Efficient Iminophosphorane-Mediated Preparation of Benzofuro-[3,2-*d*]pyrimidin-4(3*H*)-ones and Unexpected Ring Opening Products

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The carbodiimides **4**, obtained from aza-*Wittig* reactions of iminophosphorane **3** with aromatic isocyanates, reacted with secondary amines, phenols or alcohols in the presence of catalytic amounts of K_2CO_3 or sodium alkoxide to give 2-substituted benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **6**. However, when 2,2′-iminobis[ethanol] was used, the unexpected ring opening product **7** was formed instead of **6**. Reaction of **4** with primary amines RNH_2 (R = Et, Pr, Bu, *etc.*) gave guanidine intermediates **8**, which were further treated with EtONa to give only one regioisomer **9** *via* a base catalyzed cyclization. However, another regioisomer **11** was obtained when NH_3 or 'small' amines RNH_2 (R = Me, NH_2) were used in the absence of EtONa *via* a spontaneous cyclization of **8**.

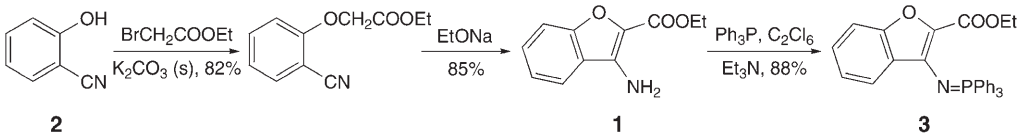
Introduction. – The derivatives of fused pyrimidinones have been the focus of great interest over many years. This is probably due to the fact that many compounds containing a fused pyrimidinone ring play a very important part in the biochemistry of the living cell [1]. The recent development of physiologically highly potent purine analogues with interesting antiviral, antiallergic, and especially with anticancer activities have promoted a great current interest in facile and general routes to these molecules in synthetically useful yields. Although some derivatives of benzofuropyrimidines have shown good analgesic, anti-inflammatory, antimicrobial, anticoccidial, and blood sugar-lowering activities [2–5], there are only few reports on the synthesis of benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones, which are of considerable interest as potential biologically active compounds or pharmaceuticals. The methods described for the preparation of some representative derivatives of this ring system involve the reaction of 3-amino-2-(ethoxycarbonyl)benzofuran with orthoformate and an amine, the rearrangement of benzofuro[3,2-*d*]oxazines by treatment with an amine, or the cyclization of 3-amino-2-(aminocarbonyl)benzofuran with acyl chlorides [6–9]. However, 2-amino or 2-aryl(alkyl)oxy substituted benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones were not easily accessible by currently existing routes.

The aza-*Wittig* reactions of iminophosphoranes have received increased attention in view of their utility in the synthesis of *N*-heterocyclic compounds [10–16]. Annelation of ring systems with *N*-heterocycles by means of an aza-*Wittig* reaction has been widely utilized because of the availability of functionalized iminophosphoranes. Recently, we have become interested in the synthesis of quinazolinones, thienopyrimidinones, and imidazolinones *via* the aza-*Wittig* reaction of α- or β-(ethoxycarbonyl)iminophosphor-

anes with aromatic isocyanates and subsequent reaction with various nucleophiles under mild conditions [17–21]. Here, we wish to report an efficient approach to benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones and unexpected ring opening products.

Results and Discussion. – Ethyl 3-amino-1-benzofuran-2-carboxylate (**1**), easily obtained by cyclization of 2-hydroxybenzonitrile (**2**), (with ethyl bromoacetate, under basic conditions [22][23]), was converted to the iminophosphorane **3** *via* the reaction with Ph_3P, hexachloroethane and Et_3N (*Scheme 1*).

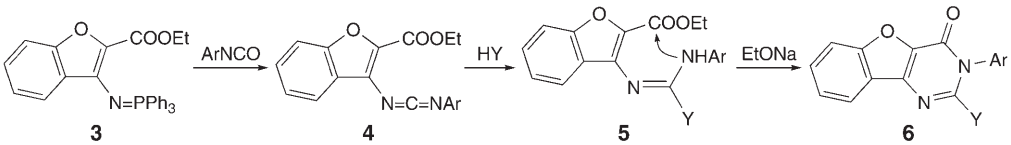
Scheme 1



2 **1** **3**

The iminophosphorane **3** reacted with aromatic isocyanates to give carbodiimides **4**, which were allowed to react with secondary amines to provide guanidine intermediates **5** ($Y = R_2N$). In the presence of catalytic amounts of EtONa, the latter were easily converted to 2-(dialkylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **6** ($Y = R_2N$) in satisfactory yields at room temperature (*Scheme 2*). It is noteworthy that the yield of isolated **6** was good even when Y was the bulky diisopropylamino group. The reaction of carbodiimides **4** with phenols produced 2-(aryloxy)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **6** (Y = ArO) in the presence of a catalytic amount of K_2CO_3 in good yields. The cyclization can be completed smoothly under mild conditions, independent of the substituents on the phenol. They can either be a halogen atom (*Table 1*, *Entries 13* and *14*) or typical electron-releasing groups (*Entries 11*, *12*, and *15*). The direct reaction of carbodiimide **4** with ROH gave 2-(alkoxy)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **6** (Y = RO) in satisfactory yields in the presence of catalytic amounts of RONa. The results are listed in *Table 1*.

Scheme 2



3 **4** **5** **6**

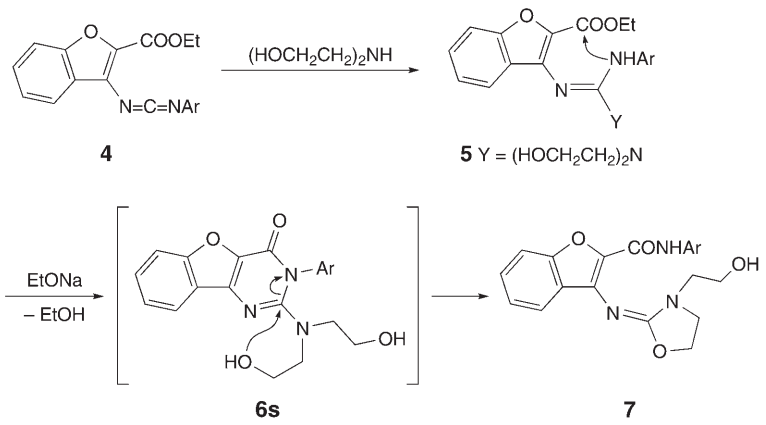
Unexpectedly, when 2,2′-iminobisethanol was used to react with carbodiimide **4**, the rearranged product **7** was obtained in the presence of catalytic amounts of EtONa (*Scheme 3*). The structure of **7** was confirmed by spectroscopic data. The formation of **7** can be explained by an initial cyclization of a guanidine intermediate of type **5** to give **6s**, which undergoes further ring cleavage to yield **7**. Furthermore, a single crystal of **7a**

Table 1. *Synthesis of Compounds* **6** *and* **7**

Entry	Compound	Ar	Y	Yield [%][a])
1	**6a**	Ph	Et_2N	86
2	**6b**	Ph	Pr_2N	82
3	**6c**	Ph	piperidin-1-yl	87
4	**6d**	Ph	$(n\text{-}C_5H_{11})_2N$	76
5	**6e**	Ph	$(i\text{-}Bu)_2N$	85
6	**6f**	Ph	(Ph)MeN	82
7	**6g**	4-Cl–C_6H_4	$(i\text{-}Pr)_2N$	75
8	**6h**	4-Cl–C_6H_4	Pr_2N	73
9	**6i**	4-Me–C_6H_4	Et_2N	83
10	**6j**	3-Me–C_6H_4	morpholin-1-yl	83
11	**6k**	Ph	3-Me–C_6H_4O	87
12	**6l**	Ph	4-MeO–C_6H_4O	90
13	**6m**	Ph	4-Cl–C_6H_4O	81
14	**6n**	4-Me–C_6H_4	4-Cl–C_6H_4O	89
15	**6o**	3-Me–C_6H_4	4-MeS–C_6H_4O	88
16	**6p**	Ph	EtO	82
17	**6q**	4-Cl–C_6H_4	MeO	78
18	**6r**	4-Me–C_6H_4	EtO	84
19	**7a**	Ph	$(HOCH_2CH_2)_2N$	88
20	**7b**	4-Me–C_6H_4	$(HOCH_2CH_2)_2N$	82

[a]) Yields of isolated product based on iminophosphorane **3**.

Scheme 3



was obtained from a CH_2Cl_2 soln., and an X-ray structure analysis verified the proposed structure (*Fig.*).

It is interesting that the reaction of carbodiimide **4** with aliphatic primary amines generates different products. The reaction of carbodiimide **4** with RNH_2 (R = Et, Pr, Bu, *etc.*) gave guanidine intermediates **8**, which in the presence of EtONa cyclized to provide 2-(alkylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **9** exclusively, *i.e.*, one

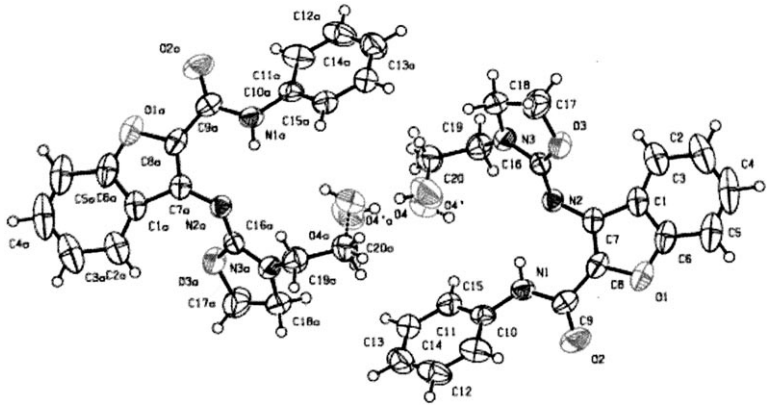


Figure. *ORTEP Diagram of the crystal structure of compound* **7a** (50% thermal ellipsoids). Compound **7a** has two unique molecules in the asymmetric unit of the monoclinic unit cell. One of the molecules is numbered with atoms C1 – C20, O1, O2, O3, O4, O4′, N1, N2, and N3, while another molecule has atoms C1a – C20a, O1a, O2a, O3a, O4a, O4a′, N1a, N2a, and N3a.

of the possible regioisomers (*Scheme 4*). The structure of **9** was deduced from their ^{1}H-NMR data. For example, the ^{1}H-NMR spectrum of **9c** (R = Bu) showed the signals of the alkylamino-NH group at 4.10 ppm and NCH_2 at 3.44 – 3.49 ppm as *multiplet*s, which strongly suggests the existence of an $NHCH_2CH_2CH_2Me$ group in **9c** [24]. The results are listed in *Table 2*. The exclusive formation of **9** can be rationalized in terms of a base catalytic cyclization of the guanidine intermediate **8** to give **9** across the arylamino group rather than the alkylamino one. This is probably due to the preferential generation of a N – Ar cycle than of the more acidic NHAr cycle under the catalysis of EtONa.

It is deduced from *Scheme 5* that if a primary amine such as 2-hydroxyethylamine is used to react with carbodiimide **4**, the ring-opening product **10** might be formed. However, the normal product **9j** was obtained, when 2-aminoethanol was utilized in the

Scheme 4

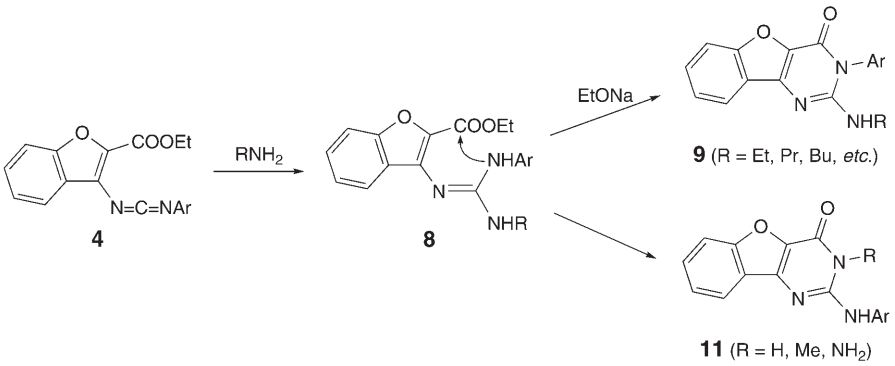
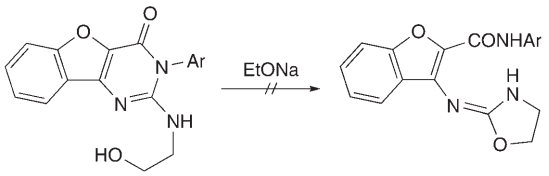


Table 2. *Synthesis of Compounds* **9** *or* **11**

Entry	Compound	Ar	R	Yield [%][a])
1	**9a**	Ph	Et	89
2	**9b**	Ph	*n*-Pr	83
3	**9c**	Ph	*n*-Bu	74
4	**9d**	Ph	i-Pr	82
5	**9e**	Ph	i-Bu	71
6	**9f**	4-Me–C_6H_4	i-Pr	87
7	**9g**	4-Me–C_6H_4	cyclohexyl	72
8	**9h**	4-Me–C_6H_4	*t*-Bu	75
9	**9i**	Ph	CH_2CH_2OH	76
10	**9j**	4-Me–C_6H_5	CH_2CH_2OH	84
11	**11a**	Ph	H	90
12	**11b**	Ph	Me	84
13	**11c**	Ph	NH_2	92
14	**11d**	4-Me–C_6H_4	NH_2	95

[a]) Isolated yields based on iminophosphorane **3**.

Scheme 5



9i,**j** **10**

presence of a catalytic amount of EtONa. The results show that the above described rearrangement of **6** to **7** might be due to steric effects in **6** which facilitate the ring opening.

On the other hand, the reaction of carbodiimides **4** with small amines, *i.e.*, NH_3, $MeNH_2$, or hydrazine, gave 2-(arylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **11**, another possible regioisomer, as the only product in the absence of EtONa (*Scheme 4*). It is deduced that the intermediate **8** is generated in a first step, which then cyclizes quickly, and catalysis by EtONa is therefore not needed. The formation of **11** can be explained in terms of a spontaneous cyclization of the guanidine intermediate **8** to give **11** directly for the slight sterically hindered and/or strong nucleophilic ammonia, methylamino or hydrazine group, but not for the arylamines.

Conclusions. – We have developed an efficient synthesis of 2-substituted benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones and previously not reported ring opening products *via* aza-*Wittig* reactions. Due to the mild reaction conditions, good yields, easily accessible starting material, and straightforward product isolation, the versatile synthetic approach discussed here compares favorably with other existing methods.

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Experimental Part

General. M.p. uncorrected. IR Spectra: *PE-983* spectrometer as KBr pellets, in cm^{-1}. NMR Spectra: in $CDCl_3$ or (D_6)DMSO on a *Varian Mercury 400* spectrometer, in ppm (δ) relative to TMS. MS: *Finnigan Trace MS* spectrometer. Elemental analyses: *Vario EL III* elemental analyzer. The diffraction data were collected on a *Bruker SMART AXS CCD* diffractometer, MoK_α, $2\theta = 1.86 - 27.50°$.

Ethyl 3-[(Triphenylphosphoranylidene)amino]-2-benzofurancarboxylate (**3**). To a mixture of *ethyl 3-amino-1-benzofuran-2-carboxylate* [22][23] (**1**; 1.64 g, 8 mmol), PPh_3 (3.14 g, 12 mmol) and C_2Cl_6 (2.84 g, 12 mmol) in dry MeCN (40 ml), was added dropwise Et_3N (2.42 g, 24 mmol) at r.t. The color of the mixture quickly turned yellow. After stirring for 4 h, the solvent was removed under reduced pressure and the residue was recrystallized from EtOH to give **3**. White solid. Yield 3.27 g, 88%. M.p.: 172 – 173°. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.23 (*t*, *J* = 7.2, Me); 4.18 (*q*, *J* = 7.2, CH_2); 6.93 – 7.80 (*m*, 19 arom. H). IR (KBr): 1755 (C=O), 1599, 1212. MS: 465 (99, M^+), 436 (31), 392 (100), 262 (37), 183 (80). Anal. calc. for $C_{29}H_{24}NO_3P$ (465.48): C 74.83, H 5.20, N 3.01; found: C 74.96, H 5.25, N 2.79.

*General Procedure for the Preparation of 2-(Dialkylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones* **6a** – **6j**. To a soln. of **3** (0.93 g, 2 mmol) in dry CH_2Cl_2 (15 ml) was added arom. isocyanate (2 mmol) under N_2 at r.t. After the mixture was kept for 8 – 12 h at 0 – 5°, the solvent was removed under reduced pressure, and Et_2O/petroleum ether (PE) (1:2, 20 ml) was added to precipitate Ph_3PO. After filtration, the solvent was removed to give carbodiimide **4**, which was used directly without further purification. To the soln. of **4** in CH_2Cl_2 (15 ml) was added dialkylamine (2 mmol), and allowed to stand for 0.5 – 6 h. Then, the soln. was concentrated, and anh. EtOH (10 ml) with several drops of EtONa in EtOH was added. The mixture was stirred for 1 – 6 h at r.t. The soln. was concentrated under reduced pressure, and the residual was recrystallized to give 2-(dialkylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **6a** – **6j**.

*2-(Diethylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6a**). White crystals, recrystallized from EtOH. Yield 0.57 g, 86%. M.p. 170 – 171°. IR (KBr): 1695 (C=O), 1529, 1346, 1074. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.85 (*t*, *J* = 7.2, 2 Me); 3.13 (*q*, *J* = 7.2, 2 CH_2N); 7.34 – 8.04 (*m*, 9 arom. H). MS: 333 (7, M^+), 304 (38), 261 (35), 214 (18), 130 (65), 102 (100). Anal. calc. for $C_{20}H_{19}N_3O_2$ (333.38): C 72.05, H 5.74, N 12.60; found: C 72.27, H 5.87, N 12.54.

*2-(Dipropylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6b**). White crystals, recrystallized from EtOH. Yield 0.59 g, 82%. M.p. 130 – 131°. IR (KBr): 1699 (C=O), 1526, 1351, 1216. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.75 (*t*, *J* = 7.2, 2 Me); 1.23 – 1.32 (*m*, 2 CH_2); 3.02 (*t*, *J* = 7.2, 2 CH_2N); 7.33 – 8.04 (*m*, 9 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 11.1 (2 C); 20.4 (2 C); 53.0 (2 C); 112.5; 121.3; 122.8; 123.1; 128.1; 128.7 (2 C); 128.8 (2 C); 129.1; 134.7; 137.4; 142.9; 154.6; 156.5; 157.2. MS: 361 (3, M^+), 318 (21), 261 (37), 214 (47), 130 (96), 102 (100). Anal. calc. for $C_{22}H_{23}N_3O_2$ (361.44): C 73.11, H 6.41, N 11.63; found: C 73.04, H 6.32, N 11.87.

*3-Phenyl-2-(piperidin-1-yl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6c**). White crystals, recrystallized from EtOH/CH_2Cl_2 (*v*/*v* = 1:1). Yield 0.60 g, 87%. M.p. 197 – 198°. IR (KBr): 1701 (C=O), 1528, 1246, 1093. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.22 – 1.45 (*m*, $(CH_2)_3$); 3.13 (*t*, *J* = 5.2, 2 CH_2N); 7.37 – 8.03 (*m*, 9 arom. H). MS: 345 (100, M^+), 316 (40), 261 (40), 160 (82), 130 (40), 102 (96). Anal. calc. for $C_{21}H_{19}N_3O_2$ (345.39): C 73.03, H 5.54, N 12.17; found: C 73.04, H 5.67, N 12.03.

*2-(Dipentylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6d**). White crystals, recrystallized from EtOH. Yield 0.63 g, 76%. M.p. 105 – 106°. IR (KBr): 1692 (C=O), 1628, 1417, 1115. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.86 (*t*, *J* = 7.2, 2 Me); 1.08 – 1.27 (*m*, 3 CH_2); 3.04 (*t*, *J* = 7.6, 2 CH_2N); 7.32 – 8.04 (*m*, 9 arom. H). MS: 417 (91, M^+), 360 (89), 346 (100), 261 (87), 159 (21), 130 (69), 102 (88). Anal. calc. for $C_{26}H_{31}N_3O_2$ (417.54): C 74.79, H 7.48, N 10.06; found: C 74.87, H 7.51, N 9.92.

*2-(Diisobutylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6e**). White crystals, recrystallized from EtOH. Yield 0.66 g, 85%. M.p. 141 – 142°. IR (KBr): 1701 (C=O), 1531, 1194. ^{1}H-NMR

(400 MHz, $CDCl_3$): 0.80 (*d*, *J* = 6.8, 4 Me); 1.81–1.88 (*m*, 2 H–C); 2.90 (*d*, *J* = 7.2, 2 CH_2N); 7.35–8.04 (*m*, 9 H). MS: 389 (35, M^+), 360 (27), 261 (38), 130 (100), 102 (79). Anal. calc. for $C_{24}H_{27}N_3O_2$ (389.49): C 74.01, H 6.99, N 10.79; found: C 74.25, H 7.04, N 10.73.

*2-[Methyl(phenyl)amino]-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6f**). White crystals, recrystallized from EtOH. Yield 0.60 g, 82%. M.p. 191–193°. IR (KBr): 1706 (C=O), 1528, 1352, 1085. ^{1}H-NMR (400 MHz, $CDCl_3$): 3.36 (*s*, MeN); 6.57–8.11 (*m*, 14 arom. H). MS: 367 (76, M^+), 290 (20), 261 (44), 235 (52), 130 (62). 77 (100). Anal. calc. for $C_{23}H_{17}N_3O_2$ (367.40): C 75.19, H 4.66, N 11.44; found: C 75.22, H 4.52, N 11.68.

*3-(4-Chlorophenyl)-2-(diisopropylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6g**). White crystals, recrystallized from EtOH. Yield 0.59 g, 75%. M.p. 167–169°. IR (KBr): 1701 (C=O), 1541, 1399, 1102. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.14 (*d*, *J* = 6.8, 4 Me); 3.49–3.56 (*m*, 2 CHN); 7.25–8.00 (*m*, 8 arom. H). MS: 395 (19, M^+), 352 (77), 295 (62), 185 (45), 130 (100). Anal. calc. for $C_{22}H_{22}ClN_3O_2$ (395.88): C 66.75, H 5.60, N 10.61; found: C 66.60, H 5.61, N 10.78.

*3-(4-Chlorophenyl)-2-(dipropylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6h**). White crystals, recrystallized from EtOH. Yield 0.58 g, 73%. M.p. 133–134°. IR (KBr): 1703 (C=O), 1539, 1490, 1093. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.77 (*t*, *J* = 7.2, 2 Me); 1.29–1.35 (*m*, 2 CH_2); 3.02 (*t*, *J* = 7.2, 2 CH_2N); 7.28–8.04 (*m*, 8 arom. H). MS: 395 (34, M^+), 352 (67), 295 (68), 185 (45), 130 (100). Anal. calc. for $C_{22}H_{22}ClN_3O_2$ (395.88): C 66.75, H 5.60, N 10.61; found: C 66.59, H 5.74, N 10.44.

*2-(Diethylamino)-3-(4-methylphenyl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6i**). White crystals, recrystallized from EtOH. Yield 0.58 g, 83%. M.p. 177–178°. IR (KBr): 1702 (C=O), 1542, 1386, 1099. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.86 (*t*, *J* = 7.2, 2 Me); 2.42 (*s*, Me); 3.13 (*q*, *J* = 7.2, 2 CH_2N); 7.21–8.05 (*m*, 8 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 12.3 (2 C); 21.0; 45.2 (2 C); 112.6; 121.3; 122.9; 123.2; 128.4 (2 C); 129.1; 129.6 (2 C); 134.8; 134.9; 138.0; 142.8; 154.8; 156.5; 157.2. MS: 347 (75, M^+), 318 (100), 275 (62), 214 (23), 130 (44), 102 (41). Anal. calc. for $C_{21}H_{21}N_3O_2$ (347.41): C 72.60, H 6.09, N 12.10; found: C 72.76, H 6.31, N 12.07.

*3-(3-Methylphenyl)-2-(morpholin-4-yl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6j**). White crystals, recrystallized from EtOH/CH_2Cl_2 (*v*/*v* = 1:1). Yield 0.60 g, 83%. M.p. 177–178°. IR (KBr): 1701 (C=O), 1528, 1400, 1118. ^{1}H-NMR (400 MHz, $CDCl_3$): 2.43 (*s*, Me); 3.16 (*t*, *J* = 4.8, 2 CH_2N); 3.44 (*t*, *J* = 4.8, 2 CH_2O); 7.19–8.02 (*m*, 8 arom. H). MS: 361 (69, M^+), 316 (78), 304 (68), 214 (45), 130 (100). Anal. calc. for $C_{21}H_{19}N_3O_3$ (361.42): C 69.79, H 5.30, N 11.63; found: C 69.84, H 5.53, N 11.47.

*General Procedure for the Preparation of 2-(Aryloxy)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones* **6k**–**6o**. To the soln. of carbodiimide **4** (ca. 2 mmol) prepared above in MeCN (15 ml) was added K_2CO_3 (0.2 mmol) and ArOH (2 mmol) in anh. MeCN (10 ml). The mixture was stirred for 6–8 h at 50–60°. The soln. was concentrated, and the residue was recrystallized to give **6k**–**6o**.

*2-(3-Methylphenoxy)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6k**). White crystals, recrystallized from EtOH. Yield 0.64 g, 87%. M.p. 230–232°. IR (KBr): 1712 (C=O), 1547, 1358, 1198. ^{1}H-NMR (400 MHz, $CDCl_3$): 2.37 (*s*, Me); 6.97–7.90 (*m*, 13 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 21.2; 112.7; 118.2; 120.9; 121.6; 122.4; 123.5; 126.6; 128.0; 129.0; 129.1; 129.4; 129.8; 134.6; 135.5; 135.6; 139.7; 141.9; 149.6; 151.8; 153.4; 153.6; 157.2. MS: 368 (8, M^+), 302 (63), 130 (23), 102 (23), 91 (27), 76 (100). Anal. calc. for $C_{23}H_{16}N_2O_3$ (368.38): C 74.99, H 4.38, N 7.60; found: C 75.23, H 4.50, N 7.52.

*2-(4-Methoxyphenoxy)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6l**). White crystals, recrystallized from EtOH. Yield 0.69 g, 90%. M.p. 231–232°. IR (KBr): 1711 (C=O), 1503, 1358, 1196. ^{1}H-NMR (400 MHz, $CDCl_3$): 3.84 (*s*, MeO); 6.91–7.90 (*m*, 13 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 55.4; 112.7; 114.2 (2 C); 121.6; 122.1 (2 C); 122.4; 123.5; 128.0 (2 C); 128.2; 129.1; 129.4 (2 C); 134.6; 135.5; 141.9; 145.3; 153.5; 153.7; 157.2 (2 C). MS: 384 (4, M^+), 265 (51), 222 (46), 130 (100), 102 (93). Anal. calc. for $C_{23}H_{16}N_2O_4$ (384.38): C 71.87, H 4.20, N 7.29; found: C 72.01, H 4.33, N 7.22.

*2-(4-Chlorophenoxy)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6m**). White crystals, recrystallized from EtOH. Yield 0.63 g, 81%. M.p. 224–226°. IR (KBr): 1717 (C=O), 1543, 1359, 1202. ^{1}H-NMR (400 MHz, $CDCl_3$): 7.12–7.88 (*m*, 13 arom. H). MS: 388 (16, M^+), 269 (38), 260 (34), 234 (27), 130 (62), 102 (100). Anal. calc. for $C_{22}H_{13}ClN_2O_3$ (388.80): C 67.96, H 3.37, N 7.21; found: C 68.01, H 3.43, N 7.16.

*2-(4-Chlorophenoxy)-3-(4-methylphenyl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6n**). White crystals, recrystallized from EtOH. Yield 0.72 g, 89%. M.p. 220–221°. IR (KBr): 1710 (C=O), 1541, 1348,

1199. ^{1}H-NMR (400 MHz, $CDCl_3$): 2.44 (*s*, Me); 7.12–7.87 (*m*, 12 arom. H). MS: 402 (14, M^+), 269 (43), 260 (27), 130 (63), 102 (100). Anal. calc. for $C_{23}H_{15}ClN_2O_3$ (402.83): C 68.58, H 3.75, N 6.95; found: C 68.63, H 3.82, N 6.78.

*3-(3-Methylphenyl)-2-[4-(methylsulfanyl)phenoxy]benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6o**). White crystals, recrystallized from EtOH. Yield 0.73 g, 88%. M.p. 222–224°. IR (KBr): 1709 (C=O), 1545, 1359, 1202. ^{1}H-NMR (400 MHz, $CDCl_3$): 2.44 (*s*, Me); 2.50 (*s*, Me); 7.10–7.88 (*m*, 12 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 16.3; 21.4; 112.8; 121.6; 121.8 (2 C); 122.3; 123.5; 124.9; 127.5 (2 C); 128.4; 129.3; 129.5; 130.1; 134.4; 135.7; 135.8; 139.5; 141.7; 149.5; 153.4; 153.6; 157.2. MS: 414 (12, M^+), 265 (47), 234 (29), 130 (52), 102 (100). Anal. calc. for $C_{24}H_{18}N_2O_3S$ (414.48): C 69.55, H 4.38, N 6.76; found: C 69.24, H 4.56, N 6.70.

*General Procedure for the Preparation of 2-(Alkoxy)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones* **6p**–**6r**. To the soln. of **4** (*ca.* 2 mmol) prepared above in anh. ROH (8 ml) was added RONa (0.2 mmol, 10% equiv) in ROH. The mixture was stirred for 4–6 h at r.t. The soln. was condensed, and the residue was recrystallized to give **6p**–**6r**.

*2-Ethoxy-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6p**). White crystals, recrystallized from MeOH. Yield 0.50 g, 82%. M.p. 148–150°. IR (KBr): 1700 (C=O), 1541, 1342, 1198. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.26 (*t*, *J* = 7.2, Me); 4.49 (*q*, *J* = 7.2, CH_2O); 7.28–8.02 (*m*, 9 arom. H). MS: 306 (100, M^+), 234 (23), 186 (84), 130 (34), 102 (98). Anal. calc. for $C_{18}H_{14}N_2O_3$ (306.32): C 70.58, H 4.61, N 9.15; found: C 70.75, H 4.72, N 9.02.

*3-(4-Chlorophenyl)-2-methoxybenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6q**). White crystals, recrystallized from MeOH. Yield 0.51 g, 78%. M.p. 205–207°. IR (KBr): 1699 (C=O), 1542, 1340, 1201. ^{1}H-NMR (400 MHz, $CDCl_3$): 4.04 (*s*, Me); 7.22–8.03 (*m*, 8 arom. H). MS: 326 (91, M^+), 295 (49), 268 (25), 186 (18), 159 (21), 130 (78), 124 (97), 102 (100). Anal. calc. for $C_{17}H_{11}ClN_2O_3$ (326.73): C 62.49, H 3.39, N 8.57; found: C 62.73, H 3.55, N 8.49.

*2-Ethoxy-3-(4-methylphenyl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**6r**). White crystals, recrystallized from MeOH. Yield 0.51 g, 84%. M.p. 186–188°. IR (KBr): 1703 (C=O), 1560, 1353, 1087. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.27 (*t*, *J* = 7.2, Me); 2.43 (*s*, Me); 4.49 (*q*, *J* = 7.2, CH_2O); 7.22–8.03 (*m*, 8 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 13.8; 21.0; 65.0; 112.6; 121.1; 122.5; 123.3; 127.6 (2 C); 129.1; 129.7 (2 C); 131.9; 135.0; 138.6; 141.9; 153.7; 153.9; 157.0. MS: 320 (87, M^+), 292 (55), 186 (100), 159 (88), 130 (35), 102 (60). Anal. calc. for $C_{19}H_{16}N_2O_3$ (320.34): C 71.24, H 5.03, N 8.74; found: C 71.30, H 5.22, N 8.49.

Preparation of 3-{[-3-(2-Hydroxyethyl)oxazolidin-2-ylidene]amino}-2-benzofurancarboxamides **7**. To the soln. of **4** prepared above in CH_2Cl_2 (15 ml) was added 2,2′-iminodiethanol (0.21 g, 2 mmol), and the mixture was allowed to stand for 0.5 h. The soln. was condensed, and anh. EtOH (10 ml) with several drops of EtONa in EtOH was added. The mixture was stirred for 1–6 h at r.t. The soln. was concentrated under reduced pressure and the residue was recrystallized from CH_2Cl_2/PE to give **7**.

*3-{[(2*E*)-3-(2-Hydroxyethyl)oxazolidin-2-ylidene]amino}-*N*-phenyl-2-benzofurancarboxamide* (**7a**). White crystals, recrystallized from CH_2Cl_2/PE 1:1. Yield 0.64 g, 88%. M.p. 184–186°. IR (KBr): 3424, 1678 (C=O), 1633, 1544, 1398, 1036. ^{1}H-NMR (400 MHz, $CDCl_3$): 3.74 (*t*, *J* = 5.2, CH_2N); 3.85 (*t*, *J* = 7.6, CH_2N); 3.97 (*t*, *J* = 5.2, CH_2O); 4.47 (*t*, *J* = 8.0, CH_2O); 7.05–7.67 (*m*, 9 arom. H); 10.07 (*s*, NH). MS: 365 (25, M^+), 322 (35), 229 (77), 186 (69), 114 (100). Anal. calc. for $C_{20}H_{19}N_3O_4$ (365.38): C 65.74, H 5.24, N 11.50; found: C 65.87, H 5.41, N 11.46.

*3-{[(2*E*)-3-(2-Hydroxyethyl)oxazolidin-2-ylidene]amino}-*N*-(4-methylphenyl)-2-benzofurancarboxamide* (**7b**). White crystals, recrystallized from CH_2Cl_2/PE 1:1. Yield 0.62 g, 82%. M.p. 170–171°. IR (KBr): 3354, 1680 (C=O), 1628, 1547, 1410, 1253, 1036. ^{1}H-NMR (400 MHz, $CDCl_3$): 2.27 (*s*, Me); 3.41 (*s*, OH); 3.70 (*t*, *J* = 5.2, CH_2N); 3.81 (*t*, *J* = 8.0, CH_2N); 3.97 (*t*, *J* = 5.2, CH_2O); 4.46 (*t*, *J* = 8.0, CH_2O); 7.05–7.52 (*m*, 8 arom. H); 10.01 (*s*, NH). MS: 379 (34, M^+), 322 (43), 229 (64), 186 (78), 114 (100). Anal. calc. for $C_{21}H_{21}N_3O_4$ (379.41): C 66.48, H 5.58, N 11.08; found: C 66.33, H 5.60, N 11.13.

*General Procedure for the Preparation of 2-(Alkylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones* (**9**). To the soln. of **4** prepared above in CH_2Cl_2 (15 ml) was added alkylamine (2 mmol), and the mixture was allowed to stand for 0.5–4 h. The soln. was condensed, and anh. EtOH (10 ml) with several drops of EtONa in EtOH was added. The mixture was stirred for 2–5 h at r.t. The soln. was concentrated under reduced pressure and the residue was recrystallized to give 2-(alkylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones **9**.

*2-(Ethylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9a**). White crystals, recrystallized from CH_2Cl_2/EtOH 2:1. Yield 0.54 g, 89%. M.p. 197–199°. IR (KBr): 3362, 1703 (C=O), 1534, 1353, 1091. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.16 (*t*, *J* = 7.2, Me); 3.48–3.55 (*q*, *J* = 7.2, CH_2N); 4.09 (*s*, NH); 7.33–8.04 (*m*, 9 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 14.6; 37.0; 112.6; 121.5; 123.0 (2 C); 128.8 (2 C); 129.1; 129.9; 130.6 (2 C); 133.1; 134.3; 144.5; 151.8; 153.7; 157.2. MS: 305 (45, M^+), 275 (25), 260 (32), 160 (41), 130 (67), 102 (100). Anal. calc. for $C_{18}H_{15}N_3O_2$ (305.33): C 70.81, H 4.95, N 13.76; found: C 70.93, H 4.70, N 13.80.

*3-Phenyl-2-(propylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9b**). White crystals, recrystallized from CH_2Cl_2/EtOH 1:1. Yield 0.53 g, 83%. M.p. 160–162°. IR (KBr): 3353, 1699 (C=O), 1540, 1342, 1112. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.86 (*t*, *J* = 7.2, Me); 1.50–1.60 (*m*, CH_2); 3.42–3.47 (*m*, CH_2N); 4.12 (*s*, NH); 7.34–8.05 (*m*, 9 arom. H). MS: 319 (29, M^+), 277 (38), 260 (28), 160 (36), 130 (65), 102 (100). Anal. calc. for $C_{19}H_{17}N_3O_2$ (319.36): C 71.46, H 5.37, N 13.16; found: C 71.43, H 5.26, N 13.28.

*2-(Butylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9c**). White crystals, recrystallized from CH_2Cl_2/EtOH 1:1. Yield 0.49 g, 74%. M.p. 163–164°. IR (KBr): 3342, 1701 (C=O), 1542, 1340, 1110. ^{1}H-NMR (400 MHz, $CDCl_3$): 0.91 (*t*, *J* = 7.2, Me); 1.26–1.52 (*m*, 2 CH_2); 3.44–3.49 (*m*, CH_2N); 4.10 (*s*, NH); 7.32–8.03 (*m*, 9 arom. H). MS: 333 (18, M^+), 289 (43), 261 (46), 160 (27), 130 (61), 102 (100). Anal. calc. for $C_{20}H_{19}N_3O_2$ (333.38): C 72.05, H 5.74, N 12.60; found: C 72.11, H 5.82, N 12.38.

*3-Phenyl-2-(isopropylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9d**). White crystals, recrystallized from CH_2Cl_2/EtOH 1:1. Yield 0.52 g, 82%. M.p. 192–194°. IR (KBr): 3332, 1702 (C=O), 1544, 1346, 1118. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.16 (*d*, *J* = 6.4, 2 Me); 3.88 (*d*, *J* = 7.2, NH); 4.32–4.38 (*m*, CHN); 7.32–8.02 (*m*, 9 arom. H). MS: 319 (100, M^+), 304 (33), 261 (69), 160 (27), 130 (49), 102 (87). Anal. calc. for $C_{19}H_{17}N_3O_2$ (319.36): C 71.46, H 5.37, N 13.16; found: C 71.25, H 5.11, N 13.22.

*2-(Isobutylamino)-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9e**). White crystals, recrystallized from CH_2Cl_2/EtOH 1:1. Yield 0.47 g, 71%. M.p. 152–154°. IR (KBr): 3328, 1698 (C=O), 1543, 1341, 1116. ^{1}H-NMR (400 MHz, $CDCl_3$): (*d*, *J* = 6.8, 2 Me); 1.82–1.86 (*m*, CH); 3.28–3.32 (*m*, CH_2N); 4.15 (*s*, NH); 7.34–8.04 (*m*, 9 arom. H). MS: 333 (80, M^+), 290 (56), 261 (61), 160 (49), 130 (51), 102 (100). Anal. calc. for $C_{20}H_{19}N_3O_2$ (333.38): C 72.05, H 5.74, N 12.60; found: C 72.17, H 5.77, N 12.57.

*3-(4-Methylphenyl)-2-(isopropylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9f**). White crystals, recrystallized from CH_2Cl_2/EtOH 2:1. Yield 0.58 g, 87%. M.p. 200–202°. IR (KBr): 3336, 1699 (C=O), 1540, 1340, 1115. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.08 (*d*, *J* = 6.4, 2 Me); 2.37 (*s*, Me); 3.87 (*d*, *J* = 7.2, NH); 4.24–4.29 (*m*, CHN); 7.10–7.94 (*m*, 8 arom. H). MS: 333 (43, M^+), 301 (33), 261 (56), 160 (54), 130 (61), 102 (100). Anal. calc. for $C_{20}H_{19}N_3O_2$ (333.38): C 72.05, H 5.74, N 12.60; found: C 72.01, H 5.86, N 12.73.

*2-(Cyclohexylamino)-3-(4-methylphenyl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9g**). White crystals, recrystallized from CH_2Cl_2/EtOH 2:1. Yield 0.54 g, 72%. M.p. 209–211°. IR (KBr): 3342, 1695 (C=O), 1541, 1213, 1091. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.06–2.00 (*m*, $(CH_2)_5$); 2.46 (*s*, Me); 4.00–4.08 (*m*, NH, CHN); 7.19–8.04 (*m*, 8 arom. H). ^{13}C-NMR (100 MHz, $CDCl_3$): 21.1; 24.4 (2 C); 25.3; 32.5 (2 C); 50.2; 112.4; 121.4; 122.8; 123.0; 128.2 (2 C); 128.9; 131.2 (2 C); 131.5; 132.9; 140.0; 144.4; 151.2; 153.8; 157.0. MS: 373 (75, M^+), 261 (71), 159 (87), 130 (75), 102 (100). Anal. calc. for $C_{23}H_{23}N_3O_2$ (373.45): C 73.97, H 6.21, N 11.25; found: C 74.21, H 6.24, N 11.15.

*2-(*tert*-Butylamino)-3-(4-methylphenyl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9h**). White crystals, recrystallized from CH_2Cl_2/EtOH 2:1. Yield 0.52 g, 75%. M.p. 205–207°. IR (KBr): 3378, 1704 (C=O), 1542, 1214, 1091. ^{1}H-NMR (400 MHz, $CDCl_3$): 1.43 (*s*, 3 Me); 2.45(*s*, Me); 4.08 (*s*, NH); 7.18–8.01 (*m*, 8 arom. H). MS: 347 (38, M^+), 331 (53), 261 (68), 159 (87), 130 (64), 102 (100). Anal. calc. for $C_{21}H_{21}N_3O_2$ (347.41): C 72.60, H 6.09, N 12.10; found: C 72.64, H 6.22, N 12.01.

*2-[(2-Hydroxyethyl)amino]-3-phenylbenzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9i**). White crystals, recrystallized from EtOH. Yield 0.49 g, 76%. M.p. 224–226°. IR (KBr): 3410, 1705 (C=O), 1544, 1342, 1117. ^{1}H-NMR (400 MHz, $CDCl_3$): 3.58–3.62 (*m*, CH_2N); 3.78 (*t*, *J* = 4.8, CH_2O); 4.18 (*s*, OH); 4.72 (*t*, *J* = 4.8, NH); 7.32–7.96 (*m*, 9 arom. H). MS: 321 (10, M^+), 302 (68), 260 (18), 130 (65), 102 (100). Anal. calc. for $C_{18}H_{15}N_3O_3$ (321.33): C 67.28, H 4.71, N 13.08; found: C 67.15, H 4.78, N 13.27.

*2-[(2-Hydroxyethyl)amino]-3-(4-methylphenyl)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**9j**). White crystals, recrystallized from EtOH. Yield 0.56 g, 84%. M.p. 210–211°. IR (KBr): 3410, 1701 (C=O), 1540, 1342, 1114. ^{1}H-NMR (400 MHz, $CDCl_3$): 2.43 (*s*, Me); 3.59–3.63 (*m*, CH_2N); 3.80 (*t*, *J* = 4.8, CH_2O); 4.28 (*s*, OH); 4.77 (*t*, *J* = 4.8, NH); 7.20–7.97 (*m*, 8 arom. H). MS: 335 (16, M^+), 320 (38), 260

(44), 130 (52), 102 (100). Anal. calc. for $C_{19}H_{17}N_3O_3$ (335.66): C 68.05, H 5.11, N 12.53; found: C 68.11, H 5.26, N 12.35.

*General Procedure for the Preparation of 2-(Arylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-ones* **11**. To the soln. of **4** prepared above in CH_2Cl_2 (15 ml) was added ammonia hydrate (0.08 g, 2 mmol, 80%), methylamine hydrate (0.19 g, 2 mmol, 33%) or hydrazine (0.21 g, 2 mmol) in MeCN (5 ml). The mixture was stirred for 0.5–1 h at r.t. The soln. was concentrated under reduced pressure and the residue was recrystallized to give **11**.

*2-(Phenylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**11a**). White crystals, recrystallized from CH_2Cl_2/EtOH 3:1. Yield 0.50 g, 90%. M.p. > 300°. IR (KBr): 3337, 3171, 1700 (C=O), 1560, 1400, 1211. ^{1}H-NMR (400 MHz, (D_6)DMSO): 7.06–8.01 (*m*, 9 arom. H); 8.82 (*s*, NH); 11.19 (*s*, NH). MS: 277 (21, M^+), 184 (23), 130 (31), 102 (48), 76 (100). Anal. calc. for $C_{16}H_{11}N_3O_2$ (277.28): C 69.31, H 4.00, N 15.15; found: C 69.24, H 3.81, N 15.19.

*3-Methyl-2-(phenylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**11b**). White crystals, recrystallized from CH_2Cl_2/EtOH 3:1. Yield 0.49 g, 84%. M.p. 269–270°. IR (KBr): 3330, 1702 (C=O), 1520, 1400, 1211. ^{1}H-NMR (400 MHz, $CDCl_3$): 3.73 (*s*, Me); 6.58 (*s*, NH); 7.19–7.98 (*m*, 9 arom. H). MS: 291 (100, M^+), 265 (23), 130 (54), 102 (48). Anal. calc. for $C_{17}H_{13}N_3O_2$ (291.30): C 70.09, H 4.50, N 14.42; found: C 70.15, H 4.63, N 14.45.

*3-Amino-2-(phenylamino)benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**11c**). White crystals, recrystallized from CH_2Cl_2/EtOH 3:1. Yield 0.54 g, 92%. M.p. 263–265°. IR (KBr): 3320, 1701 (C=O), 1542, 1402, 1199. ^{1}H-NMR (400 MHz, (D_6)DMSO): 5.85 (*s*, NH_2); 7.09–8.00 (*m*, 9 arom. H); 9.45 (*s*, NH). MS: 292 (21, M^+), 184 (23), 130 (31), 102 (48), 76 (100). Anal. calc. for $C_{16}H_{12}N_4O_2$ (292.29): C 65.75, H 4.14, N 19.17; found: C 65.58, H 4.24, N 19.15.

*3-Amino-2-[(4-methylphenyl)amino]benzofuro[3,2-*d*]pyrimidin-4(3*H*)-one* (**11d**). White crystals, recrystallized from CH_2Cl_2/EtOH 3:1. Yield 0.58 g, 95%. M.p. 292–294°. IR (KBr): 3327, 1702 (C=O), 1544, 1401, 1200, 742. ^{1}H-NMR (400 MHz, (D_6)DMSO): 2.31 (*s*, Me); 5.82 (*s*, NH_2); 7.18–7.98 (*m*, 8 arom. H); 9.35 (*s*, NH). MS: 306 (100, M^+), 288 (76), 201 (35), 130 (29). Anal. calc. for $C_{17}H_{14}N_4O_2$ (306.32): C 66.66, H 4.61, N 18.29; found: C 66.47, H 4.75, N 18.26.

X-Ray Structure of **7a**[1]). The crystal data of **7a** are as follows: formula $C_{20}H_{19}N_3O_4$; M_r 365.38; crystal system: monoclinic; space group: *P*2(1)/*c*; unit cell dimensions: $a = 13.9052(18)$ Å, $b = 10.0674(14)$ Å, $c = 13.5305(18)$ Å, $\beta = 111.119(2)°$, $V = 1766.9(4)$ Å^3; $Z = 4$; $d = 1.374$ Mg/m^3; $\mu(\mathrm{Mo}K_\alpha) = 0.097$ mm^{-1}.

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