# DOWNWIND DRIFT AND DEPOSITION OF MALATHION ON HUMAN TARGETS FROM GROUND ULTRA-LOW VOLUME MOSQUITO SPRAYS[1,2]

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ABSTRACT. Malathion was sprayed using a truck-mounted ultra-low volume (ULV) aerosol generator. The generator was operated at 41.4 kPa (6 psi) at flow rate of 128 ml (4.3 fl. oz.) per min. Malathion concentrations were measured at selected positions on live, stationary human subjects wearing protective clothing and placed along a transect at right angles to the path of the truck. Two standing subjects were exposed downwind to the malathion spray at 7.6 and 15.2 m. A third subject was exposed while jogging in the same direction as the spray vehicle and 1.5 m from the spray path. No significant differences ($P > 0.05$) in total amount of malathion deposited on subjects was demonstrated. During the last 4 sprays, average amounts of malathion deposited on ground level at 15.2, 30.4 and 91.2 m were not significantly different ($P > 0.05$). Malathion dermal residues were compared with the acute $LD_{50}$ value (4,100 mg/kg) for a 70 kg adult male. Calculated malathion dermal exposures were less than the acute lethal dose for a human subject by 4 orders-of-magnitude or more.

## INTRODUCTION

Malathion (S-[1,2-bis(ethoxycarbonyl)ethyl] phosphoro-dithioate, Cythion®, American Cyanamid Company) is applied by aircraft and trucks in Florida to control mosquitoes. Spray effectiveness has been determined by mortality of caged mosquitoes, field measures of mosquito abundance or activity and analysis of droplets collected on slides. Undesirable ecological effects of malathion used for mosquito control purposes are commonly associated with misapplication or are limited to short-term effects on sensitive crustaceans and insects (Mulla et al. 1979). Since malathion is acutely toxic to mammals in doses generally ranging from 200 to 1,000 mg/kg (US DHHS 1991), human exposures during mosquito control operations are considered inconsequential.

Increasing public awareness and concern over personal and environmental exposures to mosquito control pesticides requires that malathion deposition be quantified. The objectives of this research were: 1) to determine the amount of malathion deposited on human subjects located at various distances from the path of a spray vehicle during typical spray conditions, 2) to determine the amount of malathion deposited at ground level at various distances from the spray vehicle, and 3) to compare the deposition of malathion onto body surfaces with published dermal $LD_{50}$ values for mammalian toxicity.

## MATERIALS AND METHODS

Malathion applications were conducted on 5 occasions in 1989: three in April and May when temperatures ranged from 21 to 26°C and 2 in August and October when temperatures ranged between 25 and 28°C. Malathion (91% AI) was applied in the evening between 1715 and 1915 h using a Leco HD ULV cold aerosol generator (Lowndes Engineering Co., Valdosta, GA) at 128 ml (4.3 fl oz) per min. A vehicle speed of 16 km/h (10 mph) was maintained immediately upwind of the test area and for a sufficient distance to ensure full coverage at a nominal rate of 58.5 g/ha. Tests were conducted when the direction of prevailing breezes were perpendicular to the test plot. Wind speed was between 1.5 and 3.5 km/h except for Spray 5, when wind speed was 5.5 km/h. Droplet volume median diameter (VMD) deposited on teflon coated slides was measured using a compound microscope (Rathburn 1970). Two teflon coated slides in a rotating impinger were placed adjacent to the individual located 7.6 m downwind.

Deposition was monitored on body surfaces of 3 human subjects to quantify potential human exposure to malathion through dermal absorption. Two subjects were placed in standing, stationary positions at 7.6 m and 15.2 m downwind and facing the path of the spray vehicle. The third subject jogged in the same direction and immediately downwind (1.5 m) of the spray vehicle. Malathion droplets as deposited on body surfaces were collected by using a piece of sterile surgical gauze (280 $cm^2$) placed on the left and

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right front, left and right back of the chest, legs, and arms of each subject. A 181 $cm^2$ area was also sampled from cotton dust masks worn by test subjects. Malathion was collected on precleaned filter paper (24.0 cm diam Whatman #3 or #4 filter paper) that had been placed on styrofoam sheets covered with aluminum foil to quantify potential for deposition of malathion droplets onto smooth terrestrial or water surfaces. The styrofoam sheets were placed horizontally at ground level at sampling distances of 15.2, 30.4 and 91.2 m downwind of the spray path.

The 2 stationary human subjects remained standing for approximately 5 min while the spray cloud passed through the test area. The filter papers used to collect ground level samples remained in place for approximately 10 min.

After each spray, filter papers, gauze patches and face mask samples were placed in individual 150 ml Qorpak® bottles with not less than 100 ml of nanograde petroleum ether. Each bottle was capped, labeled, placed on ice in a cooler and shipped to the Gulf Breeze Laboratory. Samples were kept cool and in the dark until analyses were performed. Analyses were routinely completed within 24 h of collection.

All sample extracts (petroleum ether from the Qorpak bottles) were analyzed initially without preparation to determine if concentration or dilution was needed. If subsequently concentrated, sample extracts were not concentrated to less than 5 ml.

A Hewlett Packard model 5985 gas chromatograph equipped with dual nitrogen-phosphorus detectors was used for analyses. Data were collected and analyzed qualitatively and quantitatively with a Hewlett-Packard model 1000 computer system.

Malathion analytical standard was obtained from the US EPA Pesticide Repository, Las Vegas, NV. The primary standard (1.0 mg/ml) was prepared in acetone, and the working standard (0.020 $\mu$g/ml) was prepared in hexane.

Before field trials were undertaken, we determined the efficiency of extracting malathion from sterile gauze (280 $cm^2$) and filter paper (452 $cm^2$) spiked with different amounts of malathion ranging from 50 to 1,000 $\mu$g for gauze and 100–500 $\mu$g for filter paper. Overall average recovery efficiency and standard deviations of malathion from 26 gauze samples was 97.6 ± 15%. For filter papers, average recovery efficiency was 81.8 ± 22%. In the field, gauze and filter papers were spiked with known amounts of malathion and returned to the laboratory for analysis. Recovery efficiencies and standard deviations for these field samples were 89.8 ± 11% for 9 gauze patches and 93.1 ± 17% for 8 filter

Table 1. Total malathion ($\mu$g) ± SD deposited on gauze patches* attached to the front and back of human subjects or on patches attached to face masks during 5 ground ULV mosquito sprays.

| Subject Location | Location of gauze patch on front of subject | | | | | | Location of gauze patch on back of subject | | | | Mask | Total |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Torso | | Arm | | Leg | | Torso | | Arm | | Leg | |
| | Left | Right | Left | Right | Left | Right | Left | Right | Left | Right | | |
| 1.5 m Jogger | 110.8 ± 147.1 | 133.8 ± 153.7 | 105.4 ± 154.3 | 83.2 ± 79.0 | 34.8 ± 37.5 | 16.4 ± 27.7 | 36.5 ± 31.9 | 50.9 ± 55.9 | 135.2 ± 145.4 | 103.6 ± 88.2 | 38.2 ± 54.8 | 18.8 ± 24.8 | 35.5 ± 37.6 | 520.1 ± 576.5 |
| 7.6 m Stationary | 33.6 ± 42.9 | 27.8 ± 31.6 | 35.0 ± 43.5 | 34.3 ± 31.1 | 20.7 ± 34.9 | 23.5 ± 40.0 | 4.22 ± 1.79 | 2.54 ± 1.14 | 9.53 ± 2.47 | 4.98 ± 4.00 | 3.42 ± 2.50 | 4.14 ± 2.83 | 5.4 ± 2.7 | 192.8 ± 205.9 |
| 15.2 m Stationary | 30.2 ± 22.2 | 26.9 ± 13.8 | 33.4 ± 17.4 | 22.6 ± 6.3 | 14.2 ± 3.5 | 15.6 ± 6.1 | 4.22 ± 1.11 | 4.18 ± 1.98 | 5.98 ± 1.80 | 11.9 ± 5.18 | 5.34 ± 2.07 | 7.20 ± 2.98 | 7.5 ± 2.8 | 147.7 ± 54.4 |
| | | | | | | | | | | | | | | Total |
| 1.5 m Jogger | | | | | | | | | | | | | | 375.7 ± 365.4 |
| 7.6 m Stationary | | | | | | | | | | | | | | 29.1 ± 8.50 |
| 15.2 m Stationary | | | | | | | | | | | | | | 38.88 ± 7.50 |

* 280 $cm^2$ area gauze patches and 181 $cm^2$ gauze patches attached to face mask.

Table 2. Average malathion ($\mu g$) ± SD deposited per $cm^2$ on gauze surfaces placed on various body areas for human subjects during 5 ground, ULV mosquito sprays.

| Distance from source | | Torso | Arms | Legs | Head |
|---|---|---|---|---|---|
| 1.5 m Jogger | Mean | 1.19 ± 1.38 | 1.50 ± 1.67 | 0.39 ± 0.45 | 0.20 ± 0.21 |
| | *n* | 20 | 20 | 19 | 5 |
| 7.6 m Stationary | Mean | 0.27 ± 0.33 | 0.33 ± 0.34 | 0.22 ± 0.34 | 0.03 ± 0.01 |
| | *n* | 20 | 19 | 20 | 5 |
| 15.2 m Stationary | Mean | 0.23 ± 0.10 | 0.26 ± 0.08 | 0.14 ± 0.04 | 0.04 ± 0.02 |
| | *n* | 20 | 20 | 19 | 5 |

Table 3. Estimates of skin exposures for humans placed at selected distances from spray vehicle with various types of dress.*

| Distance from source (m) | Type of exposure | Sleeve | Pants | Malathion exposure (mg) |
|---|---|---|---|---|
| 1.5 | Jogger | Short | Short | 5.0 |
| | | | Long | 3.3 |
| 1.5 | Jogger | Long | Short | 2.6 |
| | | | Long | 0.91 |
| 1.5 | Jogger | Without | Short | 7.8 |
| | | | Long | 6.1 |
| 7.6 | Stationary | Short | Short | 1.6 |
| | | | Long | 0.70 |
| 7.6 | Stationary | Long | Short | 1.1 |
| | | | Long | 0.19 |
| 7.6 | Stationary | Without | Short | 2.3 |
| | | | Long | 1.4 |
| 15.2 | Stationary | Short | Short | 1.2 |
| | | | Long | 0.58 |
| 15.2 | Stationary | Long | Short | 0.78 |
| | | | Long | 0.18 |
| 15.2 | Stationary | Without | Short | 1.8 |
| | | | Long | 1.2 |

* Head exposure included in all types of dress. Short sleeve dress includes arm exposures, long sleeve dress excludes arms. No shirt dress includes arms and torso, shirts exclude torso exposures. Short pants dress includes leg exposures, long pants excludes leg exposures.

papers. When spike recovery percentage was low for field samples, we found that it was due to loss of solvent from the sample container before reaching the laboratory. Before and after each spray, procedural blanks were prepared at the spray site and analyzed to ensure that glassware, solvents, gauze patches or filter papers were not accidentally contaminated. Chemical analysis of these procedural blanks verified that field handling and transport of sample containers, solvents, gauze and filter papers had not contaminated blanks. Malathion deposition model error terms were found to deviate significantly ($P < 0.05$) from the assumption of normality based on the Shapiro-Wilk statistic (SAS Institute 1989). Therefore, Wilcoxon, Van Der Waerden and Savage scores were calculated by SAS (SAS Institute 1989).

## RESULTS AND DISCUSSION

Malathion deposits on gauze attached to the front and back of human subjects or facial masks during each of 5 treatments are shown in Table 1. Malathion deposits on upper torso, arms and legs of the jogger during Spray 1 were 2–30 times greater than that recorded during the other sprays. No reason for this difference was apparent from field observations. With the exception of Spray 1, malathion deposition on the jogger gradually increased with each successive spray, with the values peaking during Spray 5. When data were analyzed by treating data from each spray as a replicate, the amount of malathion deposited on face masks was statistically higher ($P < 0.05$) for the jogger than for the stationary subjects. Deposition on other body areas was not significantly different ($P > 0.05$) among subjects at these distances downwind of the spray source.

The amount of malathion deposited on the back of stationary subjects was less than the amounts on the front. Amounts deposited on the backs of the stationary subjects standing at 7.6 m and 15.2 m from the spray truck were not statistically different from each other ($P > 0.05$), but were less than those for the jogger. Amounts deposited on the back of the jogger varied more (greater standard deviation in proportion to the mean) than amounts deposited on the back of stationary subjects. This variation was attributed to the proximity of the jogger to the spray vehicle and the movements of the jogger. Although the VMD of the spray droplets was ≤15 $\mu$m (range = 13.2–16.2 $\mu$m) at 7.6 m downwind, larger droplets were occasionally measured. Some of these droplets probably struck the jogger and others might have settled to the ground before reaching the stationary subjects.

Estimates of total dermal exposure values for human subjects were derived in the following

Table 4. Dermal exposure (mg) for a shirtless 70 kg human, theoretical number of applications required to obtain an acute malathion $LD_{50}$ of 4,100 mg/kg, and the time required to accumulate an acute $LD_{50}$ dosage (years).*

| Distance from source | | Total deposit[1] (mg) | No. of applications[2] | Time in years[3] |
|---|---|---|---|---|
| 1.5 m Jogger | Short pants | 7.80 | 36,799 | 101 |
| | Long pants | 6.13 | 46,856 | 128 |
| 7.6 m Stationary | Short pants | 2.30 | 124,566 | 341 |
| | Long pants | 1.36 | 211,060 | 578 |
| 15.2 m Stationary | Short pants | 1.78 | 161,408 | 442 |
| | Long pants | 1.18 | 243,799 | 668 |

[1] Sum of exposed surface areas × mean deposit for respective areas.
[2] Malathion $LD_{50}$ for 70 kg male ÷ total deposit on exposed surface area.
[3] Excluding inhalation, time to accumulate $LD_{50}$ with single applications 365 days/year.
* Assuming complete degradation between sprays.

Table 5. Malathion (μg) deposited on filter paper* placed at ground level during 4 ground ULV mosquito sprays.

| Spray | Replicate | Distance from spray vehicle 15.2 m | 30.4 m | 91.2 m |
|---|---|---|---|---|
| 1 | 1 | Lost[1] | 7.3 | 5.7 |
| | 2 | 4.4 | 7.2 | 4.6 |
| | 3 | 4.7 | 9.8 | 4.8 |
| | Mean ± SD | 4.55 ± 0.21 | 8.10 ± 1.47 | 5.03 ± 0.59 |
| 2 | 1 | 5.7 | 3.4 | 4.5 |
| | 2 | 4.4 | 6.8 | 3.8 |
| | 3 | 1.9 | 3.7 | 3.7 |
| | Mean ± SD | 4.00 ± 1.93 | 4.63 ± 1.88 | 4.00 ± 0.44 |
| 3 | 1 | 8.7 | 6.5 | 11 |
| | 2 | 8.0 | 5.3 | 11 |
| | 3 | 7.8 | 9.4 | 11 |
| | Mean ± SD | 8.16 ± 0.47 | 7.06 ± 2.11 | 11 ± 0.0 |
| 4 | 1 | 32 | 38 | 3.3 |
| | 2 | 15 | 33 | 2.4 |
| | 3 | 28 | 31 | 2.3 |
| | Mean ± SD | 25.0 ± 8.89 | 34.0 ± 3.61 | 2.66 ± 0.55 |

[1] Lost = sample spilled during analysis.
* 452 $cm^2$.

manner. Malathion depositions on gauze patches in the left and right side and front and backside of arms, legs and torso were averaged over all sprays and combined to provide an estimate of total malathion deposited (Table 2). These values were then converted to estimates of malathion deposited on various body surfaces of a 70 kg adult male by using body surface area estimates obtained from measurements of one of the authors. A calculated total surface area of 1.9 $m^2$ was near that of 2 $m^2$ reported by Fitzpatrick et al. (1979) for a typical adult male. By combining deposition estimates for the various exposed body areas, we obtained a malathion dermal exposure for a 70 kg male in various types of dress (Table 3). The exposure scenarios considered were common to human exposure situations during a mosquito spray: 1) adult male with short sleeve shirt and short or long pants, 2) adult male with no shirt and short or long pants, and 3) adult male with long sleeve shirt and short or long pants.

Although their data included exposure during handling and formulation, Culver et al. (1956) reported that a mosquito control worker dressed in long trousers and a short sleeve shirt would receive less than 1.8 mg of insecticide on his exposed skin from a single passage of a spray vehicle if he were standing 9.1 m downwind. Calculated dermal exposure during this study for a person with comparable clothing and distance was 0.7 mg total.

Estimates of total dermal exposure for a shirtless subject are compared with a mammalian

(rabbit) acute $LD_{50}$ value of 4,100 mg malathion/kg (Anonymous 1982) in Table 4. Of the dress patterns compared, a shirtless individual wearing short pants represents a worst-case scenario. At 1.5 m downwind from the spray source, an adult male weighing 70 kg would require 36,799 separate applications to accumulate the reported $LD_{50}$. Assuming that there was no loss of toxicity from personal hygiene or degradation and excluding inhalation, that individual would require in excess of 100 years of daily exposure to maximum allowable rates of ULV malathion aerosols applied for adult mosquito control to accumulate the $LD_{50}$ concentration.

Malathion deposits on filter paper at ground level for the 3 distances from the spray vehicle are shown in Table 5. There were no significant differences in deposits at the various distances when data from all sprays were analyzed. Only Spray 5 showed a significant decrease in deposition at the site farthest from the spray source. Among sprays, differences between the least and greatest quantities of malathion deposited were factors of 6, 7 and 4 for 15.2, 30.4 and 91.2 m, respectively. This demonstrated the extent of variation expected between applications for reasonably similar spray conditions.

If all the malathion spray was evenly deposited within the intended treatment area (100 m), malathion should have been deposited on the filter papers at concentrations of approximately 510 ng/cm$^2$. Average amounts of malathion deposited on surface samples (Table 5) ranged from 5.9 to 75 ng/cm$^2$, a range of 1 to 14% of the expected values. Tucker et al. (1987) reported malathion deposition 16 to 17% of predicted values for a ground ULV application near saltwater marshes.

Because these theoretical expected values are used to calculate exposures for nontarget species when evaluating potential environmental hazards (Urban and Cook 1986), the potential for adverse effects during mosquito control operations under controlled and ideal conditions may be less than that predicted for worst case risk assessment. The ranges of environmental exposures at the extremes of normal application conditions still need to be evaluated to determine the margin of safety.

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## REFERENCES CITED

Anonymous, 1982. Cythion materials safety data sheet 0086-01. Cyanamid, Agriculture Division, Wayne, NJ.

Culver, D., P. Caplin and G. S. Batchlor. 1956. A summary of human exposures to malathion and chlorthion during aerosoling operations. Am. Med. Assoc. Arch. Indus. Health 13:37–50.

Fitzpatrick, T. B., A. Z. Eisen, K. Wolff, I. M. Freedburg and K. F. Austen. 1979. Dermatology in general medicine, textbook and atlas. Second edition. McGraw-Hill Book Co., New York.

Mulla, M. S., G. Majori and A. A. Arata. 1979. Impact of biological and chemical mosquito control agents on nontarget biota in aquatic ecosystems, pp. 121–173 *In:* F. A. Gunthner (ed.), Residue Reviews, Vol. 71, Springer-Verlag, New York.

Rathburn, C. B., Jr. 1970. Methods of assessing droplet size of insecticide sprays and fogs. Mosq. News 30:501–513.

SAS Institute, Inc. 1989. SAS/STAT user's guide, version 6. Fourth edition, Volume 2, Cary, NC.

Tucker, J. W., Jr., C. Q. Thompson, T. C. Wang and R. A. Lenahan. 1987. Toxicity of organophosphorus insecticides to estuarine copepods and young fish after field applications. J. Fla. Anti-Mosq. Assoc. 58:1–6.

Urban, D. R. and N. C. Cook. 1986. Hazard Evaluation Division Standard Evaluation Procedures, Ecological Risk Assessment. US EPA 540/9-85-001, USEPA, Office of Pesticide Programs, Washington, DC.

U.S. Department Health Human Services. (US DHHS). 1991. Registry of toxic effects of chemical substances. Search of computerized database of toxicity information, US Public Health Service, Natl. Inst. Occupational Safety and Health, Washington, DC.