

## **Supporting Information**

# **Cerasoidine, a Bis-aporphine Alkaloid Isolated from *Polyalthia cerasoides* During Screening for Wnt Signal Inhibitors**

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- 1) **Table S1.**  $^1\text{H}$  NMR spectroscopic data of **5**, **6**, and **9** in DMSO- $d_6$ .
- 2) **Table S2.**  $^1\text{H}$  NMR spectroscopic data of **7**, **8**, and **10** in DMSO- $d_6$ .
- 3) **Table S3**  $^{13}\text{C}$  NMR spectroscopic data of **5 - 10** in DMSO- $d_6$ .
- 4) **Figure S1.** X-ray analysis data of cerasoidine (**1**): The packing structure of **1** as indicated by a stick model.
- 5) **Table S4.** Crystal data for and structure refinement of compound **1**.
- 6) **Table S5.** Atomic coordinates ( $\times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for compound **1**. U(eq) is defined as one third of the trace of the orthogonalized  $U^{ij}$  tensor.
- 7) **Table S6.** Bond lengths [ $\text{\AA}$ ] and angles [ $^\circ$ ] for compound **1**.
- 8) **Table S7.** Anisotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for compound **1**. The anisotropic displacement factor exponent takes the form:  $-2\pi^2 [ h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12} ]$
- 9) **Table S8.** Hydrogen coordinates ( $\times 10^4$ ) and isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for compound **1**.
- 10) **Figure S2.** Chiral HPLC analysis of the isolated cerasoidine (**1**).

- 11) **Figure S3.** Comparison of the CD spectrum of cerasoidine (**1**) with that of (*S*)-(+)-9,10-dihydrodibenzo[c,g]phenanthrene<sup>6</sup>.
- 12) **Figure S4.** Comparison of CD spectra of compounds **7**, **8**, and **10** with (*S*)- and (*R*)-protoberberine alkaloids in the literature<sup>11</sup>.
- 13) **Figure S5.** Key COSY and HMBC correlations for new compounds (**1**, **5**, **6**, **7**, and **8**).
- 14) **Figure S6.** Inhibition of TCF/β-catenin transcriptional activities of compounds **1-13**.
- 15) **Figure S7.** Spectroscopic data of compound **1**.
- 16) **Figure S8.** Spectroscopic data of compound **5**.
- 17) **Figure S9.** Spectroscopic data of compound **6**.
- 18) **Figure S10.** Spectroscopic data of compound **7**.
- 19) **Figure S11.** Spectroscopic data of compound **8**.
- 20) **Figure S12.** Spectroscopic data of compound **9**, which were not available in the literature.<sup>10</sup> Therefore, we are the first to provide full spectroscopic data for **9**.
- 21) **Figure S13.** Spectroscopic data of compound **10**, which were not available in the literature.<sup>10</sup> Therefore, we are the first to provide full spectroscopic data for **10**.

1) **Table S1.**  $^1\text{H}$  NMR spectroscopic data of **5**, **6**, and **9** in  $\text{DMSO}-d_6$ .

position	<b>5<sup>a</sup></b>	<b>6<sup>b</sup></b>	<b>9<sup>a</sup></b>
1	7.23 s	7.01 s	7.37 s
2			
3			
4			6.69 s
4a			
5	2.84 t (6.2)	2.80 t (5.9)	2.79 t (6.2)
6	4.10 t (6.2)	4.09 t (5.9)	4.10 t (6.2)
8			
8a			
9			
10			
11	7.25 d (7.6)	7.26 d (8.7)	7.25 d (8.3)
12	7.28 d (7.6)	7.31 d (8.7)	7.28 d (8.3)
12a			
13	7.06 s	7.12 s	7.05 s
14			
14a			
2-OMe	3.88 s	3.88 s	3.86 s
3-OMe		3.71 s	
4-OMe	3.72 s		
9-OMe	3.77 s	3.77 s	3.76 s
3-OH	9.38 br s, 9.12 br s		9.46 br s or 9.36 br s
4-OH		9.20 s	
10-OH	9.38 br s, 9.12 br s	9.43 s	9.46 br s or 9.36 br s

(<sup>a</sup>Spectra obtained at 600 MHz. <sup>b</sup>Spectra obtained at 400 MHz).

2) **Table S2.**  $^1\text{H}$  NMR spectroscopic data of **7**, **8**, and **10** in  $\text{DMSO}-d_6$ .

position	<b>7<sup>a</sup></b>	<b>8<sup>a</sup></b>	<b>10<sup>b</sup></b>
1	6.74 s	6.51 s	6.88 s
2			
3			
4			6.59 s
4a			
5	2.86 d (15.8), 2.51 m	2.85 d (15.8), 2.40 m	2.65 m
6	4.76 ddd (13.1, 4.8, 2.8), 2.72 dt (12.4, 2.8)	4.78 ddd (12.4, 4.8, 2.1), 2.68 dt (12.4, 2.8)	4.72 dt (12.4, 3.7), 2.78 m
8			
8a			
9			
10			
11	6.96 d (8.3)	6.96 d (8.3)	6.96 d (7.8)
12	6.89 d (8.3)	6.89 d (8.3)	6.89 d (7.8)
12a			
13	3.16 dd (15.1, 2.8), 2.59 t (13.8)	3.16 dd (15.1, 2.8), 2.60 t (14.5)	3.16 dd (15.6, 3.2), 2.58 t (14.2)
14	4.65 dd (13.1, 2.8)	4.64 dd (13.1, 2.8)	4.63 dd (13.3, 3.2)
14a			
2-OMe	3.77 s	3.77 s	3.75 s
3-OMe		3.65 s	
4-OMe	3.69 s		
9-OMe	3.75 s	3.75 s	3.75 s
3-OH	8.62 s		8.96 s
4-OH		8.96 s	
10-OH	9.24 s	9.23 s	9.23 s

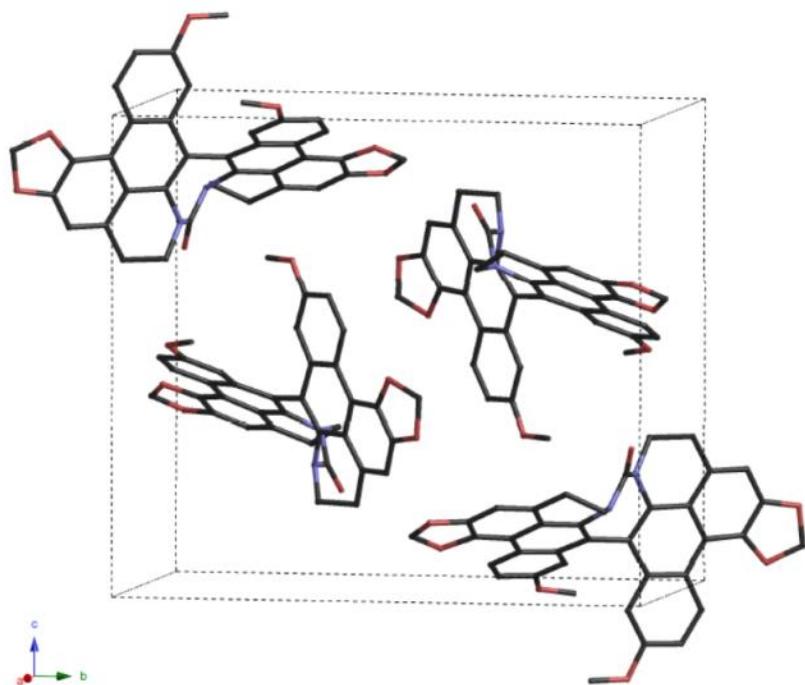
( <sup>a</sup>Spectra obtained at 600 MHz. <sup>b</sup>Spectra obtained at 400 MHz).

3) **Table S3**  $^{13}\text{C}$  NMR spectroscopic data of **5 - 10** in  $\text{DMSO}-d_6$ .

position	<b>5<sup>a</sup></b>	<b>6<sup>b</sup></b>	<b>7<sup>a</sup></b>	<b>8<sup>a</sup></b>	<b>9<sup>a</sup></b>	<b>10<sup>b</sup></b>
1	104.0	99.7	105.5	101.1	108.6	110.4
2	147.9	151.9	147.6	151.4	147.2	146.6
3	140.4	136.9	137.7	134.5	147.8	145.1
4	144.2	146.7	144.6	147.0	114.5	115.0
4a	121.7	116.0	120.8	115.6	128.3	122.8
5	21.3	21.1	22.9	28.5	27.1	28.5
6	38.6	38.6	37.3	37.2	39.0	37.7
8	158.8	158.8	161.6	161.6	158.9	161.6
8a	118.4	118.7	122.5	122.6	118.3	122.5
9	146.1	146.1	147.7	147.7	146.1	147.7
10	148.6	148.9	150.0	150.1	148.5	150.0
11	122.6	122.8	119.1	119.1	122.6	119.1
12	122.8	122.9	122.5	122.7	122.8	122.6
12a	131.2	130.9	129.7	129.7	131.3	129.8
13	100.7	101.8	38.0	38.0	100.3	38.1
14	134.7	134.5	54.2	54.4	134.8	54.3
14a	120.2	125.3	126.4	131.8	120.6	127.1
2-OMe	56.2	55.9	56.0	55.7	56.0	58.8
3-OMe		60.3		60.2		
4-OMe	61.0		59.7			
9-OMe	60.2	61.1	60.7	60.7	61.0	60.7

( <sup>a</sup>Spectra obtained at 150 MHz. <sup>b</sup>Spectra obtained at 100 MHz.).

4) **Figure S1.** X-ray analysis data of cerasoidine (**1**): The packing structure of **1** as indicated by a stick model.



Hydrogen atoms are omitted for clarity.

5) **Table S4.** Crystal data for and structure refinement of compound **1**.

Empirical formula	C37 H26 N2 O7				
Formula weight	610.6				
Temperature	173 K				
Wavelength	1.54178 Å				
Crystal system	Monoclinic				
Space group	<i>P</i> 2 <sub>1</sub> / <i>c</i>				
Unit cell dimensions	a = 8.2263(2) Å	$\alpha = 90^\circ$ .			
	b = 19.2406(5) Å	$\beta = 93.261(2)^\circ$ .			
	c = 16.9322(5) Å	$\gamma = 90^\circ$ .			
Volume	2675.67(12) Å <sup>3</sup>				
Z	4				
Density (calculated)	1.516 Mg/m <sup>3</sup>				
Absorption coefficient	0.870 mm <sup>-1</sup>				
F(000)	1272				
Crystal size	0.150 x 0.100 x 0.020 mm <sup>3</sup>				
Theta range for data collection	3.480 to 68.318°.				
Index ranges	-9<=h<=9, -22<=k<=23, -20<=l<=20				
Reflections collected	18590				
Independent reflections	4869 [R(int) = 0.0351]				
Completeness to theta = 67.679°	99.50%				
Absorption correction	Empirical				
Max. and min. transmission	0.98 and 0.88				
Refinement method	Full-matrix least-squares on F <sup>2</sup>				
Data / restraints / parameters	4869 / 0 / 417				
Goodness-of-fit on F <sup>2</sup>	1.034				
Final R indices [I>2sigma(I)]	R1 = 0.0525, wR2 = 0.1363				
R indices (all data)	R1 = 0.0784, wR2 = 0.1526				
Largest diff. peak and hole	0.487 and -0.234 e.Å <sup>-3</sup>				

6) **Table S5.** Atomic coordinates ( $x \times 10^4$ ) and equivalent isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for compound **1**.  $U(\text{eq})$  is defined as one third of the trace of the orthogonalized  $U^{ij}$  tensor.

atom	x	y	z	$U(\text{eq})$
C(1)	7725(4)	3805(1)	2709(2)	43(1)
C(2)	10251(4)	3947(1)	2068(1)	46(1)
C(3)	10623(4)	4717(2)	2048(2)	51(1)
C(4)	11514(4)	4938(1)	2808(2)	42(1)
C(5)	12514(4)	5514(2)	2811(2)	47(1)
C(6)	13236(3)	5720(1)	3520(2)	45(1)
C(7)	14415(4)	6344(2)	4485(2)	52(1)
C(8)	13017(3)	5379(1)	4214(2)	41(1)
C(9)	12031(3)	4788(1)	4255(1)	35(1)
C(10)	11270(3)	4567(1)	3519(1)	36(1)
C(11)	10226(3)	3966(1)	3520(1)	35(1)
C(12)	11675(3)	4432(1)	4982(1)	33(1)
C(13)	12305(3)	4661(1)	5733(1)	38(1)
C(14)	11917(3)	4330(1)	6415(1)	39(1)
C(15)	10858(3)	3760(1)	6377(1)	36(1)
C(16)	10220(3)	3525(1)	5660(1)	34(1)
C(17)	10640(3)	3847(1)	4946(1)	32(1)
C(18)	9981(3)	3588(1)	4188(1)	33(1)
C(19)	9526(4)	2880(2)	7084(2)	53(1)
C(20)	5169(4)	3702(2)	3342(2)	51(1)
C(21)	4271(4)	3094(2)	2944(2)	58(1)
C(22)	4857(4)	2415(2)	3294(2)	49(1)
C(23)	3851(4)	1838(2)	3228(2)	61(1)
C(24)	4408(4)	1224(2)	3554(2)	58(1)
C(25)	4753(5)	112(2)	3896(2)	82(1)
C(26)	5902(4)	1165(2)	3944(2)	50(1)
C(27)	6991(3)	1725(1)	4030(1)	40(1)
C(28)	6422(3)	2366(1)	3687(1)	40(1)
C(29)	7489(3)	2962(1)	3750(1)	36(1)
C(30)	8609(3)	1684(1)	4396(1)	37(1)
C(31)	9253(4)	1048(1)	4710(2)	46(1)

C(32)	10799(4)	1001(1)	5028(2)	49(1)
C(33)	11791(3)	1581(1)	5064(1)	42(1)
C(34)	11239(3)	2213(1)	4757(1)	36(1)
C(35)	9623(3)	2278(1)	4432(1)	34(1)
C(36)	9000(3)	2942(1)	4128(1)	33(1)
C(37)	14297(4)	2032(2)	5553(2)	56(1)
N(1)	9407(3)	3774(1)	2783(1)	38(1)
N(2)	6941(3)	3591(1)	3368(1)	41(1)
O(1)	7002(3)	4001(1)	2107(1)	61(1)
O(2)	14214(3)	6292(1)	3647(1)	56(1)
O(3)	13877(3)	5709(1)	4825(1)	49(1)
O(4)	10537(2)	3477(1)	7089(1)	44(1)
O(5)	3622(3)	593(2)	3551(2)	79(1)
O(6)	6121(3)	490(1)	4212(1)	61(1)
O(7)	13295(3)	1463(1)	5420(1)	57(1)

7) **Table S6.** Bond lengths [ $\text{\AA}$ ] and angles [ $^\circ$ ] for compound **1**.

C(1)-O(1)	1.210(3)
C(1)-N(1)	1.383(4)
C(1)-N(2)	1.384(4)
C(2)-N(1)	1.468(3)
C(2)-C(3)	1.514(4)
C(2)-H(2A)	0.99
C(2)-H(2B)	0.99
C(3)-C(4)	1.505(4)
C(3)-H(3A)	0.99
C(3)-H(3B)	0.99
C(4)-C(5)	1.380(4)
C(4)-C(10)	1.424(3)
C(5)-C(6)	1.367(4)
C(5)-H(5)	0.95
C(6)-C(8)	1.368(4)
C(6)-O(2)	1.373(3)
C(7)-O(2)	1.422(4)
C(7)-O(3)	1.432(3)
C(7)-H(7A)	0.99
C(7)-H(7B)	0.99
C(8)-O(3)	1.375(3)
C(8)-C(9)	1.400(4)
C(9)-C(10)	1.427(3)
C(9)-C(12)	1.452(3)
C(10)-C(11)	1.441(4)
C(11)-C(18)	1.370(3)
C(11)-N(1)	1.432(3)
C(12)-C(17)	1.412(3)
C(12)-C(13)	1.417(3)
C(13)-C(14)	1.372(4)
C(13)-H(13)	0.95
C(14)-C(15)	1.399(4)
C(14)-H(14)	0.95
C(15)-O(4)	1.361(3)

C(15)-C(16)	1.372(3)
C(16)-C(17)	1.419(3)
C(16)-H(16)	0.95
C(17)-C(18)	1.452(3)
C(18)-C(36)	1.482(3)
C(19)-O(4)	1.418(3)
C(19)-H(19A)	0.98
C(19)-H(19B)	0.98
C(19)-H(19C)	0.98
C(20)-N(2)	1.472(4)
C(20)-C(21)	1.521(4)
C(20)-H(20A)	0.99
C(20)-H(20B)	0.99
C(21)-C(22)	1.501(4)
C(21)-H(21A)	0.99
C(21)-H(21B)	0.99
C(22)-C(23)	1.386(5)
C(22)-C(28)	1.418(4)
C(23)-C(24)	1.372(5)
C(23)-H(23)	0.95
C(24)-C(26)	1.366(5)
C(24)-O(5)	1.375(4)
C(25)-O(5)	1.414(5)
C(25)-O(6)	1.419(4)
C(25)-H(25A)	0.99
C(25)-H(25B)	0.99
C(26)-O(6)	1.384(4)
C(26)-C(27)	1.403(4)
C(27)-C(28)	1.431(4)
C(27)-C(30)	1.439(4)
C(28)-C(29)	1.444(4)
C(29)-C(36)	1.367(4)
C(29)-N(2)	1.433(3)
C(30)-C(35)	1.414(4)
C(30)-C(31)	1.423(4)

C(31)-C(32)	1.356(4)
C(31)-H(31)	0.95
C(32)-C(33)	1.382(4)
C(32)-H(32)	0.95
C(33)-O(7)	1.364(3)
C(33)-C(34)	1.389(3)
C(34)-C(35)	1.414(4)
C(34)-H(34)	0.95
C(35)-C(36)	1.459(3)
C(37)-O(7)	1.382(4)
C(37)-H(37A)	0.98
C(37)-H(37B)	0.98
C(37)-H(37C)	0.98

O(1)-C(1)-N(1)	122.1(3)
O(1)-C(1)-N(2)	122.9(3)
N(1)-C(1)-N(2)	115.0(2)
N(1)-C(2)-C(3)	110.2(2)
N(1)-C(2)-H(2A)	109.6
C(3)-C(2)-H(2A)	109.6
N(1)-C(2)-H(2B)	109.6
C(3)-C(2)-H(2B)	109.6
H(2A)-C(2)-H(2B)	108.1
C(4)-C(3)-C(2)	110.2(2)
C(4)-C(3)-H(3A)	109.6
C(2)-C(3)-H(3A)	109.6
C(4)-C(3)-H(3B)	109.6
C(2)-C(3)-H(3B)	109.6
H(3A)-C(3)-H(3B)	108.1
C(5)-C(4)-C(10)	120.8(2)
C(5)-C(4)-C(3)	119.4(2)
C(10)-C(4)-C(3)	119.8(2)
C(6)-C(5)-C(4)	117.7(2)
C(6)-C(5)-H(5)	121.1
C(4)-C(5)-H(5)	121.1

C(5)-C(6)-C(8)	122.9(3)
C(5)-C(6)-O(2)	126.3(3)
C(8)-C(6)-O(2)	110.7(3)
O(2)-C(7)-O(3)	108.6(2)
O(2)-C(7)-H(7A)	110.0
O(3)-C(7)-H(7A)	110.0
O(2)-C(7)-H(7B)	110.0
O(3)-C(7)-H(7B)	110.0
H(7A)-C(7)-H(7B)	108.3
C(6)-C(8)-O(3)	109.7(2)
C(6)-C(8)-C(9)	122.5(3)
O(3)-C(8)-C(9)	127.8(2)
C(8)-C(9)-C(10)	115.2(2)
C(8)-C(9)-C(12)	124.7(2)
C(10)-C(9)-C(12)	120.0(2)
C(4)-C(10)-C(9)	120.9(2)
C(4)-C(10)-C(11)	121.0(2)
C(9)-C(10)-C(11)	118.1(2)
C(18)-C(11)-N(1)	119.9(2)
C(18)-C(11)-C(10)	122.9(2)
N(1)-C(11)-C(10)	117.2(2)
C(17)-C(12)-C(13)	118.4(2)
C(17)-C(12)-C(9)	119.3(2)
C(13)-C(12)-C(9)	122.3(2)
C(14)-C(13)-C(12)	121.3(2)
C(14)-C(13)-H(13)	119.3
C(12)-C(13)-H(13)	119.3
C(13)-C(14)-C(15)	120.0(2)
C(13)-C(14)-H(14)	120.0
C(15)-C(14)-H(14)	120.0
O(4)-C(15)-C(16)	124.6(2)
O(4)-C(15)-C(14)	115.1(2)
C(16)-C(15)-C(14)	120.3(2)
C(15)-C(16)-C(17)	120.7(2)
C(15)-C(16)-H(16)	119.6

C(17)-C(16)-H(16)	119.6
C(12)-C(17)-C(16)	119.2(2)
C(12)-C(17)-C(18)	120.4(2)
C(16)-C(17)-C(18)	120.4(2)
C(11)-C(18)-C(17)	118.8(2)
C(11)-C(18)-C(36)	119.6(2)
C(17)-C(18)-C(36)	121.5(2)
O(4)-C(19)-H(19A)	109.5
O(4)-C(19)-H(19B)	109.5
H(19A)-C(19)-H(19B)	109.5
O(4)-C(19)-H(19C)	109.5
H(19A)-C(19)-H(19C)	109.5
H(19B)-C(19)-H(19C)	109.5
N(2)-C(20)-C(21)	110.9(2)
N(2)-C(20)-H(20A)	109.5
C(21)-C(20)-H(20A)	109.5
N(2)-C(20)-H(20B)	109.5
C(21)-C(20)-H(20B)	109.5
H(20A)-C(20)-H(20B)	108.0
C(22)-C(21)-C(20)	110.9(2)
C(22)-C(21)-H(21A)	109.5
C(20)-C(21)-H(21A)	109.5
C(22)-C(21)-H(21B)	109.5
C(20)-C(21)-H(21B)	109.5
H(21A)-C(21)-H(21B)	108.0
C(23)-C(22)-C(28)	120.4(3)
C(23)-C(22)-C(21)	119.2(3)
C(28)-C(22)-C(21)	120.4(3)
C(24)-C(23)-C(22)	118.3(3)
C(24)-C(23)-H(23)	120.9
C(22)-C(23)-H(23)	120.9
C(26)-C(24)-C(23)	122.4(3)
C(26)-C(24)-O(5)	109.8(3)
C(23)-C(24)-O(5)	127.8(3)
O(5)-C(25)-O(6)	108.0(3)

O(5)-C(25)-H(25A)	110.1
O(6)-C(25)-H(25A)	110.1
O(5)-C(25)-H(25B)	110.1
O(6)-C(25)-H(25B)	110.1
H(25A)-C(25)-H(25B)	108.4
C(24)-C(26)-O(6)	109.4(3)
C(24)-C(26)-C(27)	122.5(3)
O(6)-C(26)-C(27)	128.0(3)
C(26)-C(27)-C(28)	115.3(3)
C(26)-C(27)-C(30)	124.9(3)
C(28)-C(27)-C(30)	119.7(2)
C(22)-C(28)-C(27)	121.0(3)
C(22)-C(28)-C(29)	120.9(3)
C(27)-C(28)-C(29)	118.1(2)
C(36)-C(29)-N(2)	119.3(2)
C(36)-C(29)-C(28)	123.0(2)
N(2)-C(29)-C(28)	117.6(2)
C(35)-C(30)-C(31)	118.2(2)
C(35)-C(30)-C(27)	120.2(2)
C(31)-C(30)-C(27)	121.6(2)
C(32)-C(31)-C(30)	121.8(3)
C(32)-C(31)-H(31)	119.1
C(30)-C(31)-H(31)	119.1
C(31)-C(32)-C(33)	120.1(2)
C(31)-C(32)-H(32)	119.9
C(33)-C(32)-H(32)	119.9
O(7)-C(33)-C(32)	113.8(2)
O(7)-C(33)-C(34)	125.5(3)
C(32)-C(33)-C(34)	120.7(3)
C(33)-C(34)-C(35)	120.1(2)
C(33)-C(34)-H(34)	119.9
C(35)-C(34)-H(34)	119.9
C(34)-C(35)-C(30)	119.0(2)
C(34)-C(35)-C(36)	121.1(2)
C(30)-C(35)-C(36)	119.8(2)

C(29)-C(36)-C(35)	118.8(2)
C(29)-C(36)-C(18)	119.0(2)
C(35)-C(36)-C(18)	122.1(2)
O(7)-C(37)-H(37A)	109.5
O(7)-C(37)-H(37B)	109.5
H(37A)-C(37)-H(37B)	109.5
O(7)-C(37)-H(37C)	109.5
H(37A)-C(37)-H(37C)	109.5
H(37B)-C(37)-H(37C)	109.5
C(1)-N(1)-C(11)	119.1(2)
C(1)-N(1)-C(2)	115.6(2)
C(11)-N(1)-C(2)	116.1(2)
C(1)-N(2)-C(29)	117.8(2)
C(1)-N(2)-C(20)	116.0(2)
C(29)-N(2)-C(20)	114.9(2)
C(6)-O(2)-C(7)	104.3(2)
C(8)-O(3)-C(7)	104.5(2)
C(15)-O(4)-C(19)	117.5(2)
C(24)-O(5)-C(25)	106.1(3)
C(26)-O(6)-C(25)	105.8(3)
C(33)-O(7)-C(37)	117.2(2)

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8) **Table S7.** Anisotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for compound **1**. The anisotropic displacement factor exponent takes the form:  $-2\pi^2 [ h^2 a^{*2} U^{11} + \dots + 2 h k a^{*} b^{*} U^{12} ]$ .

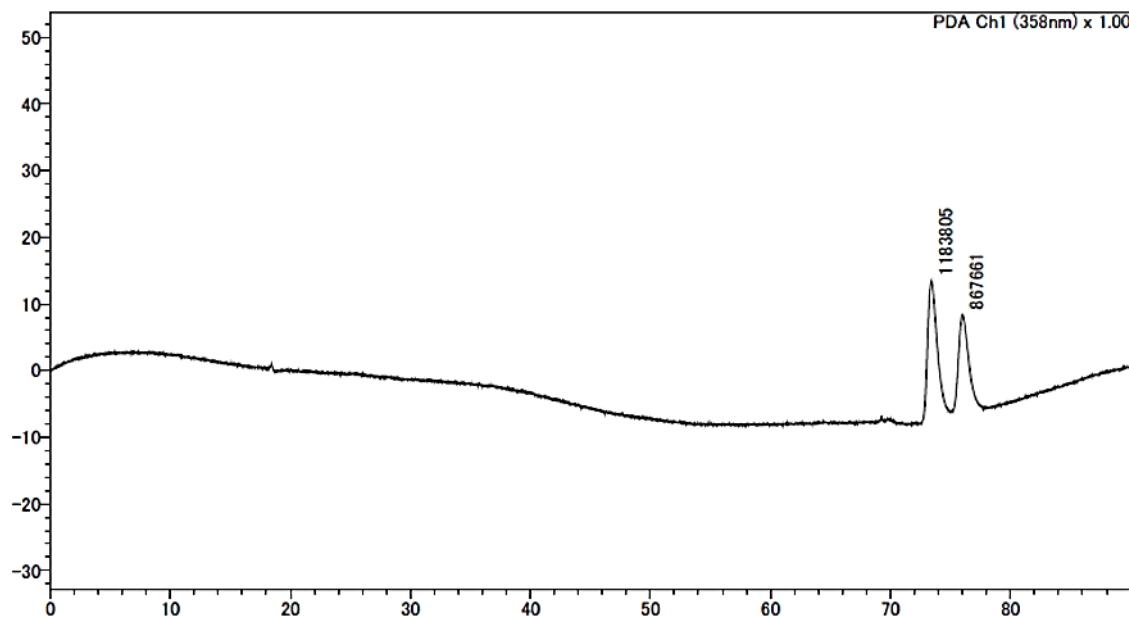
	$U^{11}$	$U^{22}$	$U^{33}$	$U^{23}$	$U^{13}$	$U^{12}$
C(1)	57(2)	36(1)	35(1)	-1(1)	-5(1)	11(1)
C(2)	68(2)	44(1)	6(1)	4(1)	7(1)	6(1)
C(3)	78(2)	47(2)	30(1)	10(1)	7(1)	3(1)
C(4)	55(2)	38(1)	36(1)	5(1)	9(1)	7(1)
C(5)	62(2)	43(1)	39(1)	10(1)	17(1)	4(1)
C(6)	46(2)	40(1)	51(2)	7(1)	13(1)	1(1)
C(7)	57(2)	42(2)	57(2)	2(1)	6(1)	-5(1)
C(8)	44(1)	39(1)	40(1)	1(1)	7(1)	5(1)
C(9)	41(1)	31(1)	35(1)	2(1)	8(1)	7(1)
C(10)	43(1)	34(1)	33(1)	4(1)	7(1)	10(1)
C(11)	44(1)	34(1)	29(1)	1(1)	3(1)	10(1)
C(12)	37(1)	32(1)	31(1)	2(1)	5(1)	7(1)
C(13)	43(1)	34(1)	36(1)	-1(1)	2(1)	0(1)
C(14)	47(1)	40(1)	31(1)	-3(1)	-3(1)	6(1)
C(15)	42(1)	36(1)	29(1)	3(1)	2(1)	8(1)
C(16)	38(1)	31(1)	32(1)	2(1)	4(1)	5(1)
C(17)	38(1)	31(1)	29(1)	3(1)	2(1)	8(1)
C(18)	38(1)	32(1)	29(1)	2(1)	3(1)	9(1)
C(19)	78(2)	45(2)	35(1)	12(1)	5(1)	-5(1)
C(20)	47(2)	62(2)	44(1)	-6(1)	-6(1)	18(1)
C(21)	48(2)	79(2)	48(2)	-6(2)	-9(1)	10(2)
C(22)	47(2)	66(2)	35(1)	-10(1)	1(1)	0(1)
C(23)	48(2)	88(2)	47(2)	-21(2)	-1(1)	-10(2)
C(24)	61(2)	66(2)	45(2)	-14(1)	7(2)	-19(2)
C(25)	100(3)	76(3)	68(2)	12(2)	-20(2)	-45(2)
C(26)	64(2)	52(2)	35(1)	-9(1)	11(1)	-13(1)
C(27)	51(2)	43(1)	27(1)	-9(1)	9(1)	-4(1)
C(28)	46(2)	49(2)	26(1)	-8(1)	5(1)	2(1)
C(29)	44(1)	39(1)	25(1)	-3(1)	4(1)	8(1)
C(30)	53(2)	33(1)	27(1)	-4(1)	9(1)	0(1)
C(31)	68(2)	30(1)	42(1)	0(1)	8(1)	-2(1)

C(32)	73(2)	31(1)	44(1)	4(1)	4(1)	9(1)
C(33)	52(2)	43(1)	32(1)	2(1)	4(1)	14(1)
C(34)	43(1)	35(1)	32(1)	-1(1)	5(1)	3(1)
C(35)	46(1)	34(1)	22(1)	-2(1)	6(1)	7(1)
C(36)	42(1)	34(1)	23(1)	-2(1)	3(1)	6(1)
C(37)	60(2)	54(2)	56(2)	10(1)	8(2)	13(1)
N(1)	53(1)	37(1)	26(1)	2(1)	1(1)	5(1)
N(2)	45(1)	44(1)	33(1)	-1(1)	-2(1)	12(1)
O(1)	70(1)	70(1)	42(1)	15(1)	-14(1)	14(1)
O(2)	63(1)	49(1)	56(1)	10(1)	9(1)	-13(1)
O(3)	56(1)	44(1)	47(1)	4(1)	2(1)	-13(1)
O(4)	58(1)	46(1)	28(1)	6(1)	1(1)	0(1)
O(5)	74(2)	79(2)	85(2)	-20(1)	4(1)	-37(1)
O(6)	80(2)	49(1)	54(1)	-5(1)	6(1)	-24(1)
O(7)	59(1)	47(1)	63(1)	5(1)	-4(1)	11(1)

9) **Table S8.** Hydrogen coordinates ( $x \times 10^4$ ) and isotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for compound **1**.

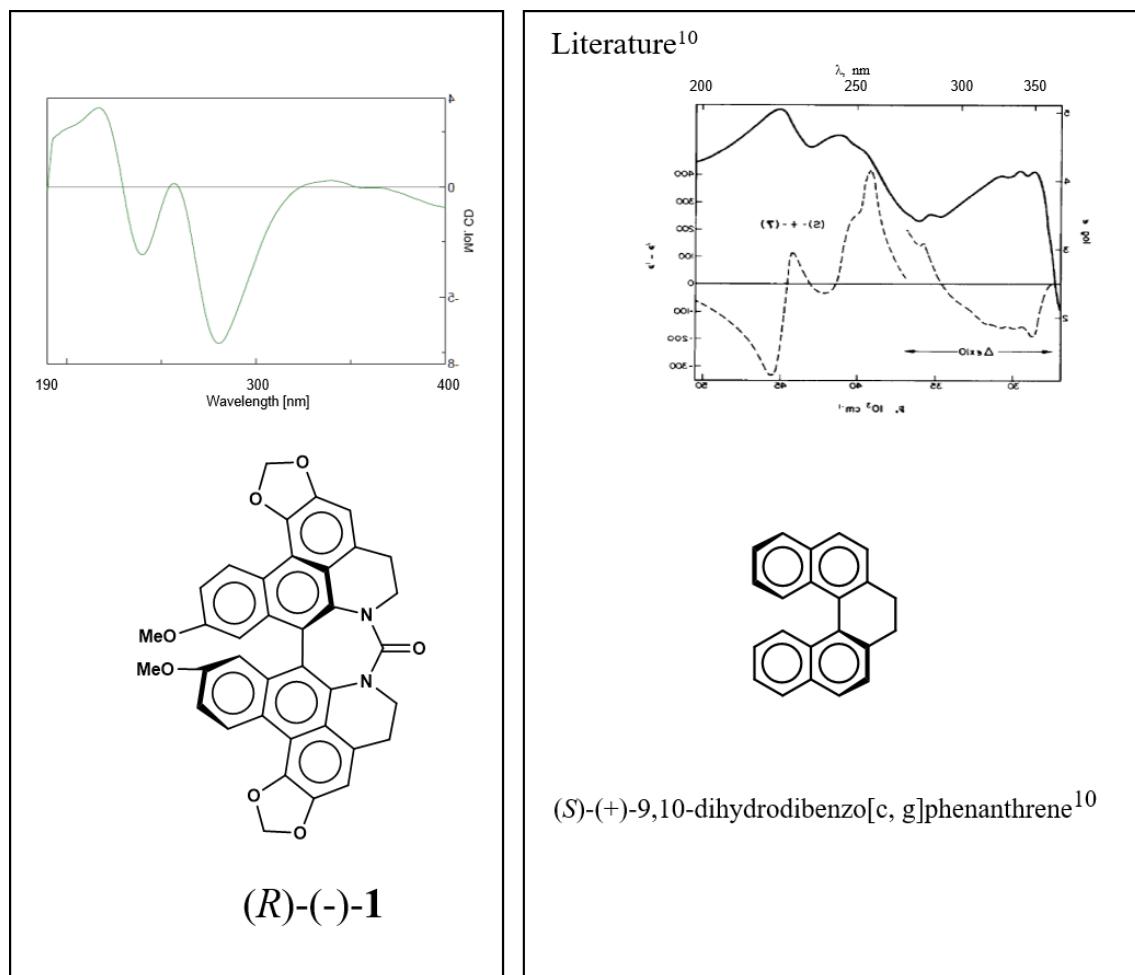
	x	y	z	U(eq)
H(2A)	9560	3817	1593	55
H(2B)	11278	3679	2061	55
H(3A)	11298	4820	1597	62
H(3B)	9594	4983	1973	62
H(5)	12695	5759	2337	57
H(7A)	13769	6739	4673	63
H(7B)	15575	6427	4646	63
H(13)	13011	5052	5766	45
H(14)	12367	4487	6912	47
H(16)	9490	3142	5642	40
H(19A)	8445	2997	6849	79
H(19B)	9428	2717	7627	79
H(19C)	10008	2512	6771	79
H(20A)	4895	4135	3048	62
H(20B)	4812	3757	3888	62
H(21A)	3088	3142	3011	70
H(21B)	4447	3098	2371	70
H(23)	2803	1866	2963	73
H(25A)	5092	-222	3493	99
H(25B)	4250	-151	4322	99
H(31)	8582	646	4696	56
H(32)	11201	569	5227	59
H(34)	11948	2603	4764	44
H(37A)	14541	2242	5046	85
H(37B)	15312	1885	5835	85
H(37C)	13747	2375	5874	85

10) **Figure S2.** Chiral HPLC analysis of isolated cerasoidine (**1**).

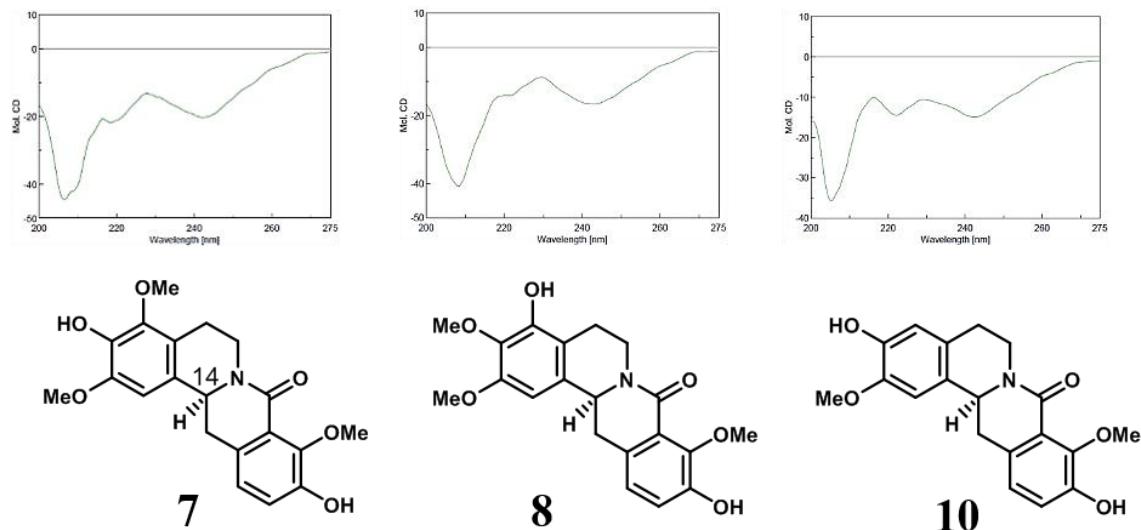


An isolated sample of **1** was subjected to a Chiral HPLC analysis [CHIRALPACK IB, φ 4.6 × 250 mm, hexane/CHCl<sub>3</sub> [100/0–75/25 (linear gradient, 0–30 min), 75/25 (30–90 min)], flow rate: 0.2 mL/min, detector wavelength 358 nm]. Peaks with retention times of 73 min and 76 min were assigned as the (R)-(-)- and (S)-(+) -enantiomers at a ratio of 57:43.

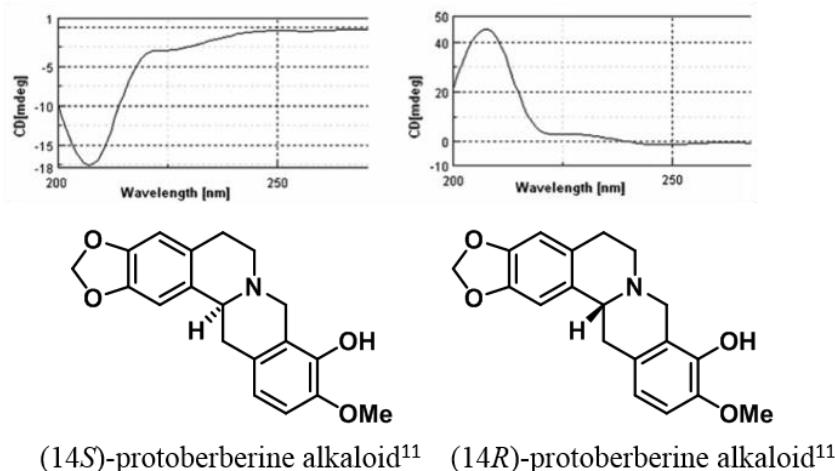
11) **Figure S3.** Comparison of the CD spectrum of cerasoidine (**1**) with that of (*S*)-(+)-9,10-dihydrodibenzo[c,g]phenanthrene<sup>10</sup>.



12) **Figure S4.** Comparison of CD spectra of compounds **7**, **8**, and **10** with those of (*S*)- and (*R*)-protoberberine alkaloids in the literature<sup>11</sup>.

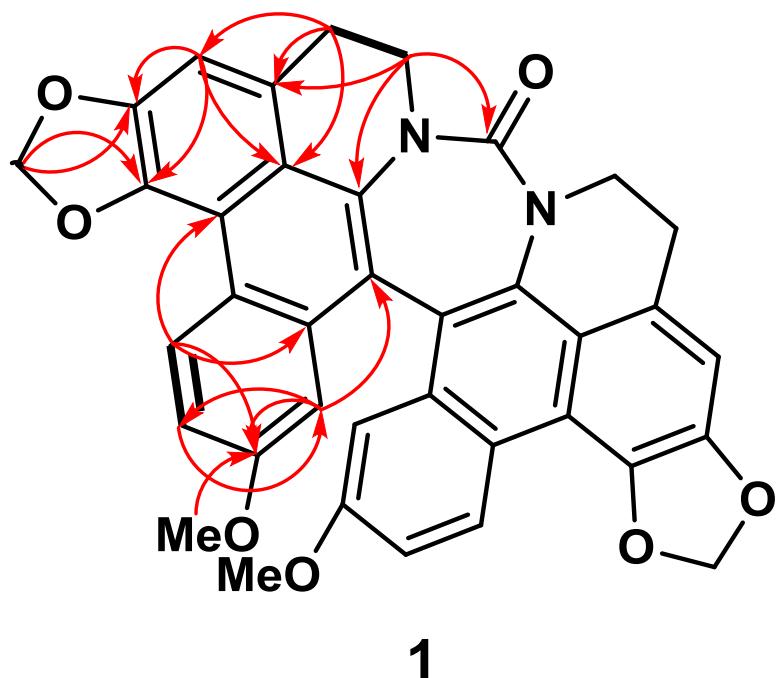


#### Literature<sup>11</sup>



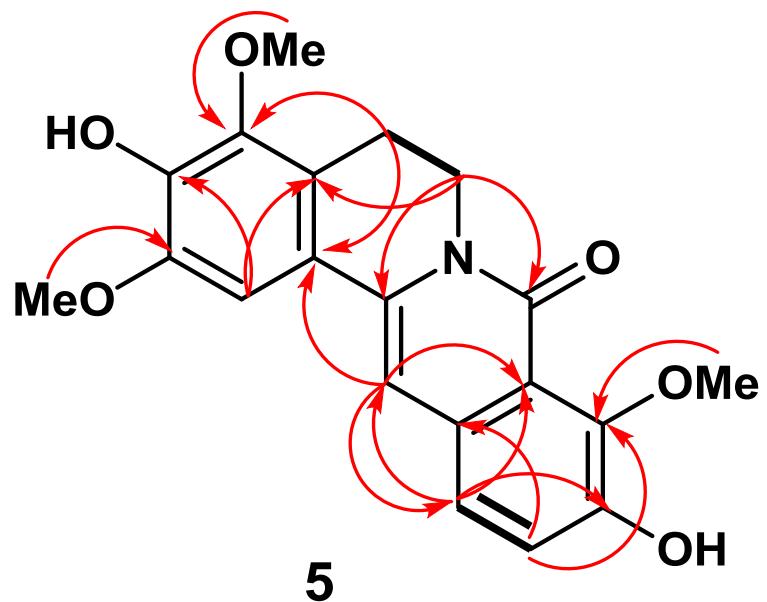
13) **Figure S5.** Key COSY and HMBC correlations for new compounds (**1**, **5**, **6**, **7**, and **8**).

**Fig S5 (A).** HMBC (arrow curves) and COSY (bold lines) correlations of **1**.

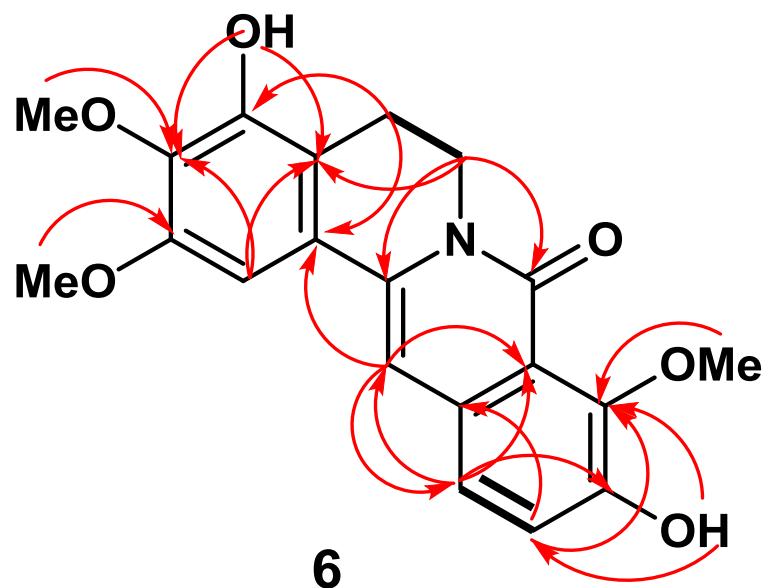


**1**

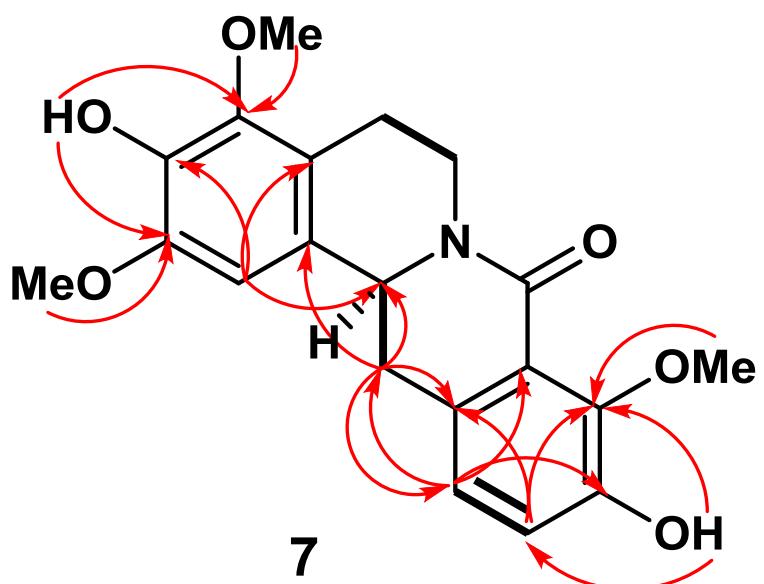
**Fig S5 (B).** HMBC (arrow curves) and COSY (bold lines) correlations of **5**.



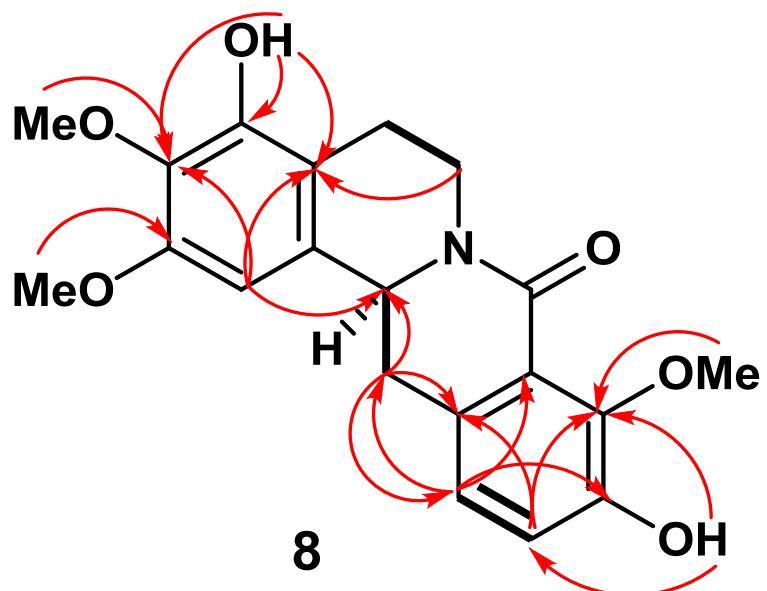
**Fig S5 (C).** HMBC (arrow curves) and COSY (bold lines) correlations of **6**.



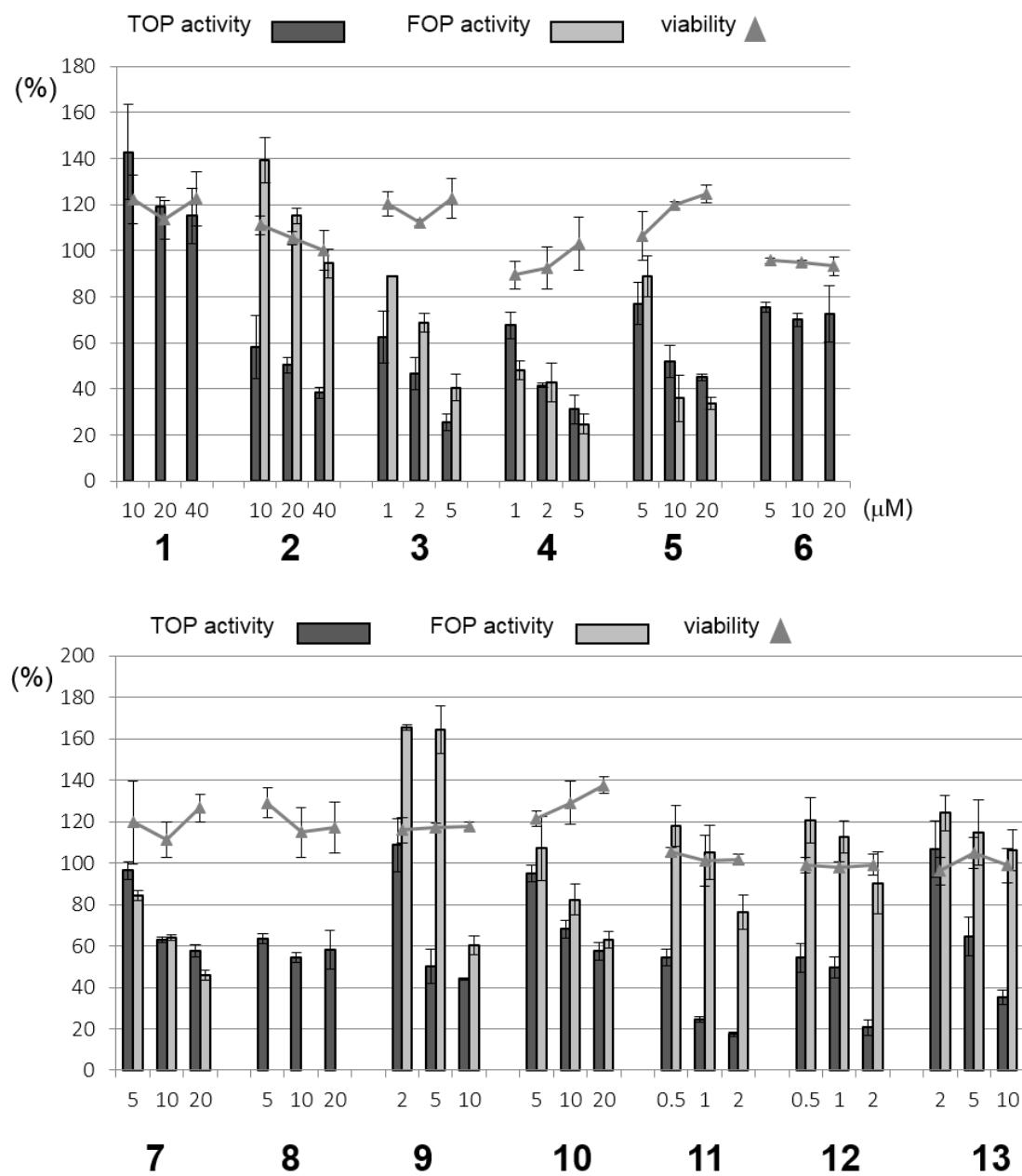
**Fig S5 (D).** HMBC (arrow curves) and COSY (bold lines) correlations of **7**.



**Fig S5 (E).** HMBC (arrow curves) and COSY (bold lines) correlations of **8**.



14) **Figure S6.** Inhibition of TCF/β-catenin transcriptional activities of compounds 1-13.



15) **Figure S7.** Spectroscopic data of compound **1**.

**Fig. S7(A)**  $^1\text{H}$  NMR spectrum of compound **1** (600 MHz,  $\text{CDCl}_3$ ).

**Fig. S7(B)**  $^{13}\text{C}$  NMR spectrum of compound **1** (100 MHz,  $\text{CDCl}_3$ ).

**Fig. S7(C)** COSY spectrum of compound **1** (600 MHz,  $\text{CDCl}_3$ ).

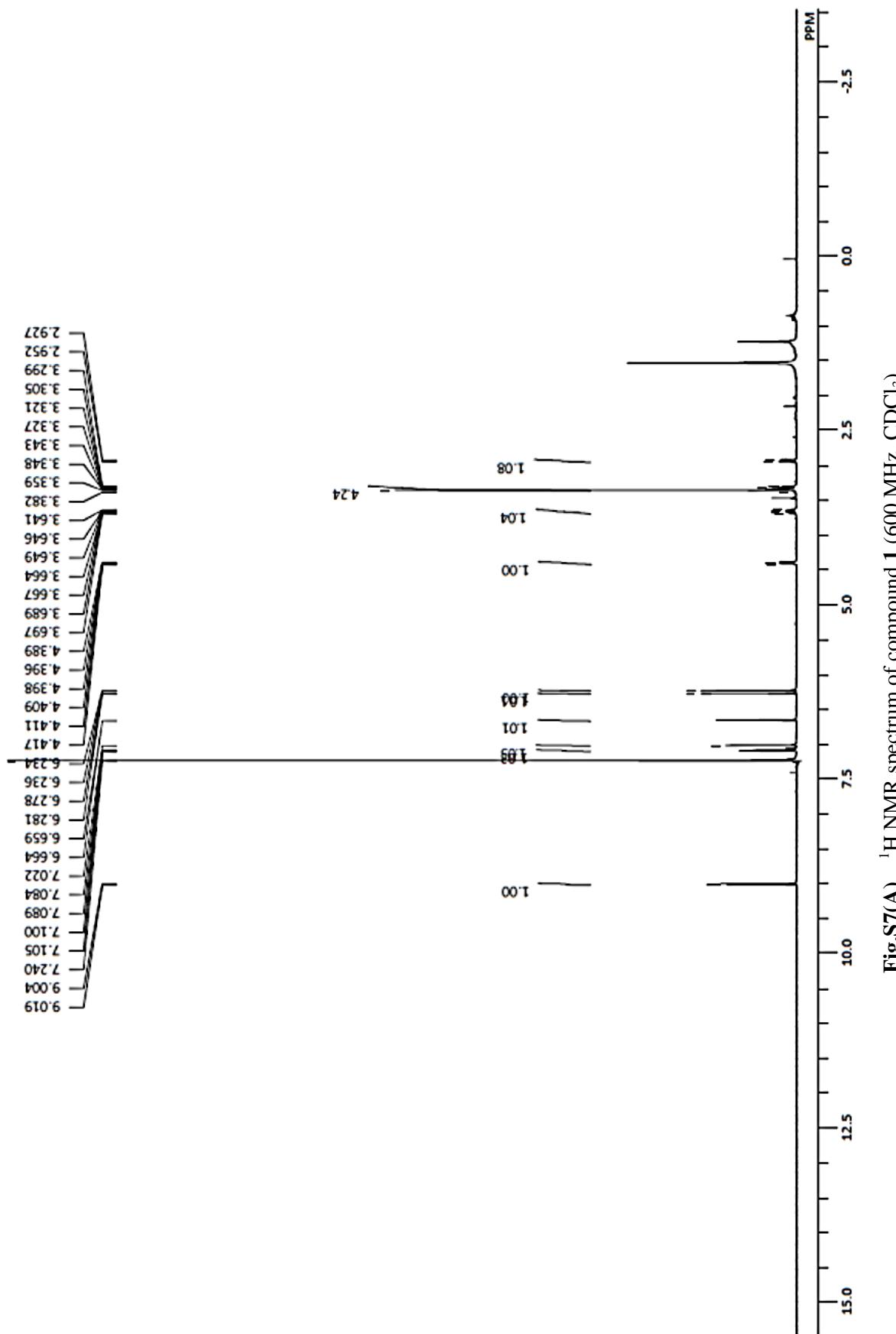
**Fig. S7(D)** HMQC spectrum of compound **1** (600 MHz,  $\text{CDCl}_3$ ).

**Fig. S7(E)** HMBC spectrum of compound **1** (600 MHz,  $\text{CDCl}_3$ ).

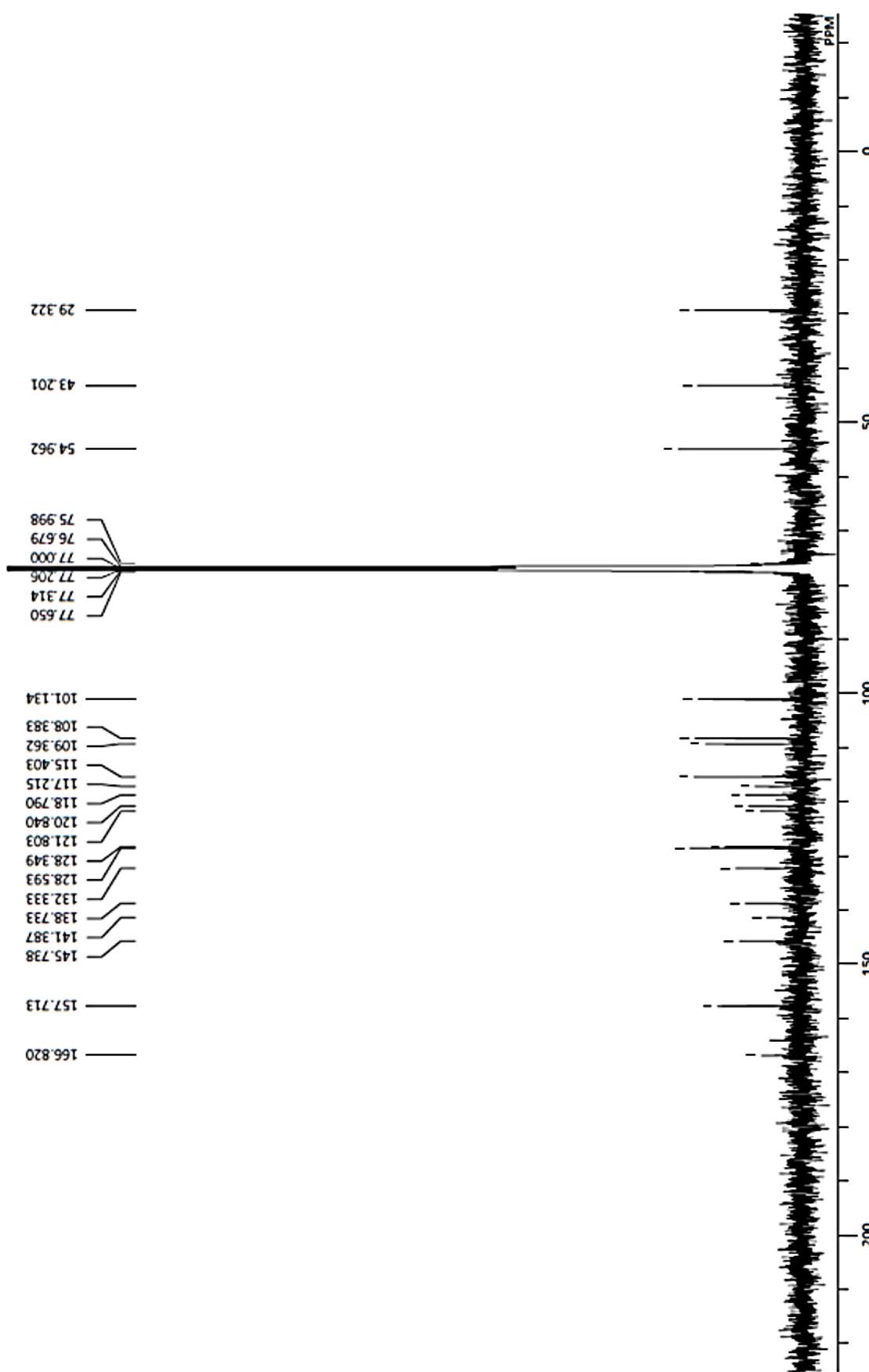
**Fig. S7(F)** HRESIMS spectrum of compound **1**.

**Fig. S7(G)** IR spectrum of compound **1**.

**Fig. S7(H)** CD spectrum of compound **1**



**Fig.S7(A)**  $^1\text{H}$  NMR spectrum of compound 1 (600 MHz,  $\text{CDCl}_3$ ).



**Fig. S7(B)**  $^{13}\text{C}$  NMR spectrum of compound 1 (100 MHz,  $\text{CDCl}_3$ ).

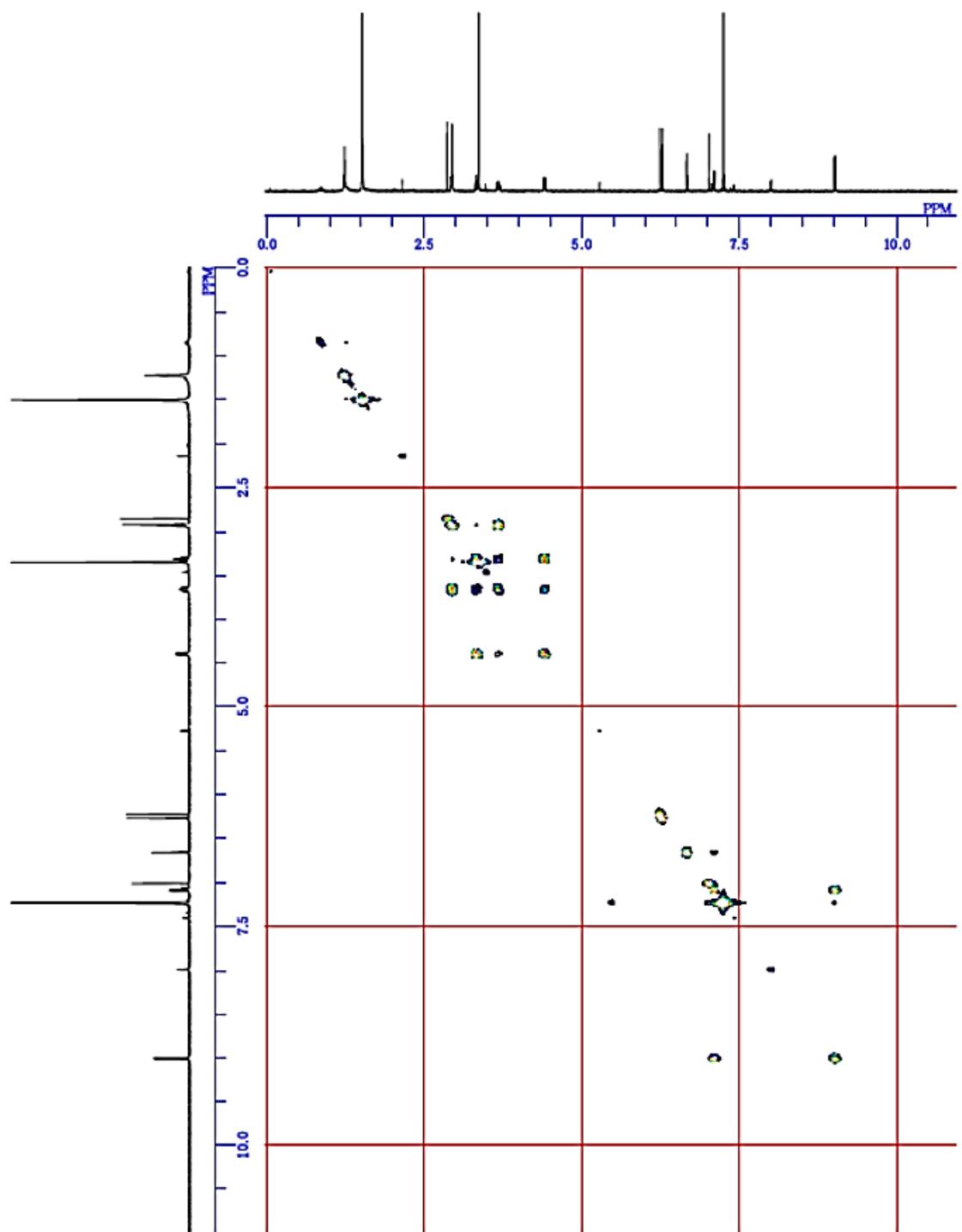


Fig. S7(C) COSY spectrum of compound 1 (600 MHz,  $\text{CDCl}_3$ ).

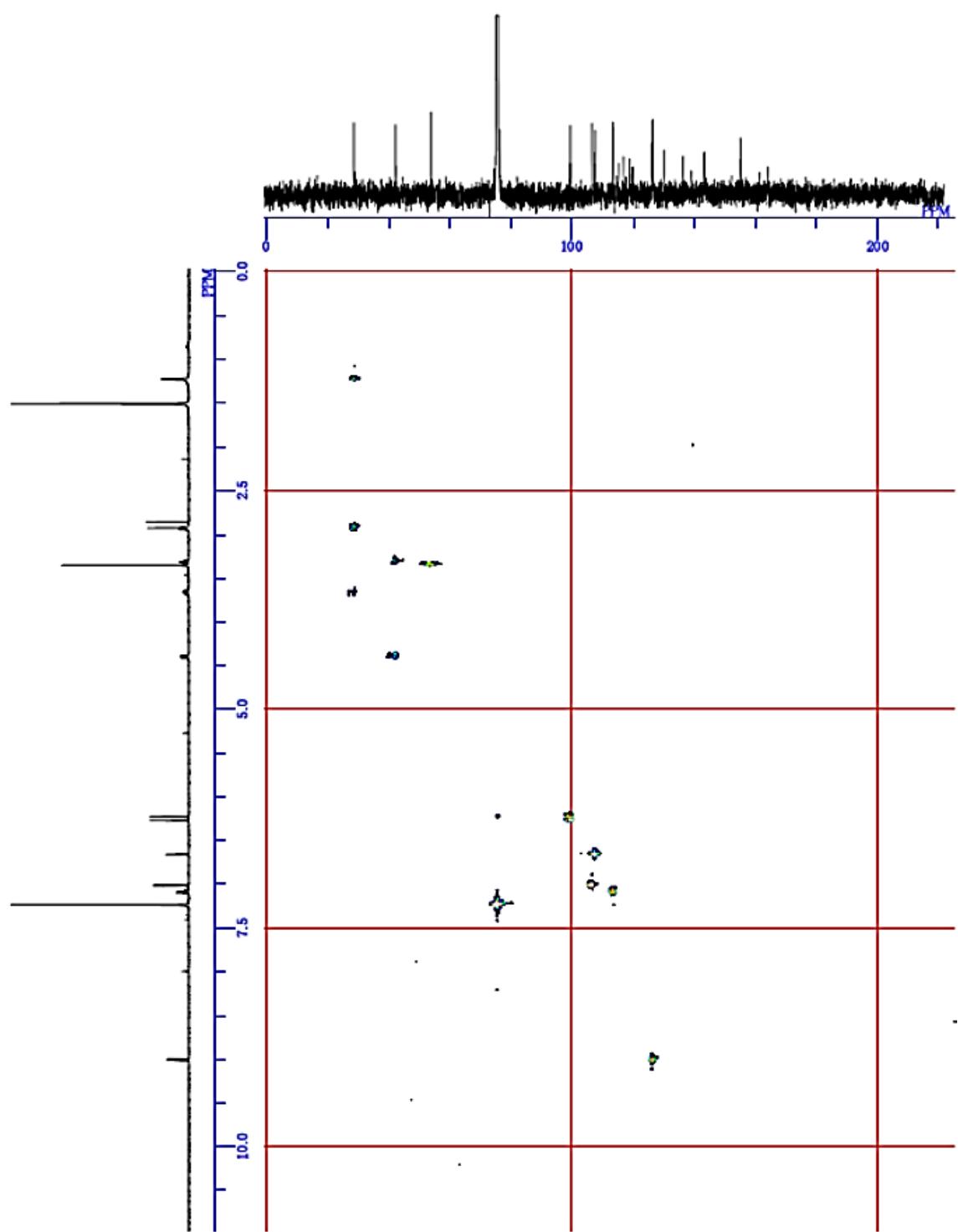
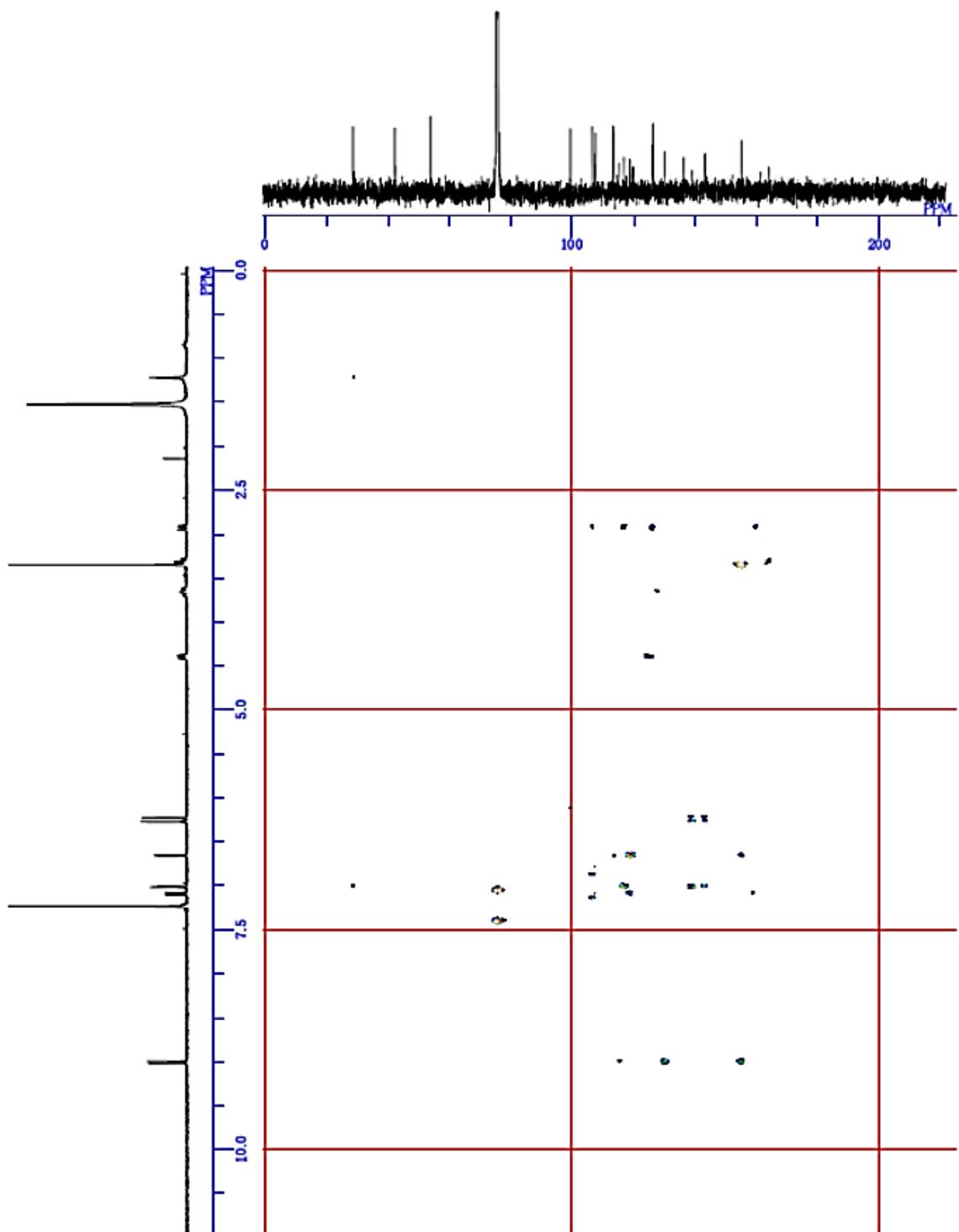
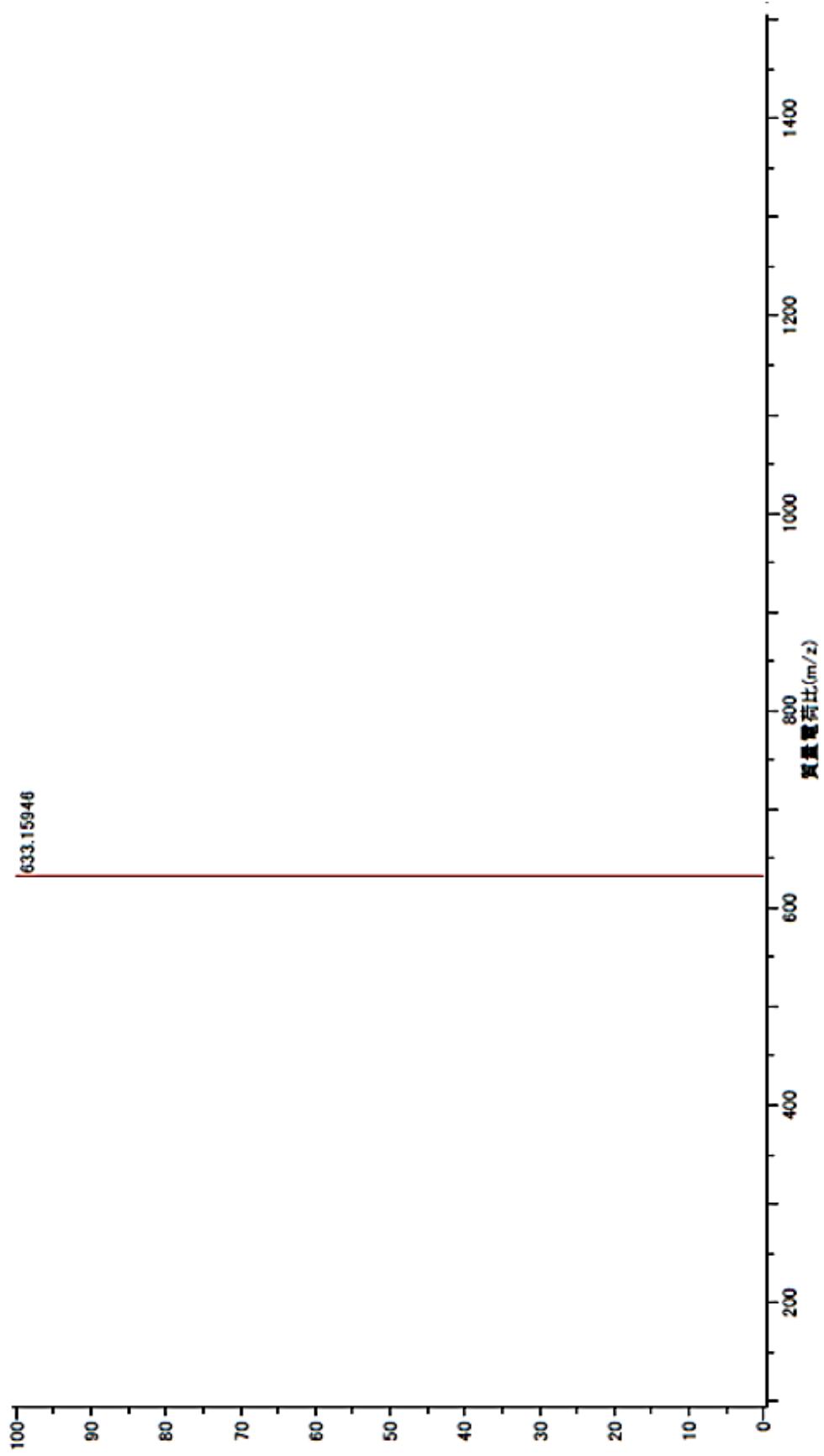


Fig. S7(D) HMQC spectrum of compound **1** (600 MHz,  $\text{CDCl}_3$ ).



**Fig. S7(E)** HMBC spectrum of compound **1** (600 MHz,  $\text{CDCl}_3$ ).



**Fig. S7(F)** HRESIMS spectrum of compound 1.

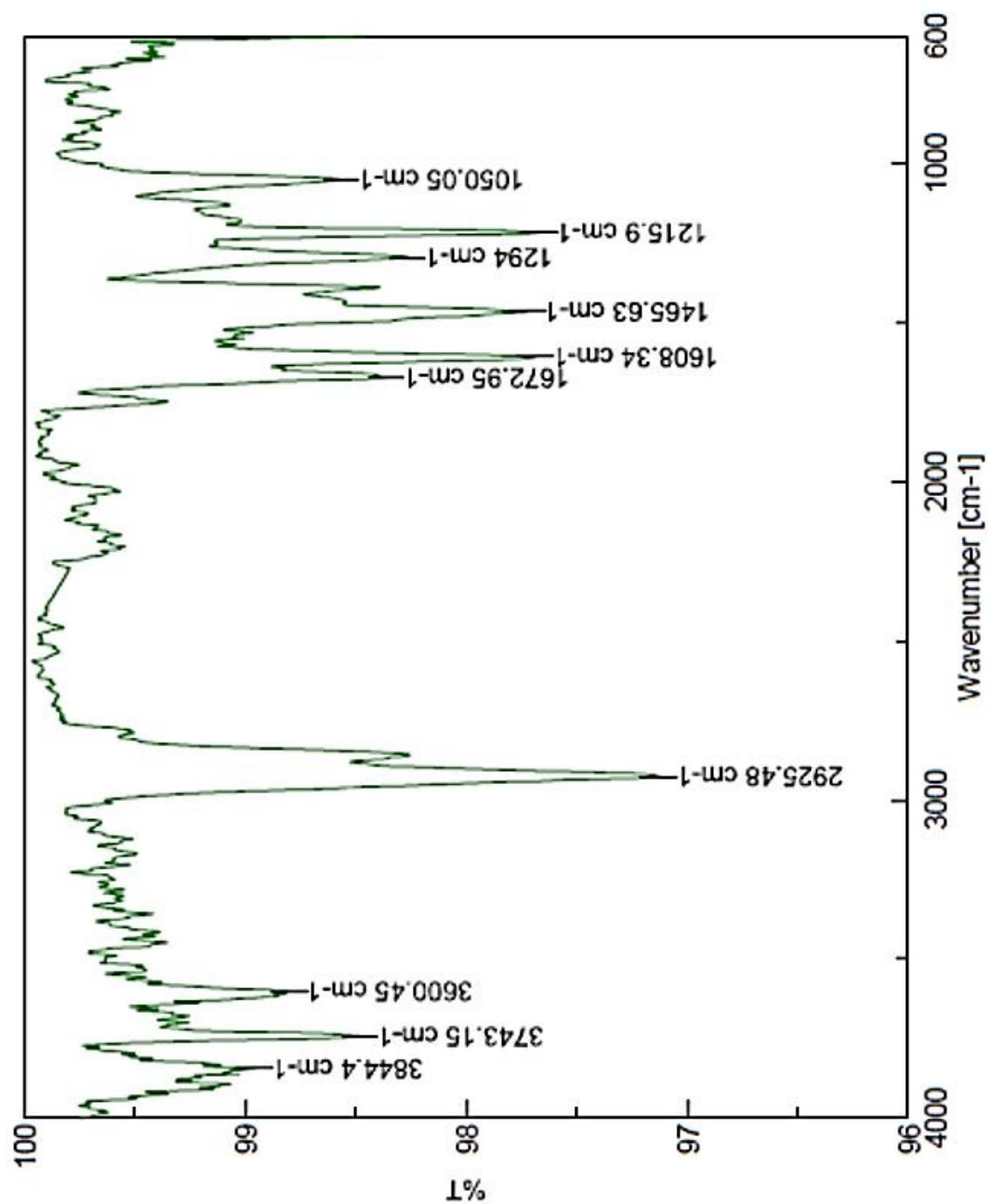


Fig. S7(G) IR spectrum of compound 1.

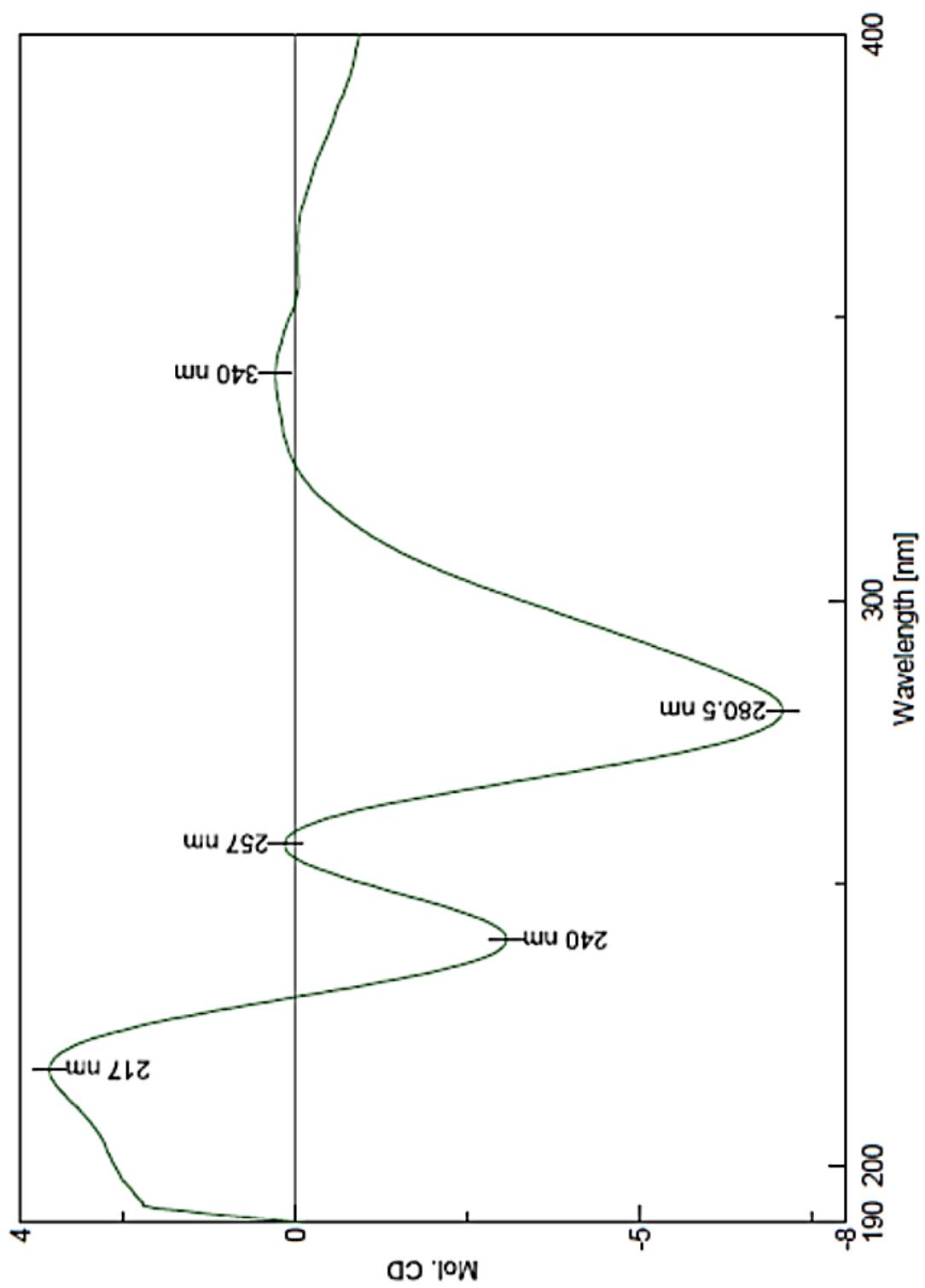


Fig. S7(H) CD spectrum of compound 1.

16) **Figure S8.** Spectroscopic data of compound **5**.

**Fig. S8(A)**  $^1\text{H}$  NMR spectrum of compound **5** (600 MHz, DMSO- $d_6$ ).

**Fig. S8(B)**  $^{13}\text{C}$  NMR spectrum of compound **5** (150 MHz, DMSO- $d_6$ ).

**Fig. S8(C)** COSY spectrum of compound **5** (600 MHz, DMSO- $d_6$ ).

**Fig. S8(D)** HMQC spectrum of compound **5** (600 MHz, DMSO- $d_6$ ).

**Fig. S8(E)** HMBC spectrum of compound **5** (600 MHz, DMSO- $d_6$ ).

**Fig. S8(F)** HRESIMS spectrum of compound **5**.

**Fig. S8(G)** IR spectrum of compound **5**.

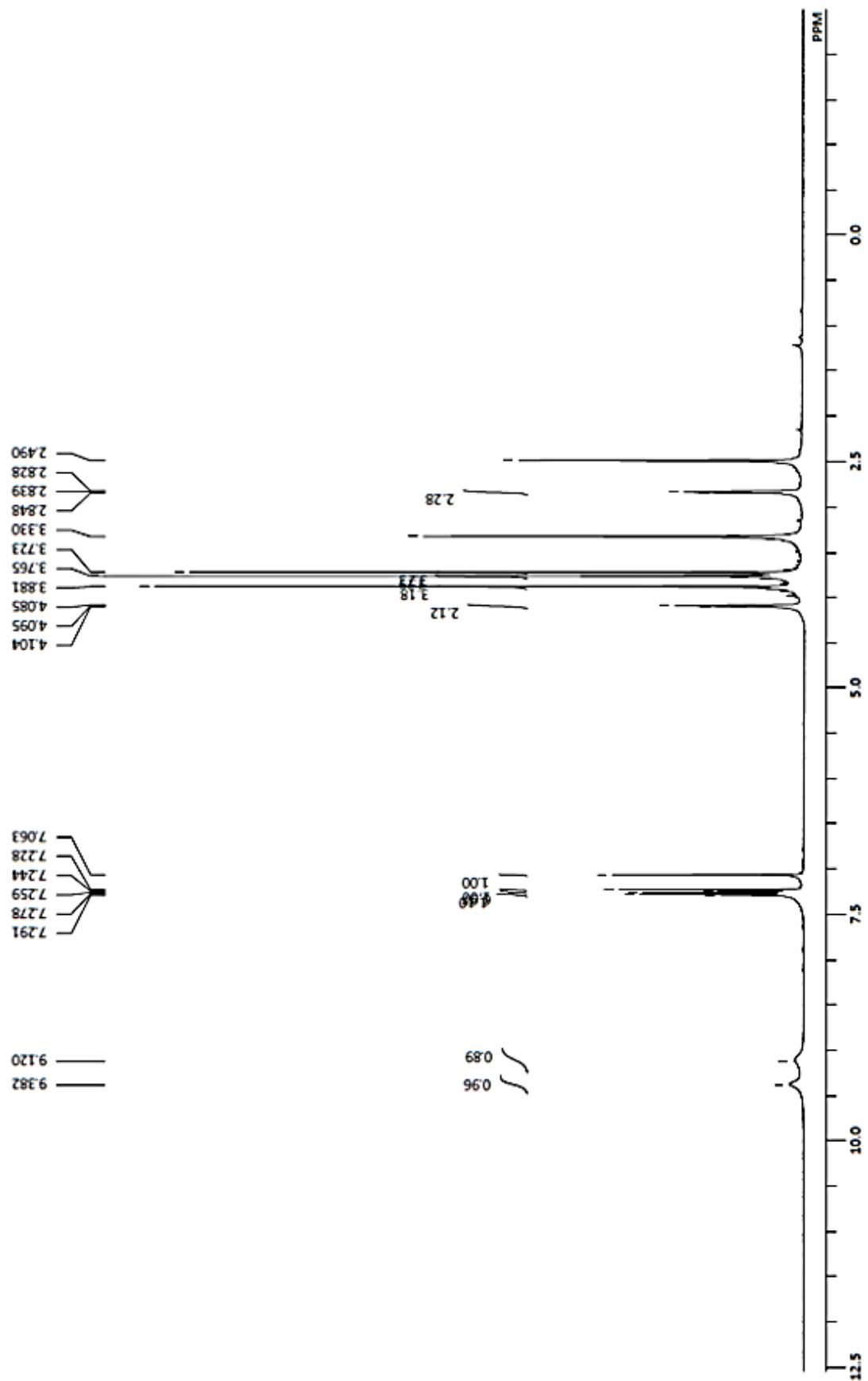
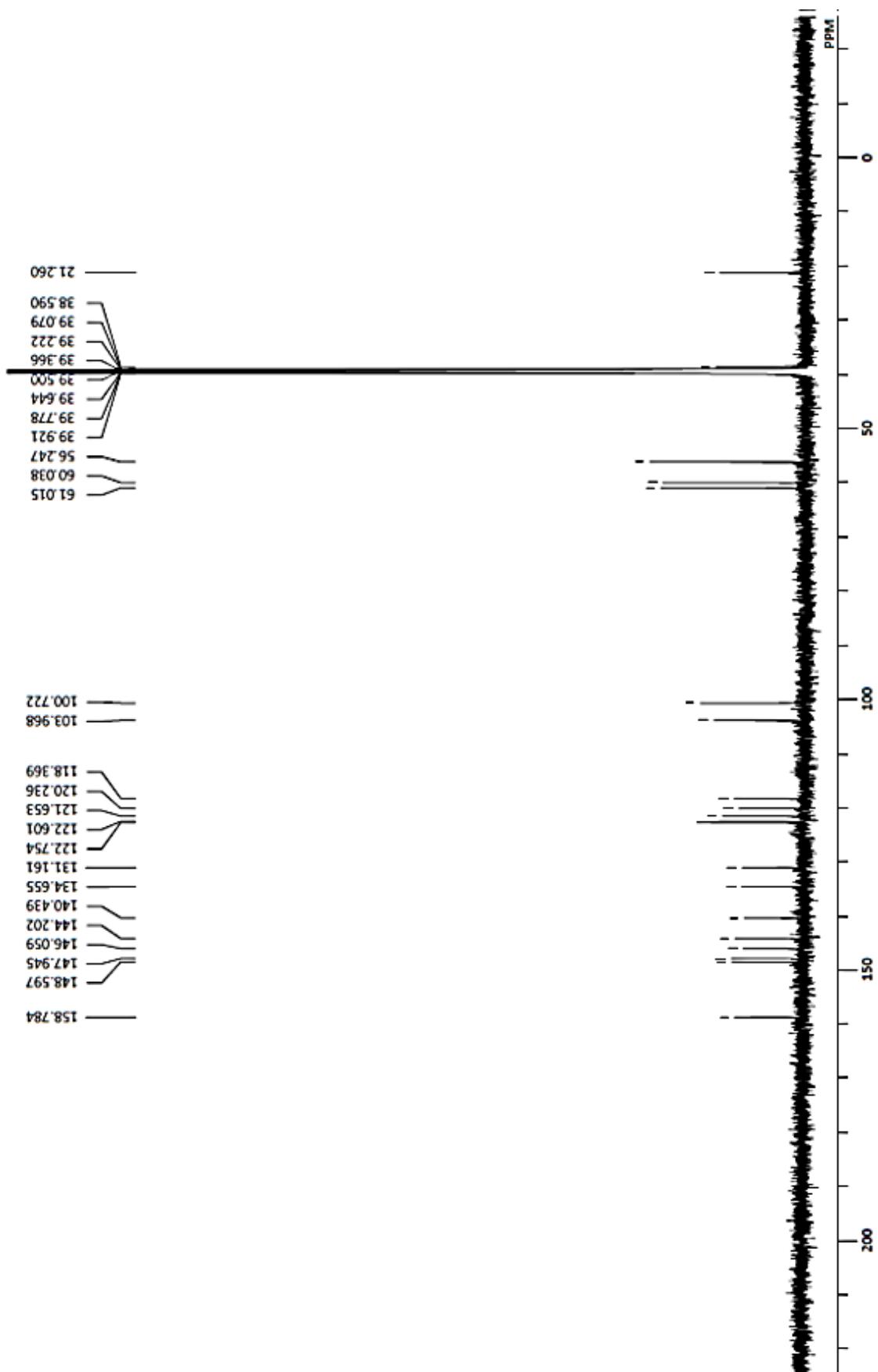


Fig. S8(A)  $^1\text{H}$  NMR spectrum of compound 5 (600 MHz,  $\text{DMSO}-d_6$ ).



**Fig. S8(B)**  $^{13}\text{C}$  NMR spectrum of compound 5 (150 MHz,  $\text{DMSO}-d_6$ ).

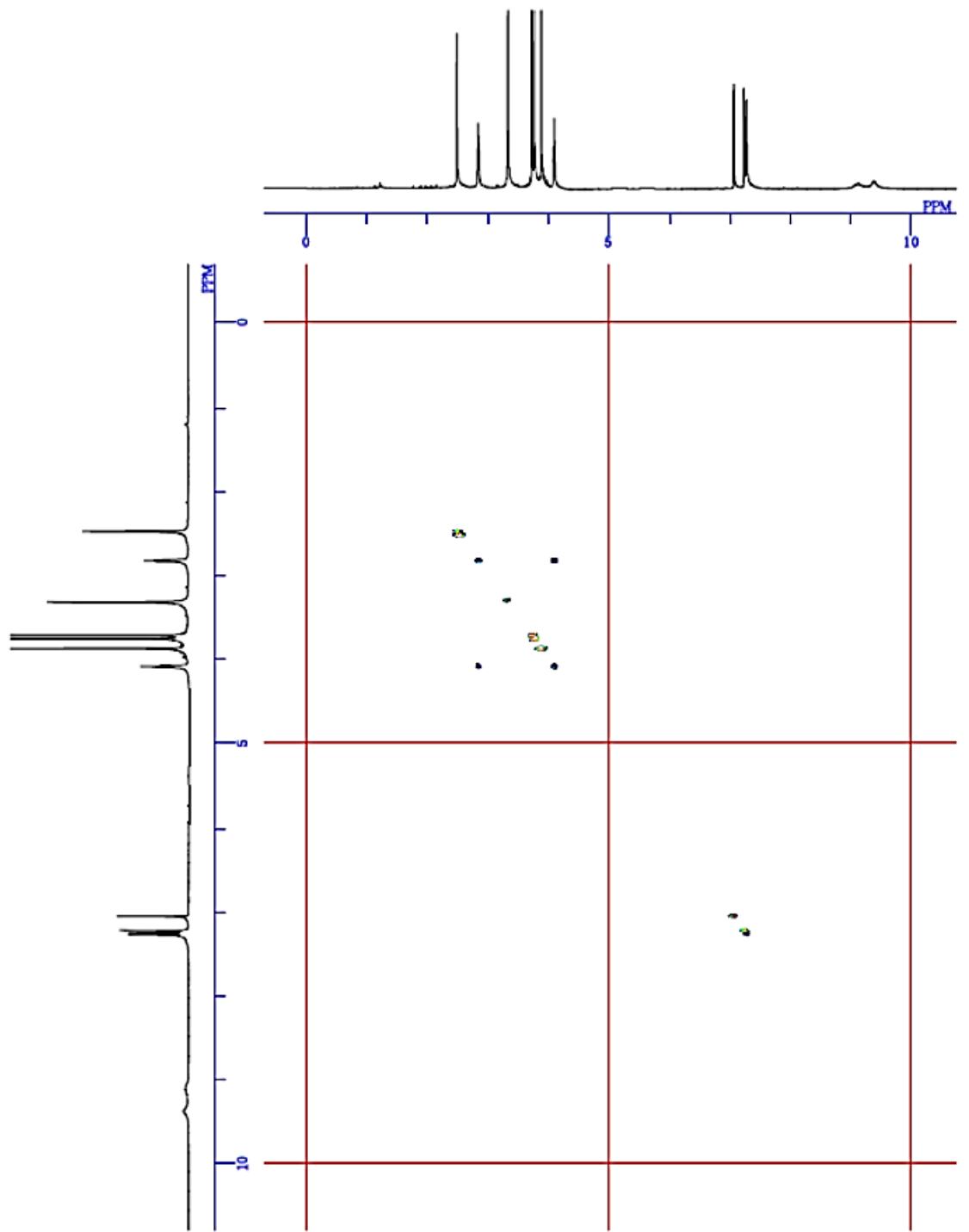


Fig. S8(C) COSY spectrum of compound 5 (600 MHz, DMSO-*d*<sub>6</sub>).

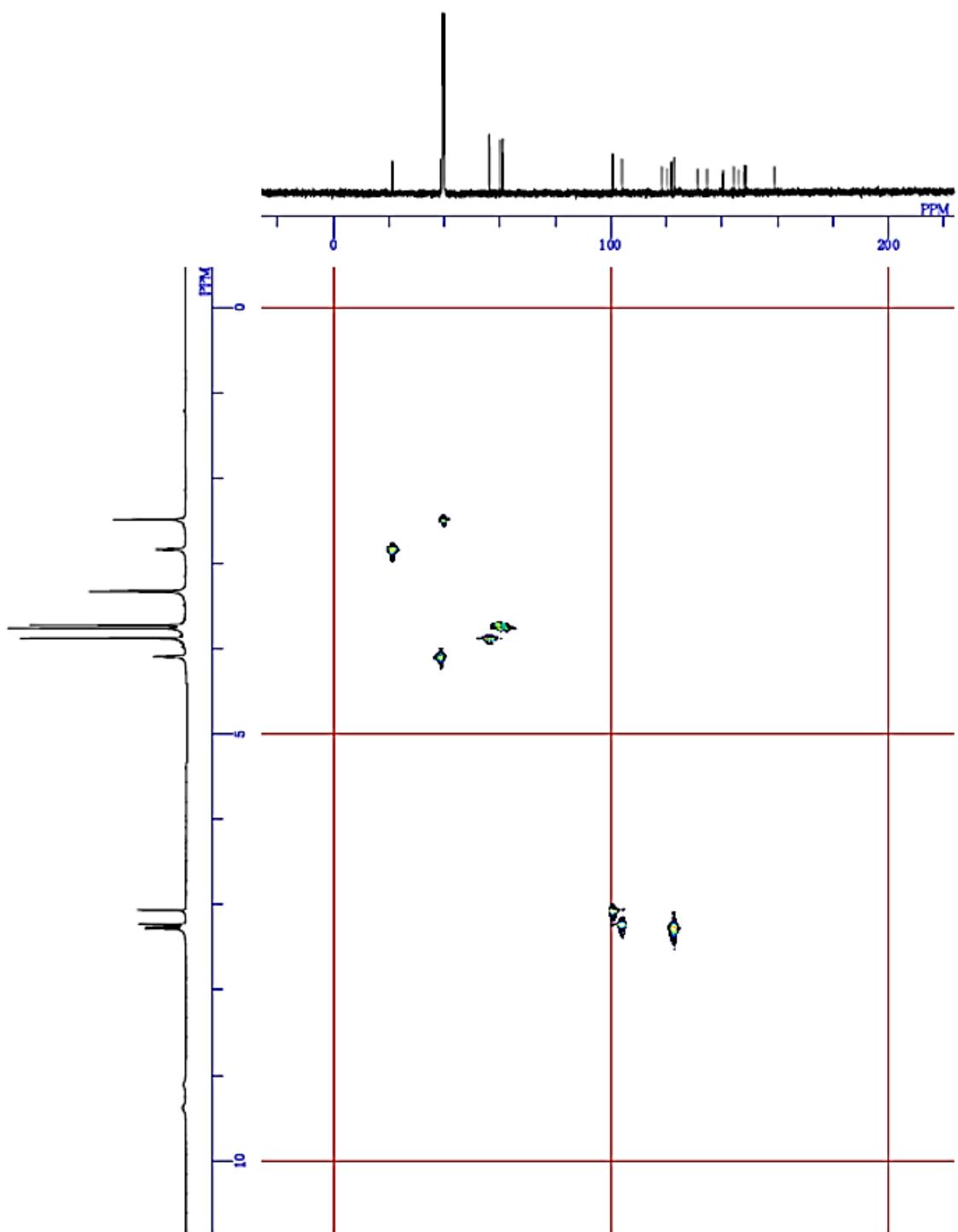
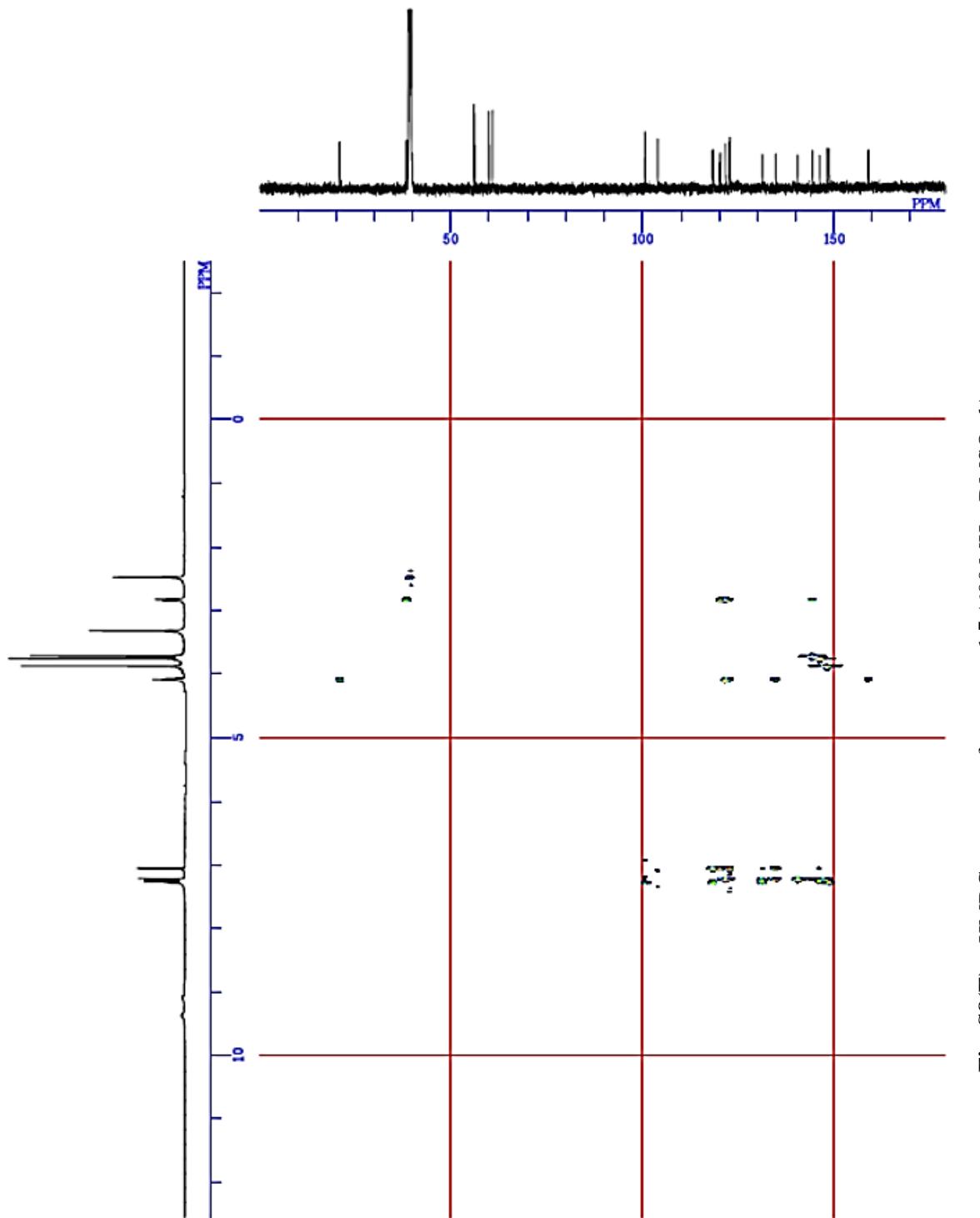


Fig. S8(D) HMQC spectrum of compound 5 (600 MHz, DMSO-*d*<sub>6</sub>).



**Fig. S8(E)** HMBC spectrum of compound **5** (600 MHz,  $\text{DMSO}-d_6$ ).

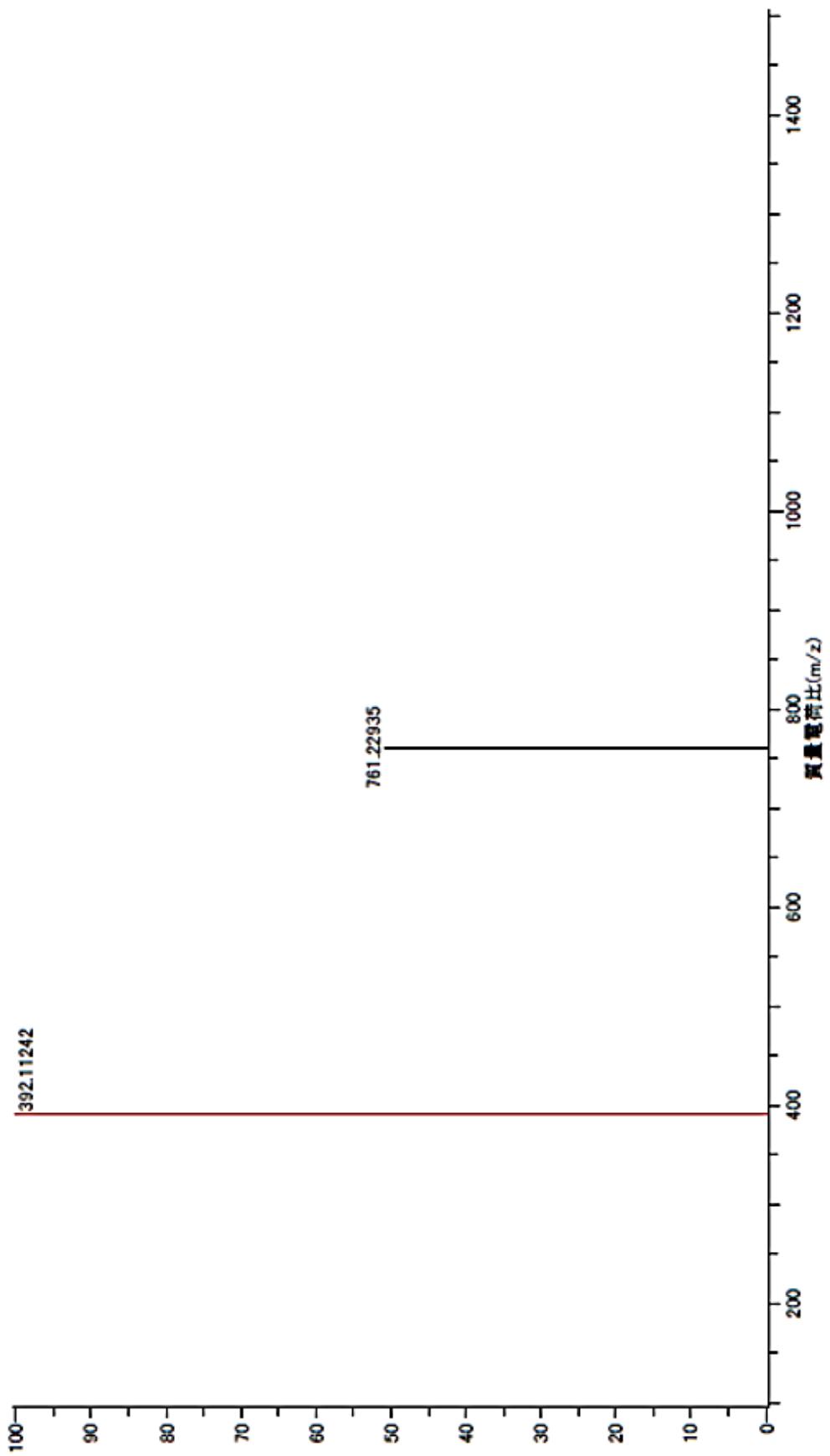


Fig.S8(F) HRESIMS spectrum of compound 5.

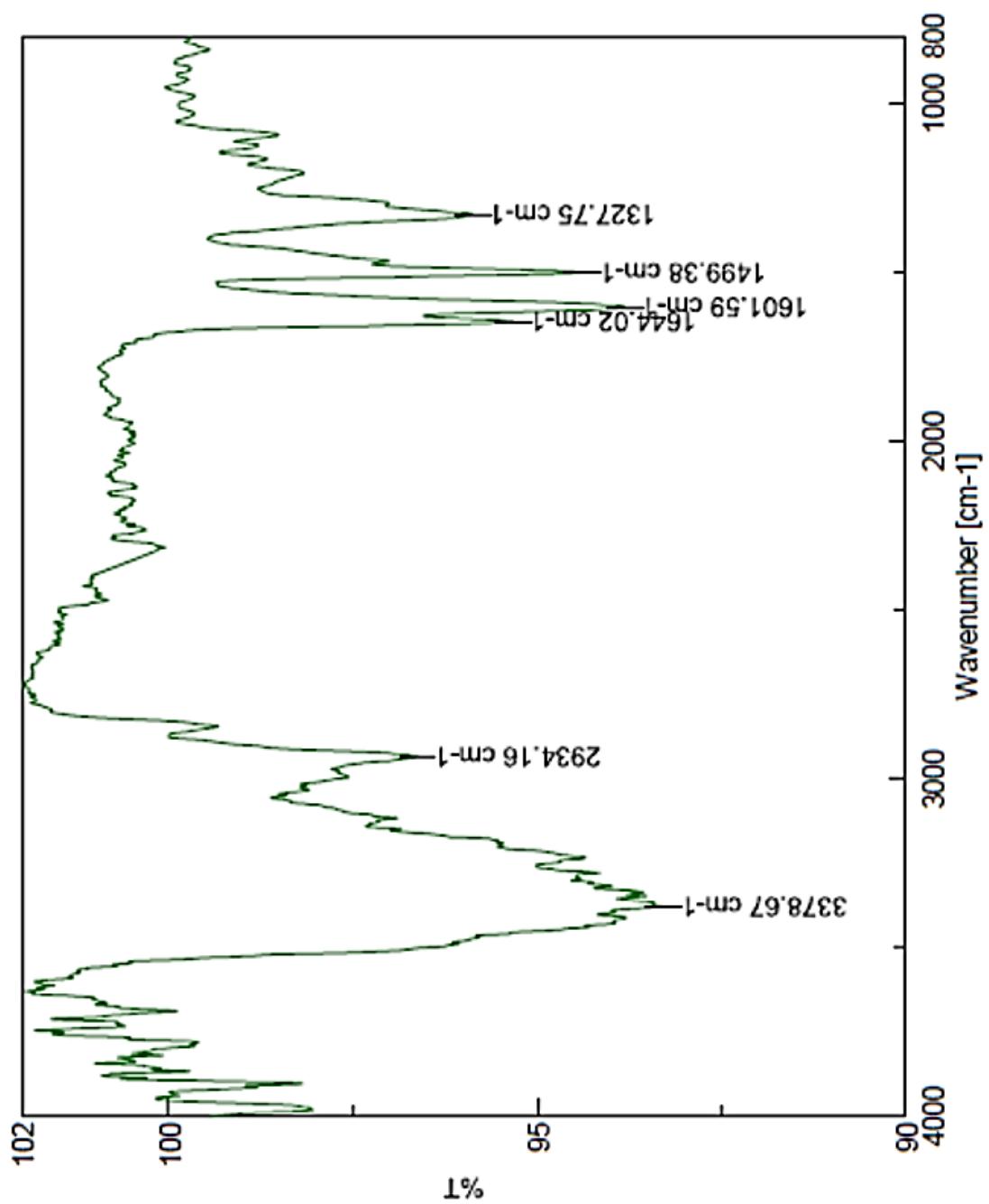


Fig. S8(G) IR spectrum of compound 5.

17) **Figure S9.** Spectroscopic data of compound **6**.

**Fig. S9(A)**  $^1\text{H}$  NMR spectrum of compound **6** (400 MHz, DMSO- $d_6$ ).

**Fig. S9(B)**  $^{13}\text{C}$  NMR spectrum of compound **6** (100 MHz, DMSO- $d_6$ ).

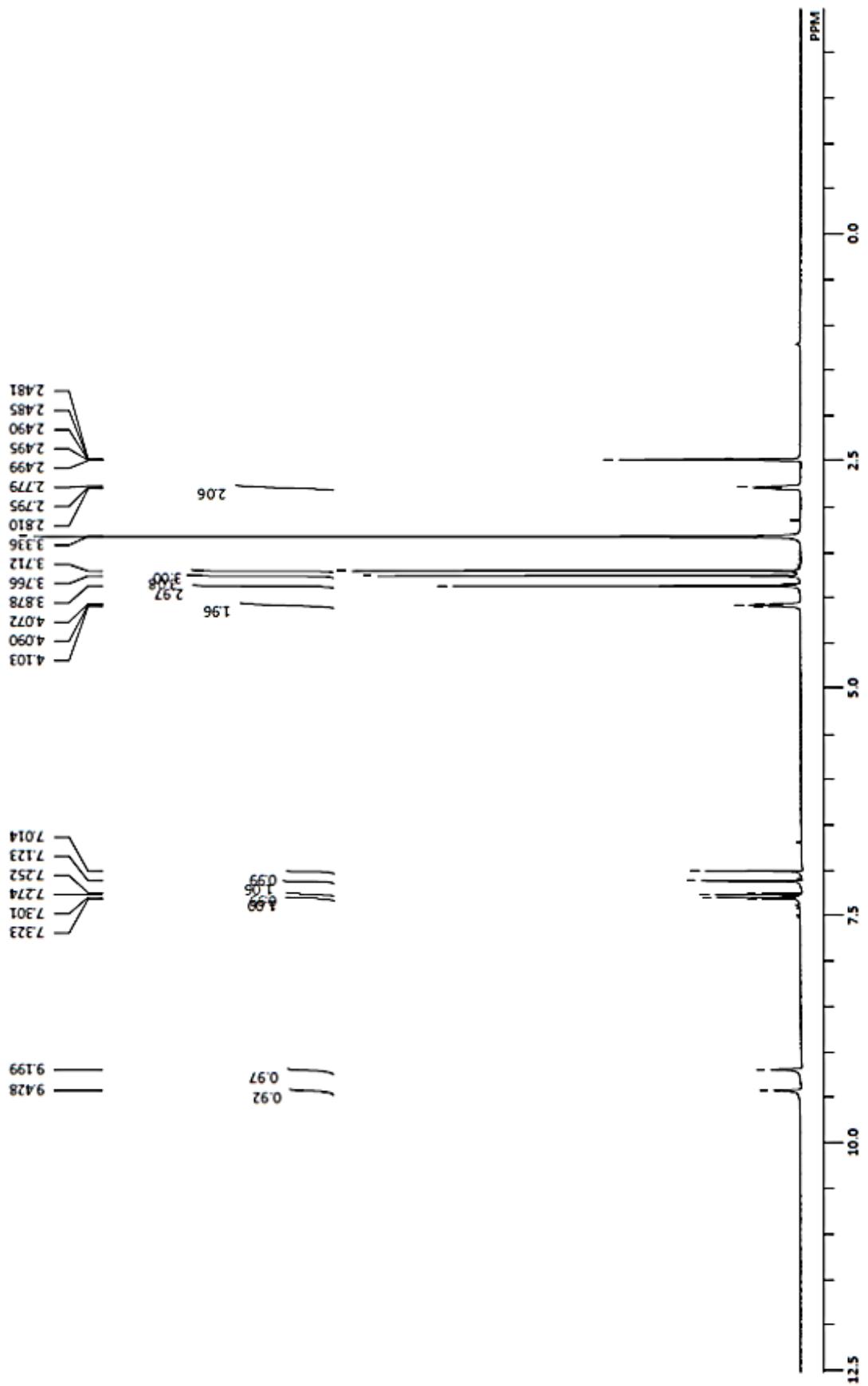
**Fig. S9(C)** COSY spectrum of compound **6** (400 MHz, DMSO- $d_6$ ).

**Fig. S9(D)** HMQC spectrum of compound **6** (400 MHz, DMSO- $d_6$ ).

**Fig. S9(E)** HMBC spectrum of compound **6** (400 MHz, DMSO- $d_6$ ).

**Fig. S9(F)** HRESIMS spectrum of compound **6**.

**Fig. S9(G)** IR spectrum of compound **6**.



**Fig. S9(A)** <sup>1</sup>H NMR spectrum of compound **6** (400 MHz, DMSO-*d*<sub>6</sub>).

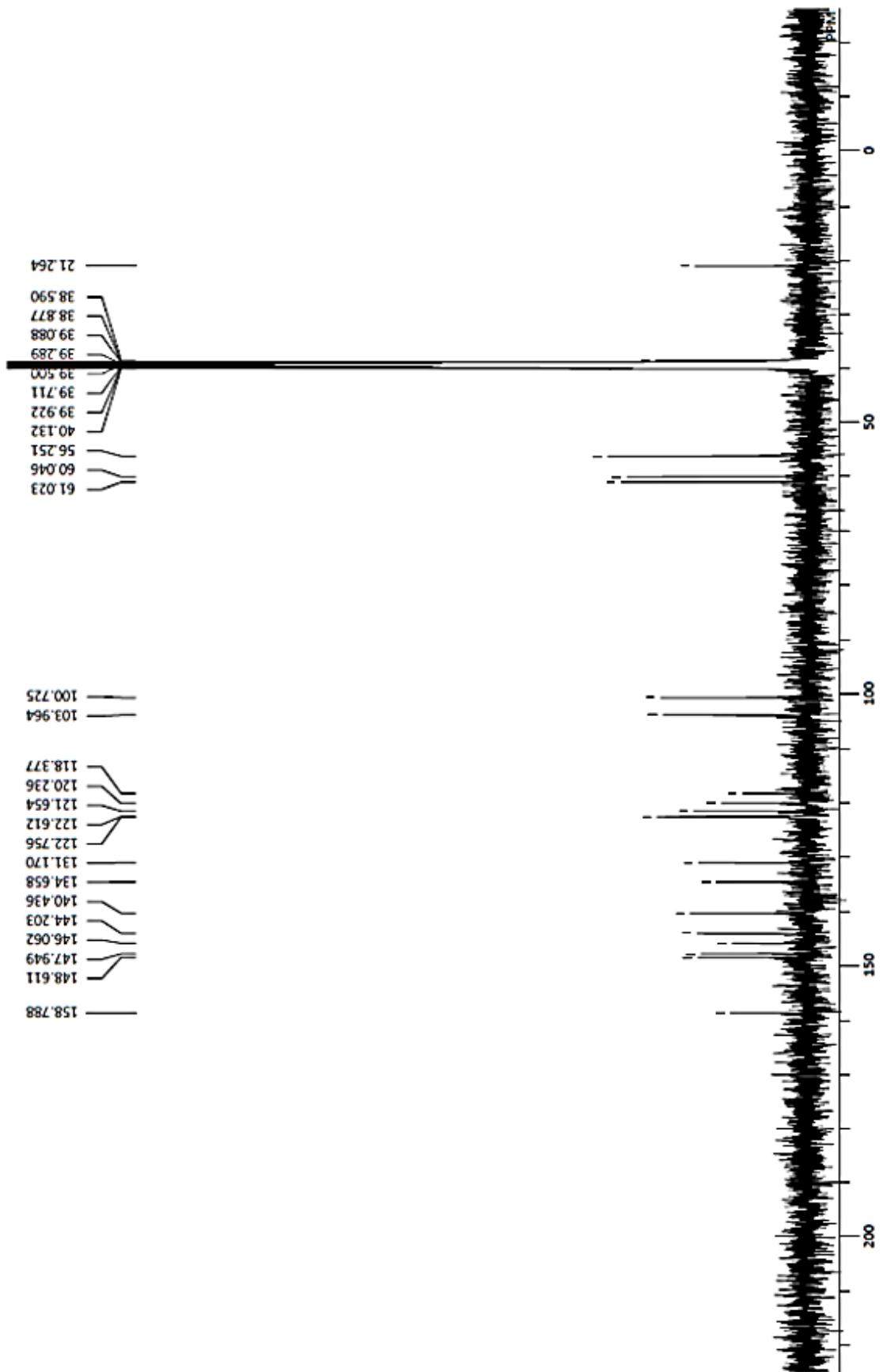


Fig. S9(B)  $^{13}\text{C}$  NMR spectrum of compound 6 (100 MHz,  $\text{DMSO}-d_6$ ).

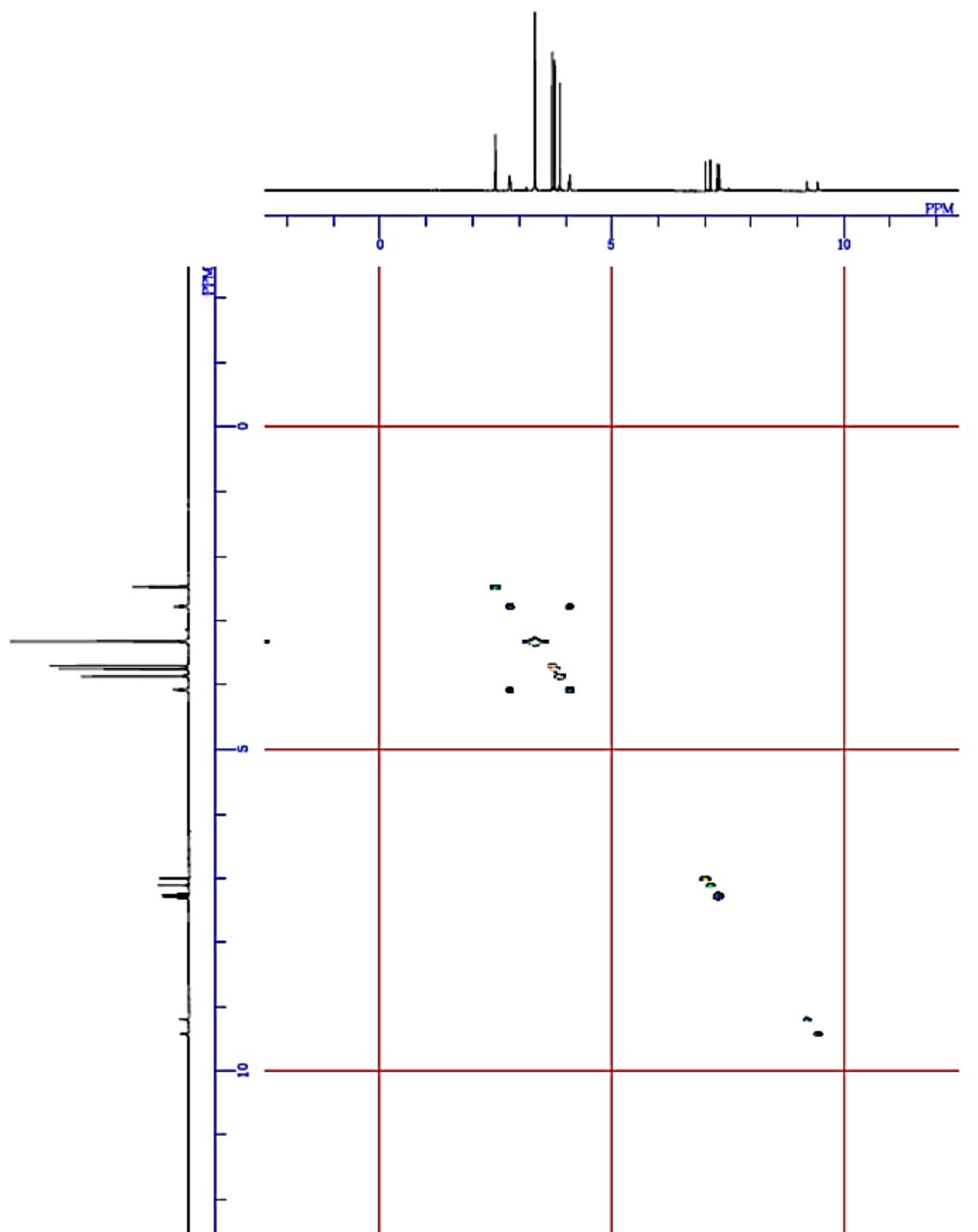


Fig. S9(C) COSY spectrum of compound **6** (400 MHz, DMSO- $d_6$ ).

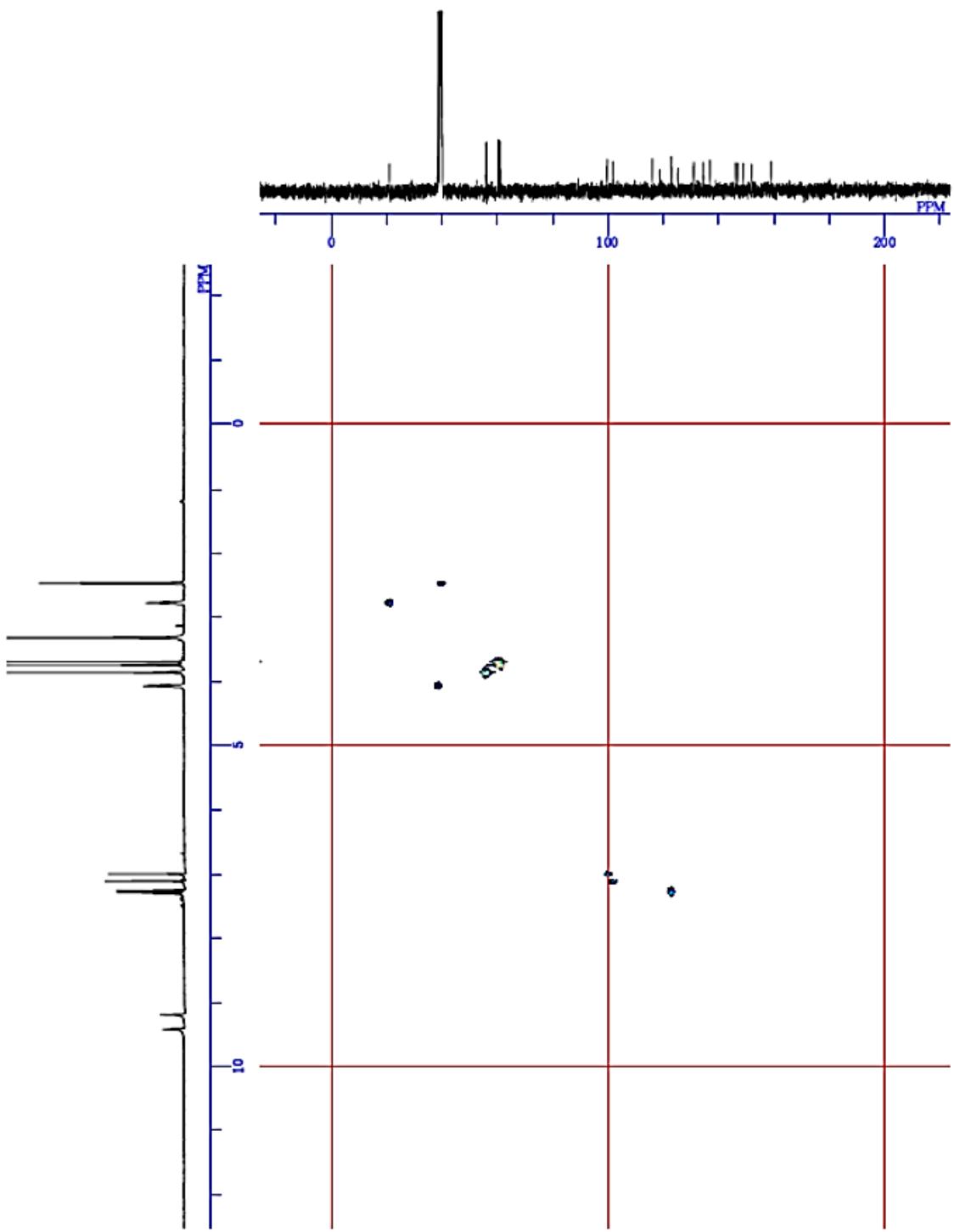
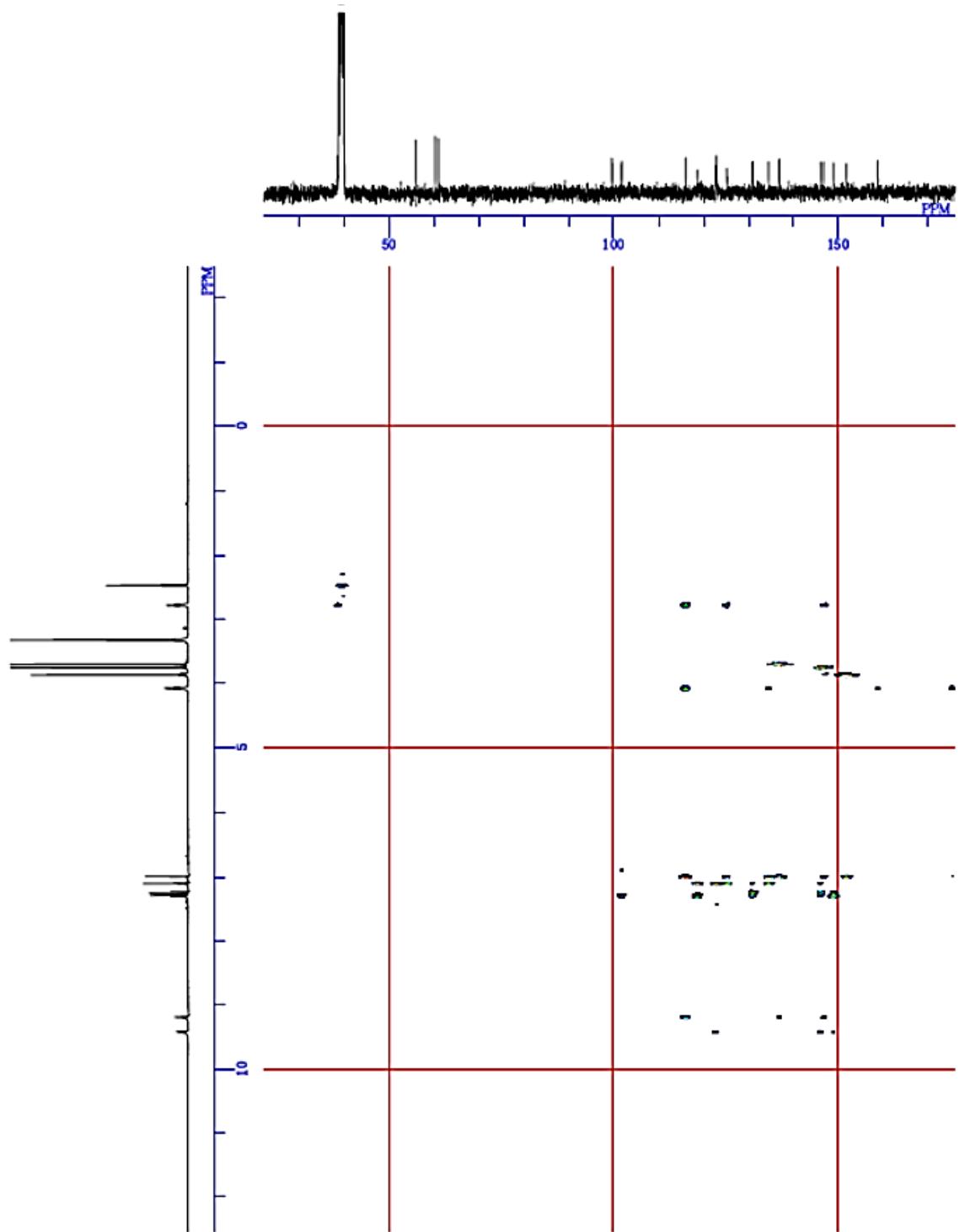


Fig. S9(D) HMQC spectrum of compound 6 (400 MHz, DMSO-*d*<sub>6</sub>).



**Fig. S9(E)** HMQC spectrum of compound **6** (400 MHz, DMSO-*d*<sub>6</sub>).

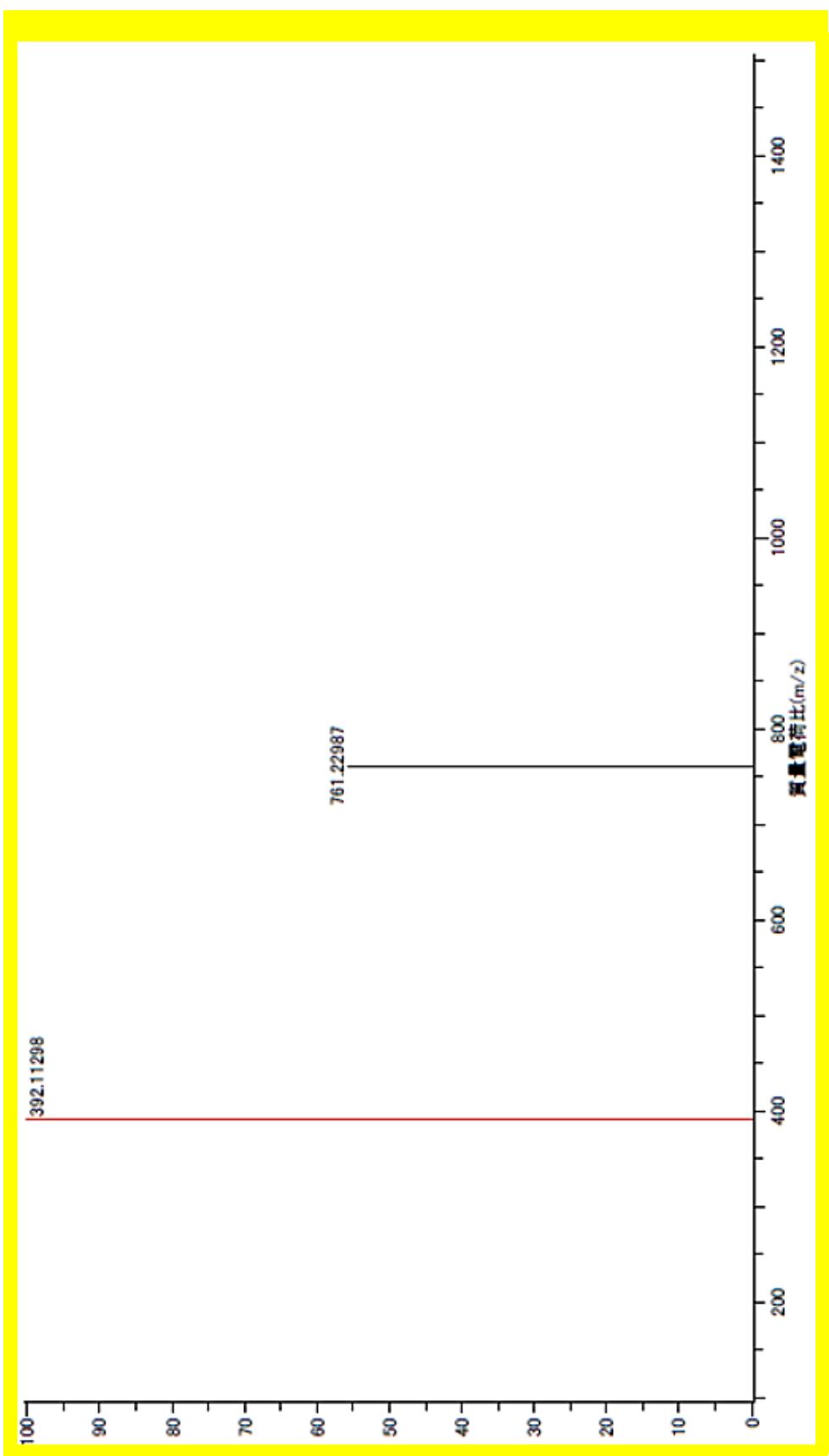


Fig. S9(F) HRESIMS spectrum of compound 6.

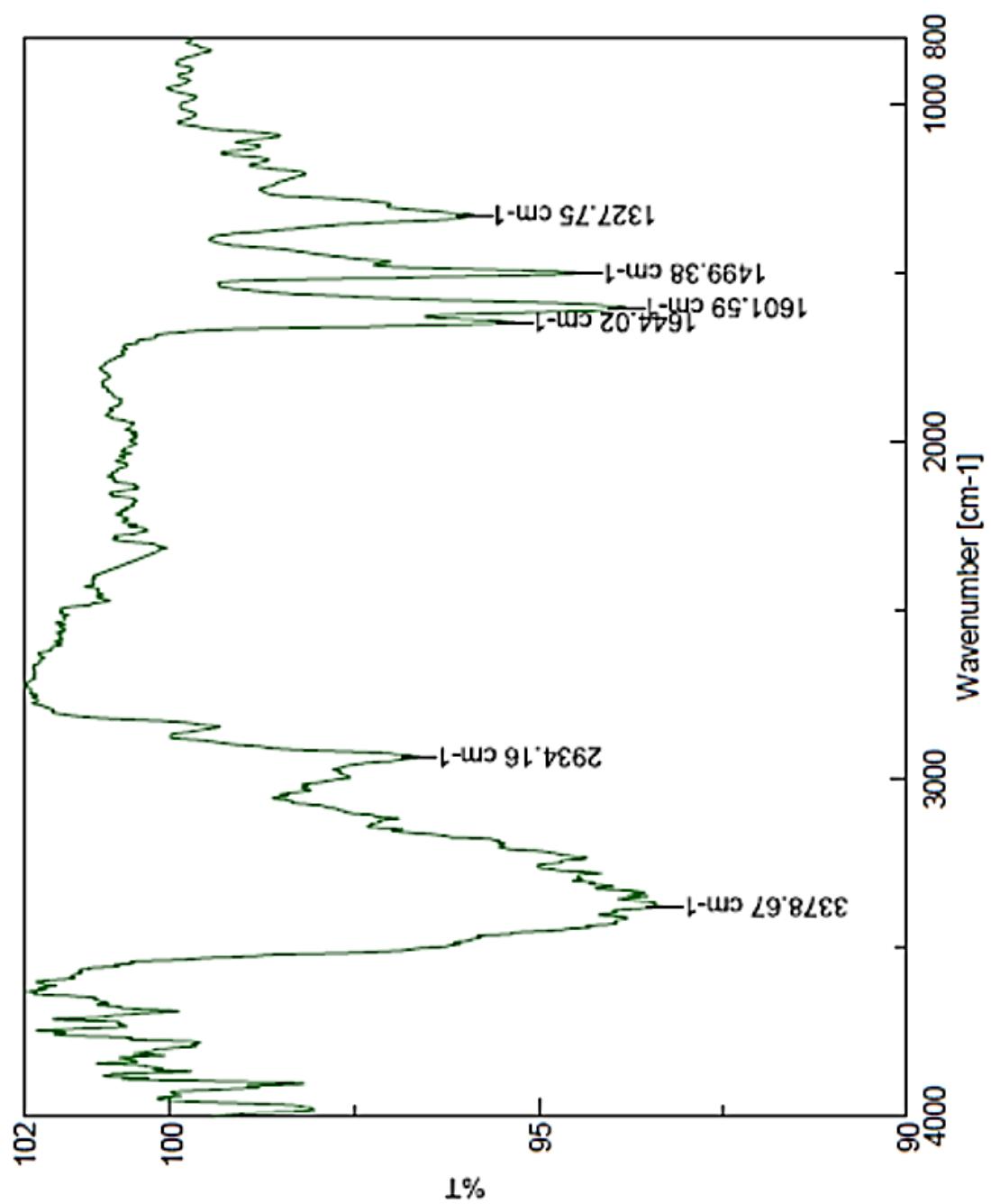


Fig. S9(G) IR spectrum of compound 6.

18) **Figure S10.** Spectroscopic data of compound 7.

**Fig. S10(A)**  $^1\text{H}$  NMR spectrum of compound 7 (600 MHz, DMSO- $d_6$ ).

**Fig. S10(B)**  $^{13}\text{C}$  NMR spectrum of compound 7 (150 MHz, DMSO- $d_6$ ).

**Fig. S10(C)** COSY spectrum of compound 7 (600 MHz, DMSO- $d_6$ ).

**Fig. S10(D)** HMQC spectrum of compound 7 (600 MHz, DMSO- $d_6$ ).

**Fig. S10(E)** HMBC spectrum of compound 7 (600 MHz, DMSO- $d_6$ ).

**Fig. S10(F)** HRESIMS spectrum of compound 7.

**Fig. S10(G)** IR spectrum of compound 7.

**Fig. S10(H)** CD spectrum of compound 7.

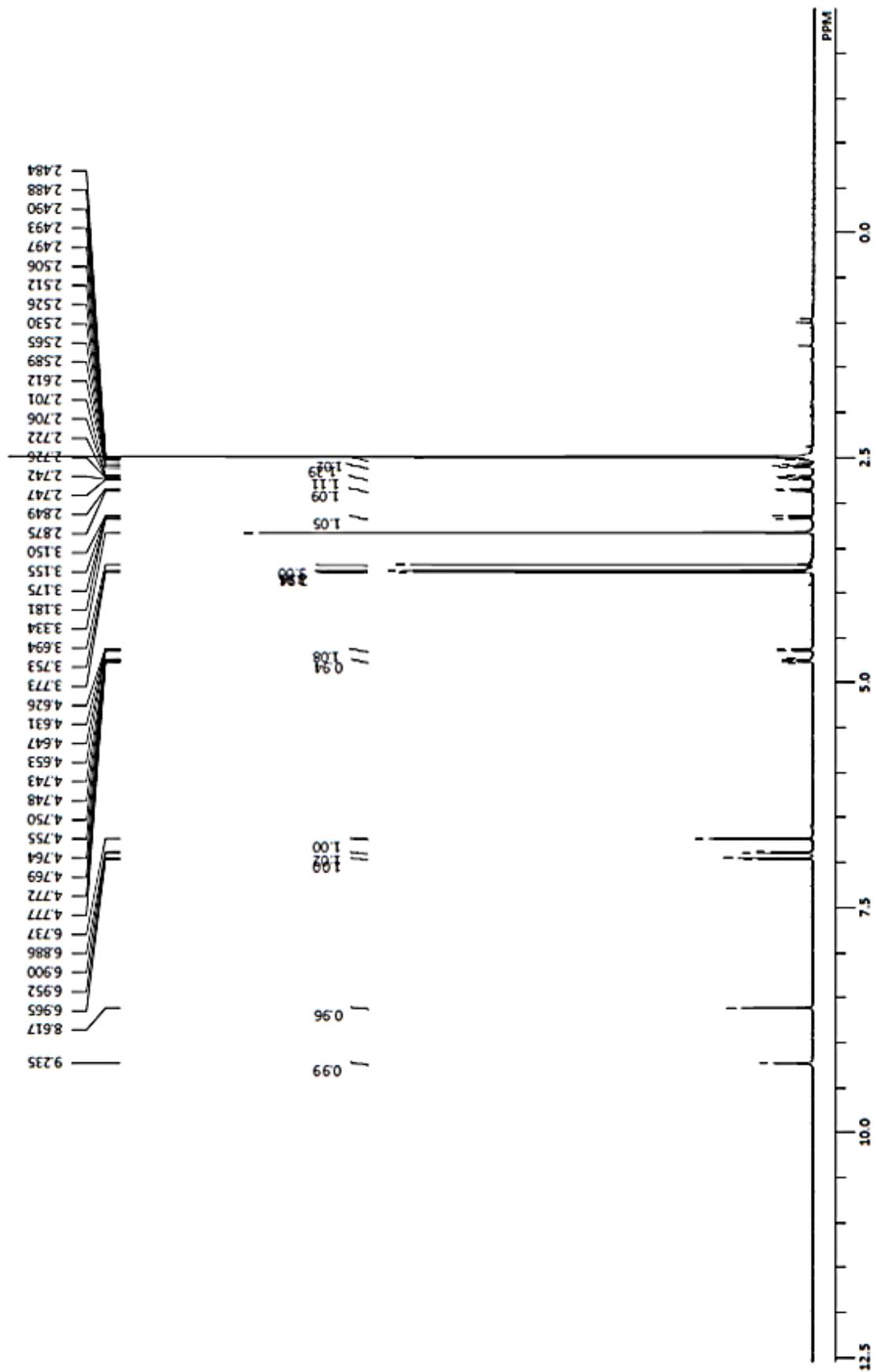
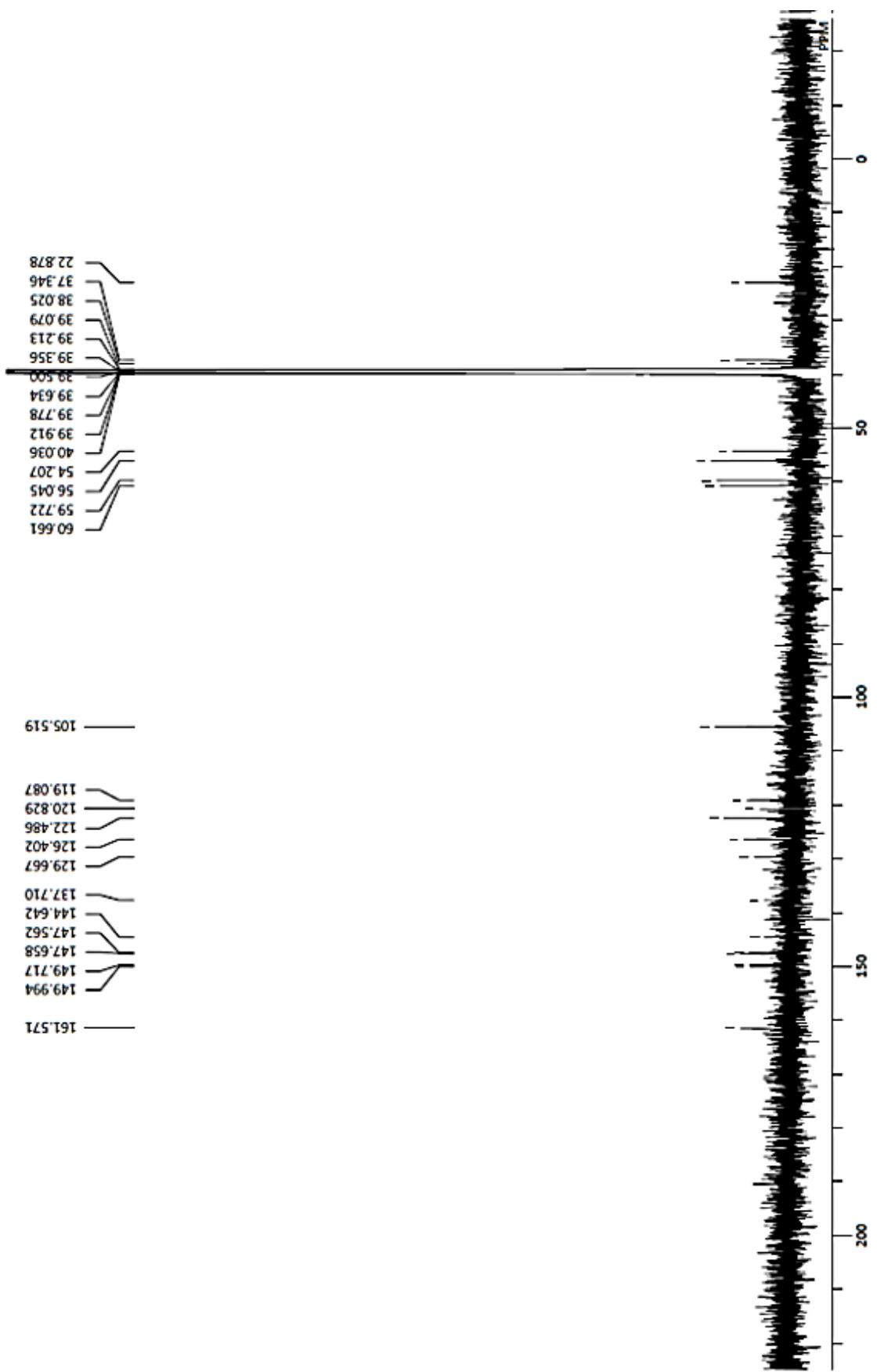


Fig. S10(A) <sup>1</sup>H NMR spectrum of compound 7 (600 MHz, DMSO-*d*<sub>6</sub>)



**Fig. S10(B)**  $^{13}\text{C}$  NMR spectrum of compound 7 (150 MHz,  $\text{DMSO}-d_6$ ).

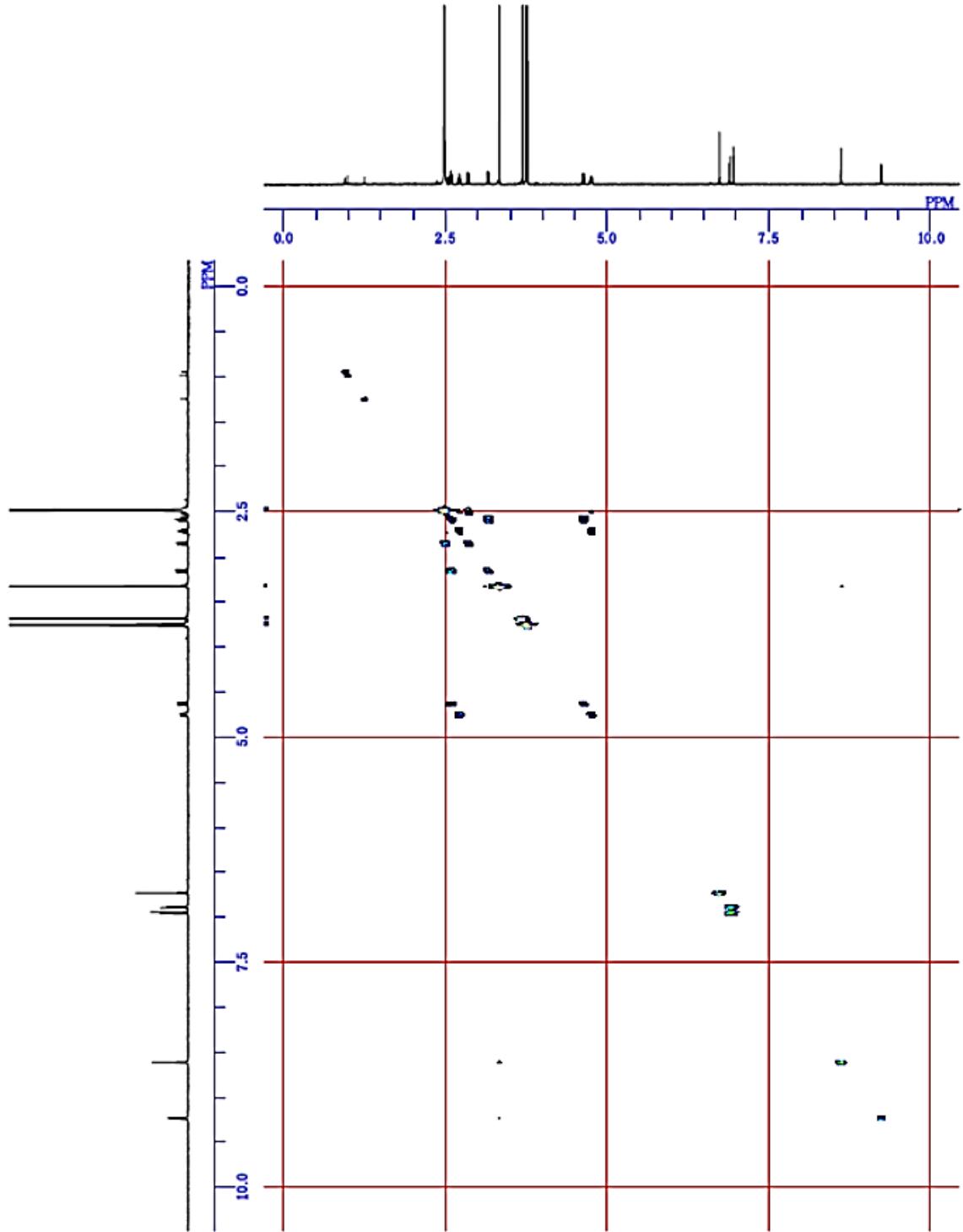
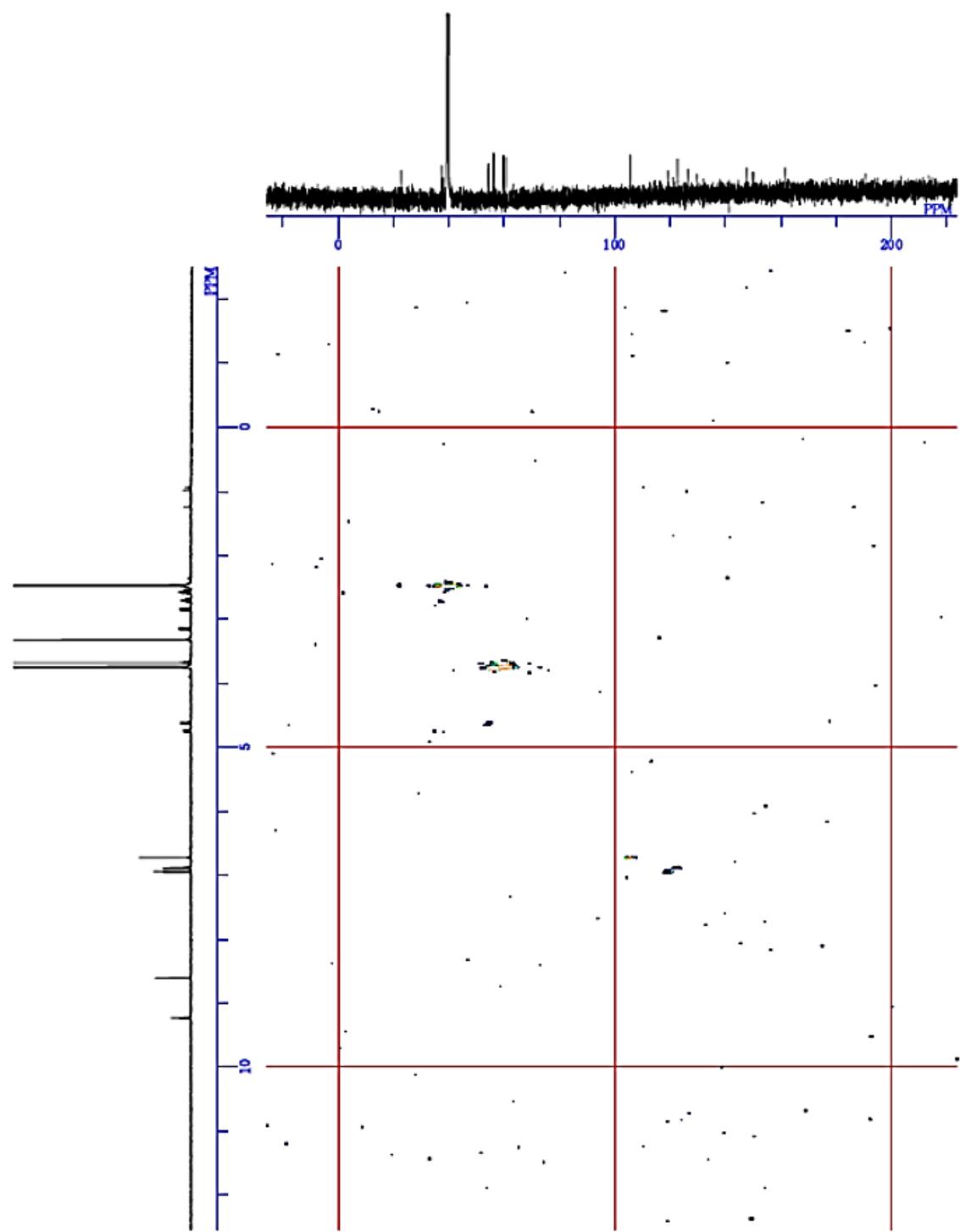
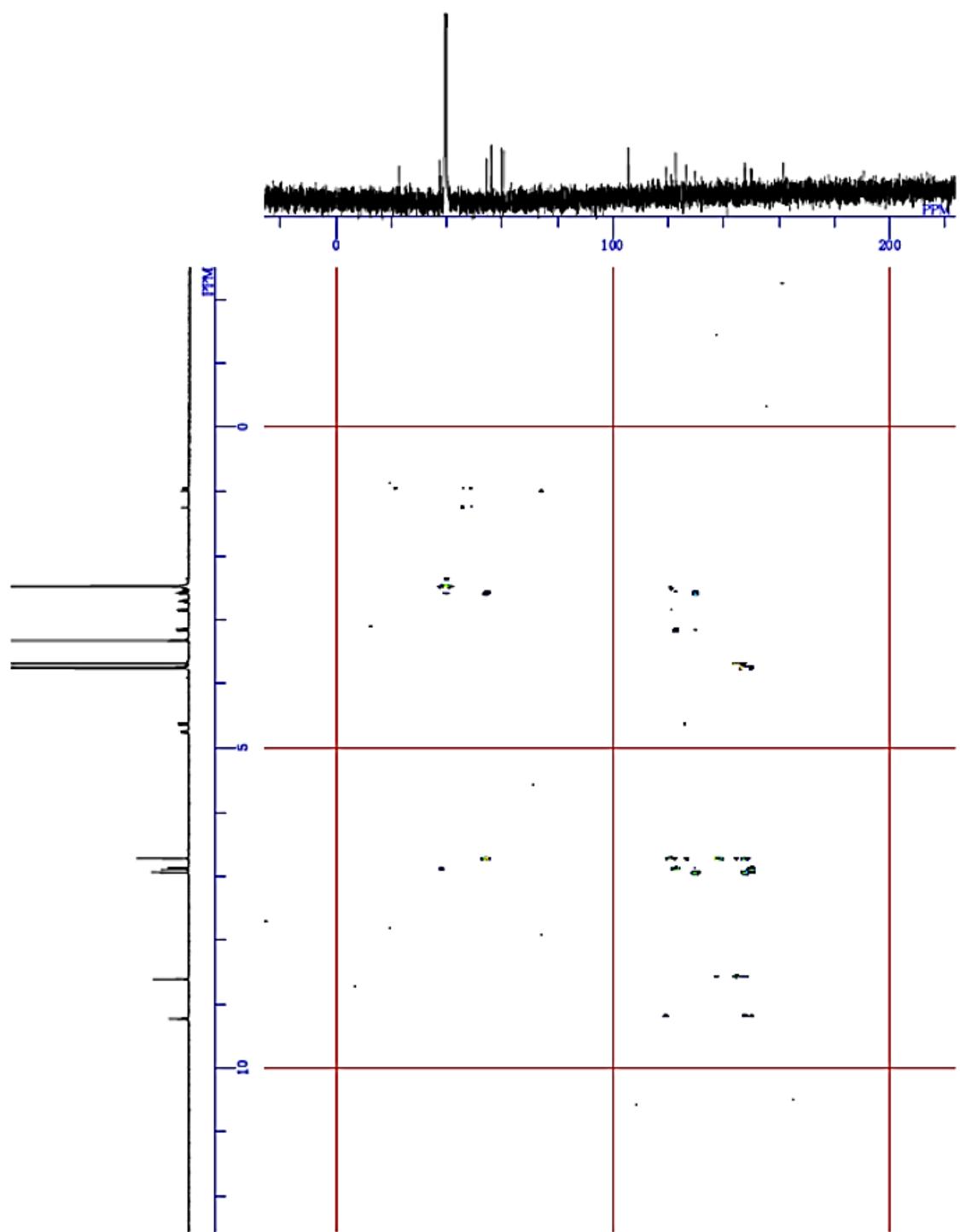


Fig. S10(C) COSY spectrum of compound 7 (600 MHz,  $\text{DMSO}-d_6$ ).



**Fig. S10(D)** HMQC spectrum of compound 7 (600 MHz,  $\text{DMSO}-d_6$ ).



**Fig. S10(E)** HMBC spectrum of compound 7 (600 MHz,  $\text{DMSO}-d_6$ ).

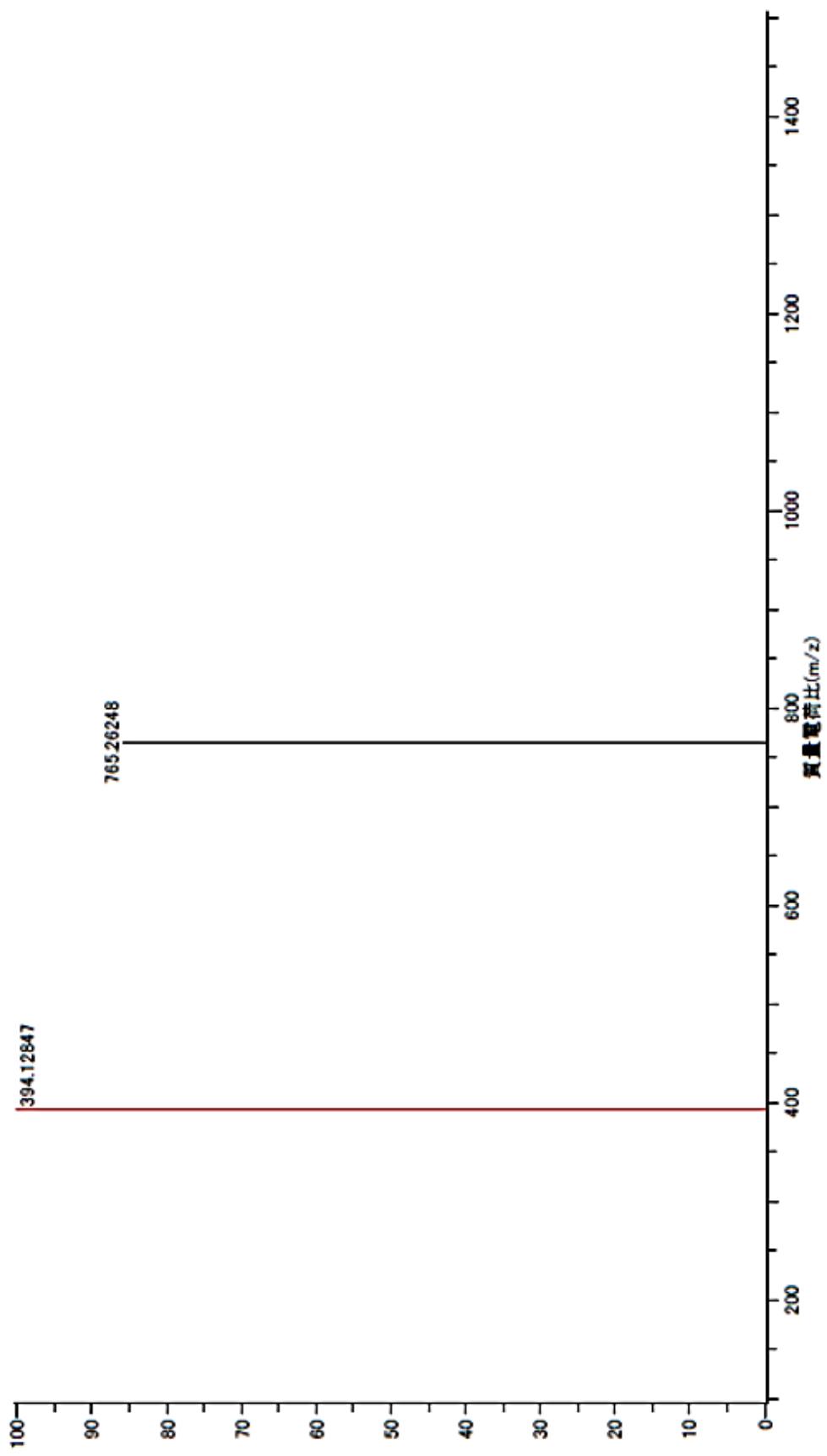


Fig. S10(F) HRESIMS spectrum of compound 7.

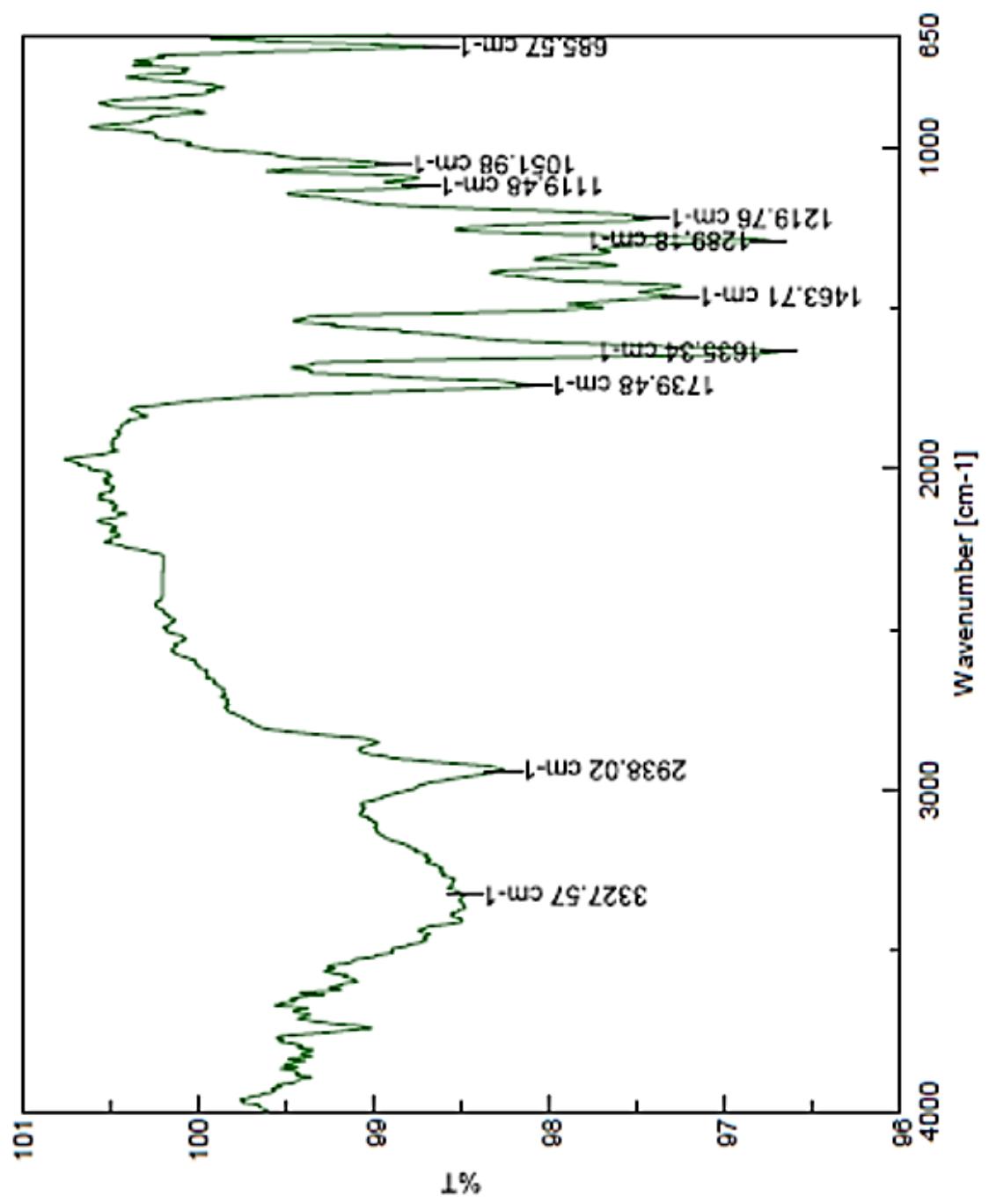


Fig. S10(G) IR spectrum of compound 7.

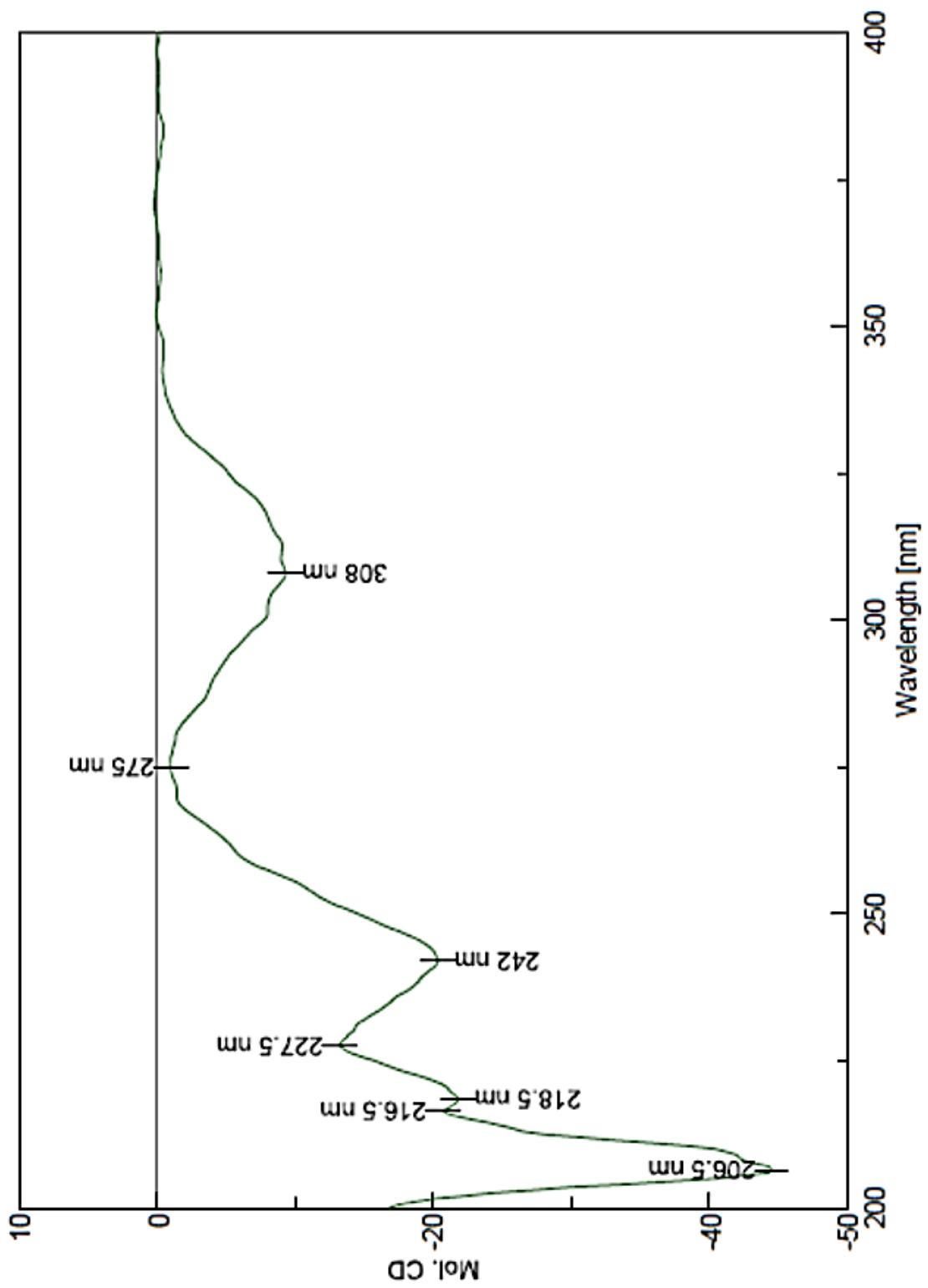


Fig. S10(H) CD spectrum of compound 7.

19) **Figure S11.** Spectroscopic data of compound **8**.

**Fig. S11(A)**  $^1\text{H}$  NMR spectrum of compound **8** (600 MHz, DMSO- $d_6$ ).

**Fig. S11(B)**  $^{13}\text{C}$  NMR spectrum of compound **8** (150 MHz, DMSO- $d_6$ ).

**Fig. S11(C)** COSY spectrum of compound **8** (600 MHz, DMSO- $d_6$ ).

**Fig. S11(D)** HMQC spectrum of compound **8** (600 MHz, DMSO- $d_6$ ).

**Fig. S11(E)** HMBC spectrum of compound **8** (600 MHz, DMSO- $d_6$ ).

**Fig. S11(F)** HRESIMS spectrum of compound **8**.

**Fig. S11(G)** IR spectrum of compound **8**.

**Fig. S11(H)** CD spectrum of compound **8**.

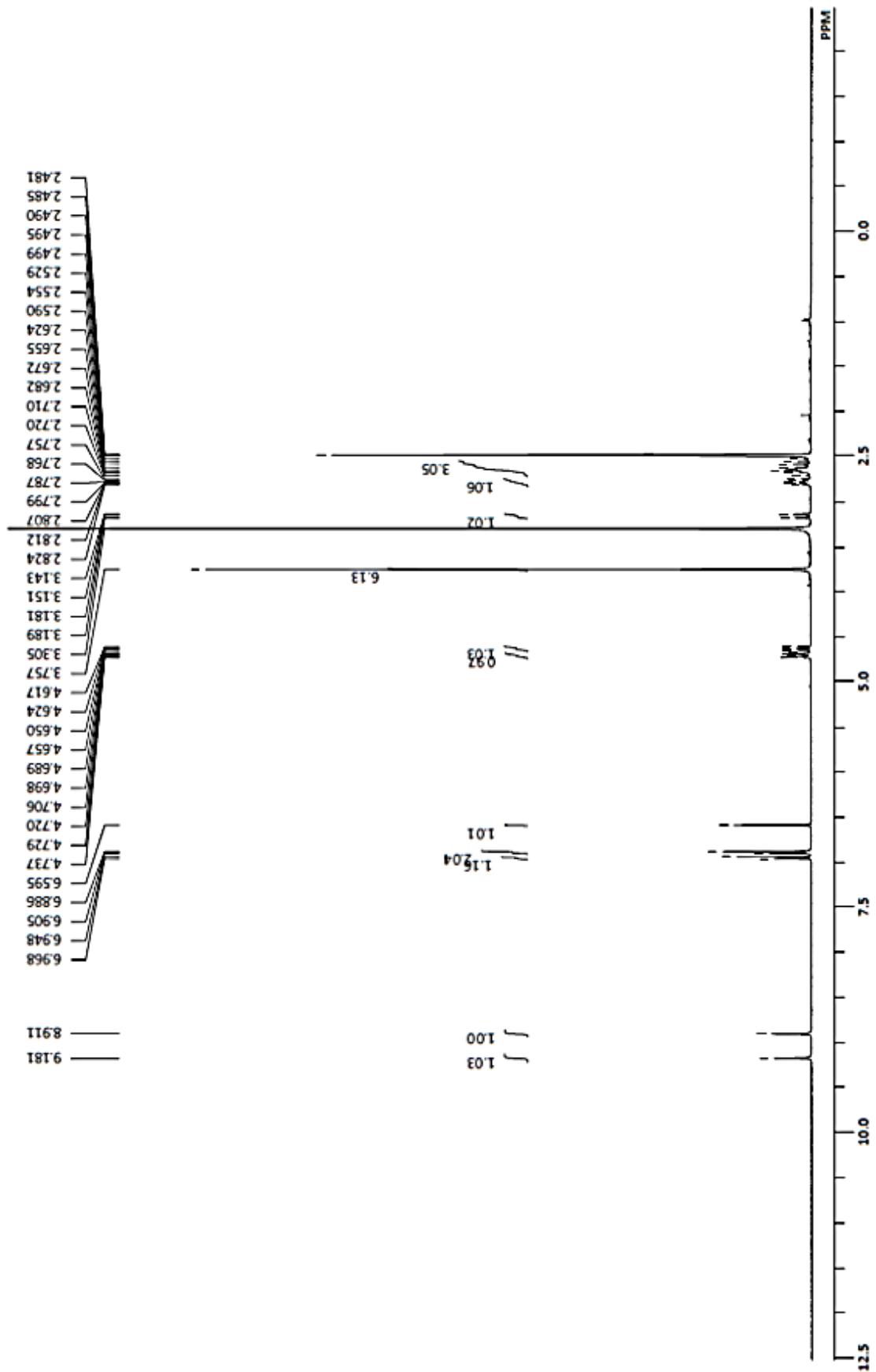
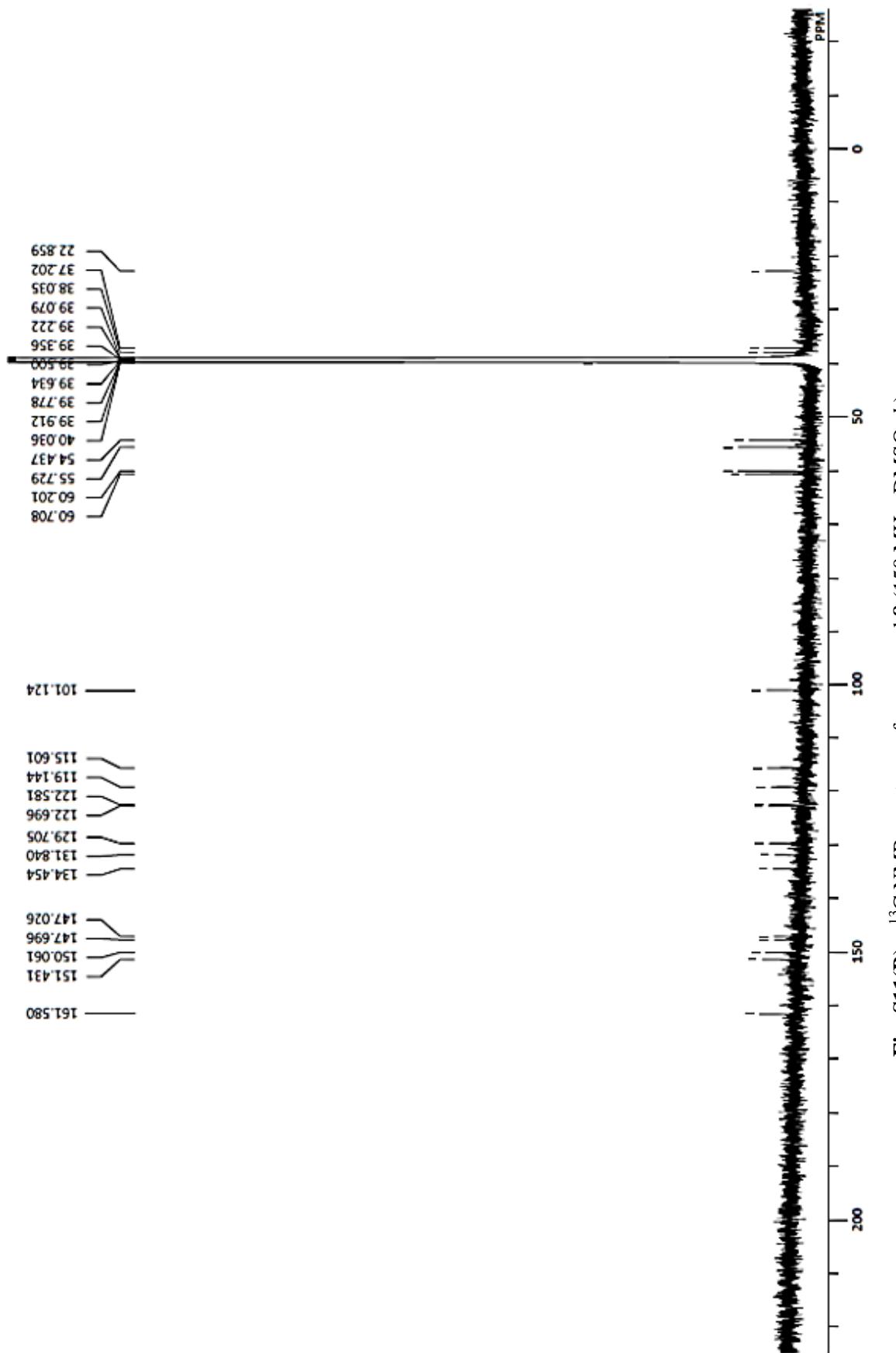


Fig. S11(A) <sup>1</sup>H NMR spectrum of compound 8 (600 MHz, DMSO-*d*<sub>6</sub>).



**Fig. S11(B)**  $^{13}\text{C}$  NMR spectrum of compound **8** (150 MHz,  $\text{DMSO}-d_6$ ).

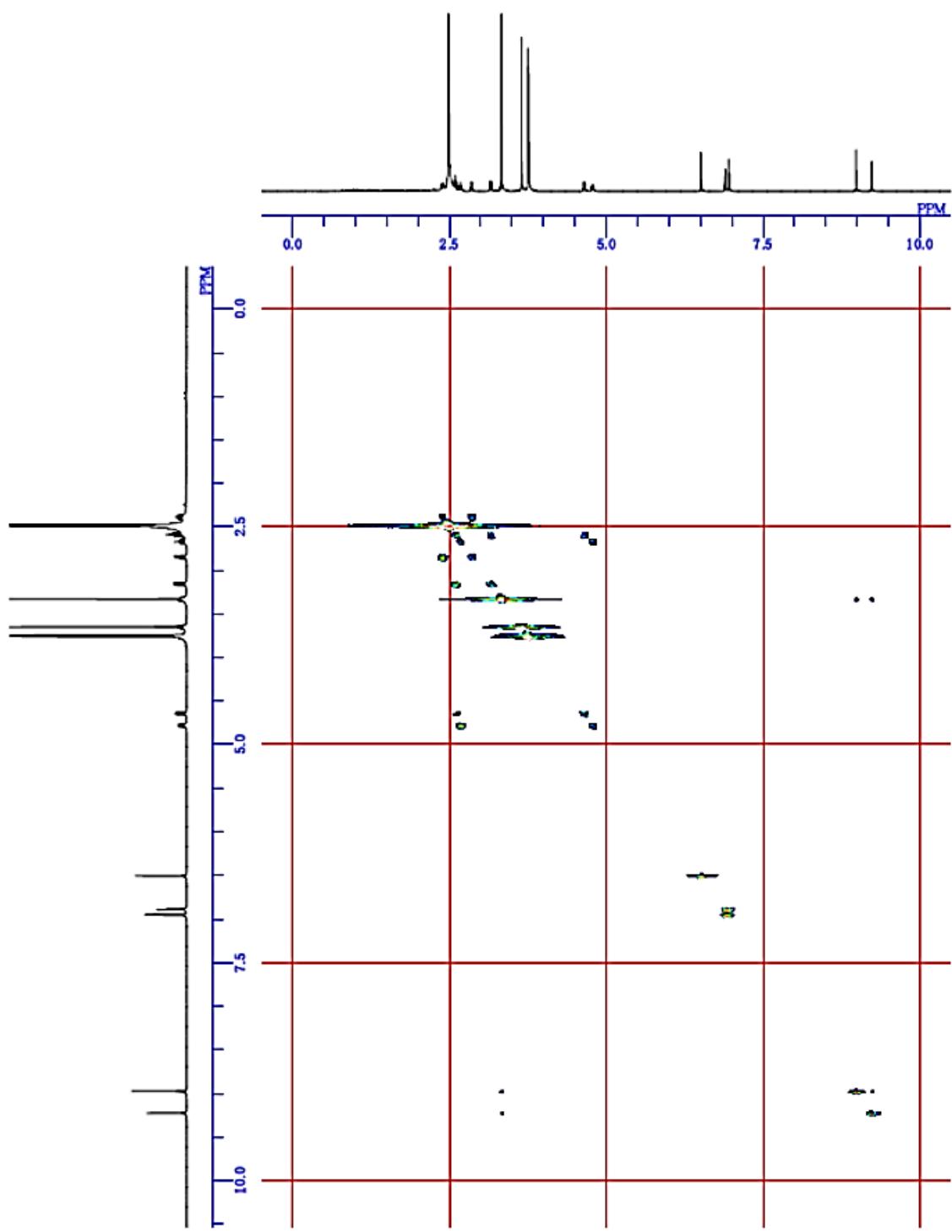
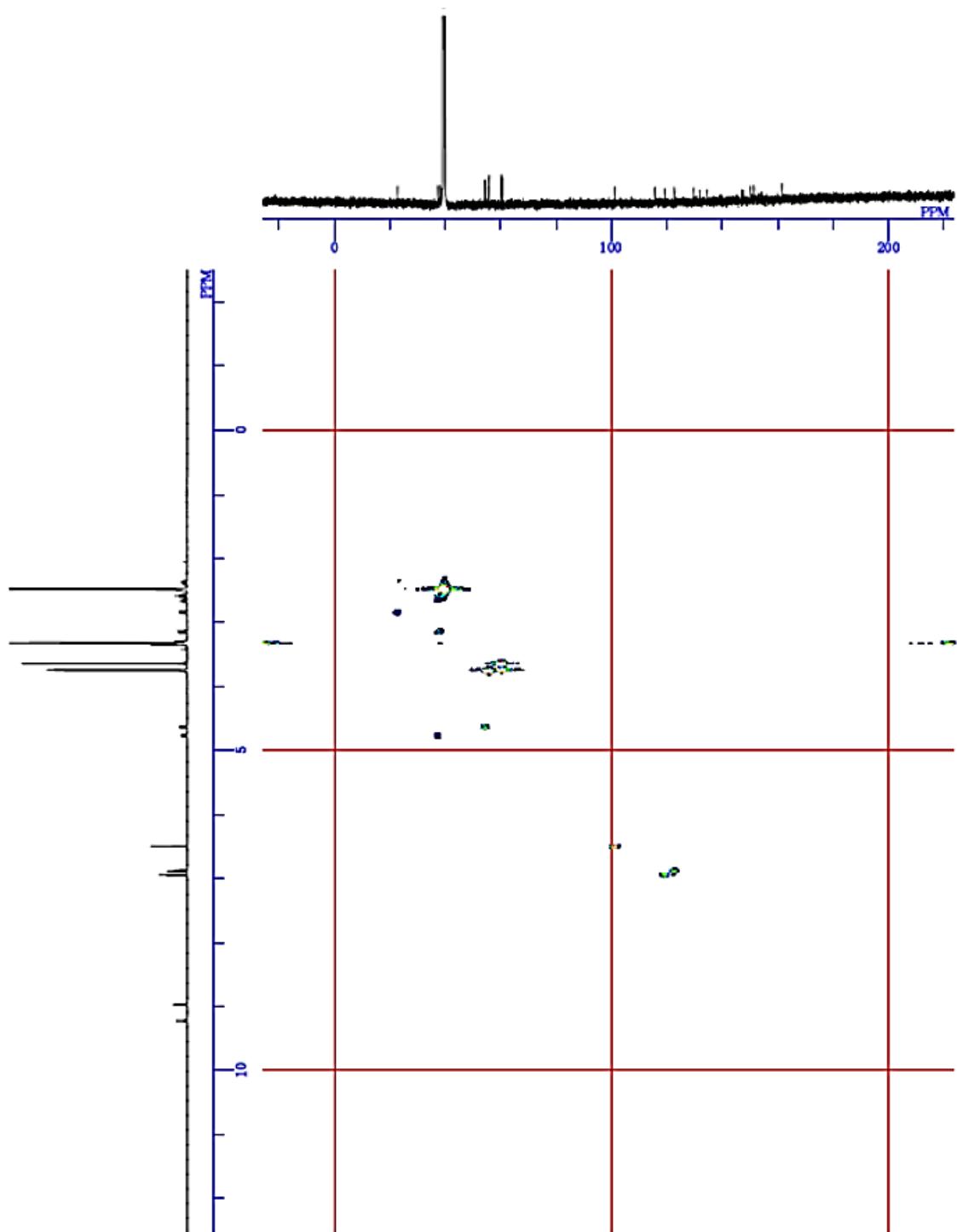


Fig. S11(C) COSY spectrum of compound **8** (600 MHz, DMSO- $d_6$ ).



**Fig. S11(D)** HMQC spectrum of compound **8** (600 MHz,  $\text{DMSO-}d_6$ ).

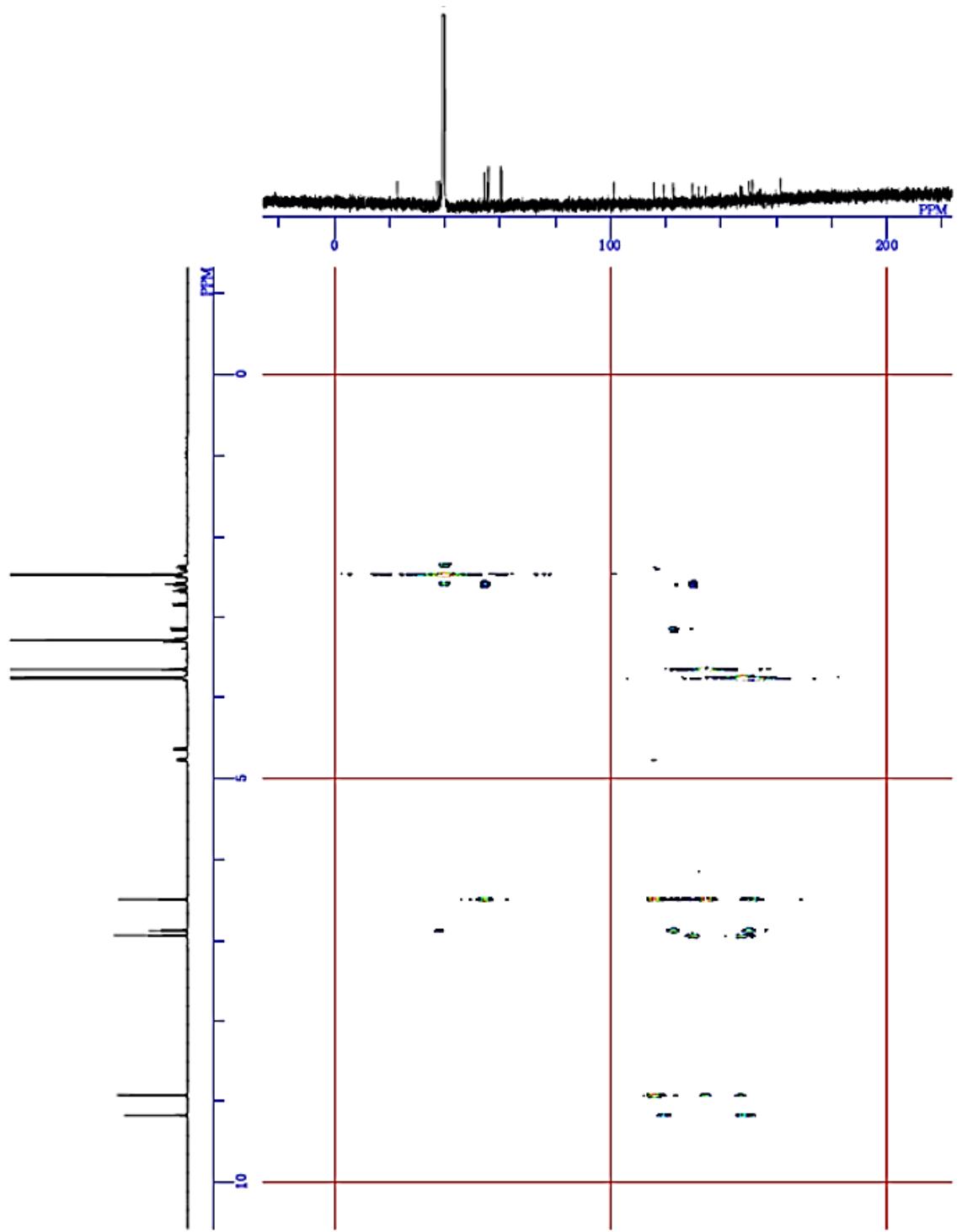


Fig. S11(E) HMBC spectrum of compound **8** (600 MHz, DMSO- $d_6$ ).

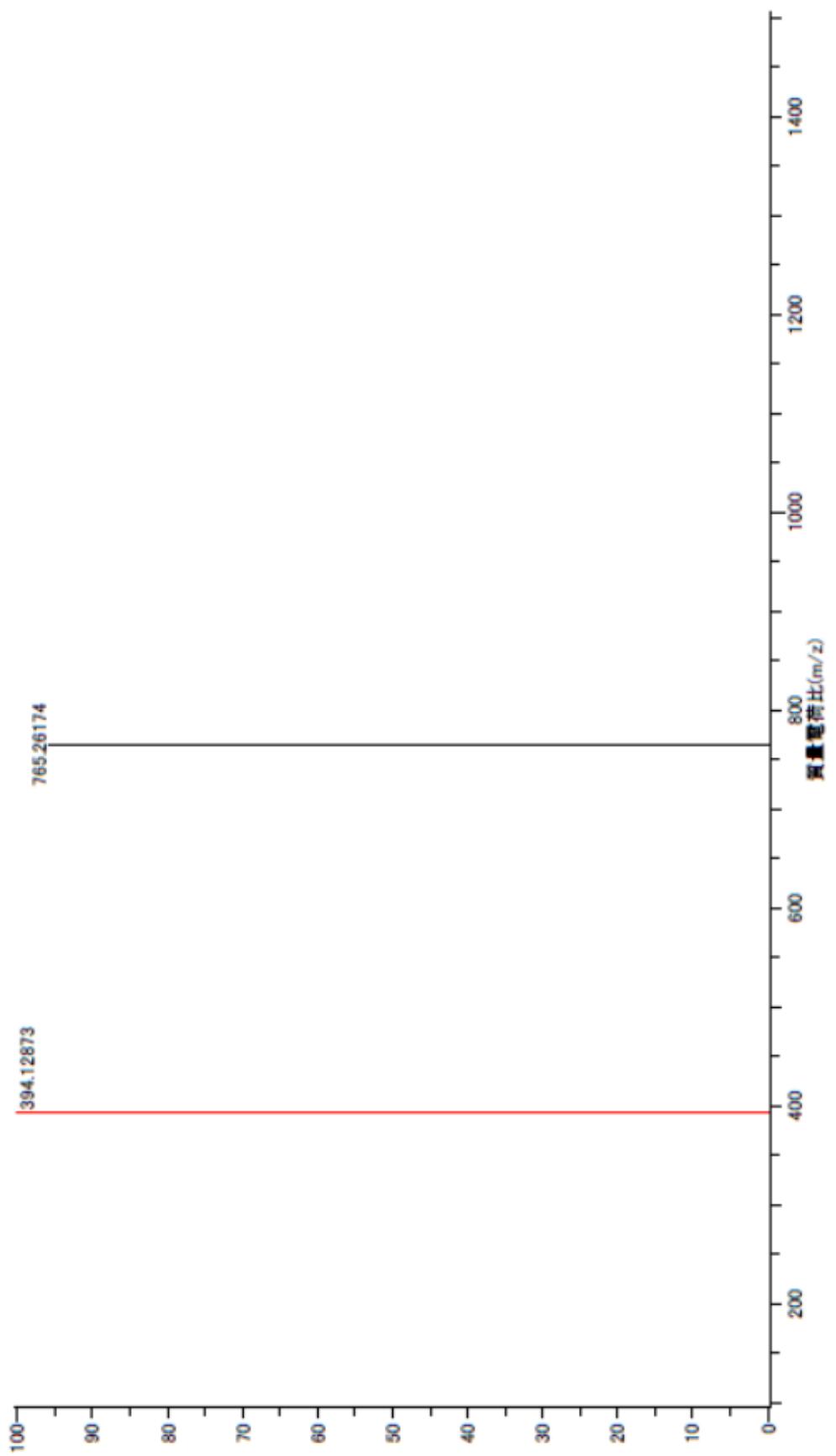


Fig. S11(F) HRESIMS spectrum of compound 8.

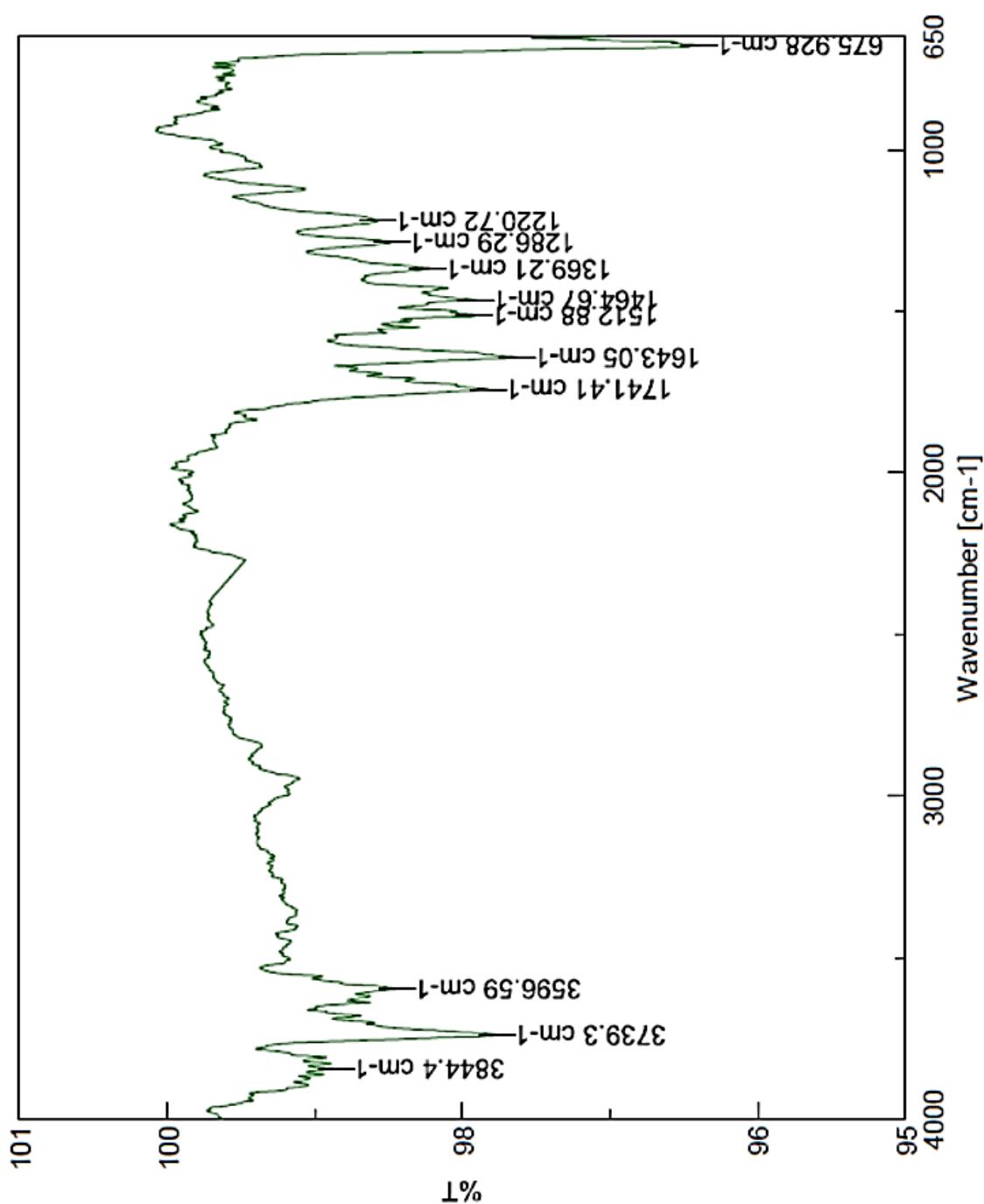


Fig. S11(G) IR spectrum of compound 8.

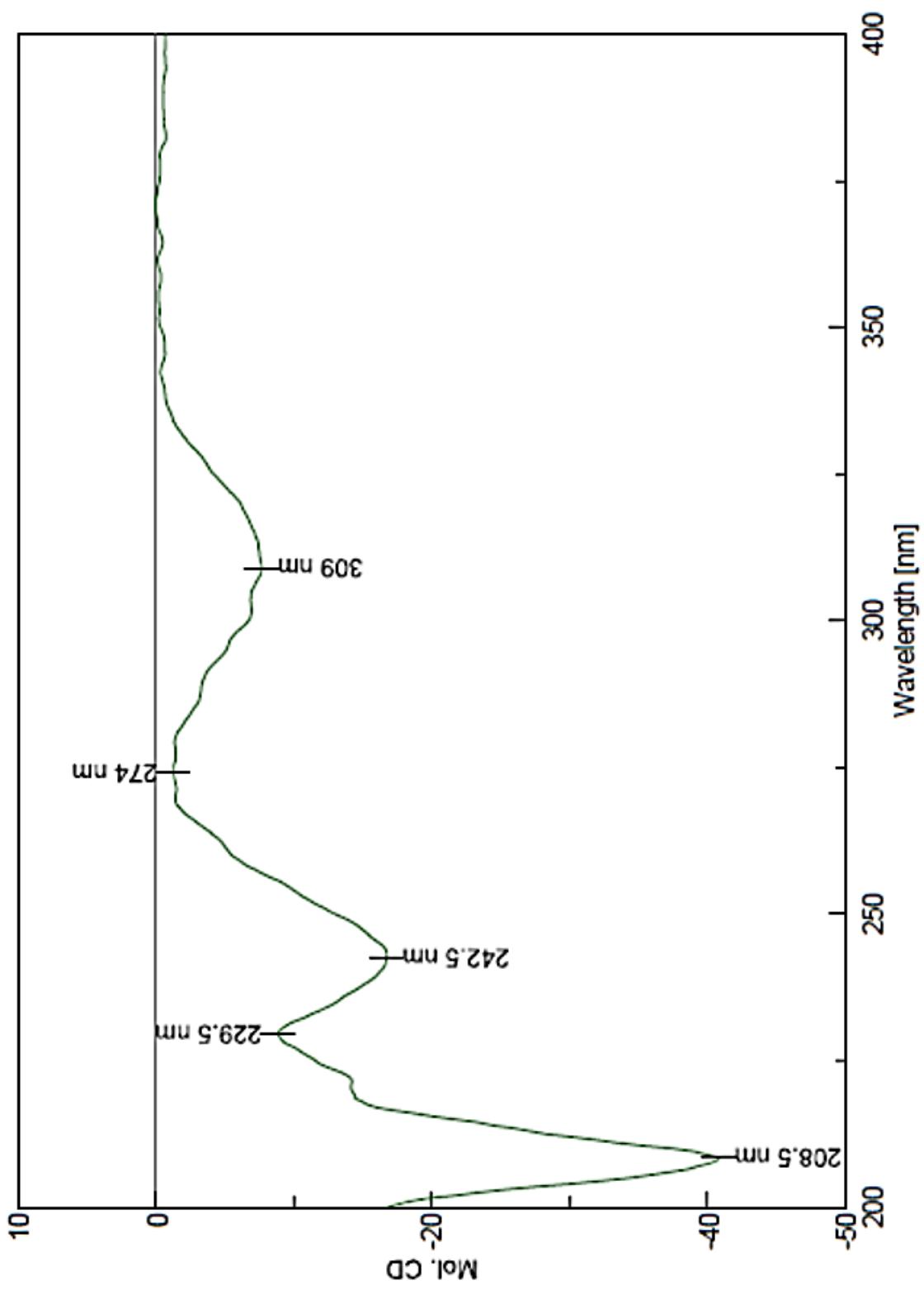


Fig. S11(H) CD spectrum of compound 8.

20) **Figure S12.** Spectroscopic data of compound **9**, which were not available in the literature.<sup>10</sup> Therefore, we are the first to provide full spectroscopic data for **9**.

**Fig. S12(A)**  $^1\text{H}$  NMR spectrum of compound **9** (600 MHz, DMSO- $d_6$ ).

**Fig. S12(B)**  $^{13}\text{C}$  NMR spectrum of compound **9** (150 MHz, DMSO- $d_6$ ).

**Fig. S12(C)** COSY spectrum of compound **9** (600 MHz, DMSO- $d_6$ ).

**Fig. S12(D)** HMQC spectrum of compound **9** (600 MHz, DMSO- $d_6$ ).

**Fig. S12(E)** HMBC spectrum of compound **9** (600 MHz, DMSO- $d_6$ ).

**Fig. S12(F)** HRESIMS spectrum of compound **9**.

**Fig. S12(G)** IR spectrum of compound **9**

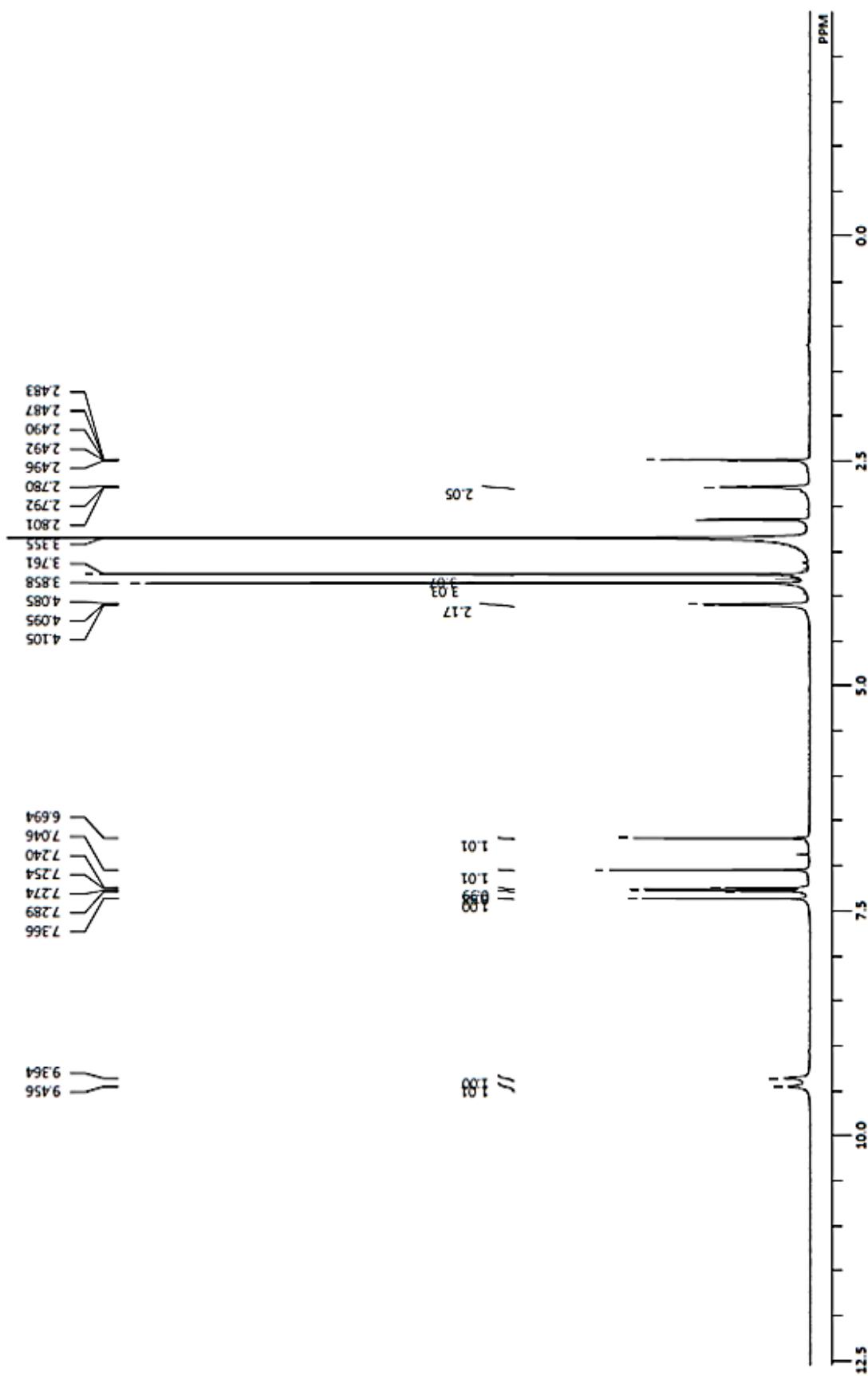


Fig. S12(A) <sup>1</sup>H NMR spectrum of compound 9 (600 MHz, DMSO-*d*<sub>6</sub>).

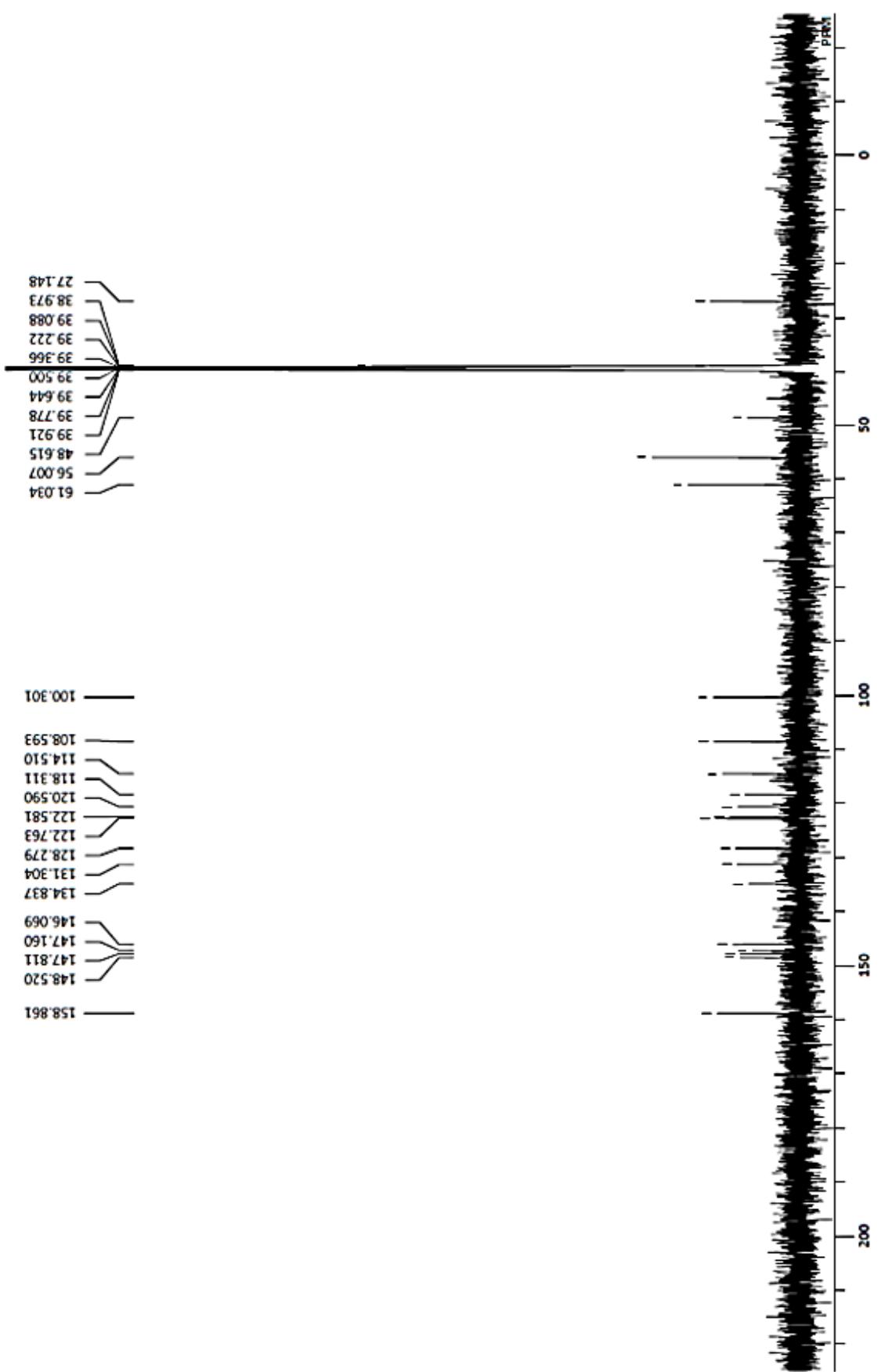


Fig. S12(B)  $^{13}\text{C}$  NMR spectrum of compound 9 (150 MHz,  $\text{DMSO}-d_6$ ).

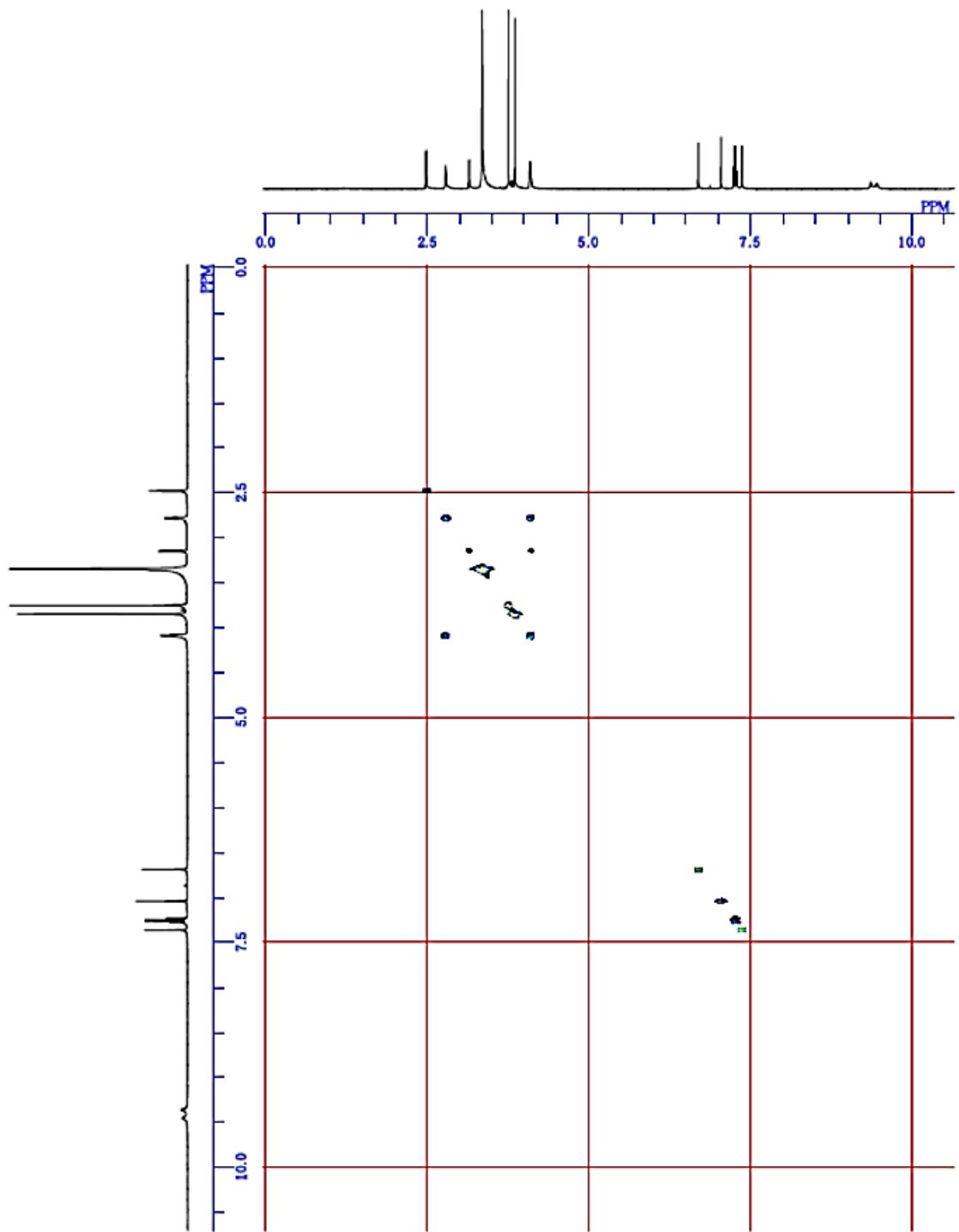


Fig. S12(C) COSY spectrum of compound **9** (600 MHz, DMSO-*d*<sub>6</sub>).

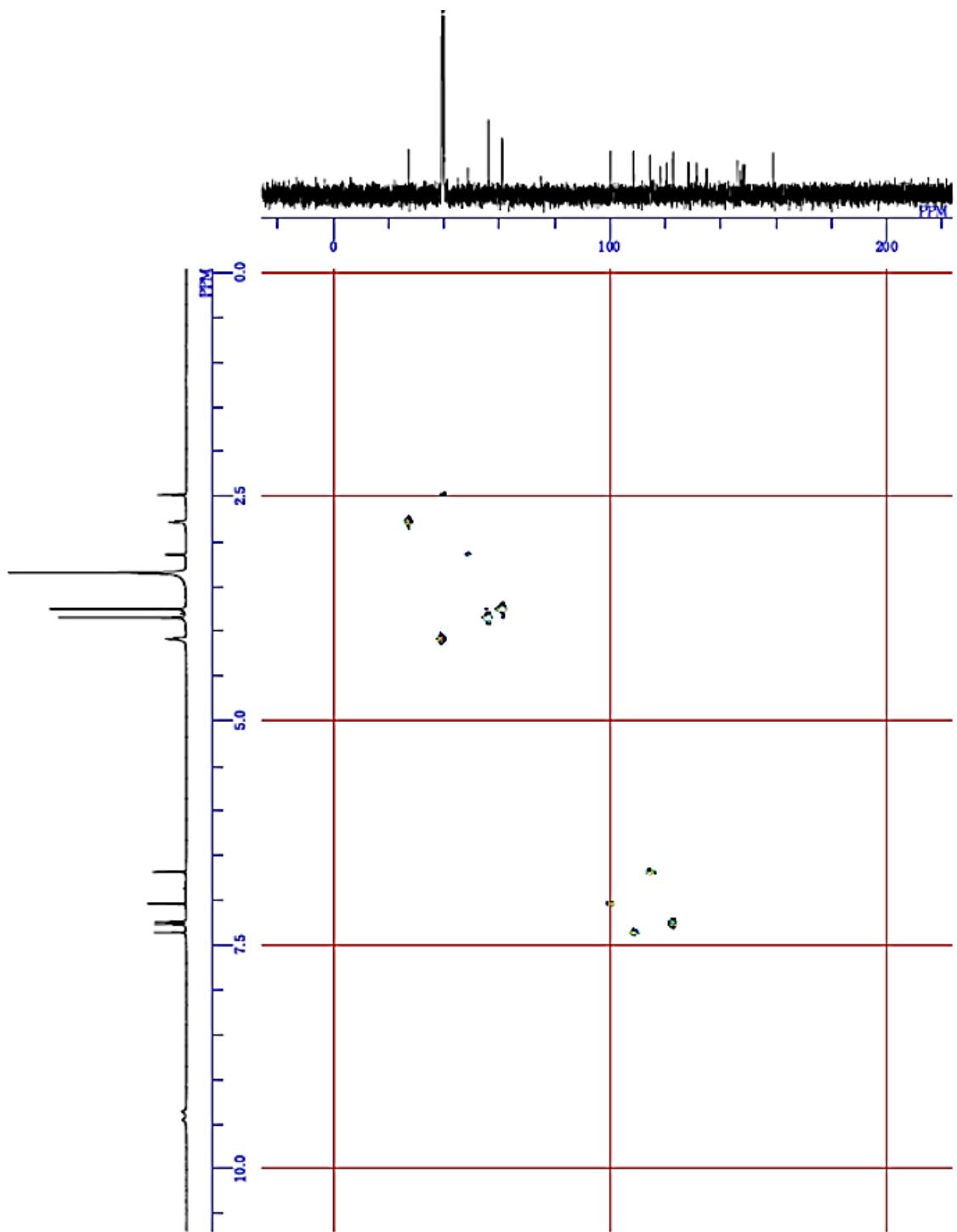


Fig. S12(D) HMQC spectrum of compound **9** (600 MHz, DMSO-*d*<sub>6</sub>).

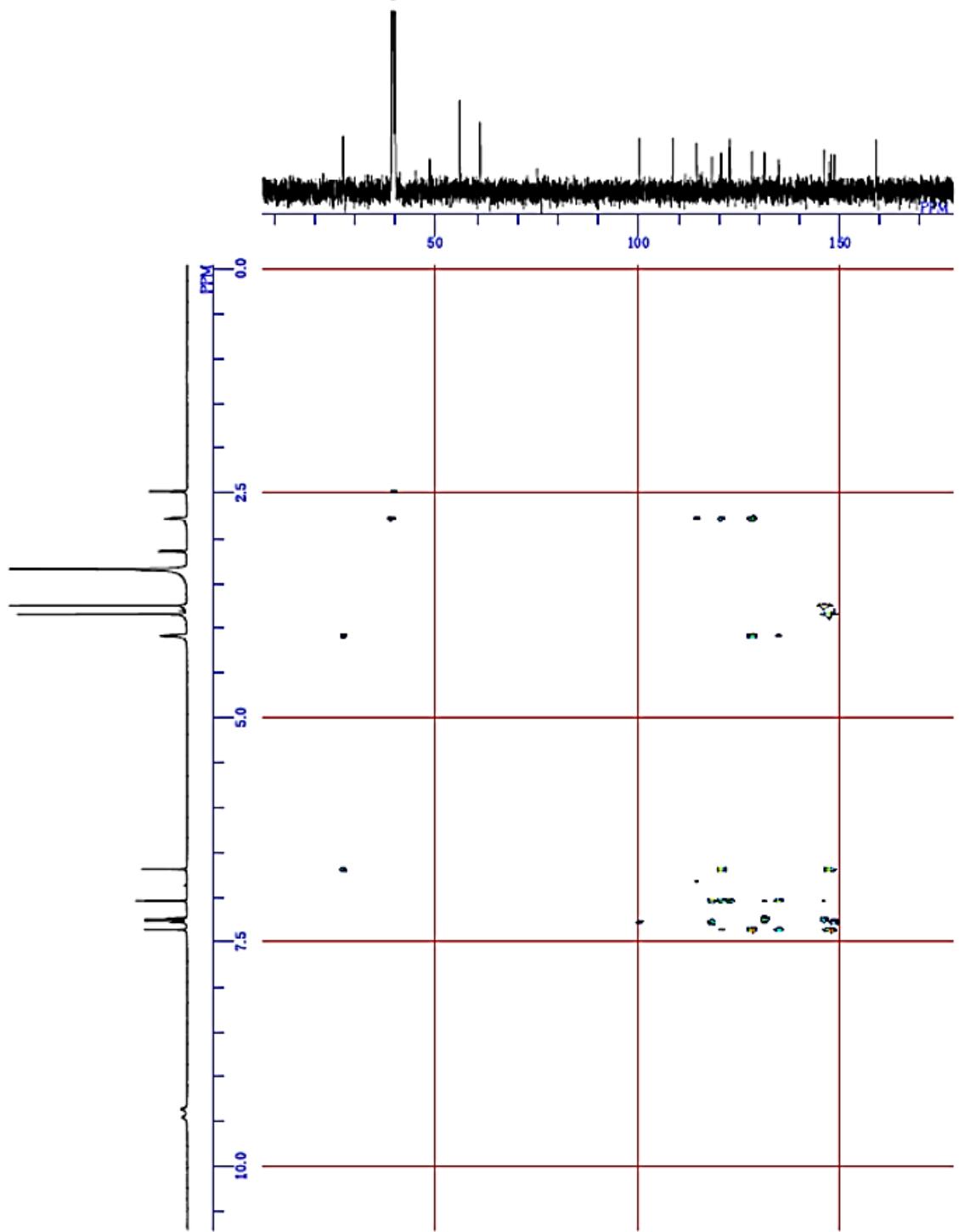


Fig. S12(E) HMBC spectrum of compound **9** (600 MHz,  $\text{DMSO}-d_6$ ).

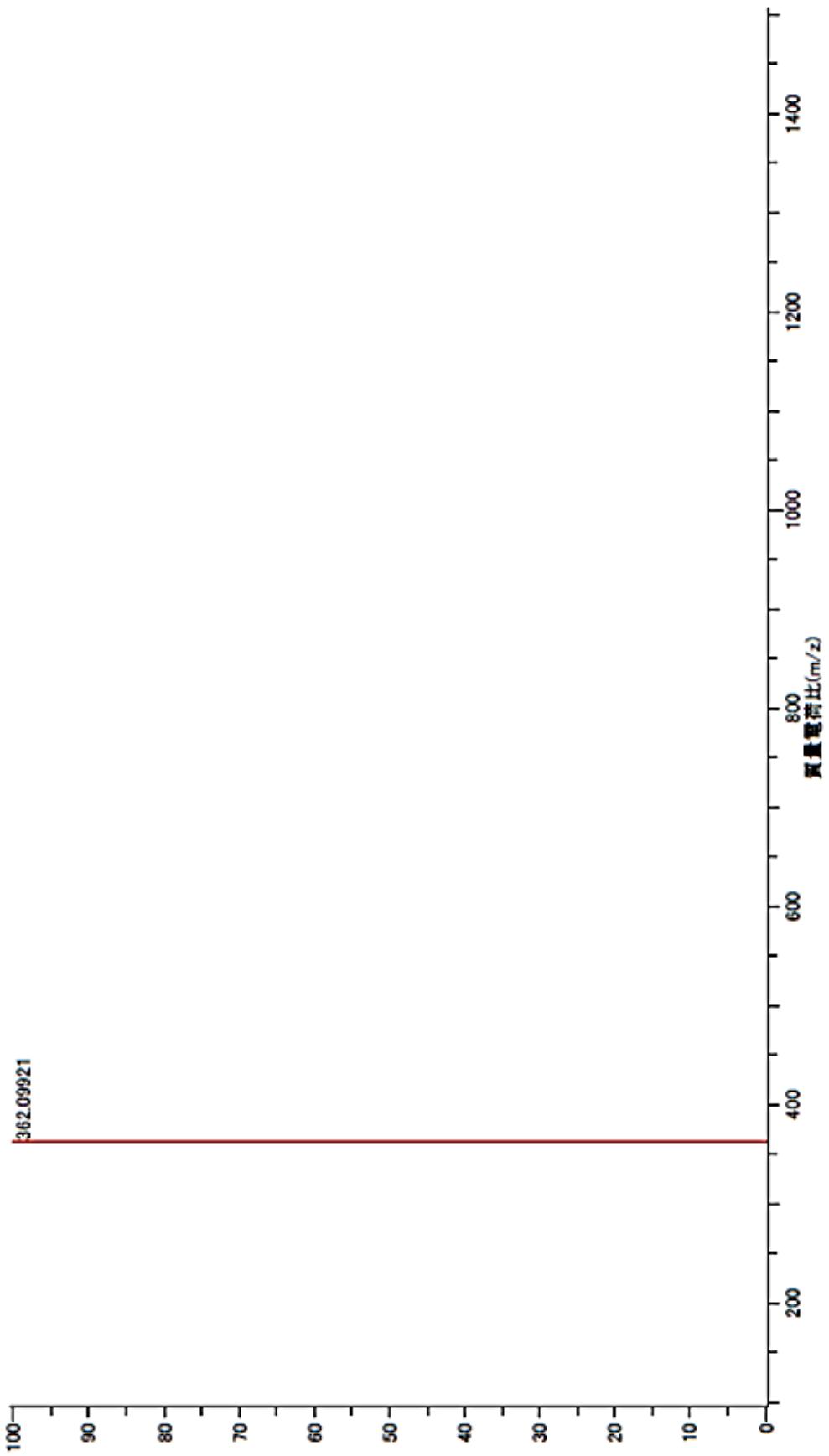


Fig. S12(F) HRESIMS spectrum of compound 9.

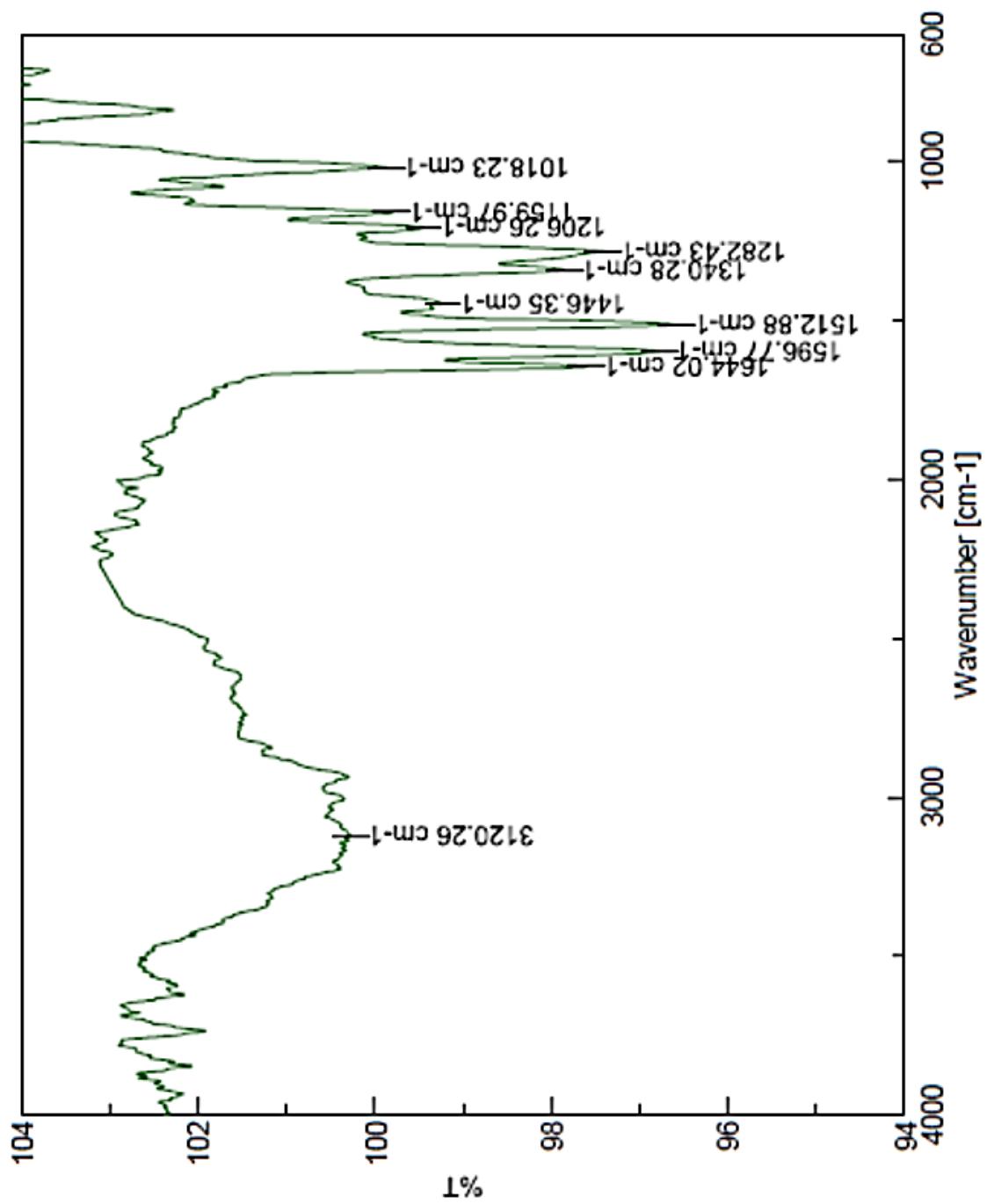


Fig. S12(G) IR spectrum of compound 9.

21) **Figure S13.** Spectroscopic data of compound **10**, which were not available in the literature.<sup>10</sup> Therefore, we are the first to provide full spectroscopic data for **10**.

**Fig. S13(A)**  $^1\text{H}$  NMR spectrum of compound **10** (400 MHz, DMSO- $d_6$ ).

**Fig. S13(B)**  $^{13}\text{C}$  NMR spectrum of compound **10** (100 MHz, DMSO- $d_6$ ).

**Fig. S13(C)** COSY spectrum of compound **10** (400 MHz, DMSO- $d_6$ ).

**Fig. S13(D)** HMQC spectrum of compound **10** (400 MHz, DMSO- $d_6$ ).

**Fig. S13(E)** HMBC spectrum of compound **10** (400 MHz, DMSO- $d_6$ ).

**Fig. S13(F)** HRESIMS spectrum of compound **10**.

**Fig. S13(G)** IR spectrum of compound **10**.

**Fig. S13(H)** CD spectrum of compound **10**.

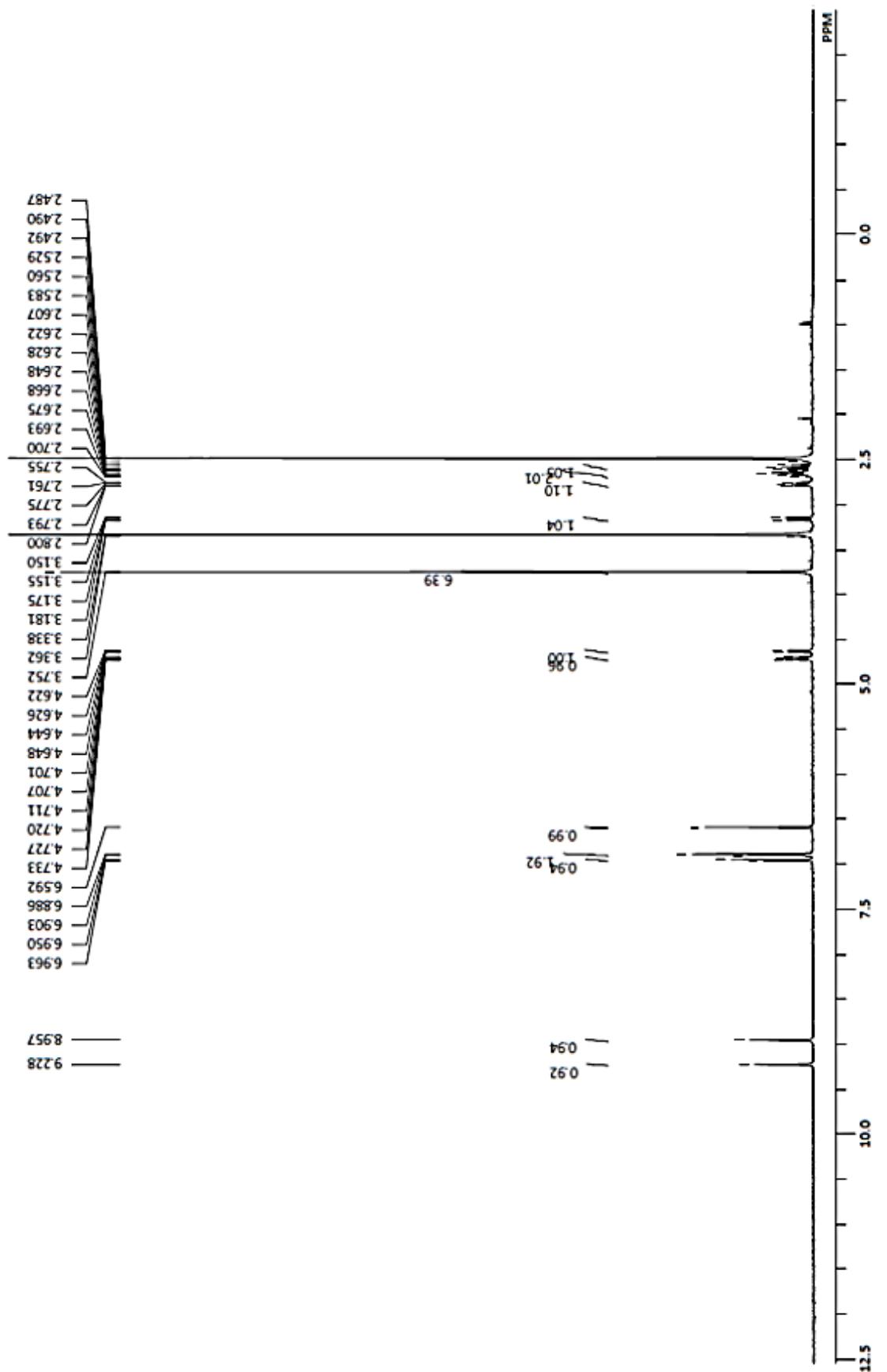
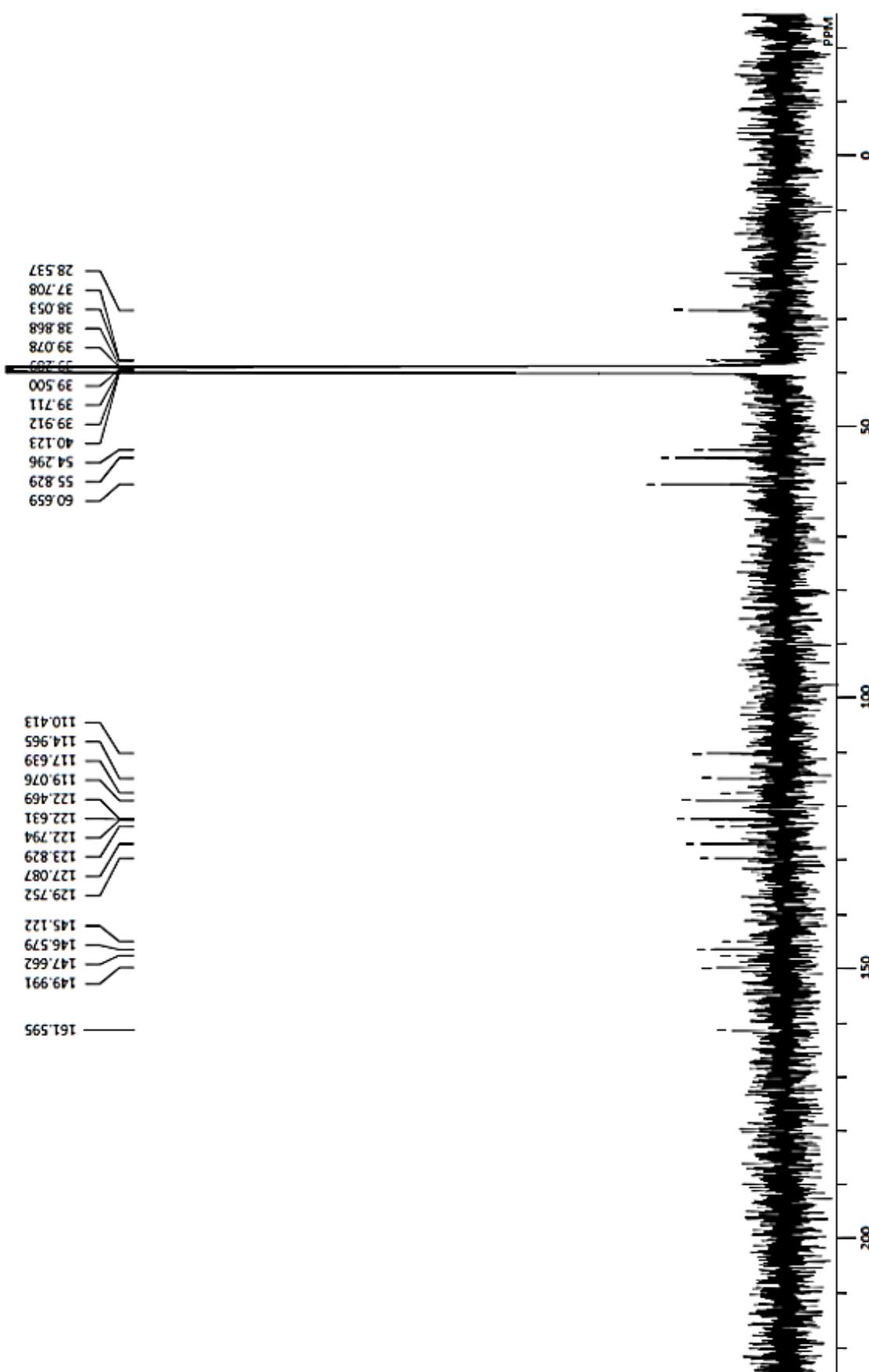


Fig. S13(A)  $^1\text{H}$  NMR spectrum of compound 10 (400 MHz, DMSO- $d_6$ ).



**Fig. S13(B)**  $^{13}\text{C}$  NMR spectrum of compound **10** (100 MHz,  $\text{DMSO}-d_6$ ).

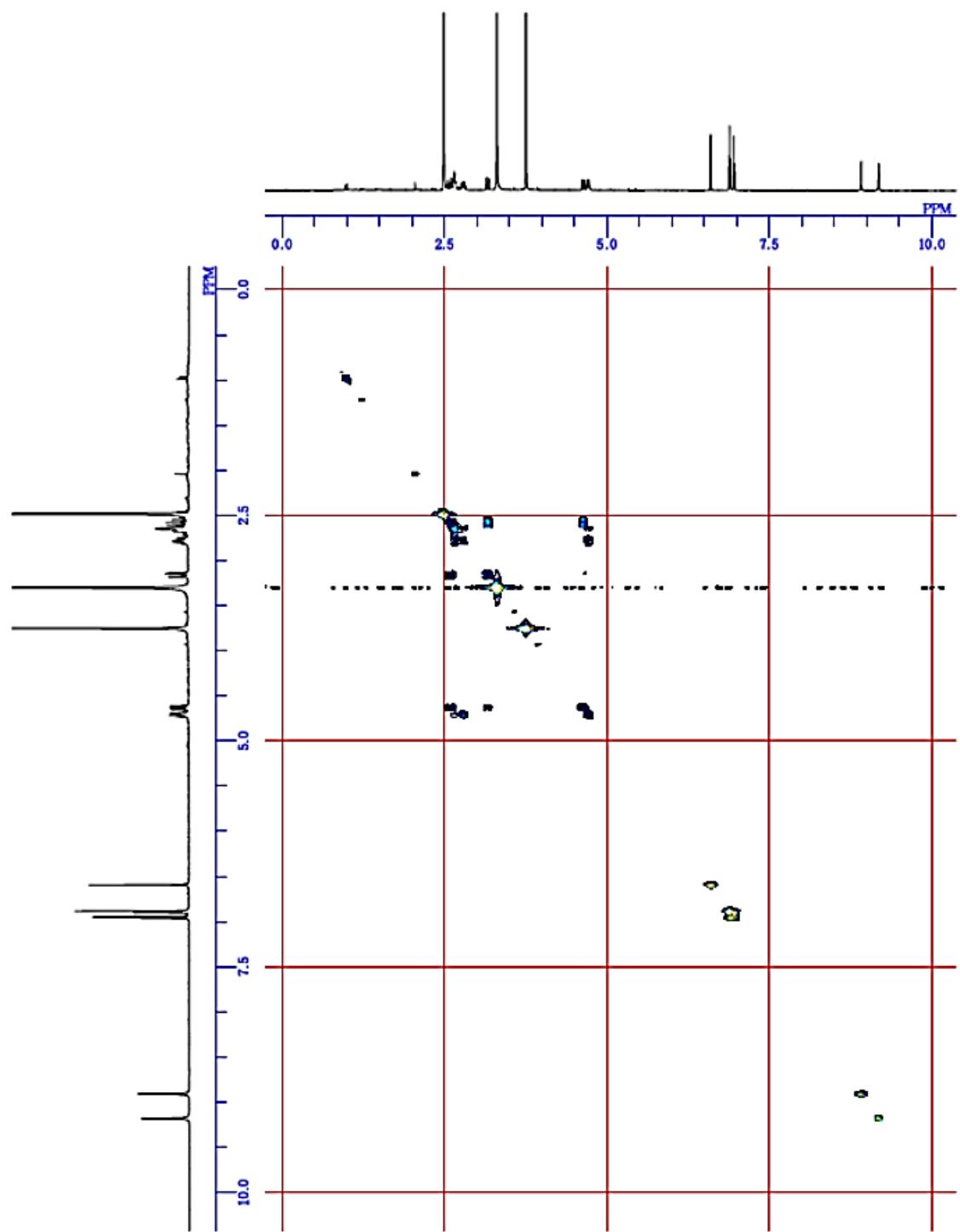
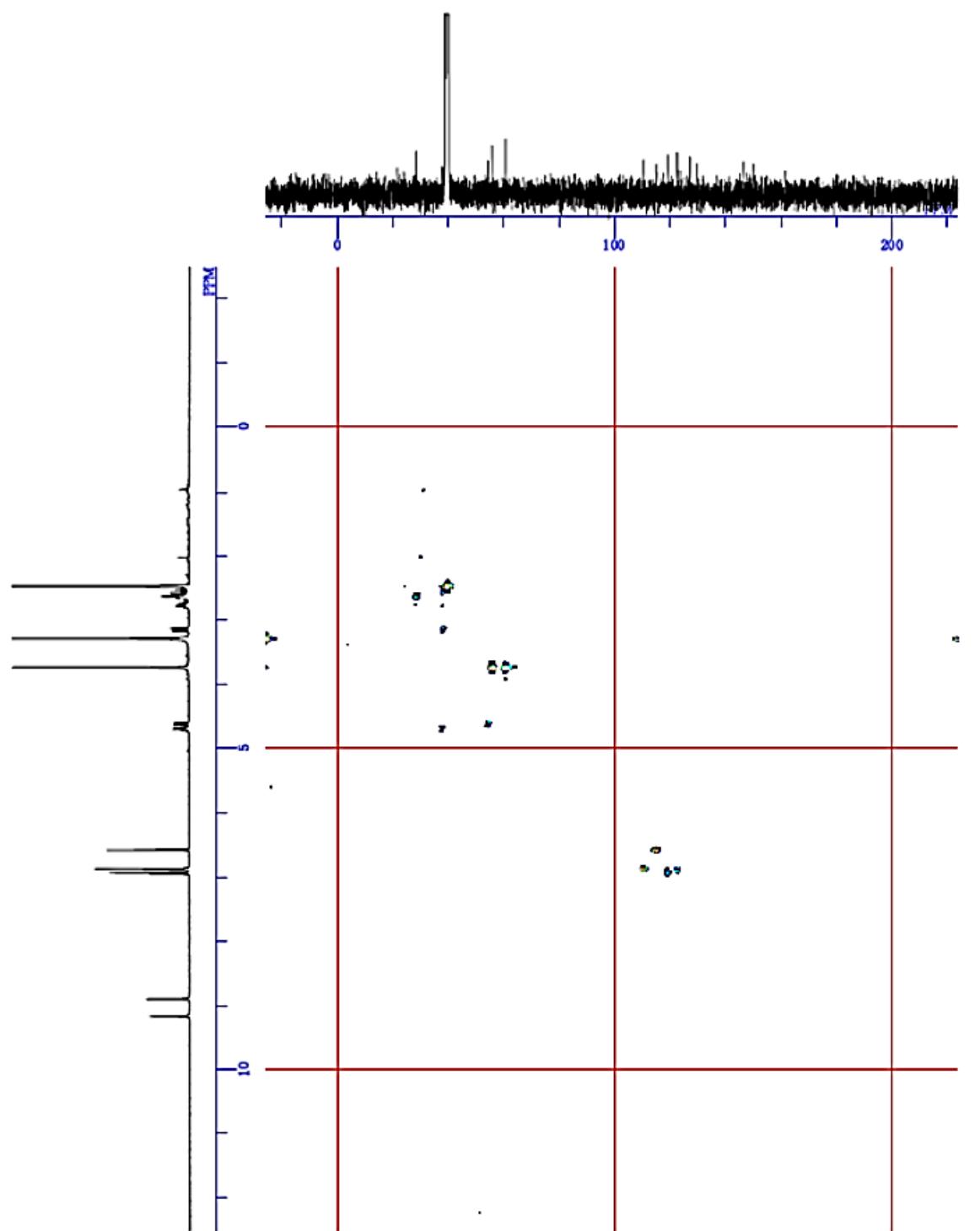


Fig. S13(C) COSY spectrum of compound 10 (400 MHz, DMSO-*d*<sub>6</sub>).



**Fig. S13(D)** HMQC spectrum of compound 10 (400 MHz,  $\text{DMSO}-d_6$ ).

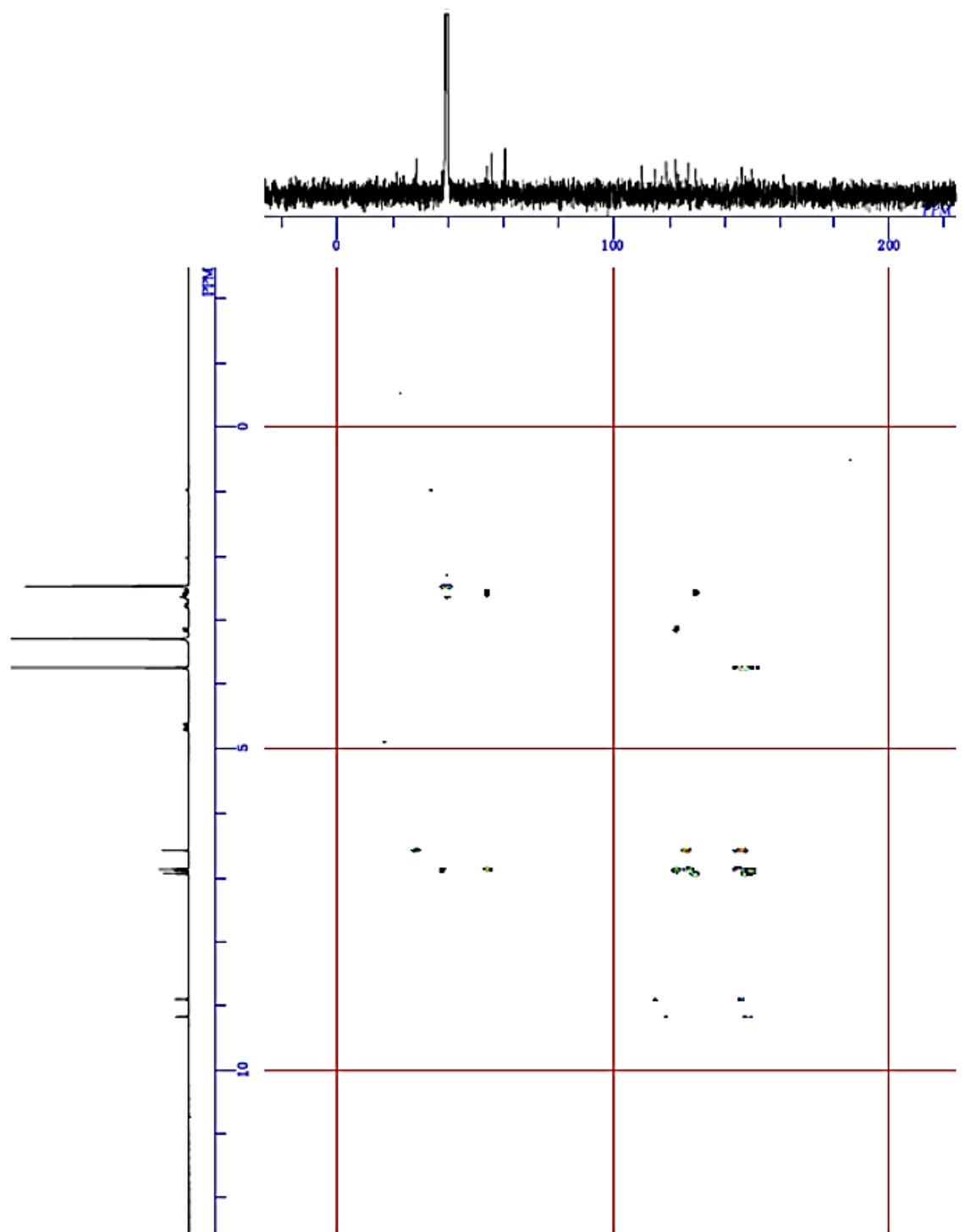


Fig. S13(E) HMQC spectrum of compound 10 (400 MHz,  $\text{DMSO}-d_6$ ).

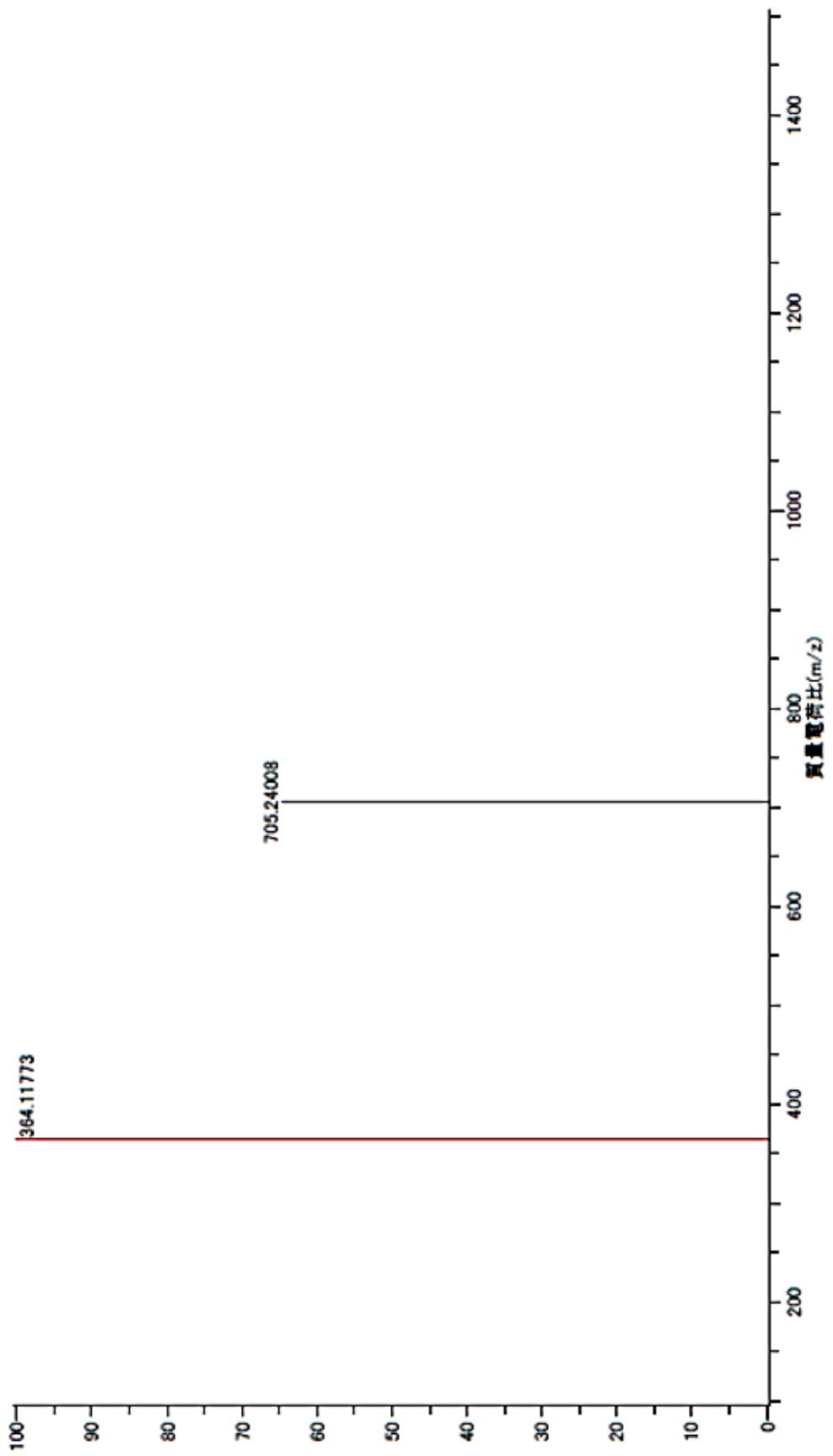


Fig. S13(F) HRESIMS spectrum of compound 10.

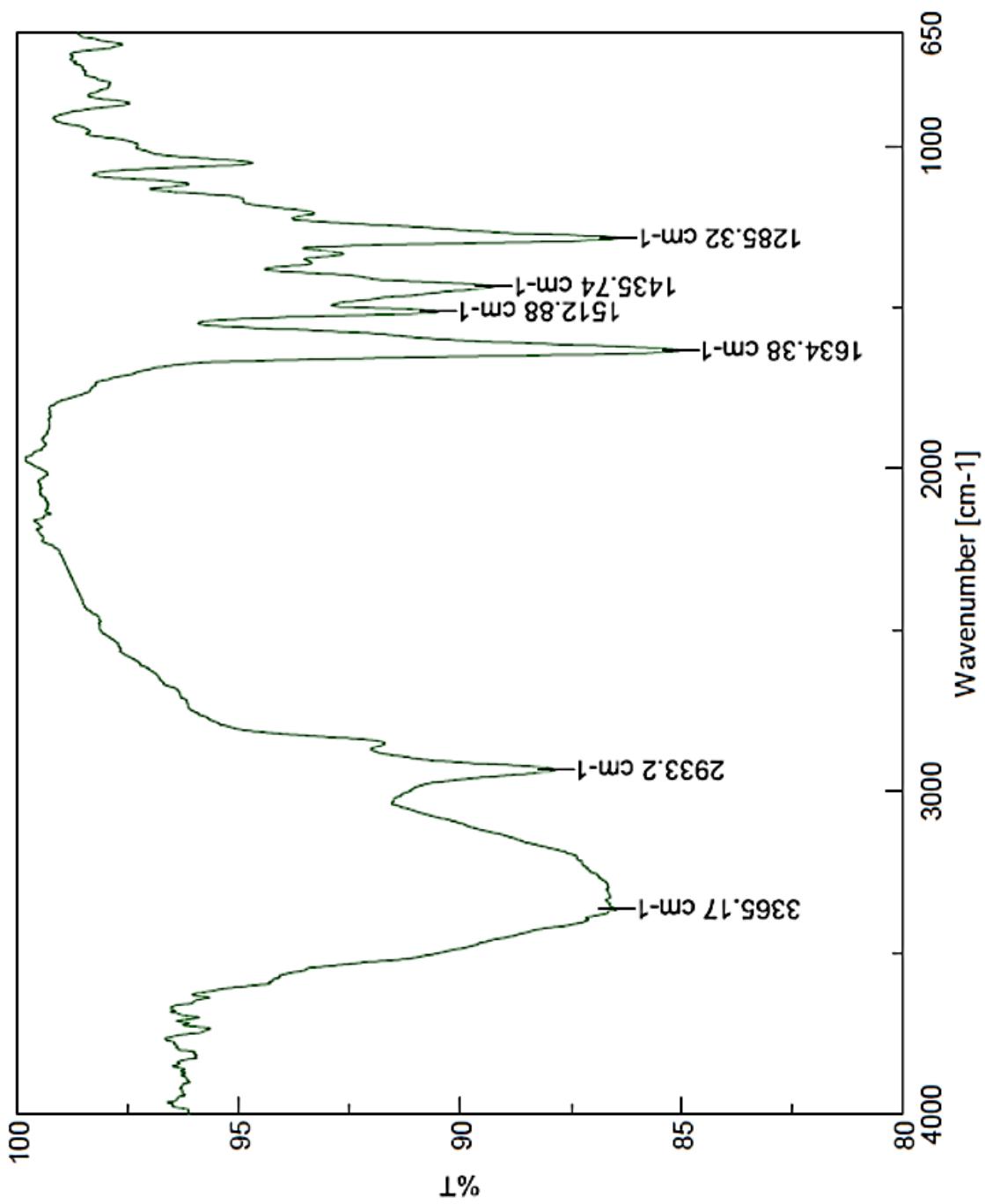


Fig. S13(G) IR spectrum of compound 10.

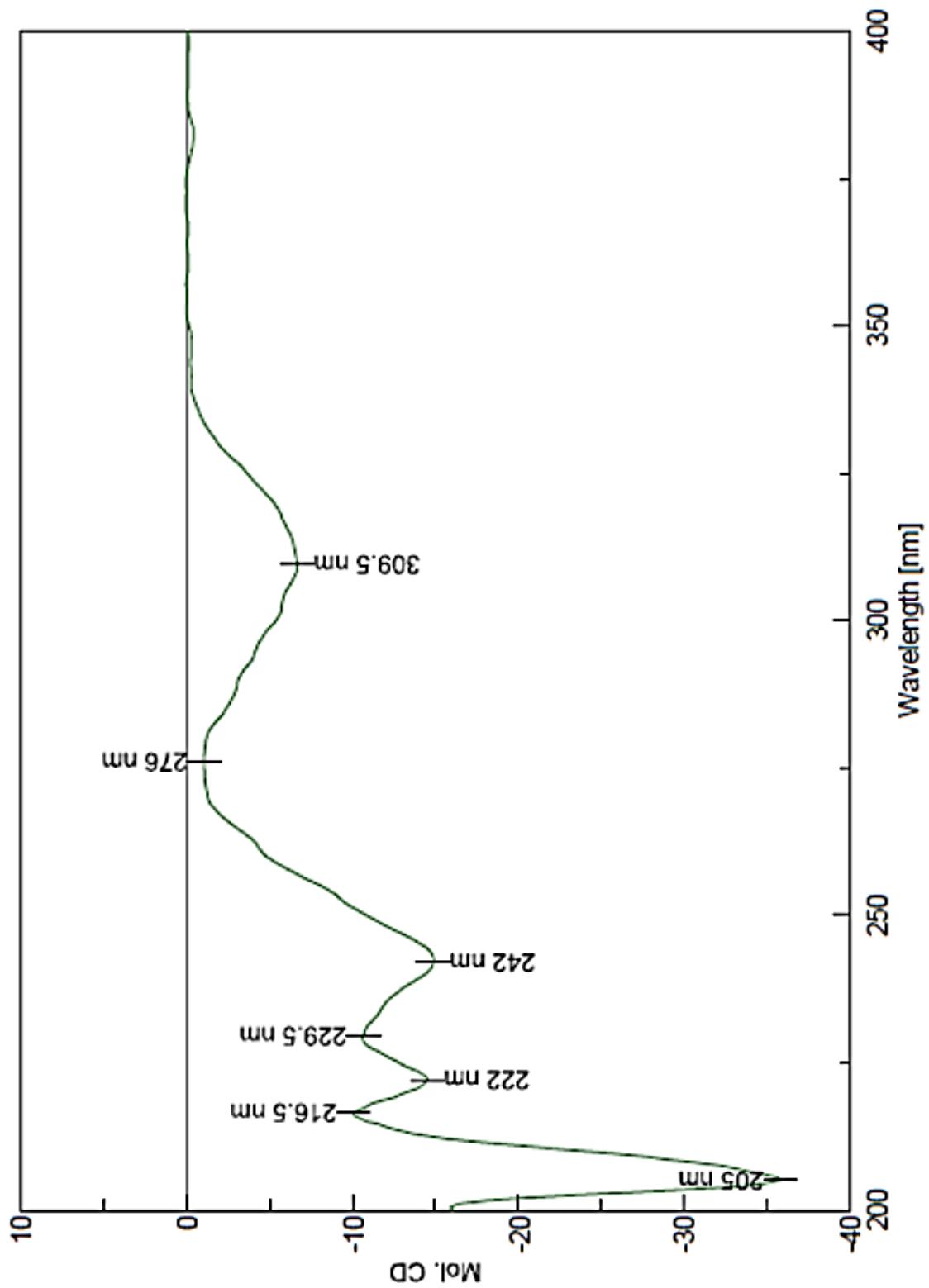


Fig. S13(H) CD spectrum of compound 10.