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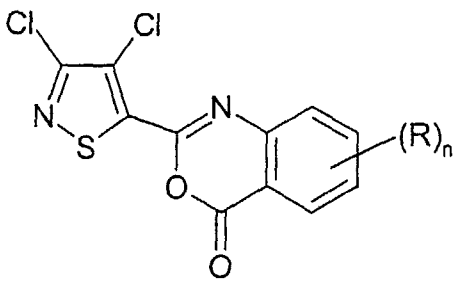
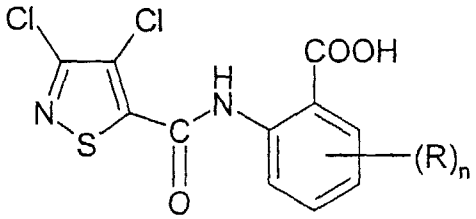
(54) Title: ISOTHIAZOLYL-BENZOXAZINE DERIVATIVES



(I)



(II)

(57) Abstract: Novel isothiazolyl-benzoxazine derivatives of the formula (I) wherein R and n have the meanings given in the specification, a process for the preparation of the new compounds and their use as microbicides Novel intermediates of the formula (II) wherein R and n have the meanings given in the specification, and a process for the preparation of these intermediates.

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

Isothiazolyl-benzoxazine Derivatives

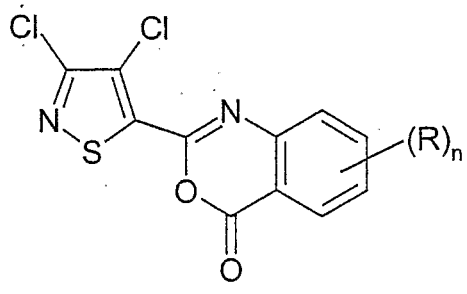
The present invention relates to novel isothiazolyl-benzoxazine derivatives, to a process for their preparation and to their use as microbicides. Further, the invention relates to novel intermediates and to a process for their preparation.

It has already been known that certain benzoxazines can be employed for the control of plant pests (cf. JP-A 43 488-1999 and JP-A 19 691-2001). The activity of such known compounds, however, is not always satisfactory.

Further, it has already been disclosed that another group of benzoxazines is herbicidally active (cf. US-A 3 914 121, JP-A 141 476-1980 and 108 776-1981). The use of such compounds as microbicides, however, has not been described as yet.

After all, it has already been known that a group of thiadiazole derivatives comprising a benzoxazine ring as a part of the molecule can be employed for the control of harmful organisms (cf. JP-A 139 566-2001). At low dosages, however, the activity of these compounds is not always satisfactory.

There have now been found novel isothiazolyl-benzoxazine derivatives of the formula



(I)

wherein

R represents halogen, alkyl, alkoxy, alkylthio, alkylsulfonyl, haloalkyl, haloalkoxy, carboxy, N,N-dialkyl-sulfamoyl, acylamino, alkoxycarbonylamino,

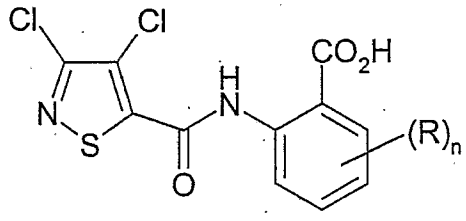
phenyl, phenoxy or nitro, or two adjacent radicals may together form a group selected from alkylene, alkenylene, alkylenedioxy or haloalkylenedioxy, and

n represents 0, 1, 2, 3 or 4.

If n represents an integer of 2 or more, the radicals may be identical or different.

Further, it has been found that the isothiazolyl-benzoxazine derivatives of the formula (I) can be prepared by a process, in which

a) isothiazolyl-caboxamides of the formula



(II)

wherein

R and n have the above-mentioned meanings,

are reacted with dehydrating agents in the presence of an inert diluent.

Finally, it has been found that the isothiazolyl-benzoxazine derivatives of the formula (I) are outstandingly active as microbicides in agriculture and horticulture, particularly as fungicides for the direct control of plant diseases or for causing resistance in plants against plant pathogens.

Surprisingly, the isothiazolyl-benzoxazine derivatives according to the invention have a much better microbicidal activity than the already known compounds, which are structurally most similar and have the same type of action.

In the present context, the terms quoted below have the following meanings:

"Halogen" and halogen in "haloalkyl", "haloalkoxy" and "haloalkylenedioxy" represents fluoro, chloro, bromo or iodo.

"Alkyl" can be a straight-chain or a branched-chain and there can be exemplarily mentioned, methyl, ethyl, n- or iso-propyl, n-, iso-, sec- or tert-butyl, n-pentyl, n-hexyl, etc.

"Alkylene" can be a straight-chain or a branched-chain and includes, for example, trimethylene, methyltrimethylene, 2-methyltrimethylene, tetramethylene, etc.

"Alkenylene" can be a straight-chain or a branched-chain comprising two or more double bonds and includes, for example, butadienylene, 1-methylbutadienylene, 2-methylbutadienylene, 1-ethylbutadienylene, 2-ethylbutadienylene, etc.

"Alkoxy" can be a straight-chain or a branched-chain and there can be exemplarily mentioned, methoxy, ethoxy, n- or iso-propoxy, n-, iso-, sec- or tert-butoxy, n-pentyloxy, n-hexyloxy, etc.

"Alkylthio" can be a straight-chain or a branched-chain and there can be exemplarily mentioned, methylthio, ethylthio, n- or iso-propylthio, n-, iso-, sec- or tert-butylthio, n-pentylthio, n-hexylthio, etc.

"Alkylsulfonyl" is an alkyl-SO_2-group, the alkyl part of which can be a straight-chain or a branched-chain, and there can be exemplarily mentioned, methylsulfonyl, ethylsulfonyl, n- or iso-propylsulfonyl, n-, iso-, sec- or tert-butylsulfonyl, n-pentylsulfonyl, n-hexylsulfonyl, etc.

"Haloalkyl" represents straight-chain or branched-chain alkyl, wherein at least one of the hydrogen atoms is substituted by halogen, and there can be exemplarily

mentioned, C_{1-4} alkyl substituted with 1-9 fluoro and/or chloro, and as specific examples there can be mentioned difluoromethyl, trifluoromethyl, chlorodifluoromethyl, 2-fluoroethyl, 2-chloroethyl, 2,2,2-trifluoroethyl, 1,1,2,2-tetrafluoroethyl, 1,1,2,2,2-pentafluoroethyl, 2-chloro-1,1,2-trifluoroethyl, 3-fluoropropyl, 3-chloropropyl, 2,2,3,3,3-pentafluoropropyl, 1,2,2,3,3,3-hexafluoropropyl, perfluorobutyl, etc.

"Haloalkoxy" is a radical the haloalkyl part of which has the above-mentioned meanings, and there can be specifically mentioned, difluoromethoxy, chlorodifluoromethoxy, trifluoromethoxy, 2-fluoroethoxy, 2-chloroethoxy, 2-bromoethoxy, 2,2,2-trifluoroethoxy, 3-chloropropoxy, etc.

As "haloalkylenedioxy" there can be exemplarily mentioned, difluoromethylenedioxy, tetrafluoroethylenedioxy, etc.

"N,N-Dialkylsulfamoyl" is a radical the alkyl parts of which have the aforementioned meanings and there can be exemplarily mentioned, N,N-dimethylsulfamoyl, N,N-diethylsulfamoyl, N,N-di-(n-propyl)sulfamoyl, N,N-di(n-butyl)-sulfamoyl, etc.

"Acylamino" represents an alkyl-CO-NH-group, the alkyl part of which can be a straight-chain or a branched-chain, and there can be specifically mentioned acetylamino, propionylamino, butyrylamino, isobutyrylamino, valerylamino, isovalerylamino, pivaloylamino, etc.

"Alkoxycarbonylamino" is an alkoxy-CO-NH-group, wherein the alkoxy part has the above-mentioned meanings and there can be exemplarily mentioned, methoxycarbonylamino, ethoxycarbonylamino, n- or iso-propoxycarbonylamino, n-, iso-, sec- or tert-butoxycarbonylamino, etc.

Formula (I) provides a general definition of the isothiazolyl-benzoxazine derivatives according to the invention. Preferred compounds of the formula (I) are those in which

R represents fluoro, chloro, bromo, iodo, alkyl having 1 or 6 carbon atoms, alkoxy having 1 to 6 carbon atoms, alkylthio having 1 to 6 carbon atoms, alkylsulfonyl having 1 to 6 carbon atoms, haloalkyl having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, haloalkoxy having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, carboxy, N,N-dialkyl-sulfamoyl having 1 to 6 carbon atoms in each of the alkyl groups, alkylcarbonylamino having 1 to 6 carbon atoms in the alkyl part, alkoxy-carbonylamino having 1 to 6 carbon atoms in the alkoxy part, phenyl, phenoxy or nitro,

or two adjacent radicals together form a group selected from alkylene having 3 to 6 carbon atoms, alkenylene having 4 to 6 carbon atoms, alkylenedioxy having 1 or 2 carbon atoms and 2 non-adjacent oxygen atoms, or halo-alkylenedioxy having 1 to 4 carbon atoms and 2 non-adjacent oxygen atoms and 1 to 5 identical or different halogen atoms, and

n is 0, 1, 2, 3, or 4.

If n represents an integer of 2 or more, the radicals may be identical or different.

Particularly, preferred are isothiazolyl-benzoxazine derivatives of the formula (I), wherein

R represents fluoro, chloro, bromo, iodo, alkyl having 1 to 4 carbon atoms, alkoxy having 1 to 4 carbon atoms, alkylthio having 1 to 4 carbon atoms, alkylsulfonyl having 1 to 4 carbon atoms, haloalkyl having 1 to 4 carbon atoms and 1 to 5 identical or different halogen atoms, haloalkoxy having 1 to

4 carbon atoms and 1 to 5 identical or different halogen atoms, carboxy, N,N-dialkyl-sulfamoyl having 1 to 4 carbon atoms in each of the alkyl groups, alkylcarbonylamino having 1 to 4 carbon atoms in the alkyl part, alkoxycarbonylamino having 1 to 4 carbon atoms in the alkoxy part, phenyl, phenoxy or nitro,

or two adjacent radicals together form a group selected from $-(CH_2)_3-$, $-(CH_2)_4-$, -CH=CH-CH=CH-, $-O-CH_2-O-$, $-O-CH_2-CH_2-O-$, $-O-CF_2-O-$ and $-O-CF_2-CF_2-O-$, and

n is 0, 1, 2, 3 or 4.

If n represents an interger of 2 or more, the radicals may be identical or different.

Very particularly preferred are isothiazolyl-benzoxazine derivatives of the formula (I), wherein

R represents fluoro, chloro, bromo, iodo, methyl, ethyl, methoxy, ethoxy, methylthio, methylsulfonyl, trifluoromethyl, trichloromethyl, difluoromethyl, trifluoromethoxy, difluoromethoxy, carboxy, N,N-dimethyl-sulfamoyl, methylcarbonylamino, ethylcarbonylamino, methoxycarbonylamino, ethoxy-carbonylamino, phenyl, phenoxy or nitro, or

two adjacent radicals represent a group selected from $-(CH_2)_3-$, $-(CH_2)_4-$, -CH=CH-CH=CH-, $-O-CH_2-O-$, $-O-CH_2-CH_2-O-$, $-O-CF_2-O-$ and $-O-CF_2-CF_2-O-$, and

n is 0, 1, 2, 3 or 4.

If n represents an integer of 2 or more, the radicals may be identical or different.

An even more preferred group of compounds are those isothiazolyl-benzoxazine derivatives of the formula (I), wherein

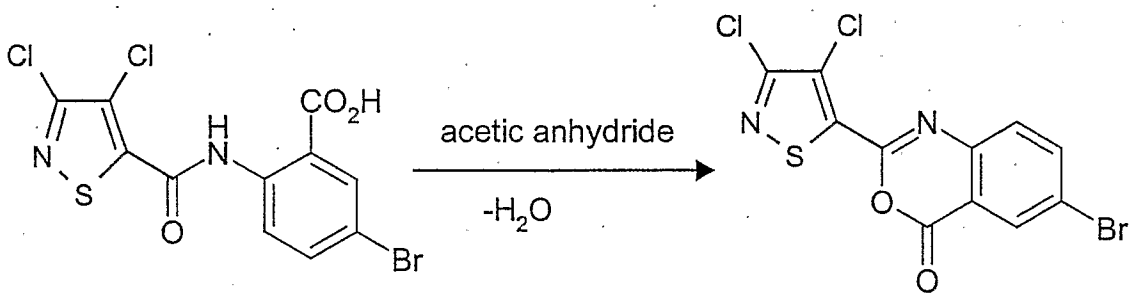
R represents fluoro, chloro, bromo, iodo, methyl, trifluoromethyl or nitro, or

two adjacent radicals represent the group -CH=CH-CH=CH-, and

n is 0, 1 or 2,

wherein the radicals may be identical or different, if n is 2.

If 5-bromo-2-(3,4-dichloro-isothiazol-5-yl-carbonylamino)-benzoic acid is used as starting material and acetic anhydride is used as dehydrating agent, the process (a) according to the invention can be illustrated by the following reaction scheme:



Formula (II) provides a general definition of the isothiazolyl-carboxamides, which are required as starting materials for carrying out process (a) according to the invention. In this formula, R and n preferably have those meanings, which have already been mentioned as preferred for this radical and this index.

The following compounds may be mentioned as specific examples of the isothiazolyl-carboxamides of the formula (II):

2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}benzoic acid,

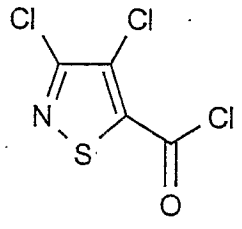
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-5-methylbenzoic acid,

2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-5-fluorobenzoic acid,

2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-5-chlorobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-5-bromobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-4-methylbenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-4-fluorobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-4-chlorobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-4-bromobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-4-nitrobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-6-methylbenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-6-fluorobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-6-chlorobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-6-bromobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-5-iodobenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-5-methoxybenzoic acid,
2-{[(3,4-dichloro-5-isothiazolyl)carbonyl]amino}-3,5-dichlorobenzoic acid and so on.

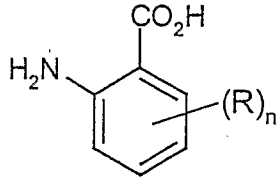
The isothiazolyl-carboxamides of the formula (II) are novel. They can be prepared by

b) reacting 3,4-dichloro-isothiazole-5-carboxylic acid chloride of the formula



(III)

with benzoic acid derivatives of the formula



(IV)

wherein

R and n have the above-mentioned meanings,

in the presence of a diluent and in the presence of an acid binding agent.

The 3,4-dichloro-isothiazole-5-carboxylic acid chloride of the formula (III) is a known compound (cf. JP-A 59024-1993).

Formula (IV) provides a general definition of the benzoic acid derivatives, which are required as reaction components for carrying out process (b) according to the invention. In this formula, R and n preferably have those meanings, which have already been mentioned in connection with the description of the substances of the formula (I) according to the invention as being preferred for this radical and this index.

The benzoic acid derivatives of the formula (IV) are known or can be prepared by known processes.

Suitable diluents for carrying out process (b) according to the invention are all customary inert organic solvents as well as water. Preference is given to using water.

Suitable acid binding agents for carrying out process (b) according to the invention are all bases, which are customary for such reactions. Preference is given to using hydroxides of alkali metals, such as lithium hydroxide, sodium hydroxide and potassium hydroxide.

When carrying out process (b) according to the invention, the reaction temperatures can be varied within a certain range. The reaction is generally carried out at a temperature between about 0°C and about 80°C, preferably between about 10°C and about 60°C.

When carrying out process (b) according to the invention, 3,4-dichloro-isothiazole-5-carboxylic acid chloride of the formula (III) and benzoic acid derivative of the formula (IV) are generally employed in equimolar amounts. However, it is also possible to use an excess of one or the other component. Working up is carried out by customary methods.

The isothiazolyl-carboxamides of the formula (II) are also suitable for the control of undesired microorganisms, in particular for controlling fungi.

When carrying out process (a) according to the invention, all customary dehydrating agents can be used for splitting off water in the course of the reaction. Examples of such dehydrating agents are acetic anhydride, benzoic anhydride and propionic anhydride.

Suitable diluents for carrying out the process (a) according to the invention are all customary inert organic solvents. Preference is given to using aliphatic, alicyclic and aromatic hydrocarbons (that may optionally be chlorinated), for example, pentane, hexane, cyclohexane, petroleum ether, ligroine, benzene, toluene, xylene, dichloromethane, chloroform, carbon tetrachloride, 1,2-dichloroethane, chlorobenzene, dichlorobenzene, etc.; ethers, for example, ethyl ether, methyl ethyl ether, isopropyl ether, butyl ether, dioxane, dimethoxyethane (DME), tetrahydrofuran (THF), diethylene glycol dimethyl ether (DGM), etc.; ketones, for example, acetone, methyl ethyl ketone (MEK), methyl isopropyl ketone, methyl isobutyl ketone (MIBK), etc.; nitriles, for example, acetonitrile, propionitrile, acrylonitrile, etc.; esters, for example, ethyl acetate, amyl acetate, etc.; acid amides, for example, dimethylformamide (DMF), dimethylacetamide (DMA), N-methylpyrrolidone, 1,3-dimethyl-2-imidazolidinone, hexamethylphosphoric triamide (HMPA), etc.; acid anhydrides, for example, acetic anhydride, benzoic anhydride, etc.; sulfones, sulfoxides, for example, dimethyl sulfoxide (DMSO), sulfolane, etc. and bases, for example, pyridine etc.

When carrying out the process (a) according to the invention, the reaction temperatures can be varied within a substantially wide range. The reaction is generally carried out at temperatures between about -10°C and about 250°C, preferably between about 20°C and about 180°C.

The process (a) according to the invention is generally carried out under atmospheric pressure but, if desired, can also be carried out under elevated or reduced pressure.

When carrying out the process (a) according to the invention, in general an isothiazolyl-carboxamide of the formula (II) is reacted in the presence of a large excess of a dehydrating agent. If acetic anhydride is used as dehydrating agent it may simultaneously serve as a diluent. Working-up is carried out by customary methods.

The compounds according to the present invention exhibit a strong microbicidal activity. Thus, they can be used for combating undesired microorganisms, such as phytopathogenic fungi and bacteriae, in agriculture and horticulture. The compounds are suitable for the direct control of undesired microorganisms as well as for generating resistance in plants against attack by undesired plant pathogens.

Resistance-inducing substances in the present context are to be understood as those substances which are capable of stimulating the defence system of plants such that the treated plants, when subsequently inoculated with undesirable microorganisms, display substantial resistance to these microorganisms.

Undesirable microorganisms in the present case are to be understood as phytopathogenic fungi and bacteriae. The substances according to the invention can thus be employed to generate resistance in plants against attack by the harmful organisms mentioned within a certain period of time after the treatment. The period of time within which resistance is brought about in general extends from 1 to 10 days, preferably 1 to 7 days, after treatment of the plants with the active compounds.

Generally, the compounds according to the invention can be used as fungicides for combating phytopathogenic fungi, such as Plasmodiophoromycetes, Oomycetes, Chytridiomycetes, Zygomycetes, Ascomycetes, Basidiomycetes and Deuteromycetes, and can also be used as bactericides for combating bacteriae, such as Pseudomonadaceae, Rhizobiaceae, Enterobacteriaceae, Corynebacteriaceae, Streptomycetaceae, Proteobacteriae and Gram-positive groups.

Some pathogens causing fungal diseases which come under the generic names listed above are mentioned as examples, but not by way of limitation:

Erwinia species, such as, for example, Erwinia amylovora;
Pythium species, such as, for example, Pythium ultimum;
Phytophthora species, such as, for example, Phytophthora infestans;
Pseudoperonospora species, such as, for example, Pseudoperonospora humuli or Pseudoperonospora cubensis;
Plasmopara species, such as, for example, Plasmopara viticola;
Bremia species, such as, for example, Bremia Lactucae;
Peronospora species, such as, for example, Peronospora pisi or P. brassicae;
Erysiphe species, such as, for example, Erysiphe graminis;
Sphaerotheca species, such as, for example, Sphaerotheca fuliginea;
Podosphaera species, such as, for example, Podosphaera leucotricha;
Venturia species, such as, for example, Venturi inaequalis;
Pyrenophora species, such as, for example, Pyrenophora teres or P. graminea (conidia form: Drechslera, syn: Helminthosporium);
Cochliobolus species, such as, for example, Cochliobolus sativus (conidia form: Drechslera,syn: Helminthosporium);
Uromyces species, such as, for example, Uromyces appendiculatus;
Puccinia species, such as, for example, Puccinia recondita;
Sclerotinia species, such as, for example, Sclerotinia sclerotiorum;
Tilletia species, such as, for example, Tilletia caries;

Ustilago species, such as, for example, Ustilago nuda or Ustilago avenae;
Pellicularia species, such as, for example, Pellicularia sasakii;
Pyricularia species, such as, for example, Pyricularia oryzae;
Fusarium species, such as, for example, Fusarium culmorum;
Botrytis species, such as, for example, Botrytis cinerea;
Septoria species, such as, for example, Leptosphaeria nodorum;
Cercospora species, such as, for example, Cercospora canescens;
Alternaria species, such as, for example, Alternaria brassicae; and
Pseudocercosporella species, such as, for example, Pseudocercosporella herpotrichoides.

The compounds according to the present invention are particularly suitable for causing resistance against infection of plants by plant pathogens, such as Pyricularia oryzae etc.

The good toleration, by plants, of the active compounds, at the concentrations required for combating plants diseases, permits treatment of above-ground parts of plants, of vegetative propagation stock and seeds, and of the soil.

The compounds according to the present invention have a low toxicity against warm-blooded animals and therefore can be used safely.

The active compounds can be converted into the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, foams, pastes, granules, tablets, aerosols, natural and synthetic materials impregnated with active compound, very fine capsules in polymeric substances, coating compositions for use on seed, and formulations used with burning equipment, such as fumigating cartridges, fumigating cans and fumigating coils, as well as ULV cold mist and warm mist formulations.

These formulations may be produced in known manner, for example by mixing the active compounds with extenders, that is to say liquid or liquefied gaseous or solid diluents or carriers, optionally with the use of surface-active agents, that is to say emulsifying agents and/or dispersing agents and/or foam-forming agents. In the case of the use of water as an extender, organic solvents can, for example, also be used as auxiliary solvents.

As liquid solvents diluents or carriers, there are suitable in the main, aromatic hydrocarbons such as xylene, toluene or alkyl naphthalenes, chlorinated aromatic or chlorinated aliphatic hydrocarbons, such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example mineral oil fractions, alcohols, such as butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl-isobutyl ketone or cyclohexanone, or strongly polar solvents, such as dimethylformamide and dimethyl-sulphoxide, as well as water.

By liquefied gaseous diluents or carriers are meant liquids which would be gaseous at normal temperature and under normal pressure, for example aerosol propellants, such as halogenated hydrocarbons as well as butane, propane, nitrogen and carbon dioxide.

As solid carriers there may be used ground natural minerals, such as kaolings, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly-dispersed silicic acid, alumina and silicates. As solid carriers for granules there may be used crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks.

As emulsifying and/or foam-forming agents there may be used non-ionic and anionic emulsifiers, such as polyoxyethylene-fatty acid esters, polyoxyethylene-fatty alcohol

ethers, for example alkylaryl polyglycol ethers, alkyl sulphonates, alkyl sulphates, aryl sulphonates as well as albumin hydrolysis products.

Dispersing agents include, for example, lignin sulphite waste liquors and methylcellulose.

Adhesives such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, can be used in the formulation.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs or metal phthalocyanine dyestuffs, and trace nutrients, such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations in general contain from 0.1 to 95 per cent by weight of active compound, preferably from 0.5 to 90 per cent by weight.

The active compounds according to the invention can be present in the formulations or in the various use forms as a mixture with other known active compounds, such as fungicides, bactericides, insecticides, acaricides, nematicides, herbicides, bird repellents, growth factors, plant nutrients and agents for improving soil structure.

In many cases, synergistic effects are achieved, i.e. the activity of the mixture exceeds the activity of the individual components.

Examples of co-components in mixtures are the following compounds:

Fungicides:

aldimorph, ampropylfos, ampropylfos potassium, andoprim, anilazine, azaconazole, azoxystrobin,

benalaxyl, benodanil, benomyl, benzamacril, benzamacril-isobutyl, bialaphos, binapacryl, biphenyl, bitertanol, blasticidin-S, bromuconazole, bupirimate, buthiobate,

calcium polysulphide, capsimycin, captafol, captan, carbendazim, carboxin, carvon, quinomethionate, chlobenthiazone, chlorfenazole, chloroneb, chloropicrin, chlorothalonil, chlozolinate, clozylacon, cufraneb, cymoxanil, cyproconazole, cyprodinil, cyprofuram, carpropamide,

debacarb, dichlorophen, diclobutrazole, diclofluanid, diclomezine, dicloran, diethofencarb, difenoconazole, dimethirimol, dimethomorph, diniconazole, diniconazole-M, dinocap, diphenylamine, dipyrithione, ditalimfos, dithianon, dodemorph, dodine, drazoxolon,

edifenphos, epoxiconazole, etaconazole, ethirimol, etridiazole,

famoxadon, fenapanil, fenarimol, fenbuconazole, fenfuram, fenitropan, fenpiclonil, fenpropidin, fenpropimorph, fentin acetate, fentin hydroxide, ferbam, ferimzone, fluazinam, flumetover, fluoromide, fluquinconazole, flurprimidol, flusilazole, flusulfamide, flutolanil, flutriafol, folpet, fosetyl-aluminium, fosetyl-sodium, fthalide, fuberidazole, furalaxyl, furametpyr, furcarbonil, furconazole, furconazole-cis, furmecyclox, fenhexamide, fluoxastrobin,

guazatine,

hexachlorobenzene, hexaconazole, hymexazole,

imazalil, imibenconazole, iminoctadine, iminoctadine albesilate, iminoctadine triacetate, iodocarb, ipconazole, iprobenfos (IBP), iprodione, irumamycin, isoprothiolane, isovaledione, iprovalicarb,

kasugamycin, kresoxim-methyl, copper preparations, such as: copper hydroxide, copper naphthenate, copper oxychloride, copper sulphate, copper oxide, oxine-copper and Bordeaux mixture,

mancopper, mancozeb, maneb, meferimzone, mepanipyrim, mepronil, metalaxyl, metconazole, methasulfocarb, methfuroxam, metiram, metomeclam, metsulfovax, mildiomycin, myclobutanil, myclozolin,

nickel dimethyldithiocarbamate, nitrothal-isopropyl, nuarimol,

ofurace, oxadixyl, oxamocarb, oxolinic acid, oxycarboxim, oxyfenthiin,

paclobutrazole, pefurazoate, penconazole, pencycuron, phosdiphen, pimaricin, piperalin, polyoxin, polyoxorim, probenazole, prochloraz, procymidone, propamocarb, propanosine-sodium, propiconazole, propineb, pyrazophos, pyrifenox, pyrimethanil, pyroquilon, pyroxyfur, prothioconazole,

quinconazole, quintozene (PCNB), quinoxyfen,

sulphur and sulphur preparations, spiroxamine,

tebuconazole, tecloftalam, tecnazene, tetcyclacis, tetraconazole, thiabendazole, thicyofen, thifluzamide, thiophanate-methyl, thiram, tioxymid, tolclofos-methyl, tolylfluanid, triadimefon, triadimenol, triazbutil, triazoxide, trichlamide, tricyclazole, tridemorph, triflumizole, triforine, triticonazole, trifloxystrobin,

uniconazole,

validamycin A, vinclozolin, viniconazole,

zarilamide, zineb, ziram and also

Dagger G,

OK-8705,

OK-8801,

α-(1,1-dimethylethyl)-β-(2-phenoxyethyl)-1H-1,2,4-triazole-1-ethanol,

α-(2,4-dichlorophenyl)-β-fluoro-β-propyl-1H-1,2,4-triazole-1-ethanol,

α-(2,4-dichlorophenyl)-β-methoxy-α-methyl-1H-1,2,4-triazole-1-ethanol,

α-(5-methyl-1,3-dioxan-5-yl)-β-[[4-(trifluoromethyl)-phenyl]-methylene]-1H-1,2,4-triazole-1-ethanol,

(5RS,6RS)-6-hydroxy-2,2,7,7-tetramethyl-5-(1H-1,2,4-triazol-1-yl)-3-octanone,

(E)-α-(methoxyimino)-N-methyl-2-phenoxy-phenylacetamide,

1-(2,4-dichlorophenyl)-2-(1H-1,2,4-triazol-1-yl)-ethanone O-(phenylmethyl)-oxime,

1-(2-methyl-1-naphthalenyl)-1H-pyrrol-2,5-dione,

1-(3,5-dichlorophenyl)-3-(2-propenyl)-2,5-pyrrolidinedione,

1-[(diiodomethyl)-sulphonyl]-4-methyl-benzene,
1-[[2-(2,4-dichlorophenyl)-1,3-dioxolan-2-yl]-methyl]-1H-imidazole,
1-[[2-(4-chlorophenyl)-3-phenyloxiranyl]-methyl]-1H-1,2,4-triazole,
1-[1-[2-[(2,4-dichlorophenyl)-methoxy]-phenyl]-ethenyl]-1H-imidazole,
1-methyl-5-nonyl-2-(phenylmethyl)-3-pyrrolidinole,
2',6'-dibromo-2-methyl-4'-trifluoromethoxy-4-trifluoro-methyl-1,3-thiazole-5-carboxanilide,
2,6-dichloro-5-(methylthio)-4-pyrimidinyl thiocyanate,
2,6-dichloro-N-(4-trifluoromethylbenzyl)-benzamide,
2,6-dichloro-N-[[4-(trifluoromethyl)-phenyl]-methyl]-benzamide,
2-(2,3,3-triiodo-2-propenyl)-2H-tetrazole,
2-[(1-methylethyl)-sulphonyl]-5-(trichloromethyl)-1,3,4-thiadiazole,
2-[[6-deoxy-4-O-(4-O-methyl-β-D-glycopyranosyl)-α-D-glucopyranosyl]-amino]-4-methoxy-1H-pyrrolo[2,3-d]pyrimidine-5-carbonitrile,
2-aminobutane,
2-bromo-2-(bromomethyl)-pentanedinitrile,
2-chloro-N-(2,3-dihydro-1,1,3-trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide,
2-chloro-N-(2,6-dimethylphenyl)-N-(isothiocyanatomethyl)-acetamide,
2-phenylphenol (OPP),
3,4-dichloro-1-[4-(difluoromethoxy)-phenyl]-1H-pyrrol-2,5-dione,
3,5-dichloro-N-[cyano[(1-methyl-2-propinyl)-oxy]-methyl]-benzamide,
3-(1,1-dimethylpropyl-1-oxo-1H-indene-2-carbonitrile,
3-[2-(4-chlorophenyl)-5-ethoxy-3-isoxazolidinyl]-pyridine,
4-chloro-2-cyano-N,N-dimethyl-5-(4-methylphenyl)-1H-imidazole-1-sulphonamide,
4-methyl-tetrazolo[1,5-a]quinazolin-5(4H)-one,
8-hydroxyquinoline sulphate,
9H-xanthene-2-[(phenylamino)-carbonyl]-9-carboxylic hydrazide,
bis-(1-methylethyl)-3-methyl-4-[(3-methylbenzoyl)-oxy] 2,5-thiophenedicarboxylate,
cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)-cycloheptanol,
cis-4-[3-[4-(1,1-dimethylpropyl)-phenyl-2-methylpropyl]-2,6-dimethyl-morpholinehydrochloride,

ethyl [(4-chlorophenyl)-azo]-cyanoacetate,
potassium hydrogen carbonate,
methanetetrathiol sodium salt,
methyl 1-(2,3-dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-imidazole-5-carboxylate,
methyl N-(2,6-dimethylphenyl)-N-(5-isoxazolylcarbonyl)-DL-alaninate,
methyl N-(chloroacetyl)-N-(2,6-dimethylphenyl)-DL-alaninate,
N-(2,6-dimethylphenyl)-2-methoxy-N-(tetrahydro-2-oxo-3-furanyl)-acetamide,
N-(2,6-dimethylphenyl)-2-methoxy-N-(tetrahydro-2-oxo-3-thienyl)-acetamide,
N-(2-chloro-4-nitrophenyl)-4-methyl-3-nitro-benzenesulphonamide,
N-(4-cyclohexylphenyl)-1,4,5,6-tetrahydro-2-pyrimidineamine,
N-(4-hexylphenyl)-1,4,5,6-tetrahydro-2-pyrimidineamine,
N-(5-chloro-2-methylphenyl)-2-methoxy-N-(2-oxo-3-oxazolidinyl)-acetamide,
N-(6-methoxy)-3-pyridinyl)-cyclopropanecarboxamide,
N-[2,2,2-trichloro-1-[(chloroacetyl)-amino]-ethyl]-benzamide,
N-[3-chloro-4,5-bis(2-propinyloxy)-phenyl]-N'-methoxy-methanimidamide,
N-formyl-N-hydroxy-DL-alanine-sodium salt,
O,O-diethyl [2-(dipropylamino)-2-oxoethyl]-ethylphosphoramidothioate,
O-methyl S-phenyl phenylpropylphosphoramidothioate,
S-methyl 1,2,3-benzothiadiazole-7-carbothioate,
spiro[2H]-1-benzopyran-2,1'(3'H)-isobenzofuran]-3'-one,

Bactericides:

bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin, octhilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin, tecloftalam, copper sulphate and other copper preparations.

Insecticides / acaricides / nematicides:

abamectin, acephate, acetamiprid, acrinathrin, alanycarb, aldicarb, aldoxycarb, alpha-cypermethrin, alphamethrin, amitraz, avermectin, AZ 60541, azadirachtin, azamethiphos, azinphos A, azinphos M, azocyclotin,

Bacillus popilliae, Bacillus sphaericus, Bacillus subtilis, Bacillus thuringiensis, baculoviruses, Beauveria bassiana, Beauveria tenella, bendiocarb, benfuracarb, bensultap, benzoximate, betacyfluthrin, bifenazate, bifenthrin, bioethanomethrin, biopermethrin, BPMC, bromophos A, bufencarb, buprofezin, butathiofos, butocarboxim, butylpyridaben,

cadusafos, carbaryl, carbofuran, carbophenothion, carbosulfan, cartap, chloethocarb, chlorethoxyfos, chlorfenapyr, chlorfenvinphos, chlorfluazuron, chlormephos, chlorpyrifos, chlorpyrifos M, chlovaporthrin, cis-resmethrin, cispermethrin, clocythrin, cloethocarb, clofentezine, cyanophos, cycloprene, cycloprothrin, cyfluthrin, cyhalothrin, cyhexatin, cypermethrin, cyromazine,

deltamethrin, demeton M, demeton S, demeton-S-methyl, diafenthiuron, diazinon, dichlorvos, diflubenzuron, dimethoat, dimethylvinphos, diofenolan, disulfoton, docusat-sodium, dofenapyn,

eflusilanate, emamectin, empenthrin, endosulfan, Entomopfthora spp., esfenvalerate, ethiofencarb, ethion, ethoprophos, etofenprox, etoxazole, etrimphos,

fenamiphos, fenazaquin, fenbutatin oxide, fenitrothion, fenothiocarb, fenoxacrim, fenoxycarb, fenpropathrin, fenpyrad, fenpyrithrin, fenpyroximate, fenvalerate, fipronil, fluazuron, flubrocythrinate, flucycloxuron, flucythrinate, flufenoxuron, flutenzine, fluvalinate, fonophos, fosmethilan, fosthiazate, fubfenprox, furathiocarb, granulosis viruses,

halofenozide, HCH, heptenophos, hexaflumuron, hexythiazox, hydroprene,

imidacloprid, isazophos, isofenphos, isoxathion, ivermectin,

lambda-cyhalothrin, lufenuron,

malathion, mecarbam, metaldehyde, methamidophos, Metharhizium anisopliae, Metharhizium flavoviride, methidathion, methiocarb, methomyl, methoxyfenozide, metolcarb, metoxadiazone, mevinphos, milbemectin, monocrotophos,

naled, nitenpyram, nithiazine, novaluron, nuclear polyhedrosis viruses,
omethoat, oxamyl, oxydemethon M,
Paecilomyces fumosoroseus, parathion A, parathion M, permethrin, phenthoat, phorat, phosalone, phosmet, phosphamidon, phoxim, pirimicarb, pirimiphos A, pirimiphos M, profenofos, promecarb, propoxur, prothiofos, prothoat, pymetrozine, pyraclofos, pyresmethrin, pyrethrum, pyridaben, pyridathion, pyrimidifen, pyriproxyfen,
quinalphos,
ribavirin,
salithion, sebufos, silafluofen, spinosad, sulfotep, sulprofos,
tau-fluvalinate, tebufenozide, tebufenpyrad, tebupirimiphos, teflubenzuron, tefluthrin, temephos, temivinphos, terbufos, tetrachlorvinphos, theta-cypermethrin, thiamethoxam, thiapronil, thiatriphos, thiocyclam hydrogen oxalate, thiodicarb, thiofanox, thuringiensin, tralocythrin, tralomethrin, triarathene, triazamate, triazophos, triazuron, trichlophenidine, trichlorfon, triflumuron, trimethacarb, thiacloprid,
vamidothion, vaniliprole, Verticillium lecanii,
YI 5302,
zeta-cypermethrin, zolaprofos,

(1R-cis)-[5-(phenylmethyl)-3-furanyl]-methyl-3-[(dihydro-2-oxo-3(2H)-furanylidene)-methyl] 2,2-dimethylcyclopropanecarboxylate,
(3-phenoxyphenyl)-methyl 2,2,3,3-tetramethylcyclopropanecarboxylate,
1-[(2-chloro-5-thiazolyl)methyl]tetrahydro-3,5-dimethyl-N-nitro-1,3,5-triazine-2(1H)-imine,
2-(2-chloro-6-fluorophenyl)-4-[4-(1,1-dimethylethyl)phenyl]-4,5-dihydro-oxazole,
2-(acetyloxy)-3-dodecyl-1,4-naphthalenedione,
2-chloro-N-[[[4-(1-phenylethoxy)-phenyl]-amino]-carbonyl]-benzamide,
2-chloro-N-[[[4-(2,2-dichloro-1,1-difluoroethoxy)-phenyl]-amino]-carbonyl]-benzamide,
3-methylphenyl propylcarbamate

4-[4-(4-ethoxyphenyl)-4-methylpentyl]-1-fluoro-2-phenoxy-benzene,
4-chloro-2-(1,1-dimethylethyl)-5-[[2-(2,6-dimethyl-4-phenoxyphenoxy)ethyl]thio]-3(2H)-pyridazinone,
4-chloro-2-(2-chloro-2-methylpropyl)-5-[(6-iodo-3-pyridinyl)methoxy]-3(2H)-pyridazinone,
4-chloro-5-[(6-chloro-3-pyridinyl)methoxy]-2-(3,4-dichlorophenyl)-3(2H)-pyridazinone,
Bacillus thuringiensis strain EG-2348,
[2-benzoyl-1-(1,1-dimethylethyl)-hydrazinobenzoic acid,
2,2-dimethyl-3-(2,4-dichlorophenyl)-2-oxo-1-oxaspiro[4.5]dec-3-en-4-yl butanoate,
[3-[(6-chloro-3-pyridinyl)methyl]-2-thiazolidinylidene]-cyanamide,
dihydro-2-(nitromethylene)-2H-1,3-thiazine-3(4H)-carboxaldehyde,
ethyl [2-[[1,6-dihydro-6-oxo-1-(phenylmethyl)-4-pyridazinyl]oxy]ethyl]-carbamate,
N-(3,4,4-trifluoro-1-oxo-3-butenyl)-glycine,
N-(4-chlorophenyl)-3-[4-(difluoromethoxy)phenyl]-4,5-dihydro-4-phenyl-1H-pyrazole-1-carboxamide,
N-[(2-chloro-5-thiazolyl)methyl]-N'-methyl-N''-nitro-guanidine,
N-methyl-N'-(1-methyl-2-propenyl)-1,2-hydrazinedicarbothioamide,
N-methyl-N'-2-propenyl-1,2-hydrazinedicarbothioamide,
O,O-diethyl [2-(dipropylamino)-2-oxoethyl]-ethylphosphoramidothioate.

The active compounds can be used as such or in the form of their formulations or the use forms prepared therefrom by further dilution, such as ready-to-use solutions, emulsions, suspensions, powders, tablets, pastes, microcapsules and granules. They are used in the customary manner, for example by watering, immersion, spraying, atomising, misting, vaporizing, injecting, forming a slurry, brushing on, dusting, scattering, dry dressing, moist dressing, wet dressing, slurry dressing or encrusting.

In the treatment of parts of plants, the active compounds concentration in the use forms can be varied within a substantial range. They are, in general, from 1 to 0.0001% by weight, preferably from 0.5 and 0.001%.

For the treatment of seed, amounts of active compound of 0.1 to 10 g, especially 1 to 5 g, are generally employed per kilogram of seed.

For the treatment of soil, active compound concentrations, at the point of action, of 0.00001 to 0.1% by weight, especially of 0.0001 to 0.02%, are generally employed.

As already mentioned above, all plants and parts of plants can be treated according to the invention. In a preferred embodiment naturally occurring plant species and plant varieties or those obtained by conventional biological breeding methods, such as crossbreeding or protoplast fusion as well as parts of such plants are treated. In an additional preferred embodiment transgenic plants and plant varieties which have been obtained by genetic engineering methods, possibly in combination with conventional methods (genetically modified organisms) and parts of such plants are treated. The term "parts" or "parts of plants" or "plant parts" is explained above.

According to the invention plants of the plant varieties commercially available or used at any particular time are very preferably treated. Plant varieties are understood to be plants with specific properties ("traits") which have been obtained both by conventional breeding, by mutagenesis or by recombinant DNA techniques. They can be varieties, biotypes or genotypes.

Depending on the species or varieties of plants, their location and growth conditions (the types of soil, climate, vegetation period and feed concerned), superadditive ("synergistic") effects can occur as a result of the treatment according to the invention. Effects such as for example reduced application rates and/or broadening of the activity spectra and/or increased activity of the compounds and compositions usable according to the invention, improved plant growth, increased tolerance of high or low temperatures, increased tolerance of dry conditions or water or ground salt contents, increased flowering capacity, facilitated harvesting, acceleration of maturity, increased crop yields, higher quality and/or increased nutritional value of

the harvested crops and increased storing quality and/or processibility of the harvested crops are possible, which are greater than those actually expected.

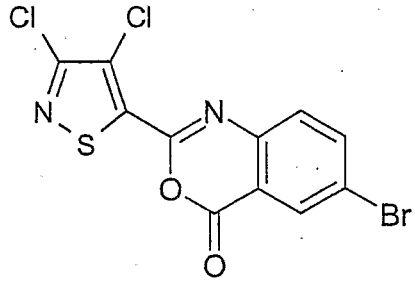
Preferred transgenic plants or plant varieties (obtained by genetic engineering) to be treated according to the invention include all plants which as a result of the genetic modification concerned have received genetic material which provides them with particularly advantageous valuable properties ("traits"). Examples of such properties are improved plant growth, increased tolerance of high or low temperatures, increased tolerance of dry conditions or water or ground salt contents, increased flowering capacity, facilitated harvesting, acceleration of maturity, increased crop yields, higher quality and/or increased nutritional value of the harvested crops and increased storing quality and/or processibility of the harvested crops. Additional and particularly noteworthy examples of such properties are increased resistance of the plants to animal and microbial pests, such as to insects, mites, phytopathogenic fungi, bacteria and/or viruses as well as increased tolerance by the plants of certain herbicidal active compounds. Examples which may be mentioned of transgenic plants are the important crop plants such as cereals (wheat and rice), corn, soybeans, potatoes, cotton, rape and fruit plants (producing apples, pears, citrus fruits and grapes), the crop plants corn, soybeans, potatoes, cotton and rape being particularly noteworthy. Particularly significant properties ("traits") are increased resistance of the plants to insects due to the toxins forming in the plants, and in particular those which are produced in the plants (hereinafter referred to as "Bt plants") by the genetic material obtained from Bacillus Thuringiensis (e.g. by the genes CrylA(a), CrylA(b), CrylA(c), CryllA, CryIIIB2, Cry9c Cry2Ab, Cry3Bb and CryIF and combinations thereof). Particularly significant properties ("traits") are the increased resistance of plants to fungi, bacteria and viruses due to systemically acquired resistance (SAR), systemin, phytoalexins, elicitors and resistance genes and correspondingly expressed proteins and toxins. Particularly significant properties ("traits") are also increased tolerance by the plants of certain herbicidal active compounds, such as for example imidazolinones, sulphonylureas, glyphosate or phosphinotricine (e.g. the "PAT" gene). The corresponding genes imparting the required properties ("traits") can also

occur in the transgenic plants in combination with each other. Examples which may be mentioned of "Bt plants" are varieties of corn, cotton, soybeans and potatoes which are sold under the trade names YIELD GARD® (e.g. corn, cotton, soybeans), KnockOut® (e.g. corn), StarLink® (e.g. corn), Bollgard® (cotton), Nucotn® (cotton) and NewLeaf® (potatoes). Examples which may be mentioned of herbicide-tolerant plants are varieties of corn, cotton and soybeans which are sold under the trade names Roundup Ready® (tolerance of glyphosate, e.g. corn, cotton, soybeans), Liberty Link® (tolerance of phosphinotricine, e.g. rape), IMI® (tolerance of imidazolinones) and STS® (tolerance of sulphonylureas, e.g. corn). Herbicide-resistant plants (bred for herbicide tolerance in the conventional manner) which may be mentioned are also the varieties (e.g. corn) sold under the name Clearfield®. The above statements do of course also apply to any plant varieties which may be developed in the future or launched onto the market in the future and which have the genetic properties ("traits") described above or developed in the future.

According to the invention the above-mentioned plants can be particularly advantageously treated with the compounds of the general formula I or the active compound mixtures according to the invention. The preferred ranges mentioned above for the active compounds or mixtures also apply to the treatment of these plants. Particularly advantageous is the treatment of plants with the compounds or mixtures specifically listed in the present text.

The preparation and the use of the compounds according to the invention is illustrated by the following examples. The invention, however, is not limited to said examples in any way.

Synthesis Example 1



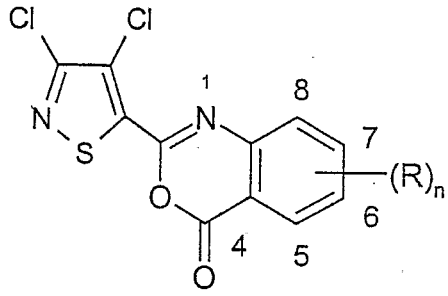
(Compound No. I -7)

15 ml of acetic anhydride were added to 1.0 g 2-(3,4-dichloro-isothiazol-5-yl-carbonylamino)-5-bromo-benzoic acid, and the resulting mixture was refluxed for 2 hours. Subsequently, the reaction mixture was cooled down to room temperature and was then further cooled by means of ice water. The precipitating crystals were filtered off and washed with methanol to obtain 2-(3,4-dichloro-isothiazol-5-yl)-6-bromo-4H-4-oxo-3,1-benzoxazine (0.6 g).

m p 169-173°C.

The compounds shown in the following Table 1 were also prepared according to the method described in Synthesis Example 1.

Table 1



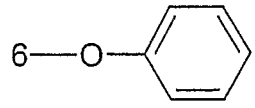
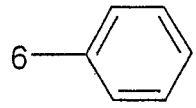
(I)

Compound No.	$(R)_n$	mp (°C) or n_D^{20}
I-1	-	191-192
I-2	8-CH_3	211-222
I-3	8-Cl	208-209
I-4	7-Cl	203-206
I-5	6-Cl	191-194
I-6	5-Cl	204-206
I-7	6-Br	169-173
I-8	6-CH_3	189-190
I-9	5-CH_3	176-177
I-10	5-F	200-202
I-11	6-OCH_3	186-187
I-12	6,7-CH=CH-CH=CH-	245-246
I-13	6,8-Cl_2	180-182
I-14	7-NO_2	216-218
I-15	7-CH_3	220-221
I-16	6-I	160-166
I-17	6,7-F_2	131-133
I-18	6,7,8-$(OCH_3)_3$	199-201
I-19	6,8-Br_2	207-210
I-20	8-OCH_3	226-228
I-21	8-CF_3	188-191

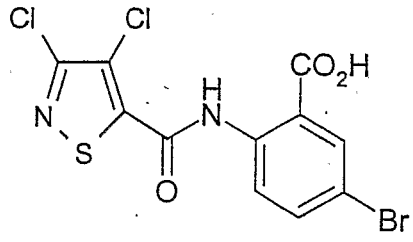
Table 1 (continued)

Compound No.	$(R)_n$	mp (°C) or n_D^{20}
I-22	7-F	
I-23	6-F	
I-24	6,8-$(CH_3)_2$	
I-25	6-NO_2	
I-26	6,7-$(OCH_3)_2$	
I-27	7,8-Cl_2	
I-28	5,8-Cl_2	
I-29	5,6,7,8-F_4	
I-30	6-CH_3,8-Br	
I-31	7-COOH	
I-32	6,8-I_2	
I-33	5-F, 6,8-Br_2	
I-34	5-COOH	
I-35	6-$NHCOCH_3$	
I-36	6,7-Br_2	
I-37	8-NO_2	
I-38	5,6,7,8-Cl_4	
I-39	5-Cl, 6,8-Br_2	
I-40	5-CF_3	
I-41	5-OCH_3	
I-42	6-$SO_2N(CH_3)_2$	
I-43	6-OCF_3	
I-44	6,7-$(CH_2)_4$-	
I-45	6,8-$(CH_3)_2$	
I-46	7-$NHCO_2C_2H_5$	

Table 1 (continued)

Compound No.	$(R)_n$	mp (°C) or n_D^{20}
I-47		
I-48		
I-49	5,8-$(CH_3)_2$	
I-50	7-CF_3	
I-51	8-CF_3	
I-52	5,6-CH=CH-CH=CH-	
I-53	6-SCH_3	
I-54	6,7-OCF_2O-	
I-55	6,8-F_2	
I-56	7-SO_2CH_3	
I-57	7-OCH_3	

Synthesis Example 2 (Intermediate)



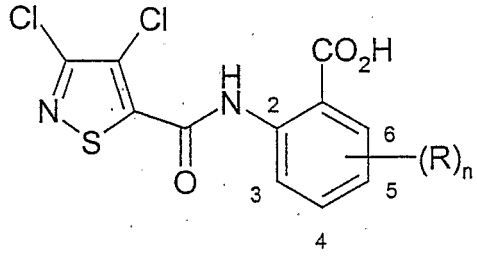
(Compound No. II - 7)

2.0 g of 3,4-dichloro-isothiazole-5-carboxylic acid chloride were added dropwise to a solution of 0.62 g of sodium hydroxide and 2.0 g of 2-amino-5-bromo-benzoic acid in 20 ml of water whilst stirring at room temperature. After stirring the reaction mixture for one more hour at room temperature, the precipitating crystals were filtered off and washed with water and then with hexane to obtain 2.37 g of 2-(3,4-dichloro-isothiazol-5-yl-carbonylamino)-5-bromo-benzoic acid.

m p >250°C

The compounds shown in the following Table 2 were also prepared according to the method described in Synthesis Example 2.

Table 2



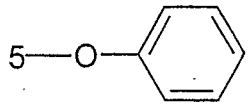
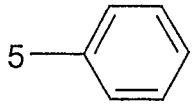
(II)

Compound No.	$(R)_n$	mp (°C) or n_D^{20}
II-1	-	>250
II-2	3-CH_3	209-213
II-3	3-Cl	195-198
II-4	4-Cl	>250
II-5	5-Cl	238-246
II-6	6-Cl	196-201
II-7	5-Br	>250
II-8	5-CH_3	229-237
II-9	6-CH_3	200-205
II-10	6-F	195-198
II-11	5-OCH_3	220-229
II-12	4,5-CH=CH-CH=CH-	>250
II-14	4-NO_2	245-248
II-13	3,5-Cl_2	
II-15	4-CH_3	234-236
II-16	5-I	232-240
II-17	4,5-F_2	207-210
II-18	3,4,5-$(OCH_3)_3$	166-169

Table 2 (continued)

Compound No.	$(R)_n$	mp (°C) or n_D^{20}
II-19	3,5-Br_2	
II-20	3-OCH_3	183-189
II-21	3-CF_3	
II-22	4-F	>250
II-23	5-F	>250
II-24	3,5-$(CH_3)_2$	
II-25	5-NO_2	
II-26	4,5-$(OCH_3)_2$	>250
II-27	3,4-Cl_2	
II-28	3,6-Cl_2	
II-29	3,4,5,6-F_4	
II-30	3- Br, 5-CH_3	
II-31	4-COOH	>250
II-32	3,5-I_2	
II-33	3,5-Br_2, 6-F,	
II-34	6-COOH	
II-35	5-$NHCOCH_3$	
II-36	4,5-Br_2	
II-37	3-NO_2	
II-38	3,4,5,6-Cl_4	
II-39	6-Cl, 3,5-Br_2	
II-40	6-CF_3	
II-41	6-OCH_3	
II-42	5-$SO_2N(CH_3)_2$	
II-43	5-OCF_3	
II-44	4,5-$(CH_2)_4$-	

Table 2 (continued)

Compound No.	$(R)_n$	mp (°C) or n_D^{20}
II-45	3,5-$(CH_3)_2$	
II-46	4-$NHCO_2C_2H_5$	
II-47		
II-48		
II-49	3,6-$(CH_3)_2$	
II-50	4-CF_3	
II-51	3-CF_3	
II-52	5,6-CH=CH-CH=CH-	
II-53	5-SCH_3	
II-54	4,5-OCF_2O-	
II-55	3,5-F_2	
II-56	4-SO_2CH_3	
II-57	4-OCH_3	

Biological Test Examples

Test Example A

Test of foliar spray effect against Pyricularia oryzae

Preparation of formulations of the compounds tested

Active compound: 30 - 40 parts by weight
Carrier: mixture of diatomaceous earth and kaolin (1:5), 55-65 parts by weight
Emulsifier: polyoxyethylene alkyl phenyl ether, 5 parts by weight

The above-mentioned amounts of active compound, carrier and emulsifier were crushed and mixed to make a wettable powder. A portion of the wettable powder comprising the prescribed amount of active compound was diluted with water and used for testing.

Testing procedure

Seedlings of paddy rice (variety: Kusabue) were cultured in plastic pots each having a diameter of 6 cm. The previously prepared solution of the prescribed concentration of active compound was sprayed over the seedlings in the 1.5 - 2 leaf stage, at a rate of 20 ml per 3 pots. 5 days after the application, a suspension of spores of artificially cultured Pyricularia oryzae was sprayed on the test plants once for inoculation, and the plants were kept at 25°C and 100% relative humidity for infection. 7 days after the inoculation, the infection rate per pot was classified and evaluated according to the following standard and the control value (%) was calculated. Phytotoxicity was tested at the same time. This test is an average of the results of 3 pots for 1 section. The evaluation of the infection rate and the calculation method of the control value are identical in each of the Test Examples A and B.

Infection rate	Percentage of lesion area in (%)
0	0
0.5	less than 2
1	2-less than 5
2	5-less than 10
3	10-less than 20
4	20-less than 40
5	more than 40

$$\text{Control value (\%)} = \left(1 - \frac{\text{Infection rate of treated section}}{\text{Infection rate of untreated section}} \right) \times 100$$

Test results

Compounds No. I-4, I-5, I-6, I-7, I-8, I-9, I-10, I-11, I-13, I-14, I-15, I-17, II-1, II-4, II-5, II-6, II-7, II-8, II-9, II-10, II-11, II-12, II-14, II-15, II-16, II-17, II-18, II-20, II-22, II-23, II-26 and II-32 showed control values of more than 80% at an active compound concentration of 500 ppm. No phytotoxicity was observed.

Test Example B

Test for the effect of seed treatment against Pyricularia oryzae.

Testing procedure

Seeds of paddy rice (variety: Kasabue) were drenched in a previously prepared diluted solution of an active compound having the prescribed concentration. 5 ml of such solution were used per 150 grains of seed. Drenching was conducted at a temperature of 20°C for 5 days. After the drenching, the air-dried seeds were sown in 2 plastic pots, each having a diameter of 9 cm, and the seeds were germinated by

placing the pots in a warmed nursery box (32°C) for 3 days. After cultivating the seedlings for 2 weeks, the plants reached the 2 - 2.5 leaf stage. A spore suspension of artificially cultured Pyricularia oryzae was then sprayed on the test plants once, and the plants were kept at a temperature of 25°C and a relative atmospheric humidity of 100% for infection. Seven days after the inoculation, the infection rate per pot was classified and evaluated and the control value (%) was calculated. Phytotoxicity was tested at the same time.

This test is an average of the results of 3 pots per one section.

Test results

Compounds No. I-1, I-4, I-5, I-6, I-8, I-9, I-11, I-12, I-13, I-15, I-16, I-21, II-1, II-3, II-4, II-5, II-6, II-10, II-16, II-17, II-26 and II-32 showed control values of more than 80% an active compound concentration of 500 ppm. No. phytotoxicity was observed.

Formulation Examples

Formulation Example I (Granules)

25 parts by weight of water were added to a mixture of 10 parts by weight of Compound No. I-1 according to the invention, 30 parts by weight of bentonite (montmorillonite), 58 parts by weight of talc and 2 parts by weight of lignin sulphonic acid salt, and the mixture was kneaded thoroughly. The resulting product was granulated by means of an extrusion granulator to form granules having a size of from 10 to 40 meshes. The granules were dried at a temperature between 40 and 50°C.

Formulation Example II (Granules)

95 parts by weight of a clay mineral having a particle size distribution within a range of from 0.2 to 2 mm were introduced into a rotary mixer. This product was uniformly wetted by spraying thereto under rotation a mixture of 5 parts by weight of Compound No. I-4 according to the invention and a liquid diluent. The granules obtained in this manner were dried at a temperature between 40 and 50°C.

Formulation Example III (Emulsifiable Concentrate)

An emulsifiable concentrate was prepared by mixing 30 parts by weight of Compound No. I-6 according to the invention, 5 parts by weight of xylene, 8 parts by weight of polyoxyethylene alkyl phenyl ether and 7 parts by weight of calcium alkylbenzene sulphonate with stirring.

Formulation Example IV (Wettable Powder)

A wettable powder was prepared by thoroughly mixing 15 parts by weight of Compound No. I-8 according to the invention, 80 parts by weight of a mixture (1:5) of White Carbon (fine powder of hydrated non-crystalline silicon oxide) and powdery clay, 2 parts by weight of sodium alkylbenzene sulphonate and 3 parts by weight of a

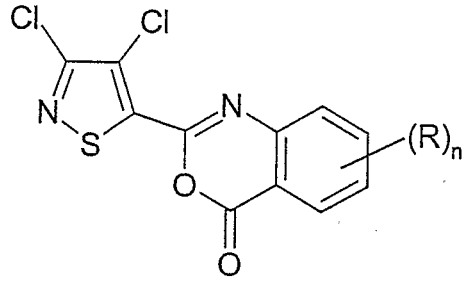
condensate of sodium alkylnaphthalene sulphonate and formaldehyde in powdery state.

Formulation Example V (Water Dispersible Granules)

20 parts by weight of Compound No. I-15 according to the invention, 30 parts by weight of sodium lignin sulphonate, 15 parts by weight of bentonite and 35 parts by weight of calcined diatomaceous earth powder were thoroughly mixed with water. The resulting product was granulated by means of extrusion through a 0.3 mm screen. After drying the product, water dispersible granules were obtained.

Patents Claims

1. Isothiazolyl-benzoxazine derivatives of the formula



(I)

wherein

R represents halogen, alkyl, alkoxy, alkylthio, alkylsulfonyl, haloalkyl, haloalkoxy, carboxy, N,N-dialkyl-sulfamoyl, acylamino, alkoxy-carbonylamino, phenyl, phenoxy or nitro, or two adjacent radicals may together form a group selected from alkylene, alkenylene, alkylene-dioxy or haloalkylenedioxy and

n represents 0, 1, 2, 3 or 4.

2. Isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1, in which

R represents fluoro, chloro, bromo, iodo, alkyl having 1 to 6 carbon atoms, alkoxy having 1 to 6 carbon atoms, alkylthio having 1 to 6 carbon atoms, alkylsulfonyl having 1 to 6 carbon atoms, haloalkyl having 1 to 6 carbon atom and 1 to 9 identical or different halogen atoms, haloalkoxy having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, carboxy, N,N-dialkyl-sulfamoyl having 1 to 6 carbon atoms in each of the alkyl groups, alkylcarbonylamino having 1 to 6 carbon atoms in the alkyl part, alkoxycarbonylamino having 1 to 6 carbon atoms in the alkoxy part, phenyl, phenoxy or nitro,

or two adjacent radicals together form a group selected from alkylene having 3 to 6 carbon atoms alkenylene having 4 to 6 carbon atoms, alkylenedioxy having 1 or 2 carbon atoms and 2 non-adjacent oxygen atoms, or haloalkylenedioxy having 1 to 4 carbon atoms and 2 non-adjacent oxygen atoms and 1 to 5 identical or different halogen atoms, and

n is 0, 1, 2, 3 or 4.

3. Isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1, in which

R represents fluoro, chloro, bromo, iodo, alkyl having 1 to 4 carbon atoms, alkoxy having 1 to 4 carbon atoms, alkylthio having 1 to 4 carbon atoms, alkylsulfonyl having 1 to 4 carbon atoms, haloalkyl having 1 to 4 carbon atoms and 1 to 5 identical or different halogen atoms, haloalkoxy having 1 to 4 carbon atoms and 1 to 5 identical or different halogen atoms, carboxy, N,N-dialkyl-sulfamoyl having 1 to 4 carbon atoms in each of the alkyl groups, alkylcarbonylamino having 1 to 4 carbon atoms in the alkyl part, alkoxycarbonylamino having 1 to 4 carbon atoms in the alkoxy part, phenyl, phenoxy or nitro,

or two adjacent radicals together form a group selected from $-(CH_2)_3-$, $-(CH_2)_4-$, $-CH=CH-CH=CH-$, $-O-CH_2-O-$, $-O-CH_2-CH_2-O-$, $-O-CF_2-O-$ and $-O-CF_2-CF_2-O-$, and

n is 0, 1, 2, 3 or 4.

4. Isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1, in which

R represents fluoro, chloro, bromo, iodo, methyl, ethyl, methoxy, ethoxy, methylthio, methylsulfonyl, trifluoromethyl, trichloromethyl, difluoromethyl, trifluoromethoxy, difluoromethoxy, carboxy, N,N-dimethyl-sulfamoyl, methylcarbonylamino, ethylcarbonylamino, methoxycarbonylamino, ethoxycarbonylamino, phenyl, phenoxy or nitro, or

two adjacent radicals represent a group selected from $-(CH_2)_3-$, $-(CH_2)_4-$, $-CH=CH-CH=CH-$, $-O-CH_2-O-$, $-O-CH_2-CH_2-O-$, $-O-CF_2-O-$ and $-O-CF_2-CF_2-O-$, and

5. Isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1, in which

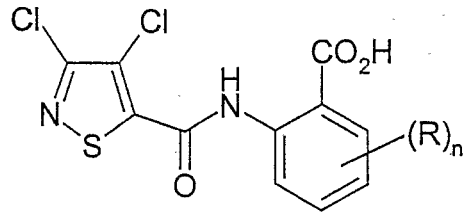
R represents fluoro, chloro, bromo, iodo, methyl, trifluoromethyl or nitro, or

two adjacent radicals represent the group -CH=CH-CH=CH-, and

n represents 0, 1 or 2,
wherein the radicals may be identical or different, if n is 2.

6. Process for the preparation of isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1, characterized in that

a) isothiazolyl-carboxamides of the formula



(II)

wherein

R and n have the meanings mentioned in claim 1

are reacted with dehydrating agents in the presence of an inert diluent.

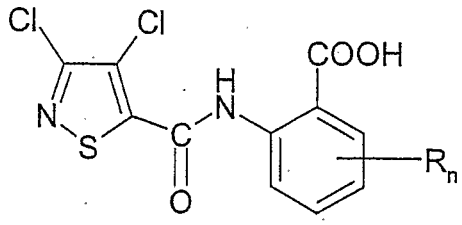
7. Microbicidal compositions, characterized in that they contain at least one isothiazolyl-benzoxazine derivative of the formula (I) according to claim 1 plus extenders and/or surface-active agents.

8. Process for combating undesired microorganisms, characterized in that isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1 are applied to the microorganisms and/or to their habitat.

9. Use of isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1 for combating undesired microorganisms.

10. Process for the preparation of microbicidal compositions, characterized in that isothiazolyl-benzoxazine derivatives of the formula (I) according to claim 1 are mixed with extenders and/or surface active agents.

11. Isothiazolyl-carboxamides of the formula



(II)

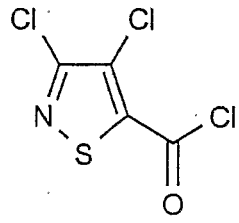
wherein

R represents halogen, alkyl, alkoxy, alkylthio, alkylsulfonyl, haloalkyl, haloalkoxy, carboxy, N,N-dialkyl-sulfamoyl, acylamino, alkoxy-carbonylamino, phenyl, phenoxy, or nitro, or two adjacent radicals may together form a group selected from alkylene, alkenylene, alkylenedioxy or haloalkylenedioxy, and

n represents 0, 1, 2, 3 or 4.

12. Process for the preparation of isothiazolyl-carboxamides of the formula (II), characterized in that

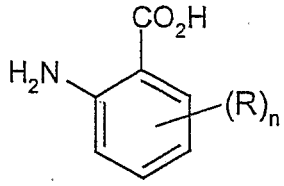
b) 3,4-dichloro-isothiazole-5-carboxylic acid chloride of the formula



(III)

is reacted

with benzoic acid derivatives of the formula



(IV)

wherein

R and n have the meanings mentioned in claim 11,

in the presence of a diluent and in the presence of an acid binding agent.

INTERNATIONAL SEARCH REPORT

PCT/EP 03/12475

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C07D417/04 A01N43/86

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C07D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

PAJ, EPO-Internal, WPI Data, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 987 257 A (NIHON NOHYAKU CO LTD) 22 March 2000 (2000-03-22) Abstract; claims 1-7; page 49, paragraph '0063!. ---	1-12
A	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 22, 9 March 2001 (2001-03-09) & JP 2001 139566 A (NIPPON NOHYAKU CO LTD), 22 May 2001 (2001-05-22) cited in the application abstract --- -/--	1-12

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
2 March 2004	18/03/2004

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Weisbrod, T

INTERNATIONAL SEARCH REPORT

PCT/EP 03/12475

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
Y	PATENT ABSTRACTS OF JAPAN vol. 2000, no. 16, 8 May 2001 (2001-05-08) & JP 2001 019691 A (NISSAN CHEM IND LTD), 23 January 2001 (2001-01-23) cited in the application abstract	1-12

INTERNATIONAL SEARCH REPORT

PCT/EP 03/12475

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. [X] Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

Although claims 8 and 9 embrace a method of treatment of the human/animal body, the search has been carried out and based on the alleged effects of the compounds.

2. [ ] Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:

3. [ ] Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. [ ] As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. [ ] As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. [ ] As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. [ ] No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

[ ] The additional search fees were accompanied by the applicant's protest.

[ ] No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

Information on patent family members

PCT/EP 03/12475

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0987257 A	22-03-2000	AU 731921 B2	05-04-2001
		AU 7453098 A	30-12-1998
		CA 2289512 A1	03-12-1998
		EP 0987257 A1	22-03-2000
		US 6348460 B1	19-02-2002
		CN 1258288 T	28-06-2000
		WO 9854163 A1	03-12-1998
		JP 11043488 A	16-02-1999
		JP 11071360 A	16-03-1999
JP 2001139566 A	22-05-2001	NONE	
JP 2001019691 A	23-01-2001	NONE	