Recyclization of chromenopyridine derivatives to pyridopyrazinediones

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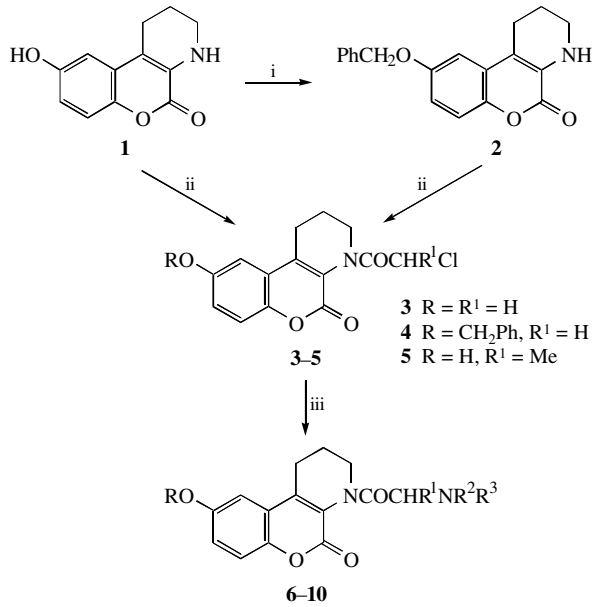
The reactions of N-ω-chloroacyl derivatives of chromeno[3,4-*b*]pyridine with primary arylalkylamines result in unusual recyclization to pyridopyrazinediones 11–16.

Recently,[1,2] we reported a new synthesis of chromeno[3,4-*b*]-pyridine derivatives *via* the condensation of quinones with α-oxolactam enamines. The presence of a cyclic secondary amino group in the chromenopyridines is responsible for the synthesis of various heterocyclic compounds of this kind with the use of reactions such as acylation.

The acylation of 9-hydroxy-1,2,3,4-tetrahydro-5*H*-chromeno-[3,4-*b*]pyridin-5-one **1**[1,2] and its O-benzyl derivative **2** with chloroacetyl and chloropropionyl chlorides gave the following N-ω-chloroacyl derivatives: 9-hydroxy-4-(2-chloroacetyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **3**, 9-benzyloxy-4-(2-chloroacetyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **4** and 9-hydroxy-4-(2-chloropropionyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **5** (Table 1). Nucleophilic substitution for the ω-chlorine atom of these compounds in reactions with secondary amines such as piperidine, morpholine and diethylamine resulted in the following ω-aminoacyl derivatives: 9-hydroxy-4-(2-piperidinoacetyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **6**, 9-hydroxy-4-(2-morpholinoacetyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **7**, 9-hydroxy-4-(2-diethylaminoacetyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **8**, 9-benzyloxy-4-(2-morpholinoacetyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **9** and 9-hydroxy-4-(2-piperidinopropionyl)-1,2,3,4-tetrahydro-5*H*-chromeno[3,4-*b*]pyridin-5-one **10** (Table 1).†

A different reaction path was observed in the interaction of compounds **3** and **4** with primary arylalkylamines $H_2N(CH_2)_xAr$ such as benzylamine, β-phenylethylamine, homoveratrylamine and tryptamine. In these cases, the reaction did not terminate at the step of nucleophilic substitution for the chlorine atom in chloroacyl residues. At the next step, intramolecular lactone ring opening took place, which was accompanied by recyclization to form piperazine-2,5-dione derivatives. The following compounds were synthesised: 2-benzyl-9-(2,5-dihydroxyphenyl)-7,8-dihydro-2*H*-pyrido[1,2-*a*]pyrazine-1,4(3*H*,6*H*)-dione **11**, 9-(2,5-dihydroxyphenyl)-2-phenethyl-7,8-dihydro-2*H*-pyrido-[1,2-*a*]pyrazine-1,4(3*H*,6*H*)-dione **12**, 9-(2,5-dihydroxyphenyl)-2-(3,4-dimethoxyphenethyl)-7,8-dihydro-2*H*-pyrido[1,2-*a*]pyrazine-1,4(3*H*,6*H*)-dione **13**, 9-(2,5-dihydroxyphenyl)-2-[2-(1*H*-indol-3-yl)ethyl]-7,8-dihydro-2*H*-pyrido[1,2-*a*]pyrazine-1,4(3*H*,6*H*)-dione **14**, 2-benzyl-9-(5-benzyloxy-2-hydroxyphenyl)-



6 R = R¹ = H, R² + R³ = $(CH_2)_5$
7 R = R¹ = H, R² + R³ = $(CH_2)_4O$
8 R = R¹ = H, R² = R³ = Et
9 R = CH_2Ph, R¹ = H, R² + R³ = $(CH_2)_4O$
10 R = H, R¹ = Me, R² + R³ = $(CH_2)_6$

Scheme 1 *Reagents and conditions*: i, anhydrous K_2CO_3 and benzyl chloride were added to a solution of compound **1** in DMF; the mixture was refluxed for 2 h and then filtered; the filtrate was evaporated, and the residue was ground with water; compound **2** was filtered off; ii, chloroacetyl chloride (or 2-chloropropionyl chloride) was added to a suspension of compound **1** (or **2**) in toluene; the mixture was refluxed with stirring for 1 h and then cooled; compound **3** (**4** or **5**) was filtered off; iii, 2 mol of an amine were added to a solution of 1 mol of compound **3** (**4** or **5**) in DMF; the mixture was stirred (20 °C, 10 h) and diluted with water; compound **6** (or **7**–**10**) was filtered off.

† The ^{1}H NMR spectra were recorded on a Bruker AC-200 spectrometer. The mass spectra were recorded on a Finnigan SSQ-710 mass spectrometer with direct sample injection into the ion source.

For **2**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.88 (q, 2H, 2-CH_2, *J* 6.4 Hz), 2.64 (t, 2H, 1-CH_2, *J* 6.4 Hz), 3.25 (br. s, 2H, 3-CH_2), 5.14 (s, 2H, 9-OCH_2), 5.92 (br. s, 1H, 4-NH), 6.89 (qd, 1H, 8-H, J_1 8.4 Hz, J_2 2.8 Hz), 6.98 (d, 1H, 10-H, *J* 2.8 Hz), 7.20 (d, 1H, 7-H, *J* 8.4 Hz), 7.32–7.49 (m, 5H, Ph). MS, *m/z*: 307 [M$^+$].

For **3**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 2.01 (q, 2H, 2-CH_2, *J* 6.4 Hz), 2.89 (t, 2H, 1-CH_2, *J* 6.4 Hz), 3.71 (br. s, 2H, 3-CH_2), 4.38 (s, 2H, $COCH_2Cl$), 7.03 (m, 2H, 8-H, 10-H), 7.26 (d, 1H, 7-H, *J* 9.4 Hz), 9.60 (s, 1H, 9-OH). MS, *m/z*: 293 [M$^+$].

For **4**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 2.02 (q, 2H, 2-CH_2, *J* 6.4 Hz), 2.95 (t, 2H, 1-CH_2, *J* 6.4 Hz), 3.72 (br. s, 2H, 3-CH_2), 4.39 (s, 2H, $COCH_2Cl$), 5.21 (s, 2H, 9-OCH_2), 7.26–7.51 (m, 8H, 7-H, 8-H, 10-H, Ph). MS, *m/z*: 383 [M$^+$].

For **5**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.57 (d, 3H, CH*Me*, *J* 6.4 Hz), 2.01 (q, 2H, 2-CH_2, *J* 6.4 Hz), 2.89 (m, 2H, 1-CH_2), 3.40 and 4.03 (2br. s, 1H, 3-CH_2), 4.89 (q, 1H, C*H*Me, *J* 6.4 Hz), 7.00–7.29 (m, 3H, 7-H, 8-H, 10-H), 9.66 (s, 1H, 9-OH). MS, *m/z*: 307 [M$^+$].

For **6**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.05 (br. s, 6H, 3′-CH_2, 4′-CH_2, 5′-CH_2), 1.93 (br. s, 2H, 2-CH_2), 2.12 (br. s, 4H, 2′-CH_2, 6′-CH_2) 2.80 (br. m, 4H, 1-CH_2, 3-CH_2), 3.40 and 4.42 (2br. s, 1H, $COCH_2$), 6.93 (m, 2H, 8-H, 10-H), 7.22 (d, 1H, 7-H, *J* 9.4 Hz), 9.51 (s, 1H, 9-OH). MS, *m/z*: 342 [M$^+$].

For **7**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.96 (q, 2H, 2-CH_2, *J* 6.4 Hz), 2.22 (t, 4H, 2′-CH_2, 6′-CH_2, *J* 6.4 Hz), 2.80 (br. m, 2H, 1-CH_2), 3.19 (br. s, 6H, 3-CH_2, 3′-CH_2, 3′-CH_2, 4′-CH_2), 3.40 and 4.40 (2br. s, 1H, $COCH_2$), 6.90–7.30 (m, 3H, 7-H, 8-H, 10-H), 9.55 (s, 1H, 9-OH). MS, *m/z*: 344 [M$^+$].

For **8**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 0.72 (t, 6H, CH_2*Me*, *J* 7.2 Hz), 1.96 (q, 2H, 2-CH_2, *J* 6.4 Hz), 2.29 (q, 4H, CH_2Me, *J* 7.2 Hz), 2.80 (br. s, 2H, 1-CH_2), 3.35 (br. s, 2H, 3-CH_2), 3.40 and 4.20 (2br. s, 1H, OCH_2), 6.90–7.30 (m, 3H, 7-H, 8-H, 10-H), 9.54 (s, 1H, 9-OH). MS, *m/z*: 330 [M$^+$].

For **9**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.96 (br. m, 2H, 2-CH_2), 2.18 (t, 4H, 2′-CH_2, 6′-CH_2, *J* 6.4 Hz), 2.85 (br. m, 2H, 1-CH_2), 3.06 (br. s, 6H, 3-CH_2, 3′-CH_2, 5′-CH_2), 3.45 and 4.41 (2br. s, 1H, $COCH_2$), 5.19 (s, 2H, 6-OCH_2), 7.10–7.60 (m, 8H, 7-H, 8-H, 10-H, Ph). MS, *m/z*: 434 [M$^+$].

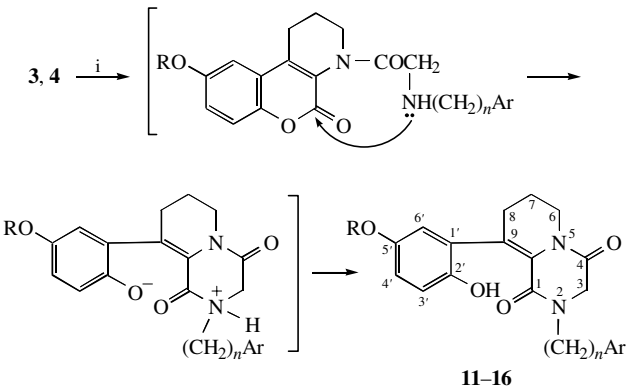
For **10**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.06 (d, 3H, CH*Me*, *J* 6.4 Hz), 1.22 (br. s, 8H, 3′-CH_2, 4′-CH_2, 5′-CH_2, 6′-CH_2), 1.97 (m, 2H, 2-CH_2), 2.41 (br. s, 4H, 2′-CH_2, 7′-CH_2), 2.60–3.20 (m, 3H, 1-CH_2, 3-H), 3.85 (q, 1H, C*H*Me, *J* 6.4 Hz), 4.42 (br. s, 1H, 3-H), 6.80–7.30 (m, 3H, 7-H, 8-H, 10-H), 9.56 (s, 1H, 9-OH). MS, *m/z*: 434 [M$^+$].

7,8-dihydro-2*H*-pyrido[1,2-*a*]pyrazine-1,4(3*H*,6*H*)-dione **15** and 9-(5-benzyloxy-2-hydroxyphenyl)-2-phenethyl-7,8-dihydro-2*H*-pyrido[1,2-*a*]pyrazine-1,4(3*H*,6*H*)-dione **16** (Table 1).‡

The structures of the prepared compounds were confirmed by NMR spectroscopy.‡ Two signals due to the protons of hydroxyl groups (δ 8.10–8.51 ppm) were the most characteristic signals in the ^{1}H NMR spectra of compounds **11**–**14**. The ^{1}H NMR spectrum of O-benzyl derivative **16** exhibited proton signals due to a hydroxyl group (δ 8.45 ppm) and an O-benzyl group (4.96 ppm, OCH_2; 7.10–7.50 ppm, Ph).

The structures of pyridopyrazines **11**–**16** were chemically supported by the readily occurring oxidation of compounds **11** and **13** to the corresponding quinolyl derivatives: 2-benzyl-9-(3,6-dioxocyclohexa-1,4-dienyl)-7,8-dihydro-2*H*-pyrido[1,2-*a*]-pyrazine-1,4(3*H*,6*H*)-dione **17** and 9-(3,6-dioxocyclohexa-1,4-dienyl)-2-(3,4-dimethoxyphenethyl)-7,8-dihydro-2*H*-pyrido-[1,2-*a*]pyrazine-1,4-(3*H*,6*H*)-dione **18**. Signals due to the protons of hydroxyl groups were absent from the ^{1}H NMR spectra of compounds **17** and **18** (Table 1).‡

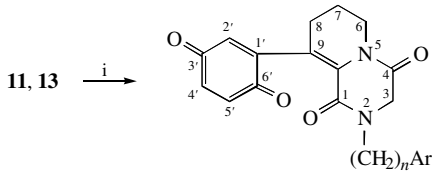
The signals of protons of the quinone moiety of derivative **17** were downfield shifted ($\Delta\delta$ = 0.3 ppm) with respect to the corresponding signals of the hydroquinone moiety of **11**. The ^{13}H NMR spectrum of compound **11** exhibited downfield signals at 145.6, 149.5 (2′-C, 5′-C) and 158.1, 162.2 (1-C, 4-C). The positions of 1-C and 4-C signals in the spectrum of compound **17** changed insignificantly (157.5 and 161.0), and downfield signals (184.6 and 187.1) appeared, which can be attributed to the 3′-C and 6′-C atoms. The signals of aromatic carbon atoms also exhibited a strong downfield shift (by 15–20 ppm) on going from compound **11** to **17**; this shift is consistent with a change from the hydroquinone to the quinone structure.



11–**16**

11 R = H, *n* = 1, Ar = Ph
12 R = H, *n* = 2, Ar = Ph
13 R = H, *n* = 2, Ar = 3,4-$(OMe)_2C_6H_3$
14 R = H, *n* = 2, Ar = 3-indolyl
15 R = CH_2Ph, *n* = 1, Ar = Ph
16 R = CH_2Ph, *n* = 2, Ar = Ph

Scheme 2 *Reagents and conditions*: i, 2 mol of an amine were added to a solution of 1 mol of compound **3** (or **4**) in DMF; the mixture was stirred (20 °C, 10 h) and diluted with water; compound **11** (or **12**–**16**) was filtered off.



Scheme 3 *Reagents and conditions*: i, an aqueous solution of potassium ferricyanide, potassium carbonate and potassium bicarbonate was added to a suspension of compound **11** (or **13**) in chloroform with stirring; the mixture was stirred (20 °C, 1 h); the organic layer was separated and evaporated; the residue was ground with diethyl ether; compound **17** (or **18**) was filtered off.

Note that laktone and chromone ring opening under the action of amine-containing compounds was described in the literature.[3–5] In this work, we found that the intramolecular nucleophilic attack of the NH group on the carbonyl carbon results in lactone ring cleavage and piperazine ring closure. The latter circumstance is of particular importance because 2,5-dioxopiperazines (cyclic dipeptides) belong to one of the most naturally widespread classes of peptide derivatives.[6] A new approach to the synthesis of these compounds is of interest in terms of their biological activity.

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‡ The mass spectra of compounds **11**–**16** do not contain the peaks of molecular ions; the peak of $[M - H_2O]^+$ is a characteristic peak in the mass spectra of these compounds.

For **11**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.82 (q, 2H, 7-CH_2, *J* 5.8 Hz), 2.35 (br. s, 2H, 8-CH_2), 3.72 (br. s, 2H, 6-CH_2), 3.96 (s, 2H, 3-CH_2), 4.46 (br. s, 2H, 2-CH_2), 6.37 (d, 1H, 6′-H, *J* 2.8 Hz), 6.43 (q, 1H, 4′-H), 6.55 (d, 1H, 3′-H, *J* 8.4 Hz), 7.19–7.37 (m, 5H, Ph), 8.21 and 8.51 (2br. s, 1H, 2′-OH, 5′-OH). ^{13}C NMR ([$^2H_6$]DMSO) δ: 20.7, 30.6, 39.5, 48.2, 49.1 (7-H, 8-H, 6-H, 3-H, 2-NCH_2), 113.9, 115.3, 115.8 (3′-C, 4′-C, 6′-C), 127.3, 127.8, 128.5, 128.9 (Ph), 124.9, 129.8, 136.4 (9-C, 1′-C, 9_a-C), 145.6, 149.5 (2′-C, 5′-C), 158.1, 162.2 (1-C, 4-C). MS, *m/z*: 346 $[M - H_2O]^+$.

For **12**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.81 (q, 2H, 7-CH_2, *J* 5.8 Hz), 2.35 (br. s, 2H, 8-CH_2), 2.73 and 3.42 (2t, 2H, 2-CH_2CH_2, *J* 7.2 Hz), 3.70 (br. s, 2H, 6-CH_2), 4.00 (s, 2H, 3-CH_2), 6.34 (d, 1H, 6′-H, *J* 2.8 Hz), 6.45 (q, 1H, 4′-H), 6.55 (d, 1H, 3′-H, *J* 8.4 Hz), 7.10–7.40 (m, 5H, Ph), 8.10 and 8.42 (2br. s, 1H, 2′-OH, 5′-OH). MS, *m/z*: 360 $[M - H_2O]^+$.

For **13**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.84 (q, 2H, 7-CH_2, *J* 5.8 Hz), 2.36 (br. s, 2H, 8-CH_2), 2.70 and 3.43 (2t, 2H, 2-CH_2CH_2, *J* 7.2 Hz), 3.74 and 3.75 (s, 3H, 3″-OMe, 4″-OMe), 4.04 (s, 2H, 3-CH_2), 6.38 (d, 1H, 6′-H, *J* 2.8 Hz), 6.48 (q, 1H, 4′-H), 6.59 (d, 1H, 3′-H, *J* 8.4 Hz), 6.70–7.00 (m, 3H, Ph), 8.17 and 8.49 (2br. s, 1H, 2′-OH, 5′-OH). MS, *m/z*: 420 $[M - H_2O]^+$.

For **14**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.82 (q, 2H, 7-CH_2, *J* 5.8 Hz), 2.33 (br. s, 2H, 8-CH_2), 2.86 and 3.49 (2t, 2H, 2-CH_2CH_2, *J* 7.2 Hz), 4.03 (s, 2H, 3-CH_2), 6.37 (d, 1H, 6′-H, *J* 2.8 Hz), 6.44 (q, 1H, 4′-H), 6.56 (d, 1H, 3′-H, *J* 8.4 Hz), 6.90–7.15 (m, 3H, 2″-H, 5″-H, 6″-H), 7.33 and 7.54 (2d, 1H, 4″-H, 7″-H, *J* 7.4 Hz), 8.10 and 8.42 (2br. s, 1H, 2′-OH, 5′-OH), 10.70 (br. s, 1H, 1″-NH). MS, *m/z*: 399 $[M - H_2O]^+$.

For **15**: ^{1}H NMR ($CDCl_3$) δ: 1.95 (m, 2H, 7-CH_2), 2.47 (t, 2H, 8-CH_2, *J* 6.2 Hz), 3.40 and 4.35 (2m, 1H, 6-CH_2), 3.95 (AB system, 2H, 3-CH_2), 4.58 (AB system, 2H, 2-CH_2), 5.01 (s, 2H, 5-OCH_2), 6.69 (d, 1H, 6′-H, *J* 2.8 Hz), 6.84 (q, 1H, 4′-H), 6.96 (d, 1H, 3′-H, *J* 8.4 Hz), 7.10–7.60 (m, 10H, 2Ph). MS, *m/z*: 346 $[M - H_2O]^+$.

For **16**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.83 (q, 2H, 7-CH_2, *J* 5.8 Hz), 2.45 (br. s, 2H, 8-CH_2), 2.73 and 3.43 (2t, 2H, 2-CH_2CH_2, *J* 7.2 Hz), 3.71 (br. s, 2H, 6-CH_2), 4.02 (s, 2H, 3-CH_2), 4.96 (s, 2H, 5′-OCH_2), 6.59–6.80 (m, 3H, 3′-H, 4′-H, 6′-H), 7.10–7.50 (m, 10H, 2Ph), 8.45 (br. s, 1H, 2′-OH). MS, *m/z*: 450 $[M - H_2O]^+$.

For **17**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.89 (m, 2H, 7-CH_2), 2.37 (m, 2H, 8-CH_2), 3.77 (m, 2H, 6-CH_2), 3.94 (s, 2H, 3-CH_2), 4.47 (AB system, 2H, CH_2Ph), 6.43 (d, 1H, 2′-H, *J* 2.2 Hz), 6.72 (q, 1H, 4′-H), 6.83 (d, 1H, 5′-H, *J* 7.0 Hz), 7.10–7.40 (m, 5H, Ph). ^{13}C NMR ($CDCl_3$), δ: 20.2, 30.4, 40.0, 48.8, 49.5 (7-C, 8-C, 6-C, 3-C, 2-NCH_2), 128.0, 128.2, 128.3, 129.0, 136.7, 137.3 (2′-C, 4′-C, 5′-C, Ph), 121.8, 134.4, 151.3 (9-C, 9_a-C, 1′-H), 157.5, 161.4 (1-C, 4-C), 184.6, 187.1 (3′-C, 6′-C). MS, *m/z*: 362 $[M^{\ddagger}]$.

For **18**: ^{1}H NMR ([$^2H_6$]DMSO) δ: 1.83 (br. s, 2H, 7-CH_2), 2.38 (br. s, 2H, 8-CH_2), 2.65 and 3.55 (2br. s, 2H, 2-CH_2CH_2), 3.71 (br. s, 8H, 6-CH_2, 2OMe), 3.95 (s, 2H, 3-CH_2), 6.40–7.00 (m, 6H, 2′-H, 4′-H, 5′-H, C_6H_3). MS, *m/z*: 436 $[M^{\ddagger}]$.

Table 1 Characterization of compounds **2**–**18**.

Compound	Empirical formula	Elemental analysis				Mp/°C	Yield (%)
		C (%) calc. (found)	H (%) calc. (found)	N (%) calc. (found)	Cl (%) calc. (found)		
2	$C_{19}H_{17}NO_3$	74.25 (74.60)	5.56 (5.50)	4.56 (5.08)		127–130 (EtOH)	98
3	$C_{14}H_{12}ClNO_4$	57.25 (57.36)	4.12 (4.12)	4.77 (4.98)	12.07 (11.75)	205–208 (EtOH)	98
4	$C_{21}H_{18}ClNO_4$	65.71 (65.73)	4.73 (5.04)	3.65 (3.61)	9.24 (8.96)	159–162 (EtOH)	91
5	$C_{15}H_{14}ClNO_4$	58.54 (58.82)	4.58 (4.52)	4.55 (4.61)	11.52 (11.00)	202–205 (EtOH)	92
6	$C_{19}H_{22}N_2O_4$	66.64 (66.61)	6.47 (6.50)	8.18 (8.23)		241–244 (acetone)	99
7	$C_{18}H_{20}N_2O_5$	62.78 (62.21)	5.85 (5.74)	8.13 (8.46)		249–251 (EtOH)	87
8	$C_{18}H_{22}N_2O_4$	65.43 (64.95)	6.71 (6.68)	8.48 (8.29)		186–188 (acetone)	91
9	$C_{25}H_{26}N_2O_5$	69.11 (69.39)	6.03 (6.00)	6.45 (6.43)		137–139	87
10	$C_{21}H_{26}N_2O_4$	68.08 (68.40)	7.07 (7.08)	7.56 (8.00)		210–212	90
11	$C_{21}H_{20}N_2O_4$	69.21 (69.82)	5.53 (5.53)	7.69 (7.15)		129–131 (acetonitrile)	83
12	$C_{22}H_{22}N_2O_4$	69.82 (70.06)	5.86 (5.90)	7.40 (7.21)		160–163 (acetonitrile)	89
13	$C_{24}H_{26}N_2O_6$	65.73 (65.58)	5.98 (6.08)	6.39 (6.35)		169–171 (acetonitrile)	93
14	$C_{24}H_{23}N_3O_4$	69.05 (69.53)	5.55 (5.67)	10.07 (9.63)		217–220 (acetone)	65
15	$C_{28}H_{26}N_2O_4$	73.99 (74.57)	5.77 (5.51)	6.16 (6.23)		148–150 (EtOH)	99
16	$C_{29}H_{28}N_2O_4$	73.67 (73.70)	6.18 (6.02)	6.13 (6.20)		134–136 (EtOH)	92
17	$C_{20}H_{18}N_2O_4$	69.60 (69.60)	5.00 (4.94)	7.73 (7.69)		158–161	92
18	$C_{24}H_{24}N_2O_6$	66.04 (65.91)	5.54 (5.49)	6.42 (6.40)		149–152	87