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(71) Applicant (for all designated States except US): **BAYER CROPSCIENCE AG** [DE/DE]; Alfred-Nobel-Str. 50, 40789 Monheim (DE).

(72) Inventor; and

(75) Inventor/Applicant (for US only): **SAKUMA, Haruhiko** [JP/JP]; 4-29-19, Higashijonan, Oyama-shi, Tochigi 323-0829 (JP).

(74) Common Representative: **BAYER CROPSCIENCE**

AG; Business Planning and Administration., Law and Patents, Patents and Licensing, Building 6100, Alfred-Nobel-Str. 50, 40789 Monheim (DE).

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(54) Title: FUNGICIDAL AND INSECTICIDAL COMPOSITION

(57) Abstract: A fungicidal and insecticidal composition comprising, as a combination: (a) 3,4-dichloro-N-(2-cyanophenyl)isothiazole-5-carboxamide and (b) fipronil or ethiprole.



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Fungicidal and Insecticidal Composition

Technical Field

The present invention relates to fungicidal and insecticidal compositions. More particularly, the present invention relates to fungicidal and insecticidal compositions to be applied to nursery boxes, and also relates to the compositions further having plant growth-promoting activity as a secondary effect.

Background Art

A method of raising paddy rice seedlings in nursery boxes involving treating the seedlings during their sowing with a long-lasting insecticide and fungicide, has been widely used and popular all over Japan as effective means to save labor and reduce the cost for paddy rice culture, since it allows reduced frequency of insects/plant diseases control work during growth of the rice plants in the field.

Each of the active ingredients contained in the compositions of the invention, although it does not affect the growth of rice under usual conditions when it is used alone for treating seeds at sowing, may cause some delay of the root growth depending on the conditions such as irrigated nursing (commonly called "pool nursing"), abnormally high temperature, low temperature, dryness and the like.

Mixed compositions containing 3,4-dichloro-N-(2-cyanophenyl)isothiazole-5-carboxamide as fungicides and known insecticides are already known (WO 2005/009131).

The inventors have found that the combinations shown below exhibit excellent efficacy in controlling paddy rice insects/plant diseases after transplantation of the paddy rice to the field, as well as in accelerating root growth resulting in shortened growth duration of the seedlings, and allowing stable growth of good root of the paddy rice seedlings even under unusual conditions, and thereby completed the invention.

Thus, the present invention provides fungicidal and insecticidal compositions comprising, as a combination,

(a) 3,4-dichloro-N-(2-cyanophenyl)isothiazole-5-carboxamide (hereinafter abbreviate to isotianil) and

(b) fipronil or ethiprole.

The invention also provides a method of controlling insects/plant diseases after transplantation of

paddy rice seedlings to the field comprising: sowing seeds of the paddy rice in nursery boxes and applying the above compositions to the seeds before covering them with soil, and a method of controlling insects/plant diseases after transplantation of paddy rice seedlings to the field comprising: mixing seedbed soil for nursery boxes or covering soil for seeds of the paddy rice with the same composition as above.

According to the invention, no publication before the application date of this application discloses the description specifying the compositions comprising the combination of above (a) isotianil and (b) fipronil or ethiprole, as well as, the specific biological effects of the compositions. So far in WO 2005/009131 fipronil and ethiprole have been disclosed within a generic formula only as possible mixing partners for compound (A) within a long list of mixing partners. In addition, these mixtures have only described in WO 2005/009131 for having very good fungicidal activity.

Surprisingly, the fungicidal and insecticidal compositions of the invention comprising, as a combination, (a) isotianil and (b) fipronil or ethiprole exhibit excellent efficacy in controlling paddy rice insects, as well as exert plant growth-promoting activity providing excellent promotion of root growth of paddy rice seedlings compared to the case without application of the compositions. As a result, growth duration of seedlings is shortened and stable root growth of paddy rice seedlings is attained even under unusual conditions. Thus, the compositions of the invention are significantly advantageous and useful for both insects/plant diseases control and productivity involving shortened growth period of seedlings.

The present invention also provides compositions further comprising, in addition to the combination of the above two ingredients (a) and (b), (c) at least one substance selected from the group consisting of imidacloprid, thiacloprid, clothianidin, dinotefuran, thiamethoxam, spinosad, thifluzamide, furametpyr, orysastrobin and BYF14182, which are also significantly advantageous and useful for both insects/plant diseases control and productivity mentioned above.

Isotianil of the ingredient (a) according to the invention is one of plant diseases-controlling agents described in Japanese Patent Publication (KOHYO) No. 2001-522840, and above mentioned fipronil and ethiprole of the ingredient (b) and the above ingredients (c), except for orysastrobin and BYF14182, are described in Pesticide Manual (13th Ed., published by BCPC, 2003). Orysastrobin is the compound described in German patent Publication No. 19539324. BYF14182 is the compound described in WO 2006/092291.

In the combination composition of the invention, the mixing ratio of the active ingredients should not be limited, and it may vary over a relatively wide range depending on specific active compounds to be mixed, predicted insects/plant diseases to be generated and the like. Generally the mixing ratio may be, for example, in the range of 0.02-3 parts by weight of (b) fipronil or ethiprole, preferably 0.05-2.5 parts by weight of (b) per part by weight of (a) isotianil. When the

ingredient (c) is also contained, (c) can be used in the range of 0.5-2 parts by weight, preferably 1-1.5 parts by weight per part by weight of (a) isotianil.

Further mixing ratios as parts by weight of (b) fipronil or ethiprole per part by weight of (a) isotianil can be used according to the present invention with increasing preference in the order
5 given are:

0.02, 0.05, 0.075, 0.1, 0.15, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1, 1.1, 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9, 2, 2.1, 2.2, 2.3, 2.4, 2.5, 2.6, 2.7, 2.8, 2.9, 3.

When the ingredient (c) is also contained,, further mixing ratios as parts by weight of ingredient(c) per part by weight of (a) isotianil can be used according to the present invention with increasing
10 preference in the order given are:

0.5, 0.6, 0.7, 0.8, 0.9, 1, 1.1, 1.2, 1.3, 1.4, 1.5, 1.6, 1.7, 1.8, 1.9, 2.

In the use of the combination compositions of the invention, insects/plant diseases-control after transplantation of rice seedlings to the field can be appropriately achieved by a method comprising sowing paddy rice seeds to nursery boxes and applying the present compositions to the seeds
15 before covering them with soil, or a method comprising mixing seedbed soil for nursery boxes or soil for covering seeds with the present compositions.

Exemplary insects/plant diseases targeted for the combination compositions of the invention include, but are not limited to, the followings:

as paddy rice insects,

20 Hemiptera: Nephotettix cincticeps, Nilaparvata lugens, Laodelphax striatellus (Fallen), Sogatella furcifera (Horvath), and etc;

Coleoptera: Oulema oryzae (Kuwayama), Lissorhoptrus oryzophilus Kuschel, and etc;

Lepidoptera: Chilo suppressalis (Walker), Cnaphalocrocis medinalis (Guenee), Parnara guttata (Bremex et Grey), and etc; and

25 as paddy rice diseases, Pyricularia oryzae, Pellicularia sasakii, Cercospora janseana, Rhizoctonia oryzae, Gaeumannomyces graminis var. graminis, Neovossia barclayana, Bipolaris oryzae, Pythium sp., Rhizoctonia solani, Achlya sp. and Sclerotium rolfsii, Sclerotium oryzae, Bipolaris oryzae, Entyloma oryzae, Rhizoctonia solani, Xanthomonas campestris pv oryzae, and etc.

According to the invention all plants and plant parts can be treated. By plants is meant all plants
30 and plant populations such as desirable and undesirable wild plants, cultivars and plant varieties (whether or not protectable by plant variety or plant breeder's rights). Cultivars and plant varieties

can be plants obtained by conventional propagation and breeding methods which can be assisted or supplemented by one or more biotechnological methods such as by use of double haploids, protoplast fusion, random and directed mutagenesis, molecular or genetic markers or by bioengineering and genetic engineering methods. By plant parts is meant all above ground and
 5 below ground parts and organs of plants such as shoot, leaf, blossom and root, whereby for example leaves, needles, stems, branches, blossoms, fruiting bodies, fruits and seed as well as roots, corms and rhizomes are listed. Crops and vegetative and generative propagating material, for example cuttings, corms, rhizomes, runners and seeds also belong to plant parts.

Among the plants that can be protected by the method according to the invention, mention may be
 10 made of major field crops like corn, soybean, cotton, Brassica oilseeds such as Brassica napus (e.g. canola), Brassica rapa, B. juncea (e.g. mustard) and Brassica carinata, rice, wheat, sugarbeet, sugarcane, oats, rye, barley, millet, triticale, flax, vine and various fruits and vegetables of various botanical taxa such as Rosaceae sp. (for instance pip fruit such as apples and pears, but also stone fruit such as apricots, cherries, almonds and peaches, berry fruits such as strawberries), Ribesioideae
 15 sp., Juglandaceae sp., Betulaceae sp., Anacardiaceae sp., Fagaceae sp., Moraceae sp., Oleaceae sp., Actinidaceae sp., Lauraceae sp., Musaceae sp. (for instance banana trees and plantings), Rubiaceae sp. (for instance coffee), Theaceae sp., Sterculiaceae sp., Rutaceae sp. (for instance lemons, oranges and grapefruit) ; Solanaceae sp. (for instance tomatoes, potatoes, peppers, eggplant), Liliaceae sp., Compositae sp. (for instance lettuce, artichoke and chicory - including root chicory, endive or
 20 common chicory), Umbelliferae sp. (for instance carrot, parsley, celery and celeriac), Cucurbitaceae sp. (for instance cucumber – including pickling cucumber, squash, watermelon, gourds and melons), Alliaceae sp. (for instance onions and leek), Cruciferae sp. (for instance white cabbage, red cabbage, broccoli, cauliflower, brussel sprouts, pak choi, kohlrabi, radish, horseradish, cress, Chinese cabbage), Leguminosae sp. (for instance peanuts, peas and beans - such as
 25 climbing beans and broad beans), Chenopodiaceae sp. (for instance mangold, spinach beet, spinach, beetroots), Malvaceae (for instance okra), Asparagaceae (for instance asparagus) ; horticultural and forest crops; ornamental plants ; as well as genetically modified homologues of these crops.

The method of treatment according to the invention can be used in the treatment of genetically modified organisms (GMOs), e.g. plants or seeds. Genetically modified plants (or transgenic
 30 plants) are plants of which a heterologous gene has been stably integrated into genome. The expression "heterologous gene" essentially means a gene which is provided or assembled outside the plant and when introduced in the nuclear, chloroplastic or mitochondrial genome gives the transformed plant new or improved agronomic or other properties by expressing a protein or polypeptide of interest or by downregulating or silencing other gene(s) which are present in the
 35 plant (using for example, antisense technology, cosuppression technology or RNA interference – RNAi - technology). A heterologous gene that is located in the genome is also called a transgene. A transgene that is defined by its particular location in the plant genome is called a transformation or

transgenic event.

Depending on the plant species or plant cultivars, their location and growth conditions (soils, climate, vegetation period, diet), the treatment according to the invention may also result in superadditive ("synergistic") effects. Thus, for example, reduced application rates and/or a widening of the activity spectrum and/or an increase in the activity of the active compounds and compositions which can be used according to the invention, better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, bigger fruits, larger plant height, greener leaf color, earlier flowering, higher quality and/or a higher nutritional value of the harvested products, higher sugar concentration within the fruits, better storage stability and/or processability of the harvested products are possible, which exceed the effects which were actually to be expected.

At certain application rates, the active compound combinations according to the invention may also have a strengthening effect in plants. Accordingly, they are also suitable for mobilizing the defense system of the plant against attack by unwanted microorganisms. This may, if appropriate, be one of the reasons of the enhanced activity of the combinations according to the invention, for example against fungi. Plant-strengthening (resistance-inducing) substances are to be understood as meaning, in the present context, those substances or combinations of substances which are capable of stimulating the defense system of plants in such a way that, when subsequently inoculated with unwanted microorganisms, the treated plants display a substantial degree of resistance to these microorganisms. In the present case, unwanted microorganisms are to be understood as meaning insects/plant diseases. Thus, the substances according to the invention can be employed for protecting plants against attack by the abovementioned pathogens within a certain period of time after the treatment. The period of time within which protection is effected generally extends from 1 to 10 days, preferably 1 to 7 days, after the treatment of the plants with the active compounds.

Plants and plant cultivars which are preferably to be treated according to the invention include all plants which have genetic material which impart particularly advantageous, useful traits to these plants (whether obtained by breeding and/or biotechnological means).

Plants and plant cultivars which are also preferably to be treated according to the invention are resistant against one or more biotic stresses, i.e. said plants show a better defense against animal and microbial pests, such as against nematodes, insects, mites, phytopathogenic fungi, bacteria, viruses and/or viroids.

Plants and plant cultivars which may also be treated according to the invention are those plants which are resistant to one or more abiotic stresses. Abiotic stress conditions may include, for example, drought, cold temperature exposure, heat exposure, osmotic stress, flooding, increased

soil salinity, increased mineral exposure, ozone exposure, high light exposure, limited availability of nitrogen nutrients, limited availability of phosphorus nutrients, shade avoidance.

- Plants and plant cultivars which may also be treated according to the invention, are those plants characterized by enhanced yield characteristics. Increased yield in said plants can be the result of, for example, improved plant physiology, growth and development, such as water use efficiency, water retention efficiency, improved nitrogen use, enhanced carbon assimilation, improved photosynthesis, increased germination efficiency and accelerated maturation. Yield can furthermore be affected by improved plant architecture (under stress and non-stress conditions), including but not limited to, early flowering, flowering control for hybrid seed production, seedling vigor, plant size, internode number and distance, root growth, seed size, fruit size, pod size, pod or ear number, seed number per pod or ear, seed mass, enhanced seed filling, reduced seed dispersal, reduced pod dehiscence and lodging resistance. Further yield traits include seed composition, such as carbohydrate content, protein content, oil content and composition, nutritional value, reduction in anti-nutritional compounds, improved processability and better storage stability.
- Plants that may be treated according to the invention are hybrid plants that already express the characteristic of heterosis or hybrid vigor which results in generally higher yield, vigor, health and resistance towards biotic and abiotic stresses). Such plants are typically made by crossing an inbred male-sterile parent line (the female parent) with another inbred male-fertile parent line (the male parent). Hybrid seed is typically harvested from the male sterile plants and sold to growers. Male sterile plants can sometimes (e.g. in corn) be produced by detasseling, i.e. the mechanical removal of the male reproductive organs (or male flowers) but, more typically, male sterility is the result of genetic determinants in the plant genome. In that case, and especially when seed is the desired product to be harvested from the hybrid plants it is typically useful to ensure that male fertility in the hybrid plants is fully restored. This can be accomplished by ensuring that the male parents have appropriate fertility restorer genes which are capable of restoring the male fertility in hybrid plants that contain the genetic determinants responsible for male-sterility. Genetic determinants for male sterility may be located in the cytoplasm. Examples of cytoplasmic male sterility (CMS) were for instance described in Brassica species (WO 92/05251, WO 95/09910, WO 98/27806, WO 05/002324, WO 06/021972 and US 6,229,072). However, genetic determinants for male sterility can also be located in the nuclear genome. Male sterile plants can also be obtained by plant biotechnology methods such as genetic engineering. A particularly useful means of obtaining male-sterile plants is described in WO 89/10396 in which, for example, a ribonuclease such as barnase is selectively expressed in the tapetum cells in the stamens. Fertility can then be restored by expression in the tapetum cells of a ribonuclease inhibitor such as barstar (e.g. WO 91/02069).
- Plants or plant cultivars (obtained by plant biotechnology methods such as genetic engineering) which may be treated according to the invention are herbicide-tolerant plants, i.e. plants made

tolerant to one or more given herbicides. Such plants can be obtained either by genetic transformation, or by selection of plants containing a mutation imparting such herbicide tolerance.

Herbicide-resistant plants are for example glyphosate-tolerant plants, i.e. plants made tolerant to the herbicide glyphosate or salts thereof. Plants can be made tolerant to glyphosate through different means. For example, glyphosate-tolerant plants can be obtained by transforming the plant with a gene encoding the enzyme 5-enolpyruvylshikimate-3-phosphate synthase (EPSPS). Examples of such EPSPS genes are the AroA gene (mutant CT7) of the bacterium *Salmonella typhimurium* (Comai et al., 1983, Science 221, 370-371), the CP4 gene of the bacterium *Agrobacterium* sp. (Barry et al., 1992, Curr. Topics Plant Physiol. 7, 139-145), the genes encoding a *Petunia* EPSPS (Shah et al., 1986, Science 233, 478-481), a Tomato EPSPS (Gasser et al., 1988, J. Biol. Chem. 263, 4280-4289), or an Eleusine EPSPS (WO 01/66704). It can also be a mutated EPSPS as described in for example EP 0837944, WO 00/66746, WO 00/66747 or WO02/26995. Glyphosate-tolerant plants can also be obtained by expressing a gene that encodes a glyphosate oxido-reductase enzyme as described in U.S. Patent Nos. 5,776,760 and 5,463,175. Glyphosate-tolerant plants can also be obtained by expressing a gene that encodes a glyphosate acetyl transferase enzyme as described in for example WO 02/36782, WO 03/092360, WO 05/012515 and WO 07/024782. Glyphosate-tolerant plants can also be obtained by selecting plants containing naturally-occurring mutations of the above-mentioned genes, as described in for example WO 01/024615 or WO 03/013226.

Other herbicide resistant plants are for example plants that are made tolerant to herbicides inhibiting the enzyme glutamine synthase, such as bialaphos, phosphinothricin or glufosinate. Such plants can be obtained by expressing an enzyme detoxifying the herbicide or a mutant glutamine synthase enzyme that is resistant to inhibition. One such efficient detoxifying enzyme is an enzyme encoding a phosphinothricin acetyltransferase (such as the bar or pat protein from *Streptomyces* species). Plants expressing an exogenous phosphinothricin acetyltransferase are for example described in U.S. Patent Nos. 5,561,236; 5,648,477; 5,646,024; 5,273,894; 5,637,489; 5,276,268; 5,739,082; 5,908,810 and 7,112,665.

Further herbicide-tolerant plants are also plants that are made tolerant to the herbicides inhibiting the enzyme hydroxyphenylpyruvate dioxygenase (HPPD). Hydroxyphenylpyruvate dioxygenases are enzymes that catalyze the reaction in which para-hydroxyphenylpyruvate (HPP) is transformed into homogentisate. Plants tolerant to HPPD-inhibitors can be transformed with a gene encoding a naturally-occurring resistant HPPD enzyme, or a gene encoding a mutated HPPD enzyme as described in WO 96/38567, WO 99/24585 and WO 99/24586. Tolerance to HPPD-inhibitors can also be obtained by transforming plants with genes encoding certain enzymes enabling the formation of homogentisate despite the inhibition of the native HPPD enzyme by the HPPD-inhibitor. Such plants and genes are described in WO 99/34008 and WO 02/36787. Tolerance of

plants to HPPD inhibitors can also be improved by transforming plants with a gene encoding an enzyme prephenate dehydrogenase in addition to a gene encoding an HPPD-tolerant enzyme, as described in WO 2004/024928.

Still further herbicide resistant plants are plants that are made tolerant to acetolactate synthase (ALS) inhibitors. Known ALS-inhibitors include, for example, sulfonylurea, imidazolinone, triazolopyrimidines, pyrimidinyoxy(thio)benzoates, and/or sulfonylaminocarbonyltriazolinone herbicides. Different mutations in the ALS enzyme (also known as acetohydroxyacid synthase, AHAS) are known to confer tolerance to different herbicides and groups of herbicides, as described for example in Tranel and Wright (2002, Weed Science 50:700-712), but also, in U.S. Patent No. 5,605,011, 5,378,824, 5,141,870, and 5,013,659. The production of sulfonylurea-tolerant plants and imidazolinone-tolerant plants is described in U.S. Patent Nos. 5,605,011; 5,013,659; 5,141,870; 5,767,361; 5,731,180; 5,304,732; 4,761,373; 5,331,107; 5,928,937; and 5,378,824; and international publication WO 96/33270. Other imidazolinone-tolerant plants are also described in for example WO 2004/040012, WO 2004/106529, WO 2005/020673, WO 2005/093093, WO 2006/007373, WO 2006/015376, WO 2006/024351, and WO 2006/060634. Further sulfonylurea- and imidazolinone-tolerant plants are also described in for example WO 07/024782.

Other plants tolerant to imidazolinone and/or sulfonylurea can be obtained by induced mutagenesis, selection in cell cultures in the presence of the herbicide or mutation breeding as described for example for soybeans in U.S. Patent 5,084,082, for rice in WO97/41218, for sugar beet in U.S. Patent 5,773,702 and WO99/057965, for lettuce in U.S. Patent 5,198,599, or for sunflower in WO01/065922.

Plants or plant cultivars (obtained by plant biotechnology methods such as genetic engineering) which may also be treated according to the invention are insect-resistant transgenic plants, i.e. plants made resistant to attack by certain target insects. Such plants can be obtained by genetic transformation, or by selection of plants containing a mutation imparting such insect resistance.

An "insect-resistant transgenic plant", as used herein, includes any plant containing at least one transgene comprising a coding sequence encoding:

- 1) an insecticidal crystal protein from *Bacillus thuringiensis* or an insecticidal portion thereof, such as the insecticidal crystal proteins listed by Crickmore et al. (1998, Microbiology and Molecular Biology Reviews, 62: 807-813), updated by Crickmore et al. (2005) at the *Bacillus thuringiensis* toxin nomenclature, online at: http://www.lifesci.sussex.ac.uk/Home/Neil_Crickmore/Bt/), or insecticidal portions thereof, e.g., proteins of the Cry protein classes Cry1Ab, Cry1Ac, Cry1F, Cry2Ab, Cry3Aa, or Cry3Bb or insecticidal portions thereof; or

- 2) a crystal protein from *Bacillus thuringiensis* or a portion thereof which is insecticidal in the presence of a second other crystal protein from *Bacillus thuringiensis* or a portion thereof, such as the binary toxin made up of the Cry34 and Cry35 crystal proteins (Moellenbeck et al. 2001, Nat. Biotechnol. 19: 668-72; Schnepf et al. 2006, Applied Environm. Microbiol. 71, 1765-1774); or
- 3) a hybrid insecticidal protein comprising parts of different insecticidal crystal proteins from *Bacillus thuringiensis*, such as a hybrid of the proteins of 1) above or a hybrid of the proteins of 2) above, e.g., the Cry1A.105 protein produced by corn event MON98034 (WO 2007/027777); or
- 4) a protein of any one of A1) to A3) above wherein some, particularly 1 to 10, amino acids have been replaced by another amino acid to obtain a higher insecticidal activity to a target insect species, and/or to expand the range of target insect species affected, and/or because of changes introduced into the encoding DNA during cloning or transformation, such as the Cry3Bb1 protein in corn events MON863 or MON88017, or the Cry3A protein in corn event MIR604;
- 5) an insecticidal secreted protein from *Bacillus thuringiensis* or *Bacillus cereus*, or an insecticidal portion thereof, such as the vegetative insecticidal (VIP) proteins listed at:
- http://www.lifesci.sussex.ac.uk/home/Neil_Crickmore/Bt/vip.html, e.g., proteins from the VIP3Aa protein class; or
- 6) a secreted protein from *Bacillus thuringiensis* or *Bacillus cereus* which is insecticidal in the presence of a second secreted protein from *Bacillus thuringiensis* or *B. cereus*, such as the binary toxin made up of the VIP1A and VIP2A proteins (WO 94/21795); or
- 7) a hybrid insecticidal protein comprising parts from different secreted proteins from *Bacillus thuringiensis* or *Bacillus cereus*, such as a hybrid of the proteins in 1) above or a hybrid of the proteins in 2) above; or
- 8) a protein of any one of 1) to 3) above wherein some, particularly 1 to 10, amino acids have been replaced by another amino acid to obtain a higher insecticidal activity to a target insect species, and/or to expand the range of target insect species affected, and/or because of changes introduced into the encoding DNA during cloning or transformation (while still encoding an insecticidal protein), such as the VIP3Aa protein in cotton event COT102.

Of course, an insect-resistant transgenic plant, as used herein, also includes any plant comprising a combination of genes encoding the proteins of any one of the above classes 1 to 8. In one embodiment, an insect-resistant plant contains more than one transgene encoding a protein of any

one of the above classes 1 to 8, to expand the range of target insect species affected when using different proteins directed at different target insect species, or to delay insect resistance development to the plants by using different proteins insecticidal to the same target insect species but having a different mode of action, such as binding to different receptor binding sites in the insect.

Plants or plant cultivars (obtained by plant biotechnology methods such as genetic engineering) which may also be treated according to the invention are tolerant to abiotic stresses. Such plants can be obtained by genetic transformation, or by selection of plants containing a mutation imparting such stress resistance. Particularly useful stress tolerance plants include:

- a. plants which contain a transgene capable of reducing the expression and/or the activity of poly(ADP-ribose)polymerase (PARP) gene in the plant cells or plants as described in WO 00/04173 or EP 04077984.5 or EP 06009836.5.
- b. plants which contain a stress tolerance enhancing transgene capable of reducing the expression and/or the activity of the PARG encoding genes of the plants or plants cells, as described e.g. in WO 2004/090140.
- c. plants which contain a stress tolerance enhancing transgene coding for a plant-functional enzyme of the nicotineamide adenine dinucleotide salvage synthesis pathway including nicotinamidase, nicotinate phosphoribosyltransferase, nicotinic acid mononucleotide adenylyl transferase, nicotinamide adenine dinucleotide synthetase or nicotine amide phosphorybosyltransferase as described e.g. in EP 04077624.7 or WO 2006/133827 or PCT/EP07/002433.

Plants or plant cultivars (obtained by plant biotechnology methods such as genetic engineering) which may also be treated according to the invention show altered quantity, quality and/or storage-stability of the harvested product and/or altered properties of specific ingredients of the harvested product such as :

- 1) transgenic plants which synthesize a modified starch, which in its physical-chemical characteristics, in particular the amylose content or the amylose/amylopectin ratio, the degree of branching, the average chain length, the side chain distribution, the viscosity behaviour, the gelling strength, the starch grain size and/or the starch grain morphology, is changed in comparison with the synthesised starch in wild type plant cells or plants, so that this is better suited for special applications. Said transgenic plants synthesizing a modified starch are disclosed, for example, in EP 0571427, WO 95/04826, EP 0719338, WO 96/15248, WO 96/19581, WO 96/27674, WO 97/11188, WO 97/26362, WO 97/32985, WO 97/42328, WO 97/44472, WO 97/45545, WO 98/27212, WO 98/40503, WO99/58688,

WO 99/58690, WO 99/58654, WO 00/08184, WO 00/08185, WO 00/08175, WO 00/28052,
 WO 00/77229, WO 01/12782, WO 01/12826, WO 02/101059, WO 03/071860, WO
 2004/056999, WO 2005/030942, WO 2005/030941, WO 2005/095632, WO 2005/095617,
 WO 2005/095619, WO 2005/095618, WO 2005/123927, WO 2006/018319, WO
 2006/103107, WO 2006/108702, WO 2007/009823, WO 00/22140, WO 2006/063862, WO
 2006/072603, WO 02/034923, EP 06090134.5, EP 06090228.5, EP 06090227.7, EP
 07090007.1, EP 07090009.7, WO 01/14569, WO 02/79410, WO 03/33540, WO
 2004/078983, WO 01/19975, WO 95/26407, WO 96/34968, WO 98/20145, WO 99/12950,
 WO 99/66050, WO 99/53072, US 6734341, WO 00/11192, WO 98/22604, WO 98/32326,
 WO 01/98509, WO 01/98509, WO 2005/002359, US 5824790, US 6013861, WO
 94/04693, WO 94/09144, WO 94/11520, WO 95/35026, WO 97/20936

2) transgenic plants which synthesize non starch carbohydrate polymers or which synthesize
 non starch carbohydrate polymers with altered properties in comparison to wild type plants
 without genetic modification. Examples are plants producing polyfructose, especially of the
 inulin and levan-type, as disclosed in EP 0663956, WO96/01904, WO96/21023, WO
 98/39460, and WO 99/24593, plants producing alpha 1,4 glucans as disclosed in WO
 95/31553, US 2002031826, US 6284479, US 5712107, WO 97/47806, WO 97/47807, WO
 97/47808 and WO 00/14249, plants producing alpha-1,6 branched alpha-1,4-glucans, as
 disclosed in WO 00/73422, plants producing alternan, as disclosed in WO 00/47727, EP
 06077301.7, US 5908975 and EP 0728213,

3) transgenic plants which produce hyaluronan, as for example disclosed in WO 2006/032538,
 WO 2007/039314, WO 2007/039315, WO 2007/039316, JP 2006304779, and WO
 2005/012529.

- Particularly useful transgenic plants which may be treated according to the
 invention are plants containing transformation events, or combination of
 transformation events, that are the subject of petitions for non-regulated status, in
 the United States of America, to the Animal and Plant Health Inspection Service
 (APHIS) of the United States Department of Agriculture (USDA) whether such
 petitions are granted or are still pending. At any time this information is readily
 available from APHIS (4700 River Road Riverdale, MD 20737, USA), for instance
 on its internet site (URL http://www.aphis.usda.gov/brs/not_reg.html).

Additional particularly useful plants containing single transformation events or combinations of
 transformation events are listed for example in the databases from various national or regional
 regulatory agencies (see for example http://gmoinfo.jrc.it/gmp_browse.aspx and
<http://www.agbios.com/dbase.php>).

Further particularly transgenic plants include plants containing a transgene in an agronomically neutral or beneficial position as described in any of the patent publications listed in Table A.

Table A

Plant species	Event	Trait	Patent reference
Corn	PV-ZMGT32 (NK603)	Glyphosate tolerance	US 2007-056056
Corn	MIR604	Insect resistance (cry3a055)	EP1737290
Corn	LY038	High lysine content	US7157281
Corn	3272	Self processing corn (alpha-amylase)	US2006-230473
Corn	PV-ZMIR13 (MON863)	Insect resistance	US2006-095986
Corn	DAS-59122-7	Insect resistance	US2006-070139
Corn	TC1507	Insect resistance	US2005-039226
Corn	Mon810	Insect resistance	US2004-180373
Corn	VIP1034	Insect resistance	WO03052073
Corn	B16	Glufosinate resistance	US2003126634
Corn	GA21	Glyphosate resistance	US6040497
Corn	GG25	Glyphosate resistance	US6040497
Corn	GJ11	Glyphosate resistance	US6040497
Corn	FI117	Glyphosate resistance	US6040497
Corn	GAT-ZM1	Glufosinate tolerance	WO01/51654
Wheat	Event 1	Fusarium resistance (trichothecene 3-O-acetyltransferase)	CA2561992
Rice	GAT-OS2	Glufosinate tolerance	WO01/83818

The combination compositions of the invention can be formulated into conventional preparation forms to use for controlling paddy rice insects/plant diseases.

- 5 Examples of the preparation forms may include aerosol dispenser, capsule suspension, cold fogging concentrate, hot fogging concentrate, encapsulated granule, fine granule, flowable concentrate for seed treatment, ready-to-use solutions, dustable powder, emulsifiable concentrate, emulsion oil in water, emulsion water in oil, macrogranule, microgranule, oil dispersible powder, oil miscible

flowable concentrate, oil miscible liquid, froths, paste, seed coated with a pesticide, suspension concentrate (flowable concentrate), suspensions-emulsions-concentrates, soluble concentrate, suspensions, soluble powder, granule, water soluble granules or tablets, water soluble powder for seed treatment, wettable powder, natural and synthetic materials impregnated with active compound, micro-encapsulation in polymeric materials and in jackets for seed, as well as ULV-cold and hot fogging formulations, gas (under pressure), gas generating product, plant rodlet, powder for dry seed treatment, solution for seed treatment, ultra low volume (ULV) liquid, ultra low volume (ULV) suspension, water dispersible granules or tablets, water dispersible powder for slurry treatment and the like.

- 10 The preparations according to the present invention can be produced according to per se known methods. The preparations can be produced, for example, by mixing above active ingredients with solid or liquid carriers or diluents, optionally with surfactants, extenders, agriculturally acceptable additives or other pharmaceutical auxiliaries.

As solid carriers and/or solid diluents which may be used in preparations of the compositions of the invention, there can be mentioned, for example, mineral ore-based inorganic substances (e.g., clays, kaolins, talc, calcium carbonate, diatomaceous earth, zeolite, bentonite, acid clay, activated earth, attapulgus clay, montmorillonite, vermiculite, pearlite, pumice, quartz sand, quartz rock, calcite, marble, sepiolite, dolomite and synthetic granules of inorganic and organic powders and the like);

synthetics (e.g., white carbons such as hydrophilic silica, hydrophobic silica and calcium silicate, aluminium oxide, synthetic zeolite, titanium dioxide and the like);

vegetable organic materials (e.g., soy powder, tobacco powder, corn cob powder, walnut powder, wheat flour, wood powder, starch, crystalline cellulose, paper, sawdust, coconut shells, corn stalks and tobacco stalks and the like);

synthetic or natural polymers (e.g., coumarone resin, petroleum resin, alkyd resin, polyalkylene glycol, ketone resin, ester gum and the like);

waxes (e.g., carnauba wax, bees wax and the like);

water-soluble substances (e.g., urea, lactose, sucrose, ammonium sulfate, potassium chloride and the like).

Suitable liquid carriers and/or liquid diluents include, for example,

30 fats and oils such as coconut oil, rapeseed oil, corn oil, soybean oil, rice bran oil and the like;

paraffinic or naphthenic hydrocarbon solvents such as kerosene, mineral oils, spindle oils, white oils, normal paraffin, isoparaffin, naphthene and the like;

aromatic hydrocarbon solvents such as xylene, alkylbenzene, alkylnaphthalene and the like;

alcohols such as ethanol, benzyl alcohol, isopropanol, cyclohexanol and the like;

polyhydric alcohols such as ethylene glycol, diethylene glycol, propylene glycol, polyethylene glycol and the like;

- 5 ether-alcohols such as ethylene glycol ethyl ether, ethylene glycol phenyl ether, diethylene glycol ethyl ether, diethylene glycol butyl ether and the like;

ketones such as methyl ethyl ketone, diisobutyl ketone, cyclohexanone, acetophenone, isophorone, gamma-butyrolactone and the like;

- 10 esters such as fatty acid methyl esters (e.g., coconut oil fatty acid methyl ester), dibasic acid methyl ester (e.g., dimethyl succinate, dimethyl glytamate, dimethyl adipate), ethyl acetate, amyl acetate, ethylene glycol acetate, diethylene glycol acetate and the like;

ethers such as dioxane, tetrahydrofuran and the like;

polar solvents such as dimethylformamide, dimethylacetamide, dimethylsulfoxide, N-alkyl pyrrolidone and the like or water etc.

- 15 In addition, surfactants, binders and other auxiliaries can be used for the purpose of emulsifying, dispersing, wetting, extending, anti-decomposition and increasing efficacy of the active ingredients and/or for the purpose of improving physical property of the preparations (control of disintegration, improvement of flowability, conferring antifreezing effect and rain-resistance and the like). The surfactants which may be used include of any type including non-ionic, anionic, cationic and
- 20 amphoteric surfactants, although non-ionic and/or anionic surfactants are generally used. Suitable non-ionic surfactants may include, for example, sorbitan fatty acid esters, polyoxyalkylene sorbitan fatty acid esters, sucrose fatty acid esters; polyoxyalkylene fatty acid esters, polyoxyalkylene resin acid esters, polyoxyalkylene fatty acid diesters; polyoxyalkylene castor oil; polyoxyalkylene hardened castor oil; polyoxyalkylene alkyl ethers; polyoxyalkylene alkyl phenyl ethers,
- 25 polyoxyalkylene dialkyl phenyl ethers, polyoxyalkylene alkyl phenyl ether-formaldehyde condensates; polyoxyethylene/polyoxypropylene block polymers, alkylpolyoxyethylene/polyoxypropylene block polymer ethers, alkylphenylpolyoxyethylene/polyoxypropylene block polymer ethers; polyoxyalkylenealkylamines, polyoxyalkylene fatty acid amides; polyoxyalkylene benzyl phenyl (or phenyl phenyl) ethers,
- 30 polyoxyalkylene styryl phenyl (or phenyl phenyl) ethers; polyoxyalkylene ether- and ester-type silicones as well as fluorinated surfactants etc.

In addition, suitable anionic surfactants may include, for example, alkylsulfate salts,

polyoxyalkylene alkyl ether sulfate salts, polyoxyalkylene alkylphenyl ether sulfate salts, polyoxyalkylene benzyl (or styryl) phenyl (or phenyl phenyl) ether sulfate salts, polyoxyethylene/polyoxypropylene block polymer sulfate salts; paraffin (alkane) sulfonate salts, alpha olefin sulfonate salts, dialkyl sulfosuccinate salts, alkylbenzene sulfonate salts, mono- or
5 dialkyl naphthalene sulfonate salts, naphthalene sulfonate/formaldehyde condensate salts, alkyldiphenyl ether disulfonate salts, ligninsulfonate salts, polyoxyalkylene alkylphenyl ether sulfonate salts, polyoxyalkylene ether sulfosuccinate half ester salts; fatty acid salts, N-methyl fatty acid sarcosinate, resin acid salts; polyoxyalkylene alkyl ether phosphate salts, polyoxyalkylene mono- or dialkylphenyl ether phosphate salts, polyoxyalkylene benzylated (or styrylated) phenyl
10 (or phenyl phenyl) ether phosphate salts, polyoxyethylene/polyoxypropylene block polymer phosphate salts; polyacrylate salts, polycarboxylate salts and the like.

The cationic surfactants used in the invention may include ammonium type surfactants such as alkyltrimethylammonium chloride, methyl-polyoxyethylene-alkyl ammonium chloride, alkyl-N-methylpyridinium bromide, mono- or dialkylmethylated ammonium chloride,
15 alkyldimethylpropylenediamine dichloride and the like; benzalkonium type surfactants such as alkyldimethyl benzalkonium chloride, benzethonium chloride and the like.

The amphoteric surfactants may include betaine type surfactants such as dialkyldiaminoethyl betaine, alkyldimethylbenzyl betaine and the like.

Furthermore, exemplary binders may include bentonite, casein, gelatin, starch, dextrin,
20 ligninsulfonate, alginate, gum Arabic, xanthan gum, carboxymethylcellulose, methylcellulose, polyvinyl alcohol, polyvinylpyrrolidone, natural phospholipids, such as cephalins and lecithins and synthetic phospholipids and the like.

In addition, other auxiliaries include, for example, disintegration controlling agents, anti-freezing agents, antiseptic agents, flowability improving agents, defoaming agents, humectants, extenders,
25 rain-resistance-conferring agents, coloring agents and the like.

Above mentioned carriers and surfactants as well as binders and various auxiliaries can be appropriately used either alone or in combination depending on the intended use, with considering preparation forms of formulations, application stages and etc.

The preparations may contain 0.1-95% by weight, preferably 0.5-90% by weight of active
30 ingredients in total.

The combination compositions of the invention may be used as such or in their various preparation forms, for example in a single "ready-mix" form, in a combined spray mixture composed from separate formulations of the single active compounds, such as a "tank-mix" at the point of use, and in a combined use of the single active ingredients when applied in a sequential manner, i.e. one

after the other with a reasonably short period, such as a few hours or days. Preferably the order of applying the compounds (a) and (b) and/or (c) is not essential for working the present invention.

Examples

Test example 1: tests for intensity and growth of paddy rice seedlings.

5 Method

Standard nursery boxes for paddy rice seedlings (30 cm x 60 cm x 3 cm) were filled with about 2.5 L of artificial nursery soil and pre-germinated seed grains (variety: Koshi-hikari) were sowed there. Immediately after irrigation, 50 g per box of granular preparations containing respective active ingredients in predetermined concentrations were uniformly applied to the nursery boxes, and 1 L
10 of artificial nursery soil was applied to cover the seeds. Then, the nursery boxes were transferred into a warming nursery chamber and allowed to stand for 2 days at about 30°C. After emergence, the boxes were transferred to a plastic greenhouse for nursing seedlings and continuously observed the growth with the naked eyes. 3 weeks after sowing (at the optimum stage for transplantation), the green stems and the roots (level of mat formation, particularly rooting volume at bottom and
15 lateral sides of the box) were observed with the naked eyes. Furthermore, the breaking limit value of pieces (10 cm x 10 cm) cut from the resulting seedling mats were determined to estimate the intensity of the seedlings in the nursery boxes. More specifically, one side of the piece of the seedling mat was anchored and the other side was pulled gradually with a mechanical force gauge, scale on which at break of the piece was read as the breaking limit value (the tensile strength).
20 Table 1 shows the average scores of the triplicate observations with the naked eyes and the average breaking limit values of the pieces (10 cm x 10 cm) cut from the seedling mats calculated from values of triplicate determinations at two sites (6 pieces in total) for each experimental group.

Evaluation criteria of the naked eye observation:

- 1 = healthy
- 25 2 = slight delay of root growth
- 3 = mild delay of root growth which does not affect their practical use adversely
- 4 = delay of root growth which slightly affects their practical use adversely
- 5 = severe delay of root growth which adversely affects their practical use

Evaluation criteria of seedling mat intensity:

- 30 (Score of 100 represents average tensile strength of non-treated pieces as measured by using a mechanical force gauge.)

1 = 95 or more,

2 = 90 to less than 95,

3 = 70 to less than 90,

4 = 50 to less than 70,

5 5 = less than 50

Table 1

<u>Treatment</u>	<u>Amount of active ingredients/nursery box (g)</u>	<u>Tensile strength (mechanical force gauge)</u>	<u>Root Growth (observed with the naked eyes)</u>
<u>Untreated</u>		<u>1 to 5</u>	<u>1 to 5</u>
<u>Isotianil</u>		<u>1</u>	<u>1</u>
	<u>2</u>	<u>2</u>	<u>1</u>
	<u>1</u>	<u>2</u>	<u>1</u>
<u>Imidacloprid</u>	<u>1</u>	<u>5</u>	<u>4</u>
<u>Fipronil</u>	<u>0.5</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Imidacloprid</u>	<u>2+1</u>	<u>5</u>	<u>4</u>
<u>Isotianil+Imidacloprid</u>	<u>1+1</u>	<u>5</u>	<u>4</u>
<u>Isotianil+Fipronil</u>	<u>2+2.5</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>2+1</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>2+0.5</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>2+0.25</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>2+0.05</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>1+2.5</u>	<u>1</u>	<u>1</u>

<u>Isotianil+Fipronil</u>	<u>1+1</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>1+0.5</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>1+0.25</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Fipronil</u>	<u>1+0.05</u>	<u>1</u>	<u>1</u>
<u>Imidacloprid+Fipronil</u>	<u>1+2.5</u>	<u>3</u>	<u>3</u>
<u>Imidacloprid+Fipronil</u>	<u>1+1</u>	<u>3</u>	<u>3</u>
<u>Imidacloprid+Fipronil</u>	<u>1+0.5</u>	<u>3</u>	<u>3</u>
<u>Imidacloprid+Fipronil</u>	<u>1+0.25</u>	<u>3</u>	<u>3</u>
<u>Imidacloprid+Fipronil</u>	<u>1+0.05</u>	<u>3</u>	<u>3</u>
<u>Isotianil+Imidacloprid+Fipronil</u>	<u>1+1+2.5</u>	<u>1</u>	<u>1</u>
<u>Isotianil+Imidacloprid+Fipronil</u>	<u>1+1+1</u>	<u>1</u>	<u>1</u>

<u>Treatment</u>	<u>Amount of active ingredients/nursery box (g)</u>	<u>Tensile strength (mechanical force gauge)</u>	<u>Root Growth (observed with the naked eyes)</u>
		1 to 5	1 to 5
<u>Isotianil+Imidacloprid+Fipronil</u>	1+1+0.5	1	1
<u>Isotianil+Imidacloprid+Fipronil</u>	1+1+0.25	1	1
<u>Isotianil+Imidacloprid+Fipronil</u>	1+1+0.05	1	1

Test example 2: estimation of insect controlling effect against *Parnara guttata* (Bremex et Grey).

Method:

Once *Oryza sativa* (variety: Koshi-hikari) raised in standard nursery boxes for paddy rice reached the optimum stage of transplantation, they were treated with 50 g per box of granular composition containing 2% (w/w) of isotianil and 1% (w/w) of fipronil, and then lightly irrigated immediately before transplantation by a rice planter. They were cultivated and managed according to local agricultural practice. 70 days after transplantation, the number of damaged stocks was determined for 600 stocks per section. Results are shown in Table 2.

Table 2

Treatment	Amount of active ingredients /nursery box (g)	70 days after transplantation	
		The number of damaged stocks	Drug-induced damage
Untreated		90	None
Isotianil+Fipronil	1+2.5	0	1
Isotianil+Fipronil	1+0.5	2	1
Isotianil+Fipronil	1+0.05	2	1

Test example 3: estimation of plant disease controlling effect against *Pyricularia oryzae*.

Method

Once *Oryza sativa* (variety: Hitomebore) raised in standard nursery boxes for paddy rice reached the optimum stage of transplantation, they were treated with the same granular compositions as in the test example 2 in an amount of 50g / box, and then lightly irrigated immediately before transplantation by a rice planter. They were cultivated and managed according to local agricultural practice. 72 days after transplantation, 120 stocks per section were subjected to check of lesion to determine the ratio of lesion area. Results are shown in Table 3.

Table 3

Treatment	Amount of active ingredients /nursery box (g)	72 days after transplantation	
		Ratio of lesion area (%)	Drug-induced damage
Untreated		8.2	None
Isotianil+Fipronil	1+2.5	0.49	1
Isotianil+Fipronil	1+0.5	0.46	1
Isotianil+Fipronil	1+0.05	0.52	1

Formulation example 1 (microgranules)

85.5 parts by weight of clay mineral granules having grain size distribution of 0.2-0.7 mm is
 5 charged to a rotary vessel type mixer, to which, while the mixer rotated, 2 parts by weight of
 isotianil and 1 part by weight of fipronil as well as 10 parts by weight of a liquid diluent are
 sprayed and 1.5 parts by weight of white carbon is then added to afford the microgranules.

Formulation example 2 (microgranules)

84.5 parts by weight of clay mineral granules having grain size distribution of 0.2-0.7 mm is
 10 charged to a rotary vessel type mixer, to which, while the mixer rotated, 2 parts by weight of
 isotianil, 1 part by weight of fipronil and 1 part by weight of imidacloprid as well as 10 parts by
 weight of a liquid diluent are sprayed and 1.5 parts by weight of white carbon is then added to
 afford the microgranules.

Formulation example 3 (water-dispersible granules)

15 30 parts by weight of isotianil, 15 parts by weight of fipronil, 15 parts by weight of sodium
 ligninsulfonate, 15 parts by weight of sodium bentonite (montmorillonite), 15 parts by weight of
 calcined diatomaceous earth and 10 parts by weight of talc are mixed thoroughly, to which water
 containing 5 parts by weight of sodium polyoxyalkylene tristyryl phenyl ether sulfate is added, and
 the mixture was then kneaded, extruded with an extruding granulating machine attached with a 0.5
 20 mm screen and dried to afford the water-dispersible granules.

CLAIMS

1. A fungicidal and insecticidal composition comprising, as a combination:
 - (a) 3,4-dichloro-N-(2-cyanophenyl)isothiazole-5-carboxamide and
 - (b) fipronil or ethiprole.
- 5 2. The composition according to claim 1 further comprising (c) at least one compound selected from the group consisting of imidacloprid, thiacloprid, clothianidin, dinotefuran, thiamethoxam, spinosad, thifluzamide, furametpyr, orysastrobin and BYF14182.
3. The composition according to claim 1 or 2 having plant growth-promoting activity as a secondary effect.
- 10 4. The composition according to claim 3 wherein said plant growth-promoting activity is rooting-promoting activity.
5. The composition according to claim 1 containing the ingredient (b) in the range of 0.02-3 parts by weight per part by weight of the ingredient (a).
6. The composition according to claim 1, 2 or 5 containing the ingredient (c) in the range of
15 0.5-2 parts by weight per part by weight of the ingredient (a).
7. A method for controlling insects/plant diseases characterized in that a composition according to claim 1 or 2 is applied to paddy rice.
8. A method for exerting plant growth-promoting activity characterized in that a composition according to claim 1 or 2 is applied to paddy rice seedlings.
- 20 9. The use of a composition according to claim 1 or 2 for controlling insects/plant diseases.
10. The use of a composition according to claim 1 or 2 for exerting plant growth-promoting activity.
11. A method for controlling insects/plant diseases after transplantation of paddy rice to the field comprising: sowing a seed of the paddy rice in a nursery box and applying the
25 composition according to claim 1 or 2 to the seed before covering the seed with soil.
12. A method for controlling insects/plant diseases after transplantation of paddy rice to the field comprising: mixing seedbed soil for a nursery box or soil for covering a seed of the paddy rice with the composition according to claim 1 or 2.