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Applicator Exposure to Fluvalinate, Chlorpyrifos, Captan, and Chlorothalonil in Florida Ornamentals[1]

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The exposure of a tractor driver applying pesticides to Florida ornamentals was assessed. The chemicals applied were fluvalinate, chlorpyrifos, captan, and chlorothalonil. Total-body exposure rates, estimated from external exposure pads, were low. Exposure rates followed application rates and were larger when the applicator pulled a boom sprayer than when he pulled a span sprayer. Pesticide on the hands of the ungloved applicator and air samples from his breathing zone were monitored. No significant difference between exposure to the right and left hands was found. The distribution of pesticide on the applicator depended strongly on which spraying device was used. Except for chlorothalonil, tank mixture samples were about 50% weaker in pesticide concentration than would be expected on the basis of complete mixing.

The pesticide exposure of greenhouse applicators is a current regulatory interest of the U.S. Environmental Protection Agency (U.S. EPA). The U.S. EPA is specifically faced with the task (1) of assessing the pesticide exposure of greenhouse applicators and (2) for pesticide label requirements, suggesting protective clothing that is both effective and comfortable. This study is a first step toward providing the data necessary for these evaluations.

The questions addressed by this study were as follows: (1) What was the *potential* for dermal exposure to the applicator; i.e., at what rate did pesticide accumulate on the body (excluding hands) of the applicator, unprotected by clothing of any kind? We term this estimated total-body accumulation rate (ETBAR) and measure it in micrograms per hour. Also, did the ETBAR depend upon the rate of pesticide leaving the spray nozzles, the kind of pesticide applied, and/or the method of application? (2) How was the ETBAR distributed over the anatomy of the applicator, and upon what factors did this distribution depend? (3) What was the accumulation rate of pesticide on the hands of the applicator? Was there a relationship between worker hand preference and exposure to the right and left hands? Did hand exposure depend upon the pesticide effluent rate, compound applied, and/or application method? (4) What was the atmospheric contamination from the pesticide in the breathing zone of the worker as he applied the compound? Did it depend upon the compound type, its effluent rate, or the application method? (5) How did samples of the spray mixture, taken pre- and postapplication, compare in pesticide concentration with that presumed to exist in the tank based on the tank mixture, the pesticide label, and an assumption of thorough mixing?

MATERIALS AND METHODS

Study Site. The study was conducted in 1985 at a commercial greenhouse facility at Cortez, FL, devoted primarily to growing chrysanthemums. The subject monitored was a tractor driver who pulled either a boom sprayer or a span sprayer. The chemicals applied were fluvalinate, chlorpyrifos, captan, and chlorothalonil (Table I), usually in some combination.

Experimental Subject. The subject chosen for this study was a 30-year-old male, height 173 cm, weight 54 kg, who was left-handed. His estimated body surface area was 1.62 m^2 (Gehan and George, 1970). He was instructed to follow his normal application procedure and wore no gloves, coveralls, boots, etc., but did wear a respirator. His outer clothing consisted of a short-sleeved cotton work shirt, denim trousers, and leather shoes.

Application Method. The subject drove a tractor that usually pulled a hydraulic boom sprayer. However, because of equipment failure, he pulled a span sprayer on Aug 14, 1985. With a span sprayer, the spray mixture is

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Table I. Compounds and Formulations

common name	chemical name	brand name	EPA reg no.	formulation
fluvalinate	(αRs,2R)-fluvalinate [(*RS*)-α-cyano-3-phenoxybenzyl (*R*)-2-[2-chloro-4-(trifluoromethyl)anilino]-3-methylbutanoate]	Mavrik	20954-123	22.3% EC
chlorpyrifos	*O*,*O*-diethyl *O*-(3,5,6-trichloro-2-pyridyl)phosphorothioate	Dursban	464-590	50% WP
captan	*N*-[(trichloromethyl)thio]-4-cyclohexene-1,2-dicarboximide	Captan	239-533AA-14775	50% WP
chlorothalonil	tetrachloroisophthalonitrile	Daconil	50534-4	75% WP

distributed by fans mounted just behind each nozzle, rather than by hydraulic pressure alone. He applied within an open-sided structure consisting of wooden poles supporting a horizontal layer of shadecloth (Siran) in an otherwise completely open environment. This structure was 5 ha in extent and was sprayed in sections having various areas.

Sampling and Preliminary Computations. Exposure pads placed on the subject were 10.16 cm × 10.16 cm and made of α-cellulose with a glassine weighing paper backing. They were taped to the subject at the following locations: one on the middle of the back, slightly higher than shoulder blade level; one on the chest just below the collarbone; one on top of each shoulder; one on each forearm, slightly below the elbow and facing outward; one on the front of each thigh, midway between hip and knee and facing frontward; one on each shin, slightly below the knee and facing frontward. These pads were outside all clothing and were entirely exposed to pesticide. Left and right pairs of pads (shoulders, forearms, thighs, shins) were combined for extraction and analysis. Timed exposure periods were at the convenience of the subject but generally lasted about 30 min. Preliminary experiments indicated that longer exposure periods than this risked loss of compound from collection media. After collection from the subject, each left and right pair of pads was wrapped in aluminum foil with exposed surfaces facing each other; back and chest pads were wrapped singly. Pads were then placed on ice in a cooler for transportation to the laboratory. During pad removal, samplers handled the pads by their outside perimeters only.

The analysis results for pesticide compound, uncorrected for recovery, divided by the pad area (one or two pads) and exposure time, give the pad fluxes appearing in supplementary material. The ETBAR was calculated from the outside pad fluxes as follows. Estimated fractional body surface areas were allotted to the head and neck, front torso, back torso, arms, upper legs, and lower legs by the proportions proposed by the U.S. EPA (1985), which are sex specific. Accumulation rates to the arms, for example, are the product of the estimated total-body surface area (1.62 m^2), the arms fraction (14.1% male), and the forearm pad flux (supplementary material). In the same way, upper leg accumulation rates were estimated from thigh pad fluxes, lower leg rates from shin pad fluxes, back torso rates from back pad fluxes, and front torso rates from chest pad fluxes. The head–neck accumulation rate was similarly derived, with the flux estimated from the average of the chest, the back, and twice the shoulder fluxes. These various accumulation rates were then summed to obtain the ETBAR. If any of these individual accumulation rates could not be determined because of a lost or missing sample, no ETBAR was calculated. Hand-wash accumulation rates were not included in the ETBAR since they were dermal exposures and measured by a different method. Left and right hand-wash accumulation rates and ETBAR's appear in the supplementary material. Hand washes were taken by placing the subject's hand in a Baggie containing 200 mL of 95% ethanol and shaking for 30 s. The Baggies were then tied and placed in 475-mL glass Mason jars for transportation on ice to the laboratory. Also appearing in the supplementary material are spray rates, the quotient of the amount of compound sprayed (calculated from the volume of mixture sprayed and the presumed concentration of compound in the mixture), and the exposure time.

The atmospheric pesticide contamination near the breathing zone of the subject is also given and is based upon a 3 L/min intake of air by a personal air sampler (Du Pont P-4000) worn by the subject. Air samplers were calibrated for this 3 L/min rate every 3 days of use. The air sampler intake was through a cylindrical polyurethane foam filter plug whose housing was placed in a downward position and clamped securely to the subject's clothing immediately below the left shoulder. A plastic tube ran from the filter housing to the air sampler pump worn on the back of the waist and supported by a belt. Following the exposure period, the filter was removed with clean tweezers, wrapped in aluminum foil, and placed in the cooler on ice, pending transportation to the laboratory. Tank mixture samples were taken pre- and postapplication directly from the tank. After they reached the laboratory, all samples were stored in a freezer at −20 °C.

Ambient air temperature and relative humidity were taken at the application site, pre- and postapplication. Means of these values appear in the supplementary material.

Extraction. Prior to exposure in the field, the α-cellulose pads and air sampler plugs were preextracted to remove contaminents. They were tumbled in a jar containing methylene chloride (10 pads/1.5 L or 8 plugs/2 L) for 10 min and air-dried. This was repeated with hexane. A glassine paper was attached to each pad; they were wrapped in foil in groups of 10 and stored. Plugs were packaged in groups of eight in Ziploc storage bags. The preextracted pads and plugs showed no interfering peaks by GLC analysis.

An identical extraction procedure was used for all four compounds. Exposed pads were center cut into a 6.35 cm × 6.35 cm (40.32-cm^2) square and quartered with a paper cutter. The blade and cutting edge were washed with acetone and methanol after each pad was quartered. The pieces were placed in a 225-mL jar with 50 mL of hexane and shaken on a flat rack shaker (New Brunswick Scientific, Model R-2) for 5 min at 350 rpm. The extract was decanted into a round-bottom flask, and the pieces were reextracted by the same process. Extracts from the two steps were combined, placed on a rotary evaporator (Brinkman Instruments, Model RE-120) at 40 °C until almost dry, and transferred into 10 mL of hexane. Exposed air sampler plugs were extracted by the same procedure.

The hand-wash sample contained in the Baggie was poured into a 500-mL separatory funnel along with 100 mL of deionized water and 50 mL of hexane. This mixture was gently shaken for 30 s and left standing until the phases separated. The hexane layer was collected. This procedure was repeated once. The extracts were combined, placed on the rotary evaporator at 40 °C until almost dry, and transferred into 10 mL of hexane.

Liquid extracts from the above samples were placed in 20-mL scintillation vials, sealed around the caps with tape,

Table II. Minimum Detection Limits (MDL) and MDL-Equivalents for Pad Flux, Hand-Wash Accumulation Rate, and Air Sample Concentration

compound	MDL, ppb	MDL-equiv[a] pad, $\mu g/cm^2 \cdot h$	hand wash, $\mu g/h$	air sample, $\mu g/L$
fluvalinate	3	0.001	0.06	0.0003
chlorpyrifos	2	0.001	0.04	0.0002
captan	10	0.005	0.20	0.0010
chlorothalonil	2	0.001	0.04	0.0002

[a] Assuming exposure time of 0.5 h.

and stored at −20 °C until GLC analysis.

Tank mixture samples were warmed to room temperature and mixed thoroughly in the sample bottle. Of the sample 1 mL was removed, weighed in a pretared 50-mL volumetric flask, and brought to volume with acetone. The sample was then transferred to an amber bottle with a Teflon-lined cap and stored at −20 °C for later GLC analysis.

Mean (±SE) recoveries from fortified media were as follows: fluvalinate, pad 89 ± 9%, hand wash 76 ± 5%, air sampler plug 67 ± 2%; chlorpyrifos, pad 91 ± 1%, hand wash 94 ± 4%, air sampler plug 77 ± 2%; captan, pad 103 ± 5%, hand wash 84 ± 4%, air sampler plug 85 ± 6%; chlorothalonil, pad 94 ± 1%, hand wash 53 ± 2%, air sampler plug 68 ± 4%. In addition, loss studies were conducted on the exposure pads and the extracts therefrom. No significant loss of compound(s) could be validated by any of these studies. Field blanks were blank, and field fortifications were within recovery ranges.

Gas Chromatographic Analysis. All analyses were done by gas chromatography using electron capture ^{63}Ni detection. Operating conditions for analyses of the various compounds were as follows: fluvalinate, Varian 6000, 10 m × 0.25 mm (i.d.) DB-1 fused silica capillary column, 0.5-μm film thickness, N_2 at 1 mL/min, oven 220 °C, injector 240 °C, detector 285 °C; chlorpyrifos, captan, and chlorothalonil, Tracor 222, 0.91 m × 2 mm (i.d.) silanized glass column with 4% SE 30/6% SP2401 on 100/120-mesh Supelcoport, N_2 at 60 mL/min, oven 170 °C, injector 210 °C, detector 250 °C. The minimum detection limit of the GLC was defined as 10 times the base-line noise level for the instrument. These minimum detection limits and their equivalents in exposed substrate units are given in Table II for all compounds. A standard curve, consisting of at least four different standard concentrations, was run jointly with samples of unknown concentrations. During an analytical run, standards were introduced no less often than every five exposed samples.

For use as standards, fluvalinate (96.2% purity) was obtained from Zoecon Corp., Palo Alto, CA 94303. Chlorpyrifos (99.9%) and chlorothalonil (99.9%) were obtained from the U.S. Environmental Protection Agency, Pesticide and Industrial Chemicals Repository, Research Triangle Park, NC 27711. Captan (99.0%) was obtained from Chevron Chemical Corp., San Francisco, CA 94105.

RESULTS AND DISCUSSION

Strictly speaking, each exposure of the subject was an experiment unto itself since variations existed in compounds, compound spray rates, exposure times, and application methods. Therefore, no exposure constituted a true replication of any other exposure because of these confounding variables that were not under our experimental control. In order to draw any general conclusions, however, some grouping of the data into classes was required. In practice, there was little variation in the presumed effluent rate (kg a.i./h) among exposures for a given compound and application method (Table III). The phrase "presumed effluent rate" suggests some ambiguity and derives from our observation that thorough mixing of the compound was not usually accomplished in the spray tank. Table IV gives the pre- and postspray tank mixture analyses for concentration, expressed as percents of the presumed concentration. While t-tests ($p < 0.05$) showed no statistical difference between pre- and postspray samples, the data indicated that, except for chlorothalonil, only about half of the calculated concentration was actually leaving the spray nozzle. With the spray rate of Table III understood in this sense, we take up the other parameters.

It was evident (Table III) that, for a given application method, the differences in mean ETBAR, hand washes, and air samples that existed among compounds were in part explainable on the basis of mean spray rate differences. Consequently, the individual exposure parameters (supplementary material) were normalized for (divided by) spray rate and the mean values recalculated. Finally, these values were divided by the subject's body weight. What resulted was a measure of the applicator's mean exposure in micrograms deposited/kilogram of body weight per kilogram sprayed, the time units having canceled out

Table III. Mean[a] Values for Spray Rate, Estimated Total-Body Accumulation Rate, Hand Washes, Air Sampler

applicn method	compound	spray rate, kg a.i./h	ETBAR,[b] $\mu g/h$	hand wash,[c] $\mu g/h$ left	hand wash,[c] $\mu g/h$ right	air sampler, $\mu g/L$
boom sprayer	fluvalinate	0.139 ± 0.009 (7)	265 ± 87 (6)	13 ± 2 (7)	17 ± 6 (7)	0.002 ± 0.001 (7)
	chlorpyrifos	0.962 ± 0.044 (11)	3958 ± 1004 (10)	257 ± 76 (11)	332 ± 51 (11)	0.027 ± 0.006 (11)
	captan	0.997 ± 0.067 (7)	2587 ± 1007 (6)	95 ± 37 (7)	131 ± 41 (7)	0.015 ± 0.005 (7)
	chlorothalonil	1.350 ± 0.011 (4)	4614 ± 1054 (4)	209 ± 20 (4)	340 ± 90 (4)	0.009 ± 0.002 (4)
span sprayer	fluvalinate	0.103 ± 0.002 (3)	3 ± 3 (3)	9 ± 2 (3)	8 ± 2 (3)	ND[d] (3)
	chlorpyrifos	0.743 ± 0.013 (3)	203 ± 18 (3)	229 ± 48 (3)	296 ± 204 (3)	0.008 ● 0.001 (3)
	captan	0.743 ± 0.013 (3)	162 ± 62 (3)	43 ± 4 (3)	51 ± 30 (3)	0.006 ± 0.003 (3)

[a] ±SE, with number of replications in parentheses. [b] Excludes hand wash. [c] Subject wore no gloves. [d] None detected.

Table IV. Mean[a] Tank Mixture Concentration, Expressed as Percent of Presumed Tank Mixture Concentration

	boom sprayer prespray	boom sprayer postspray	span sprayer prespray	span sprayer postspray	combined data
fluvalinate	57 ± 11 (7)	53 ± 10 (7)	94 ± 65 (3)	29 ± 5 (3)	57 ± 11 (20)
chlorpyrifos	45 ± 6 (11)	50 ± 6 (11)	114 ± 23 (3)	58 ± 24 (3)	56 ± 6 (28)
captan	30 ± 7 (7)	36 ± 8 (7)	91 ± 6 (3)	66 ± 26 (3)	47 ± 7 (20)
chlorothalonil	105 ± 6 (4)	109 ± 5 (4)			107 ± 4 (8)

[a] ±SE, with number of samples in parentheses.

Table V. Mean[a] Values for Estimated Total-Body Accumulation Rate, Hand Wash, and Air Sampler, Normalized for Spray Rate and Body Weight

applicn method	compound	μg deposited/kg body weight per kg sprayed		
		NETBAR[b]	total hand wash[c]	air sampler
boom sprayer	fluvalinate	39 ± 12 (6)	3.9 ± 0.9 (7)	0.030 ± 0.018 (7)
	chlorpyrifos	80 ± 20 (10)	11.1 ± 1.7 (11)	0.093 ± 0.017 (11)
	captan	50 ± 19 (6)	3.9 ± 0.9 (7)	0.048 ± 0.015 (7)
	chlorothalonil	63 ± 14 (4)	7.6 ± 1.3 (4)	0.022 ± 0.004 (4)
span sprayer	fluvalinate	0.6 ± 0.6 (3)	3.1 ± 0.6 (3)	ND[d] (3)
	chlorpyrifos	5.0 ± 0.4 (3)	13.1 ± 4.1 (3)	0.037 ± 0.004 (3)
	captan	4.1 ± 1.5 (3)	2.4 ± 0.7 (3)	0.028 ± 0.015 (3)

[a] ±SE, with number of replications in parentheses. [b] Excludes hand wash. [c] Subject wore no gloves. [d] None detected.

Table VI. Mean[a] Distribution (%) of ETBAR[b] to Various Body Regions

applicn method	compound	no. of reps	head–neck	front torso	back torso	arms	upper legs	lower legs
boom sprayer	fluvalinate	6	5 ± 1	3 ± 2	11 ± 7	29 ± 5	31 ± 6	20 ● 9
	chlorpyrifos	10	6 ± 1	6 ± 2	8 ± 1	19 ± 5	39 ± 6	22 ± 7
	captan	6	9 ± 1	11 ± 5	8 ± 5	19 ± 5	31 ± 7	21 ± 12
	chlorothalonil	4	7 ± 1	2 ± 1	13 ± 1	17 ± 3	43 ± 4	17 ± 4
	total av	26	7 ± 1	6 ± 1	10 ± 2	21 ± 3	36 ± 3	21 ± 4
span sprayer	fluvalinate	3	ND[c]	ND	ND	100 ± 100	ND	ND
	chlorpyrifos	3	9 ± 1	17 ± 4	26 ± 7	13 ± 1	18 ± 2	18 ± 2
	captan	3	10 ± 1	32 ± 18	20 ± 20	9 ± 4	21 ± 4	7 ± 7
	total av	9	9 ± 1	25 ± 9	23 ± 10	11 ± 2	20 ± 2	13 ± 4

[a] ±SE. [b] As estimated from outside pads (excludes hand washes). [c] None detected.

(Table V). Air sample values had first to be translated to micrograms per hour via the 3 L/min factor. It should be noted that the air sampler data probably underestimate the respiratory exposure of an unmasked applicator. A recent study by Adamis et al. (1985) found that greenhouse applicators inhaled air at a rate of approximately 1 m^3/h (17 L/min). Left and right mean hand-wash data (Table III) showed no significant differences ($p < 0.05$) throughout and were summed to give total hand wash prior to their inclusion in Table V.

To determine whether significant differences existed among compounds for the various normalized statistical parameters in Table V, a Duncan's multiple-range test ($p < 0.05$) was applied to each of the pesticide groups. The subject was significantly more exposed to normalized ETBAR (NETBAR) contamination from chlorpyrifos than from fluvalinate, but the difference was not large and applied only for the span sprayer. Regarding normalized hand-wash contamination, the subject was significantly more exposed to chlorpyrifos than to fluvalinate or to captan for both application methods, but the differences were not large. For normalized air sampler contamination, the subject was more exposed to chlorpyrifos than from fluvalinate or chlorothalonil (boom sprayer) and from chlorpyrifos than from fluvalinate (span sprayer). Again, differences were small. We cannot explain why chlorpyrifos was occasionally the preferred compound, on a normalized basis, to contaminate the subject. In any case, this effect was marginal.

Normalized hand exposure was unaffected by the application method (Table V) regardless of compound. This was probably because the subject's ungloved hands were exposed more through contact with contaminated machinery than by air-borne spray. Conversely, the NETBAR was 1 order of magnitude greater for the boom sprayer compared to the span sprayer. This effect probably resulted from the subject's position relative to the spray as he rode the tractor: about 2 ft higher above the spray for the span sprayer. In fact, hand exposure was usually the principal kind of exposure for the span sprayer (Table III or V), exceeding all other body areas combined. Normalized air sampler values were small, with marginally higher values occurring with boom spraying.

The distribution of the ETBAR to various body regions of the subject (excluding hands) is given in Table VI. Normalization for spray rate was not done here under the assumption that spray rate should not affect the *percent* of compound deposited on any given body area. No statistical difference in ETBAR distribution existed among compounds. Hence, total averages are also given. Distribution differences between the two application methods, however, are apparent. With the boom sprayer, 78 ± 5% of the ETBAR (hands excluded) was to the extremities versus 44 ± 5% for the span sprayer. The extremities, excluding hands, comprised 52% of the subject's estimated body area.

We are unaware of any other study monitoring exposure of a tractor driver applying pesticides to ornamentals. A comparison can be made, however, with tractor drivers pulling air-blast sprayers in Florida citrus. Some representative mean ETBAR's reported are 11.5 and 24.7 mg/h for dicofol (Nigg et al., 1986), 5.1 and 13.1 mg/h for paraquat (Wojeck et al., 1983), and 32.8 mg/h for chlorobenzilate (Nigg and Stamper, 1983). These values are about 1 order of magnitude larger than those given in Table III for boom spraying and 2 orders of magnitude larger than those for span spraying.

Future studies should determine absorbed doses in humans, either directly from skin penetration data or indirectly from urinary metabolite data. The margins of safety for these workers could be estimated, particularly for those compounds with known carcinogenic end points.

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do not necessarily reflect the views of the Agency and no official endorsement should be inferred.

Registry No. Fluvalinate, 69409-94-5; chlorpyrifos, 2921-88-2; captan, 133-06-2; chlorothalonil, 1897-45-6.

Supplementary Material Available: Tables giving exposure period, spray rate, flux onto pads, and accumulation rate for each pesticide (8 pages). Ordering information is given on any current masthead page.

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Leaching of Conversion Products of [^{14}C]Buturon from Soil during 12 Years after Application

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^{14}C-Labeled buturon [*N*-(4-chlorophenyl)-*N'*-methyl-*N'*-isobutynylurea] was applied to wheat and soil in lysimeters under outdoor conditions. In one approach, labeling was uniformly in the ring; application was in two successive years (2.98 kg/ha each). In a second approach, labeling was at the *N'*-methyl group; application was once (2 kg/ha). Leached water containing radioactivity was collected for 12 years. In water from the experiment with ring-labeled [^{14}C]buturon, ^{14}C calculated as buturon after 12 years amounted to 2.14% of total ^{14}C applied, with a concentration peak in the second year. After 12 years, the radioactivity in water comprised 4-chloroaniline, methyl *N*-(4-chlorophenyl)carbamate, and conjugated 4-chloroaniline, as identified by gas chromatography/mass spectrometry. In water from the experiment with [*N'*-*methyl*-^{14}C]buturon, ^{14}C after 12 years was 1.66% of ^{14}C applied, with decreasing leaching rates after 6 years. No chlorinated radioactive products were detected, indicating that the *N'*-methyl group was incorporated into natural substances.

The phenylurea herbicide buturon [*N*-(4-chlorophenyl)-*N'*-methyl-*N'*-isobutynylurea] is widely used since 1962 under the commercial name of Eptapur. It is classified among the less persistent pesticides. Various studies report its rapid degradability and tendency to form conversion products in the terrestrial environment. Studies on its abiotic transformation dealt with its conversion by UV irradiation (Kotzias et al., 1973, 1974). Studies with the fungus *Rhizopus japonicus* reported an elimination of the isobutynyl group from the molecule (Wallnöfer et al., 1973). The metabolism in algae, *Chlorella fusca* var. rubra (Tsorbatzoudi et al., 1976), was also examined. In plants and soil, numerous conversion products were identified (Schuphan and Ebing, 1977; Ebing and Schuphan, 1979; Haque et al., 1976, 1977; Constenla et al., 1984).

In recent years, the occurrence of pesticide residues in groundwater has become a topic of major concern (Milde and Friesel, 1987). Among the pesticides detected in trace amounts in groundwater, there were also phenylurea herbicides; e.g., isoproturon was detected in the raw water of some German drinking water production plants (Industrieverband Pflanzenschutz e.V., 1987). In order to test the leaching behavior of buturon, a 40 kg/ha dose was applied to a lysimeter, 1.35-m height and 1-m^2 diameter (Herzel and Schmidt, 1979). After 4 months, traces of buturon were detected in the leachate (mean concentration 55 ng/L). Although the results of this study cannot be extrapolated to agricultural field conditions due to the very high application rate, they show the potential of this pesticide to be vertically mobile. The leaching behavior of conversion products was not studied.

Due to their higher water solubility, the leaching potential of polar conversion products of pesticides should be higher than that of parent compounds. However, information on the leaching of pesticide metabolites is very limited. In a lysimeter study with [^{14}C]atrazine, seven conversion products were identified in leachate in addition to the parent compound (Schiavon, 1988). In the present study, both ring-labeled and *N'*-methyl-labeled [^{14}C]buturon were applied to soils in different lysimeters, in order to study the long-term leaching behavior of conversion products formed in soil. Ring-labeled [^{14}C]buturon was applied under the viewpoint of persistence. Buturon labeled at the *N'*-methyl group was applied to examine to what degree this functional group is biologically available or is incorporated into natural soil constituents. The results obtained from the analysis of soil and leachate of the experiment with ring-labeled buturon after one growing period have been published previously (Haque et al., 1977). Three months after application, about 50% of the applied radiocarbon was recovered. Of the radioactivity recovered in soil, 50% was extractable. Between one- and two-thirds

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