Reagents for new heteroannelation reactions. Part VI. 2-(Methylsulfanyl)-1,4,5,6-tetrahydropyrimidine[1]

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Dedicated to Prof. Fritz Sauter on the occasion of his 70th birthday

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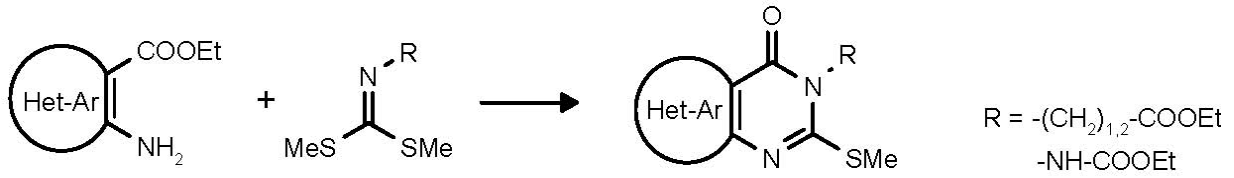
Abstract

The reaction of heteroaromatic 2-aminoesters and 2-aminonitriles with 2-(methylthio)-1,4,5,6-tetrahydropyrimidine results in annelation of a pyrimido[1,2-*a*]pyrimidine moiety in a one-pot process, providing access to a number of predominantly novel tri- and tetracyclic hetero-systems.

Keywords: 2-(Methylsulfanyl)-1,4,5,6-tetrahydropyrimidine, fused pyrimdo[1,2-*a*]pyrimidines, fused S,N-heterocycles

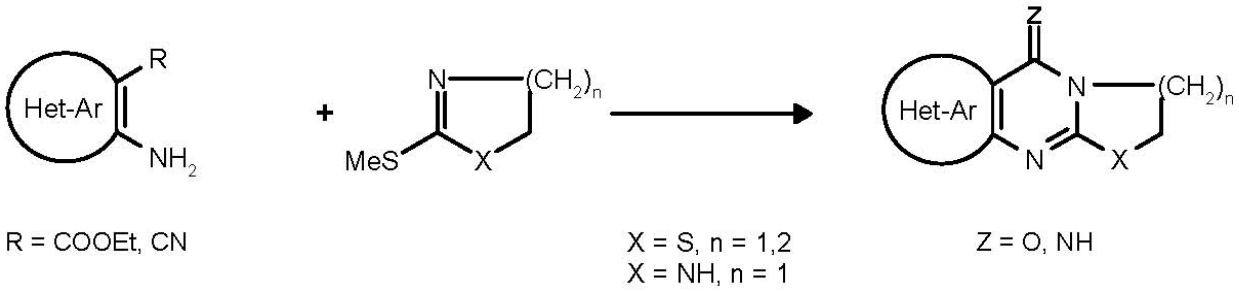
Introduction

In recent papers we have reported on the one-pot annelation of a pyrimidine ring to 2-aminoesters using compounds of the versatile *N*-[bis(methylthio)methylene]amino type (BMMA-type)[2] (Scheme 1) as well as on the extension of this methodology towards cyclic analogs of the BMMA reagents, resulting in double-reactions of imidazo[1,2-*a*]pyrimido,[1] thiazolo[2,3-*b*]pyrimido,[3] and pyrimido[2,1-*b*]thiazino[4] moieties.



Scheme 1

In addition, 2-aminonitriles were employed as starting materials and afforded the corresponding imino derivatives (Scheme 2).

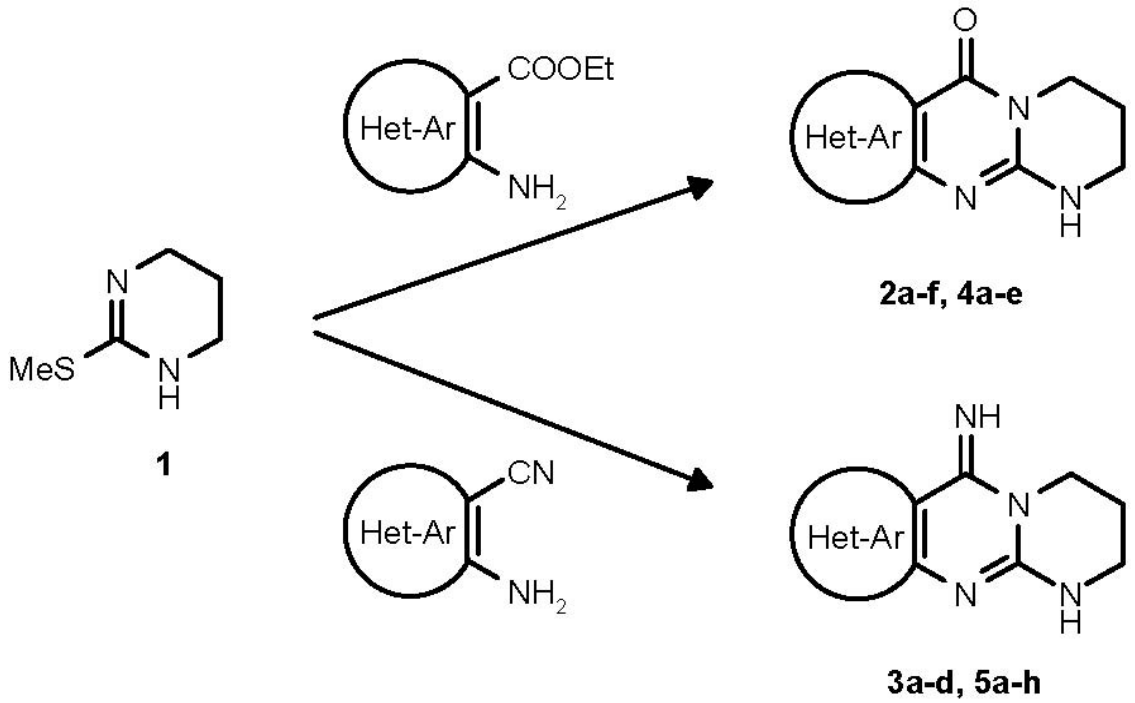


Scheme 2

In continuation of the above-mentioned work with cyclic reagents this paper describes the utilization of 2-(methylsulfanyl)-1,4,5,6-tetrahydropyrimidine (**1**) within the BMMA strategy, thus expanding the scope of this reactions towards the annelation of a pyrimido[1,2-*a*]pyrimidine unit.

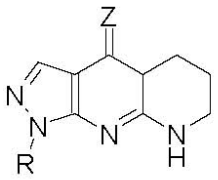
Results and Discussion

The reaction of a variety of heteroaromatic aminoesters and aminonitriles with **1** furnished the desired tri- and tetracyclic fusion products **2**, **3** and **4**, **5**, respectively (Scheme 3). In a series of experiments, solvent and temperature conditions were optimized. Heating the starting materials in HMPA to 150 °C or without solvent to 170 °C for several hours gave best results.



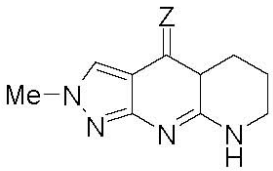
Scheme 3

The reaction of **1** with a number of monocyclic heterocycles with vicinal amino and ester functionalities gave the tricyclic oxo compounds **2**, and from the reaction of **1** with accordingly substituted heterocyclic amino nitriles the imino derivatives **3** were obtained.

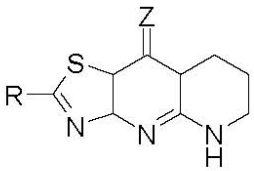


2 Z = O
3 Z = NH

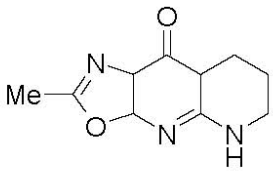
2a R = Me
2b R = Ph
3a R = H
3b R = Me



2c, **3c**



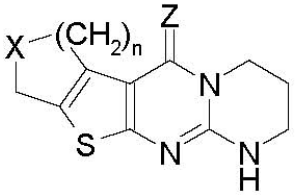
2d R = SMe
2e R = NH-Ph
3d R = SMe



2f

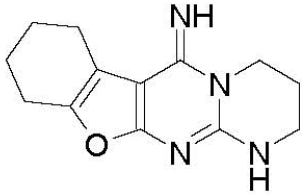
Cycloalkane- and heterocycle-fused thiophenes with 2-amino and 3-ester or 3-nitrile functionalities as well as 2-amino-4,5,6,7-tetrahydrobenzofuran-3-carbonitrile were the suitable starting materials (easily accessible from suitable cyclic ketones via the

Gewald reaction; references are provided in the Experimental part) for the preparation of the corresponding tetracyclic oxo and imino compounds **4** and **5**, respectively.



4 Z = O
5 Z = NH

	n	X
4a, **5a**	1	CH_2
4b, **5b**	2	CH_2
4c, **5c**	3	CH_2
4d, **5d**	2	NCH_2Ph
4e, **5e**	2	S
5f	2	CHMe
5g	2	NMe



5h

Compound **4b** has already been prepared by a similar reaction[5] in very poor yield, and was identified only by its molecular ion peak in a mass spectroscopic analysis. In contrast, the method described here provides easy, one-step access to polynuclear heterocyclic compounds in fair to good yields starting from relatively simple substrates.

Experimental Section

General Procedures. Melting points (mp) were determined on a Kofler hot stage apparatus. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AC 200 spectrometer (200 MHz for ^{1}H; TMS as internal standard, DMSO-d_6 as solvent, δ values in ppm). Elementary analyses were performed at the Microanalytical Laboratory, Institute of Physical Chemistry, University of Vienna (Mag. J. Theiner).
2-(Methylsulfanyl)-1,4,5,6-tetrahydropyrimidine (**1**) was prepared via a two-step procedure from 1,3-propanediamine and CS_2 followed by methylation with methyl iodide (applying procedures originally published for 2-(methylsulfanyl)-2-imidazoline[6,7]). Aminoesters and aminonitriles as starting materials were prepared according to known procedures (references given at the respective experiments).

General procedure for the cyclization reaction

2-(Methylsulfanyl)-1,4,5,6-tetrahydro-pyrimidine (1.56 g, 12 mmol) and the appropriate aminoester or aminonitrile (10 mmol) were heated under a nitrogen atmosphere either in HMPA (10 mL) to 150 °C or without solvent to 170 °C for a given period of time. After

cooling to room temperature crushed ice was added, and the mixture was stirred for 1 h. The separated product was collected by filtration and recrystallized from methanol (unless otherwise stated).

6,7,8,9-Tetrahydro-1-methylpyrazolo[3,4-*d*]pyrimido[1,2-*a*]pyrimidin-4(1*H*)-one (2a). From ethyl 5-amino-1-methylpyrazole-4-carboxylate;[8] neat at 170 °C; 2 h. Yield 1.40 g colorless crystals, 68%; mp 284 °C (ethanol/water 10:1); ^{1}H NMR (DMSO-d_6): δ 8.00 (s, 1H), 7.70 (s, 1H), 3.80 (m, 2H), 3.60 (s, 3H), 3.30 (m, 2H), 1.90 (m, 2H); ^{13}C NMR (DMSO-d_6): δ 157.12 (s), 152.82 (s), 152.33 (s), 133.94 (d), 98.43 (s), 38.78 (t), 38.68 (t), 33.04 (t), 19.84 (q).

6,7,8,9-Tetrahydro-1-phenylpyrazolo[3,4-*d*]pyrimido[1,2-*a*]pyrimidin-4(1*H*)-one (2b). From ethyl 5-amino-1-phenylpyrazole-4-carboxylate;[9] neat at 170 °C; 2 h. Yield 1.50 g colorless crystals, 56%; mp 275 °C; ^{1}H NMR (DMSO-d_6): δ 8.30 (m, 2H), 8.00 (s, 1H), 7.50 (m, 2H), 7.30 (m, 1H), 3.90 (m, 2H), 3.30 (m, 2H), 1.90 (m, 2H); ^{13}C NMR (DMSO-d_6): δ 156.99 (s), 153.11 (s), 152.60 (s), 139.14 (d), 135.90 (s), 128.61 (2d), 125.40 (d), 120.41 (2d), 99.94 (s), 43.64 (t), 43.44 (t), 19.64 (t). Anal. Calcd. for $C_{14}H_{13}N_5O$ (267.29): C, 62.91; H, 4.90; N, 26.20. Found: C, 62.62; H, 4.66; N, 26.06.

6,7,8,9-Tetrahydro-2-methylpyrazolo[3,4-*d*]pyrimido[1,2-*a*]pyrimidin-4(2*H*)-one (2c). From ethyl 3-amino-1-methyl-1*H*-pyrazole-4-carboxylate;[8] neat at 170 °C; 2 h. Yield 0.82 g colorless crystals, 40%; mp 260 °C; ^{1}H NMR (DMSO-d_6): δ 8.20 (s, 1H), 7.80 (s, 1H), 3.80 (m, 2H), 3.20 (m, 2H), 3.10 (s, 3H), 1.90 (m, 2H).

5,6,7,8-Tetrahydro-2-(methylsulfanyl)-10*H*-pyrimido[1,2-*a*]thiazolo[4,5-*d*]pyrimidin-10-one (2d). From ethyl 4-amino-2-(methylsulfanyl)thiazole-5-carboxylate;[10] in HMPA at 150 °C; 5 h. Yield 1.76 g colorless crystals, 69%; mp 286 °C; ^{1}H NMR (DMSO-d_6): δ 8.00 (s, 1H), 3.80 (m, 2H), 3.30 (m, 2H), 2.80 (s, 3H), 1.90 (m, 2H). Anal. calcd for $C_9H_{10}N_4OS_2$ (254.32): C, 42.50; H, 3.96; N, 22.03. Found: C, 42.23; H, 3.66; N, 21.76.

5,6,7,8-Tetrahydro-2-(phenylamino)-10*H*-pyrimido[1,2-*a*]thiazolo[4,5-*d*]pyrimidin-10-one (2e). From ethyl 4-amino-2-(phenylamino)thiazole-5-carboxylate;[11] in HMPA at 150 °C; 5 h. Yield 1.86 g colorless crystals, 62%; mp >320 °C; ^{1}H NMR (DMSO-d_6): δ 7.90 (s, 1H), 7.70 (m, 2H), 7.40 (m, 2H), 7.00 (m, 1H), 3.90 (m, 2H), 3.30 (m, 2H), 1.90 (m, 2H). Anal. Calcd. for $C_{14}H_{13}N_5OS$ (299.35): C, 56.17; H, 4.38; N, 23.40. Found: C, 55.72; H, 4.16; N, 22.97.

5,6,7,8-Tetrahydro-2-methyl-10*H*-oxazolo[5,4-*d*]pyrimido[1,2-*a*]pyrimidin-10-one (2f). From ethyl 5-amino-2-methyloxazole-4-carboxylate;[12] neat at 170 °C; 45 min; purified by flash chromatography (SiO_2, chloroform/acetone 9:2). Yield 0.73 g colorless crystals, 35%; mp 304 °C; ^{1}H NMR (DMSO-d_6): δ 8.10 (s, 1H), 3.90 (m, 2H), 3.30 (m,

2H), 2.40 (s, 3H), 1.90 (m, 2H); ^{13}C NMR (DMSO-d_6): δ 164.60 (s), 155.47 (s), 155.11 (s), 152.21 (s), 109.64 (s), 39.49 (t), 39.08 (t), 19.24 (t), 13.75 (q). Anal. Calcd. for $C_9H_{10}N_4O_2$ (206.20): C, 52.42; H, 4.89; N, 27.17. Found: C, 52.14; H, 4.70; N, 26.88.

6,7,8,9-Tetrahydropyrazolo[3,4-*d*]pyrimido[1,2-*a*]pyrimidin-4(1*H*)-imine (3a). From 5-amino-1*H*-pyrazol-4-carbonitrile;[13] in HMPA; 5 h. Yield 0.88 g colorless crystals, 46%; mp 230 °C; ^{1}H NMR (DMSO-d_6): δ 7.80 (s, 1H), 7.40 (s, 1H), 3.90 (m, 2H), 3.20 (m, 2H), 1.80 (m, 2H); ^{13}C NMR (DMSO-d_6): δ 154.14 (s), 151.86 (s), 151.20 (s), 134.46 (d), 97.93 (s), 39.92 (t), 38.63 (t), 20.51 (t).

6,7,8,9-Tetrahydro-1-methylpyrazolo[3,4-*d*]pyrimido[1,2-*a*]pyrimidin-4(1*H*)-imine (3b). From 5-amino-1-methyl-1*H*-pyrazol-4-carbonitrile;[14] neat at 170 °C; 4 h. Yield 1.41 g yellow crystals, 69%; mp 200 °C; ^{1}H NMR (DMSO-d_6): δ 7.80 (s, 1H), 7.60 (s,1H), 7.20 (s, 1H), 3.90 (m, 2H), 3.60 (s, 3H), 3.20 (m, 2H), 1.90 (m, 2H); ^{13}C NMR (DMSO-d_6): δ 153.78 (s), 151.90 (s), 149.30 (s), 133.94 (d), 97.98 (s), 39.85 (t), 38.58 (t), 33.86 (q), 20.34 (t).

6,7,8,9-Tetrahydro-2-methylpyrazolo[3,4-*d*]pyrimido[1,2-*a*]pyrimidin-4(2*H*)-imine (3c). From 3-amino-1-methyl-1*H*-pyrazole-4-carbonitrile;[15] neat at 170 °C; 2 h. Yield 0.98 g colorless crystals, 48%; mp 302 °C (ethanol/water 10:1); ^{1}H NMR (DMSO-d_6): δ 8.00 (s, 1H), 7.50 (s, 1H), 7.20 (s, 1H), 3.90 (m, 2H), 3.70 (s, 3H), 3.20 (m, 2H), 1.90 (m, 2H). Anal. calcd for $C_9H_{12}N_6$ (204.23): C, 52.93; H, 5.92; N, 41.15; Found: C, 52.95; H, 5.86; N, 40.82.

5,6,7,8-Tetrahydro-2-(methylsulfanyl)-10*H*-pyrimido[1,2-*a*]thiazolo[4,5-*d*]pyrimidin-10-imine (3d). From 4-amino-2-(methylsulfanyl)thiazole-5-carbonitrile;[16] in HMPA at 150 °C; 4 h. Yield 1.52 g yellow crystals, 60%; mp 258 °C; ^{1}H NMR (DMSO-d_6): δ 7.70 (s, 1H), 3.90 (m, 2H), 3.30 (m, 2H), 2.70 (s, 3H), 1.90 (m, 2H). Anal. Calcd. for $C_9H_{11}N_5S_2$ (253.34): C, 42.67; H, 4.3; N, 27.64. Found: C, 42.88; H, 4.02; N, 27.36.

1,3,4,7,8,9-Hexahydro-2*H*,6*H*-cyclopenta[4,5]thieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-one (4a). From ethyl 2-amino-5,6-dihydro-4*H*-cyclopenta[*b*]thiophene-3-carboxylate;[17] in HMPA at 150 °C; 3 h. Yield 1.88 g colorless crystals, 76%; mp 310 °C; ^{1}H NMR (DMSO-d_6): δ 7.80 (s, 1H), 3.90 (m, 2H), 3.30 (m, 2H), 2.90–2.70 (m, 4H), 2.30 (m, 2H), 1.90 (m, 2H). Anal. Calcd. for $C_{12}H_{13}N_3OS$ (247.31): C, 58.28; H, 5.30; N, 16.99. Found: C, 58.06; H, 5.21; N, 16.78.

1,2,3,4,7,8,9,10-Octahydro-6*H*-[1]benzothieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-one (4b). From ethyl 2-amino-4,5,6,7-tetrahydrobenzo[*b*]thiophene-3-carboxylate;[17] in HMPA at 150 °C; 3 h. Yield 2.17 g colorless crystals, 83%; mp 278 °C; H NMR (DMSO-d_6): δ 7.80 (s, 1H), 3.80 (m, 2H), 3.70 (m, 2H), 2.80 (m, 2H), 2.60 (m, 2H),

1.90 (m, 2H), 1.70 (m, 4H); ^{13}C NMR (DMSO-d_6): δ 165.18 (s), 157.54 (s), 150.57 (s), 130.03 (s), 123.59 (s), 112.02 (s), 38.47 (t), 38.47 (t), 25.28 (t), 24.18 (t), 22.74 (t), 21.95 (t), 19.70 (t).

1,3,4,7,8,9,10,11-Octahydro-2*H*,6*H*-cyclohepta[4,5]thieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-one (4c). From ethyl 2-amino-5,6,7,8-tetrahydro-4*H*-cyclohepta[*b*]thiophene-3-carboxy-late;[18] in HMPA at 150 °C; 3 h. Yield 2.07 g pale yellow crystals, 75%; mp 240 °C; ^{1}H NMR (DMSO-d_6): δ 7.70 (s, 1H), 3.80 (m, 2H), 3.30 (m, 2H), 3.10 (m, 2H), 2.60 (m, 2H), 1.90 (m, 2H), 1.80 (m, 2H), 1.50 (m, 4H). Anal. Calcd. for $C_{14}H_{17}N_3OS$ (275.37): C, 61.07; H, 6.22; N, 15.26. Found: C, 60.91; H, 5.99; N, 15.10.

1,2,3,4,7,8,9,10-Octahydro-9-(phenylmethyl)pyrido[4',3':4,5]thieno[2,3-*d*]pyrimido-[1,2-*a*]pyrimidin-6-one (4d). From ethyl 2-amino-4,5,6,7-tetrahydro-6-(phenylmethyl)-thieno[2,3-*c*]pyridine-3-carboxylate;[19] in HMPA at 150 °C; 3 h. Yield 2.79 g pale yellow crystals, 79%; mp 245 °C; ^{1}H NMR (DMSO-d_6): δ 7.80 (s, 1H), 7.40–7.10 (m, 5H), 3.90 (m, 2H), 3.70 (s, 2H), 3.50 (s, 2H), 3.20 (m, 2H), 2.80 (m, 2H), 2.70 (m, 2H), 1.90 (m, 2H). Anal. Calcd. for $C_{19}H_{20}N_4OS$ (352.45): C, 64.75; H, 5.72; N, 15.90. Found: C, 64.55; H, 5.48; N, 15.69.

1,3,4,7,8,10-Hexahydro-2*H*,6*H*-pyrimido[1,2-*a*]thiopyrano[4',3':4,5]thieno-[2,3-*d*]-pyrimidin-6-one (4e). From ethyl 2-amino-4,7-dihydro-5*H*-thieno[2,3-*c*]thiopyran-3-carboxylate;[20] in HMPA at 150 °C; 3 h. Yield 2.26 g colorless crystals, 81%; mp >320 °C; ^{1}H NMR (DMSO-d_6): δ 7.80 (s, 1H), 3.85 (m, 2H), 3.73 (s, 2H), 3.25 (m, 2H), 3.05 (m, 2H), 2.90 (m, 2H), 1.90 (m, 2H). Anal. Calcd. for $C_{12}H_{13}N_3OS_2$ (279.37): C, 51.59; H, 4.69; N, 15.04. Found: C, 51.32; H, 4.45; N, 14.82.

1,3,4,7,8,9-Hexahydro-2*H*,6*H*-cyclopenta[4,5]thieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-imine (5a). From 2-amino-5,6-dihydro-4*H*-cyclopenta[*b*]thiophene-3-carbonitrile;[21] in HMPA at 150 °C; 2 h. Yield 1.50 g red crystals, 61%; mp 310 °C; ^{1}H NMR (DMSO-d_6): δ 7.50 (s, 1H), 6.50 (s, 1H) 3.90 (m, 2H), 3.20 (m, 2H), 2.90 (m, 2H), 2.70 (m, 2H), 2.40 (m, 2H), 1.90 (m, 2H). Anal. Calcd. for $C_{12}H_{14}N_4S$ (246.33): C, 58.51; H, 5.73; N, 22.74. Found: C, 58.29; H, 5.65; N, 22.52.

1,2,3,4,7,8,9,10-Octahydro-6*H*-[1]benzothieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-imine (5b). From 2-amino-4,5,6,7-tetrahydrobenzo[*b*]thiophene-3-carbonitrile[17]; in HMPA at 150 °C; 2 h. Yield 1.75 g pale brown crystals, 67%; mp 277 °C; ^{1}H NMR (DMSO-d_6): δ 7.40 (s, 1H), 6.70 (s, 1H), 3.90 (m, 2H), 3.20 (m, 2H), 2.70 (m, 2H), 2.50 (m, 2H), 1.90 (m, 2H), 1.70 (m, 4H). Anal. Calcd. for $C_{13}H_{16}N_4S$ (260.36): C, 59.97; H, 6.20; N, 21.52. Found: C, 59.73; H, 6.20; N, 21.21.

1,3,4,7,8,9,10,11-Octahydro-2*H*,6*H*-cyclohepta[4,5]thieno[2,3-*d*]pyrimido[1,2-

***a*]pyrimidin-6-imine (5c).** From 2-amino-5,6,7,8-tetrahydro-4*H*-cyclohepta[*b*]thiophene-3-carbonitrile;[22] in HMPA at 150 °C; 1 h. Yield 1.79 g pale brown crystals, 65%; mp 248 °C; ^{1}H NMR (DMSO-d_6): δ 7.40 (s, 1H), 6.70 (s, 1H), 3.80 (m, 2H), 3.20 (m, 2H), 3.10 (m, 2H), 2.60 (m, 2H), 1.90 (m, 2H), 1.80 (m, 2H), 1.60 (m, 4H). Anal. Calcd. for $C_{14}H_{18}N_4S$ (274.38): C, 61.28; H, 6.61; N, 20.42. Found: C, 61.01; H, 6.33; N, 20.22.

1,2,3,4,7,8,9,10-Octahydro-9-(phenylmethyl)-6*H*-pyrido[4',3':4,5]thieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-imine (5d). From 2-amino-4,5,6,7-tetrahydro-6-(phenylmethyl)thieno[2,3-*c*]pyridine-3-carbonitrile;[23] in HMPA at 150 °C; 3 h. Yield 2.53 g pale brown crystals, 72%; mp 261 °C; ^{1}H NMR (DMSO-d_6): δ 7.40 (s, 1H), 7.30 (m, 5H), 6.60 (s, 1H), 3.90 (m, 2H), 3.60 (s, 2H), 3.50 (s, 2H), 3.20 (m, 2H), 2.90 (m, 2H), 2.70 (m, 2H), 1.90 (m, 2H). Anal. Calcd. for $C_{19}H_{21}N_5S$ (351.47): C, 64.93; H, 6.02; N, 19.93. Found: C, 64.62; H, 5.80; N, 19.67.

1,3,4,7,8,10-Hexahydro-2*H*,6*H*-pyrimido[1,2-*a*]thiopyrano[4',3':4,5]thieno[2,3-*d*]-pyrimidin-6-imine (5e). From 2-amino-4,7-dihydro-5*H*-thieno[2,3-*c*]thiopyran-3-carbo-nitrile;[23] in HMPA at 150 °C; 3 h. Yield 1.81 g red crystals, 65%; mp 295 °C; ^{1}H NMR (DMSO-d_6): δ 7.50 (s, 1H), 6.70 (s, 1H), 3.80 (m, 2H), 3.70 (s, 2H), 3.20 (m, 2H), 3.10 (m, 2H), 2.90 (m, 2H), 1.90 (m, 2H). Anal. Calcd for $C_{12}H_{14}N_4S_2$ (278.39): C, 51.77; H, 5.07; N, 20.13. Found: C, 51.58; H, 4.90; N, 19.93.

1,2,3,4,7,8,9,10-Octahydro-9-methyl-6*H*-[1]benzothieno[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-imine (5f). From 2-amino-4,5,6,7-tetrahydro-6-methylbenzo[*b*]thio-phene-3-carbonitrile;[22] in HMPA at 150 °C; 2 h. Yield 2.0 g yellow crystals, 73%; mp 274 °C; ^{1}H NMR (DMSO-d_6): δ 7.60 (s, 1H), 6.60 (s, 1H), 3.80 (m, 2H), 3.20 (m, 2H), 2.90–1.20 (m, 9H), 1.10 (d, 3H, J=7.1Hz). Anal. Calcd. for $C_{14}H_{18}N_4S$ (274.38): C, 61.28; H, 6.61; N, 20.42. Found: C, 60.97; H, 6.40; N, 20.22.

1,2,3,4,7,8,9,10-Octahydro-9-methyl-6*H*-pyrido[4',3':4,5]thieno[2,3-*d*]pyrimido[1,2-*a*]-pyrimidin-6-imine (5g). From 2-amino-4,5,6,7-tetrahydro-6-methylthieno[2,3-c]pyridine-3-carbonitrile;[24] in HMPA at 150 °C; 3 h. Yield 1.87 g yellow crystals, 68%; mp 288 °C; ^{1}H NMR (DMSO-d_6): δ 7.60 (s, 1H), 6.70 (s, 1H), 3.90 (m, 2H), 3.40 (s, 2H), 3.20 (m, 2H), 2.80 (m, 2H), 2.60 (m, 2H), 2.30 (s, 3H), 1.90 (m, 2H). Anal. Calcd. for $C_{13}H_{17}N_5S$ (275.37): C, 56.70; H, 6.22; N, 25.43. Found: C, 56.62; H, 6.12; N, 25.24.

1,2,3,4,7,8,9,10-Octahydro-6*H*-benzofuro[2,3-*d*]pyrimido[1,2-*a*]pyrimidin-6-imine (5h). From 2-amino-4,5,6,7-tetrahydrobenzofuran-3-carbonitrile;[25] in HMPA at 150 °C; 1 h. Yield 1.52 g colorless crystals, 62%; mp 290 °C; ^{1}H NMR (DMSO-d_6): δ 7.50 (s, 1H), 6.30 (s, 1H), 3.80 (m, 2H), 3.20 (m, 2H), 2.50 (m, 4H), 1.90 (m, 2H), 1.70 (m,

4H). Anal. Calcd. for $C_{13}H_{16}N_4O$ (244.30): C, 63.92; H, 6.60; N, 22.93. Found: C, 63.78; H, 6.65; N, 22.64.

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