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DEPOSITION OF MALATHION AND PERMETHRIN ON SOD GRASS AFTER SINGLE, ULTRA-LOW VOLUME APPLICATIONS IN A SUBURBAN NEIGHBORHOOD IN MICHIGAN

R. G. KNEPPER,[1] E. D. WALKER,[2] S. A. WAGNER,[1]
M. A. KAMRIN[3] AND M. J. ZABIK[4]

ABSTRACT. Deposition of malathion and permethrin onto grass surfaces, after ultra-low volume (ULV) application, was studied in a suburban neighborhood in Saginaw County, Michigan. Commercial concentrates of malathion (Cythion ULV®) and permethrin (Biomist 4 + 12®) were sprayed using a truck-mounted ULV aerosol generator. Sod-grass blocks (0.18 m^2) were placed in the frontyard and backyard of homes in the neighborhood at 4 distances to 91.4 m from the road where applications were made. Grass samples were taken from the sod blocks before application and at 15 min, 12 h, 24 h, and 36 h after application. Samples were extracted with solvent, and extractions were subjected to gas–liquid chromatography for detection of malathion and permethrin. Ranges of detection for malathion were 0.0–16.6 mg/0.18 m^2 and for permethrin were 0.0–25.9 mg/0.18 m^2. Most detections were from samples taken nearest the road at 15 min after application. Detections declined as a logarithmic function of time after application and as an exponential function of distance from the road.

INTRODUCTION

Ultra-low volume (ULV) application of high-concentrate insecticides from ground equipment has become a standard method for control of adult mosquitoes (Mount et al. 1968, Lofgren 1970, Hobbs 1976). The technology involves formulations and equipment that produce an aerosol of droplets in a restricted size range that impinge on flying mosquitoes (Weidhaas et al. 1970, Lofgren et al. 1973). Such applications present a low health risk to humans because of low dermal exposure and low mammalian toxicity of the insecticides and are thought to have low deposition onto environmental surfaces compared with expected, "theoretical" values (Tucker et al. 1987, Moore et al. 1993, Tietze et al. 1994). Information on deposition and persistence is important to address regulatory and public concerns about insecticide residues in the environment and their perceived effects on human health.

The Saginaw County Mosquito Abatement Commission is a publicly funded mosquito control agency in Michigan. Part of the program includes ULV application of malathion (formulated as Cythion ULV®) and permethrin (formulated as Biomist 4 + 12®) to reduce populations of adult mosquitoes. Because there is little information on deposition and persistence of these compounds after single applications of ground-applied ULV, especially in neighborhoods where these materials are used, we studied these phenomena for malathion and permethrin on grass in a typical suburban neighborhood in Saginaw County.

MATERIALS AND METHODS

The study was conducted in a 30 year-old housing subdivision in Saginaw County, MI, in July 1993. The landscape consisted of single family dwellings, well-kept lawns, ornamental shrubbery, mature trees, gardens, fences, and other features generally found in a suburban setting in the midwestern USA (Fig. 1). Blocks of sod grass (0.3 × 0.6 m), placed in plastic horticultural flats, were used as targets for insecticide deposition. The flats were provided with water during the experiment to prevent the grass from drying. The blocks were placed in 2 sets of 4 lines each (the backyard and frontyard sets), parallel and downwind from the course of a nearby street (Fromm Drive; see Fig. 1). The lines of sod were placed at distances of 7.6 m (25 ft.), 15.2 m (50 ft.), 30.4 m (100 ft.), and 91.4 m (300 ft.) from the edge of the street. There were sufficient blocks in each set to allow for pre-treatment (time 0) and post-treatment (15 min, 12 h, 24 h, and 36 h after application) sampling for both insecticides at each distance from the street. Grass was sampled from the sod blocks by clipping all grass from the top of the sod with shears and placing the material into clean glass jars fitted with aluminum foil seals inside the lids. Jars were then placed on wet ice for transport to the laboratory, where they were

[1] Saginaw County Mosquito Abatement Commission, 211 Congress Street, Saginaw, MI 48602.

[2] Department of Entomology, Michigan State University, East Lansing, MI 48824.

[3] Institute for Environmental Toxicology, Michigan State University, East Lansing, MI 48824.

[4] Pesticide Research Center, Michigan State University, East Lansing, MI 48824.

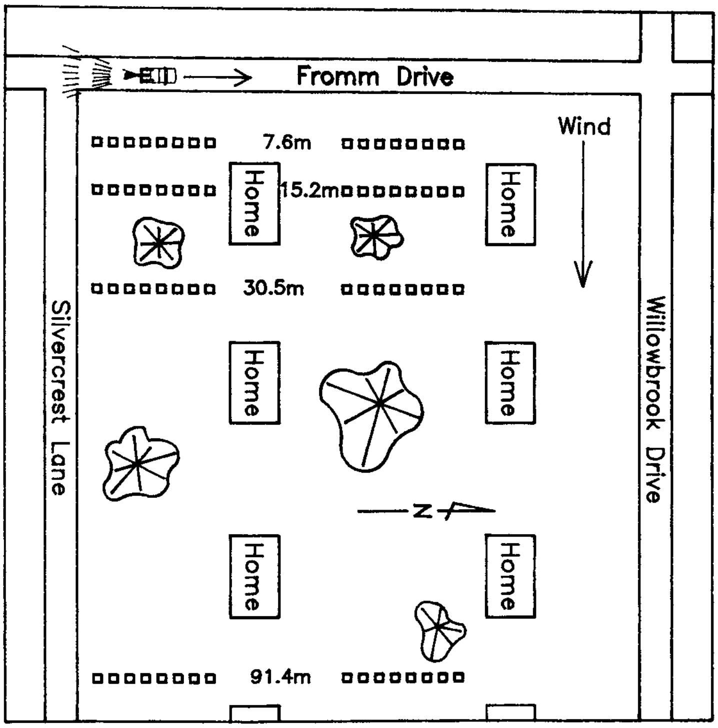

Fig. 1. Schematic diagram of the neighborhood in Saginaw County, MI, where the experimental applications took place. Truck direction was northerly on Fromm Drive. Positions of lines of sod grass blocks (the sampling stations) are shown as small squares in rows at 7.6, 15.2, 30.5, and 91.4 m to the east of Fromm Drive.

stored at −20°C. Between clippings, shears were rinsed once in acetone and then in water.

Malathion (0,0-dimethyl phosphorodithioate ester of diethyl mercaptosuccinate) was applied as a 95% commercial formulation (Cythion ULV; American Cyanamid) at a flow rate of 104 ml/min (3.5 fl. oz./min) and a truck velocity of 16.1 km/h (i.e., 10 mph). A 4% commercial formulation of permethrin ([3-phenoxyphenyl]-methyl[+]cis-trans-3-[2,2-dichloroethenyl]-2,2-dimethylcyclopropanecarboxylate) with 12% piperonyl butoxide synergist (Biomist 4 + 12; Clarke Mosquito Control Products) was applied at a flow rate of 148 ml/min (5.0 fl. oz./min) at the same truck velocity. Applications were made in the evening between 2100 and 2200 h. The application equipment consisted of a LECO™ Model 1600 cold aerosol generator mounted on a one-half-ton pickup truck. The insecticides were delivered by a positive displacement pump with a Micro-Gen™ digital flow control.

For purposes of calibration prior to application for the experiment, insecticide droplets were collected from the spray cloud using silicone-coated glass microscope slides and the hand-wave method (Mount and Pierce 1972). Droplet mass median diameters (MMDs) were calculated from slide counts using a BASIC computer program (Sofield and Kent 1984). Droplet sizes were consistent with requirements as stipulated on the insecticide labels. During the experimental application, insecticide droplets were collected at downwind distances of 7.6, 15.2, and 30.4 m from the road, using silicone-coated microscope slides mounted on mechanical slide rotators (John W. Hock Co.) set equidistantly between the duplicate sets of sod blocks and rotated at 350 rpm for 15 min post-treatment.

Weather data (temperature, relative humidity, wind speed, and wind direction) were collected during the experiment using an on-site weather station (Weather Monitor II; Davis Instruments). During the insecticide applications, temperature was 18°C, relative humidity ranged from 52 to 62%, and wind was easterly and light with a velocity of 1.6 km/h (1 mph). During the 36 h after application, temperatures ranged from 11 to 28°C, relative humidity ranged from 32 to 79%, and wind velocity ranged from 0 to 10 km/h (0–6 mph). There was no rain during the experiment.

For chemical analyses, grass samples were extracted with equal volumes of hexane and acetone, shaken for 5 min, filtered, and mixed with

5% sodium chloride, and the hexane fraction was drawn off for analysis with gas–liquid chromatography using a 60-m DB-5 column with electron capture detection. The limit of detection for permethrin analyses was 0.10 ppm with 91.6% recovery, and the limit of detection for malathion analyses was 0.05 ppm with 89.5% recovery. Curve fitting of insecticide mass (expressed as mg/0.18 m^2 area of the sod blocks) on time after application was done using least-squares regression.

RESULTS

Droplet collections on the silicone-coated slides mounted on rotators showed that the Biomist 4 + 12 droplets had MMDs of 15.7 μm at 7.6 m, 16.1 μm at 15.2 m and 8.9 μm at 30.4 m, whereas the malathion droplets had MMDs of 10.6 μm at 7.6 m, 21.2 μm at 15.2 m, and 29.6 μm at 30.4 m. The distribution of droplets of different sizes that were collected by the slide rotators, shown in histograms in Fig. 2, indicated that most droplets were collected at the 7.6-m distance from the road, compared with the two greater distances. There were significantly more droplets collected overall after the Biomist 4 + 12 application than after the Cythion ULV application (2 × 3 contingency table analysis of frequency of droplets by insecticide and distance, likelihood ratio chi-square = 28.0, df = 2, $P < 0.001$). Inspection of the histograms suggests that there were more smaller droplets of Cythion ULV collected at the 7.6-m distance compared with Biomist 4 + 12.

There were no detections of permethrin or malathion in sod blocks sampled prior to application, i.e., at "time 0" (Table 1). Of the post-treatment samples, there were 20/32 detections of permethrin and 19/32 detections of malathion. Mass of malathion ranged from 0 (undetectable) to 16.6 mg/0.18 m^2, and mass of permethrin ranged from 0 (undetectable) to 25.9 mg/0.18 m^2. Detections of both malathion and permethrin were highest in value in the 15-min post-treatment samples taken at the 7.6-m distance from the road (Table 1). Regression analyses showed that residues at the 7.6-m sampling distance declined as a logarithmic function of time after application (Fig. 3). For permethrin, the regression equation was $Y = 18.1 - 11.5[\log_{10}(X)]$ ($R^2 = 0.98$, $r = 0.99$, $P < 0.001$), and for malathion, the regression equation was $Y = 11.6 - 7.6[\log_{10}(X)$ ($R^2 = 0.98$, $r = 0.99$, $P < 0.001$), where Y is mass of the compound (mg/0.18 m^2) and X is time (h) after application. Regression analyses at the greater sampling distances over time were not done because of the low detection values (see Table 1).

Detections of permethrin and malathion declined in concentration as a negative, exponential function of distance from the road where the applications were made (Fig. 4). For permethrin, the regression equation describing this decline was $Y = 334(X)^{(-1.252)}$ ($R^2 = 0.97$, $r = 0.98$, $P < 0.001$), and for malathion, the regression equation was $Y = 337X^{(-1.549)}$ ($R^2 = 0.99$, $r = 0.99$, $P < 0.001$), where Y is mass of the compound (mg/0.18 m^2) and X is distance (m). These equations adequately describe the relationships between detection and distance within the range of distances in the experiment but should not be extrapolated beyond these ranges.

DISCUSSION

The tendency of insecticides to deposit onto objects and surfaces in the environment is rather poorly understood, particularly in patchy environments such as neighborhoods, where such applications are likely to be made. Drift and fallout of droplets within the size range of ULV aerosols are affected by MMD of droplets, wind speed, air stability, and other factors (Lofgren 1970, Tietze et al. 1994). Moore et al. (1993) evaluated deposition of malathion on human targets immediately following ULV applications, using cotton gauze patches and cotton dust masks as the collection devices, and deposition on the ground using filter paper as the collection device. Similarly, Tietze et al. (1994) examined depositional characteristics of malathion after ULV application onto filter paper targets. Both studies showed low and uniform deposition of malathion as a function of distance from the spray head to 91.4 m, whereas in our study deposition onto ground targets decreased along this distance. This result could be related to more rapid fall-out of larger sized droplets closer to the spray head (Mount 1970) and to the relatively greater number of droplets nearer the spray head than farther away as the droplets dispersed (Fig. 2). Alternatively, differences in patterns of deposition between our study and that of Moore et al. (1993) and Tietze et al. (1994) could be explained on the basis of the complexity of the neighborhood environment where our study was conducted, compared with the open field settings of those studies. Periods of air instability (Armstrong 1979) may alter patterns of deposition, thus the negative exponential model of deposition as a function of distance that we present (Fig. 4) may apply only when air temperature is stable and wind velocity is low. Tietze et al. (1994) observed an increase in deposition of malathion with distance during a single episode of air instability.

Overall, our results showed that deposition of

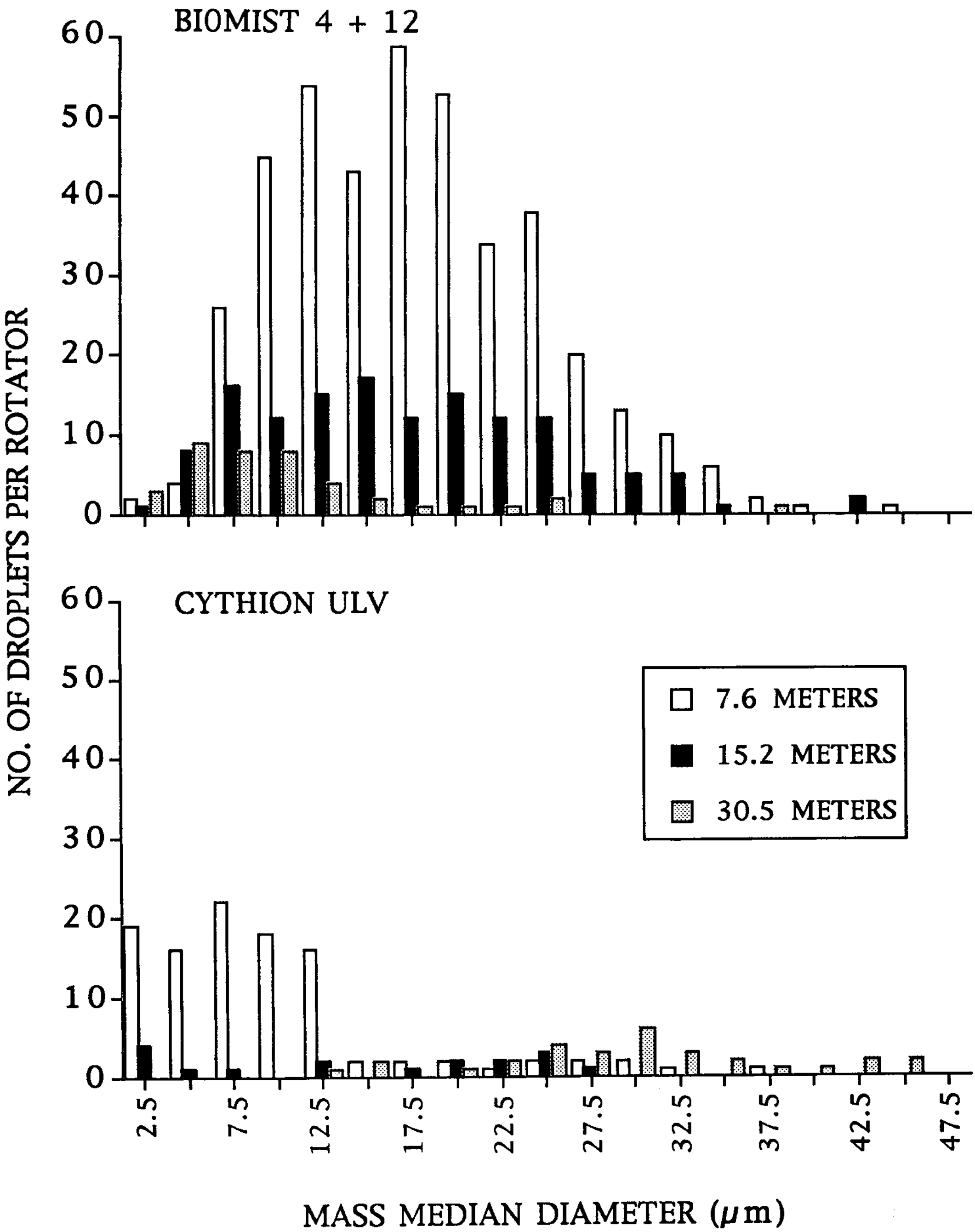

Fig. 2. Number of droplets of different mass median diameters collected by rotating slide collectors at 7.6, 15.2, and 30.5 m from the road where applications of Cythion ULV and Biomist 4 + 12 were made.

Table 1. Detection of permethrin and malathion on grass surfaces after ultra-low volume application of Biomist 4 + 12 or Cythion ULV in a suburban neighborhood in Saginaw County, MI. Values are mass (mg) per 0.18-m² surface area of the sod blocks that were used as sampling devices.

Distance from application (m)	Time after application (h)	Sod block location	Mass of permethrin	Mass of malathion
7.6	0	Frontyard	0	0
		Backyard	0	0
	0.25	Frontyard	25.9	16.6
		Backyard	23.7	15.7
	12	Frontyard	8.1	4.3
		Backyard	6.7	3.1
	24	Frontyard	1.5	0.3
		Backyard	0.2	0.4
	36	Frontyard	0.1	0
		Backyard	0	0
15.2	0	Frontyard	0	0
		Backyard	0	0
	0.25	Frontyard	10.2	4.3
		Backyard	12.1	5.1
	12	Frontyard	3.3	0.6
		Backyard	5.6	0.4
	24	Frontyard	0.3	0.3
		Backyard	1.6	0.1
	36	Frontyard	0	0
		Backyard	0	0
30.5	0	Frontyard	0	0
		Backyard	0	0
	0.25	Frontyard	7.2	1.4
		Backyard	3.8	1.6
	12	Frontyard	0.1	0.4
		Backyard	0.2	0
	24	Frontyard	0	0.2
		Backyard	0	0
	36	Frontyard	0	0
		Backyard	0	0
91.4	0	Frontyard	0	0
		Backyard	0	0
	0.25	Frontyard	1.3	0.3
		Backyard	0.9	0.4
	12	Frontyard	0.3	0
		Backyard	0	0.1
	24	Frontyard	0	0
		Backyard	0	0
	36	Frontyard	0	0
		Backyard	0	0

both malathion and permethrin tended to be high nearest to the road where the truck was driven during the applications. However, there was a discrepancy in the deposited mass of the 2 insecticides on the grass targets. The observed difference in deposition of permethrin (a 4% formulation) compared with malathion (a 95% formulation) cannot be explained by factors related to spray mechanics, because these conditions were similar except for a higher flow rate for the Biomist 4 + 12 formulation (148 ml/min) compared with Cythion ULV (104 ml/min). It is unlikely that the difference in deposition was caused by some variation in conditions at the time the individual applications were made, because there were no obvious meteorological

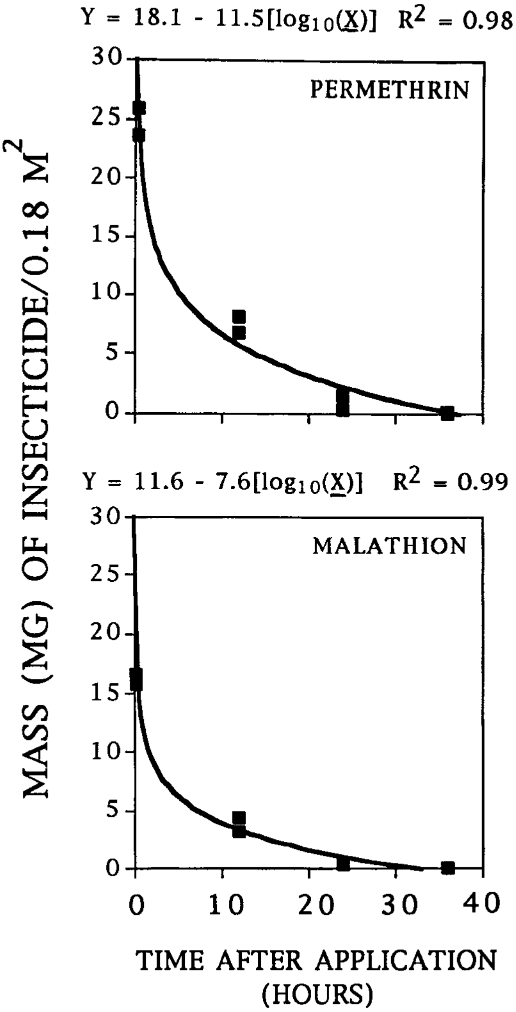

Fig. 3. Detections of mass of permethrin or malathion on grass taken from sod targets after ULV application of Biomist 4 + 12 or Cythion ULV insecticides at the 7.6-m sampling distance only. Pre-treatment sample results are not shown, but all were negative detections. Each point represents a value from one sod block (*n* = 8).

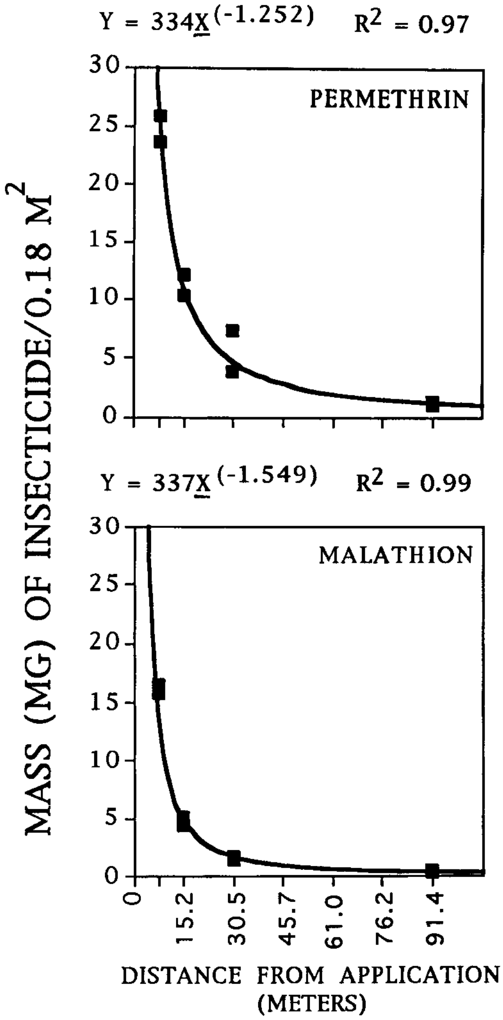

Fig. 4. Detections of mass of permethrin or malathion on grass taken from sod targets after ULV application of Biomist 4 + 12 or Cythion ULV insecticides for 15-min post-treatment samples only. Each point represents a value from one sod block (*n* = 8).

variations that could have influenced the observed deposition patterns. Possibly, the higher flow rate for Biomist 4 + 12 resulted in generation of a greater number of droplets at the spray head. There were a greater number of Biomist 4 + 12 droplets than Cythion ULV droplets collected on the slide rotators during the experiment (see Fig. 2), thus, based upon this empirical observation, one would expect greater deposition of Biomist 4 + 12 droplets than Cythion ULV droplets onto the grass targets as well.

The persistence of deposited insecticides after ULV application is not well known. Both permethrin and malathion have short half-lives in the environment (Howard 1991), but persistence of any insecticide in the environment is affected by its formulation. We found that mass of permethrin and malathion declined as a logarithmic function of time in hours, with most detections of these compounds occurring at the 15-min and 12-h sampling times after application. These data indicate that neither compound persists for very long on grass surfaces after ULV application. We do not present here a risk analysis based upon toxicity and potential exposure of humans to permethrin or malathion residues on grass surfaces, but some contact with insecticides could occur in barefooted people walking on grass near a road where an application truck passed, shortly after the time of application. Walkers are unlikely to receive a toxic dose of either permethrin or malathion after such contact because of the low mammalian toxicity of these compounds and the miniscule amount of chemical on the grass. Moore et al. (1993) concluded that direct exposures of malathion after ULV application were negligible when presented in terms of the maximum allowable daily (dermal) exposure.

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