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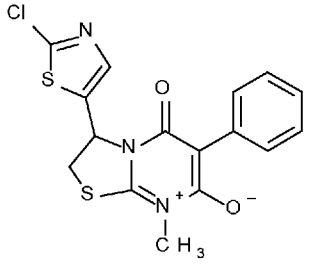
(54) Title: METHOD OF CONTROLLING RICE PESTS IN RICE

(57) Abstract: The present invention relates to methods for controlling rice pest invertebrates, which methods comprise applying pyrimidinium compounds of formula (I), the stereoisomers, salts, tautomers and N-oxides thereof, their mixtures and compositions comprising such compounds or mixtures, by seedling box application.



(I)

WO 2019/043183 A1

Method of controlling rice pests in rice

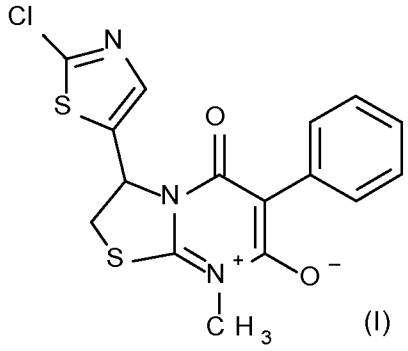
Rice (Oryza species, especially Oryza sativa) is an important basic food in the world. It is a staple food in Asia and is an important part of many cultures. Rice is therefore an important crop and is cultivated in large areas, especially in Asia.

Invertebrate pests and, in particular, insects, arthropods and nematodes cause significant damage to growing and harvested rice crop, thereby causing large economic loss to the food supply and to property. While a large number of pesticidal agents are known, due to the ability of target pests to develop resistance to said agents, there is an ongoing need for new agents for combating invertebrate pests such as insects, arachnids and nematodes. Further, rice cultivation requires special pesticides suitable for the farming methods used in rice, e.g. use of nursery boxes, paddy fields, aquatic environment and so on. Pesticides suitable for the use in rice must also be tolerated well by the rice plants. They must also be tolerated well by the environment of the rice plants, e.g. from an ecotoxicological point of view, i.e. they must not harm beneficial organisms. Further, they must survive the conditions in which they are applied, to ensure efficacy. This is especially a challenge in the aquatic environment and the high temperature climate conditions, in which rice is usually grown. On the other hand, they must degrade within a reasonable period of time so that they do not have any negative impact on the environment. They must not impact the health of the farmer and the consumer. They should not be present in the rice product later on (low or no residues), to ensure safety of the human beings consuming the rice. Furthermore, many pests have developed resistance against pesticides commonly used in rice. Therefore, pesticides suitable for the use in rice should be effective against those pests have developed resistance to other pesticides. Not all pesticides are able fulfil the requirements to be used in these conditions.

It is therefore an object of the present invention to provide compounds having a good pesticidal activity and showing a good activity spectrum against a large number of different invertebrate pests occurring in rice, especially against insects, arachnids and nematodes that are difficult to control, while still showing a good regulatory profile.

It has been found that these objectives can be achieved by S-containing pyrimidinium compounds of the general formula (I), as defined below, including their stereoisomers, their salts, in particular their agriculturally or veterinary acceptable salts, their tautomers and their N-oxides.

In a first aspect, the invention relates to a seedling box application method of controlling rice pest invertebrates in rice, which method comprises applying to said rice pest invertebrates compound of formula (I)

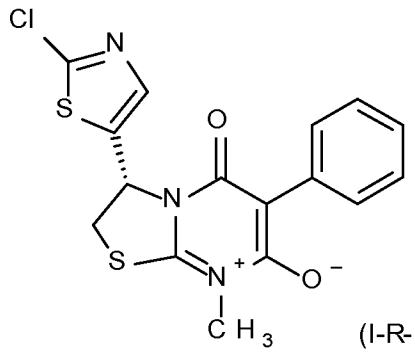


(I)

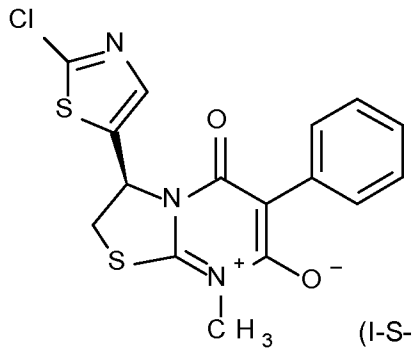
In another aspect, the invention relates to a method of controlling rice pest invertebrates in rice, which method comprises applying to said rice pest invertebrates compound of formula (I) by seedling box application.

WO 2014/167084 and PCT/EP2018/057578 describe certain substituted pyrimidinium compounds with heterocyclic substituents for combating invertebrate pests.

The compound of formula (I) is present in two enantiomeric forms I-R-1 and I-S-1 as shown below



(I-R-1)



(I-S-1)

Moreover, the present invention relates to and includes the following embodiments:

- a compound of formula (I) for use in controlling rice pests, especially rice pest invertebrates, in rice, by seedling box application;
- compositions comprising compound of formula (I), for use in controlling rice pests, especially rice pest invertebrates, in rice, by seedling box application;
- agricultural compositions comprising an amount of compound of formula (I) or an enantiomer, diasteromer or salt thereof as defined above, for controlling rice pests, especially rice pest invertebrates, in rice, by seedling box application;
- a method for combating rice pest invertebrates, infestation, or infection by rice pest invertebrates, which method comprises applying said pest or its food supply, habitat or breeding grounds with a pesticidally effective amount of compound of formula (I) as defined above or a composition thereof, by seedling box application;
- a method for controlling rice pest invertebrates, infestation, or infection by invertebrate pests, which method comprises applying said pest or its food supply, habitat or breeding grounds with a pesticidally effective amount of of formula (I) as defined above or a composition comprising compound of formula (I), by seedling box application;
- a method for preventing or protecting against rice pest invertebrates comprising contacting the rice pest invertebrates, or their food supply, habitat or breeding grounds with compound of formula (I) as defined above or a composition comprising compound of formula (I) as defined above, or a composition comprising compound of formula (I), by seedling box application;
- a method for protecting rice, rice plants, rice plant propagation material and/or growing rice plants from attack or infestation by rice pest invertebrates comprising contacting or treating the rice, rice plants, rice plant propagation material and growing rice plants, or soil, material, surface, space, area or water in which the rice, rice plants, rice plant propagation material is stored or the rice plant is growing, with a pesticidally effective amount of compound of formula (I) as defined above or a composition comprising compound of formula (I), by seedling box application;

- a method for increasing the health of rice plants, especially in paddy rice fields, comprising the treatment with compound of formula (I), by seedling box application;
- a method for increasing the yield of rice plants, comprising the treatment with compound of formula (I);

Moreover, the present invention relates to and includes the following embodiments:

- a compound of formula (I) for use in seedling box application for controlling rice pests, especially rice pest invertebrates, in rice;
- seeding box comprising a compound of formula (I), for use in controlling rice pests, especially rice pest invertebrates, in rice;
- nursery box comprising a compound of formula (I), for use in controlling rice pests, especially rice pest invertebrates, in rice;
- seeding box application method comprising agricultural compositions comprising an amount of a compound of formula (I) or an enantiomer, diasteromer or salt thereof as defined above, for use in controlling rice pests, especially rice pest invertebrates, in rice;
- a seeding box application method for combating rice pest invertebrates, infestation, or infection by rice pest invertebrates, which method comprises contacting said pest or its food supply, habitat or breeding grounds with a pesticidally effective amount of a compound of formula (I) as defined above, or a composition thereof;
- a seeding box application method for controlling rice pest invertebrates, infestation, or infection by invertebrate pests, which method comprises contacting said pest or its food supply, habitat or breeding grounds with a pesticidally effective amount of a compound of formula (I) as defined above, or a composition comprising compound of formula (I);
- a seeding box application method for preventing or protecting against rice pest invertebrates comprising contacting the rice pest invertebrates, or their food supply, habitat or breeding grounds with a compound of the formula (I) as defined above or a composition comprising a compound of formula (I) as defined above, or a composition comprising a compound of formula (I);
- a seeding box application method for protecting rice, rice plants, rice plant propagation material and/or growing rice plants from attack or infestation by rice pest invertebrates comprising contacting or treating the rice, rice plants, rice plant propagation material and growing rice plants, or soil, material, surface, space, area or water in which the rice, rice plants, rice plant propagation material is stored or the rice plant is growing, with a pesticidally effective amount of a compound of formula (I) as defined above, or a composition comprising compound of formula (I);
- a seeding box application method for increasing the health of rice plants, especially in paddy rice fields, comprising the treatment with a compound of formula (I);
- a seeding box application method for increasing the yield of rice plants, comprising the treatment with a compound of formula (I);

The present invention relates to nursery boxes comprising a growth substrate for rice, rice seeds and an aqueous formulation of a compound of formula (I) for treating said rice seeds to protect rice plants against insects for a prolonged period of time. The invention further relates to a new process for the treatment of rice crops using an insecticidal product containing a compound of formula (I), and more especially a new process for the treatment of rice crops against

parasites called stem borers (Chilo spp.) and plant hoppers (Nilparvata lugens), as well as other parasites such as weevils (Lissorhoptrus oryzophilus). Other parasites are advantageously eliminated by the process according to the invention, especially the nematode Aphelencoides besseyi, and the mining fly Hydrellia philiippina.

Rice crops are attacked by a number of diseases, especially attacks from insects such as those mentioned above. In the case of paddy rice, there is a very particular difficulty in eliminating these insects since the possible treatment products tend to pollute the water in the paddy fields. Research is thus aimed at both treating rice plants effectively and vigorously against parasites and, at the same time, reducing water pollution to the bare minimum. The difficulty in solving this problem is particularly great because the two demands conflict with each other: if the number of treatments is reduced in order to reduce pollution, the quality of the protection is reduced. If the number of treatments is increased in order to increase the quality of the protection, pollution is also increased. There is thus no obvious solution to the problem to be solved, since the only possible solutions conflict with each other.

The problem of avoiding water pollution, in the case of rice cultivation, is all the more difficult since water for rice cultivation comprises various species, including useful insects or aquatic fauna, which are beneficial and profitable. It is necessary to destroy the harmful species without harming these profitable species.

Although insecticides available on the market and in the literature are very numerous, it so happens that there is virtually no satisfactory solution to the abovementioned problem.

A new process has now been found for growing rice seedlings according to the irrigated rice method which makes it possible to solve all or part of the abovementioned problems.

According to another aspect of the invention, the latter relates to a process for treating rice plants or rice propagation material against insects with a view to obtaining a sustained and effective protection of the plant after the sowing period, characterised in that there is applied to the said plants or, preferably, to their propagation material, an effective amount of compound of formula I. More preferentially still, the propagation material used is the rice seed or, in other words, rice grains. Advantageously, the insecticidal material used is that which will be defined below.

Moreover, the present invention relates to and includes the following embodiments:

- seeding box comprising the compound of formula (I), and rice seed;
- seeding box comprising agricultural compositions comprising an amount of a compound of formula (I) or an enantiomer, diasteromer, or salt thereof as defined above;
- seeding box comprising the compound of formula (I), wherein the amount of the compound of formula (I) impregnated into the rice seeds is between about 3 and 200 g/q, preferably between about 3 and 100 g/q, more preferably between about 6 and 25 g/q.

All the compounds of the present invention including their stereoisomers, their tautomers, their salts or their N-oxides as well as compositions thereof are particularly useful for controlling invertebrate rice pests, in particular for controlling arthropods and nematodes and especially insects. Therefore, the invention relates to the use of a compound as disclosed in the present invention, for combating or controlling invertebrate rice pests, in particular invertebrate pests of the group of insects, arachnids or nematodes by seedling box application.

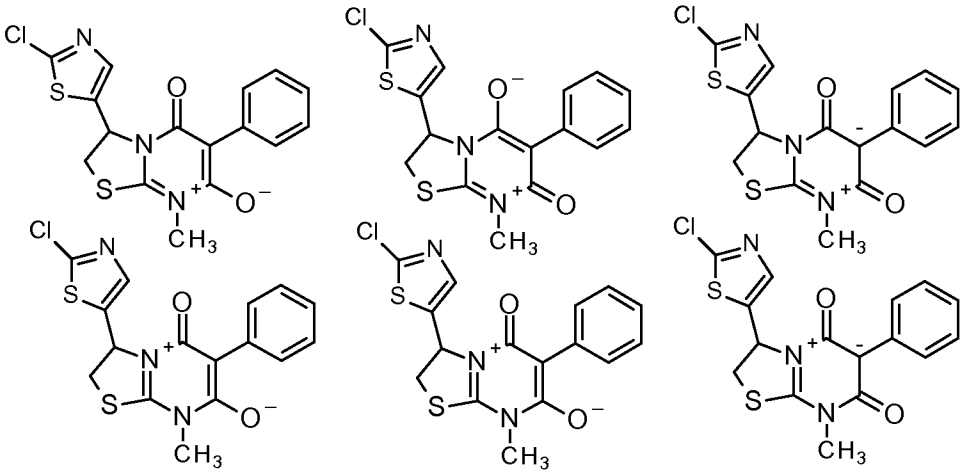
The term “Seedling box applications” refers to manual or mechanical incorporation of insecticide formulations (for eg. granules, liquid) in nursery boxes or seedling boxes containing rice seedlings before being transplanted into the main field.

The term "compound(s) according to the invention" or "compound(s) of formula (I)" or “compound I or (I)” as used in the present invention refers to and comprises the compound(s) as defined herein and/or stereoisomer(s), salt(s), tautomer(s) or N-oxide(s) thereof. The term "compound(s) of the present invention" is to be understood as equivalent to the term "compound(s) according to the invention", therefore also comprising stereoisomer(s), salt(s), tautomer(s) or N-oxide(s) of compounds of formula (I).

The term “method of invention” or method according to the invention” referes to the Seedling box applications method as described herein.

The term "composition(s) according to the invention" or "composition(s) of the present invention" encompasses composition(s) comprising compound of formula (I) according to the invention as defined above, therefore also including a stereoisomer, an agriculturally or veterinary acceptable salt, tautomer or an N-oxide of the compounds of formula (I).

The compounds of formula (I) are present in mesomeric forms. These forms may be expressed in different isoelectronic formulae, each having the formal positive and negative charges on different atoms (as shown below). The present invention extends to all representative isoelectronic structures of compounds of formula I.



The compounds of formula I-R-1 and I-R-S are also present in mesomeric forms analogous to the compound of formula I as shown above.

The compounds of formula (I) are present as mixtures of enantiomers or diastereomers. The invention provides both the single pure enantiomers or pure diastereomers of the compounds of formula (I), and their mixtures and the use according to the invention of the pure enantiomers or pure diastereomers of the compound of formula (I) or its mixtures.

The term "stereoisomer(s)" encompasses both optical isomers, such as enantiomers or diastereomers, the latter existing due to more than one center of chirality in the molecule, as well as geometrical isomers (cis/trans isomers). The present invention relates to every possible stereoisomer of the compounds of formula (I), i.e. to single enantiomers or diastereomers, as well as to mixtures thereof. Preferred embodiments of specific enantiomers are described in more detail below, as compounds of formula (I-R-1) and (I-S-1).

Salts of the compounds of the formula (I) are preferably agriculturally and/or veterinary acceptable salts. They can be formed in a customary method, e.g. by reacting the compound with an acid of the anion in question if the compound of formula (I) has a basic functionality or by reacting an acidic compound of formula (I) with a suitable base.

Suitable agriculturally or veterinary useful salts are especially the salts of those cations or the acid addition salts of those acids whose cations and anions, respectively, do not have any adverse effect on the action of the compounds according to the present invention. Suitable cations are in particular the ions of the alkali metals, preferably lithium, sodium and potassium, of the alkaline earth metals, preferably calcium, magnesium and barium, and of the transition metals, preferably manganese, copper, zinc and iron, and also ammonium (NH_4^+) and substituted ammonium in which one to four of the hydrogen atoms are replaced by C_1-C_4-alkyl, C_1-C_4-hydroxyalkyl, C_1-C_4-alkoxy, C_1-C_4-alkoxy-C_1-C_4-alkyl, hydroxy-C_1-C_4-alkoxy-C_1-C_4-alkyl, phenyl or benzyl. Examples of substituted ammonium ions comprise methylammonium, isopropylammonium, dimethylammonium, diisopropylammonium, trimethylammonium, tetramethylammonium, tetraethylammonium, tetrabutylammonium, 2-hydroxyethylammonium, 2-(2-hydroxyethoxy)ethyl-ammonium, bis(2-hydroxyethyl)ammonium, benzyltrimethylammonium and benzyltriethylammonium, furthermore phosphonium ions, sulfonium ions, preferably tri(C_1-C_4-alkyl)sulfonium, and sulfoxonium ions, preferably tri(C_1-C_4-alkyl)sulfoxonium.

Anions of useful acid addition salts are primarily chloride, bromide, fluoride, hydrogen sulfate, sulfate, dihydrogen phosphate, hydrogen phosphate, phosphate, nitrate, hydrogen carbonate, carbonate, hexafluorosilicate, hexafluorophosphate, benzoate, and the anions of C_1-C_4-alkanoic acids, preferably formate, acetate, propionate, and butyrate. They can be formed by reacting the compounds of formula I with an acid of the corresponding anion, preferably of hydrochloric acid, hydrobromic acid, sulfuric acid, phosphoric acid, or nitric acid.

The term “N-oxide” includes any compound of the present invention which has at least one tertiary nitrogen atom that is oxidized to an N-oxide moiety.

The compounds of formula (I) can be prepared as described in WO2014/167084 and EP17164175.6. The preparation of the compounds of formula (I) above may lead to them being obtained as isomer mixtures. If desired, these can be resolved by the methods customary for this purpose, such as crystallization or chromatography, also on optically active adsorbate, to give the pure isomers.

Agronomically acceptable salts of the compounds I can be formed in a customary manner, e.g. by reaction with an acid of the anion in question.

Preferences

In a particular aspect, the invention relates to a seedling box application method of controlling rice pest invertebrates in rice as described herein, wherein the compound of formula (I) is the compound I-R-1, salt, or N-oxide thereof.

In a particular aspect, the invention relates to a method of controlling rice pest invertebrates in rice by seedling box application as described herein, wherein the compound of formula (I) is the compound I-R-1, salt, or N-oxide thereof.

In another embodiment of the invention, the compound of formula (I) is the compound I-S-1.

Rice pests

In the context of this invention, rice pest invertebrates are animal pests, which occur in rice. The rice pest invertebrates include insects, acarids, and nematodes, preferably insects. Rice pest invertebrates, which are well-known in rice, include but are not limited to the following species:

Hemiptera:

brown planthopper – Nilaparvata lugens

small brown planthopper – Laodelphax striatellus

white-backed planthopper – Sogatella furcifera

white leafhopper – Cofana spectra

green leafhopper – Nephotettix virescens, N. nigriceps, N. cincticeps, N. malayanus

zig zag leafhopper – Recilia dorsalis

maize orange leafhopper – Cicadulina bipunctata

aster leafhopper - Macrosteles fascifrons

rice earhead bug, Leptocorisa oratorius, L. acuta

rice stink bugs – Nezara viridula, Pygomenida varipennis, Eysarcoris, Tibraca limbatriventris, Eysarcoris ventralis

small stink bug - Oebalus poecilus, O. pugnax

coreid bug – Eysarcoris sp

chinch bug - Blissus leucopterus leucopterus

rice mealybug, Brevennia rehi, Pseudococcus saccharicola

rice aphids, Rhopalosiphum rufiabdominalis, Macrosiphum avenae, Hysteroneura setariae, Tetraneuro nigriabdominalis

bean root aphid - Smynthurodes betae

Lepidoptera:

rice skipper – Parnara guttata, Melanitis leda ismene

rice stem borer / striped stem borer – Chilo suppressalis, Chilo polychrusus, Chilo partellus, Chilo plejadellus

rice stalk borer – Chilotraea polychrysa

pink rice borer – Sesamia inferens

yellow rice borer – Tryporyza (=Scirpophaga) incertulas

white rice borer – Tryporyza innotata

rice leafroller / leaf folder – Cnaphalocrocis medinalis, Marasmia patnalis, M. exigua

rice ear-cutting caterpillar / armyworm– Pseudaletia separata

green caterpillar – Xanthodes transversa

green rice caterpillar – Narnaga aenescens

green horned caterpillars - Melanitis leda ismene, Mycalesis sp

fall army worm – Spodoptera frugiperda

cutworm – Mythimna separata

rice case worm- Nymphula depunctalis

black hairy caterpillar, Amata sp.

hairy caterpillar- Mocis frugalis

yellow caterpillar, Psalis pennatula

rice semi-brown looper, Mocis frugalis
rice semi-looper, Chrysodeixis chalcites
grass webworm - Herpetogramma licarsisalis
sugarcane borer - Diatraea saccharalis
corn stalk borer – Elasmopalpus lignosellus
striped grass looper – Mocis latipes
european corn borer – Ostrinia nubilalis
Mexican rice borer – Eoreuma loftini
Coleoptera:
water weevil – Lissorhopterus oryzophilus
rice plant weevil – Echinocnemus squamous
rice weevil - Oryzophagus oryzae
rice hispa – Diclodispa armigera
rice leaf beetle – Oulema oryzae
rice blackbug – Scotinophora vermidulate, S. vermidulate, S. lurida, S. latiuscula
rice flea beetle – Chaetocnima basalis
grubs - Leucopholis irrorata, Leucopholis irrorata, Phyllophaga sp, Heteronychus sp
scarab beetle (bicho torito) - Diloboderus abderus
billbugs - Sphenophorus spp
grape colaspis - Colaspis brunnea, C. louisianae
rice pollen beetle, Chilolaba acuta
Diptera:
stem maggot – Chlorops oryzae
leafminer – Agromyza oryzae
rice whorl maggot / rice stem maggot – Hydrellia sasakii
rice whorl maggot / small rice leafminer – Hydrellia griseola
rice gall midge – Orseolia (=Pachydiplosis) oryzae
rice shoot fly- Atherigona oryzae
rice seed midge – Chironomus cavazzai, Chironomus spp, Cricotopus spp
Thysanoptera:
rice thrips- Chloethrips oryzae, Stenochaetothrips biformis, Perrisothrips sp., Hoplothrips sp.,
Orthoptera:
rice grasshoppers, Hieroglyphus banian, Hieroglyphus nigrorepletus, Catantops pinguis, Attractomorpha burri, A. crenulate, A. psittacina psittacina, A. Bedeli, Oxya adenttata, Oxya ebneri, Oxya hyla intricata, Acrida turricata
locusts – Locusta migratoria manilensis
mole cricket, Grylotalpa africana
field cricket: Gryllus bimaculatus, Teleogryllus occipitalis, Euscyrtus concinus
katydid – Conocephalus longipennis

Isoptera:
termites – Macrotermes gilvus, Syntermes molestans
Hymenoptera:

ants – Solenopsis geminata
rice white tip nematode – Aphelenchoides besseyi
Acari:
rice panicle mite - Steotarsonemus pinki
Crustacea:
tadpole shrimp - Triops longicaudatus. T. cancriformis
rice crayfish - Procambarus clarkii, Orconectes virilis.

In addition, rice is affected by a range of bugs including Leptocorisa chinensis, Lagynotomus elongates, Nerzara viridula, Eysacoris parvus, Leptocorisa oratorius, Oebalus pugnax, Cletus trigonus, as well as a variety of mites, caterpillars, beetles, rootworms and maggots.

In one embodiment, the rice pest invertebrate is a biting/chewing insect.

In one embodiment, the rice pest invertebrate is a piercing/sucking insect.

In one embodiment, the rice pest invertebrate is a rasping insect.

In one embodiment, the rice pest invertebrate is a siphoning insect.

In one embodiment, the rice pest invertebrate is a sponging insect.

In one embodiment, the rice pest invertebrate is selected from brown planthopper (Nilaparvata lugens), small brown planthopper (Laodelphax striatellus), white-backed planthopper (Sogatella furcifera), rice stem borer / striped stem borer (Chilo suppressalis), yellow rice borer (Tryporyza (=Scirpophaga) incertulas), rice leafroller / leaf folder (Cnaphalocrocis medinalis), water weevil (Lissorhopterus oryzophilus).

In one embodiment, the rice pest invertebreate is from the order Hemiptera or Lepidoptera.

In one embodiment, the rice pest invertebrate is from the order Hemiptera. In a further embodiment, the rice pest invertebrate is a hopper, preferably selected from brown planthopper (Nilaparvata lugens), small brown planthopper (Laodelphax striatellus), white-backed planthopper (Sogatella furcifera), green leafhopper (Nephotettix virescens). In a further embodiment, the rice pest invertebrate is selected from brown planthopper (Nilaparvata lugens) and green leafhopper (Nephotettix virescens), preferably brown planthopper (Nilaparvata lugens).

In one embodiment, the rice pest invertebrate is the brown planthopper (Nilaparvata lugens).

In one embodiment, the rice pest invertebrate is the green leafhopper (Nephotettix virescens).

In a further embodiment, the rice pest invertebrate is a stink bug, preferably selected from rice stink bugs (Nezara viridula, Pygomenida varipennis, Eysarcoris, Tibraca limbatriventris, Eysarcoris ventralis) or small stink bug (Oebalus poecilus, O. pugnax).

In one embodiment, the rice pest invertebrate is from the order Lepidoptera. In a further embodiment, the rice pest invertebrate is a borer, preferably stem borer, preferably rice stem borer (Chilo suppressalis) or yellow rice borer (Tryporyza (=Scirpophaga) incertulas).

In a further embodiment, the rice pest invertebrate is the rice leafroller / leaf folder (Cnaphalocrocis medinalis, Marasmia patnalis, M. exigua).

In one embodiment, the rice pest invertebrate is from the order Coleoptera. In a further embodiment, the rice pest invertebrate is water weevil (Lissorhopterus oryzophilus). In a further embodiment, the rice pest invertebrate is rice weevil (Oryzophagus oryzae).

In one embodiment, the rice pest invertebrate is from the family of termites (order Isoptera).

In a preferred embodiment, the rice pest invertebrate is Lissorhoptrus oryzophilus.

The present invention also relates to methods according to the invention, applying a mixture of at least one compound of the present invention with at least one mixing partner as defined herein after. Preferred are binary mixtures of one compound of the present invention as component I with one mixing partner as defined herein after as component II. Preferred weight ratios for such binary mixtures are from 5000:1 to 1:5000, preferably from 1000:1 to 1:1000, more preferably from 100:1 to 1:100, particularly preferably from 10:1 to 1:10. In such binary mixtures, components I and II may be used in equal amounts, or an excess of component I, or an excess of component II may be used.

Mixing partners can be selected from pesticides, in particular insecticides, nematicides, and acaricides, fungicides, herbicides, plant growth regulators, fertilizers, and the like. Preferred mixing partners are insecticides, nematicides and fungicides.

The following list M of pesticides, grouped and numbered according the Mode of Action Classification of the Insecticide Resistance Action Committee (IRAC), together with which the compounds of the present invention can be used and with which potential synergistic effects might be produced, is intended to illustrate the possible combinations, but not to impose any limitation:

M.1 Acetylcholine esterase (AChE) inhibitors: M.1A carbamates, e.g. aldicarb, alanycarb, bendiocarb, benfuracarb, butocarboxim, butoxycarboxim, carbaryl, carbofuran, carbosulfan, ethiofencarb, fenobucarb, formetanate, furathiocarb, isoprocarb, methiocarb, methomyl, metolcarb, oxamyl, pirimicarb, propoxur, thiodicarb, thiofanox, trimethacarb, XMC, xylylcarb and triazamate; or M.1B organophosphates, e.g. acephate, azamethiphos, azinphos-ethyl, azinphosmethyl, cadusafos, chlorethoxyfos, chlorfenvinphos, chlormephos, chlorpyrifos, chlorpyrifos-methyl, coumaphos, cyanophos, demeton-S-methyl, diazinon, dichlorvos/ DDVP, dicrotophos, dimethoate, dimethylvinphos, disulfoton, EPN, ethion, ethoprophos, famphur, fenamiphos, fenitrothion, fenthion, fosthiazate, heptenophos, imicyafos, isofenphos, isopropyl O-(methoxyaminothiophosphoryl) salicylate, isoxathion, malathion, mecarbam, methamidophos, methidathion, mevinphos, monocrotophos, naled, omethoate, oxydemeton-methyl, parathion, parathion-methyl, phenthoate, phorate, phosalone, phosmet, phosphamidon, phoxim, pirimiphos- methyl, profenofos, propetamphos, prothiofos, pyraclofos, pyridaphenthion, quinalphos, sulfotep, tebupirimfos, temephos, terbufos, tetrachlorvinphos, thiometon, triazophos, trichlorfon, and vamidothion;

M.2. GABA-gated chloride channel antagonists: M.2A cyclodiene organochlorine compounds, e.g. endosulfan or chlordane; or M.2B fiproles (phenylpyrazoles), e.g. ethiprole, fipronil, flufiprole, pyrafluprole, and pyriprole;

M.3 Sodium channel modulators from the class of M.3A pyrethroids, e.g. acrinathrin, allethrin, d-cis-trans allethrin, d-trans allethrin, bifenthrin (in particular kappa-bifenthrin), bioallethrin, bioallethrin S-cylclopentenyl, bioresmethrin, cycloprothrin, cyfluthrin, beta-cyfluthrin, cyhalothrin, lambda-cyhalothrin, gamma-cyhalothrin, cypermethrin, alpha-cypermethrin, beta-cypermethrin, theta-cypermethrin, zeta-cypermethrin, cyphenothrin, deltamethrin, empenthrin, esfenvalerate, etofenprox, fenpropathrin, fenvalerate, flucythrinate, flumethrin, tau-fluvalinate, halfenprox, heptafluthrin, imiprothrin, meperfluthrin,metofluthrin, momfluorothrin (in particular epsilon-momfluorothrin), permethrin, phenothrin, prallethrin, profluthrin, pyrethrin (pyrethrum), resmethrin,

silafluofen, tefluthrin (in particular kappa-tefluthrin), tetramethylfluthrin, tetramethrin, tralomethrin, and transfluthrin; or M.3B sodium channel modulators such as DDT or methoxychlor;

M.4 Nicotinic acetylcholine receptor agonists (nAChR): M.4A neonicotinoids, e.g. acetamiprid, clothianidin, cycloxaprid, dinotefuran, imidacloprid, nitenpyram, thiacloprid and thiamethoxam; or the compounds M.4A.1 4,5-Dihydro-N-nitro-1-(2-oxiranylmethyl)-1H-imidazol-2-amine, M.4A.2: (2E-)-1-[(6-Chloropyridin-3-yl)methyl]-N'-nitro-2-pentylidenehydrazinecarboximidamide; or M4.A.3: 1-[(6-Chloropyridin-3-yl)methyl]-7-methyl-8-nitro-5-propoxy-1,2,3,5,6,7-hexahydroimidazo[1,2-a]pyridine; or M.4B nicotine; M.4C sulfoxaflor; M.4D flupyradifurone; M.4E triflumezopyrim;

M.5 Nicotinic acetylcholine receptor allosteric activators:spinosyns, e.g. spinosad or spinetoram;

M.6 Chloride channel activators from the class of avermectins and milbemycins, e.g. abamectin, emamectin benzoate, ivermectin, lepimectin, or milbemectin;

M.7 Juvenile hormone mimics, such as M.7A juvenile hormone analogues hydroprene, kinoprene, and methoprene; or M.7B fenoxycarb, or M.7C pyriproxyfen;

M.8 miscellaneous non-specific (multi-site) inhibitors, e.g. M.8A alkyl halides as methyl bromide and other alkyl halides, M.8B chloropicrin, M.8C sulfuryl fluoride, M.8D borax, or M.8E tartar emetic;

M.9 Chordotonal organ TRPV channel modulators, e.g. M.9B pymetrozine; pyrifluquinazon;

M.10 Mite growth inhibitors, e.g. M.10A clofentezine, hexythiazox, and diflovidazin, or M.10B etoxazole;

M.12 Inhibitors of mitochondrial ATP synthase, e.g. M.12A diafenthiuron, or M.12B organotin miticides such as azocyclotin, cyhexatin, or fenbutatin oxide, M.12C propargite, or M.12D tetradifon;

M.13 Uncouplers of oxidative phosphorylation via disruption of the proton gradient, e.g. chlorfenapyr, DNOC, or sulfluramid;

M.14 Nicotinic acetylcholine receptor (nAChR) channel blockers, e.g. nereistoxin analogues bensultap, cartap hydrochloride, thiocyclam, or thiosultap sodium;

M.15 Inhibitors of the chitin biosynthesis type 0, such as benzoylureas e.g. bistrifluron, chlorfluazuron, diflubenzuron, flucycloxuron, flufenoxuron, hexaflumuron, lufenuron, novaluron, noviflumuron, teflubenzuron, or triflumuron;

M.16 Inhibitors of the chitin biosynthesis type 1, e.g. buprofezin;

M.17 Moulting disruptors, Dipteran, e.g. cyromazine;

M.18 Ecdyson receptor agonists such as diacylhydrazines, e.g. methoxyfenozide, tebufenozide, halofenozide, fufenozide, or chromafenozide;

M.19 Octopamin receptor agonists, e.g. amitraz;

M.20 Mitochondrial complex III electron transport inhibitors, e.g. M.20A hydramethylnon, M.20B acequinocyl, M.20C fluacrypyrim; or M.20D bifenazate;

M.21 Mitochondrial complex I electron transport inhibitors, e.g. M.21A METI acaricides and insecticides such as fenazaquin, fenpyroximate, pyrimidifen, pyridaben, tebufenpyrad or tolfenpyrad, or M.21B rotenone;

M.22 Voltage-dependent sodium channel blockers, e.g. M.22A indoxacarb, M.22B metaflumizone, or M.22B.1: 2-[2-(4-Cyanophenyl)-1-[3-(trifluoromethyl)phenyl]ethylidene]-N-[4-

(difluoromethoxy)phenyl]-hydrazinecarboxamide or M.22B.2: N-(3-Chloro-2-methylphenyl)-2-[(4-chlorophenyl)[4-[methyl(methylsulfonyl)amino]phenyl]methylene]-hydrazinecarboxamide;

M.23 Inhibitors of the of acetyl CoA carboxylase, such as Tetronic and Tetramic acid derivatives, e.g. spirodiclofen, spiromesifen, or spirotetramat; M.23.1 spiropidion

M.24 Mitochondrial complex IV electron transport inhibitors, e.g. M.24A phosphine such as aluminium phosphide, calcium phosphide, phosphine or zinc phosphide, or M.24B cyanide;

M.25 Mitochondrial complex II electron transport inhibitors, such as beta-ketonitrile derivatives, e.g. cyenopyrafen or cyflumetofen;

M.28 Ryanodine receptor-modulators from the class of diamides, e.g. flubendiamide, chlorantraniliprole, cyantraniliprole, tetraniliprole, M.28.1: (R)-3-Chlor-N1-{2-methyl-4-[1,2,2,2 –tetrafluoro-1-(trifluoromethyl)ethyl]phenyl}-N2-(1-methyl-2-methylsulfonylethyl)phthalamid, M.28.2: (S)-3-Chloro-N1-{2-methyl-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]phenyl}-N2-(1-methyl-2-methylsulfonylethyl)phthalamid, M.28.3: cyclaniliprole, or M.28.4: methyl-2-[3,5-dibromo-2-({[3-bromo-1-(3-chlorpyridin-2-yl)-1H-pyrazol-5-yl]carbonyl}amino)benzoyl]-1,2-dimethylhydrazinecarboxylate; or M.28.5a) N-[4,6-dichloro-2-[(diethyl-lambda-4-sulfanylidene)carbamoyl]-phenyl]-2-(3-chloro-2-pyridyl)-5-(trifluoromethyl)pyrazole-3-carboxamide; M.28.5b) N-[4-chloro-2-[(diethyl-lambda-4-sulfanylidene)carbamoyl]-6-methyl-phenyl]-2-(3-chloro-2-pyridyl)-5-(trifluoromethyl)pyrazole-3-carboxamide; M.28.5c) N-[4-chloro-2-[(di-2-propyl-lambda-4-sulfanylidene)carbamoyl]-6-methyl-phenyl]-2-(3-chloro-2-pyridyl)-5-(trifluoromethyl)pyrazole-3-carboxamide; M.28.5d) N-[4,6-dichloro-2-[(di-2-propyl-lambda-4-sulfanylidene)carbamoyl]-phenyl]-2-(3-chloro-2-pyridyl)-5-(trifluoromethyl)pyrazole-3-carboxamide; M.28.5h) N-[4,6-dibromo-2-[(diethyl-lambda-4-sulfanylidene)carbamoyl]-phenyl]-2-(3-chloro-2-pyridyl)-5-(trifluoromethyl)pyrazole-3-carboxamide; M.28.5i) N-[2-(5-Amino-1,3,4-thiadiazol-2-yl)-4-chloro-6-methylphenyl]-3-bromo-1-(3-chloro-2-pyridinyl)-1H-pyrazole-5-carboxamide; M.28.5j) 3-Chloro-1-(3-chloro-2-pyridinyl)-N-[2,4-dichloro-6-[[(1-cyano-1-methylethyl)amino]carbonyl]phenyl]-1H-pyrazole-5-carboxamide; M.28.5k) 3-Bromo-N-[2,4-dichloro-6-(methylcarbamoyl)phenyl]-1-(3,5-dichloro-2-pyridyl)-1H-pyrazole-5-carboxamide; M.28.5l) N-[4-Chloro-2-[[(1,1-dimethylethyl)amino]carbonyl]-6-methyl-phenyl]-1-(3-chloro-2-pyridinyl)-3-(fluoromethoxy)-1H-pyrazole-5-carboxamide; or

M.28.6: cyhalodiamide; or

M.29: Chordotonal organ Modulators – undefined target site, e.g. flonicamid;

M.UN. insecticidal active compounds of unknown or uncertain mode of action, e.g. afidopyropen, afoxolaner, azadirachtin, amidoflumet, benzoximate, broflanilide, bromopropylate, chinomethionat, cryolite, dicloromezotiaz, dicofol, flufenerim, flometoquin, fluensulfone, fluhexafon, fluopyram, fluralaner, metoxadiazone, piperonyl butoxide, pyflubumide, pyridalyl, tioxazafen,

M.UN.3: 11-(4-chloro-2,6-dimethylphenyl)-12-hydroxy-1,4-dioxa-9-azadispiro[4.2.4.2]-tetradec-11-en-10-one,

M.UN.4: 3-(4'-fluoro-2,4-dimethylbiphenyl-3-yl)-4-hydroxy-8-oxa-1-azaspiro[4.5]dec-3-en-2-one,

M.UN.5: 1-[2-fluoro-4-methyl-5-[(2,2,2-trifluoroethyl)sulfinyl]phenyl]-3-(trifluoromethyl)-1H-1,2,4-triazole-5-amine, or actives on basis of bacillus firmus (Votivo, I-1582);

M.UN.6: flupyrimin;

M.UN.8: fluazaindolizine; M.UN.9.a): 4-[5-(3,5-dichlorophenyl)-5-(trifluoromethyl)-4H-isoxazol-3-yl]-2-methyl-N-(1-oxothietan-3-yl)benzamide; M.UN.9.b): fluxametamide; M.UN.10: 5-[3-[2,6-dichloro-4-(3,3-dichloroallyloxy)phenoxy]propoxy]-1H-pyrazole;

M.UN.11.b) 3-(benzoylmethylamino)-N-[2-bromo-4-[1,2,2,3,3,3-hexafluoro-1-(trifluoromethyl)propyl]-6-(trifluoromethyl)phenyl]-2-fluoro-benzamide; M.UN.11.c) 3-(benzoylmethylamino)-2-fluoro-N-[2-iodo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]-6-(trifluoromethyl)phenyl]-benzamide; M.UN.11.d) N-[3-[[[2-iodo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]-6-(trifluoromethyl)phenyl]amino]carbonyl]phenyl]-N-methyl-benzamide; M.UN.11.e) N-[3-[[[2-bromo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]-6-(trifluoromethyl)phenyl]amino]carbonyl]-2-fluoro-phenyl]-4-fluoro-N-methyl-benzamide; M.UN.11.f) 4-fluoro-N-[2-fluoro-3-[[[2-iodo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]-6-(trifluoromethyl)phenyl]amino]carbonyl]phenyl]-N-methyl-benzamide; M.UN.11.g) 3-fluoro-N-[2-fluoro-3-[[[2-iodo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)-ethyl]-6-(trifluoromethyl)phenyl]amino]carbonyl]phenyl]-N-methyl-benzamide; M.UN.11.h) 2-chloro-N-[3-[[[2-iodo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]-6-(trifluoromethyl)phenyl]amino]carbonyl]phenyl]- 3-pyridinecarboxamide; M.UN.11.i) 4-cyano-N-[2-cyano-5-[[2,6-dibromo-4-[1,2,2,3,3,3-hexafluoro-1-(trifluoromethyl)propyl]phenyl]carbamoyl]phenyl]-2-methyl-benzamide; M.UN.11.j) 4-cyano-3-[(4-cyano-2-methyl-benzoyl)amino]-N-[2,6-dichloro-4-[1,2,2,3,3,3-hexafluoro-1-(trifluoromethyl)propyl]phenyl]-2-fluoro-benzamide; M.UN.11.k) N-[5-[[2-chloro-6-cyano-4-[1,2,2,3,3,3-hexafluoro-1-(trifluoromethyl)propyl]phenyl]carbamoyl]-2-cyano-phenyl]-4-cyano-2-methyl-benzamide; M.UN.11.l) N-[5-[[2-bromo-6-chloro-4-[2,2,2-trifluoro-1-hydroxy-1-(trifluoromethyl)ethyl]phenyl]carbamoyl]-2-cyano-phenyl]-4-cyano-2-methyl-benzamide; M.UN.11.m) N-[5-[[2-bromo-6-chloro-4-[1,2,2,3,3,3-hexafluoro-1-(trifluoromethyl)propyl]phenyl]carbamoyl]-2-cyano-phenyl]-4-cyano-2-methyl-benzamide; M.UN.11.n) 4-cyano-N-[2-cyano-5-[[2,6-dichloro-4-[1,2,2,3,3,3-hexafluoro-1-(trifluoromethyl)propyl]phenyl]carbamoyl]phenyl]-2-methyl-benzamide; M.UN.11.o) 4-cyano-N-[2-cyano-5-[[2,6-dichloro-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]phenyl]carbamoyl]phenyl]-2-methyl-benzamide; M.UN.11.p) N-[5-[[2-bromo-6-chloro-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]phenyl]carbamoyl]-2-cyano-phenyl]-4-cyano-2-methyl-benzamide; or

M.UN.12.a) 2-(1,3-Dioxan-2-yl)-6-[2-(3-pyridinyl)-5-thiazolyl]-pyridine; M.UN.12.b) 2-[6-[2-(5-Fluoro-3-pyridinyl)-5-thiazolyl]-2-pyridinyl]-pyrimidine; M.UN.12.c) 2-[6-[2-(3-Pyridinyl)-5-thiazolyl]-2-pyridinyl]-pyrimidine; M.UN.12.d) N-Methylsulfonyl-6-[2-(3-pyridyl)thiazol-5-yl]pyridine-2-carboxamide; M.UN.12.e) N-Methylsulfonyl-6-[2-(3-pyridyl)thiazol-5-yl]pyridine-2-carboxamide; M.UN.12.f) N-Ethyl-N-[4-methyl-2-(3-pyridyl)thiazol-5-yl]-3-methylthio-propanamide; M.UN.12.g) N-Methyl-N-[4-methyl-2-(3-pyridyl)thiazol-5-yl]-3-methylthio-propanamide; M.UN.12.h) N,2-Dimethyl-N-[4-methyl-2-(3-pyridyl)thiazol-5-yl]-3-methylthio-propanamide; M.UN.12.i) N-Ethyl-2-methyl-N-[4-methyl-2-(3-pyridyl)thiazol-5-yl]-3-methylthio-propanamide; M.UN.12.j) N-[4-Chloro-2-(3-pyridyl)thiazol-5-yl]-N-ethyl-2-methyl-3-methylthio-propanamide; M.UN.12.k) N-[4-Chloro-2-(3-pyridyl)thiazol-5-yl]-N,2-dimethyl-3-methylthio-propanamide; M.UN.12.l) N-[4-Chloro-2-(3-pyridyl)thiazol-5-yl]-N-methyl-3-methylthio-propanamide; M.UN.12.m) N-[4-Chloro-2-(3-pyridyl)thiazol-5-yl]-N-ethyl-3-methylthio-propanamide;

M.UN.14a) 1-[(6-Chloro-3-pyridinyl)methyl]-1,2,3,5,6,7-hexahydro-5-methoxy-7-methyl-8-nitro-imidazo[1,2-a]pyridine; or M.UN.14b) 1-[(6-Chloropyridin-3-yl)methyl]-7-methyl-8-nitro-1,2,3,5,6,7-hexahydroimidazo[1,2-a]pyridin-5-ol;

M.UN.16a) 1-isopropyl-N,5-dimethyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; or M.UN.16b) 1-(1,2-dimethylpropyl)-N-ethyl-5-methyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; M.UN.16c) N,5-dimethyl-N-pyridazin-4-yl-1-(2,2,2-trifluoro-1-methyl-ethyl)pyrazole-4-carboxamide; M.UN.16d) 1-[1-(1-cyanocyclopropyl)ethyl]-N-ethyl-5-methyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; M.UN.16e) N-ethyl-1-(2-fluoro-1-methyl-propyl)-5-methyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; M.UN.16f) 1-(1,2-dimethylpropyl)-N,5-dimethyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; M.UN.16g) 1-[1-(1-cyanocyclopropyl)ethyl]-N,5-dimethyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; M.UN.16h) N-methyl-1-(2-fluoro-1-methyl-propyl]-5-methyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; M.UN.16i) 1-(4,4-difluorocyclohexyl)-N-ethyl-5-methyl-N-pyridazin-4-yl-pyrazole-4-carboxamide; or M.UN.16j) 1-(4,4-difluorocyclohexyl)-N,5-dimethyl-N-pyridazin-4-yl-pyrazole-4-carboxamide,

M.UN.17a) N-(1-methylethyl)-2-(3-pyridinyl)-2H-indazole-4-carboxamide; M.UN.17b) N-cyclopropyl-2-(3-pyridinyl)-2H-indazole-4-carboxamide; M.UN.17c) N-cyclohexyl-2-(3-pyridinyl)-2H-indazole-4-carboxamide; M.UN.17d) 2-(3-pyridinyl)-N-(2,2,2-trifluoroethyl)-2H-indazole-4-carboxamide; M.UN.17e) 2-(3-pyridinyl)-N-[(tetrahydro-2-furanyl)methyl]-2H-indazole-5-carboxamide; M.UN.17f) methyl 2-[[2-(3-pyridinyl)-2H-indazol-5-yl]carbonyl]hydrazinecarboxylate; M.UN.17g) N-[(2,2-difluorocyclopropyl)methyl]-2-(3-pyridinyl)-2H-indazole-5-carboxamide; M.UN.17h) N-(2,2-difluoropropyl)-2-(3-pyridinyl)-2H-indazole-5-carboxamide; M.UN.17i) 2-(3-pyridinyl)-N-(2-pyrimidinylmethyl)-2H-indazole-5-carboxamide; M.UN.17j) N-[(5-methyl-2-pyrazinyl)methyl]-2-(3-pyridinyl)-2H-indazole-5-carboxamide,

M.UN.18a) N-[3-chloro-1-(3-pyridyl)pyrazol-4-yl]-N-ethyl-3-(3,3,3-trifluoropropylsulfanyl)propanamide; M.UN.18b) N-[3-chloro-1-(3-pyridyl)pyrazol-4-yl]-N-ethyl-3-(3,3,3-trifluoropropylsulfinyl)propanamide; M.UN.18c) N-[3-chloro-1-(3-pyridyl)pyrazol-4-yl]-3-[(2,2-difluorocyclopropyl)methylsulfanyl]-N-ethyl-propanamide; M.UN.18d) N-[3-chloro-1-(3-pyridyl)pyrazol-4-yl]-3-[(2,2-difluorocyclopropyl)methylsulfinyl]-N-ethyl-propanamide;

M.UN.19 sarolaner, M.UN.20 lotilaner;

M.UN.21 N-[4-Chloro-3-[[(phenylmethyl)amino]carbonyl]phenyl]-1-methyl-3-(1,1,2,2,2-pentafluoroethyl)-4-(trifluoromethyl)-1H-pyrazole-5-carboxamide; M.UN.22a 2-(3-ethylsulfonyl-2-pyridyl)-3-methyl-6-(trifluoromethyl)imidazo[4,5-b]pyridine, or M.UN.22b 2-[3-ethylsulfonyl-5-(trifluoromethyl)-2-pyridyl]-3-methyl-6-(trifluoromethyl)imidazo[4,5-b]pyridine;

M.UN.23a 4-[5-(3,5-dichlorophenyl)-5-(trifluoromethyl)-4H-isoxazol-3-yl]-N-[(4R)-2-ethyl-3-oxo-isoxazolidin-4-yl]-2-methyl-benzamide, or M.UN.23b 4-[5-(3,5-dichloro-4-fluoro-phenyl)-5-(trifluoromethyl)-4H-isoxazol-3-yl]-N-[(4R)-2-ethyl-3-oxo-isoxazolidin-4-yl]-2-methyl-benzamide;

M.UN.24a) N-[4-chloro-3-(cyclopropylcarbamoyl)phenyl]-2-methyl-5-(1,1,2,2,2-pentafluoroethyl)-4-(trifluoromethyl)pyrazole-3-carboxamide or M.UN.24b) N-[4-chloro-3-[(1-cyanocyclopropyl)carbamoyl]phenyl]-2-methyl-5-(1,1,2,2,2-pentafluoroethyl)-4-(trifluoromethyl)pyrazole-3-carboxamide; M.UN.25 acynonapyr; M.UN.26 benzpyrimoxan; M.UN.27 2-chloro-N-(1-cyanocyclopropyl)-5-[1-[2-methyl-5-(1,1,2,2,2-pentafluoroethyl)-4-(trifluoromethyl)pyrazol-3-yl]pyrazol-4-yl]benzamide.

The commercially available compounds of the group M listed above may be found in The Pesticide Manual, 17th Edition, C. MacBean, British Crop Protection Council (2015) among other publications. The online Pesticide Manual is updated regularly and is accessible through http://bcpcdata.com/pesticide-manual.html.

Another online data base for pesticides providing the ISO common names is http://www.alanwood.net/pesticides.

The M.4 cycloxaprid is known from WO2010/069266 and WO2011/069456. M.4A.1 is known from CN 103814937; CN105367557, CN 105481839. M.4A.2, guadipyr, is known from WO 2013/003977, and M.4A.3 (approved as paichongding in China) is known from WO 2007/101369. M.22B.1 is described in CN10171577 and M.22B.2 in CN102126994. Spiropidion M.23.1 is known from WO 2014/191271. M.28.1 and M.28.2 are known from WO2007/101540. M.28.3 is described in WO2005/077934. M.28.4 is described in WO2007/043677. M.28.5a) to M.28.5d) and M.28.5h) are described in WO 2007/006670, WO2013/024009 and WO 2013/024010, M.28.5i) is described in WO2011/085575, M.28.5j) in WO2008/134969, M.28.5k) in US2011/046186 and M.28.5l) in WO2012/034403. M.28.6 can be found in WO2012/034472. M.UN.3 is known from WO2006/089633 and M.UN.4 from WO2008/067911. M.UN.5 is described in WO2006/043635, and biological control agents on the basis of bacillus firmus are described in WO2009/124707. Flupyrimin is described in WO2012/029672. M.UN.8 is known from WO2013/055584. M.UN.9.a) is described in WO2013/050317. M.UN.9.b) is described in WO2014/126208. M.UN.10 is known from WO2010/060379. Broflanilide and M.UN.11.b) to M.UN.11.h) are described in WO2010/018714, and M.UN.11i) to M.UN.11.p) in WO 2010/127926. M.UN.12.a) to M.UN.12.c) are known from WO2010/006713, M.UN.12.d) and M.UN.12.e) are known from WO2012/000896, and M.UN.12.f) to M.UN.12.m) from WO 2010/129497. M.UN.14a) and M.UN.14b) are known from WO2007/101369. M.UN.16.a) to M.UN.16h) are described in WO2010/034737, WO2012/084670, and WO2012/143317, resp., and M.UN.16i) and M.UN.16j) are described in WO2015/055497. M.UN.17a) to M.UN.17.j) are described in WO2015/038503. M.UN.18a) to M.UN.18d) are described in US2014/0213448. M.UN.19 is described in WO2014/036056. M.UN.20 is known from WO2014/090918. M.UN.21 is known from EP2910126. M.UN.22a and M.UN.22b are known from WO2015/059039 and WO2015/190316. M.UN.23a and M.UN.23b are known from WO2013/050302. M.UN.24a and M.UN.24b are known from WO2012/126766. Acynonapyr M.UN.25 is known from WO 2011/105506. Benzpyrimoxan M.UN.26 is known from WO2016/104516. M.UN.27 is known from WO2016174049.

The following list of fungicides, in conjunction with which the compounds of the present invention can be used, is intended to illustrate the possible combinations in the methods according to the invention but does not limit them:

A) Respiration inhibitors

- Inhibitors of complex III at Qo site: azoxystrobin (A.1.1), coumethoxystrobin (A.1.2), coumoxystrobin (A.1.3), dimoxystrobin (A.1.4), enestroburin (A.1.5), fenaminstrobin (A.1.6), fenoxystrobin/flufenoxystrobin (A.1.7), fluoxastrobin (A.1.8), kresoxim-methyl (A.1.9), mandestrobin (A.1.10), metominostrobin (A.1.11), orysastrobin (A.1.12), picoxystrobin (A.1.13), pyraclostrobin (A.1.14), pyrametostrobin (A.1.15), pyraoxystrobin (A.1.16), trifloxystrobin (A.1.17), 2 (2-(3-(2,6-dichlorophenyl)-1-methyl-allylideneaminooxymethyl)-phenyl)-2 methoxyimino-N methyl-acetamide (A.1.18), pyribencarb (A.1.19), triclopyricarb/chlorodincarb (A.1.20), famoxadone (A.1.21), fenamidone (A.1.21), methyl-N-[2-[(1,4-dimethyl-5 phenyl-pyrazol-3-yl)oxylmethyl]phenyl]-N-methoxy-carbamate (A.1.22), 1-[3-chloro-2 [[1 (4-chlorophenyl)-1H-pyrazol-3-yl]oxymethyl]phenyl]-4-methyl-tetrazol-5-one (A.1.23), 1-[3-bromo-2-[[1-(4-chlorophenyl)pyrazol-3-

yl]oxymethyl]phenyl]-4-methyl-tetrazol-5-one (A.1.24), 1-[2-[[1-(4-chlorophenyl)pyrazol-3-yl]oxymethyl]-3-methyl-phenyl]-4-methyl-tetrazol-5-one (A.1.25), 1-[2-[[1-(4-chlorophenyl)pyrazol-3-yl]oxymethyl]-3-fluoro-phenyl]-4-methyl-tetrazol-5-one (A.1.26), 1-[2-[[1-(2,4-dichlorophenyl)pyrazol-3-yl]oxymethyl]-3-fluoro-phenyl]-4-methyl-tetrazol-5-one (A.1.27), 1-[3-cyclopropyl-2-[[2-methyl-4 (1 methylpyrazol-3-yl)phenoxy]methyl]phenyl]-4 methyl-tetrazol-5-one (A.1.30), 1 [3 (difluoromethoxy)-2-[[2-methyl-4-(1 methylpyrazol-3 yl)phenoxy]methyl]phenyl]-4 methyl-tetrazol-5-one (A.1.31), 1-methyl-4-[3-methyl-2 [[2 methyl-4-(1-methylpyrazol-3 yl)phenoxy]methyl]phenyl]tetrazol-5-one (A.1.32), (Z,2E)-5-[1-(2,4-dichlorophenyl)pyrazol-3-yl]-oxy-2-methoxyimino-N,3-dimethyl-pent-3-enamide (A.1.34), (Z,2E) 5 [1 (4-chlorophenyl)pyrazol-3-yl]oxy-2-methoxyimino-N,3-dimethyl-pent-3-enamide (A.1.35), pyriminostrobin (A.1.36), bifujunzhi (A.1.37), 2-(ortho-((2,5-dimethylphenyl-oxy-methylen)phenyl)-3-methoxy-acrylic acid methylester (A.1.38);

- inhibitors of complex III at Qi site: cyazofamid (A.2.1), amisulbrom (A.2.2), [(6S,7R,8R) 8 benzyl-3-[(3-hydroxy-4-methoxy-pyridine-2-carbonyl)amino]-6-methyl-4,9-dioxo-1,5-dioxonan-7-yl] 2-methylpropanoate (A.2.3), fenpicoxamid (A.2.4);
- inhibitors of complex II: benodanil (A.3.1), benzovindiflupyr (A.3.2), bixafen (A.3.3), boscalid (A.3.4), carboxin (A.3.5), fenfuram (A.3.6), fluopyram (A.3.7), flutolanil (A.3.8), fluxapyroxad (A.3.9), furametpyr (A.3.10), isofetamid (A.3.11), isopyrazam (A.3.12), mepronil (A.3.13), oxycarboxin (A.3.14), penflufen (A.3.15), penthiopyrad (A.3.16), pydiflumetofen (A.3.17), pyraziflumid (A.3.18), sedaxane (A.3.19), tecloftalam (A.3.20), thifluzamide (A.3.21), 3 (difluoromethyl)-1-methyl-N-(1,1,3-trimethylindan-4-yl)pyrazole-4 carboxamide (A.3.22), 3 (trifluoromethyl)-1-methyl-N-(1,1,3-trimethylindan-4-yl)pyrazole-4 carboxamide (A.3.23), 1,3-dimethyl-N-(1,1,3-trimethylindan-4-yl)pyrazole-4-carboxamide (A.3.24), 3-(trifluoromethyl)-1,5 dimethyl-N-(1,1,3-trimethylindan-4-yl)pyrazole-4-carboxamide (A.3.25), 1,3,5-trimethyl-N-(1,1,3-trimethylindan-4-yl)pyrazole-4-carboxamide (A.3.26), 3-(difluoromethyl)-1,5 dimethyl-N-(1,1,3-trimethylindan-4-yl)pyrazole-4-carboxamide (A.3.27), 3-(difluoromethyl)-N (7 fluoro-1,1,3-trimethyl-indan-4-yl)-1-methyl-pyrazole-4-carboxamide (A.3.28), N-[(5-chloro-2-isopropyl-phenyl)methyl]-N-cyclopropyl-5-fluoro-1,3-dimethyl-pyrazole-4-carboxamide (A.3.29), methyl (E)-2-[2-[(5-cyano-2-methyl-phenoxy)methyl]phenyl]-3-methoxy-prop-2 enoate (A.3.30), N-[(5-chloro-2-isopropyl-phenyl)methyl]-N-cyclopropyl-3-(difluoromethyl)-5 fluoro-1-methyl-pyrazole-4-carboxamide (A.3.31), 2-(difluoromethyl)-N-(1,1,3-trimethyl-indan-4-yl)pyridine-3-carboxamide (A.3.32), 2-(difluoromethyl)-N-[(3R)-1,1,3-trimethylindan-4-yl]pyridine-3-carboxamide (A.3.33), 2-(difluoromethyl)-N-(3-ethyl-1,1-dimethyl-indan-4-yl)pyridine-3-carboxamide (A.3.34), 2-(difluoromethyl)-N-[(3R)-3-ethyl-1,1-dimethyl-indan-4-yl]pyridine-3-carboxamide (A.3.35), 2-(difluoromethyl)-N-(1,1-dimethyl-3-propyl-indan-4-yl)pyridine-3-carboxamide (A.3.36), 2-(difluoromethyl)-N-[(3R)-1,1-dimethyl-3-propyl-indan-4-yl]pyridine-3-carboxamide (A.3.37), 2-(difluoromethyl)-N-(3-isobutyl-1,1-dimethyl-indan-4-yl)pyridine-3-carboxamide (A.3.38), 2-(difluoromethyl)-N-[(3R)-3-isobutyl-1,1-dimethyl-indan-4 yl]pyridine-3-carboxamide (A.3.39);
- other respiration inhibitors: diflumetorim (A.4.1); nitrophenyl derivates: binapacryl (A.4.2), dinobuton (A.4.3), dinocap (A.4.4), fluazinam (A.4.5), meptyldinocap (A.4.6), ferimzone (A.4.7); organometal compounds: fentin salts, e. g. fentin-acetate (A.4.8), fentin chloride (A.4.9) or fentin hydroxide (A.4.10); ametoctradin (A.4.11); silthiofam (A.4.12);

B) Sterol biosynthesis inhibitors (SBI fungicides)

- C14 demethylase inhibitors: triazoles: azaconazole (B.1.1), bitertanol (B.1.2), bromu-conazole (B.1.3), cyproconazole (B.1.4), difenoconazole (B.1.5), diniconazole (B.1.6), diniconazole-M (B.1.7), epoxiconazole (B.1.8), fenbuconazole (B.1.9), fluquinconazole (B.1.10), flusilazole (B.1.11), flutriafol (B.1.12), hexaconazole (B.1.13), imibenconazole (B.1.14), ipconazole (B.1.15), metconazole (B.1.17), myclobutanil (B.1.18), oxpoconazole (B.1.19), paclobutrazole (B.1.20), penconazole (B.1.21), propiconazole (B.1.22), prothioconazole (B.1.23), simeconazole (B.1.24), tebuconazole (B.1.25), tetraconazole (B.1.26), triadimefon (B.1.27), triadimenol (B.1.28), triticonazole (B.1.29), uniconazole (B.1.30), ipfentrifluconazole, (B.1.37), mefentrifluconazole (B.1.38), 2-(chloromethyl)-2-methyl-5-(p-tolylmethyl)-1 (1,2,4-triazol-1-ylmethyl)cyclopentanol (B.1.43); imidazoles: imazalil (B.1.44), pefurazoate (B.1.45), prochloraz (B.1.46), triflumizol (B.1.47); pyrimidines, pyridines and piperazines: fenarimol (B.1.49), pyrifenox (B.1.50), triforine (B.1.51), [3-(4-chloro-2-fluoro-phenyl)-5-(2,4-difluorophenyl)isoxazol-4-yl]-(3-pyridyl)methanol (B.1.52);
- Delta14-reductase inhibitors: aldimorph (B.2.1), dodemorph (B.2.2), dodemorph-acetate (B.2.3), fenpropimorph (B.2.4), tridemorph (B.2.5), fenpropidin (B.2.6), piperalin (B.2.7), spiroxamine (B.2.8);
- Inhibitors of 3-keto reductase: fenhexamid (B.3.1);
- Other Sterol biosynthesis inhibitors: chlorphenomizole (B.4.1);

C) Nucleic acid synthesis inhibitors

- phenylamides or acyl amino acid fungicides: benalaxyl (C.1.1), benalaxyl-M (C.1.2), kiralaxyl (C.1.3), metalaxyl (C.1.4), metalaxyl-M (C.1.5), ofurace (C.1.6), oxadixyl (C.1.7);
- other nucleic acid synthesis inhibitors: hymexazole (C.2.1), octhilinone (C.2.2), oxolinic acid (C.2.3), bupirimate (C.2.4), 5-fluorocytosine (C.2.5), 5-fluoro-2-(p-tolylmethoxy)pyrimidin-4 amine (C.2.6), 5-fluoro-2-(4-fluorophenylmethoxy)pyrimidin-4 amine (C.2.7), 5-fluoro-2 (4 chlorophenylmethoxy)pyrimidin-4 amine (C.2.8);

D) Inhibitors of cell division and cytoskeleton

- tubulin inhibitors: benomyl (D.1.1), carbendazim (D.1.2), fuberidazole (D1.3), thiabendazole (D.1.4), thiophanate-methyl (D.1.5), 3-chloro-4-(2,6-difluorophenyl)-6-methyl-5-phenyl-pyridazine (D.1.6), 3-chloro-6-methyl-5-phenyl-4-(2,4,6-trifluorophenyl)pyridazine (D.1.7), N ethyl-2-[(3-ethynyl-8-methyl-6-quinolyl)oxy]butanamide (D.1.8), N-ethyl-2-[(3-ethynyl-8 methyl-6 quinolyl)oxy]-2-methylsulfanyl-acetamide (D.1.9), 2-[(3-ethynyl-8-methyl-6-quinol-yl)oxy]-N (2-fluoroethyl)butanamide (D.1.10), 2-[(3-ethynyl-8-methyl-6-quinolyl)oxy]-N-(2-flu-oroethyl)-2-methoxy-acetamide (D.1.11), 2-[(3-ethynyl-8-methyl-6-quinolyl)oxy]-N-propyl-butanamide (D.1.12), 2-[(3-ethynyl-8-methyl-6-quinolyl)oxy]-2-methoxy-N-propyl-acetamide (D.1.13), 2-[(3-ethynyl-8-methyl-6-quinolyl)oxy]-2-methylsulfanyl-N-propyl-acetamide (D.1.14), 2 [(3 ethynyl-8-methyl-6-quinolyl)oxy]-N-(2-fluoroethyl)-2-methylsulfanyl-acetamide (D.1.15), 4-(2-bromo-4-fluoro-phenyl)-N-(2-chloro-6-fluoro-phenyl)-2,5-dimethyl-pyrazol-3 amine (D.1.16);
- other cell division inhibitors: diethofencarb (D.2.1), ethaboxam (D.2.2), pencycuron (D.2.3), fluopicolide (D.2.4), zoxamide (D.2.5), metrafenone (D.2.6), pyriofenone (D.2.7);

E) Inhibitors of amino acid and protein synthesis

- methionine synthesis inhibitors: cyprodinil (E.1.1), mepanipyrim (E.1.2), pyrimethanil (E.1.3);
- protein synthesis inhibitors: blasticidin-S (E.2.1), kasugamycin (E.2.2), kasugamycin

hydrochloride-hydrate (E.2.3), mildiomycin (E.2.4), streptomycin (E.2.5), oxytetracyclin (E.2.6);

F) Signal transduction inhibitors

- MAP / histidine kinase inhibitors: fluoroimid (F.1.1), iprodione (F.1.2), procymidone (F.1.3), vinclozolin (F.1.4), fludioxonil (F.1.5);
- G protein inhibitors: quinoxyfen (F.2.1);

G) Lipid and membrane synthesis inhibitors

- Phospholipid biosynthesis inhibitors: edifenphos (G.1.1), iprobenfos (G.1.2), pyrazophos (G.1.3), isoprothiolane (G.1.4);
- lipid peroxidation: dicloran (G.2.1), quintozene (G.2.2), tecnazene (G.2.3), tolclofos-methyl (G.2.4), biphenyl (G.2.5), chloroneb (G.2.6), etridiazole (G.2.7);
- phospholipid biosynthesis and cell wall deposition: dimethomorph (G.3.1), flumorph (G.3.2), mandipropamid (G.3.3), pyrimorph (G.3.4), benthiavalicarb (G.3.5), iprovalicarb (G.3.6), valifenalate (G.3.7);
- compounds affecting cell membrane permeability and fatty acides: propamocarb (G.4.1);
- inhibitors of oxysterol binding protein: oxathiapiprolin (G.5.1), 2-{3-[2-(1-{[3,5-bis(difluoromethyl-1H-pyrazol-1-yl]acetyl}piperidin-4-yl)-1,3-thiazol-4-yl]-4,5-dihydro-1,2 oxazol-5-yl}phenyl methanesulfonate (G.5.2), 2-{3-[2-(1-{[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]acetyl}piperidin-4-yl) 1,3-thiazol-4-yl]-4,5-dihydro-1,2-oxazol-5 yl}-3-chlorophenyl methanesulfonate (G.5.3), 4-[1-[2-[3-(difluoromethyl)-5-methyl-pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.4), 4-[1-[2-[3,5-bis(difluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.5), 4-[1-[2-[3-(difluoromethyl)-5-(trifluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.6), 4-[1-[2-[5-cyclopropyl-3-(difluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.7), 4-[1-[2-[5-methyl-3-(trifluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.8), 4-[1-[2-[5-(difluoromethyl)-3-(trifluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.9), 4 [1 [2-[3,5-bis(trifluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.10), (4-[1-[2-[5-cyclopropyl-3-(trifluoromethyl)pyrazol-1-yl]acetyl]-4-piperidyl]-N-tetralin-1-yl-pyridine-2-carboxamide (G.5.11);

H) Inhibitors with Multi Site Action

- inorganic active substances: Bordeaux mixture (H.1.1), copper (H.1.2), copper acetate (H.1.3), copper hydroxide (H.1.4), copper oxychloride (H.1.5), basic copper sulfate (H.1.6), sulfur (H.1.7);
- thio- and dithiocarbamates: ferbam (H.2.1), mancozeb (H.2.2), maneb (H.2.3), metam (H.2.4), metiram (H.2.5), propineb (H.2.6), thiram (H.2.7), zineb (H.2.8), ziram (H.2.9);
- organochlorine compounds: anilazine (H.3.1), chlorothalonil (H.3.2), captafol (H.3.3), captan (H.3.4), folpet (H.3.5), dichlofluanid (H.3.6), dichlorophen (H.3.7), hexachlorobenzene (H.3.8), pentachlorphenole (H.3.9) and its salts, phthalide (H.3.10), tolylfluanid (H.3.11);
- guanidines and others: guanidine (H.4.1), dodine (H.4.2), dodine free base (H.4.3), guazatine (H.4.4), guazatine-acetate (H.4.5), iminoctadine (H.4.6), iminoctadine-triacetate (H.4.7), iminoctadine-tris(albesilate) (H.4.8), dithianon (H.4.9), 2,6-dimethyl-1H,5H-[1,4]dithiino[2,3-c:5,6-c']dipyrrole-1,3,5,7(2H,6H)-tetraone (H.4.10);

I) Cell wall synthesis inhibitors

- inhibitors of glucan synthesis: validamycin (I.1.1), polyoxin B (I.1.2);

- melanin synthesis inhibitors: pyroquilon (I.2.1), tricyclazole (I.2.2), carpropamid (I.2.3), diclocymet (I.2.4), fenoxanil (I.2.5);

J) Plant defence inducers

- acibenzolar-S-methyl (J.1.1), probenazole (J.1.2), isotianil (J.1.3), tiadinil (J.1.4), prohexadione-calcium (J.1.5); phosphonates: fosetyl (J.1.6), fosetyl-aluminum (J.1.7), phosphorous acid and its salts (J.1.8), calcium phosphonate (J.1.11), potassium phosphonate (J.1.12), potassium or sodium bicarbonate (J.1.9), 4 cyclopropyl-N-(2,4-dimethoxyphenyl)thiadiazole-5-carboxamide (J.1.10);

K) Unknown mode of action

- bronopol (K.1.1), chinomethionat (K.1.2), cyflufenamid (K.1.3), cymoxanil (K.1.4), dazomet (K.1.5), debacarb (K.1.6), diclocymet (K.1.7), diclomezine (K.1.8), difenzoquat (K.1.9), difenzoquat-methylsulfate (K.1.10), diphenylamin (K.1.11), fenitropan (K.1.12), fenpyrazamine (K.1.13), flumetover (K.1.14), flusulfamide (K.1.15), flutianil (K.1.16), harpin (K.1.17), metha-sulfocarb (K.1.18), nitrapyrin (K.1.19), nitrothal-isopropyl (K.1.20), tolprocarb (K.1.21), oxin-copper (K.1.22), proquinazid (K.1.23), tebufloquin (K.1.24), tecloftalam (K.1.25), triazoxide (K.1.26), N'-(4-(4-chloro-3-trifluoromethyl-phenoxy)-2,5-dimethyl-phenyl)-N-ethyl-N methyl formamidine (K.1.27), N' (4-(4-fluoro-3-trifluoromethyl-phenoxy)-2,5-dimethyl-phenyl)-N-ethyl-N-methyl formamidine (K.1.28), N'-[4-[[3-[(4-chlorophenyl)methyl]-1,2,4-thiadiazol-5-yl]oxy]-2,5-dimethyl-phenyl]-N-ethyl-N-methyl-formamidine (K.1.29), N'-(5-bromo-6-indan-2-yloxy-2-methyl-3-pyridyl)-N-ethyl-N-methyl-formamidine (K.1.30), N'-[5-bromo-6-[1-(3,5-difluorophenyl)ethoxy]-2-methyl-3-pyridyl]-N-ethyl-N-methyl-formamidine (K.1.31), N'-[5-bromo-6-(4-isopropylcyclohexoxy)-2-methyl-3-pyridyl]-N-ethyl-N-methyl-formamidine (K.1.32), N' [5 bromo-2-methyl-6-(1-phenylethoxy)-3-pyridyl]-N-ethyl-N-methyl-formamidine (K.1.33), N'-(2-methyl-5-trifluoromethyl-4-(3-trimethylsilanyl-propoxy)-phenyl)-N-ethyl-N-methyl formamidine (K.1.34), N'-(5-difluoromethyl-2 methyl-4-(3-trimethylsilanyl-propoxy)-phenyl)-N-ethyl-N-methyl formamidine (K.1.35), 2-(4-chloro-phenyl)-N-[4-(3,4-dimethoxy-phenyl)-isoxazol-5 yl]-2-prop-2-ynyloxy-acetamide (K.1.36), 3 [5-(4-chloro-phenyl)-2,3-dimethyl-isoxazolidin-3-yl]-pyridine (pyrisoxazole) (K.1.37), 3-[5-(4-methyl-phenyl)-2,3-dimethyl-isoxazolidin-3 yl]-pyridine (K.1.38), 5-chloro-1 (4,6-dimethoxy-pyrimidin-2-yl)-2-methyl-1H-benzoimidazole (K.1.39), ethyl (Z) 3 amino-2-cyano-3-phenyl-prop-2-enoate (K.1.40), picarbutrazox (K.1.41), pentyl N-[6-[[(Z)-[(1-methyltetrazol-5-yl)-phenyl-methylene]amino]oxymethyl]-2-pyridyl]carbamate (K.1.42), but-3-ynyl N-[6-[[(Z)-[(1-methyltetrazol-5-yl)-phenyl-methylene]amino]oxymethyl]-2-pyridyl]carbamate (K.1.43), 2-[2-[(7,8-difluoro-2-methyl-3-quinolyl)oxy]-6-fluoro-phenyl]propan-2-ol (K.1.44), 2-[2-fluoro-6-[(8-fluoro-2-methyl-3-quinolyl)oxy]phen-yl]propan-2-ol (K.1.45), quinofumelin (K.1.47), 9-fluoro-2,2-dimethyl-5-(3-quinolyl)-3H 1,4 benzoxazepine (K.1.49), 2-(6-benzyl-2-pyridyl)quinazoline (K.1.50), 2-[6-(3-fluoro-4 methoxy-phenyl)-5-methyl-2-pyridyl]quinazoline (K.1.51), dichlobentiazox (K.1.52), N'-(2,5-dimethyl-4-phenoxy-phenyl)-N-ethyl-N-methyl-formamidine (K.1.53);

dipymetitrone, isoflucypram; fluindapyr, inpyrfluxam, pyrifenamine.

The fungicides described by IUPAC nomenclature, their preparation and their pesticidal activity is also known (cf. Can. J. Plant Sci. 48(6), 587-94, 1968; EP-A 141 317; EP-A 152 031; EP-A 226 917; EP-A 243 970; EP-A 256 503; EP-A 428 941; EP-A 532 022; EP-A 1 028 125; EP-A 1 035 122; EP-A 1 201 648; EP-A 1 122 244, JP 2002316902; DE 19650197; DE 10021412; DE 102005009458; US 3,296,272; US 3,325,503; WO 98/46608; WO 99/14187; WO 99/24413;

WO 99/27783; WO 00/29404; WO 00/46148; WO 00/65913; WO 01/54501; WO 01/56358; WO 02/22583; WO 02/40431; WO 03/10149; WO 03/11853; WO 03/14103; WO 03/16286; WO 03/53145; WO 03/61388; WO 03/66609; WO 03/74491; WO 04/49804; WO 04/83193; WO 05/120234; WO 05/123689; WO 05/123690; WO 05/63721; WO 05/87772; WO 05/87773; WO 06/15866; WO 06/87325; WO 06/87343; WO 07/82098; WO 07/90624, WO 10/139271, WO 11/028657, WO2012/168188, WO 2007/006670, WO 2011/77514; WO13/047749, WO 10/069882, WO 13/047441, WO 03/16303, WO 09/90181, WO 13/007767, WO 13/010862, WO 13/127704, WO 13/024009, WO 13/024010 and WO 13/047441, WO 13/162072, WO 13/092224, WO 11/135833), CN 1907024, CN 1456054, CN 103387541, CN 1309897, WO 12/84812, CN 1907024, WO 09094442, WO 14/60177, WO 13/116251, WO 08/013622, WO 15/65922, WO 94/01546, EP 2865265, WO 07/129454, WO 12/165511, WO 11/081174, WO 13/47441).

If the compounds of the present invention are mixed with one or more fungicides, the mixtures are also suitable for combating or controlling plant diseases, as caused by phytophathogenic fungi. Examples of phytophathogenic fungi in rice are

Alternaria species on rice, Bipolaris (e.g. Bipolaris oryzae), and Drechslera species on rice, Cercospora oryzae, Cochliobolus miyabeanus, Curvularia lunata, Sarocladium oryzae, S attenuatum, Entyloma oryzae, Fusarium spp such as Fusarium semitectum (and/or moniliforme

Gibberella fujikuroi (bakanae), Grainstaining complex (various pathogens), and/or Pythium ssp.

Helminthosporium. spp, for example Helminthosporium oryzae, Microdochium oryzae, Pyricularia grisea (syn. Pyricularia oryzae), Rhizoctonia species, for example Rhizoctonia solani (syn in rice Pellicularia sasakii), Corticium sasakii and Ustilaginoidea virens.

In a further embodiment, the invention relates to methods according to the invention, applying mixtures comprising a compound of formula (I) as described above, in particular I-1-R, and at least one compound II which is metaaldehyde, in particular granular metaaldehyde.

In a further embodiment, the invention relates to methods according to the invention, applying mixtures comprising a compound of formula (I) as described above, in particular I-R-1, and at least one compound II selected from the group of methiadinil, anthraquinones, beta-aminobutyric acid, laminarin, chitosan, thiamine and riboflavin.

In a further embodiment, the invention relates to methods according to the invention, applying mixtures comprising a compound of formula (I) as described above, in particular, in particular I-R-1, and at least one compound II selected from the group of oxyenadenine (also called zeatin), kinetin (oxy)enadenine, brassinolides, insecticidal extracts of Celastrus angulatus, matrine, cnidiadin, tetramycin.

In one embodiment, the methods of the present invention comprise applying a mixture of at least one compound I of the present invention with at least one mixing partner II as defined above by seedling box application. In one embodiment, the invention relates to methods applying binary mixtures of one compound I with one mixing partner II as defined above as component II by seedling box application.

Preferred weight ratios for such binary mixtures are from 5000:1 to 1:5000, preferably from 1000:1 to 1:1000, more preferably from 100:1 to 1:100, particularly preferably from 10:1 to 1:10.

In such binary mixtures, components I and II may be used in equal amounts, or an excess of component I, or an excess of component II may be used.

In the mixtures of the present invention, the ingredients may be used sequentially or in combination with each other, if appropriate also added only immediately prior to use (tank mix). For example, the plant(s) may be sprayed with compound II either before or after being treated with compound I.

In another embodiment, the invention relates to methods applying mixtures of one compound I with one mixing partner II, and optionally further pesticides by seedling box application.

In one embodiment of the invention, the mixtures of the present invention are mixtures of a compound of formula (I), preferably I-R-1, with a compound selected from the group of benomyl, carbendazim, epoxiconazole, fluquinconazole, flutriafol, flusilazole, metconazole, prochloraz, prothioconazole, tebuconazole, triticonazole, pyraclostrobin, trifloxystrobin, boscalid, dimethomorph, penthiopyrad, dodemorph, famoxadone, fenpropimorph, proquinazid, pyrimethanil, tridemorph, maneb, mancozeb, metiram, thiram, chlorothalonil, dithianon, flusulfamide,metrafenone, fluxapyroxad, bixafen, penflufen, sedaxane, isopyrazam. Especially preferred is pyraclostrobin and fluxapyroxad.

In one embodiment of the invention, the mixtures of the present invention are mixtures of a compound of formula (I), preferably I-R-1, with a compound selected from the group of imidacloprid, clothianidin, dinotefuran, chlorantraniliprole, cyantraniliprole, spinetoram, spinosad, ethiprole, fipronil, triflumezopyrim, flonicamid and tetraniliprole.

In one embodiment of the invention, the mixtures of the present invention are mixtures of a compound of formula (I), preferably I-R-1, with a compound selected from the group of probenazole, isotianil, tricyclazole, pyroquilon, isoprothiolane, tolprocarb, carpropamid, diclocymet, azoxystrobin, orysastrobin.

In one embodiment of the invention, the mixtures of the present invention are mixtures of a compound of formula (I), preferably I-R-1, with a compound selected from the group of furametpyr, thifluzamide, simeconazole, penflufen, azoxystrobin, orysastrobin,

Especially preferred mixtures in the methods according to the invention are listed in the following table M, wherein the compounds I, I-R-1, are as defined in the description:

Table M:

Mixture	Comp. I	Compound II
M-1	I	fipronil
M-2	I	alpha-cypermethrin
M-3	I	chlorfenapyr
M-4	I	metaflumizone
M-5	I	abamectin
M-6	I	pymetrozine
M-7	I	thiamethoxam
M-8	I	imidacloprid
M-9	I	dinotefuran
M-10	I	clothianidin
M-11	I	bifenthrin
M-12	I	acetamiprid

Mixture	Comp. I	Compound II
M-13	I	nitenpyram
M-14	I	cypermethrin
M-15	I	cyhalothrin
M-16	I	lambda-cyhalothrin
M-17	I	flonicamid
M-18	I	spirotetramat
M-19	I	buprofezine
M-20	I	chlorantraniliprole
M-21	I	cyantraniliprole
M-22	I	tetraniliprole
M-23	I	sulfoxaflor
M-24	I	indoxacarb

Mixture	Comp. I	Compound II
M-25	I	afidopyropen
M-26	I	broflanilide
M-27	I	pyriprole
M-28	I	triflumezopyrim
M-29	I	flupyradifurone
M-30	I	dicloromezotiaz
M-31	I	chlorpyrifos
M-32	I	dichlorvos
M-33	I	triazophos
M-34	I	cartap
M-35	I	acephate
M-36	I	carbofuran
M-37	I	carbosulfan
M-38	I	emamectin
M-39	I	ethiprole
M-40	I	etofenprox
M-41	I	spinetoram
M-42	I	spinosad
M-43	I	fluhexafon
M-44	I	tefluthrin
M-45	I	momfluorothrin
M-46	I	benzpyrimoxan
M-47	I	cyhalodiamide
M-48	I	spiropidion
M-49	I	flupyrimin
M-50	I	cyclaniliprole
M-51	I	fluxametamide
M-52	I	tioxazafen
M-53	I	fluazaindolizine
M-54	I	pyrifluquinazone
M-55	I	metaaldehyde (in particular granular)
M-56	I	benomyl
M-57	I	epoxiconazole
M-58	I	fluquinconazole
M-59	I	flutriafol
M-60	I	flusilazole
M-61	I	metconazole
M-62	I	prochloraz
M-63	I	prothioconazole
M-64	I	tebuconazole

Mixture	Comp. I	Compound II
M-65	I	triticonazole
M-66	I	pyraclostrobin
M-67	I	trifloxystrobin
M-68	I	boscalid
M-69	I	dimethomorph
M-70	I	penthiopyrad
M-71	I	dodemorph
M-72	I	famoxadone
M-73	I	fenpropimorph
M-74	I	proquinazid
M-75	I	pyrimethanil
M-76	I	tridemorph
M-77	I	maneb
M-78	I	metiram
M-79	I	thiram
M-80	I	chlorothalonil
M-81	I	dithianon
M-82	I	flusulfamide
M-83	I	metrafenone
M-84	I	fluxapyroxad
M-85	I	bixafen
M-86	I	penflufen
M-87	I	sedaxane
M-88	I	isopyrazam
M-89	I	tricyclazole
M-90	I	azoxystrobin
M-91	I	difenoconazole
M-92	I	kasugamycin
M-93	I	isoprothiolane
M-94	I	tolprocarb
M-95	I	carpropamid
M-96	I	diclocymet
M-97	I	furametpyr
M-98	I	simeconazole
M-99	I	probenazole
M-100	I	mancozeb
M-101	I	propiconazole
M-102	I	hexaconazole
M-103	I	tebuconazole
M-104	I	carbendazim
M-105	I	flutolanil

Mixture	Comp. I	Compound II
M-106	I	hymexazol
M-107	I	isotianil
M-108	I	orysastrobin
M-109	I	pencycuron
M-110	I	phthalide
M-111	I	pyroquilon
M-112	I	thifluzamide
M-113	I	thiophanate
M-114	I	thiophanate-methyl
M-115	I	tiadinil
M-116	I	validamycin
M-117	I	tebufloquin
M-118	I	benzovindiflupyr
M-119	I	picarbutrazox
M-120	I	pyraziflumid
M-121	I	dipymetitrone
M-122	I	pydiflumetofen
M-123	I	quinofumelin
M-124	I	ipfentrifluconazole
M-125	I	dichlobentiazox
M-126	I	fenpicoxamid
M-127	I	isoflucypram
M-128	I	fluindapyr
M-129	I	inpyrfluxam
M-130	I	pyrifenamine
M-131	I	mefentrifluconazole
M-132	I-R-1	fipronil
M-133	I-R-1	alpha-cypermethrin
M-134	I-R-1	chlorfenapyr
M-135	I-R-1	metaflumizone
M-136	I-R-1	abamectin
M-137	I-R-1	pymetrozine
M-138	I-R-1	thiamethoxam
M-139	I-R-1	imidacloprid
M-140	I-R-1	dinotefuran
M-141	I-R-1	clothianidin
M-142	I-R-1	bifenthrin
M-143	I-R-1	acetamiprid
M-144	I-R-1	nitenpyram
M-145	I-R-1	cypermethrin
M-146	I-R-1	cyhalothrin

Mixture	Comp. I	Compound II
M-147	I-R-1	lambda-cyhalothrin
M-148	I-R-1	flonicamid
M-149	I-R-1	spirotetramat
M-150	I-R-1	buprofezine
M-151	I-R-1	chlorantraniliprole
M-152	I-R-1	cyantraniliprole
M-153	I-R-1	tetraniliprole
M-154	I-R-1	sulfoxaflor
M-155	I-R-1	indoxacarb
M-156	I-R-1	afidopyropen
M-157	I-R-1	broflanilide
M-158	I-R-1	pyriprole
M-159	I-R-1	triflumezopyrim
M-160	I-R-1	flupyradifurone
M-161	I-R-1	dicloromezotiaz
M-162	I-R-1	chlorpyrifos
M-163	I-R-1	dichlorvos
M-164	I-R-1	triazophos
M-165	I-R-1	cartap
M-166	I-R-1	acephate
M-167	I-R-1	carbofuran
M-168	I-R-1	carbosulfan
M-169	I-R-1	emamectin
M-170	I-R-1	ethiprole
M-171	I-R-1	etofenprox
M-172	I-R-1	spinetoram
M-173	I-R-1	spinosad
M-174	I-R-1	fluhexafon
M-175	I-R-1	tefluthrin
M-176	I-R-1	momfluorothrin
M-177	I-R-1	benzpyrimoxan
M-178	I-R-1	cyhalodiamide
M-179	I-R-1	spiropidion
M-180	I-R-1	flupyrimin
M-181	I-R-1	cyclaniliprole
M-182	I-R-1	fluxametamide
M-183	I-R-1	tioxazafen
M-184	I-R-1	fluazaindolizine
M-185	I-R-1	pyrifluquinazone
M-186	I-R-1	metaaldehyde (in particular granular)

Mixture	Comp. I	Compound II
M-187	I-R-1	benomyl
M-188	I-R-1	epoxiconazole
M-189	I-R-1	fluquinconazole
M-190	I-R-1	flutriafol
M-191	I-R-1	flusilazole
M-192	I-R-1	metconazole
M-193	I-R-1	prochloraz
M-194	I-R-1	prothioconazole
M-195	I-R-1	tebuconazole
M-196	I-R-1	triticonazole
M-197	I-R-1	pyraclostrobin
M-198	I-R-1	trifloxystrobin
M-199	I-R-1	boscalid
M-200	I-R-1	dimethomorph
M-201	I-R-1	penthiopyrad
M-202	I-R-1	dodemorph
M-203	I-R-1	famoxadone
M-204	I-R-1	fenpropimorph
M-205	I-R-1	proquinazid
M-206	I-R-1	pyrimethanil
M-207	I-R-1	tridemorph
M-208	I-R-1	maneb
M-209	I-R-1	metiram
M-210	I-R-1	thiram
M-211	I-R-1	chlorothalonil
M-212	I-R-1	dithianon
M-213	I-R-1	flusulfamide
M-214	I-R-1	metrafenone
M-215	I-R-1	fluxapyroxad
M-216	I-R-1	bixafen
M-217	I-R-1	penflufen
M-218	I-R-1	sedaxane
M-219	I-R-1	isopyrazam
M-220	I-R-1	tricyclazole
M-221	I-R-1	azoxystrobin
M-222	I-R-1	difenoconazole
M-223	I-R-1	kasugamycin
M-224	I-R-1	isoprothiolane
M-225	I-R-1	tolprocarb
M-226	I-R-1	carpropamid
M-227	I-R-1	diclocymet

Mixture	Comp. I	Compound II
M-228	I-R-1	furametpyr
M-229	I-R-1	simeconazole
M-230	I-R-1	probenazole
M-231	I-R-1	mancozeb
M-232	I-R-1	propiconazole
M-233	I-R-1	hexaconazole
M-234	I-R-1	tebuconazole
M-235	I-R-1	carbendazim
M-236	I-R-1	flutolanil
M-237	I-R-1	hymexazol
M-238	I-R-1	isotianil
M-239	I-R-1	orysastrobin
M-240	I-R-1	pencycuron
M-241	I-R-1	phthalide
M-242	I-R-1	pyroquilon
M-243	I-R-1	thifluzamide
M-244	I-R-1	thiophanate
M-245	I-R-1	thiophanate-methyl
M-246	I-R-1	tiadinil
M-247	I-R-1	validamycin
M-248	I-R-1	tebufloquin
M-249	I-R-1	benzovindiflupyr
M-250	I-R-1	picarbutrazox
M-251	I-R-1	pyraziflumid
M-252	I-R-1	dipymetitrone
M-253	I-R-1	pydiflumetofen
M-254	I-R-1	quinofumelin
M-255	I-R-1	ipfentrifluconazole
M-256	I-R-1	dichlobentiazox
M-257	I-R-1	fenpicoxamid
M-258	I-R-1	isoflucypram
M-259	I-R-1	fluindapyr
M-260	I-R-1	inpyrfluxam
M-261	I-R-1	pyrifenamine
M-262	I-R-1	mefentrifluconazole

The mixtures of the present invention may be combined and applied in agriculture in mixture with further active ingredients, for example with other pesticides, insecticides, nematicides, fungicides, herbicides, safeners, fertilizers such as ammonium nitrate, urea, potash, and superphosphate, phytotoxicants and plant growth regulators.

These mixtures are also embraced by the term “mixture(s) of the present invention” or “mixture(s) according to the invention”.

These additional ingredients may be used sequentially or in combination with the mixtures of the invention, if appropriate also added only immediately prior to use (tank mix). For example, the plant(s) may be sprayed with a mixture of this invention either before or after being treated with other active ingredients.

Mixing partners can be selected from pesticides, in particular insecticides, nematicides, and acaricides, fungicides, herbicides, plant growth regulators, fertilizers, and the like. Preferred mixing partners are insecticides, nematicides and fungicides.

In one embodiment, the invention relates to ternary mixtures, comprising a compound I, a compound II and one further compound III, which is not identical to the compound I or II already present in the mixture.

In a sub-embodiment, the invention relates to a mixture of

(1) a compound of formula (I), preferably I-R-1, and

(2) a compound selected from the group of imidacloprid, clothianidin, dinotefuran, chlorantraniliprole, cyantraniliprole, spinetoram, spinosad, ethiprole, fipronil, triflumezopyrim, flonicamid and tetraniliprole, and

(3) a compound selected from the group of probenazole, isotianil, tricyclazole, pyroquilon, isoprothiolane, tolprocarb, carpropamid, diclocymet, azoxystrobin, orysastrobin.

In one embodiment, the invention relates to 4-way mixtures, comprising a compound I, a compound II and two further compounds III, which are not identical to the compound I or II already present in the mixture.

In a sub-embodiment, the invention relates to a mixture of

(1) a compound of formula (I), preferably I-R-1, and

(2) a compound selected from the group of imidacloprid, clothianidin, dinotefuran, chlorantraniliprole, cyantraniliprole, spinetoram, spinosad, ethiprole, fipronil, triflumezopyrim, flonicamid and tetraniliprole, and

(3) a compound selected from the group of probenazole, isotianil, tricyclazole, pyroquilon, isoprothiolane, tolprocarb, carpropamid, diclocymet, azoxystrobin, orysastrobin, and

(4) a compound selected from the group of furametpyr, thifluzamide, simeconazole, penflufen, azoxystrobin, orysastrobin, provided it is different from the compound under (3).

In one embodiment, the invention relates to 5-way mixtures, comprising a compound I, a compound II and three further compounds III, which are not identical to the compound I or II already present in the mixture.

The invention also relates to methods, wherein the compounds of formula (I) are provided or applied in agrochemical compositions comprising an auxiliary and compound of formula (I) or a mixture thereof.

An agrochemical composition comprises a pesticidally effective amount of a compound of the present invention or a mixture thereof. The term "pesticidally effective amount" is defined below.

The compounds of the present invention or the mixtures thereof can be converted into customary types of agro-chemical compositions, e. g. solutions, emulsions, suspensions, dusts, powders, pastes, granules, pressings, capsules, and mixtures thereof. Examples for composition types are suspensions (e.g. SC, OD, FS), emulsifiable concentrates (e.g. EC), emulsions (e.g. EW, EO, ES, ME), capsules (e.g. CS, ZC), pastes, pastilles, wettable powders or dusts (e.g. WP, SP, WS, DP, DS), pressings (e.g. BR, TB, DT), granules (e.g. WG, SG, GR, FG, GG, MG), insecticidal articles (e.g. LN), as well as gel formulations for the treatment of plant propagation materials such as seeds (e.g. GF). These and further compositions types are defined in the “Catalogue of pesticide formulation types and international coding system”, Technical Monograph No. 2, 6th Ed. May 2008, CropLife International.

The compositions are prepared in a known manner, such as described by Mollet and Grubemann, Formulation technology, Wiley VCH, Weinheim, 2001; or Knowles, New developments in crop protection product formulation, Agrow Reports DS243, T&F Informa, London, 2005.

Examples for suitable auxiliaries are solvents, liquid carriers, solid carriers or fillers, surfactants, dispersants, emulsifiers, wetters, adjuvants, solubilizers, penetration enhancers, protective colloids, adhesion agents, thickeners, humectants, repellents, attractants, feeding stimulants, compatibilizers, bactericides, anti-freezing agents, anti-foaming agents, colorants, tackifiers and binders.

Suitable solvents and liquid carriers are water and organic solvents, such as mineral oil fractions of medium to high boiling point, e.g. kerosene, diesel oil; oils of vegetable or animal origin; aliphatic, cyclic and aromatic hydrocarbons, e. g. toluene, paraffin, tetrahydronaphthalene, alkylated naphthalenes; alcohols, e.g. ethanol, propanol, butanol, benzylalcohol, cyclo¬hexanol; glycols; DMSO; ketones, e.g. cyclohexanone; esters, e.g. lactates, carbonates, fatty acid esters, gamma-butyrolactone; fatty acids; phosphonates; amines; amides, e.g. N-methylpyrrolidone, fatty acid dimethylamides; and mixtures thereof.

Suitable solid carriers or fillers are mineral earths, e.g. silicates, silica gels, talc, kaolins, limestone, lime, chalk, clays, dolomite, diatomaceous earth, bentonite, calcium sulfate, magnesium sulfate, magnesium oxide; polysaccharide powders, e.g. cellulose, starch; fertilizers, e.g. ammonium sulfate, ammonium phosphate, ammonium nitrate, ureas; products of vegetable origin, e.g. cereal meal, tree bark meal, wood meal, nutshell meal, and mixtures thereof.

Suitable surfactants are surface-active compounds, such as anionic, cationic, nonionic and amphoteric surfactants, block polymers, polyelectrolytes, and mixtures thereof. Such surfactants can be used as emusifier, dispersant, solubilizer, wetter, penetration enhancer, protective colloid, or adjuvant. Examples of surfactants are listed in McCutcheon’s, Vol.1: Emulsifiers & Detergents, McCutcheon’s Directories, Glen Rock, USA, 2008 (International Ed. or North American Ed.).

Suitable anionic surfactants are alkali, alkaline earth or ammonium salts of sulfonates, sulfates, phosphates, carboxylates, and mixtures thereof. Examples of sulfonates are alkylaryl-sulfonates, diphenylsulfonates, alpha-olefin sulfonates, lignine sulfonates, sulfonates of fatty acids and oils, sulfonates of ethoxylated alkylphenols, sulfonates of alkoxylated arylphenols,

sulfonates of condensed naphthalenes, sulfonates of dodecyl- and tridecylbenzenes, sulfonates of naphthalenes and alkyl¬naphthalenes, sulfosuccinates or sulfosuccinamates. Examples of sulfates are sulfates of fatty acids and oils, of ethoxylated alkylphenols, of alcohols, of ethoxylated alcohols, or of fatty acid esters. Examples of phosphates are phosphate esters. Examples of carboxylates are alkyl carboxylates, and carboxylated alcohol or alkylphenol eth-oxylates.

Suitable nonionic surfactants are alkoxylates, N-subsituted fatty acid amides, amine oxides, esters, sugar-based surfactants, polymeric surfactants, and mixtures thereof. Examples of alkoxylates are compounds such as alcohols, alkylphenols, amines, amides, arylphenols, fatty acids or fatty acid esters which have been alkoxylated with 1 to 50 equivalents. Ethylene oxide and/or propylene oxide may be employed for the alkoxylation, preferably ethylene oxide. Examples of N-subsititued fatty acid amides are fatty acid glucamides or fatty acid alkanolamides. Examples of esters are fatty acid esters, glycerol esters or monoglycerides. Examples of sugar-based surfactants are sorbitans, ethoxylated sorbitans, sucrose and glucose esters or alkylpolyglucosides. Examples of polymeric surfactants are homo- or copolymers of vinylpyrrolidone, vinylalcohols, or vinylacetate.

Suitable cationic surfactants are quaternary surfactants, for example quaternary ammonium compounds with one or two hydrophobic groups, or salts of long-chain primary amines. Suitable amphoteric surfactants are alkylbetains and imidazolines. Suitable block polymers are block polymers of the A-B or A-B-A type comprising blocks of polyethylene oxide and polypropylene oxide, or of the A-B-C type comprising alkanol, polyethylene oxide and polypropylene oxide. Suitable polyelectrolytes are polyacids or polybases. Examples of polyacids are alkali salts of polyacrylic acid or polyacid comb polymers. Examples of polybases are polyvinylamines or polyethyleneamines.

Suitable adjuvants are compounds, which have a neglectable or even no pesticidal activity themselves, and which improve the biological performance of the compounds of the present invention on the target. Examples are surfactants, mineral or vegetable oils, and other auxilaries. Further examples are listed by Knowles, Adjuvants and additives, Agrow Reports DS256, T&F Informa UK, 2006, chapter 5.

Suitable thickeners are polysaccharides (e.g. xanthan gum, carboxymethylcellulose), anorganic clays (organically modified or unmodified), polycarboxylates, and silicates.

Suitable bactericides are bronopol and isothiazolinone derivatives such as alkylisothiazolinones and benzisothiazolinones.

Suitable anti-freezing agents are ethylene glycol, propylene glycol, urea and glycerin.

Suitable anti-foaming agents are silicones, long chain alcohols, and salts of fatty acids.

Suitable colorants (e.g. in red, blue, or green) are pigments of low water solubility and water-soluble dyes. Examples are inorganic colorants (e.g. iron oxide, titan oxide, iron hexacyanoferrate) and organic colorants (e.g. alizarin-, azo- and phthalocyanine colorants).

Suitable tackifiers or binders are polyvinylpyrrolidons, polyvinylacetates, polyvinyl alcohols, polyacrylates, biological or synthetic waxes, and cellulose ethers.

Examples for composition types and their preparation are:

i) Water-soluble concentrates (SL, LS)

10-60 wt% of a compound I according to the invention and 5-15 wt% wetting agent (e.g. alcohol alkoxylates) are dissolved in water and/or in a water-soluble solvent (e.g. alcohols) up to 100 wt%. The active substance dissolves upon dilution with water.

ii) Dispersible concentrates (DC)

5-25 wt% of a compound I according to the invention and 1-10 wt% dispersant (e. g. polyvinylpyrrolidone) are dissolved in up to 100 wt% organic solvent (e.g. cyclohexanone). Dilution with water gives a dispersion.

iii) Emulsifiable concentrates (EC)

15-70 wt% of a compound I according to the invention and 5-10 wt% emulsifiers (e.g. calcium dodecylbenzenesulfonate and castor oil ethoxylate) are dissolved in up to 100 wt% water-insoluble organic solvent (e.g. aromatic hydrocarbon). Dilution with water gives an emulsion.

iv) Emulsions (EW, EO, ES)

5-40 wt% of a compound I according to the invention and 1-10 wt% emulsifiers (e.g. calcium dodecylbenzenesulfonate and castor oil ethoxylate) are dissolved in 20-40 wt% water-insoluble organic solvent (e.g. aromatic hydrocarbon). This mixture is introduced into up to 100 wt% water by means of an emulsifying machine and made into a homogeneous emulsion. Dilution with water gives an emulsion.

v) Suspensions (SC, OD, FS)

In an agitated ball mill, 20-60 wt% of a compound I according to the invention are comminuted with addition of 2-10 wt% dispersants and wetting agents (e.g. sodium lignosulfonate and alcohol ethoxylate), 0,1-2 wt% thickener (e.g. xanthan gum) and up to 100 wt% water to give a fine active substance suspension. Dilution with water gives a stable suspension of the active substance. For FS type composition up to 40 wt% binder (e.g. polyvinylalcohol) is added.

vi) Water-dispersible granules and water-soluble granules (WG, SG)

50-80 wt% of a compound I according to the invention are ground finely with addition of up to 100 wt% dispersants and wetting agents (e.g. sodium lignosulfonate and alcohol ethoxylate) and prepared as water-dispersible or water-soluble granules by means of technical appliances (e. g. extrusion, spray tower, fluidized bed). Dilution with water gives a stable dispersion or solution of the active substance.

vii) Water-dispersible powders and water-soluble powders (WP, SP, WS)

50-80 wt% of a compound I according to the invention are ground in a rotor-stator mill with addition of 1-5 wt% dispersants (e.g. sodium lignosulfonate), 1-3 wt% wetting agents (e.g. alcohol ethoxylate) and up to 100 wt% solid carrier, e.g. silica gel. Dilution with water gives a stable dispersion or solution of the active substance.

viii) Gel (GW, GF)

In an agitated ball mill, 5-25 wt% of a compound I according to the invention are comminuted with addition of 3-10 wt% dispersants (e.g. sodium lignosulfonate), 1-5 wt% thickener (e.g. carboxymethylcellulose) and up to 100 wt% water to give a fine suspension of the active substance. Dilution with water gives a stable suspension of the active substance.

ix) Microemulsion (ME) or nano-emulsion

5-20 wt% of a compound I according to the invention are added to 5-30 wt% organic solvent

blend (e.g. fatty acid dimethylamide and cyclohexanone), 10-25 wt% surfactant blend (e.g. alkohol ethoxylate and arylphenol ethoxylate), and water up to 100 %. This mixture is stirred for 1 h to produce spontaneously a thermodynamically stable microemulsion.

x) Microcapsules (CS)

An oil phase comprising 5-50 wt% of a compound I according to the invention, 0-40 wt% water insoluble organic solvent (e.g. aromatic hydrocarbon), 2-15 wt% acrylic monomers (e.g. methylmethacrylate, methacrylic acid and a di- or triacrylate) are dispersed into an aqueous solution of a protective colloid (e.g. polyvinyl alcohol). Radical polymerization initiated by a radi-cal initiator results in the formation of poly(meth)acrylate microcapsules. Alternatively, an oil phase comprising 5-50 wt% of a compound I according to the invention, 0-40 wt% water insolu-ble organic solvent (e.g. aromatic hydrocarbon), and an isocyanate monomer (e.g. diphenylme-thene-4,4'-diisocyanatae) are dispersed into an aqueous solution of a protective colloid (e.g. polyvinyl alcohol). The addition of a polyamine (e.g. hexamethylenediamine) results in the for-mation of a polyurea microcapsule. The monomers amount to 1-10 wt%. The wt% relate to the total CS composition.

xi) Dustable powders (DP, DS)

1-10 wt% of a compound I according to the invention are ground finely and mixed intimately with up to 100 wt% solid carrier, e.g. finely divided kaolin.

xii) Granules (GR, FG)

0.5-30 wt% of a compound I according to the invention is ground finely and associated with up to 100 wt% solid carrier (e.g. silicate). Granulation is achieved by extrusion, spray-drying or the fluidized bed.

xiii) Ultra-low volume liquids (UL)

1-50 wt% of a compound I according to the invention are dissolved in up to 100 wt% organic solvent, e.g. aromatic hydrocarbon.

The compositions types i) to xiii) may optionally comprise further auxiliaries, such as 0.1-1 wt% bactericides, 5-15 wt% anti-freezing agents, 0.1-1 wt% anti-foaming agents, and 0.1-1 wt% colorants.

The agrochemical compositions generally comprise between 0.01 and 95%, preferably between 0.1 and 90%, and most preferably between 0.5 and 75%, by weight of active sub-stance. The active substances are employed in a purity of from 90% to 100%, preferably from 95% to 100% (according to NMR spectrum).

Various types of oils, wetters, adjuvants, fertilizer, or micronutrients, and other pesticides (e.g. herbicides, insecticides, fungicides, growth regulators, safeners) may be added to the active substances or the compositions com¬prising them as premix or, if appropriate not until immediately prior to use (tank mix). These agents can be admixed with the compositions according to the invention in a weight ratio of 1:100 to 100:1, preferably 1:10 to 10:1.

In one embodiment, a suspoconcentration (SC) is preferred for the application in crop protection. In one sub-embodiment thereof, the SC agrochemical composition comprises between 50 to 500 g/L (grams per Litre), or between 100 and 250 g/L, or 100 g/L or 150g/L or 200g/L or 250 g/L.

In a further embodiment, the granules according to formulation type xii are used for the application in rice according to the present invention.

In a further embodiment, the dispersible concentrates DC according to formulation type ii are used for the application in rice according to the present invention.

In a further embodiment, the emulsifiable concentrates EC according to formulation type iii are used for the application in rice according to the present invention. ix. ME).

In a further embodiment, the microemulsions ME according to formulation type ix are used for the application in rice according to the present invention.

In a further embodiment, nano-emulsions are used for the application in rice according to the present invention.

The user applies the composition according to the invention usually from a predosage de-vice, a knapsack sprayer, a spray tank, a spray plane, or an irrigation system. Usually, the agrochemical composition is made up with water, buffer, and/or further auxiliaries to the desired application concentration and the ready-to-use spray liquor or the agrochemical composition according to the invention is thus obtained. Usually, 20 to 2000 liters, preferably 50 to 400 liters, of the ready-to-use spray liquor are applied per hectare of agricultural useful area.

According to one embodiment, individual components of the composition according to the invention such as parts of a kit or parts of a binary or ternary mixture may be mixed by the user himself in a spray tank and further auxiliaries may be added, if appropriate.

In a further embodiment, either individual components of the composition according to the invention or partially premixed components, e. g. components comprising compounds of the present invention and/or mixing partners as defined above, may be mixed by the user in a spray tank and further auxiliaries and additives may be added, if appropriate.

In a further embodiment, either individual components of the composition according to the invention or partially premixed components, e. g. components comprising compounds of the present invention and/or mixing partners as defined above, can be applied jointly (e.g. after tank mix) or consecutively.

The invention also relates to methods, wherein the compounds of formula (I) are suitable for use in protecting rice, rice plants, rice plant propagation materials, such as seeds, or soil or water, in which the rice plants are growing, from attack or infestation by rice pests, especially rice pest invertebrates. Therefore, the present invention also relates to a plant protection methods, which comprises contacting rice, rice plants, rice plant propagation materials, such as seeds, or soil or water, in which the plants are growing, to be protected from attack or infestation by rice pests, especially rice pest invertebrates, with a pesticidally effective amount of a compound of the present invention.

The present invention also relates to a method of combating or controlling rice pests, especially rice pest invertebrates, which comprises contacting the rice pests, especially rice pest invertebrates, their habitat, breeding ground, or food supply, or the rice, rice plants, rice plant propagation materials, such as seeds, or soil or water, or the area, material or environment in which the rice pests, especially rice pest invertebrates, are growing or may grow, with a pesticidally effective amount of a compound of the present invention.

The compounds of the present invention are effective through both contact and ingestion. Furthermore, the compounds of the present invention can be applied to any and all developmental stages, such as egg, larva, pupa, and adult.

The compounds of the present invention can be applied as such or in form of compositions comprising them as defined above. Furthermore, the compounds of the present invention can be applied together with a mixing partner as defined above or in form of compositions comprising said mixtures as defined above. The components of said mixture can be applied simultaneously, jointly or separately, or in succession, that is immediately one after another and thereby creating the mixture “in situ” on the desired location, e.g. the plant, the sequence, in the case of separate application, generally not having any effect on the result of the control measures.

The application can be carried out both before and after the infestation of the rice, rice plants, rice plant propagation materials, such as seeds, soil, or the area, material or environment by the pests.

Suitable application methods include inter alia soil treatment, seed treatment, in furrow application, water inlet application and foliar application. Soil treatment methods include drenching the soil, dipping roots, or soil injection. Seed treatment techniques include seed dressing, seed coating, seed dusting, seed soaking, and seed pelleting. In furrow applications typically include the steps of making a furrow in cultivated land, seeding the furrow with seeds, applying the pesticidally active compound to the furrow, and closing the furrow. Foliar application refers to the application of the pesticidally active compound to plant foliage, e.g. through spray equipment. For foliar applications, it can be advantageous to modify the behavior of the pests by use of pheromones in combination with the compounds of the present invention. Suitable pheromones for specific crops and pests are known to a skilled person and publicly available from databases of pheromones and semiochemicals, such as http://www.pherobase.com.

In the context of rice cultivation and rice crops, the following application types are of special relevance:

— “Granular application” involves manual or mechanical scattering or throwing of insecticide granules or mixtures of insecticides/fungicides and nematicides, directly into a field or nursery box, either on the surface of the soil or on standing water. The granular formulation may be mixed with a filler, carrier or fertilizer to allow for uniform distribution in the field.

— “Floating packet application” refers to the application of an insecticide or mixtures of insecticides/fungicides and nematicides in a water soluble sachet/packet by throwing into the paddy field in standing water.

— “Seedling box application” refers to manual or mechanical incorporation of insecticide formulations (for eg. Granules, liquid) in nursery boxes or seedling boxes containing rice seedlings before being transplanted into the main field.

— “Seed treatment” involves the soaking/mixing of rice seeds in a solution of an insecticide or insecticide/nematicide/fungicide mixture. This application is carried out before sowing, either before or after seed germination.

— "Foliar application" refers to application of an insecticide or an insecticide/fungicide/nematicide/selective herbicides in water or oil as a spray application using various application equipment (eg. knapsack, power sprayer, boom sprayer, etc).

— "Soil application" refers to the application of an insecticide or a mixture of an insecticide/fungicide/nematicide/selective herbicide into the soil either as drench application, water inlet application or as a granular application.

— "Aerial application" refers to the application of a granular or liquid application of an insecticide or a mixture of an insecticide/fungicide/nematicide/selective herbicide to the field using aeroplanes, helicopters or drones.

— "Dust application" involves the directed application of an insecticide or a mixture of an insecticide/fungicide/nematicide/selective herbicide as a dust formulation using specialized applicators (eg. Power dusters) directly into the field.

— "Water inlet application" is the application of a liquid formulation of an insecticide or or a mixture of an insecticide/fungicide/nematicide/selective herbicide at the point where irrigation water is released into the paddy field.

— "Encircling application" is a type of application where a liquid or granular formulation of an insecticide or a mixture of an insecticide/fungicide/nematicide/selective herbicide is applied to standing water, in a clockwork or anti clockwork direction, to the inside borders of a paddy field.

Preferred applications are granular application, seedling box application and foliar application.

In one embodiment, the invention relates to methods, in which the pesticide is applied by granular application.

In one embodiment, the invention relates to methods, in which the pesticide is applied by seedling box application.

In one embodiment, the invention relates to methods, in which the pesticide is applied by foliar application.

As used herein, the term "contacting" or "applying" includes both direct contact (applying the compounds/compositions directly on the animal pest or plant - typically to the foliage, stem or roots of the plant) and indirect contact (applying the compounds/compositions to the locus, i.e. habitat, breeding ground, plant, seed, soil, area, material or environment in which a pest is growing or may grow, of the animal pest or plant).

In the context of the present invention, the term "animal pest" includes arthropods, gastropods, and nematodes, which are rice pests, especially rice pest invertebrates, especially rice pest insects as described above. Arthropods are preferably insects and arachnids, in particular insects. Insects, which are of particular relevance, are typically referred to as crop insect pests or rice pest insects.

The term "crop" refers to both, growing and harvested rice.

In the context of the present invention, the term "plant" means preferably rice plant (Oryza species, preferably Oryza sativa). Two species of rice are most frequently cultivated, Oryza sativa and Oryza glaberrima. Numerous subspecies of Oryza sativa are commercially important including Oryza sativa subsp. indica, Oryza sativa subsp. japonica, Oryza sativa subsp. javanica, Oryza sativa subsp. glutinosa (glutinous rice), Oryza sativa Aromatica group (e.g., basmati),

and Oryza sativa (Floating rice group). The term “plant” is to be understood as including wild type plants and plants, which have been modified by either conventional breeding, or mutagenesis or genetic engineering, or by a combination thereof.

Plants, which have been modified by mutagenesis or genetic engineering, and are of particular commercial importance, include rice. In plants, which have been modified by mutagenesis or genetic engineering, one or more genes have been mutagenized or integrated into the genetic material of the plant. The one or more mutagenized or integrated genes are preferably selected from pat, epsps, cry1Ab, bar, cry1Fa2, cry1Ac, cry34Ab1, cry35AB1, cry3A, cryF, cry1F, mcry3a, cry2Ab2, cry3Bb1, cry1A.105, dfr, barnase, vip3Aa20, barstar, als, bxn, bp40, asn1, and ppo5. The mutagenesis or integration of the one or more genes is performed in order to improve certain properties of the plant. Such properties, also known as traits, include abiotic stress tolerance, altered growth/yield, disease resistance, herbicide tolerance, insect resistance, modified product quality, and pollination control. Of these properties, herbicide tolerance, e.g. imidazolinone tolerance, glyphosate tolerance, or glufosinate tolerance, is of particular importance.

It has surprisingly been found that the pesticidal activity of the compounds of the present invention may be enhanced by the insecticidal trait of a modified plant. Furthermore, it has been found that the compounds of the present invention are suitable for preventing insects to become resistant to the insecticidal trait or for combating pests, which already have become resistant to the insecticidal trait of a modified plant. Moreover, the compounds of the present invention are suitable for combating pests, against which the insecticidal trait is not effective, so that a complementary insecticidal activity can advantageously be used.

The term "plant propagation material" refers to all the generative parts of the plant such as seeds, sprouted seeds, seedlings and ratooning. Seedlings and young plants, which are to be transplanted after germination or after emergence from soil, may also be included. These plant propagation materials may be treated prophylactically with a plant protection compound either at or before planting or transplanting.

The term “seed” embraces seeds and plant propagules of all kinds including but not limited to true seeds, seed pieces, suckers, corms, fruit, grains, cuttings, cut shoots and the like, and means in a preferred embodiment true seeds.

In general, "pesticidally effective amount" means the amount of active ingredient needed to achieve an observable effect on growth, including the effects of necrosis, death, retardation, prevention, and removal, destruction, or otherwise diminishing the occurrence and activity of the target organism. The pesticidally effective amount can vary for the various compounds/compositions used in the invention. A pesticidally effective amount of the compositions will also vary according to the prevailing conditions such as desired pesticidal effect and duration, weather, target species, locus, mode of application, and the like.

In the case of soil treatment, in furrow application or of application to the pests dwelling place or nest, the quantity of active ingredient ranges from 0.0001 to 500 g per 100 m^2, preferably from 0.001 to 20 g per 100 m^2.

For use in treating rice plants, e.g. by foliar application, the rate of application of the active ingredients of this invention may be in the range of 0.0001 g to 4000 g per hectare, e.g. from 1 g to 2 kg per hectare or from 1 g to 750 g per hectare, desirably from 1 g to 100 g per hectare,

more desirably from 10 g to 50 g per hectare, e.g., 10 to 20 g per hectare, 20 to 30 g per hectare, 30 to 40 g per hectare, or 40 to 50 g per hectare.

The compounds of the present invention are particularly suitable for use in the treatment of seeds in order to protect the seeds from insect pests, in particular from soil-living insect pests, and the resulting seedling's roots and shoots against soil pests and foliar insects. The present invention therefore also relates to a method for the protection of seeds from insects, in particular from soil insects, and of the seedling's roots and shoots from insects, in particular from soil and foliar insects, said method comprising treating the seeds before sowing and/or after pregermination with a compound of the present invention. The protection of the seedling's roots and shoots is preferred. More preferred is the protection of seedling's shoots from piercing and sucking insects, chewing insects and nematodes.

The term "seed treatment" comprises all suitable seed treatment techniques known in the art, such as seed dressing, seed coating, seed dusting, seed soaking, seed pelleting, and in-furrow application methods. Preferably, the seed treatment application of the active compound is carried out by spraying or by dusting the seeds before sowing of the plants and before emergence of the plants.

The present invention also comprises seeds coated with or containing the active compound. The term "coated with and/or containing" generally signifies that the active ingredient is for the most part on the surface of the propagation product at the time of application, although a greater or lesser part of the ingredient may penetrate into the propagation product, depending on the method of application. When the said propagation product is (re)planted, it may absorb the active ingredient.

In the context of the present invention, the seed is seed of rice. The active compounds of the invention may also be used for the treatment of seeds from plants, which have been modified by mutagenisis or genetic engineering, and which e.g. tolerate the action of herbicides or fungicides or insecticides. Such modified plants have been described in detail above.

Conventional seed treatment formulations include for example flowable concentrates FS, solutions LS, suspoemulsions (SE), powders for dry treatment DS, water dispersible powders for slurry treatment WS, water-soluble powders SS and emulsion ES and EC and gel formulation GF. These formulations can be applied to the seed diluted or undiluted. Application to the seeds is carried out before sowing, either directly on the seeds or after having pregerminated the latter. Preferably, the formulations are applied such that germination is not included.

The active substance concentrations in ready-to-use formulations, which may be obtained after two-to-tenfold dilution, are preferably from 0.01 to 60% by weight, more preferably from 0.1 to 40 % by weight.

In a preferred embodiment a FS formulation is used for seed treatment. Typically, a FS formulation may comprise 1-800 g/l of active ingredient, 1-200 g/l Surfactant, 0 to 200 g/l antifreezing agent, 0 to 400 g/l of binder, 0 to 200 g/l of a pigment and up to 1 liter of a solvent, preferably water.

Especially preferred FS formulations of the compounds of the present invention for seed treatment usually comprise from 0.1 to 80% by weight (1 to 800 g/l) of the active ingredient, from 0.1 to 20 % by weight (1 to 200 g/l) of at least one surfactant, e.g. 0.05 to 5 % by weight of a wetter

and from 0.5 to 15 % by weight of a dispersing agent, up to 20 % by weight, e.g. from 5 to 20 % of an anti-freeze agent, from 0 to 15 % by weight, e.g. 1 to 15 % by weight of a pigment and/or a dye, from 0 to 40 % by weight, e.g. 1 to 40 % by weight of a binder (sticker /adhesion agent), optionally up to 5 % by weight, e.g. from 0.1 to 5 % by weight of a thickener, optionally from 0.1 to 2 % of an anti-foam agent, and optionally a preservative such as a biocide, antioxidant or the like, e.g. in an amount from 0.01 to 1 % by weight and a filler/vehicle up to 100 % by weight.

In the treatment of seed, the application rates of the compounds of the invention are generally from 0.1 g to 10 kg per 100 kg of seed, preferably from 1 g to 5 kg per 100 kg of seed, more preferably from 1 g to 1000 g per 100 kg of seed and in particular from 1 g to 200 g per 100 kg of seed, e.g. from 1 g to 100 g or from 5 g to 100 g per 100 kg of seed.

The invention therefore also relates to seed comprising a compound of the present invention, or an agriculturally useful salt thereof, as defined herein. The amount of the compound of the present invention or the agriculturally useful salt thereof will in general vary from 0.1 g to 10 kg per 100 kg of seed, preferably from 1 g to 5 kg per 100 kg of seed, in particular from 1 g to 1000 g per 100 kg of seed.

In the present invention, the compounds of the present invention may also be used for improving the health of a plant. Therefore, the present invention also relates to a method for improving plant health by treating a rice plant, rice plant propagation material and/or the locus where the rice plant is growing or is to grow with an effective and non-phytotoxic amount of a compound of the present invention.

As used herein “an effective and non-phytotoxic amount” means that the compound is used in a quantity which allows to obtain the desired effect but which does not give rise to any phytotoxic symptom on the treated plant or on the plant grown from the treated propagule or treated soil.

The terms “plant” and “plant propagation material” are defined above.

"Plant health" is defined as a condition of the plant and/or its products which is determined by several aspects alone or in combination with each other such as yield (for example increased biomass and/or increased content of valuable ingredients), quality (for example improved content or composition of certain ingredients or shelf life), plant vigour (for example improved plant growth and/or greener leaves (“greening effect”), tolerance to abiotic (for example drought) and/or biotic stress (for example disease) and production efficiency (for example, harvesting efficiency, processability).

The above identified indicators for the health condition of a plant may be interdependent and may result from each other. Each indicator is defined in the art and can be determined by methods known to a skilled person.

Examples

Compound I [3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate] is synthesized as described in EP17164175.6, and separated into the enantiomers I-R-1 and I-S-1 as described also there.

Step 1: 2-chloro-N-methoxy-N-methyl-acetamide

N-Methoxymethanamine hydrochloride (345g) and water (1.5 L) were cooled to 0°C. To this

reaction mixture, K_2CO_3 (1466g) was added in lots, then methyl tert-butyl ether (1000 mL) was added at 0 °C. The reaction mixture was cooled to -5 °C. Chloroacetylchloride (400 g) in methyl tert-butyl ether (500 ml) was added drop wise at - 5°C to 0°C and stirred for 2 hours at 0°C. The reaction mixture was allowed to come to 20-25°C. From the organic layer, the desired product was obtained as white solid (438g, 90 % yield; 98.45% HPLC purity).

Step 2: 2-chloro-1-(2-chlorothiazol-5-yl) ethanone

2-chlorothiazole (187 ml) in 750 ml tetrahydrofurane under nitrogen atmosphere were cooled to -20 °C. Isopropylmagnesium chloride x LiCl (1684 ml, 1.3 molar in tetrahydrofurane) was added drop wise and stirred at -20°C for 60 minutes. A solution of 2-chloro-N-methoxy-N-methyl-acetamide (250 g) in tetrahydrofurane was added drop wise at -20°C to -25°C. The reaction mixture was stirred at -20 °C for 90 minutes. Saturated aqueous ammonium chloride solution was added at -20°C, then the reaction mixture was brought to 20-25°C. The two phases were separated and the aqueous phase was extracted with ethyl acetate. From the combined organic layers , the desired crude product was obtained as dark brown colored oil, which was treated with activated charcoal and silica in methyl tert-butyl ether to get the crude product as pale brown colored oil (335 g) for direct use in the next step.

Step 3: N-[2-chloro-1-(2-chlorothiazol-5-yl) ethylidene]-2-methyl-propane-2-sulfinamide

To crude 2-chloro-1-(2-chlorothiazol-5-yl) ethanone (335 g) in tetrahydrofurane at 20-25°C under nitrogen atmosphere, tert-butyl sulfinamide (206 g) and $Ti(OEt)_4$ (396 ml) are added. The mixture was heated to 50 °C and stirred for 2 hours, then cooled to 20-25°C and diluted with ethyl acetate. After adding water, the mixture was stirred for 30 minutes, then filtered. The organic phase was evaporated to obtain the desired crude product as brown colored oil. After treatment with activated charcoal and silica in methyl tert-butyl ether, the crude product was obtained as pale brown colored oil (365 g) for direct use in the next step.

Step 4: N-[2-chloro-1-(2-chlorothiazol-5-yl) ethyl]-2-methyl-propane-2-sulfinamide

To N-[2-chloro-1-(2-chlorothiazol-5-yl) ethylidene]-2-methyl-propane-2-sulfinamide (365 g) in tetrahydrofurane and methanole at - 5°C, $NaBH_4$ (23g) was added lot wise and stirred for 30 minutes. Saturated aqueous ammonium chloride solution was added at 0°C. After extracting with ethyl acetate, the organic layer yielded the desired crude product as brown colored oil (310g).

Step 5: 2-chloro-1-(2-chlorothiazol-5-yl) ethanamine hydrochloride

N-[2-chloro-1-(2-chlorothiazol-5-yl) ethyl]-2-methyl-propane-2-sulfinamide was stirred with HCl in methanole (1 molar, 620 mL) at 20-25°C for 12 hours. Removal of methanole under vacuum yielded a pale yellow sticky solid (244 g), which was washed with methyl tert-butyl ether and subsequently with ethyl acetate to get a pale yellow color solid (78 g, 26 % yield over steps 2 to 5, >98% purity).

Step 6: 4-(2-chlorothiazol-5-yl)-N-methyl-thiazolidin-2-imine

2-chloro-1-(2-chlorothiazol-5-yl) ethanamine hydrochloride (285 g) in methyl tert-butyl ether and 2 molar aqueous NaOH solution (1060 mL) were stirred for 20 minutes at 23 °C. The organic layer yielded the free amine as pale brown colored oil (230 g).

The amine (230 g) in ethanole was reacted with triethylamine NEt3 (351 ml) and Me-NCS (143.2 g) at 22 to 25°C for 18 hours. The reaction mass was concentrated to obtain a brown colored residue, to which aqueous NaOH solution (114 g in 920 mL of water) was added. The resulting mixture was heated to 100 °C for 2 hours, then cooled to 20-25°C and diluted with water. After extraction with ethyl acetate, the organic layer yielded crude 4-(2-chlorothiazol-5-yl)-N-methyl-thiazolidin-2-imine as brown colored solid (256 g), which was stirred with 20% ethyl acetate in heptane (300 mL) for 30 minutes. After filtering, the product was obtained as a brown colored solid (245 g, 85% yield).

Step 7: 3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate (I-1)

4-(2-chlorothiazol-5-yl)-N-methyl-thiazolidin-2-imine (110 g) in toluene was stirred at 110 to 115°C. After adding bis(4-chlorophenyl) 2-phenylpropanedioate (226 g), the reaction mixture was stirred at this temperature for 2 hours, then cooled to 40 to 45 °C. After removal of toluene under vacuum, a brown solid was obtained, which was triturated with methyl tert-butyl ether to obtain a yellow color solid.

Stirring in methyl tert-butyl ether (1 L) at 22 to 25°C for 14 hours yielded a pale yellow solid (160 g). Further purification by dissolving in dichloromethane and precipitating with methyl tert-butyl ether yielded the desired product as fine pale yellow colored powder (129 g, 80% yield).

Separation of the enantiomers:

R-3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate and

S-3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate

The enantiomers of 3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate from the Example can be separated by preparative chiral supercritical fluid chromatography. 126 g of rac-3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate were separated. This yielded 53.4 g of R-3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate at a retention time of 1.94 min and 57.7 g of S-3-(2-chlorothiazol-5-yl)-8-methyl-7-oxo-6-phenyl-2,3-dihydrothiazolo[3,2-a] pyrimidin-8-ium-5-olate at a retention time of 1.41 min. These retention times refer to the analytical method cited below. The configuration of the chiral center was determined by X-ray analysis.

Analytical separation method:

Instrument: Thar analytical SFC

Column: Chiralpak AS-H, 150×4.6 mm i.d., 5 u

Mobile phase: A for CO_2 and B for MeOH, Gradient: B%=40%

Flow rate: 4.0 mL/min, Back pressure: 100 bar, Column temperature: 35°C

Wavelength: 220 nm

Preparative separation method:

Instrument: Thar 80 preparative SFC

Column: Chiralcel OJ-H, 250×30 mm I.D. 5 u

Mobile phase: A for CO_2 and B for CH_3CN, Gradient: B% = 50%
Flow rate: 80 g /min, Back pressure: 100 bar, Column temperature: 40°C
Wavelength: 220 nm
Cycletime: 6.5 min
Sample preparation: Racemic material was dissolved in mixed solution of MeOH-CH_3CN-DCM (1:1:0.5) to 20 mg/mL and filtrated through membrane with pore sized 0.45um.
Injection: 4 mL per injection.
After separation, the fractions were dried off via rotary evaporator at bath temperature 35°C to get the two enantiomers.
*: HPLC Method: Retention time in minutes; mass charge ratio m/z
HPLC Method A:
MSD4/5: Shimadzu Nexera UHPLC + Shimadzu LCMS 20-20, ESI
Column: Phenomenex Kinetex 1,7µm XB-C18 100A, 50 x 2,1mm
Mobile Phase: A: water + 0,1% trifluoroacetic acid; B: acetonitrile, Temperature: 60°C
Gradient: 5% B to 100% B in 1,50min; 100% B 0,25min
Flow: 0,8ml/min to 1,0ml/min in 1,51 min
MS method: ESI positive, Mass range (m/z): 100-700
HPLC Method B:
MSD4/5: Shimadzu Nexera UHPLC + Shimadzu LCMS 20-20, ESI
Column: Agilent Eclipse Plus C18, 50mm x 4.6mm x 3
Mobile phase: A=10 mM ammonium formate (0.1 % Formic Acid) B= acetonitrile (0.1 % Formic Acid), Flow= 1.2 ml/min. Column oven: 30 C
Gradient:= 10% B to 100% B - 1.5min, hold for 1min, 2.51min - 10% B; Run Time = 3.50 min

Biological test:

B.1 Rice water weevil

Micro plot trial (seedling box application): Before rice seedling transplant, granular formulation of products was manually and evenly applied to the seedling box. Seedlings were transplanted manually in the field plot (simulating commonly-used mechanical transplanting). 2% granular of I was used. Rice water weevil was infested naturally in the field. Feeding damage was estimated at 26 DAT/DAP and was expressed with an average of 10 hills. Larvae was counted with 5 hills/plot. Data was analyzed with Student-Newman-Keuls (P=0.05). Feeding damage for negative control at 26 DAT/DAP was 3.3% and average larvae number of rice water weevil for negative control at 68DAT/DAP was 29.

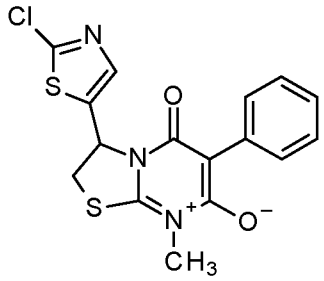
The results (Table 1) showed that the tested compound of formula I showed excellent activity against rice water weevil through granular seedling box application.

Table 1: Efficacy of I-1 against rice water weevil (Lissorhoptrus oryzophilus) by granular seedling box application

Product	rate (g. ai/ha)	% Feeding protection	% larvae control
I-1	200	93[a]	99[a]

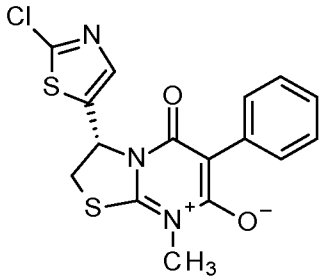
Claims:

1. A method of controlling rice pest invertebrates in rice, which method comprises applying to said rice pest invertebrates a compound of formula (I), a salt or N-oxide thereof, by seedling box application.



(I)

2. The method according to claim 1, wherein the compound of formula I is the compound I-R-1.



(I-R-1)

3. The method according to claim 1 or 2, in which the compound of formula I is applied in mixture with at least one further pesticide.

4. The method according to any of the preceding claims, wherein the rice pest invertebrate is selected from the group of
Hemiptera:
brown planthopper – Nilaparvata lugens
small brown planthopper – Laodelphax striatellus
white-backed planthopper – Sogatella furcifera
white leafhopper – Cofana spectra
green leafhopper – Nephotettix virescens, N. nigriceps, N. cincticeps, N. malayanus
zig zag leafhopper – Recilia dorsalis
maize orange leafhopper – Cicadulina bipunctata
aster leafhopper - Macrosteles fascifrons
rice earhead bug, Leptocorisa oratorius, L. acuta
rice stink bugs – Nezara viridula, Pygomenida varipennis, Eysarcoris, Tibraca limba-triventris, Eysarcoris ventralis
small stink bug - Oebalus poecilus, O. pugnax
coreid bug – Eysarcoris sp
chinch bug - Blissus leucopterus leucopterus

rice mealybug, Brevennia rehi, Pseudococcus saccharicola
rice aphids, Rhopalosiphum rufiabdominalis, Macrosiphum avenae, Hysteroneura setariae, Tetraneuro nigriabdominalis
bean root aphid - Smynthurodes betae

Lepidoptera:
rice skipper – Parnara guttata, Melanitis leda ismene
rice stem borer / striped stem borer – Chilo suppressalis, Chilo polychrusus, Chilo partellus, Chilo plejadellus
rice stalk borer – Chilotraea polychrysa
pink rice borer – Sesamia inferens
yellow rice borer – Tryporyza (=Scirpophaga) incertulas
white rice borer – Tryporyza innotata
rice leafroller / leaf folder – Cnaphalocrocis medinalis, Marasmia patnalis, M. exigua
rice ear-cutting caterpillar / armyworm– Pseudaletia separate
green caterpillar – Xanthodes transversa
green rice caterpillar – Narnaga aenescens
green horned caterpillars - Melanitis leda ismene, Mycalesis sp
fall army worm – Spodoptera frugiperda
cutworm – Mythimna separata
rice case worm- Nymphula depunctalis
black hairy caterpillar, Amata sp.
hairy caterpillar- Mocis frugalis
yellow caterpillar, Psalis pennatula
rice semi-brown looper, Mocis frugalis
rice semi-looper, Chrysodeixis chalcites
grass webworm - Herpetogramma licarsisalis
sugarcane borer - Diatraea saccharalis
corn stalk borer – Elasmopalpus lignosellus
striped grass looper – Mocis latipes
european corn borer – Ostrinia nubilalis
Mexican rice borer – Eoreuma loftini

Coleoptera:
water weevil – Lissorhopterus oryzophilus
rice plant weevil – Echinocnemus squamous
rice weevil - Oryzophagus oryzae
rice hispa – Diclodispa armigera
rice leaf beetle – Oulema oryzae
rice blackbug – Scotinophora vermidulate, S. vermidulate, S. lurida, S. latiuscula
rice flea beetle – Chaetocnima basalis
grubs - Leucopholis irrorata, Leucopholis irrorata, Phyllophaga sp, Heteronychus sp

scarab beetle (bicho torito) - Diloboderus abderus
billbugs - Sphenophorus spp
grape colaspis - Colaspis brunnea, C. louisianae
rice pollen beetle, Chilolaba acuta

Diptera:
stem maggot – Chlorops oryzae
leafminer – Agromyza oryzae
rice whorl maggot / rice stem maggot – Hydrellia sasakii
rice whorl maggot / small rice leafminer – Hydrellia griseola
rice gall midge – Orseolia (=Pachydiplosis) oryzae
rice shoot fly- Atherigona oryzae
rice seed midge – Chironomus cavazzai, Chironomus spp, Cricotopus spp

Thysanoptera:
rice thrips- Chloethrips oryzae, Stenochaetothrips biformis, Perrisothrips sp., Hoplothrips sp.,

Orthoptera:
rice grasshoppers, Hieroglyphus banian, Hieroglyphus nigrorepletus, Catantops pinguis, Attractomorpha burri, A. crenulate, A. psittacina psittacina, A. Bedeli, Oxya adenttata, Oxya ebneri, Oxya hyla intricata, Acrida turricata
locusts – Locusta migratoria manilensis
mole cricket, Grylotalpa africana
field cricket: Gryllus bimaculatus, Teleogryllus occipitalis, Euscyrtus concinus
katydid – Conocephalus longipennis

Isoptera:
termites – Macrotermes gilvus, Syntermes molestans

Hymenoptera:
ants – Solenopsis geminata
rice white tip nematode – Aphelenchoides besseyi

Acari:
rice panicle mite - Steotarsonemus pinki

Crustacea:
tadpole shrimp - Triops longicaudatus, T. cancriformis;
rice crayfish - Procambarus clarkii, Orconectes virilis.

5. The method according to any of the preceding claims, wherein the rice pest invertebrate is from the order Hemiptera, Lepidoptera, or Coleoptera;
preferably selected from hoppers,
or preferably selected from brown planthopper (Nilaparvata lugens), small brown planthopper (Laodelphax striatellus), white-backed planthopper (Sogatella furcifera), green leafhopper (Nephotettix virescens), rice stink bugs (Nezara viridula, Pygomenida varipennis, Eysarcoris, Tibraca limbatriventris, Eysarcoris ventralis), small stink bug (Oebalus poecilus, O. pugnax), rice stem borer (Chilo suppressalis), yellow rice borer (Tryporyza (=Scirpophaga) incertulas); rice leafroller / leaf folder (Cnaphalocrocis medinalis, Marasmia patnalis, M. exigua), water weevil (Lissorhopterus oryzophilus), rice weevil (Oryzophagus oryzae).

6. The method according to any of the preceding claims, wherein the pests are selected from stem borers (Chilo spp.), plant hoppers (Nilparvata lugens), weevils (Lissorhoptrus oryzophilus), nematodes Aphelencoides besseyi, and mining flies Hydrellia philiippina.

7. A method for increasing the health of rice plants, especially in paddy rice fields, comprising applying the compound of formula (I) as defined in any of claims 1 to 2 or a mixture as defined in claim 3 by seedling box application.

8. A method for increasing the yield of rice plants, comprising applying the compound of formula (I) as defined in any of claims 1 to 2 or a mixture as defined in claim 3 by seedling box application.

9. Seedling box application method for protecting rice plants, using a compound of formula (I) as defined in any of claims 1 to 2, or a mixture as defined in claim 3.

10. Seedling box application method of controlling rice pest invertebrates in rice, which method comprises applying to said rice pest invertebrates a compound of formula (I) as defined in any of claims 1 to 2, or a mixture as defined in claim 3.

11. The method according to any of the preceding claims, wherein the compound of formula (I) is applied in a granular formulation.

12. A seeding box comprising a compound of formula (I) as defined in any of claims 1 to 2, or a mixture as defined in claim 3, and rice seed.

13. A seeding box according to claim 12, wherein the amount of a compound of formula (I) impregnated into the rice seeds is between about 3 and 200 g/q.

14. A nursery box comprising a growth substrate for rice, rice seeds and an aqueous formulation of a compound of formula (I) as defined in any of claims 1 to 2, or a mixture as defined in claim 3, and rice seed.

15. A seedling box according to claims 11 or 12, or a nursery box according to claim 14, wherein the compound of formula (I) is applied in a granular formulation.

16. Use of the seeding box according to claim 11 or 12 for controlling rice pest invertebrates in rice.

17. Use of the nursery box according to claim 14 or 15 for controlling rice pest invertebrates in rice.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2018/073516

A. CLASSIFICATION OF SUBJECT MATTER
INV. A01N43/90 A01P7/04
ADD.

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)
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 practicable, search terms used)
EPO-Internal, CHEM ABS Data, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CALEB W HOLYOKE ET AL: "Mesoionic insecticides:a novel class of insecticides that modulate nicotinic acetylcholine receptors", PEST MANAG SCI, vol. 73, 1 February 2017 (2017-02-01), pages 796-806, XP002775069, the whole document -----	1-17
X	WO 2014/167084 A1 (BASF SE [DE]) 16 October 2014 (2014-10-16) cited in the application	1-17
Y	page 25 - page 31; compounds III-4, -12, -20, -28 table 12 page 93 page 47, line 32 - line 33 -----	1-17
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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

* Special categories of cited documents :

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"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

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 September 2018	27/09/2018

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, Fax: (+31-70) 340-3016	Breimaier, Waltraud

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Form PCT/ISA/210 (second sheet) (April 2005)

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 2016/055431 A1 (BASF SE) 14 April 2016 (2016-04-14) page 81, line 8 - line 24; compounds I-ex page 77; example 1; compounds C-1 page 25 - page 32; compounds III-4, -12, -20, -28, -36 page 57, line 25	1-17
A	WO 2012/092115 A1 (DU PONT [US]; PAHUTSKI THOMAS FRANCIS JR [US]) 5 July 2012 (2012-07-05) the whole document	1-17

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2018/073516

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