Conversion of 3-Hydroxy-1,2,3,4-tetrahydroquinoline-2,4-diones to 2,3a,4,5-Tetrahydrofuro[2,3-*c*]quinoline-2,4-diones *via* an Intramolecular Wittig Reaction

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Dedicated to Professor Thomas Kappe on the occasion of his 65th birthday

The preparation of 2,3a,4,5-tetrahydrofuro[2,3-*c*]quinoline-2,4-diones starting from 3-hydroxy-1,2,3,4-tetrahydroquinoline-2,4-diones and using the reaction path *via* bromoacetylderivatives and triphenylphosphonioacetyl derivatives of the initial substances is described. The nmr spectra of the products are discussed.

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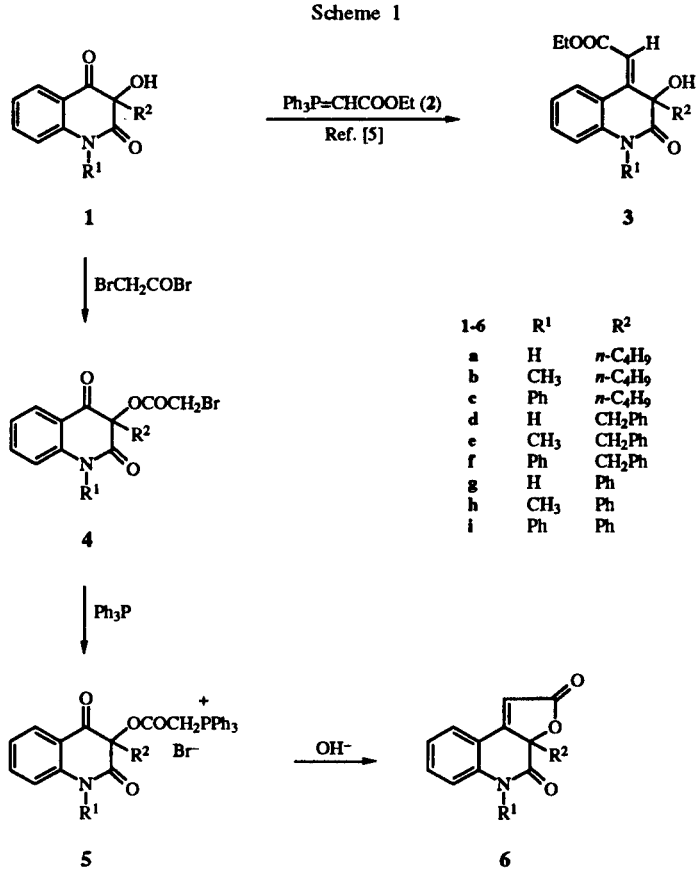
Many antiinflammatory agents are based on the structure of arylacetic and arylpropionic acids (including heteroaryl derivatives) and γ-lactones attached to an aromatic ring system exhibit especially high activity [1]. Generally, many compounds containing the 2-buten-4-olide system display appreciable bioactivity in different areas [2]. In the 2-quinolone series, some γ- and δ-lactones were prepared [3] from esters arising from the Wittig reaction of quinisatine and its *N*-methyl derivative with ethyl (triphenylphosphoranylidene)acetate (**2**).

It is known [4] that α-hydroxyketones react with **2** to give *E*-4-hydroxy-2-alkenoates. In our previous paper [5], we have described the reaction of the stabilized Wittig reagent **2** with 3-hydroxy-1,2,3,4-tetrahydroquinoline-2,4-diones **1**, known as metabolites of some *Pseudomonas* species [6,7]. This reaction proceeds stereoselectively to give *E*-ethoxycarbonylmethylene derivatives **3** and only a small quantity of 2,3a,4,5-tetrahydrofuro[2,3-*c*]quinoline-2,4-diones **6**. We found [5] that esters **3** are not changed to butenolides **6** under given reaction conditions; but, a rapid conversion of **3** to **6** takes place under irradiation with uv light or under the action of different bases and acids [8]. In some instances, we were not able to isolate the butenolides **6** due to their small quantity in the reaction mixture. Therefore, we decided ourselves to prepare these compounds by another independent way.

In this paper, we would like to describe the preparation of butenolides **6** starting from 3-hydroxy-1,2,3,4-tetrahydroquinoline-2,4-diones **1** [5] and using a known [9] path *via* triphenylphosphonioacetylderivatives of α-hydroxyketones (Scheme 1). Reactions of **1** with bromoacetylbromide and one equivalent of pyridine in benzene give esters **4** in nearly quantitative yields. In the ir spectra of esters **4**, characteristic absorption bands occured at 1659-1680 cm^{-1} (lactam) and 1704-1764 cm^{-1} (ester). In the ^1H nmr spectra of these compounds, the well distinguishable ABCD system of aromatic protons is present. The remarkable upfield shift of the H-8 proton

Scheme 1



signal was observed in the case of derivatives **4c**, **4f**, and **4i**, which is caused by circular current of π-electrons of the *N*-phenyl group [5].

Reactions of **4** with triphenylphosphane were carried out in a benzene solution to give triphenylphosphonioacetyl derivatives **5** (in yields from 50 to 90%). It is noteworthy that compounds **5** were not completely pure and contained, according to tlc and nmr, a variable quantity of butenolides **6**. All attempts to crystallize the salts **5**

Table 1

Physical and Analytical Data of Compounds **4-6**

Compound No	R^1	R^2	Method Yield (%)	Mp (°C) (Solvent)	Formula M.W.	Analysis (%) Calcd./Found C	H	N
4a	H	n-C_4H_9	85	131-132 (benzene)	$C_{15}H_{16}BrNO_4$ 354.2	50.86 50.72	4.55 4.72	3.95 3.92
4b	CH_3	n-C_4H_9	98	95-96 (c-C_6H_{12})	$C_{16}H_{18}BrNO_4$ 368.2	52.19 51.87	4.92 4.98	3.80 3.76
4c	C_6H_5	n-C_4H_9	94	60-63 (i-PrOH)	$C_{21}H_{20}BrNO_4$ 430.3	58.62 58.70	4.68 4.62	3.26 3.26
4d	H	CH_2Ph	81	178-180 (ethanol)	$C_{18}H_{14}BrNO_4$ 388.2	55.69 55.58	3.63 3.46	3.61 3.55
4e	CH_3	CH_2Ph	96	136-137 (ethanol)	$C_{19}H_{16}BrNO_4$ 402.2	56.73 56.52	4.01 4.06	3.48 3.42
4f	Ph	CH_2Ph	91	159-161 (benzene/ c-C_6H_{12})	$C_{24}H_{18}BrNO_4$ 464.3	62.08 61.93	3.91 3.75	3.02 2.97
4g	H	Ph	81	192-195 (ethanol)	$C_{17}H_{12}BrNO_4$ 374.2	54.57 54.68	3.23 3.14	3.74 3.69
4h	CH_3	Ph	61	157-160 (benzene)	$C_{18}H_{14}BrNO_4$ 388.2	55.69 55.61	3.63 3.72	3.61 3.59
4i	Ph	Ph	98	188-190 (MeOH)	$C_{23}H_{16}BrNO_4$ 450.3	61.35 61.62	3.58 3.51	3.11 2.90
5a	H	n-C_4H_9	90	146-156 dec	$C_{33}H_{31}BrNO_4P$ 616.5	64.29 63.92	5.07 5.26	2.37 2.17
5b	CH_3	n-C_4H_9	73	168-174 dec	$C_{34}H_{33}BrNO_4P$ 630.5	64.78 64.57	5.28 5.42	2.22 2.05
5c	Ph	n-C_4H_9	34	135-140 dec	$C_{39}H_{35}BrNO_4P$ .2 H_2O 728.6	64.29 64.18	5.39 5.13	1.92 1.58
5d	H	CH_2Ph	96	165-168 dec	$C_{36}H_{29}BrNO_4P$ 650.5	66.47 66.21	4.49 4.57	2.15 2.02
5e	CH_3	CH_2Ph	70	156-160 dec	$C_{37}H_{31}BrNO_4P$ 664.5	66.88 66.42	4.70 4.92	2.11 1.96
5f	Ph	CH_2Ph	61	125-135 dec	$C_{42}H_{33}BrNO_4P$ .2 H_2O 762.6	66.15 66.50	4.89 4.68	1.84 1.85
5g	H	Ph	33	130-135 dec	$C_{35}H_{27}BrNO_4P$ .3 H_2O 690.5	60.88 61.09	4.82 4.58	2.03 1.75
5h	CH_3	Ph	28	204-205 dec	$C_{36}H_{29}BrNO_4P$ 650.5	66.47 66.20	4.49 4.72	2.15 1.83
5i	Ph	Ph	61	136-142 dec	$C_{41}H_{31}BrNO_4P$ .H_2O 730.6	67.40 67.70	4.55 4.69	1.92 1.83
6a	H	n-C_4H_9	A: 81	226-228 (ethanol)	$C_{15}H_{15}NO_3$ 257.3	70.02 70.13	5.88 5.93	5.44 5.23
6b	CH_3	n-C_4H_9	A: 70	206-209 (benzene)	$C_{16}H_{17}NO_3$ 271.3	70.83 70.68	6.32 6.48	5.16 5.07
6c	Ph	n-C_4H_9	A: 57	213-216 (benzene)	$C_{21}H_{19}NO_3$ 333.4	75.66 75.49	5.74 6.04	4.20 3.91
6d	H	CH_2Ph	A: 79	293-297 (AcOH)	$C_{18}H_{13}NO_3$ 291.3	74.22 74.51	4.50 4.48	4.81 4.70
6e	CH_3	CH_2Ph	A: 46	140-142 (MeOH)	$C_{19}H_{15}NO_3$ 305.3	74.75 74.57	4.95 4.79	4.59 4.48
6f	Ph	CH_2Ph	A: 70	246-250 (benzene)	$C_{24}H_{17}NO_3$ 367.4	78.46 78.37	4.66 4.77	3.81 3.20
6g	H	Ph	B: 29	208-213	$C_{17}H_{11}NO_3$ 277.3	73.64 73.43	4.00 4.20	5.05 4.80
6h	CH_3	Ph	A: 87	216-218 (MeOH)	$C_{18}H_{13}NO_3$ 291.3	74.22 73.98	4.50 4.74	4.81 4.72
6i	Ph	Ph	A: 71	227-228 (MeOH)	$C_{23}H_{15}NO_3$ 353.4	78.17 78.02	4.28 4.37	3.96 3.92

Table 2
Spectroscopic Data of Compounds **4-6**

Compound No	IR (cm^{-1})	^{1}H-NMR δ (ppm)
4a	3210-3320, 3026, 2955, 1765, 1704, 1682, 1663, 1614, 1484, 1372, 1148	0.87 (t, J = 7.3 Hz, 3H, CH_3), 1.25-1.32 (m, 2H, H-3 of butyl), 1.41-1.47 (m, 2H, H-2 of butyl), 1.98-2.11 (m, 2H, H-1 of butyl), 4.04 (s, 2H, CH_2Br), 7.02 (d, J = 8.0 Hz, 1H, H-8), 7.16 (t, J = 7.6 Hz, 1H, H-6), 7.57 (dt, J = 7.6 and 1.0 Hz, 1 H, H-7), 7.93 (d, J = 7.6 Hz, 1H, H-5), 9.63 (s, 1H, NH)
4b	2600-3100, 1764, 1707, 1676, 1602, 1473, 1357, 1298, 1172, 1151, 1112, 1088	0.84 (t, J = 7.3 Hz, 3H, CH_3 of butyl), 1.23-1.28 (m, 2H, H-3 of butyl), 1.35-1.39 (m, 2H, H-2 of butyl), 1.93-2.02 (m, 2H, H-1 of butyl), 3.48 (s, 3H, N-CH_3), 4.02 (s, 2H, CH_2Br), 7.17-7.22 (m, 2H, H-6 and H-8), 7.66 (t, J = 7.7 Hz, 1H, H-7), 8.00 (d, J = 7.7 Hz, 1H, H-5)
4c	2960, 1754, 1713, 1680, 1600, 1490, 1463, 1384, 1367, 1350, 1300, 1289, 1273, 1166, 1154, 1068, 945, 777, 768, 710	0.89 (t, J = 7.2 Hz, 3H, CH_3), 1.27-1.41 (m, 2H, H-3 of butyl), 1.43-1.53 (m, 2H, H-2 of butyl), 2.05-2.24 (m, 2H, H-1 of butyl), 4.01 (s, 2H, CH_2Br), 6.49 (d, J = 8.2 Hz, 1H, H-8), 7.16 (dt, J = 7.5 and 0.9 Hz, 1H, H-6), 7.25-7.60 (m, 6H, H-7 and C_6H_5), 8.03 (dd, J = 7.8 and1.6 Hz, 1 H, H-5)
4d	3275, 3005, 2955, 1764, 1695, 1674, 1659, 1612, 1592, 1483, 1440, 1395, 1374, 1301, 1285, 1232, 1168, 1149, 1130, 1060, 1040, 943, 888, 783, 705	3.39 (s, 2H, CH_2Ph), 4.03 (s, 2H, CH_2Br), 6.70 (d, J = 7.8 Hz, 1H, H-8), 7.05-7.13 (m, 6H, H-6 and C_6H_5), 7.44 (dt, J = 7.7 and 1.5 Hz, 1H, H-7), 7.85 (dd, J = 7.8 and 1.4 Hz, 1 H, H-5), 8.68 (s, 1 H, NH)
4e	3010, 2960, 1757, 1713, 1682, 1606, 1474, 1360, 1300, 1171, 1152, 1078, 773, 702	3.31 (s, 3H, CH_3), 3.39 (s, 2H, CH_2Ph), 4.03 (s, 2H, CH_2Br), 6.83 (d, J = 8.4 Hz, 1 H, H-8), 6.93-7.15 (m, 6 H, H-6 and C_6H_5), 7.47 (t, J = 7.2 Hz, 1 H, H-7), 7.90 (d, J = 7.7 Hz, 1 H, H-5)
4f	3062, 3010, 1739, 1708, 1672, 1599, 1490, 1465, 1358, 1300, 1281, 1268, 1221, 1167, 1129, 1119, 1046, 950, 778, 708	3.52 (s, 2H, CH_2Ph), 4.04 (s, 2H, CH_2Br), 6.19 (d, J = 8.3 Hz, 1 H, H-8), 7.00-7.55 (m, 12 H, H-6, H-7 and two C_6H_5), 7.95 (dd, J = 7.8 and 1.7 Hz, 1 H, H-5)
4g	3279, 3005, 2960, 1755, 1704, 1680, 1662, 1618, 1599, 1488, 1451, 1402, 1292, 1231, 1165, 1139, 999, 940, 771, 750, 701	4.11 (s, 2 H, CH_2Br), 6.97 (d, J = 7.9 Hz, 1 H, H-8), 7.16 (dt, J = 7.6 and 0.9 Hz, 1 H, H-6), 7.32-7.58 (m, 6 H, H-7 and C_6H_5), 7.93 (dd, J = 7.9 and 1.3 Hz, 1 H, H-5), 8.88 (s, 1 H, NH)
4h	3002, 2946, 1751, 1707, 1673, 1599, 1492, 1473, 1300, 1249, 1142, 1130, 1062, 1008, 778, 754, 701	3.57 (s, 3H, CH_3), 4.07 and 4.10 (two doublets, J = 12.0 Hz, 2 H, CH_2Br), 7.15-7.45 (m, 7 H, H-6, H-8 and C_6H_5), 7.65 (dt, J = 7.9 and 1.6 Hz, 1 H, H-7), 7.98 (dd, J = 7.9 and 1.5 Hz, 1 H, H-5)
4i	3022, 3001, 1731, 1709, 1678, 1599, 1490, 1461, 1339, 1292, 1260, 1221, 1132, 1112, 1008, 937, 780, 772, 757, 707	4.06 and 4.11 (two doublets, J = 13.1 Hz, 2 H, CH_2Br), 6.51 (d, J = 8.3 Hz, 1 H, H-8), 7.14 (dt, J = 7.7 and 0.7 Hz, 1 H, H-6), 7.25-7.63 (m, 11H, H-7 and two C_6H_5), 8.01 (dd, J = 7.7 and 1.5 Hz, 1 H, H-5)
5a	3420, 2972, 2915, 1744, 1713, 1680, 1612, 1483, 1438, 1158, 1111, 750, 720, 688	0.63 (t, J = 6.8 Hz, 3H, CH_3), 0.92-1.05 (m, 4 H, H-2 and H-3 of butyl), 1.52-1.72 (m, 2H, H-1 of butyl), 5.15-5.25 (m, 2H, CH_2P), 6.98-7.03 (m, 2 H, H-6 and H-8), 7.50 (t, J = 7.5 Hz, 1 H, H-7), 7.60-7.85 (m, 16 H, H-5 and three C_6H_5), 10.90 (s, 1 H, NH)
5b	1748, 1708, 1670, 1601, 1472, 1439, 1362, 1338, 1302, 1176, 1110, 1082, 752, 690	0.75 (t, J = 6.8 Hz, 3 H, CH_3 of butyl), 1.04-1.18 (m, 4 H, H-2 and H-3 of butyl), 1.65-1.81 (m, 2 H, H-1 of butyl), 3.36 (s, 3 H, N-CH_3), 5.34 (d, J = 14.5 Hz, 2 H, CH_2P), 7.17 (t, J = 7.5 Hz, 1 H, H-6), 7.19 (d, J = 8.3 Hz, 1 H, H-8), 7.67 (dt, J = 7.9 and 1.5 Hz, 1 H, H-7, 7.68-7.94 (m, 16 H, H-5 and three C_6H_5)
5c	2997, 2901, 1708, 1680, 1649, 1569, 1457, 1435, 1412, 1355, 1315, 1268, 1082, 723, 662	0.82 (t, J = 6.9 Hz, 3 H, CH_3), 1.15-1.31 (m, 4 H, H-2 and H-3 of butyl), 1.87-2.00 (m, 2 H, H-1 of butyl), 5.19-5.24 and 5.27-5.46 (two m, 2H, CH_2P), 6.42 (d, J = 8.3 Hz, 1 H, H-8), 7.14 (t, J = 7.3 Hz, 1 H, H-6), 7.40 (dt, J = 7.8 and 1.5 Hz, 1 H, H-7), 7.15-7.95 (m, 20 H, four C_6H_5), 7.92 (dd, J = 7.3 and 1.5 Hz, 1 H, H-5)
5d	3400-3600, 3061, 2900, 1751, 1716, 1689, 1612, 1588, 1482, 1441, 1355, 1305, 1140, 1116, 940, 760, 728, 693	2.88 (s, 2 H, CH_2Ph), 5.34 (d, J = 15.0 Hz, 2 H, CH_2P), 6.94 (t, J = 7.8 Hz, 1 H, H-6), 7.02 (d, J = 7.6 Hz, 1 H, H-8), 7.02-7.85 (m, 21 H, H-5 and four C_6H_5), 7.40 (dt, J = 7.8 and 1.5 Hz, 1 H, H-7), 10.84 (s, 1 H, NH)
5e	2957, 1758, 1732, 1679, 1623, 1598, 1507, 1482, 1450, 1379, 1352, 1280, 1245, 1108, 757, 725	3.05-3.08 (m, 2 H, CH_2Ph), 3.18 (s, 3 H, CH_3), 5.48 (d, J = 14.4 Hz, 2H, CH_2P), 6.80-6.83 (m, 2 H, H-6 and H-8), 7.00-7.92 (m, 21 H, H-5 and four C_6H_5), 7.47 (t, J = 7.2 Hz, 1 H, H-7)
5f	3057, 2920, 1739, 1709, 1676, 1600, 1492, 1464, 1439, 1358, 1301, 1273, 1112, 752, 708, 694	3.18 and 3.27 (two d, J = 12.8 Hz, 2 H, CH_2Ph), 5.27-5.38 and 5.56-5.69 (two m, 2 H, CH_2P), 6.08 (d, J = 8.3 Hz, 1 H, H-8), 6.8-7.93 (m, 28 H, H-5, H-6, H-7 and five C_6H_5)
5g	3300-3550, 3051, 3008, 2900, 2848, 1768, 1739, 1717, 1682, 1612, 1589, 1483, 1441, 1352, 1117, 1000, 751, 725, 693	5.00-5.20 and 5.20-5.38 (two m, 2H, CH_2P), 7.12 (t, J = 7.5 Hz, 1 H, H-6), 7.22 (d, J = 7.5 Hz, 1 H, H-8), 7.45 (t, J = 7.7 Hz, 1 H, H-7), 7.02-7.96 (m, 21 H, H-5 and four C_6H_5), 11.18 (s, 1 H, NH)
5h	1748, 1713, 1670, 1600, 1470, 1439, 1362, 1336, 1324, 1160, 1110, 773, 753, 693, 683	3.47 (s, 3 H, CH_3), 5.30-5.60 (m, 2 H, CH_2P), 7.14-8.08 (m, 24 H, H-5, H-6, H-7, H-8 and four C_6H_5)

Table 2 (continued)

Compound No	IR (cm^{-1})	^{1}H-NMR δ (ppm)
5i	3055, 1740, 1713, 1681, 1645, 1600, 1495, 1456, 1446, 1342, 1300, 1112, 1001, 751, 735, 696	5.25-5.58 (m, 2 H, CH_2P), 6.42 (d, J = 8.3 Hz, 1 H, H-8), 7.11 (t, J = 7.2 Hz, 1 H, H-6), 7.25-7.95 (m, 27 H, H-5, H-7 and five C_6H_5)
6a	[5]	0.83 (t, J = 7.2 Hz, 3 H, CH_3), 1.22-1.35 (m, 4 H, H-2 and H-3 of butyl, 1.74-1.76 and 2.16-2.18 (two m, 2H, H-1 of butyl), 6.11 (s, 1 H, H-1), 7.08 (d, 1 H, H-6), 7.20 (t, J = 7.6, 1 H, H-8), 7.46 (dt, J = 8.0 and 1.2 Hz, 1 H, H-7), 7.56 (d, J = 7.6 Hz, 1 H, H-9), 8.97 (s, 1 H, NH); In hexadeuteriodimethyl sulfoxide: [5]
6b	[5]	[5]
6c	[5]	0.86 (t, J = 7.2 Hz, 3 H, CH_3), 1.25-1.33 (m, 2 H, H-3 of butyl), 1.33-1.42 (m, 2 H, H-2 of butyl), 1.81-1.88 and 2.25-2.32 (two m, 2 H, H-1 of butyl), 6.16 (s, 1 H, H-1), 6.47 (d, J = 8.3 Hz, 1 H, H-6), 7.18 (t, J = 7.5 Hz, 1 H, H-8), 7.31 (dt, J = 7.9 and 1.4 Hz, 1 H, H-7), 7.46-7.58 (m, 5 H, C_6H_5), 7.60 (dd, 7 = 7.6 and 1.2 Hz, 1 H, H-9); In hexadeuteriodimethyl sulfoxide: [5]
6d	[5]	3.06 and 3.52 (two d, J = 13.7 Hz, 2 H, CH_2), 5.83 (s, 1 H, H-1), 6.98 (d, J = 8.2 Hz, 1 H, H-6), 6.98 (t, J = 7.6 Hz, 1 H, H-8), 7.20-7.28 (m, 6 H, H-7 and C_6H_5), 7.49 (d, J = 7.6 Hz, 1 H, H-9), 7.75 (brs, 1 H, NH); In hexadeuteriodimethyl sulfoxide: [5]
6e	1763, 1686, 1641, 1601, 1469, 1456, 1360, 1338, 1283, 1167, 1079, 931, 765, 702	3.01 and 3.42 (two d, J = 13.8 Hz, 2 H, CH_2), 3.44 (s, 3 H, CH_3), 5.80 (s, 1 H, H-1), 6.92-6.94 (m, 2 H, H-6 and H-8), 7.12-7.27 (m, 5 H, C_6H_5), 7.47 (dd, J = 7.5 and 1.2 Hz, 1 H, H-9), 7.56 (dt, J = 7.9 and 1.2 Hz, 1 H, H-7)
6f	[5]	[5]; In hexadeuteriodimethyl sulfoxide: 3.31 (s, 2 H, CH_2), 6.40 (s, 1 H, H-1), 6.39 (d, J = 8.9 Hz, 1 H, H-6), 6.93-7.66 (m, 12 H, H-7, H-8 and two C_6H_5), 7.72 (dd, J = 7.5 and 1.4 Hz, 1 H, H-9)
6g	3490, 3226, 3101, 2924, 1752, 1702, 1634, 1610, 1586, 1479, 1450, 1435, 1346, 1201, 1179, 1121, 1068, 966, 905, 780, 736, 697, 645	6.29 (s, 1 H, H-1), 6.97 (d, J = 8.0 Hz, 1 H, H-6), 7.19 (dt, J = 7.5 and 1.2 Hz, 1 H, H-8), 7.25-7.72 (m, 7 H, H-7, H-9 and C_6H_5), 8.75 (s, 1 H, NH)
6h	1749, 1700, 1644, 1603, 1471, 1402, 1357, 1327, 1206, 846, 773, 736	In hexadeuteriodimethyl sulfoxide: 3.40 (s, 3 H, CH_3), 6.77 (s, 1 H, H-1), 7.20-7.43 (m, 7 H, H-6, H-8 and C_6H_5), 7.55 (t, J = 7.9 Hz, 1 H, H-7), 7.82 (d, J = 7.8 Hz, 1 H, H-9)
6i	[5]	6.36 (d, J = 8.1 Hz, 1 H, H-6), 6.37 (s, 1 H, H-1), 7.10-7.52 (m, 12 H, H-7, H-8 and two C_6H_5), 7.68 (dd, J = 7.5 and 1.2 Hz, 1 H, H-9)

failed as well as attempts of crude **5** purification by extraction with hot benzene, because even more butenolide **6** is formed under these conditions by decomposition of **5**. Some compounds **5** were amorphous and compounds **5c**, **5f**, **5g** and **5i** were identified by elemental analyses and nmr spectra as hydrates even after drying them to constant weight over phosphorus pentoxide *in vacuo* at room temperature. Because of the large number of aromatic protons in the molecules of triphenylphosphonium salts, the interpretation of their ^{1}H-nmr spectra is troublesome in some cases. But, generally it can be said that the signal positions of the aromatic protons are in accord with those of esters **4**. The signals of CH_2P protons exhibit complex splitting in most cases.

Cyclization of triphenylphosphonioacetyl derivatives **5** to butenolides **6** proceeds smoothly and was attained, in yields 60 to 90%, by shaking a chloroform solution of **5** with 0.5 *M* sodium hydroxide at room temperature for 10 minutes or by reaction of a chloroform solution of **5** with triethylamine. Compound **5** is very easily cyclized to **6** in alkaline media, thus the occurence of butenolides **6** in the reaction mixture after heating hydroxyketones **1** with Wittig reagent **2** [5] can be also explained by an ester exchange of these reaction components giving an intermediate ylide, which cyclizes rapidly to **6**. A characteristic light blue fluorescence at 254 and also at 366 nm was observed for all butenolides **6** on tlc plates. In the ir spectra of butenolides **6**, characteristic absorption bands occured at 1749-1790 cm^{-1} (lactone) and 1634-1644 cm^{-1} (lactam). The nmr spectra of butenolides **6** are generally in agreement with those of esters **4** inclusive of upfield shift of the H-6 proton doublet (corresponding to H-8 of esters **4**) of *N*-phenyl derivatives. However, the position of the signal of olefinic proton H-1 is variable. In deuteriochloroform solution, this singlet appears at 6.07-6.36 ppm in the nmr spectra of all derivatives with exception of 3a-benzyl derivatives, in which it is shifted upfield to the region of 5.80-5.88 ppm due to circular current of the π-electrons of the benzyl group. The position of the H-1 proton signal is, however, strongly influenced also by the solvent. In the hexadeuteriodimethyl sulfoxide, this singlet occured in the region of 6.51-6.91 ppm or, in the case of 3a-benzyl derivatives, at 6.23-6.40 ppm.

EXPERIMENTAL

The melting points were determined on a Kofler block or Gallenkamp apparatus and are uncorrected. The ir spectra were recorded on a Perkin-Elmer 421 or Nicolet Impact 400 spectrophotometers using samples in potassium bromide disks or by

the DRIFT method. The nmr spectra were recorded in deuteriochloroform, unless otherwise indicated, using tetramethylsilane as the internal standard. The instruments used were a Varian XL 200 at 200 MHz and a Bruker Alance DRX 500 at 500 MHz. The purity of substances was checked by thin-layer chromatography on Silufol UV 254 foils (Kavalier, Votice).

General Procedure for the Preparation of 3-Bromoacetoxy-1,2,3,4-tetrahydroquinoline-2,4-diones **4a-i**.

To the suspension of the appropriate 3-hydroxy-1,2,3,4-tetrahydroquinoline-2,4-dione **1a-f** (10 mmoles) in benzene (60 ml), pyridine (10.5 mmoles) was added and then the solution of bromoacetyl bromide (11 mmoles) in benzene (10 ml) was added with stirring at room temperature. After additional stirring at room temperature for 2 hours, the solution was heated to 60° and the precipitated pyridine hydrobromide was filtered by suction. The filtrate was evaporated to dryness *in vacuo* and the residue was crystallized.

General Procedure for the Preparation of 3-Triphenylphosphonioacetoxy-1,2,3,4-tetrahydroquinoline-2,4-dione Bromides **5a-i**.

A suspension of the appropriate 3-bromoacetoxy-1,2,3,4-tetrahydroquinoline-2,4-dione **4a-i** (10 mmoles) and triphenylphosphane (12 mmoles) in benzene (30 ml) was refluxed for 4-6 hours. During 15 minutes, the suspension became clear and after 20 minutes the precipitate, or the upper oily layer is formed. After cooling, the precipitate or glassy sediment was separated, washed with benzene and used for further experiments without crystallization.

General Procedures for the Preparation of 2,3a,4,5-Tetrahydrofuro[2,3-*b*]quinoline-2,4-diones **6a-i**.

Method A.

A solution of the appropriate bromide **5a-i** (5 mmoles) in chloroform (50 ml) was shaken with 0.5 *M* sodium hydroxide (20 ml) for 10-20 minutes. The chloroform layer was separated, washed with water, dried over anhydrous sodium sulfate, evaporated to dryness and the residue crystallized. In several cases, the product was insoluble in chloroform and precipitated from the reaction mixture. In such cases, the product was filtered by suction, washed with water and with chloroform and recrystallizated.

Method B.

To the solution of the appropriate bromide **5a-i** (5 mmoles) in chloroform (50 ml), triethylamine (10 mmoles) in benzene was added and the reaction mixture was stirred for 2 hours. Triethylamine hydrobromide which seperated was filtered with suction and the filtrate was treated as described in Method A.

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