Supporting Information

Metal-free construction of fused pyrimidines via consecutive C-C and C-N bonds formation in water

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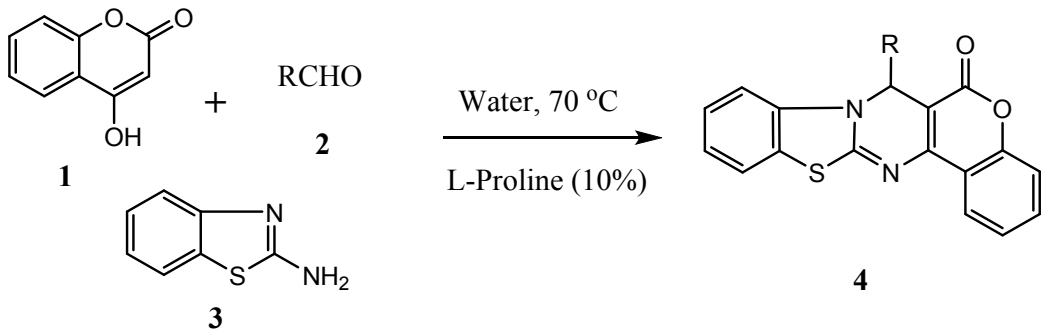
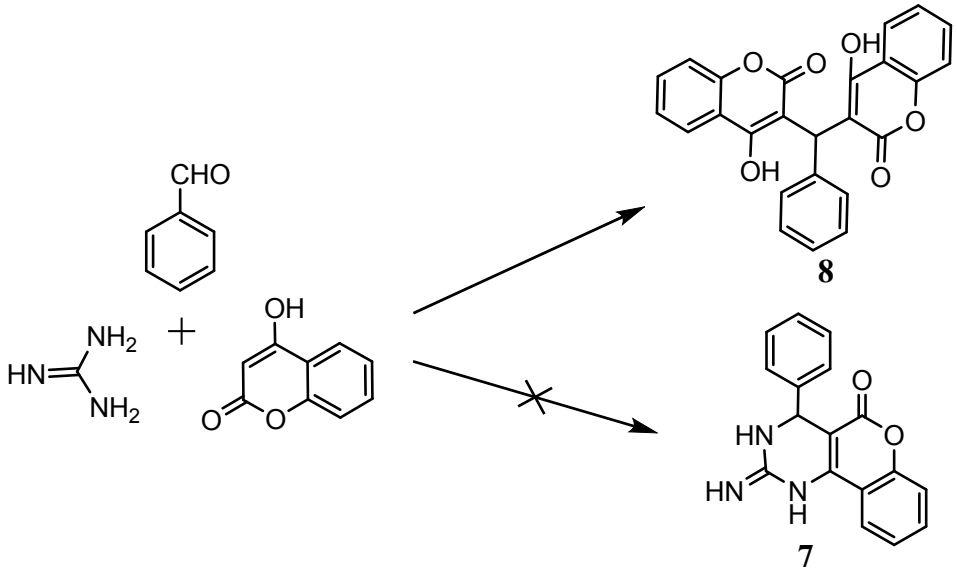
Scheme S1. Synthesis of fused pyrimidines



Scheme S2. Optimization with guanidine



Scheme S3. Proposed reaction mechanism

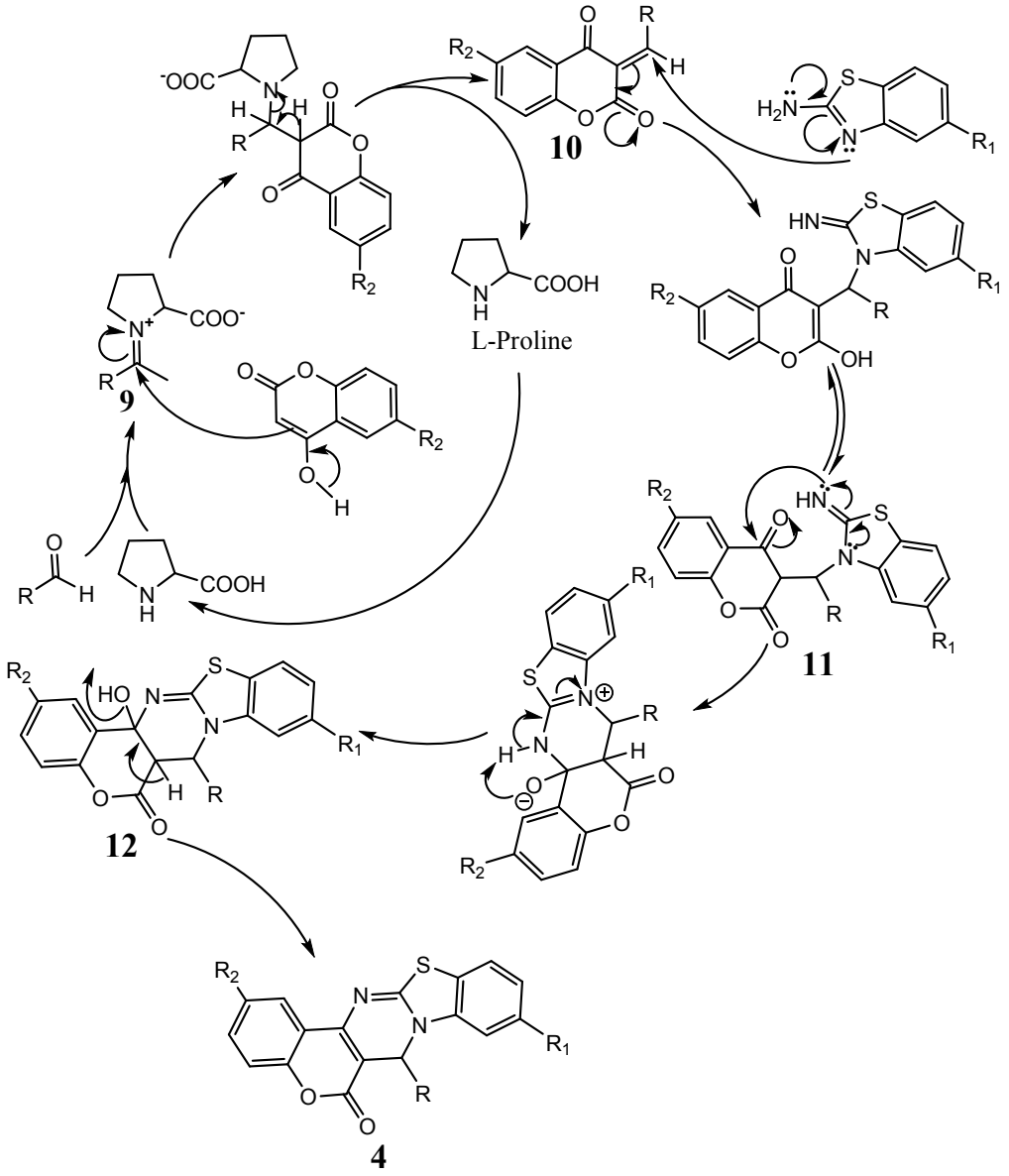
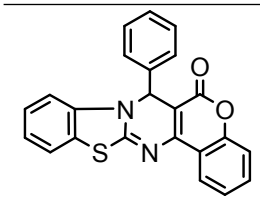


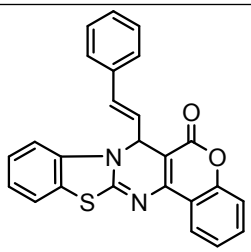
Table S1. Screening of catalysts

Entry	Catalysts	Time (h)	Yield (%)[a]
1	L-proline (2 mol%)	8.0	78
2	L-proline (5 mol%)	4.0	87
3	L-proline (10 mol%)	3.0	91
4	L-proline (15 mol%)	3.0	91
5	*p*-TSA (10 mol%)	10.0	56
6	KCl (10 mol%)	6.5	35
7	TEA (10 mol%)	8.0	74
8	$CaCl_2$ (10 mol%)	10.0	71
9	LiBr (10 mol%)	150	79
10	H_2SO_4 (10 mol%)	160	72
11	Sulphamic acid (10 mol%)	180	76
12	Pyrrolidine	4.0	71
13	Glycine	3.5	69
14	Acetic acid	4.0	81
15	Piperidine	5.0	77
16	Methylamine	5.0	69

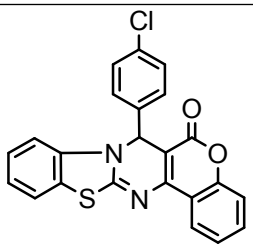
Table S2. Synthesis of library of fused pyrimidines



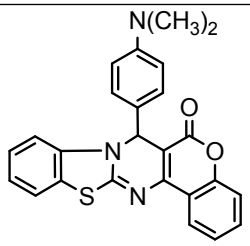
4a
3.0 h, 91%



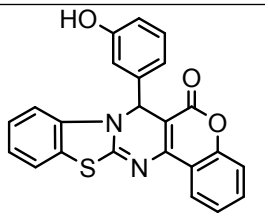
4b
3.0 h, 88%



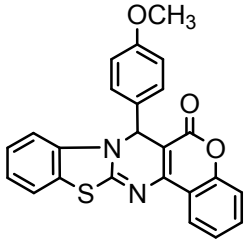
4c
3.0 h, 85%



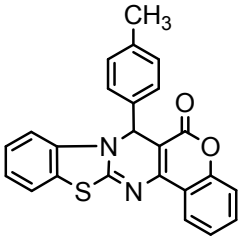
4d
3.5 h, 90%



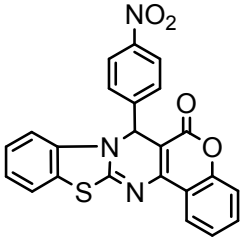
4e
3.0 h, 80%



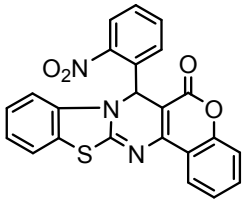
4f
3.0 h, 89%



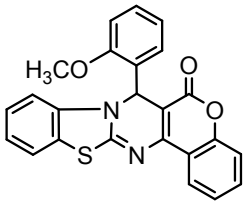
4g
3.0 h, 90%



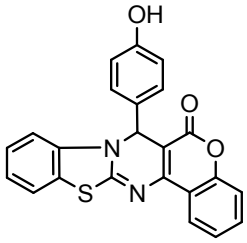
4h
4.0 h, 82%



4i
4.0 h, 80%



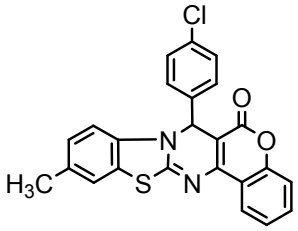
4j
4.0 h, 80%



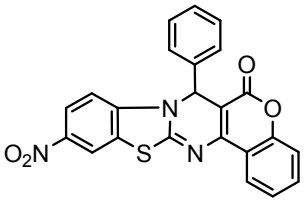
4k
2.5 h, 89%

[a]Reaction conditions: 4-hydroxy coumarin (5 mmol), aldehydes (5 mmol) and 2-aminobenzothiazole (5 mmol) using L-proline (10 mol%) in water at 70 °C.

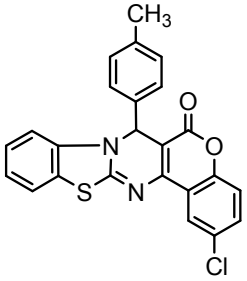
Table S3. Synthesis of fused pyrimidines using substituted benzothioazole and 4-hydroxy coumarin[a]



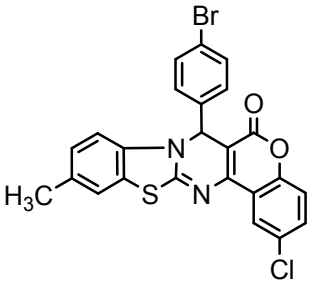
5a
3.0 h, 85%



5b
3.0 h, 86%



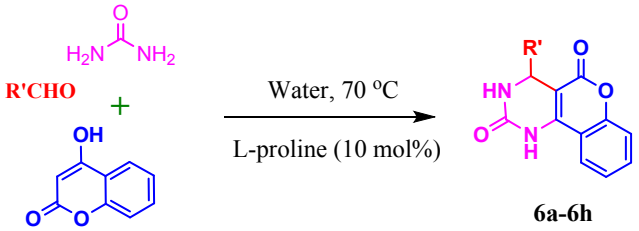
5c
3.0 h, 88%



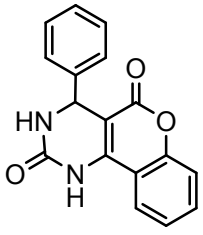
5d
3.5 h, 84%

[a]Reaction conditions: substituted 4-hydroxy coumarin (5 mmol), derived benzaldehyde (5 mmol) and substituted 2-aminobenzothiazole (5 mmol) using L-proline (10 mol%) in water at 70 °C.

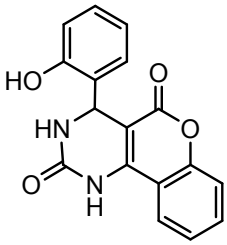
Table S4. Synthesis of library of fused pyrimidines using urea[a]



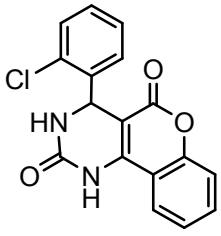
6a-6h



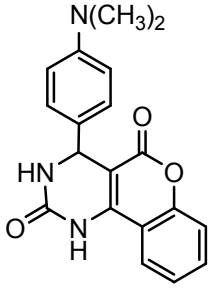
6a
3.0 h, 90%



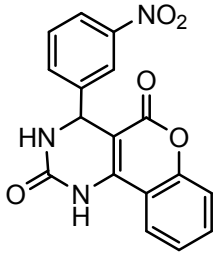
6b
4.5 h, 88%



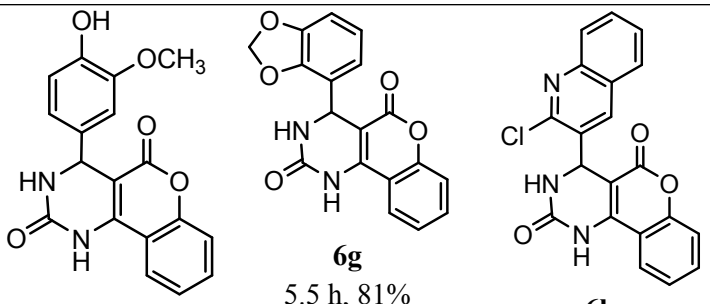
6c
5.0 h, 89%



6d
4.5 h, 83%



6e
4.5 h, 82%



6f
5.0 h, 93%

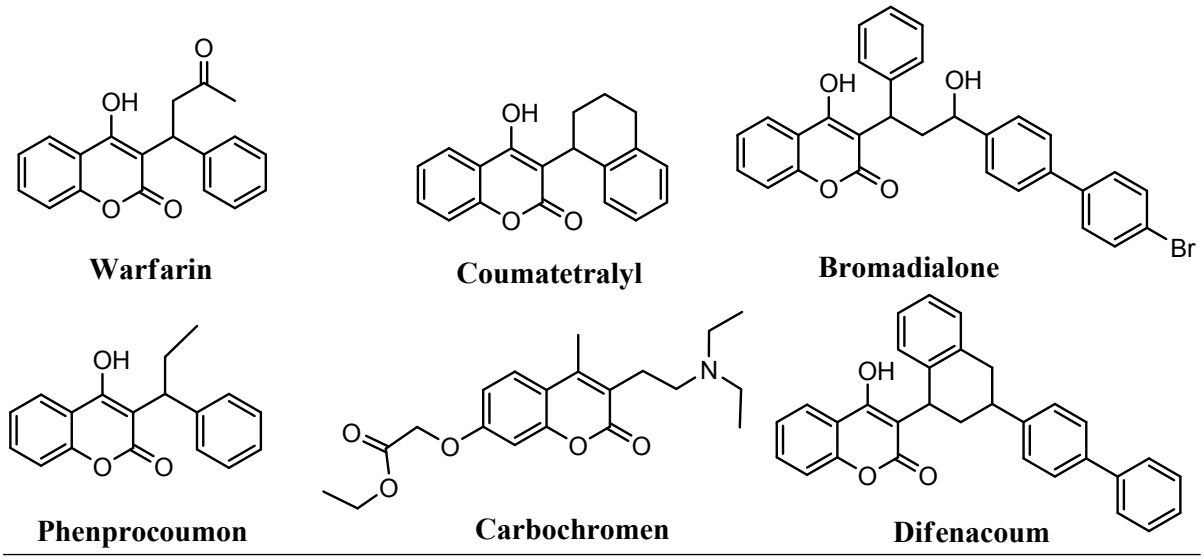
6g
5.5 h, 81%

6h
5.0 h, 83

[a]Reaction conditions: 4-hydroxy coumarin (5 mmol), aldehydes (5 mmol) and urea (5 mmol) using L-proline (10 mol%) in water at 70 °C.

Figure S1. Some drugs with coumarin and pyrimidine core

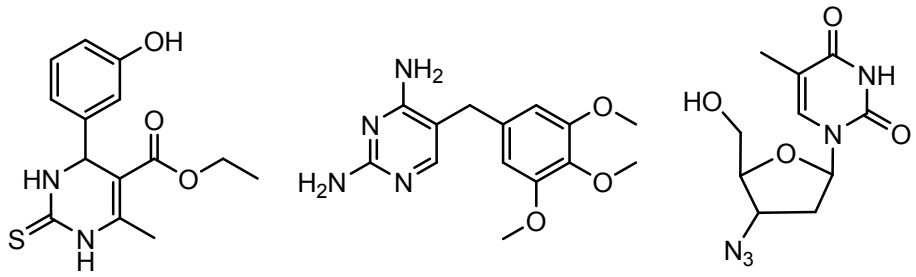
A



Warfarin **Coumatetralyl** **Bromadialone**

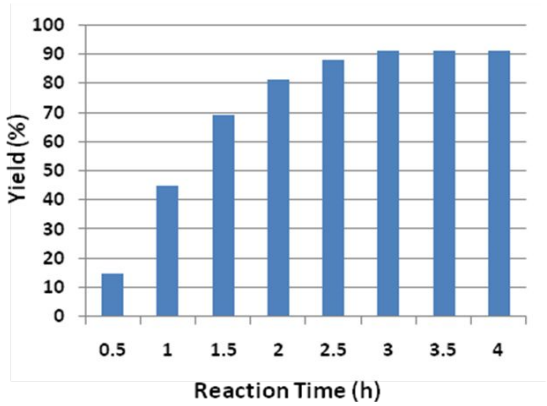
Phenprocoumon **Carbochromen** **Difenacoum**

B



Monastrol **Trimethoprim** **Zidovudine**

Figure S2. Comparison of reaction time with respect to yield



Characterization Data

7-phenylchromeno[4,3-d]benzothiazolo[3,2-a]pyrimidin-6(7H)-one (**4a**)

White powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.23 (s, 1H, -CH), 7.05-7.17 (m, 3H, Ar-H), 7.17 (t, 2H, J = 8.0 Hz, Ar-H), 7.22-7.30 (m, 4H, Ar-H), 7.38-7.51 (m, 4H); ^{13}C NMR (125 MHz, DMSO d_6): 68, 103, 114, 115, 115, 119, 122, 122, 123, 123, 124, 126, 126, 127, 127, 131, 141, 152, 164, 167, 168; ESI-MS: m/z Calculated for $C_{23}H_{14}N_2O_2S$ 382.43 Found $[M+H]^+$ 383.

7-styrylchromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4b**)

Light brown crystal, mp 180-182; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 5.63 (s, 1H, -CH), 6.26 (d, 1H, =CH), 6.70 (d, 1H, =CH), 7.19-7.38 (m, 8H, Ar-H), 7.58 (t, 2H, J = 7.5 Hz, Ar-H), 7.84 (t, 3H, J = 8.0 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 65, 101, 102, 103, 112, 114, 116, 117, 120, 122, 124, 128, 129, 131, 133, 137, 144, 151, 152, 157, 161, 163, 164, 167; ESI-MS: m/z Calculated for $C_{25}H_{16}N_2O_2S$ 408.47 Found $[M+H]^+$ 409.

7-(4-chlorophenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4c**)

Light yellow powder, mp 188-189; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.39 (s, 1H, -CH), 6.94-7.09 (m, 2H, Ar-H), 7.16 (d, 1H, J = 8.0 Hz, Ar-H), 7.21 (t, 1H, J = 7.5 Hz, Ar-H), 7.39-7.62 (m, 4H, J = 8.0 Hz), 7.82 (d, 2H, J = 8.5 Hz, Ar-H), 7.90 (d, 2H, J = 7.0 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 68, 106, 113, 118, 118, 121, 125, 125, 125, 126, 129, 130, 134, 146, 155, 163, 167, 170; ESI-MS: m/z Calculated for $C_{23}H_{13}ClN_2O_2S$ 416.88 Found $[M+H]^+$ 418.

7-(4-dimethylaminophenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4d**)

Reddish brown powder, mp 160-162; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.08 (s, 6H, -N(CH_3)$_2$), 6.26 (s, 1H, -CH), 7.15-7.41 (m, 8H, Ar-H), 7.59 (t, 2H, J = 7.5 Hz, Ar-H), 7.86 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 45, 65, 103, 111, 114, 115, 118, 118, 119, 121, 123, 124, 125, 126, 128, 131, 131, 141, 152, 164, 167; ESI-MS: m/z Calculated for $C_{25}H_{19}N_3O_2S$ 425.50 Found $[M+H]^+$ 426.

7-(3-hydroxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4e**)

Off white powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.20 (s, 1H, -CH), 6.45-6.56 (m, 3H, Ar-H), 6.94 (t, 1H, J = 7.5 Hz, Ar-H), 7.22-7.28 (m, 4H, Ar-H), 7.41-7.51 (m, 4H, Ar-H), 9.26 (br, 1H, OH); ^{13}C NMR (125 MHz, DMSO d_6): 65, 103, 112, 113, 114, 115, 117, 119, 122, 122, 123, 123, 124, 127, 128, 131, 141, 143, 152, 157, 164, 167, 168; ESI-MS: m/z Calculated for $C_{23}H_{14}N_2O_3S$ 398.43 Found $[M+H]^+$ 399.

7-(4-methoxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4f**)

Off white powder, mp 234-236 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.73 (s, 3H, OCH_3), 6.24 (s, 1H, -CH), 6.80 (d, 2H, J = 7.0 Hz, Ar-H), 7.16-7.43 (m, 4H, Ar-H), 7.51 (t, 2H, J = 7.5 Hz, Ar-H), 7.68 (d, 2H, J = 7.0 Hz, Ar-H), 7.79 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 56, 66, 103, 118, 121, 125, 128, 133, 133, 133, 134, 138, 141, 142, 145, 150, 152, 164, 166; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_3S$ 412.46 Found $[M+H]^+$ 413.

7-(4-methylphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4g**)

Yellow powder, mp >250 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.80 (s, 3H, CH_3), 6.24 (s, 1H, -CH), 7.19-7.40 (m, 8H, Ar-H), 7.51 (t, 2H, J = 7.5 Hz, Ar-H), 7.80 (d, 2H, J = 7.0 Hz, Ar-H);

^{13}C NMR (125 MHz, DMSO d_6): 27, 65, 102, 111,116, 119, 119, 123, 123, 124, 124, 128, 131, 140, 145, 152, 154, 164, 167; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_2S$ 396.46 Found $[M]^+$ 397.

7-(4-nitrophenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4h**)

Off white powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.17 (s, 1H, -CH), 7.20-7.25 (m, 5H, Ar-H), 7.29 (d, 2H, J = 7.0 Hz, Ar-H), 7.50 (t, 2H, J = 7.5 Hz, Ar-H), 7.81 (d, 2H, J = 7.5 Hz, Ar-H), 8.13 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 67, 103, 111, 115, 118, 122, 123, 123, 124, 124, 126, 128, 128, 131, 144, 152, 161, 164, 167; ESI-MS: m/z Calculated for $C_{23}H_{13}N_3O_4S$ 427.43 Found $[M+H]^+$ 429.

7-(2-methylphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4i**)

Off white powder, mp >250 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.61 (s, 3H, CH_3), 6.37 (s, 1H, -CH), 7.21-7.25 (m, 4H, Ar-H), 7.29 (t, 2H, J = 7.0 Hz, Ar-H), 7.46 (t, 2H, J = 7.5 Hz, Ar-H), 7.86 (d, 2H, J = 7.5 Hz, Ar-H), 8.13 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 38, 67, 103, 115, 118, 123, 123, 124, 124, 128, 128, 130, 131, 140, 144, 150, 152, 164, 166; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_2S$ 396.46 Found $[M+H]^+$ 397.

7-(2-methoxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4j**)

Off white powder, mp 234-236 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.73 (s, 3H, OCH_3), 6.24 (s, 1H, -CH), 6.78 (d, 2H, J = 7.0 Hz, Ar-H), 7.19-7.31 (m, 4H, Ar-H), 7.53 (t, 2H, J = 7.5 Hz, Ar-H), 7.71 (d, 2H, J = 7.0 Hz, Ar-H), 7.79 (d, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 55, 66, 106, 115, 119, 122, 122, 123, 123, 126, 131, 134, 135, 143, 149, 152, 156, 167, 169; ESI-MS: m/z Calculated for $C_{24}H_{16}N_2O_3S$ 412.46 Found $[M+H]^+$ 413.

7-(4-hydroxyphenyl)chromeno[4,3-*d*]benzothiazolo[3,2-*a*]pyrimidin-6(7*H*)-one (**4k**)

Off white powder, mp 200-202 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.16 (s, 1H, -CH), 6.54 (d, 1H, J = 7.5 Hz, Ar-H), 6.86 (d, 1H, J = 7.5 Hz, Ar-H), 7.19-7.26 (m, 4H, Ar-H), 7.35- 7.51 (m, 4H, Ar-H), 7.80 (d, 2H, J = 7.5 Hz, Ar-H), 8.91 (br, 1H, OH); ^{13}C NMR (125 MHz, DMSO d_6): 65, 101, 109, 111, 112, 113, 115, 117, 120, 120, 120, 121, 124, 125, 128, 138, 141, 149, 159, 154, 162, 164, 166, 167; ESI-MS: m/z Calculated for $C_{23}H_{14}N_2O_3S$ 398.43 Found $[M+H]^+$ 399.

7-(4-chloroyphenyl)-11-methylbenzo[4,5]thiazolo[3,2-*a*]chromeno[4,3-d]pyrimidin-6(7*H*)-one (**5a**)

Off white powder, mp 184-186 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.29 (s, 3H, CH_3), 6.29 (s, 1H, -CH), 7.14 (d, 2H, J = 7.5 Hz, Ar-H), 7.24-7.35 (m, 4H, Ar-H), 7.55-7.59 (m, 2H, Ar-H), 7.88 (d, 2H, J = 7.5 Hz, Ar-H), 7.93 (d, 1H, J = 8.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 23, 67, 104, 116, 116, 117, 117, 123, 128, 129, 129, 129, 130, 131, 132,138, 139, 152, 164, 166; ESI-MS: m/z Calculated for $C_{24}H_{15}ClN_2O_2S$ 430.91 Found $[M+H]^+$ 431.

11-nitro-7-phenyl-11-methylbenzo[4,5]thiazolo[3,2-*a*]chromeno[4,3-d]pyrimidin-6(7*H*)-one (**5b**)

Off white powder, mp 215-217 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.37 (s, 1H,-CH), 7.27 (t, 2H, J = 8.0 Hz, Ar-H), 7.33 (d, 2H, J = 8.5 Hz, Ar-H), 7.40 (d, 2H, J = 8.5 Hz, Ar-H), 7.56 (t, 2H, J = 8.0 Hz, Ar-H), 7.84 (d, 2H, J = 8.0 Hz, Ar-H), 8.09 (d, 2H, J = 9.0 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 69, 103, 111, 115, 118, 122, 122, 123, 127, 130, 131, 139, 145, 149, 151, 157, 164, 166; ESI-MS: m/z Calculated for $C_{23}H_{13}N_3O_4S$ 427.43 Found $[M]^+$ 427.5.

2-chloro-7-(p-tolyl)benzo[4,5]thiazolo[3,2-a]chromeno[4,3-d]pyrimidin-6(7H)-one **(5c)**

Off white powder, mp 225-227 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.39 (s, 3H, CH_3), 6.30 (s, 1H, -CH), 6.89-6.94 (dd, 2H, J = 8.0, 8.5 Hz, Ar-H), 7.06 (t, 2H, J = 8.0 Hz), 7.25-7.36 (m, 4H, Ar-H), 7.70 (d, 1H, J = 7.5 Hz, Ar-H), 8.10 (d, 2H, J = 8.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 21, 69, 102, 117, 118, 121, 121, 121, 126, 127, 128, 129, 129, 129, 130, 1132, 134, 143, 150, 151, 161, 163, 167; ESI-MS: m/z Calculated for $C_{24}H_{15}ClN_2O_2S$ 430.90 Found $[M]^+$ 430.9.

7-(4-bromophenyl)-2-chloro-11-methylbenzo[4,5]thiazolo[3,2-a]chromeno[4,3-d]pyrimidin-6(7H)-one **(5d)**

Off white powder, mp >250 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 2.23 (s, 3H, CH_3), 6.29 (s, 1H, -CH), 7.09-7.23 (m, 8H, Ar-H), 7.32 (t, 2H, J = 7.5 Hz, Ar-H); ^{13}C NMR (125 MHz, DMSO d_6): 22, 66, 104, 107, 113, 117, 125, 126, 128, 129, 130, 130, 134, 143, 151, 155, 162, 164; ESI-MS: m/z Calculated for $C_{24}H_{14}BrClN_2O_2S$ 509.80 Found $[M]^+$ 509.8.

4-Pheny-3,4-dihydro-1*H*-chromeno[4,3-*d*]pyrimidine-2,5-dione **(6a)**

Off white powder, mp 160-162 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.36 (s, 1H, -CH), 7.09-7.39 (m, 9H, Ar-H), 7.60 (s, 1H, NH), 7.90 (s, 1H, NH); ^{13}C NMR (125 MHz, DMSO d_6): 36 (C-1), 103 (C-2), 115 (C-13), 116 (C-17), 121 (C-14), 122 (C-15), 124 (C-16), 125 (C-6 and 10), 126 (C-8), 126 (C-7), 128 (C-9), 131 (C-5), 140 (C-12), 152 (C-3), 164 (C-4), 165 (C-11); ESI-MS: m/z Calculated for $C_{17}H_{12}N_2O_3$ 292.29 Found $[M]^+$ 292.3; C, H and N analyses Calculated for C 69.86, H 4.14, N 9.58, Found C 70.02, H 4.18, N 9.69.

4-(2-Hydroxyhenyl)-3,4-dihydro-1*H*-chromeno[4,3-*d*]pyrimidine-2,5-dione **(6b)**

Off white powder, mp 240-242 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.25 (s, 1H, -CH), 6.49-6.56 (m, 2H, Ar-H), 6.79 (d, 2H, J = 8.0 Hz, Ar-H), 6.98 (t, 1H, J = 7.5 Hz, Ar-H), 7.28-7.34 (m, 2H, Ar-H), 7.56 (t, 1H, J = 7.0 Hz, Ar-H), 7.67 (1, 1H, -NH), 7.88 (s, 1H, -NH), 9.66 (s, 1H, OH); ^{13}C NMR (125 MHz, DMSO d_6): 35 (C-1), 104 (C-2), 111 (C-13), 112 (C-17), 113 (C-14), 115 (C-15), 117 (C-16), 117 (C-7), 123 (C-8), 129 (C-9), 131 (C-10), 141 (C-5), 152 (C-6), 154 (C-12), 157 (C-3), 164 (C-4), 165 (C-11); ESI-MS: m/z Calculated for $C_{17}H_{12}N_2O_4$ 308.29 Found $[M]^+$ 309; C, H and N analyses Calculated for C 66.23, H 3.92, N 9.09, Found C 66.32, H 3.99, N 9.15.

4-(2-Chlorophenyl)-3,4-dihydro-1*H*-chromeno[4,3-*d*]pyrimidine-2,5-dione **(6c)**

Off white powder, mp 202-204 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.29 (s, 1H, CH), 7.18 (d, 2H, J = 8.5 Hz, Ar-H), 7.50-7.38 (m, 6H, Ar-H), 7.51 (s, 1H, NH), 7.85 (s, 1H, -NH); ^{13}C NMR (125 MHz, DMSO d_6): 35 (C-1), 105 (C-2), 117 (C-13), 117 (C-17), 123 (C-14), 128 (C-15), 128 (C-16), 129 (C-7), 131 (C-9), 131 (C-10), 132 (C-5), 137 (C-8), 139 (C-12), 152 (C-3), 164 (C-4), 165 (C-11); ESI-MS: m/z Calculated for $C_{17}H_{11}ClN_2O_3$ 326.73 Found $[M]^+$ 326.7; C, H and N analyses Calculated for C 62.49, H 3.39, N 8.57, Found C 62.32, H 3.49, N 8.65.

4-(4-Dimethylamino phenyl)-3,4-dihydro-1*H*-chromeno[4,3-*d*]pyrimidine-2,5-dione **(6d)**

Off white powder, mp 235-237 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.11 (s, 6H, $N(CH_3)_2$), 6.27 (s, 1H, -CH), 7.21-7.28 (m, 8H, Ar-H), 7.51 (s 1H, -NH), 7.79 (s, 1H, -NH); ^{13}C NMR (125 MHz, DMSO d_6): 35 (C-1), 45 ($N(CH_3)_2$), 103 (C-2), 111 (C-13), 115 (C-17), 119 (C-14), 122 (C-15 and 16), 124 (C-8 and 5), 128 (C-7 and 9), 131 (C-6 and 10), 131 (C-12), 152 (C-3), 164 (C-4), 167 (C-11); ESI-MS: m/z Calculated for $C_{19}H_{17}N_3O_3$ 335.36 Found $[M]^+$ 336; C, H and N analyses Calculated for C 68.05, H 5.11, N 12.53, Found C 68.12, H 5.22, N 12.61.

4-(3-Nitrophenyl)-3,4-dihydro-1*H*-chromeno[4,3-*d*]pyrimidine-2,5-dione **(6e)**

Off white powder, mp 172-174 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 6.37 (s, 1H,-CH), 7.27-7.33 (m, 4H, Ar-H), 7.38 (d, 2H, J = 8.5 Hz, Ar-H), 7.56 (t, 2H, J = 8.0 Hz, Ar-H), 7.84 (s, 1H, -NH), 8.09 (s, 1H, -NH); ^{13}C NMR (125 MHz, DMSO d_6): 36 (C-1), 103 (C-2), 115 (C-13), 118 (C-17), 123 (C-14), 123 (C-15), 124 (C-16), 124 (C-5), 128 (C-6), 130 (C-10), 131 (C-7 and 9), 145 (C-8), 150 (C-12), 152 (C-3), 164 (C-4), 166 (C-11); ESI-MS: m/z Calculated for $C_{17}H_{11}N_3O_5$ 337.29 Found $[M]^+$ 337.3; C, H and N analyses Calculated for C 60.54, H 3.29, N 12.46, Found C 60.70, H 3.41, N 12.29.

4-(4-Hydroxy-3-methoxyphenyl)-3,4-dihydro-1*H*-chromeno[4,3-*d*]pyrimidine-2,5-dione **(6f)**

Off white powder, mp 208-210 ^{o}C; ^{1}H NMR (500 MHz, DMSO d_6): δ_H 3.72 (s, 3H, OCH_3), 6.39 (s, 1H, -CH), 7.16-7.39 (m, 7H, Ar-H), 7.60 (s, 1H, -NH), 7.93 (s, 1H, -NH), 10.20 (br, 1H, OH); ^{13}C NMR (125 MHz, DMSO d_6): 35 (C-1), 56 (OCH_3), 104 (C-2), 116 (C-13), 117 (C-17), 123 (C-14), 123 (C-15), 124 (C-16), 125 (C-8), 126 (C-9), 128 (C-7), 129 (C-10), 130 (C-6), 132 (C-5), 139 (C-12), 152 (C-3), 164 (C-4), 166 (C-11); ESI-MS: m/z Calculated for $C_{18}H_{14}N_2O_5$ 338.31 Found $[M]^+$ 338.3; C, H and N analyses Calculated for C 63.90, H 4.17, N 8.28, Found C 63.76, H 4.31, N 8.11.

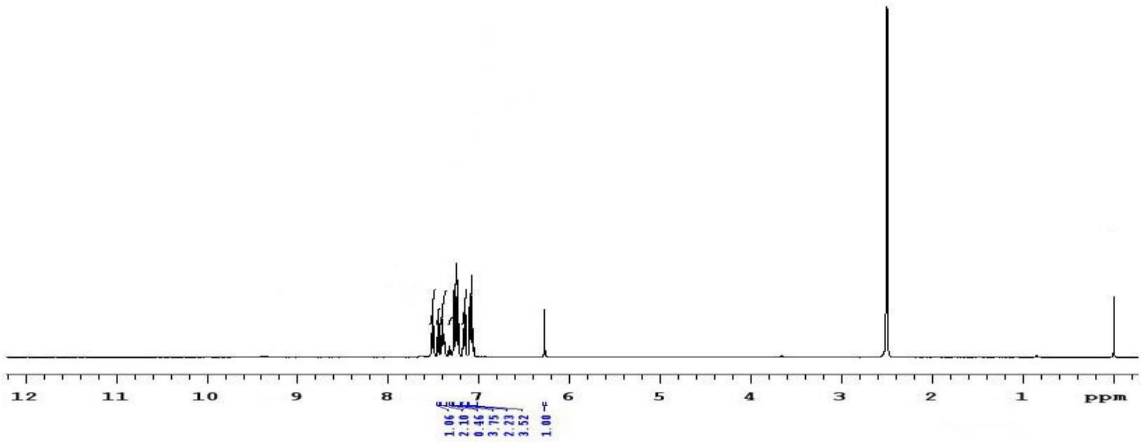


Figure S3. ^{1}H NMR spectra of **4a**

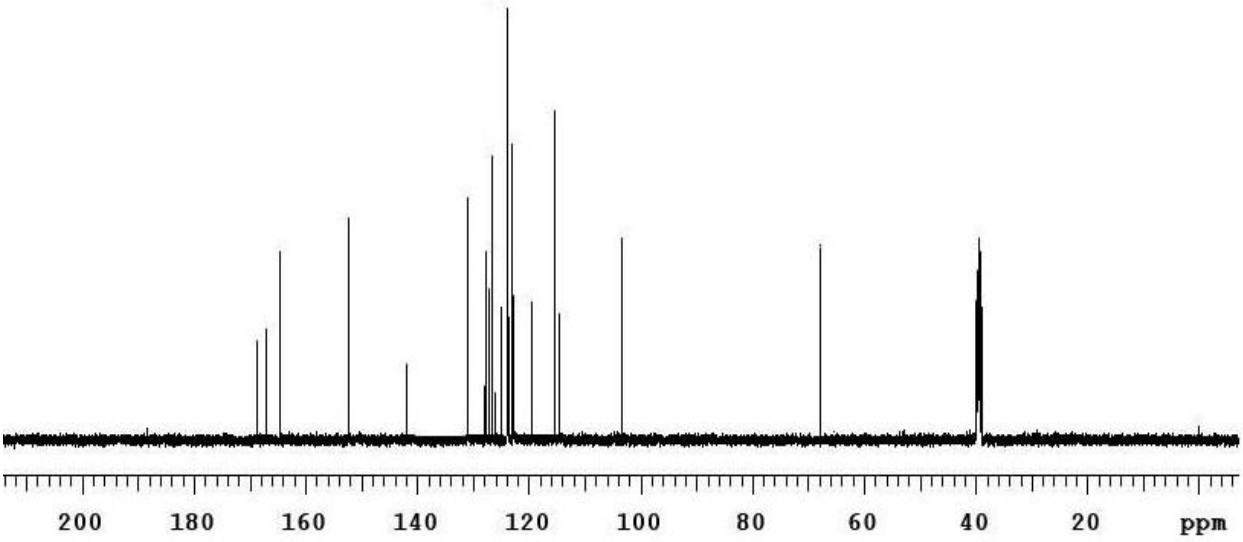


Figure S4. ^{13}C NMR spectra of **4a**

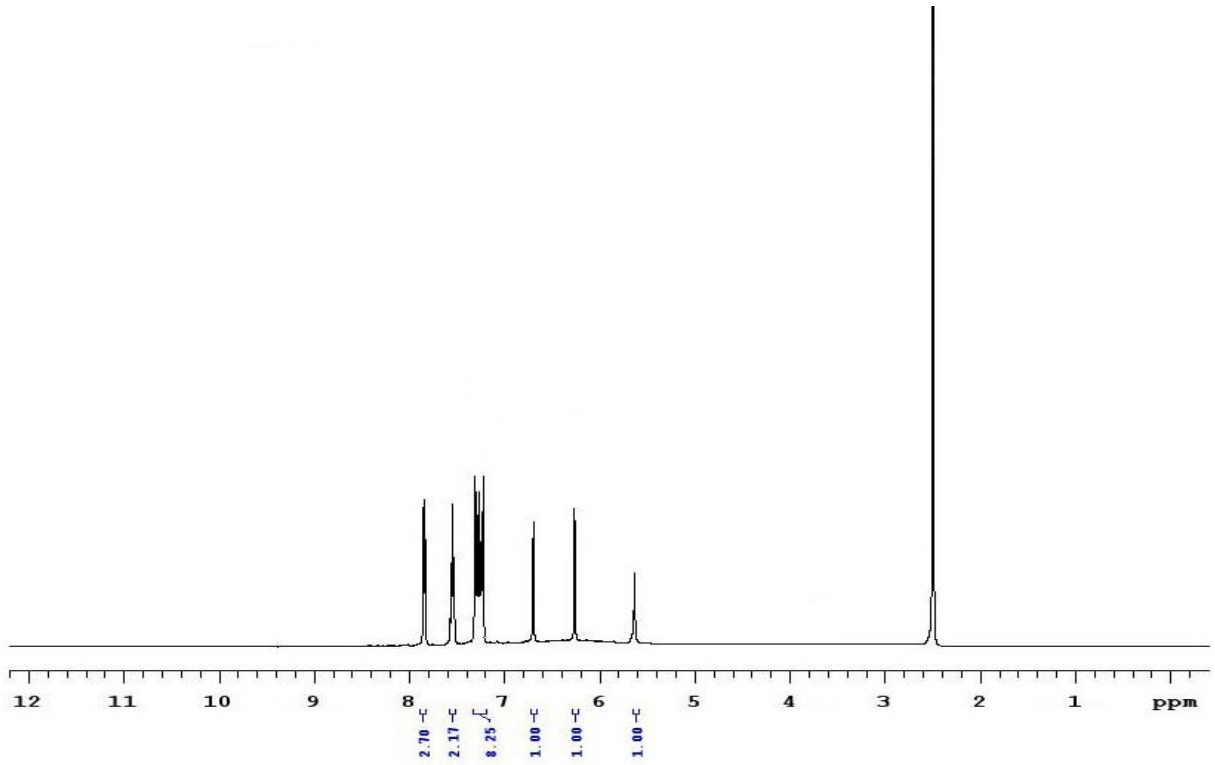


Figure S5. ^{1}H NMR spectra of **4b**

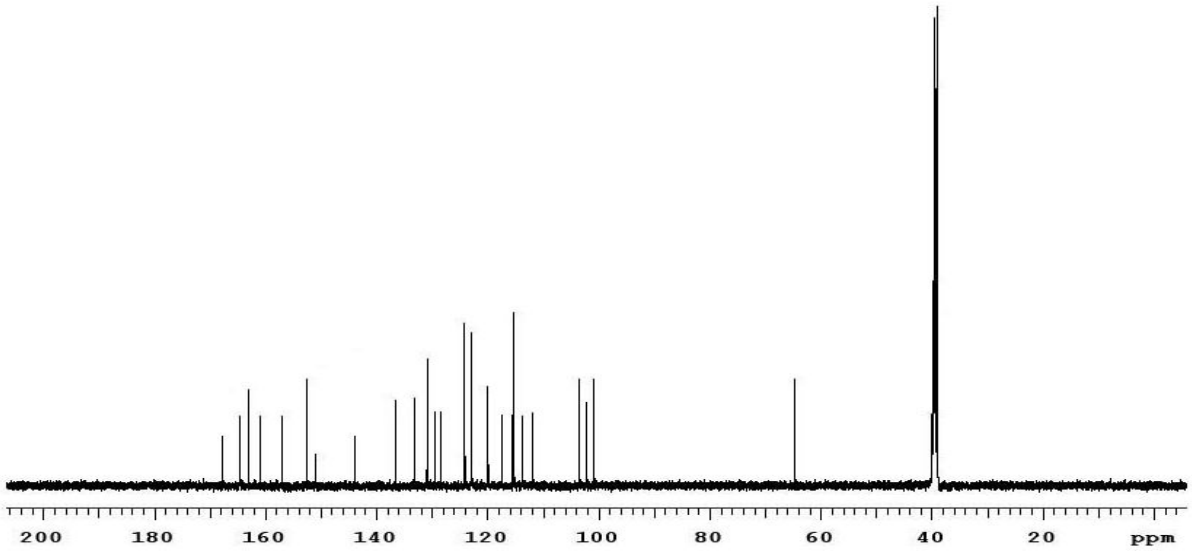


Figure S6. ^{13}C NMR spectra of **4b**

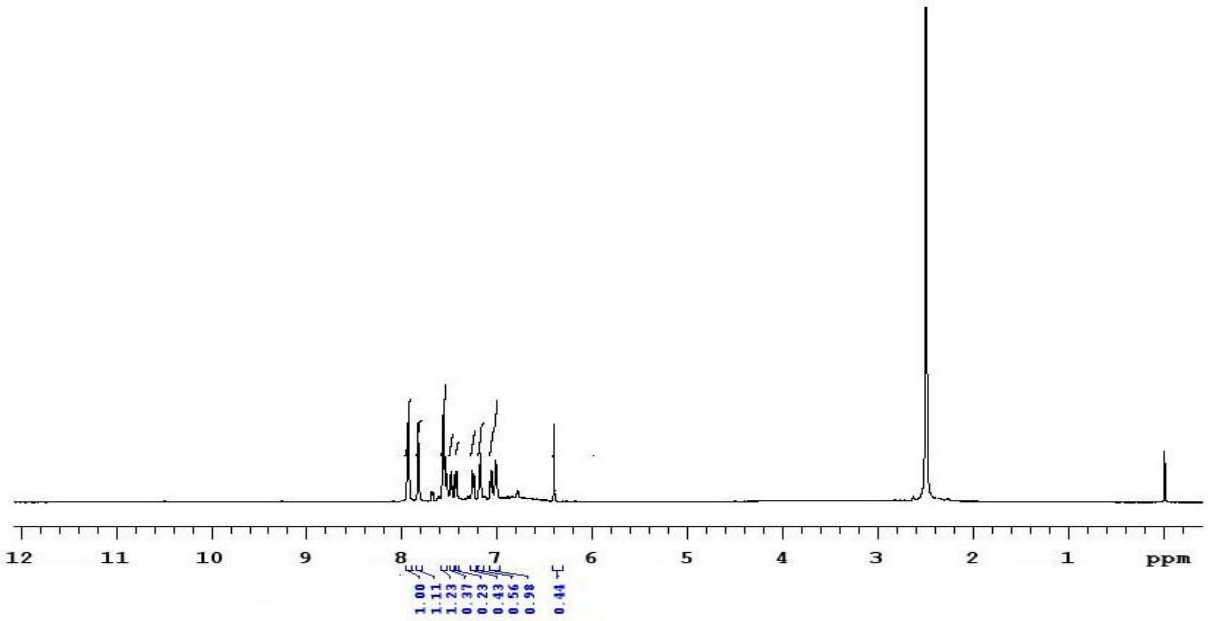


Figure S7. ^{1}H NMR spectra of **4c**

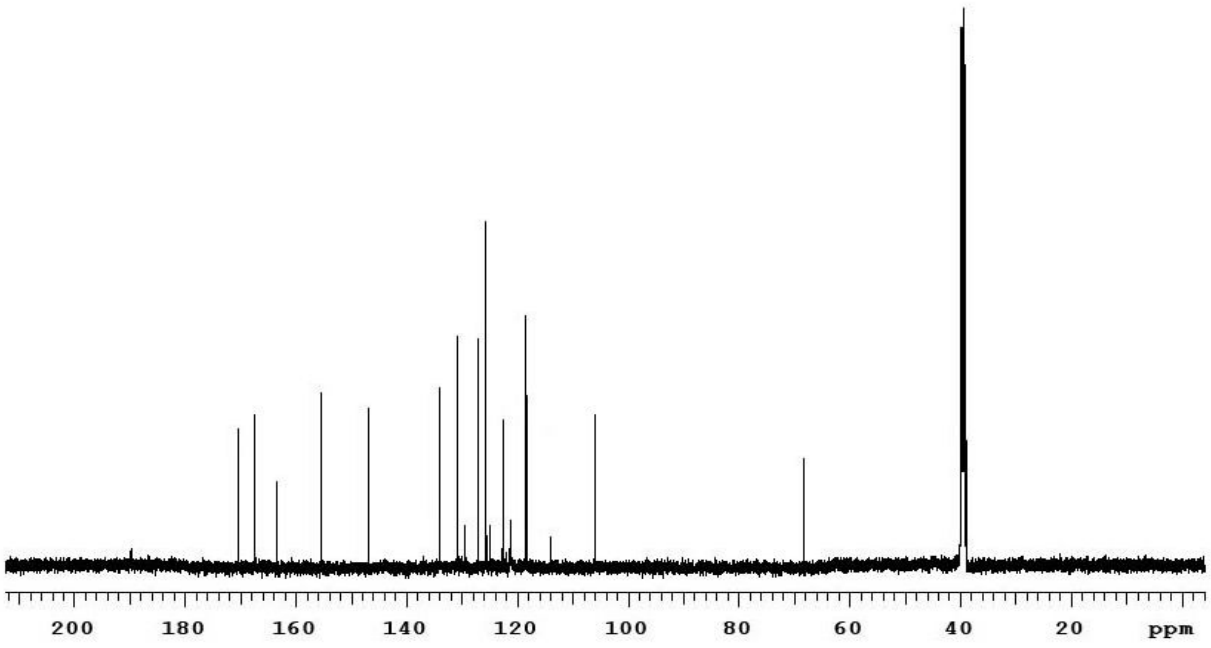


Figure S8. ^{13}C NMR spectra of **4c**

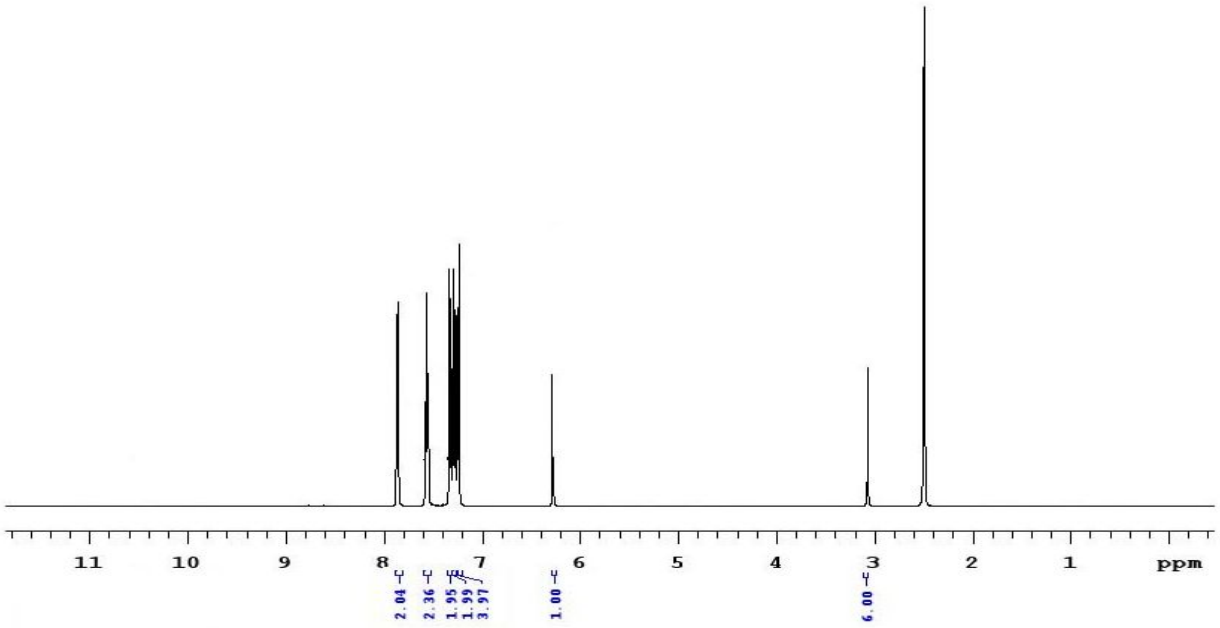


Figure S9. ^{1}H NMR spectra of **4d**

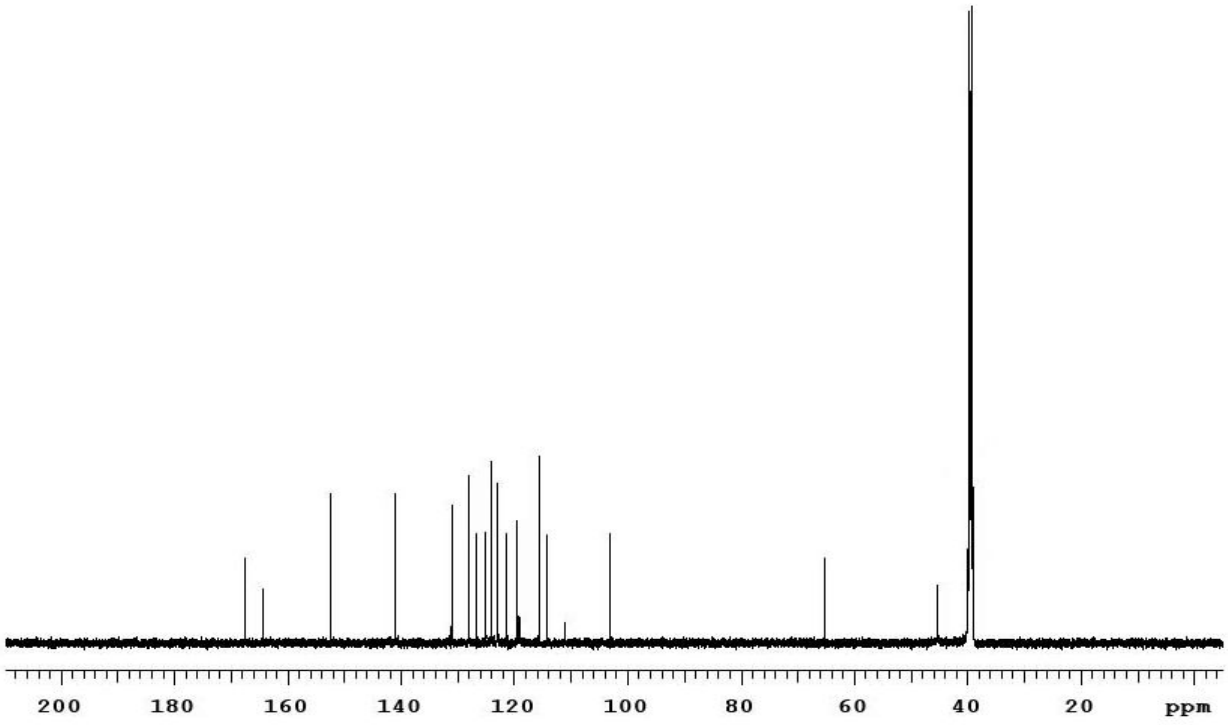


Figure S10. ^{13}C NMR spectra of **4d**

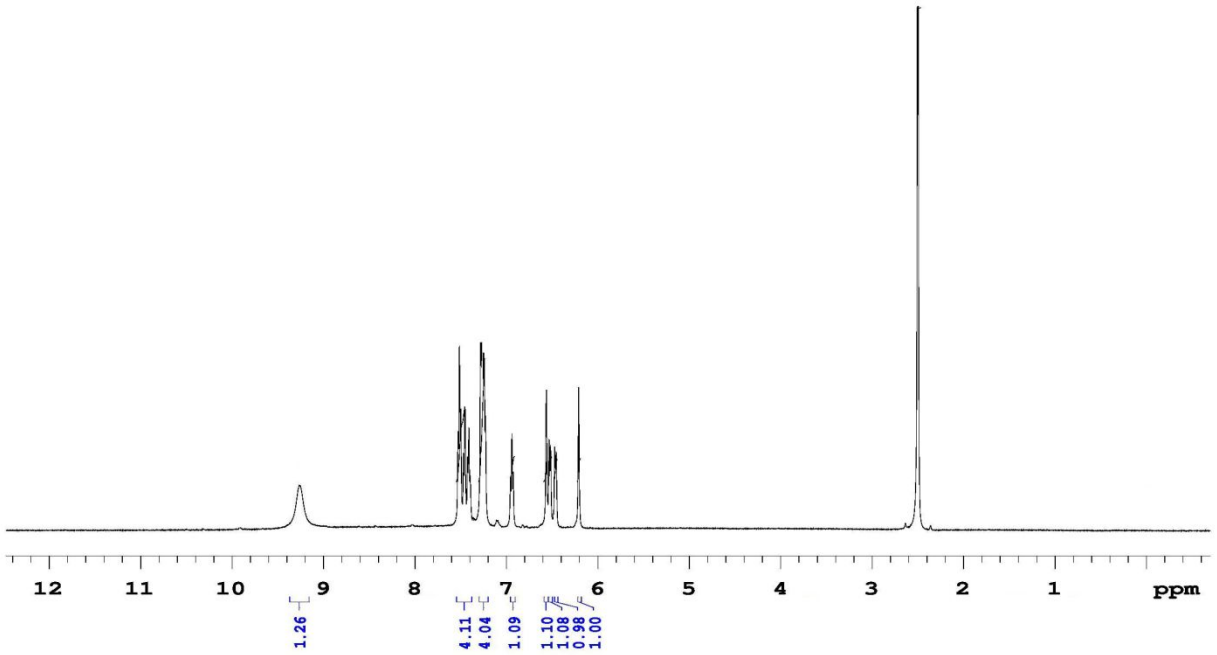


Figure S11. ^{1}H NMR spectra of **4e**

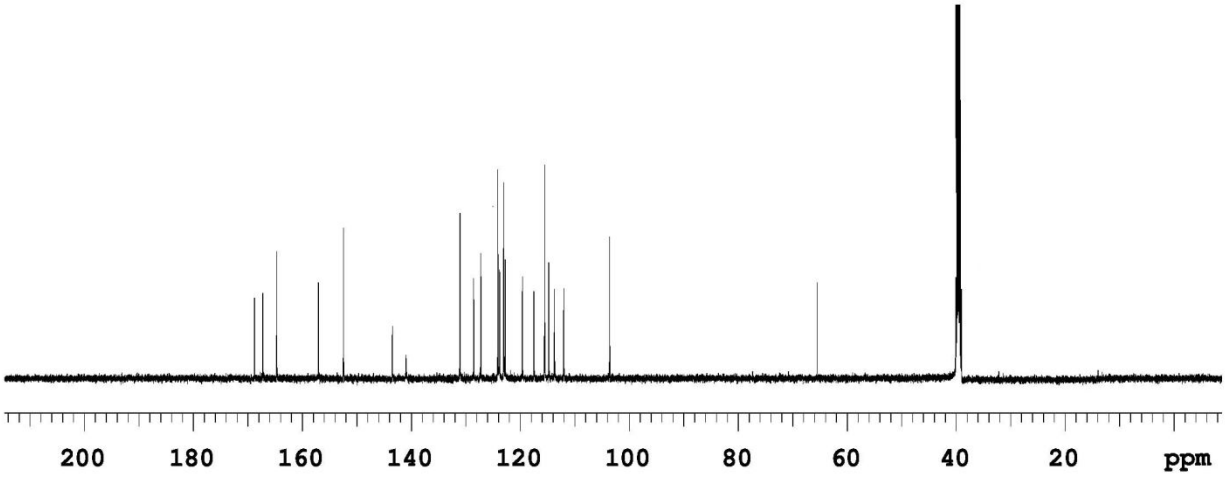


Figure S12. ^{13}C NMR spectra of **4e**

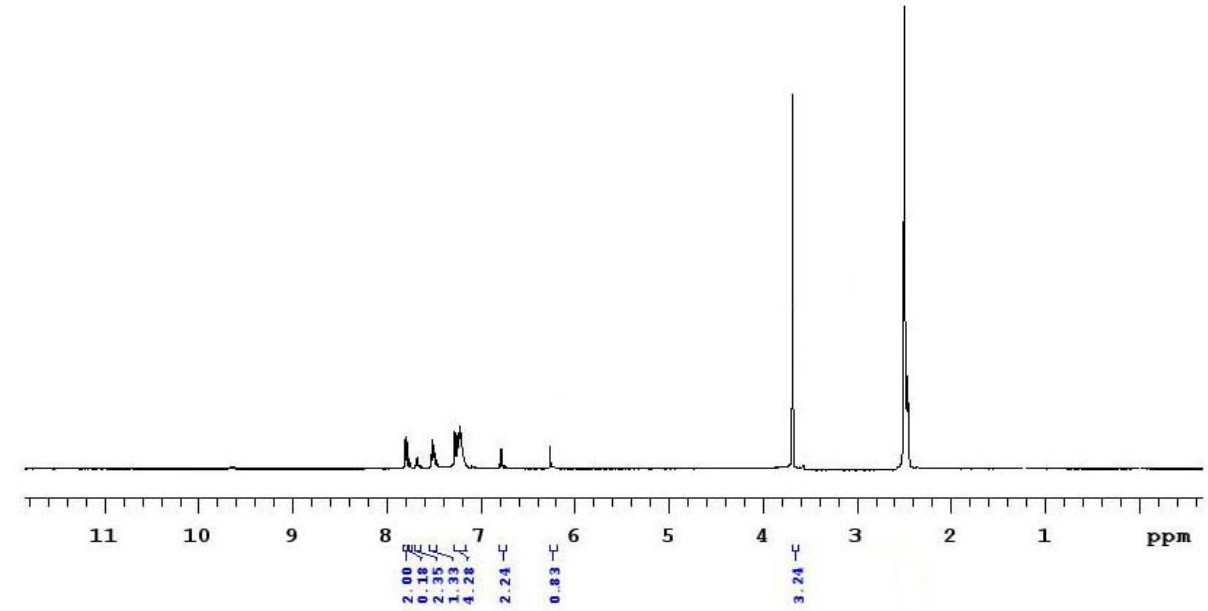


Figure S13. ^{1}H NMR spectra of **4f**

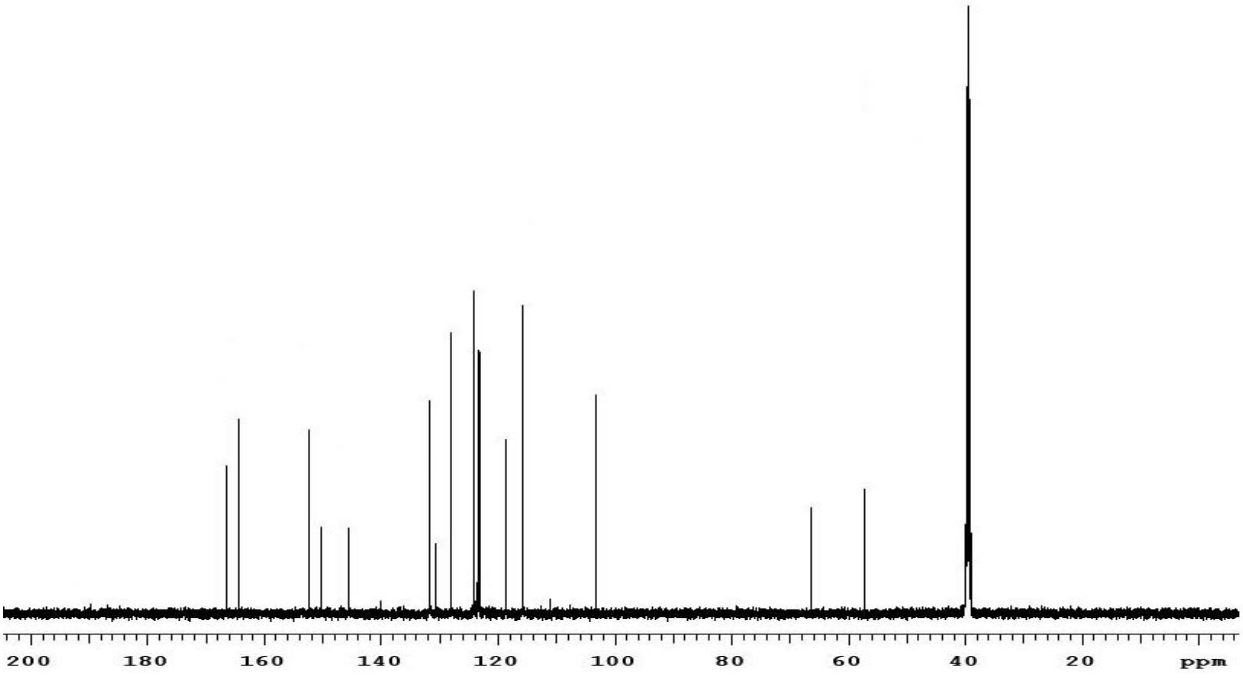


Figure S14. ^{13}C NMR spectra of **4f**

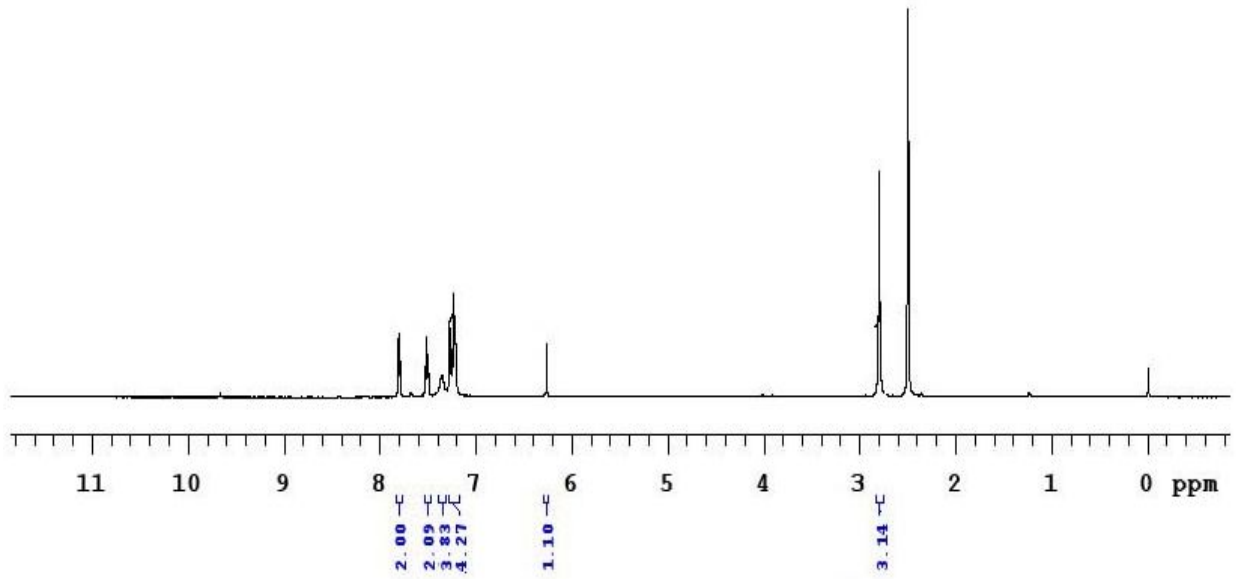


Figure S15. ^{1}H NMR spectra of **4g**

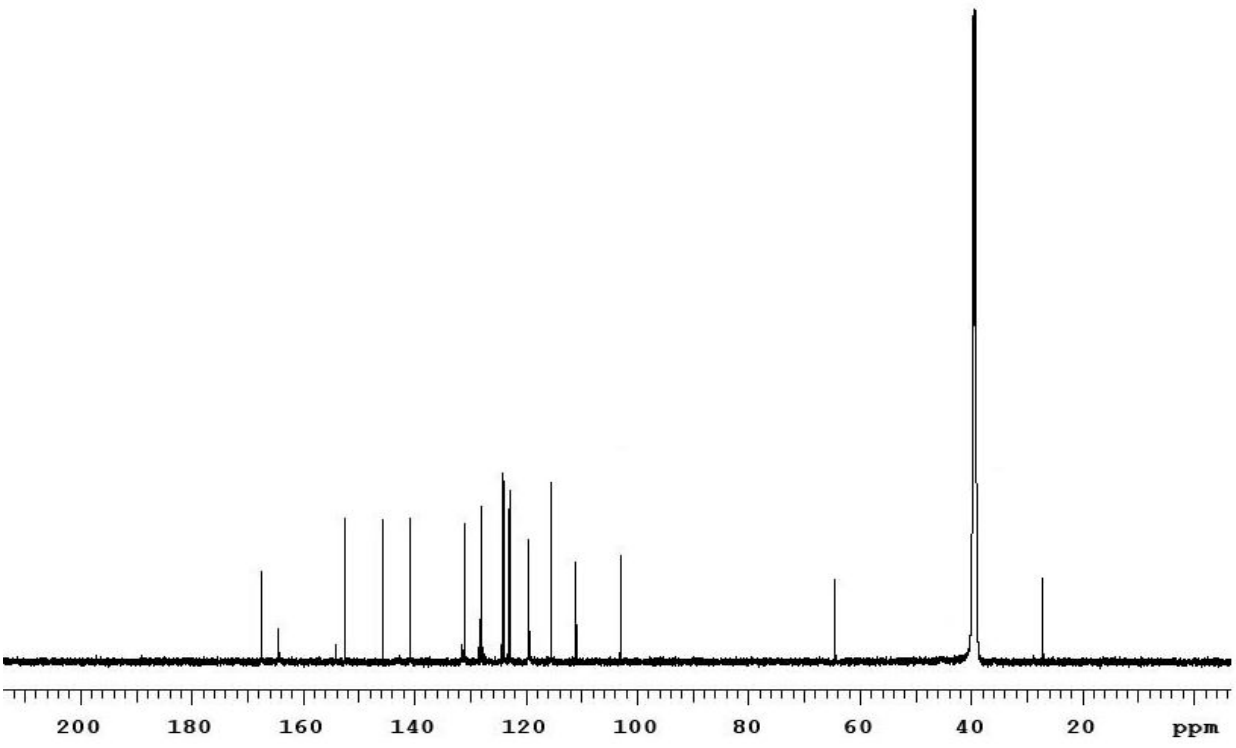


Figure S16. ^{13}C NMR spectra of **4g**

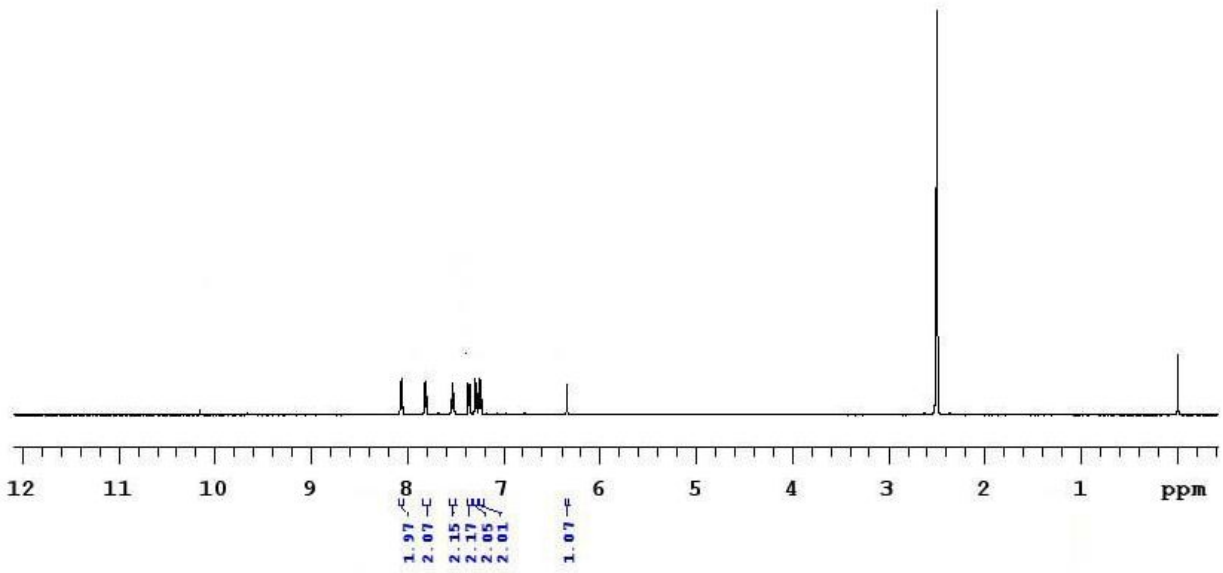


Figure S17. ^{1}H NMR spectra of **4h**

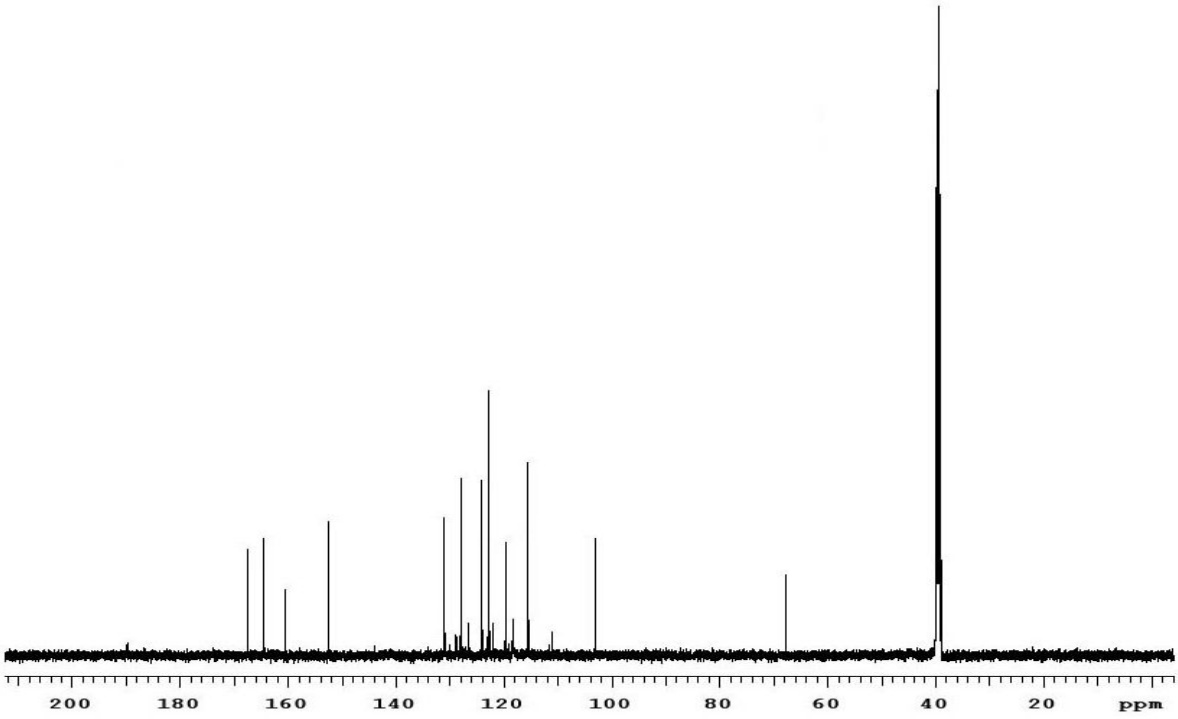


Figure S18. ^{13}C NMR spectra of **4h**

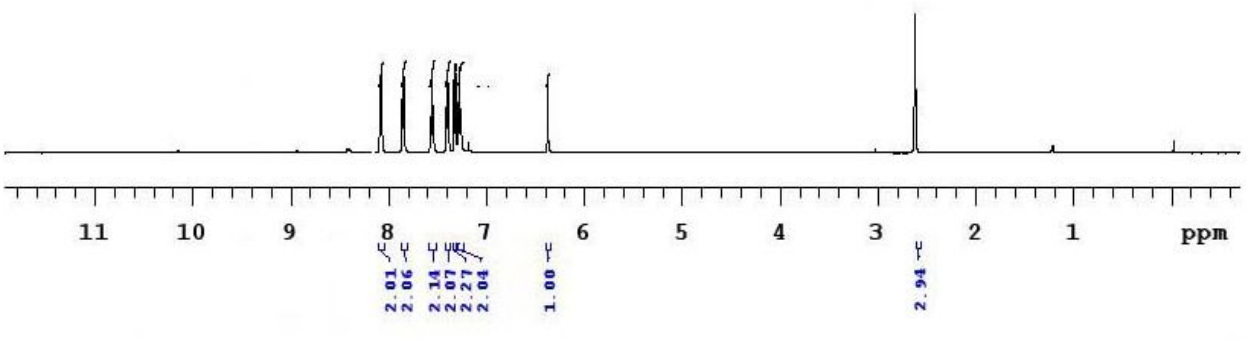


Figure S19. ^{1}H NMR spectra of **4i**

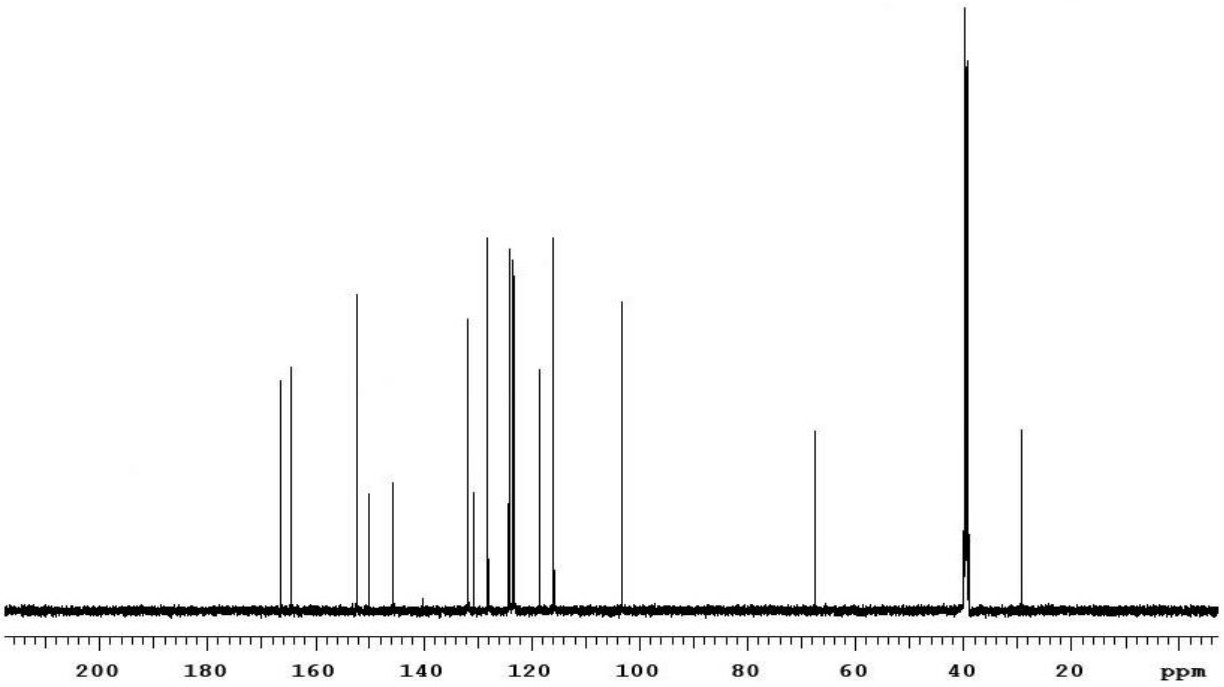


Figure S20. ^{13}C NMR spectra of **4i**

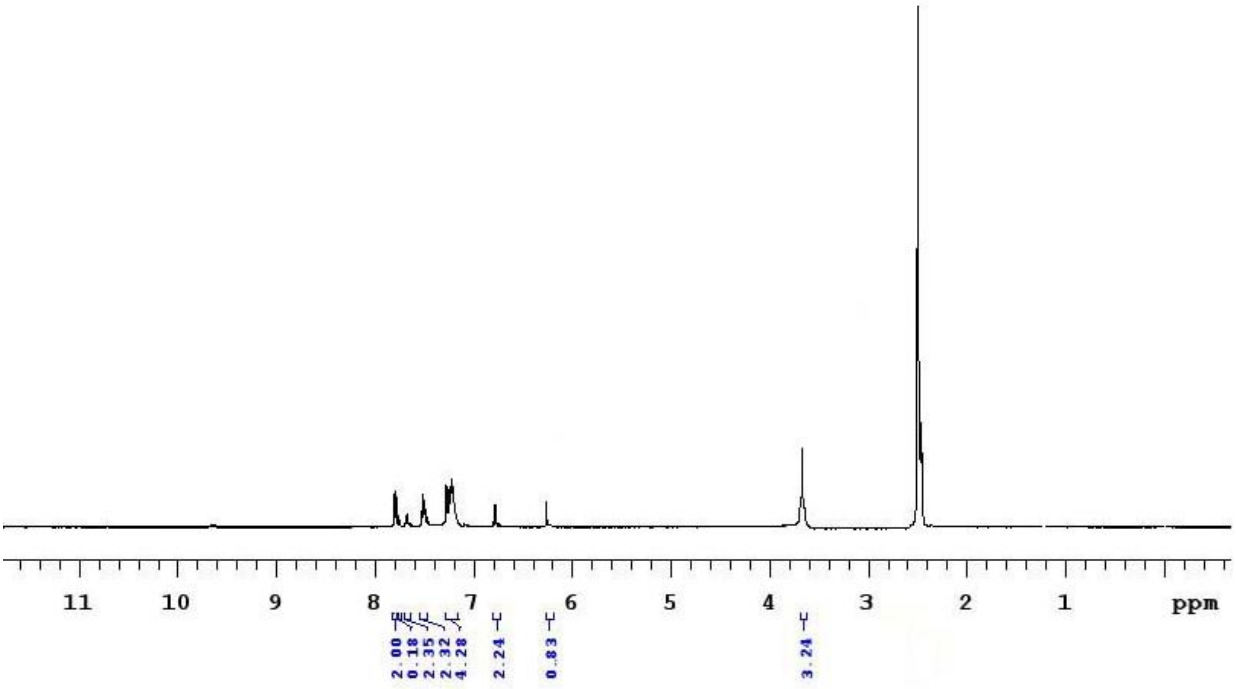


Figure S21. ^{1}H NMR spectra of **4j**

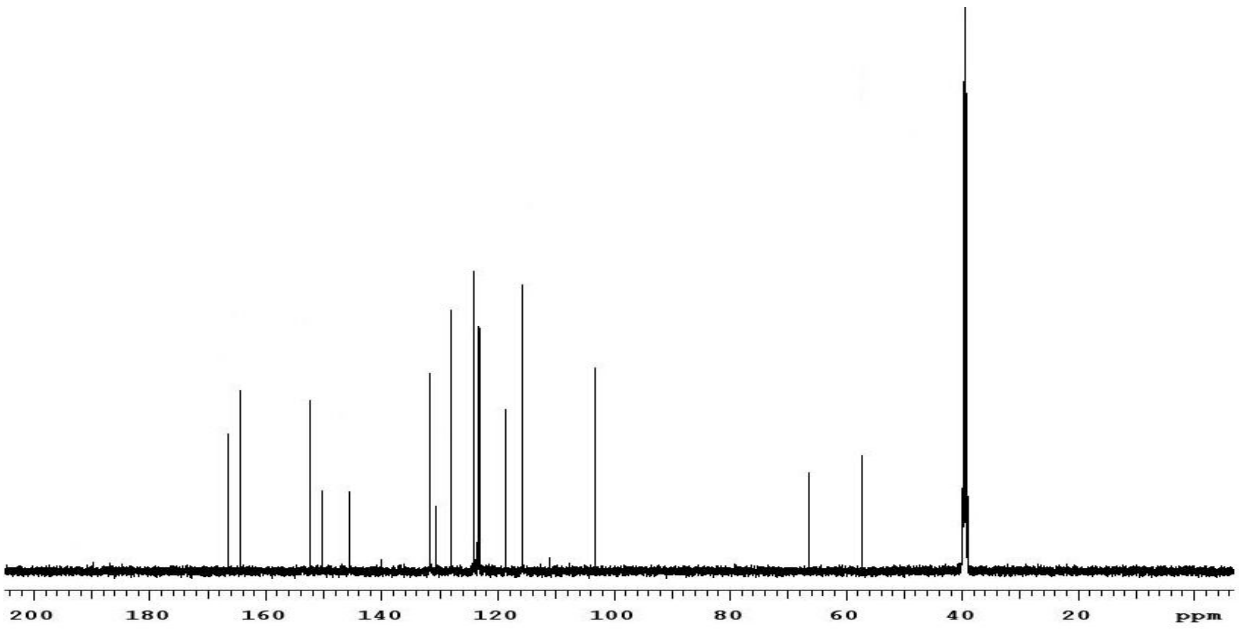


Figure S22. ^{13}C NMR spectra of **4j**

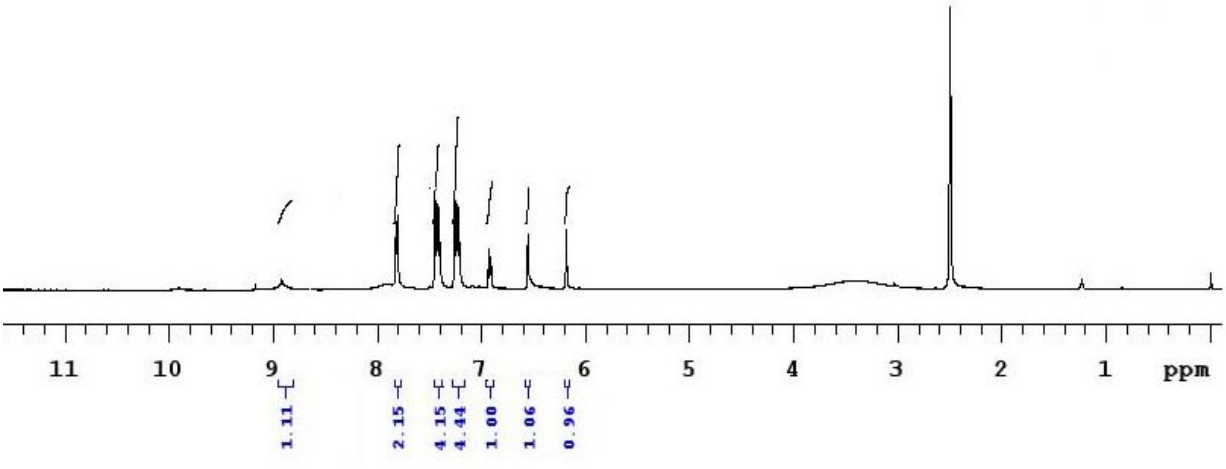


Figure S23. ^{1}H NMR spectra of **4k**

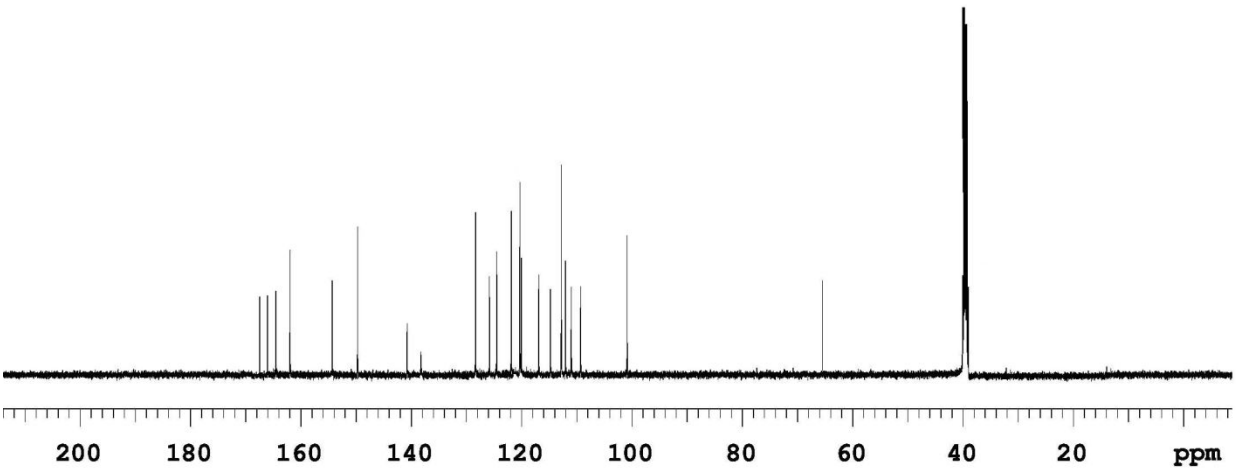


Figure S24. ^{13}C NMR spectra of **4k**

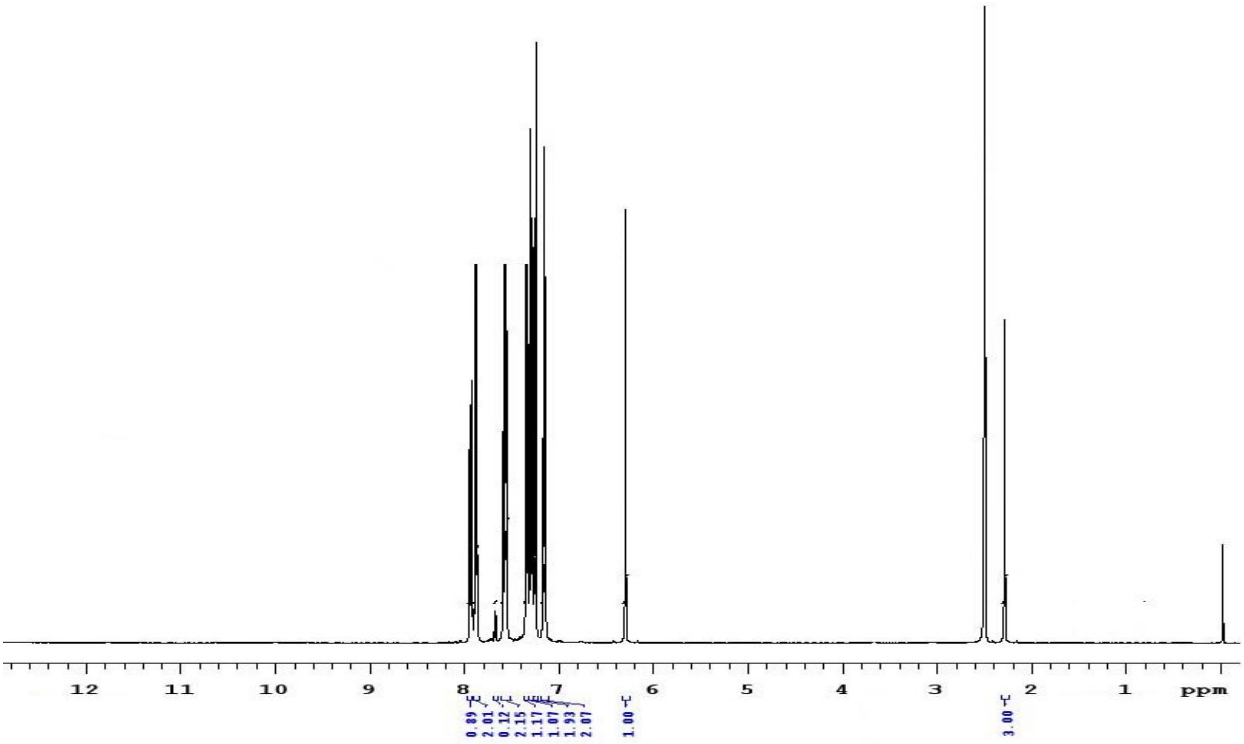


Figure S25. ^{1}H NMR spectra of **5a**

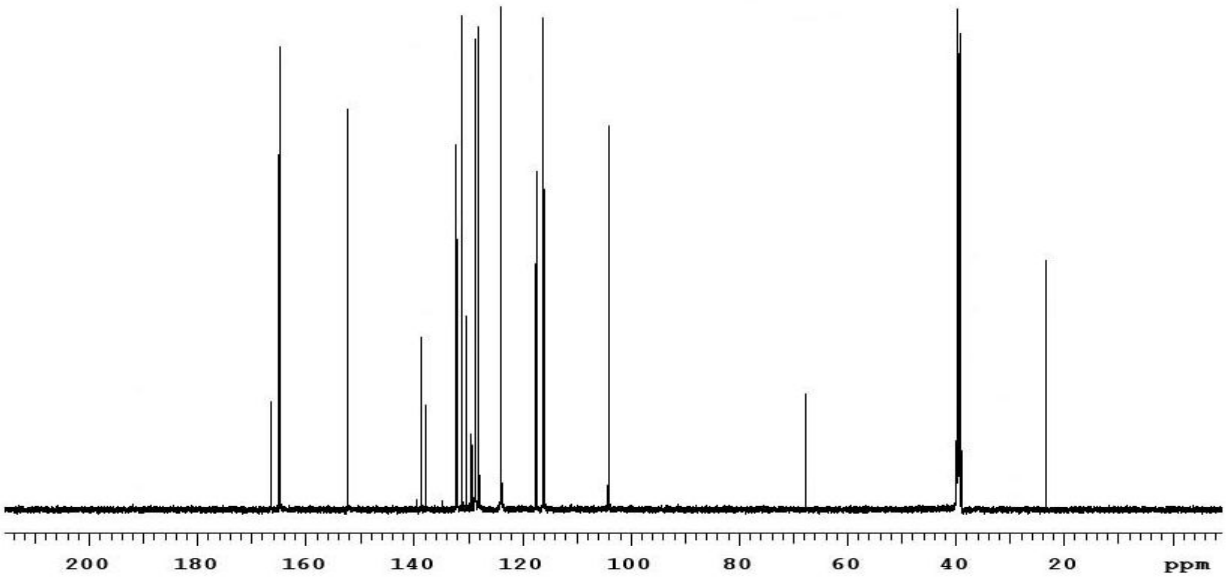


Figure S26. ^{13}C NMR spectra of **5a**

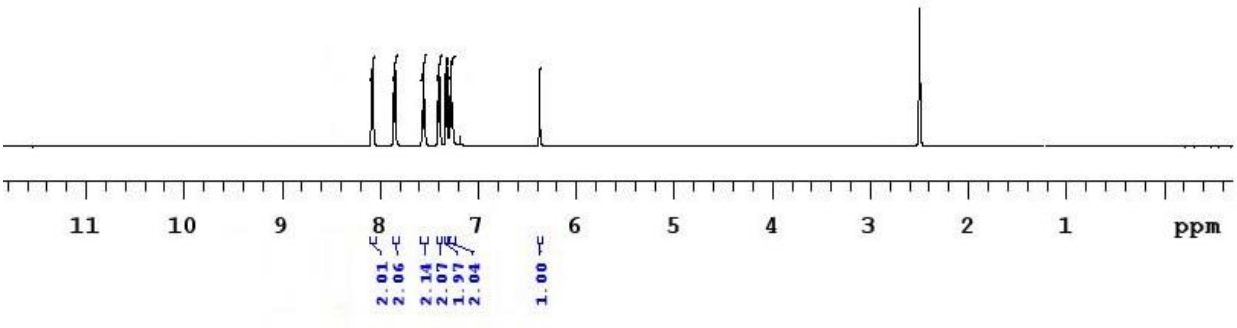


Figure S27. ^{1}H NMR spectra of **5b**

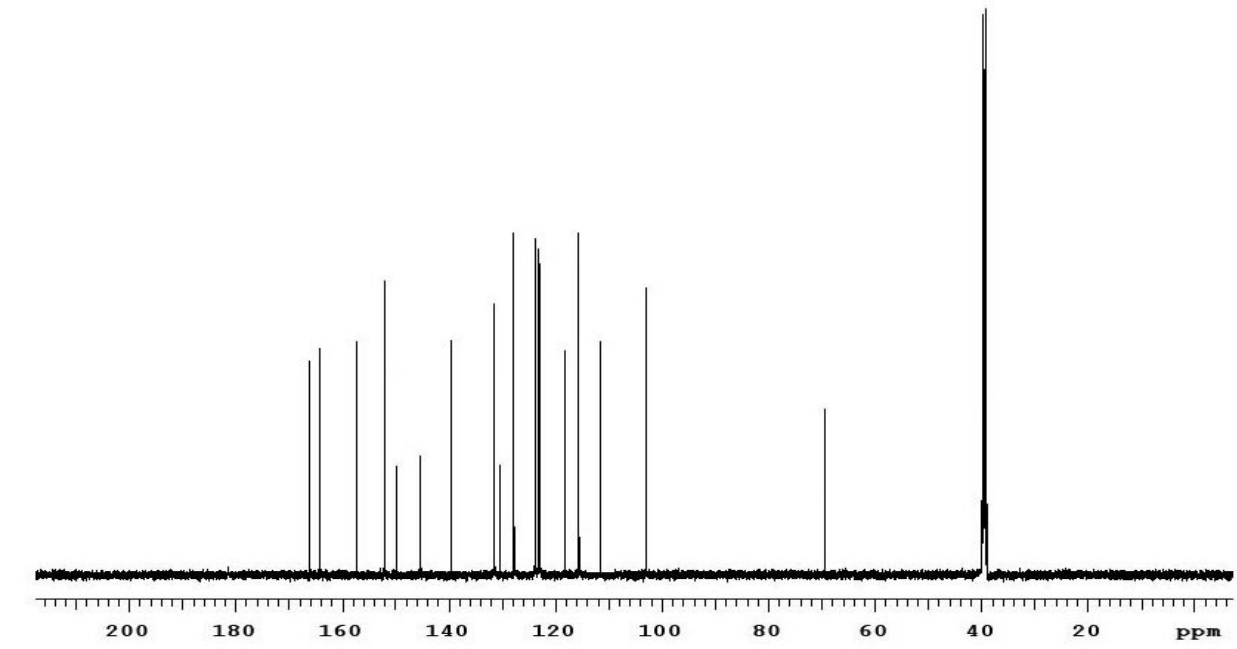


Figure S28. ^{13}C NMR spectra of **5b**

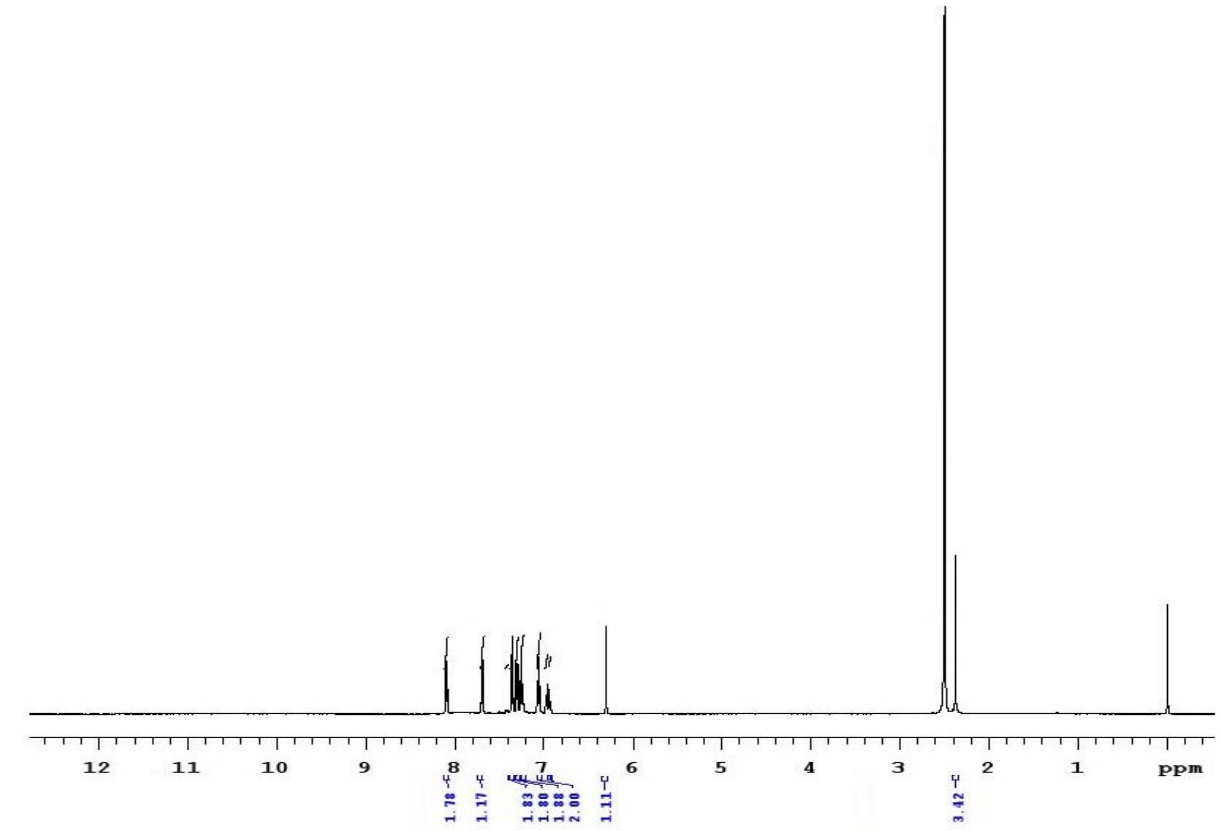


Figure S29. ^{1}H NMR spectra of **5c**

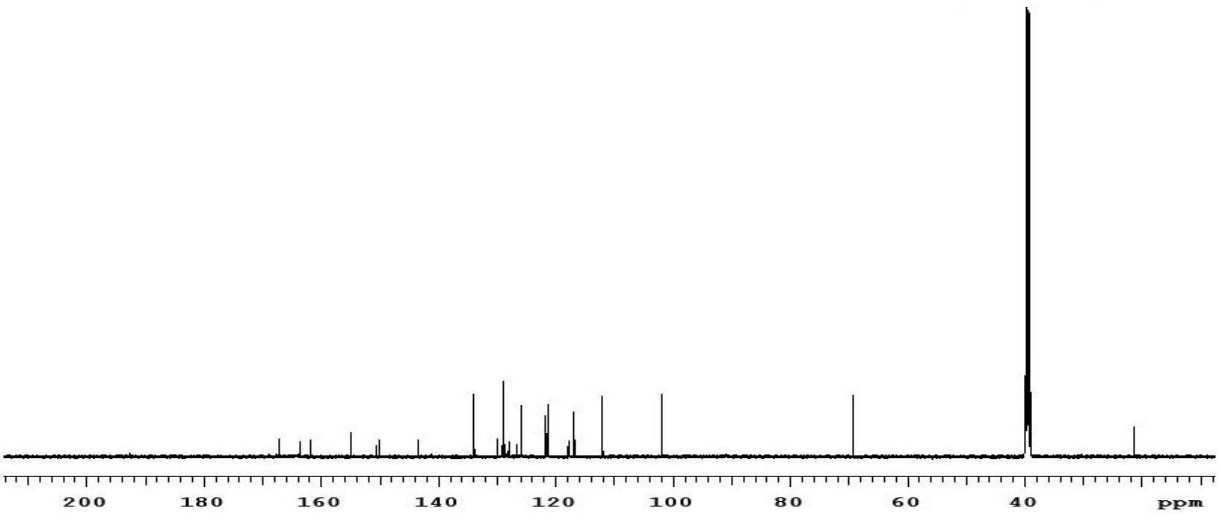


Figure S30. ^{13}C NMR spectra of **5c**

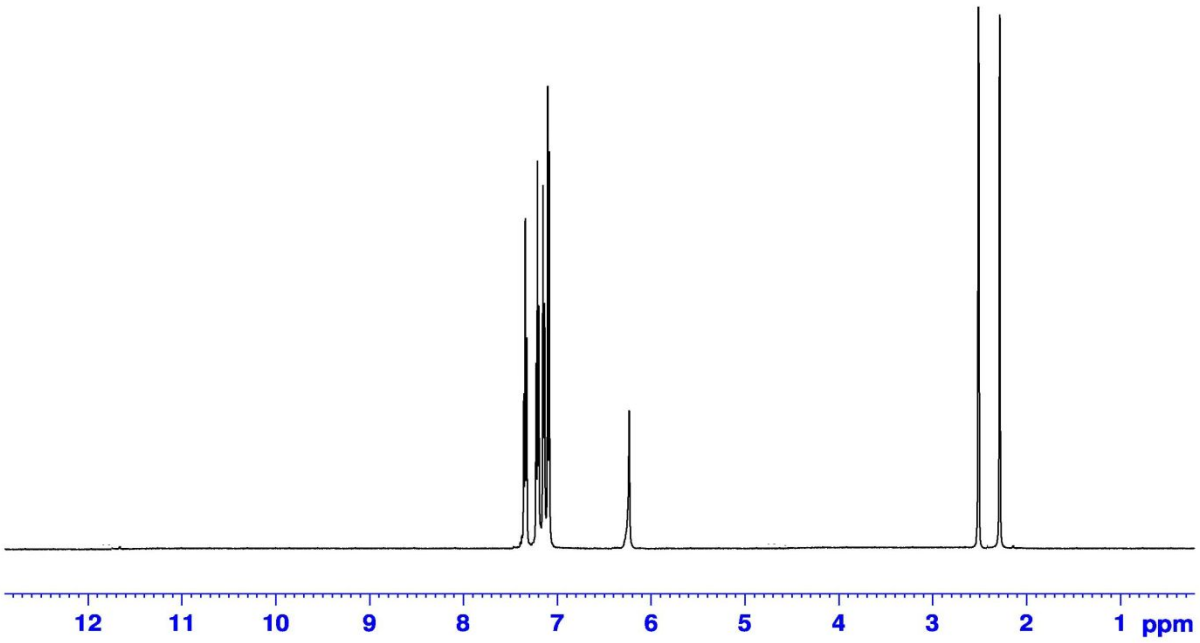


Figure S31. ^{1}H NMR spectra of **5d**

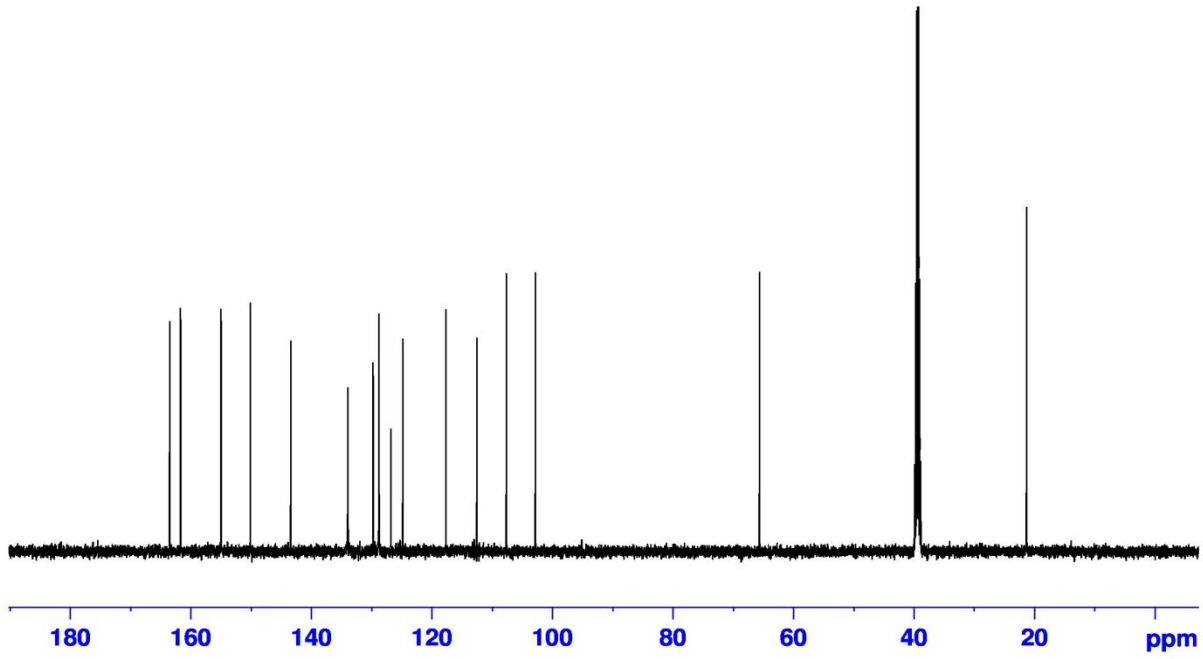


Figure S32. ^{13}C NMR spectra of **5d**

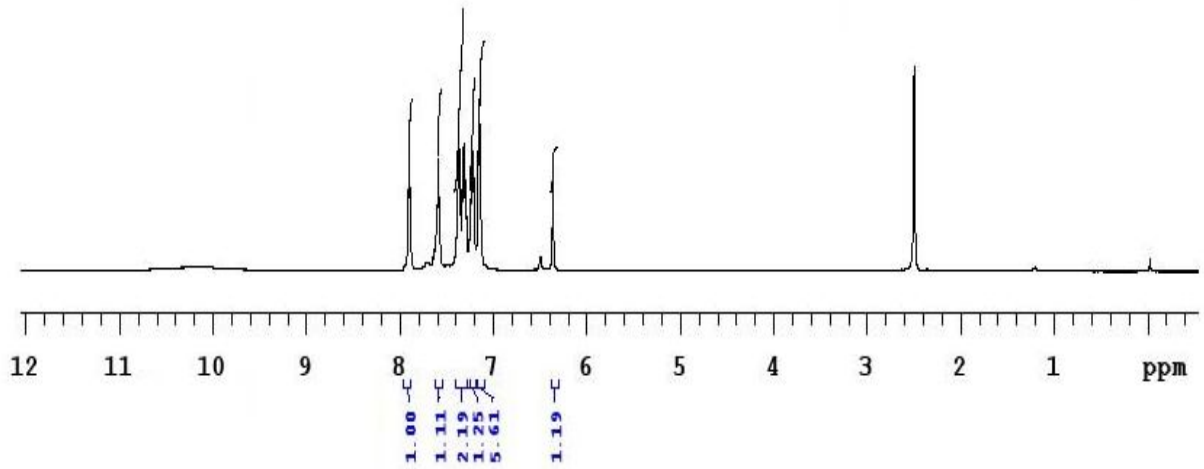


Figure S33. ^{1}H NMR spectra of **6a**

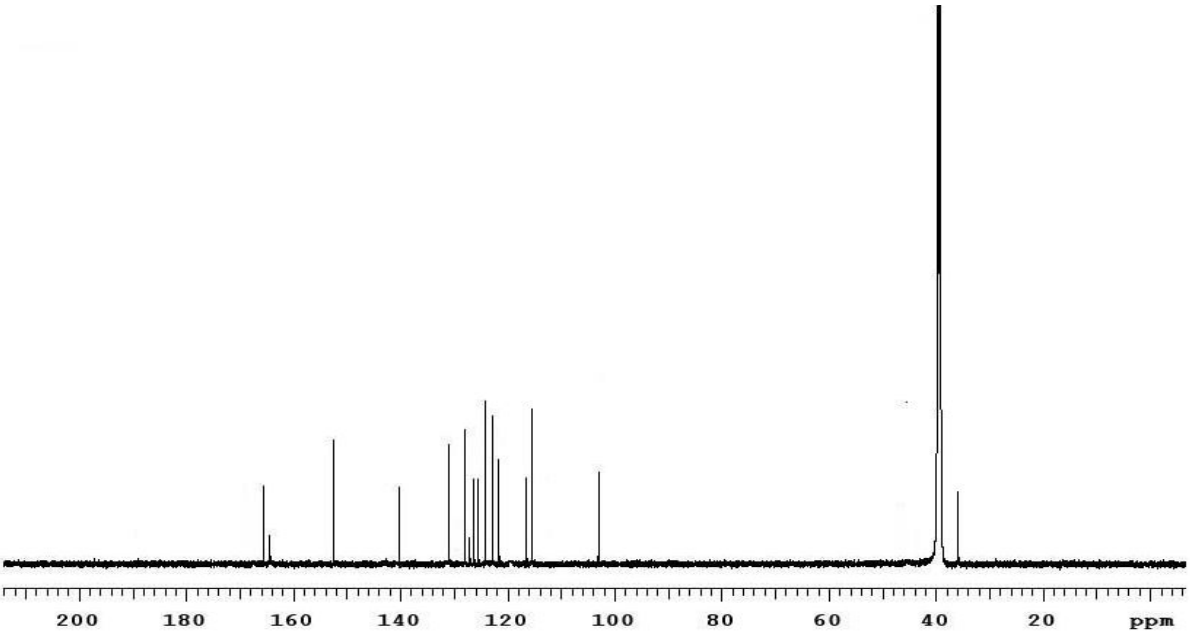


Figure S34. ^{13}C NMR spectra of **6a**

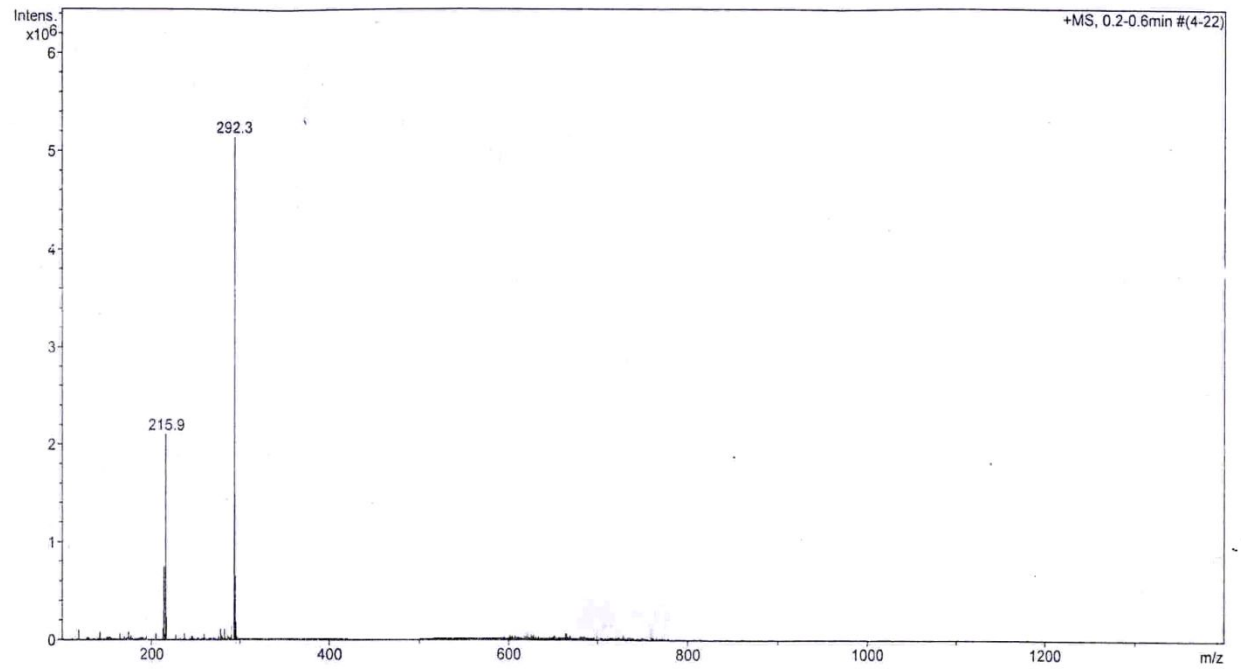


Figure S35. Mass spectra of **6a**

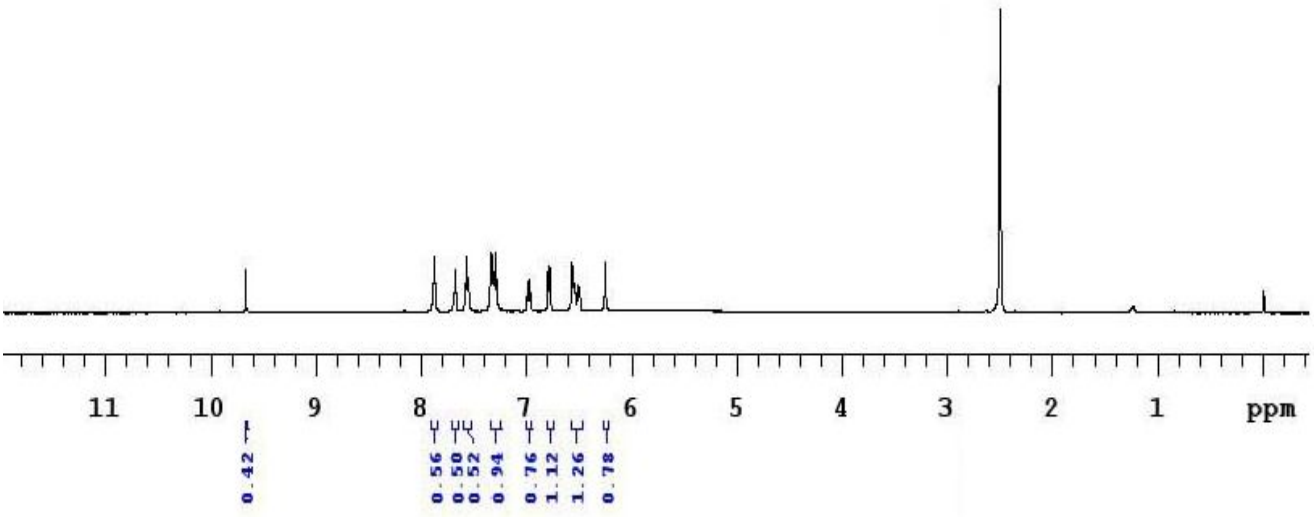


Figure S36. ^{1}H NMR spectra of **6b**

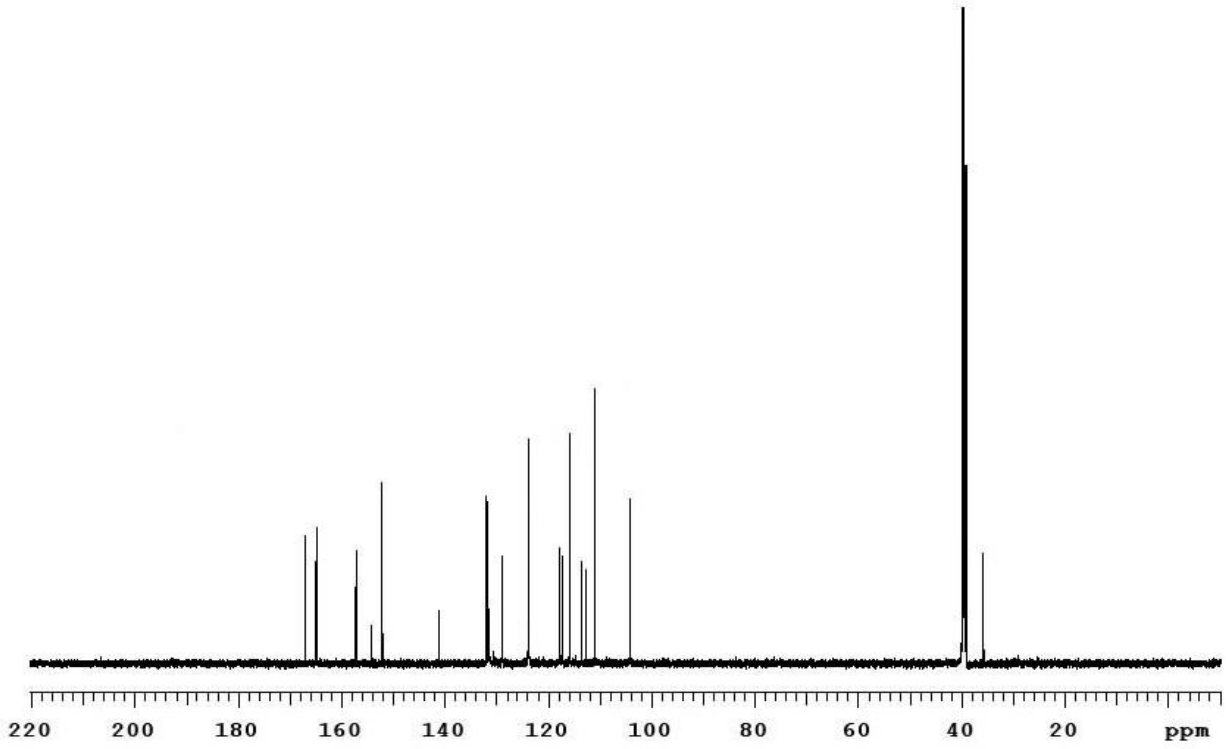


Figure S37. ^{13}C NMR spectra of **6b**

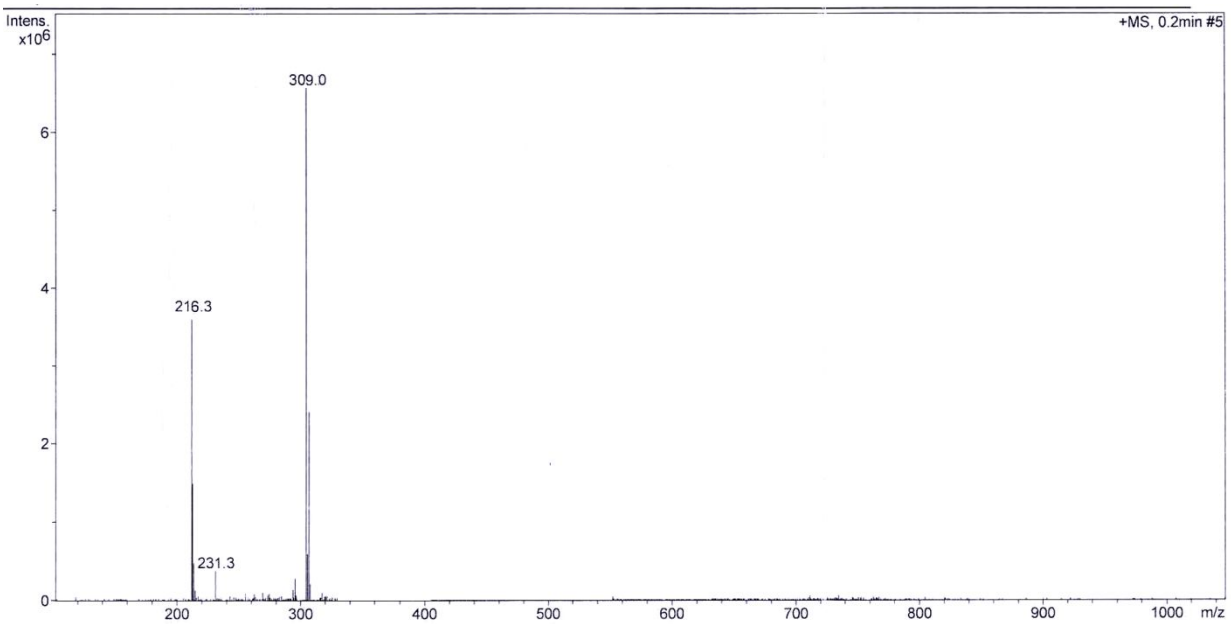


Figure S38. Mass spectra of **6b**

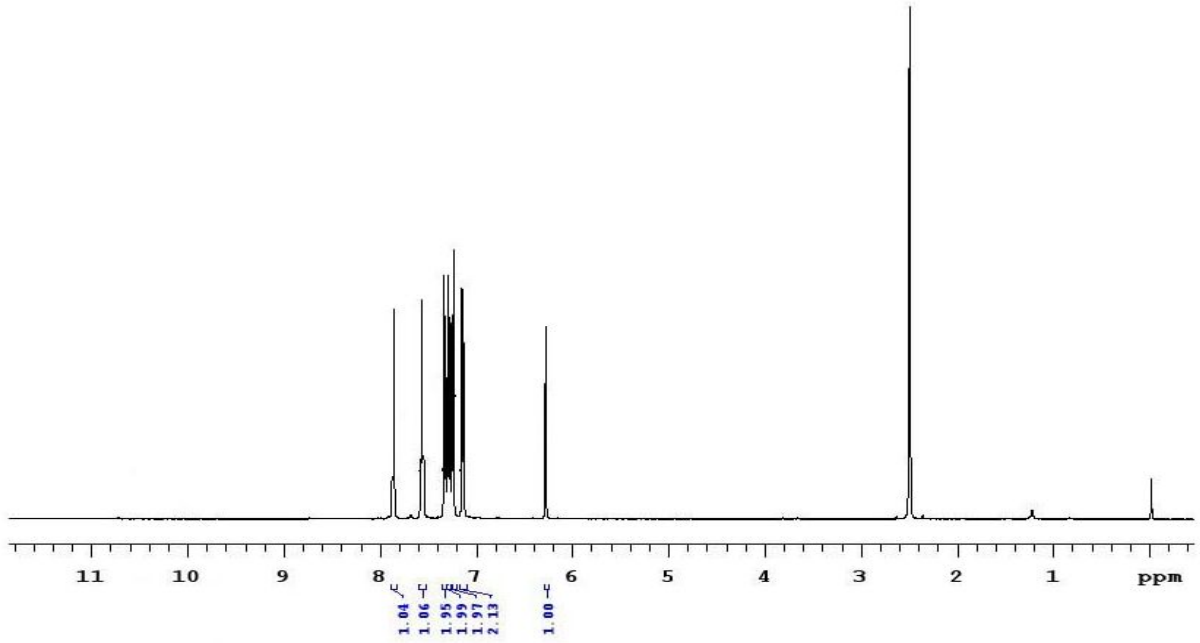


Figure S39. ^{1}H NMR spectra of **6c**

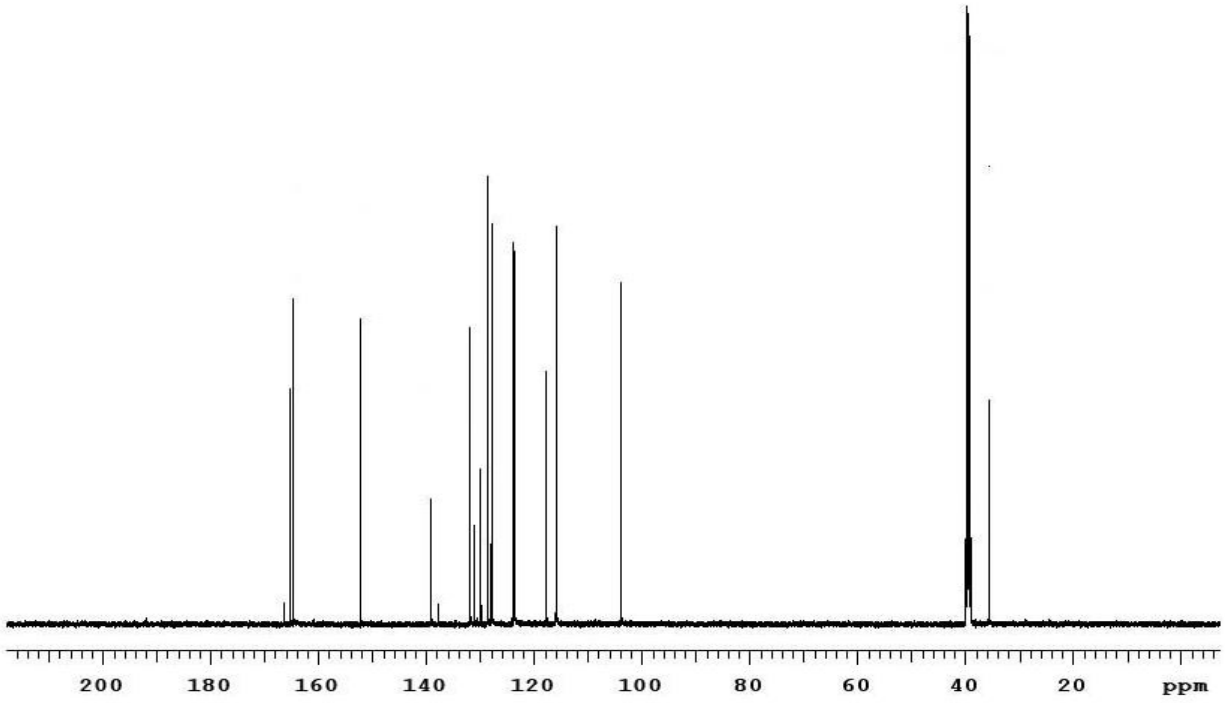


Figure S40. ^{13}C NMR spectra of **6c**

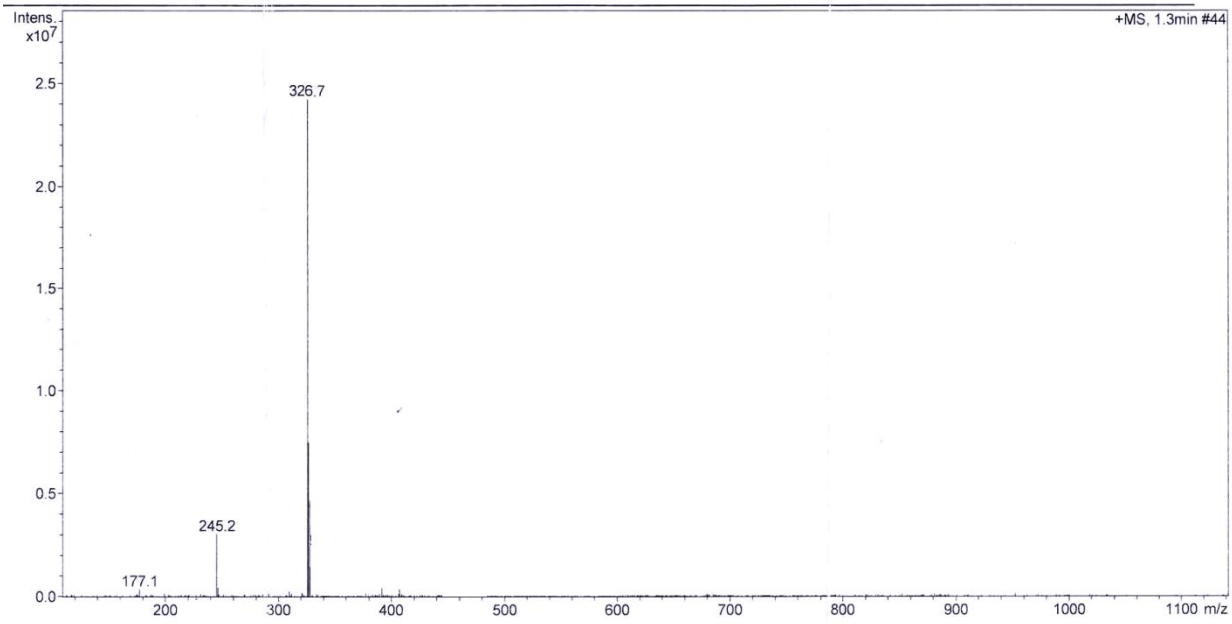


Figure S41. Mass spectra of **6c**

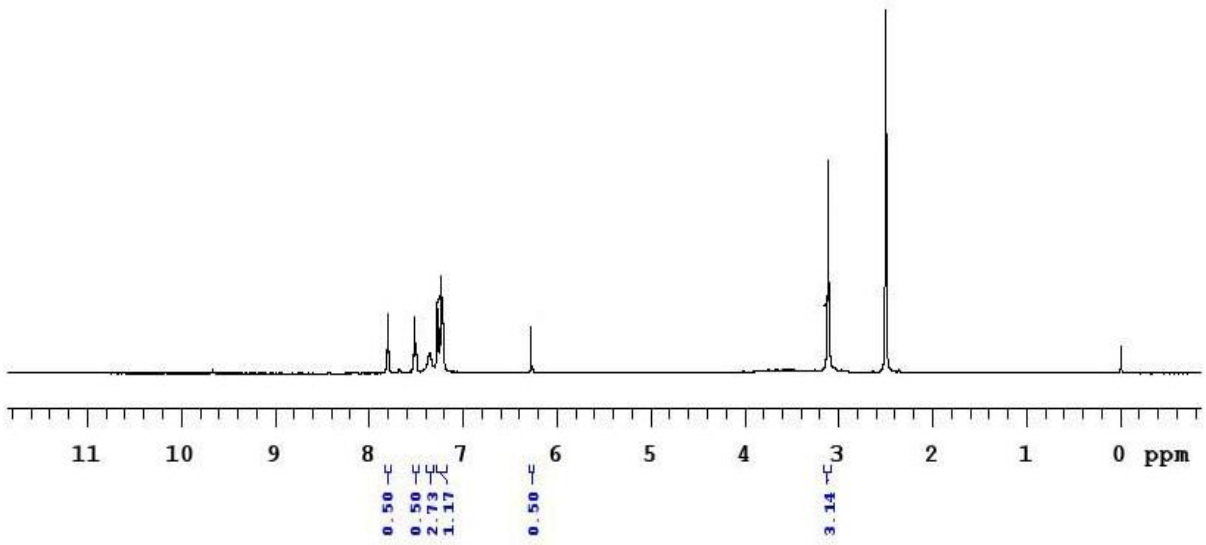


Figure S42. ^{1}H NMR spectra of **6d**

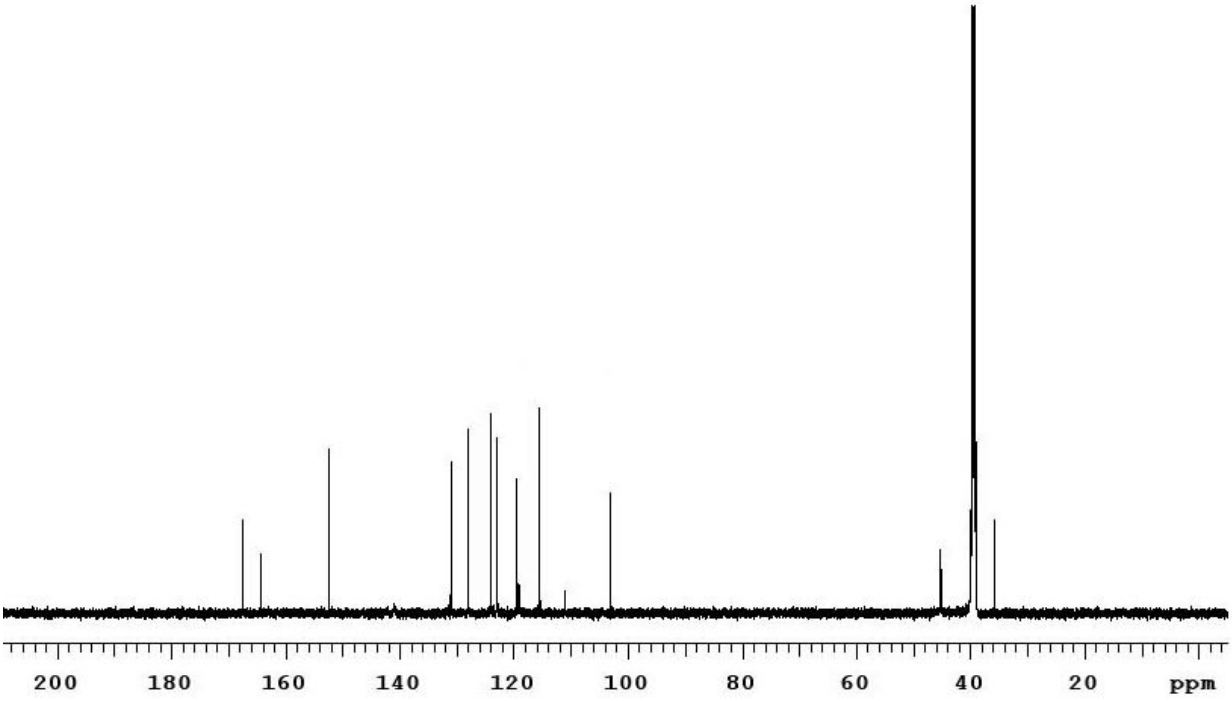


Figure S43. ^{13}C NMR spectra of **6d**

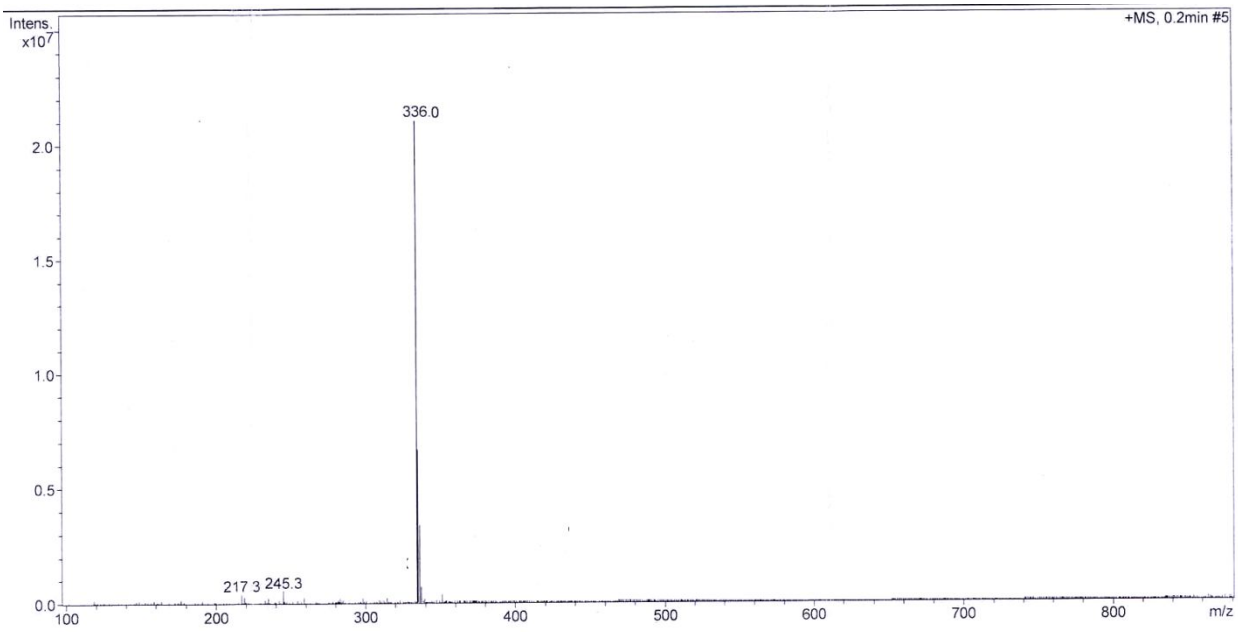


Figure S44. Mass spectra of **6d**

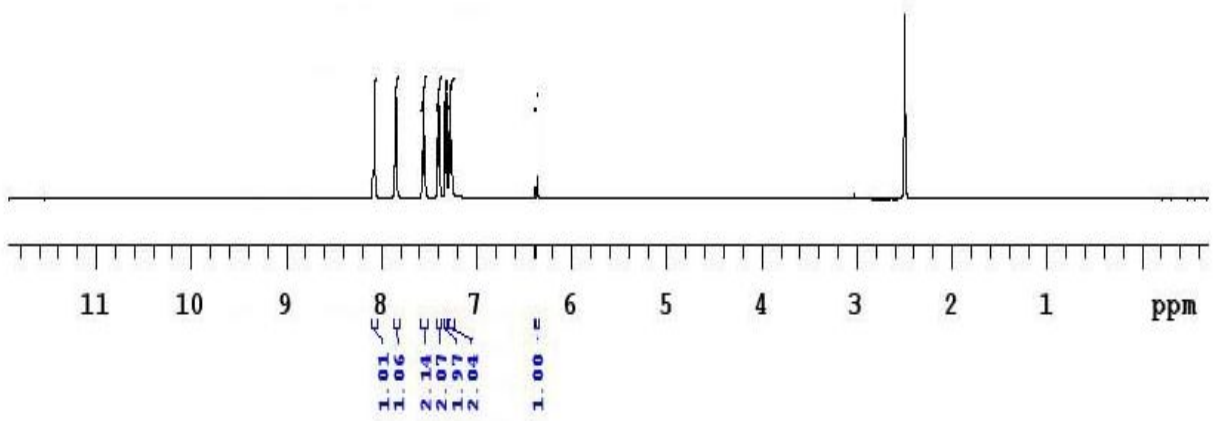


Figure S45. ^{1}H NMR spectra of **6e**

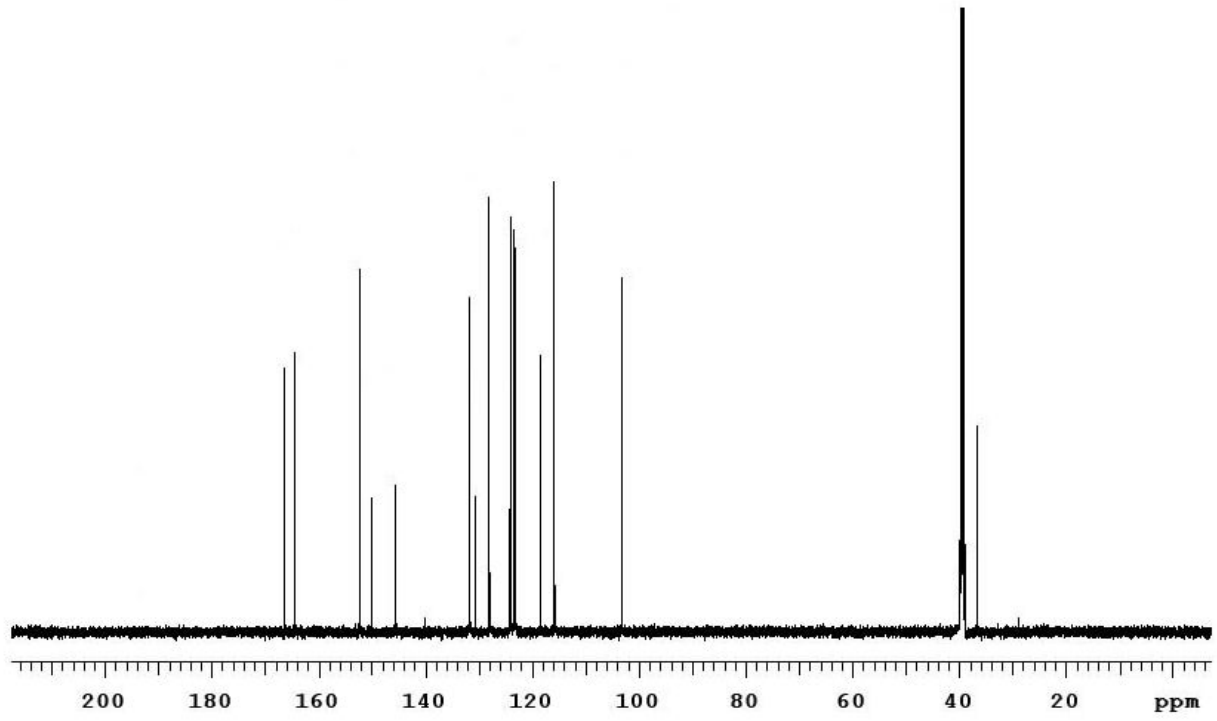


Figure S46. ^{13}C NMR spectra of **6e**

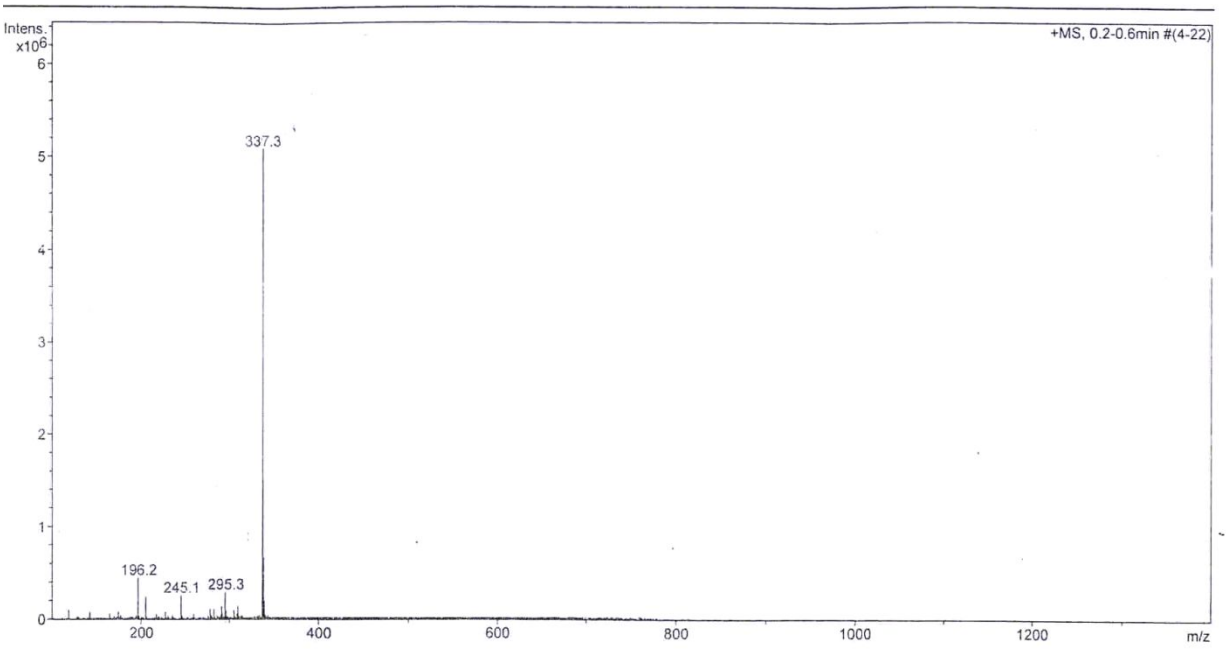


Figure S47. Mass spectra of **6e**

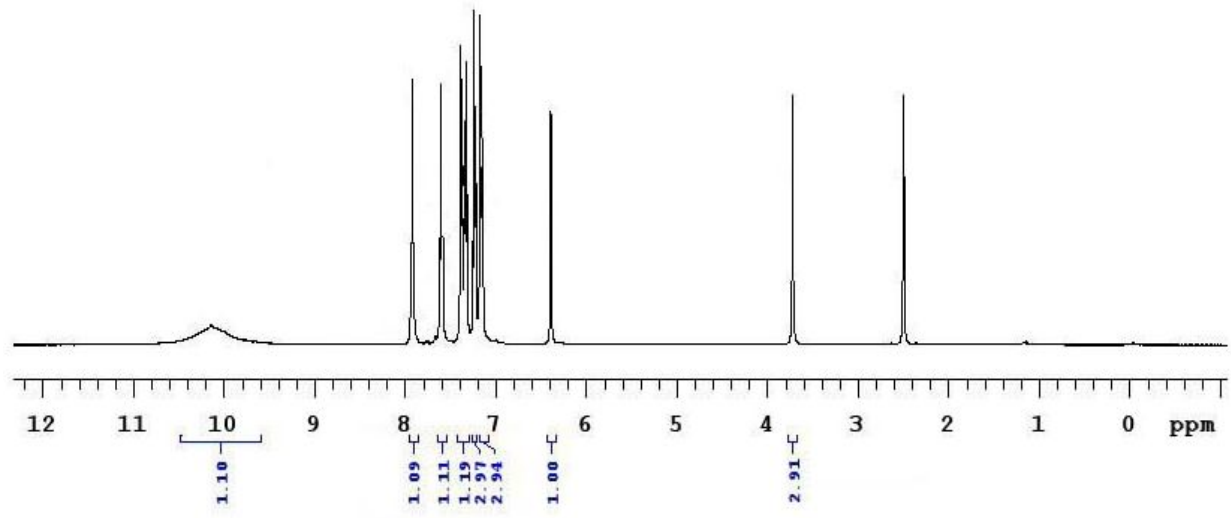


Figure S48. ^{1}H NMR spectra of **6f**

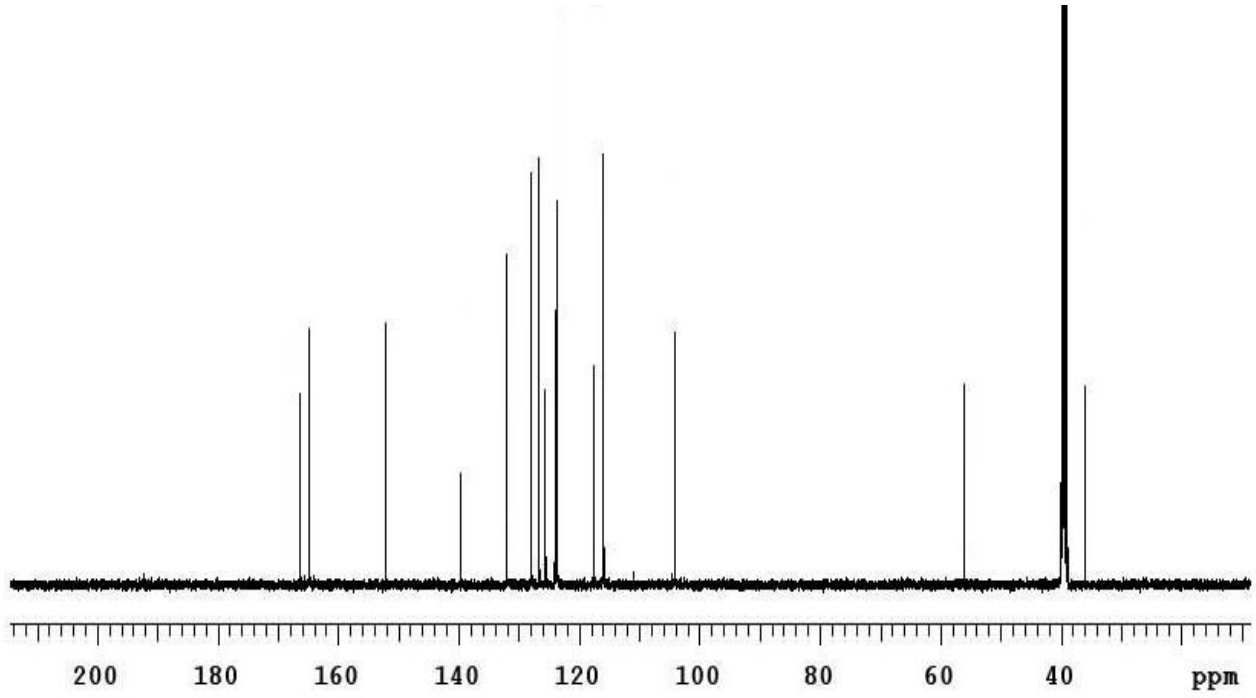


Figure S49. ^{13}C NMR spectra of **6f**

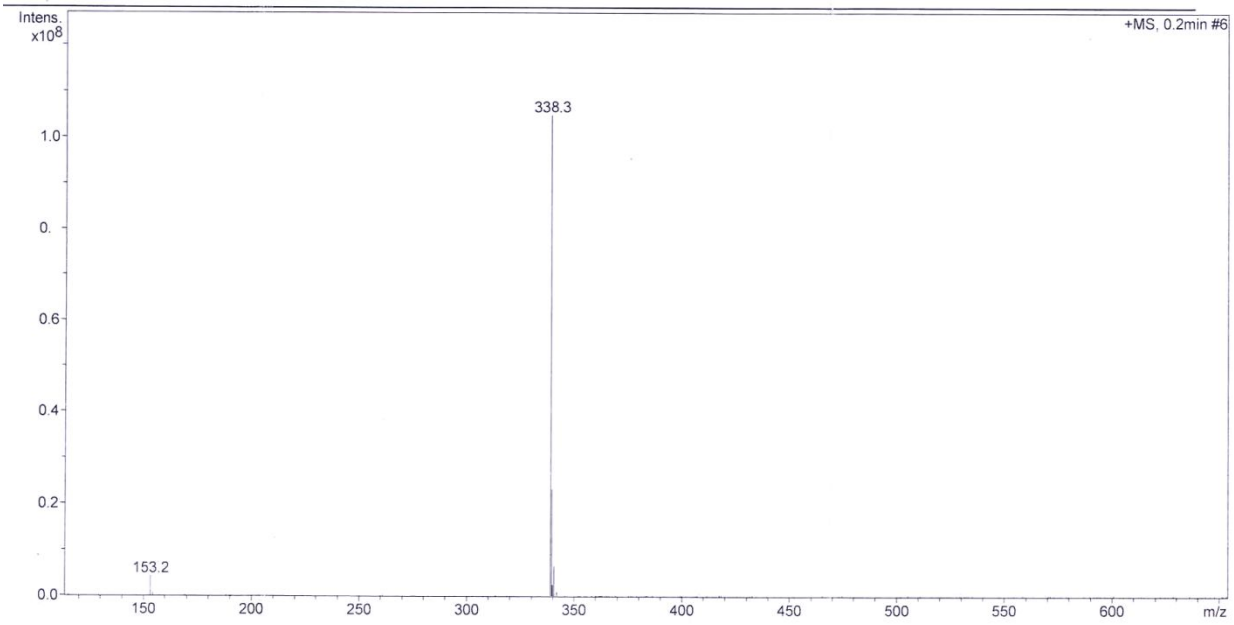


Figure S50. Mass spectra of **6f**

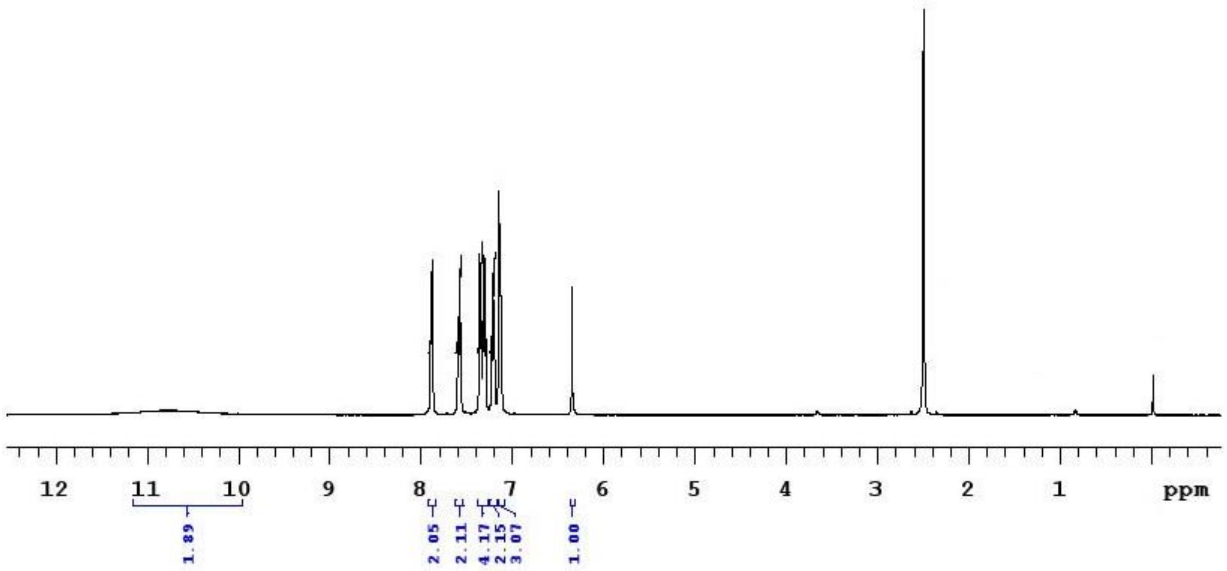


Figure S51. ^{1}H NMR spectra of **8**

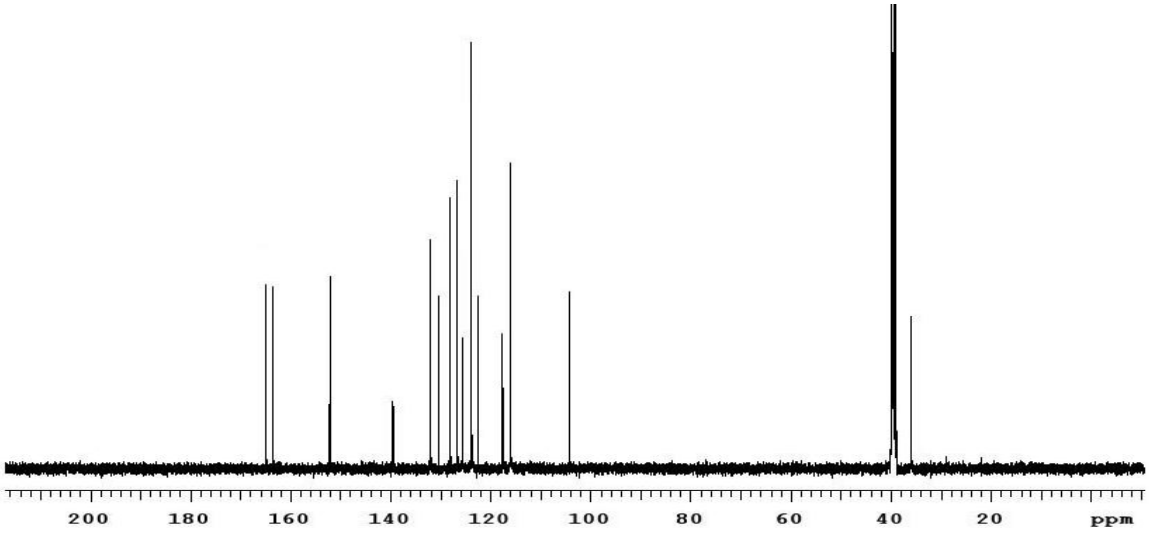


Figure S52. ^{13}C NMR spectra of **8**