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(54) DINITROANILINE DERIVATIVES

(71) We, ELI LILLY AND COMPANY, a corporation of the State of Indiana, United States of America, having a principal place of business at 307 East McCarty Street, City of Inianapolis, State of Indiana, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

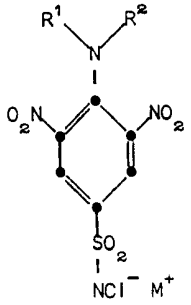
This invention relates to dinitroanilines derivatives.

In accordance with the invention there are provided certain sodium and potassium salts of N¹-chloro-3,5-dinitrosulfanilamides which exhibit potent preemergent herbicidal activity and are also useful for the control of plant fungi, especially *Plasmopara viticola*, the causative organism of downy mildew of grape.

Herbicidal 2,6-dinitroanilines have long been known as evidenced by United States Patents such as Nos. 3,111,403; 3,257,190; and 3,332,769. In United States Patent No. 3,367,949 there is disclosed a class of 2,6-dinitroanilines which are more properly named as 3,5-dinitrosulfanilamides. The compounds of U.S. Patent No. 3,367,949 serve as starting materials for the preparation of the compounds of the present invention.

Another class of 2,6-dinitroanilines containing a sulfonyl group in the molecule is disclosed in United States Patent No. 3,321,292. However, the compounds there described contain a methylsulfonyl group rather than a sulfonamido group and are only distantly related to the compounds of the present invention.

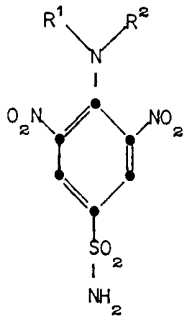
This invention provides N¹-chloro-3,5-dinitrosulfanilamides having the formula



I

wherein each of R^1 and R^2 is independently C_1—C_6 alkyl, C_2—C_4 alkenyl, halo C_1—C_6 alkyl or cyclopropylmethyl; and M is sodium or potassium.

The invention also provides a process for preparing the compounds of Formula I which comprises reacting a compound of the formula



II

wherein R^1 and R^2 are as defined above, with sodium hypochlorite in an aqueous water-miscible solvent, and optionally reacting the product so obtained with potassium hydroxide in an aqueous, water-miscible solvent.

The compounds of Formula I are useful as preemergent herbicides and in the control of phytopathogenic fungi. A method for the control of undesired vegetation by the application of a herbicidally effective amount of a compound of Formula I and a method for reducing the incidence and severity of grape downy mildew which comprises applying a fungicidally effective amount of a compound of Formula I to the foliage of the host plants are described. Pesticidal compositions comprising a compound of Formula I and an inert carrier are also described.

In the above description, all of the terms employed have the meanings normally ascribed to them in the chemical art.

The N^1-chloro-3,5-dinitrosulfanilamide sodium salts of Formula I are prepared by treating the corresponding 3,5-nitrosulfanilamide of Formula II with sodium hypochlorite. The preparation of the starting 3,5-dinitrosulfanilamides is described in United States Patent No. 3,367,949.

In preparing the compounds, a 3,5-dinitrosulfanilamide in a suitable solvent may be reacted with sodium hypochlorite in the presence of sodium hydroxide at a temperature between 0°C. and ambient temperature. A suitable solvent is a water-miscible solvent in which the 3,5-dinitrosulfanilamide is soluble. A preferred solvent is ethanol. Other suitable solvents include methanol, dioxane, and ethylene glycol dimethyl ether.

For example, a solution of the 3,5-dinitrosulfanilamide in the organic solvent is made basic by the addition of aqueous sodium hydroxide solution. The solution is then cooled to a temperature between 0°C. and the ambient temperature, and aqueous sodium hypochlorite is added. Preferably, a molar excess of between 20 and 100% of sodium hypochlorite is employed although an equimolar amount may be used. After the addition of the sodium hypochlorite is completed the reaction is allowed to proceed for a period of from 15 minutes to 2 hours. Generally, the reaction will be complete in 30 minutes. The product precipitates during the course of the reaction and is collected by a known procedure, such as filtration.

The preparation of exemplary sodium salts will be illustrated by the following examples. The examples are merely illustrative and do not limit the scope of this invention.

Example 1.

To a solution of 100 g. of 3,5-dinitro-N^4,N^4-dipropylsulfanilamide in 500 ml. of denatured ethanol and 500 ml. of 2N sodium hydroxide solution cooled to 5°C. was added rapidly dropwise 500 ml. of a 5.25% aqueous solution of sodium hypochlorite. An additional 250 ml. of water was added rapidly dropwise and the mixture was stirred in the cold for one-half hour. The product which had separated was collected by filtration. The yield was 126 g. and the product had a melting point of 88—94°C. It was identified by nuclear magnetic resonance spectroscopy as N^1-chloro-3,5-dinitro-N^4, N^4-dipropylsulfanilamide, sodium salt.

Analyses: Calculated for $C_{12}H_{16}N_4O_6SClNa$: C, 35.78; H, 4.00; N,13.91; Cl, 8.80.
Found: C, 35.58; H, 4.25; N, 13.95; Cl, 8.87.

The following additional compounds were prepared following the procedure of Example 1 and using appropriate starting materials. Many of the compounds melt over a broad range, probably due to the fact that the compounds are highly solvated.

Example 2.

N^1-chloro-3,5-dinitro-N^4-ethyl-N^4-(2-methylallyl)sulfanilamide, sodium salt, m.p. 146—148°C., from 3,5-dinitro-N^4-ethyl-N^4-(2-methylallyl)sulfanilamide, yield 74%.

Example 3.

N^1-chloro-3,5-dinitro-N^4-ethyl-N^4-propylsulfanilamide, sodium salt, m.p. 88—91°C., from 3,5-dinitro-N^4-ethyl-N^4-propylsulfanilamide, yield 96%.

Example 4.

N^1-chloro-3,5-dinitro-N^4-(2-methylallyl)-N^4-propylsulfanilamide, sodium salt, m.p. 80—88°C., from 3,5-dinitro-N^4-(2-methylallyl)-N^4-propylsulfanilamide, yield 86%.

Example 5.

N^1-chloro-N^4-cyclopropylmethyl-3,5-dinitro-N^4-propylsulfanilamide, sodium salt, m.p. 88—92°C., from N^4-cyclopropylmethyl-3,5-dinitro-N^4-propylsulfanilamide, yield 100%.

Example 6.

N^1-chloro-N^4-(2-chloroethyl)-3,5-dinitro-N^4-propylsulfanilamide, sodium salt, m.p. 83—88°C., from N^4-(2-chloroethyl)-3,5-dinitro-N^4-propylsulfanilamide, yield 86%.

Example 7.

N^4-allyl-N^1-chloro-3,5-dinitro-N^4-propylsulfanilamide, sodium salt, m.p. 83—89°C., from N^4-allyl-3,5-dinitro-N^4-propylsulfanilamide, yield 48%.

Example 8.

N^4-butyl-N^1-chloro-3,5-dinitro-N^4-ethylsulfanilamide, sodium salt, m.p. 78—81°C., from N^4-butyl-3,5-dinitro-N^4-ethylsulfanilamide, yield 34%.

Example 9.

To a solution of 1.9 g. of N^4-(2-chloroethyl)-3,5-dinitro-N^4-propylsulfanilamide in 15 ml. of denatured alcohol and 15 ml. of 2N sodium hydroxide solution in an ice bath was added dropwise 15 ml. of a 5.25 percent aqueous solution of sodium hypochlorite. The mixture was stirred in the ice bath for 30 minutes, 15 ml. of water was added, and the mixture was stirred for 30 minutes more. The reaction mixture was then placed on a rotary evaporator under vacuum at ambient temperature for 1 hour. The desired product crystallized and was collected by filtration. The yield was 1.9 g. of N^1-chloro-N^4-(2-chloroethyl)-3,5-dinitro-N^4-propylsulfanilamide, sodium salt, m.p. 83—88°C. The identity of the compound was confirmed by nuclear magnetic resonance spectroscopy.

The potassium salts of this invention are prepared by reacting the corresponding sodium salt with potassium hydroxide. The reaction is carried out in a suitable water-miscible solvent such as ethanol, methanol, dioxane, or ethylene glycol dimethyl ether. The preferred solvent is ethanol. The reaction may be carried out at a temperature ranging from 0° to ambient temperature, preferably 20° to 30°C.

The preparation of potassium salts is illustrated by the following example which is not to be interpreted as limiting the scope of the invention.

Example 10.

A solution of 5 g. of N^1-chloro-3,5-dinitro-N^4,N^4-dipropylsulfanilamide, sodium salt, in 20 ml. of denatured ethanol was added dropwise to 50 ml. of cold 2N potassium hydroxide. The mixture was stirred at room temperature for 40 minutes and then was cooled. The product which separated was collected by filtration. The yield was 3.6 g., m.p. 148—152°C., *dec.* It was confirmed by nuclear magnetic resonance spectroscopy and elemental analysis to be N^1-chloro-3,5-dinitro-N^4,N^4-dipropylsulfanilamide, potassium salt.

Analyses: Calculated for $C_{12}H_{16}N_4O_6SClK$: C, 34.41; H, 3.85; N, 13.37; Cl, 8.46.
Found: C, 34.63; H, 3.98; N, 13.15; Cl, 8.73.

The preferred compounds of Formula I are those wherein each of R^1 and R^2 is C_1—C_6 alkyl. Particularly preferred are those compounds wherein each of R^1 and R^2 is independently selected from ethyl, *n*-propyl and *n*-butyl. The sodium salts are preferred over the potassium salts.

Another preferred group of compounds are those in which R^1 is C_1—C_6 alkyl, especially ethyl or n-propyl and R^2 is C_2—C_4 alkenyl, especially allyl or 2-methylallyl. Again, the sodium salts are preferred over the potassium salts.

Still another preferred compound is the compound of Example 5.

The compounds of Formula I are useful as plant fungicides and herbicides. The compounds are particularly effective in reducing the incidence and severity of grape downy mildew infections when the compounds are applied to the foliage of the host plant

in a fungicidally effective amount. The amount of compound which is effective is dependent upon the particular compound employed and the severity of the infection. Preferably, the compounds are applied at a rate of from 10 g. to 2 kg. per hectare. When a liquid composition is used the active component is preferably present within the range of from 10 to 2000 ppm by weight. When dusts are used the active compound is preferably present at from 0.05 to 5 percent or more by weight of the composition.

For herbicidal use the compounds are applied preemergently at a rate from 0.25 to 10 kg. per hectare. The compounds may be either incorporated into the soil or surface applied. In general, less compound is required when the soil incorporation method is employed.

For either fungicidal or herbicidal use the present compounds are preferably employed in liquid, powder, or dust compositions containing one or more of such compounds and an inert diluent or carrier. The composition may also contain a surface active agent. Such compositions are prepared in accordance with standard agricultural practices by modifying the present compounds with one or more of a plurality of additives including organic solvents, petroleum distillates, water or other liquid carriers, surface active dispersing agents, and finely divided inert solids. In such compositions, the active compound can be present in a concentration from 2% to 98% by weight. Surface-active agents are present in amounts from 0.1% to 20%, and inert carriers in amounts up to 97.9% by weight.

In the preparation of dust compositions, the chlorosulfanilamides of Formula I can be compounded with any of the finely divided solids such as pyrophyllite, talc, chalk or gypsum. The finely divided carrier is ground or mixed with the chlorosulfanilamide or wet with a solution of the chlorosulfanilamide in a volatile organic solvent. Dust compositions containing the active compounds of this invention may also be prepared with various solid surface active dispersing agents such as fuller's earth, bentonite, attapulgite, and similar clays. Depending upon the proportions of ingredients, these dust compositions can be employed as concentrates and subsequently diluted with an additional solid surface active dispersing agent or with pyrophyllite, chalk, talc, gypsum, or the like to obtain the desired amount of active chlorosulfanilamide. Such dust compositions may also be dispersed in water with or without the aid of dispersing agents to form liquid, sprayable mixtures.

The chlorosulfanilamide compounds or a liquid or dust concentrate composition containing the active compounds can be incorporated in intimate mixture with surface active dispersing agents such as nonionic emulsifying agents to form spray compositions. Such compositions may be employed as such or may be dispersed in liquid carriers to form diluted sprays containing the active compound in any desired amount.

In accordance with standard practice, the active chlorosulfanilamide compound may be compounded with a suitable water-immiscible organic liquid and a surface active dispersing agent to produce emulsifiable concentrates which can be further diluted with water and/or oil to form spray mixtures in the form of oil-water emulsions. Preferred dispersing agents to be employed in these compositions are oil soluble and include the nonionic emulsifiers such as condensation products of alkylene oxides with phenols, sorbitan esters, complex ether alcohols, and the like. Suitable organic liquids which can be employed include petroleum oils and distillates, toluene and synthetic organic oils. The surface active dispersing agents are usually employed in liquid compositions in an amount of from 0.1 to 20% by weight of the composition.

The main advantage offered by the compounds of Formula I is their high degree of solubility in water and in economical mixtures of water and water-miscible solvents. Because of this property, the compounds are much more easily and economically formulated than are other, previously-known dinitroaniline herbicides and fungicides. Accordingly, the preferred herbicidal and fungicidal compositions of the compounds of Formula I are solutions in water and in aqueous water-miscible solvents.

Such solvents as C_1—C_3 alkanols, particularly ethanol, propanol and isopropanol, C_2—C_6 alkyl ketones, particularly acetone and methyl ethyl ketone, and glycols such as ethylene glycol are particularly advantageous water-miscible solvents for the formulation of such compositions.

In general, aqueous solutions according to this invention contain from 5% to 50% by weight of a compound of Formula I, dissolved in 50% to 95% by weight of water or aqueous water-miscible solvent.

When an aqueous solution composition of a compound of Formula I is used, no surface-active agents are normally necessary. The concentrated composition may be merely diluted with additional water to obtain the desired concentration for use. However, such adjuvants as spreader-stickers and the like may be added, if desired, to assist the diluted composition in adhering to the foliage of the treated plants.

The herbicidal utility of the compounds of this invention was shown by greenhouse tests using both surface application and soil incorporation methods. The compounds were prepared for use by dissolving in a 1:1 acetone/ethanol solvent containing a small amount of Toximul (Registered Trade Mark) R and S surfactants (Toximul R and Toximul S are sulfonate/nonionic blends of emulsifiers for pesticidal formulations. They are manufactured by the Stepan Chemical Company of Northfield, Illinois.) The solutions were diluted with deionized water containing 1000 ppm. Toximul R and S to obtain the desired application rates.

The test plants were grown in galvanized pans 31.5 cm. long, 21.5 cm. wide, and 8.0 cm. deep. Seeds of 14 plants were planted, seven species per pan. The seeds were planted in rows perpendicular to the long axis of the pan, one species per row. The plants employed are as follows:

A) corn (*Zea mays*)
B) foxtail millet (*Setaria italica*)
C) grain sorghum (*Sorghum vulgare*)
D) barnyardgrass (*Echinochloa crus-galli*)
E) rice (*Oryza sativa*)
F) wild oat (*Avena fatua*)
G) wheat (*Triticum aestivum*)
H) soybean (*Glycine max*)
I) pigweed (*Amaranthus retroflexus*)
J) cotton (*Gossypium barbadense*)
K) jimsonweed (*Datura stramonium*)
L) sugar beet (*Beta vulgaris*)
M) lambsquarters (*Chenopodium album*)
N) cucumber (*Cucumis sativus*)

For surface application of the test chemicals, the pans were filled two-thirds full with autoclaved greenhouse potting soil. The seeds were laid in rows and were covered with 0.5 to 1.0 cm. of autoclaved soil. The soil surface was then sprayed with the apropriate application rate of the test compound.

For soil incorporation application, the appropriate rate of test compound was mixed into 4.75 l. of autoclaved greenhouse potting soil while the soil was tumbling in a modified cement mixer. Approximately 3.785 l. of treated soil was added to a pan, the seeds were planted in rows perpendicular to the long axis of the pan and were then covered over with the remaining treated soil.

The pans were placed in a greenhouse and the plants were allowed to grow for a period of from 18 to 21 days. The herbicidal effects of the test compounds were then evaluated using a rating scale of 0 to 10, where 0 equals no injury and 10 equals complete kill or no emergence, with 1 to 9 indicating gradations between the two extremes.

The results obtained with representative compounds of this invention are reported in Tables 1 and 2. The test compounds are identified by their example numbers while the plant species are identified by the appropriate letter designation. Table 1 reports the soil incorporation results while Table 2 reports the surface application results.

TABLE 1

SOIL INCORPORATION APPLICATION

Compound No.	Rate kg./ha.	A	B	C	D	E	F	G	H	I	J	K	L	M	N
1	0.56	2	7	3	8	5	3	3	0	7	2	0	3	5	0
	1.12	3	9	4	10	8	5	3	2	10	3	2	7	7	0
	2.24	4	10	9	10	9	6	5	3	10	4	4	7	10	3
2	0.56	2	2	2	8	5	3	2	0	7	0	0	4	3	0
	1.12	4	7	3	9	6	5	3	0	8	0	2	6	9	0
	2.24	3	10	4	10	9	6	4	2	10	3	4	9	10	2
3	0.56	2	6	2	8	5	3	2	2	10	2	2	7	10	0
	1.12	2	10	3	10	8	5	2	2	10	3	3	8	10	2
	2.24	3	10	6	10	9	7	4	3	10	4	5	9	10	3
4	0.56	2	7	3	8	7	3	2	0	7	0	0	8	10	0
	1.12	3	9	4	8	7	2	3	2	10	1	2	7	10	2
	2.24	3	10	7	9	8	4	4	3	10	3	3	9	10	2
5	0.56	0	4	2	7	5	0	2	0	3	0	0	2	3	0
	1.12	2	7	3	8	7	4	3	2	10	2	2	5	10	2
	2.24	4	9.5	5	10	8	5	4	2	10	3	3	7	10	3
6	0.56	0	3	2	6	5	2	0	0	7	0	0	2	8	0
	1.12	2	7	3	8	5	3	2	2	10	2	0	3	10	0
	2.24	3	9.5	7	8	7	4	4	2	10	2	2	6	10	2
7	0.56	0	2	0	2	3	0	0	0	5	0	0	2	5	0
	1.12	2	5	2	7	5	2	2	0	7	0	0	2	6	0
	2.24	3	9	4	8	7	4	3	0	8	2	2	7	10	0
8	0.56	0	3	2	6	3	0	0	0	6	0	0	2	8	0
	1.12	3	7	3	8	5	2	2	0	9	0	2	5	9	0
	2.24	3	9.5	5	0	8	4	3	0	10	3	3	7	10	2

TABLE 2

SURFACE APPLICATION

Compound No.	Rate kg./ha.	A	B	C	D	E	F	G	H	I	J	K	L	M	N
1	1.12	0	9	0	8	3	4	0	2	7	0	0	2	8	0
	2.24	0	9	5	8	4	7	0	3	8	0	0	7	10	0
	4.48	2	10	7	10	8	7	3	4	9	0	2	7	10	0
2	1.12	0	9	0	8	4	4	0	0	7	0	0	4	9	0
	2.24	0	9	4	9	4	7	2	2	7	0	0	5	10	0
	4.48	2	9.5	6	10	6	7	3	2	8	2	4	7	10	0
3	1.12	0	9	0	8	3	5	0	0	6	0	0	4	5	0
	2.24	2	9	4	9	8	7	2	0	7	0	2	7	10	0
	4.48	2	9	5	10	8	7	4	0	7	0	3	7	10	2
4	1.12	0	8	0	8	3	4	0	0	6	0	0	3	6	0
	2.24	0	9	2	9	3	5	2	0	7	0	2	3	8	0
	4.48	2	9	4	10	5	7	2	0	8	0	2	6	9	0
5	1.12	0	9	0	8	2	5	0	0	6	0	0	2	6	0
	2.24	0	8.5	2	9	3	6	0	0	8	0	0	4	9	0
	4.48	3	9	4	9	5	7	2	3	8	2	3	5	10	0
6	1.12	0	8	2	8	2	2	0	0	6	0	0	0	5	0
	2.24	0	9	3	8	4	4	0	0	8	0	0	4	9	0
	4.48	0	9	5	8	3	6	2	2	7	2	0	5	9	0
7	1.12	0	8	0	7	0	2	0	0	5	0	0	2	7	0
	2.24	0	9	2	7	3	2	0	0	7	0	0	2	8	0
	4.48	0	9	4	9	4	4	2	0	9	0	0	5	10	0
8	1.12	0	8	0	8	3	2	0	0	7	0	0	2	8	0
	2.24	0	9	3	9	5	3	2	1	7	0	2	5	8	0
	4.48	2	9	3	9	5	5	2	2	8	0	0	4	8	2

The usefulness of the compounds of this invention to reduce the incidence and severity of grape downy mildew was also demonstrated in greenhouse tests. The test compounds were formulated by mixing 70 mg. of the compound with 2 ml. of a solution prepared from 500 ml. of acetone, 500 ml. of ethanol, and 100 ml. of Tween 20 (Registered Trade Mark). (Tween 20 is a polyoxyethylene sorbitan monolaurate made by Atlas Chemical Division of ICI America, Inc., Wilmington, Delaware.) The sample was then diluted with 175 ml. of deionized water containing one drop of Dow Corning (Registered Trade Mark) antifoam C emulsion per 2 l. of water. (Dow Corning antifoam C is a silicone complex antifoaming agent made by Dow Corning Corporation, Midland, Michigan.) The final formulation contained 400 ppm. of the test compound, 10,000 ppm. of organic solvents, and 1,000 ppm. of Tween 20, and was further diluted with deionized water to obtain lower concentrations of the test compound.

On the day the test was started, young expanding leaves were detached from grape vines grown in the greenhouse. One leaf was placed bottom side up in a plastic petri plate containing a Whatman filter paper placed on top of an expanded plastic mat to keep the leaf above the water flooding the bottom of the petri plate. A water soaked wad of cotton was wrapped around the petiole base of the leaf. The test chemical at the desired concentration was sprayed on the underside of the leaf to run off and allowed to dry. As soon as the leaf dried it was inoculated with a conidial suspension of *Plasmopara viticola* using a DeVilbiss sprayer. Conidia were obtained from recently infected leaf tissue stored in the chill room at 5°C. The conidia were washed off the leaf's surface with a brush and suspended in deionized water to obtain the inoculation suspension.

After inoculation the plates were placed in a moist chamber. Cool white fluorescent lamps above the moist chamber hood provided 200 to 400 foot-candles of light to the leaves on a cycle of 14 hours of light and 10 hours of darkness at 68°F. The leaves were

observed for disease symptoms and the results were recorded seven days after treatment. A rating scale of 1 to 5 was used to record the results where 1 equals severe disease (or no control), 2 equals moderate disease, 3 equals slight disease, 4 equals very slight disease, and 5 equals no disease (or 100% control).

The results of testing representative compounds of this invention are reported in Table 3. Blanks in the table indicate that the compound was not tested at that rate of application.

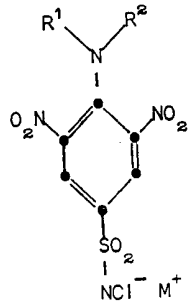
TABLE 3

ACTIVITY AGAINST GRAPE DOWNY MILDEW

Compound No.	Rate, ppm 400	200	100	50	25	12.5
1	5	5	4+	5	5	4+
2		4+	4+	4−	4	
3		4+	2+	2		
4		4+	3−	2+		
5		4	5	3		
6		3	3−	3−		
7	4+	4+	4+			
8	5	4+	5	4	3−	1

WHAT WE CLAIM IS:—

1. A N^1-chloro-3,5-dinitrosulfanilamide compound of the formula



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wherein each of R^1 and R^2 is independently C_1—C_6 alkyl, C_2—C_4 alkenyl, halo C_1—C_6 alkyl or cyclopropylmethyl; and M is sodium or potassium.

2. A compound of claim 1 wherein each of R^1 and R^2 is C_1—C_6 alkyl.

3. A compound of claim 2 wherein each of R^1 and R^2 is ethyl, *n*-propyl or *n*-butyl.

4. A compound of claim 1 wherein R^1 is C_1—C_6 alkyl and R^2 is C_2—C_4 alkenyl.

5. A compound of claim 4 wherein R^1 is ethyl or *n*-propyl and R^2 is allyl or 2-methylallyl.

6. A compound of any of claims 1—5 wherein M is sodium.

7. N^1-Chloro-3,5-dinitro-N^4,N^4-dipropylsulfanilamide, sodium salt.

8. N^4-Butyl-N^1-chloro-3,5-dinitro-N^4-ethylsulfanilamide, sodium salt.

9. N^1-Chloro-3,5-dinitro-N^4-ethyl-N^4-(2-methylallyl)sulfanilamide, sodium salt.

10. N^4-Allyl-N^1-chloro-3,5-dinitro-N^4-propylsulfanilamide, sodium salt.

11. N^1-Chloro-N^4-cyclopropylmethyl-3,5-dinitro-N^4-propylsulfanilamide, sodium salt.

12. An herbicidal and fungicidal composition which comprises a compound of Formula I as defined in any one of claims 1 to 11 in association with an insert carrier therefor.

13. A composition according to claim 12 in the form of a solution in water or an aqueous water-miscible solvent.

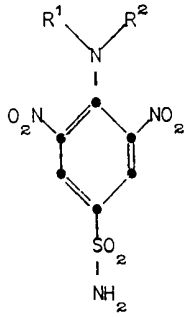
14. An herbicidal method for the control of undesired vegetation in an area which comprises applying to said area an herbicidally-effective amount of a compound of Formula I as defined in any of claims 1 to 11 or a composition according to claim 12 or 13.

15. A method according to claim 14 wherein the amount of the compound of Formula I is from 0.25 kg./ha. to 10 kg./ha.

16. A fungicidal method for reducing the incidence and severity of grape downy mildew which comprises applying to the foliage of the host plant a fungicidally effective amount of a compound of Formula I as defined in any one of claims 1 to 11 or a composition according to claim 12 or 13.

17. A method according to claim 16 wherein the amount of the compound of Formula I is from 10 g/ha. to 2 kg./ha.

18. A process for preparing a compound of Formula I as defined in any of claims 1 to 11 which comprises reacting a compound of the formula



II

wherein R^1 and R^2 are as defined in claim 1, with sodium hypochlorite in an aqueous water-miscible solvent, and optionally reacting the product so obtained with potassium hydroxide in an aqueous, water-miscible solvent.

19. A process according to claim 18 substantially as hereinbefore described with reference to any one of the Examples.

20. A compound of Formula I as defined in claim 1 whenever prepared by a process according to claim 18 or 19.

21. A compound of Formula I as defined in claim 1 as described in any one of the Examples.

22. A composition according to claim 12 substantially as hereinbefore described.

23. A method according to claim 14 substantially as hereinbefore described.

24. A method according to claim 16 substantially as hereinbefore described.

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