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Synthesis of 1,5-dihydro-3-methyl-6-trihalomethyl-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones

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N-(Methoxycarbonyl)trihaloacetimidoylchlorides react with 5-aminopyrazoles under mild conditions to give N-methoxycarbonyl-N'-(5-pyrazolyl)trihaloacetamidines, which furnish 6-trihalomethyl-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones by thermal intramolecular cyclocondensation.

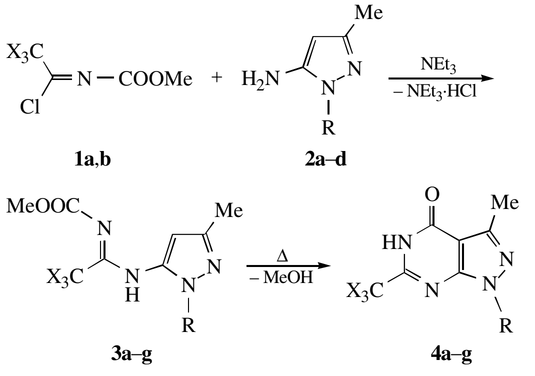
Pyrazolo[3,4-*d*]pyrimidines represent a general class of adenosine receptor antagonists.[1,2] Their effect on adenosine receptor selectivity is determined by the nature of substituents at the atoms N(1), C(4) and C(6).[3–5] The starting substances for the preparation of 4-subsituted 1*H*-pyrazolo[3,4-*d*]pyrimidines[6] are 4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones, which are obtained by the condensation of 5-amino-1*H*-pyrazole-4-carboxamides with formamide[6] or esters.[7] In particular, 4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones containing alkyl, phenyl and alkoxycarbonyl substituents at the 6-position can be synthesised. Here we report a new synthetic approach to 6-trihalomethyl-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones. The introduction of trifluoromethyl groups into the above molecules is expected to enhance their lipophilicity.[8,9] In turn, a trichloromethyl group facilitates the introduction of amino groups at the 6-position by *ipso* substitution.[10]

The method proposed is based on the use of *N*-(methoxycarbonyl)trihaloacetimidoylchlorides **1a**,**b**[11] as new synthetic units, which react with 2-aminopyrazoles **2a**–**d** to form a pyrimidine ring.

We found that compounds **1a**,**b** react with 1-alkyl(aryl, hetaryl)-5-aminopyrazoles **2a**–**d** in benzene at room temperature in the presence of triethylamine to give *N*-methoxycarbonyl-*N'*-(5-pyrazolyl)trihaloacetamidines **3a**–**g** in high yields. The structures of these compounds corroborated by elemental analysis and spectroscopy.[†] ^{1}H NMR signals from 4-H protons of the pyrazole ring observed at 5.90–6.09 ppm for compounds **3a**–**f** (and at 6.68 ppm for compound **3g**) unambiguously confirm the N-acylation of amines **2a**–**d** by imidoylchlorides **1a**,**b**.

On boiling *N*-(methoxycarbonyl)amidines **3a**–**e**,**g** in toluene and amidine **3f** in *o*-xylene for 3 h, they were converted into 3-methyl-6-trihalomethyl-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones **4a**–**g** in satisfactory or high yields.[‡]

The cyclization most likely occurs through the electrophilic attack of the carbonyl group at the π-electron-rich C-4 atom of the pyrazole ring. However, the thermolysis of *N*-(methoxycarbonyl)amidine to isocyanate,[12,13] a more electrophilic system, which in fact attacks the pyrazole nucleus, can also take place.



1a X = F
1b X = Cl

2a R = CH_2CH_2CN
2b R = Ph
2c R = 2-Cl-5-$CF_3C_6H_3$
2d R = 2-pyridyl

3a, **4a** X = F, R = CH_2CH_2CN
3b, **4b** X = Cl, R = CH_2CH_2CN
3c, **4c** X = F, R = Ph
3d, **4d** X = Cl, R = Ph
3e, **4e** X = F, R = 2-Cl-5-$CF_3C_6H_3$
3f, **4f** X = Cl, R = 2-Cl-5-$CF_3C_6H_3$
3g, **4g** X = Cl, 2-pyridyl

Scheme 1

The structure of compounds **4a**–**g** was supported by elemental analysis and IR, ^{1}H, ^{19}F and ^{13}C NMR spectroscopic data and X-ray diffraction analysis, which was carried out for **4g** (Figure 1). The N(1–4)C(1–5) bicyclic system is planar: the atom devia-

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[†] Melting points were determined using a Thomas–Hoover apparatus. The IR spectra were measured in KBr pellets. The ^{1}H, ^{13}C and ^{19}F NMR spectra were measured at 300.0, 75.5, and 282.24 MHz, respectively, in [$^{2}H_6$]DMSO with $SiMe_4$ (^{1}H and ^{13}C) and $CFCl_3$ (^{19}F) internal standards.

The general procedure for the preparation of N-*methoxycarbonyl*-N'-*(5-pyrazolyl)trihaloacetamidines* **3a**–**g**. A solution or suspension of aminopyrazole **2a**–**d** (5 mmol) and triethylamine (5 mmol, 0.69 ml) in benzene (10 ml) was added to a solution of imidochloride **1a**,**b** (5 mmol) in benzene (10 ml) with stirring at room temperature. After additional stirring for 2 h, the mixture was heated to boiling, and the resulting triethylamine hydrochloride precipitate was filtered off. The filtrate was evaporated, and the residue was crystallised from benzene.

N'-*[1-(2-Cyanoethyl)-3-methyl-1*H*-pyrazol-5-yl]*-N-*methoxycarbonyltrifluoroacetamidine* **3a**: yield 85%, mp 91–92 °C. ^{1}H NMR, δ: 2.28 (s, 3H, Me), 2.88 (t, 2H, CH_2, J 6.9 Hz), 3.79 (s, 3H, OMe), 4.59 (t, 2H, CH_2N, J 6.9 Hz), 5.91 (s, 1H, 4-$H_{pyrazole}$), 7.32 (s, 1H, NH). ^{19}F NMR, δ: –71.5 (s, CF_3). IR (ν/cm^{-1}): 3090 (NH), 2260 (CN), 1735 (C=O). Found (%): C, 43.64; H, 4.14; N, 22.78; F, 18.64. Calc. for $C_{11}H_{12}F_3N_5O_2$ (%): C, 43.57; H, 3.99; N, 23.09; F, 18.80.

N'-*[1-(2-Cyanoethyl)-3-methyl-1*H*-pyrazol-5-yl]*-N-*methoxycarbonyltrichloroacetamidine* **3b**: yield 87%, mp 138–139 °C. ^{1}H NMR, δ: 2.27 (s, 3H, Me), 2.91 (t, 2H, CH_2, J 7.2 Hz), 3.74 (s, 3H, OMe), 4.52 (t, 2H, CH_2N, J 7.2 Hz), 5.90 (s, 1H, 4-$H_{pyrazole}$), 7.34 (s, 1H, NH). IR (ν/cm^{-1}): 3075 (NH), 2260 (CN), 1730 (C=O). Found (%): C, 37.70; H, 3.44; N, 19.75; Cl, 30.35. Calc. for $C_{11}H_{12}Cl_3N_5O_2$ (%): C, 37.47; H, 3.43; N, 19.86; Cl, 30.16.

N-*Methoxycarbonyl*-N'-*(3-methyl-1-phenyl-1*H*-pyrazol-5-yl)trifluoroacetamidine* **3c**: yield 96%, mp 111–112 °C. ^{1}H NMR, δ: 2.35 (s, 3H, Me), 3.75 (s, 3H, OMe), 6.01 (s, 1H, 4-$H_{pyrazole}$), 7.31 (t, 1H, 4'-H, J 6.6 Hz), 7.42 (m, 2H, 3'-H, 5'-H), 7.51 (s, 1H, NH), 7.59 (d, 2H, 2'-H, 6'-H, J 7.6 Hz). ^{19}F NMR, δ: –71.9 (s, CF_3). IR (ν/cm^{-1}): 3100 (NH), 1740 (C=O). Found (%): C, 51.79; H, 3.83; N, 17.41; F, 17.56. Calc. for $C_{14}H_{13}F_3N_4O_2$ (%): C, 51.54; H, 4.02; N, 17.17; F, 17.47.

N-*Methoxycarbonyl*-N'-*(3-methyl-1-phenyl-1*H*-pyrazol-5-yl)trichloroacetamidine* **3d**: yield 95%, mp 112–113 °C. ^{1}H NMR, δ: 2.25 (s, 3H, Me), 3.83 (s, 3H, OMe), 5.93 (s, 1H, 4-$H_{pyrazole}$), 6.95 (t, 1H, 4'-H, J 6.8 Hz), 7.12 (m, 2H, 3'-H, 5'-H), 7.56 (s, 1H, NH), 7.90 (d, 2H, 2'-H, 6'-H, J 7.9 Hz). IR (ν/cm^{-1}): 3080 (NH), 1745 (C=O). Found (%): C, 44.38; H, 3.65; N, 15.07; Cl, 21.59. Calc. for $C_{14}H_{13}Cl_3N_4O_2$ (%): C, 44.76; H, 3.49; N, 14.92; Cl, 28.31.

N'-*{1-[2-Chloro-5-(trifuoromethyl)phenyl]-3-methyl-1*H*-pyrazol-5-yl}*-N-*methoxycarbonyltrifluoroacetamidine* **3e**: yield 68%, mp 127–128 °C. ^{1}H NMR, δ: 2.38 (s, 3H, Me), 3.82 (s, 3H, OMe), 6.06 (s, 1H, 4-$H_{pyrazole}$), 7.29 (s, 1H, NH), 7.64 (s, 2H, 3'-H, 4'-H), 7.74 (s, 1H, 6'-H). ^{19}F NMR, δ: –63.9 (s, CF_3), –71.6 (s, CF_3). IR (ν/cm^{-1}): 3100 (NH), 1740 (C=O). Found (%): C, 42.11; H, 2.69; N, 13.32; F, 26.38. Calc. for $C_{15}H_{11}ClF_6N_4O_2$ (%): C, 42.02; H, 2.59; N, 13.07; F, 26.59.

N'-*{1-[2-Chloro-5-(trifuoromethyl)phenyl]-3-methyl-1*H*-pyrazol-5-yl}*-N-*methoxycarbonyltrichloroacetamidine* **3f**: yield 77%, mp 160–161 °C. ^{1}H NMR, δ: 2.38 (s, 3H, Me), 3.77 (s, 3H, OMe), 6.09 (s, 1H, 4-$H_{pyrazole}$), 7.26 (s, 1H, NH), 7.62 (s, 2H, 3'-H, 4'-H), 7.76 (s, 1H, 6'-H). IR (ν/cm^{-1}): 3100 (NH), 1735 (C=O). Found (%): C, 37.71; H, 2.45; N, 11.47; Cl, 29.43. Calc. for $C_{15}H_{11}Cl_4F_3N_4O_2$ (%): C, 37.68; H, 2.32; N, 11.72; Cl, 29.66.

N-*Methoxycarbonyl*-N'-*[3-methyl-1-(2-pyridinyl)-1*H*-pyrazol-5-yl]trichloroacetamidine* **3g**: yield 73%, mp 139–140 °C. ^{1}H NMR, δ: 2.32 (s, 3H, Me), 3.83 (s, 3H, OMe), 6.68 (s, 1H, 4-$H_{pyrazole}$), 7.16 (m, 1H, $H_{pyridine}$), 7.86–8.04 (m, 3H, $H_{pyridine}$), 13.6 (s, 1H, NH). IR (ν/cm^{-1}): 3100 (NH), 1725 (C=O). Found (%): C, 41.69; H, 3.07; N, 18.84; Cl, 28.60. Calc. for $C_{13}H_{12}Cl_3N_5O_2$ (%): C, 41.46; H, 3.21; N, 18.59; Cl, 28.24.

tions from the least-squares plane do not exceed 0.04 Å, the dihedral angle between N(1)N(2)C(1–3) and N(3)N(4)C(3–5) cycles being only 2.9°. The N(5)C(8–12) ring is turned out from the bicyclic plane by 23.0°. In a crystal, molecules of **4g** are joined in dimeric pairs by the intermolecular hydrogen bonds N(1)–H(1)···O(1) [O(1)···N(1) 2.803(3), O(1)···H(1) 1.93(4), N(1)–H(1) 0.88(4) Å, O(1)H(1)N(1) 171(2)°].§

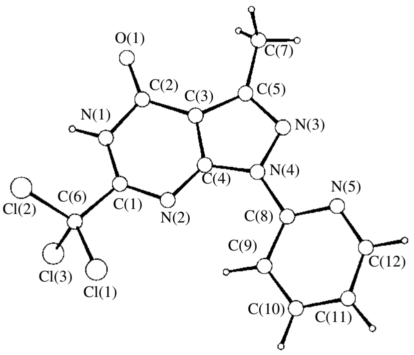

Figure 1 Molecular structure of **4g**. Selected bond lengths (Å): N(1)–C(1) 1.355(3), N(1)–C(2) 1.401(3), N(2)–C(1) 1.290(3), N(2)–C(4) 1.361(3), N(3)–N(4) 1.386(3), N(3)–C(5) 1.317(3), N(4)–C(4) 1.361(3), C(2)–C(3) 1.423(3), C(3)–C(4) 1.417(3), C(3)–C(5) 1.417(3).

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‡ *The general procedure for the preparation of 3-methyl-6-trihalomethyl-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-ones* **4a–g**. Toluene (10 ml) or *o*-xylene (10 ml, for compound **3g**) was added to compound **3a–f** (3 mmol), and the reaction mixture was refluxed for 3 h. The product precipitated on cooling was filtered off and crystallised from acetonitrile (for compounds **4a**, **4b**, **4e**, **4f**) or from an ethanol–DMF mixture (for compounds **4c**, **4d**, **4g**).

*1-(2-Cyanoethyl)-3-methyl-6-trifluoromethyl-1,5-dihydro-4*H*-pyrazolo-[3,4-*d*]pyrimidin-4-one* **4a**: yield 68%, mp 202–203 °C. ^{1}H NMR, δ: 2.50 (s, 3H, Me), 3.12 (t, 2H, CH$_2$, J 6.9 Hz), 4.52 (t, 2H, CH$_2$N, J 6.9 Hz), 13.70 (s, 1H, NH). ^{13}C NMR, δ: 13.11 (Me), 17.73 (CH$_2$), 42.38 (CH$_2$N), 104.03 [C(3a)], 117.84 (q, CF$_3$, $^1J_{\text{C–F}}$ 276.5 Hz), 118.14 (CN), 145.39 [C(3)], 145.84 [C(6), $^2J_{\text{C–F}}$ 37.4 Hz], 150.98 [C(7a)], 158.39 (C=O). ^{19}F NMR, δ: –68.9 (s, CF$_3$). IR (ν/cm^{-1}): 3170, 3080 (NH), 1690 (C=O). Found (%): C, 44.07; H, 3.17; N, 25.90; F, 21.23. Calc. for $C_{10}H_8F_3N_5O$ (%): C, 44.29; H, 2.97; N, 25.82; F, 21.02.

*1-(2-Cyanoethyl)-3-methyl-6-trichloromethyl-1,5-dihydro-4*H*-pyrazolo-[3,4-*d*]pyrimidin-4-one* **4b**: yield 62%, mp 183–184 °C. ^{1}H NMR, δ: 2.49 (s, 3H, Me), 3.16 (t, 2H, CH$_2$, J 7.1 Hz), 4.52 (t, 2H, CH$_2$N, J 7.1 Hz), 13.36 (s, 1H, NH). ^{13}C NMR, δ: 13.12 (Me), 17.55 (CH$_2$), 42.37 (CH$_2$N), 92.01 (CCl$_3$), 102.51 [C(3a)], 118.15 (CN), 145.19 [C(3)], 150.62 [C(7a)], 154.09 [C(6)], 158.60 (C=O). IR (ν/cm^{-1}): 3160, 3060 (NH), 1695 (C=O). Found (%): C, 37.20; H, 2.54; N, 22.09; Cl, 33.03. Calc. for $C_{10}H_8Cl_3N_5O$ (%): C, 37.47; H, 2.52; N, 21.85; Cl, 33.18.

*3-Methyl-1-phenyl-6-trifluoromethyl-1,5-dihydro-4*H*-pyrazolo[3,4-*d*]-pyrimidin-4-one* **4c**: yield 85%, mp 266–267 °C. ^{1}H NMR, δ: 2.57 (s, 3H, Me), 7.41 (t, 1H, 4′-H, J 6.9 Hz), 7.58 (m, 2H, 3′-H, 5′-H), 7.97 (d, 2H, 2′-H, 6′-H, J 7.8 Hz), 13.98 (s, 1H, NH). ^{13}C NMR, δ: 13.06 (Me), 105.71 [C(3a)], 117.74 (q, CF$_3$, $^1J_{\text{C–F}}$ 273.4 Hz), 121.31 [C(2′), C(6′)], 126.89 [C(4′)], 129.01 [C(3′), C(5′)], 137.68 [C(1′)], 145.87 [C(3)], 146.28 [C(6), $^2J_{\text{C–F}}$ 37.3 Hz], 150.43 [C(7a)], 158.70 (C=O). ^{19}F NMR, δ: –69.0 (s, CF$_3$). IR (ν/cm^{-1}): 3180, 3080 (NH), 1700 (C=O). Found (%): C, 53.16; H, 3.36; N, 18.79; F, 19.55. Calc. for $C_{13}H_9F_3N_4O$ (%): C, 53.07; H, 3.08; N, 19.04; F, 19.37.

*3-Methyl-1-phenyl-6-trichloromethyl-1,5-dihydro-4*H*-pyrazolo[3,4-*d*]-pyrimidin-4-one* **4d**: yield 82%, mp 252–253 °C. ^{1}H NMR, δ: 2.58 (s, 3H, Me), 7.40 (t, 1H, 4′-H, J 6.9 Hz), 7.57 (m, 2H, 3′-H, 5′-H), 8.09 (d, 2H, 2′-H, 6′-H, J 7.8 Hz), 14.02 (s, 1H, NH). ^{13}C NMR, δ: 13.22 (Me), 92.35 (CCl$_3$), 104.30 [C(3a)], 120.83 [C(2′), C(6′)], 126.76 [C(4′)], 129.16 [C(3′), C(5′)], 137.88 [C(1′)], 149.9 [C(7a)], 154.70 [C(6)], 159.20 (C=O). IR (ν/cm^{-1}): 3180, 3060 (NH), 1700 (C=O). Found (%): C, 45.36; H, 2.85; N, 16.30; Cl, 31.26. Calc. for $C_{13}H_9Cl_3N_4O$ (%): C, 45.44; H, 2.64; N, 16.31; Cl, 30.95.

*1-[2-Chloro-5-(trifluoromethyl)phenyl]-3-methyl-6-trifluoromethyl-1,5-dihydro-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-one* **4e**: yield 89%, mp 258–259 °C. ^{1}H NMR, δ: 2.59 (s, 3H, Me), 8.04 (s, 2H, 3′-H, 4′-H), 8.14 (s, 1H, 6′-H), 13.73 (s, 1H, NH). ^{13}C NMR, δ: 13.13 (Me), 104.86 [C(3a)], 117.72 (q, CF$_3$, $^1J_{\text{C–F}}$ 276.4 Hz), 123.03 (q, CF$_3$ in the aromatic ring, $^1J_{\text{C–F}}$ 272.5 Hz), 126.97 [q, C(4′), $^3J_{\text{C–F}}$ 4.5 Hz], 127.99 [q, C(6′), $^3J_{\text{C–F}}$ 3.2 Hz], 128.79 [q, C(5′), $^2J_{\text{C–F}}$ 33.4 Hz], 131.68 [C(3′)], 134.94 [C(1′)], 135.24 [C(2′)], 146.83 [C(6), $^2J_{\text{C–F}}$ 36.9 Hz], 146.85 [C(3)], 152.47 [C(7a)], 158.71 (C=O). ^{19}F NMR, δ: –61.5 (s, CF$_3$, in the aromatic ring), –69.0 (s, CF$_3$). IR (ν/cm^{-1}): 3190, 3080 (NH), 1720 (C=O). Found (%): C, 42.13; H, 1.89; N, 14.43; F, 28.97. Calc. for $C_{14}H_7ClF_6N_4O$ (%): C, 42.39; H, 1.78; N, 14.12; F, 28.74.

*1-[2-Chloro-5-(trifluoromethyl)phenyl]-3-methyl-6-trichloromethyl-1,5-dihydro-4*H*-pyrazolo[3,4-*d*]pyrimidin-4-one* **4f**: yield 40%, mp 207–208 °C. ^{1}H NMR, δ: 2.58 (s, 3H, Me), 8.01 (s, 2H, 3′-H, 4′-H), 8.16 (s, 1H, 6′-H), 13.60 (s, 1H, NH). ^{13}C NMR, δ: 13.17 (Me), 92.02 (CCl$_3$), 103.40 [C(3a)], 123.08 (q, CF$_3$ in the aromatic ring, $^1J_{\text{C–F}}$ 272.7 Hz), 126.96 [q, C(4′), $^3J_{\text{C–F}}$ 3.8 Hz], 127.72 [q, C(6′) $^3J_{\text{C–F}}$ 3.2 Hz], 128.71 [q, C(5′), $^2J_{\text{C–F}}$ 33.0 Hz], 131.67 [C(3′)], 134.95 [C(1′)], 135.02 [C(2′)], 146.78 [C(3)], 152.11 [C(7a)], 155.21 [C(6)], 158.95 (C=O). IR (ν/cm^{-1}): 3170, 3050 (NH), 1685 (C=O). Found (%): C, 37.51; H, 1.38; N, 12.71; Cl, 31.52. Calc. for $C_{14}H_7Cl_4F_3N_4O$ (%): C, 37.70; H, 1.58; N, 12.56; Cl, 31.79.

*3-Methyl-1-(2-pyridinyl)-6-trichloromethyl-1,5-dihydro-4*H*-pyrazolo-[3,4-*d*]pyrimidin-4-one* **4g**: yield 64%, mp 241–242 °C. ^{1}H NMR, δ: 2.58 (s, 3H, Me), 7.49 (m, 1H, H_{pyridine}), 8.06 (m, 2H, H_{pyridine}), 8.03 (s, 1H, NH), 8.60 (m, 1H, H_{pyridine}). ^{13}C NMR, δ: 13.22 (Me), 92.43 (CCl$_3$), 104.38 [C(3a)], 117.27 [C(3′)], 123.04 [C(5′)], 138.78 [C(4′)], 146.21 [C(3)], 148.74 [C(6′)], 149.54 [C(2′)], 150.82 [C(7a)], 155.01 [C(6)], 159.11 (C=O). IR (ν/cm^{-1}): 3160, 3070 (NH), 1670 (C=O). Found (%): C, 41.95; H, 2.38; N, 20.03; Cl, 31.09. Calc. for $C_{12}H_8Cl_3N_5O$ (%): C, 41.83; H, 2.34; N, 20.32; Cl, 30.87.

§ *Crystal data for* **4g**: $C_{12}H_8Cl_3N_5O$, M = 344.59, monoclinic, space group $P2_1/c$ (no. 14), a = 10.738(1) Å, b = 12.553(1) Å, c = 10.818(1) Å, β = = 93.06(1)°, V = 1456.1(4) Å^3, Z = 4, d_{calc} = 1.572 g cm^{-3}, μ = 58.86 cm^{-1}, F(000) = 702, crystal size of 0.53×0.53×056 mm. All data were collected using CuKα radiation (λ = 1.54178 Å) on an Enraf-Nonius CAD4 diffractometer, $\theta_{\max}$ = 70°, 293 K, 3056 reflections collected (2755 independent, R_{int} = 0.034). An empirical absorption correction based on azimuthal scan data[14] was applied. The structure was solved by direct methods and refined by a full-matrix least-squares technique in the anisotropic approximation using the CRYSTALS[15] program package. All hydrogen atoms were located in the different Fourier maps and included in the final refinement with fixed positional and thermal parameters [only the H(1) atom was refined isotropically]. Convergence was obtained at R = 0.046 and R_w = 0.050, GOF = 1.121 [2147 reflections with $I > 3\sigma(I)$, 194 refined parameters; obs/variabl. 11.1, difference electron density 0.40 and –0.46 e Å^{-3}, Chebyshev weighting scheme[16] with parameters of 12.5, –8.04, 15.0, –3.3 and 4.4]. Atomic coordinates, bond lengths, bond angles and thermal parameters have been deposited at the Cambridge Crystallographic Data Centre (CCDC). For details, see ‘Notice to Authors’, *Mendeleev Commun.*, Issue 1, 2001. Any request to the CCDC for data should quote the full literature citation and the reference number 1135/95.