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Several 2-alkylcarbamato/thiocarbamato/aryloxy/trichloromethyl-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-oxides (**4** and **6**) were synthesised by reacting 4-propoxy-*o*-phenylenediamine (**1**) with various *N*-dichlorophosphinyl carbamates (**3**), aryl phosphorodichloridates (**5a-f**) and trichloromethyl phosphonic dichloride (**5g**) in the presence of triethylamine at 45–65 °C. Their ir, ¹H, ¹³C, ³¹P nmr and mass spectral data are discussed. The compounds were screened for antifungal activity against *Curvularia lunata* and *Aspergillus niger* and for antibacterial activity against *Bacillus subtilis* and *Escherichia coli*. Most of these compounds exhibited moderate activity in the assays.

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Introduction.

Some diazaphospholes and certain phosphinyl carbamates have been demonstrated to possess insecticidal, bactericidal, antiviral, antitumour and anticarcinogenic activity [1-8]. Benzimidazol-2-yl carbamates have proven anthelmintic activity [9-10]. In view of the exhibition of potential bioactivity of these molecular skeletons, their phosphorus structural analogues, 2-substituted benzodiazaphosphole 2-oxides (**4** and **6**), which are a rare class of heterocycles have been synthesised, expecting them to possess broad spectrum of biological activity with less toxicity.

Results and Discussion.

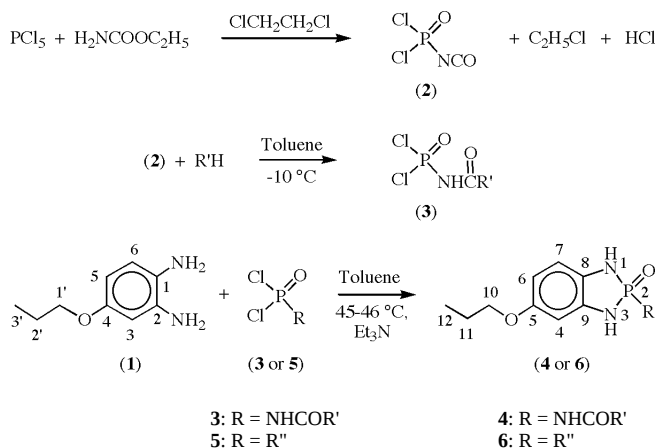
Addition of equivalent amounts of isocyanato phosphonic dichloride (**2**) [2,3] to various alcohols and thiols at -10 °C in dry toluene led to the corresponding *N*-dichlorophosphinyl carbamates (**3**). When 4-propoxy-*o*-phenylene

diamine (**1**) [11] was reacted with **3** or phosphoric and phosphinic acid dichlorides (**5**) in equimolar quantities, members of **4** or **6** were obtained, respectively (Scheme I). Presence of two equivalents of triethylamine in dry toluene at 45–65 °C served as ideal conditions for this reaction to occur. Isolation of pure products was achieved by filtering off the triethylamine hydrochloride and evaporation of the filtrate under reduced pressure. The residue on washing with water followed by 2-propanol and subsequent recrystallisation/trituration from methanol afforded analytically pure compounds. Interestingly, primary and secondary alcohols reacted readily with **2**, but tertiary alcohols failed to react under the same conditions due to steric factors.

Reaction yields, elemental analyses, ir and ³¹P nmr data of **4** and **6** are given in Table 1. Tables 2, 3 and 4 contain their ¹H, ¹³C nmr and mass spectral data. Presence of characteristic bands for P-NH (3398-3433 cm⁻¹), and P=O (1214-1292 cm⁻¹) in the ir spectra of **4** and **6** proved that cyclisation of **1** with **3/5** occurred to form the benzodiazaphosphole ring.

The ¹H nmr data (Table 2) agrees very well with the proposed structures for **4** and **6**, however it is observed that the protons of the carbamate group in **4** resonated downfield when compared to the signals of the corresponding protons in the free alcohols [17].

The ¹³C nmr chemical shifts of **4** and **6** were interpreted on the basis of additivity rules, C-P couplings, and intensity of signals. In **4**, C(4) and C(7) resonated as doublets at δ 98.6-102.1 (³J_{PC} = 12.6-13.9 Hz) and δ 111.5-119.2 (³J_{PC} = 12.6-13.8 Hz), respectively while the same carbons in **6** showed singlets at δ 101.1-101.9 and δ 115.4-116.8, respectively. Chemical shifts at δ 152.2-158.1 and δ 104.1-108.1 were assigned to C(5) and C(6), respectively. The nitrogen bearing carbons C(8) and C(9) gave signals at δ 124.8-128.4 and δ 131.5-136.9, respectively. In members of **4**, the signal for the carbonyl carbon C-1' appeared at δ 154.6-157.5, while that of C-2' of the carbamate/thiocarbamate function appeared downfield (δ 10-15) when compared to the corresponding signals in the respective free alcohols [17].



Compound	R'	Compound	R'	Compound	R''
3,4a	OCH ₃	3,4f	OC ₆ H ₁₁	5,6a	OC ₆ H ₅
b	OCH ₂ CH ₃	g	OCH ₂ C ₆ H ₅	b	OC ₆ H ₄ -CH ₃ (2')
c	OCH ₂ CH ₂ Cl	h	SCH ₂ CH ₂ CH ₃	c	OC ₆ H ₄ -CH ₃ (3')
d	OCH(CH ₃) ₂	i	S(CH ₂) ₃ CH ₃	d	OC ₆ H ₄ -CH ₃ (4')
e	OCH ₂ CH(CH ₃) ₂			e	OC ₆ H ₄ -Cl(2')
				f	OC ₆ H ₄ -Cl(4')
				g	CCl ₃

Table 1

Physical, IR and ^{31}P NMR Spectral Data of 2-Substituted-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-Oxides **4** and **6**

Compound	Yield (%)	Mp (°C)	Molecular Formula	Elemental Analysis					IR (cm ⁻¹)				³¹ P NMR data [c] ppm		
				Calcd / Found					P=O	P-NH	P-NH-CO	C=O		P-O-C(aron)	
				C	H	N	O-C	P-O							
4a	55[a]	212-214	C ₁₁ H ₁₆ N ₃ O ₄ P	46.32	5.65	14.73	1292	3398	3097	1734	-	-	-3.27,		
				46.30	5.51	14.70						17.63			
4b	52[a]	180-182	C ₁₂ H ₁₈ N ₃ O ₄ P	48.16	6.06	14.04	1221	3400	3160	1704	-	-	-3.21,		
				47.98	5.90	14.12						17.52			
4c	62[a]	196-198	C ₁₂ H ₁₇ ClN ₃ O ₄ P	43.19	5.13	12.59	1287	3400	3112	1737	-	-	17.20		
				43.08	5.20	12.72									
4d	56[a]	212-214	C ₁₃ H ₂₀ N ₃ O ₄ P	49.84	6.43	13.40	1269	3401	3109	1719	-	-	17.51		
				49.90	6.32	13.60									
4e	52[a]	228-230	C ₁₄ H ₂₂ N ₃ O ₄ P	51.37	6.77	12.83	1292	3407	3107	1728	-	-	-3.06,		
				51.20	6.65	12.74						18.14			
4f	48[a]	162-164	C ₁₆ H ₂₃ N ₃ O ₄ P	54.54	6.66	11.92	1221	3400	3176	1712	-	-	-3.04,		
				54.40	6.58	11.82						17.12			
4g	48[a]	190-192	C ₁₇ H ₂₀ N ₃ O ₄ P	56.50	5.57	11.63	1262	3402	3120	1728	-	-	-3.26,		
				56.40	5.44	11.72						17.12			
4h	40[a]	186-188	C ₁₃ H ₂₀ N ₃ O ₃ PS	47.41	6.12	12.76	1276	3398	3160	1704	-	-	-4.15,		
				47.46	6.02	12.89						3.59			
4i	38[a]	180-182	C ₁₄ H ₂₂ N ₃ O ₃ PS	48.97	6.46	12.23	1282	3404	3162	1706	-	-	-4.10,		
				48.86	6.58	12.29						3.31			
6a	58[b]	194-196	C ₁₅ H ₁₇ N ₂ O ₃ P	59.21	5.63	9.21	1218	3433	-	-	1201	966	-1.98,		
				59.12	5.65	9.12						-2.26			
6b	56[b]	205	C ₁₆ H ₁₉ N ₂ O ₃ P	60.37	6.01	8.80	1230	3402	-	-	1198	970	-2.08,		
				60.22	5.94	8.68						-2.27			
6c	50[b]	184-186	C ₁₆ H ₁₉ N ₂ O ₃ P	60.37	6.01	8.80	1223	3412	-	-	1196	964	-1.05,		
				60.50	6.13	8.88						-2.25			
6d	56[b]	204	C ₁₆ H ₁₉ N ₂ O ₃ P	60.37	6.01	8.80	1231	3428	-	-	1213	964	-1.02,		
				60.30	6.12	8.73						-2.12			
6e	46[b]	194-196	C ₁₅ H ₁₆ ClN ₂ O ₃ P	53.19	4.76	8.28	1219	3430	-	-	1190	968	-1.71,		
				52.89	4.85	8.22						0.94			
6f	48[b]	188-190	C ₁₅ H ₁₆ ClN ₂ O ₃ P	53.19	4.76	8.28	1214	3405	-	-	1177	971	-2.01,		
				53.40	4.82	8.20						0.44			
6g	32[a]	248-250	C ₁₀ H ₁₂ Cl ₃ N ₂ O ₂ P	36.43	3.67	8.50	1237	3430	-	-	-	-	6.55		
				36.32	3.52	8.48									

[a] Recrystallized from methanol, reported yields are after one recrystallisation. [b] Triturated with methanol. [c] Chemical shifts in ppm from 85% phosphoric acid.

The ^{31}P nmr signals appeared in the range of 3.04 to 3.27 ppm and 17.12 to 18.14 ppm for the alkylcarbamate compounds (**4a-g**), whereas in the thiocarbamate compounds (**4h-i**) the ^{31}P chemical shifts occurred upfield at -4.10 to -4.15 ppm and 3.31 to 3.59 ppm due to the difference in electronegativity of oxygen and sulfur [18-20]. In **6a-f**, two signals were observed at 0.44 to -2.08 and -1.71 to -2.27 ppm for ^{31}P , whereas **6g** showed only one signal for ^{31}P at 6.55 ppm. The striking appearance of two ^{31}P nmr signals in the spectra of **4** and **6** lends to the supposition that they may exist in at least two conformers [21].

In the electron impact (70 eV) mass spectra, molecular ions (M^{+}) were not observed for **4f-h**. All other compounds exhibited M^{+} and important daughter ion peaks [22] confirming the proposed structures.

Antimicrobial Activity.

All the compounds were tested at two different concentrations (250 and 500 ppm) for their antifungal activity following Benson [23] technique against *Aspergillus niger* and *Curvularia lunata*. Their antibacterial activity was evaluated on *Bacillus subtilis* and *Escherichia coli* by the method of Vincent and Vincent [24]. Most of them exhibited moderate toxicity against either the fungi or bacteria.

EXPERIMENTAL

Melting points were determined on a Mel-temp apparatus and were uncorrected. Elemental analyses were performed by the Central Drug Research Institute, Lucknow, India. The ir spectra were recorded as KBr pellets on a Perkin-Elmer 283 double

Table 2

¹H NMR Chemical shift (*J* in Hz) [a] of 2-Substituted-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-Oxides **4** and **6**

Compound	H(4,6 & 7)	H(10, 11 & 12)	R'-H
1 [b]	6.34 (d, 2.5, 3-H) 6.49 (dd, 8.9, 2.5, 5-H) 7.96 (d, 8.9, 6-H)	3.89 (t, 2H,1'-H) 1.74 -1.83 (m, 2H,2'-H) 1.02 (t, 3H, 3'-H)	4.46 (br s, 4H, 2NH ₂)
4a [b]	6.46 (s, 4-H) 6.35 (dd, 8.2,2.3, 6-H) 6.67 (d, 8.2,7-H)	3.90 (t, 2H, 10-H) 1.70 -1.85 (m, 2H,11-H) 1.02 (t, 3H,12-H)	3.60 (s, 3H, OCH ₃)
4b [b]	6.54 (s, 4-H) 6.40 (dd, 8.3,2.2, 6-H) 6.70 (d, 8.2, 7-H)	3.92 (t, 2H, 10-H) 1.76-1.82 (m, 2H, 11-H) 1.01 (t, 3H, 12-H)	4.10 (q, 2H, OCH ₂) 1.20 (t, 3H, CH ₃)
4c [b]	6.46 (s, 4-H) 6.37 (dd, 8.5, 2.3, 6-H) 6.67 (d, 8.3, 7-H)	3.86 (t, 2H, 10-H) 1.71-1.82 (m, 2H, 11-H) 1.02 (t, 3H, 12-H)	4.17 (t, 2H, OCH ₂) 3.33 (t, 2H, CH ₂ Cl)
4d [b]	6.43 (s, 4-H) 6.33 (dd, 8.3, 2.1, 6-H) 6.65 (d, 8.3, 7- H)	3.83 (t, 2H, 10-H) 1.72-1.79 (m, 2H, 11-H) 1.02 (s, 3H, 12-H)	4.74- 4.89 (m, 1H, CH) 0.89 (d, 6.2, 6H, 2CH ₃)
4e [b]	6.40 (s, 4- H) 6.34 (dd, 8.4, 2.1, 6-H) 6.61 (d, 8.3, 7-H)	3.84 (t, 2H, 10-H) 1.68-1.79 (m, 2H, 11-H) 1.01 (t, 3H, 12-H)	3.67 (d, 6.5, 2H, OCH ₂) 1.45 -1.60 (m, 1H,CH) 0.74 (d, 6.6, 6H, 2CH ₃)
4f [b]	6.84 (s, 4-H) 6.36 (dd, 8.4, 2.3, 6-H) 6.73 (d,8.6, 7-H)	3.91 (t, 2H, 10-H) 1.71-1.82 (m, 2H, 11-H) 1.01 (t, 3H, 12-H)	4.55-4.70 (m, 1H,OCH) 1.20-1.58 (m, 10H)
4g [c]	6.48 (s, 4-H) 6.37 (dd, 8.3, 2.4, 6-H) 6.69 (d, 8.6, 7-H)	3.86 (t, 2H, 10-H) 1.72-1.81 (m, 2H, 11-H) 1.01 (t, 3H, 12-H)	4.99 (s, 2H, OCH ₂) 7.25 -7.33 (m, Ar-H)
4h [c]	6.72 (s, 4-H) 6.55 (dd, 8.2,2.3, 6-H) 6.95 (d, 8.5, 7-H)	3.89 (t, 2H, 10-H) 1.72-1.8 (m, 2H,11-H) 1.02 (t, 3H, 12-H)	2.8 0(t, 2H, SCH ₂) 1.52 -1.65 (m, 2H, CH ₂) 0.96 (t, 3H, CH ₃)
4i [c]	6.73 (s, 4-H) 6.55 (dd, 8.4, 2.3, 6-H) 6.83 (d, 8.4, 7-H)	3.90 (t, 2H, 10-H) 1.74-1.78 (m, 2H, 11-H) 1.02 (t, 3H, 12-H)	2.88 (t, 2H, SCH ₂) 2.00-2.15 (m, 2H, 2'-CH ₂) 1.15-1.40 (m, 2H, 3'-CH ₂) 0.95 (t, 3H, 4'-CH ₃)
6a [b]	6.84 (s, 4-H) 6.66 (d, 8.4, 6-H) 6.85 (d, 8.3, 7-H)	3.86 (t, 2H, 10-H) 1.72-1.88 (m, 2H, 11-H) 1.02 (t, 3H, 12-H)	6.90-7.02 (m, 5H)
6b [b]	6.68 (s, 4-H) 6.52 (d, 8.4, 6-H) 6.71 (d, 8.3, 7-H)	3.83 (t, 2H, 10-H) 1.71-1.79 (m, 2H, 11-H) 0.99 (t, 3H, 12-H)	6.95-7.08 (m, 4H) 2.11 (s, 3H, CH ₃)
6c [c]	6.72 (s, 4-H) 6.54 (d, 8.2, 6-H) 6.75 (d, 8.3, 7-H)	3.78 (t, 2H, 10-H) 1.70-1.75 (m, 2H, 11-H) 0.96 (t, 3H, 12-H)	6.94-7.8 (m, 4H) 2.15 (s, 3H, CH ₃)
6d [c]	6.79 (s, 4-H) 6.52 (d, 8.3, 6-H) 6.89 (d, 8.4, 7-H)	3.76 (t, 2H, 10-H) 1.68-1.73 (m, 2H, 11-H) 0.93 (t, 2H, 12-H)	6.92 -7.10 (m, 4H) 2.21 (s, 3H, CH ₃)
6e [b]	6.96 (s, 4-H) 6.58 (d, 8.2, 6-H) 7.10 (d,8.3, 7-H)	3.81 (t, 2H, 10-H) 1.62-1.78 (m, 2H, 11-H) 0.96 (t, 3H, 12-H)	6.58-7.58 (m, 4H)

[a] Chemical shifts in δ and *J* (Hz) given in parentheses. [b] Recorded in dimethyl sulfoxide-*d*₆. [c] Recorded in acetic acid-*d*₄.

beam spectrophotometer. ¹H, ¹³C and ³¹P nmr spectra were recorded on a Varian Gemini 300 MHz spectrometer operating at 299.9 MHz for H-1, 75.46 MHz for C-13 and 121.7 MHz for P-31. All nmr data were taken in dimethyl-*d*₆ sulfoxide or acetic acid-*d*₄ solutions and were referenced to tetra-

methylsilane (¹H and ¹³C) or 85% phosphoric acid (³¹P). Mass spectra were recorded on a Auto Spec instrument using a solid probe at 70 eV.

4-Propoxy-*o*-phenylene diamine (**1**) was prepared from 4-hydroxyacetaldehyde by following the reported procedure [11].

Table 3
¹³C NMR Chemical Shift Data [a] of 2-Substituted-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-Oxides **4** and **6**

Carbon Atoms	4a[b]	4b[b]	4c[b]	4d[b]	4e[b]	4i[c]	6b[b]	6c[c]	6e[b]	6g[b]
C-4	100.6 (13.4)	100.4 (13.3)	98.6 (12.6)	100.5 (13.9)	100.0 (13.1)	102.1	101.1	101.9	101.7	101.4
C-5	154.6	154.5	152.2	154.6	154.7	156.4	-	158.1	157.3	157.7
C-6	106.8	106.6	104.1	106.7	106.8	107.5	108.1	107.8	107.9	106.2
C-7	111.6 (13.2)	111.5 (12.9)	119.2 (12.6)	111.6 (12.5)	111.6 (12.5)	109.3 (13.8)	115.4	116.4	116.8	116.7
C-8	126.6	126.5	124.8	126.8 (14.7)	126.3	126.4	126.8	128.4	126.5	126.2
C-9	133.6	133.5	132.9	133.7 (14.7)	133.4	134.9	-	131.5	136.3	136.9
C-10	72.8	72.3	68.5	71.4	73.4	68.9	67.7	68.4	68.4	68.8
C-11	23.3	23.2	20.6	23.4	23.1	22.1	20.2	21.1	21.1	21.0
C-12	10.5	10.6	7.9	10.8	10.5	10.3	7.6	9.2	9.0	9.2
C-1'	156.5	156.0	154.6	155.6	155.9	157.5	-	151.7	148.5	77.8
C-2'	54.0	60.8	64.1	71.3	71.1	72.9	-	120.0	128.7	
C-3'		18.6	42.0	23.4	32.1	37.2	-	140.9	-	
C-4'					24.0	21.3	126.8	128.1	126.5	
C-5'						13.5	127.3	128.7	123.0	
C-6'							119.1	119.2	120.4	
C-1''							18.2	19.7		

[a] Chemical shifts in ppm. [b] Recorded in dimethyl sulfoxide-*d*₆. [c] Recorded in acetic acid-*d*₄.

Table 4

Mass spectral data [a] of 2-Substituted-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-Oxides **4** & **6**

Compound	m/z (relative abundance)
1	166 [100, (M ⁺)], 124 (45.5), 107 (5.8), 95 (28.0)
4a	285 [23.9, (M ⁺)], 253 (64.1), 228 (35.8), 211 (100), 186 (38.8), 168 (70.0), 150 (50.7), 121 (25.3), 93 (18.7)
4b	299 [6.2, (M ⁺)], 253 (6.2), 228 (12.5), 211(21.5), 186 (31.2), 168 (65.6), 150 (62.5), 124 (100), 95 (37.5)
4c	333 [3.1, (M ⁺)] 253 (75), 228 (6.2), 211 (100), 186 (6.2), 168 (37.5), 150 (6.2), 121 (12.5)
4d	313 [12.0, (M ⁺)], 253 (36.0), 228 (50.0), 211 (100), 186 (32.0), 168 (44.0), 121 (12.0), 81 (12.0)
4e	327 [14.8, (M ⁺)], 253 (37), 228 (55.6), 211 (100), 186 (34.0), 168 (37.8), 121 (11.0), 93 (6.6)
4f	192(44.0), 166 (8.0), 150 (100), 122 (12.0), 95 (10.0), 82 (26.0)
4g	282 (10.2), 240 (7.7), 192 (41.0), 150 (100), 122 (8.9), 91 (12.8)
4h	192 (24.0), 166 (22.0), 150(100), 124 (44.0), 95 (24.0)
6a	304 [44.0, (M ⁺)], 262 (46.0), 166 (48.0), 125 (100), 94 (56.0), 78 (16.0)
6b	318 [44.9, (M ⁺)], 276 (38.8), 166 (30.6), 124 (100), 108 (36.7), 95 (28.6)
6d	318 [37.5, (M ⁺)], 276 (31.2), 166 (53.1), 124 (100), 108 (31.2), 95 (28.1), 78 (12.5)
6e	340 [12.0, (M ⁺ +2)], 338 [38.8 (M ⁺)], 296 (38.8), 166 (57.1), 124 (100), 95 (24.5), 78 (8.1)

[a] **4f**, **4g** and **4h** did not show M⁺. [b] **4i**, **6c**, **6f** & **6g** were not recorded.

2-Isopropylcarbamato-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-Oxide (**4d**).

A solution of 2-propanol (0.60 g, 0.01 mole) in dry toluene (20 ml) was added dropwise over a period of 20 minutes to a cold

Table 5

Antimicrobial Activity of 2-Substituted-2,3-dihydro-5-propoxy-1*H*-1,3,2-benzodiazaphosphole 2-Oxides **4** and **6**

Compound	Zone of Inhibition (mm)							
	Fungi				Bacteria			
	<i>Curvularia Lunata</i>	<i>Aspergillus niger</i>	<i>Bacillus subtilis</i>	<i>Escherichia coli</i>				
	250 500	250 500	250 500	250 500				
4a	6 12	7 14	13 18	6 10				
4b	15 23	12 19	13 19	8 13				
4c	12 20	8 12	8 12	7 12				
4d	15 24	9 17	8 12	8 16				
4e	7 12	6 10	12 19	10 18				
4f	7 14	7 12	15 25	8 16				
4g	10 19	9 18	13 25	9 17				
4h	17 28	12 19	17 23	12 20				
4i	16 31	11 18	16 26	11 21				
6a	7 14	6 11	11 19	13 20				
6b	6 11	9 18	9 16	9 16				
6c	14 26	11 19	12 18	7 13				
6d	13 21	12 20	10 18	9 18				
6e	12 21	12 23	10 18	8 14				
6f	12 20	10 16	14 20	9 16				
6g	16 28	14 24	16 30	11 19				
Penicillin					24		20	
Tetracycline					32		28	
Griseofulvin	34	34						

Concentration in ppm.

solution (-10 °C) of isocyanato phosphonic dichloride (**2**, 1.60 g, 0.01 mole) in dry toluene (20 ml). After completion of the addition, the temperature of the reaction mixture was allowed to raise slowly to room temperature and stirring was continued for an additional 2 hours. This reaction mixture was added dropwise to a cold solution (0 °C) of 4-propoxy-*o*-phenylenediamine (**3**,

1.66g, 0.01 mole) and triethylamine (2.02 g, 0.02 mole) in dry toluene (50 ml). After the addition, the temperature of the reaction mixture was allowed to raise slowly to 45-55 °C and stirring was continued for an additional 3-4 hours. Progress of the reaction was monitored by tlc analysis. The reaction mixture was filtered to separate triethylamine hydrochloride and the solvent was evaporated from the filtrate under reduced pressure. The residue after washing with water was recrystallised from methanol to give 1.75 g of pure **4d** in 56% yield, mp 212-214 °C. Physical and spectral data of **4d** are given in Tables 1-4. **4a-i** were prepared by this procedure.

2-(2-Chlorophenoxy-2,3-dihydro-1H-1,3,2-benzodiazaphosphole 2-Oxide (**6e**).

A solution of 2-chlorophenyl phosphorodichloridate (**5e**, 2.46 g, 0.01 mole) in dry toluene (25 ml) was added dropwise over 20 minutes to a stirred solution of **3** (1.66 g, 0.01 mole) and triethylamine (2.02 g, 0.02 mole) in dry toluene (60 ml). After the addition, the temperature was slowly raised to 55-65 °C and stirred for 6-7 hours. TLC analysis (silica gel) was used to monitor the progress of the reaction. The product was isolated by the procedure described above and purified by trituration with methanol to afford 1.55 g (46%) of **6e**, mp 194-196 °C. Physical and spectral data of **6e** are given in Tables 1-4. **6a-g** were prepared by this procedure.

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