A Simple and General Method for the Synthesis of Novel Hetarenonaphthyridones

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Dedicated with best wishes to Professor M. Tišler on the occasion of his 70th birthday

Reaction of hetarylacetonitriles **1** with ethyl esters of chloro- or bromo-substituted pyridinecarboxylic acids in the presence of a base affords a series of novel heterocycle-annelated [1,6]-, [2,6]-, and [2,7]naphthyridones with a bridgehead nitrogen atom.

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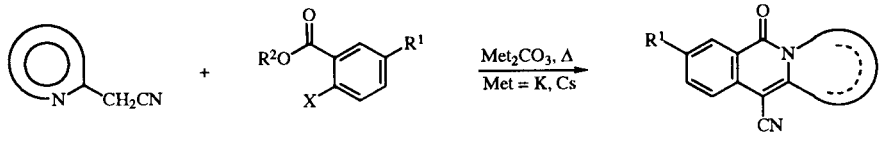
Introduction.

As shown recently, nucleophilic substitution reactions of hetarylacetonitriles of type **1** with 2-halobenzoates **2** in the presence of base give rise to the formation of fused isoquinolones of linear structure **3** shown in Scheme 1 [1]. In continuation of previous investigations of this reaction type, we became interested in replacement of the benzoic-acid-type building blocks by *N*-heterocyclic analogs. By starting from heteroaromatic carboxylic acid derivatives like nicotinoates or isonicotinoates, respectively, with an appropriate leaving group (like a halogen) in the *ortho* position to the alkoxycarbonyl function, a variety of hitherto unknown hetarenonaphthyridones should become accessible. Here we wish to report on the synthesis of a series of [*b*]-fused [2,7]naphthyridones, [*g*]-fused [1,6]naphthyridones, as well as a representative of a [*b*]-annelated [2,6]naphthyridone, using this methodology.

Scheme 1



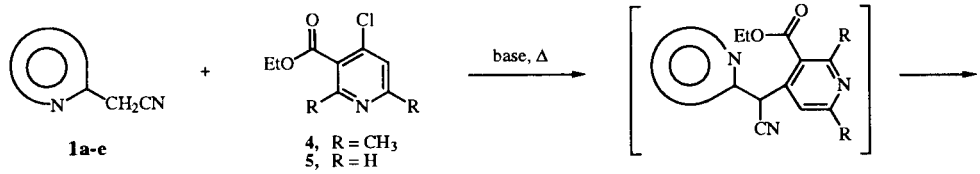
1 **2** **3**

$R^1 = H, NO_2$ $R^2 = alkyl$ X = halogen

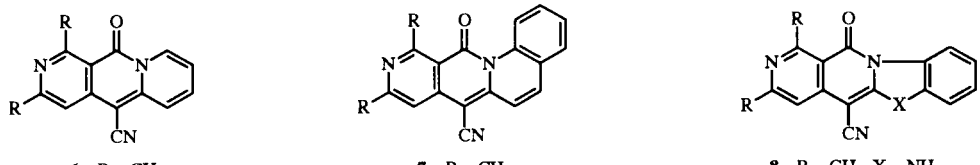
Results and Discussion.

In a first series of experiments, various hetarylacetonitriles featuring a ring nitrogen atom in the *ortho* position to the CH_2CN side chain [2-pyridylacetonitrile (**1a**), 2-quinolinylacetonitrile (**1b**), 2-benzimidazolylacetonitrile (**1c**), (1-methyl-2-benzimidazolyl)acetonitrile (**1d**), and 2-benzothiazolylacetonitrile (**1e**)] were treated with ethyl 2,6-dimethyl-4-chloronicotinoate (**4**) in dimethylformamide solution in the presence of a base. After 1.5 to 3 hours of refluxing, followed by aqueous work-up, the target naphthyridones **6-10** were isolated as almost insoluble solids in good to high yields. In most cases, employment of

Scheme 2



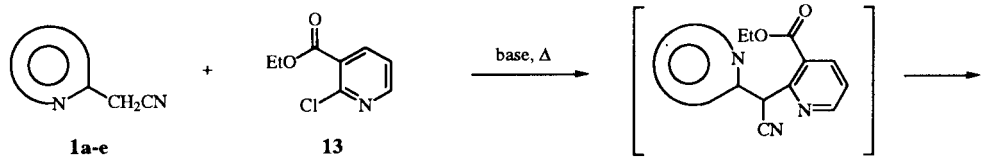
1a-e **4**, R = CH_3; **5**, R = H

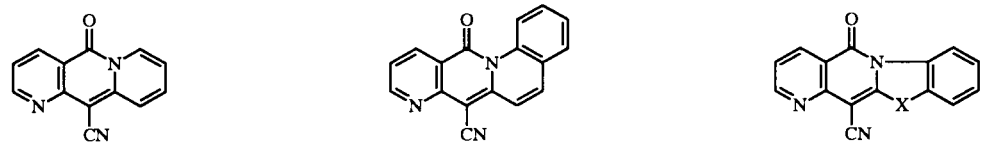


6, R = CH_3 **7**, R = CH_3

8, R = CH_3, X = NH
9, R = CH_3, X = NCH_3
10, R = CH_3, X = S
11, R = H, X = NCH_3
12, R = H, X = S

Scheme 3





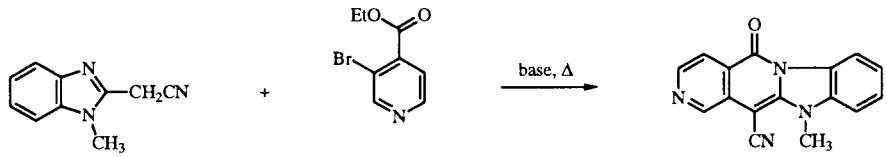
potassium *tert*-butoxide gave the best results, but also weaker bases like potassium carbonate, cesium carbonate, and 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU) were found to be suitable to effect this reaction.

The formation of the naphthyridine ring can be rationalized by nucleophilic attack of the carbanion which is generated from the hetarylacetonitrile methylene group at the pyridine carbon atom bearing the chloro substituent and - after loss of chloride - spontaneous cyclization as a result of intramolecular *N*-acylation of the heterocycle by the ester function as shown in Scheme 2.

been isolated earlier [4,5] when *o*-halogeno-substituted nicotinic or isonicotinic acid chlorides were employed as (more reactive) acylating agents.

Compounds **6-12**, **16** and **19** are representatives of new ring systems. The condensed naphthyridones thus obtained may also be regarded as interesting building blocks for the construction of other polyfunctional heterocyclic compounds, as exemplified by the conversion of 5-cyano-6,12-dihydro-1,3,6-trimethylbenzimidazo[1,2-*b*][2,7]naphthyrid-12-one (**9**) into the [1,2,4]triazolo[2,3-*b*][2,7]-naphthyridone **21**. The latter tricycle, which represents

Scheme 4



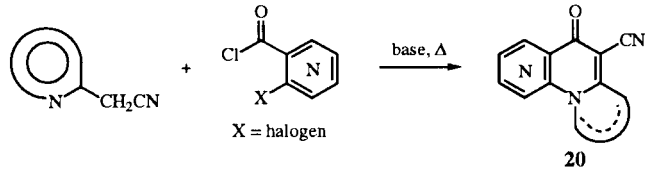
In a similar fashion, ethyl 4-chloronicotinoate (**5**) was transformed into the [*b*]-annelated [2,7]-naphthyridones **11** and **12** in yields of 78% and 86%, respectively (Scheme 2). When ethyl 2-chloronicotinoate (**13**) was employed as the starting material, the [*g*]-fused [1,6]-naphthyridones **14-17** were obtained (Scheme 3) [2]. To further demonstrate the versatility of this approach, also a representative of a [*b*]-annelated [2,6]-naphthyridone (compound **19**) was prepared, starting from ethyl 3-bromoisonicotinoate (**18**) and (1-methyl-2-benzimidazolyl)acetonitrile (**1d**) (Scheme 4).

It should be noted, that in all cases only linear annelated ring systems were obtained, although - in principle - the reaction might also take an alternative course: arylation of the ring nitrogen atom of the hetarylacetonitrile component and acylation of the activated methylene group would lead to structures of type **20** with nonlinear ring annelation. Compounds of the latter type (condensed [1,6]-, [1,7]-, and [1,8]naphthyridines; Scheme 5) had

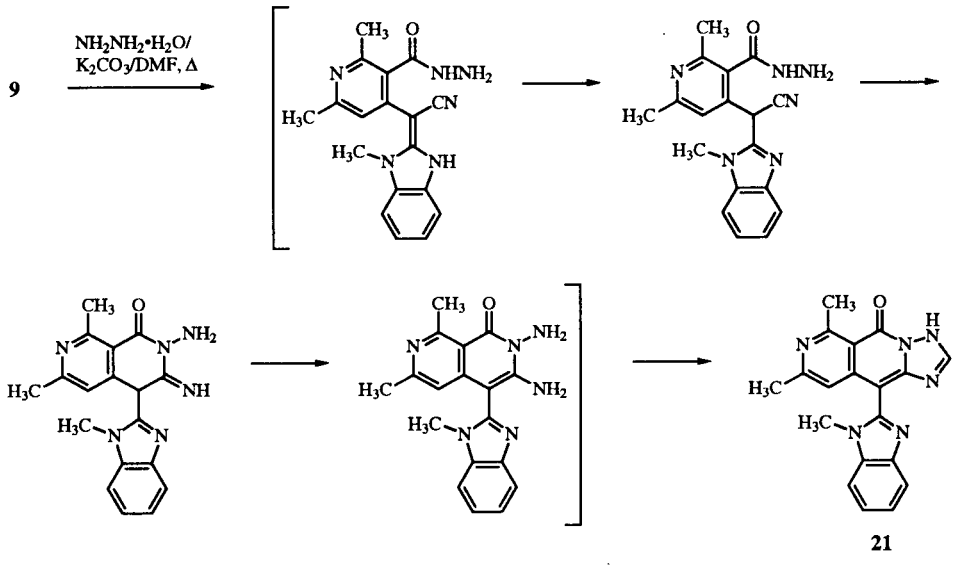
another new heterocyclic system [6], can be obtained simply by refluxing **9** with hydrazine hydrate in the presence of potassium carbonate in dimethylformamide solution - conditions which have been recently used for similar transformations in the isoquinolone series [7]. Obviously, this ring transformation is initiated by hydrazinolytic opening of the lactam ring, followed by addition of the hydrazone N-H to the nitrile function and subsequent condensation of the intermediate *vic*-diamino compound thus formed with one equivalent of the solvent (see Scheme 6).

In conclusion, it is demonstrated that the reaction of *o*-halogenosubstituted pyridinecarboxylic acid esters with

Scheme 5



Scheme 6



hetarylacetonitriles under basic conditions offers a simple and versatile method for the synthesis of condensed naphthyridones with a bridgehead nitrogen atom. The structures of all new compounds are supported by their ir and nmr spectra as well as by their microanalyses; details are given in the Experimental and in Tables 1 and 2.

Table 1

Physical and Analytical Data for Compounds **6-12**, **14-17**, and **19**

Compound No.	Method [a]	Reaction time (h)	Yield (%)	Mp, °C Recrystallization solvent	Molecular Formula	Analysis, % Calcd./Found C	H	N
6	A	1.5	82	247 DMF	$C_{15}H_{11}N_3O$	72.28 72.04	4.45 4.37	16.86 16.78
7	A	3	64	243 DMF	$C_{19}H_{13}N_3O$ x 0.1 H_2O	75.78 75.68	4.42 4.26	13.95 14.11
8	A	2	85	>330	$C_{17}H_{12}N_4O$	70.82	4.20	19.43
	B	2	47	DMF		70.64	4.02	19.29
9	A	1.5	87	321	$C_{18}H_{14}N_4O$	71.51	4.67	18.53
	B	3	47	DMF		71.45	4.61	18.74
	C	3	54					
10	A	2	51	287	$C_{17}H_{11}N_3OS$	66.87	3.63	13.76
	B	3	58	DMF		66.60	3.53	13.58
11	A	2	78	>330	$C_{16}H_{10}N_4O$	70.07	3.67	20.43
	B	2	63	DMF		69.86	3.54	20.35
12	B	2	86	276 1-propanol	$C_{15}H_7N_3OS$	64.97 64.76	2.55 2.63	15.15 15.13
14	B	2	44	283 [b] 1,4-dioxane	$C_{13}H_7N_3O$	70.58 70.31	3.19 3.41	18.99 18.89
15	B	2	37	244 2-propanol	$C_{17}H_9N_3O$	75.27 74.99	3.34 3.06	15.49 15.45
16	A	2	88	292-293	$C_{16}H_{10}N_4O$	69.61	3.72	20.29
	B	3	75	DMF	x 0.1 H_2O	69.50	3.71	20.32
	C	2	58					
17	B	2	64	275 1,4-dioxane	$C_{15}H_7N_3OS$ x 0.2 H_2O	64.14 64.04	2.66 2.62	14.96 14.89
19	B	4	52	296	$C_{16}H_{10}N_4O$	70.07	3.67	20.43
	D	3	55	DMF		69.89	3.56	20.24

[a] See Experimental. [b] Lit[3] mp 275° (from acetic acid).

Table 2

IR and ^{1}H NMR Spectral Data for Compounds **6-12**, **14-17**, and **19**

Compound No.	IR (cm^{-1})	^{1}H NMR (δ, ppm)
6	3138, 3038, 2212, 1686, 1636, 1600, 1566, 1510, 1146, 770	8.98 (d, J = 7.2 Hz, 1H, 9-H), 7.90-7.70 (m, 2H, 6-H, 7-H), 7.29 (s, 1H, 4-H), 7.22-7.14 (m, 1H, 8-H), 2.96 (s, 3H, 1-CH_3), 2.56 (s, 3H, 3-CH_3)
7	3076, 2992, 2214, 1696, 1590, 1574, 1532, 1024, 812, 754	8.88 (d, J = 9.0 Hz, 1H, 1-H), 7.93 (d, J = 9.3 Hz, 1H, 5-H), 7.83 (dd, J = 7.5 Hz and 1.5 Hz, 1H, 4-H), 7.68-7.61 (m, 1H, 2-H), 7.57-7.51 (m, 1H, 3-H), 7.48 (d, J = 9.3 Hz, 1H, 6-H), 7.27 (s, 1H, 8-H), 2.94 (s, 3H, 11-CH_3), 2.56 (s, 3H, 9-CH_3)
8	3260, 3194, 3078, 2984, 2212, 1682, 1660, 1610, 1560, 1280, 1250, 782, 764, 750	13.00-12.70 (broad s, 1H, NH), 8.39 (d, J = 7.8 Hz, 1H, 10-H), 7.61 (d, J = 7.8 Hz, 1H, 7-H), 7.41 (t, J = 7.8 Hz, 1H, 8-H), 7.25 (t, J = 7.8 Hz, 1H, 9-H), 7.05 (s, 1H, 4-H), 2.92 (s, 3H, 1-CH_3), 2.41 (s, 3H, 3-CH_3)
9	3120, 3030, 2998, 2928, 2202, 1684, 1580, 1530, 1160, 1120, 1026, 848, 756	8.65 (d, J = 8.1 Hz, 1H, 10-H), 7.76 (d, J = 8.1 Hz, 1H, 7-H), 7.59 (t, J = 8.1 Hz, 1H, 8-H), 7.43 (t, J = 8.1 Hz, 1H, 9-H), 7.29 (s, 1H, 4-H), 4.10 (s, 3H, 6-CH_3), 2.99 (s, 3H, 1-CH_3), 2.54 (s, 3H, 3-CH_3)
10	3124, 2994, 2924, 2214, 1696, 1586, 1572, 1520, 1454, 1276, 1256, 852, 766	8.96 (d, J = 7.5 Hz, 1H, 10-H), 8.10 (d, J = 7.5 Hz, 1H, 7-H), 7.65-7.54 (m, 2H, 8-H, 9-H), 7.27 (s, 1H, 4-H), 2.99 (s, 3H, 1-CH_3), 2.57 (s, 3H, 3-CH_3)
11	3128, 3034, 2196, 1694, 1594, 1538, 1476, 1032, 782, 748	9.40 (s, 1H, 1-H), 8.72 (d, J = 6.0 Hz, 1H, 3-H), 8.65 (d, J = 7.8 Hz, 1H, 10-H), 7.81(d, J = 7.8 Hz, 1H, 7-H), 7.63 (t, J = 7.8 Hz, 1H, 8-H), 7.57 (d, J = 6.0 Hz, 1H, 4-H), 7.48 (t, J = 7.8 Hz, 1H, 9-H), 4.14 (s, 3H, 6-CH_3)
12	3120, 3022, 2214, 1696, 1594, 1520, 1454, 1238, 1022, 750	9.50 (s, 1H, 1-H), 9.00 (d, J = 7.8 Hz, 1H, 10-H), 8.87 (d, J = 5.7 Hz, 1H, 3-H), 8.14 (d, J = 7.8 Hz, 1H, 7-H), 7.70-7.57 (m, 3H, 4-H, 8-H, 9-H)
14	3126, 3100, 2216, 1690, 1634, 1588, 1526, 1426, 778	9.10 (dd, J = 4.2 Hz and 1.8 Hz, 1H, 2-H), 8.96 (d, J = 7.2 Hz, 1H, 7-H), 8.71 (dd, J = 8.4 Hz and 1.8 Hz, 1H, 4-H), 7.88 (d, J = 8.1 Hz, 1H, 10-H), 7.85-7.75 (m, 1H, 9-H), 7.58 (dd, J = 8.4 Hz and 4.2 Hz, 1H, 3-H), 7.23-7.15 (m, 1H, 8-H)
15	3160, 3080, 2222, 1682, 1586, 1544, 1466, 1442, 1366, 812, 780, 746	9.28 (d, J = 8.7 Hz, 1H, 1-H), 9.09 (dd, J = 4.5 Hz and 1.8 Hz, 1H, 9-H), 8.72 (dd, J = 8.1 Hz and 1.8 Hz, 1H, 11-H), 7.95 (d, J = 9.6 Hz, 1H, 5-H), 7.88 (dd, J = 7.5 Hz and 1.5 Hz, 1H, 4-H), 7.73-7.63 (m, 3H, 2-H, 6-H, 10-H), 7.62-7.55 (m, 1H, 3-H)
16	3068, 2202, 1676, 1598, 1554, 1462, 1430, 1306, 776, 760	8.94 (dd, J = 4.5 Hz and 1.8 Hz, 1H, 2-H), 8.62-8.56 (m, 2H, 4-H, 7-H), 7.74 (d, J = 7.8 Hz, 1H, 10-H), 7.58 (t, J = 7.8 Hz, 1H, 9-H), 7.45-7.38 (m, 2H, 3-H, 8-H), 4.11 (s, 3H, 11-CH_3)
17	3068, 2996, 2220, 1682, 1590, 1544, 1458, 1298, 786, 770	9.03 (dd, J = 4.5 Hz and 1.8 Hz, 1H, 2-H), 8.95 (dd, J = 7.5 Hz and 1.8 Hz, 1H, 7-H), 8.68 (dd, J = 8.1 Hz and 1.8 Hz, 1H, 4-H), 8.10 (dd, J = 6.9 Hz and 2.1 Hz, 1H, 10-H), 7.65-7.54 (m, 3H, 3-H, 8-H, 9-H)
19	3054, 2200, 1686, 1618, 1602, 1546, 1476, 1422, 782, 756	9.12 (s, 1H, 1-H), 8.64 (d, J = 8.1 Hz, 1H, 7-H), 8.58 (d, J = 5.4 Hz, 1H, 3-H), 8.13 (d, J = 5.4 Hz, 1H, 4-H), 7.77 (d, J = 8.1 Hz, 1H, 10-H), 7.62 (t, J = 8.1 Hz, 1H, 9-H), 7.45 (t, J = 8.1 Hz, 1H, 8-H), 4.12 (s, 3H, 11-CH_3)

EXPERIMENTAL

Melting points were determined in a capillary tube and are uncorrected. The ir spectra were recorded for potassium bromide pellets on a Perkin-Elmer 1605 FT-IR spectrophotometer. The ^{1}H nmr spectra were obtained on a Varian Unity*plus* 300 (300 MHz) spectrometer, using deuteriodimethyl sulfoxide as solvent. For tlc, Merck aluminium sheets pre-coated with Kieselgel 60 F_{254} were used (eluent: chloroform/methanol, 9:1). Elemental analyses were performed at the Microanalytical Laboratory, Institute of Physical Chemistry, University of Vienna.

Potassium carbonate and cesium carbonate were freshly dried over an open flame and ground before use. The following hetarylacetonitriles were prepared according to literature procedures: 2-quinolinylacetonitrile (**1b**) [8], 2-benzimidazolylacetonitrile (**1c**) [9], (1-methyl-2-benzimidazolyl)acetonitrile (**1d**) [10], and 2-benzothiazolylacetonitrile (**1e**) [11].

The required pyridinecarboxylic acid esters were obtained following known procedures: ethyl 2,6-dimethyl-4-chloronicotinoate (**4**) [12], ethyl 4-chloronicotinoate (**5**) [13], ethyl 3-bromoisonicotinoate (**18**) [14,15], and ethyl 2-chloronicotinoate (**13**) [16].

General Procedure for the Preparation of the Condensed Naphthyridones **6-12**, **14-17**, and **19**.

Method A.

A mixture of the hetarylacetonitrile of type **1** (10 mmoles), a suitable ester, **4**, **5**, **13**, or **18**, respectively (11 mmoles) and 1.68 g (15 mmoles) of potassium *tert*-butoxide in 30 ml of dimethylformamide was stirred at reflux temperature for 1.5 to 4 hours. After cooling, the solvent was removed under reduced pressure. Water (25 ml) was added, and the reaction mixture was neutralized (*p*H 6-7) by addition of acetic acid. The precipitate was collected by filtration, washed with cold water, dried and recrystallized from a suitable solvent. For reaction times and yields, physical, analytical, and spectral data see Tables 1 and 2.

Method B.

The same reaction conditions were applied except that potassium carbonate (2.07 g, 15 mmoles) was used as a base.

Method C.

The same reaction conditions were applied except that 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU) (2.28 g, 15 mmoles) was used as a base.

Method D.

The same reaction conditions were applied except that cesium carbonate (4.89 g, 15 mmoles) was used as a base.

5-Cyano-1,3-dimethyl-11*H*-pyrido[1,2-*b*][2,7]naphthyrid-11-one (**6**).

This compound was obtained from 2-pyridylacetonitrile (**1a**) and the ester **4** as orange-red crystals.

7-Cyano-9,11-dimethyl-12*H*-quino[1,2-*b*][2,7]naphthyrid-12-one (**7**).

This compound was obtained from 2-quinolinylacetonitrile (**1b**) and the ester **4** as yellow-brown crystals.

5-Cyano-6,12-dihydro-1,3-dimethylbenzimidazo[1,2-*b*][2,7]-naphthyrid-12-one (**8**).

This compound was obtained from 2-benzimidazolylacetonitrile (**1c**) and the ester **4** as yellow crystals.

5-Cyano-6,12-dihydro-1,3,6-trimethylbenzimidazo[1,2-*b*][2,7]-naphthyrid-12-one (**9**).

This compound was obtained from (1-methyl-2-benzimidazolyl)acetonitrile (**1d**) and the ester **4** as almost colorless crystals.

5-Cyano-1,3-dimethyl-12*H*-benzothiazolo[3,2-*b*][2,7]naphthyrid-12-one (**10**).

This compound was obtained from 2-benzothiazolylacetonitrile (**1e**) and the ester **4** as almost colorless crystals.

5-Cyano-6,12-dihydro-6-methylbenzimidazo[1,2-*b*][2,7]naphthyrid-12-one (**11**).

This compound was obtained from (1-methyl-2-benzimidazolyl)acetonitrile (**1d**) and the ester **5** as yellow crystals.

5-Cyano-12*H*-benzothiazolo[3,2-*b*][2,7]naphthyrid-12-one (**12**).

This compound was obtained from 2-benzothiazolylacetonitrile (**1e**) and the ester **5** as colorless crystals.

11-Cyano-5*H*-pyrido[1,2-*g*][1,6]naphthyrid-5-one [3] (**14**).

This compound was obtained from 2-pyridylacetonitrile (**1a**) and the ester **13** as yellow crystals.

7-Cyano-12*H*-quino[1,2-*g*][1,6]naphthyrid-12-one (**15**).

This compound was obtained from 2-quinolinylacetonitrile (**1b**) and the ester **13** as red crystals.

12-Cyano-5,11-dihydro-11-methylbenzimidazo[1,2-*g*][1,6]naphthyrid-5-one (**16**).

This compound was obtained from (1-methyl-2-benzimidazolyl)acetonitrile (**1d**) and the ester **13** as pale yellow crystals.

12-Cyano-5*H*-benzothiazolo[3,2-*g*][1,6]naphthyrid-5-one (**17**).

This compound was obtained from 2-benzothiazolylacetonitrile (**1e**) and the ester **13** as almost colorless crystals.

12-Cyano-5,11-dihydro-11-methylbenzimidazo[1,2-*b*][2,6]naphthyrid-5-one (**19**).

This compound was obtained from (1-methyl-2-benzimidazolyl)acetonitrile (**1d**) and the ester **18** as yellow crystals.

3,5-Dihydro-6,8-dimethyl-10-(1-methyl-2-benzimidazolyl)-[1,2,4]triazolo[2,3-*b*][2,7]naphthyrid-5-one (**21**).

A suspension of 302 mg (1 mmole) of compound **9** and 276 mg (2 mmoles) of potassium carbonate in 15 ml of dimethylformamide was stirred at reflux temperature for 16 hours. The solvent was removed under reduced pressure, water (20 ml) was added, and the reaction mixture was neutralized (*p*H 6-7) by addition of acetic acid. The precipitate was collected, washed with water, dried, and recrystallized from dimethylformamide to give 262 mg (76%) of yellow crystals, mp 320-321°; ^{1}H nmr: δ 13.00-12.20 (broad s, 1H, NH), 8.18 (s, 1H, 2-H), 7.68 (d, 1H, J = 8.1 Hz, benzimidazole 4-H), 7.58 (d, 1H, J = 7.8 Hz, benzimidazole 7-H), 7.32-7.20 (m, 2H, benzimidazole 5-H, 6-H), 7.04 (s, 1H, 9-H), 3.57 (s, 3H, NCH_3), 3.04 (s, 3H, 6-CH_3), 2.31 (s, 3H, 8-CH_3); ir: 2922, 1656, 1556, 1536, 1458, 1340, 1248, 1238, 1186, 970, 788, 738 cm^{-1}.

Anal. Calcd. for $C_{19}H_{16}N_6O$ x 0.2 H_2O: C, 65.58; H, 4.75; N, 24.15. Found: C, 65.36; H, 4.59; N, 24.05.

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[2] Compound **14** has been described previously as an intermediate in the synthesis of allomatridine [3].

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[6] The possible tautomerism of compound **21** was not investigated; the representation as the 3*H* tautomer was chosen arbitrarily.

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