Supporting Information

The reaction of 2-(acylamino)benzonitriles with primary aromatic amines: a convenient synthesis of 2-substituted 4-(arylamino)quinazolines

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Table of contents

<table>
<thead>
<tr>
<th>Table of contents</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>General experimental details</td>
<td>S2</td>
</tr>
<tr>
<td>Experimental procedures for compounds 2, 3 and 4</td>
<td>S3</td>
</tr>
<tr>
<td>NMR Spectra ((^1)H and (^{13})C NMR) for compounds 2a-h</td>
<td>S11</td>
</tr>
<tr>
<td>NMR Spectra ((^1)H and (^{13})C NMR) for compounds 3a-l</td>
<td>S20</td>
</tr>
<tr>
<td>NMR Spectra ((^1)H and (^{13})C NMR) for compounds 4a-c</td>
<td>S32</td>
</tr>
</tbody>
</table>
**General experimental details:**

All compounds were fully characterized by elemental analysis and spectroscopic data. The NMR spectra were recorded at room temperature, on a Varian Unity Plus (\(^{1}\text{H}: 300 \text{ MHz}, ^{13}\text{C}: 75 \text{ MHz}\)), or on a Bruker Avance III 400 (\(^{1}\text{H}: 400 \text{ MHz}, ^{13}\text{C}: 100 \text{ MHz}\)) including the \(^{1}\text{H}-^{13}\text{C}\) correlation spectra (HMQC and HMBC). Deuterated DMSO was used as solvent. The chemical shifts are expressed in \(\delta\) (ppm) and the coupling constants, \(J\), are reported in hertz (Hz). The peak patterns are indicated as follows: s, singlet; d, doublet; t, triplet; m, multiplet; q, quartet and br, broad. IR spectra were recorded on a FT-IR Bomem MB 104 using Nujol mulls and NaCl cells. The reactions were monitored by thin layer chromatography (TLC) using silica gel 60 F_{254} (Merck and Macherey-Nagel). The melting points were determined on a Stuart SMP3 melting point apparatus and are uncorrected. Elemental analyses were performed on a LECO CHNS-932 instrument.
Experimental procedures for compounds 2, 3 and 4

**Synthesis of N-(2-cyanophenyl)acetamide (2a)**

\[ \text{NH} \quad \text{CN} \quad \text{O} \]
\[
\text{O} \quad \text{CH}_3
\]

Triethylamine (40 μl) was added to a solution of anthranilonitrile 1a (0.43 g; 3.64 mmol) in acetic anhydride (1.49 g; 14.56 mmol; 1375 μl; 4 equiv) at room temperature. After 10 min a white solid precipitated and was filtered and washed with water leading to the pure product N-(2-cyanophenyl)acetamide 2a (0.51 g; 3.14 mmol; 86%).

**Synthesis of N-(5-chloro-2-cyanophenyl)acetamide (2b)**

\[ \text{O} \quad \text{CH}_3
\]
\[
\text{Cl} \quad \text{NH} \quad \text{CN}
\]

Acetic anhydride (2.72 g; 26.60 mmol; 2510 μl; 4 equiv) was added to 2-amino-4-chlorobenzonitrile 1b (1.02 g; 6.65 mmol), followed by triethylamine (40 μl). The yellow suspension was stirred at room temperature. After 16 h, the starting materials were no longer present (evidence by TLC). The yellow solid was filtered and washed with water. The product was identified as N-(5-chloro-2-cyanophenyl)acetamide 2b (1.25 g; 6.40 mmol; 96%).

**Synthesis of N-(2-cyanophenyl)benzamide (2c)**

\[ \text{O} \quad \text{Ph} \]
\[
\text{NH} \quad \text{CN}
\]

Benzoic anhydride (0.95 g; 4.20 mmol; 2.1 equiv) and triethylamine (40 μl) were added to a solution of anthranilonitrile 1a (0.23 g; 1.98 mmol) in acetonitrile (3 ml), at room temperature. The solution was reflux for 2 h. The solvent was evaporated in the rotary evaporator and the oil was cooled in an ice bath. After 2 h, the white solid (0.08 g) was filtered and washed with diethyl ether. Removal of the solvent from the mother liquor and addition of diethyl ether led to the isolation of a second crop of white solid (0.18 g). Both products were identical by IR spectroscopy and were combined and identified as N-(2-cyanophenyl)benzamide 2c (0.26 g; 1.17 mmol; 59%).

**Synthesis of (2Z)-4-[(2-cyanophenyl)amino]-4-oxobut-2-enoic acid (2d)**

\[ \text{O} \quad \text{OH} \]
\[
\text{NH} \quad \text{CN}
\]

Maleic anhydride (1.92 g; 19.56 mmol; 2 equiv) was added to a solution of anthranilonitrile 1a (1.16 g; 9.78 mmol) in acetonitrile (3 ml). Triethylamine (40 μl) was also added and the yellow solution immediately evolved
to an orange solution. The reaction mixture was stirred at room temperature. After 45 min the white precipitate was filtered and washed with water. The pure product was identified as (2Z)-4-[(2-cyanophenyl)amino]-4-oxobut-2-enoic acid 2d (1.73 g; 8.02 mmol; 82%).

**Synthesis of ethyl (2-cyanophenyl)carbamate (2e)**

![Formula Image](image)

Ethyl chloroformate (0.46 g; 4.24 mmol; 404 μl; 2 equiv) was added to a yellow solution of anthraniloneitrile 1a (0.25 g; 2.12 mmol) in water (2 ml) and ethanol (2 ml). The reaction mixture was stirred at room temperature for 1.5 h. The white precipitate was filtered and washed with water. The pure product was identified as ethyl (2-cyanophenyl)carbamate 2e (0.35 g; 1.86 mmol; 88%).

**Synthesis of ethyl 5-chloro-2-cyanophenylcarbamate (2f)**

![Formula Image](image)

Ethyl chloroformate (0.18 g; 1.65 mmol; 160 μl; 1 equiv) was added to a yellow suspension of 2-amino-4-chlorobenzonitrile 1b (0.25 g; 1.65 mmol) in water (2 ml) and ethanol (2 ml). The reaction mixture was heated under reflux leading immediately to a yellow solution. Ethyl chloroformate was added in 1 M equiv portions every 15 min for a total of 2 h. A solid precipitated upon addition of water and was filtered and washed with HCl 6M and water leading the pure product ethyl 5-chloro-2-cyanophenylcarbamate 2f (0.29 g; 1.38 mmol; 79%).

**Synthesis of N-(2-cyanophenyl)formamide (2g)**

![Formula Image](image)

A solution of anthraniloneitrile 1a (0.20 g; 1.70 mmol) in formic acid (0.26 g; 5.10 mmol; 215 μl; 3 equiv) was stirred at room temperature. After 3.5 h a white solid precipitated and was filtered and washed with ethanol. The pure product was identified as N-(2-cyanophenyl)formamide 2g (0.08 g; 0.55 mmol; 33%). The solid that precipitated from the mother liquor was a mixture of product 2g and 2-(4-oxoquinazolin-3(4H)-yl)benzonitrile in a 1.3:1 molar ratio, by 1H NMR.

**Synthesis of 1-(2-cyanophenyl)-3-phenylurea (2h)**

![Formula Image](image)
Phenylisocyanate (0.33 g; 2.76 mmol; 300 μl; 1.1 equiv) was added to a yellow solution of anthranilonitrile 1a (0.30 g; 2.51 mmol) in acetonitrile (1 ml), under nitrogen atmosphere and the mixture was stirred at room temperature. After 2 h, the yellow solution was kept at -18 °C overnight. The solid precipitate (0.03 g) was filtered and washed with acetonitrile. A second crop (0.29 g) was isolated from the mother liquor upon addition of acetonitrile. The two crops were combined as they were pure by TLC and were identified as 1-(2-cyanophenyl)-3-phenylurea 2h (0.32 g; 1.33 mmol; 53%).

**Reaction of 2a and 4-methoxyaniline leading to N-(4-methoxyphenyl)-2-methylquinazolin-4-amine (3a) (lit.3)**

[Diagram of 3a]

**Method A:** 4-Methoxyaniline (0.06 g; 0.47 mmol; 1.1 equiv) and TFA (1 equiv; 35 μl) were added to a solution of N-(2-cyanophenyl)acetamide 2a (0.07 g; 0.43 mmol) in ethanol (5 ml). The brown solution was refluxed for 46 h. The solution was kept at -18 °C and after 2.5 days the yellow solid precipitate was filtered and washed with diethyl ether. The product was identified as the trifluoroacetate salt of N-(4-methoxyphenyl)-2-methylquinazolin-4-amine 3a (0.09 g; 0.24 mmol; 56%). Removal of the solvent from the mother liquor led to a mixture of the starting material 2a and product 3a (1.6:1, molar ratio), identified by 1H NMR.

**Method B:** A grey suspension of N-(2-cyanophenyl)acetamide 2a (0.07 g; 0.45 mmol) and 4-methoxyaniline (0.07 g; 0.50 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 0.3 ml) was refluxed for 45 min. After cooling, a greenish solid precipitate was filtered and washed with diethyl ether. The product was identified as N-(4-methoxyphenyl)-2-methylquinazolin-4-amine 3a (0.06 g; 0.23 mmol; 51%).

**Method C:** A brown suspension of N-(2-cyanophenyl)acetamide 2a (0.05 g; 0.34 mmol) and 4-methoxyaniline (0.05 g; 0.37 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 230 μl) was stirred at 80 °C, for 14 h. Diethyl ether was added to the cold reaction mixture leading to an off-white solid precipitate. The solid was filtered and washed with diethyl ether. The product was identified as N-(4-methoxyphenyl)-2-methylquinazolin-4-amine 3a (0.03 g; 0.10 mmol; 29%), by 1H NMR. Removal of the solvent from the mother liquor and addition of water led to the isolation of a second crop of solid. The product was identified as 3-(4-methoxyphenyl)-2-methylquinazolin-4(3H)-one 4a (0.01 g; 0.04 mmol; 10%), by 1H NMR.

**Reaction of 2a and 4-fluoroaniline leading to N-(4-fluorophenyl)-2-methylquinazolin-4-amine (3b)**

[Diagram of 3b]

**Method A:** 4-Fluoroaniline (0.08 g; 0.70 mmol; 67 μl; 1.1 equiv) and TFA (1 equiv; 35 μl) were added to a yellow solution of N-(2-cyanophenyl)acetamide 2a (0.10 g; 0.64 mmol) in ethanol (2 ml). The reaction mixture was refluxed for 9 h. The solution was kept at -18 °C overnight (16 h) and the yellow solid precipitate was filtered and washed with diethyl ether. The product was identified as the trifluoroacetate salt of N-(4-fluorophenyl)-2-methylquinazolin-4-amine 3b (0.15 g; 0.41 mmol; 64%).
Method B: A yellow solution of N-(2-cyanophenyl)acetamide 2a (0.07 g; 0.44 mmol) and 4-fluoroaniline (0.05 g; 0.48 mmol; 46 μl; 1.1 equiv) in acetic acid (11.6 equiv; 0.3 ml) was refluxed for 2 h. A saturated aqueous solution of NaHCO₃ was added to the reaction mixture. The emulsion was extracted with dichloromethane (3x 5 ml). The organic layer was dried and concentrated in the rotary evaporator. Addition of diethyl ether led to a yellow solid precipitate that was filtered and washed with diethyl ether. The product was identified as N-(4-fluorophenyl)-2-methylquinazolin-4-amine 3b (0.02 g; 0.08 mmol; 18%). The solid that precipitated from the mother liquor was a mixture of starting material 2a (45%), product 3b (14%), 4c (16%) and a compound tentatively identified as quinazoline 4 (R₃= 4-FC₆H₄, 25%) identified by ¹H NMR.

Reaction of 2a and 4-aminophenol leading to 4-(2-methylquinazolin-4-ylamino)phenol (3c)

Method A: 4-Aminophenol (0.05 g; 0.45 mmol; 1.1 equiv) and TFA (1 equiv; 25 μl) were added to a solution of N-(2-cyanophenyl)acetamide 2a (0.07 g; 0.41 mmol) in ethanol (2 ml). The brown solution was refluxed for 17 h. The solution was kept at -18 ºC overnight (17 h) and the yellow solid precipitate was filtered and washed with diethyl ether. The product was identified as the trifluoroacetate salt of 4-(2-methylquinazolin-4-ylamino)phenol 3c (0.10 g; 0.27 mmol; 66%).

Method B: A brown solution of N-(2-cyanophenyl)acetamide 2a (0.05 g; 0.32 mmol) and 4-aminophenol (0.04 g; 0.35 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 220 μl) was stirred at 60 ºC for 21 h. Addition of water led to a dark solid suspension that was filtered and washed with water (0.02 g). The solid was identified by ¹H NMR as a mixture of 3c and 4b, in a 1:6.1 molar ratio, slightly contaminated with other unidentified side-products. Removal of the solvent from the mother liquor and addition of water led to the isolation of a second crop of greenish solid (0.04 g). The product was identified as 4-(2-methylquinazolin-4-ylamino)phenol 3c (0.04 g; 0.16 mmol; 50%).

Reaction of 2a and 4-bromoaniline leading to N-(4-bromophenyl)-2-methylquinazolin-4-amine (3d)

4-Bromoaniline (0.13 g; 0.77 mmol; 1.1 equiv) and TFA (1 equiv; 25 μl) were added to a yellow solution of N-(2-cyanophenyl)acetamide 2a (0.11 g; 0.70 mmol) in ethanol (2 ml). The solution was refluxed for 9.5 h. The solution was kept at -18 ºC overnight (16 h) and the yellow precipitate was filtered and washed with diethyl ether. The product was identified as the trifluoroacetate salt of N-(4-bromophenyl)-2-methylquinazolin-4-amine 3d (0.14 g; 0.33 mmol; 47%).

Reaction of 2a and 3-bromoaniline leading to N-(3-bromophenyl)-2-methylquinazolin-4-amine (3e)
Method A: 3-Bromoaniline (0.29 g; 1.70 mmol; 185 μl; 1.2 equiv) and TFA (1 equiv; 109 μl) were added to a colorless solution of \(N\)-(2-cyanophenyl)acetamide 2a (0.22 g; 1.42 mmol) in ethanol (3 ml). The reaction mixture was refluxed and after 22 h, the yellow solid that precipitated was filtered and washed with diethyl ether. The product was identified as the trifluoroacetate salt of \(N\)-(3-bromophenyl)-2-methylquinazolin-4-amine 3e (0.07 g; 0.16 mmol; 11%). The solid that precipitated from the mother liquor was identified as the starting material 2a (13%), by IR.

Method B: A yellow solution of \(N\)-(2-cyanophenyl)acetamide 2a (0.18 g; 1.17 mmol) and 3-bromoaniline (0.24 g; 1.40 mmol; 152 μl; 1.2 equiv) in acetic acid (11.6 equiv; 0.8 ml) was refluxed for 2.5 h. Addition of water led to a yellow precipitate that was filtered and washed with water. The product was identified as \(N\)-(3-bromophenyl)-2-methylquinazolin-4-amine 3e (0.07g; 0.24 mmol; 21%).

Reaction of 2a and 4-chloroaniline leading to \(N\)-(4-chlorophenyl)-2-methylquinazolin-4-amine (3f)

A grey solution of \(N\)-(2-cyanophenyl)acetamide 2a (0.11 g; 0.67 mmol) and 4-chloroaniline (0.09 g; 0.74 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 0.5 ml) was refluxed for 2 h. Addition of water led to a yellow solid that was filtered and washed with water. The solid (0.09 g) was identified as a complex mixture, by \(^1\)H NMR. A second crop (0.05 g) was isolated from the mother liquor upon addition of NaHCO₃ (0.36 g) and was filtered and washed with water. The product was identified as \(N\)-(4-chlorophenyl)-2-methylquinazolin-4-amine 3f (0.05g; 0.19 mmol; 28%).

Reaction of 2b and 4-methoxyaniline leading to 7-chloro-\(N\)-(4-methoxyphenyl)-2-methylquinazolin-4-amine (3g)

Method A: 4-Methoxyaniline (0.18 g; 1.46 mmol; 2 equiv) was added to a white suspension of \(N\)-(5-chloro-2-cyanophenyl)acetamide 2b (0.14 g; 0.73 mmol) in ethanol (5 ml), leading to a brown suspension. TFA (1 equiv; 56 μl) was added and the suspension was refluxed leading immediately to a brown solution. After 72 h, the yellow solid precipitate was filtered and washed with diethyl ether and dichloromethane, leading to a pure product identified as the trifluoroacetate salt of 7-chloro-\(N\)-(4-methoxyphenyl)-2-methylquinazolin-4-amine 3g (0.07 g; 0.17 mmol; 23%).
Method B: A beige suspension of \( N\)-(5-chloro-2-cyanophenyl)acetamide 2b (0.07 g; 0.37 mmol) and 4-methoxyaniline (0.05 g; 0.41 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 0.3 ml) was refluxed for 2 h. Addition of water led to a beige precipitate that was filtered and washed with water and ethanol (1:1). The product was identified as 7-chloro-\( N\)-(4-methoxyphenyl)-2-methylquinazolin-4-amine 3g (0.02 g; 0.07 mmol; 19%). The solid that precipitated from the mother liquor was identified as a mixture of product 3g and a compound tentatively identified as 4 (R\(_3\) = 4-OMeC\(_6\)H\(_4\)) in a 1.7:1 molar ratio, by \( ^1\)H NMR.

Reaction of 2c and 4-methoxyaniline leading to \( N\)-(4-methoxyphenyl)-2-phenylquinazolin-4-amine (3h)

4-Methoxyaniline (0.04 g; 0.35 mmol; 1.1 equiv) was added to a suspension of \( N\)-(2-cyanophenyl)benzamide 2c (0.07 g; 0.31 mmol) in ethanol (2 ml) leading to an orange solution. TFA (1 equiv; 25 μl) was added and the solution was refluxed for 13.5 h. The yellow precipitate was filtered and washed with ethanol. The product was identified as the trifluoroacetate salt of \( N\)-(4-methoxyphenyl)-2-phenylquinazolin-4-amine 3h (0.06 g; 0.14 mmol; 45%).

Reaction of 2c and 4-chloroaniline leading to \( N\)-(4-chlorophenyl)-2-phenylquinazolin-4-amine (3i)

4-Chloroaniline (0.05 g; 0.41 mmol; 1.2 equiv) was added to a yellow solution of \( N\)-(2-cyanophenyl)benzamide 2c (0.08 g; 0.34 mmol) in ethanol (10 ml). TFA (1 equiv; 30 μl) was added and the solution was refluxed for 50 h. The solvent was removed in the rotary evaporator and diethyl ether was added to the oil leading to a solid precipitate. The yellow solid was filtered and washed with petroleum ether. The product was identified as the trifluoroacetate salt of \( N\)-(4-chlorophenyl)-2-phenylquinazolin-4-amine 3i (0.02 g; 0.06 mmol; 18%). The green solid (0.12 g) that precipitated from the mother liquor was mainly the amine contaminated with traces of starting material 2c and other unidentified by-products.

Reaction of 2h and 4-methoxyaniline leading to 4-(4-methoxyphenylamino)quinazolin-2(1H)-one (3j)
Method A: 4-Methoxyaniline (0.07 g; 0.51 mmol; 1.2 equiv) was added a solution of 1-(2-cyanophenyl)-3-phenylurea 2h (0.11 g; 0.46 mmol) in ethanol (10 ml), leading to an orange solution. TFA (1 equiv; 36 μl) was added and the solution was refluxed for 72 h. After 18 h a deep-red solution was obtained. The solution was kept at -18 ºC for 1 day and the white solid precipitate was filtered and washed with n-hexane. The product was identified as 4-(4-methoxyphenylamino)quinazolin-2(1H)-one 3j (0.05 g; 0.19 mmol; 41%).

Method B: 4-Methoxyaniline (0.08 g; 0.68 mmol; 1.1 equiv) was added to a solution of ethyl (2-cyanophenyl)carbamate 2e (0.12 g; 0.62 mmol) in ethanol (5 ml), leading to an orange solution. TFA (1 equiv; 48 μl) was added and the solution was refluxed for 97 h. The solution was kept at -18 ºC for 1 day and the white solid precipitate was filtered and washed with diethyl ether. The product was identified as the trifluoroacetate salt of 4-(4-methoxyphenylamino)quinazolin-2(1H)-one 3j (0.01 g; 0.04 mmol; 7%). The solid that precipitated from the mother liquor was the starting material 2e (13%). The aromatic amine was recovered as the third crop (7%). The fourth crop was a mixture of starting material 2e and amine (1.5:1), by 1H NMR.

Reaction of 2h and 4-chloroaniline leading to 4-(4-chlorophenylamino)quinazolin-2(1H)-one (3k)

4-Chloroaniline (0.06 g; 0.45 mmol; 1.1 equiv) was added to a solution of 1-(2-cyanophenyl)-3-phenylurea 2h (0.10 g; 0.41 mmol) in ethanol (10 ml), leading to a yellow solution. TFA (1 equiv; 32 μl) was added and the solution was refluxed for 96 h. The solution was kept at -18 ºC for 5 h and the white solid precipitate was filtered and washed with n-hexane. The product was identified as 4-(4-chlorophenylamino)quinazolin-2(1H)-one 3k (0.02 g; 0.07 mmol; 17%).

Reaction of 2g and 4-methoxyaniline leading to N-(4-methoxyphenyl)quinazolin-4-amine (3l)

4-Methoxyaniline (0.11 g; 0.86 mmol; 1.1 equiv) and concentrated HCl (0.4 equiv; 25 μl) were added to a white suspension of N-(2-cyanophenyl)formamide 2g (0.11 g; 0.78 mmol) in ethanol (3 ml), leading to a yellow solution. The reaction mixture was refluxed and after 12 h, the yellow solid that precipitated was filtered and washed with diethyl ether. The product was identified as N-(4-methoxyphenyl)quinazolin-4-amine 3l (0.09 g; 0.35 mmol; 41%).

Reaction of 2a and 4-methoxyaniline leading to 3-(4-methoxyphenyl)-2-methylquinazolin-4(3H)-one (4a)
A brown solution of \(N\)-(2-cyanophenyl)acetamide 2a (0.07 g; 0.46 mmol) and 4-methoxyaniline (0.06 g; 0.51 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 0.3 ml) was refluxed for 2 h. After cooling, water was added to the reaction mixture and the yellow solid precipitate was filtered and washed with water. The product was identified as 3-(4-methoxyphenyl)-2-methylquinazolin-4\(3H\)-one 4a (0.01 g; 0.04 mmol; 9%). The mother liquor was mainly a mixture of compound 3a and acetate salt of 4-methoxyaniline (1:1 M ratio), by \(^1\)H NMR.

**Reaction of 2a and 4-aminophenol leading to 3-(4-hydroxyphenyl)-2-methylquinazolin-4(3H)-one (4b)**

A yellow solution of \(N\)-(2-cyanophenyl)acetamide 2a (0.07 g; 0.46 mmol) and 4-aminophenol (0.06 g; 0.51 mmol; 1.1 equiv) in acetic acid (11.6 equiv; 0.3 ml) was refluxed for 2 h. A saturated aqueous solution of NaHCO\(_3\) was added to the reaction mixture. The emulsion was extracted with dichloromethane (3x 5 ml). The organic layer was dried and concentrated in the rotary evaporator. Addition of diethyl ether led to a greenish precipitate that was filtered and washed with diethyl ether. The product was identified as 3-(4-hydroxyphenyl)-2-methylquinazolin-4\(3H\)-one 4b (0.01 g; 0.04 mmol; 9%).

**Reaction of 2a and 3-chloroaniline leading to 2-methylquinazolin-4\(3H\)-one (4c)**

A yellow solution of \(N\)-(2-cyanophenyl)acetamide 2a (0.11 g; 0.71 mmol) and 3-chloroaniline (0.10 g; 0.78 mmol; 82 μl; 1.1 equiv) in acetic acid (11.6 equiv; 0.5 ml) was refluxed for 3.5 h. After cooling, water was added to the reaction mixture and the white solid precipitate was filtered and washed with water. The product was identified as 2-methylquinazolin-4\(3H\)-one 4c (0.06 g; 0.37 mmol; 52%).
$^1$H and $^{13}$C NMR spectra for compounds 2a-h
$^{1}H$ and $^{13}C$ NMR spectra for compounds 3a-l
$^1$H and $^{13}$C NMR spectra for compound 4a-c