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Supplementary Information

Switching the three-component Biginelli-like reaction conditions for the regioselective synthesis of new 2-amino[1,2,4]triazolo[1,5-*a*]pyrimidines

Martina Pacetti,[a,#] Maria Chiara Pismataro,[a,#] Tommaso Felicetti,[a,*] Federica Giammarino,[b] Anna Bonomini,[c] Matteo Tiecco,[d] Chiara Bertagnin,[c] Maria Letizia Barreca,[a] Raimondo Germani,[e] Violetta Cecchetti,[a] Ilaria Vicenti,[b] Oriana Tabarrini,[a] Maurizio Zazzi,[b] Arianna Loregian,[c] and Serena Massari.[a]

[a] Department of Pharmaceutical Sciences, University of Perugia, 06123 Perugia, Italy;

[b] Department of Medical Biotechnologies, University of Siena, 53100 Siena, Italy;

[c] Department of Molecular Medicine, University of Padua, 35121 Padua, Italy;

[d] Chemistry Interdisciplinary Project (ChIP), School of Pharmacy, University of Camerino, 62032 Camerino, MC, Italy;

[e] Department of Chemistry, Biology and Biotechnology, University of Perugia, 06123 Perugia, Italy.

*Corresponding author: Tommaso Felicetti, Department of Pharmaceutical Sciences, University of Perugia, 06123 Perugia, Italy; Tel: +39 075-5852185, *E-mail:* tommaso.felicetti@unipg.it;

[#] Co-first authors

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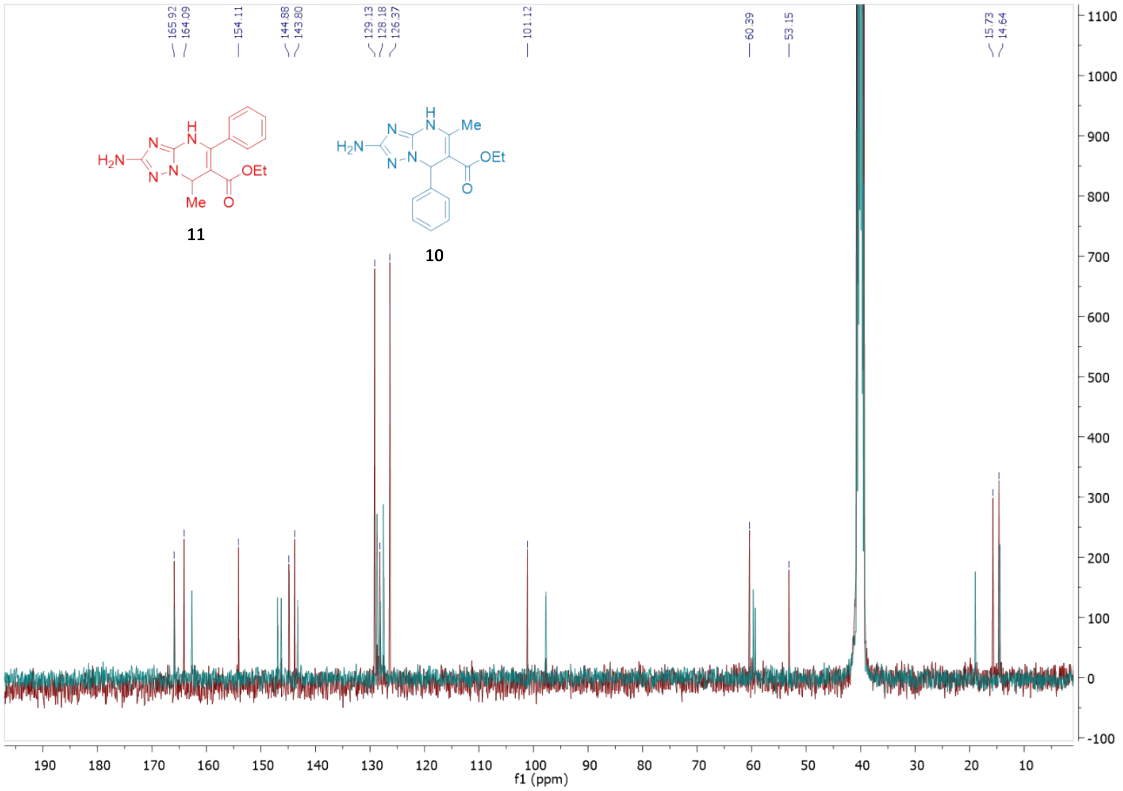


Figure S1. Superposition of ^{13}C NMR spectra of compounds **10** and **11**.

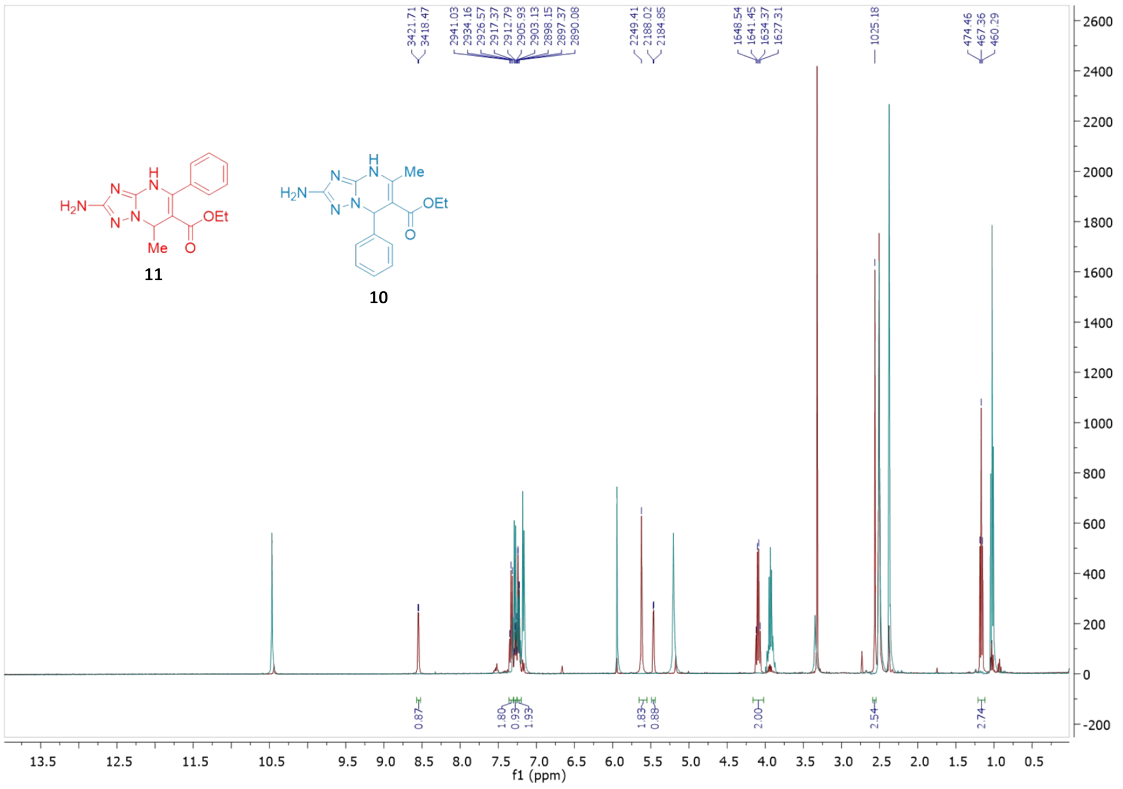


Figure S2. Superposition of ^{1}H NMR spectra of compounds **10** and **11**.

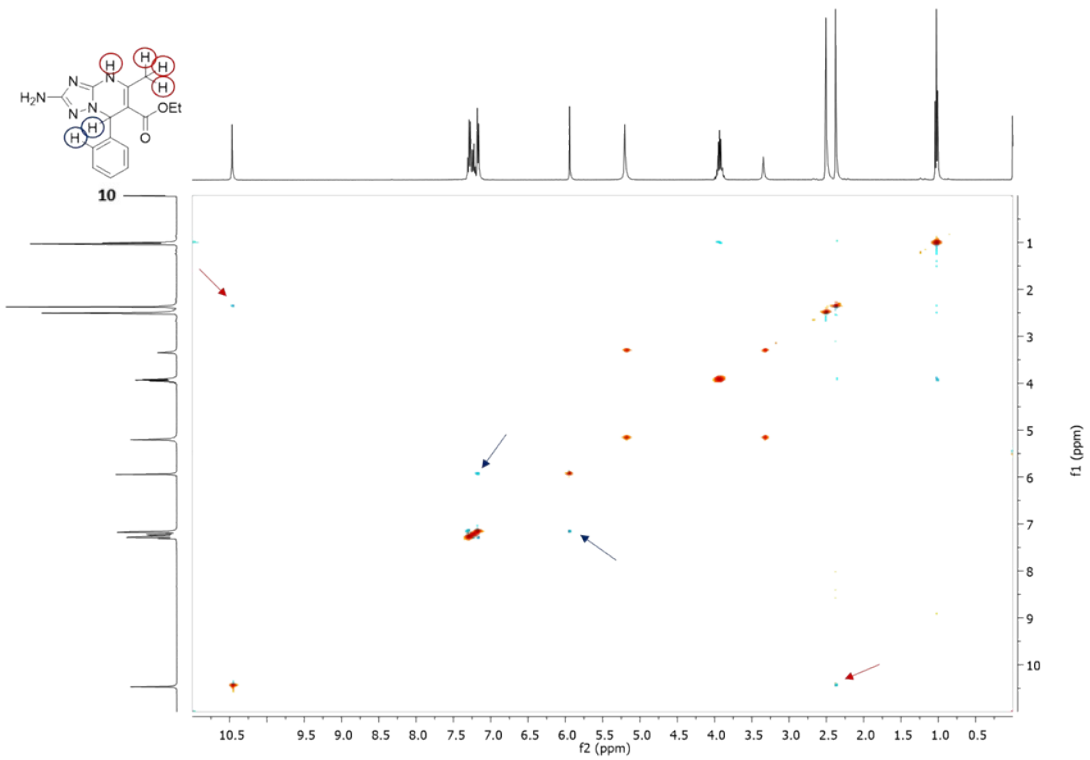


Figure S3. NOESY spectrum of compound **10**.

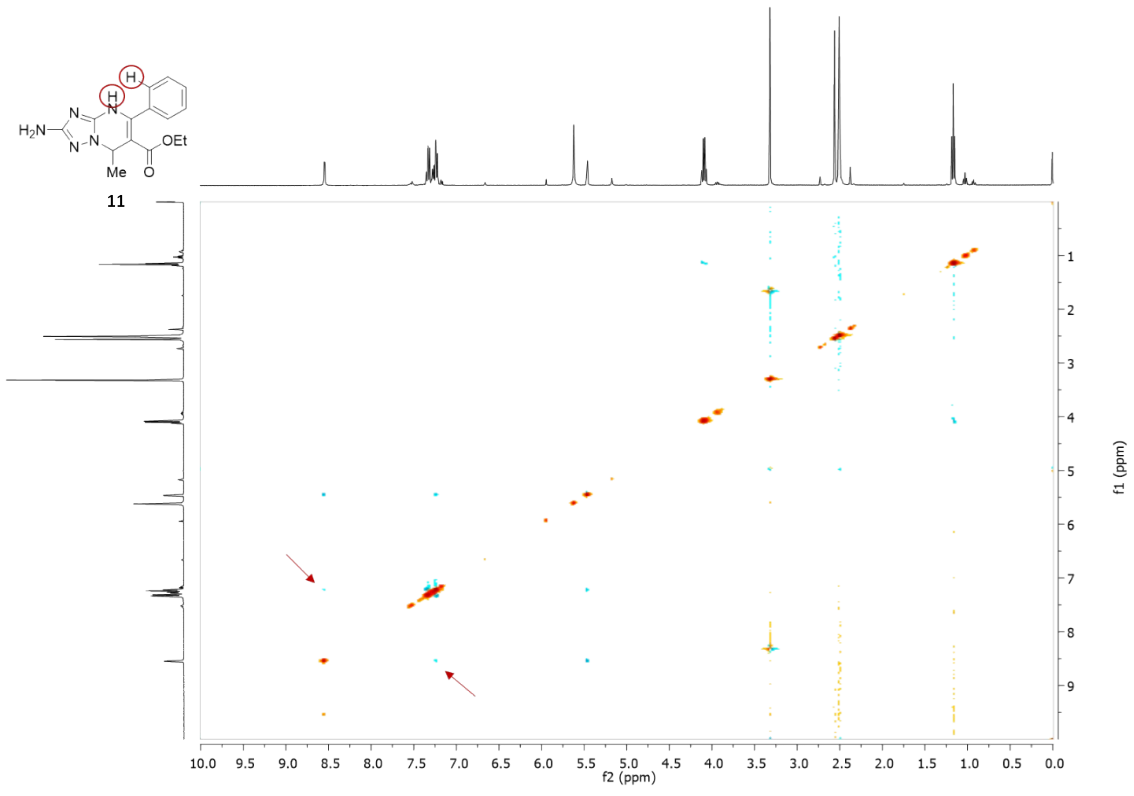


Figure S4. NOESY spectrum of compound **11**.

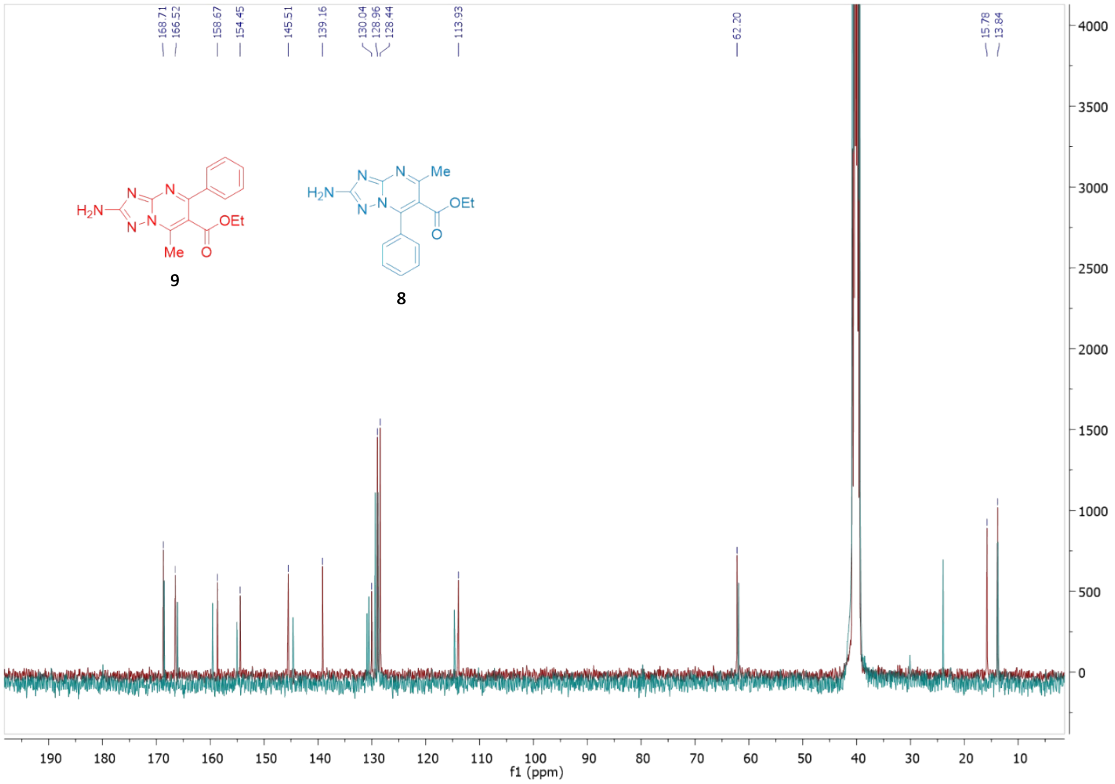


Figure S5. Superposition of ^{13}C NMR spectra of compounds **8** and **9**.

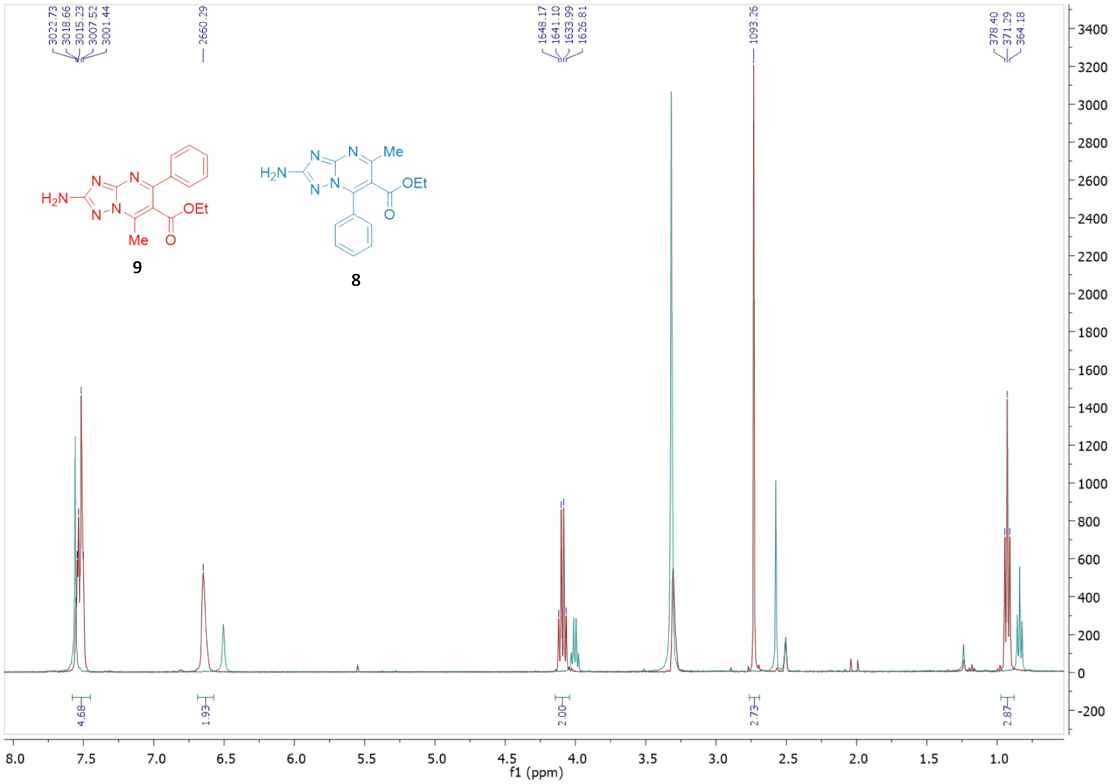
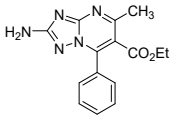
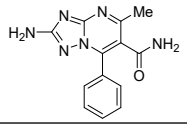
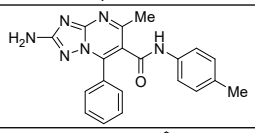
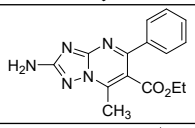
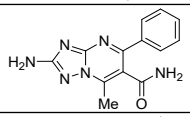
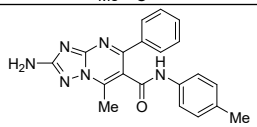
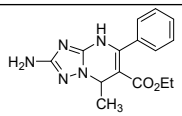
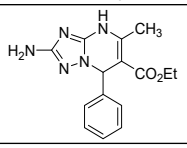


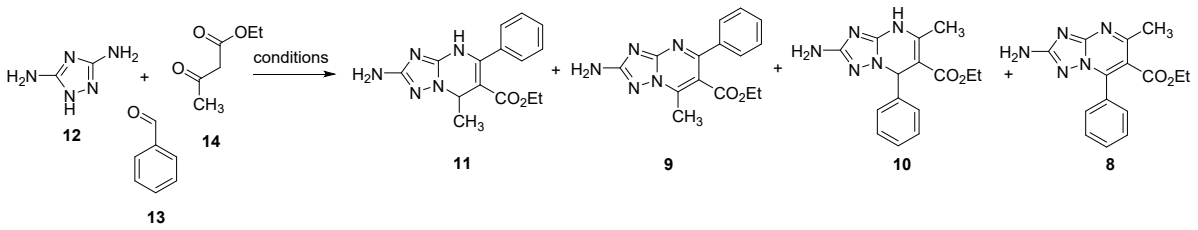
Figure S6. Superposition of ^{1}H NMR spectra of compounds **8** and **9**.

Table S1. ^{13}C NMR chemical shifts (δ, ppm) of compounds **8-11 and 16-19**.[a]

Compd	Structure	C-2	C-3	C-5	C-6	C-7	6-CO_2Et 6-CONHR	5/7-CH_3
8		159.56	144.63	168.56	114.69	155.04	166.09 61.91 13.79	23.93
17		158.87	142.24	167.99	119.80	154.69	167.16	23.24
19		159.13	142.90	168.24	119.41	154.90	163.53	23.24
9		158.67	145.51	168.71	113.9	154.45	166.52 62.20 13.84	15.79
16		155.73	142.02	167.29	117.78	153.07	166.52	14.47
18		157.24	143.85	168.61	118.61	154.41	164.01	15.68
11		164.09	144.88	154.11	101.12	53.15	165.92 60.40 14.64	15.73
10		162.68	146.28	146.97	97.74	59.73	165.87 59.32 14.46	18.98

[a] ^{13}C NMR spectra of compounds in DMSO-d_6 recorded on Bruker Avance DRX-400MHz.

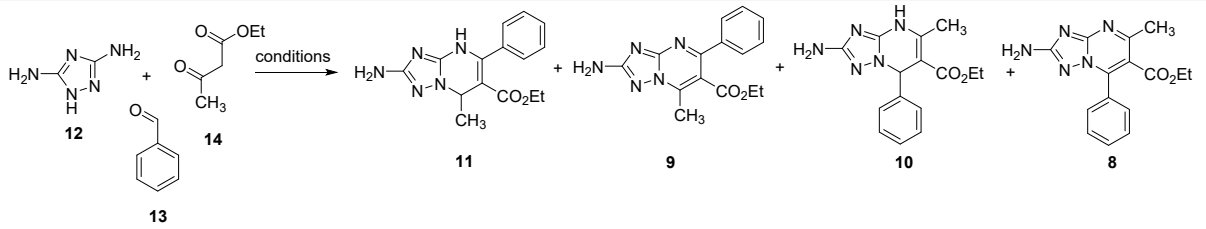
Table S2. Optimization of reaction conditions for **9** and **11** .[a]



Entry	Solvent	Ratio 12:13:14	Catalyst (equiv)	T°	Time (h)	% Ratio 11:9:10:8[b]			
						11	**9**	**10**	**8**
1	EtOH	1:1:1	citric acid (2.5)	reflux	5	66 16%[c]	22	12	-
2	EtOH	1:1:1	citric acid (5)	reflux	4	58	32	8	2
3	EtOH	1:1.5:1	citric acid (2.5)	reflux	4	64	23	12	1
4	EtOH	1:1:1	citric acid (2.5)	reflux	3.5	N.D.[d] **83%**[c]	N.D.	N.D.	N.D.
5	EtOH	1:1:1	-	100 °C µw	20’	-	-	-	-
6[e]	dry THF	1:1:1	PTSA (1)	reflux	24	36	55	7	2
7	-	1:1:1	phosphoric acid	120 °C	6	-	-	-	-
8	-	1:1:1	PPA	120 °C	6	-	-	-	-
9[e]	AcOH	1:1:1	-	reflux	6	11	61 21%[c]	18	10

[a] The reaction was performed on 1.0 mmol scale of **12** in 3 mL of solvent. [b] Percentage ratio among isomers assessed by HPLC on the crude product. [c] Isolated yield. [d] N.D. = not determined due to the presence of only compound **11** by TLC. [e] Reaction performed under nitrogen.

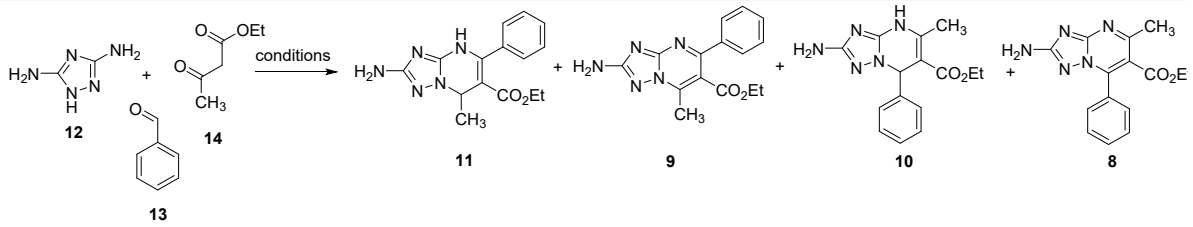
Table S3. Optimization of reaction conditions for **9**.[a]



Entry	Solvent	Ratio 12:13:14	Catalyst (equiv)	T°	Time (h)	% Ratio 11:9:10:8[b]			
						11	**9**	**10**	**8**
1	AcOH	1:1:1	-	120 °C	3	17	78	4	1
2	AcOH	1:1:1	-	120 °C	6	6	89 37%[c]	4	1
3	AcOH	1:1:1	-	120 °C	12	2	92	4	2
4	AcOH	1:1:1	-	120 °C	24	2	90	5	3
5	AcOH	1:1:1	-	60 °C	3	45	47	8	-
6	AcOH	1:1:1	-	60 °C	24	25	69	5	1
7	AcOH	2:1:1	-	120 °C	24	13	53	19	15
8	AcOH	1:2:1	-	120 °C	5	12	68	13	7
9	AcOH	1:3:1	-	120 °C	24	7	56	31	6
10	AcOH	1:1:2	-	120 °C	24	6	85	4	5
11	AcOH	1:1:3	-	120 °C	9	6	76	6	12

[a] The reaction was performed on 1.0 mmol scale of **12** in 3 mL of solvent in an open flask. [b] Percentage ratio among isomers assessed by HPLC on the crude product. [c] Isolated yield.

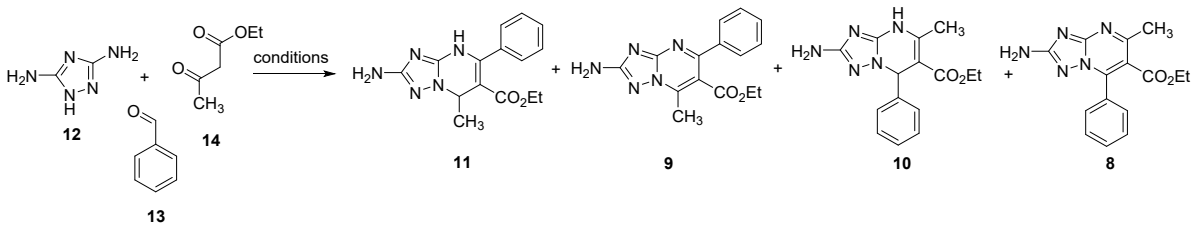
Table S4. Optimization of reaction conditions for **9** and **11**.[a]



Entry	Solvent	Ratio 12:13:14	Catalyst (equiv)	T°	Time (h)	% Ratio 11:9:10:8[b]			
						11	**9**	**10**	**8**
1	AcOH	2:1:1	-	reflux	24	6	56	19	19
2	AcOH	1:2:1	-	reflux	24	13	77 51%[c]	9	1
3	AcOH	1:2:1	-	60 °C	24	24	63 33%[c]	9	4
4	AcOH	1:3:1	-	60 °C	9	13	78 **55%**[c]	7	2
5	AcOH	1:3:1		90 °C	9	32	39	23	6
6	AcOH	1:3:1		reflux	9	29	34	25	12
7	AcOH	1:1:2	-	60 °C	24	26	70	3	1
8	AcOH	1:1:2	-	90 °C	24	21	60	11	8
9	AcOH	1:1:2	-	reflux	12	7	83 23%[c]	6	4
10	AcOH	1:3:2	-	60 °C	12	10	85 36%[c]	3	2
11	AcOH	1:3:1		60 °C µw	2.5	31	48	19	2
12	AcOH	1:3:1	H_2O_2[d]	60-110 °C µw	2.75	7	72 12%[c]	7	15
13	AcOH	1:3:1	I_2 (0.5)	60 °C	24	8	80	5	7

[a] The reaction was performed on 1.0 mmol scale of **12** in 3 mL of solvent under nitrogen. [b] Percentage ratio among isomers assessed by HPLC on the crude product. [c] Isolated yield. [d] After 2 h at 60 °C, H_2O_2 (1 mL) was added and the reaction was heated at 110 °C for 45'.

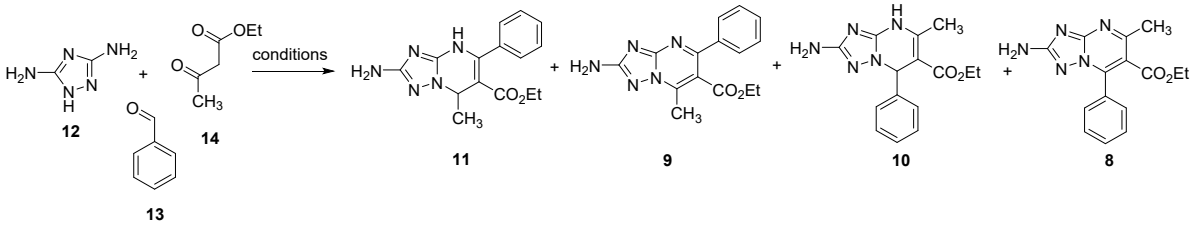
Table S5. Optimization of reaction conditions for **8** and **10**.[a]



Entry	Solvent	Ratio 12:12:14	Catalyst	T°	Time (h)	% Ratio 11:9:10:8[b]			
						11	**9**	**10**	**8**
1	BMIM MsO	1:1:1	-	120 °C	48	-	5	2	93 30%[c]
2	TBMA MsO	1:1:1	-	120 °C	24	-	-	100 **75%**[c]	-
3	BMIM TFB	1:1:1	-	120 °C	24	-	38	22	40 25%[c]
4	BMIM MsO	1:2:1	-	120 °C	12	-	-	-	-
5	TBMA MsO	1:2:1	-	120 °C	12	-	-	-	-
6	BMIM MsO	1:2:1	H_2O_2[d]	120 °C	24	-	10	3	87 **40%**[c]
7	TBMA MsO	1:2:1	H_2O_2[d]	120 °C	24	-	20	8	72
8	BMIM TFB	1:2:1	H_2O_2[d]	120 °C	24	1	29	2	68

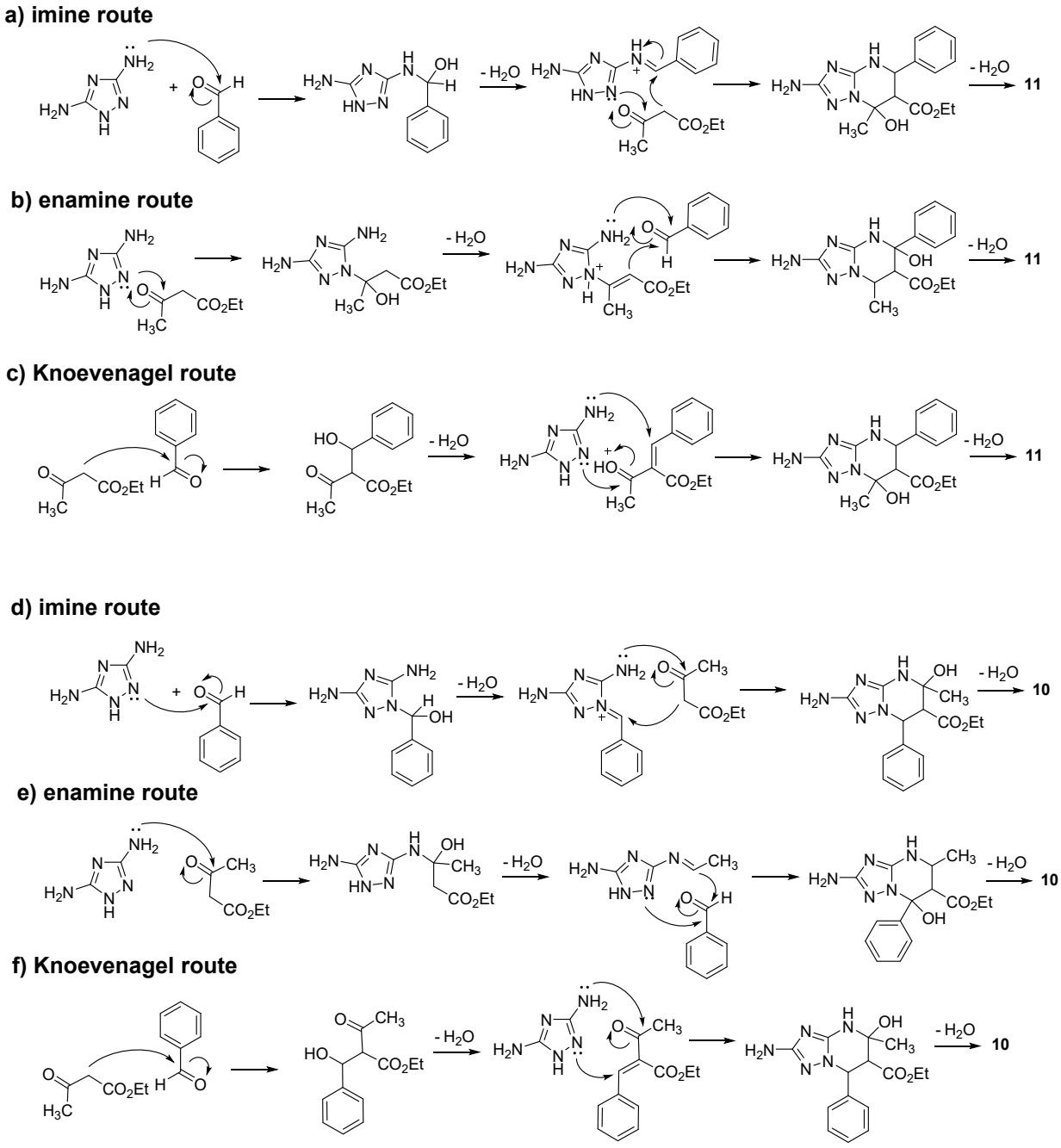
[a] The reaction was performed on 1.0 mmol scale of **12** in 0.2 g of IL in an open flask. [b] Percentage ratio among isomers assessed by HPLC on the crude product. [c] Isolated yield. [d] H_2O_2 (1 mL) was added after 12 h.

Table S6. Optimization of reaction conditions for **8**.[a]



Entry	Solvent	Ratio 12:13:14	Catalyst	T°	Time (h)	% Ratio 11:9:10:8[b]			
						11	**9**	**10**	**8**
1	TBMA TsO	1:2:1	H_2O_2[c]	120 °C	24	-	20	-	80
2	TBMP MsO	1:2:1	H_2O_2[d]	120 °C	7	1	28	25	46
3	MMIM TsO	1:2:1	H_2O_2[d]	120 °C	7	N.D.[f]	N.D.	N.D.	N.D.
4	TBA TsO	1:2:1	H_2O_2[d]	120 °C	7	1	18	31	50
5	TBA bromide	1:2:1	H_2O_2[e]	120 °C	34	-	13	-	87
6	TBA MsO	1:2:1	H_2O_2[c]	120 °C	24	-	7	11	82
7	EG/TMG	1:2:1	H_2O_2[e]	120 °C	34	1	26	1	72
8	Gly/TMG	1:2:1	H_2O_2[e]	120 °C	29	-	50	14	36
9	U/ChCl	1:2:1	H_2O_2[e]	120 °C	34	N.D.	N.D.	N.D.	N.D.

[a] The reaction was performed on 1.0 mmol scale of **12** in 0.2 g di IL or DES in an open flask. [b] Percentage ratio among isomers assessed by HPLC on the crude product. H_2O_2 (1 mL) was added after [c] 20 h, [d] 6 h, or [e] 24 h. [f] N.D. = not determined.



Scheme S1. Plausible reaction mechanisms toward **11** (a-c) and **10** (d-f). (**a**) Imine route entailing an initial direct addition of the amino group at the C(3) position of **12** on the carbonyl carbon of benzaldehyde **13** to give an iminium intermediate, which reacts with ethyl 3-oxobutanoate **14**; (**b**) enamine route entailing an initial direct addition of the N(2) of **12** on the carbonyl carbon C(3) of **14** to give a protonated enamine intermediate, which subsequently reacts with **13**; (**c**) Knoevenagel route entailing an initial direct addition of the carbon C(2) of **14** on the carbonyl carbon of **13** to give the carbenium ion intermediate (Knoevenagel's adduct), which reacts with **12** (a direct addition of the amino group at the C(3) position of **12** on the β-carbon of the adduct and a direct addition of the nucleophilic N(2) center of **12** on the carbonyl carbon C(3) of the adduct); (**d**) imine route entailing an initial direct addition of the N(2) of **12** on the carbonyl carbon of **13** to give an imine intermediate, which reacts with ethyl 3-oxobutanoate **14**; (**e**) enamine route entailing an initial direct addition of the amino group at the C(3) position of **12** on the carbonyl carbon C(3) of **14** to give an enamine intermediate, which subsequently reacts with **13**; (**f**) Knoevenagel route entailing, once formed Knoevenagel's adduct as described in point (c), a direct addition of the amino group at the C(3) position of **12** on the carbonyl carbon C(3) of the adduct and a direct addition of the nucleophilic N(2) center of **12** on the β-carbon of the adduct.

Table S7. Anti-DENV-2, anti-WNV, and anti-SARS-CoV-2 activity, and cytotoxicity of TZP derivatives **23-30** synthesized in this study.

Compd	Anti-DENV-2 activity (Huh7 cells) EC_{50}, µM[a]	Anti-WNV activity (Huh7 cells) EC_{50}, µM[a]	Cytotoxicity (Huh7 cells) CC_{50}, µM[b]	Anti-SARS-CoV-2 activity (A549 cells) EC_{50}, µM[c]	Cytotoxicity (A549 cells) CC_{50}, µM[b]
23	NA	NA	>243	NA	>243
29	NA	NA	>243	NA	>243
24	NA	NA	>243	NA	112.3
25	4.3 ± 1.5	6.7 ± 3.7	20.9	NA	9.8
26	14.1 ± 4.1	19.3 ± 1.4	141.1	NA	99.9
30	NA	NA	>243	NA	>243
27	NA	NA	>243	NA	>243
28	NA	NA	>243	NA	>243
NRM	-	-	-	0.066 ± 0.007	36
SOF	8.1 ± 1.1	5.3 ± 2.5	>243	>243	>243

[a] Activity of the compounds as determined by immunodetection assay. The EC_{50} value represents the compound concentration that reduces by 50% the expression of flavivirus envelope proteins in Huh7 cells infected with DENV or WNV. All the reported values represent the means ± SD of data derived from at least two independent experiments in duplicate. [b] Cytotoxicity of the compounds as determined by Cell Titer assay in A549 and Huh cell lines. The CC_{50} value represents the compound concentration that causes a decrease of cell viability of 50%. [c] Activity of the compounds as determined by Cell Titer. The EC_{50} value represents the compound concentration that reduces by 50% the cytopathic effect in A549 cells infected with SARS-CoV-2. All the reported values represent the means ± SD of data derived from at least two independent experiments in duplicate. [d] NA = not active. [e] ND = not determined due to solubility issues.

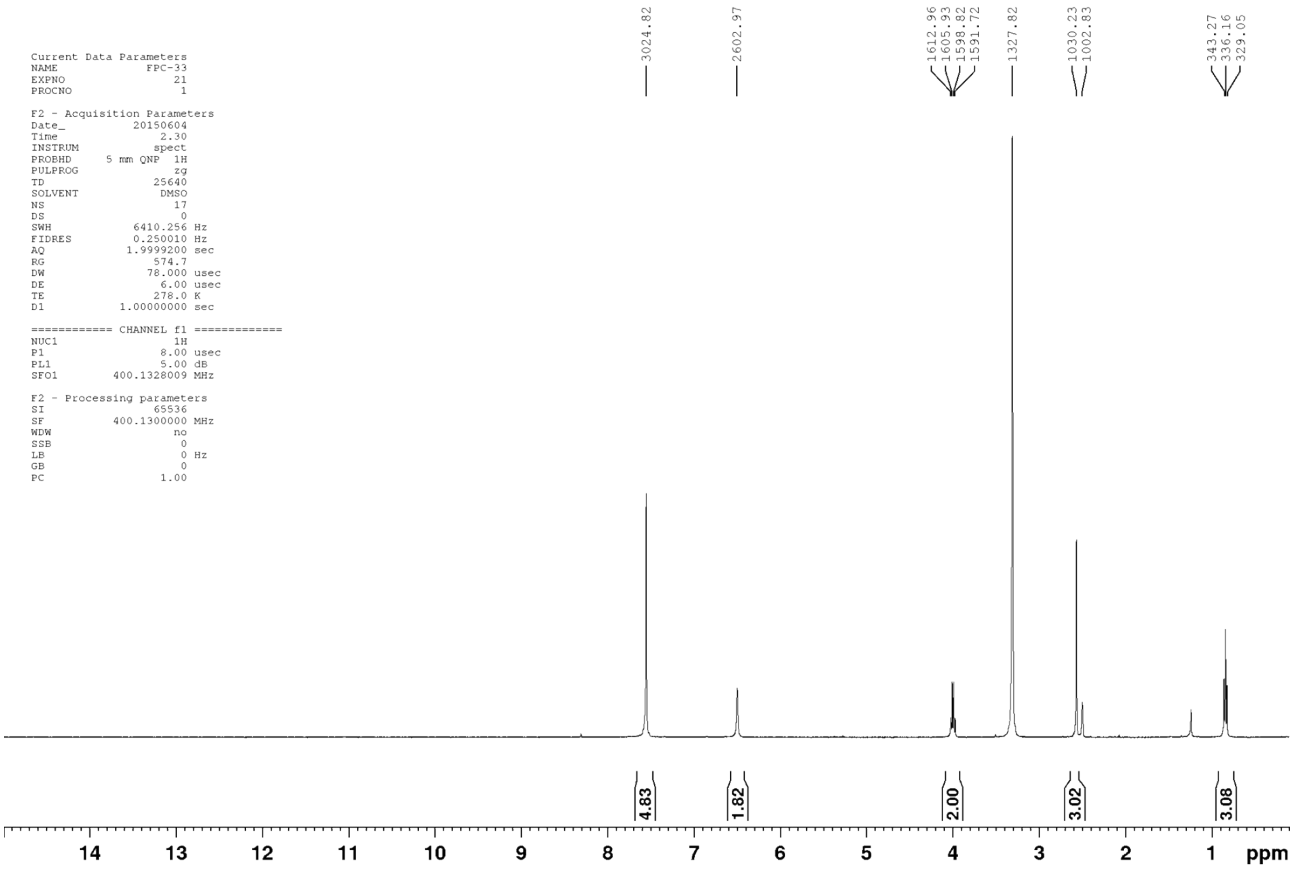


Figure S7. ^{1}H NMR spectrum of compound **8**.

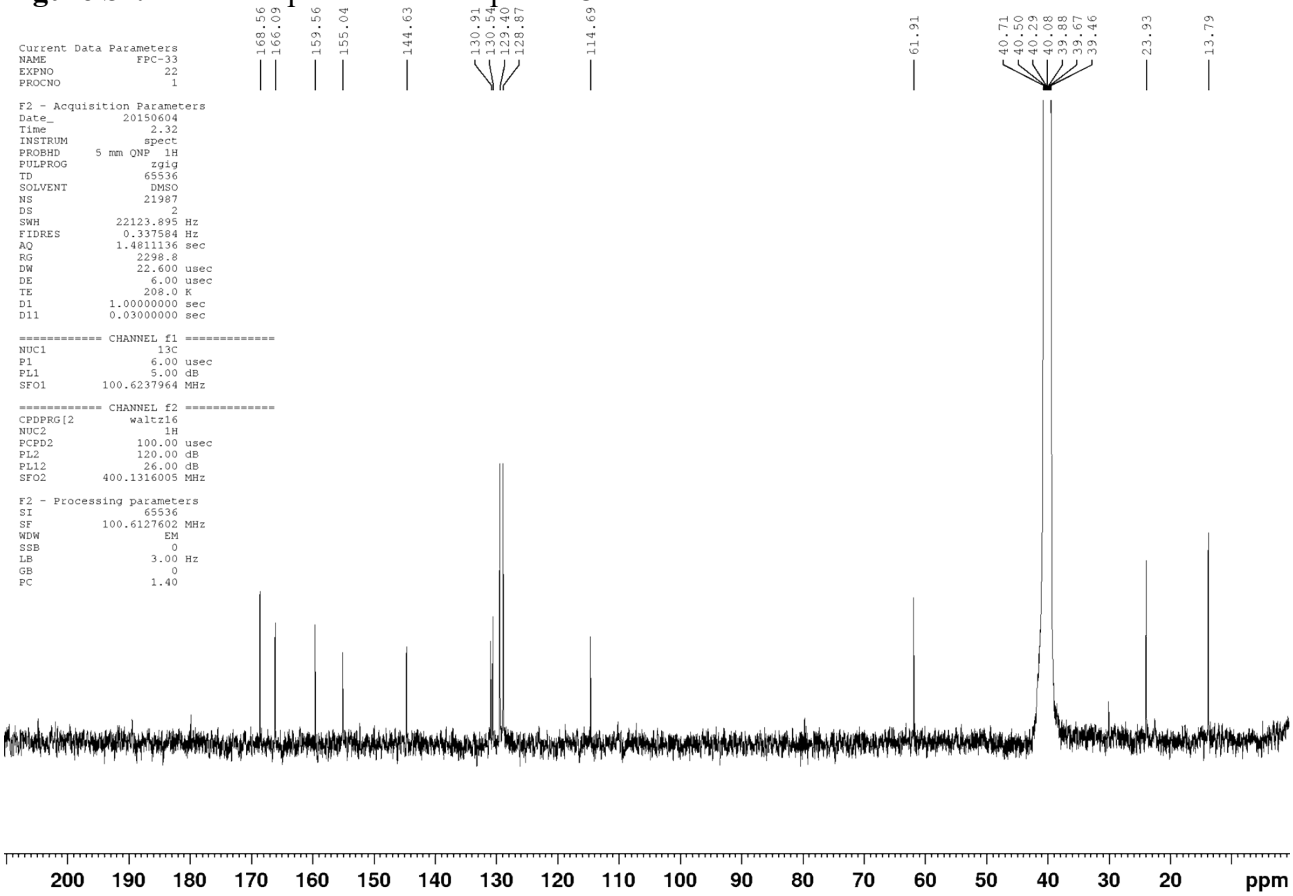


Figure S8. ^{13}C NMR spectrum of compound **8**.

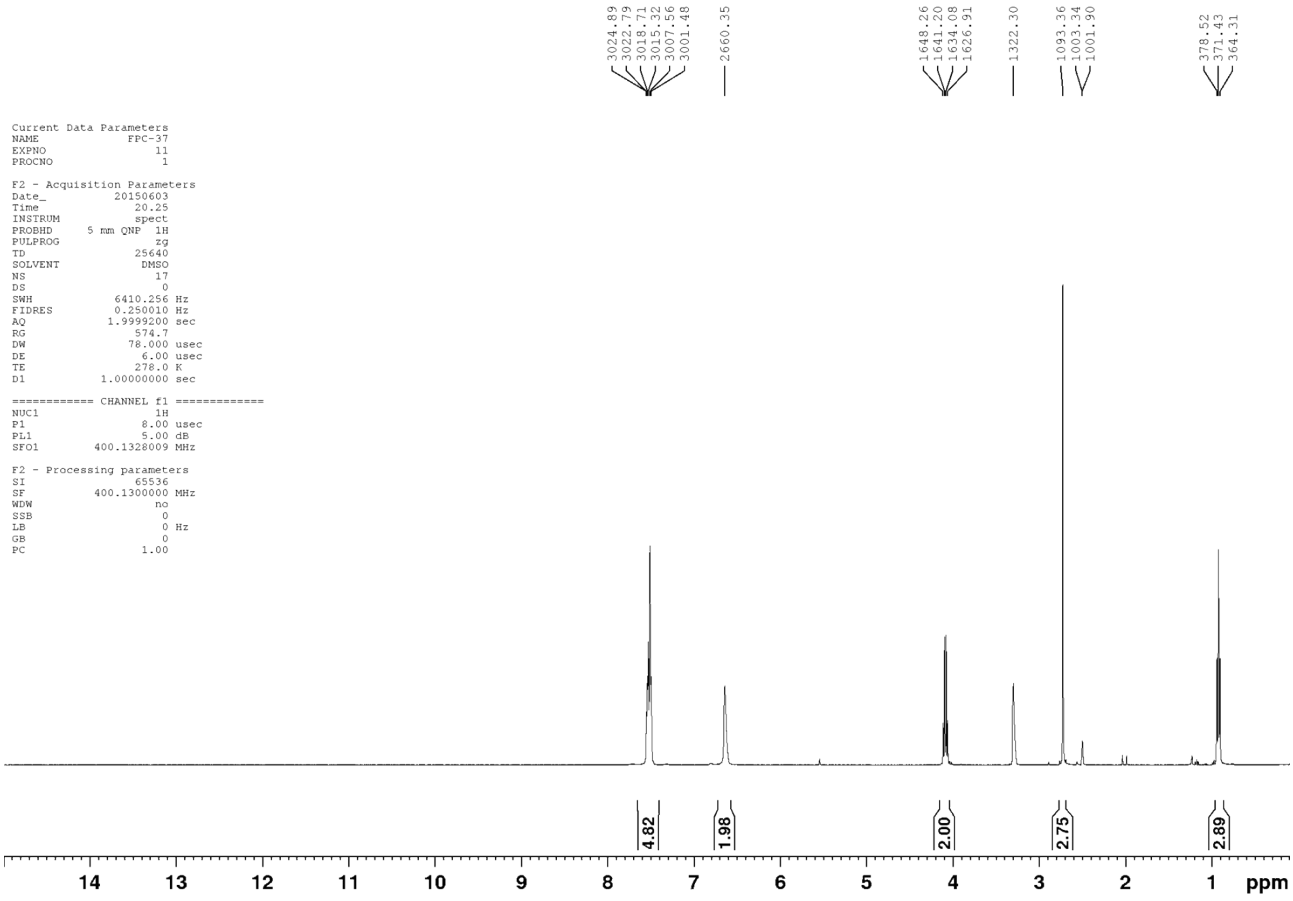


Figure S9. ^{1}H NMR spectrum of compound **9**.

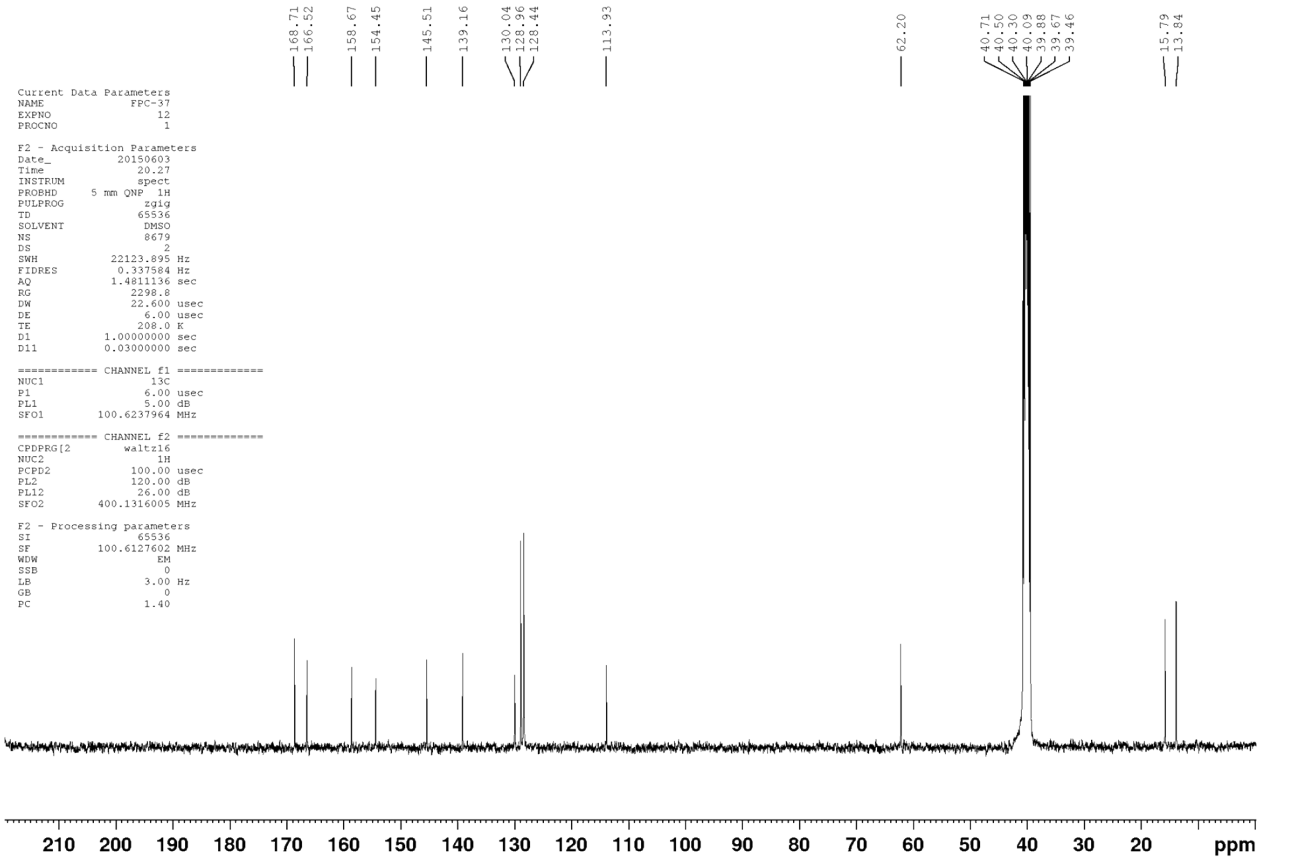


Figure S10. ^{13}C NMR spectrum of compound **9**.

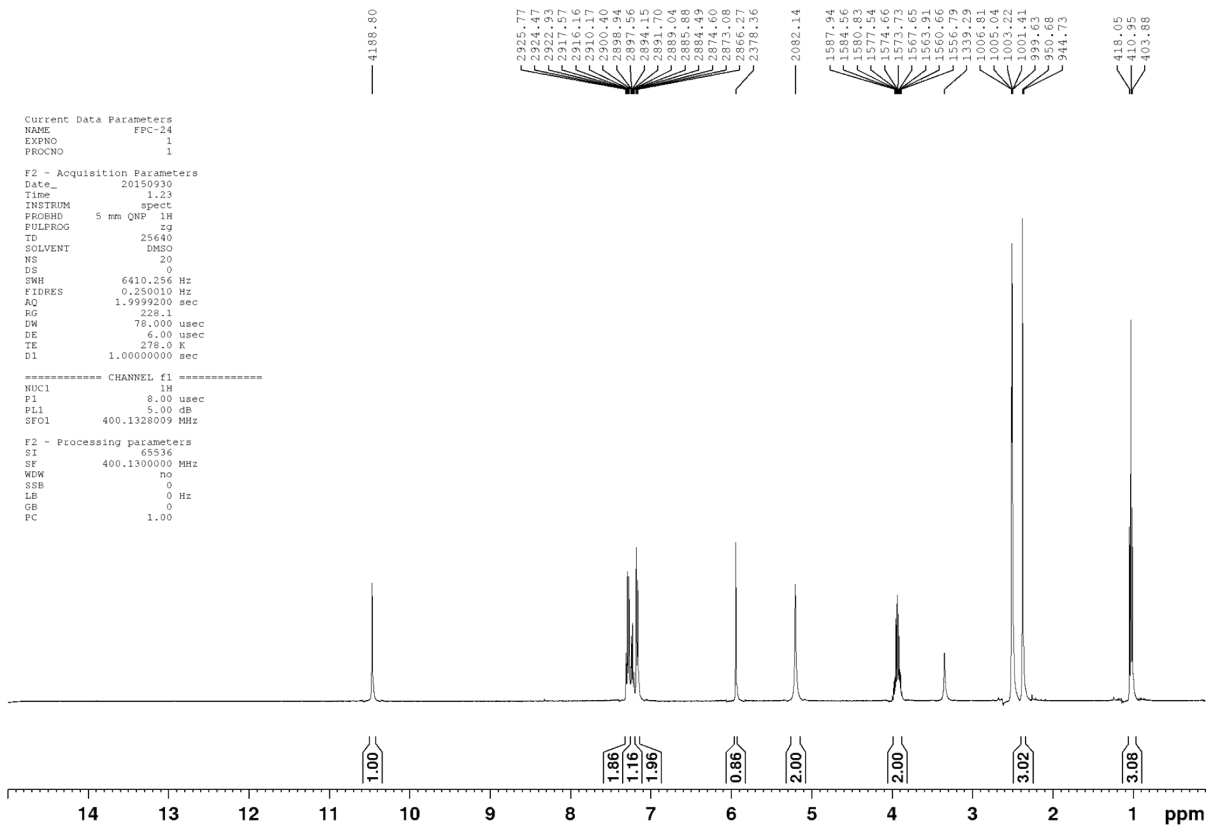


Figure S11. ^{1}H NMR spectrum of compound **10**.

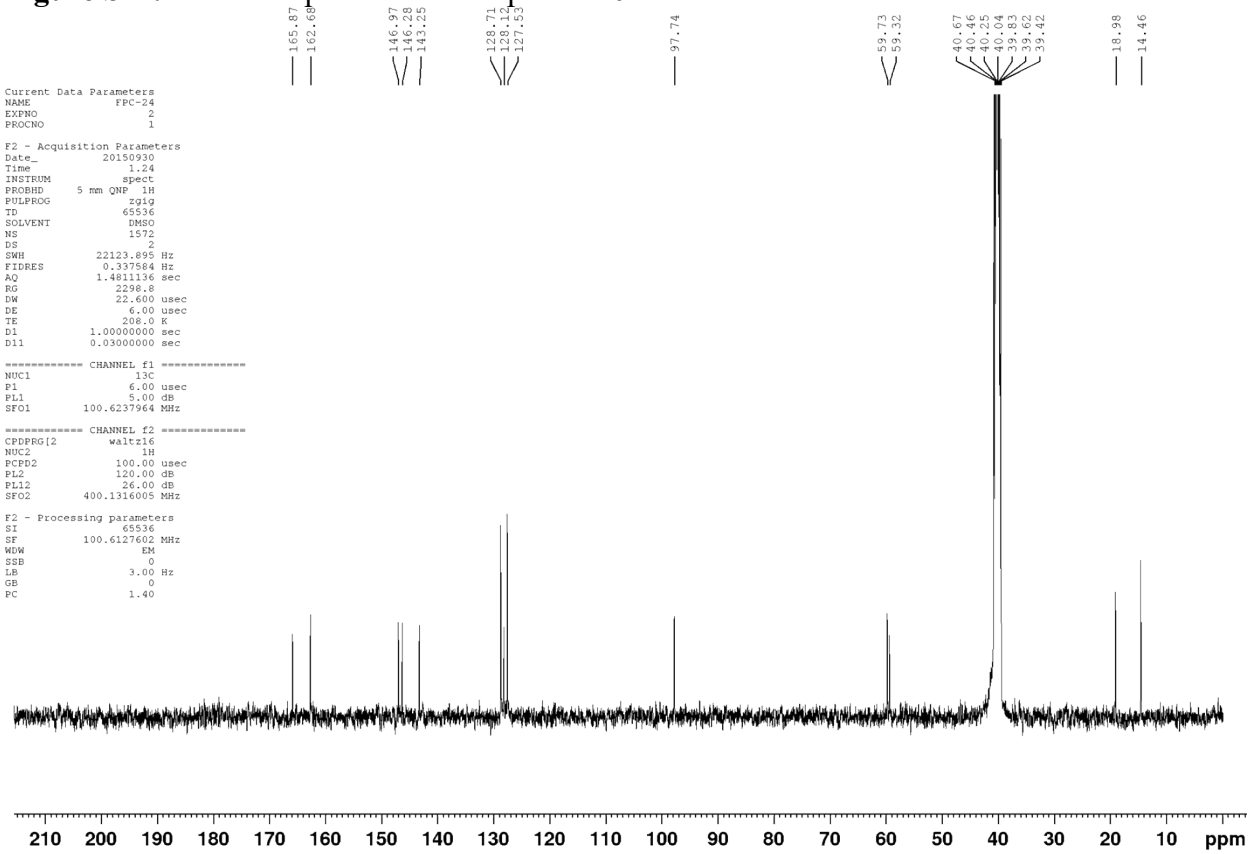


Figure S12. ^{13}C NMR spectrum of compound **10**.

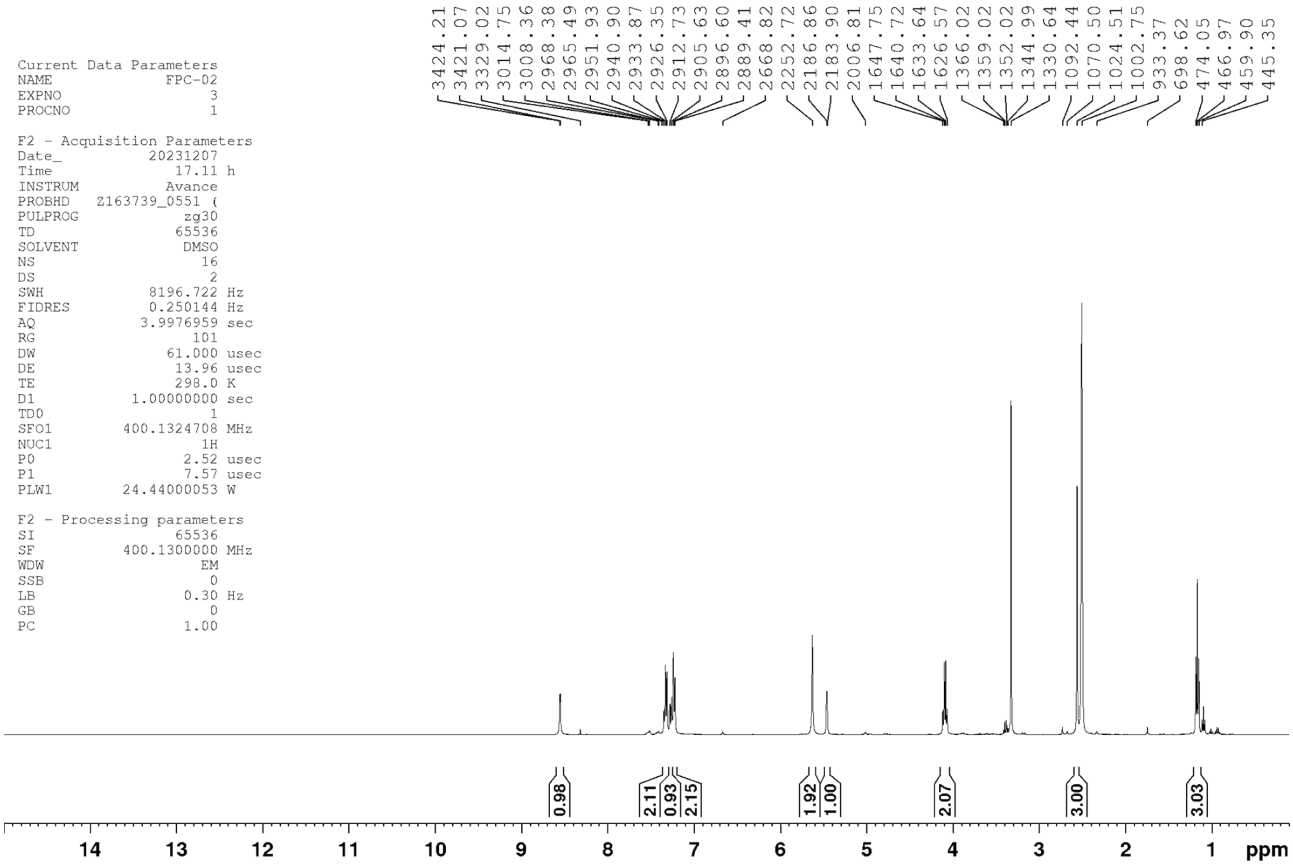


Figure S13. ^{1}H NMR spectrum of compound **11**.

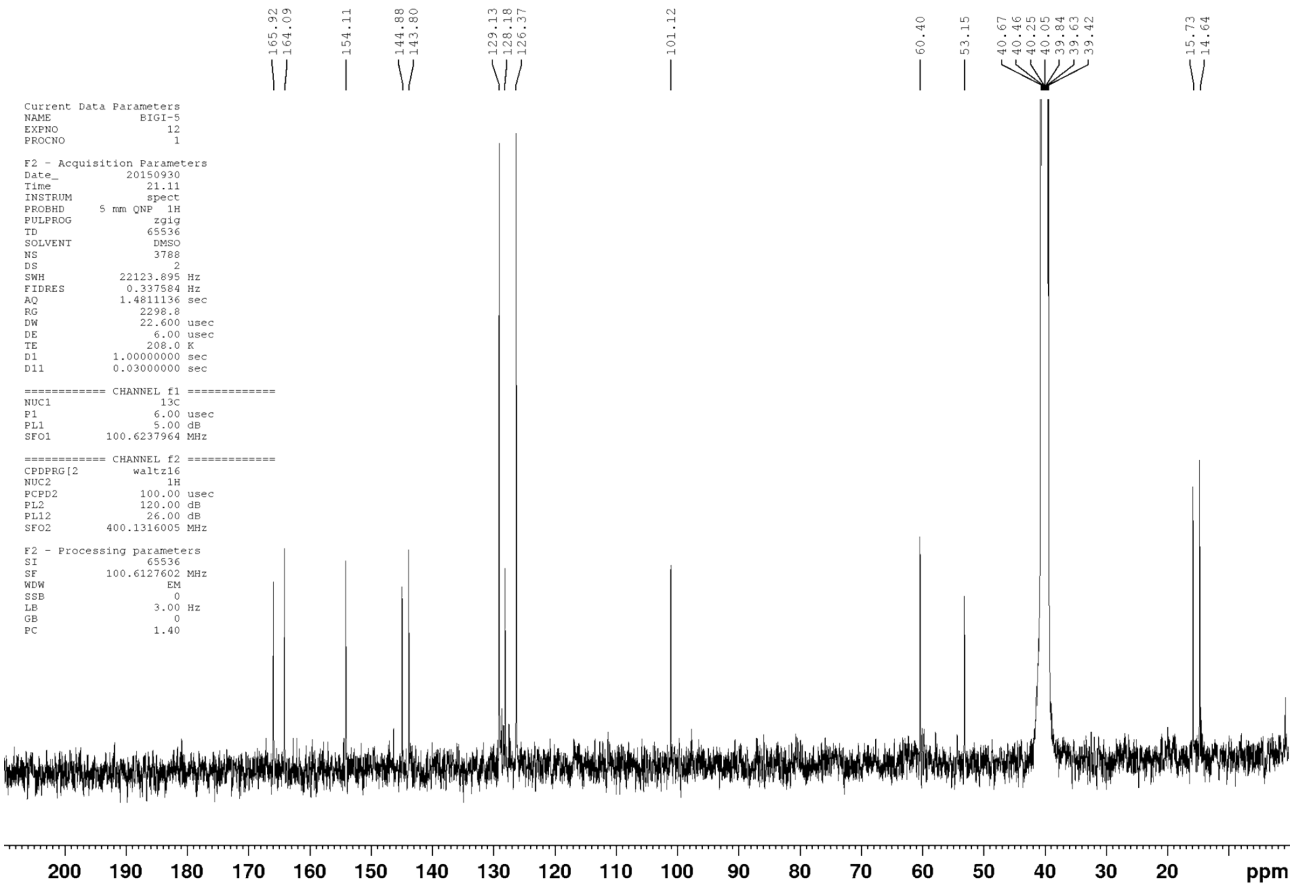


Figure S14. ^{13}C NMR spectrum of compound **11**.

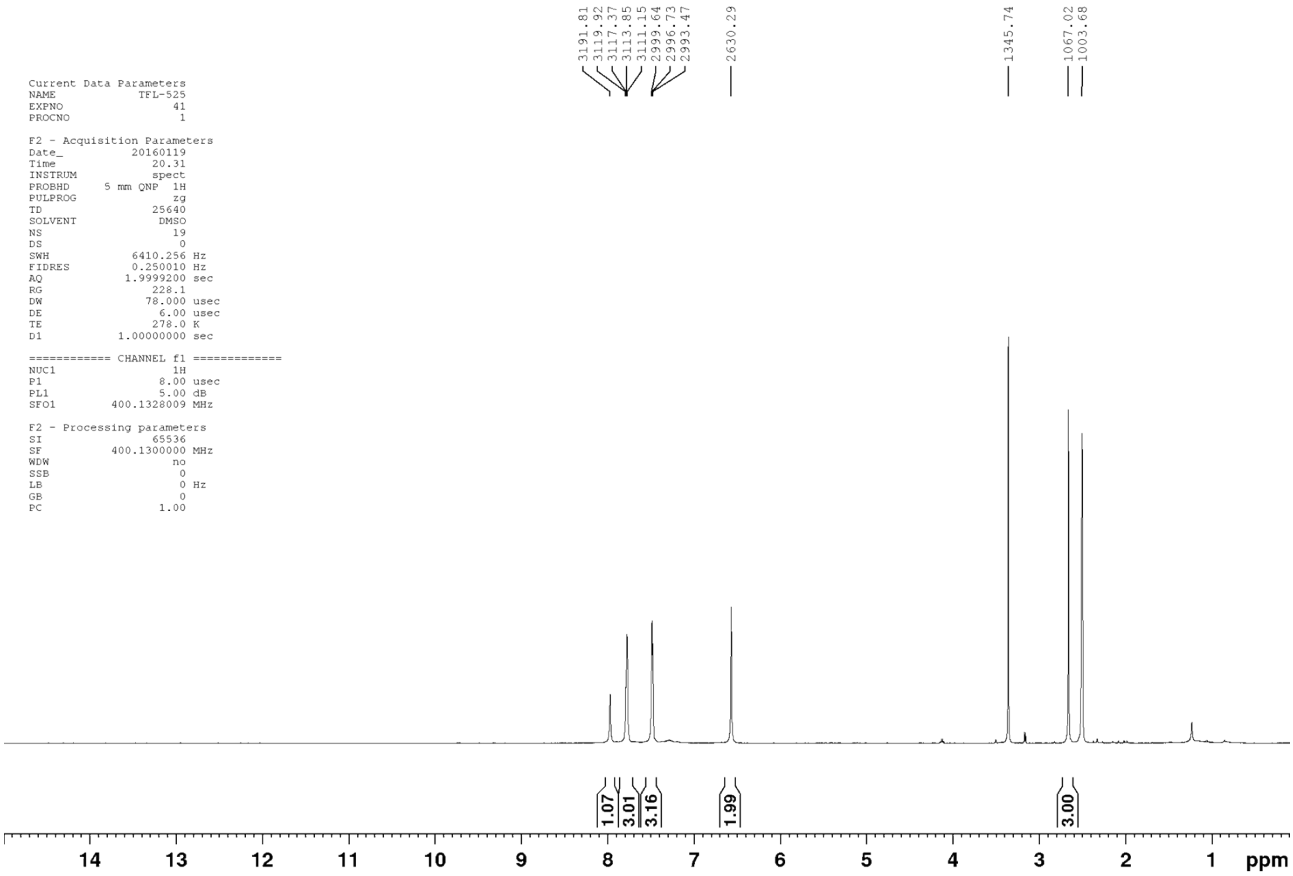


Figure S15. ^{1}H NMR spectrum of compound **16**.

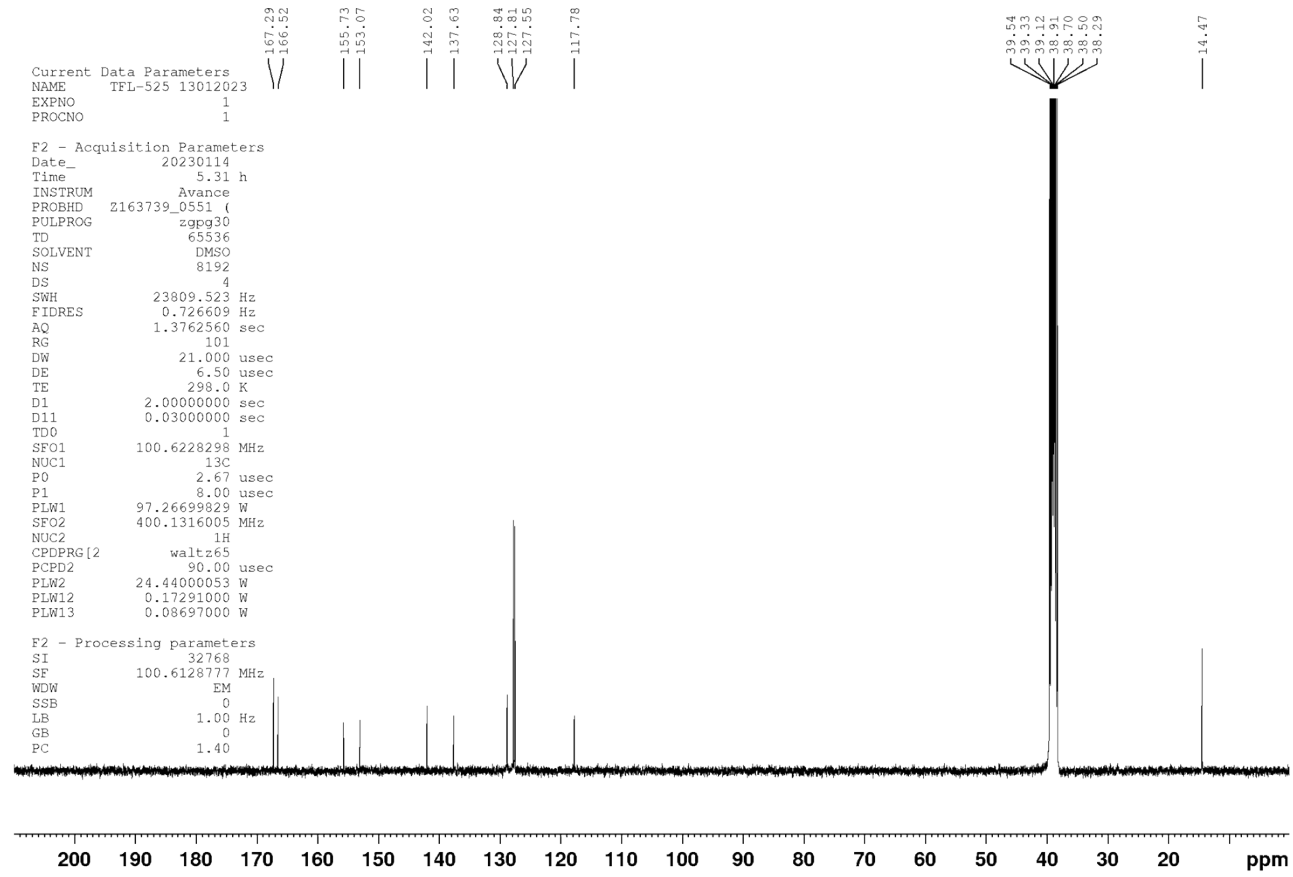


Figure S16. ^{13}C NMR spectrum of compound **16**.

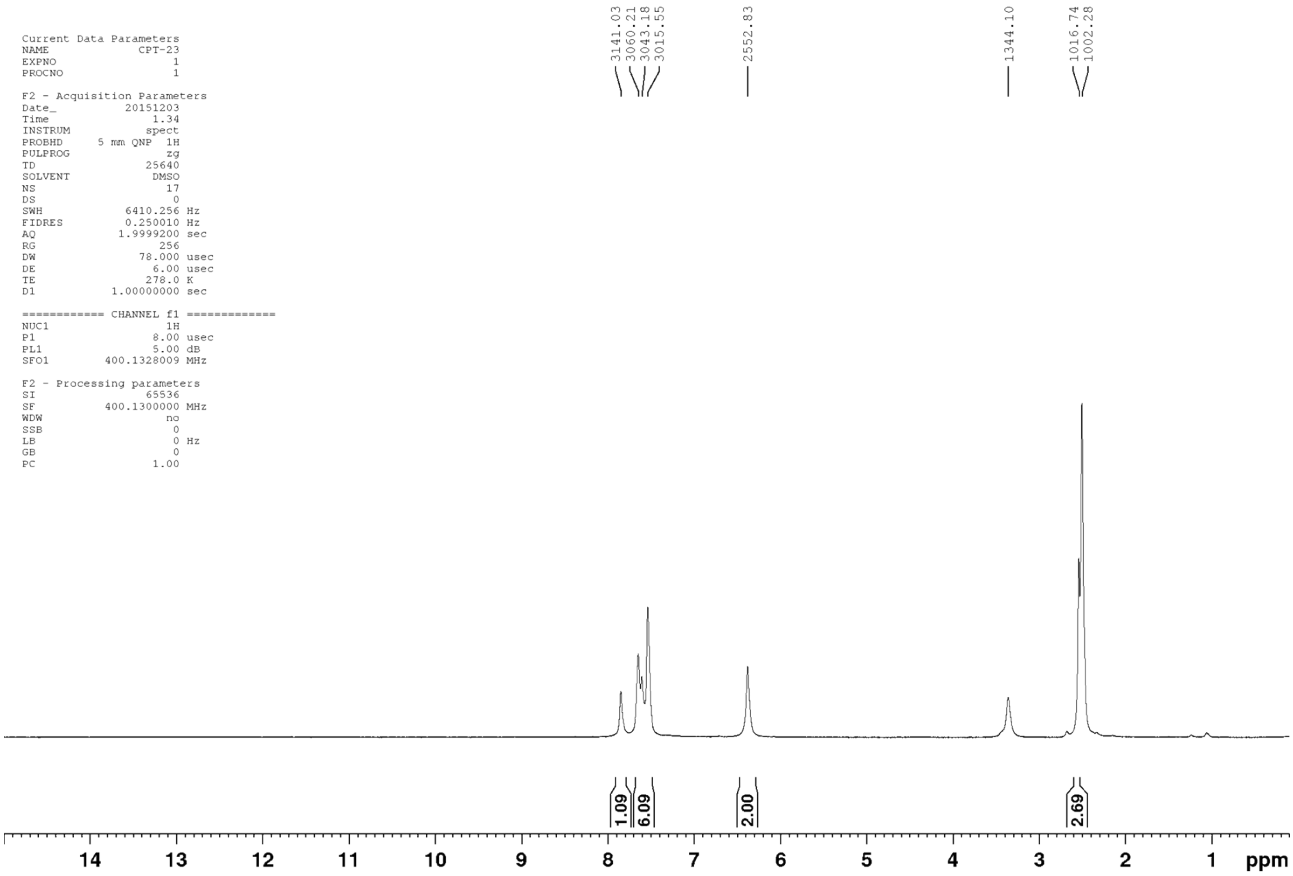


Figure S17. ^{1}H NMR spectrum of compound **17**.

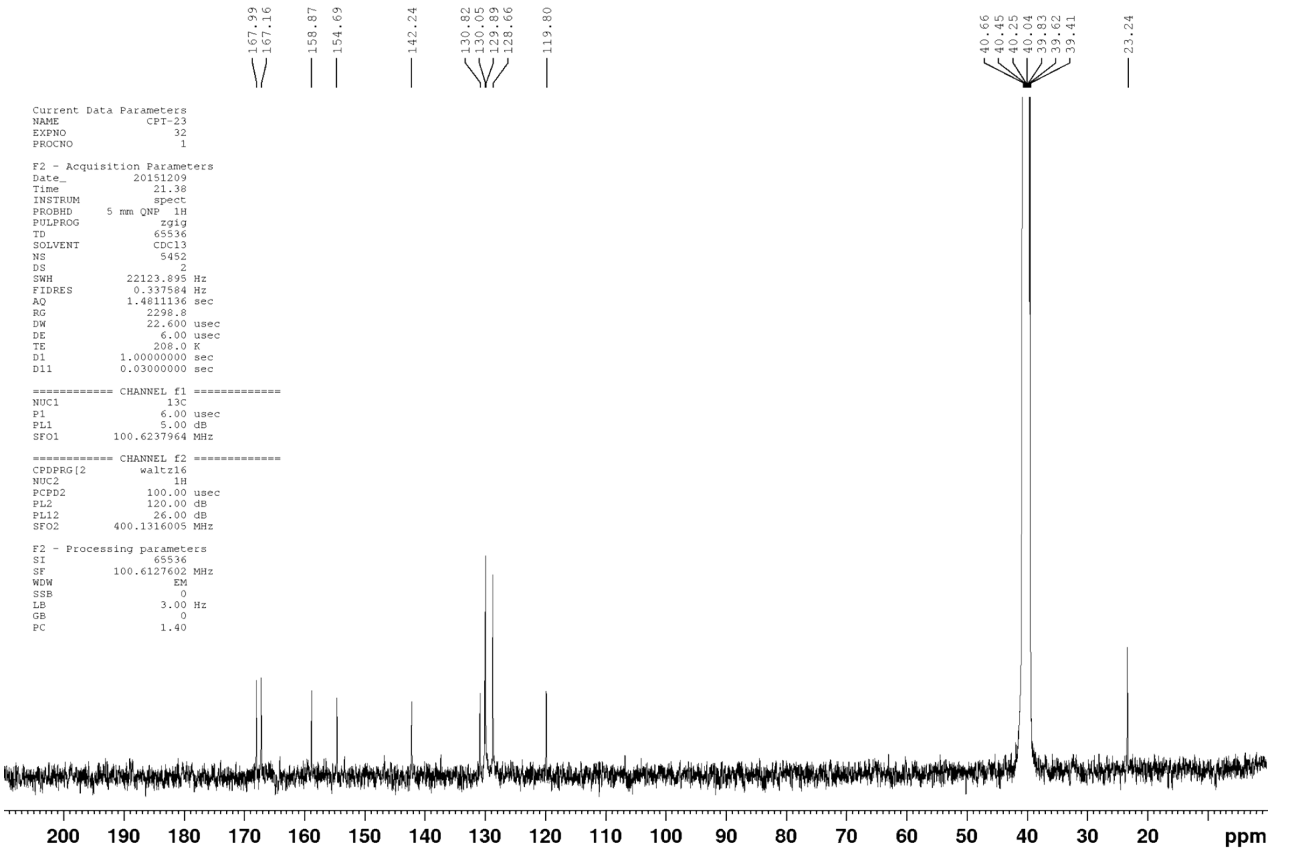


Figure S18. ^{13}C NMR spectrum of compound **17**.

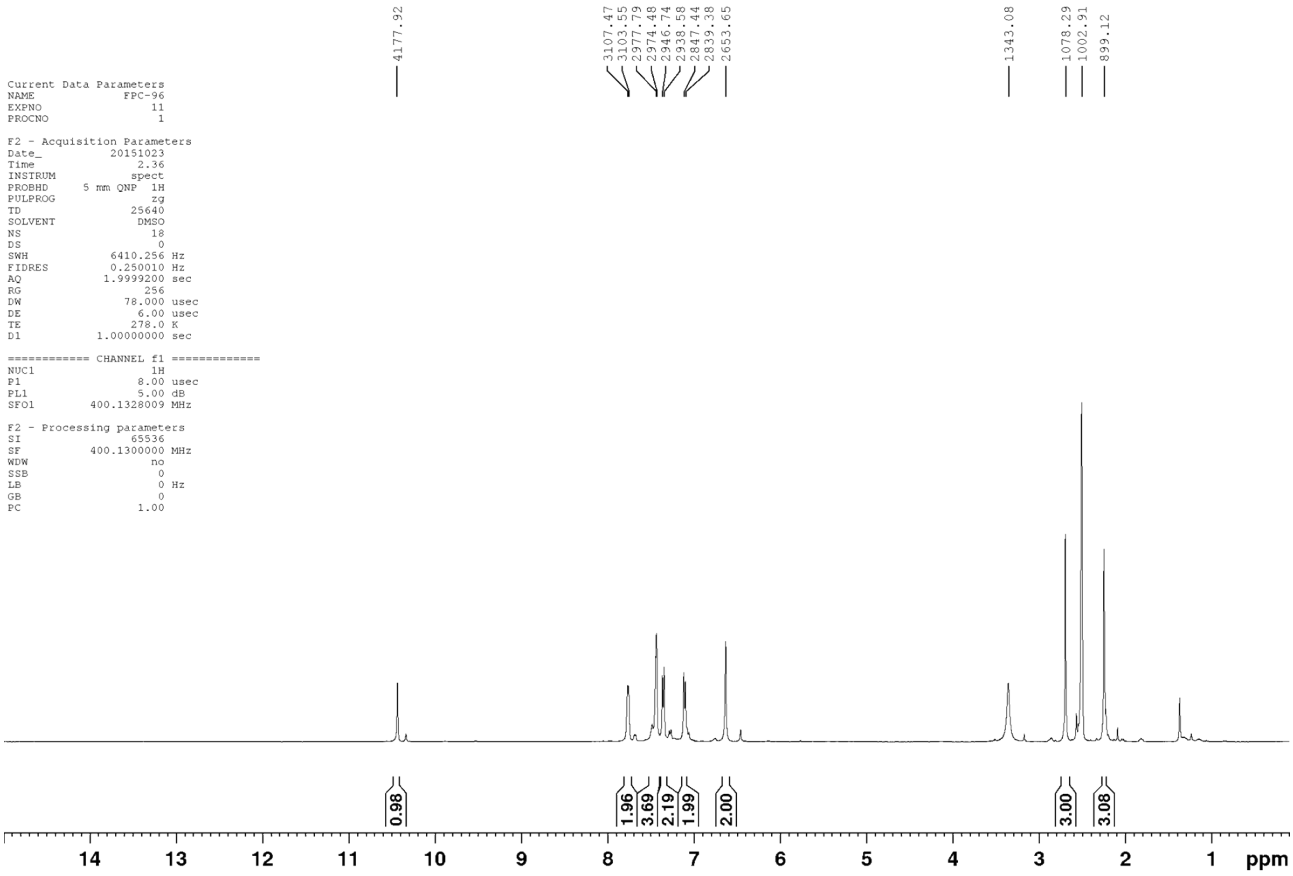


Figure S19. ^{1}H NMR spectrum of compound **18**.

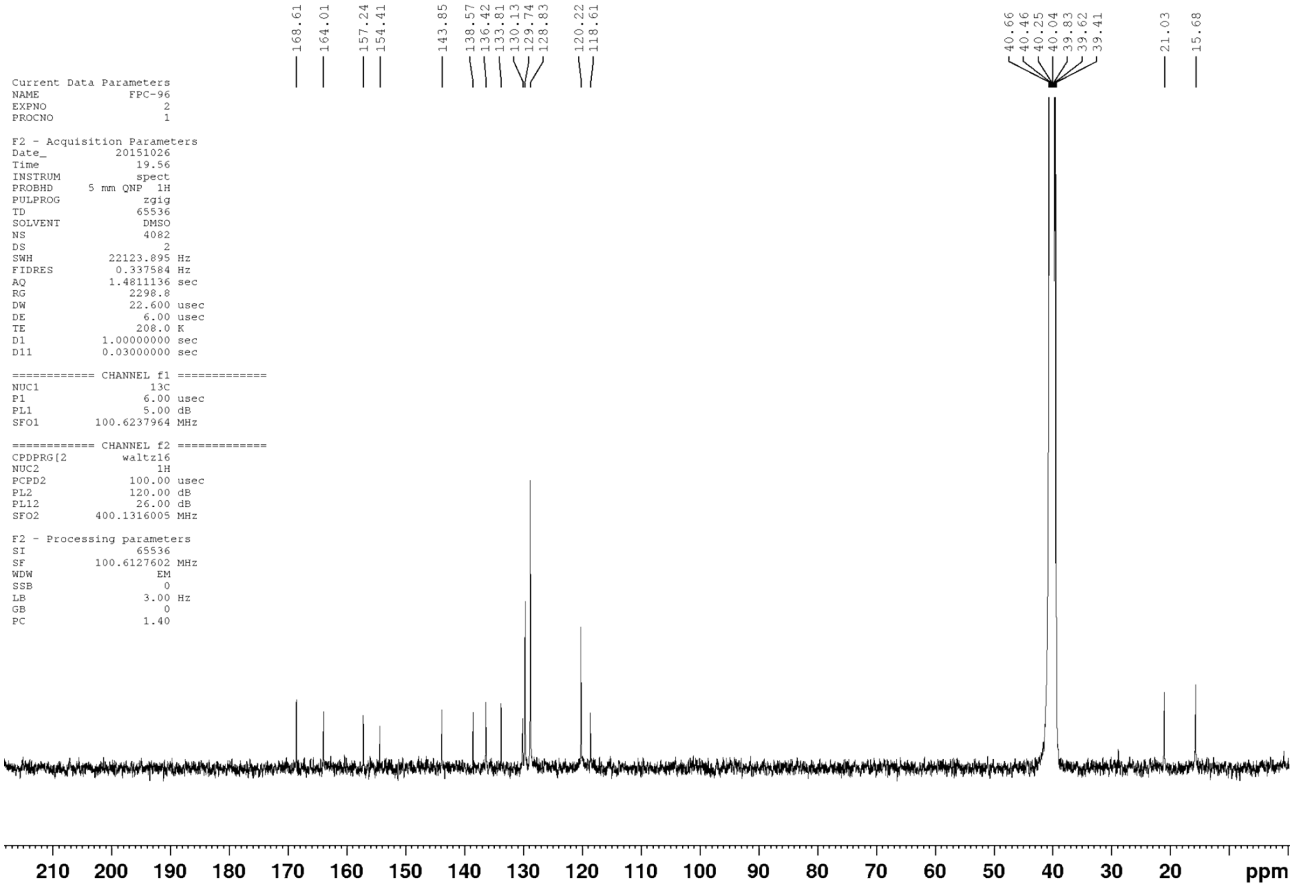


Figure S20. ^{13}C NMR spectrum of compound **18**.

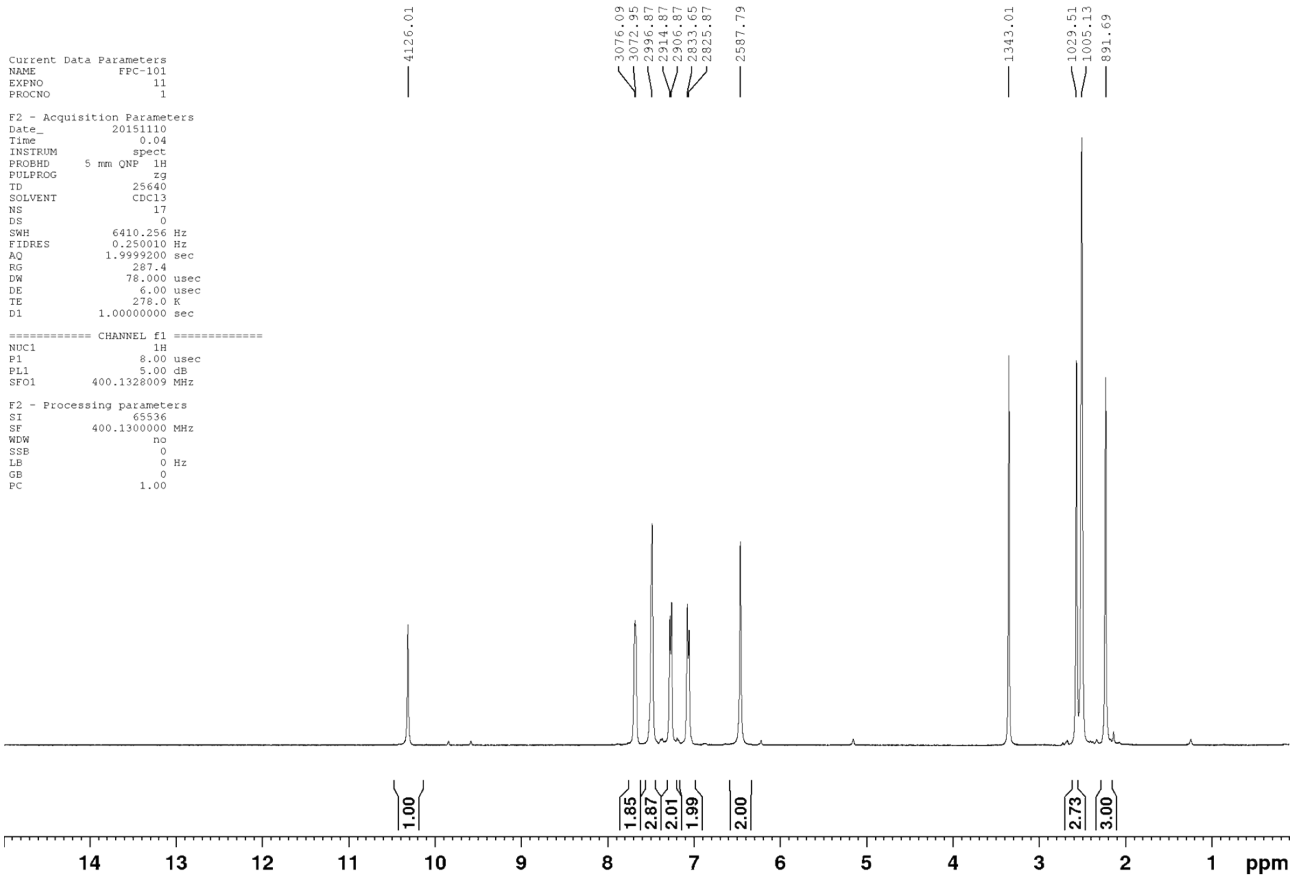


Figure S21. ^{1}H NMR spectrum of compound **19**.

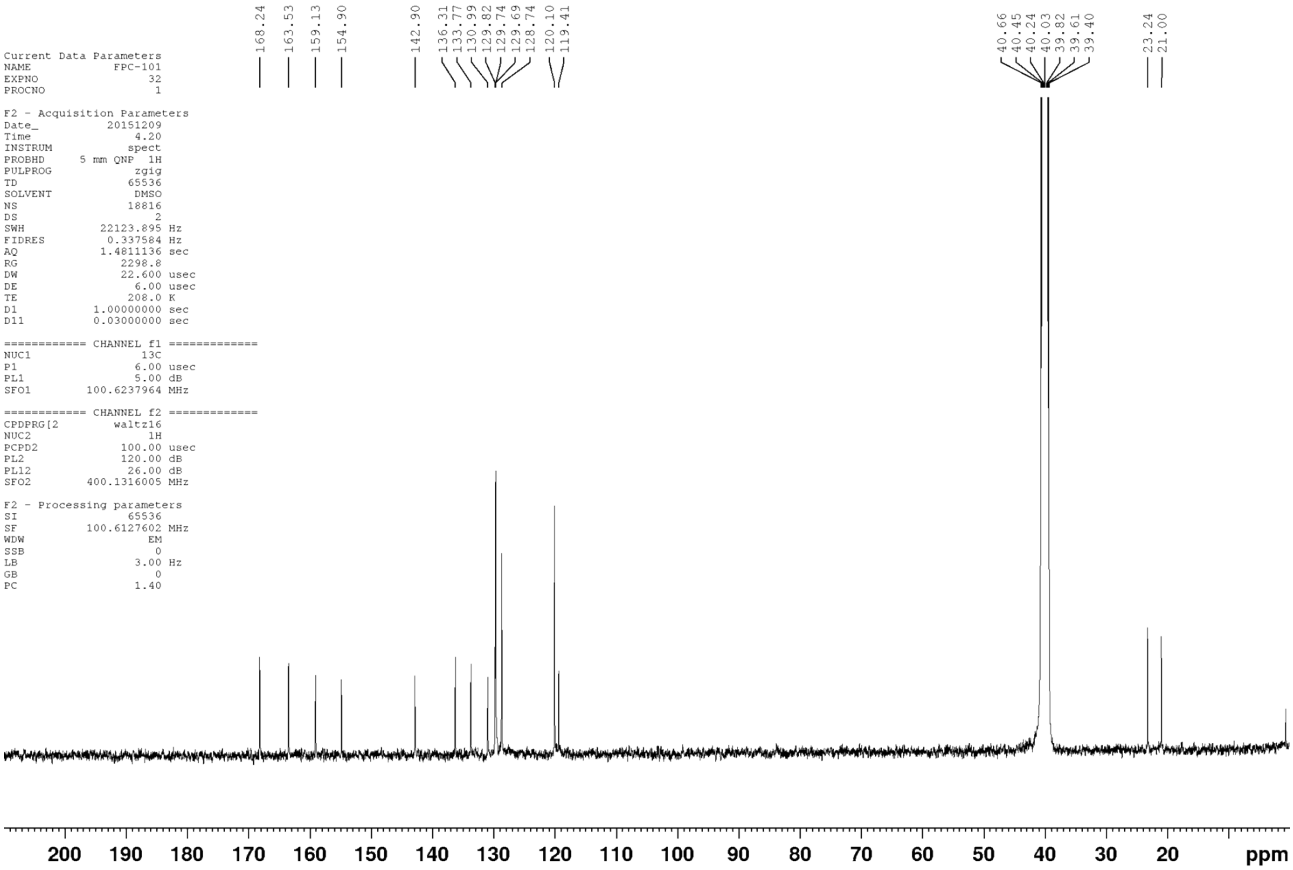


Figure S22. ^{13}C NMR spectrum of compound **19**.

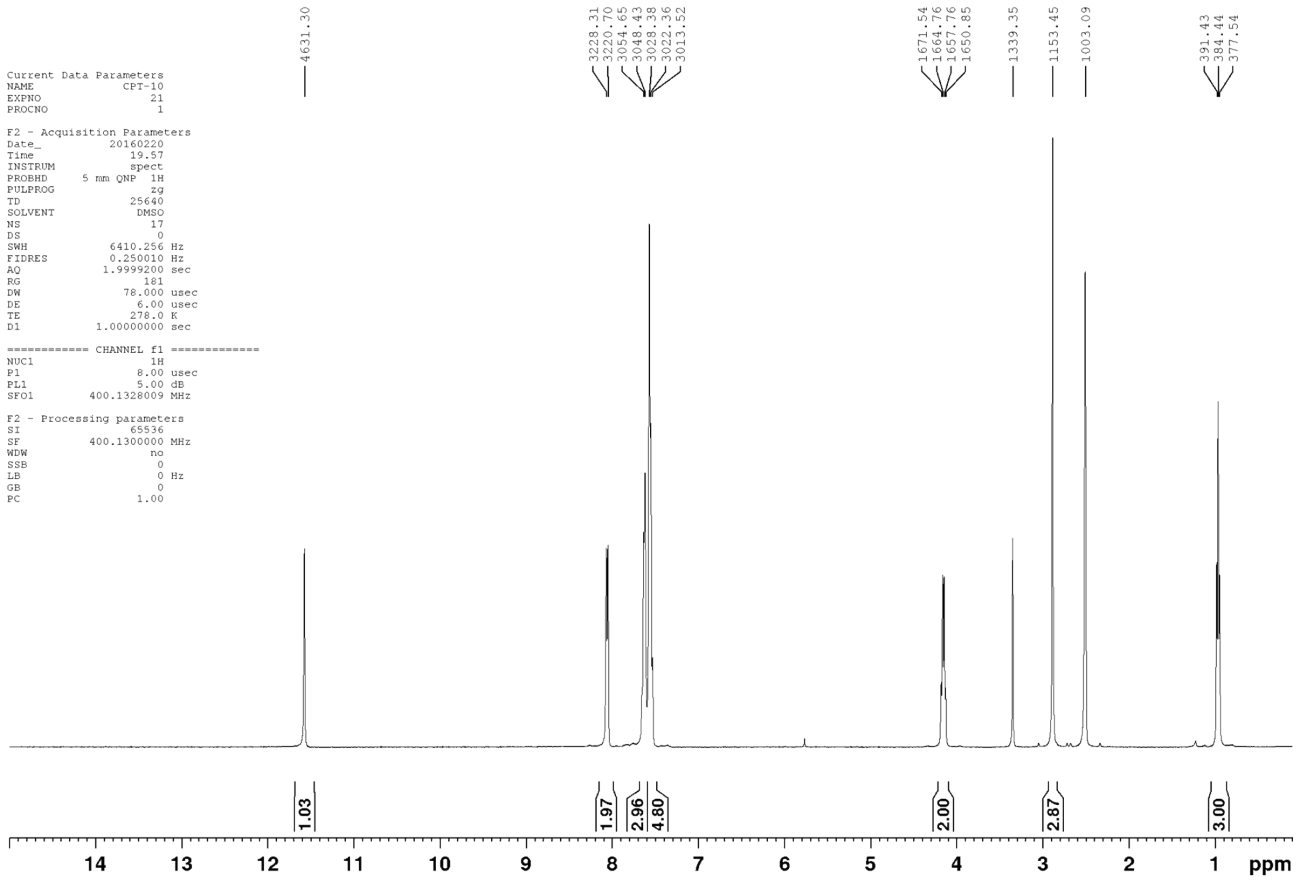


Figure S23. ^{1}H NMR spectrum of compound **23**.

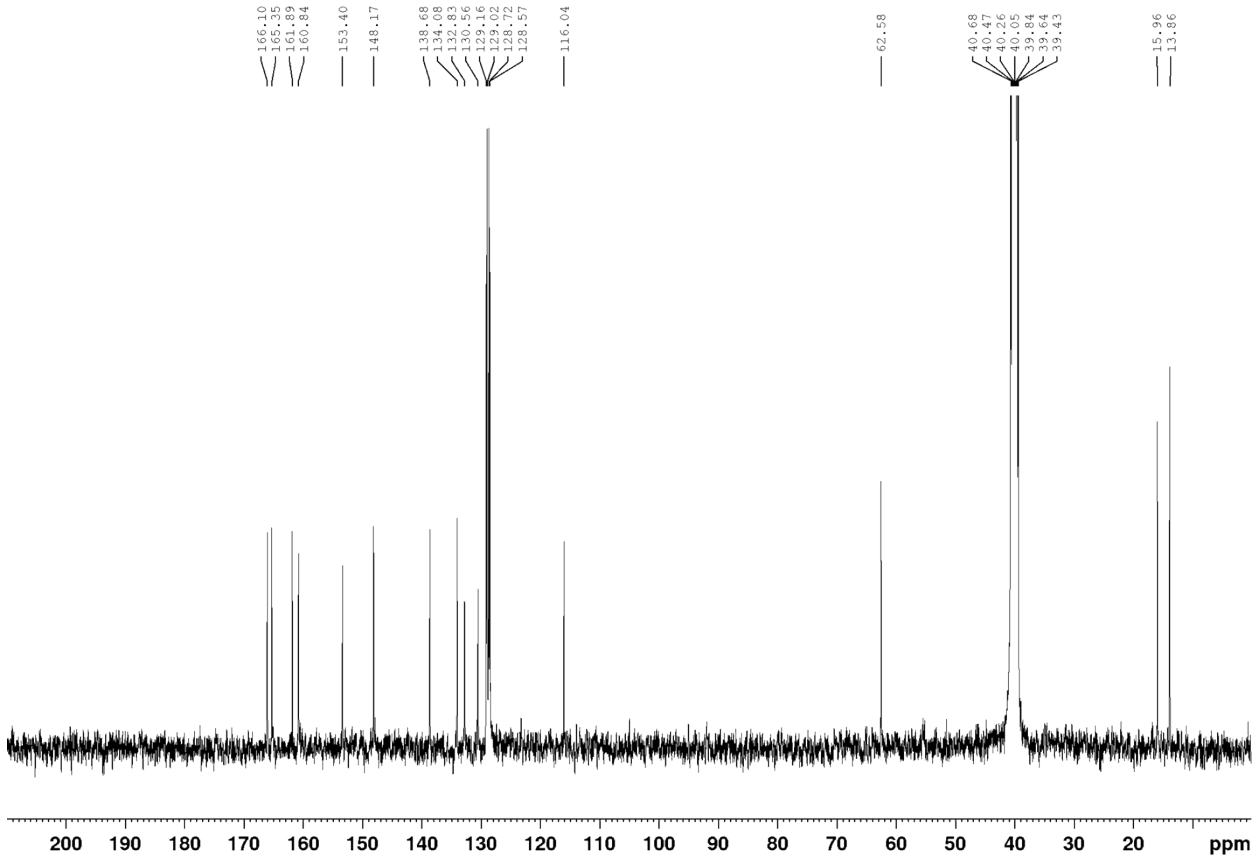


Figure S24. ^{13}C NMR spectrum of compound **23**.

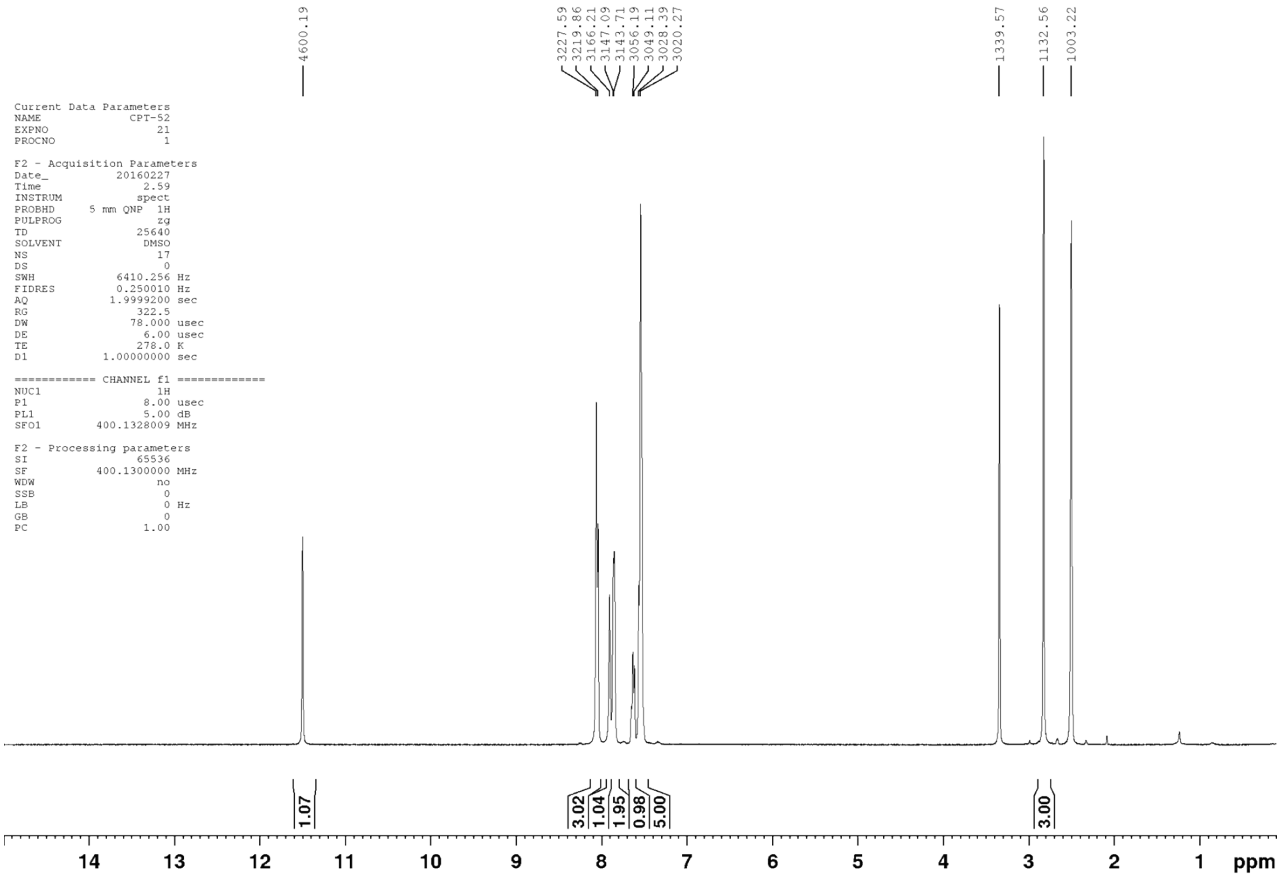


Figure S25. ^{1}H NMR spectrum of compound **24**.

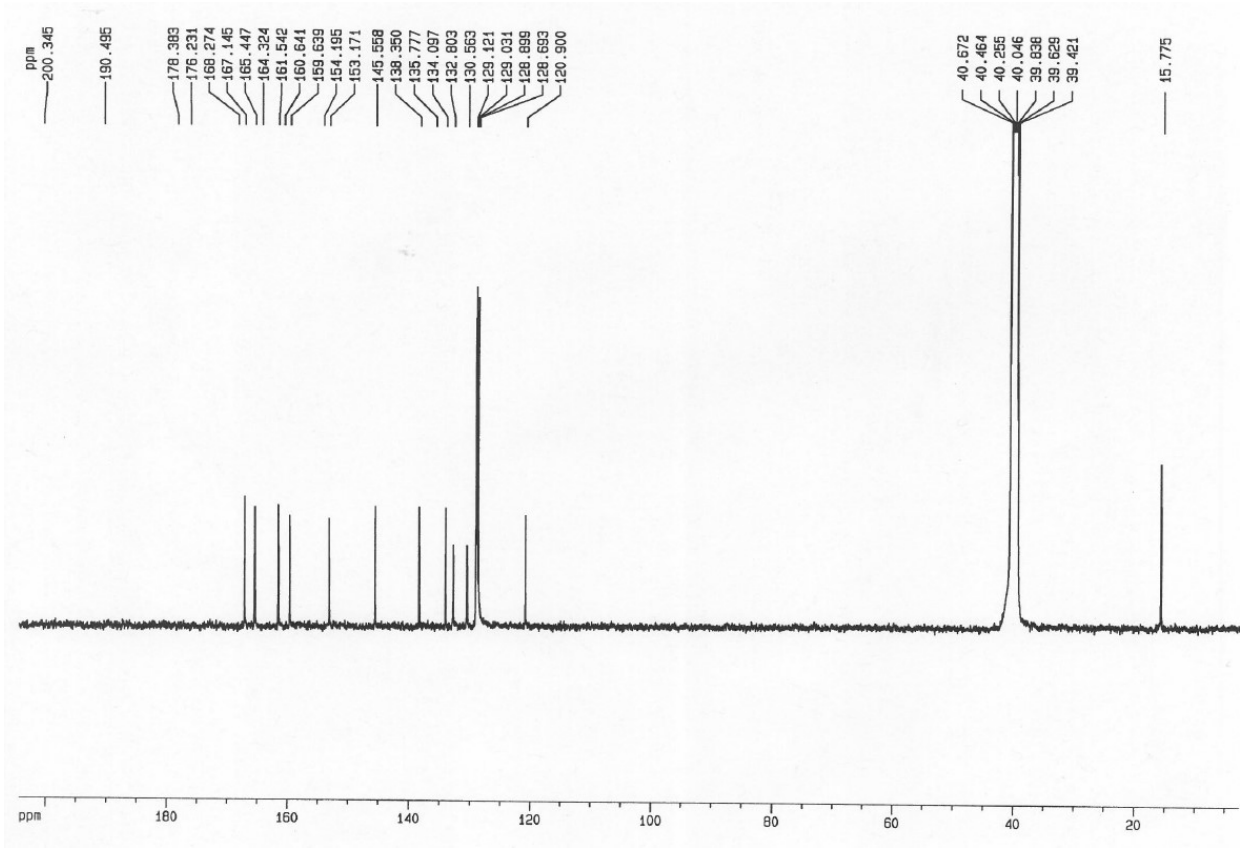


Figure S26. ^{13}C NMR spectrum of compound **24**.

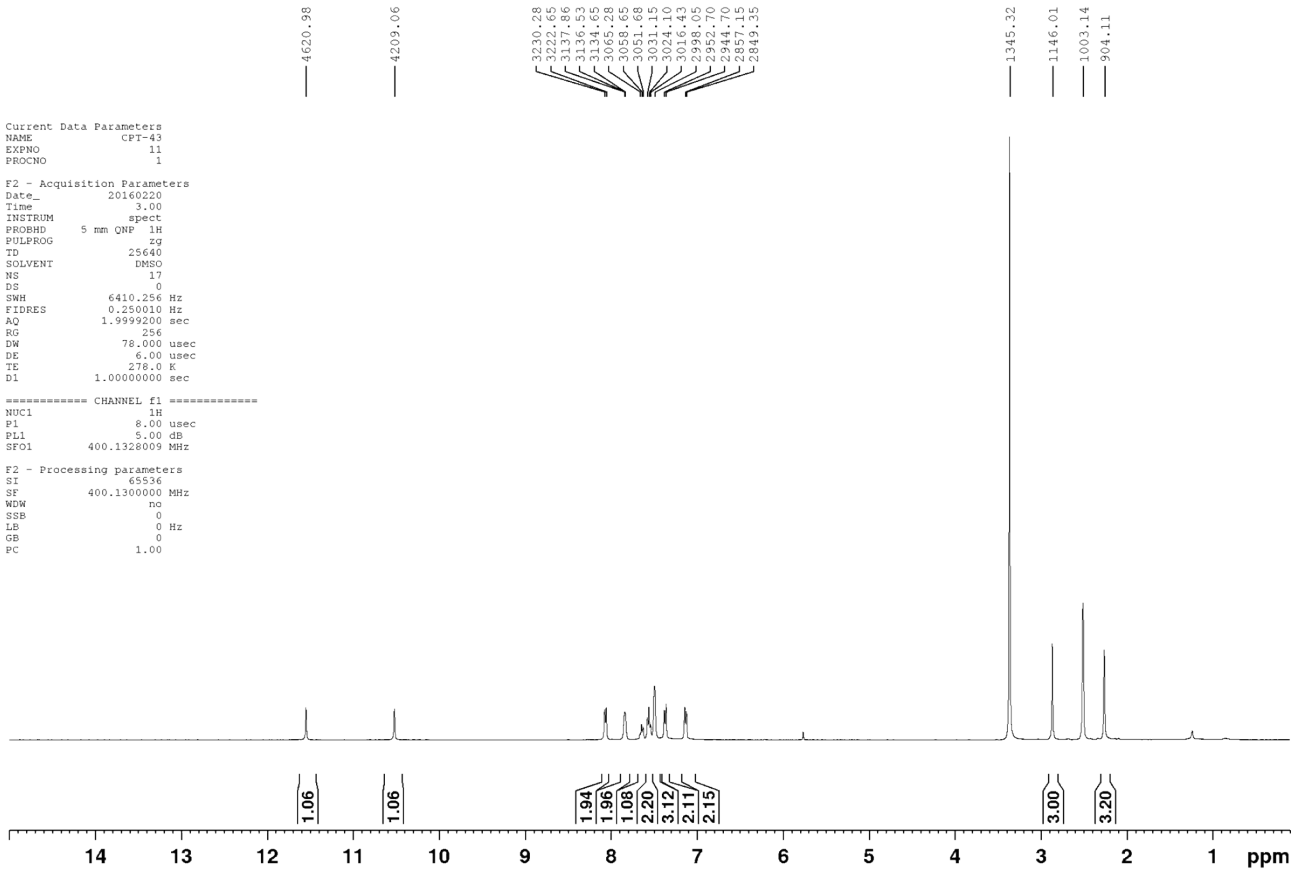


Figure S27. ^{1}H NMR spectrum of compound **25**.

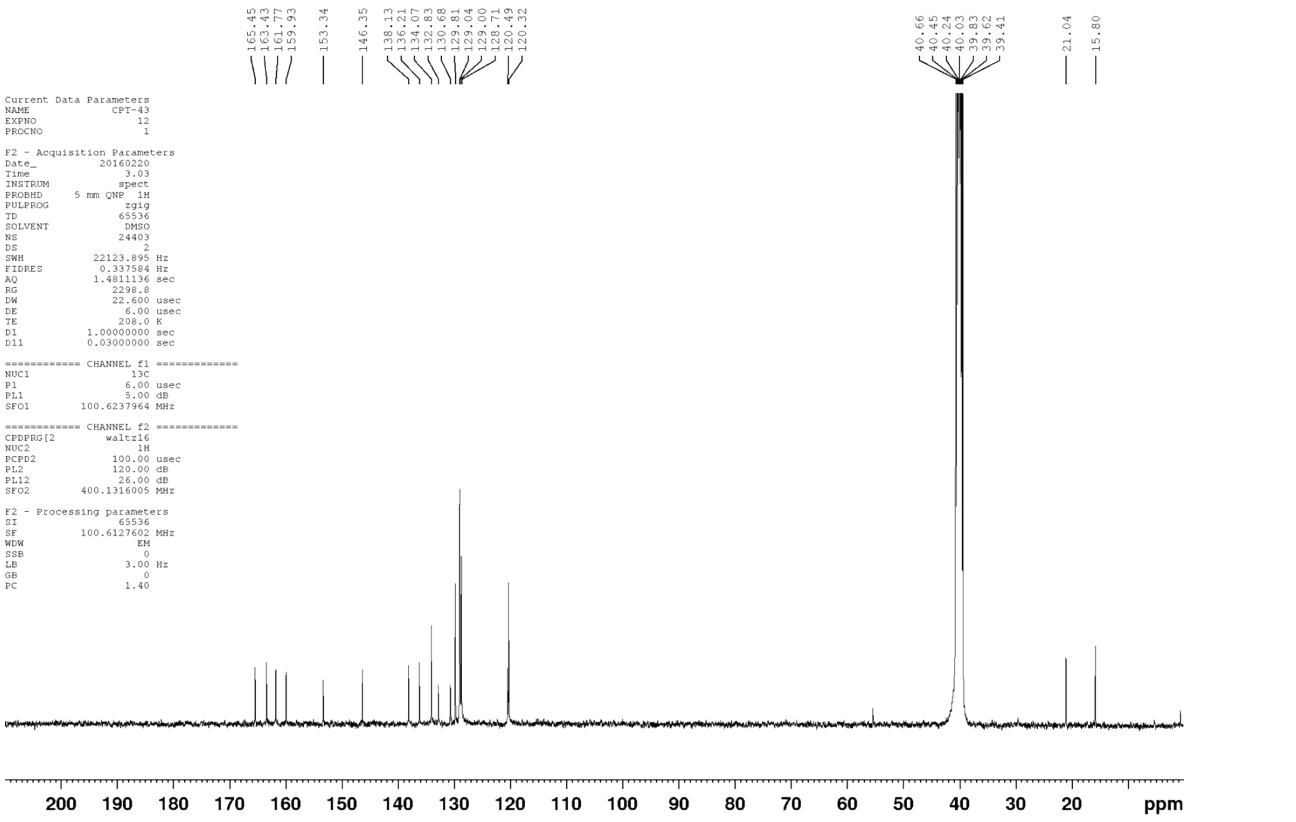


Figure S28. ^{13}C NMR spectrum of compound **25**.

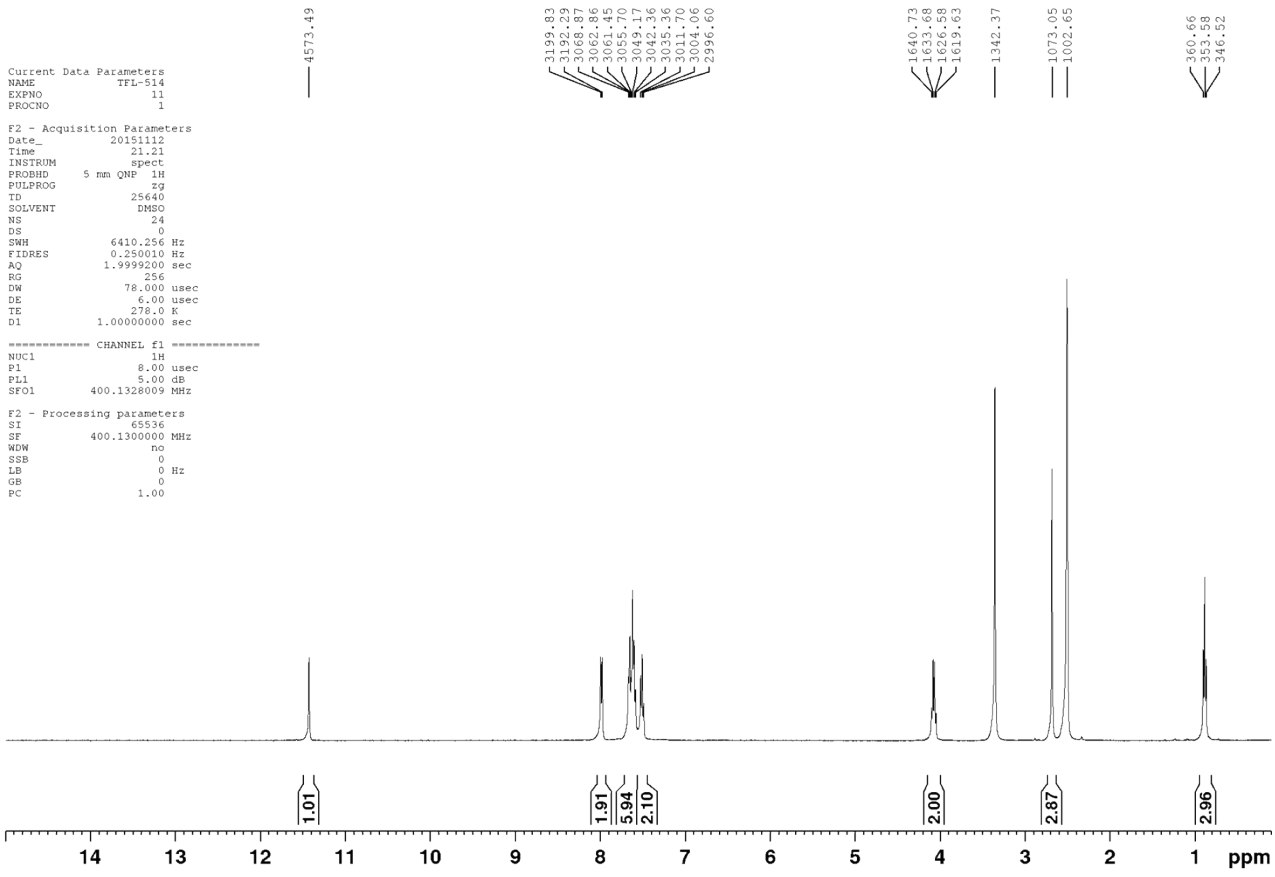


Figure S29. ^{1}H NMR spectrum of compound **26**.

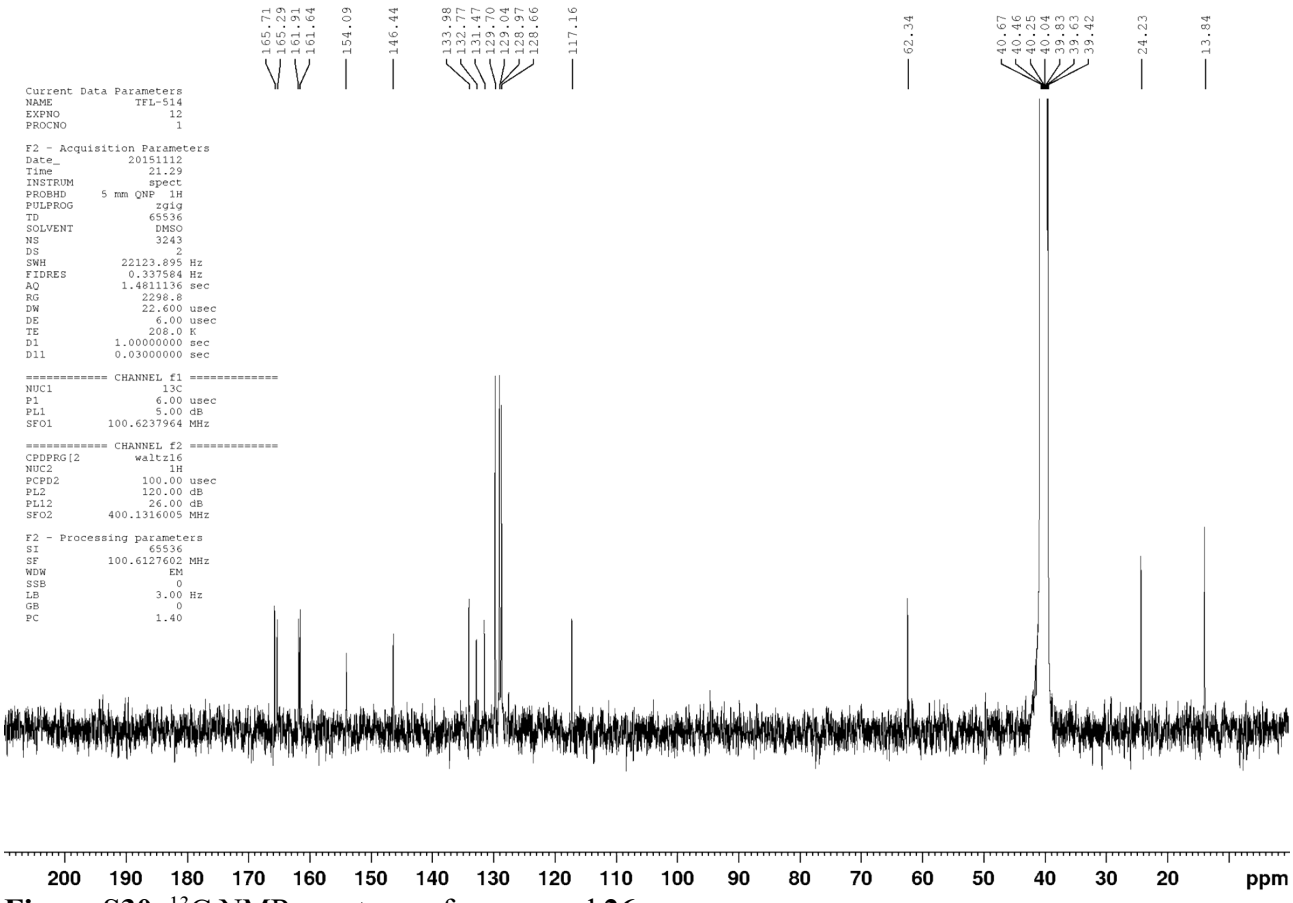


Figure S30. ^{13}C NMR spectrum of compound **26**.

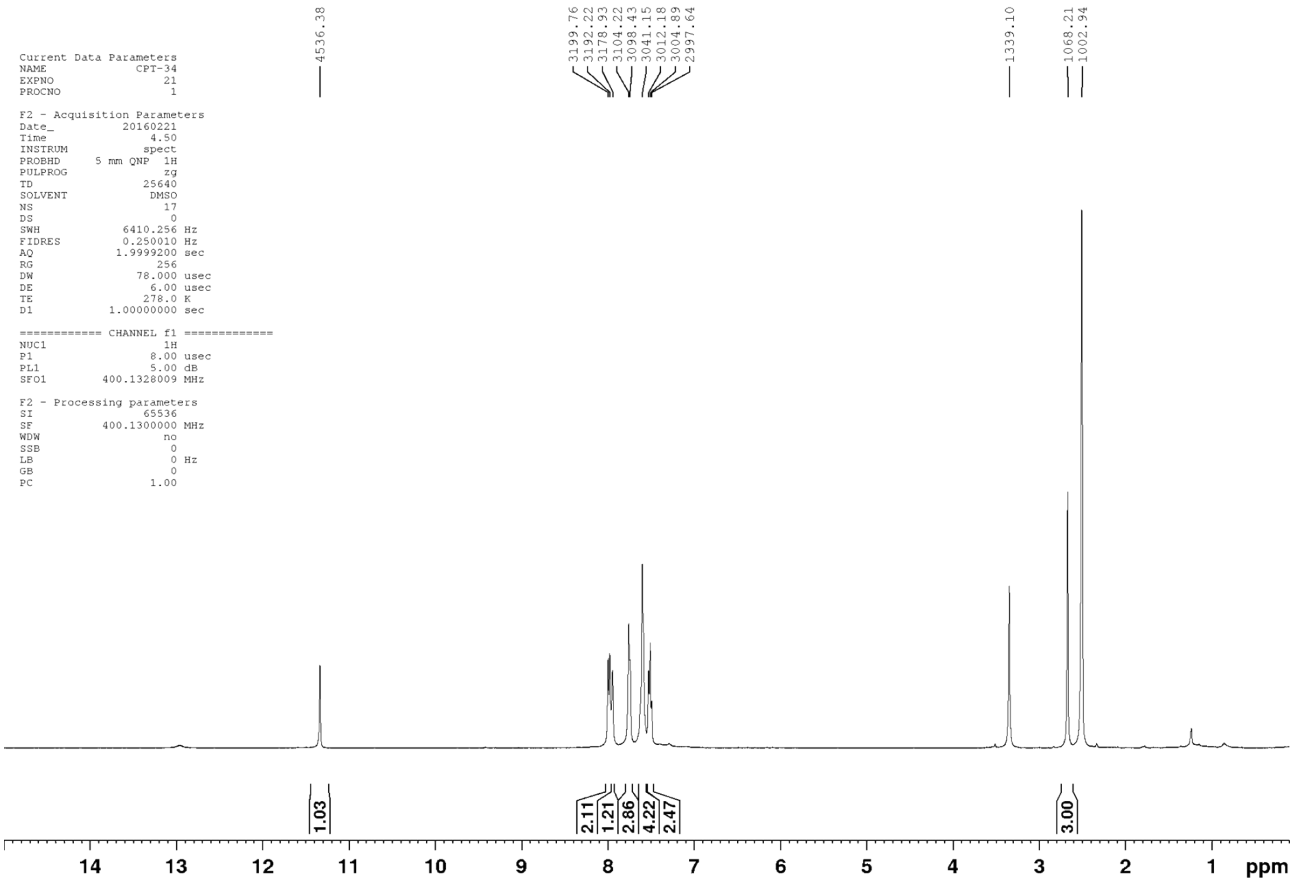


Figure S31. ^{1}H NMR spectrum of compound **27**.

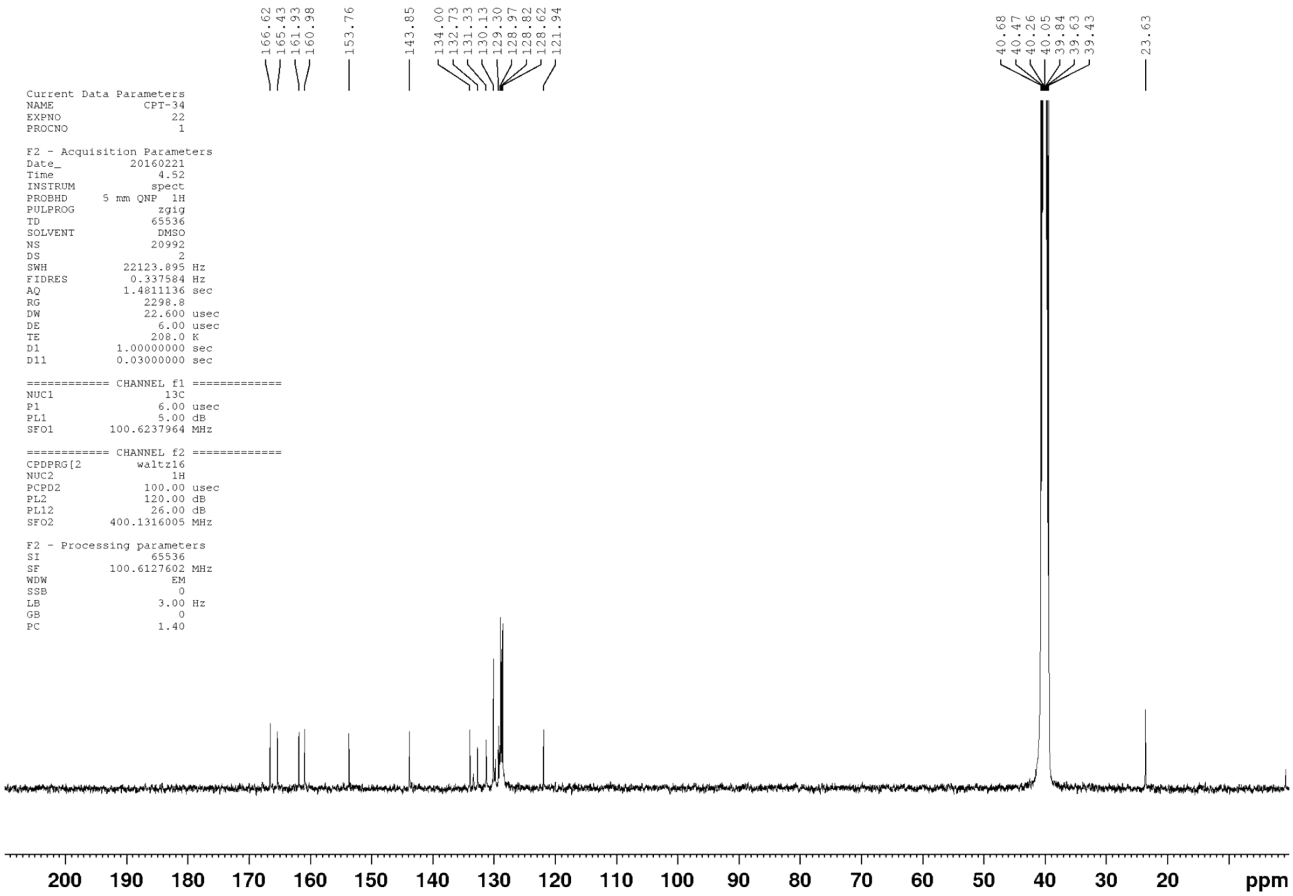


Figure S32. ^{13}C NMR spectrum of compound **27**.

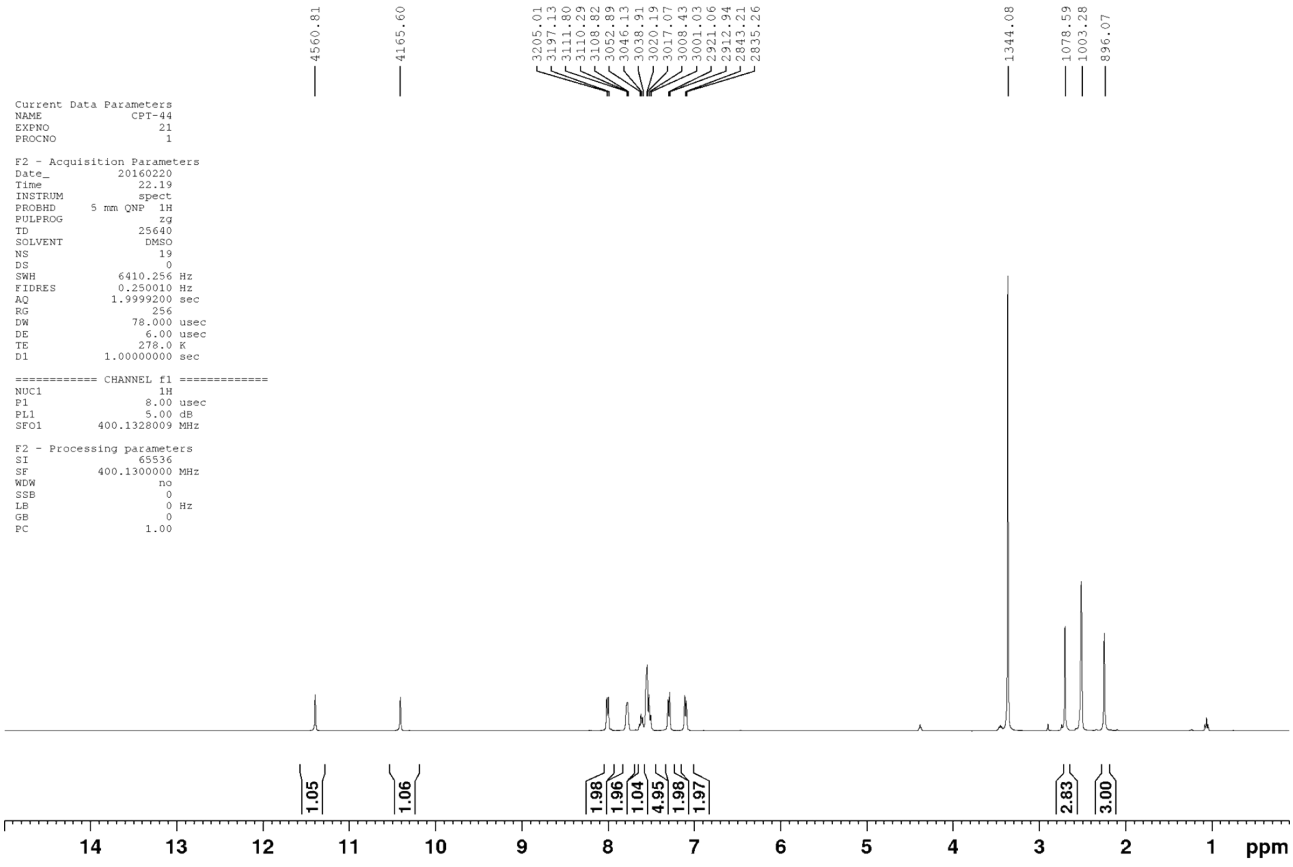


Figure S33. ^{1}H NMR spectrum of compound **28**.

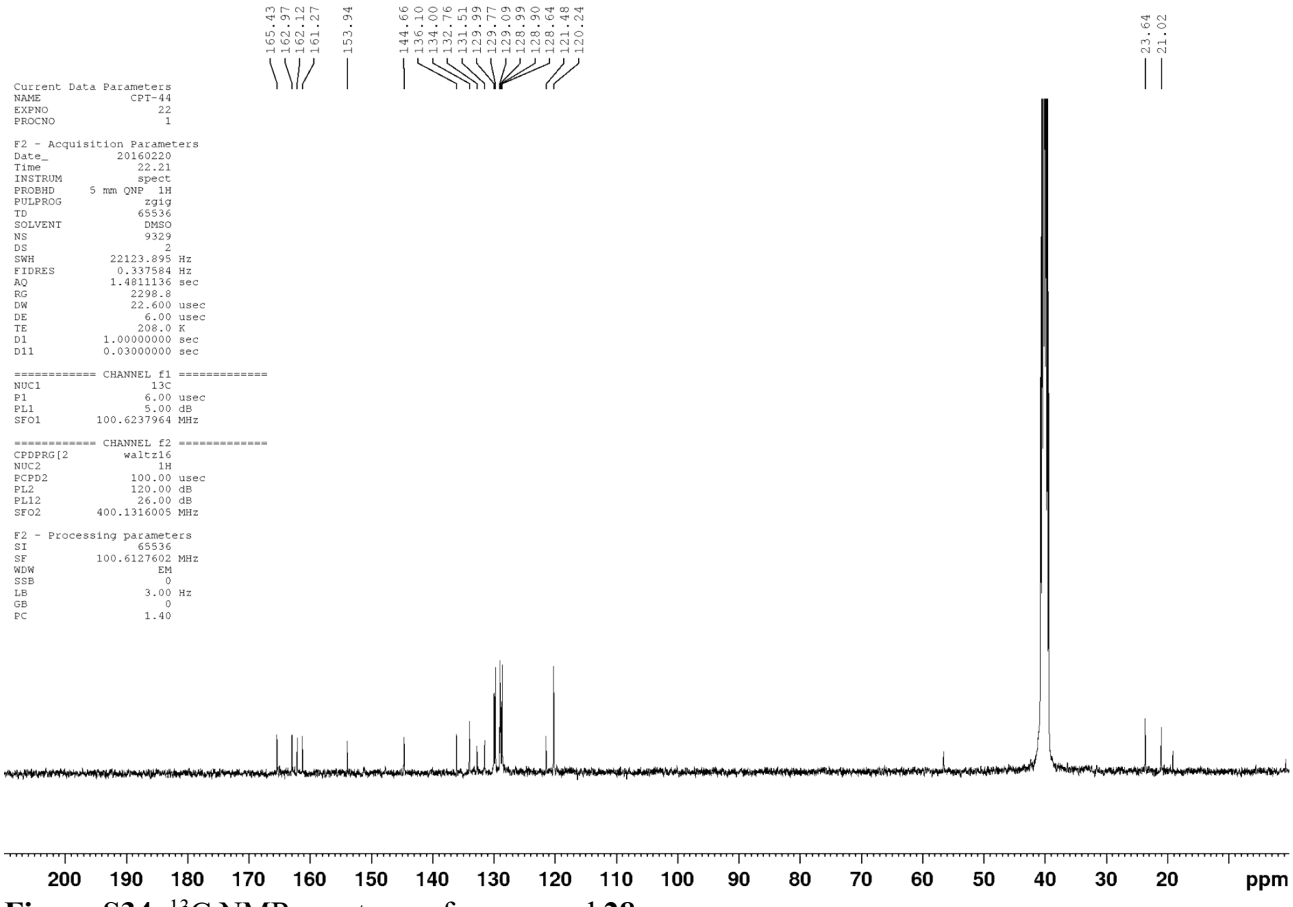


Figure S34. ^{13}C NMR spectrum of compound **28**.

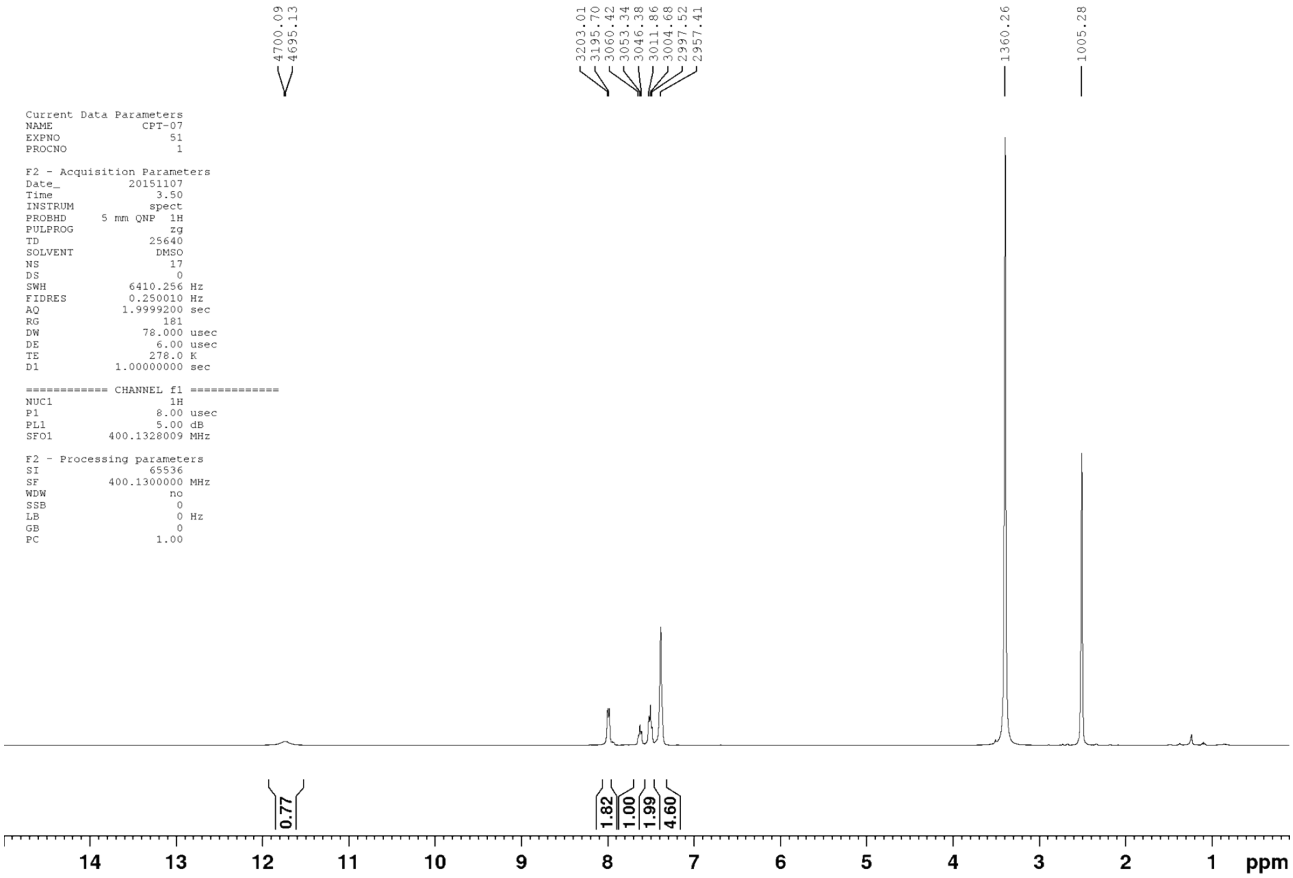


Figure S35. ^{1}H NMR spectrum of compound **29**.

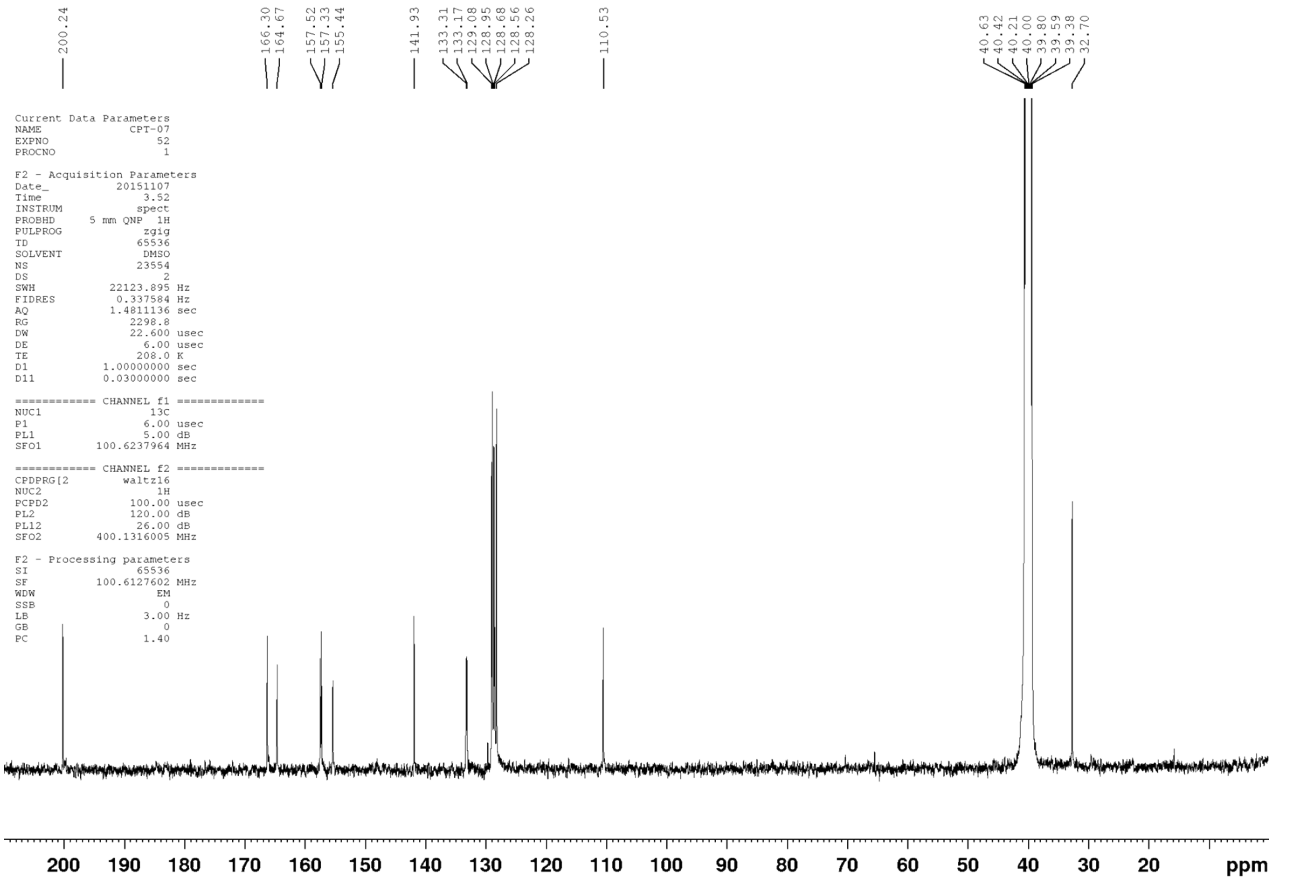


Figure S36. ^{13}C NMR spectrum of compound **29**.

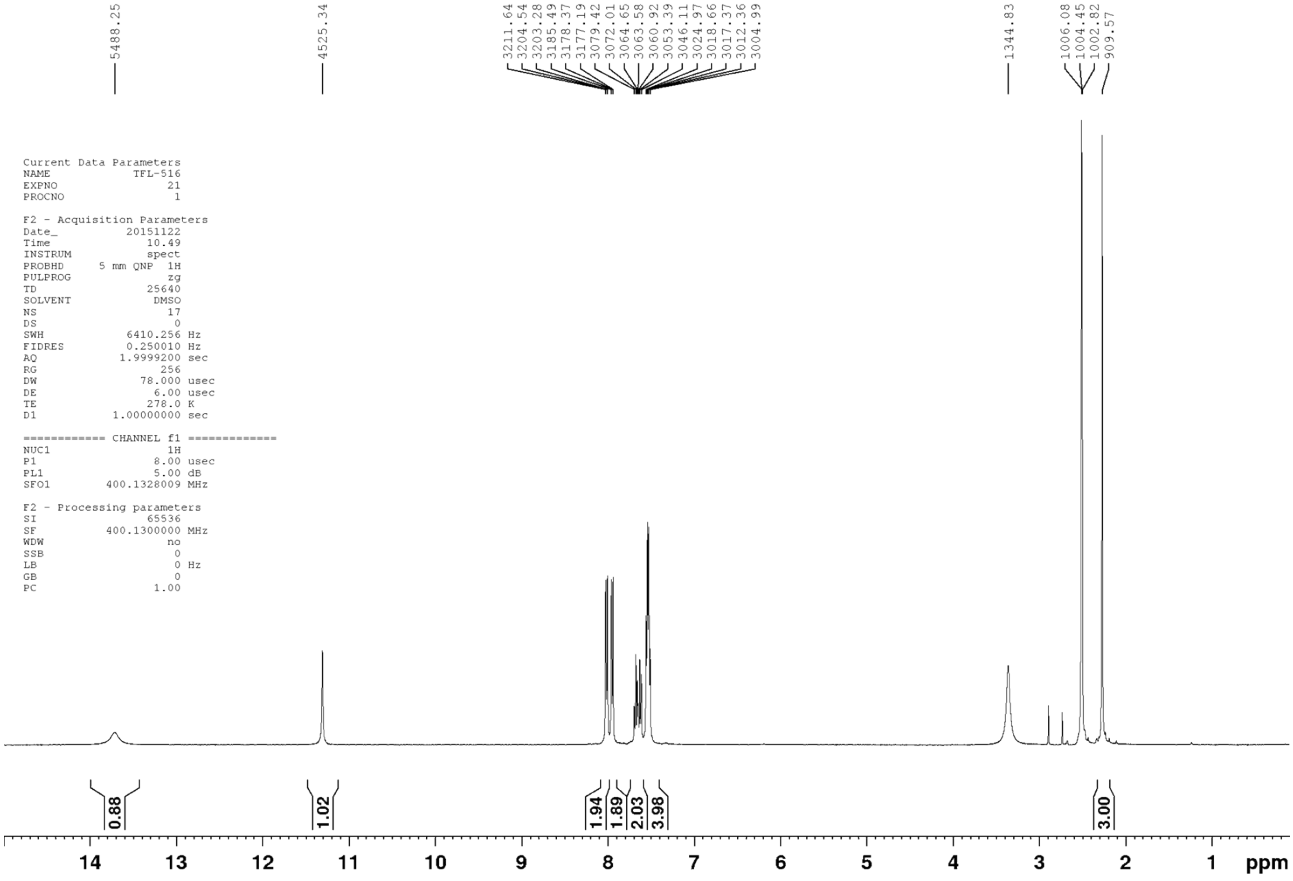


Figure S37. ^{1}H NMR spectrum of compound **30**.

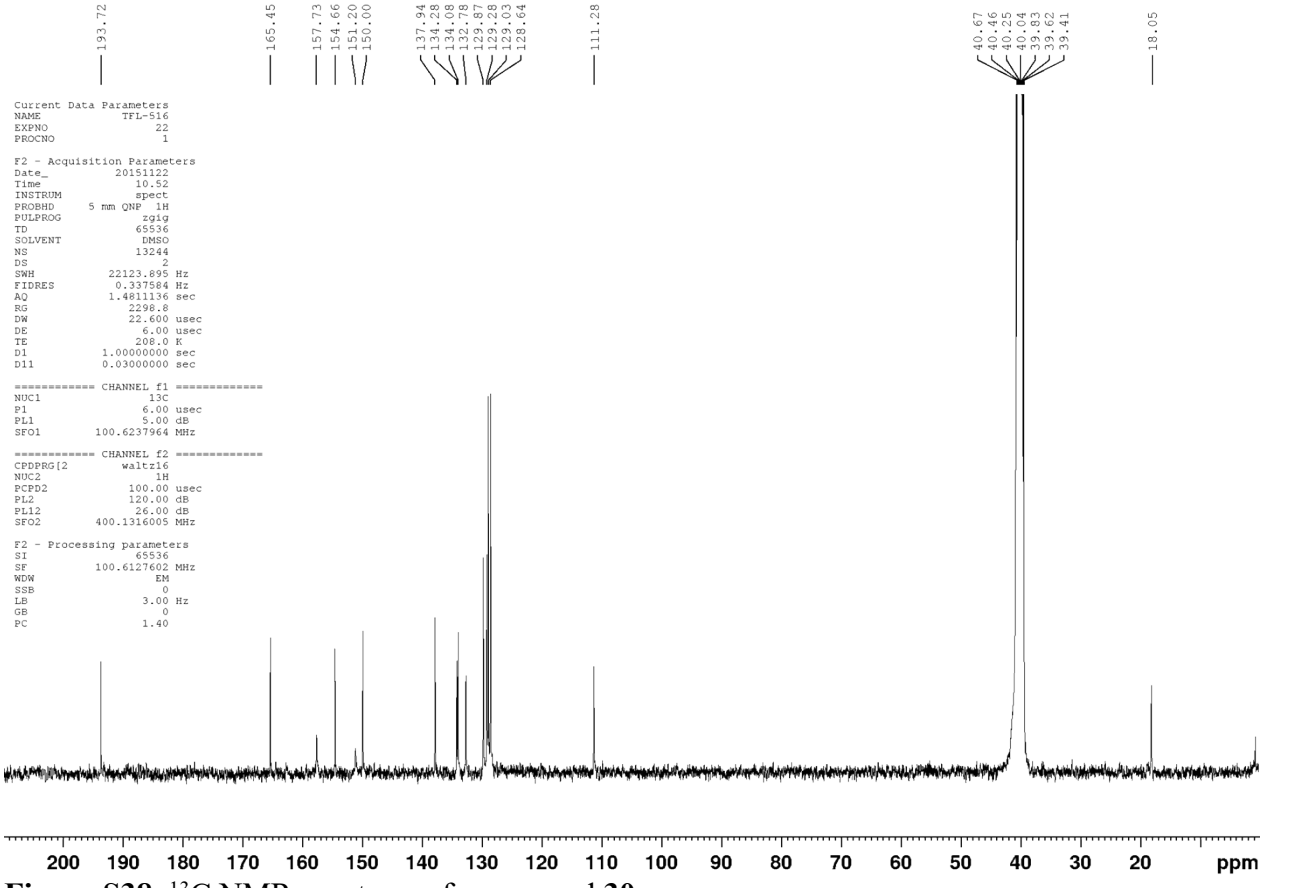


Figure S38. ^{13}C NMR spectrum of compound **30**.

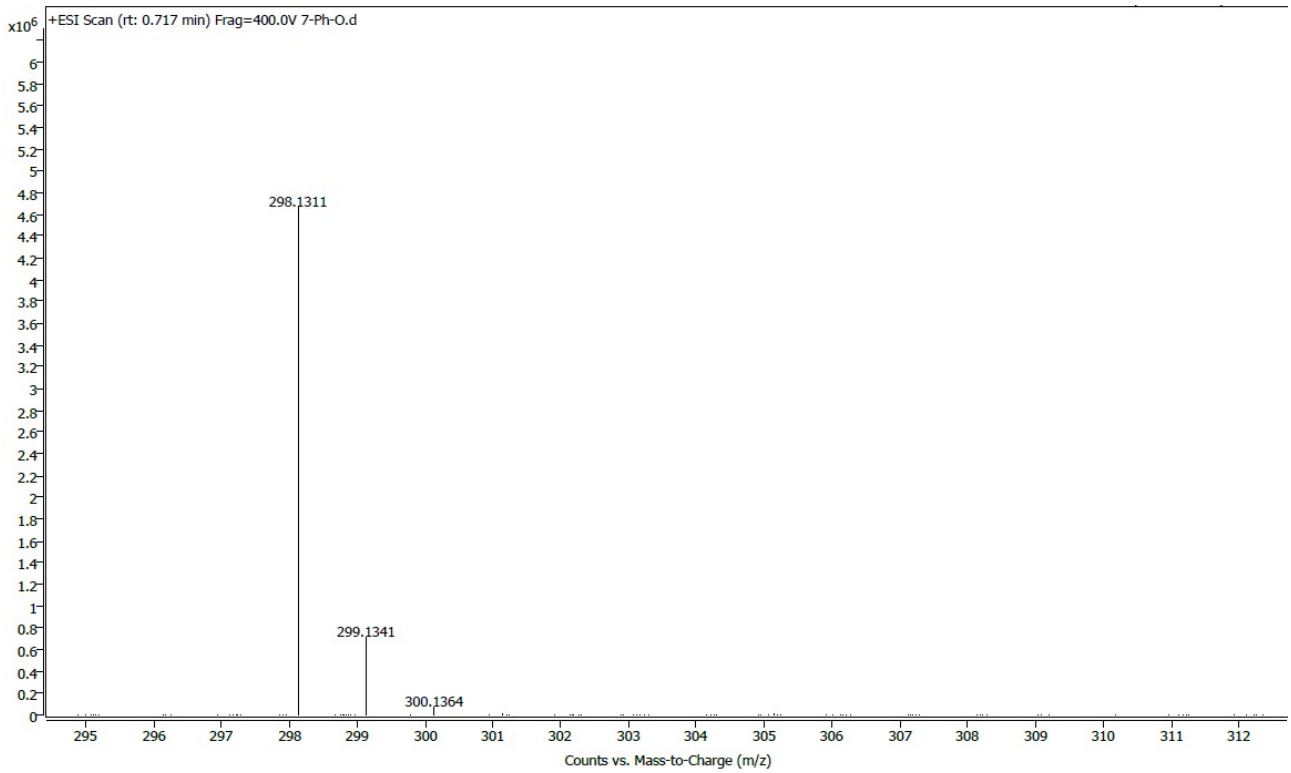


Figure S39. HRMS spectrum of compound **8**.

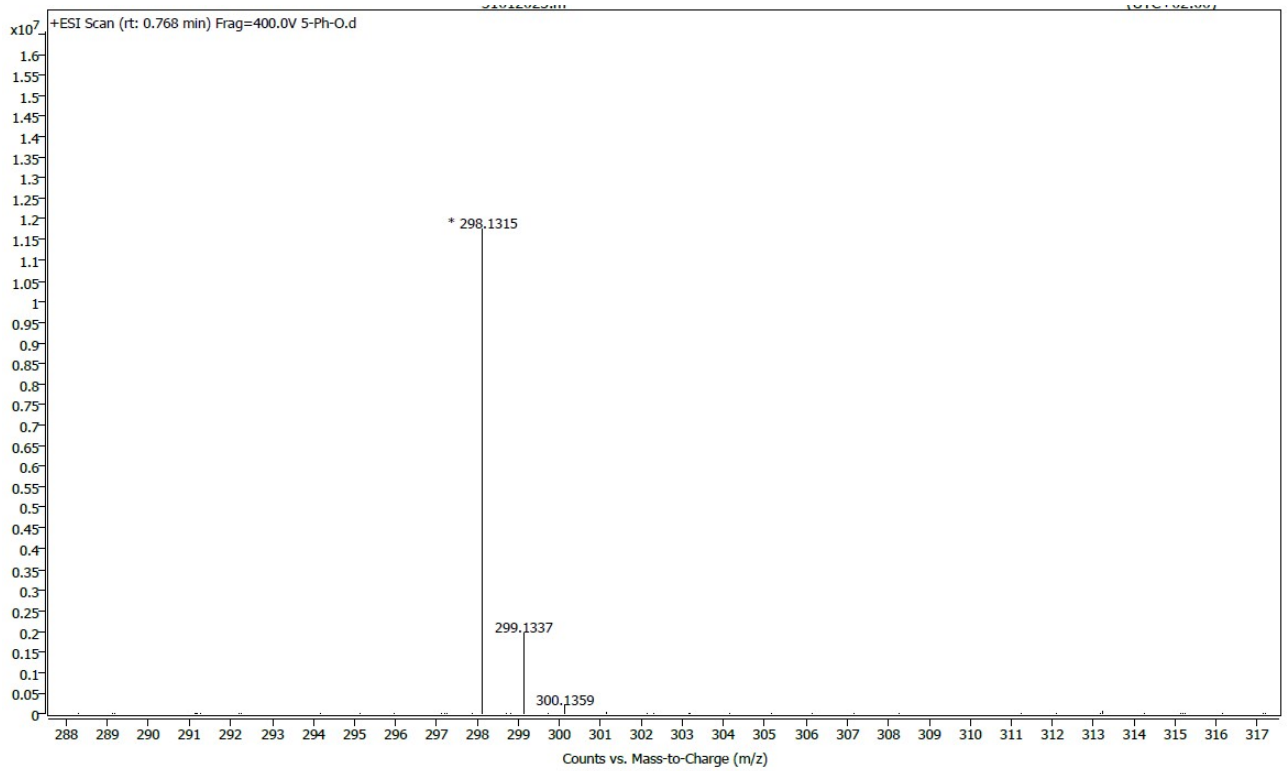


Figure S40. HRMS spectrum of compound **9**.

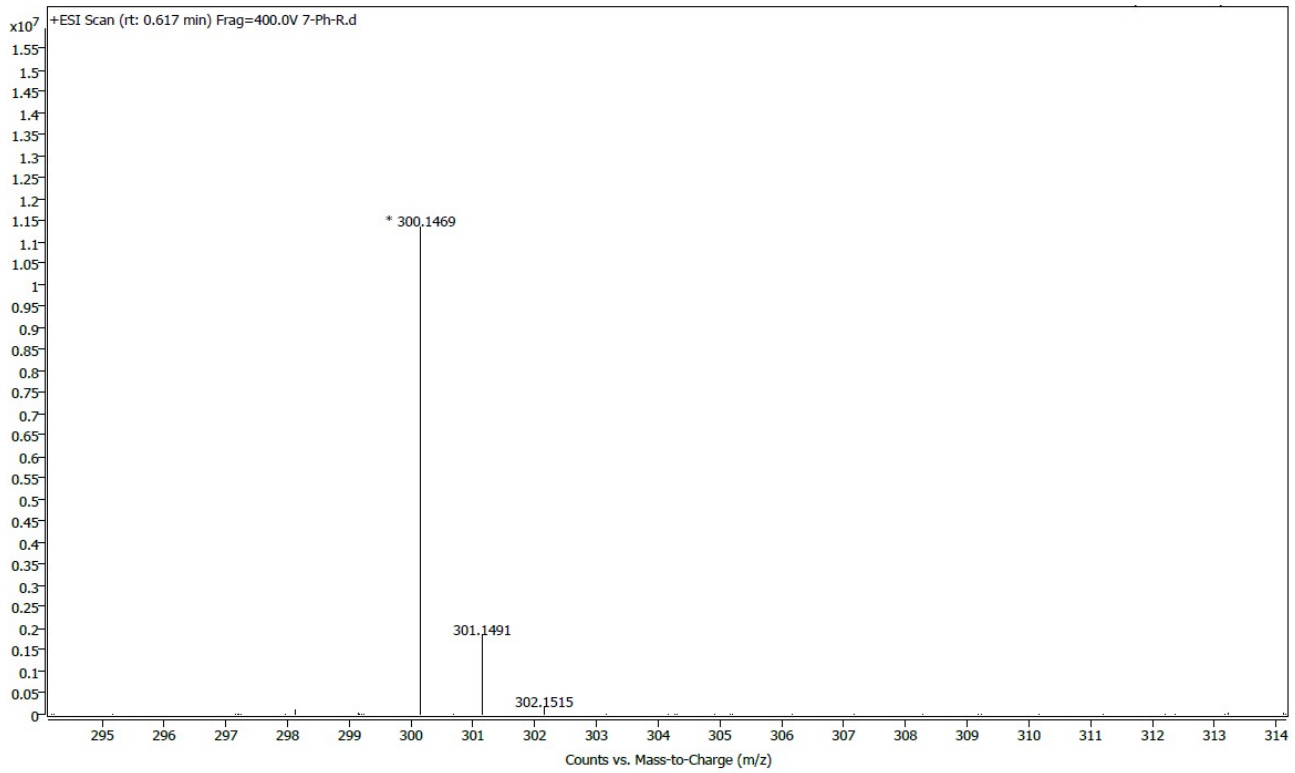


Figure S41. HRMS spectrum of compound **10**.

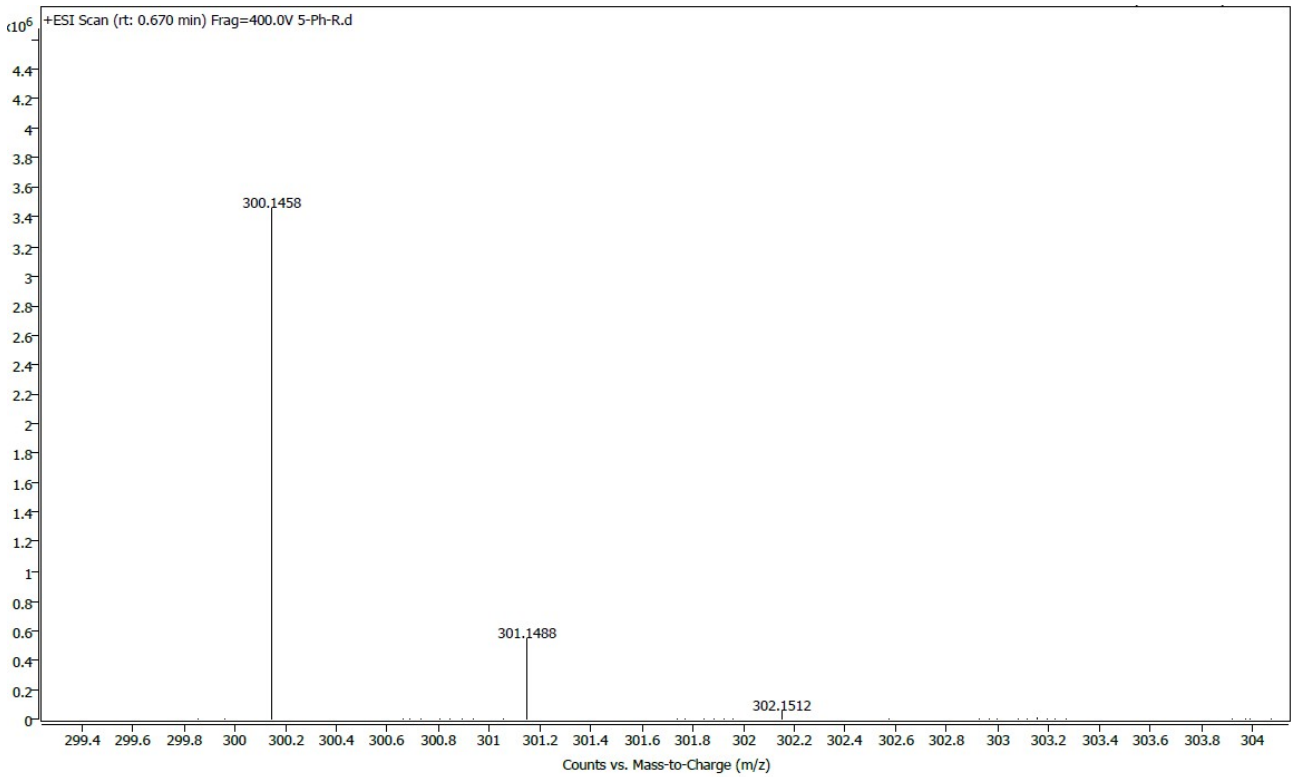
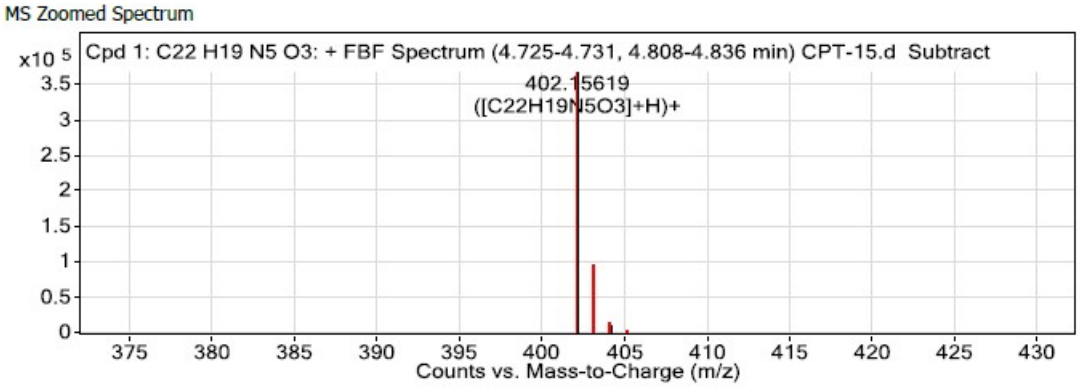


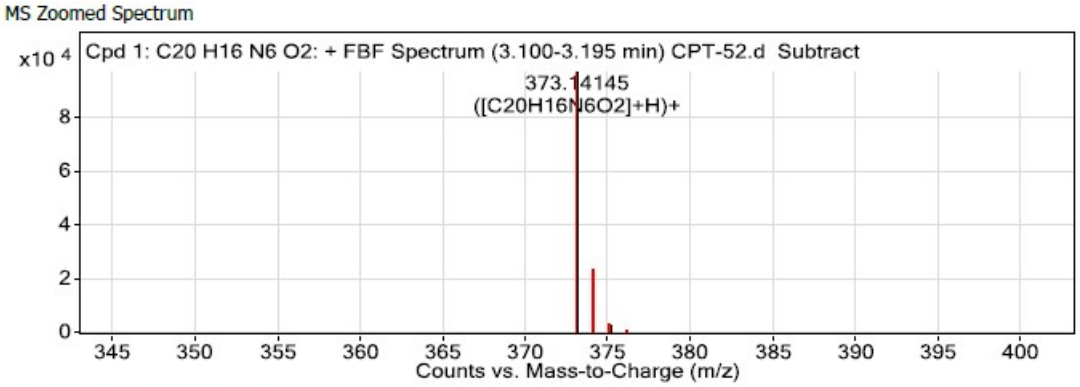
Figure S42. HRMS spectrum of compound **11**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
402.15619	1	367473.81	C22H19N5O3	(M+H)+
403.15968	1	88893.8	C22H19N5O3	(M+H)+
404.16214	1	12199.38	C22H19N5O3	(M+H)+
405.16341	1	1433.93	C22H19N5O3	(M+H)+

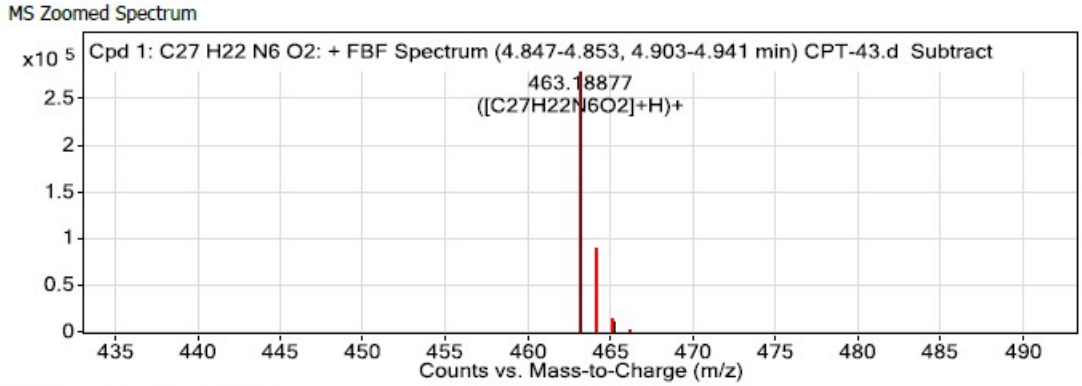
Figure S43. HRMS spectrum of compound **23**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
373.14145	1	97002.26	C20H16N6O2	(M+H)+
374.14408	1	22289.84	C20H16N6O2	(M+H)+
375.14602	1	3004.99	C20H16N6O2	(M+H)+
376.14973	1	159.01	C20H16N6O2	(M+H)+

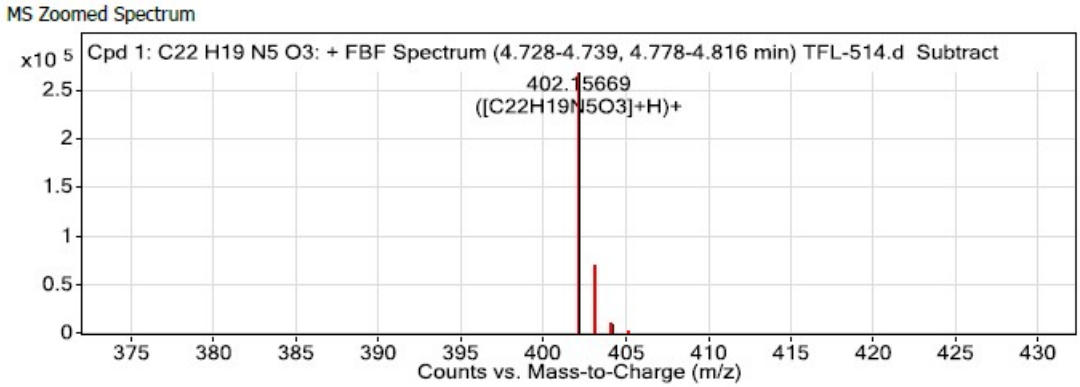
Figure S44. HRMS spectrum of compound **24**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
463.18877	1	279042.94	C27H22N6O2	(M+H)+
464.19234	1	83945.49	C27H22N6O2	(M+H)+
465.19464	1	12806.78	C27H22N6O2	(M+H)+
466.19689	1	1684.45	C27H22N6O2	(M+H)+

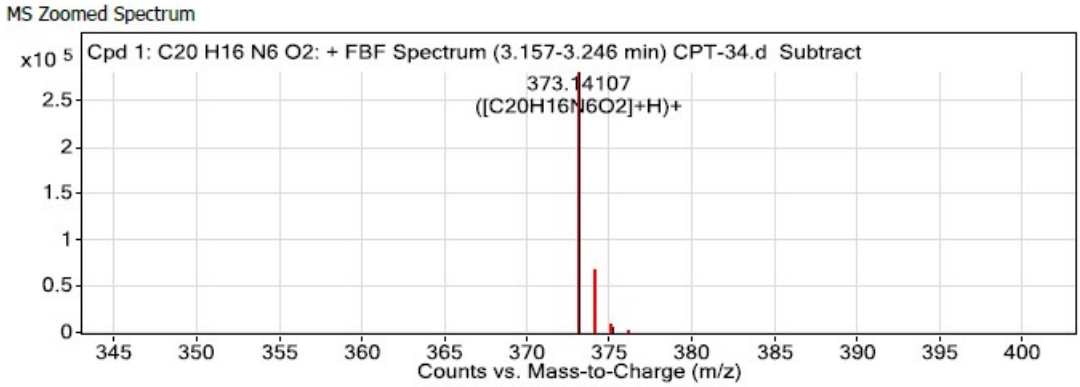
Figure S45. HRMS spectrum of compound **25**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
402.15669	1	268008.88	C22H19N5O3	(M+H)+
403.1601	1	66583.88	C22H19N5O3	(M+H)+
404.16256	1	9314.01	C22H19N5O3	(M+H)+
405.16529	1	974.6	C22H19N5O3	(M+H)+

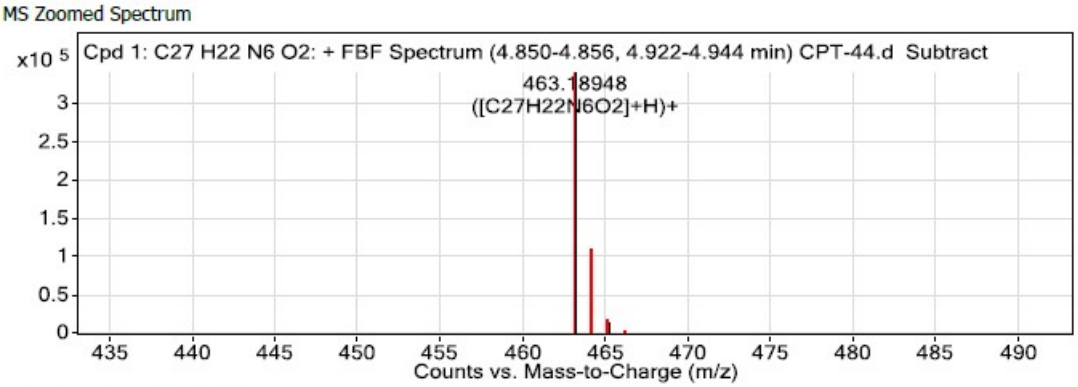
Figure S46. HRMS spectrum of compound **26**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
373.14107	1	280274.94	C20H16N6O2	(M+H)+
374.14451	1	61352.8	C20H16N6O2	(M+H)+
375.14679	1	7919.77	C20H16N6O2	(M+H)+
376.14815	1	599.68	C20H16N6O2	(M+H)+

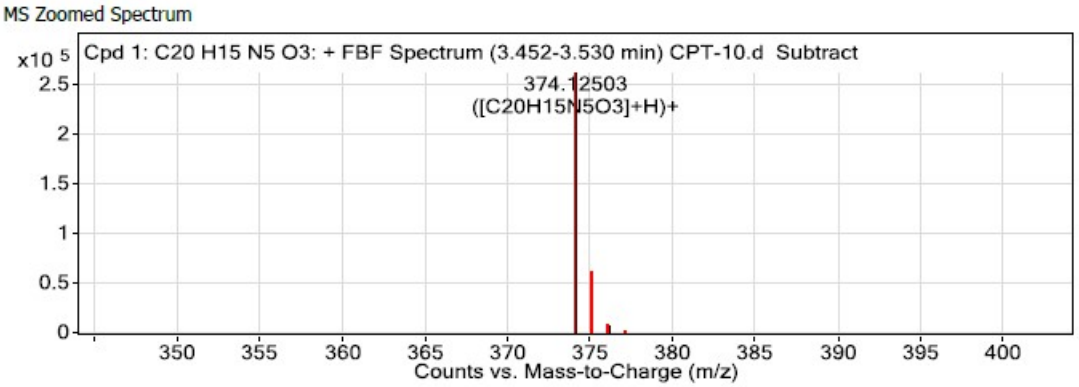
Figure S47. HRMS spectrum of compound **27**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
463.18948	1	341033.75	C27H22N6O2	(M+H)+
464.19315	1	101310.88	C27H22N6O2	(M+H)+
465.19528	1	16370.75	C27H22N6O2	(M+H)+
466.19814	1	2097.52	C27H22N6O2	(M+H)+

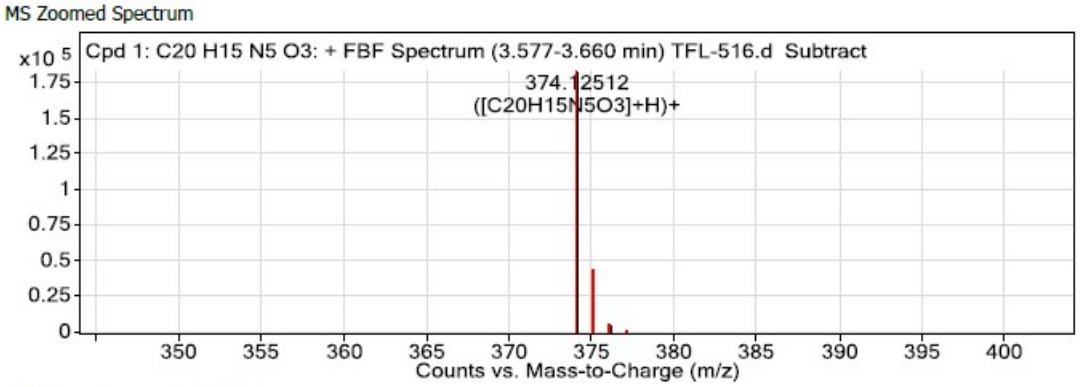
Figure S48. HRMS spectrum of compound **28**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
374.12503	1	261949.42	C20H15N5O3	(M+H)+
375.12828	1	57341.76	C20H15N5O3	(M+H)+
376.13065	1	8055.77	C20H15N5O3	(M+H)+
377.13225	1	862.65	C20H15N5O3	(M+H)+

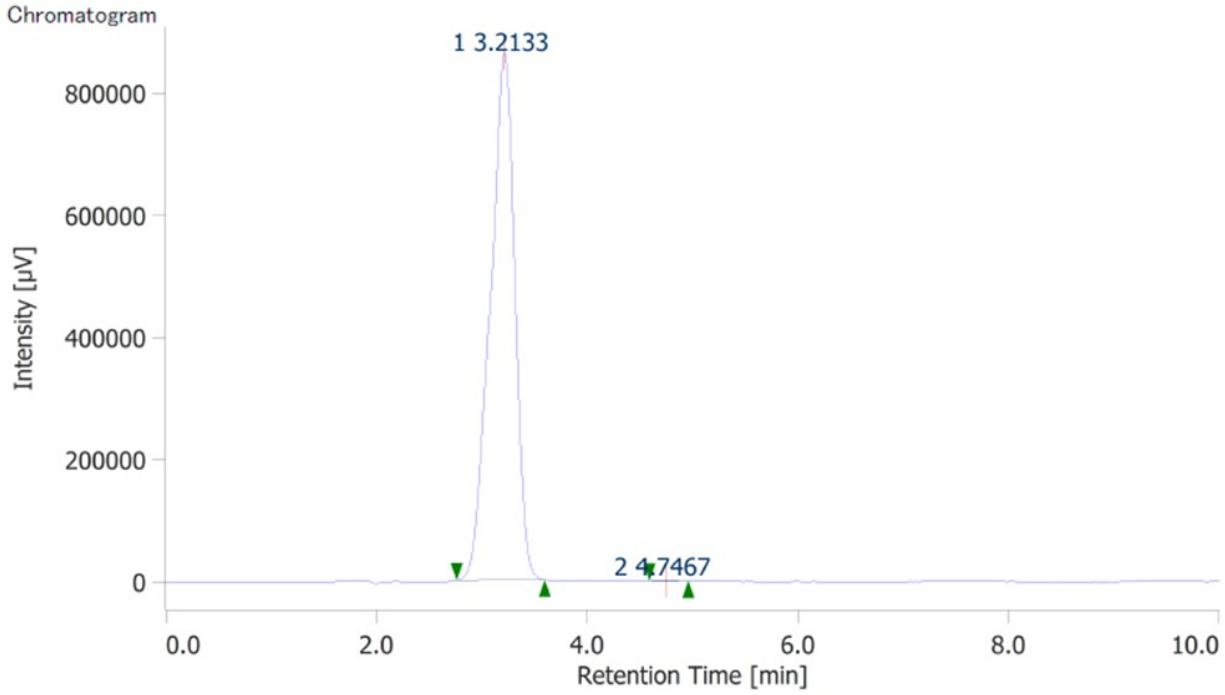
Figure S49. HRMS spectrum of compound **29**.



MS Spectrum Peak List

m/z	z	Abund	Formula	Ion
374.12512	1	182598.25	C20H15N5O3	(M+H)+
375.12821	1	41201.47	C20H15N5O3	(M+H)+
376.13063	1	5731.13	C20H15N5O3	(M+H)+
377.13311	1	379.1	C20H15N5O3	(M+H)+

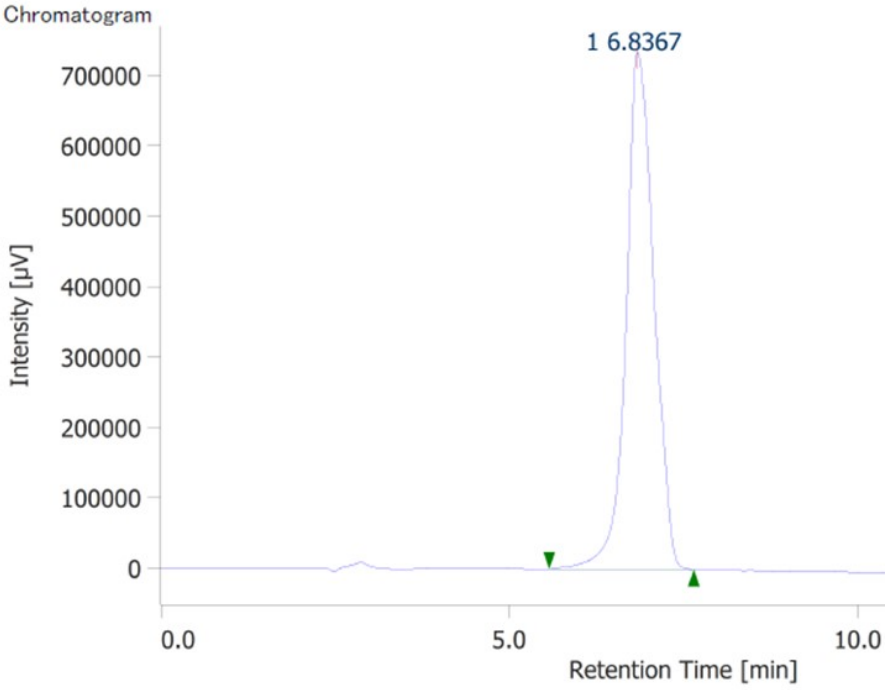
Figure S50. HRMS spectrum of compound **30**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	3.213	14019501	862507	99.930	99.920	N/A
2	Unknown	5	4.747	9783	691	0.070	0.080	N/A

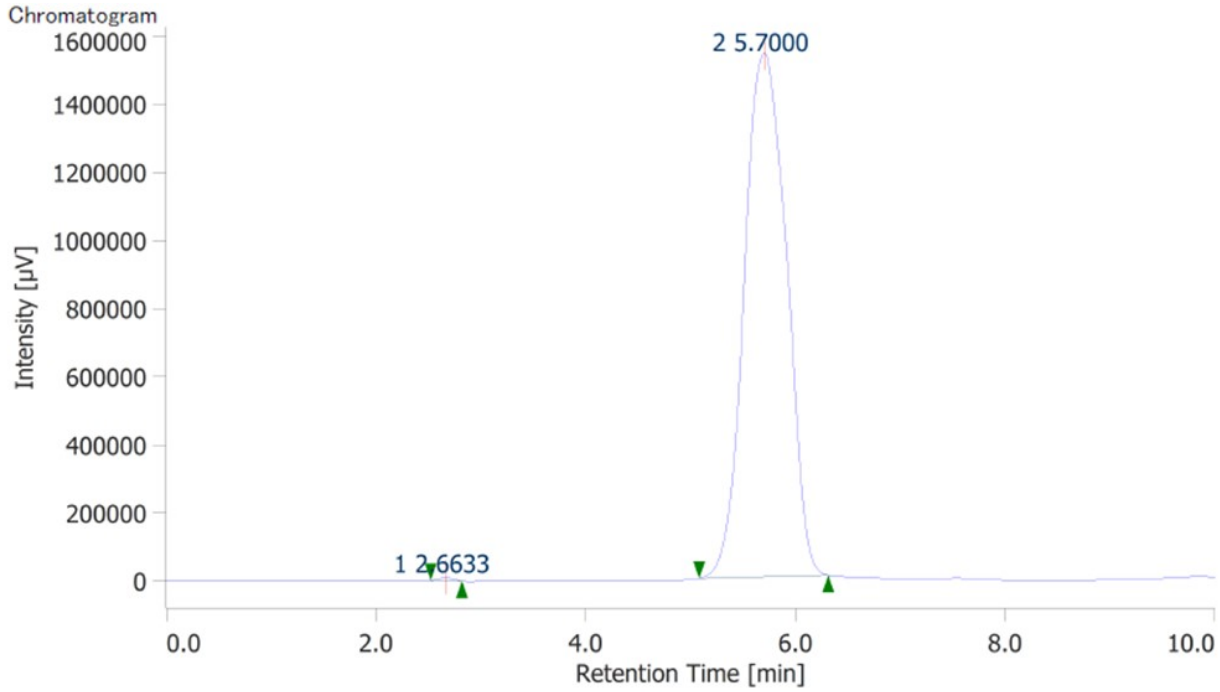
Figure S51. HPLC chromatogram of compound **23**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	6.837	20510696	734355	100.000	100.000	N/A

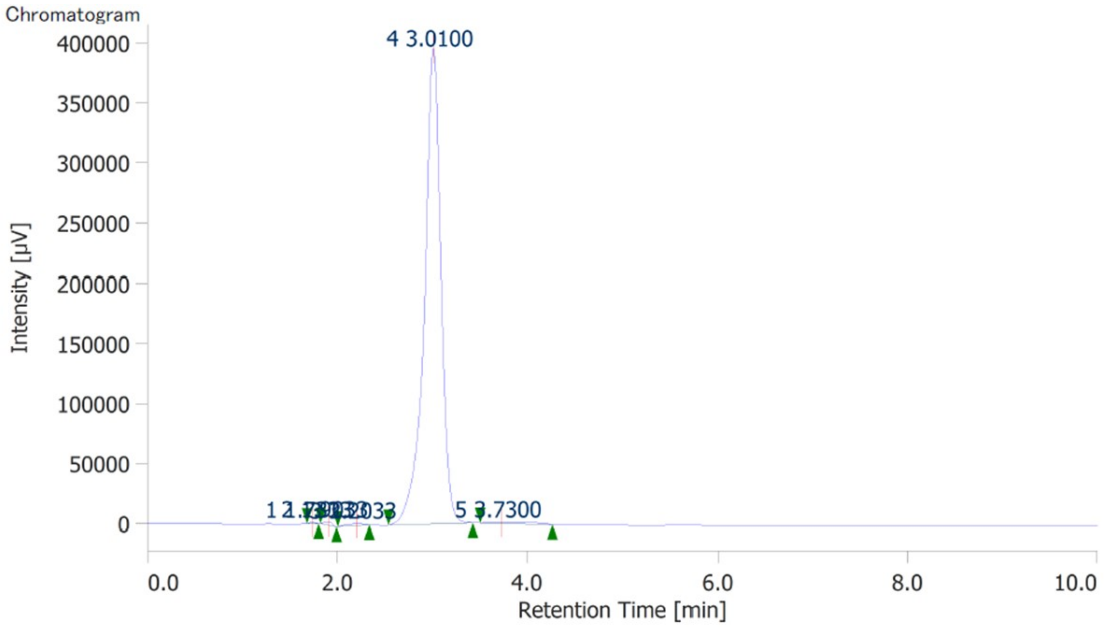
Figure S52. HPLC chromatogram of compound **24**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	2.663	90679	9931	0.210	0.642	N/A
2	Unknown	5	5.700	42996932	1537144	99.790	99.358	N/A

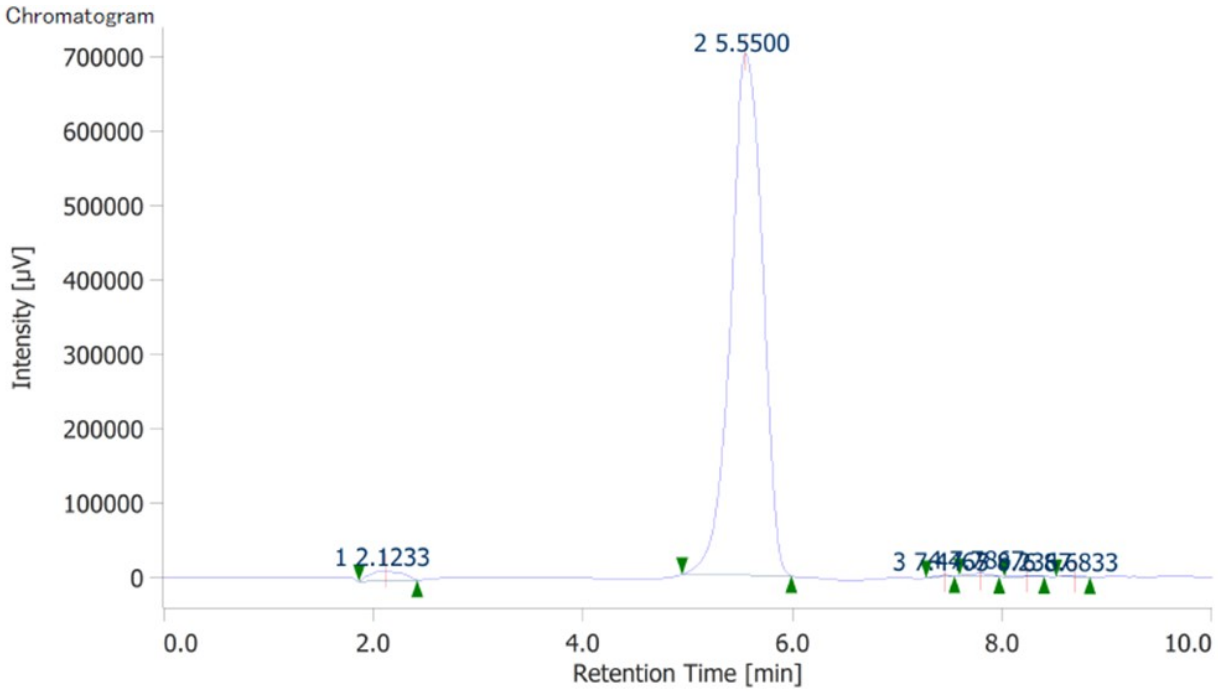
Figure S53. HPLC chromatogram of compound **25**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	1.733	4583	1366	0.094	0.339	N/A
2	Unknown	5	1.903	15724	2802	0.321	0.695	N/A
3	Unknown	5	2.203	20818	1839	0.425	0.456	N/A
4	Unknown	5	3.010	4819365	395764	98.382	98.196	N/A
5	Unknown	5	3.730	38126	1262	0.778	0.313	N/A

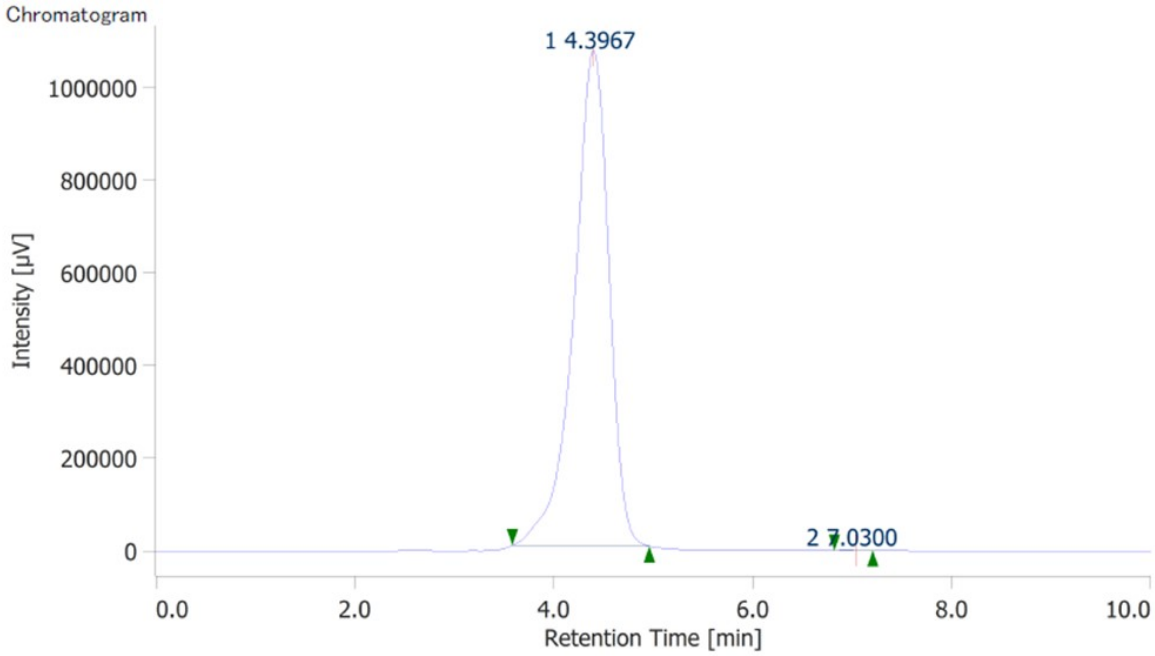
Figure S54. HPLC chromatogram of compound **26**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	2.123	287134	13863	1.898	1.917	N/A
2	Unknown	5	5.550	14741310	701137	97.437	96.939	N/A
3	Unknown	5	7.447	16253	1691	0.107	0.234	N/A
4	Unknown	5	7.787	47819	3593	0.316	0.497	N/A
5	Unknown	5	8.237	20727	1585	0.137	0.219	N/A
6	Unknown	5	8.683	15757	1405	0.104	0.194	N/A

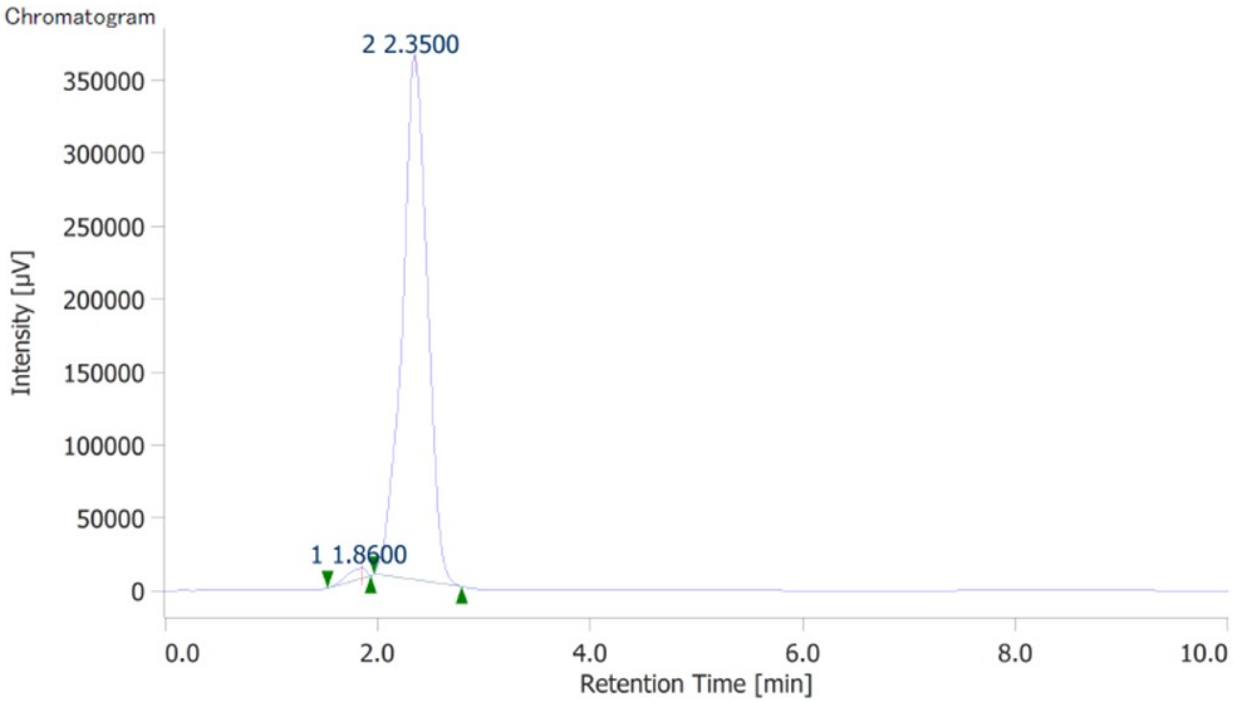
Figure S55. HPLC chromatogram of compound **27**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	4.397	26106716	1069122	99.985	99.976	N/A
2	Unknown	5	7.030	3810	255	0.015	0.024	N/A

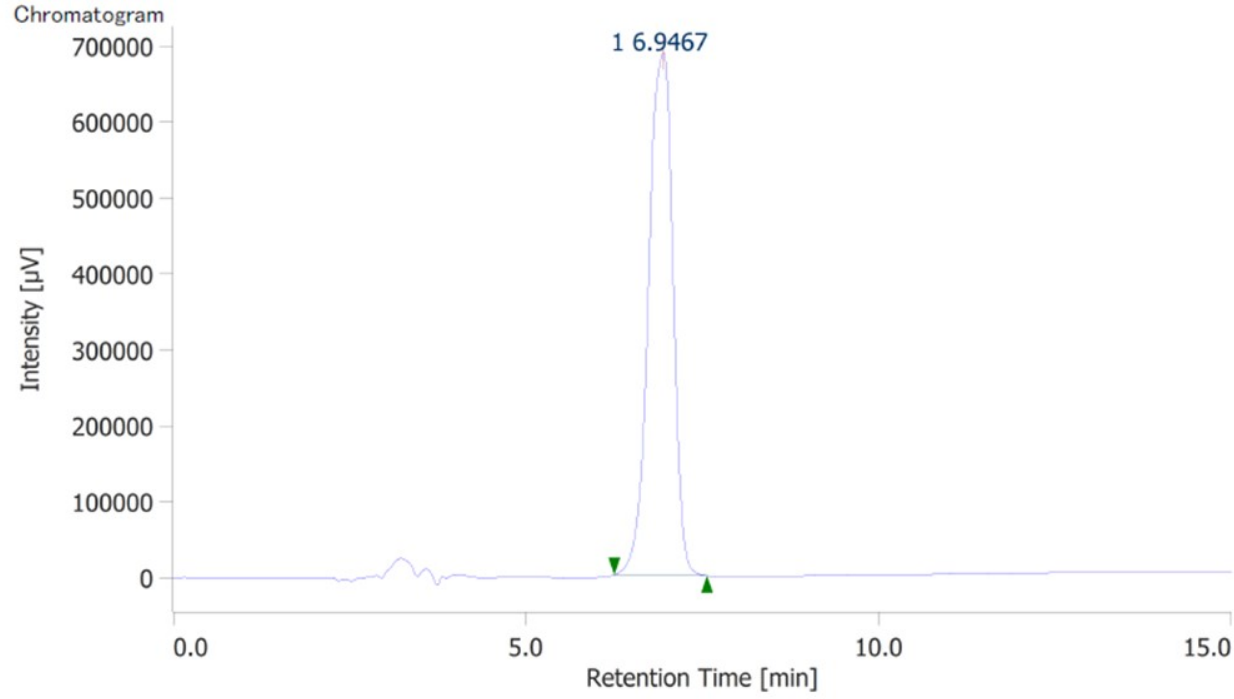
Figure S56. HPLC chromatogram of compound **28**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Peak-001	5	1.860	95154	6976	1.583	1.901	N/A
2	Peak-002	5	2.350	5915073	360046	98.417	98.099	N/A

Figure S57. HPLC chromatogram of compound **29**.



Peak Information

#	Peak Name	CH	tR [min]	Area [μV·sec]	Height [μV]	Area%	Height%	Quantity
1	Unknown	5	6.947	16586408	687921	100.000	100.000	N/A

Figure S58. HPLC chromatogram of compound **30**.

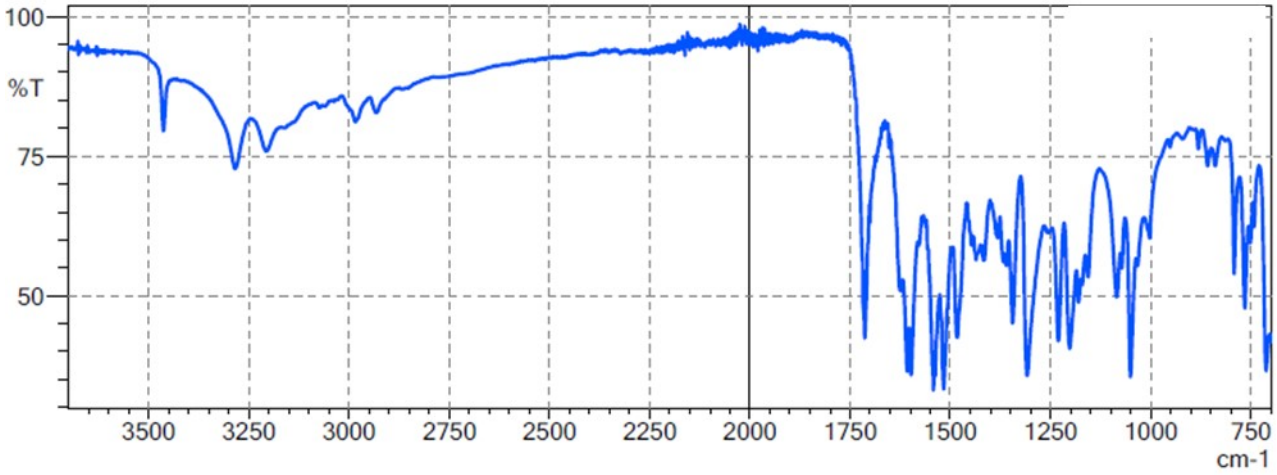


Figure S59. FT-IR spectrum of compound **8**.

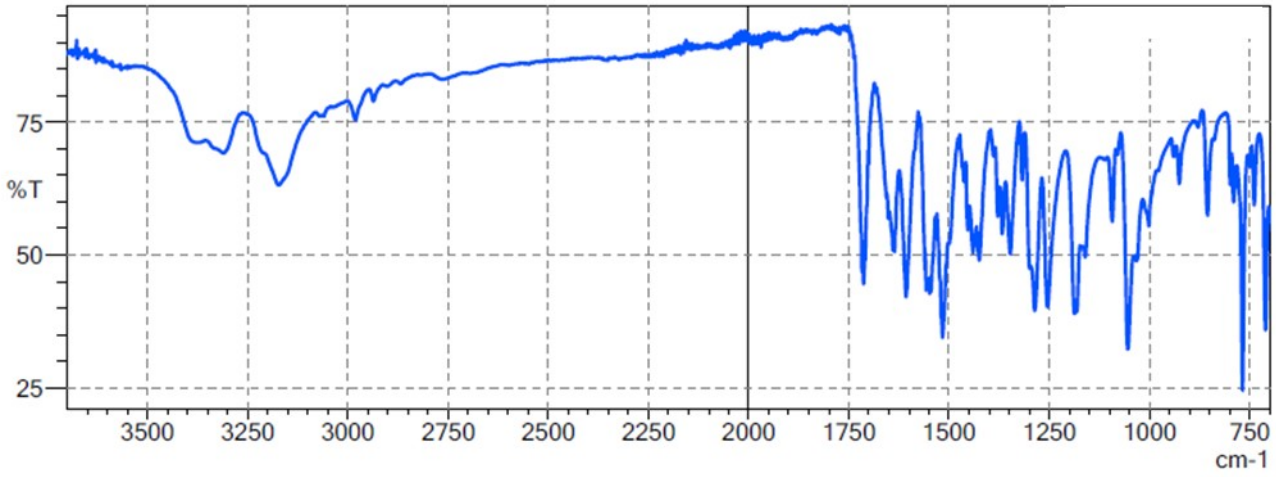


Figure S60. FT-IR spectrum of compound **9**.

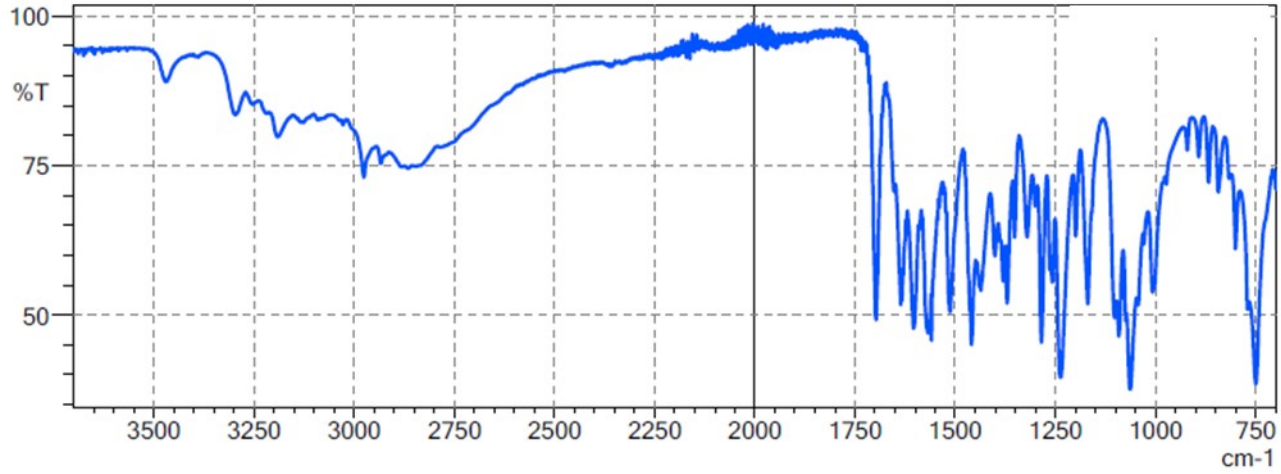


Figure S61. FT-IR spectrum of compound **10**.

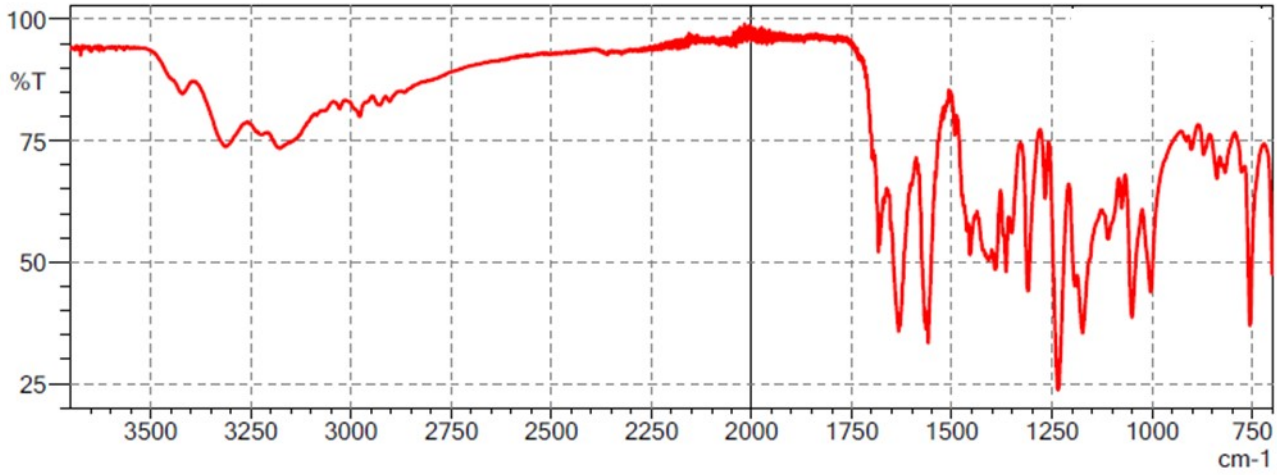


Figure S62. FT-IR spectrum of compound **11**.