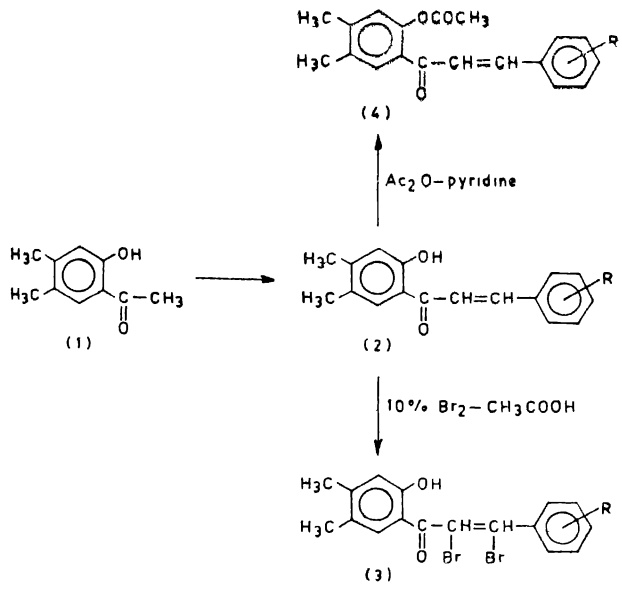

The chalcones were brominated using 10% of bromine in acetic acid to yield α,β-dibromochalcones. The acetyl derivatives of chalcones were prepared using chalcone, acetic anhydride and pyridine. Melting points are uncorrected.

TABLE 1—PHYSICAL DATA OF COMPOUNDS*

Compd. no.	R	Mol. formula	M p ** °C
2a	2-Cl	$C_{17}H_{15}O_2Cl$	94[a]
b	4-Cl	$C_{17}H_{15}O_2Cl$	110[a]
c	2,4-Cl_2	$C_{17}H_{14}O_2Cl_2$	126[a]
d	4-CH_3	$C_{18}H_{18}O_2$	81–82[b]
e	2-OCH_3	$C_{18}H_{18}O_3$	88[c]
f	3,4-$(OCH_3)_2$	$C_{19}H_{20}O_4$	104[c]
g	3,4,5-$(OCH_3)_3$	$C_{20}H_{22}O_5$	116[c]
3a	2-Cl	$C_{17}H_{15}O_2ClBr_2$	109[a]
b	4-Cl	$C_{17}H_{15}O_2ClBr_2$	118[a]
c	2,4-Cl_2	$C_{17}H_{14}O_2Cl_2Br_2$	124[b]
d	4-CH_3	$C_{18}H_{18}O_2Br_2$	138[a]
e	2-OCH_3	$C_{18}H_{18}O_3Br_2$	71[a]
f	3,4-$(OCH_3)_2$	$C_{19}H_{20}O_4Br_2$	78[b]
g	3,4,5-$(OCH_3)_3$	$C_{20}H_{22}O_5Br_2$	146–48[a]
4a	2-Cl	$C_{19}H_{17}O_3Cl$	92[b]
b	4-Cl	$C_{19}H_{17}O_3Cl$	96[b]
c	2,4-Cl_2	$C_{19}H_{16}O_3Cl_2$	98[b]
d	4-CH_3	$C_{20}H_{20}O_3$	72[a]
e	2-OCH_3	$C_{20}H_{20}O_4$	106[d]
f	3,4-$(OCH_3)_2$	$C_{21}H_{22}O_5$	98[a]
g	3,4,5-$(OCH_3)_3$	$C_{22}H_{24}O_6$	106[a]

*All compounds gave satisfactory C, H and halogen analyses

**Solvent for crystallisation : [a]C_6H_6, [b]absolute EtOH, [c]glacial AcOH, [d]MeOH

The paper disc method was used to test antibacterial activity of the chalcones against *Staphylococcus aureus* and *Escherichia coli*. The inocula were obtained from agar solidified nutrient broth media and the growth was checked at 37° after 24 h and the zone of inhibition was measured.

As compared to chloramphenicol and 4,4′-sulphonyldianiline, the chalcones (**2a**–**f**) were found much less active against the representative strains of organisms ; **2g** was found comparatively most active.

Acknowledgement

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Syntheses of some 2-(2′-Methoxyphenyl)-chromones

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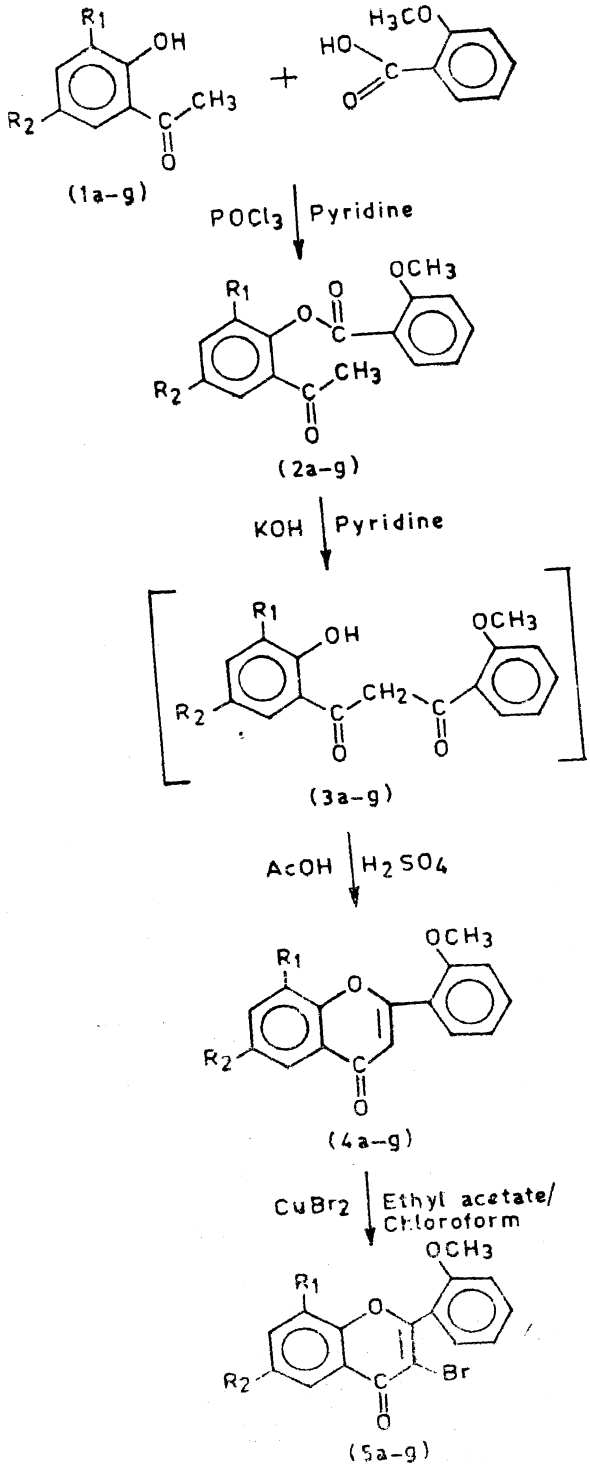
IN continuation of our investigation on chromones[1] we report here the synthesis of some 2-(2′-methoxyphenyl)chromones. These chromones have been obtained (Scheme 1) from the *o*-hydroxyacetophenones (**1a**–**g**).

The *o*-hydroxyacetophenone were converted to the respective *o*-benzoyloxy derivatives (**2a**–**g**) in good yield, by direct union of **1a**–**g** with *o*-methoxybenzoic acid using $POCl_3$ as condensing agent[2]. At higher temperature the yield decreased sharply. Baker-Venkataraman rearrangement of the esters (**2a**–**g**) gave the yellow β-diketones, but isolation of the diketones **3a**–**g** could not be achieved as acidification with ice and dilute H_2SO_4 gave the chromones directly.

The structures of all the chromones were confirmed by elemental analyses and spectral data. An additional experimental confirmation was also achieved. A number of chromones, had earlier been brominated in our laboratory[3] with the help of copper(II) bromide and the corresponding 3-bromo-



Scheme 1

a ; R_1=H, R_2=CH_3 **b** ; R_1=Br, R_2=CH_3
c ; R_1=H, R_2=Cl **d** ; R_1=Br, R_2=Cl
e ; R_1=CH_3, R_2=Br **f** ; R_1=H, R_2=Br
g ; R_1=R_2=Br

chromones were obtained. In the present cases too, the 3-bomochromones (**5a**–**g**), except **5c** and **5f** were prepared in the same manner. In the pmr spectra of these bromo compounds, the diagonistic peak of the C-3 proton was absent. In fact, lhe region δ 6.0–7.0 was blank, thus lending an additional support to the chromone structure.

Experimental

The melting points of all the compounds were recorded in open capillary and are uncorrected. Infrared spectra (nujol/KBr) were recorded on a Backman spectrometer and pmr spectra ($CDCl_3$) were recorded on a Perkin-Elmer spectrometer (90 MHz).

2'-Methoxybenzoyloxy acetophenones (**2a**–**g**) : A mixture of hydroxyacetophenone (0.0065 mol) and *o*-methoxybenzoic acid (0.007 mol) dissolved in pyridine (5–10 ml) was treated with $POCl_3$ (0.3 ml) with stirring and cooling (0–5°). The solids obtained on acidification, were washed successively with water, dilute sodium carbonate solution, dilute NaOH solution and water and recrystallised from suitable solvents. The compounds were crystallised from MeOH, except **2d** which was from EtOH. All the compounds analysed correctly for carbon and hydrogen : **a** (yield 60%), m.p. 48–49° ; **b** (87%), 71–72° ; **c** (87%), 51–52° ; **d** (84%), 84–85° ; **e** (85%), 81–82° ; **f** (88%), 65–67 ; **g** (94%), 108–09°.

2-(2'-Methoxyphenyl)chromones (**4a**–**g**) : A solution of the ester (**2** ; 0.01 mol) in dry pyridine was stirred for 2 h with powdered KOH (0.03 mol) at 60° and then cooled and decomposed with ice and sulphuric acid solution. To maximise the yield, the crude products were refluxed in glacial acetic acid solution for 1 h with catalytic amount of concentrated HCl and worked up. All the chromones (Table 1) gave correct elemental analysis.

TABLE 1—PHYSICAL DATA OF COMPOUNDS **4** AND **5**

Compd. no.	Yield. %	M.p. °C	ν_{max} (KBr) cm^{-1} C=O	C=C	C–O–C
4a	80	58	1 650	1 570	1 235
b	90	97	1 660	1 580	1 240
c	90	60–61	1 640	1 600	1 230
d	90	108	1 655	1 610	1 240
e	90	77–78	1 645	1 580	1 235
f	84	65–66	1 650	1 610	1 240
g	95	116–17	1 660	1 590	1 235
5a	60	54[a]	1 665	1 610	1 220
b	65	93[a]	1 660	1 580	1 240
d	60	103[b]	1 650	1 605	1 240
e	62	71–72[b]	—	—	—
g	70	112[a]	1 655	1 590	1 238

Pmr : **4a** δ 2.68 (3H, s, CH_3), 3.4 (3H, s, OCH_3), 6.6 (1H, s, C-3), 7.2–8.6 (m, ArH) ; **4d** δ 3.2 (3H, s, OCH_3), 6.8 (1H, s, C-3), 7.2–8.6 (m, ArH) ; **4e** δ 2.7 (3H, s, CH_3), 3.45 (3H, s, OCH_3), 6.8 (1H, s, C-3), 7.2–8.6 (m, ArH) ; **5b** δ 2.6 (3H, s, CH_3), 3.42 (3H, s, OCH_3), 7.2–8.6 (m, ArH) ; **5e** δ 2.7 (3H, s, CH_3), 3.45 (3H, s, OCH_3), 7.2–8.4 (m, ArH).

*Crystallisation solvent : [a]EtOH, [b]MeOH.

3-Bromo-2-(2′-methoxyphenyl)chromones (**5a–g**) : Cupric bromide (3 mol) was dissolved in refluxing chloroform-ethyl acetate mixture (1 : 2). The chromone (**4a–g**) (**1** mol) was then added to it and the reflux continued for 8–12 h. The grey white precipitate of copper(I) bromide was filtered and the filterate evaporated to dryness and the solid residue was crystallised from suitable solvents. All the compounds (Table 1) gave satisfactory elemental analyses.

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Synthesis of some Tetrahydropyrimidines

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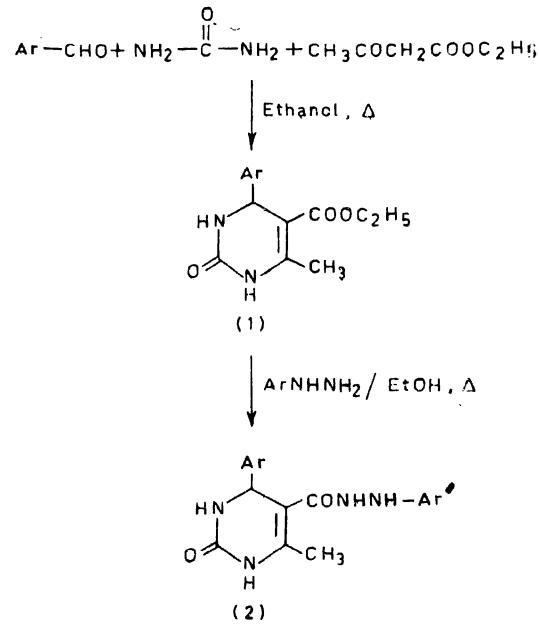
Hydropyrimidines[1] and hydrazine derivatives[2] are reported to possess diverse biological and pharmacological properties. Keeping this in view, we have undertaken the synthesis and antimicrobial activities of some 4-aryl-5-(arylhydrazinocarbamoyl)-6-methyl-1, 2, 3, 4-tetrahydropyrimidin-2-ones (Scheme 1).

Aromatic aldehydes undergo Biginati's reaction to give pyrimidine derivatives[3], which react with various hydrazine derivatives to form the titled compounds (**2**). Aldehydes used were 5-bromosalicyldehyde, 5-bromo- and 5-iodovaniline.

In the ir spectra of compounds **2**, the disappearance of the peak at 1 720 cm^{-1} (C=O ester) and the appearance of a peak at 1 640 cm^{-1} (C=O amide) proved the conversion of **1** to **2**.

Experimental

All melting points are uncorrected. The purity of the compounds was checked by tlc. The ir



1a ; Ar=2-OH-5-Br.C_6H_3
1b ; Ar=4-OH-5-I-3-OCH_3.C_6H_2
1c ; Ar=4-OH-5-Br-3-OCH_3.C_6H_2

Compounds **2** :

2a–e ; Ar=2-OH-5-Br.C_6H_3
2f–j ; Ar=4-OH-5-I-3-OCH_3.C_6H_2
2k–o ; Ar=4-OH-5-Br-3-OCH_3.C_6H_2

2a, f, k ; Ar′=H **2d, i, n** ; Ar′=COC_6H_5
2b, g, l ; Ar′=C_6H_5 **2e, j, o** ; Ar′=COC_6H_5-NH_2-4
2c, h, m ; Ar′=2,4-$(NO_2)_2$.C_6H_3.

Scheme 1

spectra (KBr) were recorded on a Perkin-Elmer spectrophotometer.

2-Oxo-6-methyl-5-carbethoxy-4-aryl-1,2,3,4-tetrahydropyrimidines (**1**). *General procedure :* A mixture of the aldehyde (0.5 mol), urea (0.5 mol) and ethyl acetoacetate (0.75 mol) in absolute ethanol (200 ml) and concentrated HCl (a few drops) was refluxed for 3 h. It was then chilled and the product was filtered. The filtrate was refluxed for 1.5 h and the solvent partially distilled off and then cooled to yield solid product. The solid was then recrystallised from ethanol (Table 1).

4-Aryl-5-(arylhydrazinocarbamoyl)-6-methyl-1,2,3,4-tetrahydropyrimidin-2-ones (**2**) : A mixture of compound **1** (0.01 mol) and various hydrazine derivatives (0.01 mol) was refluxed in ethanol (30 ml) for 3 h. The excess solvent was then distilled off and the resulting solid was dried and recrystallised from ethanol (Table 1) : ν_{max} (KBr) 3 300 (NH), 1 690 (C=O cyclic), 1 640 (C=O non-cyclic), 1 320 (C–N bend) and 1 490 cm^{-1} (C=C bend).

Results and Discussion

In the antibacterial screening, out of 15 compounds tested, 8 compounds (**2a, e, f, i, k, l, n, o**)