**N-alkylation of organonitrogen compounds catalyzed by methylene-linked bis-NHC half-sandwich ruthenium complexes**

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1. Optimization of reaction conditions

Table S1. Initial Screening of Reaction Conditions for N-alkylation of Aniline with Benzyl Alcohol

<table>
<thead>
<tr>
<th>entry</th>
<th>catalyst (mol %)</th>
<th>base (mol %)</th>
<th>yield 6a (%)&lt;sup&gt;b&lt;/sup&gt;</th>
<th>yield 7a (%)&lt;sup&gt;b&lt;/sup&gt;</th>
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<tbody>
<tr>
<td>1</td>
<td>1 (0.5)</td>
<td>K&lt;sub&gt;2&lt;/sub&gt;CO&lt;sub&gt;3&lt;/sub&gt; (10)</td>
<td>10</td>
<td>31</td>
</tr>
<tr>
<td>2</td>
<td>2 (0.5)</td>
<td>K&lt;sub&gt;2&lt;/sub&gt;CO&lt;sub&gt;3&lt;/sub&gt; (10)</td>
<td>18</td>
<td>48</td>
</tr>
<tr>
<td>3</td>
<td>3 (0.5)</td>
<td>K&lt;sub&gt;2&lt;/sub&gt;CO&lt;sub&gt;3&lt;/sub&gt; (10)</td>
<td>11</td>
<td>44</td>
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<tr>
<td>4</td>
<td>1 (0.5)</td>
<td>KOiPr (10)</td>
<td>70</td>
<td>14</td>
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<tr>
<td>5</td>
<td>2 (0.5)</td>
<td>KOiPr (10)</td>
<td>82</td>
<td>7</td>
</tr>
<tr>
<td>6</td>
<td>3 (0.5)</td>
<td>KOiPr (10)</td>
<td>75</td>
<td>9</td>
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<td>7</td>
<td>1 (0.5)</td>
<td>NaOH (10)</td>
<td>93</td>
<td>5</td>
</tr>
<tr>
<td>8</td>
<td>2 (0.5)</td>
<td>NaOH (10)</td>
<td>89</td>
<td>8</td>
</tr>
<tr>
<td>9</td>
<td>3 (0.5)</td>
<td>NaOH (10)</td>
<td>85</td>
<td>11</td>
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<tr>
<td>10</td>
<td>1 (0.5)</td>
<td>tBuOK (10)</td>
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<tr>
<td>11</td>
<td>2 (0.5)</td>
<td>tBuOK (10)</td>
<td>92</td>
<td>7</td>
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<tr>
<td>12</td>
<td>3 (0.5)</td>
<td>tBuOK (10)</td>
<td>85</td>
<td>14</td>
</tr>
<tr>
<td>13</td>
<td>1 (0.5)</td>
<td>KOH (10)</td>
<td>93</td>
<td>5</td>
</tr>
<tr>
<td>14</td>
<td>2 (0.5)</td>
<td>KOH (10)</td>
<td>&gt;99</td>
<td>-</td>
</tr>
<tr>
<td>15</td>
<td>3 (0.5)</td>
<td>KOH (10)</td>
<td>96</td>
<td>2</td>
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<tr>
<td>16</td>
<td>-</td>
<td>KOH (10)</td>
<td>-</td>
<td>5</td>
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<tr>
<td>17</td>
<td>2 (0.5)</td>
<td>-</td>
<td>-</td>
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<sup>a</sup>Unless noted otherwise, reactions were carried out with aniline (1.0 mmol), benzyl alcohol (1.0 mmol), base (10 mol %) and Ru cat. (0.5 mol %) in 2 mL of toluene at 100 ºC.<sup>b</sup>Determined by GC-MS after 2h.
Table S2. Solvent Screening for N-alkylation of Aniline with Benzyl Alcohol Using Complex 2\(^a\)

![Reaction Scheme]

<table>
<thead>
<tr>
<th>entry</th>
<th>solvent</th>
<th>yield 6a (%)(^b)</th>
<th>yield 7a (%)(^b)</th>
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<tr>
<td>1</td>
<td>toluene</td>
<td>&gt;99</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>p-xylene</td>
<td>&gt;99</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>1,4-dioxane</td>
<td>78</td>
<td>3</td>
</tr>
<tr>
<td>4</td>
<td>DMF</td>
<td>-</td>
<td>3</td>
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<tr>
<td>5</td>
<td>tBuOH</td>
<td>49</td>
<td>30</td>
</tr>
<tr>
<td>6</td>
<td>-</td>
<td>98</td>
<td>2</td>
</tr>
</tbody>
</table>

\(^a\)Unless noted otherwise, reactions were carried out with aniline (1.0 mmol), benzyl alcohol (1.0 mmol), KOH (10 mol %) and complex 2 (0.5 mol %) in 2 mL of solvent at 100 ºC.

\(^b\)Determined by GC-MS after 2h.

Table S3. Temperature Optimization for N-alkylation of Aniline with Benzyl Alcohol Using Complex 2\(^a\)

![Reaction Scheme]

<table>
<thead>
<tr>
<th>entry</th>
<th>T (ºC)</th>
<th>yield 6a (%)(^b)</th>
<th>yield 7a (%)(^b)</th>
</tr>
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<td>-</td>
</tr>
<tr>
<td>2</td>
<td>80</td>
<td>&gt;99</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>60</td>
<td>75</td>
<td>25</td>
</tr>
</tbody>
</table>

\(^a\)Unless noted otherwise, reactions were carried out with aniline (1.0 mmol), benzyl alcohol (1.0 mmol), KOH (10 mol %) and complex 2 (0.5 mol %) in 2 mL of toluene. \(^b\)Determined by GC-MS after 2h.
**Table S4.** Optimization of the Amount of Complex 2 for N-alkylation of Aniline with Benzyl Alcohol$^a$

<table>
<thead>
<tr>
<th>entry</th>
<th>cat 2 (mol %)</th>
<th>yield 6a (%)$^b$</th>
<th>yield 7a (%)$^b$</th>
<th>TON$^c$</th>
<th>TOF (h$^{-1}$)$^d$</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>0.5</td>
<td>&gt;99</td>
<td>-</td>
<td>198</td>
<td>99</td>
</tr>
<tr>
<td>2</td>
<td>0.1</td>
<td>94</td>
<td>5</td>
<td>940</td>
<td>470</td>
</tr>
<tr>
<td>3$^e$</td>
<td>0.1</td>
<td>6</td>
<td>78</td>
<td>60</td>
<td>30</td>
</tr>
<tr>
<td>4</td>
<td>0.05</td>
<td>90</td>
<td>10</td>
<td>1800</td>
<td>900</td>
</tr>
</tbody>
</table>

$^a$Unless noted otherwise, reactions were carried out with aniline (2.0 mmol), benzyl alcohol (2.0 mmol), KOH (10 mol %) in 2 mL of toluene at 80ºC. $^b$Determined by CG-MS after 2h. $^c$Turnover number (moles of aniline converted to 6a per mole of catalyst). $^d$Turnover frequency (moles of aniline converted to 6a per mole of catalyst per hour). $^e$The reaction was carried out at 60ºC.
2. General method for the *N*-alkylation of amines with alcohols. Characterization data for compounds 6a-p, 8a-q, 9, 10a-b and 11.

The corresponding primary alcohols (1.0 mmol) and amines (1.0 mmol) were added to a solution of the ruthenium catalyst **2** (0.001 mmol, 0.8 mg) and KOH (0.1 mmol, 5.6 mg) in degassed toluene (2 mL) under argon. The reaction mixture was stirred at 80 ºC for 2 h and then evaluated by TLC and GC-MS. The reaction mixture was cooled to room temperature and the solvent evaporated under reduced pressure. The resulting crude product was purified by column chromatography on silica gel using mixtures of hexanes and ethyl acetate as eluents to afford the corresponding *N*-alkylated products.

**N**-benzylaniline (6a). Pale yellow solid, mp 38-40 ºC (181.2 mg, 99% isolated yield) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl₃): δ 4.06 (br s, NH), 4.33 (s, 2H), 6.65 (d, J = 8.1 Hz, 2H), 6.72 (tt, J = 7.3, 1.1 Hz, 1H), 7.15-7.19 (m, 2H), 7.27-7.38 (m, 5H) ppm. 13C NMR (100 MHz, CDCl₃): δ 148.1, 139.4, 129.2 (2C), 128.6 (2C), 127.4 (2C), 127.2, 117.5, 112.8 (2C), 48.2 ppm.

**N**-(2-methylbenzyl)aniline (6b). Yellow oil (173.6 mg, 88%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl₃): δ 2.38 (s, 3H), 3.90 (br s, NH), 4.28 (s, 2H), 6.65 (d, J = 7.6 Hz, 2H), 6.73 (tt, J = 7.3, 1.1 Hz, 1H), 7.17-7.22 (m, 5H), 7.34 (d, J = 6.6 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl₃): δ 148.2, 136.9, 136.2, 130.3, 129.2 (2C), 128.1, 127.3, 126.0, 117.3, 112.6 (2C), 46.2, 18.8 ppm.

**N**-(3-methylbenzyl)aniline (6c). White solid, mp 42-44 ºC (189.4 mg, 96%) (flash chromatography 40% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl₃): δ 2.35 (s, 3H), 4.07 (br s, NH), 4.28 (s, 2H), 6.64 (d, J = 8.6 Hz, 2H), 6.72 (tt, J = 7.3, 1.1 Hz, 1H), 7.09 (d, J = 8.1 Hz, 1H), 7.16-7.23 (m, 5H) ppm. 13C NMR (100 MHz, CDCl₃): δ 148.1, 139.3, 138.1, 129.1 (2C), 128.4, 128.2, 127.9, 124.5, 117.4, 112.7 (2C), 117.3, 112.6 (2C), 48.2, 21.3 ppm.

**N**-(4-methylbenzyl)aniline (6d). White solid, mp 42-44 ºC (177.5 mg, 90%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl₃): δ 2.45 (s, 3H), 4.36 (s, 2H), 6.72 (d, J = 8.2 Hz, 2H), 6.82 (tt, J = 7.3, 1.1 Hz, 1H), 7.24-7.29 (m, 4H), 7.35 (d, J = 8.2 Hz, 2H) ppm. 13C NMR (100 MHz, CDCl₃): δ 148.2, 136.8, 136.3, 129.3 (2C), 129.2 (2C), 127.5 (2C), 117.5, 112.8 (2C), 48.1, 21.1 ppm.

**N**-(4-fluorobenzyl)aniline (6e). Yellow oil (199.2 mg, 99%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl₃): δ 4.06 (br s, NH), 4.38 (s, 2H), 6.74 (d, J = 7.5 Hz, 2H), 6.82 (tt, J = 7.5, 1.1 Hz, 1H), 7.15 (d, J = 8.7 Hz, 2H), 7.32 (dd, J = 8.7, 7.5 Hz, 2H), 7.41-7.46 (m, 2H) ppm. 13C NMR (100 MHz, CDCl₃): δ 148.2 (d, JCF = 245.0 Hz), 147.9, 135.0 (d, JCF = 8.0 Hz, 2C), 117.7, 115.4 (d, JCF = 21.4 Hz, 2C), 112.9 (2C), 47.6 ppm. 19F NMR (376 MHz, CDCl₃): δ -115.6 ppm.

**N**-(4-chlorobenzyl)aniline (6f). Yellow oil (204.6 mg, 94%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl₃): δ 4.33 (s, 2H), 6.65 (d, J = 7.7 Hz, 2H), 6.79 (tt, J = 7.3, 1.1 Hz, 1H), 7.20-7.26 (m, 2H), 7.34 (m, 4H) ppm. 13C NMR (100 MHz, CDCl₃): δ 147.8, 138.0, 132.8, 129.3 (2C), 128.73 (2C), 128.67 (2C), 117.8, 112.9 (2C), 47.6 ppm.
**N-benzylpyridin-2-amine (6g).** White solid, mp 94-96 °C (182.4 mg, 99%) (flash chromatography 25% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.51 (d, J = 5.8 Hz, 2H), 5.02 (br s, 2H), 6.37 (d, J = 8.4 Hz, 1H), 6.59 (dd, J = 7.1, 5.2, 0.9 Hz, 1H), 7.24-7.43 (m, 6H), 8.10 (d, J = 5.2 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 158.6, 148.1, 139.1, 137.4, 128.6 (2C), 127.3 (2C), 127.2, 113.1, 106.7, 46.3 ppm.

**N-(4-methylbenzyl)pyridin-2-amine (6h).** White solid, mp 74-76 °C (182.4 mg, 92%) (flash chromatography 25% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 2.32 (s, 3H), 4.41 (d, J = 5.7 Hz, 2H), 5.01 (br s, 2H), 6.33 (d, J = 8.6 Hz, 1H), 6.54 (dd, J = 7.2, 5.0, 0.9 Hz, 1H), 7.12 (d, J = 7.8 Hz, 2H), 7.22 (d, J = 7.8 Hz, 2H), 7.36 (dd, J = 8.6, 7.2, 1.9 Hz, 1H), 8.05 (d, J = 5.0 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 158.6, 148.0, 137.4, 136.7, 136.0, 129.2 (2C), 127.3 (2C), 112.9, 106.6, 46.0, 21.0 ppm.

**N-(4-methoxybenzyl)pyridin-2-amine (6i).** White solid, mp 128-131 °C (212.1 mg, 99%) (flash chromatography 35% ethyl acetate/petroleum ether). 1H NMR (500 MHz, CDCl3): δ 3.79 (s, 3H), 4.41 (d, J = 5.4 Hz, 2H), 4.97 (br s, 2H), 6.36 (dt, J = 8.4, 1.0 Hz, 1H), 6.57 (dd, J = 7.1, 5.1, 1.0 Hz, 1H), 6.85-6.88 (m, 2H), 7.26-7.29 (m, 2H), 7.39 (dd, J = 8.4, 7.2, 1.9 Hz, 1H), 8.08 (dd, J = 5.1, 1.9, 1.0 Hz, 1H) ppm. 13C NMR (125 MHz, CDCl3): δ 158.8, 158.6, 148.0, 137.4, 131.1, 128.6 (2C), 113.9 (2C), 113.0, 106.7 55.2, 45.7 ppm.

**N-(4-chlorobenzyl)pyridin-2-amine (6j).** White solid, mp 104-106 °C (216.5 mg, 99%) (flash chromatography 30% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.46 (d, J = 6.0 Hz, 2H), 5.04 (br s, 2H), 6.33 (d, J = 8.4 Hz, 1H), 6.59 (dd, J = 7.1, 5.1, 0.9 Hz, 1H), 7.25-7.29 (m, 4H), 7.38 (dd, J = 8.4, 7.1, 1.9 Hz, 1H), 8.07 (d, J = 5.1 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 158.4, 148.1, 137.8, 137.5, 132.8, 128.7 (2C), 113.9 (2C), 113.0, 106.7 55.2, 45.7 ppm.

**N-(Benzo[1,3]dioxol-5-ylmethyl)aniline (6k).** White solid, mp 78-80 °C (184.1 mg, 81%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 3.98 (br s, 2H), 4.23 (s, 2H), 5.94 (s, 2H), 6.61-6.65 (m, 2H), 6.72 (t, J = 7.6, 1.1 Hz, 1H), 6.77 (d, J = 7.6 Hz, 1H), 6.82-6.85 (m, 1H), 6.86-6.88 (m, 1H), 7.18 (dd, J = 8.6, 7.2 Hz, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 148.0, 147.9, 146.7, 133.3, 129.2 (2C), 120.6, 117.6, 112.8 (2C), 108.3, 108.0, 101.0, 48.1 ppm.

**N-(3-((tert-butyldimethylsilyl)oxy)benzyl)aniline (6l).** Colourless oil (163.0 mg, 52%) (flash chromatography 3% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 0.15 (s, 6H), 0.95 (s, 9H), 4.23 (s, 2H), 5.94 (s, 2H), 6.61-6.65 (m, 2H), 6.72 (t, J = 7.6, 1.1 Hz, 1H), 6.77 (d, J = 7.6 Hz, 1H), 6.82-6.85 (m, 1H), 6.86-6.88 (m, 1H), 7.18 (dd, J = 8.6, 7.2 Hz, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 155.9, 147.3, 140.4, 129.6, 129.3, 120.6, 119.4, 119.0, 118.2, 113.5, 48.6, 25.7, 18.2, -4.42 ppm.

**N-phenethylaniline (6m).** Colourless oil (193.3 mg, 98%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 0.15 (s, 6H), 0.95 (s, 9H), 4.23 (s, 2H), 5.94 (s, 2H), 6.61-6.65 (m, 2H), 6.72 (t, J = 7.6, 1.1 Hz, 1H), 6.77 (d, J = 7.6 Hz, 1H), 6.82-6.85 (m, 1H), 6.86-6.88 (m, 1H), 7.18 (dd, J = 8.6, 7.2 Hz, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 148.0, 147.9, 146.7, 133.3, 129.2 (2C), 120.6, 117.6, 112.8 (2C), 108.3, 108.0, 101.0, 48.1 ppm.

**N-(cyclopropylmethyl)aniline (6n).** Yellow oil (145.7 mg, 99%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 0.32 (dt, J = 5.9, 4.4 Hz, 2H), 0.60-0.67
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(m, 2H), 1.12-1.24 (m, 1H), 3.04 (d, J = 6.9 Hz, 2H), 3.73 (br s, NH), 6.70 (d, J = 7.4 Hz, 2H), 6.79 (tt, J = 7.4, 1.1 Hz, 1H), 7.21-7.31 (m, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 148.4, 129.2 (2C), 117.3, 112.8 (2C), 49.1, 10.9, 3.4 (2C) ppm.

N-ethylpyridin-2-amine (6o). White solid, mp 78-80 °C (120.9 mg, 99%) (flash chromatography 10% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 1.24 (t, J = 7.2 Hz, 3H), 3.28 (quint, J = 7.2 Hz, 2H), 4.45 (br s, NH), 6.35 (d, J = 8.6 Hz, 1H), 6.54 (ddd, J = 8.4, 1.9 Hz, 1H), 7.40 (ddd, J = 8.6, 7.2, 1.8 Hz, 1H), 8.06 (dd, J = 5.1, 1.8 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 158.8, 148.2, 137.4, 112.7 106.3, 36.9, 14.8 ppm.

N-methylpyridin-2-amine (6p). 10 Yellow oil (56.2 mg, 52%) (flash chromatography 50% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 2.91 (d, J = 4.0 Hz, 3H), 4.64 (br s, NH), 6.39 (dt, J = 8.4, 1.0 Hz, 1H), 6.57 (ddd, J = 7.2, 5.1, 0.9 Hz, 1H), 7.44 (ddd, J = 8.4, 7.2, 1.9 Hz, 1H), 8.06 (ddd, J = 5.1, 1.9, 0.9 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 159.5, 147.4, 137.9, 112.6, 106.3, 29.1 ppm.

N-benzyl-2-bromoaniline (8a).3 White solid, mp 52-54 °C (215.0 mg, 82%) (flash chromatography 10% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.41 (d, J = 5.7 Hz, 2H), 4.76 (br s, NH), 6.56-6.63 (m, 2H), 7.13 (ddd, J = 8.5, 7.4, 1.5 Hz, 1H), 7.27-7.32 (m, 1H), 7.33-7.39 (m, 4H), 7.45 (dd, J = 7.9, 1.5 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 147.0, 138.8, 131.9 (2C), 128.7 (2C), 127.4 (2C), 127.3, 123.1, 120.1, 115.3, 111.4, 47.9 ppm.

N-benzyl-4-bromoaniline (8c).11 White solid, mp 52-54 °C (215.0 mg, 82%) (flash chromatography 10% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.08 (br s, NH), 4.31 (s, 2H), 6.51 (d, J = 8.7 Hz, 2H), 7.25 (d, J = 8.7 Hz, 2H), 7.27-7.41 (m, 5H) ppm. 13C NMR (100 MHz, CDCl3): δ 147.0, 138.8, 131.9 (2C), 128.7 (2C), 127.4 (2C), 127.3, 123.1, 120.1, 115.3, 111.4, 47.9 ppm.

N-benzyl-4-chloroaniline (8d).11 Colourless oil (213.3 mg, 98%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.32 (s, 2H), 6.57 (d, J = 8.9 Hz, 2H), 7.14 (d, J = 8.9 Hz, 2H), 7.28-7.40 (m, 5H) ppm. 13C NMR (100 MHz, CDCl3): δ 146.6, 138.9, 129.0 (2C), 128.6 (2C), 127.36 (2C), 127.32, 122.0, 113.9 (2C), 48.3 ppm.

4-(benzylamino)benzonitrile (8e).12 White solid, mp 62-64 °C (202.0 mg, 97%) (flash chromatography 10% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.37 (s, 2H), 4.60 (br s, NH), 6.54-6.63 (m, 2H), 7.28-7.37 (m, 5H), 7.38-7.44 (m, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 151.0, 137.8, 133.7 (2C), 128.8 (2C), 127.7, 127.3 (2C), 120.3, 112.4 (2C), 99.1, 47.5 ppm.

N-benzyl-4-methylaniline (8f).4 Yellow oil (195.3 mg, 99%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 2.36 (s, 3H), 3.98 (br s, NH), 4.39 (s, 2H), 6.79 (tt, J = 7.4, 1.1 Hz, 1H).
6.66 (d, J = 8.3 Hz, 2H), 7.10 (d, J = 8.3 Hz, 2H), 7.38 (m, 1H), 7.42-7.51 (m, 4H) ppm. 13C NMR (100 MHz, CDCl3): δ 145.8, 139.6, 129.6 (2C), 128.4 (2C), 128.1, 127.4 (2C), 127.0, 112.9 (2C), 48.5, 20.3 ppm.

N-benzynaphthalen-1-amine (8g). Brown solid, mp 58-60 ºC (231.0 mg, 99%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (500 MHz, CDCl3): δ 4.52 (s, 2H), 4.84 (br s, N-H), 6.69 (dd, J = 7.4, 1.2 Hz, 1H), 7.32-7.54 (m, 9H), 7.84-7.89 (m, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 143.0, 139.0, 134.2, 128.5 (2C), 128.4, 127.6, 125.7, 124.7, 123.3, 119.9, 117.7, 104.8, 48.6 ppm.

N-benzylquinolin-2-amine (8h). Yellow solid, mp 91-93 ºC (231.9 mg, 99%) (flash chromatography 20% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.75 (d, J = 5.5 Hz, 2H), 5.26 (br s, N-H), 6.61 (d, J = 8.9 Hz, 1H), 7.23-7.39 (m, 4H), 7.41-7.46 (m, 2H), 7.54-7.65 (m, 2H), 7.78-7.82 (m, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 156.6, 147.9, 139.3, 137.2, 129.4, 128.5 (2C), 127.6 (2C), 127.3, 127.1, 123.4, 122.0, 111.2, 45.7 ppm.

N-benzylpyrimidin-2-amine (8i). White solid, mp 74-76 ºC (183.4 mg, 99%) (flash chromatography 25% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.63 (d, J = 5.7 Hz, 2H), 6.44 (t, J = 4.8 Hz, 1H), 6.67 (br s, N-H), 7.23-7.40 (m, 5H), 8.08 (br s, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 162.2, 157.8 (2C), 139.0, 128.4 (2C), 127.5 (2C), 127.1, 110.4, 45.4 ppm.

N-benzylbenzo[d]thiazol-2-amine (8j). White solid, mp 154-156 ºC (209.2 mg, 99%) (flash chromatography 20% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.64 (s, 2H), 6.37 (br s, N-H), 7.08 (dt, J = 7.6, 1.2 Hz, 1H), 7.23-7.43 (m, 6H), 7.45 (dd, J = 8.2, 1.2 Hz, 1H), 7.57 (dd, J = 7.9, 1.2 Hz, 1H) ppm. 13C NMR (100 MHz, CDCl3): δ 167.6, 152.2, 137.4, 130.4 (2C), 128.8, 127.8 (2C), 127.6, 126.0, 121.6, 120.8, 118.9, 49.4 ppm.

(E)-N-benzyl-4-styrylaniline (8k). White solid, mp 88-90 ºC (171.2 mg, 60%) (flash chromatography 3% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 4.36 (s, 2H), 6.64 (d, J = 8.5 Hz, 2H), 6.89 (d, J = 16.3 Hz, 1H), 7.01 (d, J = 16.3 Hz, 1H), 7.16-7.22 (m, 1H), 7.44-7.47 (m, 2H) ppm. 13C NMR (100 MHz, CDCl3): δ 147.7, 150.6, 128.7 (2C), 128.5 (2C), 127.7 (2C), 127.4, 127.3 (2C), 127.2, 126.7, 126.0 (2C), 124.5, 112.9, 48.1 ppm.

Dibenzylamine (8l). Colourless oil (148.0 mg, 75%) (flash chromatography 20% ethyl acetate/petroleum ether). 1H NMR (400 MHz, CDCl3): δ 1.57 (br s, N-H), 3.74 (s, 4H), 7.16-7.28 (m, 10H) ppm. 13C NMR (100 MHz, CDCl3): δ 140.3 (2C), 128.4 (4C), 128.1 (4C), 126.9 (2C), 53.2 (2C) ppm.

N-benzyl-1-phenylethanamine (8m). Colourless oil (171.1 mg, 81%) (flash chromatography 5% ethyl acetate/petroleum ether). 1H NMR (500 MHz, CDCl3): δ 1.39 (br s, N-H), 3.74 (s, 4H), 7.16-7.28 (m, 10H) ppm. 13C NMR (100 MHz, CDCl3): δ 145.5, 140.6, 128.4 (2C), 128.3 (2C), 128.1 (2C), 126.9, 126.8, 126.7 (2C), 57.5, 51.6, 24.5 ppm.

N-benzylcyclohexanamine (8n). Pale yellow oil (153.4 mg, 81%) (flash chromatography petroleum ether/ethyl acetate/Et3N 4:1:0.05). 1H NMR (400 MHz, CDCl3): δ 1.11-1.23 (m, 5H),
1.61-1.66 (m, 1H), 1.73-1.82 (m, 2H), 1.88-1.92 (m, 1H), 3.76 (s, 2H), 7.19-7.22 (m, 1H), 7.27-7.31 (m, 4H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 141.1, 128.3 (2C), 128.1 (2C), 126.8, 55.9, 50.5, 33.0 (2C), 25.9, 24.9 (2C) ppm.

1-benzylpyrrolidine (8o). Yellow oil (132.2 mg, 82%) (flash chromatography 2% MeOH/DCM). $^1$H NMR (500 MHz, CDCl$_3$): $\delta$ 1.76-1.78 (m, 4H), 2.51-2.54 (m, 4H), 3.63 (s, 2H), 7.22-7.34 (m, 5H) ppm. $^{13}$C NMR (125 MHz, CDCl$_3$): $\delta$ 138.7, 128.9 (2C), 128.1 (2C), 126.9, 60.4, 53.9 (2C), 23.2 (2C) ppm.

1-(4-methoxybenzyl)pyrrolidine (8p). Pale yellow oil (118.6 mg, 62%) (flash chromatography petroleum ether/ethyl acetate/Et$_3$N 7:1:0.2). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 1.74-1.80 (m, 4H), 2.47-2.51 (m, 4H), 3.55 (s, 2H), 3.79 (s, 3H), 6.81-6.88 (m, 2H), 7.24 (d, $J = 8.6$ Hz, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 158.6, 130.0 (2C), 128.2, 113.5 (2C), 60.0, 55.2, 54.0 (2C), 23.4 (2C) ppm.

1-benzylpiperidine (8q). Light orange oil (61.3 mg, 35%) (flash chromatography petroleum ether/ethyl acetate/Et$_3$N 4:1:0.05). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 1.41-1.46 (m, 2H), 1.56-1.61 (m, 4H), 2.40 (br s, 4H), 3.50 (s, 2H), 7.30-7.31 (m, 3H), 7.35-7.39 (m, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 138.0, 129.4 (2C), 128.1 (2C), 127.0, 63.6, 54.3 (2C), 25.7 (2C), 24.2 ppm.

N-phenylpyrrolidine (9). Colourless oil (75.1 mg, 51%) (flash chromatography 5% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 2.02-2.08 (m, 4H), 3.31-3.37 (m, 4H), 6.63 (d, $J = 7.7$ Hz, 2H), 6.71 (t, $J = 7.7$ Hz, 1H), 7.25-7.31 (m, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 147.9, 129.1 (2C), 115.4, 111.6 (2C), 47.6 (2C), 25.4 (2C) ppm.

1,2,3,4-Tetrahydroquinoxaline (10a). Yellow solid, mp 99-101 ºC (41.6 mg, 31%) (flash chromatography 15% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 3.41 (s, 4H), 6.47-6.52 (m, 2H), 6.56-6.60 (m, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 133.6 (2C), 118.8 (2C), 114.7 (2C), 41.4 (2C) ppm.

1,4-dihydroquinoxaline (10b). Amorphous yellow solid (66.1 mg, 50%) (flash chromatography 15% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.74-7.81 (m, 2H), 8.08-8.15 (m, 2H), 8.84 (s, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 145.0 (2C), 143.0 (2C), 130.1 (2C), 129.5 (2C) ppm.

Indole (11). The title compound was prepared according to the general procedure using 2-aminophenethyl alcohol (2744.0 mg, 20 mmol) and was purified by flash chromatography (10% ethyl acetate/petroleum ether) to yield the title compound (2319.6 mg, 99% isolated yield) as a white solid, mp 52-53 ºC. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 6.57 (ddd, $J = 3.1, 2.0, 1.0$ Hz, 1H), 7.13 (ddd, $J = 8.1, 7.1, 1.1$ Hz, 1H), 7.18-7.23 (m, 2H), 7.40 (ddd, $J = 8.1, 1.8, 8.0$ Hz, 1H), 7.66 (ddd, $J = 8.1, 1.8, 0.8$ Hz, 1H), 8.12 (br s, NH) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 135.7, 127.8, 124.1, 122.0, 120.7, 119.8, 111.0, 102.6 ppm.


The corresponding primary alcohols (1.0 mmol) and amides (1.0 mmol) were added to a solution of the ruthenium catalyst 2 (0.005 mmol, 3.86 mg) and KOH (0.1 mmol, 5.6 mg) in degassed p-
xylene (2 mL) under argon. The reaction mixture was stirred at 140 ºC for 16 h and then evaluated by TLC and GC-MS. The mixture was cooled to room temperature. The reaction crude was purified by silica gel column using petroleum ether and ethyl acetate mixtures to afford compounds 13a-h.

**N-benzylbenzamide (13a).**
White solid, mp 99-101 ºC (209.1 mg, 99%) (flash chromatography 50% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (400 MHz, CDCl}_3\] : \( \delta \) 4.60 (d, \( J = \) 5.8 Hz, 2H), 6.76 (br s, 1H), 7.25-7.34 (m, 5H), 7.39 (tt, \( J = \) 8.4, 1.4 Hz, 2H), 7.45-7.50 (m, 1H), 7.77-7.80 (m, 2H) ppm. 

\[ ^13C \text{NMR (100 MHz, CDCl}_3\] : \( \delta \) 167.4, 138.2, 134.3, 131.4, 128.6 (2C), 128.4 (2C), 127.8 (2C), 127.4, 126.9 (2C), 44.0 ppm.

**N-(4-fluorobenzyl)benzamide (13b).**
White solid, mp 113-115 ºC (226.9 mg, 99%) (flash chromatography 50% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (500 MHz, CDCl}_3\] : \( \delta \) 4.53 (d, \( J = \) 5.8 Hz, 2H), 6.95-7.00 (m, 3H), 7.25 (dd, \( J = \) 8.5, 5.5 Hz, 2H), 7.37 (t, \( J = \) 7.7 Hz, 1H), 7.47 (tt, \( J = \) 7.7, 1.1 Hz, 1H), 7.78 (d, \( J = \) 7.0 Hz, 2H) ppm. 

\[ ^13C \text{NMR (125 MHz, CDCl}_3\] : \( \delta \) 167.4, 162.0 (d, \( J_{C-F} = \) 245.5 Hz), 134.12, 134.06 (d, \( J_{C-F} = \) 3.3 Hz), 131.5, 129.3 (d, \( J_{C-F} = \) 8.1 Hz), 128.4 (2C), 126.9 (2C), 115.4 (d, \( J_{C-F} = \) 21.5 Hz), 43.2 ppm. 

**N-(4-methoxybenzyl)benzamide (13c).**
White solid, mp 95-96º (219.6 mg, 91%) (flash chromatography 55% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (400 MHz, CDCl}_3\] : \( \delta \) 3.76 (s, 3H), 4.51 (d, \( J = \) 5.6 Hz, 2H), 6.84 (m, 3H), 7.23 (d, \( J = \) 8.8 Hz, 2H), 7.26-7.41 (m, 2H), 7.42-7.51 (m, 1H), 7.69-7.90 (m, 2H) ppm. 

\[ ^13C \text{NMR (100 MHz, CDCl}_3\] : \( \delta \) 167.3, 158.8, 134.3, 131.3, 130.3, 129.1 (2C), 128.4 (2C), 126.9 (2C), 113.9 (2C), 55.2, 43.4 ppm.

**N-(cyclopropylmethyl)benzamide (13d).**
White solid, mp 77-79 ºC (173.5 mg, 99%) (flash chromatography 50% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (400 MHz, CDCl}_3\] : \( \delta \) 0.19-0.27 (m, 2H), 0.47-0.55 (m, 2H), 0.98-1.09 (m, 1H), 3.28 (t, \( J = \) 6.4 Hz, 2H), 6.50 (br s, 1H), 7.39 (d, \( J = \) 7.6 Hz, 2H), 7.46 (t, \( J = \) 7.6 Hz, 1H), 7.77 (d, \( J = \) 7.6 Hz, 2H) ppm. 

\[ ^13C \text{NMR (100 MHz, CDCl}_3\] : \( \delta \) 167.4, 134.7, 131.2, 128.4 (2C), 126.8 (2C), 44.8, 10.7, 3.4 ppm.

**N-pentylbenzamide (13e).**
Pale yellow oil (141.5 mg, 74%) (flash chromatography 40% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (400 MHz, CDCl}_3\] : \( \delta \) 0.88 (t, \( J = \) 6.8 Hz, 3H), 1.27-1.39 (m, 4H), 1.54-1.64 (m, 2H), 3.37-3.44 (m, 2H), 6.38 (br s, 1H), 7.38 (t, \( J = \) 7.4 Hz, 2H), 7.42-7.51 (m, 1H), 7.75 (d, \( J = \) 7.4 Hz, 2H) ppm. 

\[ ^13C \text{NMR (100 MHz, CDCl}_3\] : \( \delta \) 167.5, 134.8, 131.2, 128.4 (2C), 126.8 (2C), 40.0, 29.3, 29.1, 22.3, 13.9 ppm.

**N-benzyl-4-methoxybenzamide (13f).**
White solid, mp 126-128 ºC (224.4 mg, 96%) (flash chromatography 50% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (400 MHz, CDCl}_3\] : \( \delta \) 3.82 (s, 3H), \( J_{C-F} = \) 1.27-1.39 (m, 4H), 1.54-1.64 (m, 2H), 3.37-3.44 (m, 2H), 6.38 (br s, 1H), 7.38 (t, \( J = \) 7.4 Hz, 2H), 7.42-7.51 (m, 1H), 7.75 (d, \( J = \) 7.4 Hz, 2H) ppm. 

\[ ^13C \text{NMR (100 MHz, CDCl}_3\] : \( \delta \) 167.0, 162.2, 138.5, 128.8 (2C), 128.7 (2C), 127.8 (2C), 127.4, 126.6, 113.7 (2C), 55.4, 44.0 ppm.

**N-benzylisonicotinamide (13g).**
White solid, mp 85-87 ºC (210.1 mg, 99%) (flash chromatography 60% ethyl acetate/petroleum ether). 

\[ ^1H \text{NMR (400 MHz, CDCl}_3\] : \( \delta \) 3.82 (s, 3H), \( J_{C-F} = \) 1.27-1.39 (m, 4H), 1.54-1.64 (m, 2H), 3.37-3.44 (m, 2H), 6.38 (br s, 1H), 7.38 (t, \( J = \) 7.4 Hz, 2H), 7.42-7.51 (m, 1H), 7.75 (d, \( J = \) 7.4 Hz, 2H) ppm. 

\[ ^13C \text{NMR (100 MHz, CDCl}_3\] : \( \delta \) 167.5, 134.8, 131.2, 128.4 (2C), 126.8 (2C), 40.0, 29.3, 29.1, 22.3, 13.9 ppm.

S10
**N-benzylpropionamide (13h).** White solid, mp 50-52 °C (161.6 mg, 99%) (flash chromatography 50% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 1.18 (t, $J = 7.6$ Hz, 3H), 2.25 (q, $J = 7.6$ Hz, 2H), 4.44 (d, $J = 5.7$ Hz, 2H), 5.75 (br s, 1H), 7.25-7.36 (m, 5H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 173.6, 138.4, 128.7 (2C), 127.8 (2C), 127.5, 43.6, 29.7, 9.8 ppm.

4. **General method for the N-alkylation of sulfonamides with alcohols.**

Characterization data for compounds 15a-j.

The corresponding primary alcohols (1 mmol) and sulfonamides (1 mmol) were added to a solution of the ruthenium catalyst 2 (0.025 mmol, 19.3 mg) and KOH (0.1 mmol, 5.6 mg) in degassed p-xylene (2 mL) under argon. The reaction mixture was stirred at 140 °C for 16 h and then evaluated by TLC and GC-MS. The mixture was cooled to room temperature. The reaction crude was purified by silica gel column using petroleum ether and ethyl acetate mixtures to afford compounds 15a-j.

**N-benzylbenzenesulfonamide (15a).** White solid, mp 78-80 °C (205.2 mg, 83%) (flash chromatography 20% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 4.15 (d, $J = 6.2$ Hz, 2H), 4.79 (t, $J = 6.2$ Hz, 1H), 7.16-7.22 (m, 2H), 7.23-7.29 (m, 3H), 7.47-7.55 (m, 2H), 7.56-7.61 (m, 1H), 7.85-7.90 (m, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 139.9, 136.1, 132.7, 129.1 (2C), 128.7 (2C), 128.1, 127.9, 127.8 (2C), 127.1 (2C), 127.0 (2C), 114.0 (2C), 55.3, 46.8 ppm.

**N-(4-methoxybenzyl)benzenesulfonamide (15b).** White solid, mp 105-107 °C (133.1 mg, 48%) (flash chromatography 30% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 3.76 (s, 3H), 4.06 (d, $J = 6.1$ Hz, 2H), 4.71 (t, $J = 6.1$ Hz, 1H), 6.78 (d, $J = 8.8$ Hz, 2H), 7.08 (d, $J = 8.8$ Hz, 2H), 7.54-7.61 (m, 1H), 7.84-7.89 (m, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 159.3, 139.9, 132.6, 129.2 (2C), 129.1 (2C), 128.1, 127.1 (2C), 114.0 (2C), 55.3, 46.8 ppm.

**N-(4-chlorobenzyl)benzenesulfonamide (15c).** White solid, mp 112-114 °C (183.1 mg, 65%) (flash chromatography 20% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 4.10 (d, $J = 6.3$ Hz, 2H), 5.00 (t, $J = 6.3$ Hz, 1H), 7.12 (d, $J = 8.4$ Hz, 2H), 7.22 (d, $J = 8.4$ Hz, 2H), 7.50 (dd, $J = 8.3$, 6.9 Hz, 2H), 7.56-7.62 (m, 1H), 7.84 (d, $J = 8.3$ Hz, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 139.8, 134.8, 133.7, 132.8, 129.16, 129.15, 128.8, 127.0, 46.5 ppm.

**N-(pentyl)benzenesulfonamide (15d).** Yellow oil (122.7 mg, 54%) (flash chromatography 10% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 0.82 (t, $J = 6.8$ Hz, 3H), 1.19-1.26 (m, 4H), 1.43 (quint, $J = 7.0$ Hz, 2H), 2.93 (td, $J = 7.0$, 6.2 Hz, 2H), 4.58 (t, $J = 6.2$ Hz, 1H), 7.47-7.59 (m, 3H), 7.85-7.88 (m, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 140.0, 133.4, 132.5, 129.0 (2C), 127.0 (2C), 43.2, 29.2, 28.6, 22.1, 13.8 ppm.

**N-Benzyl-4-methylbenzenesulfonamide (15e).** White solid, mp 161-163 °C (224.7 mg, 86%) (flash chromatography 20% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 0.82 (t, $J = 6.8$ Hz, 3H), 1.19-1.26 (m, 4H), 1.43 (quint, $J = 7.0$ Hz, 2H), 2.93 (td, $J = 7.0$, 6.2 Hz, 2H), 4.58 (t, $J = 6.2$ Hz, 1H), 7.19-7.23 (m, 2H), 7.25-7.34 (m, 5H), 7.77 (t, $J = 8.2$ Hz, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 143.4, 136.8, 136.3, 129.7 (2C), 128.6, 127.80 (2C), 127.77 (2C), 127.1, 47.2, 21.5 ppm.

**N-Benzyl-3-methoxybenzenesulfonamide (15f).** White solid, mp 80-81 °C (210.8 mg, 76%) (flash chromatography 25% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 3.82
(s, 3H), 4.15 (d, J = 6.2 Hz, 2H), 5.24 (t, J = 6.2 Hz, 1H), 7.10 (ddd, J = 8.0, 2.6, 1.1 Hz, 1H), 7.19-7.30 (m, 5H), 7.38 (t, J = 1.8, Hz, 1H), 7.41 (d, J = 8.0, Hz, 1H), 7.46 (ddd, J = 8.0, 1.8, 1.1 Hz, 1H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 159.8, 140.9, 136.2, 130.1, 128.5 (2C), 127.76 (2C), 127.74, 119.15, 119.12, 111.6, 55.5, 47.2 ppm.

$^N$-Benzyl-(phenyl)methanesulfonamide (15g).$^{31}$ White solid, mp 145-146 $^\circ$C (235.2 mg, 90%) (flash chromatography 20% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 4.05 (d, J = 6.1 Hz, 2H), 4.12 (s, 2H), 4.43 (t, J = 6.1 Hz, 1H), 7.18-7.29 (m, 10H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 136.8, 130.6 (2C), 129.2, 128.80 (2C), 128.78 (2C), 128.7, 128.1, 128.0 (2C), 59.4, 47.6 ppm.

$^N$-Benzylmethanesulfonamide (15h).$^{27}$ White solid, mp 58-61 $^\circ$C (157.4 mg, 85%) (flash chromatography 20% ethyl acetate/petroleum ether). $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 2.82 (s, 3H), 4.29 (d, J = 6.2 Hz, 2H), 4.96 (br s, 1H), 7.38-7.27 (m, 5H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 136.7, 128.8 (2C), 128.0, 127.8 (2C), 47.1, 41.0 ppm.

$^N$-(4-(diethylamino)benzyl)-4-methoxybenzenesulfonamide (15i). The title compound was prepared according to the general procedure using 4-methoxybenzenesulfonamide (3744.0 mg, 20 mmol) and 4-(diethylamino)benzyl alcohol (3580.0 mg, 20 mmol) and was purified by flash chromatography (20% ethyl acetate/petroleum ether) to yield the title compound (250.9 mg, 72% isolated yield) as a white solid, mp 83-85 $^\circ$C. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$1.12 (t, J = 7.2 Hz, 6H), 3.30 (q, J = 7.2 Hz, 4H), 3.86 (s, 3H), 3.97 (d, J = 5.9 Hz, 2H), 4.62 (br s, 1H), 6.54 (d, J = 8.8 Hz, 2H), 6.95 (d, J = 8.8 Hz, 2H), 6.98 (d, J = 8.8 Hz, 2H), 7.79 (d, J = 8.8 Hz, 2H) ppm. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$162.7, 147.3, 131.5, 129.2 (4C), 122.3, 114.1 (2C), 111.6 (2C), 55.5, 46.9, 44.2 (2C), 12.4 (2C) ppm. IR (film): 3271, 2976, 1615, 1524, 1314, 1258, 1147, 1042, 890, 837, 802, 557 cm$^{-1}$. HRMS (ESI/TOF) m/z: calcd for C$_{28}$H$_{32}$SO$_3$N$_2$ ([M+H]$^+$), 349.1586; found, 349.1609.
5. NMR spectrum

$^1$H NMR (400 MHz, CDCl$_3$) for 6a
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6a
$^1$H NMR (400 MHz, CDCl$_3$) for 6b
$^{13}\text{C} \text{ NMR (100 MHz, CDCl}_3\text{) for 6b}$
$^1H$ NMR (400 MHz, CDCl$_3$) for 6c

![NMR Spectrum Image]
$^1$H NMR (400 MHz, CDCl$_3$) for 6d
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6d
$^1$H NMR (400 MHz, CDCl$_3$) for 6e
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6e
$^{19}\text{F NMR (376 MHz, CDCl}_{3}\text{) for 6e}$
\[ ^1\text{H NMR (400 MHz, CDCl}_3\text{)} \text{ for 6f} \]
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6f
$^1$H NMR (400 MHz, CDCl$_3$) for 6g
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6g
$^1$H NMR (500 MHz, CDCl$_3$) for $6$
$^{13}$C NMR (125 MHz, CDCl$_3$) for 6i
$^1$H NMR (400 MHz, CDCl$_3$) for 6j
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6j
$^1$H NMR (400 MHz, CDCl$_3$) for 6k
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6I
$^1$H NMR (400 MHz, CDCl$_3$) for 6m
$^{13}\text{C NMR (100 MHz, CDCl}_3\text{) for 6m}$
\(^1\)H NMR (400 MHz, CDCl\(_3\)) for 6n
$^{13}\text{C NMR (100 MHz, CDCl}_3\text{) for 6n}$
$^1$H NMR (400 MHz, CDCl$_3$) for 6a
$^{13}$C NMR (100 MHz, CDCl$_3$) for 6p
$^1$H NMR (400 MHz, CDCl$_3$) for 8a
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8a
$\text{NH}$

$\text{Br}$

$^1\text{H NMR}$ (400 MHz, CDCl$_3$) for 8b
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8b
$^1$H NMR (400 MHz, CDCl$_3$) for 8c
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8c
$^1$H NMR (400 MHz, CDCl$_3$) for 8d
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8d
^H NMR (400 MHz, CDCl_3) for 8e
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8e
$^1$H NMR (400 MHz, CDCl$_3$) for 8f
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8f
$^1$H NMR (400 MHz, CDCl$_3$) for 8$^g$
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8g
$^1$H NMR (400 MHz, CDCl$_3$) for 8h
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8h
$^1$H NMR (400 MHz, CDCl$_3$) for 8i
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8i
$^1$H NMR (400 MHz, CDCl$_3$) for 8j
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8k
$^1$H NMR (400 MHz, CDCl$_3$) for 8I
$^{13}$C NMR (100 MHz, CDCl$_3$) for 81
"$^{1}$H NMR (500 MHz, CDCl$_3$) for 8m"
$^{13}$C NMR (125 MHz, CDCl$_3$) for 8m
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8n
$^1$H NMR (500 MHz, CDCl$_3$) for 8a
$^1$H NMR (400 MHz, CDCl$_3$) for 8p

MeO

[Chemical structure image]
$^1$H NMR (100 MHz, CDCl$_3$) for 8p
$^1$H NMR (400 MHz, CDCl$_3$) for 8q
$^{13}$C NMR (100 MHz, CDCl$_3$) for 8q
$^{13}$C NMR (100 MHz, CDCl$_3$) for 9
$^1$H NMR (400 MHz, CDCl$_3$) for 10a
$^{13}$C NMR (100 MHz, CDCl$_3$) for 10a
$^1$H NMR (400 MHz, CDCl$_3$) for 10b
$^{13}$C NMR (100 MHz, CDCl$_3$) for 10b
$^1$H NMR (400 MHz, CDCl$_3$) for 11
\(^{13}\text{C NMR (100 MHz, CDCl}_3\text{)}\) for 11
$^1$H NMR (400 MHz, CDCl$_3$) for 13a
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13a

The diagram shows a chemical structure and a graph with various peaks, indicating the chemical shifts observed in the NMR experiment.
$^1$H NMR (400 MHz, CDCl$_3$) for 13b
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13b
$^{19}F$ NMR (376 MHz, CDCl$_3$) for 13b
$^1$H NMR (400 MHz, CDCl$_3$) for 13c
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13c
$^1$H NMR (400 MHz, CDCl$_3$) for 13d
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13d
$^1$H NMR (400 MHz, CDCl$_3$) for 13e
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13e
$^1$H NMR (400 MHz, CDCl$_3$) for 13f
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13g
$^1$H NMR (400 MHz, CDCl$_3$) for 13h
$^{13}$C NMR (100 MHz, CDCl$_3$) for 13h
$^1$H NMR (400 MHz, CDCl$_3$) for 15a
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15a
$^1$H NMR (400 MHz, CDCl$_3$) for 15b
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15b
$^1$H NMR (400 MHz, CDCl$_3$) for 15c
$^{13}$C NMR (400 MHz, CDCl$_3$) for **15c**
$^1$H NMR (400 MHz, CDCl$_3$) for 15d
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15d
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15e
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15f
1H NMR (400 MHz, CDCl3) for 15g
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15g
$^1$H NMR (400 MHz, CDCl$_3$) for 15h
$^{13}\text{C NMR (100 MHz, CDCl}_3\text{) for 15h}$
$^1$H NMR (400 MHz, CDCl$_3$) for 15i
$^{13}$C NMR (100 MHz, CDCl$_3$) for 15i
6. References


