Synthesis of α -(acylamino)polyhaloalkylphosphoryl compounds by the reaction of trivalent phosphorus chlorides with N-(α -hydroxypolyhaloalkyl)amides

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N-(α -Hydroxypolyhaloalkyl)amides react with trivalent phosphorus chlorides to give α -(α -Hydroxypolyhaloalkylphosphoryl compounds via phosphorotropic rearrangement of intermediate phosphites or phosphinites.

Key words: halogen-containing organic compounds, amidoalkylation; rearrangements; α -aminophosphonates, α -aminoalkylphosphinoxides.

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Derivatives of a-aminophosphonic acids have attracted the constant attention of researchers, which is evidenced by a continuously increasing number of publications concerning this problem. First of all, this is related to the diverse biological activity of such compounds. As phosphorus-containing analogs of a-aminocarboxylic acids, they have been used in the production of enzyme inhibitors, antibacterial and neuroactive compounds, plant growth regulators, etc.¹⁻³ In 1952, M. I. Kabachnik et al.4 and E. K. Fields⁵ independently of one another elaborated a simple and quite versatile method for creating a P-C bond in the α -position with respect to the amino group by reaction of ammonia (amines) with carbonyl compounds and dialkyl phosphites (the Kabachnik-Fields reaction), which largely stimulated the development of the chemistry of α -aminophosphoryl derivatives. Later, other methods have been proposed (see the latest reviews^{2,3}), but all these impose some restrictions. That is why a search for new approaches to the synthesis of compounds containing the phosphoryl and amino groups in the α -position with respect to each other still remains an urgent problem. In particular, traditional approaches are often insufficiently convenient for the synthesis of α -aminophosphoryl derivatives with polyhaloalkyl groups.

Earlier,⁶ the N-acylated derivatives of α -aminopolyhaloalkylphosphoryl compounds have been obtained mostly by the addition of hydrophosphoryl compounds to N-acylimines or by the reaction of α -chloroalkylamides with the alkoxy derivatives of P^{III}. We elaborated a simpler method based on the reaction of N-(α -hydroxypolyhaloalkyl)amides 1 with trivalent phosphorus chlorides 2 (Scheme 1)^{7,8} with subsequent phosphorotropic rearrangement 3 \rightarrow 4.

The initial hydroxyamides 1 can be obtained easily by condensation of polyhaloaldehydes with amides. The Scheme 1



D	CCI3	CH ₂ F	ElO	ĸ	CCI3	EtO	El2N
С	CCl ₃	CF ₃	EtO	ł	CCl ₃	Ph	EtO
d	CCl ₃	EtO	EtO	m	$H(CF_2)_4$	MeO	EtO
е	CCl ₃	2-CIC ₆ H ₄	EtO	n	$H(CF_2)_4$	EtO	EtO
f	CCI ₃	Me	Ph	0	$H(CF_2)_4$	EtO	Et ₂ N
g	CCl ₃	CH ₂ F	Ph	р	$H(CF_2)_4$	EtO	Ph
h	CCI3	CF ₃	Ph	q	$H(CF_2)_6$	EtO	Ph
ĭ	CCI ₃	2-CIC6H4	Ph	Г	CCI3	EtO	[2-0C ₆ H ₄ O]

reaction of hydroxyamides with phosphorus chlorides proceeds under mild conditions (10-20 °C), first resulting in O-phosphorylated derivatives (3) and then in C-phosphorylated ones (4). The ease of the phosphorotropic isomerization $3 \rightarrow 4$ is greatly influenced by the substituents at the C atom of the carbonyl group and the nature of the phosphorous group. An increase in the electron-withdrawing properties of the amide substituent R and the nucleophilicity of the P^{III} atom favor the rearrangement. When the phosphorus atom has donor substituents, the isomerization $3 \rightarrow 4$ easily occurs un-

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der the conditions of the synthesis of these phosphorylated derivatives from compounds 1 and 2 (Ph_2P , $(Et_2N)_2P > (EtO)_2P \gg (o-OC_6H_4O)P)$. When hydroxyamide 1 (R = EtO, $R_{Hal} = CCl_3$) reacts with (EtO)₂PCl or (Et₂N)₂PCl in the presence of Et₃N (ether, 15-20 °C), the ratio of isomers 4 : 3 amounts (after 1 h) to 1: 10 and 4: 1, respectively, whereas no intermediate phosphinite 3 (R' = Ph) was detected in the reaction of this hydroxyamide with Ph2PCl under similar conditions. This indicates that the rate-limiting stage in the latter case is the formation of compound 3 rather than its isomerization. The nucleophilicity of phosphorus in phenylenedioxyphosphite 3 ($R_{Hal} = CCl_3$ and $R'_2 =$ o-OC₆H₄O) (δ P 135) is lower, and heating is required to isomerize it. During isolation, the corresponding phosphonate 4r (δP 32.2) undergoes opening of the phospholane ring to give monoester 5 (Scheme 2).8

Scheme 2



To estimate the influence of the amide substituents on the rate of the phosphorotropic rearrangement $3 \rightarrow 4$, we determined the ratio between isomers 3 and 4 ($R_{Hal} = CCI_3$, R' = EtO) for different substituents R by ³¹P NMR spectroscopy, 1 and 3 h after mixing the reagents (ether, 15-20 °C):

R	4	3	R	4:3	
	1 h	3 h		1 h	3 h
Ph	>95 : 1	100 : 0	Me		1.5 : 1.0
CF ₁	12:1	100 : 0	EtO	1 : 10	4 : 10
CH ₂ F	<1 : 20	5:1			

From the above data, it follows that the nature of amide substituents R substantially influences the rate of isomerization $3 \rightarrow 4$ (Ph > CF₃ > CH₂F > Me > EtO), the influence of electron-withdrawing properties becoming particularly pronounced when small electrone-gative F atoms are introduced successively into the methyl group (CF₃ > CH₂F > CH₃).

A replacement of N-acyl substituent by the phosphoryl group does not hinder the rearrangement (Scheme 3, $6 \rightarrow 7$).

The presence of a polyhaloalkyl substituent R_{Hal} at the α -C atom is not required for the isomerization. Thus, the reaction of N-hydroxymethyltrifluoroacetamide with (EtO)₂PCl in the presence of Et₃N yields phosphite **8** (δ P 140.6), which is easily isomerized to give phosphonate **9** (Scheme 4). Twenty minutes after mixing the reagents (ether, 15–20 °C) the **9** to **8** ratio is equal to 10 : 1, and after 1.5 h no more isomer **8** is present in the reaction mixture. Thus, the rearrange-



ment $8 \rightarrow 9$ occurs even faster than it does in the case of trichloromethyl analog 3 (R_{Hal} = CCl₃, R⁺ = EtO), which can be explained by the steric influence of the trichloromethyl group.

The decisive factor for the rearrangement is the presence of an amide group in the type 3, 6, and 8 derivatives. Thus, compounds 10 and 11 are not isomerized into the corresponding phosphonates under similar conditions.



Phosphite 10 (δ P 140.1) remains unchanged with heating (100 °C, 2 h) or when distilled. Compound 11 (δ P 14 and 140.6) is stable at room temperature. When heated, it decomposes to give a series of unidentified products.

Compounds 4 ($R_{Hal} = CCl_3$) in the presence of bases are easily dehydrochlorinated (especially at R = EtO, Et_2N) to give the corresponding vinylamides 12 (cf. Ref. 6 and 9).



12: R = EtO, R' = EtO (a), Et_2N (b); R = Me, R' = EtO (c)

Com- pound	Yield (%)	M.p./°C {B.p./°C	<u>Found</u> Calcul	ated (%)	Molecular formula	$\frac{\delta^{3!} \mathbf{p}}{(\mathbf{A} \pm \mathbf{B})^a}$	IR, v/cm ⁻¹
		(p/Torr)]	Cl (F)	Р			
4a	62	115-116	<u>32.39</u> 32.57	<u>9.26</u> 9.49	C ₈ H ₁₅ Cl ₃ NO ₄ P	15.1	1050 (POC); 1255 (P=O); 1705 (C=O); 3270 (NH)
4b	48	98	<u>30.72</u> 30.87	<u>8.93</u> 8.99	C ₈ H ₁₄ Cl ₃ FNO ₄ P	12.8	
4c	38	110-112	<u>27.61</u> 27.95	<u>7.92</u> 8.14	$C_8H_{12}Cl_3F_3NO_4P$	12.1	1050 (POC); 1265 (P=O); 1735 (C=O); 3210 (NH)
4e	57	120	<u>33.84</u> 33.52	<u>7.30</u> 7.32	$C_{13}H_{16}Cl_4NO_4P$	13.0	1035 (POC); 1275 (P=O); 1680 (C=O); 3180 (NH)
4f	45	175	<u>26.93</u> 27.22	<u>8.21</u> 7.93	C ₁₆ H ₁₅ Cl ₃ NO ₂ P	30.7 (A); 31.4 (B) (5 : 1)	1200 (P=O); 1685 (C=O); 3190, 3270 (NH)
4g	49	212	<u>25.80</u> 26.02	<u>7.73</u> 7.58	C ₁₆ H ₁₄ Cl ₃ FNO ₂ P	28.3	1120 (C-F); 1195 (P=O); 1705 (C=O); 3200 (NH)
4h	56	168-170	<u>24.03</u> 23.92	<u>7.02</u> 6.97	C ₁₆ H ₁₂ Cl ₃ F ₃ NO ₂ P	28.7 (A); 27.8 (B) (20 : 1)	1170, 1210 (CF ₃ , P=O); 1725 (C=O); 3170 (NH)
4 i	52	220-222	<u>29.21</u> 29.11	<u>6.31</u> 6.36	$C_{21}H_{16}Cl_4NO_2P$	28.8	1205 (P=O); 1685 (C=O); 3150 (NH)
41	66	120-122	-		-	14.7	
4m	52	[130—134 (0.04)]	(<u>36.81</u>) (37.14)	<u>7.82</u> 7.57	C ₁₁ H ₁₆ F ₈ NO ₄ P	14.0	1275 (P=O); 1725 (C=O); 3210 (NH)
4q	24	157	-	<u>5.45</u> 5.13	$C_{22}H_{18}F_{12}NO_3P$	30.5	-
7	52	44	-	<u>12.13</u> 12.31	$C_{13}H_{23}F_8NO_6P_2$	7.6 (N-P); 16.2 (C-P)	_
9	63	[130—136 (0.05)]	(<u>21.30</u>) (21.66)	<u>11.94</u> 11.77	$C_7H_{13}F_3NO_4P$	20.7	

Table 1. The yields, elemental analysis data, some physicochemical characteristics, and 1R spectra of compounds 4, 7, and 9

^a The main and the minor isomers, respectively.

^b Literature data¹⁵: m.p. 121-122 °C.

That is why one should avoid using an excess of base in their synthesis. According to the literature data, 10 the preparation of compounds 4 ($R_{Hal} = CCl_3$, R' = Ph) by the Arbuzov reaction from tetrachloroethylamides CCI₃CH(CI)NHCOR and Ph₂POEt is accompanied by the formation of vinylamides 12, which prevents phosphinoxides 4 from being isolated in the individual state. At R' = EtO, the accompanying dehydrochlorination is not so characteristic. We believe that the dehydrochlorination in the course of the Arbuzov reaction occurs in a quasiphosphonium intermediate whose lifetime at R' = Ph is sufficient for partial realization of this process (cf. Ref. 11). Use of ester amides of phosphorous acid in the synthesis of compounds 4 is also not suitable. At the same time, the synthesis of phosphinoxides 4 by the reaction of hydroxyalkylamides 1 with Ph_2PCI or $(R_2N)_2PCI$ creates no problems.

The compositions and structures of N-acylated aminophosphoryl compounds 4, 7, 9, and 12 were confirmed by data from elemental analysis and ${}^{1}H$, ${}^{31}P$, and ${}^{19}F$ NMR and 1R spectroscopy (Tables 1 and 2). In particular, chemical shifts in the ³¹P NMR spectrum that correspond to the phosphorus atom of the phosphonate or phosphinoxide group as well as a characteristic value of spin-spin coupling constant ${}^{2}J_{HP} = 5-20$ Hz correlate well with the presence of the P--C bond. The signal for the CHP proton in the ¹H NMR spectra of compounds 4, recorded in solvents containing traces of water (e.g., acetone-d₆), appears as a superposition of a doublet of doublets (splitting on the P atom and the NH proton) and a doublet (splitting only on the P atom). When D_2O is added, this multiplet is transformed into a doublet. In the spectra of polyfluoroalkyl derivatives $(R_{Hal} = H(CF_2)_4$ and $H(CF_2)_6)$, the signal of the CHP proton takes the form of a complex multiplet owing to an additional spin-spin coupling with the diastereotopic F atoms.

A hindered rotation around the amide bond in the molecules of α -acylaminopolyhaloalkylphosphoryl compounds implies their geometric isomerism. Derivatives 4 and 12 obtained by us exist mainly as one isomer, but in some cases (see Tables 1 and 2 and Experimental),

Com-	,	¹ H NMR,	¹⁹ F NMR, δ	
pound	$\frac{\text{CHP}}{(^2 J_{\text{HP}}/^3 J_{\text{HH}})}$	NH (³ J _{HH})	Other signals	(<i>J</i> /Hz)
4a ^{<i>n</i>}	5.38 (dd, J = 18.5/10)	8.38 (d. $J = 10$)	1.30, 1.33 (both t, 6 H, 2 C \underline{H}_3 CH ₂ , $J = 7$); 2.12 (s, 3 H, MeCO); 4.12 (m, 4 H, 2 OCH ₂)	
4b ^a	5.35 (dd, J = 19/10.5)	8.03 (d, J = 10.5)	1.31, 1.33 (both t, 6 H, 2 CH ₃ CH ₂ , $J = 7$); 4.12 (m, 4 H, OCH ₂); 5.07 (d, 2 H, CH ₂ F, ${}^{2}J_{HF} = 47.1$)	-68.67 (t, $J_{\rm FH} = 47$)
4c ^b	5.26 (dd, J = 18.5/10.2)	8.57 (br.d, $J = 10$)	1.35, 1.39 (both t, 6 H, 2 C \underline{H}_3 CH ₂ , $J = 7$); 4.2 (m, 4 H, 2 OCH ₂)	-77.2 (s)
4e ^b	5.52 (dd, J = 18.6/10.5)	8.03 (d, J = 10.5)	1.29, 1.32 (both t, 6 H, 2 Me, $J = 7.5$); 4.2 (m, 4 H, 2 OCH ₂); 7.25-7.45 (m, 3 H, Ar); 7.66 (d, 1 H, Ar, $J = 9$)	_
4f°	5.95 (dd, J = 6.5/10.8); 5.73 (d, $J = 6/$)	8.03 (d, J = 10.8); 8.8 (br)	1.63, 1.91 (both s, the 5 : 1 ratio, Me); 7.3-8.1 (m, 10 H, Ph)	_
4 g ^{<i>a</i>}	6.02 (dd, J = 6.3/10.5)	8.23 (d, J = 10.5)	4.60, 4.82 (both dd, CH ₂ , AB system, $J_{H_AH_B} = 14.4$, ${}^{2}J_{HF} = 47$); 7.5–7.7 (m, 6 H, Ph); 7.9–8.1 (m, 4 H, Ph)	-66.61 (t, $J_{\rm FH} = 47$)
4h ^a	5.70 (dd, J = 4.5/10)		7.5-8.0 (m, 11 H, Ph, NH)	-75.0 (s)
4i ^b	6.08 (dd, J = 5/10.5)		7.5-8.0 (m, 15 H, Ph, NH)	
4m ^a	5.0 (m, J = -/10.5)	7.36 (d, $J = 10.5$)	1.30, 1.33 (both t, 6 H, 2 CH ₃ CH ₂ , $J = 7$); 3.69 (s, 3 H, MeO); 4.2 (m, 4 H, 2 OCH ₂); 6.71 (tt, 1 H, CHF ₂ , ² $J_{HF} = 51$, ³ $J_{HF} = 5.5$)	-136.8 (m, 2 F); $-130.2(1 F); -128.8 (1 F); -123.0(2 F); -115.4, -111.6 (m,2 F, CF_{A}F_{B}, J_{AB} = 290)$
4q ⁶	5.5 (m)	5.78 (d, J = 10.5)	1.02 (t, 3 H, Me, $J = 7$); 3.91 (q, 2 H, CH ₂ , $J = 7$); 6.00 (tt, 1 H, CHF ₂ , $J_{HF} = 52$, ${}^{3}J_{HF} = 5.5$); 7.6 (m, 6 H, Ph); 7.9 (m, 4 H, Ph)	_
7 a	4.5 (m)	4.95 (m, J = 11)	1.2–1.4 (m, 12 H, 4 Me); 3.9–4.3 (m, 8 H, 4 CH ₂); 6.73 (tt, 1 H, CHF ₂ , ${}^{2}J_{HF} = 51$, ${}^{3}J_{HF} = 5.5$)	-139.3 to -139.8 (m, 2 F); -131.3 (s, 2 F); -124.1, -122.2 (m, 2 F, CF_AF_B , $J_{AB} = 300$); -115.2, -112.0 (m, 2 F, CF_AF_B , $J_{AB} = 284$)
9 <i>a</i>	3.82 ($J = 12.3/6$)	9.4 (br)	1.30 (t, 6 H, 2 Me, $J = 7$); 4.15 (quint, 4 H, 2 OCH ₂ , $J_{\text{HP}} \approx J_{\text{HH}} \approx 7$)	-

Table 2. The parameters of the 1 H and 19 F NMR spectra of compounds 4, 7, and 9

^a In acetone-d₆.

^b In CDCl₃.

^c In DMSÖ-d₆.

signals for a minor isomer were also observed in the ¹H and ³¹P NMR spectra (cf. Ref. 9). The presence of a chiral center in compounds 4 and 7 predetermines the splitting of signals for the diastereotopic groups (EtO, CH₂, and CF₂) in the ¹H and ¹⁹F NMR spectra (see Table 2). When hydrogen chloride is eliminated, the chiral center disappears and that is why vinyl phosphonates **12** are not diastereotopic.

Thus, the reaction of N-(α -hydroxypolyhaloalkyl)amides with trivalent phosphorus chlorides is a convenient method of synthesis of N-acylated and N-phosphory-lated derivatives of α -aminopolyhaloalkylphosphonic acids and phosphinoxides. This approach successfully supplements the method developed in recent years¹²⁻¹⁴ and based on the reaction of carbonyl compounds with carboxamides and P^{III} chlorides because the latter method is unsuitable for α -halocarbonyl compounds.¹² It is also

significant that despite the apparent similarity of these processes, they differ principally. Detailed investigations suggest that it is nucleophilic "activated" derivatives of the Cl₂POH, Cl₂POAc, ClP(OH)₂, and H₃PO₃ types formed *in situ* from P^{III} chlorides and acetic acid that take an important part in the latter case, as in the reactions of hydroxymethylamides RCONHCH₂OH with PCl₃, ^{13,14}

Experimental

¹H, ¹⁹F, and ³¹P NMR spectra were recorded on a Varian VXR-300 spectrometer (299.95, 282.20, and 121.42 MHz, respectively). Chemical shifts are given with respect to the tetramethylsilane (δ^{-1} H) and CFCl₃ (δ^{-19} F) as the internal standards and 85% H₃PO₄ (δ^{-31} P) as the external standard. IR spectra were recorded on a UR-20 instrument (KBr or thin film).

N-(1-Hydroxypolyfluoroalkyl)amides I (R = H(CF₂)₄ and H(CF₂)₆) have been described by us earlier.^{7,16} N-(1-Hydroxy-trichloroethyl)amides were synthesized according to the known procedure.⁶

N-(1-Hydroxy-2,2,2-trichloroethyl)fluoroacetamide, yield 72%, m.p. 98–100 °C (from benzene). Found (%): Cl, 47.27. C₄H₅Cl₃FNO₂. Calculated (%): Cl, 47.39. ¹H NMR (acetone-d₆), δ: 4.94 (d, 2 H, ²J_{HF} = 47.1 Hz); 5.98 (d, 1 H, ³J_{H-NH} = 8.7 Hz); 6.35 (br.s, OH); 7.84 (br.d, NH).

N-(1-Hydroxy-2,2,2-trichloroethyl)-2-chlorobenzamide, yield 75%, m.p. 148-149 °C. Found (%): Cl, 46.59. C₉H₇Cl₄NO₂. Calculated (%): Cl, 46.81. IR (KBr), v/cm^{-1} : 1670 (C=O); 3100, 3280 (NH, OH).

The reaction of hydroxyamides 1 with trivalent phosphorus chlorides (general procedure). A solution of chloride 2 (10 mmol) was added to a stirred solution of compound 1 (10 mmol) and Et₃N (12 mmol) in ether or benzene at 10---15 °C. The reaction mixture was stirred at 20 °C for 3 h. After 12 h, the precipitate that formed was filtered off, and the filtrate was washed with water and dried over MgSO₄. The solvent was removed and the residue was washed with petro-leum ether and purified by recrystallization. If the phosphorus containing product precipitate was washed on the filter with water and dried to give compound 4. Compounds 4d,j,k,a-m and 5 have been described in previous reports.^{7,8} The characteristics of the new compounds are given in Tables 1 and 2.

0,0-Diethyl 1-ethoxycarbonylamino-2,2-dichlorovinyl phosphonate (12a). Et₃N (4 mmol) was added to a benzene solution of phosphonate **4d** (3 mmol) and stirred at 40-60 °C for 10 h. The precipitate of Et₃N · HCl was filtered off, and the filtrate was washed with water and dried over MgSO₄. The solvent was evaporated to give compound **12a** (liquid), yield 89%. Found (%): Cl, 21.56; P, 9.46. C₉H₁₆Cl₂NO₅P. Calculated (%): Cl, 22.15; P, 9.68. IR (thin film), v/cm⁻¹: 1070 (POC); 1275 (P=O); 1590 (C=C); 1750 (C=O); 3270 (NH). ¹H NMR (CDCl₃), δ : 1.28 (t, 3 H, Me, J = 7 Hz); 1.37 (t, 6 H, 2 Me, J = 7 Hz); 4.2 (m, 6 H, 3 OCH₂); 6.74 (br.d, 1 H, NH, ³J_{HP} = 2.5 Hz). ³¹P NMR (CDCl₃), δ : 9.15.

Bis(diethylamido)-1-ethoxycarbonylamino-2,2-dichlorovinyl phosphonate (12b) was obtained similarly, yield 78%, m.p. 103–104 °C. Found (%): CI, 18.57; P, 8.37. $C_{13}H_{26}CI_2N_3O_3P$. Calculated (%): CI, 18.95; P, 8.28. ¹H NMR (acetone-d₆-CCl₄, 1 : 1), δ : 1.12 (t, 12 H, 4 CH₃CH₂N, J = 7 Hz); 1.36 (t, 6 H, 2 CH₃CH₂O, J = 7 Hz); 4.10 (q, 2 H, OCH₂, J =7 Hz); 6.99 (br.s, 1 H, NH). ³¹P NMR (CDCl₃), δ : 21.7, 23.4 (10 : 1). 0,0-Diethyl 1-acetylamino-2,2-dichlorovinyl phosphonate (12c) was obtained similarly, liquid, yield 60%. Found (%): Cl, 24.98; P, 10.32. C₈H₁₄Cl₂NO₄P. Calculated (%): Cl, 24.44; P, 10.68. ¹H NMR (acetone-d₆-CCl₄, 1 : 1), δ : 1.31 (t, 6 H, 2 CH₃CH₂, J = 7 Hz); 1.96 (s, 3 H, MeCO); 4.08 (quint, 4 H, 2 OCH₂, ${}^{3}J_{HH} \approx {}^{3}J_{HP} \approx 7$ Hz); 8.84 (br.s, 1 H, NH). ³¹P NMR, δ : 9.64.

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