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Synthesis of New Polyfunctional 5,6,7,8-Tetrahydroimidazo-[1,5-*c*]pyrimidin-5-ones by the Aza-Wittig Reaction Followed by Intramolecular Cyclization and 1,3-Prototropic Shift

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Abstract—Ethyl 2-oxo-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylates reacted with organic isocyanates according to the aza-Wittig pattern, and the subsequent intramolecular ring closure and 1,3-H shift resulted in the formation of ethyl 3-alkyl(aryl)amino-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylates.

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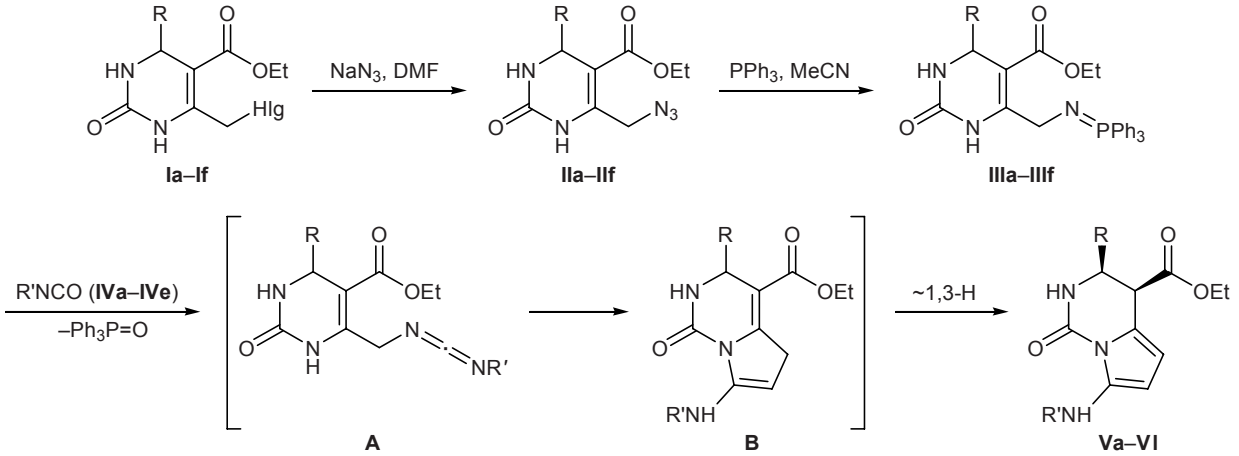
Imidazo[1,5-*c*]pyrimidines may be regarded as analogs of purines which constitute a pharmacologically important class of fused systems. One of the most familiar derivatives of this series is alkaloid Zapotidine (6-methylimidazo[1,5-*c*]tetrahydropyrimidine-5-thione) [1, 2] isolated from the seeds of *Casimiroa edulis* Llave *et* Lex. (*Rutaceae*); it was found to exhibit hypnotic, sedative, and hypotensive effects. Tetrahydroimidazo[1,5-*c*]pyrimidin-5-one derivatives were identified as a new class of adenosine antagonists [3]. A semisynthetic glycopeptide antibiotic containing a tetrahydroimidazo[1,5-*c*]pyrimidine fragment was shown to be more active than the known antibiotic Vancomycin [4].

Imidazo[1,5-*c*]pyrimidines can be synthesized by intramolecular cyclization of histamine with the use of *N*,*N*′-carbonyldiimidazole [5, 6], *N*,*N*′-thiocarbonyldiimidazole [7], bis(4-nitrophenyl) carbonate [8], and *S*-phenyl chlorothiocarbonate [9] as condensing agents. However, these procedures make it possible to obtain only the corresponding 5-oxo or 5-thioxo derivatives. Published data on imidazo[1,5-*c*]pyrimidines having functional substituents in the imidazole or pyrimidine ring are very limited. Chivikas and Hodges [10] reported on the synthesis of methyl (7*S*)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-7-carboxylate from L-histidine methyl ester [10], and imidazo[1,5-*c*]-pyrimidines containing chlorine atoms or methylsulfanyl groups in the pyrimidine ring were prepared on the basis of the corresponding 6-aminomethyl-substituted pyrimidines [11]. Nevertheless, imidazo[1,5-*c*]-pyrimidine derivatives with functional groups capable of undergoing further transformations may be regarded as quite promising building blocks for the design of small libraries of potential biologically active compounds. Therefore, the present work was aimed at developing a synthetic approach to new imidazo[1,5-*c*]-pyrimidine derivatives having an alkyl(aryl)amino group in position *3* and ester moiety in position *8*. The approach was based on annulation to pyrimidine ring of exocyclic carbodiimide moiety generated *in situ* via the aza-Wittig reaction.

As starting compounds we selected ethyl 3-substituted 6-halomethyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylates **Ia**–**If**. These accessible compounds are widely used in both organic synthesis [12] and biomedical studies [13]. 6-Bromomethylpyrimidines **Ia**–**Ic** were synthesized according to the procedure described in [14], by bromination of the corresponding 6-methylpyrimidines in chloroform. To obtain 6-chloromethylpyrimidines **Id**–**If**, we have developed by analogy with the Biginelli reaction [15] a one-step procedure utilizing ethyl 4-chloro-3-oxobutanoate as β-dicarbonyl component. As a result, compounds **Id**–**If** were prepared in 78–84% yield by heating a mixture of aromatic aldehyde, ethyl 4-chloro-3-oxobutanoate, and

Scheme 1.



I, Hlg = Br, R = H (**a**), Me (**b**), Ph (**c**); Hlg = Cl, R = 4-ClC_6H_4 (**d**), 4-$MeOC_6H_4$ (**e**), 4-*t*-BuC_6H_4 (**f**); **II**, **III**, R = H (**a**), Me (**b**), Ph (**c**), 4-ClC_6H_4 (**d**), 4-$MeOC_6H_4$ (**e**), 4-*t*-BuC_6H_4 (**f**); **IV**, R′ = *cyclo*-C_6H_{11} (**a**), 4-ClC_6H_4 (**b**), 3-$F_3CC_6H_4$ (**c**), 4-$MeOC_6H_4$ (**d**), 3-Cl-4-MeC_6H_3 (**e**); **V**, R = H, R′ = 4-ClC_6H_4 (**a**); R = Me, R′ = 4-ClC_6H_4 (**b**), 4-$MeOC_6H_4$ (**c**); R = Ph, R′ = *cyclo*-C_6H_{11} (**d**), 4-ClC_6H_4 (**e**); R = 4-ClC_6H_4, R′ = *cyclo*-C_6H_{11} (**f**), 4-ClC_6H_4 (**g**), 4-$MeOC_6H_4$ (**h**); R = R′ = 4-$MeOC_6H_4$ (**i**); R = 4-*t*-BuC_6H_4, R′ = 3-$F_3CC_6H_4$ (**j**), 4-$MeOC_6H_4$ (**k**), 3-Cl-4-MeC_6H_3 (**l**).

urea at a ratio of 1:1:1.5 in acetic acid at 45–50°C over a period of 48 h.

Kappe [16] reported on the replacement of the bromine atom in 6-bromomethylpyrimidine **Ic** by azido group by treatment with sodium azide in HMPA (3 days). We found that both bromo- and chloromethyl derivatives **Ia**–**If** smoothly reacted with NaN_3 in DMF at room temperature to give in 12 h the corresponding 6-azidomethylpyrimidines **IIa**–**IIf** (Scheme 1). The latter were heated with triphenylphosphine in acetonitrile solution to obtain new 1,2,3,4-tetrahydropyrimidin-2-one derivatives, methyliminophosphoranes **IIIa**–**IIIf**, whose structure was consistent with their ^1H and ^{31}P NMR spectra.

In the past two decades, aza-Wittig reactions of *N*-alkyl(alkenyl, aryl, hetaryl)iminophosphoranes have become a powerful tool in the synthesis of various heterocycles and fused heterocyclic systems [17, 18]. It was shown that 2-aminoimidazoles can be synthesized by condensation of amines with carbodiimides generated by the aza-Wittig reaction [19–21]. However, examples of reactions of carbodiimides with amino groups in partially hydrogenated pyrimidines were not reported. We have found that iminophosphoranes **IIIa**–**IIIf** react with isocyanates **IVa**–**IVe** in boiling chlorobenzene to produce ethyl 3-R-amino-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylates **Va**–**Vl** in satisfactory yield. Presumably, initial aza-Wittig reaction gives pyrimidinylmethylcarbodiimides **A** which, despite reduced basicity of the N^1 atom in the pyrimidine ring, undergo intramolecular cyclization on heating in the absence of a catalyst to form 1,5,6,7-tetrahydroimidazo[1,5-*c*]pyrimidines **B**. The subsequent 1,3-H shift in the C^1–C^{8a}=C^8 allylic system yields more stable structure **V** (Scheme 1). This transformation is an example of hydrogenation of pyrimidine ring via aromatization of imidazole ring fused thereto.

The structure of compounds **Va**–**Vl** was determined on the basis of their IR, ^1H and ^{13}C NMR, and mass spectra. The IR spectra of **Va**–**Vl** contained absorption bands belonging to stretching vibrations of carbonyl groups in the cyclic ureide fragment (1700–1710 cm^{-1}) and ester moiety (1730–1740), as well as N–H absorption bands in the region 3230–3390 cm^{-1}. According to the ^1H NMR data, analytically pure samples of some compounds **V** (obtained by recrystallization from propan-2-ol) were mixtures of diastereoisomers. In particular, double sets of signals from almost all protons were observed in the ^1H NMR spectra of **Vb**, **Vc**, **Ve**, **Vh**, **Vi**, and **Vl**; the signal intensity ratios were 3:1 (**Vb**), 5:1 (**Vc**), 1:1 (**Ve**), 10:1 (**Vh**, **Vi**), and 8:1 (**Vl**). After repeated recrystallization from ethanol–DMF (3:1), signals belonging to the minor isomer disappeared from the spectra of compounds **Vb**, **Vc**, **Vh**, **Vi**, and **Vl**, whereas the spectral pattern of **Ve** almost did not change. These findings suggests that the 1,3-H shift leading to the target products is stereoselective (in most cases, one of the four possible diastereoisomers prevails). The minor product is removed by recrystallization of the diastereoisomer mixture.

Analysis of the NMR spectra of racemic compound **Va**, pure diastereoisomers **Va**–**Vd** and **Vf**–**Vl**, and diastereoisomer mixture **Ve** allowed us to reliably assign the most typical signals. The 8-H proton in major diastereoisomers resonated in the ^{1}H NMR spectra at δ 3.94–4.36 ppm as a doublet with a coupling constant *J* of 3.7–4.2 Hz, while the 7-H signal appeared as a multiplet in the region δ 4.90–5.06 ppm. The singlet at δ 6.23–6.64 ppm was assigned to 1-H. The presence in the ^{1}H NMR spectra of **Vd** and **Vf** of a doublet signal from the NH proton in the exocyclic cyclohexylamino group (δ 6.5 ppm, *J* 7.6 Hz) rules out isomeric 3-imino structure. Minor diastereoisomers are characterized by insignificant shifts of the above signals, and the coupling constant for 8-H is 5.0 Hz. The coupling constants for 8-H in the major (*J* = 3.7–4.2 Hz) and minor diastereoisomers (*J* = 5.0 Hz) indicate small difference in the corresponding torsion angles; therefore, equatorial–axial orientation of 7-H and 8-H was presumed. The ^{13}C NMR spectra of individual diastereoisomers **Vd** and **Vf**–**Vl** contained signals from carbon atoms in the imidazo[1,5-*c*]pyrimidine system at δ_C ~43 (C^8), ~55 (C^7), 115–117 (C^{8a}), 122–123 (C^1), and 144–149 ppm (C^3). In the ^{13}C NMR spectrum of diastereoisomer mixture **Ve**, the corresponding signals were doubled, but the difference in the chemical shifts was fairly small ($\Delta\delta_C$ ~0.3–1 ppm).

EXPERIMENTAL

The IR spectra were recorded in KBr on a UR-20 instrument. The ^{1}H NMR spectra were measured on a Varian Gemini spectrometer at 299.94 MHz using tetramethylsilane as internal reference. The ^{31}P NMR spectra were obtained on a Varian Gemini instrument at 80.95 MHz relative to 85% H_3PO_4 as external reference. The ^{13}C NMR spectra were run on a Bruker Avance DRX-500 spectrometer (125.75 MHz) using tetramethylsilane as internal reference. All NMR spectra were recorded from solutions in DMSO-d_6.

Ethyl 4-aryl-6-chloromethyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylates Id–If (*general procedure***).** A mixture of 0.02 mol of the corresponding aromatic aldehyde, 3.3 g (0.02 mol) of ethyl 4-chloro-3-oxobutanoate, and 1.8 g (0.03 mol) of urea in 50 ml of acetic acid was heated for 48 h at 45–50°C. The mixture was then poured into 200 ml of water, and the precipitate was filtered off, dried, and recrystallized from 60% aqueous ethanol.

Ethyl 2-oxo-6-chloromethyl-4-(4-chlorophenyl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate (Id). Yield 84%, mp 180–181°C. IR spectrum, ν, cm^{-1}: 3370, 3240 (N–H); 1690 (C=O, ester); 1640 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.14 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.0 Hz), 4.04 m (2H, OCH_2), 4.56 d (1H, CH_2Cl, *J* = 10.5 Hz), 4.76 d (1H, CH_2, *J* = 10.5 Hz), 5.18 d (1H, 4-H, *J* = 3.3 Hz), 7.24 d (2H, H_{arom}, *J* = 8.6 Hz), 7.33 d (2H, H_{arom}, *J* = 8.6 Hz), 7.81 s and 9.47 s (1H each, NH). Found, %: C 51.19; H 4.30; N 8.57. $C_{14}H_{14}Cl_2N_2O_3$. Calculated, %: C 51.08; H 4.29; N 8.51.

Ethyl 6-chloromethyl-4-(4-methoxyphenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (Ie). Yield 79%, mp 185–186°C. IR spectrum, ν, cm^{-1}: 3365, 3290 (N–H), 1690 (C=O, ester); 1640 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.14 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.0 Hz), 3.72 s (3H, OCH_3), 4.02 m (2H, OCH_2), 4.58 d (1H, CH_2Cl, *J* = 10.6 Hz), 4.75 d (1H, CH_2Cl, *J* = 10.6 Hz), 5.13 d (1H, 4-H, *J* = 3.3 Hz), 6.84 d (2H, H_{arom}, *J* = 8.7 Hz), 7.16 d (2H, H_{arom}, *J* = 8.7 Hz), 7.71 s and 9.37 s (1H, NH). Found, %: C 55.59; H 5.30; N 8.59. $C_{15}H_{17}ClN_2O_4$. Calculated, %: C 55.48; H 5.28; N 8.63.

Ethyl 4-(4-*tert*-butylphenyl)-6-chloromethyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (If). Yield 76%, mp 169–171°C. IR spectrum, ν, cm^{-1}: 3370, 3290 (N–H); 1695 (C=O, ester); 1645 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.13 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.0 Hz), 1.25 s (9H, *t*-Bu), 4.06 m (2H, OCH_2), 4.61 d (1H, CH_2Cl, *J* = 10.7 Hz), 4.75 d (1H, CH_2Cl, *J* = 10.7 Hz), 5.16 d (1H, 4-H, *J* = 3.4 Hz), 7.18 d (2H, H_{arom}, *J* = 8.4 Hz), 7.35 d (2H, H_{arom}, *J* = 8.4 Hz), 7.80 s and 9.47 s (1H each, NH). Found, %: C 61.59; H 6.60; N 8.00. $C_{18}H_{23}ClN_2O_3$. Calculated, %: C 61.62; H 6.61; N 7.98.

Ethyl 4-alkyl(aryl)-6-azidomethyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylates IIa–IIf (*general procedure***).** A mixture of 14 mmol of compound **Ia**–**If**, 1 g (15.4 mmol) of sodium azide, and a catalytic amount of tetrabutylammonium iodide in 8 ml of DMF was stirred for 5 h and left overnight. The mixture was then poured into 100 ml of water, and the precipitate was filtered off and recrystallized from ethanol.

Ethyl 6-azidomethyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIa). Yield 67%, mp 149–150°C. IR spectrum, ν, cm^{-1}: 3330, 3240 (N–H); 2150 (N_3); 1705 (C=O, ester); 1640 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.23 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.0 Hz), 3.96 s (2H, CH_2), 4.11 m (2H, OCH_2), 4.29 s (2H, CH_2), 7.08 s and 9.05 s (1H each, NH). Found, %: C 41.19;

H 4.90; N 31.00. $C_8H_{11}N_5O_3$. Calculated, %: C 2.67; H 4.92; N 31.10.

Ethyl 6-azidomethyl-4-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIb). Yield 62%, mp 144–145°C. IR spectrum, ν, cm^{-1}: 3320, 3230 (N–H); 2150 (N_3); 1705 (C=O, ester); 1645 (C=O). ^1H NMR spectrum, δ, ppm: 1.17 d (3H, 4-CH_3, J = 6.6 Hz), 1.24 t (3H, CH_2CH_3, J = 7.0 Hz), 4.05–4.29 m (4H, OCH_2, CH_2N_3, 4-H), 4.38 d (1H, CH_2N_3, J = 13 Hz), 7.25 s and 9.14 s (1H each, NH). Found, %: C 44.99; H 5.41; N 29.20. $C_9H_{13}N_5O_3$. Calculated, %: C 45.19; H 5.48; N 29.27.

Ethyl 6-azidomethyl-2-oxo-4-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIc). Yield 65%, mp 234–236°C. IR spectrum, ν, cm^{-1}: 3330 (N–H), 2150 (N_3), 1705 (C=O, ester), 1640 (C^2=O). ^1H NMR spectrum, δ, ppm: 1.14 t (3H, CH_2CH_3, J = 7.0 Hz), 4.05 m (2H, OCH_2), 4.35 d (1H, CH_2N_3, J = 12.7 Hz), 4.46 d (1H, CH_2N_3, J = 12.7 Hz), 5.21 d (1H, 4-H, J = 3.2 Hz), 7.29 m (5H, Ph), 7.80 s and 9.39 s (1H each, NH). Found, %: C 55.79; H 5.01; N 23.20. $C_{14}H_{15}N_5O_3$. Calculated, %: C 55.81; H 5.02; N 23.24.

Ethyl 6-azidomethyl-4-(4-chlorophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IId). Yield 67%, mp 119–120°C. IR spectrum, ν, cm^{-1}: 3330 (N–H), 2150 (N_3), 1705 (C=O, ester), 1640 (C^2=O). ^1H NMR spectrum, δ, ppm: 1.14 t (3H, CH_2CH_3, J = 7.0 Hz), 4.04 m (2H, OCH_2), 4.30 d (1H, CH_2N_3, J = 13 Hz), 4.58 d (1H, CH_2N_3, J = 13 Hz), 5.20 d (1H, 4-H, J = 3.2 Hz), 7.24–7.38 m (4H, H_{arom}), 7.82 s and 9.43 s (1H each, NH). Found, %: C 50.00; H 4.11; N 21.00. $C_{14}H_{14}ClN_5O_3$. Calculated, %: C 50.08; H 4.20; N 20.86.

Ethyl 6-azidomethyl-4-(4-methoxyphenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIe). Yield 70%, mp 132–136°C. IR spectrum, ν, cm^{-1}: 3360, 3255 (N–H); 2135 (N_3); 1690 (C=O, ester); 1640 (C^2=O). ^1H NMR spectrum, δ, ppm: 1.14 t (3H, CH_2CH_3, J = 7.0 Hz), 3.73 s (3H, OCH_3), 4.04 m (2H, OCH_2), 4.31 d (1H, CH_2N_3, J = 13 Hz), 4.45 d (1H, CH_2N_3, J = 13 Hz), 5.15 d (1H, 4-H, J = 3.2 Hz), 6.83 d (2H, H_{arom}, J = 8.5 Hz), 7.17 d (2H, H_{arom}, J = 8.5 Hz), 7.72 s and 9.33 s (1H each, NH). Found, %: C 54.32; H 5.13; N 21.09. $C_{15}H_{17}N_5O_4$. Calculated, %: C 54.38; H 5.17; N 21.14.

Ethyl 6-azidomethyl-4-(4-*tert*-butylphenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIf). Yield 71%, mp 107–108°C. IR spectrum, ν, cm^{-1}: 3390, 3330 (N–H); 2120 (N_3); 1705 (C=O, ester); 1660 (C^2=O). ^1H NMR spectrum, δ, ppm: 1.16 t (3H, CH_2CH_3, J = 7.0 Hz), 1.27 s (9H, *t*-Bu), 4.04 m (2H, OCH_2), 4.29 d (1H, CH_2N_3, J = 13 Hz), 4.48 d (1H, CH_2N_3, J = 13 Hz), 5.17 d (1H, 4-H, J = 3.2 Hz), 7.18 d (2H, H_{arom}, J = 8.5 Hz), 7.30 d (2H, H_{arom}, J = 8.5 Hz), 7.74 s and 9.35 s (1H each, NH). Found, %: C 60.33; H 6.43; N 19.67. $C_{18}H_{23}N_5O_3$. Calculated, %: C 60.49; H 6.49; N 19.59.

Ethyl 4-alkyl(aryl)-2-oxo-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylates IIIa–IIIf (*general procedure*). A solution of 1.834 g (7 mmol) of triphenylphosphine in 20 ml of acetonitrile was added to a solution of 7 mmol of compound **IIa–IIf** in 25 ml of hot acetonitrile, the mixture was left overnight at room temperature, and the precipitate was filtered off.

Ethyl 2-oxo-6-[(triphenyl-λ^5-phosphanylidene)-aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIIa). Yield 88%, mp 159–160°C. IR spectrum, ν, cm^{-1}: 3260 (N–H), 1685 (C=O, ester), 1635 (C^2=O). ^1H NMR spectrum, δ, ppm: 1.11 t (3H, CH_2CH_3, J = 7.1 Hz), 3.88–3.98 m (4H, OCH_2, 4-H), 4.19 d (1H, 6-CH_2, J = 11.5 Hz), 7.08 s (1H, NH), 7.58 m (15H, Ph), 8.76 s (1H, NH). ^{31}P NMR spectrum: δ_P 18.80 ppm. Found, %: C 68.04; H 5.77; N 9.17. $C_{26}H_{26}N_3O_3P$. Calculated, %: C 67.96; H 5.70; N 9.14.

Ethyl 4-methyl-2-oxo-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIIb). Yield 87%, mp 176–179°C. IR spectrum, ν, cm^{-1}: 3270 (N–H), 1685 (C=O, ester), 1630 (C^2=O). ^1H NMR spectrum, δ, ppm: 1.12 m (6H, CH_2CH_3, 4-CH_3), 3.85–4.31 m (5H, OCH_2, 6-CH_2, 4-H), 7.28 s (1H, NH), 7.59 m (15H, Ph), 8.89 s (1H, NH). ^{31}P NMR spectrum: δ_P 14.54 ppm. Found, %: C 68.54; H 5.97; N 8.82. $C_{27}H_{28}N_3O_3P$. Calculated, %: C 68.49; H 5.96; N 8.87.

Ethyl 2-oxo-4-phenyl-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIIc). Yield 88%, mp 155–157°C. IR spectrum, ν, cm^{-1}: 3210 (N–H), 1680 (C=O, ester), 1640 (C=O). ^1H NMR spectrum, δ, ppm: 1.00 t (3H, CH_2CH_3, J = 7.0 Hz), 3.89 m (2H, OCH_2), 4.30 m (2H, 6-CH_2), 5.15 d (1H, 4-H, J = 3.4 Hz), 7.19–7.34 m (5H, Ph), 7.62 m (15H, Ph), 7.89 s and 9.06 s (1H each, NH). ^{31}P NMR spectrum: δ_P 16.07 ppm. Found, %: C 71.64; H 5.69; N 7.83. $C_{32}H_{30}N_3O_3P$. Calculated, %: C 71.76; H 5.65; N 7.85.

Ethyl 4-(4-chlorophenyl)-2-oxo-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIId). Yield 85%, mp 190–193°C. IR spectrum, ν, cm^{-1}: 3210 (N–H),

1675 (C=O, ester), 1638 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.02 t (3H, $CH_2C\mathbf{H}_3$, J = 7.0 Hz), 3.92 m (2H, OCH_2), 4.32 m (2H, 6-CH_2), 5.17 d (1H, 4-H, J = 3.2 Hz), 7.22 d (2H, H_{arom}, J = 8.0 Hz), 7.38 d (2H, H_{arom}, J = 8.0 Hz), 7.61 m (15H, Ph), 7.92 s and 9.11 s (1H each, NH). ^{31}P NMR spectrum: δ 16.14 ppm. Found, %: C 67.55; H 5.15; N 7.42. $C_{32}H_{29}ClN_3O_3P$. Calculated, %: C 67.43; H 5.13; N 7.37.

Ethyl 4-(4-methoxyphenyl)-2-oxo-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIIe). Yield 76%, mp 145–146°C. IR spectrum, ν, cm^{-1}: 3215 (N–H), 1680 (C=O, ester), 1640 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.01 t (3H, $CH_2C\mathbf{H}_3$, J = 7.0 Hz), 3.71 s (3H, OCH_3), 3.87 m (2H, OCH_2), 4.29 m (2H, 6-CH_2), 5.11 d (1H, 4-H, J = 3.2 Hz), 6.86 d (2H, H_{arom}, J = 8.2 Hz), 7.11 d (2H, H_{arom}, J = 8.2 Hz), 7.61 m (15H, Ph), 7.85 s and 9.06 s (1H each, NH). ^{31}P NMR spectrum: δ_P 16.30 ppm. Found, %: C 70.05; H 5.72; N 7.41. $C_{33}H_{32}N_3O_4P$. Calculated, %: C 70.08; H 5.70; N 7.43.

Ethyl 4-(4-*tert*-butylphenyl)-2-oxo-6-[(triphenyl-λ^5-phosphanylidene)aminomethyl]-1,2,3,4-tetrahydropyrimidine-5-carboxylate (IIIf). Yield 69%, mp 174–176°C. IR spectrum, ν, cm^{-1}: 3210 (N–H), 1680 (C=O, ester), 1640 (C^2=O). ^{1}H NMR spectrum, δ, ppm: 1.02 t (3H, $CH_2C\mathbf{H}_3$, J = 7.0 Hz), 1.25 s (9H, *t*-Bu), 3.88 m (2H, OCH_2), 4.27 m (2H, 6-CH_2), 5.12 d (1H, 4-H, J = 3.2 Hz), 7.14 d (2H, H_{arom}, J = 8.0 Hz), 7.31 d (2H, H_{arom}, J = 8.0 Hz), 7.59 m (15H, Ph), 7.86 s and 9.05 s (1H each, NH). ^{31}P NMR spectrum: δ_P 16.25 ppm. Found, %: C 73.01; H 6.53; N 7.12. $C_{36}H_{38}N_3O_3P$. Calculated, %: C 73.08; H 6.47; N 7.10.

Ethyl 3-alkyl(aryl)amino-7-alkyl(aryl)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylates Va–Vl (*general procedure*). A mixture of 2 mmol of compound **IIIa**–**IIIf** and 2 mmol of isocyanate **IVa**–**IVe** in 10 ml of chlorobenzene was heated for 5–6 h under reflux, the mixture was evaporated, and the residue was recrystallized from propan-2-ol. Compounds **Vb**, **Vc**, **Ve**, **Vh**, **Vi**, and **Vl** were additionally purified by recrystallization from ethanol–DMF (3:1).

Ethyl 3-(4-chlorophenylamino)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Va). Yield 52%, mp 176–177°C. IR spectrum, ν, cm^{-1}: 3245 (N–H), 1740 (C=O, ester), 1700 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.24 t (3H, $CH_2C\mathbf{H}_3$, J = 6.5 Hz), 3.56 m (3H, 7-H, 8-H), 4.17 m (2H, OCH_2), 6.58 s (1H, 1-H), 7.26 d (2H, H_{arom}, J = 8.7 Hz), 7.71 d (2H, H_{arom}, J = 8.7 Hz), 8.39 s and 9.40 s (1H each, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.95 ($CH_2\mathbf{C}H_3$), 36.77 (C^7), 42.08 (C^8), 61.59 (OCH_2), 118.90 (CH_{arom}), 122.21 (C^1), 125.03 (C^{8a}), 129.03 (CH_{arom}), 139.14 (C_{arom}), 145.44 (C^3), 151.45 (C^5), 169.88 (C=O). Mass spectrum: m/z 335 $[M + 1]^+$. Found, %: C 53.94; H 4.56; N 16.71. $C_{15}H_{15}ClN_4O_3$. Calculated, %: C 53.82; H 4.52; N 16.74. M 334.76.

Ethyl 3-(4-chlorophenylamino)-7-methyl-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vb). Yield 27%, mp 195–196°C. IR spectrum, ν, cm^{-1}: 3245 (N–H), 1735 (C=O, ester), 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.21 m (6H, CH_3), 3.94 m (2H, 7-H, 8-H), 4.15 m (2H, OCH_2), 6.62 s (1H, 1-H), 7.32 d (2H, H_{arom}, J = 8.5 Hz), 7.77 d (2H, H_{arom}, J = 8.5 Hz), 8.48 s and 9.39 s (1H each, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.78 ($CH_2\mathbf{C}H_3$), 19.78 (7-CH_3), 42.51 (C^8), 48.62 (C^7), 61.10 (OCH_2), 117.61 (C^{8a}), 118.52 (CH_{arom}), 122.66 (C^1), 124.39 (C_{arom}), 128.49 (CH_{arom}), 138.45 (C_{arom}), 144.69 (C^3), 150.07 (C^5), 169.39 (C=O). Mass spectrum: m/z 349 $[M + 1]^+$. Found, %: C 55.23; H 4.99; N 16.11. $C_{16}H_{17}ClN_4O_3$. Calculated, %: C 55.10; H 4.91; N 16.06. M 348.79.

Ethyl 3-(4-methoxyphenylamino)-7-methyl-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vc). Yield 60%, mp 155–157°C. IR spectrum, ν, cm^{-1}: 3245 (N–H), 1735 (C=O, ester), 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.18 d (3H, 7-CH_3, J = 6.7 Hz), 1.23 t (3H, $CH_2C\mathbf{H}_3$, J = 7.0 Hz), 3.72 s (3H, OCH_3), 3.92 m (1H, 7-H), 4.01 d (1H, 8-H, J = 4.1 Hz), 4.13 m (2H, OCH_2), 6.55 s (1H, 1-H), 7.83 d (2H, H_{arom}, J = 8.5 Hz), 7.58 d (2H, H_{arom}, J = 8.5 Hz), 8.32 s and 9.16 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 14.03 ($CH_2\mathbf{C}H_3$), 17.10 (7-CH_3), 41.12 (C^8), 48.10 (C^7), 55.07 (OCH_3), 60.56 (OCH_2), 114.00 (CH_{arom}), 117.67 (C^{8a}), 118.36 (CH_{arom}), 122.05 (C^1), 132.99 (C_{arom}), 145.62 (C^3), 150.62 (C^5), 153.81 (C_{arom}), 168.55 (C=O). Mass spectrum: m/z 345 $[M + 1]^+$. Found, %: C 59.22; H 5.87; N 16.25. $C_{17}H_{20}N_4O_4$. Calculated, %: C 59.29; H 5.85; N 16.27. M 344.37.

Ethyl 3-cyclohexylamino-5-oxo-7-phenyl-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vd). Yield 48%, mp 191–192°C. IR spectrum, ν, cm^{-1}: 3235 (N–H), 1730 (C=O, ester), 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.20–1.47 m (8H, CH_3, *cyclo*-C_6H_{11}), 1.50–1.83 m (3H, *cyclo*-C_6H_{11}), 1.98 m (2H, *cyclo*-C_6H_{11}), 3.55 m (1H, *cyclo*-C_6H_{11}), 4.09 m (3H, OCH_2, 8-H), 4.90 m (1H, 7-H), 6.22 s (1H, 1-H), 6.55 d (1H, NH, J = 7.6 Hz), 7.22–7.33 m (5H, Ph), 8.49 d (1H, NH, J = 2.5 Hz). ^{13}C NMR spectrum, δ_C, ppm: 13.75 ($CH_2\mathbf{C}H_3$), 24.22 (*cyclo*-C_6H_{11}), 25.18

(*cyclo*-C_6H_{11}), 32.46 (*cyclo*-C_6H_{11}), 43.78 (C^8), 50.37 (*cyclo*-C_6H_{11}), 55.80 (C^7), 61.02 (OCH_2), 115.40 (C^{8a}), 123.06 (C^1), 126.03 (CH_{arom}), 127.71 (CH_{arom}), 128.40 (CH_{arom}), 139.66 (C_{arom}), 149.15 (C^3), 150.62 (C^5), 169.40 (C=O). Mass spectrum: *m*/*z* 383 $[M + 1]^+$. Found, %: C 65.89; H 6.90; N 14.55. $C_{21}H_{26}N_4O_3$. Calculated, %: C 65.95; H 6.85; N 14.65. *M* 382.46.

Ethyl 3-(4-chlorophenylamino)-5-oxo-7-phenyl-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Ve). Yield 59%, mp 206–208°C. IR spectrum, ν, cm^{-1}: 3230 (N–H), 1730 (C=O, ester), 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 0.95 t and 1.19 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.0 Hz), 3.86 m and 4.12 m (2H, OCH_2), 4.23 d (0.5H, 8-H, *J* = 4.0 Hz), 4.40 d (0.5H, 8-H, *J* = 5.0 Hz), 4.95–5.11 m (1H, 7-H), 6.49 s and 6.65 s (1H, 1-H), 7.13–7.40 m (7H, H_{arom}), 7.74 m (2H, H_{arom}), 8.85 s and 8.86 s (1H, NH), 9.44 s and 9.55 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.51 and 13.84 ($CH_2\mathbf{C}H_3$), 42.72 and 43.63 (C^8), 55.99 and 56.04 (C^7), 60.51 and 61.25 (OCH_2), 117.00 and 117.47 (C^{8a}), 118.63 and 118.65 (CH_{arom}), 122.63 and 123.10 (C^1), 124.48 and 124.50 (C_{arom}), 126.15 and 126.74 (CH_{arom}), 127.96 and 128.18 (C_{arom}), 128.29 and 128.53 (C_{arom}), 128.56 and 128.60 (C_{arom}), 137.19 and 138.39 (CH_{arom}), 138.46 and 139.25 (CH_{arom}), 144.70 and 144.93 (C^3), 150.45 and 150.88 (C^5), 167.84 and 169.15 (C=O). Mass spectrum: *m*/*z* 411 $[M + 1]^+$. Found, %: C 61.45; H 4.72; N 13.60. $C_{21}H_{19}ClN_4O_3$. Calculated, %: C 61.39; H 4.66; N 13.64. *M* 410.86.

Ethyl 7-(4-chlorophenyl)-3-cyclohexylamino-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vf). Yield 44%, mp 189–191°C. IR spectrum, ν, cm^{-1}: 3230, 3390 (N–H); 1740 (C=O, ester); 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.20–1.45 m (8H, CH_3, *cyclo*-C_6H_{11}), 1.55–1.82 m (3H, *cyclo*-C_6H_{11}), 1.98 m (2H, *cyclo*-C_6H_{11}), 3.54 m (1H, *cyclo*-C_6H_{11}), 4.09 m (1H, OCH_2, 8-H), 4.91 m (1H, 7-H), 6.23 s (1H, 1-H), 6.52 d (1H, NH, *J* = 7.6 Hz), 7.25 d (2H, H_{arom}, *J* = 8.6 Hz), 7.35 d (2H, H_{arom}, *J* = 8.6 Hz), 8.51 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.75 ($CH_2\mathbf{C}H_3$), 24.28 (*cyclo*-C_6H_{11}), 25.21 (*cyclo*-C_6H_{11}), 32.48 (*cyclo*-C_6H_{11}), 43.48 (C^8), 50.42 (*cyclo*-C_6H_{11}), 55.23 (C^7), 61.11 (OCH_2), 115.31 (C^{8a}), 123.15 (C^1), 128.23 (CH_{arom}), 128.46 (CH_{arom}), 132.55 (C_{arom}), 138.55 (C_{arom}), 149.19 (C^3), 150.53 (C^5), 169.21 (C=O). Mass spectrum: *m*/*z* 417 $[M + 1]^+$. Found, %: C 60.78; H 6.00; N 13.53. $C_{21}H_{25}ClN_4O_3$. Calculated, %: C 60.50; H 6.04; N 13.44. *M* 416.91.

Ethyl 7-(4-chlorophenyl)-3-(4-chlorophenylamino)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vg). Yield 47%, mp >250°C. IR spectrum, ν, cm^{-1}: 3230, 3335 (N–H); 1735 (C=O, ester), 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.16 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.0 Hz), 4.13 m (2H, OCH_2), 4.37 d (1H, 8-H, *J* = 4.2 Hz), 5.06 m (1H, 7-H), 6.57 s (1H, 1-H), 7.33–7.45 m (6H, H_{arom}), 7.77 d (2H, H_{arom}, *J* = 8.4 Hz), 8.96 s and 9.42 s (1H each, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.87 ($CH_2\mathbf{C}H_3$), 43.44 (C^8), 55.35 (C^7), 61.32 (OCH_2), 116.86 (C^{8a}), 118.63 (CH_{arom}), 123.06 (C^1), 124.56 (C_{arom}), 128.19 (CH_{arom}), 128.57 (CH_{arom}), 128.59 (CH_{arom}), 132.65 (C_{arom}), 138.19 (C_{arom}), 138.37 (C_{arom}), 144.74 (C^3), 150.33 (C^5), 168.97 (C=O). Mass spectrum: *m*/*z* 445 $[M + 1]^+$. Found, %: C 56.63; H 4.10; N 12.52. $C_{21}H_{18}Cl_2N_4O_3$. Calculated, %: C 56.64; H 4.07; N 12.58. *M* 445.30.

Ethyl 7-(4-chlorophenyl)-3-(4-methoxyphenylamino)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vh). Yield 46%, mp 236–237°C. IR spectrum, ν, cm^{-1}: 3230, 3340 (N–H); 1735 (C=O, ester); 1710 (C^5=O). ^{1}H NMR spectrum, δ, ppm: 1.16 t (3H, $CH_2C\mathbf{H}_3$, *J* = 6.9 Hz), 3.71 s (3H, OCH_3), 4.14 m (2H, OCH_2), 4.32 d (1H, 8-H, *J* = 3.8 Hz), 5.05 m (1H, 7-H), 6.52 s (1H, 1-H), 6.87 d (2H, H_{arom}, *J* = 8.5 Hz), 7.33 d (2H, H_{arom}, *J* = 8.4 Hz), 7.44 d (2H, H_{arom}, *J* = 8.2 Hz), 7.64 d (2H, H_{arom}, *J* = 8.5 Hz), 8.89 s and 9.17 s (1H each, NH). ^{13}C NMR spectrum, δ_C, ppm: 14.41 ($CH_2\mathbf{C}H_3$), 44.02 (C^8), 55.68 (OCH_3), 55.87 (C^7), 61.82 (OCH_2), 114.59 (CH_{arom}), 116.72 (C^{8a}), 119.00 (CH_{arom}), 123.72 (C^1), 128.81 (CH_{arom}), 129.13 (CH_{arom}), 133.17 (C_{arom}), 133.37 (C_{arom}), 138.87 (C_{arom}), 146.12 (C^3), 151.11 (C^5), 154.43 (C_{arom}), 169.62 (C=O). Mass spectrum: *m*/*z* 441 $[M + 1]^+$. Found, %: C 59.93; H 4.77; N 12.63. $C_{22}H_{21}ClN_4O_4$. Calculated, %: C 59.93; H 4.80; N 12.71. *M* 440.89.

Ethyl 7-(4-methoxyphenyl)-3-(4-methoxyphenylamino)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vi). Yield 30%, mp 199–201°C. IR spectrum, ν, cm^{-1}: 3230, 3340 (N–H); 1735 (C=O, ester); 1710 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.19 t (3H, $CH_2C\mathbf{H}_3$, *J* = 7.1 Hz), 3.72 s (6H, OCH_3), 4.14 m (3H, OCH_2, 8-H), 4.92 m (1H, 7-H), 6.43 s (1H, 1-H), 6.84 m (4H, H_{arom}), 7.16 d (2H, H_{arom}, *J* = 8.6 Hz), 7.59 d (2H, H_{arom}, *J* = 8.6 Hz), 8.72 d (1H, NH, *J* = 3.0 Hz), 9.18 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.91 ($CH_2\mathbf{C}H_3$), 43.81 (C^8), 55.06 and 55.17 (OCH_3), 55.54 (C^7), 61.19 (OCH_2), 113.97 (CH_{arom}), 114.08 (CH_{arom}), 116.59 (C^{8a}), 118.44 (CH_{arom}), 122.99 (C^1), 127.47 (CH_{arom}), 131.22 (C_{arom}), 132.91 (C_{arom}), 145.51 (C^3), 150.68 (C^5), 153.88 (C_{arom}), 158.91 (C_{arom}), 169.31 (C=O). Found, %: C 62.98; H 5.47; N 12.80. $C_{23}H_{24}N_4O_5$. Calculated, %: C 63.29; H 5.54; N 12.84.

Ethyl 7-(4-*tert*-butylphenyl)-5-oxo-3-[3-(trifluoromethyl)phenylamino]-5,6,7,8-tetrahydroimidazo-[1,5-*c*]pyrimidine-8-carboxylate (Vj). Yield 58%, mp 200–201°C. IR spectrum, ν, cm^{-1}: 3230, 3335 (N–H); 1735 (C=O, ester); 1710 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.20 t (3H, CH_2CH_3, J = 7.2 Hz), 1.25 s (9H, *t*-Bu), 4.13 m (2H, OCH_2), 4.27 d (1H, 8-H, J = 3.8 Hz), 4.98 m (1H, 7-H), 6.56 s (1H, 1-H), 7.11–7.40 m (5H, H_{arom}), 7.47 t (1H, H_{arom}, J = 7.8 Hz), 7.77 d (1H, H_{arom}, J = 8.5 Hz), 8.32 s (1H, H_{arom}), 8.89 d (1H, NH, J = 2.3 Hz), 9.63 s (1H, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.69 (CH_2CH_3), 30.81 [$C(CH_3)_3$], 33.98 [$C(CH_3)_3$], 43.36 (C^8), 55.64 (C^7), 61.22 (OCH_2), 113.02 (CH_{arom}), 117.14 (CH_{arom}), 117.33 (C^{8a}), 120.95 (CH_{arom}), 123.10 (C^1), 124.23 q (CF_3, J_{CF} = 271 Hz), 125.31 (CH_{arom}), 125.79 (CH_{arom}), 129.50 (C_{arom}), 129.74 (CH_{arom}), 136.29 (C_{arom}), 140.19 (C_{arom}), 144.46 (C^3), 150.31 (C_{arom}), 150.41 (C^5), 169.21 (C=O). Mass spectrum: m/z 501 $[M + 1]^+$. Found, %: C 62.29; H 5.41; N 11.22. $C_{26}H_{27}F_3N_4O_3$. Calculated, %: C 62.39; H 5.44; N 11.19. M 500.52.

Ethyl 7-(4-*tert*-butylphenyl)-3-(4-methoxyphenylamino)-5-oxo-5,6,7,8-tetrahydroimidazo[1,5-*c*]pyrimidine-8-carboxylate (Vk). Yield 30%, mp 168–170°C. IR spectrum, ν, cm^{-1}: 3335, 3240 (N–H); 1735 (C=O, ester), 1710 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.19 t (3H, CH_2CH_3, J = 6.9 Hz), 1.25 s (9H, *t*-Bu), 3.72 s (3H, OCH_3), 4.10–4.21 m (3H, OCH_2, 8-H), 4.95 m (1H, 7-H), 6.45 s (1H, 1-H), 6.83 d (2H, H_{arom}, J = 8.3 Hz), 7.16 d (2H, H_{arom}, J = 8.3 Hz), 7.33 d (2H, H_{arom}, J = 8.0 Hz), 7.61 d (2H, H_{arom}, J = 8.0 Hz), 8.76 s and 9.21 s (1H each, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.75 (CH_2CH_3), 30.85 [$C(CH_3)_3$], 34.17 [$C(CH_3)_3$], 43.41 (C^8), 54.90 (OCH_3), 55.55 (C^7), 61.22 (OCH_2), 114.04 (CH_{arom}), 116.43 (C^{8a}), 118.53 (CH_{arom}), 123.02 (C^1), 125.36 (CH_{arom}), 125.77 (C_{arom}), 132.80 (C_{arom}), 136.49 (C_{arom}), 145.51 (C^3), 150.37 (C_{arom}), 150.65 (C^5), 153.95 (C_{arom}), 169.38 (C=O). Found, %: C 67.74; H 6.48; N 12.29. $C_{26}H_{30}N_4O_4$. Calculated, %: C 67.51; H 6.54; N 12.11.

Ethyl 7-(4-*tert*-butylphenyl)-3-(3-chloro-4-methylphenylamino)-5-oxo-5,6,7,8-tetrahydroimidazo-[1,5-*c*]pyrimidine-8-carboxylate (Vl). Yield 41%, mp 201–202°C. IR spectrum, ν, cm^{-1}: 3335, 3240 (N–H); 1735 (C=O, ester); 1710 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.15 t (3H, CH_2CH_3, J = 6.9 Hz), 1.23 s (9H, *t*-Bu), 2.26 s (3H, CH_3), 4.15 m (2H, OCH_2), 4.36 d (1H, 8-H, J = 3.7 Hz), 5.00 m (1H, 7-H), 6.59 s (1H, 1-H), 7.09–7.48 m (6H, H_{arom}), 8.09 s (1H, H_{arom}), 8.92 s and 9.40 s (1H each, NH). ^{13}C NMR spectrum, δ_C, ppm: 13.85 (CH_2CH_3), 18.68 (CH_3), 30.95 [$C(CH_3)_3$], 34.17 [$C(CH_3)_3$], 43.49 (C^8), 55.72 (C^7), 61.24 (OCH_2), 116.05 (CH_{arom}), 116.84 (CH_{arom}), 117.02 (C^{8a}), 123.21 (C^1), 125.40 (CH_{arom}), 125.40 (CH_{arom}), 125.85 (C_{arom}), 127.34 (C_{arom}), 131.11 (CH_{arom}), 133.19 (C_{arom}), 136.34 (C_{arom}), 138.58 (C_{arom}), 144.68 (C^3), 150.40 (C_{arom}), 150.45 (C^5), 169.27 (C=O). Found, %: C 65.07; H 6.09; N 11.78. $C_{26}H_{29}ClN_4O_3$. Calculated, %: C 64.93; H 6.08; N 11.65.

REFERENCES

1. Kincl, F.A., Romo, J., Rosenkranz, G., and Sondheimer, F., *J. Chem. Soc*., 1956, p. 4163.
2. Mechoulam, R., Sondheimer, F., Melera, A., and Kincl, F.A., *J. Am. Chem. Soc*., 1961, vol. 83, p. 2022.
3. Siddigi, S.M., Melman, N., Olah, M.E., Jain, R., Evans, R., Glashofer, M., Radget, W., Cohen, L.A., Daly, J.W., Stiles, G.L., and Jacobson, U.A., *Nucleosides Nucleotides*, 1996, vol. 15, p. 693.
4. Yoshida, O., Yasukata, T., Sumino, Y., Munekage, T., Narukawa, Y., and Nishitani, Y., *Bioorg. Med. Chem. Lett*., 2002, vol. 12, p. 3027.
5. Potvin, P.G. and Wong, M.H., *J. Chem. Soc., Chem. Commun*., 1987, p. 672.
6. Narayanan, S., Vangapandu, S., and Jain, R., *Bioorg. Med. Chem. Lett*., 2001, vol. 11, p. 1133.
7. Borchers, A. and Schunack, W., *Arch. Pharm*., 1984, vol. 317, p. 455.
8. Collman, J.P., Zhong, M., and Costanzo, S., *J. Chem. Res., Synop*., 2001, no. 5, p. 195.
9. Schlögl, K and Woidich, H., *Monatsh. Chem*., 1956, vol. 87, p. 679.
10. Chivikas, C.J. and Hodges, J.C., *J. Org. Chem*., 1987, vol. 52, p. 3591.
11. Wade, J.J., *J. Heterocycl. Chem*., 1986, vol. 23, p. 981.
12. Kappe, C.O., *Tetrahedron*, 1993, vol. 49, p. 6937.
13. Dallinger, D., Stadler, A., and Kappe, C.O., *Pure Appl. Chem*., 2004, vol. 76, p. 1017.
14. Zigeuner, G., Hamberger, H., Blasche, H., and Sterk, H., *Monatsh. Chem*., 1966, vol. 97, p. 1408.
15. Biginelli, P., *Gazz. Chim. Ital*., 1893, vol. 23, p. 360.
16. Kappe, C.O., *Justus Liebigs Ann. Chem*., 1990, p. 505.
17. Molina, P. and Vilaplana, M.J., *Synthesis*, 1994, no. 12, p. 1197.
18. Palacios, F., Alouso, C., Aparicio, D., Rubiales, G., and de los Santoc, J.M., *Tetrahedron*, 2007, vol. 63, p. 523.
19. Molina, P., Díaz, I., and Tárraga, A., *Synlett*, 1995, no. 10, p. 1031.
20. Ding, M.-W., Tu, H.-Y., and Liu, Z.-J., *Synth. Commun*., 1997, vol. 27, p. 3657.
21. Palacios, F., Legido, M., Perez de Heredia, I., Rubiales, G., and Ezpeleta, J.M., *Heterocycles*, 2001, vol. 55, p. 1641.