

## Synthesis and Herbicidal Activity of N-Aryl-2-heteroaryloxy-N-isopropyl acetamide

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A series of novel N-aryl-2-heteroaryloxy-N-isopropylacetamide derivatives were synthesized by multi-step reactions. Their structures were characterized by <sup>1</sup>H NMR, MS and elemental analyses. The target compounds were evaluated for their herbicidal activities against *Echinochloa crusgalli*, *Digitaria sanguinalis* Scop., *Abutilon theophrasti*, *Setaria viridis*, *Zinnia elegans* and *Acalypha australis*. The results indicated that some of the title compounds displayed excellent herbicidal activities.

**Key Words:** N-Aryl-2-heteroaryloxy-N-isopropyl acetamide, Aryloxyacetamide, Synthesis, Herbicidal activity.

## INTRODUCTION

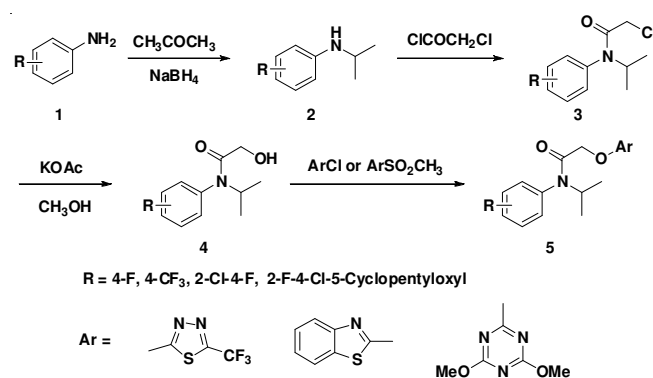
Recent years, heterocyclic compounds had received considerable attentions in medicinal and pesticidal fields<sup>1</sup>. Aryloxyacetamide derivatives<sup>2</sup> have been widely studied in recent years because of its outstanding biological activity, high selectivity, easy degradation and safety for the plants. Among them, some compounds have been developed as commercial herbicides, such as napropamide and flufenacet, which exhibited excellent activities. Furthermore, it is reported that many heterocyclic compounds with nitrogen-containing heterocycles, especially thiadiazole<sup>3</sup>, benzothiazole<sup>4</sup> and triazine<sup>5</sup>, possess a diverse range of functions. For example, thiadiazoles, benzothiazoles and triazines displayed herbicidal, fungicidal, insecticidal, anticancer and antiinflammatory activities. In view of these facts and also as a part of our work on the development of bioactive heterocyclic compounds, herein we reported the synthesis, characterization and biological of a series of aryloxyacetamide derivatives containing a thiadiazole, benzothiazole or triazine moiety.

## EXPERIMENTAL

Melting points were determined with a BÜCHI Melting Point B-450 apparatus (Büchi Labortechnik, Switzerland). The <sup>1</sup>H NMR spectra were recorded in DMSO-*d*<sub>6</sub> or CDCl<sub>3</sub> on Bruker Avance DMX 500 at 500 MHz (chemical shifts are expressed as  $\delta$  values relative to TMS as internal standard). ESI (positive) was recorded on an Esquire-LC-00075 spectrometer. Element analyses were performed on an Eager 300 instrument. All reactions were monitored by thin-layer

chromatography (TLC). All reagents were obtained from commercial sources and used without further purification unless stated.

**Synthesis of compounds:** The title compounds were synthesized according to the route as shown in Fig. 1. The intermediate **2**<sup>6</sup>, **3**<sup>7</sup>, **4**<sup>8</sup>, 2-chlorobenzo[*d*]thiazole<sup>9</sup>, 2-chloro-4,6-dimethoxy-1,3,5-triazine<sup>10</sup> and 2-(methylsulfonyl)-5-(trifluoro-methyl)-1,3,4-thiadiazole<sup>11</sup> are synthesized.

Fig. 1. Synthesis route for compounds **5a-5j**

**General procedure for the synthesis of N-aryl-N-isopropyl-2-(5-(trifluoromethyl)-1,3,4-thiadiazol-2-yloxy)-acetamide (**5a-c**):** A mixture of N-aryl-2-hydroxyl-N-isopropylacetamide (**4**) (10 mmol), 2-(methylsulfonyl)-5-(trifluoromethyl)-1,3,4-thiadiazole (10 mmol), tetraethylammonium bromide (1 mmol), anhydrous potassium carbonate (10.1 mmol) and acetone (20 mL) was stirred at room temperature

for 20 h. After that, the mixture was filtered and the filtrate was concentrated *in vacuo*. The residue was recrystallized from ethanol to afford **5a-c**.

**N-(2-Chloro-4-fluorophenyl)-N-isopropyl-2-(5-(trifluoromethyl)-1,3,4-thiadiazol-2-yloxy)acetamide (5a):** White solid, m.p. 87-89 °C, yield 61.5 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.05 (d, *J* = 7.0 Hz, 3H), 1.26 (d, *J* = 7.0 Hz, 3H), 4.69-4.88 (m, 3H), 7.13-7.17 (m, 1H), 7.33-7.35 (dd, *J* = 7.5, 2.5 Hz, 1H), 7.41-7.44 (m, 1H); MS (ESI), *m/z*: 398 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 42.29; H, 3.41; N, 10.53; found: C, 42.27; H, 3.34; N, 10.56.

**N-Isopropyl-2-(5-(trifluoromethyl)-1,3,4-thiadiazol-2-yloxy)-N-(4-(trifluoromethyl)phenyl)acetamide (5b):** White solid, m.p. 100-102 °C, yield 50.9 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.12 (d, *J* = 6.5 Hz, 6H), 4.32 (s, 2H), 4.96-5.01 (m, 1H), 7.41 (d, *J* = 8.0 Hz, 2H), 7.79 (d, *J* = 8.0 Hz, 2H); MS (ESI), *m/z*: 414 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 43.59; H, 3.17; N, 10.17; found: C, 43.74; H, 3.15; N, 10.05.

**N-(4-Chloro-5-(cyclopentylloxy)-2-fluorophenyl)-N-isopropyl-2-(5-(trifluoromethyl)-1,3,4-thiadiazol-2-yloxy)-acetamide (5c):** White solid, m.p. 79-81 °C, yield 54.7 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.05 (d, *J* = 7.0 Hz, 3H), 1.18 (d, *J* = 7.0 Hz, 3H), 1.64-1.68 (m, 2H), 1.82-1.89 (m, 6H), 3.70-3.77 (m, 2H), 4.72-4.74 (m, 1H), 4.93-4.98 (m, 1H), 6.69 (d, *J* = 7.0 Hz, 1H), 7.25 (d, *J* = 9.0 Hz, 1H); MS (ESI), *m/z*: 482 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 47.36; H, 4.18; N, 8.72; found: C, 47.18; H, 4.03; N, 8.83.

**General procedure for the synthesis of N-aryl-2-(benzo[d]thiazol-2-yloxy)-N-isopropylacetamide (5d-g):** 2-Chloro-benzo[d]thiazole (15.6 mmol) was added dropwise to a mixture of *N*-aryl-2-hydroxyl-*N*-isopropylacetamide (**4**) (15.6 mmol), potassium hydroxide (26.8 mol) and isopropanol (20 mL) at 0-5 °C. After addition, the mixture was stirred for 3 h at room temperature, then poured into ice water. The generated solid was collected by filtration and recrystallized from ethanol to get **5d-g**.

**2-(Benzo[d]thiazol-2-yloxy)-N-(4-fluorophenyl)-N-isopropylacetamide (5d):** White solid, m.p. 100-102 °C, yield 81.5 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.09 (d, *J* = 7.0 Hz, 6H), 4.71 (s, 2H), 4.90-5.05 (m, 1H), 7.17-7.22 (m, 3H), 7.27-7.29 (m, 2H), 7.33-7.36 (m, 1H), 7.62-7.64 (m, 2H); MS (ESI), *m/z*: 345 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 62.77; H, 4.98; N, 8.13; found: C, 62.98; H, 5.01; N, 7.99.

**2-(Benzo[d]thiazol-2-yloxy)-N-(2-chloro-4-fluorophenyl)-N-isopropylacetamide (5e):** White solid, m.p. 98-100 °C, yield 77.9 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.05 (d, *J* = 7.0 Hz, 3H), 1.27 (d, *J* = 7.0 Hz, 3H), 4.67-4.79 (m, 2H), 4.87-4.93 (m, 1H), 7.12-7.16 (m, 1H), 7.20-7.24 (m, 1H), 7.32-7.37 (m, 2H), 7.40-7.43 (m, 1H), 7.60-7.64 (m, 2H); MS (ESI), *m/z*: 379 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 57.07; H, 4.26; N, 7.39; found: C, 57.07.55; H, 4.27; N, 7.31.

**2-(Benzo[d]thiazol-2-yloxy)-N-isopropyl-N-(4-(trifluoromethyl)phenyl)acetamide (5f):** White solid, m.p. 112-114 °C, yield 83.3 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.13 (d, *J* = 7.0 Hz, 6H), 4.72 (s, 2H), 5.03-5.06 (m, 1H), 7.21-7.24 (m, 1H), 7.33-7.37 (m, 1H), 7.46 (d, *J* = 8.0 Hz, 2H), 7.62-7.64 (m, 2H), 7.78 (d, *J* = 8.0 Hz, 2H); MS (ESI), *m/z*: 395 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 57.86; H, 4.34; N, 7.10; found: C, 58.01; H, 4.40; N, 7.02.

**2-(Benzo[d]thiazol-2-yloxy)-N-(4-chloro-5-(cyclopentylloxy)-2-fluorophenyl)-N-isopropylacetamide (5g):** White solid, m.p. 70-72 °C, yield 74.5 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.08 (d, *J* = 7.0 Hz, 3H), 1.20 (d, *J* = 7.0 Hz, 3H), 1.63-1.68 (m, 2H), 1.86-1.95 (m, 6H), 4.72 (d, *J* = 14.0 Hz, 1H), 4.77-4.80 (m, 1H), 4.89 (d, *J* = 14.0 Hz, 1H), 4.97-5.02 (m, 1H), 6.86 (d, *J* = 7.0 Hz, 1H), 7.20-7.24 (m, 1H), 7.30-7.36 (m, 2H), 7.59-7.65 (m, 2H); MS (ESI), *m/z*: 463 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 59.67; H, 5.23; N, 6.05; found: C, 59.55; H, 5.19; N, 6.00.

**General procedure for the synthesis of N-Aryl-2-(4,6-dimethoxy-1,3,5-triazin-2-yloxy)-N-isopropylacetamide (5h-j):** A mixture of *N*-aryl-2-hydroxyl-*N*-isopropylacetamide (**4**) (7.5 mmol), 2-chloro-4,6-dimethoxy-1,3,5-triazine (7.5 mmol), anhydrous potassium carbonate (7.5 mmol) and toluene (20 mL) was refluxed for 20 h. After cooling, the mixture was filtered and the filtrate was concentrated *in vacuo*. The residue was recrystallized from acetone/hexane to get **5h-j**.

**2-(4,6-Dimethoxy-1,3,5-triazin-2-yloxy)-N-(4-fluorophenyl)-N-isopropylacetamide (5h):** White solid, m.p. 118-120 °C, yield 82.5 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.07 (d, *J* = 7.0 Hz, 6H), 4.01 (s, 6H), 4.53 (s, 2H), 4.92-4.97 (m, 1H), 7.14-7.18 (m, 2H), 7.23-7.26 (m, 2H); MS (ESI), *m/z*: 351 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 54.85; H, 5.47; N, 15.99; found: C, 54.73; H, 5.54; N, 15.82.

**2-(4,6-Dimethoxy-1,3,5-triazin-2-yloxy)-N-isopropyl-N-(4-(trifluoromethyl)phenyl)acetamide (5i):** White solid, m.p. 109-111 °C, yield 85.4 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.10 (d, *J* = 7.0 Hz, 6H), 4.02 (s, 6H), 4.52 (s, 2H), 4.92-4.97 (m, 1H), 7.43 (d, *J* = 8.0 Hz, 2H), 7.76 (d, *J* = 8.0 Hz, 2H); MS (ESI), *m/z*: 401 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 51.00; H, 4.78; N, 13.99; found: C, 50.82; H, 4.83; N, 14.06.

**N-(4-Chloro-5-(cyclopentylloxy)-2-fluorophenyl)-2-(4,6-dimethoxy-1,3,5-triazin-2-yloxy)-N-isopropylacetamide (5j):** White solid, m.p. 148-150 °C, yield 85.4 %; <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz), δ: 1.05 (d, *J* = 7.0 Hz, 3H), 1.17 (d, *J* = 7.0 Hz, 3H), 1.63-1.68 (m, 2H), 1.86-1.91 (m, 6H), 4.00 (s, 6H), 4.58-4.66 (m, 2H), 4.78-4.80 (m, 1H), 4.93-4.96 (m, 1H), 6.86 (d, *J* = 7.0 Hz, 1H), 7.25 (d, *J* = 7.0 Hz, 1H); MS (ESI), *m/z*: 469 (M+1)<sup>+</sup>. Elemental anal. (%), calculated: C, 53.79; H, 5.59; N, 11.95; found: C, 53.65; H, 5.62; N, 11.81.

#### Herbicidal activities assay

Three dicotyledonous weeds, viz., *Abutilon theophrasti*, *Zinnia elegans* and *Acalypha australis*, and three monocotyledonous crops, viz., *Echinochloa crusgalli* and *Digitaria sanguinalis* and *Setaria viridis*, were used to test the herbicidal activities of the title compounds. Acetochlor was obtained as a commercial material.

**Culture method:** The seeds were planted in 8 cm diameter plastic boxes containing artificial mixed soil. Before plant emergence, the boxes were covered with plastic film to retain moisture. Plants were grown in the green house. The fresh weight of the above ground tissues was measured 17 days after treatment. The inhibition per cent was used to describe the control efficiency of the compounds.

**Treatment:** The dosage (activity ingredient) for each compound is corresponded to 750 g/ha. The purified compounds were dissolved in 100 µL of *N,N*-dimethylformamide

TABLE-1  
 HERBICIDAL ACTIVITY OF TITLE COMPOUNDS *in vivo* (750 g/hm<sup>2</sup>)

| Compd. No. | <i>Echinochloa crusgalli</i> |       | <i>Setaria viridis</i> |       | <i>Digitaria sanguinalis</i> |       | <i>Acalypha australis</i> |       | <i>Abutilon theophrasti</i> |       | <i>Zinnia elegans</i> |       |
|------------|------------------------------|-------|------------------------|-------|------------------------------|-------|---------------------------|-------|-----------------------------|-------|-----------------------|-------|
|            | A                            | B     | A                      | B     | A                            | B     | A                         | B     | A                           | B     | A                     | B     |
| <b>5a</b>  | 68.60                        | 97.04 | 81.58                  | 97.44 | 90.18                        | 100   | 55.74                     | 100   | 56.12                       | 4.07  | 75.07                 | 54.20 |
| <b>5b</b>  | 30.00                        | 79.14 | 75.40                  | 72.74 | 73.13                        | 100   | 63.08                     | 100   | 70.85                       | 9.48  | 60.08                 | 52.67 |
| <b>5c</b>  | 23.60                        | 19.68 | 52.71                  | 25.84 | 79.12                        | 29.77 | 50.35                     | 71.21 | 56.25                       | 7.59  | 48.38                 | 49.94 |
| <b>5d</b>  | 43.96                        | 52.11 | 53.80                  | 27.74 | 62.62                        | 49.77 | 45.15                     | 62.83 | 58.79                       | 11.48 | 69.43                 | 35.22 |
| <b>5e</b>  | 33.38                        | 6.67  | 26.76                  | 66.67 | 48.08                        | 23.25 | 69.41                     | 8.33  | 39.46                       | 32.22 | 68.23                 | 39.97 |
| <b>5f</b>  | 54.71                        | 37.92 | 94.35                  | 45.17 | 82.31                        | 40.00 | 59.64                     | 16.67 | 70.48                       | 3.52  | 83.99                 | 45.96 |
| <b>5g</b>  | 19.81                        | 31.67 | 74.05                  | 29.58 | 83.10                        | 3.26  | 74.08                     | 48.48 | 28.59                       | 30.92 | 51.09                 | 25.90 |
| <b>5h</b>  | 29.70                        | 2.11  | 56.28                  | 16.74 | 56.04                        | 15.81 | 80.47                     | 56.06 | 20.32                       | 10.55 | 48.27                 | 24.97 |
| <b>5i</b>  | 36.37                        | 6.92  | 30.16                  | 20.69 | 61.05                        | 15.81 | 36.33                     | 12.88 | 26.85                       | 2.22  | 53.24                 | 28.13 |
| <b>5j</b>  | 38.22                        | 0.85  | 52.68                  | 34.12 | 22.10                        | 7.44  | 80.95                     | 2.27  | 4.73                        | 8.70  | 44.64                 | 43.67 |
| Acetochlor |                              | 98.52 |                        | 94.78 |                              | 100   |                           | 100   |                             | 12.34 |                       | 21.34 |

A = Postmergence; B = Premergence

with 0.1  $\mu$ L of Tween 20 and then prepared for spraying with a laboratory belt sprayer delivering a 750 L/ha spray volume. Compounds and Acetochlor were sprayed immediately after seed planting (premergence treatment) or after the expansion of the first true leaf (postmergence treatment). The mixture of same amount of water, *N,N*-dimethylformamide and Tween 20 was sprayed as the blank. Each treatment was triplicated. The activity numbers represented the per cent displaying herbicidal damage as compared to the blank. The error of the experiments was 2 %.

## RESULTS AND DISCUSSION

A group of 10 new aryloxyacetamide derivatives was synthesized from substituted arylamines by the process of *N*-alkylation, *N*-acylation, esterification, transesterification and condensation reactions. To reducing impurities in the process of synthesizing amide **3**, nitrogen gas was bubbled into the reaction system to remove the generated HCl, instead of using acid blinding agent such as pyridine, triethylamine, potassium carbonate or sodium hydroxide base. *N*-Aryl-2-hydroxyl-*N*-isopropylacetamide **4** was prepared by one-pot reaction by using compound **3**, potassium acetate and methanol as reactants. This method has the advantages of good yields, short reaction time and mild reaction conditions.

**Herbicidal activity:** Herbicidal activities of target compounds **5a-j** against *Echinochloa crusgalli*, *Setaria viridis*, *Digitaria sanguinalis* Scop, *Acalypha australis*, *Abutilon theophrasti* and *Zinnia elegans* were summarized in Table-1. From the Table-1, it is indicated that compounds had better activity under the premergence condition than that of postmergence. Compounds **5a** and **5b** have excellent herbicidal activities against *Acalypha australis* and *Digitaria sanguinalis* (100 %) under the premergence condition. Under the postmergence, however, same herbicidal activities were observed for compound **5a** and **5b** has moderate effects on the *Echinochloa crusgalli*, *Setaria viridis*, *Digitaria sanguinalis* Scop, *Acalypha australis*, *Abutilon theophrasti* and *Zinnia elegans*. All these compounds did not display obvious herbicidal activities against *Abutilon theophrasti* and *Zinnia elegans*. For the *Setaria viridis*, it was indicated that most of the compounds had weak herbicidal activity, except that compound **5a** (81.58 %)

and **5f** (94.35 %) displayed good control effect against *Setaria viridis*. The data given in Table-1 indicated that the change of heterocycles affects the herbicidal activity. These compounds have lower herbicidal activities with the benzo[d]-thiazole, triazine than with thiadiazole. It is benefit to further design work.

## Conclusion

A series of new aryloxyacetamides with a heterocycle moiety was designed and synthesized. Their structures were confirmed through <sup>1</sup>H NMR, ESI-MS and Elemental analysis. The herbicidal activity results show that some of these compounds had good herbicidal activity against *Echinochloa crusgalli*, *Setaria viridis*, *Digitaria sanguinalis* scop, *Acalypha australis*, *Abutilon theophrasti* and *Zinnia elegans*.

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