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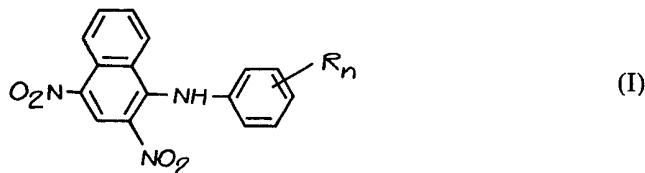
(54) NOVEL 1-ARYLAMINO-2,4-DINITRONAPHTHALENES
 AND THEIR USE AS PESTICIDES

(71) We, BAYER AKTIENGESELLSCHAFT, a body corporate, organised under the laws of Germany, of Leverkusen, Federal Republic of Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed to be particularly described in and by the following statement:—

The present invention relates to certain new 1-aryl-amino-2,4-dinitronaphthalenes, to a process for their preparation and to their use as pesticides.

It has already been disclosed that arylaminonaphthalenes, of which the naphthyl radical is substituted by halogen, exhibit insecticidal, acaricidal, fungicidal and bactericidal properties. Their action, however, is not fully satisfactory, especially if low amounts are used (see German Offenlegungsschrift (German Published Specification) 2,213,058).

The present invention provides, as new compounds, the 1-arylamino-2,4-dinitronaphthalenes of the general formula



in which

n is 1, 3, 3, 4 or 5 and

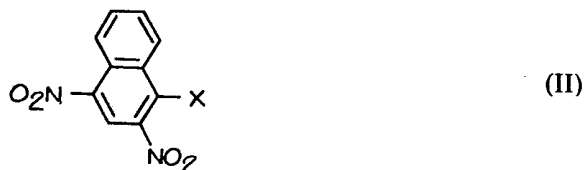
R represents hydrogen, alkyl, halogenoalkyl, alkenyl, alkynyl, alkoxy, alkylmercapto, alkylsulphonyl, sulphonate, halogenoalkoxy, halogenoalkylmercapto, acylamino, phenylazo, halogen, CN, NO₂ or alkoxy-carbonyl, the radicals R being selected independently of one another when n is 2 or more, and two radicals R, in the ortho-position to one another, conjointly with the two adjoining carbon atoms of the phenyl ring, can form an optionally halogen-substituted 1,3-dioxane ring.

The compounds of the formula (I) are distinguished by powerful pesticidal properties.

Preferably, each R is selected independently from hydrogen, alkyl with 1—4 carbon atoms (especially methyl), alkoxy with 1—4 carbon atoms, alkylmercapto with 1—4 carbon atoms, alkylsulphonyl with 1—4 carbon atoms, sulphonate, (especially sodium sulphonate), halogenoalkyl, halogenoalkoxy and halogenoalkylmercapto, each with 1—4 carbon atoms and with 3—5 halogen

atoms, acylamino, methoxycarbonyl, phenylazo, halogen, CN and NO₂, or two radicals R in the ortho-position to one another, conjointly with the two adjoining C atoms of the phenyl ring, form a 1,3-dioxane ring which is optionally substituted by fluorine.

5 The present invention also provides a process for the preparation of a 1-arylamino-2,4-dinitronaphthalene of the formula (I) in which 5
(a) a compound of the general formula



in which

10 X represents fluorine, chlorine, bromine or an aliphatic or aromatic sulphonic acid ester radical, 10
is reacted with an aniline of the general formula



in which

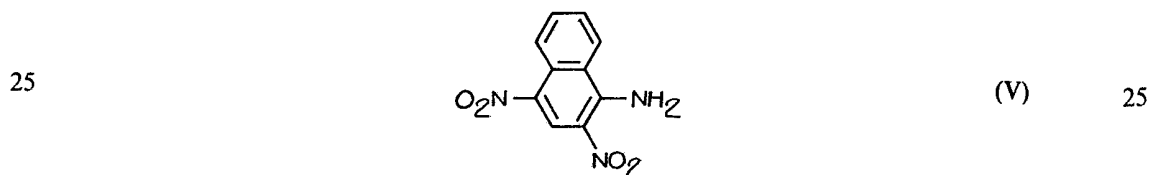
15 R and n have the above-mentioned meanings, 15
in the presence of an acid-binding agent and, if appropriate, in the presence of a diluent, or
(b) a compound of the general formula



20 in which

X represents fluorine, chlorine, bromine or an aliphatic or aromatic sulphonic acid ester radical and 20

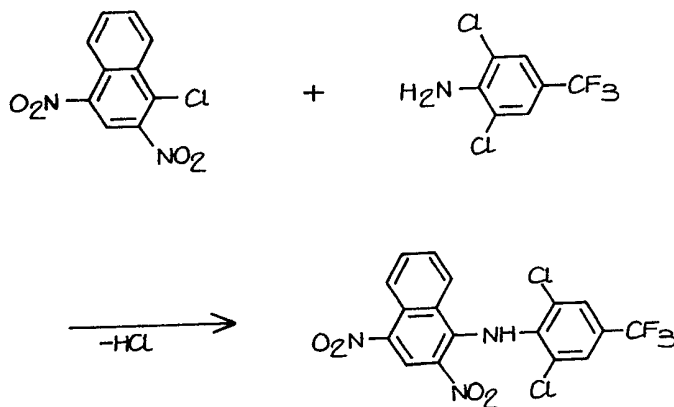
R and n have the above-mentioned meanings,
is reacted with 3,4-dinitro-1-naphthylamine, of the formula



in the presence of an acid-binding agent and, if appropriate, in the presence of a diluent.

30 Surprisingly, the 1-arylamino-2,4-dinitronaphthalenes of the present invention show a substantially higher insecticidal, acaricidal, development-inhibiting, fungicidal and bactericidal potency than the known halogen-substituted 30
arylamino-naphthalenes which are chemically the nearest compounds of the same type of action. The compounds according to the invention thus represent an enrichment of the art.

35 If 1-chloro-2,4-dinitronaphthalene and 2,6-dichloro-4-trifluoromethylaniline are used as starting materials, the course of the reaction can be represented by the following equation: 35



The compounds of the formulae (II), (III), (IV) and (V) to be used as starting materials are known or can be prepared in accordance with known processes.

Particularly preferred compounds are prepared by reacting 1-chloro-2,4-dinitronaphthalene with an aniline of the formula (III), in which each R represents hydrogen, nitro, cyano, methylsulphonyl, sulphonate, trifluoromethyl, trifluoromethylmercapto, trifluoromethoxy, alkyl or alkoxy with 1-4 carbon atoms, acylamino, phenylazo or halogen (such as fluorine, chlorine or bromine) and *n* represents 1, 2, 3 or 5.

The following may be mentioned as examples of anilines of the formula (III) which can be used according to the invention: 2,4-dichloroaniline, 2,5-dichloroaniline, 2,6-dichloroaniline, 3,4-dichloroaniline, 3,5-dichloroaniline, 3-chloro-4-fluoroaniline, 2,4-difluoroaniline, 2,4,5-trichloroaniline, 2,4,6-trichloroaniline, 4-nitroaniline, 2-chloro-4-nitroaniline, 3-chloro-4-nitroaniline, 4-chloro-2-nitroaniline, 4-chloro-3-nitroaniline, 2,4-dichloro-6-nitroaniline, 2,6-dichloro-4-nitroaniline, 2-bromo-4-nitroaniline, 2-bromo-4,6-dinitroaniline, 2-chloro-4,6-dinitroaniline, 3-chloro-6-methyl-4-nitroaniline, 5-chloro-2-methoxy-4-nitroaniline, 4-methyl-2-nitroaniline, 2,4-dinitroaniline, 2,6-dinitroaniline, methyl 4-amino-2-chloro-benzoate, 4-nitro-2-trifluoromethylaniline, 2-nitro-4-trifluoromethylaniline, 2,4-dinitro-6-trifluoromethylaniline, 3-trifluoromethylaniline, 2,4-bis-trifluoromethylaniline, 3,5-bis-trifluoromethylaniline, 4-chloro-2-trifluoromethylaniline, 4-chloro-3-trifluoromethylaniline, 3-chloro-4-trifluoromethylaniline, 2-chloro-5-trifluoromethylaniline, 2,6-dichloro-4-trifluoromethylaniline, 2,3,5,6-tetrachloro-4-trifluoromethylaniline, 4-trifluoromethoxyaniline, 3-chloro-4-trifluoromethoxyaniline, 3-chloro-4-trifluoromethylmercapto-aniline, 6-amino-2,2,4,4-tetrafluoro-1,3-benzodioxane, 2-bromo-4-nitro-6-trifluoromethylaniline, 2-amino-5-nitrobenzotrile, 2-amino-3,5-dinitrobenzotrile, 2-amino-3-bromo-5-nitrobenzotrile, 4-amino-1,3-phthalodinitrile, 2-amino-5-trifluoromethylbenzotrile, 4-ethoxyaniline, 4-methoxy-2-nitroaniline, 4-methoxy-3-nitroaniline, 4-methoxy-3-trifluoromethylaniline, 4-amino-3-nitro-azobenzene, 4-amino-3,5-dibromo-azobenzene, 4-methylsulphonylaniline, 2-methylsulphonyl-4-nitroaniline, the sodium salt of 3-amino-benzenesulphonic acid, 3-amino-acetanilide and 4-amino-acetanilide.

Suitable diluents for process variants (a) and (b) are inert organic solvents. Preferably, dimethylformamide or tetrahydrofuran is used. At times it is also advantageous to carry out the process in aqueous suspension.

Suitable acid-binding agents are bases, such as alkali metal hydroxides, alkali metal carbonates or alkali metal hydrides. The use of potassium hydroxide or sodium hydride is preferred.

The reaction can also be carried out in a two-phase system with the aid of phase transfer catalysts such as alkyl-ammonium salts or alkylphosphonium salts. In that case, the aqueous phase contains the acid-binding agent (for example potassium hydroxide) and the organic phase (for example toluene) contains the starting materials.

The reaction temperatures can be varied within a substantial range. In general, the reaction is carried out at from -10° to $+100^{\circ}\text{C}$, preferably at from 10° to 40°C .

Usually, the reactants are employed in equimolar amounts, but one or other component can also be used in excess.

As already mentioned, the compounds according to the invention are active as pesticides. They have a powerful fungitoxic and bacteriotoxic action. They do not damage crop plants in the concentrations required for combating fungi and bacteria. For these reasons, they are suitable for use as plant protection agents for combating fungi and bacteria. Fungitoxic agents in plant protection are employed for combating *Plasmodiophoromycetes*, *Oomycetes*, *Chytridiomycetes*, *Zygomycetes*, *Ascomycetes*, *Basidiomycetes* and *Deuteromycetes*.

The active compounds according to the invention have a broad spectrum of action and can be used against parasitic fungi which attach above-ground parts of plants or attack the plants through the soil, as well as against seed-borne pathogens.

The active compounds according to the invention exhibit a particularly good activity against parasitic fungi which attack above-ground parts of plants, such as rust diseases of cereals, caused by species of *Puccinia*, and bean rust (*Uromyces phaseoli*), as well as against powdery mildew caused by species of *Erysiphe*, and powdery mildew of apples (*Podosphaera leucotricha*) and, in the case of rice, against *Pyricularia oryzae* and *Pellicularia sasakii*. On above-ground parts of plants, the compounds are also active against species of *Botrytis*, species of *Septoria*, species of *Helminthosporium* and species of *Cercospora*. The active compounds are effective and of particular practical importance when they are used as seed dressings or soil treatment agents against phytopathogenic fungi which adhere to the seed and occur in the soil, and cause seedling diseases, root rot, tracheomycoses and seed diseases in crop plants, such as species of *Fusarium*, species of *Rhizoctonia*, *Verticillium albo-atrum* and *Phialophora cinerescens*.

When used in higher amounts, the active compounds according to the invention are also herbicidally active.

The active compounds are well tolerated by plants, have a favourable level of toxicity to warm-blooded animals, and can be used for combating arthropod pests, especially insects and acarids, which are encountered in agriculture, in forestry, in the protection of stored products and of materials, and in the hygiene field. They are active against normally sensitive and resistant species and against all or some stages of development. The abovementioned pests include:

from the class of the *Isopoda*, for example *Oniscus asellus*, *Armadillidium vulgare* and *Porcellio scaber*;

from the class of the *Diplopoda*, for example *Blaniulus guttulatus*;

from the class of the *Chilopoda*, for example *Geophilus carpophagus* and *Scutigera spec.*;

from the class of the *Symphyla*, for example *Scutigera immaculata*;

from the order of the *Thysanura*, for example *Lepisma saccharina*;

from the order of the *Collembola*, for example *Onychiurus armatus*;

from the order of the *Orthoptera*, for example *Blatta orientalis*, *Periplaneta americana*, *Leucophaea maderae*, *Blattella germanica*, *Acheta domesticus*, *Grylotalpa* spp., *Locusta migratoria migratorioides*, *Melanoplus differentialis* and *Schistocerca gregaria*;

from the order of the *Dermaptera*, for example *Forficula auricularia*;

from the order of the *Isoptera*, for example *Reticulitermes* spp.;

from the order of the *Anoplura*, for example *Phylloxera vastatrix*, *Pemphigus* spp., *Pediculus humanus corporis*, *Haematopinus* spp. and *Linognathus* spp.;

from the order of the *Mallophaga*, for example *Trichodectes* spp. and *Damalinea* spp.;

from the order of the *Thysanoptera*, for example *Hercinothrips femoralis* and *Thrips tabaci*;

from the order of the *Heteroptera*, for example *Eurygaster* spp., *Dysdercus intermedius*, *Piesma quadrata*, *Cimex lectularius*, *Rhodnius prolixus* and *Triatoma* spp.;

from the order of the *Homoptera*, for example *Aleurodes brassicae*, *Bemisia tabaci*, *Trialeurodes vaporariorum*, *Aphis gossypii*, *Brevicoryne brassicae*, *Cryptomyzus ribis*, *Doralis fabae*, *Doralis pomi*, *Eriosoma lanigerum*, *Hyalopterus arundinis*, *Macrosiphum avenae*, *Myzus* spp., *Phorodon humuli*, *Rhopalosiphum padi*, *Empoasca* spp., *Euscelis bilobatus*, *Nephotettix cincticeps*, *Lecanium corni*, *Saissetia oleae*, *Laodelphax striatellus*, *Nilaparvata lugens*, *Aonidiella aurantii*, *Aspidiotus hederiae*, *Pseudococcus* spp. and *Psylla* spp.;

from the order of the *Lepidoptera*, for example *Pectinophora gossypiella*, *Bupalus piniarius*, *Cheimatobia brumata*, *Lithocolletis blancardella*, *Hyponomeuta padella*, *Plutella maculipennis*, *Malacosoma neustria*, *Euproctis chrysoorhoea*,

- 5 *Lymantria* spp., *Bucculatrix thurberiella*, *Phyllonistis citella*, *Agrotis* spp., *Euxoa* spp., *Feltia* spp., *Earias insulana*, *Helithis* spp., *Laphygma exigua*, *Mamestra brassicae*, *Panolis flammea*, *Prodenia Litura*, *Spodoptera* spp., *Trichopulsia ni*,
5 *Carpocapsa pomonella*, *Pieris* spp., *Chilo* spp., *Pyrausta nubilalis*, *Ephestia kuehniella*,
Galleria mellonella, *Cacoecia podana*, *Capua reticulana*, *Choristoneura fumiferana*,
Clysia ambiguella, *Homona magnanima* and *Tortrix viridana*; 5
- 10 from the order of the *Coleoptera*, for example *Anobium punctatum*, *Rhizopertha dominica*, *Bruchidius obtectus*, *Acanthoscelides obtectus*, *Hylotrupes bajulus*, *Agelastica alni*, *Leptinotarsa decemlineata*, *Phaedon cochleriae*, *Diabrotica* spp., *Psylliodes chrysocephala*, *Epilachna varivestis*, *Atomaria* spp., *Oryzaephilus surinamensis*,
10 *Anthonomus* spp., *Sitophilus* spp., *Otiorrhynchus sulcatus*, *Cosmopholites sordidus*,
Ceuthorrhynchus assimilis, *Hypera postica*, *Dermestes* spp., *Trogoderma* spp.,
Anthrenus spp., *Attagenus* spp., *Lyctus* spp., *Meligethes aeneus*, *Ptinuu* spp., *Niptus hololeucus*, *Gibbium psylloides*, *Tribolium* spp., *Tenebrio molitor*, *Agriotes* spp.,
15 *Conoderus* spp., *Melolontha melolontha*, *Amphimallon solstitialis* and *Costelytra zealandica*;
15 from the order of the *Hymenoptera*, for example *Diprion* spp., *Hoplocampa* spp.,
Lasius spp., *Monomorium pharaonis* and *Vespa* spp.;
- 20 from the order of the *Diptera*, for example *Aedes* spp., *Anopheles* spp., *Culex* spp., *Brosophilia melanogaster*, *Musca* spp., *Fannia* spp., *Calliphora erythrocephala*,
20 *Lucilia* spp., *Chrysomyia* spp., *Cuterebra* spp., *Gastrophilus* spp., *Hyppobosca* spp.,
Stomoxys spp., *Oestrus* spp., *Hypoderma* spp., *Tabanus* spp., *Tannia* spp., *Biblio hortulanus*, *Oscinella frit*, *Phorbia* spp., *Pegomyia hyoscyami*, *Ceratitis capitata*, *Dacus oleae* and *Tipula paludosa*;
25 from the order of the *Siphonaptera*, for example *Xenopsylla cheopis* and
25 *Ceratophyllus* spp.;
- 30 from the class of the *Arachnida*, for example *Scorpio maurus* and *Latrodectus mactans*;
30 from the order of the *Acarina*, for example *Acarus siro*, *Argas* spp., *Ornithodoros* spp.,
Dermanyssus gallinae, *Eriophyes ribis*, *Phyllocoptruta oleivora*, *Boophilus* spp.,
30 *Rhipicephalus* spp., *Amblyomma* spp., *Hyalomma* spp., *Ixodes* spp., *Psoroptes* spp.,
Choriotes spp., *Sarcoptes* spp., *Tarsonemus* spp., *Bryobia praetiosa*, *Panonychus* spp.
and *Tetranychus* spp.
- 35 In low use concentrations, the active compounds according to the invention exhibit a development-inhibiting activity on some or all stages of development of
35 insects.
- 40 The active compounds can be converted into the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, dusting agents, foams, pastes, soluble powders, granules, aerosols, suspension-emulsion concentrates, seed-treatment powders, natural and synthetic materials impregnated with active compound, very fine capsules in polymeric substances, coating compositions for use on seed, and formulations used with burning equipment, such as fumigating cartridges, fumigating cans and fumigating coils, as well as ULV cold mist and warm mist fomulations.
45
- 50 These formulations may be produced in known manner, for example by mixing the acting compounds with extenders, that is to say liquid or liquefied gaseous or solid diluents or carriers, optionally with the use of surface-active agents, that is to say emulsifying agents and/or dispersing agents and/or foam-forming agents. In the case of the use of water as an extender, organic solvents can, for example, also be used as auxiliary solvents.
50
- 55 As liquid solvents diluents or carriers, especially solvents, there are suitable in the main, aromatic hydrocarbons, such as xylene, toluene or alkyl naphthalenes, chlorinated aromatic or chlorinated aliphatic hydrocarbons, such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic or alicyclic hydrocarbons, such as cyclohexane or paraffins, for example mineral oil fractions, alcohols, such as butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, or strongly polar solvents, such as dimethylformamide and dimethylsulphoxide, as well as water.
55
- 60 By liquefied gaseous diluents or carriers are meant liquids which would be gaseous at normal temperature and under normal pressure, for example aerosol propellants, such as halogenated hydrocarbons as well as butane, propane, nitrogen and carbon dioxide.
60
- 65 As solid carriers there may be used ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgitte montmorillonite or diatomaceous earth, and
65

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

As emulsifying and/or foam-forming agents there may be used non-ionic and anionic emulsifiers, such as polyoxyethylene-fatty acid esters, polyoxyethylene-fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkyl sulphonates, alkyl sulphates, aryl sulphonates as well as albumin hydrolysis products. Dispersing agents include, for example, lignin sulphite waste liquors and methylcellulose.

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

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

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

The active compounds according to the invention may be used in the form of their formulations of the types that are commercially available or in the use forms prepared from these formulations.

The active compound content of the use forms prepared from the formulations of the types that are commercially available can vary within wide ranges. The active compound concentration of the use forms can be from 0.0000001 to 100% by weight of active compound, preferably from 0.01 to 10% by weight.

The compounds may be employed in a customary manner appropriate for the particular use forms.

When used against pests harmful to health and pests of stored products, the active compounds are distinguished by an excellent residual activity on wood and clay as well as a good stability to alkali on limed substrates.

The present invention also provides an arthropodicidal, fungicidal or bactericidal composition containing as active ingredient a compound of the present invention in admixture with a solid or liquefied gaseous diluent or carrier or in admixture with a liquid diluent or carrier containing a surface-active agent.

The present invention also provides a method of combating arthropods (especially insects or acarids), fungi or bacteria which comprises applying to the arthropods, fungi or bacteria, or to a habitat thereof, a compound of the present invention alone or in the form of a composition containing as active ingredient a compound of the present invention in admixture with a diluent or carrier.

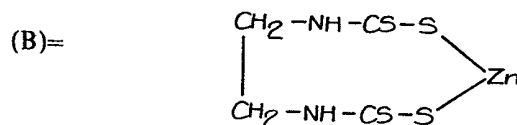
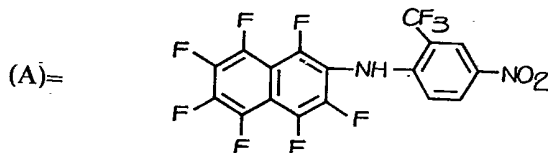
The present invention further provides crops protected from damage by arthropods, fungi or bacteria by being grown in areas in which immediately prior to and/or during the time of the growing a compound of the present invention was applied alone or in admixture with a diluent or carrier.

It will be seen that the usual methods of providing a harvested crop may be improved by the present invention.

The pesticidal activity of the compounds of this invention is illustrated by the following biotest Examples.

In these Examples, the compounds according to the present invention are each identified by the number (given in brackets) of the corresponding preparative Example, which will be found later in this specification.

The known comparison compounds are identified as follows:



Example A

Phaedon larvae test

Solvent: 3 parts by weight of dimethylformamide

Emulsifier: 1 part by weight of alkylaryl polyglycol ether.

5 To produce a suitable preparation of active compound, 1 part by weight of the active compound was mixed with the stated amount of solvent containing the stated amount of emulsifier and the concentrate was diluted with water to the desired concentration. 5

10 Cabbage leaves (*Brassica oleracea*) were sprayed with the preparation of the active compound until dripping wet and were then infested with mustard beetle larvae (*Phaedon cochleariae*). 10

After the specified periods of time, the degree of destruction was determined in %: 100% meant that all of the beetle larvae had been killed whereas 0% meant that none of the beetle larvae had been killed.

15 The active compounds, the concentrations of the active compounds, the valuation times and the results can be seen from the following table: 15

TABLE A
(Insects which damage plants)
Phaedon larvae test

Active compounds (A)	Active compound concentration in %	Degree of destruction in % after 3 days
	0.1	20
	0.01	0
(36)	0.1	100
	0.01	100
(6)	0.1	100
	0.01	70
(11)	0.1	100
	0.01	90
(37)	0.1	100
	0.01	100
(45)	0.1	100
	0.01	100
(49)	0.1	100
	0.01	100
(4)	0.1	100
	0.01	95
(9)	0.1	100
	0.01	90
(25)	0.1	100
	0.01	90
(23)	0.1	100
	0.01	100
(29)	0.1	100
	0.01	100
(8)	0.1	100
	0.01	90
(26)	0.1	100
	0.01	95

Example B

Tetranychus test (resistant)

Solvent: 3 parts by weight of dimethylformamide

Emulsifier: 1 part by weight of alkylaryl polyglycol ether.

50 To produce a suitable preparation of active compound, 1 part by weight of the active compound was mixed with the stated amount of solvent and the stated amount of emulsifier and the concentrate was diluted with water to the desired concentration. 55

60 Bean plants (*Phaseolus vulgaris*) which were heavily infested with the two-spotted spider mite (*Tetranychus urticae*) in all stages of development were sprayed with the preparation of the active compound until dripping wet. 60

After the specified periods of time, the degree of destruction was determined as a percentage: 100% meant that all of the spider mites were killed whereas 0% meant that none of the spider mites were killed.

The active compounds, the concentrations of the active compounds, the evaluation times and the results can be seen from the following table:

TABLE B
(Mites which damage plants)

Tetranychus test

Active compounds	Active compound concentration in %	Degree of destruction in % after 2 days
(A)	0.1	0
(44)	0.1	100
(45)	0.1	100
(4)	0.1	99

Example C

Fusicladium test (apple)/protective
Solvent: 4.7 parts by weight of acetone
Emulsifier: 0.3 part by weight of alkylaryl polyglycol ether
Water: 95 parts by weight.

The amount of active compound required for the desired concentration of the active compound in the spray liquid was mixed with the stated amount of solvent, and the concentrate was diluted with the stated amount of water which contained the stated amount of emulsifier.

Young apple seedlings in the 4—6 leaf stage were sprayed with the spray liquid until dripping wet. The plants remained in a greenhouse for 24 hours at 20 degrees C and at a relative atmospheric humidity of 70%. They were then inoculated with an aqueous conidium suspension of the apple scab causative organism (*Fusicladium dendriticum*) and incubated for 18 hours in a humidity chamber at 18—20 degrees C and at a relative atmospheric humidity of 100%.

The plants were then brought into a greenhouse again for 14 days. 15 days after inoculation, the infection of the seedlings was determined. The assessment data were converted to percent infection. 0% meant no infection; 100% meant that the plants were totally infected.

The active compounds, the concentrations of the active compounds and the results can be seen from the following table:

TABLE C
Fusicladium test (apple)/protective

Active compound	Infection in % at an active compound concentration of 0.0025%
(A)	32
(26)	19
(4)	2
(36)	20
(6)	7
(9)	20
(7)	2
(33)	17

Example D

Uromyces test (beanrust)/protective
Solvent: 4.7 parts by weight of acetone
Emulsifier: 0.3 part by weight of alkylaryl polyglycol ether
Water: 95 parts by weight.

The amount of active compound required for the desired concentration of active compound in the spray liquor was mixed with the stated amount of the solvent and the concentrate was diluted with the stated amount of water which contained the stated amount of emulsifier.

The young bean plants, which were in the 2-leaf stage, were sprayed with the

spray liquor until dripping wet. The plants remained in a greenhouse for 24 hours at 20—22 deg. C and a relative atmospheric humidity of 70% in order to dry. They were then inoculated with an aqueous uredospore suspension of the causative organism of bean rust (*Uromyces phaseole*) and incubated for 24 hours in a dark humidity chamber at 20—22 deg. C and 100% relative atmospheric humidity.

The plants were then set up in a greenhouse under intensive illumination for 9 days at 20—22 deg. C and a relative atmospheric humidity of 70—80%.

10 days after the inoculation, the infection of the plants was determined. The assessment data were converted to % infection. 0% denoted no infection and 100% denoted that the plants were completely infected.

The active compounds, active compound concentrations and results can be seen from the following table:

TABLE D
Uromyces test/protective

Active compound	Infection in % at an active compound concentration of 0.001%
(A)	19
(26)	2
(4)	9
(30)	12

Example E

Phytophthora test (tomato)/protective

Solvent: 4.7 parts by weight of acetone

Emulsifier: 0.3 part by weight of alkylaryl polyglycol ether

Water: 95 parts by weight.

The amount of the active compound required for the desired concentration of the active compound in the spray liquid was mixed with the stated amount of solvent and the concentrate was diluted with the stated amount of water which contained the stated additions.

Young tomato plants with 2 to 4 foliage leaves were sprayed with the spray liquid until dripping wet. The plants remained in a greenhouse for 24 hours at 20 deg. C and at a relative atmospheric humidity of 70%. The tomato plants were then inoculated with an aqueous spore suspension of *Phytophthora infestans*. The plants were brought into a moist chamber with an atmospheric humidity of 100% and a temperature of 18—20 deg. C.

After 5 days the infection of the tomato plants was determined. The assessment data were converted to percent infection: 0% meant no infection; 100% meant that the plants were totally infected.

The active compounds, the concentrations of the active compound and the results can be seen from the following table:

TABLE E
Phytophthora test (tomatoes)/protective

Active compound	Infection in % at an active compound concentration of 0.001%
(B)	44
(26)	1

Example F

Shoot treatment test/cereal rust (leaf-destructive mycosis)/protective

To produce a suitable preparation of active compound, 0.25 part by weight of active compound was taken up in 25 parts by weight of dimethylformamide and 0.06 part by weight of alkylaryl polyglycol ether and then 975 parts by weight of water were added. The concentrate was diluted with water to the desired final concentration of the spray liquor.

To test the protective activity, one-leaved young wheat plants of the Michigan Amber variety were inoculated with a uredospore suspension of *Puccinia recondita* in 0.1% strength aqueous agar. After the spore suspension had dried on, the wheat

plants were sprayed with the preparation of active compound until dew-moist and were placed, for incubation, in a greenhouse for 24 hours at about 20 deg. C and 100% relative atmospheric humidity.

After 10 days' dwell time of the plants at a temperature of 20 deg. C and 80—90% atmospheric humidity, the occurrence of rust pustules on the plant was evaluated. The degree of infection was expressed as a percentage of the infection of the untreated control plants. 0% denoted no infection and 100% denoted the same degree of infection as in the case of the untreated control. The active compound was the more active, the lower was the degree of rust infection.

The active compounds active compound concentrations in the spray liquor and degrees of infection can be seen from the table which follows:

TABLE F
Shoot treatment test/cereal rust/protective

Active compounds untreated	Active compound concentration in the spray liquor in % by weight	Infection in % of the untreated control
(A)	0.01	50.0
(23)	0.01	27.5
(4)	0.01	21.3
(36)	0.01	33.8
(37)	0.01	5.0
(45)	0.01	3.8
(49)	0.01	33.8
(6)	0.01	21.3
(8)	0.01	21.3
(9)	0.01	28.8
(3)	0.01	28.8
(38)	0.01	16.3

Example G

Pyricularia and *Pellicularia* test

Solvent: 11.75 parts by weight of acetone

Dispersing agent: 0.75 parts by weight of alkylaryl polyglycol ether

Water: 987.50 parts by weight.

The amount of active compound required for the desired concentration of active compound in the spray liquid was mixed with the stated amount of the solvent and of the dispersing agent and the concentrate was diluted with the stated amount of water.

2×30 rice plants about 2—4 weeks old were sprayed with the spray liquid until dripping wet. The plants remained in a greenhouse at temperatures of 22 to 24 deg. C and a relative atmospheric humidity of about 70% until they were dry. Thereafter, the plants were inoculated with an aqueous suspension of 100,000 to 200,000 spores/ml of *Pyricularia oryzae* and placed in a chamber at 24 to 26 deg. C and 100% relative atmospheric humidity.

Other rice plants which had been sprayed and dried in the above-described manner, were infected with a culture of *Pellicularia sasakii* grown on malt agar and were set up at 28 to 30 deg. C and 100% relative atmospheric humidity.

5 to 8 days after the inoculation, the infection of all the leaves present at the time of inoculation with *Pyricularia oryzae* was determined as a percentage of the untreated but also inoculated control plants.

In the case of the plants infected with *Pellicularia sasakii*, the infection at the leaf sheaths after the same time was determined, again in relation to the untreated but infected control.

The evaluation was made on a scale of from 1 to 9. 1 denoted 100% action, 3 denoted good action, 5 denoted moderate action and 9 denoted no action.

The active compounds, the concentrations of the active compounds and the results can be seen from the table which follows.

TABLE G
Pyricularia (a) and *Pellicularia* (b) test

	Active compound	Action at an active compound concentration of 0.025%		
		(a)	(b)	
5	(A)	5	2	5
	(4)	—	2	
	(36)	3	—	
	(37)	—	1	
10	(45)	—	1	10
	(49)	5	2	
	(8)	—	1	
	(9)	3	—	
	(10)	3	—	

Example H

15 Mycelium growth test 15

Nutrient medium used:

20 parts by weight of agar-agar
 200 parts by weight of potato decoction
 5 parts by weight of malt
 15 parts by weight of dextrose
 5 parts by weight of peptone
 2 parts by weight of disodium hydrogen phosphate
 0.3 part by weight of calcium nitrate.

20 20

25 Ratio of solvent mixture to nutrient medium:
 2 parts by weight of solvent mixture
 100 parts by weight of agar nutrient medium. 25

Composition of the solvent mixture:

0.19 part by weight of dimethylformamide
 0.01 part by weight of emulsifier (alkylaryl polyglycol ether)
 1.80 parts by weight of water.

30 30

The amount of active compound required for the desired active compound concentration in the nutrient medium was mixed with the stated amount of solvent mixture. The concentrate was thoroughly mixed, in the stated proportion, with the liquid nutrient medium (which had been cooled to 42 deg. C) and was then poured into Petri dishes of 9 cm diameter. Control plates to which the preparation had not been added were also set up.

35 35

When the nutrient medium had cooled and solidified, the plates were inoculated with the species of fungi stated in the table and incubated at about 21 deg. C.

40 40

Evaluation was carried out after 4—10 days, dependent upon the speed of growth of the fungi. When evaluation was carried out the radial growth of the mycelium on the treated nutrient media was compared with the growth on the control nutrient medium. In the evaluation of the fungus growth, the following characteristic values were used:

45 45

1 no fungus growth
 up to 3 very strong inhibition of growth
 up to 5 medium inhibition of growth
 up to 7 slight inhibition of growth
 9 growth equal to that of untreated control.

50 50

The active compounds, the active compound concentrations and the results can be seen from the following table:

Active compounds (concentration =10 ppm)	Mycelium growth test		Fungi and 1 bacterium	
(B)	9	9	9	9
(23)	5	5	5	5
(29)	—	5	—	—
(4)	5	1	—	—
(36)	5	1	—	—
(37)	—	1	—	—
(49)	3	1	3	3
(8)	3	1	5	2
(9)	3	1	5	1
(10)	—	1	—	—
(11)	5	2	—	—
	<i>Fusarium culmorum</i>		<i>Pyricularia oryzae</i>	
	<i>Sclerotinia sclerotiorum</i>		<i>Phialophora cinerescens</i>	
	<i>Fusarium nivale</i>		<i>Helminthosporium gramineum</i>	
	<i>Colletotrichum coffeanum</i>		<i>Mycosphaerella musicola</i>	
	<i>Rhizoctonia solani</i>		<i>Phytophthora cactorum</i>	
	<i>Pythium ultimum</i>		<i>Venturia inaequalis</i>	
	<i>Cochliobolus miyabeanus</i>		<i>Pellicularia sasakii</i>	
	<i>Botrytis cinerea</i>		<i>Xanthomonas oryzae</i>	
	<i>Verticillium alboatrum</i>			

TABLE H

Example J

Bacteria test/*Xanthomonas oryzae*

Solvent: 11.75 parts by weight of acetone

Dispersing agent: 0.75 part by weight of alkylaryl polyglycol ether

Water: 987.50 parts by weight.

5

5

The amount of active compound required for the desired active compound concentration in the spray liquor was mixed with the stated amount of the solvent and of the dispersing agent and the concentrate was diluted with the stated amount of water.

10

30 rice plants which were about 40 days old were sprayed with the spray liquor until dripping wet. The plants remained in a greenhouse, at temperatures of 22 to 24 deg. C and a relative atmospheric humidity of about 70%, until they had dried. Needles were then dipped into an aqueous bacterial suspension of *Xanthomonas oryzae* and the plants were inoculated by pricking the leaves. After the inoculation, the leaves stood for 24 hours at 100% relative atmospheric humidity and thereafter in a room at 26 to 28 deg. C and 80% relative atmospheric humidity.

10

15

15

10 days after the inoculation, the infection of all pricked inoculated leaves of plants which had beforehand been treated with the preparation was evaluated, using a scale of from 1 to 9. 1 denoted 100% action, 3 denoted a good action, 5 denoted a moderate action and 9 denoted no action.

20

20

The active compounds, active compound concentrations and results can be seen from the table which follows:

TABLE J
Bacteria test/*Xanthomonas oryzae*

25

25

Active compound	Action at an active compound concentration of 0.025%
(A)	9
(37)	3
(1)	5
(5)	5
(32)	3
(33)	3
(31)	5
(3)	5
(42)	5
(43)	5
(2)	5
(46)	3

30

30

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35

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40

The experiments given below show the arthropod development-inhibiting action of the compounds according to the invention without intending to imply a limitation in respect of the breadth of action of these compounds. In these tests, the morphological changes, such as partially pupated animals, incompletely slipped larvae or caterpillars, defective wings and pupal cuticula in imagines, were assessed as mis-developments over the entire stated development of the test animals.

45

45

Example K

Development-inhibiting action/ingestion test

Test animals: *Plutella maculipennis* (caterpillars in the 4th stage of development, 20 specimens)Feed plants: Cabbage plants (*Brassica oleracea*)

Solvent: 4 parts by weight of acetone

Emulsifier: 1 part by weight of alkylaryl polyglycol ether.

50

50

To produce a suitable preparation of active compound, 2 parts by weight of active compound were mixed with the stated amount of solvent, emulsifier and sufficient water to give a 1% strength mixture, which was diluted with water to the desired concentration.

55

55

The test animals were fed with leaves of the feed plants, which were provided with a uniform spray covering of the active compound mixture of the desired concentrations, so that the stated amounts of active compound in ppm (parts per million) were obtained on the leaves, until the image developed.

As a control, leaves provided only with solvent and emulsifier of the stated concentration were used as the feed.

The sum of the morphological malformations and of the destruction was determined as a percentage. 100% meant that all of the animals were either killed or morphologically damaged whilst 0% meant that no damage had taken place.

The results can be seen from the table which follows:

TABLE K
Development-inhibiting action/ingestion test

10	Active compound	<i>Plutella macul.</i>		10
		10 ppm	1 ppm	
	Control (untreated)	0%	0%	
	(A)	100%	50%	
	(26)	100%	100%	
	(27)	100%	100%	
15	(28)	100%	100%	15
	(29)	100%	70%	
	(30)	100%	70%	
	(35)	100%	60%	
	(4)	100%	100%	
20	(36)	100%	100%	20
	(22)	100%	100%	

Example L

Development-inhibiting action

Test animal: *Ceratitis capitata* (20 eggs)

Feed: Artificial feed (carrot powder and yeast powder)

Solvent: 20 parts by weight of acetone

Emulsifier: 5 parts by weight of alkylaryl polyglycol ether.

To produce a suitable preparation of active compound, 4 parts by weight of active compound were mixed with the stated amount of solvent, emulsifier and sufficient water to give a 1% strength mixture which was diluted with water to the desired concentration.

The eggs of the test animals were placed on a dish with the artificial feed, into which the chosen concentration of the active compound mixture had been incorporated so that the stated amount of active compound in ppm (parts per million) was achieved. The development was observed until the flies slipped.

As a control, artificial feed mixed only with solvent and emulsifier of the stated concentration was offered.

The sum of the morphological malformations and of the destruction was determined as a percentage. 100% meant that all of the animals were either killed or morphologically damaged whilst 0% meant that no damage had taken place.

The results can be seen from the table which follows:

TABLE L
Development-inhibiting action

Test animals: *Ceratitis capitata*

45	Active compound	Damage at a concentration of 10 ppm	45
	(A)	20%	
	(22)	100%	
50	(26)	100%	50
	(30)	100%	

Example M

Development-inhibiting action

Test animal: *Aedes aegypti* (larvae in the 3rd stage of development, 20 specimens)

Solvent: 10 parts by weight of acetone

Emulsifier: 1 part by weight of polyoxyethylene-(20) sorbitan monolaurate.

To produce a suitable preparation of active compound, 2 parts by weight of active compound were mixed with the stated amount of solvent, emulsifier and

sufficient water to produce a mixture containing 10 ppm, which was diluted with water to the desired concentration.

The test animals were introduced into 90 ml of these active compound solutions and were observed until the imago slipped. As a control, test animals were introduced into a solvent/emulsifier/water mixture of the stated concentration and observed until the imago slipped.

The sum of the morphological malformations and of the destruction was determined as a percentage. 100% meant that all of the animals were either killed or morphologically damaged whilst 0% meant that no damage had taken place.

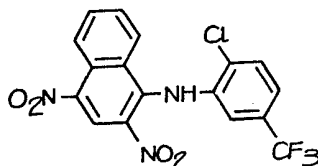
The results can be seen from the table which follows:

TABLE M
Development-inhibiting action/mosquito test
Test animals: *Aedes aegypti*

Active compound	Damage at a concentration of 0.1 ppm
Control (untreated)	0%
(A)	0%
(25)	100%
(26)	100%
(28)	100%
(30)	100%

Preparative examples

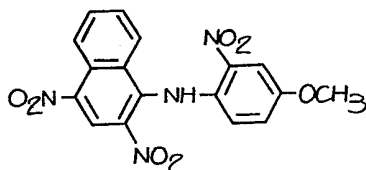
Example 1



(1)

6.0 g (107 m mols) of pulverulent potassium hydroxide were added, in portions, to a solution of 11.73 g (60 m mols) of 3 - amino - 4 - chloro - benzotrifluoride in 60 ml of dry dimethylformamide at room temperature. A solution of 15.15 g (60 m mols) of 1 - chloro - 2,4 - dinitronaphthalene in 100 ml of dry dimethylformamide was then added dropwise at 25°C. After stirring for three days at room temperature the mixture was filtered and the filtrate was diluted with 50 ml of glacial acetic acid and poured out onto water/ice. 17.6 g of a yellow product were filtered off, from which 11.8 g of 1 - (2 - chloro - 5 - trifluoromethyl - phenylamino) - 2,4 - dinitronaphthalene were obtained, in the form of golden yellow flakes, by recrystallisation from isopropanol. Melting point 153°C.

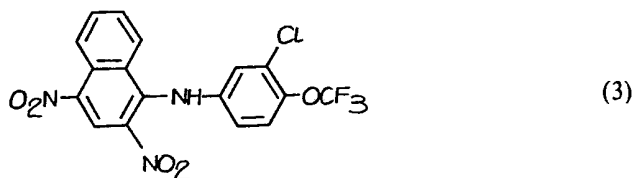
Example 2



(2)

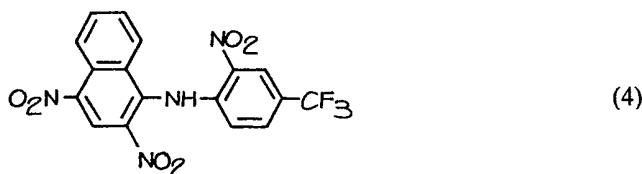
A mixture of 3.70 g (66 m mols) of potassium hydroxide and 3.71 g (10 m mols) of triphenylethylphosphonium bromide in 50 ml of water, together with 15.15 g (60 m mols) of 1-chloro-2,4-dinitronaphthalene and 10.2 g (60 m mols) of 3-nitro-4-aminoanisole in 150 ml of toluene was stirred for 8 hours at 80°C. 20 g of potassium hydroxide were added and stirring was continued for 5 hours at 80°C. After the mixture had cooled, a dark powder was filtered off, washed with water and water/acetone (12:1) and dissolved in 100 ml of hot glacial acetic acid. A few drops of concentrated hydrochloric acid were added and after cooling 15.0 g of 1 - (2 - nitro - 4 - methoxy - phenylamino) - 2,4 - dinitronaphthalene of melting point 173°C were isolated. Molecular weight 384 (determined by mass spectrometry).

Example 3



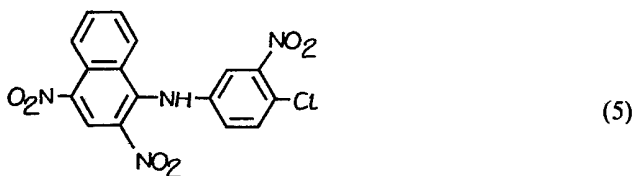
5 A suspension of 12.7 g (60 m mols) of 3-chloro-4-trifluoromethoxyaniline and 15.15 g (60 m mols) of 1-chloro-2,4-dinitronaphthalene in 150 ml of water was heated to the boil. 5.6 g (67 m mols) of sodium bicarbonate were introduced in the course of 30 minutes and heating was continued for 2 hours under reflux. After the mixture had cooled to 20°C, the product was filtered off. 24 g of 1 - (3 - chloro - 4 - trifluoromethoxyphenylamino) - 2,4 - dinitronaphthalene of melting point 140°C were obtained; after recrystallisation from ethanol, 17.4 g of melting point 159°C were obtained.

Example 4



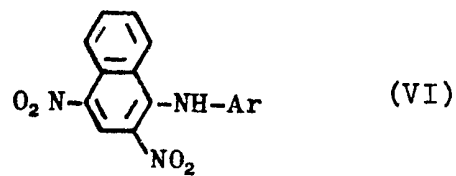
15 A solution of 14.0 g of 2,4-dinitro-1-naphthylamine in 50 ml of dimethylformamide was added dropwise, at 0°C, to a suspension of 2.88 g of 80% strength sodium hydride (20% paraffin) in 30 ml of dry dimethylformamide. The mixture was allowed to come to room temperature and was stirred for 1 hour. A solution of 12.55 g of 4-fluoro-3-nitrobenzotrifluoride in 30 ml of dimethylformamide was then added dropwise at 10°C. The batch was stirred overnight at room temperature and a further 0.3 g of 80% strength sodium hydride and 1.25 g of 4-fluoro-3-nitrobenzotrifluoride were added. After stirring for 24 hours, the mixture was filtered, 500 ml of glacial acetic acid were added to the filtrate and the whole was poured out onto 400 g of ice. The yellow precipitate of 1 - (2 - nitro - 4 - trifluoromethyl - phenylamino) - 2,4 - dinitronaphthalene was filtered off and dried; 21.2 g of melting point 145°C. After recrystallisation from amyl alcohol, the melting point rose to 159—160°C.

Example 5

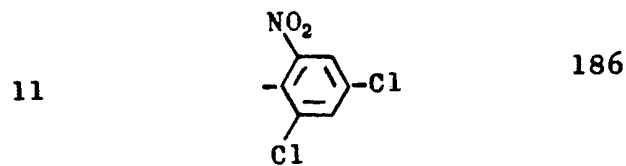
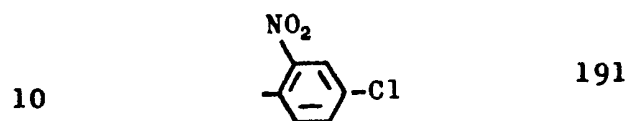
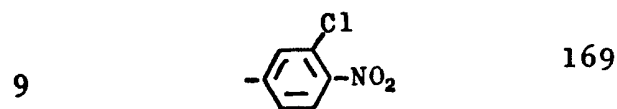
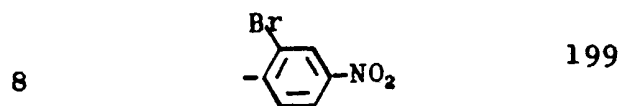
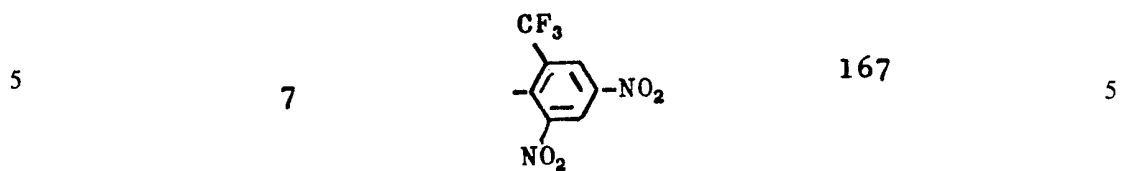
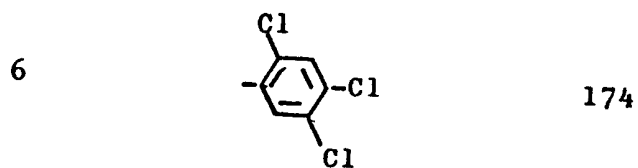


30 A solution of 17.3 g (0.1 mol) of 4-chloro-3-nitroaniline in 200 ml of dimethylformamide was added at 0°C to a suspension of 5 g of sodium hydride (coated with 20% of paraffin) in 100 ml of dimethylformamide. The mixture was stirred for 1 hour at 25°C and was then cooled to 10°C, and a solution of 25.3 g (0.1 mol) of 1-chloro-2,4-dinitronaphthalene in 250 ml of dimethylformamide was added dropwise. After standing overnight, the mixture was filtered, mixed with 500 ml of glacial acetic acid and poured onto ice. The crystals of 1 - (3 - nitro - 4 - chloro - phenylamino) - 2,4 - dinitronaphthalene were filtered off and washed with ether. Yield 25 g, melting point 182°C.

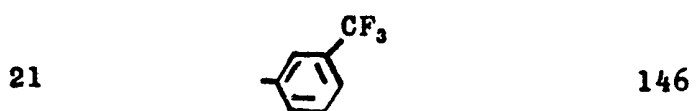
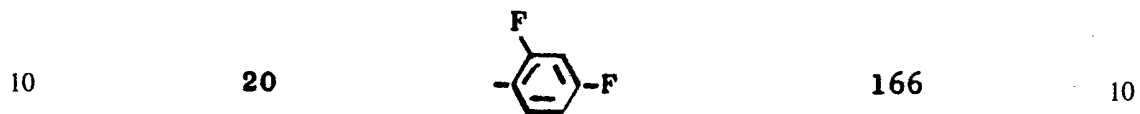
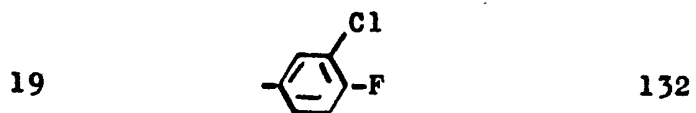
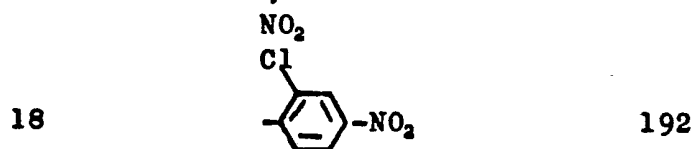
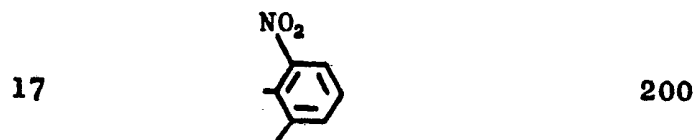
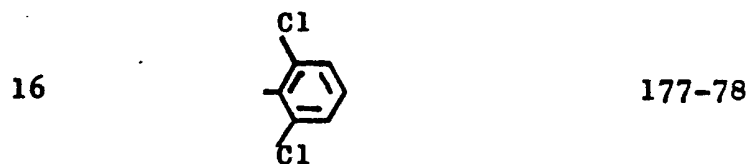
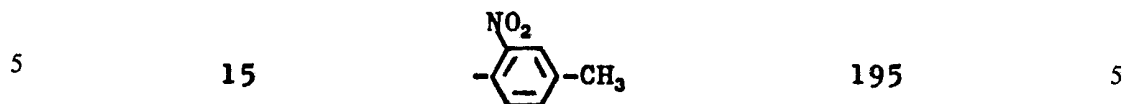
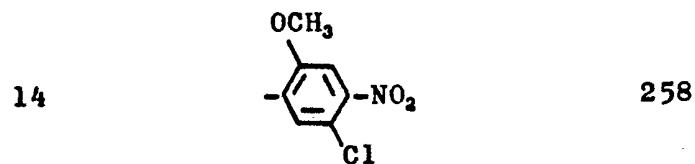
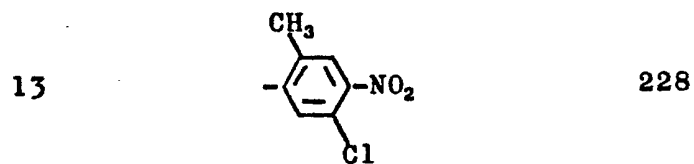
35 The following compounds could be prepared analogously to Example 5:

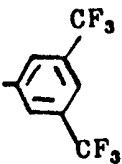
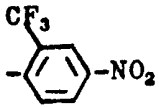
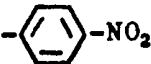
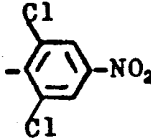
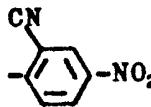
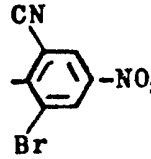
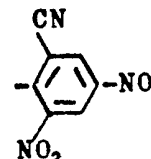
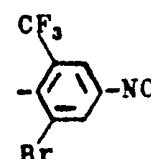
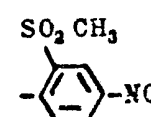


Example No.	Ar	Melting point (°C)
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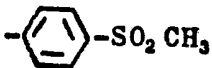


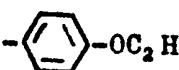
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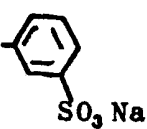


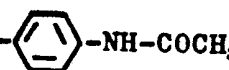
Example No.	Ar	Melting point (°C)		
22		183		
23		184-85		
5	24		219	5
25		210		
26		216		
27		212		
28		239		
10	29		196	10
30		248		

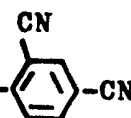
Example No.	Ar	Melting point (°C)
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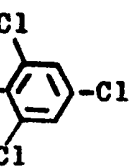
31		228
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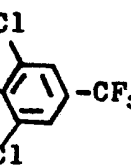
32		155-56
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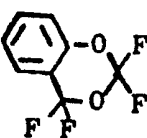
5	33		110-113 (free acid)	5
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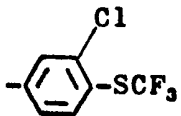
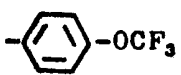
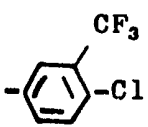
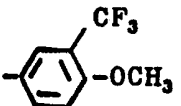
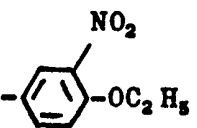
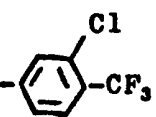
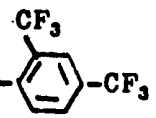
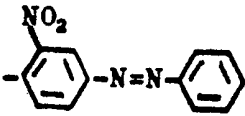
34		252-53
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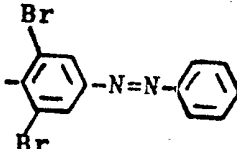
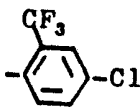
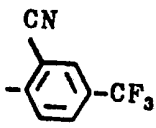
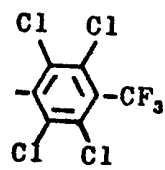
35		236
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36		199
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37		181
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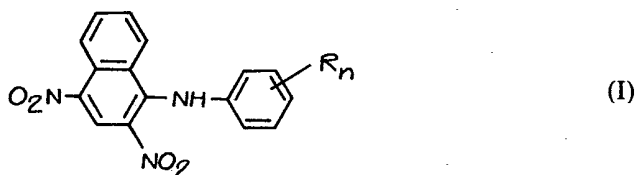
10	38		184	10
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Example No.	Ar	Melting point (°C)	
39		190	
40		159	
5 41		198	5
42		219-220	
43		208-209	
44		163-165	
45		165-167	
10 46		208-209	10

Example No.	Ar	Melting point (°C)
47		216-218
48		184
49		157-158
50		211

WHAT WE CLAIM IS:—

1. 1-arylamino-2,4-dinitronaphthalenes of the general formula



10 in which

n is 1, 2, 3, 4 or 5,

R represents hydrogen, alkyl, halogenoalkyl, alkenyl, alkynyl, alkoxy, alkylmercapto, alkylsulphonyl, sulphonate, halogenoalkoxy, halogenoalkylmercapto, acylamino, alkoxy-carbonyl, phenylazo, halogen, CN or nitro, the radicals R being selected independently of one another when n is 2 or more, and two radicals R in the ortho-position to one another can, conjointly with the two adjoining carbon atoms of the phenyl ring, form an optionally halogen-substituted 1,3-dioxane ring.

2. Compounds according to Claim 1, in which each R is selected independently from hydrogen, alkyl with 1-4 carbon atoms, alkoxy with 1-4 carbon atoms, alkylmercapto with 1-4 carbon atoms, alkylsulphonyl with 1-4 carbon atoms, sulphonate, halogenoalkyl, halogenoalkoxy and halogenoalkylmercapto, each with 1-4 carbon atoms and with 3-5 halogen atoms, acylamino, methoxycarbonyl, phenylazo, halogen, CN and NO_2 , or two radicals R in the ortho-position to one another, conjointly with the two adjoining carbon atoms of the phenyl ring, form a 1,3-dioxane ring which is optionally substituted by fluorine.

3. The compound according to Claim 1 that is disclosed in any one of Examples 1 to 50.

4. A process for the preparation of 1-arylamino-2,4-dinitronaphthalene according to Claim 1, in which

5 (a) a compound of the general formula



in which

X represents fluorine, chlorine, bromine or an aliphatic or aromatic sulphonic acid ester radical,

10 is reacted with an aniline of the general formula



in which

R and *n* have the meanings stated in Claim 1, in the presence of an acid-binding agent and, if appropriate, in the presence of a diluent, or

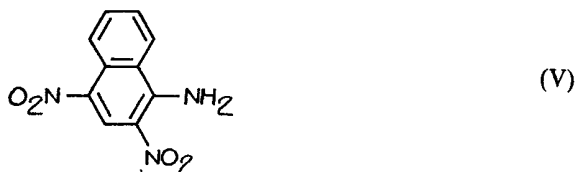
15 (b) a compound of the general formula



in which

X represent fluorine, chlorine, bromine or an aliphatic or aromatic sulphonic acid ester radical and

R and *n* have the meanings stated in Claim 1, is reacted with 2,4-dinitro-1-naphthylamine, of the formula



25 in the presence of an acid-binding agent and, if appropriate, in the presence of a diluent.

5. A process according to Claim 4, in which the reaction is effected in the presence of an inert organic solvent.

6. A process according to Claim 5, in which the solvent is dimethylformamide or tetrahydrofuran.

30 7. A process according to Claim 4, 5 or 6, in which the acid-binding agent is an alkali metal hydroxide, an alkali metal carbonate or alkali metal hydride.

8. A process according to Claim 7, in which the acid-binding agent is potassium hydroxide or sodium hydride.

35 9. A process according to any of Claims 4 to 8, in which the reaction is effected at from -10° to $+100^{\circ}\text{C}$.

10. A process according to Claim 9, in which the reaction is effected at from 10° to 40°C .

11. A process according to any of Claims 4 to 10 in which the reactants are employed in equimolar amounts.

40 12. A process according to any of Claims 4 to 11, in which variant (a) is employed and X is chlorine, each R represents hydrogen, nitro, cyano, methylsulphonyl, sulphonate, trifluoromethyl, trifluoromethylmercapto,

trifluoromethoxy, alkyl or alkoxy with 1—4 carbon atoms, acylamino, phenylazo, fluorine, chlorine or bromine, and n is 1, 2, 3 or 5.

5 13. A process according to any of Claims 4 to 12 in which variant (a) is employed and the compound (III) is one that is hereinbefore specifically mentioned. 5

14. A process for the preparation of a compound according to Claim 1, substantially as described in any of Examples 1 to 5.

10 15. Compounds according to Claim 1 whenever prepared by a process according to any of Claims 4 to 14. 10

16. An arthropodicidal, fungicidal or bactericidal composition containing as active ingredient a compound according to any of Claims 1 to 3 and 15 in admixture with a solid or liquefied gaseous diluent or carrier or in admixture with a liquid diluent or carrier containing a surface-active agent.

15 17. A composition according to Claim 16 containing from 0.1 to 95% of the active compound, by weight. 15

18. A method of combating arthropods, fungi or bacteria which comprises applying to the arthropods, fungi or bacteria, or to a habitat thereof, a compound according to any of Claims 1 to 3 and 15 alone or in the form of a composition containing as active ingredient a compound according to any of Claims 1 to 3 and 15, in admixture with a diluent or carrier. 20

19. A method according to Claim 18 in which a composition is used containing from 0.0000001 to 100% of the active compound, by weight. 20

20. A method according to Claim 19 in which a composition is used containing from 0.01 to 10% of the active compound, by weight. 25

25 21. A method according to Claim 18, 19 or 20 in which the active compound is applied against insects or acarids. 25

22. Crops protected from damage by arthropods, fungi or bacteria, by being grown in areas in which immediately prior to and/or during the time of the growing a compound according to any of Claims 1 to 3 and 15 was applied alone or in admixture with a diluent or carrier. 30

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