



Features of the synthesis of 5,7-di(het)aryltriazolo[1,5-a]pyrimidine derivatives

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Abstract

To date, the synthesis of 5,7-di(het)arylazolo[1,5-a]pyrimidines, including 6-nitro-substituted derivatives, is represented by a limited number of original studies. To expand the synthetic approaches in this field of azaheterocyclic chemistry, we have developed a universal method for the preparation of 5,7-di(het)aryl-6-nitro-4,5,6,7-tetrahydro[1,2,4]triazolo[1,5-a]pyrimidines and the corresponding 6-unsubstituted 5,7-di(hetero)aryl[1,2,4]triazolo[1,5-a]pyrimidines using imines and nitrostyrenes as starting reagents. It was shown that the reaction of 3-substituted-(*E*)-1-(4-nitrophenyl)-*N*-(1*H*-1,2,4-triazol-5-yl)methanimines (1) with nitrostyrene derivatives (2) in acetonitrile and one equivalent of NEt_3 results in the products of denitration-aromatization – 7-(het)aryl-2-(methylthio)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidines (3). This process of nitro group elimination with concomitant aromatization is also observed for 5,7-di(het)aryl-6-nitro-4,5,6,7-tetrahydro[1,2,4]triazolo[1,5-a]pyrimidines and depends on the electronegativity of the substituent at the C-5 atom, the nature of the basic or acidic activators used, and the solvent. A series of experiments were conducted to establish the mechanism of nitro group elimination using 4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (6a) as the starting reagent. During the study, the nitro group reduction product – 7-(4-methoxyphenyl)-2-(methylthio)-5-(pyridin-3-yl)-[1,2,4]triazolo[1,5-a]pyrimidin-6-amine (8) was obtained.

Key findings

- New 5,7-di(het)aryl-6-nitro-4,5,6,7-tetrahydro[1,2,4]triazolo[1,5-a]pyrimidines and 5,7-di(het)aryl-6*H*-[1,2,4]triazolo[1,5-a]pyrimidines derivatives were obtained (yields range from 14-92%)
- The mechanism of nitro group elimination with concomitant aromatization was proposed
- A product of nitro group reduction with concomitant aromatization was obtained using thiophenol as a reducing agent

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1. Introduction

Azolopyrimidines and azoloazines with a bridge nitrogen atom are known for their applicability in various research fields. In particular, due to their similarity to natural purines, such derivatives exhibit a wide range of biological ac-

tivity [1], including antiviral [2, 3], antitumor, anticancer [5, 6] and antifungal [7] activity. Synthetic approaches to the preparation of azolopyrimidine derivatives with a bridge nitrogen atom are diverse, and this structural diversity is virtually unlimited [8].

Nitro-substituted azolopyrimidines are of particular interest. The presence of such a pharmacophore fragment as

a nitro group imparts a variety of properties to the compounds, including a wide range of biological activity such as antidiabetic activity [9], antiglycating action [10], antiviral activity [11], anti-tumor activity [12].

Special attention should be paid to partially hydrogenated and 5,7-di(het)aryl substituted azolopyrimidines (Figure 1), which are also known as broad-spectrum biologically active compounds. The use of 5,7-disubstituted triazolo[1,5-a]pyrimidine derivatives as protective agents against LPS (lipopolysaccharide)-induced acute lung injury has been reported [13].

Complexes of various metals with 5,7-disubstituted 1,2,4-triazolo[1,5-a]pyrimidines exhibit dose-dependent cytotoxic activity against a number of cancer cell lines [14]. For a derivative of this series with an amino group as a substituent in position 6, inhibitory activity against α -glucosidase, one of the molecular targets for the treatment of type 2 diabetes mellitus, has been experimentally established [15].

It is noted that partially hydrogenated 5,7-diphenyl-substituted derivatives of triazolo[1,5-a]pyrimidine are known as inhibitors of the secretion of the surface antigen of the hepatitis B virus [16]. Also, some derivatives of this series are considered as potential components in the complex therapy of viral hepatitis [17] and as blockers dipeptidyl peptidase 4 (DPP-4), serving as a prototype of an antidiabetic agent [9, 18].

Synthetic approaches to the preparation of such 5,7-di(het)aryl-substituted derivatives are scarcely represented in the literature. The first report on the synthesis of these derivatives can be considered the reaction of aminoazoles with chalcones under basic condition [19, 20, 21]. This method has been used for several decades to synthesize potential drugs based on 5,7-di(het)aryl-substituted azolopyrimidines [22, 23, 24]. The aim of the methods is to develop a series of 5,7-di(het)aryl-6H-substituted azolopyrimidine derivatives.

Examples of using other approaches for the synthesis of such compounds are scarce. One such example is multicomponent reactions [25] and the use of organomagnesium compounds [26, 27].

Previously, in our works [28, 29], we proposed new methods for the synthesis and aromatization of 5,7-di(het)aryl-6-nitro-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-a]pyrimidines and also conducted studies of their regio- and stereospecificity using NMR methods.

During the optimization of the conditions for the synthesis of partially hydrogenated 5,7-di(het)aryl-substituted triazolo[1,5-a]pyrimidines, we discovered that a change in the base and its ratio radically changes the reaction result.

2. Results and Discussion

By carrying out a typical reaction for the preparation of partially hydrogenated derivatives of 5,7-di(het)aryl-6-nitro-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-a]pyrimidines under basic condition in acetonitrile using NEt_3 as a base and 3-substituted-(*E*)-1-(4-nitrophenyl)-*N*-(1*H*-1,2,4-triazol-5-yl)methanimines as starting reagents, unexpected products of nitro group elimination concomitant with aromatization were obtained instead of the expected partially hydrogenated derivatives. (Scheme 1).

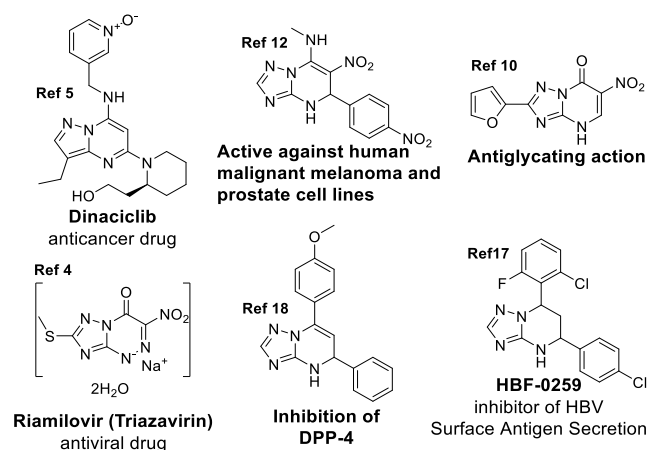
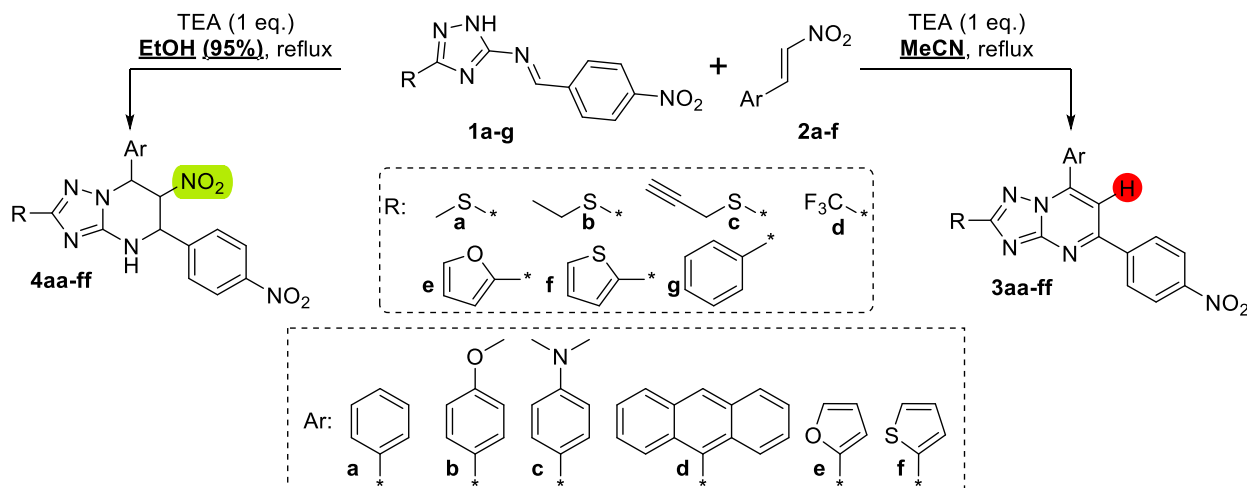


Figure 1 Known biologically active derivatives of azolopyrimidines



Scheme 1 Synthesis of 2-substituted-7-(het)aryl-5-(4-nitrophenyl) derivatives of triazolo[1,5-a]pyrimidines

Table 1 Substituents of the resulting compounds, the reaction conditions and the resulting yields for 5-(4-nitrophenyl) derivatives **3** and **4**

R	Ar	Yield of product, %	
		Cond. A*	Cond. B**
SMe	4-methoxyphenyl	3ab 14%	4ab 40%
	<i>N,N</i> -dimethylaniline-4-yl	3ac 5%	-
	anthracen-9-yl	3ad 32%	4ad 14%
	furan-2-yl	3ae 17%	-
	thiophen-2-yl	-	4af 24%
SEt	4-methoxyphenyl	3bb 29%	-
	4-methoxyphenyl	3cb 54%	4cb 18%
	<i>N,N</i> -dimethylaniline-4-yl	3cc 10%	-
	anthracen-9-yl	3cd 45%	4cd 77%
	furan-2-yl	3ce 45%	-
CF ₃	4-methoxyphenyl	3db 40%	-
	<i>N,N</i> -dimethylaniline-4-yl	3dc 24%	-
	anthracen-9-yl	3dd 36%	-
	thiophen-2-yl	3df 31%	-
	phenyl	3ea 11%	-
furan-2-yl	4-methoxyphenyl	3eb 13%	-
	<i>N,N</i> -dimethylaniline-4-yl	3ec 26%	-
	anthracen-9-yl	3ed 20%	4ed 80%
	furan-2-yl	3ee 14%	-
	thiophen-2-yl	3ef 20%	-
thiophen-2-yl	4-methoxyphenyl	3fb 36%	-
	<i>N,N</i> -dimethylaniline-4-yl	3fc 26%	-
	anthracen-9-yl	-	4fd 92%
	thiophen-2-yl	3ff 58%	4ff 40%
phenyl	4-methoxyphenyl	3gb 18%	-

* Condition A: reflux in MeCN with NET₃ for 8 h** Condition B: reflux in EtOH (95%) with NET₃ for 2-8 h

Table 1 shows substituents of the resulting compounds the reaction conditions and the resulting yields for 5-(4-nitrophenyl) derivatives.

During the study of the observed transformation, the reaction conditions were optimized to determine the influence of solvent, base and their ratio. Based on the results of the selected conditions, it was found that use of other bases, both organic and inorganic, in acetonitrile leads to product of nitro group elimination concomitant with aromatization (hereinafter called “denitration”) (Table 2). Optimization was performed using a model compound **3ab**.

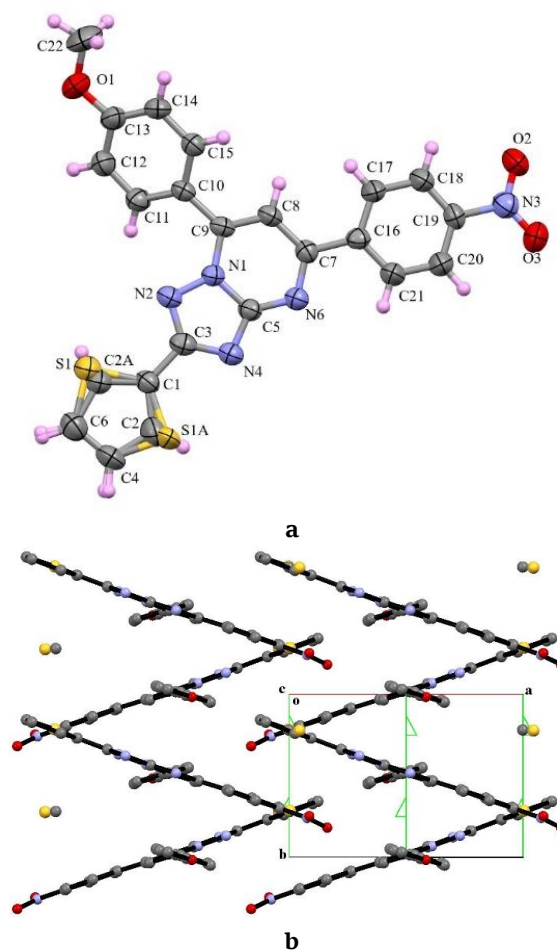
It was found that use of bases such as NET₃, DIPEA, DBU, and DABCO in acetonitrile leads to the formation of a denitration product; however, the yields are extremely low. Catalysis with K₂CO₃ in MeCN_(dry) did not result in the formation product of denitration. Process of denitration was also observed in EtOH (95%) in the presence of DBU (1 eq.). The formation of partially hydrogenated derivatives was observed in the presence of NET₃ in EtOH (95%) and MeOH.

The resulting denitration products were characterized by NMR and IR spectroscopy, Mass spectrometry, and X-ray diffraction analysis (XRD), which clearly correlates with the proposed structure (Figure 2).

According XRD data, the compound is crystallized in the non-centrosymmetric space group of the monoclinic system. The compound is non-planar due to rotating of the 4-MeOPh-group toward the plane of the heterocycle on the angle 35°. The thienyl substitute is disordered in the plane into two positions with occupancy coefficients 0.7/0.3. The bond distances and angles for the molecule of the compound are near to expectation (Figure 2a).

Table 2 Reaction optimization conditions for the preparation of 5-(4-nitrophenyl)-substituted triazolo[1,5-a]pyrimidine derivatives

N°	Solvent	Base	Base eq.	Yield of product, %	
				3ab	4ab
1	MeCN	NET ₃ ^a	1	14%	-
2	MeCN	NET ₃	0.5	8%	-
3	Dioxane	NET ₃	1	10%	-
4	EtOH (95%)	NET ₃	1	-	41%
5	EtOH (95%)	NET ₃	1.5	-	40%
6	MeOH	NET ₃	1	-	35%
7	MeCN	DIPEA ^b	1	9%	-
8	MeCN	DBU ^c	0.1	8%	-
9	MeCN	DBU	0.5	8%	-
10	EtOH (95%)	DBU	1	5%	-
11	MeCN _(dry)	K ₂ CO ₃	1	-	-
12	MeCN	Cs ₂ CO ₃	1	9%	-
13	MeCN	DABCO ^d	1	6%	-
14	MeCN	DMAP ^e	1	-	-
15	MeCN	-	-	-	-
16	Solvent free	-	-	trace	-

^a Triethylamine^b *N,N*-Diisopropylethylamine^c 1,8-Diazabicyclo[5.4.0]undec-7-ene^d 1,4-diazabicyclo[2.2.2]octane^e 4-Dimethylaminopyridine**Figure 2** a - The compound **3fb** according XRD data in the thermal ellipsoids of the 50 % probability level; b - The unit cell and crystal packing of the molecules of the compound **3fb** in the crystal (the H-atoms are omitted).

The crystal packing of the molecules is ordered by presence the screw axes (Figure 2b). The significant π - π -interactions (lesser than 3.35 Å) between molecules in the crystal do not observed. The shortened intermolecular contacts

are presented by the weak H-bonds of the C_{Ar}-H...O type with participation of the NO₂-group and the O-atom of the MeO-group. In particular, the conformation of the molecule is fixed by the H-bonds between the C(8), C(15) atoms and the O(2) atom [-2-x, y-0.5, -z-1] of the NO₂-group of the nearest molecule

It is worth noting that such reaction turned out to be specific for compounds containing an aromatic fragment with electron-withdrawing properties (4-nitrophenylene) and not for electron-donating (4-methoxyphenylene) substituents [28].

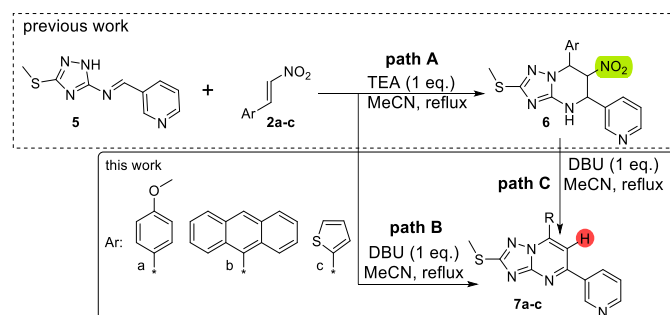
Analyzing this transformation and the influence of the imine configuration on the reaction course, it became interesting to know whether a similar reaction could occur with 3-(methylthio)-(E)-1-(pyridin-3-yl)-N-(1H-1,2,4-triazol-5-yl)methanimines, described in our previous work [29].

We have previously established that the reaction of imine and nitroethylene under basic conditions using NEt₃ (1 eq.) in MeCN affords partially saturated azolo[1,5-a]pyrimidine derivatives in the *trans-trans* configuration (Scheme 2, Pathway A). The formation of 5,7-di(het)aryl-6-nitro-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (**6**) was found using DBU (0.1 eq.) in acetonitrile.

Following the newly discovered process, the reaction was carried out using DBU (1 eq.) in MeCN (Scheme 2, Pathway B). The reaction yielded a product of denitration (**7a-c**).

c). It's worth noting that refluxing the previously isolated tetrahydro azolopyrimidine derivative with DBU (1 eq.) also yielded the product of denitration, even though in lower yields, and in some cases, a mixture of the starting tetrahydro and product of denitration.

To identify possible intermediates in the reaction between imines and nitroethylenes (Scheme 2, path B), an NMR experiment was conducted. Samples were taken from the reaction mixture at the following time points: 0 min – loading of all components, then 5, 30, 60, 120, and 540 minutes after loading all reagents. ¹H NMR spectra were recorded in DMSO-*d*₆. The resulting NMR spectra revealed new signals in the 4-7 ppm range, characteristic of possible dihydro intermediates (Figure 3).



Scheme 2 Synthesis of 7-(het)aryl-5-(pyridine-3-yl)-2-(methylthio)-substituted triazolo[1,5-a]pyrimidine

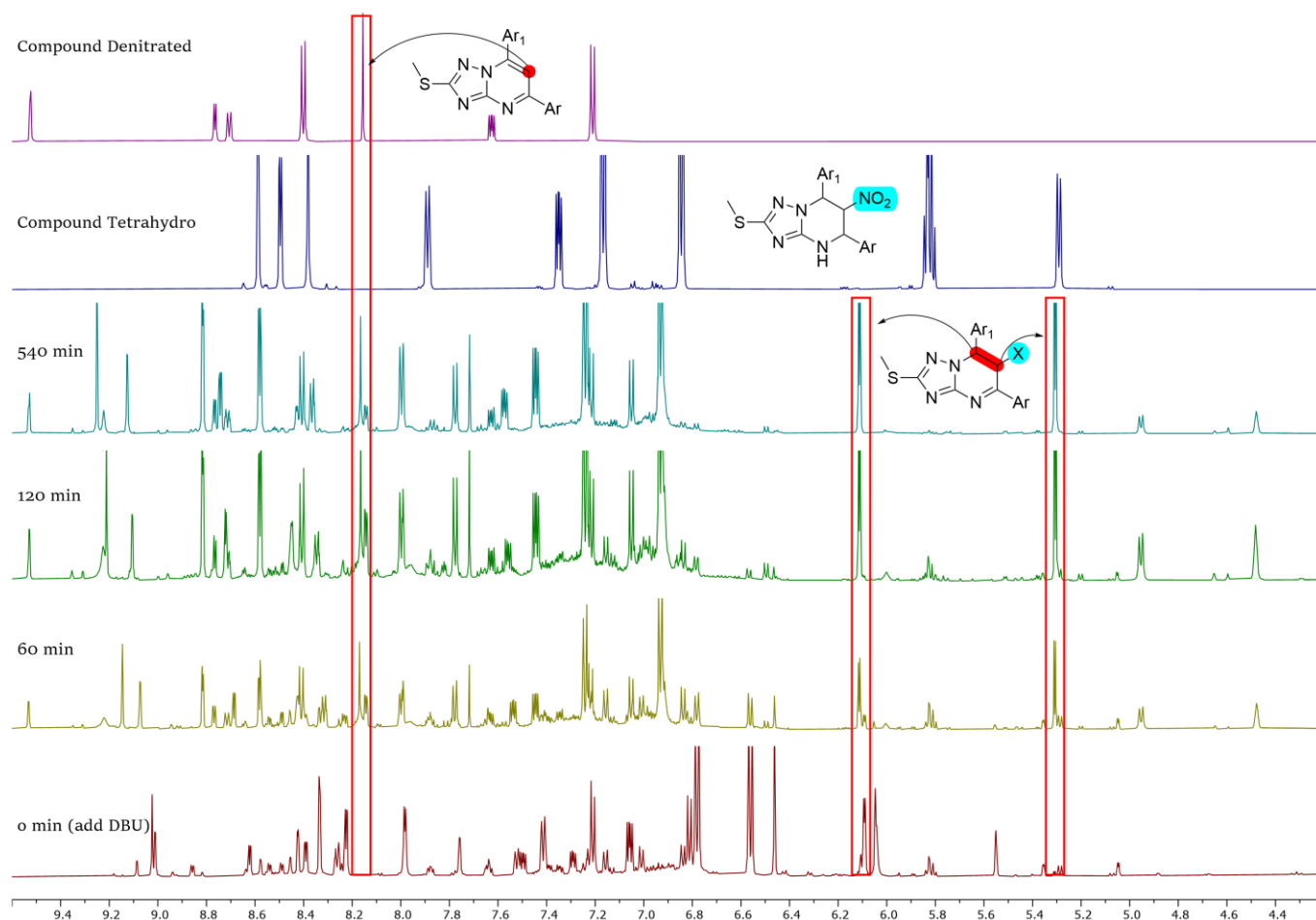


Figure 3 ¹H NMR spectra of the reaction mass experiment with compound **7a**

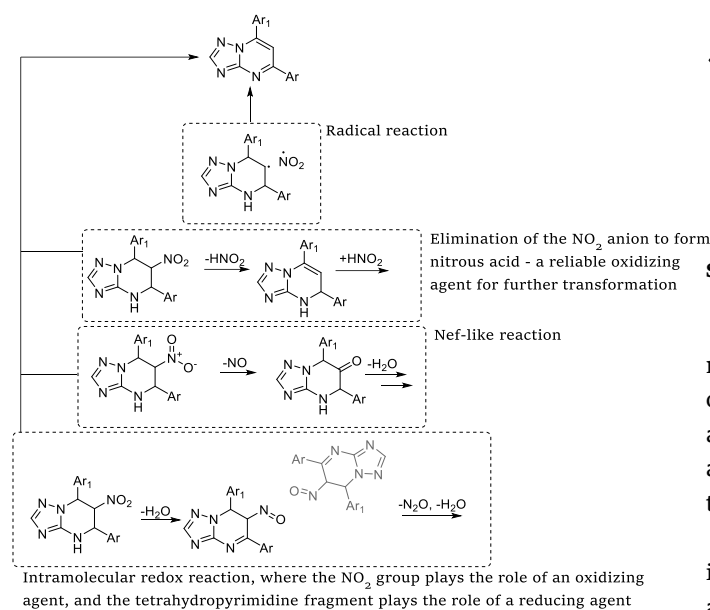
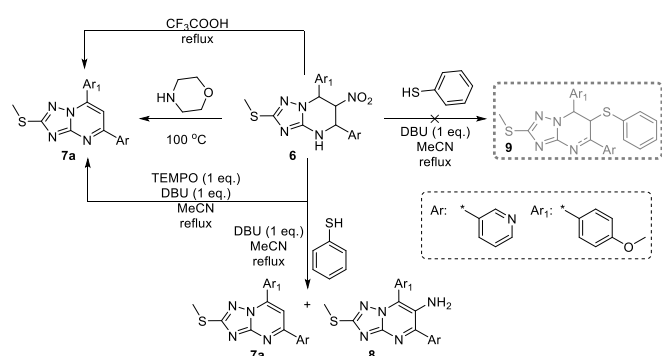


Figure 4 Possible mechanisms of the denitration reaction

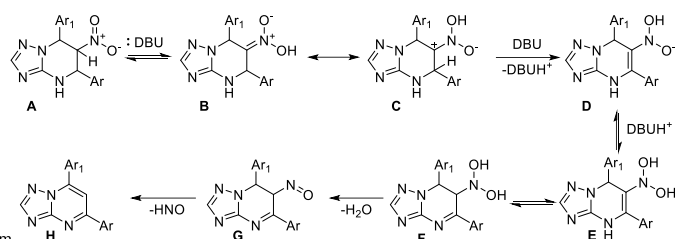


Scheme 3 Reactions of the tetrahydro derivative (**6a**) with nucleophiles, in CF_3COOH and the presence of TEMPO

Thus, the preliminarily isolated tetrahydro derivatives of azolopyrimidines served as a starting point for further discussion of the reaction mechanism.

The process of nitro group elimination with accompanying aromatization of the heterocycle, can occur according to several mechanisms (Figure 4), among which the following can be distinguished: the Nef reaction [30], a radical process [31], NAE – nitrous acid elimination [32, 33] followed by further oxidation with atmospheric oxygen or eliminated HNO_2 , or intramolecular partial reduction of the nitro group followed by HNO or $\text{N}_2\text{O} + \text{H}_2\text{O}$ elimination.

To establish the possibility of a radical process for the elimination of the nitro group, an experiment was conducted with a TEMPO radical trap (Scheme 3). As a result, the product of denitration was obtained in an even higher yield. This effect can apparently be achieved due to the independent participation of TEMPO as a co-oxidant [34, 35, 36]. A GC-MS study of the reaction mixture composition revealed no intermediates corresponding to known mechanisms, such as Nef-type reaction intermediates (such as the 6-one derivative) or adducts with TEMPO. The reaction was also carried out in an inert atmosphere, resulting in the isolation of the product of denitrated.



Scheme 5 Proposed mechanism of the denitration reaction

To establish the crucial role of the aci-nitro form in the reaction of denitration from the tetrahydro derivative of azolopyrimidines, a similar reaction was carried out under acidic conditions with DBU replaced by trifluoroacetic acid as the solvent and activator. This allowed the isolation of the desired aromatic product (**7a**), but in lower yield.

To study the possibilities of nitro group elimination during nucleophilic substitution processes (S_N processes) (Nef and NAE reactions, mechanistic scheme), a reaction was carried out starting from derivative **6a**, with the addition of thiophenol as an external nucleophile [37, 38]. Unexpectedly, a mixture of denitrated and aromatic 6-amino derivatives (**8**) was obtained. A similar process of ring aromatization during nitro group reduction was previously noted by Gorbunov et al [39].

However, additional products from the addition of thiophenol to the heterocycle (**9**), as well as the replacement of thiophenol with morpholine, did not yield any adducts.

Thus, of all the probable mechanisms, the remaining process involves nitro group reduction concomitant with the oxidation of tetrahydropyrimidine ring and further HNO elimination (Scheme 5).

According to the proposed mechanism, the “denitration” step is determined by the formation of dihydroxylamine **E** from the aci-nitro form of starting derivative **A**. These steps depend on the dissociability of the acidic proton at the C5 atom and, correspondingly, on the electron-withdrawal effect provided by the aromatic substituent at the C5 atom, as demonstrated in the experiments described above.

Next, the tautomeric transformation of dihydroxylamine into the **F** form occurs, followed by dehydration to form nitroso **G**, followed by the elimination of HNO . Since it is not entirely clear whether this process is completely intramolecular, we also assumed that the elimination of the nitrosyl group can occur by a bimolecular mechanism with the formation of N_2O and H_2O .

3. Limitations

The proposed approach makes it possible to obtain derivatives of the 5,7-di(het)aryl substituted azolopyrimidines from the corresponding tetrahydro precursors or through the initial reaction between imines and β -nitrostyrenes by adjusting the starting conditions. The conducted studies of the mechanism of the proposed transformation make it possible to expand the general understanding of the methods of elimination of the nitro group in partially hydrogenated

azaheterocyclic systems. The main limitation of the transformation with the elimination of the nitro group is the presence of a pronounced electron acceptor substituent at the C5 atom in the studied series of azolo[1,5-a]pyrimidines.

4. Conclusions

In conclusion, we have discovered a novel denitration process concomitant with aromatization in the series of partially hydrogenated 6-nitroazolo[1,5-a]pyrimidines (**6**) and proposed some structure/condition-reactivity relationship. Based on the experimental data, a reaction mechanism is proposed. The discovered process was successfully used to develop a synthetic procedure for the preparation of new 5,7-di(het)aryl-6H-[1,2,4]triazolo[1,5-a]pyrimidine (**3** and **7**) derivatives. Furthermore, a series of previously undescribed 5-(4-nitrophenyl)-7-(hetero)aryl-6-nitro-4,5,6,7-tetrahydro[1,2,4]triazolo[1,5-a]pyrimidines (**4**) were synthesized. The structures of all newly obtained compounds were confirmed by NMR and IR spectroscopy, mass spectrometry and elemental analysis. The structure of the denitration product was unambiguously established by XRD for (**3fb**).

5. Materials and Methods

All experimental data were obtained in Laboratory of Complex Research and Expert Evaluation of Organic Materials, [Center for Collective Use of unique equipment of the Ural Federal University](#).

All solvents and commercially available reagents were used as received unless otherwise stated. Non-commercial starting materials were prepared as described below or according to the literature procedures. One-dimensional ^1H and ^{13}C NMR spectra were acquired at 25°C from solutions in DMSO- d_6 on a «Bruker AVANCE NEO – 600» (600 MHz for ^1H , 151 MHz for ^{13}C MHz, 565 MHz for ^{19}F) NMR spectrometer equipped with CryoProbe Prodigy, and Topspin 4.0.5 software was used for all NMR data acquisition. Chemical shifts were referenced to the residual peaks of solvent as an internal standard: 2.50 ppm for ^1H NMR and 39.52 ppm for ^{13}C NMR in DMSO- d_6 . The residual signal in the ^1H NMR spectrum referenced to DMSO- d_5 . Chemical shifts are expressed in δ (parts per million, ppm) values, and coupling constants are expressed in hertz (Hz). The following abbreviations are used for the multiplicity of NMR signals: s, singlet; d, doublet; t, triplet; q, quartet; dd, doublet of doublets; br.s, broadened signal; m, multiplet.

The reaction course was monitored by TLC on plates (0.25 mm) with silica gel (Merck 60F 254) using ethyl acetate as eluent. IR spectra were recorded on a Bruker Alpha FTIR spectrometer with an attenuated total internal reflectance (ATR) accessory and with KBr. Mass spectra were detected on a Shimadzu GCMS-QP2010 mass spectrometer (electron impact ionization, 70 eV). Elemental analysis was

performed using a Perkin-Elmer 2400 Series II CHNS/O analyzer. Melting points were determined on a Stuart S.PL.3 instrument at a heating rate of 10°C min $^{-1}$.

The XRD analysis was carried out using equipment of the Center for Joint Use “Spectroscopy and Analysis of Organic Compounds” at the Postovsky Institute of Organic Synthesis of the Russian Academy of Sciences (Ural Branch). The experiment was accomplished on the automated X-ray diffractometer «Xcalibur 3» with CCD detector on standard procedure (MoK α -irradiation, graphite monochromator, ω -scans with 10 step at T= 295(2) K). Empirical absorption correction was applied. Using Olex2 [40], the structure was solved with the ShelXS [41] structure solution program using Direct Methods and refined with the ShelXL [42] refinement package using Least Squares minimization. The H-atoms at C-H bonds were placed in the calculated positions and were refined in the “rider” model. The crystallographic data for structures **3fb** was presented in Table S1 in Supplementary materials. The XRD data were deposited in the Cambridge Structural Database with number CCDC 2544393. This data can be requested free of charge via www.ccdc.cam.ac.uk.

2-nitrovinyl derivatives (β -nitrostyrenes) were synthesized according to the reported procedure in (2-nitrovinyl)benzene (β -nitrostyrene) (**2a**), 1-methoxy-4-(2-nitrovinyl)benzene (**2b**), 2-(2-nitrovinyl)furan (**2e**), 2-(2-nitrovinyl)thiophene (**2f**) [43], *N,N*-dimethyl-4-(2-nitrovinyl)Aniline (**2c**) [44], 9-(2-nitrovinyl)anthracene [45] (**2d**).

New imines (**1a-g**) were synthesized as described in literature [28].

(E)-N-(3-(methylthio)-1H-1,2,4-triazol-5-yl)-1-(4-nitrophenyl)methanimine (1a). Yellow powder (83%). m.p.: 267-269 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 14.26 (br.s, 1H), 9.30 (s, 1H), 8.36 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 8.25 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 2.61 (s, 3H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 162.3, 156.5, 149.3, 140.6, 130.2, 124.1, 13.9. IR ν/cm^{-1} : 3192, 3144 (N-H), 3084 (C-H $_{\text{Ar}}$), 1622 (C-N=C), 1595 (C-H $_{\text{Ar}}$), 1515, 1345 (NO $_2$), 1449, 1443 (CH $_3$). Mass spectrum (EI, 70 eV, m/z): 263 [M] $^+$. Calcd for C $_{10}$ H $_9$ N $_5$ O $_2$ S: C, 45.71; H, 3.52; N, 26.68; S, 12.23, found C, 45.62; H, 3.45; N, 26.60; S, 12.18.

(E)-N-(3-(ethylthio)-1H-1,2,4-triazol-5-yl)-1-(4-nitrophenyl)methanimine (1b). Yellow powder (45%). m.p.: 181-183 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 14.29 (br. d, 1H), 9.31 (s, 1H), 8.36 (d, $^3J_{\text{HH}} = 8.4$ Hz, 2H), 8.26 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 3.15 (s, 2H), 1.33 (t, $^3J_{\text{HH}} = 7.3$ Hz, 3H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 162.6, 162.2, 155.3, 149.3, 140.7, 130.2, 124.0, 25.8, 15.1. IR ν/cm^{-1} : 3204 (N-H), 3086 (C-H $_{\text{Ar}}$), 2897-2730 (-CH $_2$ -CH $_3$), 1618 (C-N=C), 1593 (C-H $_{\text{Ar}}$), 1514, 1343 (NO $_2$). Mass spectrum (EI, 70 eV, m/z): 277 [M] $^+$. Calcd for C $_{11}$ H $_{11}$ N $_5$ O $_2$ S: C, 47.70; H, 4.09; N, 25.35; S, 11.63, found C, 47.65; H, 4.00; N, 25.26; S, 11.56.

(E)-1-(4-nitrophenyl)-N-(3-(prop-2-yn-1-ylthio)-1H-1,2,4-triazol-5-yl)methanimine (1c). Yellow powder (72%). m.p.: 220-222 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 14.51 (s, 1H), 9.33 (s, 1H), 8.37 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 8.26 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 4.00 (s, 2H), 3.22 (s, 1H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 163.2, 162.4, 155.2, 149.9, 141.0, 130.8, 124.5, 80.4, 74.6, 20.3. IR ν/cm^{-1} : 3304 (C \equiv CH), 3253 (N-H), 3000 (C-H $_{\text{Ar}}$), 1619 (C-N=C), 1594 (C-H $_{\text{Ar}}$), 1508,

1329 (NO₂). Mass spectrum (EI, 70 eV, m/z): 287 [M]⁺. Calcd for C₁₂H₉N₅O₂S: C, 50.10; H, 3.10; N, 24.30; S, 11.10, found C, 50.17; H, 3.16; N, 24.38; S, 11.16.

(E)-1-(4-nitrophenyl)-N-(3-(trifluoromethyl)-1H-1,2,4-triazol-5-yl)methanimine (1d). Yellow powder (35%). m.p.: 258-260 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 9.43 (s, 1H), 8.39-8.35 (m, 2H), 8.32-8.27 (m, 2H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 166.5, 159.7, 151.2 (q, ²J_{CF} = 38 Hz), 149.9, 139.7, 130.9, 124.1, 119.4 (q, ¹J_{CF} = 270 Hz). ¹⁹F NMR (565 MHz, DMSO-*d*₆) δ -64.40. IR ν/cm⁻¹: 3289 (N-H), 2954, 2910 (C-H_{Ar}), 1625 (C-N=C), 1592 (C-H_{Ar}), 1520, 1345 (NO₂). Mass spectrum (EI, 70 eV, m/z): 285 [M]⁺. Calcd for C₁₀H₆F₃N₅O₂: C, 42.10; H, 2.10; N, 24.50, found C, 42.12; H, 2.12; N, 24.56.

(E)-N-(3-(furan-2-yl)-1H-1,2,4-triazol-5-yl)-1-(4-nitrophenyl)methanimine (1e). Yellow powder (83%). m.p.: 279-281 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 14.59 (br.s, 1H), 9.38 (s, 1H), 8.35 (d, ³J_{HH} = 8.6 Hz, 2H), 8.27 (d, ³J_{HH} = 8.6 Hz, 2H), 7.87 (d, ³J_{HH} = 1.8 Hz, 1H), 7.03 (d, ³J_{HH} = 3.4 Hz, 1H), 6.68 (dd, ³J_{HH} = 3.4, 1.8 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 162.7, 162.2, 150.3, 149.3, 144.5, 144.3, 140.7, 130.2, 124.0, 111.9, 109.9. IR ν/cm⁻¹: 3110 (N-H), 3054 (C-H_{Ar}), 1619 (C-N=C), 1594 (C-H_{Ar}), 1512, 1350 (NO₂). Mass spectrum (EI, 70 eV, m/z): 283 [M]⁺. Calcd for C₁₃H₉N₅O₃: C, 55.20; H, 3.29; N, 24.81, found C, 55.13; H, 3.20; N, 24.73.

(E)-1-(4-nitrophenyl)-N-(3-(thiophen-2-yl)-1H-1,2,4-triazol-5-yl)methanimine (1f). Yellow powder (70%). m.p.: 269-271 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 14.57 (br.s, 1H), 9.37 (s, 1H), 8.36 (d, ³J_{HH} = 8.4 Hz, 2H), 8.29 (d, ³J_{HH} = 8.3 Hz, 2H), 7.70 (d, ³J_{HH} = 4.2 Hz, 2H), 7.20 (t, ³J_{HH} = 4.4 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 162.2, 153.5, 149.3, 140.7, 131.8, 130.3, 128.1, 126.4, 124.0. IR ν/cm⁻¹: 3258 (N-H), 3000 (C-H_{Ar}), 1621 (C-N=C), 1596 (C-H_{Ar}), 1515, 1338 (NO₂). Mass spectrum (EI, 70 eV, m/z): 299 [M]⁺. Calcd for C₁₃H₉N₅O₂S: C, 52.10; H, 3.03; N, 23.35; S, 10.70, found C, 52.17; H, 3.03; N, 23.40; S, 10.71.

(E)-1-(4-nitrophenyl)-N-(3-phenyl-1H)-1,2,4-triazol-5-yl)methanimine (1g). Yellow powder (71%). m.p.: 264-266 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 14.56 (s, 1H), 9.40 (s, 1H), 8.34 (d, ³J_{HH} = 8.7 Hz, 2H), 8.27 (d, ³J_{HH} = 8.8 Hz, 2H), 8.06-8.01 (m, 2H), 7.53 (t, ³J_{HH} = 7.3 Hz, 2H), 7.51-7.46 (m, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 163.4, 161.7, 156.9, 149.2, 140.9, 130.1, 129.8, 128.9, 125.9, 124.0. IR ν/cm⁻¹: 3288 (N-H), 3167, 3056 (C-H_{Ar}), 1622 (C-N=C), 1596 (C-H_{Ar}), 1513, 1337 (NO₂). Mass spectrum (EI, 70 eV, m/z): 293 [M]⁺. Calcd for C₁₅H₁₁N₅O₂: C, 61.40; H, 3.70; N, 23.80, found C, 61.43; H, 3.78; N, 23.88.

7-(het)aryl-2-(methylthio)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidines (3) (general methods). Imines **1** (1 mmol) and β-nitroethylenes **2** (1 mmol) was suspended in 5 mL of solvent MeCN in a flask, treated with NEt₃ (1 mmol), and refluxed for 8 h. The hot solution was filtered precipitate was washed with MeCN 3-5 mL and dried to a constant weight. The filtrate was evaporated under reduced pressure to dryness oil, treated with hot MeCN 3-5 mL, filtered and dried in air to a constant weight in order to extract an additional amount of the target product.

7-(4-methoxyphenyl)-2-(methylthio)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ab). Imines **1a** (1 mmol) and β-nitroethylenes **2b** (1 mmol) was suspended in 5 mL of solvent MeCN in a flask, treated with NEt₃ (1 mmol), and refluxed for 8 h. The hot solution was

filtered precipitate was washed with MeCN 3-5 mL. The filtrate was poured into an evaporation dish. The product crystallized from the filtrate for 30 minutes. The precipitate was filtered, washed with 2-3 ml MeCN, and dried to a constant weight in air. Yellow powder (14%). m.p.: 237-239 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.59 - 8.54 (m, 2H), 8.39 - 8.36 (m, 2H), 8.36 - 8.33 (m, 2H), 8.12 (s, 1H), 7.21 - 7.16 (m, 2H), 3.89 (s, 3H), 2.68 (s, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 168.3, 162.1, 157.0, 156.3, 148.7, 146.3, 142.0, 131.8, 128.9, 123.8, 121.4, 114.0, 105.4, 55.6, 13.3. IR (KBr) ν/cm⁻¹: 3107, 3074 (C-H_{Ar}), 2932, 2837 (C-H_{alif}), 1604 (C-H_{Ar}), 1545, 1345 (NO₂). Mass spectrum (EI, 70 eV, m/z): 393 [M]⁺. Calcd for C₁₉H₁₅N₅O₃S: C, 58.05; H, 3.88; N, 17.86; S, 8.18, found C, 58.01; H, 3.84; N, 17.80; S, 8.15.

N,N-dimethyl-4-(2-(methylthio)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidin-7-yl)aniline (3ac). Burgundy powder (5%). m.p.: 274-276 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.59 (d, ³J_{HH} = 8.8 Hz, 2H), 8.42 (d, ³J_{HH} = 9.1 Hz, 2H), 8.37 (d, ³J_{HH} = 8.8 Hz, 2H), 8.09 (s, 1H), 6.88 (d, ³J_{HH} = 9.2 Hz, 2H), 3.08 (s, 6H), 2.71 (s, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 167.9, 156.7, 156.7, 152.5, 148.6, 146.8, 142.5, 131.4, 128.9, 123.9, 115.1, 111.1, 103.7, 39.9, 13.3. IR ν/cm⁻¹: 3060, 3004 (C-H_{Ar}), 2979, 2934 (C-H_{alif}), 1601 (C-H_{Ar}), 1530, 1349 (NO₂). Mass spectrum (EI, 70 eV, m/z): 406 [M]⁺. Calcd for C₂₀H₁₈N₆O₂S: C, 59.13; H, 4.49; N, 20.71; S, 7.91, found C, 59.10; H, 4.46; N, 20.68; S, 7.89.

7-(anthracen-9-yl)-2-(methylthio)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ad). Brown powder (32%) m.p.: 325-327 (decompos) °C. ¹H NMR (400 MHz, DMSO-*d*₆) δ 8.97 (s, 1H), 8.69 - 8.63 (m, 2H), 8.41 (d, ³J_{HH} = 3.5 Hz, 2H), 8.39 (s, 1H), 8.27 (d, ³J_{HH} = 8.5 Hz, 2H), 7.66 - 7.58 (m, 4H), 7.48 (ddd, ³J_{HH} = 8.4, 6.6, 1.3 Hz, 2H), 2.49 (s, 3H). ¹³C NMR (101 MHz, DMSO-*d*₆) δ 168.7, 157.4, 156.1, 148.9, 145.2, 141.9, 130.6, 130.1, 129.2, 129.1, 128.6, 127.5, 125.8, 125.1, 124.0, 123.6, 111.1, 13.3. IR (KBr) ν/cm⁻¹: 3082, 3050, 3006 (C-H_{Ar}), 2930, 2852 (C-H_{alif}), 1624 (C-H_{Ar}), 1513, 1337 (NO₂). Mass spectrum (EI, 70 eV, m/z): 463 [M]⁺. Calcd for C₂₆H₁₇N₅O₂S: C, 67.40; H, 3.72; N, 15.16; S, 6.96, found C, 67.37; H, 3.70; N, 15.11; S, 6.92.

2-(methylthio)-5-(4-nitrophenyl)-7-(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ae). Yellow powder (17%). m.p.: 254-256 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.80 (dd, ³J_{HH} = 4.0, 1.2 Hz, 1H), 8.65 - 8.60 (m, 2H), 8.54 (s, 1H), 8.41 - 8.37 (m, 2H), 8.24 (dd, ³J_{HH} = 5.0, 1.2 Hz, 1H), 7.47 (dd, ³J_{HH} = 5.0, 3.9 Hz, 1H), 2.77 (s, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 168.5, 156.9, 156.0, 148.8, 142.1, 140.5, 136.3, 133.8, 129.8, 128.9, 128.2, 124.0, 102.7, 13.5. IR (KBr) ν/cm⁻¹: 3103, 3087, 3071 (C-H_{Ar}), 2925, 2836 (C-H_{alif}), 1593 (C-H_{Ar}), 1553, 1345 (NO₂). Mass spectrum (EI, 70 eV, m/z): 369 [M]⁺. Calcd for C₁₆H₁₁N₅O₂S₂: C, 52.06; H, 3.02; N, 18.99; S, 17.39, found C, 52.02; H, 3.00; N, 18.96; S, 17.36.

2-(ethylthio)-7-(4-methoxyphenyl)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3bb). Brown powder (29%). m.p.: 185-187 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.64-8.56 (m, 2H), 8.41-8.33 (m, 4H), 8.15 (s, 1H), 7.23 - 7.16 (m, 2H), 3.90 (s, 3H), 3.27 (q, ³J_{HH} = 7.3 Hz, 2H), 1.43 (t, ³J_{HH} = 7.3 Hz, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 167.7, 162.1, 157.1, 156.3, 148.7, 146.4, 142.0, 131.8, 128.9, 123.9, 121.5, 114.0, 105.5, 55.6, 25.0, 15.0. IR ν/cm⁻¹: 3068, 3012, 2972 (C-H_{Ar}), 2938, 2841 (C-H_{alif}), 1604 (C-H_{Ar}), 1520, 1341 (NO₂). MS (EI, m/z): 407 [M]⁺. Calcd for C₂₀H₁₇N₅O₃S: C, 58.99; H, 4.28; N, 17.21; S, 7.89, found C, 58.96; H, 4.21; N, 17.19; S, 7.87.

7-(4-methoxyphenyl)-5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-[1,2,4]triazolo[1,5-a]pyrimidine (3cb). Yellow powder (54%). m.p.: 189-191 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.63 – 8.59 (m, 2H), 8.43 – 8.40 (m, 2H), 8.39 – 8.36 (m, 2H), 8.19 (s, 1H), 7.21 – 7.18 (m, 2H), 4.15 (d, ³J_{HH} = 2.6 Hz, 2H), 3.90 (s, 3H), 3.26 (t, ³J_{HH} = 2.6 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 166.1, 162.2, 157.4, 156.3, 148.8, 146.6, 142.0, 131.9, 129.0, 123.9, 121.3, 114.0, 105.8, 79.9, 73.9, 55.6, 19.1. IR ν/cm⁻¹: 3273 (C≡C-H), 3016 (C-H_{Ar}), 2963, 2937, 2897, 2839 (C-H_{alif}), 1602 (C-H_{Ar}), 1511, 1340 (NO₂). Mass spectrum (EI, 70 eV, m/z): 417 [M]⁺. Calcd for C₂₁H₁₅N₅O₃S: C, 60.46; H, 3.65; N, 16.79; S, 7.69, found C, 60.42; H, 3.62; N, 16.78; S, 7.68.

***N,N*-dimethyl-4-(5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-[1,2,4]triazolo[1,5-a]pyrimidin-7-yl)aniline (3cc).** Burgundy-colored powder (10%). m.p.: 230-232 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.64 – 8.60 (m, 2H), 8.47 – 8.44 (m, 2H), 8.41 – 8.37 (m, 2H), 8.14 (s, 1H), 6.91 – 6.87 (m, 2H), 4.16 (d, ³J_{HH} = 2.6 Hz, 2H), 3.26 (t, ³J_{HH} = 2.6 Hz, 1H), 3.08 (s, 6H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 165.7, 156.8, 156.6, 152.4, 148.6, 146.9, 142.3, 131.4, 128.9, 123.8, 114.9, 111.0, 103.8, 80.0, 73.8, 39.5, 19.0. IR ν/cm⁻¹: 3271 (C≡C-H), 3106, 3085 (C-H_{Ar}), 2972, 2909, 2888, 2860, 2806 (C-H_{alif}), 1607 (C-H_{Ar}), 1518, 1343 (NO₂). Mass spectrum (EI, 70 eV, m/z): 430 [M]⁺. Calcd for C₂₂H₁₈N₆O₂S: C, 61.40; H, 4.23; N, 19.56; S, 7.48, found C, 61.38; H, 4.21; N, 19.52; S, 7.45.

7-(anthracen-9-yl)-5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-[1,2,4]triazolo[1,5-a]pyrimidine (3cd). Green powder (45%). m.p.: 250-252 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.97 (s, 1H), 8.69 – 8.64 (m, 2H), 8.44 (s, 1H), 8.42 – 8.38 (m, 2H), 8.27 (d, ³J_{HH} = 8.5 Hz, 2H), 7.68 – 7.64 (m, 2H), 7.63 – 7.58 (m, 2H), 7.51 – 7.47 (m, 2H), 4.01 (d, ³J_{HH} = 2.6 Hz, 2H), 3.15 (t, ³J_{HH} = 2.6 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 166.6, 157.7, 156.0, 148.9, 145.5, 141.8, 130.6, 130.2, 129.2, 129.2, 128.7, 127.5, 125.9, 125.1, 124.0, 123.4, 111.4, 79.8, 73.9, 19.0. IR ν/cm⁻¹: 3295 (C≡C-H), 3157, 3110, 3073 (C-H_{Ar}), 2970, 2924 (C-H_{alif}), 1600 (C-H_{Ar}), 1520, 1342 (NO₂). Mass spectrum (EI, 70 eV, m/z): 487 [M]⁺. Calcd for C₂₈H₁₇N₅O₂S: C, 69.00; H, 3.55; N, 14.39; S, 6.60, found C, 68.98; H, 3.51; N, 14.37; S, 6.58.

7-(furan-2-yl)-5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-[1,2,4]triazolo[1,5-a]pyrimidine (3ce). Brown powder (25%). m.p.: 218-220 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.53 (dd, ³J_{HH} = 6.8, 4.9 Hz, 2H), 8.34 (d, ³J_{HH} = 7.4 Hz, 2H), 8.27 (d, ³J_{HH} = 1.9 Hz, 1H), 8.14 (d, ³J_{HH} = 3.4 Hz, 2H), 6.98 (dd, ³J_{HH} = 3.6, 1.7 Hz, 1H), 4.21 (d, ³J_{HH} = 2.6 Hz, 2H), 3.26 (t, ³J_{HH} = 2.6 Hz, 1H). ¹³C NMR (151 MHz, DMSO) δ 166.8, 157.1, 155.8, 148.8, 148.4, 142.4, 141.7, 135.9, 128.9, 124.0, 121.0, 113.9, 101.4, 79.9, 73.9, 19.1. IR ν/cm⁻¹: 3288 (C≡C-H), 3138, 3120, 3067 (C-H_{Ar}), 1606 (C-H_{Ar}), 1521, 1337 (NO₂). Mass spectrum (EI, 70 eV, m/z): 377 [M]⁺. Calcd for C₁₈H₁₁N₅O₃S: C, 57.30; H, 2.99; N, 18.59; S, 8.56, found C, 57.29; H, 2.94; N, 18.56; S, 8.50.

5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-7-(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (3cf). Orange powder (25%). m.p.: 239-241 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.79 (dd, ³J_{HH} = 4.0, 1.2 Hz, 1H), 8.63 – 8.60 (m, 2H), 8.54 (s, 1H), 8.40 – 8.37 (m, 2H), 8.24 (dd, ³J_{HH} = 5.0, 1.2 Hz, 1H), 7.46 (dd, ³J_{HH} = 5.0, 3.9 Hz, 1H), 4.21 (d, ³J_{HH} = 2.6 Hz, 2H), 3.26 (t, ³J_{HH} = 2.6 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 166.3, 157.0, 155.9, 148.8, 141.9, 140.7, 136.4, 133.9, 129.7, 128.9, 128.2, 123.9, 103.0, 79.8, 74.0, 19.2. IR ν/cm⁻¹: 3296 (C≡C-H), 3098, 3086, 3070 (C-H_{Ar}), 1594 (C-

H_{Ar}), 1517, 1344 (NO₂). Mass spectrum (EI, 70 eV, m/z): 393 [M]⁺. Calcd for C₁₈H₁₁N₅O₂S₂: C, 54.98; H, 2.88; N, 17.86; S, 16.34, found C, 54.95; H, 2.82; N, 17.80; S, 16.30.

7-(4-methoxyphenyl)-5-(4-nitrophenyl)-2-(trifluoromethyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3db). Yellow powder (40%). m.p.: 251-253 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.67 – 8.61 (m, 2H), 8.41 – 8.36 (m, 3H), 8.34 – 8.30 (m, 2H), 7.25 – 7.20 (m, 2H), 3.90 (s, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 162.4, 159.8, 156.0, 155.6 (q, ²J_{CF} = 153.8 Hz), 149.1, 148.7, 141.4, 131.9, 129.3, 124.0, 119.4 (q, ¹J_{CF} = 272.3 Hz), 120.8, 114.2, 108.2, 55.6. ¹⁹F NMR (565 MHz, DMSO-*d*₆) δ -64.57. IR (KBr) ν/cm⁻¹: 3070, 3052 (C-H_{Ar}), 1605 (C-H_{Ar}), 1513, 1351 (NO₂). Mass spectrum (EI, 70 eV, m/z): 415 [M]⁺. Calcd for C₁₉H₁₂F₃N₅O₃: C, 54.99; H, 2.97; N, 16.89, found (%): C, 54.95; H, 2.91; N, 16.86.

***N,N*-dimethyl-4-(5-(4-nitrophenyl)-2-(trifluoromethyl)-[1,2,4]triazolo[1,5-a]pyrimidin-7-yl)aniline (3dc).** Burgundy powder (24%). m.p.: 277-279 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.66 – 8.62 (m, 2H), 8.42 – 8.39 (m, 2H), 8.39 – 8.36 (m, 2H), 8.32 (s, 1H), 6.93 – 6.89 (m, 2H), 3.09 (s, 6H). ¹³C NMR (151 MHz, DMSO) δ 159.2, 156.4, 155.4 (q, ²J_{CF} = 38.4 Hz), 152.7, 149.0, 148.9, 141.9, 131.6, 129.2, 123.9, 119.4 (q, ¹J_{CF} = 269.1 Hz), 114.4, 111.2, 106.1, 39.5. ¹⁹F NMR (565 MHz, DMSO-*d*₆) δ -64.57. IR ν/cm⁻¹: 2911, 2867, 2823 (C-H_{alif}), 1592 (C-H_{Ar}), 1523, 1347 (NO₂). Mass spectrum (EI, 70 eV, m/z): 428 [M]⁺. Calcd for C₂₀H₁₅F₃N₆O₂: C, 56.10; H, 3.56; N, 19.62, found C, 56.08; H, 3.53; N, 19.62.

7-(anthracen-9-yl)-5-(4-nitrophenyl)-2-(trifluoromethyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3dd). Yellow powder (36%). m.p.: 328-330 (decompose) °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 9.01 (s, 1H), 8.73 – 8.70 (m, 3H), 8.46 – 8.42 (m, 2H), 8.31 – 8.27 (m, 2H), 7.74 (dt, ³J_{HH} = 8.8, 1.0 Hz, 2H), 7.62 (ddd, ³J_{HH} = 8.5, 6.5, 1.1 Hz, 2H), 7.49 (ddd, ³J_{HH} = 8.9, 6.5, 1.2 Hz, 2H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 160.4, 156.1, 155.8 (q, ²J_{CF} = 38.2 Hz), 149.3, 147.4, 141.3, 130.6, 129.6, 129.5, 128.6, 127.6, 125.9, 125.2, 124.2, 122.7, 119.2 (q, ¹J_{CF} = 271.2 Hz), 114.1. ¹⁹F NMR (565 MHz, DMSO-*d*₆) δ -64.46. IR ν/cm⁻¹: 3096, 3061, 2990 (C-H_{Ar}), 1604 (C-H_{Ar}), 1526, 1348 (NO₂). Mass spectrum (EI, 70 eV, m/z): 485 [M]⁺. Calcd for C₂₆H₁₅F₃N₆O₂: C, 56.13; H, 3.58; N, 19.66, found C, 56.08; H, 3.53; N, 19.62.

5-(4-nitrophenyl)-7-(thiophen-2-yl)-2-(trifluoromethyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3df). Beige powder (31%). m.p.: 271-273 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.82 (d, ³J_{HH} = 4.0 Hz, 1H), 8.75 – 8.72 (m, 1H), 8.66 (d, ³J_{HH} = 8.6 Hz, 2H), 8.42 – 8.39 (m, 2H), 8.27 (dd, ³J_{HH} = 5.0, 1.2 Hz, 1H), 7.48 (t, ³J_{HH} = 4.5 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 159.5, 155.7, 155.4 (q, ²J_{CF} = 39.4 Hz), 149.1, 142.9, 141.4, 136.9, 134.5, 129.2, 129.2, 128.4, 124.0, 119.1 (q, ¹J_{CF} = 271.9 Hz), 105.4. ¹⁹F NMR (565 MHz, DMSO) δ -64.63. IR ν/cm⁻¹: 3078 (C-H_{Ar}), 1601 (C-H_{Ar}), 1512, 1346 (NO₂). Mass spectrum (EI, 70 eV, m/z): 391 [M]⁺. Calcd for C₁₆H₈F₃N₅O₂S: C, 49.13; H, 2.08; N, 17.96; S, 8.21, found C, 49.11; H, 2.06; N, 17.90; S, 8.19.

2-(furan-2-yl)-5-(4-nitrophenyl)-7-phenyl-[1,2,4]triazolo[1,5-a]pyrimidine (3ea). Yellow powder (11%). m.p.: 254-256 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.68 – 8.64 (m, 2H), 8.43 – 8.40 (m, 2H), 8.35 – 8.32 (m, 2H), 8.28 (s, 1H), 7.97 (dd, ³J_{HH} = 1.8, 0.8 Hz, 1H), 7.74 – 7.68 (m, 3H), 7.29 (dd, ³J_{HH} = 3.4, 0.8 Hz, 1H), 6.75 (dd, ³J_{HH} = 3.4, 1.7 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 158.2, 158.1, 156.2, 148.9, 147.8, 145.5, 145.5, 141.9, 131.8, 129.9, 129.7, 129.1, 128.6, 124.0, 112.8, 112.3, 107.5. IR ν/cm⁻¹: 3102, 3076 (C-H_{Ar}), 1607 (C-H_{Ar}), 1517, 1342 (NO₂). Mass spectrum (EI, 70

eV, m/z): 383 [M]⁺. Calcd for C₂₁H₁₃N₅O₃: C, 65.81; H, 3.46; N, 18.30, found C, 65.79; H, 3.42; N, 18.27.

2-(furan-2-yl)-7-(4-methoxyphenyl)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ed). Yellow powder (13%). m.p.: 279-281 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.61 – 8.58 (m, 2H), 8.42 – 8.38 (m, 2H), 8.38 – 8.34 (m, 2H), 8.16 (s, 1H), 7.97 (dd, ³J_{HH} = 1.8, 0.8 Hz, 1H), 7.25 (dd, ³J_{HH} = 3.4, 0.8 Hz, 1H), 7.23 – 7.19 (m, 2H), 6.73 (dd, ³J_{HH} = 3.4, 1.7 Hz, 1H), 3.90 (s, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 162.1, 158.0, 157.8, 156.3, 148.7, 147.3, 145.6, 145.5, 142.0, 131.8, 128.9, 123.9, 121.5, 114.0, 112.7, 112.2, 106.1, 55.6. IR (KBr) ν/cm⁻¹: 3147, 3098, 3073 (C-H_{Ar}), 2963, 2935, 2915 (C-H_{alif}), 1603 (C-H_{Ar}), 1525, 1346 (NO₂). Mass spectrum (EI, 70 eV, m/z): 413 [M]⁺. Calcd for C₂₂H₁₅N₅O₄: C, 63.96; H, 3.68; N, 16.99, found C, 63.92; H, 3.66; N, 16.94.

4-(2-(furan-2-yl)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidin-7-yl)-*N,N*-dimethylaniline (3ec). Burgundy powder (26%). m.p.: 299-301 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.62 (d, ³J_{HH} = 8.6 Hz, 2H), 8.47 (d, ³J_{HH} = 8.9 Hz, 2H), 8.39 (d, ³J_{HH} = 8.6 Hz, 2H), 8.13 (s, 1H), 7.99 (dd, ³J_{HH} = 1.7, 0.8 Hz, 1H), 7.29 – 7.26 (m, 1H), 6.91 (d, ³J_{HH} = 9.0 Hz, 2H), 6.75 (dd, ³J_{HH} = 3.4, 1.7 Hz, 1H), 3.09 (s, 6H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 157.8, 157.4, 156.6, 152.5, 148.7, 147.7, 145.8, 145.4, 142.5, 131.5, 128.9, 123.9, 115.2, 112.5, 112.2, 111.2, 104.4, 39.9. IR ν/cm⁻¹: 3120, 3112, 3071 (C-H_{Ar}), 2915 (C-H_{alif}), 1590 (C-H_{Ar}), 1517, 1340 (NO₂). Mass spectrum (EI, 70 eV, m/z): 426 [M]⁺. Calcd for C₂₃H₁₈N₆O₃: C, 64.80; H, 4.26; N, 19.78, found C, 64.78; H, 4.25; N, 19.71.

7-(anthracen-9-yl)-2-(furan-2-yl)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ed). Green powder (20%). m.p.: > 370 °C (decompose). ¹H NMR (600 MHz, DMSO-*d*₆) δ 9.00 (s, 1H), 8.70 – 8.66 (m, 2H), 8.47 (s, 1H), 8.43 – 8.40 (m, 2H), 8.29 (d, ³J_{HH} = 8.6 Hz, 2H), 7.76 – 7.73 (m, 1H), 7.69 (d, ³J_{HH} = 8.9 Hz, 2H), 7.64 – 7.59 (m, 2H), 7.48 (ddd, ³J_{HH} = 8.8, 6.4, 1.2 Hz, 2H), 7.16 (d, ³J_{HH} = 3.5 Hz, 1H), 6.62 (dd, ³J_{HH} = 3.5, 1.7 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 158.3, 158.2, 156.2, 148.9, 146.3, 145.4, 145.3, 141.9, 130.7, 130.1, 129.3, 129.2, 128.7, 127.5, 125.9, 125.20, 124.1, 123.7, 112.8, 112.2, 111.8. IR ν/cm⁻¹: 3113, 3062 (C-H_{Ar}), 1598 (C-H_{Ar}), 1527, 1344 (NO₂). Mass spectrum (EI, 70 eV, m/z): 483 [M]⁺. Calcd for C₂₉H₁₇N₅O₃: C, 72.06; H, 3.56; N, 14.50, found C, 72.04; H, 3.54; N, 14.49.

2,7-di(furan-2-yl)-5-(4-nitrophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ee). Brown powder (14%). m.p.: 272-274 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.56 (d, ³J_{HH} = 8.4 Hz, 2H), 8.37 (d, ³J_{HH} = 8.6 Hz, 2H), 8.28 (s, 1H), 8.22 (d, ³J_{HH} = 3.6 Hz, 1H), 8.19 (s, 1H), 8.01 (d, ³J_{HH} = 1.9 Hz, 1H), 7.37 (d, ³J_{HH} = 3.4 Hz, 1H), 7.00 (dd, ³J_{HH} = 3.6, 1.7 Hz, 1H), 6.77 (dd, ³J_{HH} = 3.4, 1.7 Hz, 1H). ¹³C NMR (151 MHz, DMSO) δ 158.4, 157.5, 155.8, 148.8, 148.3, 145.7, 145.4, 142.6, 141.8, 136.6, 128.9, 124.0, 120.9, 113.9, 113.0, 112.3, 101.9. IR ν/cm⁻¹: 3142, 3120, 3106, 3079 (C-H_{Ar}), 1613 (C-H_{Ar}), 1518, 1343 (NO₂). Mass spectrum (EI, 70 eV, m/z): 373 [M]⁺. Calcd for C₁₉H₁₁N₅O₄: C, 61.15; H, 2.9; N, 18.79, found C, 61.13; H, 2.97; N, 18.76.

2-(furan-2-yl)-5-(4-nitrophenyl)-7-(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ef). Orange powder (20%). m.p.: 318-320 °C (decompose). ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.81 (dd, ³J_{HH} = 3.9, 1.2 Hz, 1H), 8.66 – 8.61 (m, 2H), 8.56 (s, 1H), 8.41 – 8.37 (m, 2H), 8.24 (dd, ³J_{HH} = 5.0, 1.2 Hz, 1H), 8.02 (d, ³J_{HH} = 2.1 Hz, 1H), 7.47 (dd, ³J_{HH} = 5.0, 3.9 Hz, 1H), 7.32 (d, ³J_{HH} = 3.4 Hz, 1H), 6.78 (dd, ³J_{HH} = 3.4,

1.7 Hz, 1H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 157.8, 157.4, 155.8, 148.8, 145.7, 145.4, 142.0, 141.4, 136.2, 133.8, 129.88, 128.9, 128.2, 123.9, 113.0, 112.3, 103.4. IR ν/cm⁻¹: 3152, 3121, 3113 (C-H_{Ar}), 1606 (C-H_{Ar}), 1520, 1340 (NO₂). Mass spectrum (EI, 70 eV, m/z): 389 [M]⁺. Calcd for C₁₉H₁₁N₅O₃S: C, 58.63; H, 2.88; N, 18.01; S, 8.25, found C, 58.61; H, 2.85; N, 17.99; S, 8.23.

7-(4-methoxyphenyl)-5-(4-nitrophenyl)-2-(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (3fb). Orange powder (36%). m.p.: 272-274 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.53 (d, ³J_{HH} = 8.4 Hz, 2H), 8.37 (d, ³J_{HH} = 8.4 Hz, 2H), 8.30 (d, ³J_{HH} = 8.4 Hz, 2H), 8.07 (s, 1H), 7.83 (d, ³J_{HH} = 3.6 Hz, 1H), 7.78 (d, ³J_{HH} = 4.9 Hz, 1H), 7.22 (t, ³J_{HH} = 4.3 Hz, 1H), 7.17 (d, ³J_{HH} = 8.4 Hz, 2H), 3.89 (s, 3H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 162.1, 161.2, 157.4, 156.3, 148.6, 146.9, 141.8, 133.0, 131.8, 129.6, 128.8, 128.4, 128.3, 123.8, 121.4, 114.0, 105.9, 55.5. IR ν/cm⁻¹: 3098, 3078 (C-H_{Ar}), 2961, 2916, 2837 (C-H_{alif}), 1604 (C-H_{Ar}), 1523, 1347 (NO₂). Mass spectrum (EI, 70 eV, m/z): 429 [M]⁺. Calcd for C₂₂H₁₅N₅O₃S: C, 61.566; H, 3.56; N, 16.35; S, 7.49, found C, 61.53; H, 3.52; N, 16.31; S, 7.47.

***N,N*-dimethyl-4-(5-(4-nitrophenyl)-2-(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidin-7-yl)aniline (3fc)**. m.p.: 304-306 °C. Burgundy Powder (26%). m.p.: 304-306 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.67 – 8.64 (m, 2H), 8.53 – 8.49 (m, 2H), 8.44 – 8.41 (m, 2H), 8.19 (s, 1H), 7.93 (dd, ³J_{HH} = 3.4, 1.0 Hz, 1H), 7.82 (dd, ³J_{HH} = 5.0, 1.2 Hz, 1H), 7.28 (dd, ³J_{HH} = 4.9, 3.6 Hz, 1H), 6.95 (dd, ³J_{HH} = 9.4, 2.5 Hz, 2H), 3.11 (s, 6H). ¹³C NMR (151 MHz, DMSO-*d*₆) δ 161.5, 157.7, 157.2, 153.0, 149.2, 148.0, 143.0, 133.7, 132.0, 130.1, 129.4, 128.9, 128.8, 124.4, 115.7, 111.7, 104.9, 40.3. IR ν/cm⁻¹: 3069 (C-H_{Ar}), 2914 (C-H_{alif}), 1591 (C-H_{Ar}), 1515, 1341 (NO₂). Mass spectrum (EI, 70 eV, m/z): 442 [M]⁺. Calcd for C₂₃H₁₈N₆O₂S: C, 62.47; H, 4.13; N, 19.01; S, 7.27, found C, 62.43; H, 4.10; N, 18.99; S, 7.25.

5-(4-nitrophenyl)-2,7-di(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (3ff). Orange powder (58%). m.p.: 328-330 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.88 (dd, ³J_{HH} = 3.9, 1.2 Hz, 1H), 8.72 – 8.69 (m, 2H), 8.65 (s, 1H), 8.46 (dd, ³J_{HH} = 9.1, 2.3 Hz, 2H), 8.29 (dd, ³J_{HH} = 4.9, 1.1 Hz, 1H), 8.00 (dd, ³J_{HH} = 3.6, 1.2 Hz, 1H), 7.87 (dd, ³J_{HH} = 5.0, 1.2 Hz, 1H), 7.52 (dd, ³J_{HH} = 5.0, 3.9 Hz, 1H), 7.32 (dd, ³J_{HH} = 5.0, 3.6 Hz, 1H). ¹³C NMR (151 MHz, DMSO) δ 161.2, 157.5, 156.1, 148.9, 142.2, 141.3, 136.3, 133.8, 132.9, 130.1, 130.0, 129.0, 128.8, 128.6, 128.3, 124.0, 103.6. IR ν/cm⁻¹: 3091, 3069 (C-H_{Ar}), 1599 (C-H_{Ar}), 1517, 1344 (NO₂). Mass spectrum (EI, 70 eV, m/z): 405 [M]⁺. Calcd for C₁₉H₁₁N₅O₂S₂: C, 56.29; H, 2.73; N, 17.27; S, 15.81, found C, 56.29; H, 2.73; N, 17.27; S, 15.81.

7-(4-methoxyphenyl)-5-(4-nitrophenyl)-2-phenyl-[1,2,4]triazolo[1,5-a]pyrimidine (3gb). Yellow powder (18%). m.p.: 256-258 °C. ¹H NMR (600 MHz, DMSO-*d*₆) δ 8.64 – 8.60 (m, 2H), 8.50 – 8.45 (m, 2H), 8.38 – 8.35 (m, 2H), 8.25 – 8.22 (m, 2H), 8.19 (s, 1H), 7.59 – 7.53 (m, 3H), 7.25 – 7.21 (m, 2H), 3.91 (s, 3H). ¹³C NMR (151 MHz, DMSO) δ 164.9, 162.1, 157.5, 156.6, 148.7, 147.1, 142.1, 131.9, 130.7, 130.2, 128.9, 128.9, 126.9, 123.9, 121.6, 114.1, 106.0, 55.6. IR ν/cm⁻¹: 3111, 3041 (C-H_{Ar}), 2976, 2936, 2842 (C-H_{alif}), 1607 (C-H_{Ar}), 1544, 1350 (NO₂). Mass spectrum (EI, 70 eV, m/z): 423 [M]⁺. Calcd for C₂₄H₁₇N₅O₃: C, 68.09; H, 4.06; N, 16.58, found C, 68.08; H, 4.05; N, 16.54.

2-substituted-7-(het)aryl-5(4-nitrophenyl)-6-nitro-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-a]pyrimidine (general method) Imines **1** (1 mmol) and β-nitroethylenes **2** (1

mmol) was suspended in 5 mL of EtOH (95%) in a flask, treated with NEt_3 (1 mmol), and refluxed for 2-8 h. The hot solution was filtered precipitate was washed with EtOH (95%) 3 mL and dried to a constant weight. The filtrate was evaporated under reduced pressure to dryness oil, treated with hot EtOH (95%) 3-5 mL, filtered and dried in air to a constant weight in order to extract an additional amount of the target product.

7-(4-methoxyphenyl)-2-(methylthio)-6-nitro-5-(4-nitrophenyl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4ab). White powder (40%). m.p.: 267-269 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.52 (d, $^3J_{\text{HH}} = 2.3$ Hz, 1H), 8.12 - 8.06 (m, 2H), 7.65 - 7.61 (m, 2H), 7.04 - 7.00 (m, 2H), 6.75 - 6.72 (m, 2H), 5.93 (d, $^3J_{\text{HH}} = 6.2$ Hz, 1H), 5.89 (t, $^3J_{\text{HH}} = 6.4$ Hz, 1H), 5.48 (dd, $^3J_{\text{HH}} = 6.8$, 2.3 Hz, 1H), 3.65 (s, 3H), 2.40 (s, 3H). ^{13}C NMR (151 MHz, $\text{DMSO-}d_6$) δ 159.2, 158.4, 153.5, 147.4, 143.6, 128.8, 128.7, 125.9, 123.4, 113.6, 88.0, 60.1, 56.0, 55.0, 13.5. IR (KBr) ν/cm^{-1} : 3181, 3149, 3109 (C-H_{Ar}), 3014, 2991, 2932 (CH₃), 1619 (C-H_{Ar}), 1564, 1523, 1356 (NO₂). Mass spectrum (EI, 70 eV, m/z): 442 [M]⁺. Calcd for C₁₉H₁₈N₆O₅S: C, 51.59; H, 4.16; N, 19.01; S, 7.28, found C, 51.58; H, 4.10; N, 18.99; S, 7.25.

7-(anthracen-9-yl)-2-(methylthio)-6-nitro-5-(4-nitrophenyl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4ad). Brown powder (14%). m.p.: 284-286 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.78 (s, 1H), 8.70 (s, 1H), 8.48 (d, $^3J_{\text{HH}} = 9.2$ Hz, 1H), 8.30-8.24 (m, 2H), 8.19 (dd, $^3J_{\text{HH}} = 7.6$, 1.9 Hz, 1H), 8.15 (dd, $^3J_{\text{HH}} = 8.7$, 1.3 Hz, 1H), 7.91-7.87 (m, 2H), 7.69 (d, $^3J_{\text{HH}} = 10.1$ Hz, 1H), 7.65 (ddd, $^3J_{\text{HH}} = 9.1$, 6.5, 1.3 Hz, 1H), 7.62-7.51 (m, 4H), 5.87 (t, $^3J_{\text{HH}} = 9.9$ Hz, 1H), 5.68 (d, $^3J_{\text{HH}} = 9.8$ Hz, 1H), 2.24 (s, 3H). ^{13}C NMR (151 MHz, $\text{DMSO-}d_6$) δ 158.4, 154.0, 148.3, 142.3, 131.5, 131.2, 130.8, 130.7, 130.0, 129.6, 129.4, 128.7, 127.7, 127.6, 125.3, 125.1, 124.0, 123.0, 122.5, 122.4, 88.8, 57.9, 56.5, 13.7. IR (KBr) ν/cm^{-1} : 3188, 3057 (C-H_{Ar}), 2930, 2878, 2860 (CH₃), 1615 (C-H_{Ar}), 1562, 1519, 1347 (NO₂). Mass spectrum (EI, 70 eV, m/z): 512 [M]⁺. Calcd for C₂₆H₂₀N₆O₄S: C, 60.96; H, 3.96; N, 16.43; S, 6.29, found C, 60.93; H, 3.93; N, 16.40; S, 6.26.

2-(methylthio)-6-nitro-5-(4-nitrophenyl)-7-(thiophen-2-yl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4af). White powder (24%). m.p.: 257-259 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.60 (d, $^3J_{\text{HH}} = 2.4$ Hz, 1H), 8.11 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 7.64 (d, $^3J_{\text{HH}} = 8.7$ Hz, 2H), 7.40 (dd, $^3J_{\text{HH}} = 5.0$, 1.1 Hz, 1H), 6.87 (d, $^3J_{\text{HH}} = 3.5$ Hz, 1H), 6.79 (dd, $^3J_{\text{HH}} = 4.9$, 3.6 Hz, 1H), 6.31 (d, $^3J_{\text{HH}} = 5.4$ Hz, 1H), 6.01 (t, $^3J_{\text{HH}} = 5.8$ Hz, 1H), 5.54 (d, $^3J_{\text{HH}} = 3.7$ Hz, 1H), 2.43 (s, 3H). ^{13}C NMR (151 MHz, $\text{DMSO-}d_6$) δ 158.6, 153.1, 147.3, 143.7, 136.7, 128.6, 127.6, 127.3, 126.6, 123.42, 87.4, 56.5, 55.9, 13.4. IR (KBr) (ν/cm^{-1}): 3205, 3173, 3139 (C-H_{Ar}), 3028, 2929, 2879 (C-H_{alif}), 1615 (C-H_{Ar}), 1566, 1524, 1325 (NO₂). Mass spectrum (EI, 70 eV, m/z): 418 [M]⁺. Calcd for C₁₆H₁₄N₆O₄S₂: C, 45.93; H, 3.37; N, 20.08; S, 15.32, found C, 45.93; H, 3.37; N, 20.08; S, 15.32.

7-(4-methoxyphenyl)-6-nitro-5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4cb). White powder (18%). m.p.: 236-238 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.60 (d, $^3J_{\text{HH}} = 2.1$ Hz, 1H), 8.11 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 7.65 (d, $^3J_{\text{HH}} = 8.7$ Hz, 2H), 7.06 (d, $^3J_{\text{HH}} = 8.7$ Hz, 2H), 6.76 (d, $^3J_{\text{HH}} = 8.8$ Hz, 2H), 5.94 (d, $^3J_{\text{HH}} = 6.5$ Hz, 1H), 5.90 (t, $^3J_{\text{HH}} = 6.8$ Hz, 1H), 5.49 (d, $^3J_{\text{HH}} = 9.3$ Hz, 1H), 3.81 (t, $^3J_{\text{HH}} = 3.1$ Hz, 2H), 3.66 (s, 3H), 3.17 (t, $^3J_{\text{HH}} = 2.6$ Hz, 1H). ^{13}C NMR (151 MHz, DMSO-

d_6) δ 159.3, 156.1, 153.6, 147.5, 143.5, 128.9, 128.8, 125.7, 123.4, 113.7, 88.1, 80.3, 73.7, 60.3, 56.1, 55.1, 19.1. IR (KBr) ν/cm^{-1} : 3386 (C≡CH), 3283, 3264, 3209 (C-H_{Ar}), 2971, 2938, 2841 (C-H_{alif}), 1616 (C-H_{Ar}), 1563, 1530, 1324 (NO₂). Mass spectrum (EI, 70 eV, m/z): 446 [M]⁺. Calcd for C₂₁H₁₈N₆O₅S: C, 54.09; H, 3.91; N, 18.05; S, 6.90, found C, 54.07; H, 3.89; N, 18.02; S, 6.87.

7-(anthracen-9-yl)-6-nitro-5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4cd). Brown powder (77%). m.p.: 244-246 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.84 (s, 1H), 8.79 (s, 1H), 8.50 (d, $^3J_{\text{HH}} = 9.2$ Hz, 1H), 8.31 - 8.26 (m, 2H), 8.19 (dd, $^3J_{\text{HH}} = 7.9$, 1.9 Hz, 1H), 8.15 (dd, $^3J_{\text{HH}} = 8.6$, 1.3 Hz, 1H), 7.95 - 7.90 (m, 2H), 7.75 (d, $^3J_{\text{HH}} = 10.1$ Hz, 1H, CH), 7.69 - 7.63 (m, 1H), 7.61 - 7.54 (m, 3H), 7.53 (d, $^3J_{\text{HH}}^3J_{\text{HH}} = 8.5$ Hz, 1H), 5.91 (t, $^3J_{\text{HH}} = 9.9$ Hz, 1H), 5.74 (d, $^3J_{\text{HH}} = 9.8$ Hz, 1H), 3.71 (d, $^3J_{\text{HH}} = 2.6$ Hz, 2H), 3.05 (t, $^3J_{\text{HH}} = 2.6$ Hz, 1H). ^{13}C NMR (151 MHz, $\text{DMSO-}d_6$) δ 156.2, 154.0, 148.3, 142.2, 131.5, 131.2, 130.8, 130.7, 130.0, 129.7, 129.4, 128.81, 127.7, 127.6, 125.3, 125.2, 124.0, 123.0, 122.5, 122.3, 88.6, 80.1, 73.7, 57.9, 56.6, 19.3. IR ν/cm^{-1} : 3306 (C≡CH), 3267, 3169, 3063 (C-H_{Ar}), 2915, 2873, 2824 (C-H_{alif}), 1589 (C-H_{Ar}), 1539, 1343 (NO₂). Mass spectrum (EI, 70 eV, m/z): 536 [M]⁺. Calcd for C₂₈H₂₀N₆O₄S: C, 62.68; H, 3.76; N, 15.66; S, 5.98, found C, 62.68; H, 3.76; N, 15.66; S, 5.98.

6-nitro-5-(4-nitrophenyl)-2-(prop-2-yn-1-ylthio)-7-(thiophen-2-yl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4cf). Yellow powder (45%). m.p.: 240-242 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.69 (d, $^3J_{\text{HH}} = 2.4$ Hz, 1H, NH), 8.16 - 8.09 (m, 2H), 7.67 (d, $^3J_{\text{HH}} = 8.7$ Hz, 2H), 7.42 (dd, $^3J_{\text{HH}} = 5.1$, 1.3 Hz, 1H), 6.91 (dt, $^3J_{\text{HH}} = 3.6$, 1.1 Hz, 1H), 6.81 (dd, $^3J_{\text{HH}} = 5.0$, 3.6 Hz, 1H), 6.34 (d, $^3J_{\text{HH}} = 5.8$ Hz, 1H, CH), 6.02 (t, $^3J_{\text{HH}} = 6.1$ Hz, 1H, CH), 5.55 (dd, $^3J_{\text{HH}} = 6.5$, 2.4 Hz, 1H, CH), 3.84 (t, $^3J_{\text{HH}} = 2.8$ Hz, 2H), 3.17 (t, $^3J_{\text{HH}} = 2.6$ Hz, 1H). ^{13}C NMR (151 MHz, $\text{DMSO-}d_6$) δ 156.4, 153.2, 147.4, 143.5, 136.4, 128.7, 127.9, 127.4, 126.6, 123.4, 87.5, 80.2, 73.8, 56.6, 56.1, 19.1. IR ν/cm^{-1} : 3273 (C≡CH), 3212, 3152 (C-H_{Ar}), 2922, 2868 (C-H_{alif}), 1615 (C-H_{Ar}), 1564, 1526, 1348 (NO₂). Mass spectrum (EI, 70 eV, m/z): 392 [M]⁺. Calcd for C₁₈H₁₄N₆O₄S₂: C, 48.89; H, 3.20; N, 19.00; S, 14.51, found C, 48.86; H, 3.19; N, 18.99; S, 14.49.

7-(anthracen-9-yl)-2-(furan-2-yl)-6-nitro-5-(4-nitrophenyl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4ed). Yellow powder (80%). m.p.: 289-291 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.81 (s, 1H), 8.75 (s, 1H), 8.54 - 8.50 (m, 1H), 8.31 - 8.28 (m, 2H), 8.22 - 8.19 (m, 1H), 8.19 - 8.16 (m, 1H), 7.95 - 7.91 (m, 2H), 7.80 (d, $^3J_{\text{HH}} = 10.1$ Hz, 1H), 7.68 (ddd, $^3J_{\text{HH}} = 9.1$, 6.5, 1.3 Hz, 1H), 7.61 - 7.58 (m, 1H), 7.58 - 7.53 (m, 4H), 6.71 (dd, $^3J_{\text{HH}} = 3.4$, 0.8 Hz, 1H), 6.47 (dd, $^3J_{\text{HH}} = 3.4$, 1.7 Hz, 1H), 5.94 (t, $^3J_{\text{HH}} = 10.0$ Hz, 1H), 5.74 (d, $^3J_{\text{HH}} = 9.8$ Hz, 1H). ^{13}C NMR (151 MHz, $\text{DMSO-}d_6$) δ 153.8, 152.4, 148.3, 146.2, 143.3, 142.3, 131.6, 131.3, 130.8, 130.8, 130.0, 129.7, 129.4, 128.7, 127.8, 127.7, 125.3, 125.1, 124.0, 122.9, 122.5, 122.5, 111.4, 108.8, 88.8, 58.0, 56.5. IR ν/cm^{-1} : 3349 (NH), 3067, 3011 (C-H_{Ar}), 1629 (C-H_{Ar}), 1564, 1516, 1324 (NO₂). Mass spectrum (EI, 70 eV, m/z): 532 [M]⁺. Calcd for C₂₉H₂₀N₆O₅: C, 65.46; H, 3.81; N, 15.79, found C, 65.41; H, 3.79; N, 15.78.

2-(furan-2-yl)-6-nitro-5-(4-nitrophenyl)-7-(thiophen-2-yl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4ff). White powder (40%). m.p.: 254-256 °C. ^1H NMR (600 MHz, $\text{DMSO-}d_6$) δ 8.66 (s, 1H, NH), 8.27 (d, $^3J_{\text{HH}} = 8.4$ Hz, 2H), 7.76 (s, 1H), 7.65 (t, $^3J_{\text{HH}} = 5.8$ Hz, 3H), 7.29 (d, $^3J_{\text{HH}} = 3.6$ Hz, 1H), 7.13 (t, $^3J_{\text{HH}} = 4.3$ Hz, 1H), 6.86 (d,

$^3J_{HH} = 3.3$ Hz, 1H), 6.64 – 6.60 (m, 1H), 6.58 (s, 1H), 5.67 – 5.61 (m, 1H), 5.36 (d, $^3J_{HH} = 3.2$ Hz, 1H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 153.0, 152.5, 147.6, 146.5, 143.6, 142.8, 139.9, 128.6, 127.5, 127.2, 127.0, 123.5, 111.6, 109.0, 86.0, 55.2, 52.6. IR ν/cm^{-1} : 3205, 3111, 3068 (C-H_{Ar}), 2934, 2866 (C-H_{alif}), 1616 (C-H_{Ar}), 1563, 1530, 1324 (NO₂). Mass spectrum (EI, 70 eV, m/z): 438 [M]⁺. Calcd for C₁₉H₁₄N₆O₅S: C, 52.07; H, 3.28; N, 19.19; S, 7.39, found C, 52.05; H, 3.22; N, 19.17; S, 7.31.

7-(anthracen-9-yl)-6-nitro-5-(4-nitrophenyl)-2-(thiophen-2-yl)-4,5,6,7-tetrahydro-[1,2,4]triazolo[1,5-a]pyrimidine (4fd). Yellow powder (92%). m.p.: 287–289 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 8.81 (s, 1H), 8.76 (s, 1H), 8.52 (d, $^3J_{HH} = 9.2$ Hz, 1H), 8.31 – 8.26 (m, 2H), 8.22 – 8.19 (m, 1H), 8.18 (d, $^3J_{HH} = 8.4$ Hz, 1H), 7.94 – 7.89 (m, 2H), 7.79 (d, $^3J_{HH} = 10.1$ Hz, 1H), 7.68 (ddd, $^3J_{HH} = 9.1, 6.5, 1.4$ Hz, 1H), 7.60 – 7.53 (m, 4H), 7.44 (dd, $^3J_{HH} = 5.0, 1.2$ Hz, 1H), 7.29 (dd, $^3J_{HH} = 3.7, 1.2$ Hz, 1H), 6.98 (dd, $^3J_{HH} = 5.0, 3.7$ Hz, 1H), 5.90 (t, $^3J_{HH} = 9.9$ Hz, 1H), 5.72 (d, $^3J_{HH} = 9.8$ Hz, 1H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 155.4, 154.0, 148.3, 142.3, 134.0, 131.5, 131.3, 130.9, 130.8, 130.1, 129.6, 129.4, 128.7, 127.8, 127.7, 127.7, 126.6, 125.5, 125.4, 125.2, 124.0, 123.0, 122.6, 89.0, 58.0, 56.6. IR ν/cm^{-1} : 3345 (NH), 3057 (C-H_{alif}), 1609 (C-H_{Ar}), 1561, 1512, 1343 (NO₂). Mass spectrum (EI, 70 eV, m/z): 548 [M]⁺. Calcd for C₂₉H₂₀N₆O₄S: C, 63.53; H, 3.69; N, 15.38; S, 5.88, found C, 63.50; H, 3.67; N, 15.32; S, 5.84.

7-(het)aryl-2-(methylthio)-5-(pyridin-3-yl)-[1,2,4]triazolo[1,5-a]pyrimidines (7a-c) (general methods). Imines **5** (1 mmol) and β -nitroethylenes **2a-c** (1 mmol) was suspended in 5 mL of solvent MeCN in a flask, treated with DBU (1 mmol), and refluxed for 8 h. The hot solution was filtered, precipitate was washed with MeCN 3–5 mL and dried to a constant weight. The filtrate was evaporated under reduced pressure to dryness oil, treated with hot MeCN 3–5 mL, filtered and dried in air to a constant weight in order to extract an additional amount of the target product.

7-(4-methoxyphenyl)-2-(methylthio)-5-(pyridin-3-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (7a). Beige powder (14%). m.p.: 239–241 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 9.52 (dd, $^3J_{HH} = 2.4, 0.9$ Hz, 1H), 8.77 (dd, $^3J_{HH} = 4.7, 1.6$ Hz, 1H), 8.71 (ddd, $^3J_{HH} = 8.0, 2.4, 1.6$ Hz, 1H), 8.42–8.38 (m, 2H), 8.16 (s, 1H), 7.63 (ddd, $^3J_{HH} = 8.0, 4.8, 0.9$ Hz, 1H), 7.23–7.18 (m, 2H), 3.90 (s, 3H), 2.70 (s, 3H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 167.9, 162.0, 157.7, 156.5, 151.6, 148.8, 146.4, 135.1, 131.9, 131.8, 123.9, 121.6, 114.1, 105.0, 55.6, 13.3. IR (KBr) ν/cm^{-1} : 3081, 3016 (C-H_{Ar}), 2995, 2936, 2843 (C-H_{alif}), 1603 (C-H_{Ar}). Mass spectrum (EI, 70 eV, m/z): 349 [M]⁺. Calcd for C₁₈H₁₅N₅OS: C, 61.89; H, 4.36; N, 20.08; S, 9.19, found C, 61.87; H, 4.33; N, 20.04; S, 9.18.

7-(anthracen-9-yl)-2-(methylthio)-5-(pyridin-3-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (7b). Brown powder (40%). m.p.: 320–322 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 9.54 (s, 1H), 8.97 (s, 1H), 8.76 (dd, $^3J_{HH} = 4.8, 1.6$ Hz, 1H), 8.74 (dd, $^3J_{HH} = 8.2, 2.1$ Hz, 1H), 8.36 (d, $^3J_{HH} = 1.4$ Hz, 1H), 8.27 (d, $^3J_{HH} = 8.5$ Hz, 2H), 7.66–7.58 (m, 5H), 7.49 (dd, $^3J_{HH} = 8.7, 6.7$ Hz, 2H), 2.48 (s, 3H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 168.3, 157.8, 156.2, 151.8, 148.9, 145.1, 135.2, 131.7, 130.6, 130.1, 129.2, 128.7, 127.5, 125.9, 125.1, 124.0, 123.7, 110.5, 13.3. IR (KBr) ν/cm^{-1} : 3082, 3049, 3005 (C-H_{Ar}), 2929, 2852 (C-H_{alif}), 1603 (C-H_{Ar}). Mass spectrum (EI, 70 eV, m/z): 419 [M]⁺. Calcd for C₂₅H₁₇N₅S: C, 71.60; H, 4.10; N, 16.70; S, 7.68, found: C, 71.58; H, 4.08; N, 16.69; S, 7.64.

2-(methylthio)-5-(pyridin-3-yl)-7-(thiophen-2-yl)-[1,2,4]triazolo[1,5-a]pyrimidine (7c). Grey powder (15%). m.p.: 214–216 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 9.55 (dd, $^3J_{HH} = 2.4, 0.8$ Hz, 1H), 8.80 (dd, $^3J_{HH} = 3.9, 1.2$ Hz, 1H), 8.78 (dd, $^3J_{HH} = 4.7, 1.6$ Hz, 1H), 8.72 (ddd, $^3J_{HH} = 8.1, 2.4, 1.7$ Hz, 1H), 8.54 (s, 1H), 8.23 (dd, $^3J_{HH} = 5.0, 1.2$ Hz, 1H), 7.64 (ddd, $^3J_{HH} = 8.0, 4.7, 0.9$ Hz, 1H), 7.47 (dd, $^3J_{HH} = 5.0, 3.9$ Hz, 1H), 2.77 (s, 3H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 168.1, 157.4, 156.1, 151.6, 148.8, 140.4, 136.1, 135.0, 133.6, 131.9, 129.8, 128.2, 123.9, 102.1, 13.5. IR ν/cm^{-1} : 3083, 3067, 3019 (C-H_{Ar}), 2924 (C-H_{alif}), 1594 (C-H_{Ar}). Mass spectrum (EI, 70 eV, m/z): 325 [M]⁺. Calcd for C₁₅H₁₁N₅S₂: C, 55.40; H, 3.45; N, 21.56; S, 19.73, found C, 55.37; H, 3.41; N, 21.52; S, 19.70.

7-(4-methoxyphenyl)-2-(methylthio)-5-(pyridin-3-yl)-[1,2,4]triazolo[1,5-a]pyrimidin-6-amine (8). Tetrahydropyrimidine (**6a**) (0.001 mol) was suspended in 5 ml of MeCN in a flask, then thiophenol (0.001 mol) was added, then DBU (0.001 mol) was added, reflux for 9 hours, then left overnight at room temperature. The resulting suspension was filtered, the precipitate was washed with 3–5 ml MeCN until the color disappeared and dried to obtain a constant mass. The filtrate was evaporated at reduced pressure to dry the oil, treated with 3–5 ml MeCN, filtered and dried in air until a constant weight was obtained in order to extract an additional amount of the target product. Next, the crude product was purified by chromatographically on sili-cagel, eluent ethylacetate:hexane (1:0.5). Yellow powder (10%). m.p.: 238–240 °C. ^1H NMR (600 MHz, DMSO- d_6) δ 9.00 (dd, $^3J_{HH} = 2.3, 0.9$ Hz, 1H), 8.72 (dd, $^3J_{HH} = 4.8, 1.7$ Hz, 1H), 8.23 (dt, $^3J_{HH} = 7.9, 2.0$ Hz, 1H), 7.70 – 7.65 (m, 2H), 7.59 (ddd, $^3J_{HH} = 7.9, 4.8, 0.9$ Hz, 1H), 7.22 – 7.16 (m, 2H), 4.60 (s, 2H), 3.87 (s, 3H), 2.57 (s, 3H). ^{13}C NMR (151 MHz, DMSO- d_6) δ 165.12, 160.56, 151.07, 150.82, 150.36, 149.28, 136.35, 132.95, 131.47, 130.06, 129.32, 123.64, 119.78, 114.55, 55.39, 13.44. IR ν/cm^{-1} : 3416, 3329 (NH₂), 3065, 3023 (C-H_{Ar}), 2938, 2832 (CH₃), 1608 (C-H_{Ar}). Mass spectrum (EI, 70 eV, m/z): 364 [M]⁺. Calcd for C₁₈H₁₆N₆OS: C, 59.33; H, 4.43; N, 23.06; S, 8.80, found C, 59.33; H, 4.43; N, 23.06; S, 8.80.

Supplementary materials

This manuscript contains supplementary materials, which are available on the corresponding online page

Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Conflict of interest

The authors declare no conflict of interests.

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