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(54) Title: HERBICIDAL COMPOSITION

(57) Abstract: The present invention provides a herbicidal composition comprising a first herbicide and a second herbicide in which the first herbicide is 6-chloro-N-ethyl-N'- (1-methylethyl)-1,3,5-triazine-2,4-diamine and the second herbicide is 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl- 2,4(1H,3H)pyrimidinedione and an herbicidal method of use thereof.

HERBICIDAL COMPOSITION

This application claims the benefit of U.S. Provisional Application No. 60/819,571 filed July 10, 2006.

FIELD OF THE INVENTION

This invention relates to herbicidal compositions containing 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and 3-(4-chloro-6-fluoro-2-trifluoromethyl-benzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione.

BACKGROUND OF THE INVENTION

U. S. Patent 6,077,812 discloses 3-(4-chloro-6-fluoro-2-trifluoromethyl-benzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione.

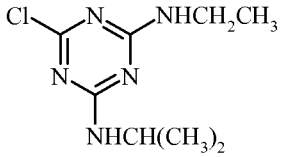
SUMMARY OF THE INVENTION

In accordance with the present invention, it has been found that an herbicidal composition comprising a first herbicide and a second herbicide in which the first herbicide is 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and the second herbicide is 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione, wherein the first herbicide and the second herbicide are present in an herbicidally effective amount, has unexpected advantageous properties.

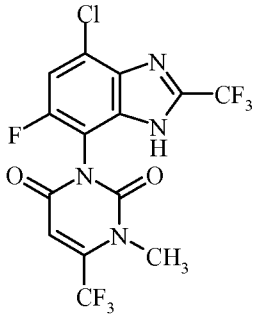
DETAILED DESCRIPTION OF THE INVENTION

It has been unexpectedly found that a composition comprising a first herbicide and a second herbicide in which the first herbicide is 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and the second herbicide is 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione has unexpected advantageous properties in weed control, for example, the mixture controls a broader variety of weeds at low rates. The combination can also be useful in overcoming reduced sensitivity by a weed to either of the individual components

The structural formula of containing 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine is as follows:



The structural formula for 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione is as follows:



The terms "weed" and "weeds" refer to any unwanted vegetation in agricultural crops and orchards, as well as, but not limited to, around buildings, along fences, roadways and rail lines.

The ratio of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine to 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione varies over a wide range but is usually in the range 160:1 to30:1, preferably 63:1 to 12:1.

Other herbicides can be employed in conjunction with the first and second herbicides described above providing they do not adversely affect the interaction between the components of this invention. For example it can sometimes be useful to include additional herbicides to extend the range of activity in order to control a wider spectrum of weeds.

Another embodiment of the present invention is a method for controlling broadleaf and grass weeds in crops such as rice or sugar cane which comprises applying a composition containing 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione, either together or sequentially, to a locus where weeds are present or are expected to be present.

The compositions of the present invention may be employed in many forms and are often most conveniently prepared in aqueous form immediately prior to use.

One method of preparing such a composition is referred to as "tank mixing" in which the ingredients in their commercially available form, either with or without other additives, are mixed together by the user in a quantity of water.

In addition to tank mixing immediately prior to use the compositions containing 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and 3-(7-chloro-5-fluoro-2-trifluoromethylbenzimidazol-4-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pryimidinedione may be formulated into a more concentrated primary composition which is diluted with water or other diluent before use. Such compositions may comprise a surface active agent in addition to the active ingredients and examples of such compositions are set forth below.

The herbicidal compounds of the present invention may be formulated as a granule of relatively large particle size (for example, 8/16 or 4/8 US Mesh), as water-soluble or water-dispersible granules, as powdery dusts, as wettable powders, as emulsifiable concentrates, as aqueous emulsions, as solutions, or as any of the other known types of agriculturally-useful formulations, depending on the desired mode of application to the areas in which suppression of vegetation is desired. These formulations may contain as little as 0.1%, 0.2% or 0.5% to as much as 95% or more by weight of the total of the two herbicides.

The composition can be in the form of a dispersible solution which comprises the herbicides dissolved in a water-miscible solvent with the addition of a dispersing agent. Alternatively it can comprise the herbicides in the form of a finely ground powder in association with a dispersing agent and intimately mixed with water to give a paste or cream which can if desired be added to an emulsion of oil in water to give a dispersion of the two herbicides in an aqueous oil emulsion.

Alternatively, the composition can be in the form of water-soluble or water-dispersible granules that disperse readily in water or other dispersant. Water-soluble or water-dispersible granules normally are prepared to contain about 5-80% of the herbicides, depending on the absorbency of the carrier, and usually also contain a wetting, dispersing or emulsifying agent to facilitate dispersion and may contain a preservative. Typical carriers for water-soluble or water-dispersible granules include Fuller's earth, natural clays, silicas, and other highly absorbent, readily wet inorganic diluents. For example, a useful water-soluble or water-dispersible granule formulation contains 26.71 parts of the herbicidal compound, 30.90 parts of

ammonium sulfate, 30.89 parts of continental clay, 10.00 parts of sodium lignosulfonate as a dispersant, 1.00 part of sodium dioctylsuccinate as a wetting agent and 0.50 part of citric acid as a preservative. The mixture is milled, diluted with water to form a paste and the paste is extruded and dried to produce granules.

Other alternatives that may be employed are dusts which are free flowing admixtures of the herbicides with finely divided solids such as talc, natural clays, kieselguhr, flours such as walnut shell and cottonseed flours, and other organic and inorganic solids which act as dispersants and carriers for the herbicides. These finely divided solids have an average particle size of less than about 50 microns. A typical dust formulation useful herein is one containing 1.0 part or less of the herbicidal compounds and 99.0 parts of talc.

Also useful formulations for the herbicidal compositions of the present invention are wettable powders in the form of finely divided particles that disperse readily in water or other dispersant. The wettable powder is ultimately applied to the locus where weed control is needed either as a dry dust or as an emulsion in water or other liquid. Typical carriers for wettable powders include Fuller's earth, kaolin clays, silicas, and other highly absorbent, readily wet inorganic diluents. Wettable powders are prepared to contain about 5-80% of the herbicides, depending on the absorbency of the carrier, and usually also contain a small amount of a wetting, dispersing or emulsifying agent to facilitate dispersion. For example, a useful wettable powder formulation contains 80.0 parts of the herbicidal compounds, 17.9 parts of Palmetto clay, and 1.0 part of sodium lignosulfonate and 0.3 part of sulfonated aliphatic polyester as wetting agents. Additional wetting agents and/or oils will frequently be added to a tank mix to facilitate dispersion on the foliage of the plant.

Other useful formulations for the herbicidal compositions of the present invention are emulsifiable concentrates (ECs) which are homogeneous liquid compositions dispersible in water or other dispersant, and may consist entirely of the herbicidal compounds and a liquid or solid emulsifying agent, or may also contain a liquid carrier, such as xylene, heavy aromatic naphthas, isophorone, or other non-volatile organic solvents. For herbicidal application these concentrates are dispersed in water or other liquid carriers and applied as a spray to the area to be treated. The

percentage by weight of the herbicidal compounds may vary according to the manner in which the composition is to be applied, but in general comprises 0.5 to 95% of the herbicidal compounds by weight of the total composition.

Suspension concentrate formulations may also be employed. These are similar to ECs, except that the herbicidal compounds are suspended in a liquid carrier, generally water. Suspension concentrates, like ECs, may include a small amount of a surfactant, and will typically contain the herbicidal compounds in the range of 0.5 to 95%, frequently from 10 to 50%, by weight of the total composition. For example, a useful suspension concentrate formulation contains 22.0 parts of the herbicidal compounds, 2.6 parts of an ethoxylated/propoxylated block copolymer surfactant, 0.4 part phosphate ester surfactant, 0.8 part thickening agent, 6.0 parts antifreeze agent, 0.1 antifoam agent, 0.05 part anti-bactericide and 44.0 parts distilled water. For herbicidal application, suspension concentrates may be diluted in water or other liquid vehicle, and are normally applied as a spray to the area to be treated.

Other useful formulations include suspensions of the herbicidal compounds in a relatively non-volatile solvent such as water, corn oil, kerosene, propylene glycol, or other suitable solvents.

Still other useful formulations for these herbicidal compositions include simple solutions of the herbicides in a solvent in which it is completely soluble at the desired concentration, such as acetone, alkylated naphthalenes, xylene, or other organic solvents. Granular formulations, wherein the herbicides are carried on relative coarse particles, are of particular utility for aerial distribution or for penetration of cover crop canopy. Pressurized sprays, typically aerosols wherein the herbicides are dispersed in finely divided form as a result of vaporization of a low-boiling dispersant solvent carrier may also be used.

Another useful formulation for the herbicidal compositions of the present invention is micro encapsulation. This method of formulation is a process in which either one or both of the herbicidal compounds may be encapsulated in a shell of polyurea, polyamide or amide-urea copolymer as disclosed in EP 0792100 B1 and U. S. Patent 5,583,090.

In some circumstances it may be desirable to combine two types of formulation e.g. one of the herbicidal compounds is used as an emulsifiable concentrate and the second herbicidal compound is dispersed as a powder in this concentrate.

The concentrate of the first and second herbicides (when used as the sole active components) in a composition for direct application to the crop by conventional ground methods is preferably within the range of 0.001 to 10 % by weight of the composition, especially 0.005 to 5 % by weight, but more concentrated compositions containing up to 40 % may be desirable.

Typical wetting, dispersing or emulsifying agents that may be used in the compositions of the present invention include, but are not limited to, the alkyl and alkylaryl sulfonates and sulfates and their sodium salts; alkylaryl polyether alcohols; sulfated higher alcohols; polyethylene oxides; sulfonated animal and vegetable oils; sulfonated petroleum oils; fatty acid esters of polyhydric alcohols and the ethylene oxide addition products of such esters; and the addition product of long-chain mercaptans and ethylene oxide. Many other types of useful surface-active agents are available in commerce. Surface-active agents, when used, normally comprise 1 to 15% by weight of the composition.

The invention is illustrated in the following Examples which describe experiments in which a beneficial effect was observed. The desired concentration of the active ingredients was achieved by diluting, with water, the commercially available herbicide formulation of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine (Atrazine 4L Herbicide, Agriliance, LLC) and 22% by weight suspension concentrate formulation (22 SC) of 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione, each of which contained conventional surfactants.

EXAMPLE 1

Post-emergent herbicidal evaluation of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine, 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione and Mixtures Thereof

The compositions of the present invention were tested for herbicidal efficacy as compared with the individual components used in the composition in the following manner:

Test compositions of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine (Atrazine 4L) were prepared in a spray tank that provided the appropriate application rate of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine. A crop oil concentrate was added to these test compositions.

Test compositions of 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione (22 SC) were prepared in a spray tank that provided the appropriate application rate of 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione. A crop oil concentrate was added to these test compositions.

Test compositions of mixtures of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine (Atrazine 4L) and 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione (22 SC) were prepared in a tank mix that provided appropriate rates of application of combinations of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-m bl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione. A crop oil concentrate was added to these test compositions.

The test weeds include ivyleaf morningglory (*Ipomoea hederacea*), redroot pigweed (*Amaranthus retroflexus*), velvetleaf (*Abutilon theophrasti*), common cocklebur (*Xanthium strumarium* L) and field bindweed (*Convolvulus arvenisi*).

For post-emergence testing, three disposable fiber flats (8 cm.times.15 cm.times.25 cm) for each rate of application of each herbicide were filled to an approximate depth of 6.5 cm with steam-sterilized sandy loam soil. The soil was leveled and impressed with a template to provide five evenly spaced furrows 13 cm long and 0.5 cm deep in each flat. Seeds of the test weeds were planted in the furrows of the flats. The five-row template was employed to firmly press the seeds into place. A topping soil of equal portions of sand and sandy loam soil was placed uniformly on top of each flat to a depth of approximately 0.5 cm. Flats for post-emergence testing were placed in a greenhouse and watered for 16 days, thus allowing the seeds to germinate and the foliage to develop.

The flats were placed on a conveyor belt and the conveyor belt fed under a spray nozzle mounted about ten inches above the postemergent foliage. The spray of herbicidal solution was commenced and once stabilized; the flats were passed under the spray at a speed to receive a coverage equivalent of 30 gallons per acre. The application rates are those shown in Table 1 below for the individual herbicidal solutions and mixtures thereof. The post-emergence flats were immediately placed in the green-house and not watered until 24 hours after treatment with the test solution. Thereafter they were regularly watered at ground level. After 6, 14 and 27 days the plants were examined and the phytotoxicity data were recorded. Untreated control flats were included in the test.

Percent control was determined by a method similar to the 0 to 100 rating system disclosed in "Research Methods in Weed Science," 2nd ed., B. Truelove, Ed.; Southern Weed Science Society; Auburn University, Auburn, Ala., 1977. The rating system is as follows:

Herbicide Rating System		
Rating Percent Control	Description of Main Categories	Weed Description
0	No Effect	No weed control
10		Very poor weed control
20	Slight Effect	Poor weed control
30		Poor to deficient weed control
40		Deficient weed control
50	Moderate Effect	Deficient to moderate weed control
60		Moderate weed control
70		Control somewhat less than satisfactory
80	Severe	Satisfactory to good weed control
90		Very good to excellent weed control
100	Complete Effect	Complete weed destruction

TABLE 1
Post-emergence Herbicidal Activity 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine (Cmpd A), 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione (Cmpd B) and Mixtures Thereof

Rate of application pound/acre		Weed Species* % Control at 6 Days After Treatment				
Cmpd A	Cmpd B	A	B	C	D	E
--	--	0	0	0	0	0
0.25	--	20	0	0	0	0
0.5		30	10	10	10	10
1.0	--	80	20	30	10	5
--	0.008	90	30	75	10	20
--	0.016	95	80	75	20	70
.025	0.008	**100**	**60**	60	**60**	**65**
0.5	0.008	75	**40**	75	**20**	**25**
1.0	0.008	**100**	**70**	70	**50**	**60**
0.25	0.016	80	75	75	**60**	50
0.5	0.016	**100**	**85**	75	**60**	**75**
1.0	0.016	**100**	**85**	75	**30**	**75**

Rate of application pound/acre		Weed Species* % Control at 14 Days After Treatment				
Cmpd A	Cmpd B	A	B	C	D	E
--	--	0	0	0	0	0
0.25	--	20	2	10	15	2
0.5		40	20	25	40	5
1.0	--	90	20	85	75	10
--	0.008	85	10	60	30	20
--	0.016	90	75	70	30	70
.025	0.008	**100**	**25**	20	**50**	20
0.5	0.008	60	20	30	25	**30**
1.0	0.008	**100**	**60**	70	**80**	**50**
0.25	0.016	90	30	70	20	60
0.5	0.016	**100**	**80**	60	**60**	**80**
1.0	0.016	**100**	70	70	50	70

Rate of application pound/acre		Weed Species* % Control at 27 Days After Treatment				
Cmpd A	Cmpd B	A	B	C	D	E
--	--	0	0	0	0	0
0.25	--	10	5	10	20	2
0.5		20	20	40	50	5
1.0	--	90	25	75	75	10
--	0.008	75	10	50	25	10
--	0.016	80	70	60	30	70
.025	0.008	**100**	**25**	20	**50**	**20**
0.5	0.008	25	10	30	20	**25**
1.0	0.008	100	30	60	75	**50**
0.25	0.016	**90**	25	**70**	25	70
0.5	0.016	**100**	70	40	**60**	**80**
1.0	0.016	**100**	50	60	60	70

Highlighted numbers indicate more advantageous herbicidal properties.
* A – field bindweed. B - – common cocklebur, C – ivyleaf morningglory, D – redroot pigweed, E – velvetleaf

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Those of ordinary skill in the art will appreciate that variations of the invention may be used and that it is intended that the invention may be practiced otherwise than as specifically described herein. Accordingly, this invention includes all modifications encompassed within the spirit and scope of the invention as defined by the following claims.

What is claimed is:

1. An herbicidal composition, comprising a first herbicide and a second herbicide in which the first herbicide is 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine and the second herbicide is 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione.

2. The composition according to claim 1 wherein the ratio of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine to 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione is in the range of from 160:1 to 3:1.

3. The composition according to claim 2, wherein the ratio of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine to 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione is in the range of from 63:1 to 12:1.

4. A method for controlling weeds comprising applying a composition of claim 1 to a locus where weeds are present or are expected to be present.

5. A method according to claim 4 wherein the ratio of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine to 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione in the composition is in the range of from 160:1 to 3:1.

6. A method according to claim 5 wherein the ratio of 6-chloro-N-ethyl-N'-(1-methylethyl)-1,3,5-triazine-2,4-diamine to 3-(4-chloro-6-fluoro-2-trifluoromethylbenzimidazol-7-yl)-1-methyl-6-trifluoromethyl-2,4(1H,3H)pyrimidinedione is in the range of from 63:1 to 12:1.