

PATENT SPECIFICATION

(11) 1 429 080

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(21) Application No. 43855/73 (22) Filed 18 Sept. 1973

(23) Complete Specification filed 19 Oct. 1973

(44) Complete Specification published 24 March 1976

(51) INT CL² A01N 9/24

(52) Index at acceptance

A5B 1A3B 1A3C 1A5A1 1C12C 1C14 1C15B1 1C15B3
1C1A6 1C9B 1C9D

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(54) FUNGICIDAL COMPOSITION

(71) We, TAKASAGO PERFUMERY CO., LTD., a Japanese Company, of No. 4-1, Hatchobori 1-Chome, Chuo-Ku, Tokyo, Japan, 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 a fungicide for use against fungi attacking vascular crop plants.

Many controlling agents against pathogenic fungi which attack crops have been well known. However, there are problems in their effects on controlling *Fusarium fungus* which causes root-rot of black pepper; methyl - 1 - (butyl - carbamoyl) - 2 - benzimidazol - carbamate, marketed by E. I. Du Pont as a wettable powder under the trade mark "Benlate", is, for example, actually used but with unsatisfactory effect.

Moreover, for controlling downy mildew (*Pseudoperonospora humuli* Wilson) of hop, organic phosphides have been used and recently antibiotic agents such as kasugamycin and polyoxin have been studied. However, none of these can provide satisfactory mildew control.

It has been reported that gum trees suffer serious damage due to mouldy rot caused by *Ceratocystis fimbriata*. The mouldy rot due to *C. fimbriata* generally occurs together with olive brown mycelium when the bark of gum trees is cut for collecting a gum milk and, after infection with these fungi, the cut portion becomes hardened and the bark for collecting the gum milk is no longer reproduced.

In the past, a dilute aqueous solution of Benlate wettable powder or Difolton (N-tetrachloroethyl thiotetrahydrophthalimide) has been sprayed or applied to the cut portion to control these infectious fungi, but the fungicidal effect of these agents has not been adequate.

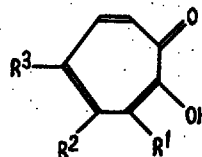
It is also known that coffee plants suffer serious damage due to an infection caused by *Hemileia vastatrix* and, in particular, the damage is remarkable in flat land. Agents

containing copper are generally used for controlling this infection, but the best way to avoid the above infection is to permit the coffee plants to grow to a large size whilst keeping them healthy.

Typical infections in cacao include such plant diseases as witches' broom caused by *Marasmius perniciosus* and white root disease caused by *Rigidoporus lignosis*. In particular, witches' broom causes a great deal of damage, and the whole tree of cacao often acquires a "broom-like" appearance if the control of this disease is insufficient. Fungicidal agents which sufficiently control this infection have not yet been developed, and the only way to control this infection presently available is to cut off the infected portions.

It is said that the root-rot of black pepper is caused by the invasion of *Fusarium fungus*, a soil micro-organism, from the cut end of a cutting or from a wound caused by a nematode, which fungus propagates itself in vessels to close the same, thereby stopping the growth of the plant or producing poisonous gas, so that the roots or stems are attacked and finally leaves fall off the plant and dies. When a pepper plant is attacked with the root-rot, it takes about 2 to 3 years before its symptoms can be confirmed, by which time it is too late to cure the disease; therefore, the only treatment is for the diseased plant to be dug up and burned to ash in order to prevent the infection from spreading. There is a great need for an effective fungicide against root-rot.

We have now found that a compound having the general formula:



wherein R¹, R² and R³ are each a hydrogen atom or an alkyl radical having 1 to 3 carbon atoms, is effective for controlling pathogenic

fungi which attack vascular plants, for example fungi such as *Fusarium* which causes root-rot of the pepper plant, downy mildew which attacks hop, *C. fimbriate* which causes mouldy rot of the gum tree, *H. vastatrix* which causes orange rust of the coffee plant, *M. perniciosus* which causes witches' broom, *Botrytis cinerea* Pers which attacks the strawberry plant, *Alternaria kikuchiana* which attacks pear, *Piricularia oryzae* which attacks rice, *Cladosporium fulvum* which attacks tomato, *Fusarium oxysporum* which attacks tomato and *Venturia inaequalis* which attacks apple, and is also effective when applied to the soil for controlling a wide range of pathogenic fungi which attack vascular plants.

The compounds used as fungicides in this invention are those having a tropolone skeleton. The general formula described above in which R^1 , R^2 and R^3 are each a hydrogen atom represents tropolone; that in which R^1 is an isopropyl radical and R^2 and R^3 are each a hydrogen atom represents α -thujaplicin; that in which R^2 is an isopropyl radical and R^1 and R^3 are each a hydrogen atom represents β -thujaplicin (hinokitiol); and that in which R^3 is an isopropyl radical and R^1 and R^2 are each a hydrogen atom represents γ -thujaplicin.

Hinokitiol is naturally present in Formosan hinoki oil, Japanese hiba oil and Western red cedar oil. α -, β - and γ -thujaplicins have all been described as having antifungal properties (see Dictionary of Organic Compounds, 5 (1965), page 3060).

A process for synthesizing these compounds is disclosed by Dr. Bartlett in U.S. Patent No. 3,448,155. According to this process, tropolone is produced by reacting cyclopentadiene directly with dichloroacetyl chloride. Thujaplicin is obtained by reacting cyclopentadiene with isopropylbromide to produce isopropyl cyclopentadiene, reacting it with dichloroacetyl chloride and treating with glacial acetic acid and sodium acetate. All of the above products are in the form of crystals.

The fungicidal composition of the invention contains as active ingredient at least one compound according to the aforesaid general formula and is in the form of a liquid emulsifiable concentrate which contains 2 to 25% by weight of active ingredient or in the form of solid granules which incorporates a solid carrier and contains 0.1 to 5% by weight of active ingredient.

The invention also provides a method of controlling a fungus disease of a vascular plant which comprises applying a preparation containing a tropolone of the aforesaid general formula to the surface of the plant, injecting it into the plant or applying it to the soil in which the plants are growing.

An emulsifiable concentrate is a solution of one or more of the aforesaid tropolones

together with a surface-active agent, which solution can be emulsified in water before use. For use, it is preferred that the emulsifiable concentrate is diluted with water to about 1/200 to 1/20,000 solution for spraying. Excellent results are obtained through the absorption of the active ingredient by directly treating the plant bodies with a solution of a concentration of about 1/7,500 to 1/10,000 or by injecting the solution into the plant bodies, particularly the cut end of a stem or branch.

Solvents which may be suitably used in the composition are one or more organic compounds such as alcohols (e.g., ethyl alcohol, propylene glycol or diacetone alcohol), ketones (e.g., acetone or methyl ethyl ketone), aliphatic hydrocarbons (e.g. hexane or petroleum benzene), aromatic hydrocarbons (e.g., benzene or toluene), halogenated hydrocarbons (e.g., chloroform or dichloroethane) and esters (e.g., ethyl acetate or butyl acetate).

Solid carriers which may be used in the composition include vegetable powders (e.g., soybean powder, wheat flour and wood meal), mineral powders (e.g., clays such as kaolin, bentonite and acid clay, talcs such as talc powder and agalmatolite and other siliceous powders such as diatomaceous earth and mica powder) as well as alumina and active carbon. They may be used alone or as a mixture.

Surface active agents used as an emulsifying agent or a dispersing agent in the concentrate include non-ionic surface active agents such as polyoxyethylene fatty alcohol ethers, polyoxyethylene alkyl aryl ethers, polyoxyethylene fatty acid esters, polyoxyethylene sorbitan fatty acid esters, polyoxyethylene alkyl ether phosphate, polyoxyethylene fatty acid amines and amides, polyoxyethylene alkyl phosphate, polyoxyethylene alkyl aryl ether phosphate, polyoxyethylene alkyl aryl phosphate, glyceryl fatty acid monoesters, and glycol fatty acid esters; casein, gelatin, vegetable gums such as gum arabic and gum tragacanth, and carbohydrate dispersing agents such as dextrin. These agents also serve, when applied to plants, as a film-forming substance.

The present composition may be used in admixture with another germicide, insecticide, trace element, perfume or other additive.

The present composition may be applied by directly dip-treating plants in a dilute aqueous solution of the fungicide, spraying or spreading on the surfaces of plants, directly injecting into plants or spraying or spreading on soil for the purpose of sterilizing soil.

The rate of use of the fungicide of this invention, the amount of any other agents used therewith and the ratio thereof depend on various factors such as the kind of plant to be treated, its growth stage, growth state, cultivation method, kind of disease, state of appearance of disease, time of applying the agent, environmental conditions, method of

application and economical use of the agent.

The following Examples 1 to 5 illustrate the preparation of compositions according to the invention. Examples 7, 8 and 12 give the results of laboratory tests of antifungal activity. The other Examples show the antifungal use of compositions and methods of the present invention.

EXAMPLE 1.

550 Grams of isopropyl alcohol and 200 g of xylene were mixed with polyoxyethylene nonylphenyl ether (marketed under the trade mark "Nikkol—NP—10" by Nikko Chemical Co., Ltd.), in which 150 g of β -thujaplicin was dissolved with heating to produce an emulsifiable concentrate.

EXAMPLE 2.

50 Grams of γ -thujaplicin were dissolved with heating in 500 g of propylene glycol, with which 200 g of polyoxyethylene oleyl ether (marked under the trade mark "Nikkol—BO—7" by Nikko Chemical Co., Ltd.) and 250 g of water was mixed and dissolved to produce an emulsifiable concentrate.

EXAMPLE 3.

An aqueous solution was prepared which comprised 600 g of a 30% aqueous solution of gum arabic, 80 g of propylene glycol and 170 g of water. An oily solution was made by dissolving with heating 20 g of tropolone in 110 g turpentine oil and 20 g of natural resin. The aqueous solution was vigorously stirred while the oily solution was added dropwise therein and thoroughly mixed to form therein oily droplets. The resultant composition was an emulsifiable concentrate.

EXAMPLE 4.

400 Grams of methyl alcohol, 450 g of diacetone alcohol and 50 g of "Tween-80" (Atlas Co., Ltd., polyoxyethylene sorbitan mono-oleate) were mixed, in which 50 g of β -thujaplicin and 50 g of γ -thujaplicin were dissolved, to produce an emulsifiable concentrate.

EXAMPLE 5.

10 Grams of tropolone and 30 g of glyceryl monostearate were pulverized and mixed; they were further thoroughly mixed with 960 g of powdered talc, to produce uniform granules.

The following Examples illustrate the use of the composition of the invention.

EXAMPLE 6.

A Barancotto kind of pepper (*Piper nigrum* L.) was used as a test plant; saplings thereof were kept uninfected so as to avoid effects caused by other pathogenic fungi. Then, they were inoculated with *Fusarium fungi*. Comparison was made between plants injected

with thujaplicin and those untreated. At the time when the difference in growth was observed, both groups of the above plants were taken out of the soil, their roots were washed with water and the effect resulting from thujaplicin treatment was studied.

More specifically, from among 20 plants to be tested which were cultivated for layering sapling, 9 plants of about the same size were selected and transplanted in large pots. After confirming that they took root, *Fusarium fungus* which was separately cultured was soil-inoculated for all of the 9 plants. The inoculation was made in such a manner that the surrounding soil was dug out slopewise and 100 ml of a bran medium containing *Fusarium fungus* was poured into each of the 11 pots.

Thujaplicin treatment was effected as follows: ampoules were used containing about 1/10,000 solution of a 50:50 mixture of γ -thujaplicin and β -thujaplicin; the mouths of the ampoules were opened and then inserted upside down into the cut ends of 4 pepper branches (the diameter of the cut end of branch and that of the mouth of ampoule were approximately the same), and a synthetic resin adhesive was applied around the periphery of the ampoule mouth and cut end of branch so as to prevent leakage of the thujaplicin solution.

The thujaplicin treatment was effected about 1 week after the inoculation of *Fusarium fungus*. After the treatment, changes with time of untreated plants and treated plants were observed. One of 5 untreated plants shed leaves and died only one month after the inoculation. On the 133rd day after the inoculation with *Fusarium fungus*, the remaining 8 plants which survived were taken out and their rhizomes were washed with water.

On soil-inoculating with *Fusarium fungus*, the symptoms firstly appeared in a part of roots, which diseased roots turned to brown black. With thujaplicin-treated plants, diseased roots turned to black but new roots were very healthy and had many branched roots. With untreated plants, scarcely any new roots or branched roots were observed.

From the above facts, it is apparent that thujaplicin prevented the growth of *Fusarium fungus* in living plant bodies, which effect could be readily deduced from the states of growth of overground and underground parts. The growth of the overground part, of course, depends upon that of the underground part and thus 4 plants not treated with thujaplicin were just before death already presenting deciduous phenomenon, while 4 treated plants put forth new buds. Moreover, the effects of thujaplicin were sufficiently proved in that standard plants (not inoculated with *Fusarium fungus* and not treated with thujaplicin) grew healthily without falling of the leaves.

The microscopic examination was performed as follows. Sections were prepared by transversely cutting the stem at a point about 5 cm above the ground and subjected to the microscopic examination with the following results:

(1) Untreated infected plants.

In some of the vessels hyphae were observed (but not conidiospores) and cell walls turned to brown. In the ground part, hyphae spread in the vessels and further to the cortex part but did not spread to the external part through the epidermis. With vessels in which hyphae were not observed, it was often observed that their cell walls turned to brown and callus-like substances clogged them.

(2) Uninfected plants.

The stems remained healthy without turning to black and hyphae were not observed in vessels or epidermis. The roots were healthy but big roots had small swellings at several portions and nematodes were observed therein.

(3) Thujaplicin-treated plants.

In the stems, hyphae were observed as in untreated plants. Several roots had swellings

at several portions in which nematodes were observed. Only in these portions, hyphae were observed in the epidermis. One of two roots rotted but the others were healthy. The vessels were clogged with a brown callus-like substance as in untreated plants. Although this is closely similar to the cases of disease caused by *Fusarium*, it was confirmed that thujaplicin was an effective controlling agent, judging from the condition of vigour of plants; colour of leaves and new bud and new root germination.

EXAMPLE 7.

Antimicrobial Test of Various Compounds against *Fusarium fungus* (1).

Tropolone, β - and γ -thujaplicins were added to potato-glucose agar plates to 100, 75 and 50 ppm concentration. The plates were inoculated with one loopful of *Fusarium fungus* respectively and left in a constant temperature vessel kept at 37°C. For comparison, Benlate was tested in the same manner. The results are shown in the following Table 1.

TABLE 1

Compound	Concentration	Days in Culture Days				
		1	2	3	5	10
Tropolone	50 ppm	-	±	±	+	+
	75	-	-	±	±	±
	100	-	-	-	-	-
β -Thujaplicin	50	-	±	±	+	+
	75	-	-	±	±	±
	100	-	-	-	-	-
γ -Thujaplicin	50	-	±	±	+	+
	75	-	-	±	±	±
	100	-	-	-	-	-
Benlate	100	+	++	+++	+++	+++
	1,000	-	-	-	±	+
	2,000	-	-	-	-	±
Untreated Lot		+	++	+++	+++	+++

-: no growth of hypha, ±: growth difficult to judge, +: a little growth, ++: moderate growth, +++: significant growth.

EXAMPLE 8.

Antimicrobial Test of Various Compounds against *Fusarium fungus* (2).

- 5 Tropolone, β -thujaplicin, γ -thujaplicin, 3-hydroxy - 5 - methylisoxazol and Benlate were diluted with water. *Fusarium* hyphae were

immersed in these aqueous solutions for 15 to 20 min and inoculated on potato-glucose agar plates. The state of growth was observed and the results are shown in the following Table 2. 10

TABLE 2

Compound	Concentration	Time for Immersion			
		15	30	60	120 ^{min}
Tropolone	1/10,000	-	-	-	-
β -Thujaplicin	1/10,000	-	-	-	-
γ -Thujaplicin	1/10,000	-	-	-	-
Benlate	1/1,000	-	-	-	-
"	1/3,000	+	+	+	-
Hydroxy-Methyl-Isoxazol	1/1,500	+	+	+	+
"	1/3,000	+	+	+	+
Untreated	-	+	+	+	+

-: no growth of fungi, +: growth of fungi

EXAMPLE 9.

Controlling of Root-Rot of Pepper (1).

- 15 Six soil seed-beds for rearing pepper saplings were prepared, and in each bed was incorporated 0.1% or 0.02% of tropolone, β -thujaplicin or γ -thujaplicin. A seventh bed was left untested as a control. Pepper saplings were transplanted into each bed. At the time when roots and buds were being put forth from the saplings, the saplings were inoculated with *Fusarium fungus* and observed to see the progress of disease.

- 25 It was found that in the plants of the control untreated bed and in the 0.02% treated lot, the growth of *Fusarium hyphae* was remarkable and more than 80% of the plants were observed to be diseased. In the three 0.1% treated samples, there was scarcely observed any disease and hyphae did not grow and the growth of the saplings was best.

EXAMPLE 10.

35 Control of Root-Rot of Pepper (2).

Each test was effected on 5 young plants which were attacked with the root-rot of pepper.

- 40 (A) 6 ppm aqueous solution of β -thujaplicin
(B) mixed solution of 6 ppm of β -thujaplicin and 20 ppm of copper sulphate

(C) 100 ppm aqueous solution of Benlate.

Each one plant was sprayed on its leaves with 100 ml of one of the above aqueous solutions (A), (B) or (C). After 2 months, the state of growth of the plant was observed and the results were as follows:

(A) β -thujaplicin treated plants.

One plant died and the other 4 plants grew with 20% decrease in height.

(B) β -thujaplicin and copper sulphate treated plants.

All 5 plants did not die and grew with 20% decrease in height.

(C) benlate treated plants.

2 plants died and the other 3 plants grew with 50% decrease in height.

(D) untreated plants.

All 5 plants died.

EXAMPLE 11.

Controlling Downy Mildew of Hop.

The injection method was effected against young hop plants which apparently suffered from the downy mildew. One bine was selected from a hop plant having 5 to 6 bines and it was cut at 20 to 30 cm distance from a crown. To the cut part was attached a 100 ml ampoule which was charged with an aqueous solution of hinokitiol. The effect of

various concentrations of solution was tried on different bines.

Fifteen days after this treatment the plants were observed, with the following results:

	Height	β -Thujaplicin Concentration	Absorbed Amount of Solution	State of Disease
1	150 cm	1/10,000	10 ml	a little mildew on leaves remained
2	200	1/10,000	50	completely cured
3	160	1/7,500	20	completely cured
4	145	1/7,500	30	mildew on leaves in lateral branch

EXAMPLE 12.

Tropolone and α -, β - and γ -thujaplicins were added to glucose-peptone agar plates to 100 and 50 ppm concentration. The plates were inoculated with fungi to be tested and cultured at 27°C for 96 hours. They were ob-

served visually and estimated as follows:

+ : growth observed

- : not observed

$\pm$: difficult to judge

The results are shown in the following Table 3.

10

15

TABLE 3

	Concentration ppm	Fungi				
		Alternaria Kikuchianae	Piricularia Oryzae	Clado- sporium Fulvum	Fusarium Oxysporum	Venturia Inaequalis
Tropolone	100	-	-	-	-	-
	50	±	-	-	-	-
α -Thujaplicin	100	-	-	-	-	-
	50	±	-	-	-	-
β -Thujaplicin	100	-	-	-	-	-
	50	-	-	-	-	-
γ -Thujaplicin	100	-	-	-	-	-
	50	±	±	-	-	-

EXAMPLE 13.

Controlling Mouldy Rot of Gum Tree.

- 5 A test gum tree was incised for controlling a gum milk with a knife contaminated with *C. fimbriata*. A dilute aqueous solution of each of Difoltan, Benlate and γ -Thujaplicin was then applied to the infected cut portions with a brush for consecutive four weeks (3 times
- 10 a week) in each case, and the results were observed one month after the infection. The growth of each of the fungi was inhibited, and it was found that the bark began to show normal recovery. The concentration of each
- 15 of the agents used in this test was as follows:

Difoltan 0.5%
Benlate 0.1%
 γ -Thujaplicin 0.015%

EXAMPLE 14.

Controlling Orange Rust of Coffee Plants. 20

Thirty healthy young coffee plants which had been inoculated with *H. vastatrix* and observed to be infected were used as test trees.

These test trees were then treated by the leaf spray method with a dilute emulsion which had been adjusted to contain 100 ppm of an equivalent mixture of β - and γ -Thujaplicin. The amount of the liquid sprayed was 25

250 ml per the tree, and the observation of infections 10 days after infection showed that the disease progress was inhibited in each case.

- 5 In a test for determining the prophylactic effect, the above described 100 ppm dilute emulsion was uniformly sprayed in an amount of 250 ml per the tree. The tree thus treated
- 10 was then inoculated with *H. vastatrix*, and, after a lapse of 30 days, a normal incubation period, observation of the trees showed no indication of infections.

EXAMPLE 15.

Controlling Witches' Broom of Cacao Plant.

- 15 Twenty healthy young plants which had been inoculated with *M. perniciosus* and observed to be infected were used as test trees.

These test trees were then treated in the same manner as described in Example 14.

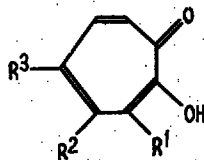
- 20 Observation of the infections showed that the disease progress was inhibited with no indication of infections in each case.

From the above tests, it is apparent that compositions of this invention have excellent fungicidal effects.

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WHAT WE CLAIM IS:—

1. A fungicidal composition suitable for use against diseases of vascular plants, which composition contains as active ingredient at
- 30 least one tropolone having the general formula:



- wherein R^1 , R^2 and R^3 are each a hydrogen atom or an alkyl radical having 1 to 3 carbon atoms, and is in the form of a liquid emulsifiable concentrate which contains 2 to 25% by weight of said tropolone, or in the form of solid granules which incorporate a solid carrier and which contain 0.1 to 5% by weight of said tropolone.
- 35

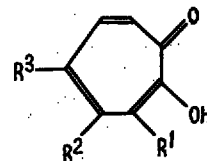
2. A composition according to Claim 1, in which said concentrate contains as emulsifying agent a non-ionic surface-active agent, vegetable gum or a carbohydrate dispersing agent.

3. A composition according to Claim 1, in which said solid carrier is a vegetable powder or mineral powder.

4. A composition according to Claim 1, substantially as hereinbefore described with reference to the product of any of Examples 1 to 5.
- 50

5. A method of controlling a fungus disease of a vascular plant, wherein a preparation containing at least one tropolone having the general formula:

55



- wherein R^1 , R^2 and R^3 are each a hydrogen atom or an alkyl radical having 1 to 3 carbon atoms is applied to the surface of the plant, injected into the plant or applied to the soil in which the plant is growing.
- 60

6. A method for controlling a fungus disease of a vascular plant, wherein a preparation made by dilution with water of an emulsifiable concentrate according to any of Claims 1, 2 or 4 is applied to the surface of the plant, injected into the plant or applied to the soil in which the plant is growing.
- 65

7. A method for controlling a fungus disease of a vascular plant wherein solid granules according to any one of Claims 1, 3 or 4 are applied to the surface of the plant, injected into the plant or applied to the soil in which the plant is growing.
- 70

8. A method for controlling diseases of plants according to any of Claims 5 to 7, in which the plant treated is black pepper and the disease of the plant is root-rot of pepper.
- 75

9. A method for controlling diseases of plants according to any of Claims 5 to 7, in which the plant treated is hop and the disease of the plant is downy mildew of hop.
- 80

10. A method for controlling diseases of plants according to any of Claims 5 to 7, wherein the plant treated is gum tree and the disease of the plant is mouldy rot.
- 85

11. A method for controlling diseases of plants according to any of Claims 5 to 7, wherein the plant treated is coffee plant and the disease of the plant is orange rust.
- 90

12. A method for controlling diseases of plant according to any of Claims 5 to 7, wherein the plant treated is cacao plant and the disease of the plant is witches' broom.

13. A method for controlling fungus diseases of vascular plants, according to Claim 5, which is carried out substantially as hereinbefore described with reference to the procedure of any of Examples 6, 9 to 11, and 13 to 15.
- 95

14. Plants treated by a method according to any of Claims 5 to 13.
- 100



250 ml per the tree, and the observation of infections 10 days after infection showed that the disease progress was inhibited in each case.

- 5 In a test for determining the prophylactic effect, the above described 100 ppm dilute emulsion was uniformly sprayed in an amount of 250 ml per the tree. The tree thus treated was then inoculated with *H. vastatrix*, and, after a lapse of 30 days, a normal incubation period, observation of the trees showed no indication of infections.

EXAMPLE 15.

Controlling Witches' Broom of Cacao Plant.

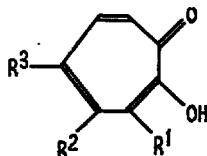
- 15 Twenty healthy young plants which had been inoculated with *M. pernicius* and observed to be infected were used as test trees.

- 20 These test trees were then treated in the same manner as described in Example 14. Observation of the infections showed that the disease progress was inhibited with no indication of infections in each case.

- 25 From the above tests, it is apparent that compositions of this invention have excellent fungicidal effects.

WHAT WE CLAIM IS:—

- 30 1. A fungicidal composition suitable for use against diseases of vascular plants, which composition contains as active ingredient at least one tropolone having the general formula:



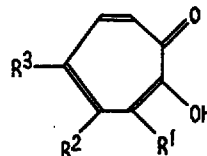
- 35 wherein R^1 , R^2 and R^3 are each a hydrogen atom or an alkyl radical having 1 to 3 carbon atoms, and is in the form of a liquid emulsifiable concentrate which contains 2 to 25% by weight of said tropolone, or in the form of solid granules which incorporate a solid carrier and which contain 0.1 to 5% by weight of said tropolone.

- 40 2. A composition according to Claim 1, in which said concentrate contains as emulsifying agent a non-ionic surface-active agent, vegetable gum or a carbohydrate dispersing agent.

- 45 3. A composition according to Claim 1, in which said solid carrier is a vegetable powder or mineral powder.

- 50 4. A composition according to Claim 1, substantially as hereinbefore described with reference to the product of any of Examples 1 to 5.

5. A method of controlling a fungus disease of a vascular plant, wherein a preparation containing at least one tropolone having the general formula:



- wherein R^1 , R^2 and R^3 are each a hydrogen atom or an alkyl radical having 1 to 3 carbon atoms is applied to the surface of the plant, injected into the plant or applied to the soil in which the plant is growing.

6. A method for controlling a fungus disease of a vascular plant, wherein a preparation made by dilution with water of an emulsifiable concentrate according to any of Claims 1, 2 or 4 is applied to the surface of the plant, injected into the plant or applied to the soil in which the plant is growing.

7. A method for controlling a fungus disease of a vascular plant wherein solid granules according to any one of Claims 1, 3 or 4 are applied to the surface of the plant, injected into the plant or applied to the soil in which the plant is growing.

8. A method for controlling diseases of plants according to any of Claims 5 to 7, in which the plant treated is black pepper and the disease of the plant is root-rot of pepper.

9. A method for controlling diseases of plants according to any of Claims 5 to 7, in which the plant treated is hop and the disease of the plant is downy mildew of hop.

10. A method for controlling diseases of plants according to any of Claims 5 to 7, wherein the plant treated is gum tree and the disease of the plant is mouldy rot.

11. A method for controlling diseases of plants according to any of Claims 5 to 7, wherein the plant treated is coffee plant and the disease of the plant is orange rust.

12. A method for controlling diseases of plant according to any of Claims 5 to 7, wherein the plant treated is cacao plant and the disease of the plant is witches' broom.

13. A method for controlling fungus diseases of vascular plants, according to Claim 5, which is carried out substantially as hereinbefore described with reference to the procedure of any of Examples 6, 9 to 11, and 13 to 15.

14. Plants treated by a method according to any of Claims 5 to 13.

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Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1976.
Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from
which copies may be obtained.

