

**Synthesis of new heterocyclic systems  
fused at pyrazolo[3,4-*c*]-2,7-naphthyridine core**

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**Material and methods**

<sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded in DMSO-*d*<sub>6</sub>, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub> (1:3) or CDCl<sub>3</sub> solutions (300 MHz for <sup>1</sup>H and 75 MHz for <sup>13</sup>C, respectively) on a Varian Mercury 300VX spectrometer. Chemical shifts were reported as δ (parts per million) relative to TMS as internal standard. IR spectra were recorded on Nicolet Avatar 330-FT-IR spectrophotometer and the reported wave numbers were given in cm<sup>-1</sup>. All melting points were determined in an open capillary and were uncorrected. Elemental analyses were performed on Elemental Analyzer Euro EA 3000. Compounds **1a–g**, **4** and **5b** were described previously.

*Compounds 2a–g and 5a (general procedure).* A mixture of compound **1** or **4** (1 mmol) and hydrazine hydrate (501 mg, 10 mmol) in absolute ethanol (15 ml) was refluxed for 5 h. Ethanol was distilled off to dryness, and water (30 ml) was added to the residue. The precipitated crystals were filtered off, washed with water, dried and recrystallized from ethanol.

**7-Methyl-5-piperidin-1-yl-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-naphthyridin-1-amine**

**2a.** Colorless solid, yield 79%, mp 244–246 °C. IR  $\nu/\text{cm}^{-1}$ : 3461, 3367 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 1.57–1.74 (m, 6H, C<sub>5</sub>H<sub>10</sub>N), 2.41 (s, 3H, NCH<sub>3</sub>), 2.66 (t, *J* 6.1 Hz, 2H, 8-CH<sub>2</sub>), 2.99–3.05 (m, 4H, N(CH<sub>2</sub>)<sub>2</sub>), 3.19 (t, *J* 6.0 Hz, 2H, 9-CH<sub>2</sub>), 3.35 (s, 2H, 6-CH<sub>2</sub>), 4.47 (s, 2H, NH<sub>2</sub>), 11.28 (s, 1H, NH). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 24.1, 25.6, 26.2, 45.7, 50.9, 51.4, 54.5, 100.7, 113.9, 138.7, 147.1, 150.2, 160.4. Found (%): C, 63.24; H, 7.93; N, 29.59. Calc. for C<sub>15</sub>H<sub>22</sub>N<sub>6</sub> (%): C, 62.91; H, 7.74; N, 29.35.

**5-(4-Benzylpiperidin-1-yl)-7-methyl-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-naphthyridin-**

**1-amine 2b.** Colorless solid, yield 73%, mp 138–140 °C. IR  $\nu/\text{cm}^{-1}$ : 3433, 3206 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 1.33–1.48 and 1.62–1.79 (both m, 2H and 3H, C<sub>5</sub>H<sub>9</sub>N), 2.41 (s, 3H, NCH<sub>3</sub>), 2.60 (d, *J* 6.6 Hz, 2H, CH<sub>2</sub>Ph), 2.61–2.74 (m, 4H, 8-CH<sub>2</sub>, N(CH<sub>2</sub>)<sub>2</sub>), 3.19 (t, *J* 5.9 Hz, 2H, 9-CH<sub>2</sub>), 3.34 (s, 2H, 6-CH<sub>2</sub>), 3.35–3.40 (m, 2H, N(CH<sub>2</sub>)<sub>2</sub>), 4.47 (s, 2H, NH<sub>2</sub>), 7.10–7.16 (m, 3H, Ph), 7.20–7.27 (m, 2H, Ph), 11.30 (s, 1H, NH). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 26.2, 31.9, 37.6, 42.7, 45.7, 50.2, 51.4, 54.6, 100.7, 113.9, 125.2, 127.6, 128.4, 138.8, 139.8, 147.1, 150.2, 160.1. Found (%): C, 70.53; H, 7.73; N, 22.59. Calc. for C<sub>22</sub>H<sub>28</sub>N<sub>6</sub> (%): C, 70.18; H, 7.50; N, 22.32.

**5-[4-(Diphenylmethyl)piperazin-1-yl]-7-methyl-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-**

**naphthyridin-1-amine 2c.** Colorless solid, yield 81%, mp 218–220 °C. IR  $\nu/\text{cm}^{-1}$ : 3654, 3338 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.36 (s, 3H, NCH<sub>3</sub>), 2.50–2.56 (m, 4H, CHN(CH<sub>2</sub>)<sub>2</sub>), 2.63 (t, *J* 6.0 Hz, 2H, 8-CH<sub>2</sub>), 3.08–3.14 (m, 4H, N(CH<sub>2</sub>)<sub>2</sub>), 3.19 (t, *J* 5.9 Hz, 2H, 9-CH<sub>2</sub>), 3.28 (s, 2H, 6-CH<sub>2</sub>), 4.28 (s, 1H, CH), 4.46 (s, 2H, NH<sub>2</sub>), 7.11–7.17 (m, 2H, Ph), 7.22–7.29 (m, 4H, Ph), 7.40–7.44 (m, 4H, Ph), 11.30 (s, 1H, NH). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 26.3, 45.8, 49.6, 51.4, 51.6, 54.7, 75.7, 100.9, 113.7, 126.3, 127.3, 127.9, 139.0, 142.4, 147.2, 150.2, 159.4. Found (%): C, 71.81; H, 7.07; N, 21.87. Calc. for C<sub>27</sub>H<sub>31</sub>N<sub>7</sub> (%): C, 71.50; H, 6.89; N, 21.62.

**7-Methyl-5-(4-methylphenyl)-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-naphthyridine-1,5-diamine 2d.** Colorless solid, yield 84%, mp 232-234 °C. IR  $\nu/\text{cm}^{-1}$ : 3502, 3434, 3301 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.31 (s, 3H, NCH<sub>3</sub>), 2.48 (s, 3H, CH<sub>3</sub>Ph), 2.67 (t, *J* 5.6 Hz, 2H, 8-CH<sub>2</sub>), 3.19 (t, *J* 5.4 Hz, 2H, 9-CH<sub>2</sub>), 3.44 (s, 2H, 6-CH<sub>2</sub>), 4.38 (s, 2H, NH<sub>2</sub>), 6.99–7.05 (m, 2H, C<sub>6</sub>H<sub>4</sub>), 7.28 (s, 1H, NH), 7.49–7.54 (m, 2H, C<sub>6</sub>H<sub>4</sub>), 11.08 (s, 1H, NH<sub>pyrazole</sub>). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 20.3, 26.2, 45.7, 50.6, 52.9, 98.6, 108.7, 120.8, 128.0, 129.6, 137.6, 138.6, 147.1, 150.1, 151.9. Found (%): C, 66.60; H, 6.78; N, 27.52. Calc. for C<sub>17</sub>H<sub>20</sub>N<sub>6</sub> (%): C, 66.21; H, 6.54; N, 27.25.

**5-(4-Methoxyphenyl)-7-methyl-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-naphthyridine-1,5-diamine 2e.** Brown solid, yield 77%, mp 220-222 °C. IR  $\nu/\text{cm}^{-1}$ : 3115, 3197, 3302 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.48 (s, 3H, NCH<sub>3</sub>), 2.67 (t, *J* 5.7 Hz, 2H, 8-CH<sub>2</sub>), 3.13 (t, *J* 5.6 Hz, 2H, 9-CH<sub>2</sub>), 3.42 (s, 2H, 6-CH<sub>2</sub>), 3.77 (s, 3H, OCH<sub>3</sub>), 4.37 (s, 2H, NH<sub>2</sub>), 6.76–6.82 (m, 2H, C<sub>6</sub>H<sub>4</sub>), 7.25 (s, 1H, NH), 7.48–7.54 (m, 2H, C<sub>6</sub>H<sub>4</sub>), 11.06 (s, 1H, NH<sub>pyrazole</sub>). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 26.2, 45.8, 50.6, 53.0, 54.7, 98.4, 108.4, 113.0, 122.9, 134.2, 137.5, 147.2, 150.3, 152.4, 154.3. Found (%): C, 63.31; H, 6.40; N, 26.17. Calc. for C<sub>17</sub>H<sub>20</sub>N<sub>6</sub>O (%): C, 62.95; H, 6.21; N, 25.91.

**7-Methyl-5-(2-phenylethyl)-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-naphthyridine-1,5-diamine 2f.** Colorless solid, yield 82%, mp 208-210 °C. IR  $\nu/\text{cm}^{-1}$ : 3187, 3439 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.43 (s, 3H, NCH<sub>3</sub>), 2.62 (t, *J* 5.7 Hz, 2H, 8-CH<sub>2</sub>), 2.88–2.95 (m, 2H, CH<sub>2</sub>Ph), 3.07 (t, *J* 5.6 Hz, 2H, 9-CH<sub>2</sub>), 3.15 (s, 2H, 6-CH<sub>2</sub>), 3.57–3.66 (m, 2H, NHCH<sub>2</sub>), 4.30 (s, 2H, NH<sub>2</sub>), 5.42 (t, *J* 5.5 Hz, 1H, NHCH<sub>2</sub>), 7.12–7.29 (m, 5H, Ph), 10.95 (s, 1H, NH). Found (%): C, 67.44; H, 7.10; N, 26.35. Calc. for C<sub>18</sub>H<sub>22</sub>N<sub>6</sub> (%): C, 67.06; H, 6.88; N, 26.07.

**5-[2-(3,4-Dimethoxyphenyl)ethyl]-7-methyl-6,7,8,9-tetrahydro-3H-pyrazolo[3,4-c]-2,7-naphthyridine-1,5-diamine 2g.** Light yellow solid, yield 71%, mp 125-127 °C. IR  $\nu/\text{cm}^{-1}$ : 3333, 3417 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.43 (s, 3H, NCH<sub>3</sub>), 2.62 (t, *J* 5.7 Hz, 2H, 8-CH<sub>2</sub>), 2.80–2.87 (m, 2H, CH<sub>2</sub>Ph), 3.06 (t, *J* 5.6 Hz, 2H, 9-CH<sub>2</sub>), 3.15 (s, 2H, 6-CH<sub>2</sub>), 3.54–3.63 (m, 2H, NHCH<sub>2</sub>), 3.77 (s, 3H, OCH<sub>3</sub>), 3.78 (s, 3H, OCH<sub>3</sub>), 4.28 (s, 2H, NH<sub>2</sub>), 5.35 (t, *J* 5.4 Hz, 1H, NHCH<sub>2</sub>), 6.69–6.79 (m, 3H, C<sub>6</sub>H<sub>3</sub>), 10.91 (s, 1H, NH). Found (%): C, 63.13; H, 7.02; N, 22.21. Calc. for C<sub>20</sub>H<sub>26</sub>N<sub>6</sub>O<sub>2</sub> (%): C, 62.81; H, 6.85; N, 21.97.

**10-Methyl-8,9,10,11-tetrahydro-5H-pyrazolo[3,4-c][1,2,4]triazolo[3,4-a]-2,7-naphthyridin-7-amine 5a.** Light yellow solid, yield 83%, mp 287-289 °C. IR  $\nu/\text{cm}^{-1}$ : 3437, 3311, 3198 (NH, NH<sub>2</sub>). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.48 (s, 3H, NCH<sub>3</sub>), 2.71 (t, *J* 5.7 Hz, 2H, 9-CH<sub>2</sub>), 2.98–3.04 (m, 2H, 8-CH<sub>2</sub>), 3.63 (s, 2H, 11-CH<sub>2</sub>), 5.47 (s, 2H, NH<sub>2</sub>), 8.86 (s, 1H, CH), 11.80 (s, 1H, NH). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 25.6, 45.4, 50.9, 52.3, 92.9, 108.8, 129.7, 133.7, 144.3, 148.5. Found (%): C, 54.66; H, 5.59; N, 40.57. Calc. for C<sub>11</sub>H<sub>13</sub>N<sub>7</sub> (%): C, 54.31; H, 5.39; N, 40.30.

*Compounds 3a–g and 6a,b (general procedure).* A mixture of compound **2a-g** or **5a,b** (1 mmol) and acetylacetone (15 ml) was refluxed for 5 h. The reaction mixture was cooled, the precipitated crystals were filtered off, washed with water, dried, and recrystallized from ethanol.

**3,9,11-Trimethyl-5-piperidin-1-yl-1,2,3,4-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-c]-2,7-naphthyridine 3a.** Light yellow solid, yield 69%, mp 198-200 °C. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 1.61–1.82 (m, 6H, C<sub>5</sub>H<sub>10</sub>N), 2.46 (s, 3H, NCH<sub>3</sub>), 2.64 (s, 3H, CH<sub>3</sub>), 2.80 (t, *J* 6.1 Hz, 2H, 2-CH<sub>2</sub>), 2.84 (s, 3H, CH<sub>3</sub>), 3.18–3.26 (m, 4H, N(CH<sub>2</sub>)<sub>2</sub>), 3.41–3.51 (m, 4H, 1,4-CH<sub>2</sub>), 7.00 (s, 1H, CH). Found (%): C, 68.87; H, 7.66; N, 24.23. Calc. for C<sub>20</sub>H<sub>26</sub>N<sub>6</sub> (%): C, 68.54; H, 7.48; N, 23.98.

**5-(4-Benzylpiperidin-1-yl)-3,9,11-trimethyl-1,2,3,4-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-c]-2,7-naphthyridine 3b.** Light yellow solid, yield 72%, mp 224-225 °C. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 1.39–1.55 and 1.73–1.86 (both m, 2H and 3H, C<sub>5</sub>H<sub>9</sub>N), 2.46 (s, 3H, NCH<sub>3</sub>), 2.62 (d, *J* 6.8 Hz, 2H, CH<sub>2</sub>Ph), 2.64 (s, 3H, CH<sub>3</sub>), 2.73–2.90 (m, 4H, 1,2-CH<sub>2</sub>), 2.83 (s, 3H, CH<sub>3</sub>), 3.43 (s, 2H, 4-CH<sub>2</sub>), 3.42–3.48 (m, 2H, N(CH<sub>2</sub>)<sub>2</sub>), 3.57–3.66 (m, 2H, N(CH<sub>2</sub>)<sub>2</sub>), 6.99 (s, 1H, CH), 7.11–7.18 (m, 3H, Ph), 7.21–7.28 (m, 2H, Ph). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 17.0, 23.9, 27.0, 31.8, 37.6, 42.7, 45.7, 49.8, 51.3, 54.8, 99.5, 110.2, 116.4, 125.2, 127.6, 128.4, 139.7, 139.9, 142.2, 143.7, 154.6, 158.1, 162.8. Found (%): C, 73.96; H, 7.53; N, 19.35. Calc. for C<sub>27</sub>H<sub>32</sub>N<sub>6</sub> (%): C, 73.60; H, 7.32; N, 19.07.

**5-[4-(Diphenylmethyl)piperazin-1-yl]-3,9,11-trimethyl-1,2,3,4-tetrahydropyrimido[1',2':1,5]-pyrazolo[3,4-c]-2,7-naphthyridine 3c.** Yellow solid, yield 70%, mp 227-229 °C. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.43 (s, 3H, NCH<sub>3</sub>), 2.55–2.62 (m, 4H, CHN(CH<sub>2</sub>)<sub>2</sub>), 2.64 (s, 3H, CH<sub>3</sub>), 2.73–2.81 (m, 2H, 2-CH<sub>2</sub>), 2.85 (s, 3H, CH<sub>3</sub>), 3.28–3.49 (m, 8H, N(CH<sub>2</sub>)<sub>2</sub>, 1,4-CH<sub>2</sub>), 4.31 (s, 1H, CHN(CH<sub>2</sub>)<sub>2</sub>), 7.01 (s, 1H, 10-CH), 7.12–7.19 (m, 2H, Ph), 7.23–7.31 (m, 4H, Ph), 7.42–7.48 (m, 4H, Ph). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 17.0, 23.9, 26.6, 45.1, 49.4, 50.9, 51.4, 54.4,

75.6, 99.4, 110.4, 126.3, 127.2, 127.9, 139.8, 142.3, 143.8, 154.9, 157.9, 162.0. Found (%): C, 74.59; H, 7.00; N, 19.20. Calc. for C<sub>32</sub>H<sub>35</sub>N<sub>7</sub> (%): C, 74.25; H, 6.81; N, 18.94.

**3,9,11-Trimethyl-N-(4-methylphenyl)-1,2,3,4-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-c]-2,7-naphthyridin-5-amine 3d.** Yellow solid, yield 74%, mp 216-218 °C. IR  $\nu/\text{cm}^{-1}$ : 3460, 3333 (NH). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.35 (s, 3H, NCH<sub>3</sub>), 2.53 (s, 3H, CH<sub>3</sub>), 2.61 (s, 3H, CH<sub>3</sub>), 2.75 (t, *J* 5.7 Hz, 2H, 2-CH<sub>2</sub>), 2.80 (s, 3H, CH<sub>3</sub>C<sub>6</sub>H<sub>4</sub>), 3.40 (t, *J* 5.6 Hz, 2H, 1-CH<sub>2</sub>), 3.56 (s, 2H, 4-CH<sub>2</sub>), 6.89 (s, 1H, 10-CH), 7.07–7.12 (m, 2H, C<sub>6</sub>H<sub>4</sub>), 7.64 (s, 1H, NH), 7.74–7.80 (m, 2H, C<sub>6</sub>H<sub>4</sub>). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 17.0, 20.3, 23.8, 26.8, 45.7, 50.4, 52.8, 97.8, 109.3, 112.1, 121.1, 127.9, 130.2, 138.0, 138.2, 142.3, 143.5, 154.2, 154.5, 158.4. Found (%): C, 71.31; H, 6.71; N, 22.85. Calc. for C<sub>22</sub>H<sub>24</sub>N<sub>6</sub> (%): C, 70.94; H, 6.49; N, 22.56.

**N-(4-Methoxyphenyl)-3,9,11-trimethyl-1,2,3,4-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-c]-2,7-naphthyridin-5-amine 3e.** Yellow solid, yield 71%, mp 237-239 °C. IR  $\nu/\text{cm}^{-1}$ : 3337 (NH). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.54 (s, 3H, NCH<sub>3</sub>), 2.62 (s, 3H, CH<sub>3</sub>), 2.76 (t, *J* 5.7 Hz, 2H, 2-CH<sub>2</sub>), 2.80 (s, 3H, CH<sub>3</sub>), 3.41 (t, *J* 5.3 Hz, 2H, 1-CH<sub>2</sub>), 3.56 (s, 2H, 4-CH<sub>2</sub>), 3.80 (s, 3H, OCH<sub>3</sub>), 6.82–6.90 (m, 3H, 10-CH, C<sub>6</sub>H<sub>4</sub>), 7.64 (s, 1H, NH), 7.73–7.79 (m, 2H, C<sub>6</sub>H<sub>4</sub>). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3)  $\delta$ : 17.0, 23.8, 26.7, 45.7, 50.4, 52.8, 54.6, 97.7, 109.3, 111.9, 112.8, 122.9, 133.6, 137.9, 142.3, 143.4, 154.4, 154.5, 158.5. Found (%): C, 68.37; H, 6.44; N, 21.90. Calc. for C<sub>22</sub>H<sub>24</sub>N<sub>6</sub>O (%): C, 68.02; H, 6.23; N, 21.63.

**3,9,11-Trimethyl-N-(2-phenylethyl)-1,2,3,4-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-c]-2,7-naphthyridin-5-amine 3f.** Yellow solid, yield 68%, mp 204-206 °C. IR  $\nu/\text{cm}^{-1}$ : 3315 (NH). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>)  $\delta$ : 2.46 (s, 3H, NCH<sub>3</sub>), 2.56 (s, 3H, CH<sub>3</sub>), 2.69–2.79 (m, 2H, 2-CH<sub>2</sub>), 2.75 (s, 3H, CH<sub>3</sub>), 2.92–2.99 (m, 2H, CH<sub>2</sub>Ph), 3.24–3.35 (m, 4H, 1,4-CH<sub>2</sub>), 3.64–3.73 (m, 2H, NHCH<sub>2</sub>), 6.55 (t, *J* 5.4 Hz, 1H, NH), 7.04 (s, 1H, 10-CH), 7.18–7.36 (m, 5H, Ph). <sup>13</sup>C NMR (75 MHz, DMSO-*d*<sub>6</sub>)  $\delta$ : 17.1, 24.1, 26.7, 34.7, 42.4, 45.7, 50.3, 52.7, 96.4, 109.7, 125.9, 128.2, 128.6, 128.6, 137.2, 140.1, 142.5, 144.2, 155.6, 156.9, 159.4. Found (%): C, 71.80; H, 6.95; N, 21.99. Calc. for C<sub>23</sub>H<sub>26</sub>N<sub>6</sub> (%): C, 71.48; H, 6.78; N, 21.74.

**N-[2-(3,4-Dimethoxyphenyl)ethyl]-3,9,11-trimethyl-1,2,3,4-tetrahydropyrimido[1',2':1,5]-pyrazolo[3,4-c]-2,7-naphthyridin-5-amine 3g.** Yellow solid, yield 69%, mp 184-186 °C. IR  $\nu/\text{cm}^{-1}$ : 3317 (NH). <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>/CCl<sub>4</sub>, 1:3)  $\delta$ : 2.44 (s, 3H, NCH<sub>3</sub>), 2.56 (s, 3H, CH<sub>3</sub>), 2.70 (br t, *J* 5.4 Hz, 2H, 2-CH<sub>2</sub>), 2.74 (s, 3H, CH<sub>3</sub>), 2.85–2.92 (m, 2H, CH<sub>2</sub>C<sub>6</sub>H<sub>3</sub>), 3.23–3.33 (m, 4H, 1,4-CH<sub>2</sub>), 3.62–3.73 (m, 2H, NHCH<sub>2</sub>), 3.72 (s, 3H, OCH<sub>3</sub>), 3.76 (s, 3H, OCH<sub>3</sub>),

6.48 (t, *J* 5.4 Hz, 1H, NH), 6.75–6.80 (m, 1H, C<sub>6</sub>H<sub>3</sub>), 6.86–6.90 (m, 2H, C<sub>6</sub>H<sub>3</sub>), 7.04 (s, 1H, 10-CH). <sup>13</sup>C NMR (75 MHz, DMSO/CCl<sub>4</sub>, 1/3) δ: 17.0, 24.1, 26.8, 34.3, 42.5, 45.8, 50.4, 52.8, 55.3, 55.5, 96.4, 109.6, 111.7, 111.9, 112.6, 120.4, 132.7, 137.2, 142.5, 144.2, 147.1, 148.6, 155.5, 156.9, 159.4. Found (%): C, 67.62; H, 6.99; N, 19.11. Calc. for C<sub>25</sub>H<sub>30</sub>N<sub>6</sub>O<sub>2</sub> (%): C, 67.24; H, 6.77; N, 18.82.

**5,9,11-Trimethyl-4,5,6,7-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-*c*][1,2,4]triazolo[3,4-*a*]-2,7-naphthyridine 6a.** Colorless solid, yield 71%, mp > 340 °C. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ: 2.61 (s, 3H, NCH<sub>3</sub>), 2.69 (s, 3H, CH<sub>3</sub>), 2.86 (s, 3H, CH<sub>3</sub>), 2.90 (t, *J* 5.7 Hz, 2H, 6-CH<sub>2</sub>), 3.44–3.51 (m, 2H, 7-CH<sub>2</sub>), 3.97 (t, *J* 2.0 Hz, 2H, 4-CH<sub>2</sub>), 6.87 (s, 1H, 10-CH), 9.21 (s, 1H, 1-CH). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ: 17.5, 24.9, 26.8, 46.1, 51.6, 52.8, 98.0, 110.8, 114.0, 130.1, 133.7, 143.8, 143.9, 146.0, 149.2, 159.8. Found (%): C, 62.86; H, 5.77; N, 32.16. Calc. for C<sub>16</sub>H<sub>17</sub>N<sub>7</sub> (%): C, 62.52; H, 5.58; N, 31.90.

**5,9,11-Trimethyl-4,5,6,7-tetrahydropyrimido[1',2':1,5]pyrazolo[3,4-*c*]tetrazolo[5,1-*a*]-2,7-naphthyridine 6b.** Yellow solid, yield 73%, mp > 300 °C. IR ν/cm<sup>-1</sup>: 1635 (C=C<sub>Ar</sub>). <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ: 2.63 (s, 3H, NCH<sub>3</sub>), 2.73 (s, 3H, CH<sub>3</sub>), 2.93 (t, *J* 5.7 Hz, 2H, 6-CH<sub>2</sub>), 2.98 (s, 3H, CH<sub>3</sub>), 3.53–3.59 (m, 2H, 7-CH<sub>2</sub>), 4.01 (t, *J* 2.0 Hz, 2H, 4-CH<sub>2</sub>), 7.00 (s, 1H, 10-CH). <sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ: 17.8, 25.1, 27.1, 46.0, 51.4, 52.9, 100.0, 112.1, 112.9, 135.3, 143.3, 144.2, 146.9, 149.2, 160.6. Found (%): C, 58.74; H, 5.40; N, 36.58. Calc. for C<sub>15</sub>H<sub>16</sub>N<sub>8</sub> (%): C, 58.43; H, 5.23; N, 36.34.

201

Spice

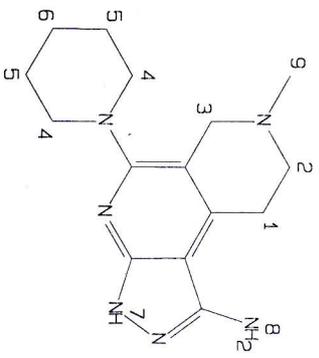
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

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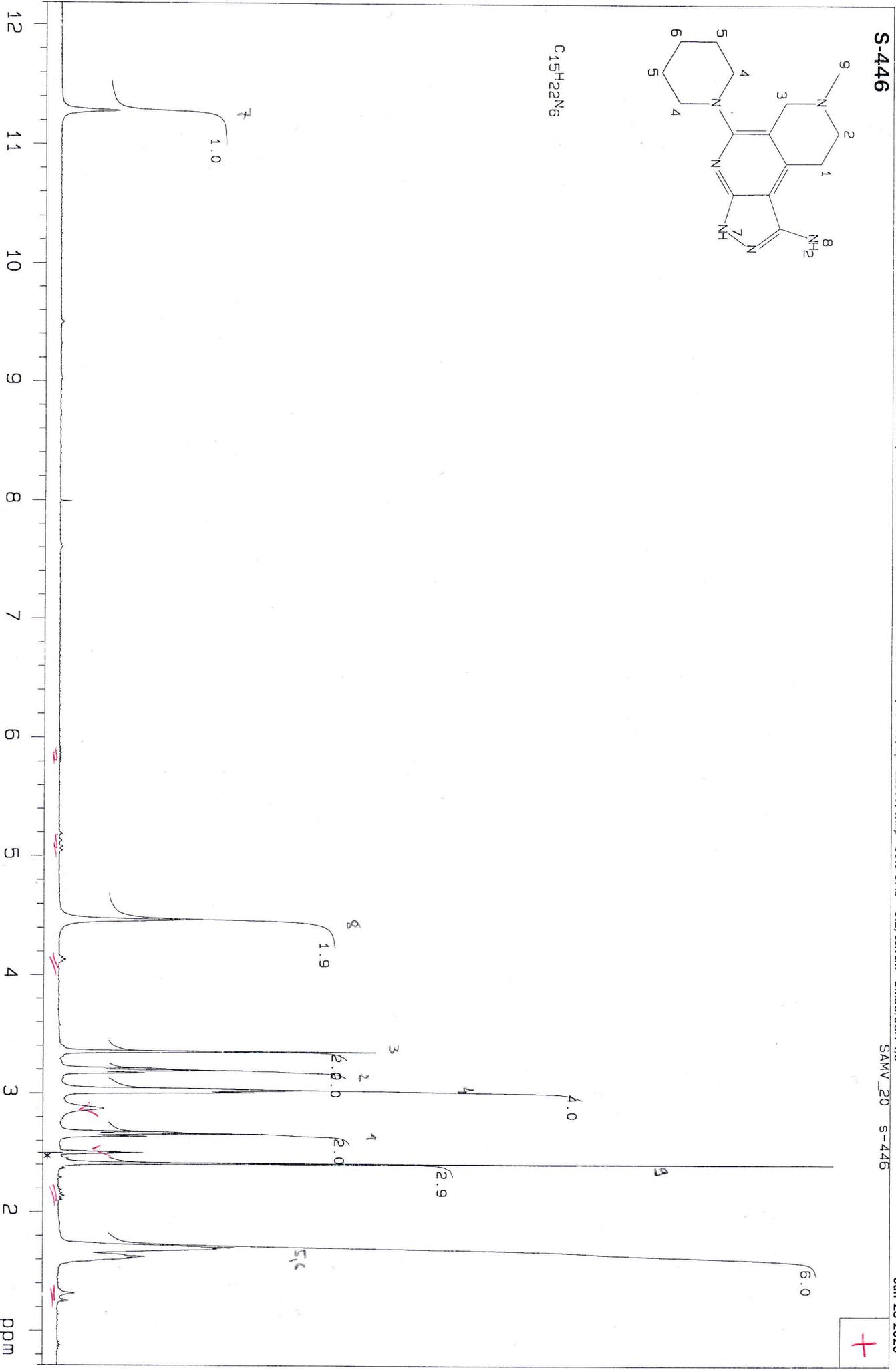
Jan 23 2020

S-446

SAMV\_20 s-446



C<sub>15</sub>H<sub>22</sub>N<sub>6</sub>



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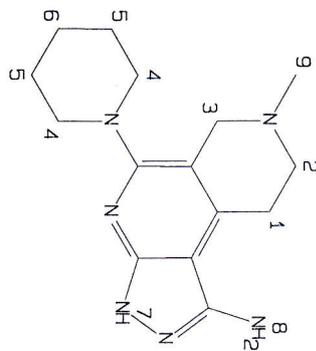
S-446

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

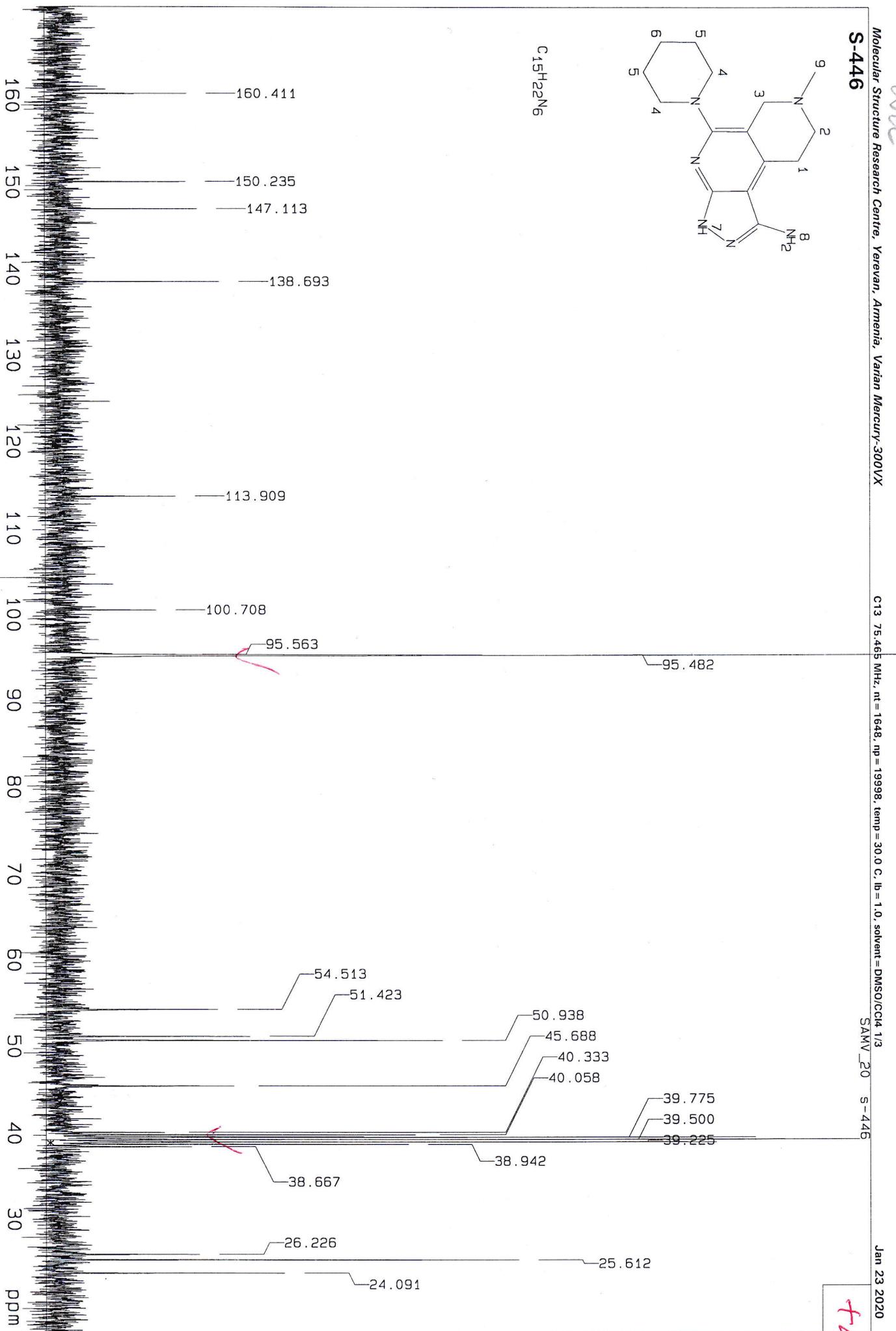
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SAMV\_20 S-446

Jan 23 2020



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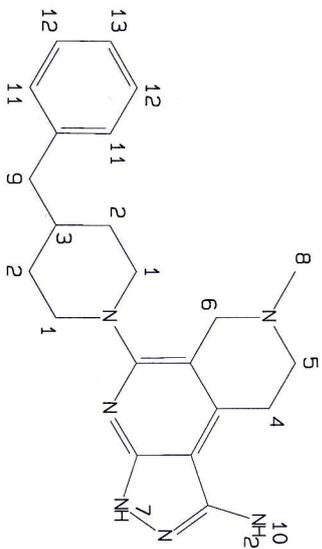
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

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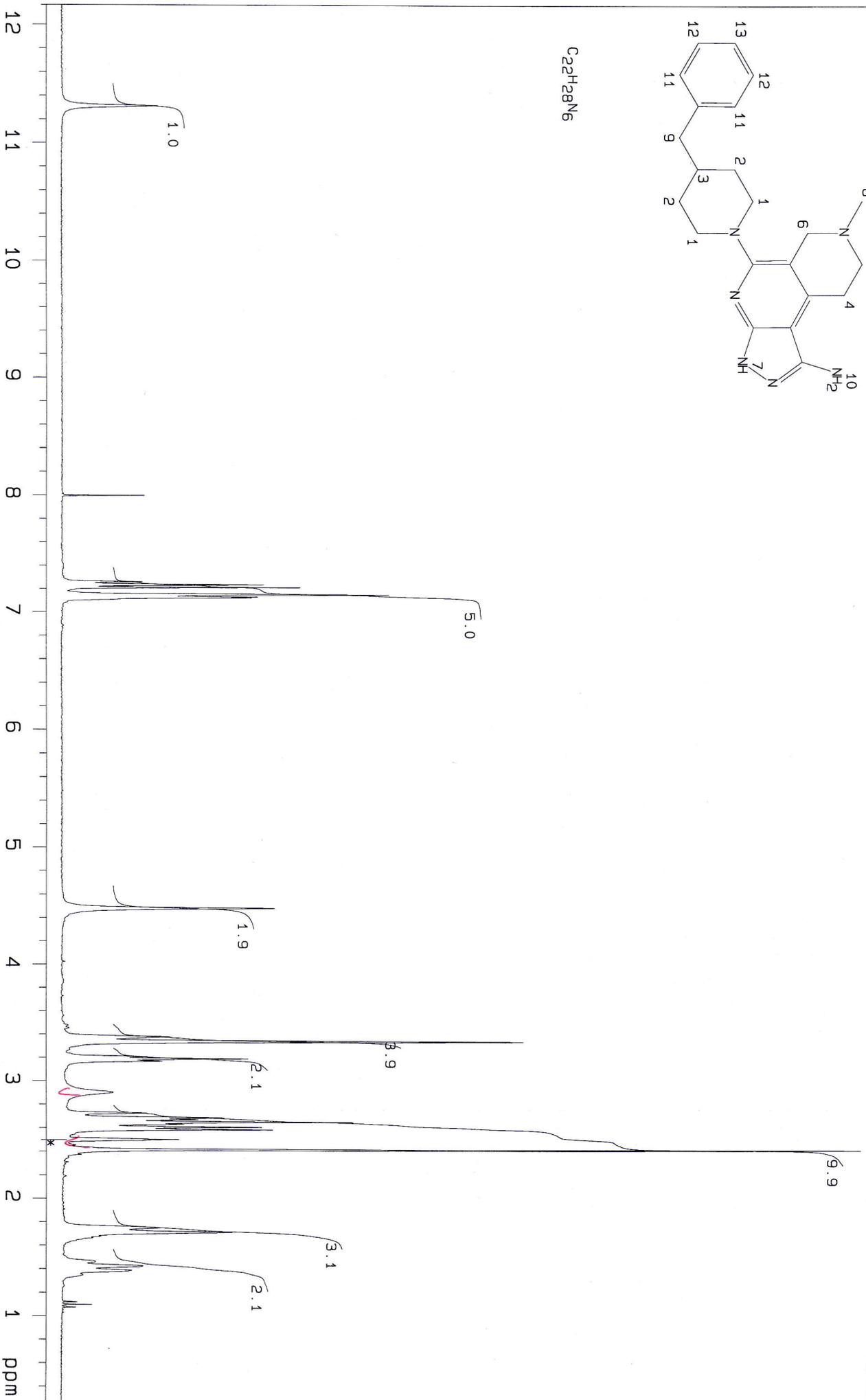
Oct 4 2021

AE-S0358-0034

NOCI\_21 ae-S0358-0034



C<sub>22</sub>H<sub>28</sub>N<sub>6</sub>



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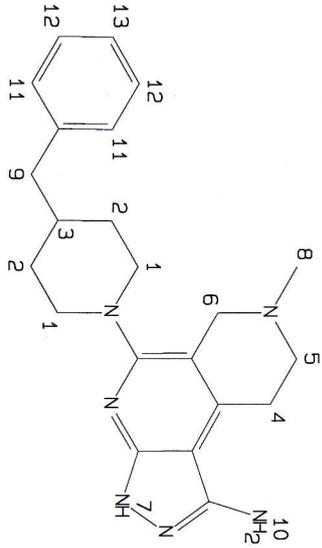
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Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
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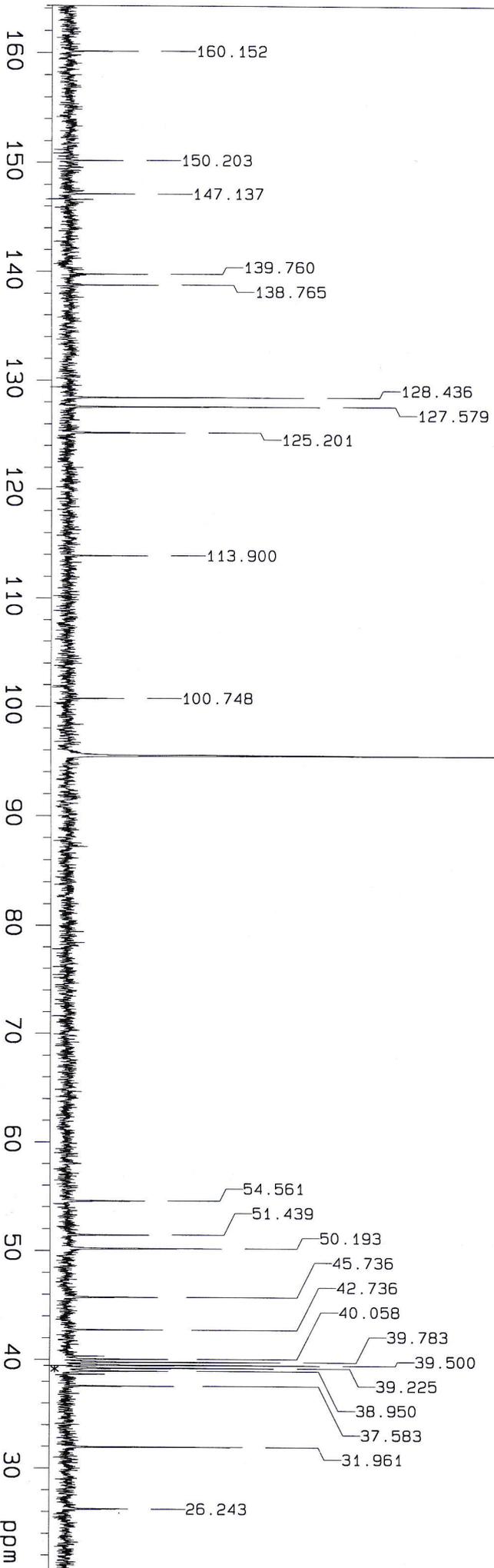
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NOCI\_21 ae-s0358-0034

Oct 4 2021



C<sub>22</sub>H<sub>28</sub>N<sub>6</sub>

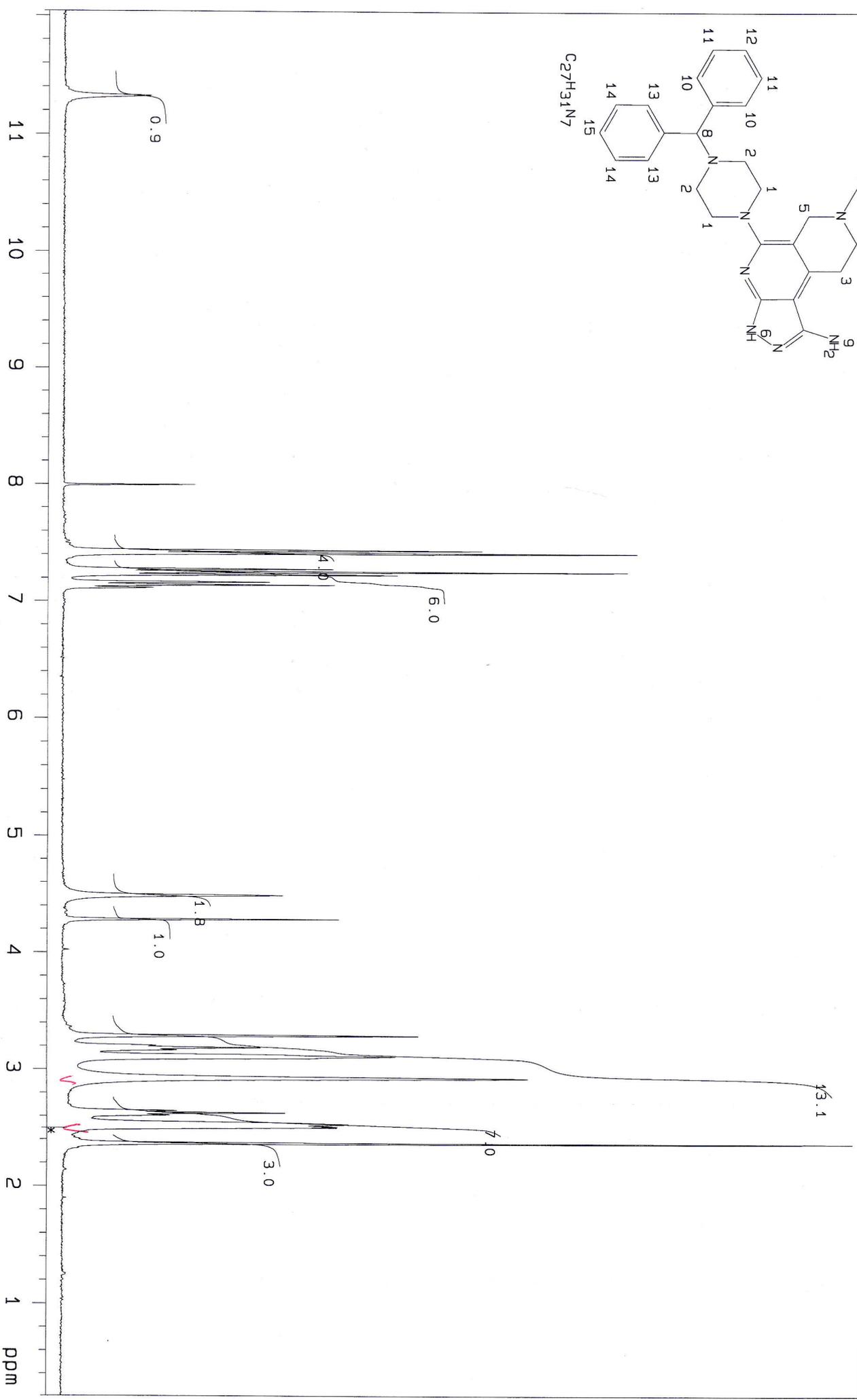
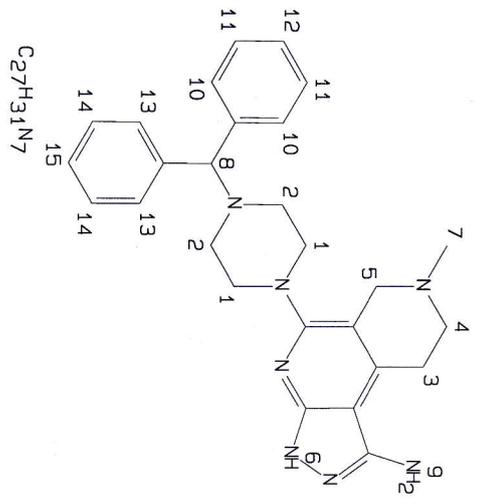


2c

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
**AE-S0358-0038**

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NOCT\_21 ae-s0358-0038

Oct 4 2021



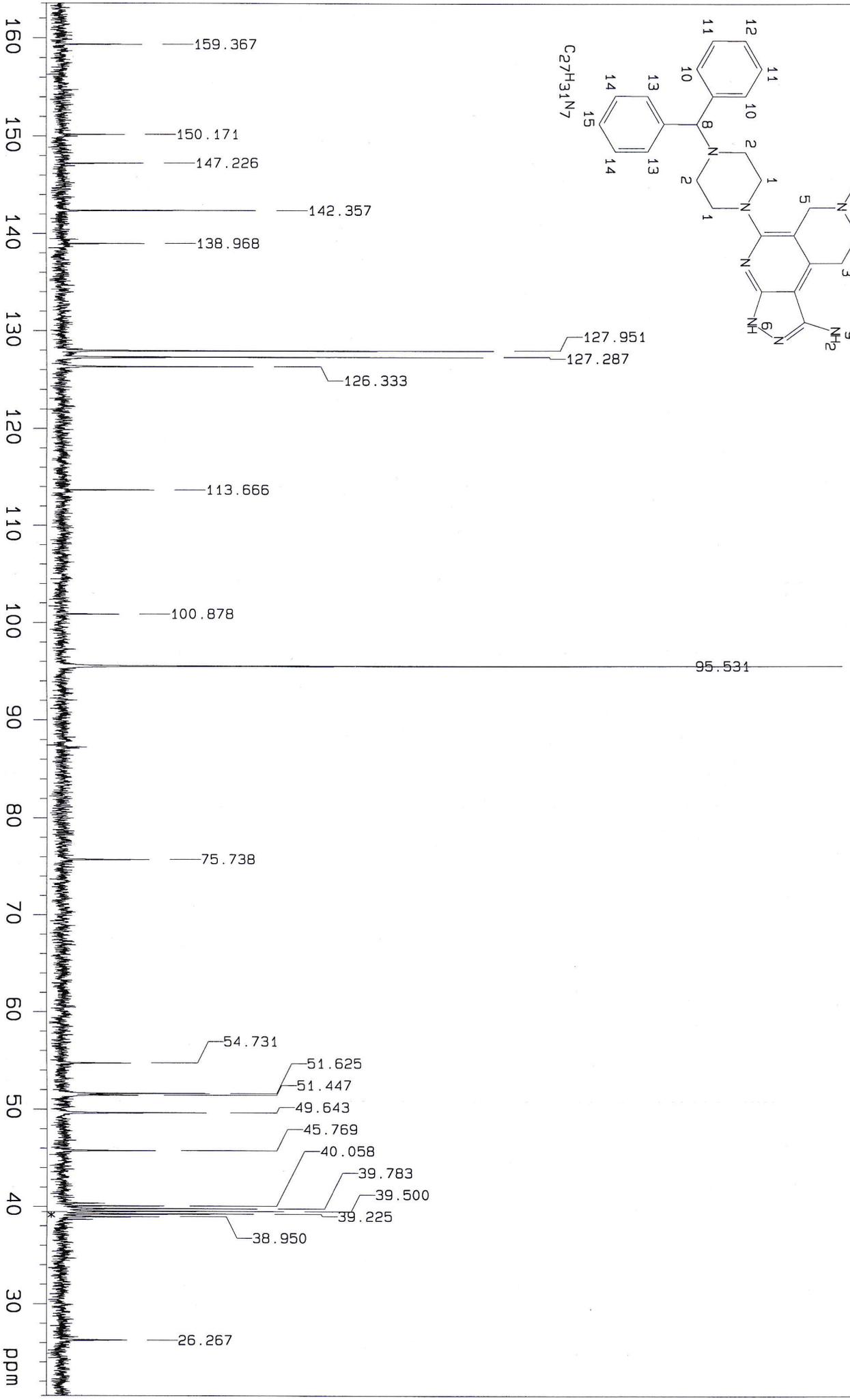
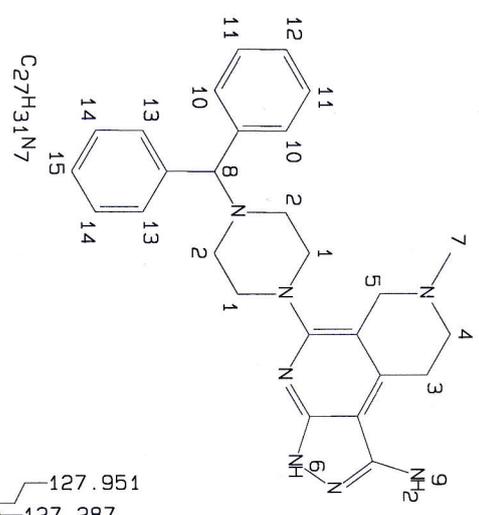
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Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
AE-S0358-0038

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NOCT\_21 ae-s0358-0038

Oct 4 2021



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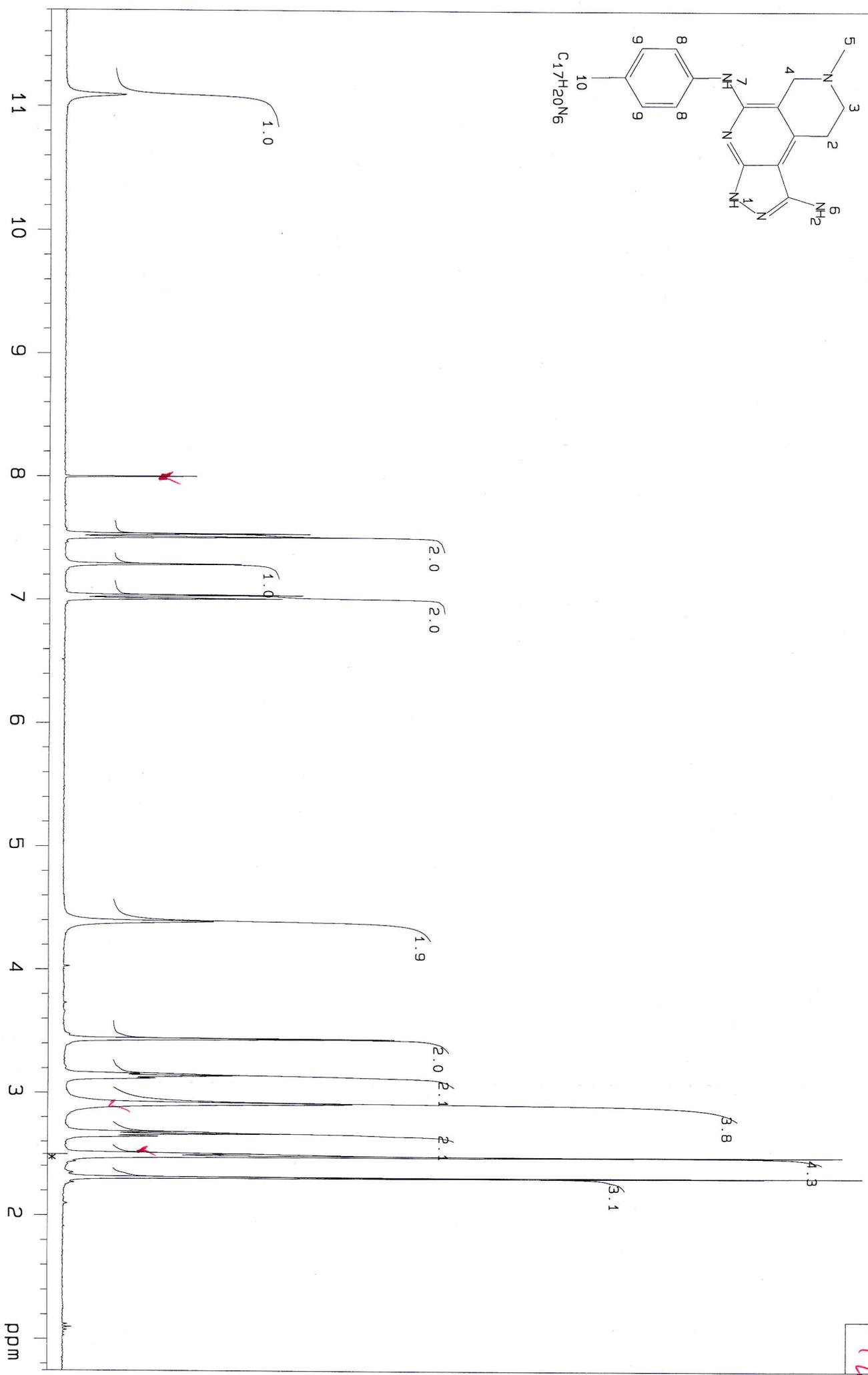
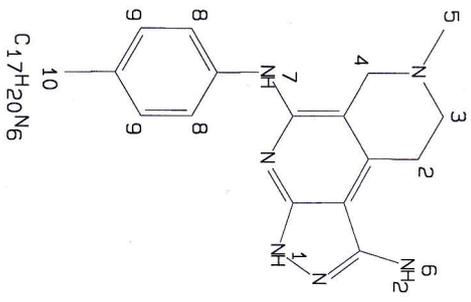
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Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
**SC-0358-0056**

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Oct 5 2021



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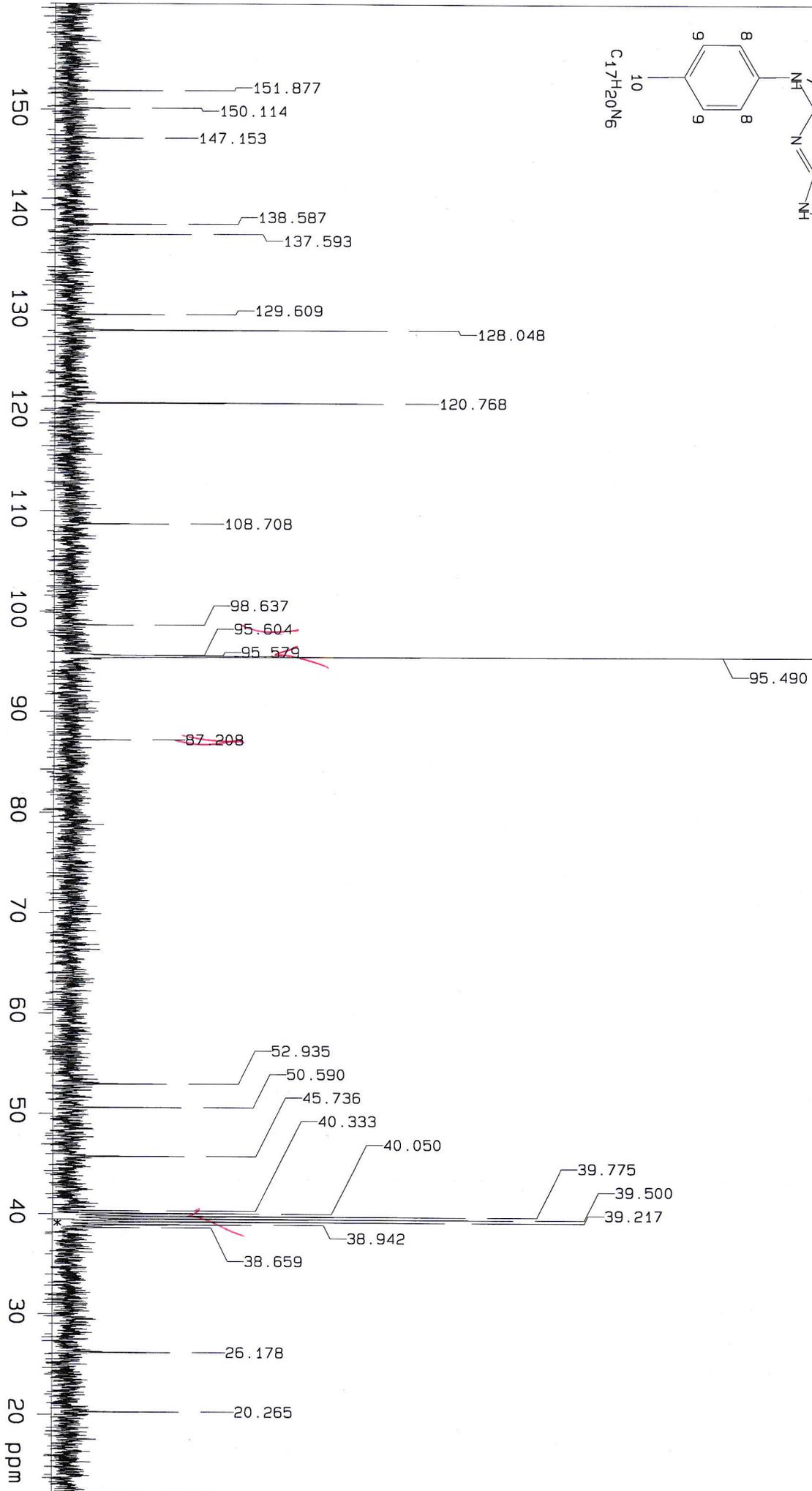
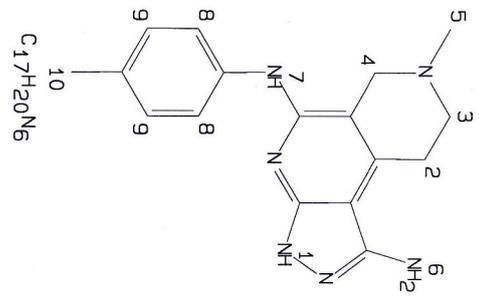
221

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
**SC-0358-0056**

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ANUSH\_TEMA SC-0358-0056

Oct 5 2021



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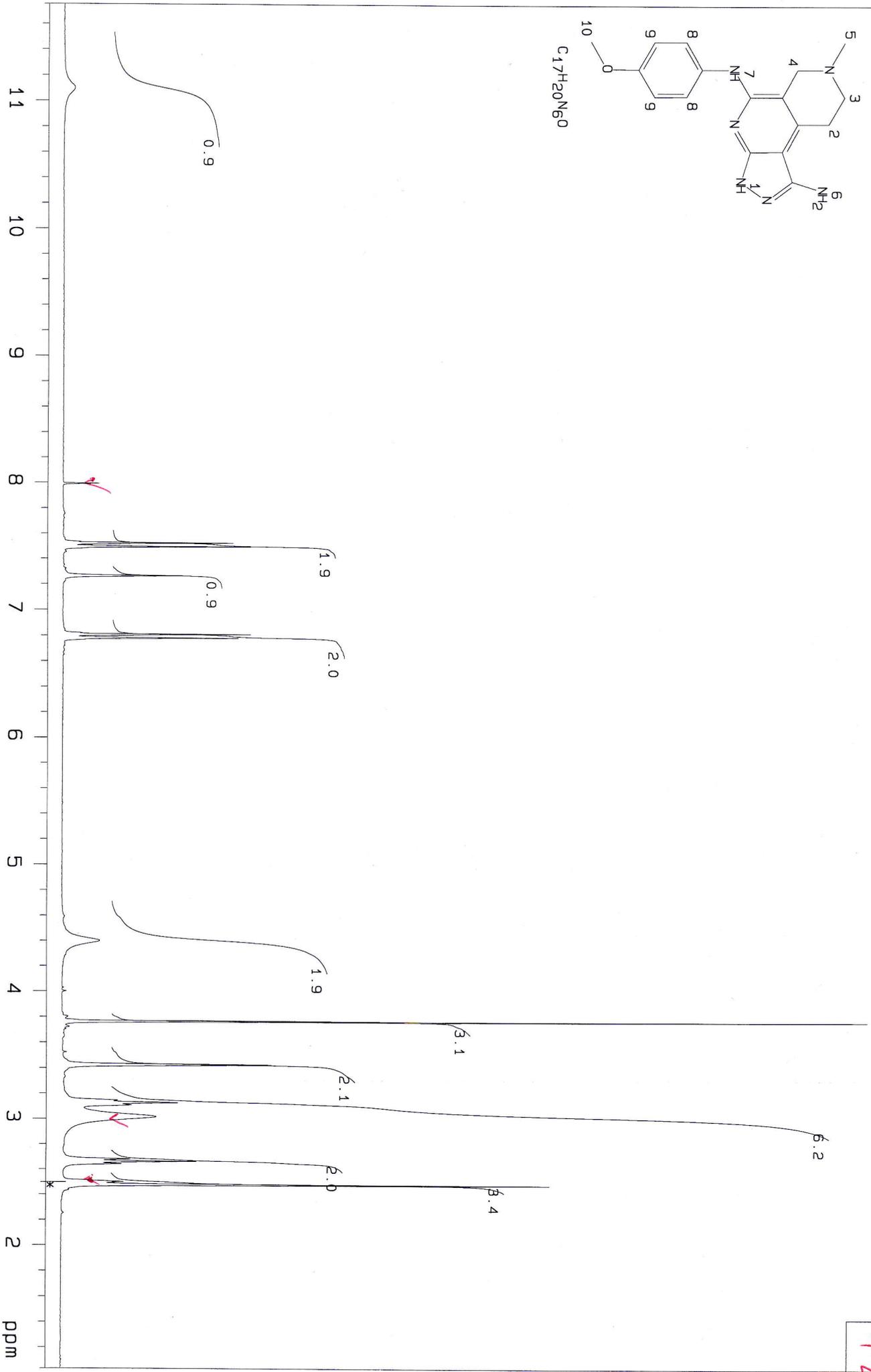
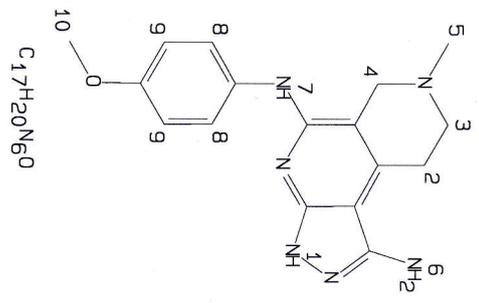
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

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ANUSH\_TEMA SC-0358-0057

Oct 5 2021

SC-0358-0057



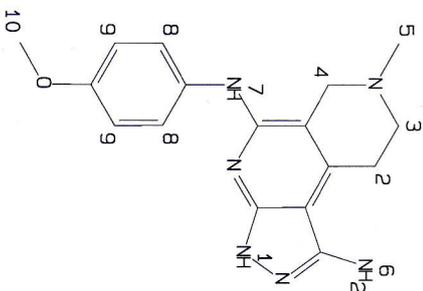
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22

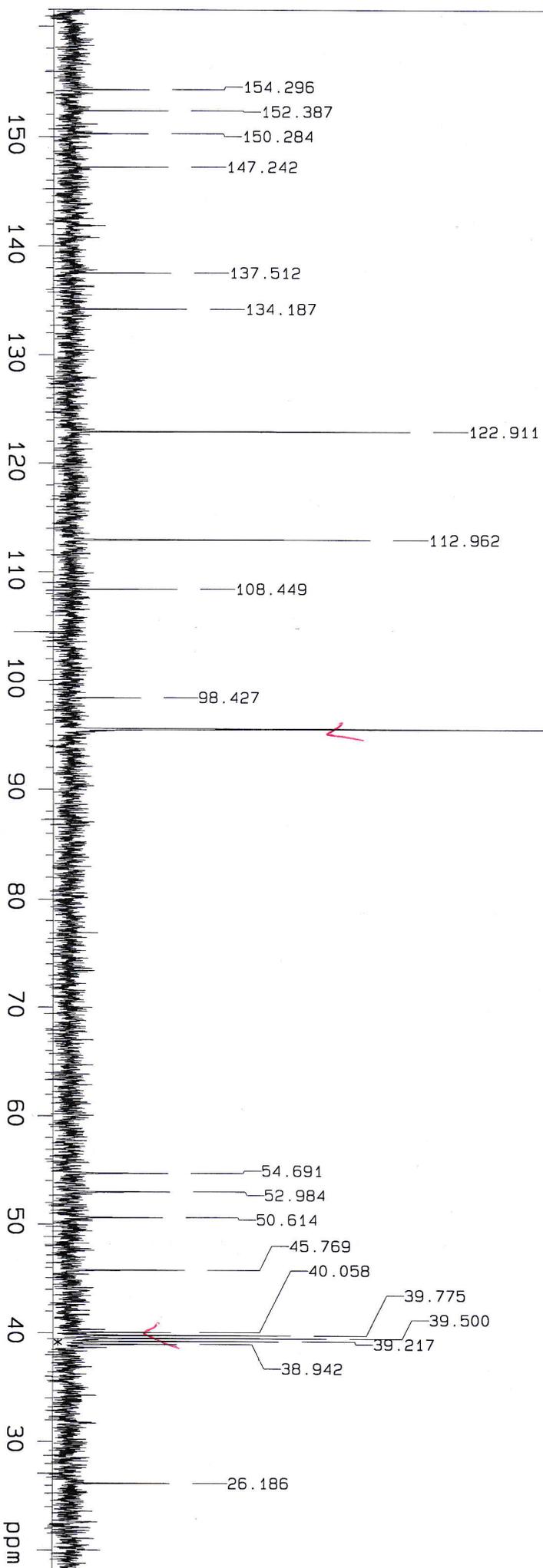
SC-0358-0057

ANUSH\_TEMA SC-0358-0057

Oct 5 2021



C<sub>17</sub>H<sub>20</sub>N<sub>6</sub>O



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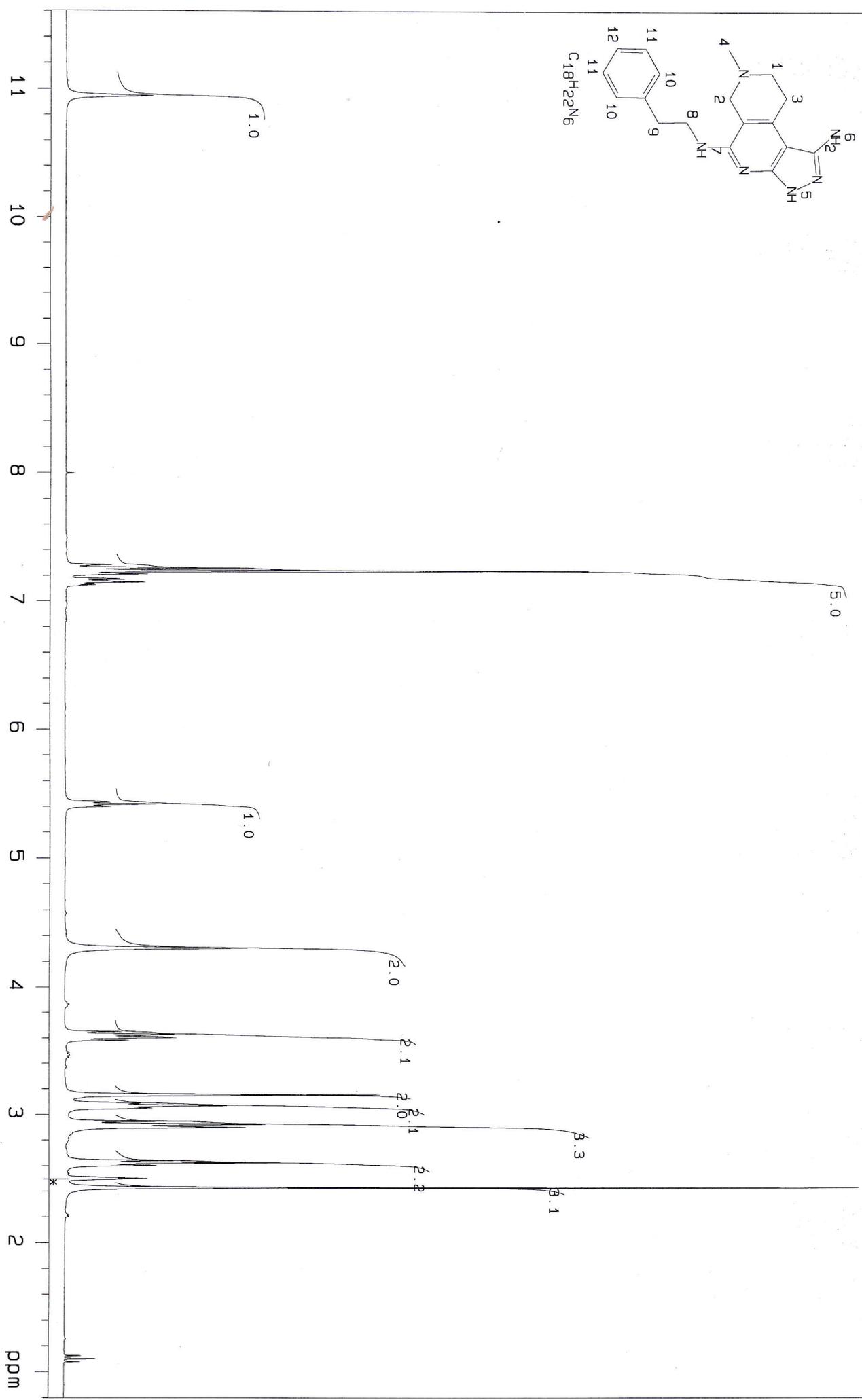
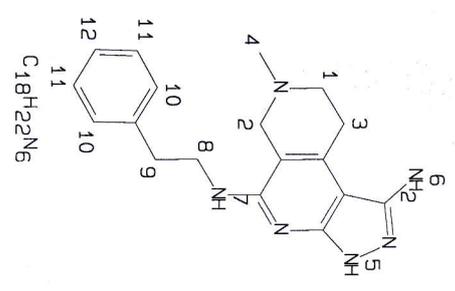
2f

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
AE-S0358-0018

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SAMV\_04A ae-s0358-0018

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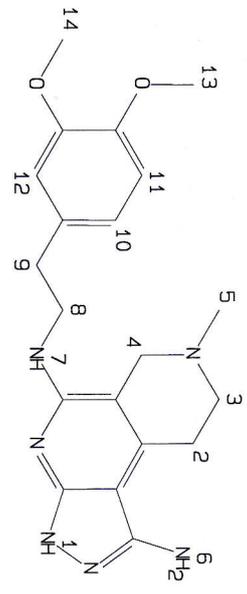
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Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
AE-S0958-0047

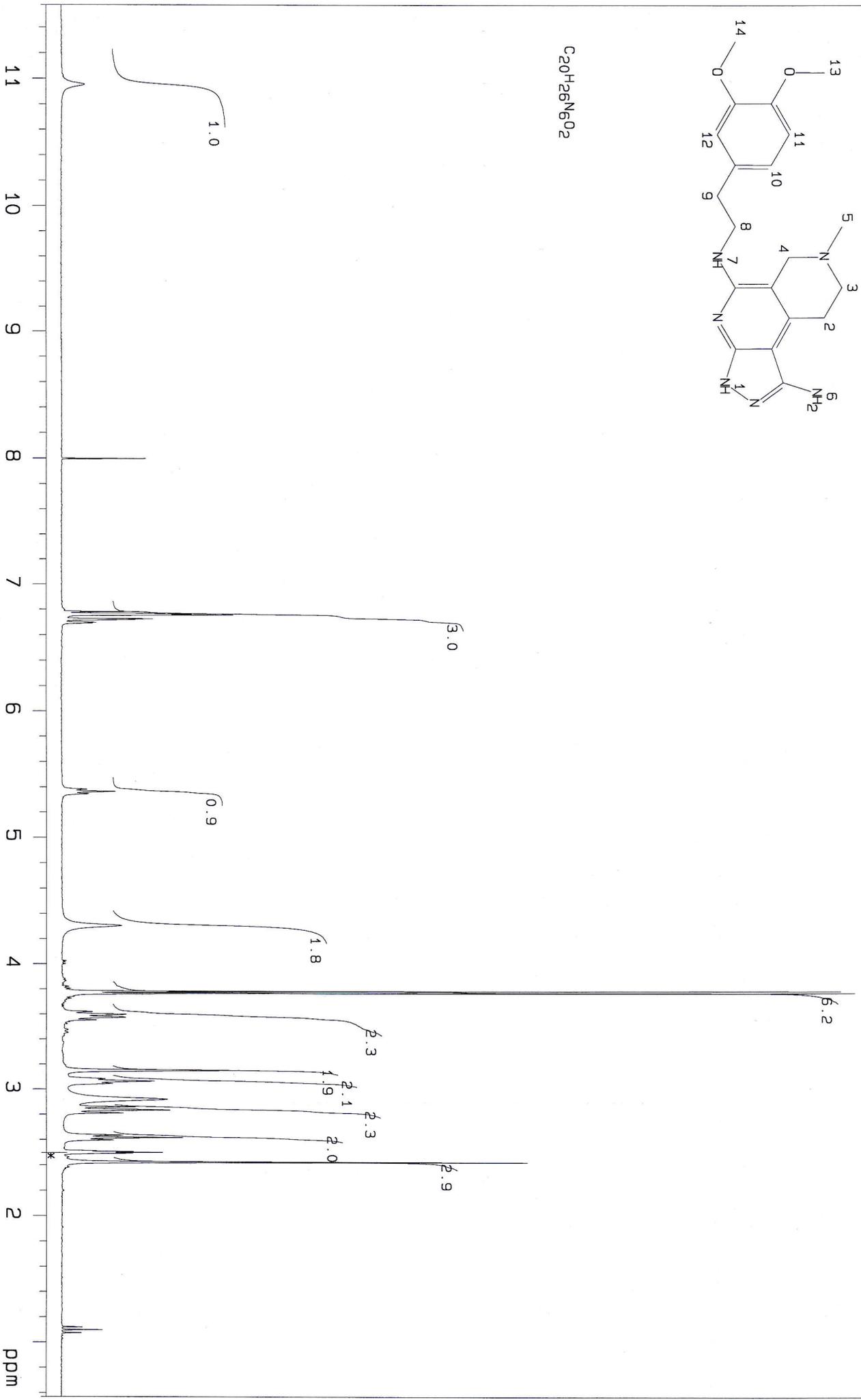
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ANUSH\_TEMA ae-s0958-0047

Oct 6 2021



C<sub>20</sub>H<sub>26</sub>N<sub>6</sub>O<sub>2</sub>



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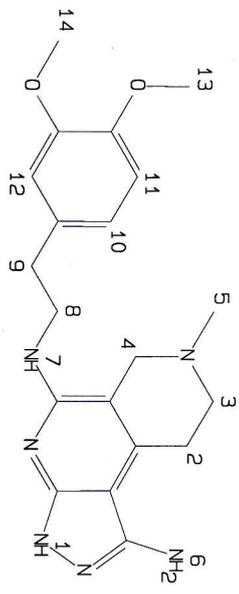
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Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
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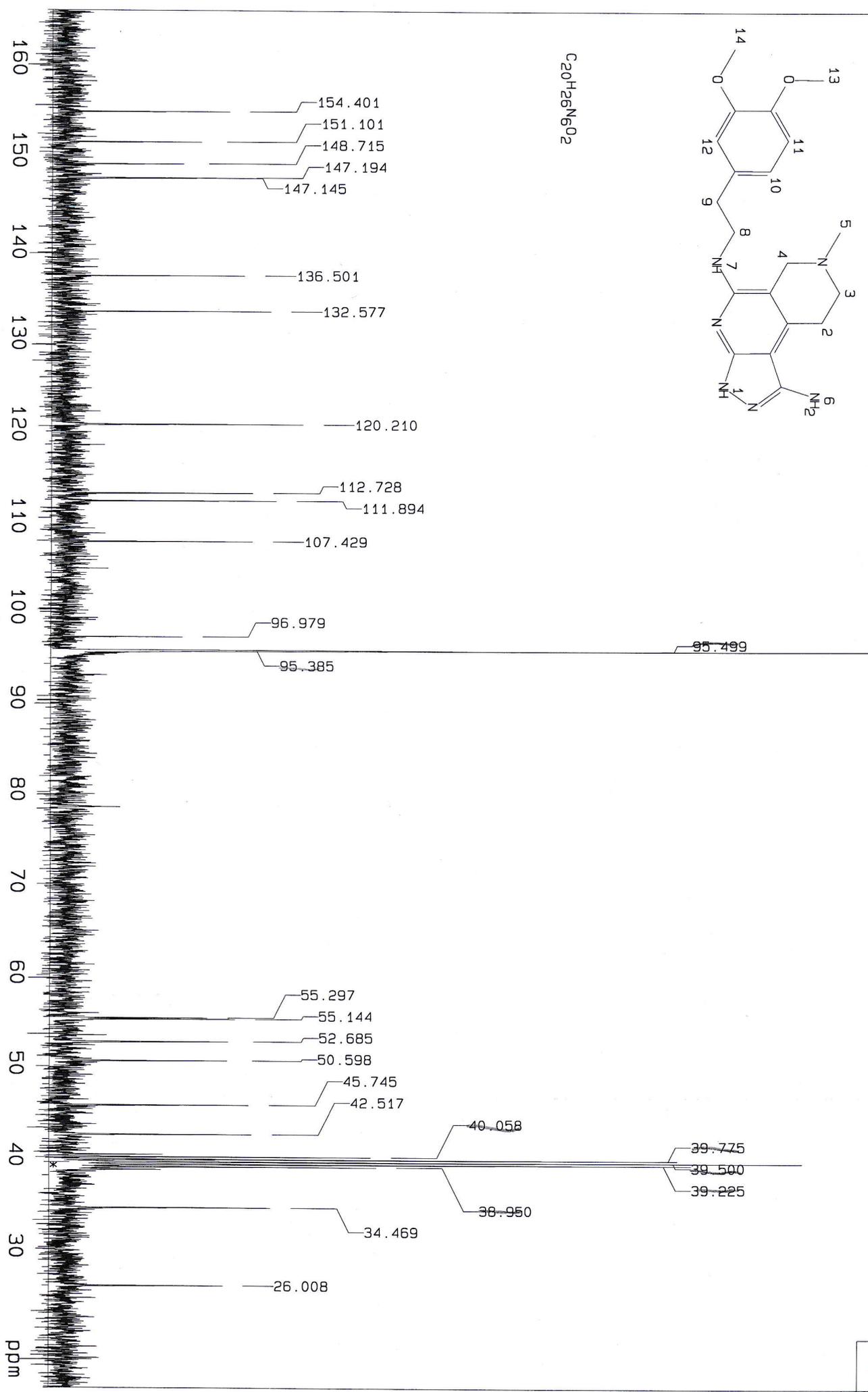
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ANUSH\_TEMA ae-s0958-0047

Oct 6 2021



C<sub>20</sub>H<sub>26</sub>N<sub>6</sub>O<sub>2</sub>



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3a

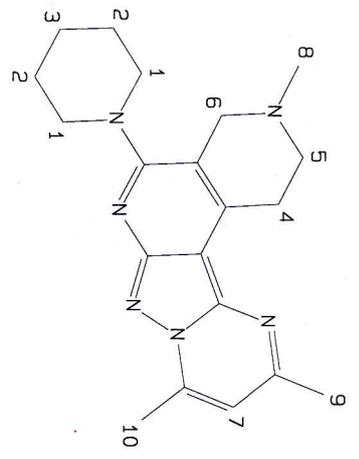
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

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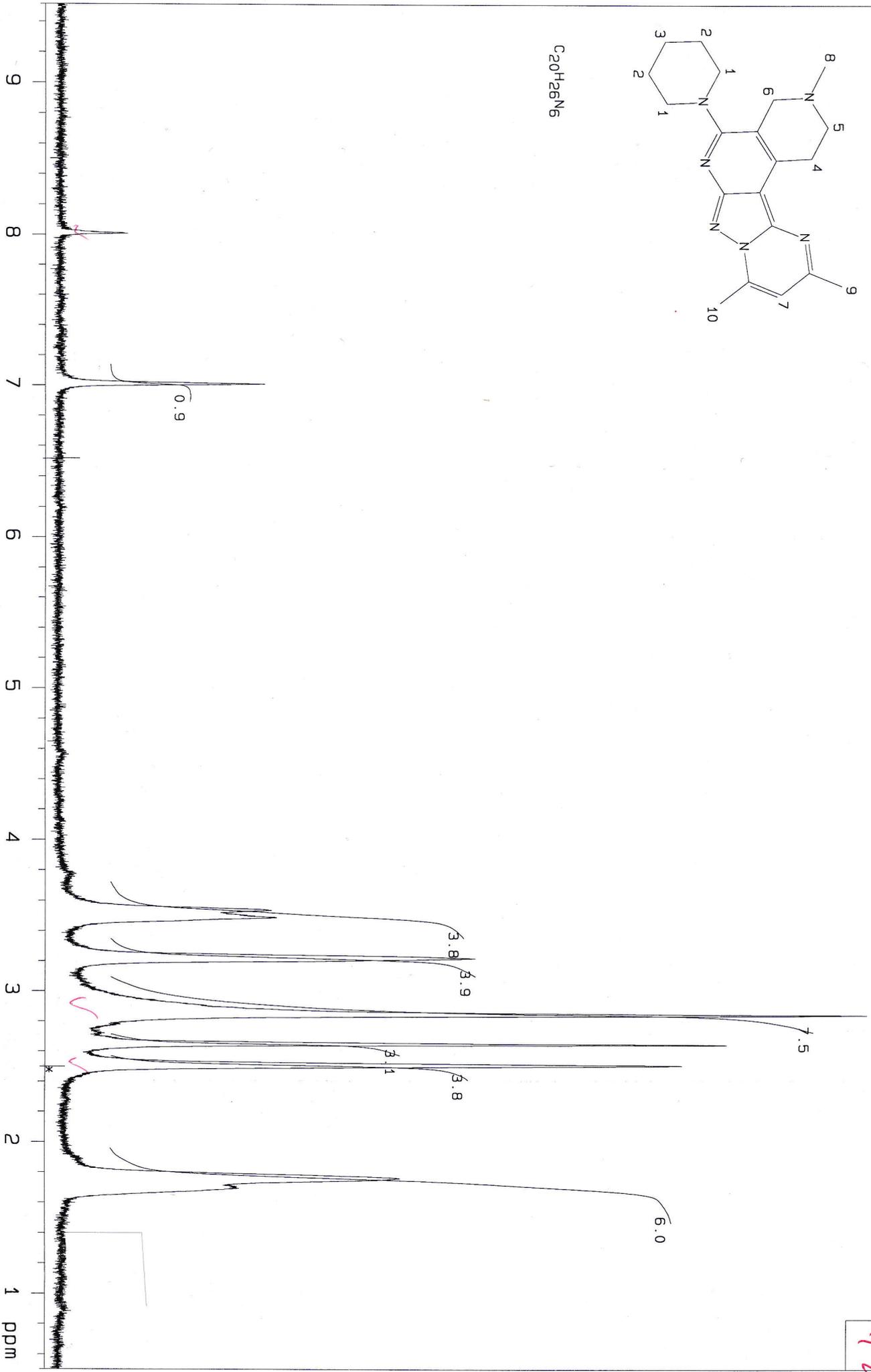
ANUSH\_TEMA ha-1077

Oct 13 2021

HA-1077



C<sub>20</sub>H<sub>26</sub>N<sub>6</sub>



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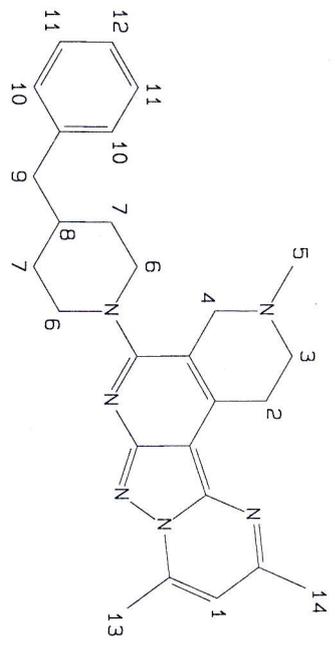
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

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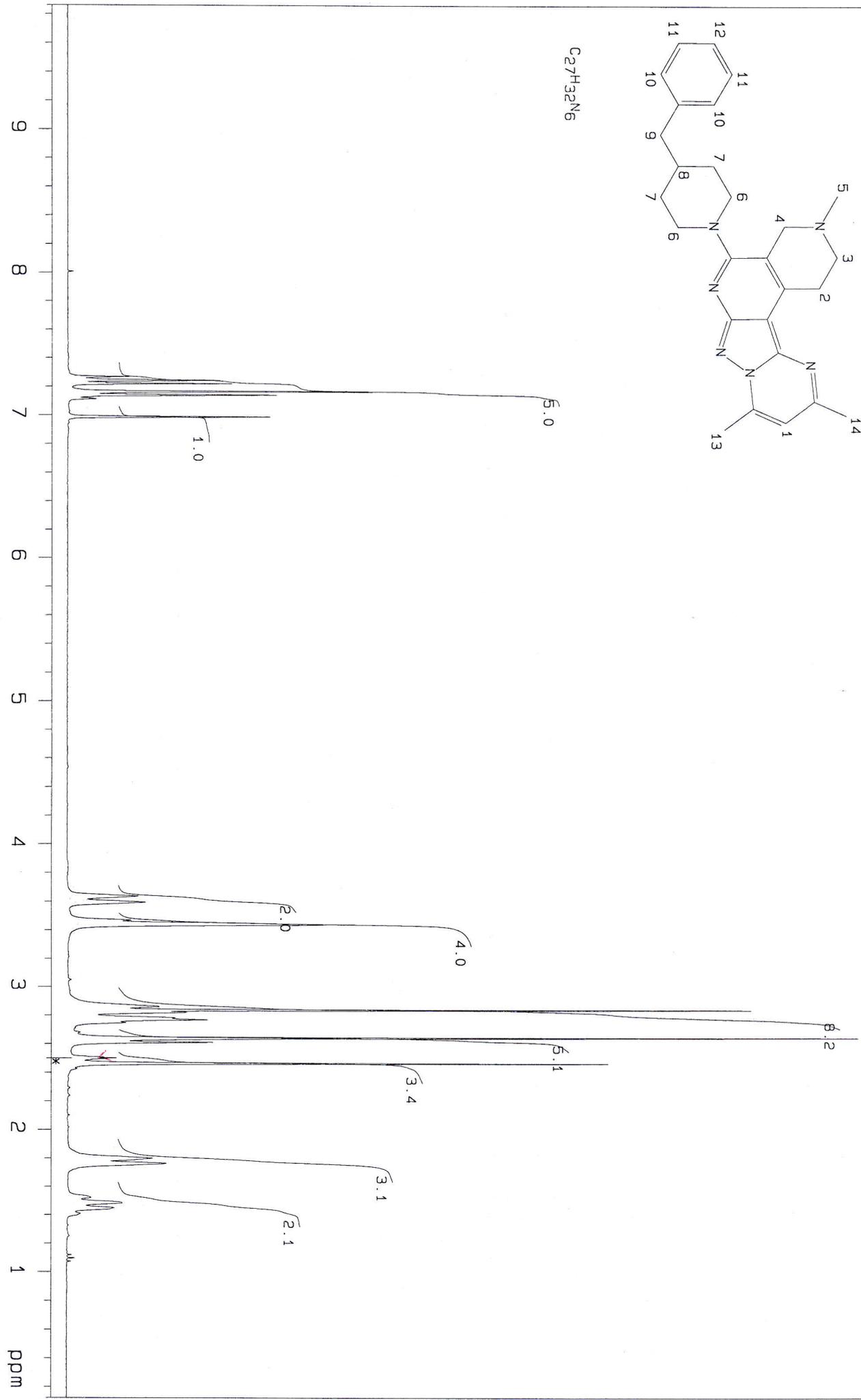
SAMV\_20 ha-1075

Feb 1 2020

HA-1075



C<sub>27</sub>H<sub>32</sub>N<sub>6</sub>



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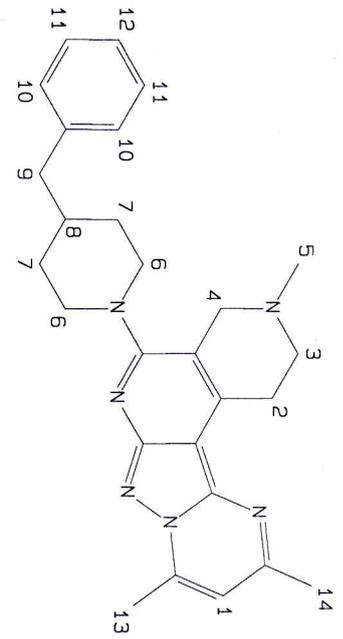
36

HA-1075

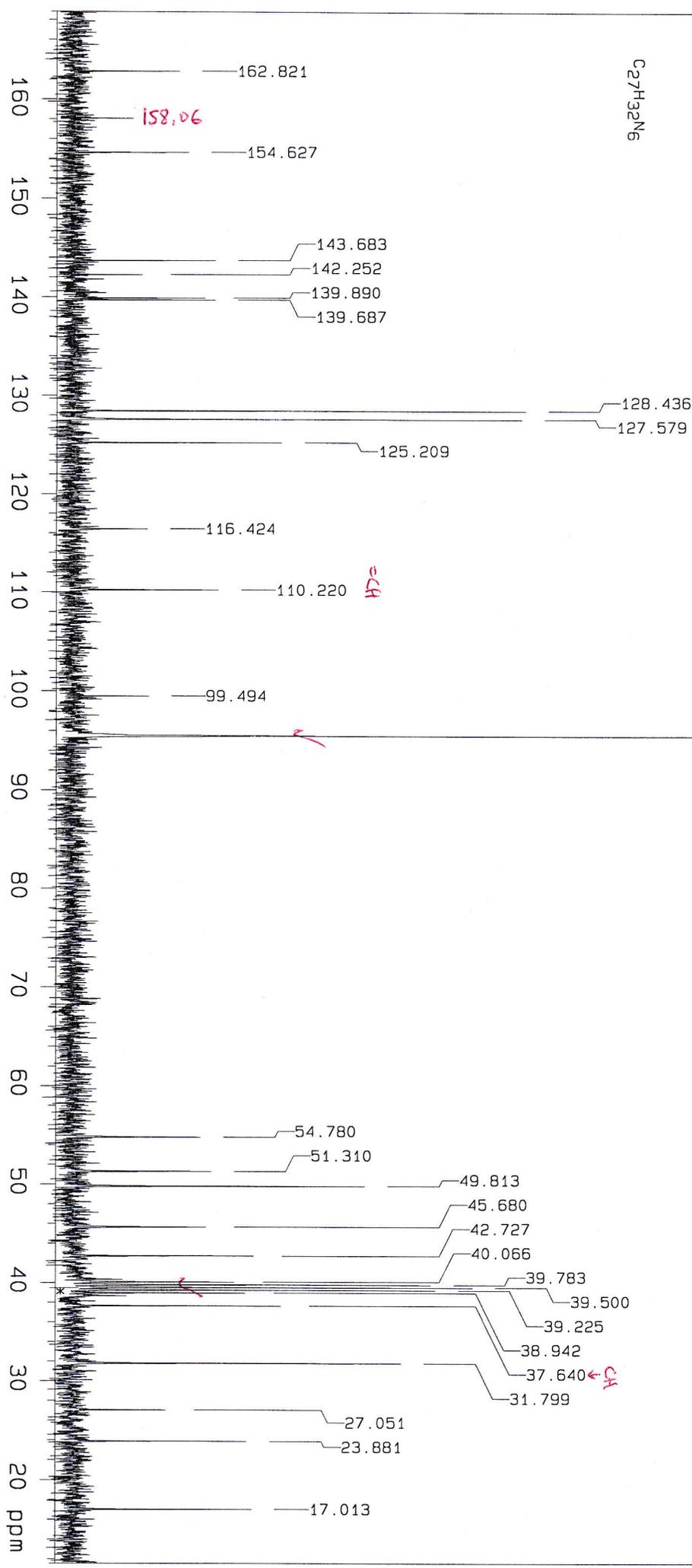
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SAMV\_20 ha-1075

Feb 1 2020



C<sub>27</sub>H<sub>32</sub>N<sub>6</sub>



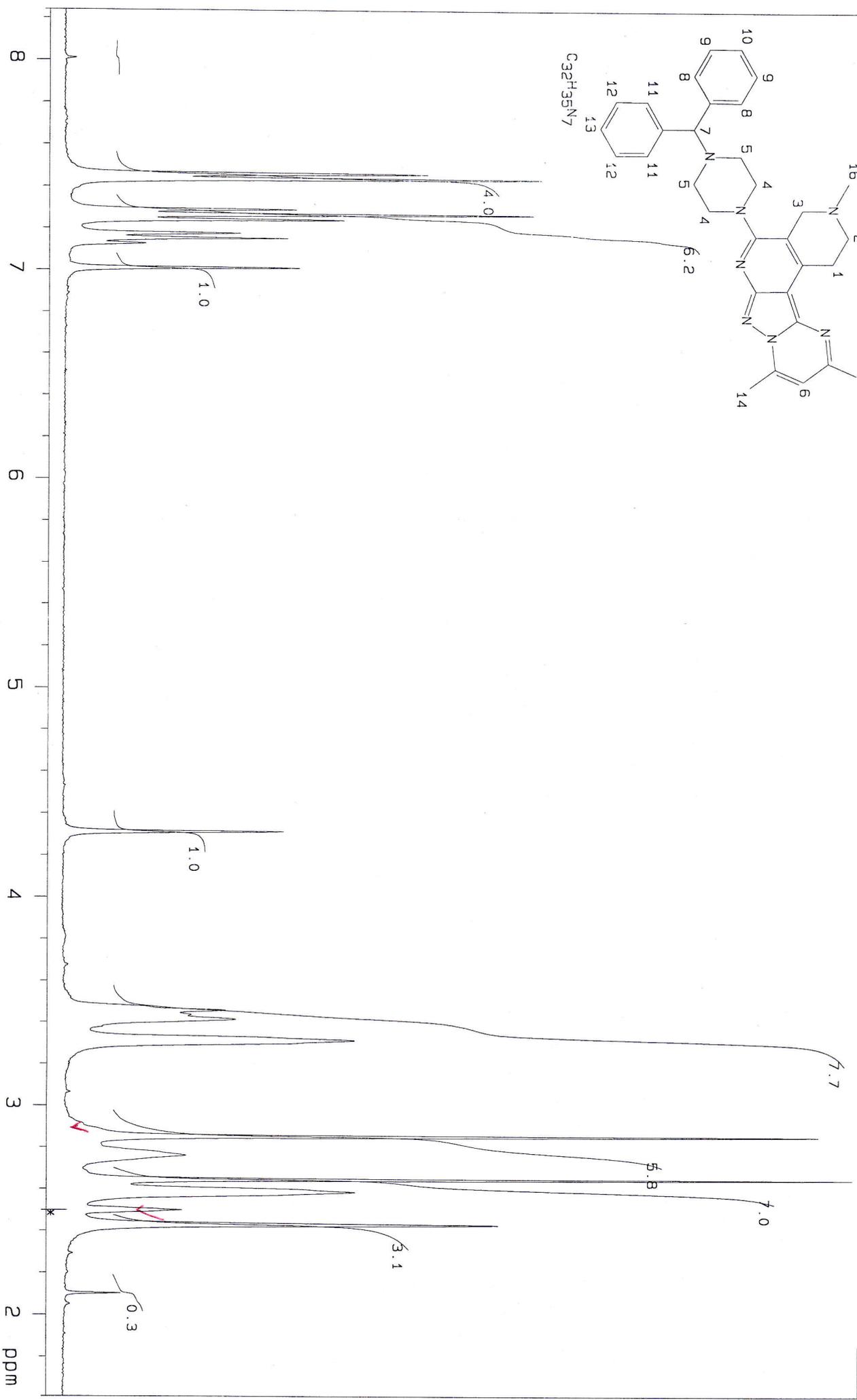
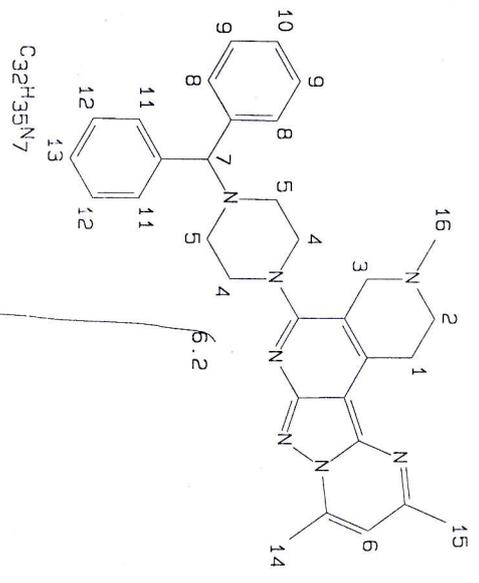
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3c

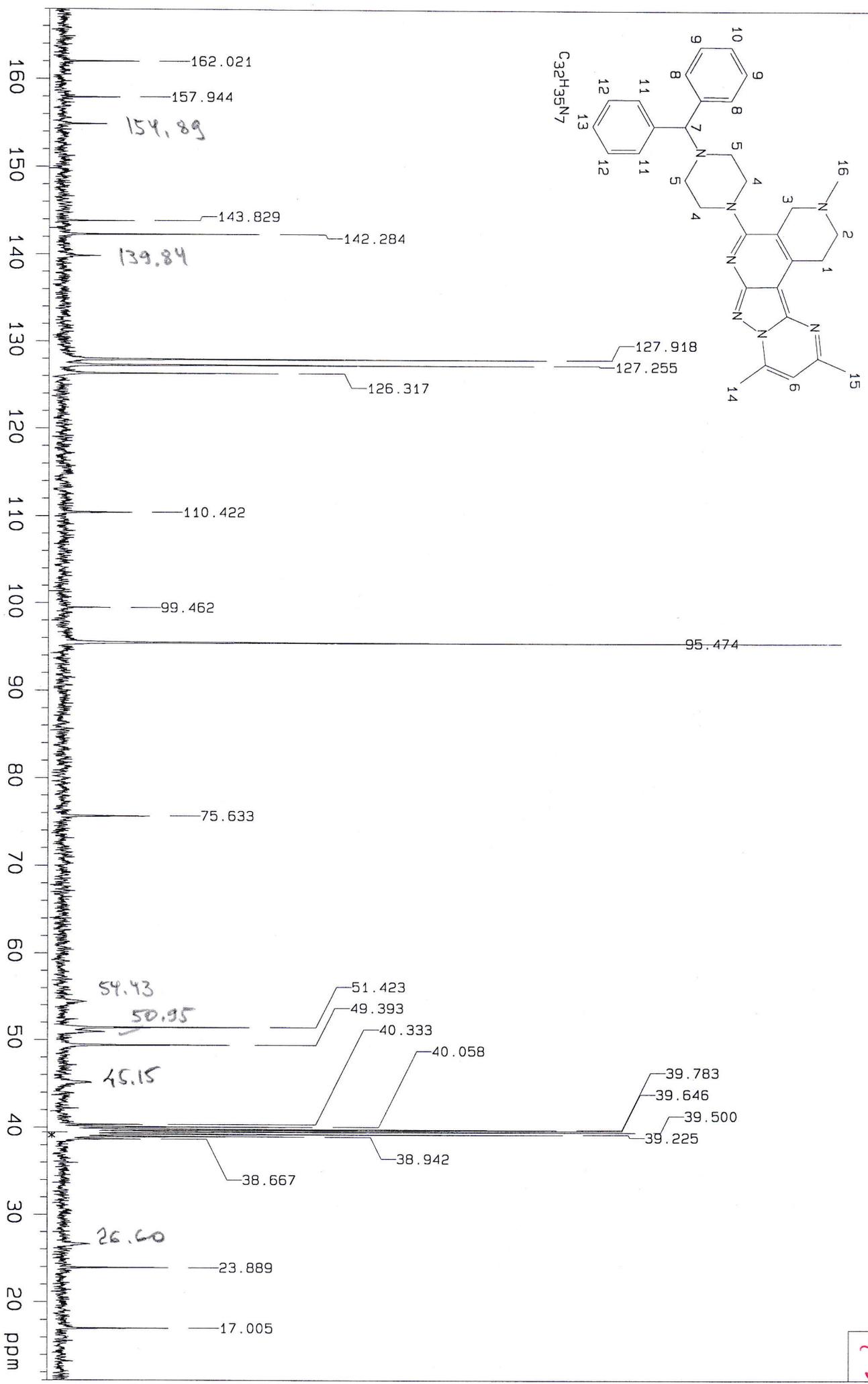
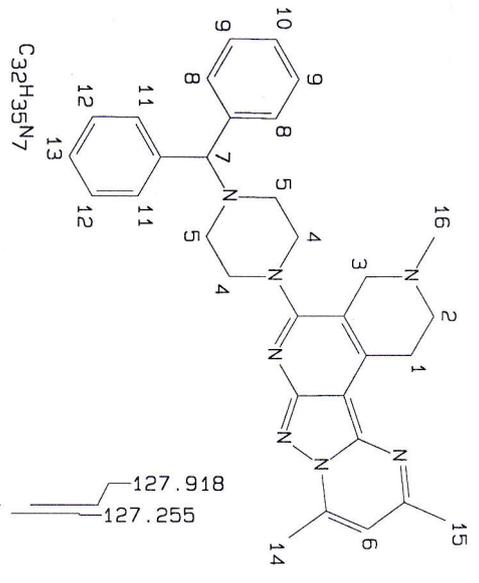
HA-1071

SAMV\_20 ha-1071

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3c



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301

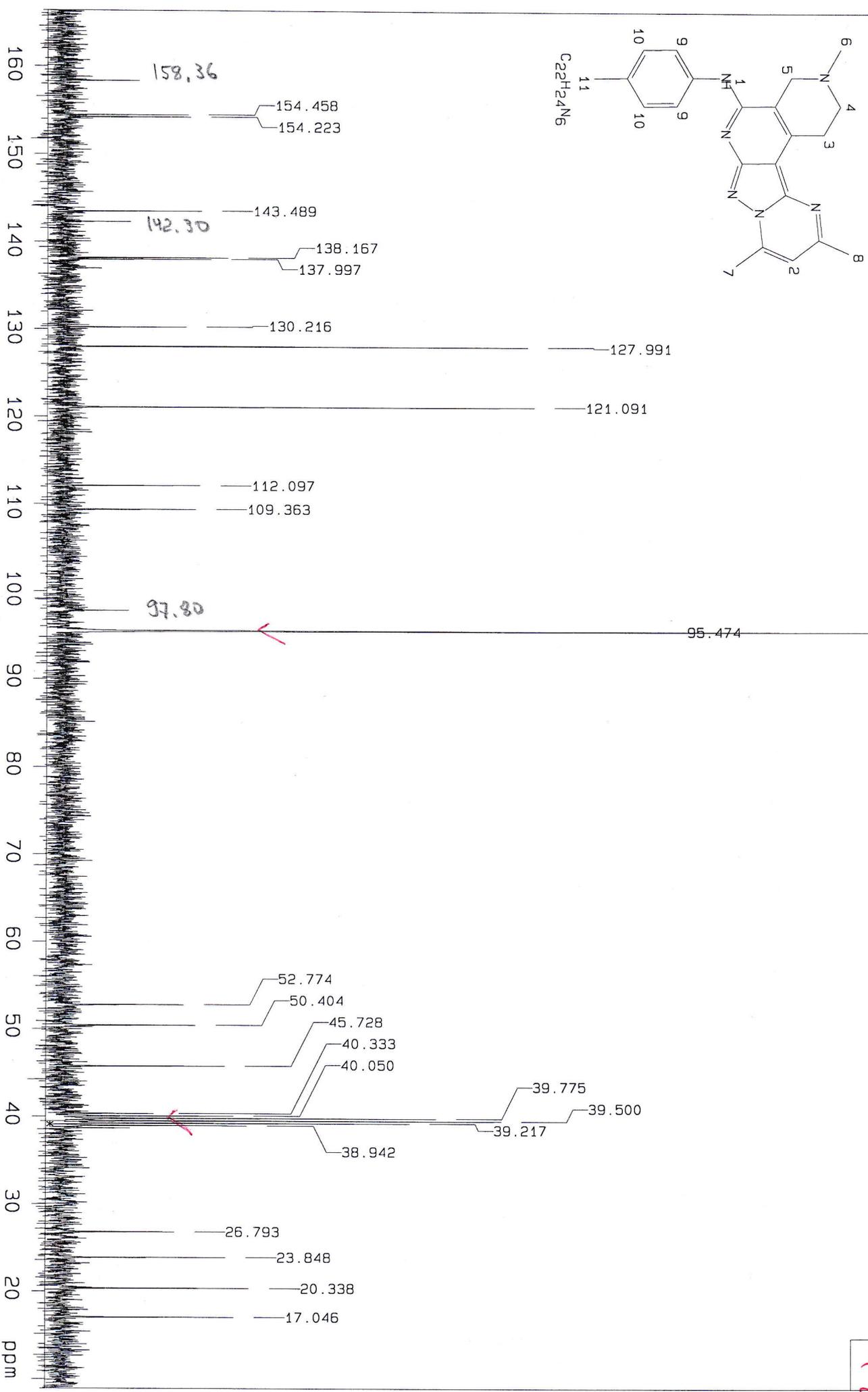
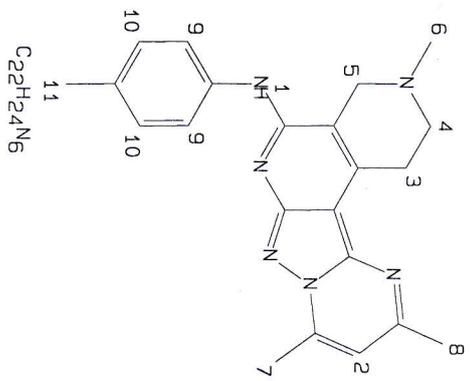
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

HA-1079

CD 3 75.464 MHz, nt = 352, np = 19998, temp = 30.0 C, lb = 1.0, solvent = DMSO/CD4 1/3

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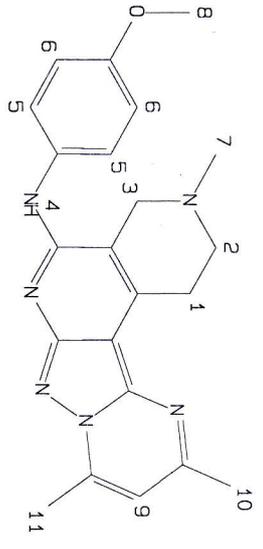
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

HA-1080

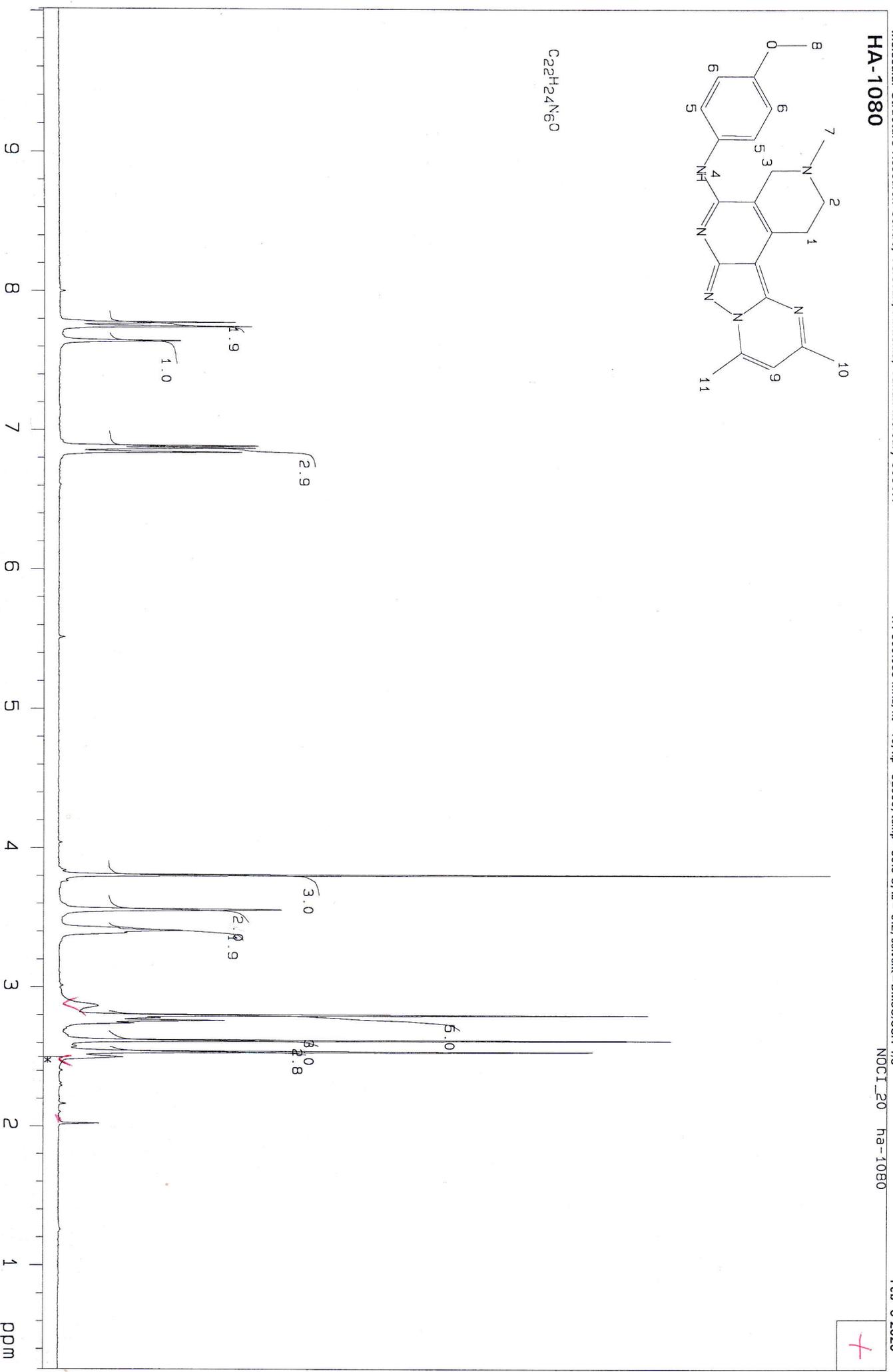
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NOCI\_20 ha-1080

Feb 6 2020



C<sub>22</sub>H<sub>24</sub>N<sub>6</sub>O



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*[Signature]*

3e

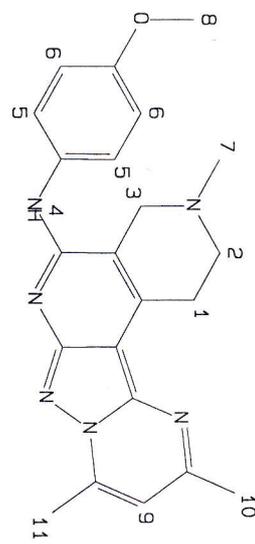
HA-1080

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

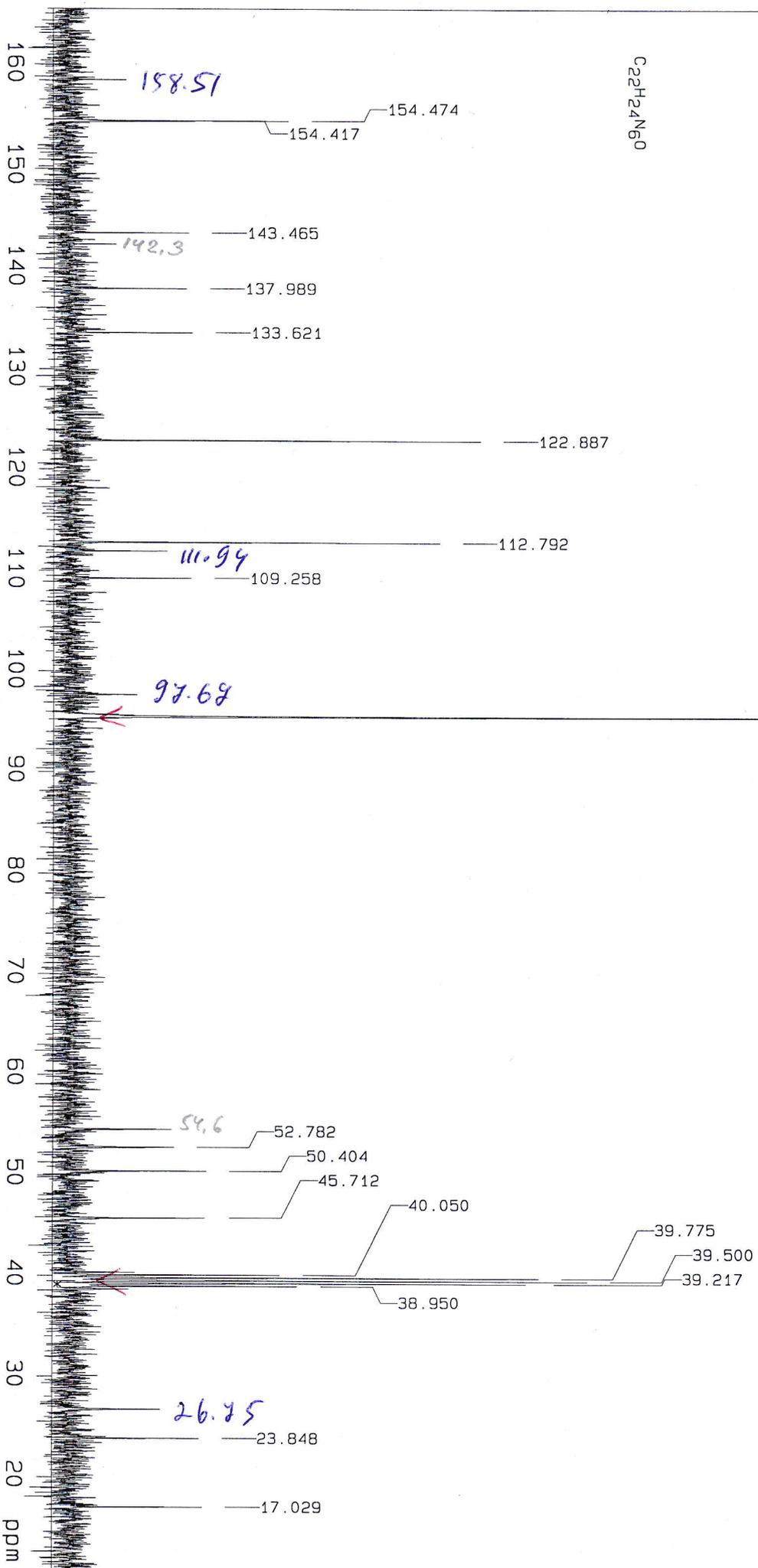
C13 75.464 MHz, nt=528, np=19998, temp=30.0 C, lb=1.0, solvent=DMSO/C14 1/3

NOCI\_20 ha-1080

Feb 6 2020



C<sub>22</sub>H<sub>24</sub>N<sub>6</sub>O



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3f

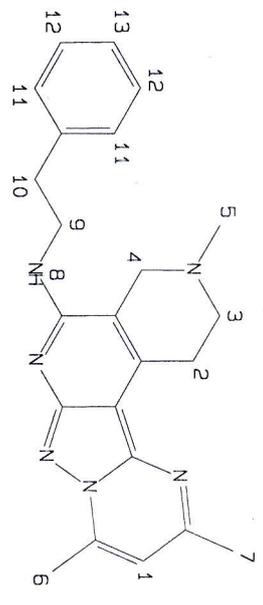
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

H1 300.088 MHz, nt = 16, np = 32000, temp = 30.0 C, lb = -0.2, solvent = DMSO-d6

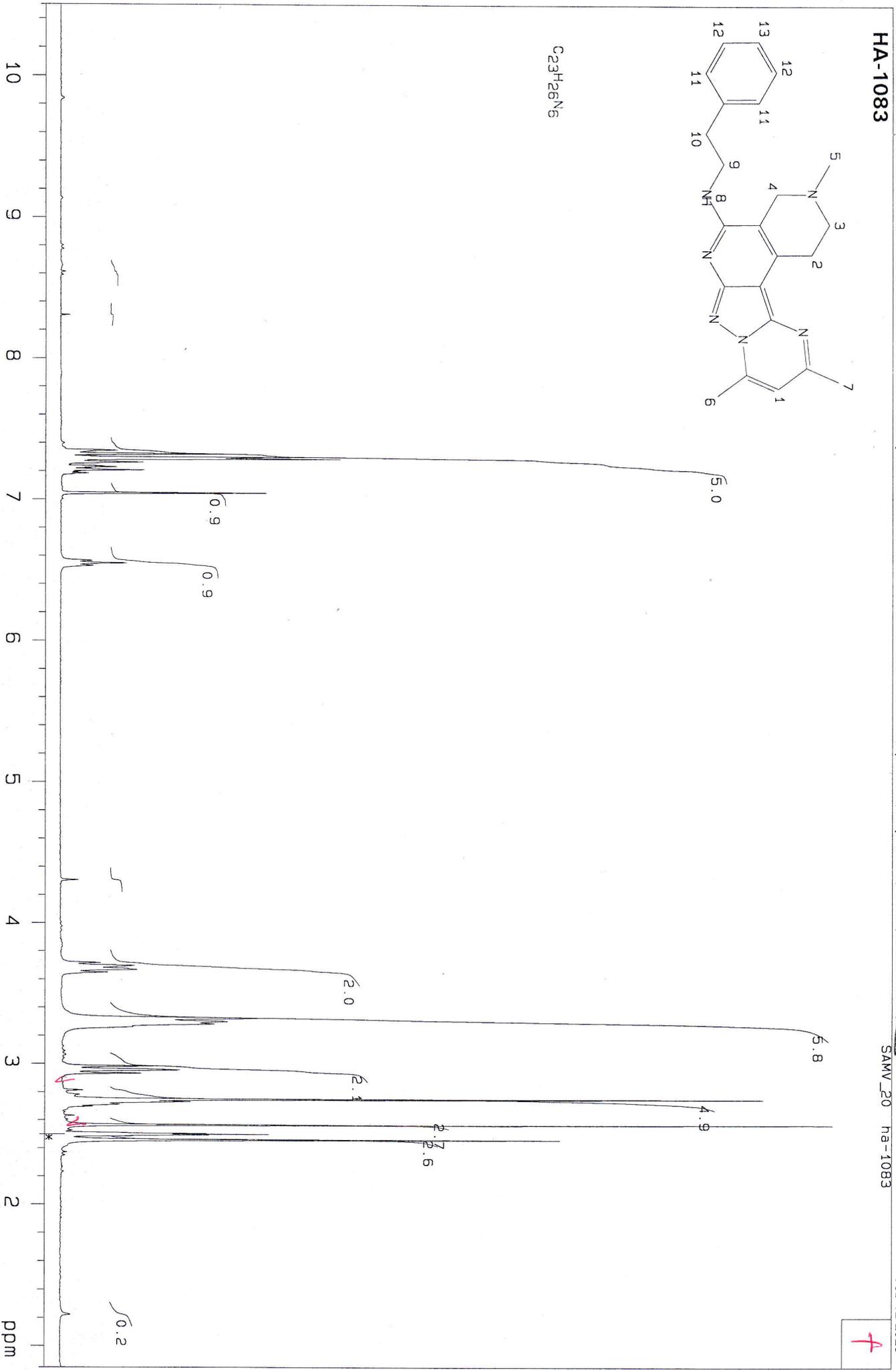
HA-1083

SAMV\_20 ha-1083

Feb 12 2020



C<sub>23</sub>H<sub>26</sub>N<sub>6</sub>



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38

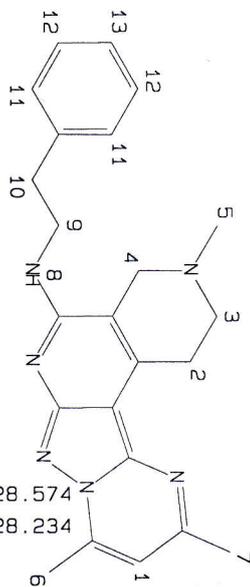
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

HA-1083

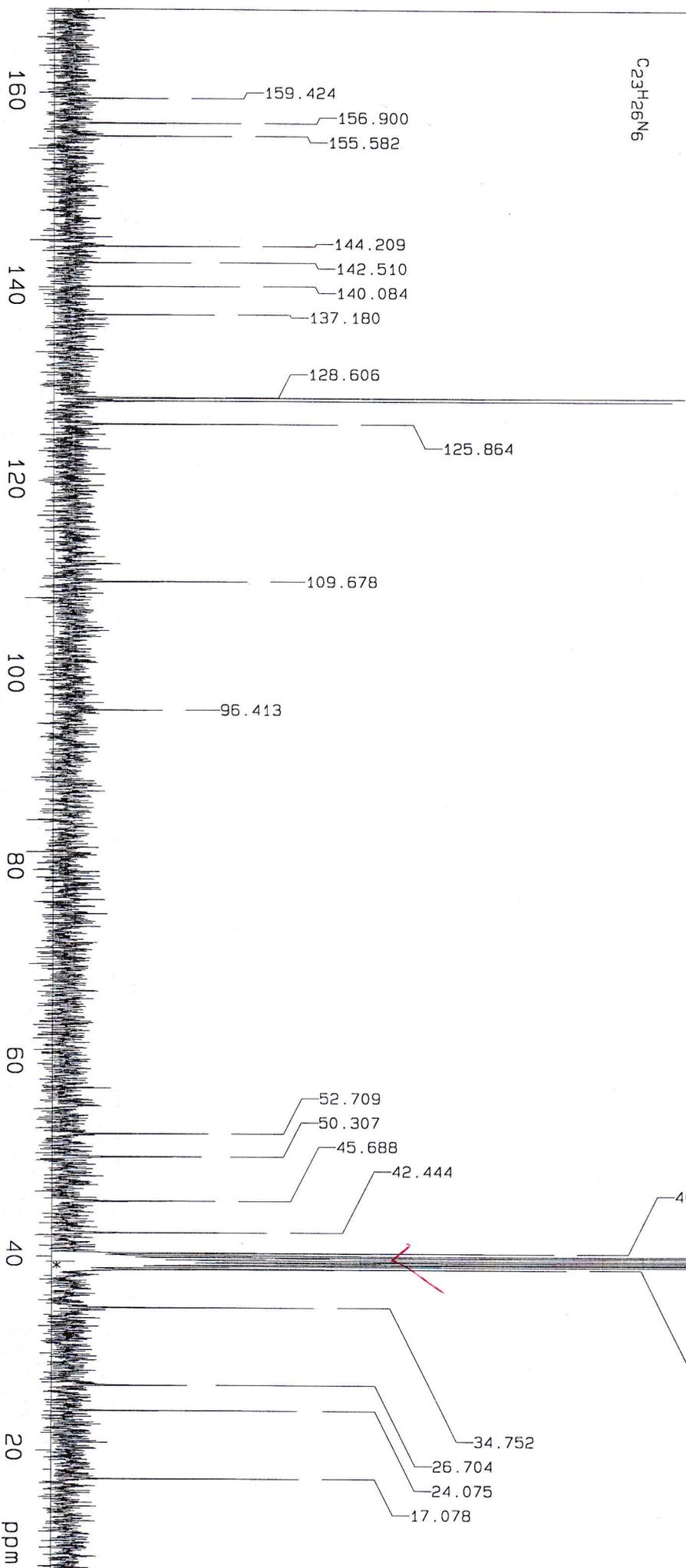
C13 75.465 MHz, nt = 1024, np = 19998, temp = 30.0 C, lb = 1.0, solvent = DMSO-d6

SAMV 20 ha-1083

Feb 12 2020



C<sub>23</sub>H<sub>26</sub>N<sub>6</sub>



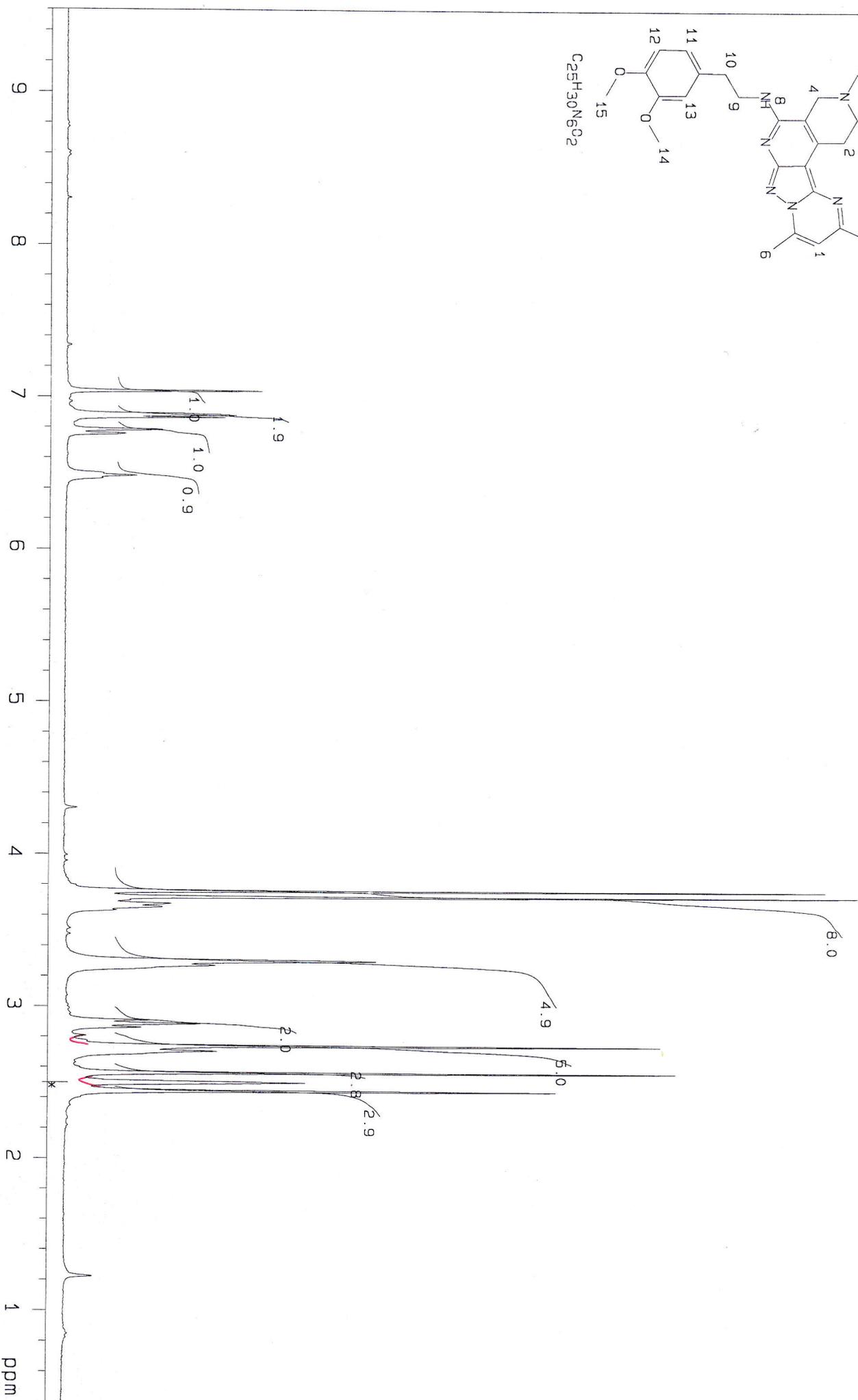
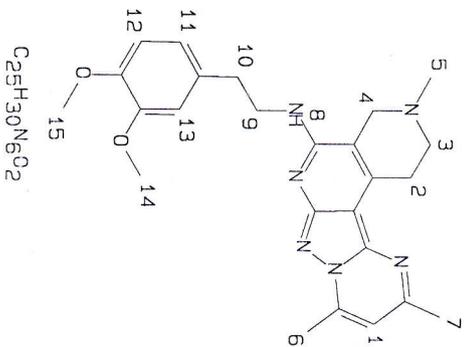
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39

HA-1081

SAMV\_20 ha-1081

Feb 13 2020



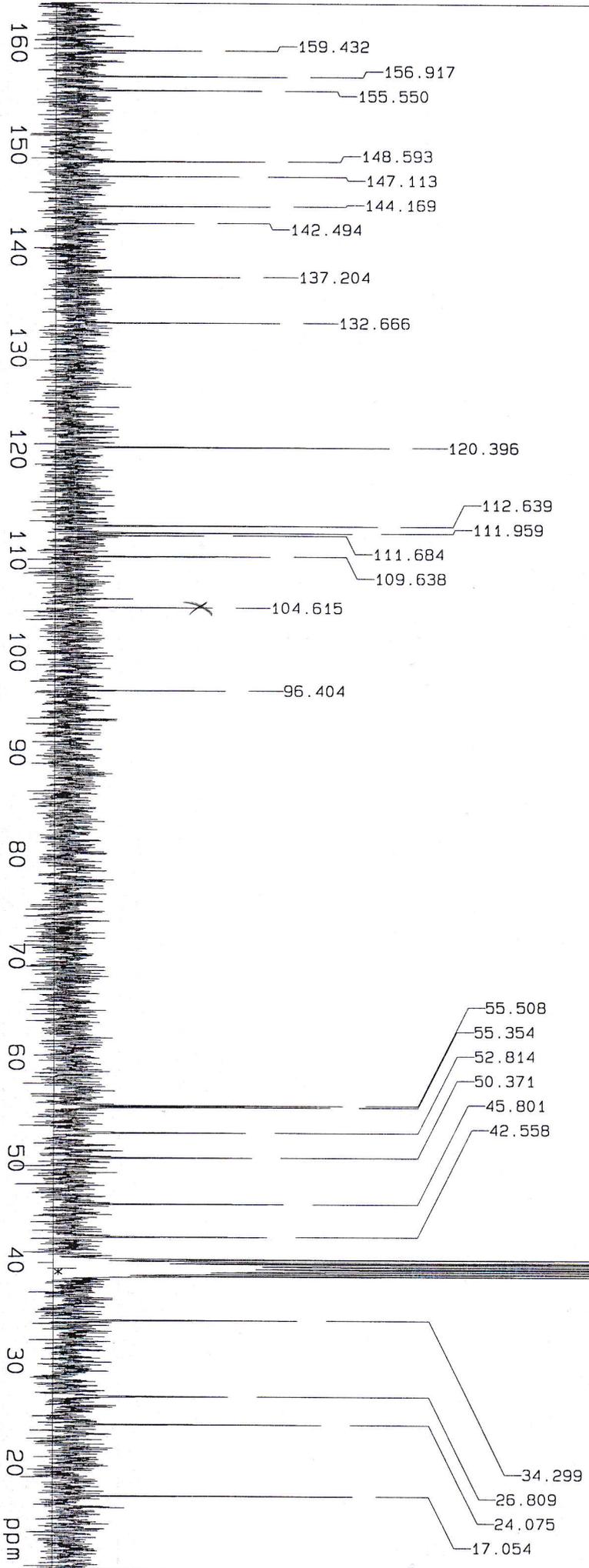
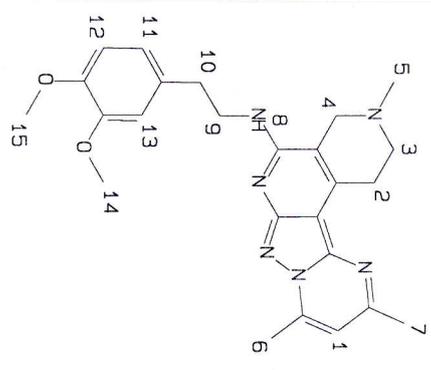
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39  
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
HA-1081

C13 75.465 MHz, nt=960, np=19998, temp=30.0 C, lb=1.0, solvent=DMSO-d6

NOCI\_20  
ha-1081

Feb 13 2020



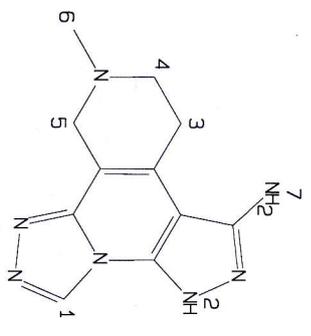
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5

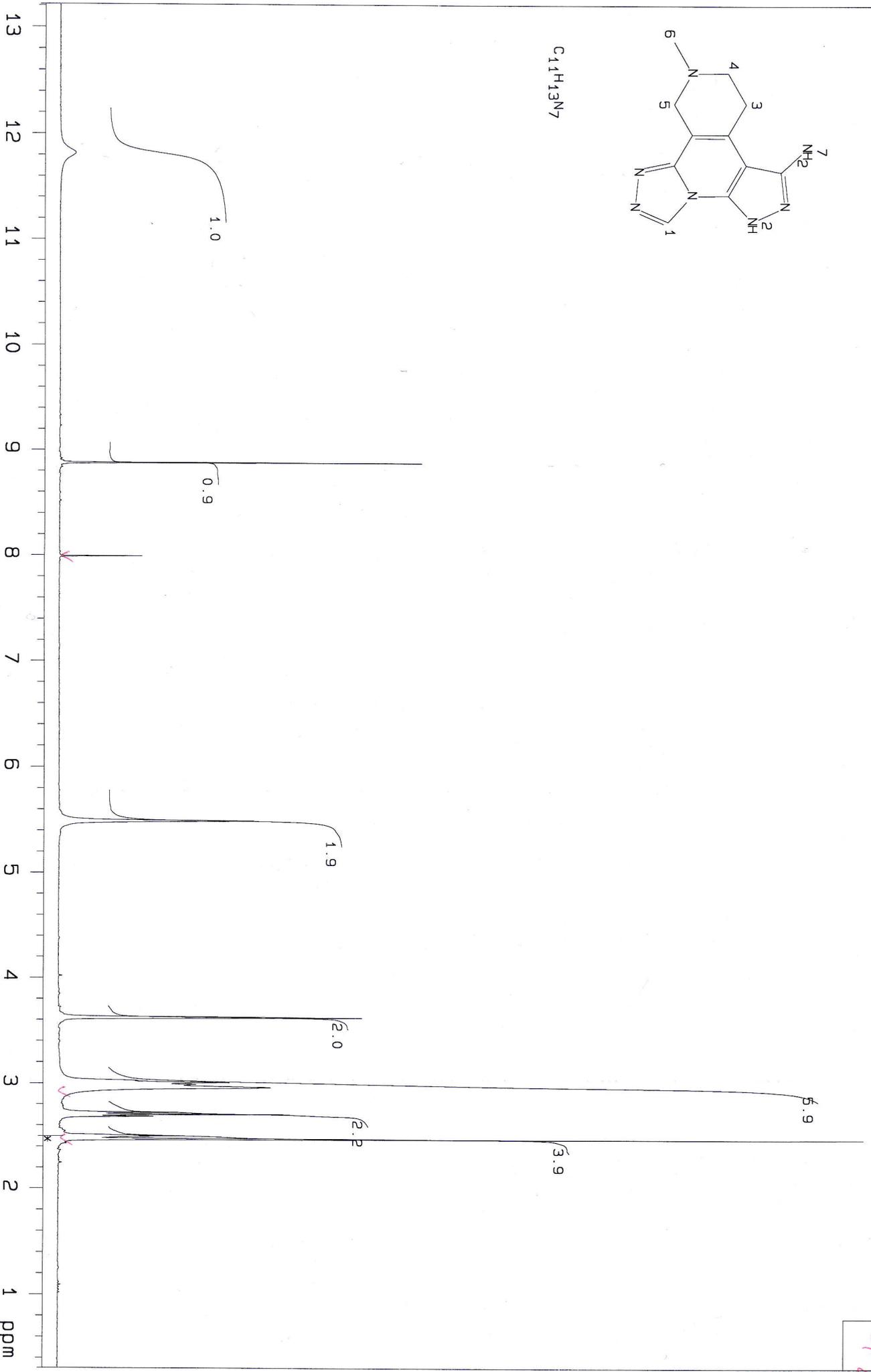
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
**NAR-024**

H1 300.088 MHz, nt = 16, mp = 32000, temp = 30.0 C, lb = -0.2, solvent = DMSO/CDCl4 1/3  
NOCI\_21 nar-024

Oct 12 2021



C<sub>11</sub>H<sub>13</sub>N<sub>7</sub>



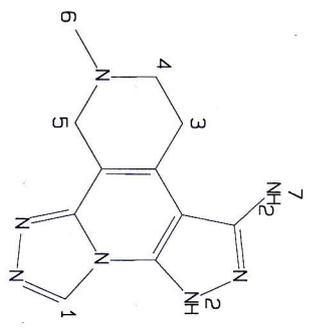
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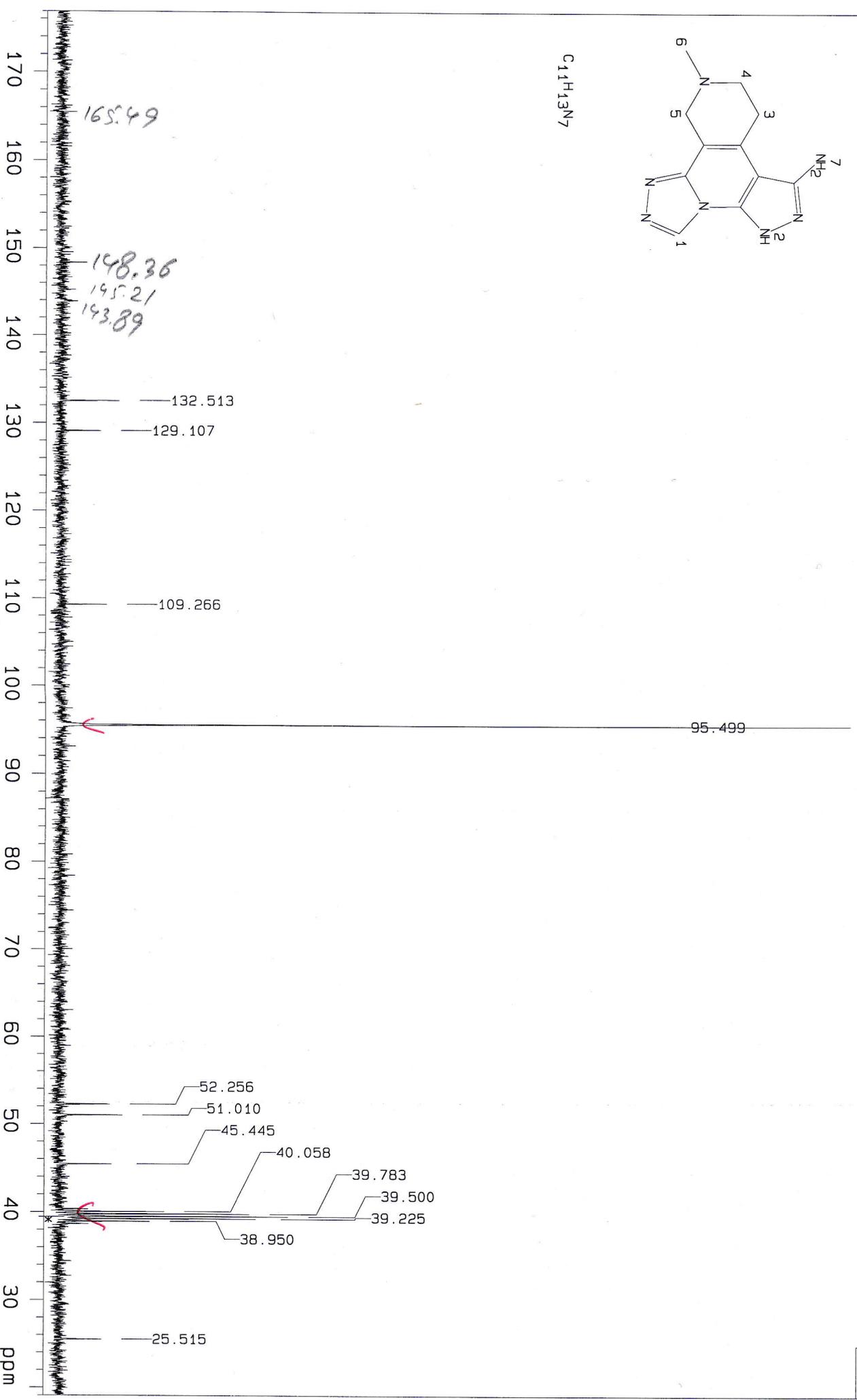
Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
**NAR-024**

C13 75.465 MHz, n=720, np=19998, temp=30.0 C, lb=1.0, solvent=DMSO/C14 1/3  
NOCI\_21 nar-024

Oct 12 2021



C<sub>11</sub>H<sub>13</sub>N<sub>7</sub>



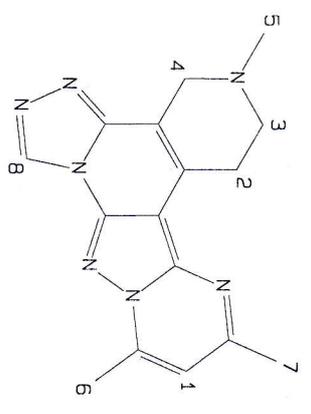
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Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
HA-1084-1A

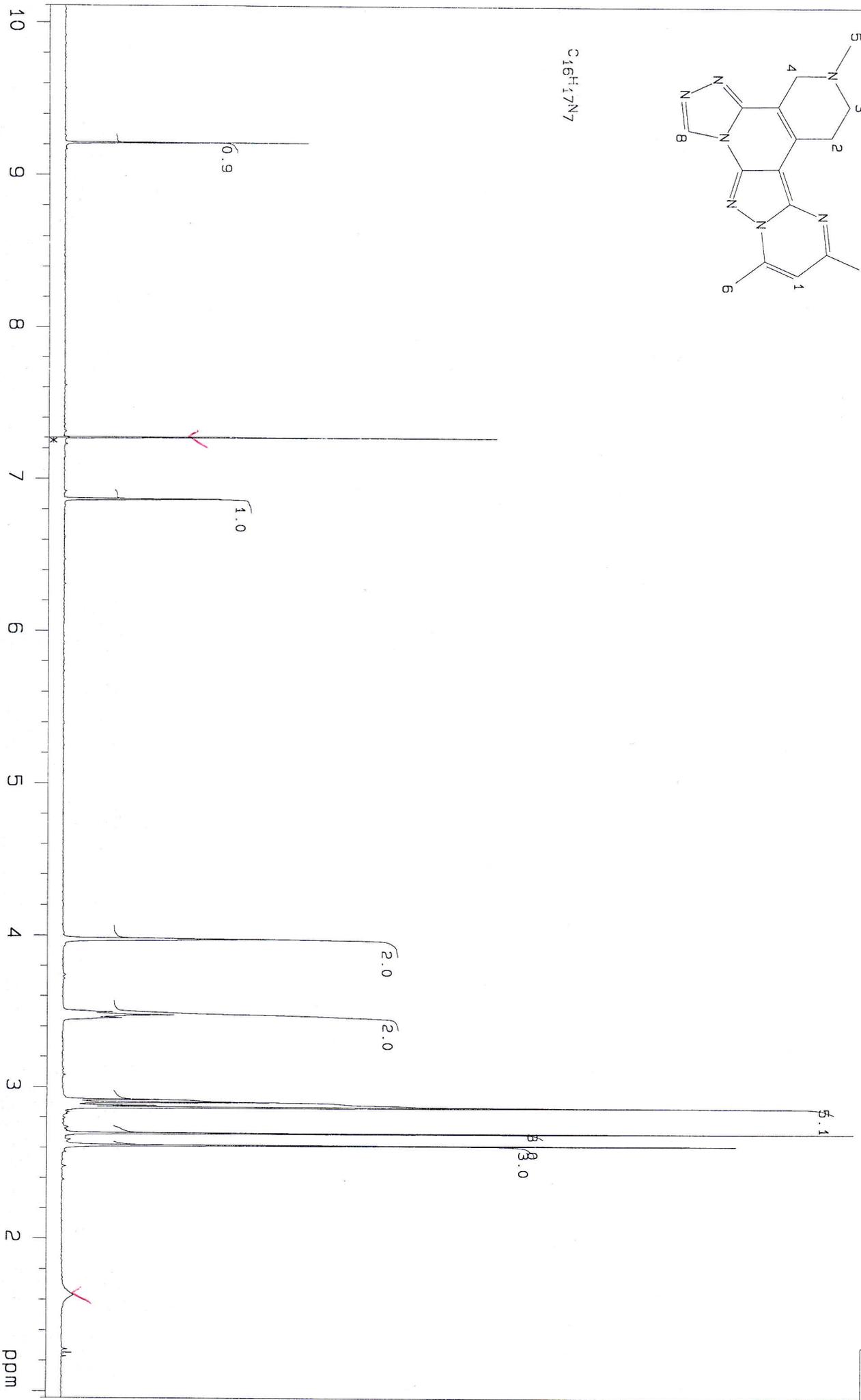
H1 300.086 MHz, nt = 16, np = 32000, temp = 30.0 C, lb = -0.2, solvent = CDCl3

SAMV\_20 ha-1084-1

Feb 13 2020



C<sub>16</sub>H<sub>17</sub>N<sub>7</sub>

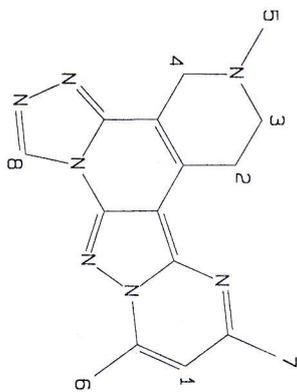


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[Signature]

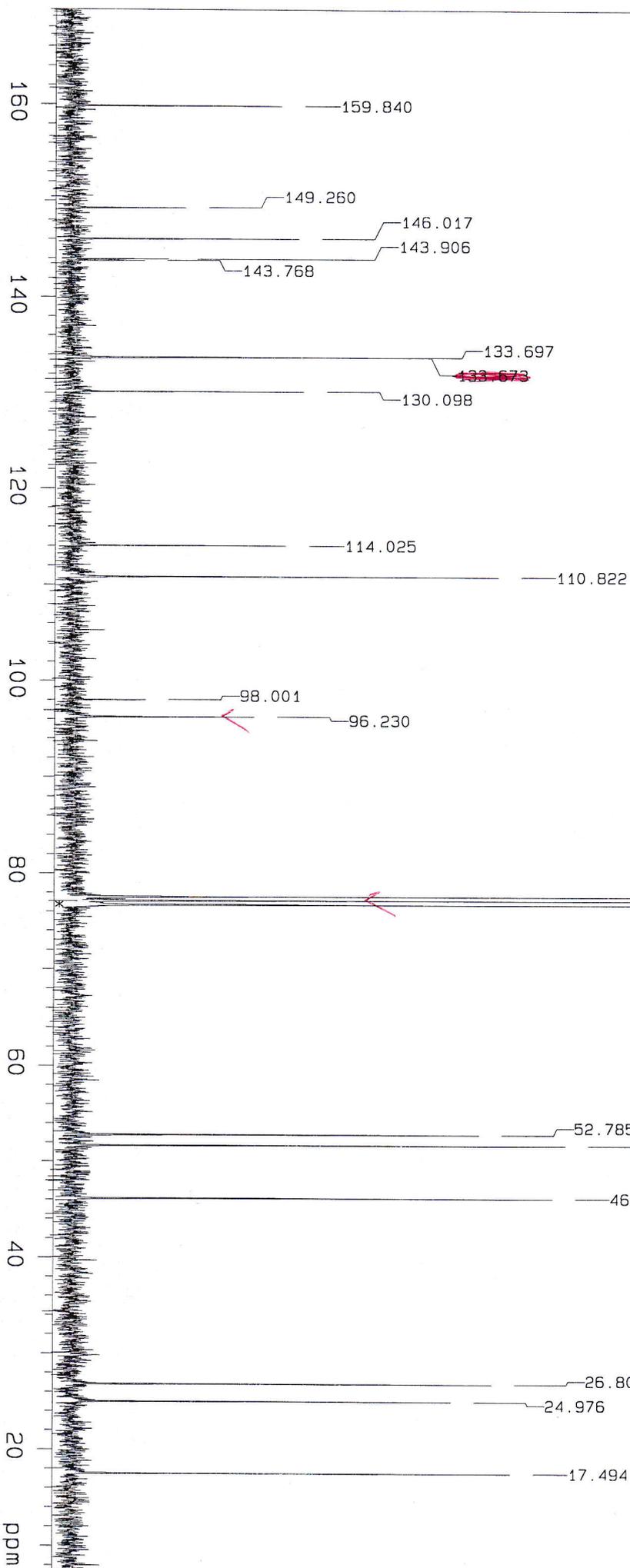
HA-1084-1

SAMV\_20 ha-1084-1

Feb 13 2020



C<sub>16</sub>H<sub>17</sub>N<sub>7</sub>



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*Signature*

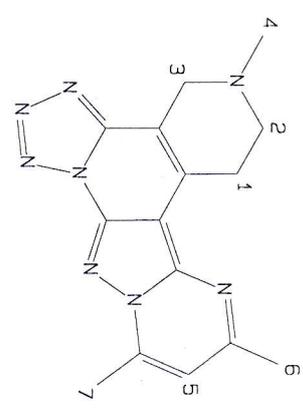
HA-1088

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX

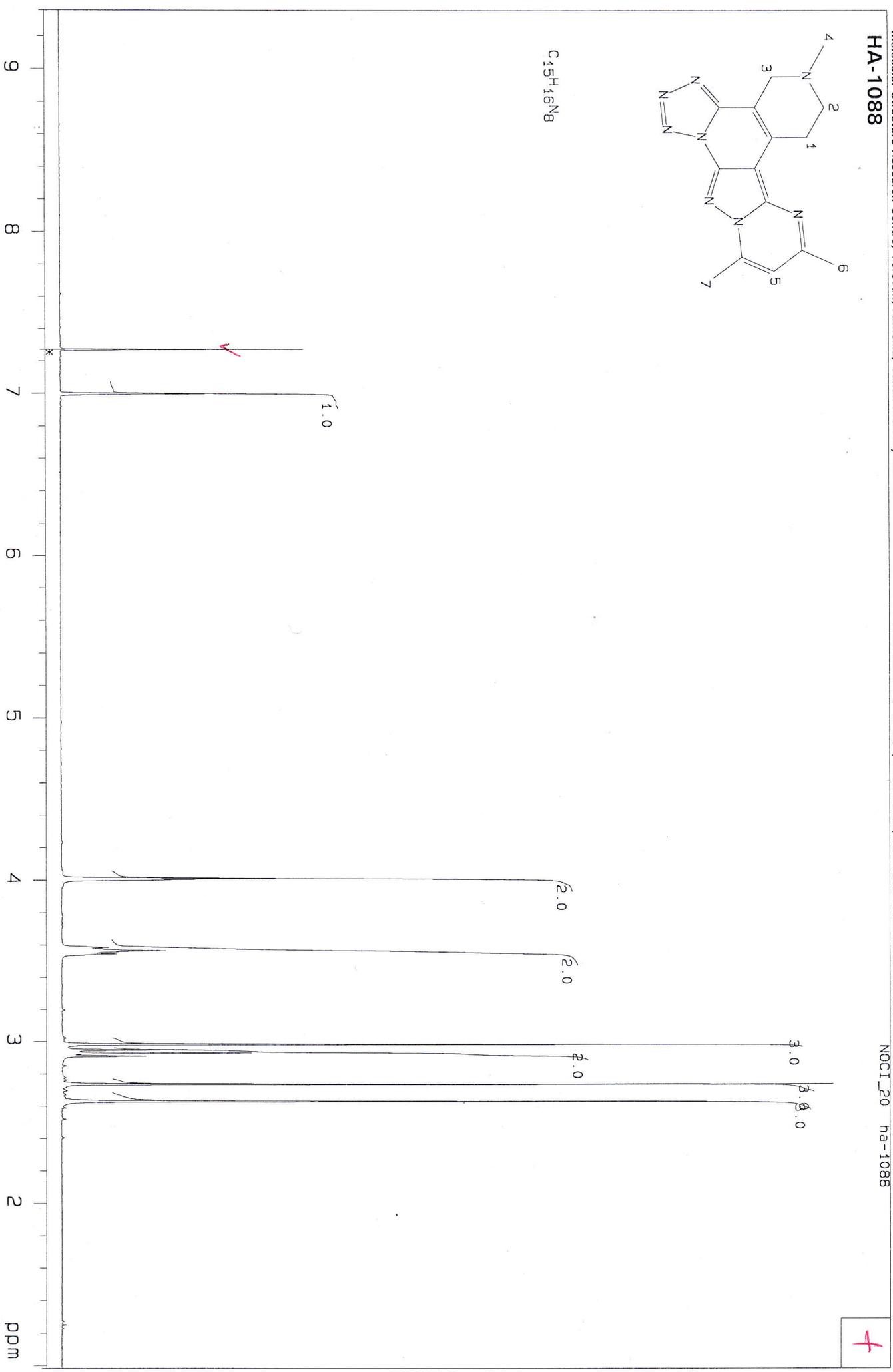
H1 300.086 MHz, nt = 16, np = 32000, temp = 30.0 C, lb = 0.2, solvent = CDCl3

NOCI\_20 ha-1088

Mar 4 2020



C<sub>15</sub>H<sub>16</sub>N<sub>8</sub>



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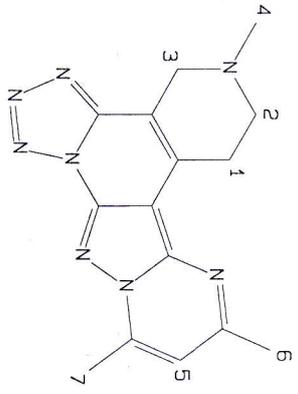
8

Molecular Structure Research Centre, Yerevan, Armenia, Varian Mercury-300VX  
HA-1088

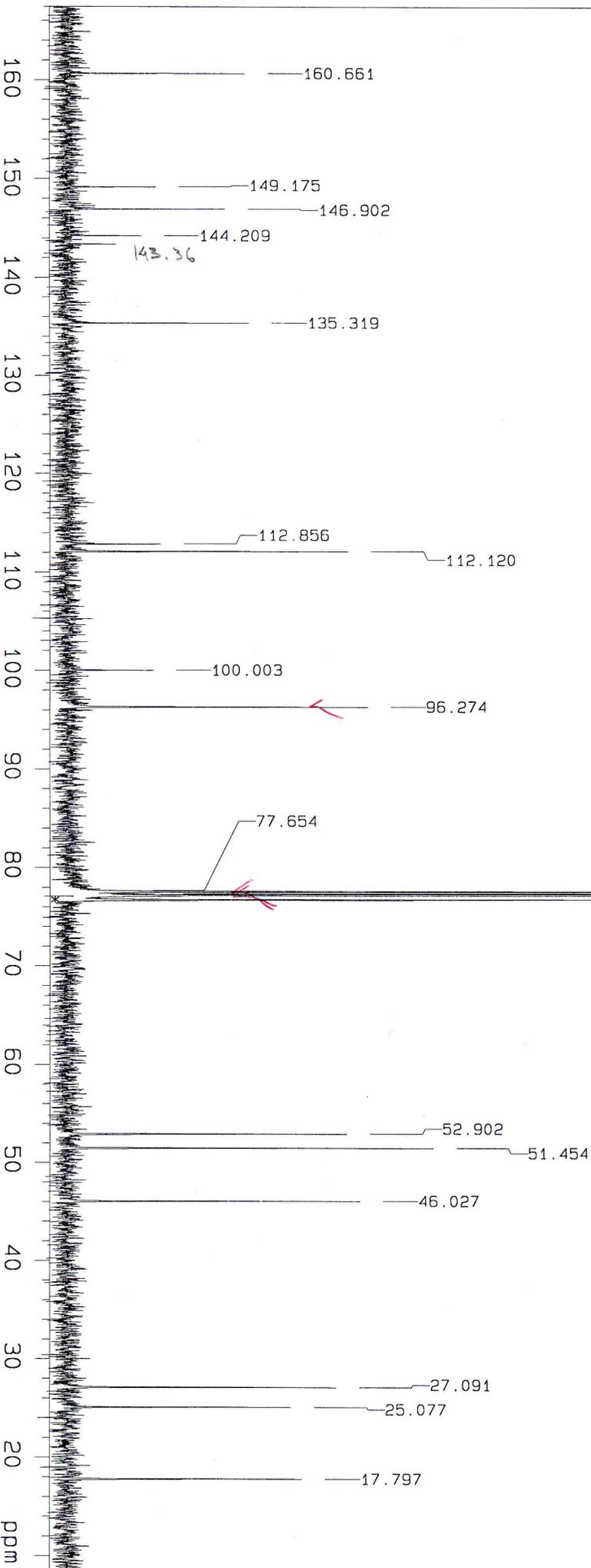
C13 75.464 MHz, nt = 3536, nr = 19998, temp = 30.0 C, lb = 1.0, solvent = CDCl3

NOCI\_20 na-1088

Mar 4 2020



C<sub>15</sub>H<sub>16</sub>N<sub>8</sub>



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