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New efficient synthesis of 2,3,4-trisubstituted 3,4-dihydroquinazolines by a Ugi 4CC/Staudinger/aza-Wittig sequence

Ying Zhong, Long Wang, Ming-Wu Ding *

Key Laboratory of Pesticide and Chemical Biology of Ministry of Education, Central China Normal University, Wuhan 430079, PR China

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ABSTRACT

A series of 2,3,4-trisubstituted 3,4-dihydroquinazoline **3** was prepared by intramolecular aza-Wittig reaction of amide carbonyl groups with methyldiphenyl iminophosphorane, which was obtained from a Ugi 4CC/Staudinger sequence. Further intramolecular Heck reaction of **3d′**–**g′** in the presence of catalytic amount of $Pd(OAc)_2$ gave 6,12-dihydroindolo[2,1-*b*]quinazolines **4a**–**d** in good yields.

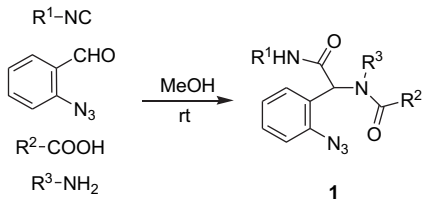
1. Introduction

3,4-Dihydroquinazolines have widespread biological activities and a wide variety of derivatives of this ring system have been used as BACE-1 inhibitor, α-MSH-induced melanin inhibitor, anti-inflammatory, anti-allergic, anti-malarial agent, and especially as cancer treatment agent.[1] For example, some 3,4-dihydroquinazolines were reported to have potent T-type channel blocking activity and anti-cancer activity, their growth inhibition of human cancer cells is similar to that of Doxorubicin and has little cytotoxicity.[2] There are many known methods for the synthesis of 3,4-dihydroquinazolines.[3] However, multisubstituted 4-aminocarbonyl-3,4-dihydroquinazolines have not yet been prepared previously probably due to the fact that they are not easily accessible by routine synthetic method. Multisubstituted 4-aminocarbonyl-3,4-dihydro quinazoline is a very attractive target for biochemical studies and drug discovery, due to its large number of possible substitution patterns and interesting combination of potential hydrogen bond donors and acceptors on the amide moiety.

Ugi reaction is a powerful, atom-economical I-MCRs starting from isonitrile, amine, aldehyde (or ketone), and carboxylic acid components that generates a significantly more complex α-acylamino amide adduct.[4] The sequences of classical Ugi isocyanide multicomponent reactions, followed by post-condensation transformations, constitute extremely powerful synthetic tools for the preparation of structurally diverse heterocyclic compounds.[5] On the other hand, aza-Wittig reactions have received increased attention in view of their utility in the synthesis of nitrogen-containing heterocycles.[6] Thus, it is envisioned that combining the efficiency of the Ugi condensation with a post-condensation aza-Wittig reaction would facilitate access to a series of biologically useful heterocycles. Indeed, the sequence of Ugi and Passerini reaction starting from some azide precursors, followed by post-condensation of Staudinger and aza-Wittig reaction, has been recently utilized in synthesis of various heterocycles, such as 1,4-benzodiazepine-5-ones,[7] dibenzo[*b*,*f*]-1,5-diazocine-6(5*H*)-ones,[8] 5-oxo-1,4-diazepines,[9] 4*H*-3,1-benzoxazine,[10] 3,4-dihydroquinazolines,[11] indolo[1,2-*c*]quinazolines,[12] and oxazolines.[13] Continuing our interest in the synthesis of *N*-heterocycles via the aza-Wittig reaction,[14] we report herein a new efficient synthesis of trisubstituted 3,4-dihydroquinazolines by a Ugi 4CC/Staudinger/aza-Wittig sequence.

2. Results and discussion

A mixture of 2-azidobenzaldehyde (1 equiv), amine (1 equiv), isocyanide (1 equiv), and acid (1 equiv) was stirred in methanol at room temperature for 12–36 h until solid precipitated (Scheme 1). The Ugi reaction was carried out smoothly and the azide products **1** were obtained in satisfactory yields after recrystallization (Scheme 1 and Table 1).[11]



Scheme 1. Preparation of compounds **1**.

Initially, the reactions of azides **1** with triphenylphosphine were examined in toluene at room temperature for 2 h, followed by heating at reflux for 6–24 h. Nitrogen evolution via the Staudinger reaction ceased during the first 2 h to give triphenyl

* Corresponding author. Tel.: +86 27 63158845; fax: +86 27 67862041; e-mail address: ding5229@yahoo.com.cn (M.-W. Ding).

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Table 1
Preparation of azides **1** via Ugi 4CC reaction

	R^1	R^2	R^3	Time (h)	Yield[a] (%)
1a	*t*-Bu	H	4-ClC_6H_4	24	74
1b	*n*-Bu	H	4-MeC_6H_4	24	85
1c	*n*-Bu	CF_3	4-ClC_6H_4	12	67
1d	*t*-Bu	CF_3	Ph	12	66
1e	*t*-Bu	CH_3	4-ClC_6H_4	12	92
1f	*t*-Bu	CH_3	4-MeC_6H_4	12	81
1g	*t*-Bu	CH_3CH_2	4-ClC_6H_4	12	78
1h	*t*-Bu	4-$NO_2C_6H_4$	4-MeC_6H_4	12	85
1i	*t*-Bu	4-$NO_2C_6H_4$	4-ClC_6H_4	36	73
1j	*n*-Bu	Ph	4-ClC_6H_4	36	90
1k	*t*-Bu	4-MeC_6H_4	4-ClC_6H_4	36	87
1l	*t*-Bu	2-FC_6H_4	4-ClC_6H_4	24	83
1m	*t*-Bu	2-MeC_6H_4	4-ClC_6H_4	24	69
1q	*t*-Bu	Ph	*c*-C_6H_{11}	36	70
1r	*t*-Bu	4-ClC_6H_4	*t*-Bu	24	81
1s	*t*-Bu	4-MeC_6H_4	*t*-Bu	36	77
1t	*t*-Bu	Ph	*t*-Bu	36	75
1u	*t*-Bu	Ph	4-MeC_6H_4	24	91
1v	*n*-Bu	Ph	4-MeC_6H_4	24	83
1w	*n*-Bu	4-MeC_6H_4	4-ClC_6H_4	24	88
1x	*n*-Bu	4-MeC_6H_4	4-MeC_6H_4	36	82
1z	*n*-Bu	4-ClC_6H_4	4-MeC_6H_4	36	86
1a′	*t*-Bu	Ph	4-$EtOC_6H_4$	36	82
1b′	*t*-Bu	Ph	2-MeC_6H_4	36	74
1c′	*t*-Bu	Ph	3-ClC_6H_4	24	87
1d′	*t*-Bu	PhCH=CH–	2-BrC_6H_4	36	74
1e′	*n*-Bu	PhCH=CH–	2-BrC_6H_4	36	78
1f′	*t*-Bu	4-ClC_6H_4CH=CH–	2-BrC_6H_4	36	72
1g′	*n*-Bu	4-ClC_6H_4CH=CH–	2-BrC_6H_4	36	70

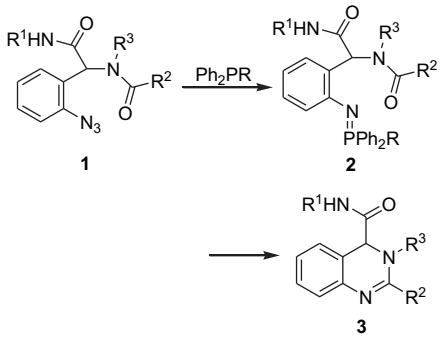
[a] Isolated yields of **1** based on 2-azidobenzaldehyde.

iminophosphorane **2**, but further heating of the triphenyl iminophosphorane **2** gave disappointed results (Table 2). As indicated in Table 2, no or low yields of the 3,4-dihydroquinazolines **3** were obtained when R^2 or R^3 is alkyl or aromatic group. Good yields were reached only in case that R^2 is a strong electro-withdrawing CF_3 group or a small H (**3a**–**d**, Table 2). The above cyclization involves intramolecular aza-Wittig reaction between amide carbonyls and triphenyl iminophosphorane. The low yield of the cyclization reaction is due to the low electrophilicity of the amido group. Although some examples of intramolecular aza-Wittig reactions involving triphenyl iminophosphorane and 'activated' amide carbonyl groups (with electron-withdrawing substituent on the nitrogen atom, i.e., imide or sulfonyl amide groups) gave good yield of various heterocycles,[15] simple amide carbonyls are typically inert toward intramolecular aza-Wittig reactions.[16] This problem has been recognized for many years and triphenyl iminophosphorane was replaced by more reactive trialkyl iminophosphorane to provide better results in some cases (Scheme 2).[15d,c,17]

Table 2
Preparation of 3,4-dihydroquinazoline **3** via intramolecular aza-Wittig reaction of triphenyl iminophosphorane **2** (R=Ph)

	R^1	R^2	R^3	Time (h)	Yield[a] (%)
3a	*t*-Bu	H	4-ClC_6H_4	12	87
3b	*n*-Bu	H	4-MeC_6H_4	12	72
3c	*n*-Bu	CF_3	4-ClC_6H_4	6	83
3d	*t*-Bu	CF_3	Ph	6	80
3e	*t*-Bu	CH_3	4-ClC_6H_4	12	5
3f	*t*-Bu	CH_3	4-MeC_6H_4	12	0
3g	*t*-Bu	CH_3CH_2	4-ClC_6H_4	12	0
3h	*t*-Bu	4-$NO_2C_6H_4$	4-MeC_6H_4	12	37
3i	*t*-Bu	4-$NO_2C_6H_4$	4-ClC_6H_4	12	52
3j	*n*-Bu	Ph	4-ClC_6H_4	24	14
3k	*t*-Bu	4-MeC_6H_4	4-ClC_6H_4	24	10
3l	*t*-Bu	2-FC_6H_4	4-ClC_6H_4	24	0
3m	*t*-Bu	2-MeC_6H_4	4-ClC_6H_4	24	0
3n	*t*-Bu	Ph	Me	24	0
3o	*t*-Bu	Ph	Et	24	0
3p	*t*-Bu	Ph	*i*-Pr	24	0
3t	*t*-Bu	Ph	*t*-Bu	24	0

[a] Isolated yields based on azides **1**.



Scheme 2. Preparation of compounds **3**.

We have now observed that a simple change in phosphorus substituents from triphenyl to methyldiphenyl iminophosphorane dramatically increases the reactivity of intramolecular aza-Wittig reactions involving such less reactive amide carbonyl groups. The reactions of azides **1** with methyldiphenyl phosphine (prepared conveniently from triphenylphosphine[18] and is more easily handled or stored than trialkyl phosphine due to its sufficient stability to air) gave diphenylmethyl iminophosphorane **2** (R=Me) at room temperature. Further heating of **2** in toluene produced 3,4-dihydroquinazolines **3** in satisfactory yields (Scheme 2, Table 3). Compared with the condition needed for cyclization in Tables 2 and 3, an obvious improvement was observed when methyldiphenyl iminophosphorane **2** was used. In the preparation of compounds **3a**–**d** from diphenylmethyl iminophosphorane, shorter reaction

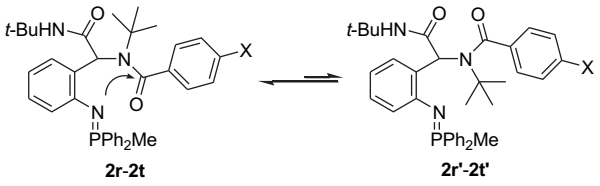
Table 3
Preparation of 3,4-dihydroquinazoline **3** via intramolecular aza-Wittig reaction of diphenylmethyl iminophosphorane **2** (R=Me)

	R^1	R^2	R^3	Time (h)	Yield[a] (%)
3a	*t*-Bu	H	4-ClC_6H_4	1	92
3b	*n*-Bu	H	4-MeC_6H_4	2	82
3c	*n*-Bu	CF_3	4-ClC_6H_4	1	85
3d	*t*-Bu	CF_3	Ph	1	88
3e	*t*-Bu	CH_3	4-ClC_6H_4	18	80
3f	*t*-Bu	CH_3	4-MeC_6H_4	19	60
3g	*t*-Bu	CH_3CH_2	4-ClC_6H_4	12	82
3h	*t*-Bu	4-$NO_2C_6H_4$	4-MeC_6H_4	2	93
3i	*t*-Bu	4-$NO_2C_6H_4$	4-ClC_6H_4	2	95
3j	*n*-Bu	Ph	4-ClC_6H_4	2	91
3k	*t*-Bu	4-MeC_6H_4	4-ClC_6H_4	2	88
3l	*t*-Bu	2-FC_6H_4	4-ClC_6H_4	18	91
3m	*t*-Bu	2-MeC_6H_4	4-ClC_6H_4	36	35
3n	*t*-Bu	Ph	Me	36	40
3o	*t*-Bu	Ph	Et	36	50
3p	*t*-Bu	Ph	*i*-Pr	36	62
3q	*t*-Bu	Ph	*c*-C_6H_{11}	36	67
3r	*t*-Bu	4-ClC_6H_4	*t*-Bu	6	85
3s	*t*-Bu	4-MeC_6H_4	*t*-Bu	6	88
3t	*t*-Bu	Ph	*t*-Bu	6	93
3u	*t*-Bu	Ph	4-MeC_6H_4	2	88
3v	*n*-Bu	Ph	4-MeC_6H_4	3	90
3w	*n*-Bu	4-MeC_6H_4	4-ClC_6H_4	2	92
3x	*n*-Bu	4-MeC_6H_4	4-MeC_6H_4	3	89
3y	*n*-Bu	4-ClC_6H_4	4-ClC_6H_4	2	91
3z	*n*-Bu	4-ClC_6H_4	4-MeC_6H_4	2	90
3a′	*t*-Bu	Ph	4-$EtOC_6H_4$	3	92
3b′	*t*-Bu	Ph	2-MeC_6H_4	5	88
3c′	*t*-Bu	Ph	3-ClC_6H_4	4	84
3d′	*t*-Bu	PhCH=CH–	2-BrC_6H_4	12	82
3e′	*n*-Bu	PhCH=CH–	2-BrC_6H_4	12	73
3f′	*t*-Bu	4-ClC_6H_4CH=CH–	2-BrC_6H_4	12	85
3g′	*n*-Bu	4-ClC_6H_4CH=CH–	2-BrC_6H_4	12	80

[a] Isolated yields based on azides **1**.

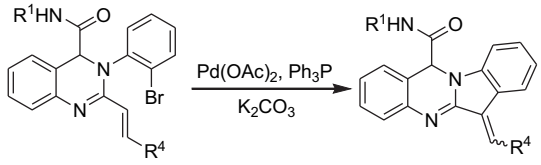
times were required in the comparison of that from triphenyl iminophosphorane (compare **3a**–**d** in Table 3 with **3a**–**d** in Table 2). In case that no or low yield of the 3,4-dihydroquinazolines **3** were obtained (**3e**–**l**) in Table 2, good yield of the cyclized products (**3e**–**l**, **3u**–**g′**) was obtained in Table 3 when the reaction mixture was heated for 2–24 h. The steric hindrance of R^2 group has remarkable influence on the reaction yield: when R^2 is a steric 2-methylphenyl (**3m**), low yield of the products (35%) was obtained even after heating for 36 h in Table 3. However, it is not the case with R^3 group: as R^3 is the steric 2-bromophenyl (**3d′**–**g′**), good yields of the products were obtained after heating for 12 h.

It is interesting that in Table 3 there is even an inverse influence of the steric hindrance of R^3 group on the reaction yields. In preparation of compounds **3n**–**t**, the yields of the product increased with the increasing steric hindrance of R^3 group. When R^3 is changed from Me, Et, *i*-Pr to cyclohexyl, the yield is increased slightly from 40 to 67% after heating for 36 h. However, as R^3 is the bulky *t*-Bu group, the yields are promoted remarkably to 85–93% after heating for only 6 h (**3r**–**t**). This is probably due to the conformation of the iminophosphorane **2r**–**t**. The bulky *t*-Bu group in conformation of **2r**–**t** repels the benzoyl group to the iminophosphorane moiety, which is entropically favorable for the cyclization (Scheme 3).



Scheme 3.

Some of the products **3** can be further transferred into fused quinazolines. For eample, intramolecular Heck reaction of 3,4-dihydroquinazolines **3d′**–**g′** in the presence of catalytic amount of $Pd(OAc)_2$ and Ph_3P produced 6,12-dihydroindolo[2,1-*b*]quinazolines **4a**–**d** in 58–77% yields (Scheme 4, Table 4). It provides a rapid synthesis of 6,12-dihydroindolo[2,1-*b*]quinazoline via a sequential Ugi/Staudinger/aza-Wittig/Heck reaction, starting from easily accessible 2-bromobenzenamine, cinnamic acids, 2-azidobenzaldehyde and isocyanide.



Scheme 4. Preparation of compounds **4**.

Table 4
Preparation of 6,12-dihydroindolo[2,1-*b*]quinazolines **4a**–**d** via intramolecular Heck reaction

	R^1	R^4	Time (h)	Yield[a] (%)
4a	*t*-Bu	Ph	6	58
4b	*n*-Bu	Ph	6	73
4c	*t*-Bu	4-ClC_6H_4	5	62
4d	*n*-Bu	4-ClC_6H_4	5	77

[a] Isolated yields.

3. Conclusion

We report a new multicomponent synthesis of 3,4-dihydroquinazoline using Ugi reaction followed by Staudinger and intramolecular aza-Wittig reaction of methyldiphenyl iminophosphorane. The used amines, isocyanides, and acids can be varied broadly, producing products with three potential points of diversity. Some of the 3,4-dihydroquinazoline can be further transferred into fused quinazolines. Due to the easy availability of the synthetic approach and the neutral ring closure condition, this new synthetic approach discussed here has the potential in synthesis of various 2,3,4-trisubstituted 3,4-dihydroquinazolines or fused quinazolines, which are of considerable interest as potential biological active compounds or pharmaceuticals.

4. Experimental

4.1. General

Melting points were uncorrected. MS were measured on a Finnigan Trace MS spectrometer. NMR were recorded in $CDCl_3$ or DMSO-d_6 on a Varian Mercury 400 or 600 spectrometer and resonances relative to TMS. Elementary analyses were taken on a Vario EL III elementary analysis instrument.

4.2. Synthesis of azides 1 via Ugi reaction

*4.2.1. 2-(2-Azidophenyl)-N-tert-butyl-2-(N-(4-chlorophenyl) formamido)acetamide (**1a**).* 2-Azidobenzaldehyde (0.29 g, 2 mmol), formic acid (0.09 g, 2 mmol), and *tert*-butylisocyanide (0.17 g, 2 mmol) were added sequentially to a solution of 4-chlorophenyl amine (0.25 g, 2 mmol) in methanol (15 mL) at room temperature. The reaction mixture was stirred at ambient temperature for 24 h until solid precipitated completely. After the solvent was evaporated, the crude reaction mixture was purified by recrystallization from methylene dichloride/petroleum ether to give 0.57 g (74%) of azide **1a** as white solid. Mp: 183–185 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.35 (s, 1H, CHO), 7.30–6.96 (m, 8H, Ar–H), 6.08 (s, 1H, CH), 5.59 (s, 1H, NH), 1.38 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 167.9, 162.6, 138.9, 136.9, 133.5, 131.0, 130.2, 129.7, 128.7, 124.7, 118.0, 58.0, 51.8, 28.5. Anal. Calcd for $C_{19}H_{20}ClN_5O_2$: C, 59.14; H, 5.22; N, 18.15. Found: C, 59.57; H, 5.43; N, 18.12.

*4.2.2. 2-(2-Azidophenyl)-N-butyl-2-(N-p-tolylformamido) acetamide (**1b**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol) and 4-methylphenyl amine (0.22 g, 2 mmol), compound **1b** (0.62 g, 85%) was also isolated as white solid. Mp: 174–176 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.35 (s, 1H, COH), 7.29–6.95 (m, 8H, Ar–H), 6.17 (s, 1H, CH), 5.81 (s, 1H, NH), 3.34–3.29 (m, 2H, CH_2), 2.25 (s, 3H, CH_3), 1.50–1.45 (m, 2H, CH_2), 1.34–1.26 (m, 2H, CH_2), 0.90 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.6, 163.1, 138.9, 137.7, 135.9, 131.4, 130.0, 129.3, 128.0, 124.5, 117.9, 57.9, 39.6, 31.4, 20.9, 20.0, 13.7. Anal. Calcd for $C_{20}H_{23}N_5O_2$: C, 65.73; H, 6.34; N, 19.16. Found: C, 65.97; H, 6.18; N, 19.33.

*4.2.3. N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-N-(4-chlorophenyl)-2,2,2-trifluoroacetamide (**1c**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol) and 2,2,2-trifluoroacetic acid (0.23 g, 2 mmol), compound **1c** (0.61 g, 67%) was also isolated as white solid. Mp: 136–137 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.80–6.59 (m, 8H, Ar–H), 6.23 (s, 1H, CH), 5.61 (s, 1H, NH), 3.33–3.28 (m, 2H, NCH_2), 1.50–1.43 (m, 2H, CH_2), 1.34–1.25 (m, 2H, CH_2), 0.86 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 167.9, 157.3, 157.0, 139.4, 135.0, 134.2, 132.7, 131.6, 131.0, 130.8, 128.6, 128.0, 124.8, 123.3,

117.9, 60.4, 39.7, 31.2, 19.9, 13.5 Anal. Calcd for $C_{20}H_{19}ClF_3N_5O_2$: C, 52.93; H, 4.22; N, 15.43. Found: C, 53.22; H, 4.37; N, 15.18.

*4.2.4. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-2,2,2-trifluoro-N-phenylacetamide (**1d**).* Operation as above with phenylamine (0.19 g, 2 mmol) and 2,2,2-trifluoroacetic acid (0.23 g, 2 mmol), compound **1d** (0.55 g, 66%) was also isolated as white solid. Mp: 186–187 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.80–6.69 (m, 9H, Ar–H), 6.12 (s, 1H, CH), 5.40 (s, 1H, N–H), 1.36 (s, 9H, 3×CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 167.1, 157.4, 157.2, 139.4, 135.8, 131.6, 131.2, 130.5, 129.7, 128.9, 128.3, 127.8, 124.6, 123.9, 117.8, 60.9, 52.0, 28.6. Anal. Calcd for $C_{20}H_{20}F_3N_5O_2$: C, 57.28; H, 4.81; N, 16.70. Found: C, 57.56; H, 4.93; N, 16.55.

*4.2.5. 2-(2-Azidophenyl)-N-(tert-butyl)-2-(N-(4-chlorophenyl)acetamido)acetamide (**1e**).* Operation as above with acetic acid (0.12 g, 2 mmol), compound **1e** (0.74 g, 92%) was also isolated as white solid. Mp: 209–211 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.27–6.85 (m, 8H, Ar–H), 6.17 (s, 1H, CH), 5.47 (s, 1H, NH), 1.87 (s, 3H, CH_3), 1.35 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.6, 168.8, 139.0, 138.7, 133.6, 131.3, 129.9, 128.7, 125.6, 124.6, 117.8, 58.8, 51.6, 28.5, 23.1 Anal. Calcd for $C_{20}H_{22}ClN_5O_2$: C, 60.07; H, 5.55; N, 17.51. Found: C, 59.83; H, 5.55; N, 17.68.

*4.2.6. 2-(2-Azidophenyl)-N-(tert-butyl)-2-(N-(p-tolyl) acetamido) acetamide (**1f**).* Operation as above with 4-methylphenyl amine (0.22 g, 2 mmol) and acetic acid (0.12 g, 2 mmol), compound **1f** (0.61 g, 81%) was also isolated as white solid. Mp: 206–208 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.27–6.82 (m, 8H, Ar–H), 6.18 (s, 1H, CH), 5.56 (s, 1H, NH), 2.24 (s, 3H, CH_3), 1.86 (s, 3H, CH_3), 1.35 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 171.1, 168.8, 139.0, 137.6, 137.5, 131.5, 129.6, 126.0, 124.3, 117.6, 58.9, 51.4, 28.5, 23.1, 20.9. Anal. Calcd for $C_{21}H_{25}N_5O_2$: C, 66.47; H, 6.64; N, 18.46. Found: C, 66.18; H, 6.45; N, 18.55.

*4.2.7. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(4-chlorophenyl)propionamide (**1g**).* Operation as above with propionic acid (0.15 g, 2 mmol), compound **1g** (0.65 g, 78%) was also isolated as white solid. Mp: 176–178 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.30–6.84 (m, 8H, Ar–H), 6.22 (s, 1H, CH), 5.72 (s, 1H, N–H), 2.05–2.03 (m, 2H, CH_2), 1.35 (s, 9H, 3CH_3), 1.04–1.03 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 173.9, 169.0, 139.0, 138.3, 133.6, 131.4, 129.8, 128.7, 125.7, 124.6, 117.8, 58.9, 51.5, 28.4, 28.3, 28.2. Anal. Calcd for $C_{21}H_{24}ClN_5O_2$: C, 60.94; H, 5.84; N, 16.92. Found: C, 60.88; H, 5.73; N, 16.62.

*4.2.8. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-4-nitro-N-p-tolylbenzamide (**1h**).* Operation as above with 4-methylphenyl amine (0.22 g, 2 mmol) and 4-nitrobenzoic acid (0.33 g, 2 mmol), compound **1h** (0.83 g, 85%) was also isolated as white solid. Mp: 179–181 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.99–6.75 (m, 12H, Ar–H), 6.32 (s, 1H, CH), 5.56 (s, 1H, NH), 2.12 (s, 3H, CH_3), 1.39 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.8, 168.2, 147.5, 142.6, 139.2, 137.5, 136.6, 131.5, 130.0, 129.8, 129.2, 128.9, 125.3, 124.6, 122.8, 117.9, 60.5, 51.8, 30.8, 28.5, 20.8. Anal. Calcd for $C_{26}H_{26}N_6O_4$: C, 64.19; H, 5.39; N, 17.27. Found: C, 64.52; H, 5.54; N, 17.48.

*4.2.9. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(4-chlorophenyl)-4-nitrobenzamide (**1i**).* Operation as above with 4-nitrobenzoic acid (0.33 g, 2 mmol), compound **1i** (0.74 g, 73%) was also isolated as white solid. Mp: 235–237 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.03–6.94 (m, 12H, Ar–H), 6.35 (s, 1H, CH), 5.54 (s, 1H, N–H), 1.40 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.6, 168.2, 147.8, 142.1, 139.2, 137.8, 133.5, 131.5, 131.4, 130.4, 129.2, 128.5, 124.8, 123.0, 118.0, 60.3, 52.0, 30.9, 28.6. Anal. Calcd for $C_{25}H_{23}ClN_6O_4$: C, 59.23; H, 4.57; N, 16.58. Found: C, 59.00; H, 4.31; N, 16.67.

*4.2.10. N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-N-(4-chlorophenyl)benzamide (**1j**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1j** (0.83 g, 90%) was also isolated as white solid. Mp: 149–152 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.30–6.93 (m, 13H, Ar–H), 6.45 (s, 1H, CH), 5.94 (s, 1H, NH), 3.38–3.30 (m, 2H, NCH_2), 1.52–1.47 (m, 2H, CH_2), 1.33–1.30 (m, 2H, CH_2), 0.90 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.8, 169.4, 139.1, 135.7, 132.7, 131.5, 130.1, 129.5, 128.4, 128.2, 127.7, 125.4, 124.7, 117.9, 60.1, 39.6, 31.3, 20.0, 13.6. Anal. Calcd for $C_{25}H_{24}ClN_5O_2$: C, 65.00; H, 5.24; N, 15.16. Found: C, 65.32; H, 5.11; N, 15.02.

*4.2.11. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(4-chlorophenyl)-4-methylbenzamide (**1k**).* Operation as above with 4-methylbenzoic acid (0.27 g, 2 mmol), compound **1k** (0.83 g, 87%) was also isolated as white solid. Mp: 180–182 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.27–6.92 (m, 12H, Ar–H), 6.31 (s, 1H, CH), 5.68 (s, 1H, NH), 2.43 (s, 3H, CH_3), 1.38 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.8, 168.6, 139.8, 139.3, 139.1, 132.8, 132.5, 131.4, 130.0, 128.6, 128.4, 128.1, 125.7, 124.7, 117.9, 60.6, 51.7, 28.6, 21.3. Anal. Calcd for $C_{26}H_{26}ClN_5O_2$: C, 65.61; H, 5.51; N, 14.71. Found: C, 65.48; H, 5.39; N, 14.56.

*4.2.12. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(4-chlorophenyl)-2-fluorobenzamide (**1l**).* Operation as above with 2-fluorobenzoic acid (0.28 g, 2 mmol), compound **1l** (0.80 g, 83%) was also isolated as white solid. Mp: >300 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.27–6.83 (m, 12H, Ar–H), 6.37 (s, 1H, CH), 5.78 (s, 1H, NH), 1.40 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.1, 166.9, 158.4, 156.8, 139.2, 137.6, 133.2, 131.4, 131.2, 131.0, 130.9, 130.1, 129.0, 128.0, 125.2, 125.0, 124.8, 124.6, 123.8, 117.9, 115.4, 115.3, 59.9, 51.8, 28.5. Anal. Calcd for $C_{25}H_{23}ClFN_5O_2$: C, 62.56; H, 4.83; N, 14.59. Found: C, 62.83; H, 4.96; N, 14.59.

*4.2.13. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(4-chlorophenyl)-2-methylbenzamide (**1m**).* Operation as above with 2-methylbenzoic acid (0.27 g, 2 mmol), compound **1m** (0.66 g, 69%) was also isolated as white solid. Mp: 176–179 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.27–6.85 (m, 12H, Ar–H), 6.34 (s, 1H, CH), 5.72 (s, 1H, NH), 2.40 (s, 3H, CH_3), 1.40 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 171.4, 168.6, 139.0, 137.9, 136.0, 134.5, 132.8, 131.3, 131.1, 129.9, 128.5, 127.9, 127.1, 125.4, 124.8, 124.7, 117.9, 59.6, 51.7, 28.5, 19.3. Anal. Calcd for $C_{26}H_{26}ClN_5O_2$: C, 65.61; H, 5.51; N, 14.71. Found: C, 65.94; H, 5.68; N, 14.45.

*4.2.14. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-cyclohexylbenzamide (**1q**).* Operation as above with benzoic acid (0.24 g, 2 mmol) and cyclohexylamine (0.20 g, 2 mmol), compound **1q** (0.61 g, 70%) was also isolated as white solid. Mp: 162–164 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.69–7.18 (m, 9H, Ar–H), 5.47 (s, 1H, N–H), 5.08 (s, 1H, CH), 3.62–3.37 (m, 1H, NCH), 2.05–1.04 (m, 19H, 5CH_2, and 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 172.2, 168.0, 137.6, 136.9, 129.7, 129.3, 128.3, 125.9, 125.2, 117.9, 59.9, 58.1, 51.2, 31.8, 31.4, 28.3, 25.6, 25.3, 24.9. Anal. Calcd for $C_{25}H_{31}N_5O_2$: C, 69.26; H, 7.21; N, 16.15. Found: C, 69.58; H, 7.48; N, 16.42.

*4.2.15. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-tert-butyl-4-chlorobenzamide (**1r**).* Operation as above with *tert*-butylamine (0.15 g, 2 mmol) and 4-chlorobenzoic acid (0.31 g, 2 mmol), compound **1r** (0.72 g, 81%) was also isolated as white solid. Mp: 208–210 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.66–7.15 (m, 8H, Ar–H), 5.49 (s, 1H, N–H), 5.19 (s, 1H, CH), 1.28 (s, 9H, 3CH_3), 1.21 (s, 9H, 3CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 173.2, 168.4, 139.1, 137.8, 134.9, 130.9, 129.5, 129.1, 128.3, 128.0, 125.3, 117.9, 62.5, 58.6, 51.2, 30.6, 28.4. Anal. Calcd for $C_{23}H_{28}ClN_5O_2$: C, 62.51; H, 6.39; N, 15.85. Found: C, 62.64; H, 6.62; N, 15.61.

*4.2.16. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-tert-butyl-4-methylbenzamide (**1s**).* Operation as above with *tert*-

butylamine (0.15 g, 2 mmol) and 4-methylbenzoic acid (0.27 g, 2 mmol), compound **1s** (0.65 g, 77%) was also isolated as white solid. Mp: 187–189 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.71–7.15 (m, 8H, Ar–H), 5.81 (s, 1H, N–H), 5.20 (s, 1H, CH), 2.36 (s, 3H, CH_3), 1.30 (s, 9H, $3CH_3$), 1.22 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 174.5, 168.9, 139.0, 137.7, 131.1, 129.2, 128.5, 126.5, 125.1, 117.8, 62.7, 58.4, 51.1, 30.6, 28.4, 21.3. Anal. Calcd for $C_{24}H_{31}N_5O_2$: C, 68.38; H, 7.41; N, 16.61. Found: C, 68.16; H, 7.22; N, 16.53.

*4.2.17. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-tert-butylbenzamide (**1t**).* Operation as above with *tert*-butylamine (0.15 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1t** (0.61 g, 75%) was also isolated as white solid. Mp: 173–175 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.71–7.16 (m, 9H, Ar–H), 5.65 (s, 1H, NH), 5.22 (s, 1H, CH), 1.29 (s, 9H, $3CH_3$), 1.21 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 174.3, 168.7, 140.7, 137.9, 131.1, 129.4, 129.0, 128.0, 126.5, 125.2, 117.9, 62.7, 58.6, 51.2, 30.7, 28.4. Anal. Calcd for $C_{23}H_{29}N_5O_2$: C, 67.79; H, 7.17; N, 17.19. Found: C, 68.05; H, 7.23; N, 17.37.

*4.2.18. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-p-tolylbenzamide (**1u**).* Operation as above with 4-methylphenyl amine (0.22 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1u** (0.80 g, 91%) was also isolated as white solid. Mp: 173–174 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.33–6.75 (m, 13H, Ar–H), 6.30 (s, 1H, CH), 5.78 (s, 1H, NH), 2.12 (s, 3H, CH_3), 1.39 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 171.0, 168.6, 139.1, 137.9, 136.7, 136.1, 131.5, 129.6, 129.2, 128.7, 128.5, 127.5, 126.0, 124.5, 117.8, 61.0, 51.6, 28.6, 20.9. Anal. Calcd for $C_{26}H_{27}N_5O_2$: C, 70.73; H, 6.16; N, 15.86. Found: C, 70.69; H, 6.31; N, 15.64.

*4.2.19. N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-N-p-tolylbenzamide (**1v**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol), 4-methylphenyl amine (0.22 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1v** (0.73 g, 83%) was also isolated as white solid. Mp: 142–144 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.33–6.77 (m, 13H, Ar–H), 6.38 (s, 1H, CH), 5.98 (s, 1H, NH), 3.36–3.32 (m, 2H, CH_2), 2.13 (s, 3H, CH_3), 1.51–1.47 (m, 2H, CH_2), 1.34–1.30 (m, 2H, CH_2), 0.90 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 171.0, 169.4, 139.0, 138.0, 136.7, 136.0, 131.6, 129.6, 129.2, 128.7, 128.4, 127.4, 125.9, 124.5, 117.8, 60.7, 39.5, 31.3, 20.8, 19.9, 13.6. Anal. Calcd for $C_{26}H_{27}N_5O_2$: C, 70.73; H, 6.16; N, 15.86. Found: C, 70.98; H, 6.04; N, 15.58.

*4.2.20. N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-N-(4-chlorophenyl)-4-methylbenzamide (**1w**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol) and 4-methylbenzoic acid (0.27 g, 2 mmol), compound **1w** (0.84 g, 88%) was also isolated as white solid. Mp: 183–186 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.30–6.94 (m, 12H, Ar–H), 6.42 (s, 1H, CH), 5.93 (s, 1H, N–H), 3.37–3.30 (m, 2H, CH_2), 2.25 (s, 3H, CH_3), 1.51–1.48 (m, 2H, CH_2), 1.34–1.29 (m, 2H, CH_2), 0.79 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.9, 169.4, 139.8, 139.4, 139.1, 132.6, 131.6, 131.4, 130.0, 128.6, 128.3, 128.2, 125.6, 124.7, 117.9, 60.3, 39.6, 31.4, 21.3, 20.0, 13.7. Anal. Calcd for $C_{26}H_{26}ClN_5O_2$: C, 65.61; H, 5.51; N, 14.71. Found: C, 65.97; H, 5.65; N, 14.44.

*4.2.21. N-(1-(2-azidophenyl)-2-(butylamino)-2-oxoethyl)-4-methyl-N-p-tolylbenzamide (**1x**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol), 4-methylphenyl amine (0.22 g, 2 mmol) and 4-methylbenzoic acid (0.27 g, 2 mmol), compound **1x** (0.75 g, 82%) was also isolated as white solid. Mp: 152–154 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.32–6.79 (m, 12H, Ar–H), 6.36 (s, 1H, CH), 6.06 (s, 1H, NH), 3.35–3.32 (m, 2H, CH_2), 2.22 (s, 3H, CH_3), 2.15 (s, 3H, CH_3), 1.52–1.47 (m, 2H, CH_2), 1.33–1.30 (m, 2H, CH_2), 0.90 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 171.1, 169.4, 139.5, 139.0, 138.4, 136.7, 133.0, 131.7, 129.7, 129.5, 128.8, 128.7, 128.1, 126.0, 124.5, 117.8, 61.0, 39.5, 31.4, 21.3, 20.9, 20.0, 13.7. Anal. Calcd for $C_{27}H_{29}N_5O_2$: C, 71.19; H, 6.42; N, 15.37. Found: C, 71.43; H, 6.57; N, 15.15.

*4.2.22. N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-4-chloro-N-p-tolylbenzamide (**1z**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol), 4-methylphenyl amine (0.22 g, 2 mmol) and 4-chlorobenzoic acid (0.31 g, 2 mmol), compound **1z** (0.82 g, 86%) was also isolated as white solid. Mp: 199–201 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.28–6.78 (m, 12H, Ar–H), 6.38 (s, 1H, CH), 5.91 (s, 1H, NH), 3.36–3.31 (m, 2H, CH_2), 2.15 (s, 3H, CH_3), 1.51–1.47 (m, 2H, CH_2), 1.33–1.29 (m, 2H, CH_2), 0.90 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.9, 169.2, 139.1, 137.7, 137.1, 135.2, 34.5, 131.7, 130.0, 129.7, 128.9, 127.7, 125.7, 124.6, 117.8, 60.7, 39.6, 31.4, 20.9, 20.0, 13.6. Anal. Calcd for $C_{26}H_{26}ClN_5O_2$: C, 65.61; H, 5.51; N, 14.71. Found: C, 65.89; H, 5.50; N, 14.66.

*4.2.23. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(4-ethoxyphenyl)benzamide (**1a′**).* Operation as above with 4-ethoxyphenyl amine (0.27 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1a′** (0.77 g, 82%) was also isolated as white solid. Mp: 197–198 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.33–6.45 (m, 13H, Ar–H), 6.31 (s, 1H, CH), 5.74 (s, 1H, NH), 3.82 (q, J=7.2 Hz, 2H, OCH_2), 1.39 (s, 9H, $3CH_3$), 1.30 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 171.1, 168.7, 157.4, 139.1, 136.2, 133.0, 131.6, 131.1, 129.7, 129.1, 128.4, 127.5, 126.0, 124.5, 117.8, 113.6, 63.2, 60.7, 51.6, 28.6, 14.6. Anal. Calcd for $C_{27}H_{29}N_5O_3$: C, 68.77; H, 6.20; N, 14.85. Found: C, 68.47; H, 6.32; N, 14.66.

*4.2.24. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-o-tolylbenzamide (**1b′**).* Operation as above with 2-methylphenyl amine (0.22 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1b′** (0.65 g, 74%) was also isolated as white solid. Mp: 164–166 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.82–6.73 (m, 13H, Ar–H), 6.51 (s, 0.3H, 0.3CH), 5.94 (s, 0.7H, 0.7CH), 5.78 (s, 1H, NH), 1.98 (s, 0.8H, $0.8CH_3$), 1.80 (s, 2.2H, $2.2CH_3$), 1.39 (s, 6H, $3CH_3$), 1.33 (s, 3H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.9, 168.7, 139.2, 138.6, 136.6, 136.1, 132.5, 131.2, 130.4, 129.8, 129.4, 128.3, 128.2, 127.8, 127.3, 125.8, 124.9, 124.1, 117.6, 59.5, 51.6, 30.9, 28.6, 28.5, 17.3. Anal. Calcd for $C_{26}H_{27}N_5O_2$: C, 70.73; H, 6.16; N, 15.86. Found: C, 70.54; H, 6.22; N, 15.61.

*4.2.25. N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(3-chlorophenyl)benzamide (**1c′**).* Operation as above with 3-chlorophenyl amine (0.25 g, 2 mmol) and benzoic acid (0.24 g, 2 mmol), compound **1c′** (0.82 g, 87%) was also isolated as white solid. Mp: 165–167 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.34–6.85 (m, 13H, Ar–H), 6.30 (s, 1H, CH), 5.64 (s, 1H, NH), 1.39 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.6, 168.4, 141.6, 139.0, 135.6, 133.2, 131.3, 130.1, 130.0, 129.5, 128.7, 128.5, 128.3, 127.6, 127.0, 125.5, 124.5, 117.9, 65.7, 60.6, 51.6, 30.8, 28.5, 15.1. Anal. Calcd for $C_{25}H_{24}ClN_5O_2$: C, 65.00; H, 5.24; N, 15.16. Found: C, 65.33; H, 5.37; N, 15.12.

*4.2.26. (E)-N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxoethyl)-N-(2-bromophenyl)cinnamamide (**1d′**).* Operation as above with 2-bromophenyl amine (0.34 g, 2 mmol) and cinnamic acid (0.30 g, 2 mmol), compound **1d′** (0.79 g, 74%) was also isolated as white solid. Mp: 211–213 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.68–6.76 (m, 14H, Ar–H), 6.61 (s, 0.8H, 0.8CH), 6.21 (s, 0.2H, 0.2CH), 6.15–6.08 (m, 1H, =CH), 5.92 (s, 0.2H, 0.2NH), 5.82 (s, 0.8H, 0.8NH), 1.36 (s, 7.2H, $3CH_3$), 1.31 (s, 1.8H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.8, 166.3, 143.2, 139.9, 137.9, 135.0, 133.4, 132.9, 132.3, 129.8, 129.6, 128.5, 128.1, 127.9, 126.0, 124.3, 123.7, 118.2, 117.9, 57.9, 51.6, 28.6, 28.4. Anal. Calcd for $C_{27}H_{26}BrN_5O_2$: C, 60.91; H, 4.92; N, 13.15. Found: C, 60.62; H, 4.76; N, 13.37.

*4.2.27. (E)-N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-N-(2-bromophenyl)cinnamamide (**1e′**).* Operation as above with *n*-

butylisocyanide (0.17 g, 2 mmol), 2-bromophenyl amine (0.34 g, 2 mmol) and cinnamic acid (0.30 g, 2 mmol), compound **1e′** (0.83 g, 78%) was also isolated as white solid. Mp: 214–215 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.04–6.76(m, 14H, Ar–H), 6.70 (s, 0.75H, 0.75CH), 6.28 (s, 0.25H, 0.25CH), 6.13–6.08 (m, 1H, =CH), 5.95 (s, 0.75H, 0.75N–H), 5.90 (s, 0.25H, 0.25N–H), 3.34–3.27 (m, 2H, CH_2), 1.51–1.46 (m, 2H, CH_2), 1.35–1.26 (m, 2H, CH_2), 0.92–0.87 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.7, 166.4, 143.4, 140.0, 137.8, 135.0, 133.6, 132.9, 132.4, 129.9, 129.6, 128.6, 128.1, 127.9, 126.1, 124.2, 123.8, 118.1, 118.0, 57.6, 39.6, 31.5, 20.0, 13.7. Anal. Calcd for $C_{27}H_{26}BrN_5O_2$: C, 60.91; H, 4.92; N, 13.15. Found: C, 60.62; H, 4.74; N, 13.00.

*4.2.28. (E)-N-(1-(2-Azidophenyl)-2-(tert-butylamino)-2-oxo ethyl)-N-(2-bromophenyl)-3-(4-chlorophenyl)acrylamide (**1f′**).* Operation as above with 2-bromophenyl amine (0.34 g, 2 mmol) and (*E*)-3-(4-chlorophenyl)acrylic acid (0.36 g, 2 mmol), compound **1f′** (0.82 g, 72%) was also isolated as white solid. Mp: 232–233 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.03–6.76 (m, 13H, Ar–H), 6.59 (s, 0.8H, 0.8CH), 6.13–6.04 (m, 1H, =CH), 5.88 (s, 0.2H, 0.2CH), 5.73 (s, 0.8H, 0.8N–H), 5.30 (s, 0.2H, 0.2N–H), 1.36 (s, 7.25H, $3CH_3$), 1.31 (s, 1.75H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.8, 166.1, 141.8, 139.9, 137.8, 135.4, 133.5, 133.0, 132.3, 129.9, 129.1, 128.8, 128.1, 126.0, 124.2, 123.8, 118.8, 118.0, 58.0, 51.6, 28.6, 28.5. Anal. Calcd for $C_{27}H_{25}BrClN_5O_2$: C, 57.21; H, 4.45; N, 12.35. Found: C, 57.55; H, 4.67; N, 12.16.

*4.2.29. (E)-N-(1-(2-Azidophenyl)-2-(butylamino)-2-oxoethyl)-N-(2-bromophenyl)-3-(4-chlorophenyl)acrylamide (**1g′**).* Operation as above with *n*-butylisocyanide (0.17 g, 2 mmol), 2-bromophenyl amine (0.34 g, 2 mmol) and (*E*)-3-(4-chlorophenyl)acrylic acid (0.36 g, 2 mmol), compound **1g′** (0.79 g, 70%) was also isolated as white solid. Mp: 215–216 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.06–6.76(m, 12H, Ar–H), 6.69 (s, 0.75H, 0.75CH), 6.20 (s, 0.25H, 0.25CH), 6.10–6.04 (m, 1H, =CH), 5.89 (s, 0.75H, 0.75N–H), 5.86 (s, 0.25H, 0.25N–H), 3.33–3.27 (m, 2H, CH_2), 1.50–1.45 (m, 2H, CH_2), 1.34–1.26 (m, 2H, CH_2), 0.92–0.87 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.7, 166.2, 142.0, 140.0, 137.7, 135.4, 133.6, 133.5, 133.0, 132.3, 130.0, 129.1, 128.8, 128.2, 126.0, 124.1, 123.8, 118.6, 118.0, 57.7, 39.7, 31.4, 20.0, 13.7. Anal. Calcd for $C_{27}H_{25}BrClN_5O_2$: C, 57.21; H, 4.45; N, 12.35. Found: C, 57.66; H, 4.43; N, 12.64.

4.3. Synthesis of the 3,4-dihydroquinazoline 3 via intramolecular aza-Wittig reaction

*4.3.1. N-tert-Butyl-3-(4-chlorophenyl)-3,4-dihydro quinazoline-4-carboxamide (**3a**).* Methyldiphenyl phosphine (0.20 g, 1 mmol) was added to a solution of azide **1a** (0.39 g, 1 mmol) in dry toluene (10 mL) at stirred condition. After stirring for about 2 h, iminophosphorane **2a** was formed which was monitored by TLC. Then the solution was heated to reflux for 1 h without isolated. After the reaction was completed, the solvent was evaporated and the residue was purified by column chromatography to give 0.31 g (92%) of 3,4-dihydroquinazoline **3a** as white solid. Mp: 200–203 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.64 (s, 1H, N=CH), 7.40–7.17 (m, 8H, Ar–H), 5.64 (s, 1H, NH), 5.30 (s, 1H, CH), 1.25 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.9, 144.6, 141.0, 139.2, 130.8, 129.7, 129.5, 126.7, 126.1, 125.4, 120.8, 120.6, 63.0, 51.8, 28.3. MS *m*/*z*: 341 (M^+, 2), 256 (12), 241 (100). Anal. Calcd for $C_{19}H_{20}ClN_3O$: C, 66.76; H, 5.90; N, 12.29. Found: C, 66.93; H, 5.99; N, 12.36.

*4.3.2. N-Butyl-3-p-tolyl-3,4-dihydroquinazoline-4-carboxamide (**3b**).* Operation as above with azide **1b** (0.37 g, 1 mmol), compound **3b** (0.26 g, 82%) was also isolated as white solid. Mp: 184–186 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.62 (s, 1H, Ar–H), 7.32–7.08 (m, 8H, Ar–H), 6.24 (s, 1H, NH), 5.45 (s, 1H, CH), 3.28–3.09 (m, 2H, CH_2), 2.35 (s, 3H, CH_3), 1.38–1.30 (m, 2H, CH_2), 1.16–1.10 (m, 2H, CH_2), 0.80 (t, *J*=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.1, 145.2, 139.9, 139.3, 135.3, 130.2, 129.3, 126.4, 126.3, 124.9, 120.7, 119.3, 62.6, 62.5, 39.4, 31.3, 20.7, 19.7. MS *m*/*z*: 321 (M^+, 1) [M^+], 221 (100). Anal. Calcd for $C_{20}H_{23}N_3O$: C, 74.74; H, 7.21; N, 13.07. Found: C, 74.85; H, 7.33; N, 13.21.

*4.3.3. N-Butyl-3-(4-chlorophenyl)-2-(trifluoromethyl)-3,4-dihydroquinazoline-4-carboxamide (**3c**).* Operation as above with azide **1c** (0.45 g, 1 mmol), compound **3c** (0.35 g, 85%) was also isolated as white solid. Mp: 185–187 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.48–7.19 (m, 8H, Ar–H), 5.77 (s, 1H, NH), 5.09 (s, 1H, CH), 3.26–3.20 (m, 2H, CH_2), 1.43–1.37 (m, 2H, CH_2), 1.26–1.21 (m, 2H, CH_2), 0.87 (t, *J*=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.7, 144.1, 143.8, 141.1, 138.6, 133.9, 129.9, 128.4, 127.7, 126.3, 126.0, 121.3, 119.8, 117.0, 66.6, 39.7, 31.2, 19.8, 13.5. MS *m*/*z*: 409 (M^+, 1), 308 (100), 221 (31). Anal. Calcd for $C_{20}H_{19}ClF_3N_3O$: C, 58.61; H, 4.67; N, 10.25. Found: C, 58.75; H, 4.58; N, 10.12.

*4.3.4. N-tert-Butyl-3-phenyl-2-(trifluoromethyl)-3,4-dihydro quinazoline-4-carboxamide (**3d**).* Operation as above with azide **1d** (0.42 g, 1 mmol), compound **3d** (0.33 g, 88%) was also isolated as white solid. Mp: 189–190 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.44–7.23 (m, 9H, Ar–H), 5.95 (s, 1H, NH), 5.04 (s, 1H, CH), 1.31 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.2, 142.8, 138.4, 129.6, 128.4, 128.0, 126.9, 125.7, 121.9, 119.5, 117.7, 67.3, 51.9, 29.0, 28.5. MS *m*/*z*: 375 (M^+, 1), 275 (100). Anal. Calcd for $C_{20}H_{20}F_3N_3O$: C, 63.99; H, 5.37; N, 11.19. Found: C, 63.72; H, 5.26; N, 11.31.

*4.3.5. N-tert-Butyl-3-(4-chlorophenyl)-2-methyl-3,4-dihydro quinazoline-4-carboxamide (**3e**).* Operation as above with azide **1e** (0.40 g, 1 mmol), compound **3e** (0.28 g, 80%) was also isolated as white solid. Mp: 196–199 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.38–7.08 (m, 8H, Ar–H), 5.52 (s, 1H, NH), 5.02 (s, 1H, CH), 2.10 (s, 3H, CH_3), 1.25 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.4, 154.8, 142.1, 140.8, 133.1, 129.6, 129.4, 128.1, 125.3, 125.2, 124.3, 121.0, 66.5, 51.6, 28.4, 23.2. MS *m*/*z*: 355 (M^+, 1), 255 (100). Anal. Calcd for $C_{20}H_{22}ClN_3O$: C, 67.50; H, 6.23; N, 11.81. Found: C, 67.72; H, 6.41; N, 11.77.

*4.3.6. N-tert-Butyl-2-methyl-3-p-tolyl-3,4-dihydro quinazoline-4-carboxamide (**3f**).* Operation as above with azide **1f** (0.38 g, 1 mmol), compound **3f** (0.20 g, 60%) was also isolated as white solid. Mp: 164–167 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.29–7.09 (m, 8H, Ar–H), 5.64 (s, 1H, NH), 5.06 (s, 1H, CH), 2.37 (s, 3H, CH_3), 2.07 (s, 3H, CH_3), 1.26 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.8, 155.2, 141.0, 140.9, 137.5, 130.1, 129.1, 126.6, 125.3, 125.0, 124.1, 121.0, 67.0, 51.5, 28.4, 23.3, 21.0. MS *m*/*z*: 335 (M^+, 1), 235 (100). Anal. Calcd for $C_{21}H_{25}N_3O$: C, 75.19; H, 7.51; N, 12.53. Found: C, 75.53; H, 7.63; N, 12.50.

*4.3.7. N-tert-Butyl-3-(4-chlorophenyl)-2-ethyl-3,4-dihydro quinazoline-4-carboxamide (**3g**).* Operation as above with azide **1g** (0.41 g, 1 mmol), compound **3g** (0.30 g, 82%) was also isolated as white solid. Mp: 158–160 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.34–7.08 (m, 8H, Ar–H), 5.56 (s, 1H, NH), 4.96 (s, 1H, CH), 2.50–2.34 (m, 2H, CH_2), 1.87 (t, *J*=7.2 Hz, 3H, CH_3), 1.25 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.5, 159.1, 142.4, 141.0, 132.6, 129.5, 129.4, 127.7, 125.6, 125.5, 124.5, 121.1, 66.4, 51.6, 28.6, 28.4, 11.7. MS *m*/*z*: 369 (M^+, 1), 269 (100). Anal. Calcd for $C_{21}H_{24}ClN_3O$: C, 68.19; H, 6.54; N, 11.36. Found: C, 68.46; H, 6.28; N, 11.22.

*4.3.8. N-tert-Butyl-2-(4-nitrophenyl)-3-p-tolyl-3,4-dihydroquinazoline-4-carboxamide (**3h**).* Operation as above with azide **1h** (0.49 g, 1 mmol), compound **3h** (0.41 g, 93%) was also isolated as white solid. Mp: 98–101 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.12–6.97 (m,

12H, Ar–H), 5.50 (s, 1H, NH), 5.25 (s, 1H, CH), 2.24 (s, 3H, CH_3), 1.23 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.2, 152.9, 148.3, 142.2, 142.1, 141.4, 135.2, 130.7, 129.7, 126.7, 126.0, 125.2, 124.2, 123.4, 122.1, 65.6, 51.8, 28.4, 20.8. MS *m*/*z*: 442 (M^+, 2), 342 (100), 296 (16). Anal. Calcd for $C_{26}H_{26}N_4O_3$: C, 70.57; H, 5.92; N, 12.66. Found: C, 70.86; H, 5.75; N, 12.33.

*4.3.9. N-tert-Butyl-3-(4-chlorophenyl)-2-(4-nitrophenyl)-3,4-dihydroquinazoline-4-carboxamide (**3i**).* Operation as above with azide **1i** (0.51 g, 1 mmol), compound **3i** (0.44 g, 95%) was also isolated as white solid. Mp: 105–107 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 8.15–7.11 (m, 12H, Ar–H), 5.40 (s, 1H, NH), 5.24 (s, 1H, CH), 1.22 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.8, 152.7, 148.5, 143.1, 141.7, 141.4, 130.8, 130.7, 129.9, 129.1, 127.0, 125.9, 125.5, 125.4, 123.5, 122.0, 65.2, 51.9, 28.4. MS *m*/*z*: 462 (M^+, 2), 362 (100), 316 (14). Anal. Calcd for $C_{25}H_{23}ClN_4O_3$: C, 64.86; H, 5.01; N, 12.10. Found: C, 64.68; H, 5.17; N, 12.26.

*4.3.10. N-Butyl-3-(4-chlorophenyl)-2-phenyl-3,4-dihydro quinazoline-4-carboxamide (**3j**).* Operation as above with azide **1j** (0.46 g, 1 mmol), compound **3j** (0.38 g, 91%) was also isolated as white solid. Mp: 179–181 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.73 (d, J=7.8 Hz, 2H, Ar–H), 7.52–7.11 (m, 11H, Ar–H), 5.69 (s, 1H, NH), 5.31 (s, 1H, CH), 3.21–3.17 (m, 2H, CH_2), 1.36–1.31 (m, 2H, CH_2), 1.18–1.15 (m, 2H, CH_2), 0.79 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.0, 154.8, 143.9, 141.8, 135.3, 130.2, 130.0, 129.8, 129.7, 128.7, 128.3, 126.2, 126.0, 125.4, 125.2, 121.9, 65.0, 65.0, 39.6, 31.2, 19.8. MS *m*/*z*: 417 (M^+, 1), 317 (100). Anal. Calcd for $C_{25}H_{24}ClN_3O$: C, 71.85; H, 5.79; N, 10.05. Found: C, 72.03; H, 5.56; N, 10.30.

*4.3.11. N-tert-Butyl-3-(4-chlorophenyl)-2-p-tolyl-3,4-dihydroquinazoline-4-carboxamide (**3k**).* Operation as above with azide **1k** (0.48 g, 1 mmol), compound **3k** (0.38 g, 88%) was also isolated as white solid. Mp: 233–235 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.61–7.08 (m, 12H, Ar–H), 5.65 (s, 1H, N–H), 5.20 (s, 1H, CH), 2.32 (s, 3H, CH_3), 1.22 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.2, 154.4, 144.2, 141.6, 140.6, 132.4, 129.9, 129.6, 129.5, 129.2, 128.8, 126.3, 126.0, 125.2, 125.0, 122.3, 65.8, 51.7, 28.4, 21.4. MS *m*/*z*: 431 (M^+, 1), 331 (100). Anal. Calcd for $C_{26}H_{26}ClN_3O$: C, 72.29; H, 6.07; N, 9.73. Found: C, 72.47; H, 6.22; N, 9.52.

*4.3.12. N-tert-Butyl-3-(4-chlorophenyl)-2-(2-fluorophenyl)-3,4-dihydroquinazoline-4-carboxamide (**3l**).* Operation as above with azide **1l** (0.48 g, 1 mmol), compound **3l** (0.40 g, 91%) was also isolated as white solid. Mp: 114–117 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.89–6.70 (m, 13H, Ar–H, and N–H), 5.18 (s, 1H, CH), 1.34 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.2, 160.2, 158.6, 149.8, 142.9, 140.3, 132.4, 132.3, 132.2, 131.2, 129.2, 128.8, 127.1, 126.8, 125.3, 123.9, 123.8, 121.7, 115.8, 115.6, 66.6, 66.6, 51.9, 28.5. MS *m*/*z*: 435 (M^+, 1), 335 (100). Anal. Calcd for $C_{25}H_{23}ClFN_3O$: C, 68.88; H, 5.32; N, 9.64. Found: C, 68.67; H, 5.16; N, 9.38.

*4.3.13. N-tert-Butyl-3-(4-chlorophenyl)-2-o-tolyl-3,4-dihydroquinazoline-4-carboxamide (**3m**).* Operation as above with azide **1m** (0.48 g, 1 mmol), compound **3m** (0.15 g, 35%) was also isolated as white solid. Mp: 224–227 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.48–7.05 (m, 12H, Ar–H), 5.47 (s, 1H, NH), 5.24 (s, 1H, CH), 2.53 (s, 3H, CH_3), 1.23 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.1, 155.1, 143.1, 141.5, 137.7, 134.9, 130.9, 130.5, 129.7, 129.5, 128.6, 126.1, 125.9, 125.8, 125.7, 125.0, 121.4, 65.8, 51.7, 28.3, 20.1. MS *m*/*z*: 431 (M^+, 1), 331 (100). Anal. Calcd for $C_{26}H_{26}ClN_3O$: C, 72.29; H, 6.07; N, 9.73. Found: C, 72.01; H, 6.07; N, 9.99.

*4.3.14. N-tert-Butyl-3-methyl-2-phenyl-3,4-dihydro quinazoline-4-carboxamide (**3n**).* Operation as above with azide **1n** (0.37 g, 1 mmol), compound **3n** (0.13 g, 40%) was also isolated as white solid. Mp: 219–221 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.61–7.14 (m, 9H, Ar–H), 5.65 (s, 1H, NH), 4.84 (s, 1H, CH), 3.11 (s, 3H, CH_3), 1.28 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 169.5, 157.7, 141.8, 135.8, 129.8, 129.3, 128.5, 128.3, 125.7, 125.2, 124.9, 120.4, 65.7, 51.5, 41.4, 28.5. MS *m*/*z*: 321 (M^+, 2), 221 (100), 193 (8). Anal. Calcd for $C_{20}H_{23}N_3O$: C, 74.74; H, 7.21; N, 13.07. Found: C, 74.97; H, 7.31; N, 13.01.

*4.3.15. N-tert-Butyl-3-ethyl-2-phenyl-3,4-dihydro quinazoline-4-carboxamide (**3o**).* Operation as above with azide **1o** (0.38 g, 1 mmol), compound **3o** (0.17 g, 50%) was also isolated as white solid. Mp: 215–217 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.63–7.15 (m, 9H, Ar–H), 5.83 (s, 1H, NH), 4.93 (s, 1H, CH), 3.52–3.40 (m, 2H, CH_2), 1.28 (s, 9H, $3CH_3$), 1.16 (t, J=6.6 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 170.1, 157.7, 141.8, 136.0, 129.9, 129.2, 128.7, 128.2, 126.0, 125.4, 124.8, 121.1, 62.3, 51.4, 47.5, 28.5, 14.0. MS *m*/*z*: 335 (M^+, 1), 235 (100), 207 (18). Anal. Calcd for $C_{21}H_{25}N_3O$: C, 75.19; H, 7.51; N, 12.53. Found: C, 75.42; H, 7.63; N, 12.33.

*4.3.16. N-tert-Butyl-3-isopropyl-2-phenyl-3,4-dihydro quinazoline-4-carboxamide (**3p**).* Operation as above with azide **1p** (0.39 g, 1 mmol), compound **3p** (0.22 g, 62%) was also isolated as white solid. Mp: 160–161 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.74–7.16 (m, 9H, Ar–H), 6.05 (s, 1H, NH), 4.95 (s, 1H, CH), 4.09–4.05 (m, H, CH), 1.36 (d, J=6.6 Hz, 3H, CH_3), 1.26 (s, 9H, $3CH_3$), 1.02 (d, J=6.6 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.5, 157.7, 141.8, 136.3, 130.4, 128.9, 128.8, 128.2, 126.2, 125.7, 124.6, 122.4, 56.9, 52.2, 51.3, 28.5, 21.4, 20.9. MS *m*/*z*: 349 (M^+, 1), 249 (100), 207 (96). Anal. Calcd for $C_{22}H_{27}N_3O$: C, 75.61; H, 7.79; N, 12.02. Found: C, 75.93; H, 7.87; N, 12.05.

*4.3.17. N-tert-Butyl-3-cyclohexyl-2-phenyl-3,4-dihydroquinazoline-4-carboxamide (**3q**).* Operation as above with azide **1q** (0.43 g, 1 mmol), compound **3q** (0.26 g, 67%) was also isolated as white solid. Mp: 189–191 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.75–7.16 (m, 9H, Ar–H), 6.08 (s, 1H, N–H), 4.98 (s, 1H, CH), 3.61–3.57 (m, 1H, NCH), 2.16–1.03 (m, 19H, $5CH_2$, and $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.4, 157.6, 141.7, 136.1, 130.3, 128.7, 128.1, 126.2, 125.5, 124.4, 122.4, 60.4, 57.8, 51.2, 31.5, 28.4, 25.5, 25.3, 24.8. MS *m*/*z*: 389 (M^+, 1), 289 (63), 207 (100). Anal. Calcd for $C_{25}H_{31}N_3O$: C, 77.08; H, 8.02; N, 10.79. Found: C, 76.83; H, 8.25; N, 10.68.

*4.3.18. N,3-Di-tert-butyl-2-(4-chlorophenyl)-3,4-dihydroquinazoline-4-carboxamide (**3r**).* Operation as above with azide **1r** (0.44 g, 1 mmol), compound **3r** (0.34 g, 85%) was also isolated as white solid. Mp: 192–194 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.85–7.25 (m, 8H, Ar–H), 5.43 (s, 1H, N–H), 5.02 (s, 1H, CH), 1.15 (s, 9H, $3CH_3$), 1.13 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.4, 158.5, 142.5, 139.2, 136.0, 130.4, 129.0, 128.3, 125.8, 125.3, 124.0, 59.5, 58.8, 51.3, 31.1, 28.2. MS *m*/*z*: 397 (M^+, 1), 297 (13), 241 (100). Anal. Calcd for $C_{23}H_{28}ClN_3O$: C, 69.42; H, 7.09; N, 10.56. Found: C, 69.31; H, 7.17; N, 10.72.

*4.3.19. N,3-Di-tert-butyl-2-p-tolyl-3,4-dihydroquinazoline-4-carboxamide (**3s**).* Operation as above with azide **1s** (0.42 g, 1 mmol), compound **3s** (0.33 g, 88%) was also isolated as white solid. Mp: 182–184 °C, ^1H NMR ($CDCl_3$, 600 MHz): δ 7.77–7.22 (m, 8H, Ar–H), 5.71 (s, 1H, N–H), 5.02 (s, 1H, CH), 2.40 (s, 3H, CH_3), 1.15 (s, 18H, $6CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.6, 159.2, 142.5, 140.3, 137.7, 128.9, 128.7, 126.1, 125.6, 125.4, 123.9, 59.5, 58.9, 51.2, 31.0, 28.3, 21.3. MS *m*/*z*: 377 (M^+, 1), 277 (13), 221 (100). Anal. Calcd for $C_{24}H_{31}N_3O$: C, 76.35; H, 8.28; N, 11.13. Found: C, 76.41; H, 8.47; N, 11.07.

*4.3.20. N,3-Di-tert-butyl-2-phenyl-3,4-dihydroquinazoline-4-carboxamide (**3t**).* Operation as above with azide **1t** (0.41 g, 1 mmol),

compound **3t** (0.34 g, 93%) was also isolated as white solid. Mp: 172–173 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.89–7.24 (m, 9H, Ar–H), 5.62 (s, 1H, NH), 5.03 (s, 1H, CH), 1.16 (s, 9H, $3CH_3$), 1.15 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.5, 159.3, 142.6, 140.6, 130.0, 129.0, 128.8, 128.0, 126.0, 125.8, 125.3, 124.0, 59.5, 59.0, 51.2, 31.0, 28.2. MS *m*/*z*: 363 (M^+, 1), 263 (9), 207 (100). Anal. Calcd for $C_{23}H_{29}N_3O$: C, 76.00; H, 8.04; N, 11.56. Found: C, 76.31; H, 8.17; N, 11.42.

*4.3.21. N-tert-Butyl-2-phenyl-3-p-tolyl-3,4-dihydro quinazoline-4-carboxamide (**3u**).* Operation as above with azide **1u** (0.44 g, 1 mmol), compound **3u** (0.35 g, 88%) was also isolated as white solid. Mp: 160–162 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.74 (d, J=7.2 Hz, 2H, Ar–H), 7.48–6.95 (m, 11H, Ar–H), 5.83 (s, 1H, NH), 5.22 (s, 1H, CH), 2.23 (s, 3H, CH_3), 1.25 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 169.6, 154.5, 143.0, 141.5, 135.8, 134.4, 129.9, 129.5, 129.4, 129.2, 128.2, 126.4, 126.0, 124.9, 124.0, 122.3, 66.1, 51.6, 28.4, 20.7. MS *m*/*z*: 397 (M^+, 1), 297 (100). Anal. Calcd for $C_{26}H_{27}N_3O$: C, 78.56; H, 6.85; N, 10.57. Found: C, 78.74; H, 6.96; N, 10.38.

*4.3.22. N-Butyl-2-phenyl-3-p-tolyl-3,4-dihydroquinazoline-4-carboxamide (**3v**).* Operation as above with azide **1v** (0.44 g, 1 mmol), compound **3v** (0.36 g, 90%) was also isolated as white solid. Mp: 182–183 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.75 (d, J=6.6 Hz, 2H, Ar–H), 7.50–6.95 (m, 11H, Ar–H), 5.83 (s, 1H, N–H), 5.33 (s, 1H, CH), 3.26–3.15 (m, 2H, CH_2), 2.22 (s, 3H, CH_3), 1.38–1.33 (m, 2H, CH_2), 1.20–1.16 (m, 2H, CH_2), 0.80 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.4, 154.8, 142.9, 141.8, 135.7, 134.3, 129.8, 129.6, 129.3, 128.1, 126.1, 125.8, 124.9, 124.1, 121.9, 65.4, 65.4, 39.4, 31.2, 20.7, 19.7, 13.5. MS *m*/*z*: 397 (M^+, 1), 297 (100). Anal. Calcd for $C_{26}H_{27}N_3O$: C, 78.56; H, 6.85; N, 10.57. Found: C, 78.80; H, 6.69; N, 10.76.

*4.3.23. N-Butyl-3-(4-chlorophenyl)-2-p-tolyl-3,4-dihydro quinazoline-4-carboxamide (**3w**).* Operation as above with azide **1w** (0.48 g, 1 mmol), compound **3w** (0.40 g, 92%) was also isolated as white solid. Mp: 163–165 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.62–7.07 (m, 12H, Ar–H), 5.71 (s, 1H, NH), 5.30 (s, 1H, CH), 3.22–3.15 (m, 2H, CH_2), 2.31 (s, 3H, CH_3), 1.36–1.30 (m, 2H, CH_2), 1.19–1.14 (m, 2H, CH_2), 0.79 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 170.1, 154.7, 144.1, 141.9, 140.6, 132.3, 129.9, 129.7, 129.6, 129.1, 128.7, 126.1, 126.0, 125.3, 125.1, 121.9, 65.1, 39.6, 31.2, 21.4, 19.8, 13.5. MS *m*/*z*: 431 (M^+, 1), 331 (100). Anal. Calcd for $C_{26}H_{26}ClN_3O$: C, 72.29; H, 6.07; N, 9.73. Found: C, 72.46; H, 6.15; N, 9.56.

*4.3.24. N-Butyl-2,3-di(p-tolyl)-3,4-dihydroquinazoline-4-carboxamide (**3x**).* Operation as above with azide **1x** (0.46 g, 1 mmol), compound **3x** (0.37 g, 89%) was also isolated as white solid. Mp: 175–177 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.63–6.95 (m, 12H, Ar–H), 5.86 (s, 1H, NH), 5.31 (s, 1H, CH), 3.27–3.13 (m, 2H, CH_2), 2.30 (s, 3H, CH_3), 2.23 (s, 3H, CH_3), 1.37–1.32 (m, 2H, CH_2), 1.23–1.14 (m, 2H, CH_2), 0.80 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 170.5, 154.8, 143.2, 141.9, 140.2, 134.3, 132.8, 129.7, 129.4, 129.0, 126.3, 125.8, 124.9, 124.1, 122.1, 65.6, 39.5, 31.3, 21.4, 20.7, 19.8, 13.5. MS *m*/*z*: 411 (M^+, 1), 311 (100). Anal. Calcd for $C_{27}H_{29}N_3O$: C, 78.80; H, 7.10; N, 10.21. Found: C, 78.56; H, 6.98; N, 10.28.

*4.3.25. N-Butyl-2,3-bis(4-chlorophenyl)-3,4-dihydro quinazoline-4-carboxamide (**3y**).* Operation as above with azide **1y** (0.50 g, 1 mmol), compound **3y** (0.41 g, 91%) was also isolated as white solid. Mp: 166–169 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.70–7.13 (m, 12H, Ar–H), 5.59 (s, 1H, NH), 5.29 (s, 1H, CH), 3.20–3.15 (m, 2H, CH_2), 1.35–1.30 (m, 2H, CH_2), 1.19–1.14 (m, 2H, CH_2), 0.80 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.9, 153.8, 143.6, 141.8, 136.2, 133.8, 131.2, 130.2, 129.7, 128.8, 128.5, 126.3, 125.9, 125.4, 125.2, 121.8, 64.8, 39.6, 31.2, 19.7, 13.5. MS *m*/*z*: 451 (M^+, 1), 351 (100). Anal. Calcd for $C_{25}H_{23}Cl_2N_3O$: C, 66.38; H, 5.12; N, 9.29. Found: C, 66.61; H, 5.27; N, 9.27.

*4.3.26. N-Butyl-2-(4-chlorophenyl)-3-p-tolyl-3,4-dihydro quinazoline-4-carboxamide (**3z**).* Operation as above with azide **1z** (0.48 g, 1 mmol), compound **3z** (0.39 g, 90%) was also isolated as white solid. Mp: 178–180 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.71–6.96 (m, 12H, Ar–H), 5.69 (s, 1H, NH), 5.31 (s, 1H, CH), 3.22–3.16 (m, 2H, CH_2), 2.25 (s, 3H, CH_3), 1.35–1.31 (m, 2H, CH_2), 1.20–1.15 (m, 2H, CH_2), 0.81 (t, J=7.2 Hz, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 170.3, 154.1, 142.1, 141.9, 136.0, 134.7, 131.2, 129.5, 128.5, 126.1, 125.0, 124.2, 121.9, 65.3, 39.6, 31.3, 20.8, 19.8, 13.6. MS *m*/*z*: 431 (M^+, 1), 331 (100). Anal. Calcd for $C_{26}H_{26}ClN_3O$: C, 72.29; H, 6.07; N, 9.73. Found: C, 72.47; H, 6.22; N, 9.61.

*4.3.27. N-tert-Butyl-3-(4-ethoxyphenyl)-2-phenyl-3,4-dihydroquinazoline-4-carboxamide (**3a′**).* Operation as above with azide **1a′** (0.47 g, 1 mmol), compound **3a′** (0.39 g, 92%) was also isolated as white solid. Mp: 211–213 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.23–6.66 (m, 13H, Ar–H), 5.80 (s, 1H, NH), 5.19 (s, 1H, CH), 3.96 (q, J=7.2 Hz, 2H, CH_2), 1.35 (t, J=7.2 Hz, 3H, CH_3), 1.25 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.7, 156.2, 154.6, 141.6, 138.6, 135.8, 129.9, 129.6, 129.3, 128.2, 126.3, 125.9, 125.7, 124.9, 122.1, 114.5, 66.4, 63.5, 51.6, 28.5, 14.7. MS *m*/*z*: 427 (M^+, 1), 327 (100), 299 (9), 269 (8). Anal. Calcd for $C_{27}H_{29}N_3O_2$: C, 75.85; H, 6.84; N, 9.83. Found: C, 75.61; H, 6.98; N, 9.57.

*4.3.28. N-tert-Butyl-2-phenyl-3-o-tolyl-3,4-dihydro quinazoline-4-carboxamide (**3b′**).* Operation as above with azide **1b′** (0.44 g, 1 mmol), compound **3b′** (0.35 g, 88%) was also isolated as white solid. Mp: 184–187 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.93–6.88 (m, 13H, Ar–H), 5.79 (s, 1H, NH), 5.06 (s, 1H, CH), 1.78 (s, 3H, CH_3), 1.26 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.8, 156.1, 144.6, 135.5, 134.5, 131.0, 130.2, 129.7, 129.2, 127.9, 127.4, 127.1, 126.9, 125.9, 125.8, 124.7, 122.5, 66.6, 51.7, 28.5, 18.0. MS *m*/*z*: 397 (M^+, 1), 297 (100). Anal. Calcd for $C_{26}H_{27}N_3O$: C, 78.56; H, 6.85; N, 10.57. Found: C, 78.33; H, 6.76; N, 10.71.

*4.3.29. N-tert-Butyl-3-(3-chlorophenyl)-2-phenyl-3,4-dihydroquinazoline-4-carboxamide (**3c′**).* Operation as above with azide **1c′** (0.46 g, 1 mmol), compound **3c′** (0.35 g, 84%) was also isolated as white solid. Mp: 163–164 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.76–6.95 (m, 13H, Ar–H), 5.60 (s, 1H, NH), 5.25 (s, 1H, CH), 1.23 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.0, 154.4, 146.5, 141.5, 135.2, 134.4, 130.3, 129.6, 129.5, 128.4, 126.3, 126.2, 125.2, 124.6, 123.9, 122.5, 122.3, 65.5, 65.5, 51.8, 28.4. MS *m*/*z*: 417 (M^+, 1), 317 (100). Anal. Calcd for $C_{25}H_{24}ClN_3O$: C, 71.85; H, 5.79; N, 10.05. Found: C, 72.07; H, 5.98; N, 10.01.

*4.3.30. (E)-3-(2-Bromophenyl)-N-tert-butyl-2-styryl-3,4-dihydroquinazoline-4-carboxamide (**3d′**).* Operation as above with azide **1d′** (0.53 g, 1 mmol), compound **3d′** (0.40 g, 82%) was also isolated as white solid. Mp: 174–176 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.89–7.06 (m, 14H, Ar–H), 6.54 (s, 0.3H, 0.3NH), 5.84 (s, 0.7H, 0.7NH), 6.34–6.31 (m, 1H, =CH), 5.20 (s, 0.3H, 0.3CH), 5.04 (s, 0.7H, 0.7CH), 1.26 (s, 7H, $3CH_3$), 1.20 (s, 2H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.7, 152.4, 142.2, 141.4, 138.7, 135.5, 134.1, 133.7, 131.6, 130.7, 130.4, 130.3, 129.4, 129.1, 128.9, 128.6, 128.5, 128.5, 128.4, 127.5, 125.3, 125.2, 124.8, 123.3, 121.6, 120.7, 66.0, 51.5, 28.4, 28.2, 16.6, 16.1. MS *m*/*z*: 487 (M^+, 1), 389 (100), 307 (11), 231 (36). Anal. Calcd for $C_{27}H_{26}BrN_3O$: C, 66.40; H, 5.37; N, 8.60. Found: C, 66.68; H, 5.51; N, 8.42.

*4.3.31. (E)-3-(2-Bromophenyl)-N-butyl-2-styryl-3,4-dihydro quinazoline-4-carboxamide (**3e′**).* Operation as above with azide **1e′** (0.53 g, 1 mmol), compound **3e′** (0.36 g, 73%) was also isolated as

white solid. Mp: 202–203 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.91–7.14 (m, 13H, Ar–H), 6.53–6.31 (m, 1H, =CH), 5.90 (s, 0.8H, 0.8N–H), 5.55 (s, 0.2H, 0.2NH), 5.23 (s, 0.2H, 0.2CH), 5.11 (s, 0.8H, 0.8CH), 3.22–3.19 (m, 2H, CH_2), 1.41–1.37 (m, 2H, CH_2), 1.22–1.18 (m, 2H, CH_2), 0.84–0.78 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.6, 152.5, 142.3, 141.6, 139.0, 135.6, 133.8, 130.7, 129.5, 129.4, 129.1, 128.7, 128.5, 127.6, 125.5, 125.4, 124.9, 123.3, 121.3, 120.7, 65.5, 39.5, 31.3, 19.8, 13.6. MS *m*/*z*: 487 (M^+, 1), 387 (100), 307 (9), 231 (30). Anal. Calcd for $C_{27}H_{26}BrN_3O$: C, 66.40; H, 5.37; N, 8.60. Found: C, 66.78; H, 5.49; N, 8.76.

*4.3.32. (E)-2-(4-Chlorostyryl)-3-(2-bromophenyl)-N-tert-butyl-3,4-dihydroquinazoline-4-carboxamide (**3f′**).* Operation as above with azide **1f′** (0.57 g, 1 mmol), compound **3f′** (0.44 g, 85%) was also isolated as white solid. Mp: 179–182 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.91–6.12 (m, 14H, Ar–H), 5.77 (s, 0.75H, 0.75NH), 5.54 (s, 0.25H, 0.25NH), 5.28 (s, 0.25H, 0.25CH), 5.04 (s, 0.75H, 0.75CH), 1.26 (s, 6.7H, $3CH_3$), 1.20 (s, 2.3H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.4, 152.9, 141.9, 140.5, 138.2, 137.9, 135.0, 133.9, 130.8, 129.8, 129.4, 128.8, 126.6, 125.6, 125.3, 124.8, 124.4, 123.3, 121.3, 120.6, 66.1, 51.7, 28.4, 28.2. MS *m*/*z*: 523 (M^+, 1), 423 (100), 231 (23). Anal. Calcd for $C_{27}H_{25}BrClN_3O$: C, 62.02; H, 4.82; N, 8.04. Found: C, 62.38; H, 4.64; N, 8.32.

*4.3.33. (E)-2-(4-Chlorostyryl)-3-(2-bromophenyl)-N-butyl-3,4-dihydroquinazoline-4-carboxamide (**3g′**).* Operation as above with azide **1g′** (0.57 g, 1 mmol), compound **3g′** (0.42 g, 80%) was also isolated as white solid. Mp: 166–167 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.94–7.13 (m, 13H, Ar–H), 6.48 (s, 0.2H, 0.2NH), 6.45–6.27 (m, 1H, =CH), 5.80 (s, 0.8H, 0.8NH), 5.27 (s, 0.2H, 0.2CH), 5.10 (s, 0.8H, 0.8CH), 3.22–3.19 (m, 2H, CH_2), 1.40–1.34 (m, 2H, CH_2), 1.23–1.16 (m, 2H, CH_2), 0.84–0.78 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 170.5, 152.3, 142.3, 141.6, 137.6, 134.9, 134.1, 133.9, 130.8, 129.6, 129.5, 129.0, 128.8, 128.7, 125.6, 125.5, 125.0, 123.3, 121.3, 65.5, 39.5, 31.3, 19.8, 13.6. MS *m*/*z*: 523 (M^+, 1), 423 (100), 231 (32). Anal. Calcd for $C_{27}H_{25}BrClN_3O$: C, 62.02; H, 4.82; N, 8.04. Found: C, 62.43; H, 4.77; N, 8.35.

4.4. Synthesis of the 6,12-dihydroindolo[2,1-*b*]quinazolines 4 via the intramolecular Heck reaction

*4.4.1. 6-Benzylidene-N-tert-butyl-6,12-dihydroindolo[2,1-b]quinazoline-12-carboxamide (**4a**).* To a solution of 3,4-dihydroquinazoline **3d′** (0.49 g, 1 mmol) in DMF (10 mL) were added solid K_2CO_3 (0.14 g, 1 mmol), $Pd(OAc)_2$ (0.02 g, 0.1 mmol), and Ph_3P (0.05 g, 0.2 mmol). The reaction mixture was stirred for additional 6 h at 110 °C. Afterward the solvent was evaporated under reduced pressure and the residue was purified by column chromatography to give 0.24 g (58%) of **4a** as red solid. Mp: >300 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.11–6.88 (m, 14H, Ar–H), 5.57 (s, 1H, CH), 5.55 (s, 1H, NH), 1.19 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.9, 153.3, 143.4, 135.4, 133.0, 130.0, 129.6, 129.1, 128.6, 126.5, 126.1, 125.9, 123.1, 122.5, 122.2, 119.0, 108.7, 60.3, 51.7, 28.3. MS *m*/*z*: 407 (M^+, 2), 307 (100). Anal. Calcd for $C_{27}H_{25}N_3O$: C, 79.58; H, 6.18; N, 10.31. Found: C, 79.33; H, 6.15; N, 10.03.

*4.4.2. 6-Benzylidene-N-butyl-6,12-dihydroindolo[2,1-b] quinazoline-12-carboxamide (**4b**).* Operation as above with 3,4-dihydroquinazoline **3e′** (0.49 g, 1 mmol), compound **4b** (0.30 g, 73%) was also isolated as red solid. Mp: 180–183 °C, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.97–6.87 (m, 14H, Ar–H), 6.37 (s, 1H, NH), 5.70 (s, 1H, CH), 3.29–3.08 (m, 2H, CH_2), 1.34–1.27 (m, 2H, CH_2), 1.09–1.03 (m, 2H, CH_2), 0.75–0.71 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 100 MHz): δ 169.7, 153.2, 143.2, 140.4, 135.1, 132.9, 130.0, 129.6, 129.1, 128.5, 126.3, 126.1, 125.8, 123.0, 122.4, 122.2, 118.6, 108.5, 59.4, 39.3, 31.1, 19.7, 13.5. MS *m*/*z*: 407 (M^+, 2), 307 (100). Anal. Calcd for $C_{27}H_{25}N_3O$: C, 79.58; H, 6.18; N, 10.31. Found: C, 79.86; H, 6.03; N, 10.53.

*4.4.3. 6-(4-Chlorobenzylidene)-N-tert-butyl-6,12-dihydro indolo[2,1-b]quinazoline-12-carboxamide (**4c**).* Operation as above with 3,4-dihydroquinazoline **3f′** (0.52 g, 1 mmol), compound **4c** (0.27 g, 62%) was also isolated as red solid. Mp: 235–237 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.99–6.88 (m, 13H, Ar–H), 5.59 (s, 1H, NH), 5.54 (s, 1H, CH), 1.20 (s, 9H, $3CH_3$). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 168.8, 153.1, 143.5, 140.3, 134.9, 133.8, 131.2, 130.5, 130.2, 129.6, 129.2, 128.9, 126.5, 126.1, 126.0, 123.0, 122.2, 119.0, 108.8, 60.3, 51.7, 28.3. MS *m*/*z*: 441 (M^+, 2), 341 (100), 305 (7). Anal. Calcd for $C_{27}H_{24}ClN_3O$: C, 73.38; H, 5.47; N, 9.51. Found: C, 73.70; H, 5.24; N, 9.59.

*4.4.4. 6-(4-Chlorobenzylidene)-N-butyl-6,12-dihydroindolo [2,1-b] quinazoline-12-carboxamide (**4d**).* Operation as above with 3,4-dihydroquinazoline **3g′** (0.52 g, 1 mmol), compound **4d** (0.34 g, 77%) was also isolated as red solid. Mp: 226–227 °C, ^{1}H NMR ($CDCl_3$, 600 MHz): δ 7.92–6.88 (m, 13H, Ar–H), 6.14 (s, 1H, NH), 5.69 (s, 1H, CH), 3.24–3.09 (m, 2H, CH_2), 1.30–1.26 (m, 2H, CH_2), 1.07–1.02 (m, 2H, CH_2), 0.75–0.70 (m, 3H, CH_3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 169.5, 153.0, 143.3, 140.3, 135.0, 133.7, 131.3, 130.5, 130.3, 129.6, 129.1, 128.9, 126.4, 126.3, 126.0, 123.0, 122.2, 122.2, 118.7, 108.7, 59.3, 39.4, 31.1, 19.6, 13.5. MS *m*/*z*: 441 (M^+, 1), 341 (100), 305 (7). Anal. Calcd for $C_{27}H_{24}ClN_3O$: C, 73.38; H, 5.47; N, 9.51. Found: C, 73.68; H, 5.33; N, 9.68.

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Supplementary data

Supplementary data associated with this article can be found in online version at doi:10.1016/j.tet.2011.03.056. These data include MOL files and InChIKeys of the most important compounds described in this article.

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