Bull. Environ. Contam. Toxicol. (1994) 53:493–500

Environmental Contamination and Toxicology

Droplet Size Characterization of Three Aerial Malathion Spray Programs

T. Younglove, P. M. McCool

Statewide Air Pollution Research Center, University of California—Riverside, Riverside, California 92521, USA

Received: 25 October 1993/Accepted: 18 January 1994

The Mediterranean fruit fly, Ceratitis capitata, and the Mexican fruit fly, Anastrepha ludens are major agricultural pests, capable of destroying a wide variety of fruit. It is the policy of the California Department of Food and Agriculture (CDFA) to pursue eradication of fruit fly populations to prevent establishment in California. A component of the eradication efforts in the past has been the aerial application of a malathion ULV/Nu-Lure® bait mixture. Three aerial spray programs have been monitored by the Environmental Hazards Assessment Program of CDFA. Specifically, these were the eradication of the Mediterranean fruit fly (Medfly), in the Santa Clara Valley in 1981 (Oshima et. al., 1982) and the Los Angeles area in 1990 (Segawa et. al., 1991) and the Mexican fruit fly (Mexfly) in the El Cajon (San Diego) area in 1990 (Turner et. al., 1991). Extensive monitoring of the 1981 application and resulting public and environmental exposure have been presented (Discher, 1982; Finlayson et. al., 1982; Grether et. al., 1987; Oshima et. al., 1982). The two 1990 eradication programs were less extensivly monitored.

One of the components of the monitoring programs for these sprays was the collection of droplet size data. Because of the possibility of additional aerial applications of malathion/bait in future eradication projects, additional analysis of the droplet size data from the three previous studies was undertaken. Continuing public concern over the aerial application of malathion/bait warranted additional analysis of droplet size because of possible effects of droplet size on drift and volatilization of malathion. Therefore, the primary focus of the analysis was on droplet size differences between spray programs and on droplet size as it affects human and environmental exposure. The differences in aerial applications between 1981 and 1990 add some confounding effects to the analysis. However, given the large amounts of time and effort which have gone into the collection of the monitoring data in the past and the expense that would be required to conduct a controlled aerial exposure for experimental monitoring purposes, this analysis is the only realistic option available at the present time.

MATERIALS AND METHODS

Detailed descriptions of the aerial applications are reported in Oshima et. al., 1982; Segawa et. al., 1991 and Turner et. al., 1991. The 1981 malathion/bait formulation consisted of 71 ml (2.4 oz) technical grade malathion (91% active ingredient) mixed with 284 ml (9.6 oz) of Staleys Protein Bait applied by helicopter at an altitude of 91.4 m (300 feet) early in the eradication program and by a combination of helicopter and fixed wing aircraft later in the program. The 1990 malathion/bait

Correspondence to: P. M. McCool

formulation for both El Cajon and Los Angeles consisted of 83 ml (2.8 oz) technical grade malathion (95% active ingredient) mixed with 284 ml (9.6 oz) of Staleys Protein Bait applied by four to six Bell 204 helicopters flying at an altitude of 152.4 m (500 feet) above the ground in Los Angeles and at a minimum of 91 m (298) feet in El Cajon where the ground height was much more variable. The calculated application rate of malathion for the 1981 Medfly spray program was 1836 $\mu g/ft^2$, compared to 2212 $\mu g/ft^2$ for the two 1990 spray programs. The 1981 spray program applied the material through smaller Tee Jet 8003 nozzles (Spraying Systems Co., Wheaton, IL) compared to the 8010 nozzles used in the two 1990 applications.

In addition to the spray application differences there were differences in local topography which may have effected the micro-meteorology at the monitoring sites. All sprays for all three eradication programs occurred at night generally between 9:00 pm and 1:00 am. Some larger scale meteorological differences also existed, particularly between the 1990 Los Angeles Medfly sprays which occurred from February through April and the 1981 Medfly sprays which occurred in July and August. The 1990 El Cajon sprays all occurred in May and June and were similar to the 1981 Medfly sprays both in time of year and distance from the Pacific ocean. The overnight low temperatures for the 1981 Medfly spray dates ranged from a low of 8.9° C to a high of 15.0° C which compared favorably to the El Cajon low of 13.3° C and high of 14.4° C. The 1990 Medfly spray dates on the other hand were cooler with overnight lows ranging from -1.1° C to a high of 12.2° C. Wind speed was not considered a factor in droplet size because of the far greater dispersing effects of helicopter rotor wash in combination with the 128.75 km/hr (80 mile/hr) forward air speed of the helicopters. Aerial applications of the malathion/bait were not conducted during windy conditions in order to minimize drift.

All droplet size monitoring sites for these aerial spray programs were assessed for trees and other obstructions which could have interfered with the descent of the malathion/bait. All non-publicly owned locations were used with written permission of the property owner. The 1981 Medfly monitoring sites were located a minimum of 400 meters apart while the two 1990 studies had sites a minimum of 300 meters apart. Air monitoring sites were selected which had both indoor and outdoor monitoring areas which were accessible with electricity available.

Monitoring utilized droplet size cards, consisting of Kromekote® Cover 65 lb glossy paper approximately 9 cm x 13 cm fixed within a cardboard holder. Additionally, mass deposition absorbent paper towels with plastic backing were attached to cardboard sampling platforms with push pins. One droplet size card was placed at each mass deposition site in each of the studies under consideration. Different spread factors were calculated for the 1981 droplet sizes and the 1990 droplet sizes. For this analysis the spread factor for each year was used for that year as opposed to using the 1990 spread factor for all data. The individual spread factors were used because of the somewhat different formulations of the malathion/bait sprayed in 1981 and 1990. Spray dates which did not have at least 10 droplet card samples taken were not used in this analysis because of the large size of the spray areas. The varying topography and micro-meteorological conditions could not be properly characterized with fewer than 10 samples. Cards from flagged (non-spray) sites and cards without droplets were not used.

Air samples were collected extensively during the 1981 eradication program and on a more limited basis for the 1990 Medfly and 1990 Mexfly programs. Indoor and outdoor samples were collected with low volume air samplers at private residences because of noise considerations and with high volume air samplers at schools and hospitals. Results were reported in µg/m^3 for both high and low volume samplers which allows for comparison between types. Sampling and chemical analysis as well as statistical comparison of the 1990 Medfly and 1981 Medfly air samples has been detailed elsewhere (Segawa et. al., 1991). There were up to 41 total air sampling sites per week in the 1981 Medfly monitoring program, 5 per date in the 1990 Medfly monitoring program and 4 per date in the 1990 Mexfly monitoring program.

Statistical analysis of mean droplet size and distribution had not been conducted previously because of an observed dependency among droplet counts obtained from the same cards (Segawa et. al., 1991; Turner et. al., 1991). This dependency was eliminated by calculating the mean droplet size per card to average out the dependency. The mean droplet size per card was calculated following the same method as previously used to calculate the overall mean droplet size but done on a per card basis; the arithmetic mean of each size category was multiplied by the proportion of droplets in the category, then the values for each category were summed (Segawa et. al., 1991). The largest size category in each study was not used in this analysis because it was not possible to measure the size of these droplets with the instrumentation employed (Segawa et. al., 1991). Because of the small number of droplets found in the largest size category in comparison to the total number of droplets this had a minimal effect on the mean size calculations.

RESULTS AND DISCUSSION

In the 1980 Medfly aerial spray program there were 20 dates with at least 10 nonzero droplet size cards collected. All 10 of the spray dates in the two 1990 aerial applications had at least 20 droplet size cards collected and were included. A total of 821 droplet size cards were used in the combined study, with a minimum of 11 on 7/23/81 and a maximum of 58 on 8/17/81. There were a total of 556 cards in the 1981 Medfly monitoring program, 202 cards in the 1990 Medfly monitoring program and 63 cards taken for the 1990 Mexfly monitoring program. The mean droplet size per study as calculated by the card mean method differed slightly from the mean calculated over all droplets (Table 1). The droplet size dependency precluded the calculation of standard deviations for the three spray programs with the overall droplet size method. However, the means and standard deviations for the mean droplet size per card were easily calculated from the card means.

Table 1. Droplet Size Characterization for Three Eradication Efforts.

Study	Overall Droplet Mean	Mean of Card Means (S.D.)*
1981 Medfly	252 µm	274 µm (104.42)
1990 Medfly	308 µm	346 µm (163.64)
1990 Mexfly	259 µm	268 µm (97.80)

S.D. = Standard Deviation

Significant differences in mean and variance of droplet size were found between the 1990 Medfly spray and both the 1981 Medfly and 1990 Mexfly sprays (Table 2). No significant differences in the mean and variance of the droplet size were found between the 1981 Medfly and 1990 Mexfly sprays.

Table 2. Statistical Analysis of Droplet Size Characterization for Three Eradication Efforts.

ANOVA for Card Mean

Source	DF	Sum of Squares	Mean Square	F-value	
STUDY	2	820983.583	410491.583	27.919	p<0.0001
ERROR	818	12027191.07	14703.168		

Fisher's PLSD test for Card Mean

Comparison	Mean Difference	Critical Difference	p-value
MED1990/MEX1990	78.24	34.35	<0.0001
MED1990/MED1981	72.79	19.55	<0.0001
MEX1990/MED1981	-5.45	31.64	0.7353

F-ratio test for Difference in Variance

MED1990/MED1981 = 26779/10903 = 2.46	p<0.0001
MED1990/MEX1990 = 26779/9564 = 2.80	p<0.0001
MED1981/MEX1990 = 10903/9564 = 1.14	p>0.1000

The observed differences in variance were of some concern because of their possible effect on the significance levels of the standard statistical tests. Transformations were applied to the data in order to equalize the variance between sprays. However, there were still significant differences in variances which were correlated to the means. Given the relatively small differences in variance between monitoring programs, the inability of the transformations to remove the heterogeneity and the high significance of the statistical tests performed, the analysis was conducted on the untransformed data. The results are unbiased but will have lost the minimum variance property (Rohatgi, 1976; Draper and Smith, 1981).

The 1990 Medfly and 1990 Mexfly monitoring programs each had multiple sprays on the same locations on different dates which provided an opportunity to test for significant differences in droplet size between dates without possible confounding effects of location differences. The Rosemead/Monrovia spray area was treated on four dates, 2/21/90, 3/14/90, 4/4/90 and 4/26/90. All three of the spray dates for the El Cajon study were done on the same area. A one-way analysis of variance was run on each area separately to test for significant differences. This was done to obtain the maximum number of error degrees of freedom and to exclude between-site variability from the error term for this test. No significant difference was found between the dates for the Rosemead/Monrovia spray area (p=0.3088). Significant differences were found between the mean droplet size in the El Cajon dates (p=0.0423) with the 5/21/90 date having a mean droplet size significantly larger than the 6/18/90 and 6/4/90 dates.

The 1981 weekly mean droplet size values were regressed against the malathion and malaoxon air concentrations during spray (Spray), first post spray (1st Post) and 2nd post spray (2nd Post) indoors and outdoors. These regressions were conducted separately for Residences, Hospitals, Nursing Homes and the combined 1990 data set. All air samples for each date in the 1990 studies were combined for analysis because of the small number of total samples. A significance level of 0.01 was used as the cut off for the droplet size-air concentration regressions because of the large number of regressions examined.

The regression significance levels for malathion and malaoxon air concentrations are presented in Table 3. There was a significant correlation between mean droplet size and indoor air concentrations of malathion ($p=0.0042$, $R^2=0.896$). The malathion air concentrations during spray had a significant negative correlation with the mean droplet size of the spray (Figure 1). The smaller the mean droplet size the higher the residential indoor air concentrations during the spray.

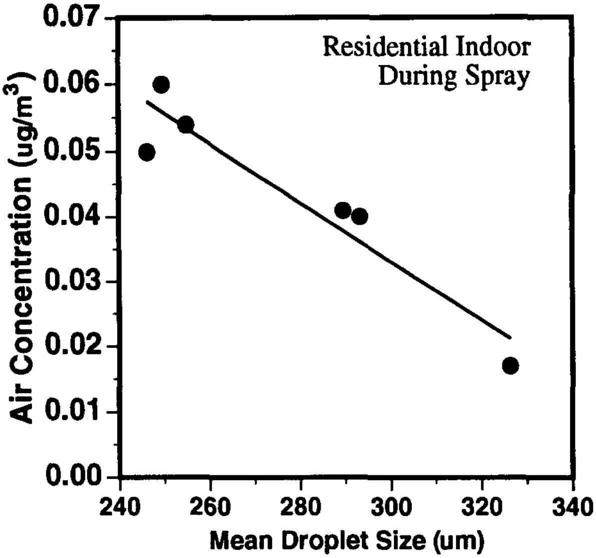

Figure 1. Relationship of residential indoor malathion air concentrations during spray with mean droplet size.

Table 3. Significance levels (p-value) for regressions of mean droplet size on malathion and malaoxon air concentrations. Samples taken during spray (Spray), up to 24 hours post spray (1st) and up to 48 hours post spray (2nd).

Malathion

		Indoor			Outdoor		
Year	Study/Location	Spray	1st	2nd	Spray	1st	2nd
1981	Medfly Residence	.0042**	.0013**	.3307	.1095	.1210	.4826
1981	Medfly Hospital	.1268	.3468	.2813	.1247	.0098**	.0992
1981	Medfly Nursing	.6248	.7142	.1782	.1390	.5986	.6131
1990	Medfly and Mexfly	.2167	.3100	.9948	.7303	.2401	.4450

Malaoxon

		Indoor			Outdoor		
Year	Study/Location	Spray	1st	2nd	Spray	1st	2nd
1981	Medfly Residence	.2932	.4681	.4916	.0755	.4296	.0342
1981	Medfly Hospital	.0995	.5414	.2581	.3729	.3656	.0298
1981	Medfly Nursing	.2512	.3930	.8638	.7216	.6874	.3443
1990	Medfly and Mexfly	.0621	.0160	.1404	.9844	.7373	.3071

** denotes significance at the .01 level

There was also a significant regression of mean droplet size against Indoor 1st Post Spray air concentrations of malathion ($p=0.0013$, $R^2=0.943$). The Indoor 1st Post Spray air concentrations also followed a negative linear trend (Figure 2). The smaller the mean droplet size the higher the residential indoor air concentrations during the 24 hours following the spray.

The indoor air concentrations, while low, were significantly correlated during spray and in the first period after the sprays with the mean droplet size. The smaller the mean droplet size the higher the residential indoor air concentrations during and 24 hours after the spray. No significant correlation's were found between outdoor air concentrations of malathion and mean droplet size at private residences. The air concentrations of malathion increased during and after each spray application in all cases but it was only the indoor 'during spray' and first post spray air concentrations which had increases correlated to the mean droplet size.

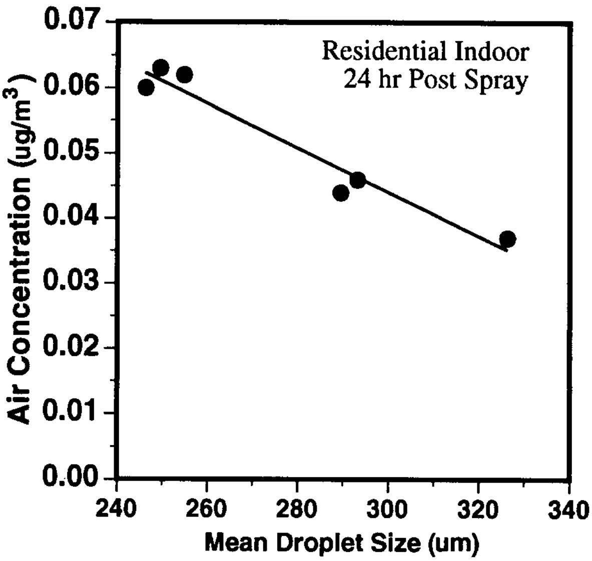

Figure 2. Relationship of residential indoor malathion air concentrations during the 24 hours post spray with mean droplet size.

None of the residence malaoxon air concentrations were significantly correlated with droplet size. The residence 2nd Post spray malaoxon air concentration re gression had a p-value of 0.0342 but with only four observations. Two sample periods did not have malaoxon data available which reduced the power of the regression. Given the large number of regressions examined, the 2nd post spray malaoxon regression was assumed to be a spurious correlation.

It should be noted that while the residences and nursing homes were located within the spray zone the hospital sites were located in flagged areas and should not have received direct spraying of malathion/bait. This may have reduced the 'during spray' air concentrations because of the unknown amount of time required for the malathion droplets to drift to the hospital monitoring sites. Significant mass deposition of malathion was reported at some of the flagged sites during each spray period (Oshima et. al., 1982).

The regression significance levels for Hospital air concentrations are also presented in Table 3. There was a significant regression of mean droplet size with Outdoor 1st Post Spray air concentrations of malathion ($p=0.0098$, $R^2=0.842$). Similar to the residential samples, the air concentrations followed a negative linear trend (Figure 3). No significant correlation's were found between air concentrations of malathion and mean droplet size at any of the indoor hospital samples.

None of the hospital malaoxon air concentrations were significantly correlated with droplet size. The Hospital 2nd Post spray malaoxon concentrations had a regression p-value of 0.0298, but this also was assumed to be spurious because it did not meet the 0.01 significance level chosen because of the large number of tests on the mean droplet size data.

The regression significance levels for Nursing Home air concentrations are presented in Table 3. There were no significant regressions for mean droplet size against air concentrations of either malathion or malaoxon during any time period.

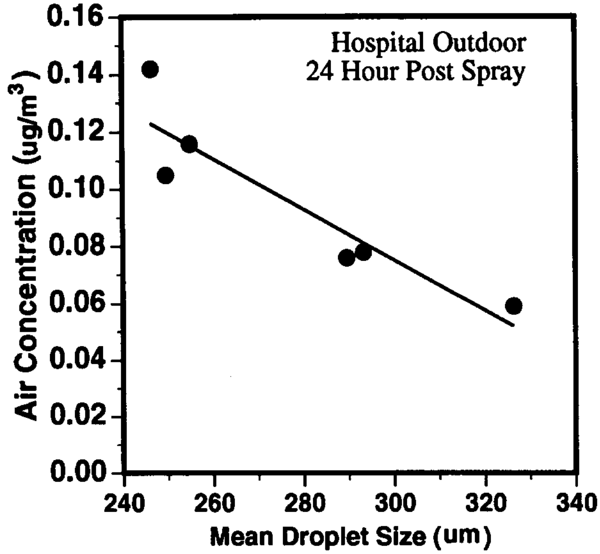

Figure 3. Relationship of hospital outdoor malathion air concentrations during 24 hours post spray with mean droplet size.

The 1990 Medfly monitoring program air monitoring consisted of two or three residence sites and two or three Hospital/School/ Day Care/ Convalescent Hospital sites for a total of five sites per spray. The 1990 Mexfly monitoring program consisted of three School sites and one Hospital site for a total of four sites for each spray date. For analysis, the sites were all averaged together for each spray date because of the small number of samples. The regression significance levels are presented in Table 3. There were no significant ($p<0.01$) regressions of mean droplet size against malathion or malaoxon air concentrations. The 1st post spray malaoxon air concentration regression was nearly significant ($p= 0.016$) but examination of the regression plot indicated that the slope of the regression was entirely determined by two small mean droplet size dates from the 1990 Mexfly program. There may be an effect of droplet size on indoor malaoxon concentrations but it is difficult to determine with these data.

In the Executive Summary of the 1990 Medfly monitoring program it was theorized that temperature may have accounted in part for the differences observed in monitoring between the 1981 and 1990 Medfly programs (Oshima, 1991). The primary influence of temperature would be on droplet size, both as a result of increased viscosity of the malathion/bait mixture and due to changes in air density. The temperature at the time of the spray was not readily available for all of the spray dates, particularly the 1981 Medfly spray. For this reason the overnight low temperature from the nearest national weather service monitoring station was used for the analysis. This may have introduced more error into the 1990 Medfly monitoring program than the other two aerial spray programs because of the number of months and seasons during which the monitoring occurred.

The regression of mean droplet size against overnight low temperature over all three of the monitoring programs had a significant slope at the 0.05 level ($p=0.0140$). An examination of the regression plot revealed that the negative slope of the regression was being determined by four spray dates with temperatures 4.4 ° C (40° F) or below which had higher droplet sizes. All four of the dates with the coldest overnight low temperatures were from the 1990 Medfly program in the winter and spring in Los Angeles which also had a higher overall mean droplet size than the other two spray monitoring programs. A second set of regressions was run on each of the studies separately and no significant regressions were found. Unfortunately the 1981 Medfly study which had the greatest amount of data covered only the upper end of the temperature range.

Significant differences in mean and variance of droplet size were found between aerial spray programs. Differences in mean droplet size were significantly correlated with indoor malathion air concentrations at residences but only outdoors at hospitals which were not located within the spray zones. No significant correlations were found between air concentration of malathion and droplet size at the nursing home sites. Smaller mean droplet size was correlated with increased malathion air concentrations and illustrates the potential for human exposure in residences within the spray zones.

Acknowledgments. We thank Min Poe of the Statewide Air Pollution Research Center, U.C. Riverside and Roberto R. Teso and Dr. Rosemary H. Neal of the California Department of Pesticide Regulation, Riverside office, for their efforts in archiving the data and for assistance in preparing this manuscript. This work was supported by CDPR Contract #93-0120. The mention of commercial products, their source or use in connection with material reported herein is not to be construed as an actual or implied endorsement of such product.

REFERENCES

Discher, DP (Chmn). 1982. Report to the Governor: Medfly Eradication Program. Medfly Health Advisory Committee.

Draper, NR, Smith H. 1981. Applied regression analysis. Second Edition. John Wiley and Sons, New York.

Finlayson, BJ, Faggella G, Jong H, Littrell E, Lew T. 1982. Impact on fish and wildlife from broadscale aerial applications in South San Francisco Bay region. Environmental Services Branch Administrative Report 82-2. California Department of Fish and Game, Rancho Cordova, California.

Grether, JK, Harris JA, Neutra R, Kizer KW. 1987. Exposure to aerial malathion application and the occurrence of low birthweight. Public Health 77:1009-1010.

Oshima, RJ 1991 Executive Summary of Report EH-91-3 "Environmental monitoring of malathion aerial applications used to eradicate Mediterranean fruit flies in Southern California, 1990. " California Department of Food and Agriculture.

Oshima, RJ, Neher LA, Mischke TM, Weaver DJ, Leifson OS . 1982. A characterization of sequential aerial malathion applications in the Santa Clara Valley of California, 1981. California Department of Food and Agriculture. Report EH 82-01

Rohatgi, VK 1976. An introduction to probability theory and mathematical statistics. John Wiley and Sons, New York.

Segawa, RT, Sitts JA, White JH, Marade SJ, Powell SJ. 1991. Environmental monitoring of malathion aerial applications used to eradicate Mediterranean fruit flies in Southern California, 1990. California Department of Food and Agriculture. Report 91-3

Turner, B, Waithman J, Segawa RT. 1991 Environmental monitoring results of the Mexican fruit fly eradication program, San Diego County, Spring 1990. California Department of Food and Agriculture. Report 91-4