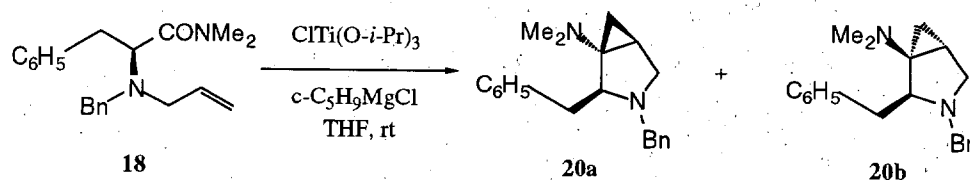


SUPPORTING INFORMATION

General

All solvents were reagent grade and were distilled before use. Tetrahydrofuran (THF) was distilled over sodium and benzophenone. Reagent cyclopentyl magnesium chloride was purchased from Aldrich. Proton magnetic resonance spectra (^1H NMR) and carbon magnetic resonance spectra (^{13}C NMR) were recorded on a Bruker AMX-500 spectrometer. Chemical shifts are in parts per million (ppm) relative to the solvent as the internal reference. Infrared spectra were obtained on a Perkin-Elmer Model 281-B spectrometer. Absorptions were reported in wavenumber (cm^{-1}). Optical rotations (in degrees) were measured with a Perkin-Elmer Model 241 polarimeter. Flash column chromatography was carried out on E. Merck silica gel 60 (240-400 mesh) using the solvent systems listed under individual experiments.

Representative Procedure for the Intramolecular Aminocyclopropanation



To a solution of 2-(allylbenzylamino)-*N,N*-dimethyl-3-phenylpropionamide (**18**, 50mg, 0.155 mmol) in 1.55 mL anhydrous THF was added $\text{ClTi(O-}i\text{-Pr)}_3$ (0.155 mL of 1.0 M solution in hexanes, 1.0 equiv), cyclopentyl magnesium chloride (0.349 mL of 2.0 M solution in diethyl ether, 4.5 equiv) was added dropwise at room temperature over a period of 1 h. The reaction mixture was stirred for an additional 2 h, poured into ice water (5 mL), and stirred for 30 min. THF was evaporated under reduced pressure and 10 mL Et_2O was added to the residue. The organic layer was separated, and the aqueous layer was extracted with ether (3x10 mL). The combined extracts were washed with aqueous NaHCO_3 (10 mL), followed by brine (10 mL), and dried over Na_2SO_4 . Evaporation of the solvent, followed by silica gel column chromatography (acetone-hexanes, 10:90) of the residue afforded 28 mg (60% yield) of **20a** as a light yellow solid, and 11mg (23%) of **20b** as a light yellow oil.

(1*R*,2*S*,6*S*)-2,3-Dibenzyl-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**20a**)

- Mp: 32.8-33.8 °C; R_f 0.2 (ethyl acetate:petroleum ether, 20:80); $[\alpha]_D^{20} +18.4$ ($c=1.0$, CHCl_3); ^1H NMR (500MHz, CDCl_3) δ 7.14-7.32 (m, 10H), 3.81 (d, 1H, $J = 13.3$ Hz), 3.18 (m, 1H), 3.14-3.19 (dd, 1H, $J = 4.2$ Hz, 13.7 Hz), 3.02 (d, 1H, $J = 13.3$ Hz), 2.86 (dd, 1H, $J = 13.7$ Hz, 4.8 Hz), 2.72 (d, 1H, $J = 9.0$ Hz), 2.38 (s, 6H), 2.30 (dd, 1H, $J = 3.9$ Hz, 9.0Hz), 1.24 (m, 1H), 0.89 (dd, 1H, $J = 4.5$ Hz), 0.59 (dd, 1H, $J = 4.5$ Hz, 8.7Hz); ^{13}C NMR (500MHz, CDCl_3) δ 140.4, 139.8, 129.6, 128.4, 128.0, 128.0, 126.6, 125.8, 62.9, 58.3, 55.4, 54.6, 42.2, 38.7, 21.7, 14.5; IR (neat): 3026, 2926.7, 2817.4, 2776.9, 1602.1, 1494.0, 1452.2, 1376.8, 1351.6, 737.2, 697.1 cm^{-1} ; HRMS calcd for $\text{MH}^+ \text{C}_{21}\text{H}_{27}\text{N}_2$, 307.2174; found, 307.2160.

(1S,2S,6R)-2,3-Dibenzyl-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**20b**)

- R_f 0.2 (ethyl acetate:petroleum ether, 5:95); $[\alpha]_D^{20}$ -48.0 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.07-7.23 (m, 10H), 3.68 (d, 1H, J = 13.8 Hz), 3.55 (d, 1H, J = 13.8 Hz), 3.42 (dd, 1H, J = 4.7 Hz, 6.4 Hz), 3.10 (dd, 1H, J = 13.8 Hz, 4.5 Hz), 2.79 (dd, 1H, J = 3.2 Hz, 9.3 Hz), 2.73 (dd, 1H, J = 6.4 Hz, 13.8Hz), 2.51 (d, 1H, J = 9.3 Hz), 2.12 (s, 6H), 1.55-1.58 (m, 1H), 0.72 (dd, 1H, J = 4.2 Hz, 8.5 Hz), 0.63 (dd, 1H, J = 4.0 Hz); ¹³C NMR (500MHz, CDCl₃) δ 142.5, 140.5, 129.6, 128.2, 127.9, 127.8, 126.4, 125.0, 68.0, 57.0, 55.8, 51.9, 42.1, 34.5, 22.8, 8.45; IR (neat): 3025.2, 2936.1, 2817.4, 2774.8; 1602.1, 1493.4, 1452.7, 1348.6, 730.7, 697.3 cm⁻¹; HRMS calcd for MH⁺ C₂₁H₂₇N₂, 307.2174; found, 307.2160.

(1R,2S,6S)-3-Benzyl-2-(4-hydroxybenzyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**22a**)

- Mp: 143-144 °C; R_f 0.35 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ +14.4 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.15-7.29 (m, 7H), 6.70 (d, 2H, J = 8.1 Hz), 5.88 (br, s, 1H), 3.84 (d, 1H, J = 13.3 Hz), 3.16 (m, 1H), 3.02-3.10 (m, 2H), 2.81 (d, 1H, J = 10.5 Hz), 2.72 (d, 1H, J = 8.4 Hz), 2.38 (s, 6H), 2.31 (m, 1H), 1.28 (m, 1H), 0.89 (m, 1H), 0.62 (d, 1H, J = 3.0 Hz); ¹³C NMR (500MHz, CDCl₃) δ 154.0, 139.5, 132.1, 130.6, 128.5, 128.1, 126.6, 115.2, 62.8, 58.3, 55.4, 54.5, 42.3, 37.7, 21.9, 14.5; IR (neat): 3300 (br), 2930.5, 2785.1, 1612.6; 1514.3, 1453.5, 1240.1, 1170.0, 827.1, 751.7, 699.2 cm⁻¹; HRMS calcd for MH⁺ C₂₁H₂₇N₂O, 323.2123; found, 323.2111.

(1S,2S,6R)-3-Benzyl-2-(4-hydroxybenzyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**22b**)

- R_f 0.55 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ -34.2 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.08-7.24 (m, 7H), 6.67 (d, 2H, J = 8.4 Hz), 4.91 (br, s, 1H), 3.68 (d, 1H, J = 13.8 Hz), 3.56 (d, 1H, J = 13.8 Hz), 3.35 (m, 1H), 3.01 (dd, 1H, J = 13.7 Hz, 3.6 Hz), 2.79 (dd, 1H, J = 6.9 Hz, 8.7 Hz), 2.68 (dd, 1H, J = 6.1 Hz, 13.6 Hz), 2.49 (d, 1H, J = 9.4 Hz), 2.13 (s, 6H), 1.56 (m, 1H), 0.74 (dd, 1H, J = 3.6 Hz, 7.7Hz), 0.59 (m, 1H); ¹³C NMR (500MHz, CDCl₃) δ 153.2, 140.5, 134.4, 130.7, 128.2, 128.0, 126.4, 114.7, 68.2, 57.3, 55.8, 52.0, 42.0, 33.8, 22.9, 8.73; IR (neat): 3318.8 (br), 2934.8, 2817.8, 2776.1, 1612.3, 1513.5, 1453.3, 1352.2, 1243.2, 1170.4, 828.5, 740.0, 698.4 cm⁻¹; HRMS calcd for MH⁺ C₂₁H₂₇N₂O, 323.2123; found, 323.2118.

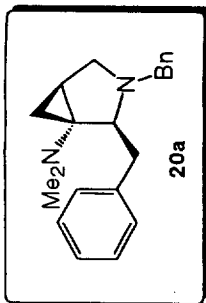
(1R,2S,6S)-3-Benzyl-2-(1H-indol-3-ylmethyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**27a**)

- R_f 0.35 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ -9.8 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.97 (br, s, 1H), 7.72 (d, 1H, J = 7.7 Hz), 7.30 (d, 1H, J = 7.8 Hz), 7.10-7.23 (m, 7H), 7.05 (d, 1H, J = 1.9 Hz), 3.80 (d, 1H, J = 13.3 Hz), 3.36 (dd, 1H, J = 4.1 Hz, 6.1 Hz), 3.30 (dd, 1H, J = 4.0 Hz, 15.3 Hz), 2.98-3.03 (m, 2H), 2.77 (d, 1H, J = 9.0 Hz), 2.42 (s, 6H), 2.33 (dd, 1H, J = 3.9 Hz, 9.0Hz), 1.30 (m, 1H), 1.09 (dd, 1H, J = 4.3 Hz), 0.65 (dd, 1H, J = 4.4 Hz, 8.6Hz); ¹³C NMR (500MHz, CDCl₃) δ 140.0, 136.3, 128.4, 127.9, 127.9, 126.5, 122.1, 121.8, 119.3, 119.1, 114.8, 111.0, 61.2, 58.8, 56.0, 54.9, 42.2, 28.9, 22.4, 14.5; IR (neat): 3420.3, 3058.2, 2922.9, 2783.6, 1619.7, 1493.8, 1477.5, 1453.8, 1353.2, 738.7, 699.2 cm⁻¹; HRMS calcd for M⁺ C₂₃H₂₇N₃, 345.2205; found, 345.2203.

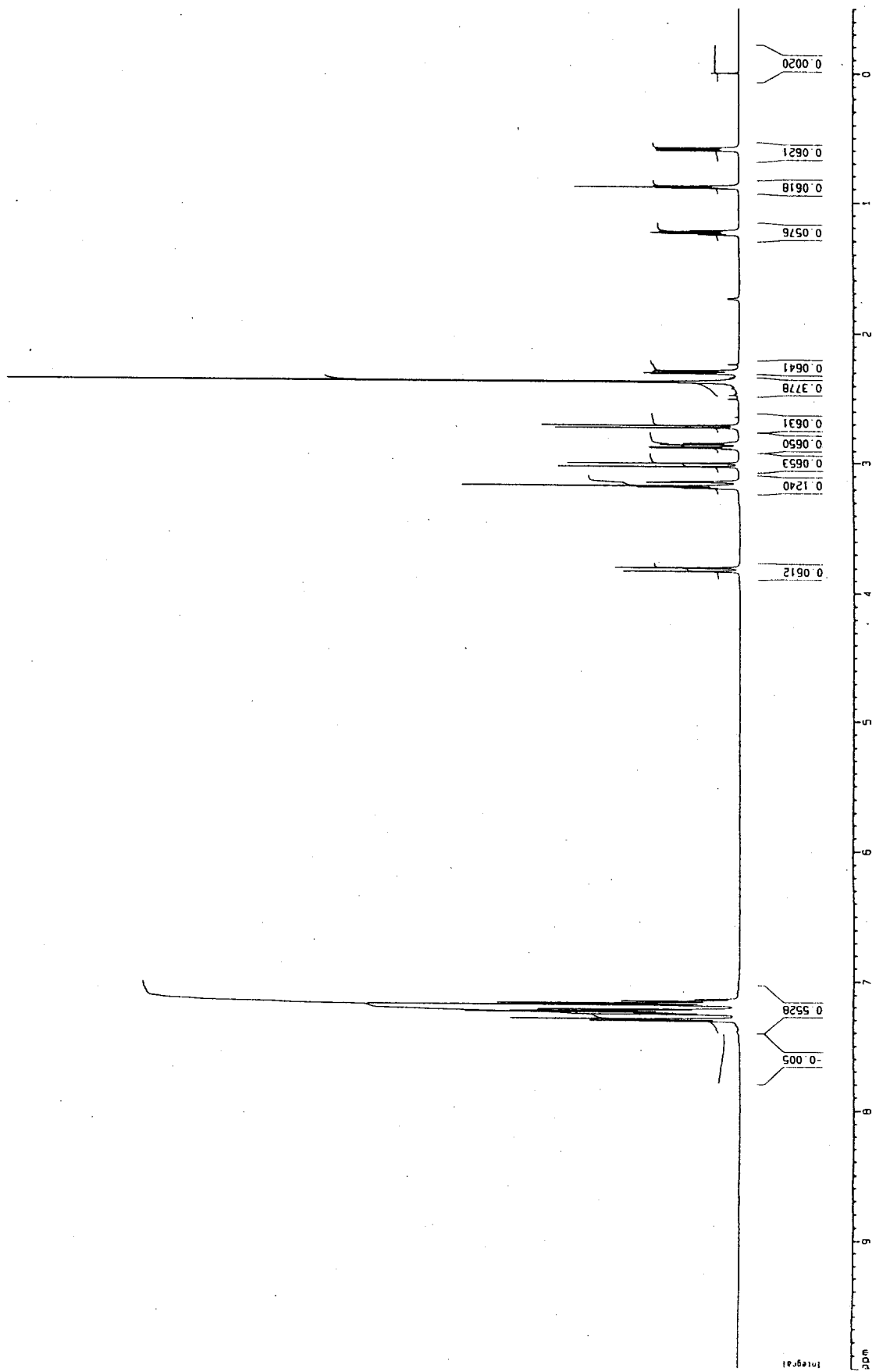
(1S,2S,6R)-3-Benzyl-2-(1H-indol-3-ylmethyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**27b**)

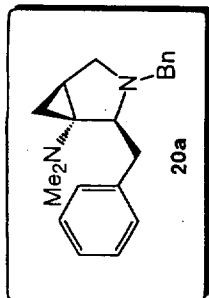
- R_f 0.55 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ -30.3 ($c=1.0$, CHCl_3); ^1H NMR (500MHz, CDCl_3) δ 7.81 (br, s, 1H), 7.60 (d, 1H, $J = 7.8$ Hz), 7.28 (d, 1H, $J = 8.0$ Hz), 7.06-7.21 (m, 7H), 6.92 (d, 1H, $J = 1.7$ Hz), 3.73 (d, 1H, $J = 14.0$ Hz), 3.56 (d, 1H, $J = 14.0$ Hz), 3.51 (dd, 1H, $J = 4.0$ Hz, 6.9 Hz), 3.29 (dd, 1H, $J = 3.9$ Hz, 14.8 Hz), 2.91 (dd, 1H, $J = 3.2$ Hz, 9.3 Hz), 2.84 (dd, 1H, $J = 7.0$ Hz, 14.8 Hz), 2.50 (d, 1H, $J = 9.3$ Hz), 2.21 (s, 6H), 1.61 (m, 1H), 0.77 (dd, 1H, $J = 4.2$ Hz, 8.5 Hz), 0.62 (dd, 1H, $J = 4.0$ Hz); ^{13}C NMR (500MHz, CDCl_3) δ 140.9, 136.1, 128.1, 128.0, 127.9, 126.3, 122.5, 121.4, 119.2, 118.8, 115.9, 110.9, 67.2, 57.2, 55.9, 52.2, 42.4, 23.8, 23.0, 9.34; IR (neat): 3421.1, 3057.1, 2935.8, 2817.0, 2774.6, 1617.9, 1492.5, 1454.1, 1351.1, 738.1, 698.0 cm^{-1} ; HRMS calcd for MH^+ $\text{C}_{23}\text{H}_{28}\text{N}_3$, 346.2283; found, 346.2273.

HS



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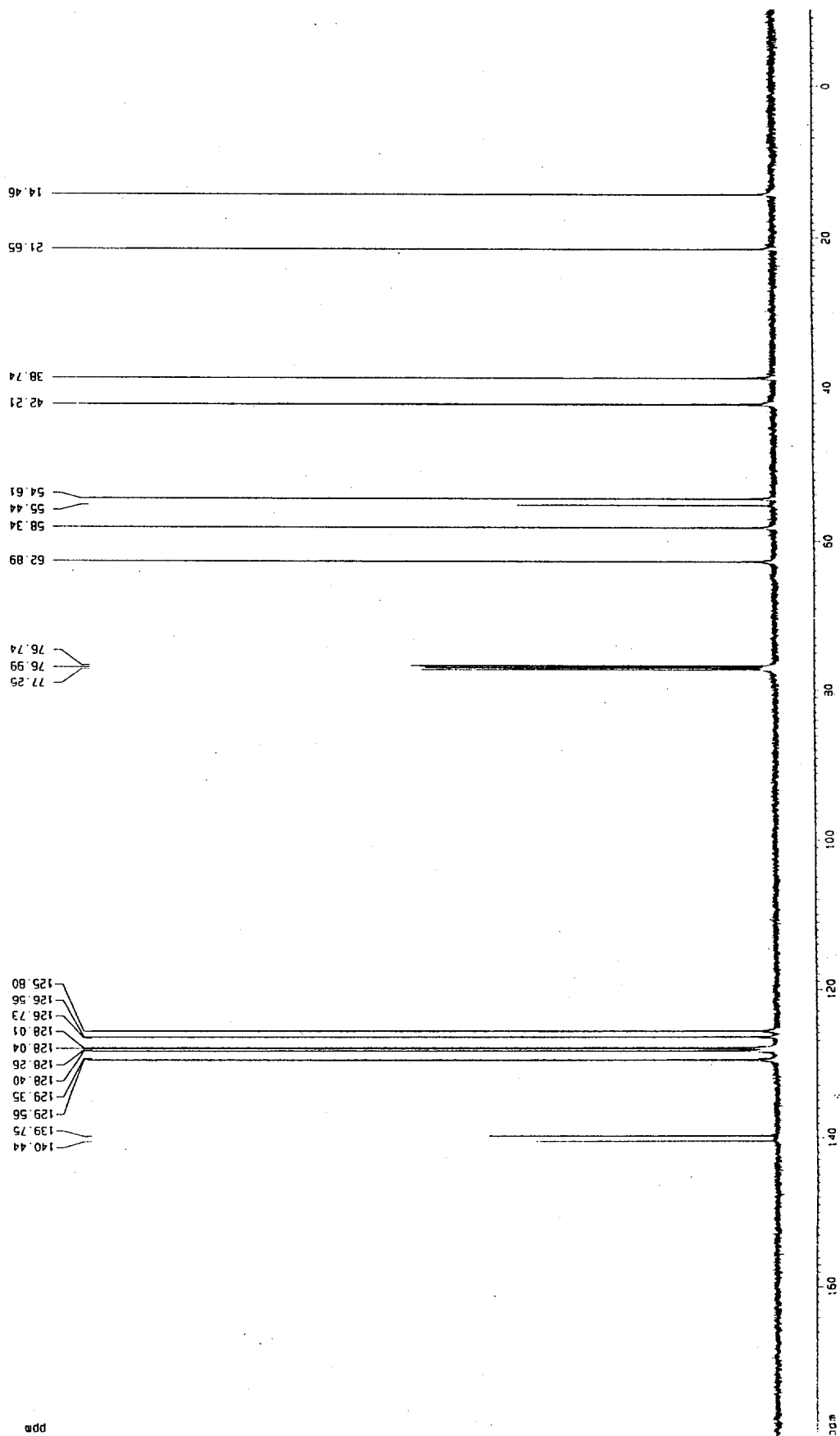


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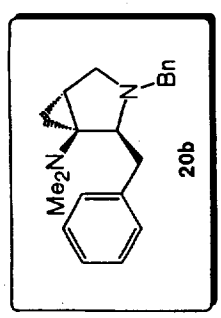
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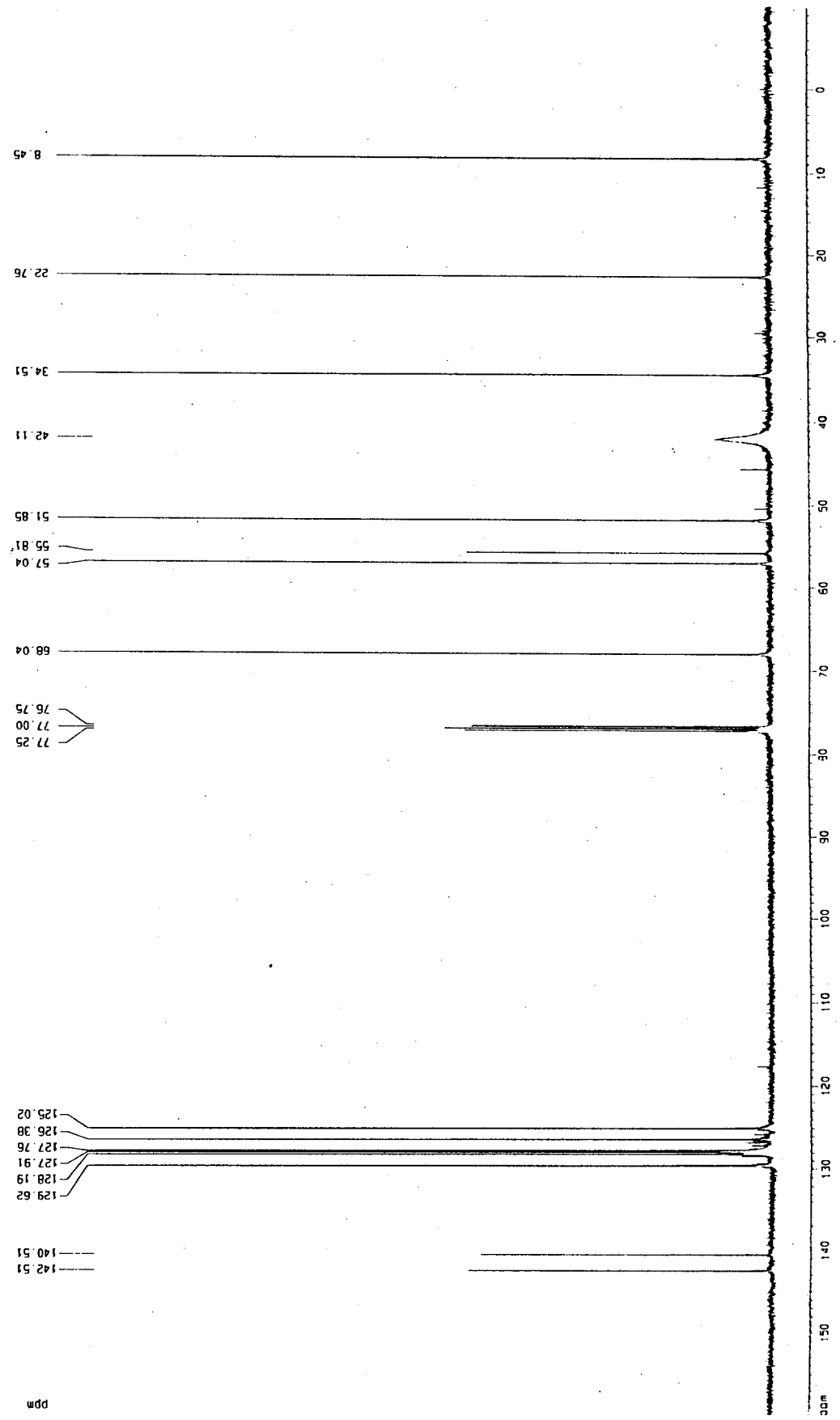
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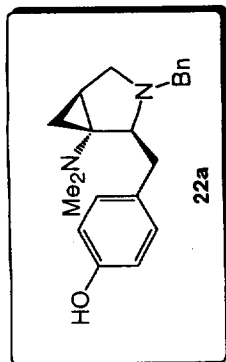


¹³C-NMR



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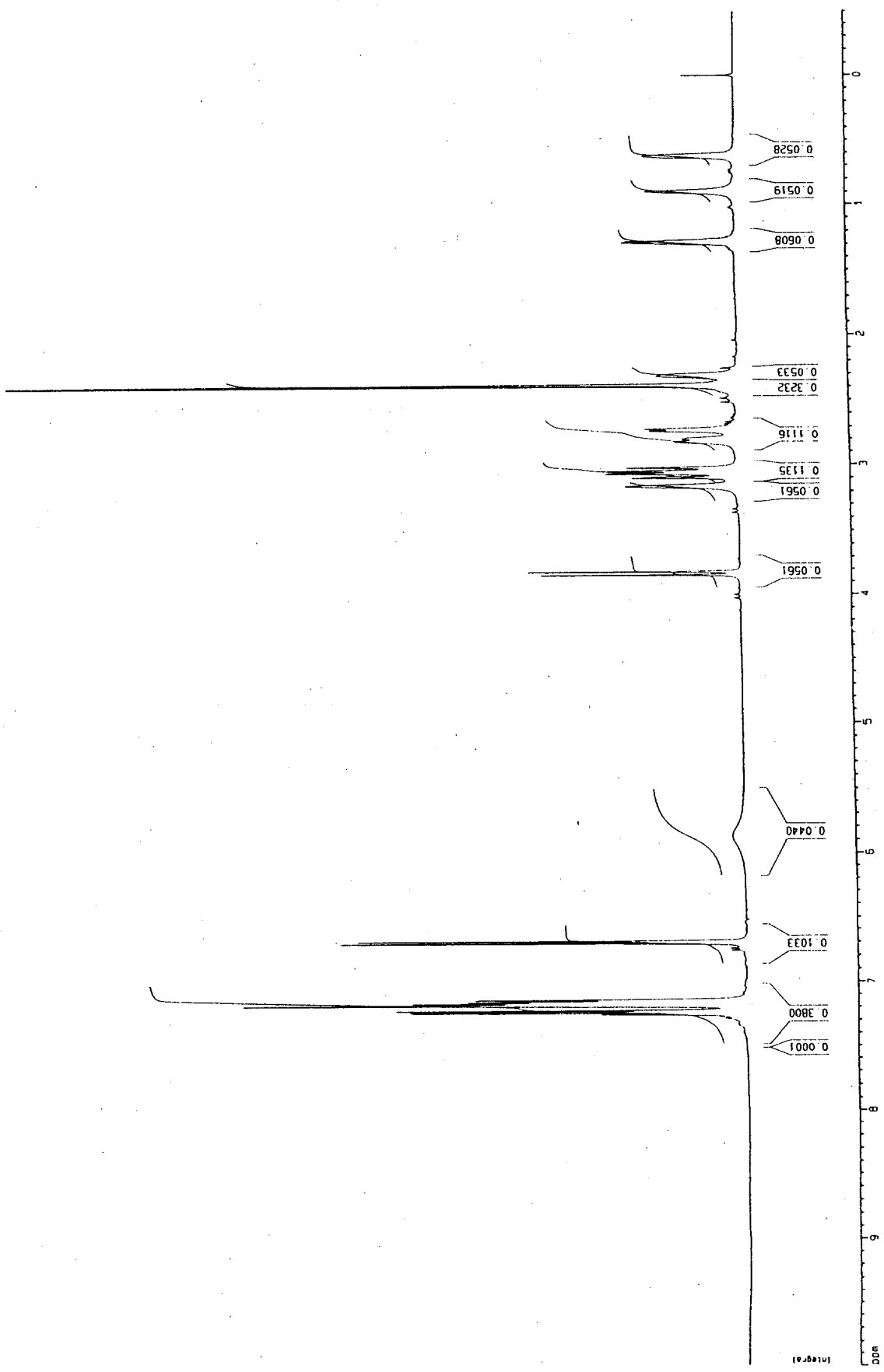


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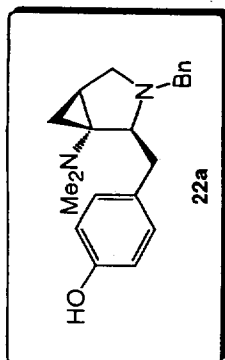
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65

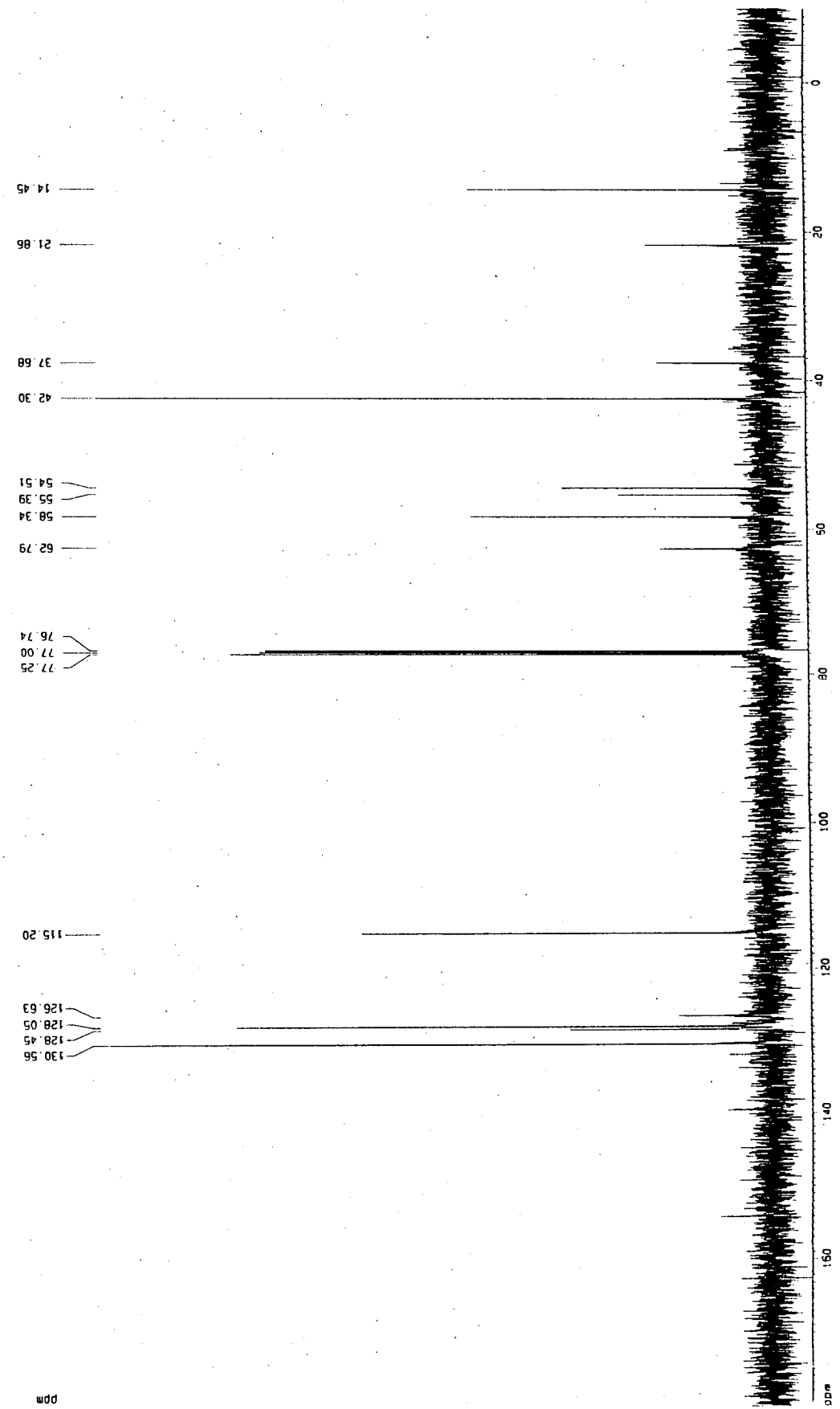


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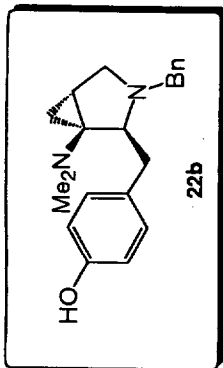
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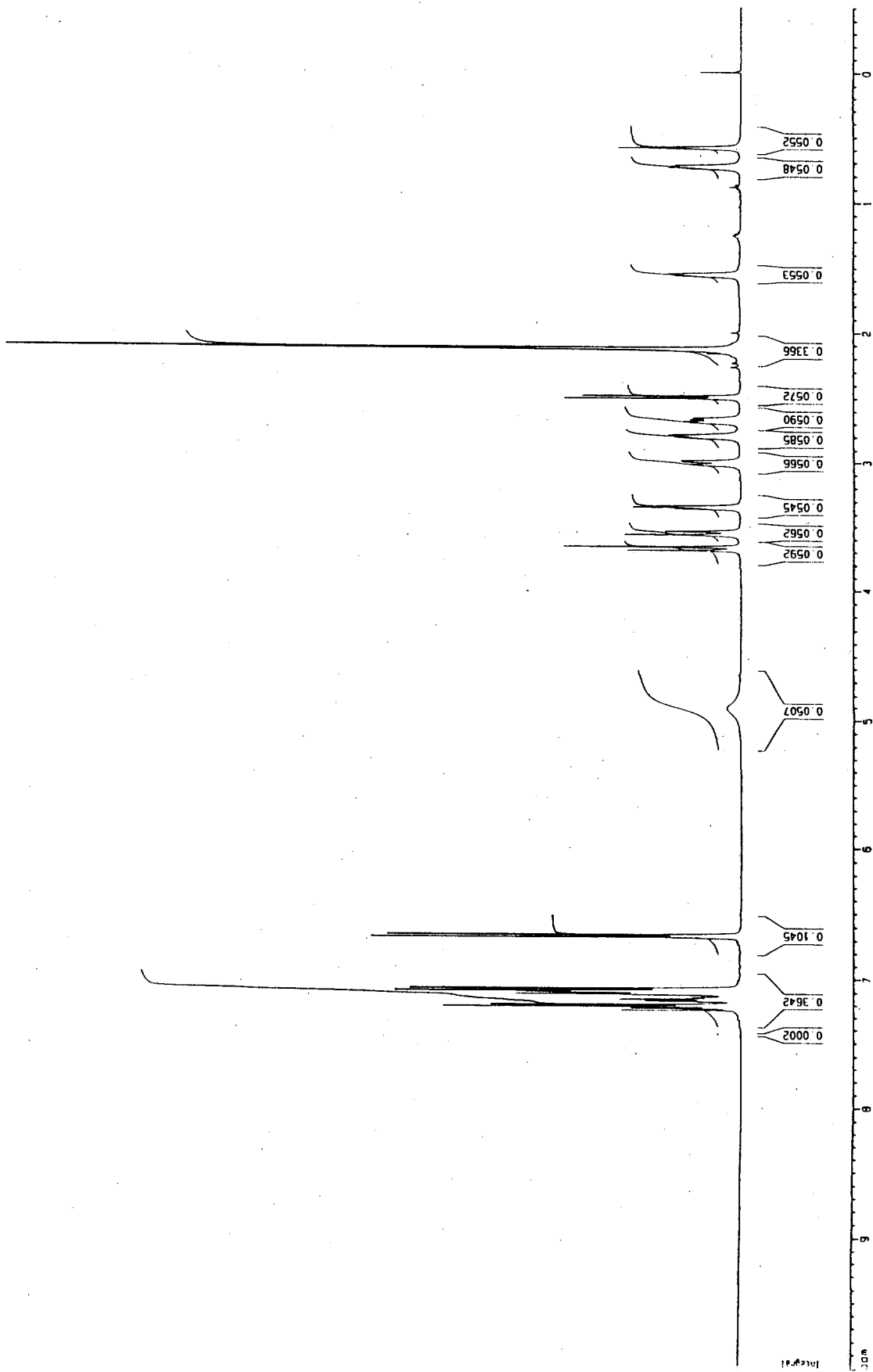
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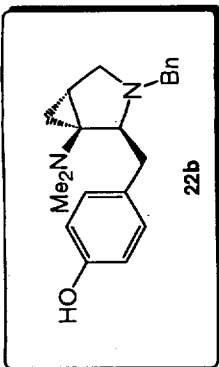


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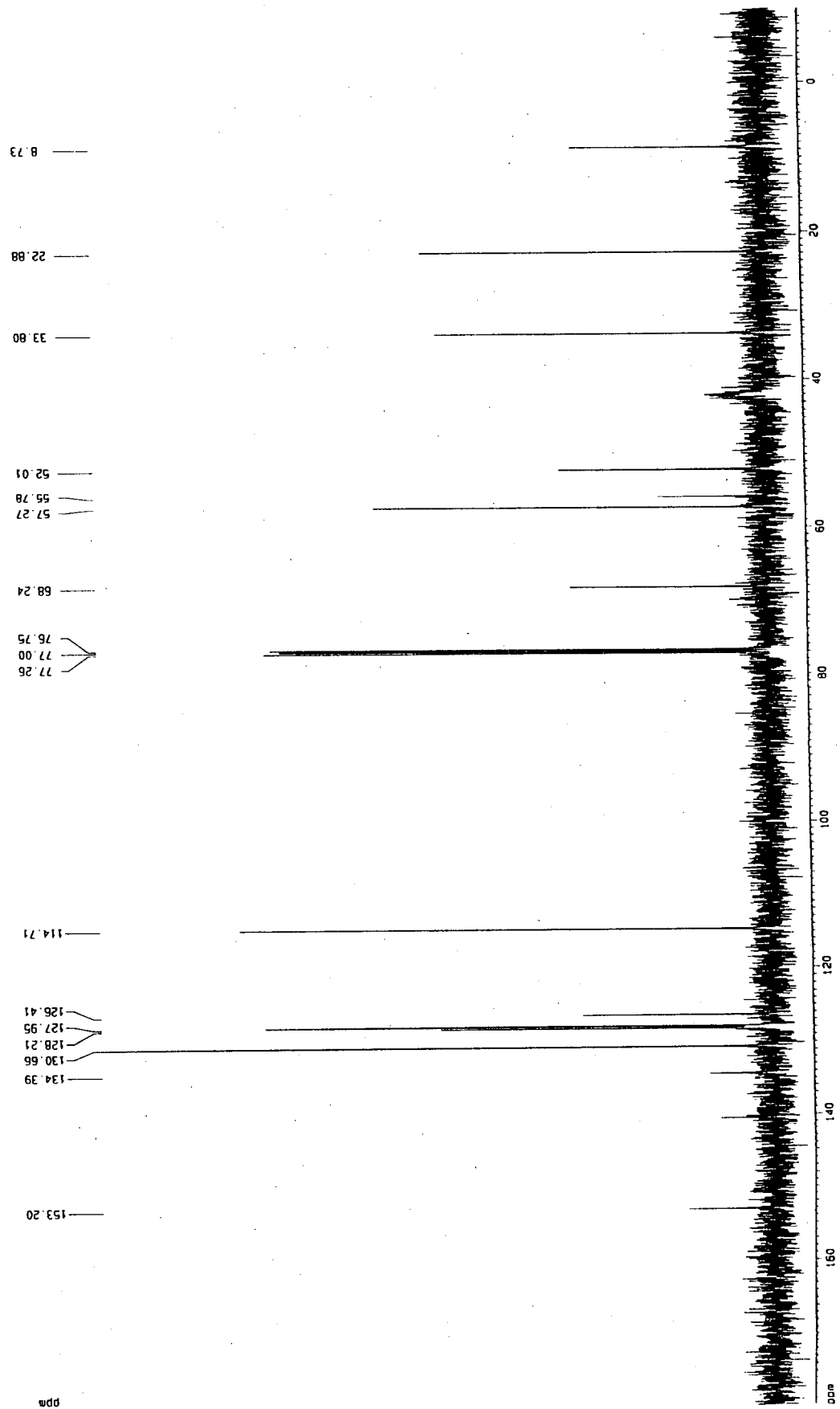
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 F19 118.370000000 MHz
 F20 118.370000000 MHz
 F21 118.370000000 MHz
 F22 118.370000000 MHz
 F23 118.370000000 MHz
 F24 118.370000000 MHz
 F25 118.370000000 MHz
 F26 118.370000000 MHz
 F27 118.370000000 MHz
 F28 118.370000000 MHz
 F29 118.370000000 MHz
 F30 118.370000000 MHz
 F31 118.370000000 MHz
 F32 118.370000000 MHz
 F33 118.370000000 MHz
 F34 118.370000000 MHz
 F35 118.370000000 MHz
 F36 118.370000000 MHz
 F37 118.370000000 MHz
 F38 118.370000000 MHz
 F39 118.370000000 MHz
 F40 118.370000000 MHz
 F41 118.370000000 MHz
 F42 118.370000000 MHz
 F43 118.370000000 MHz
 F44 118.370000000 MHz
 F45 118.370000000 MHz
 F46 118.370000000 MHz
 F47 118.370000000 MHz
 F48 118.370000000 MHz
 F49 118.370000000 MHz
 F50 118.370000000 MHz
 F51 118.370000000 MHz
 F52 118.370000000 MHz
 F53 118.370000000 MHz
 F54 118.370000000 MHz
 F55 118.370000000 MHz
 F56 118.370000000 MHz
 F57 118.370000000 MHz
 F58 118.370000000 MHz
 F59 118.370000000 MHz
 F60 118.370000000 MHz
 F61 118.370000000 MHz
 F62 118.370000000 MHz
 F63 118.370000000 MHz
 F64 118.370000000 MHz
 F65 118.370000000 MHz
 F66 118.370000000 MHz
 F67 118.370000000 MHz
 F68 118.370000000 MHz
 F69 118.370000000 MHz
 F70 118.370000000 MHz
 F71 118.370000000 MHz
 F72 118.370000000 MHz
 F73 118.370000000 MHz
 F74 118.370000000 MHz
 F75 118.370000000 MHz
 F76 118.370000000 MHz
 F77 118.370000000 MHz
 F78 118.370000000 MHz
 F79 118.370000000 MHz
 F80 118.370000000 MHz
 F81 118.370000000 MHz
 F82 118.370000000 MHz
 F83 118.370000000 MHz
 F84 118.370000000 MHz
 F85 118.370000000 MHz
 F86 118.370000000 MHz
 F87 118.370000000 MHz
 F88 118.370000000 MHz
 F89 118.370000000 MHz
 F90 118.370000000 MHz
 F91 118.370000000 MHz
 F92 118.370000000 MHz
 F93 118.370000000 MHz
 F94 118.370000000 MHz
 F95 118.370000000 MHz
 F96 118.370000000 MHz
 F97 118.370000000 MHz
 F98 118.370000000 MHz
 F99 118.370000000 MHz
 F100 118.370000000 MHz

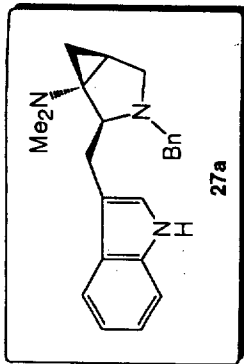




Current Data Parameters
 NAME [-888C13]
 EXPNO 1
 PROCNO 1
 F2 - Acquisition Parameters
 Date_ 99/12/24
 Time 15:11
 INSTRUM spect
 PROBNM 5 ac Data 13
 PULPROG zgpg30
 ID 85378
 SOLVENT CDCl3
 NS 20
 DS 2
 SWH 3333.33 Hz
 FIDRES 0.00036 Hz
 AQ 0.000000 sec
 SFORES 0.983000 sec
 RG 65384
 CH 0
 CT 15.000 usec
 DE 18.75 usec
 TE 300.0 K
 FL1 40 dB
 D11 0.000000 sec
 D1 2.000000 sec
 D2 2.000000 sec
 CQPRG8 waltz16
 P31 107.00 usec
 S3 18 dB
 P1 5.00 usec
 DE 18.75 usec
 SFO1 125.830000 MHz
 ACQRES 132
 F2 - Processing parameters
 SI 32768
 SF 125.8231705 MHz
 WHW 0
 SSB 0
 LB 1.30 Hz
 GB 0
 PC 1.40
 10 NMR plot parameters
 CX 20.00 cm
 FIP 180.000 ppm
 F1 22648.37 Hz
 F2 -1298.23 Hz
 F3
 PRCM 5.42857 ppm/
 K1C4 683.04010 Hz/Hz



S12

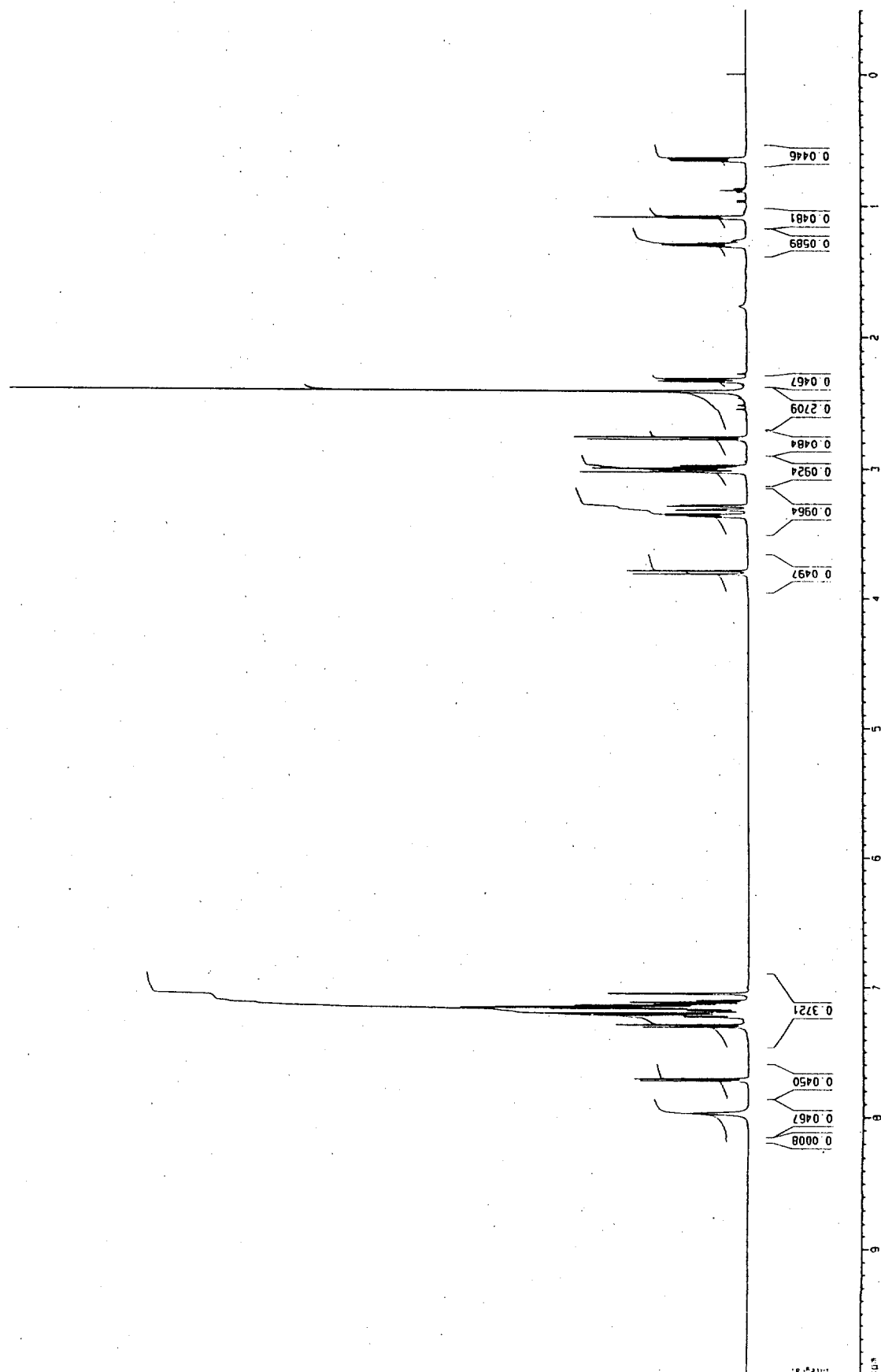


Current Data Parameters
 Name: [REDACTED]
 Sample: 1
 P0000: 1

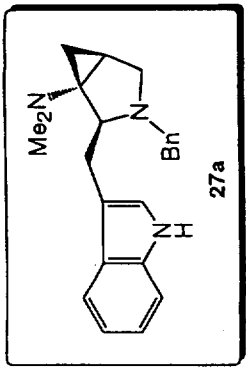
F2 - Acquisition Parameters
 Date_: 990121
 Time: 14.23
 INSTRUM: ANAL500
 P0000: 5 mm Dual 13
 PULPROG: zg
 ID: 32765
 SOLVENT: CDCl3
 NS: 1000
 DS: 2
 SFO1: 500.362518 MHz
 NUC1: 13C
 FIDRES: 6024.096 Hz
 A0: 0.183841 Hz
 RG: 128
 DM: 83.000 uSFC
 DE: 118.57 uSFC
 EQ: 300.0 Hz
 AL1: 1.0000000 Hz
 P1: 12.00 uSFC
 DE: 118.57 uSFC
 SFO1: 500.362518 MHz
 NUC1: 13C

F2 - Processing parameters
 SI: 6554
 SF: 500.362518 MHz
 WDW: EM
 SSB: 0
 LB: 0.10 Hz
 GB: 0
 PC: 1.40

10 MHz plot parameters
 SI: 6554
 SF: 10.000 MHz
 P1: 503.90 Hz
 F2: -250.20 Hz
 SFO1: 0.30000 MHz
 HZCN: 180.11702 MHz



S13

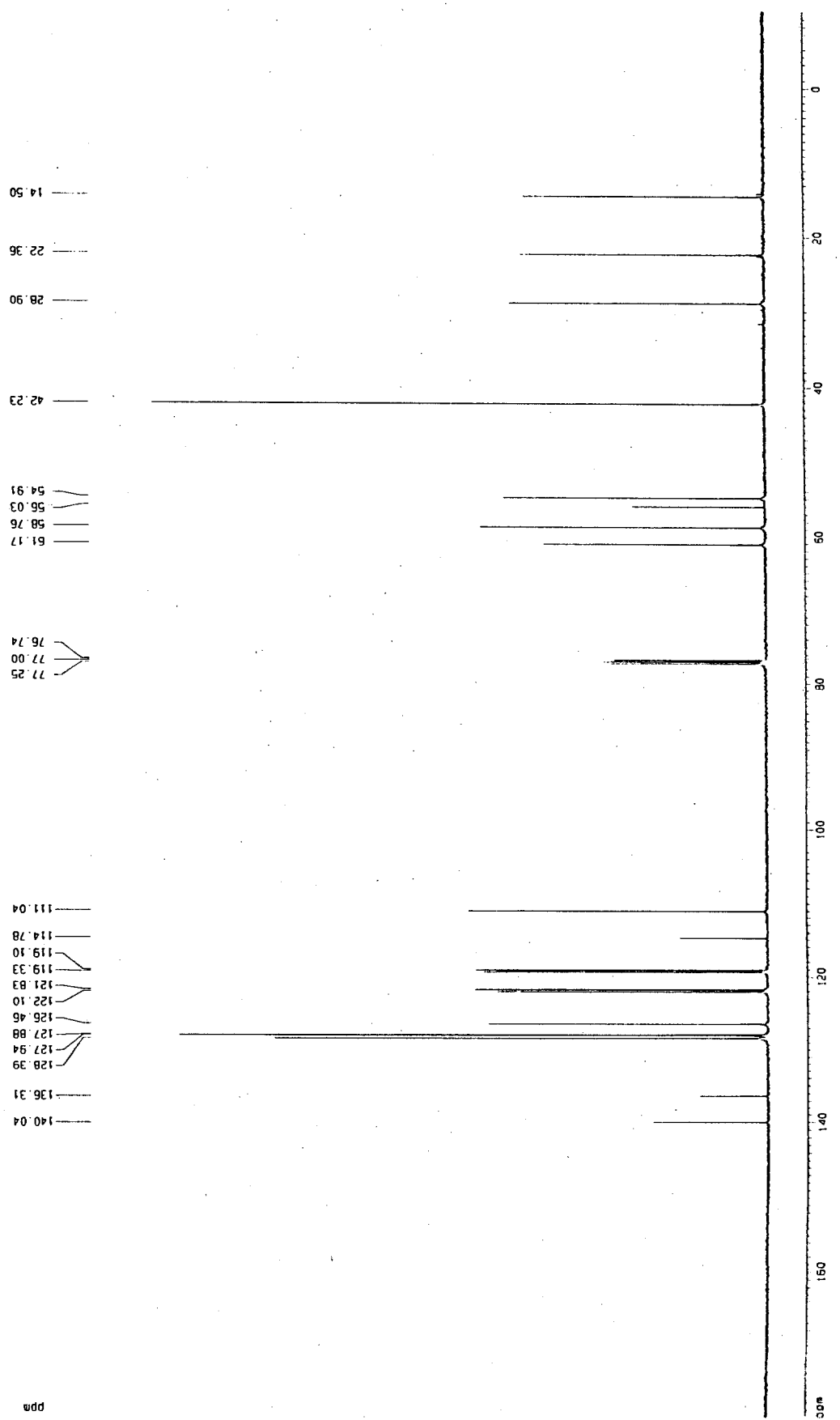


Current Data Parameters
 NAME 1-686C13
 EXPNO 1
 PROCNO 1

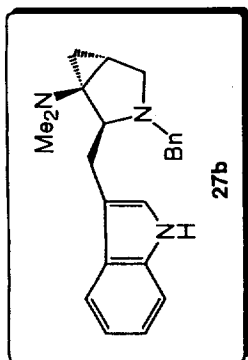
F2 - Acquisition Parameters
 Date_ 990121
 Time 14.37
 INSTRUM spect
 PULPROG zgpg30
 ID 68336
 SOLVENT CDCl3
 NS 633
 DS 2
 SWH 3333.332 Hz
 FIDRES 0.308268 Hz
 AQ 0.397456 SEC
 RG 63.84
 DM 15.000 USEC
 DE 18.75 USEC
 TE 300.0 K
 K1 40 dB
 D11 0.0000000 SEC
 S4 24 dB
 CDPRG2 waltz16
 CPDPRG2 107.00 USEC
 P31 18 dB
 S2 18 dB
 P1 5.00 USEC
 DE 18.75 USEC
 SF01 125.638148 MHz
 NUC16S 13C

F2 - Processing parameters
 SI 32768
 SF 125.631748 MHz
 WDW EM
 SSB 0
 LB 1.50 Hz
 GB 0
 PC 1.49

ID MS plot parameters
 CX 35.00 cm
 FIP 180.000 Da
 FI 28648.17 Hz
 F2 -10.000 Da
 F3 125.23 Hz
 F4 1.000 Hz
 K12M 683.0410 Hz/c1



HS

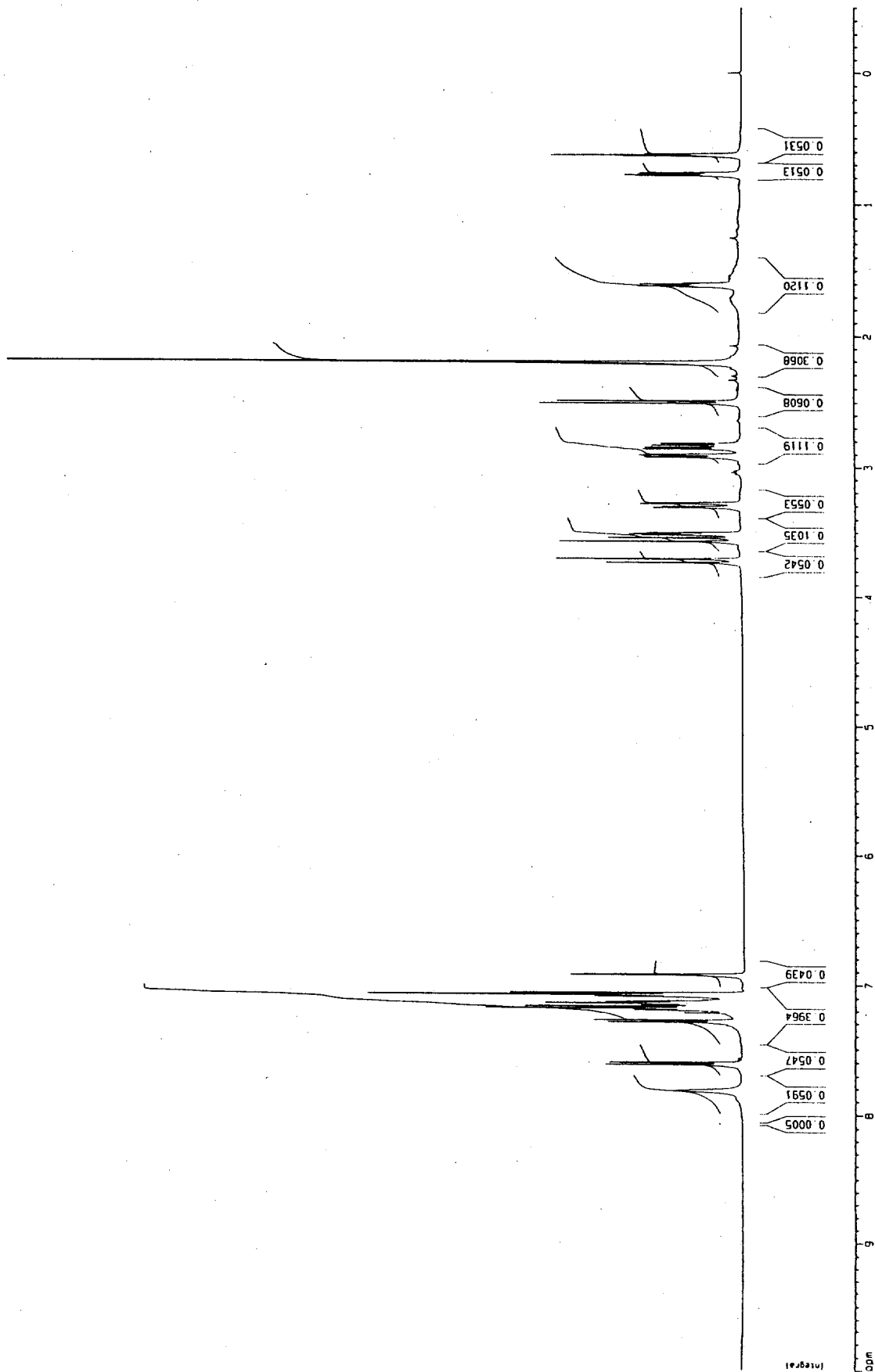


Current Data Parameters
 Name: 1-06
 EXPNO: 1
 PROCNO: 1

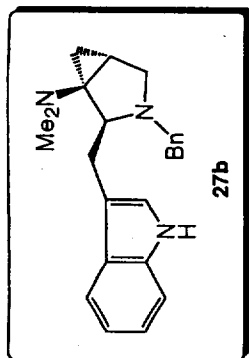
F2 - Acquisition Parameters
 Date_: 980122
 Time: 15 41
 INSTRUM: sm500
 PULPROG: zgpg30
 5 mm Outl 13
 TD: 32768
 SOLVENT: CDCl3
 NS: 17
 DS: 2
 SWH: 6024.096 Hz
 FIDRES: 0.183641 Hz
 AQ: 2.7197940 sec
 RG: 128
 DE: 83.460 uV
 TE: 118.57
 300.0 K
 K1: 3.00
 P1: 1.00000000 sec
 P2: 12.00 uV
 DE: 118.57 uV
 SFO1: 500.1362718 MHz
 NUC1: 13C

F2 - Processing parameters
 SI: 16384
 SF: 500.1362718 MHz
 WDW: EM
 SSB: 0
 LB: 0.10 Hz
 GB: 0
 PC: 1.40

10 MHz D100 parameters
 C1: 35.00 sec
 F1P: 10.000 ppm
 F1: 5003.50 Hz
 F2P: -0.500 ppm
 F2: -250.20 Hz
 RNUC1: 0.30000 ppm
 NUC2: 150.11702 Hz/C



S15

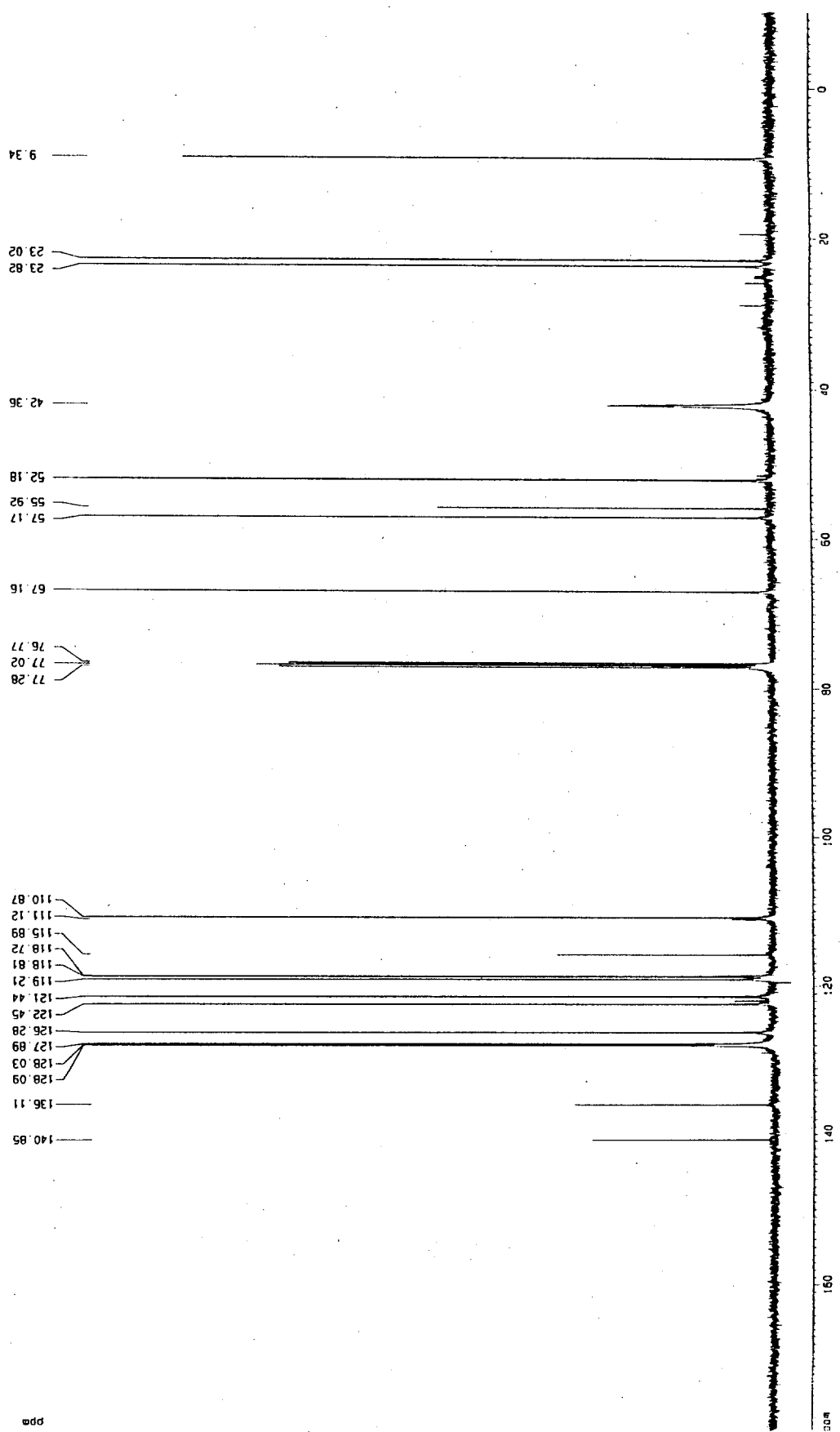


Current Data Parameters
 NAME I-80413
 EXPNO 1
 PROCNO 1

F2 - Acquisition Parameters
 DATE_ 991222
 TIME 15.59
 INSTRUM sm500
 PROBO 5 mm QNP 13
 PULPROG zgpg30
 ID 6336
 SOLVENT CDCl3
 NS 2
 DS 2
 SWH 3333.332 Hz
 FIDRES 0.508236 Hz
 AQ 0.9820900 sec
 RG 16384
 DM 15.000 usec
 DE 18.75 usec
 TE 300.2
 HL 1.00 cm
 H1 10.00 MHz
 H2 107.00 usec
 P1 18.00 usec
 P2 18.00 usec
 DE 18.75 usec
 SF01 125.005148 MHz
 NUC1EUS 13C

F2 - Processing parameters
 SI 32768
 SF 125.005148 MHz
 WDM 0
 SSB 0
 LB 1.50 Hz
 GB 0
 PC 1.40

ID Web plot parameters
 C1 30.00 usec
 F1P 100.000 MHz
 F1 22848.17 Hz
 F2P -10.000 MHz
 F2 -1254.23 Hz
 PPMCH 5.42857 usec
 N2CM 503.0410 Hz/cm



ppm