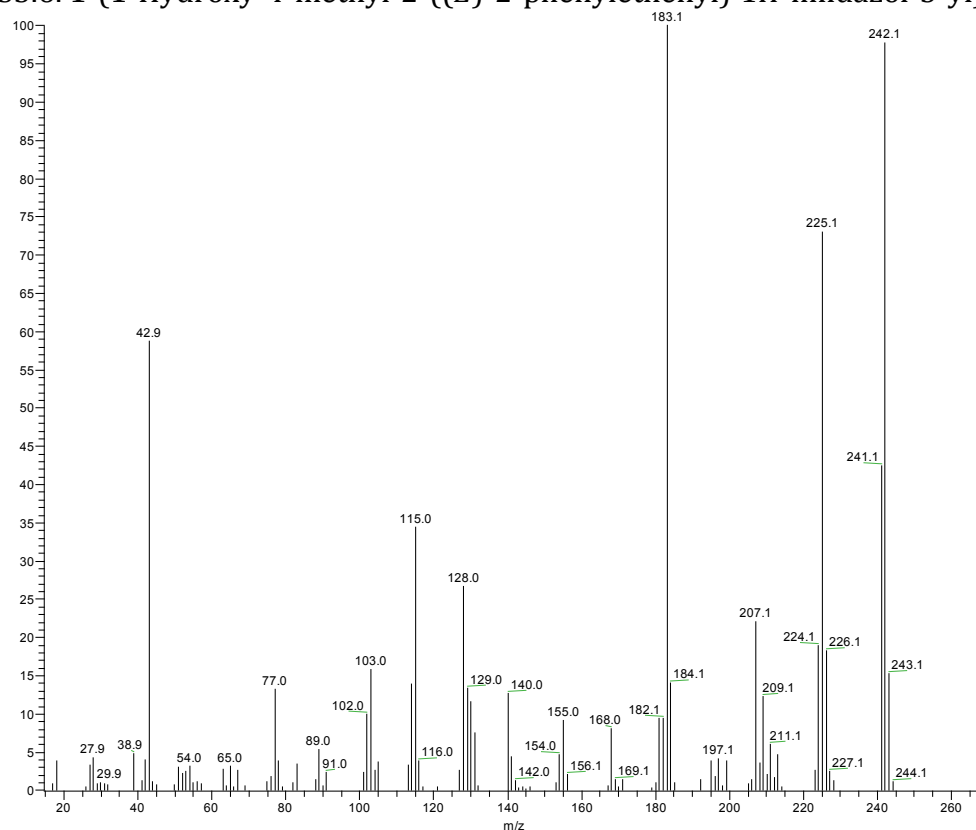
S5.4. 4,5-Dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazol-1-ol (**3d**).



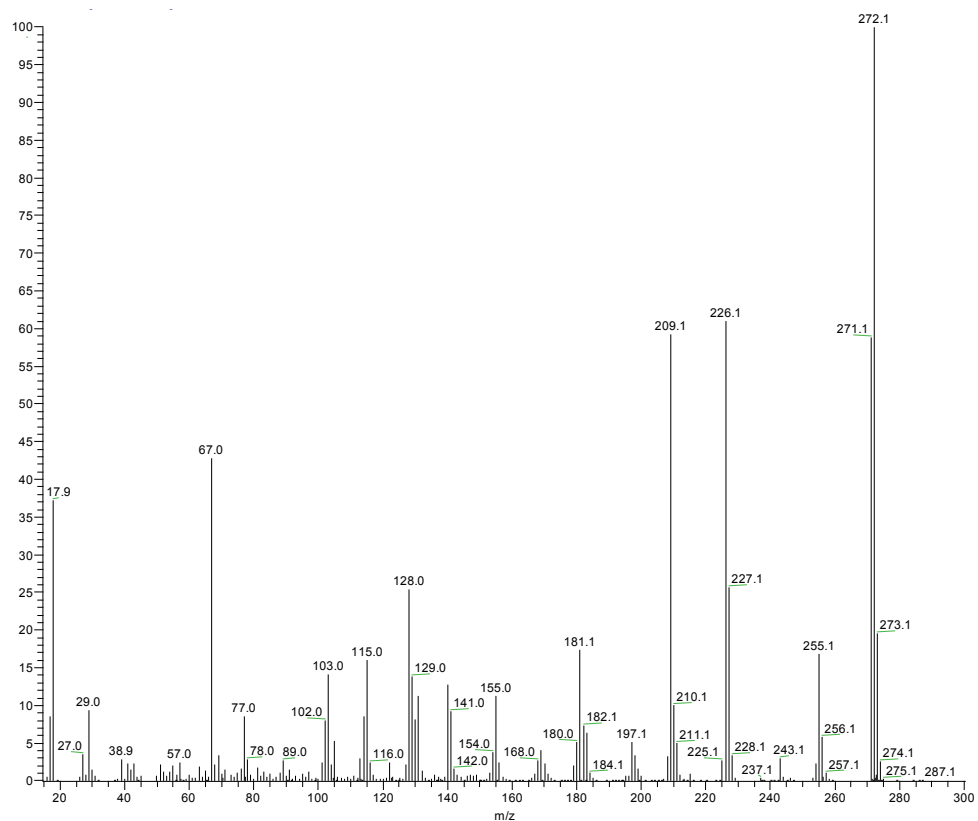
S5.5. 3-Hydroxy-6,6-dimethyl-2-((*E*)-2-phenylethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (**3e**).



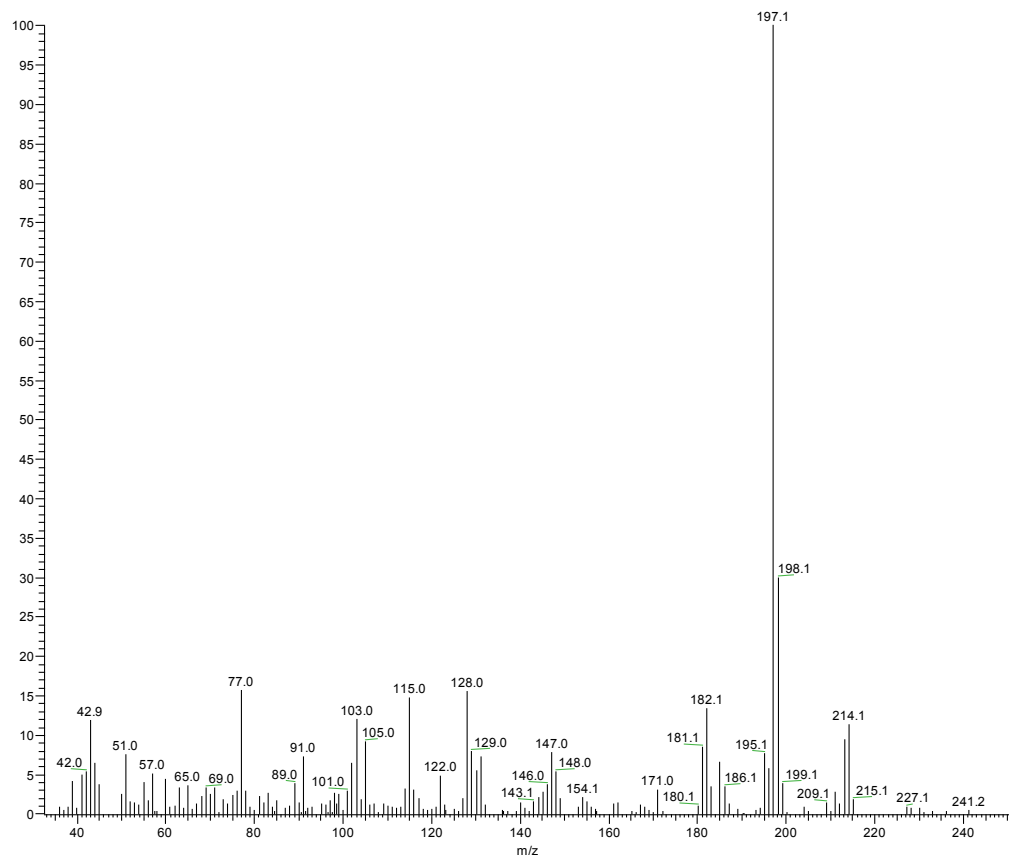
S5.6. 1-(1-Hydroxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazol-5-yl)ethanone (**3f**).



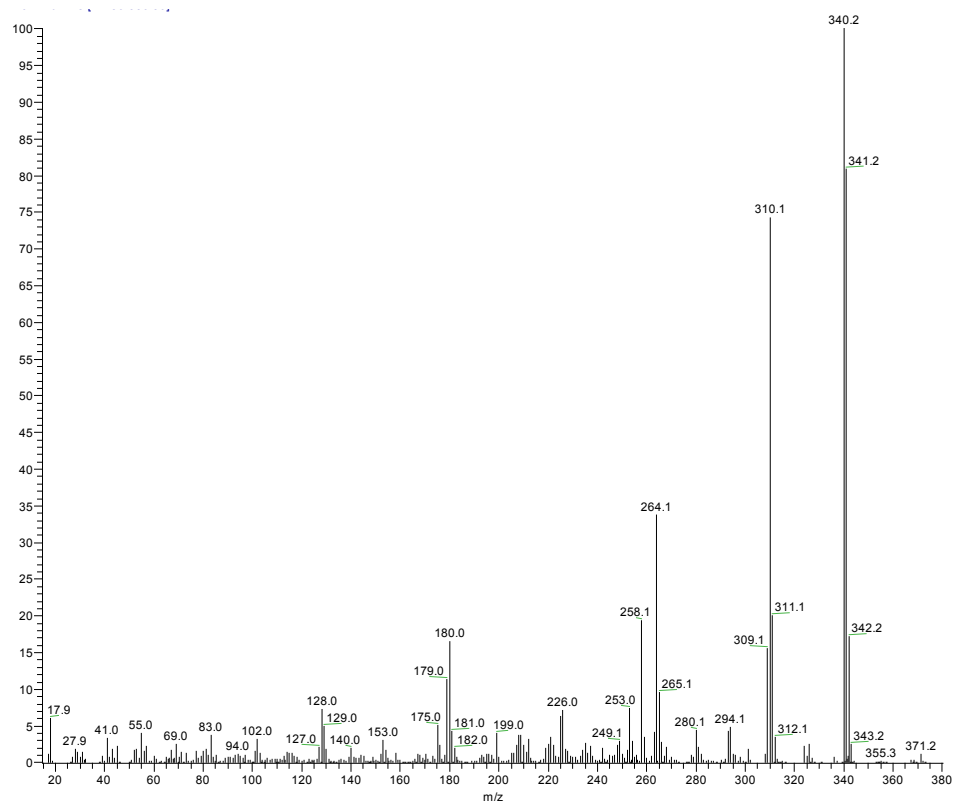
S5.7. Ethyl 1-hydroxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole-5-carboxylate (**3g**).



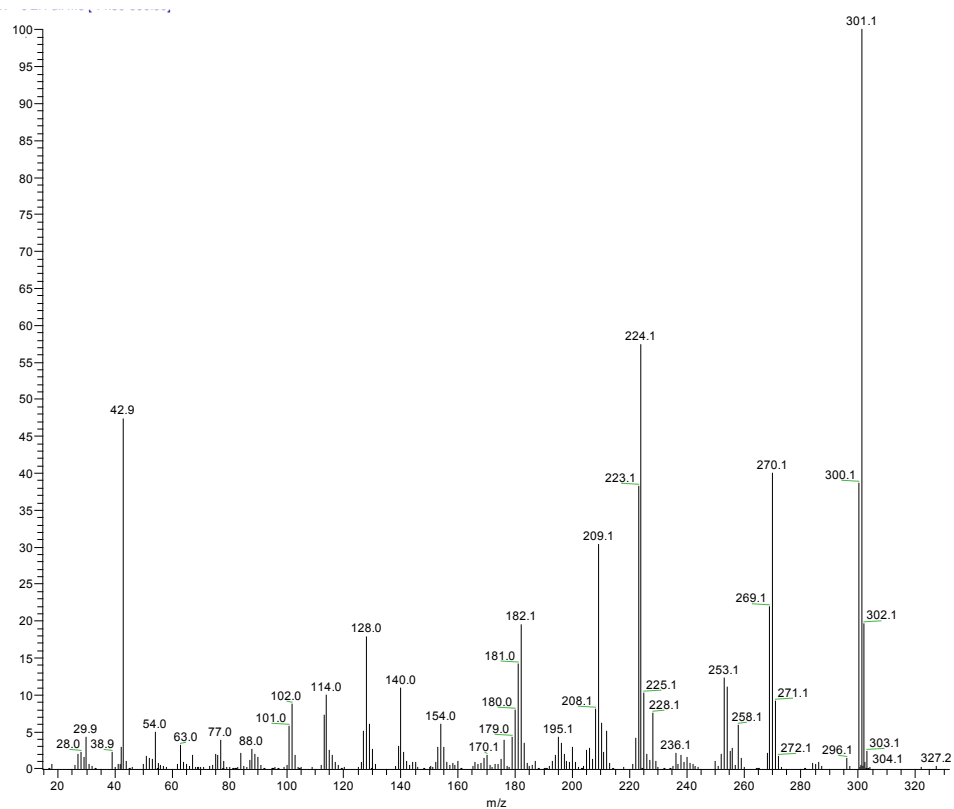
S5.8. 4,5-Dimethyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazol-1-ol (**3h**).



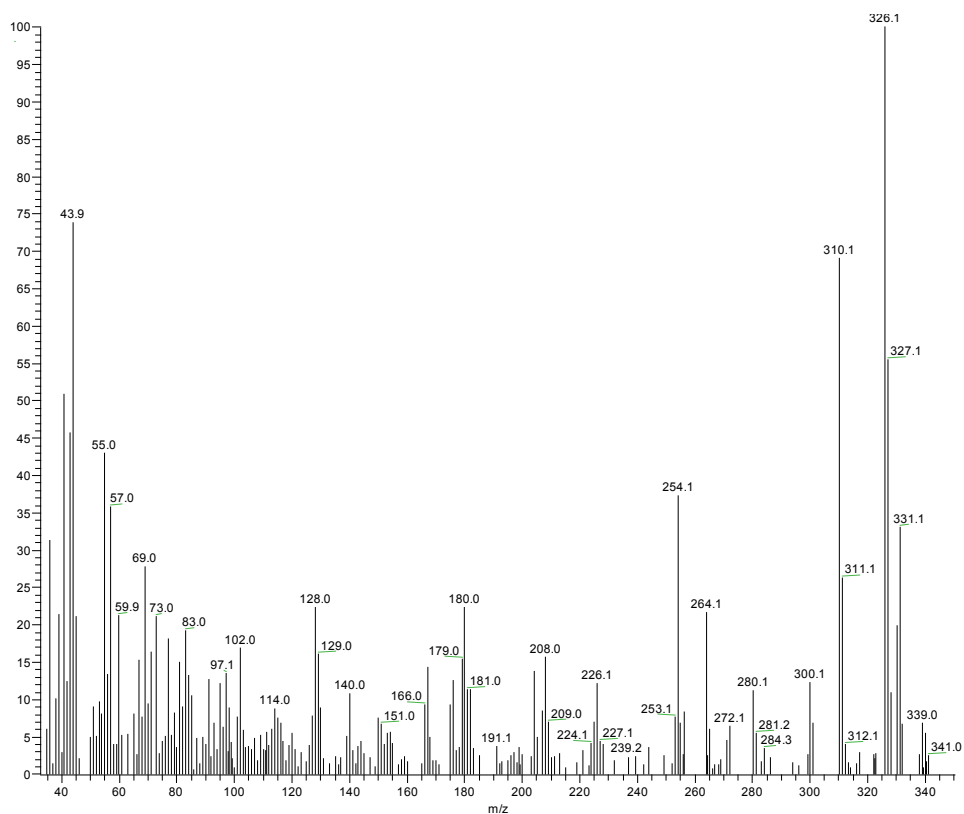
S5.9. 3-Methoxy-6,6-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (**4a**).



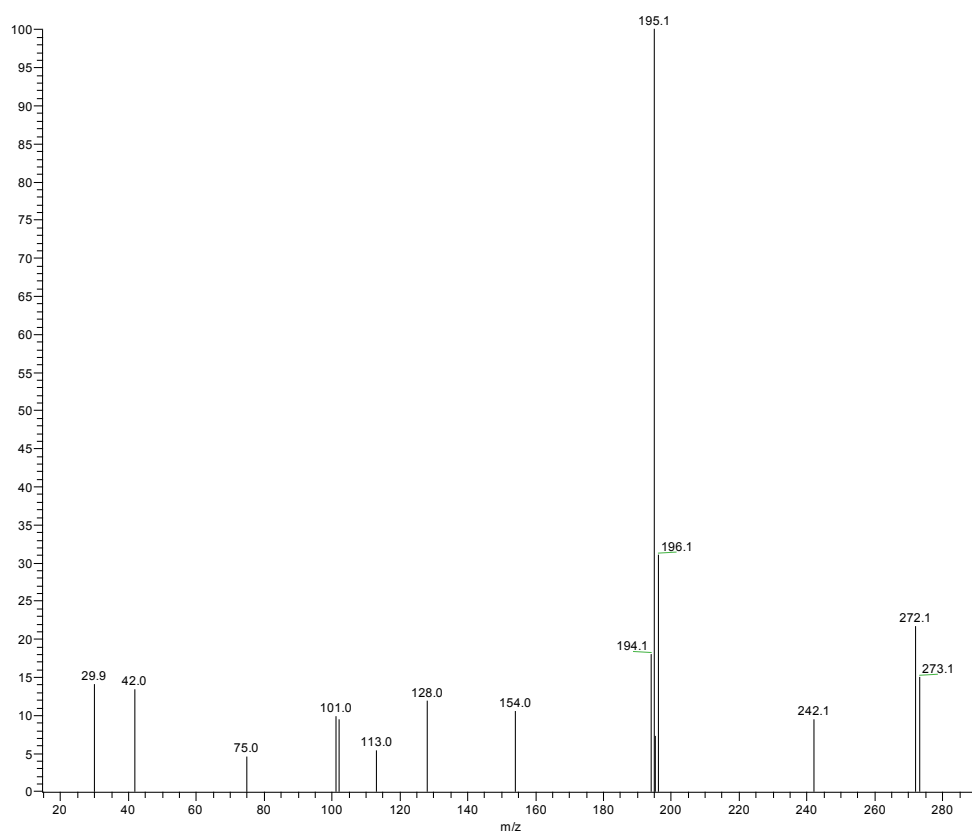
S.5.10. 1-(1-Methoxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazol-5-yl)ethanone (**4b**).



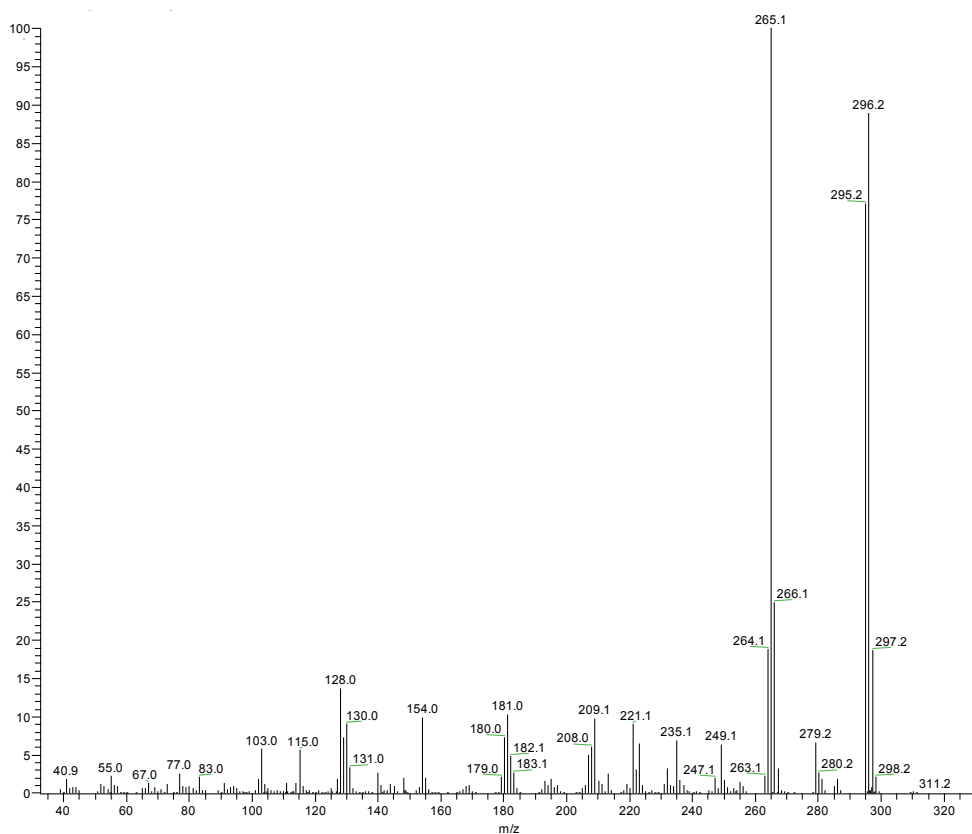
S.5.11. Ethyl 1-methoxy-4-methyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole-5-carboxylate (**4c**).



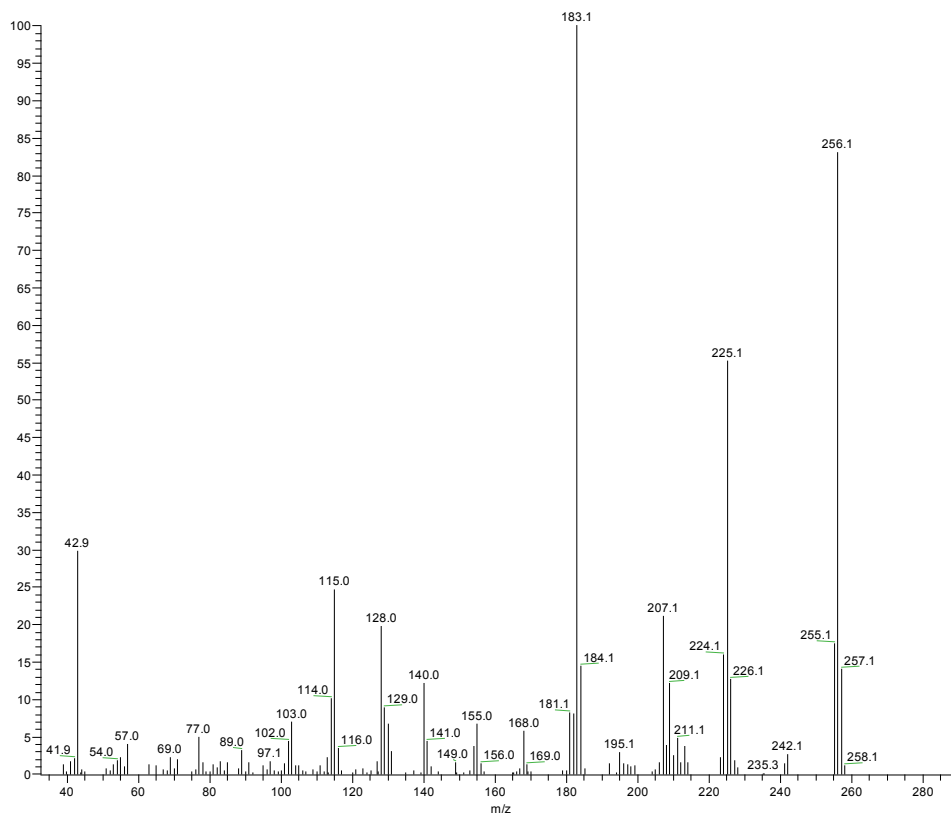
S.5.12. 1-Methoxy-4,5-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole (**4d**).



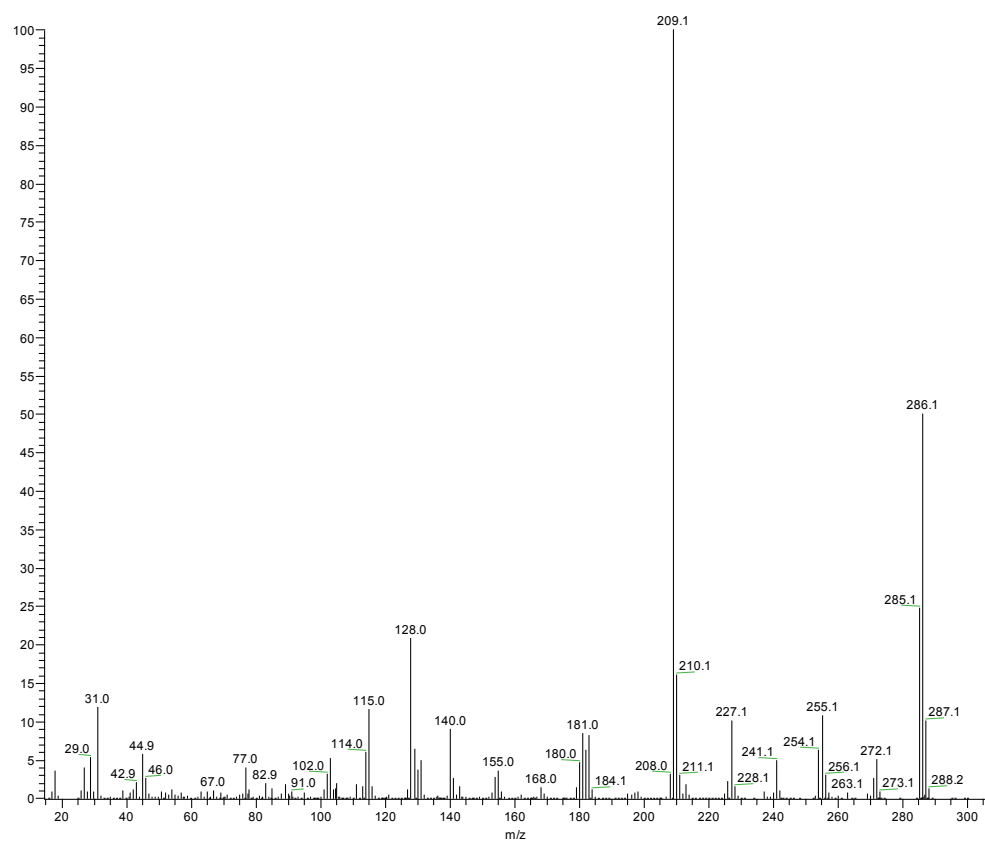
S.5.13. 3-Methoxy-6,6-dimethyl-2-((*E*)-2-phenylethenyl)-3,5,6,7-tetrahydro-4*H*-benzimidazol-4-one (**4e**).



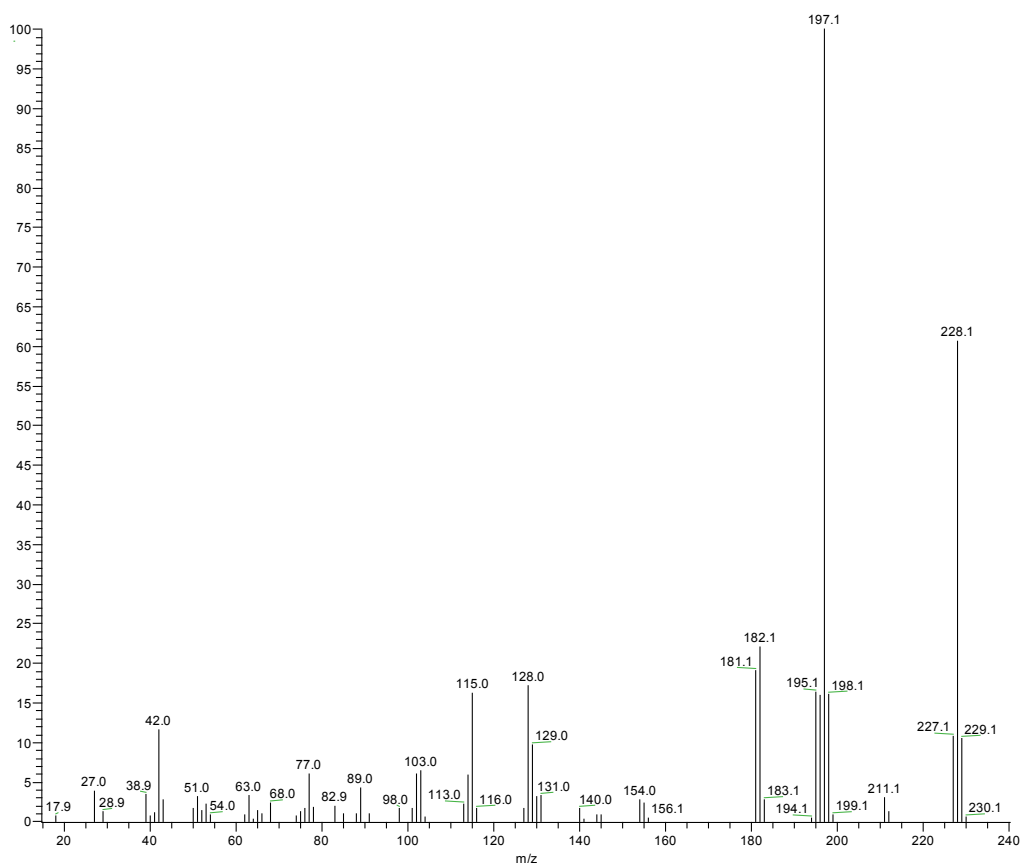
S.5.14. 1-(1-Methoxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazol-5-yl)ethanone (**4f**).



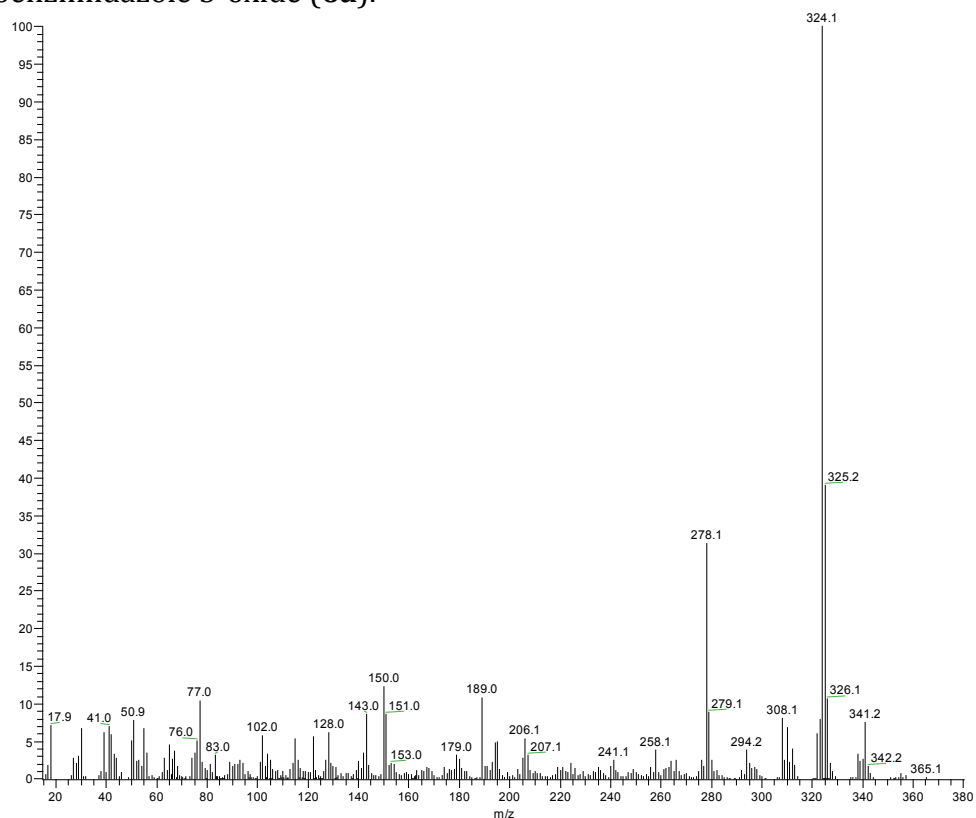
S.5.15. Ethyl 1-methoxy-4-methyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole-5-carboxylate (**4g**).



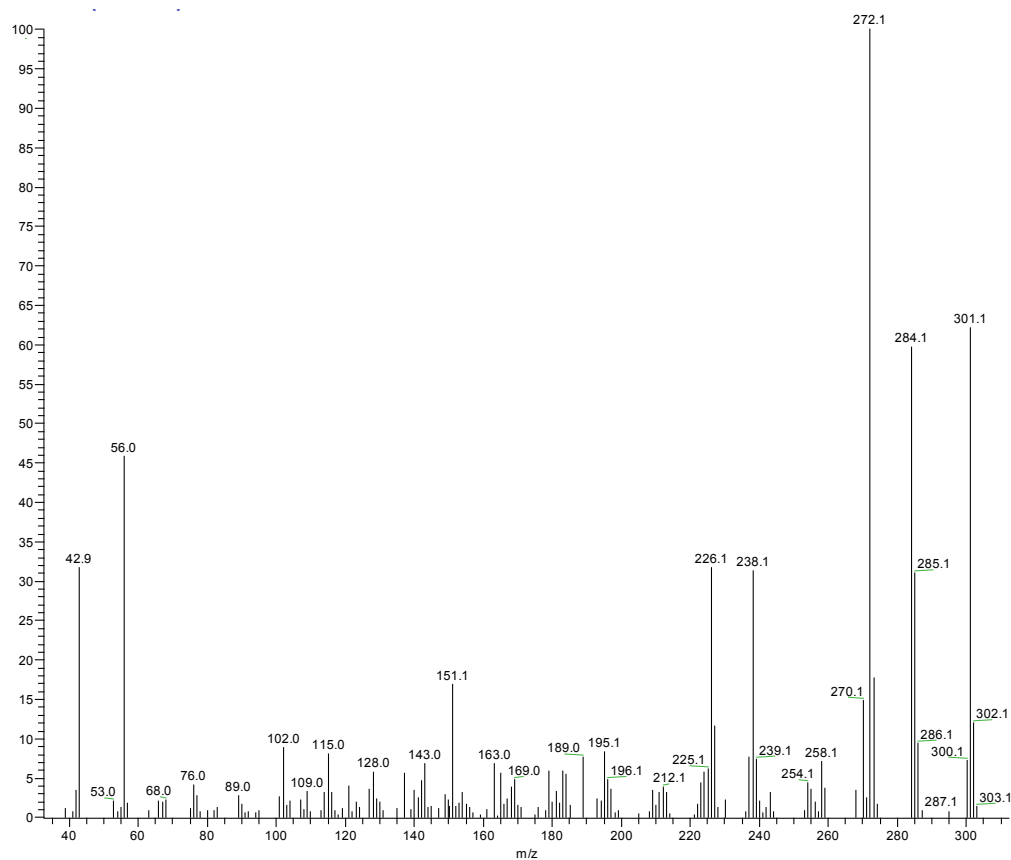
S.5.16. 1-Methoxy-4,5-dimethyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole (**4h**).



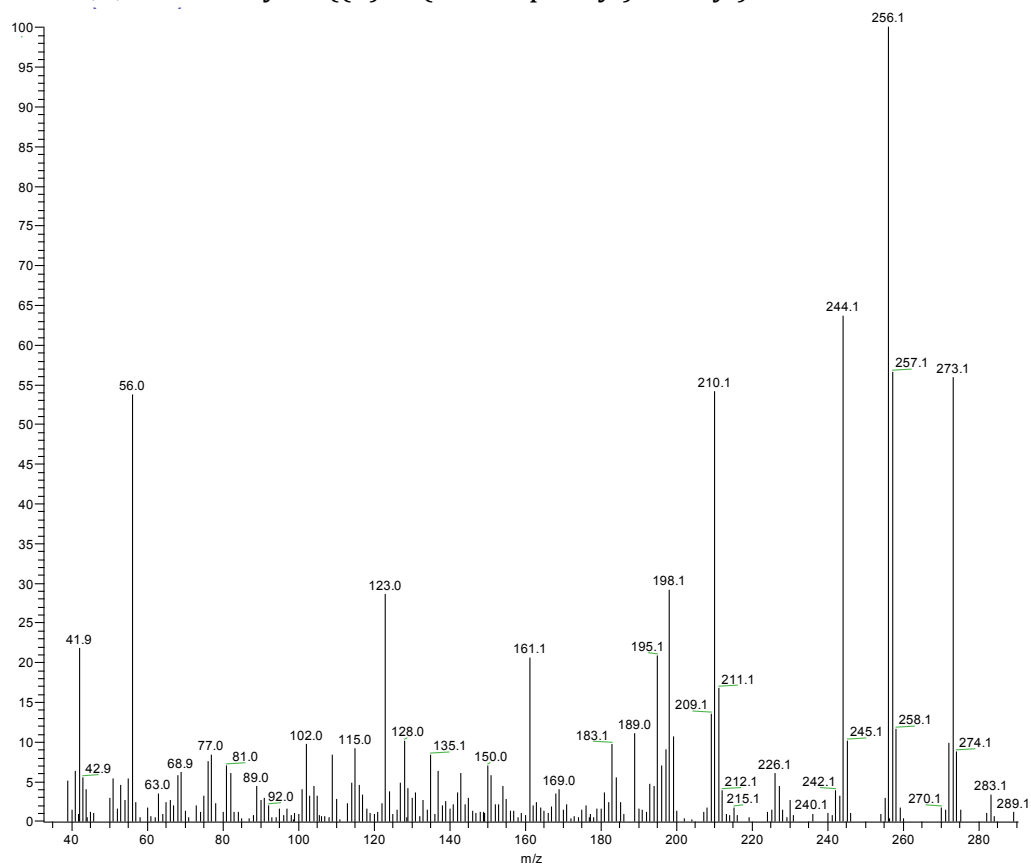
S.5.17. 1,6,6-Trimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-4-oxo-4,5,6,7-tetrahydro-1*H*-benzimidazole 3-oxide (**6a**).



S.5.18. 4-Acetyl-1,5-dimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole 3-oxide (**6b**).



S.5.19. 1,4,5-Trimethyl-2-((*E*)-2-(4-nitrophenyl)ethenyl)-1*H*-imidazole 3-oxide (**6c**).



S.5.20. 1,4,5-Trimethyl-2-((*E*)-2-phenylethenyl)-1*H*-imidazole 3-oxide (**6d**).

