

## Supporting Information

# Selective Autoxidation of Benzylamines: Application to the Synthesis of Some Nitrogen Heterocycles

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## General information

Reagents were obtained from commercial supplier and used without further purification. Analytical thinlayer chromatography (TLC) was purchased from Merck KGaA (silica gel 60 F254). Visualization of the chromatogram was performed by UV light (254 nm) or phosphomolybdic acid or vanilline stains. Flash column chromatography was carried out using kieselgel 35-70 µm particle sized silica gel (230-400 mesh). NMR Chemical shifts are reported in ( $\delta$ ) ppm relative to tetramethylsilane (TMS) with the residual solvent as internal reference ( $\text{CDCl}_3$ ,  $\delta$  7.26 ppm for  $^1\text{H}$  and  $\delta$  77.0 ppm for  $^{13}\text{C}$ ;  $\text{DMSO-d}_6$ ,  $\delta$  2.50 ppm for  $^1\text{H}$  and  $\delta$  39.5 ppm for  $^{13}\text{C}$ ;  $\text{CD}_3\text{OD}$ ,  $\delta$  3.31 ppm for  $^1\text{H}$  and  $\delta$  49.0 ppm for  $^{13}\text{C}$ ). Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet), coupling constants (Hz) and integration.

## General procedures

### Oxydation of benzylamines (1) to *N*-benzylbenzaldimine (2)

A 20 mL tube containing benzylamine **1** (5 mmol) was flushed with  $\text{O}_2$  and equipped with an  $\text{O}_2$  balloon. The reaction was stirred at 100 °C for 24 h. In cases where purification was necessary, the reaction crude could be rapidly chromatographed on silica gel column previously washed with a solution of triethylamine (0.1% volumn) in heptane.

### Synthesis of benzimidazole (4)

A 20 mL tube containing benzylamine **1** (6 mmol), *o*-phenylenediamine **2** (5 mmol) and AcOH (1 mmol, 60 mg) was flushed with  $\text{O}_2$  and equipped with an  $\text{O}_2$  balloon. The reaction

was stirred at 110 °C for 24 h. The solid crude mixture was stirred vigorously with CH<sub>2</sub>Cl<sub>2</sub> (2 mL) or diethyl ether (2 mL) or CH<sub>2</sub>Cl<sub>2</sub> : heptane (2 mL : 2 mL), filtered and washed with the same solvent mixture (2 mL × 3). The solid residue was dissolved in MeOH, filtered through a short pad of celite. The filtrate was concentrated to afford the crystalline product. In case of **4ah**, the product was chromatographed by silica gel column (heptane : AcOEt 4 : 1).

### Synthesis of benzothiazole (**6**)

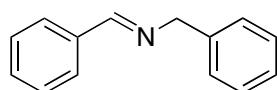
A 20 mL tube containing benzylamine **1** (10 mmol), *o*-aminothiophenol **5** (5 mmol) and AcOH (1 mmol, 60 mg) was flushed with O<sub>2</sub> and equipped with an O<sub>2</sub> balloon. The reaction was stirred at 100 °C for 24 h. The product was chromatographed by silica gel column (heptane : AcOEt 98 : 2).

### Synthesis of 2,3-dihydroquinazolin-4(1*H*)-one (**8**) and quinazolin-4(3*H*)-one (**9**)

A 20 mL tube containing benzylamine **1** (5 mmol), anthranilamide **7** (2.5 mmol) and AcOH (1 mmol, 60 mg) was flushed with O<sub>2</sub> and equipped with an O<sub>2</sub> balloon. The reaction was stirred at 120 °C or 150 °C for 24 h (See Table 4 of the manuscript). The solid crude mixture was stirred vigorously with CH<sub>2</sub>Cl<sub>2</sub> (2 mL) (for **8**) or EtOH (2 mL) (for **9**), filtered and washed with the same solvent (2 ml × 3), then dried (0.01 mmHg, 60 °C).

## Characterizations of Products

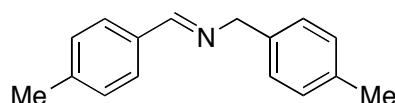
### *N*-Benzylidene-1-phenylmethanamine (**2a**)<sup>1</sup>



<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.44 (s, 1H), 7.85-7.82 (m, 2H), 7.49-7.28 (m, 8H), 4.88 (s, 2H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 162.2, 139.5, 136.3, 130.9, 128.8, 128.7, 128.5, 128.2, 127.2, 65.2.

### *N*-(4-Methylbenzylidene)-1-(*p*-tolyl)methanamine (**2b**)<sup>2</sup>



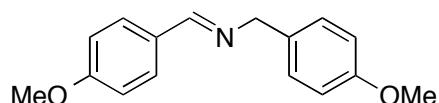
<sup>1</sup> Wendlandt, A. E.; Stahl S. S. *Org. Lett.* **2012**, *14*, 2850.

<sup>2</sup> Wu, X. F.; Petrosyan, A.; Ghochikyan, T. V.; Saghyan, A. S.; Langer, P. *Tetrahedron Lett.* **2013**, *54*, 3158.

<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.38 (s, 1H), 7.71 (d, *J* = 7.9 Hz, 2H), 7.28-7.24 (m, 4H), (d, *J* = 7.9 Hz, 2H), 4.81 (s, 2H), 2.42 (s, 3H), 2.38 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 161.8, 141.1, 136.7, 136.5, 133.8, 129.5, 129.3, 128.4, 128.1, 65.0, 21.7, 21.3.

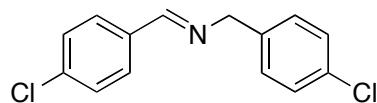
**N-(4-Methoxybenzylidene)-1-(4-methoxyphenyl)methanamine (2c)<sup>1</sup>**



<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.33 (s, 1H), 7.76-7.73 (m, 2H), 7.29-7.27 (m, 2H), 6.97-6.09 (m, 4H), 4.76 (s, 2H), 3.86 (s, 3H), 3.82 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 161.8, 161.1, 158.8, 131.8, 130.0, 129.3, 114.1, 114.1, 64.6, 55.5, 55.5.

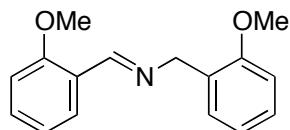
**N-(4-Chlorobenzylidene)-1-(4-chlorophenyl)methanamine (2d)<sup>1</sup>**



<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.36 (s, 1H), 7.76-7.72 (m, 2H), 7.44-7.40 (m, 2H), 7.36-7.28 (m, 4H), 4.79 (m 2H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 161.0, 137.8, 137.0, 134.6, 133.0, 131.1, 129.6, 129.4, 129.1, 128.8, 64.3.

**N-(2-Methoxybenzylidene)-1-(2-methoxyphenyl)methanamine (2e)<sup>3</sup>**

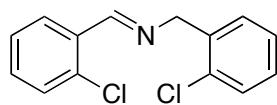


<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.88 (s, 1H), 8.07 (dd, *J* = 7.8, 1.7 Hz, 1H), 7.44-7.25 (m, 3H), 7.04-6.90 (m, 4H), 4.87 (s, 2H), 3.90 (s, 3H), 3.88 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 159.0, 158.5, 157.2, 131.9, 129.3, 128.3, 128.1, 127.7, 125.1, 120.9, 120.7, 111.2, 110.4, 59.8, 55.7, 55.5.

**N-(2-Chlorobenzylidene)-1-(2-chlorophenyl)methanamine (2f)<sup>3</sup>**

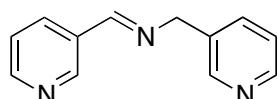
<sup>3</sup> Huang, B.; Tian, H.; Lin, S.; Xie, M.; Yu, X.; Xu, Q. *Tetrahedron Lett.* **2013**, *54*, 2861.



<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.91 (s, 1H), 8.16 (dd, *J* = 7.5, 1.8 Hz, 1H), 7.48- 7.22 (m, 7H), 4.98 (s, 2H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 159.9, 137.0, 135.5, 133.6, 133.3, 131.9, 130.0, 129.9, 129.5, 128.7, 128.5, 127.2, 127.1, 62.4.

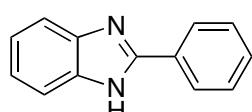
### 1-(Pyridin-3-yl)-*N*-(pyridin-3-ylmethylene)methanamine (2g)<sup>2</sup>



<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.92-8.91 (m, 1H), 8.70-8.54 (m, 3H), 8.49 (s, 1H), 8.19-8.15 (m, 1H), 7.72-7.66 (m, 1H), 7.40-7.27 (m, 2H), 4.86 (s, 2H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 159.9, 152.0, 150.5, 149.5, 148.8, 135.7, 134.7, 134.5, 131.5, 123.8, 123.6, 62.6.

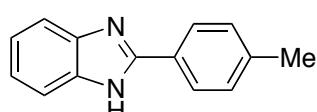
### 2-Phenylbenzimidazole (4aa)<sup>4</sup>



<sup>1</sup>H NMR (500 MHz, CD<sub>3</sub>OD) δ 8.09-8.07 (m, 2H), 7.62-7.59 (m, 2H), 7.53-7.46 (m, 3H), 7.27-7.23 (m, 2H).

<sup>13</sup>C NMR (125 MHz, CD<sub>3</sub>OD) δ 153.5, 140.4, 131.4, 131.1, 130.2, 127.9, 124.0, 116.0.

### 2-(*p*-Tolyl)benzimidazole (4ba)<sup>4</sup>

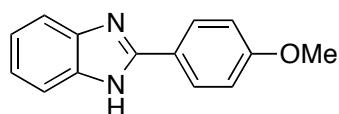


<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 7.96 (d, *J* = 8.1 Hz, 2H), 7.60-7.56 (m, 2H), 7.33 (d, *J* = 8.1 Hz, 2H), 7.26-7.21 (m, 2H), 2.40 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 153.8, 142.0, 130.9, 128.5, 127.9, 123.9, 118.6, 113.1, 21.6.

### 2-(4-Methoxyphenyl)benzimidazole (4ca)<sup>4</sup>

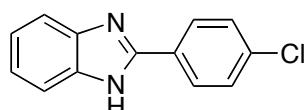
<sup>4</sup> Nguyen, T. B., Ermolenko, L.; Dean, W. A. Al-Mourabit, A. *Org. Lett.* **2012**, *14*, 5948.



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.02-7.99 (m, 2H), 7.57-7.54 (m, 2H), 7.22-7.20 (m, 2H), 7.08-7.03 (m, 2H), 3.84 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 162.8, 153.4, 129.2, 123.5, 123.3, 115.3, 55.7.

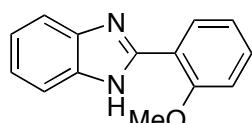
#### 2-(4-Chlorophenyl)benzimidazole (4da)<sup>4</sup>



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.07-8.02 (m, 2H), 7.62-7.57 (m, 2H), 7.55-7.51 (m, 2H), 7.29-7.23 (m, 2H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 152.2, 137.3, 130.3, 129.7, 129.3, 124.1, 115.3.

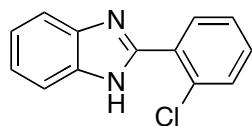
#### 2-(2-Methoxyphenyl)benzimidazole (4ea)



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.23 (dd, *J* = 7.8, 1.7 Hz, 1H), 7.64-7.61 (m, 2H), 7.47-7.41 (m, 1H), 7.25-7.22 (m, 2H), 7.15 (dd, *J* 8.5, 0.9 Hz, 1H), 7.12-7.07 (m 1H), 4.02 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 158.7, 151.1, 139.4, 132.8, 131.0, 123.7, 122.2, 118.9, 115.9, 112.8, 56.2.

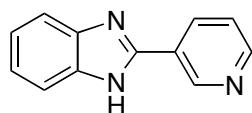
#### 2-(2-Chlorophenyl)benzimidazole (4fa)



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 7.83-7.79 (m, 1H), 7.67-7.41 (m, 5H), 7.31-7.25 (m, 2H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 151.2, 139.9, 134.0, 133.2, 132.6, 131.6, 131.1, 128.5, 124.2, 116.1.

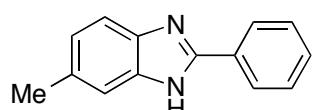
#### 2-(Pyridin-3-yl)benzimidazole (4ga)<sup>4</sup>



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 9.23 (m, 1H), 8.63 (dd, *J* = 4.9, 1.7 Hz, 1H), 8.47-8.43 (m, 1H), 7.62-7.55 (m, 3H), 7.30-7.24 (m, 2H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 151.4, 150.3, 148.5, 136.1, 128.0, 125.7, 125.6, 124.6, 116.1 (broad peak).

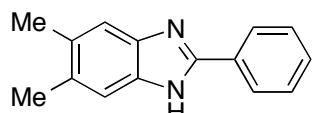
#### 6-Methyl-2-phenylbenzimidazole (4ab)<sup>4</sup>



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.06-8.03 (m, 2H), 7.52-7.37 (m, 5H), 7.08-7.05 (m, 1H), 2.44 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 153.2, 134.1, 131.3, 130.2, 127.8, 121.3, 118.6, 118.3, 115.6, 21.9.

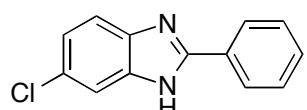
#### 5,6-Dimethyl-2-phenylbenzimidazole (4ac)<sup>4</sup>



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.05-8.03 (m, 2H), 7.53-7.43 (m, 3H), 7.35 (s, 2H), 2.35 (s, 5H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 152.4, 138.7, 133.0, 131.2, 131.0, 130.1, 127.6, 115.9, 20.5.

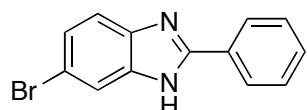
#### 6-Chloro-2-phenylbenzimidazole (4ad)<sup>4</sup>



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.05-8.03 (m, 2H), 7.56-7.47 (m, 5H), 7.22-7.20 (m, 1H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 154.8, 131.8, 130.7, 130.3, 129.5, 128.0, 124.4, 118.9, 113.9.

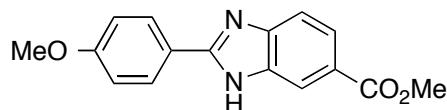
#### 6-Bromo-2-phenylbenzimidazole (4ae)



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.05-8.03 (m, 2H), 7.72 (m, 2H), 7.53-7.47 (m, 4H), 7.32 (dd, *J* = 8.6, 1.6 Hz, 1H).

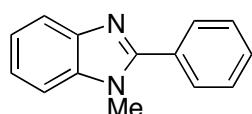
<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 154.7, 131.8, 130.7, 130.3, 128.0, 127.1, 119.2, 116.8.

**Methyl 2-(4-methoxyphenyl)benzimidazole-6-carboxylate (4cf)**



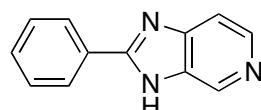
<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD) δ 8.23 (d, *J* = 1.6 Hz, 1H), 8.03-8.00 (m, 2H), 7.91 (dd, *J* = 8.7, 1.6 Hz, 1H), 7.58 (d, *J* = 8.7 Hz), 7.09-7.06 (m, 2H), 3.92 (s, 3H), 3.87 (s, 3H).  
<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD) δ 182.1, 169.2, 163.5, 129.8, 125.6, 125.3, 122.8, 115.7, 56.1, 52.7.

**1-Methyl-2-phenylbenzimidazole (4cg)<sup>4</sup>**



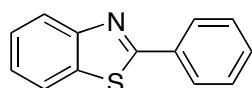
<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 7.88-7.85 (m, 1H), 7.80-7.77 (m, 2H), 7.58-7.51 (m, 3H), 7.42-7.32 (m, 3H), 3.86 (s, 3H).  
<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 153.9, 143.1, 136.7, 130.4, 129.9, 129.6, 128.8, 122.9, 122.6, 120.0, 109.8, 31.8.

**2-Phenyl-3*H*-imidazo[4,5-*c*]pyridine (4ah)<sup>5</sup>**



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 13.36 (broad s, 1H), 8.95 (s, 1H), 8.32 (d, *J* = 5.4 Hz, 1H), 8.24-8.21 (m, 2H), 7.62-7.52 (m, 4H).  
<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 158.0, 145.8, 142.6, 140.7, 139.9, 133.2, 132.3, 131.2, 129.3, 111.5.

**2-Phenylbenzothiazole (6a)<sup>4</sup>**

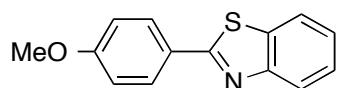


<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.09-8.06 (m, 3H), 7.09 (dd, *J* = 8.3, 0.9 Hz, 1H), 7.49-7.47 (m, 4H), 7.39-7.36 (m, 1H).

<sup>5</sup> Sharghi, H.; Asemani, O.; Khalifeh, R. *Synth. Commun.* **2008**, 38, 1128.

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 168.3, 154.3, 135.3, 133.8, 131.2, 129.2, 127.8, 126.5, 125.4, 123.5, 121.8.

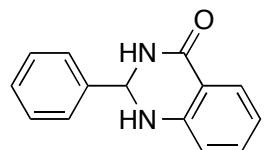
**2-(4-Methoxyphenyl)benzothiazole (6c)<sup>6</sup>**



<sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>) δ 8.03-8.00 (m, 3H), 7.85 (d, *J* = 8.1 Hz), 7.48-7.42 (m, 1H), 7.36-7.31 (m, 1H), 7.00-6.96 (m, 2H), 3.85 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CDCl<sub>3</sub>) δ 168.0, 162.1, 154.4, 135.0, 129.3, 126.6, 126.4, 125.0, 123.0, 121.7, 114.5, 55.6.

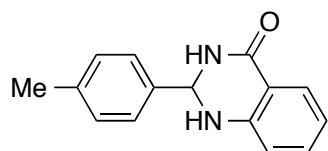
**2-Phenyl-2,3-dihydroquinazolin-4(1*H*)-one (8a)<sup>7</sup>**



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 8.29 (broad s, 1H), 7.62 (dd, *J* = 7.6, 1.6 Hz, 1H), 7.52-7.48 (m, 2H), 7.43-7.32 (m, 3H), 7.27-7.21 (m, 1H), 7.10 (broad s, 1H), 6.76 (d, *J* = 8.4 Hz, 1H), 6.67 (1H, t, *J* = 7.4 Hz, 1H), 5.76 (broad s, 1H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 163.6, 147.8, 141.6, 133.3, 128.4, 128.3, 127.3, 126.8, 117.1, 114.9, 114.4, 66.5.

**2-(*p*-Tolyl)-2,3-dihydroquinazolin-4(1*H*)-one (8b)<sup>7</sup>**



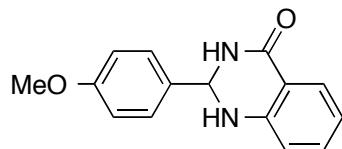
<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 8.23 (1H, br s), 7.62 (dd, *J* = 8.0, 1.6 Hz, 1H), 7.39-7.36 (m, 2H), 7.27-7.18 (m, 3H), 7.05 (broad s, 1H), 6.74 (d, *J* = 8.0 Hz, 1H), 6.70-6.65 (m, 1H), 5.72 (broad s, 1H), 2.30 (3H, s).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 163.6, 147.9, 138.6, 137.7, 133.2, 128.8, 127.3, 126.8, 117.0, 115.0, 114.4, 66.3, 20.7.

<sup>6</sup> Liu, J.; Gui, Q.; Yang, Z.; Tan, Z.; Guo, R.; Shi, J. *Synthesis* **2013**, 943.

<sup>7</sup> J. A. Watson, A. J. A.; Maxwell, A. C.; Williams, J. M. *J. Org. Biomol. Chem.* **2012**, 10, 240.

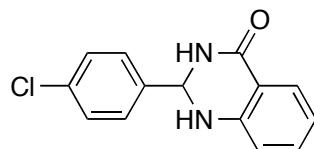
**2-(4-Methoxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (8c)<sup>7</sup>**



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 8.19 (m, 1H), 7.62 (dd, *J* = 7.6, 1.6 Hz, 1H), 7.44-7.41 (m, 2H), 7.27-7.21 (m, 1H), 7.02 (broad s, 1H), 6.96-6.93 (m, 2H), 6.75 (d, *J* = 7.6 Hz, 1H), 6.70-6.65 (m, 1H), 5.71 (broad s, 1H), 3.75 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 163.7, 159.4, 148.0, 133.4, 133.2, 128.2, 127.3, 117.0, 115.0, 114.4, 113.6, 66.3, 55.1.

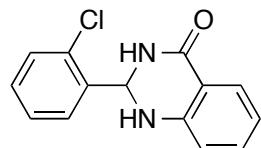
**2-(4-Chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (8d)<sup>7</sup>**



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD/CD<sub>3</sub>SOCD<sub>3</sub>) δ 7.63-7.61 (m, 1H), 7.74-7.45 (m, 2H), 7.37-7.34 (m, 2H), 7.24-7.19 (m, 1H), 6.70-6.65 (m, 2H), 5.74 (s, 1H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD/CD<sub>3</sub>SOCD<sub>3</sub>) δ 165.8, 149.0, 140.8, 135.0, 134.7, 130.3, 129.7, 129.4, 128.4, 118.7, 115.7, 115.4, 67.6.

**2-(2-Chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (8e)<sup>8</sup>**

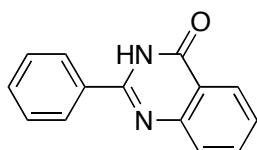


<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>OD/CD<sub>3</sub>SOCD<sub>3</sub>) δ 7.67 (m, 2H), 7.42-7.20 (m, 4H), 6.73-6.67 (m, 2H), 6.17 (s, 1H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>OD/CD<sub>3</sub>SOCD<sub>3</sub>) δ 166.3, 149.4, 139.2, 135.3, 133.8, 131.8, 131.2, 130.1, 129.0, 128.8, 119.4, 116.1, 65.4.

**2-Phenylquinazolin-4(3*H*)-one (9a)<sup>7</sup>**

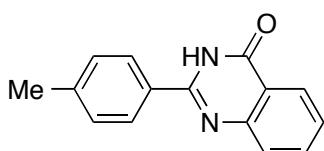
<sup>8</sup> Gao, L.; Ji, H.; Rong, L.; Tang, D.; Zha, Y.; Shi, Y.; Tu, S. J. Heterocycl. Chem. **2011**, *48*, 957



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 12.55 (s, 1H), 8.21-8.15 (m, 3H), 7.86-7.73 (m, 2H), 7.61-7.49 (m, 4H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 162.2, 152.3, 148.7, 134.5, 132.7, 131.3, 128.5, 127.7, 127.5, 126.5, 125.8, 121.0.

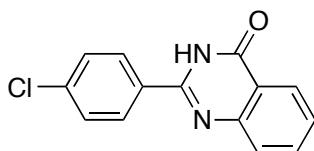
**2-(*p*-Tolyl)quinazolin-4(3*H*)-one (9b)<sup>7</sup>**



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 12.44 (s, 1H), 8.16-8.07 (m, 3H), 7.84-7.79 (m, 1H), 7.73 (d, *J* = 7.9 Hz, 1H), 7.52-7.47 (m, 1H), 7.34 (d, *J* = 8.1 Hz, 2H), 2.38 (s, 3H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 162.2, 152.2, 148.8, 141.4, 134.5, 129.8, 129.1, 127.6, 127.3, 126.3, 125.8, 120.8, 118.2, 21.0.

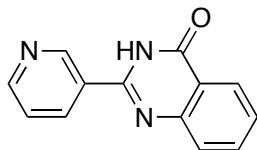
**2-(4-Chlorophenyl)quinazolin-4(3*H*)-one (9d)<sup>9</sup>**



<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 12.57 (s, 1H), 8.22-8.14 (m, 3H), 7.87-7.82 (m, 1H), 7.74 (d, *J* = 7.9 Hz, 1H), 7.63 (d, *J* = 8.6 Hz, 2H), 7.56-7.51 (m, 1H).

<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 218.6, 162.1, 151.3, 148.5, 136.2, 134.6, 131.5, 129.6, 128.7, 127.5, 126.8, 125.8, 120.9, 39.5.

**2-(Pyridin-3-yl)quinazolin-4(3*H*)-one (9g)<sup>7</sup>**



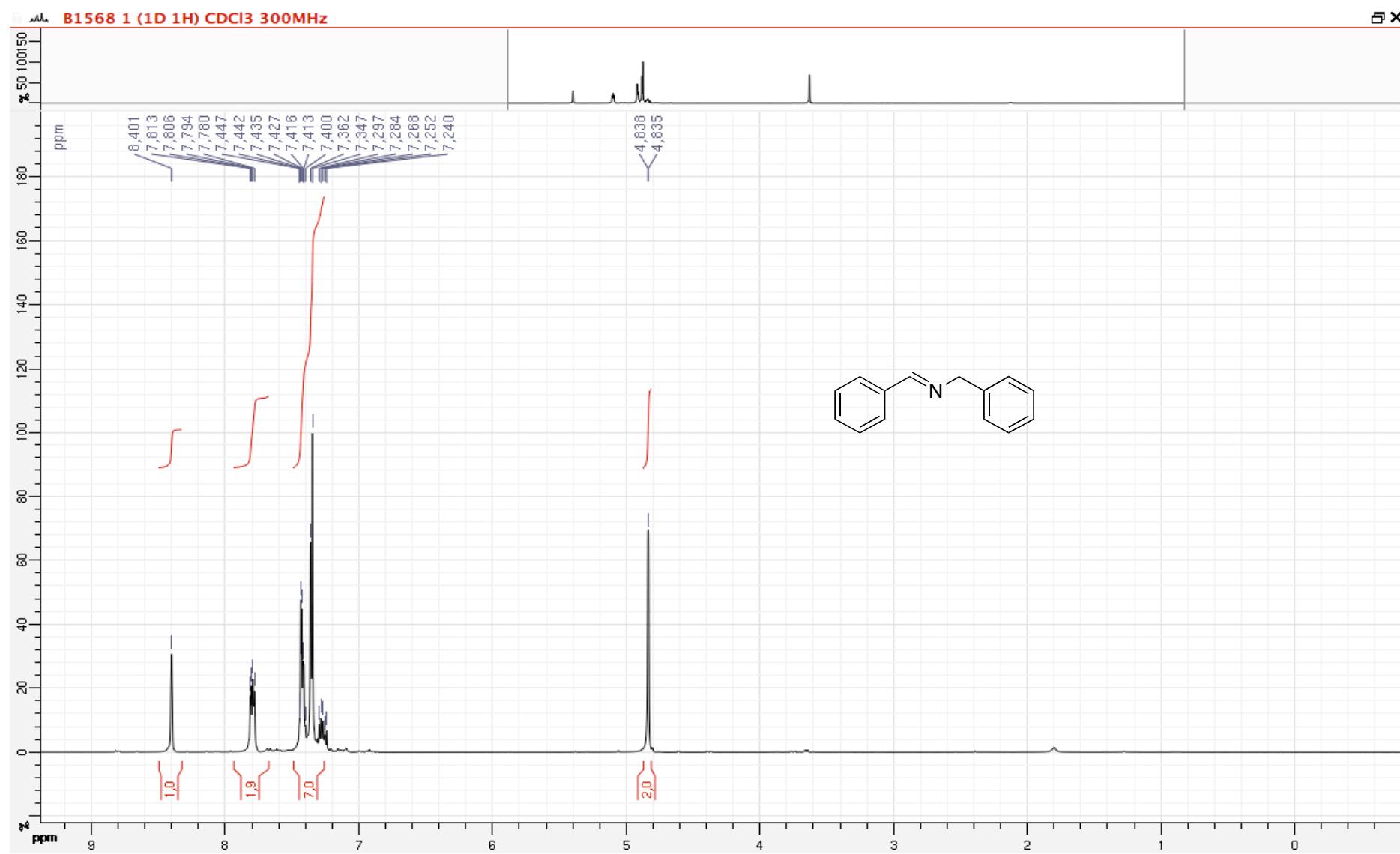
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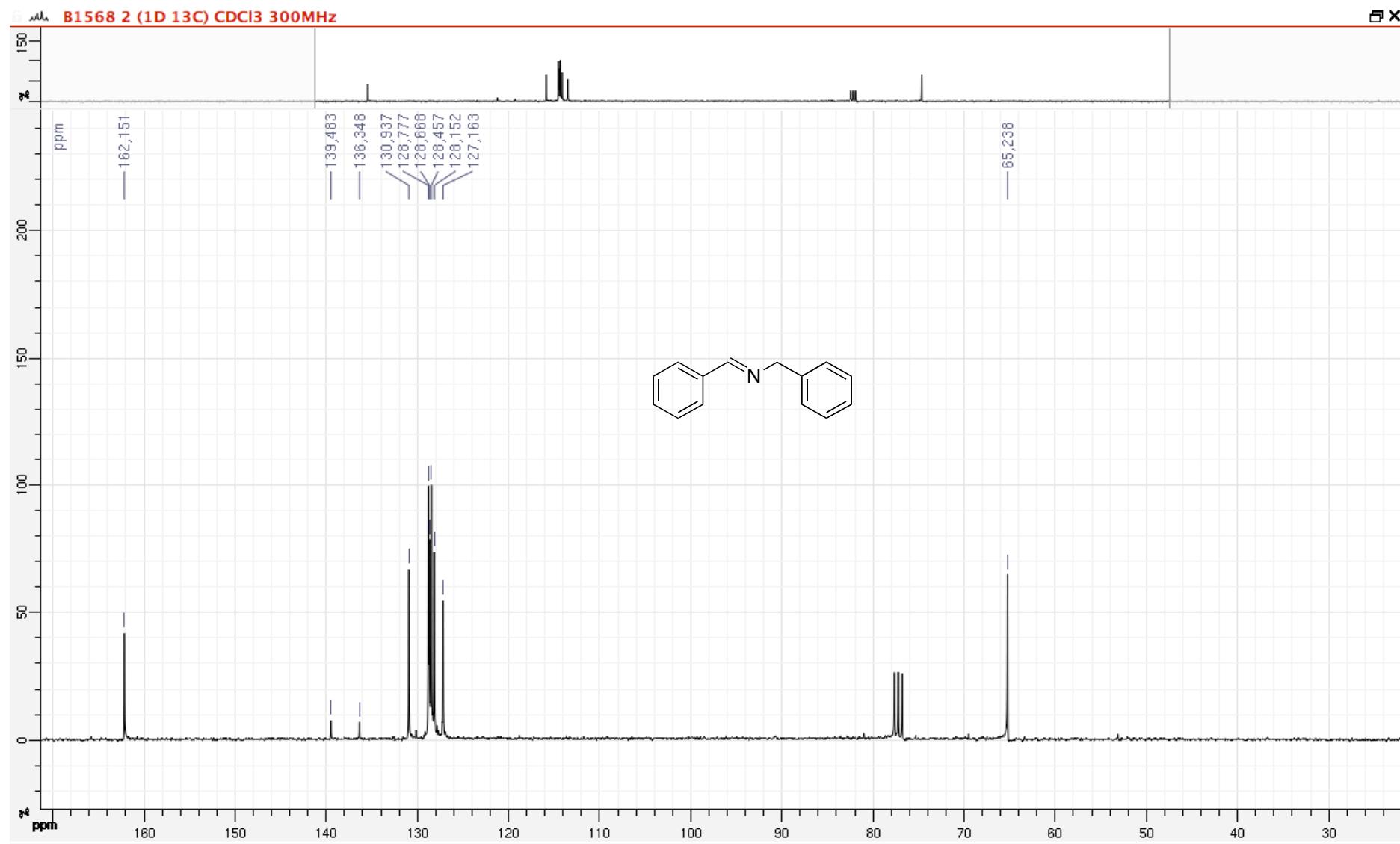
<sup>9</sup> Zeng, L.; Cai, C. *J. Heterocycl. Chem.* **2010**, *47*, 1035.

<sup>1</sup>H NMR (300 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 12.74 (s, 1H), 8.76 (dd, *J* = 4.9, 1.5 Hz, 1H), 8.50 (m, 1H), 8.17 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.85-7.83 (m, *J* = 1H), 7.77 (d, *J* = 7.7 Hz, 1H), 7.61-7.52 (m, 2H).

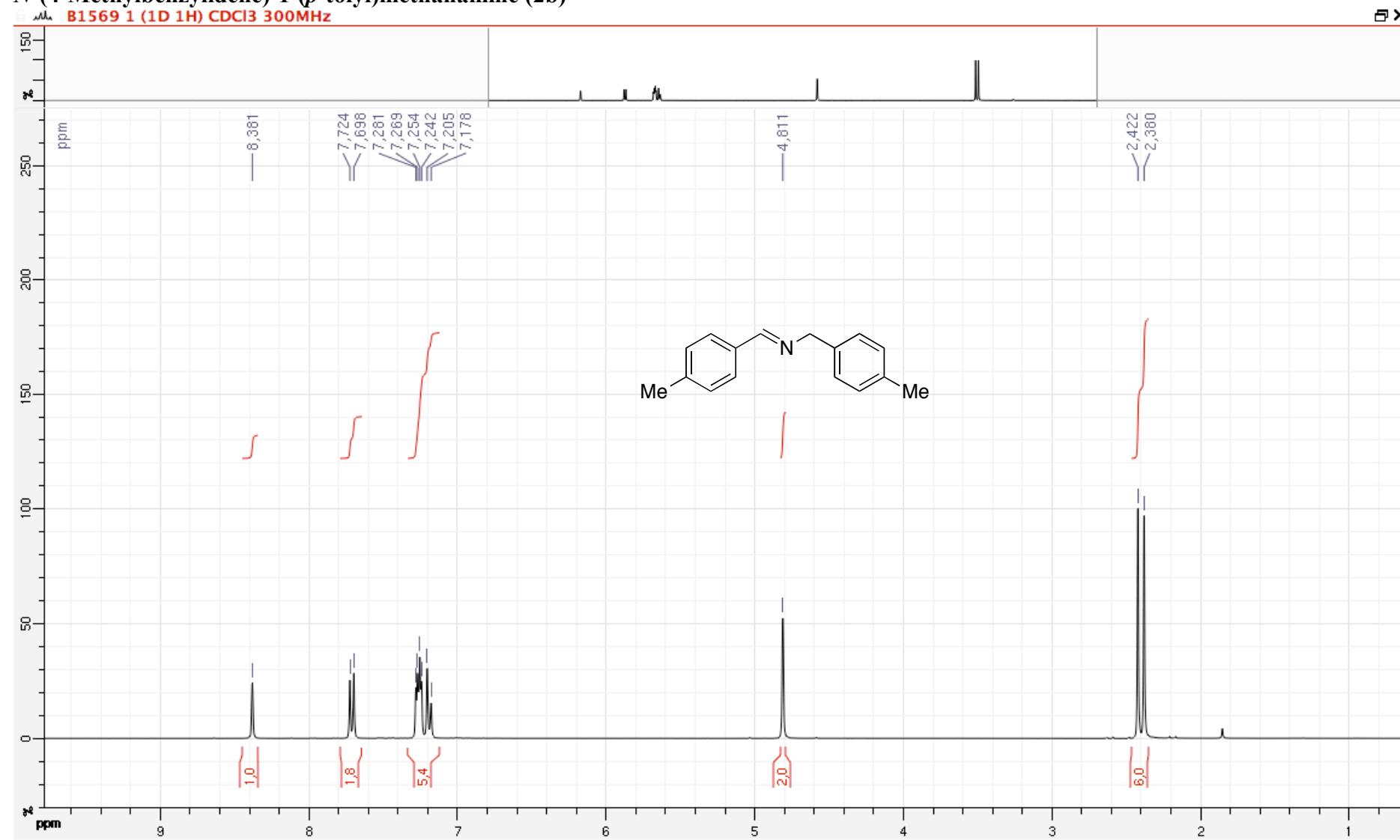
<sup>13</sup>C NMR (75 MHz, CD<sub>3</sub>SOCD<sub>3</sub>) δ 162.1, 151.8, 150.8, 148.7, 148.5, 135.4, 134.7, 128.7, 127.5, 126.9, 125.9, 123.5, 121.1.

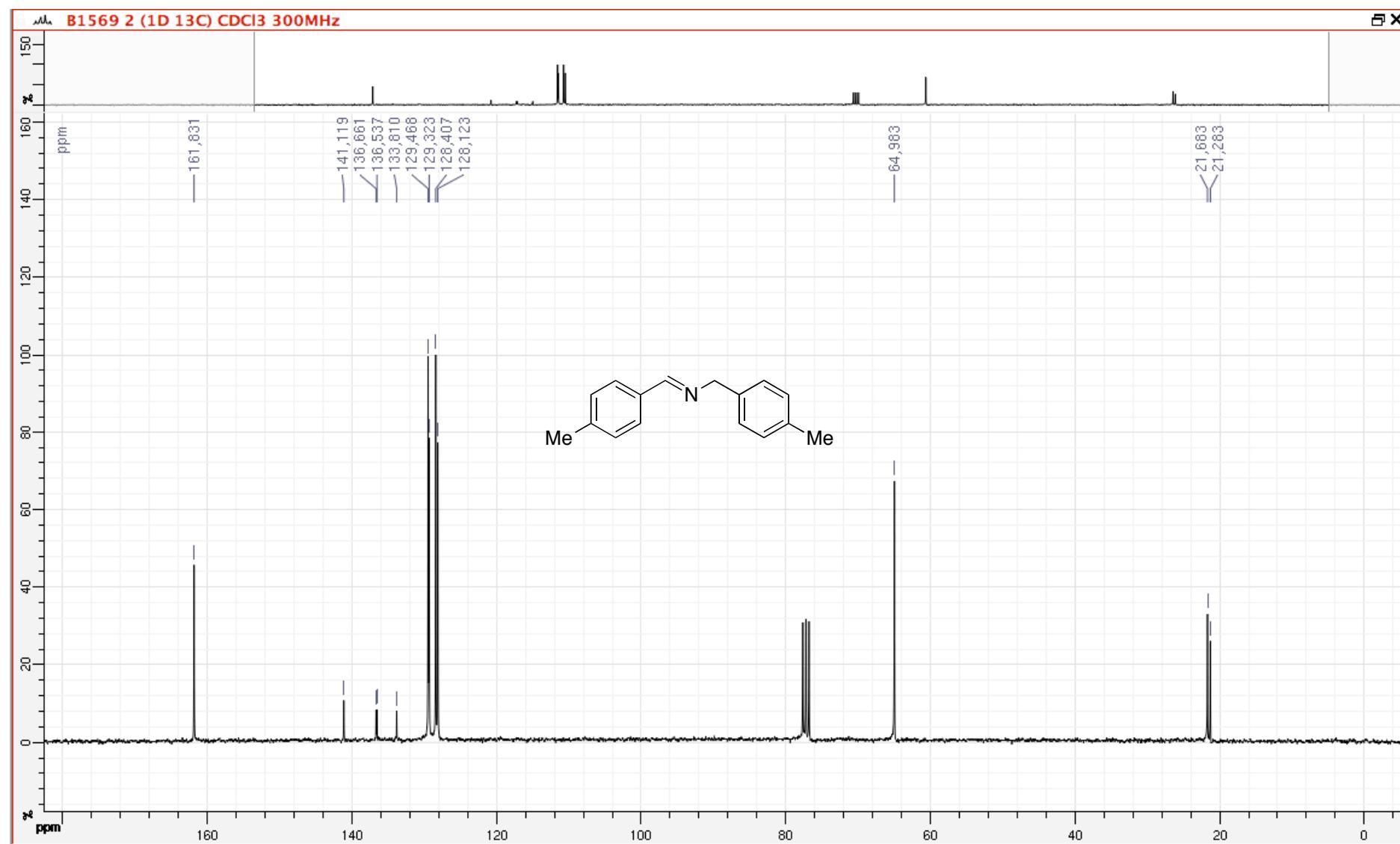
**N-Benzylidene-1-phenylmethanamine (2a)**



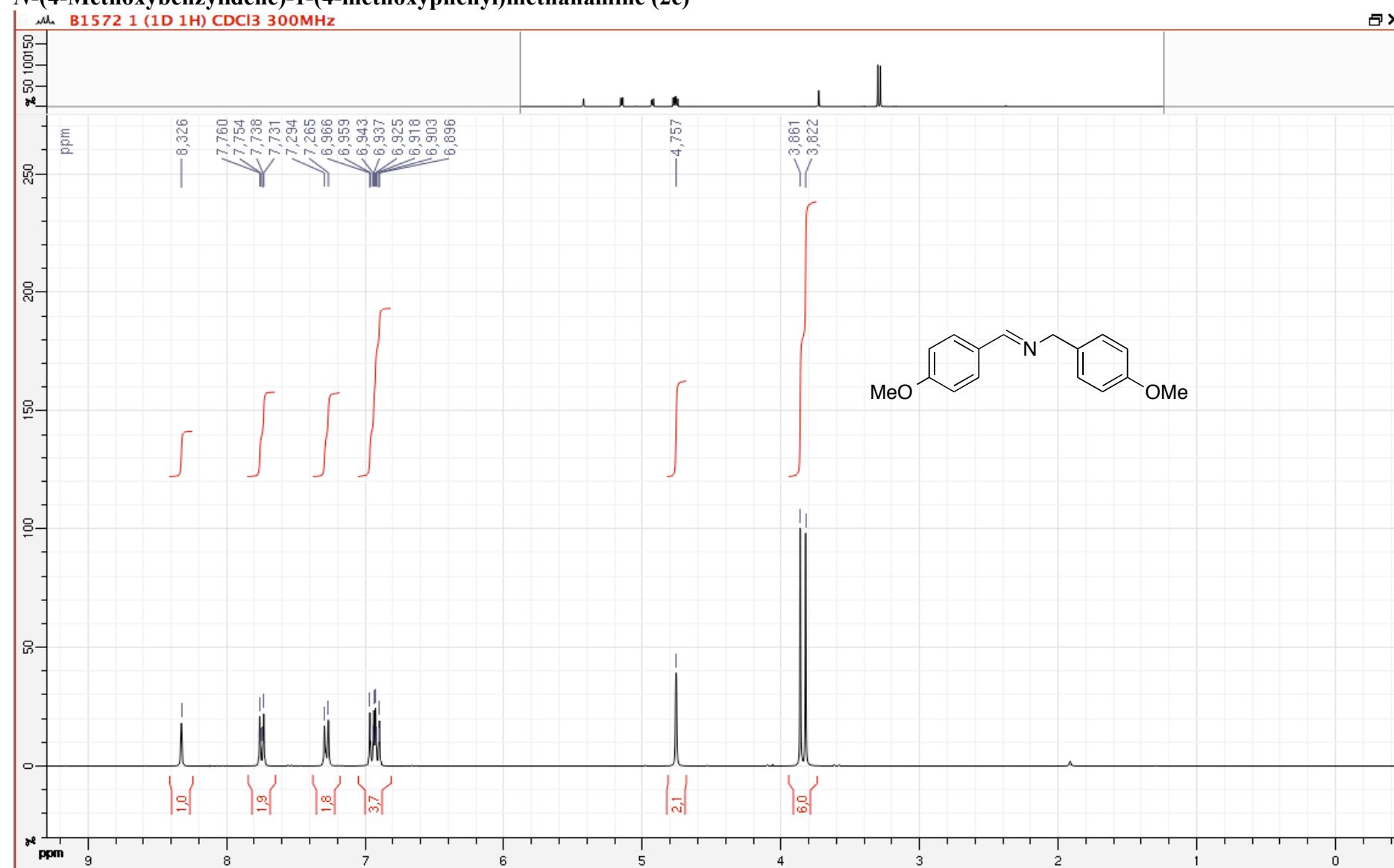


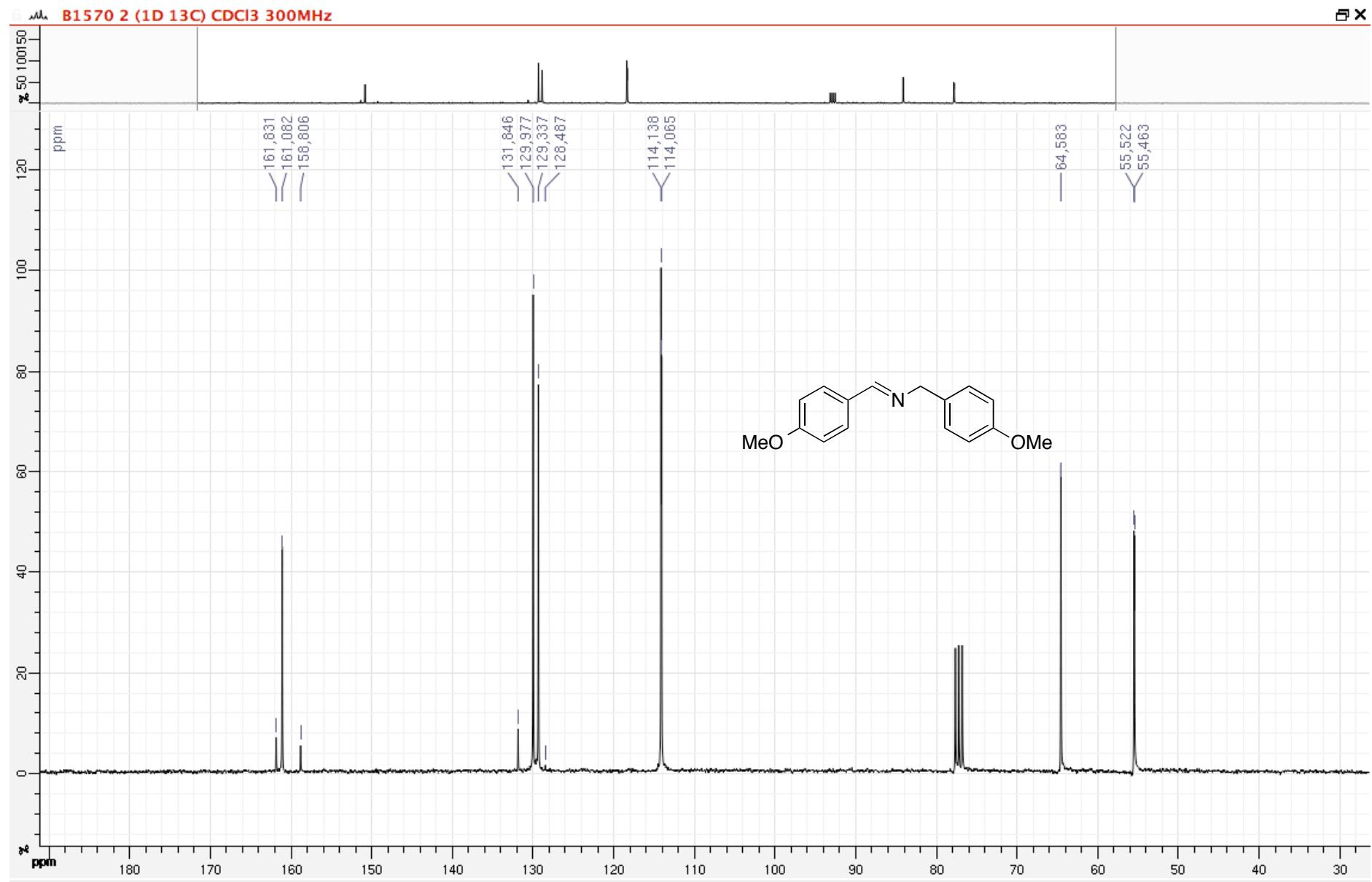
**N-(4-Methylbenzylidene)-1-(*p*-tolyl)methanamine (2b)**



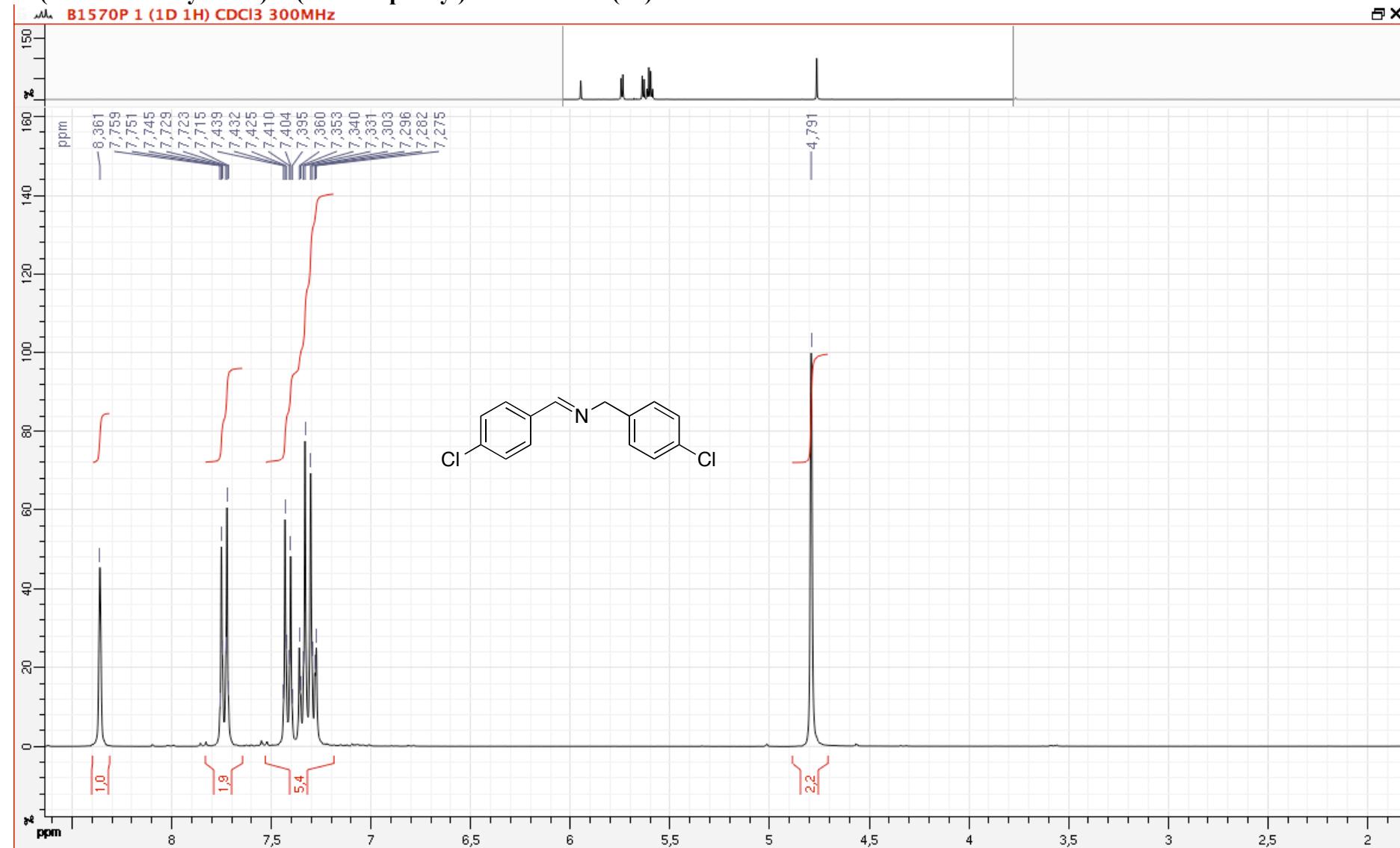


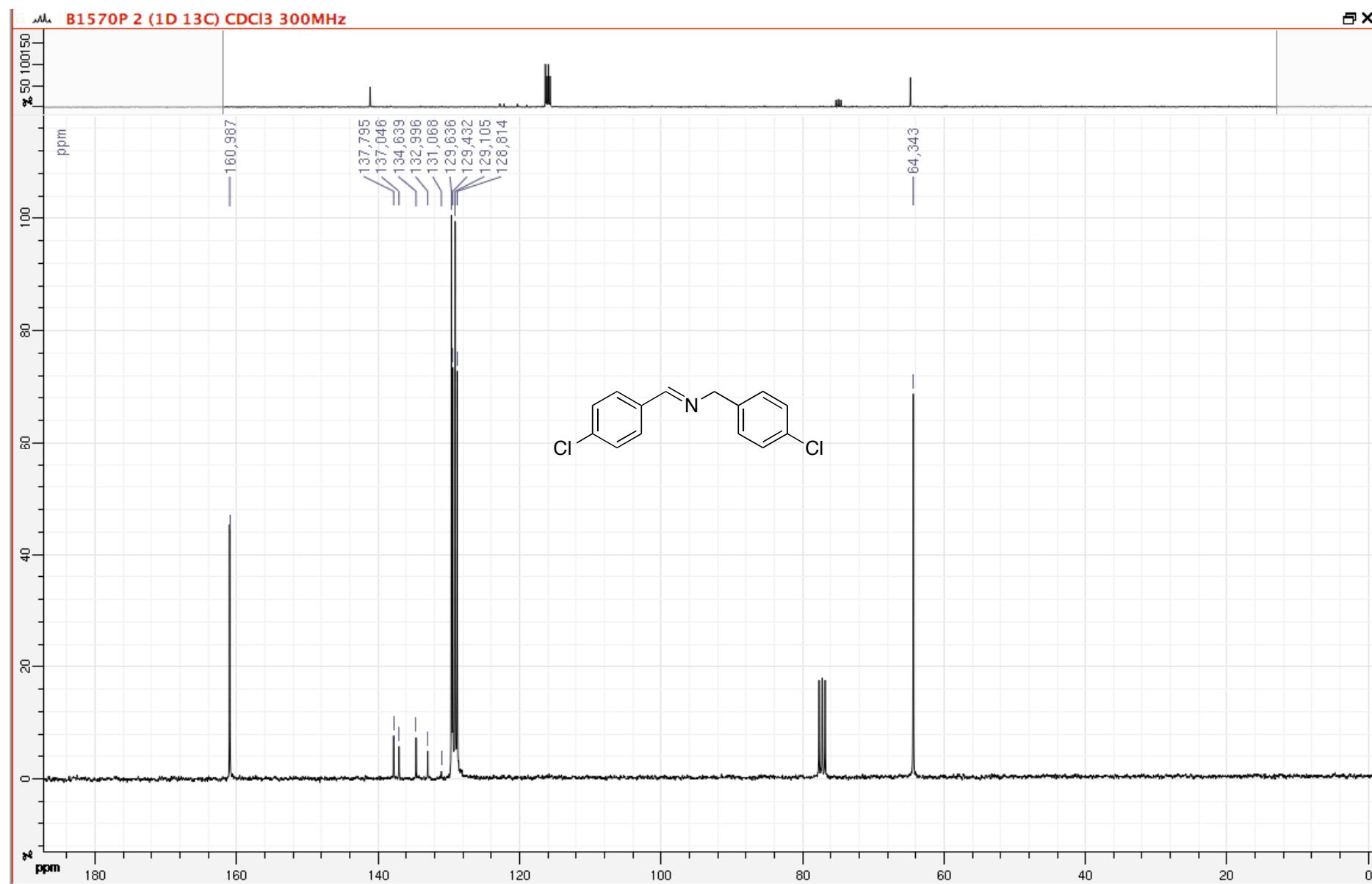
**N-(4-Methoxybenzylidene)-1-(4-methoxyphenyl)methanamine (2c)**



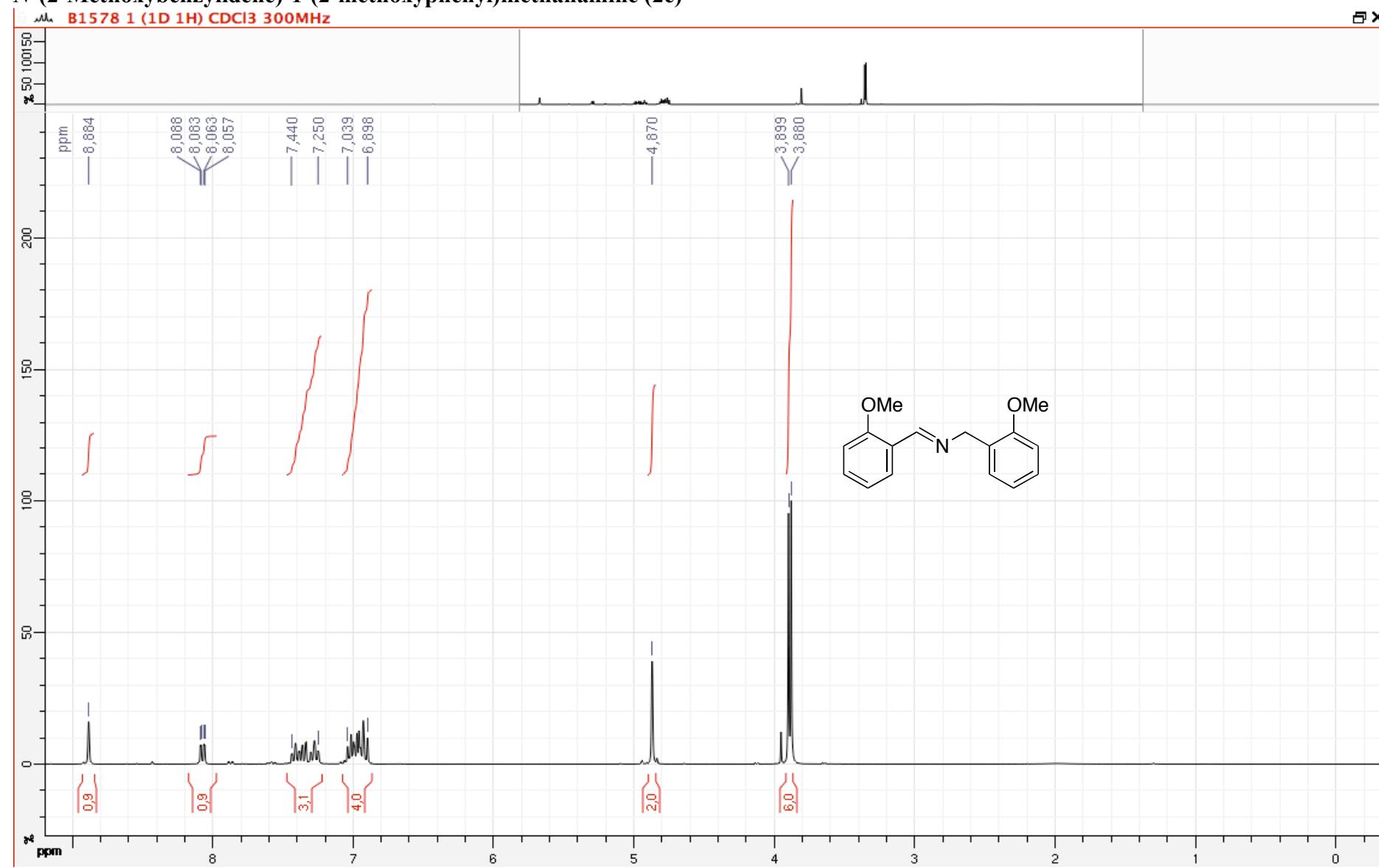


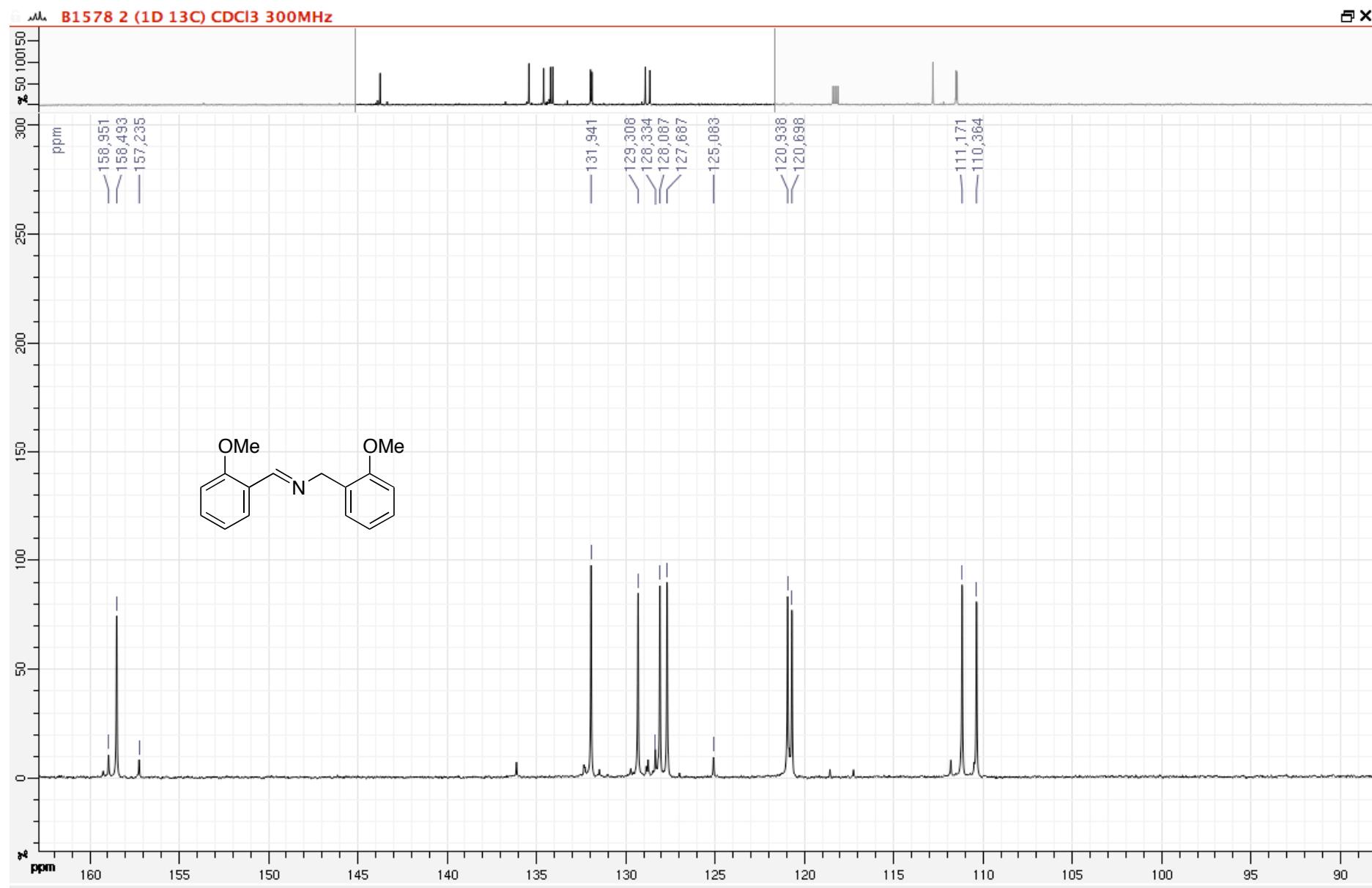
**N-(4-Chlorobenzylidene)-1-(4-chlorophenyl)methanamine (2d)**



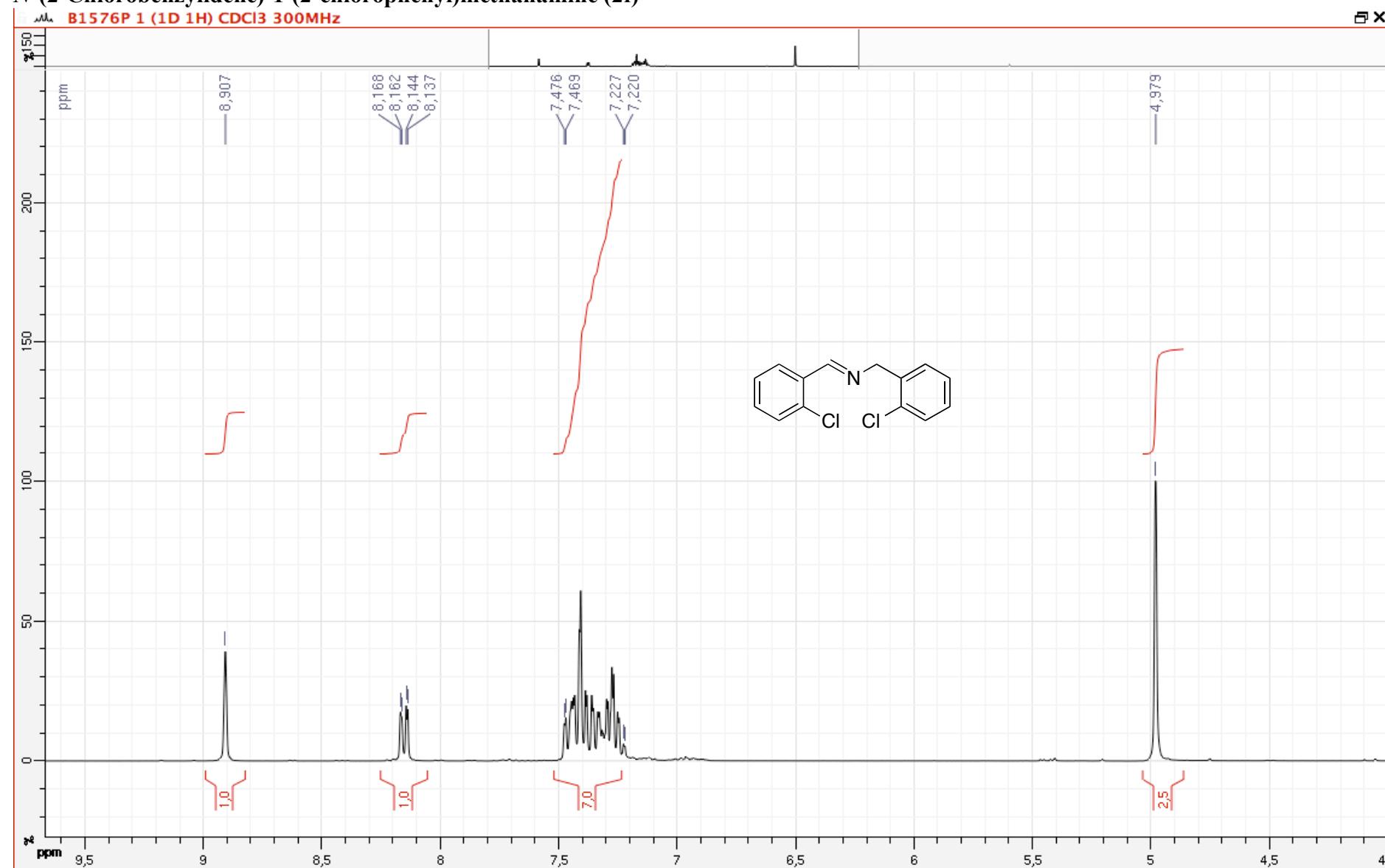


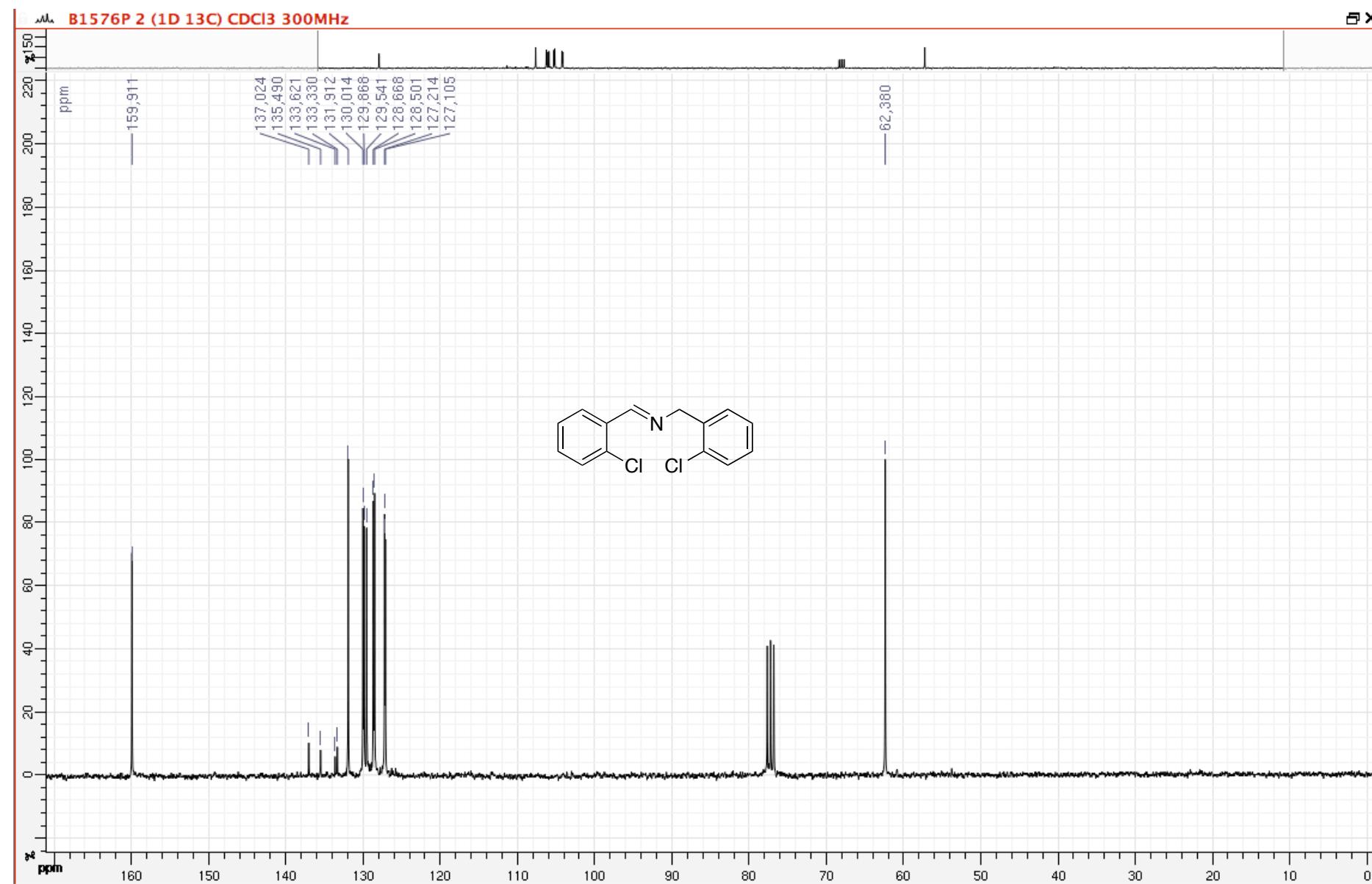
**N-(2-Methoxybenzylidene)-1-(2-methoxyphenyl)methanamine (2e)**



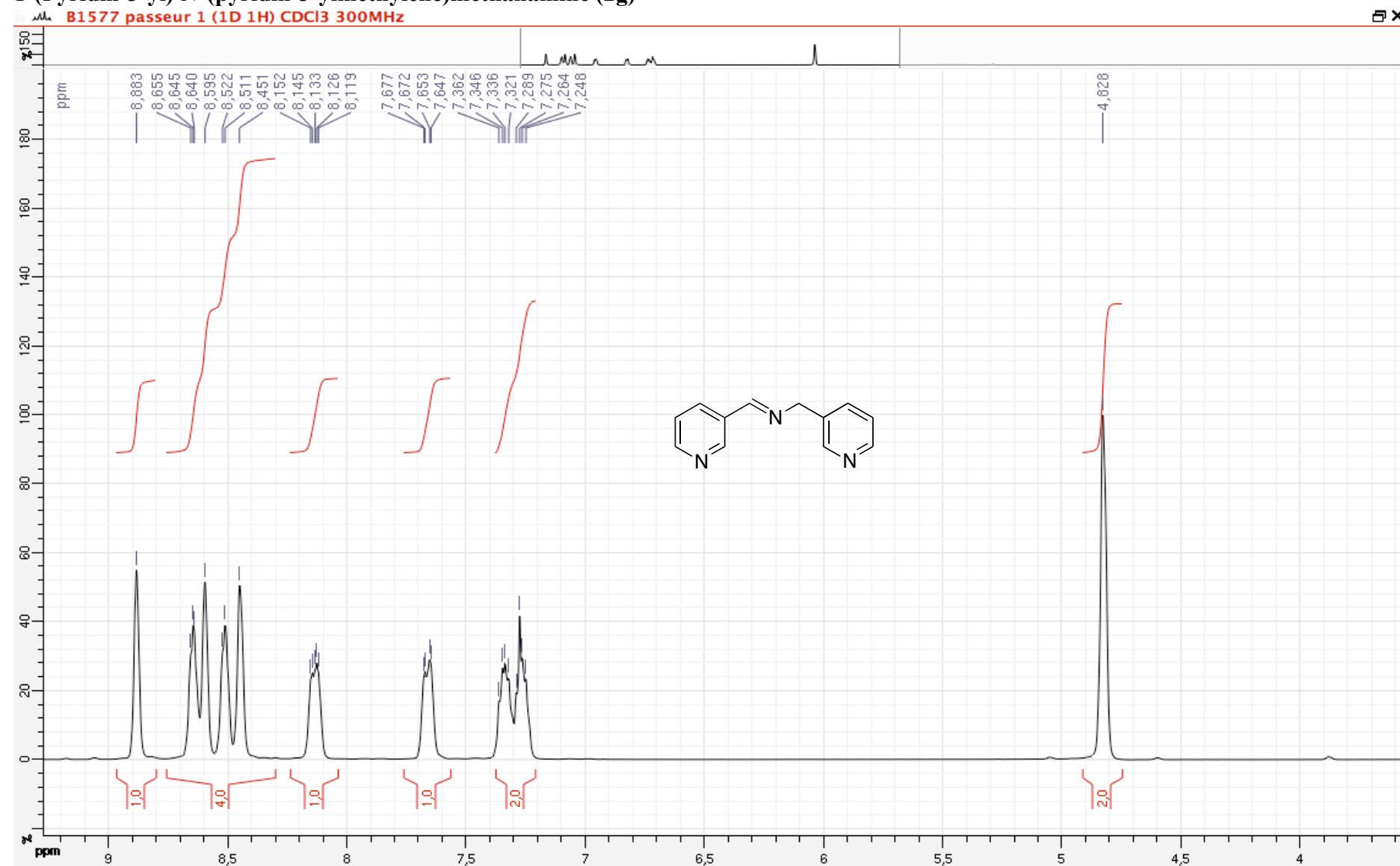


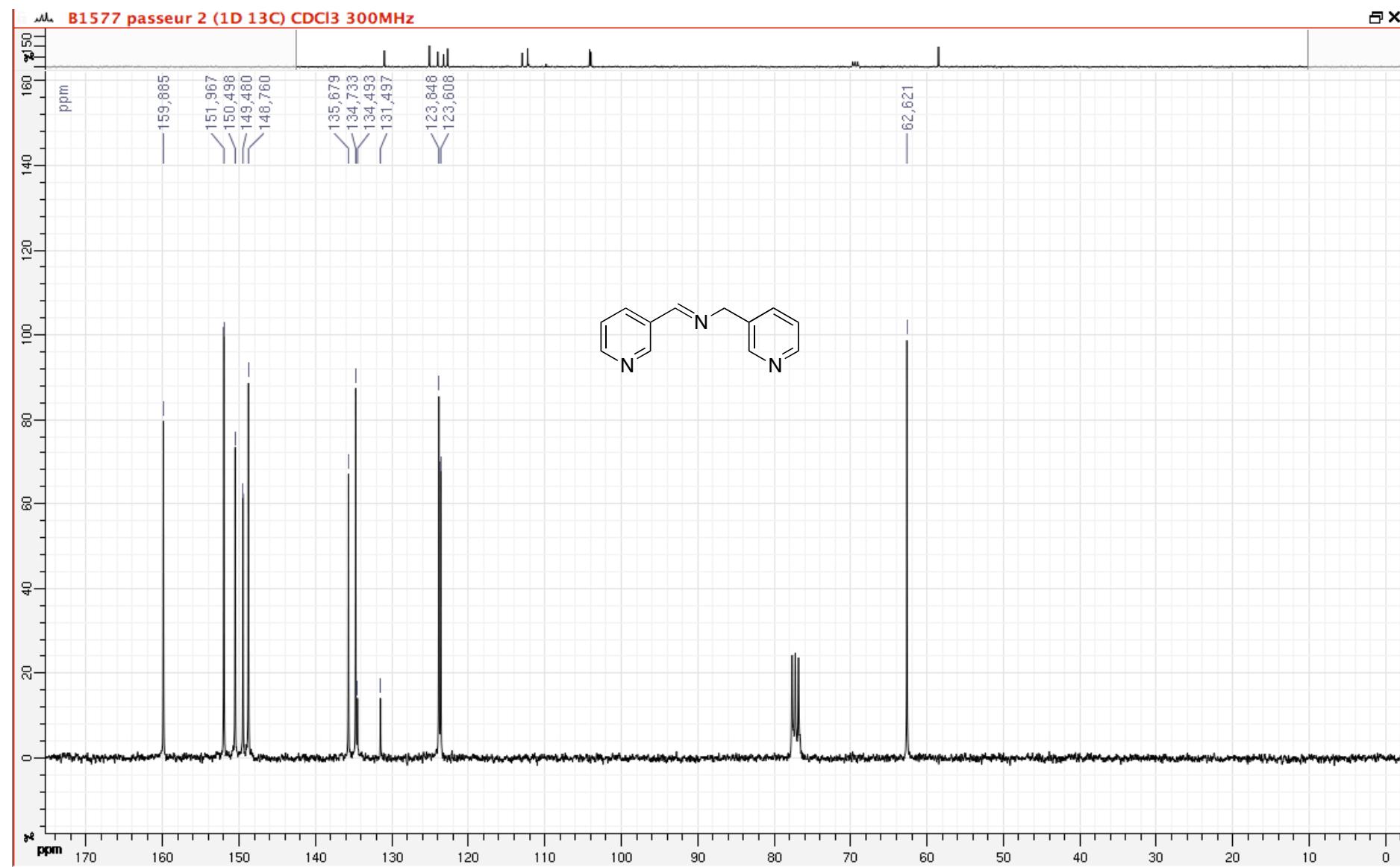
**N-(2-Chlorobenzylidene)-1-(2-chlorophenyl)methanamine (2f)**



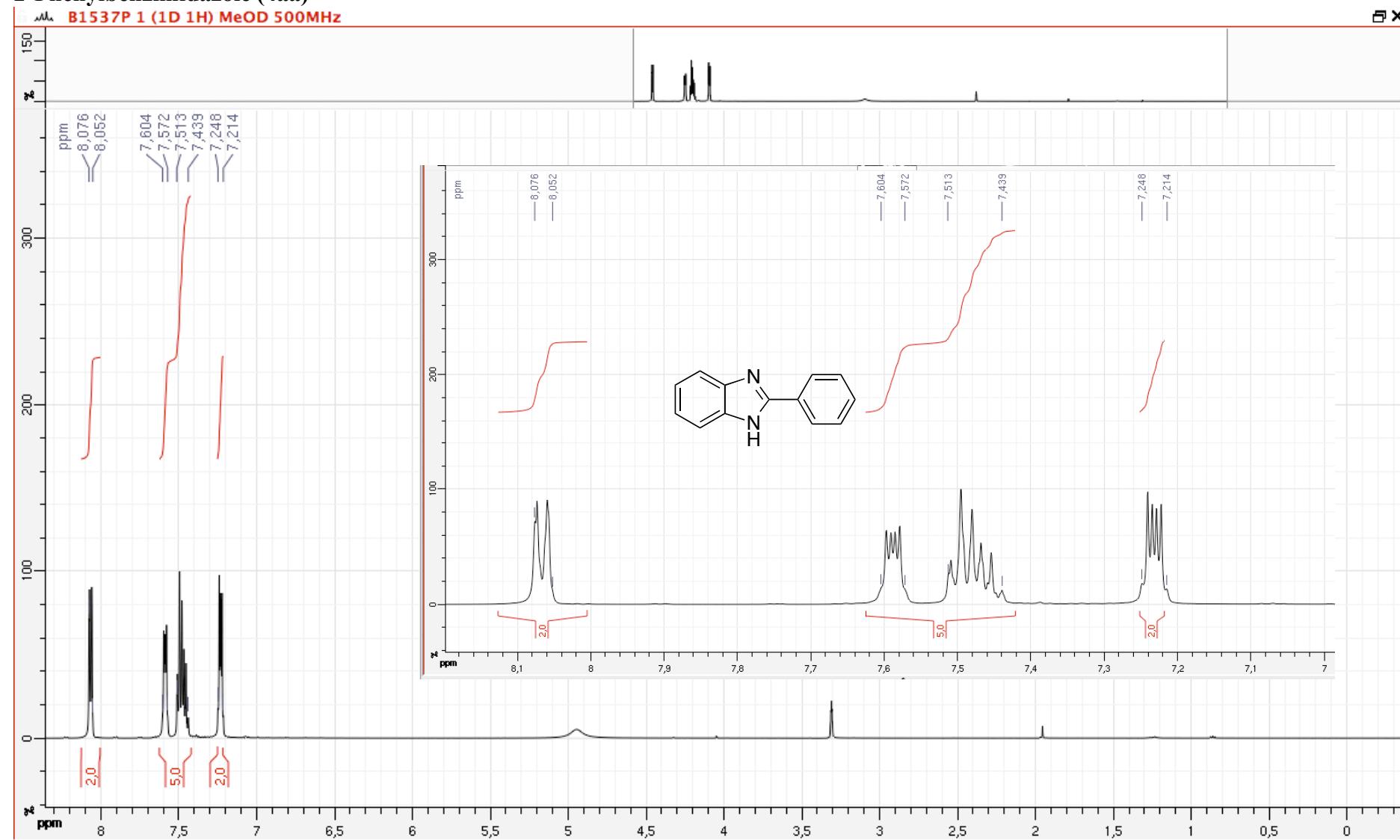


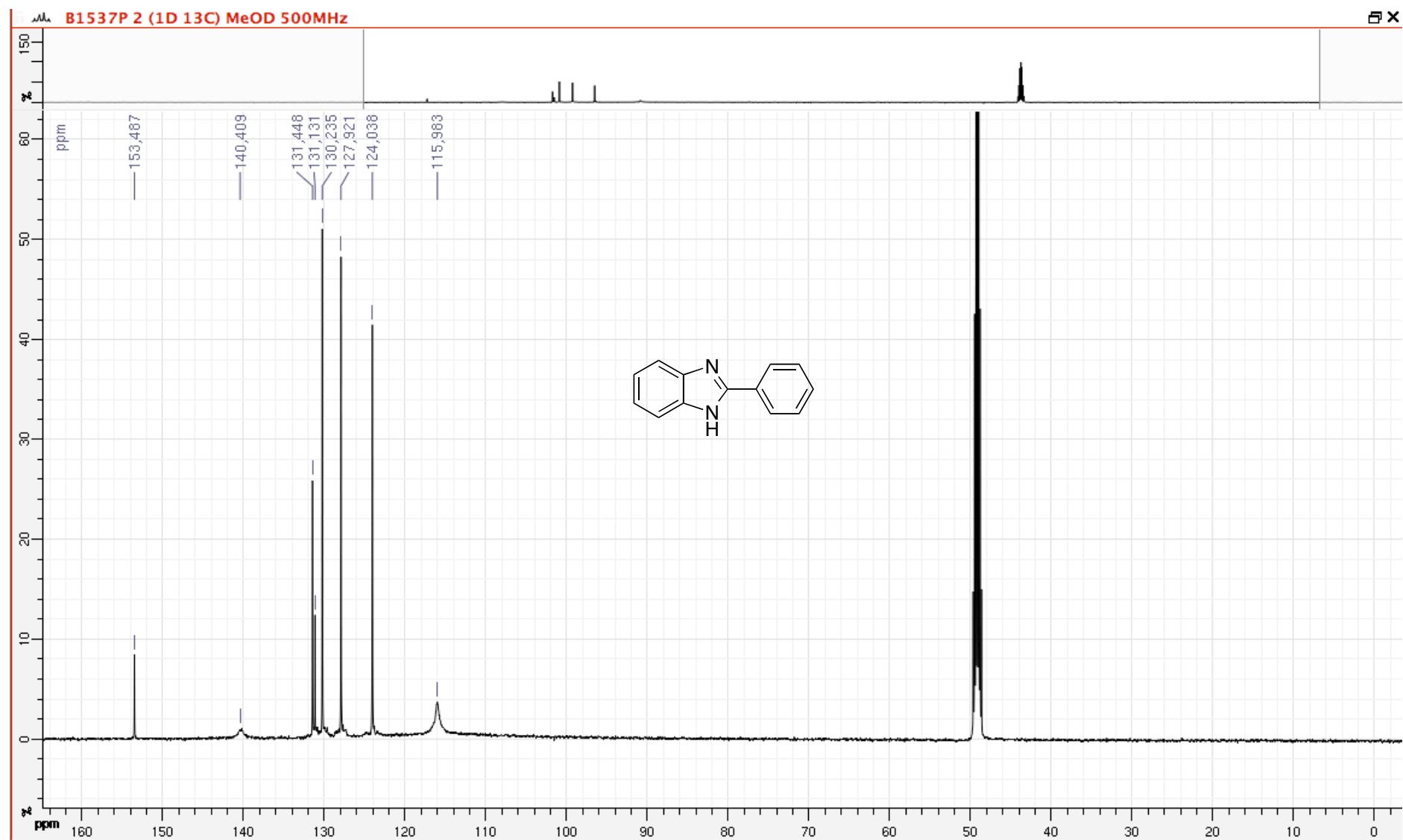
**1-(Pyridin-3-yl)-N-(pyridin-3-ylmethylene)methanamine (2g)**



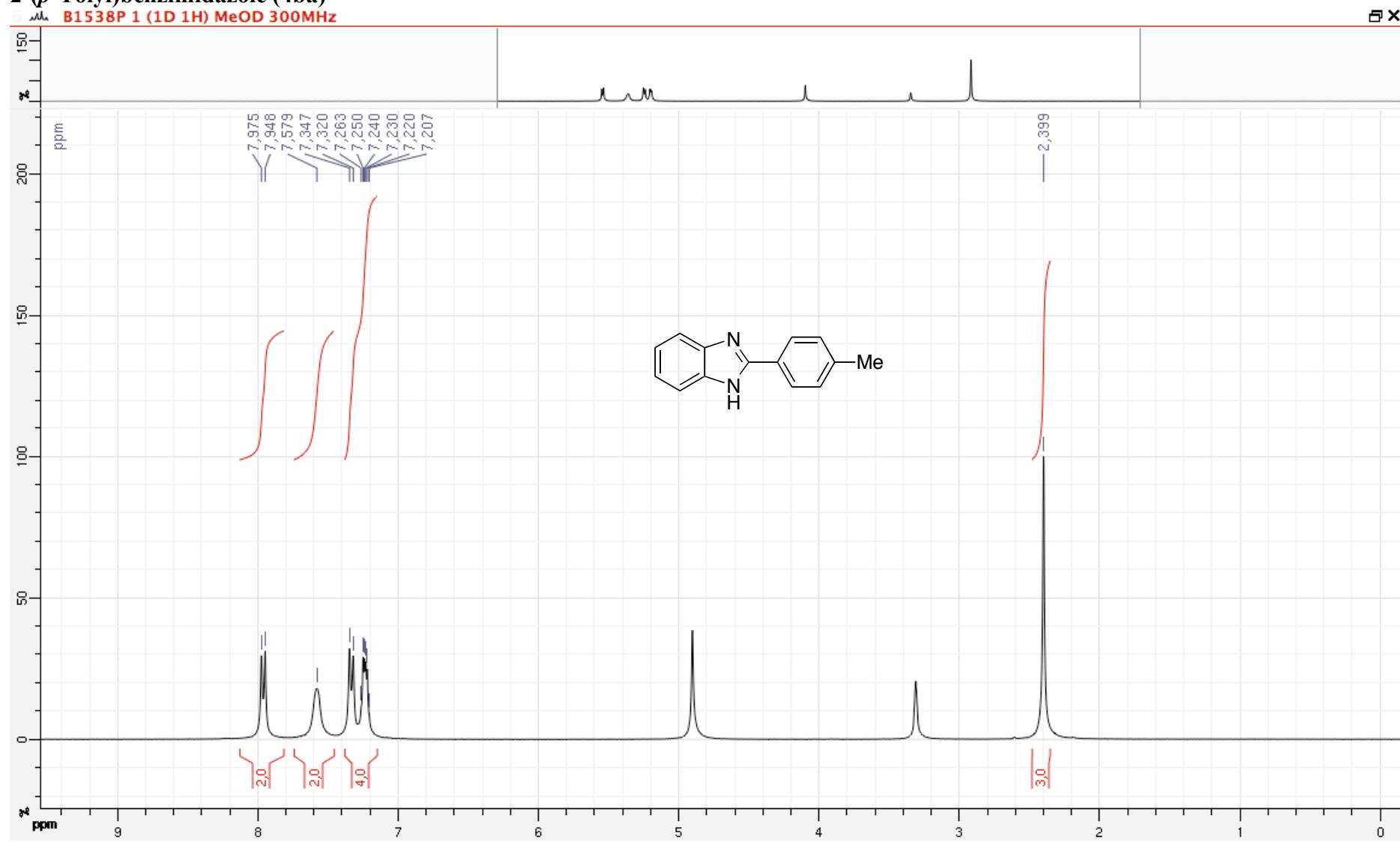


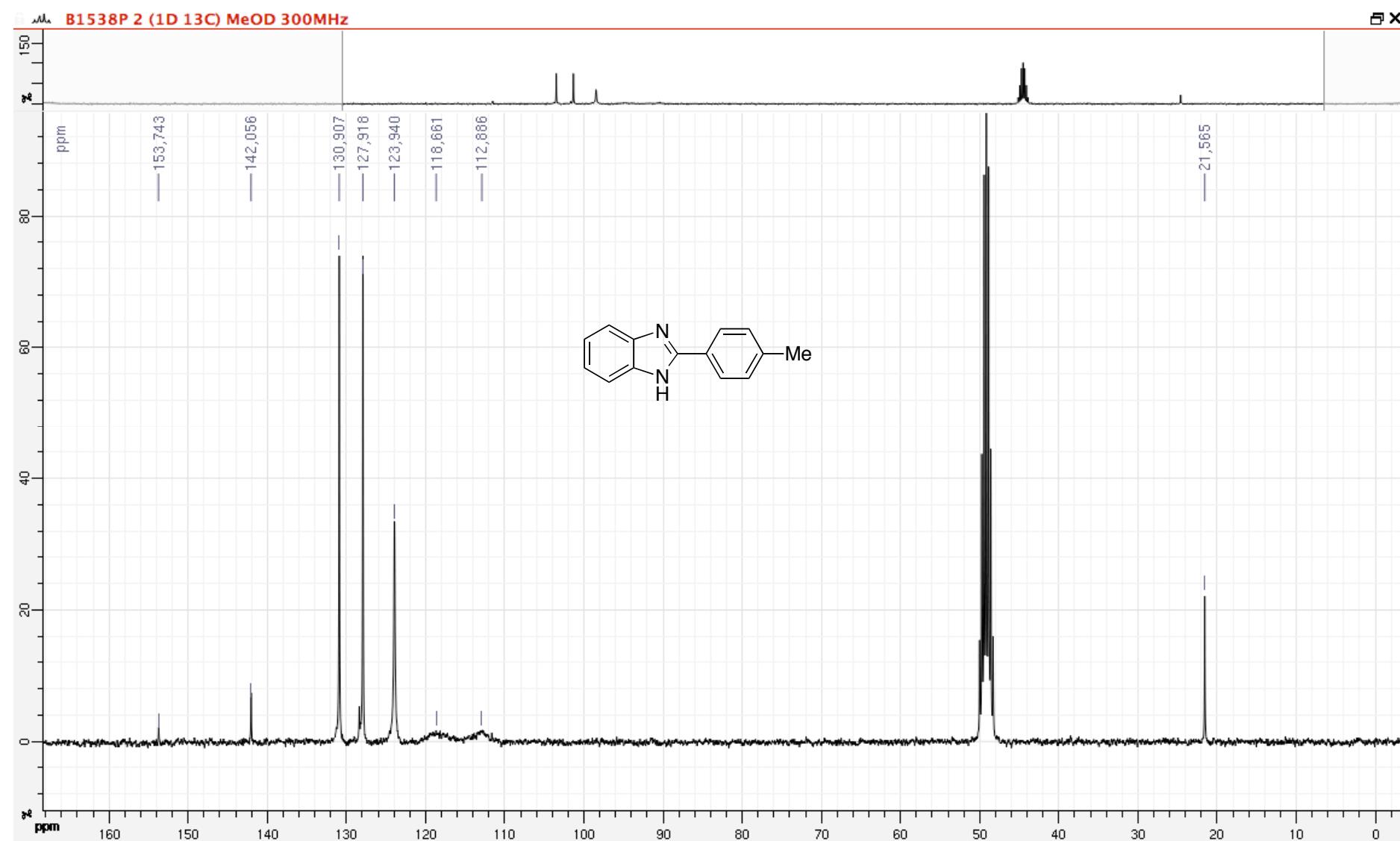
**2-Phenylbenzimidazole (4aa)**



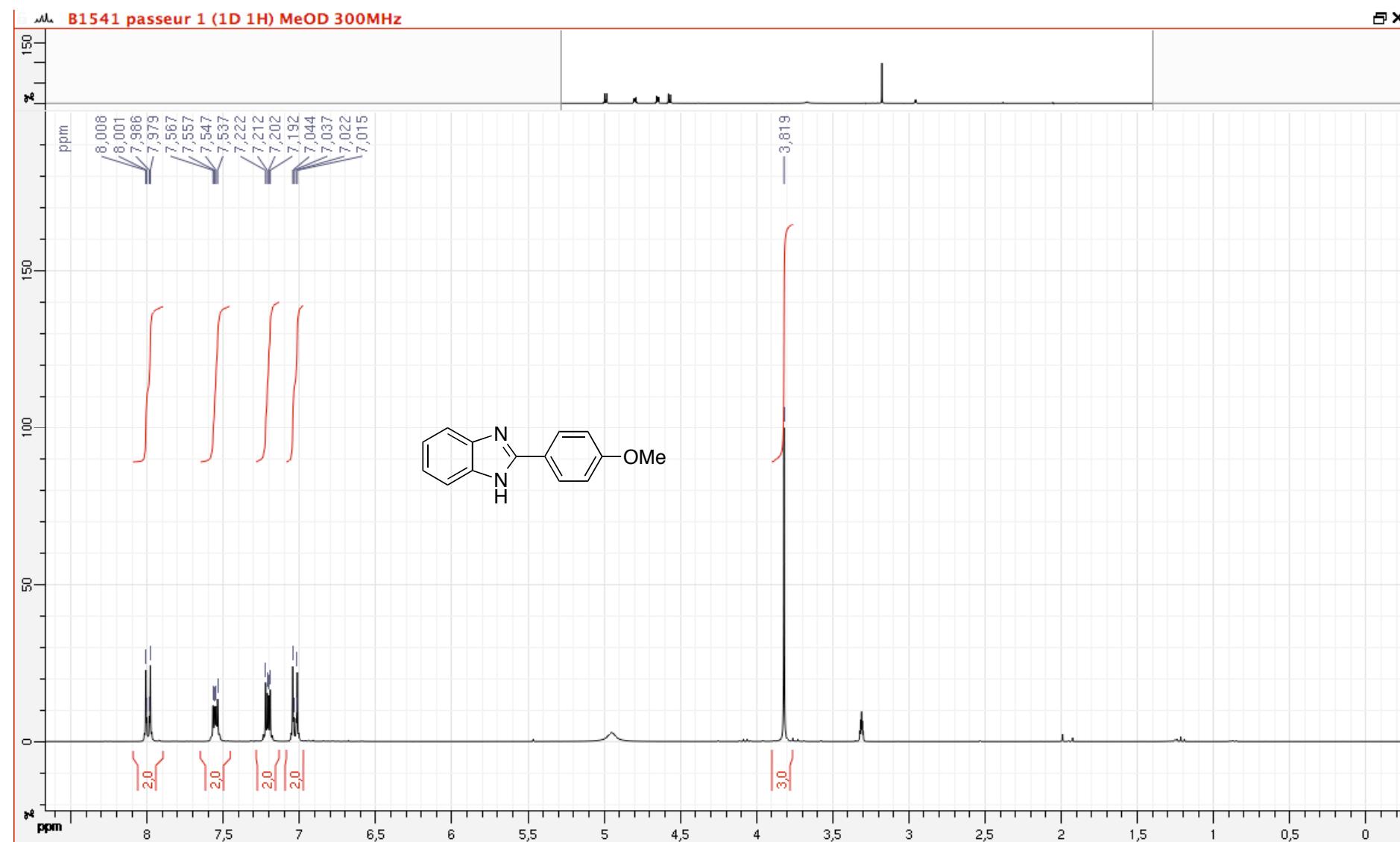


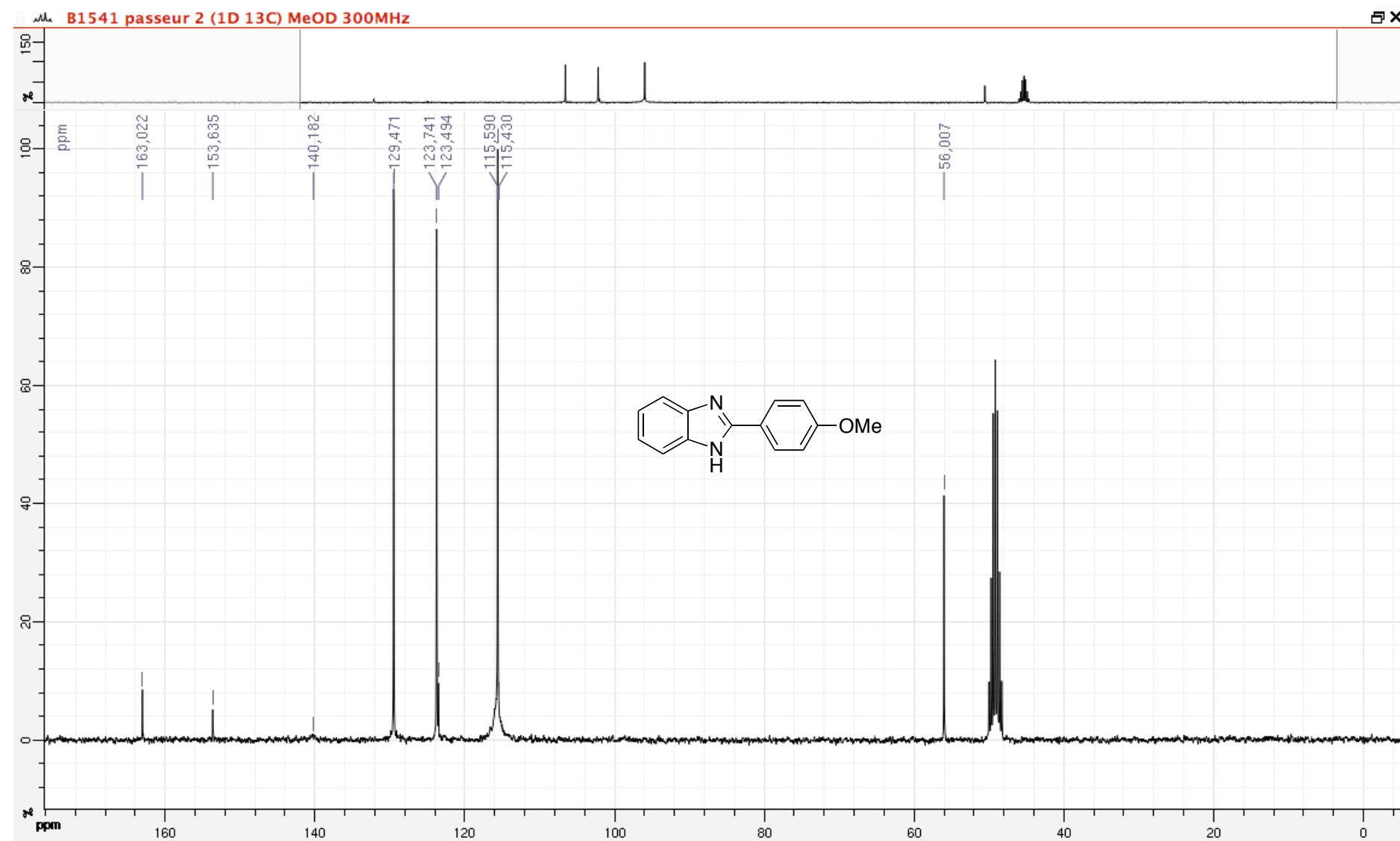
**2-(*p*-Tolyl)benzimidazole (**4ba**)**



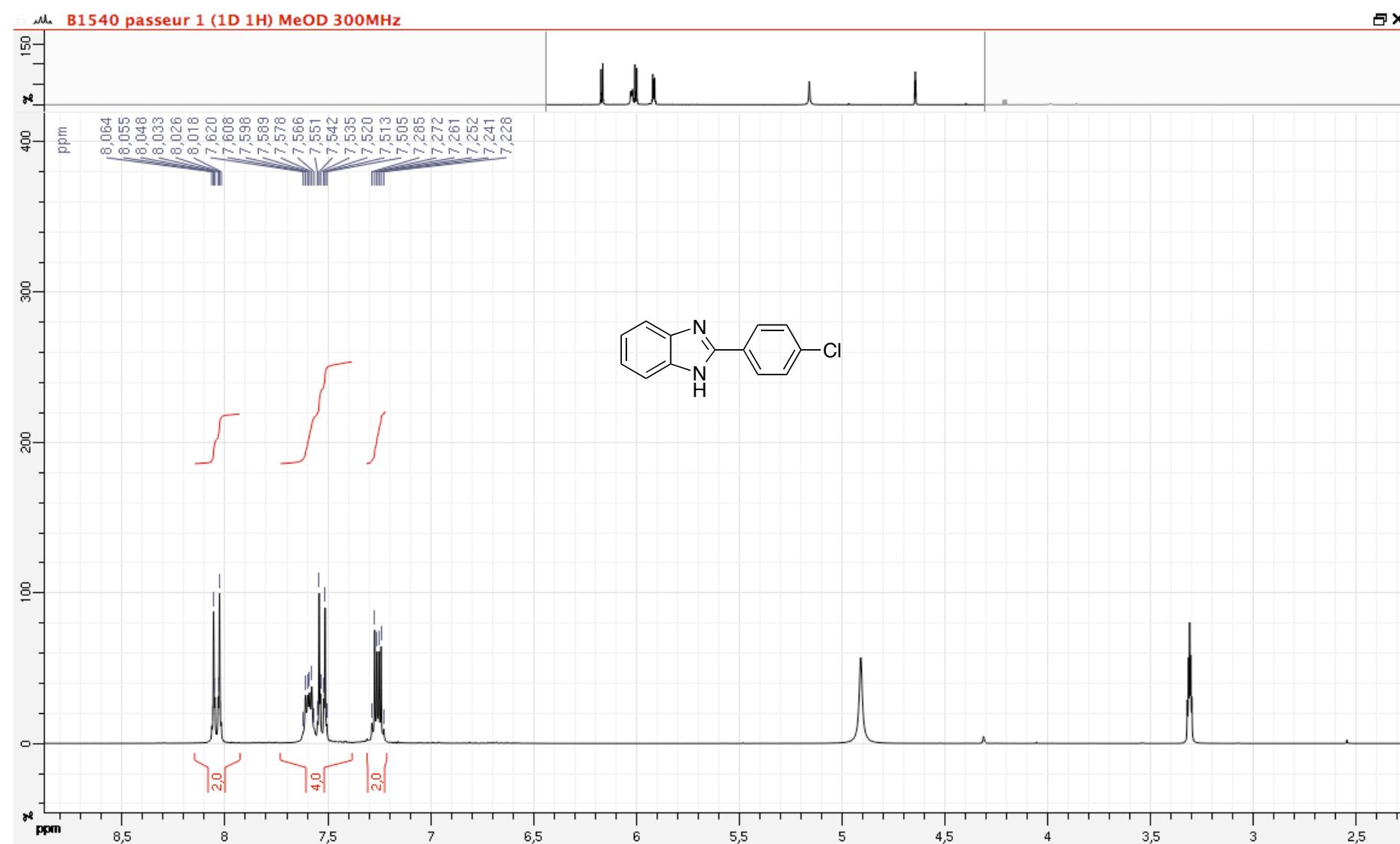


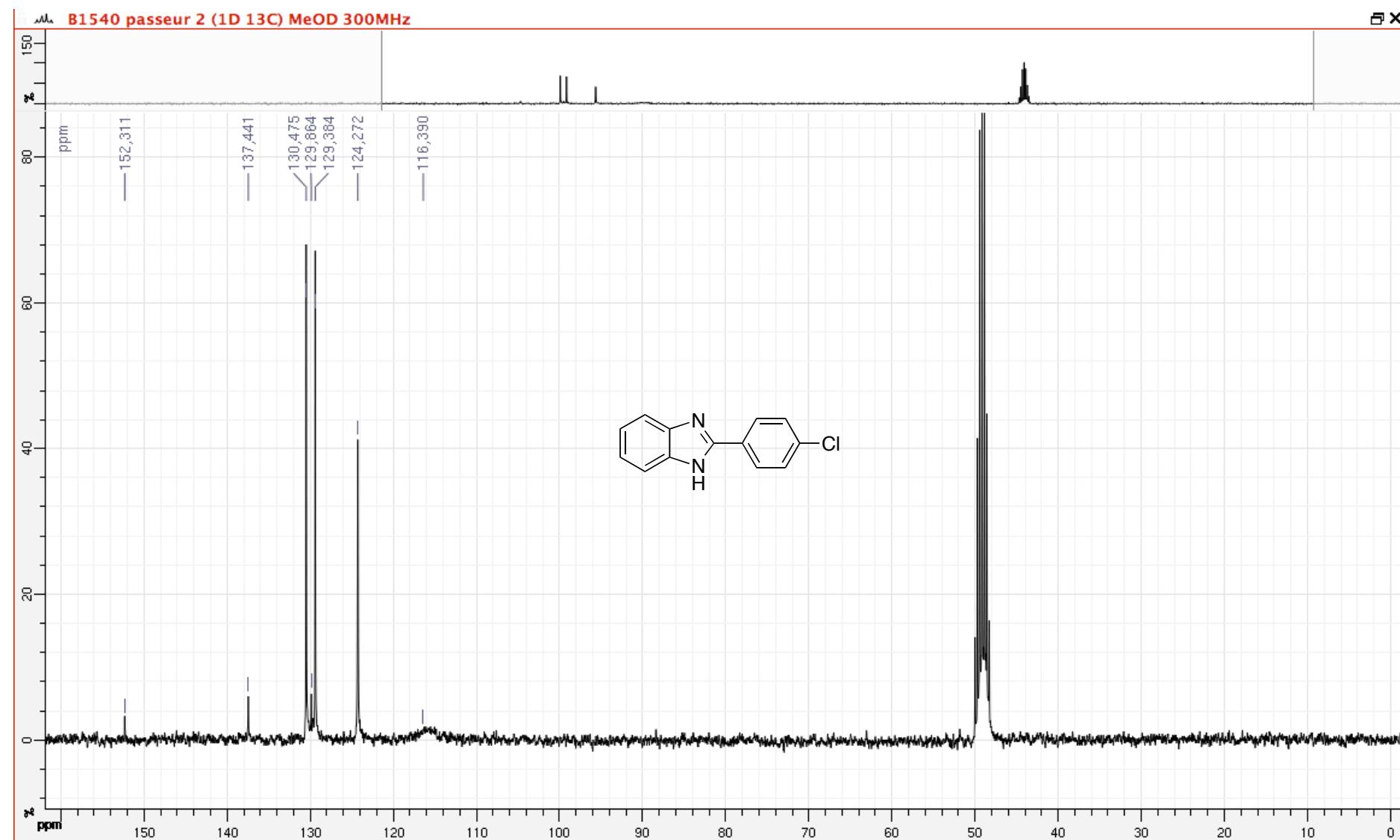
**2-(4-Methoxyphenyl)benzimidazole (4ca)**



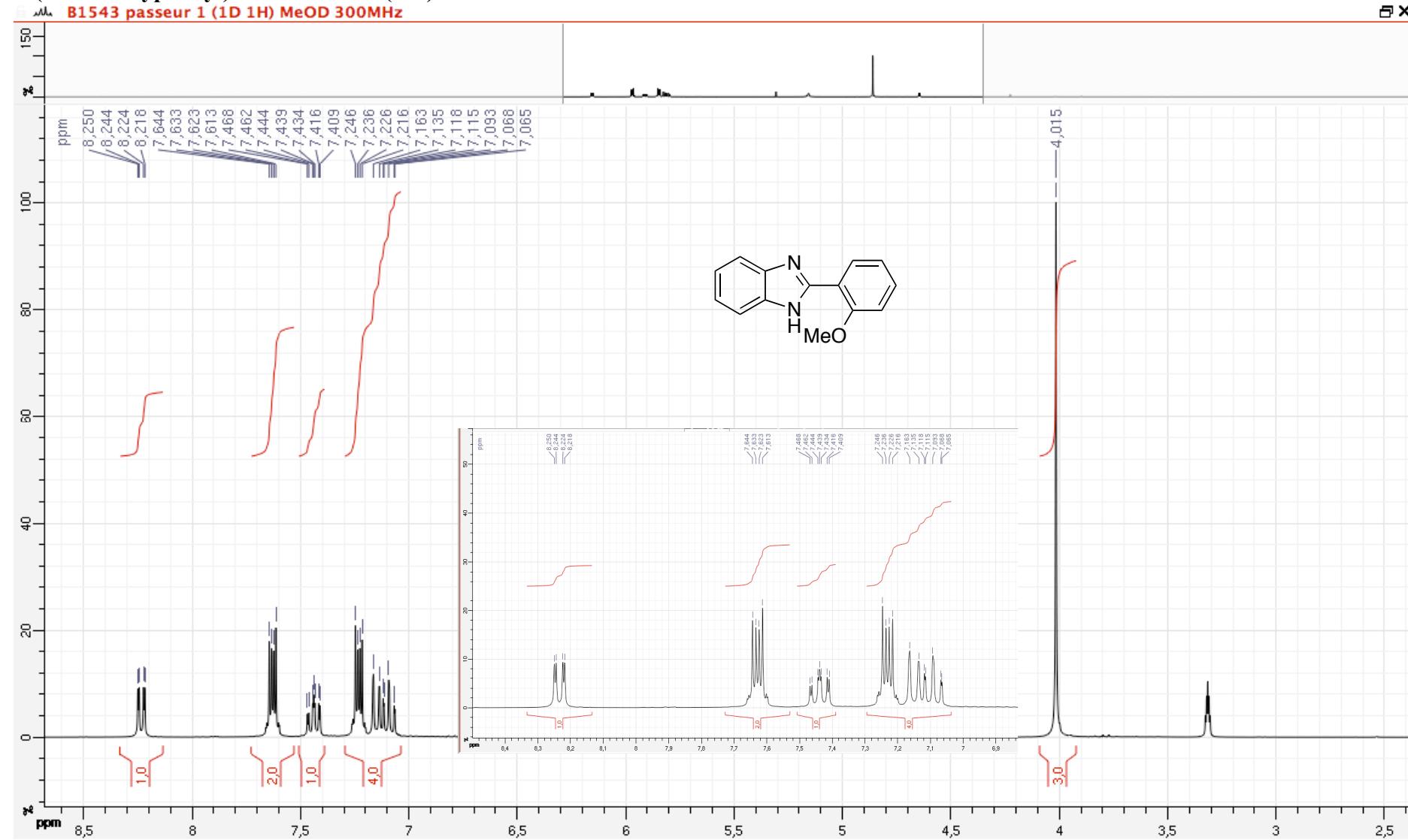


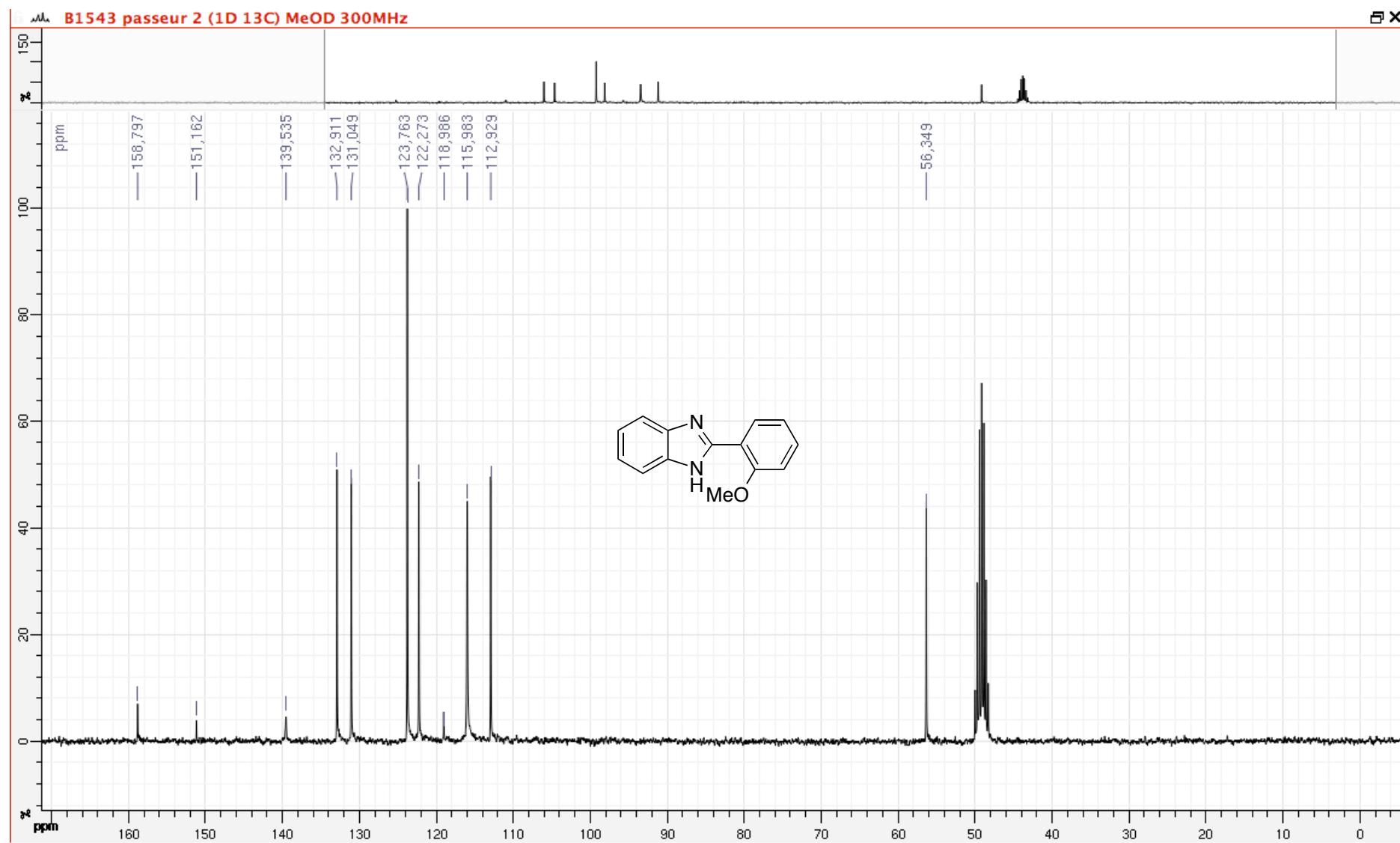
**2-(4-Chlorophenyl)benzimidazole (4da)**



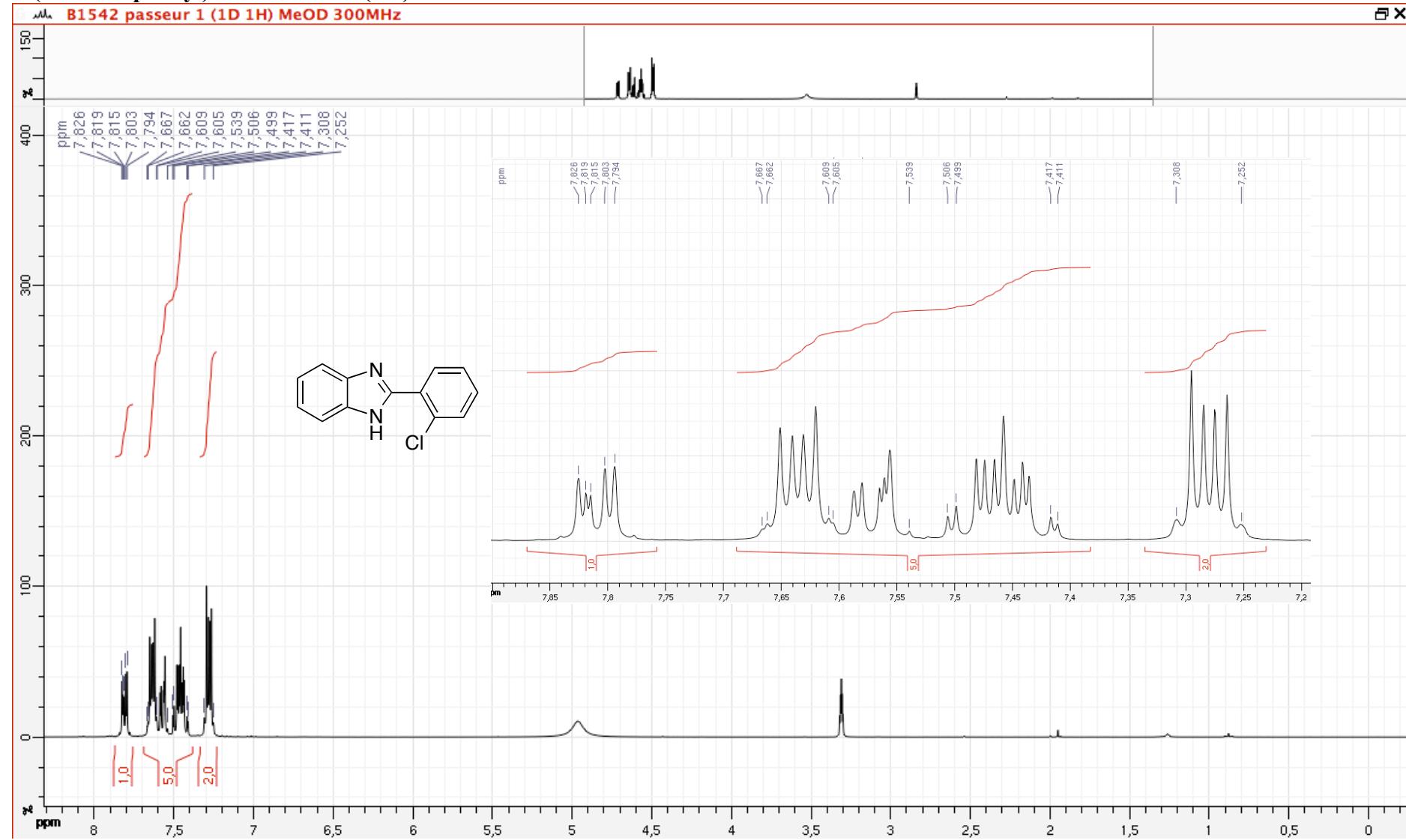


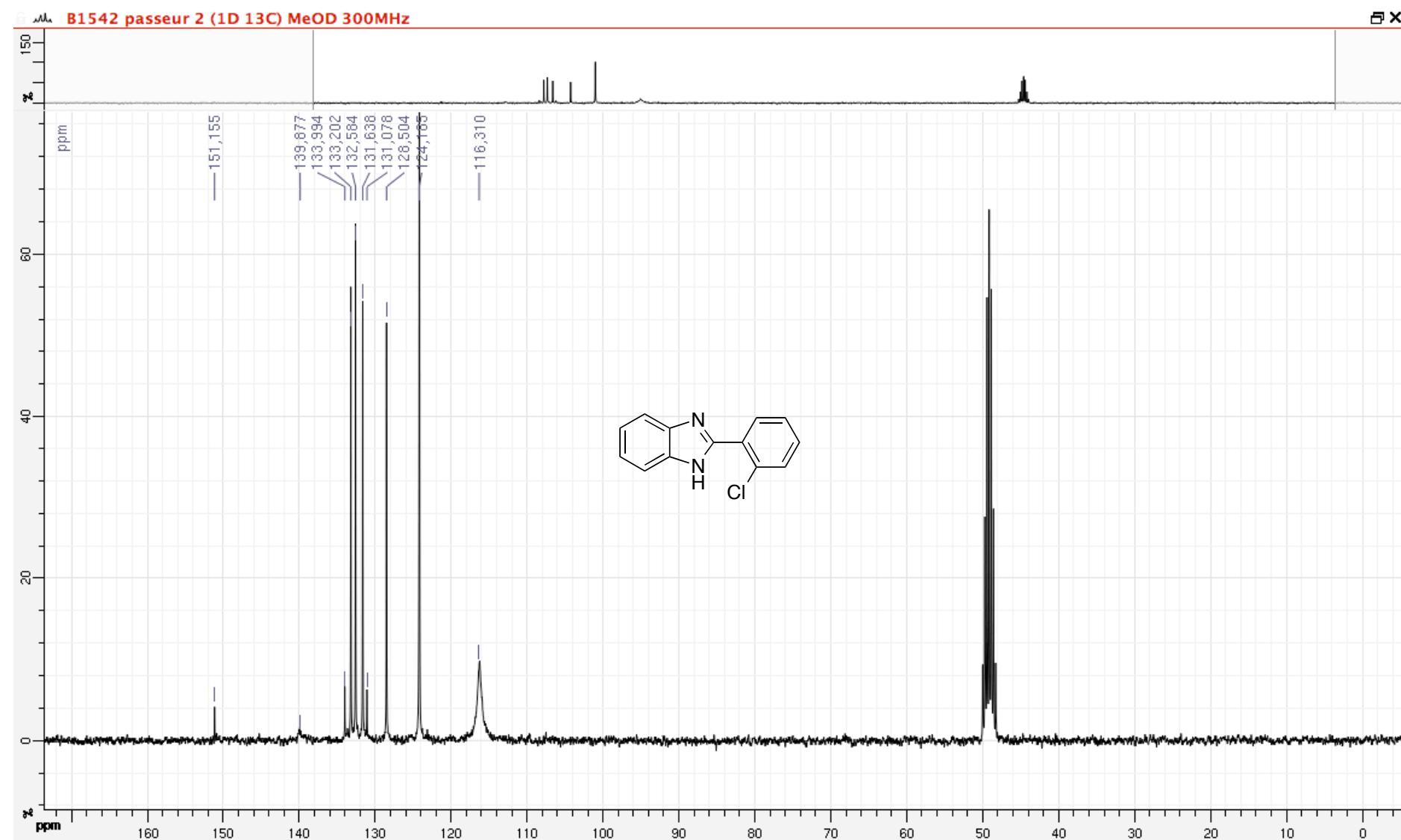
**2-(2-Methoxyphenyl)benzimidazole (4ea)**



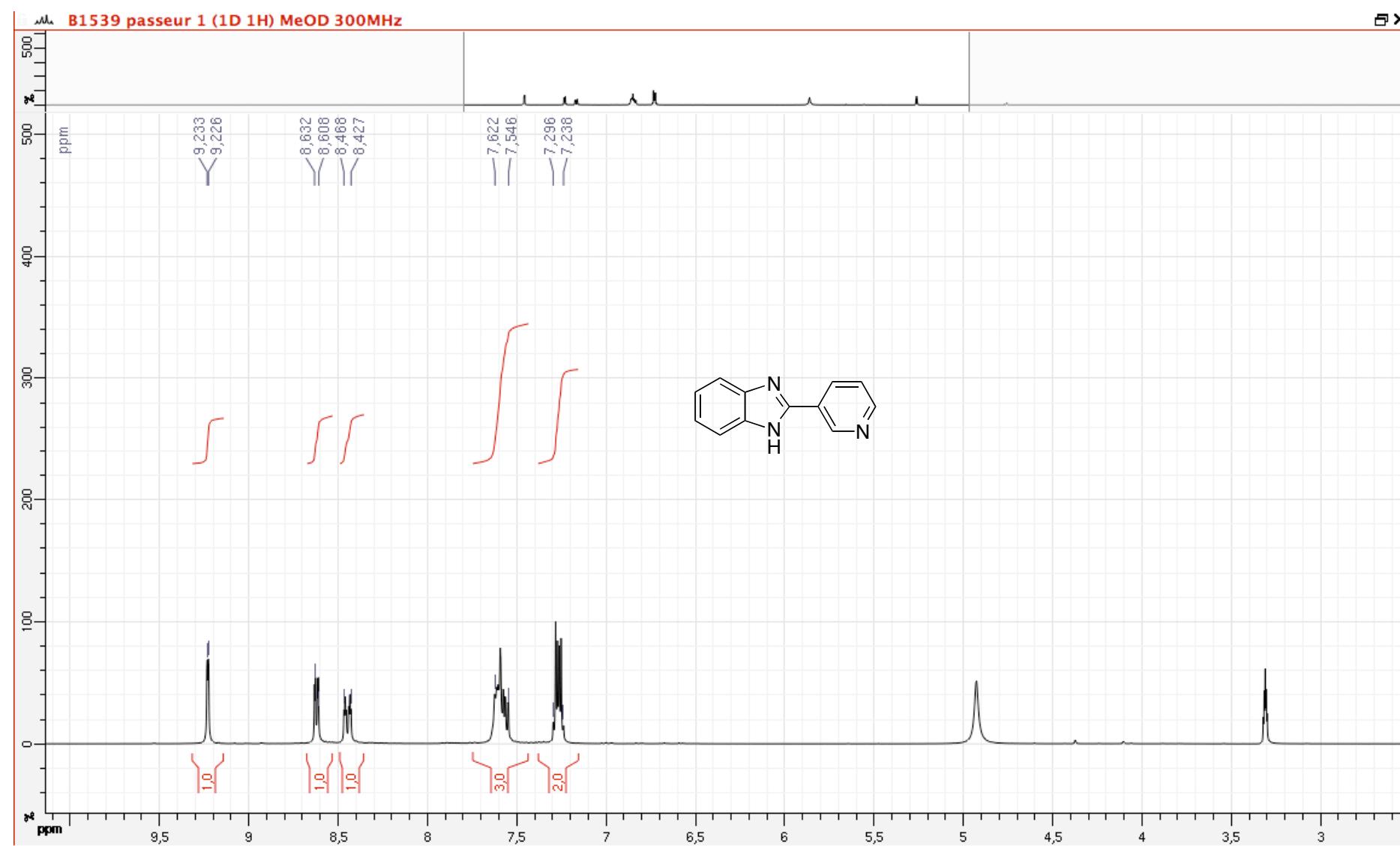


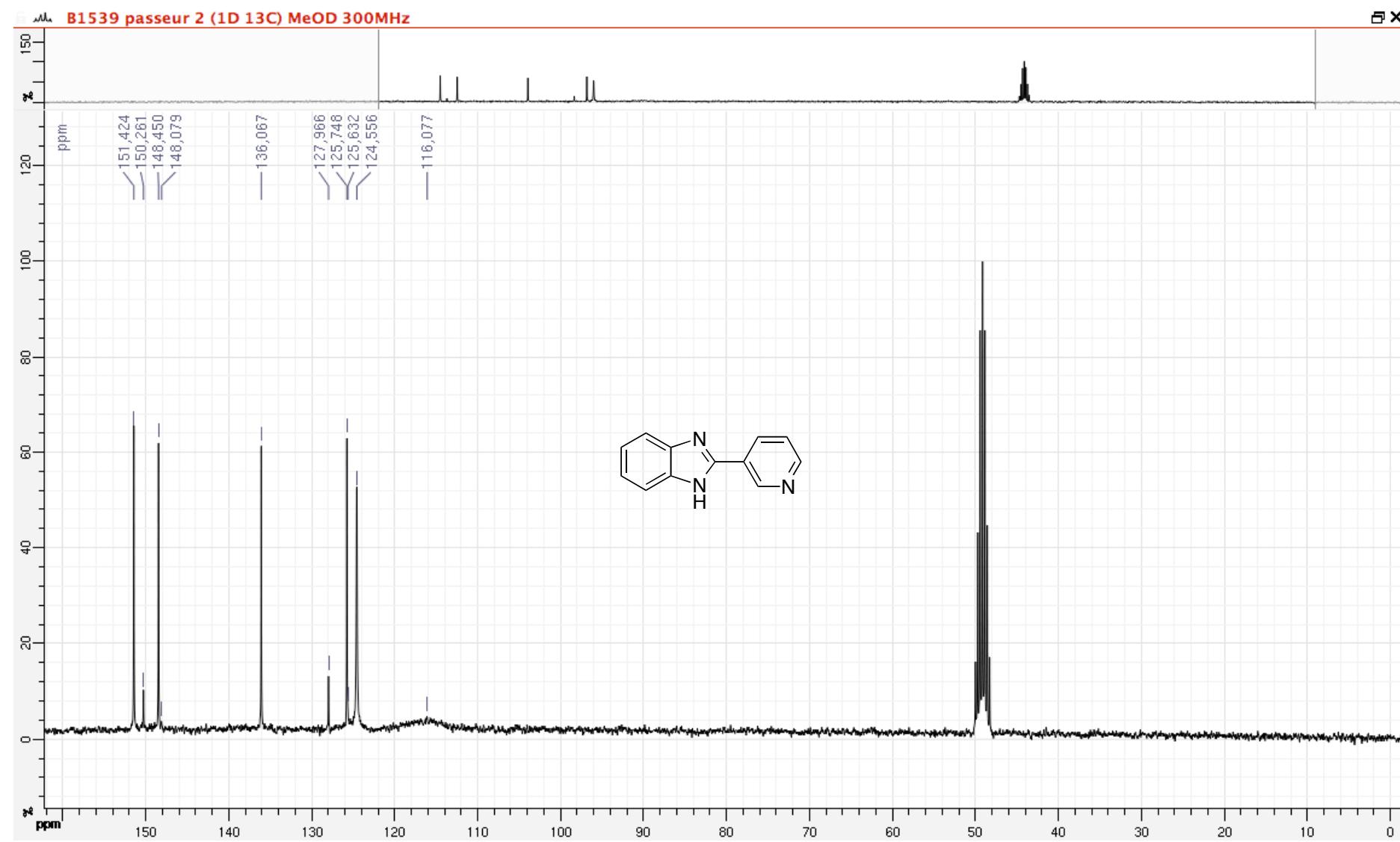
**2-(2-Chlorophenyl)benzimidazole (4fa)**



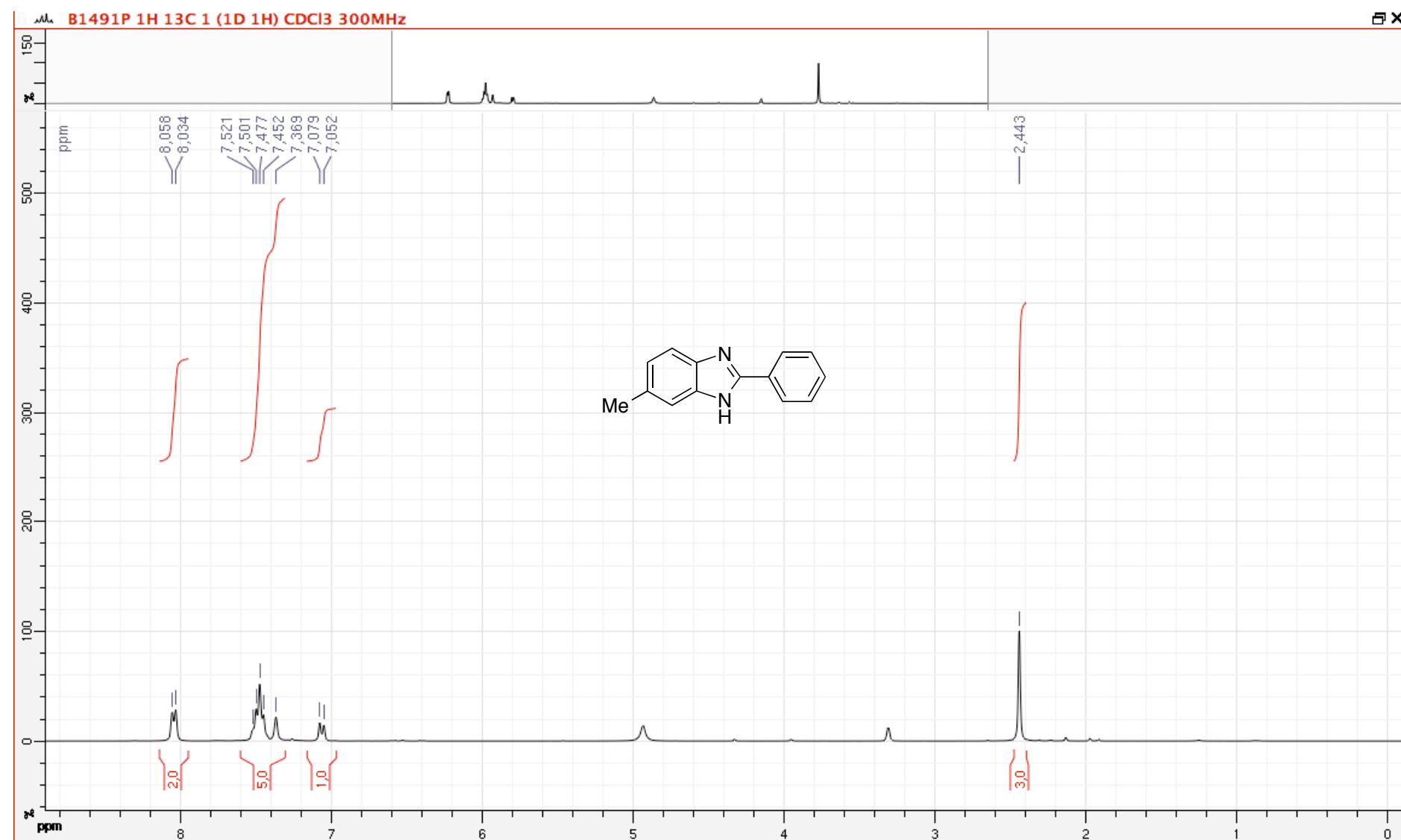


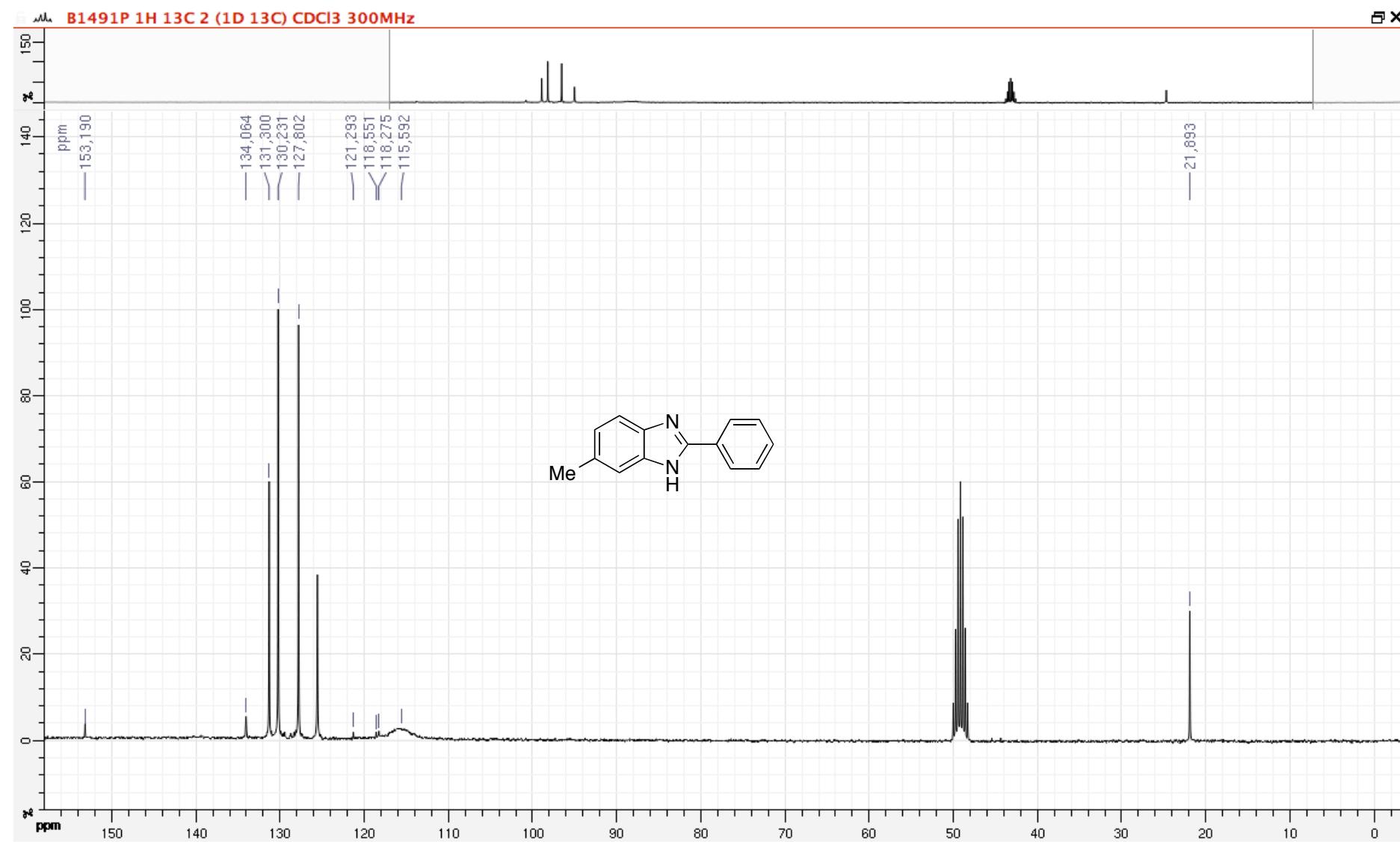
**2-(Pyridin-3-yl)benzimidazole (4ga)**



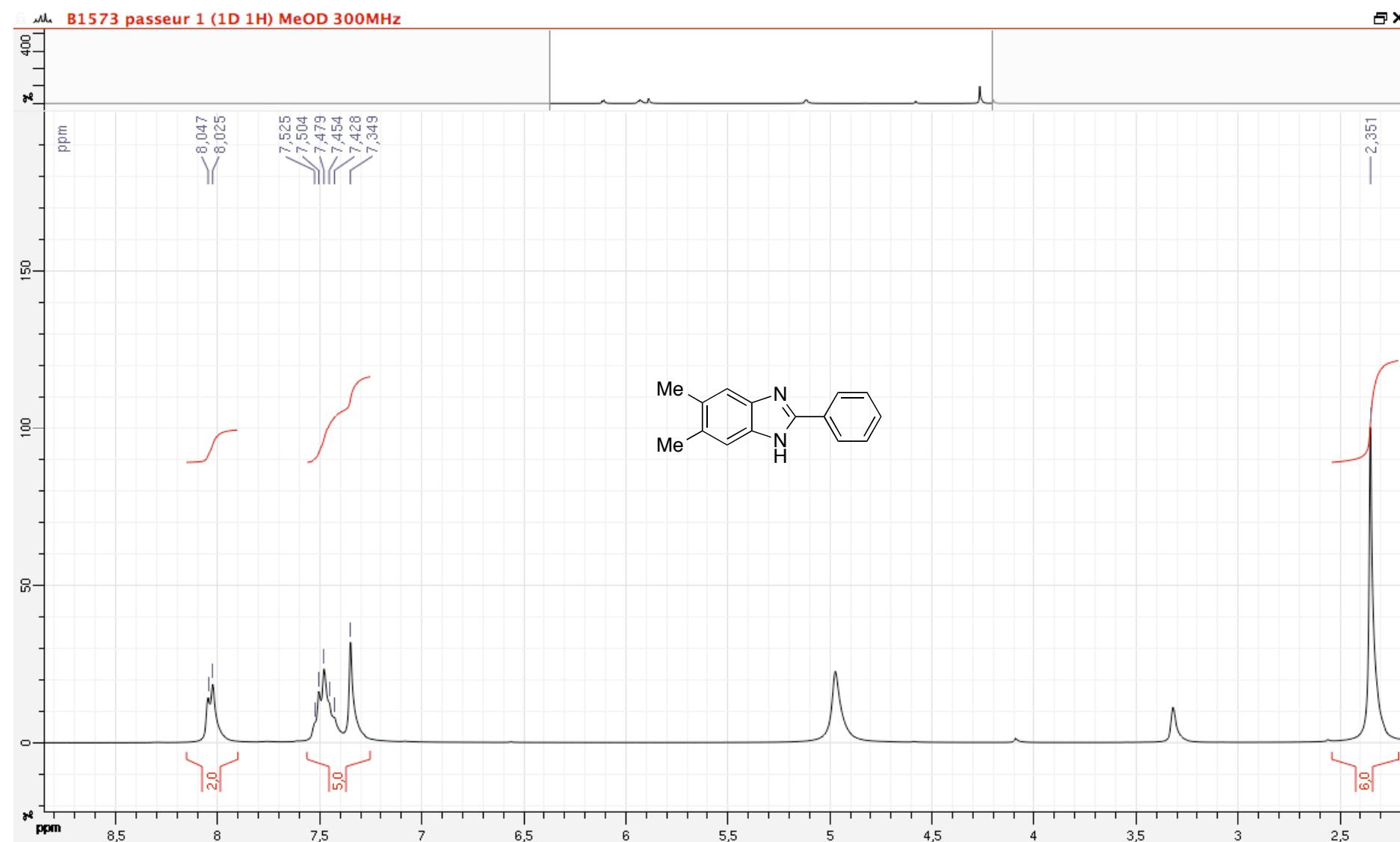


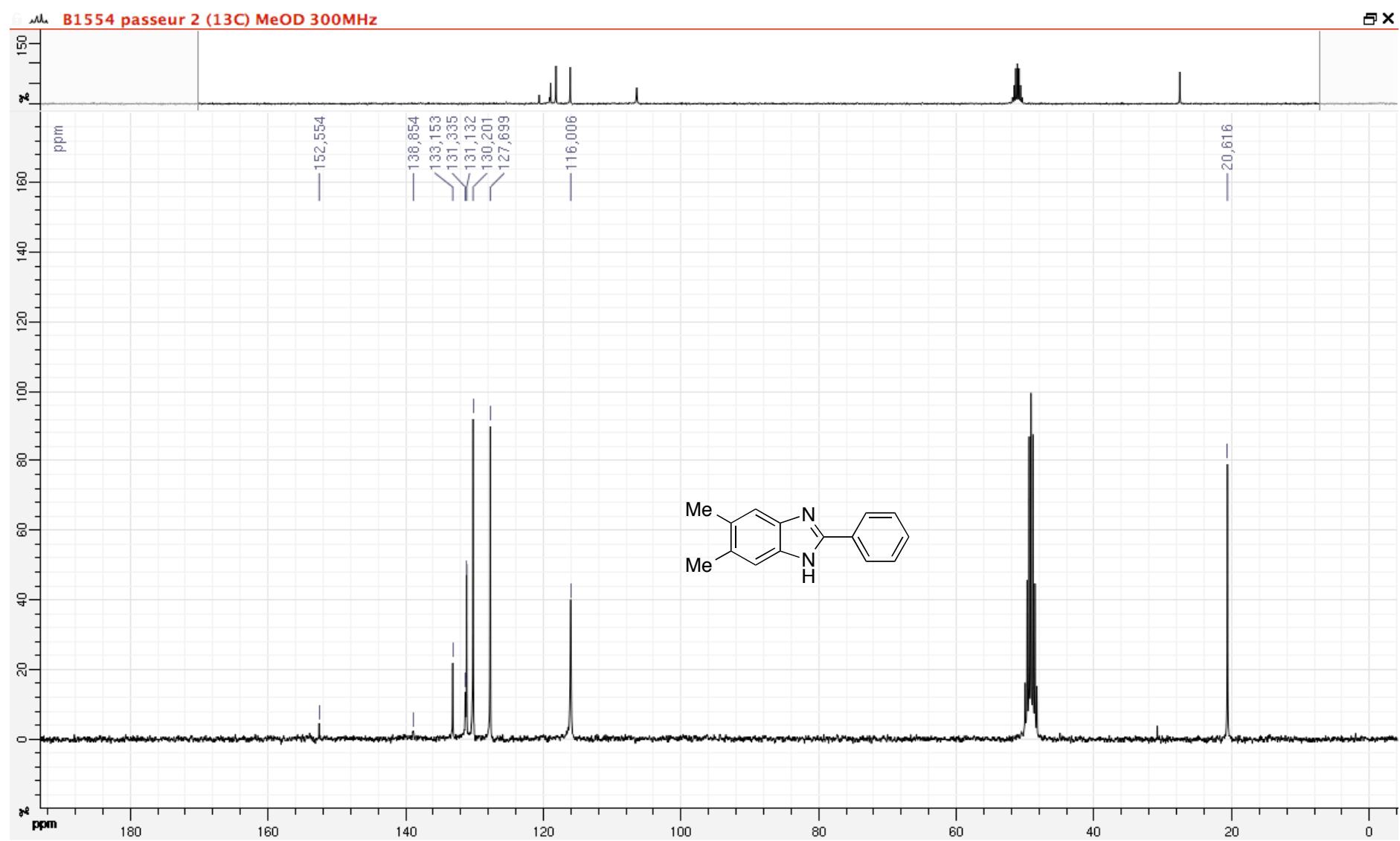
**6-Methyl-2-phenylbenzimidazole (4ab)**



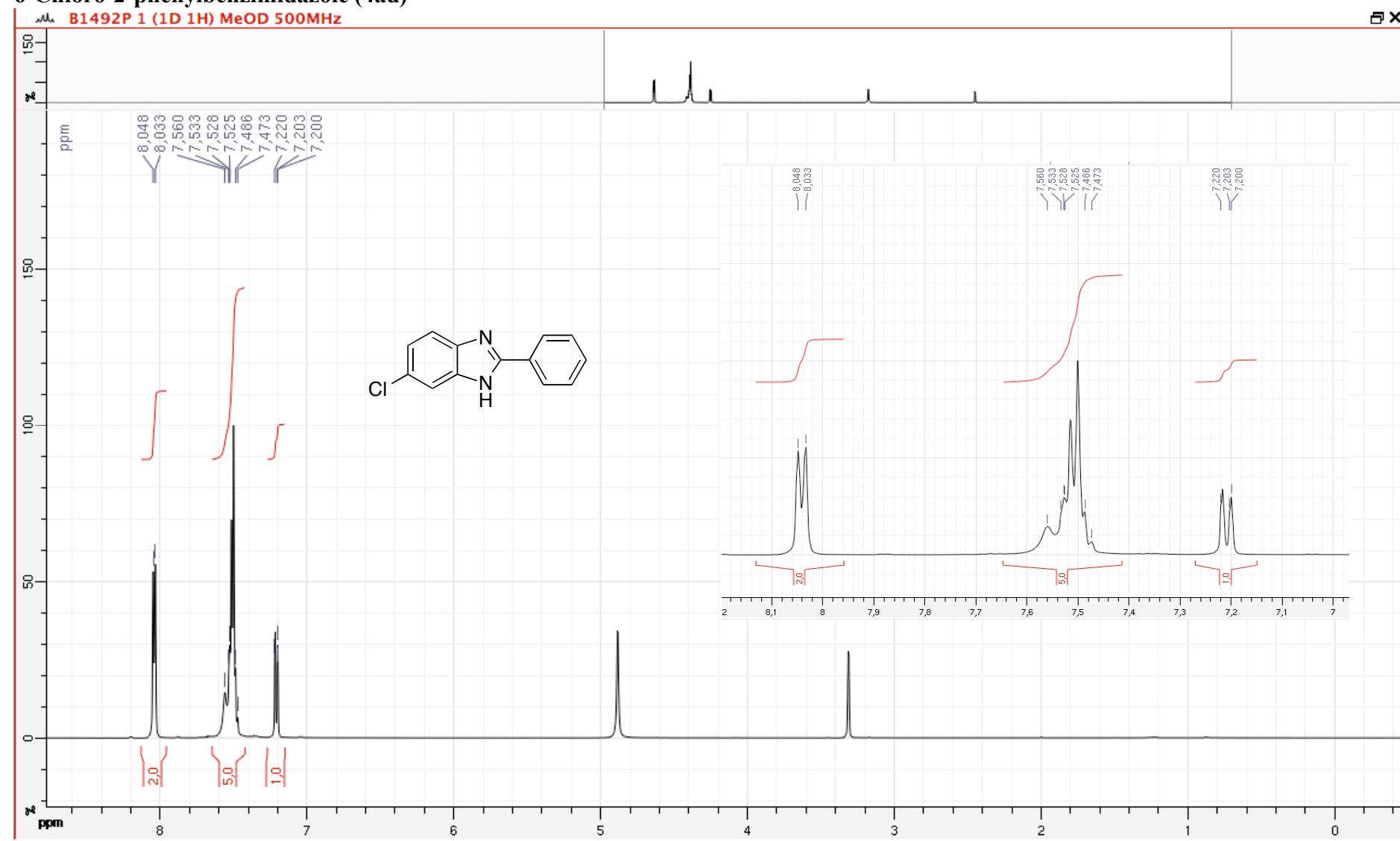


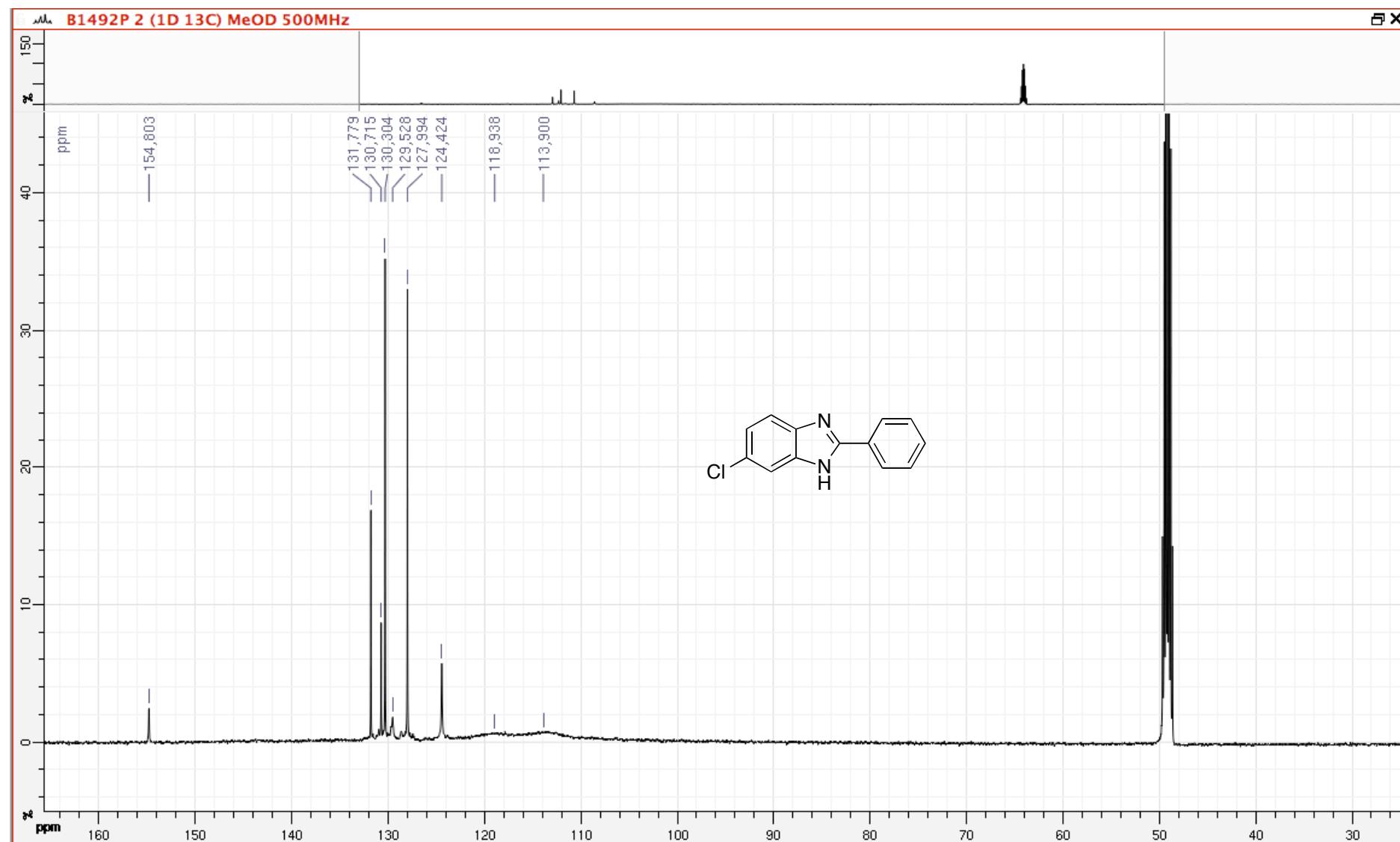
**5,6-Dimethyl-2-phenylbenzimidazole (4ac)**



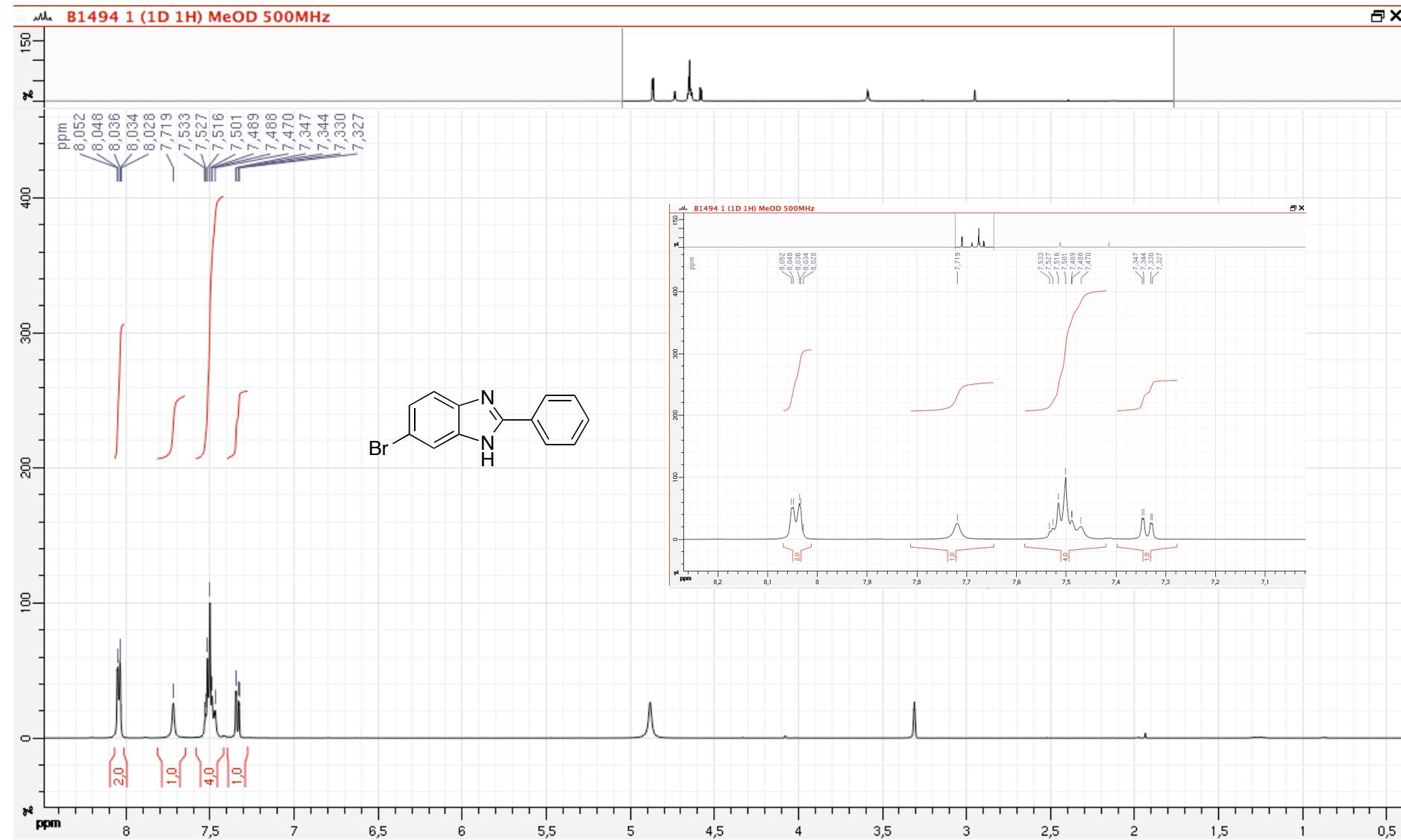


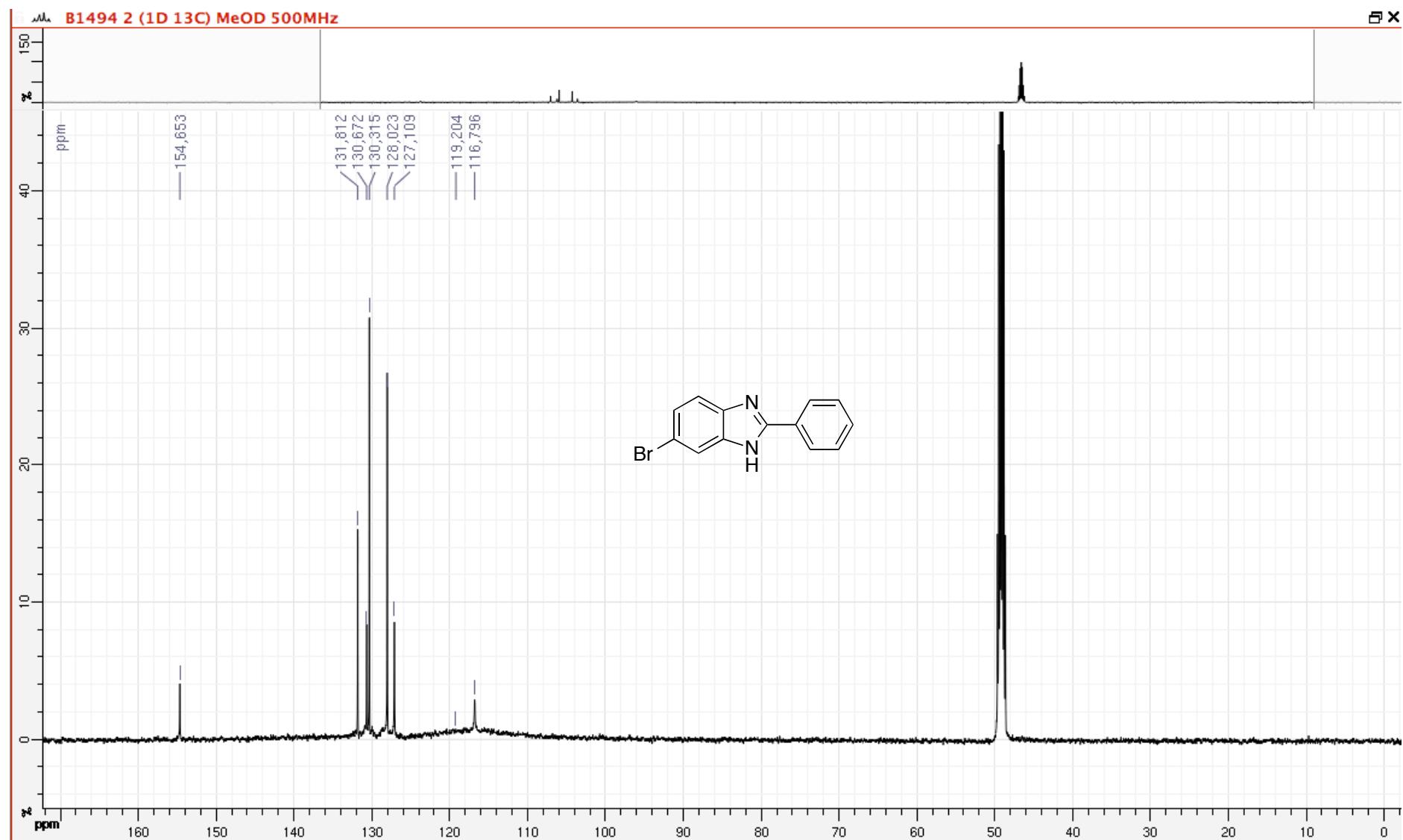
**6-Chloro-2-phenylbenzimidazole (4ad)**



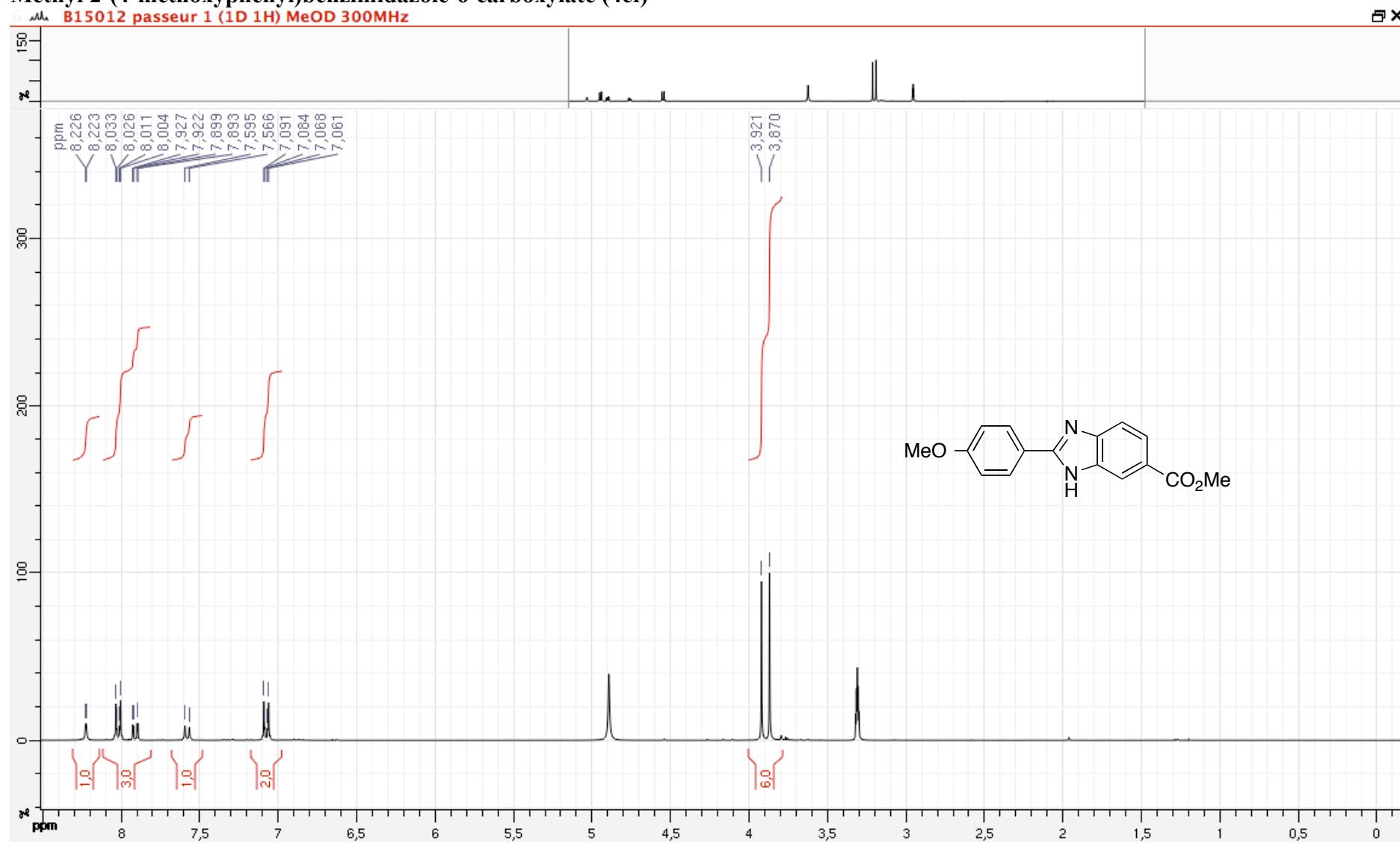


**6-Bromo-2-phenylbenzimidazole (4ae)**



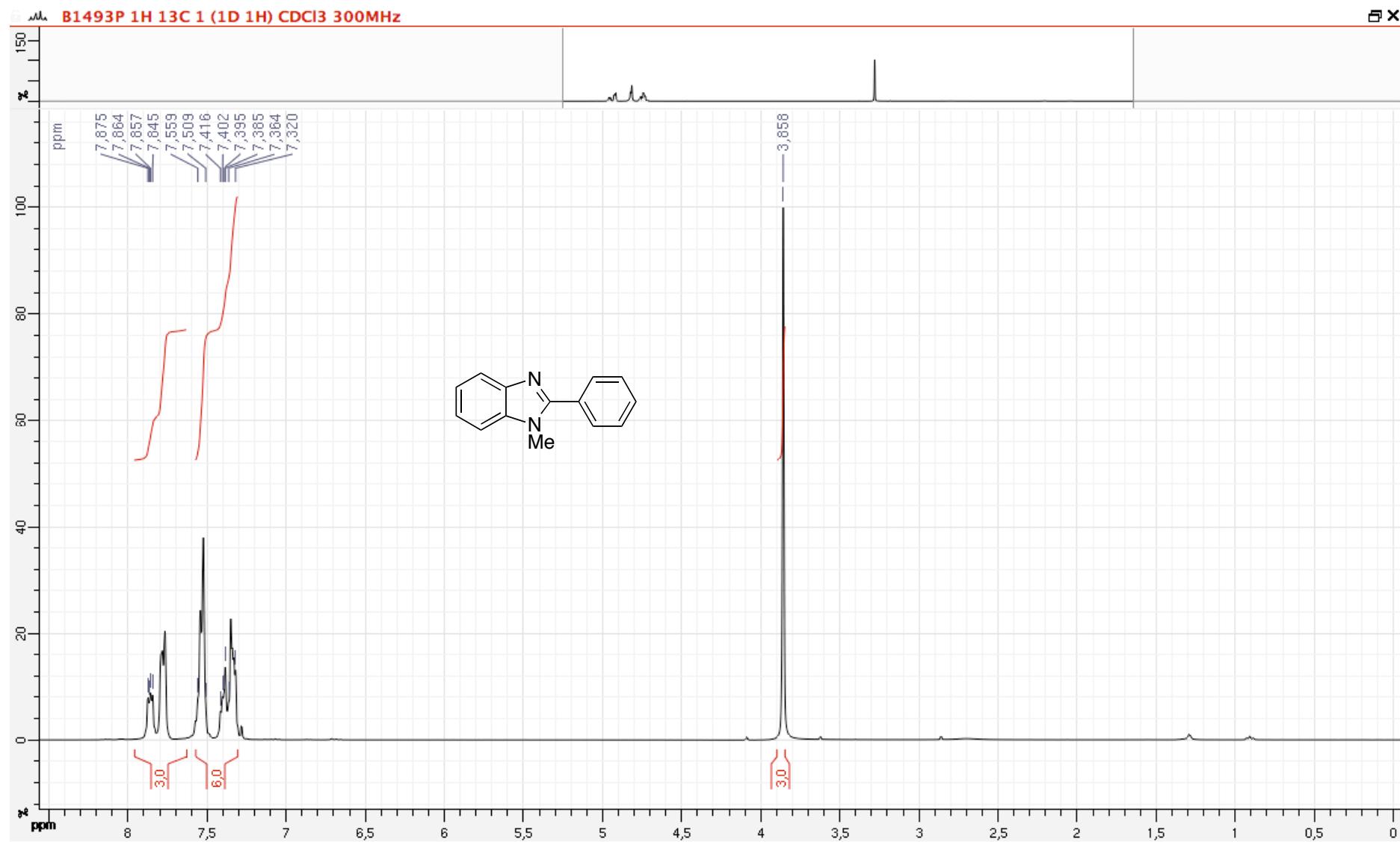


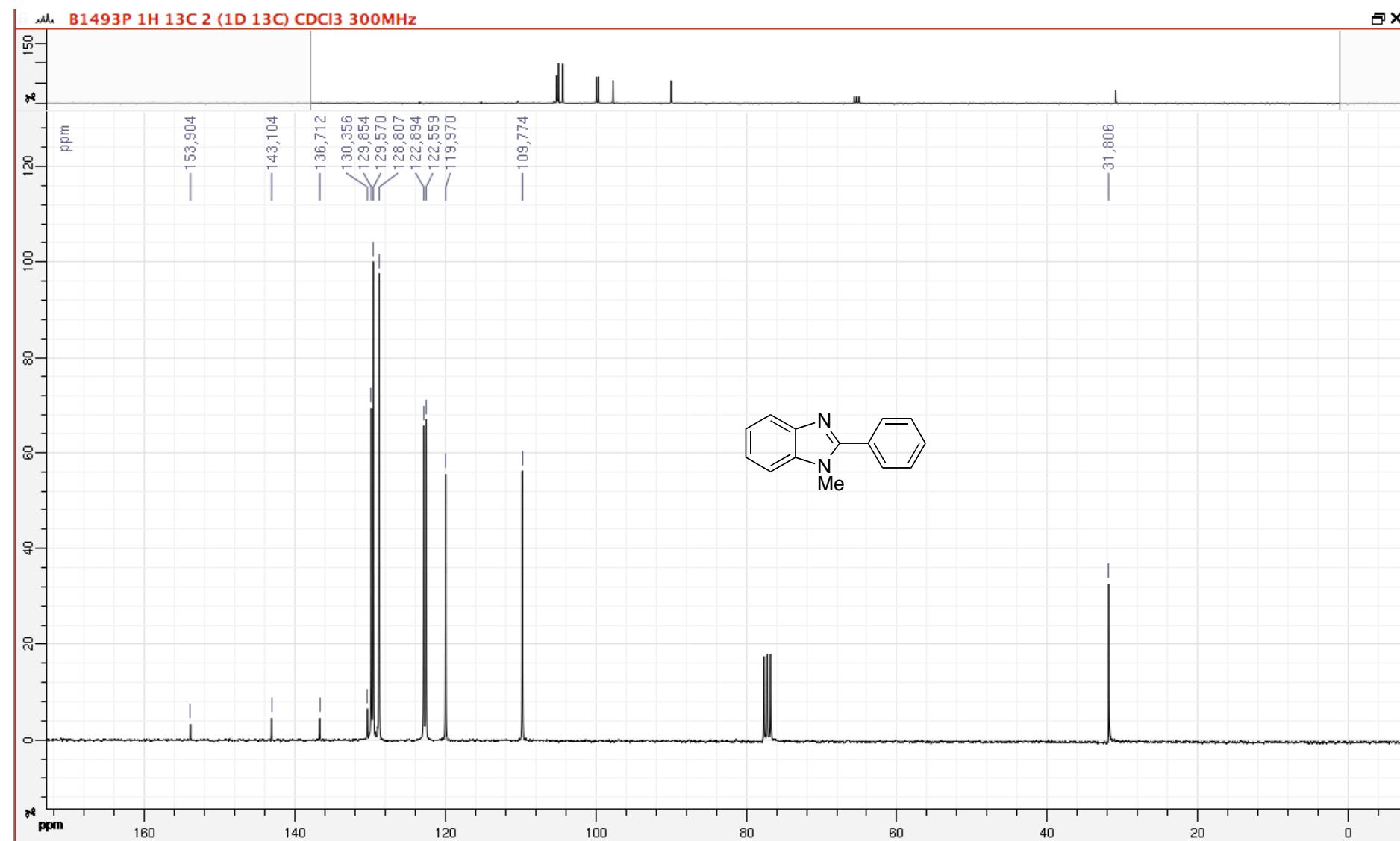
**Methyl 2-(4-methoxyphenyl)benzimidazole-6-carboxylate (4cf)**



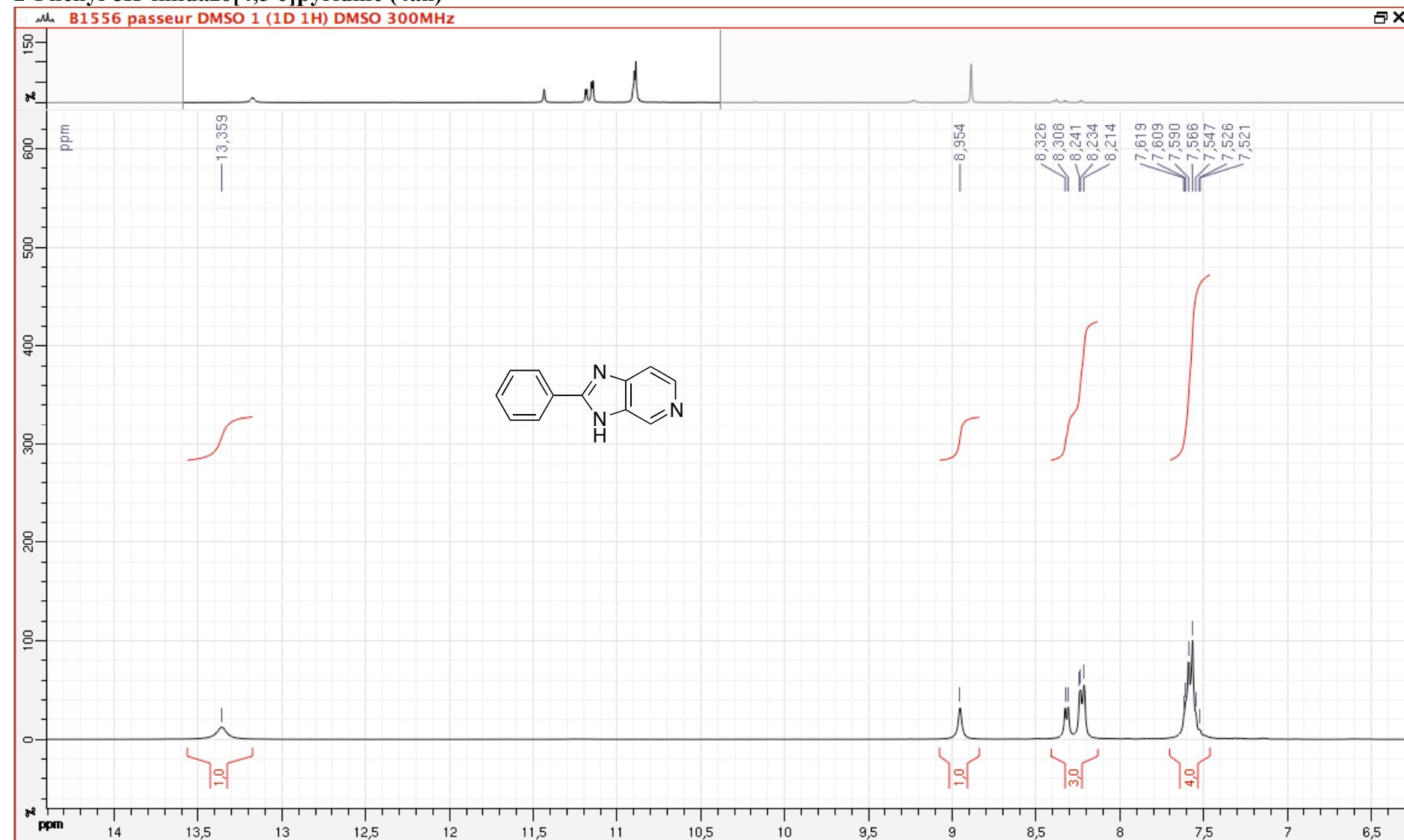


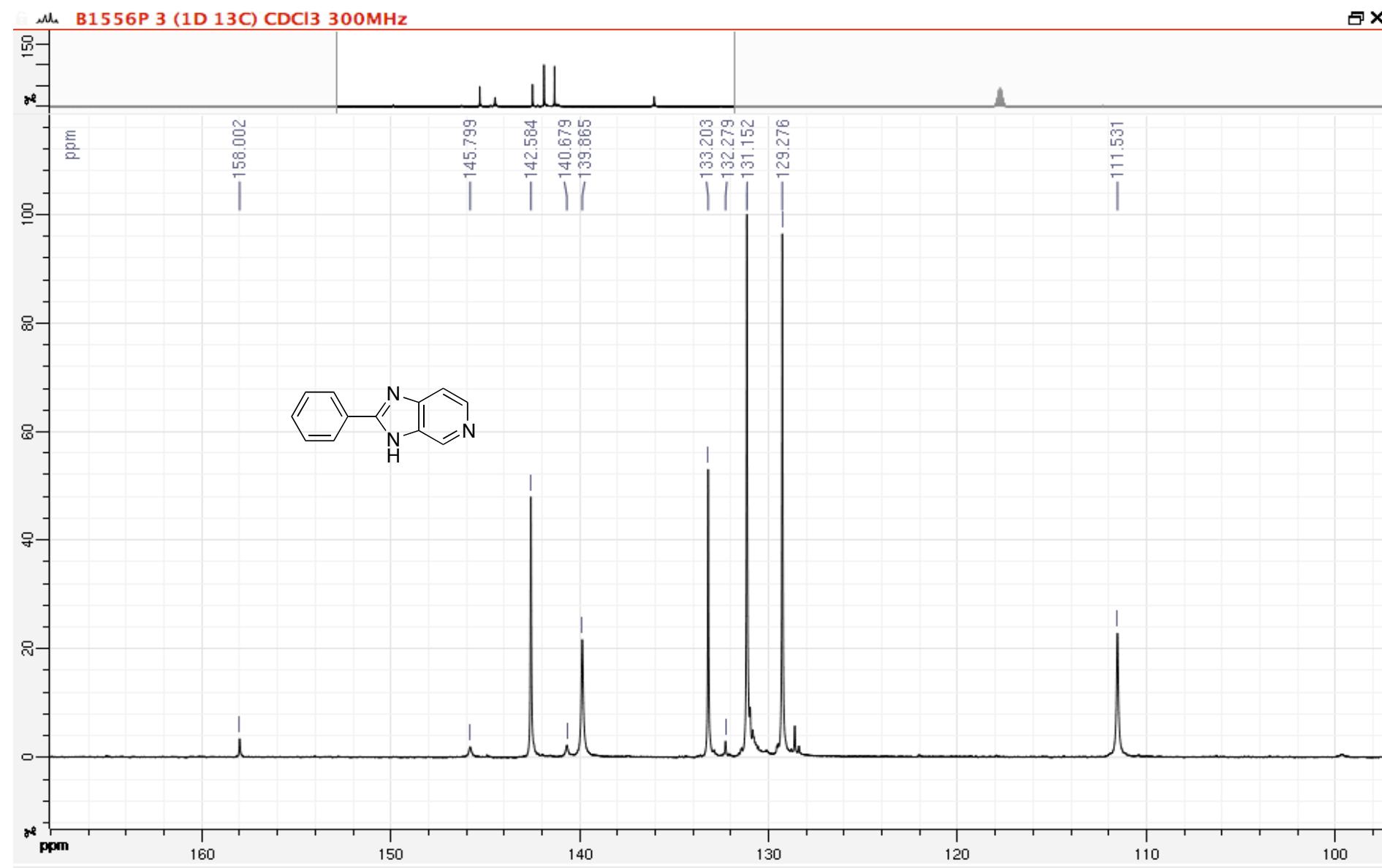
**1-Methyl-2-phenylbenzimidazole (4cg)**



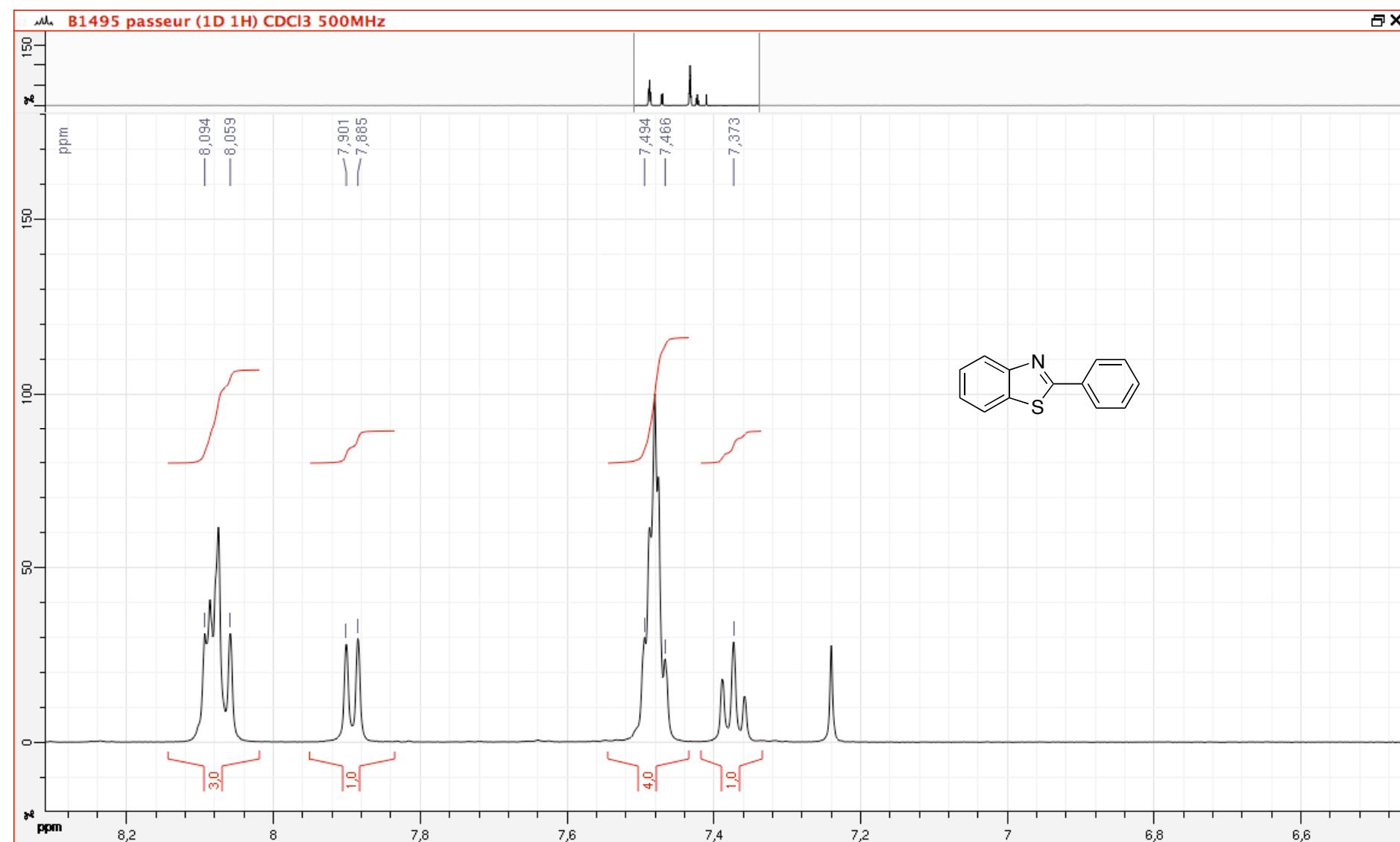


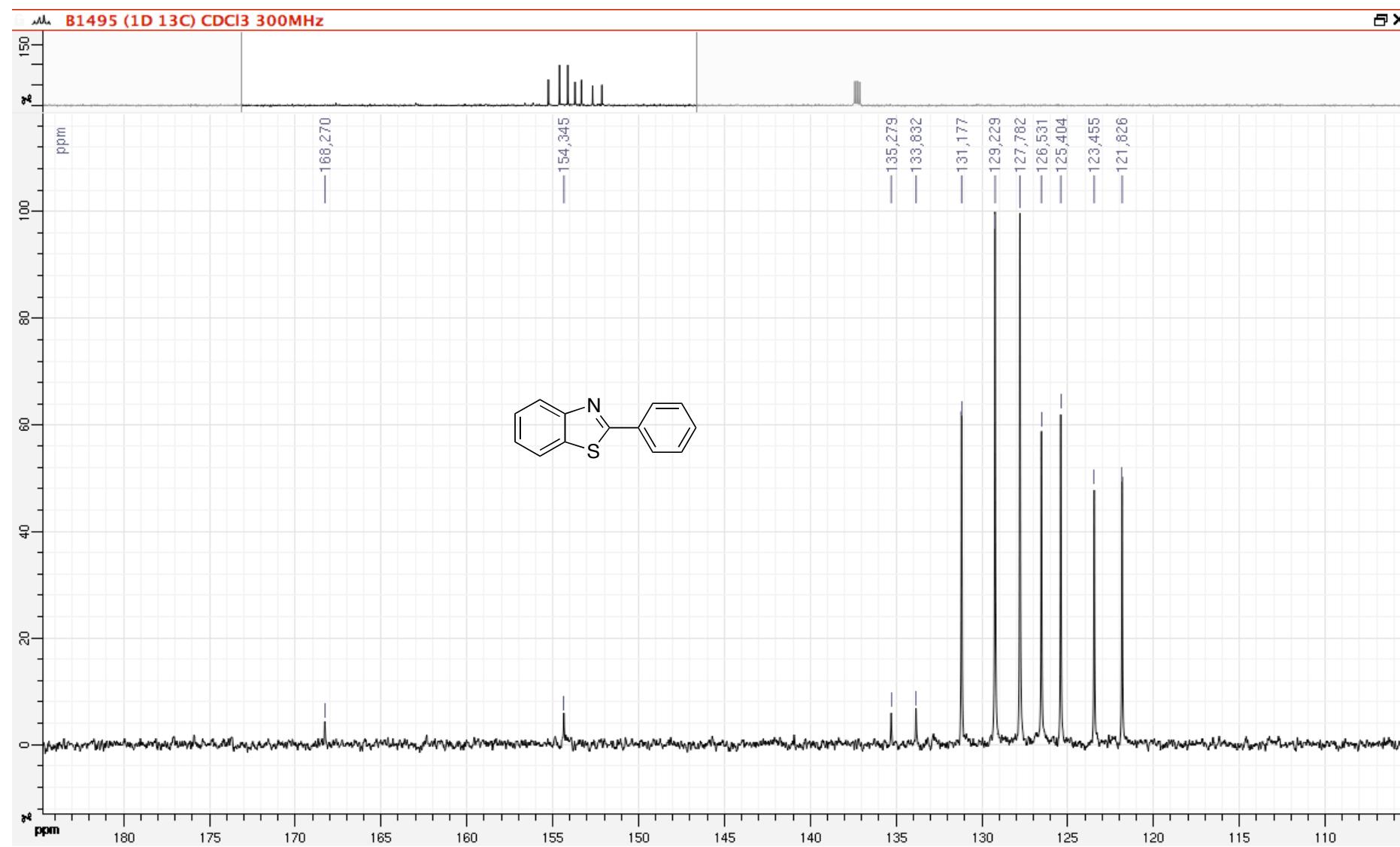
**2-Phenyl-3*H*-imidazo[4,5-*c*]pyridine (4ah)**



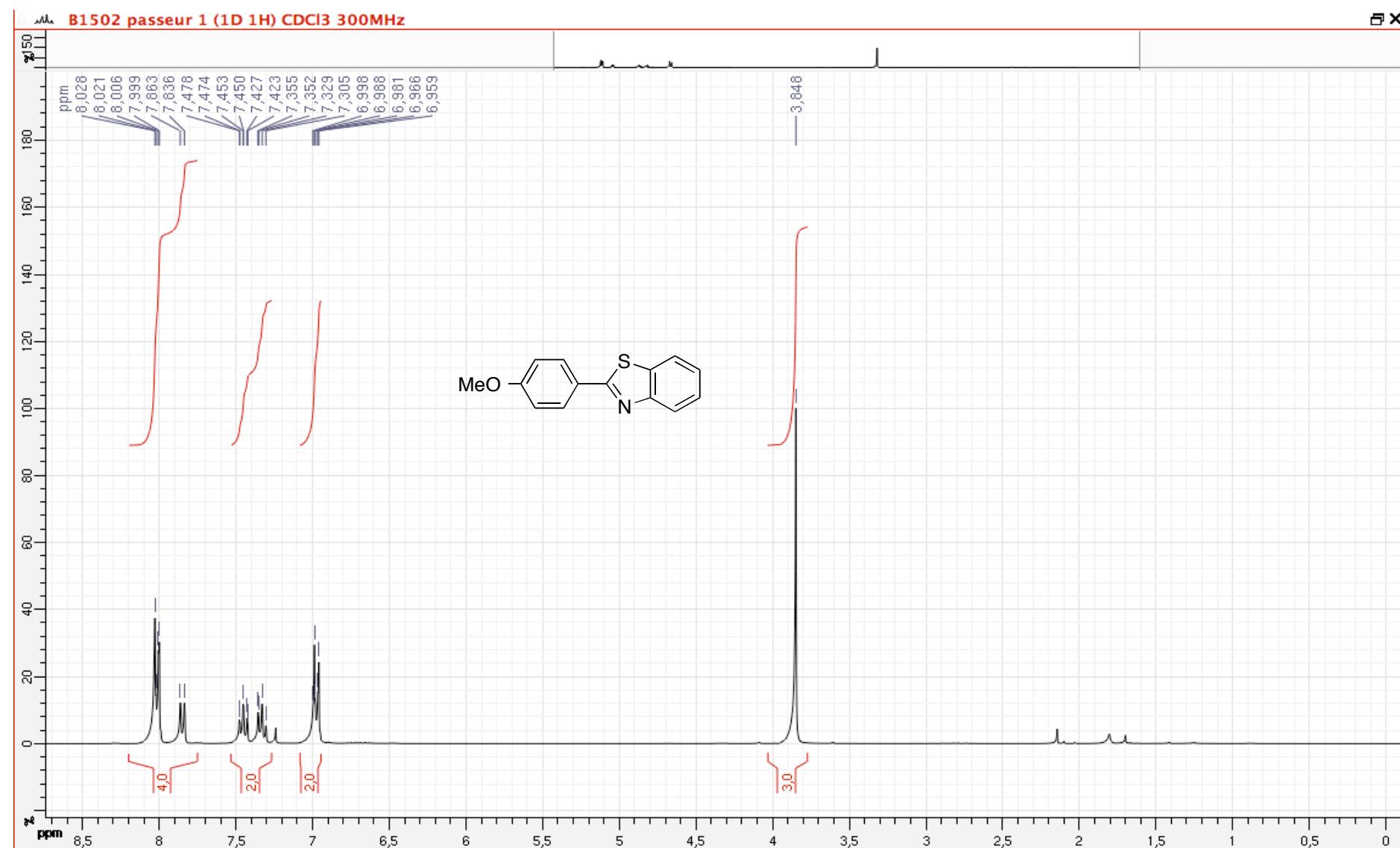


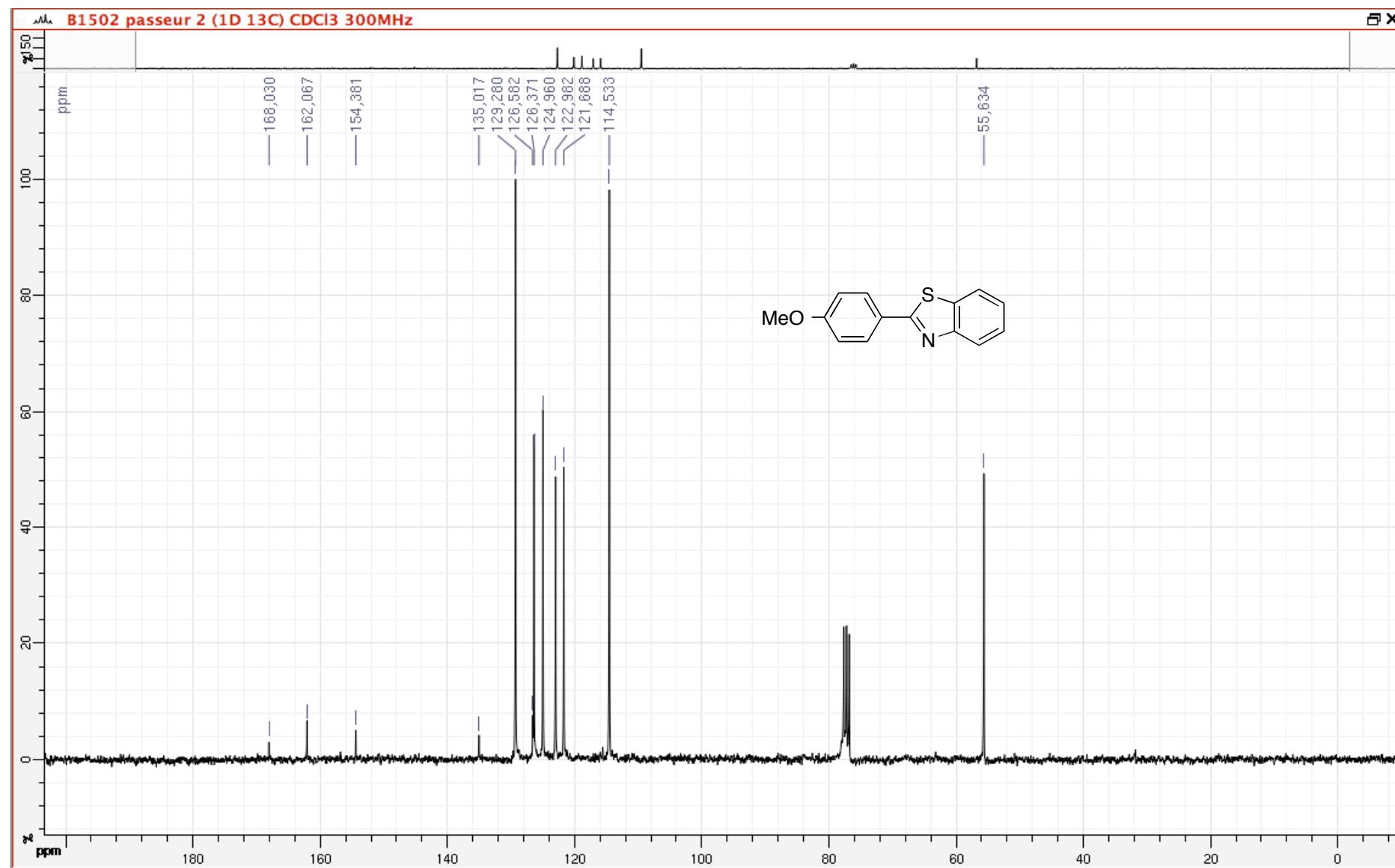
**2-Phenylbenzothiazole (6a)**



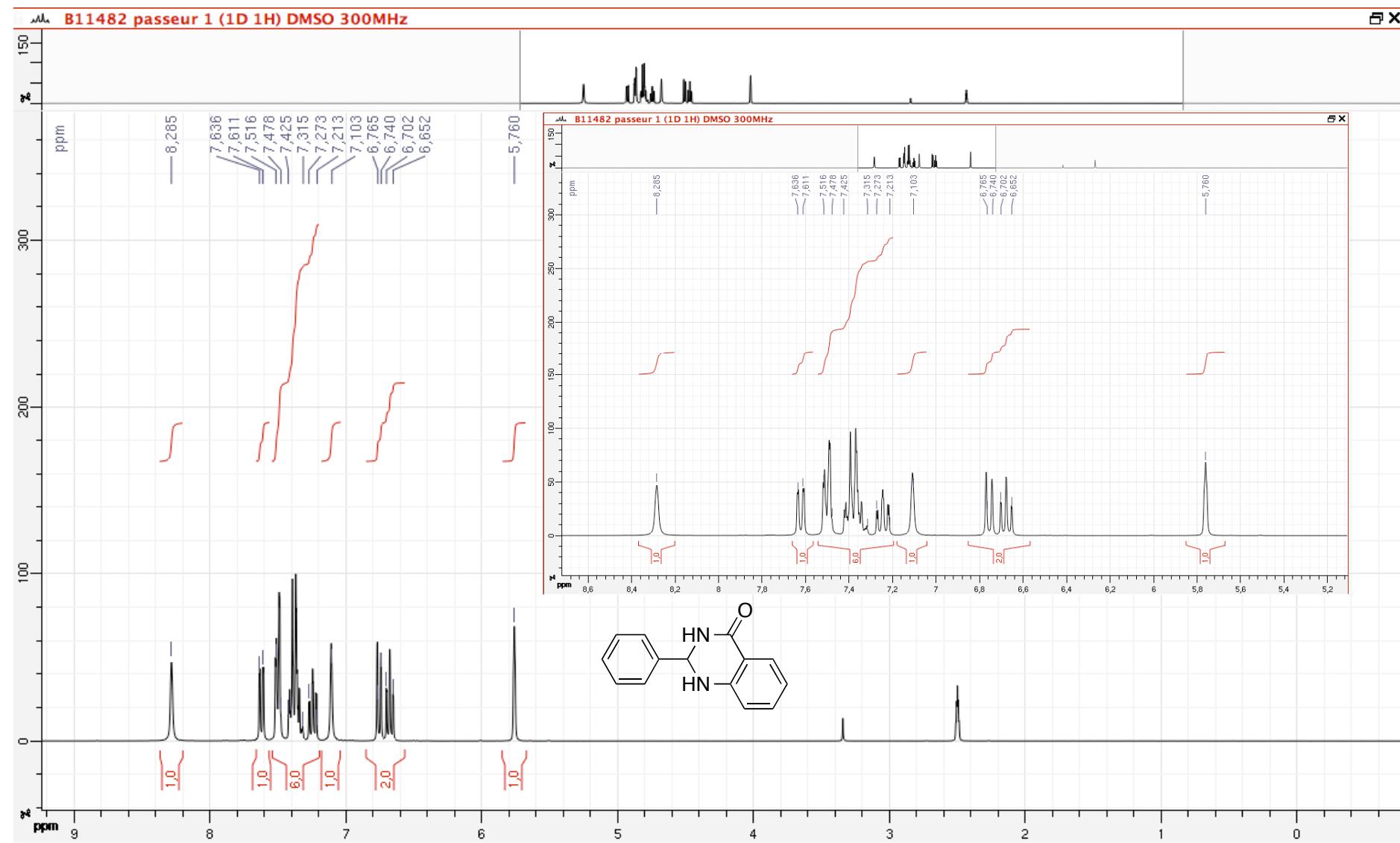


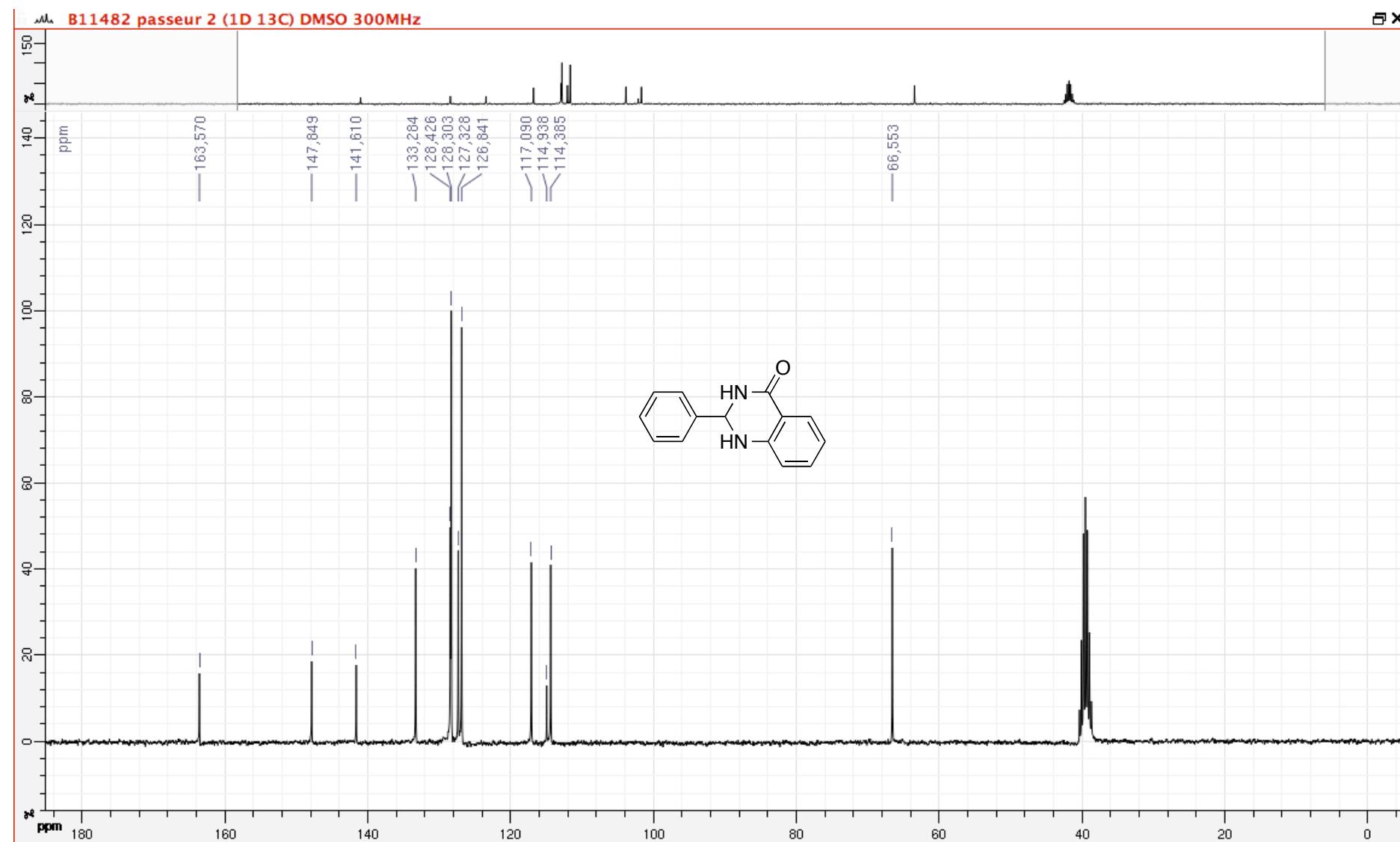
**2-(4-Methoxyphenyl)benzothiazole (6c)**



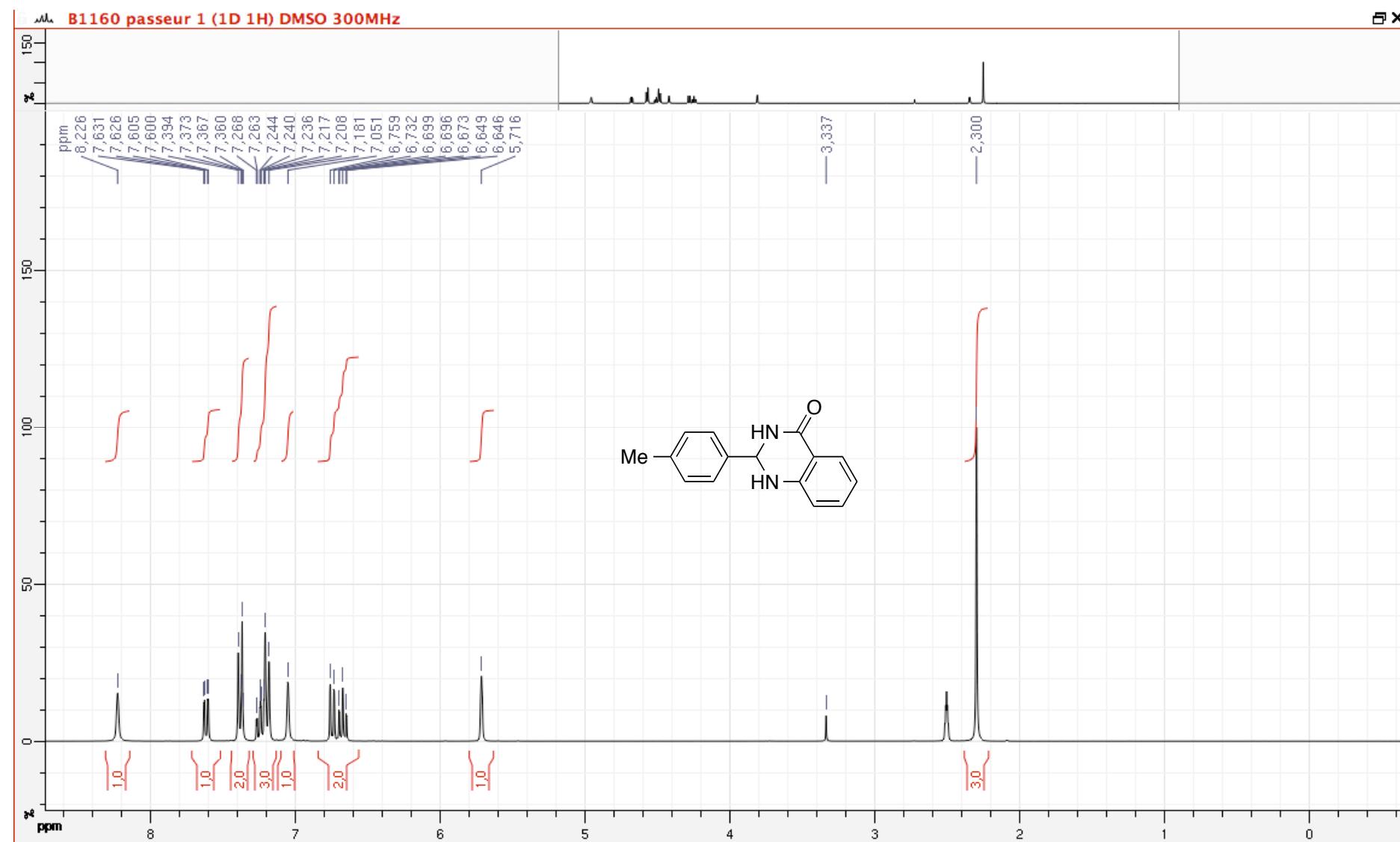


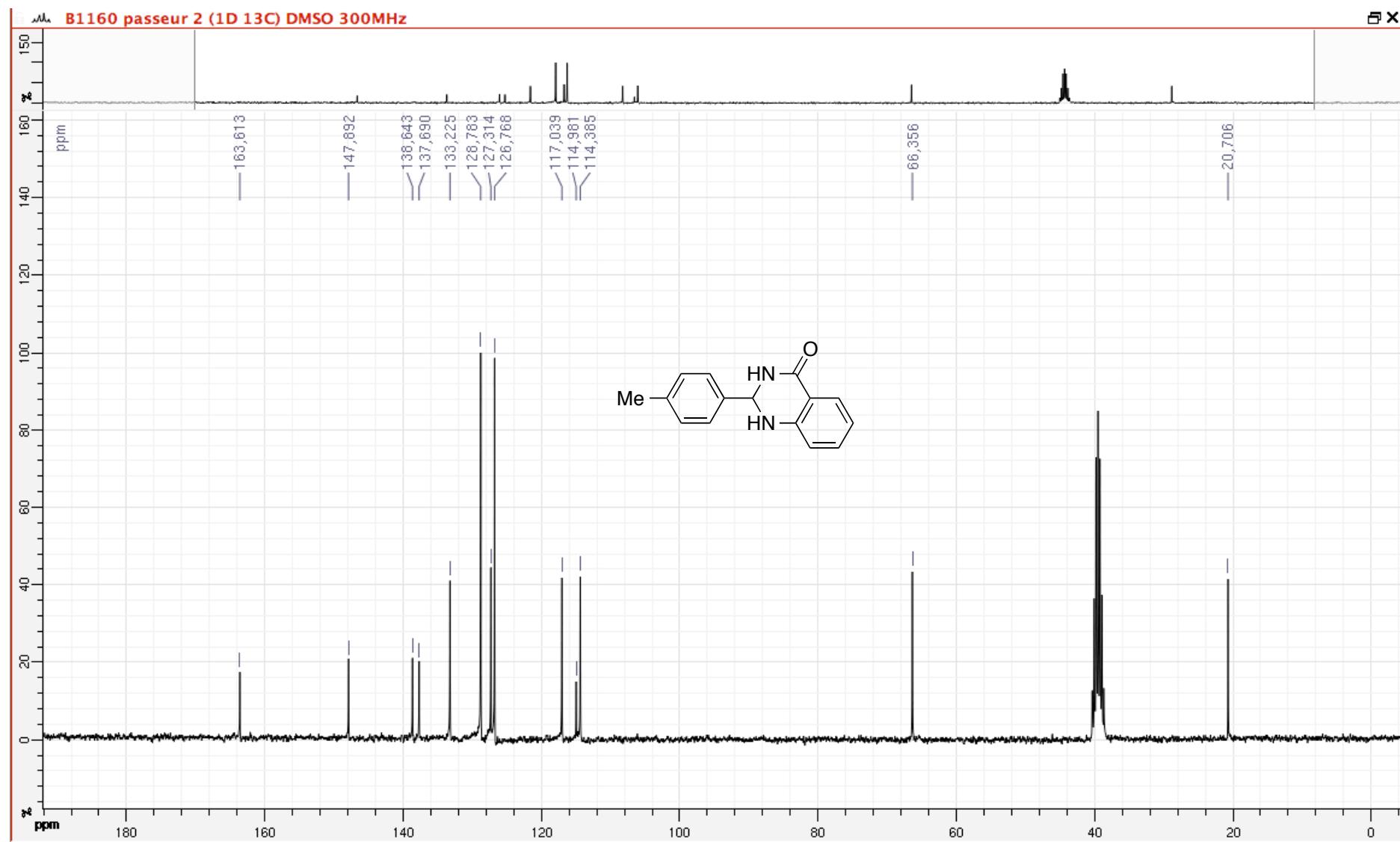
**2-Phenyl-2,3-dihydroquinazolin-4(1H)-one (8a)**



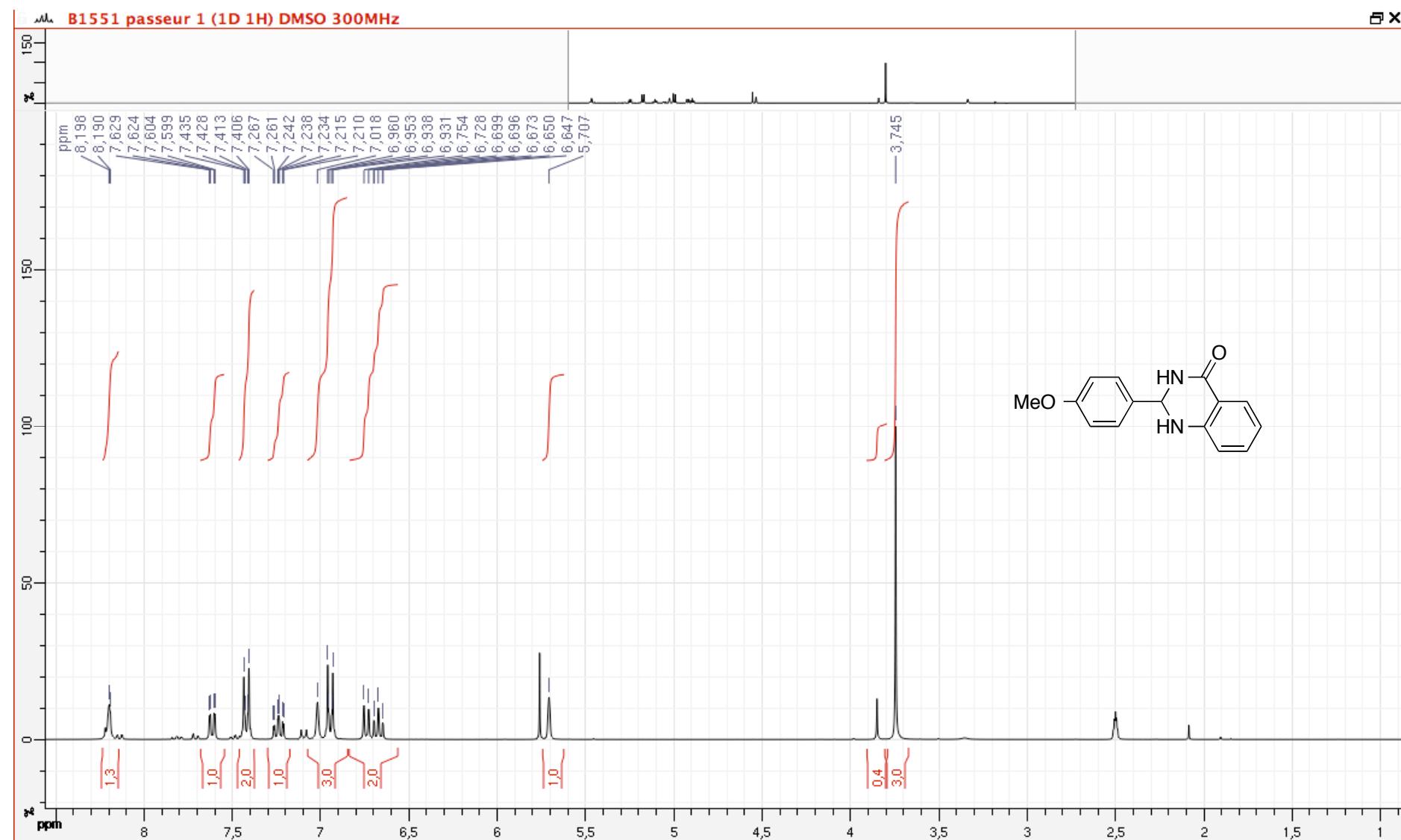


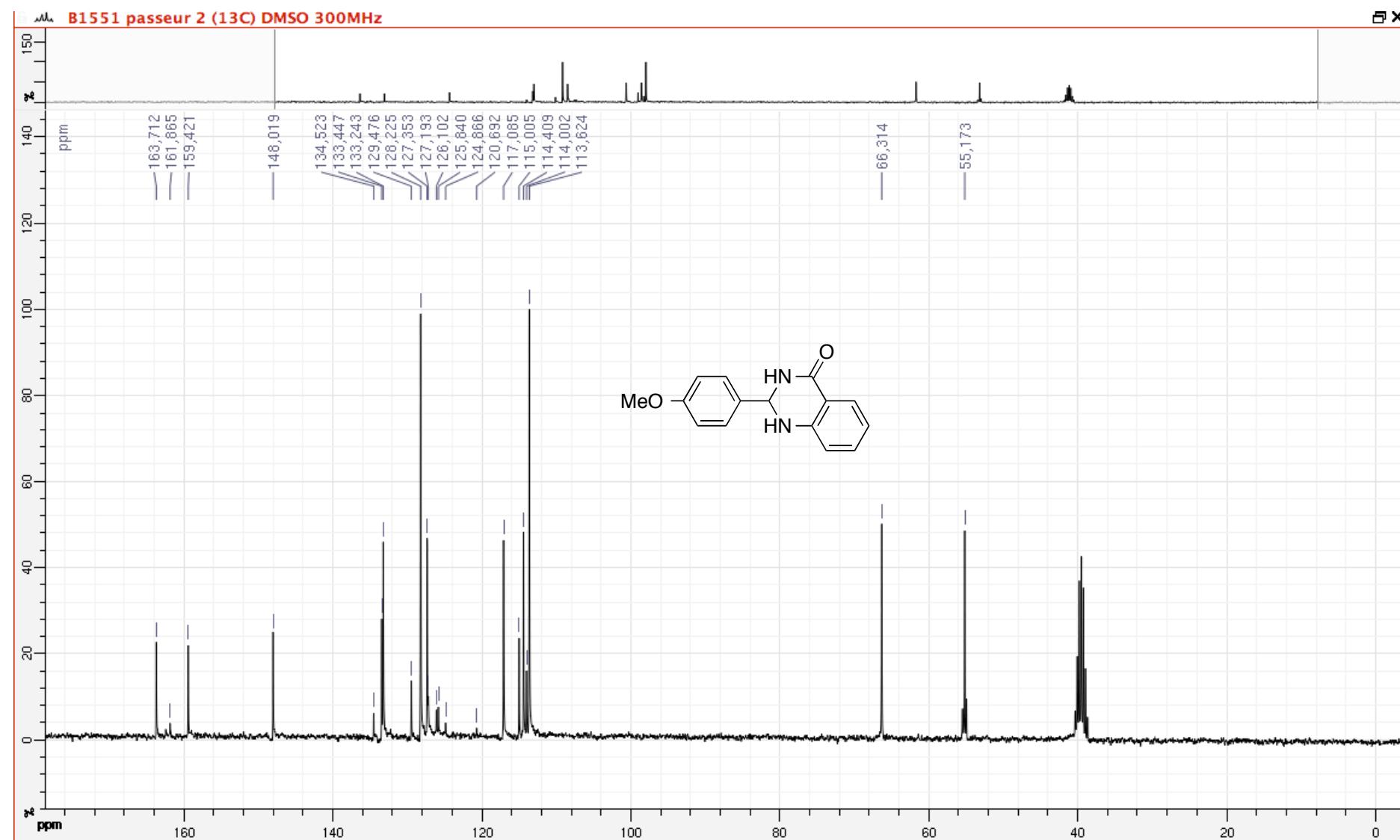
**2-(*p*-Tolyl)-2,3-dihydroquinazolin-4(1*H*)-one (**8b**)**



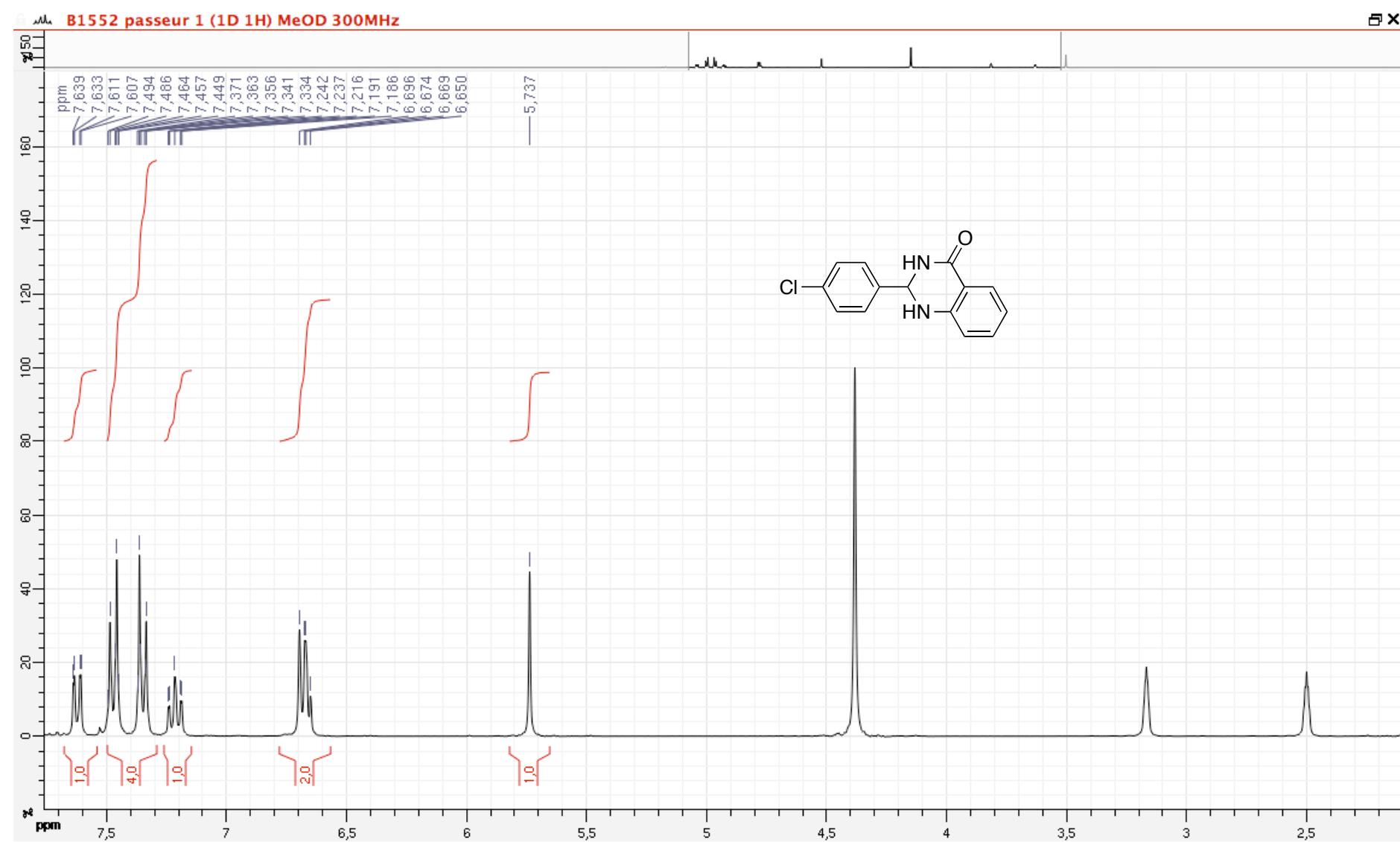


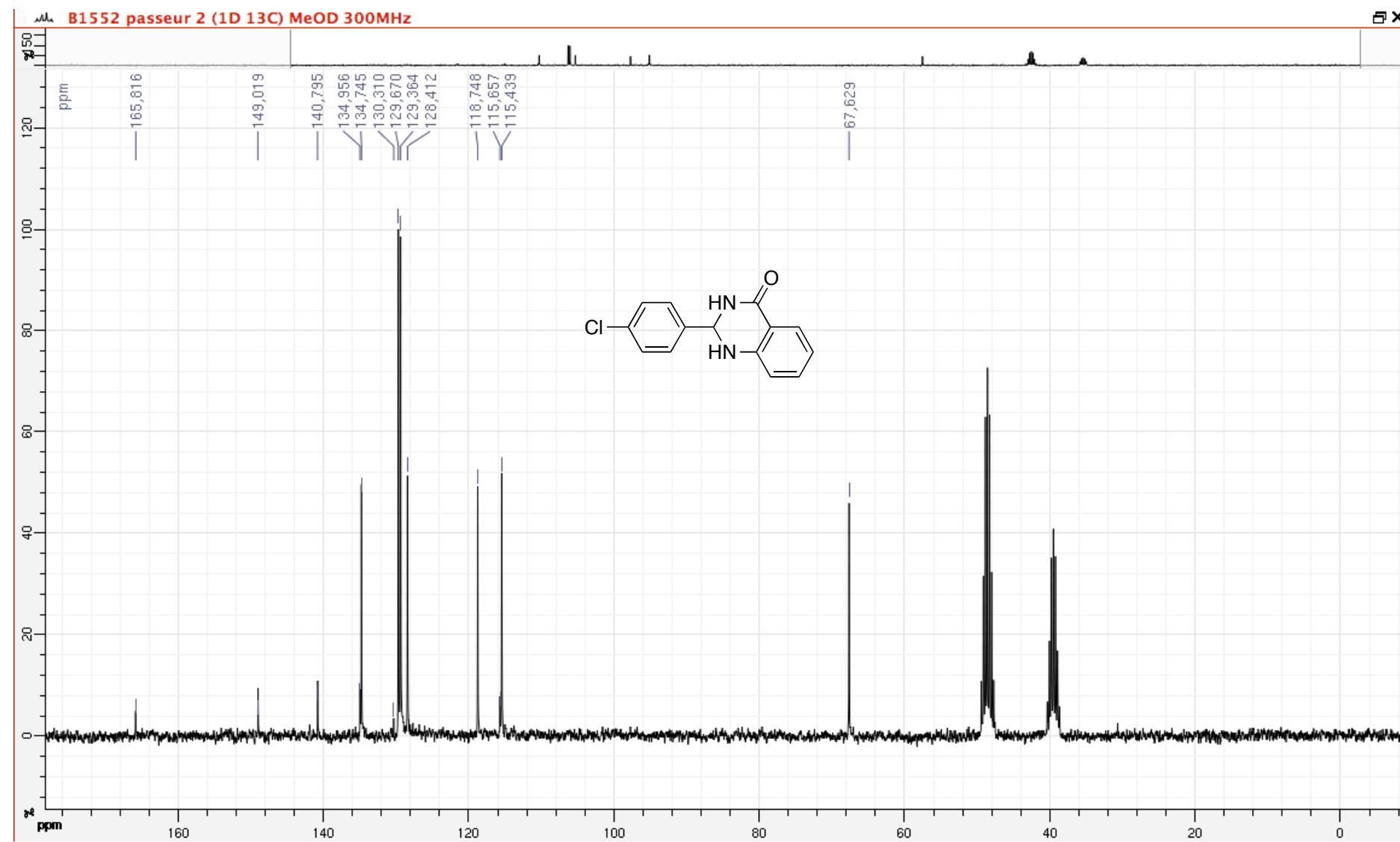
**2-(4-Methoxyphenyl)-2,3-dihydroquinazolin-4(1H)-one (8c)**



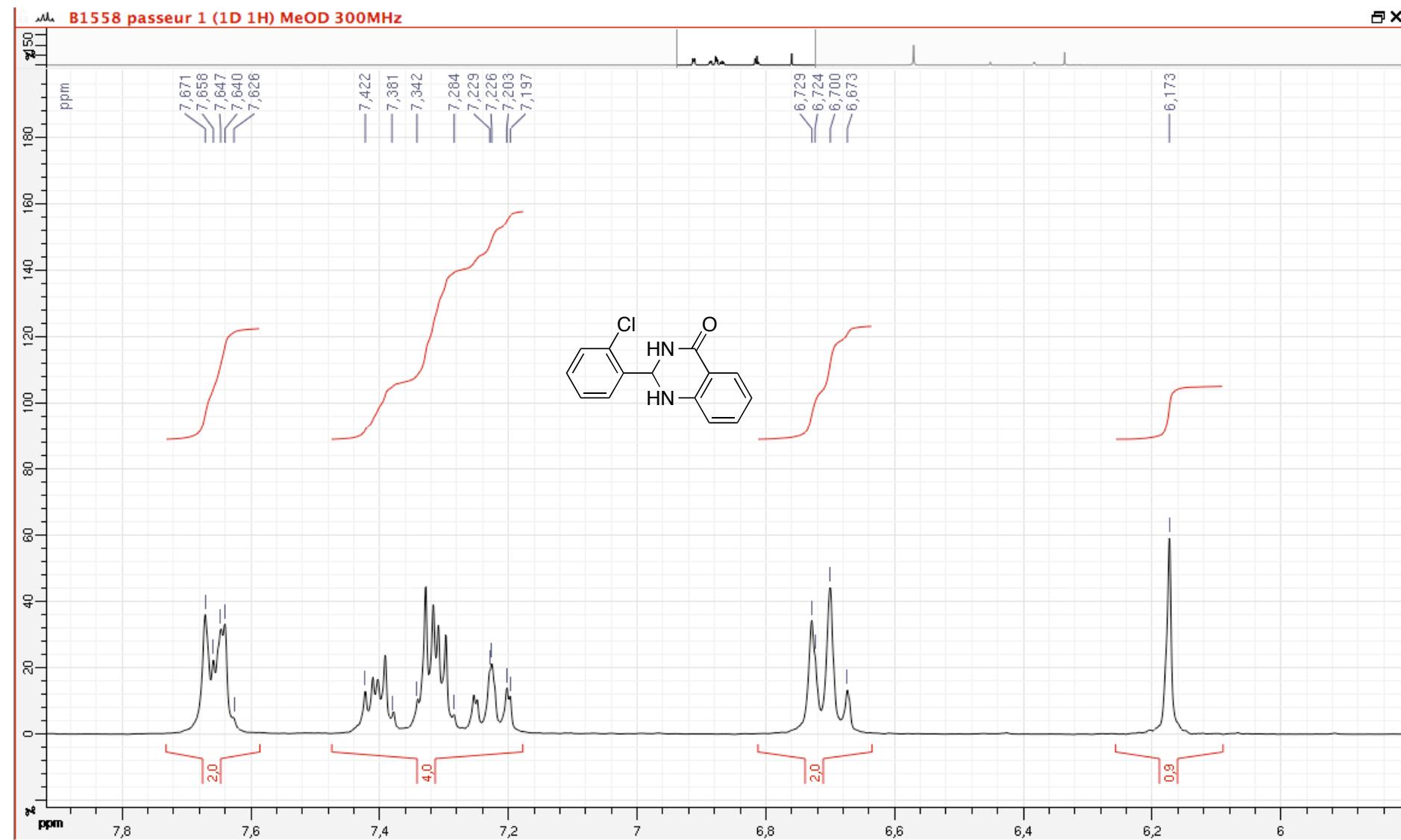


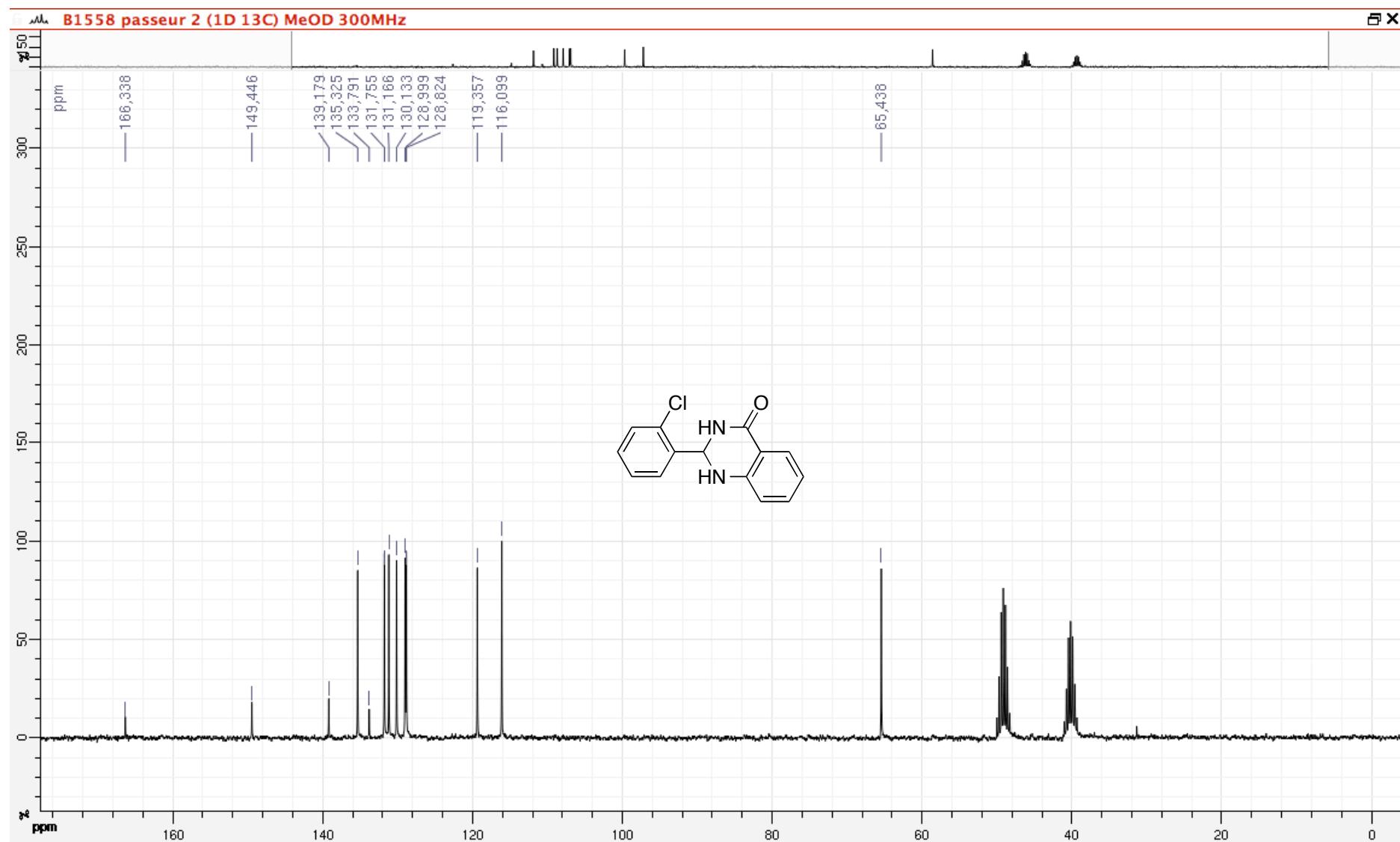
**2-(4-Chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (8d)**



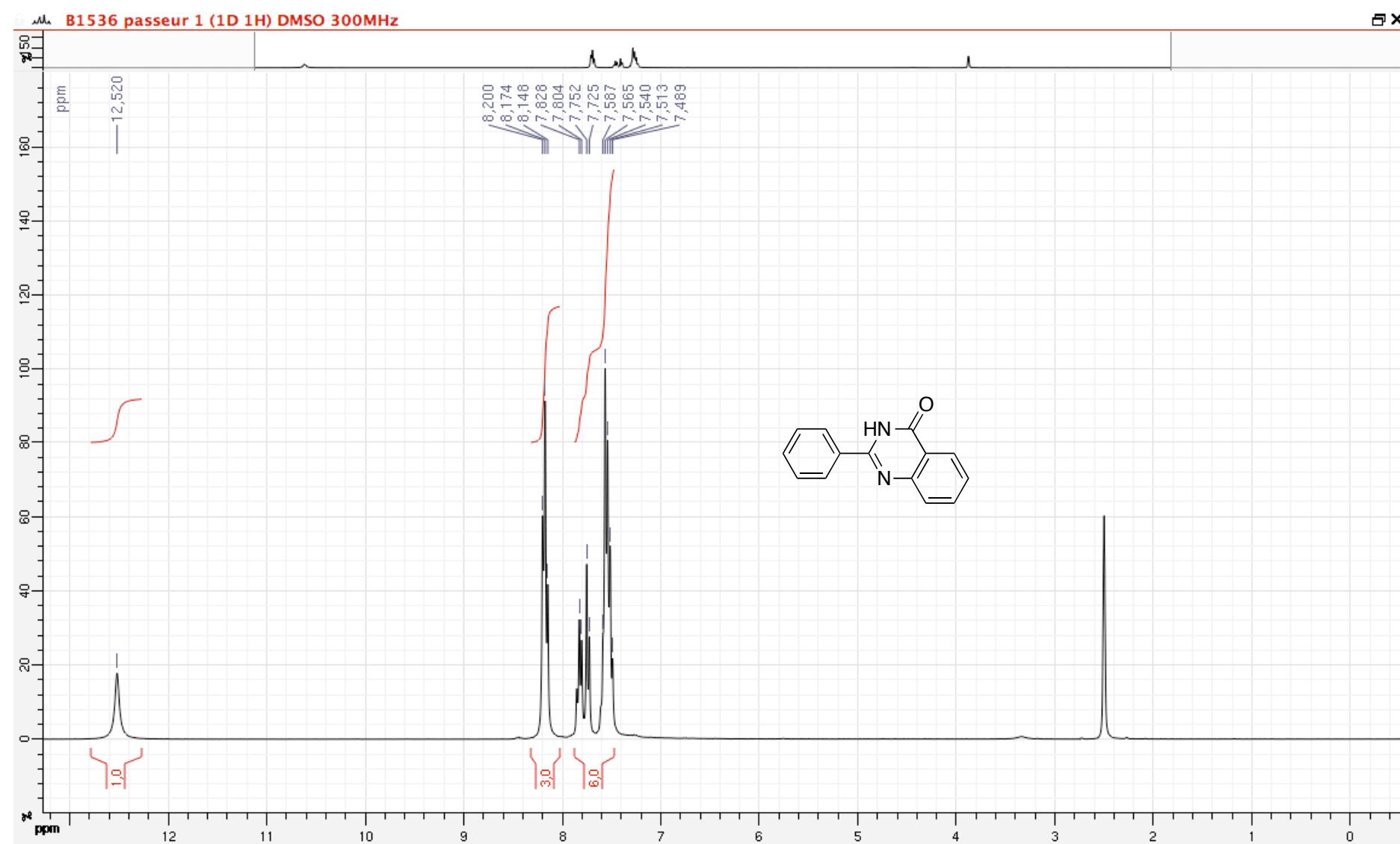


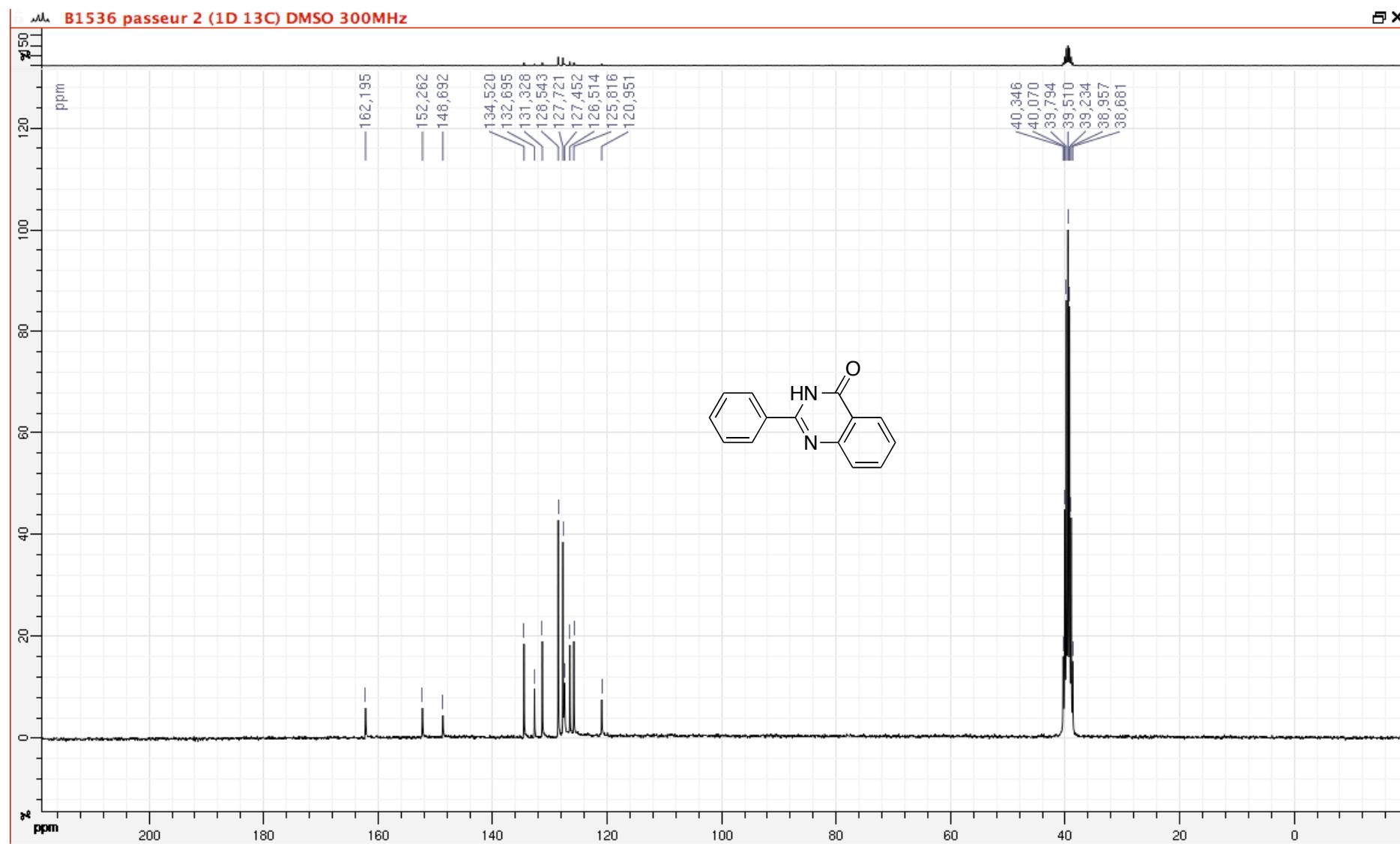
2-(2-Chlorophenyl)-2,3-dihydroquinazolin-4(1H)-one (8e)



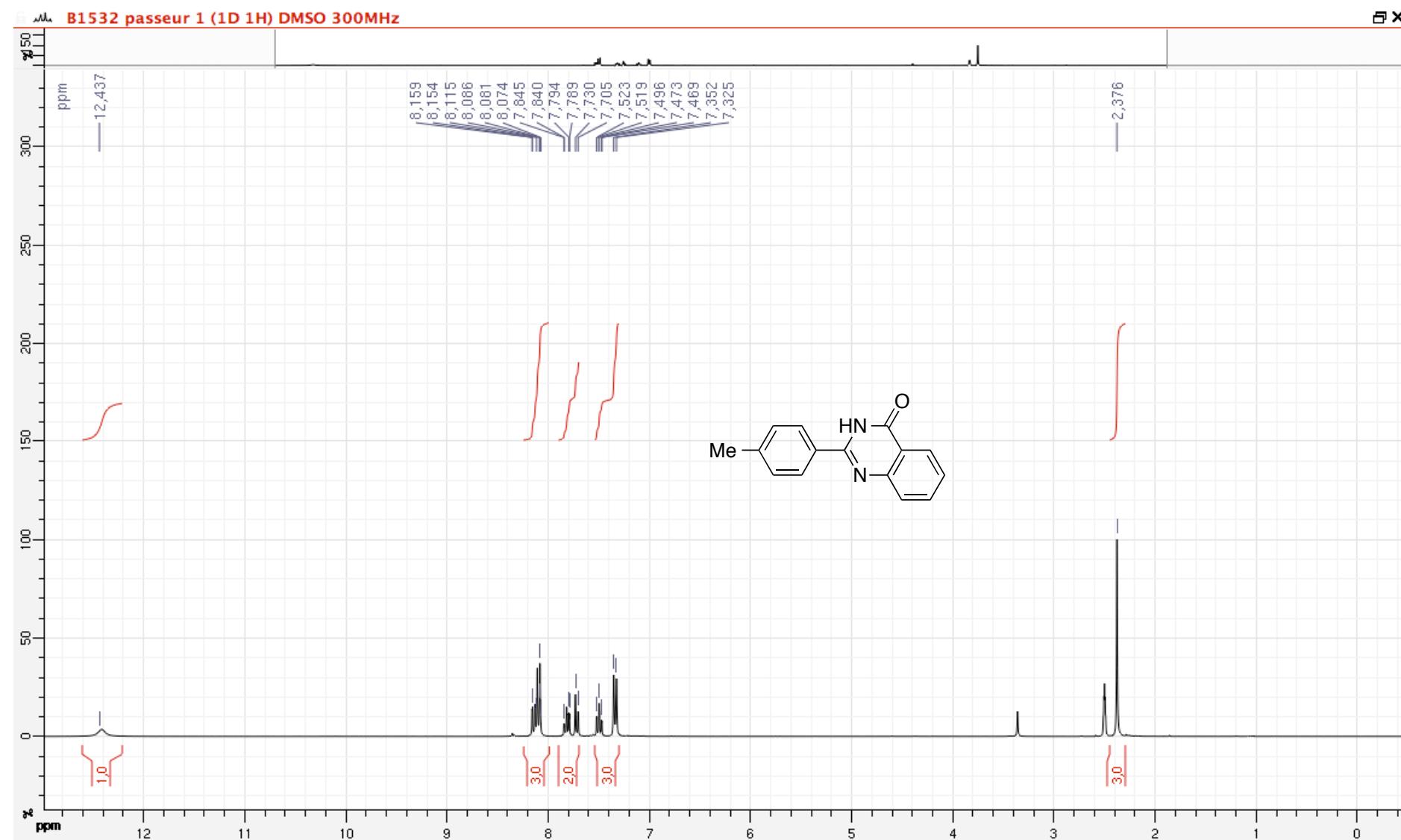


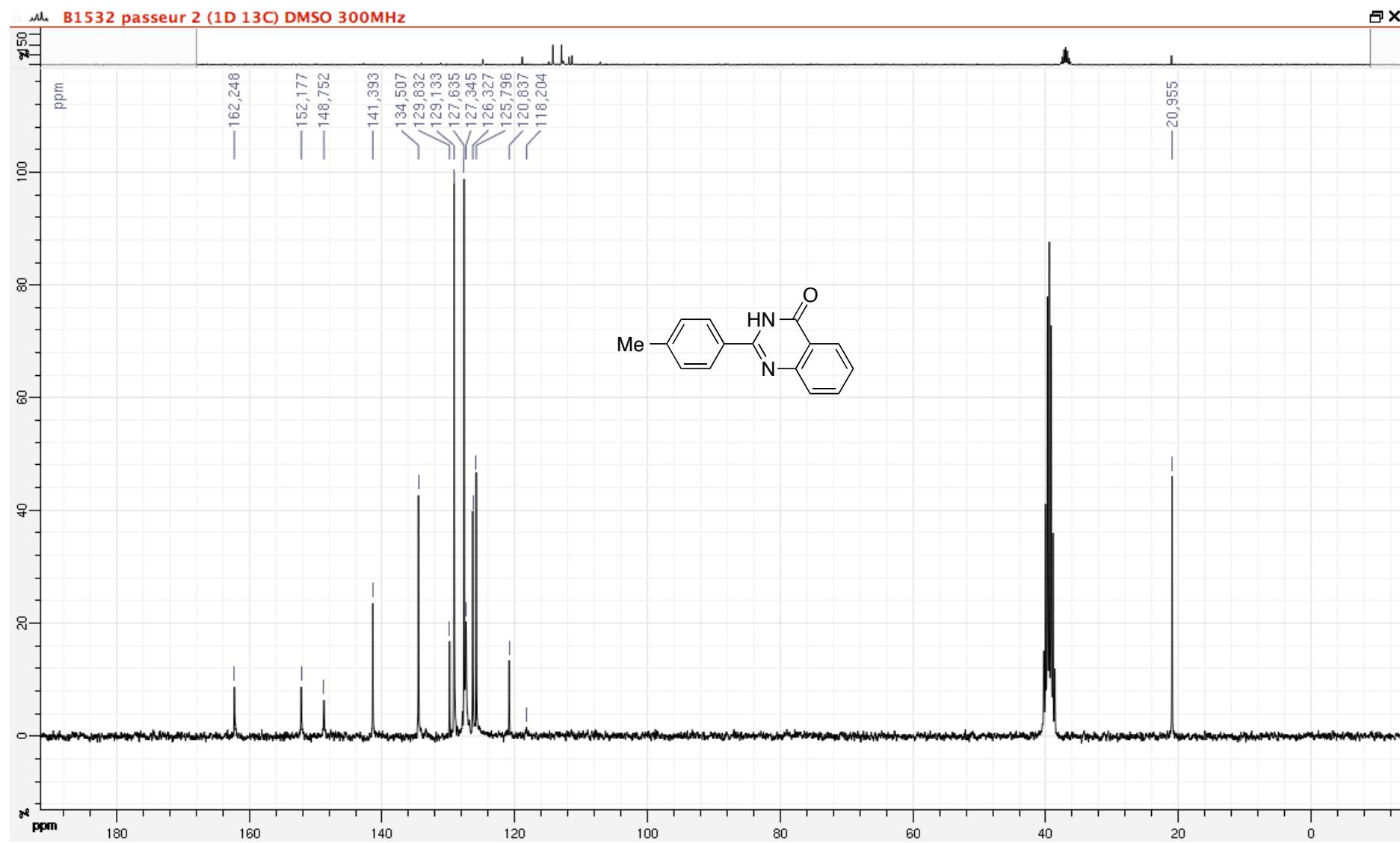
**2-Phenylquinazolin-4(3H)-one (9a)**



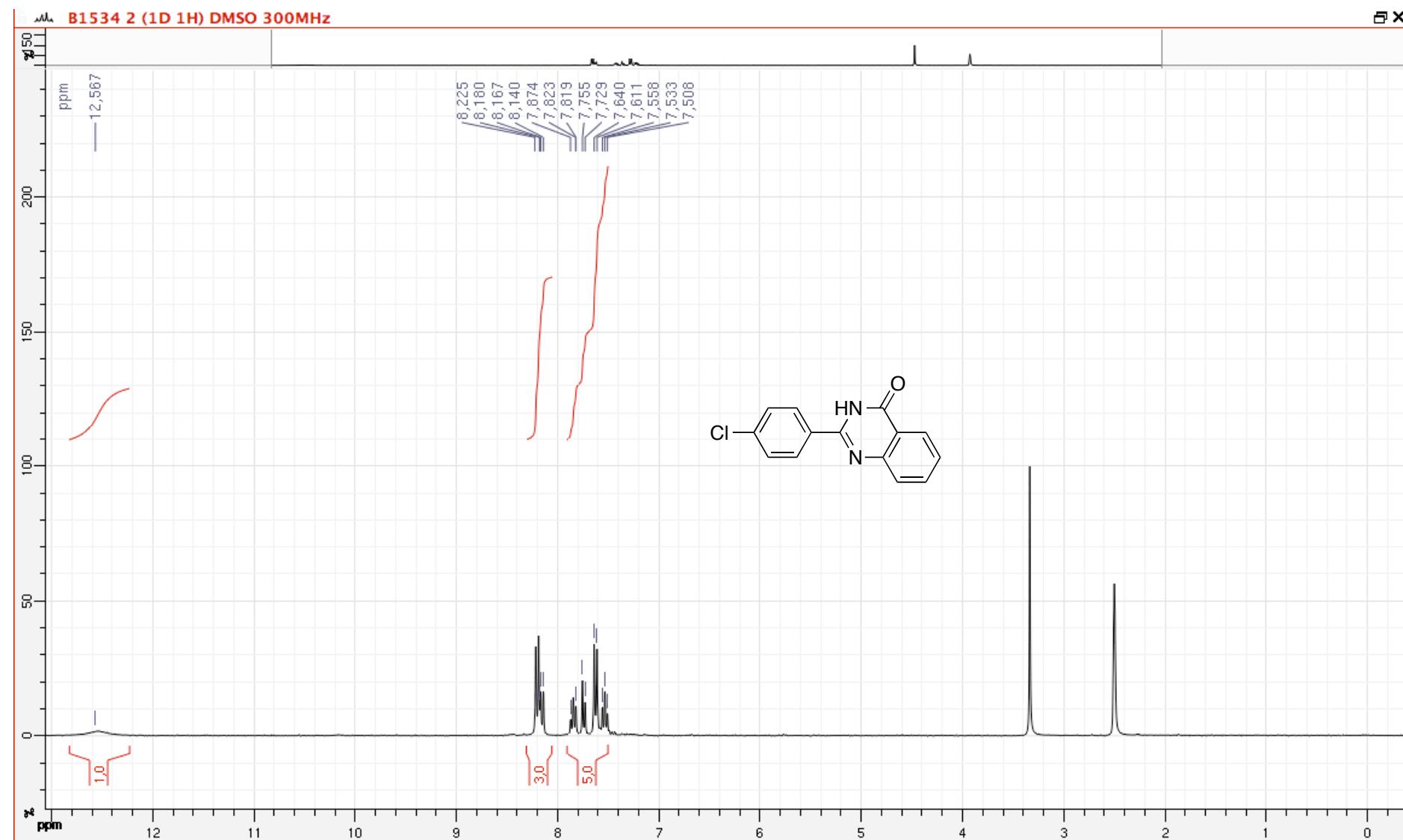


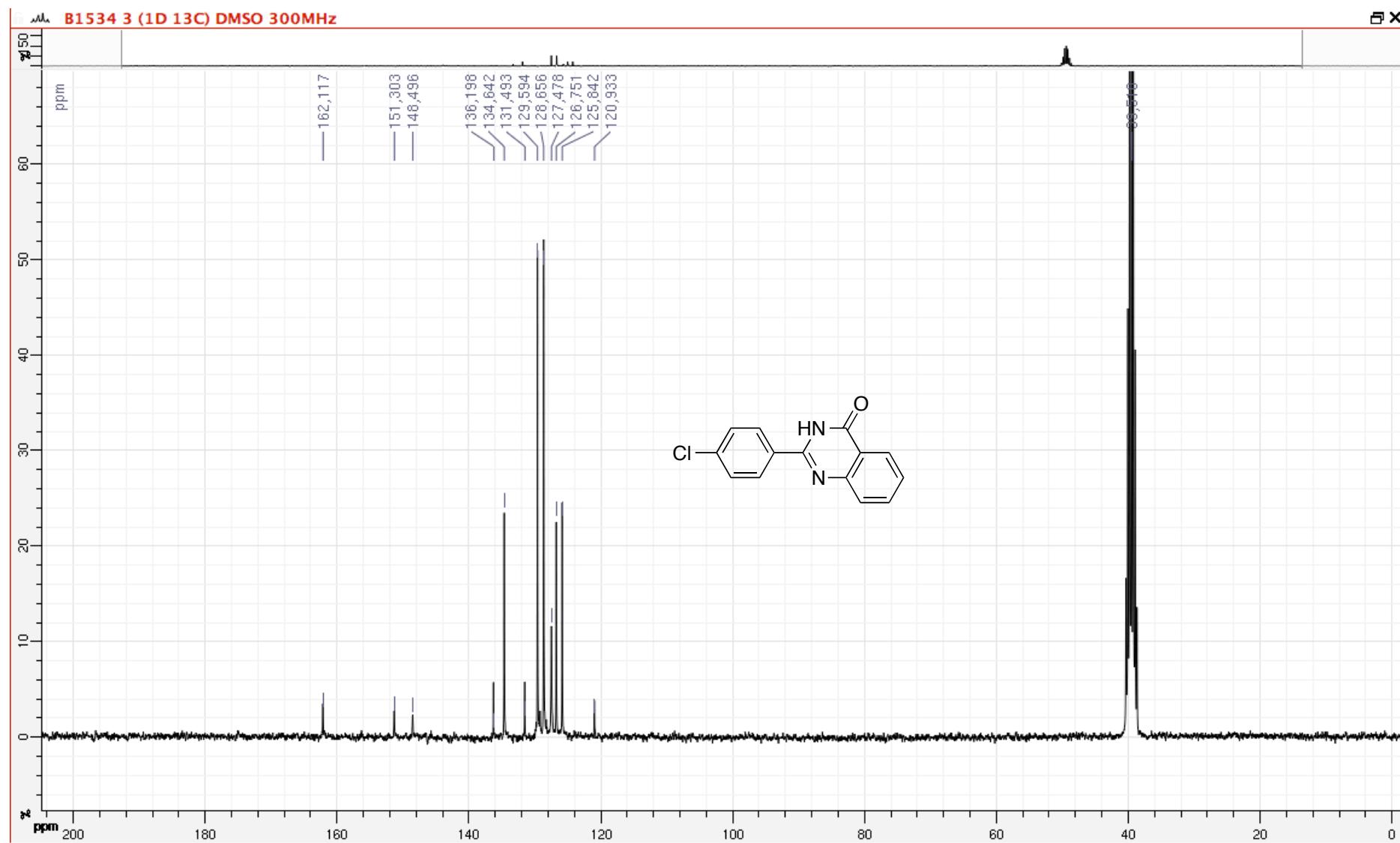
**2-(*p*-Tolyl)quinazolin-4(3*H*)-one (9b)**





**2-(4-Chlorophenyl)quinazolin-4(3H)-one (9d)**





**2-(Pyridin-3-yl)quinazolin-4(3H)-one (9g)**

