

at 215° (dec.) $\lambda_{\text{max}}^{\text{Nujol}}$ 3.04 μ (NH); 3.75 μ (OH); 6.28s, 6.62s, 13.54 μ s (*o*-disubst. phenyl).

Anal. Calcd. for $\text{C}_{14}\text{H}_{16}\text{N}_2\text{O}_2$: C, 68.8; H, 6.6; N, 11.5. Found: C, 68.8; H, 6.2; N, 11.6.

XIV was converted to *N,N'*-ethylenebis(2'-hydroxyacetanilide), diacetate, by warming with an excess of acetate-acetic anhydride. The solid, obtained by extraction with water, was recrystallized from benzene to give colorless prisms, m.p. 171–173°.

Anal. Calcd. for $\text{C}_{22}\text{H}_{24}\text{N}_2\text{O}_6$: C, 64.1; H, 5.9; N, 6.8. Found: C, 63.9; H, 6.2; N, 6.9.

Treatment of XIV with 2 mols. of acetate-acetic anhydride gave *N,N'*-ethylenebis-*o*-hydroxyacetanilide, m.p. 242–243° (dec.), as tan crystals from alcohol. This same product could be obtained by careful alkaline hydrolysis of the diacetate.

Anal. Calcd. for $\text{C}_{18}\text{H}_{20}\text{N}_2\text{O}_4$: C, 65.8; H, 6.1; N, 8.5. Found: C, 65.5; H, 6.3; N, 8.6.

N,N'-Ethylenebis-(*N*-*o*-hydroxyphenyl)glycine, di- δ -lactone (XVI). An alkaline solution of 1 g. of XIV was refluxed with an excess of sodium chloroacetate under nitrogen for 2 hr. until the pH was about 7. The solution was cooled, the pH adjusted to 5 with hydrochloric acid, and the solid collected and combined with an additional crop obtained on concentrating the filtrate; total yield 1.2 g. of buff powder, m.p. 160–170°. An analytical sample, obtained as very fine white needles from alcohol, melted at 176–178°. It was insoluble in water and acid and slowly soluble in dilute base. $\lambda_{\text{max}}^{\text{Nujol}}$ no NH or OH; 5.66s (δ -lactone- γ,δ -unsatd.).

Anal. Calcd. for $\text{C}_{18}\text{H}_{18}\text{N}_2\text{O}_4$: C, 66.7; H, 5.0; N, 8.6. Found: C, 66.5; H, 5.2; N, 8.8.

N-Methyl-phenmorpholone-2 (XXI). A solution of 0.5 g. (0.004 mol.) of *N*-methyl-*o*-aminophenol (prepared by the method of Clark²⁶) and 0.5 g. (0.005 mol.) of chloroacetic acid was maintained at pH 8 by the addition of 10% sodium hydroxide solution, while refluxed under nitrogen. After 1 hr.

the clear solution was cooled, acidified, and refrigerated overnight. The precipitated tan plates weighed 0.2 g. and melted at 51–52°. $\lambda_{\text{max}}^{\text{Nujol}}$ no NH or OH; 5.62s (δ -lactone, γ,δ -unsatd.).

Anal. Calcd. for $\text{C}_9\text{H}_9\text{NO}_2$: N, 8.6. Found: N, 8.4.

N-Methyl-*N*-formyl-*o*-aminophenoxyacetic acid (XXXII). This was prepared by the usual procedure from 1.5 g. (0.01 mol.) of *N*-methyl-*o*-hydroxyformanilide (m.p. 108–109°, lit.,²⁶ 103–104°), 0.94 g. (0.01 mol.) of chloroacetic acid and 0.08 g. (0.02 mol.) of caustic in 20 ml. of water. Acidification yielded 0.8 g. of crude product, m.p. 157–163°, and 0.5 g. of starting material was isolated from the filtrate. Recrystallization from 25 ml. of boiling water (charcoal) gave 0.5 g. (ca. 30%) of white microcrystalline powder, m.p. 174–174.5°. $\lambda_{\text{max}}^{\text{Nujol}}$ no NH or OH; 5.72s (COOH); 6.16s (amide I); 6.30s (?).

Anal. Calcd. for $\text{C}_{10}\text{H}_{11}\text{NO}_4$: C, 57.4; H, 5.3; N, 6.7. Found: C, 57.6; H, 5.4; N, 6.6.

XXXII was converted to *N*-methyl-phenmorpholone-3 (XXXIII) by dissolving in warm 1*N* hydrochloric acid and allowing the product to crystallize. The white needles had m.p. 59.5–60° (lit.²⁶ 58–59°).

N-Acetyl-phenmorpholone-3 (XXVIII). One g. of *N*-acetyl-*o*-aminophenoxyacetic acid²⁴ (XXVII) was boiled briefly with 2 g. of acetic anhydride, cooled, and treated with 15 ml. of water. The oily product crystallized on standing and after recrystallization from 50 ml. of alcohol consisted of 0.7 g. (80%) of fine white needles, m.p. 79–79.5° (lit.²³ 77°).

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[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, OHIO UNIVERSITY]

Substituted Aryl Phosphonic and Phosphinic Acids^{1,2}

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A number of new, substituted arylphosphonic and phosphinic acids have been prepared from the corresponding anilines. These include the 2,5-dibromophenyl-, 2-bromo-3-nitrophenyl-, 2,3-dichlorophenyl-, 3,5-dichlorophenyl-, 2,3,6-trichlorophenyl-, and 2,4,5-trichlorophenylphosphonic and phosphinic acids. Ethyl esters of several such acids are also reported. These compounds are being tested for plant-growth activity.

Halogen-substituted benzoic acids have been examined in detail as plant growth substances and factors relating structure and growth activity have

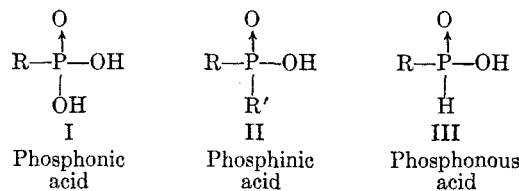
(1) Taken in part from the M.S. thesis of Joseph M. Denham, Ohio University, 1956.

(2) This work was supported by a contract with the U. S. Army Chemical Corps., Fort Detrick, Frederick, Md.

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been suggested.^{3–5} It was of interest to prepare and test the phosphonic and phosphinic acid analogs of the active benzoic acids; the present paper reports initial investigations of this problem.

To date, there has been no report of the testing of halogen-substituted arylphosphorus acids as plant growth substances. Maguire and Shaw have reported the preparation and testing of 2,4-dichloro-

phenoxyethylphosphonic acid⁶ and of 2,4-dichlorophenoxyethylphosphonous acid⁷ (phosphorus analogs of 2,4-D). Also, 3-indolemethylphosphonic acid,⁸⁻¹⁰ and unsubstituted phenyl- and naphthyl-phosphorus acids^{8,9} have been reported.

The substituted phosphonic acid phosphinic acids were prepared *via* the diazonium salts by the method of Doak and Freedman.¹¹ The use of a two-to-one ratio of diazonium salt to phosphorus tribromide, rather than the more usual one-to-one ratio, should be more favorable for the formation of the desired phosphinic acids. Both ratios were run for the preparation of the 2,5-dichlorophenyl derivatives; using a two-to-one ratio, the yield of phosphinic acid was nearly doubled and the yield of phosphonic acid increased approximately fifty per cent over the yields obtained with a one-to-one ratio. A similar change of ratio of reactants in the preparation of arsonic and arsinic acids has been stated to have no significant effect on the yields obtained.¹² The two-to-one ratio has been employed in the work herein reported with the hope of obtaining maximum amounts of the phosphinic acids.

All of the compounds reported here are new, with three exceptions. *m*-Tolylphosphonic acid¹³ and 2,5-dichlorophenylphosphonic acid¹⁴ have been previously reported but are included in the tables because of a significant difference in melting point. *m*-Iodophenylphosphonic acid was prepared earlier by a less direct method.¹⁵

It is of interest that, contrary to expectation, the 2,4,5-trichlorophenylphosphonic acid has a higher melting point than the bis(2,4,5-trichlorophenyl)-phosphinic acid.

Preliminary tests indicate that there is no correlation between growth activities of the phosphorus acids and of the corresponding benzoic acids. For example, the phosphorous analog of the markedly active 2,3,6-trichlorobenzoic acid is inactive; also, bis(*m*-bromophenyl)phosphinic acid shows considerable activity while the *m*-halobenzoic acids are

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inactive. Detailed biological results will be published elsewhere by Dr. R. L. Weintraub, U. S. Army Chemical Corps, Fort Detrick, Frederick, Md.

EXPERIMENTAL¹⁶

Typical procedures for each of the general types of reactions are described below.

3,5-Dichlorobenzene diazonium fluoborate. A method similar to that of Roe¹⁷ was used for the preparation of all diazonium compounds. To a cold mixture (0-10°) of 62 ml. of concentrated hydrochloric acid, 120 ml. of water, 41.5 g. (0.38 mole) of sodium fluoborate, and 48.6 g. (0.30 mole) of 3,5-dichloroaniline in a 1-liter beaker was added, dropwise, a solution of 20.7 g. (0.30 mole) of sodium nitrite in 40 ml. of water. A broad-bladed stirrer was used to ensure effective stirring of the continually-thickening mixture. The precipitate was collected on a filter and washed with 50 ml. of cold 5% fluoboric acid, 50 ml. of cold methanol, and several 60-ml. portions of ethyl ether to give, when dry, 45 g. (58%) of the diazonium salt. The product was dried by spreading it thinly over filter paper supported on a wire screen and allowing it to stand overnight; it was used in the next reaction without further purification.

3,5-Dichlorophenylphosphonic acid (Table I). The following method¹¹ was used for the preparation of all derivatives in Tables I and II; modifications were introduced when necessary.

A mixture of 250 ml. of absolute ethyl acetate and 45.0 g. (0.17 mole) of 3,5-dichlorobenzene diazonium fluoborate was placed in a 2-liter, 3-necked flask. The flask was fitted with a mechanical stirrer and a reflux condenser containing a wide-bore delivery tube leading to a gas trap. With stirring, 23.3 g. (0.086 mole) of phosphorus tribromide was added, followed by 2.0 g. of cuprous bromide. The mixture changed from a red to a violet color and foamed vigorously for a brief period. The foaming was held under control by application of an ice bath and soon subsided. Stirring for an additional 30 min. at room temperature and heating to 50° for 5 min. failed to produce evidence of further reaction. The mixture was cooled to room temperature and hydrolyzed by the dropwise addition of 25 ml. of water.

The ethyl acetate was removed by steam distillation; when a separate water layer began forming, the receiver was changed and the steam distillation was continued until all steam-volatile, water-insoluble material ceased collecting in the distillate (approximately one liter). Filtration of the distillate yielded 6.0 g. of yellow solid melting at 74-75.5°. One recrystallization from ethanol raised the melting point to 75-76.5°. No depression of melting point occurred when this sample was mixed with authentic 3,5-dichlorobromobenzene.

A red oil had separated in the residual steam distillation mixture. The hot water layer was decanted through a filter and allowed to cool to give a pink precipitate of crude phosphonic acid. The precipitate was collected on a filter. The red oil, composed principally of phosphinic acid, hardened somewhat on cooling and solidified on extraction with 150 ml. of boiling 6*N* hydrochloric acid. The crude phosphonic acid was collected by filtration and this filtrate was added to the main filtrate. The combined filtrates were evaporated to approximately 50 ml. on a steam bath to obtain a second portion of crude phosphonic acid.

The pure phosphonic acid was isolated in two portions. The first was obtained by treating the pink solid obtained

(16) All melting points are uncorrected.

(17) A. Roe, *Org. Reactions*, **V**, 205 (1949). A brief previous mention of the 3,5-dichlorobenzene diazonium fluoborate appears in the literature (see Roe, *loc. cit.*, p. 217); a detailed procedure is included as typical of the fluoborates used in this work.

TABLE I
 SUBSTITUTED PHENYLPHOSPHONIC ACIDS^a

I, R =	Yield, %	M.P., °C.	Formula	Analysis			
				Halogen, %		Phosphorus, %	
				Calcd.	Found	Calcd.	Found
<i>m</i> -Tolyl- ^b	8.7	125-126	C ₇ H ₉ O ₃ P	—	—	18.0	17.38
<i>m</i> -Iodo- ^c	20	183-184	C ₆ H ₆ IO ₃ P	—	—	—	—
2,5-Dibromophenyl-	25	204-208	C ₆ H ₃ Br ₂ O ₃ ·H ₂ O	47.86	47.6	9.28	9.1
2-Bromo-3-nitrophenyl- ^{d,e}	12	224 (dec.)	C ₆ H ₃ BrNO ₂ P	28.35	28.0	10.99	10.8
2,3-Dichlorophenyl- ^f	14	200-202	C ₆ H ₃ Cl ₂ O ₃ P·H ₂ O	28.94	29.5	12.64	12.4
3,5-Dichlorophenyl-	19 ^g	188-190	C ₆ H ₃ Cl ₂ O ₃ P	31.24	31.69	13.65	13.80
2,3,6-Trichlorophenyl- ^e	^h	203-204	C ₆ H ₃ Cl ₃ O ₃ P	40.69	40.24	11.85	11.5
2,4,5-Trichlorophenyl-	19 ⁱ	252-254	C ₆ H ₃ Cl ₃ O ₃ P	40.69	40.36	11.85	11.64

^a All of these acids were recrystallized from 6*N* hydrochloric acid; the *m*-tolylphosphonic acid was also recrystallized from benzene. ^b Reported m.p. 121° (see ref. 13) and 116-117° [see A. Michaelis, *Ann.*, **293**, 261 (1896)]. ^c Prepared previously *via* nitration of phenylphosphonic acid (see ref. 15). ^d Calcd. for N: 4.97; found: 4.95. ^e The required aniline was prepared from the corresponding benzoic acid *via* the Hofmann reaction. ^f Also dried to constant weight for analysis. Calcd. for C₆H₃Cl₂O₃P: Cl, 31.24; P, 13.65. Found: Cl, 31.28; P, 13.41. ^g Yield based on product melting at 186.5-190°. ^h Only 0.25 g. was obtained, with a theoretical yield of 0.064 mole. ⁱ The initial reaction was very vigorous, resulting in some loss of product; yield based on product melting at 250-252°.

 TABLE II
 SUBSTITUTED PHENYLPHOSPHONIC ACIDS

II, R = R' =	Yield, %	M.P., °C.	Solvent for Recrystn.	Formula	Analysis			
					Halogen, %		Phosphorus, %	
					Calcd.	Found	Calcd.	Found
<i>m</i> -Tolyl-	5.5	166-168	EtOH-H ₂ O	C ₁₄ H ₁₆ O ₂ P	—	—	12.58	12.33
<i>m</i> -Iodophenyl-	8.0	212.5-213.5	EtOH-H ₂ O	C ₁₂ H ₉ I ₂ O ₂ P	54.01	53.51	6.59	6.61
2,5-Dibromophenyl-	1.6	277-279	EtOAc	C ₁₂ H ₇ Br ₂ O ₂ P	59.88	60.2	5.80	5.8
2-Bromo-3-nitrophenyl- ^a	5.4	296-297 (dec.)	AcOH	C ₁₂ H ₇ Br ₂ N ₂ O ₆ P	34.30	34.22	6.65	6.11
2,3-Dichlorophenyl-	9.3	278-280	EtOH-H ₂ O	C ₁₂ H ₇ Cl ₂ O ₂ P	39.84	39.82	8.70	8.66
2,5-Dichlorophenyl- ^b	16	221-222	EtOH-H ₂ O	C ₁₂ H ₇ Cl ₂ O ₂ P	39.84	39.56	8.70	8.6
3,5-Dichlorophenyl-	13 ^c	243-244.5	EtOH-H ₂ O	C ₁₂ H ₇ Cl ₂ O ₂ P	39.84	39.81	8.70	8.76
2,3,6-Trichlorophenyl-	6.4	287-288.5	EtOH	C ₁₂ H ₅ Cl ₃ O ₂ P	50.07	49.57	7.29	7.25
2,4,5-Trichlorophenyl-	11 ^d	244.5-246	EtOH-H ₂ O; EtOAc	C ₁₂ H ₅ Cl ₃ O ₂ P	50.07	49.97	7.29	7.35
R = Phenyl- R' = <i>m</i> -Bromophenyl- ^e	50	161.5-162	EtOH-H ₂ O	C ₁₂ H ₁₀ BrO ₂ P	26.90	27.04	10.43	10.35

^a Dried to constant weight for analysis. Calcd. for N: 6.01; found: 5.92. ^b This compound, m.p. 232-233°, has been prepared by Freedman and co-workers (see ref. 14); additional recrystallization did not change the melting point. ^c Yield based on product recrystallized from aqueous ethanol, m.p. 241-243°. ^d Yield based on product m.p. 240-242°, recrystallized from aqueous ethanol. ^e Phenylphosphine dichloride used in place of phosphorus tribromide; yield based on product melting at 160-161°.

from the cooled water layer of the steam distillation mixture with 20% sodium hydroxide and isolation of the hemi-sodium salt. The salt was precipitated by acidifying the alkaline solution to the Congo Red point with concentrated hydrochloric acid. The alkaline solution was filtered prior to acidification and, if necessary, was decolorized with charcoal. Two recrystallizations from 6*N* hydrochloric acid gave 1.5 g. of 3,5-dichlorophenylphosphonic acid as pink needles melting at 186.5-190°.

The base treatment separated a tan oil which solidified on standing. Recrystallization from aqueous ethanol gave 1.4 g. of white needles melting at 50-51°; no depression of melting point occurred when the solid was mixed with an authentic sample of 3,5-dichloroaniline.

The second portion of pure phosphonic acid was isolated by treating in a similar manner the residue obtained from the evaporated filtrates. Two recrystallizations gave 2.15 g. of pink needles melting at 186.5-190°. The combined yield was 3.65 g. (19%, based on a theoretical yield of 0.086 mole).

Two additional recrystallizations from 6*N* hydrochloric acid produced an analytical sample as colorless needles melting at 188-190°.

Bis(3,5-dichlorophenyl)phosphinic acid (Table II). The crude phosphinic acid from the above preparation was dissolved in dilute sodium hydroxide solution. The alkaline solution was decolorized with charcoal and filtered, and the filtrate was acidified with concentrated hydrochloric acid to isolate the free acid; the acid was then recrystallized from aqueous ethanol. The first two treatments with water produced a trace of amorphous solid; the third gave 5.2 g. of pink-to-red crystals melting at 239-240.5°. Extraction with 30 ml. of warm ethyl acetate left 4.0 g. (13%, based on a theoretical yield of 0.086 mole) of bis(3,5-dichlorophenyl)phosphinic acid melting at 241-243°. An additional recrystallization from aqueous ethanol and a final recrystallization from absolute ethanol produced an analytical sample as pink, hexagonal platelets melting at 243-244.5°.

Several other phosphonic and phosphinic acids were prepared in this manner and the results are listed in Tables I and II. Variations in the above procedure were necessary with two of the acids and these modifications are given below. In most cases only a single run was made and, therefore, the yields probably are not optimum.

2,3,6-Trichlorophenylphosphonic acid (Table I). A very small quantity of crude phosphonic acid was isolated as

TABLE III
ETHYL ESTERS OF SUBSTITUTED PHENYLPHOSPHONIC AND PHENYLPHOSPHINIC ACIDS

	Yield, %	B.P., °C.	Pres- sure, Mm.	n_D^{25}	Formula	Analysis			
						Halogen, %		Phosphorus, %	
						Calcd.	Found	Calcd.	Found
Phosphonate Esters									
<i>m</i> -Chlorophenyl-	41	101	0.25	1.5005	C ₁₆ H ₁₄ ClO ₂ P	14.26	14.74	12.46	12.12
<i>m</i> -Bromophenyl-	30	109-113	0.22	1.5200	C ₁₆ H ₁₄ BrO ₂ P	27.26	27.19	10.57	10.42
2,5-Dibromophenyl-	26	134-139	0.30	1.5475	C ₁₆ H ₁₂ Br ₂ O ₂ P	42.97	42.38	8.33	8.19
Phosphinate Esters									
Bis(<i>m</i> -chlorophenyl)-	48	183-187	0.70	1.5794	C ₁₄ H ₁₂ Cl ₂ O ₂ P	22.50	22.66	9.83	9.51
Phenyl- <i>m</i> -chlorophenyl-	58	170-174	0.60	1.5701	C ₁₄ H ₁₂ ClO ₂ P	12.63	12.04	11.03	11.14

above from the steam distillation mixture. Attempts to prepare the hemi-sodium salt gave only a green colored, gummy mass; thus, excess hydrochloric acid was added to effect solution of the product. The solution was evaporated to dryness and the residue then extracted with acetone. Removal of the acetone left a residue that gave, after recrystallization from 6*N* hydrochloric acid, 0.25 g. of 2,3,6-trichlorophenylphosphonic acid as white needles.

Bis(2,4,5-trichlorophenyl)phosphinic acid (Table II). The residue containing the phosphinic acid, isolated in the usual manner from the steam distillation mixture, failed to dissolve appreciably in 100 ml. of 10% sodium hydroxide solution. The mixture was diluted with one liter of water and heated to reflux. The boiling mixture was treated with charcoal and filtered through a heated funnel to prevent crystallization of the salt. The free acid was isolated by acidification with concentrated hydrochloric acid.

m-Chlorophenylphosphonyl dichloride.¹⁸ To 10.0 g. (0.052 mole) of *m*-chlorophenylphosphonic acid in a 200-ml. flask fitted with condenser and drying tube, was added 24.1 g. (0.10 mole plus 10% excess) of phosphorus pentachloride. The solids fused after a period of 15 min. and rapid refluxing followed. When the reaction subsided, the mixture was warmed to maintain refluxing for 30 min. and then allowed to stand at room temperature overnight. The phosphorus oxychloride and excess phosphorus pentachloride were removed at water pump pressure and a water bath temperature of 60°. The residue was vacuum distilled to give 10.1 g. (84%) of colorless liquid, boiling at 86-89° (0.4 mm.). The acid chloride was used in the ester preparation without further purification.

m-Bromophenylphosphonyl dichloride (b.p. 94-100°/0.25 mm.) and 2,5-dibromophenylphosphonyl dichloride (m.p. 107-114°, without purification) were prepared in a similar manner in yields of 92% and 85%, respectively. Bis(*m*-chlorophenyl)phosphinyl chloride and phenyl-*m*-chlorophenylphosphinyl chloride were prepared using a one-molar ratio of phosphorus pentachloride plus 10% excess. The application of heat was required in some cases to initiate the reaction of the acid with phosphorus pentachloride. The phosphinyl chlorides were used directly after removal of the phosphorus oxychloride and excess phosphorus pentachloride.

(18) Method of L. D. Freedman, *et al.*; see reference 14.

Diethyl m-chlorophenylphosphonate (Table III).¹⁹ A mixture of 2.0 g. (0.044 mole) of absolute ethanol, 3.9 g. (0.049 mole) of dry pyridine, and 20 ml. of absolute ethyl ether was added, in 3 portions, to a cooled solution of 5.0 g. (0.022 mole) of *m*-chlorophenylphosphonyl dichloride and 30 ml. of dry ether in a 100-ml. flask fitted with condenser and drying tube. Refluxing occurred after each portion was added and pyridinium chloride precipitated. The mixture was allowed to stand overnight and then filtered. The solvent was removed by distillation and the residue was vacuum distilled. A small amount of pyridinium chloride sublimed initially; this was removed from the column and the distillation continued to give 2.25 g. (41%) of diethyl *m*-chlorophenylphosphonate as a colorless liquid, b.p. 100-101° (0.30 mm.), n_D^{25} 1.5005.

The *m*-bromo-, 2,5-dibromo-, and phenyl-*m*-bromophenyl- derivatives were also prepared by this procedure; results may be found in Table III.

Ethyl bis(m-chlorophenyl)phosphinate (Table III). Bis(*m*-chlorophenyl)phosphinyl chloride, prepared from 5.0 g. (0.017 mole) of the phosphinic acid, was dissolved in 9 ml. of carbon tetrachloride. To this solution was added 1.5 ml. of absolute ethanol; some warming occurred on adding the ethanol. The mixture was refluxed for 2 hr. (hydrogen chloride was evolved) and the solvent and excess ethanol were removed by distillation at water-pump pressure. The residue was vacuum-distilled to give 1.75 g. of ethyl phosphate, b.p. 47-50° (0.7 mm.) and 1.45 g. (48%, based on amount of acid not recovered) of ethyl bis(*m*-chlorophenyl)phosphinate, b.p. 183-187° (0.7 mm.), n_D^{25} 1.5794.

The dark brown residue from the distillation was dissolved in 50 ml. of hot ethanol and decolorized with charcoal; dilution with water and cooling gave 2.1 g. (42% recovery) of white solid melting at 133-152°. Recrystallization from petroleum ether (b.p. 90-120°) brought the melting point to 162-165°; a mixture melting point with the starting acid showed no depression.

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(19) Method of M. I. Kabachnik, P. A. Rossiiskaya, and N. N. Novikova, *Bull. acad. sci., U.R.S.S., Classe sci. chim.*, 97 (1947) [*Chem. Abstr.*, 42, 4132 (1948)].