# Synthesis of novel 9,9-dimethyl-8,12-dihydro-9*H*-chromeno[3,2-*e*] [1,2,4] triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives

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Novel 9,9-dimethyl-8,12-dihydro-9*H*-chromeno[3,2-*e*][1,2,4]triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives were synthesised by the reactions of triethyl orthoformate or aromatic aldehydes with chromeno[2,3-*d*]pyrimidine derivatives, which were prepared by the reactions of hydrazine monohydrate and formimidate derivatives. Formimidate were synthesised by the reactions of 2-amino-7,7-dimethyl-5-oxo-4-aryl-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile derivatives and triethyl orthoformate. The structures of 12-(4-methoxyphenyl)-9,9-dimethyl-8,12-dihydro-9*H*-chromeno[3,2-*e*][1,2,4]triazolo[1,5-*c*] pyrimidin-11(10*H*)-one was determined by single crystal X-ray diffraction analysis.

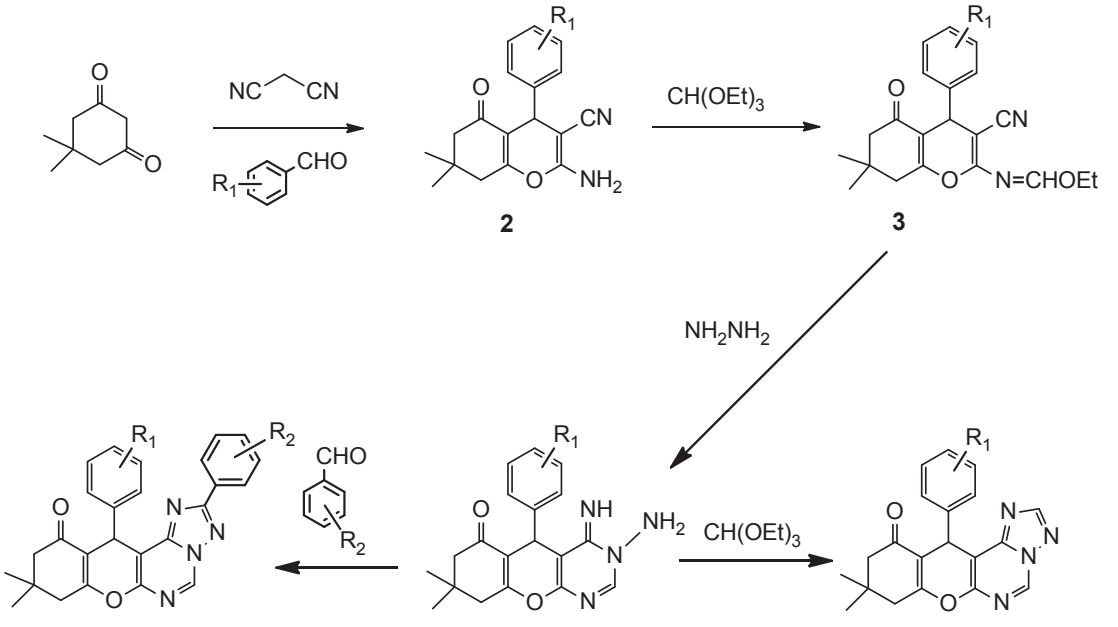
**Keywords:** chromeno[3,2-*e*][1,2,4]triazolo[1,5-*c*]pyrimidine-11(10*H*)-one, formimidate, chromeno[2,3-*d*]pyrimidine

The triazolo heterocyclic ring system has received considerable attention among synthetic chemists in recent years, because molecules bearing this feature display a wide range of biological activities in medicinal chemistry, such as antioxidant, antimalarial, anti-ocular hypertension properties.[1–4] Furthermore, 1,2,4-triazolo[1,5-*c*]pyrimidines exhibited antimicrobial activities such as antiviral, antibacterial and antithrombotic agents.[5–7] The pyrazolo[4,3-*e*] [1,2,4]triazolo[1,5-*c*]pyrimidine nucleus has been recognised as a promising template for the development of new adenosine receptor antagonists.[8] In addition, antigenotoxic activity has been found in the pyrano[3,2-*e*][1,2,4]triazolo[1,5-*c*] pyrimidine derivatives.[9]

We now report the structural modification of certain biological active heterocyclic nuclei through the study of the synthesis of several novel analogues of chromeno[3,2-*e*][1,2,4] triazolo[1,5-*c*]pyrimidine derivatives with the aim of enhancing their biological activities (Scheme 1).

## Results and discussion

The synthetic route to the title compounds are shown in Scheme 1. 2-Amino-7,7- dimethyl-5-oxo-4-aryl-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile derivatives **2** were synthesised by the reaction of 5,5-dimethylcyclohexane-1,3-dione with 2-arylylidene malononitrile derivatives **1**,[10] which were prepared by the reaction of aromatic aldehydes and malononitrile with $KF{\cdot}2H_2O$ as catalyst in ethanol.[11] Compounds **2** and excess triethyl orthoformate were heated to give ethyl *N*-(4-aryl-3-cyano-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromen-2-yl)formimidate derivatives **3**, which reacted with hydrazine monohydrate to afford 3-amino-5-aryl 4-imino-8,8-dimethyl-5,7,8,9-tetrahydro-3*H*-chromeno[2,3-*d*] pyrimidin-6(4*H*)-one derivatives **4**. 12-Aryl-9,9-Dimethyl-8,12-dihydro-9*H*-chromeno[3,2-*e*][1,2,4]triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives **5** and **6** were synthesised by the reaction of compounds **4** with excess triethyl orthoformate or aromatic aldehydes. The melting points and yields of compounds **3**, **4**, **5** and **6** are shown in Tables 1, 2, 3 and 4, respectively.

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The data of IR, $^1$H NMR, elemental analysis shown in the experimental section are in accordance with the chemical structures of the target compounds.

In the $^1$H NMR spectrum of compound **3c**, a sharp single proton peak at δ 4.46 was the characteristic absorption proton peak of 4-H. The structures of these compounds were further testified by their IR spectra through several typical absorption bands of 1624 cm$^{-1}$ for (C=N), 2220 cm$^{-1}$ for (–CN), 2931 cm$^{-1}$ for (–$CH_2$), 2980 cm$^{-1}$ for (–$CH_3$), 3053 cm$^{-1}$ for (CH-aromatic).

In the $^1$H NMR spectrum of compound **4c**, a sharp single proton peak at δ 4.66 was the characteristic absorption proton peak of 5-H. The structures of these compounds were further testified by their IR spectra through several typical absorption bands of 1660 cm$^{-1}$ for (C=O), 2959 cm$^{-1}$ for (–$CH_3$), 3190 cm$^{-1}$ for (cyclohexane–$CH_2$), and 3283 cm$^{-1}$ for (N–H).

In the $^1$H NMR spectrum of compound **5c**, a sharp single proton peak at δ 5.48 was the characteristic absorption proton peak of 12-H. The structures of these compounds were further proved by their IR spectra through several typical absorption bands of 1656 cm$^{-1}$ for (C=O), 2837, 2967 cm$^{-1}$ for (–$CH_3$), and 3083 cm$^{-1}$ for (aromatic-H).

In the $^1$H NMR spectrum of compound **6c**, a sharp single proton peak at δ 5.44 was the characteristic absorption proton peak of 12-H. The structures of these compounds were further proved by their IR spectra through several typical absorption bands of 1659 cm$^{-1}$ for (C=O), 2892, 2952 cm$^{-1}$ for (–$CH_3$), and 3118 cm$^{-1}$ for (aromatic-H).

Interaction of compounds **3** with hydrazine monohydrate in absolute ethanol gave the key intermediates compounds **4**. Note that compounds **3** can be reduced to afford the raw material **2** if excess hydrazine monohydrate 99% was used in the reaction. This may be attributed to the reaction of excess hydrazine monohydrate 99% with the iminoether **3** that took place with preferential elimination of ethyl formate hydrazone rather than cyclisation to give the pyrimidine ring.[12] At the high activity, hydrazine monohydrate was added dropwise to the reaction system; the optimal reaction temperature was 0–5 °C. If the temperature of the reaction was higher, the reaction was too intense to yield compounds **4**. Moreover, all reagents used in reaction were dried.

Originally, the final compounds **5** were obtained by reaction of compounds **4** with triethyl orthoformate (1 : 1) with piperidine or KF·2$H_2O$ as catalysts under reflux in ethanol. However, we found the yield was very low (*ca* 20%). In order to increase the productivity, we directly added excessive triethyl orthoformate at reflux. In this way, the reaction temperature was improved from 78 to 143 °C, and the yields reached up to around 80%. In addition, the reactant compounds **4** and triethyl orthoformate were all purified.

In the synthesis process of compounds **6**, a series of aromatic aldehydes were involved. However, it was found that the yield of aromatic aldehydes containing the electron-withdrawing

**Table 1** Synthesis of 4-aryl-ethyl *N*-(3-cyano-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromen-2-yl)formimidate derivatives **3a**–**g**[a]

| Derivative | R | M.p./°C | Yield/%[b] |
|---|---|---|---|
| **3a** | H | 216–218 | 72 |
| **3b** | 4-$OCH_3$ | 124–126 | 75 |
| **3c** | 3,4-$(OCH_3)_2$ | 126–128 | 70 |
| **3d** | 4-$CH_3$ | 116–118 | 80 |
| **3e** | 2,4-$Cl_2$ | 212–214 | 76 |
| **3f** | 3-$NO_2$ | 114–116 | 72 |
| **3g** | 2-Cl | 154–156 | 70 |

[a]Reaction conditions: 15 mL triethyl orthoformate, 10 mmol compounds **2**, reflux.
[b]Isolated yield.

**Table 2** Synthesis of 3-amino-4-imino-8,8-dimethyl-3,4,8,9-5-phenyl-tetrahydro-5*H*-chromeno[2,3-*d*]pyrimidin-6(7*H*)-one derivatives **4a**–**g**[a]

| Derivative | R | M.p./°C | Yield/%[b] |
|---|---|---|---|
| **4a** | H | 214–216 | 61 |
| **4b** | 4-$OCH_3$ | 178–180 | 70 |
| **4c** | 3,4-$(OCH_3)_2$ | 174–176 | 73 |
| **4d** | 4-$CH_3$ | 176–178 | 79 |
| **4e** | 2,4-$Cl_2$ | 178–180 | 75 |
| **4f** | 3-$NO_2$ | 202–204 | 76 |
| **4g** | 2-Cl | 166–168 | 72 |

[a]Reaction conditions: 10 mL ethanol, 5 mmol 99% hydrazine monohydrate, 5 mmol formimidate derivatives **3**, room temperature.
[b]Isolated yield.

**Table 3** Synthesis of 12-aryl-9,9-dimethyl-8,12-dihydro-9*H*-chromeno [3,2-*e*][1,2,4]triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives **5a**–**g**[a]

| Derivative | R | M.p./°C | Yield/%[b] |
|---|---|---|---|
| **5a** | H | 188–190 | 65 |
| **5b** | 4-$OCH_3$ | 208–210 | 72 |
| **5c** | 3,4-$(OCH_3)_2$ | 202–204 | 76 |
| **5d** | 4-$CH_3$ | 206–208 | 73 |
| **5e** | 2,4-$Cl_2$ | 222–224 | 63 |
| **5f** | 3-$NO_2$ | 214–216 | 63 |
| **5g** | 2-Cl | 238–240 | 66 |

[a]Reaction condition: 10 mL triethyl orthoformate, 5 mmol compounds **4**, reflux.
[b]Isolated yield.

**Table 4** Synthesis of 2,12-diaryl-9,9-dimethyl-8,12-dihydro-9*H*-chromeno [3,2-*e*][1,2,4]triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives **6a**–**g**[a]

| Derivative | $R^1$ | $R^2$ | M.p./°C | Yield/%[b] |
|---|---|---|---|---|
| **6a** | H | 4-$OCH_3$ | 208–210 | 70 |
| **6b** | 4-$OCH_3$ | 4-$OCH_3$ | 136–138 | 75 |
| **6c** | 4-$OCH_3$ | 3,4-$(OCH_3)_2$ | 132–134 | 73 |
| **6d** | 3,4-$(OCH_3)_2$ | 4-$OCH_3$ | 118–120 | 72 |
| **6e** | 3,4-$(OCH_3)_2$ | 3,4-$(OCH_3)_2$ | 156–158 | 77 |
| **6f** | 4-$CH_3$ | 4-$OCH_3$ | 118–120 | 68 |
| **6g** | 4-$CH_3$ | 3,4-$(OCH_3)_2$ | 214–216 | 73 |

[a]Reaction condition: 15 mL ethanol, 10 mmol compounds **4**, 10 mmol aromatic aldehydes, 1 drop piperidine, reflux.
[b]Isolated yield.

**Table 5** Crystallographic data for compound **5b**

| Empirical formula | $C_{21}H_{20}N_4O_3$ |
|---|---|
| Formula weight | 376.41 |
| Wavelength/nm | 0.71073 |
| Crystal system | Orthorhombic |
| Space group | $P2_12_12_1$ |
| a/Å | 6.898(3) |
| b/Å | 13.722(6) |
| c/Å | 19.927(8) |
| α/(°) | 90.00 |
| β/(°) | 90.00 |
| γ/(°) | 90.00 |
| Volume/Å$^3$ | 1886.4(14) |
| Z | 4 |
| Calculated density/g·cm$^{-3}$ | 1.325 |
| Absorption coefficient/mm$^{-1}$ | 0.091 |
| F (000) | 792 |
| Final R indices [I > 2s(I)] | $R^1$ = 0.0526, $wR^2$ = 0.1088 |
| R indices (all data) | $R^1$ = 0.1188, $wR^2$ = 0.1522 |

group was lower than that containing the electron-donating group (such as methoxy) in the same reaction time. It was in the presence of the electron-donating group, that the activity of aromatic aldehydes was increased. So, aromatic aldehydes containing the methoxy group were incorporated into the experiment.

## Conclusion

The present study synthesised a series of novel 9,9-dimethyl-8,12-dihydro-9*H*-chromeno[3,2-*e*][1,2,4]triazolo[1,5-*c*] pyrimidin-11(10*H*)-one derivatives from chromeno [2,3-*d*] pyrimidine in good yields in a route that is also useful for drug research. Moreover, the new synthetic route offers several advantages including simple operation, available raw materials, and good yields, which makes it an attractive process for the synthesis of the title compounds.

## Experimental

Melting points were determined by an electrothermal apparatus and the temperature was uncorrected. Microanalysis was performed by the PerkinElmer 2400 Microanalytical Service. IR spectra were determined on a PerkinElmer 1700 spectrophotometer. $^1$H NMR spectra were recorded on a Bruker ARX-400 instrument. Mass spectra were recorded by JMS-DX300 mass spectrometer with an ionisation potential of 70 eV. All reactions were monitored by TLC, which carried out on 0.2 mm silica GF254 (Merck) plates using UV light (254 and 365 nm) for detection.

*Synthesis of ethyl N-(4-aryl-3-cyano-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate derivatives (***3a**–**g***); general procedure*

The ethyl 2-amino-4-aryl-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile derivatives **2** (10 mmol) was added to excess triethyl orthoformate (15 mL) and refluxed for 6 h. The resulting dark brown solution was allowed to cool to room temperature and evaporated under reduced pressure to give a brown residue. Then the crude product was filtered off and recrystallised from ethanol.

*Synthesis of 3-amino-5-aryl-4-imino-8,8-dimethyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one derivatives (***4a**–**g***); general procedure*

Compounds **3** (5 mmol) were dissolved in ethanol (10 mL), and 99% hydrazine monohydrate (5 mmol) was added dropwise to the solution. The solution was stirred at 0–5 °C for 3 h, during which time precipitate separated out from the solution. The precipitate was filtered off and recrystallised from ethanol to afford 3-amino-5-aryl-4-imino-8,8-dimethyl-3,4,8,9-tetrahydro-5*H*-chromeno[2,3-*d*] pyrimidin-6(7*H*)-one derivatives **4** as crystalline powder.

*Synthesis of 12-aryl-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one derivatives (***5a**–**g***); general procedure*

A solution of compounds **4** (5 mmol) and triethyl orthoformate (10 mL) was heated to reflux for 6 h. After cooling to room temperature, the precipitate was filtered off and recrystallised from ethanol to yield 12-aryl-9,9-dimethyl-8,12-dihydro-9*H*-chromeno-[3,2-*e*][1,2,4] triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives **5** as crystalline powder.

*Synthesis of 2,12-diaryl-9,9-dimethyl-8,12-dihydro-9H-chromeno [3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one derivatives (***6a**–**g***); general procedure*

Piperidine (one drop) as a catalyst was added to a solution of the corresponding compounds **4** (10 mmol) and aromatic aldehydes (10 mmol) in ethanol (15 mL). The mixture was stirred at 80 °C for 4 h, the progress of the reaction was monitored by TLC. Then the resultant mixture was cooled to room temperature. The filtrate was purified by silica gel flash chromatography using ethyl acetate/petroleum ether (2 : 1) as the eluent to obtain pure 2,12-diaryl-9,9-dimethyl-8,12-dihydro-9*H*-chromeno-[3,2-*e*][1,2,4]triazolo[1,5-*c*]pyrimidin-11(10*H*)-one derivatives **6**.

*Single-crystal X-ray crystallography*

X-ray diffraction experiments were carried out using Bruker SMART APEX CCD with MoKα radiation (λ = 0.71073 Å) at 291(2) K. Data collection, cell refinement, and data reduction were performed by using the Crystal Clear software package. The structures of **5b** were solved by direct methods and refined by the full-matrix method based on F2 using the SHELXTL software package. All non-hydrogen atoms were refined anisotropically, and the positions of all hydrogen atoms were generated geometrically. Crystallographic data (excluding structure factors) for the structure in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication CCDC Nos. 1005559. Crystal data for **5b**: $C_{21}H_{20}N_4O_3$, $M_w$ = 376.41, orthorhombic $P2_12_12_1$, $a$ = 6.898(3) Å, $b$ = 13.722(6) Å, $c$ = 19.927(8) Å, V = 1886.4(14) Å$^3$, Z = 4, $D_c$ = 1.325 g cm$^{-3}$, R$^1$ (I > 2σ) = 0.0526, wR$^2$ = 0.1088, μ = 0.091 mm$^{-1}$, S = 1.000; Flack value = 0.4 (10).

The single crystal structure of **5b** was determined by single-crystal X-ray diffraction analysis (Figs 1 and 2) and a summary of the crystal data and structure refinement is presented in Table 5.

*Ethyl N-(3-cyano-7,7-dimethyl-5-oxo-4-phenyl-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3a**): Yield 72%; m.p. 216–218 °C. IR (KBr, ν, cm$^{-1}$): 3053, 2978, 2935, 2213, 1621; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.08 (s, 3H, –$CH_3$), 1.14 (s, 3H, –$CH_3$), 1.31–1.40 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.21–2.31 (m, 2H, C$^8$–H), 2.48–2.56 (m,

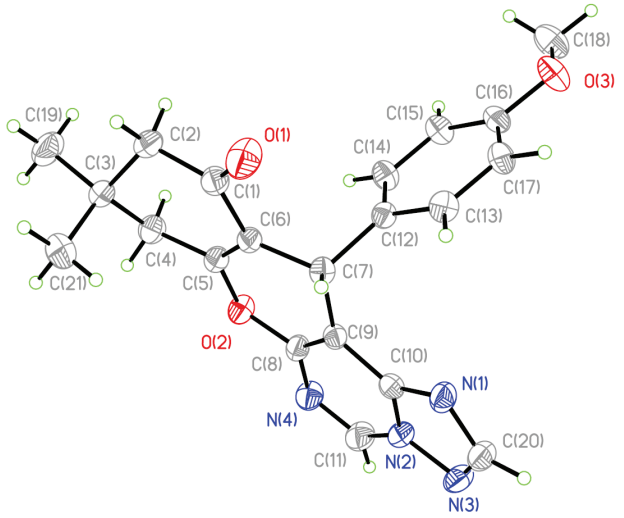


**Fig. 1** Molecular structure of compound **5b**.

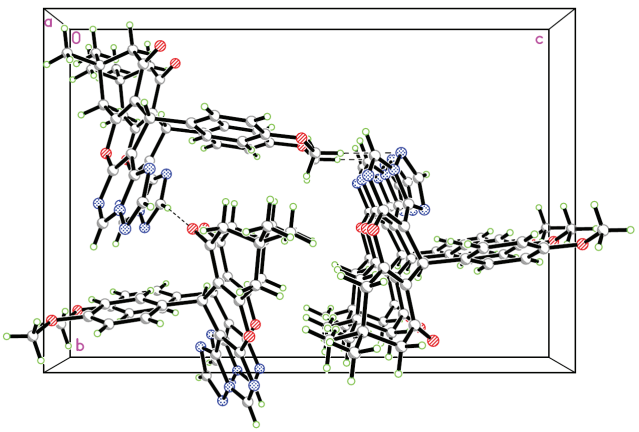


**Fig. 2** Packing diagram of compound **5b** in the unit cell.

2H, $C^6$–H), 4.38–4.44 (q, $J$ = 7.1 Hz, 2H, –$OCH_2CH_3$), 4.54 (s, 1H, $C^4$–H), 7.22–7.35 (m, 5H, Ph–H), 8.26 (s, 1H, –N=CHOEt). Anal. calcd for $C_{21}H_{22}N_2O_3$: C, 71.98; H, 6.33; N, 7.99; found C, 71.83; H, 6.20; N, 7.81%.

*Ethyl N-(3-cyano-4-(4-methoxyphenyl)-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3b**): Yield 75%; m.p. 124–126 °C. IR (KBr, ν, $cm^{-1}$): 3050, 2985, 2924, 2223, 1620; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.05 (s, 3H, –$CH_3$), 1.12 (s, 3H, –$CH_3$), 1.35–1.38 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.19–2.28 (dd, $J_1$ = 16.4 Hz, $J_2$ = 21.7 Hz, 2H, $C^8$–H), 2.48 (s, 2H, $C^6$–H), 3.77 (s, 3H, –$OCH_3$), 4.36–4.42 (q, $J$ = 7.0 Hz, 2H, –$OCH_2CH_3$), 4.47 (s, 1H, $C^4$–H), 6.82–6.85 (d, $J$ = 8.4 Hz, 2H, 3′+5′-H), 7.17–7.19 (d, $J$ = 8.5 Hz, 2H, 2′+6′-H), 7.26 (s, 1H, –N=CHOEt). Anal. calcd for $C_{22}H_{24}N_2O_4$: C, 69.46; H, 6.36; N, 7.36; found C, 69.32; H, 6.25; N, 7.30%.

*Ethyl N-(3-cyano-4-(3,4-dimethoxyphenyl)-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3c**): Yield 70%; m.p. 126–128 °C. IR (KBr, ν, $cm^{-1}$): 3053, 2980, 2931, 2220, 1624; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.08 (s, 3H, –$CH_3$), 1.13 (s, 3H, –$CH_3$), 1.35–1.38 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.21–2.30 (dd, $J_1$ = 16.5 Hz, $J_2$ = 20.1 Hz, 2H, $C^8$–H), 2.49 (s, 2H, $C^6$–H), 3.84–3.87 (d, 6H, 2×–$OCH_3$), 4.37–4.42 (q, $J$ = 7.0 Hz, 2H, –$OCH_2CH_3$), 4.46 (s, 1H, $C^4$–H), 6.76–6.82 (m, 3H, Ph–H), 8.23 (s, 1H, –N=CHOEt). Anal. calcd for $C_{23}H_{26}N_2O_5$: C, 67.30; H, 6.38; N, 6.82; found C, 67.21; H, 6.31; N, 6.90%.

*Ethyl N-(3-cyano-7,7-dimethyl-5-oxo-4-p-tolyl-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3d**): Yield 80%; m.p. 116–118 °C. IR (KBr, ν, $cm^{-1}$): 3052, 2980, 2921, 2224, 1618; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.06 (s, 3H, –$CH_3$), 1.12 (s, 3H, –$CH_3$), 1.34–1.38 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.18–2.29 (dd, $J_1$ = 16.4 Hz, $J_2$ = 22.3 Hz, 2H, $C^8$–H), 2.29 (s, 3H, –$CH_3$), 2.48 (s, 2H, $C^6$–H), 4.35–4.41 (q, $J$ = 7.1 Hz, 2H, –$OCH_2CH_3$), 4.48 (s, 1H, $C^4$–H), 7.09–7.16 (m, 4H, Ph–H), 8.23 (s, 1H, –N=CHOEt). Anal. calcd for $C_{22}H_{24}N_2O_3$: C, 72.50; H, 6.64; N, 7.69; found C, 72.38; H, 6.50; N, 7.82%.

*Ethyl N-(3-cyano-4-(2,4-dichlorophenyl)-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3e**): Yield 76%; m.p. 212–214 °C. IR (KBr, ν, $cm^{-1}$): 3047, 2981, 2934, 2218, 1622; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.07 (s, 3H, –$CH_3$), 1.12 (s, 3H, –$CH_3$), 1.35–1.38 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.18–2.28 (dd, $J_1$ = 16.4 Hz, $J_2$ = 26.2 Hz, 2H, $C^8$–H), 2.48 (s, 2H, $C^6$–H), 4.37–4.42 (q, $J$ = 7.1 Hz, 2H, –$OCH_2CH_3$), 4.94 (s, 1H, $C^4$–H), 7.20 (s, 2H, 5′+6′-H), 7.37 (s, 1H, 3′-H), 8.24 (s, 1H, –N=CHOEt). Anal. calcd for $C_{21}H_{20}Cl_2N_2O_3$: C, 60.15; H, 4.81; N, 6.68; found C, 59.92; H, 4.69; N, 6.80%.

*Ethyl N-(3-cyano-7,7-dimethyl-4-(3-nitrophenyl)-5-oxo-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3f**): Yield 72%; m.p. 114–116 °C. IR (KBr, ν, $cm^{-1}$): 3052, 2983, 2924, 2220, 1621; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.07 (s, 3H, –$CH_3$), 1.14 (s, 3H, –$CH_3$), 1.36–1.39 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.20–2.31 (dd, $J_1$ = 16.4 Hz, $J_2$ = 26.2 Hz, 2H, $C^8$–H), 2.49–2.60 (dd, $J_1$ = 17.9 Hz, $J_2$ = 24.1 Hz, 2H, $C^6$–H), 4.39–4.44 (q, $J$ = 7.1 Hz, 2H, –$OCH_2CH_3$), 4.65 (s, 1H, $C^4$–H), 7.50–7.54 (t, $J$ = 7.9 Hz, 1H, 5′-H), 7.72–7.74 (d, $J$ = 7.5 Hz, 1H, 6′-H), 8.07–8.12 (m, 2H, 2′+4′-H), 8.29 (s, 1H, –N=CHOEt). Anal. calcd for $C_{21}H_{21}N_3O_5$: C, 63.79; H, 5.35; N, 10.63; found C, 63.61; H, 5.24; N, 10.71%.

*Ethyl N-(4-(2-chlorophenyl)-3-cyano-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4H-chromen-2-yl)formimidate* (**3g**): Yield 70%; m.p. 154–156 °C. IR (KBr, ν, $cm^{-1}$): 3051, 2978, 2934, 2217, 1625; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.07 (s, 3H, –$CH_3$), 1.12 (s, 3H, –$CH_3$), 1.34–1.38 (t, $J$ = 7.1 Hz, 3H, –$OCH_2CH_3$), 2.17–2.28 (dd, $J_1$ = 16.3 Hz, $J_2$ = 25.9 Hz, 2H, $C^8$–H), 2.49 (s, 2H, $C^6$–H), 4.36–4.41 (q, $J$ = 7.0 Hz, 2H, –$OCH_2CH_3$), 4.99 (s, 1H, $C^4$–H), 7.15–7.35 (m, 4H, Ph–H), 8.24 (s, 1H, –N=CHOEt). Anal. calcd for $C_{21}H_{21}ClN_2O_3$: C, 65.54; H, 5.50; N, 7.28; found C, 65.43; H, 5.38; N, 7.41%.

*3-Amino-4-imino-8,8-dimethyl-5-phenyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4a**): Yield 61%; m.p. 214–216 °C. IR (KBr, ν, $cm^{-1}$): 3397, 3328, 3215, 2964, 1662; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.06 (s, 3H, –$CH_3$), 1.13 (s, 3H, –$CH_3$), 2.19–2.29 (dd, $J_1$ = 16.3 Hz, $J_2$ = 21.8 Hz, 2H, $C^9$–H), 2.48 (s, 2H, $C^7$–H), 4.43 (s, 1H, $C^5$–H),4.56 (s, 2H, –$NH_2$),7.22–7.33 (m, 6H, Ph+$C^2$–H). Anal. calcd for $C_{19}H_{20}N_4O_2$: C, 67.84; H, 5.99; N, 16.66; found C, 67.32; H, 6.08; N, 16.76%.

*3-Amino-4-imino-5-(4-methoxyphenyl)-8,8-dimethyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4b**): Yield 70%; m.p. 178–180 °C. IR (KBr, ν, $cm^{-1}$): 3315, 3277, 3191, 2964, 1662; $^1$H NMR ($CDCl_3$, 400 MHz) δ 0.99 (s, 3H, –$CH_3$), 1.11 (s, 3H, –$CH_3$), 2.17–2.28 (dd, $J_1$ = 16.4 Hz, $J_2$ = 27.1 Hz, 2H, $C^9$–H), 2.54 (s, 2H, $C^7$–H), 3.76 (s, 3H, –$OCH_3$), 4.68 (s, 1H, $C^5$–H), 4.76 (s, 2H, –$NH_2$), 6.80–6.82 (d, $J$ = 8.6 Hz, 2H, 3′+5′-H), 7.26–7.28 (d, $J$ = 8.6 Hz, 2H, 2′+6′-H), 8.03 (m, 1H, $C^2$–H). Anal. calcd for $C_{20}H_{22}N_4O_3$: C, 65.56; H, 6.05; N, 15.29; found C, 65.42; H, 6.25; N, 15.34%.

*3-Amino-5-(3,4-dimethoxyphenyl)-4-imino-8,8-dimethyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4c**): Yield 73%; m.p. 174–176 °C. IR (KBr, ν, $cm^{-1}$): 3283, 3190, 2959, 1660; $^1$H NMR ($CDCl_3$, 400 MHz) δ 0.99 (s, 3H, –$CH_3$), 1.11 (s, 3H, –$CH_3$), 2.18–2.28 (dd, $J_1$ = 16.4 Hz, $J_2$ = 24.5 Hz, 2H, $C^9$–H), 2.53 (s, 2H, $C^7$–H), 3.81–3.84 (d, 6H, 2×–$OCH_3$), 4.66 (s, 1H, $C^5$–H), 4.77 (s, 2H, –$NH_2$), 6.75–6.77 (d, $J$ = 8.7 Hz, 1H, 2′-H), 6.86–6.88 (d, $J$ = 6.1 Hz, 2H, 5′+6′-H), 8.03 (s, 1H, $C^2$–H). Anal. calcd for $C_{21}H_{24}N_4O_4$: C, 63.62; H, 6.10; N, 14.13; found C, 63.42; H, 6.34; N, 14.17%.

*3-Amino-5-(p-tolyl)-4-imino-8,8-dimethyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4d**): Yield 69%; m.p. 176–178 °C. IR (KBr, ν, $cm^{-1}$): 3318, 3287, 3195, 2966, 1659; $^1$H NMR ($CDCl_3$, 400 MHz) δ 0.98 (s, 3H, –$CH_3$), 1.10 (s, 3H, –$CH_3$), 2.16–2.27 (dd, $J_1$ = 16.4 Hz, $J_2$ = 28.5 Hz, 2H, $C^9$–H), 2.27 (s, 3H, 4′–$CH_3$), 2.53 (s, 2H, $C^7$–H), 4.67 (s, 1H, $C^5$–H), 4.78 (s, 2H, –$NH_2$), 7.06–7.08 (d, $J$ = 7.9 Hz, 2H, 3′+5′-H), 7.21–7.28 (d, $J$ = 8.0 Hz, 2H, 2′+6′-H), 7.99 (s, 1H, $C^2$–H). Anal. calcd for $C_{20}H_{22}N_4O_2$: C, 68.55; H, 6.33; N, 15.99; found C, 68.67; H, 6.21; N, 15.89%.

*3-Amino-5-(2,4-dichlorophenyl)-4-imino-8,8-dimethyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4e**): Yield 75%; m.p. 178–180 °C. IR (KBr, ν, $cm^{-1}$): 3335, 3293, 3195, 2957, 1663; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.02 (s, 3H, –$CH_3$), 1.14 (s, 3H, –$CH_3$), 2.17–2.31 (dd, $J_1$ = 16.4 Hz, $J_2$ = 37.1 Hz, 2H, $C^9$–H), 2.57 (s, 2H, $C^7$–H), 4.78 (s, 2H, –$NH_2$), 5.13 (s, 1H, $C^5$–H), 7.20 (s, 2H, 5′+6′-H), 7.37 (s, 1H, 3′-H), 8.04 (m, 1H, $C^2$–H). Anal. calcd for $C_{19}H_{18}Cl_2N_4O_2$: C, 56.31; H, 4.48; N, 13.82; found C, 56.43; H, 4.38; N, 13.75%.

*3-Amino-4-imino-8,8-dimethyl-5-(3-nitrophenyl)-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4f**): Yield 76%; m.p. 202–204 °C. IR (KBr, ν, $cm^{-1}$): 3331, 3290, 3188, 2957, 1659; $^1$H NMR ($CDCl_3$, 400 MHz) δ 0.99 (s, 3H, –$CH_3$), 1.13 (s, 3H, –$CH_3$), 2.19–2.30 (dd, $J_1$ = 16.4 Hz, $J_2$ = 40.9 Hz, 2H, $C^9$–H), 2.55–2.63 (dd, $J_1$ = 18.0 Hz, $J_2$ = 22.6 Hz, 2H, $C^7$–H), 4.71 (s, 2H, –$NH_2$), 4.88 (s, 1H, $C^5$–H), 7.48–7.52 (t, $J$ = 7.9 Hz, 1H, 5′-H), 7.82–7.83 (d, $J$ = 7.6 Hz, 1H, 6′-H), 8.08 (s, 1H, 4′-H), 8.10 (s, 1H, $C^2$–H), 8.16 (s, 1H, 2′-H). Anal. calcd for $C_{19}H_{19}N_5O_4$: C, 59.84; H, 5.02; N, 18.36; found C, 59.79; H, 5.12; N, 18.33%.

*3-Amino-5-(2-chlorophenyl)-4-imino-8,8-dimethyl-3,4,8,9-tetrahydro-5H-chromeno[2,3-d]pyrimidin-6(7H)-one* (**4g**): Yield 72%; m.p. 166–168 °C. IR (KBr, ν, $cm^{-1}$): 3329, 3297, 3187, 2948, 1670; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.02 (s, 3H, –$CH_3$), 1.13 (s, 3H, –$CH_3$), 2.17–2.31 (dd, $J_1$ = 16.4 Hz, $J_2$ = 43.4 Hz, 2H, $C^9$–H), 2.57 (s, 2H, $C^7$–H), 4.72 (s, 2H, –$NH_2$), 5.17 (s, 1H, $C^5$–H), 7.13–7.16 (m, 1H, 6′-H), 7.19–7.22 (m, 1H, 4′-H), 7.25–7.28 (m, 1H, 5′-H), 7.34–7.35 (d, $J$ = 7.8 Hz, 1H, 3′-H), 8.02 (s, 1H, $C^2$–H). Anal. calcd for $C_{19}H_{19}ClN_4O_2$: C, 61.54; H, 5.16; N, 15.11; found C, 61.43; H, 5.20; N, 15.07%.

*9,9-Dimethyl-12-phenyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5a**): Yield 65%; m.p. 188–190 °C. IR (KBr, ν, $cm^{-1}$): 2962, 2929, 2880, 1654; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.12 (s, 3H, –$CH_3$), 1.18 (s, 3H, –$CH_3$), 2.27–2.37 (dd, $J_1$ = 16.5 Hz, $J_2$ = 23.2 Hz, 2H, $C^8$–H), 2.66–2.76 (dd, $J_1$ = 18.1 Hz, $J_2$ = 24.1 Hz, 1H, $C^{10}$–H), 5.52 (s, 1H, $C^{12}$–H), 7.16–7.46 (m, 5H, Ph–H), 8.32 (s, 1H, $C^5$–H), 9.13 (s, 1H, $C^2$–H). Anal. calcd for $C_{20}H_{18}N_4O_2$: C, 69.35; H, 5.24; N, 16.17; found C, 69.45; H, 5.10; N, 16.27%.

*12-(4-Methoxyphenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5b**): Yield 72%; m.p. 208–210 °C. IR (KBr, ν, $cm^{-1}$): 3112, 2950, 2895, 1660; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.12 (s, 3H, –$CH_3$), 1.17 (s, 3H,–$CH_3$),

2.27–2.37 (dd, $J_1$ = 16.3 Hz, $J_2$ = 22.5 Hz, 2H, $C^8$–H), 2.64–2.75 (dd, $J_1$ = 17.7 Hz, $J_2$ = 22.7 Hz, 2H, $C^{10}$–H), 3.74 (s, 3H, 12-$OCH_3$), 5.47 (s, 1H, $C^{12}$–H), 6.79–6.81 (d, $J$ = 8.7 Hz, 2H, 12-(3′+5′)-H), 7.35–7.37 (d, $J$ = 8.7 Hz, 2H, 12-(2′+6′)-H), 8.32 (s, 1H, $C^5$–H), 9.12 (s, 1H, $C^2$–H). Anal. calcd for $C_{21}H_{20}N_4O_3$: C, 67.01; H, 5.36; N, 14.88; found C, 67.21; H, 5.21; N, 14.74%.

*12-(3,4-Dimethoxyphenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5c**): Yield 76%; m.p. 202–204 °C. IR (KBr, ν, $cm^{-1}$): 3083, 2967, 2837, 1656; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.14 (s, 3H, –$CH_3$), 1.18 (s, 3H,–$CH_3$), 2.29–2.38 (dd, $J_1$ = 16.3 Hz, $J_2$ = 20.2 Hz, 2H, $C^8$–H), 2.66–2.75 (t, $J$ = 18.5 Hz, 2H, $C^{10}$–H), 3.78 (s, 3H, 12-$OCH_3$), 3.88 (s, 3H, 12-$OCH_3$), 5.48 (s, 1H, $C^{12}$–H), 6.74–6.76 (d, $J$ = 8.3 Hz, 1H, 12-6′-H), 6.88–6.91 (m, 1H, 12-5′-H), 7.09–7.10 (d, $J$ = 2.0 Hz, 1H, 12-2′-H), 8.33 (s, 1H, $C^5$–H), 9.13 (s, 1H, $C^2$–H). Anal. calcd for $C_{22}H_{22}N_4O_4$: C, 65.01; H, 5.46; N, 13.78; found C, 65.34; H, 5.32; N, 13.65%.

*9,9-Dimethyl-12-(p-tolyl)-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5d**): Yield 73%; m.p. 206–208 °C. IR (KBr, ν, $cm^{-1}$): 3063, 2957, 2932, 2873, 1662; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.12 (s, 3H, –$CH_3$), 1.18 (s, 3H,–$CH_3$), 2.26 (s, 3H, 12-$CH_3$), 2.27–2.37 (dd, $J_1$ = 16.2 Hz, $J_2$ = 23.3 Hz, 2H, $C^8$–H), 2.65–2.75 (dd, $J_1$ = 17.7 Hz, $J_2$ = 23.1 Hz, 2H, $C^{10}$–H), 5.48 (s, 1H, $C^{12}$–H), 7.07–7.09 (d, $J$ = 7.9 Hz, 2H, 12-(3′+5′)-H), 7.33–7.35 (d, $J$ = 8.1 Hz, 2H, 12-(2′+6′)-H), 8.32 (s, 1H, $C^5$–H), 9.12 (s, 1H, $C^2$–H). Anal. calcd for $C_{21}H_{20}N_4O_2$: C, 69.57; H, 5.60; N, 15.43; found C, 69.29; H, 5.51; N, 15.27%. *12-(2,4-Dichlorophenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5e**): Yield 63%; m.p. 222–224 °C. IR (KBr, ν, $cm^{-1}$): 3056, 2958, 2931, 2873, 1660; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.12 (s, 3H, –$CH_3$), 1.17 (s, 3H,–$CH_3$), 2.25–2.36 (dd, $J_1$ = 16.4 Hz, $J_2$ = 25.6 Hz, 2H, $C^8$–H), 2.67 (s, 2H, $C^{10}$–H), 5.74 (s, 1H, $C^{12}$–H), 7.22–7.24 (m, 1H, 12-6′-H), 7.28 (s, 1H, 12-5′-H), 7.54–7.56 (d, $J$ = 8.2 Hz, 1H, 12-3′-H), 8.30 (s, 1H, $C^5$–H), 9.16 (s, 1H, $C^2$–H). Anal. calcd for $C_{20}H_{16}Cl_2N_4O_2$: C, 57.84; H, 3.88; N, 13.49; found C, 57.69; H, 3.98; N, 13.61%.

*9,9-Dimethyl-12-(3-nitrophenyl)-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5f**): Yield 63%; m.p. 214–216 °C. IR (KBr, ν, $cm^{-1}$): 3052, 2963, 2927, 2868, 1662; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.14 (s, 3H, –$CH_3$), 1.19 (s, 3H,–$CH_3$), 2.29–2.38 (dd, $J_1$ = 16.4 Hz, $J_2$ = 30.6 Hz, 2H, $C^8$–H), 2.70–2.81 (dd, $J_1$ = 17.8 Hz, $J_2$ = 38.1 Hz, 2H, $C^{10}$–H), 5.62 (s, 1H, $C^{12}$–H), 7.47–7.51 (t, $J$ = 7.9 Hz, 1H, 12-5′-H), 7.97–7.98 (d, $J$ = 7.6 Hz, 1H, 12-6′-H), 8.04–8.06 (d, $J$ = 8.1 Hz, 1H, 12-4′-H), 8.17 (s, 1H, 12-2′-H), 8.31 (s, 1H, $C^5$–H), 9.17 (s, 1H, $C^2$–H). Anal. calcd for $C_{20}H_{17}N_5O_4$: C, 61.38; H, 4.38; N, 17.89; found C, 61.45; H, 4.22; N, 17.73%.

*12-(2-Chlorophenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**5g**): Yield 66%; m.p. 238–240 °C. IR (KBr, ν, $cm^{-1}$): 3048, 2962, 2927, 2869, 1672; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.12 (s, 3H, –$CH_3$), 1.17 (s, 3H,–$CH_3$), 2.26–2.34 (dd, $J_1$ = 16.4 Hz, $J_2$ = 28.4 Hz, 2H, $C^8$–H), 2.63–2.71 (dd, $J_1$ = 18.0 Hz, $J_2$ = 20.1 Hz, 2H, $C^{10}$–H), 5.79 (s, 1H, $C^{12}$–H), 7.13–7.16 (m, 1H, 12-6′-H), 7.24–7.26 (m, 2H, 12-(4′+5′)-H), 7.60–7.61 (d, $J$ = 6.7 Hz, 1H, 12-3′-H), 8.29 (s, 1H, $C^5$–H), 9.14 (s, 1H, $C^2$–H). Anal. calcd for $C_{20}H_{17}ClN_4O_2$: C, 63.08; H, 4.50; N, 14.71; found C, 63.12; H, 4.44; N, 14.82%.

*2-(4-Methoxyphenyl)-9,9-dimethyl-12-phenyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6a**): Yield 70%; m.p. 208–210 °C. IR (KBr, ν, $cm^{-1}$): 3050, 2960, 2928, 2869, 1658; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.14 (s, 3H, –$CH_3$), 1.27 (s, 3H, –$CH_3$), 2.31–2.41 (m, 2H, $C^8$–H), 2.67–2.73 (m, 1H, $C^{10}$–H), 3.16–3.23 (m, 1H, $C^{10}$–H), 3.81 (s, 3H, –$OCH_3$), 5.42 (s, 1H, $C^{12}$–H), 6.82–6.96 (m, 2H, 2-(3′+5′)-H), 7.11–7.23 (m, 5H, Ph–H), 7.45–7.57 (m, 2H, 2-(2′+6′)-H), 8.18 (s, 1H, $C^5$–H). Anal. calcd for $C_{27}H_{24}N_4O_3$: C, 71.67; H, 5.35; N, 12.38; found C, 71.45; H, 5.43; N, 12.45%.

*2,12-bis(4-Methoxyphenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6b**): Yield 75%; m.p. 136–138 °C. IR (KBr, ν, $cm^{-1}$): 3120, 2957, 2887, 1663; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.01 (s, 3H, –$CH_3$), 1.13 (s, 3H, –$CH_3$), 2.21–2.32 (dd, $J_1$ = 16.4 Hz, $J_2$ = 26.7 Hz, 2H, $C^8$–H), 2.60 (s, 2H, $C^{10}$–H), 3.75 (s, 3H, 12-$OCH_3$), 3.86 (s, 3H, 2-$OCH_3$), 5.26 (s, 1H, $C^{12}$–H), 6.80–6.82 (d, $J$ = 8.7 Hz, 2H, 12-(3′+5′)-H), 6.93–6.95 (d, $J$ = 8.8 Hz, 2H, 2-(3′+5′)-H), 7.30–7.31 (m, 1H, 12-2′-H), 7.65–7.71 (m, 3H, 12-6′+2-(2′+6′)-H), 8.48 (s, 1H, $C^5$–H). Anal. calcd for $C_{28}H_{26}N_4O_4$: C, 69.70; H, 5.43; N, 11.61; found C, 69.83; H, 5.33; N, 11.56%.

*2-(3,4-Dimethoxyphenyl)-12-(4-methoxyphenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6c**): Yield 73%; m.p. 132–134 °C. IR (KBr, ν, $cm^{-1}$): 3118, 2952, 2892, 1659; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.00 (s, 3H, –$CH_3$), 1.36 (s, 3H,–$CH_3$), 2.21–2.32 (dd, $J_1$ = 16.4 Hz, $J_2$ = 26.2 Hz, 2H, $C^8$–H), 2.60 (s, 2H, $C^{10}$–H), 3.74 (s, 3H, 12-$OCH_3$), 3.94 (s, 3H, 2-$OCH_3$), 4.05 (s, 3H, 2-$OCH_3$), 5.44 (s, 1H, $C^{12}$–H), 6.78–6.80 (d, $J$ = 8.7 Hz, 2H, 12-(3′+5′)-H), 6.88–6.90 (d, $J$ = 8.3 Hz, 1H, 2-5′-H), 7.10–7.12 (d, $J$ = 8.3 Hz, 1H, 12-2′-H), 7.30 (s, 1H, 12-6′-H), 7.52 (s, 1H, 2-2′-H), 7.63 (s, 1H, 2-6′-H), 8.45 (s, 1H, $C^5$–H). Anal. calcd for $C_{29}H_{28}N_4O_5$: C, 67.96; H, 5.51; N, 10.93; found C, 67.76; H, 5.61; N, 10.98%.

*12-(3,4-Dimethoxyphenyl)-2-(4-methoxyphenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6d**): Yield 72%; m.p. 118–120 °C. IR (KBr, ν, $cm^{-1}$): 3080, 2961, 2841, 1662; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.03 (s, 3H, –$CH_3$), 1.15 (s, 3H, –$CH_3$), 2.24–2.34 (dd, $J_1$ = 16.3 Hz, $J_2$ = 23.8 Hz, 2H, $C^8$–H), 2.61 (s, 2H, $C^{10}$–H), 3.71 (s, 3H, 2-$OCH_3$), 3.82–3.86 (d, $J$ = 16.0 Hz, 6H, 2 × 12-$OCH_3$), 5.41 (s, 1H, $C^{12}$–H), 6.75–6.77 (d, $J$ = 8.3 Hz, 1H, 12-6′-H), 6.83–6.86 (m, 1H, 12-5′-H), 6.95–6.97 (d, $J$ = 8.8 Hz, 3H, 12-2′+2-(3′+5′)-H), 7.71–7.73 (d, $J$ = 8.6 Hz, 2H, 2-(2′+6′)-H), 8.48 (s, 1H, $C^5$–H). Anal. calcd for $C_{29}H_{28}N_4O_5$: C, 67.96; H, 5.51; N, 10.93; found C, 67.76; H, 5.65; N, 10.98%.

*2,12-bis(3,4-Dimethoxyphenyl)-9,9-dimethyl-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6e**): Yield 77%; m.p. 156–158 °C. IR (KBr, ν, $cm^{-1}$): 3073, 2972, 2841, 1658; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.02 (s, 3H, –$CH_3$), 1.14 (s, 3H, –$CH_3$), 2.22–2.32 (dd, $J_1$ = 16.4 Hz, $J_2$ = 23.5 Hz, 2H, $C^8$–H), 2.60 (s, 2H, $C^{10}$–H), 3.65 (s, 3H, 12-4′–$OCH_3$), 3.80 (s, 3H, 12-3′–$OCH_3$), 3.94 (s, 3H, 2-4′–$OCH_3$), 4.08 (s, 3H, 2-3′–$OCH_3$), 5.60 (s, 1H, $C^{12}$–H), 6.72–6.75 (d, $J$ = 8.3 Hz, 1H, 12-6′-H), 6.81–6.84 (m, 1H, 12-5′-H), 6.88–6.90 (d, $J$ = 8.3 Hz, 1H, 12-2′-H), 6.95 (s, 1H, 2-5′-H), 7.55–7.62 (d, $J$ = 24.5 Hz, 2H, 2-(2′+6′)-H), 8.43 (s, 1H, $C^5$–H). Anal. calcd for $C_{30}H_{30}N_4O_6$: C, 66.41; H, 5.57; N, 10.33; found C, 66.65; H, 5.31; N, 10.41%.

*2-(4-Methoxyphenyl)-9,9-dimethyl-12-(p-tolyl)-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6f**): Yield 68%; m.p. 118–120 °C. IR (KBr, ν, $cm^{-1}$): 3058, 2963, 2930, 2869, 1660; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.01 (s, 3H, –$CH_3$), 1.14 (s, 3H, –$CH_3$), 2.28 (m, 5H, 12-4′+$C^8$–H), 2.60 (s, 2H, $C^{10}$–H), 3.86 (s, 3H, –$OCH_3$), 5.24 (s, 1H, $C^{12}$–H), 6.93–6.95 (d, $J$ = 8.9 Hz, 2H, 12-(3′+5′)-H), 7.08–7.10 (d, $J$ = 7.9 Hz, 2H, 12-(2′+6′)-H), 7.26–7.28 (d, $J$ = 8.2 Hz, 2H, 2-(3′+5′)-H), 7.69–7.71 (d, $J$ = 8.7 Hz, 2H, 2-(2′+6′)-H), 8.49 (s, 1H, $C^5$–H). Anal. calcd for $C_{28}H_{26}N_4O_3$: C, 72.09; H, 5.62; N, 12.01; found C, 72.29; H, 5.56; N, 11.90%.

*2-(3,4-Dimethoxyphenyl)-9,9-dimethyl-12-(p-tolyl)-8,12-dihydro-9H-chromeno[3,2-e][1,2,4]triazolo[1,5-c]pyrimidin-11(10H)-one* (**6g**): Yield 73%; m.p. 214–216 °C. IR (KBr, ν, $cm^{-1}$): 3061, 2955, 2933, 2870, 1661; $^1$H NMR ($CDCl_3$, 400 MHz) δ 1.01 (s, 3H, –$CH_3$), 1.14 (s, 3H, –$CH_3$), 2.21–2.31 (m, 5H, 12-4′+$C^8$–H), 2.60 (s, 2H, $C^{10}$–H), 3.94 (s, 3H, –$OCH_3$), 4.05 (s, 3H, –$OCH_3$), 5.45 (s, 1H, $C^{12}$–H), 6.87–6.90 (d, $J$ = 8.3 Hz, 1H, 2-5′-H), 7.06–7.27 (m, 5H, (12-Ph+2-2′)-H), 7.63 (s, 1H, 2-6′-H), 8.45 (s, 1H, $C^5$–H). Anal. calcd for $C_{29}H_{28}N_4O_4$: C, 70.15; H, 5.68; N, 11.28; found C, 70.34; H, 5.48; N, 11.31%.

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