Synthesis of Novel 2-Alkoxy(aralkoxy)-4*H*-[1,2,4]triazolo[1,5-*a*] quinazolin-5-ones Starting with Dialkyl-*N*-Cyanoimidocarbonates

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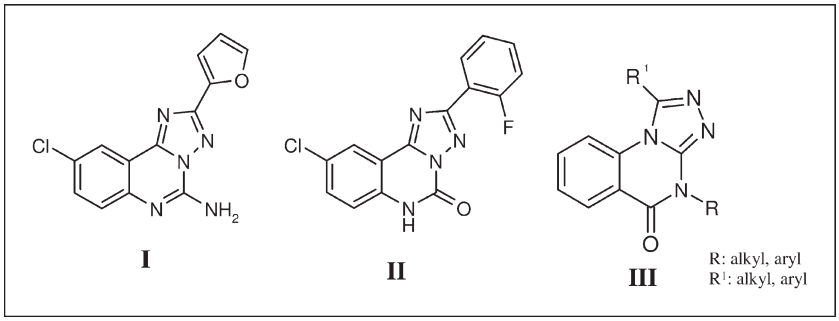

A novel series of 2-alkoxy(aralkoxy)-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolines were synthesized employing *N*-cyanoimidocarbonates and 2-hydrazinobenzoic acid as building blocks. Chemical transformation of the inherent lactam moiety in the targeted 2-alkoxy(aralkoxy)[1,2,4]triazolo[1,5-*a*]quinazolines was offered access to a variety of derivatives.

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INTRODUCTION

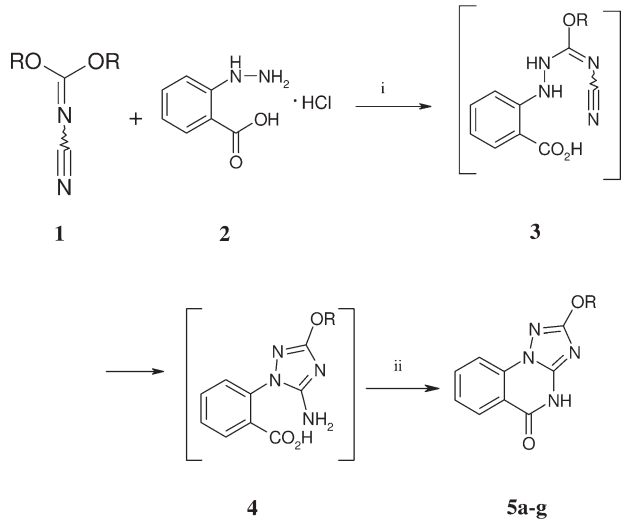
Compounds with 1,2,4-triazoloquinazoline moiety have been shown to exhibit diverse biological activities. For example, the novel compound **I** is effective adenosine antagonist whereas the related compound **II** was found to be benzodiazepine receptor antagonist [1–4]. The recently reported 1,2,4-triazoloquinazolines of type **III** were also found to exhibit promising antihistaminic activity against histamine induced bronchospasm and showed negligible sedation compared to chlorpheniramine maleate and could therefore serve as lead molecules for further modification to obtain a clinically useful class of non-sedative antihistamines [5,6]. In view of the beforementioned biological activities of diverse triazoloquinazolines and continuation of our ongoing studies dealing with the chemistry of *N*-cyanoimidocarbonates and their precursors, we wish to report herein the results of our study of cyclocondensation of *N*-cyanoimidocarbonates with hydrazinobenzoic acid to give 2-alkoxy(aralkoxy)[1,2,4]triazolo[1,5-*a*]quinazolin-5-ones.

RESULTS AND DISCUSSION

The preparation of several dialkyl *N*-cyanoimidocarbonates **1** from equimolar amounts of cyanogen bromide and the corresponding alcohol according to an established literature procedure was reported [7]. Based on the high reactivity of *N*-cyanoimidocarbonates towards hydrazines to produce 1,2,4-triazole derivatives [8–10], analogously, reaction of **1** with **2** in ethanol under ice cooling in the presence of triethylamine provided the intermediate 1,2,4-triazole derivative **4**, which upon treatment with hydrochloric acid (36%) produced the 2-alkoxy(aralkoxy)[1,2,4]triazolo[1,5-*a*]quinazolin-5-ones compounds **5a–g** in 40–60% yield (Scheme 1) [11]. The structure of the novel [1,2,4]triazolo[1,5-*a*]quinazolin-5-ones **5a–g** was confirmed by IR, ^{1}H NMR, ^{13}C NMR spectra, and microanalysis (see experimental). The IR spectra of compounds **5a–g** are characterized by a strong (C=O)-stretching band at 1685–1705 cm^{-1}.

Alkylation of lactams with alkyl halides may give rise to N- or/and O-alkylated products, the outcome of the reaction being dependent on the pH of the reaction, temperature, the nature of solvents, and the reactivity of the alkylating agents [12–14]. Accordingly, when the [1,2,4]triazolo[1,5-*a*]quinazolin-5-ones **5a,b** were allowed to react with alkyl halides in a molar ratio of 1:1.5 in absolute dimethyl formamide at room temperature in the presence of potassium carbonate, the corresponding 4-alkyl[1,2,4]triazolo[1,5-*a*]quinazolin-5-ones **6a–h** resulted in 62–87% yield [15]. Under these conditions, formation of the

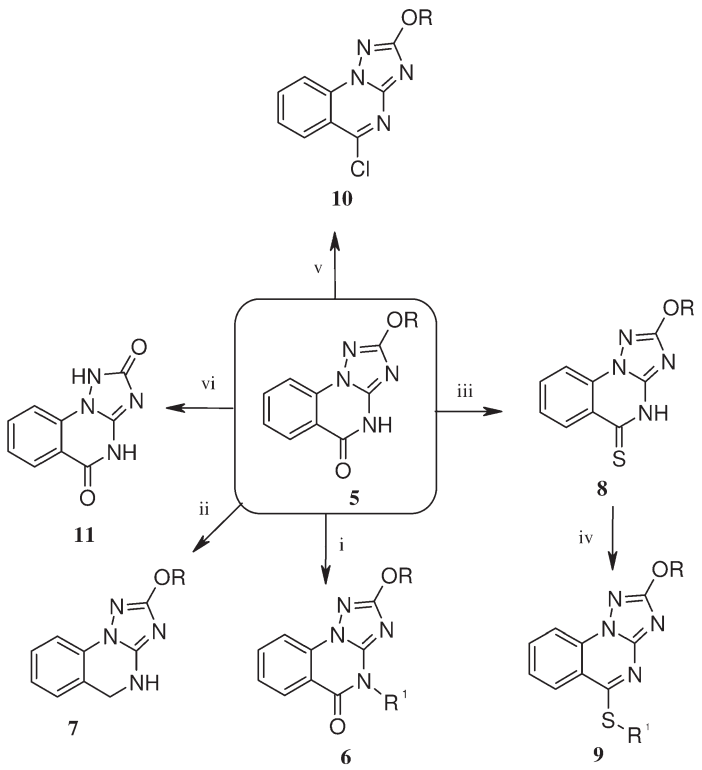
Scheme 1



i : Et_3N, EtOH ii : conc. HCl, 80 °C

isomeric lactim ethers was not observed (Scheme 2). The products **6a–h** were obtained as colored solid compounds and their IR spectra display a strong (C=O)-absorption band at 1670–1685 cm^{-1}. Treatment of compounds **5a,b,e,f,g** with lithium aluminum hydride in absolute tetrahydrofuran at room temperature furnished the aimed 4,5-dihydro[1,2,4]triazolo[1,5-*a*] quinazolines **7a–e** in 45–70% yield [16]. The compounds **7a–e** were obtained after column chromatography as colorless solids, and their structure was verified by elemental analyses and spectral (NMR, MS, and IR) data. The IR revealed the disappearance of a (C=O) absorption band at 1685–1705 cm^{-1} (previously found in compound **5**) confirmed the formation of the products **7**. When equimolar amounts of [1,2,4]triazolo[1,5-*a*] quinazolin-5-ones **5a,b,e,f,g** and phosphorus pentasulfide were allowed to react in absolute pyridine under reflux for 2 h, the targeted 2-alkoxy(aralkoxy)-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-thiones **8a–e** could be isolated in excellent yields of 85–97% as yellow solids [17]. In the IR spectra, the compounds **8a–e** displayed a weak absorption band of a (C=S) around 1244–

Scheme 2



R, R^1 : alky l, aralkyl, aryl

i : alkyl halides, DMF ii : $LiAlH_4$, THF iii : P_2S_5, pyridine iv : alkyl halides, $NaOH/H_2O$
v : $POCl_3$, benzene or $C_2O_2Cl_2$, trichloroethane vi : Pd/C -H_2, THF

1257 cm^{-1}, and the ^{13}C NMR spectra are characterized by a (C=S) resonance at 184.9–185.6 ppm. Reaction of the [1,2,4]triazolo[1,5-*a*]quinazolin-5-thiones **8a,d** with different alkyl halides in aqueous 0.5 *M* sodium hydroxide solution afforded smoothly the expected thioethers **9a–d** in 58–73% yield [18]. Conversion of [1,2,4]triazoloquinazolin-5-ones **5** into 5-chloro-[1,2,4]triazolo[1,5-*a*] quinazolines **10** has been successfully achieved by chlorination with either oxalyl chloride in boiling 1,1,2-trichloroethane for 19 h [10] or with phosphorus oxychloride in boiling benzene for 2 h, followed by titration with a saturated aqueous solution of potassium carbonate [19]. Although both methods gave acceptable yields, the reaction of **5** with phosphorus oxychloride is more advantageous with regard to short reaction time and higher yields. The formation of **10** was accompanied by the gradual disappearance of the characteristic (C=O) band of **5** at 1685–1705 cm^{-1}. Hydrogenolysis of **5f** on Pd/C in tetrahydrofuran cleanly afforded 1,2,4,5-tetrahydro[1,2,4]triazolo[1,5-*a*]quinazolin-2,5-dione **11** as a colorless solid in excellent yield of 95% and its structure was followed unambiguously from IR spectrum which showed two (C=O) absorption bands at 1707 and 1686 cm^{-1}.

EXPERIMENTAL

Melting points were determined on open glass capillaries using a Mettler FP 62 apparatus and are uncorrected. Elemental analyses were carried out with a Heraeus CHN-O-Rapid Instrument. The IR (KBr) spectra were recorded on a Shimadzu FTIR 8300. ^1H NMR (400.1 MHz) and ^{13}C NMR spectra were recorded on a Bruker AMX 400 spectrometer and chemical shifts are giving in a (ppm) downfield from tetramethylsilane (TMS) as an internal standard, DMSO is using as solvent. Mass spectra were recorded on a Finnigan MAT 311A and on a VG 70-250S (VG Analytical) instrument. Follow-up of the reactions and checking the purity of compounds was made by TLC on DC-Mikrokarten polygram SIL G/UV_{254}, from the Macherey-Nagel Firm, Duren Thickness: 0.25 m. Column chromatography was conducted on silica gel (ICN Silica 100–200, active 60 Å).

2-Alkoxy(aralkoxy)-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-ones (5a–g). General procedure. 10 mmol of 2-hydrazinobenzoic acid **2** was added portion wise to a stirred solution of **1** (10 mmol) in EtOH (20 mL) at 0°C. Afterwards, triethylamine (30 mmol) was added drop-wise over a period of 30 min. After the addition was complete, the reaction mixture was left to stir overnight at room temperature. Acidification of the mixture was performed by conc. HCl under ice cooling followed by refluxing for 1–3 h. After cooling, the mixture was poured into ice/water, the resulting solid was filtered, washed with water and dried. Recrystallization from THF gave analytically pure colored **5a–g**.

2-Methoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5a). Yellow solid; (yield: 60%), m.p. 228°C (THF). IR (KBr) 1685 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 3.99 (s, 3H, CH_3), 7.48–8.15 (m, 4H, ArH), 13.15 (s, 1H, NH), ^{13}C NMR (DMSO-d_6): δ_C 57.16 (CH_3), 114.25, 116.83, 125.50, 128.58, 135.72, 136.12 (C_{Arom}), 147.87 (C-guanidine), 159.93 (C=O), 168.26 (C-isourea). ms: *m/z* (%): 216 (M^+, 100), 201 (M^+-methyl, 15), 187 (30), 145 (27), 104 (17). Anal. Calcd. for $C_{10}H_8N_4O_2$ (216.20): C, 55.56; H, 3.73; N, 25.91. Found: C, 55.38; H, 3.83; N, 25.99.

2-Ethoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5b). Yellow solid; (yield: 60%), m.p. 244°C (THF). IR (KBr) 1689 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 1.38 (t, J = 7.07 Hz, 3H, CH_3), 4.35 (q, J = 14.13 Hz, 2H, CH_2), 7.47–8.16 (m, 4H, ArH), 13.01 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 14.86 (CH_3), 65.64 (CH_2), 114.26, 116.79, 125.50, 128.59, 135.75, 136.12 (C_{Arom}), 147.74 (C-guanidine), 159.92 (C=O), 167.56 (C-isourea). ms: *m/z* (%): 230 (M^+, 95), 201 (M^+-ethyl, 90), 187 (7), 160 (4), 134 (100), 104 (25). Anal. Calcd. for $C_{11}H_{10}N_4O_2$ (230.23): C, 57.39; H, 4.38; N, 24.34. Found: C, 57.18; H, 4.49; N, 24.40.

2-Isopropoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5c). Pale brown solid; (yield: 40%), m.p. 221°C (THF). IR (KBr) 1691 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 1.37 (d, J = 7.35 Hz, 6H, —CH(CH_3)$_2$), 4.95–5.02 (m, 1H, C*H*(CH_3)$_2$), 6.96–8.16 (m, 4H, ArH), 11.34 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 22.16 (CH_3), 73.10 (CH), 114.25, 116.83, 125.46, 128.61, 135.73, 136.64 (C_{Arom}), 147.60 (C-guanidine), 158.85 (C=O), 167.47 (C-isourea). ms: *m/z* (%): 244 (M^+, 35), 202 (M^+-isopropyl, 100), 160 (7), 134 (80), 105 (40), 77 (30). Anal. Calcd. for $C_{12}H_{12}N_4O_2$ (244.26): C, 59.01; H, 4.95; N, 22.94. Found: C, 59.37; H, 4.88; N, 22.55.

2-Pentyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5d). Yellow solid; (yield: 50%), m.p. 234°C (THF). IR (KBr) 1692 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 0.91 (t, J = 7.41 Hz, 3H, —$CH_2CH_2CH_2CH_2CH_3$), 1.33–1.42 (m, 4H, —$CH_2CH_2CH_2CH_2CH_3$), 1.73–1.79 (m, 2H, —$CH_2CH_2CH_2CH_2CH_3$), 4.30 (t, J = 7.60 Hz, 2H, —$CH_2CH_2CH_2CH_2CH_3$), 7.45–8.16 (m, 4H, ArH), 12.98 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 14.27 (—$CH_2CH_2CH_2CH_2CH_3$), 22.14 (—$CH_2CH_2CH_2CH_2CH_3$), 27.63 (—$CH_2CH_2CH_2CH_2CH_3$), 28.47 ($CH_2CH_2CH_2CH_2CH_3$), 69.74 (—$CH_2CH_2CH_2CH_2CH_3$), 114.21, 116.80, 125.45, 128.6, 135.72, 136.11 (C_{Arom}), 147.74 (C-guanidine), 159.91 (C=O), 167.70 (C-isourea). ms: *m/z* (%): 272 (M^+, 15), 202 (M^+-pentyl, 100), 160 (5), 134 (60), 104 (15), 43 (67). Anal. Calcd. for $C_{14}H_{16}N_4O_2$ (272.31): C, 61.75; H, 5.92; N, 20.57. Found: C, 61.68; H, 6.02; N, 20.52.

2-Allyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5e). Yellow solid; (yield: 55%), m.p. 215°C (THF). IR (KBr) 1696 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 4.86 (d, J = 5.68 Hz, 2H, —CH_2=$CHCH_2$), 5.42–5.65 (m, 2H, —CH_2=$CHCH_2$), 6.05–6.15 (m, 1H, —CH_2=$CHCH_2$), 7.48–8.17 (m, 4H, ArH), 13.01 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 69.60 (—CH_2=$CHCH_2$), 113.82 (—CH_2=$CHCH_2$), 116.44, 118.25, 125.16, 128.13 (C_{Arom}), 134.11 (—CH_2=$CHCH_2$), 135.30, 135.62 (C_{Arom}), 147.30 (C-guanidine), 159.45 (C=O), 166.92 (C-isourea). ms: *m/z* (%): 242 (M^+, 100), 202 (M^+-allyl, 23), 187 (9), 160 (5), 134 (18), 104 (37), 41 (40). Anal. Calcd. for $C_{12}H_{10}N_4O_2$ (242.24): C, 59.50; H, 4.16; N, 23.13. Found: C, 59.20; H, 4.42; N, 22.85.

2-Benzyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5f). White solid; (yield: 58%), m.p. 258°C (THF). IR (KBr) 1701 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 5.39 (s, 2H, CH_2), 7.37–8.16 (m, 9H, ArH), 13.04 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 71.18 (CH_2), 114.34, 116.81, 125.53, 127.74, 128.03, 128.85, 135.75, 136.11, 136.77 (C_{Arom}), 147.11 (C-guanidine), 160.40 (C=O), 167.17 (C-isourea). ms: *m/z* (%): 292 (M^+,

35), 250 (6), 248 (17), 201(12), 91(100). Anal. Calcd. for $C_{16}H_{12}N_4O_2$ (292.30): C, 65.75; H, 4.14; N, 19.17. Found: C, 65.39; H, 4.04; N,19.06.

2-Phenethyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (5g). White solid; (yield: 56%), m.p. 227°C (THF). IR (KBr) 1705 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 3.20 (t, J = 7.50 Hz, 2H, —OCH_2CH_2Ph), 4.50 (t, J = 7.51 Hz, 2H, —OCH_2CH_2Ph), 7.20–8.19 (m, 9H, ArH), 13.75 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 34.94 (—OCH_2CH_2Ph), 70.23 (—OCH_2CH_2Ph), 116.81, 114.32, 126.80, 125.51, 128.59, 128.74, 129.37, 136.14, 138.33 (C_{Arom}), 147.72 (C-guanidine), 159.91 (C=O), 167.57 (C-isourea). ms: *m/z* (%): 306 (M^+, 53), 292 (M^+—CH_2, 15), 202 (M^+-phenethyl, 80), 134 (15), 105 (100), 91 (28). Anal. Calcd. for $C_{17}H_{14}N_4O_2$ (306.33): C, 66.66; H, 4.61; N, 18.29. Found: C, 66.32; H, 4.94; N, 18.33.

2-Alkoxy(aralkoxy)-4-alkyl(aralkyl)-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-ones (6a–h). General procedure. To a solution of **5a,b** (1 mmol) in DMF (5 mL) was added potassium carbonate (1.2 mmol) portion wise over a period of 10 min at room temperature. After stirring for 20 min, the appropriate alkyl halide (1.5 mmol) was added drop wise, and the reaction mixture was stirred for 18 h at room temperature. The mixture was poured into ice/water, the precipitate was filtered off, washed with water and dried. Analytically pure products **6a–h** were obtained after recrystallization from THF.

4-(4-Bromobenzyl)-2-methoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6a). Yellow solid; (yield: 83%), m.p. 194°C (THF). IR (KBr) 1676 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 4.12 (s, 3H, CH_3), 5.37 (s, 2H, CH_2), 7.50–8.33 (m, 8H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 46.21 (CH_2), 57.23 (CH_3), 114.28, 116.13, 118.45, 121.03, 125.88, 128.96, 130.50, 131.68, 135.65, 136.02 (C_{Arom}), 148.80 (C-guanidine), 159.02 (C=O), 167.84 (C-isourea). ms: *m/z* (%): 385 (M^+, 100), 355 (10), 305 (13), 201 (70), 171 (90), 90 (30). Anal. Calcd. for $C_{17}H_{13}BrN_4O_2$ (385.22): C, 53.01; H, 3.40; N, 14.54. Found: C, 52.78; H, 3.47; N, 14.43.

2-Methoxy-4-phenethyl-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6b). White solid; (yield: 85%), m.p. 155°C (THF). IR (KBr) 1675 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 3.04 (t, J = 7.54 Hz, 2H, —NCH_2CH_2Ph), 4.02 (s, 3H, OCH_3), 4.31 (t, J = 7.51 Hz, 2H, —NCH_2CH_2Ph), 7.22–8.20 (m, 9H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 33.08 (—NCH_2CH_2Ph), 44.66 (—NCH_2CH_2Ph), 57.21(OCH_3), 114.18, 116.11, 125.82, 126.88, 128.87, 135.42, 135.84, 138.39 (C_{Arom}), 148.59 (C-guanidine), 158.76 (C=O), 167.97 (C-isourea). ms: *m/z* (%): 320 (M^+, 90), 229 (70), 216 (100), 188 (15), 160 (11), 145 (12), 104 (40). Anal. Calcd. for $C_{18}H_{16}N_4O_2$ (320.35): C, 67.49; H, 5.03; N, 17.49. Found: C, 67.49; H, 5.15; N, 17.18.

4-Benzyl-2-methoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6c). Yellow solid; (yield: 82%), m.p. 134°C (THF). IR (KBr) 1678 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 4.13 (s, 3H, CH_3), 5.34 (s, 2H, CH_2), 7.50–8.30 (m, 9H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 46.75 (CH_2), 57.21(CH_3), 114.24, 116.86, 117.79, 125.80, 128.89, 129.11, 131.66, 132.45, 135.86, 135.52 (C_{Arom}), 148.85 (C-guanidine), 158.96 (C=O), 167.86 (C-isourea). ms: *m/z* (%): 306 (M^+, 100), 277 (8), 262 (5), 201 (22), 91 (55). Anal. Calcd. for $C_{17}H_{14}N_4O_2$ (306.33): C, 66.66; H, 4.61; N, 18.29. Found: C, 66.46; H, 4.69; N, 17.93.

4-Ethyl-2-methoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6d). White solid; (yield: 62%), m.p. 135°C (THF). IR (KBr) 1672 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 1.37 (t, J = 7.04 Hz, 3H, NCH_2CH_3), 4.24 (q, J = 14.31 Hz, 2H, NCH_2CH_3), 4.11 (s, 3H, OCH_3), 7.58–8.29 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 12.81 (NCH_2CH_3), 52.34 (NCH_2CH_3), 57.18 (OCH_3), 114.13, 116.22, 125.71, 128.79, 135.20, 135.42 (C_{Arom}), 148.49 (C-guanidine), 158.64 (C=O), 167.99 (C-isourea). ms: *m/z* (%): 244 (M^+, 67), 230 (M^+-methyl, 35), 216 (87), 91 (22). Anal. Calcd. for $C_{12}H_{12}N_4O_2$ (244.26): C, 59.01; H, 4.95; N, 22.94. Found: C, 58.79; H, 5.04; N, 22.58.

4-Allyl-2-methoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6e). Yellow solid; (yield: 82%), m.p. 132°C (THF). IR (KBr) 1680 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 4.13 (s, 3H, CH_3), 4.83 (d, J = 4.60 Hz, 2H, —CH_2=$CHCH_2$), 5.30–5.42 (m, 2H, —CH_2=$CHCH_2$), 6.04–6.30 (m, 1H, —CH_2=$CHCH_2$), 7.46–8.31 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 45.53 (—CH_2=$CHCH_2$), 57.14 (CH_3), 114.47 (C_{Arom}), 116.10 (—CH_2=$CHCH_2$), 117.18, 125.83, 128.82, 131.66 (C_{Arom}), 135.42 (—CH_2=$CHCH_2$), 135.86 (C_{Arom}), 148.79 (C-guanidine), 157.81 (C=O), 168.43 (C-isourea). ms: *m/z* (%): 256 (M^+, 70), 216 (100), 187 (11), 160 (5), 104 (42), 41 (53). Anal. Calcd. for $C_{13}H_{12}N_4O_2$ (256.27): C, 60.93; H, 4.72; N, 21.86. Found: C, 60.79; H, 5.01; N, 21.76.

2-Ethoxy-4-prop-2-ynyl-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6f). White solid; (yield: 87%), m.p. 147°C (THF). IR (KBr) 1685 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 1.64 (t, J = 7.26 Hz, 3H, OCH_2CH_3), 3.58 (s, 1H, $HC{\equiv}CCH_2$), 4.64 (q, J = 14.32 Hz, 2H, OCH_2CH_3), 5.10 (s, 2H, $HC{\equiv}CCH_2$), 7.77–8.48 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 14.18 (CH_3), 32.99 ($HC{\equiv}CCH_2$), 65.77 (OCH_2), 74.88 ($HC{\equiv}CCH_2$), 78.13 ($HC{\equiv}CCH_2$), 122.45, 125.83, 129.68, 131.61(C_{Arom}), 148.46 (C-guanidine), 163.58 (C=O), 167.53 (C-isourea). ms: *m/z* (%): 268 (M^+, 78), 253 (12), 239 (10), 197 (80), 169 (12), 145 (13), 107 (25), 39 (100). Anal. Calcd. for $C_{14}H_{12}N_4O_2$ (268.28): C, 62.68; H, 4.51; N, 20.88. Found: C, 62.45; H, 4.75; N, 21.08.

4-Cyclopropylmethyl-2-ethoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6g). White solid; (yield: 70%), m.p. 116°C (THF). IR (KBr) 1677 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 0.63–0.69 (m, 5H, —$CH_2C_3H_5$), 1.56 (t, J = 7.12 Hz, 3H, CH_3), 4.17 (s, 2H, —$CH_2C_3H_5$), 4.47 (q, J = 14.34 Hz, 2H, OCH_2), 7.68–8.38 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 3.93 (2C-cyclopropyl), 9.81 (C-cyclopropyl), 14.85 (CH_3), 48.37 (—$CH_2C_3H_5$), 65.80 (OCH_2), 114.17, 116.23, 125.85, 128.93, 135.43, 135.97 (C_{Arom}), 148.78 (C-guanidine), 158.94 (C=O), 167.20 (C-isourea). ms: *m/z* (%): 284 (M^+, 76), 256 (13), 230 (28), 145 (34), 104 (11), 91 (88). Anal. Calcd. for $C_{15}H_{16}N_4O_2$ (284.32): C, 63.37; H, 5.67; N, 19.71. Found: C, 63.55; H, 5.74; N, 19.50.

4-(2,4-Dichlorobenzyl)-2-ethoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-one (6h). White solid; (yield: 73%), m.p. 203°C (THF). IR (KBr) 1676 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 1.63 (t, J =7.02 Hz, 3H, CH_3), 4.61 (q, J = 14.02 Hz, 2H, CH_2), 5.31 (s, 2H, CH_2Ph), 7.31–8.19 (m, 7H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 14.73 (CH_3), 44.96 (CH_2Ph), 65.80 (CH_2), 114.34, 116.1, 125.81, 127.83, 129.01, 129.70, 131.62, 132.40, 133.15, 135.83, 136.17 (C_{Arom}), 148.63 (C-guanidine), 159.27 (C=O), 167.19 (C-isourea). ms: *m/z* (%): 390 (M^++1, 18), 389 (M^+, 54), 230 (23), 187 (5), 143 (12), 90 (58). Anal. Calcd. for $C_{18}H_{14}Cl_2N_4O_2$ (389.24): C, 55.54; H, 3.63; N, 14.39. Found: C, 55.17; H, 3.75; N, 14.37.

2-Alkoxy(aralkoxy)-4,5-dihydro[1,2,4]triazolo[1,5-*a*]quinazolines (7a–e). General procedure. A solution of **5** (1 mmol) in dry THF (5 mL) was added drop wise to a stirred

suspension of $LiAlH_4$ (3 mmol) in dry THF (10 mL). After stirring at room temperature for 3 h, water (0.4 mL) was added carefully and the mixture was stirred for an additional 30 min. The reaction mixture was filtered and the solvent removed under reduced pressure, the residue was dissolved in THF and passed through a short column chromatography, the solvent was removed under reduced pressure, and the obtained solid was recrystallized from EtOAc/n-hexane.

4,5-Dihydro-2-methoxy[1,2,4]triazolo[1,5-a]quinazoline (7a). White solid; (yield: 60%), m.p. 133°C (EtOAc-hexane). ^{1}H NMR (DMSO-d_6): δ_H 3.90 (s, 3H, CH_3), 4.20 (s, 2H, CH_2-quinazoline), 7.28–7.82 (m, 4H, ArH), 7.95 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 43,22 (*C*H_2-quinazoline), 56.35 (OCH_3), 112.72, 119.64, 124.50, 126.23, 130.75, 134.16 (C_{Arom}), 155.18 (C-guanidine), 165.29 (C-isourea). ms: *m/z* (%): 201 (M^+-1, 100), 186 (14), 143 (5), 129 (11), 89 (9). Anal. Calcd. for $C_{10}H_{10}N_4O$ (202.22): C, 59.40; H, 4.98; N, 27.71. Found: C, 59.15; H, 5.18; N, 27.38.

4,5-Dihydro-2-ethoxy[1,2,4]triazolo[1,5-a]quinazoline (7b). White solid; (yield: 61%), m.p. 142°C (EtOAc-hexane). ^{1}H NMR (DMSO-d_6): δ_H 1.31 (t, J = 7.21 Hz, 3H, CH_3), 4.24 (q, J = 14.40 Hz, 2H, CH_2), 4.49 (s, 2H, CH_2-quinazoline), 7.08–7.33 (m, 4H, ArH), 7.76 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 14.94 (CH_3), 42.38 (*C*H_2-quinazoline), 64.71 (OCH_2), 112.71, 119.70, 124.52, 126.79, 128.79, 134.14 (C_{Arom}), 154.99 (C-guanidine), 167.37 (C-isourea). ms: *m/z* (%): 216 (M^+, 100), 187 (8), 145 (13), 104 (11), 76 (18). Anal. Calcd. for $C_{11}H_{12}N_4O$ (216.24): C, 61.10; H, 5.59; N, 25.91. Found: C, 60.86; H, 5.57; N, 25.63.

2-Allyloxy-4,5-dihydro[1,2,4]triazolo[1,5-a]quinazoline (7c). White solid; (yield: 55%), m.p. 105°C (EtOAc-hexane). ^{1}H NMR (DMSO-d_6): δ_H 4.76 (d, J = 6.76 Hz, 2H, —CH_2=CHCH_2), 4.92 (s, 2H, CH_2-quinazoline), 5.32–5.43 (m, 2H, —CH_2=CHCH_2), 6.09–6.16 (m, 1H, —CH_2=C*H*CH_2), 7.48–8.10 (m, 4H, ArH), 8.25 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 69.63 (*C*H_2-quinazoline), 113.87 (—*C*H_2=CHCH_2), 116.45 (—CH_2=C*H*CH_2), 118.20, 119.24, 125.33, 128.12, 134.57 (C_{Arom}), 135.20 (—CH_2=*C*HCH_2), 135.52 (C_{Arom}), 159.37 (C-guanidine), 166.70 (C-isourea). ms: *m/z* (%): 228 (M^+, 100), 209 (3), 187 (85), 116 (25). Anal. Calcd. for $C_{12}H_{12}N_4O$ (228.26) : C, 63.15; H, 5.30; N, 24.55. Found: C, 63.43; H, 5.23; N, 24.42.

2-Benzyloxy-4,5-dihydro[1,2,4]triazolo[1,5-a]quinazoline (7d). White solid; (yield: 70%), m.p. 158°C (EtOAc-hexane). ^{1}H NMR (DMSO-d_6): δ_H 4.50 (s, 2H, CH_2-quinazoline), 5.26 (s, 2H, OCH_2Ph), 7.11–7.46 (m, 9H, ArH), 7.81 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 69.94 (O*C*H_2Ph), 112.27, 119.23, 124.10, 126.30, 127.02, 127.95, 128.27, 128.95, 133.67, 136.40 (C_{Arom}), 154.55 (C-guanidine), 166.87 (C-isourea). ms: *m/z* (%): 278 (M^+, 100), 233 (7), 201 (6), 187 (18), 91 (100). Anal. Calcd. for $C_{16}H_{14}N_4O$ (278.32): C, 69.05; H, 5.07; N, 20.13. Found: C, 69.35; H, 5.10; N,19.83.

4,5-Dihydro-2-phenethyloxy[1,2,4]triazolo[1,5-a]quinazoline (7e). White solid; (yield: 64%), m.p. 119°C (EtOAc-hexane). ^{1}H NMR (DMSO-d_6): δ_H 3.04 (t, J = 7.40 Hz, 2H, OCH_2CH_2Ph), 4.39 (t, J = 7.51 Hz, 2H, OCH_2C*H*$_2$Ph), 4.48 (s, 2H, CH_2-quinazoline), 7.10–7.32 (m, 9H, ArH), 7.77 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 34.90 (OCH_2*C*H_2Ph), 68.95 (O*C*H_2CH$_2$Ph), 113.33, 119.27, 126.80, 128.51, 129.37, 135.74, 136.11, 138.33 (C_{Arom}), 154.90 (C-guanidine), 166.85 (C-isourea). ms: *m/z* (%): 292 (M^+, 39), 188 (100), 173 (5), 145 (6), 105 (35). Anal. Calcd. for $C_{17}H_{16}N_4O$ (292.34): C, 69.85; H, 5.52; N, 19.16. Found: C, 69.51; H, 5.56; N, 18.93.

2-Alkoxy(aralkoxy)-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-thiones (8a–e). General procedure. Compound **5** (1 mmol) was refluxed with phosphorus pentasulfide (1 mmol) in absolute pyridine (5 mL) for 2 h. Afterwards the reaction mixture was cooled and poured into ice/water, the yellow precipitate was separated by filtration and washed thoroughly with water. Recrystallization from aqueous DMF furnished analytically pure **8a–e**.

2-Methoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-thione (8a). Yellow solid; (yield: 85%), m.p. 230°C (DMF). IR (KBr) 1250 cm^{-1}. ^{1}H NMR (DMSO-d_6): δ_H 4.02 (s, 3H, CH_3), 7.52–7.96 (m, 4H, ArH), 14.72 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 56.84 (CH_3), 114.21, 122.43, 125.83, 131.77, 132.41, 135.88 (C_{Arom}), 149.59 (C-guanidine), 162.78 (C-isourea), 185.01(C=S). ms: *m/z* (%): 232 (M^+, 100), 216 (40), 203 (4), 175 (7), 120 (23), 102 (13). Anal. Calcd. for $C_{10}H_8N_4OS$ (232.27): C, 51.71; H, 3.47; N, 24.12; S, 13.80. Found: C, 51.99; H, 3.22; N, 4.52; S,13.65.

2-Ethoxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-thione (8b). Yellow solid; (yield: 92%), m.p. 226°C (DMF). IR (KBr) 1248 cm^{-1}. ^{1}H NMR (DMSO-d_6): δ_H 1.39 (t, J = 7.28 Hz, 3H, CH_3), 4.40 (q, J = 14.20 Hz, 2H, CH_2), 7.51–8.62 (m, 4H, ArH), 14.70 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 14.39 (CH_3), 65.46 (CH_2), 114.20, 122.90, 125.79, 131.39, 132.39 (C_{Arom}), 145.63 (C-guanidine), 162.12 (C-isourea), 184.94 (C=S). ms: *m/z* (%): 246 (M^+,100), 230 (10), 218 (45), 202 (8), 150 (42). Anal. Calcd. for $C_{11}H_{10}N_4OS$ (246.29): C, 53.64; H, 4.09; N, 22.75; S, 13.02. Found: C, 53.42; H, 3.87; N, 22.32; S, 13.17.

2-Allyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-thione (8c)? Yellow solid; (yield: 95%), m.p. 190°C (DMF). IR (KBr) 1244 cm^{-1}. ^{1}H NMR (DMSO-d_6): δ_H 4.85 (d, J = 6.36 Hz, 2H, —CH_2=CHCH_2), 5.31–5.46 (m, 2H, —CH_2=CHCH_2), 6.08–6.15 (m, 1H, —CH_2=C*H*CH_2), 7.48–8.62 (m, 4H, ArH), 14.72 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 69.92 (—*C*H_2=CHCH_2), 114.27 (C_{Arom}), 118.39 (—CH_2=CH*C*H_2), 122.53, 125.92, 128.21, 131.83 (C_{Arom}), 132.42 (—CH_2=*C*HCH$_2$), 135.92 (C_{Arom}), 145.75 (C-guanidine), 167.31 (C-isourea), 185.08 (C=S). ms: *m/z* (%): 258 (M^+, 94), 242 (25), 216 (20), 150 (11), 120 (15). Anal. Calcd. for $C_{12}H_{10}N_4OS$ (258.30): C, 55.80; H, 3.90; N, 21.69; S, 12.41. Found: C, 55.65; H, 3.97; N, 21.73; S, 12.18.

2-Benzyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-thione (8d). Yellow solid; (yield: 97%), m.p. 210°C (DMF). IR (KBr) 1253 cm^{-1}. ^{1}H NMR (DMSO-d_6): δ_H 5.42 (s, 2H, CH_2), 7.37–8.62 (m, 9H, ArH), 14.74 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 70.60 (CH_2), 114.24, 122.40, 125.37, 128.06, 128.10, 128.90, 131.72, 132.33, 135.38 (C_{Arom}), 145.90 (C-guanidine), 167.34 (C-isourea), 185.62 (C=S). ms: *m/z* (%): 308 (M^+, 25), 275 (10), 218 (100), 186 (13), 150 (45), 91 (60). Anal. Calcd. for $C_{16}H_{12}N_4OS$ (308.36): C, 62.32; H, 3.92; N, 18.17; S, 10.40. Found: C, 61.96; H, 4.05; N, 17.87; S,10.06.

2-Phenethyloxy-4H-[1,2,4]triazolo[1,5-a]quinazolin-5-thione (8e). Yellow solid; (yield: 89%), m.p. 221°C (DMF). IR (KBr) 1257 cm^{-1}. ^{1}H NMR (DMSO-d_6): δ_H 3.11 (t, J = 6.35 Hz, 2H, OCH_2C*H*$_2$Ph), 4.55 (t, J = 6.63 Hz, 2H, OC*H*$_2$CH$_2$Ph), 7.24–8.61 (m, 9H, ArH), 14.70 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 34.45 (OCH_2*C*H_2Ph), 69.72 (O*C*H_2CH$_2$Ph),

114.23, 122.42, 125.00, 125.82, 126.30, 128.28, 128.85, 131.76, 135.83, 137.80 (C_{Arom}), 145.63 (C-guanidine), 165.20 (C-isourea), 184.91 (C=S). ms: *m/z* (%): 322 (M^+, 78), 218 (31), 187 (11) 134 (15), 91 (28). Anal. Calcd. for $C_{17}H_{14}N_4OS$ (322.39): C, 63.34; H, 4.38; N, 17.38; S, 9.95. Found: C, 62.95; H, 4.65; N, 17.02; S,10.03.

2-Alkoxy(aralkoxy)-5-alkyl(aralkyl)sulfanyl[1,2,4]triazolo[1,5-*a*]quina-zolines (9a–d). General procedure. Compound **8a,d** (1 mmol) was dissolved in aqueous 0.5 *M* sodium hydroxide solution (10 mL), alkyl halide (1.5 mmol) was added drop wise over a period 2 min, the mixture was left to stirr for 5–20 min at room temperature, and the obtained solid was separated by filtration, washed thoroughly with water and dried. Recrystallization of the crude products from EtOH afforded **9a–d** as colored pure solids.

5-Allylsulfanyl-2-methoxy[1,2,4]triazolo[1,5-a]quinazoline (9a). White solid; (yield: 70%), m.p. 123°C (EtOH). ^{1}H NMR (DMSO-d_6): δ_H 4.07 (s, 3H, CH_3), 4.09 (d, J = 10.12 Hz, 2H, —C*H*$_2$=CHCH$_2$), 5.22–5.50 (m, 2H, —CH$_2$=CHC*H*$_2$) 5.99–6.09 (m, 1H, —CH$_2$=C*H*CH$_2$), 7.63–8.22 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 32.03 (—*C*H$_2$=CHCH$_2$), 56.67 (CH_3), 114.81, 116.99 (C_{Arom}), 118.94 (—CH$_2$=CH*C*H$_2$), 125.54 (C_{Arom}), 132.59 (—CH$_2$=*C*HCH$_2$), 133.54 (C_{Arom}), 135.57 (C-guanidine), 165.88 (C-thioether), 169.24 (C-isourea). ms: *m/z* (%): 272 (M^+, 31), 258 (5), 200 (11), 145 (20), 104 (34). Anal. Calcd. for $C_{13}H_{12}N_4OS$ (272.33): C, 57.34; H, 4.44; N, 20.57; S, 11.77. Found: C, 57.70; H, 4.32; N, 20.53; S, 11.48.

2-Benzyloxy-5-methylsulfanyl[1,2,4]triazolo[1,5-a]quinazoline (9b). Yellow solid; (yield: 73%), m.p. 185°C (EtOH). ^{1}H NMR (DMSO-d_6): δ_H 2.45 (s, 3H, CH_3), 5.34 (s, 2H, CH_2), 7.20–8.86 (m, 9H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 12.69 (CH_3), 69.78 (CH_2), 113.19, 114.79, 123.25, 125.45, 125.78, 127.78, 128.26, 131.87, 132.14, 135.45 (C_{Arom}), 136.69 (C-guanidine), 152.67 (C-thioether), 168.11 (C-isourea). ms: *m/z* (%): 322 (M^+, 24), 232 (13), 201 (22), 104 (43), 91 (25). Anal. Calcd. for $C_{17}H_{14}N_4OS$ (322.39): C, 63.34; H, 4.38; N, 17.38; S, 9.95. Found: C, 63.02; H, 4.32; N, 17.24; S, 10.01.

2-Methoxy-5-prop-2-ynylsulfanyl[1,2,4]triazolo[1,5-a]quinazoline (9c). Pale brown solid; (yield: 61%), m.p. 190°C (EtOH). ^{1}H NMR (DMSO-d_6): δ_H 3.23 (s, 1H, *H*C≡CCH$_2$-), 4.20 (s, 3H, CH_3), 4.45 (s, 2H, HC≡CC*H*$_2$-), 7.60–8.34 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 18.45 (HC≡C*C*H$_2$-), 57.11(CH_3), 79.57 (HC≡*C*CH$_2$), 80.74 (HC≡*C*CH$_2$-), 115.23, 117.04, 125.64, 125.79, 126.28, 133.91, 136.16 (C_{Arom}), 151.02 (C-guanidine), 165.08 (C-thioether), 169.60 (C-isourea). ms: *m/z* (%): 270 (M^+, 100), 201 (8), 187 (13), 104 (4), 91 (21). Anal. Calcd. for $C_{13}H_{10}N_4OS$ (270.31): C, 57.76; H, 3.73; N, 20.73; S, 11.86. Found: C, 57.61; H, 3.90; N, 21.08; S, 11.55.

2-Methoxy-5-methylsulfanyl[1,2,4]triazolo[1,5-a]quinazoline (9d). Yellow solid; (yield: 60%), m.p. 146°C (EtOH). ^{1}H NMR (DMSO-d_6): δ_H 2.73 (s, 3H, SCH_3), 4.07 (s, 3H, OCH_3), 7.66–8.22 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 31.45 (SCH_3), 56.64 (OCH_3), 114.21, 122.27, 125.32, 131.81, 132.52, 135.88 (C_{Arom}), 140.47 (C-guanidine), 159.22 (C-thioether), 166.20 (C-isourea). ms: *m/z* (%): 246 (M^+, 37), 232 (54), 145 (6), 120 (8), 102 (31). Anal. Calcd. for $C_{11}H_{10}N_4OS$ (246.29): C, 53.64; H, 4.09; N, 22.75; S, 13.02. Found: C, 53.91; H, 3.86; N, 22.45; S, 12.81.

2-Alkoxy(aralkoxy)-5-chloro[1,2,4]triazolo[1,5-*a*]quinazolines (10a–e). General procedure. Method-A: Compound **5** (2 mmol) was refluxed with oxalyl chloride (6 mmol) in 1,1,2-trichloroethane (12 mL) for 19 h at 105°C. The solution was cooled and MeOH (0.2 mL) was added drop wise, the obtained solid was filtered, washed with hexane, dried and recrystallized from THF-hexane.

Method-B: Compound **5** (1 mmol) was refluxed with Phosphorus oxychloride (1 mL) in benzene (7 mL) for 2 h. The solvent was evaporated and the residue was treated with saturated aqueous solution of potassium carbonate. The solid was filtered, washed thoroughly with water, dried and recrystallized from THF-hexane.

5-Chloro-2-methoxy[1,2,4]triazolo[1,5-a]quinazoline (10a). White solid; (yield: 80%), m.p. 148°C (THF-hexane). ^{1}H NMR (DMSO-d_6): δH 3.99 (s, 3H, CH_3), 7.48–8.15 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 57.16 (CH_3), 114.20, 116.83, 125.51, 128.57, 135.74, 136.19 (C_{Arom}), 141.11 (C-guanidine), 159.90 (C—Cl), 168.26 (C-isourea). ms: *m/z* (%): 234 (M^+, 100), 216 (50), 169 (40), 128 (10), 102 (15). Anal. Calcd. for $C_{10}H_7ClN_4O$ (234.65): C, 51.19; H, 3.01; N, 23.88. Found: C, 51.12; H, 3.18; N, 23.98.

5-Chloro-2-ethoxy[1,2,4]triazolo[1,5-a]quinazoline (10b). White solid; (yield: 89%), m.p. 134°C (THF-hexane). ^{1}H NMR (DMSO-d_6): δ_H 1.37 (t, J = 7.07 Hz, 3H, CH_3), 4.34 (q, J = 14.13 Hz, 2H, CH_2), 7.49–8.15 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 14.83 (CH_3), 65.67 (CH_2), 114.17, 116.75, 125.52, 128.80, 135.18, 136.13 (C_{Arom}), 142.20 (C-guanidine), 159.92 (C—Cl), 167.38 (C-isourea). ms: *m/z* (%): 248 (M^+, 78), 220 (32), 201 (4), 128 (12), 91 (50). Anal. Calcd. for $C_{11}H_9ClN_4O$ (248.67): C, 53.13; H, 3.65; N, 22.53. Found: C, 53.33; H, 3.98; N, 22.33.

5-Chloro-2-pentyloxy[1,2,4]triazolo[1,5-a]quinazoline (10c). Pale brown solid; (yield: 81%), m.p. 110°C (THF-hexane). ^{1}H NMR (DMSO-d_6): δ_H 0.96 (t, J = 7.45 Hz, 3H, —CH$_2$CH$_2$CH$_2$CH$_2$C*H*$_3$), 1.37–1.47 (m, 4H, —CH$_2$CH$_2$C*H*$_2$C*H*$_2$CH$_3$), 1.83–1.89 (m, 2H, —CH$_2$C*H*$_2$CH$_2$CH$_2$CH$_3$), 4.43 (t, J = 7.60 Hz, 2H, —C*H*$_2$CH$_2$CH$_2$CH$_2$CH$_3$), 7.45–8.16 (m, 4H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 13.75 (—CH$_2$CH$_2$CH$_2$CH$_2$*C*H$_3$), 21.70 (CH$_2$CH$_2$CH$_2$*C*H$_2$CH$_3$), 27.35 (—CH$_2$CH$_2$*C*H$_2$CH$_2$CH$_3$), 28.16 (—CH$_2$*C*H$_2$CH$_2$CH$_2$CH$_3$), 69.52 (—*C*H$_2$CH$_2$CH$_2$CH$_2$CH$_3$), 114.70, 116.81, 126.54, 127.95, 135.57 (C_{Arom}), 146.63 (C-guanidine), 155.33 (C—Cl), 166.57 (C-isourea). ms: *m/z* (%): 290 (M^+, 66), 220 (90), 160 (5), 134 (18), 104 (31), 43 (27). Anal. Calcd. for $C_{14}H_{15}ClN_4O$ (290.75): C, 57.83; H, 5.20; N, 19.27. Found: C, 57.93; H, 5.29; N, 18.98.

2-Benzyloxy-5-chloro[1,2,4]triazolo[1,5-a]quinazoline (10d). White solid; (yield: 90%), m.p. 130°C (THF-hexane). ^{1}H NMR (DMSO-d_6): δ_H 5.79 (s, 2H, CH_2), 7.37–8.45 (m, 9H, ArH), ^{13}C NMR (DMSO-d_6): δ_C 71.34 (CH_2), 115.20, 117.42, 125.50, 126.71, 127.14, 128.07, 128.70, 132.41, 135.90, 136.11 (C_{Arom}), 136.77 (C-guanidine), 155.93 (C—Cl), 165.25 (C-isourea). ms: *m/z* (%): 310 (M^+, 89), 220 (32), 189 (4), 104 (21), 91 (100). Anal. Calcd. for $C_{16}H_{11}ClN_4O$ (310.75): C, 61.84; H, 3.57; N, 18.03. Found: C, 61.80; H, 3.82; N, 17.88.

5-Chloro-2-phenethyloxy[1,2,4]triazolo[1,5-a]quinazoline (10e). White solid; (yield: 91%), m.p. 140°C (THF-hexane). ^{1}H NMR (DMSO-d_6): δ_H 3.15 (t, J = 7.50 Hz, 2H, C*H*$_2$CH$_2$Ph), 4.65 (t, J = 7.51 Hz, 2H, CH$_2$C*H*$_2$Ph), 7.22–8.37 (m, 9H, ArH); ^{13}C NMR (DMSO-d_6): δ_C 35.09 (CH$_2$*C*H$_2$Ph), 69.61 (*C*H$_2$CH$_2$Ph), 114.59, 124.40, 124.83, 126.72, 128.74, 129.30, 134.29, 134.94, 138.49 (C_{Arom}), 153.37 (C-guanidine), 156.84 (C—Cl), 168.61 (C-isourea). ms: *m/z* (%): 324 (M^+, 100), 220 (3), 145 (32), 104 (11), 90 (34). Anal. Calcd. for $C_{17}H_{13}ClN_4O$ (324.77): C, 62.87; H, 4.03; N, 17.25. Found: C, 62.57; H, 4.22; N, 17.15.

1,2,4,5-Tetrahydro[1,2,4]triazolo[1,5-*a*]quinazolin-2,5-dione (11). A mixture of **5f** (1 mmol) and Pd-C 10% (120 mg) as a catalyst was hydrogenated in THF (75 mL) for 2 h. The suspension was filtered off and the solvent evaporated. The resulting solid was suspended in EtOAc (2 mL) and filtered again to afford analytically pure **11** as white solid; (yield: 95%), m.p. 177°C (EtOAc). IR (KBr) 1707, 1686 cm^{-1}. ^1H NMR (DMSO-d_6): δ_H 7.45–8.15 (m, 4H, ArH), 11.84 (s, 1H, NH), 12.98 (s, 1H, NH); ^{13}C NMR (DMSO-d_6): δ_C 114.18, 116.64, 125.11, 128.58, 135.71, 136.24 (C_{Arom}), 147.43 (C-guanidine), 160.04 (C=O), 167.01 (C=O). ms: *m/z* (%) 202 (M^+, 100), 106 (3), 134 (70), 105 (25), 76 (15). Anal. Calcd. for $C_9H_6N_4O_2$ (202.17): C, 53.47; H, 2.99; N, 27.71. Found: C, 53.67; H, 3.02; N, 27.52.

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