

PATENT SPECIFICATION

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(54) METHODS OF DEFOLIATION OF PLANTS

(71) We, SCHERING AKTIENGESELLSCHAFT, a Body Corporate organised according to the laws of Germany, of Berlin and Bergkamen, Germany, 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:—

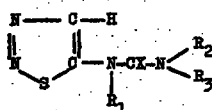
The present invention is concerned with methods of defoliating the plants defined below with 1,2,3-thiadiazolyl-ureas.

In the normal vital functions of a large number of types of plants a group of special cells, for example, at the base of the leaf stem, is changed into meristematic cells, owing to environmental conditions or certain processes within the plant. They form a separating layer such that finally the leaves fall off. However, this does not often occur at the time or to the extent that would be desirable, and, therefore, attempts have been made to control this process so as to take place at a predetermined time or stage of development of the plant.

Such controlled defoliation has special economic importance owing to the facilitation of harvesting and the acceleration of ripening obtainable thereby in agricultural and horticultural crops or also in the course of indirect measures to combat insects, fungi or bacteria that attack plants.

For this purpose there have already been proposed aliphatic thiosphosphates, especially tri-*n*-butyl trithiophosphate in United States Patent Specification No. 2,965,467. However, this active substance does not always have a satisfactory action and, furthermore, owing to its unpleasant odour troubles persons handling it.

It has now been found that compounds of the general formula I



(I)

in which

R₁ represents a hydrogen atom or an alkyl group containing 1 to 3 carbon atoms,

R₂ represents a hydrogen atom or an alkyl group which may be interrupted by at least one hetero atom selected from oxygen and sulphur atoms and

R₃ represents an alkyl group which may be interrupted by at least one hetero atom selected from oxygen and sulphur atoms, a cycloaliphatic hydrocarbon group which may be substituted by at least one alkyl group, or an aromatic hydrocarbon group which may be substituted by at least one substituent selected from alkyl groups, halogen atoms, alkyl-mercapto groups, alkoxy groups, trifluoromethyl groups and a nitro group, or

R₂ and R₃ together with the adjacent nitrogen atom represent a morpholino, piperidino or pyrrolidino group, and

X represents an oxygen or sulphur atom, are distinctly superior to the known tri-*n*-butyl trithiophosphate in their action and avoid the disadvantages thereof when used for the plants defined below.

The present invention accordingly provides a method of defoliating a living

plant selected from cotton plants, apple trees, hibiscus plants, grape vines and rose bushes, wherein the living plant is treated with a compound of the general formula I.

The present invention also provides a method of defoliating a growing cotton, apple, hibiscus, grape or rose crop, wherein the area of land containing the growing crop is treated with a compound of the general formula I.

As active substances of the general formula I given above there are especially suitable those compounds in which the symbol R_1 has the meaning given above, representing, for example, a methyl or ethyl group, the symbol R_2 represents a hydrogen atom or an alkyl group containing 1 to 4 carbon atoms, for example a methyl or ethyl group, the symbol R_3 represents an alkyl group containing 1 to 4 carbon atoms, for example a methyl or ethyl group, a cycloalkyl or alkyl-cycloalkyl group containing 5 to 8 carbon atoms, for example a cyclopentyl, cyclohexyl or methylcyclohexyl group, or a phenyl, halogeno-phenyl, methyl-phenyl or methoxy-phenyl group, and X has the meaning given above.

With regard to their defoliating action the compounds used in accordance with the present invention have a high activity and can be used, for example, for apple orchards, grapevines and rose bushes and especially in cotton. In this way plants or parts of plants to be harvested are in an advantageous manner both rendered more accessible and their ripening is considerably accelerated. In suitable environmental conditions plants so treated later form healthy normal foliage.

While the cultivated plants mentioned above shed leaves only under special physiological conditions or under special environmental conditions, *Malvaceae* react extremely sensitively and, after treatment with the compounds of the general formula I, shed their leaves with considerably less dependence on special conditions. The compounds of the general formula I may therefore be used for defoliating hibiscus plants.

The compounds used in accordance with the present invention are especially suitable for defoliating cotton plants, which enables gathering machines to be used for the capsule harvesting to an extent hitherto unobtainable.

The rates of application for the desired defoliation are within the range of from 1 to 10,000 grams of active substance per hectare, and preferably within the range of from 10 to 1000 grams of active substance per hectare. When two or more compounds of the general formula I are used these ranges refer of course to the total amount applied of these compounds.

The compounds used in accordance with the present invention may be applied either singly or in the form of mixtures of two or more such compounds; furthermore, the compounds of the general formula I may be applied in admixture with other active substances. If desired, other defoliating, plant protecting or pest combating agents may be added depending on the purpose desired.

The defoliating action and speed of defoliating action of the compounds of the general formula I can be promoted, for example, by one or more additives that are capable of increasing the action, for example solvents, especially organic solvents, preferably dimethyl sulphoxide, wetting agents and oils, preferably a non-phytotoxic paraffin oil. This enables the rate of application of the active substance proper to be reduced. Such additives may be used in a total amount of 1 to 300 parts by weight for each part by weight of the compound(s) of the general formula I.

Advantageously, the active substances of the general formula I or mixtures thereof with the other additives indicated above are used in the form of defoliating preparations, for example powders, strewable preparations, granules, solutions, emulsions or suspensions, with the addition of liquid and/or solid vehicles or diluents and, if desired, of surface-active agents, for example wetting agents, adherent agents, emulsifiers and/or dispersing agents.

Thus, the compounds of the general formula I may be used for the methods of the present invention in the form of a preparation comprising one or more compounds of the general formula I in admixture or conjunction with a suitable inert carrier and/or at least one additive capable of increasing the defoliating action of the compound(s) of the general formula I.

Suitable liquid carriers are, for example, water, aliphatic hydrocarbons, aromatic hydrocarbons, for example benzene, toluene and xylene, cyclohexanone, isophorone, dimethyl sulphoxide and dimethylformamide, and also mineral oil fractions.

As solid carriers there are suitable mineral earths, for example tonsil, silica gel,

talcum, kaolin, attaclay, limestone and silicic acid, and vegetable products, for example meals.

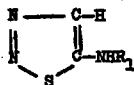
As surface-active agents there may be mentioned, for example, calcium lignin sulphonate, polyoxyethylene octylphenol ethers, naphthalene sulphonic acids and salts thereof, phenol sulphonic acids and salts thereof, formaldehyde condensates, fatty alcohol sulphates and also substituted benzene sulphonic acids and salts thereof.

The total amount of the active compound or compounds of the general formula I in the various preparations may vary within wide limits. For example, the preparations may contain approximately 10 to 80% by weight of active compound(s), approximately 90 to 20% by weight of liquid or solid carrier and, if desired, up to 20% by weight of surface-active agent(s).

The distribution of the compounds of the general formula I may be carried out in the usual manner, for example, with water as carrier in quantities of spray liquor from about 100 to 1000 litres per hectare. It is also possible to apply the compounds by the so-called "low volume method" and "ultra-low volume method" and they may also be applied in the form of so-called microgranules.

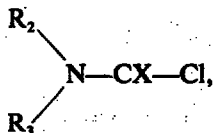
The compounds used in accordance with the present invention are known and may be prepared by known methods. They may be prepared, for example, by reacting the corresponding amino-compound with a carbamoyl halide, chloroformic acid ester or isocyanate, or with phosgene and then reacting the resulting isocyanate or carbamoyl halide with a corresponding amine to form the desired product. The thio-compounds are prepared with the use of the analogous sulphur-containing starting compounds.

Thus, the compounds of the great formula I may be prepared by reacting a compound of the general formula



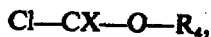
in which R_1 has the meaning given above,

- (a) in the presence of an acid-binding agent, advantageously an inorganic or a tertiary organic base, for example sodium carbonate or triethylamine, with a carbamoyl or thiocarbamoyl chloride of the general formula

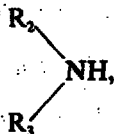


in which R_2 , R_3 and X have the meanings given above, or

- (b) in the presence of an acid-binding agent, for example triethylamine, with a chloroformic acid ester or chlorothioformic acid ester of the general formula



in which X has the meaning given above and R_4 represents a lower alkyl group, for example a methyl or ethyl group, or a phenyl group, and reacting the resulting compound with an amine of the general formula



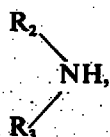
- (c) in the process of an acid-binding agent, for example N,N-dimethylaniline, with phosgene or thiophosgene and reacting the resulting isocyanate, isothiocyanate, carbamoyl chloride or thiocarbamoyl chloride with an amine of the general formula

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in which R_2 and R_3 have the meanings given above, or
(d) when R_2 represents a hydrogen atom, in the presence of a catalyst, advantageously an organic base, for example triethylamine, with an isocyanate or isothiocyanate of the general formula



in which R_3 and X have the meanings given above.
Examples 1 and 2 below illustrate the preparation of the active compounds used in accordance with the present invention.

The active compounds listed in the following Table can be prepared in a manner analogous to that described in Examples 1 and 2. (In the Table D denotes with decomposition).

TABLE I

Active compound	Physical constant
N-(4-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 256°C (D)
N-Cyclohexyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 215°C (D)
N-(3-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 244°C (D)
N-(4-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 228°C (D)
N-(3-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 208°C (D)
N-(3,4-Dichlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 236°C (D)
N-Methyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 174°C
N,N-Dimethyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 222°C (D)
N,N-Dimethyl-N'-methyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 129°C
N-Methyl-N'-methyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 221°C
N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-thiourea	M.p.: 205°C (D)
N-(4-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-thiourea	M.p.: 213°C (D)
N-Methyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 184°C (D)

TABLE I cont'd.

Active compound	Physical constant
N-Propyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 190°C (D)
N-Butyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 198°C (D)
N-(2-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 237°C (D)
N-(2-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 197°C (D)
N-(2-Nitrophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 229°C (D)
N-(3-Nitrophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 252°C (D)
N-Methyl-N-(2-methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	M.p.: 215°C (D)

These are crystalline substances which are very sparingly soluble, for example, in aliphatic and aromatic hydrocarbons and in water, and dissolve well in polar organic solvents, for example acetone, cyclohexanone, isophorone, dimethyl sulphoxide and dimethylformamide, depending on the particular substitution.

The starting compounds for the preparation of the compounds used in accordance with the present invention are known or can be prepared by methods known *per se*.

Some of the compounds used in accordance with the present invention have already been proposed as agents for regulating the growth of plants in general. In this case the agents exhibit an inhibiting effect which has the result of retarding vegetative growth (German Offenlegungsschrift No. 2,214,632).

The discovery of an action of these compounds influencing the leaf shedding of plants must therefore be especially surprising, because this action is directly opposed to the known retarding effect and is therefore all the more unexpected.

The following Examples 3 to 8 illustrate the invention. Examples 1 and 2 illustrate the preparation of the active compounds of the general formula I and Examples 9 and 10 illustrate preparations which may be used for the methods of the present invention.

Example 1.

N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.

10.1 grams (0.1 mole) of 5-amino-1,2,3-thiadiazole, 75 ml of tetrahydrofuran and 11.5 ml (0.106 mole) of phenyl isocyanate were mixed together. There was also added as catalyst 0.1 ml of triethylamine. After standing overnight the first crystals were precipitated. The solvent was removed by evaporation *in vacuo*, and the residue was recrystallized from isopropanol.

M.p.: 217°C (with decomposition)

Yield: 16.9 grams = 76.7% of the theoretical yield.

Example 2.

N-Ethyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.

11.05 Grams (0.05 mole) of 5-phenoxy-carbonylamino-1,2,3-thiadiazole, prepared in a known manner from 5-amino-1,2,3-thiadiazole and chloroformic acid phenyl ester, were dissolved in 100 ml of N,N-dimethylformamide, and 12.6 grams (0.1 mole) of N-ethylaniline were added. The mixture was maintained for 6 hours at 90 to 100°C while stirring. After cooling, the reaction mixture was diluted with

water, and the resulting urea precipitated in the form of a pale brown precipitate. The product was filtered off with suction, washed with benzene and dried. There were obtained 10.2 grams (82.1% of the theoretical yield) of the pure urea melting at 200°C.

M.p.: 200°C (with decomposition).

Yield: 10.2 grams = 82.1% of the theoretical yield.

Example 3.

In a series of tests potted young plants of cotton were treated at the stage of 5 to 6 unfolded true leaves. The quantities in which the compound used in accordance with the present invention and the agents used for comparison were applied in 500 litres of water per hectare are given in the Table below.

14 Days after the treatment the number of foliage leaves still present was determined. The results are expressed as the percentage defoliation as compared with the number of foliage leaves present before the treatment.

Active substance	kg of active substance per hectare	Percentage defoliation
N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	0.5	94.4
Agent for comparison		
Tri- <i>n</i> -butyl trithio-phosphate	0.7	38.9
Sodium chlorate	1	0
Untreated	0	0

Analogous effects were also obtained with the following active substances.

Active substances	
N-(4-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Cyclohexyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(3-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(4-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(3-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(3,4-Dichlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Methyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N,N-Dimethyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N,N-Dimethyl-N'-methyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Methyl-N'-methyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-thiourea.	
N-(4-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-thiourea.	
N-Methyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Ethyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Propyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Butyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(2-Chlorophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(2-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(2-Nitrophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-(3-Nitrophenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	
N-Methyl-N-(2-methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea.	

Example 4.

Potted cotton plants were treated as described in Example 3 at the stage of the beginning of the formation of flower buds.

The results are given in the following Table.

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	<u>Active substance</u>	<u>kg of active substance per hectare</u>	<u>Percentage defoliation</u>	
	N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	0.5	91.0	
5	N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-thiourea	0.5	90.4	5
	N-(2-Methylphenyl)-N'-(1,2,3-thiadiazol-5-yl)-urea	0.5	87.1	
10	N-Methyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	0.5	92.3	10
	<u>Agent for comparison</u>			
15	Tri-n-butyl trithio-phosphate	0.7	72.0	15
	Untreated	0	0	

Example 5.

20 Cotton plants were treated as described in Example 3 at the stage of flower bud formation.
 20 The aqueous suspensions contained in some cases as a further additive dimethyl sulphoxide in the concentrations given in the Table below. The results in the Table show an increase in action caused by the addition of dimethyl sulphoxide.

25		<u>kg/ha</u>		<u>Percentage defoliation</u>	25
		I	II		
	N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea (I)	0.05		81.8	
	Dimethyl sulphoxide (II)		15.0	0	
30	I + II	0.05	0.15	83.7	30
	I + II	0.05	15.0	95.6	

Example 6.

35 Cotton plants were treated as described in Example 3 at the stage of capsule ripeness. The aqueous suspensions contained as further additive a non-phytotoxic paraffin oil in some cases in combination with dimethyl sulphoxide in the concentrations given in the Table below.
 35 The results are apparent from the Table and show an increase in defoliation caused by the additives.

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	I	kg/ha II	III	Percentage defoliation	
5	N-Phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea (I)	0.05		45.4	5
	Paraffin oil (II)	5.0		0	
	Dimethyl sulphoxide (III)		5.0	0	
	I + II	0.05	0.5	61.8	
10	I + II	0.05	5.0	81.8	10
	I + II + III	0.05	2.5	94.1	

Example 7.

15 In an open field experiment apple trees (Cox Orange Rennet) were sprayed in separate tests 14 days after blossoming with an aqueous suspension of N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea of 0.03% and 0.09% strength, respectively. This caused a shedding of leaves on the young long shoots, but their further growth was, however, not affected. 15

Example 8.

20 Young plants of *Hibiscus rosa-sinensis* were sprayed in separate experiments with aqueous suspensions of N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea at rates of application corresponding to 0.005 kg/hectare, 0.05 kg/hectare and 0.5 kg/hectare. Irrespective of the actual dosages of active substance applied in this case the foliage was completely shed a few days after the treatment. 20

25 N-Butyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea produced a similar effect, whereas a tri-*n*-butyl trithiophosphate caused no defoliation either with 0.05 or 0.5 kg of active substance per hectare. 25

Example 9.

The preparation of a spray powder

30	50.0 per cent by weight of	N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea as active substance	30
	1.0 per cent by weight of	wetting agent based on sodium alkyl-naphthalene sulphonate	
	20.0 per cent by weight of	surface-active substance based on calcium lignin sulphonate	
35	29.0 per cent by weight of	aluminium silicate as carrier material	35

The constituents were ground to a finely dispersed powder in a rod mill, and gave a preparation ready for use; such a preparation may be diluted with water to form the desired suspensions.

Example 10.

40 The preparation of a solution of active substance

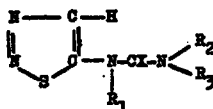
	20.0 per cent by weight of	N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea	
	40.0 per cent by weight of	dimethyl sulphoxide	
45	40.0 per cent by weight of	xylene	45

The active substance was mixed with the other constituents while stirring and gave a solution ready for use; such a solution can be applied by the so-called ultra-low volume method, for example, with the use of aircraft.

After admixture with surface-active agents, for example emulsifiers, there are formed so-called emulsion concentrates, which may be used after dilution with water.

WHAT WE CLAIM IS:—

1. A method of defoliating a living plant selected from cotton plants, apple trees, hibiscus plants, grape vines and rose bushes, wherein the living plant is treated with a compound of the general formula I



in which

R₁ represents a hydrogen atom or containing 1 to 3 carbon atoms,

R₂ represents a hydrogen atom or an alkyl group which may be interrupted by at least one hetero atom selected from oxygen and sulphur atoms and

R₃ represents an alkyl group which may be interrupted by at least one hetero atom selected from oxygen and sulphur atoms, a cycloaliphatic hydrocarbon group which may be substituted by at least one alkyl group, or an aromatic hydrocarbon group which may be substituted by at least one substituent selected from alkyl groups, halogen atoms, alkyl-mercapto groups, alkoxy groups, trifluoromethyl groups and a nitro group, or

R₂ and R₃ together with the adjacent nitrogen atom represent a morpholino, piperidino or pyrrolidino group, and

X represents an oxygen or sulphur atom,

2. A method as claimed in claim 1, wherein R₁ represents a methyl or ethyl group.

3. A method as claimed in claim 1 or 2, wherein R₂ represents an alkyl group containing 1 to 4 carbon atoms.

4. A method as claimed in claim 3, wherein R₃ represents a methyl or ethyl group.

5. A method as claimed in any one of claims 1 to 4, wherein R₃ represents an alkyl group containing 1 to 4 carbon atoms.

6. A method as claimed in claim 5, wherein R₃ represents a methyl or ethyl group.

7. A method as claimed in any one of claims 1 to 4, wherein R₃ represents a cycloalkyl or alkyl-cycloalkyl group containing 5 to 8 carbon atoms.

8. A method as claimed in claim 7, wherein R₃ represents a cyclopentyl, cyclohexyl or methylcyclohexyl group.

9. A method as claimed in any one of claims 1 to 4, wherein R₃ represents a phenyl, halogeno-phenyl, methyl-phenyl or methoxy-phenyl group.

10. A method as claimed in any one of claims 1 to 9, wherein the plant is treated with N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.

11. A method as claimed in any one of claims 1 to 9, wherein the plant is treated with at least one compound selected from N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea, N-ethyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea and the compounds listed in Table I herein.

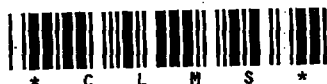
12. A method as claimed in any one of claims 1 to 11, wherein a single compound of the general formula I is used for the treatment in an amount within the range of from 1 to 10,000 grams per hectare.

13. A method as claimed in claim 12, wherein the amount is within the range of from 10 to 1000 grams per hectare.

14. A method as claimed in any one of claims 1 to 11, wherein two or more compounds of the general formula I are used for the treatment in a total amount within the range of from 1 to 10,000 grams per hectare.

15. A method as claimed in claim 14, wherein the total amount is within the range of from 10 to 1000 grams per hectare.

16. A method of defoliating a growing cotton, apple, hibiscus, grape or rose crop, wherein the area of land containing the growing crop is treated with a



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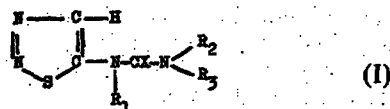
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The active substance was mixed with the other constituents while stirring and gave a solution ready for use; such a solution can be applied by the so-called ultra-low volume method, for example, with the use of aircraft.

After admixture with surface-active agents, for example emulsifiers, there are formed so-called emulsion concentrates, which may be used after dilution with water.

WHAT WE CLAIM IS:—

1. A method of defoliating a living plant selected from cotton plants, apple trees, hibiscus plants, grape vines and rose bushes, wherein the living plant is treated with a compound of the general formula I



in which

R₁ represents a hydrogen atom or containing 1 to 3 carbon atoms,

R₂ represents a hydrogen atom or an alkyl group which may be interrupted by at least one hetero atom selected from oxygen and sulphur atoms and

R₃ represents an alkyl group which may be interrupted by at least one hetero atom selected from oxygen and sulphur atoms, a cycloaliphatic hydrocarbon group which may be substituted by at least one alkyl group, or an aromatic hydrocarbon group which may be substituted by at least one substituent selected from alkyl groups, halogen atoms, alkyl-mercapto groups, alkoxy groups, trifluoromethyl groups and a nitro group, or

R₂ and R₃ together with the adjacent nitrogen atom represent a morpholino, piperidino or pyrrolidino group, and

X represents an oxygen or sulphur atom,

2. A method as claimed in claim 1, wherein R₁ represents a methyl or ethyl group.

3. A method as claimed in claim 1 or 2, wherein R₂ represents an alkyl group containing 1 to 4 carbon atoms.

4. A method as claimed in claim 3, wherein R₂ represents a methyl or ethyl group.

5. A method as claimed in any one of claims 1 to 4, wherein R₃ represents an alkyl group containing 1 to 4 carbon atoms.

6. A method as claimed in claim 5, wherein R₃ represents a methyl or ethyl group.

7. A method as claimed in any one of claims 1 to 4, wherein R₃ represents a cycloalkyl or alkyl-cycloalkyl group containing 5 to 8 carbon atoms.

8. A method as claimed in claim 7, wherein R₃ represents a cyclopentyl, cyclohexyl or methylcyclohexyl group.

9. A method as claimed in any one of claims 1 to 4, wherein R₃ represents a phenyl, halogeno-phenyl, methyl-phenyl or methoxy-phenyl group.

10. A method as claimed in any one of claims 1 to 9, wherein the plant is treated with N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.

11. A method as claimed in any one of claims 1 to 9, wherein the plant is treated with at least one compound selected from N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea, N-ethyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea and the compounds listed in Table I herein.

12. A method as claimed in any one of claims 1 to 11, wherein a single compound of the general formula I is used for the treatment in an amount within the range of from 1 to 10,000 grams per hectare.

13. A method as claimed in claim 12, wherein the amount is within the range of from 10 to 1000 grams per hectare.

14. A method as claimed in any one of claims 1 to 11, wherein two or more compounds of the general formula I are used for the treatment in a total amount within the range of from 1 to 10,000 grams per hectare.

15. A method as claimed in claim 14, wherein the total amount is within the range of from 10 to 1000 grams per hectare.

16. A method of defoliating a growing cotton, apple, hibiscus, grape or rose crop, wherein the area of land containing the growing crop is treated with a



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compound of the general formula I given in claim 1, in which R_1 , R_2 , R_3 , and X have the meanings given in claim 1.

17. A method as claimed in claim 16, wherein R_1 represents a methyl or ethyl group.

18. A method as claimed in claim 16 or 17, wherein R_2 represents an alkyl group containing 1 to 4 carbon atoms.

19. A method as claimed in claim 18, wherein R_2 represents a methyl or ethyl group.

20. A method as claimed in any one of claims 16 to 19, wherein R_3 represents an alkyl group containing 1 to 4 carbon atoms.

21. A method as claimed in claim 20, wherein R_3 represents a methyl or ethyl group.

22. A method as claimed in any one of claims 16 to 19, wherein R_3 represents a cycloalkyl or alkyl-cycloalkyl group containing 5 to 8 carbon atoms.

23. A method as claimed in claim 22, wherein R_3 represents a cyclopentyl, cyclohexyl or methylcyclohexyl group.

24. A method as claimed in any one of claims 16 to 19, wherein R_3 represents a phenyl, halogeno-phenyl, methyl-phenyl or methoxy-phenyl group.

25. A method as claimed in any one of claims 16 to 24, wherein the area of land is treated with N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea.

26. A method as claimed in any one of claims 16 to 24 wherein the area of land is treated with at least one compound selected from N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea, N-ethyl-N-phenyl-N'-(1,2,3-thiadiazol-5-yl)-urea and the compounds listed in Table I herein.

27. A method as claimed in any one of claims 16 to 26, wherein a single compound of the general formula I is used for the treatment in an amount within the range of from 1 to 10,000 grams per hectare.

28. A method as claimed in claim 27, wherein the amount is within the range of from 10 to 10,000 grams per hectare.

29. A method as claimed in any one of claims 16 to 26, wherein two or more compounds of the general formula I are used for the treatment in a total amount within the range of from 1 to 10,000 grams per hectare.

30. A method as claimed in claim 29, wherein the total amount is within the range of from 10 to 1000 grams per hectare.

31. A method as claimed in any one of claims 1 to 30, wherein the compound(s) of the general formula I is/are used for the treatment in the form of a preparation comprising the compound(s) of the general formula I in admixture or conjunction with a suitable inert carrier and/or at least one additive capable of increasing the defoliating action of the compound(s) of the general formula I.

32. A method as claimed in claim 31, wherein the additive(s) is/are a solvent capable of increasing the defoliating action and/or an oil.

33. A method as claimed in claim 32, wherein the solvent is dimethyl sulphoxide.

34. A method as claimed in claim 32 or 33, wherein the oil is a non-phytotoxic paraffin oil.

35. A method as claimed in any one of claims 31 to 34, wherein the preparation contains a single additive capable of increasing the defoliating action in an amount of 1 to 300 parts by weight for each part by weight of the compound(s) of the general formula I.

36. A method as claimed in any one of claims 31 to 34, wherein the preparation contains two or more additives capable of increasing the defoliating action in a total amount of 1 to 300 parts by weight for each part by weight of the compound(s) of the general formula I.

37. A method as claimed in any one of claims 31 to 36, wherein the preparation is in the form of a powder, a strewable preparation, granules, a solution, an emulsion or a suspension.

38. A method as claimed in any one of claims 31 to 37, wherein the preparation contains a single compound of the general formula I in an amount of approximately 10 to 80% by weight.

39. A method as claimed in any one of claims 31 to 37, wherein the preparation contains two or more compounds of the general formula I in a total amount of approximately 10 to 80% by weight.

40. A method as claimed in any one of claims 31 to 39, wherein the preparation contains a single surface-active agent in an amount of up to 20% by weight.

41. A method as claimed in any one of claims 31 to 39, wherein the preparation



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11 contains two or more surface-active agents in a total amount of up to 20% by weight.

42. A method as claimed in claim 31, wherein the preparation has a composition substantially as described in Example 9 or 10 herein.

5 43. A method as claimed in claim 1, conducted substantially as described in any one of Examples 3, 4 and 8 herein.

44. A method as claimed in claim 1, conducted substantially as described in Example 5 or 6 herein.

10 45. A method as claimed in claim 16, conducted substantially as described in Example 7 herein.

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