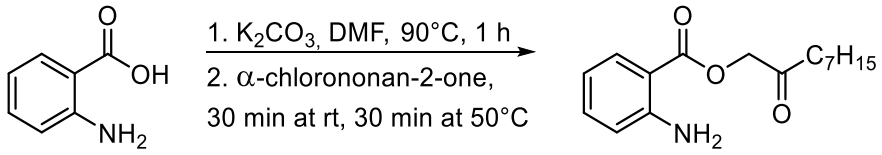
Supporting Information

Chemical synthesis

Synthesis of 2-oxononyl 2´-aminobenzoate **4**

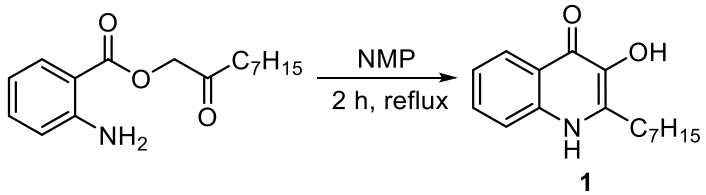


4

Anthranilic acid (3.26 g, 23.8 mmol, 1.2 eq.) was dissolved in DMF (44 mL) and K_2CO_3 (2.93 g, 21.19 mmol, 1.07 eq.) was added. The mixture was heated at 90°C for 1 h. After reaching room temperature, α-chlorononan-2-one (3.5 g, 19.8 mmol, 1.0 eq.) was added. The mixture was stirred for 30 min at room temperature and 30 min at 50°C. After letting cool to room temperature, the mixture was poured in ice-water and the precipitate collected by filtration and dried.

2-oxononyl 2-aminobenzoate (**4**) was obtained as a white solid (5.15 g, 94 %): ^{1}H-NMR ($CDCl_3$ 400.13 MHz) δ (ppm): 0.88 (t, *J* = 7.0 Hz, 3H, -CH_3), 1.21 – 1.35 (m, 8H, -(**CH**$_2$)$_4$-CH_3), 1.63 (m, 2H, -CO-CH_2-C**H**$_2$-), 2.50 (t, *J* = 7.4 Hz, 2H, -CO-C**H**$_2$-CH_2-), 4.83 (s, 2H, COO-CH_2-CO-), 5.68 (s, br, 2H, -NH_2), 6.66 (m, 2H, H-5 and H-3), 7.29 (m, 1H, H-4), 7.93 (dd, *J* = 8.5 Hz, J = 1.7 Hz, 1H, H-6). ^{13}C-NMR ($CDCl_3$ 100.61 MHz) δ (ppm): 14.1 (-CH_3), 22.7, 23.4, 29.1, 29.2, 31.7 (-(**C**H_2)$_5$-CH_3), 39.0 (-CO-**C**H_2-), 68.1 (-COO-**C**H_2-CO-), 110.0 (C-1), 116.7 (C-5 and C-3), 131.5 (C-6), 134.7 (C-4), 150.9 (C-2), 167.3 (-COO-), 204.9 (-CO-). ESI-HRMS: m/z = 278.1752 $[M+H]^+$, calc. for $C_{16}H_{23}NO_3 + H^+$ = 278.1751; m/z = 300.1574 $[M+Na]^+$, calc. for $C_{16}H_{23}NO_3 + Na^+$ = 300.1570.

Synthesis of 2-heptyl-3-hydroxy-4-quinolone (**1**)

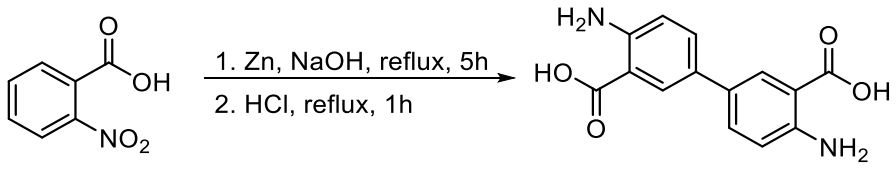


1

Compound **4** (1.91 g, 6.9 mmol) was dissolved in NMP (19.2 mL) and the solution was refluxed for 2 h. After reaching room temperature, the solution was poured in ice-water and the precipitated collected by filtration and dried.

2-heptyl-3-hydroxyquinolin-4(1*H*)-one (**1**) was obtained as white solid (1.50 g, 84 %): ^{1}H-NMR (DMSO-d_6 400.13 MHz) δ (ppm): 0.84 (t, *J* = 6.9 Hz, 3H, -CH_3), 1.20 – 1.37 (m, 8H, -(C**H**$_2$)$_4$-CH_3), 1.67 (m, 2H, -C**H**$_2$-(CH_2)$_4$-CH_3), 2.72 (t, *J* = 7.9 Hz, 2H, -C**H**$_2$-(CH_2)$_5$-CH_3), 7.21 (ddd, *J* = 2.6 Hz, 5.4 Hz, 8.1 Hz, 1H, H-6), 7.52 (m, 2H, H-7 and H-8), 8.09 (d, *J* = 8.1 Hz, 1H, H-5), 11.40 (s, 1H, NH). ^{13}C-NMR (DMSO-d_6 100.61 MHz) δ (ppm): 13.9 (-CH_3), 22.0, 28.4, 28.7, 31.2 (-(**C**H_2)$_4$-CH_3), 27.8 (-**C**H_2-(CH_2)$_4$-CH_3), 28.1 (-**C**H_2-(CH_2)$_5$-CH_3), 117.7 (C-8), 121.5 (C-6), 122.1 (C-4a), 124.5 (C-5), 129.9 (C-7), 135.5 (C-2), 137.4 (C-8a), 137.8 (C-3), 168.9 (C-4). ESI-HRMS: m/z = 260.1651 $[M+H]^+$, calc. for $C_{16}H_{21}NO_2 + H^+$ = 260.1645; m/z = 282.1470 $[M+Na]^+$, calc. for $C_{16}H_{21}NO_2 + Na^+$ = 282.1465.

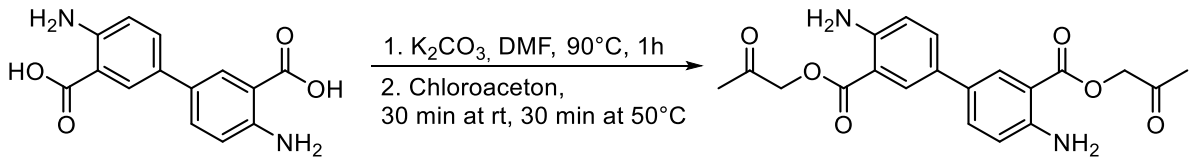
Synthesis of 4,4'-diamino-[1,1'-biphenyl]-3,3'-dicarboxylic acid (**5**)



5

o-Nitrobenzoic acid (6.685 g, 0.04 mol) was dissolved in 200 mL of 12.5 M aq. NaOH to result in a 0.2 M solution. To the solution was added 50 g Zn (0.765 mol) and the mixture heated to 100°C and kept at this temperature for 5 h. After the solution reached room temperature, 100 mL conc. HCl was slowly added and the mixture heated to 100°C for 1 h. After reaching room temperature, the precipitate was isolated by filtration under vacuum and washed with water. The compound was obtained as a green-yellow solid (3.87 g, 71 %). ^{1}H-NMR (DMSO-d_6 400.1 MHz) δ (ppm): 6.80 (d, 2H, J = 8.7 Hz, H-5 and H-5`), 7.57 (dd, 2H, J = 8.7 Hz, J = 2.3 Hz, H-6 and H-6´), 7.84 (d, 2H, J = 2.3 Hz, H-2 and H-2´). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 109.9 (2C, C-3 and C-3´), 117.9 (2C, C-5 and C-5´), 126.6 (2C, C-1 and C-1´), 127.5 (2C, C-2 and C-2´), 131.4 (2C, C-6 and C-6´), 150.1 (2C, C-4 and C-4´), 169.6 (2C, 2xCOOH).

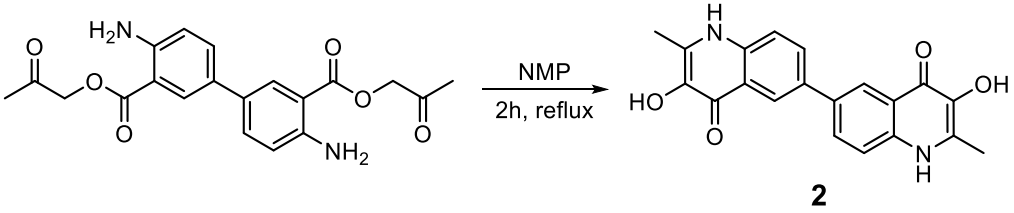
Synthesis of bis(2-oxopropyl) 4,4'-diamino-[1,1'-biphenyl]-3,3'-dicarboxylate (**6**)



6

Compound **5** (1.718 g, 6.31 mmol) was dissolved in 25 mL DMF and K_2CO_3 (9.465 mmol, 1.5 eq.) was added. The mixture was heated at 90°C for 1 h. After reaching room temperature, chloroacetone (1.75 ml, 3 eq.) was added. The mixture was stirred for 30 min at room temperature and 30 min at 50°C. At room temperature, the mixture was poured in ice-water. The mixture was extracted with ethyl acetate, the combined organic phases washed with brine, filtered and the solvent evaporated leaving a brown solid. The solid was suspended in a small volume of ethyl acetate and collected by filtration. The solid was washed with small volumes of ethyl acetate and dried leaving the clean product as a brown solid (1.25 g, 51.5%). ^{1}H-NMR (DMSO-d_6 400.1 MHz) δ (ppm): 2.16 (s, 6H, 2x -CH_3), 4.98 (s, 4H, 2x -CH_2-), 6.67 (s, br, 4H, 2x -NH_2), 6.87 (d, 2H, J = 8.6 Hz, H-5 and H-5`), 7.52 (dd, 2H, J = 8.6 Hz, J = 2.3 Hz, H-6 and H-6´), 7.90 (d, 2H, J = 2.3 Hz, H-2 and H-2´). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 25.9 (2C, 2x -CH_3), 68.3 (2C, 2x -CH_2-), 108.3 (2C, C-3 and C-3´), 117.4 (2C, C-5 and C-5´), 126.5 (2C, C-1 and C-1´), 126.9 (2C, C-2 and C-2´), 132.1 (2C, C-6 and C-6´), 150.2 (2C, C-4 and C-4´), 166.5 (2C, 2x COOH), 202.2 (2C, 2x –CO-). ESI-HRMS: m/z = 385.1396 $[M+H]^+$, calc. for $C_{20}H_{20}N_2O_6$ + H^+ = 385.1394; m/z = 407.1217 $[M+Na]^+$, calc. for $C_{20}H_{20}N_2O_6$ + Na^+ = 407.1214.

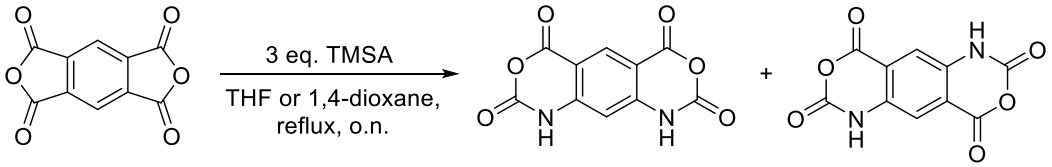
Synthesis of 3,3'-dihydroxy-2,2'-dimethyl-[6,6'-biquinoline]-4,4'(*1H,1'H*)-dione (**2**)



2

Compound **6** (1.0 g, 2.6 mmol) was dissolved in 10 mL NMP and the solution was refluxed for 2 h. After reaching room temperature, the solution was poured in ice-water and the precipitated collected by filtration. The slightly impure product (780 mg) was purified by washing with THF (5x 15 mL). The pure product was obtained as a light brown solid (680 mg, 75%). ^{1}H-NMR (DMSO-d_6 600.2 MHz) δ (ppm): 2.40 (s, 6H, 2x -CH_3), 7.63 (d, 2H, J = 8.7 Hz, H-8 and H-8`), 7.96 (dd, 2H, J = 8.7 Hz, J = 2.1 Hz, H-7 and H-7´), 8.16 (s, br, 2H, 2x –OH), 8.40 (d, 2H, J = 2.1 Hz, H-5 and H-5´), 11.62 (s, br, 2x –NH). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 14.1 (2C, 2x -CH_3), 118.6 (2C, C-8 and C-8´), 121.5 (2C, C-5 and C-5´), 122.6 (2C, C-4a and C-4a´), 128.6 (2C, C-7 and C-7´), 131.7 (2C, C-2 and C-2´), 132.8 (2C, C-6 and C-6´), 136.5 (2C, C-8a and C-8a´) 138.3 (C-3 and C-3´), 168.7 (C-4 and C-4´). ESI-HRMS: m/z = 349.1184 $[M+H]^+$, calc. for $C_{20}H_{16}N_2O_4$ + H^+ = 349.1183; m/z = 371.1007 $[M+Na]^+$, calc. for $C_{20}H_{16}N_2O_4$ + Na^+ = 371.1002; m/z = 719.2117 $[2M+Na]^+$, calc. for $C_{40}H_{32}N_4O_8$ + Na^+ = 719.2112.

Synthesis of 1,9-dihydro-2H,4H-benzo[1,2-d:5,4-d']bis([1,3]oxazine)-2,4,6,8-tetraone (**7a**) and 1,6-dihydrobenzo[1,2-d:4,5-d']bis([1,3]oxazine)-2,4,7,9-tetraone (**7b**)



7a **7b**

Pyromellitic dianhydride (1 eq.) was suspended in THF or 1,2-dioxane (0.8 M) and TMSA (3eq.) was added dropwise. The colourless mixture was refluxed overnight and turned yellow/orange. The mixture was allowed to cool to room temperature and the precipitated collected by centrifugation. The precipitate was washed with THF or 1,4-dioxane and dried under vacuum.

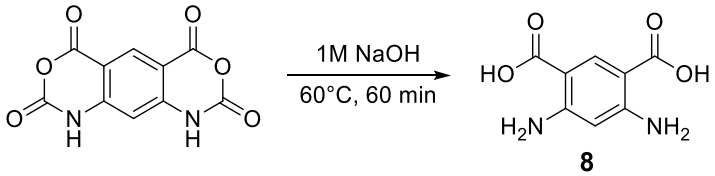
The reaction in THF resulted in a yellow solution with light-yellow precipitate containing a mixture of isomers **7a**:**7b** in a ratio of 1:0.85 (23 %) that could not be separated.

The reaction in 1,4-dioxane gave an orange reaction solution with white precipitate. After extensive washing of the precipitate with 1,4-dioxane, pure **7a** was obtained as white solid (21%). The supernatants were combined, the solvent evaporated and the precipitate washed again extensively with 1,4-dioxane. After several repetitions of this method a small amount of pure **7b** could be isolated as yellow solid for characterization.

1,9-dihydro-2H,4H-benzo[1,2-d:5,4-d']bis([1,3]oxazine)-2,4,6,8-tetraone (**7a**): ^{1}H-NMR (DMSO-d_6 400.13 MHz) δ (ppm): 6.79 (s, 1H, H-10), 8.31 (s, 1H, H-5), 12.09 (s, br, 2H, 2x –NH). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 99.4 (C-10), 106.7 (2C, C-4a and C-5a), 131.9 (C-5), 146.7 (2C, C-9a and C-10a), 146.8 (2C, C-2 and C-8), 158.4 (2C, C-4 and C-6). ESI-HRMS: m/z = 249.0149 $[M+H]^+$, calc. for $C_{10}H_4N_2O_6$ + H^+ = 249.0142.

1,6-dihydrobenzo[1,2-d:4,5-d']bis([1,3]oxazine)-2,4,7,9-tetraone (**7b**): ^{1}H-NMR (DMSO-d_6 400.13 MHz) δ (ppm): 7.60 (s, 2H, H-5 and H-10), 11.83 (s, 2H, 2x –NH). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 114.7 (2C, C-5 and C-10), 117.3 (2C, C-5a and C-10a), 136.0 (2C, C-4a and C-9a), 146.4 (2C, C-2 and C-7), 159.1 (2C, C-4 and C-9).

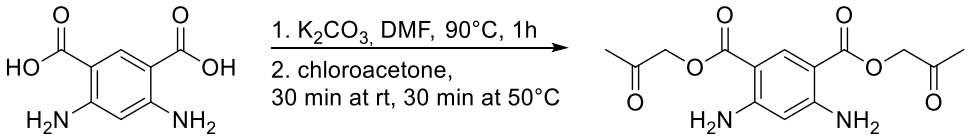
Synthesis of 4,6-diaminoisophthalic acid **8**



8

The compound **7a** was dissolved in 1 M NaOH (250 mM) while the reaction was warmed up to 60°C. The reaction was stirred at 60°C for 1 h and afterwards cooled with an ice-bath and brought to pH 4 with acetic acid. The precipitated was filtered, washed with water, and dried under vacuum. The product was obtained as a white solid (59 %).^{1}H-NMR (DMSO-d_6 400.13 MHz) δ (ppm): 5.84 (s, 1H, H-5), 6.89 (s, 4H, 2x –NH_2), 8.29 (s, 1H, H-2). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 97.2 (C-5), 100.7 (2C, C-1 and C-3), 137.9 (C-2), 154.9 (2C, C-4 and C-6), 168.9 (2C, 2x -COOH). ESI-HRMS: m/z = 197.0558 $[M+H]^+$, calc. for $C_8H_8N_2O_4$ + H^+ = 197.0557; m/z = 219.0377 $[M+Na]^+$, calc. for $C_8H_8N_2O_4$ + Na^+ = 219.0376; m/z = 415.0860 $[2M+Na]^+$, calc. for $C_{16}H_{16}N_4O_8$ + Na^+ 415.0860.

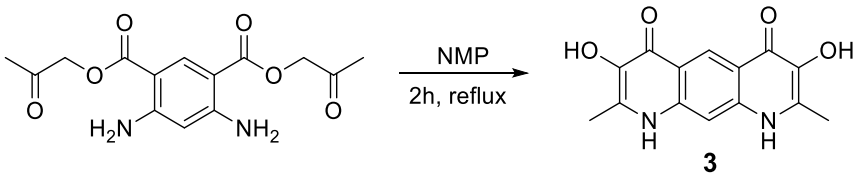
Synthesis of bis(2-oxopropyl) 4,6-diaminoisophthalate (**9**)



9

4,6-Diaminoisophthalic acid (**8**) (0.358 g, 1.825 mmol) was dissolved in 8 mL DMF and 378 mg K_2CO_3 (2.73 mmol, 1.5 eq.) was added. The mixture was heated at 90°C for 1 h. After reaching room temperature, 440 µL chloroacetone (5.475 mmol, 3.0 eq.) was added. The mixture was stirred for 30 min at room temperature and 30 min at 50°C. At room temperature, the mixture was poured in water and extracted with ethyl acetate. The combined organic phases were washed with water, dried over Na_2SO_4, filtered and the solvent evaporated. The residue was dissolved in small amounts of DCM and added on a silica gel column equilibrated with petrol ether/ethyl acetate 1:1. The residue was purified by column chromatography with petrol ether/ethyl acetate 1:1 and 1:2. The product was obtained as slightly yellow solid (226 mg, 40%). R_f = 0.23 (petrol ether/ethyl acetate 1:1). ^{1}H-NMR (DMSO-d_6 400.13 MHz) δ (ppm): 2.12 (s, 6H, 2x -CH_3), 4.89 (s, 4H, 2x -CH_2-), 5.94 (s, 1H, H-5), 6.92 (s, 4H, 2x –NH_2), 8.47 (s, 1H, H-2). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 25.8 (2C, 2x -CH_3), 67.9 (2C, 2x -CH_2-), 97.1 (C-5), 99.9 (2C, C-1 and C-3), 137.5 (C-2), 154.8 (2C, C-4 and C-6), 165.7 (2C, 2x –COO-), 202.4 (2C, 2x –CO-). ESI-HRMS: m/z = 331.0904 $[M+Na]^+$, calc. for $C_{14}H_{16}N_2O_6$ + Na^+ = 331.0901; m/z = 639.1905 $[2M+Na]^+$, calc. for $C_{28}H_{32}N_4O_{12}$ + Na^+ 639.1909.

Synthesis of 3,7-dihydroxy-2,8-dimethylpyrido[3,2-g]quinoline-4,6(1H,9H)-dione (**3**)



3

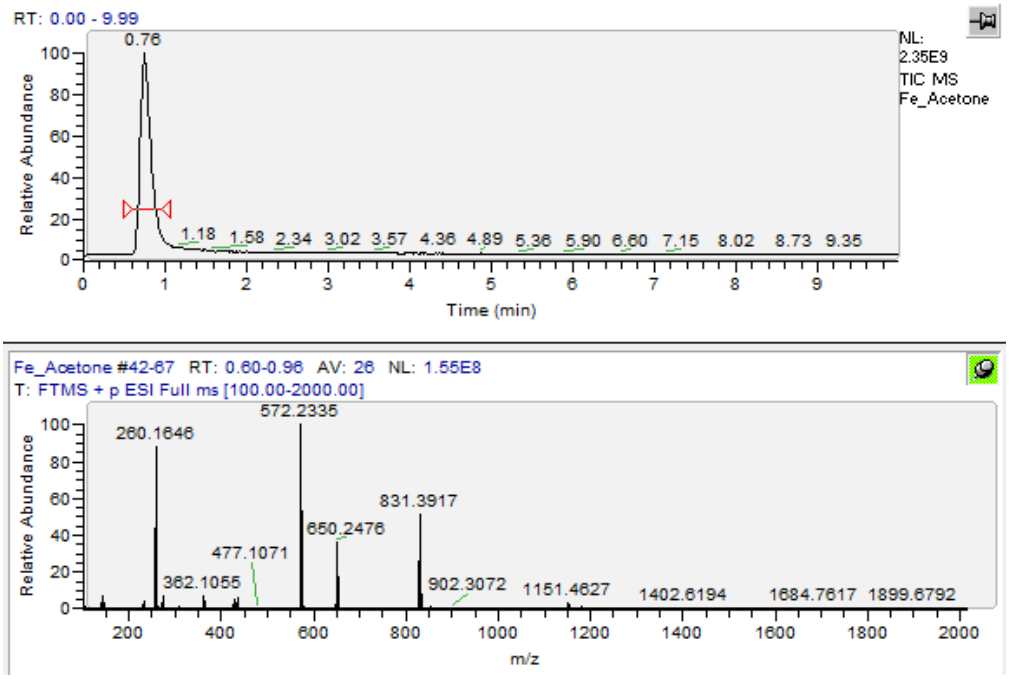
Bis(2-oxopropyl) 4,6-diaminoisophthalate **9** (1.46 g, 4.736 mmol) was dissolved in 20 mL NMP and refluxed for 2 h. The reaction was cooled to room temperature and the precipitate collected by filtration, washed with water, and dried. The product was obtained as a yellowish-brown solid (m = 789 mg, 61.2%). ^{1}H-NMR (DMSO-d_6 400.13 MHz) δ (ppm): 2.37 (s, 6H, 2x -CH_3), 7.47 (s, 1H, H-10), 8.06 (s, br, 2H, 2x -OH), 8.95 (s, 1H, H-5), 11.33 (s, 2H, 2x -NH). ^{13}C-NMR (DMSO-d_6 100.6 MHz) δ (ppm): 14.3 (2C, 2x -CH_3), 101.3 (C-10), 118.9 (2C, C-4a and C-5a), 123.6 (C-5), 133.4 (2C, C-2 and C-8), 136.2 (2C, C-3 and C-7), 138.2 (2C, C-9a and C-10a), 170.0 (2C, C-4 and C-6). ESI-HRMS: m/z = 295.0690 $[M+Na]^+$, calc. for $C_{14}H_{12}N_2O_4 + Na^+$ = 295.0689; m/z = 567.1482 $[2M+Na]^+$, calc. for $C_{28}H_{24}N_4O_8 + Na^+$ = 567.1486.

Table S1. Conditions used for formation experiments and resulting PQS-metal ion complexes.

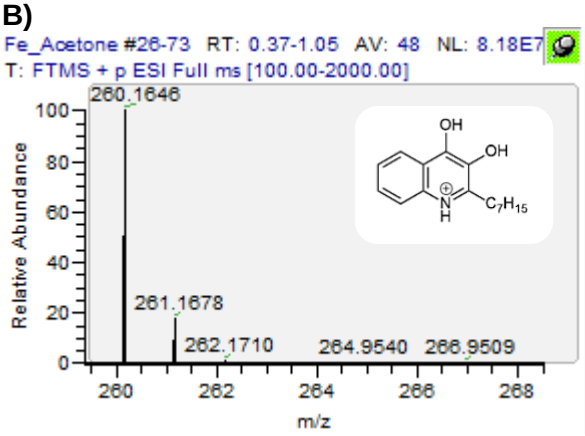
Salt	Final molarity of salt*, μM	Final molarity of PQS*, μM	Detection of metal-PQS complexes in MeOH	Detection of metal-PQS complexes in Acetone
$CuCl_2$	30	60	-	-
$ZnCl_2$	30	60	-	-
$NiCl_2$	30	60	-	-
$CoCl_2$	30	60	-	-
$MnCl_2$	30	60	-	-
$AlCl_3$	30	90	+	+
$FeCl_3$	30	90	+	+
Na_3VO_4	30	150	+	+
Na_2MoO_4	30	180	+	+

* Stock solutions were prepared in DMSO (DMSO/H_2O (1:1) for Na_2MoO_4).

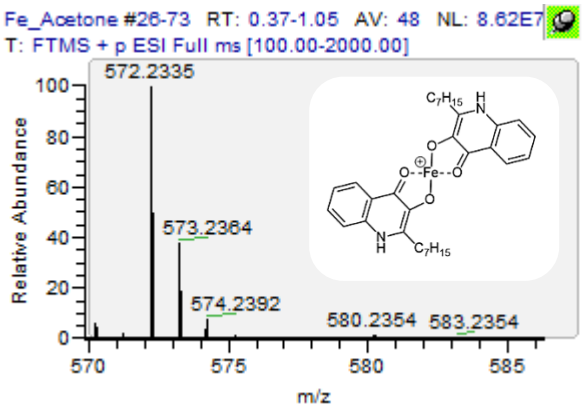
A)



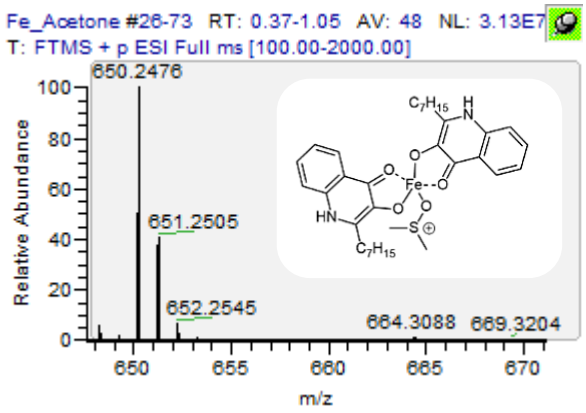
B)



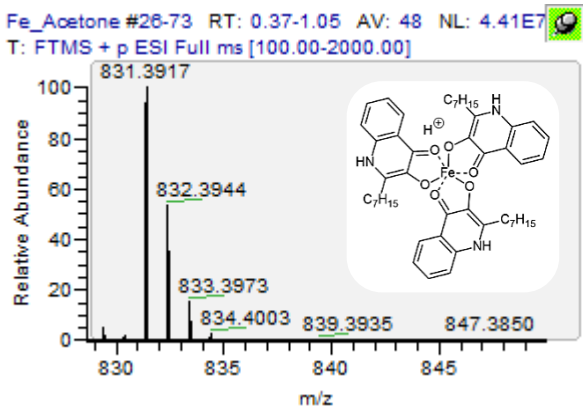
Calc. m/z: 260.1646 (100.0%), 261.1679 (17.3%), 262.1713 (1.4%).



Calc. m/z: 572.2332 (100.0%), 573.2366 (34.6%), 570.2379 (6.4%), 574.2400 (5.8%), 573.2337 (2.3%), 571.2413 (2.2%).



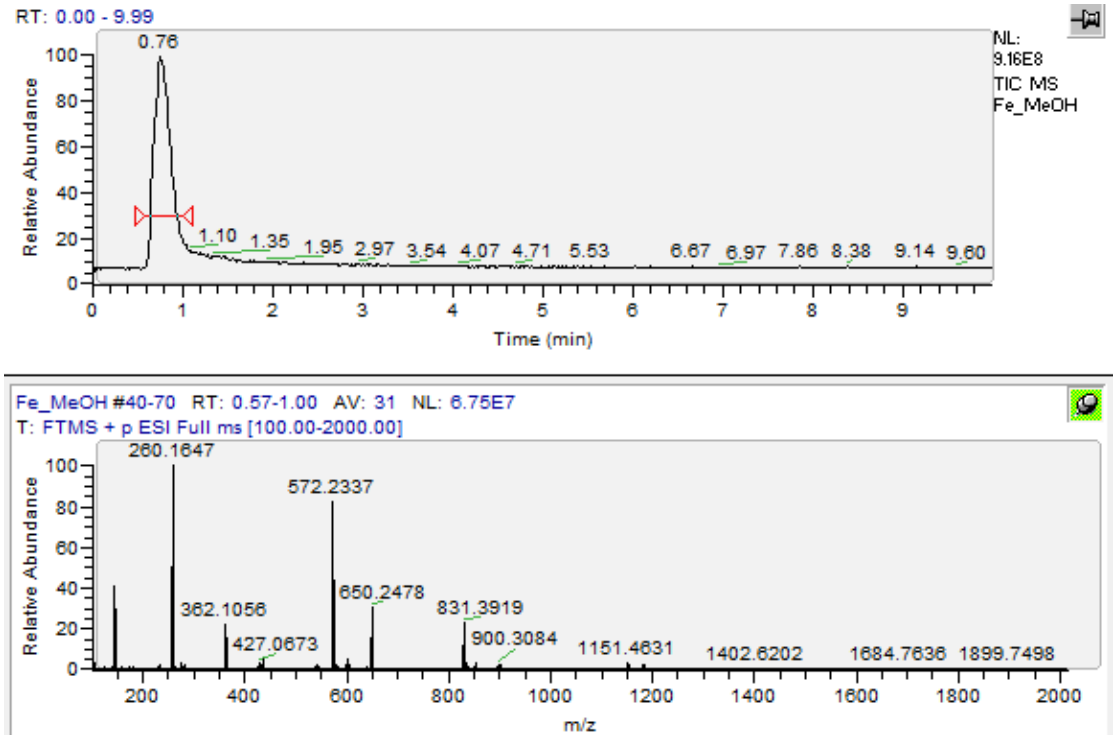
Calc. m/z: 650.2472 (100.0%), 651.2505 (36.8%), 652.2539 (6.6%), 648.2519 (6.4%), 652.2430 (4.5%), 649.2552 (2.3%), 651.2476 (2.3%), 653.2463 (1.7%), 652.2514 (1.0%).



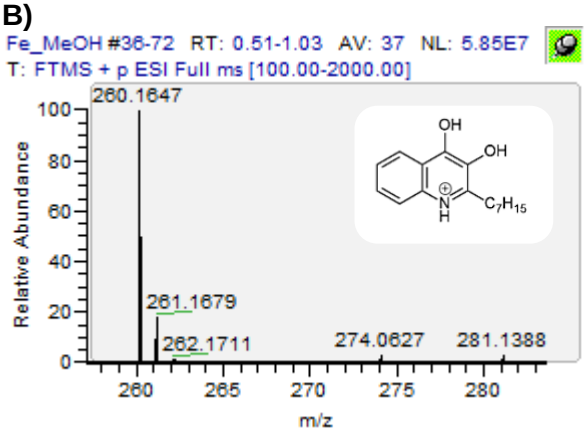
Calc. m/z: 831.3905 (100.0%), 832.3938 (51.9%), 833.3972 (13.2%), 829.3952 (6.4%), 830.3985 (3.3%), 832.3909 (2.3%), 834.4005 (2.2%), 833.3947 (1.2%), 833.3943 (1.2%), 832.3875 (1.1%).

Figure S1. **A)** Full HRMS-spectrum of Fe(III)-PQS complexes in acetone. **B)** Detected and calculated m/z for PQS and complexes. Insets: proposed complexes structures.

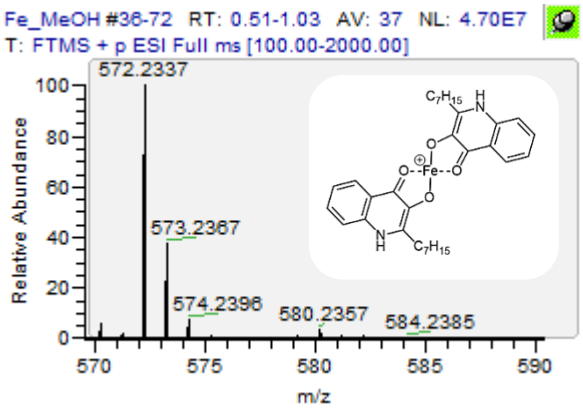
A)



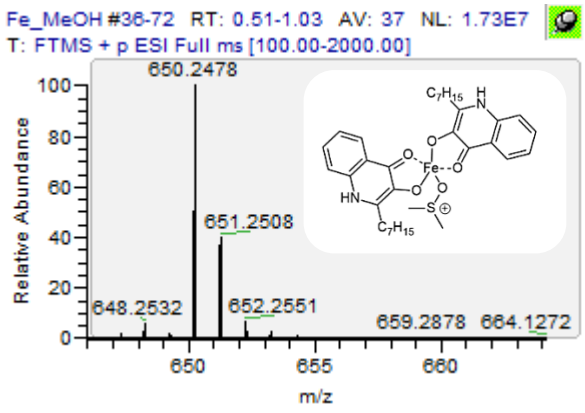
B)



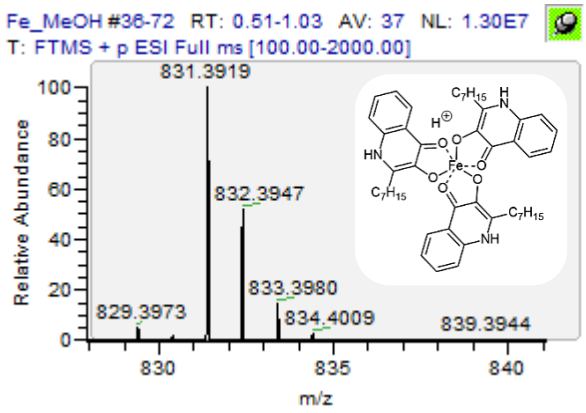
Calc. m/z: 260.1646 (100.0%), 261.1679 (17.3%), 262.1713 (1.4%).



Calc. m/z: 572.2332 (100.0%), 573.2366 (34.6%), 570.2379 (6.4%), 574.2400 (5.8%), 573.2337 (2.3%), 571.2413 (2.2%).



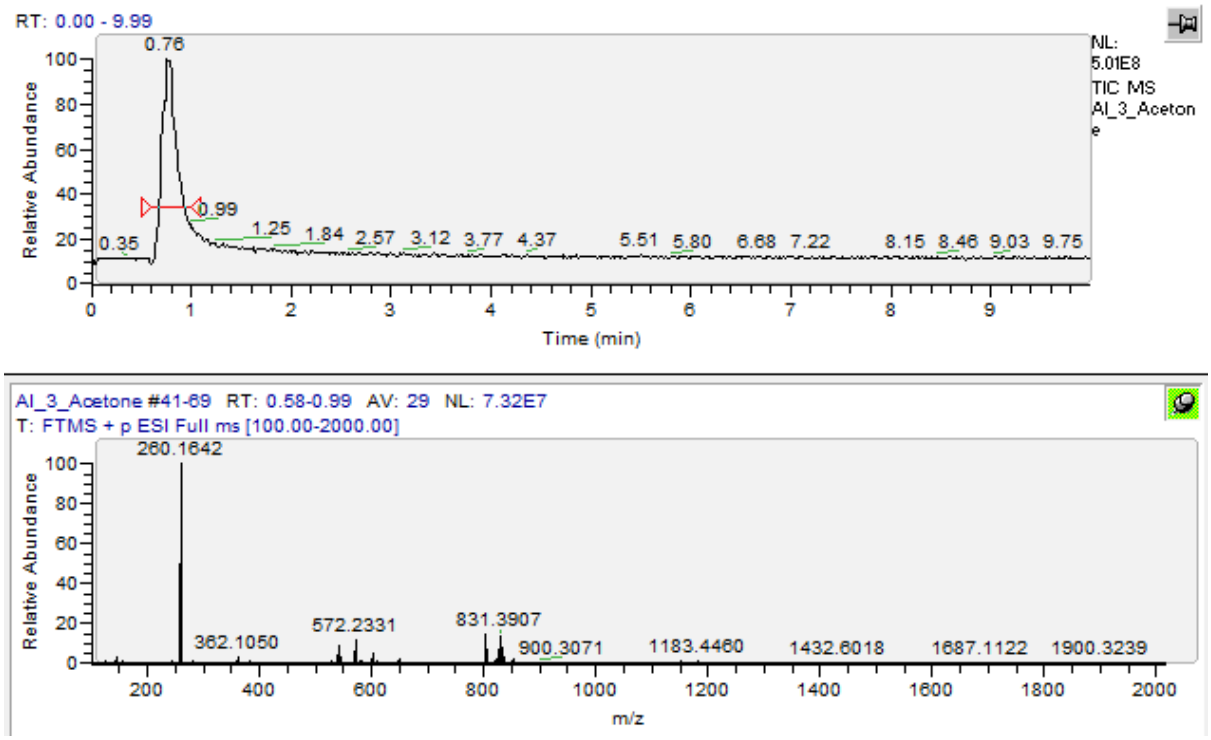
Calc. m/z: 650.2472 (100.0%), 651.2505 (36.8%), 652.2539 (6.6%), 648.2519 (6.4%), 652.2430 (4.5%), 649.2552 (2.3%), 651.2476 (2.3%), 653.2463 (1.7%), 652.2514 (1.0%).



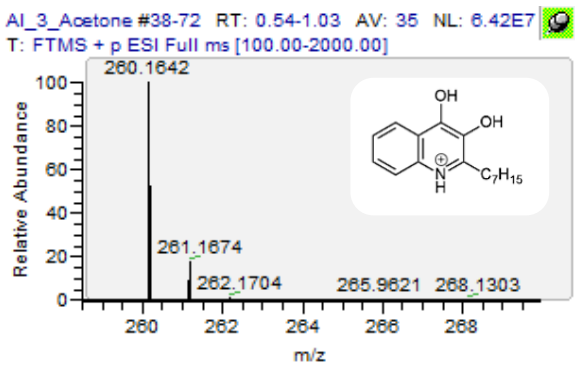
Calc. m/z: 831.3905 (100.0%), 832.3938 (51.9%), 833.3972 (13.2%), 829.3952 (6.4%), 830.3985 (3.3%), 832.3909 (2.3%), 834.4005 (2.2%), 833.3947 (1.2%), 833.3943 (1.2%), 832.3875 (1.1%).

Figure S2. **A)** Full HRMS-spectrum of Fe(III)-PQS complexes in methanol. **B)** Detected and calculated m/z for PQS and complexes. Insets: proposed complexes structures.

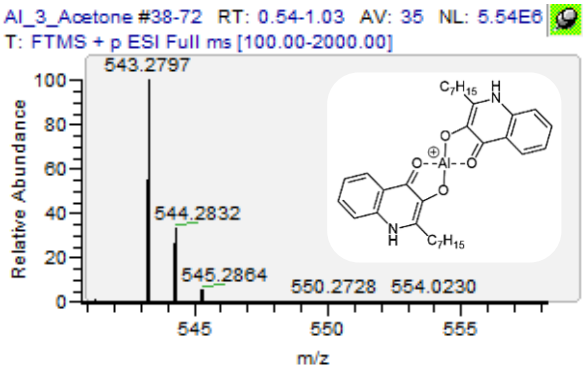
A)



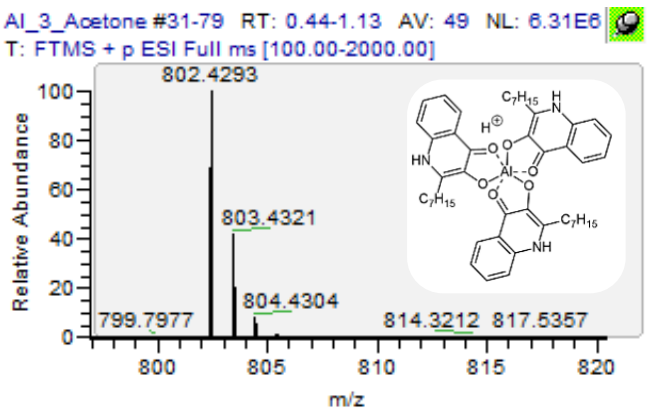
B)



Calc. m/z: 260.1646 (100.0%), 261.1679 (17.3%), 262.1713 (1.4%).



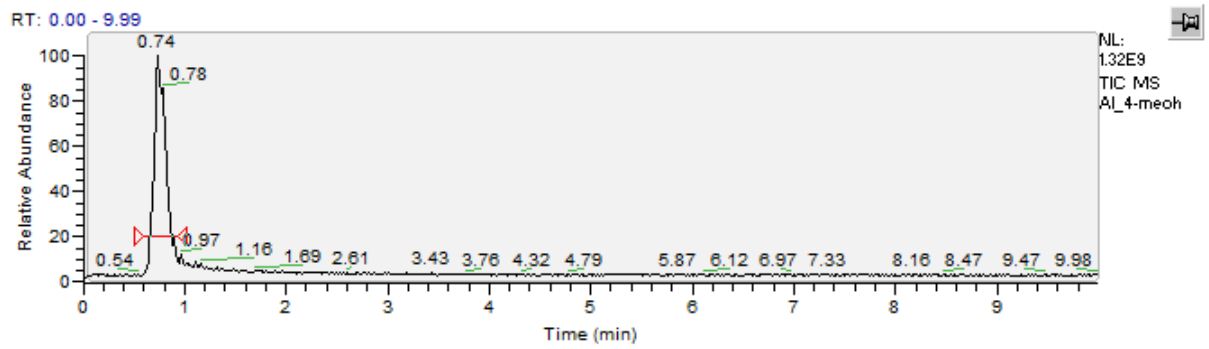
Calc. m/z: 543.2798 (100.0%), 544.2832 (34.6%), 545.2866 (5.8%).

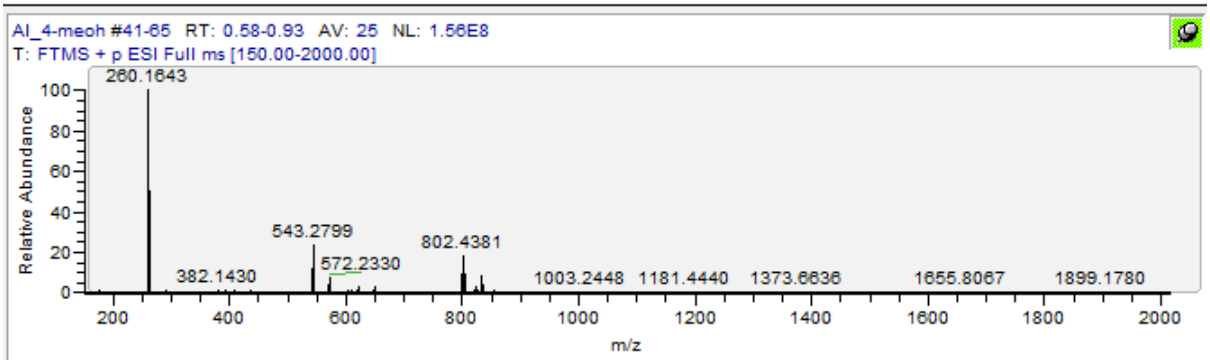


Calc. m/z: 802.4371 (100.0%), 803.4404 (51.9%), 804.4438 (13.2%), 805.4471 (2.2%), 804.4413 (1.2%), 803.4341 (1.1%).

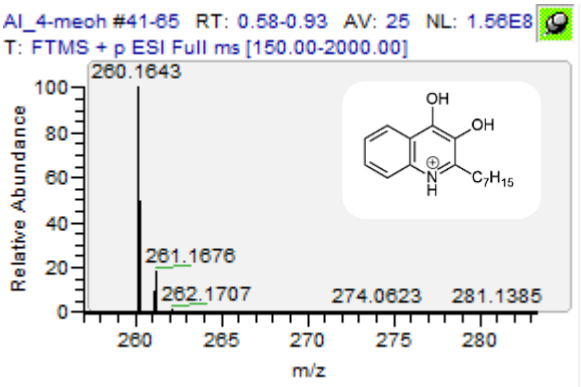
Figure S3. **A)** Full HRMS-spectrum of Al-PQS complexes in acetone. **B)** Detected and calculated m/z for PQS and complexes. Insets: proposed complexes structures.

A)

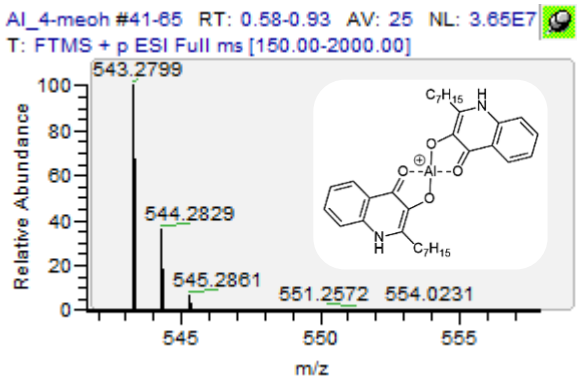




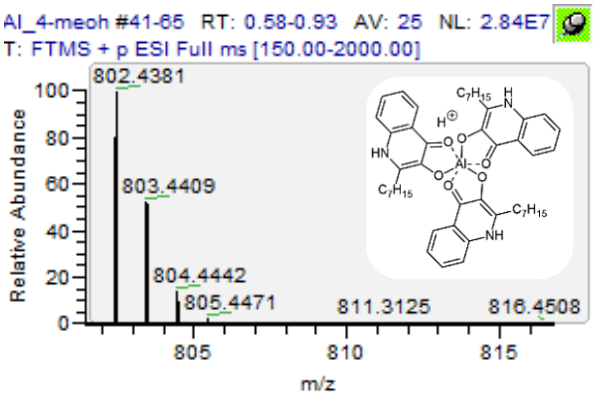
B)



Calc. m/z: 260.1646 (100.0%), 261.1679 (17.3%), 262.1713 (1.4%).



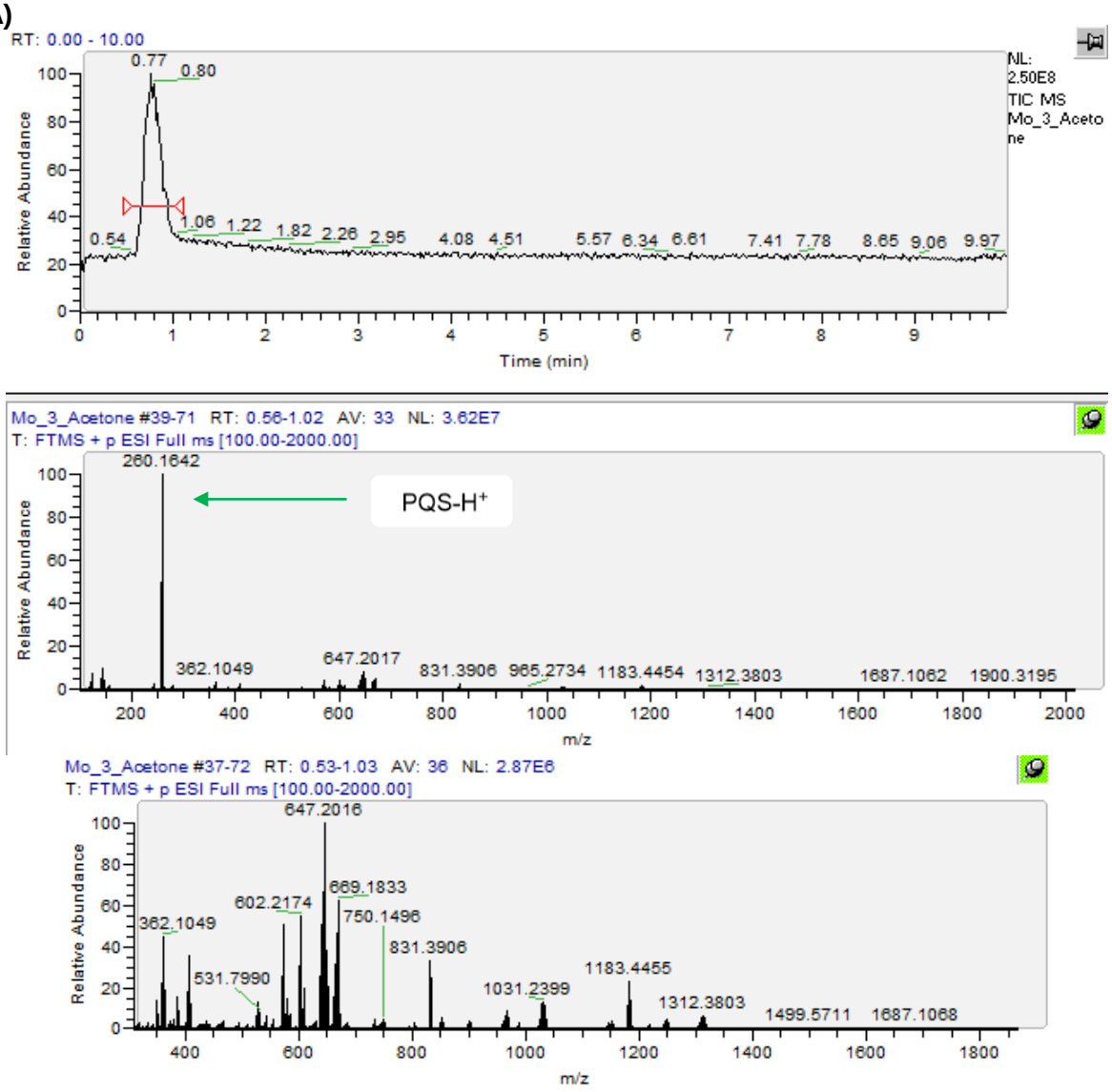
Calc. m/z: 543.2798 (100.0%), 544.2832 (34.6%), 545.2866 (5.8%).



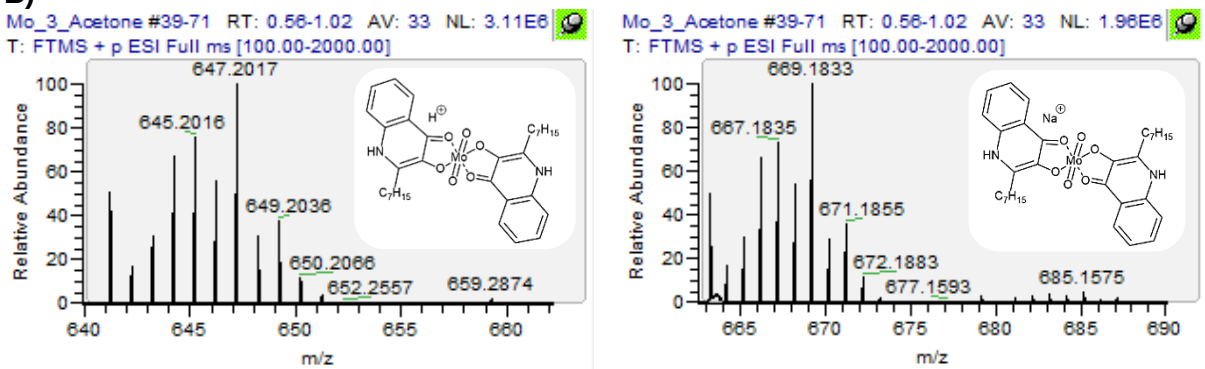
Calc. m/z: 802.4371 (100.0%), 803.4404 (51.9%), 804.4438 (13.2%), 805.4471 (2.2%), 804.4413 (1.2%), 803.4341 (1.1%).

Figure S4. **A)** Full HRMS-spectrum of Al-PQS complexes in methanol. **B)** Detected and calculated m/z for PQS and complexes. Insets: proposed complexes structures.

A)



B)

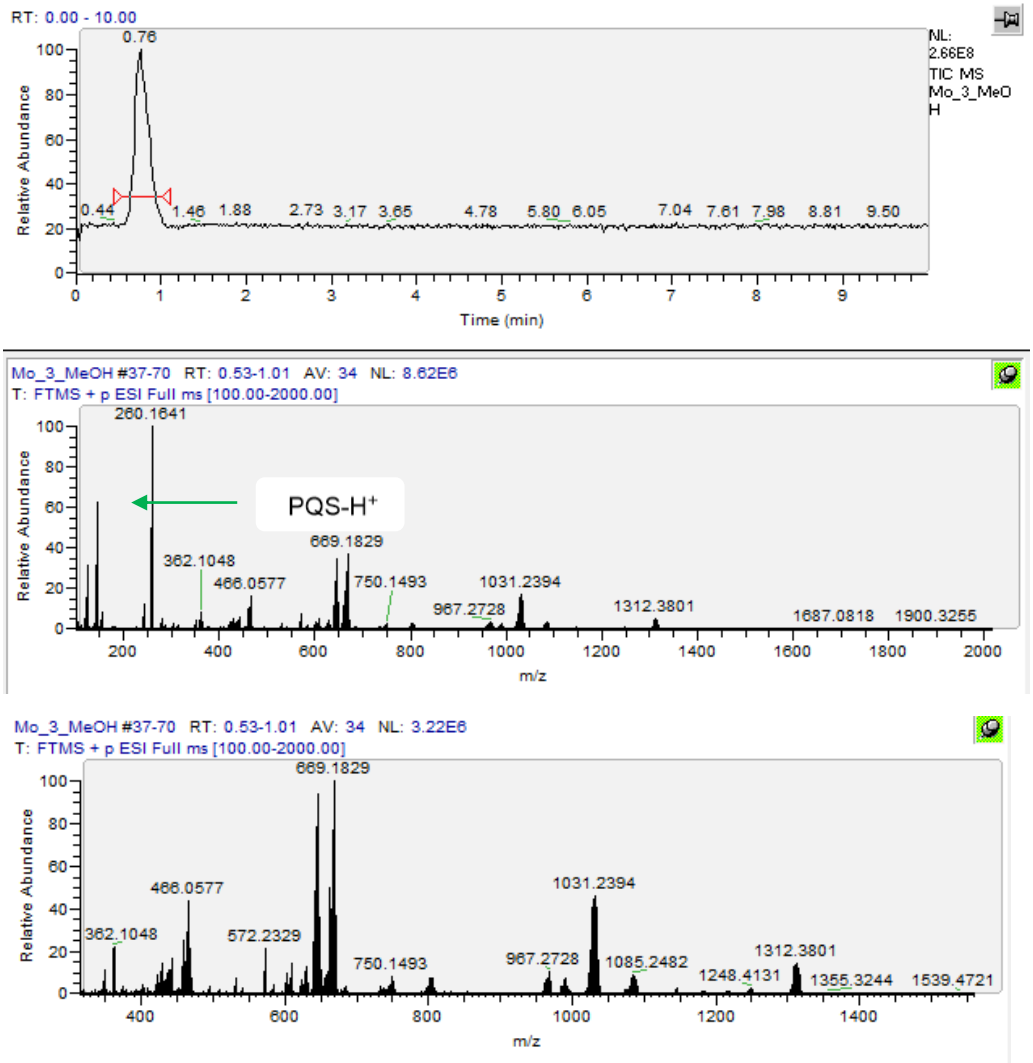


Calc. m/z: 647.2014 (100.0%), 645.2006 (69.1%), 644.2018 (66.0%), 641.2028 (61.5%), 649.2034 (39.9%), 646.2020 (39.6%), 643.2010 (38.3%), 648.2047 (34.6%), 646.2040 (23.9%), 645.2052 (22.8%), 642.2061 (21.3%), 650.2068 (13.8%), 647.2053 (13.7%), 644.2044 (13.3%), 649.2081 (5.8%), 647.2074 (4.0%), 646.2085 (3.8%), 643.2095 (3.6%), 651.2101 (2.3%), 648.2087 (2.3%), 645.2078 (2.2%), 649.2056 (1.2%).

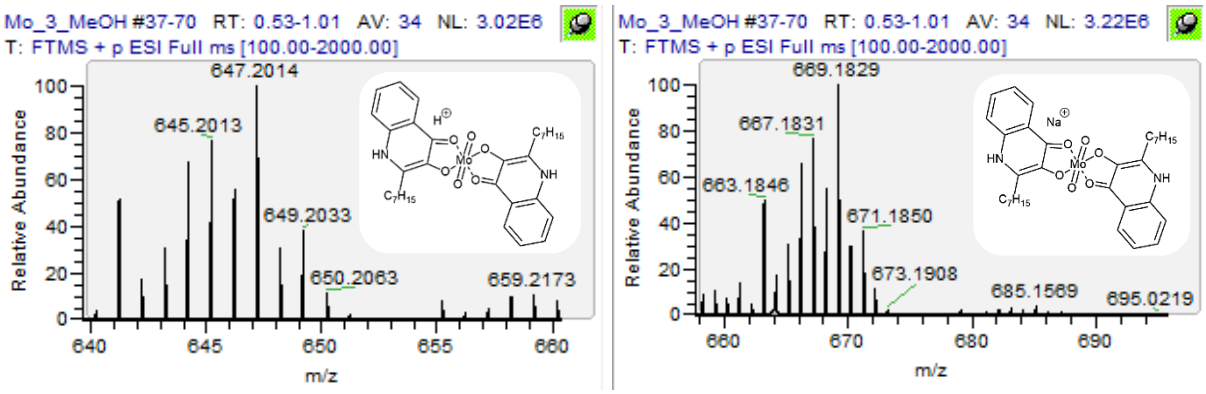
Calc. m/z: 669.1833 (100.0%), 667.1826 (69.1%), 666.1837 (66.0%), 663.1847 (61.5%), 671.1854 (39.9%), 668.1839 (39.6%), 665.1830 (38.3%), 670.1867 (34.6%), 668.1859 (23.9%), 667.1871 (22.8%), 664.1881 (21.3%), 672.1887 (13.8%), 669.1873 (13.7%), 666.1863 (13.3%), 671.1900 (5.8%), 669.1893 (4.0%), 668.1905 (3.8%), 665.1914 (3.6%), 673.1921 (2.3%), 670.1906 (2.3%), 667.1897 (2.2%), 671.1876 (1.2%).

Figure S5. **A)** Full HRMS-spectrum of Mo(VI)-PQS complexes in acetone. **B)** Detected and calculated m/z for complexes and their proposed structures.

A)



B)

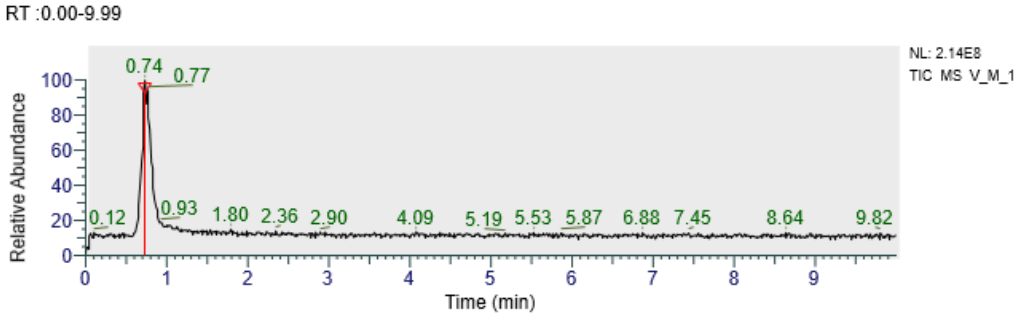


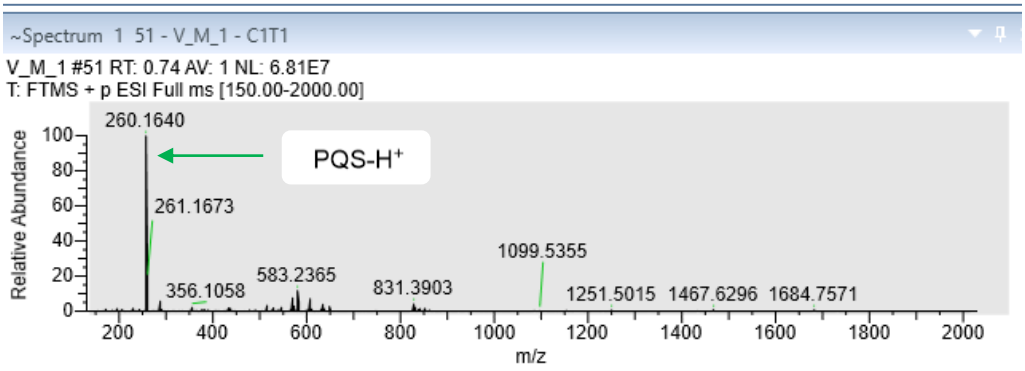
Calc. m/z: 647.2014 (100.0%), 645.2006 (69.1%), 644.2018 (66.0%), 641.2028 (61.5%), 649.2034 (39.9%), 646.2020 (39.6%), 643.2010 (38.3%), 648.2047 (34.6%), 646.2040 (23.9%), 645.2052 (22.8%), 642.2061 (21.3%), 650.2068 (13.8%), 647.2053 (13.7%), 644.2044 (13.3%), 649.2081 (5.8%), 647.2074 (4.0%), 646.2085 (3.8%), 643.2095 (3.6%), 651.2101 (2.3%), 648.2087 (2.3%), 645.2078 (2.2%), 649.2056 (1.2%).

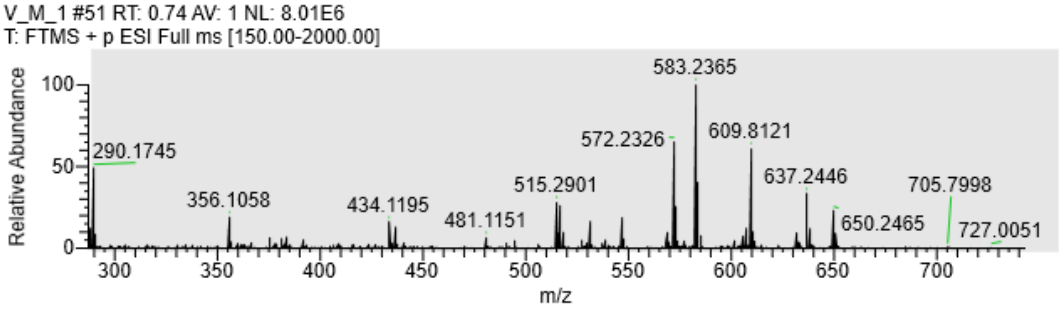
Calc. m/z: 669.1833 (100.0%), 667.1826 (69.1%), 666.1837 (66.0%), 663.1847 (61.5%), 671.1854 (39.9%), 668.1839 (39.6%), 665.1830 (38.3%), 670.1867 (34.6%), 668.1859 (23.9%), 667.1871 (22.8%), 664.1881 (21.3%), 672.1887 (13.8%), 669.1873 (13.7%), 666.1863 (13.3%), 671.1900 (5.8%), 669.1893 (4.0%), 668.1905 (3.8%), 665.1914 (3.6%), 673.1921 (2.3%), 670.1906 (2.3%), 667.1897 (2.2%), 671.1876 (1.2%)

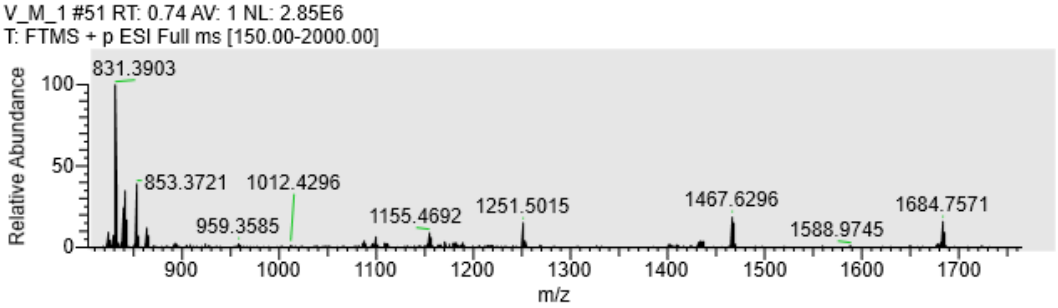
Figure S6. **A)** Full HRMS-spectrum of Mo (VI)-PQS complexes in methanol. **B)** Detected and calculated m/z for complexes and their proposed structures.

A)

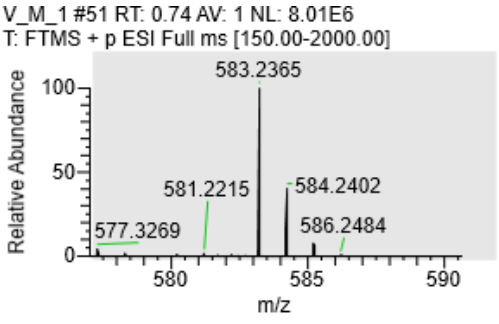

RT :0.00-9.99
NL: 2.14E8
TIC MS V_M_1
Relative Abundance
Time (min)
0.74
0.77
0.12
0.93
1.80
2.36
2.90
4.09
5.19
5.53
5.87
6.88
7.45
8.64
9.82

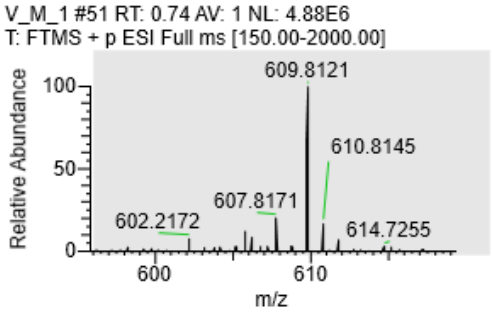

~Spectrum 1 51 - V_M_1 - C1T1
V_M_1 #51 RT: 0.74 AV: 1 NL: 6.81E7
T: FTMS + p ESI Full ms [150.00-2000.00]
Relative Abundance
m/z
260.1640
PQS-H+
261.1673
1099.5355
356.1058
583.2365
831.3903
1251.5015
1467.6296
1684.7571


V_M_1 #51 RT: 0.74 AV: 1 NL: 8.01E6
T: FTMS + p ESI Full ms [150.00-2000.00]
Relative Abundance
m/z
583.2365
290.1745
572.2326
609.8121
637.2446
705.7998
356.1058
434.1195
515.2901
481.1151
650.2465
727.0051


V_M_1 #51 RT: 0.74 AV: 1 NL: 2.85E6
T: FTMS + p ESI Full ms [150.00-2000.00]
Relative Abundance
m/z
831.3903
853.3721
1012.4296
959.3585
1155.4692
1251.5015
1467.6296
1588.9745
1684.7571

B)


V_M_1 #51 RT: 0.74 AV: 1 NL: 8.01E6
T: FTMS + p ESI Full ms [150.00-2000.00]
Relative Abundance
m/z
583.2365
581.2215
584.2402
577.3269
586.2484


V_M_1 #51 RT: 0.74 AV: 1 NL: 4.88E6
T: FTMS + p ESI Full ms [150.00-2000.00]
Relative Abundance
m/z
609.8121
610.8145
607.8171
602.2172
614.7255

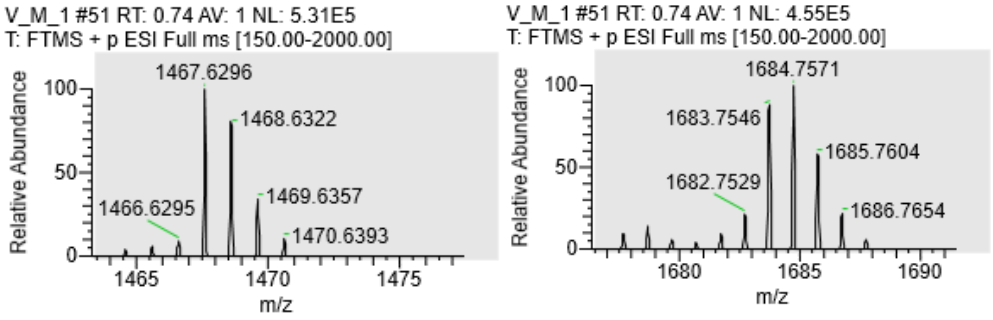


Figure S7. **A)** Full HRMS-spectrum of V (V)-PQS complexes in methanol. B) Detailed excerpts of unidentified V (V)-PQS complexes.

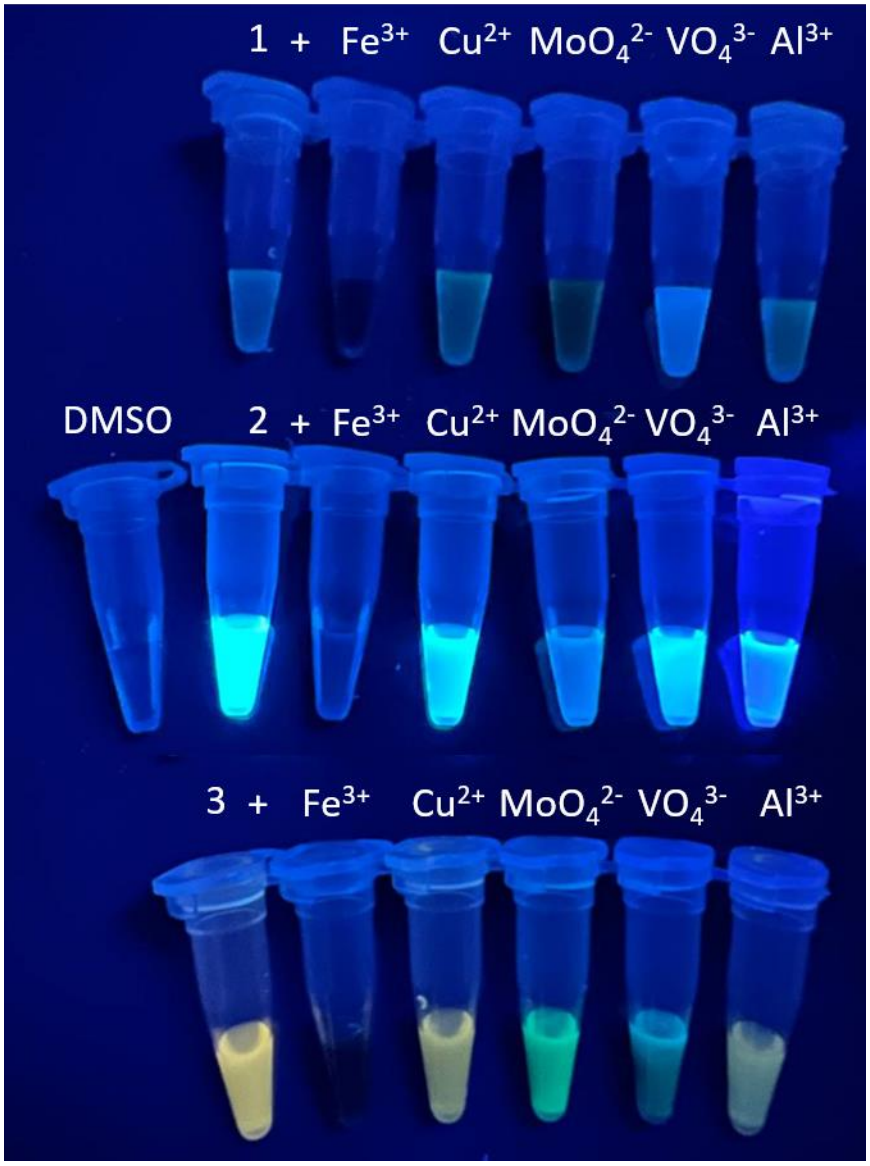
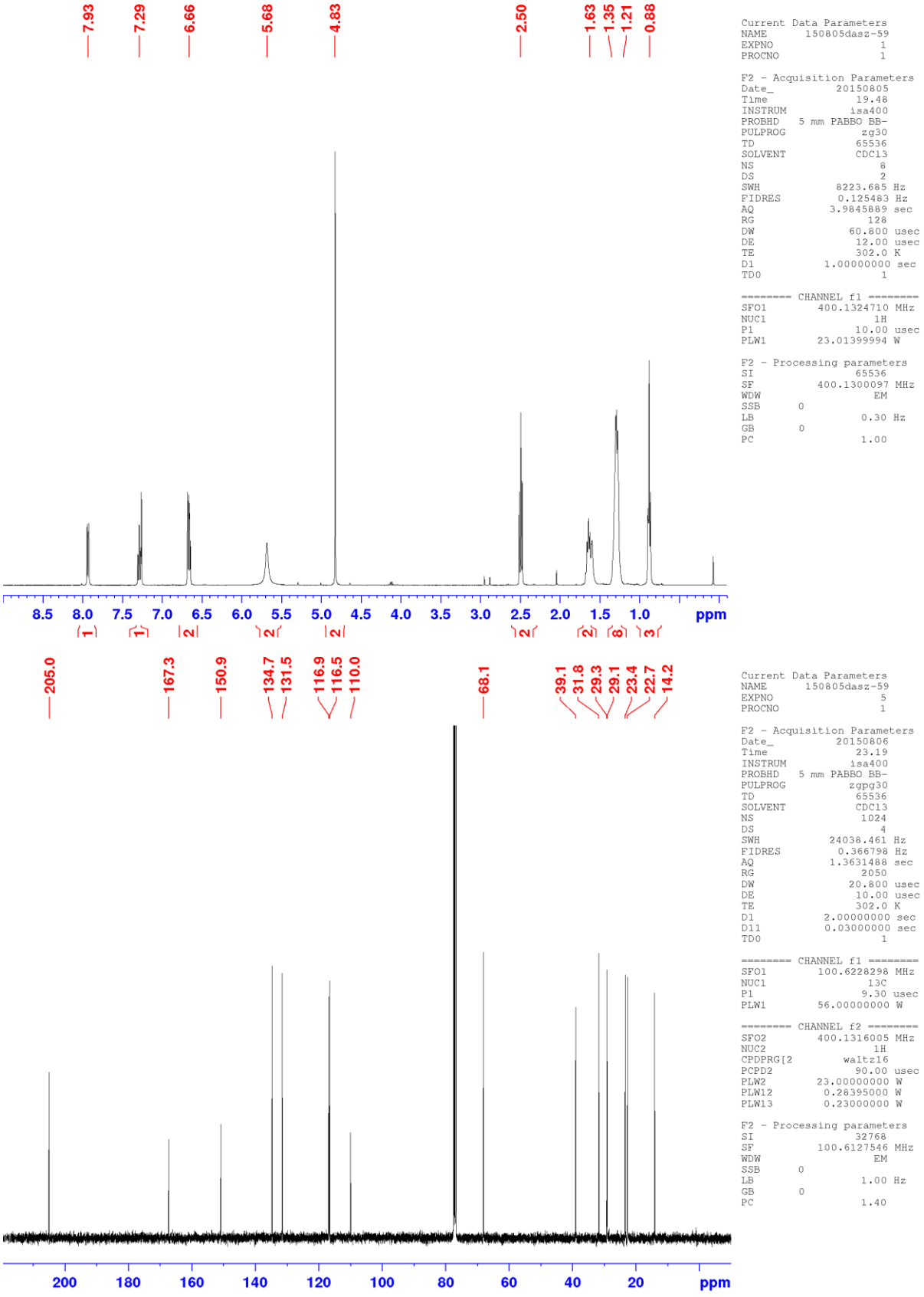


Figure S8. DMSO solutions of compound **1** (top), **2** (middle) and **3** (bottom) with metal ions under UV light (365 nm).

NMR spectra

^{1}H- and ^{13}C-NMR spectra of **4** in $CDCl_3$

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EXPNO                 1
PROCNO                1

F2 - Acquisition Parameters
Date_           20150805
Time              19.48
INSTRUM          isa400
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PULPROG            zg30
TD                65536
SOLVENT            CDCl3
NS                    8
DS                    2
SWH            8223.685 Hz
FIDRES         0.125483 Hz
AQ             3.9845889 sec
RG                  128
DW               60.800 usec
DE               12.00 usec
TE                302.0 K
D1            1.00000000 sec
TD0                   1

======== CHANNEL f1 ========
SFO1         400.1324710 MHz
NUC1                 1H
P1                10.00 usec
PLW1         23.01399994 W

F2 - Processing parameters
SI                 65536
SF           400.1300097 MHz
WDW                   EM
SSB       0
LB             0.30 Hz
GB        0
PC                 1.00
```

```
Current Data Parameters
NAME       150805dasz-59
EXPNO                 5
PROCNO                1

F2 - Acquisition Parameters
Date_           20150806
Time              23.19
INSTRUM          isa400
PROBHD   5 mm PABBO BB-
PULPROG          zgpg30
TD                65536
SOLVENT            CDCl3
NS                 1024
DS                    4
SWH           24038.461 Hz
FIDRES         0.366798 Hz
AQ             1.3631488 sec
RG                 2050
DW               20.800 usec
DE               10.00 usec
TE                302.0 K
D1            2.00000000 sec
D11           0.03000000 sec
TD0                   1

======== CHANNEL f1 ========
SFO1         100.6228298 MHz
NUC1                 13C
P1                9.30 usec
PLW1         56.00000000 W

======== CHANNEL f2 ========
SFO2         400.1316005 MHz
NUC2                 1H
CPDPRG[2          waltz16
PCPD2             90.00 usec
PLW2         23.00000000 W
PLW12         0.28395000 W
PLW13         0.23000000 W

F2 - Processing parameters
SI                 32768
SF           100.6127546 MHz
WDW                   EM
SSB       0
LB             1.00 Hz
GB        0
PC                 1.40
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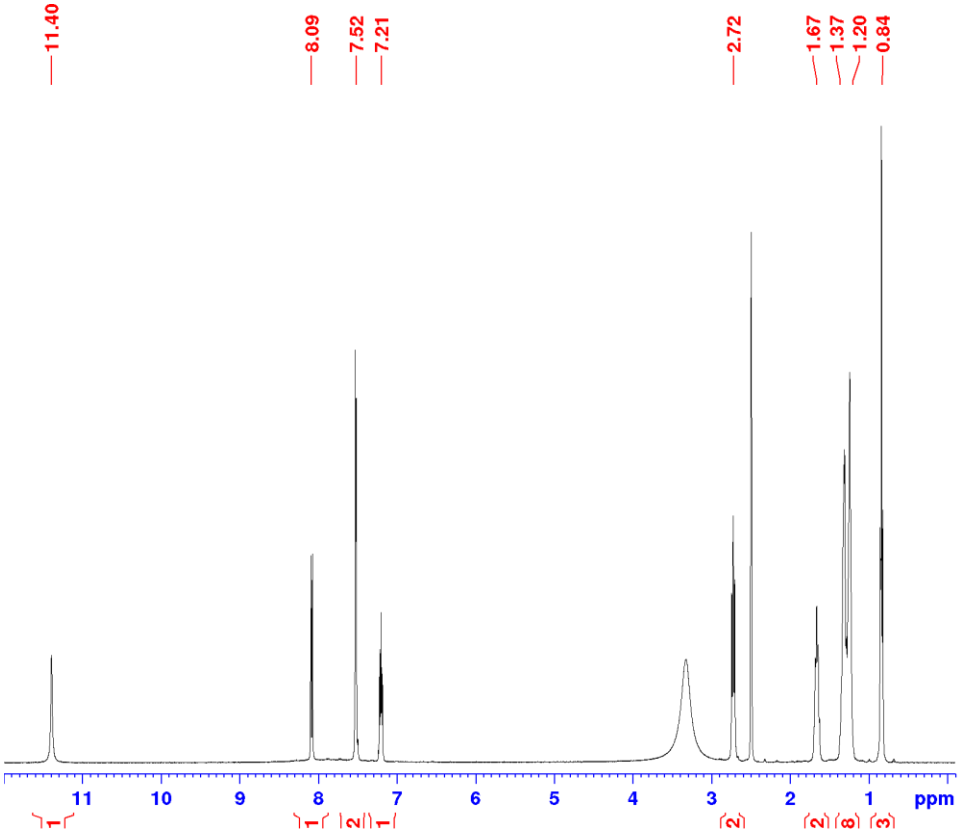
^{1}H- and ^{13}C-NMR spectra of **1** in DMSO-d_6

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EXPNO                  1
PROCNO                 1

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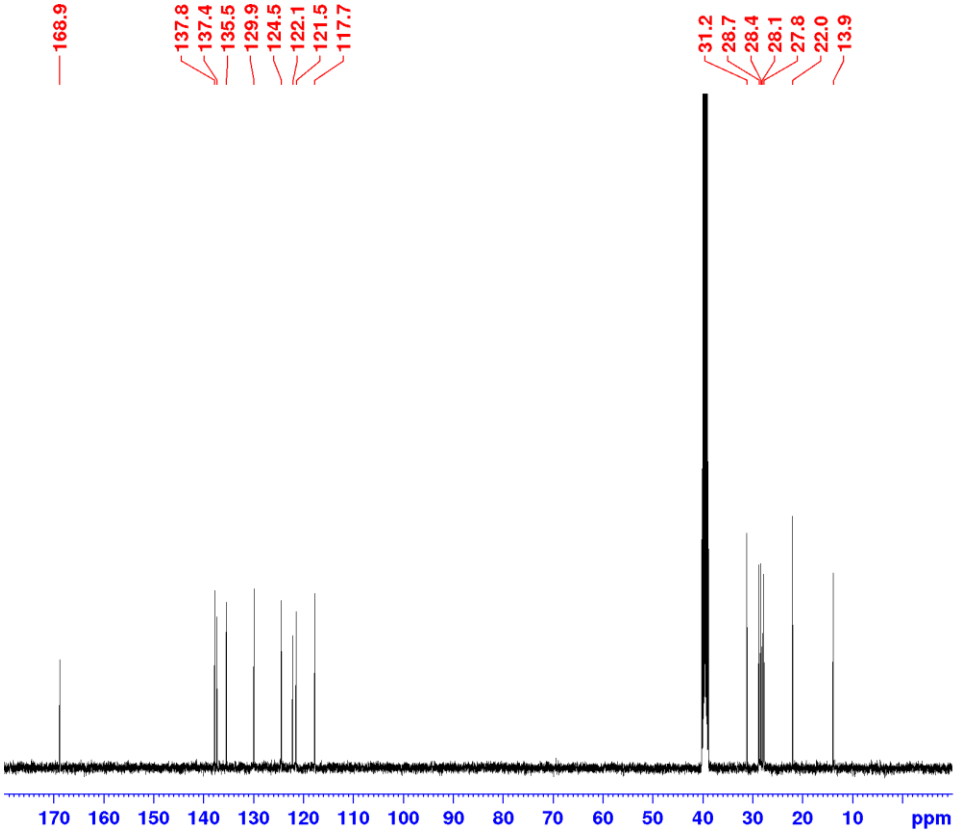
Date_           20150807
Time              20.28
INSTRUM           isa400
PROBHD    5 mm PABBO BB-
PULPROG             zg30
TD                 65536
SOLVENT             DMSO
NS                     8
DS                     2
SWH            8223.685 Hz
FIDRES          0.125483 Hz
AQ             3.9845889 sec
RG                   128
DW                60.800 usec
DE                12.00 usec
TE                302.1 K
D1             1.00000000 sec
TD0                    1

======== CHANNEL f1 ========
SFO1         400.1324710 MHz
NUC1                   1H
P1                 10.00 usec
PLW1         23.01399994 W

F2 - Processing parameters
SI                 65536
SF           400.1300054 MHz
WDW                   EM
SSB       0
LB                 0.30 Hz
GB        0
PC                 1.00
```




```
Current Data Parameters
NAME      150807dasz-18 (new)
EXPNO                  5
PROCNO                 1

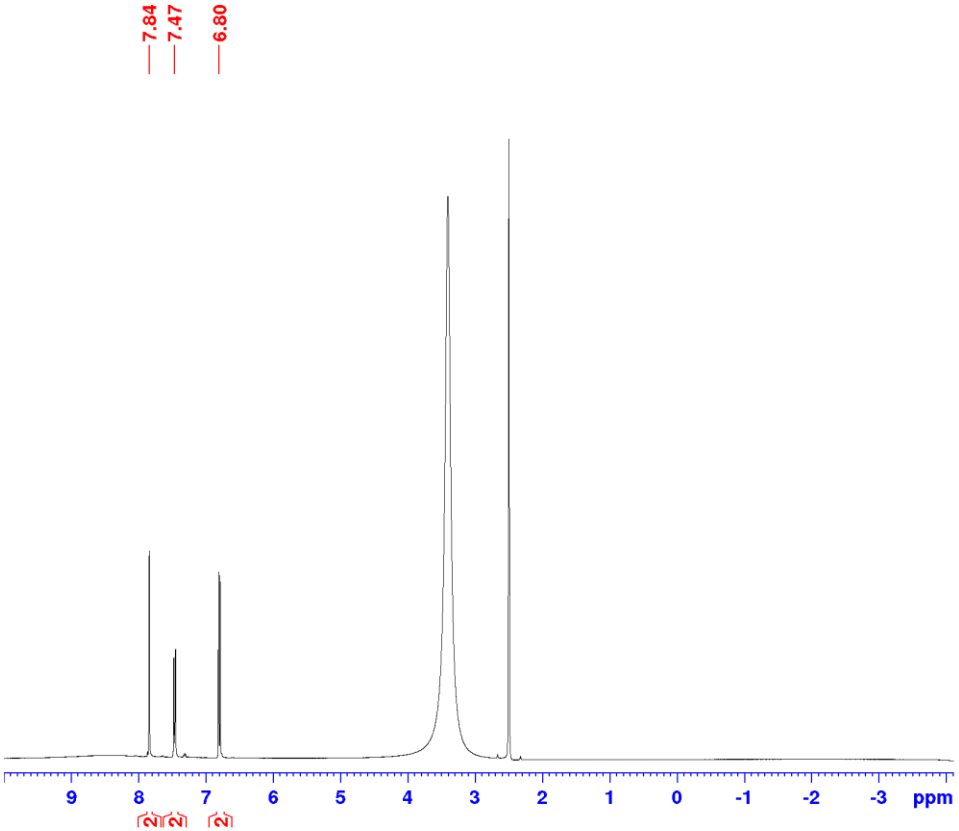
F2 - Acquisition Parameters
Date_           20150808
Time              12.06
INSTRUM           isa400
PROBHD    5 mm PABBO BB-
PULPROG           zgpg30
TD                 65536
SOLVENT             DMSO
NS                  1024
DS                     4
SWH           24038.461 Hz
FIDRES         0.366798 Hz
AQ             1.3631488 sec
RG                  2050
DW                20.800 usec
DE                10.00 usec
TE                302.0 K
D1             2.00000000 sec
D11            0.03000000 sec
TD0                    1

======== CHANNEL f1 ========
SFO1         100.6228298 MHz
NUC1                  13C
P1                 9.30 usec
PLW1         56.00000000 W

======== CHANNEL f2 ========
SFO2         400.1316005 MHz
NUC2                   1H
CPDPRG[2          waltz16
PCPD2             90.00 usec
PLW2         23.00000000 W
PLW12         0.28395000 W
PLW13         0.23000000 W

F2 - Processing parameters
SI                 32768
SF           100.6128183 MHz
WDW                   EM
SSB       0
LB                 1.00 Hz
GB        0
PC                 1.40
```

^{1}H- and ^{13}C-NMR spectra of **5** in DMSO-d_6

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PROCNO                 1

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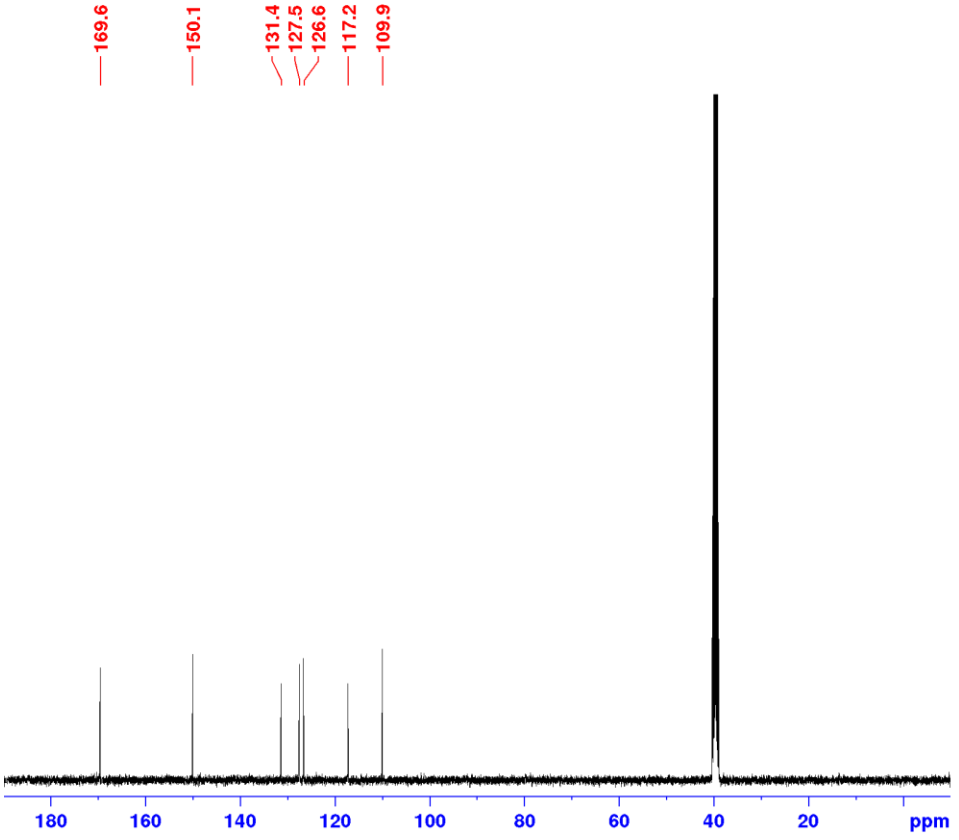
Date_           20170205
Time              16.30
INSTRUM          isa400
PROBHD   5 mm PABBO BB-
PULPROG           zg30
TD               65536
SOLVENT           DMSO
NS                  32
DS                   2
SWH           8223.685 Hz
FIDRES         0.125483 Hz
AQ            3.9845889 sec
RG                90.5
DW              60.800 usec
DE              10.00 usec
TE              300.0 K
D1           1.00000000 sec
TD0                  1

======== CHANNEL f1 ========
SFO1        400.1324710 MHz
NUC1               1H
P1               12.50 usec
PLW1        23.01399994 W

F2 - Processing parameters
SI               65536
SF          400.1300036 MHz
WDW                EM
SSB       0
LB              0.30 Hz
GB        0
PC              1.00
```

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EXPNO                  5
PROCNO                 1

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Time               5.44
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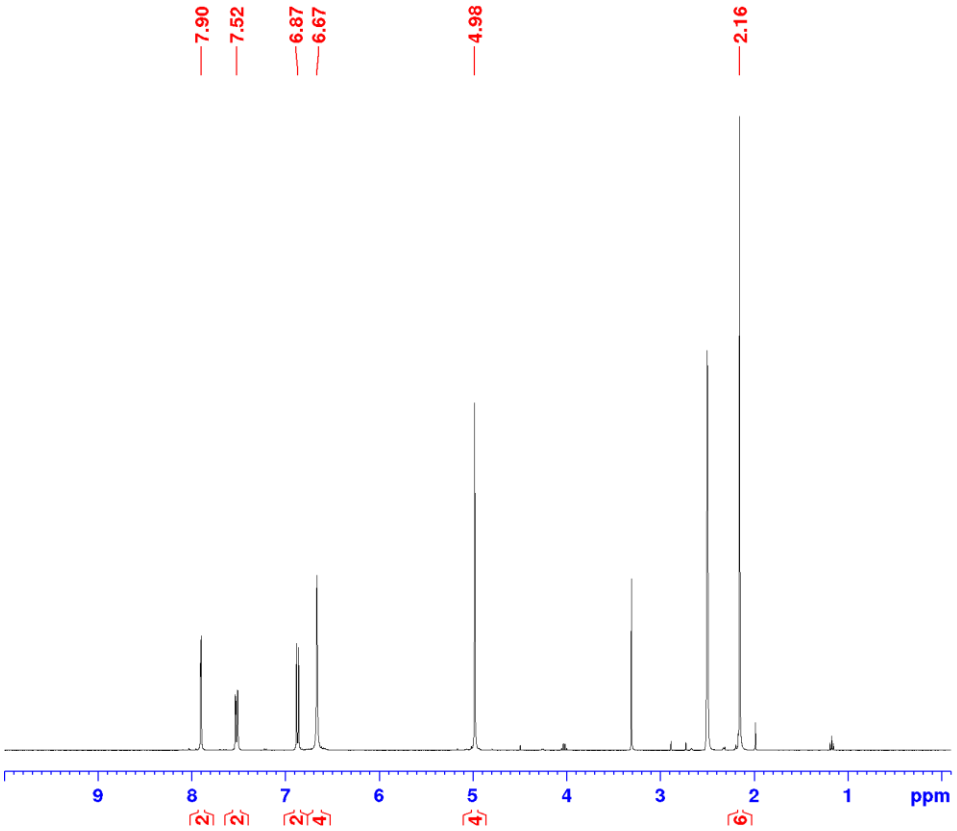
PULPROG         zgpg30
TD               65536
SOLVENT           DMSO
NS                1024
DS                   4
SWH          24038.461 Hz
FIDRES         0.366798 Hz
AQ            1.3631488 sec
RG                2050
DW              20.800 usec
DE              10.00 usec
TE              300.0 K
D1           2.00000000 sec
D11          0.03000000 sec
TD0                  1

======== CHANNEL f1 ========
SFO1        100.6228298 MHz
NUC1               13C
P1                9.25 usec
PLW1        56.00000000 W

======== CHANNEL f2 ========
SFO2        400.1316005 MHz
NUC2               1H
CPDPRG[2         waltz16
PCPD2            90.00 usec
PLW2        23.00000000 W
PLW12        0.44367000 W
PLW13        0.22316000 W

F2 - Processing parameters
SI               32768
SF          100.6128098 MHz
WDW                EM
SSB       0
LB              1.00 Hz
GB        0
PC              1.40
```

^{1}H- and ^{13}C-NMR spectra of **6** in DMSO-d_6

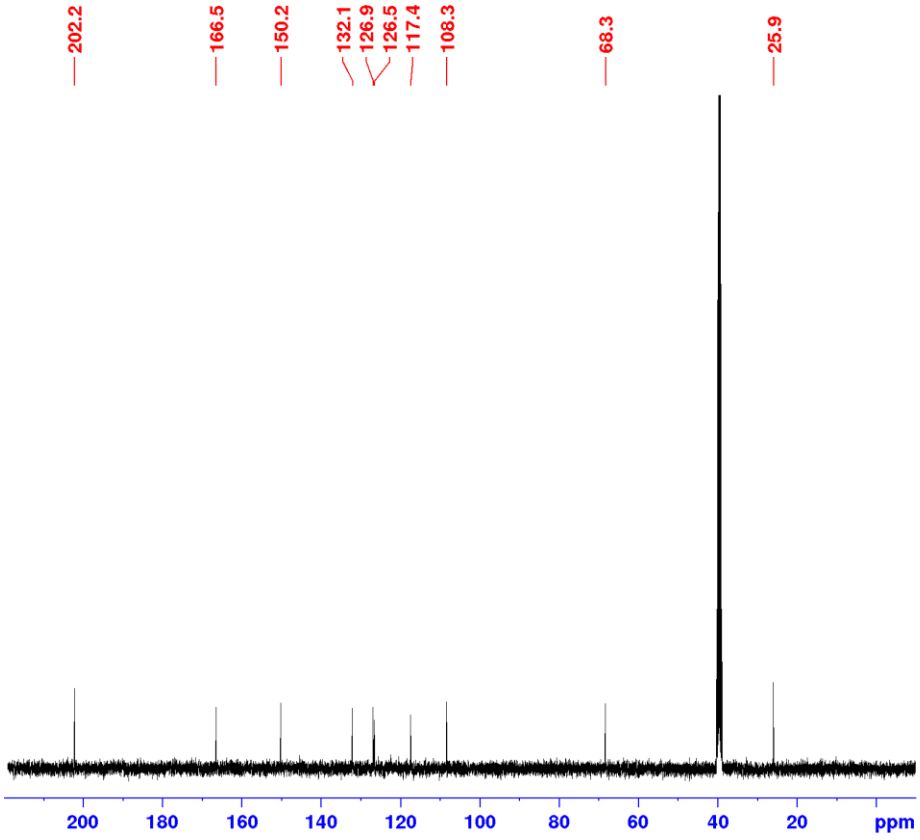



```
Current Data Parameters
NAME       170206dasz-54
EXPNO                 1
PROCNO                1

F2 - Acquisition Parameters
Date_           20170206
Time              20.34
INSTRUM          isa400
PROBHD   5 mm PABBO BB-
PULPROG            zg30
TD                65536
SOLVENT            DMSO
NS                    8
DS                    2
SWH            8223.685 Hz
FIDRES        0.125483 Hz
AQ            3.9845889 sec
RG                  287
DW               60.800 usec
DE               10.00 usec
TE                300.1 K
D1            1.00000000 sec
TD0                   1

======== CHANNEL f1 ========
SFO1         400.1324710 MHz
NUC1                 1H
P1                12.50 usec
PLW1         23.01399994 W

F2 - Processing parameters
SI                65536
SF           400.1300036 MHz
WDW                  EM
SSB       0
LB                0.30 Hz
GB        0
PC                1.00
```




```
Current Data Parameters
NAME       170206dasz-54
EXPNO                 3
PROCNO                1

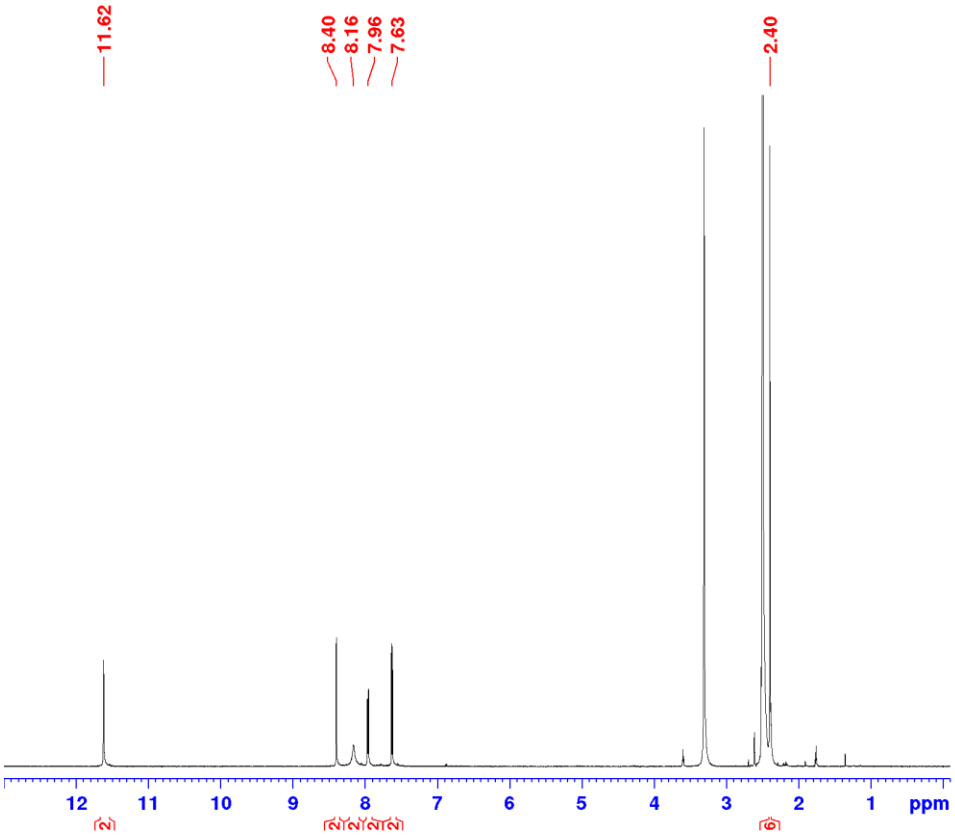
F2 - Acquisition Parameters
Date_           20170206
Time              20.47
INSTRUM          isa400
PROBHD   5 mm PABBO BB-
PULPROG          zgpg30
TD                65536
SOLVENT            DMSO
NS                  128
DS                    4
SWH           24038.461 Hz
FIDRES        0.366798 Hz
AQ            1.3631488 sec
RG                 2050
DW               20.800 usec
DE               10.00 usec
TE                300.0 K
D1            2.00000000 sec
D11           0.03000000 sec
TD0                   1

======== CHANNEL f1 ========
SFO1         100.6228298 MHz
NUC1                 13C
P1                9.25 usec
PLW1         56.00000000 W

======== CHANNEL f2 ========
SFO2         400.1316005 MHz
NUC2                 1H
CPDPRG[2         waltz16
PCPD2             90.00 usec
PLW2         23.00000000 W
PLW12         0.44367000 W
PLW13         0.22316000 W

F2 - Processing parameters
SI                32768
SF           100.6128192 MHz
WDW                  EM
SSB       0
LB                1.00 Hz
GB        0
PC                1.40
```

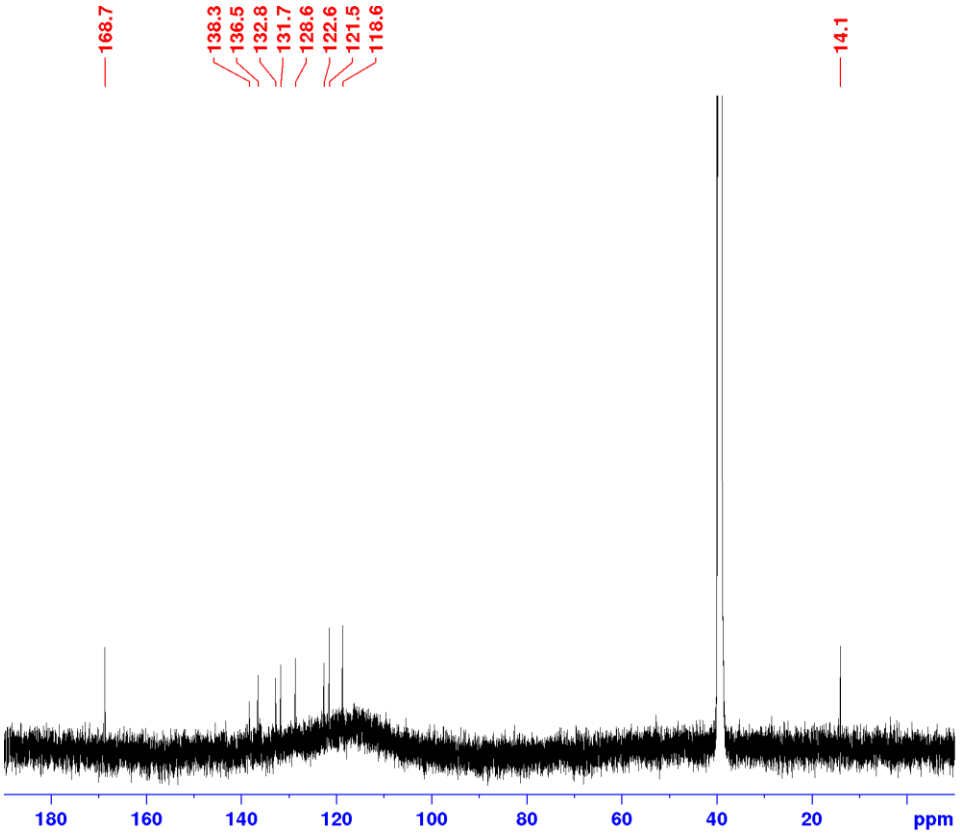
^{1}H- and ^{13}C-NMR spectra of **2** in DMSO-d_6




```
Current Data Parameters
NAME      170213-dasz-NB117
EXPNO                    1
PROCNO                   1

F2 - Acquisition Parameters
Date_             20170213
Time                15.58 h
INSTRUM             spect
PROBHD    Z865401_0004 (
PULPROG               zg30
TD                   65536
SOLVENT               DMSO
NS                      16
DS                       2
SWH             12019.230 Hz
FIDRES           0.366798 Hz
AQ               2.7262976 sec
RG                     287
DW                41.600 usec
DE                11.00 usec
TE                 300.0 K
D1              1.00000000 sec
TD0                      1
SFO1           600.1737060 MHz
NUC1                   1H
P1                  10.31 usec
PLW1           10.00000000 W

F2 - Processing parameters
SI                   65536
SF             600.1700047 MHz
WDW                    EM
SSB       0
LB                0.30 Hz
GB        0
PC                1.00
```

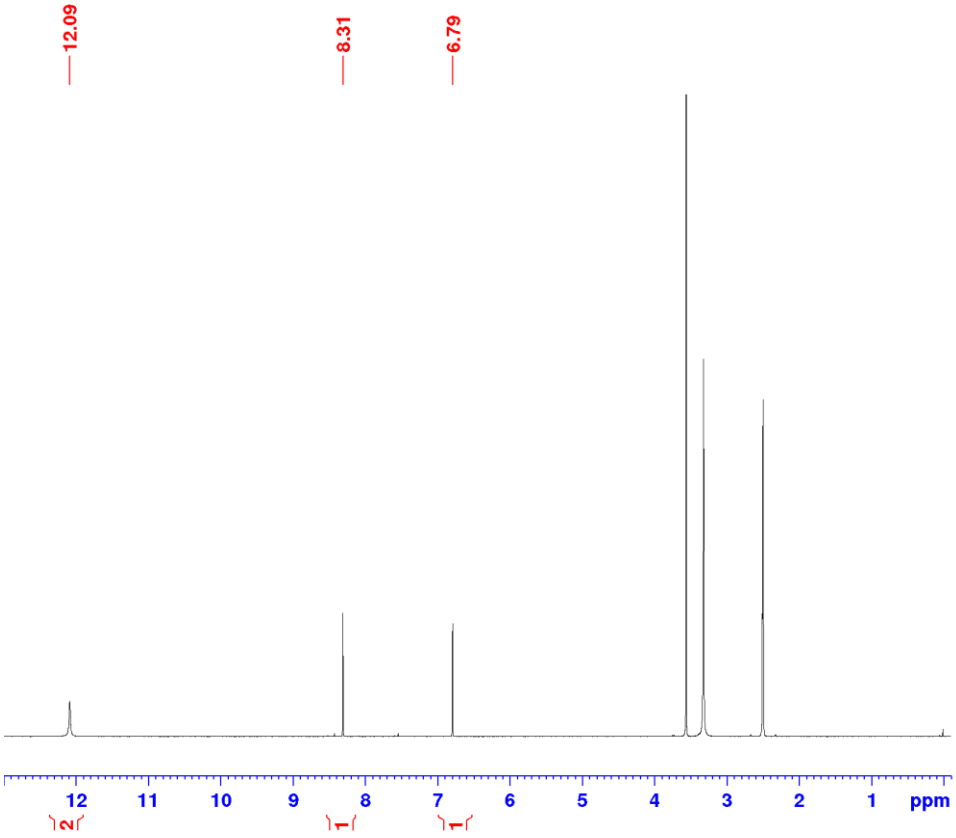



```
Current Data Parameters
NAME      170213-dasz-NB117
EXPNO                    4
PROCNO                   1

F2 - Acquisition Parameters
Date_             20170214
Time                8.21 h
INSTRUM             spect
PROBHD    Z865401_0004 (
PULPROG            zgpg30
TD                   65536
SOLVENT               DMSO
NS                   15416
DS                       4
SWH             36057.691 Hz
FIDRES           1.100393 Hz
AQ               0.9087659 sec
RG                    2050
DW                13.867 usec
DE                11.00 usec
TE                 300.0 K
D1              2.00000000 sec
D11             0.03000000 sec
TD0                      1
SFO1           150.9279571 MHz
NUC1                   13C
P1                  14.50 usec
PLW1           132.00000000 W
SFO2           600.1724007 MHz
NUC2                   1H
CPDPRG[2          waltz16
PCPD2               70.00 usec
PLW2           10.00000000 W
PLW12           0.21693000 W
PLW13           0.10911000 W

F2 - Processing parameters
SI                   32768
SF             150.9129419 MHz
WDW                    EM
SSB       0
LB                1.00 Hz
GB        0
PC                1.40
```

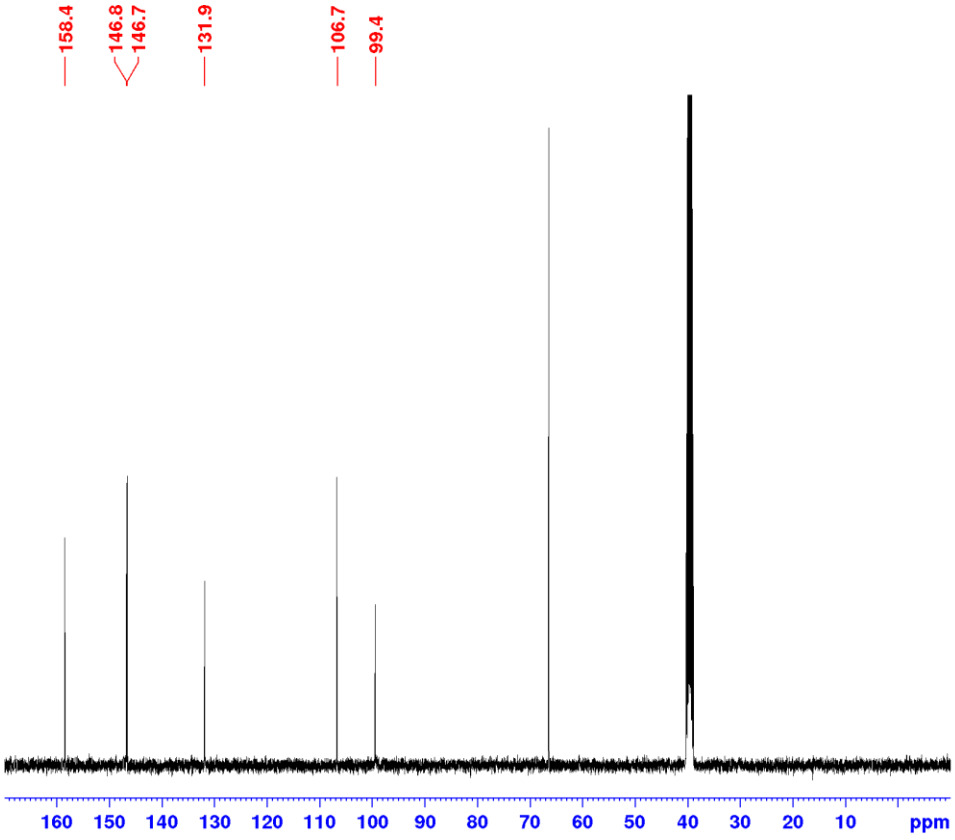
^{1}H- and ^{13}C-NMR spectra of **7a** in DMSO-d_6




```
Current Data Parameters
NAME       190726dasz-1
EXPNO                 1
PROCNO                1

F2 - Acquisition Parameters
Date_           20190726
Time              14.23 h
INSTRUM           isa400
PROBHD   Z863001_0018 (
PULPROG            zg30
TD                65536
SOLVENT            DMSO
NS                    8
DS                    2
SWH            8223.685 Hz
FIDRES        0.250967 Hz
AQ            3.9845889 sec
RG                  456
DW               60.800 usec
DE               16.26 usec
TE                300.0 K
D1           1.00000000 sec
TD0                   1
SFO1         400.1324710 MHz
NUC1                 1H
P1                12.90 usec
PLW1        16.00000000 W

F2 - Processing parameters
SI                65536
SF          400.1300031 MHz
WDW                  EM
SSB       0
LB                0.30 Hz
GB        0
PC                1.00
```

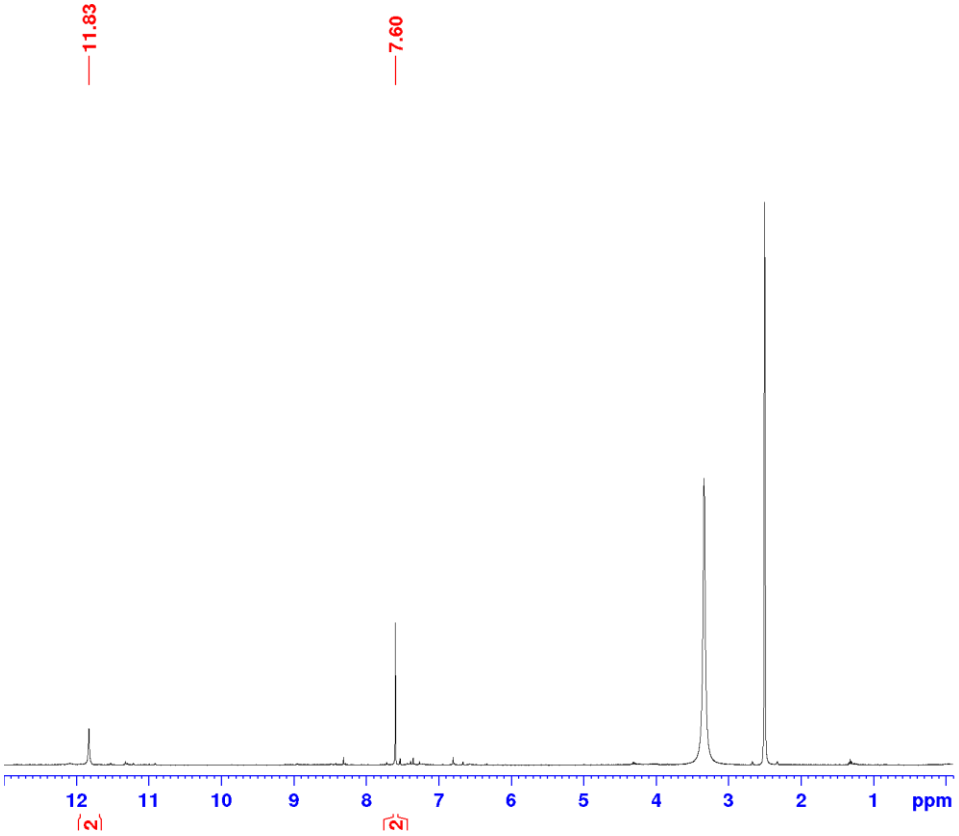



```
Current Data Parameters
NAME       190726dasz-1
EXPNO                 5
PROCNO                1

F2 - Acquisition Parameters
Date_           20190727
Time              3.19 h
INSTRUM           isa400
PROBHD   Z863001_0018 (
PULPROG          zgpg30
TD                65536
SOLVENT            DMSO
NS                 1024
DS                    4
SWH           24038.461 Hz
FIDRES        0.733596 Hz
AQ            1.3631488 sec
RG                 2050
DW               20.800 usec
DE               12.38 usec
TE                300.0 K
D1           2.00000000 sec
D11          0.03000000 sec
TD0                   1
SFO1         100.6228298 MHz
NUC1                 13C
P1                9.00 usec
PLW1        56.00000000 W
SFO2         400.1316005 MHz
NUC2                 1H
CPDPRG[2         waltz16
PCPD2             90.00 usec
PLW2        16.00000000 W
PLW12        0.32870999 W
PLW13        0.16534001 W

F2 - Processing parameters
SI                32768
SF          100.6128162 MHz
WDW                  EM
SSB       0
LB                1.00 Hz
GB        0
PC                1.40
```

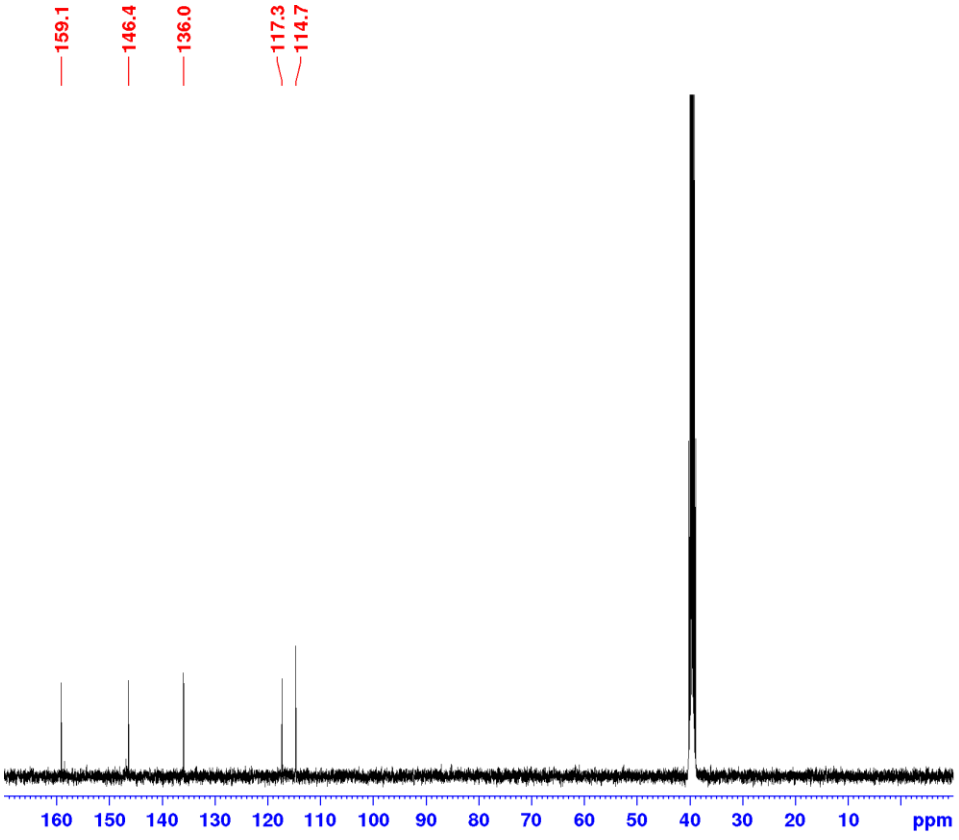
^{1}H- and ^{13}C-NMR spectra of **7b** in DMSO-d_6




```
Current Data Parameters
NAME        190726dasz-2
EXPNO                  1
PROCNO                 1

F2 - Acquisition Parameters
Date_            20190726
Time               14.28 h
INSTRUM           isa400
PROBHD    Z863001_0018 (
PULPROG             zg30
TD                 65536
SOLVENT             DMSO
NS                     8
DS                     2
SWH             8223.685 Hz
FIDRES          0.250967 Hz
AQ              3.9845889 sec
RG                   512
DW                60.800 usec
DE                16.26 usec
TE                300.0 K
D1             1.00000000 sec
TD0                    1
SFO1          400.1324710 MHz
NUC1                  1H
P1                 12.90 usec
PLW1          16.00000000 W

F2 - Processing parameters
SI                 65536
SF            400.1300033 MHz
WDW                   EM
SSB       0
LB                 0.30 Hz
GB        0
PC                 1.00
```

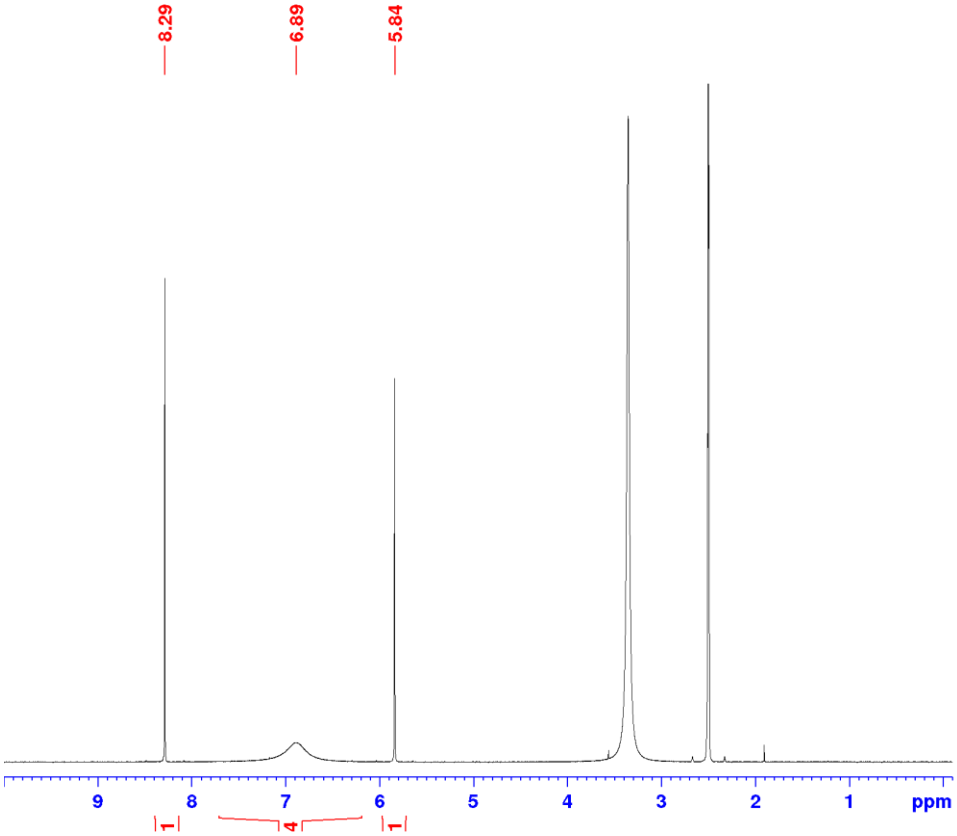



```
Current Data Parameters
NAME        190726dasz-2
EXPNO                  5
PROCNO                 1

F2 - Acquisition Parameters
Date_            20190727
Time                5.02 h
INSTRUM           isa400
PROBHD    Z863001_0018 (
PULPROG           zgpg30
TD                 65536
SOLVENT             DMSO
NS                  1024
DS                     4
SWH            24038.461 Hz
FIDRES          0.733596 Hz
AQ              1.3631488 sec
RG                  2050
DW                20.800 usec
DE                12.38 usec
TE                300.0 K
D1             2.00000000 sec
D11            0.03000000 sec
TD0                    1
SFO1          100.6228298 MHz
NUC1                  13C
P1                 9.00 usec
PLW1          56.00000000 W
SFO2          400.1316005 MHz
NUC2                  1H
CPDPRG[2         waltz16
PCPD2             90.00 usec
PLW2          16.00000000 W
PLW12          0.32870999 W
PLW13          0.16534001 W

F2 - Processing parameters
SI                 32768
SF            100.6128114 MHz
WDW                   EM
SSB       0
LB                 1.00 Hz
GB        0
PC                 1.40
```

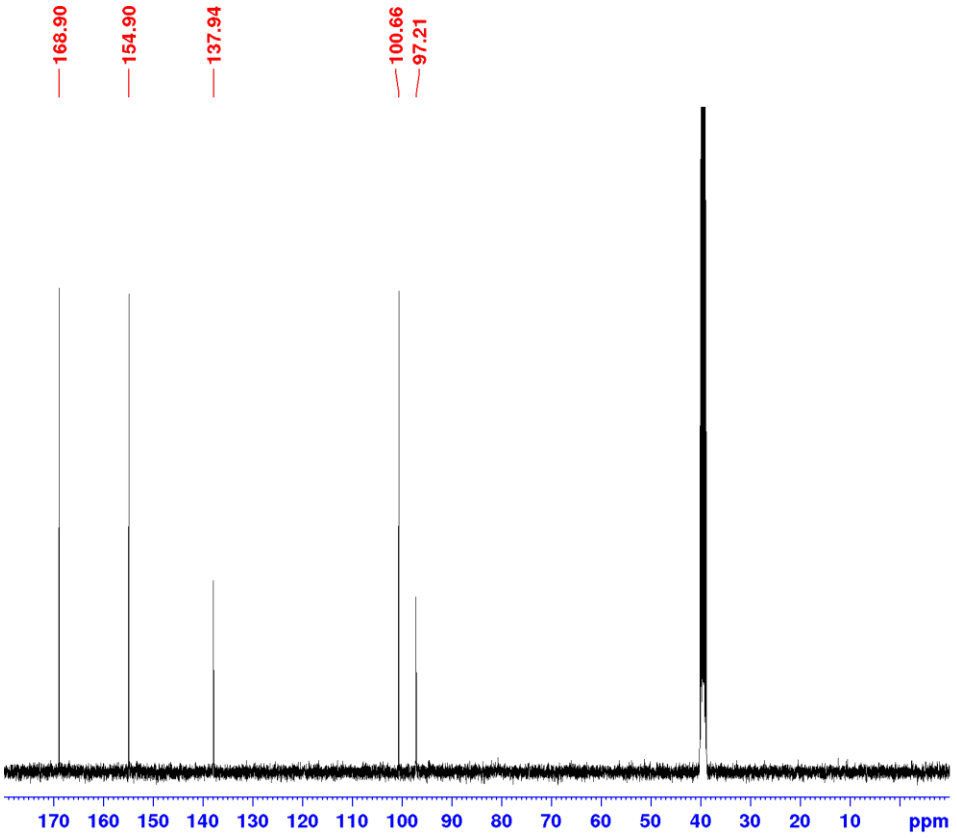
^{1}H- and ^{13}C-NMR spectra of **8** in DMSO-d_6




```
Current Data Parameters
NAME      190802dasz_FastTrack-31
EXPNO                1
PROCNO               1

F2 - Acquisition Parameters
Date_             20190802
Time                11.51 h
INSTRUM           isa400
PROBHD    Z863001_0018 (
PULPROG             zg30
TD                 65536
SOLVENT             DMSO
NS                     8
DS                     2
SWH             8223.685 Hz
FIDRES          0.250967 Hz
AQ              3.9845889 sec
RG                   322
DW                60.800 usec
DE                16.26 usec
TE                300.0 K
D1             1.00000000 sec
TD0                    1
SFO1          400.1324710 MHz
NUC1                  1H
P1                 12.90 usec
PLW1          16.00000000 W

F2 - Processing parameters
SI                 65536
SF            400.1300037 MHz
WDW                   EM
SSB       0
LB                 0.30 Hz
GB        0
PC                 1.00
```

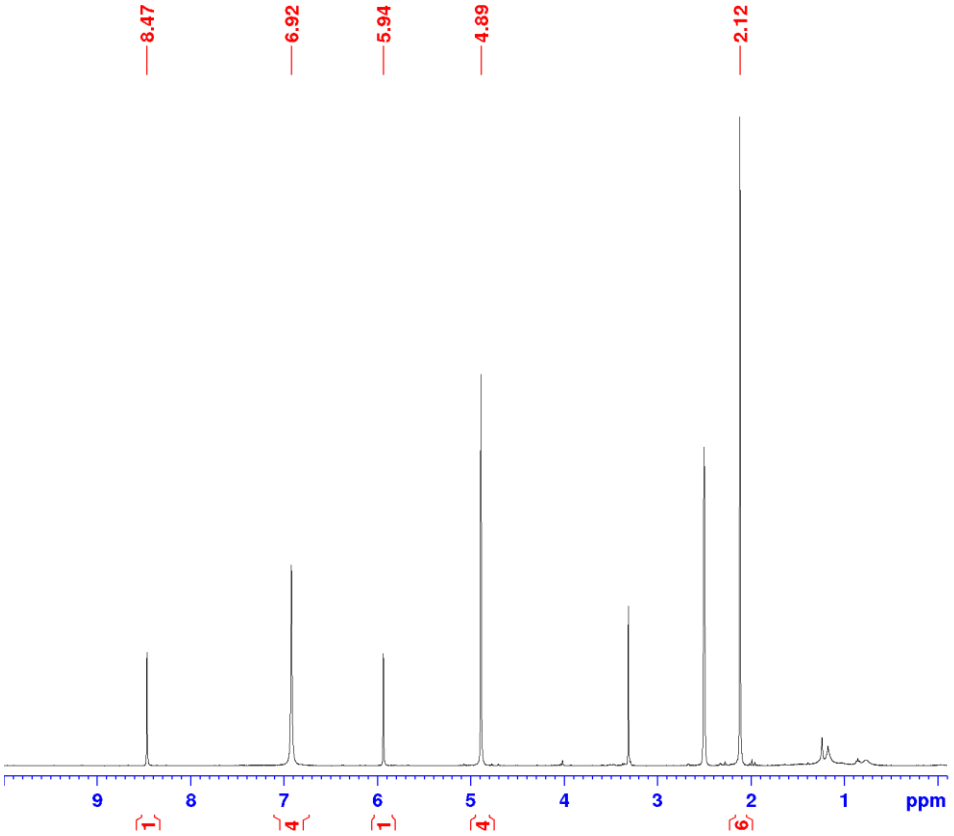



```
Current Data Parameters
NAME       190802dasz-31
EXPNO                1
PROCNO               1

F2 - Acquisition Parameters
Date_             20190803
Time                6.18 h
INSTRUM           isa400
PROBHD    Z863001_0018 (
PULPROG           zgpg30
TD                 65536
SOLVENT             DMSO
NS                  1024
DS                     4
SWH            24038.461 Hz
FIDRES          0.733596 Hz
AQ              1.3631488 sec
RG                  2050
DW                20.800 usec
DE                12.38 usec
TE                300.0 K
D1             2.00000000 sec
D11            0.03000000 sec
TD0                    1
SFO1          100.6228298 MHz
NUC1                  13C
P1                 9.00 usec
PLW1          56.00000000 W
SFO2          400.1316005 MHz
NUC2                  1H
CPDPRG[2          waltz16
PCPD2             90.00 usec
PLW2          16.00000000 W
PLW12          0.32870999 W
PLW13          0.16534001 W

F2 - Processing parameters
SI                 32768
SF            100.6128135 MHz
WDW                   EM
SSB       0
LB                 1.00 Hz
GB        0
PC                 1.40
```

^{1}H- and ^{13}C-NMR spectra of **9** in DMSO-d_6

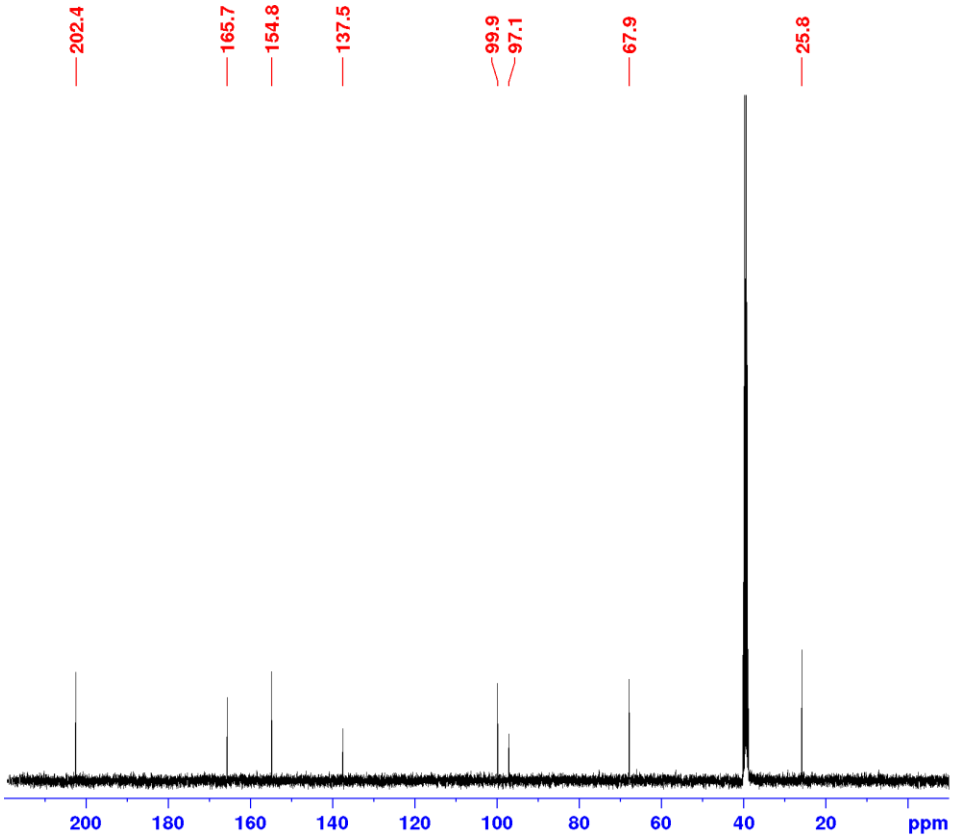



```
Current Data Parameters
NAME       170123badu-60
EXPNO                  1
PROCNO                 1

F2 - Acquisition Parameters
Date_           20170123
Time              14.42
INSTRUM           isa400
PROBHD   5 mm PABBO BB-
PULPROG             zg30
TD                 65536
SOLVENT             DMSO
NS                     8
DS                     2
SWH             8223.685 Hz
FIDRES          0.125483 Hz
AQ              3.9845889 sec
RG                   406
DW                60.800 usec
DE                10.00 usec
TE                300.1 K
D1             1.00000000 sec
TD0                    1

======== CHANNEL f1 ========
SFO1          400.1324710 MHz
NUC1                  1H
P1                 12.50 usec
PLW1         23.01399994 W

F2 - Processing parameters
SI                 65536
SF            400.1300033 MHz
WDW                   EM
SSB       0
LB                 0.30 Hz
GB        0
PC                 1.00
```




```
Current Data Parameters
NAME       170123badu-60
EXPNO                  2
PROCNO                 1

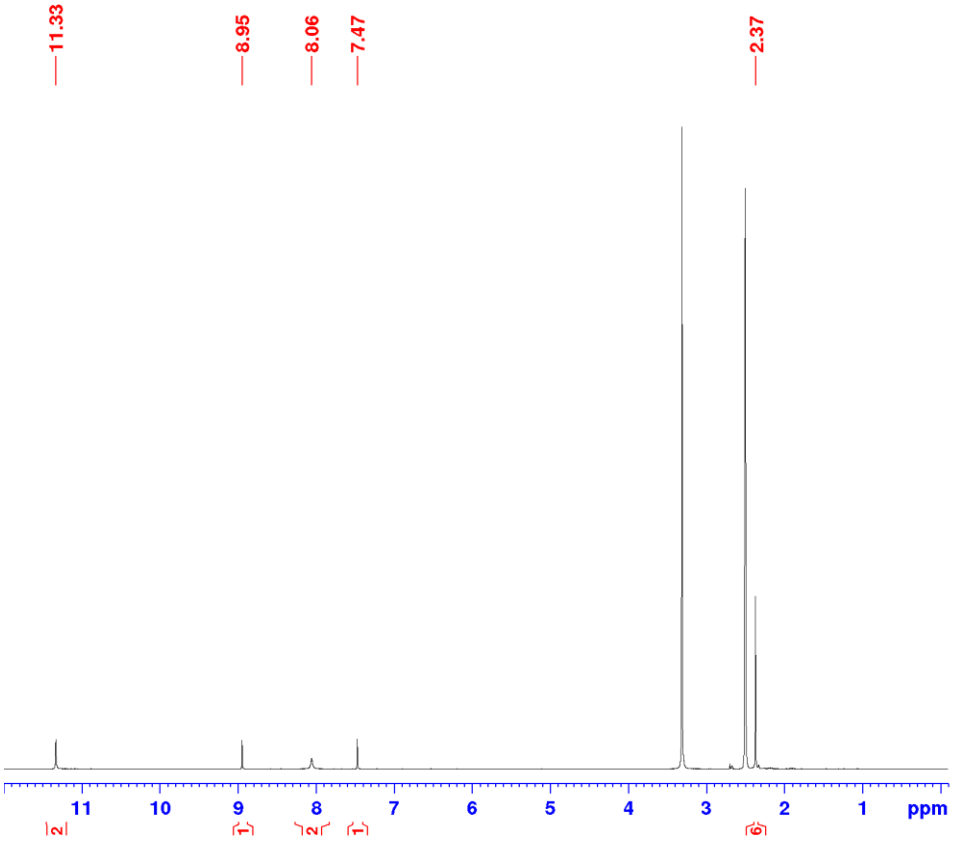
F2 - Acquisition Parameters
Date_           20170123
Time              14.51
INSTRUM           isa400
PROBHD   5 mm PABBO BB-
PULPROG           zgpg30
TD                 65536
SOLVENT             DMSO
NS                   128
DS                     4
SWH            24038.461 Hz
FIDRES          0.366798 Hz
AQ              1.3631488 sec
RG                  2050
DW                20.800 usec
DE                10.00 usec
TE                300.0 K
D1             2.00000000 sec
D11            0.03000000 sec
TD0                    1

======== CHANNEL f1 ========
SFO1          100.6228298 MHz
NUC1                  13C
P1                 9.25 usec
PLW1         56.00000000 W

======== CHANNEL f2 ========
SFO2          400.1316005 MHz
NUC2                  1H
CPDPRG[2          waltz16
PCPD2              90.00 usec
PLW2         23.00000000 W
PLW12         0.44367000 W
PLW13         0.22316000 W

F2 - Processing parameters
SI                 32768
SF            100.6128185 MHz
WDW                   EM
SSB       0
LB                 1.00 Hz
GB        0
PC                 1.40
```

^{1}H- and ^{13}C-NMR spectra of **3** in DMSO-d_6

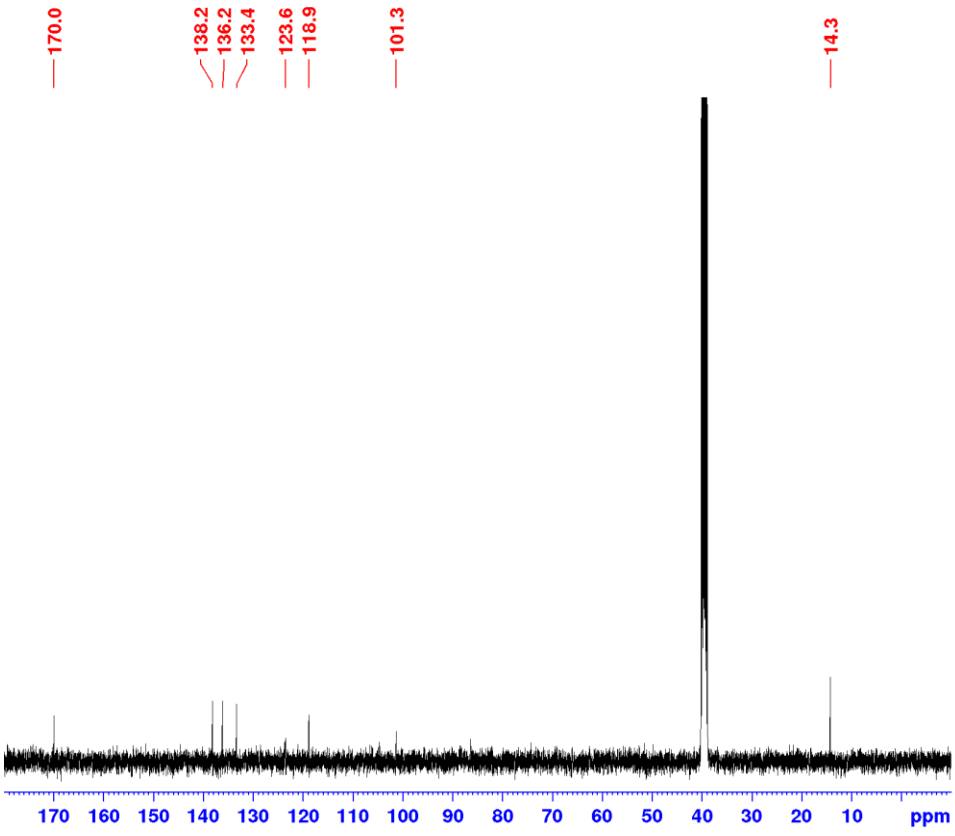



```
Current Data Parameters
NAME      170129dasz-27
EXPNO                1
PROCNO               1

F2 - Acquisition Parameters
Date_           20170129
Time              16.23
INSTRUM           isa400
PROBHD   5 mm PABBO BB-
PULPROG             zg30
TD                 65536
SOLVENT             DMSO
NS                    32
DS                     2
SWH             8223.685 Hz
FIDRES          0.125483 Hz
AQ              3.9845889 sec
RG                   456
DW                60.800 usec
DE                10.00 usec
TE                300.0 K
D1            1.00000000 sec
TD0                  1

======== CHANNEL f1 ========
SFO1         400.1324710 MHz
NUC1                 1H
P1                12.50 usec
PLW1         23.01399994 W

F2 - Processing parameters
SI                 65536
SF           400.1300034 MHz
WDW                  EM
SSB       0
LB                0.30 Hz
GB        0
PC                1.00
```




```
Current Data Parameters
NAME      170129dasz-27
EXPNO                7
PROCNO               1

F2 - Acquisition Parameters
Date_           20170129
Time              22.19
INSTRUM           isa400
PROBHD   5 mm PABBO BB-
PULPROG           zgpg30
TD                 65536
SOLVENT             DMSO
NS                  1024
DS                     4
SWH            24038.461 Hz
FIDRES          0.366798 Hz
AQ              1.3631488 sec
RG                  2050
DW                20.800 usec
DE                10.00 usec
TE                300.0 K
D1            2.00000000 sec
D11           0.03000000 sec
TD0                  1

======== CHANNEL f1 ========
SFO1         100.6228298 MHz
NUC1                 13C
P1                9.25 usec
PLW1         56.00000000 W

======== CHANNEL f2 ========
SFO2         400.1316005 MHz
NUC2                 1H
CPDPRG[2         waltz16
PCPD2             90.00 usec
PLW2         23.00000000 W
PLW12         0.44367000 W
PLW13         0.22316000 W

F2 - Processing parameters
SI                 32768
SF           100.6128187 MHz
WDW                  EM
SSB       0
LB                1.00 Hz
GB        0
PC                1.40
```