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(54) Title: CONTROLLING SPRAY DRIFT OF PESTICIDES WITH SELF-EMULSIFIABLE ESTERS

(57) Abstract: Spray drift during the application of agricultural chemicals is reduced by incorporating a self-emulsifiable ester into the liquid to be sprayed.



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CONTROLLING SPRAY DRIFT OF PESTICIDES WITH SELF-EMULSIFIABLE ESTERS

This application claims the benefit of United States Provisional Application Serial Number 61/110,060 filed on October 31, 2008.

5 The present invention concerns a novel method to reduce spray drift during the application of agricultural chemicals by incorporating a self-emulsifiable ester into the liquid to be sprayed. Agricultural spraying by economical and available technologies uses hydraulic spray nozzles that inherently produce a wide spectrum of spray droplet sizes. The potential for these spray droplets to drift from the initial, desired site of application is found to be a
10 function of droplet size, with smaller droplets having a higher propensity for off-target movement. Significant research efforts, involving numerous field trials, wind tunnel tests and subsequent generation of predictive math models have led to a greatly enhanced understanding of the relationship between spray droplet size and potential for off-target drift. Although other factors such as meteorological conditions and spray boom height contribute to
15 the potential for drift, spray droplet size distribution has been found to be a predominant factor. Teske et. al. (Teske M. E., Hewitt A. J., Valcore, D. L. 2004. *The Role of Small Droplets in Classifying Drop Size Distributions* ILASS Americas 17th Annual Conference: Arlington VA) have reported a value of <156 microns (μ) as the fraction of the spray droplet distribution that contributes to drift. Wolf (www.bac.ksu.edu/faculty/wolf/drift.htm) cites a
20 value of <200 μ as the driftable fraction. A good estimation of droplet size likely to contribute to drift, therefore, is the fraction below 175 μ .

 The negative consequences of off-target movement can be quite pronounced. Some herbicides have demonstrated very sensitive phytotoxicity to particular plant species at extremely low parts per million (ppm) or even parts per billion (ppb) levels, resulting in
25 restricted applications around sensitive crops, orchards and residential plantings. For example, the California Dept of Pesticide Regulation imposes buffers of ½ - 2 miles (0.8 - 3.2 kilometers) for propanil containing herbicides applied aerially in the San Joaquin valley.

 High molecular weight, water-soluble polymers are currently added to spray compositions as a tank mix to increase droplet size and thereby reduce drift (see, for example,
30 WO 2008/101818 A2 and U.S. 6,214,771 B1). However, high molecular weight, water-soluble polymers are not entirely satisfactory because they are expensive to use at the

concentrations required to substantially increase droplet size. Furthermore, research has shown that many commercially available drift retardants typically do not work with many aerially applied herbicide tank mixtures, due to pump shear, wind shear and other performance issues, which are more pronounced in high speed aerial application conditions.

- 5 See Hewitt, A.J. (2003) Drift Control Adjuvants in Spray Applications: Performance and Regulatory Aspects. *Proc. Third Latin American Symposium on Agricultural Adjuvants, Sao Paulo, Brazil.*

It has now been found that by incorporating a self-emulsifiable ester into an agricultural spray mixture that spray drift during application can be reduced. The present invention concerns a method to reduce spray drift during the application of a pesticide which comprises incorporating into the pesticidal spray from 0.01 to 5 percent vol/vol of a self-emulsifiable ester or mixture thereof. The reduction in spray drift may result from a variety of factors including a reduction in the production of fine spray droplets (<175 μ in diameter) and an increase in the volume median diameter (VMD) of the spray droplets. For a given spray apparatus, application, and condition, and based on the self-emulsifiable ester, the median diameter of the plurality of spray droplets is increased above that of a spray composition without said self-emulsifiable ester.

Another embodiment of the invention is a premix formulation which comprises from 1 to 90 weight percent of a pesticide, and from 0.05 to 30 weight percent of a self-emulsifiable ester. The premix formulation is preferably a solution, emulsion, suspension, wettable or soluble powder, or water-dispersible or water-soluble granule formulation.

The method to reduce spray drift applies to the application of any pesticide or crop protection agent including herbicides, fungicides and insecticides. Particularly preferred herbicides to which this method applies include cyhalofop-butyl, haloxyfop, penoxsulam, flumetsulam, cloransulam-methyl, florasulam, pyroxsulam, diclosulam, fluroxypyr, clopyralid, acetochlor, triclopyr, isoxaben, 2,4-D, MCPA, dicamba, MSMA, oxyfluorfen, oryzalin, trifluralin, benfluralin, ethalfluralin, aminopyralid, atrazine, picloram, tebuthiuron, pendimethalin, propanil, propyzamide, glyphosate and glufosinate. Particularly preferred insecticides to which this method applies include organophosphates such as chlorpyrifos, MAC's such as halofenozide, methoxyfenozide and tebufenozide, pyrethroids such as *gamma*-cyhalothrin and deltamethrin, and biopesticides such as spinosad and spinetoram. Particularly preferred fungicides to which this method applies include mancozeb,

myclobutanil, fenbuconazole, zoxamide, propiconazole, quinoxifen and thifluzamide. The present invention is particularly useful for the application of herbicides, most particularly with herbicides that are subject to restricted applications around sensitive crops which have not been modified to be tolerant of them, such as 2,4-D, dicamba, glyphosate and glufosinate.

5 Self-emulsifiable esters (SEEs) used in the present invention are characterized as molecules that combine oil (hydrophobic), hydrophilic nonionic, and, optionally, anionic functionality in a single molecule that can form uniform stable emulsions in an aqueous phase. Contrary to conventional emulsions in which one or more oils would be blended with one or more surfactants (emulsifiers), no such additives are necessary for the emulsion
10 disclosed in the present invention. Uniform, stable aqueous emulsions can be formed with these SEEs without the use of additional emulsifiers or oils by little to moderate agitation of the SEE and water mixture. Examples of these SEEs include, but are not limited to, the following: (1) trimer acid based self-emulsifiable esters produced by the polymerization of oleic and linoleic acids having a C₅₄ lipophilic backbone and an ester portion of the molecule
15 containing both nonionic and anionic surfactant functionality (see, for example, U.S. Patents 5,688,750 and 5,707,945; commercially available under the trademark Priolube products from Croda Uniqema, Inc); (2) esters prepared by esterification of ethoxylated trimethylolpropane by fatty acids and dicarboxylic acid anhydrides (see, for example, WO1990/005714); (3) esters derived from high molecular weight dibasic acids,
20 polyoxyalkylene glycols and monofunctional aliphatic alcohols (see, for example, U.S. Patent 3,912,642); (4) self emulsifying ester compounds prepared by reacting an ethoxylated trimethylol propane with a carboxylic acid or a reactive derivative thereof, such as an anhydride, as disclosed in U.S. Patent 5,219,479; (5) succinate triglyceride oil derived from maleating triglyceride oil from a plant or land animal (see, for example, WO 2005/071050
25 A1) commercially available as the trademark VEG-ESTER products of Lubrizol, Inc.; (6) ethoxylated fatty acid esters (see, for example, WO 1996/022109); (7) alkoxylate esters prepared by reacting an alcohol with ethylene oxide and propylene oxide and/or butylene oxide and capping the resulting alkoxylate with an alkanoic or aromatic acid as disclosed in U.S. Patent 4,559,226, available commercially as the trademark Hetester products from the
30 Bernel Chemical Company, Inc., a division of Alzo International, Inc.; and (8) alkoxylated triglycerides commercially available as the trademark Agnique RSO and Agnique SBO products from Cognis, Inc.

The self-emulsifiable esters can be incorporated into the pesticidal spray by being tank-mixed directly with the diluted pesticidal formulation or by being provided as a pre-mix with the pesticidal formulation prior to dilution to the final spray volume. The self-emulsifiable ester is incorporated at a concentration from 0.01 to 5 volume percent of the final spray volume, preferably from 0.05 to 1.0 volume percent of the final spray volume, and most preferably from 0.05 to 0.2 volume percent of the final spray volume.

The present method reduces off-target movement of the pesticide spray in both aerial and ground applications.

The optimum droplet size depends on the application for which the composition is used. If droplets are too large, there will be less coverage by the spray; i.e., large droplets will land in certain areas while areas in between will receive little or no spray composition. The maximum acceptable droplet size may depend on the amount of composition being applied per unit area and the need for uniformity in spray coverage. Smaller droplets provide more even coverage, but are more prone to drift during spraying. If it is particularly windy during spraying, larger droplets may be preferred, whereas on a calmer day smaller droplets may be preferred.

The spray droplet size may also depend on the spray apparatus; e.g., nozzle size and configuration. One skilled in the art will readily be able to adjust the percentage of surfactant and/or polymer in the composition to provide the desired droplet size for a given apparatus, application, and condition. In any event, for a given spray apparatus, application, and condition, and based on the self-emulsifiable ester, the median diameter of the plurality of spray droplets is increased above that of a spray composition without said self-emulsifiable ester.

In addition to the method set forth above, the present invention also embraces premix formulations comprising from 1 to 90 weight percent, preferably from 5 to 70 weight percent, and most preferably from 20 to 60 weight percent of a pesticide and from 0.05 to 30 weight percent, preferably from 1.0 to 20 weight percent, and most preferably from 1.0 to 10 weight percent of a self-emulsifiable ester.

Optionally, the composition of the present invention may contain a surfactant. The surfactants can be anionic, cationic or nonionic in character. Typical surfactants include salts of alkyl sulfates, such as diethanolammonium lauryl sulfate; alkylarylsulfonate salts, such as

calcium dodecylbenzenesulfonate; alkyl and/or arylalkylphenol-alkylene oxide addition products, such as nonylphenol-C₁₈ ethoxylate; alcohol-alkylene oxide addition products, such as tridecyl alcohol-C₁₆ ethoxylate; soaps, such as sodium stearate; alkylnaphthalenesulfonate salts, such as sodium dibutylnaphthalenesulfonate; dialkyl esters of sulfosuccinate salts, such as sodium di(2-ethylhexyl) sulfosuccinate; sorbitol esters, such as sorbitol oleate; quaternary amines, such as lauryl trimethylammonium chloride; ethoxylated amines, such as tallowamine ethoxylated; betaine surfactants, such as cocoamidopropyl betaine; polyethylene glycol esters of fatty acids, such as polyethylene glycol stearate; block copolymers of ethylene oxide and propylene oxide; salts of mono and dialkyl phosphate esters; and mixtures thereof. The surfactant or mixture of surfactants is usually present at a concentration of from 1 to 20 weight percent of the formulation.

In addition to the formulations set forth above, the present invention also embraces formulations in combination with one or more additional compatible ingredients. Other additional ingredients may include, for example, one or more other pesticides, dyes, and any other additional ingredients providing functional utility, such as, for example, stabilizers, fragrances, viscosity-lowering additives, and freeze-point depressants.

The premix formulation is preferably a solution, emulsion, suspension, wettable or soluble powder, or water-dispersible or water-soluble granule formulation.

The following Examples illustrate the invention.

Example 1

To make each concentrated 2,4-D formulations A to G as in Table 1, a stainless steel beaker equipped with a mechanical stirrer was first charged with 90 grams DMA 6 SEQUESTERED, containing 68.8% 2,4-D dimethylammonium salt in an aqueous solution. 10 grams total of the different combinations of the Priolube products were then added according to the compositions in Table 1. Each liquid mixture was stirred to homogenize the composition and to furnish the formulation.

The aqueous spray solution was made by adding 1 mL of the each formulation to 99 mL of deionized water to make a 1%v/v dilution. The spray solution was then sprayed through a TeeJet 8002 flat fan nozzle at 40psi (276 kilopascal (kPa)) and the droplet size was measured by using a Sympatec Helos particle sizer. The measurements were made with the

- tip of the nozzle at either 6 or 12 inches (15.24 or 30.48 centimeters (cm)) from the measurement zone of the particle sizer. The results are reported in Table 2 (6 inches; 15.24 cm) and Table 3 (12 inches; 30.48 cm). As can be seen from these results, the driftable fines with droplet size less than 175 μ m by the spray composition of the present invention are
- 5 largely reduced. The present invention efficiently reduces driftable fine droplets by narrowing the drop size distribution profile without significantly increasing the large droplets, thus with minimal effect on spray coverage and quality.

Table 1. Compositions for Formulations of Example 1.

Formulation #	DMA 6 SEQ *, g	Priolube 3955, g	Priolube 3952, g	Priolube 3953, g
DMA 6 SEQ *	100	0	0	0
A	90	0	0	10
B	90	0	5	5
C	90	0	10	0
D	90	3.3	3.3	3.3
E	90	5	0	5
F	90	5	5	0
G	90	10	0	0

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* DMA 6 SEQUESTERED, a 6 lb (2.72 kilograms (kg)) ae/gal 2,4-D-dimethylammonium Manufacturing Use Concentrate of Dow AgroSciences, LLC.

Table 2. Spray Performance of Example 1 Compositions 6 Inches (15.24 cm) from Nozzle.

Spray solution	VMD *, μm	Percent < 100** μm	Percent < 175*** μm
DMA 6 SEQ 1%	213.00	15.99	38.72
A 1%	223.60	14.21	35.64
B 1%	287.44	4.36	16.24
C 1%	281.19	4.65	17.55
D 1%	278.43	5.27	18.76
E 1%	267.02	6.03	21.37
F 1%	255.07	8.52	25.99
G 1%	238.08	10.86	30.62

* VMD – volume median diameter

** Percent of spray volume with droplet size less than 100 μm 5 *** Percent of spray volume with droplet size less than 175 μm

Table 3. Spray Performance of Example 1 Compositions 12 Inches (30.48 cm) from Nozzle.

Spray solution	VMD *, μm	Percent < 100** μm	Percent < 175*** μm
DMA 6 SEQ 1%	164	20.8	53.7
A 1%	204	13.7	40.6
B 1%	251	7.2	25.9
C 1%	248	7.9	27.6
D 1%	227	10.3	33.4
E 1%	237	9.0	30.5
F 1%	231	9.8	32.4
G 1%	190	15.4	44.8

Example 2

The spray solution of formulations of H to N at 1%v/v was made following the same procedure as in Example 1. To each spray solution was then added the proper amount of glyphosate dimethylammonium salt to achieve an acid equivalent ratio between 2,4-D and glyphosate of 1:1. The dilution was stirred to homogenous to furnish the spray solution. It was then sprayed following the same procedure and settings as described in Example 1. The results are shown in Table 4 (measured 6 inches (15.24 cm) from spray nozzle) and Table 5 (measured 12 inches (30.48 cm) from nozzle). As can be seen from these results, the addition of glyphosate does not significantly affect the spray distribution from previous example, indicating the robustness of the present invention in controlling the driftable fines.

Table 4. Spray Performance of Example 2 Compositions 6 Inches (15.24 cm) from Nozzle.

Spray solution	Spray Composition	VMD, μm	Percent < 100 μm	Percent < 175 μm
DMA 6 SEQ 1%		213.00	15.99	38.72
H	A 1% + glyphosate	240.35	10.02	30.09
I	B 1% + glyphosate	279.53	4.58	17.28
J	C 1% + glyphosate	272.80	5.06	19.02
K	D 1% + glyphosate	260.34	6.09	22.43
L	E 1% + glyphosate	257.79	6.54	23.32
M	F 1% + glyphosate	254.81	7.06	24.41
N	G 1% + glyphosate	235.76	10.24	30.60

Table 5. Spray Performance of Example 2 Compositions 12 Inches (30.48 cm) from Nozzle.

Spray solution	Spray Composition	VMD, μm	Percent < 100 μm	Percent < 175 μm
DMA 6 SEQ 1% + glyphosate 1%		148	25.2	60.2
H	A 1% + glyphosate	180	18.0	48.2
I	B 1% + glyphosate	250	7.4	26.5
J	C 1% + glyphosate	235	9.2	30.8
K	D 1% + glyphosate	230	10.0	32.7
L	E 1% + glyphosate	224	10.6	34.3
M	F 1% + glyphosate	230	9.7	32.4
N	G 1% + glyphosate	207	12.9	39.6

Example 3

To make each of the concentrated herbicide formulations O to Q shown in Table 4, a stainless steel beaker equipped with a mechanical stirrer was first charged with 95 g of the herbicide formulation (either DMA6 SEQ, Garlon 3A*, or Milestone**) and 5 g of Hetester PCA (obtained from Alzo International Inc.). Each liquid mixture was stirred to homogenize the composition and to furnish the formulation for subsequent dilution and spray analysis.

The aqueous spray solutions of each were made by adding 2 ml of each of the formulations to 98 ml of deionized water to make 2% v/v dilutions. The solutions were then sprayed following the same procedure and settings as described in Example 1, with the spray nozzle 12 inches (30.48 cm) from particle sizer measurement zone. The results are shown in

Table 6. As can be seen from these results, the present invention does effectively reduce the driftable fine droplets by narrowing the drop size distribution profile without significantly increasing the large droplets, thus having minimal effect on spray coverage and quality.

* Garlon 3A, a 3 lb (1.36 kg) ae/gal triclopyr triethylamine salt commercial product of Dow AgroSciences, LLC.

** Milestone, a 2 lb (0.91 kg) ae/gal aminopyralid triisopropanolamine salt commercial product of Dow AgroSciences, LLC.

Table 6. Spray Performance of Example 3 Compositions.

Spray Solution	Spray Composition	VMD, μm	Percent < 100 μm	Percent < 175 μm
DMA6 SEQ 2%		163.15	21.8	54.2
Garlon 3A 2%		181.20	17.9	47.8
Milestone 2%		152.11	24.9	58.5
O 2%	DMA6 SEQ + Hetester PCA	165.72	21.2	53.2
P 2%	Garlon 3A + Hetester PCA	255.11	8.8	25.5
Q 2%	Milestone + Hetester PCA	243.62	8.6	26.7

Example 4

To make each of the herbicide formulations R through T shown in Table 7, a sample jar was first charged with 294 mL of deionized water, followed by 6 mL of the commercial herbicide formulation (either DMA6 SEQ, Clarity*, or Accord XRT II**) to make a 2% v/v dilution of the formulation. The sample was then shaken until homogeneous. To each of these solutions Veg-Ester GY-350 (Lubrizol, Inc.) was then added in an amount equal to 0.1% w/w of the diluted herbicide formulation. The sample was once again shaken until homogeneous. The solutions were then sprayed following the same procedure and settings as described in Example 1, with the spray nozzle 12 inches (30.48 cm) from particle sizer measurement zone. The results are shown in Table 5. As can be seen from these results, the present invention does effectively reduce the driftable fine droplets by altering the drop size distribution profile without significantly increasing the large droplets, thus having minimal effect on spray coverage and quality.

*Clarity, a 4 lb (1.81 kg) ae/gal dicamba diglycolamine salt commercial product of BASF corp.

**Accord XRT II, a 4 lb (1.81 kg) ae/gal glyphosate dimethylamine salt commercial product of Dow AgroSciences, LLC.

Table 7. Spray Performance of Example 4 Compositions.

Spray Solution	Spray Composition	VMD, μm	Percent < 100 μm	Percent < 175 μm
Clarity 2%		159.21	22.55	55.8
Accord XRT II 2%		142.49	27.73	63.1
DMA6 SEQ 2%		163.15	21.79	54.2
R 2%	Clarity 2% + GY-350 0.1%	192.48	16.39	44.2
S 2%	Accord XRT II 2% + GY- 350 0.1%	160.58	21.52	55.4
T 2%	DMA6 SEQ 2% + GY-350 0.1%	175.55	20.21	49.7

Example 5

1.40 ml of an aqueous solution of Dicamba dimethylammonium salt (Dicamba DMA, 46.9% w/w a.e., 560.92 g a.e./L) and 0.649 g of Priolube 3952 were added to 324 ml of deionized water and hand shaken to yield a solution containing 0.43% v/v of Dicamba DMA and 0.2% w/w of Priolube 3952. In a similar manner, 1.45 ml of the Dicamba dimethylammonium salt solution and 0.70 g of Agnique SBO-10 were added to 335 ml of deionized water to yield a solution containing 0.43% v/v of Dicamba DMA and 0.2% w/w of Agnique SBO-10. As a control, 1.45 ml of the Dicamba DMA concentrate was added to 336 ml of deionized water to yield a 0.43% v/v solution. Finally, a 0.43% v/v solution of Clarity was prepared for comparison. The resulting solutions were sprayed and their droplet size distributions measured as described in Example 1, with the spray nozzle 12 inches (30.48 cm) from particle sizer measurement zone. The results are summarized in Table 8. As can be seen from these results, the present invention does effectively reduce the driftable fine droplets by

altering the drop size distribution profile without significantly increasing the large droplets, thus having minimal effect on spray coverage and quality.

Table 8. Spray Performance of Example 5 Compositions.

Spray Solution	VMD, μm	Percent <100 μm	Percent <175 μm
0.43% Dicamba DMA	159	22.4	55.4
0.43% Dicamba DMA + 0.2% Priolube 3952	260	6.2	23.1
0.43% Dicamba DMA + 0.2% Agnique SBO-10	280	4.9	17.7
0.43% Clarity	161	21.5	55.1

5 Example 6

To 359.07 g of deionized water was added in order: 2.75 g of 2,4-D dimethylethanolammonium salt solution (53.6% a.e.), 0.15 g of Agnique SBO-10, 0.38 g of propylene glycol, and 3.52 g glyphosate dimethylammonium salt solution (42.2% a.e.). As a control, a second sample was prepared as just described, except the Agnique SBO-10 was replaced with 0.15 g of additional deionized water. The resulting solutions were briefly hand-shaken and then analyzed for their spray droplet distributions as described in Example 1, with the spray nozzle 12 inches (30.48 cm) from particle sizer measurement zone. The results are shown in Table 9.

Table 9. Spray Performance of Example 6 Compositions.

Spray Solution	VMD, μm	Percent < 100 μm	Percent < 175 μm
0.40% 2,4-D a.e., 0.41% Glyphosate a.e.	159	22.4	56.1%
0.40% 2,4-D a.e., 0.41% Glyphosate a.e. 0.04% +SBO-10	269	6.2	22.2%

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As used herein, except where the context requires otherwise, the term "comprise" and variations of the term, such as "comprising", "comprises" and "comprised", are not intended to exclude other additives, components, integers or steps.

Reference to any prior art in the specification is not, and should not be taken as, an acknowledgment, or any form of suggestion, that this prior art forms part of the common general knowledge in Australia or any other jurisdiction or that this prior art could reasonably be expected to be ascertained, understood and regarded as relevant by a person skilled in the art.

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We claim

1. A method to reduce spray drift during the application of a pesticide which comprises incorporating into the pesticidal spray from 0.01 to 5 percent vol/vol of a self-emulsifiable ester or mixture thereof, in which the self-emulsifiable ester is (1) trimer acid based self-emulsifiable esters produced by the polymerization of oleic and linoleic acids having a C₅₄ lipophilic backbone and an ester portion of the molecule containing both nonionic and anionic surfactant functionality; (2) esters prepared by esterification of ethoxylated trimethylolpropane by fatty acids and dicarboxylic acid anhydrides; (3) esters derived from high molecular weight dibasic acids, polyoxyalkylene glycols and monofunctional aliphatic alcohols; (4) self emulsifying ester compounds prepared by reacting an ethoxylated trimethylol propane with a carboxylic acid or a reactive derivative thereof; (5) succinate triglyceride oil derived from maleating triglyceride oil from a plant or land animal; (6) ethoxylated fatty acid esters; and (7) alkoxylated triglycerides.

2. The method of Claim 1 in which the pesticide is an herbicide.

3. The method of Claim 2 in which the herbicide is 2,4-D, glyphosate, triclopyr, aminopyralid, dicamba or mixtures thereof.

4. The method of Claim 1 in which the pesticide is an insecticide.

5. The method of Claim 1 in which the pesticide is a fungicide.

6. A premix formulation which comprises from 1 to 90 weight percent of a pesticide and from 0.05 to 30 weight percent of a self-emulsifiable ester, in which the self-emulsifiable ester is

(1) trimer acid based self-emulsifiable esters produced by the polymerization of oleic and linoleic acids having a C₅₄ lipophilic backbone and an ester portion of the molecule containing both nonionic and anionic surfactant functionality; (2) esters prepared by esterification of ethoxylated trimethylolpropane by fatty acids and dicarboxylic acid anhydrides; (3) esters derived from high molecular weight dibasic acids, polyoxyalkylene glycols and monofunctional aliphatic alcohols; (4) self emulsifying ester compounds prepared by reacting an ethoxylated trimethylol propane with a carboxylic acid or a reactive derivative thereof; (5) succinate triglyceride oil derived from maleating triglyceride oil from a plant or land animal; (6) ethoxylated fatty acid esters; and (7) alkoxylated triglycerides.

7. The premix formulation of Claim 6 which comprises from 20 to 60 weight percent of a pesticide and from 1.0 to 10 weight percent of a self-emulsifiable ester.

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8. The method of Claim 1, substantially as herein described.
9. The premix formulation of Claim 6, substantially as herein described.