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A systematic investigation of the reactivity of 2,4,6-trichloropyrimidine with phenoxide nucleophiles has been conducted. Conditions have been described which lead to mono-, di-, and trisubstituted-pyrimidines. Unexpected product distributions for mono- and disubstituted products were observed and explanations for these results are offered.

We have begun to explore the reactivity of 2,4,6-trichloropyrimidine 1 with a variety of nucleophiles with the goal of developing multisubstituted pyrimidines in which different substituents could be introduced sequentially. There is a significant body of literature that describes the reaction of 1 with neutral nucleophiles such as ammonia [1], methylamine [2], dimethylamine [3], and benzylamine [4] among others. Our initial report in this area described the reaction of 1 with ethanolamine and diethanolamine [5].

Replacement of the chlorine atoms by oxygen nucleophiles, however, is less extensive. Reactions involving 1 and a variety of alkoxides [6-8] proceeds quite readily.

We describe here our systematic investigations involving a variety of 4-substituted phenols under considerably milder conditions. A preliminary report included one example from this study [11].

Treatment of equimolar amounts of 1 with various 4-substituted phenols 2 in the presence of an equivalent amount of sodium hydroxide at room temperature afforded very good to excellent yields of monosubstituted products 3 and 4, see Figure 1. While the formation of isomeric monosubstituted products was expected, the ratio of 3 to 4 was most surprising. Ratios ranging from 8:2 to 9:1 were initially isolated from the reaction. This assessment of the two isomers was based on ^{1}H nmr analysis of the mixture. Until



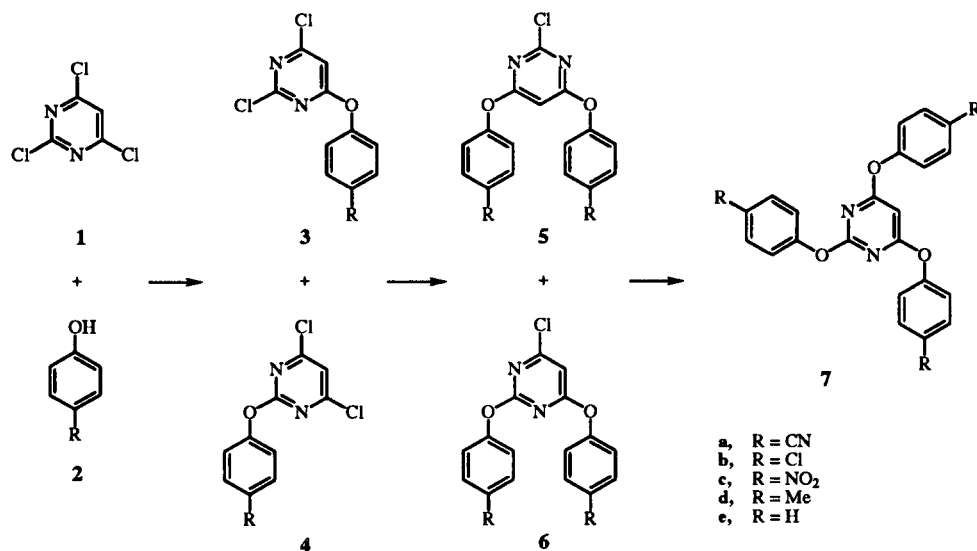a, R = CN
b, R = Cl
c, R = NO$_2$
d, R = Me
e, R = H

Figure 1

Since a suggested order of reactivity of nucleophiles places aryloxides considerably below alkoxides [9], we believed some selectivity could be achieved with substituted phenoxides as nucleophiles. Only one example involving phenols has been described [10]. In that case all three chlorine atoms of 1 were replaced by heating several phenols with potassium hydroxide at 215° for twenty minutes resulting in a 90% yield of the corresponding triaryloxypyrimidines.

now the ratio of 4-substituted-pyrimidines to 2-substituted-pyrimidines arising from 1 has usually been in the order of 2:1 or 3:1 [2,3,5]. These previous observations have led to the assumption that there is little difference in reactivity for the C-Cl bond in either position 2 or 4. Statistically, a ratio of 2:1 should occur if both positions are equally reactive. However, these earlier reports all dealt with neutral nucleophiles. In the present circumstance the anionic nucleophile definitely favors substitution at the C-Cl bond of position 4.

One tempting explanation for these observations is found in a consideration of the mechanistic pathway. Most substitution reactions of this type follow a $S_NAr$ (addition-elimination) process. However, it has been postulated that the intermediate Meisenheimer complex involving a *para*-quinoid structure is more favored than the corresponding *ortho*-quinoid structure [12-15], which is illustrated in Figure 2. Providing some support for this explanation is the observation that in the monochloropyridine series replacement of the chlorine by methoxide is in the order 4-Cl >> 2-Cl > 3-Cl. It is postulated that the difference between 4-Cl and 2-Cl is attributable to the Meisenheimer intermediate where the ring nitrogen *para* to the tetrahedral carbon can bear the negative charge better than the *ortho* nitrogen [12,13]. There is parallel behavior with 4-Cl and 2-Cl pyrimidines [14]. In a study of thiophenoxide with tetrafluorophthalonitrile, reaction in methanol leads to an 8:1 ratio of two products. The major product is obtained by replacement of the two fluorine atoms *para* to the cyano groups. The minor product is derived from replacement of all four fluorine atoms. The formation of the major product is attributable to a *para*-quinoid intermediate [15].
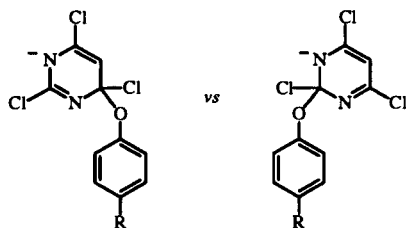


Figure 2

Purification of the crude product by recrystallization afforded only pure 3. The assignment of structure 3 is based on elemental analysis, mass spectral data, $^1H$ nmr, and $^{13}C$ nmr. The $^1H$ nmr spectrum is particularly useful for structural assignment because it displays a signal for the C-5 proton that is distinct from the aromatic ring protons. Furthermore, this signal is highly influenced by the electronic effects of neighboring groups. Thus, chlorine atoms in both positions 4 and 6 place the C-5 proton signal further downfield than the corresponding C-5 proton signal when the neighboring atoms are chlorine and oxygen.

When 1 was allowed to react with two equivalents of the phenols 2 in the presence of an excess of sodium carbonate, good yields of a mixture of isomeric disubstituted products 5 and 6 were obtained. The ratio of the two disubstituted products is approximately 1:1. This observation was even more unexpected than the results obtained during monosubstitution since the formation of 5 was not anticipated. To our knowledge no 4,6-disubstituted products

have ever arisen from similar reactions of 1 with other nucleophiles.

Compound 5 can only arise from 3 while 6 can come from either 3 or 4. Assuming that the ratio of ~9:1 for the 3/4 mixture obtained during the disubstitution process the rate of formation of 6 from 4 must be faster than either of the other rates, namely $3 \rightarrow 5$ and $3 \rightarrow 6$. It seems less probable that very much of 6 could be obtained from 3 due to the failure of position 2 to participate during the monosubstitution process.

It is possible that the argument offered to explain the preponderant formation of 3 over 4, namely enhanced stability of a *para*-quinoid intermediate, is appropriate here as well. Since the formation only of 5 would have been predicted using the *para*-substitution explanation, these results suggest a very strong competing factor that would decrease the yield of 5 by half.

We also considered that an alternative mechanism such as $S_{RN}1$ could be involved [16]. This process operates through a radical ion intermediate that would not necessarily follow the pattern exhibited by $S_NAr$ processes in the past. To test this hypothesis, 1 was treated with 2b under the same conditions except that radical scavengers were added. No effect on either the ratio of 5 to 6 or the overall yield was observed which eliminated this pathway as a viable explanation.

Again, purification of the mixture was accomplished by recrystallization which yielded only 5 in pure form. Characterization by elemental analysis, mass spectra, $^1H$ nmr, and $^{13}C$ nmr led to this assignment of the structure. Similar observations of the $^1H$ nmr signal for the C-5 protons flanked by heteroatoms contributed to this assignment.

In order to ascertain less vigorous conditions sufficient to achieve trisubstitution as reported earlier [10], we subjected 1 to treatment with 2 as well as unsubstituted phenol. The triaryloxypyrimidines 7 were obtained in high yields upon treatment of 1 with three equivalents of 2 and three equivalents of sodium hydroxide in dimethyl sulfoxide at 50°. Under these same conditions we also obtained the previously reported triphenoxypyrimidine 7e in 92% yield. Even though 7b-e were previously described [10] we subjected these compounds to full characterization, in part because of differences between the literature melting points and those we obtained. Spectral data and elemental analysis supported these structural assignments.

## EXPERIMENTAL

with tetramethylsilane as the internal standard. Relative integrals of peak areas are in agreement with the assigned structures. Ultraviolet spectra were recorded on a Cary 1 UV-Visible spectrophotometer. Mass spectra were measured on a Hewlett Packard 5995A GC/MS instrument using a direct insertion probe. Elemental analyses were performed by Galbraith Laboratories, Knoxville, TN.

**2,6-Dichloro-4-(4'-cyanophenoxy)pyrimidine (3a).**

2,4,6-Trichloropyrimidine (0.92 g, 5.0 mmoles) was dissolved in 10 ml of acetone and the solution cooled to 0° and stirred. Meanwhile a solution of 4-cyanophenoxide was prepared by dissolving 4-cyanophenol (0.595 g, 5.0 mmoles) and sodium hydroxide (0.205 g, 5.1 mmoles) in water (10 ml). The aqueous phenoxide solution was added slowly to the acetone solution. A white precipitate formed rapidly and the mixture was slowly warmed to room temperature and stirred for an additional 3 hours. After dilution with water (20 ml) the crude product was filtered to give 1.2 g (90%). The crude product contained the other isomer, 4,6-dichloro-2-(4'-cyanophenoxy)pyrimidine **4a**, as a minor component in a ratio of 90:10.

This was recrystallized from methanol/ether to give pure **3a**, mp 136-138°; ir (potassium bromide): ν 3113, 2220 (CN), 1547, 1493, 1402, 1338, 1270, 1195, 1109, 852, 543 cm$^{-1}$; uv (methanol): λ max 207 nm (ε 22,300), 231 nm (ε 20,750), 256 nm (ε 11,000); $^1$H nmr (dimethyl-d$_6$ sulfoxide): d 7.54 (d, 2H, J = 8.4 Hz, aromatic), 7.62 (s, 1H, H-5), 8.00 (d, 2H, J = 8.4 Hz, aromatic); $^{13}$C (dimethyl-d$_6$ sulfoxide): d 171.0, 162.8, 158.9, 155.8, 135.4, 123.6, 119.1, 110.0, 108.6; ms: m/z 267 (21), 265 (35), 230 (100), 163 (25).

*Anal.* Calcd. For C$_{11}$H$_5$N$_3$Cl$_2$O: C, 49.62; H, 1.88; N, 15.79. Found: C, 49.60; H, 1.82; N, 15.61.

**2,6-Dichloro-4-(4'-chlorophenoxy)pyrimidine (3b).**

This compound was prepared using the procedure described for **3a**. The crude product was obtained in 85% yield and contained the other isomer, 4,6-dichloro-2-(4'-chlorophenoxy)-pyrimidine **4b**, as a minor component in a ratio of 80:20.

This was recrystallized from hexane to give pure **3b**, mp 78-80°; ir (potassium bromide): ν 3100, 1547, 1477, 1402, 1338, 1258, 1189, 1103, 970, 847 cm$^{-1}$; uv (methanol): λ max 205 nm (ε 20,400), 258 nm (ε 8,100); $^1$H nmr (deuteriochloroform): δ 6.86 (s, 1H, H-5), 7.10 (d, 2H, J = 9.0 Hz, aromatic), 7.40 (m, 2H, J = 8.7 Hz, aromatic); $^{13}$C (deuteriochloroform): δ 163.2, 160.4, 150.4, 132.3, 130.4, 123.2, 106.6; ms: m/z 276 (27), 239 (100), 241 (70), 172 (34), 111 (29).

*Anal.* Calcd. For C$_{10}$H$_5$N$_2$Cl$_3$O: C, 43.56; H, 1.81; N, 10.16. Found: C, 43.73; H, 1.92; N, 10.30.

**2,6-Dichloro-4-(4'-nitrophenoxy)pyrimidine (3c).**

This compound was prepared using the procedure described for **3a**. The crude product was obtained in 68% yield and contained the other isomer, 4,6-dichloro-2-(4'-nitrophenoxy)pyrimidine **4c**, as a minor component in a ratio of 85:15.

This was recrystallized from chloroform/hexane to give pure **3c**, mp 124-128°; ir (potassium bromide): ν 3091, 1552, 1488, 1397, 1199, 1109, 980, 852 cm$^{-1}$; uv (methanol): λ max 204 nm (ε 26,500), 266 nm (ε 15,300); $^1$H nmr (deuteriochloroform): δ 7.01 (s, 1H, H-5), 7.35 (d, 2H, J = 9.3 Hz, aromatic), 8.30 (d, 2H, J = 9.3 Hz, aromatic); $^{13}$C (deuteriochloroform): δ 170.1, 163.7, 160.3, 156.5, 146.1, 126.0, 122.6, 107.3; ms: m/z 287 (28), 285 (40), 250 (100), 204 (19), 183 (12), 137 (3)

*Anal.* Calcd. For C$_{10}$H$_5$N$_3$Cl$_2$O$_3$: C, 41.96; H, 1.75; N, 14.69. Found: C, 41.86; H, 1.71; N, 14.56.

**2,6-Dichloro-4-(4'-methylphenoxy)pyrimidine (3d).**

This compound was prepared using the procedure described for **3a**. The crude product was obtained in 85% yield and contained the other isomer, 4,6-dichloro-2-(4'-methylphenoxy)-pyrimidine **4d**, as a minor component in a ratio of 85:15.

This was recrystallized from methanol to give pure **3d**, mp 49-52°; ir (potassium bromide): ν 3100, 1547, 1498, 1403, 1258, 1183, 1103, 980, 847 cm$^{-1}$; uv (methanol): λ max 206 nm (ε 27,200), 251 nm (ε 12,000); $^1$H nmr (deuteriochloroform): δ 2.38 (s, 3H, -CH$_3$ ), 6.75 (s, 1H, H-5), 7.02 (d, 2H, J = 8.4 Hz, aromatic), 7.23 (d, 2H, J = 8.4 Hz, aromatic); $^{13}$C (deuteriochloroform): δ 171.6, 162.9, 160.3, 136.6, 130.9, 121.3, 106.2, 21.2; ms: m/z 256 (30), 254 (38), 221 (60), 219 (100), 183 (2), 152 (42), 91 (51).

*Anal.* Calcd. For C$_{11}$H$_8$N$_2$Cl$_2$O: C, 51.76; H, 3.14; N, 10.98. Found: C, 51.51; H, 3.14; N, 10.81.

**2-Chloro-4,6-di-(4'-cyanophenoxy)pyrimidine (5a).**

2,4,6-Trichloropyrimidine (0.92 g, 5.0 mmoles), 4-cyanophenol (1.19 g, 10.0 mmoles), and sodium carbonate (1.59 g, 15.0 mmoles) were taken up in ethanol (20 ml) and the mixture heated to reflux, with stirring, for 3 hours. After cooling to room temperature the mixture was diluted with water (100 ml) and the solid filtered, washed with water, and dried in air to give 1.57 g (90%) of a mixture of disubstituted products **5a** and **6a** in a ratio of 55:45.

This was recrystallized from acetonitrile to give pure **5a**, mp 250-252°; ir (potassium bromide): ν 3070, 2215 (CN), 1579, 1493, 1413, 1263, 1210, 1012, 938, 836, 537 cm$^{-1}$; uv (methanol): λ max 210 nm (ε 15,500), 234 nm (ε 19,800), 251 nm (ε 14,400); $^1$H nmr (dimethyl-d$_6$ sulfoxide): δ 6.96 (s, 1H, H-5), 7.49 (d, 4H, J = 8.4 Hz, aromatic), 7.98 (d, 4H, J = 8.4 Hz, aromatic); $^{13}$C (dimethyl-d$_6$ sulfoxide): δ 171.8, 158.8, 156.3, 135.3, 123.3, 118.9, 109.8, 94.1; ms: m/z 348 (23), 313 (78), 246 (22), 218 (29), 163 (97), 102 (100).

*Anal.* Calcd. For C$_{18}$H$_9$N$_4$ClO$_2$: C, 61.98; H, 2.58; N, 16.07. Found: C, 61.48; H, 2.54; N, 15.77

**2-Chloro-4,6-di-(4'-chlorophenoxy)pyrimidine (5b).**

This compound was prepared using the procedure described for **5a**. The crude product, 83%, consisted in a mixture of **5b** and **6b** in a ratio of 50:50.

This was recrystallized from hexane to give pure **5b**, mp 160-161°; ir (potassium bromide): ν 3120, 1611, 1578, 1482, 1413, 1263, 1210, 1087, 1012, 943, 825, 670 cm$^{-1}$; uv (methanol): λ max 204 nm (ε 23,400), 219 nm (ε 20,000), 249 nm (ε 8,400); $^1$H nmr (deuteriochloroform): δ 6.21 (s, 1H, H-5), 7.10 (d, 4H, J = 8.7 Hz, aromatic), 7.40 (d, 4H, J = 8.7 Hz, aromatic); $^{13}$C (deuteriochloroform): δ 172.8, 160.5, 151.8, 132.2, 130.0, 124.2, 91.5; ms: m/z 368 (27), 333 (36), 331 (66), 263 (19), 236 (12), 172 (63), 111 (100).

*Anal.* Calcd. For C$_{16}$H$_9$N$_2$Cl$_3$O$_2$•0.25H$_2$O: C, 51.64; H, 2.57; N, 7.53. Found: C, 51.72; H, 2.32; N, 7.46.

**2-Chloro-4,6-di-(4'-nitrophenoxy)pyrimidine (5c).**

This compound was prepared using the procedure described for **5a**. The crude product, 76%, consisted in a mixture of **5c** and **6c** in a ratio of 50:50.

This was recrystallized from chloroform to give pure **5c**, mp 248-52°; ir (potassium bromide): ν 3102, 1568, 1520, 1488, 1413, 1349, 1258, 1205, 852 cm$^{-1}$; uv (methanol): λ max 204 nm (ε 41,000), 274 nm (ε 26,500), 249 nm (ε 8,400); $^1$H nmr (dimethyl-d$_6$ sulfoxide): δ 7.05 (s, 1H, H-5), 7.55 (d, 4H, J = 8.7 Hz, aromatic), 8.35 (d, 4H, J = 8.7 Hz, aromatic); $^{13}$C (dimethyl-d$_6$ sulfoxide): δ 171.8, 159.2, 157.6, 145.9, 126.6, 123.3, 94.4; ms: m/z 388 (48), 360 (100), 353 (796), 258 (18), 183 (70).

*Anal.* Calcd. For C$_{16}$H$_9$N$_4$ClO$_6$•0.25H$_2$O : C, 48.87; H, 2.43; N, 14.25. Found: C, 48.81; H, 2.46; N, 14.48.

2-Chloro-4,6-di-(4'-methylphenoxy)pyrimidine (**5d**).

This compound was prepared using the procedure described for **5a**. The crude product, 87%, consisted in a mixture of **5d** and **6d** in a ratio of 55:45.

This was recrystallized from hexane to give pure **5d**, mp 105-108°; ir (potassium bromide): ν 3020-2920, 1616, 1584, 1552, 1498, 1413, 1263, 1210, 1016, 979, 940, 815 cm$^{-1}$; uv (methanol): λ max 209 nm (ε 16,700), 243 nm (ε 9,050); $^1$H nmr (deuteriochloroform): δ 2.45 (s, 6H, -CH$_3$), 6.05 (s, 1H, H-5), 7.05 (d, 4H, J = 8.7 Hz, aromatic), 7.21 (d, 4H, J = 9.0 Hz, aromatic); $^{13}$C (deuteriochloroform): δ 172.8, 160.1, 150.4, 136.2, 130.8, 121.3, 90.2, 21.3; ms: m/z 326 (36), 298 (80), 291 (100), 223 (24), 152 (44), 91 (80).

*Anal.* Calcd. For C$_{18}$H$_{15}$N$_2$ClO$_2$•0.25H$_2$O : C, 65.26; H, 4.72; N, 8.46. Found: C, 65.35; H, 4.55; N, 8.37.

2,4,6-Tri-(4'-cyanophenoxy)pyrimidine (**7a**).

2,4,6-Trichloropyrimidine (0.92 g, 5.0 mmoles), 4-cyanophenol (1.85 g, 15.5 mmoles), and powdered sodium hydroxide (0.62 g, 15.5 mmoles) were taken up in dimethyl sulfoxide (20 ml) and the mixture heated at ~50°, with stirring, for 4 hours. After cooling to room temperature the mixture was diluted with water (100 ml) and the solid filtered, washed with water, and dried in air to give 88% of crude **7a**.

This was recrystallized from tetrahydrofuran/hexane to give pure **7a**, mp 202-205°; ir (potassium bromide): ν 3090, 2226 (CN), 1579, 1498, 1429, 1360, 1221, 1066, 1023, 836, 542 cm$^{-1}$; uv (methanol): λ max 206 nm (ε 25,900), 233 nm (ε 21,900), 257 nm (ε 16,000); $^1$H nmr (dimethyl-d$_6$ sulfoxide): δ 6.65 (s, 1H, H-5), 7.35 (d, 2H, J = 8.4 Hz, aromatic), 7.45 (d, 4H, J = 8.7 Hz, aromatic), 7.8 (d, 2H, J = 8.7 Hz, aromatic), 8.0 (d, 4H, J = 8.7 Hz, aromatic); $^{13}$C (dimethyl-d$_6$ sulfoxide): δ 172.5, 163.4, 156.3, 156.2, 135.0, 134.6, 123.5, 119.1, 109.4, 108.9, 89.7; ms: m/z 431 (12), 313 (100), 246 (8), 213 (28), 169 (36), 102 (30).

*Anal.* Calcd. For C$_{25}$H$_{13}$N$_5$O$_3$•0.25H$_2$O: C, 68.89; H, 3.02; N, 16.07. Found: C, 68.54; H, 2.97; N, 15.82

2,4,6-Tri-(4'-chlorophenoxy)pyrimidine (**7b**).

This compound was prepared using the procedure described for **7a**. The crude product was obtained in 91% yield.

This was recrystallized from chloroform/hexane to give pure **7b**, mp 124-126° (lit.107° [10]); ir (potassium bromide): ν 3080, 1600, 1573, 1482, 1424, 1349, 1215, 1087, 1010, 836, 804, 660 cm$^{-1}$; uv (methanol): λ max 205 nm (ε 23,500), 221 nm (ε 21,800), 253 nm (ε 11,000); $^1$H nmr (deuteriochloroform): δ 6.00 (s, 1H, H-5), 6.95-7.00 (m, 6H, aromatic), 7.20-7.40 (m, 6H, aromatic); $^{13}$C (deuteriochloroform): δ 173.1, 164.4, 151.3, 130.0, 129.5, 123.3, 86.9; ms: m/z 460 (19), 331 (100), 305 (8), 222 (7), 178 (20), 111 (18).

*Anal.* Calcd. For C$_{22}$H$_{13}$N$_2$Cl$_3$O$_3$: C, 57.45; H, 2.83; N, 6.09. Found: C, 57.42; H, 2.92; N, 6.49.

2,4,6-Tri-(4'-nitrophenoxy)pyrimidine (**7c**).

This compound was prepared using the procedure described for **7a**. The crude product was obtained in 82% yield.

This was recrystallized from chloroform/hexane to give pure **7c**, mp 134-136°; ir (potassium bromide): ν 3091, 1568, 1520, 1482, 1434, 1328, 1221, 1109, 852 cm$^{-1}$; uv (methanol): λ max 206 nm (ε 38,200), 274 nm (ε 26,500); $^1$H nmr (deuteriochloroform): δ 6.3 (s, 1H, H-5), 7.20 (d, 2H, J = 8.7, aromatic), 7.30 (d, 4H, J = 9.0, aromatic), 8.10 (d, 2H, J = 9.3, aromatic), 8.30 (d, 4H, J = 8.7, aromatic); $^{13}$C (deuteriochloroform): δ 172.4, 163.4, 157.2, 145.7, 145.4, 125.8, 125.4, 122.6, 89.2; ms: m/z 491 (20), 353 (100), 307 (22), 233 (60), 171 (80).

*Anal.* Calcd. for C$_{22}$H$_{13}$N$_5$O$_9$: C, 53.77; H, 2.65; N, 14.26. Found: C, 53.76; H, 2.75; N, 14.40.

2,4,6-Tri-(4'-methylphenoxy)pyrimidine (**7d**).

This compound was prepared using the procedure described for **7a**. The crude product was obtained in 85% yield.

This was recrystallized from chloroform/methanol to give pure **7d**, mp 125-127° (lit 118° [10]); ir (potassium bromide): ν 3027-2920, 1584, 1504, 1424, 1360, 1215, 1066, 1039, 1012, 809 cm$^{-1}$; uv (methanol): λ max 206 nm (ε 41,000), 253 nm (ε 14,600); $^1$H nmr (deuteriochloroform): δ 2.34 (s, 3H, -CH$_3$), 2.37 (s, 6H, -CH$_3$), 5.8 (s, 1H, H-5), 7.0-7.20 (m, 12H, aromatic); $^{13}$C (deuteriochloroform): δ 173.6, 165.0, 150.6, 135.6, 134.9, 130.5, 130.0, 121.5, 85.8, 21.2; ms: m/z 398 (12), 291 (100), 197 (7), 158 (10), 135 (5), 91 (7).

*Anal.* Calcd. for C$_{25}$H$_{22}$N$_2$O$_3$: C, 75.38; H, 5.53; N, 7.04. Found: C, 75.16; H, 5.68; N, 7.41.

2,4,6-Tri-(phenoxy)pyrimidine (**7e**).

This compound was prepared using the procedure described for **7a**. The crude product was obtained in 92% yield.

This was recrystallized from methanol to give pure **7e**, mp 164-165° (lit 156° [10]); ir (potassium bromide): ν 3059, 1605, 1573, 1488, 1429, 1354, 1205, 1066, 1010, 810, 761, 667 cm$^{-1}$; uv (methanol): λ max 207 nm (ε 33,600), 253 nm (ε 14,000); $^1$H nmr (deuteriochloroform): δ 5.85 (s, 1H, H-5), 7.0-7.40 (m, 15H, aromatic); $^{13}$C (deuteriochloroform): δ 173.5, 164.9, 152.8, 130.1, 129.6, 126.1, 125.5, 122.1, 121.8, 86.4 ppm; ms:m/z 356 (19), 279 (3), 263 (100), 237 (6), 169 (8), 77 (20).

*Anal.* Calcd. for C$_{22}$H$_{16}$N$_2$O$_3$: C, 74.14; H, 4.53; N, 7.86. Found: C, 73.83; H, 4.57; N, 7.77.

Acknowledgement.

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