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I. General Experimental Protocols

^{13}C and ^{1}H NMR spectra were recorded on a Bruker HD-500, AV-500, AV-400, or AX-400 spectrometer. Proton chemical shifts are referenced to TMS (δ 0.00 ppm) in $CDCl_3$ solutions and to the residual CHD_5 (δ 7.16 ppm) in benzene-d_6 solutions. A non-first order multiplet, doublet, or doublet of doublets in a ^{1}H NMR spectrum is denoted as a 'nfom', 'nfod', or 'nfodd', respectively. For the latter two, the coupling constant is listed as an apparent value (J_{app}), because the spacing between the two major lines for the population of molecules having magnetically equivalent protons (ca. 50%) is actually the value of $J_o + J_p$). Resonances are reported in the following format: chemical shift (ppm) [multiplicity, coupling constant (s) (in Hz), integral value (to the nearest integer), and assignment of the substructural environment within the structure]. First-order coupling constants were analyzed using methods we have published elsewhere.[1,2] The ^{13}C NMR chemical shifts are taken from the "1D" spectrum where possible, although some were deduced from HMBC correlations. Carbon chemical shifts are referenced to δ 77.16 ppm in $CDCl_3$ solutions and to δ 128.06 for C_6D_6 solutions.

Infrared spectra were recorded using a Bruker Alpha II Spectrometer. Samples were prepared as thin films on a diamond window in the attenuated total reflectance (ATR) mode. Absorption maxima are given in cm^{-1}.

The high-resolution **mass spectrometry** (HRMS) measurements were made in the ESI mode using a Thermo Orbitrap Velos instrument (mass accuracy of ≤3 ppm). An external calibrant was used (Pierce™ LTQ) and the samples were directly injected into the ion source.

Medium pressure liquid **chromatography** (MPLC) was often used to purify newly synthesized materials. Hand-packed silica gel columns (normal-phase, 25-200 psi, 20-40 μm, 60 Å pore size, Teledyne RediSep Rf Gold®) were used. The apparatus consisted of a Waters HPLC pump (model 510), a Gilson (111 UV) detector, and a Waters (R401) differential refractive index detector. Preparative flash chromatography was performed on columns packed with Agela silica gel (230-400 mesh). Thin layer chromatography (TLC) was performed on silica-gel coated, plastic-backed plates that were visualized by UV light and/or by a solution of potassium permanganate and heating.

The indicated reaction temperature refers to the temperature of the external cooling or heating bath. HDDA reactions, including those performed at temperatures higher than the boiling point of the reaction solvent, were done in a screw-top culture tube that was capped with an inert, Teflon®-lined closure.

Poly-yne substrates **8**[3], **19a**[4], and **19b**[4] were synthesized according to reported methods.

II. Preparation procedures and characterization data for all compounds

A. General Procedure for trapping of HDDA-generated arynes with imines

The polyyne precursor (1 equiv) and the imine (1-3 equiv) were combined in a screw-capped culture tube. 1,2-Dichloroethane was added (0.05 M) and the resulting solution was placed in an oil bath maintained at 90 °C and allowed to react overnight. Subsequently, the solvent was removed under reduced pressure, and the crude material was purified using MPLC with the elution solvent mixture indicated for each compound.

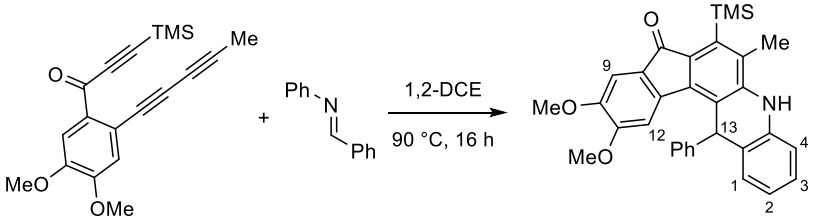
B. General Procedure for oxidation of 1,4-dihydroacridines to their respective acridines

A scintillation vial or a culture tube was charged with a stir bar and the respective 1,4-dihydroacridine (1 equiv). Chloroform or dichloromethane (0.005 M) was added, along with MnO_2 (ca. 10-20 equiv). The resulting slurry was allowed to stir at ambient temperature until the reaction was observed to be complete by TLC analysis. The reaction mixture was filtered through Celite® and the filtrate was concentrated in vacuo.

C. General Procedure for one-pot synthesis of acridines from HDDA-generated benzynes and imines

The HDDA polyyne precursor (1 equiv) and the imine (1-3 equiv) were added to a screw-cap culture tube. 1,2-Dichloroethane was added (0.05 M), and the resulting solution was placed in an oil bath maintained at 90 °C and allowed to react overnight. MnO_2 (ca. 10-20 equiv) and a stir bar were added. The slurry was then stirred at ambient temperature until the reaction was observed to be complete by TLC. The reaction mixture was filtered through Celite® and the filtrate was concentrated in vacuo.

10,11-Dimethoxy-6-methyl-13-phenyl-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (11)



8 ***2a*** ***11***, 98%

N-Benzylideneaniline (**2a**) was prepared according to a reported procedure.[5]

Following general procedure A, 1-(4,5-dimethoxy-2-(penta-1,3-diyn-1-yl)phenyl)-3-(trimethylsilyl)prop-2-yn-1-one (**8**, 0.024 g, 0.074 mmol, 1 equiv), (*E*)-*N*,1-diphenylmethanimine (**2a**, 0.015 g, 0.083 mmol, 1.1 equiv), and dichloroethane (2 mL) were used to prepare the 1,4-dihydroacridine **11**. Purification of the crude product by MPLC (2:1 hexanes:EtOAc) yielded **11** (0.038 g, 0.076 mmol, 98%) as an orange crystalline solid.

^{1}H NMR (500 MHz, $CDCl_3$): δ 7.45 (d, J = 7.8, 1.4 Hz, 1H, *H1*), 7.29 (nfod, J_{app} = 7.5 Hz, 2H, ArH_o), 7.22 (nfodd, J_{app} = 7.7, 7.7 Hz, 2H, PhH_m), 7.126 (ddd, J = 7.4, 7.4, 1.5 Hz, 1H, *H3*), 7.125 (tt, J = 7.4, 1.5 Hz, 1H, ArH_p), 7.10 (s, 1H, *H9*), 7.09 (s, 1H, *H12*), 6.95 (dd, J = 7.4, 7.4, 1.1 Hz, 1H, *H2*), 6.81 (dd, J = 7.9, 1.3 Hz, 1H, *H4*), 6.48 (s, 1H, N*H*), 5.77 (s, 1H, *H13*), 3.90 (s, 3H, OCH_3), 3.86 (s, 3H, OCH_3), 2.45 (s, 3H, ArCH_3), and 0.46 (s, 9H, Si(CH_3)$_3$).

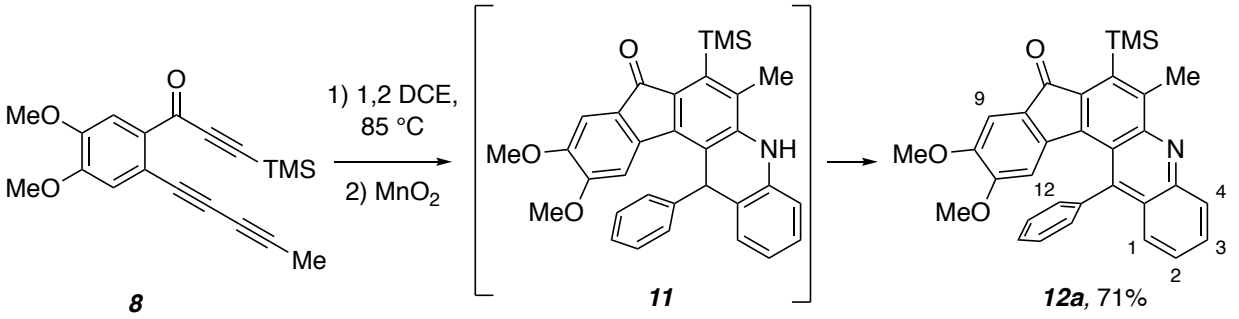
^{13}C NMR (126 MHz, $CDCl_3$): 193.6, 153.0, 149.0, 144.7, 143.1, 142.6, 141.2, 137.6, 137.5, 133.8, 129.1, 128.70, 128.67, 127.8, 127.1, 127.0, 125.3, 123.9, 122.3, 118.8, 115.2, 107.8, 106.7, 56.6, 56.2, 44.7, 18.2, and 3.2.

HRMS (ESI-TOF): Calcd for $C_{32}H_{32}NO_3Si^+$ $[M+H^+]^+$ requires 506.2146; found 506.2150.

IR (neat): 3440, 3390, 3059, 3001, 2924, 2853, 2359, 2342, 2069, 2034, 1976, 1961, 1944, 1694, 1605, 1577, 1546, 1489, 1465, 1432, 1416, 1375, 1343, 1324, 1299, 1283, 1259, 1245, 1215, 1173, 1158, 1091, 1045, 1027, 991, 936, 873, 854, 797, 771, 751, 727, 699, 676, 645, 630, 613, 599, 575, 519, 493, 449, and 408 cm^{-1}.

mp: 249-250 °C.

10,11-Dimethoxy-6-methyl-13-phenyl-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12a)



8 ***11*** ***12a***, 71%

Following general procedure C, 1-(4,5-dimethoxy-2-(penta-1,3-diyn-1-yl)phenyl)-3-(trimethylsilyl)prop-2-yn-1-one (**8**, 0.010 g, 0.030 mmol, 1 equiv), (*E*)-*N*,1-diphenylmethanimine (**2a**, 0.007 g, 0.038 mmol, 1.3 equiv), MnO_2 (xs), and dichloroethane (2 mL) were used to prepare acridine **12a**. Purification of the crude product yielded acridine **12a** (0.011 g, 0.022 mmol, 71%) as a purple crystalline solid.

^{1}H NMR (500 MHz, $CDCl_3$): δ 8.26 (d, *J* = 8.6 Hz, 1H, *H4*), 8.14 (d, *J* = 8.9 Hz, 1H, *H1*), 7.78 (nfod, J_{app} = 7.2 Hz, 2H, PhH_o), 7.77 (br dd, *J* = 8.8, 6.8 Hz, 1H, *H3*), 7.56 (nfodd, J_{app} = 7.5, 7.5 Hz, 2H, PhH_m), 7.53 (tt, *J* = 6.9, 1.7 Hz, 1H, PhH_p), 7.48 (br dd, *J* = 8.6, 6.6 Hz, 1H, *H2*), 7.04 (s, 1H, *H9*), 5.64 (s, 1H, *H12*), 3.83 (s, 3H, C10OCH_3), 3.51 (s, 3H, C11OCH_3), 3.06 (s, 1H, ArCH_3), and 0.51 (s, 9H, Si(CH_3)$_3$).

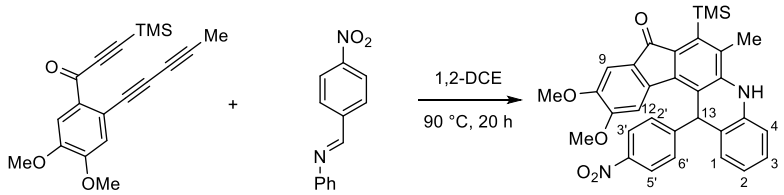
^{13}C NMR (126 MHz, $CDCl_3$): 195.4, 153.0, 150.6, 149.0, 148.0, 146.9, 145.1, 143.0, 140.1, 138.6, 137.3, 136.1, 132.8, 130.9, 130.3, 129.5, 129.0, 126.7, 126.5, 125.2, 125.0, 120.8, 109.3, 106.5, 56.7, 56.1, 20.7, and 2.7.

HRMS (ESI-TOF): Calcd for $C_{32}H_{30}NO_3Si^+$ $[M+H^+]^+$ requires 504.1989; found 504.1983.

IR (neat): 3062, 2956, 2922, 2852, 2357, 2171, 2099, 2041, 2023, 1984, 1758, 1709, 1595, 1579, 1498, 1463, 1442, 1418, 1405, 1376, 1336, 1285, 1250, 1224, 1180, 1144, 1109, 1059, 1026, 1012, 972, 896, 842, 816, 799, 764, 734, 702, 671, 642, 619, 604, 568, 544, 519, 507, and 412 cm^{-1}.

mp: 96-97 °C.

10,11-Dimethoxy-6-methyl-13-(4-nitrophenyl)-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (SI-1):



8 *2b* ***SI-1*** , 68%

1-(4-Nitrophenyl)-*N*-phenylmethanimine (**2b**) was prepared according to a reported procedure.[6]

Following general procedure A, 1-(4,5-dimethoxy-2-(penta-1,3-diyn-1-yl)phenyl)-3-(trimethylsilyl)prop-2-yn-1-one (**8**, 0.009 g, 0.029 mmol, 1 equiv), (*E*)-1-(4-nitrophenyl)-N-phenylmethanimine (**2b**, 0.023 g, 0.093 mmol, 3.2 equiv), and dichloroethane (2 mL) were used to prepare the 1,4-dihydroacridine **SI-1**. Purification of the crude product by MPLC (2:1 hexanes:EtOAc) yielded **SI-1** (0.011 g, 0.020 mmol, 68%) as an orange amorphous solid.

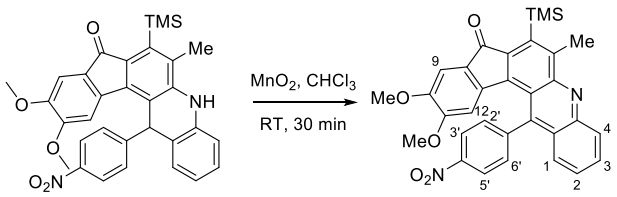
^{1}H NMR (500 MHz, $CDCl_3$): δ 8.09 (nfod, J_{app} = 8.6 Hz, 2H, *H3'* and *H5'*), 7.43 (nfod, J_{app} = 9.0 Hz, 2H, *H2'* and *H6'*), 7.43 (dd, *J* = 7.5, 1.5 Hz, 1H, *H1*), 7.18 (ddd, *J* = 7.6, 7.6, 1.3 Hz, 1H, *H3*), 7.12 (s, 1H, *H9*), 7.02 (s, 1H, *H12*), 6.99 (ddd, *J* = 7.5, 7.5, 1.8 Hz, 1H, *H2*), 6.86 (dd, *J* = 7.6, 1.8 Hz, 1H, *H4*), 6.53 (s, 1H, N*H*), 5.90 (s, 1H, *H13*), 3.92 (s, 3H, OCH_3), 3.88 (s, 3H, OCH_3), 2.46 (s, 3H, ArCH_3), and 0.46 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (100 MHz, $CDCl_3$): 193.2, 153.1, 151.4, 149.4, 146.9, 142.7, 142.4, 142.1, 137.8, 137.0, 134.0, 128.8, 128.6, 128.5, 127.9, 125.8, 124.4, 122.6, 122.0, 117.3, 115.5, 107.3, 107.0, 56.6, 56.3, 44.6, 18.2, and 3.2.

HRMS (ESI-TOF): Calcd for $C_{32}H_{31}N_2O_5Si^+$ $[M+H^+]^+$ requires 551.1997; found 551.1958 (major ion) and Calcd for $C_{32}H_{29}N_2O_5Si^+$ $[M-2H+H^+]^+$ requires 549.1840; found 549.1817.

IR (neat): 3433, 3103, 3057, 3002, 2943, 2899, 2835, 2452, 2346, 2220, 2122, 1925, 1696, 1604, 1593, 1577, 1546, 1521, 1489, 1465, 1431, 1415, 1374, 1344, 1299, 1282, 1259, 1244, 1214, 1110, 1090, 1044, 1022, 1014, 990, 937, 852, 797, 770, 751, 711, 696, 677, 642, 628, 603, 576, 544, 519, 494, 457, and 433 cm^{-1}.

10,11-Dimethoxy-6-methyl-13-(4-nitrophenyl)-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12b):



Following General Procedure B, 10,11-dimethoxy-6-methyl-13-(4-nitrophenyl)-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (**SI-1**, 0.009 g, 0.015 mmol, 1 equiv), MnO_2 (xs), and $CHCl_3$ (10 mL) were used to prepare acridine **12b**. Purification of the crude product yielded **12b** (0.008 g, 0.014 mmol, 90%) as a purple amorphous solid.

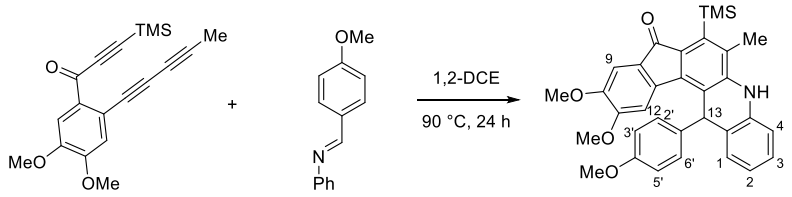
^{1}H NMR (500 MHz, $CDCl_3$): δ 8.42 (nfod, J_{app} = 8.7 Hz, 2H, *H3'* and *H5'*), 8.30 (dd, *J* = 8.3, 1.1 Hz, 1H, *H4*), 7.99 (nfod, J_{app} = 8.7 Hz, 2H, *H2'* and *H6'*), 7.98 (dd, *J* = 8.3, 1.1 Hz, 1H, *H1*), 7.80 (ddd, *J* = 8.8, 6.5, 1.3 Hz, 1H, *H3*), 7.53 (ddd, *J* = 8.8, 6.5, 1.3 Hz, 1H, *H2*), 7.06 (s, 1H, *H9*), 5.60 (s, 1H, *H12*), 3.83 (s, 3H, C10-OCH_3), 3.50 (s, 3H, C11-OCH_3), 3.07 (s, 3H, ArCH_3), and 0.51 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (125 MHz, $CDCl_3$): 194.9, 152.9, 150.3, 148.9, 148.6, 148.1, 147.4, 145.0, 141.8, 141.7, 139.3, 137.9, 136.6, 133.6, 131.3, 130.5, 127.7, 125.4, 125.2, 124.3, 124.0, 120.1, 108.9, 107.1, 56.6, 56.2, 20.6, and 2.6.

HRMS (ESI-TOF): Calcd for $C_{32}H_{29}N_2O_5Si^+$ $[M+H^+]^+$ requires 549.1840; found 549.1827.

IR (neat): 3104, 3074, 2999, 2945, 2900, 2838, 2702, 2456, 2036, 2007, 1974, 1950, 1707, 1598, 1586, 1524, 1493, 1460, 1406, 1379, 1367, 1346, 1295, 1248, 1217, 1182, 1165, 1133, 1099, 1060, 1014, 981, 944, 884, 854, 812, 793, 762, 735, 704, 680, 662, 644, 627, 619, 606, 580, 569, 530, 493, 485, 451, and 414 cm^{-1}.

10,11-Dimethoxy-13-(4-methoxyphenyl)-6-methyl-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (SI-2)



8 *2c* ***SI-2*** , 78%

1-(4-Methoxyphenyl)-*N*-phenylmethanimine (**2c**) was prepared according to a reported procedure.[7]

Following General Procedure A, 1-(4,5-dimethoxy-2-(penta-1,3-diyn-1-yl)phenyl)-3-(trimethylsilyl)prop-2-yn-1-one (**8**, 0.020 g, 0.063 mmol, 1 equiv), (*E*)-1-(4-methoxyphenyl)-*N*-phenylmethanimine (**2c**, 0.041 g, 0.179 mmol, 2.9 equiv), and dichloroethane (2 mL) were used to prepare the 1,4-dihydroacridine **SI-2**. Purification of the crude product by MPLC (2:1 hexanes:EtOAc) yielded **SI-2** (0.027 g, 0.048 mmol, 78%) as an orange amorphous solid.

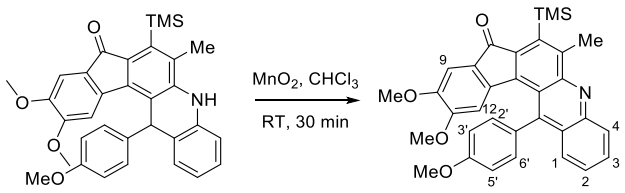
^{1}H NMR (500 MHz, $CDCl_3$): δ 7.41 (dd, *J* = 7.6, 1.4 Hz, 1H, *H1*), 7.18 (nfod, J_{app} = 8.8 Hz, 2H, *H2'* and *H6'*), 7.12 (ddd, *J* = 7.6, 7.6, 1.5 Hz, 1H, *H3*), 7.098 (s, 1H, *H9* or *H12*), 7.096 (s, 1H, *H9* or *H12*), 6.95 (ddd, *J* = 7.5, 7.5, 1.2 Hz, 1H, *H2*), 6.80 (dd, *J* = 7.9, 1.2 Hz, 1H, *H4*), 6.75 (nfod, J_{app} = 8.8 Hz, 2H, *H3'* and *H5'*), 6.47 (s, 1H, N*H*), 5.71 (s, 1H, *H13*), 3.91 (s, 3H, OCH_3), 3.86 (s, 3H, OCH_3), 3.68 (s, 3H, C4'-OCH_3), 2.43 (s, 3H, ArCH_3), and 0.46 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (125 MHz, $CDCl_3$): 193.6, 158.5, 153.0, 149.0, 143.0, 142.5, 141.1, 137.6, 137.5, 136.9, 133.7, 128.7, 128.6, 128.1, 127.7, 125.3, 124.2, 122.2, 119.1, 115.1, 114.4, 107.8, 106.6, 56.5, 56.2, 55.3, 18.2, and 3.2.

HRMS (ESI-TOF): Calcd for $C_{33}H_{32}NO_4Si^+$ $[M-2H+H^+]^+$ requires 534.2095; found 534.2078.

IR (neat): 3445, 3388, 3055, 2999, 2948, 2902, 2835, 2358, 2344, 2197, 2174, 2144, 2121, 2068, 2027, 2014, 1981, 1963, 1950, 1926, 1693, 1607, 1577, 1546, 1507, 1489, 1464, 1431, 1415, 1374, 1342, 1324, 1298, 1283, 1246, 1214, 1179, 1110, 1090, 1030, 990, 874, 846, 797, 788, 770, 748, 699, 676, 646, 631, 607, 588, 557, 529, 487, 474, 453, 435, and 417 cm^{-1}.

10,11-Dimethoxy-13-(4-methoxyphenyl)-6-methyl-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12c)



SI-2 *12c*, 86%

Following General Procedure B, 10,11-dimethoxy-13-(4-methoxyphenyl)-6-methyl-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (**SI-2**, 0.025 g, 0.045 mmol, 1 equiv), MnO_2 (xs), and $CHCl_3$ (10 mL) were used to prepare acridine **12c**. Purification of the crude product yielded **12c** (0.021 g, 0.039 mmol, 86%) as a purple amorphous solid.

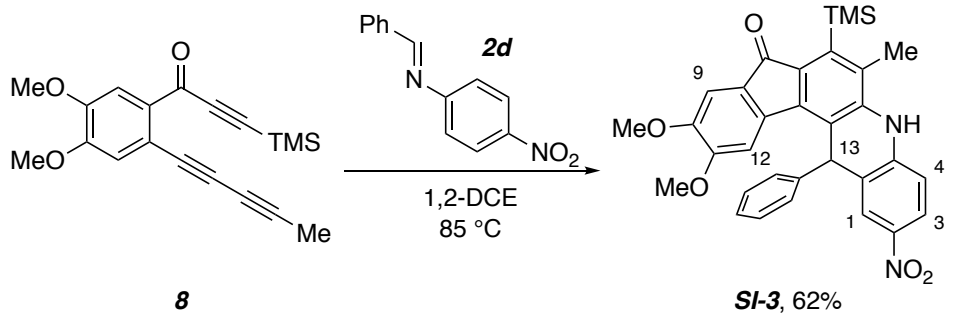
^{1}H NMR (500 MHz, $CDCl_3$): δ 8.25 (dd, *J* = 8.7, 1.2 Hz, 1H, *H4*), 8.16 (dd, *J* = 8.9, 1.2 Hz, 1H, *H1*), 7.75 (ddd, *J* = 8.7, 6.5, 1.3 Hz, 1H, *H3*), 7.69 (nfod, J_{app} = 8.7 Hz, 2H, *H2'* and *H6'*), 7.47 (ddd, *J* = 8.9, 6.5, 1.3 Hz, 1H, *H2*), 7.07 (nfod, J_{app} = 8.8 Hz, 2H, *H3'* and *H5'*), 7.05 (s, 1H, *H9*), 5.67 (s, 1H, *H12*), 3.87 (s, 3H, OCH_3), 3.84 (s, 3H, OCH_3), 3.55 (s, 3H, C11-OCH_3), 3.05 (s, 3H, Ar-CH_3), and 0.51 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (125 MHz, $CDCl_3$): 195.4, 160.7, 153.1, 150.7, 149.0, 148.0, 146.8, 145.0, 143.2, 140.1, 137.0, 136.0, 134.0, 131.0, 130.9, 130.2, 126.5, 125.4, 125.3, 121.0, 114.5, 109.6, 106.4, 56.6, 56.1, 55.6, 20.6, and 2.6. (missing one aromatic C; one of the resonances at 134.0, 126.5, and 114.4 ppm is likely two non-resolved resonances)

HRMS (ESI-TOF): Calcd for $C_{33}H_{32}NO_4Si^+$ $[M+H^+]^+$ requires 534.2095; found 534.2081.

IR (neat): 3068, 3000, 2945, 2901, 2838, 2023, 1915, 1703, 1605, 1586, 1539, 1494, 1459, 1406, 1378, 1367, 1329, 1294, 1250, 1217, 1177, 1133, 1112, 1099, 1061, 1029, 1017, 949, 842, 813, 790, 763, 733, 701, 670, 652, 633, 621, 608, 588, 570, 529, 505, 493, 476, 450, 420, and 406 cm^{-1}.

10,11-Dimethoxy-6-methyl-2-nitro-13-phenyl-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (SI-3):



8 ***SI-3***, 62%

N-(4-Nitrophenyl)-1-phenylmethanimine (**2d**) was prepared according to a reported procedure with the modification that elevated temperature was used to increase the rate of imine formation.[8]

Triynone **8** (20 mg, 0.0616 mmol, 1 equiv) and the imine **2d** (45 mg, 0.185 mmol, 3 equiv) were added to a screw-capped culture tube containing freshly activated 4Å molecular sieves. 1,2-DCE was added (0.05 M) and the resulting solution was placed in an oil bath maintained at 85 °C and kept overnight. The solvent was partially removed under reduced pressure, chloroform was added, and the mixture was directly loaded onto the MPLC column and eluted (2:1 hexanes:EtOAc) to give **SI-3** (21 mg, 62%) as a dark red amorphous solid.

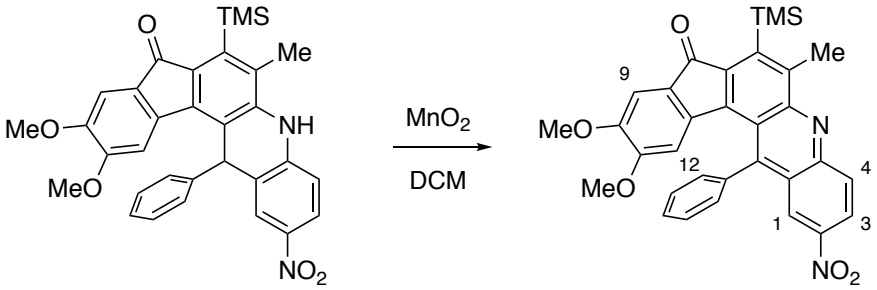
The extremely low solubility of this material, likely due to strong hydrogen bonding in the crystal lattice of this *p*-nitroaniline derivative, precluded obtaining a suitable ^{13}C NMR spectrum.

^{1}H NMR (500 MHz, $CDCl_3$): δ 8.39 (d, *J* = 2.6 Hz, 1H, *H1*), 8.05 (dd, *J* = 8.8, 2.5 Hz, 1H, *H3*), 7.34–7.30 (m, PhH_o and PhH_m), 7.18 (tt, J = 8.6, 1.5 Hz, PhH_p), 7.11 (s, 1H, *H9* or *H12*), 7.04 (s, 1H, *H9* or *H12*), 6.91 (br s, 1H, N*H*), 6.88 (d, *J* = 8.9 Hz, 1H, *H4*), 5.81 (s, 1H, *H13*), 3.94 (s, 3H, OCH_3), 3.87 (s, 3H, OCH_3), 2.50 (s, 3H, ArCH_3), and 0.47 (s, 9H, Si(CH_3)$_3$).

HRMS (ESI-TOF): Calcd for $C_{32}H_{31}N_2O_5Si^+$ $[M+H^+]^+$ requires 551.1997; found 551.1945 (major ion) and Calcd for $C_{32}H_{29}N_2O_5Si^+$ $[M-2H+H^+]^+$ requires 549.1840; found 549.1817 (minor ion).

IR (neat): 3379, 3084, 2954, 2904, 2852, 2368, 2136, 2074, 2041, 1952, 1700, 1675, 1600, 1576, 1545, 1522, 1490, 1457, 1427, 1408, 1375, 1321, 1298, 1244, 1213, 1191, 1086, 1045, 1023, 990, 967, 950, 864, 841, 797, 780, 769, 741, 675, 657, 639, 617, 607, 577, 550, 520, 495, 450, 420, and 409 cm^{-1}.

10,11-Dimethoxy-6-methyl-2-nitro-13-phenyl-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12d):



SI-3 ***12d***, 86%

The acridine **12d** was synthesized according to General Procedure B: 14 mg of amine **SI-3** (1 equiv) and 22 mg of MnO_2 (10 equiv). The reaction mixture was filtered through Celite® (DCM elution) to afford **12d** (12 mg, 82%) as a purple amorphous solid.

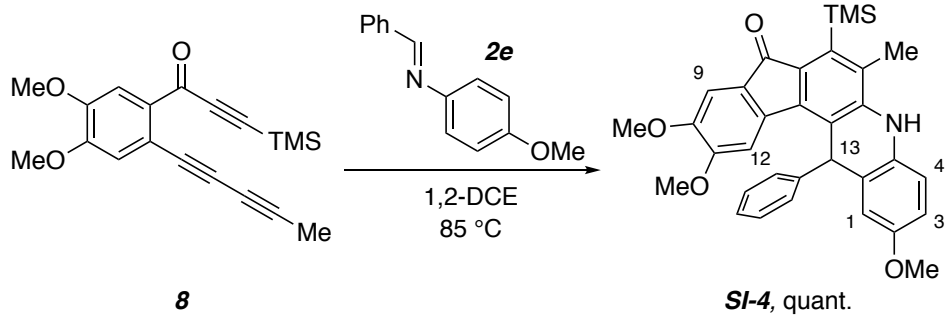
^{1}H NMR (500 MHz, $CDCl_3$): δ 9.15 (d, 2.4 Hz, 1H, *H1*), 8.47 (dd, *J* = 9.5, 2.3 Hz, 1H, *H3*), 8.36 (d, *J* = 9.5 Hz, 1H, *H4*), 7.81–7.77 (nfom, 2H, PhH_o), 7.66–7.62 (m, 3H, PhH_m and PhH_p), 7.06 (s, 1H, *H9*), 5.60 (s, 1H, *H12*), 3.84 (s, 3H, OCH_3), 3.51 (s, 3H, OCH_3), 3.05 (s, 3H, ArCH_3), and 0.52 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 194.9, 153.3, 152.2, 149.6, 148.7, 148.5, 147.2, 145.6, 142.9, 139.6, 139.2, 138.3, 137.4, 132.8, 132.7, 130.6, 129.6, 125.0, 124.9, 123.2, 123.1, 121.4, 109.1, 106.7, 56.7, 56.2, 20.6, and 2.6.

HRMS (ESI-TOF): Calcd for $C_{32}H_{29}N_2O_5Si^+$ $[M+H^+]^+$ requires 549.1840; found 549.1827.

IR (neat): 3058, 3001, 2943, 2903, 2851, 2194, 2152, 2069, 2021, 1977, 1707, 1620, 1602, 1585, 1560, 1540, 1518, 1494, 1465, 1456, 1417, 1401, 1370, 1335, 1293, 1246, 1218, 1182, 1159, 1131, 1104, 1086, 1064, 1028, 1014, 971, 928, 909, 867, 839, 814, 803, 788, 767, 738, 707, 679, 664, 642, 623, 586, 565, 542, 520, 494, 458, 433, and 414 cm^{-1}.

2,10,11-Trimethoxy-6-methyl-13-phenyl-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (SI-4)



8 ***SI-4,*** quant.

N-(4-Methoxyphenyl)-1-phenylmethanimine (**2e**) was prepared according to a reported procedure.[9]

The dihydroacridine **SI-4** was synthesized according to General Procedure A using the triyne **8** (25 mg) and the imine **2e** (49 mg, 3 equiv). Purification by MPLC (2:1 hexanes:EtOAc) afforded (41 mg, 100%) as a red amorphous solid.

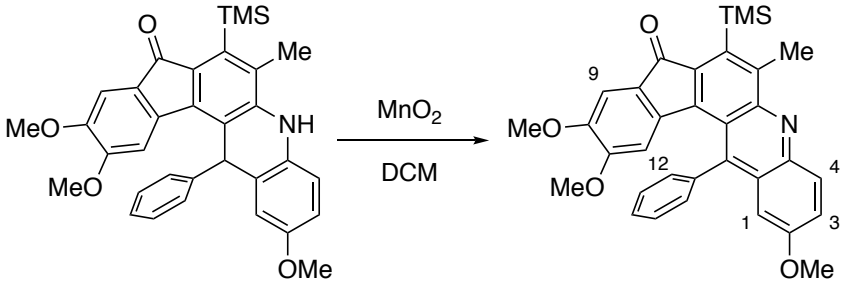
^{1}H NMR (500 MHz, $CDCl_3$): δ 7.29 (nfod, J_{app} = 8.2 Hz, 2H, ArH_o), 7.22 (nfodd, J_{app} = 7.5, 7.5 Hz, 2H, PhH_m), 7.12 (tt, J = 7.3, 1.4 Hz, 1H, ArH_p), 7.09 (s, 1H, *H9*), 7.08 (s, 1H, *H12*), 6.99 (d, J = 2.6 Hz, 1H, *H1*), 6.75 (d, J = 8.6 Hz, 1H, *H4*), 6.70 (dd, J = 8.6, 2.7 Hz, 1H, *H3*), 6.39 (s, 1H, N*H*), 5.73 (s, 1H, *H13*), 3.90 (s, 3H, OCH_3), 3.85 (s, 3H, OCH_3), 3.78 (s, 3H, OCH_3), 2.42 (s, 3H, ArCH_3), and 0.46 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (125 MHz, $CDCl_3$): 193.4, 155.2, 152.9, 149.0, 144.6, 143.4, 142.6, 141.1, 137.4, 133.3, 131.5, 129.0, 128.8, 127.1, 127.0, 125.1, 124.8, 118.1, 115.9, 114.0, 113.4, 107.8, 106.6, 56.6, 56.2, 55.9, 45.0, 18.2, and 3.2.

HRMS (ESI-TOF): Calcd for $C_{33}H_{34}NO_4Si^+$ $[M+H^+]^+$ requires 536.2252; found 536.2219 (major ion) and Calcd for $C_{33}H_{32}NO_4Si^+$ $[M-2H+H^+]^+$ requires 534.2095; found 534.2084 (minor ion).

IR (neat): 3440, 3392, 3058, 3020, 2999, 2941, 2900, 2834, 2361, 2339, 2225, 2182, 2153, 2028, 1957, 1692, 1602, 1575, 1546, 1492, 1462, 1442, 1411, 1374, 1343, 1309, 1284, 1259, 1245, 1212, 1173, 1164, 1153, 1131, 1118, 1091, 1043, 1029, 990, 952, 863, 841, 797, 768, 734, 701, 644, 627, 609, 567, 552, 535, 514, 493, 459, 451, 416, and 405 cm^{-1}.

2,10,11-Trimethoxy-6-methyl-13-phenyl-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12e):



SI-4 ***12e***, 94%

The acridine **12e** was synthesized according to General Procedure B (36 mg of amine **SI-4**). The reaction mixture was filtered through Celite® (DCM elution) to afford **12e** (36 mg, quant.) as a purple oil. This material was purified by MPLC on silica gel (2:1 hexanes:EtOAc) to give **12e** (34 mg, 94%) as a purple amorphous solid.

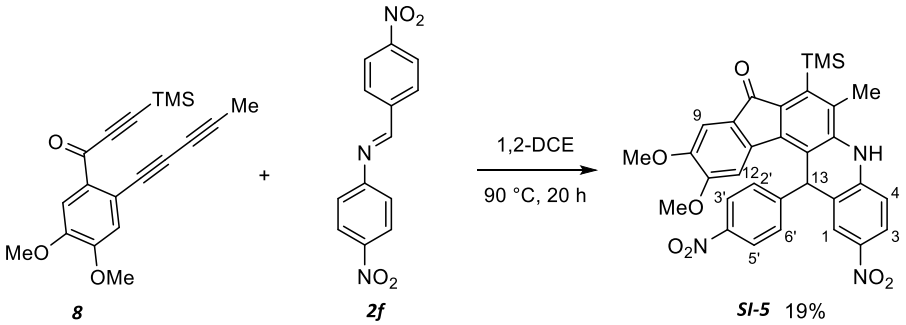
^{1}H NMR (500 MHz, $CDCl_3$): δ 8.15 (d, 9.4 Hz, 1H, *H4*), 7.78 (nfod, J_{app} = 6.9 Hz, 2H, PhH_o), 7.55 (nfodd, J_{app} = 7.1, 7.1 Hz, 2H, PhH_m), 7.49 (tt, J = 7.3, 1.5 Hz, 1H, PhH_p), 7.45 (br dd, J = 9.4, 2.7 Hz, 1H, *H3*), 7.37 (d, J = 2.6 Hz, 1H, *H1*), 7.03 (s, 1H, *H9*), 5.65 (s, 1H, *H12*), 3.831 (s, 3H, C2- or C10-OCH_3), 3.828 (s, 3H, C2- or C10-OCH_3), 3.52 (s, 3H, C11-OCH_3), 3.04 (s, 1H, ArCH_3), and 0.50 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 195.5, 158.0, 153.0, 149.2, 147.9, 146.9, 146.2, 142.4, 142.3, 140.3, 139.1, 137.5, 134.9, 132.5, 132.4, 129.3, 129.0, 126.0, 125.3, 125.0, 121.0, 109.2, 106.4, 101.9, 56.6, 56.1, 55.5, 20.6, and 2.7.

HRMS (ESI-TOF): Calcd for $C_{33}H_{32}NO_4Si^+$ $[M+H^+]^+$ requires 534.2095; found 534.2079.

IR (neat): 3120, 3057, 2999, 2947, 2902, 2835, 2356, 2194, 2048, 1970, 1703, 1604, 1586, 1552, 1524, 1512, 1493, 1467, 1442, 1406, 1375, 1369, 1354, 1293, 1261, 1247, 1227, 1210, 1171, 1157, 1120, 1102, 1066, 1028, 1016, 976, 868, 839, 831, 808, 792, 768, 739, 706, 676, 654, 636, 613, 587, 577, 554, 540, 521, 497, 460, 437, 424, and 410 cm^{-1}.

10,11-Dimethoxy-6-methyl-2-nitro-13-(4-nitrophenyl)-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (SI-5)



8 *2f* ***SI-5*** 19%

Following general procedure A, 1-(4,5-dimethoxy-2-(penta-1,3-diyn-1-yl)phenyl)-3-(trimethylsilyl)prop-2-yn-1-one (**8**, 0.025 g, 0.078 mmol, 1 equiv), *N*,1-bis(4-nitrophenyl)methanimine (**2f**, 0.063 g, 0.23 mmol, 3.0 equiv), and 1,2-dichloroethane (2 mL) were used to prepare the 1,4-dihydroacridine **SI-5**. Purification of the crude product by MPLC (3:1 hexanes:EtOAc) yielded **SI-5** (0.009 g, 0.015 mmol, 19%) as an orange amorphous solid.

^{1}H NMR (500 MHz, $CDCl_3$): δ 8.38 (d, J = 2.4 Hz, 1H, Ar*H1*), 8.14 (nfod, J_{app} = 8.8 Hz, 2H, ArH*3'* and Ar*H5'*), 8.10 (dd, J = 8.9, 2.5 Hz, 1H, *H3*), 7.47 (nfod, J_{app} = 8.8 Hz, 2H, Ar*H2'* and Ar*H6'*), 7.14 (s, 1H, Ar*H9*), 6.97 (s, 1H, Ar*H12*), 6.95 (br s, 1H, N*H*), 6.92 (d, J = 8.9 Hz, 1H, *H4*) 5.95 (s, 1H, *H13*), 3.96 (s, 3H, OCH_3), 3.88 (s, 3H, OCH_3), 2.51 (s, 3H, ArCH_3), and 0.47 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 192.6, 153.6, 150.3, 149.8, 147.4, 143.0, 143.0, 142.5, 142.2, 140.3, 136.6, 135.9, 128.5, 127.9, 126.7, 125.2, 124.8, 124.8, 122.3, 116.7, 115.3, 107.2, 107.1, 56.7, 56.3, 44.2, 18.3, and 3.1.

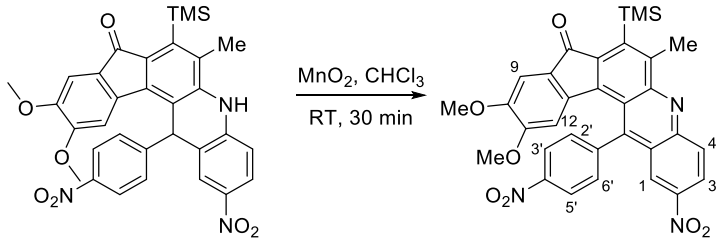
HRMS (ESI-TOF): Calcd for $C_{32}H_{30}N_3O_7Si^+$ $[M+H^+]^+$ requires 596.1848; found 596.1814.

IR (neat): 3628, 3380, 3006, 2946, 2837, 1890, 1700, 1602, 1551, 1523, 1495, 1462, 1409, 1375, 1345, 1327, 1301, 1245, 1213, 1111, 1089, 1046, 1015, 990, 949, 865, 854, 833, 797, 773, 726, 699, 675, 667, 640, 609, 581, 565, 555, 522, 492, 482, 455, 435, and 412 cm^{-1}.

N,1-bis(4-Nitrophenyl)methanimine (**2f**) was prepared according to the following procedure:

p-Nitroaniline (0.30 g, 2.2 mmol, 1 equiv) and *p*-nitrobenzaldehyde (0.33 g, 2.2 mmol, 1 equiv) were added to a 50 mL round-bottom flask and suspended in 20 mL of toluene. The solution was heated to refluxed, during which time it became homogeneous, for 12 hours. The reaction solution was allowed to cool to room temperature. During this time, a yellow solid crystallized from the solution. The crystals were collected via vacuum filtration and washed with cold hexanes. Yellow needles of *N*,1-bis(4-nitrophenyl)methanimine (**2f**, ca. 0.5 g) were obtained and used without further purification. The ^{1}H NMR spectral data were consistent with previously reported data.[10]

10,11-Dimethoxy-6-methyl-2-nitro-13-(4-nitrophenyl)-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12f):



2f **12f**, 56%

Following General Procedure B, 10,11-dimethoxy-6-methyl-2-nitro-13-(4-nitrophenyl)-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (**SI-5**, 0.009 g, 0.015 mmol, 1 equiv), MnO_2 (xs), and $CHCl_3$ (10 mL) were used to prepare acridine **12f**. This oxidation reaction was accompanied by formation of a second, bright yellow compound of higher polarity, which required separation by MPLC (3:1 hexanes:EtOAc). This byproduct was not fully identified, but its ^{1}H NMR spectrum suggested that, at least, the aromatic methyl group had been oxidized to an aldehyde. Purification of the crude product yielded **12f** (0.004 g, 0.007 mmol, 56%) as a dark green amorphous solid.

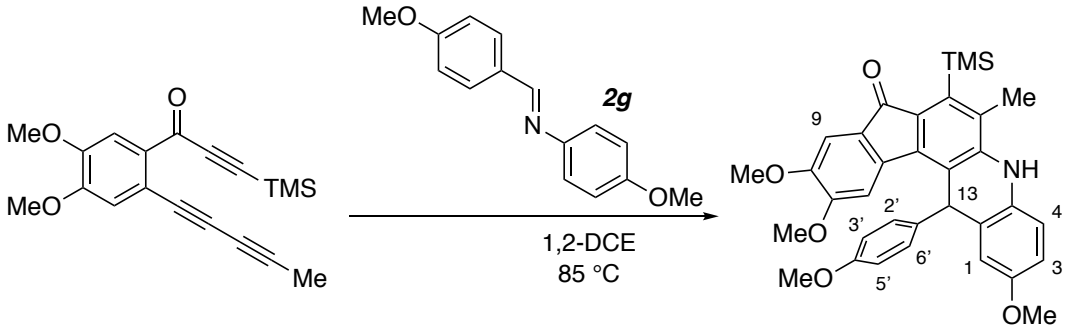
^{1}H NMR (500 MHz, $CDCl_3$): δ 8.99 (d, J = 2.4 Hz, 1H, *H1*), 8.52 (dd, J = 9.4, 2.4 Hz, 1H, *H3*), 8.50 (nfod, J_{app} = 8.8 Hz, 2H, *H3'* and *H5'*), 8.42 (d, J = 9.4 Hz, 1H, *H4*), 8.01 (nfod, J_{app} = 8.7 Hz, 2H, *H2'* and *H6'*), 7.08 (s, 1H, *H9*), 5.56 (s, 1H, *H12*), 3.84 (s, 3H, OCH_3), 3.50 (s, 3H, OCH_3), 3.07 (s, 1H, ArCH_3), and 0.53 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 194.4, 153.2, 151.9, 149.4, 149.0, 148.7, 147.8, 146.1, 145.3, 143.5, 141.8, 139.8, 138.9, 138.8, 133.5, 133.3, 125.2, 124.5, 123.4, 123.3, 122.6, 120.9, 108.7, 107.3, 56.6, 56.2, 20.7, and 2.5.

HRMS (ESI-TOF): Calcd for $C_{32}H_{28}N_3O_7Si^+$ $[M+H^+]^+$ requires 594.1691; found 594.1676.

IR (neat): 3103, 3078, 3000, 2944, 2902, 2873, 2854, 2840, 1948, 1707, 1620, 1599, 1586, 1561, 1521, 1494, 1464, 1455, 1416, 1401, 1347, 1336, 1294, 1246, 1217, 1181.3, 1158.4, 1132, 1108, 1086, 1065, 1012, 990, 925, 906, 869, 854, 838, 813, 800, 787, 755, 726, 704, 682, 667, 652, 639, 606, 586, 566, 542, 519, 495, 484, 453, and 416 cm^{-1}.

2,10,11-Trimethoxy-13-(4-methoxyphenyl)-6-methyl-7-(trimethylsilyl)-5,13-dihydro-8H-indeno[1,2-a]acridin-8-one (SI-6)



8 ***SI-6***, 88%

N,1-bis(4-Methoxyphenyl)methanimine (**2g**) was prepared according to a reported procedure.[11]

The dihydroacridine **SI-6** was synthesized according to General Procedure A using the triyne **8** (25 mg) and the imine **2g** (56 mg, 3 equiv). Purification by MPLC (2:1 hexanes:EtOAc) afforded (38 mg, 88%) as a red amorphous solid.

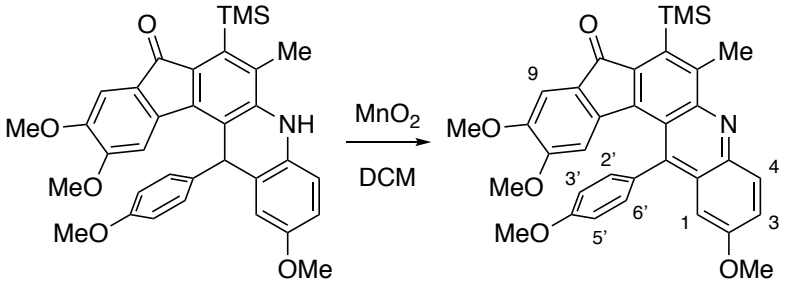
^{1}H NMR (400 MHz, $CDCl_3$): δ 7.18 (nfod, J_{app} = 8.7 Hz, 2H, Ar*H2'* and Ar*H6'*), 7.099 (s, 1H, Ar*H9* or Ar*H12*), 7.096 (s, 1H, Ar*H9* or Ar*H12*), 6.97 (d, *J* = 2.5 Hz, 1H, *H1*), 6.76 (nfod, J_{app} = 8.8 Hz, 2H, Ar*H3'* and Ar*H5'*), 6.75 (d, *J* = 8.6 Hz, 1H, *H4*), 6.72 (dd, *J* = 8.6, 2.7 Hz, 1H, *H3*), 6.36 (s, 1H, N*H*), 5.68 (s, 1H, *H13*), 3.92 (s, 3H, OCH_3), 3.86 (s, 3H, OCH_3), 3.79 (s, 3H, OCH_3), 3.69 (s, 3H, OCH_3), 2.42 (s, 3H, ArCH_3), and 0.45 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (125 MHz, $CDCl_3$): 193.5, 158.5, 155.3, 152.9, 149.0, 143.4, 142.6, 141.0, 137.4, 136.8, 133.3, 131.5, 128.8, 128.1, 125.2, 125.0, 118.4, 115.9, 114.4, 114.0, 113.3, 107.8, 106.6, 56.6, 56.2, 55.9, 55.3, 44.1, 18.2, and 3.2.

HRMS (ESI-TOF): Calcd for $C_{34}H_{34}NO_5Si^+$ $[M-2H]^+$ requires 564.2201; found 564.2193 (major ion) and Calcd for $C_{34}H_{36}NO_5Si^+$ $[M+H^+]^+$ requires 566.2357; found 566.2316 (minor ion).

IR (neat): 3440, 3386, 2998, 2948, 2904, 2834, 2337, 2320, 2074, 2009, 1732, 1689, 1604, 1576, 1506, 1492, 1442, 1413, 1374, 1343, 1309, 1285, 1177, 1152, 1131, 1111, 1091, 1032, 990, 952, 862, 839, 792, 739, 687, 642, 629, 607, 567, 544, 480, 447, and 409 cm^{-1}.

2,10,11-Trimethoxy-13-(4-methoxyphenyl)-6-methyl-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (12g):



SI-6 ***12g,*** 89%

The acridine **12g** was synthesized according to General Procedure B (19 mg of amine **SI-6**). The reaction mixture was filtered through Celite® (DCM elution) to afford crude **12g** as a purple oil. This material was purified by MPLC on silica gel (1:1 hexanes:EtOAc) to give **12g** (17 mg, 89%) as a purple amorphous solid.

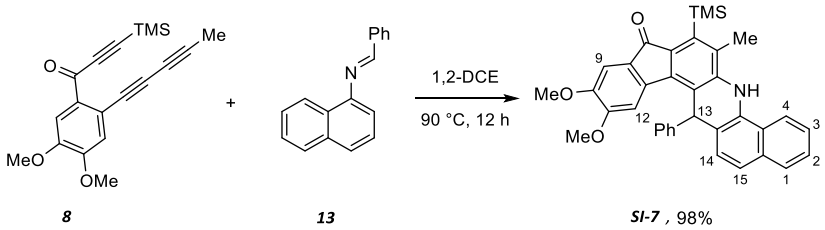
^{1}H NMR (400 MHz, $CDCl_3$): δ 8.14 (d, J = 9.3 Hz, 1H, *H4*), 7.70 (nfod, J_{app} = 8.7 Hz, 2H, Ar*H2'* and Ar*H6'*), 7.45 (dd, J = 9.2, 2.7 Hz, 1H, *H3*), 7.39 (d, J = 2.6 Hz, 1H, *H1*), 7.07 (nfod, J_{app} = 8.7 Hz, 2H, Ar*H3'* and Ar*H5'*), 7.05 (s, 1H, *H9*), 5.68 (s, 1H, *H12*), 3.87 (s, 3H, OCH_3), 3.85 (s, 3H, OCH_3), 3.84 (s, 3H, OCH_3), 3.56 (s, 3H, OCH_3), 3.04 (s, 1H, ArCH_3), and 0.50 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (100 MHz, $CDCl_3$): 195.5, 160.5, 158.0, 153.1, 149.3, 147.9, 146.9, 146.2, 142.6, 142.3, 140.3, 137.3, 134.9, 133.6, 132.5, 131.4, 126.2, 125.4, 124.9, 121.3, 114.5, 109.6, 106.4, 102.0, 56.6, 56.1, 55.6, 55.6, 20.5, and 2.7.

HRMS (ESI-TOF): Calcd for $C_{34}H_{34}NO_5Si^+$ $[M-2H]^+$ requires 564.2201; found 564.2182

IR (neat): 3057, 3000, 2836, 2682, 2544, 2437, 2355, 2254, 2044, 1915, 1701, 1623, 1604, 1585, 1513, 1492, 1464, 1440, 1405, 1291, 1246, 1223, 1207, 1175, 1115, 1099, 1066, 1026, 1014, 868, 807, 766, 646, 612, 489, and 424 cm^{-1}.

10,11-Dimethoxy-6-methyl-13-phenyl-7-(trimethylsilyl)-5,13-dihydro-8H-benzo[h]indeno[1,2-a]acridin-8-one (SI-7)



8 *13* *SI-7*, 98%

N-(Naphthalen-1-yl)-1-phenylmethanimine (**13**) was prepared according to a reported procedure.[12]

Following general procedure A, triynone **8** (0.010 g, 0.030 mmol, 1 equiv), (*E*)-*N*-(naphthalen-1-yl)-1-phenylmethanimine (**13**, 0.023 g, 0.10 mmol, 3.3 equiv), and dichloroethane (1 mL) were used to prepare the 1,4-dihydroacridine **SI-7**. Purification of the crude product by MPLC (2:1 hexanes:EtOAc) yielded **SI-7** (0.016 g, 0.029 mmol, 98%) as an orange amorphous solid.

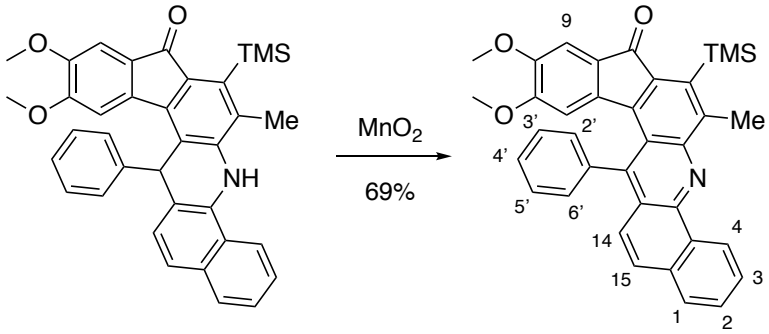
^{1}H NMR (500 MHz, $CDCl_3$): δ 7.814 (dd, *J* = 7.9, 0.9 Hz, 1H, *H1* or *H4*), 7.809 (d, *J* = 8.0, 1.6 Hz, 1H, *H1* or *H4*), 7.57 (d, *J* = 8.4 Hz, 1H, *H14*), 7.55 (dd, *J* = 8.4, 6.8, 1.4 Hz, 1H, *H2* or *H3*), 7.47 (dd, *J* = 8.1, 6.8, 1.2 Hz, 1H, *H2* or *H3*), 7.47 (br d, *J* = 8.5 Hz, 1H, *H15*), 7.35 (nfod, J_{app} = 7.3 Hz, 2H, PhH_o), 7.27 (s, 1H, N*H*), 7.22 (nfodd, J_{app} = 7.5, 7.5 Hz, 2H, PhH_m), 7.15 (s, 1H, *H9* or *H12*), 7.11 (s, 1H, *H9* or *H12*), 7.13 (tt, *J* = 7.7, 1.5 Hz, 1H, PhH_p), 5.90 (s, 1H, *H13*), 3.93 (s, 3H, OCH_3), 3.87 (s, 3H, OCH_3), 2.62 (s, 3H, ArCH_3), and 0.49 (s, 9H, Si(CH_3)$_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 193.6, 153.1, 149.1, 144.9, 142.7, 142.6, 141.2, 137.5, 134.4, 133.2, 131.8, 129.11, 129.08, 128.6, 127.2, 127.1, 126.8, 126.2, 125.97, 125.95, 122.2, 122.0, 119.0, 118.6, 118.2, 107.8, 106.7, 56.6, 56.2, 45.2, 18.2, and 3.2.

HRMS (ESI-TOF): Calcd for $C_{36}H_{32}NO_3Si^+$ $[M-2H]^+$ requires 554.2146; found 554.2132 (most intense ion) and Calcd for $C_{36}H_{34}NO_3Si^+$ $[M+H^+]^+$ requires 556.2302; found 556.2260 (minor ion).

IR (neat): 3467, 3370, 3290, 3057, 3002, 2935, 2904, 854, 2837, 2359, 2183, 2123, 2048, 2007, 1977, 1949, 1712, 1694, 1614, 1578, 1550, 1516, 1492, 1460, 1406, 1375, 1349, 1310, 1293, 1277, 1251, 1214, 1173, 1130, 1094, 1018, 971, 946, 853, 804, 793, 771, 752, 736, 715, 703, 679, 668, 643, 626, 598, 582, 563, 542, 527, 496, 443, 432, 421, and 409 cm^{-1}.

10,11-Dimethoxy-6-methyl-13-phenyl-7-(trimethylsilyl)-8H-benzo[h]indeno[1,2-a]acridin-8-one (14)



SI-7 ***14***

Acridine **14** was prepared according to General Procedure B starting with 16 mg of amine **SI-7**. Purification by MPLC (3:1 hexanes:EtOAc) afforded **14** (11 mg, 69%) as a purple amorphous solid.

^{1}H NMR (500 MHz, C_6D_6): 9.47 (dd, *J* = 8.2, 1.2 Hz, 1H, *H4*), 7.79 (d, *J* = 9.4 Hz, 1H, *H14*), 7.63 (ddd, *J* = 8.2, 7.0, 1.3 Hz, 1H, *H3* or *H2*), 7.60 (dd, *J* = 7.9, 1.6 Hz, 1H, *H1*), 7.50 (ddd, *J* = 8.2, 7.0, 1.4 Hz, 1H, *H3* or *H2*), 7.39 (d, *J* = 9.4, 0.9 Hz, 1H, *H15*), 7.32–7.30 (nfom, 2H, *H2'* and *H6'*), 7.12 (s, 1H, *H9*), 7.02–6.97 (m, 3H, *H3', H4'* and *H5'*), 5.57 (s, 1H, *H12*), 3.39 (s, 3H, OCH_3), 3.23 (s, 3H, OCH_3), 3.20 (s, 1H, ArCH_3), and 0.79 (s, 9H, Si(CH_3)$_3$).

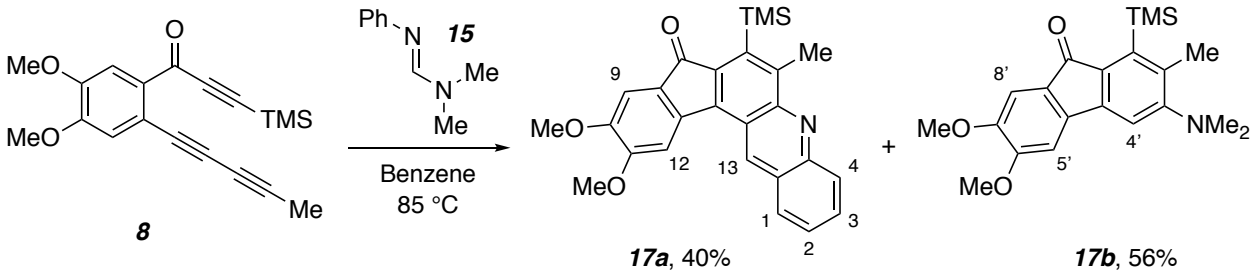
^{13}C NMR (126 MHz, C_6D_6): δ 195.2, 153.9, 149.7, 149.3, 147.8, 147.2, 144.4, 143.1, 140.3, 138.9, 138.2, 136.8, 134.1, 132.9, 132.8, 129.7, 129.0, 128.8, 128.3, 127.9, 126.5, 126.1, 123.9, 123.5, 121.9, 110.4, 107.3, 56.4, 55.3, 21.0, and 3.1. (one aromatic signal wasn't observed, perhaps obscured by the solvent resonances).

^{13}C NMR (126 MHz, $CDCl_3$): 195.4, 153.1, 149.1, 148.1, 147.5, 146.9, 144.0, 142.6, 140.2, 138.7, 137.6, 136.6, 133.7, 132.8, 132.3, 129.5, 129.4, 129.0, 128.4, 127.7, 127.6, 126.0, 125.5, 123.6, 123.2, 121.4, 109.4, 106.4, 56.7, 56.1, 20.6, and 2.8.

HRMS (ESI-TOF): Calcd for $C_{36}H_{32}NO_3Si^+$ $[M+H^+]^+$ requires 554.2146; found 554.2128.

IR (neat): 3121, 3057, 2999, 2947, 2900, 2874, 2851, 2837, 2684, 2315, 2180, 2052, 1949, 1703, 1602, 1584, 1558, 1492, 1464, 1441, 1415, 1395, 1376, 1364, 1352, 1329, 1295, 1247, 1229, 1215, 1184, 1154, 1102, 1080, 1067, 1037, 1021, 1008, 978, 889, 850, 834, 803, 776, 747, 704, 683, 661, 637, 624, 600, 572, 552, 526, 501, 456, and 413 cm^{-1}.

10,11-Dimethoxy-6-methyl-7-(trimethylsilyl)-8H-indeno[1,2-a]acridin-8-one (17a)
and
3-(Dimethylamino)-6,7-dimethoxy-2-methyl-1-(trimethylsilyl)-9H-fluoren-9-one (17b)



Amidine **15** was prepared according to a reported procedure.[13]

Triynone **8** (20 mg, 0.0616 mmol, 1 equiv) and the formamidine **15** (28 mg, 0.185 mmol, 3 equiv) were added to a screw-capped culture tube containing freshly activated 4Å molecular sieves. Benzene was added (0.05 M) and the resulting solution was placed in an oil bath maintained at 85 °C and kept overnight. The solvent was then removed under reduced pressure, and the residue was purified by MPLC (2:1 hexanes:EtOAc) to give a coeluting mixture of **17a** (40% yield) and **17b** (56% yield).

Data for 17a and 17b (a mixture of coeluting products in a ratio of 2:3)

Data for 17a, the acridine adduct, minor product:

^{1}H NMR (500 MHz, $CDCl_3$): δ 9.23 (s, 1H, *H13*), 8.24 (dd, *J* = 8.9, 1.0 Hz, 1H, *H4*), 8.04 (dd, *J* = 8.6, 1.5 Hz, 1H, *H1*), 7.81 (ddd, *J* = 8.7, 6.6, 1.4 Hz, 1H, *H3*), 7.61 (s, 1H, *H12*), 7.57 (ddd, *J* = 8.1, 6.6, 1.3 Hz, 1H, *H2*), 7.23 (s, 1H, *H9*), 4.15 (s, 3H, OCH_3), 3.96 (s, 3H, OCH_3), 3.11 (s, 3H, ArCH_3), and 0.52 (s, 9H, Si(CH_3)$_3$).

HRMS (ESI-TOF): Calcd for $C_{26}H_{26}NO_3Si^+$ $[M+H^+]^+$ requires 428.1676; found 428.1674.

^{13}C NMR (126 MHz, $CDCl_3$): 195.2, 153.3, 150.1, 149.3, 146.0, 142.0, 138.7, 137.4, 135.8, 132.6, 131.2, 130.6, 128.4, 126.8, 126.8, 126.6, 122.0, 107.7, 107.3, 102.6, 56.8, 56.3, 20.2, and 3.2.

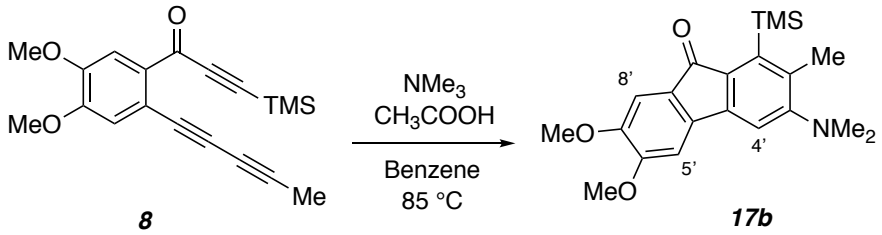
Data for 17b, the amine-trapped adduct, major product:

^{1}H NMR (500 MHz, $CDCl_3$): δ 7.11 (s, 1H, *H8'*), 7.01 (s, 1H, *H5'*), 6.93 (s, 1H, *H4'*), 4.01 (s, 3H, OCH_3), 3.96 (s, 3H, OCH_3), 3.11 (s, 3H, ArCH_3), and 0.52 (s, 9H, Si(CH_3)$_3$).

HRMS (ESI-TOF): Calcd for $C_{21}H_{28}NO_3Si^+$ $[M+H^+]^+$ requires 370.1833; found 370.1832.

^{13}C NMR (125 MHz, $CDCl_3$): 193.9, 157.6, 154.0, 149.5, 144.0, 143.9, 138.6, 135.7, 133.8, 127.7, 109.9, 106.8, 102.6, 56.5, 56.3, 43.9, 21.1, and 2.8.

An authentic sample of the amine-trapped product **17b** was also prepared using the following procedure:



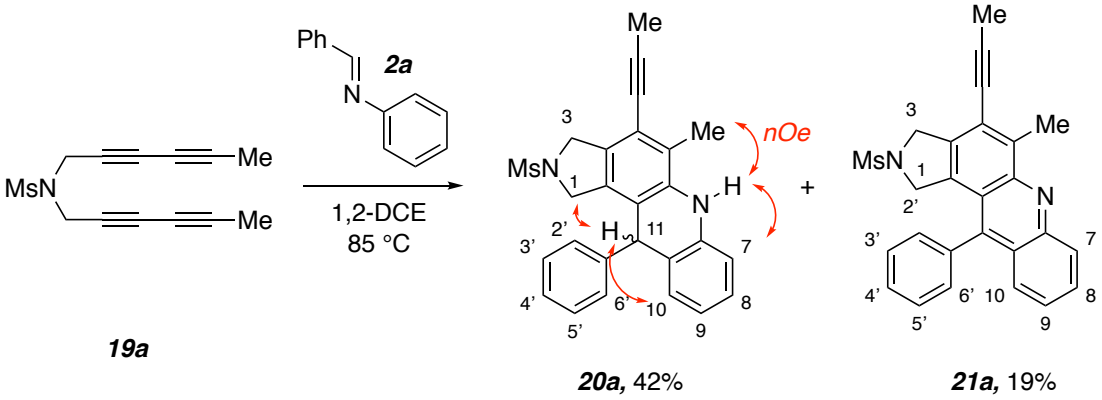
8 ***17b***

Triynone **8** (20 mg, 0.0616 mmol, 1 equiv) and acetic acid (5.42 μL, 0.092 mmol, 1.5 equiv) were added to a screw-capped culture tube. Benzene was added (0.05 M) and trimethylamine gas was bubbled through the mixture. This solution was placed in an oil bath maintained at 85 °C and kept overnight. The solvent was then removed under reduced pressure, and the residue was purified by MPLC (2:1 hexanes:EtOAc) to give **17b** as an orange oil. ^{1}H and ^{13}C spectrum for **17b** from this experiment were used as a reference to aid in the interpretation of the NMR data obtained from the sample of a mixture of the acridine and amine adducts **17a** and **17b** (from the above experiment).

5-Methyl-2-(methylsulfonyl)-11-phenyl-4-(prop-1-yn-1-yl)-2,3-dihydro-1H-pyrrolo[3,4-a]acridine (20a)

and

5-Methyl-2-(methylsulfonyl)-11-phenyl-4-(prop-1-yn-1-yl)-2,3-dihydro-1H-pyrrolo[3,4-a]acridine (21a):



This reaction was performed using general Procedure A: 20 mg of polyyne **19a** (1 equiv) and 55 mg of the imine **2a** (3 equiv). Purification by MPLC (2:1 hexanes:EtOAc) afforded the 1,4-dihydroacridine **20a** (18 mg, 42%) as a pale yellow crystalline solid along with the corresponding acridine **21a** (8 mg, 19%), which presumably arises through air oxidation, also as a yellow crystalline solid.

The assignment of the structure of the dihydroacridine **20a** was based upon observed (difference) nOe interactions between i) the amine proton (*NH*) with the adjacent aryl methyl group and the H7 aromatic proton; and ii) the methine proton (*H11*) with the adjacent methylene (C1*H2*) and H10 aromatic proton.

Data for acridine 21a, the faster eluting minor product:

^{1}H NMR (400 MHz, $CDCl_3$): δ 8.28 (ddd, *J* = 8.8, 1.0, 1.0 Hz, 1H, *H7*), 7.75 [(ddd, *J* = 8.3, 6.3, 1.6 Hz, 1H, *H9*], 7.63 (tt, *J* = 7.4, 1.4 Hz, 1H, *H4'*), 7.56 (nfom, 1H, *H3'* and *H5'*), 7.47–7.38 [overlapped m, 2H, *H10* and *H8*], 7.36 (nfod, J_{app} = 6.8 Hz, 2H, *H2'* and *H6'*), 4.79 (br t, *J* = 3.3 Hz, 2H, *C3H$_2$*), 4.04 (br t, *J* = 3.2 Hz, 2H, *C1H$_2$*), 3.09 (s, 3H, CH_3SO_2N), 2.72 (s, 3H, NArCH3), and 2.20 (s, 3H, C≡CCH_3).

^{13}C NMR (100 MHz, $CDCl_3$): 148.2, 147.7, 145.2, 141.4, 137.5, 135.6, 130.3, 130.2, 130.0, 129.4, 128.7, 128.4, 126.7, 126.5, 126.1, 120.6, 119.5, 96.8, 76.5, 55.8, 54.8, 34.9, 16.8, and 5.0.

HRMS (ESI-TOF): Calcd for $C_{26}H_{23}N_2O_2S^+$ $[M+H^+]^+$ requires 427.1475; found 427.1465.

IR (neat): 3404, 3059, 3024, 2915, 2852, 2357, 2227, 2045, 2014, 1960, 1721, 1609, 1584, 1496, 1467, 1445, 1412, 1379, 1333, 1286, 1269, 1255, 1079, 1044, 894, 853, 752, 737, 679, 643, 629, 609, 573, 555, 450, 437, and 418 cm^{-1}.

mp: 262-264 °C (with decomposition).

Data for 1,4-dihydroacridine 20a, the slower eluting major product:

^{1}H NMR (500 MHz, C_6D_6): δ 7.10 (m, 2H, *H2'* and *H6'*), 7.05 (dd, *J* = 7.8, 1.6 Hz, 1H, *H10*), 6.977 (ddd, *J* = 7.5, 7.5, 1.6 Hz, 2H, *H8*), 6.975 (nfodd, J_{app} = 7.4, 7.4 Hz, 2H, *H3'* and *H5'*), 6.89 (tt , *J* = 6.8, 1.3 Hz, 1H, *H4'*), 6.79 (ddd, *J* = 7.4, 7.4, 1.2 Hz, 1H, *H9*), 6.41 (dd, *J* = 8.0, 1.1 Hz, 1H, *H7*), 5.65 (s, 1H, *NH*), 4.85 (dddd, *J* = 13.3, 2.9, 0.9, 0.9 Hz, 1H, MsN*C3H$_a$*C3H$_b$), 4.81 (dd, *J* = 0.6, 0.6 Hz, 1H, *H11*), 4.57 (br dddd, *J* = 13.6, 3.0, 0.9, 0.9 Hz, 1H, MsN*C1H$_a$*C1H$_b$), 4.48 (ddd, *J* = 13.4, 3.1, 1.7 Hz, 1H, MsNC1H$_a$*C1H$_b$*), 4.18 (br d, *J* = 13.5 Hz, 1H, MsNC1H$_a$*C1H$_b$*), 2.14 (s, 3H, CH_3SO_2N), 1.94 (s, 3H, NArCH3), and 1.69 (s, 3H, C≡CCH_3).

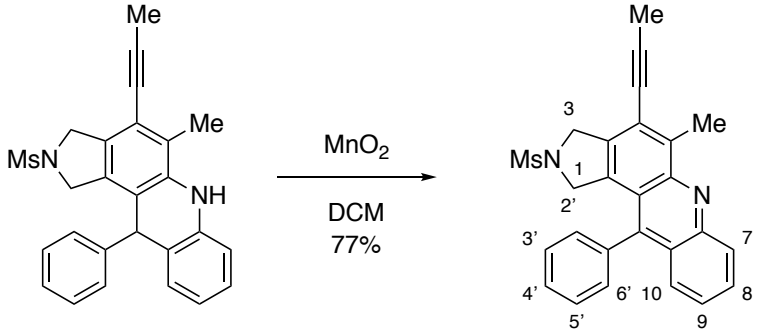
^{13}C NMR (126 MHz, $CDCl_3$): 146.2, 137.8, 137.2, 132.9, 130.7, 129.5, 129.0, 127.7, 127.3, 126.9, 123.1, 122.7, 121.6, 117.8, 117.4, 114.8, 93.9, 75.9, 54.5, 53.5, 45.9, 34.4, 14.8, and 4.7.

HRMS (ESI-TOF): Calcd for $C_{26}H_{25}N_2O_2S^+$ $[M+H^+]^+$ requires 429.1631; found 429.1621 (minor ion) and Calcd for $C_{26}H_{23}N_2O_2S^+$ $[M-2H]^+$ requires 427.1475; found 427.1469 (major ion).

IR (neat): 3052, 3033, 3006, 2924, 2873, 2852, 2241, 2222, 2161, 2019, 1715, 1622, 1601, 1560, 1541, 1526, 1492, 1475, 1452, 1445, 1415, 1379, 1355, 1332, 1308, 1294, 1265, 1192, 1156, 1088, 1032, 994, 976, 923, 911, 862, 841, 824, 790, 762, 749, 712, 674, 646, 611, 571, 563, 518, 496, 460, 423, and 415 cm^{-1}.

mp: 304-306 °C (with decomposition).

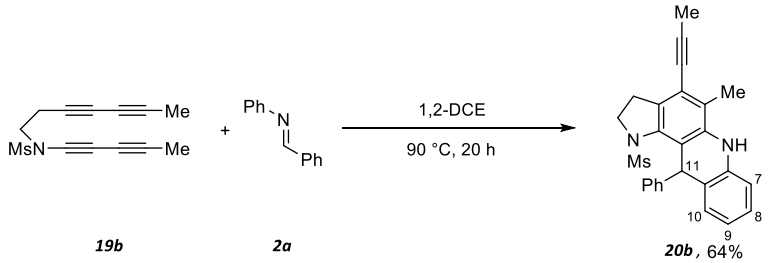
5-Methyl-2-(methylsulfonyl)-11-phenyl-4-(prop-1-yn-1-yl)-2,3-dihydro-1H-pyrrolo[3,4-a]acridine (21a):



20a ***21a***

The acridine **21a** was synthesized according to General Procedure B (40 mg of amine **20a**). Purification by MPLC (2:1 hexanes:EtOAc) afforded **21a** (30 mg, 77%) as a yellow crystalline solid (characterization data given above).

5-Methyl-1-(methylsulfonyl)-11-phenyl-4-(prop-1-yn-1-yl)-2,3,6,11-tetrahydro-1H-pyrrolo[2,3-a]acridine (20b)



Following general procedure A, *N*-(hepta-3,5-diyn-1-yl)-*N*-(penta-1,3-diyn-1-yl)methanesulfonamide (**19b**, 0.020 g, 0.081 mmol, 1 equiv), (*E)-N*,1-diphenylmethanimine (**2a**, 0.015 g, 0.088 mmol, 1.1 equiv), and 1,2-dichloroethane (2 mL) were used to prepare the 1,4-dihydroacridine **20b**. Purification of the crude product by MPLC (2:1 hexanes:EtOAc) yielded **20b** (0.023 g, 0.053 mmol, 64%) as a pale yellow amorphous solid.

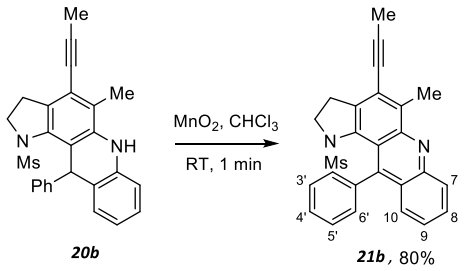
^{1}H NMR (500 MHz, $CDCl_3$): δ 7.30 (br d, *J* = 7.5 Hz, 1H, *H10*), 7.16 (ddd, *J* = 9.0, 7.9, 1.5 Hz, 1H, *H8*), 7.10 (br dd, *J* = 7.6, 7.6 Hz, 2H, PhH_m), 7.05–7.02 (overlapping m's, 3H, PhH_o and PhH_p), 6.95 (ddd, *J* = 9.0, 7.5, 1.2 Hz, 1H, *H9*), 6.87 (br d, *J* = 7.9 Hz, 1H, *H7*), 6.36 (s, 1H, *H11*), 6.22 (br s, 1H, N*H*), 4.29 (dd, *J* = 13.0, 7.8 Hz, 1H, MsNCH_aH$_b$CH$_2$), 3.82 (ddd , *J* = 12.9, 12.9, 8.3 Hz, 1H, MsNCH$_a$$H_bCH_2$), 3.21 (ddd, *J* = 16.2, 12.0, 7.8, 1H, MsNCH$_2$CH_aH$_b$), 2.84 (dd, *J* = 16.2, 8.2 Hz, 1H, MsNCH$_2$CH$_a$$H_b$), 2.72 (s, 3H, C$H_3SO_2$N), 2.40 (s, 3H, ArC$H_3$), and 2.11 (s, 3H, C≡CC$H_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 145.5, 139.0, 138.5, 138.3, 130.8, 129.6, 128.3, 127.4, 127.1, 126.2, 123.7, 122.0, 121.3, 119.4, 117.5, 114.3, 93.1, 76.6, 53.5, 42.6, 36.1, 29.8, 14.9, and 4.7.

HRMS (ESI-TOF): Calcd for $C_{26}H_{25}N_2O_2S^+$ $[M+H^+]^+$ requires 429.1631; found 429.1613.

IR (neat): 3407, 3057, 3024, 3004, 2955, 2915, 2852, 2342, 2235, 2142, 2000, 1609, 1598, 1577, 1494, 1465, 1434, 1381, 1339, 1328, 1295, 1279, 1252, 1241, 1198, 1154, 1121, 1076, 1045, 1031, 1014, 964, 937, 901, 885, 850, 804, 752, 734, 699, 664, 647, 617, 592, 561, 542, 513, 460, 445, 423, and 405 cm^{-1}.

5-Methyl-1-(methylsulfonyl)-11-phenyl-4-(prop-1-yn-1-yl)-2,3-dihydro-1H-pyrrolo[2,3-a]acridine (21b)



20b *21b*, 80%

Following general procedure B, 5-methyl-1-(methylsulfonyl)-11-phenyl-4-(prop-1-yn-1-yl)-2,3,6,11-tetrahydro-1H-pyrrolo[2,3-a]acridine (**20b**, 0.024 g, 0.057 mmol, 1 equiv), MnO_2 (xs) and $CHCl_3$ (10 mL) were used to prepare acridine **21b**. Purification of the crude product yielded acridine **21b** (0.019 g, 0.045 mmol, 80%) as a light green amorphous solid.

^{1}H NMR (500 MHz, $CDCl_3$): δ 8.23 (dd, *J* = 8.7, 0.6 Hz, 1H, *H7*), 7.88 (br d, *J* = 7.6 Hz, 1H, *H2'* or *H6'),* 7.80 (dd, *J* = 8.8, 0.7 Hz, 1H, *H10*), 7.69 (ddd, *J* = 8.5, 6.5, 1.4 Hz, 1H, *H8*), 7.59 (ddd, *J* = 7.5, 7.5, 1.0 Hz, 1H, *H3'* or *H5'*), 7.47 (dddd, *J* = 7.5, 7.5, 1.4, 1.4 Hz, 1H, *H4'*), 7.39 (ddd, *J* = 8.5, 7.7, 0.9 Hz, 1H, *H9*), 7.38 (dd, *J* = 7.6, 7.6, 1.4 Hz, 1H, *H3'* or *H5'*), 7.11 (ddd, *J* = 7.8, 1.4, 1.4 Hz, 1H, *H2'* or *H6'*), 3.99 (dd, *J* = 12.0, 7.0 Hz, 1H, $MsNC\mathit{H}_aH_bCH_2$), 3.70 (ddd, *J* = 11.9, 11.9, 7.9 Hz, 1H, $MsNCH_a\mathit{H}_bCH_2$), 3.57 (ddd, *J* = 16.2, 11.7, 7.0 Hz, 1H, $MsNCH_2C\mathit{H}_aH_b$), 3.09 (s, 3H, $C\mathit{H}_3SO_2N$), 3.01 (dd, *J* = 16.2, 7.8 Hz, 1H, $MsNCH_2CH_a\mathit{H}_b$), 2.21 (overlapping s, 3H, $ArC\mathit{H}_3$), 2.21 (overlapping s, 3H, $C{\equiv}CC\mathit{H}_3$).

^{13}C NMR (126 MHz, $CDCl_3$): 148.04, 148.00, 144.7, 140.8, 137.8, 137.0, 133.9, 133.1, 132.1, 130.5, 129.5, 127.7, 127.4, 126.6, 126.5, 126.2, 125.6, 120.9, 119.4, 95.8, 77.1, 52.0, 37.8, 32.0, 17.0, and 5.0.

HRMS (ESI-TOF): Calcd for $C_{26}H_{23}N_2O_2S^+$ $[M+H^+]^+$ requires 427.1475; found 427.1468.

IR (neat): 3055, 3031, 2955, 2917, 2850, 2233, 1730, 1619, 1593, 1548, 1442, 1413, 1353, 1321, 1263, 1150, 1103, 1015, 963, 921, 848, 803, 763, 736, 701, 670, 650, 623, 606, 536, 511, and 470 cm^{-1}.

Supporting Information References

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[3] S. Arora, J. Zhang, V. Pogula, T. R. Hoye, *Chem. Sci.* **2019**, *10*, 9069–9076.

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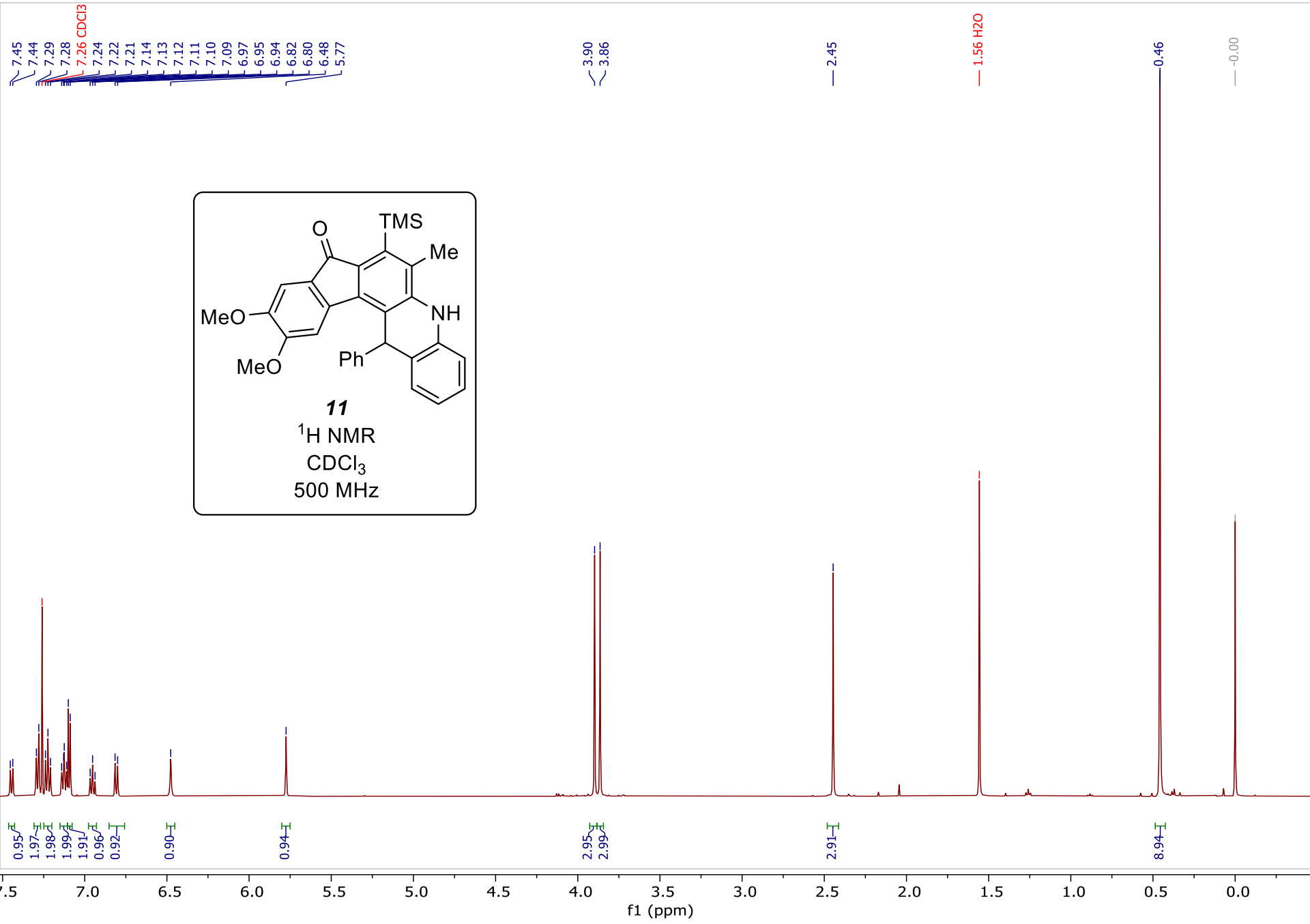
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6.95
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3.86
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Ph
11
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500 MHz
0.95
1.97
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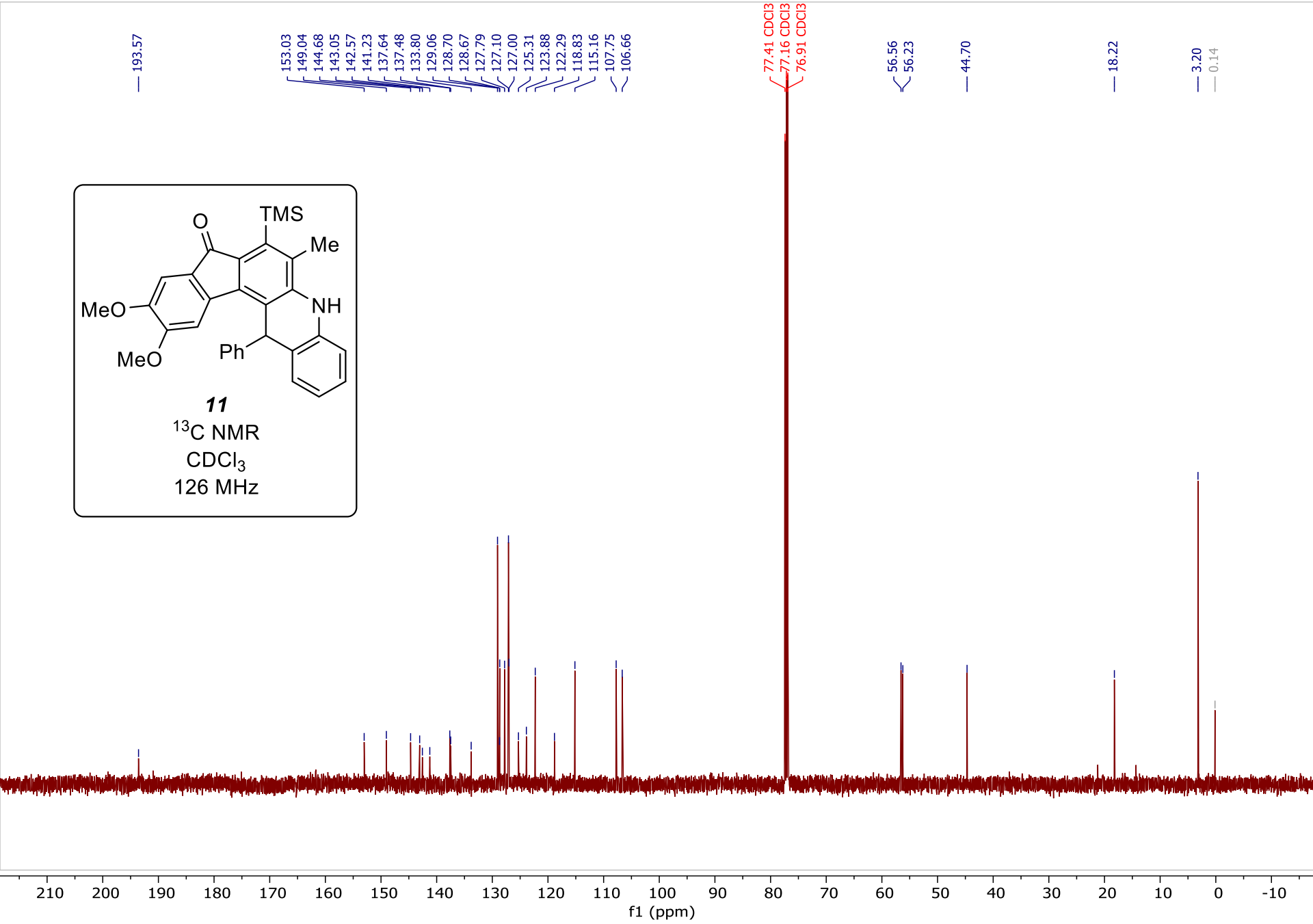
f1 (ppm)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56.56
56.23
44.70
18.22
3.20
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Me
MeO
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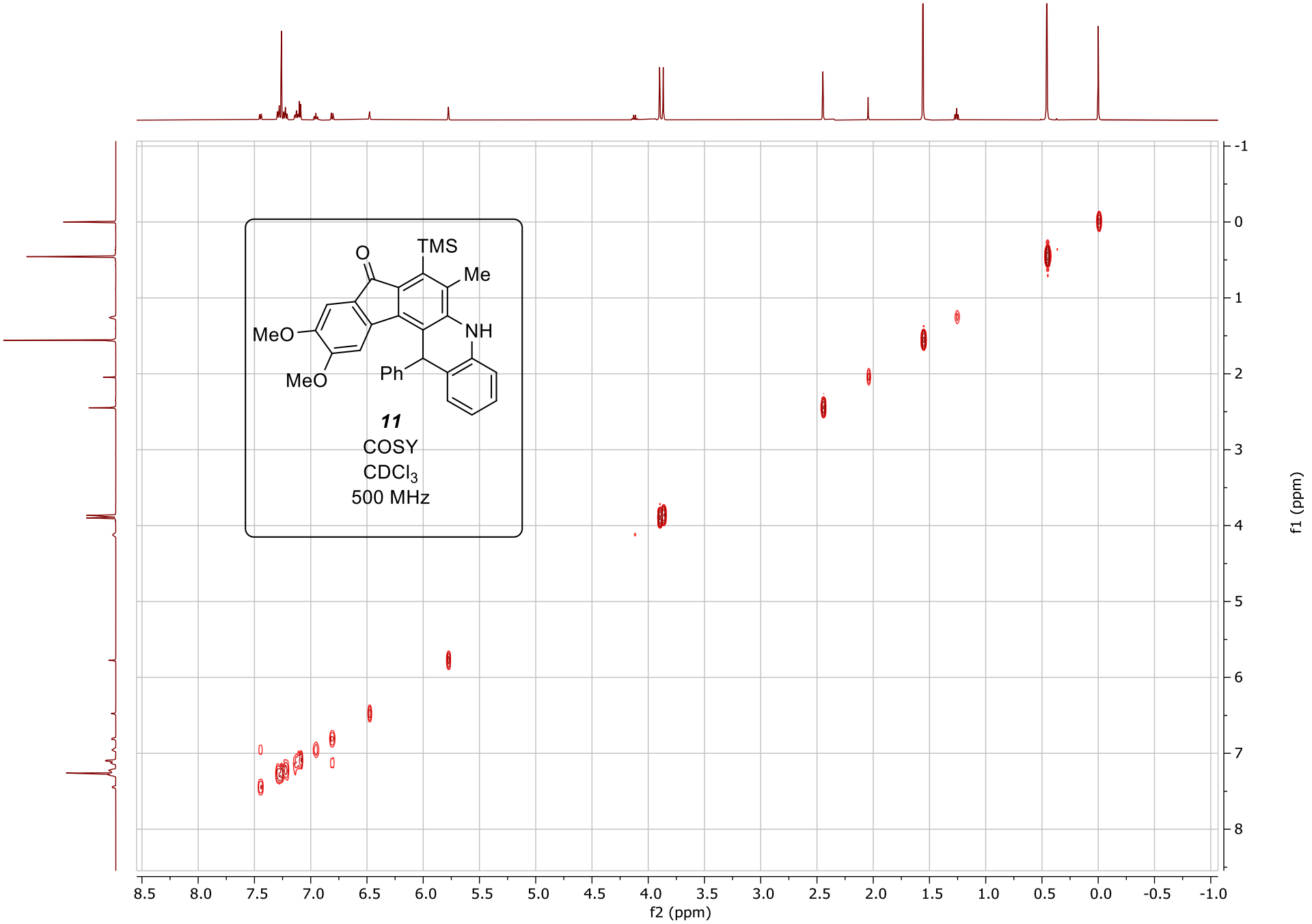
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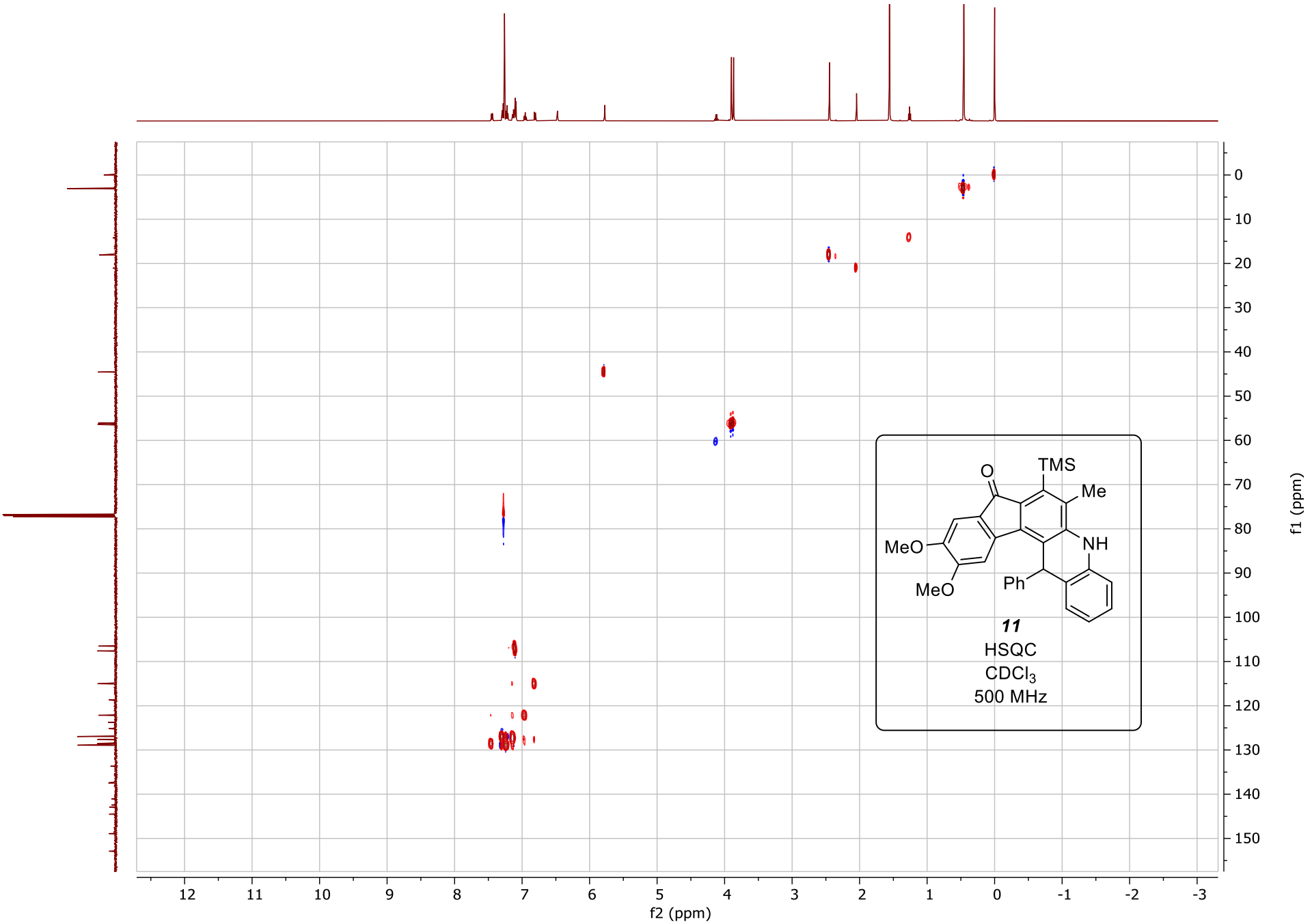
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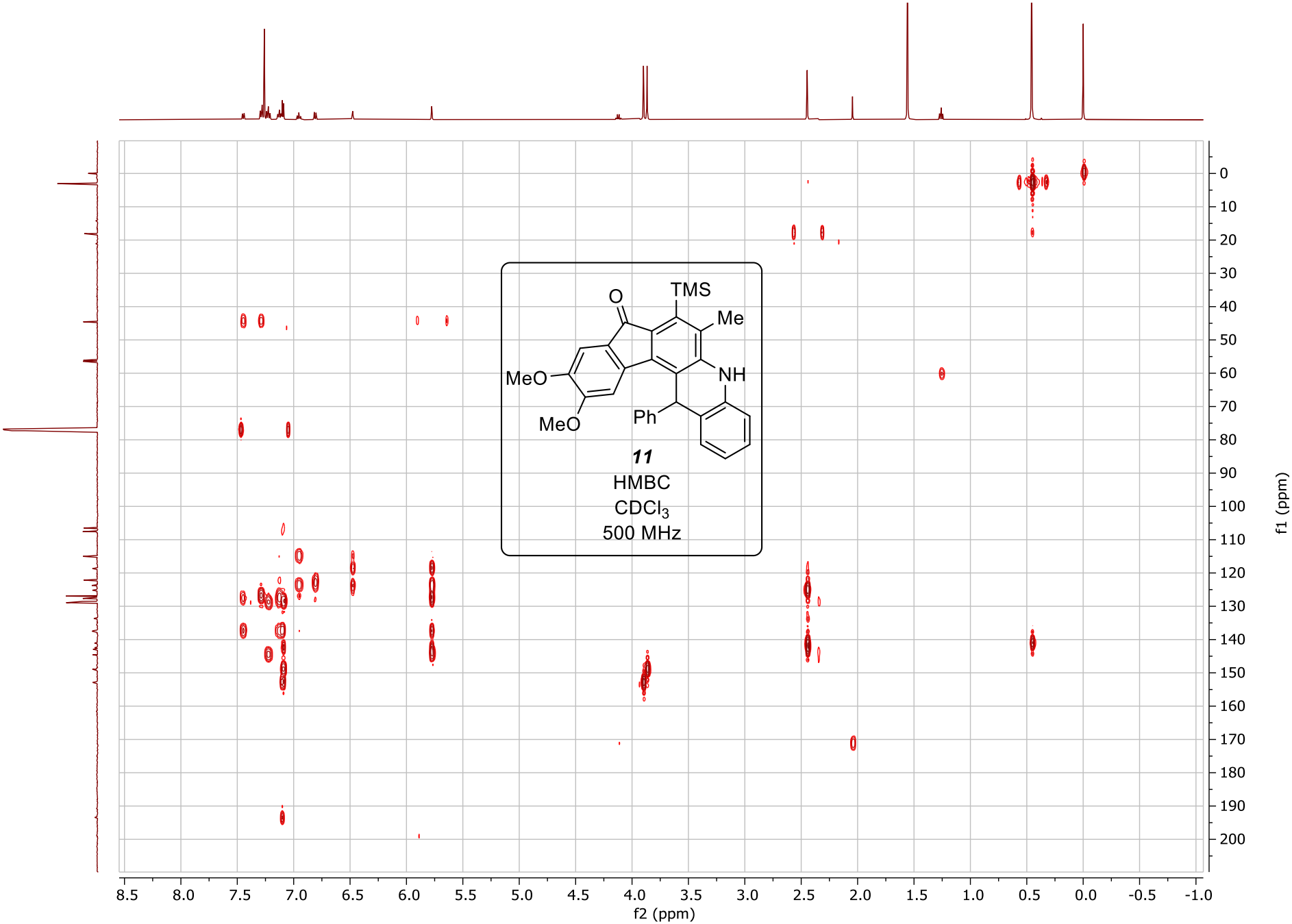
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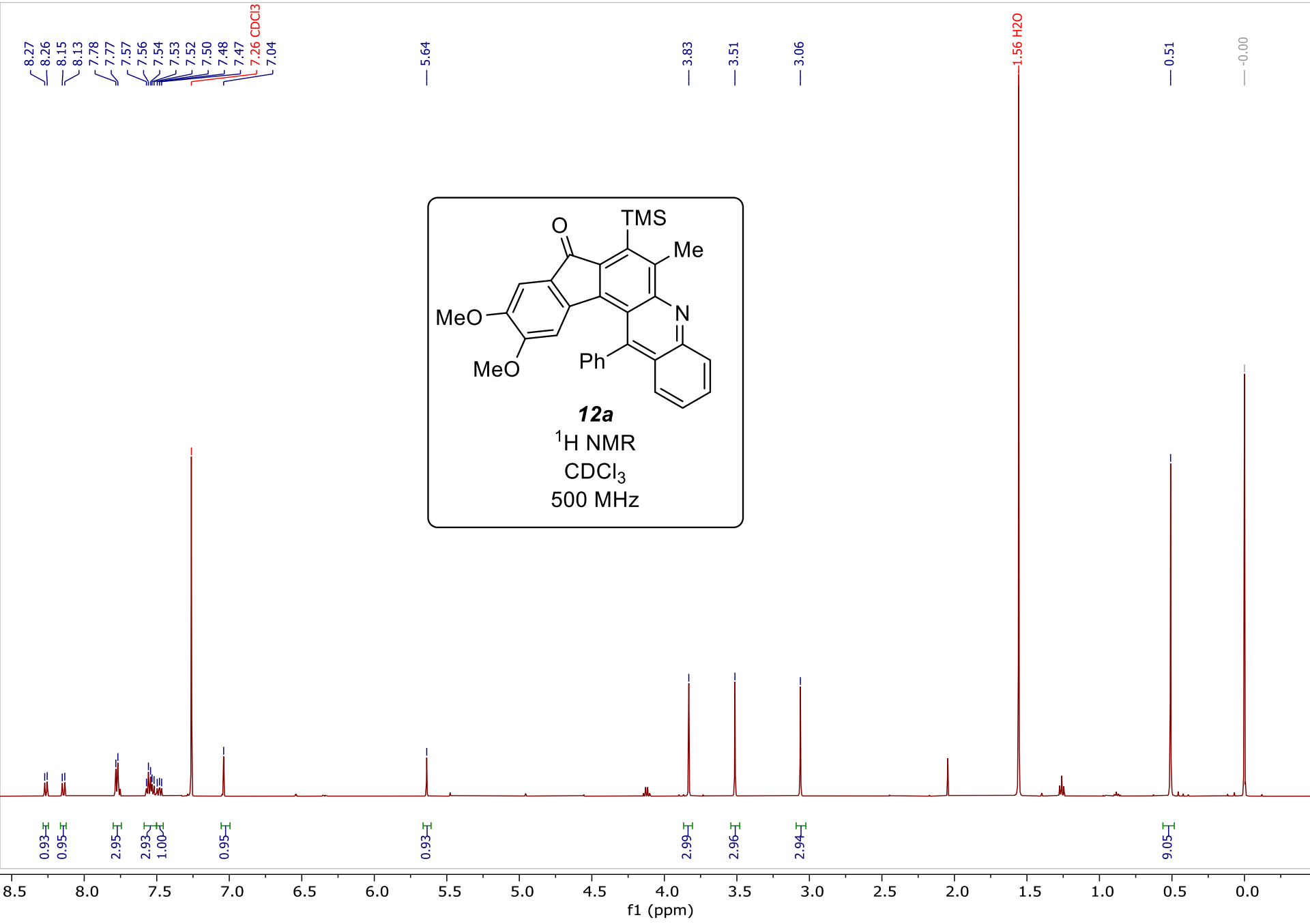
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N
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Ph
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7.0
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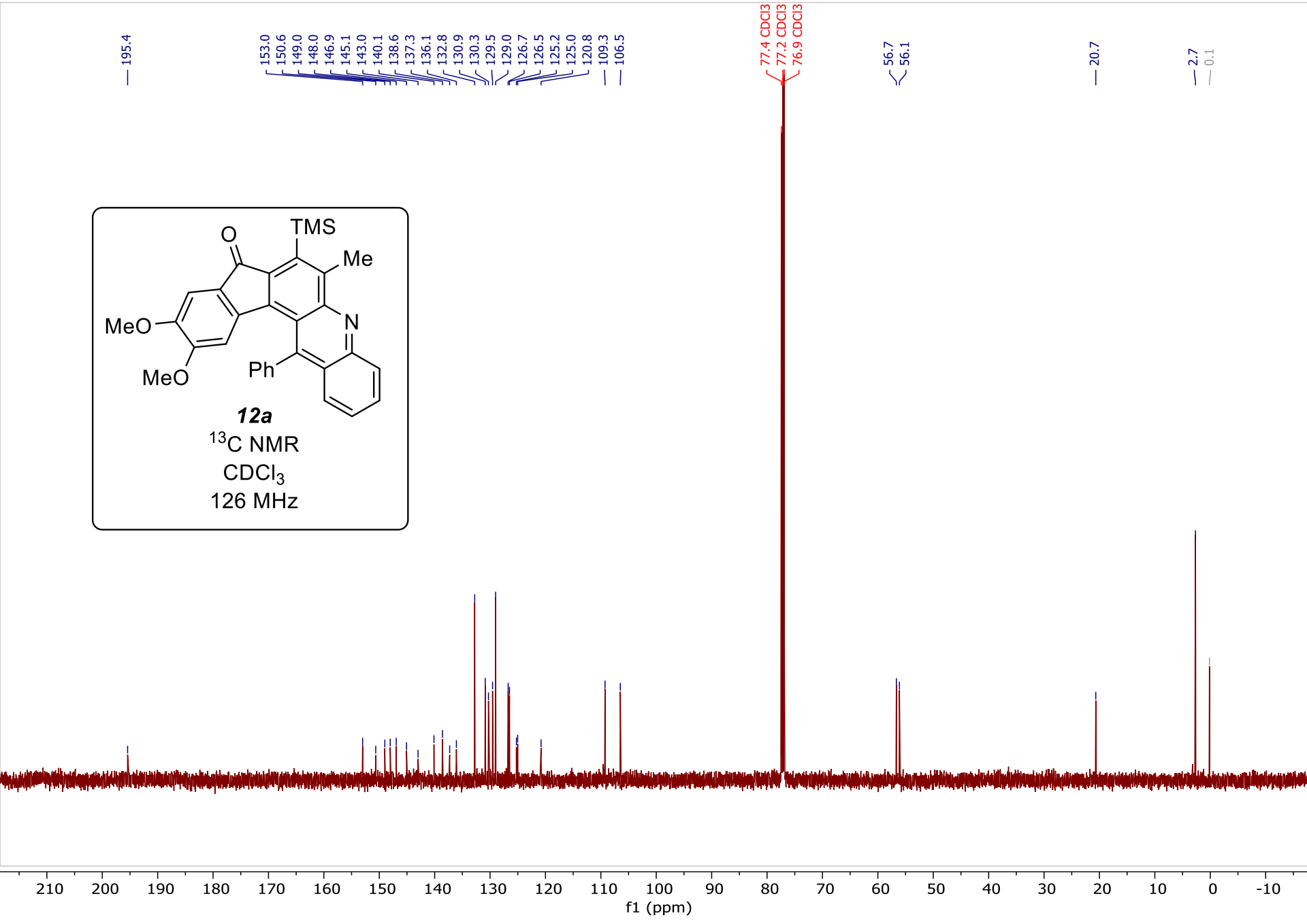
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77.2 CDCl3
76.9 CDCl3
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56.1
20.7
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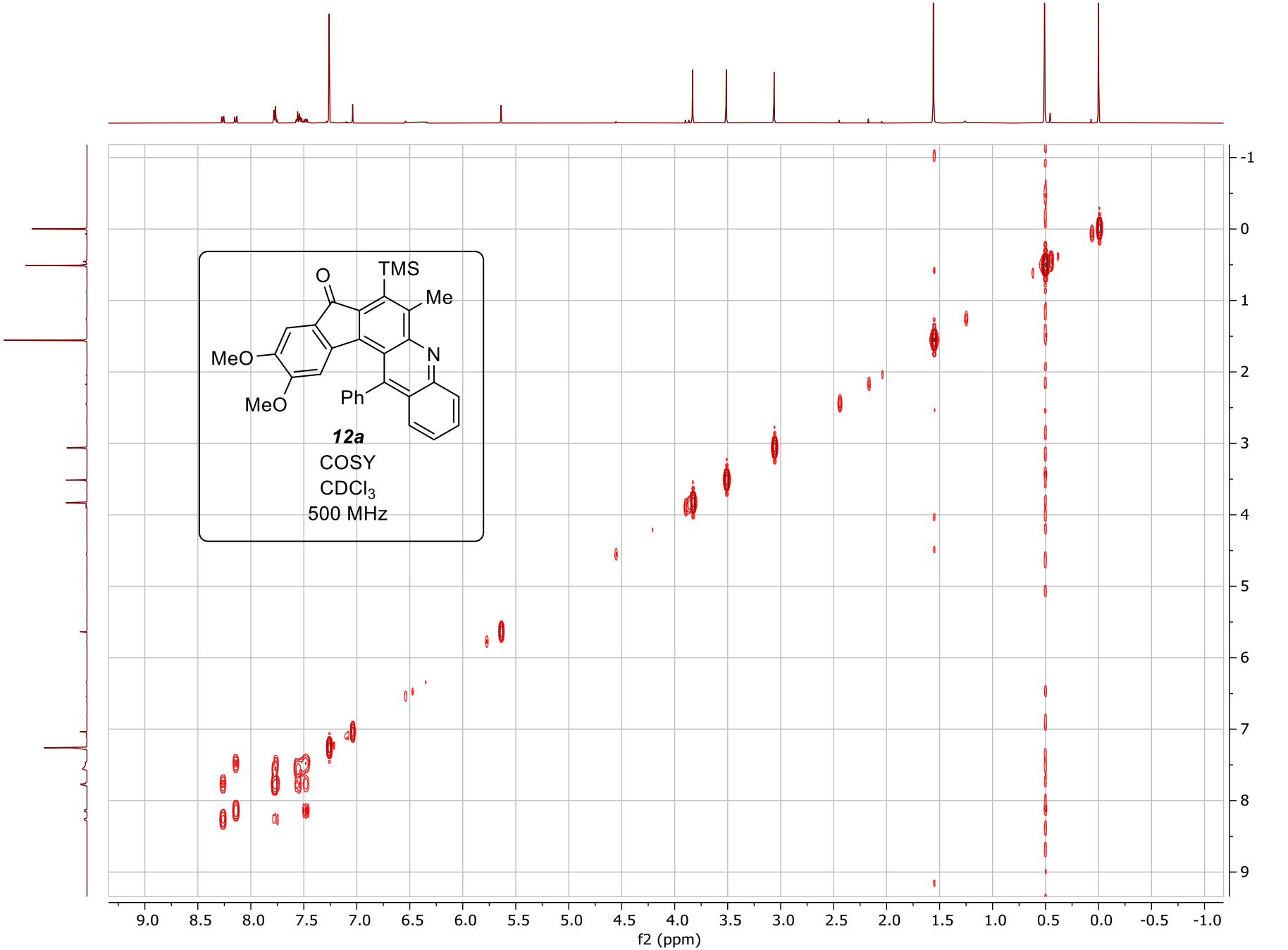
126 MHz
f1 (ppm)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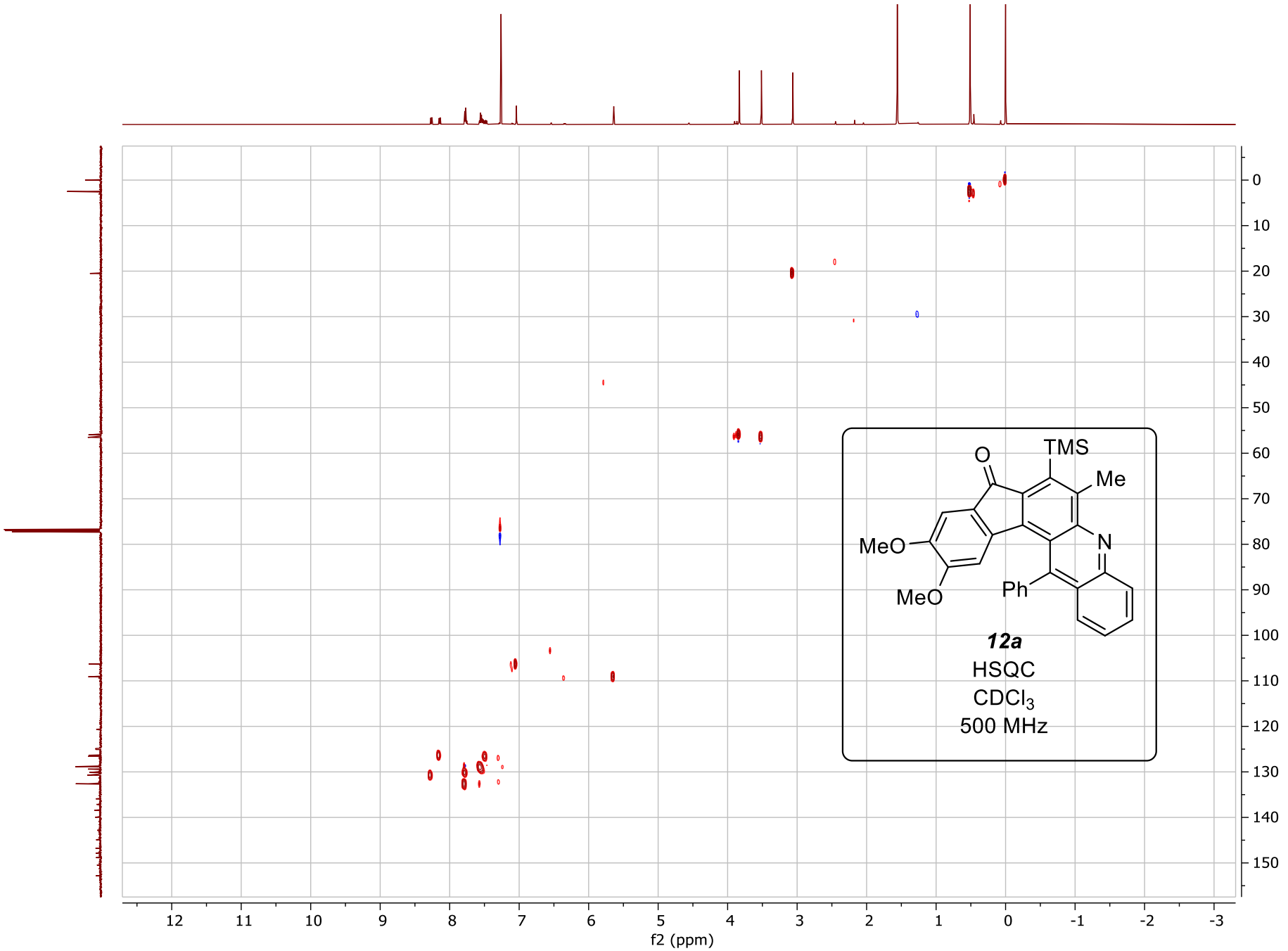
f1 (ppm)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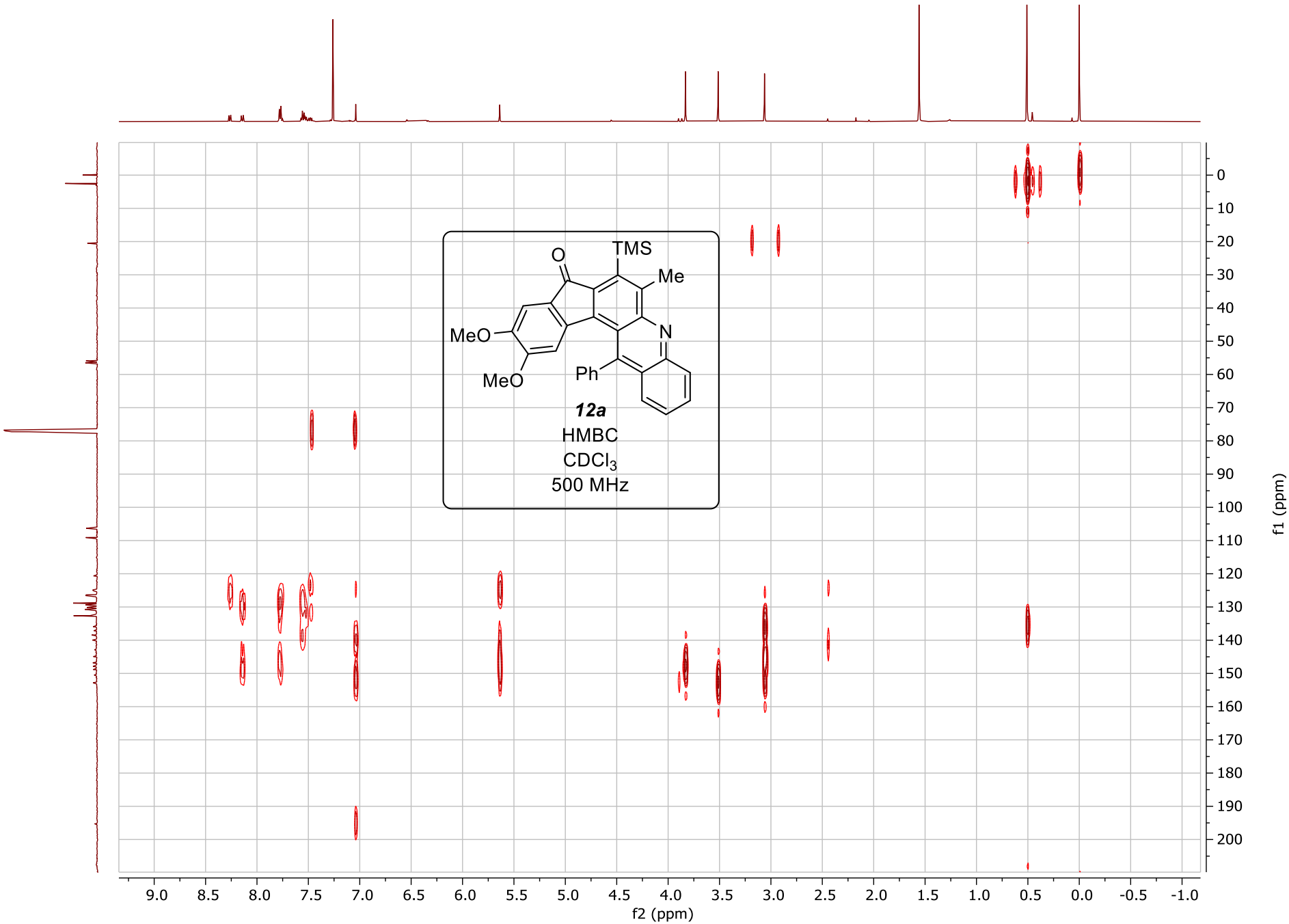
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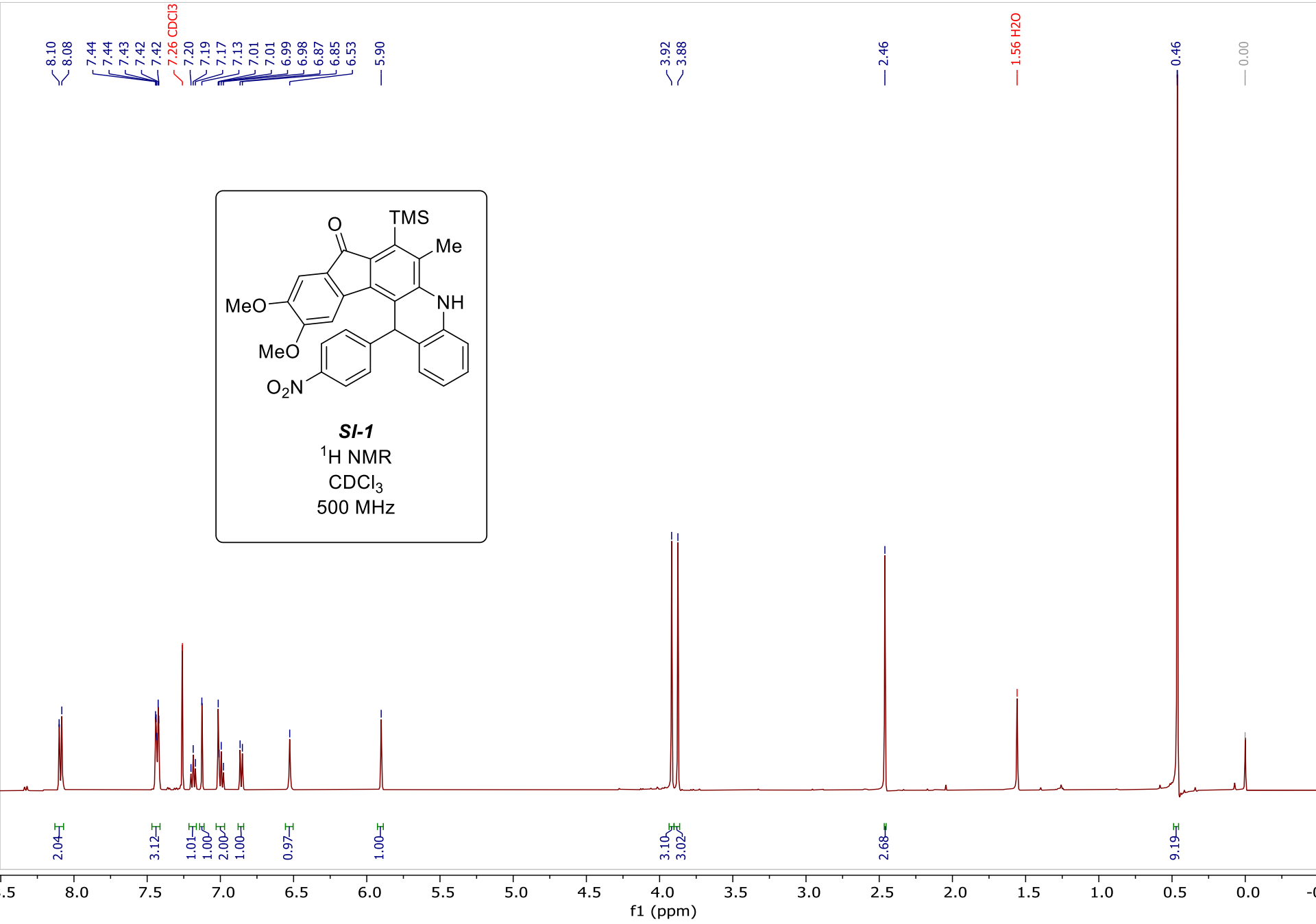
f1 (ppm)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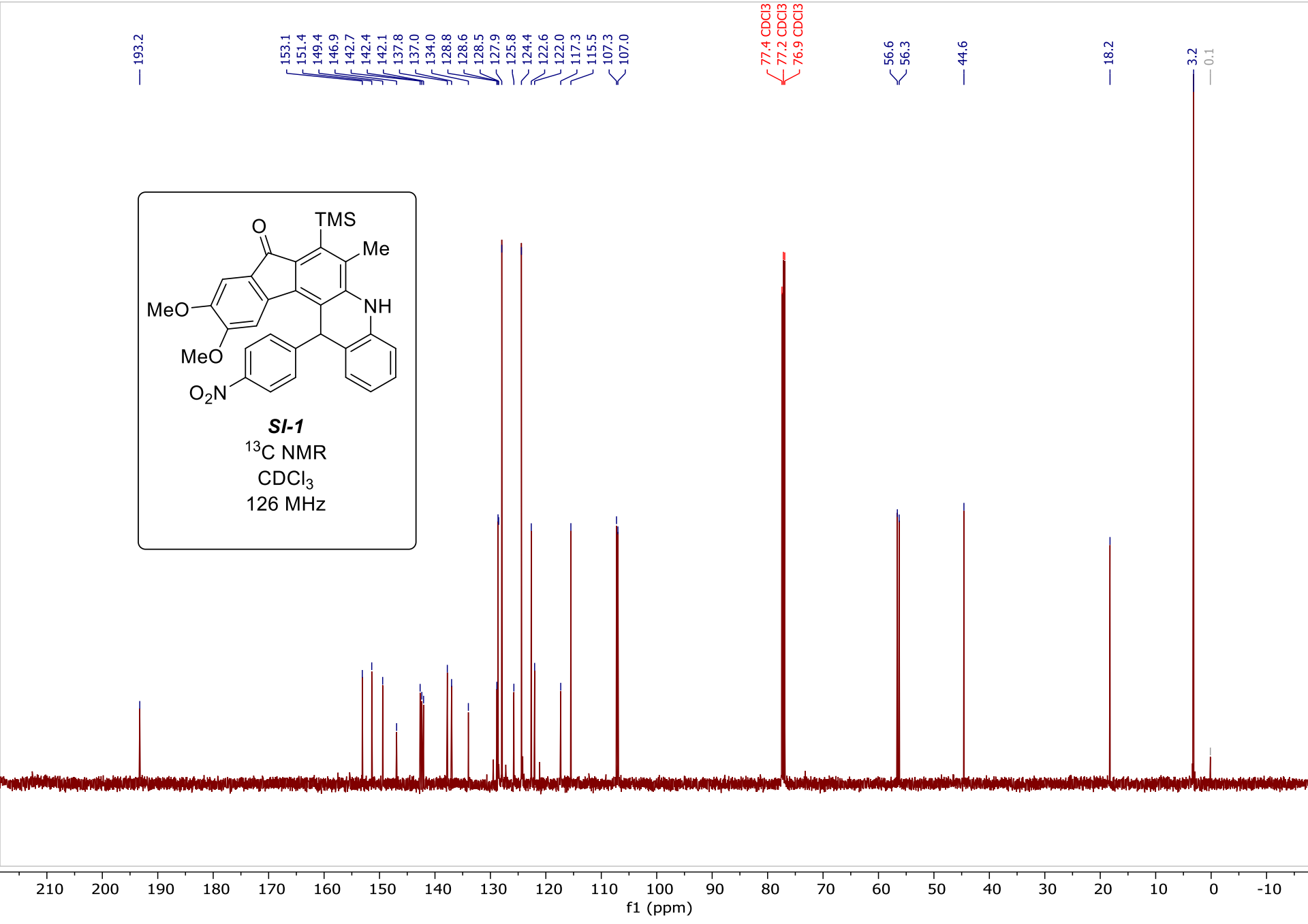
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77.2 CDCl3
76.9 CDCl3
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56.3
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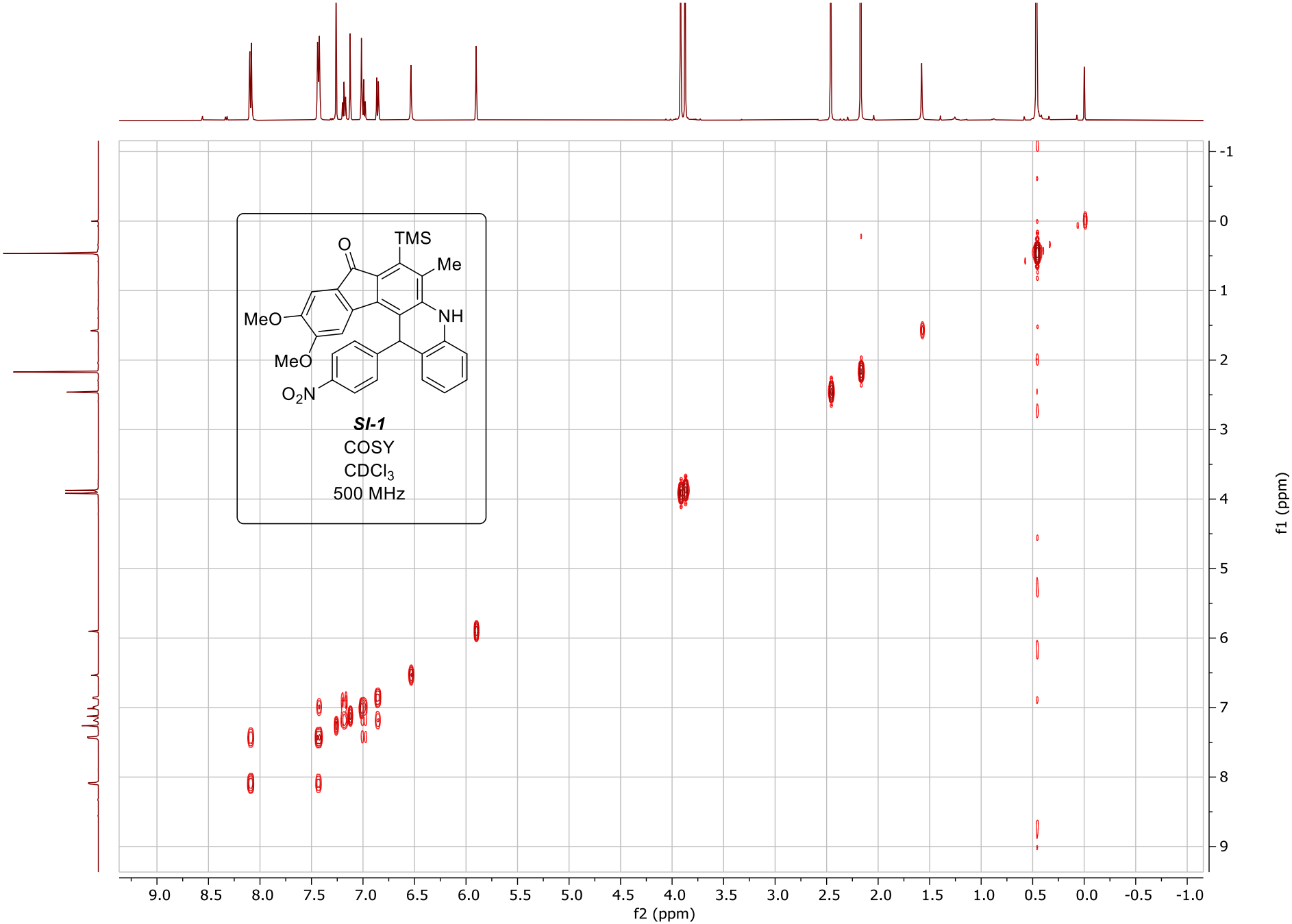
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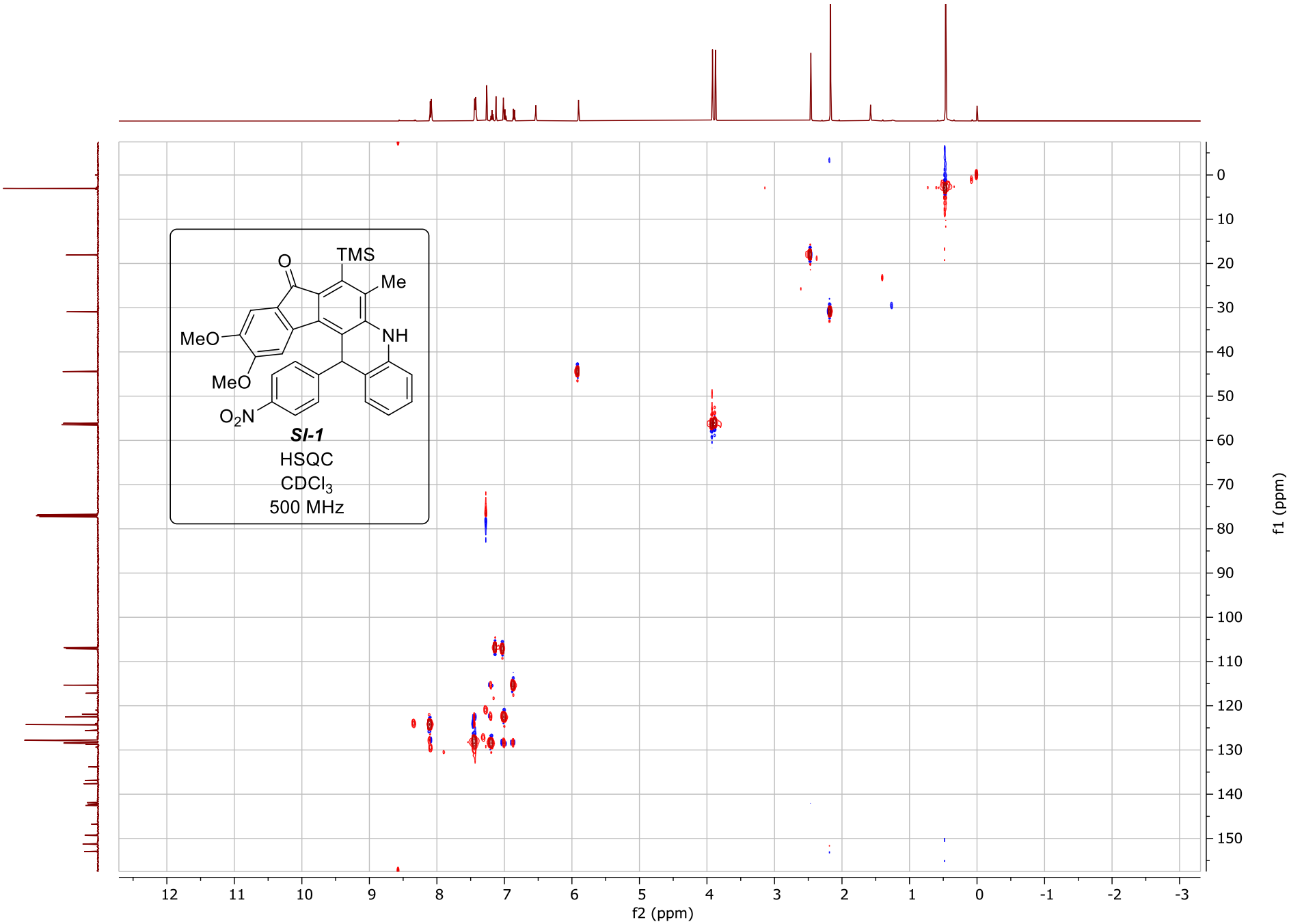
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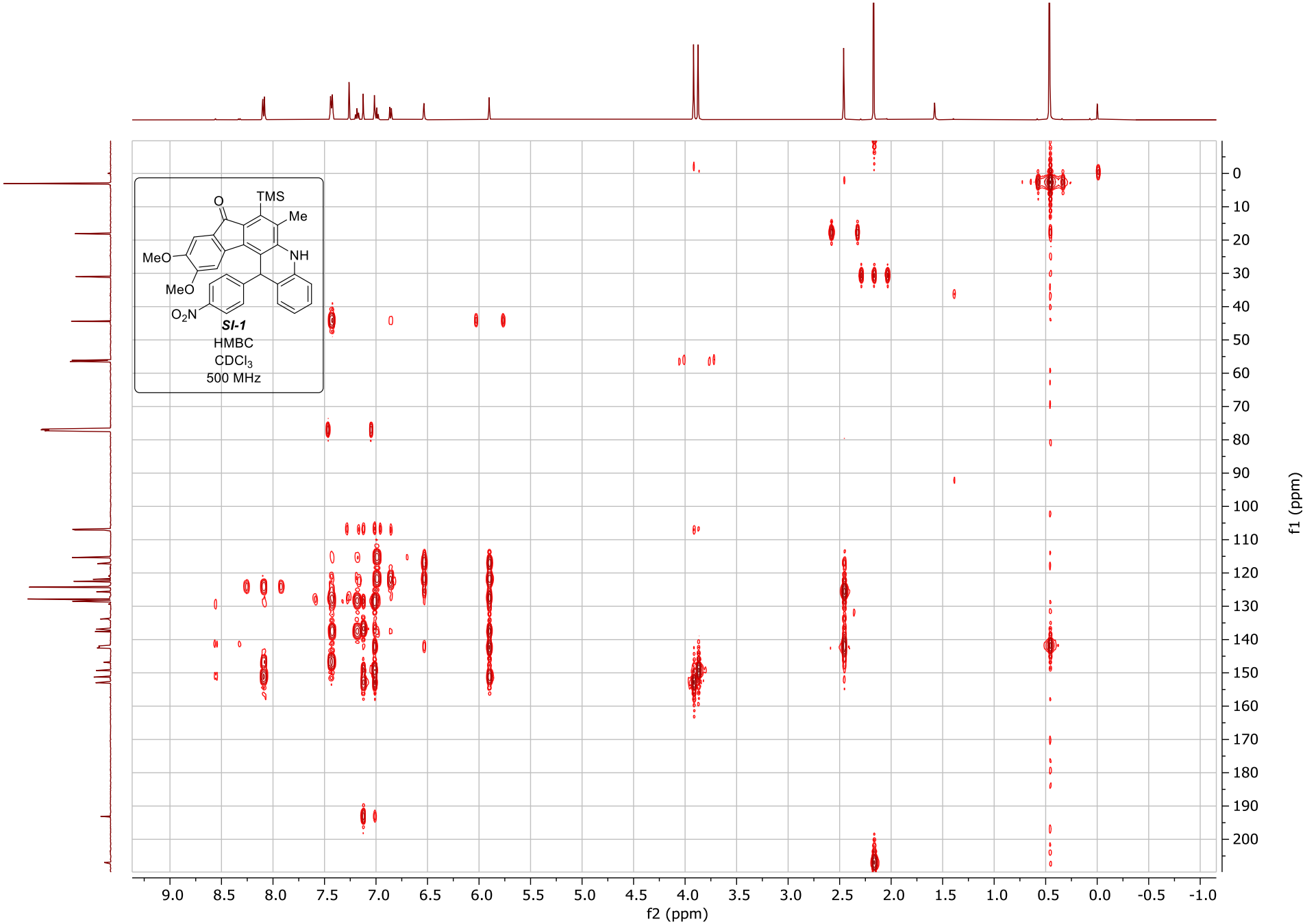
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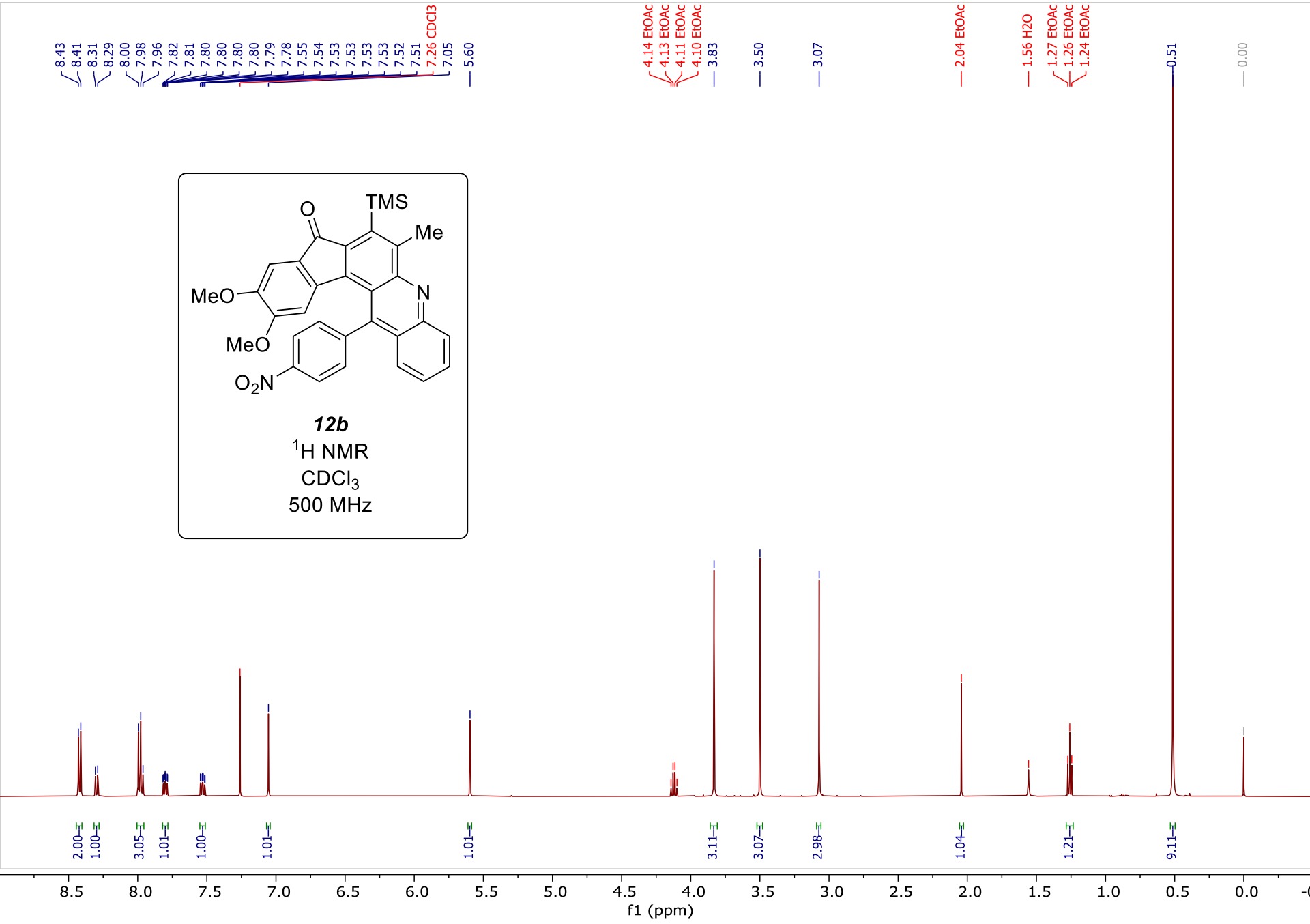
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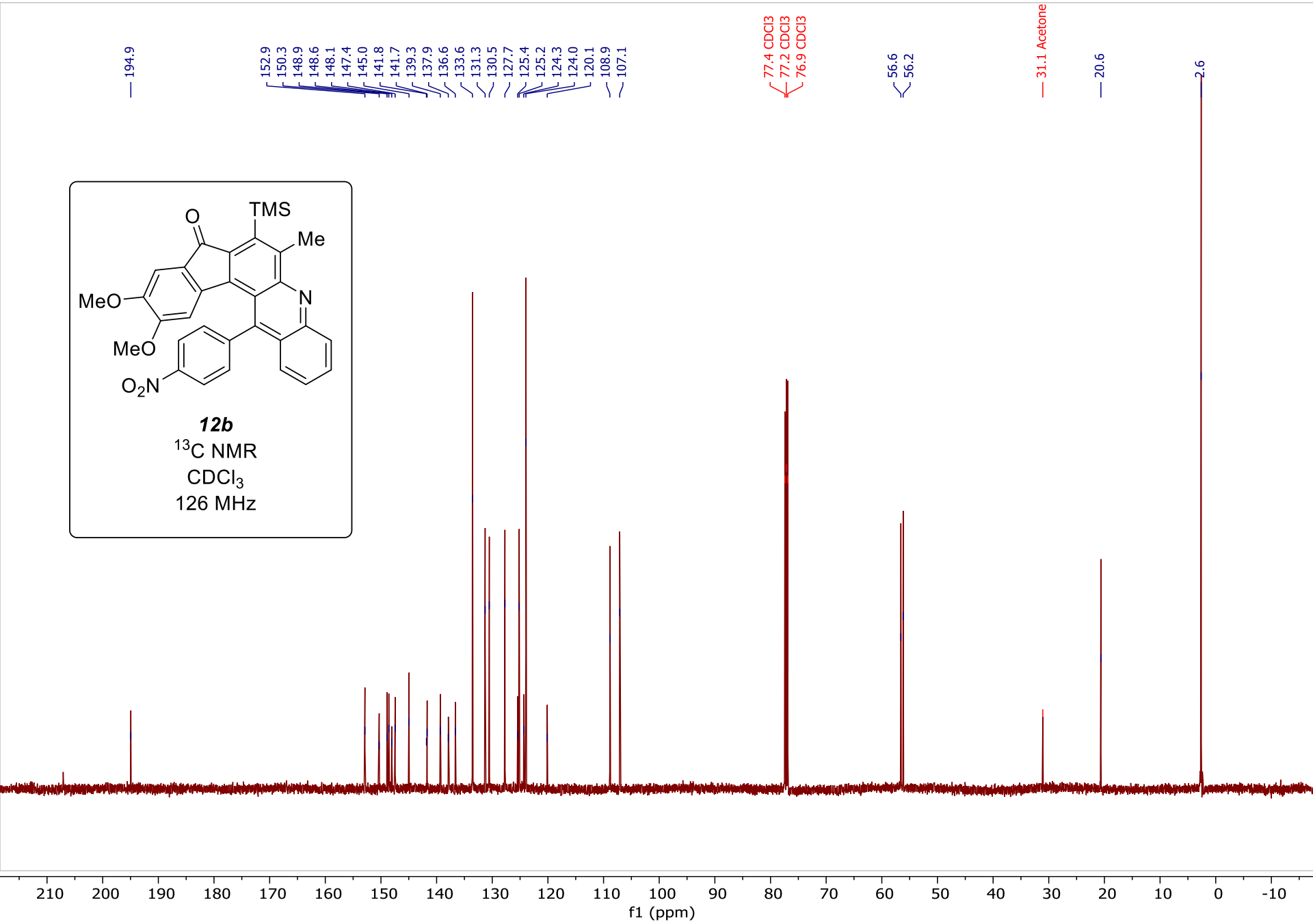
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77.2 CDCl3
76.9 CDCl3
56.6
56.2
31.1 Acetone
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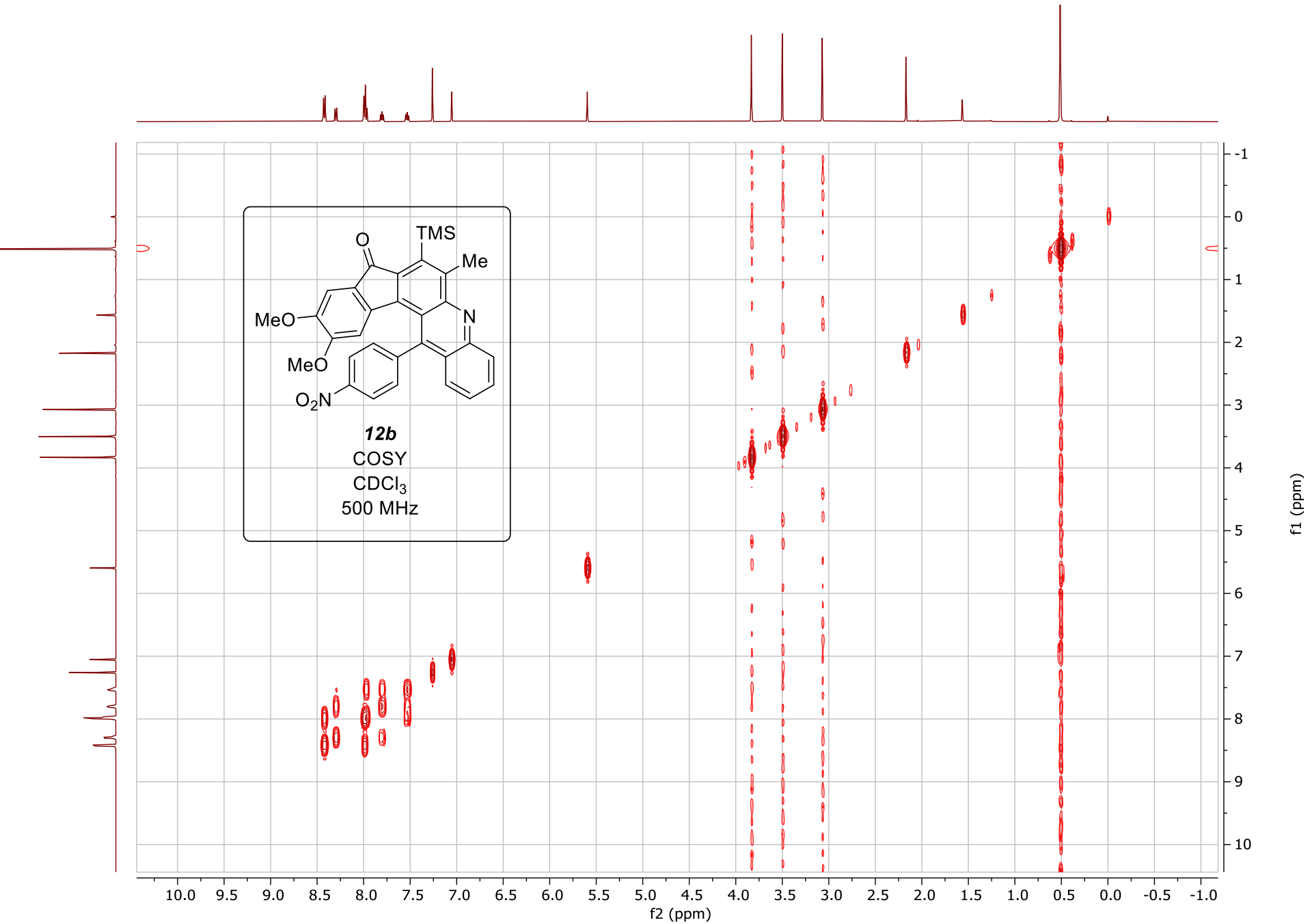
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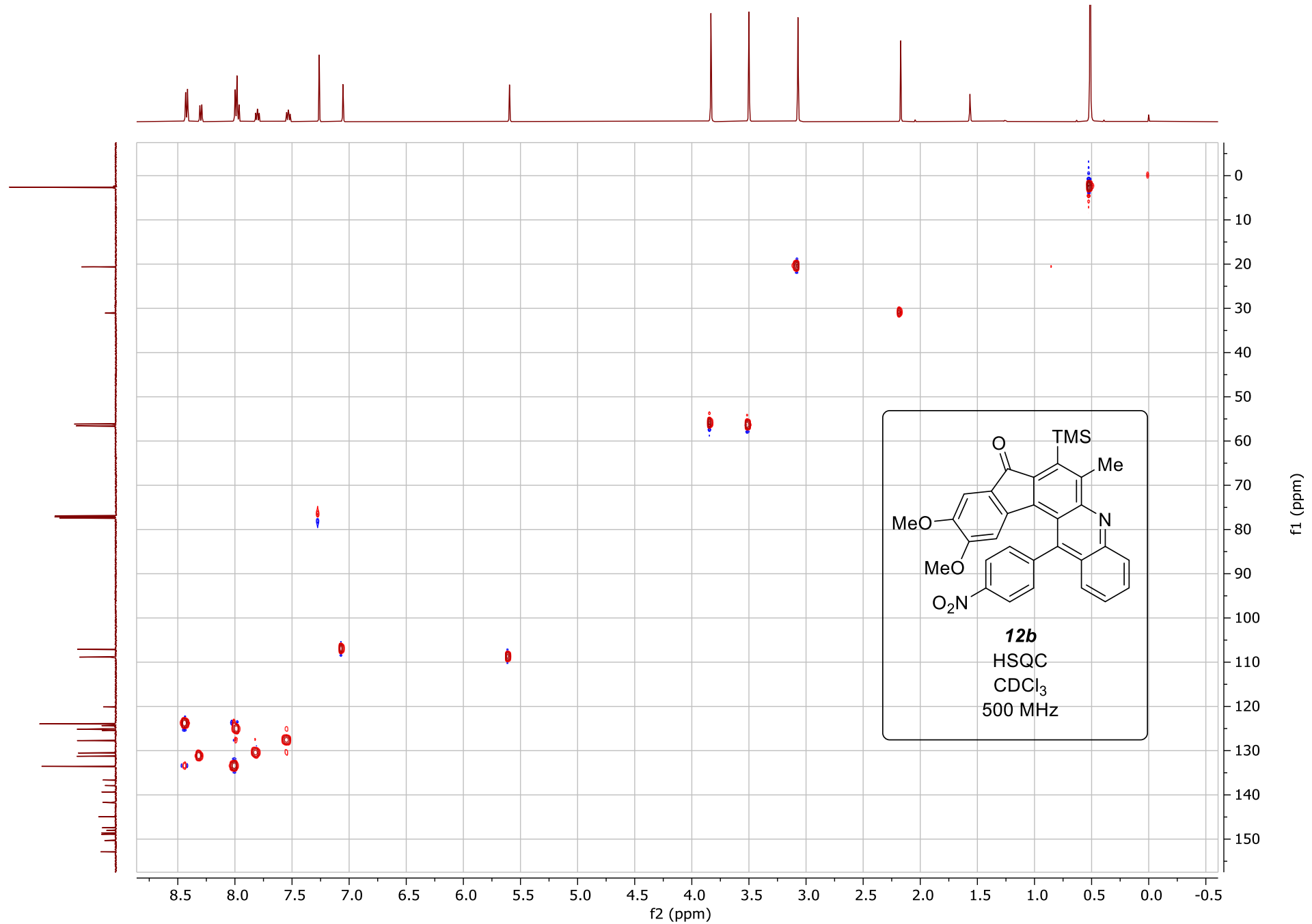
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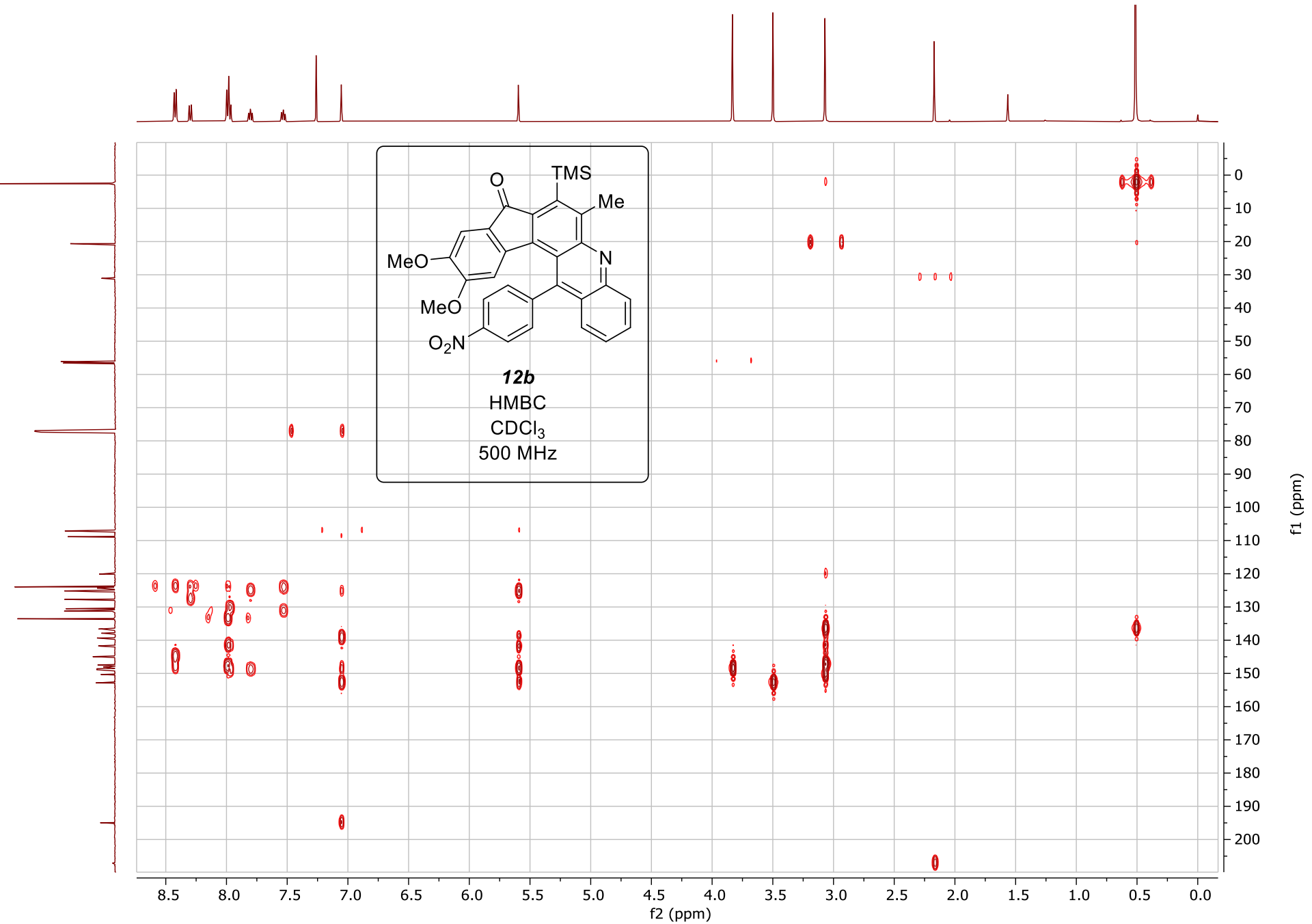
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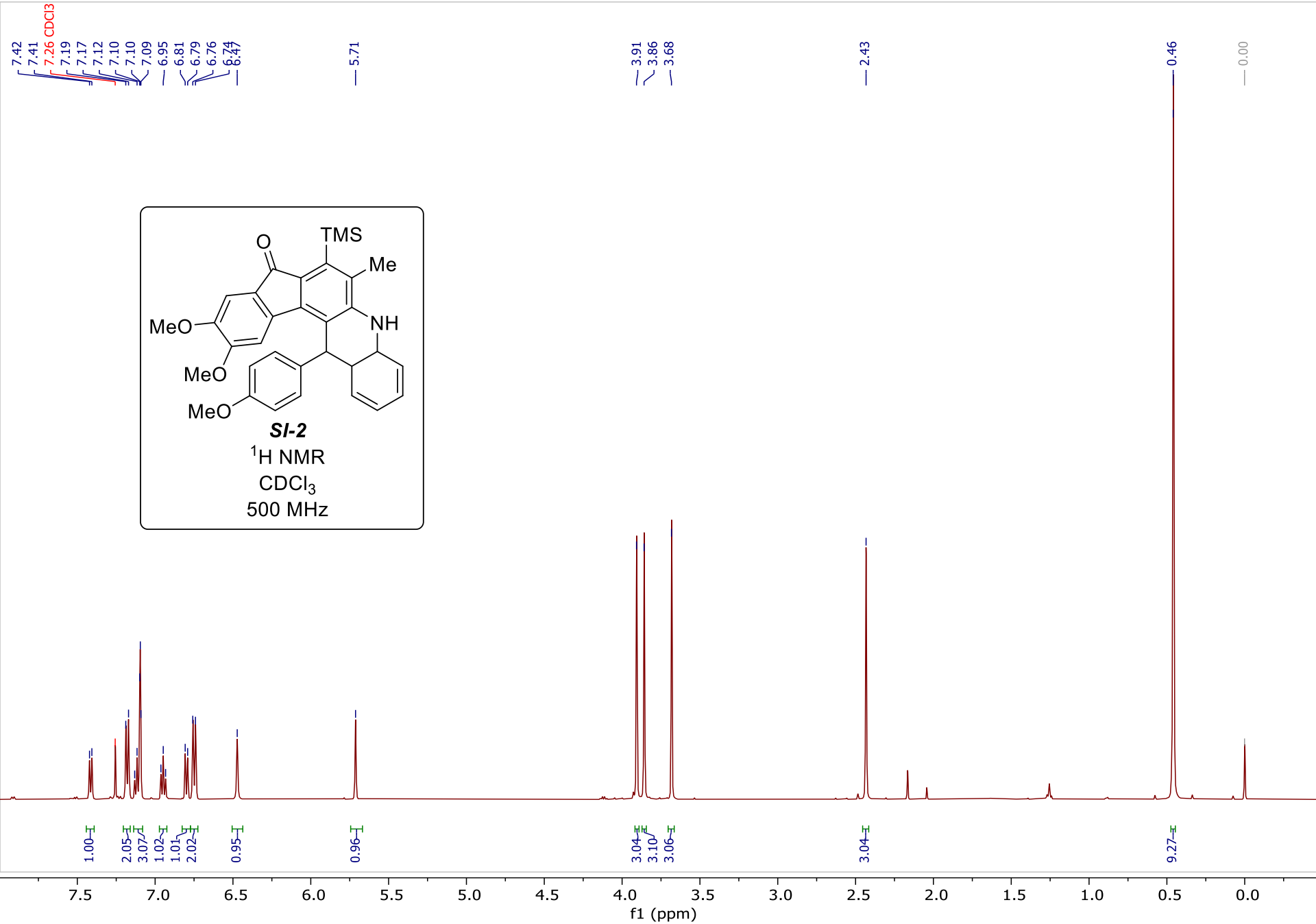
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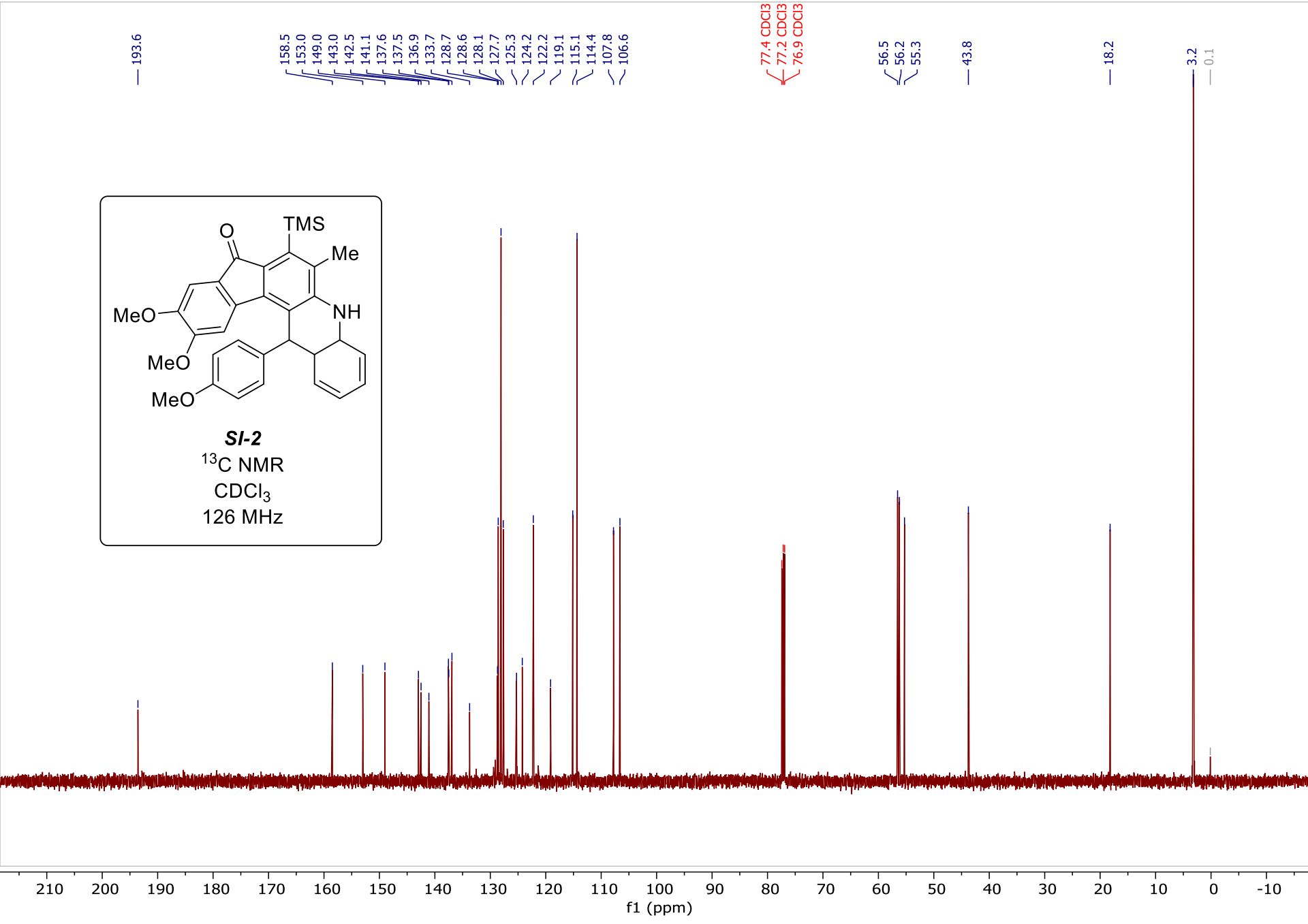
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77.2 CDCl3
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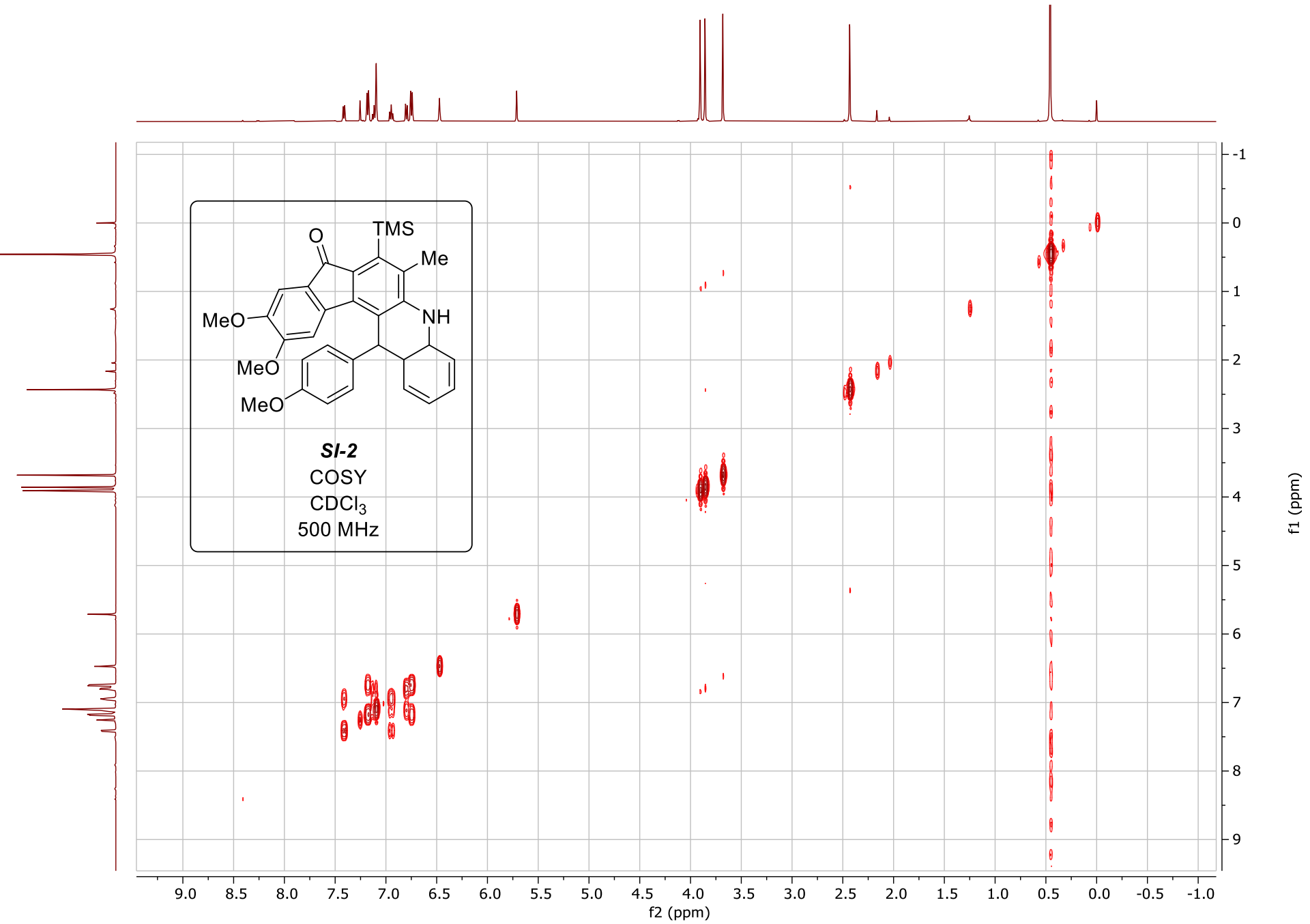
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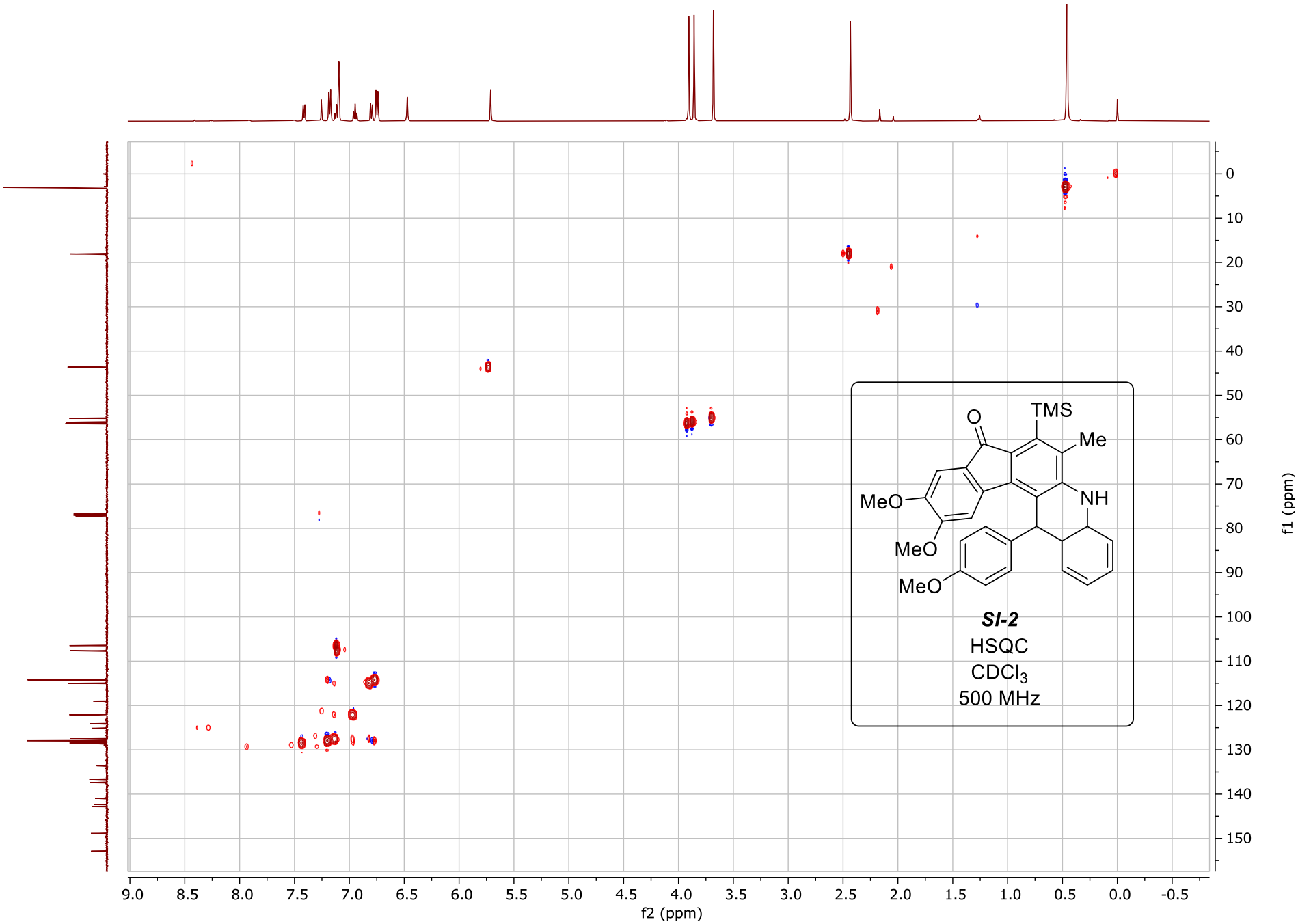
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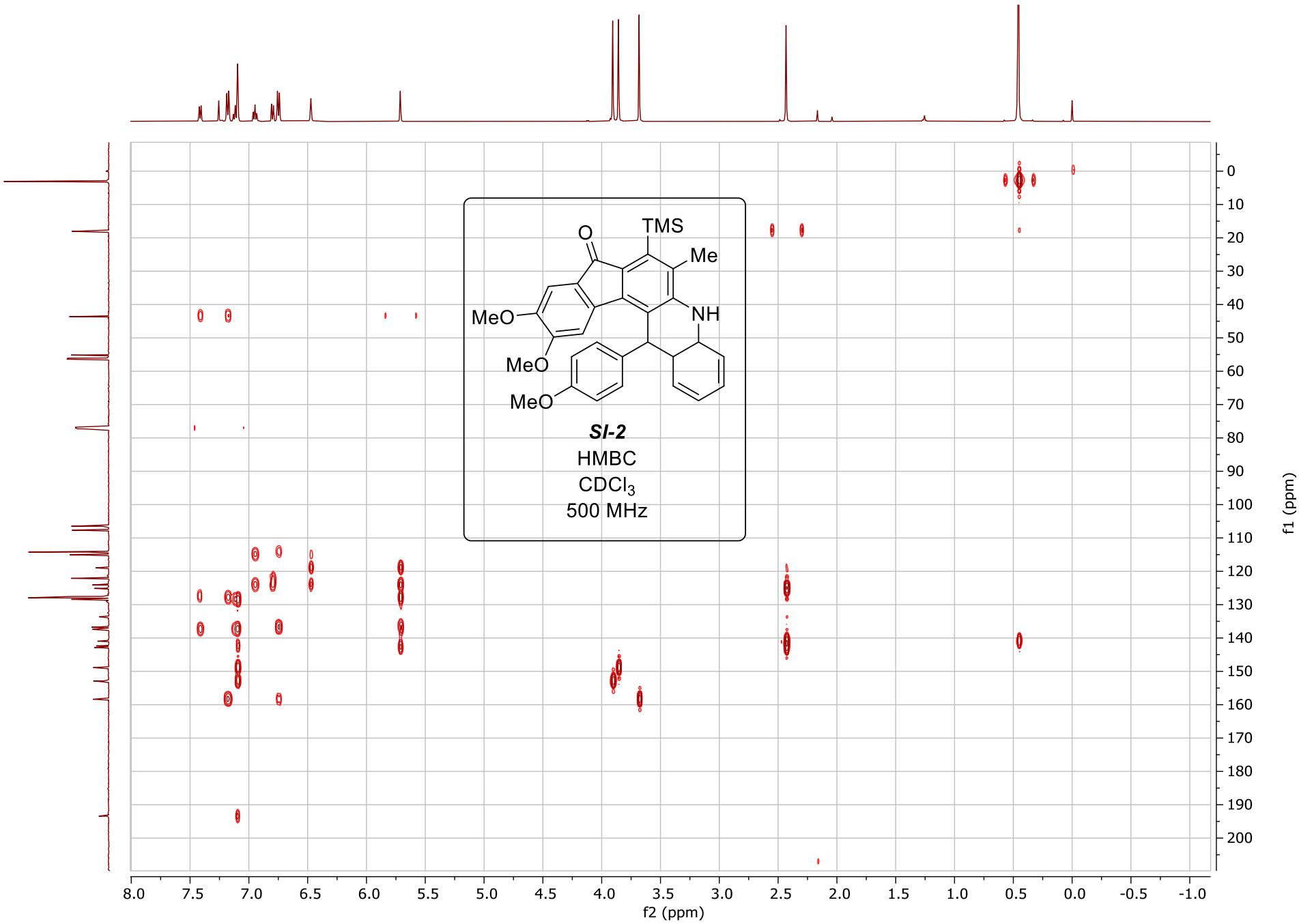
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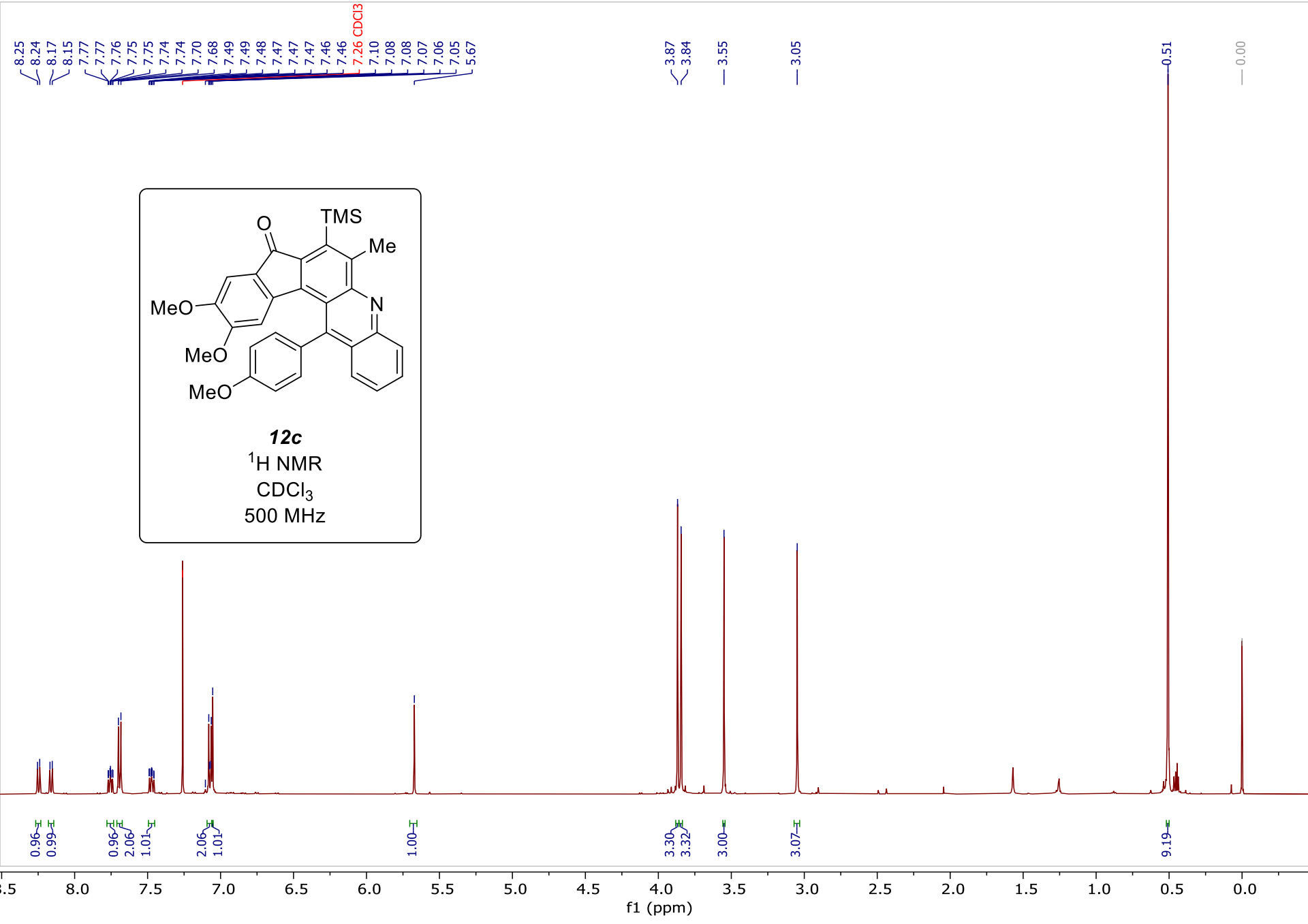
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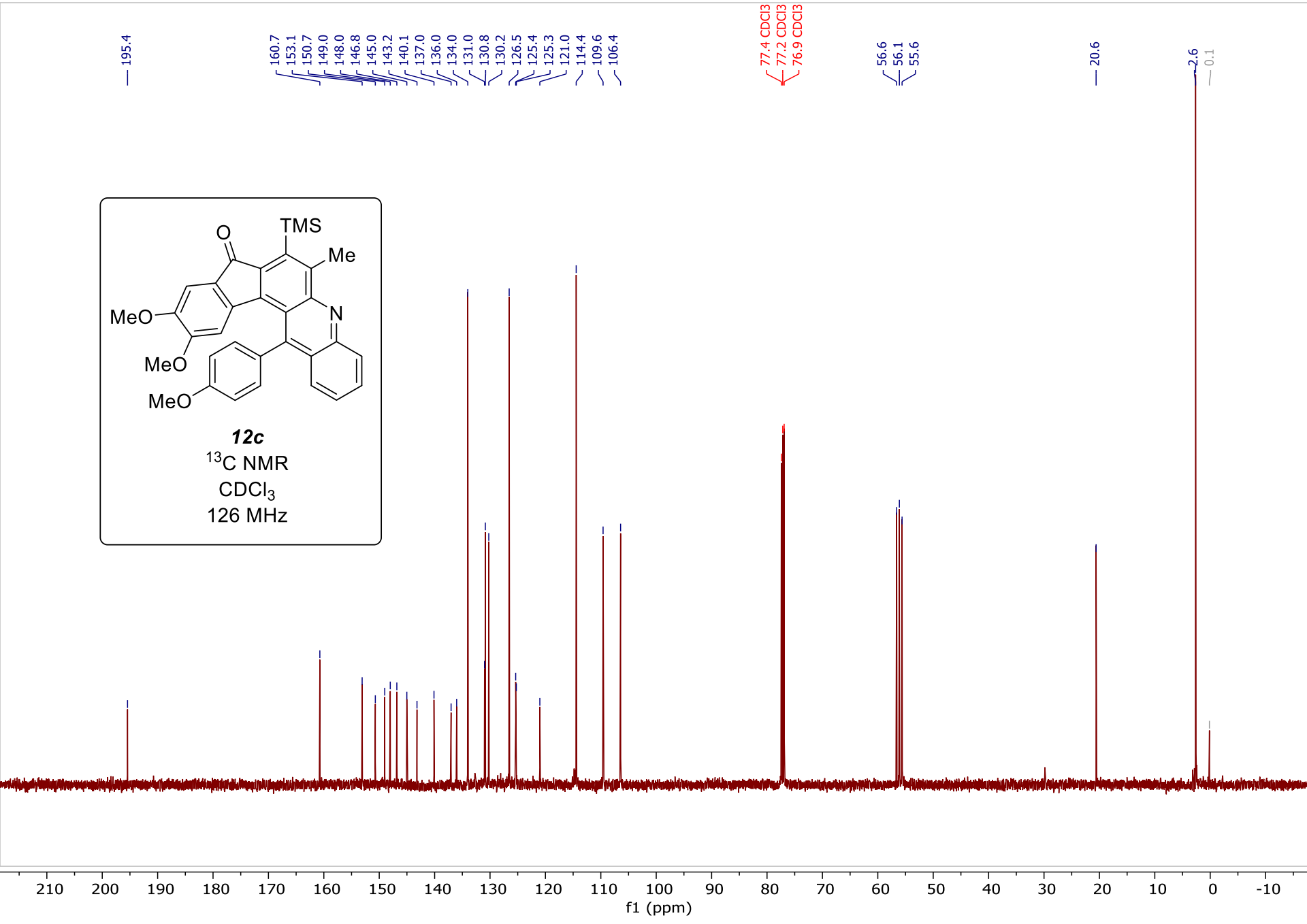
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126.5
125.4
125.3
121.0
114.4
109.6
106.4
77.4 CDCl3
77.2 CDCl3
76.9 CDCl3
56.6
56.1
55.6
20.6
2.6
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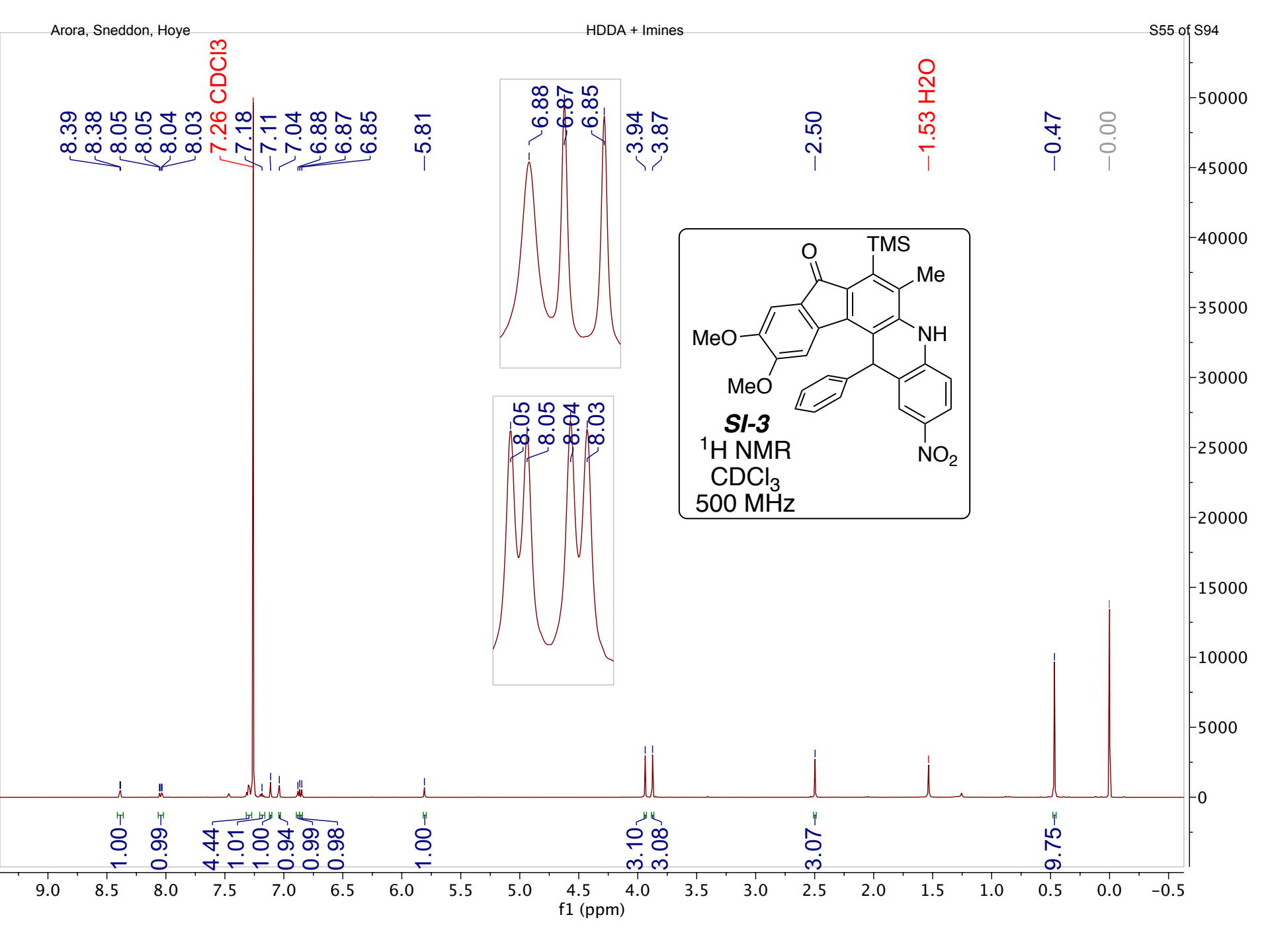
TMS
Me
MeO
N
MeO
MeO
12c
13C NMR
CDCl3
126 MHz
f1 (ppm)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8.38
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8.05
8.04
8.03
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7.18
7.11
7.04
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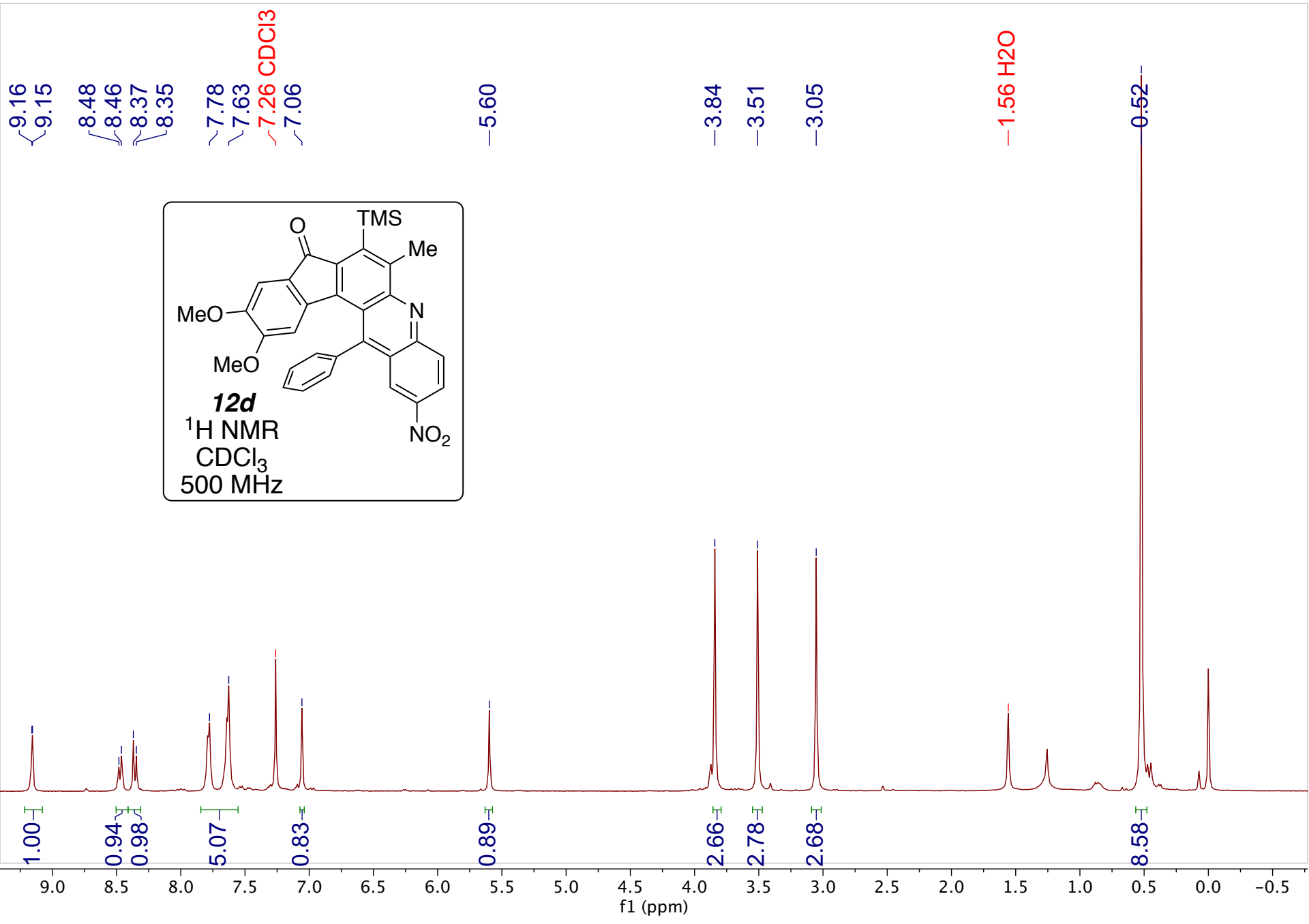
6.87
6.85
5.81
6.88
6.87
6.85
3.94
3.87
2.50
1.53 H2O
0.47
0.00
8.05
8.05
8.04
8.03
O
TMS
Me
MeO
NH
MeO
NO2
SI-3
1H NMR
CDCl3
500 MHz
1.00
0.99
4.44
1.01
1.00
0.94
0.99
0.98
1.00
3.10
3.08
3.07
9.75
f1 (ppm)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7.63
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7.06
5.60
3.84
3.51
3.05
1.56 H2O
0.52
TMS
Me
O
N
MeO
MeO
NO2
12d
1H NMR
CDCl3
500 MHz
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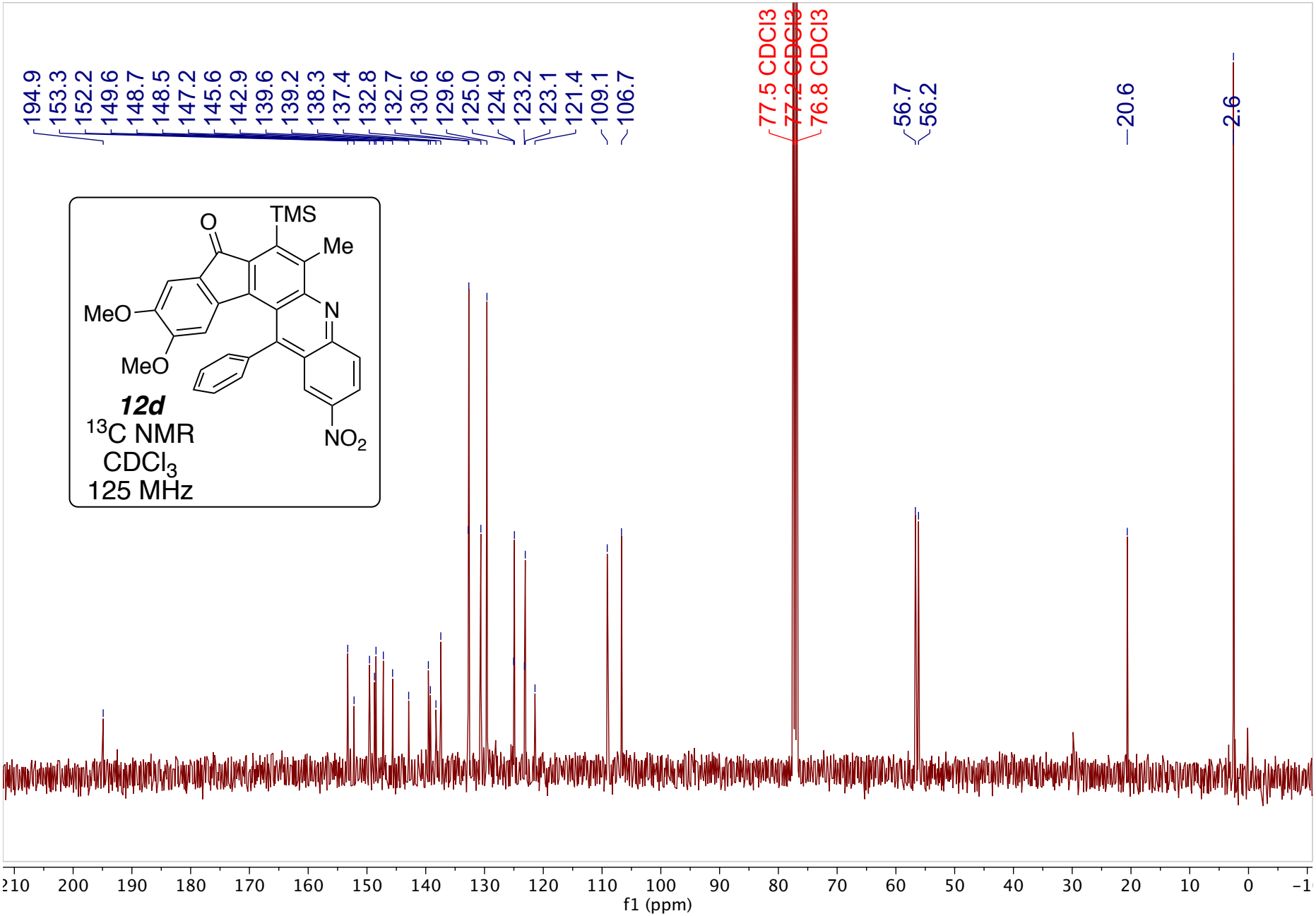
0.94
0.98
5.07
0.83
0.89
2.66
2.78
2.68
8.58
f1 (ppm)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152.2
149.6
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148.5
147.2
145.6
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139.2
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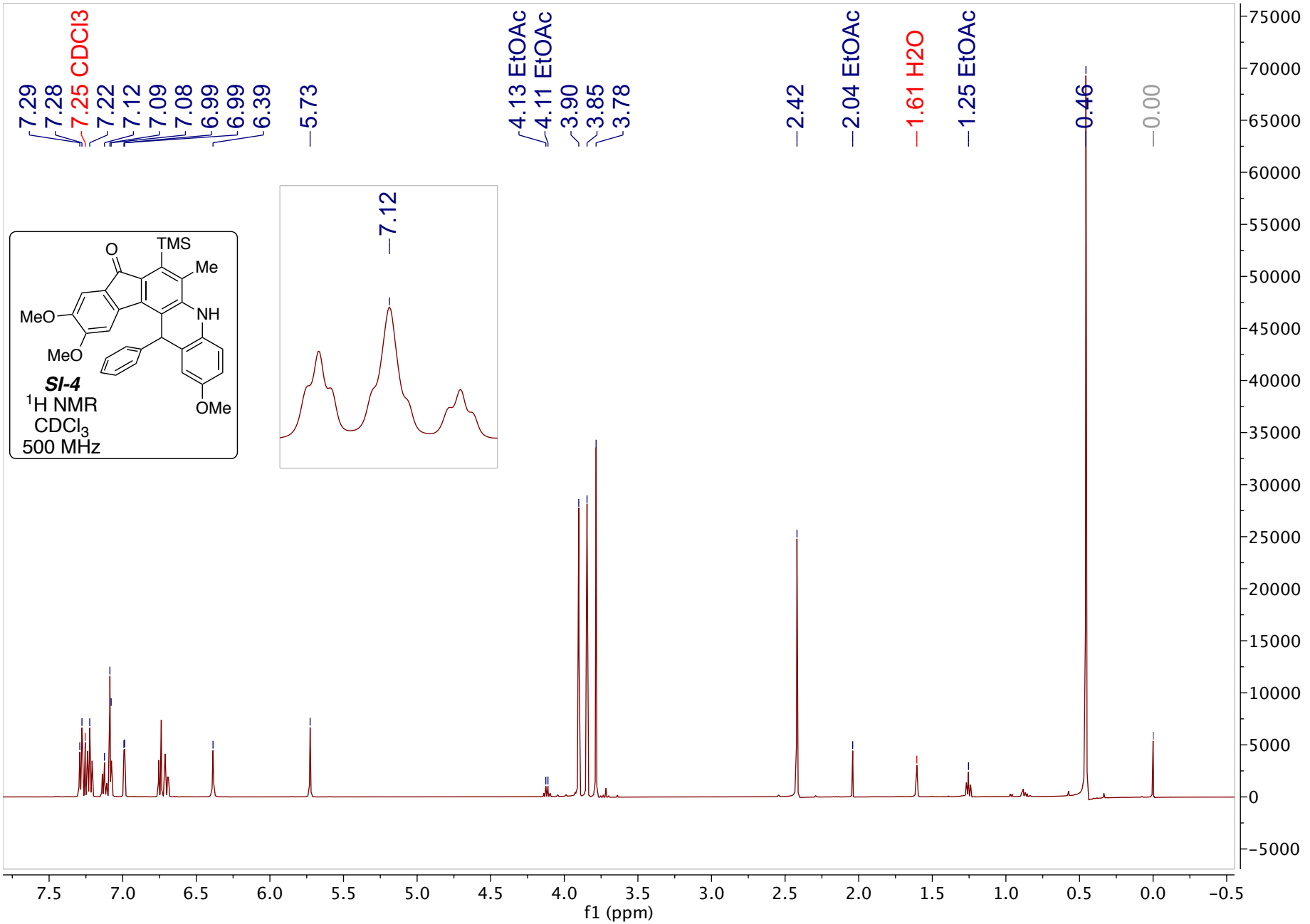
137.4
132.8
132.7
130.6
129.6
125.0
124.9
123.2
123.1
121.4
109.1
106.7
77.5 CDCl3
77.2 CDCl3
76.8 CDCl3
56.7
56.2
20.6
2.6
TMS
Me
MeO
MeO
N
NO2
12d
13C NMR
CDCl3
125 MHz
f1 (ppm)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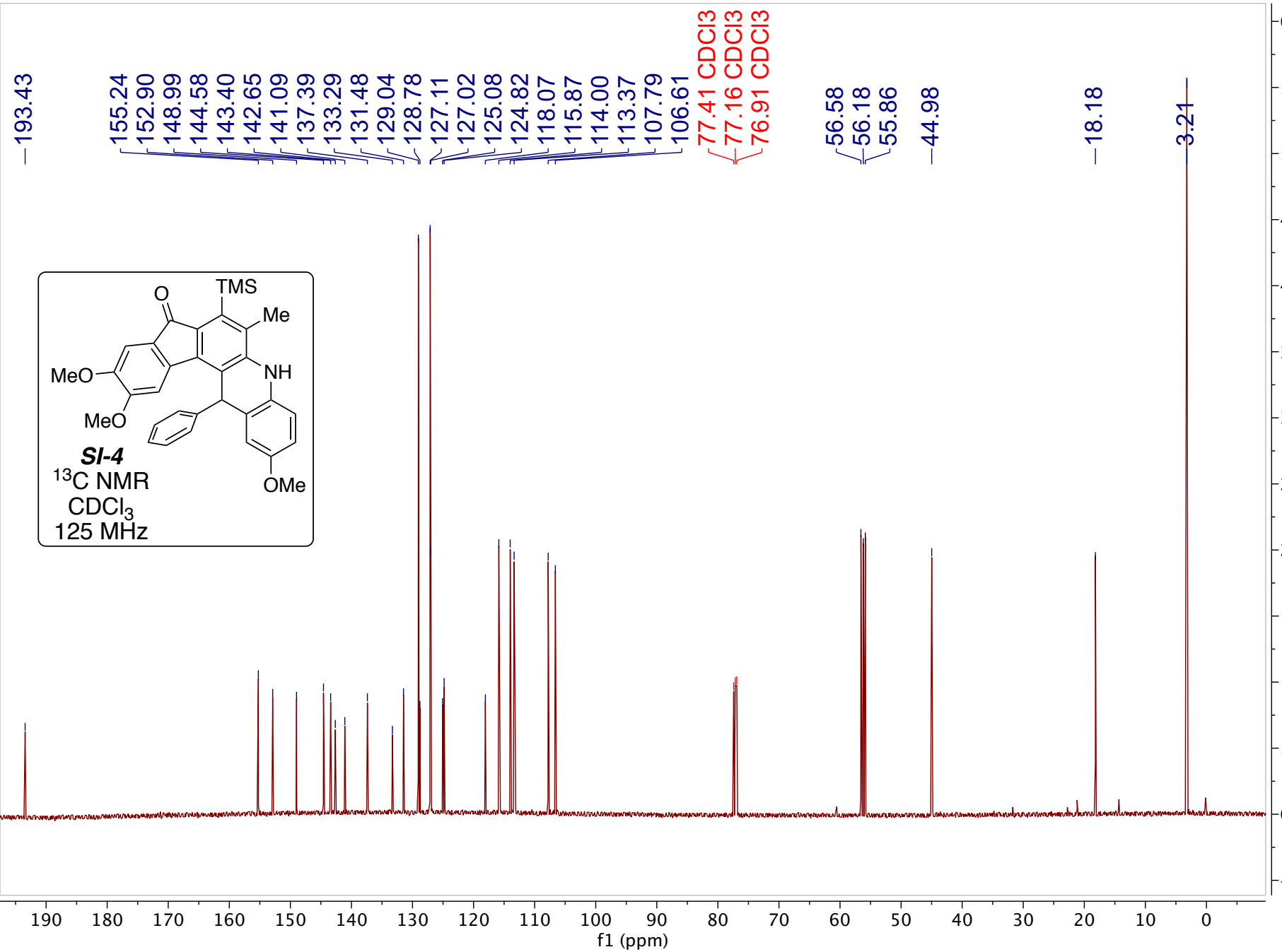
7.28
7.25 CDCl3
7.22
7.12
7.09
7.08
6.99
6.99
6.39
5.73
4.13 EtOAc
4.11 EtOAc
3.90
3.85
3.78
2.42
2.04 EtOAc
1.61 H2O
1.25 EtOAc
0.46
0.00
7.12
TMS
O
Me
MeO
NH
MeO
OMe
SI-4
1H NMR
CDCl3
500 MHz
f1 (ppm)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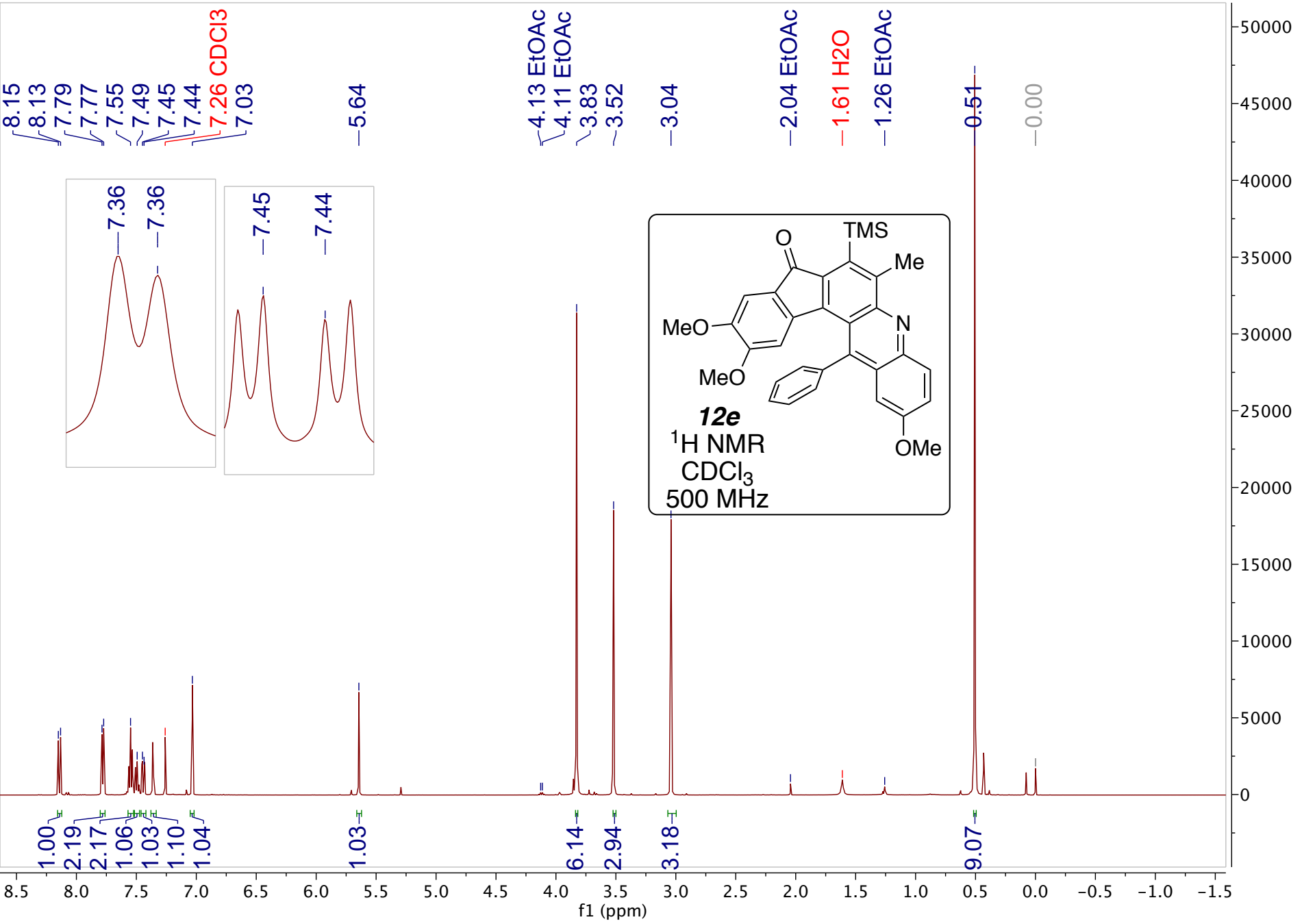
128.78
127.11
127.02
125.08
124.82
118.07
115.87
114.00
113.37
107.79
106.61
77.41 CDCl3
77.16 CDCl3
76.91 CDCl3
56.58
56.18
55.86
44.98
18.18
3.21
O
TMS
Me
MeO
NH
MeO
OMe
SI-4
13C NMR
CDCl3
125 MHz
6000
5500
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
-500
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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8.13
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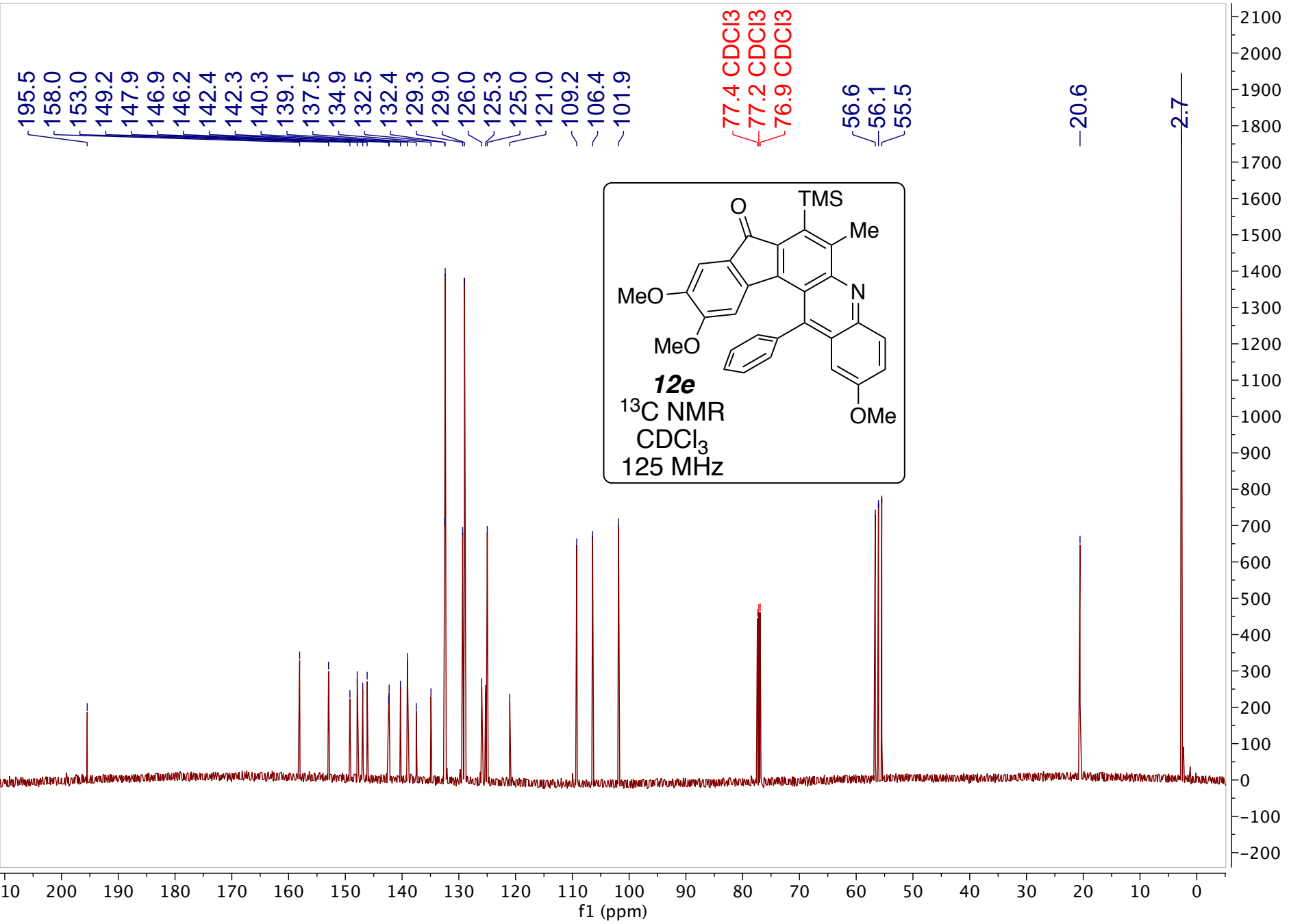
7.77
7.55
7.49
7.45
7.44
7.26 CDCl3
7.03
5.64
4.13 EtOAc
4.11 EtOAc
3.83
3.52
3.04
2.04 EtOAc
1.61 H2O
1.26 EtOAc
0.51
0.00
7.36
7.36
7.45
7.44
TMS
O
Me
MeO
N
MeO
OMe
12e
1H NMR
CDCl3
500 MHz
1.00
2.19
2.17
1.06
1.03
1.10
1.04
1.03
6.14
2.94
3.18
9.07
f1 (ppm)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153.0
149.2
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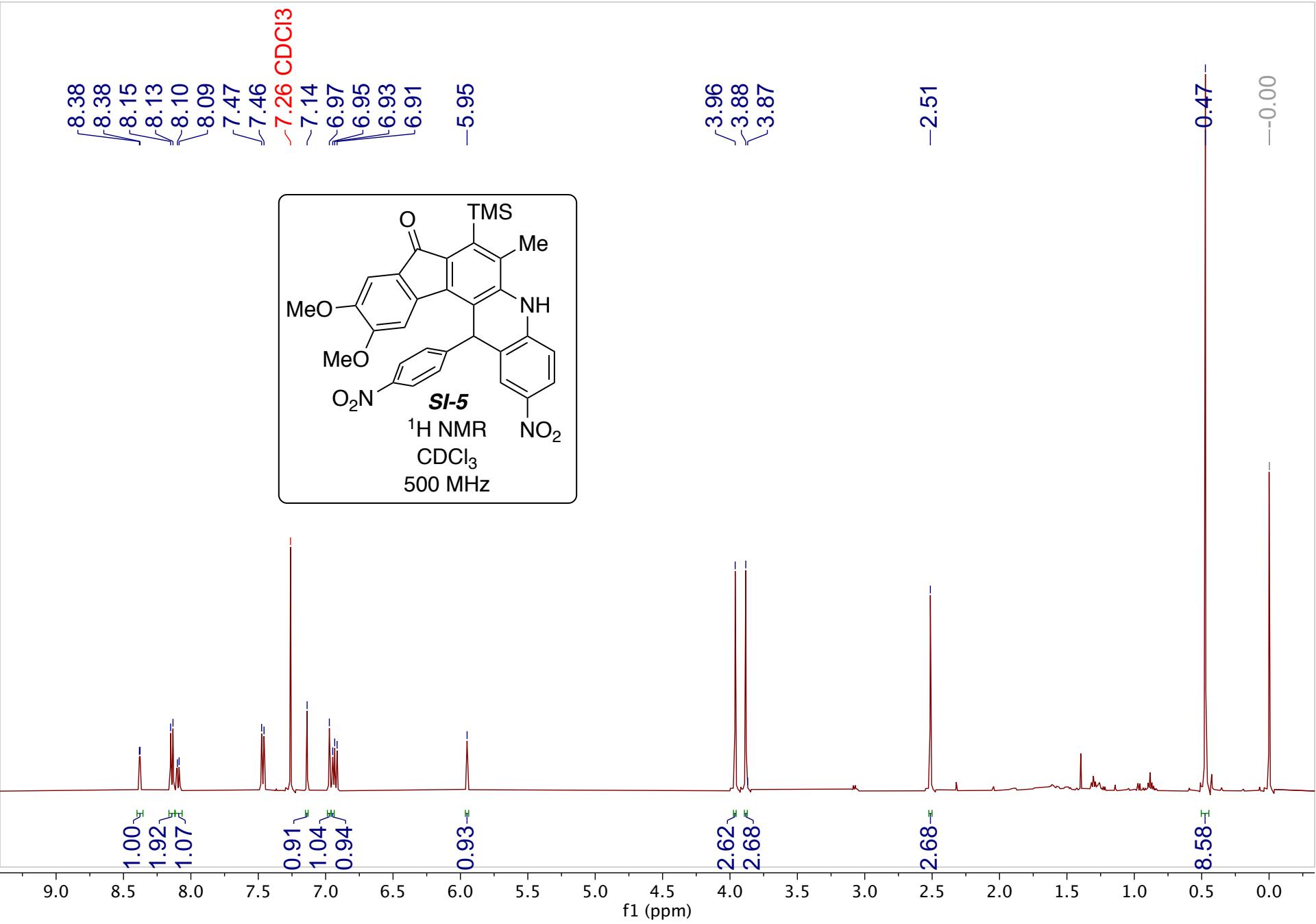
146.9
146.2
142.4
142.3
140.3
139.1
137.5
134.9
132.5
132.4
129.3
129.0
126.0
125.3
125.0
121.0
109.2
106.4
101.9
77.4 CDCl3
77.2 CDCl3
76.9 CDCl3
56.6
56.1
55.5
20.6
2.7
TMS
O
Me
N
MeO
MeO
OMe
12e
13C NMR
CDCl3
125 MHz
f1 (ppm)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8.38
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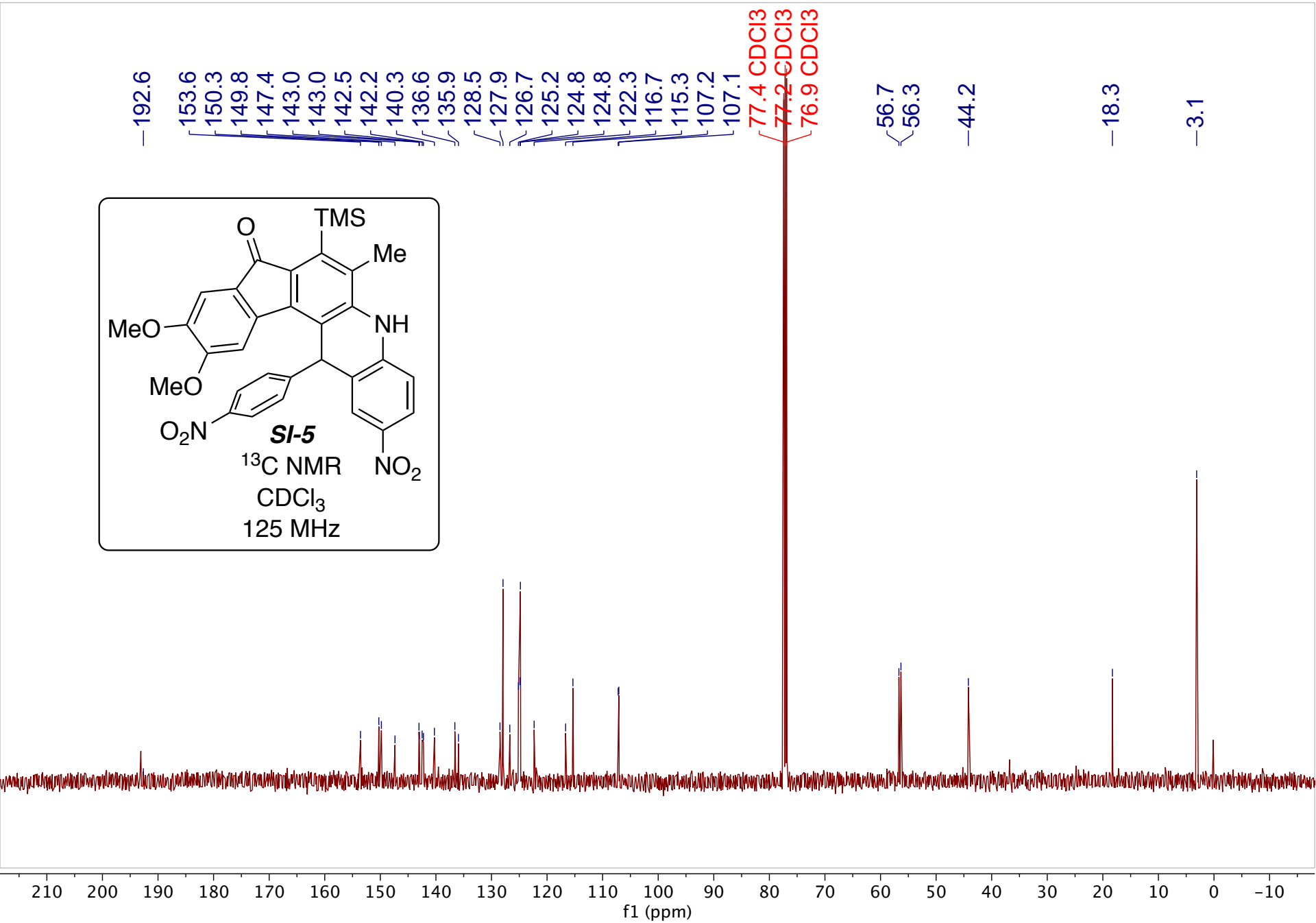
8.09
7.47
7.46
7.26 CDCl3
7.14
6.97
6.95
6.93
6.91
5.95
3.96
3.88
3.87
2.51
0.47
0.00
TMS
O
Me
MeO
NH
MeO
O2N
NO2
SI-5
1H NMR
CDCl3
500 MHz
1.00
1.92
1.07
0.91
1.04
0.94
0.93
2.62
2.68
2.68
8.58
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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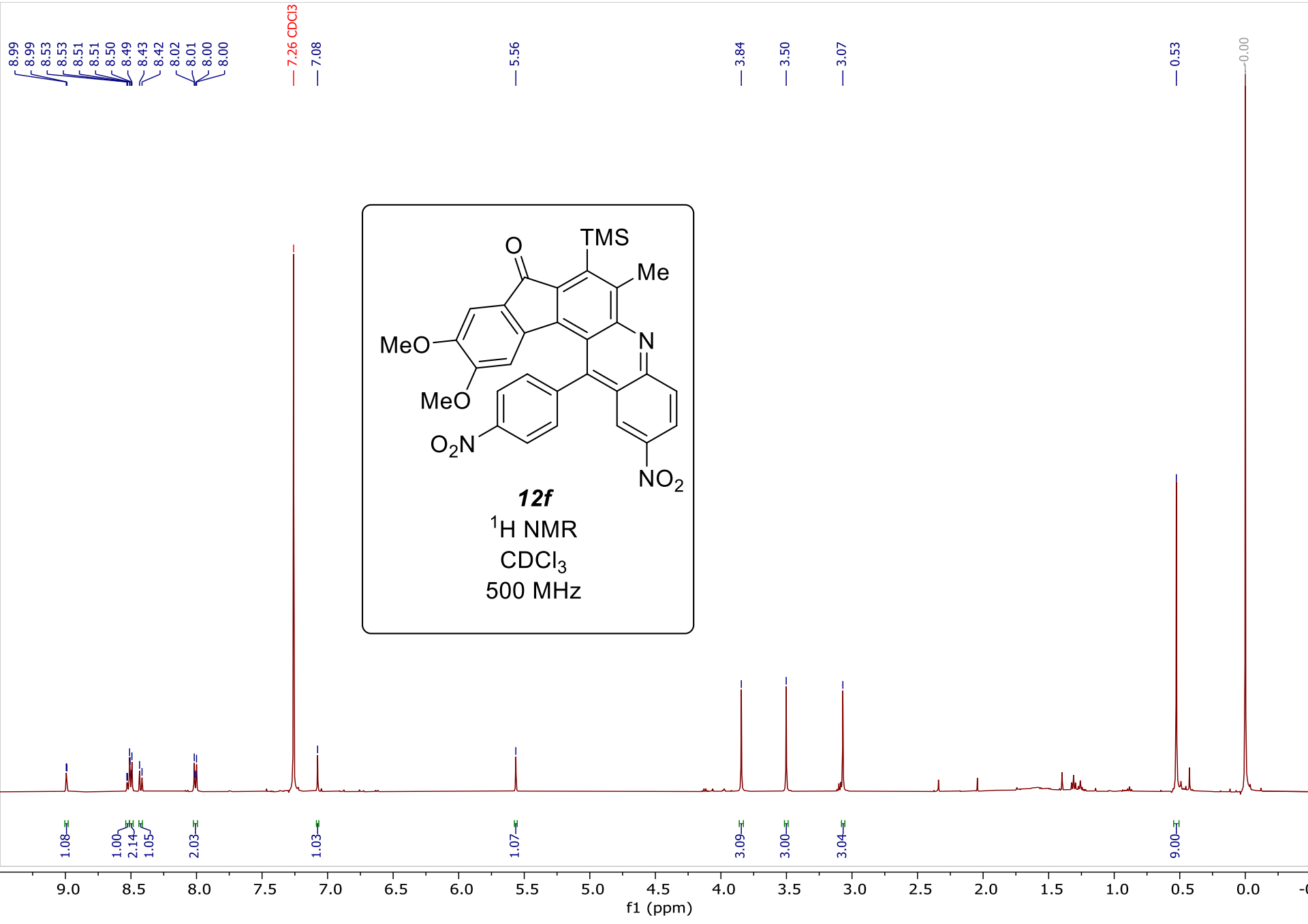
153.6
150.3
149.8
147.4
143.0
143.0
142.5
142.2
140.3
136.6
135.9
128.5
127.9
126.7
125.2
124.8
124.8
122.3
116.7
115.3
107.2
107.1
77.4 CDCl3
77.2 CDCl3
76.9 CDCl3
56.7
56.3
44.2
18.3
3.1
TMS
O
Me
MeO
NH
MeO
O2N
NO2
SI-5
13C NMR
CDCl3
125 MHz
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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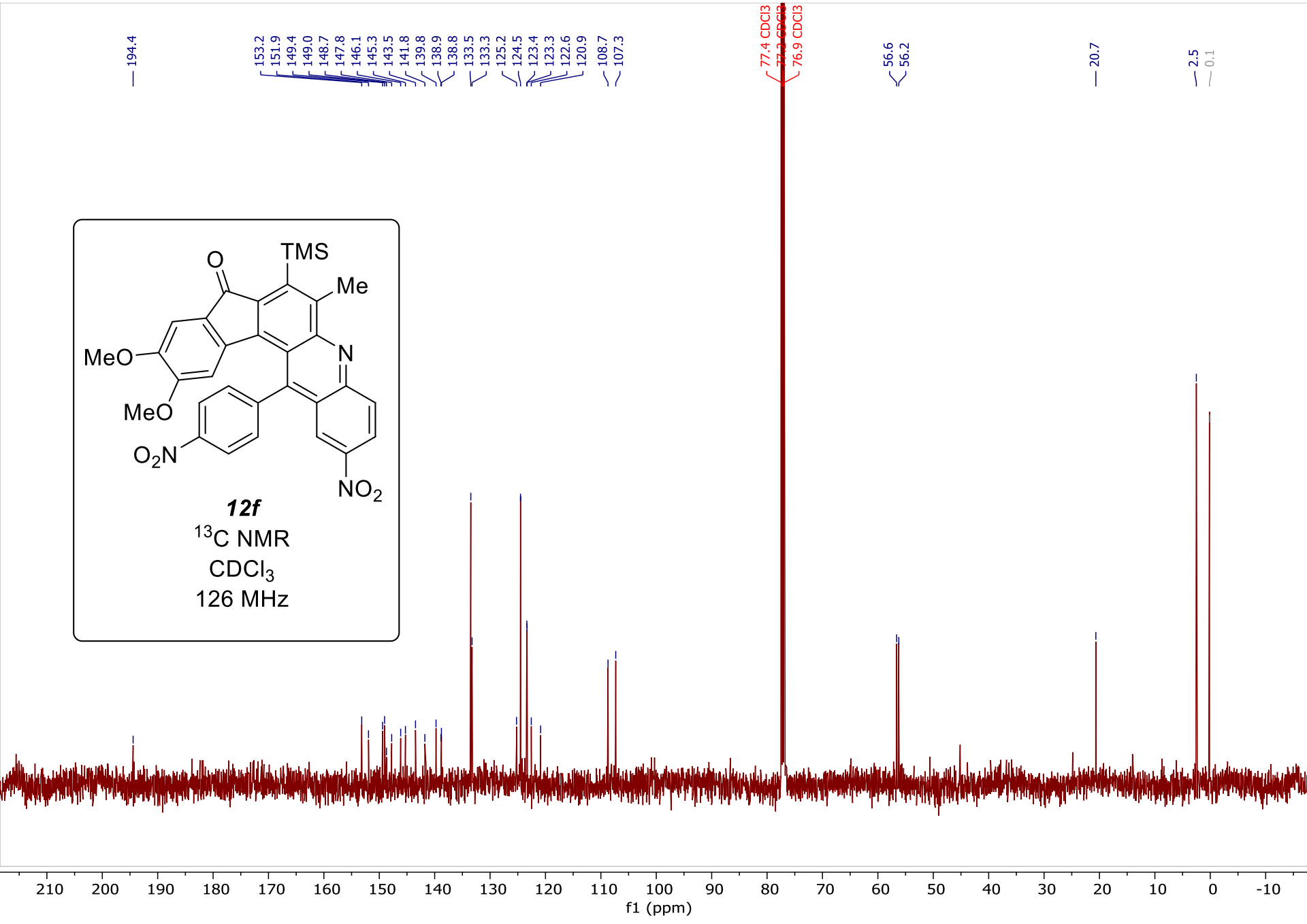
8.99
8.53
8.53
8.51
8.51
8.50
8.49
8.43
8.42
8.02
8.01
8.00
8.00
7.26 CDCl3
7.08
5.56
3.84
3.50
3.07
0.53
0.00
TMS
O
Me
MeO
N
MeO
O2N
NO2
12f
1H NMR
CDCl3
500 MHz
1.08
1.00
2.14
1.05
2.03
1.03
1.07
3.09
3.00
3.04
9.00
f1 (ppm)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151.9
149.4
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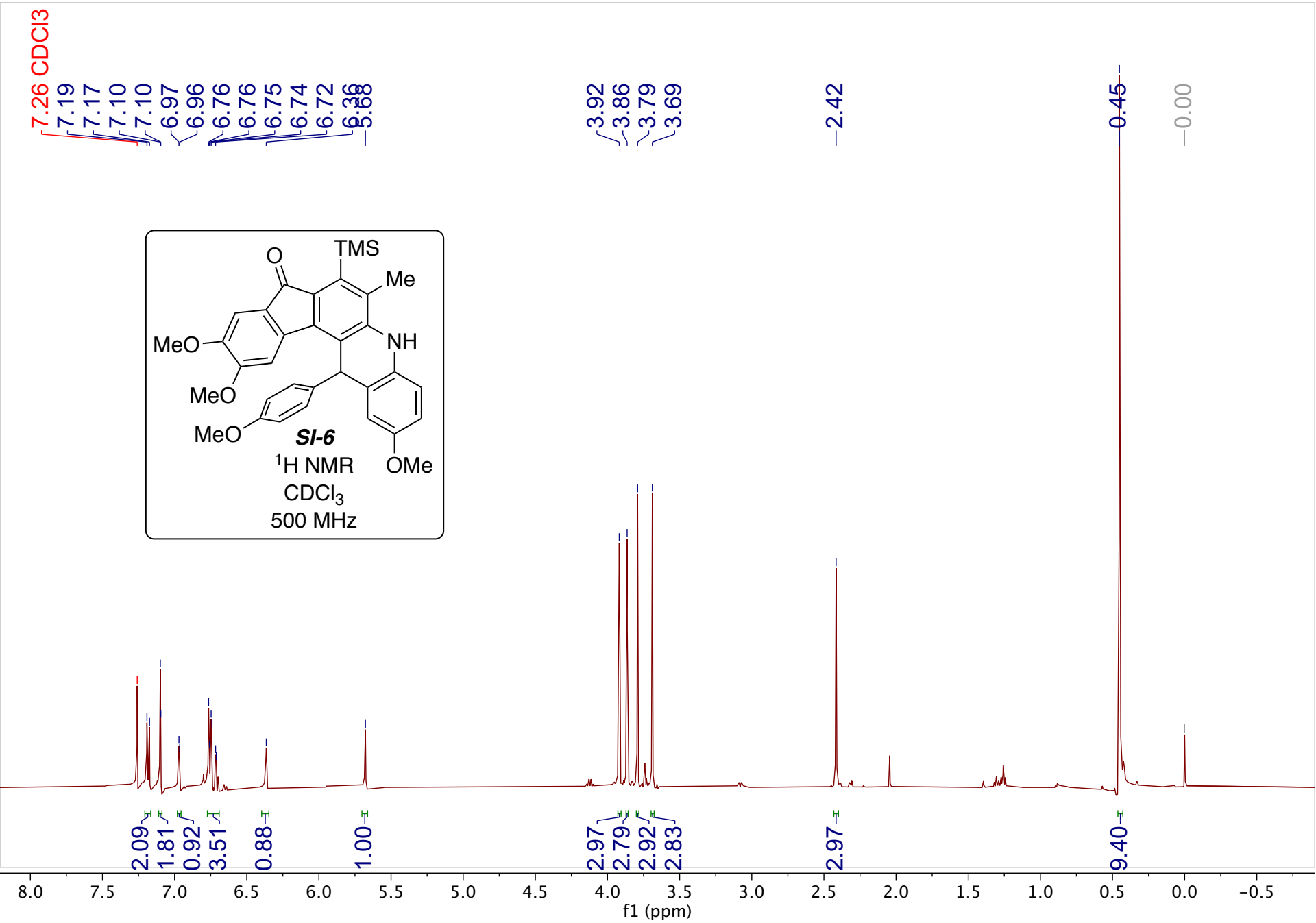
148.7
147.8
146.1
145.3
143.5
141.8
139.8
138.9
138.8
133.5
133.3
125.2
124.5
123.4
123.3
122.6
120.9
108.7
107.3
77.4 CDCl3
76.9 CDCl3
56.6
56.2
20.7
2.5
0.1
O
TMS
Me
MeO
N
MeO
O2N
NO2
12f
13C NMR
CDCl3
126 MHz
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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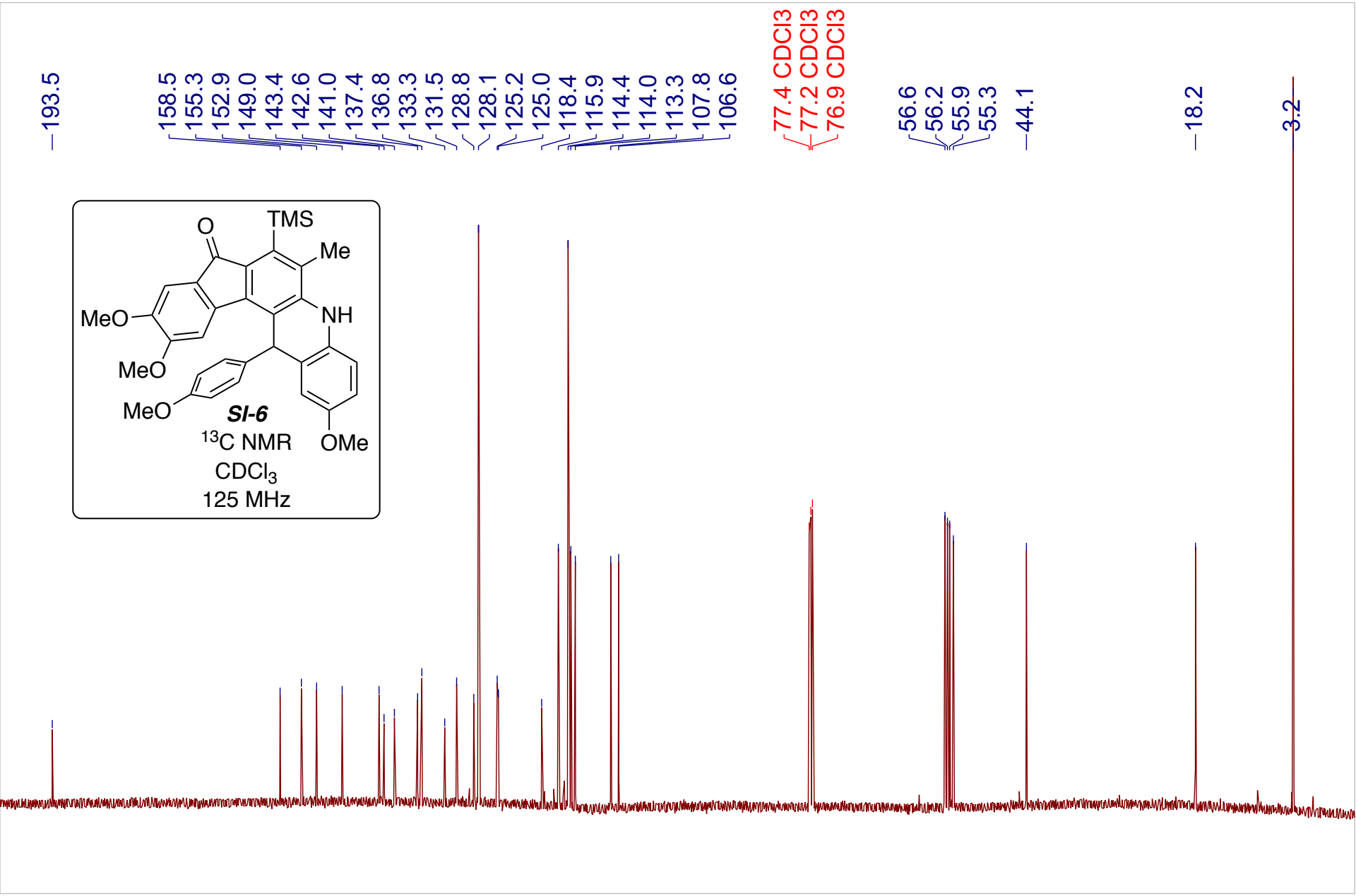
7.19
7.17
7.10
7.10
6.97
6.96
6.76
6.76
6.75
6.74
6.72
6.36
5.68
3.92
3.86
3.79
3.69
2.42
0.45
0.00
TMS
O
Me
MeO
NH
MeO
MeO
OMe
SI-6
1H NMR
CDCl3
500 MHz
2.09
1.81
0.92
3.51
0.88
1.00
2.97
2.79
2.92
2.83
2.97
9.40
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
f1 (ppm)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152.9
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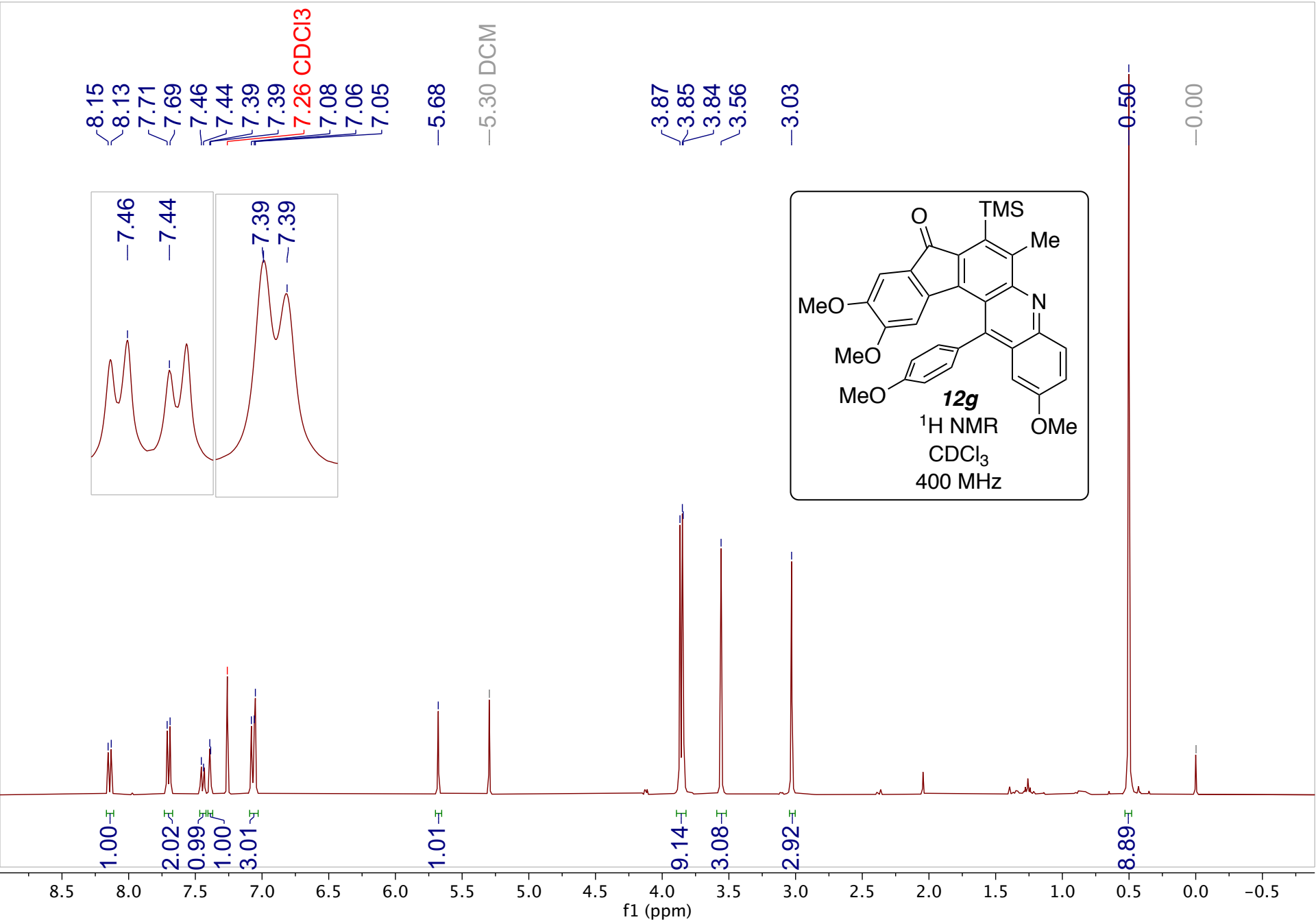
143.4
142.6
141.0
137.4
136.8
133.3
131.5
128.8
128.1
125.2
125.0
118.4
115.9
114.4
114.0
113.3
107.8
106.6
77.4 CDCl3
77.2 CDCl3
76.9 CDCl3
56.6
56.2
55.9
55.3
44.1
18.2
3.2
O
TMS
Me
MeO
NH
MeO
MeO
OMe
SI-6
13C NMR
CDCl3
125 MHz
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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8.13
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7.46
7.44
7.39
7.39
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7.08
7.06
7.05
5.68
5.30 DCM
3.87
3.85
3.84
3.56
3.03
0.50
0.00
TMS
O
Me
MeO
N
MeO
MeO
OMe
12g
1H NMR
CDCl3
400 MHz
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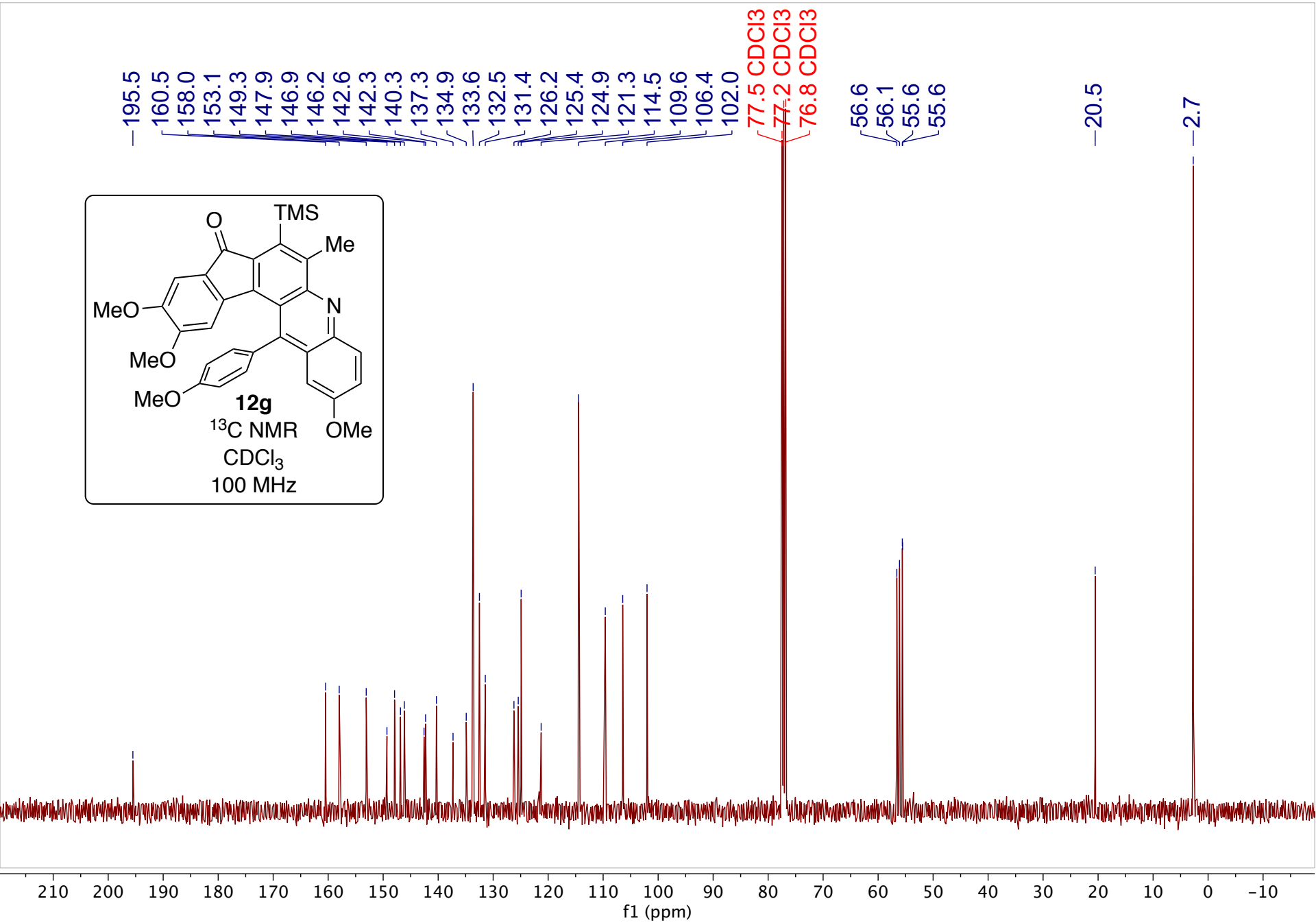
0.99
1.00
3.01
1.01
9.14
3.08
2.92
8.89
f1 (ppm)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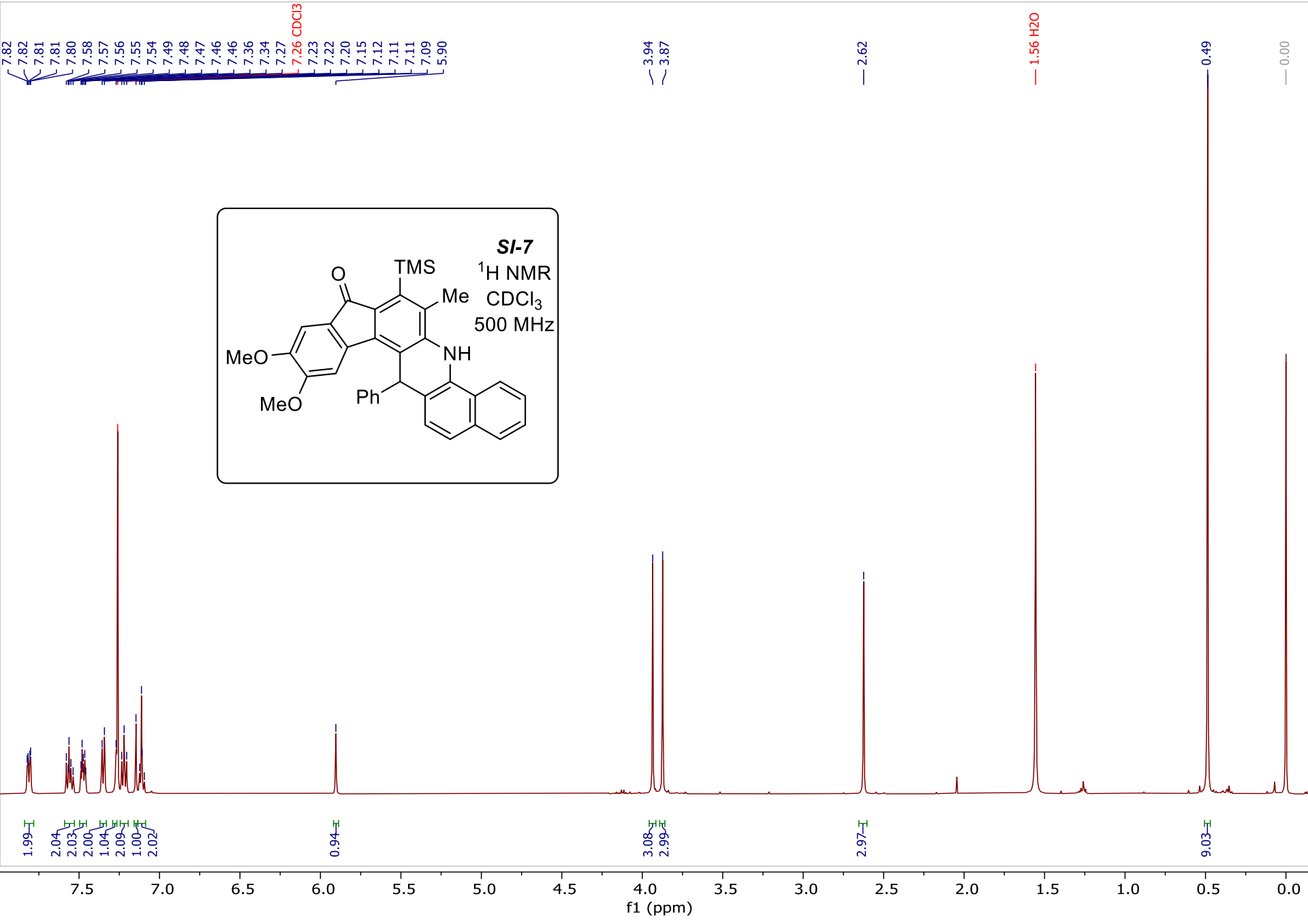
147.9
146.9
146.2
142.6
142.3
140.3
137.3
134.9
133.6
132.5
131.4
126.2
125.4
124.9
121.3
114.5
109.6
106.4
102.0
77.5 CDCl3
77.2 CDCl3
76.8 CDCl3
56.6
56.1
55.6
55.6
20.5
2.7
TMS
O
Me
MeO
N
MeO
MeO
OMe
12g
13C NMR
CDCl3
100 MHz
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)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7.81
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7.58
7.57
7.56
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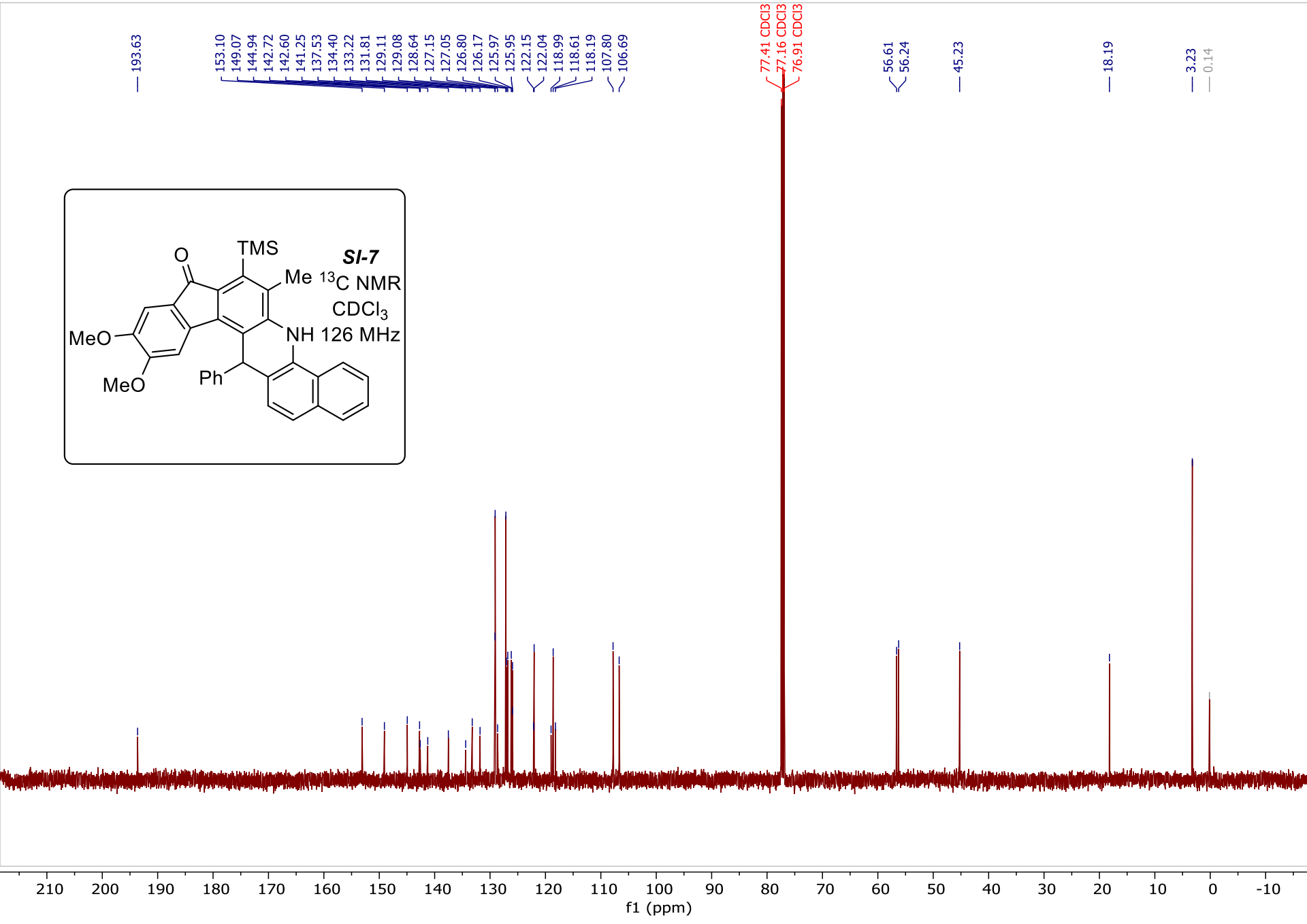
7.54
7.49
7.48
7.47
7.46
7.46
7.36
7.34
7.27
7.26 CDCl3
7.23
7.22
7.20
7.15
7.12
7.11
7.11
7.09
5.90
3.94
3.87
2.62
1.56 H2O
0.49
0.00
SI-7
1H NMR
CDCl3
500 MHz
O
TMS
Me
NH
MeO
MeO
Ph
1.99
2.04
2.03
2.00
1.04
2.09
1.00
2.02
0.94
3.08
2.99
2.97
9.03
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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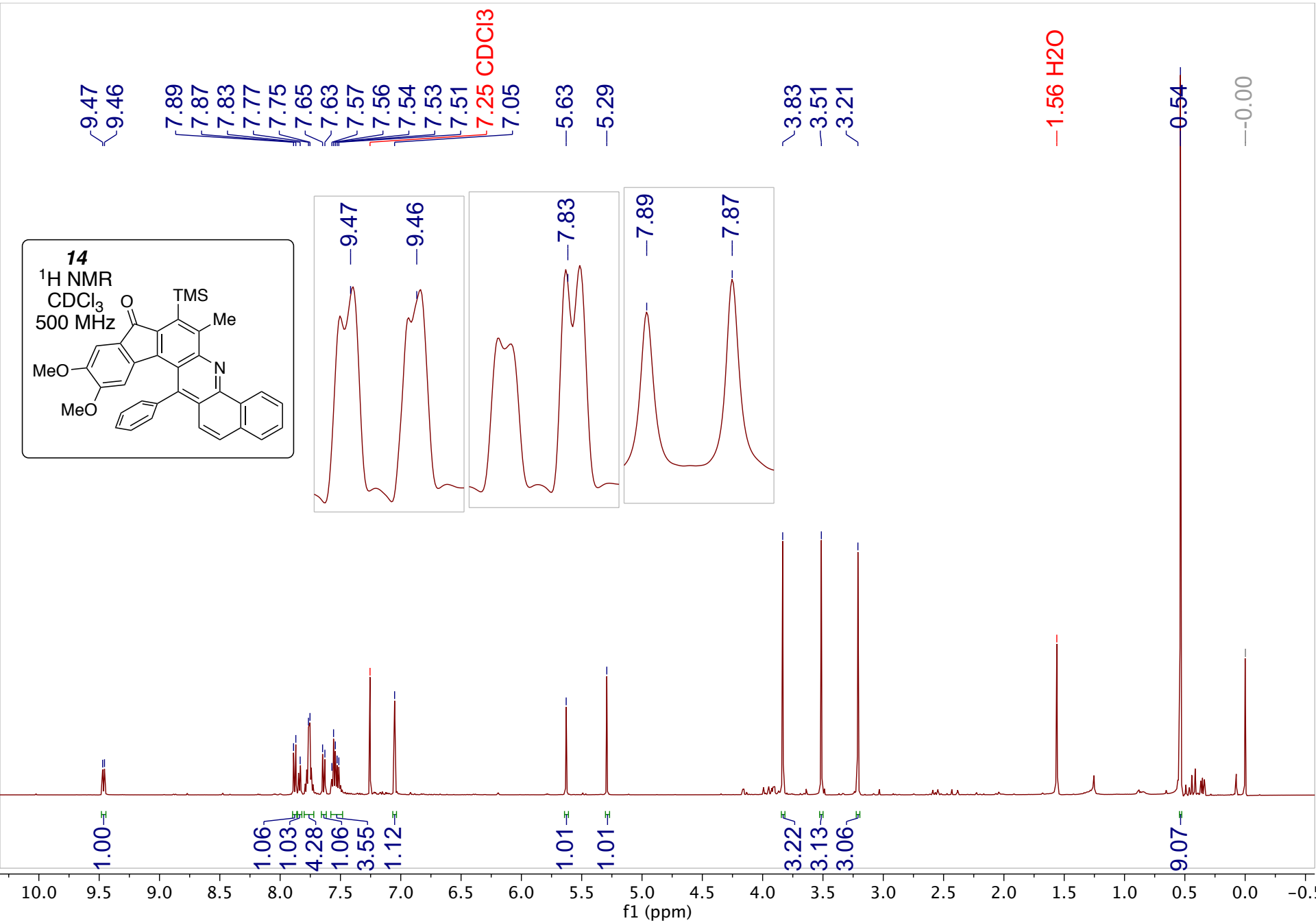
133.22
131.81
129.11
129.08
128.64
127.15
127.05
126.80
126.17
125.97
125.95
122.15
122.04
118.99
118.61
118.19
107.80
106.69
77.41 CDCl3
77.16 CDCl3
76.91 CDCl3
56.61
56.24
45.23
18.19
3.23
0.14
TMS
O
Me
MeO
MeO
Ph
NH
SI-7
13C NMR
CDCl3
126 MHz
f1 (ppm)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9.46
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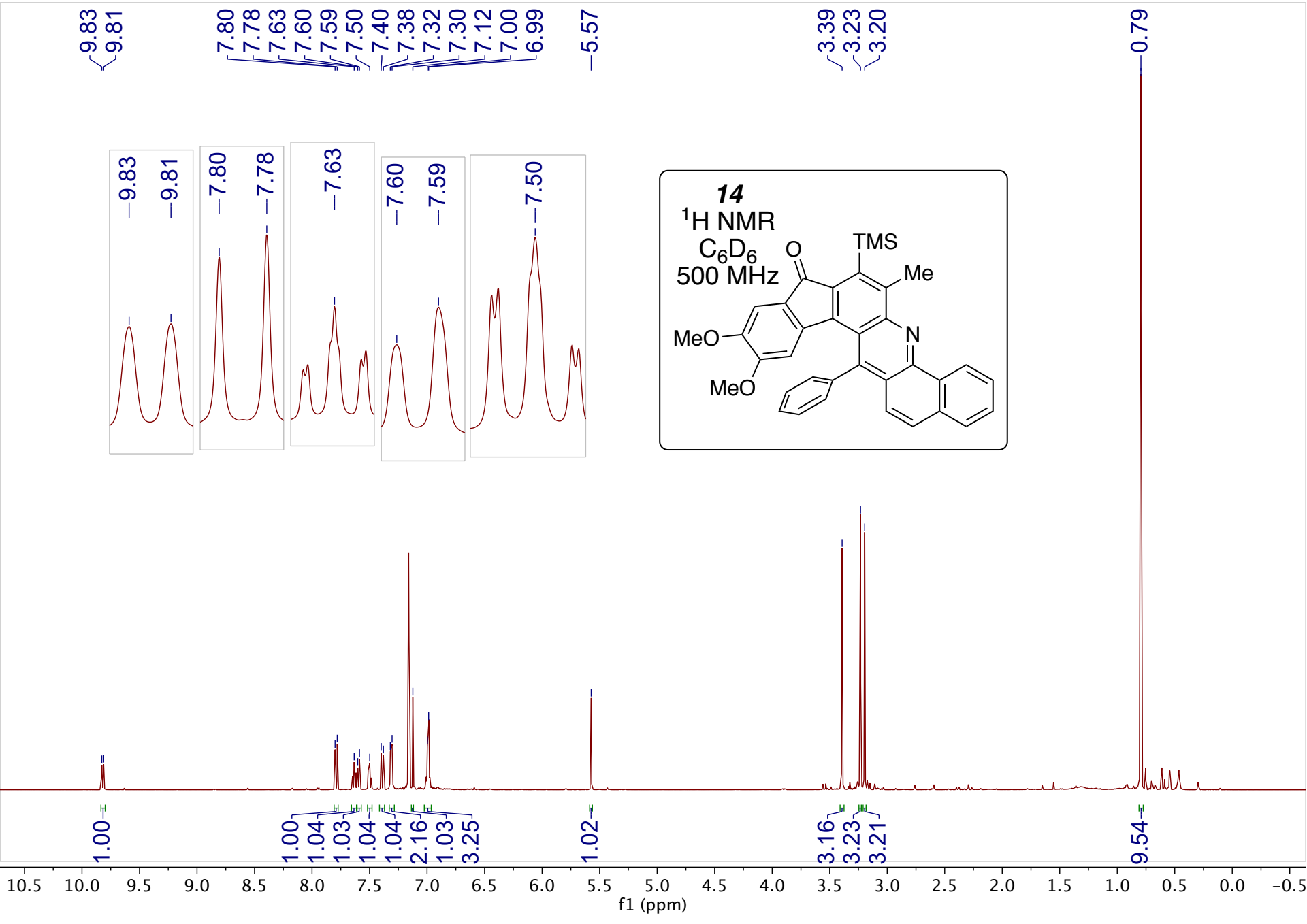
7.77
7.75
7.65
7.63
7.57
7.56
7.54
7.53
7.51
7.25 CDCl3
7.05
5.63
5.29
3.83
3.51
3.21
1.56 H2O
0.54
0.00
14
1H NMR
CDCl3
500 MHz
O
TMS
Me
MeO
MeO
N
1.00
1.06
1.03
4.28
1.06
3.55
1.12
1.01
1.01
3.22
3.13
3.06
9.07
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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9.81
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7.78
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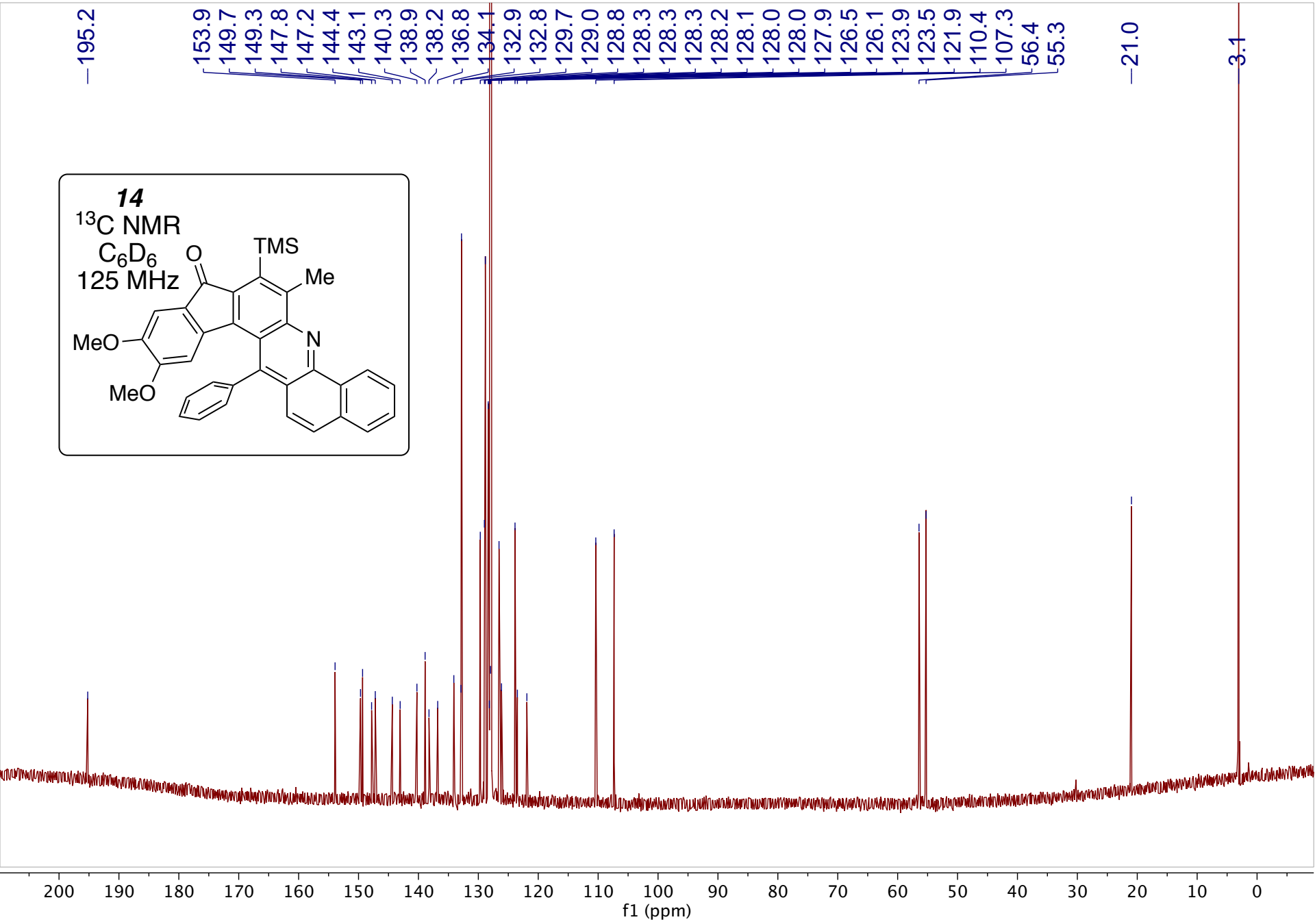
7.60
7.59
7.50
7.40
7.38
7.32
7.30
7.12
7.00
6.99
5.57
3.39
3.23
3.20
0.79
14
1H NMR
C6D6
500 MHz
O
TMS
Me
MeO
MeO
N
1.00
1.00
1.04
1.03
1.04
1.04
2.16
1.03
3.25
1.02
3.16
3.23
3.21
9.54
f1 (ppm)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147.2
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143.1
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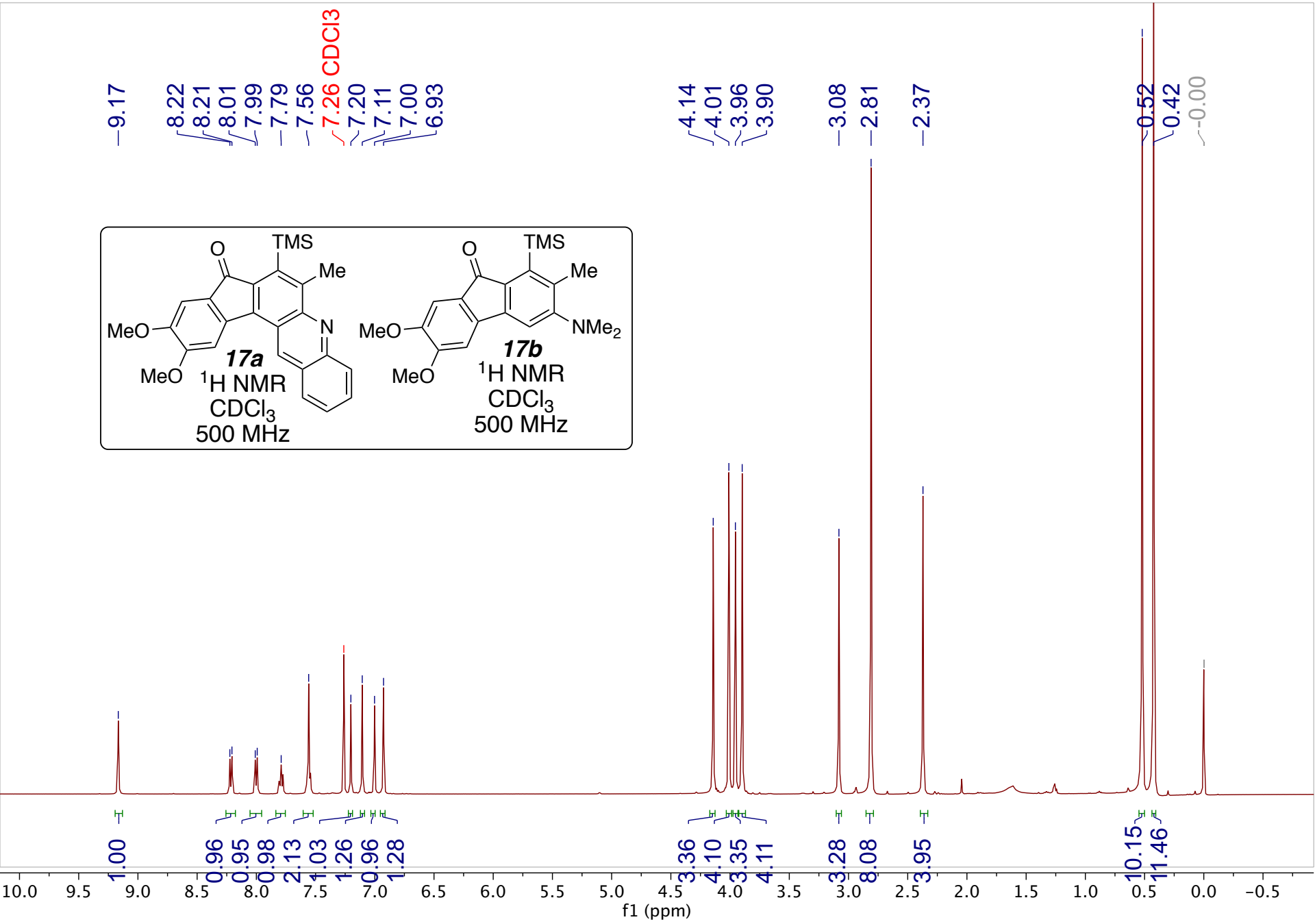
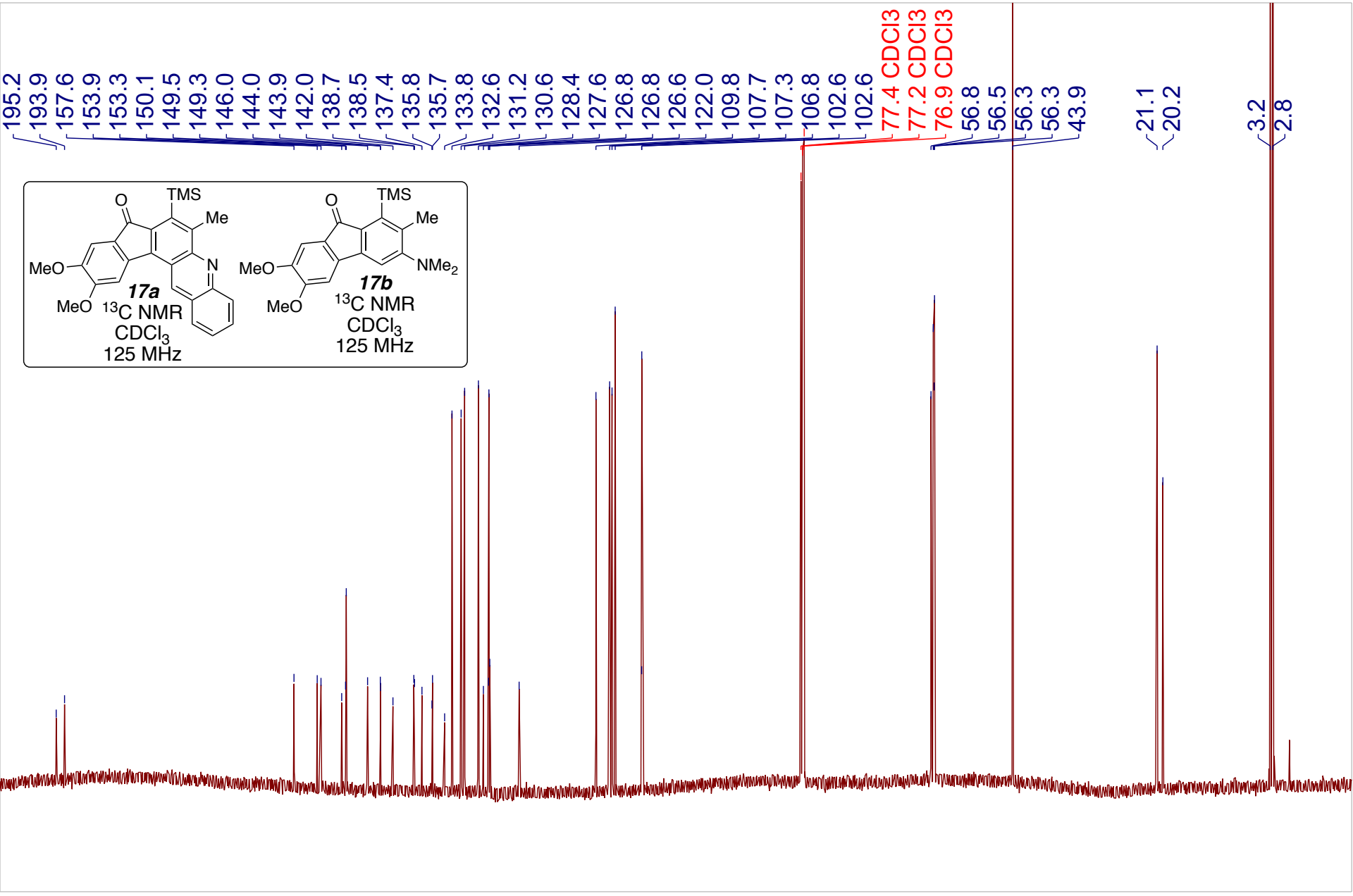
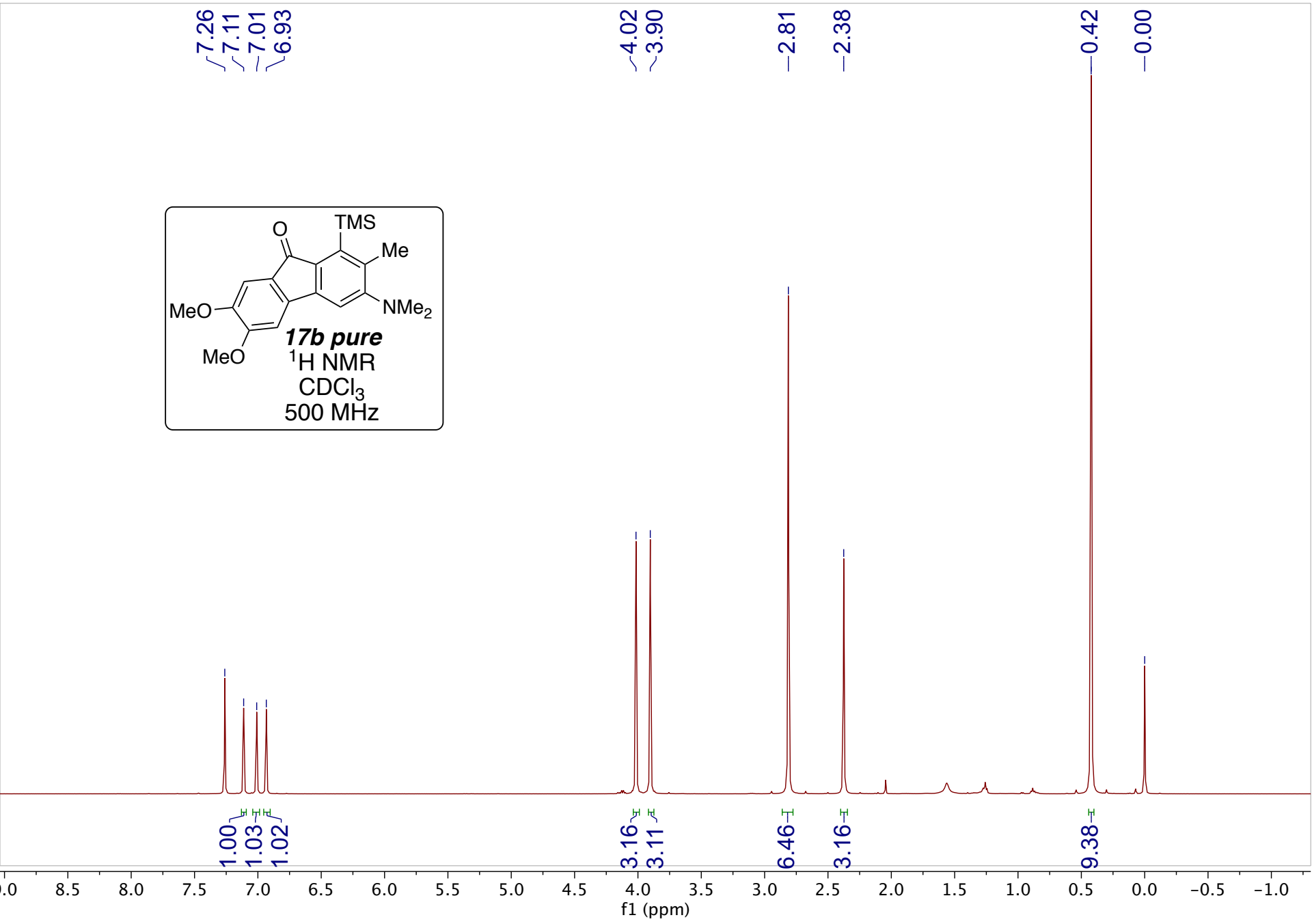
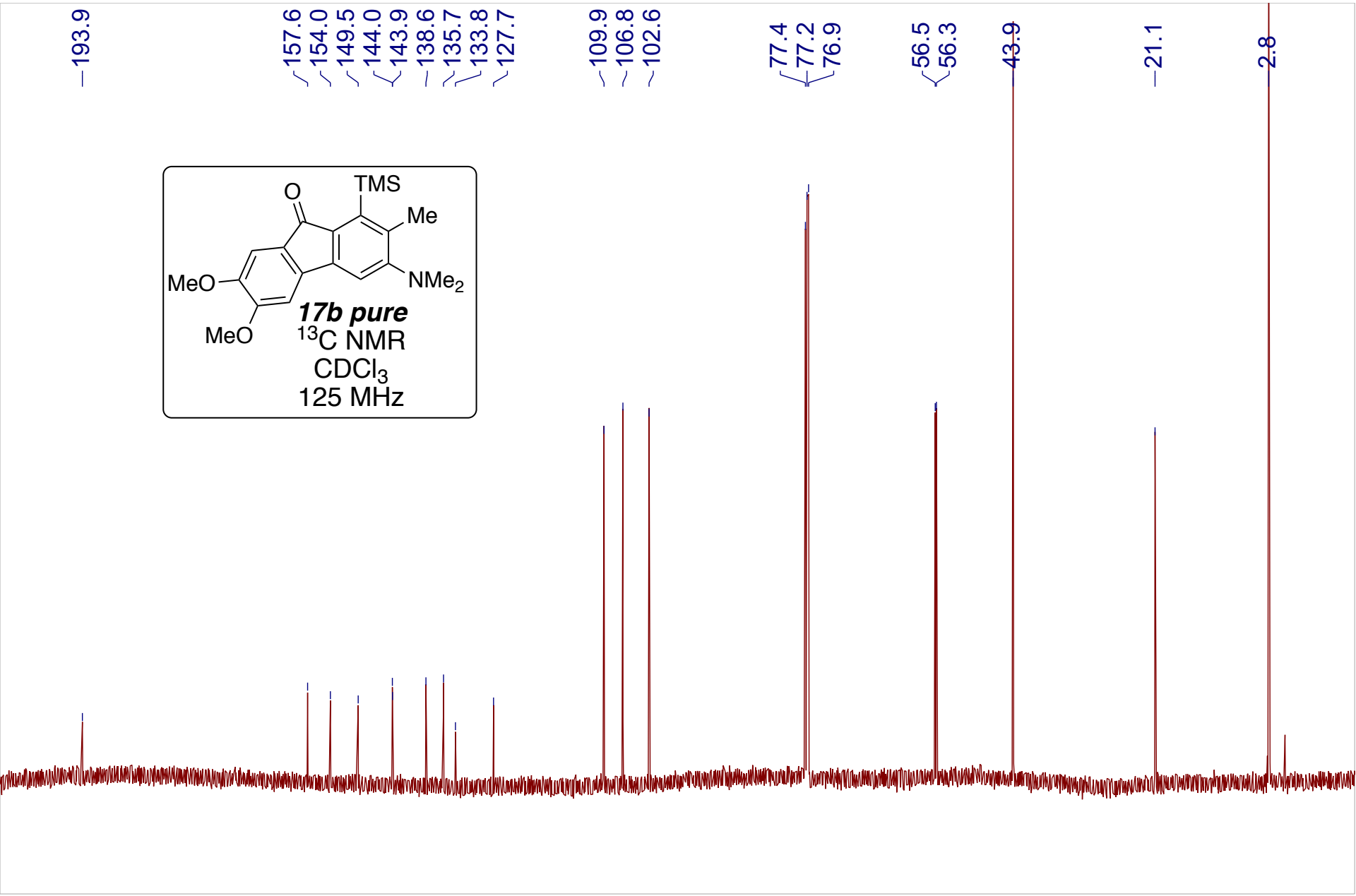
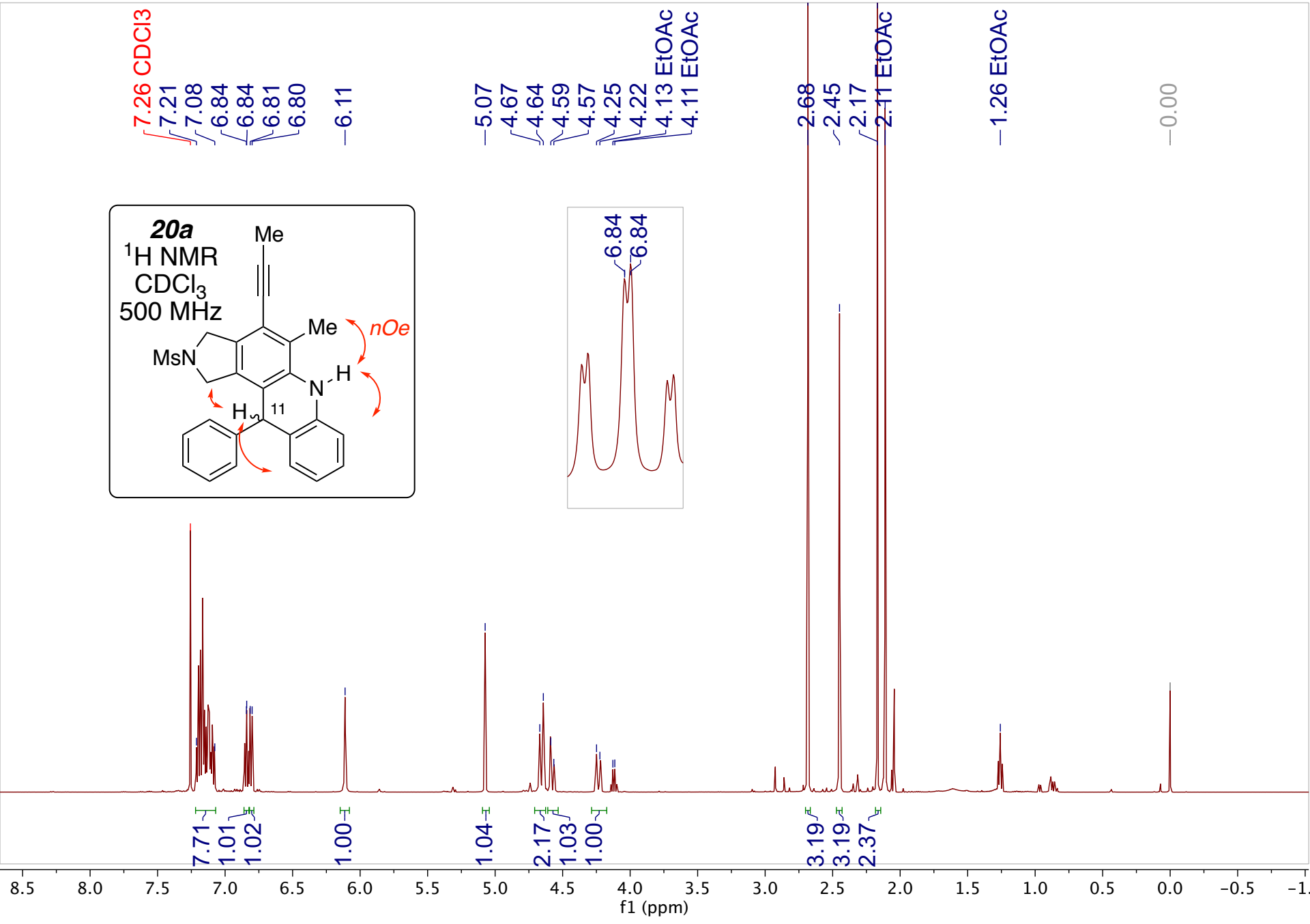
138.2
136.8
134.1
132.9
132.8
129.7
129.0
128.8
128.3
128.3
128.3
128.2
128.1
128.0
128.0
127.9
126.5
126.1
123.9
123.5
121.9
110.4
107.3
56.4
55.3
21.0
3.1
14
13C NMR
C6D6
125 MHz
O
TMS
Me
MeO
MeO
N
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

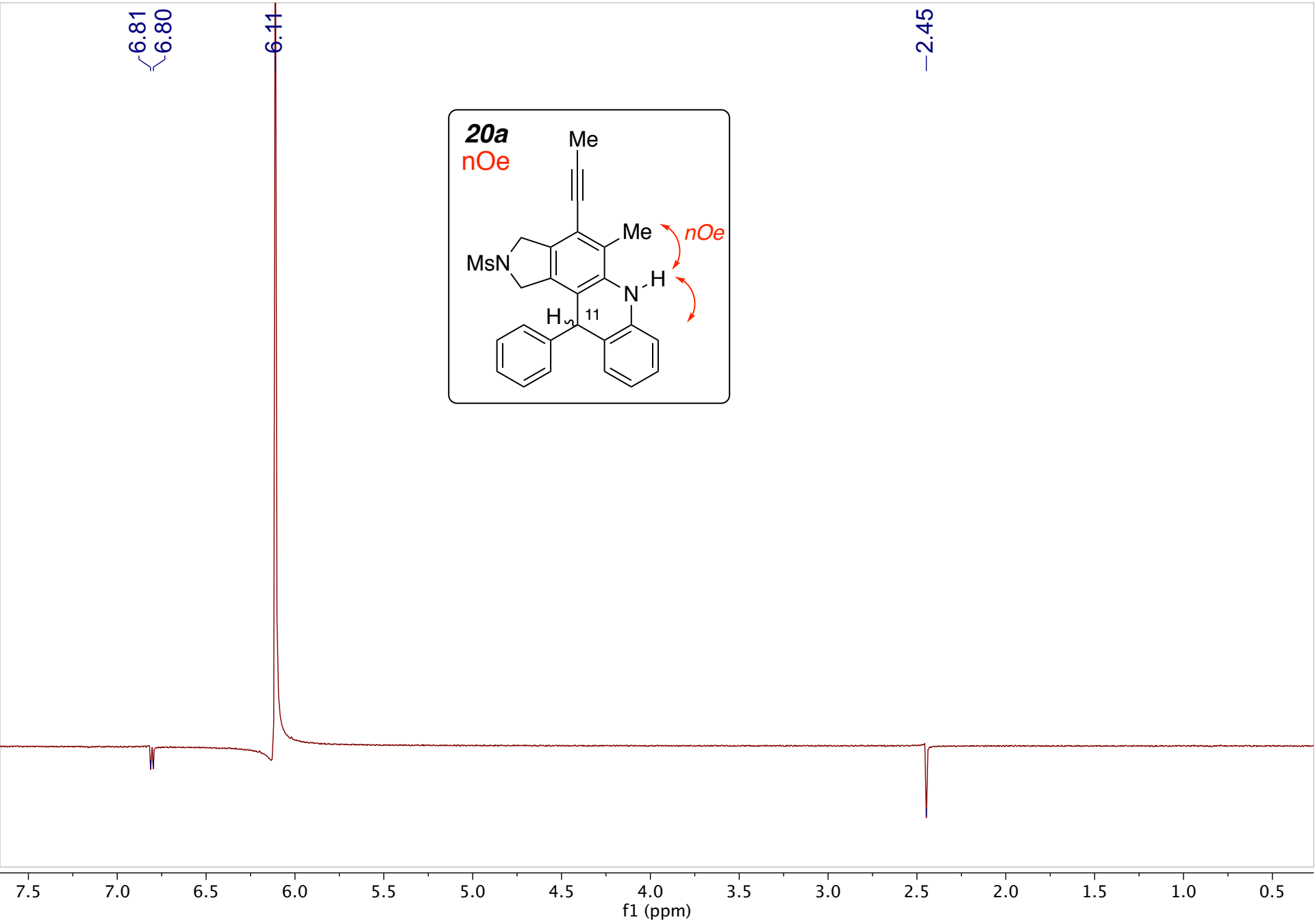

9.17
8.22
8.21
8.01
7.99
7.79
7.56
7.26 CDCl3
7.20
7.11
7.00
6.93
4.14
4.01
3.96
3.90
3.08
2.81
2.37
0.52
0.42
-0.00
TMS
O
Me
MeO
N
MeO
17a
1H NMR
CDCl3
500 MHz
TMS
O
Me
NMe2
MeO
MeO
17b
1H NMR
CDCl3
500 MHz
1.00
0.96
0.95
0.98
2.13
1.03
1.26
0.96
1.28
3.36
4.10
3.35
4.11
3.28
8.08
3.95
10.15
11.46
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
f1 (ppm)

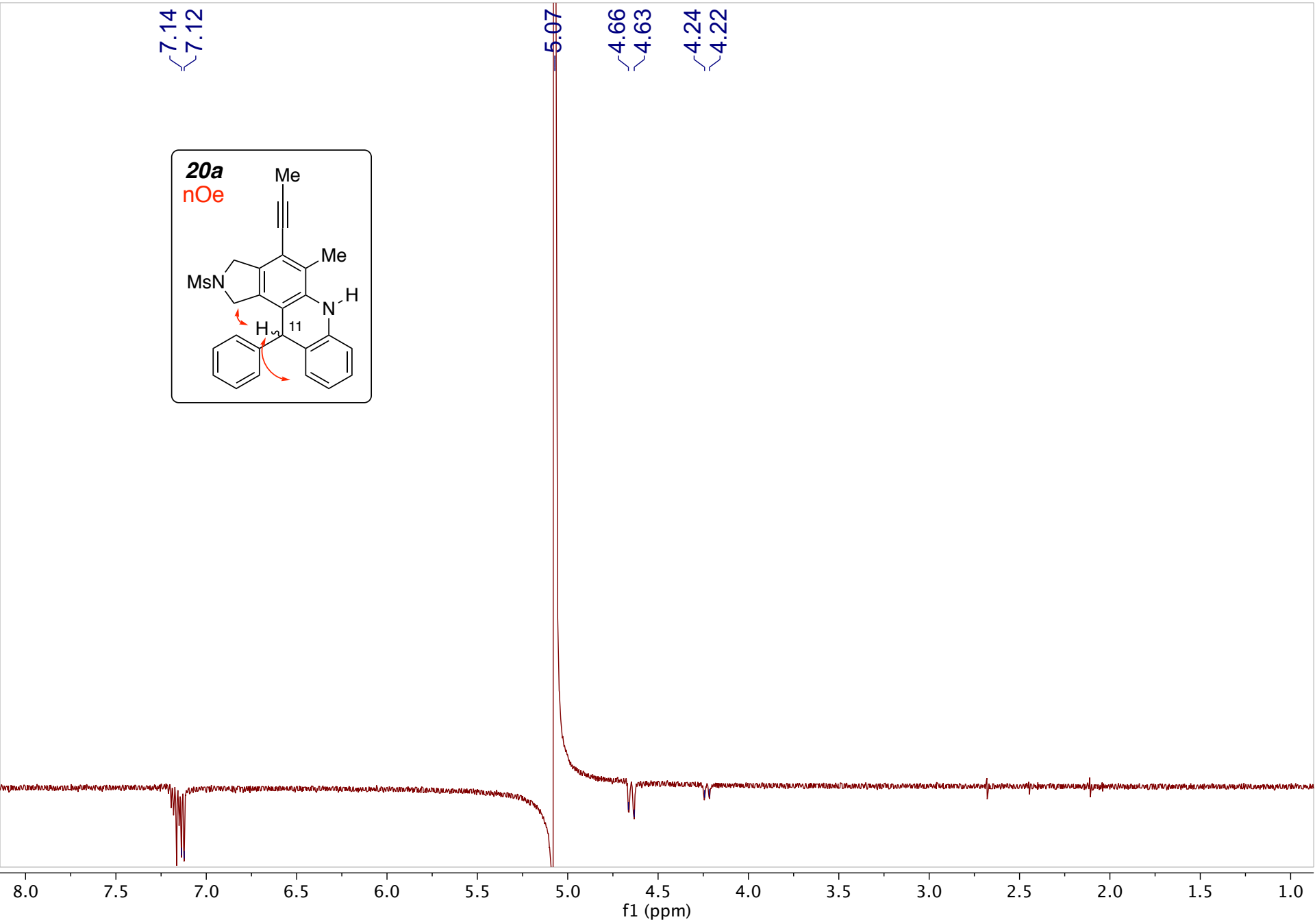

195.2
193.9
157.6
153.9
153.3
150.1
149.5
149.3
146.0
144.0
143.9
142.0
138.7
138.5
137.4
135.8
135.7
133.8
132.6
131.2
130.6
128.4
127.6
126.8
126.8
126.6
122.0
109.8
107.7
107.3
106.8
102.6
102.6
77.4 CDCl3
77.2 CDCl3
76.9 CDCl3
56.8
56.5
56.3
56.3
43.9
21.1
20.2
3.2
2.8
TMS
O
Me
MeO
N
MeO
17a
13C NMR
CDCl3
125 MHz
TMS
O
Me
MeO
NMe2
MeO
17b
13C NMR
CDCl3
125 MHz
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

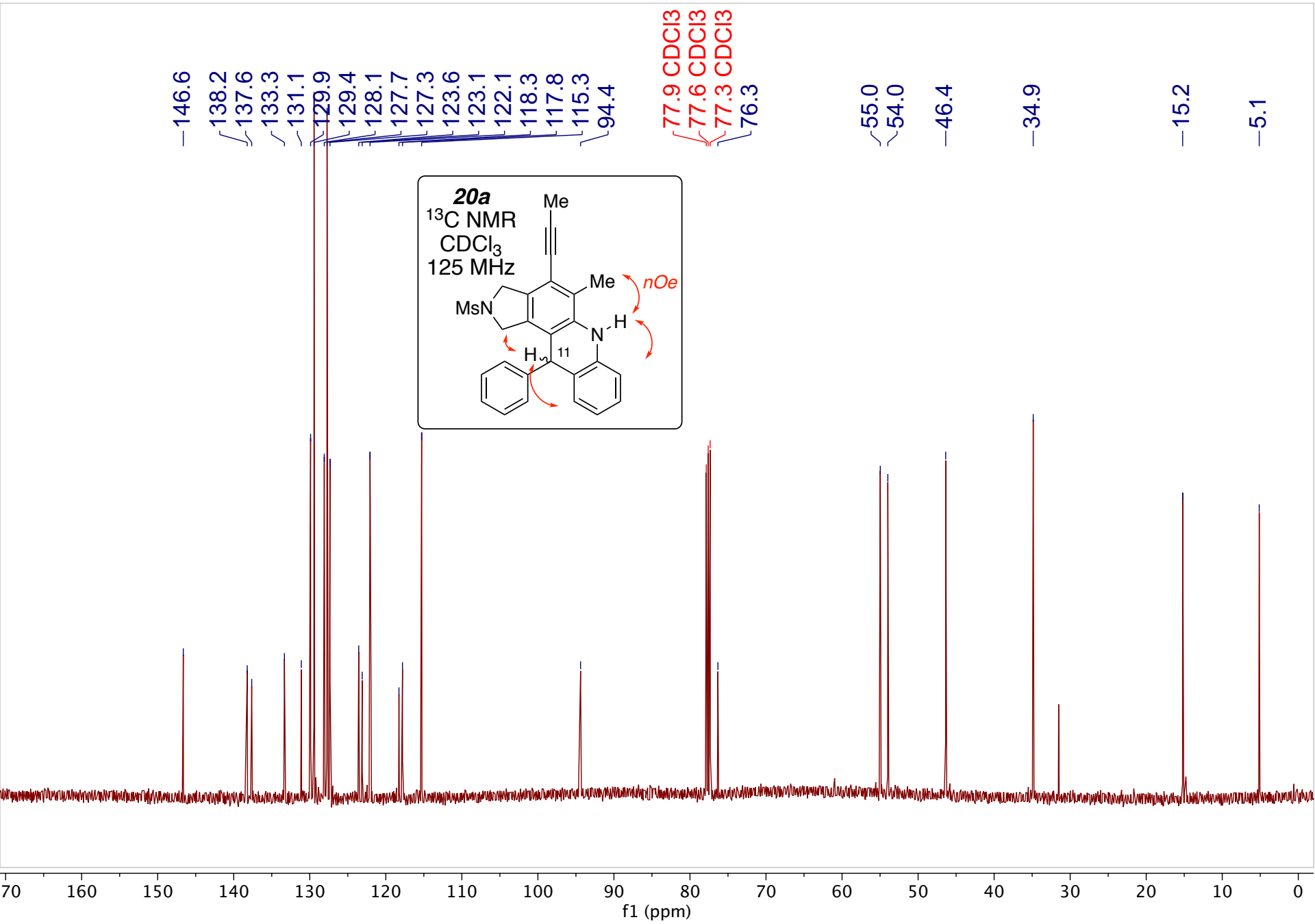

7.26
7.11
7.01
6.93
4.02
3.90
2.81
2.38
0.42
0.00
TMS
O
Me
MeO
NMe2
MeO
17b pure
1H NMR
CDCl3
500 MHz
1.00
1.03
1.02
3.16
3.11
6.46
3.16
9.38
f1 (ppm)

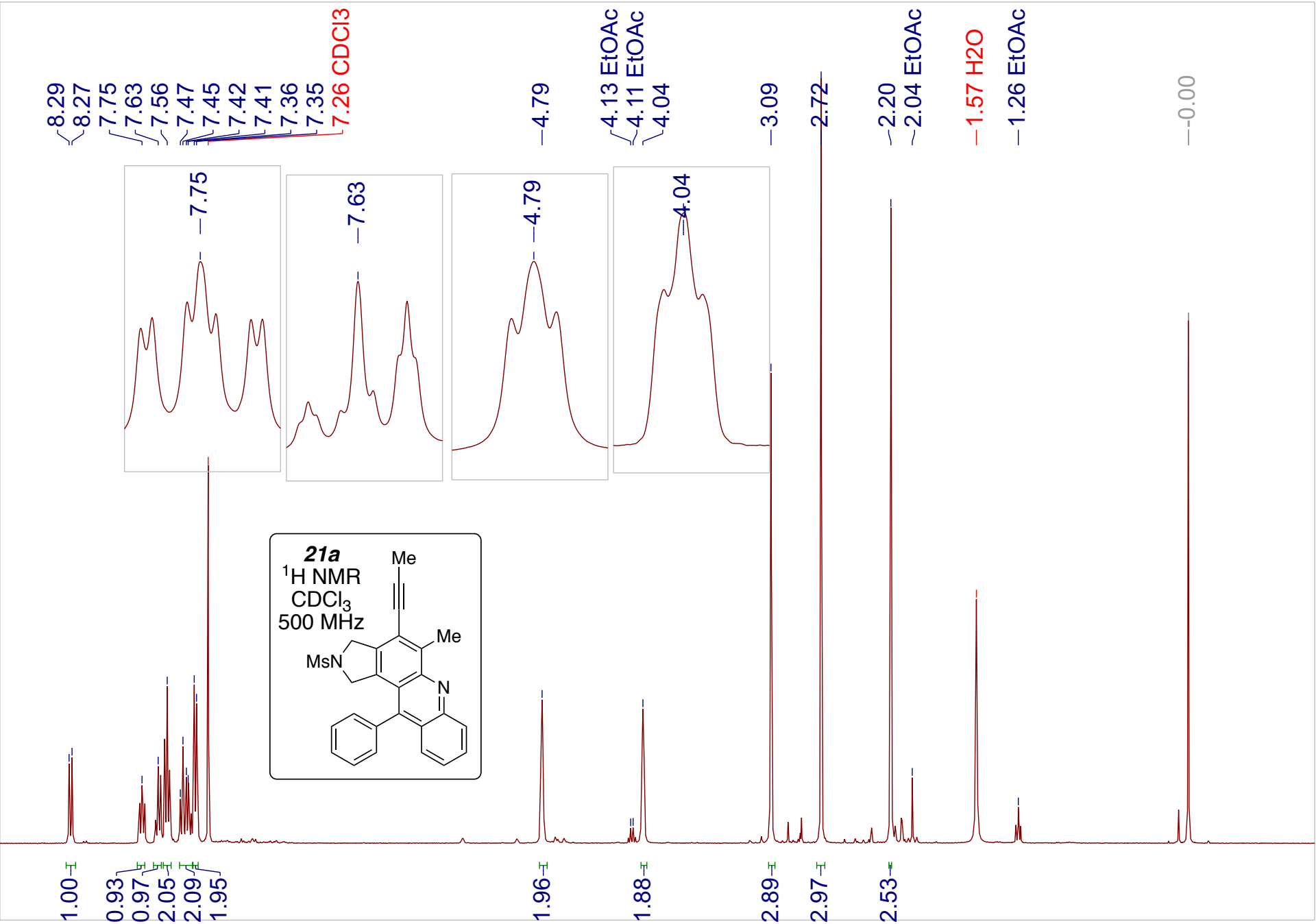

193.9
157.6
154.0
149.5
144.0
143.9
138.6
135.7
133.8
127.7
109.9
106.8
102.6
77.4
77.2
76.9
56.5
56.3
43.9
21.1
2.8
O
TMS
Me
MeO
NMe2
MeO
17b pure
13C NMR
CDCl3
125 MHz
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
-10
f1 (ppm)

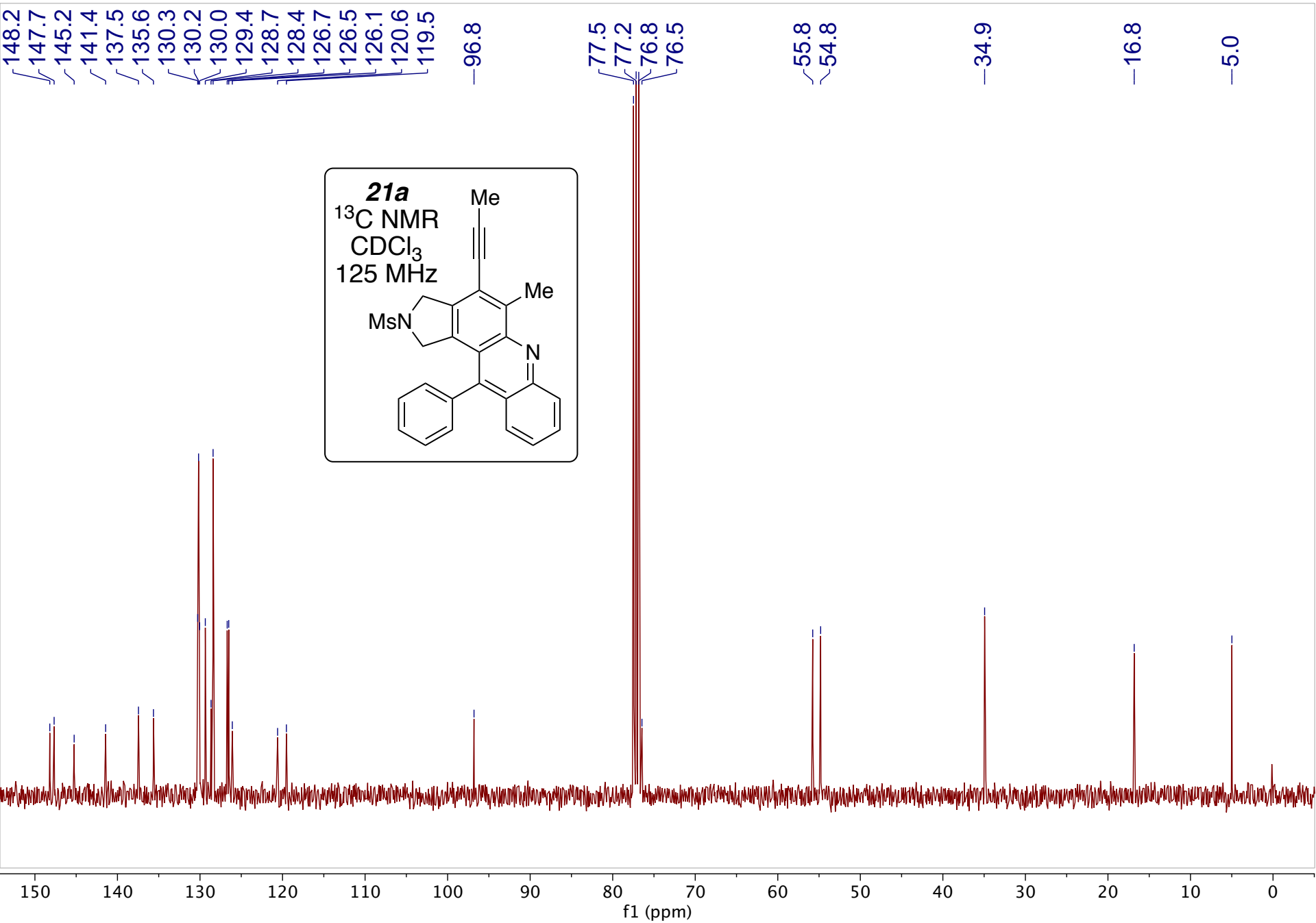

7.26 CDCl3
7.21
7.08
6.84
6.84
6.81
6.80
6.11
5.07
4.67
4.64
4.59
4.57
4.25
4.22
4.13 EtOAc
4.11 EtOAc
2.68
2.45
2.17
2.11 EtOAc
1.26 EtOAc
0.00
20a
1H NMR
CDCl3
500 MHz
Me
Me
nOe
MsN
N
H
H
11
6.84
6.84
7.71
1.01
1.02
1.00
1.04
2.17
1.03
1.00
3.19
3.19
2.37
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
f1 (ppm)

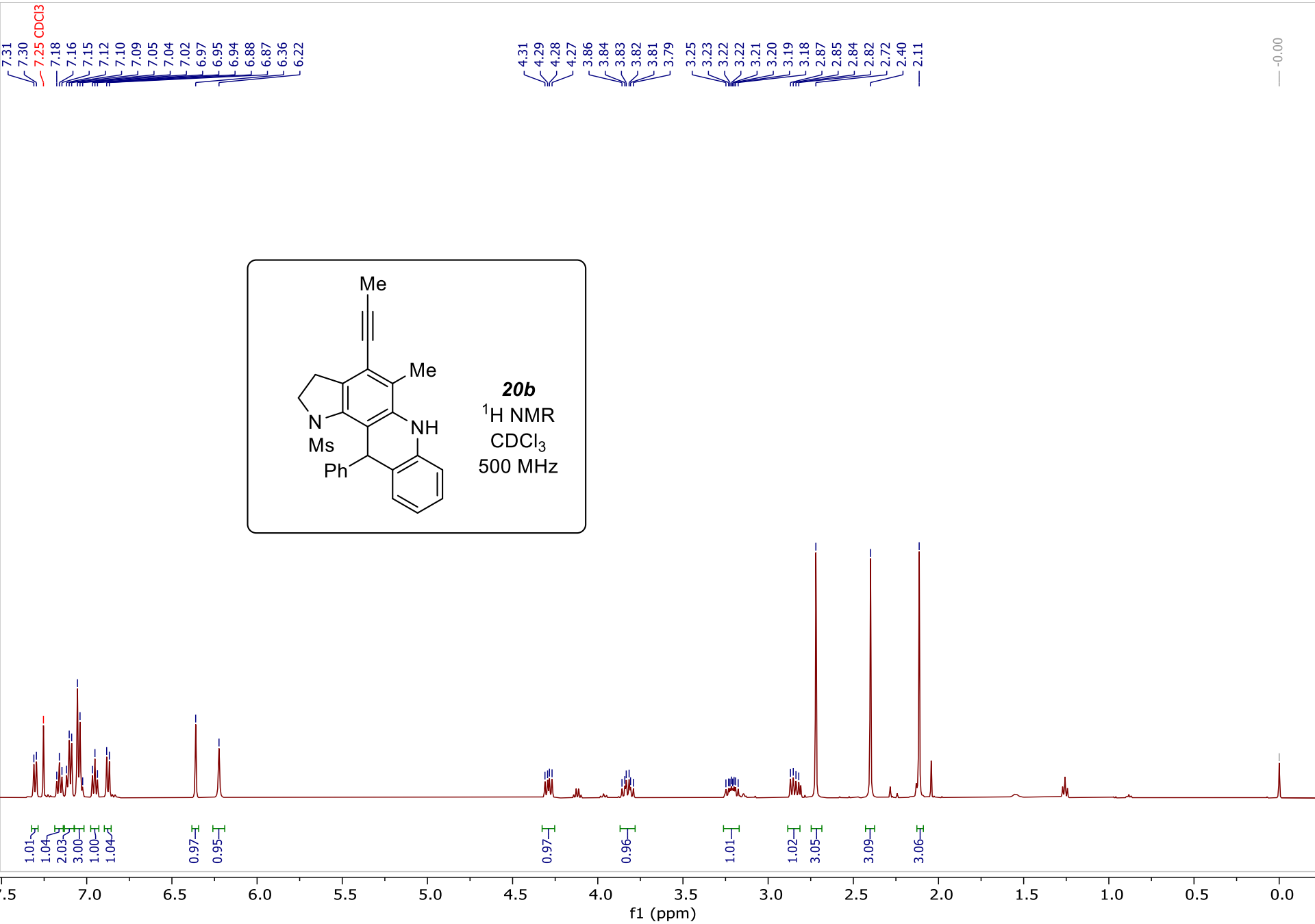

6.81
6.80
6.11
2.45
20a
nOe
Me
Me
nOe
MsN
N
H
H
11
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)

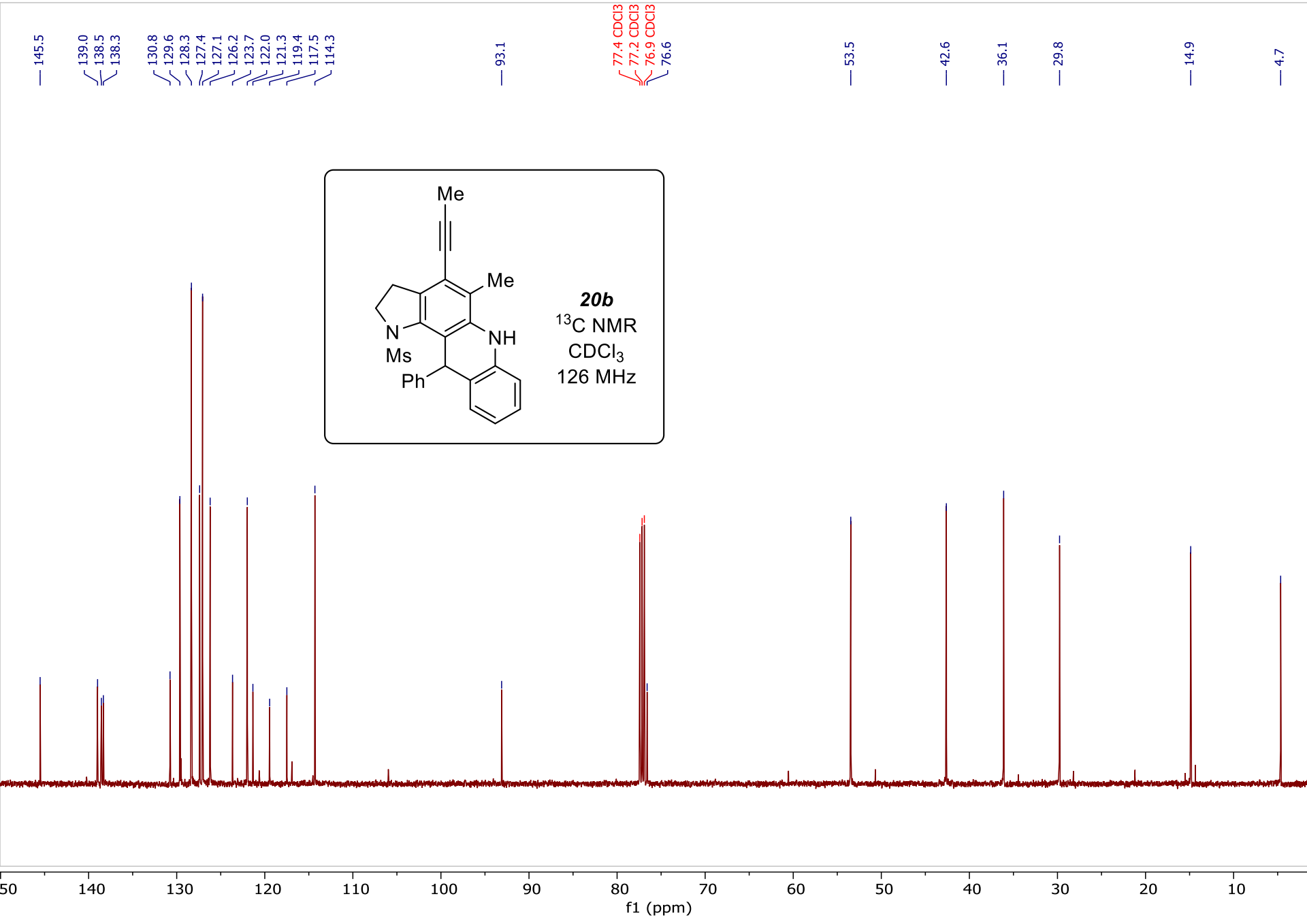

7.14
7.12
5.07
4.66
4.63
4.24
4.22
20a
nOe
Me
Me
MsN
N
H
H
11
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
f1 (ppm)

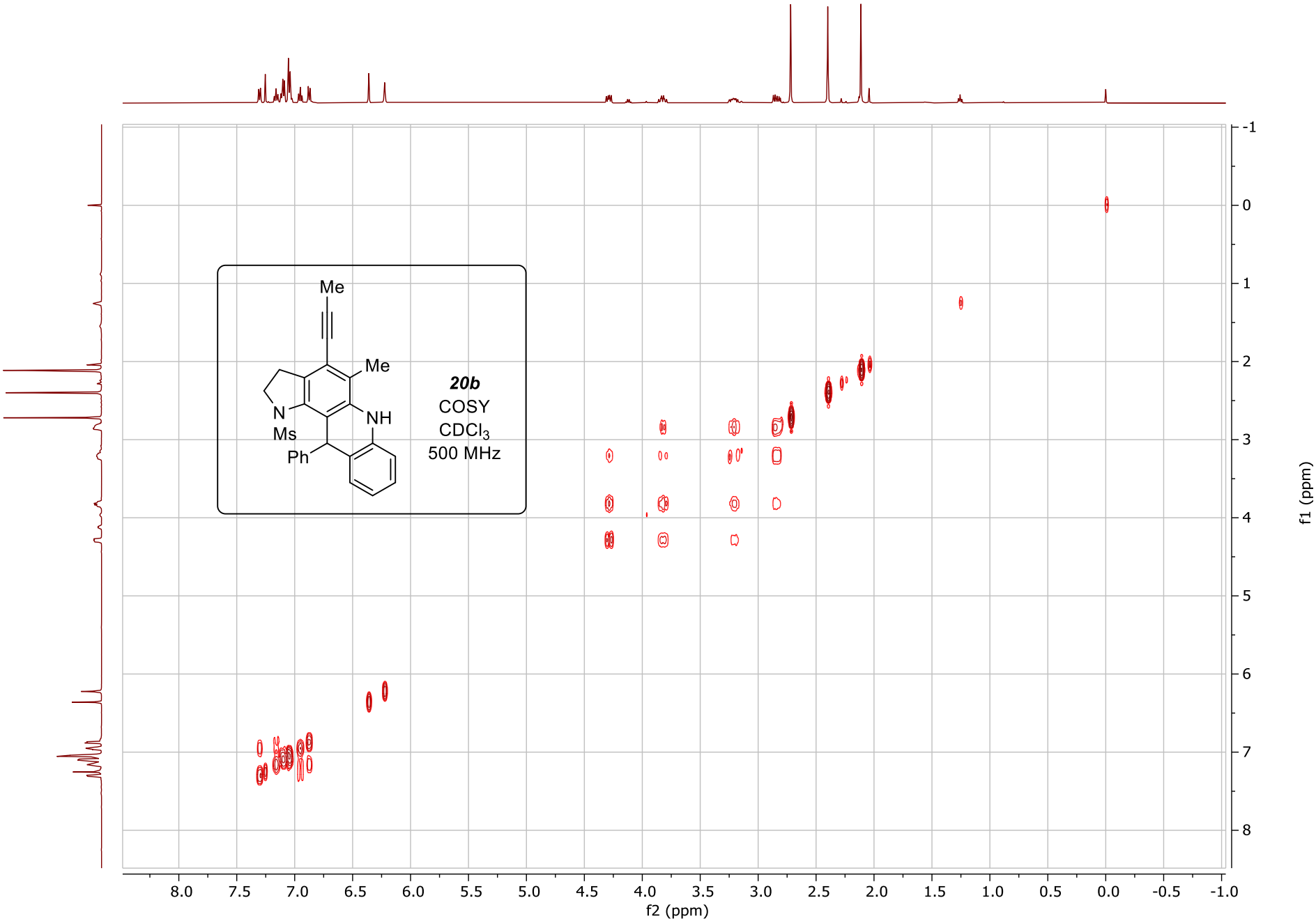

146.6
138.2
137.6
133.3
131.1
129.9
129.4
128.1
127.7
127.3
123.6
123.1
122.1
118.3
117.8
115.3
94.4
77.9 CDCl3
77.6 CDCl3
77.3 CDCl3
76.3
55.0
54.0
46.4
34.9
15.2
5.1
20a
13C NMR
CDCl3
125 MHz
Me
Me
nOe
MsN
N
H
H
11
f1 (ppm)

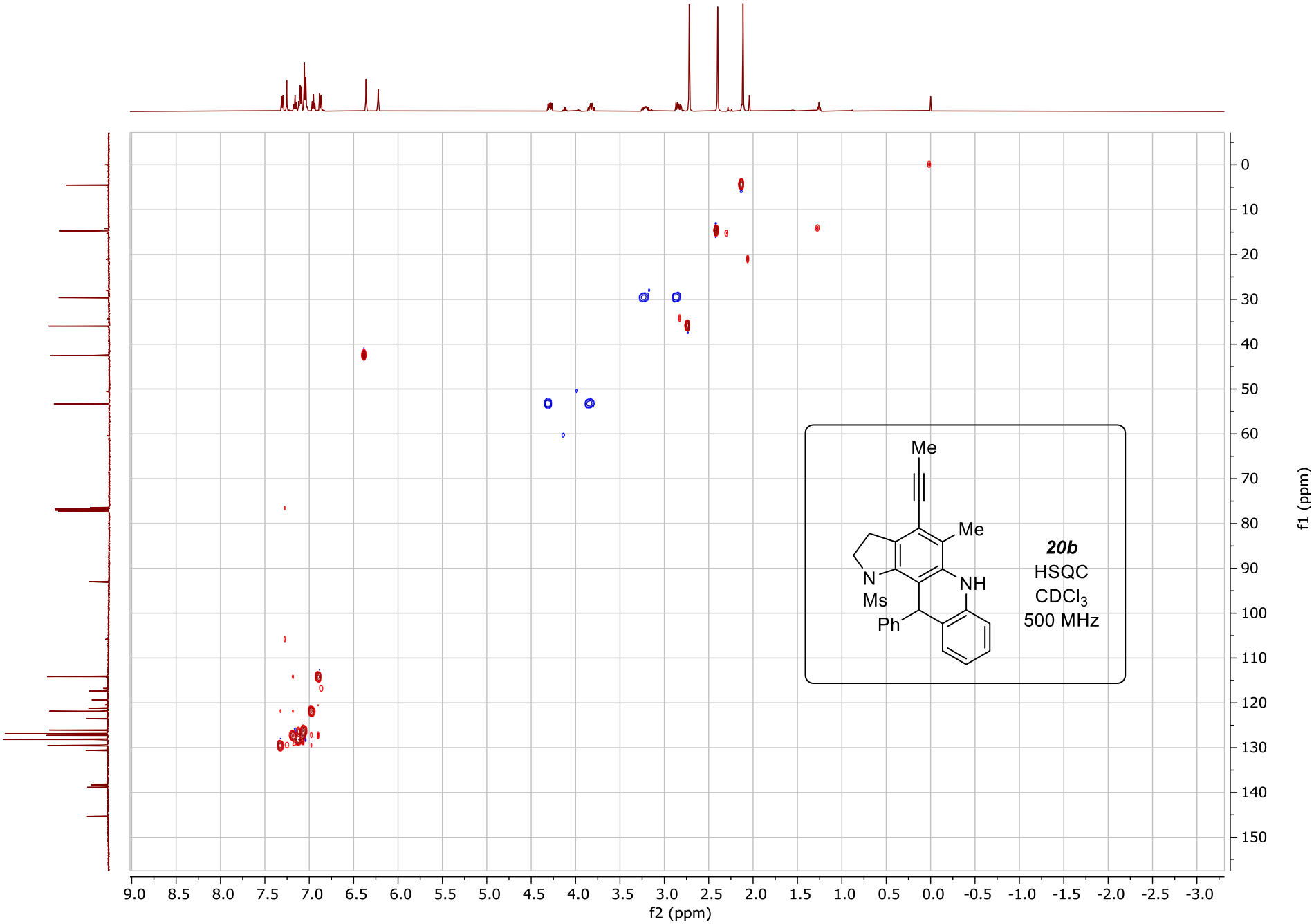

8.29
8.27
7.75
7.63
7.56
7.47
7.45
7.42
7.41
7.36
7.35
7.26 CDCl3
4.79
4.13 EtOAc
4.11 EtOAc
4.04
3.09
2.72
2.20
2.04 EtOAc
1.57 H2O
1.26 EtOAc
0.00
7.75
7.63
4.79
4.04
21a
1H NMR
CDCl3
500 MHz
Me
Me
MsN
N
1.00
0.93
0.97
2.05
2.09
1.95
1.96
1.88
2.89
2.97
2.53

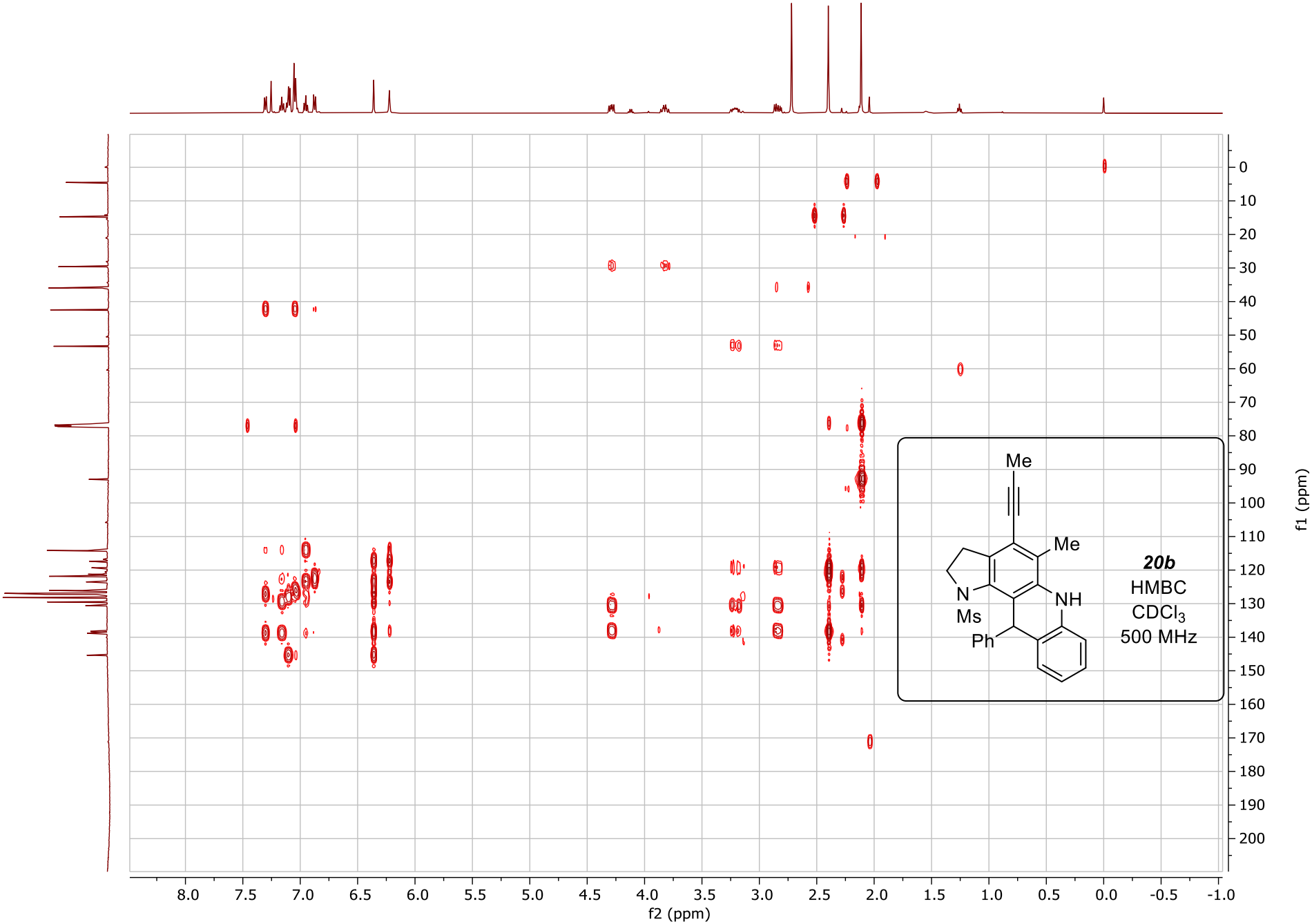

148.2
147.7
145.2
141.4
137.5
135.6
130.3
130.2
130.0
129.4
128.7
128.4
126.7
126.5
126.1
120.6
119.5
96.8
77.5
77.2
76.8
76.5
55.8
54.8
34.9
16.8
5.0
21a
13C NMR
CDCl3
125 MHz
Me
Me
MsN
N
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

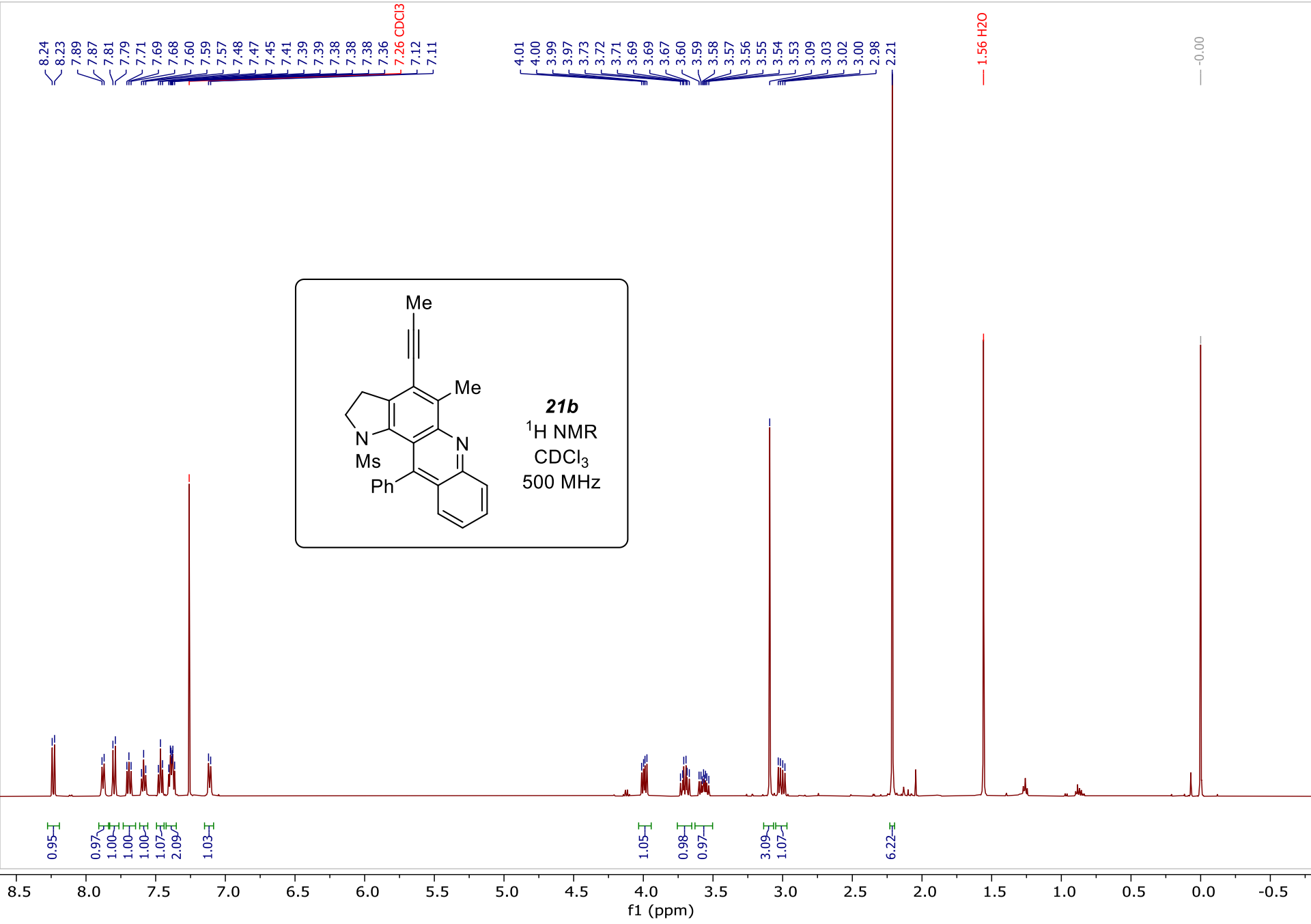

7.31
7.30
7.25 CDCl3
7.18
7.16
7.15
7.12
7.10
7.09
7.05
7.04
7.02
6.97
6.95
6.94
6.88
6.87
6.36
6.22
4.31
4.29
4.28
4.27
3.86
3.84
3.83
3.82
3.81
3.79
3.25
3.23
3.22
3.22
3.21
3.20
3.19
3.18
2.87
2.85
2.84
2.82
2.72
2.40
2.11
-0.00
Me
Me
N
Ms
NH
Ph
20b
1H NMR
CDCl3
500 MHz
1.01
1.04
2.03
3.00
1.00
1.04
0.97
0.95
0.97
0.96
1.01
1.02
3.05
3.09
3.06
f1 (ppm)

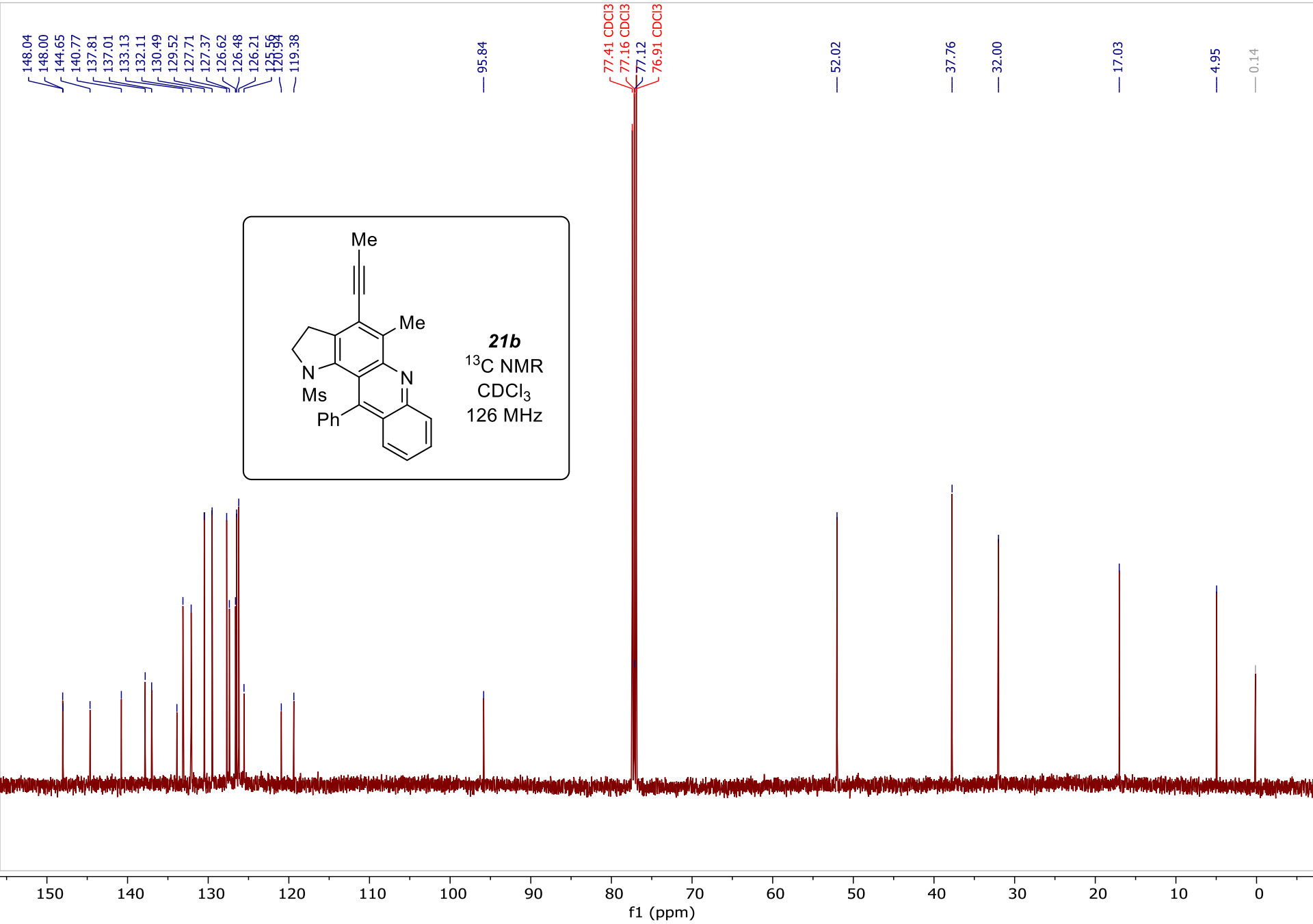

145.5
139.0
138.5
138.3
130.8
129.6
128.3
127.4
127.1
126.2
123.7
122.0
121.3
119.4
117.5
114.3
93.1
77.4 CDCl3
77.2 CDCl3
76.9 CDCl3
76.6
53.5
42.6
36.1
29.8
14.9
4.7
Me
Me
N
Ms
NH
Ph
20b
13C NMR
CDCl3
126 MHz
f1 (ppm)

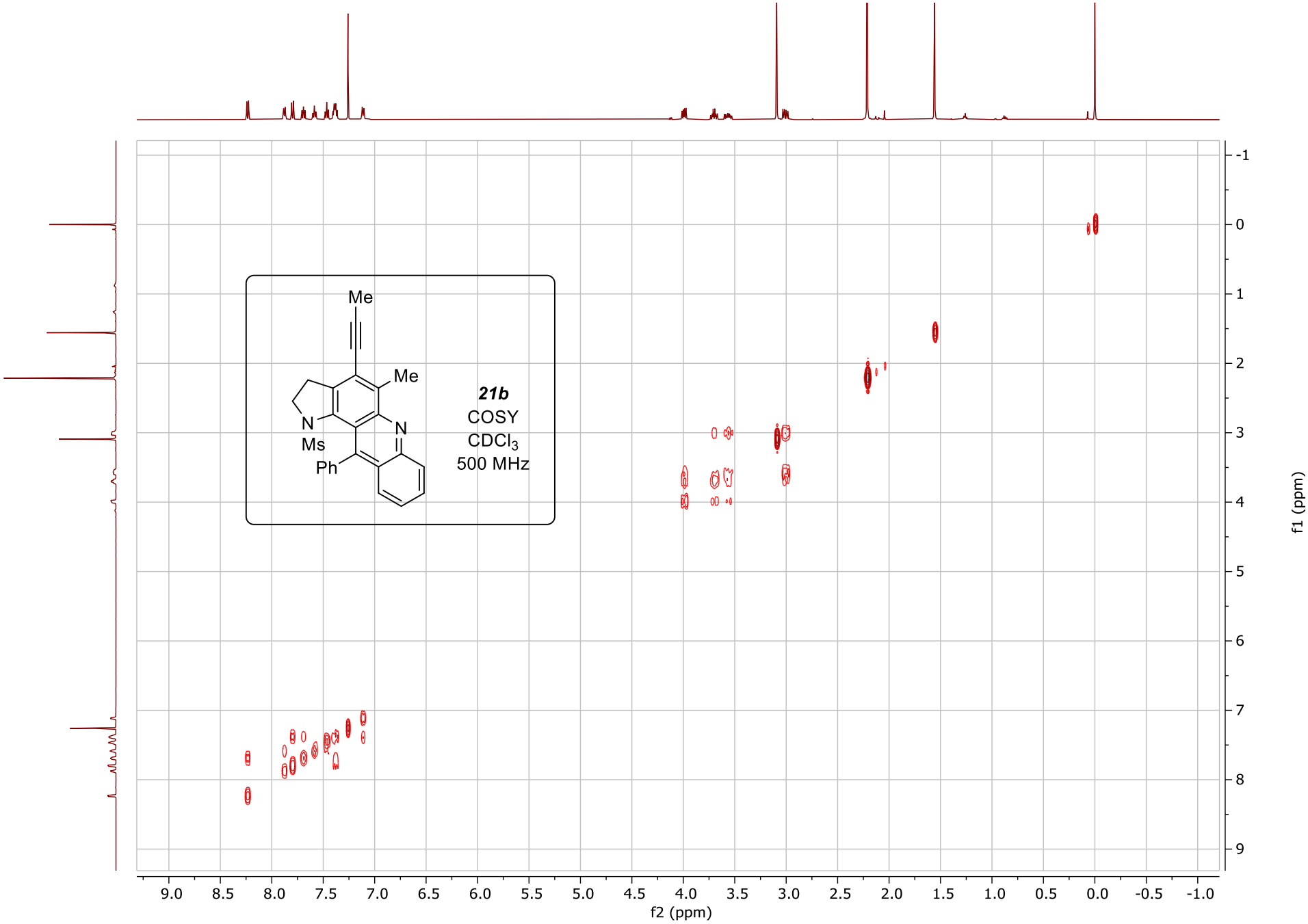

Me
Me
N
Ms
NH
Ph
20b
COSY
CDCl3
500 MHz
f2 (ppm)
f1 (ppm)

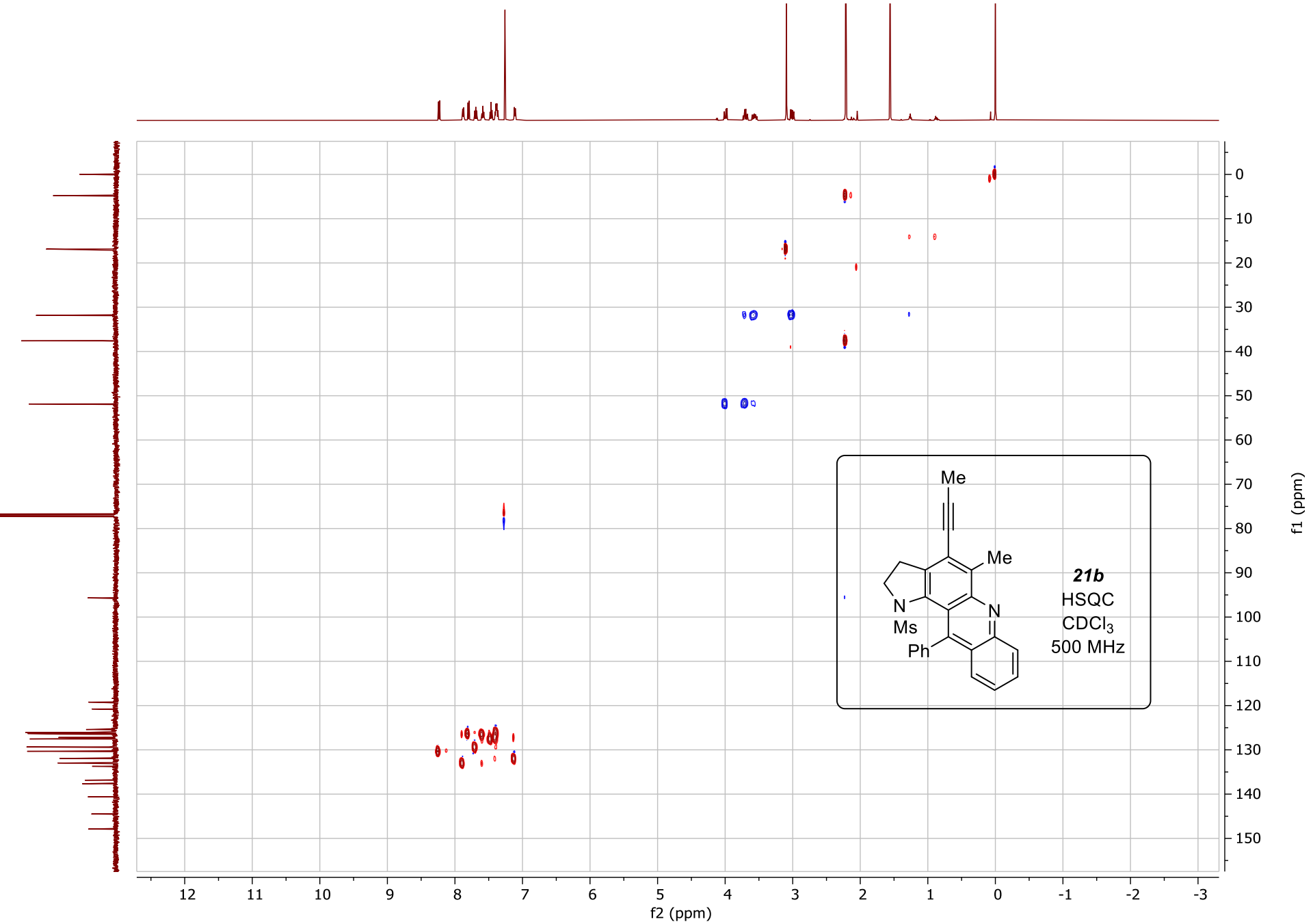

Me
Me
N
Ms
NH
Ph
20b
HSQC
CDCl3
500 MHz
f2 (ppm)
f1 (ppm)

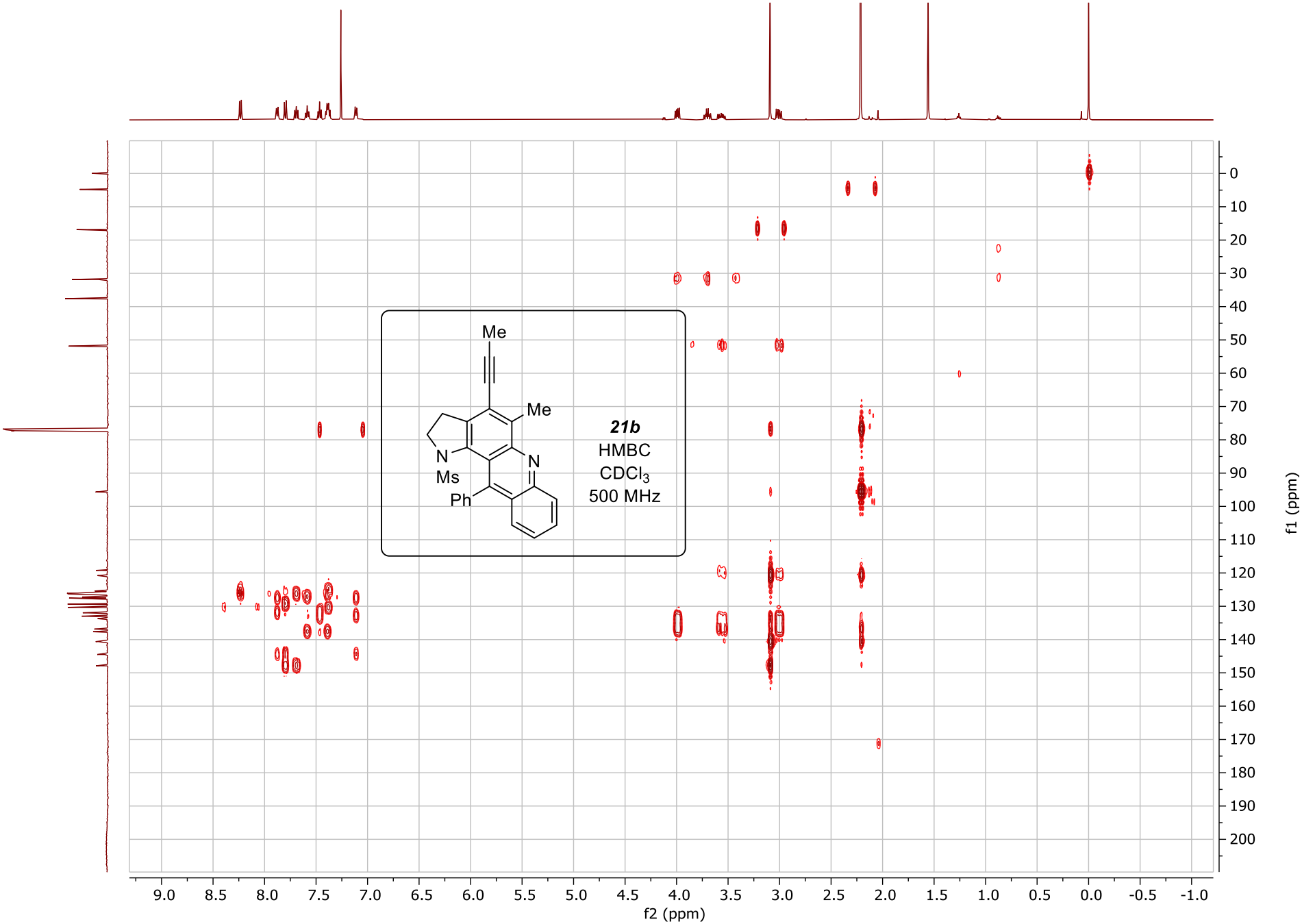

Me
Me
N
NH
Ms
Ph
20b
HMBC
CDCl3
500 MHz
f2 (ppm)
f1 (ppm)


8.24
8.23
7.89
7.87
7.81
7.79
7.71
7.69
7.68
7.60
7.59
7.57
7.48
7.47
7.45
7.41
7.39
7.39
7.38
7.38
7.38
7.36
7.26 CDCl3
7.12
7.11
4.01
4.00
3.99
3.97
3.73
3.72
3.71
3.69
3.69
3.67
3.60
3.59
3.58
3.57
3.56
3.55
3.54
3.53
3.09
3.03
3.02
3.00
2.98
2.21
1.56 H2O
-0.00
Me
Me
N
Ms
N
Ph
21b
1H NMR
CDCl3
500 MHz
0.95
0.97
1.00
1.00
1.00
1.07
2.09
1.03
1.05
0.98
0.97
3.09
1.07
6.22
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
f1 (ppm)


148.04
148.00
144.65
140.77
137.81
137.01
133.13
132.11
130.49
129.52
127.71
127.37
126.62
126.48
126.21
125.56
120.94
119.38
95.84
77.41 CDCl3
77.16 CDCl3
77.12
76.91 CDCl3
52.02
37.76
32.00
17.03
4.95
0.14
Me
Me
N
Ms
N
Ph
21b
13C NMR
CDCl3
126 MHz
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Me
Me
N
Ms
N
Ph
21b
COSY
CDCl3
500 MHz
f2 (ppm)
f1 (ppm)


Me
Me
N
Ms
N
Ph
21b
HSQC
CDCl3
500 MHz
f2 (ppm)
f1 (ppm)


Me
Me
N
Ms
N
Ph
21b
HMBC
CDCl3
500 MHz
f2 (ppm)
f1 (ppm)