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(*E*)-2-(4-Arylbut-1-en-3-yn-1-yl)chromones as Synthons for the Synthesis of Xanthone-1,2,3-triazole Dyads

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Xanthone-1,2,3-triazole dyads have been synthesized by two different approaches, both starting from novel (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)chromones, prepared through a base-catalyzed aldol reaction of 2-methylchromone and arylpropargyl aldehydes. In the first method, the xanthone moiety is built by Diels–Alder reaction of the referred unsaturated chromones with *N*-methylmaleimide under microwave irradiation, followed by oxidation of the obtained adducts with DDQ, whereas the 1,2,3-triazole ring results from the cycloaddition reaction of the acetylene moiety with sodium azide. The second strategy first involves the cycloaddition reaction with sodium azide to provide the 1,2,3-triazole ring, followed by methylation of the triazole NH group prior to Diels–Alder reaction with *N*-methylmaleimide. The last step in this synthesis of novel xanthone-1,2,3-triazole dyads entails oxidation of the cycloadducts with DDQ.

Introduction

Xanthones or 9*H*-xanthen-9-ones are one of the most important classes of naturally occurring oxygenated heterocyclic compounds possessing a dibenzo-γ-pyrone framework. The parent xanthone has not been reported as a natural product.[1] However, in 2002 Oldenburg et al. described the occurrence of this xanthone in crude oils from offshore Norway. The authors suggest that this product can be formed by oxidation of xanthene in the reservoir or obtained by geosynthesis from aromatic precursors.[2] Natural xanthones often appear as highly substituted derivatives, bearing methoxy, hydroxy, alkyl, isopentenyl and glycosyl groups in their monomeric, dimeric, polycyclic or xanthonolignoide forms.[3] In the past few years, a great number of studies have emphasized the biological and pharmacological properties of both natural and synthetic xanthones,[4] including anti-inflammatory,[5] cancer chemopreventive,[6] antimalarial,[7] and radical scavenging activities.[8] The inhibitory activities of these agents against enzymes such as cyclooxygenase and cholinesterase have also been reported.[4,9]

Likewise, among the nitrogen heterocycles, triazoles have particular relevance due to their application in several research fields such as biochemistry, pharmaceutical and material science. Triazoles have been used as drugs,[10] and are known to possess anti-HIV-type I protease,[11] anti-hyperglycemic,[12] and antimicrobial activities, among others.[13] In terms of medicinal chemistry, it has been demonstrated that 1,4- and 1,5-disubstituted 1,2,3-triazoles can participate in important binding interactions with biological targets, maintaining a good pharmacokinetic profile.[14] In addition, they are commercially used as anticorrosive agents,[15] agrochemicals,[16] photostabilizers and dyes.[17]

Considering the biological properties exhibited by these two classes of heterocycles, the development of new production methods for xanthone-1,2,3-triazole dyads aimed specifically at developing potentially improved therapeutic agents is a high priority. This idea was recently exemplified by a study carried out by Zou et al. in which xanthones bearing a 1,4-disubstituted-1,2,3-triazole moiety showed promising antitumor activity.[18] Another study indicated that 3,6-dihydroxyxanthone, known as a good fluorophore, increased its fluorescence upon triazole formation.[19] With this rational in mind, we designed and synthesized novel chromone derivatives to be used as building blocks in the synthesis of xanthone-1,2,3-triazole dyads. The first reports dealing with the reactivity of chromone derivatives in Diels–Alder (DA) reactions *en route* to xanthone derivatives have been summarized by our group in 1993.[20] At that time, xanthene adducts were not characterized and were assumed to have the expected 1,2,3,9a-tetrahydroxanthene structure. Subsequently, other studies revealed that the products formed were, in fact, 1,2,3,4-tetrahydroxanthenes.[21] In 2000, 2-vinylchromones were also used as starting materials in [4 + 2] cycloaddition reactions with enamines for the synthesis of new xanthone derivatives.[22] More recently, studies

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on DA reactions of 3-styrylchromones under microwave irradiation in solvent-free conditions resulted in a new strategy for the synthesis of novel xanthones.[23] In 2009, 2- and 3-styrylchromones were used in cyclization reactions to afford new polyhydroxylated 2,3-diarylxanthones.[24] Following the previously described studies, 2-(4-arylbuta-1,3-dien-1-yl)chromones underwent electrocyclization and oxidation processes in order to prepare 1-arylxanthones.[25] In this paper, we disclose our synthesis of new chromone derivatives, 2-(4-arylbut-1-en-3-yn-1-yl)chromones **4a**–**d**, which were further used as synthons to generate new xanthone-1,2,3-triazole dyads. Such chromones **4a**–**d** possess two unsaturated systems (a diene and an alkyne) anticipated to display different reactivities in cycloaddition reactions. Moreover, these new chromones can participate in two consecutive types of cycloaddition reactions; namely, DA and Huisgen azide–alkyne 1,3-dipolar cycloadditions ("click chemistry"). Herein, we present two different synthetic strategies enabling facile access to new chromone-1,2,3-triazole derivatives as well as xanthone-1,2,3-triazole dyads.

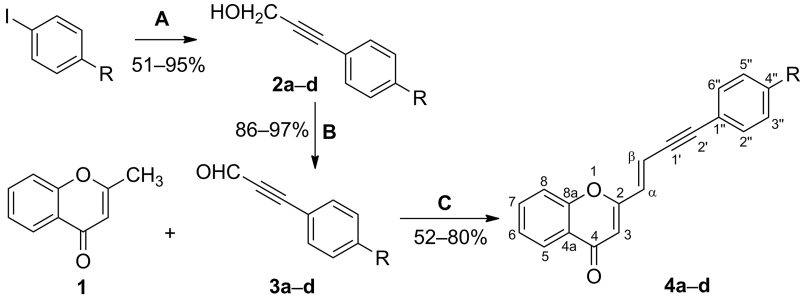
Results and Discussion

Syntheses

Novel (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)chromones **4a**–**d** were obtained in moderate to good yields (52–80%) using a base-catalyzed aldol reaction of 2-methylchromone **1** with arylpropargyl aldehydes **3a**–**d** in ethanolic solutions (Scheme 1).[25] The required starting material 2-methylchromone (**1**) was prepared by a three-step Baker–Venkataraman sequence in good overall yields.[26] Since arylpropargyl aldehydes **3b**–**d** are not commercially available, we generated them using a two-step approach: i) palladium-catalyzed cross-coupling reaction of aryl iodides with propargyl alcohol[27] in order to prepare the corresponding arylpropargyl alcohols **2b**–**d**, in good yields (79–95%); ii) oxidation of the arylpropargyl alcohols **2b**–**d** with activated MnO_2 to afford desired arylpropargyl aldehydes **3b**–**d**, in excellent yields (86–97%). The first step of this methodology was slightly improved by using toluene as solvent in the presence of 1.5 equiv. piperidine instead of in neat diethylamine (Scheme 1).

In our first approach to the target xanthone-1,2,3-triazole dyads, newly synthesized chromones **4a**–**d** were used as dienes in DA reactions with the poor dienophile *N*-methyl-maleimide (Scheme 2). The structure of each conjugated diene suggests that it could not be very reactive due to extended electron delocalization. Thus, we tried highly energetic reaction conditions using 1,2,4-trichlorobenzene (TCB) under refluxing conditions (Table 1, Entry 1). No DA reaction took place and after a reaction time of 7 d only degradation of the starting material was observed when using two different derivatives **4a** and **4c** (Table 1, Entries 1 and 2). Slightly less energetic conditions (DMF at reflux for 48 h) once again failed to produce any DA reaction despite the inclusion of the Lewis acid $Sc(OTf)_3$ (Table 1, Entries 3 and 4). The failure of the normal (or "typical") electron demand DA reaction to produce meaningful results inspired us to attempt the reaction using an electron rich dienophile (dihydropyran) in neat conditions (Table 1, Entry 5). This DA reaction also failed to take place; we postulate that this is a consequence of the low reaction temperature imposed by the low boiling point of dihydropyran.

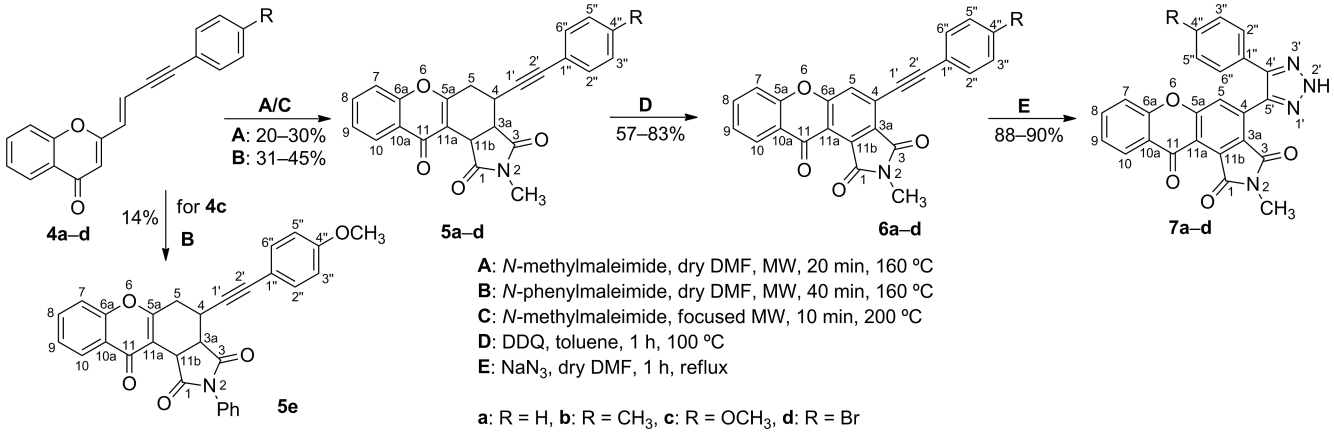
On the basis of previous findings by our group[23,28] we performed the DA reaction under microwave irradiation (multimode apparatus) conditions. Several experiments were carried out in which reaction times, solvent, Lewis acids, amounts of dienophile and microwave potencies were varied (Table 1, Entries 6–15). The best results were achieved in a reaction time of 20 min, at 160 °C using a few drops of DMF as solvent (Table 1, Entries 6, 16–18). DA adducts **5a**–**d** were obtained in low yields (20–30%) although it is worth noting that a significant amount of starting chromone could be readily recovered (65–70%). Thus, the poor yields can be explained by the competition of the retro-DA reaction, which is often promoted at such temperatures.[29] When the reaction of **4c** with *N*-methylmaleimide was performed under the optimized conditions



A: Toluene, $Pd(PPh_3)_2Cl_2$, piperidine, CuI, propargyl alcohol, 60 °C, 2 h;
B: Ethyl acetate, MnO_2, reflux, 1 h;
C: Na, EtOH, r.t.

a: R = H; **b**: R = CH_3; **c**: R = OCH_3; **d**: R = Br

Scheme 1. Synthesis of (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)chromones **4a**–**d**.



Scheme 2. Strategy for the synthesis of xanthone-1,2,3-triazole dyads **7a**–**d**.

Table 1. Optimization of the Diels–Alder reaction conditions for the synthesis of 4-(arylethynyl)-2-methyl/phenyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11-triones **5a**–**d**.

Entry	Heating method	Reactant	Solvent	Time	*T* [°C]	Catalyst	Isolated yield [%]	Recovered starting material [%]
1	oil bath	**4a**	1,2,4-TCB	7 d	reflux	–	no reaction	–
2	oil bath	**4c**	1,2,4-TCB	7 d	reflux	–	no reaction	–
3	oil bath	**4c**	DMF	48 h	reflux	$Sc(OTf)_3$	no reaction	–
4	oil bath	**4c**	DMF	48 h	reflux	–	no reaction	–
5	oil bath	**4b**	–[e]	24 h	80	–	no reaction	–
6	MW (800 W)	**4c**	DMF[a]	40 min	160	–	30	67
7	MW (800 W)	**4c**	DMF[a]	40 min	160	$AlCl_3$	30	65
8	MW (800 W)	**4c**	DMF[a]	40 min	160	$Sc(OTf)_3$	30	66
9	MW (800 W)	**4c**	DMF[a]	20 min	160	–	30	68
10	MW (800 W)	**4c**	DMF[a]	10 min	160	–	14	76
11	MW (800 W)	**4c**	DMF[c]	40 min	160	–	no reaction	–
12	MW (800 W)	**4c**	NMP[a]	40 min	160	–	no reaction	–
13	MW (600 W)	**4c**	NMP[a]	40 min	150	–	5	70
14	MW (600 W)	**4c**	NMP[b]	40 min	150	–	no reaction	–
15	MW (600 W)	**4c**	DMF[b]	40 min	150	–	no reaction	–
16	MW (800 W)	**4a**	DMF[a]	20 min	160	–	28	67
17	MW (800 W)	**4b**	DMF[a]	20 min	160	–	20	72
18	MW (800 W)	**4d**	DMF[a]	20 min	160	–	30	66
19	MW (800 W)	**4b**	DMF[d]	40 min	160	–	14	10

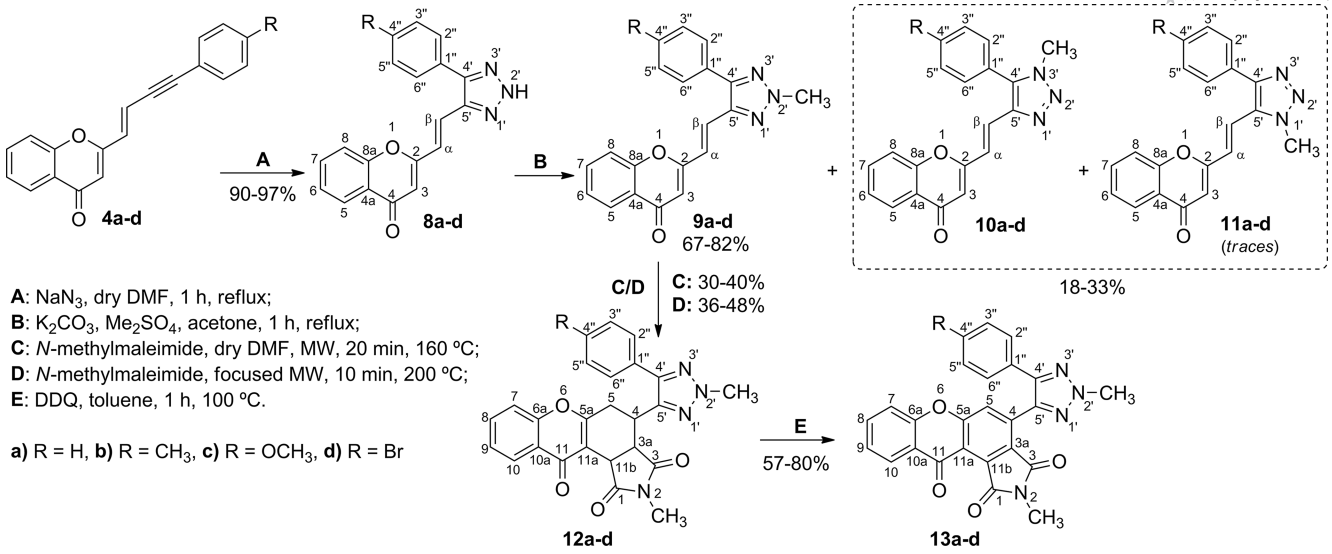
[a] A few drops of DMF and 5 equiv. of *N*-methylmaleimide. [b] 5 mL of solvent and 10 equiv. *N*-methylmaleimide. [c] Dimethyl acetylenedicarboxylate. [d] *N*-phenylmaleimide. [e] Dihydropyran.

(160 °C, using a few drops of DMF as solvent) for 10 min cycloadduct **5c** was isolated in 14% yield; reaction times of 20 and 40 min afforded the same yield (30%) (Table 1, Entries 6, 9 and 10). At lower reaction temperature (100 °C), cycloadduct **5c** was obtained in 1% yield and 80% of the starting material was recovered.

A single attempt to test *N*-phenylmaleimide as the dienophile led to no improvements in the DA reaction and led to only a 14% yield of cycloadduct **5e** and 10% recovered starting material (Table 1, Entry 19, Scheme 2). The DA reaction has also been studied using monomode microwave equipment in solvent-free conditions. As an example, chromone **4c** was used in three experiments performed at 5, 10 and 20 min of irradiation at 200 °C. The best results (31%) were not significantly better than those obtained under the multimode microwave conditions (30%). However, it was possible to achieve similar results in shorter reaction times (10 min). Thus, after 5 min of irradiation it was possible to isolate **5c** in 23% yield; with 10 or 20 min of irradiation the yields increased to 31%. Applying these optimized conditions to the other derivatives, we can conclude that the yields were slightly improved (36–45%) using this environmentally friendlier technique since the reaction occurs in solvent-free conditions and in a shorter reaction time.

The next step of our first strategy was the oxidation/aromatization of the DA adducts to afford xanthone derivatives. The reaction was performed in toluene with DDQ and the corresponding xanthone derivatives **6a**–**d** were achieved in good yields (Scheme 2, 57–83%). The synthesized xanthones possess alkyne substitution in position 4 of the heterocycle, which, upon reacting with sodium azide in refluxing DMF provided new xanthone-1,2,3-triazole dyads **7a**–**d** in excellent yields (Scheme 2, 88–90%).

In the second strategy an inverse approach was used, starting with the synthesis of the triazole ring and then the xanthone skeleton. First, the 1,3-cycloaddition reaction of



Scheme 3. Strategy for the synthesis of xanthone-1,2,3-triazole dyads **13a**–**d**.

chromones **4a**–**d** with sodium azide afforded the 1,2,3-triazole derivatives **8a**–**d** in excellent yields (Scheme 3, 90–97%). Subsequently and before the optimization of the DA reaction we protected the NH of the triazole since it could undergo a Michael addition reaction with the *N*-methylmaleimide dienophile.[30] N-Protection with the methyl group was achieved using dimethyl sulfate in refluxing acetone and the expected three isomers of 1,2,3-triazoles **9a**–**d**, **10a**–**d** and **11a**–**d** were prepared in good overall yields (Scheme 3): 2′-*N*CH_3 triazoles **9a**–**d** (higher R_f value) were isolated as major isomers (67–82%); whereas 3′-*N*CH_3 triazoles **10a**–**d** together with 1′-*N*CH_3 isomers **11a**–**d** were obtained as an inseparable mixture in low yields (18–33%). Analysis of the NMR spectra of this mixture allowed us to conclude that 1′-*N*CH_3 isomers **11a**–**d** were obtained in trace amounts (see NMR discussion).

At this point and being aware that the DA reaction of chromone **4a**–**d** only occurs under microwave irradiation conditions, the reaction of protected 1,2,3-triazole derivatives **9a**–**d** with *N*-methylmaleimide was optimized using similar heating conditions. Once more, the lack of reactivity of the diene resulted in moderate yields of cycloadducts **12a**–**d** (30–40%) and 50–64% of recovered **9a**–**d** when the reactions were performed in DMF at 160 °C for 20 min, with microwave irradiation (multimode apparatus). The optimization of the reaction using a monomode microwave apparatus was carried out using 1,2,3-triazole derivative **9c**, in solvent-free conditions, with irradiation times of 5, 10 and 20 min at 200 °C. The best yield of compound **12c** was obtained with a 10 min irradiation; when applying these conditions to the other derivatives, the yields of cycloadducts **12a**–**d** (36–48%) were slightly better than those obtained using multimode microwave equipment. Furthermore, no side products were detected and 37–51% of starting materials **9a**–**d** were recovered from the reaction mixtures. Finally, dehydrogenation of cycloadducts **12a**–**d** leading to xanthones **13a**–**d** occurred in the presence of DDQ oxidant and with good results (Scheme 3, 57–80%).

Nuclear Magnetic Resonance Spectroscopy

The most important features in the ^{1}H NMR spectra of the (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)chromones **4a**–**d** are, in each case: i) two doublets, at δ = 6.66–6.71 and 6.89–6.93 ppm corresponding to the vinylic protons α-H and β-H respectively, in a *trans* configuration ($J_{\alpha H,\beta H}$ 15–16 Hz), and ii) a singlet at δ = 6.26–6.28 ppm corresponding to 3-H. HMBC spectra enabled the assignment of the most relevant quaternary carbons: C-1′ and C-2′ at δ = 86.7–88.4 and 96.5–98.4 ppm, respectively, C-2 at δ = 160.0–160.5 ppm, C-8a and C-4a at δ = 155.9 and 124.1 ppm, respectively, and C-4 at δ = 178.3 ppm (Figure 1). In the NOESY spectra of compounds **4a**–**d** NOE cross-peaks were observed between the 3-H signal and those of α-H and β-H indicating free rotation around the C2–Cα bond.

The ^{1}H NMR spectra of DA cycloadducts **5a**–**d**, revealed the disappearance of the structural features mentioned for **4a**–**d**. Furthermore, the ^{1}H NMR spectra showed the presence of five new signals: i) two multiplets at δ = 3.02–3.08 ppm and δ = 3.70–3.77 ppm, corresponding to 5-H and 4-H; ii) a double doublet at δ = 3.28–3.29 ppm, corresponding to 3a-H; iii) a doublet at δ = 4.37–4.39 ppm, corresponding to 11b-H; and iv) a singlet at δ = 2.96–2.98 ppm, corresponding to the 2-NCH_3 belonging to *N*-methylmaleimide moiety. All these structural features of compounds **5a**–**d** support the disappearance of structural features characteristic of compounds **4a**–**d** indicating that the DA reaction proceeded thereby affording desired cycloadducts **5a**–**d**. Analysis of the ^{13}C NMR spectra enabled assignment of three carbonyl carbons: C-11 from the chromone moiety, and C-1 and C-3 corresponding to *N*-methyl-

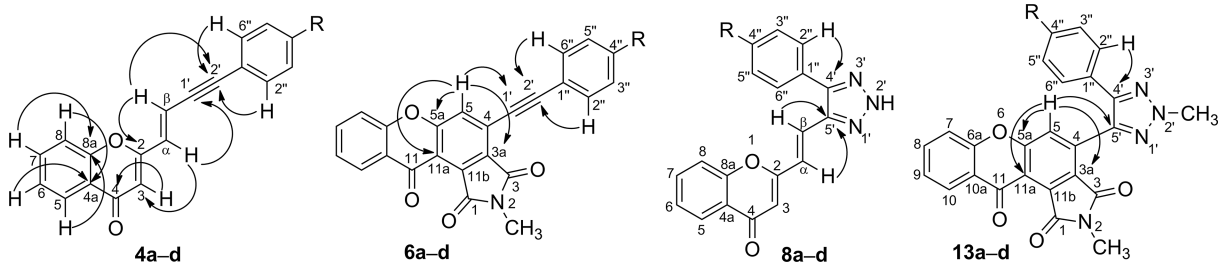


Figure 1. Important connectivities found in the HMBC spectra of the compounds **4a**–**d**, **6a**–**d**, **8a**–**d** and **13a**–**d**.

maleimide. Furthermore, the HMBC spectra of derivatives **5a**–**d**, allowed the assignment of carbons C-1′ (δ = 83.3–84.8 ppm) and C-2′ (δ = 85.7–86.0 ppm) through the connectivity with aliphatic protons and with H-2′′,6′′, respectively.

The most important characteristics in the ^{1}H NMR spectra of 4-(arylethynyl)-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones **6a**–**d** are the presence in each case of two singlets, at δ = 7.85–7.89 ppm corresponding to the aromatic protons 5-H, and at δ = 3.26 ppm corresponding to the protons of the 2-NCH_3 group of the *N*-methylmaleimide moiety. Also, the aliphatic protons characteristic of DA adducts **5a**–**d** do not appear in the aliphatic regions of the spectra.

The most relevant structural differences between compounds **6a**–**d** and **7a**–**d** are found in the ^{13}C NMR spectra. In compounds **6a**–**d** the signals corresponding to C-1′ and C-2′, assigned through HMBC analysis, appear at δ = 83.4–85.0 and 99.0–101.2 ppm, respectively. These signals are not present in the ^{13}C NMR spectra of compounds **7a**–**d**; instead, analysis of their ^{13}C NMR and HMBC spectra showed the presence of two signals at δ = 136.3–139.3 ppm and δ = 142.1–142.7 ppm, corresponding to C-5′ and C-4′, respectively, of the 1,2,3-triazole moiety.

The multiplicity of the ^{1}H NMR spectra of the (*E*)-2-[2-(4-aryl-2*H*-1,2,3-triazol-5-yl)vinyl]chromones **8a**–**d** is pretty much the same as in (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)chromones **4a**–**d**. However, the signals of α-H, β-H and 3-H (δ = 7.30–7.35, 7.61–7.68 and 6.58–6.61 ppm, respectively) appear at higher frequency than those of the same protons in compounds **4a**–**d**. The deshielding effect of the triazole moiety is more pronounced for α-H and β-H than for the 3-H protons. As in the case of **4a**–**d**, the NOESY spectra of compounds **8a**–**d** present NOE cross-peaks between the signal of 3-H and those of α-H and β-H indicating free rotation around the C2–Cα bond. In the case of the ^{13}C NMR spectra of compounds **8a**–**d**, the most important feature is the set of signals for C-4′ and C-5′, from 1,2,3-triazole moiety, at δ = 137.6–138.3 and 141.8–145.1 ppm, respectively, assigned from HMBC spectra (Figure 1).

In the ^{1}H NMR spectra of (*E*)-2-[2-(4-aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)vinyl]chromones **9a**–**d** the most relevant feature is the singlet at δ = 4.28–4.29 ppm corresponding to 2′-NCH_3 of the methylated 1,2,3-triazole moiety. The unequivocal identification of isomers **9a**–**d** and **10a**–**d** was based on the connectivity found in the HMBC spectra. Thus, 2′-NCH_3 protons of isomers **9a**–**d** do not correlate with any carbon of the 1,2,3-triazole ring. The position of the methyl group in the triazole ring of isomers **10a**–**d** was identified by correlations observed between 2′′,6′′-H and 3′-NCH_3 with C-4′ of the 1,2,3-triazole ring. Traces of isomers **11a**–**d** were detected by the presence of less intense signals of the 1′-NCH_3 protons and 3-H in the ^{1}H NMR spectra of the mixture containing both **10a**–**d** and **11a**–**d** isomers.

Once again, the ^{1}H NMR spectra of the 4-(4-aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones **12a**–**d** do not show the structural features described for compounds **8a**–**d** and **9a**–**d**. Indeed, new signals are present in these spectra: i) two multiplets at δ = 2.96–3.24 and 3.79–3.91 ppm, corresponding to 5-H and 4-H; ii) a double duplet at δ = 3.46–3.48 ppm, due to 3a-H; iii) a doublet at δ = 4.63–4.64 ppm, assigned to 11b-H; and iv) a singlet at δ = 2.81–2.82 ppm, attributed to 2-NCH_3 of the *N*-methylmaleimide moiety. The key feature in the ^{1}H NMR spectra of 4-(5-aryl-2-methyl-2*H*-1,2,3-triazol-4-yl)-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones **13a**–**d** is that of a singlet at δ = 7.83–7.85 ppm assigned to the 5-H aromatic proton in each case. Moreover, the ^{1}H NMR spectra of compounds **13a**–**d** do not show aliphatic protons indicating successful aromatization of DA cycloadducts **12a**–**d**.

Conclusions

We have reported the preparation of novel (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)chromone derivatives, which were further used as starting materials in two main synthetic routes to xanthone-1,2,3-triazole dyads. The first strategy involves the Diels–Alder reaction of the chromone derivatives with *N*-methylmaleimide followed by aromatization of the cycloadducts and reaction with sodium azide to give desired xanthone-1,2,3-triazole dyads. In the second synthetic approach an additional step of 1,2,3-triazole protection is required. Thus, chromone derivatives first react with sodium azide to provide chromone-1,2,3-triazole dyads, in excellent yields. After methylation of the triazole NH group, DA reaction with *N*-methylmaleimide and subsequent oxidation with DDQ affords the corresponding cycloadducts.

The DA reactions were performed under both monomode and multimode microwave irradiation conditions. The former case appears to be a more sustainable and environmentally friendly process since the reaction occurs in sol-

vent-free conditions, a shorter reaction time and consequently less energy is required. Moreover, yields obtained in this "greener" fashion were slightly improved relative to those obtained from the multimode microwave irradiation conditions.

Experimental Section

General Remarks: Melting points were measured with a Büchi Melting Point B-540 apparatus. NMR spectra were recorded with a Bruker Avance 300 spectrometer (300.13 MHz for ^{1}H and 75.47 MHz for ^{13}C) or Bruker Avance 500 spectrometer (500.13 MHz for ^{1}H and 125.77 MHz for ^{13}C). Chemical shifts (δ) are reported in ppm and coupling constants (J) in Hz; the internal standard was TMS. Unequivocal ^{13}C assignments were made with the aid of 2D *g*HSQC and *g*HMBC (delays for one-bond and long-range J C/H couplings were optimised for 145 and 7 Hz, respectively) experiments. Positive-ion ESI mass spectra were acquired with a QTOF 2 instrument [dilution of 1 μL of the sample in chloroform solution (ca. 10^{-5} M) in 200 μL of 0.1% trifluoroacetic acid/methanol solution. Nitrogen was used as nebuliser gas and argon as collision gas. The needle voltage was set at 3000 V, with the ion source at 80 °C and the desolvation temperature at 150 °C. The cone voltage was 35 V]. Other low- and high-resolution mass spectra (EI, 70 eV) were measured with VG Autospec Q and M spectrometers. Elemental analyses were obtained with a LECO 932 CHNS analyser. Preparative thin-layer chromatography was performed with Merck silica gel (60 DGF254). Column chromatography was performed with Merck silica gel (60, 70–230 mesh). All chemicals and solvents used were obtained from commercial sources and used as received or dried by standard procedures. 3-Phenylpropiolaldehyde (**3a**) was not prepared since it is commercially available (Sigma–Aldrich).

General Procedure for the Synthesis of Arylpropargyl Alcohols 2b–d: Propargyl alcohol (1.75 mL, 30 mmol) was added to a solution of the appropriate iodobenzene (20 mmol), bis(triphenylphosphine) palladium(II) chloride (280 mg, 0.04 mmol), piperidine (3.96 mL, 40 mmol) and copper iodide (114 mg, 0.6 mmol) in toluene (10 mL), under a nitrogen atmosphere. The mixture was stirred at 60 °C for 2 h and then filtered through Celite 345 and washed with chloroform. The solvent was evaporated and the residue was purified by silica gel column chromatography using CH_2Cl_2 as eluent. The solvent was evaporated to dryness in each case, and expected 3-arylprop-2-yn-1-ols **2b–d** were obtained in good yields (79–95%).

3-(4-Methylphenyl)prop-2-yn-1-ol (2b): Yield 2.31 g (79%). Oil. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.34 (s, 3 H, 4′-CH_3), 4.48 (d, J = 5.2 Hz, 2 H, 1-H), 7.11 (d, J = 7.9 Hz, 2 H, 3′,5′-H), 7.33 (d, J = 7.9 Hz, 2 H, 2′,6′-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.4 (4′-*C*H$_3$), 51.6 (C-1), 85.7 (C-3), 86.5 (C-2), 119.3 (C-1′), 129.0 (C-3′,5′), 131.5 (C-2′,6′), 138.6 (C-4′) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{10}H_{11}O$ [M + H]$^+$ 147.0810, found 147.0801. MS (ESI$^+$): *m*/*z* (%) = 147 (100) [M + H]$^+$, 315 (35) [2M + Na]$^+$.

3-(4-Methoxyphenyl)prop-2-yn-1-ol (2c): Yield 2.56 g (79%). This compound showed spectroscopic and analytical data identical to previously reported data.[31]

3-(4-Bromophenyl)prop-2-yn-1-ol (2d): Yield 4.01 g (95%). This compound showed spectroscopic and analytical data identical to previously reported data.[32]

General Procedure for the Synthesis of Arylpropargyl Aldehydes 3b–d: Activated MnO_2 (6.9 g, 79.6 mmol) was added to a solution of the appropriate arylpropargyl alcohols **2b–d** (15.9 mmol) in EtOAc (40 mL). The resulting mixture was then refluxed for 1 h. After that period, the mixture was filtered through Celite 345 and washed with EtOAc and CH_2Cl_2. After evaporation of the solvent, the residue was purified by silica gel column chromatography using CH_2Cl_2 as eluent. The solvent was evaporated to dryness to give the expected arylpropynals **3b–d** in good yields (86–97%).

3-(4-Methylphenyl)propiolaldehyde (3b): Yield 1.97 g (86%). Oil. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.40 (s, 3 H, 4′-CH_3), 7.22 (d, J = 8.0 Hz, 2 H, 3′,5′-H), 7.51 (d, J = 8.0 Hz, 2 H, 2′,6′-H), 9.41 (s, 1 H, 1-H) ppm. ^{13}C NMR (300 MHz, $CDCl_3$): δ = 21.8 (4′-*C*H$_3$), 88.4 (C-2), 96.0 (C-3), 116.3 (C-1′), 129.5 (C-3′,5′), 133.3 (C-2′,6′), 142.2 (C-4′), 176.9 (C-1) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{10}H_9O$ [M + H]$^+$ 145.0653, found 145.0644. MS (ESI$^+$): *m*/*z* (%) = 189 (55) [M + 2Na]$^+$, 145 (30) [M + H]$^+$.

3-(4-Methoxyphenyl)propiolaldehyde (3c): Yield 2.34 g (92%). This compound showed spectroscopic and analytical data identical to previously reported data.[31]

3-(4-Bromophenyl)propiolaldehyde (3d): Yield 3.22 g (97%). This compound showed spectroscopic and analytical data identical to previously reported data.[32]

General Procedure for the Synthesis of 2-(4-Arylbut-1-en-3-yn-1-yl)-4*H*-chromen-4-ones 4a–d: Sodium (0.11 g, 4.8 mmol) was gradually added to 5 mL of dry ethanol and the mixture was stirred until the reaction mixture reached room temperature. 2-Methylchromone **1** (0.2 g, 1.2 mmol) and the appropriate aldehyde **3a–d** (1.8 mmol) were added and the reaction mixture allowed to stand at room temperature until complete disappearance of chromone **1**. After this period, the solution was poured into ice (20 g) and water (30 mL) and the pH adjusted to 4 with dilute HCl. The solid was removed by filtration, taken in CH_2Cl_2 and purified by silica gel column chromatography using CH_2Cl_2 as eluent. The solvent was evaporated to dryness and the residue were recrystallized from ethanol to give the (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)-4*H*-chromen-4-ones **4a–d** in good yields (52–80%).

(*E*)-2-(4-Phenylbut-1-en-3-yn-1-yl)-4*H*-chromen-4-one (4a): Yield 166 mg (52%), m.p. 127–128 °C (recrystallized from ethanol). ^{1}H NMR (300 MHz, $CDCl_3$): δ = 6.28 (s, 1 H, 3-H), 6.71 (d, J = 15.7 Hz, 1 H, α-H), 6.93 (d, J = 15.7 Hz, 1 H, β-H), 7.35–7.43 (m, 4 H, 6-H, 3′′,4′′,5′′-H), 7.48 (d, J = 7.7 Hz, 1 H, 8-H), 7.48–7.52 (m, 2 H, 2′′,6′′-H), 7.69 (dt, J = 7.7, 1.7 Hz, 1 H, 7-H), 8.19 (dd, J = 7.9, 1.7 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 87.4 (C-1′), 97.8 (C-2′), 111.1 (C-3), 117.2 (C-β), 117.9 (C-8), 122.4 (C-1′′), 124.1 (C-4a), 125.2 (C-6), 125.7 (C-5), 128.5 (C-3′′,5′′), 129.2 (C-4′′), 131.9 (C-2′′,6′′), 132.1 (C-α), 134.0 (C-7), 155.9 (C-8a), 160.2 (C-2), 178.3 (C-4) ppm. $C_{19}H_{12}O_2$: C, 83.81; H, 4.44; found C, 83.48; H, 4.40. MS (ESI$^+$): *m*/*z* (%) = 273 (100) [M + H]$^+$, 295 (54) [M + Na]$^+$.

(*E*)-2-[4-(4-Methylphenyl)but-1-en-3-yn-1-yl]-4*H*-chromen-4-one (4b): Yield 286 mg (80%), m.p. 148–150 °C (recrystallized from ethanol). ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.38 (s, 3 H, 4′′-CH_3), 6.27 (s, 1 H, 3-H), 6.69 (d, J = 15.8 Hz, 1 H, α-H), 6.93 (d, J = 15.8 Hz, 1 H, β-H), 7.18 (d, J = 8.1 Hz, 2 H, 3′′,5′′-H), 7.400 (ddd, J = 7.6, 7.5, 1.0 Hz, 1 H, 6-H), 7.402 (d, J = 8.1 Hz, 2 H, 2′′,6′′-H), 7.48 (dd, J = 7.9, 1.0 Hz, 1 H, 8-H), 7.69 (ddd, J = 7.9, 7.6, 1.7 Hz, 1 H, 7-H), 8.19 (dd, J = 7.6, 1.7 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.6 (4′′-*C*H$_3$), 87.0 (C-1′′), 98.3 (C-2′), 111.0 (C-3), 117.4 (C-β), 117.9 (C-8), 119.3 (C-1′), 124.1 (C-4a), 125.1 (C-6), 125.7 (C-5), 129.3 (C-3′′,5′′), 131.6 (C-α), 131.8 (C-2′′,6′′), 133.9 (C-7), 139.6 (C-4′′), 155.9 (C-8a), 160.4 (C-2), 178.3 (C-4) ppm. $C_{20}H_{14}O_2$: C, 83.90; H, 4.93; found C, 83.51; H, 4.91. MS (ESI$^+$): *m*/*z* (%) = 287 (100) [M + H]$^+$, 325 (12) [M + K]$^+$, 595 (8) [2M + Na]$^+$.

(*E*)-2-[4-(4-Methoxyphenyl)but-1-en-3-yn-1-yl]-4*H*-chromen-4-one (4c): Yield 302 mg (80%), m.p. 162–163 °C (recrystallized from ethanol). ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.85 (s, 3 H, 4′′-OCH_3), 6.26 (s, 1 H, 3-H), 6.66 (d, J = 15.7 Hz, 1 H, α-H), 6.89 (d, J = 8.6 Hz, 2 H, 3′′,5′′-H), 6.93 (d, J = 15.7 Hz, 1 H, β-H), 7.40 (dd, J = 7.9, 7.8 Hz, 1 H, 6-H), 7.45 (d, J = 8.6 Hz, 2 H, 2′′,6′′-H), 7.48 (d, J = 7.6 Hz, 1 H, 8-H), 7.69 (ddd, J = 7.8, 7.6, 1.6 Hz, 1 H, 7-H), 8.19 (dd, J = 7.9, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 55.4 (4′′-O*C*H$_3$), 86.7 (C-1′), 98.4 (C-2′), 110.8 (C-3), 114.2 (C-3′′,5′′), 114.4 (C-1′′), 117.5 (C-β), 117.8 (C-8), 124.1 (C-4a), 125.1 (C-6), 125.7 (C-5), 131.1 (C-α), 133.5 (C-2′′,6′′), 133.9 (C-7), 155.9 (C-8a), 160.4 (C-4′′), 160.5 (C-2), 178.3 (C-4) ppm. $C_{20}H_{14}O_3$: C, 79.46; H, 4.67; found C, 79.61; H, 4.83. MS (ESI$^+$): m/z (%) = 303 (86) [M + H]$^+$, 325 (53) [M + Na]$^+$, 627 (100) [2M + Na]$^+$.

(*E*)-2-[4-(4-Bromophenyl)but-1-en-3-yn-1-yl]-4*H*-chromen-4-one (4d): Yield 259 mg (59%), m.p. 164–166 °C (recrystallized from ethanol). ^{1}H NMR (300 MHz, $CDCl_3$): δ = 6.28 (s, 1 H, 3-H), 6.71 (d, J = 15.8 Hz, 1 H, α-H), 6.89 (d, J = 15.8 Hz, 1 H, β-H), 7.36 (d, J = 8.6 Hz, 2 H, 2′′,6′′-H), 7.40 (dt, J = 7.8, 0.9 Hz, 1 H, 6-H), 7.48 (dd, J = 8.7, 0.9 Hz, 1 H, 8-H), 7.51 (d, J = 8.6 Hz, 2 H, 3′′,5′′-H), 7.69 (ddd, J = 8.7, 7.8, 1.7 Hz, 1 H, 7-H), 8.19 (dd, J = 7.8, 1.7 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 88.4 (C-1′), 96.5 (C-2′), 111.3 (C-3), 116.7 (C-β), 117.8 (C-8), 121.3 (C-1′′), 123.6 (C-4′′), 124.1 (C-4a), 125.2 (C-6), 125.7 (C-5), 131.8 (C-3′′,5′′), 132.5 (C-α), 133.2 (C-2′′,6′′), 134.0 (C-7), 155.9 (C-8a), 160.0 (C-2), 178.3 (C-4) ppm. $C_{19}H_{11}BrO_2$: C, 64.98; H, 3.16; found C, 64.71; H, 3.23. MS (ESI$^+$): m/z (%) = 351 (39) [M + H]$^+$ (^{79}Br); 353 (37) [M + H]$^+$ (^{81}Br).

General Procedure for the Synthesis of 4-(Arylethynyl)-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones 5a–e. Method A: *N*-Methylmaleimide (0.10 g, 0.92 mmol) was added to a solution of the appropriate (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)-4*H*-chromen-4-one (**4a–d**) (0.18 mmol) in dry DMF (5 μL). The mixture was heated at 160 °C under microwave irradiation (multimode apparatus) for 20 min. The residue was dissolved in CH_2Cl_2 and purified by preparative TLC using CH_2Cl_2 as eluent to give desired cycloadducts **5a–d**: **5a** (19.7 mg, 28%), **5b** (19.3 mg, 20%), **5c** (22.3 mg, 30%), **5d** (25.0 mg, 30%). Adduct **5e** was obtained in 14% using the same methodology, under irradiation for 40 min.

Method B: *N*-Methylmaleimide (0.10 g, 0.92 mmol) was mixed with the appropriate (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)-4*H*-chromen-4-one **4a–d** (0.18 mmol) in a closed vessel. The mixture was heated at 200 °C under microwave irradiation (monomode apparatus) for 10 min. The residue was dissolved in CH_2Cl_2 and purified by preparative TLC using CH_2Cl_2 as eluent to give desired cycloadducts **5a–d**: **5a** (25.3 mg, 36%), **5b** (40.5 mg, 42%), **5c** (23.0 mg, 31%), **5d** (37.5 mg, 45%).

2-Methyl-4-(phenylethynyl)-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]-isoindole-1,3,11(2*H*)-trione (5a): M.p. 169–171 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.98 (s, 3 H, CH_3), 3.04–3.08 (m, 2 H, 5-H), 3.29 (dd, J = 7.6, 5.0 Hz, 1 H, 3a-H), 3.73–3.77 (m, 1 H, 4-H), 4.39 (d, J = 7.6 Hz, 1 H, 11b-H), 7.21–7.28 (m, 5 H, 2′′,3′′,4′′,5′′,6′′-H), 7.39–7.45 (m, 2 H, 7-H, 9-H), 7.67 (ddd, J = 8.6, 7.1, 1.7 Hz, 1 H, 8-H), 8.29 (dd, J = 8.2, 1.7 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.8 (N*C*H$_3$), 27.8 (C-4), 33.2 (C-5), 37.8 (C-11b), 43.2 (C-3a), 84.7 (C-1′), 85.8 (C-2′), 112.1 (C-11a), 117.6 (C-7), 121.7 (C-1′′), 123.5 (C-10a), 125.2 (C-9), 126.4 (C-10), 128.2 (C-2′′,6′′), 128.6 (C-4′′), 131.6 (C-3′′,5′′), 133.7 (C-8), 155.7 (C-6a), 162.1 (C-5a), 174.2 (C-3 and C-11), 176.1 (C-1) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{24}H_{18}NO_4$ [M + H]$^+$ 384.1236, found 384.1218. MS (ESI$^+$): m/z (%) = 384 (32) [M + H]$^+$, 406 (94) [M + Na]$^+$, 789 (100) [2M + Na]$^+$.

2-Methyl-4-[(4-methylphenyl)ethynyl]-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (5b): M.p. 197–199 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.28 (s, 3 H, 4′′-CH_3), 2.97 (s, 3 H, *N*-CH_3), 3.03–3.07 (m, 1 H, 5-H), 3.28 (dd, J = 7.6, 5.0 Hz, 1 H, 3a-H), 3.72–3.76 (m, 1 H, 4-H), 4.37 (d, J = 7.6 Hz, 1 H, 11b-H), 7.01 (d, J = 7.1 Hz, 2 H, 3′′,5′′-H), 7.16 (d, J = 7.1 Hz, 2 H, 2′′,6′′-H), 7.39–7.44 (m, 2 H, 7-H, 9-H), 7.66 (ddd, J = 8.5, 7.1, 1.7 Hz, 1 H, 8-H), 8.29 (dd, J = 8.2, 1.7 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.4 (4′′-*C*H$_3$), 24.8 (N-*C*H$_3$), 27.8 (C-4), 33.2 (C-5), 37.7 (C-11b), 43.3 (C-3a), 84.0 (C-1′), 85.9 (C-2′), 112.1 (C-11a), 117.6 (C-9), 118.6 (C-1′′), 123.5 (C-10a), 125.2 (C-7), 126.4 (C-10), 129.0 (C-3′′,5′′), 131.4 (C-2′′,6′′), 133.7 (C-8), 138.8 (C-4′′), 155.7 (C-6a), 162.2 (C-5a), 174.3 (C-3), 176.2 (C-1 and C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{20}NO_4$ [M + H]$^+$ 398.1392, found 398.1372. MS (ESI$^+$): m/z (%) = 398 (100) [M + H]$^+$, 420 (21) [M + Na]$^+$, 817 (27) [2M + Na]$^+$.

4-[(4-Methoxyphenyl)ethynyl]-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (5c): M.p. 203–204 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.97 (s, 3 H, NCH_3), 3.02–3.07 (m, 2 H, 5-H), 3.28 (dd, J = 7.6, 5.0 Hz, 1 H, 3a-H), 3.71–3.75 (m, 1 H, 4-H), 3.75 (s, 3 H, OCH_3), 4.37 (d, J = 7.6 Hz, 1 H, 11b-H), 6.73 (d, J = 8.9 Hz, 2 H, 3′′,5′′-H), 7.20 (d, J = 8.9 Hz, 2 H, 2′′,6′′-H), 7.39–7.44 (m, 2 H, 7-H, 9-H), 7.66 (ddd, J = 8.6, 7.1, 1.7 Hz, 1 H, 8-H), 8.29 (dd, J = 8.2, 1.7 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.7 (N*C*H$_3$), 27.8 (C-4), 33.3 (C-5), 37.7 (C-11b), 43.3 (C-3a), 55.2 (O*C*H$_3$), 83.3 (C-1′), 85.7 (C-2′), 112.1 (C-11a), 113.7 (C-1′′), 113.8 (C-3′′,5′′), 117.6 (C-7), 123.5 (C-10a), 125.2 (C-9), 126.4 (C-10), 133.0 (C-2′′,6′′), 133.7 (C-8), 155.7 (C-6a), 159.7 (C-4′′), 162.3 (C-5a), 174.3 (C-1), 176.2 (C-3 and C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{20}NO_5$ [M + H]$^+$ 414.1341, found 414.1329. MS (ESI$^+$): m/z (%) = 414 (22) [M + H]$^+$, 436 (38) [M + Na]$^+$, 849 (100) [2M + Na]$^+$.

4-[(4-Bromophenyl)ethynyl]-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (5d): M.p. 214–215 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.96 (s, 3 H, CH_3), 3.03–3.08 (m, 2 H, 5-H), 3.29 (dd, J = 7.6, 5.0 Hz, 1 H, 3a-H), 3.70–3.75 (m, 1 H, 4-H), 4.39 (d, J = 7.6 Hz, 1 H, 11b-H), 7.14 (d, J = 8.5 Hz, 2 H, 2′′-H, 6′′-H), 7.36 (d, J = 8.5 Hz, 2 H, 3′′-H, 5′′-H), 7.40–7.45 (m, 2 H, 7-H, 9-H), 7.67 (ddd, J = 8.5, 7.1, 1.6 Hz, 1 H, 8-H), 8.29 (dd, J = 8.2, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.8 (N*C*H$_3$), 27.8 (C-4), 33.0 (C-5), 37.8 (C-11b), 43.2 (C-3a), 84.8 (C-1′), 86.0 (C-2′), 112.1 (C-11a), 117.6 (C-7), 120.6 (C-1′′), 123.0 (C-4′′), 123.8 (C-10a), 125.3 (C-9), 126.4 (C-10), 131.5 (C-3′′ and C-5′′), 133.0 (C-2′′ and C-6′′), 133.8 (C-8), 155.7 (C-6a), 162.3 (C-5a), 175.9 (C-1), 176.0 (C-3 and C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{24}H_{17}{}^{79}BrNO_4$ [M + H]$^+$ 462.0341, found 462.0325; calcd. for $C_{24}H_{17}{}^{81}BrNO_4$ [M + H]$^+$ 464.0320, found 464.0302. MS (ESI$^+$): m/z (%) = 462 (35) [M + H]$^+$, 947 (100) [2M + Na + 2H]$^+$.

4-[(4-Methoxyphenyl)ethynyl]-2-phenyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (5e): M.p. 164–166 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.10–3.13 (m, 2 H, 5-H), 3.37 (dd, J = 7.7, 4.9 Hz, 1 H, 3-H), 3.71 (s, 3 H, 4′′-OCH_3), 3.87–3.90 (m, 1 H, 4-H), 4.56 (d, J = 4.9 Hz, 1 H, 11b-H), 6.64 (d, J = 6.9 Hz, 2 H, 3′′,5′′-H), 7.08 (d, J = 6.9 Hz, 2 H, 2′′,6′′-H), 7.27–7.34 (m, 5 H, 2′′′,3′′′,4′′′,5′′′,6′′′-H), 7.39–7.43 (m, 1 H, 9-H), 7.43 (d, J = 7.7 Hz, 1 H, 7-H), 7.66 (ddd, J = 7.8, 7.7, 1.6 Hz, 1 H, 8-H), 8.28 (dd, J = 8.3, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 27.9 (C-4), 33.8 (C-5), 38.1 (C-11b), 43.0 (C-3a), 55.2 (O*C*H$_3$), 83.5 (C-1′), 86.5 (C-2′), 112.0 (C-11a), 113.66 (C-1′′),

113.73 (C-3′′,5′′), 117.7 (C-7), 123.6 (C-10a), 125.3 (C-9), 126.4 (C-10), 126.6 (C-2′′′,6′′′), 128.4 (C-4′′′), 128.9 (C-3′′′,5′′′), 131.8 (C-1′′′), 133.3 (C-2′′,6′′), 133.7 (C-8), 155.8 (C-6a), 159.7 (C-4′′), 162.4 (C-5a), 173.0 (C-3), 175.3 (C-1), 176.2 (C-11) ppm.

General Procedure for the Synthesis of 4-(Arylethynyl)-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones 6a–d: 2,3-Dichloro-5,6-dicyanobenzoquinone (DDQ) (121 mg, 534 μmol) was added to a solution of the appropriate 4-(arylethynyl)-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones **5a**–**d** (178 μmol) in toluene (10 mL). The mixture was stirred at 100 °C for 1 h. After that period, the solvent was evaporated to dryness and the residue was purified by preparative TLC using CH_2Cl_2 as eluent to give desired 4-(arylethynyl)-2-methylchromeno[3,2-*e*] isoindole-1,3,11-triones **6a**–**d** in good yields (57–83%).

2-Methyl-4-(phenylethynyl)chromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (6a): Yield 42.5 mg (63%), m.p. 273–274 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.26 (s, 3 H, CH_3), 7.42–7.47 (m, 4 H, 9-H, 3′′,4′′,5′′-H), 7.50 (dd, J = 8.3, 0.6 Hz, 1 H, 7-H), 7.71–7.73 (m, 2 H, 2′′,6′′-H), 7.77 (ddd, J = 8.3, 7.1, 1.6 Hz, 1 H, 8-H), 7.89 (s, 1 H, 5-H), 8.36 (dd, J = 8.0, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.5 (NCH_3), 84.0 (C-1′), 100.4 (C-2′), 117.6 (C-7), 118.9 (C-11a), 121.8 (C-11b), 123.0 (C-10a), 124.8 (C-4), 125.1 (C-9), 127.0 (C-5), 127.4 (C-10), 128.6 (C-3′′,5′′), 128.8 (C-3a), 129.9 (C-4′′), 132.4 (C-2′′,6′′), 133.6 (C-1′′), 135.5 (C-8), 155.0 (C-6a), 159.6 (C-5a), 164.1 and 165.9 (C-1 and C-3), 174.0 (C-11) ppm. HRMS (ESI): m/z calcd. for $C_{24}H_{14}NO_4$ [M + H]$^+$ 380.0923, found 380.0907. MS (ESI$^+$): m/z (%) = 380 (40) [M + H]$^+$, 402 (21) [M + Na]$^+$, 781 (100) [2M + Na]$^+$, 797 (27) [2M + K]$^+$.

2-Methyl-4-[(4-methylphenyl)ethynyl]chromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (6b): Yield 53.9 mg (77%), m.p. 326–327 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.41 (s, 3 H, CH_3), 3.26 (s, 3 H, NCH_3), 7.24 (d, J = 8.0 Hz, 2 H, 3′′,5′′-H), 7.45 (t, J = 7.8 Hz, 1 H, 9-H), 7.50 (d, J = 8.3 Hz, 1 H, 7-H), 7.62 (d, J = 8.0 Hz, 2 H, 2′′,6′′-H), 7.77 (ddd, J = 8.3, 7.8, 1.5 Hz, 1 H, 8-H), 7.88 (s, 1 H, 5-H), 8.37 (dd, J = 7.8, 1.5 Hz, 1 H, 10-H) ppm. ^{13}C NMR (500 MHz, $CDCl_3$): δ = 21.8 (CH_3), 24.5 (NCH_3), 83.7 (C-1′), 100.9 (C-2′), 117.6 (C-7), 118.7 (C-1′′), 118.8 (C-3a), 123.0 (C-10a), 125.07 (C-11b), 125.12 (C-9), 126.8 (C-5), 127.4 (C-10), 128.7 (C-11a), 129.4 (C-3′′,5′′), 132.3 (C-2′′,6′′), 133.6 (C-4), 135.4 (C-8), 140.5 (C-4′′), 155.0 (C-6a), 159.7 (C-5a), 164.1 and 166.0 (C-1 and C-3), 174.1 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{16}NO_4$ [M + H]$^+$ 394.1079, found 394.1060. MS (ESI$^+$): m/z (%) = 394 (40) [M + H]$^+$, 416 (72) [M + Na]$^+$, 432 (33) [M + K]$^+$, 809 (100) [2M + Na]$^+$, 825 (43) [2M + K]$^+$.

4-[(4-Methoxyphenyl)ethynyl]-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (6c): Yield 60.5 mg (83%), m.p. 295–297 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 3.26 (s, 3 H, NCH_3), 3.87 (s, 3 H, OCH_3), 6.95 (d, J = 8.9 Hz, 2 H, 3′′,5′′-H), 7.44 (ddd, J = 7.8, 7.3, 1.0 Hz, 1 H, 9-H), 7.50 (dd, J = 8.4, 1.0 Hz, 1 H, 7-H), 7.68 (d, J = 8.9 Hz, 2 H, 2′′,6′′-H), 7.77 (ddd, J = 8.4, 7.3, 1.6 Hz, 1 H, 8-H), 7.85 (s, 1 H, 5-H), 8.37 (dd, J = 7.8, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (126 MHz, $CDCl_3$): δ = 24.5 (NCH_3), 55.4 (OCH_3), 83.4 (C-1′), 101.2 (C-2′), 113.8 (C-1′′), 114.3 (C-3′′,5′′), 117.6 (C-7), 118.6 (C-11a), 123.0 (C-10a), 125.1 (C-9), 125.3 (C-11b), 126.5 (C-5), 127.4 (C-10), 128.5 (C-3a), 133.6 (C-4), 134.2 (C-2′′,6′′), 135.4 (C-8), 155.0 (C-6a), 159.7 (C-5a), 161.0 (C-4′′), 164.2 and 166.1 (C-1 and C-3), 174.1 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{16}NO_5$ [M + H]$^+$ 410.1028, found 410.1007. MS (ESI$^+$): m/z (%) = 410 (14) [M + H]$^+$, 432 (30) [M + Na]$^+$, 448 (15) [M + K]$^+$, 841 (100) [2M + Na]$^+$, 857 (47) [2M + K]$^+$.

4-[(4-Bromophenyl)ethynyl]-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (6d): Yield 46.5 mg (57%), m.p. 332–333 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 3.26 (s, 3 H, NCH_3), 7.45 (ddd, J = 8.0, 7.1, 0.9 Hz, 1 H, 9-H), 7.51 (d, J = 8.2 Hz, 1 H, 7-H), 7.56–7.60 (m, 4 H, 2′′,6′′-H, 3′′,5′′-H), 7.78 (ddd, J = 8.2, 7.1, 1.6 Hz, 1 H, 8-H), 7.88 (s, 1 H, 5-H), 8.37 (dd, J = 8.0, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (126 MHz, $CDCl_3$): δ = 24.5 (NCH_3), 85.0 (C-1′), 99.0 (C-2′), 117.6 (C-7), 119.1 (C-11a), 120.7 (C-1′′), 123.0 (C-10a), 124.3 and 124.5 (C-4 and C-11b), 125.2 (C-9), 126.9 (C-5), 127.5 (C-10), 128.8 (C-3a), 132.0 (C-3′′,5′′), 133.5 (C-4′′), 133.7 (C-2′′,6′′), 135.5 (C-8), 155.0 (C-6a), 159.7 (C-5a), 164.1 and 166.2 (C-1 and C-3), 174.0 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{24}H_{13}{}^{79}BrNO_4$ [M + H]$^+$ 458.0028, found 458.0006; calcd. for $C_{24}H_{13}{}^{81}BrNO_4$ [M + H]$^+$ 460.0007, found 459.9986. MS (ESI$^+$): m/z (%) = 480 (23) [M + Na]$^+$, 497 (12) [M + K]$^+$, 939 (19) [2M + Na]$^+$.

General Procedure for the Synthesis of 4-(4-Aryl-2*H*-1,2,3-triazol-5-yl)-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones 7a–d: Sodium azide (43 mg, 660 μmol) was added to a solution of the appropriate 4-(arylethynyl)-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione **6a**–**d** (132 μmol) in dry DMF (5 mL). The mixture was stirred under reflux for 1 h under nitrogen atmosphere. After that period, the solution was poured into ice (30 g) and water (30 mL), and the pH was adjusted to 4 with diluted HCl. The mixture was vigorously stirred for 15 min, the precipitate was removed by filtration, washed with water (3 × 20 mL) and light petroleum (3 × 20 mL), and recrystallized from ethanol to afford desired xanthone-1,2,3-triazole dyads **7a**–**d** in excellent yields (88–90%).

2-Methyl-4-(4-phenyl-2*H*-1,2,3-triazol-5-yl)chromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (7a): Yield 49.6 mg (89%), m.p. 304–306 °C. ^{1}H NMR [500 MHz, $(CD_3)_2SO$ + TFA]: δ = 2.94 (s, 3 H, NCH_3), 7.30–7.34 (m, 3 H, 3′′,4′′,5′′-H), 7.47 (dd, J = 7.8, 1.7 Hz, 2 H, 2′′,6′′-H), 7.55 (ddd, J = 7.7, 7.3, 0.9 Hz, 1 H, 9-H), 7.68 (d, J = 8.4 Hz, 1 H, 7-H), 7.91 (ddd, J = 8.4, 7.3, 1.5 Hz, 1 H, 8-H), 8.04 (s, 1 H, 5-H), 8.24 (dd, J = 7.7, 1.5 Hz, 1 H, 10-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 24.2 (NCH_3), 118.0 (C-7), 119.4 (C-11a), 122.7 (C-10a), 125.3 (C-9), 126.4 (C-5), 126.5 (C-10), 126.9 (C-2′′,6′′), 127.5 (C-3a), 128.4 (C-4′′), 128.9 (C-3′′,5′′), 129.9 (C-1′′), 133.7 and 133.8 (C-4 and C-11b), 136.0 (C-8), 136.8 (C-5′), 142.7 (C-4′), 154.8 (C-6a), 159.5 (C-5a), 163.9 and 165.4 (C-1 and C-3), 173.6 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{24}H_{15}N_4O_4$ [M + H]$^+$ 423.1093, found 423.1076. MS (ESI$^+$): m/z (%) = 423 (100) [M + H]$^+$, 445 (10) [M + Na]$^+$.

2-Methyl-4-[4-(4-methylphenyl)-2*H*-1,2,3-triazol-5-yl]chromeno-[3,2-*e*]isoindole-1,3,11(2*H*)-trione (7b): Yield 51.8 mg (90%), m.p. 318–320 °C. ^{1}H NMR [500 MHz, $(CD_3)_2SO$ + TFA]: δ = 2.23 (s, 3 H, CH_3), 2.92 (s, 3 H, NCH_3), 7.09 (d, J = 8.1 Hz, 2 H, 3′′,5′′-H), 7.32 (d, J = 8.1 Hz, 2 H, 2′′,6′′-H), 7.49 (t, J = 7.6 Hz, 1 H, 9-H), 7.61 (d, J = 8.0 Hz, 1 H, 7-H), 7.86 (ddd, J = 8.0, 7.6, 1.6 Hz, 1 H, 8-H), 7.96 (s, 1 H, 5-H), 8.21 (dd, J = 7.6, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 20.9 (CH_3), 24.3 (NCH_3), 118.2 (C-7), 119.7 (C-11a), 123.0 (C-10a), 125.5 (C-9), 126.6 (C-5), 126.8 (C-10), 127.1 (C-2′′,6′′), 127.2 (C-1′′), 127.8 (C-3a), 129.7 (C-3′′,5′′), 134.0 and 134.2 (C-4 and C-11b), 136.1 (C-8), 136.8 (C-5′), 138.2 (C-4′′), 142.6 (C-4′), 155.1 (C-6a), 159.8 (C-5a), 164.2 and 165.7 (C-1 and C-3), 173.9 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{17}N_4O_4$ [M + H]$^+$ 437.1250, found 437.1228. MS (ESI$^+$): m/z (%) = 437 (100) [M + H]$^+$, 459 (71) [M + Na]$^+$, 475 (38) [M + K]$^+$, 895 (78) [2M + Na]$^+$.

4-[4-(4-Methoxyphenyl)-2*H*-1,2,3-triazol-5-yl]-2-methylchromeno-[3,2-*e*]isoindole-1,3,11(2*H*)-trione (7c): Yield 52.5 mg (88%), m.p. 297–299 °C. ^{1}H NMR [500 MHz, $(CD_3)_2SO$ + TFA]: δ = 2.93 (s, 3

H, NCH_3), 3.69 (s, 3 H, OCH_3), 6.84 (d, J = 8.9 Hz, 2 H, 3′′,5′′-H), 7.36 (d, J = 8.9 Hz, 2 H, 2′′,6′′-H), 7.50 (ddd, J = 7.8, 7.3, 0.8 Hz, 1 H, 9-H), 7.63 (d, J = 8.3 Hz, 1 H, 7-H), 7.87 (ddd, J = 8.3, 7.3, 1.5 Hz, 1 H, 8-H), 7.97 (s, 1 H, 5-H), 8.22 (dd, J = 7.8, 1.5 Hz, 1 H, 10-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 24.2 (N*C*H$_3$), 55.2 (O*C*H$_3$), 114.5 (C-3′′,5′′), 118.1 (C-7), 119.5 (C-11a), 122.2 (C-1′′), 122.9 (C-10a), 125.4 (C-9), 126.5 (C-5), 126.6 (C-10), 127.7 (C-3a), 128.4 (C-2′′,6′′), 133.8 and 134.2 (C-4 and C-11b), 136.1 (C-8), 136.3 (C-5′), 142.1 (C-4′), 154.9 (C-6a), 159.6 and 159.7 (C-4′′ and C-5a), 164.1 and 165.6 (C-1 and C-3), 173.8 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{17}N_4O_5$ [M + H]$^+$ 453.1199, found 453.1177. MS (ESI$^+$): m/z (%) = 437 (16) [M – CH$_3$]$^+$, 453 (100) [M + H]$^+$, 475 (77) [M + Na]$^+$, 491 (33) [M + K]$^+$, 927 (64) [2M + Na]$^+$.

4-[4-(4-Bromophenyl)-2*H*-1,2,3-triazol-5-yl]-2-methylchromeno-[3,2-*e*]isoindole-1,3,11(2*H*)-trione (7d): Yield 59.5 mg (90%), m.p. 319–320 °C. ^{1}H NMR [500 MHz, $(CD_3)_2SO$ + TFA]: δ = 2.92 (s, 3 H, NCH_3), 7.39 (d, J = 8.7 Hz, 2 H, 2′′,6′′-H), 7.47 (d, J = 8.7 Hz, 2 H, 3′′,5′′-H), 7.51 (ddd, J = 7.8, 7.3, 0.9 Hz, 1 H, 9-H), 7.64 (dd, J = 8.2, 0.9 Hz, 1 H, 7-H), 7.88 (ddd, J = 8.2, 7.3, 1.6 Hz, 1 H, 8-H), 8.02 (s, 1 H, 5-H), 8.22 (dd, J = 7.8, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 24.3 (N*C*H$_3$), 118.2 (C-7), 119.7 (C-11a), 121.9 (C-4′′), 122.9 (C-10a), 125.5 (C-9), 126.6 (C-5), 126.7 (C-10), 127.6 (C-3a), 129.0 (C-2′′,6′′), 129.7 (C-1′′), 132.1 (C-3′′,5′′), 133.5 and 134.0 (C-4 and C-11b), 136.2 (C-8), 139.3 (C-5′), 142.5 (C-4′), 155.0 (C-6a), 159.8 (C-5a), 164.1 and 165.7 (C-1 and C-3), 173.8 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{24}H_{14}{}^{79}BrN_4O_4$ [M + H]$^+$ 501.0198, found 501.0180; calcd. for $C_{24}H_{14}{}^{81}BrN_4O_4$ [M + H]$^+$ 503.0178, found 503.0155. MS (ESI$^+$): m/z (%) = 501 (100) [M + H]$^+$.

General Procedure for the Synthesis of (*E*)-2-[2-(4-Aryl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-ones (8a–d): Sodium azide (117 mg, 1.8 mmol) was added to a solution of the appropriate (*E*)-2-(4-arylbut-1-en-3-yn-1-yl)-4*H*-chromen-4-ones **4a**–**d** (367 μmol) in dry DMF (5 mL). The mixture was stirred under reflux for 1 h under nitrogen atmosphere. After that period, the solution was poured into ice (30 g) and water (30 mL), and the pH was adjusted to 4 with diluted HCl. The mixture was vigorously stirred for 15 min, the precipitate was washed with water (3 × 20 mL) and light petroleum (3 × 20 mL), removed by filtration and recrystallized from ethanol to give (*E*)-2-[2-(4-aryl-2*H*-1,2,3-triazol-5-yl) vinyl]-4*H*-chromen-4-ones **8a**–**d** in excellent yields (90–97%).

(*E*)-2-[2-(4-Phenyl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-one (8a): Yield 0.10 g (90%), m.p. 240–241 °C. ^{1}H NMR [300 MHz, $(CD_3)_2SO$ + TFA]: δ = 6.58 (s, 1 H, 3-H), 7.35 (d, J = 15.8 Hz, 1 H, α-H), 7.46 (t, J = 7.7 Hz, 1 H, 6-H), 7.53 (t, J = 7.5 Hz, 1 H, 4′′-H), 7.61 (t, J = 7.5 Hz, 2 H, 3′′,5′′-H), 7.65 (d, J = 8.1 Hz, 1 H, 8-H), 7.68 (d, J = 15.8 Hz, 1 H, β-H), 7.74 (d, J = 7.5 Hz, 2 H, 2′′,6′′-H), 7.77 (ddd, J = 8.1, 7.7, 1.5 Hz, 1 H, 7-H), 8.06 (dd, J = 7.7, 1.5 Hz, 1 H, 5-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 111.1 (C-3), 118.7 (C-8), 123.1 (C-α), 123.9 (C-4a), 124.0 (C-β), 125.3 (C-5), 125.6 (C-6), 128.5 (C-2′′,6′′), 129.3 (C-1′′), 129.4 (C-4′′), 129.6 (C-3′′,5′′), 134.5 (C-7), 138.3 (C-4′), 142.6 (C-5′), 156.0 (C-8a), 161.5 (C-2), 177.7 (C-4) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{19}H_{14}N_3O_2$ [M + H]$^+$ 316.1086, found 316.1075. MS (ESI$^+$): m/z (%) = (100) 316 [M + H]$^+$, 653 (22) [2M + Na]$^+$.

(*E*)-2-{2-[4-(4-Methylphenyl)-2*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (8b): Yield 0.12 g (98%), m.p. 239–240 °C. ^{1}H NMR [300 MHz, $(CD_3)_2SO$ + TFA]: δ = 2.42 (s, 3 H, CH_3), 6.61 (s, 1 H, 3-H), 7.34 (d, J = 15.8 Hz, 1 H, α-H), 7.41 (d, J = 7.7 Hz, 2 H, 3′′,5′′-H), 7.48 (t, J = 7.7 Hz, 1 H, 6-H), 7.61 (d, J = 7.7 Hz, 2 H, 2′′,6′′-H), 7.64 (d, J = 15.8 Hz, 1 H, β-H), 7.70 (d, J = 8.0 Hz, 1 H, 8-H), 7.80 (ddd, J = 8.0, 7.7, 1.4 Hz, 1 H, 7-H), 8.03 (dd, J = 7.7, 1.4 Hz, 1 H, 5-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 21.0 (*C*H$_3$), 110.7 (C-3), 118.5 (C-8), 122.4 (C-α), 123.6 (C-β), 123.8 (C-4a), 124.9 (C-5), 125.4 (C-6), 125.7 (C-1′′), 128.1 (C-2′′ and C-6′′), 129.9 (C-3′′ and C-5′′), 134.3 (C-7), 137.8 (C-4′), 138.8 (C-4′′), 141.8 (C-5′), 155.5 (C-8a), 161.1 (C-2), 177.3 (C-4) ppm. HRMS-EI: m/z calcd. for $C_{20}H_{15}O_2N_3$ M$^{+\cdot}$ 329.1164, found 329.1166. MS (ESI$^+$): m/z (%) = 330 (100) [M + H]$^+$, 352 (15) [M + Na]$^+$.

(*E*)-2-{2-[4-(4-Methoxyphenyl)-2*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (8c): Yield 0.12 g (98%), m.p. 237–238 °C. ^{1}H NMR [500 MHz, $(CD_3)_2SO$ + TFA]: δ = 3.85 (s, 3 H, OCH_3), 6.60 (s, 1 H, 3-H), 7.16 (d, J = 8.8 Hz, 2 H, 3′′,5′′-H), 7.32 (d, J = 15.8 Hz, 1 H, α-H), 7.46 (ddd, J = 7.7, 7.2, 1.0 Hz, 1 H, 6-H), 7.62 (d, J = 15.8 Hz, 1 H, β-H), 7.64 (d, J = 8.8 Hz, 2 H, 2′′,6′′-H), 7.70 (d, J = 8.1 Hz, 1 H, 8-H), 7.79 (ddd, J = 8.1, 7.2, 1.6 Hz, 1 H, 7-H), 8.01 (dd, J = 7.7, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR [126 MHz, $(CD_3)_2SO$ + TFA]: δ = 55.4 (O*C*H$_3$), 110.6 (C-3), 114.8 (C-3′′,5′′), 118.5 (C-8), 120.7 (C-1′′), 122.2 (C-α), 123.6 (C-4a), 123.9 (C-β), 124.9 (C-5), 125.4 (C-6), 129.6 (C-2′′,6′′), 134.3 (C-7), 137.6 (C-4′), 145.1 (C-5′), 155.5 (C-8a), 160.1 (C-4′′), 161.2 (C-2), 177.2 (C-4) ppm. HRMS-EI: m/z calcd. for $C_{20}H_{15}O_3N_3$ M$^{+\cdot}$ 345.1113, found 345.1116. MS (ESI$^+$): m/z (%) = 346 (100) [M + H]$^+$, 368 (15) [M + Na]$^+$.

(*E*)-2-{2-[4-(4-Bromophenyl)-2*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (8d): Yield 0.14 g (97%), m.p. 293–294 °C. ^{1}H NMR [300 MHz, $(CD_3)_2SO$ + TFA]: δ = 6.58 (s, 1 H, 3-H), 7.30 (d, J = 15.8 Hz, 1 H, α-H), 7.42 (ddd, J = 7.7, 7.4, 1.3 Hz, 1 H, 6-H), 7.61 (d, J = 15.8 Hz, 1 H, β-H), 7.64 (d, J = 8.5 Hz, 2 H, 2′′,6′′-H), 7.68 (d, J = 7.8 Hz, 1 H, 8-H), 7.73–7.78 (m, 1 H, 7-H), 7.75 (d, J = 8.5 Hz, 2 H, 3′′,5′′-H), 8.00 (dd, J = 7.7, 1.3 Hz, 1 H, 5-H) ppm. ^{13}C NMR [75 MHz, $(CD_3)_2SO$ + TFA]: δ = 111.1 (C-3), 118.8 (C-8), 122.8 (C-4′′), 123.5 (C-α, C-β), 123.9 (C-4a), 125.1 (C-5), 125.6 (C-6), 128.7 (C-1′′), 130.3 (C-2′′,6′′), 132.5 (C-3′′,5′′), 134.5 (C-7), 138.2 (C-4′), 142.2 (C-5′), 155.8 (C-8a), 161.2 (C-2), 177.6 (C-4) ppm. HRMS-EI: m/z calcd. for $C_{19}H_{12}O_2N_3{}^{79}Br$ M$^{+\cdot}$ 393.0113, found 393.0122; calcd. for $C_{19}H_{12}O_2N_3{}^{81}Br$ M$^{+\cdot}$ 395.0092, found 395.0106. MS (ESI$^+$): m/z (%) = 394 (39) [M + H]$^+$ (^{79}Br); (37) 396 [M + H]$^+$ (^{81}Br).

General Procedure for the Synthesis of (*E*)-2-[2-(4-Aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-ones 9a–d, (*E*)-2-[2-(4-Aryl-3-methyl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-ones 10a–d and (*E*)-2-[2-(4-Aryl-1-methyl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-ones 11a–d: Dimethyl sulfate (0.1 mL, 1.1 mmol) was added dropwise to a solution of the appropriate (*E*)-2-[2-(4-aryl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-one **8a**–**d** (1 mmol) in acetone (10 mL) with potassium carbonate (0.36 g, 3 mmol). The mixture was stirred under reflux for 1 h. The inorganic salts were removed by filtration and washed with acetone (3 × 30 mL). The solvent was evaporated to dryness and the residue was purified by silica column chromatography using CH_2Cl_2 as eluent. Two fractions were obtained: 1,2,3-triazole derivatives **9a**–**d** (higher R_f value) in 67–82% yield and a mixture of **10a**–**d** and **11a**–**d** in 18–33% yield.

(*E*)-2-[2-(2-Methyl-4-phenyl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-one (9a): Yield 221 mg (67%), m.p. 169–171 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 4.29 (s, 3 H, NCH_3), 6.34 (s, 1 H, 3-H), 7.12 (d, J = 15.8 Hz, 1 H, α-H), 7.39 (ddd, J = 8.0, 7.1, 0.8 Hz, 1 H, 6-H), 7.47 (dd, J = 8.2, 0.8 Hz, 1 H, 8-H), 7.48–7.57 (m, 3 H, 3′′,4′′,5′′-H), 7.61 (d, J = 15.8 Hz, 1 H, β-H), 7.64–7.70 (m, 3 H, 7-H, 2′′,6′′-H), 8.19 (dd, J = 8.0, 1.5 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 42.2 (N*C*H$_3$), 111.3 (C-3), 117.9

(C-8), 123.2 (C-α), 124.0 (C-β), 124.1 (C-4a), 125.1 (C-6), 125.7 (C-5), 128.3 (C-2′′,6′′), 128.9 (C-4′′), 129.0 (C-3′′,5′′), 130.1 (C-1′′), 133.8 (C-8), 140.5 (C-5′), 147.4 (C-4′), 155.9 (C-8a), 161.1 (C-2), 178.5 (C-4) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{20}H_{16}N_3O_2$ [M + H]$^+$ 330.1243, found 330.1231. MS (ESI$^+$): *m*/*z* (%) = 330 (100) [2M + H]$^+$, 659 (7) [2M + H]$^+$.

Mixture of 10a and 11a (86:14): Yield 109 mg (33%). **(*E*)-2-[2-(3-Methyl-4-phenyl-1*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-one (10a)**: ^{1}H NMR (500 MHz, $CDCl_3$): δ = 4.02 (s, 3 H, NCH_3), 6.30 (s, 1 H, 3-H), 7.21 (d, *J* = 15.8 Hz, 1 H, α-H), 7.36 (d, *J* = 15.8 Hz, 1 H, β-H), 7.38 (ddd, *J* = 7.7, 7.5, 1.0 Hz, 1 H, 6-H), 7.41–7.43 (m, 3 H, 8-H, 2′′,6′′-H), 7.61–7.65 (m, 4 H, 7-H, 3′′,4′′,5′′-H), 8.17 (dd, *J* = 7.5, 1.5 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 35.5 (N*C*H$_3$), 110.9 (C-3), 117.9 (C-8), 122.0 (C-α), 124.0 (C-β), 124.1 (C-4a), 125.0 (C-6), 125.7 (C-5), 126.1 (C-1′′), 129.5 (C-2′′,6′′), 129.6 (C-3′′,5′′), 130.2 (C-4′′), 133.6 (C-7), 137.0 (C-4′), 141.3 (C-5′), 155.9 (C-8a), 161.4 (C-2), 178.5 (C-4) ppm. **(*E*)-2-[2-(1-Methyl-4-phenyl-1*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-one (11a)**: ^{1}H NMR (500 MHz, $CDCl_3$): δ = 4.27 (s, 3 H, NCH_3), 6.27 (s, 1 H, 3-H) ppm.

(*E*)-2-{2-[2-Methyl-4-(4-methylphenyl)-2*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (9b): Yield 257 mg (75%), m.p. 192–194 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.46 (s, 3 H, CH_3), 4.29 (s, 3 H, NCH_3), 6.49 (s, 1 H, 3-H), 7.13, (d, *J* = 15.8 Hz, 1 H, α-H), 7.34 (d, *J* = 8.0 Hz, 2 H, 3′′,5′′-H), 7.42 (ddd, *J* = 7.7, 7.4, 1.1 Hz, 1 H, 6-H), 7.50 (dd, *J* = 8.4, 1.1 Hz, 1 H, 8-H), 7.55 (d, *J* = 8.0 Hz, 2 H, 2′′,6′′-H), 7.64 (d, *J* = 15.8 Hz, 1 H, β-H), 7.70 (ddd, *J* = 8.4, 7.4, 1.6 Hz, 1 H, 7-H), 8.21 (dd, *J* = 7.7, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.4 (*C*H$_3$), 42.2 (N*C*H$_3$), 110.8 (C-3), 118.0 (C-8), 122.7 (C-α), 123.6 (C-4a), 124.9 (C-β), 125.3 (C-6), 125.7 (C-5), 127.1 (C-1′′), 128.2 (C-2′′,6′′), 129.8 (C-3′′,5′′), 134.1 (C-7), 139.1 (C-4′), 140.3 (C-5′), 147.7 (C-4′′), 156.0 (C-8a), 161.9 (C-2), 178.5 (C-4) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{21}H_{18}N_3O_2$ [M + H]$^+$ 344.1399, found 344.1381. MS (ESI$^+$): *m*/*z* (%) = 344 (100) [M + H]$^+$, 687 (11) [2M + H]$^+$.

Mixture of 10b and 11b (81:19): Yield 86 mg (25%). **(*E*)-2-{2-[3-Methyl-4-(4-methylphenyl)-1*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (10b)**: ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.51 (s, 3 H, CH_3), 4.01 (s, 3 H, NCH_3), 6.31 (s, 1 H, 3-H), 7.20 (d, *J* = 15.9 Hz, 1 H, α-H), 7.30 (d, *J* = 8.1 Hz, 2 H, 2′′,6′′-H), 7.32–7.38 (m, 1 H, 6-H), 7.37 (d, *J* = 15.9 Hz, 1 H, β-H), 7.41–7.45 (m, 3 H, 8-H, 3′′,5′′-H), 7.64 (ddd, *J* = 8.5, 7.1, 1.6 Hz, 1 H, 7-H), 8.17 (dd, *J* = 7.9, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.5 (*C*H$_3$), 35.5 (N*C*H$_3$), 110.8 (C-3), 117.9 (C-8), 121.8 (C-α), 123.0 (C-4a), 124.0 (C-1′′), 124.3 (C-β), 125.0 (C-6), 125.7 (C-5), 129.4 (C-2′′,6′′), 130.2 (C-3′′,5′′), 133.7 (C-7), 137.2 (C-4′), 140.6 (C-4′′), 141.1 (C-5′), 155.9 (C-8a), 161.6 (C-2), 178.5 (C-4) ppm. **(*E*)-2-{2-[1-Methyl-4-(4-methylphenyl)-1*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (11b)**: ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.2 (s, 3 H, CH_3), 3.97 (s, 3 H, NCH_3), 6.40 (s, 1 H, 3-H) ppm.

(*E*)-2-{2-[4-(4-Methoxyphenyl)-2-methyl-2*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (9c): Yield 280 mg (78%), m.p. 186–187 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.90 (s, 3 H, OCH_3), 4.28 (s, 3 H, NCH_3), 6.34 (s, 1 H, 3-H), 7.06 (d, *J* = 8.8 Hz, 2 H, 3′′,5′′-H), 7.10 (d, *J* = 15.8 Hz, 1 H, α-H), 7.40 (ddd, *J* = 7.7, 7.3, 1.1 Hz, 1 H, 6-H), 7.48 (d, *J* = 8.2 Hz, 1 H, 8-H), 7.59 (d, *J* = 15.8 Hz, 1 H, β-H), 7.59 (d, *J* = 8.8 Hz, 2 H, 2′′,6′′-H), 7.67 (ddd, *J* = 8.2, 7.3, 1.6 Hz, 1 H, 7-H), 8.20 (dd, *J* = 7.7, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 42.1 (N*C*H$_3$), 55.4 (O*C*H$_3$), 111.1 (C-3), 114.4 (C-3′′,5′′), 117.9 (C-8), 122.5 (C-1′′), 122.9 (C-α), 124.1 (C-4a), 124.2 (C-β), 125.0 (C-6), 125.7 (C-5), 129.6 (C-2′′,6′′), 133.7 (C-7), 140.2 (C-5′), 147.3 (C-4′), 155.9 (C-8a), 160.2 (C-4′′), 161.1 (C-2), 178.5 (C-4) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{21}H_{18}N_3O_3$ [M + H]$^+$ 360.1348, found 360.1332. MS (ESI$^+$): *m*/*z* (%) = 344 (4) [M – CH_3]$^+$, 360 (100) [M + H]$^+$, 719 (10) [2M + H]$^+$.

Mixture of 10c and 11c (87:13): Yield 79 mg (22%). **(*E*)-2-{2-[4-(4-Methoxyphenyl)-3-methyl-1*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (10c)**: ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.94 (s, 3 H, OCH_3), 4.00 (s, 3 H, NCH_3), 6.30 (s, 1 H, 3-H), 7.13 (d, *J* = 8.8 Hz, 2 H, 3′′,5′′-H), 7.19 (d, *J* = 15.9 Hz, 1 H, α-H), 7.33–7.40 (m, 4 H, β-H, 6-H, 2′′,6′′-H), 7.43 (dd, *J* = 8.5, 0.6 Hz, 1 H, 8-H), 7.64 (ddd, *J* = 8.5, 7.0, 1.6 Hz, 1 H, 7-H), 8.16 (dd, *J* = 7.9, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 35.4 (N*C*H$_3$), 55.5 (O*C*H$_3$), 110.8 (C-3), 115.0 (C-3′′,5′′), 117.9 (C-8), 121.6 (C-α), 124.1 (C-1′′), 124.3 (C-β), 125.0 (C-6), 125.7 (C-5), 130.9 (C-2′′,6′′), 133.6 (C-7), 137.0 (C-4′), 141.2 (C-5′), 155.9 (C-8a), 161.0 (C-4′′), 161.5 (C-2), 178.5 (C-4) ppm. **(*E*)-2-{2-[4-(4-Methoxyphenyl)-1-methyl-1*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (11c)**: ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.88 (s, 3 H, OCH_3), 4.25 (s, 3 H, NCH_3), 6.27 (s, 1 H, 3-H) ppm.

(*E*)-2-{2-[4-(4-Bromophenyl)-2-methyl-2*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (9d): Yield 335 mg (82%), m.p. 223–224 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 4.29 (s, 3 H, NCH_3), 6.50 (s, 1 H, 3-H), 7.15 (d, *J* = 15.8 Hz, 1 H, α-H), 7.43 (dt, *J* = 7.7, 0.8 Hz, 1 H, 6-H), 7.52 (dd, *J* = 7.6, 0.8 Hz, 1 H, 8-H), 7.54 (d, *J* = 8.5 Hz, 2 H, 2′′,6′′-H), 7.58 (d, *J* = 15.8 Hz, 1 H, β-H), 7.67 (d, *J* = 8.5 Hz, 2 H, 3′′,5′′-H), 7.68–7.74 (m, 1 H, 7-H), 8.21 (dd, *J* = 7.7, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 42.2 (N*C*H$_3$), 111.1 (C-3), 117.9 (C-8), 123.3 (C-4′′), 123.5 (C-β), 123.7 (C-α), 124.1 (C-4a), 125.2 (C-6), 125.7 (C-5), 129.1 (C-1′′), 129.8 (C-2′′,6′′), 132.2 (C-3′′,5′′), 133.9 (C-7), 140.5 (C-5′), 146.3 (C-4′), 155.9 (C-8a), 160.9 (C-2), 178.5 (C-4) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{20}H_{15}{}^{79}BrN_3O_2$ [M + H]$^+$ 408.0348, found 408.0330; calcd. for $C_{20}H_{15}{}^{81}BrN_3O_2$ [M + H]$^+$ 410.0327, found 410.0306. MS (ESI$^+$): *m*/*z* (%) = 408 (100) [M + H]$^+$, 430 (15) [M + Na]$^+$.

Mixture of 10d and 11d (93:7): Yield 73 mg (18%). **(*E*)-2-{2-[4-(4-Bromophenyl)-1-methyl-1*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (10d)**: ^{1}H NMR (500 MHz, $CDCl_3$): δ = 4.02 (s, 3 H, NCH_3), 6.33 (s, 1 H, 3-H), 7.26–7.31 (m, 2 H, α-H, β-H), 7.30 (d, *J* = 8.5 Hz, 2 H, 2′′,6′′-H), 7.38 (ddd, *J* = 7.9, 7.1, 0.7 Hz, 1 H, 6-H), 7.45 (dd, *J* = 8.5, 0.7 Hz, 1 H, 8-H), 7.65 (ddd, *J* = 8.5, 7.1, 1.6 Hz, 1 H, 7-H), 7.77 (d, *J* = 8.5 Hz, 2 H, 3′′,5′′-H), 8.18 (dd, *J* = 7.9, 1.6 Hz, 1 H, 5-H) ppm. ^{13}C NMR (125 MHz, $CDCl_3$): δ = 35.5 (N*C*H$_3$), 111.2 (C-3), 117.9 (C-8), 122.5 (C-β), 123.3 (C-α), 124.1 (C-4a), 124.9 and 125.0 (C-1′′ and C-4′′), 125.1 (C-6), 125.7 (C-5), 131.1 (C-2′′,6′′), 132.9 (C-3′′,5′′), 133.7 (C-7), 135.9 (C-4′), 141.5 (C-5′), 155.9 (C-8a), 161.2 (C-2), 178.4 (C-4) ppm. **(*E*)-2-{2-[4-(4-Bromophenyl)-3-methyl-1*H*-1,2,3-triazol-5-yl]vinyl}-4*H*-chromen-4-one (11d)**: ^{1}H NMR (500 MHz, $CDCl_3$): δ = 4.03 (s, 3 H, NCH_3), 6.30 (s, 1 H, 3-H) ppm.

General Procedure for the Synthesis of 4-(4-Aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones (12a–d). Method A: *N*-Methylmaleimide (0.10 g, 0.92 mmol) was added to a solution of the appropriate (*E*)-2-[2-(4-aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)vinyl]-4*H*-chromen-4-ones **9a**–**d** (0.18 mmol) in dry DMF (5 μL). The mixture was heated at 160 °C under microwave irradiation (multimode apparatus) for 20 min. The residue was dissolved in CH_2Cl_2 and purified by preparative TLC using CH_2Cl_2/EtOAc (4:1) as eluent to give desired cycloadducts **12a**–**d** in the following yields: **12a** (29.3 mg, 37%), **12b** (27.0 mg, 33%), **12c** (33.9 mg, 40%), **12d** (28.0 mg, 30%).

Method B: *N*-Methylmaleimide (0.10 g, 0.92 mmol) was mixed with the appropriate (*E*)-2-[2-(4-aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)

vinyl]-4*H*-chromen-4-ones **9a**–**d** (0.18 mmol) in a close vessel. The mixture was heated at 200 °C under microwave irradiation (monomode apparatus) for 10 min. The residue was dissolved in CH_2Cl_2 and purified by preparative TLC using CH_2Cl_2/EtOAc (4:1) as eluent to give desired cycloadducts **12a**–**d** in the following yields: **12a** (28.5 mg, 36%), **12b** (39.3 mg, 48%), **12c** (33.9 mg, 40%), **12d** (35.5 mg, 38%).

2-Methyl-4-(2-methyl-4-phenyl-2*H*-1,2,3-triazol-5-yl)-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (12a): M.p. 152–153 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.82 (s, 3 H, 2-NCH_3), 2.98–3.24 (m, 2 H, 5-H), 3.47 (dd, J = 8.8, 6.4 Hz, 1 H, 3a-H), 3.84–3.91 (m, 1 H, 4-H), 4.18 (s, 3 H, 2′-NCH_3), 4.64 (d, J = 8.8 Hz, 1 H, 11b-H), 7.40–7.49 (m, 5 H, 7-H, 9-H, 3′′,4′′,5′′-H), 7.61 (dd, J = 8.1, 1.5 Hz, 2 H, 2′′,6′′-H), 7.67 (ddd, J = 8.5, 7.1, 1.6 Hz, 1 H, 8-H), 8.30 (dd, J = 8.4, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.9 (2-NCH_3), 30.6 (C-5), 32.4 (C-4), 37.9 (C-11b), 41.2 (C-3a), 41.7 (2′-NCH_3), 113.3 (C-11a), 117.7 (C-7), 123.7 (C-10a), 125.3 (C-9), 126.4 (C-10), 127.7 (C-2′′,6′′), 128.6 (C-4′′), 129.0 (C-3′′,5′′), 130.9 (C-1′′), 133.7 (C-8), 142.3 (C-5′), 145.1 (C-4′), 155.7 (C-6a), 164.1 (C-5a), 174.7 (C-3), 175.2 (C-1), 176.1 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{21}N_4O_4$ [M + H]$^+$ 441.1563, found 441.1539. MS (ESI$^+$): m/z (%) = 441 (100) [M + H]$^+$, 463 (17) [M + Na]$^+$, 479 (10) [M + K]$^+$.

2-Methyl-4-[2-methyl-4-(4-methylphenyl)-2*H*-1,2,3-triazol-5-yl]-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (12b): M.p. 143–145 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.40 (s, 3 H, CH_3), 2.82 (s, 3 H, 2-NCH_3), 2.98–3.24 (m, 2 H, 5-H), 3.48 (dd, J = 8.8, 6.4 Hz, 1 H, 3a-H), 3.82–3.89 (m, 1 H, 4-H), 4.17 (s, 3 H, 2′-NCH_3), 4.63 (d, J = 8.8 Hz, 1 H, 11b-H), 7.26 (d, J = 8.0 Hz, 2 H, 3′′,5′′-H), 7.40–7.45 (m, 1 H, 9-H), 7.41 (d, J = 8.2 Hz, 1 H, 7-H), 7.49 (d, J = 8.0 Hz, 2 H, 2′′,6′′-H), 7.67 (ddd, J = 8.2, 7.2, 1.5 Hz, 1 H, 8-H), 8.29 (dd, J = 8.2, 1.5 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 21.3 (CH_3), 24.9 (2-NCH_3), 30.5 (C-5), 32.4 (C-4), 37.9 (C-11b), 41.1 (C-3a), 41.7 (2′-NCH_3), 113.3 (C-11a), 117.7 (C-7), 123.7 (C-10a), 125.3 (C-9), 126.4 (C-10), 127.5 (C-2′′,6′′), 127.9 (C-1′′), 129.7 (C-3′′,5′′), 133.7 (C-8), 138.6 (C-4′′), 142.2 (C-5′), 145.1 (C-4′), 155.7 (C-6a), 164.2 (C-5a), 174.8 (C-3), 175.2 (C-1), 176.0 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{26}H_{23}N_4O_4$ [M + H]$^+$ 455.1719, found 455.1699. MS (ESI$^+$): m/z (%) = 455 (100) [M + H]$^+$, 477 (26) [M + Na]$^+$.

4-[4-(4-Methoxyphenyl)-2-methyl-2*H*-1,2,3-triazol-5-yl]-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (12c): M.p. 167–169 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.82 (s, 3 H, 2-NCH_3), 2.97–3.23 (m, 2 H, 5-H), 3.48 (dd, J = 8.8, 6.4 Hz, 1 H, 3a-H), 3.79–3.90 (m, 1 H, 4-H), 3.85 (s, 3 H, OCH_3), 4.16 (s, 3 H, 2′-NCH_3), 4.64 (d, J = 8.8 Hz, 1 H, 11b-H), 6.98 (d, J = 8.8 Hz, 2 H, 3′′,5′′-H), 7.41 (d, J = 8.2 Hz, 1 H, 7-H), 7.40–7.45 (m, 1 H, 9-H), 7.53 (d, J = 8.8 Hz, 2 H, 2′′,6′′-H), 7.67 (ddd, J = 8.2, 7.1, 1.6 Hz, 1 H, 8-H), 8.29 (dd, J = 8.4, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.9 (2-NCH_3), 30.6 (C-5), 32.4 (C-4), 37.9 (C-11b), 41.2 (C-3a), 41.7 (2′-NCH_3), 55.3 (OCH_3), 113.3 (C-11a), 114.4 (C-3′′,5′′), 117.7 (C-7), 123.2 (C-1′′), 123.6 (C-10a), 125.3 (C-9), 126.4 (C-10), 129.0 (C-2′′,6′′), 133.7 (C-8), 142.0 (C-4′), 145.0 (C-5′), 155.7 (C-6a), 159.9 (C-4′′), 164.1 (C-5a), 174.7 (C-3), 175.2 (C-1), 176.0 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{26}H_{23}N_4O_5$ [M + H]$^+$ 471.1668, found 471.1645. MS (ESI$^+$): m/z (%) = 471 (100) [M + H]$^+$, 493 (8) [M + Na]$^+$.

4-[4-(4-Bromophenyl)-2-methyl-2*H*-1,2,3-triazol-5-yl]-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (12d): M.p. 145–146 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.81 (s, 3 H, 2-NCH_3), 2.96–3.24 (m, 2 H, 5-H), 3.46 (dd, J = 8.8, 6.4 Hz, 1 H, 3a-H), 3.82–3.88 (m, 1 H, 4-H), 4.16 (s, 3 H, 2′-NCH_3), 4.64 (d, J = 8.8 Hz, 1 H, 11b-H), 7.40–7.45 (m, 1 H, 9-H), 7.41 (d, J = 8.1 Hz, 1 H, 7-H), 7.49 (d, J = 8.6 Hz, 2 H, 2′′,6′′-H), 7.59 (d, J = 8.6 Hz, 2 H, 3′′,5′′-H), 7.67 (ddd, J = 8.1, 7.2, 1.6 Hz, 1 H, 8-H), 8.29 (dd, J = 8.2, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.9 (2-NCH_3), 30.7 (C-5), 32.3 (C-4), 37.9 (C-11b), 41.2 (C-3a), 41.8 (2′-NCH_3), 113.3 (C-11a), 117.7 (C-7), 122.9 (C-4′′), 123.6 (C-10a), 125.4 (C-9), 126.4 (C-10), 129.3 (C-2′′,6′′), 129.8 (C-1′′), 132.2 (C-3′′,5′′), 133.8 (C-8), 142.4 (C-5′), 144.1 (C-4′), 155.7 (C-6a), 163.8 (C-5a), 174.6 (C-3), 175.2 (C-1), 175.9 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{20}{}^{79}BrN_4O_4$ [M + H]$^+$ 519.0668, found 519.0649; calcd. for $C_{25}H_{20}{}^{81}BrN_4O_4$ [M + H]$^+$ 521.0647, found 521.0627. MS (ESI$^+$): m/z (%) = 519 (20) [M + H]$^+$, 541 (15) [M + Na]$^+$.

General Procedure for the Synthesis of 4-(5-Aryl-2-methyl-2*H*-1,2,3-triazol-4-yl)-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (13a–d): 2,3-Dichloro-5,6-dicyanobenzoquinone (DDQ) (121 mg, 534 μmol) was added to a solution of the appropriate 4-(4-aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)-2-methyl-3a,4,5,11b-tetrahydrochromeno[3,2-*e*]isoindole-1,3,11(2*H*)-triones **12a**–**d** (178 μmol) in toluene (10 mL). The mixture was stirred at 100 °C for 1 h. After that period, the solvent was evaporated to dryness. The residue was purified by preparative TLC using CH_2Cl_2 as eluent to give desired 4-(4-aryl-2-methyl-2*H*-1,2,3-triazol-5-yl)-2-methylchromeno[3,2-*e*]-isoindole-1,3,11(2*H*)-triones **13a**–**d** in good yields (57–80%).

2-Methyl-4-(2-methyl-4-phenyl-2*H*-1,2,3-triazol-5-yl)chromeno-[3,2-*e*]isoindole-1,3,11(2*H*)-trione (13a): Yield 57.5 mg (74%), m.p. 310–312 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 3.07 (s, 3 H, 2-NCH_3), 4.35 (s, 3 H, 2′-NCH_3), 7.28–7.31 (m, 3 H, 3′′,4′′,5′′-H), 7.39–7.43 (m, 2 H, 2′′,6′′-H), 7.44–7.49 (m, 1 H, 9-H), 7.50 (d, J = 7.3 Hz, 1 H, 7-H), 7.78 (ddd, J = 8.6, 7.3, 1.6 Hz, 1 H, 8-H), 7.85 (s, 1 H, 5-H), 8.40 (dd, J = 8.0, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (75 MHz, $CDCl_3$): δ = 24.4 (2-NCH_3), 42.1 (2′-NCH_3), 117.6 (C-7), 119.7 (C-11a), 123.0 (C-10a), 125.2 (C-9), 126.3 (C-5), 127.0 (C-2′′,6′′), 127.4 (C-3a), 127.5 (C-10), 128.5 (C-4′′), 128.8 (C-3′′,5′′), 130.3 (C-1′′), 134.1 and 134.2 (C-4 and C-11b), 135.5 (C-8), 138.7 (C-5′), 146.5 (C-4′), 155.0 (C-6a), 159.7 (C-5a), 164.4 and 165.4 (C-1 and C-3), 174.3 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{25}H_{17}N_4O_4$ [M + H]$^+$ 437.1250, found 437.1229. MS (ESI$^+$): m/z (%) = 895 (100) [M + Na]$^+$, 437 (88) [M + H]$^+$, 911 (35) [2M + K]$^+$.

2-Methyl-4-[2-methyl-4-(4-methylphenyl)-2*H*-1,2,3-triazol-5-yl]-chromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (13b): Yield 64.1 mg (80%), m.p. 302–303 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.31 (s, 3 H, CH_3), 3.08 (s, 3 H, 2-NCH_3), 4.34 (s, 3 H, 2′-NCH_3), 7.09 (d, J = 8.1 Hz, 2 H, 3′′,5′′-H), 7.29 (d, J = 8.1 Hz, 2 H, 2′′,6′′-H), 7.44–7.49 (m, 1 H, 9-H), 7.50 (d, J = 7.3 Hz, 1 H, 7-H), 7.78 (ddd, J = 8.5, 7.3, 1.6 Hz, 1 H, 8-H), 7.83 (s, 1 H, 5-H), 8.39 (dd, J = 8.0, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (126 MHz, $CDCl_3$): δ = 21.3 (CH_3), 24.4 (2-NCH_3), 42.1 (2′-NCH_3), 117.6 (C-7), 119.7 (C-11a), 123.0 (C-10a), 125.2 (C-9), 126.4 (C-5), 126.9 (C-2′′,6′′), 127.4 (C-3a), 127.5 (C-10), 129.5 (C-3′′,5′′), 134.1 and 134.4 (C-11b and C-4), 135.5 (C-8), 138.4 (C-5′), 138.5 (C-1′′, C-4′′), 146.5 (C-4′), 155.0 (C-6a), 159.7 (C-5a), 164.3 and 165.5 (C-1 and C-3), 174.2 (C-11) ppm. HRMS (ESI$^+$): m/z calcd. for $C_{26}H_{19}N_4O_4$ [M + H]$^+$ 451.1406, found 451.1382. MS (ESI$^+$): m/z (%) = 451 (100) [M + H]$^+$, 489 (63) [M + K]$^+$, 923 (16) [2M + Na]$^+$, 939 (20) [2M + K]$^+$.

4-[4-(4-Methoxyphenyl)-2-methyl-2*H*-1,2,3-triazol-5-yl]-2-methyl-chromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (13c): Yield 68.9 mg (83%), m.p. 286–288 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 3.09 (s, 3 H, 2-NCH_3), 3.78 (s, 3 H, OCH_3), 4.33 (s, 3 H, 2′-NCH_3), 6.81

(d, J = 8.9 Hz, 2 H, 3′′,5′′-H), 7.34 (d, J = 8.9 Hz, 2 H, 2′′,6′′-H), 7.46 (ddd, J = 8.0, 7.1, 0.7 Hz, 1 H, 9-H), 7.50 (dd, J = 8.3, 0.7 Hz, 1 H, 7-H), 7.78 (ddd, J = 8.3, 7.1, 1.6 Hz, 1 H, 8-H), 7.84 (s, 1 H, 5-H), 8.40 (dd, J = 8.0, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (126 MHz, $CDCl_3$): δ = 24.4 (2-N*C*H$_3$), 42.1 (2′-N*C*H$_3$), 55.3 (O*C*H$_3$), 114.3 (C-3′′,5′′), 117.6 (C-7), 119.6 (C-11a), 122.8 (C-1′′), 123.0 (C-10a), 125.2 (C-9), 126.4 (C-5), 127.4 (C-3a), 127.5 (C-10), 128.3 (C-2′′,6′′), 134.1 and 134.4 (C-4 and C-11b), 135.5 (C-8), 138.1 (C-5′), 146.3 (C-4′), 155.0 (C-6a), 159.76 (C-4′′), 159.75 (C-5a), 164.3 and 165.4 (C-1 and C-3), 174.3 (C-11) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{26}H_{19}N_4O_5$ [M + H]$^+$ 467.1355, found 467.1332. MS (ESI$^+$): *m*/*z* (%) = 467 (100) [M + H]$^+$, 489 (27) [M + Na]$^+$, 505 (20) [M + K]$^+$.

4-[4-(4-Bromophenyl)-2-methyl-2*H*-1,2,3-triazol-5-yl]-2-methylchromeno[3,2-*e*]isoindole-1,3,11(2*H*)-trione (13d): Yield 52.3 mg (57%), m.p. 312–313 °C. ^{1}H NMR (500 MHz, $CDCl_3$): δ = 3.09 (s, 3 H, 2-NC*H*$_3$), 4.34 (s, 3 H, 2′-NC*H*$_3$), 7.29 (d, J = 8.6 Hz, 2 H, 2′′,6′′-H), 7.42 (d, J = 8.6 Hz, 2 H, 3′′,5′′-H), 7.47 (ddd, J = 8.0, 7.1, 0.7 Hz, 1 H, 9-H), 7.51 (dd, J = 8.3, 0.7 Hz, 1 H, 7-H), 7.79 (ddd, J = 8.3, 7.1, 1.6 Hz, 1 H, 8-H), 7.84 (s, 1 H, 5-H), 8.40 (dd, J = 8.0, 1.6 Hz, 1 H, 10-H) ppm. ^{13}C NMR (126 MHz, $CDCl_3$): δ = 24.5 (2-N*C*H$_3$), 42.2 (2′-N*C*H$_3$), 117.6 (C-7), 119.8 (C-11a), 122.7 (C-4′′), 123.0 (C-10a), 125.3 (C-9), 126.3 (C-5), 127.2 (C-3a), 127.5 (C-10), 128.4 (C-2′′,6′′), 129.4 (C-1′′), 132.0 (C-3′′,5′′), 133.8 and 134.1 (C-4 and C-11b), 135.6 (C-8), 138.7 (C-5′), 145.5 (C-4′), 155.0 (C-6a), 159.8 (C-5a), 164.1 and 165.4 (C-1 and C-3), 174.1 (C-11) ppm. HRMS (ESI$^+$): *m*/*z* calcd. for $C_{25}H_{16}{}^{79}BrN_4O_4$ [M + H]$^+$ 515.0355, found 515.0330; calcd. for $C_{25}H_{16}{}^{81}BrN_4O_4$ [M + H]$^+$ 517.0334, found 517.0484. MS (ESI$^+$): *m*/*z* (%) = 515 (15) [M + H]$^+$, 537 (24) [M + Na]$^+$, 555 (21) [M + K]$^+$.

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