creased solubility as dipole moment increases), and van der Waals forces.

Although anion and cation combinations may have different effects on s-triazine salting out, the solubility values obtained with the chloride salts indicate that salts in the soil solution (~0.02 N) would have an insignificant effect on solubility. Richards (1954) reported that saturated saline soils do not exceed 0.1 N salt concentration and that most agricultural soils have salt levels below 0.02 N.

It may be possible, however, that under the right management conditions, i.e., band application of fertilizers with herbicides and low-moisture conditions, salt levels may become high enough to precipitate the s-triazines. The condition would probably be relieved upon application of water.

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Fate of the Insecticide *O*-[4-[(4-Chlorophenyl)thio]phenyl] *O*-Ethyl *S*-Propyl Phosphorothioate (RH-0994) in Water

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The stability of a radiolabeled preparation of the organic phosphate insecticide *O*-[4-[(4-chlorophenyl)thio]phenyl] *O*-ethyl *S*-propyl phosphorothioate (RH-0994) was determined in buffered water, in the dark, at pH 4.0, 7.0, and 10.0. RH-0994 degraded rapidly at neutral or alkaline pH but much more slowly under acidic conditions. The half-lives of 0.5-ppm solutions of RH-0994 under the conditions of study were as follows: pH 10.0, <1 day; pH 7.0, ~14 days; pH 4.0, ≫28 days. Degradation occurred primarly by hydrolysis of the phosphorus–O–phenyl ester linkage and by oxidation of the diphenyl thioether sulfur to sulfoxide derivatives.

There is a continual need for the development of insect control agents that are both efficacious and highly selective in toxicity toward pest species. Further, as pest insects acquire resistance to existing insecticides, the development of newer chemicals that circumvent resistance becomes a highly desirable and in fact critical need.

Certain major insect pests of cotton, such as the cotton bollworm, *Heliothis zea* (Boddie), and the tobacco budworm, *Heliothis virescens* (F.), have developed a high degree of resistance to many of the insecticides used against them for the past two or more decades. Due to resistance, control strategies for these insects must be adaptable as the insects themselves change. The *Heliothis* complex in cotton is presently managed by a number of approaches, including continued reliance on older insecticides, the use of newer chemicals such as the synthetic pyrethroids, and various integrated pest management (IPM) techniques. There is little doubt that management of the *Heliothis* complex in cotton will, in the future, require further refinements; thus, research toward the development of newer and more efficacious insecticides is clearly warranted.

The experimental organic phosphate insecticide *O*-[4-[(4-chlorophenyl)thio]phenyl] *O*-ethyl *S*-propyl phosphorothioate (RH-0994 of the Rohm and Haas Co., Philadelpha, PA) is being developed for possible use in controlling *Heliothis* populations, as well as other insect species. The compound has good selectivity, particularly in its relatively low toxicity to mammals (Bull and Ivie, 1981). RH-0994 thus offers promise as an environmentally acceptable insecticide for use against *Heliothis* and other insects attacking cotton and other crops. As a necessary prerequisite to its potential commercial use, environmental studies are needed. We have previously reported on the fate of RH-0994 in cotton (Bull and Ivie, 1981); the current studies were designed to evaluate the fate of the chemical in water, in the dark, under acidic, neutral, and alkaline conditions.

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MATERIALS AND METHODS

Chemicals. [^{14}C]RH-0994 (6.15 mCi/g) was supplied for these studies by the Rohm and Haas Co., Spring House, PA. The radiochemical was uniformly incorporated into

Table I. Thin-Layer Chromatographic (TLC) Behavior of the Organophosphorus Insecticide RH-0994 and Certain of Its Analogues[a]

product[b]	R_f in solvent system[c]							
	1	2	3	4	5	6	7	8
RH-0994	0.58	0.66	0.73	0.45	0.65	0.79	0.44	0.75
RH-0994 sulfoxide	0.33	0.27	0.64	0.11	0.49	0.76	0.13	0.64
RH-0994 sulfone	0.50	0.54	0.73	0.28	0.54	0.78	0.31	0.69
phenol	0.39	0.71	0.59	0.46	0.60	0.64	0.54	0.73
phenol sulfoxide	0.16	0.31	0.32	0.09	0.42	0.53	0.16	0.60
phenol sulfone	0.21	0.55	0.50	0.21	0.48	0.59	0.33	0.65

[a] 0.25 mm thick silica gel chromatoplates, Brinkman Silplate F-22, with fluorescent indicator. [b] Structures of these compounds are shown in Figure 1. [c] Solvent systems as follows: (1) benzene-ethanol-acetic acid (93:7:1), (2) ether-acetic acid (99:1), (3) methylene chloride-methanol (20:1), (4) benzene (saturated with formic acid)-ether (5:1), (5) hexane-acetone-acetic acid (50:50:1), (6) chloroform-ethanol (10:1), (7) ether-hexane-acetic acid (60:20:1), and (8) hexane-ethyl acetate-methanol-acetic acid (20:20:10:1).

the phosphorus–O–phenyl ring. Also provided by Rohm and Haas were samples of unlabeled RH-0994, its sulfoxide and sulfone analogues, the phenolic hydrolysis product of RH-0994, and the phenol sulfoxide and phenol sulfone. Chemical names for these compounds are found in a previous report (Bull and Ivie, 1981).

Exposure Media. Stability studies with [^{14}C]RH-0994 were done in the following three buffer systems, each prepared with glass distilled water: 0.01 M sodium acetate–acetic acid, pH 4.0; 0.01 M boric acid–sodium hydroxide, pH 7.0; 0.01 M boric acid–sodium hydroxide, pH 10.0. So that the possibility of microbial degradation of [^{14}C]RH-0994 during the exposure is minimized, the prepared buffers were autoclaved (121 °C; 30 min) prior to use.

Sample Preparation and Exposure. The radiolabeled RH-0994 sample as supplied was determined by thin-layer chromatographic (TLC) and radioautographic analysis to contain appreciable (~8%) radiochemical impurities; it was therefore purified on TLC (solvent system 1; vide infra) prior to use. The purified compound was eluted from the appropriate gel region with ether and was subsequently found to be of >99% radiochemical purity upon two-dimensional TLC analysis (solvent systems 1 and 2; vide infra).

Samples for exposure were prepared by adding 10 μg (approximately 140 000 dpm) of [^{14}C]RH-0994 in acetone solution to each of several 50 mL capacity glass ampules that had been heat sterilized. The acetone carrier was evaporated with a gentle stream of nitrogen, and then 20 mL of the appropriate buffer solution was added to each ampoule, giving an RH-0994 concentration equivalent to 0.5 ppm. The ampules were heat sealed, placed in a rack, and subjected to continuous gentle shaking in the dark. The sample temperature throughout the exposures was approximately 35 °C; the slight elevation above room temperature was caused by added heat from the electric shaker. Preliminary studies with the equivalent of 0.5 ppm of [^{14}C]RH-0994 added to each of the three buffers studied showed that after 1 h of continuous shaking, >80% of the ^{14}C added to each sample was apparently in solution or at least fine suspension, as evidenced by LSC of 0.2-mL aliquots. Thus, the parameters chosen here were considered acceptable for studies of the fate of [^{14}C]RH-0994 in water.

Extraction and Analysis. At appropriate intervals, samples were removed and extracted in the following manner: four ampules for each pH and exposure intervals were opened and the pH was adjusted to ~2.0 with 1.0 N HCl. To two of the four replicates was immediately added 25 μL of an acetone solution containing 30 μg each of unlabeled RH-0994 and its five degradation products (vide supra). These additions were made to facilitate characterization of the ^{14}C-labeled degradation products upon two-dimensional TLC chromatography of the extracts. The contents of each ampule were then transferred to a 50-mL screw-cap tube, the ampule was rinsed thoroughly with 15 mL of diethyl ether, and the ether was added to the appropriate aqueous sample. After partitioning, the ether was drawn off, and the aqueous phase was partitioned twice again with 15-mL volumes of ether. Radiocarbon in the aqueous and ether extracts was quantitated by liquid scintillation counting (LSC) of 0.2-mL aliquots, and then the ether fractions were dried over anhydrous sodium sulfate, concentrated under a gentle stream of nitrogen to near dryness, and analyzed by two-dimensional TLC.

Product Resolution and Characterization. Degradation products of [^{14}C]RH-0994 in water were resolved by two-dimensional TLC. The precoated silica gel plates used were Brinkman Silplate F-22, 20 × 20 cm, 0.25-mm gel thickness, with a fluorescent indicator. The concentrated sample extracts were spotted as a short (~2-cm) band in one corner of the plates, and then the plates were developed two dimensionally, first in a system consisting of benzene–ethanol–glacial acetic acid (93:7:1; solvent system 1) and then in the second dimension in a system consisting of ether-glacial acetic acid (99:1; solvent system 2). The developed plates were exposed for 2 weeks to medical X-ray film (Kodak No-Screen) for visualization of the ^{14}C-labeled components present.

Characterizations of the ^{14}C-labeled components in the sample extracts were made by comparing the TLC behavior of the radiolabeled components present with that of the added unlabeled compounds of known structure. The positions of the unlabeled compounds on the developed plates were determined by examination under short-wavelength ultraviolet light. Identical TLC behavior of a radiocarbon-labeled RH-0994 water degradation product and an unlabeled standard after two-dimensional TLC constituted tentative product characterization; however, all identifications were ultimately confirmed by additional TLC cochromatography studies in six additional solvent systems: (3) methylene chloride–methanol (20:1); (4) benzene (saturated with formic acid)–ether (5:1); (5) hexane–acetone–glacial acetic acid (50:50:1); (6) chloroform–ethanol (10:1); (7) ether–hexane–glacial acetic acid (60:20:1); (8) hexane–ethyl acetate–methanol–glacial acetic acid (20:20:10:1). TLC R_f values for RH-0994 and its analogues studied are shown in Table I.

The relative distribution of [^{14}C]RH-0994 water degradation products formed in individual samples was determined by sampling the appropriate gel regions after two-dimensional TLC and radioautography and subjecting them to direct LSC analysis. In these and other LSC measurements during this study, a toluene–2-methoxyethanol based scintillation cocktail was used, and correc-

Table II. Degradation Products of [^{14}C]RH-0994 after Exposure in the Dark as 0.5-ppm Solutions in Acidic, Neutral, and Alkaline Buffers[a]

pH	product[c,d] or fraction	% (±SE) product or fraction at the time interval[b] of						
		0 day	1 day	4 days	7 days	14 days	21 days	28 days
4.0	RH-0994	98.5 ± 0.1	94.2 ± 1.2	94.7 ± 0.0	95.7 ± 0.2	92.4 ± 0.3	92.0 ± 0.9	90.5 ± 0.9
	RH-0994 sulfoxide	0.5 ± 0.1	2.4 ± 0.9	2.2 ± 0.2	2.1 ± 0.2	4.0 ± 0.4	4.3 ± 0.3	4.0 ± 0.4
	phenol	0	0	0	0	0	0.3 ± 0.1	0.2 ± 0.0
	unknowns[e]	0.4 ± 0.2	0.1 ± 0.1	0.4 ± 0.4	0	0.8 ± 0.1	0.3 ± 0.1	0.7 ± 0.2
	water soluble[f]	0.6 ± 0.1	3.4 ± 0.3	2.8 ± 0.2	2.3 ± 0.1	2.9 ± 0.1	3.3 ± 1.2	4.5 ± 0.4
7.0	RH-0994	96.6 ± 1.4	92.2 ± 1.0	78.7 ± 0.8	70.0[h]	50.6 ± 1.3	39.1 ± 2.1	23.5 ± 2.0
	RH-0994 sulfoxide	2.9 ± 1.9	2.5 ± 1.6	1.8 ± 1.1	0.9[h]	1.5 ± 0.2	1.8 ± 0.1	1.6 ± 0.8
	phenol	0	4.3 ± 0.4	17.7 ± 2.4	27.3[h]	44.8 ± 2.0	54.3 ± 2.8	66.7 ± 4.8
	phenol sulfoxide	0	0.4 ± 0.1	1.1 ± 0.3	1.1[h]	2.8 ± 0.1	4.2 ± 0.3	6.1 ± 2.0
	unknowns[g]	0	0.5 ± 0.3	0.5 ± 0.0	0.6[h]	0.2 ± 0.2	0.6 ± 0.2	1.4 ± 0.5
	water soluble[f]	0.5 ± 0.4	0.2 ± 0.1	0.3 ± 0.2	0.1[h]	0.2 ± 0.2	0.2 ± 0.1	0.4 ± 0.1
10.0	RH-0994	98.0 ± 0.2	19.1 ± 1.8	15.5 ± 2.6	3.3 ± 0.7	1.0 ± 0.1	0.5 ± 0.3	0.4 ± 0.0
	RH-0994 sulfoxide	0.9 ± 0.3	1.5 ± 1.2	0.3 ± 0.2	0	0	0	0
	phenol	0.9 ± 0.2	70.9 ± 4.0	80.6 ± 2.8	92.8 ± 1.6	93.8 ± 1.6	78.8 ± 3.6	86.0 ± 1.2
	phenol sulfoxide	0.1 ± 0.0	7.6 ± 4.8	2.6 ± 0.1	1.9 ± 1.2	4.3 ± 1.4	18.5 ± 3.2	10.5 ± 1.0
	unknowns[i]	0	1.0 ± 0.3	0.8 ± 0.3	1.2 ± 0.3	0.3 ± 0.0	2.1 ± 0.4	2.6 ± 0.0
	water soluble[f]	0.1 ± 0.1	0.2 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	0.6 ± 0.2	0.4 ± 0.1	0.6 ± 0.2

[a] 10 μg of [^{14}C]RH-0994 in 20 mL of 0.01 M sodium acetate–acetic acid (pH 4.0), 0.01 M boric acid–sodium hydroxide (pH 7.0), or 0.01 M boric acid–sodium hydroxide (pH 10.0) in sealed glass ampules (50-mL capacity). [b] After exposure for the appropriate period, but prior to extraction and analysis, 30 μg each of authentic unlabeled RH-0994 and five of its potential degradation products (see the text) was added to each sample to facilitate characterization studies. [c] As resolved by two-dimensional TLC (see the text). [d] See Figure 1 for structures of products. [e] Cumulative totals for zero to three unidentified products, depending upon the sample. [f] Radiocarbon remaining in the aqueous fraction after extraction of the acidified samples. [g] Cumulative totals for zero to six unidentified products, depending upon the sample. [h] Data from one sample only. [i] Cumulative totals for zero to eight unidentified products, depending upon the sample.

tions were made for quench as appropriate.

RESULTS

Dilute aqueous solutions of RH-0994 degrade to several products in the dark, and the rate of degradation is highly dependent upon pH (Table II). In pH 4.0 buffer, RH-0994 is quite stable, and even after exposure for 28 days, only about 10% of the radiochemical is degraded. The major identified product is RH-0994 sulfoxide, but trace amounts of RH-0994 phenol are also observed. As many as three unidentified products are seen in these samples, but they do not collectively comprise >0.8% of the total radiocarbon in any sample (Table II).

At pH 7.0, [^{14}C]RH-0994 degrades at a moderate rate, with a half-life of ~14 days (Table II). The phenolic hydrolysis product is by far the major degradation product observed in the samples, but much lesser amounts of the phenol sulfoxide and RH-0994 sulfoxide are also generated. Water-soluble radiocarbon comprises a very minor part of the total, and although as many as six unidentified products are seen in certain samples, their contribution to the total residue is very small (Table II).

In contrast to the high stability of RH-0994 at pH 4.0 and its moderate stability at pH 7.0, RH-0994 degrades very rapidly at pH 10.0. The half-life of the compound under these parameters is ≪1 day, and within 7 days, the compound is almost totally degraded (Table II). As is seen at pH 7.0, the major route of degradation is ester hydrolysis to the phenol, but sulfur oxidation prior to hydrolysis (to RH-0994 sulfoxide) also occurs as a pathway of minor significance. Moderate amounts of the phenol sulfoxide are also detected in all samples. Radiocarbon not extracted from the aqueous phase and ^{14}C-labeled components not identified (as many as eight, depending upon the sample) comprise very small proportions of the total radiocarbon in all samples (Table II). Radiocarbon recovery did not vary significantly with respect to either pH of the exposure media or time of exposure, indicating that RH-0994 did not appreciably degrade to volatile products under the conditions of study.

Table II shows data from samples fortified, after exposure but prior to extraction and analysis, with 30 μg each of unlabeled RH-0994 and several of its analogues. The original intent of these additions was to facilitate TLC characterization of the ^{14}C-labeled components present. However, upon analysis of the data from these samples, and from others identical with them in every respect except for the absence of added unlabeled standards, it became obvious that chemical transformations of some of the ^{14}C-labeled components occurred during the extraction and analysis procedures and that the presence of unlabeled RH-0994 analogues suppressed these transformations. In particular, concentrations of RH-0994 and its phenol tended to be considerably lower, and levels of RH-0994 sulfoxide and the phenol sulfoxide higher, in the "unprotected" samples than in those with added unlabeled standards. These differences were often quite dramatic. For example, in samples exposed for 1 day at pH 7.0, analysis indicated the following percentage of the four identified components present: for unlabeled standards added (Table II), RH-0994, 92.2; RH-0994 sulfoxide, 2.5; phenol, 4.3; phenol sulfoxide, 0.4; for no standards added, RH-0994, 45.8; RH-0994 sulfoxide, 48.0; phenol, 1.2; phenol sulfoxide, 3.0. These data indicate that, although the extractions and analyses were conducted by quite conventional methods, RH-0994 and its phenolic hydrolysis product, particularly in low concentrations, underwent rather rapid sulfur oxidation to sulfoxide analogues. After completion of the studies reported here, Morelli (1980) confirmed that extraction of dilute aqueous solutions of RH-0994 with ether does in fact result in oxidation of the diphenyl thioether sulfur, a reaction that may be attributable to peroxide contaminants in the ether because the oxidation was much less pronounced when heptane was used as the extracting solvent (Morelli, 1980).

DISCUSSION

The degradation of RH-0994 in buffered water proceeds through straightforward and perhaps predictable pathways, as are indicated in Figure 1. The compound is quite

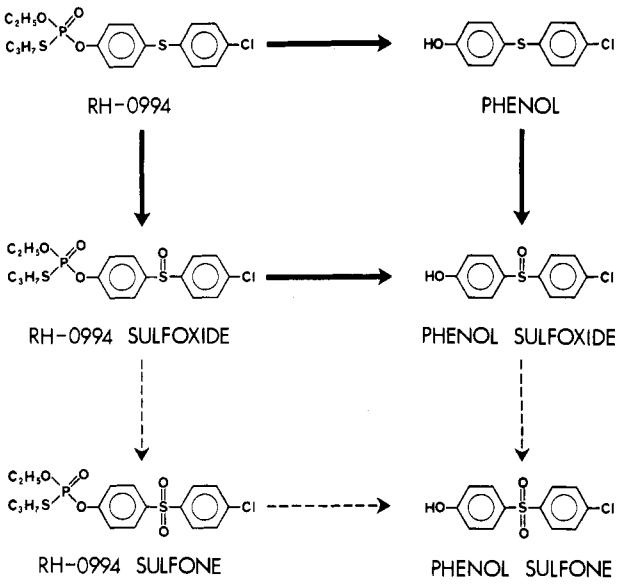

Figure 1. Degradation of RH-0994 in water in the dark. The RH-0994 sulfone and phenol sulfone analogues either were not formed under the conditions of study or were generated in amounts too low for detection.

stable under moderately acidic aqueous conditions but is rapidly degraded at alkaline pH, where hydrolysis of the phosphorus–O–phenyl ester linkage is the major initial degradation step (Table II). Many organic phosphate insecticides are known to be readily susceptible to base- but not acid-catalyzed ester hydrolysis (O'Brien, 1967); thus our data on the pH-dependent degradation of RH-0994 are not at all surprising.

The finding that RH-0994 and its phenolic hydrolysis product readily oxidized to sulfoxide analogues during our sample preparation and analysis procedures suggests that caution may be required in the development and utilization of analytical methods for this compound and its derivatives. Alternatively, because sulfides, sulfoxides, and sulfones in a given series are usually considered to be toxicologically equivalent from a regulatory standpoint (Ivie and Bandal, 1981), the sulfur oxidation artifacts observed in the current study may be of little or no significance regarding the development of analytical methods for quantitation of RH-0994 residues. At any rate, we in our studies fortuitously minimized such problems by the addition of unlabeled RH-0994 and its analogues prior to extraction and analysis. With these additions, oxidation of RH-0994 during sample workup appeared to be minimal, on the basis of 0-day data (Table II). However, it is possible that oxidation of phenol to phenol sulfoxide may have occurred during sample workup; thus caution in accepting these data as absolute values is warranted.

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Received for review March 2, 1981. Accepted August 17, 1981. This paper reports the results of research only. Mention of a pesticide does not constitute a recommendation for use by the U.S. Department of Agriculture nor does it imply registration under FIFRA as amended. Mention of a trade name, proprietary product, vendor, or specific equipment likewise does not constitute a guarantee or warranty and does not imply its approval to the exclusion of other products or vendors that may be suitable.

Metabolism of *N*-(2,3-Dichlorophenyl)-3,4,5,6-tetrachlorophthalamic Acid (Techlofthalam) in Paddy Soil and Rice

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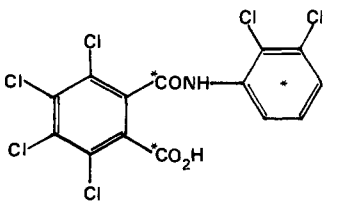
The metabolism of the bactericide *N*-(2,3-dichlorophenyl)-3,4,5,6-tetrachlorophthalamic acid, techlofthalam, has been studied, under controlled conditions, in paddy soil and after application to rice plants by using the ^{14}C-labeled compound. Reductive dechlorination of the tetrachlorophthalamic acid moiety was shown to be the major degradative pathway in paddy soil stored in laboratory flasks. Monodechlorinated products were detected after 2 weeks of incubation, and after 32 weeks more than 90% of the extractable radioactivity, equivalent to about 30% of the applied radioactivity, was associated with two or possibly more monodechlorinated products. Nine percent of the applied radioactivity was converted to $^{14}CO_2$ during 32 weeks. The imide of techlofthalam was a minor metabolite in paddy soil but was the major transformation product detected in rice leaves treated with [^{14}C]techlofthalam.

The compound *N*-(2,3-dichlorophenyl)-3,4,5,6-tetrachlorophthalamic acid, techlofthalam, is a new systemic bactericide for the control of bacterial leaf blight (*Xanthomonas oryzae*) in rice (Nakagami et al., 1980). Techlofthalam possesses low acute mammalian toxicity with



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