

PATENT SPECIFICATION

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SCIENCE REFERENCE LIBRARY

(54) HERBICIDAL COMPOSITIONS

(71) We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, Imperial Chemical House, Millbank, London, SW1P 3JF, a British Company, 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 herbicidal compositions containing a herbicidal bipyridylum quaternary salt as an active ingredient.

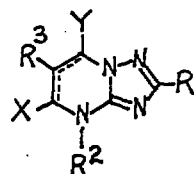
Over the years, a wide range of pesticides has been developed for agricultural use in the control of fungal and insect pests and weeds. While these substances are necessarily toxic to certain forms of life, when used with due care and in accordance with governmentally approved codes of practice, they present no hazard to human life. However, in spite of efforts to encourage those concerned with pesticides to adopt safe handling practices instances of misuse of pesticides do occur. One particular unsafe practice in the case of liquid pesticides is for an operator to transfer a small amount of the concentrated pesticide to a domestic container such as a beverage bottle for subsequent use at home. The risk attached to this practice is of course that a child or incautious adult coming upon the bottle may swallow the contents with possibly serious consequences.

We have now found it possible to reduce the likelihood of serious consequences of such accidental swallowing by induction of emesis. This can result in some cases in the rapid removal of the pesticidal composition from the stomach and digestive tract before lethal amounts of the pesticide can be assimilated by the body.

We have further discovered that the admixture of a known triazolo[1,5-a]pyrimidine derivative of formula (I), as specified hereinafter, to a herbicidal bipyridylum quaternary salt in a herbicidal composition produces a composition which if swallowed tends to in-

duce emesis, and thereby, expulsion of the composition.

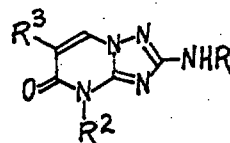
According to the invention there is provided a herbicidal composition of reduced health hazard comprising a salt of a herbicidal bipyridylum quaternary cation and a s - triazolo - [1,5 - a]pyrimidine derivative of the formula:



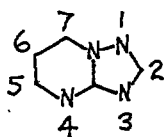
(I) 55

wherein R¹ is an amino, alkylamino, phenyl-alkylamino, dialkylamino, ureido, carboxyl, hydroxyalkyl or carbazoyl group; R² is an alkyl, cycloalkyl or alkenyl group; R³ is hydrogen, halogen or an alkyl or hydroxyalkyl group; one of X and Y stands for an-oxo or thioxo radical, and the other of X and Y stands for hydrogen or an alkyl radical and when X stands for an oxo or thioxo radical, the nucleus contains a double bond between the carbon atoms in positions 6 and 7, and when Y stands for an oxo or thioxo radical, the nucleus contains a double bond between the carbon atoms in positions 5 and 6; or a base addition salt of a triazolo - pyrimidine derivative as defined above which contains an acidic group; or an acylated derivative, or base addition salt thereof.

Preferably the invention provides a herbicidal composition of reduced health hazard comprising a salt of a herbicidal bipyridylum quaternary cation and a s - triazolo - [1,5-a]pyrimidine derivative of the formula:



wherein R^1 is hydrogen, a C_{1-4} alkyl radical, or a $COOEt$ radical, R^2 is a C_{1-4} alkyl radical or an allyl radical, and R^3 is a C_{1-4} alkyl radical; and addition salts thereof. The s-triazolo[1,5-a]pyrimidine ring structure is numbered as shown below:

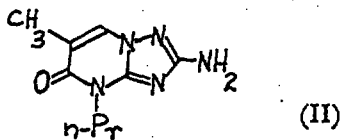


There are some indications that the reduced toxicity of the compositions of the invention may not be due solely to their emetic properties. It seems possible (though this is yet unproven) that in certain circumstances the triazolopyrimidine may delay uptake by the blood of the toxic chemical from the stomach and digestive tract even if emesis does not take place.

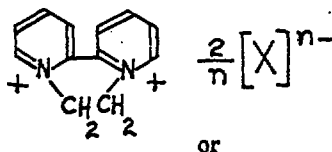
Particular derivatives of 5-oxo-4,5-dihydro-s-triazolo[1,5-a]pyrimidine of use in the practice of the invention are:

- 6-methyl-4-n-propyl-2-n-propylamino-
- 2-amino-6-methyl-4-allyl-
- 2-amino-4,6-di-n-propyl-
- 2-ethoxycarbonylamino-6-methyl-4-n-propyl-

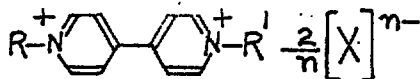
An especially useful triazolo-pyrimidine for use in the compositions of the invention is 2-amino-6-methyl-5-oxo-4-n-propyl-4,5-dihydro-s-triazolo[1,5-a]pyrimidine (II).



Preferred herbicidal bipyridylium quaternary salts for use in the compositions of the invention are those of the following formulae:



or



wherein R and R^1 , which may be the same or different, stand for alkyl radicals of from 1 to 4 carbon atoms which may be substituted by hydroxyl, halogen, carboxyl, alkoxy,

alkylcarbonyl, alkoxy carbonyl, carbamoyl or N-substituted carbamoyl; $[X]^{n-}$ represents an anion and n is an integer from 1 to 4 inclusive.

Particularly preferred herbicidal bipyridylium quaternary salts are those listed below:

- 1,1'-dimethyl-4,4'-bipyridylium dimethylsulphate (paraquat dimethylsulphate)
- 1,1'-ethylene-2,2'-bipyridylium dibromide (diquat dibromide)
- 1,1'-dimethyl-4,4'-bipyridylium dichloride (paraquat dichloride)
- 1,1'-di-2-hydroxyethyl-4,4'-bipyridylium dichloride
- 1-(2-hydroxyethyl)-1'-methyl-4,4'-bipyridylium dichloride
- 1,1'-di-carbamoylmethyl-4,4'-bipyridylium dichloride
- 1,1'-bis-N,N-dimethylcarbamoylmethyl-4,4'-bipyridylium dichloride
- 1,1'-dimethyl-4,4'-bipyridylium sulphate (paraquat sulphate)
- 1,1'-bis-N,N-diethylcarbamoylmethyl-4,4'-bipyridylium dichloride
- 1,1'-diacetonyl-4,4'-bipyridylium dichloride
- 1,1'-diethoxycarbonylmethyl-4,4'-bipyridylium dibromide

The names in brackets alongside some of the compounds in the above list are the accepted common names for these compounds.

Thus 'paraquat' is the common name for the 1,1'-dimethyl-4,4'-bipyridylium cation. Paraquat is a particularly preferred bipyridylium cation for use in the compositions of the invention. A particularly preferred anion $[X]^{n-}$ is the chloride anion, for reasons of convenience and economy, but any anion which gives rise to a conveniently water-soluble salt may be used if desired. The herbicidal action is due solely to the cation and for this reason the concentration of a herbicidal bipyridylium salt composition is frequently given in terms of the cation alone. The amount of herbicidal bipyridylium quaternary salt present in the compositions of the invention is generally from 1.0 to 99.9% by weight.

The compositions of the invention may comprise a carrier or diluent, and may be solids, e.g. granules, or liquids, e.g. aqueous solutions.

In a preferred aspect the invention provides a concentrated herbicidal composition comprising an aqueous solution of a salt of a herbicidal bipyridylium quaternary cation and a triazolo-pyrimidine as hereinbefore defined. Preferably the herbicidal bipyridylium quaternary cation is paraquat.

The amount of herbicidal bipyridylium quaternary cation present in the aqueous solu-

tion is preferably from 0.5 to 4.0 pounds per Imperial gallon (50 grams to 400 grams per litre).

Preferably the composition also comprises a surface active agent.

Surface-active agents may be cationic, non-ionic or anionic. Generally speaking cationic and non-ionic surface-active agents are preferred to anionic surface-active agents for use in the compositions of the invention, since the latter may interact undesirably with the bipyridylum quaternary salt in the compositions. Examples of non-ionic surface-active agents for use in the compositions of the invention include the condensation products of ethylene oxide with alkyl phenols such as octylphenol, nonylphenol and octylcresol. Other non-ionic agents are the partial esters derived from long chain fatty acids and hexitol anhydrides, for example sorbitan monolaurate; the condensation products of the said partial esters with ethylene oxide; and the lecithins. Examples of cationic surface-active agents include quaternary salts and condensates of ethylene oxide with amines, for example the substances sold under the Trade Mark "Ethomeen", "Ethoduomeen", "Duoquad" and "Arquad".

Particularly preferred surface-active agents are the combinations of surface-active agents described in U.K. Patent No. 998,264 for use in formulations of herbicidal bipyridylum quaternary salts.

The preparation of the triazolo - pyrimidines (I) has been described in U.K. Patent Specification No. 1,234,635.

The emetic properties of the composition are primarily determined by the amount of triazolo - pyrimidine it contains. In deciding the most appropriate amounts of triazolo - pyrimidine to use in any composition, regard must be had to the efficacy of the triazolo - pyrimidine relative to the toxicity of the herbicide. The amount of triazolo - pyrimidine to be included is preferably such that any sample of the composition containing a potentially lethal dose of herbicide will contain sufficient of the triazolo - pyrimidine to give it an emetic action. However, clearly neither lethal doses nor emetic doses can be directly measured in man; they can only be inferred from animal data.

Compositions according to the invention preferably contain from 0.1 to 5 parts by weight of the triazolo - pyrimidine (e.g. the triazolopyrimidine II) per 100 parts of the herbicidal bipyridylum quaternary cation (e.g. paraquat). Conveniently the amount of the triazolo - pyrimidine (e.g. the triazolopyrimidine II) used is 0.25 to 2.0 parts per 100 parts of herbicidal bipyridylum quaternary cation (e.g. paraquat).

Concentrated aqueous formulations of the invention are corrosive. They must be handled with care, to avoid splashing of the eyes or

skin, and they should not be allowed to come into contact with corrodeable metals prior to dilution.

The compositions according to the invention may also comprise coloured dyestuff or pigment compounds to give them a characteristic or distinguishing colour. Examples of such compounds are "Monastral Blue BNV Paste" and "Lissamine Turquoise VN 150".

The compositions according to the invention may also comprise a stenching agent. Examples of such stenching agents are: alkyl pyridines as described in U.K. Patent Specification No. 1,406,881; *n* - valeric acid; and tetrahydrothiophen.

If desired, the triazolo - pyrimidines (I) may be incorporated in thixotropic formulations of herbicidal quaternary salts. In particular the triazolo - pyrimidines (I) may be incorporated in the formulations of herbicidal bipyridylum quaternary salts described in our U.K. Patent Specification No. 1,395,502. These formulations comprise an aqueous solution of a herbicidal bipyridylum quaternary salt containing a gelling agent, for example finely divided silica, or a combination of the xanthan gum sold under the trade mark "Kelzan" with sodium metaborate. Xanthan gum is a complex polysaccharide.

The compositions according to the invention may also comprise a herbicide other than a herbicidal bipyridylum quaternary salt as hereinbefore defined. Examples of such herbicides are:

Amides (e.g. *N,N* - diallylchloroacetamide, 3,4 - dichloropropionanilide, *N* - (3 - chloro - 4 - methyl - phenyl) - 2 - methylpentamide).

Carbamates (e.g. Isopropyl - *N* - phenylcarbamate, isopropyl - *N* - (3 - chlorophenyl) carbamate, 4 - chloro - 2 but - 2 - ynyl - *N* - (3 - chlorophenyl) carbamate, 2 - chlorallyl - *N,N* - diethyl - dithiocarbamate).

Ureas/anilides (e.g. *N,N'* - di - (2,2,2-trichloro - 1 - hydroxy - ethyl)urea, 3,4-dichloroacetanilide, *O* - chloroisobutyranilide, α - bromo - 3,4 - dichloroacetanilide, 3,4-dichloroformanilide, 2 - acetamido - 3 - chlorotoluene).

Diazines (e.g. 3,4,5,6 - tetrahydro - 3,5-dimethyl - 2 - thio - 2H - 1,3,5 - thiadiazine, 5 - bromo - 3 - isopropyl - 6 - methyluracil, 5 - amino - 4 - chloro - 2 - phenyl - 3-pyridazone, 1,2,3,6 - tetrahydro - 3,6 - dioxypyridazine).

Triazines (e.g. 2 - chloro - 4,6 - bisethylamino - 1,3,5 - triazine, 2 - chloro - 4 - ethylamino - 6 - isopropylamino - 1,3,5 - triazine, 2 - chloro - 4,6 - bisisopropylamino - 1,3,5-triazine, 4 - ethylamino - 6 - isopropylamino - 2 - methoxy - 1,3,5 - triazine, 4,6 - bisisopropylamino - 2 - methoxy - 1,3,5 - triazine).

The inclusion of the triazolo - pyrimidine (I) in the compositions according to the invention has no significant adverse effect upon the herbicidal activity of the compositions.

The invention is illustrated by the following Examples.

EXAMPLE 1

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - *n*-propyl - 4,5 - dihydro - s - triazolo[1,5 - a]-pyrimidine (II) in aqueous solution.

Ingredients	% w/v
Paraquat concentrate (II)	x 0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

Paraquat concentrate is a solution of paraquat dichloride containing 25% to 30% by weight of 1,1' - dimethyl - 4,4' - bipyridylium cation. The amount used was chosen to give a composition containing 20% weight/volume paraquat cation. The composition was prepared by a simple agitation of the ingredients together.

EXAMPLE 2

This Example illustrates a composition according to the invention which comprises the triazolo - pyrimidine (II) in aqueous solution. The composition comprises the following ingredients.

Ingredients	% w/v
Paraquat concentrate	x
Sodium metaborate	1.3
Sodium benzoate	2.0
Lissapol NX	1.1
DS 4392	4.1
Silcolapse 5000 (II)	0.06 0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

"Lissapol" NX is a Trade Mark for a surface-active agent comprising a condensate of from 7 to 8 molar proportions of ethylene oxide with 1 molar proportion of *p* - nonylphenol.

DS 4392 is a code number for a surface-active agent comprising a mixture of amines derived from soya bean fatty acids condensed with approximately 15 molar proportions of ethylene oxide.

"Silcolapse" is a Trade Mark for an anti-foaming agent comprising a silicone derivative.

The composition described above was prepared by simple agitation of the ingredients together.

EXAMPLE 3

This Example illustrates a composition according to the invention which comprises a gelling agent in addition to the triazolo-pyrimidine (II). The composition comprises the following ingredients.

Ingredients	Amount in Grams
Paraquat concentrate "Kelzan"	x 0.4 (added as 1% solution in water)
Sodium metaborate	0.014
Lissapol NX	1.1
DS 4392 (II)	4.1 0.05
Silcolapse 5000	0.01
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat ion)

The composition described above was prepared by simple agitation of the ingredients together and was sufficiently fluid when prepared to be poured into containers. On standing for 15 to 20 minutes the composition formed a gel and was not pourable unless vigorously shaken.

EXAMPLE 4

This Example illustrates a dry free flowing granular composition which is both stable in storage and is readily convertible into an aqueous solution for application as a spray.

A solution was made up having the following composition in which percentages are by weight.

Paraquat dichloride	33.0	90
2-amino-6-methyl-5-oxo-4- <i>n</i> -propyl-4,5-dihydro-s-triazolo[1,5-a]pyrimidine (II)	0.1	
Sodium metaborate	2.5	95
Potassium phosphate	1.6	
Lissapol NX	24.0	
Water	38.8	
	100	

310 gms of the above solution were then added in a thin stream to 690 gms of dried magnesium sulphate contained in the bowl of a Hobart C.E. 100 dough mixer ("Hobart" is a Trade Mark). The resulting product which was dry was then passed through a granulating machine and finally agitated in a sieve having 30 meshes per linear inch to remove dust. The resulting granules had a size of at least 0.9 mm and possessed a solution rate of 150 seconds.

EXAMPLE 5

This Example illustrates a composition

according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - *n*-propyl - 4,5 - dihydro - *s* - triazolo[1,5-*a*]-pyrimidine (II) and a stenching agent in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
<i>n</i> -valeric acid	1.0
II	0.05
DS 4392	4.0
Silcolapse	0.01
Lissapol NX	1.0
Water	to 100 mls

(where x gives 200 g/l paraquat ion).

The composition described above was prepared by simple agitation of the ingredients together.

EXAMPLE 6

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - *n*-propyl - 4,5 - dihydro - *s* - triazolo[1,5-*a*]-pyrimidine (II) in a thickened, coloured, aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
Synperonic 2	2.5
Synperonic 16	2.5
Nansa 1106	8.5
Monastral BNVS Paste	1.0
Pyridine Base	1.0
II	0.05
Water	to 100 mls

(where x gives 200 g/l paraquat ion).

"Synperonic" and "Nansa" are Trade Marks. Synperonic 2 is a condensation product of a mixture of 67% C₁₃ and 33% C₁₅ aliphatic alcohols with two equivalents of ethylene oxide. Synperonic 16 is a condensation product of a mixture of 67% C₁₃ and 33% C₁₅ aliphatic alcohols with sixteen equivalents of ethylene oxide. Nansa 1106 is sodium dodecyl (substantially C₁₂ straight chain) benzene sulphonate. Monastral BNVS paste is a dispersion comprising 15% w/v copper phthalocyanine pigment in water. Pyridine base is a mixture consisting substantially of alkyl pyridines.

EXAMPLE 7

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - *n*-propyl - 4,5 - dihydro - *s* - triazolo[1,5-*a*]-pyrimidine (II) in aqueous solution.

Ingredients	% w/v
diquat dibromide	x
sodium molybdate	0.18
potassium phosphate (as a mixture of dipotassium hydrogen phosphate and potassium dihydrogen phosphate)	2.75
(II)	0.05
water	to 100 ml

(where x gives 140 ± 5 g/l diquat ion).

Diquat ion is 1,1' - ethylene - 2,2' - bipyridylum cation. The composition described above was prepared by simple agitation of the ingredients together, and had a pH of 6.5 ± 0.3.

EXAMPLE 8

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - *n*-propyl - 4,5 - dihydro - *s* - triazolo[1,5-*a*]-pyrimidine (II) in aqueous solution.

Ingredients	% w/v
Morfamquat dichloride	x
II	0.05
water	to 100 ml

(where x gives 300 ± 5 g/l morfamquat ion).

Morfamquat ion is 1,1' - bis(3,5 - dimethylmorpholinocarbonylmethyl - 4,4' - bipyridylum ion. The composition described above was prepared by simple agitation of the ingredients together.

EXAMPLE 9

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - *n*-propyl - 4,5 - dihydro - *s* - triazolo[1,5-*a*]-pyrimidine (II) in aqueous solution.

Ingredients	% w/v
1,1'-bis(diethylcarbamoylmethyl-4,4'-bipyridylum dichloride	x
Tween 20	8
(II)	0.05
water	to 100 ml

(where x gives 200 ± 5 g/l 1,1' - bis(diethylcarbamoylmethyl - 4,4' - bipyridylum ion). The composition described above was prepared by simple agitation of the ingredients together.

"Tween" is a Trade Mark. Tween 20 is a condensation product of one mole of sorbitan monolaurate with twenty moles of ethylene oxide.

EXAMPLE 10

This Example illustrates a composition according to the invention which comprises 2 - *n* - propylamino - 6 - methyl - 5 - oxo - 4 - *n* - propyl - 4,5 - dihydro - s - triazolo[1,5 - a] pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
2- <i>n</i> -propylamino-6-methyl-5-oxo-4- <i>n</i> -propyl-4,5-dihydro-s-triazolo[1,5-a]-pyrimidine	0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

EXAMPLE 11

This Example illustrates a composition according to the invention which comprises 2 - amino - 5 - oxo - 4,6 - di - *n* - propyl - 4,5 - dihydro - s - triazolo[1,5 - a]pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
2-amino-5-oxo-4,6-di- <i>n</i> -propyl-4,5-dihydro-s-triazolo[1,5-a]pyrimidine	0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

EXAMPLE 12

This Example illustrates a composition according to the invention which comprises 2 - ethoxycarbonylamino - 6 - methyl - 5 - oxo - 4 - *n* - propyl - 4,5 - dihydro - s - triazolo[1,5 - a]pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
2-ethoxycarbonylamino-6-methyl-5-oxo-4- <i>n</i> -propyl-4,5-dihydro-s-triazolo[1,5-a]-pyrimidine	0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

EXAMPLE 13

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - allyl - 4,5 - dihydro - s - triazolo[1,5 - a]pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x

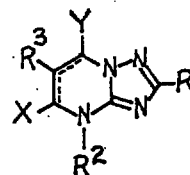
2-amino-6-methyl-5-oxo-4-allyl-4,5-dihydro-s-triazolo[1,5-a]pyrimidine

0.05
to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

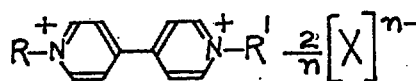
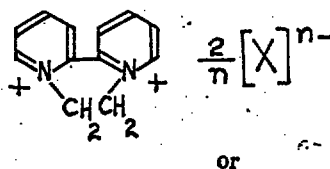
WHAT WE CLAIM IS:—

1. A herbicidal composition comprising a salt of a herbicidal bipyridylium quaternary cation and a s - triazolo[1,5 - a]pyrimidine derivative of the formula:



wherein R¹ is an amino, alkylamino, phenyl-alkylamino, dialkylamino, ureido, carboxyl, hydroxyalkyl or carbazoyl group; R² is an alkyl, cycloalkyl or alkenyl group; R³ is hydrogen, halogen or an alkyl or hydroxy-alkyl group; one of X and Y stands for an oxo or thioxo radical, and the other of X and Y stands for hydrogen or an alkyl radical and when X stands for an oxo or thioxo radical, the nucleus contains a double bond between the carbon atoms in positions 6 and 7, and when Y stands for an oxo or thioxo radical, the nucleus contains a double bond between the carbon atoms in positions 5 and 6; or a base addition salt of a triazolo - pyrimidine derivative as defined above which contains an acidic group; or an acylated derivative, or base addition salt thereof.

2. A composition as claimed in Claim 1, in which the salt of the herbicidal bipyridylium cation is a compound of the formula:



wherein R and R¹, which may be the same or different, stand for alkyl radicals of from 1 to 4 carbon atoms which may be substituted by hydroxyl, halogen, carboxyl, alkoxy, alkylcarbonyl, alkoxy carbonyl, carbamoyl or N - substituted carbamoyl; [X]ⁿ⁻ represents an anion and n is an integer from 1 to 4 inclusive.

3. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bi-

EXAMPLE 10

This Example illustrates a composition according to the invention which comprises 2 - *n* - propylamino - 6 - methyl - 5 - oxo - 4 - *n* - propyl - 4,5 - dihydro - *s* - triazolo[1,5 - *a*] pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
2- <i>n</i> -propylamino-6-methyl-5-oxo-4- <i>n</i> -propyl-4,5-dihydro- <i>s</i> -triazolo[1,5- <i>a</i>]-pyrimidine	0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

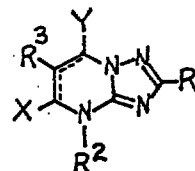
2-amino-6-methyl-5-oxo-4-allyl-4,5-dihydro-*s*-triazolo[1,5-*a*]pyrimidine

0.05
to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

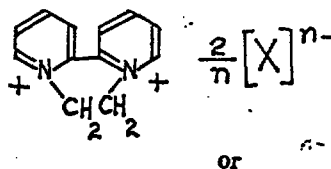
WHAT WE CLAIM IS:—

1. A herbicidal composition comprising a salt of a herbicidal bipyridylium quaternary cation and a *s* - triazolo[1,5 - *a*]pyrimidine derivative of the formula:

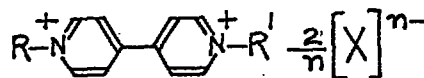


wherein R¹ is an amino, alkylamino, phenyl-alkylamino, dialkylamino, ureido, carboxyl, hydroxyalkyl or carbazoyl group; R² is an alkyl, cycloalkyl or alkenyl group; R³ is hydrogen, halogen or an alkyl or hydroxy-alkyl group; one of X and Y stands for an oxo or thioxo radical, and the other of X and Y stands for hydrogen or an alkyl radical and when X stands for an oxo or thioxo radical, the nucleus contains a double bond between the carbon atoms in positions 6 and 7, and when Y stands for an oxo or thioxo radical, the nucleus contains a double bond between the carbon atoms in positions 5 and 6; or a base addition salt of a triazolo - pyrimidine derivative as defined above which contains an acidic group; or an acylated derivative, or base addition salt thereof.

2. A composition as claimed in Claim 1, in which the salt of the herbicidal bipyridylium cation is a compound of the formula:



or



wherein R and R¹, which may be the same or different, stand for alkyl radicals of from 1 to 4 carbon atoms which may be substituted by hydroxyl, halogen, carboxyl, alkoxy, alkylcarbonyl, alkoxycarbonyl, carbamoyl or N - substituted carbamoyl; [X]ⁿ⁻ represents an anion and n is an integer from 1 to 4 inclusive.

3. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bi-

EXAMPLE 11

This Example illustrates a composition according to the invention which comprises 2 - amino - 5 - oxo - 4,6 - di - *n* - propyl - 4,5 - dihydro - *s* - triazolo[1,5 - *a*]pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
2-amino-5-oxo-4,6-di- <i>n</i> -propyl-4,5-dihydro- <i>s</i> -triazolo[1,5- <i>a</i>]pyrimidine	0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

EXAMPLE 12

This Example illustrates a composition according to the invention which comprises 2 - ethoxycarbonylamino - 6 - methyl - 5 - oxo - 4 - *n* - propyl - 4,5 - dihydro - *s* - triazolo[1,5 - *a*]pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x
2-ethoxycarbonylamino-6-methyl-5-oxo-4- <i>n</i> -propyl-4,5-dihydro- <i>s</i> -triazolo[1,5- <i>a</i>]-pyrimidine	0.05
Water	to 100 ml

(where x gives 200 ± 5 g/litre paraquat cation)

EXAMPLE 13

This Example illustrates a composition according to the invention which comprises 2 - amino - 6 - methyl - 5 - oxo - 4 - allyl - 4,5 - dihydro - *s* - triazolo[1,5 - *a*]pyrimidine in aqueous solution.

Ingredients	% w/v
Paraquat concentrate	x



pyridylum quaternary cation is 1,1 - dimethyl - 4,4' - bipyridylum di(methylsulphate).

4. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - ethylene - 2,2' - bipyridylum dibromide.

5. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - dimethyl - 4,4' - bipyridylum dichloride.

6. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - di - 2-hydroxyethyl - 4,4' - bipyridylum dichloride.

7. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1 - (2 - hydroxyethyl) - 1' - methyl - 4,4' - bipyridylum dichloride.

8. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - dicarbamoymethyl - 4,4' - bipyridylum dichloride.

9. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - bis-N,N - dimethylcarbamoymethyl - 4,4' - bipyridylum dichloride.

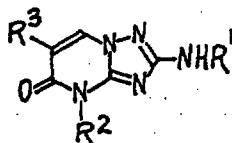
10. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - dimethyl - 4,4' - bipyridylum sulphate.

11. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - bis-N,N - diethylcarbamoymethyl - 4,4' - bipyridylum dichloride.

12. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - diacetonyl - 4,4' - bipyridylum dichloride.

13. A composition as claimed in Claim 1 or 2 in which the salt of the herbicidal bipyridylum quaternary cation is 1,1' - diethoxycarbonylmethyl - 4,4' - bipyridylum dibromide.

14. A composition as claimed in any of the preceding Claims in which the s - triazolo[1,5 - a]pyrimidine is a compound of the formula:



wherein R¹ is hydrogen or a C₁₋₄-alkyl radical, or a COOEt radical, R² is a C₁₋₄-alkyl radical, or an allyl radical, and R³ is a C₁₋₄-alkyl radical.

15. A composition as claimed in any of the preceding Claims in which the s - tri-

azolo[1,5 - a]pyrimidine is 2 - amino - 6-methyl - 5 - oxo - 4 - n - propyl - 4,5-dihydro - s - triazolo[1,5 - a]pyrimidine.

16. A composition as claimed in any one of the Claims 1 to 14 in which the s - triazolo[1,5 - a]pyrimidine is 6 - methyl - 5-oxo - 4 - n - propyl - 2 - n - propylamino-4,5 - dihydro - s - triazolo[1,5 - a]pyrimidine.

17. A composition as claimed in any one of Claims 1 to 14 in which the s - triazolo[1,5 - a]pyrimidine is 4 - allyl - 2 - amino-6 - methyl - 5 - oxo - 4,5 - dihydro - s-triazolo[1,5 - a]pyrimidine.

18. A composition as claimed in any one of Claims 1 to 14 in which the s - triazolo[1,5 - a]pyrimidine is 2 - amino - 4,6 - di-n - propyl - 5 - oxo - 4,5 - dihydro - s-triazolo[1,5 - a]pyrimidine.

19. A composition as claimed in any one of Claims 1 to 14 in which the s - triazolo[1,5 - a]pyrimidine is 2 - ethoxycarbonylamino - 6 - methyl - 5 - oxo - 4 - n - propyl-4,5 - dihydro - s - triazolo[1,5 - a]pyrimidine.

20. A composition as claimed in Claim 15 in which the amount of 2 - amino - 6-methyl - 5 - oxo - 4 - n - propyl - 4,5-dihydro - s - triazolo[1,5 - a]pyrimidine is 0.1 to 5.0 parts per 100 parts of herbicidal bipyridylum quaternary cation.

21. A composition as claimed in Claim 15 in which the amount of 2 - amino - 6-methyl - 5 - oxo - 4 - n - propyl - 4,5-dihydro - s - triazolo[1,5 - a]pyrimidine is 0.25 to 2.0 parts per 100 parts of herbicidal bipyridylum quaternary cation.

22. A composition as claimed in any of the preceding Claims comprising a carrier or diluent.

23. A composition as claimed in Claim 22 in which the carrier or diluent is a solid.

24. A composition as claimed in Claim 22 in which the carrier or diluent is aqueous.

25. A composition as claimed in any of Claims 22 to 24 comprising a surface active agent.

26. A composition as claimed in any of Claims 22 to 25 comprising a stenching agent.

27. A composition as claimed in Claim 26 in which the stenching agent is an alkyl pyridine base.

28. A composition as claimed in any of Claims 22 to 27 comprising a coloured dye-stuff or pigment compound.

29. A composition as claimed in Claim 24 comprising a gelling agent.

30. A composition as claimed in any of the preceding Claims comprising any other herbicidal compound.

31. A herbicidal composition comprising an aqueous solution of a salt of a paraquat cation containing 50 to 400 grams per litre of the paraquat cation and from 0.1 to 5



parts by weight of 2 - amino - 6 - methyl- substantially as herein described with particular reference to any one of the Examples.
5 - oxo - 4 - n - propyl - 4,5 - dihydro-
s - triazolo[1,5 - a]pyrimidine per 100 parts
by weight paraquat cation.

5 32. Compositions as claimed in Claim 1

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