

SUPPLEMENTARY MATERIAL TO  
**Synthesis, antioxidant and antimicrobial activity of  
carbohydrazones**

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SPECTRAL DATA FOR COMPOUNDS **1–14**

*[(2-Hydroxyphenyl)methylidene]carbonohydrazide (1)*<sup>1</sup>. White solid (solvent used for crystallization: ethanol). Yield: 66 %; m.p. 180–181°C (lit m.p. –). Anal. Calcd. for C<sub>8</sub>H<sub>9</sub>N<sub>4</sub>O<sub>2</sub> (*Mw* = 245.24 g mol<sup>-1</sup>): C, 49.74; H, 4.70; N, 29.00 %. Found: C, 49.68; H, 4.66; N, 28.93 %. IR (KBr, cm<sup>-1</sup>): 3353 (OH), 3282 (NH<sub>2</sub>), 3096 (NH), 1680 (C=O), 1640 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 4.16 (2H, *s*, H<sub>2</sub>–N<sub>4</sub>), 6.79–6.89 (2H, *m*, H–C<sub>5</sub>, H–C<sub>3</sub>), 7.18 (1H, *td*, H–C<sub>4</sub>, <sup>3</sup>J<sub>4,5</sub> = 7.7 Hz, <sup>4</sup>J<sub>4,6</sub> = 1.4 Hz), 7.64 (1H, *s*, H–C<sub>6</sub>), 7.92 (1H, *s*, H–N<sub>3</sub>), 8.22 (1H, *s*, H–C<sub>7</sub>), 10.08–10.73 (2H, *br.m.ovlp.*, OH, H–N<sub>2</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 116.09 (C<sub>3</sub>), 119.22 (C<sub>5</sub>), 120.06 (C<sub>2</sub>), 127.86 (C<sub>6</sub>), 130.23 (C<sub>4</sub>), 140.22 (C<sub>7</sub>), 156.24 (C<sub>1</sub>), 157.92 (C<sub>8</sub>). lit. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 4.12 (2H, H<sub>2</sub>–N<sub>4</sub>), 6.81 (1H, H–C<sub>5</sub>), 6.84 (1H, H–C<sub>3</sub>), 7.18 (1H, H–C<sub>4</sub>), 7.66 (1H, H–C<sub>6</sub>), 7.90 (1H, H–N<sub>3</sub>), 8.20 (1H, H–C<sub>7</sub>), 10.40 (2H, OH, H–N<sub>2</sub>). <sup>13</sup>C NMR (90 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 116.48 (C<sub>3</sub>), 119.62 (C<sub>5</sub>), 120.49 (C<sub>2</sub>), 128.12 (C<sub>6</sub>), 130.61 (C<sub>4</sub>), 140.05 (C<sub>7</sub>), 156.60 (C<sub>1</sub>), 157.30 (C<sub>8</sub>).

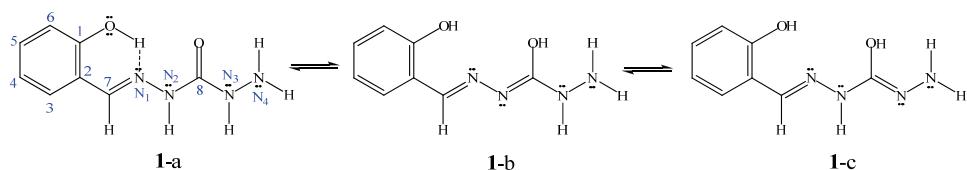


Fig. S-1. Equilibrium of tautomeric forms and geometrical isomers of **1** with numeration of the atom of interest.

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(2-pyridinylmethylidene)carbonohydrazide (**2**).

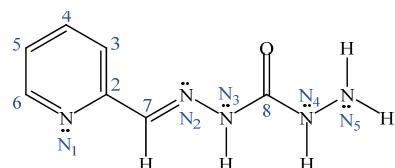


Fig. S-2. (*E*)-isomer of compound **2** with numeration of atom of interest.

White solid (acetonitrile). Yield: 67 %; m.p. 173–174 °C. Anal. Calcd. for  $C_7H_9N_5O$  ( $M_w = 179.18 \text{ g mol}^{-1}$ ): C, 46.92; H, 5.06; N, 39.09 %. Found: C, 46.88; H, 5.01; N, 39.11 %. IR (KBr,  $\text{cm}^{-1}$ ): 3313 (NH<sub>2</sub>), 3208 (NH), 1678 (C=O), 1635 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>,  $\delta$  / ppm): 4.11 (2H, s, H<sub>2</sub>–N<sub>5</sub>), 7.31 (1H, *ddd*, H–C<sub>5</sub>, <sup>3</sup>J<sub>5,4</sub> = 7.5 Hz, <sup>3</sup>J<sub>5,6</sub> = 4.9 Hz, <sup>4</sup>J<sub>5,3</sub> = 1.1 Hz), 7.78 (1H, *td*, H–C<sub>4</sub>, <sup>3</sup>J<sub>4,5</sub> = 7.5 Hz, <sup>4</sup>J<sub>4,6</sub> = 1.5 Hz), 7.89 (1H, s, H–C<sub>7</sub>), 8.10–8.28 (2H, *br.m.ovlp.*, H–C<sub>3</sub>, H–N<sub>4</sub>), 8.51 (1H, *ddd*, H–C<sub>6</sub>, <sup>3</sup>J<sub>6,5</sub> = 4.9 Hz, <sup>4</sup>J<sub>6,4</sub> = 1.5 Hz, <sup>5</sup>J<sub>6,3</sub> = 0.9 Hz), 10.64 (1H, s, H–N<sub>3</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>,  $\delta$  / ppm): 119.85 (C<sub>3</sub>), 123.54 (C<sub>5</sub>), 136.53 (C<sub>4</sub>), 140.59 (C<sub>7</sub>), 149.09 (C<sub>6</sub>), 153.77 (C<sub>2</sub>), 156.85 (C<sub>8</sub>). <sup>15</sup>N NMR (derived from 2D HMBC,  $\delta$  / ppm): 51.10 (N<sub>5</sub>), 99.70 (N<sub>4</sub>), 153.60 (N<sub>3</sub>), 312.20 (N<sub>1</sub>), 326.00 (N<sub>2</sub>).

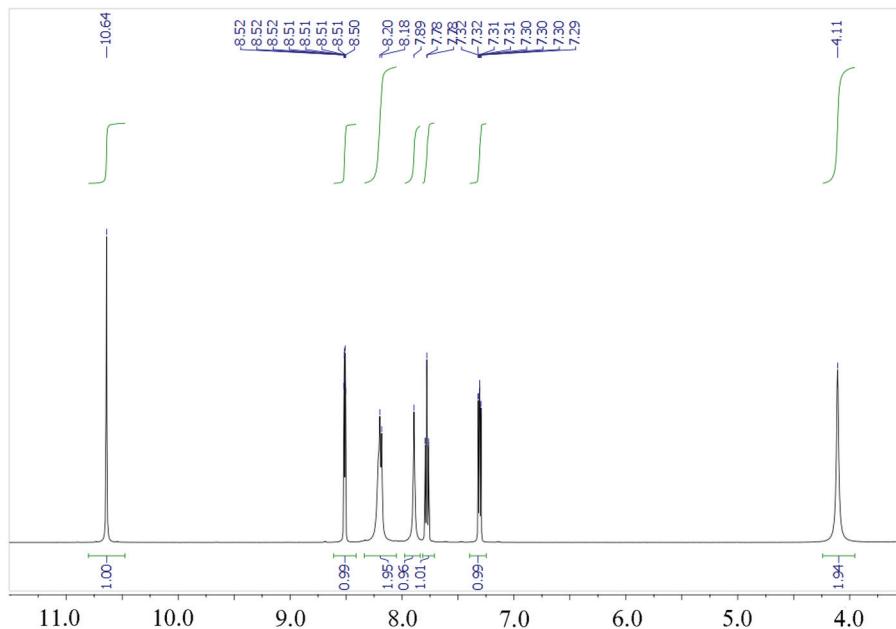
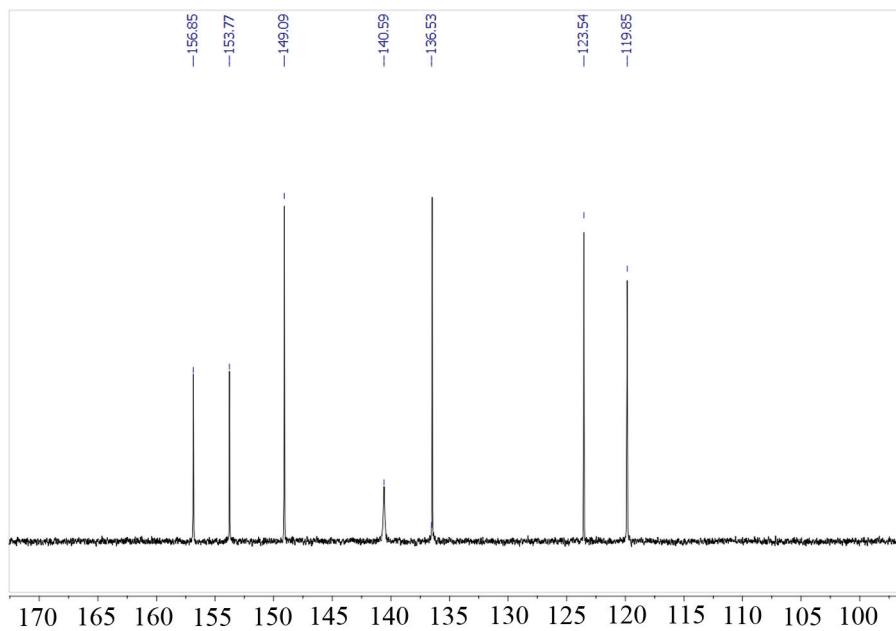
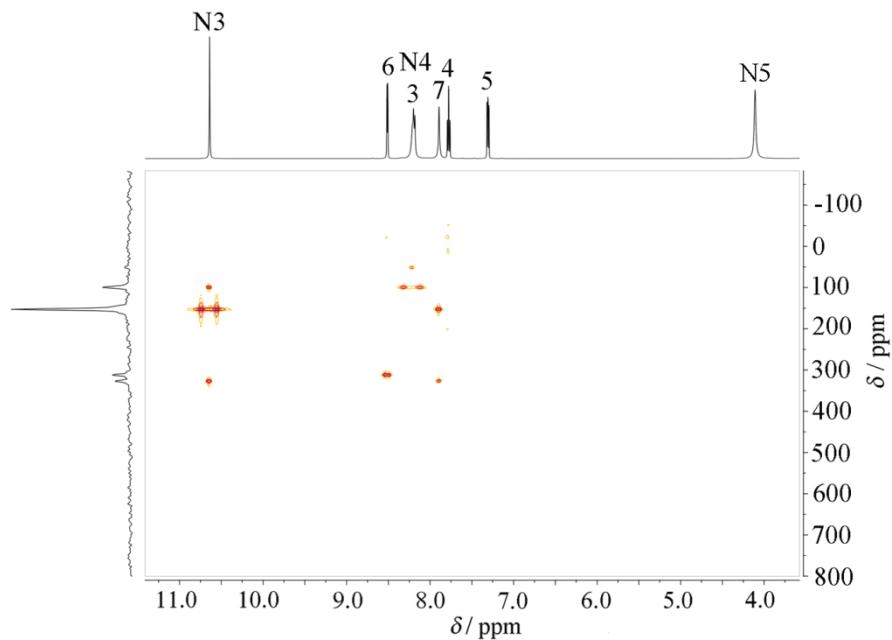


Fig. S-3. <sup>1</sup>H NMR spectrum of **2** in DMSO-*d*<sub>6</sub> recorded at 298 K

Fig. S-4.  $^{13}\text{C}$ -NMR spectrum of **2** in  $\text{DMSO}-d_6$  recorded at 298 KFig. S-5. 2D  $^{15}\text{N}$ -HMBC spectrum of **2** in  $\text{DMSO}-d_6$  recorded at 298 K (x-axis,  $^1\text{H}$ ; y-axis,  $^{15}\text{N}$ -NMR chemical shift).

*[1-(2-Pyridinyl)ethylidene]carbonohydrazide (3)<sup>2</sup>.* White solid (ethanol). Yield: 72.0 %; m.p. 203 °C (lit m.p. 202-203°C). Anal. Calcd. for C<sub>8</sub>H<sub>11</sub>N<sub>5</sub>O (*Mw* = 193.21 g mol<sup>-1</sup>): C, 47.73; H, 5.74; N, 36.25 %. Found: C, 47.61; H, 5.82; N, 36.18 %. IR (KBr, cm<sup>-1</sup>): 3308 (NH<sub>2</sub>), 3197 (NH), 1674 (C=O), 1631 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 2.25 (3H, *s*, H<sub>3</sub>-CH<sub>3</sub>), 4.15 (2H, *s*, H<sub>2</sub>-N<sub>5</sub>), 7.36 (1H, *dd*, H-C<sub>5</sub>, <sup>3</sup>J<sub>5,4</sub> = 6.4 Hz, <sup>3</sup>J<sub>5,6</sub> = 4.6 Hz), 7.78 (1H, *ddd*, H-C<sub>4</sub>, <sup>3</sup>J<sub>4,3</sub> = 8.1 Hz, <sup>3</sup>J<sub>4,5</sub> = 6.4 Hz, <sup>4</sup>J<sub>4,6</sub> = 1.5 Hz), 8.20 (1H, *s*, H-N<sub>4</sub>), 8.41 (1H, *d*, H-C<sub>3</sub>, <sup>3</sup>J<sub>3,4</sub> = 8.1 Hz), 8.54 (1H, *d*, H-C<sub>6</sub>, <sup>3</sup>J<sub>6,5</sub> = 4.6 Hz), 9.78 (1H, *s*, H-N<sub>3</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 11.60 (CH<sub>3</sub>), 120.58 (C<sub>3</sub>), 123.57 (C<sub>5</sub>), 136.27 (C<sub>4</sub>), 145.14 (C<sub>7</sub>), 148.10 (C<sub>6</sub>), 155.01 (C<sub>2</sub>), 157.40 (C<sub>8</sub>). lit. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 2.26 (3H, *s*, CH), 4.14 (2H, *br*, NH), 7.34 (1H, *t*, py), 7.76 (1H, *t*, py), 8.18 (1H, *br*, NH), 8.38 (1H, *d*, py), 8.52 (1H, *d*, py), 9.76 (1H, *br*, NH).

*[Phenyl(2-pyridinyl)methylidene]carbonohydrazide (4).*

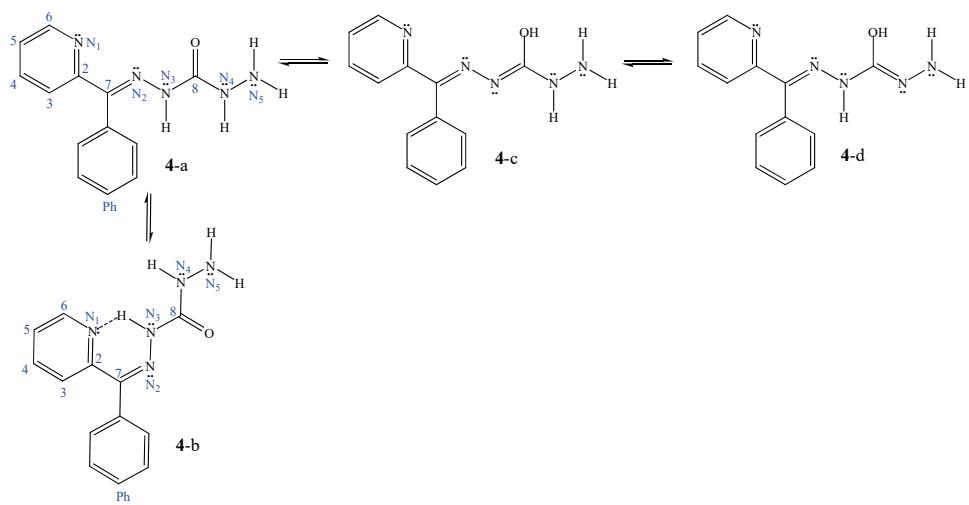
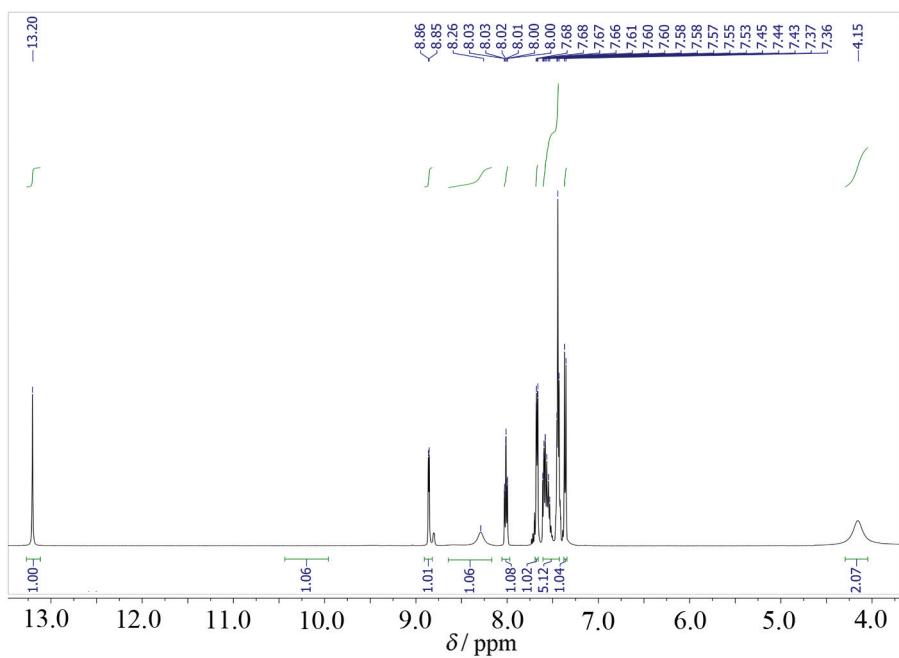
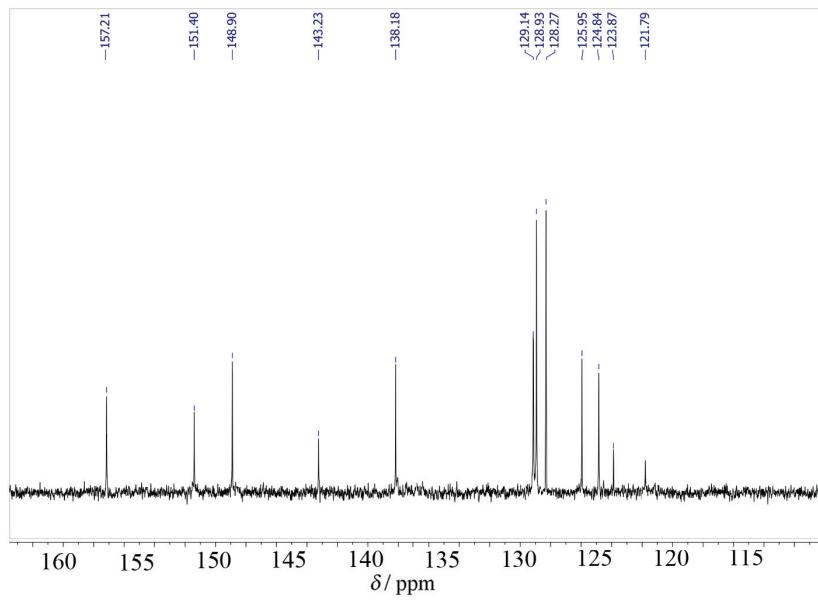


Fig. S-6. Equilibrium of tautomeric forms (4-a, 4-c and 4-d) and geometrical isomers (4-a and 4-b) of compound 4 with numeration of the atom of interest.

White solid (ethanol). Yield: 84 %; m.p. 203-205 °C. Anal. Calcd. for C<sub>13</sub>H<sub>13</sub>N<sub>5</sub>O (*Mw* = 255.11 g mol<sup>-1</sup>): C, 61.17; H, 5.13; N, 27.43 %. Found: C, 61.02; H, 4.98; N, 27.15 %. IR (KBr, cm<sup>-1</sup>): 3304 (NH<sub>2</sub>), 3215 (NH), 1674 (C=O), 1623 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 4.15 (2H, *s*, H<sub>2</sub>-N<sub>5</sub>), 7.36 (1H, *d*, H-C<sub>3</sub>, <sup>3</sup>J<sub>3,4</sub> = 7.8 Hz), 7.40-7.60 (5H, *m*, Ph), 7.68 (1H, *dd*, H-C<sub>4</sub>, <sup>3</sup>J<sub>4,3</sub> = 7.8 Hz, <sup>3</sup>J<sub>4,5</sub> = 7.5 Hz), 8.01 (1H, *dd*, H-C<sub>5</sub>, <sup>3</sup>J<sub>5,4</sub> = 7.5 Hz, <sup>3</sup>J<sub>5,6</sub> = 4.5 Hz), 8.26 (1H, *s*, H-N<sub>4</sub>), 8.85 (1H, *d*, H-C<sub>6</sub>, <sup>3</sup>J<sub>6,5</sub> = 4.5 Hz), 13.20 (1H, *s*, H-N<sub>3</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 121.79 (C<sub>3</sub>), 123.87 (C<sub>5</sub>), 124.84 (Ph), 125.95 (Ph), 128.27 (Ph), 128.93 (Ph), 129.14 (Ph), 138.18 (C<sub>4</sub>), 143.23 (C<sub>7</sub>), 148.90 (C<sub>6</sub>), 151.40 (C<sub>2</sub>), 157.21 (C<sub>8</sub>). <sup>15</sup>N NMR (derived from 2D HMBC, δ / ppm): 57.62 (N<sub>5</sub>), 100.07 (N<sub>4</sub>), 155.1 (N<sub>3</sub>), 306.33 (N<sub>1</sub>), 315.85 (N<sub>2</sub>).

Fig. S-7.  $^1\text{H}$  NMR spectrum of **4** in  $\text{DMSO}-d_6$  recorded at 298 KFig. S-8.  $^{13}\text{C}$  NMR spectrum of **4** in  $\text{DMSO}-d_6$  recorded at 298 K.

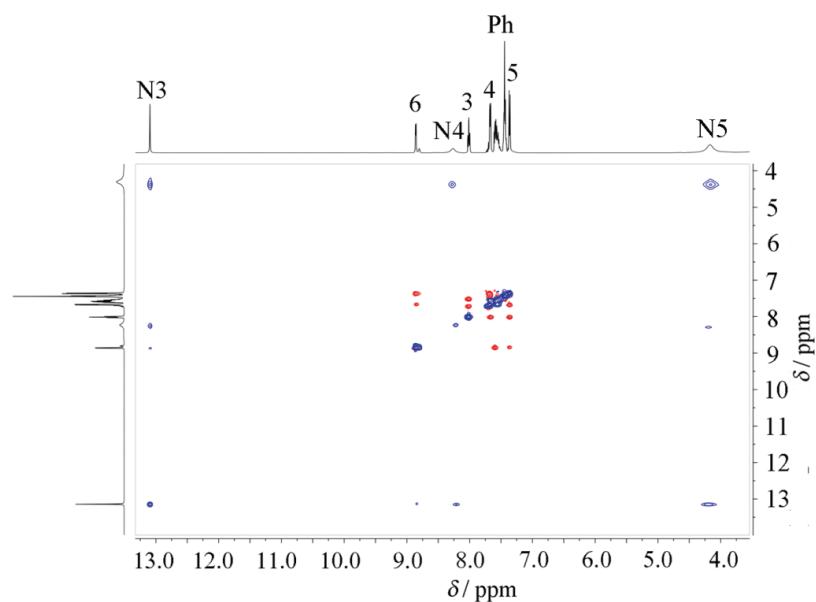


Fig. S-9. 2D NOESY of **4** in  $\text{DMSO}-d_6$  recorded at 298 K (x- and y-axes,  $^1\text{H}$ -NMR chemical shift).

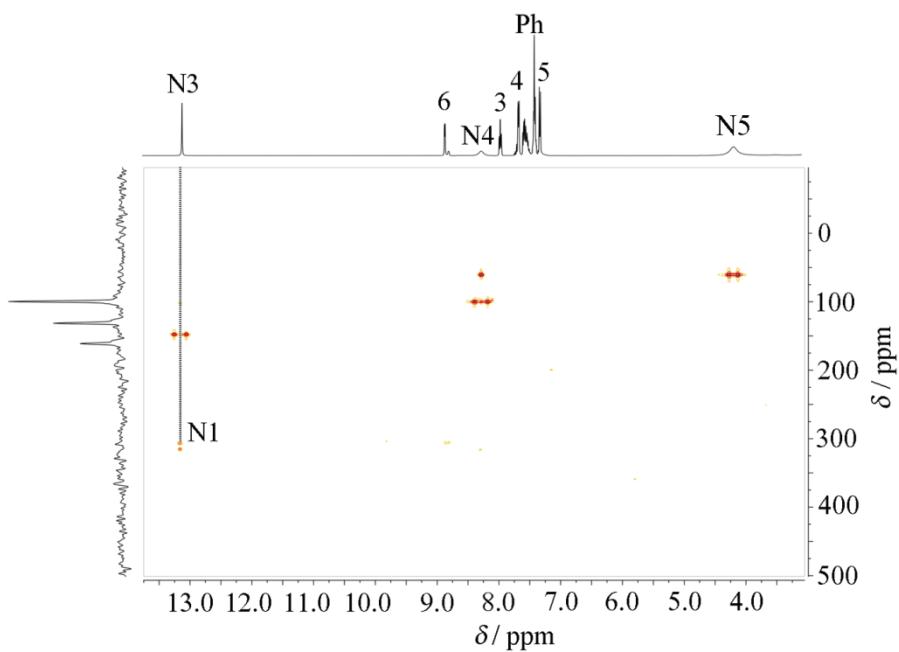


Fig. S-10. 2D  $^{15}\text{N}$ -HMBC of **4** in  $\text{DMSO}-d_6$  at 298 K (x-axis,  $^1\text{H}$ ; y-axis,  $^{15}\text{N}$ -NMR chemical shift).

*(2-Quinolinylmethylidene)carbonohydrazide (5)*<sup>3</sup>. White solid (methanol). Yield: 56 %; m.p. 183 °C (lit m.p. 183°C). Anal. Calcd. for C<sub>11</sub>H<sub>11</sub>N<sub>5</sub>O (*Mw* = 229.24 g mol<sup>-1</sup>): C, 57.63; H, 4.84; N, 30.55 %. Found: C, 57.58; H, 4.62; N, 30.69 %. IR (KBr, cm<sup>-1</sup>): 3297 (NH<sub>2</sub>), 3188 (NH), 1679 (C=O), 1638 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 4.15 (2H, s, H-N<sub>5</sub>), 7.58 (1H, *ddd*, H-C<sub>6</sub>, <sup>3</sup>J<sub>6,7</sub> = 8.2 Hz, <sup>3</sup>J<sub>6,5</sub> = 7.0 Hz, <sup>4</sup>J<sub>6,8</sub> = 1.1 Hz), 7.74 (1H, *ddd*, H-C<sub>7</sub>, <sup>3</sup>J<sub>7,6</sub> = 8.2 Hz, <sup>3</sup>J<sub>7,8</sub> = 6.9 Hz, <sup>4</sup>J<sub>7,5</sub> = 1.4 Hz), 7.93-7.99 (2H, *br.m.ovlp.*, H-C<sub>5</sub>, H-C<sub>8</sub>), 8.03 (1H, s, H-C<sub>9</sub>), 8.27 (1H, *d*, H-C<sub>4</sub>, <sup>3</sup>J<sub>4,3</sub> = 8.4 Hz), 8.34-8.46 (2H, *br.m.ovlp.*, H-C<sub>3</sub>, H-N<sub>4</sub>, <sup>3</sup>J<sub>3,4</sub> = 8.4 Hz), 10.84 (1H, *s*, H-N<sub>3</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 118.03 (C<sub>3</sub>), 126.84 (C<sub>6</sub>), 127.66 (C<sub>4a</sub>), 127.92 (C<sub>5</sub>), 128.69 (C<sub>8</sub>), 129.82 (C<sub>7</sub>), 136.19 (C<sub>4</sub>), 140.64 (C<sub>9</sub>), 147.26 (C<sub>8a</sub>), 154.34 (C<sub>2</sub>), 156.76 (C<sub>10</sub>).

*[(8-Hydroxy-2-quinolinyl)methylidene]carbonohydrazide (6)*<sup>3</sup>. Yellow solid (methanol). Yield: 72 %; m.p. 214-215 °C (lit m.p. 214-215°C). Anal. Calcd. for C<sub>11</sub>H<sub>11</sub>N<sub>5</sub>O<sub>2</sub> (*Mw* = 245.24 g mol<sup>-1</sup>): C, 53.83; H, 4.525; N, 28.56 %. Found: C, 53.66; H, 4.68; N, 28.74 %. IR (KBr, cm<sup>-1</sup>): 3371 (OH), 3335 (NH<sub>2</sub>), 3198 (NH), 1696 (C=O), 1600 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 4.14 (2H, s, H-N<sub>5</sub>), 7.08 (1H, *dd*, H-C<sub>7</sub>, <sup>3</sup>J<sub>7,6</sub> = 7.4 Hz, <sup>4</sup>J<sub>7,5</sub> = 1.4 Hz), 7.36 (1H, *dd*, H-C<sub>5</sub>, <sup>3</sup>J<sub>5,6</sub> = 8.1 Hz, <sup>4</sup>J<sub>5,7</sub> = 1.4 Hz), 7.41 (1H, *m*, H-C<sub>6</sub>), 8.09 (1H, s, H-C<sub>9</sub>), 8.24 (1H, *d*, H-C<sub>4</sub>, <sup>3</sup>J<sub>4,3</sub> = 8.6 Hz), 8.30-8.50 (2H, *br.m.ovlp.*, H-C<sub>3</sub>, H-N<sub>4</sub>), 9.71 (1H, s, OH), 10.88 (1H, *s*, H-N<sub>3</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 111.59 (C<sub>7</sub>), 117.74 (C<sub>5</sub>), 118.35 (C<sub>3</sub>), 127.73 (C<sub>6</sub>), 128.52 (C<sub>4a</sub>), 136.06 (C<sub>4</sub>), 137.93 (C<sub>8a</sub>), 140.50 (C<sub>9</sub>), 152.25 (C<sub>2</sub>), 153.24 (C<sub>8</sub>), 156.83 (C<sub>10</sub>).

*(8-Quinolinylmethylidene)carbonohydrazide (7)*<sup>3</sup>. Yellow solid (methanol). Yield: 64 %; m.p. 185 °C (lit m.p. 185°C). Anal. Calcd. for C<sub>11</sub>H<sub>11</sub>N<sub>5</sub>O (*Mw* = 229.24 g mol<sup>-1</sup>): C, 57.63; H, 4.84; N, 30.55 %. Found: C, 57.71; H, 4.78; N, 30.62 %. IR (KBr, cm<sup>-1</sup>): 3316 (NH<sub>2</sub>), 3200 (NH), 1681 (C=O), 1621 (C=N). <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 4.12 (2H, s, H-N<sub>5</sub>), 7.57 (1H, *dd*, H-C<sub>3</sub>, <sup>3</sup>J<sub>3,4</sub> = 8.3 Hz, <sup>3</sup>J<sub>3,2</sub> = 4.1 Hz), 7.63 (1H, *t*, H-C<sub>6</sub>, <sup>3</sup>J<sub>6,5</sub> = 7.7 Hz), 7.98 (1H, *dd*, H-C<sub>5</sub>, <sup>3</sup>J<sub>5,6</sub> = 7.7 Hz, <sup>4</sup>J<sub>5,7</sub> = 1 Hz), 8.16 (1H, *s*, H-N<sub>4</sub>), 8.39 (1H, *dd*, H-C<sub>4</sub>, <sup>3</sup>J<sub>4,3</sub> = 8.3 Hz, <sup>4</sup>J<sub>4,2</sub> = 2.0 Hz), 8.58 (1H, *d*, H-C<sub>7</sub>, <sup>3</sup>J<sub>7,6</sub> = 7.4 Hz), 8.94 (1H, *dd*, H-C<sub>2</sub>, <sup>3</sup>J<sub>2,3</sub> = 4.1 Hz, <sup>3</sup>J<sub>2,4</sub> = 2.0 Hz), 9.14 (1H, *s*, H-C<sub>9</sub>), 10.65 (1H, *s*, H-N<sub>3</sub>). <sup>13</sup>C NMR (126 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 121.67 (C<sub>3</sub>), 125.61 (C<sub>7</sub>), 126.45 (C<sub>6</sub>), 127.94 (C<sub>4a</sub>), 128.90 (C<sub>5</sub>), 131.59 (C<sub>8</sub>), 136.55 (C<sub>4</sub>), 136.89 (C<sub>9</sub>), 145.01 (C<sub>8a</sub>), 150.08 (C<sub>2</sub>), 157.21 (C<sub>10</sub>).

*1,5-Bis[2-hydroxyphenyl)methylidene]carbonohydrazide (8)*<sup>4</sup>. Yellow cristal (ethanol). Yield: 78 %; m.p. 219°C (lit m.p. 216°C); Anal. Calcd. for C<sub>15</sub>H<sub>14</sub>N<sub>4</sub>O<sub>3</sub> (*Mw* = 298.11 g mol<sup>-1</sup>): C, 60.35; H, 4.74; N, 18.79 %, Found: C, 60.22; H, 4.62; N, 18.93 %; IR (KBr, cm<sup>-1</sup>): 3344 (OH), 3284 (NH), 1704 (C=O), 1622 (C=N). <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 6.86-6.90 (4H, *m*), 7.22-7.26 (2H, *m*), 7.68-7.71 (2H, *m*), 8.43 (2H, *s*), 10.84 (4H, *br*). <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 116.10, 119.10, 119.60, 128.10, 130.60, 142.60, 151.90, 156.60.

*1,5-Bis(2-pyridinylmethylidene)carbonohydrazide (9)*<sup>5</sup>. White solid (ethanol). Yield: 88 %; m.p. 185°C (lit m.p. 190-191°C). Anal. Calcd. for C<sub>13</sub>H<sub>12</sub>N<sub>6</sub>O (*Mw* = 268.11 g mol<sup>-1</sup>): C, 58.20; H, 4.51; N, 31.33 %. Found: C, 58.12; H, 4.88; N, 30.98 %. IR (KBr, cm<sup>-1</sup>): 3201 (NH), 1696 (C=O), 1604 (C=N). <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 11.08 (1H, *s*, N-H<sub>urea</sub>), 8.59 (1H, *d*, *J* = 4.4 Hz, C-H<sub>ar</sub>), 8.25 (1H, *s*, C-H<sub>imine</sub>), 8.15 (1H, *s*, N-H<sub>urea</sub>), 7.87 (1H, *t*, *J* = 7.6, C-H<sub>ar</sub>), 7.39 (2H, *t*, *J* = 6.0, C-H<sub>ar</sub>). <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>, δ / ppm): 151.68, 148.56, 144.59, 143.83, 126.01, 123.88.

*1,5-Bis[1-(2-pyridinylethylidene]carbonohydrazide (10)*<sup>2</sup>. White solid (ethanol). Yield: 88,0 %; m.p.(decomp.) 187 °C (lit m.p.(decomp.) 186°C). Anal. Calcd. for C<sub>15</sub>H<sub>16</sub>N<sub>6</sub>O (*Mw* = 296.14 g mol<sup>-1</sup>): C, 60.80; H, 5.44; N, 28.36 %. Found: C, 60.61; H, 5.31; N, 28.62 %. IR

(KBr,  $\text{cm}^{-1}$ ): 3206 (NH), 1698 (C=O), 1611 (C=N).  $^1\text{H}$  NMR (500 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 2.31 (6H, *s*, H<sub>3</sub>-CH<sub>3</sub>), 7.42 (2H, *dd*, H-C<sub>5</sub> = H-C<sub>13</sub>,  $^3J_{5,4} = ^3J_{13,12} = 8.1$  Hz,  $^3J_{5,6} = ^3J_{13,14} = 4.6$  Hz), 7.88 (2H, *ddd*, H-C<sub>4</sub> = H-C<sub>12</sub>,  $^3J_{4,3} = ^3J_{12,11} = 7.9$  Hz,  $^3J_{4,5} = ^3J_{12,13} = 6.2$  Hz,  $^4J_{4,6} = ^4J_{12,14} = 1.7$  Hz), 8.16 (2H, *d*, H-C<sub>3</sub> = H-C<sub>11</sub>,  $^3J_{3,4} = ^3J_{11,12} = 7.9$  Hz), 8.58 (2H, *dd*, H-C<sub>6</sub> = H-C<sub>14</sub>,  $^3J_{6,5} = ^3J_{14,13} = 4.6$  Hz,  $^4J_{6,4} = ^4J_{14,12} = 1.7$  Hz), 10.31 (2H, *s*, H-N<sub>3</sub> = H-N<sub>4</sub>).  $^{13}\text{C}$  NMR (126 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 13.25 (CH<sub>3</sub>), 123.58 (C<sub>3</sub> = C<sub>11</sub>), 133.70 (C<sub>5</sub> = C<sub>13</sub>), 145.81 (C<sub>4</sub> = C<sub>12</sub>), 147.14 (C<sub>7</sub> = C<sub>9</sub>), 149.61 (C<sub>6</sub> = C<sub>14</sub>), 151.45 (C<sub>2</sub> = C<sub>10</sub>), 152.12 (C<sub>8</sub>). lit.  $^1\text{H}$  NMR (300.00 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 2.38 (6H, *s*, CH<sub>3</sub>), 7.38 (2H, *t*, py), 7.85 (2H, *t*, py), 8.09 (2H, *d*, py), 8.59 (2H, *d*, py), 10.30 (2H, *br*, NH).

*1,5-Bis[phenyl(2-pyridinyl)methylidene]carbonohydrazide (11)*<sup>2,6</sup>. White solid (ethanol). Yield: 91.0 %; m.p. 223 °C (lit M.p. 225–226°C). Anal. Calcd. for C<sub>25</sub>H<sub>20</sub>N<sub>6</sub>O ( $M_w = 420.17$  g mol<sup>-1</sup>): C, 71.41; H, 4.79; N, 19.99 %. Found: C, 71.16; H, 4.82; N, 20.04 %. IR (KBr,  $\text{cm}^{-1}$ ): 3177 (NH), 1702 (C=O), 1612 (C=N).  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 7.20–8.79 (18H, *m*, C-H<sub>ar</sub>), 9.90 (1H, *s*, N–H), 12.74 (1H, *s*, N–H).  $^{13}\text{C}$  NMR (62.90 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 123.70, 124.60, 127.20, 128.70, 129.00, 129.50, 131.00, 133.40, 136.80, 138.10, 148.80, 149.00, 162.40.

*1,5-Bis(2-quinolinylethylidene)carbonohydrazide (12)*<sup>3</sup>. White solid (DMF/methanol mixture 1 : 9 v/v). Yield 78 %; m.p. 162–164 °C (lit m.p. 162–164°C). Anal. Calcd. for C<sub>21</sub>H<sub>16</sub>N<sub>6</sub>O ( $M_w = 368.14$  g mol<sup>-1</sup>): C, 68.47; H, 4.38; N, 22.81 %. Found: C, 68.81; H, 4.80; N, 22.56 %. IR (KBr,  $\text{cm}^{-1}$ ): 3392 (NH), 1708 (C=O), 1630 (C=N).  $^1\text{H}$  NMR (500 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 7.63 (2H, *ddd*, H-C<sub>6</sub> = H-C<sub>16</sub>,  $^3J_{6,5} = ^3J_{16,15} = 8.2$  Hz,  $^3J_{6,7} = ^3J_{16,17} = 6.8$  Hz,  $^4J_{6,8} = ^4J_{16,18} = 1.2$  Hz), 7.79 (2H, *ddd*, H-C<sub>7</sub> = H-C<sub>17</sub>,  $^3J_{7,8} = ^3J_{17,18} = 7.9$  Hz,  $^3J_{7,6} = ^3J_{17,16} = 6.8$  Hz,  $^4J_{7,5} = ^3J_{17,15} = 1.7$  Hz), 7.99–8.13 (4H, *m*, H-C<sub>5</sub> = H-C<sub>15</sub>, H-C<sub>8</sub> = H-C<sub>18</sub>), 8.31 (2H, *s*, H-C<sub>9</sub> = H-C<sub>11</sub>), 8.38–8.60 (4H, *br.m.ovlp.*, H-C<sub>3</sub> = H-C<sub>13</sub>, H-C<sub>4</sub> = H-C<sub>14</sub>), 11.31 (2H, *s*, H-N<sub>3</sub> = H-N<sub>4</sub>).  $^{13}\text{C}$  NMR (126 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 117.78 (C<sub>3</sub> = C<sub>13</sub>), 127.11 (C<sub>7</sub> = C<sub>17</sub>), 127.79 (C<sub>6</sub> = C<sub>16</sub>), 127.99 (C<sub>4a</sub> = C<sub>14a</sub>), 128.84 (C<sub>5</sub> = C<sub>15</sub>), 130.00 (C<sub>8</sub> = C<sub>18</sub>), 136.47 (C<sub>4</sub> = C<sub>14</sub>), 144.06 (C<sub>9</sub> = C<sub>11</sub>), 147.35 (C<sub>8a</sub> = C<sub>18a</sub>), 151.67 (C<sub>2</sub> = C<sub>12</sub>), 153.99 (C<sub>10</sub>).

*1,5-Bis(8-hydroxy-2-quinolinyl)methylidene]carbonohydrazide (13)*<sup>3</sup>. Yellow solid (DMF/methanol mixture 1 : 9 v/v). Yield: 66 %; m.p. 248–249 °C (lit m.p. 248–249°C). Anal. Calcd. for C<sub>21</sub>H<sub>16</sub>N<sub>6</sub>O<sub>3</sub> ( $M_w = 400.39$  g mol<sup>-1</sup>): C, 62.99; H, 4.03; N, 20.99 %. Found: C, 62.84; H, 4.11; N, 21.22 %. IR (KBr,  $\text{cm}^{-1}$ ): 3408 (OH), 3116 (NH), 1684 (C=O), 1601 (C=N).  $^1\text{H}$  NMR (500 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 7.12 (2H, *dd*, H-C<sub>7</sub> = H-C<sub>17</sub>,  $^3J_{7,6} = ^3J_{17,16} = 7.4$  Hz,  $^4J_{7,5} = ^4J_{17,15} = 1.5$  Hz), 7.41 (2H, *dd*, H-C<sub>5</sub> = H-C<sub>15</sub>,  $^4J_{5,7} = ^4J_{15,17} = 1.5$  Hz), 7.45 (2H, *t*, H-C<sub>6</sub> = H-C<sub>16</sub>,  $^3J_{6,7} = ^3J_{16,17} = 7.4$  Hz), 8.17–8.39 (4H, *br.m.ovlp.*, H-C<sub>3</sub> = H-C<sub>13</sub>, H-C<sub>4</sub> = H-C<sub>14</sub>), 8.48 (2H, *s*, H-C<sub>9</sub> = H-C<sub>11</sub>), 9.80 (2H, *s*, H-O<sub>1</sub> = H-O<sub>2</sub>), 11.34 (2H, *s*, H-N<sub>3</sub> = H-N<sub>4</sub>).  $^{13}\text{C}$  NMR (126 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 112.13 (C<sub>7</sub> = C<sub>17</sub>), 117.90 (C<sub>5</sub> = C<sub>15</sub>), 118.12 (C<sub>3</sub> = C<sub>13</sub>), 128.13 (C<sub>6</sub> = C<sub>16</sub>), 128.75 (C<sub>4a</sub> = C<sub>14a</sub>), 136.40 (C<sub>4</sub> = C<sub>14</sub>), 138.13 (C<sub>8a</sub> = C<sub>18a</sub>), 144.02 (C<sub>9</sub> = C<sub>11</sub>), 151.92 (C<sub>2</sub> = C<sub>12</sub>), 153.37 (C<sub>8</sub> = C<sub>18</sub>), 162.45 (C<sub>10</sub>).

*1,5-Bis(8-quinolinylethylidene)carbonohydrazide (14)*<sup>3</sup>. Yellow solid (methanol). Yield: 54 %; m.p. 219–220 °C (lit m.p. 219–220°C). Anal. Calcd. for C<sub>21</sub>H<sub>16</sub>N<sub>6</sub>O ( $M_w = 368.14$  g mol<sup>-1</sup>): C, 68.47; H, 4.38; N, 22.81 %. Found: C, 68.32; H, 4.91; N, 22.73 %. IR (KBr,  $\text{cm}^{-1}$ ): 3331 (NH), 1707 (C=O), 1614 (C=N).  $^1\text{H}$  NMR (500 MHz, DMSO- $d_6$ ,  $\delta$  / ppm): 7.61 (2H, *dd*, H-C<sub>3</sub> = H-C<sub>17</sub>,  $^3J_{3,2} = ^3J_{17,18} = 4.1$  Hz,  $^3J_{3,4} = ^3J_{17,16} = 8.3$  Hz), 7.72 (2H, *t*, H-C<sub>6</sub> = H-C<sub>14</sub>,  $^3J_{6,5} = ^3J_{14,15} = 7.9$  Hz); 8.04 (2H, *dd*, H-C<sub>5</sub> = H-C<sub>15</sub>,  $^3J_{5,6} = ^3J_{15,14} = 7.9$  Hz), 8.43 (2H, *dd*, H-C<sub>4</sub> = H-C<sub>16</sub>,  $^3J_{4,3} = ^3J_{16,17} = 8.3$  Hz,  $^4J_{4,2} = ^4J_{16,18} = 1.75$  Hz), 8.60 (2H, *d*, H-C<sub>7</sub> = H-C<sub>13</sub>,  $^3J_{7,6} = ^3J_{13,14} = 0.4$  Hz), 8.99 (2H, *dd*, H-C<sub>2</sub> = H-C<sub>18</sub>,  $^3J_{2,3} = ^3J_{18,17} = 4.1$  Hz,  $^4J_{2,4} = ^4J_{18,16} = 1.75$  Hz), 9.50 (2H, *s*, H-C<sub>9</sub> = H-C<sub>11</sub>), 11.09 (2H, *s*, H-N<sub>3</sub>, H-N<sub>4</sub>).  $^{13}\text{C}$  NMR (126 MHz, DMSO- $d_6$ ,  $\delta$  / ppm):

121.77 ( $C_3 = C_{17}$ ), 125.72 ( $C_7 = C_{13}$ ), 126.49 ( $C_6 = C_{14}$ ), 128.02 ( $C_{4a} = C_{15a}$ ), 129.28 ( $C_5 = C_{15}$ ), 131.65 ( $C_8 = C_{12}$ ), 136.62 ( $C_4 = C_{16}$ ), 139.89 ( $C_9 = C_{11}$ ), 145.19 ( $C_{8a} = C_{12a}$ ), 150.16 ( $C_2 = C_{18}$ ), 152.28 ( $C_{10}$ ).

TABLE S-I. *In vitro* antifungal activity of the compounds tested by the well-diffusion agar assay expressed as the diameter (mm) of the inhibition zone (includes diameter of the well of 8 mm)

Tested compound	<i>C. albicans</i>	<i>S. cerevisiae</i>	<i>A. brasiliensis</i>
<b>1</b>	—	—	—
<b>2</b>	—	—	—
<b>3</b>	—	—	—
<b>4</b>	—	—	—
<b>5</b>	10	14	12
<b>6</b>	12	14	12
<b>7</b>	12	16	16
<b>8</b>	—	—	—
<b>9</b>	—	—	—
<b>10</b>	—	—	—
<b>11</b>	—	—	—
<b>12</b>	—	—	—
<b>13</b>	12	10	10
<b>14</b>	14	10	10
Nystatin	34	56	32

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