Reactions of Alkyl (*Z*)-2-[(*E*)-2-ethoxycarbonyl-2-(2-pyridinyl)-ethenyl]amino-3-dimethylaminopropenoates with *C*- and *N*-Nucleophiles. The Synthesis of Fused Pyranones, Pyridinones and Pyrimidinones

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Alkyl 2-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-dimethylaminopropenoates **3** and **4** were transformed with *C*-and *N*-nucleophiles into β-heteroaryl-α,β-didehydro-α-amino acid derivatives **13-16**, substituted 3-amino-4*H*-quinolizin-4-one **17**, 2*H*,5*H*-benzo[*b*]pyran-2,5-dione **18** and **19**, 2*H*,5*H*-pyrano[4,3-*b*]pyran-2,5-dione **20**, 2*H*,5*H*-pyrano[3,2-*c*]benzo[*b*]pyran-2,5-dione **21**, 2*H*-1-benzopyran-2-one **22** and **24**, pyrido[1,2-*a*]pyrimidin-4-one **31-34** and **39** derivatives, and *N*-heteroaryl-1*H*-imidazole-4-carboxylates **37** and **38**.

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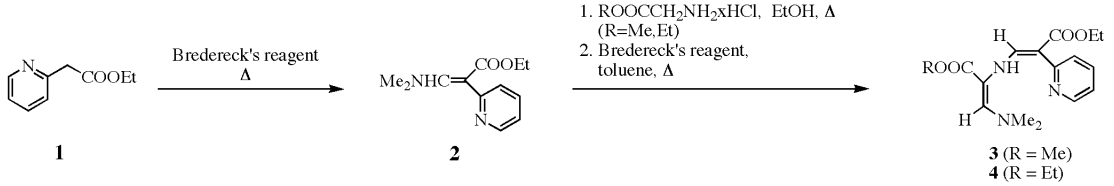
Quinolizines, pyridopyrimidines, benzopyrans, pyranopyrans and related fused systems are the basic structures of many naturally occurring alkaloids and their synthetic derivatives exhibiting various biological activity [1-5].

Recently, alkyl 2-substituted 3-(dimethylamino)propenoates and their cyclic analogs have been shown to be versatile and efficient reagents for the preparation of various heterocyclic systems [6], including some natural products, such as aplysinopsins and analogs [7]. This methodology has opened also an easy access to substituted 4*H*-quinolizin-4-ones, pyridopyrimidines and other heterocyclic systems with an amino group in 3 position of the newly formed heterocyclic system [8-10]. The substituents attached at the 2,2-disubstituted ethenyl group of the substituted amino group are ester groups or a combinations of an ester and an acyl, two acyl, an ester and an amino, an ester and a cyano, two cyano, or an ester and a phenyl group [6].

In this communication we report the transformations of 2-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-dimethylaminopropenoates **3** and **4**, prepared from ethyl 2-pyridinylacetate (**1**) in two steps (Scheme 1) [11], with *C*- and *N*-nucleophiles in order to introduce a heteroaryl substituted ethenylamino group into the newly formed heterocyclic system.

Scheme 1

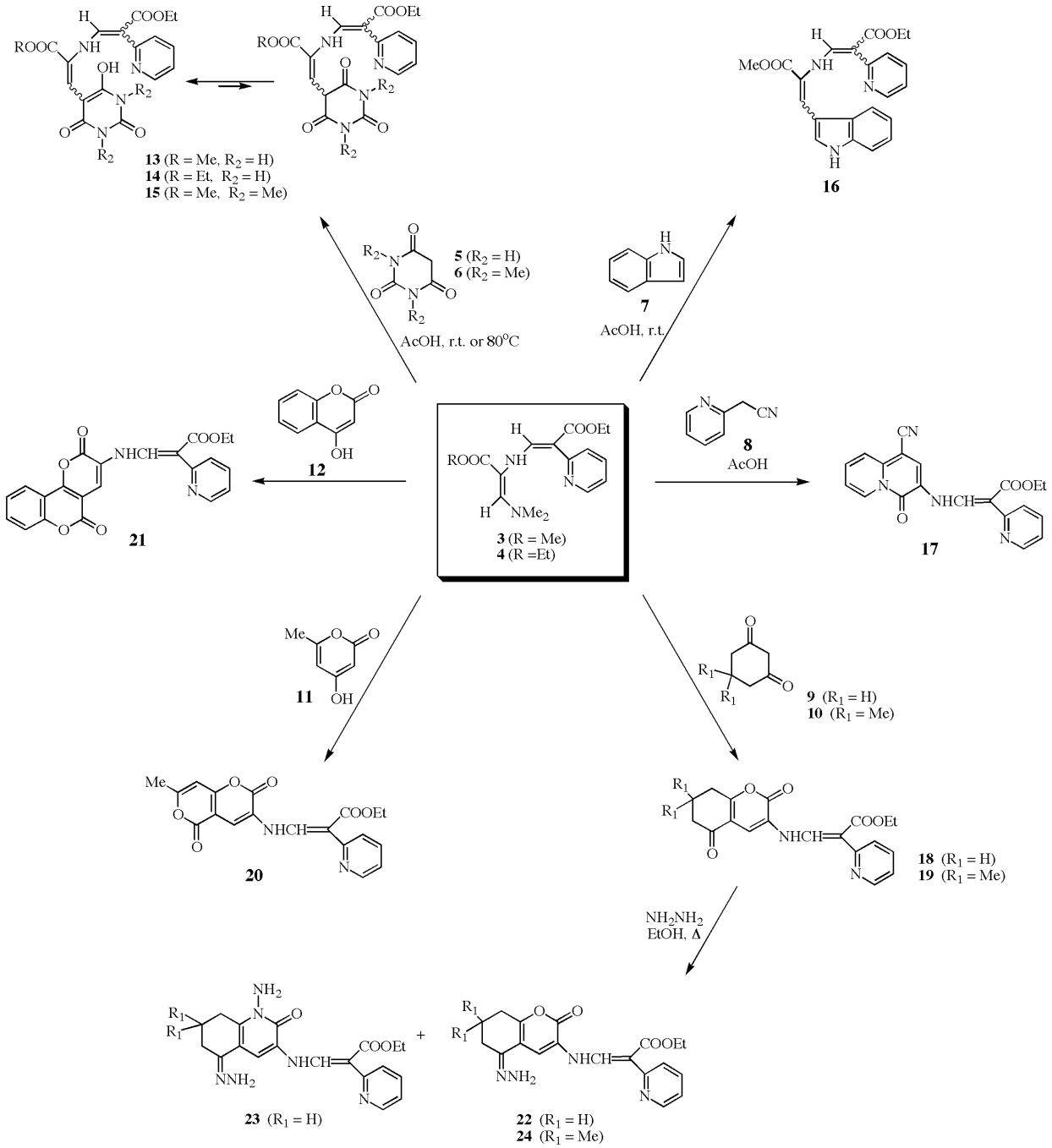


1 **2** **3** (R = Me) **4** (R = Et)

Alkyl (*Z*)-2-[(*E*)-2-ethoxycarbonyl-2-(2-pyridinyl)-ethenyl]amino-3-dimethylaminoprope-noates **3** and **4** were treated with barbituric acid (**5**) or its 1,3-dimethyl derivative (**6**) in acetic acid at room temperature for 3-5 hours or at 80 °C for 0.5-2.5 hours to form the corresponding 2-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(4-hydroxy-2,6-dioxo-5-pyridinyl)propenoates **13-15** in 65-94% yield, and with indole (**7**) at room temperature for 6 days the corresponding 3-(3-indolyl)propenoate **16** in low yield. By treatment of **3** or **4** with other *C*-nucleophiles in acetic acid at room temperature, 80-100 °C or by refluxing for several hours the cyclic products, such as fused pyranones, pyridinones, pyrimidinones, and *N*-heteroaryl substituted imidazoles were obtained. In this respect, **3** and/or **4** were transformed with 2-pyridinylacetonitrile (**8**) into substituted amino-4*H*-quinolizin-4-one **17**, cyclohexane-1,3-dione (**9**) and its 5,5-dimethyl derivative (**10**) into 5,6,7,8-tetrahydro-2*H*,5*H*-benzo[*b*]pyran-2,5-diones **18** and **19**, 4-hydroxy-6-methyl-2*H*-pyran-2-one (**11**) into 2*H*,5*H*-pyrano[4,3-*b*]-pyran-2,5-dione **20**, 4-hydroxy-2*H*-benzo[*b*]pyran-2-one (**12**) into 2*H*,5*H*-pyrano[3,2-*c*]benzo[*b*]pyran-2,5-dione (**21**). Compound **18** gave by treatment with hydrazine in refluxing ethanol the corresponding 5-hydrazono derivative **22**, and 1-aminoquinolin-2(1*H*)-one derivative **23**, while from **19** only 5-hydrazono derivative **24** was formed. (Scheme 2).

N-Nucleophiles react in two different manners: with sterically unhindered heteroarylamines the reactions proceed according to path A and the fused pyrimidin-4-ones are formed, while with sterically hindered heteroarylamines the reactions proceed according to path B resulting in the formation of 1-heteroaryl-1*H*-imidazole-4-carboxylates. This phenomenon has been observed earlier in reactions with some other 2-[2,2-disubstituted ethenyl]amino-3-dimethylaminopropenoates [12]. Thus, compounds **3** and **4** react

Scheme 2

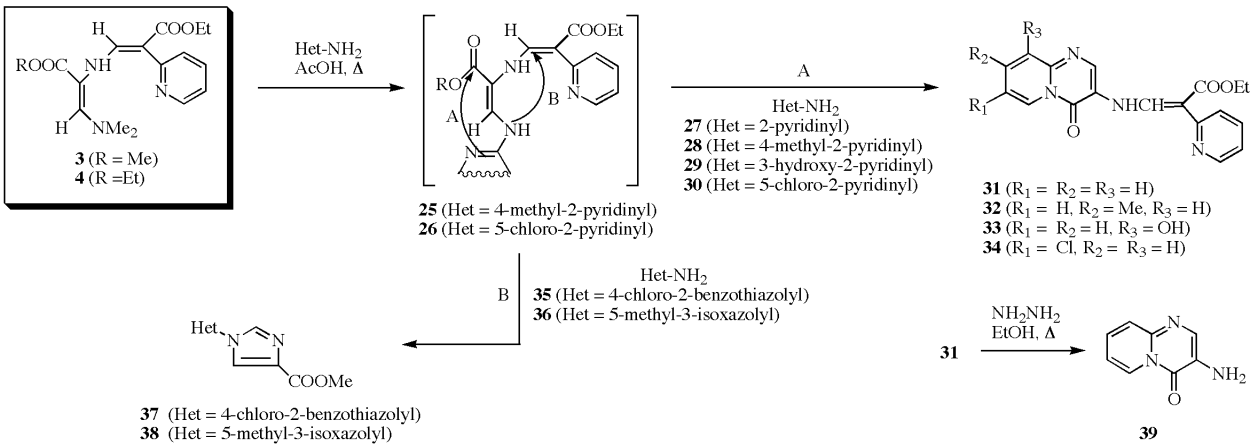


with *N*-nucleophiles, such as α-heteroarylamines 2-aminopyridine (**27**) and its derivatives **28-30**, to give 4*H*-pyrido[1,2-*a*]pyrimidin-4-one derivatives **31-34**, while compound **3** reacts with 2-amino-4-chlorobenzothiazole (**35**) and 3-amino-5-methylisoxazole (**36**) to form the corresponding *N*-heteroaryl-1*H*-imidazole-4-carboxylates **37** and **38**. Deprotection of compound **31** by heating with hydrazine in boiling ethanol produced the parent 3-amino-4*H*-pyrido[1,2-*a*]pyrimidin-4-one (**39**) [13]. (Scheme 3).

Structure Determination.

The structures of the compounds were determined on the basis of their mass spectra, elemental analyses for *C*, *H*, and *N* and ^{1}H nmr spectra. The chemical shifts for protons attached to the cyclic structure are in agreement with the

Scheme 3



data reported in the literature for other derivatives of these systems [6]. The proton attached at the newly fused pyridine ring in compound **17** and pyrimidine ring in compounds **31-34** and **37** appears in ^{1}H nmr spectra in all cases as a sharp singlet at δ = 7.68 ppm and δ = 8.33-8.38 ppm, respectively indicating that this proton is attached at the 2-position. In alternative structures, 2-ones, the proton at 4-position should have appeared as doublet due to the long-range coupling to proton at 9-position characteristic for pyrido[1,2-*a*]pyrimidin-2-ones [14]. Furthermore, the transformation of the corresponding 3-diazonium salts into alkyl 1-(substituted pyridin-2-yl)-1*H*-1,2,3-triazole-4-carboxylates, supported by an X-ray analyses, is an additional proof, which speaks in favor of 4-one structures [15]. The protons attached to the side chain show the following characteristics. The C*H*NH appear as doublets in the range of δ = 8.27-8.56 ppm and CHN*H* as doublets in the range of δ = 12.96-13.54 ppm with the coupling constants J_{CH-NH} = 11.4-12.8 Hz. From the chemical shifts one can conclude that the orientation around the C=C bond is *trans* and from the magnitude of coupling constants that the orientation around the CHNH is *trans* (*antiperiplanar*). (Figure 1).

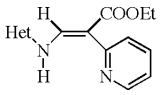


Figure 1

EXPERIMENTAL

Melting points were taken on a Kofler micro hot stage. The ^{1}H nmr spectra were obtained on a Bruker Avance DPX 300 (300 MHz) spectrometer in such solvent as dimethyl sulfoxide-d_6 and deuteriochloroform with tetramethylsilane as internal standard, MS spectra on an AutoSpecQ spectrometer, IR spectra on a Perkin-Elmer 1310 infrared spectrophotometer and elemental analyses for C, H, and N on a Perkin-Elmer CHN Analyser 2400. Experimental and analytical data are given in Tables 1 and 2.

Ethyl 3-Dimethylamino-2-(2-pyridinyl)propenoate (**2**) was Prepared According to the Procedure Described in the Literature [16].

Methyl and ethyl (*Z*)-2-[(*E*)-2-ethoxycarbonyl-2-(2-pyridinyl)-ethenyl]amino-3-dimethylaminopro-penoate **3** and **4**, methyl 2-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(4-methyl-2-pyridinyl)aminopropenoate (**25**) and methyl 2-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(5-chloro-2-pyri-dinyl)amino-propenoate (**26**) were prepared according to the procedures described in the literature [11].

Preparation of β-Heteroaryl-α,β-didehydro-α-amino Acid Derivatives **13-16**.

Method A.

To compounds **3** or **4** (0.5 mmole) the corresponding *C*-nucleophilic compounds **5-7** (0.5 mmole) and acetic acid (2 ml) were added and stirred at room temperature from 3 hours to 6 days. The formed precipitate was collected by filtration and washed with ethanol. The exception is the isolation of product **16**, where the volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added and the precipitate was collected by filtration.

Method B.

To compound **3** (0.5 mmole) the corresponding *C*-nucleophilic compounds **5, 6** (0.5 mmole) and acetic acid (2 ml) were added and the reaction mixture was stirred at 80 °C from 30 minutes to 2.5 hours. The formed precipitate was collected by filtration and washed with ethanol.

Methyl 2-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(4-hydroxy-2,6-dioxo-5-pyrimidinyl)-propenoate (**13**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and barbituric acid (**5**) (0.5 mmole, 64 mg) by method A, 3 hours and by method B, 30 minutes.

Ethyl 2-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(4-hydroxy-2,6-dioxo-5-pyrimidinyl)-propenoate (**14**).

This compound was prepared from compound **4** (0.5 mmole, 167 mg) and barbituric acid (**5**) (0.5 mmole, 64 mg) by method A, 5 hours.

Methyl 2-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(4-hydroxy-1,3-dimethyl-2,6-dioxo-5-pyrimidinyl)propenoate (**15**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 1,3-dimethylbarbituric acid (**6**) (0.5 mmole, 78 mg) by method A, 3 hours and by method B, 2.5 hours.

Table 1

Experimental and Analytical Data

Compound	Yield (%)	mp (°C)	Molecular formula Analyses	MS M+ (m/z)	IR (cm-1)
13	65[a]	239-242	$C_{18}H_{18}N_4O_7$	403 MH	3420 (OH)
	94[b]	from ethanol	Calcd: C, 52.56; H, 4.65; N, 13.62[g]		
			Found: C, 52.15; H, 4.60; N, 13.21		
14	93	224-227	$C_{19}H_{20}N_4O_7$	417 MH	3420 (OH)
		from ether	Calcd: C, 53.65; H, 4.98; N, 13.17[g]		
			Found: C, 53.91; H, 5.33; N, 12.91		
15	89[a]	227-229	$C_{20}H_{22}N_4O_7$	430	3440 (OH)
	69[b]	from	Calcd: C, 55.81; H, 5.15; N, 13.02	431 (MH)	
		ethanol/toluene	Found:C, 55.95; H, 4.92; N, 12.82		
16	6	181-182	$C_{22}H_{21}N_3O_4$		
		from ethanol	Calcd: C, 67.51; H, 5.41; N, 10.73		
			Found:C, 67.44; H, 5.75; N, 10.66		
17	26[a]	265-268	$C_{20}H_{16}N_4O_3$		
	53[b]	from ethanol	Calcd: C, 66.66; H, 4.48; N, 15.55		2210 (CN)
			Found:C, 66.96; H, 4.25; N, 15.45		
18	30[a]	176-178	$C_{19}H_{18}N_2O_5$		
	7[b]	from ethanol	Calcd: C, 64.40; H, 5.12; N, 7.91		
	7[d]		Found:C, 64.53; H, 5.33; N, 7.95		
19	56[a]	146-151	$C_{21}H_{22}N_2O_5$		
	44[b]	from ethanol	Calcd: C, 65.96; H, 5.80; N, 7.33		
	6[d]		Found:C, 66.22; H, 5.84; N, 7.39		
20	16[b]	225-228	$C_{19}H_{16}N_2O_6$		
	9[d]	from	Calcd: C, 61.95; H, 4.38; N, 7.61		
		ethanol/toluene	Found:C, 62.03; H, 4.43; N, 7.77		
21	30[b]	235-240	$C_{22}H_{16}N_2O_6$		
	22[d]	from	Calcd: C, 65.35; H, 3.99; N, 6.93		
		ethanol/toluene	Found:C, 65.68; H, 4.04; N, 6.97		
22	82	212-215	$C_{19}H_{20}N_4O_4$		
		from ethanol	Calcd: C, 61.95; H, 5.47; N, 15.21		
			Found:C, 62.09; H, 5.55; N, 15.23		
23	5	165-172	$C_{19}H_{22}N_6O_3$		
		from ethanol	Calcd: C, 58.30; H, 5.92; N, 21.45[g]	382	
			Found: C, 58.27; H, 5.58; N, 21.73		
24	37	211-213	$C_{21}H_{24}N_4O_4$	396	
		from ethanol	Calcd: C, 63.62; H, 6.10; N, 14.13	HRMS:	
			Found:C, 63.69; H, 6.02; N, 14.00	Calcd:396.179756	
				Found: 396.180550	
31	30	149-156	$C_{18}H_{16}N_4O_3$	336	
		from	Calcd: C, 63.43; H, 4.88; N, 16.44[f]	HRMS:	
		ethanol/toluene	Found: C, 63.46; H, 5.07; N, 15.61	Calcd: 336.122241	
				Found: 336.123040	
32	55[a]	173-179	$C_{19}H_{18}N_4O_3$		
	21[e]	from	Calcd: C, 65.13; H, 5.18; N, 15.99		
	15[d]	ethanol/toluene	Found:C, 64.83; H, 5.38; N, 15.76		
33	17[b]	208-212	$C_{18}H_{16}N_4O_4$		
	13[d]	from	Calcd: C, 61.36; H, 4.58; N, 15.90		
		ethanol/toluene	Found:C, 61.32; H, 4.68; N, 15.94		
34	26[a]	201-203	$C_{18}H_{15}ClN_4O_3$		
	18[c]	from	Calcd: C, 58.31; H, 4.08; N, 15.11		
	12[d]	ethano/toluene	Found:C, 58.15; H, 4.08; N, 15.08		
37	67	185-186	$C_{12}H_8ClN_3O_2S$		
		from ethanol	Calcd: C, 49.07; H, 2.75; N, 14.31	293	
			Found:C, 49.07; H, 2.72; N, 14.32		
38	37	197-198	$C_9H_9N_3O_3$		
		from ethanol	Calcd: C, 52.17; H, 4.38; N, 20.28		
			Found:C, 52.35; H, 4.47; N, 19.98		

[a] By method A. [b] By method B. [c] By method C, from compound **5.** [d] By method C, from compound **6.** [e] From compound **5.** [f] Calcd. for compound **33** + 0.25 H_2O. [g] Calcd. for compound **13**, **14**, and **33** + 0.5 H_2O.

Table 2

^{1}H NMR Data

Compound	300 MHz Solvent	δ (tetramethylsilane)
13	DMSO-d_6	1.29 (3H, t, $COOCH_2CH_3$), 3.74 (3H, s, COOMe), 4.33 (2H, q, $COOCH_2CH_3$), 7.56 (2H, br.s, H_5, H_4), 7.91 (1H, d, H_3), 8.27-8.30 (1H, m, H_6), 8.35 (1H, br.s, CH), 8.55 (1H, br.s, CHNH), 9.96 (2H, s, 2x H), 12.54 (1H, br.s, CHNH), 14.32 (1H, br.s, OH), J_{CH2CH3} = 6.8 Hz.
14	DMSO-d_6	1.26, 1.29 (6H, 2x t, 2x $COOCH_2CH_3$), 4.21 (2H, q, $COOCH_2CH_3$), 4.31 (2H, q, $COOCH_2CH_3$), 7.47-7.53 (2H, m, H_5, H_4), 7.90 (1H, d, H_3), 8.23 (1H, dd, H_6), 8.32 (1H, s, CH), 8.54 (1H, br.s, CHNH), 9.90 (2H, s, 2x NH), 11.96 (1H, br.s, CHNH), J_{H3H4}= 8.7 Hz, J_{H5H6} = 7.7 Hz, J_{CH2CH3} = 6.9 Hz. OH exchanged.
15	DMSO-d_6	1.26 (3H, t, $COOCH_2CH_3$), 3.11 (6H, s, 2x Me), 3.75 (3H, s, COOMe), 4.22 (2H, q, $COOCH_2CH_3$), 7.40-7.80 (2H, m, H_4, H_5), 7.93 (1H, d, H_3), 8.20-8.55 (3H, m, H_6, CH, CHNH), 12.42 (1H, br.s, CHNH), 14.64 (1H, br.s, OH), J_{H3H4} = 8,7 Hz, J_{CH2CH3} = 7.1 Hz.
	$CDCl_3$	1.45 (3H, t, $COOCH_2CH_3$), 3.35 (6H, s, 2x Me), 3.83 (3H, s, COOMe), 4.58 (2H, q, $COOCH_2CH_3$), 7.11 (1H, ddd, H_5), 7.69 (1H, ddd, H_3), 7.92 (1H, ddd, H_4), 7.95 (1H, ddd, H_6), 8.16 (1H, s, CH), 8.88 (1H, br.s, CHNH), 14.64 (1H, br.s, OH), J_{H3H4} = 9.1 Hz, J_{H3H5} = 1.0 Hz, J_{H3H6} = 1.0 Hz, J_{H4H5} = 7.2 Hz, J_{H4H6} = 1.7 Hz, J_{H5H6} = 6.2 Hz, J_{CH2CH3} = 7.1 Hz. OH exchanged.
16	DMSO-d_6	1.19 (3H, t, $COOCH_2CH_3$), 3.85 (3H, s, COOMe), 4.13 (2H, q, $COOCH_2CH_3$), 7.14 (1H, ddd, H_5), 7.19 (1H, ddd, H_5'), 7.23 (1H, ddd, H_6), 7.49 (1H, ddd, H_7), 7.55 (1H, s, CH), 7.76 (1H, ddd, H_4), 7.82 (1H, ddd, H_4'), 8.11 (1H, d, H_2), 8.24 (1H, ddd, H_3'), 8.37 (1H, d, CHNH), 8.45 (1H, ddd, H_6'), 11.91 (1H, br.s, H_1), 12.27 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 1.9 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H4H5} = 7.9 Hz, J_{H4H6} = 1.2 Hz, J_{H4H7} = 1.0 Hz, J_{H5H6} = 7.0 Hz, J_{H5H7} = 1.0 Hz, J_{H6H7} = 8.1 Hz, J_{H1H2} = 2.4 Hz, J_{CHNH} = 12.6 Hz, J_{CH2CH3} = 7.1 Hz.
	$CDCl_3$	1.28 (3H, t, $COOCH_2CH_3$), 3.92 (3H, s, COOMe), 4.23 (2H, q, $COOCH_2CH_3$), 6.99 (1H, ddd, H_5'), 7.24 (1H, ddd, H_5), 7.29 (1H, ddd, H_6), 7.42 (1H, ddd, H_7), 7.63 (1H, s, CH), 7.66 (1H, ddd, H_4'), 7.82 (1H, ddd, H_4), 8.09 (1H, d, H_2), 8.33 (1H, ddd, H_6'), 8.37 (1H, ddd, H_3'), 8.40 (1H, d, CHNH), 8.59 (1H, br.s, H_1), 12.46 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H4H5} = 7.7 Hz, J_{H4H6} = 1.3 Hz, J_{H4H7} = 1.0 Hz, J_{H5H6} = 7.1 Hz, J_{H5H7} = 1.3 Hz, J_{H6H7} = 8.0 Hz, J_{H1H2} = 2.9 Hz, J_{CHNH} = 12.4 Hz, J_{CH2CH3} = 7.1 Hz.
17	$CDCl_3$	1.43 (3H, t, $COOCH_2CH_3$), 4.36 (2H, q, $COOCH_2CH_3$), 7.11 (1H, ddd, H_5'), 7.15 (1H, ddd, H_7), 7.48 (1H, ddd, H_8), 7.68 (1H, s, H_2), 7.70 (1H, ddd, H_4'), 7.93 (1H, ddd, H_9), 8.32 (1H, ddd, H_3'), 8.42 (1H, d, CHNH), 8.70 (1H, ddd, H_6'), 9.11 (1H, ddd, H_6), 13.41 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 1.9 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H6H7} = 7.4 Hz, J_{H6H8} = 1.2 Hz, J_{H6H9} = 1.0 Hz, J_{H7H8} = 6.6 Hz, J_{H7H9} = 1.4 Hz, J_{H8H9} = 9.0 Hz, J_{CHNH} = 12.2 Hz, J_{CH2CH3} = 7.1 Hz.
18	$CDCl_3$	1.40 (3H, t, $COOCH_2CH_3$), 2.14-2.23 (2H, m, CH_2), 2.57-2.61 (2H, m, CH_2), 2.88 (2H, t, CH_2), 4.35 (2H, q, $COOCH_2CH_3$), 7.10 (1H, ddd, H_5'), 7.36 (1H, s, H_4), 7.70 (1H, ddd, H_4'), 8.28 (1H, ddd, H_3'), 8.29 (1H, d, CHNH), 8.63 (1H, ddd, H_6'), 13.13 (1H, d, CHNH), $J_{H3'H4'}$ = 8.5 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 4.9 Hz, J_{CH2CH2} = 6.4 Hz, J_{CHNH} = 12.4 Hz, J_{CH2CH3} = 7.1 Hz.
19	$CDCl_3$	1.16 (6H, s, 2x 7-Me), 1.39 (3H, t, $COOCH_2CH_3$), 2.45 (2H, s, CH_2), 2.74 (2H, s, CH_2), 4.34 (2H, q, $COOCH_2CH_3$), 7.10 (1H, ddd, H_5'), 7.35 (1H, s, H_4), 7.69 (1H, ddd, H_4'), 8.28 (1H, ddd, H_3'), 8.29 (1H, d, CHNH), 8.63 (1H, ddd, H_6'), 13.14 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.3 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{CHNH} = 11.6 Hz, J_{CH2CH3} = 7.1 Hz.
20	$CDCl_3$	1.41 (3H, t, $COOCH_2CH_3$), 2.36 (3H, d, 7-Me), 4.36 (2H, q, $COOCH_2CH_3$), 6.23 (1H, qd, H_8), 7.12 (1H, ddd, H_5'), 7.37 (1H, br.s, H_4), 7.71 (1H, ddd, H_4'), 8.27 (1H, d, CHNH), 8.31 (1H, ddd, H_3'), 8.63 (1H, ddd, H_6'), 13.39 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H4H8} = 0.7 Hz, $J_{7\text{-}MeH8}$ = 0.9 Hz, J_{CHNH} = 11.4 Hz, J_{CH2CH3} = 7.1 Hz.
21	$CDCl_3$	1.42 (3H, t, $COOCH_2CH_3$), 4.38 (2H, q, $COOCH_2CH_3$), 7.15 (1H, ddd, H_5'), 7.41 (1H, dd, H_7), 7.42 (1H, ddd, H_9), 7.49 (1H, s, H_4), 7.62 (1H, ddd, H_8), 7.73 (1H, ddd, H_4'), 8.04 (1H, dd, H_{10}), 8.31 (1H, d, CHNH), 8.32 (1H, ddd, H_3'), 8.66 (1H, ddd, H_6'), 13.54 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 4.9 Hz, J_{H7H8} = 8.3 Hz, J_{H7H9} = 1.2 Hz, J_{H8H9} = 7.3 Hz, J_{H8H10} = 1.5 Hz, J_{H9H10} = 8.2 Hz, J_{CHNH} = 11.8 Hz, J_{CH2CH3} = 7.1 Hz.
22	$CDCl_3$	1.40 (3H, t, $COOCH_2CH_3$), 2.14-2.22 (2H, m, CH_2), 2.55-2.59 (2H, m, CH_2), 3.13-3.17 (2H, m, CH_2), 4.35 (2H, q, $COOCH_2CH_3$), 5.12 (2H, s, =N-NH_2), 7.08 (1H, ddd, H_5'), 7.68 (1H, ddd, H_4'), 7.70 (1H, s, H_4), 8.29 (1H, ddd, H_3'), 8.42 (1H, d, CHNH), 8.64 (1H, ddd, H_6'), 13.05 (1H, d, CHNH), $J_{H3'H4'}$ = 8.6 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.1 Hz, $J_{H4'H5'}$ = 7.2 Hz, $J_{H4'H6'}$ = 1.9 Hz, $J_{H5'H6'}$ = 4.9 Hz, J_{CHNH} = 12.4 Hz, J_{CH2CH3} = 7.2 Hz.
23	$CDCl_3$	1.39 (3H, t, $COOCH_2CH_3$), 1.96-2.04 (2H, m, CH_2), 2.41-2.46 (2H, m, CH_2), 2.96-3.00 (2H, m, CH_2), 4.33 (2H, q, $COOCH_2CH_3$), 5.11 (2H, s, =N-NH_2), 5.26 (2H, br.s, NH_2), 7.05 (1H, ddd, H_5'), 7.66 (1H, ddd, H_4'), 7.83 (1H, s, H_4), 8.25 (1H, ddd, H_3'), 8.46 (1H, d, CHNH), 8.66 (1H, ddd, H_6'), 12.96 (1H, d, CHNH), $J_{H3'H4'}$ = 8.6 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 0.8 Hz, $J_{H4'H5'}$ = 7.3 Hz, $J_{H4'H6'}$ = 1.9 Hz, $J_{H5'H6'}$ = 4.9 Hz, J_{CHNH} = 12.8 Hz, J_{CH2CH3} = 7.2 Hz.
24	$CDCl_3$	1.14 (6H, s, 2x CH_3), 1.40 (3H, t, $COOCH_2CH_3$), 2.43 (2H, s, CH_2), 3.01 (2H, s, CH_2), 4.34 (2H, q, $COOCH_2CH_3$), 5.13 (2H, s, =N-NH_2), 7.08 (1H, ddd, H_5'), 7.68 (1H, ddd, H_4'), 7.69 (1H, s, H_4), 8.30 (1H, ddd, H_3'), 8.42 (1H, d, CHNH), 8.64 (1H, ddd, H_6'), 13.06 (1H, br.s, CHNH), $J_{H3'H4'}$ = 8.3 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.1 Hz, $J_{H4'H5'}$ = 7.5 Hz, $J_{H4'H6'}$ = 1.9 Hz, $J_{H5'H6'}$ = 4.9 Hz, J_{CHNH} = 12.4 Hz, J_{CH2CH3} = 7.1 Hz.
31	$CDCl_3$	1.39 (3H, t, $COOCH_2CH_3$), 4.33 (2H, q, $COOCH_2CH_3$), 6.96 (1H, dd, H_7), 7.07 (1H, ddd, H_5'), 7.42 (1H, ddd, H_9), 7.68 (1H, ddd, H_4'), 8.33 (1H, ddd, H_3'), 8.38 (1H, s, H_2), 8.56 (1H, d, CHNH), 8.58 (1H, d, H_8), 8.66 (1H, ddd, H_6'), 8.91 (1H, d, H_6), 13.12 (1H, d, CHNH), $J_{H3'H4'}$ = 8.5 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H6H7} = 7.4 Hz, J_{H6H9} = 0.9 Hz, J_{H7H8} = 1.9 Hz, J_{H7H9} = 1.0 Hz, J_{H8H9} = 2.0 Hz, J_{CHNH} = 12.7 Hz, J_{CH2CH3} = 7.1 Hz.

Table 2 (continued)

Compound	300 MHz Solvent	δ (tetramethylsilane)
32	$CDCl_3$	1.39 (3H, t, $COOCH_2CH_3$), 2.47 (3H, 2x s, 8-Me), 4.33 (2H, q, $COOCH_2CH_3$), 6.96 (1H, dd, H_7), 7.07 (1H, ddd, H_5'), 7.42 (1H, dd, H_9), 7.68 (1H, ddd, H_4'), 8.33 (1H, ddd, H_3'), 8.38 (1H, s, H_2), 8.56 (1H, d, CHNH), 8.66 (1H, ddd, H_6'), 8.91 (1H, d, H_6), 13.12 (1H, d, CHNH), $J_{H3'H4'}$ = 8.4 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 4.9 Hz, J_{H6H7} = 7.4 Hz, J_{H6H9} = 0.9 Hz, J_{H7H9} = 2.0 Hz, J_{CHNH} = 12.5 Hz, J_{CH2CH3} = 7.1 Hz.
33	$CDCl_3$	1.40 (3H, t, $COOCH_2CH_3$), 4.34 (2H, q, $COOCH_2CH_3$), 7.01 (1H, dd, H_8), 7.05 (1H, dd, H_7), 7.09 (1H, ddd, H_5'), 7.69 (1H, ddd, H_4'), 8.31 (1H, ddd, H_3'), 8.33 (1H, s, H_2), 8.52 (1H, dd, H_6), 8.53 (1H, d, CHNH), 8.67 (1H, ddd, H_6'), 13.18 (1H, d, CHNH), $J_{H3'H4'}$ = 8.5 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H6H7} = 6.8 Hz, J_{H6H8} = 1.8 Hz, J_{H7H8} = 7.4 Hz, J_{CHNH} = 12.0 Hz, J_{CH2CH3} = 7.1 Hz. OH exchanged.
34	$CDCl_3$	1.40 (3H, t, $COOCH_2CH_3$), 4.34 (2H, q, $COOCH_2CH_3$), 7.10 (1H, ddd, H_5'), 7.46 (1H, dd, H_8), 7.58 (1H, dd, H_9), 7.70 (1H, ddd, H_4'), 8.31 (1H, ddd, H_3'), 8.38 (1H, s, H_2), 8.54 (1H, d, CHNH), 8.67 (1H, ddd, H_6'), 9.01 (1H, dd, H_6), 13.24 (1H, d, CHNH), $J_{H3'H4'}$ = 8.5 Hz, $J_{H3'H5'}$ = 1.1 Hz, $J_{H3'H6'}$ = 1.0 Hz, $J_{H4'H5'}$ = 7.4 Hz, $J_{H4'H6'}$ = 2.0 Hz, $J_{H5'H6'}$ = 5.0 Hz, J_{H6H8} = 2.3 Hz, J_{H6H9} = 0.8 Hz, J_{H8H9} = 9.5 Hz, J_{CHNH} = 12.4 Hz, J_{CH2CH3} = 7.1 Hz.
37	$CDCl_3$	3.96 (3H, s, COOMe), 7.39 (1H, dd, H_6'), 7.58 (1H, dd, H_7'), 7.77 (1H, dd, H_5'), 8.29 (1H, d, H_5), 8.38 (1H, d, H_2), J_{H2H5} = 1.4 Hz, $J_{H5'H7'}$ = 1.1 Hz, $J_{H5'H6'}$ = $J_{H6'H7'}$ = 8.0 Hz.
38	$CDCl_3$	2.53 (3H, d, 5'-Me), 3.94 (3H, s, COOMe), 6.23 (1H, q, H_4'), 8.02 (1H, d, H_5), 8.05 (1H, d, H_2), J_{H2H5} = 1.4 Hz, $J_{H4'\ 5\text{-}Me}$ = 0.9 Hz.

Methyl 2-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyll]amino-3-(3-indolyl)propenoate (**16**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and indole (**7**) (0.5 mmole, 59 mg) by method A, 6 days.

1-Cyano-3-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-4*H*-quinolizin-4-one (**17**).

To compound **3** (0.5 mmole, 160 mg) 2-pyridinylacetonitrile (**8**) (0.5 mmole, 59 mg) and acetic acid (2 ml) were added and stirred at 80 °C for 2.5 hours (method A) or at room temperature for 4 days (method B). The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added, the precipitate was collected by filtration and washed with ethanol.

Preparation of Pyranones **18**-**22**, **24**.

Method A.

To compound **3** (0.5 mmole) the corresponding *C*-nucleophilic compounds **9**, **10** (0.5 mmole) and acetic acid (2 ml) were added and stirred at room temperature from 5 hours to 5 days. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added and the precipitate was collected by filtration.

Method B.

To compound **3** (0.5 mmole) the corresponding *C*-nucleophilic compounds **9**-**12** (0.5 mmole) and acetic acid (2 ml) were added and the reaction mixture was stirred at 80-100 °C for 1.5-4 hours. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added and the precipitate was collected by filtration.

Method C.

To compound **4** (0.5 mmole) the corresponding *C*-nucleophilic compounds **9**-**12** (0.5 mmole) and acetic acid (2 ml) were added and refluxed for 5 hours. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added and the precipitate was collected by filtration.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-5,6,7,8-tetrahydro-2*H*,5*H*-benzo[*b*]pyran-2,5-dione (**18**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and cyclohexane-1,3-dione (**9**) (0.5 mmole, 56 mg) by method A, 5 hours, and by method B, 110 °C, 2 hours, and from compound **4** (0.5 mmole, 167 mg) and cyclohexane-1,3-dione (**9**) (0.5 mmole, 56 mg) by method C, 5 hours.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-7,7-dimethyl-5,6,7,8-tetrahydro-2*H*,5*H*-benzo[*b*]pyran-2,5-dione (**19**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 5,5-dimethylcyclohexane-1,3-dione (**10**) (0.5 mmole, 70 mg) by method A, 5 days, and by method B, 80 °C, 4 hours, and from compound **4** (0.5 mmole, 167 mg) and 5,5-dimethylcyclohexane-1,3-dione (**10**) (0.5 mmole, 70 mg) by method C, 5 hours.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-7-methyl-2*H*,5*H*-pyrano[4,3-*b*]pyran-2,5-dione (**20**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 4-hydroxy-6-methyl-2*H*-pyran-2-one (**11**) (0.5 mmole, 63 mg) by method B, 80 °C, 4 hours, and from compound **4** (0.5 mmole, 167 mg) and 4-hydroxy-6-methyl-2*H*-pyran-2-one (**11**) (0.5 mmole, 63 mg) by method C, 5 hours.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-2*H*,5*H*-pyrano[3,2-*c*]benzo[*b*]pyran-2,5-dione (**21**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 4-hydroxy-2*H*-benzo[*b*]pyran-2-one (**12**) (0.5 mmole, 81 mg) by method B, 110 °C, 1.5 hours, and from compound **4** (0.5 mmole, 167 mg) and 4-hydroxy-2*H*-benzo[*b*]pyran-2-one (**12**) (0.5 mmole, 81 mg) by method C, 5 hours.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-5-hydrazono-5,6,7,8-tetrahydro-2*H*-1-benzopy-ran-2-one (**22**).

To compound **18** (0.28 mmole, 100 mg) 4 mL 0.5 *M* solution of hydrazine hydrate in ethanol was added and the mixture was refluxed for 1 hour. After cooling, the precipitate that formed was collected by filtration and washed with ethanol. The filtrate was left in the refrigirator for one week. The precipitate, 1-amino-3-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-5-hydrazono-5,6,7,8-tetrahydro-quinolin-2(1*H*)-one (**23**), which was formed, was collected by filtration and washed with ethanol.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-5-hydrazono-7,7-dimethyl-5,6,7,8-tetrahydro-2*H*-1-benzopyran-2-one (**24**).

To compound **19** (0.44 mmole, 168 mg) 4 mL 0.5 *M* solution of hydrazine hydrate in ethanol was added and the mixture was refluxed for 1 hour. After cooling, the precipitate that formed was collected by filtration and washed with ethanol.

General Procedures for the Preparation of Pyrido[1,2-*a*]pyrimidin-4-ones **31**-**34**.

Method A.

Methyl 2-[2-ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-3-(heteroaryl)amino-propenoates **25** or **26** were dissolved in acetic acid (3-4 mL/1 mmole of starting compound) and the reaction mixtures were refluxed for 2.5-4 hours. The volatile compounds were evaporated *in vacuo*, ether (3 ml) was added, the precipitate was collected by filtration and washed with ether.

Method B.

To compound **3** (0.5 mmole) the corresponding heteroarylamines **27**, **29** (0.5 mmole) and acetic acid (2 ml) were added and stirred at 60-80 °C for 9 hours. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added, the precipitate was collected by filtration and washed with ethanol.

Method C.

To compounds **3** or **4** (0.5 mmole) the corresponding heteroarylamines **28**-**30** (0.5 mmole) and acetic acid (2 ml) were added and the reaxtion mixture was refluxed for 1-1.5 hour. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added, the precipitate was collected by filtration and washed with ethanol.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-4*H*-pyrido-[1,2-*a*]pyrimidin-4-one (**31**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 2-aminopyridine (**27**) (47 mg, 0.5 mmole) by method B, 9 hours.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-8-methyl-4*H*-pyrido[1,2-*a*]pyrimidin-4-one (**32**).

This compound was prepared from compound **25** (0.05 mmole, 20 mg) by method A, 4 hours, and from compound **4** (0.5 mmole, 167 mg) and 2-amino-4-methylpyridine (**28**) (0.5 mmole, 54 mg) by method C, 1.5 hours. Compound **32** was also prepared from compound **3** (0.5 mmole, 160 mg) and 2-amino-4-methylpyridine (**28**) (0.5 mmole, 54 mg). The reaction mixture was stirred at room temperature for 2 hours and afterwards at 110 °C for 8 hours. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added, the precipitate was collected by filtration and washed with ethanol.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-9-hydroxy-4*H*-pyrido[1,2-*a*]pyrimidin-4-one (**33**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 2-amino-3-hydroxypyridine (**29**) (0.5 mmole, 55 mg) by method B, 9 hours, and from compound **4** (0.5 mmole, 167 mg) and 2-amino-3-hydroxypyridine (**29**) (0.5 mmole, 55 mg) by method C, 1 hour.

3-[2-Ethoxycarbonyl-2-(2-pyridinyl)ethenyl]amino-7-chloro-4*H*-pyrido[1,2-*a*]pyrimidin-4-one (**34**).

This compound was prepared from compound **26** (0.5 mmole, 201 mg) by method A, 2.5 hours, from compound **3** (0.5 mmole, 160 mg) and 2-amino-5-chloropyridine (**30**) (0.5 mmole, 64 mg) by method C, 1 hour, and from compound **4** (0.5 mmole, 167 mg) and 2-amino-5-chloropyridine (**30**) (0.5 mmole, 64 mg) by method C, 1 hour.

General Procedure for the Preparation of *N*-Heteroaryl-1*H*-imidazol-4-carboxylates **37**, **38**.

To compound **3** (0.5 mmole) the corresponding heteroarylamines **35**, **36** (0.5 mmole) and acetic acid (2 ml) were added and the reaxtion mixtures were refluxed for 1-1.5 hour. The volatile compounds were evaporated *in vacuo*, ethanol (3 ml) was added, the precipitate was collected by filtration and washed with ethanol.

Methyl *N*-(4-chloro-2-benzothiazolyl)-1*H*-imidazole-4-carboxylate (**37**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 2-amino-4-chlorobenzothiazole (**35**) (0.5 mmole, 185 mg), 1 hour.

Methyl *N*-(5-Methyl-3-isoxazolyl)-1*H*-imidazole-4-carboxylate (**38**).

This compound was prepared from compound **3** (0.5 mmole, 160 mg) and 3-amino-5-methylisoxazole (**36**) (0.5 mmole, 50 mg), 1.5 hours.

3-Amino-4*H*-pyrido[1,2-*a*]pyrimidin-4-one (**39**).

To 3-[2-Ethoxycarbonyl-2-(2-pyridinyl)-ethenyl]amino-4*H*-pyrido[1,2-*a*]pyrimidin-4-one (**31**) (0.2 mmole, 67 mg) 0.5 *M* solution of hydrazine hydrate in ethanol (2 mL) was added and the mixture was refluxed for 5.5 hours. The volatile compounds were evaporated *in vacuo*, ethanol (3 mL) was added, the precipitate was collected by filtration and washed with ethanol in 67.1% yield, mp 177-178 °C (from ethanol), lit [13] mp 178-179 °C; ^{1}H nmr ($CDCl_3$): δ 4.04 (2H, br.s, NH_2), 6.99 (1H, ddd, H_7), 7.38 (1H, ddd, H_8), 7.52 (1H, ddd, H_9), 8.00 (1H, s, H_2), 8.88 (1H, ddd, H_6), J_{H6H7} = 7.3 Hz, J_{H6H8} = 1.5 Hz, J_{H6H9} = 0.9 Hz, J_{H7H8} = 6.5 Hz, J_{H7H9} = 1.4 Hz, J_{H8H9} = 9.1 Hz.

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