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Review

A different look at experiments on pesticide distribution

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Abstract

Experiments on the biological consequences of differences in pesticide distribution include testing differences in application equipment, differences in formulation, and more direct tests of the influence of droplet size, droplet number, or application volume on efficacy for insecticides, herbicides, or fungicides applied as atomized sprays. While these tests have been conducted for at least 60 years, there are continued calls for improving the efficiency of the application process to address ecological, social, and economic concerns of producers and the public about our food and fiber supply. In designing equipment or formulations to address these issues, we need to understand how droplet size, numbers of droplets, toxicant per droplet, and total dose applied influence efficacy. Our solution involves changing our conceptual and experimental framework from a factorial model to a mixture model, and changing our focus from pest management in the field to an individual pest interacting with one pesticide deposit.
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Keywords: Application technology; Experimental design; Mixture design; Insecticides; Fungicides; Herbicides

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1. Introduction

The goal of application technology is ‘‘the placement on targets of just sufficient active ingredient to achieve a desired biological result with safety and economy

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(Hislop, 1987).'' While there are four parts to this goal, we focus on ''just sufficient active ingredient.'' We interpret this statement to mean that every pesticide molecule applied finds its intended target site. The degree to which we fail to achieve this goal is the efficiency of the application process. The estimated efficiency for insecticides is between 0.02% and 3% (Graham-Bryce, 1977). We assume that if there was an innovation that increased efficiency there would be a concomitant decline in application rates, and such a device would take over the market. While shielded sprayers, air assist, and electrostatic sprayers all may improve some aspect of application, they are a long way from taking over the market.

Another indication that little has changed in application efficiency are repeated calls for an improvement in pesticide application (Anderson, 1948; Graham-Bryce, 1977; Hall and Barry, 1995; Hellqvist, 1956; Matthews, 1989). These calls have not resulted in any universal improvement in the application of crop protection agents (Wolf and Downer, 1998) despite the fact that the process is known to be inefficient (Bukovac, 1985; Graham-Bryce, 1977; Herzog et al., 1983; Hislop, 1987; Lawrie et al., 1997; Munthali and Wyatt, 1986; Pimentel and Levitan, 1986; Wolf and Downer, 1998). While it is possible that the problems cannot be solved, we suggest an alternative view of the application process and the mechanisms of pesticide efficacy may permit progress in crop protectant utilization.

2. Background

In the broadest sense the dose transfer process covers everything from the manufacture and distribution of the pesticide to the final biological effect before the last molecule degrades (Ebert et al., 1999a; Young, 1986). For this paper, we focus on the portion of the dose transfer process from deposit formation to biological effect (Fig. 1). We omit literature discussing a dose retained by foliage, because ''a high-level deposit badly distributed is less efficient than a low-level deposit well distributed (Frick, 1970).'' This effect of distribution has been recognized in several systems: herbicides (Holly, 1952), fungicides (Robinson and Garnet, 1984), insecticides (Frick, 1970; Johnstone, 1973; Matthews, 1973).

Table 1 is a list of papers where the primary focus of the paper was to look at the influence of droplet size, droplet number, toxicant concentration, or application volume. We omit papers where only one paper reported on a particular toxicant, and we left out papers on herbicides summarized by Knoche (1994). We converted many papers to a common system of units. Sometimes, droplet size was manipulated through changes in application equipment, and in these cases volume median diameter (VMD), number median diameter

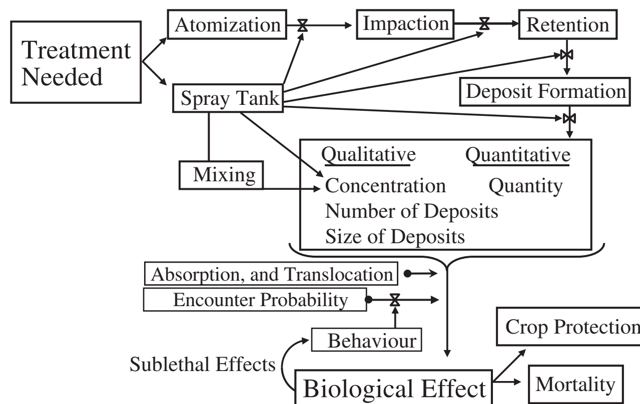

Fig. 1. A simplified version of the dose transfer process that emphasizes processes between deposit formation and biological effect.

(NMD), or mass median diameter (MMD) are often provided. VMD, also referred to as $D_{v0.5}$, is the droplet diameter in μm where half the spray volume is in droplets larger than the listed droplet size, and is equivalent to MMD. NMD is the diameter of droplets where half the numbers are larger than the listed size.

There are many studies that looked for differences in application equipment. Such studies are relevant to this discussion because changes in equipment alter the cloud of atomized droplets. These alterations influence the dispersion and quantity of pesticide on the treated surface, and that translates into a change in the biological result. In a study designed to compare sprayer A against sprayer B, we need to understand how sprayer A could influence efficacy. The sprayer is a mechanical device. The device is filled with fluid containing a pesticide. The pest manager uses the fluid filled sprayer to distribute the pesticide over the crop at some rate of travel ($m\,s^{-1}$) with fluid leaving the sprayer at some rate ($l\,s^{-1}$). We suggest that the primary way a mechanical device influences efficacy is by changing properties of the droplet spectrum: droplet size, droplet number, and droplet velocity.

2.1. Herbicides

Application equipment differences were evaluated by applying glyphosate with Drift Guard, Turbo TeeJet, AI TeeJet, TurboDrop, and flat fan nozzles. No significant differences were found in retention of spray on redroot pigweed (*Amaranthus retroflexus* L) or common lambsquarters (*Chenopodium album* L.). Efficacy was tested against oat (*Avena sativa* L.), proso millet (*Panicum miliaceum* L.), and foxtail millet (*Setaria italica* (L.) P. Beauv.), but no differences were found between different application devices (Ramsdale and Messersmith, 2001). Glyphosate control of common cocklebur (*Xanthium strumarium* (L.)) and broadleaf signalgrass (*Brachiaria platyphylla* (Griseb.) Nash) was evaluated using three nozzles (Delavan Raindrop Ultra (RU), AI TeeJet (AI),

Table 1
Fate of efficacy with changes in droplet size (in μm), numbers, and concentration

Toxicant	Size	Number	Concentration	Units[a]	Citation
Acaricide					
Clofentezine	181–187 = 221–232*V*		0.37 = 1.25	g l^{-1}	(Cross et al., 2000)
Clofentezine			0.08 > 2	g l^{-1}	(Peregrine et al., 1986)
Dicofol	112 > 320	50 > 600	2 < 32	g l^{-1}	(Fisher et al., 1974)
Dicofol	18 > 146		0.5 < 10 > 40	g l^{-1}	(Munthali and Wyatt, 1986)
Dicofol	20 > 100		0.5 < 11.8 > 40	g l^{-1}	(Munthali, 1984)
Dicofol	20 > 120	5 < 200	2.5 > 40	g l^{-1}	(Munthali and Scopes, 1982)
Dicofol	575 > 1560	13 < 100	0.13 < 0.46	g l^{-1}	(Fisher and Morgan, 1968)
Formetanate	100 = 300		0.19 < 8.94	g l^{-1}	(Hall and Reichard, 1978)
Formetanate	100 = 300		0.186 > 8.9	g l^{-1}	(Hall and Reichard, 1978)
Fungicide					
Dinocap	100 < 175 > 400		31 < 500	ppm	(Frick, 1970)
Dinocap	100 > 300		30 > 240	mg l^{-1}	(Falchieri and Cesari, 1993)
Herbicide					
2,4-D	2255	1 = 6	250 = 3000	ppm	(Mullison, 1953)
2,4-D	1970	4 < 100	0.05 > 1.2	g l^{-1}	(Hellqvist, 1956)
2,4-D	985 > 2673		10 < 200	ul leaf^{-1}	(Knoche et al., 1998)
Colletotrichum orbiculare			250 = 1000	l ha^{-1}	(Klein and Auld, 1995)
Colletotrichum truncatum	104 < 421*V*				(Egley et al., 1993)
Colletotrichum truncatum	900 > 2100				(Egley et al., 1993)
Daminozide	1241 > 2673		10 < 200	ul leaf^{-1}	(Knoche and Bukovac, 1995)
Daminozide	985 > 2673		10 < 200	ul leaf^{-1}	(Knoche et al., 1998)
Dinoseb			0.44 > 2.8	g l^{-1}	(Hellqvist, 1956)
Dinoseb			0.0003 > 0.03	g l^{-1}	(Hellqvist, 1956)
Dinoseb			0.44 > 2.8	g l^{-1}	(Hellqvist, 1956)
Glyphosate			1.2 < 3.6	g l^{-1}	(Kogan and Zúñiga, 2001)
Glyphosate	205 = 370 > 670*V*		45 < 125	l ha^{-1}	(Howarth et al., 2004)
Glyphosate	205 = 670*V*		45 < 85 = 125	l ha^{-1}	(Howarth et al., 2004)
Stagonospora sp.	161 < 250*N*				(Lawrie et al., 1997)
Insecticide					
Azinphosmethyl	100 > 300				(Burt et al., 1970)
Azinphosmethyl	100 > 300				(Smith et al., 1975)
B. thuringiensis			0.0125 > 0.05	ml l^{-1}	(Zehnder and Speese, 1991)
B. thuringiensis	50 = 98*N*,120 = 180*V*	3–5 = 5–10	5 = 16.67	BIU l^{-1}	(van Frankenhuyzen et al., 1989)
B. thuringiensis	90–130 = 150–350*V*	4–10 = 10–28	5 = 16.67	BIU l^{-1}	(van Frankenhuyzen et al., 1991)
B. thuringiensis	100 > 300	1 < 80	5795	IU μl^{-1}	(Bryant and Yendol, 1988)
B. thuringiensis	103 = 134*V*	2 = 8	4 = 13	BIU l^{-1}	(Dubois et al., 1993)
B. thuringiensis	110 = 163*V*				(Dubois et al., 1994)
B. thuringiensis		43 > 466	20 < 160		(Falchieri and Cesari, 1993)
B. thuringiensis		43 > 466	20 > 160		(Falchieri and Cesari, 1993)
B. thuringiensis	50 = 155*N*		3.2 = 12.5	BIU l^{-1}	(Morris 1984)
	98 = 205*V*				
Baculovirus heliothis			$9.6 \times 10^8 > 1.9 \times 10^9$	PIB l^{-1}	(Smith et al., 1979)
Bifenthrin	985 > 2122	1 < 10			(Hall et al., 1990)
Bifenthrin	150 < 170	1 < 200			(Adams et al., 1991)
Bifenthrin		1 < 500	0.075 > 7.5	g l^{-1}	(Adams and Hall, 1990)
Bifenthrin	97 < 337*V*				(Womac et al., 1994)
Fluvalinate in oil	42 = 299*V*	0.33 = 321	4.5 < 22.7	g l^{-1}	(Smith and Luttrell, 1987)
Fluvalinate in water	97 = 390*V*	0.08 = 201	0.25 < 1.5	g l^{-1}	(Smith and Luttrell, 1987)
Fluvalinate + oil	42 = 299	0.3 = 321	4.5 < 22.7	g l^{-1}	(Luttrell and Smith, 1990)
Fluvalinate + water	97 = 390	0.1 = 201	0.3 = 1.5	g l^{-1}	(Luttrell and Smith, 1990)
Heterorhabditis sp.			1500 < 12000	IJ ml^{-1}	(Mason et al., 1999)
Lambdacyhalothrin			0.08 > 0.8	g l^{-1}	(Attique et al., 2001)
Lambdacyhalothrin	121 = 302		0.005 < 0.02	g l^{-1}	(Reed and Smith, 2001)
Lambdacyhalothrin	100 > 1000				(Hall and Thacker, 1994)
Monocrotophos			2 > 20	g l^{-1}	(Attique et al., 2001)
Monocrotophos	254 = 556		9.35 = 28.05	l ha^{-1}	(Jimenez et al., 1976)
Permethrin	100 > 1000				(Hall and Thacker, 1994)
Permethrin			103 > 298	l ha^{-1}	(Zehnder and Speese III, 1991)
Permethrin	100 = 220	50	0.4	g l^{-1}	(Adams et al., 1992)
Permethrin	110	25 < 200	0.4	g l^{-1}	(Hoy et al., 1990)
Permethrin	31 > 108				(Adams et al., 1987)

Table 1 (*continued*)

Toxicant	Size	Number	Concentration	Units[a]	Citation
Permethrin in oil	24.1 > 413.5 *V*	0.1 > 189.4	15.1 = 60.6	ml l^{-1}	(Wofford et al., 1987)
Permethrin in water	89.5 = 336.1 *V*	0.8 < 387.2	2.5 < 40	ml l^{-1}	(Wofford et al., 1987)
Permethrin + oil	24 > 413	0.1 > 198	5.8 = 23.2	g l^{-1}	(Luttrell and Smith, 1990)
Permethrin + water	89 = 336	0.8 < 387	0.9 > 15.4	g l^{-1}	(Luttrell and Smith, 1990)
Resmethrin	10 < 15 = 25 > 40 *V*				(Owens and Bennett, 1978)
Resmethrin	20 > 78 *V*				(Mboob, 1975)
Sabadilla			0.03 = 0.05 < 0.15	g l^{-1}	(Yee et al., 2001)
Steinernema sp.			1500 < 12000	IJ ml^{-1}	(Mason et al., 1999)

Size is given as *V* = volume median diameter, *N* = number median diameter, while no letter is given if actual droplet size is used. Greater than, less than, and equal signs indicate direction of improving efficacy: a < b indicates b had greater efficacy than a. Empty cells indicate missing data. IUPAC names for chemicals are given in Table 2.

[a]s = spores, IU = International Units, BIU = billion IU, IJ = infective juveniles, PIB = polyhedral inclusion bodies.

and XR TeeJet (XR)) at two concentrations (50, 100 l ha^{-1}). For both weeds, nozzles were ranked AI = XR > RU, and these nozzles were ranked large to small by VMD RU > AI > XR (Etheridge et al., 2001).

2.2. Fungicides

Over a 2-year spray program for control of apple powdery mildew (*Podosphaera leucotricha*) and apple scab (*Venturia inaequalis*) on apples using a variety of fungicides, efficacy improved with decreasing droplet size (range 90–140 μm), and decreasing concentration (range 50–200 l ha^{-1}) (Cross and Berrie, 1995). Fungicides were mixed with insecticides and adjuvants in a variety of combinations unique to each of the 12 spray dates reported over the 2-year period.

Note: Application volume is inversely proportional to pesticide concentration for a fixed pesticide application rate.

2.3. Acaricides

A conventional sprayer at 613 l ha^{-1} versus an electrostatic sprayer at 31 l ha^{-1}, showed that the electrostatic sprayer improved the efficacy of avermectin and one experimental acaricide, but had no effect on the other three tested (Tjosvold et al., 1996) against twospotted spider mite (*Tetranychus urticae*) on greenhouse grown roses.

2.4. Insecticides

Verticillium lecanii (Zimm.) Viegas was applied with an electrostatic rotary atomizer (APE-80) and a conventional high-volume sprayer for control of cotton aphid (*Aphis gossypii* Glover) and chrysanthemum aphid (*Macrosiphoniella sanborni*) on chrysanthemums. Infection of aphids occurred sooner and peak populations were lower in plots treated with the APE-80. This may have been due to better underleaf deposition with the APE-80 (Sopp et al., 1990).

Four ULV sprayers were tested for deposition patterns on *Pelargonium* and *Impatiens* and for efficacy against greenhouse whitefly (*Trialeurodes vaporariorum*). While the type of sprayer significantly affected the pesticide distribution, there was no effect on efficacy (Sopp and Palmer, 1990).

Viral insect pathogens: The nucleopolyhedrosis virus of velvetbean caterpillar (AgMNPV) was evaluated over four growing seasons in Brazil. Efficacy was not influenced by changes in nozzle type: Cone JD 10-1, Cone JA 02, XR TeeJet 11002, TwinJet 11002, Turbo TeeJet TT 11003 (Silva and Moscardi, 2002).

Helicoverpa armigera nuclear polyhedrosis virus efficacy against cotton bollworm in Thailand improved with a spinning disc sprayer (11.1 l ha^{-1}) versus a mist blower (55.5 l ha^{-1}) (Parnell et al., 1999).

3. Four confounded variables

Droplet size, droplet number, and pesticide concentration influence efficacy for all pesticides in laboratory, greenhouse, and field studies (Tables 1 and 2). However, there is no clear pattern that would allow one to make statements like: decreasing droplet size usually improves efficacy. As many authors point out, their results are subject to alternative conclusions due to the confounded nature of the variables: (Ebert et al., 1999a; Munthali and Scopes, 1982; Wolf and Downer, 1998) to cite three examples. In the example of *H. armigeria* NPV efficacy against cotton bollworm (Parnell et al., 1999), we could ask: were the results due to a change in the equipment or a change in the application volume? There is no way to answer this question with the data presented. So how should we design these experiments to answer the questions being asked?

As a preamble, we need to clarify droplets versus deposits. Deposits are the pesticides left on the plant

Table 2
List of toxicants, and IUPAC chemical names used in papers cited in this manuscript

Common name	Chemical name
2,4-D	2,4-dichlorophenoxyacetic acid
Avermectin	Extract from fermentation with the bacterium *Streptomyces avermitilis*
Azinphosmethyl	S-(3,4-dihydro-4-oxybenzo[d]-[1,2,3]-triazin-3-ylmethyl) O,O-dimethyl phosphordithioate
Bifenthrin	2-methyl-1,1-biphenyl-3yl)-methyl-3-(2-chloro-3,3,3-trifluoro-1-propenyl)-2,2-dimethyl cyclopropanecarboxylate
Clofentezine	3,6-Bis(2-chlorophenyl)-1,2,4,5-tetrazine
Daminozide	N-dimethylaminosuccinamic acid
Dicofol	1,1-bis(*p*-chlorophenyl)2,2,2-trichloroethanol
Dinocap	2(*or* 4)-isooctyl-4,6 (*or* 2,6)-dinitrophenyl (*E*)-2-butenoate
Dinoseb	2-(1-methylpropyl)-4,6-dinitrophenol
Fluvalinate	(*RS*)-α-cyano-3-phenoxybenzyl *N*-(2-chloro-α,α,α-trifluoro-*p*-tolyl)-DL-valinate
Formetanate	*N,N*-dimethyl-*N*?-[3-[[(methylamino)carbonyl]oxy]phenyl]methanimidamide
Glyphosate	N-(phosphonomethyl) glycine
Lambdacyhalothrin	[1?(*S**),3?(*Z*)]-cyano(3-phenoxyphenyl)methyl 3-(2-chloro-3,3,3-trifluoro-1-propenyl)-2,2-dimethylcyclopropanecarboxylate
Monocrotophos	Dimethyl (*E*)-1-methyl-3-(methylamino)-3-oxo-1-propenyl phosphate
Permethrin	(3-phenoxyphenyl)methyl 3-(2,2,-dichloroethenyl)-2,2-dimethylcyclopropanecarboxylate
Resmethrin	5-benzyl-3-furylmethyl (1*RS*)-*cis-trans*-2,2-dimethyl-3-(2-methylprop-1-enyl)cyclopropanecarboxylate
Sabadilla	Extract from the plant *Schoenocaulon officinale* Grey

after droplets containing pesticides have impacted, been retained, and coalesced. It is difficult to directly manipulate deposits to create a distribution typical of an agricultural spray. In contrast, droplets are easy to manipulate through changes in equipment or chemistry. So, we will talk about droplets because that is what we can change even though it is deposits that are biologically interesting.

A droplet of fixed radius r has a specific volume calculated as $4/3\ \pi r^3$. At any fixed concentration of pesticide, this droplet contains a specific quantity of toxicant. The number of these droplets applied to a specified area will result in a specific quantity of pesticide per unit area. In general: Dose in the environment = sum for all droplet sizes (S) of the numbers of droplets (N) * droplet volume (V) * concentration of pesticide in each droplet (C) or:

$$Dose_{env} = \sum_{s=0\ \mu m}^{\infty} N_s V_s C_s. \tag{1}$$

Any experiment that changes droplet size, or pesticide concentration must also change some other variable to keep Eq. (1) balanced. These variables are confounded physically, and require an experimental design that deals with confounded variables. The general statistical approach is a mixture model as described by Cornell (1990). The Cornell model assumes that the variables are additive, and this is achieved by taking the logarithm of both sides of Eq. (1). To work with Eq. (1), we can assume a monosized droplet distribution and eliminate the summation sign. For modeling a biological response (y) Eq. (1) can be rewritten as:

$$y = \log(Dose) + \log(N) + \log(V) + \log(C). \tag{2}$$

While Eq. (2) looks like any other relationship suitable for a factorial experimental design, such is not the case. While the mathematical relationship is linear, this has not eliminated the problem that knowing any three of the independent variables in this model allows one to calculate the fourth. These factors cannot be made orthogonal. In the Cornell mixture model, N, V, and C are mixture variables while $Dose$ is a process variable.

3.1. Fluidity of the mixture model

The minimum size and minimum concentration are at the discretion of individual researchers. Because the minima are arbitrary, the response surface is not fixed. Consider the central point J (Fig. 2), with a size = 93 μm, number = 609, and concentration = 0.003 g l^{-1}. If the minimum concentration changes to 0.0005 g l^{-1}, then this point changes to size = 56 μm, number = 131, and concentration = 0.07 g l^{-1}. This sensitivity allows us to study the dose transfer process more accurately, but it also makes comparing the results between studies difficult.

3.2. Revised efficacy assays

We need to revisit toxicity bioassays: assays that coat an arena with pesticide and then introduce a pest into the arena, or assays that thoroughly mix pesticide with diet and allow the pest to feed. In each case, the goal is to distribute the pesticide uniformly (at all spatial scales) in the test environment. However, spraying crops has the potential to create deposit patterns anywhere within the triangular graph (Fig. 2). So laboratory bioassays are trying to estimate the entire response surface by

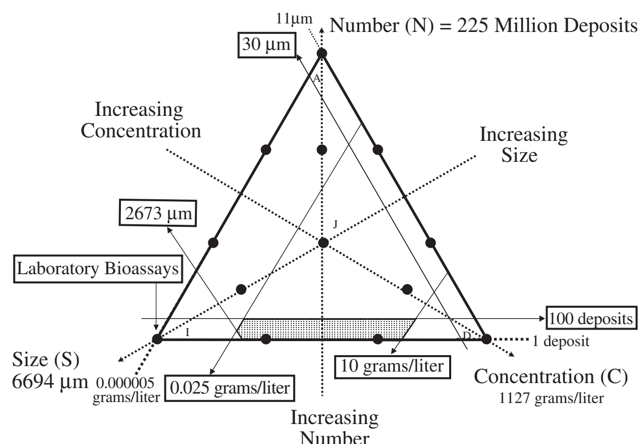

Fig. 2. A mixture design covering most of the possible range in pesticide distribution from the left-hand side of Eq. (1). The shaded area is the range tested for 2,4-D efficacy based on the published ranges in droplet size, droplet numbers, and herbicide concentration. The shaded area overestimates the true proportion tested because it does not account for differences in the dose tested.

assaying at a single point. Therefore it should not be surprising when good laboratory results fail to translate into good field performance.

Experiments for testing equipment differences often fail to clearly identify what is being tested. For example, in evaluating an electrostatic sprayer for control of cotton pests, it was compared with a conventional hydraulic boom sprayer (Herzog et al., 1983). The electrostatic sprayer won the contest, but it applied $9.35\,l\,ha^{-1}$ while the conventional applied $65.5\,l\,ha^{-1}$. So, was the biological effect the result of using the electrostatic sprayer or of applying $9.35\,l\,ha^{-1}$? Could we get the same effect by applying $9.35\,l\,ha^{-1}$ with the conventional sprayer fitted with a smaller orifice nozzle? Likewise, sprayer tests that evaluate retention have reported improved retention on plants sprayed with the electrostatic sprayer (Arnold et al., 1984a; Arnold et al., 1984b; Coates and Palumbo, 1997), but we do not know if the improved retention would result in improved efficacy. This problem is not unique to evaluating electrostatic sprayers (e.g. Holland et al., 1997; McKinlay, 1985). In fairness, if the goal of the research was to evaluate different application techniques to enable farmers to select the best current methodology, then these considerations are unimportant (e.g.Welty et al., 1995). If the goal was scientific, then factorial experimental designs are inadequate (e.g. Holland et al., 1997; Reed and Smith, 2001).

4. The design space

The design space is a coordinate system upon which every possible treatment could be plotted. For factorial designs with one variable and a response the design space is a plane while two variables and a response is plotted in a cube. The variables are orthogonal, or the coordinate plane can be rotated to make them orthogonal. Mixture designs are different, because the independent variables must sum to a total: $A+B+C=100\%$ of the total. The design space with two mixture variables is a line from A to B with the response plotted above the line. With three variables the design space is a triangle with the response plotted as contours on a triangular plane (Fig. 2). Unlike a factorial design, a mixture design is bounded, and it is impossible to go outside these bounds. Mixture variables are not orthogonal, and cannot be made orthogonal. So, what are the bounds in the design space in Eq. (2)?

Fig. 2, shows an example of mixture design space with three variables. The minimum droplet number is 1. The theoretical minimum droplet size is 1 molecule. We do not use this value, because the maximum of the other variables is calculated using the minimum of the remaining variables. Using the theoretical minimum droplet size forces the numbers of droplets to be large (on the order of 10^{15} or more). We arbitrarily chose $11\,\mu m$ as our minimum droplet size to cover most application systems, though a droplet range of $1–2000\,\mu m$ has been reported for agricultural sprayers (Downer and Hall, 1994). Our minimum pesticide concentration was arbitrarily chosen as $0.000005\,g\,l^{-1}$, which is below the minimum concentration used in any paper in the literature review. While the theoretical minimum concentration is infinitely close to zero, using this value forces the other factors to get infinitely large. Given these minima, the maxima were calculated using the fixed total and the minimum values of the other two variables (Ebert et al., 1999a; Ebert et al., 1999b). Within this design space, the gray area in Fig. 2 shows the portion of the design space examined by all studies on 2,4-D efficacy based on the droplet sizes and concentrations examined. The area tested for most other pesticides is much smaller than that of 2,4-D. What happens in the area we have not examined?

5. Common assumptions

Most statements are true within limits. One statement that is frequently encountered is: the quantity of pesticide retained by the foliage determines efficacy. This must be true. At a dose of zero, there is no effect. At some level just above zero we begin to get a biological response. However, once there is sufficient toxicant to get the desired biological effect given 100% efficiency, it is the distribution of the pesticide that determines efficacy. A large concentrated dose may burn a hole in a leaf thereby making the remaining dose inaccessible. For insecticides, high concentrations of

pesticide results in a decrease in deposit size or decrease in numbers of deposits (given a fixed dose). It is therefore less likely that the insect will encounter a dose. It is easy to design an experiment where high doses distributed in small concentrated deposits have a lower efficacy than lower doses distributed as larger and less concentrated deposits (Ebert and Derksen, 2004). The same could be said for herbicides and fungicides, though the causes are different. So what are the bounds within which it is reasonable to say dose determines efficacy? We suggest that for agricultural applications, dose has little to do with efficacy because there is already sufficient pesticide to kill all the pests in the field many times over.

Another common statement is that uniformity of spray coverage is necessary for maximal product efficacy. The key to defining the limits in this statement center around uniformity and spatial scale. For example, if there is one deposit every meter, and I sample the field in 1 m^2 quadrats, I will probably detect a uniform toxicant distribution. For insecticides: At 1 km^2, uniformity is essential for product efficacy. If all the toxicant is at one end of the field, the other end will be eaten. At 1 m^2, the same result is likely. Somewhere between 1 m^2 and 1 μm^2, the rules change. At this scale, individual insects encounter individual deposits. The probability of contact and the dose acquired per contact influence the rate that the pesticide is acquired (Ebert and Derksen, 2004). Given a fixed dose, and, for the sake of simplicity, that this dose is just barely sufficient to cause mortality, then as uniformity increases the probability of contact increases and the toxicant acquired per contact declines. Consider a single lethal insecticide deposit on a leaf. The phytophagous insect might encounter it as soon as it gets to the leaf, or it might eat the entire leaf before encountering the deposit. Given thousands of such encounters, the average result is that half the leaf will be consumed. In contrast if the same deposit is divided into 200 deposits each deposit with 1/200 of a lethal dose, then the contact per deposit will be less, and the insect will have time to eat more of the leaf. Therefore, the uniform distribution will maximize sub-lethal contact and provide more time for the insect to grow and toxicant to decay. One could increase dose, but then the insect dies sooner leaving more toxicant wasted on the plant surface (Ebert and Derksen, 2004). Similar situations occur with herbicides and fungicides so long as there are differential benefits to pesticide at different locations. So, what are the bounds within which uniformity improves efficacy and what is the implied spatial scale for measuring this uniformity?

6. Alternatives

One option to solving the problem of confounded variables is to demonstrate that the biological response is due to some function of size and number, e.g. contact area of deposits (Knoche and Bukovac, 1999), or perimeter of deposits (Adams et al., 1987; Salt and Ford, 1995), and point source diffusion (Sharkey et al., 1987). These experimental designs can work because contact area or perimeter can be held constant through appropriate manipulation of size and number. However, it is hard to see how to translate an optimal perimeter of deposits into a recommendation for sprayer design. One would need to develop a model that translates deposit perimeter into sprayer properties: droplet size, droplet number, and velocity. Furthermore, if a droplet lands on a surface it will spread, and in the case of water-sensitive paper, this spread factor is roughly two. Thus the deposit created by a droplet will be about twice the diameter of the droplet. However, volume increases by the cube of the diameter. The pesticide per droplet will therefore increase proportionately to the cube of the droplet size. Therefore, the pesticide concentration ($g\,cm^{-1}$) will be higher in deposits created from larger droplets. Deposit perimeters may be important when modeling the effects of volatile pesticides. However, the perimeter–dose relationship is similar to the area–dose relationship and the same problems apply. Localized variability in leaf surface composition will add additional complexity to these relationships through their effect on spread factors and boundary layer diffusion.

7. Future directions

Experimental designs for studying the influence of toxicant distribution on efficacy using a mixture approach require the application of monosized droplets. For a polydispersed spray, this experimental design works only if the biological effect of the polydispersed spray is the sum of its parts. This has yet to be proven. What is needed is an estimate of the total dose and some measure of the dispersion of that dose over the treated surface. While dispersion statistics are available (Downer, 1998) , we suggest that fractal dimension will be a better approach. In part this is suggested because fractal geometry may be capable of spanning the range in spatial scales involved in the application process (Baudry, 1993). One approach is to perform a lacunarity analysis (Plotnick et al., 1993, 1996) on digital images of deposits. Fig. 3 shows a comparison of agricultural spray versus the distribution of glands on a leaf surface. The initial high lacunarity values indicate that the frame size is smaller than the differences in spacing between any two deposits. As frame size increases, apparent uniformity also increases. Thus natural deposits are more evenly spaced than agricultural deposits. While we do not know if this difference has a biological consequence, our existing model of insects interacting with discrete pesticide deposits suggests that it should

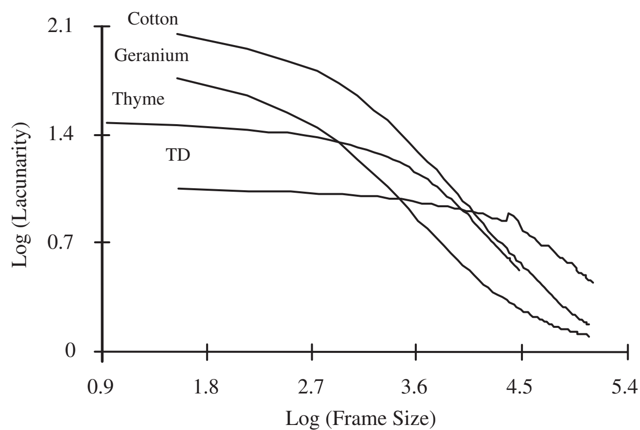

Fig. 3. Lacunarity versus frame size (measured in mm^2) for deposits on surfaces. TD is a turbo drop nozzle from Spraying Systems Co. Wheaton IL used to apply dye to paper. Cotton, geranium, and thyme are from the analysis of the distribution of surface glands on a leaf. All labels are above the curve.

(Ebert and Derksen, 2004). Furthermore, we suggest that the agricultural system could be a model for studying these affects at all spatial scales and working out the problems associated with transitioning from an individual insect to a population.

The conclusion from this review is that we need to study pesticide application as an integrated multi-disciplinary process. This approach was the foundation for Frank Hall's program that resulted in the formation of The Laboratory for Pest Control Application Technology at The Ohio State University (Hall, 1985). However, rethinking every step in the application process is a monumental task. We cannot take short-cuts and assume a single variable (like retention) will provide all the information necessary to predict biological outcomes. Every experiment needs to measure both retention and biological effect. Knowing both is critical to interpret the results properly. Finally, we need to take a much closer look at dispersion and how to measure it in terms that are biologically relevant. Until we start doing this, we will remain where we were 60 years ago—bemoaning the inefficiency of the process and calling for improvements.

The impediments to progress are almost as complex as the application process itself. (1) Education: in all my college classes, pest control was "spray and die." There were long lists of different insecticides, different formulations, and application equipment—but that is all they were, lists. Application technology is worthy of a graduate level course that integrates biology, chemistry, physics, and mathematics. (2) Mixture designs: while the mixture design has been around for quite some time, it has not been a standard feature in statistics or biology classes. The first general text on the subject was published in 1990. (3) Confounded variables: it is easy to take two machines or two formulations, spray and count mortality. Understanding how these treatments resulted in differences in mortality is more difficult. The variables determining the application rate (grams pesticide per hectare) are all confounded: ground speed over the crop, numbers of nozzles per meter of spray boom, pressure, nozzle orifice size, viscosity, and pesticide concentration. It might not look like these variables are confounded—after all one can change tractor speed without changing viscosity, or change pressure without changing nozzle orifice size. However, we know that dose influences efficacy (directly or indirectly) so, try keeping total dose constant and changing viscosity. Viscosity changes flow rates, droplet sizes, and droplet velocities. To compensate, you need to change tractor speed, or pressure, or nozzle orifice—yet these factors also change droplet sizes, numbers, and velocities. A possible solution is to note that travel speed only influences atomization when velocity produces air sheer. Below this velocity, travel speed only changes the numbers of droplets applied, but the size distribution of those droplets remains unchanged. It would appear that changing velocity often has the fewest number of confounded effects.

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