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SUPPLEMENTARY MATERIAL TO

Synthesis of various fused pyrimidine rings and their pharmacological and antimicrobial evaluation

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SOME PHYSICAL, ANALYTIC AND SPECTRAL DATA FOR THE PREPARED COMPOUNDS

*6-(1,3-Benzodioxol-5-yl)-4-oxo-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylic acid (**1g**)*. Yield: 53 %; Pale brown crystals; m.p.: 223 °C; Anal. Calcd. for $C_{12}H_8N_2O_5S$: C, 49.31; H, 2.76; N, 9.58; S, 10.97 %. Found: C, 49.15; H, 2.70, N, 9.72; S, 10.98 %; IR (KBr, cm^{-1}): 3416 (–OH), 3280/2978 (–NH), 1718 (–C=O acid), 1657 (–C=O amide), 1635 (–C=N), 1595 (C=C– aromatic ring), 1195/2587 (C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 6.09 (2H, *s*, OCH_2O), 6.94–7.34 (3H, *m*, aromatic), 11.2 (1H, *s*, OH, D_2O exchangeable), 12.2, 12.8 (2H, *s*, 2 NH, D_2O exchangeable); ^{13}C-NMR (75 MHz, DMSO-d_6,, δ / / ppm): 171.3 (C_2), 162.8 (C_4), 98.5 (C_5), 168.9 (C_6), 165.2 (COOH), 101.4 (O–CH_2–O), 130.1 (Ar-C), 117.7 (Ar-C), 111.7 (Ar-C), 147.4 (Ar-C), 149.1 (Ar-C), 110.3 (Ar-C); MS (*m*/*z*, (relative abundance, %)): 292 (M^+, 13), 248, 128, 122, 98, 84, 78, 72, 60 (BP, 100).

*4-(1,3-Benzodioxol-5-yl)-2-hydrazino-6-oxo-1,6-dihydropyrimidine-5-carbonitrile (**1h**)*. Yield: 65 % from **1a**, and 72 % from **1c**; Brown crystals; m.p.: 240 °C; Anal. Calcd. for $C_{12}H_9N_5O_3$: C, 53.14; H, 3.34; N, 25.82 %. Found: C, 53.45; H, 3.27; N, 25.76 %; IR (KBr, cm^{-1}): 3584, 3435, 3327 (–NH, –NH_2), 2213 (–C≡N), 1686 (–C=O), 1634 (–C=N), 1594 (C=C– aromatic ring); ^{1}H--NMR (300 MHz, DMSO-d_6, δ / ppm): 6.19 (2H, *s*, OCH_2O), 7.82–7.16 (3H, *m*, aromatic), 8.43 (2H, *s*, 2 NH, D_2O exchangeable), 13.18 (2H, *s*, NH_2, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 273 ([M + 2]$^+$, 37), 271 (M^+, 44), 257 (BP, 100), 256, 241, 215, 214, 186, 148, 121, 118, 93, 76, 64, 63.

*Ethyl 2-{[4-(1,3-benzodioxol-5-yl)-5-cyano-6-oxo-1,6-dihydropyrimidin-2--yl]thio}acetate (**2a**)*. Yield: 71 %; Brown crystals; m.p.: 221 °C; Anal. Calcd. for $C_{16}H_{13}N_3O_5S$: C, 53.48; H, 3.65; N, 11.69; S, 8.92 %. Found: C, 53.65; H, 3.77;

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N, 11.84; S, 9.02 %; IR (KBr, cm^{-1}): 3453 (–NH), 2795 ($–CH_2–S$), 2215 (–C≡N), 1737 (–C=O ester), 1658 (–C=O amide), 1632 (–C=N), 1610 (C=C– aromatic ring); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.26 (3H, *t*, *J* = 7.0 Hz, CH_3), 4.10 (2H, *q*, *J* = 7.0 Hz, CH_2), 4.13 (2H, *s*, CH_2S), 6.17 (2H, *s*, OCH_2O), 7.81–7.12 (3H, *m*, aromatic), 10.92 (1H, *s*, NH, D_2O exchangeable); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 159.98 (C_2), 170.19 (C_4), 101.19 (C_5), 166.42 (C_6), 112.81 (CN), 49.69 (SCH_2), 168.34 (OCO), 61.09 (OCH_2), 13.93 (CH_3), 102.12 (OCH_2O), 147.34 (2 Ar-C), 111.81 (2 Ar-C), 121.73 (Ar-C), 124.44 (Ar-C); MS (*m*/*z*, (relative abundance, %)): 359 (M^+, 6.13), 273, 229, 178 (BP, 100), 122, 83.

*Ethyl 2-{[5-cyano-4-(3,4-dimethoxyphenyl)-6-oxo-1,6-dihydropyrimidin-2--yl]thio}acetate (**2b**)*. Yield: 72 %; m.p.: 178 °C; orange crystals; Anal. Calcd. for $C_{17}H_{17}N_3O_5S$: C, 54.39; H, 4.56; N, 11.19; S, 8.54 %. Found: C, 54.46; H, 4.49; N, 11.12; S, 8.62 %; IR (KBr, cm^{-1}): 3446 (–NH), 2835 ($–CH_2–S$), 2218 (–C≡N), 1746 (–C=O ester), 1648 (–C=O amide), 1630 (–C=N), 1610 (C=C– aromatic ring); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.32 (3H, *t*, *J* = 7.0 Hz, CH_3), 3.85, 3.88 (6H, 2 *s*, 2 OCH_3), 4.08 (2H, *s*, SCH_2), 4.46 (2H, *q*, *J* = 7.0 Hz, CH_2), 7.76–7.12 (3H, *m*, aromatic), 12.35 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 375 (M^+, 46.7), 330, 194 (BP, 100), 151 (60.0).

*Ethyl 2-{[6-(1,3-benzodioxol-5-yl)-5-cyano-2-thioxo-1,2-dihydropyrimidin--4-yl]oxy}acetate (**3a**)*. Yield: 57 %; m.p.: 220 °C, brown crystals; Anal. Calcd. for $C_{16}H_{13}N_3O_5S$: C, 53.48; H, 3.65; N, 11.69; S, 8.92 %. Found: C, 53.29; H, 3.48; N, 11.82; S, 9.05 %; IR (KBr, cm^{-1}): 3335/2985 (–NH), 2810 ($O–CH_2C=O$), 2216 (–C≡N), 1728 (–C=O ester), 1635 (–C=N), 1608 (C=C– aromatic ring), 1168/2592 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.25 (3H, *t*, *J* = 7.0 Hz, CH_3), 4.13 (2H, *s*, OCH_2CO), 4.00 (2H, *q*, *J* = 7.0 Hz, CH_2), 6.17 (2H, *s*, OCH_2O), 7.82–7.31 (3H, *m*, aromatic), 11.17 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 359 (M^+, 15), 334, 314, 287, 273, 115 (BP, 100), 109, 93, 77.

*Ethyl 2-{[5-cyano-6-(3,4-dimethoxyphenyl)-2-thioxo-1,2-dihydropyrimidin--4-yl]oxy}acetate (**3b**)*. Yield: 61 %; m.p.: 178 °C; orange crystals; Anal. Calcd. for $C_{17}H_{17}N_3O_5S$: C, 54.39; H, 4.56; N, 11.19; S, 8.63 %. Found: C 54.46, H 4.48, N 11.39, S 8.74 %; IR (KBr, cm^{-1}): 3345/2955 (–NH), 2208 (–C≡N), 1732 (–C=O ester), 1628 (–C=N), 1598 (C=C– aromatic ring), 1210/2594 (–C=S/–SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.37 (3H, *t*, *J* = 7.0 Hz, CH_3), 3.87, 3.83 (6H, 2*s*, 2 OCH_3), 4.08 (2H, *s*, OCH_2), 4.43 (2 H, *q*, *J* = 7.0 Hz, CH_2), 7.76––7.15 (3H, *m*, aromatic), 11.36 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 375 (M^+, 47), 330, 323, 230, 195, 194 (BP, 100), 77, 76, 69, 64, 63, 62, 53, 50.

*Ethyl 2-{[5-acetyl-6-(3,4-dimethoxyphenyl)-2-thioxo-1,2-dihydropyrimidin--4-yl]oxy}acetate (**3c**)*. Yield: 70 %; m.p.: 280 °C dec.; yellow crystals; Anal.

Calcd. for $C_{18}H_{20}N_2O_6S$: C, 55.09; H, 5.14; N, 7.14; S, 8.17 %. Found: C, 55.24; H 5.10; N, 7.20; S, 8.22 %; IR (KBr, cm^{-1}): 3115/3023/2934 (–NH), 2685 (–CH_2CO), 2218 (–C≡N), 1750 (–C=O ester), 1678 (–C=O acetyl), 1639 (–C=N), 1605 (–C=C aromatic ring), 1280/2610 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.32 (3H, *t*, *J* = 7.0 Hz, CH_3), 2.06 (3H, *s*, $COCH_3$), 3.75, 3.82 (6H, 2 *s*, 2 OCH_3), 4.22 (2H, *q*, *J* = 7.0 Hz, CH_2), 5.02 (2H, *s*, OCH_2), 6.68––7.38 (3H, *m*, aromatic), 11.72 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 392 (M^+, 10), 360, 328, 209, 207 (BP, 100), 177, 97, 95, 90, 77, 63, 56, 55, 50.

*Ethyl 6-(3,4-dimethoxyphenyl)-4-(2-ethoxy-2-oxoethoxy)-2-thioxo-1,2-dihydropyrimidine-5-carboxylate (**3d**)*. Yield: 64 %; m.p.: > 300 °C; pale brown crystals; Anal. Calcd. for $C_{19}H_{22}N_2O_7S$: C, 54.02; H, 5.25; N, 6.63; S, 7.59 %. Found: C, 53.94; H, 5.30; N, 6.56; S, 7.70 %; IR (KBr, cm^{-1}): 3119/3023 (–NH), 2682 (–CH_2CO), 2216 (–C≡N), 1732 (–C=O ester), 1714 (–C=O ester), 1640 (–C=N), 1597 (C=C– aromatic ring), 1265/2595 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.31 (6H, 2 *t*, *J* = 7.0 Hz , 2 CH_3), 3.85 (6H, *s*, 2 OCH_3), 4.28, 4.45 (4H, 2 *q*, *J* = 7.0 Hz, 2 CH_2), 5.12 (2H, *s*, OCH_2), 6.56–7.44 (3H, *m*, aromatic), 11.89 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 422 (M^+, 24), 377, 348, 142 (BP, 100), 141, 128, 126, 51.

*Ethyl 5-amino-4-(1,3-benzodioxol-5-yl)-2-thioxo-1,2-dihydrofuro[2,3-*d*]pyrimidine-6-carboxylate (**4a**)*.Yield: 63 %; m.p.: 192 °C dec. brown crystals; Anal. Calcd. for $C_{16}H_{13}N_3O_5S$: C, 53.48; H, 3.65; N, 11.69; S, 8.92 %. Found: C, 54.16; H, 3.48; N, 11.74; S, 8.73 %; IR (KBr, cm^{-1}): 3382 /3194 /2910 (–NH_2/ /–NH), 1744 (–C=O ester), 1635 (–C=N stretching), 1612 (C=C– aromatic ring), 1208/2592 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.37 (3H, *t*, *J* = 7.0 Hz, CH_3), 4.54 (2H, *q*, *J* = 7.0 Hz , CH_2), 6.18 (2H, *s*, OCH_2O), 7.76––7.15 (3H, *m*, aromatic), 10.85 (1H, *s*, NH_2, D_2O exchangeable), 13.17 (2H, *s*, NHC=S, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 359 (M^+, 44), 314, 313, 286, 285 (BP, 100), 271, 216, 148, 51.

*Ethyl 5-amino-4-(3,4-dimethoxyphenyl)-2-thioxo-1,2-dihydrofuro[2,3-*d*]pyrimidine-6-carboxylate (**4b**)*. Yield: 61 %; mp > 300 °C; brown crystals; Anal. Calcd. for $C_{17}H_{17}N_3O_5S$: C, 54.39; H, 4.56; N, 11.19; S, 8.54 %. Found: C, 54.32; H, 3.68; N, 11.14; S, 8.65 %; IR (KBr, cm^{-1}): 3372/3185/2928 (–NH_2/ /–NH), 1738 (–C=O ester), 1640 (–C=N), 1608 (C=C– aromatic ring), 1185/2584 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.35 (3H, *t*, J = 7.0 Hz, CH_3), 3.85 (6H, *s*, 2 OCH_3), 4.46 (2H, *q*, *J* = 7.0 Hz, CH_2), 7.16–7.45 (3H, *m*, aromatic), 10.48 (1H, *s*, NH_2, D_2O exchangeable), 13.37 (2H, *s*, NHC=S, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 376 ([M–1]$^+$, 8.2), 274, 149 (BP, 100), 148, 104, 60.

*Ethyl 4-(3,4-dimethoxyphenyl)-5-methyl-2-thioxo-1,2-dihydrofuro[2,3-*d*]pyrimidine-6-carboxylate (**4c**)*. Yield: 66 %; m.p.: 262 °C dec. brown crystals; Anal.

Calcd. for $C_{18}H_{18}N_2O_5S$: C, 57.74; H, 4.85; N, 7.48; S, 8.56 %. Found: C, 57.62; H, 3.68; N, 7.60; S, 8.65 %; IR (KBr, cm^{-1}): 3185/3094 (–NH), 1716 (–C=O ester), 1650 (–C=N), 1598 (C=C– aromatic ring), 1205/2678 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.92 (3H, *t*, *J* = 7.0 Hz, CH_3), 2.58 (3H, *s*, CH_3), 3.38 (6H, *s*, 2 OCH_3), 4.39 (2H, *q*, *J* = 7.0 Hz, CH_2), 6.58–7.36 (3H, *m*, aromatic), 10.81 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 374 (M^+, 9), 342, 149 (BP, 100), 94, 68, 66, 65, 55, 51, 50.

Ethyl 4-(3,4-dimethoxyphenyl)-5-oxo-2-thioxo-1,2,5,6-tetrahydrofuro[2,3-d]pyrmidine-6-carboxylate (***4d***). Yield: 55 %; m.p.: 190 °C; brown crystals; Anal. Calcd. for $C_{17}H_{16}N_2O_6S$: C, 54.25; H, 4.29; N, 7.44; S, 8.52 %. Found: C, 54.58; H, 4.33; N, 7.62; S, 8.48 %; IR (KBr, cm^{-1}): 3235 (–NH), 1723 (–C=O ester), 1646 (–C=O), 1606 (C=C– aromatic ring), 1215/2595 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 1.26 (3H, *t*, *J* = 7.0 Hz, $CH_2C\mathbf{H}_3$), 3.65, 3.72 (6H, 2 *s*, 2 OCH_3), 4.30 (2H, *q*, *J* = 7.0 Hz, CH_2), 5.31 (1H, *s*, $CHCO_2$), 6.78–7.44 (3H, *m*, aromatic), 10.87 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 376 (M^+, 18), 323, 319, 190, 184, 178, 150, 149 (BP, 100), 122, 75, 65, 50.

5-(1,3-Benzodioxol-5-yl)-7-oxo-2-thioxo-1,2,3,7-tetrahydro[1,2,4]triazolo-[1,5-a]pyrimidine-6-carbonitrile (***6***). Yield: 76 %; brown crystals, m.p.: 270 °C; Anal. Calcd. for $C_{13}H_7N_5O_3S$: C, 49.84; H, 2.25; N, 22.35; S, 10.23 %. Found: C, 50.11; H, 2.39; N, 22.18; S, 10.41 %; IR (KBr, cm^{-1}): 3383/3275 (–NH), 2230 (–C≡N), 1705 (–C=O), 1626 (–C=N), 1605/2565 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 6.07 (2H, *s*, OCH_2O), 7.56–8.20 (3H, *m*, aromatic), 8.68 (1H, *s*, NHC=N, D_2O exchangeable), 12.08 (1H, *s*, NNHC=S, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 313 (M^+, 15), 273, 250, 214, 192, 148 , 76, 56, 41 (BP 100).

3,3′-[(4-Fluorophenyl)methylene]bis[6-(1,3-benzodioxol-5-yl)-4-oxo-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carbonitrile] (***7***). Yield: 73 %; white crystals; m.p.: 128 °C; Anal. Calcd. for $C_{31}H_{17}FN_6O_6S_2$: C, 57.05; H, 2.63; N, 12.88; S, 9.83 %. Found: C, 56.87; H, 2.71; N, 12.48; S, 9.80 %; IR (KBr, cm^{-1}): 3380/3185 (–NH), 2225 (–C≡N), 1658 (–C=O stretching), 1628 (–C=N), 1605 (C=C– aromatic ring); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 5.7 (1H, *s*, CHAr), 6.08 (4H, *s*, 2 OCH_2O), 6.88–7.85 (10H, *m*, aromatic), 12.35 (2H, 2*s*, 2 NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 652 (M^+, 65), 272, 245, 214, 186, 149, 124, 91 (BP, 100) 50.

3,3′-(3,6-Dioxocyclohexa-1,4-diene-1,4-diyl)bis(1,3-benzodioxol-5-yl)-4-oxo-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carbonitrile (***9***). Yield: 68 %; pale brown crystals; m.p.: 216 °C; Anal. Calcd. for $C_{30}H_{14}N_6O_8S_2$: C, 55.38; H, 2.17; N, 12.92; S, 9.86 %. Found: C, 55.53; H, 2.09; N, 13.04; S, 9.98 %; IR (KBr, cm^{-1}): 3235/3165 (–NH), 2216 (–C≡N), 1704 (–C=O quinonoid), 1648 (–C=O amide), 1622 (–C=N), 1595 (C=C– aromatic ring); ^{1}H-NMR (300 MHz, DMSO-

-d_6, δ / ppm): 6.04 (4H, *s*, 2 OCH_2O), 6.18, 6.37 (2H, 2 *s*, 2 CHC=O) 6.82–7.68 (6H, *m*, aromatic), 12.43 (2H, 2 *s*, 2 NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 650 (M^+, 44.12), 604, 217 (BP, 100), 78, 77.

*5-(1,3-Benzodioxol-5-yl)-7-oxo-1,7-dihydrotetrazolo[1,5-*a*]pyrimidine-6-carbonitrile (**10**)*. Yield: 52 %; brown crystals; m.p.: 166 °C; Anal. Calcd. for $C_{12}H_6N_6O_3$: C, 51.07; H, 2.14; N, 29.78 %. Found: C, 50.96; H, 2.15; N, 29.89 %; IR (KBr, cm^{-1}): 3310 (–NH), 2231 (–C≡N), 1695 (–C=O), 1626 (–C=N), 1610 (C=C– aromatic ring), 1582 (–N=N) ; ^{1}H-NMR (300 MHz, DMSO-d_6, δ / / ppm): 6.19 (2H, *s*, OCH_2O), 7.54–7.22 (3H, *m*, aromatic), 12.18 (1H, *s*, NH, D_2O exchangeable); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 151.37 (C_2), 164.41 (C_4), 90.19 (C_5), 171.91 (C_6), 115.23 (CN), 101.59 (OCH_2O), 149.68 (Ar-C), 147.08 (Ar-C), 108.93 (Ar-C), 108.73 (Ar-C), 124.17 (Ar-C), 122.46 (Ar-C); MS (*m*/*z*, (relative abundance, %)): 161 (34.12), 136 (31.76), 77 (30.59), 69 (BP, 100).

*6-(1,3-Benzodioxol-5-yl)-4-chloro-2-thioxo-1,2-dihydropyrimidine-5-carbonitrile (**11a**)*.Yield: 74 %; brown crystals; m.p.: 106 °C; Anal. Calcd. for $C_{12}H_6ClN_3O_2S$: C, 49.41; H, 2.07; Cl, 12.15; N, 14.40; S, 10.99 %. Found: C, 49.50; H, 1.99; Cl, 12.21; N, 14.35; S, 11.05 %; IR (KBr, cm^{-1}): 3144 (–NH), 2217 (–C≡N), 1681 (–C=N), 1185/2630 (–C=S/C–SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 6.10 (2H, *s*, OCH_2O), 6.74–7.33 (3H, *m*, aromatic), 12.4 (1H, *s*, NH, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 276, 275, 274, 257, 216 (BP, 100), 170, 121, 77.

*4-Chloro-6-(3,4-dimethoxyphenyl)-2-thioxo-1,2-dihydropyrimidine-5-carbonitrile (**11b**)*.Yield: 62 %; m.p.: 183 °C; brown crystals; Anal. Calcd. for $C_{13}H_{10}ClN_3O_2S$: C, 50.73; H, 3.28; Cl, 11.52; N, 13.65; S, 10.42 %. Found: C, 50.49; H, 3.79; Cl, 11.21; N, 13.35; S, 11.05 %; IR (KBr, cm^{-1}): 3195 (–NH), 3089 (–CH aromatic), 2993 (–CH aliphatic), 2229 (–C≡N nitrile), 1619 (–C=N), 1539 (C=C– aromatic ring), 1168/2620 (–C=S/C–SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 3.81 (6H, *s*, 2 OCH_3), 6.82–7.24 (3 H, *m*, aromatic), 12.76 (1H, *s*, NH, D_2O exchangeable); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 178.20 (C_2), 162.10 (C_4), 101.37 (C_5),153.80 (C_6), 114.42 (CN), 147.88 (Ar-C), 128.97 (Ar-C), 121.94 (Ar-C), 109.03 (Ar-C), 108.53 (Ar-C), 55.81 (2 OCH_3); MS (*m*/*z*, (relative abundance, %)): 307 (M+, 75.34), 276, 246, 221, 188, 85 (BP, 100), 86.

*4-(1,3-Benzodioxol-5-yl)-6-chloro-2-(methylthio)pyrimidine-5-carbonitrile (**11c**)*. Yield: 64 %; m.p.: 296 °C; brown crystals; Anal. Calcd. for $C_{13}H_8ClN_3O_2S$: C, 51.07; H, 2.64; Cl, 11.60; N, 13.74; S, 10.49 %. Found: C, 51.31; H, 2.68; Cl, 11.09; N, 13.99; S, 10.89 %; IR (KBr, cm^{-1}): 2216 (–C≡N), 1635 (–C=N), 1605 (C=C– aromatic ring); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / / ppm): 172.1 (C_2), 160.0 (C_4), 108.6 (C_5), 158.8 (C_6), 114.8 (CN), 109.2 (**C**CN), 14.4 (SCH_3), 103.3 (OCH_2O), 162.5 (Ar-C), 126.3 (Ar-C), 120.8 (Ar-C), 110.2

(Ar-C), 145.8 (2 Ar-C); MS, (*m*/*z*, (relative abundance, %)): 288 , 287 (BP, 100), 286, 241, 200, 148, 120, 100, 63, 62, 53, 50.

*7-(1,3-Benzodioxol-5-yl)-5-thioxo-5,6-dihydrotetrazolo[1,5-*c*]pyrimidine-8--carbonitrile (**13**)*. Yield: 54 %; yellow crystals; m.p.: 202 °C; Anal. Calcd. for $C_{12}H_6N_6O_2S$: C, 48.32; H, 2.03; N, 28.17; S, 10.75 %. Found: C, 48.29; H, 2.11; N, 28.33; S, 10.73 %; IR (KBr, cm^{-1}): 3148 (–NH), 2213 (–C≡N nitrile), 1642 (–C=N), 1210/2585 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 6.10 (2H, *s*, OCH_2O), 8.24–7.57 (3H, *m*, aromatic), 8.77 (1H, *s*, NH, D_2O exchangeable); ^{13}C-NMR (75 MHz, DMSO, δ / ppm): 158.4 (C_5), 154.1 (C_7), 134.9 (C_9), 106 (C_8), 138.1 (Ar-C), 137.9 (Ar-C), 131.90 (Ar-C), 131.97 (C≡N), 116.9 (Ar-C), 117.1 (Ar-C), 113 (O–CH_2–O); 70.5 (Ar-C); MS (*m*/*z*, (relative abundance, %)): 300 ($[M + 2]^+$, 23), 299 ($[M +1]^+$, 30), 298 (M^+, 72), 272, 244 (BP, 100), 238, 234, 211, 184, 152, 121, 109, 64, 54.

*4-Amino-5-(1,3-benzodioxol-5-yl)pyrimido[4,5-*d*]pyrimidine-2,7(1*H*,3*H*)-di-thione (**15**)*. Yield: 62 %; yellow crystals; m.p.: 244 °C; Anal. Calcd. for $C_{13}H_9N_5O_2S_2$: C, 47.12; H, 2.74; N, 21.13; S, 19.35 %. Found: C, 47.20; H, 2.65; N, 21.18; S, 19.23 %; IR (KBr, cm^{-1}): 3422, 3342 (–NH_2/NH), 1654 (–C=N), 1605 (C=C– aromatic ring), 1595/2615 (–C=S/SH); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 3.40 (2H, *s*, NH_2, D_2O exchangeable), 6.15 (2H, *s*, OCH_2O), 7.33–7.78 (3H, *m*, Ar-H); 12.3 (s, 2H, 2NH, D_2O exchangeable) MS (*m*/*z*, (relative abundance, %)): 331 (M^+, 96.27), 301, 280, 252, 134 (BP, 100), 78.

*5-Amino-4-(1,3-benzodioxol-5-yl)-6-phenyl-1,3-dihydro-2*H*-pyrrolo[2,3-*d*]-pyrimidine-2-thione* (**17a**).Yield: 72 %; brown crystals; m.p.: 208 °C; Anal. Calcd. for $C_{19}H_{14}N_4O_2S$: C, 62.97; H, 3.89; N, 15.46; S, 8.85 %. Found: C, 63.12; H, 3.95; N, 15.43; S, 8.76 %; IR (KBr, cm^{-1}): 3448 (–NH, NH_2), 2895 (–CH aliphatic), 1616 (–C=N); ^{1}H-NMR) (300 MHz, DMSO-d_6, δ / ppm): 1.94 (1H, *s*, NH, D_2O exchangeable), 4.14 (2H, *s*, NH_2, D_2O exchangeable), 6.2 (2H, *s*, OCH_2O), 7.28–7.85 (8H, *m*, aromatic), 8.12 (1H, *s*, NHC=S, D_2O exchangeable); MS (*m*/*z*, (relative abundance, %)): 362 (M^+, 59.78), 318, 241, 112 (BP, 100), 122, 78.

*5-Amino-4-(3,4-dimethoxyphenyl)-6-phenyl-1,3-dihydro-2*H*-pyrrolo[2,3-*d*]-pyrimidine-2-thione (**17b**)*. Yield: 67 %; brown crystals; m.p.: 190 °C; Anal. Calcd. for $C_{20}H_{18}N_4O_2S$: C, 63.47; H, 4.79; N, 14.80; S 8.47 %. Found: C, 63.52; H, 4.89; N, 14.95; S, 8.62 %; IR (KBr, cm^{-1}): 3466 (–NH,NH_2), 3090 (–CH aromatic), 1632 (–C=N); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 2.1 (1H, *s*, NH, D_2O exchangeable), 3.58 (6H, *s*, 2 OCH_3), 4.78 (2H, *s*, NH_2, D_2O exchangeable), 6.78–7.55 (8H, *m*, aromatic), 8.28 (1H, *s*, NHC=S, D_2O exchangeable), ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 174.4 (C_2), 159.8 (C_4), 109.3 (C_5), 161.6 (C_6), 118.7 (**C**=CNH_2), 121.7 (CNH_2), 51.1 (2 OCH_3), 147.7(Ar-C), 147.1 (Ar-C), 135.9 (Ar-C), 126.5 (Ar-C), 126.1 (Ar-C), 125.8 (Ar-C), 124.4

(Ar-C), 123.8 (Ar-C), 119.6 (Ar-C), 119.0 (Ar-C), 115.8 (Ar-C), 111.9 (Ar-C); MS, (*m*/*z*, (relative abundance, %)): 378 (M^+, 42.31), 301, 287,109, 55 (BP, 100).