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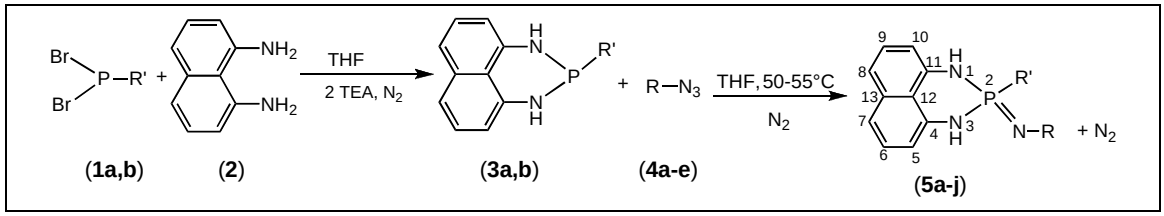

The title compounds (**5a-j**) were synthesized by a two step process. 4-Chlorophenyl and bis(2-chloroethyl) amino-dibromophosphites (**1a** and **1b**) were reacted with 1,8-diaminonaphthalene (**2**) to form the diazaphosphinines (**3a** and **3b**). They were further reacted with different alkyl azides (**4a-e**) in THF at 50-55 °C under N_2 atmosphere to afford the corresponding iminophosphoranes (**5a-j**). Their structures were established by elemental analysis, IR, ^{1}H, ^{13}C, ^{31}P NMR and Mass spectral data. All of them exhibited moderate antibacterial activity.

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INTRODUCTION

Reaction of a tertiary phosphine with an organic azide to produce an iminophosphorane [1], known as the Staudinger reaction [2] is a versatile tool in organic synthesis [3]. Nitrogen mustard derivative of cyclic organophosphorus compound, such as cyclophosphamide is known to exhibit antitumour activity [4]. In the primary imination process, phosphazides are important intermediates that have been isolated [5] or trapped *via* an intramolecular reaction [6]. But in most cases such phosphazides lose nitrogen at room temperature or even at lower temperature to give the corresponding iminophosphoranes in practically quantitative yields. Stable and isolable phosphazides [7] were formed in the case of sterically hindered and the electron releasing substituents on phosphorus and electron decreasing groups on the α-N of the azides [8,9]. Iminophosphosharanes play an important role in heterocyclic synthesis [10-12]. The versatility of phosphinimines in the synthesis of heterocycles embedded with high-valent transition metals is well documented [13]. The asymmetrically substituted phosphine ligands are also good chelating agents due to the presence of the phosphine and imine nitrogen electron rich centres [14].

In the course of our studies on six-membered phosphorus fused heterocycles having externally linked iminophosphorane moiety from 1,8-diamino naphthalene by Staudinger reaction, synthesis of the title compounds (**5a-j**) has been accomplished.

RESULTS AND DISCUSSION

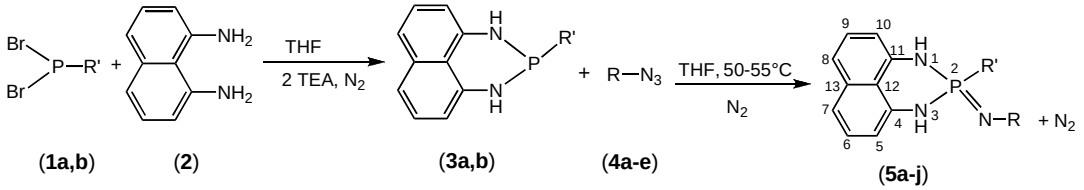
p-Chlorophenol/bis(2-chloroethyl) amine hydrochloride on reaction with phosphorus tribromide in the presence of triethylamine (TEA) in dry tetrahydrofuran (THF) under N_2 atmosphere at room temperature form the corresponding trivalent dibromophosphites (**1a** and **1b**). Reaction of **1a** and **1b** with 1,8-diaminonaphthalene (**2**) in two moles of TEA in dry THF under N_2 atmosphere affords 2-(4-chlorophenoxy)-2,3-dihydro-1*H*-naphtho[1,8-*de*][1,3,2]diazaphosphinine and *N*,*N*-di(2-chloroethyl)-*N*-(2,3-dihydro-1*H*-naphtho[1,8-*de*][1,3,2]diazaphosphinin-2-yl)amine (**3a** and **3b**). Different alkyl azides (**4a-e**) when treated with **3a** and **3b** in dry THF at 50-55°C under similar conditions afforded **5a-j** in 64-73% yield. They melt in the range of 220-252°C. Thin layer chromatography (TLC) was employed to monitor the reaction progress and to determine the purity of the products.

IR absorptions in the region [15-18] 1220-1235, 1370-1385, 1020-1035 and 3163-3289 cm^{-1} are assigned for P=N, P-N, N-C and P-NH stretching frequencies respectively.

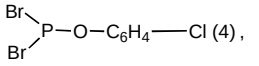
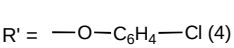
Naphthalene moiety [15] protons of **5a-j** resonated as three doublets at δ 6.61-6.75 (5 & 10-H, J = 6.2 – 6.8 Hz), 7.31-7.49 (6 & 9-H, J = 7.9-8.1 Hz) and δ 7.21-7.35 (7 & 8-H, J = 7.9-8.3 Hz). The hydrogens of 4-chloro-phenoxy moiety (**5a-j**) gave two doublets at δ 7.15-7.25 (2′ & 6′-H, J = 7.5-8.0 Hz) and δ 7.21-7.41 (3′ & 5′-H, J =8.6-9.2 Hz). The –N-*CH*$_2$-CH$_2$-Cl protons gave signals as multiplet at δ 2.52-2.75 and the –N-CH$_2$-*CH*$_2$-Cl protons resonated as triplet at δ 3.68-3.71 (J = 11.4-11.7 Hz). The –NH protons [19] resonated as a doublet at δ 7.98-8.29 ($^2J_{P\text{-}N\text{-}H}$ = 4.0-4.5 Hz) due to coupling with phosphorus. The alkyl side chain protons exhibited signals in the expected range [20].

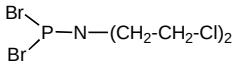
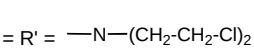
Compounds **5a, 5d, 5f** and **5j** exhibited only five ^{13}C NMR signals [15,20] for the ten naphthalene carbons because of their symmetrical disposition with respect to

Scheme 1



(**1a,b**) (**2**) (**3a,b**) (**4a-e**) (**5a-j**)

1a = R' =  , **3a** = R' = 

1b = R' =  , **3b** = R' = 

Compd.	R	Compd.	R′
5a,5f	$CH(CH_3)_2$	**5a-e**	$O\text{-}C_6H_4\text{-}Cl(4)$
5b,5g	$CH_2\text{-}CH{=}CH_2$	**5f-j**	$N\text{-}(CH_2\text{-}CH_2\text{-}Cl)_2$
5c,5h	$CH_2\text{-}CH_3$		
5d,5i	$CH_2\text{-}(CH_2)_2\text{-}CH_3$		
5e,5j	$CH_2\text{-}CH\text{-}(CH_3)_2$		

iminophosphorane system. The N-CH_2-CH_2-Cl in compounds **5f** and **5j** gave doublet at δ 43.0-43.1 ($^2J_{PNC}$ = 9.5-9.6 Hz) and –N-CH_2-CH_2-Cl resonated at δ 47.4-47.0. **5a** and **5d** exhibited ^{13}C NMR signals in the expected region. In the side chain, first carbon of the imino group (-P=N-*C*) gave doublet at δ 33.1-33.2 ($^2J_{P=N-C}$ = 9.7-10.0 Hz) due to coupling with phosphorus [21].

^{31}P NMR chemical shifts [17,22] (**5a-j**) appeared in the region -2.14 to 6.95 ppm.

Compounds **5a, 5d, 5f** and **5j** exhibited their respective molecular ions at m/z 370 (19.2, M-1), 383 (14.1, M-2), 384 (60.5, M-1) and 399 (22.8, $M^{+\bullet}$) in the FAB mass spectra [15].

Thus, the combined analytical, IR, NMR and mass spectral data conclusively agreed with the proposed structures for the title compounds (**5a-j**).

CONCLUSION

We successfully synthesized a series of novel iminophosphoranes in high yields by Staudinger reaction adopting a simple and straight forward procedure. The advantages are short reaction times, low cost of the starting chemicals, simple experimental procedure. These compounds exhibited moderate antibacterial activity.

EXPERIMENTAL

Solvents were used after purifying them by the established procedures. Progress of the reactions and purity of the compounds were monitored by thin layer chromatography (TLC) using *n*-hexane and ethyl acetate (1:1 by volume) as eluting system on silica gel (60-120 mesh) and iodine as visualizing agent. Melting points were determined in open capillary tubes on Mel-temp apparatus and were corrected. Microanalysis was performed at CDRI, Lucknow, India. IR spectra were recorded in KBr pellets on a Nicolet 380 FT double beam spectrophotometer ($\overline{\upsilon}$ in cm^{-1}) in Environmental Engineering Lab, S.V. University, Tirupati. ^{1}H, ^{13}C and ^{31}P NMR spectra were recorded at IISc, Bangalore, Bruker AMX 400 MHz spectrometer operating at 400 MHz, for ^{1}H and 100 MHz for ^{13}C, 161.9 MHz for ^{31}P NMR in CD_3OD. The ^{1}H and ^{13}C chemical shifts were referenced to tetramethyl silane and ^{31}P chemical shifts to 85% H_3PO_4. Mass spectra were recorded on a Jeol S X 102 DA/600 mass spectrometer using Argon / Xenon (6 keV, 10 mA) as the FAB (fast atom bombardment) gas and also a Shimadzu QP-2000, GC-MS instrument.

Synthesis of butyl azide (4d). In a dry 100 mL round bottomed flask fitted with dropping funnel, calcium chloride tube, sodium azide (0.32 g, 0.005 mole) and 10 mL of dry THF were placed and stirred. Butyl bromide (0.53 g, 0.005 mole) in 100 mL of dry THF was added to it at room temperature. Temperature of the reaction mixture was raised to 40-45°C stirred for 3-4 hrs, after cooling to room temperature to remove sodium bromide. The filtrate was used for the next step reaction.

Synthesis of [2-(4-Chloro-phenoxy)-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phospha-phenalen -2-ylidene]-butyl-amine (5d). *p*-Chlorophenol (0.57 g, 0.005 mole) in 10 mL of dry THF was added at 0-5°C to phosphorus tribromide (0.909 g, 0.005 mole) in the presence of TEA (0.005 mole) in dry THF (20 mL) under nitrogen atmosphere and stirred at room temperature for 2 hrs to form **1a**. The reaction mixture was filtered to remove triethylamine hydrobromide. To the filtrate a solution of 1,8-diaminonaphthalene (**2**) (0.79 g, 0.005 mole) in 20 mL of dry THF was added in the presence of triethyl amine (0.01 mole) under N_2 atmosphere and the reaction mixture was stirred for two hours to form 2-(4-chlorophenoxy)-2,3-dihydro-1*H*-naphtho[1,8-*de*][1,3,2]diaza-

phosphinine (**3a**). The reaction mixture was filtered under nitrogen atmosphere to remove triethylamine hydrobromide. To the filtrate butyl azide (**4d**) (0.005 mole) were added at room temperature and the mixture was stirred at 50-55°C under N_2 atmosphere for 2-3 hrs. The solvent was removed in a rotoevaporator. The crude residue was further purified by column chromatography using silica gel (60-120 mesh) as adsorbent and hexane and ethyl acetate (1:1) as an eluent to afford pure iminophosphorane (**5d**) with yield 2.06 g (66%), mp. 230-232°C. Compounds **5a-c** and **5e-j** were prepared by adopting the above procedure.

Physical, Analytical and Spectral Data for the test compounds (5a-j).

Characterization of [2-(4-chloro-phenoxy)-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phosphaphenalen-2-ylidene]-isopropyl-amine (5a). This compounds was obtained as brown color needles (ethanol), Yield: 1.98 g (65%), mp 220-222°C; ir: P=N 1235, P-N 1370, N-C 1020, P-NH 3163 cm^{-1}; ^{31}P nmr: δ 2.98; ^{1}H nmr: δ 6.75 (d, *J* = 6.3 Hz, 2H, 5 & 10-H), 7.45 (d, *J* = 8.1 Hz, 2H, 6 & 9-H), 7.34 (d, *J* = 8.2 Hz, 2H, 7 & 8-H), 7.18 (d, *J* = 7.5 Hz, 2H, 2′ & 6′-H), 7.24 (d, *J* = 9.2 Hz, 3′ & 5′-H), 8.27 (d, *J* = 4.2 Hz, 2H, P-NH), 2.09-2.00 (m, H, -*CH*-$(CH_3)_2$), 1.50-1.45 (m, 6H, -CH-$(CH_3)_2$); ^{13}C nmr: δ 109.1 (C-5 & C-10), 128.0 (C-6 & C-9), 122.7 (C-7 & C-8), 130.2 (C-12), 131.3 (C-13), 112.4 (C-4 & C-11), 148.2 (C-1′), 123.0 (C-2′ & C-6′), 129.6 (C-3′ & C-5′), 124.0 (C-4′), 33.1 ($^2J_{PNC}$ = 9.8 Hz, -*CH*-$(CH_3)_2$), 25.4 (-CH-$(CH_3)_2$); ms: m/z 370 (19.2, M-1), 328 (49.1), 290 (35), 254 (33.3), 203 (66.6), 146 (40.5), 120 (100), 83 (96.1). *Anal*. Calcd for $C_{19}H_{19}N_3POCl$: C, 61.37; H, 5.15; N, 11.30. Found: C, 61.32; H, 5.11; N, 11.26.

Characterization of [2-(4-chloro-phenyl)-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phosphaphenalen-2-ylidene]-propenyl-amine (5b). This compounds was obtained as brown color prisms (ethanol), Yield: 2.02 g (67%); mp 228-230°C; ir: P=N 1238, P-N 1373, N-C 1022, P-NH 3220 cm^{-1}; ^{31}P nmr: δ 0.31; ^{1}H nmr: δ 6.68 (d, *J* = 6.4 Hz, 2H, 5 & 10-H), 7.44 (d, *J* = 8.0 Hz, 2H, 6 & 9-H), 7.27 (d, *J* = 8.1 Hz, 2H, 7 & 8-H), 7.17 (d, *J* = 7.8 Hz, 2H, 2′ & 6′-H), 7.21 (d, *J* = 9.0 Hz, 2H, 3′ & 5′-H), 7.98 (d, *J* = 4.3 Hz, P-NH), 3.55-3.49 (m, H, -*CH*$_2$-CH=CH$_2$), 5.98-5.90 (m, H, -CH$_2$-*CH*=CH$_2$), 5.22 (d, *J* = 5.7 Hz, 2H, -CH$_2$-CH=*CH*$_2$). *Anal*. Calcd for $C_{19}H_{17}N_3POCl$: C, 61.71; H, 4.63; N, 11.36. Found: C, 61.65; H, 4.60; N, 11.31.

Characterization of [2-(4-chloro-phenoxy)-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phospha-phenalen-2-ylidene]-ethyl-amine (5c). This compound was obtained as brown color needles (ethanol), Yield: 2.01 g (68%); mp 224-226°C; ir: P=N 1230, P-N 1375, N-C 1027, P-NH 3289 cm^{-1}; ^{31}P nmr: δ 1.12; ^{1}H nmr: δ 6.72 (d, *J* = 6.2 Hz, 2H, 5 & 10-H), 7.41 (d, *J* = 7.9 Hz, 2H, 6 & 9-H), 7.31 (d, *J* = 8.0 Hz, 2H, 7 & 8-H), 7.15 (d, *J* = 7.7 Hz, 2H, 2′ & 6′-H), 7.22 (d, *J* = 8.9 Hz, 2H, 3′ & 5′-H), 8.29 (d, *J* = 4.5 Hz, 2H, P-NH), 2.09-2.02 (m, 2H, -*CH*$_2$-CH$_3$), 1.24 (t, *J* = 11.5 Hz, 3H, -CH$_2$-*CH*$_3$). *Anal*. Calcd for $C_{18}H_{17}N_3POCl$: C, 60.42; H, 4.78; N, 11.74. Found: C, 60.35, H, 4.74; N, 11.70.

Characterization of [2-(4-chloro-phenoxy)-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phospha-phenalen-2-ylidene]-butyl-amine (5d). This compound was obtained as brown color needles (ethanol), Yield: 2.06 g (66%); mp 230-232°C; ir: P=N 1225, P-N 1380, N-C 1025, P-NH 3223 cm^{-1}; ^{31}P nmr: δ 6.95; ^{1}H nmr: δ 6.73 (d, *J* = 6.5 Hz, 2H, 5 & 10-H), 7.43 (d, *J* = 8.0 Hz, 2H, 6 & 9-H), 7.35 (d, *J* = 8.3 Hz, 2H, 7 & 8 –H), 7.20 (d, *J* = 7.9 Hz, 2H, 2′ & 6′-H), 7.31 (d, *J* = 8.6 Hz, 2H, 3′ & 5′-H), 8.28 (d, *J* = 4.1 Hz, 2H, P-NH), 2.15-2.10 (m, 2H, -*CH*$_2$-CH$_2$-CH$_2$-CH$_3$), 1.60-1.54 (m, 2H, -CH$_2$-*CH*$_2$-CH$_2$-CH$_3$), 1.45-1.40 (m, 2H, -CH$_2$-CH$_2$-*CH*$_2$-CH$_3$), 0.96 (t, *J* = 6.2 Hz, 3H, -CH$_2$-CH$_2$-CH$_2$-*CH*$_3$); ^{13}C nmr: δ 110.1 (C-5 & C-10), 128.1 (C-6 & C-9), 122.6 (C-7 & C-8), 129.5 (C-12), 130.1 (C-13), 112.7 (C-4 & C-11), 147.3 (C-1′), 123.9 (C-2′ & C-6′), 127.2 (C-3′ & C-5′), 120.8 (C-4′), 33.0 ($^2J_{PNC}$=9.9 Hz, -*CH*$_2$-CH$_2$-CH$_2$-CH$_3$), 30.7 (*J* = 5.2 Hz, -CH$_2$-*CH*$_2$-CH$_2$-CH$_3$), 26.0 (-CH$_2$-CH$_2$-*CH*$_2$-CH$_3$), 14.1 (-CH$_2$-CH$_2$-CH$_2$-*CH*$_3$). ms: m/z 383 (14.1, M-2), 330 (50.3), 295 (46.2), 250 (76.2), 231 (76.5), 190 (100), 170 (48.5), 120 (85.2), 83 (95.5). *Anal*. Calcd. for $C_{20}H_{21}N_3POCl$: C, 62.26; H, 5.48; N, 10.89. Found: C, 62.20; H, 5.44; N, 10.83.

Characterization of [2-(4-chloro-phenoxy)-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phospha-phenalen-2-ylidene]-isobutyl-amine (5e). This compound was obtained as brown color prisms (ethanol), Yield: 2.00 g (64%); mp 235-237°C; ir: P=N 1231, P-N 1382, N-C 1023, P-NH 3215 cm^{-1}; ^{31}P nmr: δ -2.14; ^{1}H nmr: δ 6.75 (d, *J* = 6.7 Hz, 2H, 5 & 10-H), 7.49 (d, *J* = 8.1 Hz, 2H, 6 & 9-H), 7.34 (d, *J* = 7.9 Hz, 2H, 7 & 8 –H), 7.25 (d, *J* = 8.0 Hz, 2H, 2′ & 6′-H), 7.41 (d, *J* = 8.8 Hz, 2H, 3′ & 5′-H), 8.27 (d, *J* = 4.0 Hz, 2H, P-NH), 1.75-1.68 (m, 2H, -*CH*$_2$-CH-$(CH_3)_2$), 3.50-3.42 (m, H, -CH$_2$-*CH*-$(CH_3)_2$), 1.42 (d, *J* = 13.5 Hz, 6H, (-CH$_2$-CH-$(CH_3)_2$). *Anal*. Calcd. for $C_{20}H_{21}N_3POCl$: C, 62.26; H, 5.48; N, 10.89. Found: C, 62.20; H, 5.43; N, 10.84.

Characterization of 2-[bis-(2-chloro-ethyl)]-(2-isopropyl-imino-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phosphaphenalen-2-yl)-amine (5f). This compound was obtained as brown color needles (ethanol), Yield: 1.88 g (71%); mp 240-242°C; ir: P=N 1222, P-N

Table 1

Antibacterial activity[a] of compounds (**5a-j**).

Compd.	*Staphylococcus aureus*		*Bacillus faecalis*		*Escherichia coli*		*Klebsiella pneumonia*	
	100 µg/mL	200 µg/mL	100 µg/mL	200 µg/mL	100 µg/mL	200 µg/mL	100 µg/mL	200 µg/mL
5a	6	8	6	12	6	12	6	10
5b	6	9	6	10	6	8	6	12
5c	6	10	6	7	6	9	6	10
5d	6	12	6	7	6	10	8	13
5e	6	10	6	8	6	7	6	10
5f	7	13	6	10	7	12	7	11
5g	7	10	7	14	6	8	6	6
5h	7	10	7	15	6	10	6	7
5i	7	7	7	10	10	15	7	11
5j	7	9	7	10	7	10	6	9
Pencillin[b]	12		11		9		12	

[a]Zone of inhibition in mm; [b]Standard antibacterial compound.

1376, N-C 1039, P-NH 3219 cm^{-1}; ^{31}P nmr: δ 5.14; ^{1}H nmr: δ 6.68 (d, *J* = 6.8 Hz, 2H, 5 & 10-H), 7.48 (d, *J* = 8.0 Hz, 2H, 6 & 9-H), 7.30 (d, *J* = 8.1 Hz, 2H, 7 & 8-H), 2.75-2.69 (m, 4H, -N-*CH*$_2$-CH$_2$-Cl), 3.71 (t, *J* = 11.6 Hz, 4H, -N-CH$_2$-*CH*$_2$-Cl), 8.17 (d, *J* = 4.1 Hz, 2H, P-NH), 2.10-1.98 (m, H, -*CH*-(CH$_3$)$_2$), 1.61-1.52 (m, 6H, -CH-(*CH*$_3$)$_2$). ^{13}C nmr: δ 109.4 (C-5 & C-10), 128.2 (C-6 & C-9), 122.5 (C-7 & C-8), 129.5 (C-12), 130.1 (C-13), 112.4 (C-4 & C-11), 43.0 (*J* = 9.5 Hz, -N-*CH*$_2$-CH$_2$-Cl), 47.4 (-N-CH$_2$-*CH*$_2$-Cl), 33.0 ($^{2}J_{PNC}$ = 10.0 Hz, -*CH*-(CH$_3$)$_2$), 23.7 (-CH-(*CH*$_3$)$_2$); ms: m/z 384 (60.5, m-1), 350 (46.5), 321 (55.2), 273 (68.4), 208 (100), 178 (20.5), 120 (80.3), 87 (24.5). *Anal*. Calcd. for $C_{17}H_{23}N_4PCl_2$; C, 52.99; H, 6.01; N, 14.54; Found: C, 52.93; H, 5.57; N, 14.50.

Characterization of 2-[bis-(2-chloro-ethyl)]-(2-propenyl-imino-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phosphaphenalen-2-yl)-amine (5g). This compound was obtained as brown color needles (ethanol), Yield: 1.80 g (70%); mp 245-247°C; ir: 1229 (P=N), 1374 (P-N), 1034 (N-C), 3203 (P-NH) cm^{-1}; ^{31}P nmr: δ 0.22; ^{1}H nmr: δ 6.63 (d, *J* = 6.9 Hz, 2H, 5 & 10-H), 7.42 (d, *J* = 7.9 Hz, 2H, 6 & 9-H), 7.24 (d, *J* = 8.3 Hz, 2H, 7 & 8-H), 2.62-2.54 (m, 4H, -N-*CH*$_2$-CH$_2$-Cl), 3.71 (t, *J* = 11.7 Hz, 4H, -N-CH$_2$-*CH*$_2$-Cl), 8.18 (d, *J* = 4.2 Hz, 2H, P-NH), 3.58-3.47 (m, 2H, -*CH*$_2$-CH=CH$_2$), 6.03-5.94 (m, H, -CH$_2$-*CH*=CH$_2$), 5.31 (d, *J* = 5.8 Hz, 2H, -CH$_2$-CH=*CH*$_2$). *Anal*. Calcd. for $C_{17}H_{21}N_4PCl_2$; C, 53.27; H, 5.52; N, 14.61; Found: C, 53.20; H, 5.48; N, 14.56.

Characterization of 2-[bis-(2-chloro-ethyl)]-(2-ethylimino-2,3-dihydro-1*H*-1,3-diaza-2λ5-phosphaphenalen-2-yl)-amine (5h). This compound was obtained as brown color prisms (ethanol), Yield: 1.85 g (72%); mp 250-252°C; ir: P=N 1223, P-N 1384, N-C 1039, P-NH 3280 cm^{-1}; ^{31}P nmr: δ 1.16; ^{1}H nmr: δ 6.62 (d, *J* = 6.4 Hz, 2H, 5 & 10-H), 7.32 (d, *J* = 8.0 Hz, 2H, 6 & 9-H), 7.30 (d, *J* = 7.9 Hz, 2H, 7 & 8-H), 2.60-2.52 (m, 4H, -N-*CH*$_2$-CH$_2$-Cl), 3.68 (t, *J* = 11.5 Hz, 4H, -N-CH$_2$-*CH*$_2$-Cl), 8.15 (d, *J* = 4.0 Hz, 2H, P-NH), 2.08-2.00 (m, 2H, -*CH*$_2$-CH$_3$), 1.30 (t, *J* = 11.6 Hz, 3H, -CH$_2$-*CH*$_3$). *Anal*. Calcd. for $C_{16}H_{21}N_4PCl_2$; C, 51.76; H, 5.70; N, 15.09; Found: C, 51.70; H, 5.65; N, 15.03.

Characterization of 2-[bis-(2-chloro-ethyl)]-(2-butylimino-2,3-dihydro-1*H*-1,3-diaza-2λ^5-phosphaphenalen-2-yl)-amine (5i). This compound was obtained as brown color needles (ethanol), Yield: 1.94 g (71%); mp 244-246°C; ir: P=N 1220, P-N 1382, N-C 1035, P-NH 3283 cm^{-1}; ^{31}P nmr: δ 0.21; ^{1}H nmr: δ 6.62 (d, *J* = 6.5 Hz, 2H, 5 & 10-H), 7.31 (d, *J* = 7.9 Hz, 2H, 6 & 9-H), 7.23 (d, *J* = 8.2Hz, 2H, 7 & 8-H), 2.61-2.53 (m, 4H, -N-*CH*$_2$-CH$_2$-Cl), 3.69 (t, *J* = 11.4 Hz, 4H, -N-CH$_2$-*CH*$_2$-Cl), 8.12 (d, *J* = 4.1 Hz, 2H, P-NH), 2.09-2.03 (m, 2H, -*CH*$_2$-CH$_2$-CH$_2$-CH$_3$), 1.78-1.72 (m, 2H, -CH$_2$-*CH*$_2$-CH$_2$-CH$_3$), 1.48-1.41 (m, 2H, -CH$_2$-CH$_2$-*CH*$_2$-CH$_3$), 0.95 (t, *J* = 6.3 Hz, 3H, -CH$_2$-CH$_2$-CH$_2$-*CH*$_3$). *Anal*. Calcd. for $C_{18}H_{25}N_4PCl_2$; C, 54.41; H, 6.31; N, 14.03; Found: C, 54.34; H, 6.26; N, 13.97.

Characterization of 2-[bis-(2-chloro-ethyl)]-(2-isobutyl-imino-2,3-dihydro-1*H*-1,3-diaza-2λ5-phosphaphenalen-2-yl)-amine (5j). This compound was obtained as brown color prisms (ethanol), Yield: 2.00 g (73%); mp 247-249°C; ir: P=N 1226, P-N 1385, N-C 1031, P-NH 3282 cm^{-1}; ^{31}P nmr: δ 0.99; ^{1}H nmr: δ 6.61 (d, *J* = 6.6 Hz, 2H, 5 & 10-H), 7.33 (d, *J* = 8.0 Hz, 2H, 6 & 9-H), 7.21 (d, *J* = 8.1 Hz, 2H, 7 & 8-H), 2.61-2.54 (m, 4H, -N-*CH*$_2$-CH$_2$-Cl), 3.70 (t, *J* = 11.5 Hz, 4H, -N-CH$_2$-*CH*$_2$-Cl), 8.08 (d, *J* = 4.0 Hz, 2H, P-NH), 1.73-1.70 (m, 2H, -*CH*$_2$-CH-(CH$_3$)$_2$), 3.52-3.48 (m, 1H, -CH$_2$-*CH*-(CH$_3$)$_2$), 1.38-1.27 (m, 6H, -CH$_2$-CH-(*CH*$_3$)$_2$); ^{13}C nmr: δ 109.9 (C-5 & C-10), 128.0 (C-6 & C-9), 122.0 (C-7 & C-8), 128.3 (C-12), 129.5 (C-13), 112.5 (C-4 & C-11), 43.1 (*J* = 9.6 Hz, -N-*CH*$_2$-CH$_2$-Cl), 47.7 (-N-CH$_2$-*CH*$_2$-Cl), 33.2 ($^{2}J_{PNC}$ = 9.7 Hz, -*CH*$_2$-CH-(CH$_3$)$_2$), 30.7 (*J* = 5.2 Hz, -CH$_2$-*CH*-(CH$_3$)$_2$), 20.2 (-CH$_2$-CH-(*CH*$_3$)$_2$); ms: m/z (%): 399 (22.8, M-1), 381 (16.2), 342 (100), 297 (37.8), 222 (32.4), 175 (13.5), 127 (20.5), 97 (35.2). *Anal*. Calcd. for $C_{18}H_{25}N_4PCl_2$; C, 54.14; H, 6.31; N, 14.03; Found: C, 54.34; H, 6.27; N, 13.98.

Antibacterial Activity. Compounds (**5a-j**) were screened for their antibacterial activity (Table 1) against gram positive bacteria, *Staphylococcus aureus*, *Bacillus faecalis* and gram negative bacteria, *Escherichia coli*, *Klebsiella pneumania* by the disc diffusion method [23,24] in luria bertani nutrient agar medium at two concentrations (100, 200 μg/mL). The solutions containing 10^6 cells/mL were added to each filter paper disc (6 mm diameter) and DMSO was used as the control. The freshly prepared agar medium containing bacterial species was loaded to the discs. These plates were incubated at 35°C and examined for zone of inhibition around each disc after 24 hrs. The results were compared with the activity of the standard antibiotic (100 μg/mL).

All the compounds **5a-j** exhibited moderate antibacterial activity, but **5i** showed more antibacterial activity against *Escherichia coli* when compared to that of the standard.

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