

SPIROHETEROCYCLIC PYRROLIDINE DIONE DERIVATIVES USEFUL AS PESTICIDES

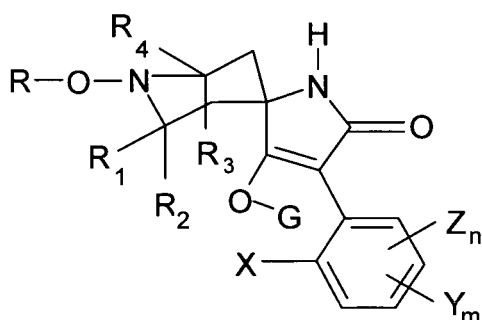
The present invention relates to new spiroheterocyclic pyrrolidine dione derivatives, to processes for preparing them, to pesticidal, in particular insecticidal, acaricidal,
 5 molluscicidal and nematocidal compositions comprising them and to methods of using them to combat and control pests such as insect, acarine, mollusc and nematode pests.

Spiroheterocyclic pyrrolidine dione derivatives are disclosed for example in US 6555567, US 6479489, US 6774133, EP 596298, WO 98/05638 and WO 99/48869.

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It has now surprisingly been found that certain new spiroheterocyclic pyrrolidine dione derivatives with nitrogen interrupting the spirocycle, and especially those ones being N-oxy substituted, have good insecticidal properties.

15 The present invention therefore provides compounds of formula I



(I) ,

wherein

X , Y and Z, independently of each other, are C₁₋₄alkyl, C₃₋₆cycloalkyl, C₁₋₄haloalkyl,

20 C₁₋₄alkoxy, halogen, phenyl or phenyl substituted by C₁₋₄alkyl or halogen, C₁₋₄haloalkyl, halogen or cyano,

m and n, independently of each other, are 0, 1, 2 or 3, where m+n is 0, 1, 2 or 3,

G is hydrogen, a metal, ammonium, sulfonium or latentating group,

R is hydrogen, C₁₋₆alkyl, C₁₋₆haloalkyl, C₁₋₆cyanoalkyl, benzyl, C₁₋₄alkoxy(C₁₋₄)alkyl,

25 C₁₋₄alkoxy(C₁₋₄)alkoxy(C₁₋₄)alkyl or a group selected from G, and

R₁, R₂, R₃ and R₄, independently of each other, are hydrogen or methyl, or an agrochemically acceptable salt or an N-oxide thereof.

In the compounds of the formula I, each alkyl moiety either alone or as part of a larger group is a straight or branched chain and is, for example, methyl, ethyl, n-propyl, n-butyl, iso-propyl, sec-butyl, iso-butyl, tert-butyl, n-pentyl, iso-pentyl and n-hexyl.

- 5 Alkoxy groups preferably have a preferred chain length of from 1 to 4 carbon atoms. Alkoxy is, for example, methoxy, ethoxy, propoxy, i-propoxy, n-butoxy, isobutoxy, sec-butoxy and tert-butoxy. Such groups can be part of a larger group such as alkoxyalkyl and alkoxyalkoxyalkyl. Alkoxyalkyl groups preferably have a chain length of 1 to 4 carbon atoms. Alkoxyalkyl is, for example, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, n-propoxymethyl, n-propoxyethyl or isopropoxymethyl.
- 10

Halogen is generally fluorine, chlorine, bromine or iodine. This also applies, correspondingly, to halogen in combination with other meanings, such as haloalkyl.

- 15 Haloalkyl groups preferably have a chain length of from 1 to 6 carbon atoms. Haloalkyl is, for example, fluoromethyl, difluoromethyl, trifluoromethyl, chloromethyl, dichloromethyl, trichloromethyl, 2,2,2-trifluoroethyl, 2-fluoroethyl, 2-chloroethyl, pentafluoroethyl, 1,1-difluoro-2,2,2-trichloroethyl, 2,2,3,3-tetrafluoroethyl and 2,2,2-trichloroethyl; preferably trichloromethyl, difluorochloromethyl, difluoromethyl, trifluoromethyl and dichlorofluoromethyl.
- 20

The cycloalkyl groups preferably have from 3 to 6 ring carbon atoms, for example cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl.

- 25 Phenyl, also as part of a substituent such as benzyl, may be substituted, preferably by alkyl groups. In this case, the substituents can be in ortho, meta and/or para position. The preferred substituent positions are the ortho and para positions to the ring attachment point.

- The latentating groups G are selected to allow its removal by one or a combination of biochemical, chemical or physical processes to afford compounds of formula I where G is hydrogen before, during or following application to the treated area or plants. Examples of these processes include enzymatic cleavage, chemical hydrolysis and photolysis.
- 30
- Compounds bearing such groups G may offer certain advantages, such as improved penetration of the cuticula of the plants treated, increased tolerance of crops, improved compatibility or stability in formulated mixtures containing other herbicides, herbicide
- 35
- safeners, plant growth regulators, fungicides or insecticides, or reduced leaching in soils.

The latentating group G is preferably selected from the groups C₁-C₈alkyl, C₂-C₈haloalkyl, phenylC₁-C₈alkyl (wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano or by nitro), heteroarylC₁-C₈alkyl (wherein the heteroaryl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano or by nitro), C₃-C₈alkenyl, C₃-C₈haloalkenyl, C₃-C₈alkynyl, C(X^a)-R^a, C(X^b)-X^c-R^b, C(X^d)-N(R^c)-R^d, -SO₂-R^e, -P(X^e)(R^f)-R^g or CH₂-X^f-R^h wherein X^a, X^b, X^c, X^d, X^e and X^f are independently of each other oxygen or sulfur;

R^a is H, C₁-C₁₈alkyl, C₂-C₁₈alkenyl, C₂-C₁₈alkynyl, C₁-C₁₀haloalkyl, C₁-C₁₀cyanoalkyl, C₁-C₁₀nitroalkyl, C₁-C₁₀aminoalkyl, C₁-C₅alkylaminoC₁-C₅alkyl, C₂-C₈dialkylaminoC₁-C₅alkyl, C₃-C₇cycloalkylC₁-C₅alkyl, C₁-C₅alkoxyC₁-C₅alkyl, C₃-C₅alkenyloxyC₁-C₅alkyl, C₃-C₅alkynylC₁-C₅oxyalkyl, C₁-C₅alkylthioC₁-C₅alkyl, C₁-C₅alkylsulfinylC₁-C₅alkyl, C₁-C₅alkylsulfonylC₁-C₅alkyl, C₂-C₈alkylideneaminoxyC₁-C₅alkyl, C₁-C₅alkylcarbonylC₁-C₅alkyl, C₁-C₅alkoxycarbonylC₁-C₅alkyl, aminocarbonylC₁-C₅alkyl, C₁-C₅alkylaminocarbonylC₁-C₅alkyl, C₂-C₈dialkylaminocarbonylC₁-C₅alkyl, C₁-C₅alkylcarbonylaminoC₁-C₅alkyl, *N*-C₁-C₅alkylcarbonyl-*N*-C₁-C₅alkylaminoC₁-C₅alkyl, C₃-C₆trialkylsilylC₁-C₅alkyl, phenylC₁-C₅alkyl (wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), heteroarylC₁-C₅alkyl, (wherein the heteroaryl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), C₂-C₅haloalkenyl, C₃-C₈cycloalkyl, phenyl or phenyl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, heteroaryl or heteroaryl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro,

R^b is C₁-C₁₈alkyl, C₃-C₁₈alkenyl, C₃-C₁₈alkynyl, C₂-C₁₀haloalkyl, C₁-C₁₀cyanoalkyl, C₁-C₁₀nitroalkyl, C₂-C₁₀aminoalkyl, C₁-C₅alkylaminoC₁-C₅alkyl, C₂-C₈dialkylaminoC₁-C₅alkyl, C₃-C₇cycloalkylC₁-C₅alkyl, C₁-C₅alkoxyC₁-C₅alkyl, C₃-C₅alkenyloxyC₁-C₅alkyl, C₃-C₅alkynyloxyC₁-C₅alkyl, C₁-C₅alkylthioC₁-C₅alkyl, C₁-C₅alkylsulfinylC₁-C₅alkyl, C₁-C₅alkylsulfonylC₁-C₅alkyl, C₂-C₈alkylideneaminoxyC₁-C₅alkyl, C₁-C₅alkylcarbonylC₁-C₅alkyl, C₁-C₅alkoxycarbonylC₁-C₅alkyl, aminocarbonylC₁-C₅alkyl, C₁-C₅alkylaminocarbonylC₁-C₅alkyl, C₂-C₈dialkylaminocarbonylC₁-C₅alkyl, C₁-C₅alkylcarbonylaminoC₁-C₅alkyl, *N*-C₁-C₅alkylcarbonyl-*N*-C₁-C₅alkylaminoC₁-C₅alkyl, C₃-C₆trialkylsilylC₁-C₅alkyl, phenylC₁-C₅alkyl (wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen,

cyano, or by nitro), heteroarylC₁-C₅alkyl, (wherein the heteroaryl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), C₃-C₅haloalkenyl, C₃-C₈cycloalkyl, phenyl or phenyl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, heteroaryl or heteroaryl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro,

R^c and R^d are each independently of each other hydrogen, C₁-C₁₀alkyl, C₃-C₁₀alkenyl, C₃-C₁₀alkynyl, C₂-C₁₀haloalkyl, C₁-C₁₀cyanoalkyl, C₁-C₁₀nitroalkyl, C₁-C₁₀aminoalkyl, C₁-C₅alkylaminoC₁-C₅alkyl, C₂-C₈dialkylaminoC₁-C₅alkyl, C₃-C₇cycloalkylC₁-C₅alkyl, C₁-C₅alkoxyC₁-C₅alkyl, C₃-C₅alkenyloxyC₁-C₅alkyl, C₃-C₅alkynyloxyC₁-C₅alkyl, C₁-C₅alkylthioC₁-C₅alkyl, C₁-C₅alkylsulfinylC₁-C₅alkyl, C₁-C₅alkylsulfonylC₁-C₅alkyl, C₂-C₈alkylideneaminoxyC₁-C₅alkyl, C₁-C₅alkylcarbonylC₁-C₅alkyl, C₁-C₅alkoxycarbonylC₁-C₅alkyl, aminocarbonylC₁-C₅alkyl, C₁-C₅alkylaminocarbonylC₁-C₅alkyl, C₂-C₈dialkylaminocarbonylC₁-C₅alkyl, C₁-C₅alkylcarbonylaminoC₁-C₅alkyl, *N*-C₁-C₅alkylcarbonyl-*N*-C₂-C₅alkylaminoalkyl, C₃-C₆trialkylsilylC₁-C₅alkyl, phenylC₁-C₅alkyl (wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), heteroarylC₁-C₅alkyl, (wherein the heteroaryl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), C₂-C₅haloalkenyl, C₃-C₈cycloalkyl, phenyl or phenyl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, heteroaryl or heteroaryl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, heteroaryl-amino or heteroaryl-amino substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, diheteroaryl-amino or diheteroaryl-amino substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, phenyl-amino or phenyl-amino substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or by nitro, diphenyl-amino or diphenyl-amino substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or by nitro or C₃-C₇cycloalkyl-amino, di-C₃-C₇cycloalkyl-amino or C₃-C₇cycloalkoxy or R^c and R^d may join together to form a 3-7 membered ring, optionally containing one heteroatom selected from O or S,

R^e is C₁-C₁₀alkyl, C₂-C₁₀alkenyl, C₂-C₁₀alkynyl, C₁-C₁₀haloalkyl, C₁-C₁₀cyanoalkyl, C₁-C₁₀nitroalkyl, C₁-C₁₀aminoalkyl, C₁-C₅alkylaminoC₁-C₅alkyl, C₂-C₈dialkylaminoC₁-C₅alkyl, C₃-C₇cycloalkylC₁-C₅alkyl, C₁-C₅alkoxyC₁-C₅alkyl, C₃-C₅alkenyloxyC₁-C₅alkyl, C₃-C₅alkynyloxyC₁-C₅alkyl, C₁-C₅alkylthioC₁-C₅alkyl, C₁-C₅alkylsulfinylC₁-C₅alkyl, C₁-

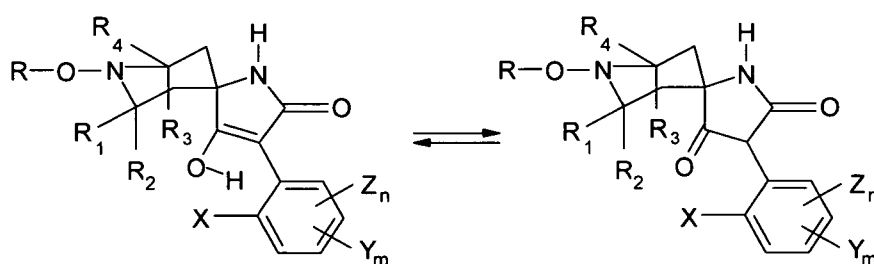
C₅alkylsulfonylC₁-C₅alkyl, C₂-C₈alkylideneaminoxyC₁-C₅alkyl, C₁-C₅alkylcarbonylC₁-C₅alkyl,
 C₁-C₅alkoxycarbonylC₁-C₅alkyl, aminocarbonylC₁-C₅alkyl, C₁-C₅alkylaminocarbonylC₁-
 C₅alkyl, C₂-C₈dialkylaminocarbonylC₁-C₅alkyl, C₁-C₅alkylcarbonylaminoC₁-C₅alkyl, *N*-C₁-
 C₅alkylcarbonyl-*N*-C₁-C₅alkylaminoC₁-C₅alkyl, C₃-C₆trialkylsilylC₁-C₅alkyl, phenylC₁-C₅alkyl
 5 (wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-
 C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen,
 cyano, or by nitro), heteroarylC₁-C₅alkyl (wherein the heteroaryl may optionally be
 substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-
 C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), C₂-C₅haloalkenyl, C₃-
 10 C₈cycloalkyl, phenyl or phenyl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-
 C₃haloalkoxy, halogen, cyano or nitro, heteroaryl or heteroaryl substituted by C₁-C₃ alkyl,
 C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or by nitro, heteroaryl amino
 or heteroaryl amino substituted by C₁-C₃ alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy,
 halogen, cyano or by nitro, diheteroaryl amino or diheteroaryl amino substituted by C₁-C₃
 15 alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, phenyl amino or
 phenyl amino substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy,
 halogen, cyano or nitro, diphenyl amino, or diphenyl amino substituted by C₁-C₃alkyl, C₁-
 C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, or C₃-
 C₇cycloalkyl amino, diC₃-C₇cycloalkyl amino or C₃-C₇cycloalkoxy, C₁-C₁₀alkoxy, C₁-
 20 C₁₀haloalkoxy, C₁-C₅alkyl amino or C₂-C₈dialkyl amino
 R^f and R^g are each independently of each other C₁-C₁₀alkyl, C₂-C₁₀alkenyl, C₂-
 C₁₀alkynyl, C₁-C₁₀alkoxy, C₁-C₁₀haloalkyl, C₁-C₁₀cyanoalkyl, C₁-C₁₀nitroalkyl, C₁-
 C₁₀aminoalkyl, C₁-C₅alkyl aminoC₁-C₅alkyl, C₂-C₈dialkyl aminoC₁-C₅alkyl, C₃-C₇cycloalkylC₁-
 C₅alkyl, C₁-C₅alkoxyC₁-C₅alkyl, C₃-C₅alkenyl oxyC₁-C₅alkyl, C₃-C₅alkynyl oxyC₁-C₅alkyl, C₁-
 25 C₅alkylthioC₁-C₅alkyl, C₁-C₅alkylsulfinylC₁-C₅alkyl, C₁-C₅alkylsulfonylC₁-C₅alkyl, C₂-
 C₈alkylideneaminoxyC₁-C₅alkyl, C₁-C₅alkylcarbonylC₁-C₅alkyl, C₁-C₅alkoxycarbonylC₁-
 C₅alkyl, aminocarbonylC₁-C₅alkyl, C₁-C₅alkylaminocarbonylC₁-C₅alkyl, C₂-
 C₈dialkylaminocarbonylC₁-C₅alkyl, C₁-C₅alkylcarbonylaminoC₁-C₅alkyl, *N*-C₁-
 C₅alkylcarbonyl-*N*-C₂-C₅alkylaminoalkyl, C₃-C₆trialkylsilylC₁-C₅alkyl, phenylC₁-C₅alkyl
 30 (wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-
 C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen,
 cyano, or by nitro), heteroarylC₁-C₅alkyl (wherein the heteroaryl may optionally be
 substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkylthio, C₁-
 C₃alkylsulfinyl, C₁-C₃alkylsulfonyl, halogen, cyano, or by nitro), C₂-C₅haloalkenyl, C₃-
 35 C₈cycloalkyl, phenyl or phenyl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-
 C₃haloalkoxy, halogen, cyano or nitro, heteroaryl or heteroaryl substituted by C₁-C₃ alkyl,

C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or by nitro, heteroaryl amino
 or heteroaryl amino substituted by C₁-C₃ alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy,
 halogen, cyano or by nitro, diheteroaryl amino or diheteroaryl amino substituted by C₁-C₃
 alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, phenyl amino or
 5 phenyl amino substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy,
 halogen, cyano or nitro, diphenyl amino, or diphenyl amino substituted by C₁-C₃alkyl, C₁-
 C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen, cyano or nitro, or C₃-
 C₇cycloalkyl amino, diC₃-C₇cycloalkyl amino or C₃-C₇cycloalkoxy, C₁-C₁₀haloalkoxy, C₁-
 C₅alkyl amino or C₂-C₈dialkyl amino, benzyloxy or phenoxy, wherein the benzyl and phenyl
 10 groups may in turn be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-
 C₃haloalkoxy, halogen, cyano or nitro, and
 R^h is C₁-C₁₀alkyl, C₃-C₁₀alkenyl, C₃-C₁₀alkynyl, C₁-C₁₀haloalkyl, C₁-C₁₀cyanoalkyl, C₁-
 C₁₀nitroalkyl, C₂-C₁₀aminoalkyl, C₁-C₅alkyl amino C₁-C₅alkyl, C₂-C₈dialkyl amino C₁-C₅alkyl, C₃-
 C₇cycloalkyl C₁-C₅alkyl, C₁-C₅alkoxy C₁-C₅alkyl, C₃-C₅alkenyl oxy C₁-C₅alkyl, C₃-
 15 C₅alkynyl oxy C₁-C₅alkyl, C₁-C₅alkyl thio C₁-C₅alkyl, C₁-C₅alkyl sulfinyl C₁-C₅alkyl, C₁-
 C₅alkyl sulfonyl C₁-C₅alkyl, C₂-C₈alkylidene amino oxy C₁-C₅alkyl, C₁-C₅alkyl carbonyl C₁-C₅alkyl,
 C₁-C₅alkoxy carbonyl C₁-C₅alkyl, aminocarbonyl C₁-C₅alkyl, C₁-C₅alkyl aminocarbonyl C₁-
 C₅alkyl, C₂-C₈dialkyl aminocarbonyl C₁-C₅alkyl, C₁-C₅alkyl carbonyl amino C₁-C₅alkyl, *N*-C₁-
 C₅alkyl carbonyl-*N*-C₁-C₅alkyl amino C₁-C₅alkyl, C₃-C₆trialkyl silyl C₁-C₅alkyl, phenyl C₁-C₅alkyl
 20 (wherein wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-
 C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkyl thio, C₁-C₃alkyl sulfinyl, C₁-C₃alkyl sulfonyl, halogen,
 cyano or by nitro), heteroaryl C₁-C₅alkyl (wherein the heteroaryl may optionally be
 substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkyl thio, C₁-
 C₃alkyl sulfinyl, C₁-C₃alkyl sulfonyl, halogen, cyano or by nitro), phenoxy C₁-C₅alkyl (wherein
 25 wherein the phenyl may optionally be substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy,
 C₁-C₃haloalkoxy, C₁-C₃alkyl thio, C₁-C₃alkyl sulfinyl, C₁-C₃alkyl sulfonyl, halogen, cyano or by
 nitro), heteroaryl oxy C₁-C₅alkyl (wherein the heteroaryl may optionally be substituted by C₁-
 C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, C₁-C₃alkyl thio, C₁-C₃alkyl sulfinyl, C₁-
 C₃alkyl sulfonyl, halogen, cyano or by nitro), C₃-C₅haloalkenyl, C₃-C₈cycloalkyl, phenyl or
 30 phenyl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy, C₁-C₃haloalkoxy, halogen or
 by nitro, or heteroaryl, or heteroaryl substituted by C₁-C₃alkyl, C₁-C₃haloalkyl, C₁-C₃alkoxy,
 C₁-C₃haloalkoxy, halogen, cyano or by nitro.

In particular, the latentating group G is a group -C(X^a)-R^a or -C(X^b)-X^c-R^b, and the
 35 meanings of X^a, R^a, X^b, X^c and R^b are as defined above.

It is preferred that G is hydrogen, an alkali metal or alkaline earth metal, or an ammonium or sulfonium group, where hydrogen is especially preferred.

Depending on the nature of the substituents, compounds of formula I may exist in different isomeric forms. When G is hydrogen, for example, compounds of formula I may exist in different tautomeric forms:



This invention covers all such isomers and tautomers and mixtures thereof in all proportions. Also, when substituents contain double bonds, *cis*- and *trans*-isomers can exist. These isomers, too, are within the scope of the claimed compounds of the formula I.

The invention relates also to the agriculturally acceptable salts which the compounds of formula I are able to form with transition metal, alkali metal and alkaline earth metal bases, amines, quaternary ammonium bases or tertiary sulfonium bases.

Among the transition metal, alkali metal and alkaline earth metal salt formers, special mention should be made of the hydroxides of copper, iron, lithium, sodium, potassium, magnesium and calcium, and preferably the hydroxides, bicarbonates and carbonates of sodium and potassium.

Examples of amines suitable for ammonium salt formation include ammonia as well as primary, secondary and tertiary C₁-C₁₈alkylamines, C₁-C₄hydroxyalkylamines and C₂-C₄alkoxyalkyl-amines, for example methylamine, ethylamine, *n*-propylamine, *i*-propylamine, the four butylamine isomers, *n*-amylamine, *i*-amylamine, hexylamine, heptylamine, octylamine, nonylamine, decylamine, pentadecylamine, hexadecylamine, heptadecylamine, octadecylamine, methylethylamine, methylisopropylamine, methylhexylamine, methylnonylamine, methylpentadecylamine, methyloctadecylamine, ethylbutylamine, ethylheptylamine, ethyloctylamine, hexylheptylamine, hexyloctylamine, dimethylamine, diethylamine, di-*n*-propylamine, di-*i*-propylamine, di-*n*-butylamine, di-*n*-

amylamine, di-*i*-amylamine, dihexylamine, diheptylamine, dioctylamine, ethanolamine, *n*-propanolamine, *i*-propanolamine, *N,N*-diethanolamine, *N*-ethylpropanolamine, *N*-butylethanolamine, allylamine, *n*-but-2-enylamine, *n*-pent-2-enylamine, 2,3-dimethylbut-2-enylamine, dibut-2-enylamine, *n*-hex-2-enylamine, propylenediamine, trimethylamine, triethylamine, tri-*n*-propylamine, tri-*i*-opropylamine, tri-*n*-butylamine, tri-*i*-butylamine, tri-*sec*-butylamine, tri-*n*-amylamine, methoxyethylamine and ethoxyethylamine; heterocyclic amines, for example pyridine, quinoline, isoquinoline, morpholine, piperidine, pyrrolidine, indoline, quinuclidine and azepine; primary arylamines, for example anilines, methoxyanilines, ethoxyanilines, *o*-, *m*- and *p*-toluidines, phenylenediamines, benzidines, naphthylamines and *o*-, *m*- and *p*-chloroanilines; but especially triethylamine, *i*-propylamine and di-*i*-propylamine.

Preferred quaternary ammonium bases suitable for salt formation correspond, for example, to the formula $[N(R_a R_b R_c R_d)]OH$, wherein R_a , R_b , R_c and R_d are each independently of the others hydrogen or C_1 - C_4 alkyl. Further suitable tetraalkylammonium bases with other anions can be obtained, for example, by anion exchange reactions.

Preferred tertiary sulfonium bases suitable for salt formation correspond, for example, to the formula $[SR_e R_f R_g]OH$, wherein R_e , R_f and R_g are each independently of the others C_1 - C_4 alkyl. Trimethylsulfonium hydroxide is especially preferred. Suitable sulfonium bases may be obtained from the reaction of thioethers, in particular dialkylsulfides, with alkylhalides, followed by conversion to a suitable base, for example a hydroxide, by anion exchange reactions.

It should be understood that in those compounds of formula I, where G is a metal, ammonium or sulfonium as mentioned above and as such represents a cation, the corresponding negative charge is largely delocalised across the $O-C=C-C=O$ unit.

The compounds of formula I according to the invention also include hydrates which may be formed during the salt formation.

In the compounds of the formula I, the substituent R is preferably C_1 - C_4 alkyl, in particular methyl or ethyl.

Preferably, X, Y and Z denote C₁-C₄alkyl, C₃-C₆cycloalkyl, C₁-C₄alkoxy or halogen, in particular methyl, ethyl, cyclopropyl, methoxy, fluoro or chloro, when m+n is 1-3, in particular, when m+n is 1-2.

Alternatively, Y and Z, independently of each other, denote C₁-C₄alkyl, C₃-C₆cycloalkyl, C₁-C₄alkoxy, halogen, phenyl or phenyl substituted by C₁₋₄alkyl or halogen, in particular methyl, ethyl, cyclopropyl, methoxy, fluoro, chloro, phenyl or phenyl substituted with halogen, in particular fluoro or chloro, in particular in 4-position, when m+n is 1-3, in particular, when m+n is 1-2.

10 In preferred group of compounds of the formula I, R₁ to R₄ are hydrogen.

In another preferred group of compounds of the formula (I), R is methyl or ethyl, X is methyl, ethyl, cyclopropyl, methoxy, fluoro or chloro, Y and Z, independently of each other, are methyl, ethyl, cyclopropyl, methoxy, fluoro, chloro, phenyl or phenyl substituted by halogen or C₁-C₂alkyl, G is hydrogen, and R₁, R₂, R₃ and R₄ are hydrogen.

The invention covers also salts of the compounds of the formula I with amines, alkali metal and alkaline earth metal bases or quaternary ammonium bases.

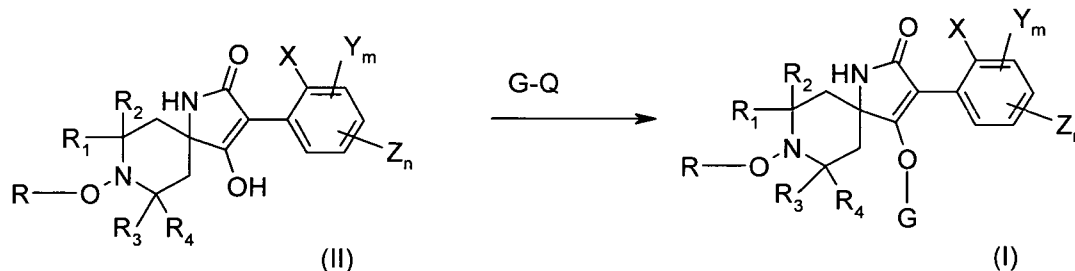
Among the alkali metal and alkaline earth metal hydroxides as salt formers, special mention should be made of the hydroxides of lithium, sodium, potassium, magnesium and calcium, but especially the hydroxides of sodium and potassium. The compounds of formula I according to the invention also include hydrates which may be formed during the salt formation.

Examples of amines suitable for ammonium salt formation include ammonia as well as primary, secondary and tertiary C₁-C₁₈alkylamines, C₁-C₄hydroxyalkylamines and C₂-C₄alkoxyalkylamines, for example methylamine, ethylamine, n-propylamine, isopropylamine, the four butylamine isomers, n-amylamine, isoamylamine, hexylamine, heptylamine, octylamine, nonylamine, decylamine, pentadecylamine, hexadecylamine, heptadecylamine, octadecylamine, methylethylamine, methylisopropylamine, methylhexylamine, methylnonylamine, methylpentadecylamine, methyloctadecylamine, ethylbutylamine, ethylheptylamine, ethyloctylamine, hexylheptylamine, hexyloctylamine, dimethylamine, diethylamine, di-n-propylamine, diisopropylamine, di-n-butylamine, di-n-amylamine, diisoamylamine, dihexylamine, diheptylamine, dioctylamine, ethanolamine, n-propanolamine, isopropanolamine, N,N-diethanolamine, N-ethylpropanolamine, N-butylethanolamine, allylamine, n-but-2-enylamine, n-pent-2-enylamine, 2,3-dimethylbut-2-enylamine, dibut-2-enylamine, n-hex-2-enylamine, propylenediamine, trimethylamine,

triethylamine, tri-n-propylamine, triisopropylamine, tri-n-butylamine, triisobutylamine, tri-sec-butylamine, tri-n-amylamine, methoxyethylamine and ethoxyethylamine; heterocyclic amines, for example pyridine, quinoline, isoquinoline, morpholine, piperidine, pyrrolidine, indoline, quinuclidine and azepine; primary arylamines, for example anilines, methoxyanilines, ethoxyanilines, o-, m- and p-toluidines, phenylenediamines, benzidines, naphthylamines and o-, m- and p-chloroanilines; but especially triethylamine, isopropylamine and diisopropylamine.

Preferred quaternary ammonium bases suitable for salt formation correspond, for example, to the formula $[N(R_a R_b R_c R_d)]OH$ wherein R_a , R_b , R_c and R_d are each independently of the others C_1 - C_4 alkyl. Further suitable tetraalkylammonium bases with other anions can be obtained, for example, by anion exchange reactions.

The compounds of the invention may be made by a variety of methods. For example, the compounds of formula I, wherein the substituents have the meanings assigned to them above, can be prepared by means of processes known *per se*, e.g. by treating compounds of formula II with an alkylating, acylating, phosphorylating or sulfonylating agent G-Q in the presence of at least one equivalent of a base, where G is the alkyl, acyl, phosphoryl or sulfonyl group to be incorporated and Q is a nucleofuge:



Compounds of formula I, wherein G is a latentating group of the formula $-C(X^a)-R^a$, $C(X^b)-X^c-R^b$ or $-C(X^d)-NR^cR^d$ may be prepared by known procedures as described, for example, in US 6774133, US 6555567 and US 6479489. Typically, compounds of formula II are treated with an acylating agent such as an acid halide (especially acid chloride), acid anhydride, haloformate (especially chloroformate), halothioformate (especially chlorothioformate), isocyanate, isothiocyanate, carbamoyl halide (especially carbamoyl chloride) or thiocarbamoyl chloride (especially thiocarbamoyl chloride) in the presence of at least one equivalent of a suitable base, optionally in the presence of a suitable solvent. The base may be inorganic such as an alkali metal carbonate or hydroxide or a metal hydride, or an organic base such as a tertiary amine or metal alkoxide. Examples of suitable inorganic

bases include sodium carbonate, sodium or potassium hydroxide, sodium hydride, and suitable organic bases include trialkylamines such as trimethylamine and triethylamine, pyridines or other amine bases such as 1,4-diazobicyclo[2.2.2]octane and 1,8-diazabicyclo[5.4.0]undec-7-ene. Preferred bases include triethylamine and pyridine.

5 Suitable solvents for this reaction are selected to be compatible with the reagents and include ethers such as tetrahydrofuran and 1,2-dimethoxyethane and halogenated solvents such as dichloromethane and chloroform. Certain bases, such as pyridine and triethylamine, may be employed successfully as both base and solvent. For cases, where the acylating agent is a carboxylic acid, acylation is preferably effected in the presence of a
10 coupling agent such as 2-chloro-1-methylpyridinium iodide, *N,N'*-dicyclohexycarbodiimide, 1-(3-dimethylaminopropyl)-3-ethylcarbodiimide and *N,N'*-carbodiimidazole, and a base such as triethylamine or pyridine in a suitable solvent such as tetrahydrofuran, dichloromethane and acetonitrile. Suitable procedures are described, for example, in US 6774133, US 6555567 and US 6479489.

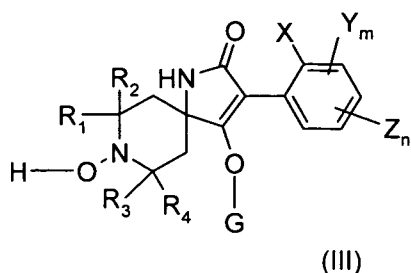
15 Compounds of formula I, wherein G is a latentating group of the formula $C(X^b)-X^c-R^b$ or $C(X^d)-NR^cR^d$, may be also be prepared by treating compounds of formula II with phosgene or a phosgene equivalent, optionally in the presence of a solvent such as toluene or ethyl acetate, and a base and reacting the resultant chloroformate, or equivalent, with an alcohol,
20 thiol or amine under known conditions, as described, for example, in US 6774133, US 6555567 and US 6479489.

Compounds of formula I, wherein G is a latentating group of the formula $-P(X^e)R^fR^g$ may be prepared from compounds of formula II using procedures described, for example, in
25 US 6774133, US 6555567 and US 6479489.

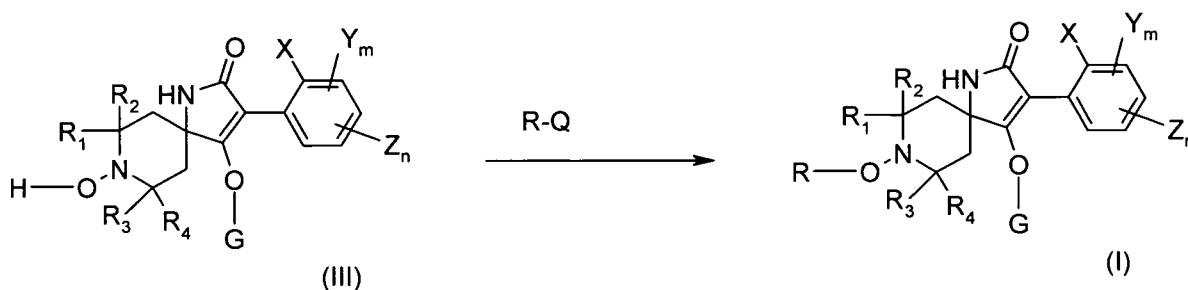
Compounds of formula I, wherein G is a latentating group of the formula $-SO_2R^e$, may be prepared by reaction of compounds of formula II with an alkyl or aryl sulfonyl halide, preferably in the presence of at least one equivalent of base.

30 Compounds of formula I, wherein G is C_1-C_6 alkyl, C_2-C_6 alkenyl, C_3-C_6 alkynyl or a latentating group of the formula $CH_2-X^f-R^h$, may be prepared by treatment of a compound of formula II with a compound of formula G-Y wherein Y is a halogen (especially bromine or iodine), sulfonate (especially mesylate or tosylate) or a sulfate preferably in the presence of
35 a base, under known conditions.

Compounds of formula III

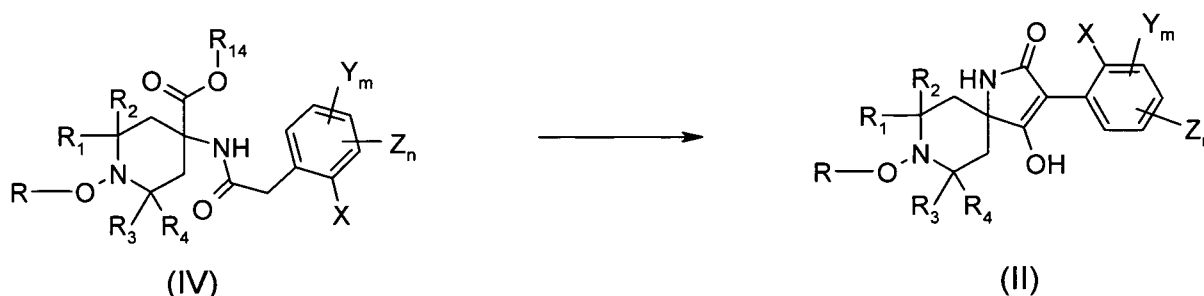


- 5 can be obtained by catalytic hydrogenation of compounds of formula I, in which R is represented by a benzyl group.

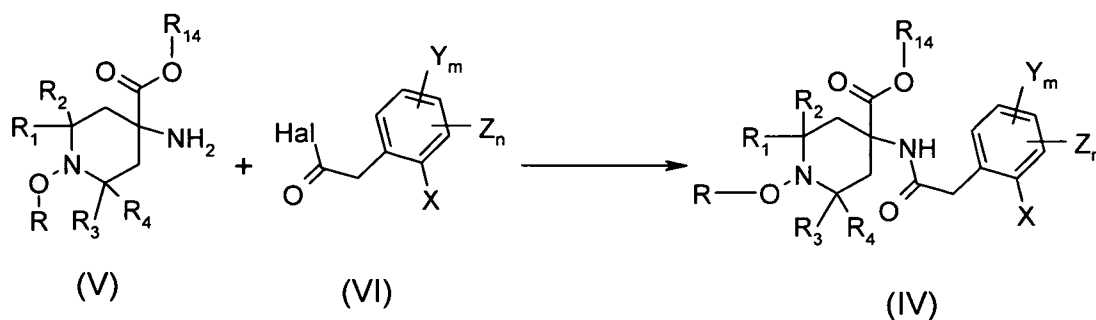


- 10 Compounds of formula I, in which R represents $-\text{C}(\text{X}^a)-\text{R}^a$, $\text{C}(\text{X}^b)-\text{X}^c-\text{R}^b$ or $-\text{C}(\text{X}^d)-\text{NR}^c\text{R}^d$, SO_2R^e , $\text{P}(\text{X}^e)\text{R}^f\text{R}^g$ or $\text{CH}_2-\text{X}^f-\text{R}^h$, can be obtained by treating compounds of formula III with an alkylating, acylating, phosphorylating or sulfonylating agent R-Q, wherein Q represents a nucleofuge, in the presence of at least one equivalent of a base.

- 15 Suitable conditions are the same as described above for the conversion of compounds of formula II to compounds of formula I.



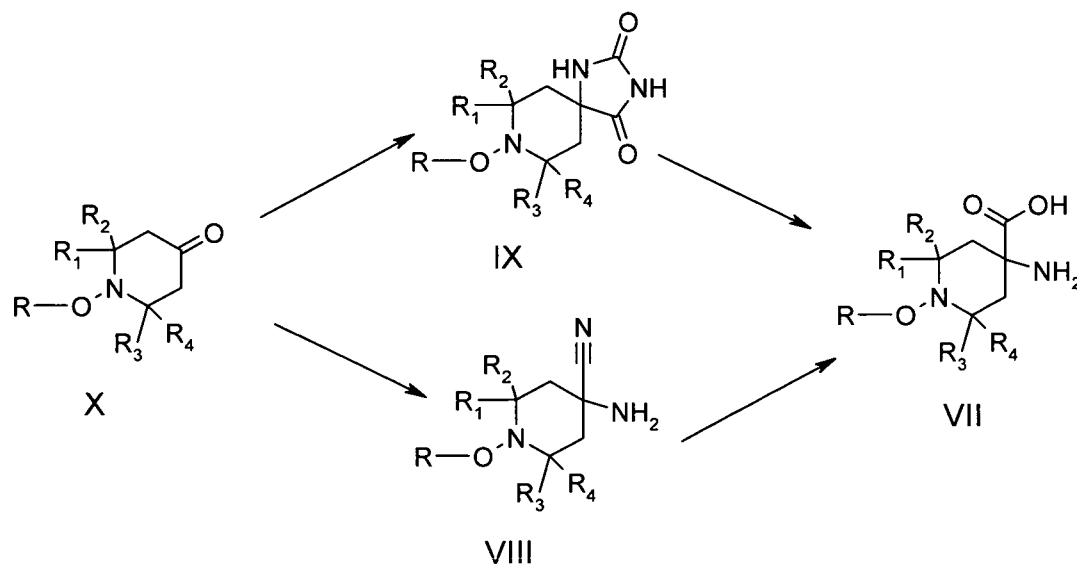
- 20 Compounds of formula II may be prepared via the cyclisation of compounds of formula IV, wherein R_{14} is C_{1-4} alkyl, preferably in the presence of base, and optionally in the presence of a suitable solvent, by methods described, for example, in US 6774133, US 6555567 and US 6479489.



Compounds of formula IV may be prepared by reacting amino acid derivatives of formula V with phenylacetyl halides of formula VI, preferably in the presence of base in a suitable solvent by known methods described, for example, in US 6774133, US 6555567 and US 6479489.

Phenylacetyl halides of formula VI, wherein Hal is Cl or Br, are known compounds or can be prepared by known methods as described, for example, in US 6774133, US 6555567 and US 6479489.

Amino acid esters of formula V, wherein R₁₄ is C₁-C₄alkyl, can be prepared by known methods from amino acids of formula VII. These compounds can be isolated as free amines or amine salts.

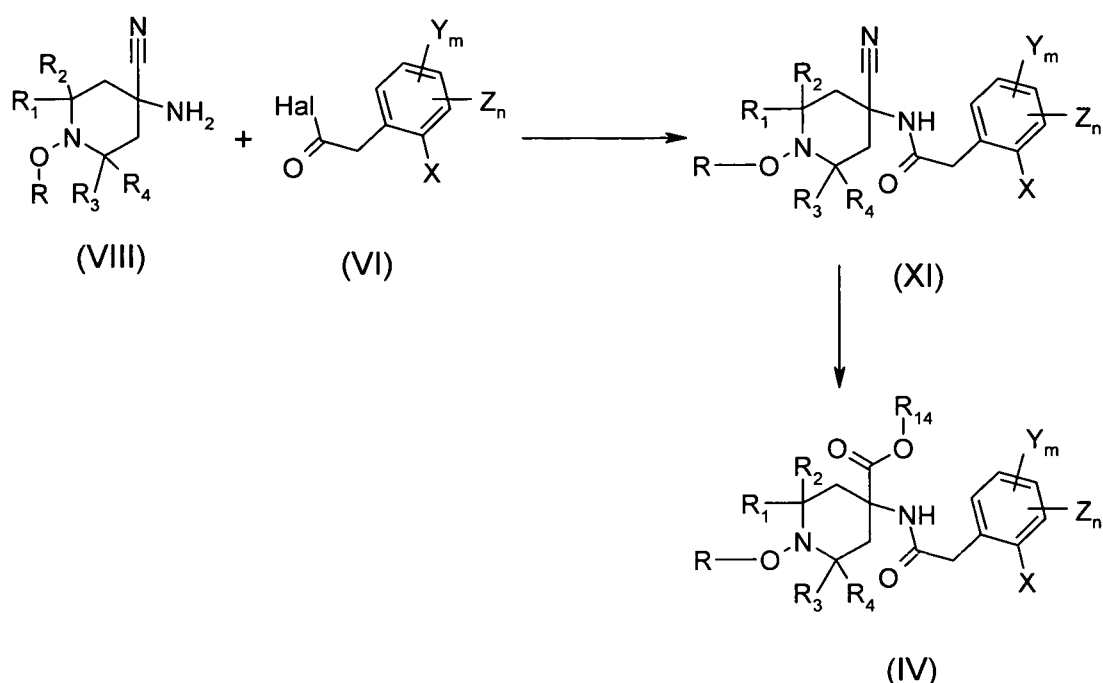


Amino acids of formula VII can be prepared from ketons of formula X by means of Strecker Synthesis via amino nitriles of formula VIII.

Alternatively, amino acids of formula VII can be prepared from ketones of formula X by means of Bucherer Berge reaction via hydantoin of formula IX.

Compounds of formula X, where R_1 , R_2 , R_3 and R_4 are hydrogen and R is C_{1-4} alkyl, C_{1-4} haloalkyl, C_3-C_6 alkenyl, C_3-C_6 alkynyl or a benzyl group, are known or can be obtained, for example, according to Journal of Organic Chemistry (1961), 26, 1867-74).

Compounds of formula X, where R_1 , R_2 , R_3 and R_4 are CH_3 and R is a C_{1-4} alkyl or C_3-C_6 alkenyl, are known or can be obtained in analogy to procedures described, for example, in WO9854174.



Alternatively, compounds of formula IV may be prepared by subjecting derivatives of formula XI to alcoholysis with $R_{14}OH$, preferably in acidic media by known methods described, for example, in US 6774133, US 6555567 and US 6479489.

Compounds of formula XI may be themselves prepared by reacting amino nitriles of formula VIII with phenylacetyl halides of formula VI, preferably in the presence of base in a suitable solvent by known methods described, for example, in US 6774133, US 6555567 and US 6479489.

The compounds of the formulae IV, V, VII, VIII, IX and XI, and salts thereof in the case of compounds V, VII and VIII, are novel and have been specifically designed for the synthesis of the compounds of the formula I. The remaining starting compounds and intermediates of

the reaction schemes are known or can be prepared according to methods known to a person skilled in the art.

The reactants can be reacted in the presence of a base. Examples of suitable bases are
5 alkali metal or alkaline earth metal hydroxides, alkali metal or alkaline earth metal hydrides, alkali metal or alkaline earth metal amides, alkali metal or alkaline earth metal alkoxides, alkali metal or alkaline earth metal acetates, alkali metal or alkaline earth metal carbonates, alkali metal or alkaline earth metal dialkylamides or alkali metal or alkaline earth metal alkylsilylamides, alkylamines, alkylenediamines, free or N-alkylated saturated or
10 unsaturated cycloalkylamines, basic heterocycles, ammonium hydroxides and carbocyclic amines. Examples which may be mentioned are sodium hydroxide, sodium hydride, sodium amide, sodium methoxide, sodium acetate, sodium carbonate, potassium tert-butoxide, potassium hydroxide, potassium carbonate, potassium hydride, lithium diisopropylamide, potassium bis(trimethylsilyl)amide, calcium hydride, triethylamine, diisopropylethylamine,
15 triethylenediamine, cyclohexylamine, N-cyclohexyl-N,N-dimethylamine, N,N-diethylaniline, pyridine, 4-(N,N-dimethylamino)pyridine, quinuclidine, N-methylmorpholine, benzyltrimethylammonium hydroxide and 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU).

The reactants can be reacted with each other as such, i.e. without adding a solvent or diluent.
20 In most cases, however, it is advantageous to add an inert solvent or diluent or a mixture of these. If the reaction is carried out in the presence of a base, bases which are employed in excess, such as triethylamine, pyridine, N-methylmorpholine or N,N-diethylaniline, may also act as solvents or diluents.

25 The reaction is advantageously carried out in a temperature range from approximately -80°C to approximately +140°C, preferably from approximately -30°C to approximately +100°C, in many cases in the range between ambient temperature and approximately +80°C.

30 A compound I can be converted in a manner known per se into another compound I by replacing one or more substituents of the starting compound I in the customary manner by (an)other substituent(s) according to the invention.

Depending on the choice of the reaction conditions and starting materials which are suitable
35 in each case, it is possible, for example, in one reaction step only to replace one substituent

by another substituent according to the invention, or a plurality of substituents can be replaced by other substituents according to the invention in the same reaction step.

Salts of compounds I can be prepared in a manner known per se. Thus, for example, acid
5 addition salts of compounds I are obtained by treatment with a suitable acid or a suitable ion exchanger reagent and salts with bases are obtained by treatment with a suitable base or with a suitable ion exchanger reagent.

Salts of compounds I can be converted in the customary manner into the free compounds I,
10 acid addition salts, for example, by treatment with a suitable basic compound or with a suitable ion exchanger reagent and salts with bases, for example, by treatment with a suitable acid or with a suitable ion exchanger reagent.

Salts of compounds I can be converted in a manner known per se into other salts of
15 compounds I, acid addition salts, for example, into other acid addition salts, for example by treatment of a salt of inorganic acid such as hydrochloride with a suitable metal salt such as a sodium, barium or silver salt, of an acid, for example with silver acetate, in a suitable solvent in which an inorganic salt which forms, for example silver chloride, is insoluble and thus precipitates from the reaction mixture.

20 Depending on the procedure or the reaction conditions, the compounds I, which have salt-forming properties can be obtained in free form or in the form of salts.

The compounds I and, where appropriate, the tautomers thereof, in each case in free form
25 or in salt form, can be present in the form of one of the isomers which are possible or as a mixture of these, for example in the form of pure isomers, such as antipodes and/or diastereomers, or as isomer mixtures, such as enantiomer mixtures, for example racemates, diastereomer mixtures or racemate mixtures, depending on the number, absolute and relative configuration of asymmetric carbon atoms which occur in the molecule
30 and/or depending on the configuration of non-aromatic double bonds which occur in the molecule; the invention relates to the pure isomers and also to all isomer mixtures which are possible and is to be understood in each case in this sense hereinabove and hereinbelow, even when stereochemical details are not mentioned specifically in each case.

35 Diastereomer mixtures or racemate mixtures of compounds I, in free form or in salt form, which can be obtained depending on which starting materials and procedures have been

chosen can be separated in a known manner into the pure diastereomers or racemates on the basis of the physicochemical differences of the components, for example by fractional crystallization, distillation and/or chromatography.

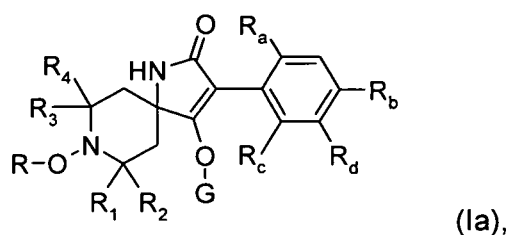
5 Enantiomer mixtures, such as racemates, which can be obtained in a similar manner can be resolved into the optical antipodes by known methods, for example by recrystallization from an optically active solvent, by chromatography on chiral adsorbents, for example high-performance liquid chromatography (HPLC) on acetyl cellulose, with the aid of suitable microorganisms, by cleavage with specific, immobilized enzymes, via the formation of
10 inclusion compounds, for example using chiral crown ethers, where only one enantiomer is complexed, or by conversion into diastereomeric salts, for example by reacting a basic end-product racemate with an optically active acid, such as a carboxylic acid, for example camphor, tartaric or malic acid, or sulfonic acid, for example camphorsulfonic acid, and separating the diastereomer mixture which can be obtained in this manner, for example by
15 fractional crystallization based on their differing solubilities, to give the diastereomers, from which the desired enantiomer can be set free by the action of suitable agents, for example basic agents.

Pure diastereomers or enantiomers can be obtained according to the invention not only by
20 separating suitable isomer mixtures, but also by generally known methods of diastereoselective or enantioselective synthesis, for example by carrying out the process according to the invention with starting materials of a suitable stereochemistry.

It is advantageous to isolate or synthesize in each case the biologically more effective isomer, for example enantiomer or diastereomer, or isomer mixture, for example enantiomer
25 mixture or diastereomer mixture, if the individual components have a different biological activity.

The compounds I and, where appropriate, the tautomers thereof, in each case in free form
30 or in salt form, can, if appropriate, also be obtained in the form of hydrates and/or include other solvents, for example those which may have been used for the crystallization of compounds which are present in solid form.

The compounds in Tables 1 to 22 below illustrate the compounds of the invention.

Table 1: This table discloses the 105 compounds T1.001 to T1.105 of the formula Ia:

- 5 wherein R is CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined below:

No.	R _a	R _b	R _c	R _d
T1.001	Br	H	H	H
T1.002	Cl	H	H	H
T1.003	CH ₃	H	H	H
T1.004	CH ₂ CH ₃	H	H	H
T1.005	OCH ₃	H	H	H
T1.006	Br	Cl	H	H
T1.007	Cl	Br	H	H
T1.008	Cl	Cl	H	H
T1.009	Cl	CH ₃	H	H
T1.010	CH ₃	Cl	H	H
T1.011	CH ₃	CH ₃	H	H
T1.012	Cl	H	Cl	H
T1.013	Cl	H	CH ₃	H
T1.014	Cl	H	CH ₂ CH ₃	H
T1.015	Cl	H	OCH ₃	H
T1.016	CH ₃	H	CH ₃	H
T1.017	CH ₃	H	CH ₂ CH ₃	H
T1.018	CH ₃	H	OCH ₃	H
T1.019	CH ₂ CH ₃	H	CH ₂ CH ₃	H
T1.020	CH ₂ CH ₃	H	OCH ₃	H
T1.021	OCH ₃	H	OCH ₃	H
T1.022	Br	H	H	Cl
T1.023	Br	H	H	CH ₃
T1.024	Br	H	H	4-Cl-C ₆ H ₄
T1.025	Cl	H	H	Cl
T1.026	Cl	H	H	CH ₃

No.	R _a	R _b	R _c	R _d
T1.027	Cl	H	H	4-Cl-C ₆ H ₄
T1.028	CH ₃	H	H	Br
T1.029	CH ₃	H	H	Cl
T1.030	CH ₃	H	H	CH ₃
T1.031	CH ₃	H	H	C ₆ H ₅
T1.032	CH ₃	H	H	4-Cl-C ₆ H ₄
T1.033	CH ₂ CH ₃	H	H	CH ₃
T1.034	CH ₂ CH ₃	H	H	4-Cl-C ₆ H ₄
T1.035	OCH ₃	H	H	CH ₃
T1.036	OCH ₃	H	H	4-Cl-C ₆ H ₄
T1.037	Cl	H	Cl	Br
T1.038	CH ₃	H	CH ₃	Br
T1.039	CH ₃	H	CH ₃	Cl
T1.040	CH ₃	H	CH ₃	4-Cl-C ₆ H ₄
T1.041	Br	Cl	H	CH ₃
T1.042	Br	CH ₃	H	CH ₃
T1.043	Cl	Cl	H	Cl
T1.044	Cl	Br	H	CH ₃
T1.045	Cl	Cl	H	CH ₃
T1.046	Cl	CH ₃	H	Cl
T1.047	Cl	CH ₃	H	CH ₃
T1.048	CH ₃	Br	H	CH ₃
T1.049	CH ₃	Cl	H	CH ₃
T1.050	CH ₃	CH ₃	H	CH ₃
T1.051	CH ₃	CH ₃	H	4-Cl-C ₆ H ₄
T1.052	Br	Br	CH ₃	H
T1.053	Br	Cl	CH ₃	H
T1.054	Br	CH ₃	Br	H
T1.055	Br	CH ₃	Cl	H
T1.056	Cl	Br	CH ₃	H
T1.057	Cl	Cl	Cl	H
T1.058	Cl	Cl	CH ₃	H
T1.059	Cl	CH ₃	Cl	H
T1.060	Cl	CH ₃	CH ₂ CH ₃	H
T1.061	Cl	CH ₃	OCH ₃	H
T1.062	Cl	4-Cl-C ₆ H ₄	Cl	H
T1.063	Cl	4-Cl-C ₆ H ₄	CH ₃	H
T1.064	Cl	4-Cl-C ₆ H ₄	CH ₂ CH ₃	H

No.	R _a	R _b	R _c	R _d
T1.065	Cl	4-Cl-C ₆ H ₄	OCH ₃	H
T1.066	CH ₃	Br	CH ₃	H
T1.067	CH ₃	Cl	CH ₃	H
T1.068	CH ₃	CH ₃	Br	H
T1.069	CH ₃	CH ₃	Cl	H
T1.070	CH ₃	CH ₃	CH ₃	H
T1.071	CH ₃	CH ₃	CH ₂ CH ₃	H
T1.072	CH ₃	CH ₃	OCH ₃	H
T1.073	CH ₃	4-Cl-C ₆ H ₄	CH ₃	H
T1.074	CH ₃	4-Cl-C ₆ H ₄	CH ₂ CH ₃	H
T1.075	CH ₃	4-Cl-C ₆ H ₄	OCH ₃	H
T1.076	CH ₂ CH ₃	Br	Br	H
T1.077	CH ₂ CH ₃	Br	Cl	H
T1.078	CH ₂ CH ₃	Br	CH ₃	H
T1.079	CH ₂ CH ₃	Br	CH ₂ CH ₃	H
T1.080	CH ₂ CH ₃	Br	OCH ₃	H
T1.081	CH ₂ CH ₃	Cl	Br	H
T1.082	CH ₂ CH ₃	Cl	Cl	H
T1.083	CH ₂ CH ₃	Cl	CH ₃	H
T1.084	CH ₂ CH ₃	Cl	CH ₂ CH ₃	H
T1.085	CH ₂ CH ₃	Cl	OCH ₃	H
T1.086	CH ₂ CH ₃	CH ₃	Br	H
T1.087	CH ₂ CH ₃	CH ₃	Cl	H
T1.088	CH ₂ CH ₃	CH ₃	CH ₂ CH ₃	H
T1.089	CH ₂ CH ₃	CH ₃	OCH ₃	H
T1.090	CH ₂ CH ₃	CH ₂ CH ₃	CH ₃	H
T1.091	CH ₂ CH ₃	CH ₂ CH ₃	CH ₂ CH ₃	H
T1.092	CH ₂ CH ₃	4-Cl-C ₆ H ₄	Br	H
T1.093	CH ₂ CH ₃	4-Cl-C ₆ H ₄	CH ₂ CH ₃	H
T1.094	CH ₂ CH ₃	4-Cl-C ₆ H ₄	OCH ₃	H
T1.095	OCH ₃	Br	CH ₃	H
T1.096	OCH ₃	Cl	CH ₃	H
T1.097	OCH ₃	CH ₃	Br	H
T1.098	OCH ₃	CH ₃	Cl	H
T1.099	OCH ₃	CH ₃	OCH ₃	H
T1.100	OCH ₃	4-Cl-C ₆ H ₄	OCH ₃	H
T1.101	CH ₃	CH ₃	CH ₃	F
T1.102	CH ₃	CH ₃	CH ₃	Cl

No.	R _a	R _b	R _c	R _d
T1.103	CH ₃	CH ₃	CH ₃	Br
T1.104	CH ₃	CH ₃	CH ₃	CH ₃
T1.105	CH ₃	CH ₃	CH ₃	4-Cl-C ₆ H ₄

Table 2: This table discloses the 105 compounds T2.001 to T2.105 of the formula Ia, wherein R is CH₂CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

5

Table 3: This table discloses the 105 compounds T3.001 to T3.105 of the formula Ia, wherein R is n-C₃H₇, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

10 Table 4: This table discloses the 105 compounds T4.001 to T4.105 of the formula Ia, wherein R is i-C₃H₇, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

15 Table 5: This table discloses the 105 compounds T5.001 to T5.105 of the formula Ia, wherein R is allyl, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

20 Table 6: This table discloses the 105 compounds T6.001 to T6.105 of the formula Ia, wherein R is benzyl, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

25 Table 7: This table discloses the 105 compounds T7.001 to T7.105 of the formula Ia, wherein R is C(=O)-CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

Table 8: This table discloses the 105 compounds T8.001 to T8.105 of the formula Ia, wherein R is C(=O)-CH₂CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

30 Table 9: This table discloses the 105 compounds T9.001 to T9.105 of the formula Ia, wherein R is C(=O)-n-C₃H₇, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

Table 10: This table discloses the 105 compounds T10.001 to T10.105 of the formula Ia, wherein R is C(=O)O-CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

5

Table 11: This table discloses the 105 compounds T11.001 to T11.105 of the formula Ia, wherein R is C(=O)O-CH₂CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

10 Table 12: This table discloses the 105 compounds T12.001 to T12.105 of the formula Ia, wherein R is C(=O)O-n-C₃H₇, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

15 Table 13: This table discloses the 105 compounds T13.001 to T13.105 of the formula Ia, wherein R is C(=O)NH-CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

20 Table 14: This table discloses the 105 compounds T14.001 to T14.105 of the formula Ia, wherein R is C(=O)NH-CH₂CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

25 Table 15: This table discloses the 105 compounds T15.001 to T15.105 of the formula Ia, wherein R is C(=O)NH-n-C₃H₇, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

Table 16: This table discloses the 105 compounds T16.001 to T16.105 of the formula Ia, wherein R is hydrogen, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

30 Table 17: This table discloses the 105 compounds T17.001 to T17.105 of the formula Ia, wherein R is CH₂-O-CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

35 Table 18: This table discloses the 105 compounds T18.001 to T18.105 of the formula Ia, wherein R is CH₂-O-C₂H₅, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

Table 19 This table discloses the 105 compounds T19.001 to T19.105 of the formula Ia, wherein R is CH₂-O-C₂H₄-O-CH₃, R₁, R₂, R₃ and R₄ are hydrogen, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

5

Table 20 This table discloses the 105 compounds T20.001 to T20.105 of the formula Ia, wherein R is hydrogen, R₁, R₂, R₃ and R₄ are CH₃, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

10 Table 21 This table discloses the 105 compounds T21.001 to T21.105 of the formula Ia, wherein R is CH₃, R₁, R₂, R₃ and R₄ are CH₃, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

15 Table 22 This table discloses the 105 compounds T22.001 to T22.105 of the formula Ia, wherein R is C₂H₅, R₁, R₂, R₃ and R₄ are CH₃, G is hydrogen and R_a, R_b, R_c and R_d are as defined in Table 1.

The compounds according to the invention are preventively and/or curatively valuable active ingredients in the field of pest control, even at low rates of application, which have a very
20 favorable biocidal spectrum and are well tolerated by warm-blooded species, fish and plants. The active ingredients according to the invention act against all or individual developmental stages of normally sensitive, but also resistant, animal pests, such as insects or representatives of the order Acarina. The insecticidal or acaricidal activity of the active ingredients according to the invention can manifest itself directly, i. e. in destruction
25 of the pests, which takes place either immediately or only after some time has elapsed, for example during ecdysis, or indirectly, for example in a reduced oviposition and/or hatching rate, a good activity corresponding to a destruction rate (mortality) of at least 50 to 60%.

The compounds of formula I can be used to combat and control infestations of insect pests
30 such as Lepidoptera, Diptera, Hemiptera, Thysanoptera, Orthoptera, Dictyoptera, Coleoptera, Siphonaptera, Hymenoptera and Isoptera and also other invertebrate pests, for example, acarine, nematode and mollusc pests. Insects, acarines, nematodes and molluscs are hereinafter collectively referred to as pests. The pests which may be combated and controlled by the use of the invention compounds include those pests
35 associated with agriculture (which term includes the growing of crops for food and fibre products), horticulture and animal husbandry, companion animals, forestry and the storage

of products of vegetable origin (such as fruit, grain and timber); those pests associated with the damage of man-made structures and the transmission of diseases of man and animals; and also nuisance pests (such as flies).

- 5 Examples of pest species which may be controlled by the compounds of formula I include: *Myzus persicae* (aphid), *Aphis gossypii* (aphid), *Aphis fabae* (aphid), *Lygus* spp. (capsids), *Dysdercus* spp. (capsids), *Nilaparvata lugens* (planthopper), *Nephotettix inciticeps* (leafhopper), *Nezara* spp. (stinkbugs), *Euschistus* spp. (stinkbugs), *Leptocorisa* spp. (stinkbugs), *Frankliniella occidentalis* (thrip), *Thrips* spp. (thrips), *Leptinotarsa decemlineata* (Colorado potato beetle), *Anthonomus grandis* (boll weevil), *Aonidiella* spp. (scale insects),
10 *Trialeurodes* spp. (white flies), *Bemisia tabaci* (white fly), *Ostrinia nubilalis* (European corn borer), *Spodoptera littoralis* (cotton leafworm), *Heliothis virescens* (tobacco budworm), *Helicoverpa armigera* (cotton bollworm), *Helicoverpa zea* (cotton bollworm), *Sylepta derogata* (cotton leaf roller), *Pieris brassicae* (white butterfly), *Plutella xylostella* (diamond
15 back moth), *Agrotis* spp. (cutworms), *Chilo suppressalis* (rice stem borer), *Locusta migratoria* (locust), *Chortiocetes terminifera* (locust), *Diabrotica* spp. (rootworms), *Panonychus ulmi* (European red mite), *Panonychus citri* (citrus red mite), *Tetranychus urticae* (two-spotted spider mite), *Tetranychus cinnabarinus* (carmine spider mite), *Phyllocoptruta oleivora* (citrus rust mite), *Polyphagotarsonemus latus* (broad mite),
20 *Brevipalpus* spp. (flat mites), *Boophilus microplus* (cattle tick), *Dermacentor variabilis* (American dog tick), *Ctenocephalides felis* (cat flea), *Liriomyza* spp. (leafminer), *Musca domestica* (housefly), *Aedes aegypti* (mosquito), *Anopheles* spp. (mosquitoes), *Culex* spp. (mosquitoes), *Lucilia* spp. (blowflies), *Blattella germanica* (cockroach), *Periplaneta americana* (cockroach), *Blatta orientalis* (cockroach), termites of the Mastotermitidae (for
25 example *Mastotermes* spp.), the Kalotermitidae (for example *Neotermes* spp.), the Rhinotermitidae (for example *Coptotermes formosanus*, *Reticulitermes flavipes*, *R. speratu*, *R. virginicus*, *R. hesperus*, and *R. santonensis*) and the Termitidae (for example *Globitermes sulphureus*), *Solenopsis geminata* (fire ant), *Monomorium pharaonis* (pharaoh's ant), *Damalinia* spp. and *Linognathus* spp. (biting and sucking lice), and
30 *Deroceras reticulatum* (slug).

The active ingredients according to the invention can be used for controlling, i. e. containing or destroying, pests of the abovementioned type which occur in particular on plants, especially on useful plants and ornamentals in agriculture, in horticulture and in forests, or on organs, such as fruits, flowers, foliage, stalks, tubers or roots, of such plants, and in

some cases even plant organs which are formed at a later point in time remain protected against these pests.

Suitable target crops are, in particular, cereals, such as wheat, barley, rye, oats, rice, maize or sorghum; beet, such as sugar or fodder beet; fruit, for example pomaceous fruit, stone
5 fruit or soft fruit, such as apples, pears, plums, peaches, almonds, cherries or berries, for example strawberries, raspberries or blackberries; leguminous crops, such as beans, lentils, peas or soya; oil crops, such as oilseed rape, mustard, poppies, olives, sunflowers, coconut, castor, cocoa or ground nuts; cucurbits, such as pumpkins, cucumbers or melons; fibre plants, such as cotton, flax, hemp or jute; citrus fruit, such as oranges, lemons,
10 grapefruit or tangerines; vegetables, such as spinach, lettuce, asparagus, cabbages, carrots, onions, tomatoes, potatoes or bell peppers; Lauraceae, such as avocado, Cinnamonum or camphor; and also tobacco, nuts, coffee, eggplants, sugarcane, tea, pepper, grapevines, hops, the plantain family, latex plants and ornamentals.

The term "crops" is to be understood as including also crops that have been rendered
15 tolerant to herbicides like bromoxynil or classes of herbicides (such as, for example, HPPD inhibitors, ALS inhibitors, for example primisulfuron, prosulfuron and trifloxysulfuron, EPSPS (5-enol-pyrovyl-shikimate-3-phosphate-synthase) inhibitors, GS (glutamine synthetase) inhibitors) as a result of conventional methods of breeding or genetic engineering. An example of a crop that has been rendered tolerant to imidazolinones, e.g. imazamox, by
20 conventional methods of breeding (mutagenesis) is Clearfield® summer rape (Canola). Examples of crops that have been rendered tolerant to herbicides or classes of herbicides by genetic engineering methods include glyphosate- and glufosinate-resistant maize varieties commercially available under the trade names RoundupReady® and LibertyLink®.

25 The term "crops" is also to be understood as including also crop plants which have been so transformed by the use of recombinant DNA techniques that they are capable of synthesising one or more selectively acting toxins, such as are known, for example, from toxin-producing bacteria, especially those of the genus *Bacillus*.

30 Toxins that can be expressed by such transgenic plants include, for example, insecticidal proteins, for example insecticidal proteins from *Bacillus cereus* or *Bacillus popilliae*; or insecticidal proteins from *Bacillus thuringiensis*, such as δ -endotoxins, e.g. CryIA(b), CryIA(c), CryIF, CryIF(a2), CryIIA(b), CryIIIA, CryIIIB(b1) or Cry9c, or vegetative insecticidal proteins (VIP), e.g. VIP1, VIP2, VIP3 or VIP3A; or insecticidal proteins of bacteria
35 colonising nematodes, for example *Photorhabdus* spp. or *Xenorhabdus* spp., such as

Photorhabdus luminescens, Xenorhabdus nematophilus; toxins produced by animals, such as scorpion toxins, arachnid toxins, wasp toxins and other insect-specific neurotoxins; toxins produced by fungi, such as Streptomyces toxins, plant lectins, such as pea lectins, barley lectins or snowdrop lectins; agglutinins; proteinase inhibitors, such as trypsin inhibitors, serine protease inhibitors, patatin, cystatin, papain inhibitors; ribosome-inactivating proteins (RIP), such as ricin, maize-RIP, abrin, luffin, saporin or bryodin; steroid metabolism enzymes, such as 3-hydroxysteroidoxidase, ecdysteroid-UDP-glycosyl-transferase, cholesterol oxidases, ecdysone inhibitors, HMG-CoA-reductase, ion channel blockers, such as blockers of sodium or calcium channels, juvenile hormone esterase, diuretic hormone receptors, stilbene synthase, bibenzyl synthase, chitinases and glucanases.

In the context of the present invention there are to be understood by δ -endotoxins, for example CryIA(b), CryIA(c), CryIF, CryIF(a2), CryIIA(b), CryIIIA, CryIIIB(b1) or Cry9c, or vegetative insecticidal proteins (VIP), for example VIP1, VIP2, VIP3 or VIP3A, expressly also hybrid toxins, truncated toxins and modified toxins. Hybrid toxins are produced recombinantly by a new combination of different domains of those proteins (see, for example, WO 02/15701). Truncated toxins, for example a truncated CryIA(b), are known. In the case of modified toxins, one or more amino acids of the naturally occurring toxin are replaced. In such amino acid replacements, preferably non-naturally present protease recognition sequences are inserted into the toxin, such as, for example, in the case of CryIIIA055, a cathepsin-D-recognition sequence is inserted into a CryIIIA toxin (see WO 03/018810).

Examples of such toxins or transgenic plants capable of synthesising such toxins are disclosed, for example, in EP-A-0 374 753, WO 93/07278, WO 95/34656, EP-A-0 427 529, EP-A-451 878 and WO 03/052073.

The processes for the preparation of such transgenic plants are generally known to the person skilled in the art and are described, for example, in the publications mentioned above. CryI-type deoxyribonucleic acids and their preparation are known, for example, from WO 95/34656, EP-A-0 367 474, EP-A-0 401 979 and WO 90/13651.

The toxin contained in the transgenic plants imparts to the plants tolerance to harmful insects. Such insects can occur in any taxonomic group of insects, but are especially

commonly found in the beetles (Coleoptera), two-winged insects (Diptera) and butterflies (Lepidoptera).

Transgenic plants containing one or more genes that code for an insecticidal resistance and
5 express one or more toxins are known and some of them are commercially available.
Examples of such plants are: YieldGard® (maize variety that expresses a CryIA(b) toxin);
YieldGard Rootworm® (maize variety that expresses a CryIIIB(b1) toxin); YieldGard Plus®
(maize variety that expresses a CryIA(b) and a CryIIIB(b1) toxin); Starlink® (maize variety
that expresses a Cry9(c) toxin); Herculex I® (maize variety that expresses a CryIF(a2) toxin
10 and the enzyme phosphinothricine N-acetyltransferase (PAT) to achieve tolerance to the
herbicide glufosinate ammonium); NuCOTN 33B® (cotton variety that expresses a CryIA(c)
toxin); Bollgard I® (cotton variety that expresses a CryIA(c) toxin); Bollgard II® (cotton
variety that expresses a CryIA(c) and a CryIIA(b) toxin); VIPCOT® (cotton variety that
expresses a VIP toxin); NewLeaf® (potato variety that expresses a CryIIIA toxin); Nature-
15 Gard® Agrisure® GT Advantage (GA21 glyphosate-tolerant trait), Agrisure® CB Advantage
(Bt11 corn borer (CB) trait) and Protecta®.

Further examples of such transgenic crops are:

- 20 1. Bt11 Maize from Syngenta Seeds SAS, Chemin de l'Hobit 27, F-31 790 St. Sauveur,
France, registration number C/FR/96/05/10. Genetically modified *Zea mays* which has been
rendered resistant to attack by the European corn borer (*Ostrinia nubilalis* and *Sesamia
nonagrioides*) by transgenic expression of a truncated CryIA(b) toxin. Bt11 maize also
transgenically expresses the enzyme PAT to achieve tolerance to the herbicide glufosinate
25 ammonium.
2. Bt176 Maize from Syngenta Seeds SAS, Chemin de l'Hobit 27, F-31 790 St. Sauveur,
France, registration number C/FR/96/05/10. Genetically modified *Zea mays* which has been
rendered resistant to attack by the European corn borer (*Ostrinia nubilalis* and *Sesamia
30 nonagrioides*) by transgenic expression of a CryIA(b) toxin. Bt176 maize also transgenically
expresses the enzyme PAT to achieve tolerance to the herbicide glufosinate ammonium.
3. MIR604 Maize from Syngenta Seeds SAS, Chemin de l'Hobit 27, F-31 790 St. Sauveur,
France, registration number C/FR/96/05/10. Maize which has been rendered insect-
35 resistant by transgenic expression of a modified CryIIIA toxin. This toxin is Cry3A055

modified by insertion of a cathepsin-D-protease recognition sequence. The preparation of such transgenic maize plants is described in WO 03/018810.

4. MON 863 Maize from Monsanto Europe S.A. 270-272 Avenue de Tervuren, B-1150
5 Brussels, Belgium, registration number C/DE/02/9. MON 863 expresses a CryIIIB(b1) toxin and has resistance to certain Coleoptera insects.

5. IPC 531 Cotton from Monsanto Europe S.A. 270-272 Avenue de Tervuren, B-1150
Brussels, Belgium, registration number C/ES/96/02.

10

6. 1507 Maize from Pioneer Overseas Corporation, Avenue Tedesco, 7 B-1160 Brussels, Belgium, registration number C/NL/00/10. Genetically modified maize for the expression of the protein Cry1F for achieving resistance to certain Lepidoptera insects and of the PAT protein for achieving tolerance to the herbicide glufosinate ammonium.

15

7. NK603 × MON 810 Maize from Monsanto Europe S.A. 270-272 Avenue de Tervuren, B-1150 Brussels, Belgium, registration number C/GB/02/M3/03. Consists of conventionally bred hybrid maize varieties by crossing the genetically modified varieties NK603 and MON 810. NK603 × MON 810 Maize transgenically expresses the protein CP4 EPSPS, obtained
20 from *Agrobacterium* sp. strain CP4, which imparts tolerance to the herbicide Roundup® (contains glyphosate), and also a CryIA(b) toxin obtained from *Bacillus thuringiensis subsp. kurstaki* which brings about tolerance to certain Lepidoptera, include the European corn borer.

25 Transgenic crops of insect-resistant plants are also described in BATS (Zentrum für Biosicherheit und Nachhaltigkeit, Zentrum BATS, Clarastrasse 13, 4058 Basel, Switzerland) Report 2003.

The term "crops" is to be understood as including also crop plants which have been so
30 transformed by the use of recombinant DNA techniques that they are capable of synthesising antipathogenic substances having a selective action, such as, for example, the so-called "pathogenesis-related proteins" (PRPs, see e.g. EP-A-0 392 225). Examples of such antipathogenic substances and transgenic plants capable of synthesising such antipathogenic substances are known, for example, from EP-A-0 392 225, WO 95/33818,
35 and EP-A-0 353 191. The methods of producing such transgenic plants are generally

known to the person skilled in the art and are described, for example, in the publications mentioned above.

Antipathogenic substances which can be expressed by such transgenic plants include, for example, ion channel blockers, such as blockers for sodium and calcium channels, for example the viral KP1, KP4 or KP6 toxins; stilbene synthases; bibenzyl synthases; chitinases; glucanases; the so-called "pathogenesis-related proteins" (PRPs; see e.g. EP-A-0 392 225); antipathogenic substances produced by microorganisms, for example peptide antibiotics or heterocyclic antibiotics (see e.g. WO 95/33818) or protein or polypeptide factors involved in plant pathogen defence (so-called "plant disease resistance genes", as described in WO 03/000906).

Further areas of use of the compounds according to the invention are the protection of stored goods and storerooms and the protection of raw materials, such as wood, textiles, floor coverings or buildings, and also in the hygiene sector, especially the protection of humans, domestic animals and productive livestock against pests of the mentioned type.

In the hygiene sector, the compounds according to the invention are active against ectoparasites such as hard ticks, soft ticks, mange mites, harvest mites, flies (biting and licking), parasitic fly larvae, lice, hair lice, bird lice and fleas.

Examples of such parasites are:

Of the order Anoplurida: Haematopinus spp., Linognathus spp., Pediculus spp. and Phtirus spp., Solenopotes spp..

Of the order Mallophagida: Trimenopon spp., Menopon spp., Trinoton spp., Bovicola spp., Werneckiella spp., Lepikentron spp., Damalina spp., Trichodectes spp. and Felicola spp..

Of the order Diptera and the suborders Nematocerina and Brachycerina, for example Aedes spp., Anopheles spp., Culex spp., Simulium spp., Eusimulium spp., Phlebotomus spp., Lutzomyia spp., Culicoides spp., Chrysops spp., Hybomitra spp., Atylotus spp., Tabanus spp., Haematopota spp., Philipomyia spp., Braula spp., Musca spp., Hydrotaea spp., Stomoxys spp., Haematobia spp., Morellia spp., Fannia spp., Glossina spp., Calliphora spp., Lucilia spp., Chrysomyia spp., Wohlfahrtia spp., Sarcophaga spp., Oestrus spp., Hypoderma spp., Gasterophilus spp., Hippobosca spp., Lipoptena spp. and Melophagus spp..

Of the order Siphonaptera, for example *Pulex* spp., *Ctenocephalides* spp., *Xenopsylla* spp., *Ceratophyllus* spp..

Of the order Heteroptera, for example *Cimex* spp., *Triatoma* spp., *Rhodnius* spp.,
5 *Panstrongylus* spp..

Of the order Blattaria, for example *Blatta orientalis*, *Periplaneta americana*,
Blattella germanica and *Supella* spp..

Of the subclass Acaria (Acarida) and the orders Meta- and Meso-stigmata, for example
Argas spp., *Ornithodoros* spp., *Otobius* spp., *Ixodes* spp., *Amblyomma* spp., *Boophilus*
10 spp., *Dermacentor* spp., *Haemophysalis* spp., *Hyalomma* spp., *Rhipicephalus* spp.,
Dermanyssus spp., *Raillietia* spp., *Pneumonyssus* spp., *Sternostoma* spp. and *Varroa* spp..

Of the orders Actinieda (Prostigmata) and Acaridida (Astigmata), for example *Acarapis*
spp., *Cheyletiella* spp., *Ornithocheyletia* spp., *Myobia* spp., *Psorergates* spp., *Demodex*
spp., *Trombicula* spp., *Listrophorus* spp., *Acarus* spp., *Tyrophagus* spp., *Caloglyphus* spp.,
15 *Hypodectes* spp., *Pterolichus* spp., *Psoroptes* spp., *Chorioptes* spp., *Otodectes* spp.,
Sarcoptes spp., *Notoedres* spp., *Knemidocoptes* spp., *Cytodites* spp. and *Laminosioptes*
spp..

The compounds according to the invention are also suitable for protecting against insect
infestation in the case of materials such as wood, textiles, plastics, adhesives, glues, paints,
20 paper and card, leather, floor coverings and buildings.

The invention therefore provides a method of combating and controlling insects, acarines,
nematodes or molluscs which comprises applying an insecticidally, acaricidally,
nematicidally or molluscicidally effective amount of a compound of formula I, or a
composition containing a compound of formula I, to a pest, a locus of pest, or to a plant
25 susceptible to attack by a pest, The compounds of formula I are preferably used against
insects or acarines.

The term "plant" as used herein includes seedlings, bushes and trees.

The invention therefore also relates to pesticidal compositions such as emulsifiable
30 concentrates, suspension concentrates, directly sprayable or dilutable solutions, spreadable
pastes, dilute emulsions, soluble powders, dispersible powders, wettable powders, dusts,

granules or encapsulations in polymeric substances, which comprise - at least - one of the active ingredients according to the invention and which are to be selected to suit the intended aims and the prevailing circumstances.

5 In these compositions, the active ingredient is employed in pure form, a solid active ingredient for example in a specific particle size, or, preferably, together with - at least - one of the auxiliaries conventionally used in the art of formulation, such as extenders, for example solvents or solid carriers, or such as surface-active compounds (surfactants).

10 Examples of suitable solvents are: unhydrogenated or partially hydrogenated aromatic hydrocarbons, preferably the fractions C8 to C12 of alkylbenzenes, such as xylene mixtures, alkylated naphthalenes or tetrahydronaphthalene, aliphatic or cycloaliphatic hydrocarbons, such as paraffins or cyclohexane, alcohols such as ethanol, propanol or butanol, glycols and their ethers and esters such as propylene glycol, dipropylene glycol
15 ether, ethylene glycol or ethylene glycol monomethyl ether or ethylene glycol monoethyl ether, ketones, such as cyclohexanone, isophorone or diacetone alcohol, strongly polar solvents, such as N-methylpyrrolid-2-one, dimethyl sulfoxide or N,N-dimethylformamide, water, unepoxidized or epoxidized vegetable oils, such as unexpodized or epoxidized rapeseed, castor, coconut or soya oil, and silicone oils.

20

Solid carriers which are used for example for dusts and dispersible powders are, as a rule, ground natural minerals such as calcite, talc, kaolin, montmorillonite or attapulgite. To improve the physical properties, it is also possible to add highly disperse silicas or highly disperse absorbtive polymers. Suitable particulate adsorptive carriers for granules are
25 porous types, such as pumice, brick grit, sepiolite or bentonite, and suitable non-sorptive carrier materials are calcite or sand. In addition, a large number of granulated materials of inorganic or organic nature can be used, in particular dolomite or comminuted plant residues.

30 Suitable surface-active compounds are, depending on the type of the active ingredient to be formulated, non-ionic, cationic and/or anionic surfactants or surfactant mixtures which have good emulsifying, dispersing and wetting properties. The surfactants mentioned below are only to be considered as examples; a large number of further surfactants which are conventionally used in the art of formulation and suitable according to the invention are
35 described in the relevant literature.

Suitable non-ionic surfactants are, especially, polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, of saturated or unsaturated fatty acids or of alkyl phenols which may contain approximately 3 to approximately 30 glycol ether groups and approximately 8 to approximately 20 carbon atoms in the (cyclo)aliphatic hydrocarbon radical or approximately 6 to approximately 18 carbon atoms in the alkyl moiety of the alkyl phenols. Also suitable are water-soluble polyethylene oxide adducts with polypropylene glycol, ethylenediaminopolypropylene glycol or alkyl polypropylene glycol having 1 to approximately 10 carbon atoms in the alkyl chain and approximately 20 to approximately 250 ethylene glycol ether groups and approximately 10 to approximately 100 propylene glycol ether groups. Normally, the abovementioned compounds contain 1 to approximately 5 ethylene glycol units per propylene glycol unit. Examples which may be mentioned are nonylphenoxypolyethoxyethanol, castor oil polyglycol ether, polypropylene glycol/polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol or octylphenoxypolyethoxyethanol. Also suitable are fatty acid esters of polyoxyethylene sorbitan, such as polyoxyethylene sorbitan trioleate.

The cationic surfactants are, especially, quarternary ammonium salts which generally have at least one alkyl radical of approximately 8 to approximately 22 C atoms as substituents and as further substituents (unhalogenated or halogenated) lower alkyl or hydroxyalkyl or benzyl radicals. The salts are preferably in the form of halides, methylsulfates or ethylsulfates. Examples are stearyltrimethylammonium chloride and benzylbis(2-chloroethyl)ethylammonium bromide.

Examples of suitable anionic surfactants are water-soluble soaps or water-soluble synthetic surface-active compounds. Examples of suitable soaps are the alkali, alkaline earth or (unsubstituted or substituted) ammonium salts of fatty acids having approximately 10 to approximately 22 C atoms, such as the sodium or potassium salts of oleic or stearic acid, or of natural fatty acid mixtures which are obtainable for example from coconut or tall oil; mention must also be made of the fatty acid methyl taurates. However, synthetic surfactants are used more frequently, in particular fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylaryl sulfonates. As a rule, the fatty sulfonates and fatty sulfates are present as alkali, alkaline earth or (substituted or unsubstituted) ammonium salts and they generally have an alkyl radical of approximately 8 to approximately 22 C atoms, alkyl also to be understood as including the alkyl moiety of acyl radicals; examples which may be mentioned are the sodium or calcium salts of lignosulfonic acid, of the dodecylsulfuric ester or of a fatty alcohol sulfate mixture prepared from natural fatty acids.

This group also includes the salts of the sulfuric esters and sulfonic acids of fatty alcohol/ethylene oxide adducts. The sulfonated benzimidazole derivatives preferably contain 2 sulfonyl groups and a fatty acid radical of approximately 8 to approximately 22 C atoms. Examples of alkylarylsulfonates are the sodium, calcium or triethanolammonium salts of decylbenzenesulfonic acid, of dibutyl-naphthalenesulfonic acid or of a naphthalenesulfonic acid/formaldehyde condensate. Also possible are, furthermore, suitable phosphates, such as salts of the phosphoric ester of a p-nonylphenol/(4-14)ethylene oxide adduct, or phospholipids.

- 10 As a rule, the compositions comprise 0.1 to 99%, especially 0.1 to 95%, of active ingredient and 1 to 99.9%, especially 5 to 99.9%, of at least one solid or liquid adjuvant, it being possible as a rule for 0 to 25%, especially 0.1 to 20%, of the composition to be surfactants(% in each case meaning percent by weight). Whereas concentrated compositions tend to be preferred for commercial goods, the end consumer as a rule uses
15 dilute compositions which have substantially lower concentrations of active ingredient. Preferred compositions are composed in particular as follows (% = percent by weight):

Emulsifiable concentrates:

- | | |
|--------------------|---------------------------------|
| active ingredient: | 1 to 95%, preferably 5 to 20% |
| 20 surfactant: | 1 to 30%, preferably 10 to 20 % |
| solvent: | 5 to 98%, preferably 70 to 85% |

Dusts:

- | | |
|--------------------|-------------------------------------|
| active ingredient: | 0.1 to 10%, preferably 0.1 to 1% |
| 25 solid carrier: | 99.9 to 90%, preferably 99.9 to 99% |

Suspension concentrates:

- | | |
|--------------------|---------------------------------|
| active ingredient: | 5 to 75%, preferably 10 to 50% |
| water: | 94 to 24%, preferably 88 to 30% |
| 30 surfactant: | 1 to 40%, preferably 2 to 30% |

Wettable powders:

- | | |
|--------------------|---------------------------------|
| active ingredient: | 0.5 to 90%, preferably 1 to 80% |
| surfactant: | 0.5 to 20%, preferably 1 to 15% |
| 35 solid carrier: | 5 to 99%, preferably 15 to 98% |

Granulates:

active ingredient: 0.5 to 30%, preferably 3 to 15%

solid carrier: 99.5 to 70%, preferably 97 to 85%

- 5 The compositions can also comprise further solid or liquid auxiliaries, such as stabilizers, for example unepoxidized or epoxidized vegetable oils (for example epoxidized coconut oil, rapeseed oil or soya oil), antifoams, for example silicone oil, preservatives, viscosity regulators, binders and/or tackifiers, fertilizers or other active ingredients for achieving specific effects, for example bactericides, fungicides, nematocides, plant activators,
10 molluscicides or herbicides.

The compositions according to the invention are prepared in a manner known per se, in the absence of auxiliaries for example by grinding, screening and/or compressing a solid active ingredient and in the presence of at least one auxiliary for example by intimately mixing
15 and/or grinding the active ingredient with the auxiliary (auxiliaries). These processes for the preparation of the compositions and the use of the compounds I for the preparation of these compositions are also a subject of the invention.

The application methods for the compositions, that is the methods of controlling pests of the
20 abovementioned type, such as spraying, atomizing, dusting, brushing on, dressing, scattering or pouring - which are to be selected to suit the intended aims of the prevailing circumstances - and the use of the compositions for controlling pests of the abovementioned type are other subjects of the invention. Typical rates of concentration are between 0.1 and 1000 ppm, preferably between 0.1 and 500 ppm, of active ingredient. The
25 rate of application per hectare is generally 1 to 2000 g of active ingredient per hectare, in particular 10 to 1000 g/ha, preferably 10 to 600 g/ha.

A preferred method of application in the field of crop protection is application to the foliage of the plants (foliar application), it being possible to select frequency and rate of application
30 to match the danger of infestation with the pest in question. Alternatively, the active ingredient can reach the plants via the root system (systemic action), by drenching the locus of the plants with a liquid composition or by incorporating the active ingredient in solid form into the locus of the plants, for example into the soil, for example in the form of granules (soil application). In the case of paddy rice crops, such granules can be metered into the
35 flooded paddy-field.

The compositions according to the invention are also suitable for the protection of plant propagation material, for example seeds, such as fruit, tubers or kernels, or nursery plants, against pests of the abovementioned type. The propagation material can be treated with the compositions prior to planting, for example seed can be treated prior to sowing.

5 Alternatively, the compositions can be applied to seed kernels (coating), either by soaking the kernels in a liquid composition or by applying a layer of a solid composition. It is also possible to apply the compositions when the propagation material is planted to the site of application, for example into the seed furrow during drilling. These treatment methods for plant propagation material and the plant propagation material thus treated are further
10 subjects of the invention.

In order to apply a compound of formula I as an insecticide, acaricide, nematocide or molluscicide to a pest, a locus of pest, or to a plant susceptible to attack by a pest, a compound of formula I is usually formulated into a composition which includes, in addition to
15 the compound of formula I, a suitable inert diluent or carrier and, optionally, a formulation adjuvant in form of a surface active agent (SFA) as described herein or, for example, in EP-B-1062217. SFAs are chemicals which are able to modify the properties of an interface (for example, liquid/solid, liquid/air or liquid/liquid interfaces) by lowering the interfacial tension and thereby leading to changes in other properties (for example dispersion, emulsification
20 and wetting). It is preferred that all compositions (both solid and liquid formulations) comprise, by weight, 0.0001 to 95%, more preferably 1 to 85%, for example 5 to 60%, of a compound of formula I. The composition is generally used for the control of pests such that a compound of formula I is applied at a rate of from 0.1g to 10kg per hectare, preferably from 1g to 6kg per hectare, more preferably from 1g to 1kg per hectare.

25 When used in a seed dressing, a compound of formula I is used at a rate of 0.0001g to 10g (for example 0.001g or 0.05g), preferably 0.005g to 10g, more preferably 0.005g to 4g, per kilogram of seed.

30 In another aspect the present invention provides an insecticidal, acaricidal, nematocidal or molluscicidal composition comprising an insecticidally, acaricidally, nematocidally or molluscicidally effective amount of a compound of formula I and a suitable carrier or diluent therefor.

35 In a still further aspect the invention provides a method of combating and controlling pests at a locus which comprises treating the pests or the locus of the pests with an insecticidally,

acaricidally, nematocidally or molluscicidally effective amount of a composition comprising a compound of formula I.

The compositions can be chosen from a number of formulation types, including dustable
5 powders (DP), soluble powders (SP), water soluble granules (SG), water dispersible
granules (WG), wettable powders (WP), granules (GR) (slow or fast release), soluble
concentrates (SL), oil miscible liquids (OL), ultra low volume liquids (UL), emulsifiable
concentrates (EC), dispersible concentrates (DC), emulsions (both oil in water (EW) and
10 water in oil (EO)), micro-emulsions (ME), suspension concentrates (SC), aerosols,
fogging/smoke formulations, capsule suspensions (CS) and seed treatment formulations.
The formulation type chosen in any instance will depend upon the particular purpose en-
visaged and the physical, chemical and biological properties of the compound of formula I.

Dustable powders (DP) may be prepared by mixing a compound of formula I with one or
15 more solid diluents (for example natural clays, kaolin, pyrophyllite, bentonite, alumina,
montmorillonite, kieselguhr, chalk, diatomaceous earths, calcium phosphates, calcium and
magnesium carbonates, sulphur, lime, flours, talc and other organic and inorganic solid
carriers) and mechanically grinding the mixture to a fine powder.

20 Soluble powders (SP) may be prepared by mixing a compound of formula I with one or
more water-soluble inorganic salts (such as sodium bicarbonate, sodium carbonate or
magnesium sulphate) or one or more water-soluble organic solids (such as a
polysaccharide) and, optionally, one or more wetting agents, one or more dispersing agents
or a mixture of said agents to improve water dispersibility/solubility. The mixture is then
25 ground to a fine powder. Similar compositions may also be granulated to form water
soluble granules (SG).

Wettable powders (WP) may be prepared by mixing a compound of formula I with one or
more solid diluents or carriers, one or more wetting agents and, preferably, one or more
30 dispersing agents and, optionally, one or more suspending agents to facilitate the
dispersion in liquids. The mixture is then ground to a fine powder. Similar compositions
may also be granulated to form water dispersible granules (WG).

Granules (GR) may be formed either by granulating a mixture of a compound of formula I
35 and one or more powdered solid diluents or carriers, or from pre-formed blank granules by
absorbing a compound of formula I (or a solution thereof, in a suitable agent) in a porous

granular material (such as pumice, attapulgite clays, fuller's earth, kieselguhr, diatomaceous earths or ground corn cobs) or by adsorbing a compound of formula I (or a solution thereof, in a suitable agent) on to a hard core material (such as sands, silicates, mineral carbonates, sulphates or phosphates) and drying if necessary. Agents which are commonly used to aid absorption or adsorption include solvents (such as aliphatic and aromatic petroleum solvents, alcohols, ethers, ketones and esters) and sticking agents (such as polyvinyl acetates, polyvinyl alcohols, dextrans, sugars and vegetable oils). One or more other additives may also be included in granules (for example an emulsifying agent, wetting agent or dispersing agent).

Dispersible Concentrates (DC) may be prepared by dissolving a compound of formula I in water or an organic solvent, such as a ketone, alcohol or glycol ether. These solutions may contain a surface active agent (for example to improve water dilution or prevent crystallisation in a spray tank).

Emulsifiable concentrates (EC) or oil-in-water emulsions (EW) may be prepared by dissolving a compound of formula I in an organic solvent (optionally containing one or more wetting agents, one or more emulsifying agents or a mixture of said agents). Suitable organic solvents for use in ECs include aromatic hydrocarbons (such as alkylbenzenes or alkylnaphthalenes, exemplified by SOLVESSO 100, SOLVESSO 150 and SOLVESSO 200; SOLVESSO is a Registered Trade Mark), ketones (such as cyclohexanone or methylcyclohexanone) and alcohols (such as benzyl alcohol, furfuryl alcohol or butanol), N-alkylpyrrolidones (such as N-methylpyrrolidone or N-octylpyrrolidone), dimethyl amides of fatty acids (such as C₈-C₁₀ fatty acid dimethylamide) and chlorinated hydrocarbons. An EC product may spontaneously emulsify on addition to water, to produce an emulsion with sufficient stability to allow spray application through appropriate equipment. Preparation of an EW involves obtaining a compound of formula I either as a liquid (if it is not a liquid at room temperature, it may be melted at a reasonable temperature, typically below 70°C) or in solution (by dissolving it in an appropriate solvent) and then emulsifying the resultant liquid or solution into water containing one or more SFAs, under high shear, to produce an emulsion. Suitable solvents for use in EWs include vegetable oils, chlorinated hydrocarbons (such as chlorobenzenes), aromatic solvents (such as alkylbenzenes or alkylnaphthalenes) and other appropriate organic solvents which have a low solubility in water.

Microemulsions (ME) may be prepared by mixing water with a blend of one or more solvents with one or more SFAs, to produce spontaneously a thermodynamically stable isotropic liquid formulation. A compound of formula I is present initially in either the water or the solvent/SFA blend. Suitable solvents for use in MEs include those hereinbefore
5 described for use in ECs or in EWs. An ME may be either an oil-in-water or a water-in-oil system (which system is present may be determined by conductivity measurements) and may be suitable for mixing water-soluble and oil-soluble pesticides in the same formulation. An ME is suitable for dilution into water, either remaining as a microemulsion or forming a conventional oil-in-water emulsion.

10

Suspension concentrates (SC) may comprise aqueous or non-aqueous suspensions of finely divided insoluble solid particles of a compound of formula I. SCs may be prepared by ball or bead milling the solid compound of formula I in a suitable medium, optionally with one or more dispersing agents, to produce a fine particle suspension of the compound.
15 One or more wetting agents may be included in the composition and a suspending agent may be included to reduce the rate at which the particles settle. Alternatively, a compound of formula I may be dry milled and added to water, containing agents hereinbefore described, to produce the desired end product.

20 Aerosol formulations comprise a compound of formula I and a suitable propellant (for example *n*-butane). A compound of formula I may also be dissolved or dispersed in a suitable medium (for example water or a water miscible liquid, such as *n*-propanol) to provide compositions for use in non-pressurised, hand-actuated spray pumps.

25 A compound of formula I may be mixed in the dry state with a pyrotechnic mixture to form a composition suitable for generating, in an enclosed space, a smoke containing the compound.

Capsule suspensions (CS) may be prepared in a manner similar to the preparation of EW
30 formulations but with an additional polymerisation stage such that an aqueous dispersion of oil droplets is obtained, in which each oil droplet is encapsulated by a polymeric shell and contains a compound of formula I and, optionally, a carrier or diluent therefor. The polymeric shell may be produced by either an interfacial polycondensation reaction or by a coacervation procedure. The compositions may provide for controlled release of the
35 compound of formula I and they may be used for seed treatment. A compound of formula I

may also be formulated in a biodegradable polymeric matrix to provide a slow, controlled release of the compound.

5 A composition of the present invention may include one or more additives to improve the biological performance of the composition (for example by improving wetting, retention or distribution on surfaces; resistance to rain on treated surfaces; or uptake or mobility of a compound of formula I). Such additives include surface active agents, spray additives based on oils, for example certain mineral oils or natural plant oils (such as soy bean and rape seed oil), and blends of these with other bio-enhancing adjuvants (ingredients which
10 may aid or modify the action of a compound of formula I).

A compound of formula I may also be formulated for use as a seed treatment, for example as a powder composition, including a powder for dry seed treatment (DS), a water soluble powder (SS) or a water dispersible powder for slurry treatment (WS), or as a liquid
15 composition, including a flowable concentrate (FS), a solution (LS) or a capsule suspension (CS). The preparations of DS, SS, WS, FS and LS compositions are very similar to those of, respectively, DP, SP, WP, SC and DC compositions described above. Compositions for treating seed may include an agent for assisting the adhesion of the composition to the seed (for example a mineral oil or a film-forming barrier).

20

Wetting agents, dispersing agents and emulsifying agents may be surface active agents (SFAs) of the cationic, anionic, amphoteric or non-ionic type.

Suitable SFAs of the cationic type include quaternary ammonium compounds (for example
25 cetyltrimethyl ammonium bromide), imidazolines and amine salts.

Suitable anionic SFAs include alkali metals salts of fatty acids, salts of aliphatic monoesters of sulphuric acid (for example sodium lauryl sulphate), salts of sulphonated aromatic compounds (for example sodium dodecylbenzenesulphonate, calcium
30 dodecylbenzenesulphonate, butylnaphthalene sulphonate and mixtures of sodium di-*isopropyl*- and tri-*isopropyl*-naphthalene sulphonates), ether sulphates, alcohol ether sulphates (for example sodium laureth-3-sulphate), ether carboxylates (for example sodium laureth-3-carboxylate), phosphate esters (products from the reaction between one or more fatty alcohols and phosphoric acid (predominately mono-esters) or phosphorus pentoxide
35 (predominately di-esters), for example the reaction between lauryl alcohol and

tetraphosphoric acid; additionally these products may be ethoxylated), sulphosuccinamates, paraffin or olefine sulphonates, taurates and lignosulphonates.

Suitable SFAs of the amphoteric type include betaines, propionates and glycines.

5

Suitable SFAs of the non-ionic type include condensation products of alkylene oxides, such as ethylene oxide, propylene oxide, butylene oxide or mixtures thereof, with fatty alcohols (such as oleyl alcohol or cetyl alcohol) or with alkylphenols (such as octylphenol, nonylphenol or octylcresol); partial esters derived from long chain fatty acids or hexitol anhydrides; condensation products of said partial esters with ethylene oxide; block polymers (comprising ethylene oxide and propylene oxide); alkanolamides; simple esters (for example fatty acid polyethylene glycol esters); amine oxides (for example lauryl dimethyl amine oxide); and lecithins.

10

15

Suitable suspending agents include hydrophilic colloids (such as polysaccharides, polyvinylpyrrolidone or sodium carboxymethylcellulose) and swelling clays (such as bentonite or attapulgite).

20

A compound of formula I may be applied by any of the known means of applying pesticidal compounds. For example, it may be applied, formulated or unformulated, to the pests or to a locus of the pests (such as a habitat of the pests, or a growing plant liable to infestation by the pests) or to any part of the plant, including the foliage, stems, branches or roots, to the seed before it is planted or to other media in which plants are growing or are to be planted (such as soil surrounding the roots, the soil generally, paddy water or hydroponic culture systems), directly or it may be sprayed on, dusted on, applied by dipping, applied as a cream or paste formulation, applied as a vapour or applied through distribution or incorporation of a composition (such as a granular composition or a composition packed in a water-soluble bag) in soil or an aqueous environment.

25

30

A compound of formula I may also be injected into plants or sprayed onto vegetation using electrodynamic spraying techniques or other low volume methods, or applied by land or aerial irrigation systems.

35

Compositions for use as aqueous preparations (aqueous solutions or dispersions) are generally supplied in the form of a concentrate containing a high proportion of the active ingredient, the concentrate being added to water before use. These concentrates, which

may include DCs, SCs, ECs, EWs, MEs SGs, SPs, WPs, WGs and CSs, are often required to withstand storage for prolonged periods and, after such storage, to be capable of addition to water to form aqueous preparations which remain homogeneous for a sufficient time to enable them to be applied by conventional spray equipment. Such aqueous
5 preparations may contain varying amounts of a compound of formula I (for example 0.0001 to 10%, by weight) depending upon the purpose for which they are to be used.

A compound of formula I may be used in mixtures with fertilisers (for example nitrogen-, potassium- or phosphorus-containing fertilisers). Suitable formulation types include
10 granules of fertiliser. The mixtures suitably contain up to 25% by weight of the compound of formula I.

The invention therefore also provides a fertiliser composition comprising a fertiliser and a compound of formula I.

15

The compositions of this invention may contain other compounds having biological activity, for example micronutrients or compounds having fungicidal activity or which possess plant growth regulating, herbicidal, insecticidal, nematocidal or acaricidal activity.

20 The compound of formula I may be the sole active ingredient of the composition or it may be admixed with one or more additional active ingredients such as a pesticide (insect, acarine, mollusc and nematode pesticide), fungicide, synergist, herbicide or plant growth regulator where appropriate. An additional active ingredient may: provide a composition having a broader spectrum of activity or increased persistence at a locus; synergise the
25 activity or complement the activity (for example by increasing the speed of effect or overcoming repellency) of the compound of formula I; or help to overcome or prevent the development of resistance to individual components. The particular additional active ingredient will depend upon the intended utility of the composition. Examples of suitable pesticides include the following:

- 30 a) Pyrethroids, such as permethrin, cypermethrin, fenvalerate, esfenvalerate, deltamethrin, cyhalothrin (in particular lambda-cyhalothrin), bifenthrin, fenpropathrin, cyfluthrin, tefluthrin, fish safe pyrethroids (for example ethofenprox), natural pyrethrin, tetramethrin, s-bioallethrin, fenfluthrin, prallethrin or 5-benzyl-3-furylmethyl-(E)-(1R,3S)-2,2-dimethyl-3-(2-oxothiolan-3-ylidenemethyl)cyclopropane carboxylate;
- 35 b) Organophosphates, such as, profenofos, sulprofos, acephate, methyl parathion, azinphos-methyl, demeton-s-methyl, heptenophos, thiometon, fenamiphos, monocrotophos,

profenofos, triazophos, methamidophos, dimethoate, phosphamidon, malathion, chlorpyrifos, phosalone, terbufos, fensulfthion, fonofos, phorate, phoxim, pirimiphos-methyl, pirimiphos-ethyl, fenitrothion, fosthiazate or diazinon;

5 c) Carbamates (including aryl carbamates), such as pirimicarb, triazamate, cloethocarb, carbofuran, furathiocarb, ethiofencarb, aldicarb, thiofurox, carbosulfan, bendiocarb, fenobucarb, propoxur, methomyl or oxamyl;

d) Benzoyl ureas, such as diflubenzuron, triflumuron, hexaflumuron, flufenoxuron or chlorfluazuron;

e) Organic tin compounds, such as cyhexatin, fenbutatin oxide or azocyclotin;

10 f) Pyrazoles, such as tebufenpyrad and fenpyroximate;

g) Macrolides, such as avermectins or milbemycins, for example abamectin, emamectin benzoate, ivermectin, milbemycin, spinosad or azadirachtin;

h) Hormones or pheromones;

15 i) Organochlorine compounds such as endosulfan, benzene hexachloride, DDT, chlordane or dieldrin;

j) Amidines, such as chlordimeform or amitraz;

k) Fumigant agents, such as chloropicrin, dichloropropane, methyl bromide or metam;

l) Chloronicotinyl compounds such as imidacloprid, thiacloprid, acetamiprid, nitenpyram or thiamethoxam;

20 m) Diacylhydrazines, such as tebufenozide, chromafenozide or methoxyfenozide;

n) Diphenyl ethers, such as diofenolan or pyriproxifen;

o) Indoxacarb;

p) Chlorfenapyr; or

q) Pymetrozine.

25

In addition to the major chemical classes of pesticide listed above, other pesticides having particular targets may be employed in the composition, if appropriate for the intended utility of the composition. For instance, selective insecticides for particular crops, for example stemborer specific insecticides (such as cartap) or hopper specific insecticides (such as buprofezin) for use in rice may be employed. Alternatively insecticides or acaricides specific for particular insect species/stages may also be included in the compositions (for example acaricidal ovo-larvicides, such as clofentezine, flubenzimine, hexythiazox or tetradifon; acaricidal motilicides, such as dicofol or propargite; acaricides, such as bromopropylate or chlorobenzilate; or growth regulators, such as hydramethylnon, cyromazine, methoprene, chlorfluazuron or diflubenzuron).

30

35

The following mixtures of the compounds of formula I with active ingredients are preferred, wherein, preferably, the term "COMPOUND OF FORMULA I" refers to a compound selected from the following Tables 1 to 22:

an adjuvant selected from the group of substances consisting of petroleum oils

5 (alternative name) (628) + COMPOUND OF FORMULA I,

an acaricide selected from the group of substances consisting of 1,1-bis(4-chloro-phenyl)-2-ethoxyethanol (IUPAC name) (910) + COMPOUND OF FORMULA I, 2,4-dichlorophenyl benzenesulfonate (IUPAC/Chemical Abstracts name) (1059) + COMPOUND OF FORMULA I, 2-fluoro-*N*-methyl-*N*-1-naphthylacetamide (IUPAC name) (1295) +
10 COMPOUND OF FORMULA I, 4-chlorophenyl phenyl sulfone (IUPAC name) (981) + COMPOUND OF FORMULA I, abamectin (1) + COMPOUND OF FORMULA I, acequinocyl (3) + COMPOUND OF FORMULA I, acetoprole [CCN] + COMPOUND OF FORMULA I, acrinathrin (9) + COMPOUND OF FORMULA I, aldicarb (16) + COMPOUND OF FORMULA I, aldoxycarb (863) + COMPOUND OF FORMULA I, alpha-
15 cypermethrin (202) + COMPOUND OF FORMULA I, amidithion (870) + COMPOUND OF FORMULA I, amidoflumet [CCN] + COMPOUND OF FORMULA I, amidothioate (872) + COMPOUND OF FORMULA I, amiton (875) + COMPOUND OF FORMULA I, amiton hydrogen oxalate (875) + COMPOUND OF FORMULA I, amitraz (24) + COMPOUND OF FORMULA I, aramite (881) + COMPOUND OF FORMULA I, arsenous oxide (882) +
20 COMPOUND OF FORMULA I, AVI 382 (compound code) + COMPOUND OF FORMULA I, AZ 60541 (compound code) + COMPOUND OF FORMULA I, azinphos-ethyl (44) + COMPOUND OF FORMULA I, azinphos-methyl (45) + COMPOUND OF FORMULA I, azobenzene (IUPAC name) (888) + COMPOUND OF FORMULA I, azocyclotin (46) + COMPOUND OF FORMULA I, azothoate (889) + COMPOUND OF FORMULA I,
25 benomyl (62) + COMPOUND OF FORMULA I, benoxafos (alternative name) [CCN] + COMPOUND OF FORMULA I, benzoximate (71) + COMPOUND OF FORMULA I, benzyl benzoate (IUPAC name) [CCN] + COMPOUND OF FORMULA I, bifenazate (74) + COMPOUND OF FORMULA I, bifenthrin (76) + COMPOUND OF FORMULA I, binapacryl (907) + COMPOUND OF FORMULA I, brofenvalerate (alternative name) +
30 COMPOUND OF FORMULA I, bromocyclen (918) + COMPOUND OF FORMULA I, bromophos (920) + COMPOUND OF FORMULA I, bromophos-ethyl (921) + COMPOUND OF FORMULA I, bromopropylate (94) + COMPOUND OF FORMULA I, buprofezin (99) + COMPOUND OF FORMULA I, butocarboxim (103) + COMPOUND OF FORMULA I, butoxycarboxim (104) + COMPOUND OF FORMULA I, butylpyridaben (alternative name)
35 + COMPOUND OF FORMULA I, calcium polysulfide (IUPAC name) (111) + COMPOUND OF FORMULA I, camphechlor (941) + COMPOUND OF FORMULA I, carbanolate (943)

+ COMPOUND OF FORMULA I, carbaryl (115) + COMPOUND OF FORMULA I, carbofuran (118) + COMPOUND OF FORMULA I, carbophenothion (947) + COMPOUND OF FORMULA I, CGA 50'439 (development code) (125) + COMPOUND OF FORMULA I, chinomethionat (126) + COMPOUND OF FORMULA I, chlordbenside (959) + COMPOUND OF FORMULA I, chlordimeform (964) + COMPOUND OF FORMULA I, chlordimeform hydrochloride (964) + COMPOUND OF FORMULA I, chlorfenapyr (130) + COMPOUND OF FORMULA I, chlorfenethol (968) + COMPOUND OF FORMULA I, chlorfenson (970) + COMPOUND OF FORMULA I, chlorfensulphide (971) + COMPOUND OF FORMULA I, chlorfenvinphos (131) + COMPOUND OF FORMULA I, chlorobenzilate (975) + COMPOUND OF FORMULA I, chloromebuform (977) + COMPOUND OF FORMULA I, chloromethiuron (978) + COMPOUND OF FORMULA I, chloropropylate (983) + COMPOUND OF FORMULA I, chlorpyrifos (145) + COMPOUND OF FORMULA I, chlorpyrifos-methyl (146) + COMPOUND OF FORMULA I, chlorthiophos (994) + COMPOUND OF FORMULA I, cinerin I (696) + COMPOUND OF FORMULA I, cinerin II (696) + COMPOUND OF FORMULA I, cinerins (696) + COMPOUND OF FORMULA I, clofentezine (158) + COMPOUND OF FORMULA I, closantel (alternative name) [CCN] + COMPOUND OF FORMULA I, coumaphos (174) + COMPOUND OF FORMULA I, crotamiton (alternative name) [CCN] + COMPOUND OF FORMULA I, crotoxyphos (1010) + COMPOUND OF FORMULA I, cufraneb (1013) + COMPOUND OF FORMULA I, cyanthoate (1020) + COMPOUND OF FORMULA I, cyflumetofen (CAS Reg. No.: 400882-07-7) + COMPOUND OF FORMULA I, cyhalothrin (196) + COMPOUND OF FORMULA I, cyhexatin (199) + COMPOUND OF FORMULA I, cypermethrin (201) + COMPOUND OF FORMULA I, DCPM (1032) + COMPOUND OF FORMULA I, DDT (219) + COMPOUND OF FORMULA I, demephion (1037) + COMPOUND OF FORMULA I, demephion-O (1037) + COMPOUND OF FORMULA I, demephion-S (1037) + COMPOUND OF FORMULA I, demeton (1038) + COMPOUND OF FORMULA I, demeton-methyl (224) + COMPOUND OF FORMULA I, demeton-O (1038) + COMPOUND OF FORMULA I, demeton-O-methyl (224) + COMPOUND OF FORMULA I, demeton-S (1038) + COMPOUND OF FORMULA I, demeton-S-methyl (224) + COMPOUND OF FORMULA I, demeton-S-methylsulphon (1039) + COMPOUND OF FORMULA I, diafenthion (226) + COMPOUND OF FORMULA I, dialifos (1042) + COMPOUND OF FORMULA I, diazinon (227) + COMPOUND OF FORMULA I, dichlofluanid (230) + COMPOUND OF FORMULA I, dichlorvos (236) + COMPOUND OF FORMULA I, dicliphos (alternative name) + COMPOUND OF FORMULA I, dicofol (242) + COMPOUND OF FORMULA I, dicrotophos (243) + COMPOUND OF FORMULA I, dienochlor (1071) + COMPOUND OF FORMULA I, dimefox (1081) + COMPOUND OF FORMULA I, dimethoate (262) +

COMPOUND OF FORMULA I, dinactin (alternative name) (653) + COMPOUND OF
FORMULA I, dinex (1089) + COMPOUND OF FORMULA I, dinex-diclexine (1089) +
COMPOUND OF FORMULA I, dinobuton (269) + COMPOUND OF FORMULA I, dinocap
(270) + COMPOUND OF FORMULA I, dinocap-4 [CCN] + COMPOUND OF FORMULA I,
5 dinocap-6 [CCN] + COMPOUND OF FORMULA I, dinocton (1090) + COMPOUND OF
FORMULA I, dinopenton (1092) + COMPOUND OF FORMULA I, dinosulfon (1097) +
COMPOUND OF FORMULA I, dinoterbon (1098) + COMPOUND OF FORMULA I,
dioxathion (1102) + COMPOUND OF FORMULA I, diphenyl sulfone (IUPAC name) (1103)
+ COMPOUND OF FORMULA I, disulfiram (alternative name) [CCN] + COMPOUND OF
10 FORMULA I, disulfoton (278) + COMPOUND OF FORMULA I, DNOC (282) +
COMPOUND OF FORMULA I, dofenapyn (1113) + COMPOUND OF FORMULA I,
doramectin (alternative name) [CCN] + COMPOUND OF FORMULA I, endosulfan (294) +
COMPOUND OF FORMULA I, endothion (1121) + COMPOUND OF FORMULA I, EPN
(297) + COMPOUND OF FORMULA I, eprinomectin (alternative name) [CCN] +
15 COMPOUND OF FORMULA I, ethion (309) + COMPOUND OF FORMULA I, ethoate-
methyl (1134) + COMPOUND OF FORMULA I, etoxazole (320) + COMPOUND OF
FORMULA I, etrimfos (1142) + COMPOUND OF FORMULA I, fenazaflor (1147) +
COMPOUND OF FORMULA I, fenazaquin (328) + COMPOUND OF FORMULA I,
fenbutatin oxide (330) + COMPOUND OF FORMULA I, fenothiocarb (337) + COMPOUND
20 OF FORMULA I, fenpropathrin (342) + COMPOUND OF FORMULA I, fenpyrad
(alternative name) + COMPOUND OF FORMULA I, fenpyroximate (345) + COMPOUND
OF FORMULA I, fenson (1157) + COMPOUND OF FORMULA I, fentrifanil (1161) +
COMPOUND OF FORMULA I, fenvalerate (349) + COMPOUND OF FORMULA I, fipronil
(354) + COMPOUND OF FORMULA I, fluacrypyrim (360) + COMPOUND OF FORMULA I,
25 fluazuron (1166) + COMPOUND OF FORMULA I, flubenzimine (1167) + COMPOUND OF
FORMULA I, flucycloxuron (366) + COMPOUND OF FORMULA I, flucythrinate (367) +
COMPOUND OF FORMULA I, fluenetil (1169) + COMPOUND OF FORMULA I,
flufenoxuron (370) + COMPOUND OF FORMULA I, flumethrin (372) + COMPOUND OF
FORMULA I, fluorbenside (1174) + COMPOUND OF FORMULA I, fluvalinate (1184) +
30 COMPOUND OF FORMULA I, FMC 1137 (development code) (1185) + COMPOUND OF
FORMULA I, formetanate (405) + COMPOUND OF FORMULA I, formetanate
hydrochloride (405) + COMPOUND OF FORMULA I, formothion (1192) + COMPOUND
OF FORMULA I, formparanate (1193) + COMPOUND OF FORMULA I, gamma-HCH
(430) + COMPOUND OF FORMULA I, glyodin (1205) + COMPOUND OF FORMULA I,
35 halfenprox (424) + COMPOUND OF FORMULA I, heptenophos (432) + COMPOUND OF
FORMULA I, hexadecyl cyclopropanecarboxylate (IUPAC/Chemical Abstracts name)

(1216) + COMPOUND OF FORMULA I, hexythiazox (441) + COMPOUND OF FORMULA I, iodomethane (IUPAC name) (542) + COMPOUND OF FORMULA I, isocarbophos (alternative name) (473) + COMPOUND OF FORMULA I, isopropyl O-(methoxyaminothiophosphoryl)salicylate (IUPAC name) (473) + COMPOUND OF FORMULA I, ivermectin (alternative name) [CCN] + COMPOUND OF FORMULA I, jasmolin I (696) + COMPOUND OF FORMULA I, jasmolin II (696) + COMPOUND OF FORMULA I, jodfenphos (1248) + COMPOUND OF FORMULA I, lindane (430) + COMPOUND OF FORMULA I, lufenuron (490) + COMPOUND OF FORMULA I, malathion (492) + COMPOUND OF FORMULA I, malonoben (1254) + COMPOUND OF FORMULA I, mecarbam (502) + COMPOUND OF FORMULA I, mephosfolan (1261) + COMPOUND OF FORMULA I, mesulfen (alternative name) [CCN] + COMPOUND OF FORMULA I, methacrifos (1266) + COMPOUND OF FORMULA I, methamidophos (527) + COMPOUND OF FORMULA I, methidathion (529) + COMPOUND OF FORMULA I, methiocarb (530) + COMPOUND OF FORMULA I, methomyl (531) + COMPOUND OF FORMULA I, methyl bromide (537) + COMPOUND OF FORMULA I, metolcarb (550) + COMPOUND OF FORMULA I, mevinphos (556) + COMPOUND OF FORMULA I, mexacarbate (1290) + COMPOUND OF FORMULA I, milbemectin (557) + COMPOUND OF FORMULA I, milbemycin oxime (alternative name) [CCN] + COMPOUND OF FORMULA I, mipafox (1293) + COMPOUND OF FORMULA I, monocrotophos (561) + COMPOUND OF FORMULA I, morphothion (1300) + COMPOUND OF FORMULA I, moxidectin (alternative name) [CCN] + COMPOUND OF FORMULA I, naled (567) + COMPOUND OF FORMULA I, NC-184 (compound code) + COMPOUND OF FORMULA I, NC-512 (compound code) + COMPOUND OF FORMULA I, nifluridide (1309) + COMPOUND OF FORMULA I, nikkomycins (alternative name) [CCN] + COMPOUND OF FORMULA I, nitrilacarb (1313) + COMPOUND OF FORMULA I, nitrilacarb 1:1 zinc chloride complex (1313) + COMPOUND OF FORMULA I, NNI-0101 (compound code) + COMPOUND OF FORMULA I, NNI-0250 (compound code) + COMPOUND OF FORMULA I, omethoate (594) + COMPOUND OF FORMULA I, oxamyl (602) + COMPOUND OF FORMULA I, oxydeprofos (1324) + COMPOUND OF FORMULA I, oxydisulfoton (1325) + COMPOUND OF FORMULA I, pp'-DDT (219) + COMPOUND OF FORMULA I, parathion (615) + COMPOUND OF FORMULA I, permethrin (626) + COMPOUND OF FORMULA I, petroleum oils (alternative name) (628) + COMPOUND OF FORMULA I, phenkapton (1330) + COMPOUND OF FORMULA I, phenthoate (631) + COMPOUND OF FORMULA I, phorate (636) + COMPOUND OF FORMULA I, phosalone (637) + COMPOUND OF FORMULA I, phosfolan (1338) + COMPOUND OF FORMULA I, phosmet (638) + COMPOUND OF FORMULA I, phosphamidon (639) + COMPOUND OF FORMULA I,

phoxim (642) + COMPOUND OF FORMULA I, pirimiphos-methyl (652) + COMPOUND OF FORMULA I, polychloroterpenes (traditional name) (1347) + COMPOUND OF FORMULA I, polynactins (alternative name) (653) + COMPOUND OF FORMULA I, proclonol (1350) + COMPOUND OF FORMULA I, profenofos (662) + COMPOUND OF FORMULA I,
5 promacyl (1354) + COMPOUND OF FORMULA I, propargite (671) + COMPOUND OF FORMULA I, propetamphos (673) + COMPOUND OF FORMULA I, propoxur (678) + COMPOUND OF FORMULA I, prothidathion (1360) + COMPOUND OF FORMULA I, prothoate (1362) + COMPOUND OF FORMULA I, pyrethrin I (696) + COMPOUND OF FORMULA I, pyrethrin II (696) + COMPOUND OF FORMULA I, pyrethrins (696) +
10 COMPOUND OF FORMULA I, pyridaben (699) + COMPOUND OF FORMULA I, pyridaphenthion (701) + COMPOUND OF FORMULA I, pyrimidifen (706) + COMPOUND OF FORMULA I, pyrimitate (1370) + COMPOUND OF FORMULA I, quinalphos (711) + COMPOUND OF FORMULA I, quintiofos (1381) + COMPOUND OF FORMULA I, R-1492 (development code) (1382) + COMPOUND OF FORMULA I, RA-17 (development
15 code) (1383) + COMPOUND OF FORMULA I, rotenone (722) + COMPOUND OF FORMULA I, schradan (1389) + COMPOUND OF FORMULA I, sebufos (alternative name) + COMPOUND OF FORMULA I, selamectin (alternative name) [CCN] + COMPOUND OF FORMULA I, SI-0009 (compound code) + COMPOUND OF FORMULA I, sophamide (1402) + COMPOUND OF FORMULA I, spirodiclofen (738) + COMPOUND
20 OF FORMULA I, spiromesifen (739) + COMPOUND OF FORMULA I, SSI-121 (development code) (1404) + COMPOUND OF FORMULA I, sulfiram (alternative name) [CCN] + COMPOUND OF FORMULA I, sulfluramid (750) + COMPOUND OF FORMULA I, sulfotep (753) + COMPOUND OF FORMULA I, sulfur (754) + COMPOUND OF FORMULA I, SZI-121 (development code) (757) + COMPOUND OF FORMULA I, tau-
25 fluvalinate (398) + COMPOUND OF FORMULA I, tebufenpyrad (763) + COMPOUND OF FORMULA I, TEPP (1417) + COMPOUND OF FORMULA I, terbam (alternative name) + COMPOUND OF FORMULA I, tetrachlorvinphos (777) + COMPOUND OF FORMULA I, tetradifon (786) + COMPOUND OF FORMULA I, tetranactin (alternative name) (653) + COMPOUND OF FORMULA I, tetrasul (1425) + COMPOUND OF FORMULA I, thiafenox
30 (alternative name) + COMPOUND OF FORMULA I, thiocarboxime (1431) + COMPOUND OF FORMULA I, thiofanox (800) + COMPOUND OF FORMULA I, thiometon (801) + COMPOUND OF FORMULA I, thioquinox (1436) + COMPOUND OF FORMULA I, thuringiensin (alternative name) [CCN] + COMPOUND OF FORMULA I, triamiphos (1441) + COMPOUND OF FORMULA I, triarathene (1443) + COMPOUND OF FORMULA I,
35 triazophos (820) + COMPOUND OF FORMULA I, triazuron (alternative name) + COMPOUND OF FORMULA I, trichlorfon (824) + COMPOUND OF FORMULA I,

trifenofos (1455) + COMPOUND OF FORMULA I, trinactin (alternative name) (653) + COMPOUND OF FORMULA I, vamidothion (847) + COMPOUND OF FORMULA I, vaniliprole [CCN] and YI-5302 (compound code) + COMPOUND OF FORMULA I,

an algicide selected from the group of substances consisting of bethoxazin [CCN] +
5 COMPOUND OF FORMULA I, copper dioctanoate (IUPAC name) (170) + COMPOUND
OF FORMULA I, copper sulfate (172) + COMPOUND OF FORMULA I, cybutryne [CCN]
+ COMPOUND OF FORMULA I, dichlone (1052) + COMPOUND OF FORMULA I,
dichlorophen (232) + COMPOUND OF FORMULA I, endothal (295) + COMPOUND OF
FORMULA I, fentin (347) + COMPOUND OF FORMULA I, hydrated lime [CCN] +
10 COMPOUND OF FORMULA I, nabam (566) + COMPOUND OF FORMULA I,
quinoclamine (714) + COMPOUND OF FORMULA I, quinonamid (1379) + COMPOUND
OF FORMULA I, simazine (730) + COMPOUND OF FORMULA I, triphenyltin acetate
(IUPAC name) (347) and triphenyltin hydroxide (IUPAC name) (347) + COMPOUND OF
FORMULA I,

15 an anthelmintic selected from the group of substances consisting of abamectin (1) +
COMPOUND OF FORMULA I, crufomate (1011) + COMPOUND OF FORMULA I,
doramectin (alternative name) [CCN] + COMPOUND OF FORMULA I, emamectin (291) +
COMPOUND OF FORMULA I, emamectin benzoate (291) + COMPOUND OF FORMULA
I, eprinomectin (alternative name) [CCN] + COMPOUND OF FORMULA I, ivermectin
20 (alternative name) [CCN] + COMPOUND OF FORMULA I, milbemycin oxime (alternative
name) [CCN] + COMPOUND OF FORMULA I, moxidectin (alternative name) [CCN] +
COMPOUND OF FORMULA I, piperazine [CCN] + COMPOUND OF FORMULA I,
selamectin (alternative name) [CCN] + COMPOUND OF FORMULA I, spinosad (737) and
thiophanate (1435) + COMPOUND OF FORMULA I,

25 an avicide selected from the group of substances consisting of chloralose (127) +
COMPOUND OF FORMULA I, endrin (1122) + COMPOUND OF FORMULA I, fenthion
(346) + COMPOUND OF FORMULA I, pyridin-4-amine (IUPAC name) (23) and strychnine
(745) + COMPOUND OF FORMULA I,

a bactericide selected from the group of substances consisting of 1-hydroxy-1*H*-
30 pyridine-2-thione (IUPAC name) (1222) + COMPOUND OF FORMULA I, 4-(quinoxalin-2-
ylamino)benzenesulfonamide (IUPAC name) (748) + COMPOUND OF FORMULA I, 8-
hydroxyquinoline sulfate (446) + COMPOUND OF FORMULA I, bronopol (97) +
COMPOUND OF FORMULA I, copper dioctanoate (IUPAC name) (170) + COMPOUND
OF FORMULA I, copper hydroxide (IUPAC name) (169) + COMPOUND OF FORMULA I,
35 cresol [CCN] + COMPOUND OF FORMULA I, dichlorophen (232) + COMPOUND OF
FORMULA I, dipyrithione (1105) + COMPOUND OF FORMULA I, dodicin (1112) +

COMPOUND OF FORMULA I, fenaminosulf (1144) + COMPOUND OF FORMULA I, formaldehyde (404) + COMPOUND OF FORMULA I, hydrargaphen (alternative name) [CCN] + COMPOUND OF FORMULA I, kasugamycin (483) + COMPOUND OF FORMULA I, kasugamycin hydrochloride hydrate (483) + COMPOUND OF FORMULA I, nickel bis(dimethyldithiocarbamate) (IUPAC name) (1308) + COMPOUND OF FORMULA I, nitrapyrin (580) + COMPOUND OF FORMULA I, octhilinone (590) + COMPOUND OF FORMULA I, oxolinic acid (606) + COMPOUND OF FORMULA I, oxytetracycline (611) + COMPOUND OF FORMULA I, potassium hydroxyquinoline sulfate (446) + COMPOUND OF FORMULA I, probenazole (658) + COMPOUND OF FORMULA I, streptomycin (744) + COMPOUND OF FORMULA I, streptomycin sesquisulfate (744) + COMPOUND OF FORMULA I, tecloftalam (766) + COMPOUND OF FORMULA I, and thiomersal (alternative name) [CCN] + COMPOUND OF FORMULA I,

a biological agent selected from the group of substances consisting of *Adoxophyes orana* GV (alternative name) (12) + COMPOUND OF FORMULA I, *Agrobacterium radiobacter* (alternative name) (13) + COMPOUND OF FORMULA I, *Amblyseius* spp. (alternative name) (19) + COMPOUND OF FORMULA I, *Anagrapha falcifera* NPV (alternative name) (28) + COMPOUND OF FORMULA I, *Anagrus atomus* (alternative name) (29) + COMPOUND OF FORMULA I, *Aphelinus abdominalis* (alternative name) (33) + COMPOUND OF FORMULA I, *Aphidius colemani* (alternative name) (34) + COMPOUND OF FORMULA I, *Aphidoletes aphidimyza* (alternative name) (35) + COMPOUND OF FORMULA I, *Autographa californica* NPV (alternative name) (38) + COMPOUND OF FORMULA I, *Bacillus firmus* (alternative name) (48) + COMPOUND OF FORMULA I, *Bacillus sphaericus* Neide (scientific name) (49) + COMPOUND OF FORMULA I, *Bacillus thuringiensis* Berliner (scientific name) (51) + COMPOUND OF FORMULA I, *Bacillus thuringiensis* subsp. *aizawai* (scientific name) (51) + COMPOUND OF FORMULA I, *Bacillus thuringiensis* subsp. *israelensis* (scientific name) (51) + COMPOUND OF FORMULA I, *Bacillus thuringiensis* subsp. *japonensis* (scientific name) (51) + COMPOUND OF FORMULA I, *Bacillus thuringiensis* subsp. *kurstaki* (scientific name) (51) + COMPOUND OF FORMULA I, *Bacillus thuringiensis* subsp. *tenebrionis* (scientific name) (51) + COMPOUND OF FORMULA I, *Beauveria bassiana* (alternative name) (53) + COMPOUND OF FORMULA I, *Beauveria brongniartii* (alternative name) (54) + COMPOUND OF FORMULA I, *Chrysoperla carnea* (alternative name) (151) + COMPOUND OF FORMULA I, *Cryptolaemus montrouzieri* (alternative name) (178) + COMPOUND OF FORMULA I, *Cydia pomonella* GV (alternative name) (191) + COMPOUND OF FORMULA I, *Dacnusa sibirica* (alternative name) (212) + COMPOUND OF FORMULA I, *Diglyphus isaea* (alternative name) (254) + COMPOUND OF FORMULA

- I, *Encarsia formosa* (scientific name) (293) + COMPOUND OF FORMULA I, *Eretmocerus eremicus* (alternative name) (300) + COMPOUND OF FORMULA I, *Helicoverpa zea* NPV (alternative name) (431) + COMPOUND OF FORMULA I, *Heterorhabditis bacteriophora* and *H. megidis* (alternative name) (433) + COMPOUND OF FORMULA I, *Hippodamia convergens* (alternative name) (442) + COMPOUND OF FORMULA I, *Leptomastix dactylopii* (alternative name) (488) + COMPOUND OF FORMULA I, *Macrolophus caliginosus* (alternative name) (491) + COMPOUND OF FORMULA I, *Mamestra brassicae* NPV (alternative name) (494) + COMPOUND OF FORMULA I, *Metaphycus helvolus* (alternative name) (522) + COMPOUND OF FORMULA I, *Metarhizium anisopliae* var. *anisopliae* (scientific name) (523) + COMPOUND OF FORMULA I, *Metarhizium anisopliae* var. *anisopliae* (scientific name) (523) + COMPOUND OF FORMULA I, *Neodiprion sertifer* NPV and *N. lecontei* NPV (alternative name) (575) + COMPOUND OF FORMULA I, *Orius* spp. (alternative name) (596) + COMPOUND OF FORMULA I, *Paecilomyces fumosoroseus* (alternative name) (613) + COMPOUND OF FORMULA I, *Phytoseiulus persimilis* (alternative name) (644) + COMPOUND OF FORMULA I, *Spodoptera exigua* multicapsid nuclear polyhedrosis virus (scientific name) (741) + COMPOUND OF FORMULA I, *Steinernema bibionis* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema carpocapsae* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema feltiae* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema glaseri* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema riobrave* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema riobravis* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema scapterisci* (alternative name) (742) + COMPOUND OF FORMULA I, *Steinernema* spp. (alternative name) (742) + COMPOUND OF FORMULA I, *Trichogramma* spp. (alternative name) (826) + COMPOUND OF FORMULA I, *Typhlodromus occidentalis* (alternative name) (844) and *Verticillium lecanii* (alternative name) (848) + COMPOUND OF FORMULA I,
- a soil sterilant selected from the group of substances consisting of iodomethane (IUPAC name) (542) and methyl bromide (537) + COMPOUND OF FORMULA I,
- a chemosterilant selected from the group of substances consisting of apholate [CCN] + COMPOUND OF FORMULA I, bisazir (alternative name) [CCN] + COMPOUND OF FORMULA I, busulfan (alternative name) [CCN] + COMPOUND OF FORMULA I, diflubenzuron (250) + COMPOUND OF FORMULA I, dimatif (alternative name) [CCN] + COMPOUND OF FORMULA I, hemel [CCN] + COMPOUND OF FORMULA I, hempa [CCN] + COMPOUND OF FORMULA I, metepa [CCN] + COMPOUND OF FORMULA I, methiotepa [CCN] + COMPOUND OF FORMULA I, methyl apholate [CCN] + COMPOUND

OF FORMULA I, morzid [CCN] + COMPOUND OF FORMULA I, penfluron (alternative name) [CCN] + COMPOUND OF FORMULA I, tepa [CCN] + COMPOUND OF FORMULA I, thiohempa (alternative name) [CCN] + COMPOUND OF FORMULA I, thiotepa (alternative name) [CCN] + COMPOUND OF FORMULA I, tretamine (alternative name) [CCN] and uredepa (alternative name) [CCN] + COMPOUND OF FORMULA I,

an insect pheromone selected from the group of substances consisting of (*E*)-dec-5-en-1-yl acetate with (*E*)-dec-5-en-1-ol (IUPAC name) (222) + COMPOUND OF FORMULA I, (*E*)-tridec-4-en-1-yl acetate (IUPAC name) (829) + COMPOUND OF FORMULA I, (*E*)-6-methylhept-2-en-4-ol (IUPAC name) (541) + COMPOUND OF FORMULA I, (*E,Z*)-tetradeca-4,10-dien-1-yl acetate (IUPAC name) (779) + COMPOUND OF FORMULA I, (*Z*)-dodec-7-en-1-yl acetate (IUPAC name) (285) + COMPOUND OF FORMULA I, (*Z*)-hexadec-11-enal (IUPAC name) (436) + COMPOUND OF FORMULA I, (*Z*)-hexadec-11-en-1-yl acetate (IUPAC name) (437) + COMPOUND OF FORMULA I, (*Z*)-hexadec-13-en-11-yn-1-yl acetate (IUPAC name) (438) + COMPOUND OF FORMULA I, (*Z*)-icos-13-en-10-one (IUPAC name) (448) + COMPOUND OF FORMULA I, (*Z*)-tetradec-7-en-1-al (IUPAC name) (782) + COMPOUND OF FORMULA I, (*Z*)-tetradec-9-en-1-ol (IUPAC name) (783) + COMPOUND OF FORMULA I, (*Z*)-tetradec-9-en-1-yl acetate (IUPAC name) (784) + COMPOUND OF FORMULA I, (7*E*,9*Z*)-dodeca-7,9-dien-1-yl acetate (IUPAC name) (283) + COMPOUND OF FORMULA I, (9*Z*,11*E*)-tetradeca-9,11-dien-1-yl acetate (IUPAC name) (780) + COMPOUND OF FORMULA I, (9*Z*,12*E*)-tetradeca-9,12-dien-1-yl acetate (IUPAC name) (781) + COMPOUND OF FORMULA I, 14-methyloctadec-1-ene (IUPAC name) (545) + COMPOUND OF FORMULA I, 4-methylnonan-5-ol with 4-methylnonan-5-one (IUPAC name) (544) + COMPOUND OF FORMULA I, alpha-multistriatin (alternative name) [CCN] + COMPOUND OF FORMULA I, brevicomin (alternative name) [CCN] + COMPOUND OF FORMULA I, codlure (alternative name) [CCN] + COMPOUND OF FORMULA I, codlemone (alternative name) (167) + COMPOUND OF FORMULA I, cuelure (alternative name) (179) + COMPOUND OF FORMULA I, disparlure (277) + COMPOUND OF FORMULA I, dodec-8-en-1-yl acetate (IUPAC name) (286) + COMPOUND OF FORMULA I, dodec-9-en-1-yl acetate (IUPAC name) (287) + COMPOUND OF FORMULA I, dodeca-8 + COMPOUND OF FORMULA I, 10-dien-1-yl acetate (IUPAC name) (284) + COMPOUND OF FORMULA I, dominicalure (alternative name) [CCN] + COMPOUND OF FORMULA I, ethyl 4-methyloctanoate (IUPAC name) (317) + COMPOUND OF FORMULA I, eugenol (alternative name) [CCN] + COMPOUND OF FORMULA I, frontalinal (alternative name) [CCN] + COMPOUND OF FORMULA I, gossyplure (alternative name) (420) + COMPOUND OF FORMULA I, grandlure (421) + COMPOUND OF FORMULA I, grandlure I (alternative name) (421) +

COMPOUND OF FORMULA I, grandlure II (alternative name) (421) + COMPOUND OF FORMULA I, grandlure III (alternative name) (421) + COMPOUND OF FORMULA I, grandlure IV (alternative name) (421) + COMPOUND OF FORMULA I, hexalure [CCN] + COMPOUND OF FORMULA I, ipsdienol (alternative name) [CCN] + COMPOUND OF FORMULA I, ipsenol (alternative name) [CCN] + COMPOUND OF FORMULA I, japonilure (alternative name) (481) + COMPOUND OF FORMULA I, lineatin (alternative name) [CCN] + COMPOUND OF FORMULA I, litlure (alternative name) [CCN] + COMPOUND OF FORMULA I, looplure (alternative name) [CCN] + COMPOUND OF FORMULA I, medlure [CCN] + COMPOUND OF FORMULA I, megatomoic acid (alternative name) [CCN] + COMPOUND OF FORMULA I, methyl eugenol (alternative name) (540) + COMPOUND OF FORMULA I, muscalure (563) + COMPOUND OF FORMULA I, octadeca-2,13-dien-1-yl acetate (IUPAC name) (588) + COMPOUND OF FORMULA I, octadeca-3,13-dien-1-yl acetate (IUPAC name) (589) + COMPOUND OF FORMULA I, orfralure (alternative name) [CCN] + COMPOUND OF FORMULA I, oryctalure (alternative name) (317) + COMPOUND OF FORMULA I, ostramone (alternative name) [CCN] + COMPOUND OF FORMULA I, siglure [CCN] + COMPOUND OF FORMULA I, sordidin (alternative name) (736) + COMPOUND OF FORMULA I, sulcatol (alternative name) [CCN] + COMPOUND OF FORMULA I, tetradec-11-en-1-yl acetate (IUPAC name) (785) + COMPOUND OF FORMULA I, trimedlure (839) + COMPOUND OF FORMULA I, trimedlure A (alternative name) (839) + COMPOUND OF FORMULA I, trimedlure B₁ (alternative name) (839) + COMPOUND OF FORMULA I, trimedlure B₂ (alternative name) (839) + COMPOUND OF FORMULA I, trimedlure C (alternative name) (839) and trunc-call (alternative name) [CCN] + COMPOUND OF FORMULA I,

an insect repellent selected from the group of substances consisting of 2-(octylthio)-ethanol (IUPAC name) (591) + COMPOUND OF FORMULA I, butopyronoxyl (933) + COMPOUND OF FORMULA I, butoxy(polypropylene glycol) (936) + COMPOUND OF FORMULA I, dibutyl adipate (IUPAC name) (1046) + COMPOUND OF FORMULA I, dibutyl phthalate (1047) + COMPOUND OF FORMULA I, dibutyl succinate (IUPAC name) (1048) + COMPOUND OF FORMULA I, diethyltoluamide [CCN] + COMPOUND OF FORMULA I, dimethyl carbate [CCN] + COMPOUND OF FORMULA I, dimethyl phthalate [CCN] + COMPOUND OF FORMULA I, ethyl hexanediol (1137) + COMPOUND OF FORMULA I, hexamide [CCN] + COMPOUND OF FORMULA I, methoquin-butyl (1276) + COMPOUND OF FORMULA I, methylneodecanamide [CCN] + COMPOUND OF FORMULA I, oxamate [CCN] and picaridin [CCN] + COMPOUND OF FORMULA I,

an insecticide selected from the group of substances consisting of 1-dichloro-1-nitroethane (IUPAC/Chemical Abstracts name) (1058) + COMPOUND OF FORMULA I, 1,1-dichloro-2,2-bis(4-ethylphenyl)ethane (IUPAC name) (1056), + COMPOUND OF FORMULA I, 1,2-dichloropropane (IUPAC/Chemical Abstracts name) (1062) + COMPOUND OF FORMULA I, 1,2-dichloropropane with 1,3-dichloropropene (IUPAC name) (1063) + COMPOUND OF FORMULA I, 1-bromo-2-chloroethane (IUPAC/Chemical Abstracts name) (916) + COMPOUND OF FORMULA I, 2,2,2-trichloro-1-(3,4-dichlorophenyl)ethyl acetate (IUPAC name) (1451) + COMPOUND OF FORMULA I, 2,2-dichlorovinyl 2-ethylsulfinyethyl methyl phosphate (IUPAC name) (1066) + COMPOUND OF FORMULA I, 2-(1,3-dithiolan-2-yl)phenyl dimethylcarbamate (IUPAC/ Chemical Abstracts name) (1109) + COMPOUND OF FORMULA I, 2-(2-butoxyethoxy)ethyl thiocyanate (IUPAC/Chemical Abstracts name) (935) + COMPOUND OF FORMULA I, 2-(4,5-dimethyl-1,3-dioxolan-2-yl)phenyl methylcarbamate (IUPAC/ Chemical Abstracts name) (1084) + COMPOUND OF FORMULA I, 2-(4-chloro-3,5-xylyloxy)ethanol (IUPAC name) (986) + COMPOUND OF FORMULA I, 2-chlorovinyl diethyl phosphate (IUPAC name) (984) + COMPOUND OF FORMULA I, 2-imidazolidone (IUPAC name) (1225) + COMPOUND OF FORMULA I, 2-isovalerylindan-1,3-dione (IUPAC name) (1246) + COMPOUND OF FORMULA I, 2-methyl(prop-2-ynyl)aminophenyl methylcarbamate (IUPAC name) (1284) + COMPOUND OF FORMULA I, 2-thiocyanatoethyl laurate (IUPAC name) (1433) + COMPOUND OF FORMULA I, 3-bromo-1-chloroprop-1-ene (IUPAC name) (917) + COMPOUND OF FORMULA I, 3-methyl-1-phenylpyrazol-5-yl dimethylcarbamate (IUPAC name) (1283) + COMPOUND OF FORMULA I, 4-methyl(prop-2-ynyl)amino-3,5-xylyl methylcarbamate (IUPAC name) (1285) + COMPOUND OF FORMULA I, 5,5-dimethyl-3-oxocyclohex-1-enyl dimethylcarbamate (IUPAC name) (1085) + COMPOUND OF FORMULA I, abamectin (1) + COMPOUND OF FORMULA I, acephate (2) + COMPOUND OF FORMULA I, acetamiprid (4) + COMPOUND OF FORMULA I, acethion (alternative name) [CCN] + COMPOUND OF FORMULA I, acetoprole [CCN] + COMPOUND OF FORMULA I, acrinathrin (9) + COMPOUND OF FORMULA I, acrylonitrile (IUPAC name) (861) + COMPOUND OF FORMULA I, alanycarb (15) + COMPOUND OF FORMULA I, aldicarb (16) + COMPOUND OF FORMULA I, aldoxycarb (863) + COMPOUND OF FORMULA I, aldrin (864) + COMPOUND OF FORMULA I, allethrin (17) + COMPOUND OF FORMULA I, allosamidin (alternative name) [CCN] + COMPOUND OF FORMULA I, allyxycarb (866) + COMPOUND OF FORMULA I, alpha-cypermethrin (202) + COMPOUND OF FORMULA I, alpha-ecdysone (alternative name) [CCN] + COMPOUND OF FORMULA I, aluminium phosphide (640) + COMPOUND OF FORMULA I, amidithion (870) + COMPOUND OF FORMULA I, amidothioate (872) + COMPOUND OF FORMULA I, aminocarb (873) +

COMPOUND OF FORMULA I, amiton (875) + COMPOUND OF FORMULA I, amiton
hydrogen oxalate (875) + COMPOUND OF FORMULA I, amitraz (24) + COMPOUND OF
FORMULA I, anabasine (877) + COMPOUND OF FORMULA I, athidathion (883) +
COMPOUND OF FORMULA I, AVI 382 (compound code) + COMPOUND OF FORMULA
5 I, AZ 60541 (compound code) + COMPOUND OF FORMULA I, azadirachtin (alternative
name) (41) + COMPOUND OF FORMULA I, azamethiphos (42) + COMPOUND OF
FORMULA I, azinphos-ethyl (44) + COMPOUND OF FORMULA I, azinphos-methyl (45)
+ COMPOUND OF FORMULA I, azothoate (889) + COMPOUND OF FORMULA I,
Bacillus thuringiensis delta endotoxins (alternative name) (52) + COMPOUND OF
10 FORMULA I, barium hexafluorosilicate (alternative name) [CCN] + COMPOUND OF
FORMULA I, barium polysulfide (IUPAC/Chemical Abstracts name) (892) + COMPOUND
OF FORMULA I, barthrin [CCN] + COMPOUND OF FORMULA I, Bayer 22/190
(development code) (893) + COMPOUND OF FORMULA I, Bayer 22408 (development
code) (894) + COMPOUND OF FORMULA I, bendiocarb (58) + COMPOUND OF
15 FORMULA I, benfuracarb (60) + COMPOUND OF FORMULA I, bensultap (66) +
COMPOUND OF FORMULA I, beta-cyfluthrin (194) + COMPOUND OF FORMULA I,
beta-cypermethrin (203) + COMPOUND OF FORMULA I, bifenthrin (76) + COMPOUND
OF FORMULA I, bioallethrin (78) + COMPOUND OF FORMULA I, bioallethrin S-
cyclopentenyl isomer (alternative name) (79) + COMPOUND OF FORMULA I,
20 bioethanomethrin [CCN] + COMPOUND OF FORMULA I, biopermethrin (908) +
COMPOUND OF FORMULA I, bioresmethrin (80) + COMPOUND OF FORMULA I, bis(2-
chloroethyl) ether (IUPAC name) (909) + COMPOUND OF FORMULA I, bistrifluron (83) +
COMPOUND OF FORMULA I, borax (86) + COMPOUND OF FORMULA I,
brofenvalerate (alternative name) + COMPOUND OF FORMULA I, bromfenvinfos (914) +
25 COMPOUND OF FORMULA I, bromocyclen (918) + COMPOUND OF FORMULA I,
bromo-DDT (alternative name) [CCN] + COMPOUND OF FORMULA I, bromophos (920) +
COMPOUND OF FORMULA I, bromophos-ethyl (921) + COMPOUND OF FORMULA I,
bufencarb (924) + COMPOUND OF FORMULA I, buprofezin (99) + COMPOUND OF
FORMULA I, butacarb (926) + COMPOUND OF FORMULA I, butathiofos (927) +
30 COMPOUND OF FORMULA I, butocarboxim (103) + COMPOUND OF FORMULA I,
butonate (932) + COMPOUND OF FORMULA I, butoxycarboxim (104) + COMPOUND OF
FORMULA I, butylpyridaben (alternative name) + COMPOUND OF FORMULA I,
cadusafos (109) + COMPOUND OF FORMULA I, calcium arsenate [CCN] + COMPOUND
OF FORMULA I, calcium cyanide (444) + COMPOUND OF FORMULA I, calcium
35 polysulfide (IUPAC name) (111) + COMPOUND OF FORMULA I, camphechlor (941) +
COMPOUND OF FORMULA I, carbanolate (943) + COMPOUND OF FORMULA I,

carbaryl (115) + COMPOUND OF FORMULA I, carbofuran (118) + COMPOUND OF FORMULA I, carbon disulfide (IUPAC/Chemical Abstracts name) (945) + COMPOUND OF FORMULA I, carbon tetrachloride (IUPAC name) (946) + COMPOUND OF FORMULA I, carbophenothion (947) + COMPOUND OF FORMULA I, carbosulfan (119) + COMPOUND OF FORMULA I, cartap (123) + COMPOUND OF FORMULA I, cartap hydrochloride (123) + COMPOUND OF FORMULA I, cevadine (alternative name) (725) + COMPOUND OF FORMULA I, chlorbicyclen (960) + COMPOUND OF FORMULA I, chlordane (128) + COMPOUND OF FORMULA I, chlordecone (963) + COMPOUND OF FORMULA I, chlordimeform (964) + COMPOUND OF FORMULA I, chlordimeform hydrochloride (964) + COMPOUND OF FORMULA I, chlorethoxyfos (129) + COMPOUND OF FORMULA I, chlorfenapyr (130) + COMPOUND OF FORMULA I, chlorfenvinphos (131) + COMPOUND OF FORMULA I, chlorfluazuron (132) + COMPOUND OF FORMULA I, chlormephos (136) + COMPOUND OF FORMULA I, chloroform [CCN] + COMPOUND OF FORMULA I, chloropicrin (141) + COMPOUND OF FORMULA I, chlorphoxim (989) + COMPOUND OF FORMULA I, chlorprazophos (990) + COMPOUND OF FORMULA I, chlorpyrifos (145) + COMPOUND OF FORMULA I, chlorpyrifos-methyl (146) + COMPOUND OF FORMULA I, chlorthiophos (994) + COMPOUND OF FORMULA I, chromafenozide (150) + COMPOUND OF FORMULA I, cinerin I (696) + COMPOUND OF FORMULA I, cinerin II (696) + COMPOUND OF FORMULA I, cinerins (696) + COMPOUND OF FORMULA I, cis-resmethrin (alternative name) + COMPOUND OF FORMULA I, cismethrin (80) + COMPOUND OF FORMULA I, clocythrin (alternative name) + COMPOUND OF FORMULA I, cloethocarb (999) + COMPOUND OF FORMULA I, closantel (alternative name) [CCN] + COMPOUND OF FORMULA I, clothianidin (165) + COMPOUND OF FORMULA I, copper acetoarsenite [CCN] + COMPOUND OF FORMULA I, copper arsenate [CCN] + COMPOUND OF FORMULA I, copper oleate [CCN] + COMPOUND OF FORMULA I, coumaphos (174) + COMPOUND OF FORMULA I, coumithoate (1006) + COMPOUND OF FORMULA I, crotamiton (alternative name) [CCN] + COMPOUND OF FORMULA I, crotoxyphos (1010) + COMPOUND OF FORMULA I, cruformate (1011) + COMPOUND OF FORMULA I, cryolite (alternative name) (177) + COMPOUND OF FORMULA I, CS 708 (development code) (1012) + COMPOUND OF FORMULA I, cyanofenphos (1019) + COMPOUND OF FORMULA I, cyanophos (184) + COMPOUND OF FORMULA I, cyanthoate (1020) + COMPOUND OF FORMULA I, cyclethrin [CCN] + COMPOUND OF FORMULA I, cycloprothrin (188) + COMPOUND OF FORMULA I, cyfluthrin (193) + COMPOUND OF FORMULA I, cyhalothrin (196) + COMPOUND OF FORMULA I, cypermethrin (201) + COMPOUND OF FORMULA I, cyphenothrin (206) + COMPOUND OF FORMULA I, cyromazine (209) + COMPOUND OF FORMULA I,

cythioate (alternative name) [CCN] + COMPOUND OF FORMULA I, *d*-limonene (alternative name) [CCN] + COMPOUND OF FORMULA I, *d*-tetramethrin (alternative name) (788) + COMPOUND OF FORMULA I, DAEP (1031) + COMPOUND OF FORMULA I, dazomet (216) + COMPOUND OF FORMULA I, DDT (219) + COMPOUND OF FORMULA I, decarbofuran (1034) + COMPOUND OF FORMULA I, deltamethrin (223) + COMPOUND OF FORMULA I, demephion (1037) + COMPOUND OF FORMULA I, demephion-O (1037) + COMPOUND OF FORMULA I, demephion-S (1037) + COMPOUND OF FORMULA I, demeton (1038) + COMPOUND OF FORMULA I, demeton-methyl (224) + COMPOUND OF FORMULA I, demeton-O (1038) + COMPOUND OF FORMULA I, demeton-O-methyl (224) + COMPOUND OF FORMULA I, demeton-S (1038) + COMPOUND OF FORMULA I, demeton-S-methyl (224) + COMPOUND OF FORMULA I, demeton-S-methylsulphon (1039) + COMPOUND OF FORMULA I, diafenthion (226) + COMPOUND OF FORMULA I, dialifos (1042) + COMPOUND OF FORMULA I, diamidafos (1044) + COMPOUND OF FORMULA I, diazinon (227) + COMPOUND OF FORMULA I, dicapthon (1050) + COMPOUND OF FORMULA I, dichlofenthion (1051) + COMPOUND OF FORMULA I, dichlorvos (236) + COMPOUND OF FORMULA I, dicliphos (alternative name) + COMPOUND OF FORMULA I, dicresyl (alternative name) [CCN] + COMPOUND OF FORMULA I, dicrotophos (243) + COMPOUND OF FORMULA I, dicyclanil (244) + COMPOUND OF FORMULA I, dieldrin (1070) + COMPOUND OF FORMULA I, diethyl 5-methylpyrazol-3-yl phosphate (IUPAC name) (1076) + COMPOUND OF FORMULA I, diflubenzuron (250) + COMPOUND OF FORMULA I, dilor (alternative name) [CCN] + COMPOUND OF FORMULA I, dimefluthrin [CCN] + COMPOUND OF FORMULA I, dimefox (1081) + COMPOUND OF FORMULA I, dimetan (1085) + COMPOUND OF FORMULA I, dimethoate (262) + COMPOUND OF FORMULA I, dimethrin (1083) + COMPOUND OF FORMULA I, dimethylvinphos (265) + COMPOUND OF FORMULA I, dimetilan (1086) + COMPOUND OF FORMULA I, dinex (1089) + COMPOUND OF FORMULA I, dinex-diclexine (1089) + COMPOUND OF FORMULA I, dinoprop (1093) + COMPOUND OF FORMULA I, dinosam (1094) + COMPOUND OF FORMULA I, dinoseb (1095) + COMPOUND OF FORMULA I, dinotefuran (271) + COMPOUND OF FORMULA I, diofenolan (1099) + COMPOUND OF FORMULA I, dioxabenzofos (1100) + COMPOUND OF FORMULA I, dioxacarb (1101) + COMPOUND OF FORMULA I, dioxathion (1102) + COMPOUND OF FORMULA I, disulfoton (278) + COMPOUND OF FORMULA I, dithicrofos (1108) + COMPOUND OF FORMULA I, DNOC (282) + COMPOUND OF FORMULA I, doramectin (alternative name) [CCN] + COMPOUND OF FORMULA I, DSP (1115) + COMPOUND OF FORMULA I, ecdysterone (alternative name) [CCN] + COMPOUND OF FORMULA I, EI 1642

(development code) (1118) + COMPOUND OF FORMULA I, emamectin (291) + COMPOUND OF FORMULA I, emamectin benzoate (291) + COMPOUND OF FORMULA I, EMPC (1120) + COMPOUND OF FORMULA I, empenthrin (292) + COMPOUND OF FORMULA I, endosulfan (294) + COMPOUND OF FORMULA I, endothion (1121) +
5 COMPOUND OF FORMULA I, endrin (1122) + COMPOUND OF FORMULA I, EPBP (1123) + COMPOUND OF FORMULA I, EPN (297) + COMPOUND OF FORMULA I, epofenonane (1124) + COMPOUND OF FORMULA I, eprinomectin (alternative name) [CCN] + COMPOUND OF FORMULA I, esfenvaterate (302) + COMPOUND OF FORMULA I, etaphos (alternative name) [CCN] + COMPOUND OF FORMULA I,
10 ethiofencarb (308) + COMPOUND OF FORMULA I, ethion (309) + COMPOUND OF FORMULA I, ethiprole (310) + COMPOUND OF FORMULA I, ethoate-methyl (1134) + COMPOUND OF FORMULA I, ethoprophos (312) + COMPOUND OF FORMULA I, ethyl formate (IUPAC name) [CCN] + COMPOUND OF FORMULA I, ethyl-DDD (alternative name) (1056) + COMPOUND OF FORMULA I, ethylene dibromide (316) + COMPOUND
15 OF FORMULA I, ethylene dichloride (chemical name) (1136) + COMPOUND OF FORMULA I, ethylene oxide [CCN] + COMPOUND OF FORMULA I, etofenprox (319) + COMPOUND OF FORMULA I, etrimfos (1142) + COMPOUND OF FORMULA I, EXD (1143) + COMPOUND OF FORMULA I, famphur (323) + COMPOUND OF FORMULA I, fenamiphos (326) + COMPOUND OF FORMULA I, fenazaflor (1147) + COMPOUND OF
20 FORMULA I, fenchlorphos (1148) + COMPOUND OF FORMULA I, fenethacarb (1149) + COMPOUND OF FORMULA I, fenfluthrin (1150) + COMPOUND OF FORMULA I, fenitrothion (335) + COMPOUND OF FORMULA I, fenobucarb (336) + COMPOUND OF FORMULA I, fenoxacrim (1153) + COMPOUND OF FORMULA I, fenoxycarb (340) + COMPOUND OF FORMULA I, fenpirithrin (1155) + COMPOUND OF FORMULA I,
25 fenpropathrin (342) + COMPOUND OF FORMULA I, fenpyrad (alternative name) + COMPOUND OF FORMULA I, fensulfothion (1158) + COMPOUND OF FORMULA I, fenthion (346) + COMPOUND OF FORMULA I, fenthion-ethyl [CCN] + COMPOUND OF FORMULA I, fenvalerate (349) + COMPOUND OF FORMULA I, fipronil (354) + COMPOUND OF FORMULA I, flonicamid (358) + COMPOUND OF FORMULA I,
30 flubendiamide (CAS. Reg. No.: 272451-65-7) + COMPOUND OF FORMULA I, flucofuron (1168) + COMPOUND OF FORMULA I, flucycloxuron (366) + COMPOUND OF FORMULA I, flucythrinate (367) + COMPOUND OF FORMULA I, fluenetil (1169) + COMPOUND OF FORMULA I, flufenerim [CCN] + COMPOUND OF FORMULA I, flufenoxuron (370) + COMPOUND OF FORMULA I, flufenprox (1171) + COMPOUND OF
35 FORMULA I, flumethrin (372) + COMPOUND OF FORMULA I, fluvalinate (1184) + COMPOUND OF FORMULA I, FMC 1137 (development code) (1185) + COMPOUND OF

FORMULA I, fonofos (1191) + COMPOUND OF FORMULA I, formetanate (405) +
COMPOUND OF FORMULA I, formetanate hydrochloride (405) + COMPOUND OF
FORMULA I, formothion (1192) + COMPOUND OF FORMULA I, formparanate (1193) +
COMPOUND OF FORMULA I, fosmethilan (1194) + COMPOUND OF FORMULA I,
5 fospirate (1195) + COMPOUND OF FORMULA I, fosthiazate (408) + COMPOUND OF
FORMULA I, fosthietan (1196) + COMPOUND OF FORMULA I, furathiocarb (412) +
COMPOUND OF FORMULA I, furethrin (1200) + COMPOUND OF FORMULA I, gamma-
cyhalothrin (197) + COMPOUND OF FORMULA I, gamma-HCH (430) + COMPOUND OF
FORMULA I, guazatine (422) + COMPOUND OF FORMULA I, guazatine acetates (422)
10 + COMPOUND OF FORMULA I, GY-81 (development code) (423) + COMPOUND OF
FORMULA I, halfenprox (424) + COMPOUND OF FORMULA I, halofenozide (425) +
COMPOUND OF FORMULA I, HCH (430) + COMPOUND OF FORMULA I, HEOD
(1070) + COMPOUND OF FORMULA I, heptachlor (1211) + COMPOUND OF FORMULA
I, heptenophos (432) + COMPOUND OF FORMULA I, heterophos [CCN] + COMPOUND
15 OF FORMULA I, hexaflumuron (439) + COMPOUND OF FORMULA I, HHDN (864) +
COMPOUND OF FORMULA I, hydramethylnon (443) + COMPOUND OF FORMULA I,
hydrogen cyanide (444) + COMPOUND OF FORMULA I, hydroprene (445) +
COMPOUND OF FORMULA I, hyquincarb (1223) + COMPOUND OF FORMULA I,
imidacloprid (458) + COMPOUND OF FORMULA I, imiprothrin (460) + COMPOUND OF
20 FORMULA I, indoxacarb (465) + COMPOUND OF FORMULA I, iodomethane (IUPAC
name) (542) + COMPOUND OF FORMULA I, IPSP (1229) + COMPOUND OF FORMULA
I, isazofos (1231) + COMPOUND OF FORMULA I, isobenzan (1232) + COMPOUND OF
FORMULA I, isocarbophos (alternative name) (473) + COMPOUND OF FORMULA I,
isodrin (1235) + COMPOUND OF FORMULA I, isofenphos (1236) + COMPOUND OF
25 FORMULA I, isolane (1237) + COMPOUND OF FORMULA I, isoprocab (472) +
COMPOUND OF FORMULA I, isopropyl O-(methoxyaminothiophosphoryl)salicylate
(IUPAC name) (473) + COMPOUND OF FORMULA I, isoprothiolane (474) + COMPOUND
OF FORMULA I, isothioate (1244) + COMPOUND OF FORMULA I, isoxathion (480) +
COMPOUND OF FORMULA I, ivermectin (alternative name) [CCN] + COMPOUND OF
30 FORMULA I, jasmolin I (696) + COMPOUND OF FORMULA I, jasmolin II (696) +
COMPOUND OF FORMULA I, jodfenphos (1248) + COMPOUND OF FORMULA I,
juvenile hormone I (alternative name) [CCN] + COMPOUND OF FORMULA I, juvenile
hormone II (alternative name) [CCN] + COMPOUND OF FORMULA I, juvenile hormone III
(alternative name) [CCN] + COMPOUND OF FORMULA I, kelevan (1249) + COMPOUND
35 OF FORMULA I, kinoprene (484) + COMPOUND OF FORMULA I, lambda-cyhalothrin
(198) + COMPOUND OF FORMULA I, lead arsenate [CCN] + COMPOUND OF

FORMULA I, lepimectin (CCN) + COMPOUND OF FORMULA I, leptophos (1250) +
COMPOUND OF FORMULA I, lindane (430) + COMPOUND OF FORMULA I, lirimfos
(1251) + COMPOUND OF FORMULA I, lufenuron (490) + COMPOUND OF FORMULA I,
lythidathion (1253) + COMPOUND OF FORMULA I, *m*-cumenyl methylcarbamate (IUPAC
5 name) (1014) + COMPOUND OF FORMULA I, magnesium phosphide (IUPAC name)
(640) + COMPOUND OF FORMULA I, malathion (492) + COMPOUND OF FORMULA I,
malonoben (1254) + COMPOUND OF FORMULA I, mazidox (1255) + COMPOUND OF
FORMULA I, mecarbam (502) + COMPOUND OF FORMULA I, mecarphon (1258) +
COMPOUND OF FORMULA I, menazon (1260) + COMPOUND OF FORMULA I,
10 mephosfolan (1261) + COMPOUND OF FORMULA I, mercurous chloride (513) +
COMPOUND OF FORMULA I, mesulfenfos (1263) + COMPOUND OF FORMULA I,
metaflumizone (CCN) + COMPOUND OF FORMULA I, metam (519) + COMPOUND OF
FORMULA I, metam-potassium (alternative name) (519) + COMPOUND OF FORMULA I,
metam-sodium (519) + COMPOUND OF FORMULA I, methacrifos (1266) + COMPOUND
15 OF FORMULA I, methamidophos (527) + COMPOUND OF FORMULA I,
methanesulfonyl fluoride (IUPAC/Chemical Abstracts name) (1268) + COMPOUND OF
FORMULA I, methidathion (529) + COMPOUND OF FORMULA I, methiocarb (530) +
COMPOUND OF FORMULA I, methocrotophos (1273) + COMPOUND OF FORMULA I,
methomyl (531) + COMPOUND OF FORMULA I, methoprene (532) + COMPOUND OF
20 FORMULA I, methoquin-butyl (1276) + COMPOUND OF FORMULA I, methothrin
(alternative name) (533) + COMPOUND OF FORMULA I, methoxychlor (534) +
COMPOUND OF FORMULA I, methoxyfenozide (535) + COMPOUND OF FORMULA I,
methyl bromide (537) + COMPOUND OF FORMULA I, methyl isothiocyanate (543) +
COMPOUND OF FORMULA I, methylchloroform (alternative name) [CCN] + COMPOUND
25 OF FORMULA I, methylene chloride [CCN] + COMPOUND OF FORMULA I, metofluthrin
[CCN] + COMPOUND OF FORMULA I, metolcarb (550) + COMPOUND OF FORMULA I,
metoxadiazone (1288) + COMPOUND OF FORMULA I, mevinphos (556) + COMPOUND
OF FORMULA I, mexacarbate (1290) + COMPOUND OF FORMULA I, milbemectin
(557) + COMPOUND OF FORMULA I, milbemycin oxime (alternative name) [CCN] +
30 COMPOUND OF FORMULA I, mipafox (1293) + COMPOUND OF FORMULA I, mirex
(1294) + COMPOUND OF FORMULA I, monocrotophos (561) + COMPOUND OF
FORMULA I, morphothion (1300) + COMPOUND OF FORMULA I, moxidectin
(alternative name) [CCN] + COMPOUND OF FORMULA I, naftalofos (alternative name)
[CCN] + COMPOUND OF FORMULA I, naled (567) + COMPOUND OF FORMULA I,
35 naphthalene (IUPAC/Chemical Abstracts name) (1303) + COMPOUND OF FORMULA I,
NC-170 (development code) (1306) + COMPOUND OF FORMULA I, NC-184 (compound

code) + COMPOUND OF FORMULA I, nicotine (578) + COMPOUND OF FORMULA I, nicotine sulfate (578) + COMPOUND OF FORMULA I, nifluridide (1309) + COMPOUND OF FORMULA I, nitenpyram (579) + COMPOUND OF FORMULA I, nithiazine (1311) + COMPOUND OF FORMULA I, nitrilacarb (1313) + COMPOUND OF FORMULA I,
5 nitrilacarb 1:1 zinc chloride complex (1313) + COMPOUND OF FORMULA I, NNI-0101 (compound code) + COMPOUND OF FORMULA I, NNI-0250 (compound code) + COMPOUND OF FORMULA I, nor nicotine (traditional name) (1319) + COMPOUND OF FORMULA I, novaluron (585) + COMPOUND OF FORMULA I, noviflumuron (586) + COMPOUND OF FORMULA I, O-5-dichloro-4-iodophenyl O-ethyl ethylphosphonothioate
10 (IUPAC name) (1057) + COMPOUND OF FORMULA I, O,O-diethyl O-4-methyl-2-oxo-2H-chromen-7-yl phosphorothioate (IUPAC name) (1074) + COMPOUND OF FORMULA I, O,O-diethyl O-6-methyl-2-propylpyrimidin-4-yl phosphorothioate (IUPAC name) (1075) + COMPOUND OF FORMULA I, O,O,O',O'-tetrapropyl dithiopyrophosphate (IUPAC name) (1424) + COMPOUND OF FORMULA I, oleic acid (IUPAC name) (593) + COMPOUND
15 OF FORMULA I, omethoate (594) + COMPOUND OF FORMULA I, oxamyl (602) + COMPOUND OF FORMULA I, oxydemeton-methyl (609) + COMPOUND OF FORMULA I, oxydeprofos (1324) + COMPOUND OF FORMULA I, oxydisulfoton (1325) + COMPOUND OF FORMULA I, pp'-DDT (219) + COMPOUND OF FORMULA I, para-dichlorobenzene [CCN] + COMPOUND OF FORMULA I, parathion (615) + COMPOUND OF FORMULA I,
20 parathion-methyl (616) + COMPOUND OF FORMULA I, penfluron (alternative name) [CCN] + COMPOUND OF FORMULA I, pentachlorophenol (623) + COMPOUND OF FORMULA I, pentachlorophenyl laurate (IUPAC name) (623) + COMPOUND OF FORMULA I, permethrin (626) + COMPOUND OF FORMULA I, petroleum oils (alternative name) (628) + COMPOUND OF FORMULA I, PH 60-38 (development code)
25 (1328) + COMPOUND OF FORMULA I, phenkapton (1330) + COMPOUND OF FORMULA I, phenothrin (630) + COMPOUND OF FORMULA I, phenthoate (631) + COMPOUND OF FORMULA I, phorate (636) + COMPOUND OF FORMULA I, phosalone (637) + COMPOUND OF FORMULA I, phosfolan (1338) + COMPOUND OF FORMULA I, phosmet (638) + COMPOUND OF FORMULA I, phosnichlor (1339) + COMPOUND OF
30 FORMULA I, phosphamidon (639) + COMPOUND OF FORMULA I, phosphine (IUPAC name) (640) + COMPOUND OF FORMULA I, phoxim (642) + COMPOUND OF FORMULA I, phoxim-methyl (1340) + COMPOUND OF FORMULA I, pirimetaphos (1344) + COMPOUND OF FORMULA I, pirimicarb (651) + COMPOUND OF FORMULA I, pirimiphos-ethyl (1345) + COMPOUND OF FORMULA I, pirimiphos-methyl (652) +
35 COMPOUND OF FORMULA I, polychlorodicyclopentadiene isomers (IUPAC name) (1346) + COMPOUND OF FORMULA I, polychloroterpenes (traditional name) (1347) +

COMPOUND OF FORMULA I, potassium arsenite [CCN] + COMPOUND OF FORMULA I,
potassium thiocyanate [CCN] + COMPOUND OF FORMULA I, prallethrin (655) +
COMPOUND OF FORMULA I, precocene I (alternative name) [CCN] + COMPOUND OF
FORMULA I, precocene II (alternative name) [CCN] + COMPOUND OF FORMULA I,
5 precocene III (alternative name) [CCN] + COMPOUND OF FORMULA I, primidophos
(1349) + COMPOUND OF FORMULA I, profenofos (662) + COMPOUND OF FORMULA I,
profluthrin [CCN] + COMPOUND OF FORMULA I, promacyl (1354) + COMPOUND OF
FORMULA I, promecarb (1355) + COMPOUND OF FORMULA I, propaphos (1356) +
COMPOUND OF FORMULA I, propetamphos (673) + COMPOUND OF FORMULA I,
10 propoxur (678) + COMPOUND OF FORMULA I, prothidathion (1360) + COMPOUND OF
FORMULA I, prothiofos (686) + COMPOUND OF FORMULA I, prothoate (1362) +
COMPOUND OF FORMULA I, protrifenbute [CCN] + COMPOUND OF FORMULA I,
pymetrozine (688) + COMPOUND OF FORMULA I, pyraclofos (689) + COMPOUND OF
FORMULA I, pyrazophos (693) + COMPOUND OF FORMULA I, pyresmethrin (1367) +
15 COMPOUND OF FORMULA I, pyrethrin I (696) + COMPOUND OF FORMULA I,
pyrethrin II (696) + COMPOUND OF FORMULA I, pyrethrins (696) + COMPOUND OF
FORMULA I, pyridaben (699) + COMPOUND OF FORMULA I, pyridalyl (700) +
COMPOUND OF FORMULA I, pyridaphenthion (701) + COMPOUND OF FORMULA I,
pyrimidifen (706) + COMPOUND OF FORMULA I, pyrimitate (1370) + COMPOUND OF
20 FORMULA I, pyriproxyfen (708) + COMPOUND OF FORMULA I, quassia (alternative
name) [CCN] + COMPOUND OF FORMULA I, quinalphos (711) + COMPOUND OF
FORMULA I, quinalphos-methyl (1376) + COMPOUND OF FORMULA I, quinothion
(1380) + COMPOUND OF FORMULA I, quintiofos (1381) + COMPOUND OF FORMULA
I, R-1492 (development code) (1382) + COMPOUND OF FORMULA I, rafoxanide
25 (alternative name) [CCN] + COMPOUND OF FORMULA I, resmethrin (719) +
COMPOUND OF FORMULA I, rotenone (722) + COMPOUND OF FORMULA I, RU
15525 (development code) (723) + COMPOUND OF FORMULA I, RU 25475
(development code) (1386) + COMPOUND OF FORMULA I, ryania (alternative name)
(1387) + COMPOUND OF FORMULA I, ryanodine (traditional name) (1387) +
30 COMPOUND OF FORMULA I, sabadilla (alternative name) (725) + COMPOUND OF
FORMULA I, schradan (1389) + COMPOUND OF FORMULA I, sebufos (alternative
name) + COMPOUND OF FORMULA I, selamectin (alternative name) [CCN] +
COMPOUND OF FORMULA I, SI-0009 (compound code) + COMPOUND OF FORMULA
I, SI-0205 (compound code) + COMPOUND OF FORMULA I, SI-0404 (compound code)
35 + COMPOUND OF FORMULA I, SI-0405 (compound code) + COMPOUND OF
FORMULA I, silafluofen (728) + COMPOUND OF FORMULA I, SN 72129 (development

code) (1397) + COMPOUND OF FORMULA I, sodium arsenite [CCN] + COMPOUND OF
FORMULA I, sodium cyanide (444) + COMPOUND OF FORMULA I, sodium fluoride
(IUPAC/Chemical Abstracts name) (1399) + COMPOUND OF FORMULA I, sodium
hexafluorosilicate (1400) + COMPOUND OF FORMULA I, sodium pentachlorophenoxide
5 (623) + COMPOUND OF FORMULA I, sodium selenate (IUPAC name) (1401) +
COMPOUND OF FORMULA I, sodium thiocyanate [CCN] + COMPOUND OF FORMULA
I, sophamide (1402) + COMPOUND OF FORMULA I, spinosad (737) + COMPOUND OF
FORMULA I, spiromesifen (739) + COMPOUND OF FORMULA I, spirotetmat (CCN) +
COMPOUND OF FORMULA I, sulcofuron (746) + COMPOUND OF FORMULA I,
10 sulcofuron-sodium (746) + COMPOUND OF FORMULA I, sulfluramid (750) +
COMPOUND OF FORMULA I, sulfotep (753) + COMPOUND OF FORMULA I, sulfuryl
fluoride (756) + COMPOUND OF FORMULA I, sulprofos (1408) + COMPOUND OF
FORMULA I, tar oils (alternative name) (758) + COMPOUND OF FORMULA I, tau-
fluvalinate (398) + COMPOUND OF FORMULA I, tazimcarb (1412) + COMPOUND OF
15 FORMULA I, TDE (1414) + COMPOUND OF FORMULA I, tebufenozide (762) +
COMPOUND OF FORMULA I, tebufenpyrad (763) + COMPOUND OF FORMULA I,
tebupirimfos (764) + COMPOUND OF FORMULA I, teflubenzuron (768) + COMPOUND
OF FORMULA I, tefluthrin (769) + COMPOUND OF FORMULA I, temephos (770) +
COMPOUND OF FORMULA I, TEPP (1417) + COMPOUND OF FORMULA I, terallethrin
20 (1418) + COMPOUND OF FORMULA I, terbam (alternative name) + COMPOUND OF
FORMULA I, terbufos (773) + COMPOUND OF FORMULA I, tetrachloroethane [CCN] +
COMPOUND OF FORMULA I, tetrachlorvinphos (777) + COMPOUND OF FORMULA I,
tetramethrin (787) + COMPOUND OF FORMULA I, theta-cypermethrin (204) +
COMPOUND OF FORMULA I, thiacloprid (791) + COMPOUND OF FORMULA I,
25 thiafenox (alternative name) + COMPOUND OF FORMULA I, thiamethoxam (792) +
COMPOUND OF FORMULA I, thicrofos (1428) + COMPOUND OF FORMULA I,
thiocarboxime (1431) + COMPOUND OF FORMULA I, thiocyclam (798) + COMPOUND
OF FORMULA I, thiocyclam hydrogen oxalate (798) + COMPOUND OF FORMULA I,
thiodicarb (799) + COMPOUND OF FORMULA I, thiofanox (800) + COMPOUND OF
30 FORMULA I, thiometon (801) + COMPOUND OF FORMULA I, thionazin (1434) +
COMPOUND OF FORMULA I, thiosultap (803) + COMPOUND OF FORMULA I,
thiosultap-sodium (803) + COMPOUND OF FORMULA I, thuringiensin (alternative name)
[CCN] + COMPOUND OF FORMULA I, tolfenpyrad (809) + COMPOUND OF FORMULA I,
tralomethrin (812) + COMPOUND OF FORMULA I, transfluthrin (813) + COMPOUND OF
35 FORMULA I, transpermethrin (1440) + COMPOUND OF FORMULA I, triamiphos (1441)
+ COMPOUND OF FORMULA I, triazamate (818) + COMPOUND OF FORMULA I,

- triazophos (820) + COMPOUND OF FORMULA I, triazuron (alternative name) +
COMPOUND OF FORMULA I, trichlorfon (824) + COMPOUND OF FORMULA I,
trichlormetaphos-3 (alternative name) [CCN] + COMPOUND OF FORMULA I, trichloronat
(1452) + COMPOUND OF FORMULA I, trifenofos (1455) + COMPOUND OF FORMULA I,
5 triflumuron (835) + COMPOUND OF FORMULA I, trimethacarb (840) + COMPOUND OF
FORMULA I, triprene (1459) + COMPOUND OF FORMULA I, vamidothion (847) +
COMPOUND OF FORMULA I, vaniliprole [CCN] + COMPOUND OF FORMULA I,
veratridine (alternative name) (725) + COMPOUND OF FORMULA I, veratrine (alternative
name) (725) + COMPOUND OF FORMULA I, XMC (853) + COMPOUND OF FORMULA I,
10 xylylcarb (854) + COMPOUND OF FORMULA I, YI-5302 (compound code) +
COMPOUND OF FORMULA I, zeta-cypermethrin (205) + COMPOUND OF FORMULA I,
zetamethrin (alternative name) + COMPOUND OF FORMULA I, zinc phosphide (640) +
COMPOUND OF FORMULA I, zolaprofos (1469) and ZXI 8901 (development code) (858)
+ COMPOUND OF FORMULA I,
- 15 a molluscicide selected from the group of substances consisting of bis(tributyltin)
oxide (IUPAC name) (913) + COMPOUND OF FORMULA I, bromoacetamide [CCN] +
COMPOUND OF FORMULA I, calcium arsenate [CCN] + COMPOUND OF FORMULA I,
cloethocarb (999) + COMPOUND OF FORMULA I, copper acetoarsenite [CCN] +
COMPOUND OF FORMULA I, copper sulfate (172) + COMPOUND OF FORMULA I,
20 fentin (347) + COMPOUND OF FORMULA I, ferric phosphate (IUPAC name) (352) +
COMPOUND OF FORMULA I, metaldehyde (518) + COMPOUND OF FORMULA I,
methiocarb (530) + COMPOUND OF FORMULA I, niclosamide (576) + COMPOUND OF
FORMULA I, niclosamide-olamine (576) + COMPOUND OF FORMULA I,
pentachlorophenol (623) + COMPOUND OF FORMULA I, sodium pentachlorophenoxide
25 (623) + COMPOUND OF FORMULA I, tazimcarb (1412) + COMPOUND OF FORMULA I,
thiodicarb (799) + COMPOUND OF FORMULA I, tributyltin oxide (913) + COMPOUND OF
FORMULA I, trifenmorph (1454) + COMPOUND OF FORMULA I, trimethacarb (840) +
COMPOUND OF FORMULA I, triphenyltin acetate (IUPAC name) (347) and triphenyltin
hydroxide (IUPAC name) (347) + COMPOUND OF FORMULA I,
- 30 a nematocide selected from the group of substances consisting of AKD-3088
(compound code) + COMPOUND OF FORMULA I, 1,2-dibromo-3-chloropropane
(IUPAC/Chemical Abstracts name) (1045) + COMPOUND OF FORMULA I, 1,2-
dichloropropane (IUPAC/ Chemical Abstracts name) (1062) + COMPOUND OF FORMULA
I, 1,2-dichloropropane with 1,3-dichloropropene (IUPAC name) (1063) + COMPOUND OF
35 FORMULA I, 1,3-dichloropropene (233) + COMPOUND OF FORMULA I, 3,4-
dichlorotetrahydrothiophene 1,1-dioxide (IUPAC/Chemical Abstracts name) (1065) +

COMPOUND OF FORMULA I, 3-(4-chlorophenyl)-5-methylrhodanine (IUPAC name) (980)
+ COMPOUND OF FORMULA I, 5-methyl-6-thioxo-1,3,5-thiadiazinan-3-ylacetic acid
(IUPAC name) (1286) + COMPOUND OF FORMULA I, 6-isopentenylaminopurine
(alternative name) (210) + COMPOUND OF FORMULA I, abamectin (1) + COMPOUND
5 OF FORMULA I, acetoprole [CCN] + COMPOUND OF FORMULA I, alanycarb (15) +
COMPOUND OF FORMULA I, aldicarb (16) + COMPOUND OF FORMULA I, aldoxycarb
(863) + COMPOUND OF FORMULA I, AZ 60541 (compound code) + COMPOUND OF
FORMULA I, benclothiaz [CCN] + COMPOUND OF FORMULA I, benomyl (62) +
COMPOUND OF FORMULA I, butylpyridaben (alternative name) + COMPOUND OF
10 FORMULA I, cadusafos (109) + COMPOUND OF FORMULA I, carbofuran (118) +
COMPOUND OF FORMULA I, carbon disulfide (945) + COMPOUND OF FORMULA I,
carbosulfan (119) + COMPOUND OF FORMULA I, chloropicrin (141) + COMPOUND OF
FORMULA I, chlorpyrifos (145) + COMPOUND OF FORMULA I, cloethocarb (999) +
COMPOUND OF FORMULA I, cytokinins (alternative name) (210) + COMPOUND OF
15 FORMULA I, dazomet (216) + COMPOUND OF FORMULA I, DBCP (1045) +
COMPOUND OF FORMULA I, DCIP (218) + COMPOUND OF FORMULA I, diamidafos
(1044) + COMPOUND OF FORMULA I, dichlofenthion (1051) + COMPOUND OF
FORMULA I, dicliphos (alternative name) + COMPOUND OF FORMULA I, dimethoate
(262) + COMPOUND OF FORMULA I, doramectin (alternative name) [CCN] +
20 COMPOUND OF FORMULA I, emamectin (291) + COMPOUND OF FORMULA I,
emamectin benzoate (291) + COMPOUND OF FORMULA I, eprinomectin (alternative
name) [CCN] + COMPOUND OF FORMULA I, ethoprophos (312) + COMPOUND OF
FORMULA I, ethylene dibromide (316) + COMPOUND OF FORMULA I, fenamiphos
(326) + COMPOUND OF FORMULA I, fenpyrad (alternative name) + COMPOUND OF
25 FORMULA I, fensulfothion (1158) + COMPOUND OF FORMULA I, fosthiazate (408) +
COMPOUND OF FORMULA I, fosthietan (1196) + COMPOUND OF FORMULA I,
furfural (alternative name) [CCN] + COMPOUND OF FORMULA I, GY-81 (development
code) (423) + COMPOUND OF FORMULA I, heterophos [CCN] + COMPOUND OF
FORMULA I, iodomethane (IUPAC name) (542) + COMPOUND OF FORMULA I,
30 isamidofos (1230) + COMPOUND OF FORMULA I, isazofos (1231) + COMPOUND OF
FORMULA I, ivermectin (alternative name) [CCN] + COMPOUND OF FORMULA I,
kinetin (alternative name) (210) + COMPOUND OF FORMULA I, mecarphon (1258) +
COMPOUND OF FORMULA I, metam (519) + COMPOUND OF FORMULA I, metam-
potassium (alternative name) (519) + COMPOUND OF FORMULA I, metam-sodium (519)
35 + COMPOUND OF FORMULA I, methyl bromide (537) + COMPOUND OF FORMULA I,
methyl isothiocyanate (543) + COMPOUND OF FORMULA I, milbemycin oxime

(alternative name) [CCN] + COMPOUND OF FORMULA I, moxidectin (alternative name) [CCN] + COMPOUND OF FORMULA I, *Myrothecium verrucaria* composition (alternative name) (565) + COMPOUND OF FORMULA I, NC-184 (compound code) + COMPOUND OF FORMULA I, oxamyl (602) + COMPOUND OF FORMULA I, phorate (636) +
5 COMPOUND OF FORMULA I, phosphamidon (639) + COMPOUND OF FORMULA I, phosphocarb [CCN] + COMPOUND OF FORMULA I, sebufos (alternative name) + COMPOUND OF FORMULA I, selamectin (alternative name) [CCN] + COMPOUND OF FORMULA I, spinosad (737) + COMPOUND OF FORMULA I, terbam (alternative name) + COMPOUND OF FORMULA I, terbufos (773) + COMPOUND OF FORMULA I,
10 tetrachlorothiophene (IUPAC/ Chemical Abstracts name) (1422) + COMPOUND OF FORMULA I, thiafenox (alternative name) + COMPOUND OF FORMULA I, thionazin (1434) + COMPOUND OF FORMULA I, triazophos (820) + COMPOUND OF FORMULA I, triazuron (alternative name) + COMPOUND OF FORMULA I, xlenols [CCN] + COMPOUND OF FORMULA I, YI-5302 (compound code) and zeatin (alternative name)
15 (210) + COMPOUND OF FORMULA I,
a nitrification inhibitor selected from the group of substances consisting of potassium ethylxanthate [CCN] and nitrapyrin (580) + COMPOUND OF FORMULA I,
a plant activator selected from the group of substances consisting of acibenzolar (6) + COMPOUND OF FORMULA I, acibenzolar-S-methyl (6) + COMPOUND OF FORMULA I,
20 probenazole (658) and *Reynoutria sachalinensis* extract (alternative name) (720) + COMPOUND OF FORMULA I,
a rodenticide selected from the group of substances consisting of 2-isovalerylindan-1,3-dione (IUPAC name) (1246) + COMPOUND OF FORMULA I, 4-(quinoxalin-2-ylamino)benzenesulfonamide (IUPAC name) (748) + COMPOUND OF FORMULA I,
25 alpha-chlorohydrin [CCN] + COMPOUND OF FORMULA I, aluminium phosphide (640) + COMPOUND OF FORMULA I, antu (880) + COMPOUND OF FORMULA I, arsenous oxide (882) + COMPOUND OF FORMULA I, barium carbonate (891) + COMPOUND OF FORMULA I, bithiosemi (912) + COMPOUND OF FORMULA I, brodifacoum (89) + COMPOUND OF FORMULA I, bromadiolone (91) + COMPOUND OF FORMULA I,
30 bromethalin (92) + COMPOUND OF FORMULA I, calcium cyanide (444) + COMPOUND OF FORMULA I, chloralose (127) + COMPOUND OF FORMULA I, chlorophacinone (140) + COMPOUND OF FORMULA I, cholecalciferol (alternative name) (850) + COMPOUND OF FORMULA I, coumachlor (1004) + COMPOUND OF FORMULA I, coumafuryl (1005) + COMPOUND OF FORMULA I, coumatetralyl (175) + COMPOUND
35 OF FORMULA I, crimidine (1009) + COMPOUND OF FORMULA I, difenacoum (246) + COMPOUND OF FORMULA I, difethialone (249) + COMPOUND OF FORMULA I,

diphacinone (273) + COMPOUND OF FORMULA I, ergocalciferol (301) + COMPOUND OF FORMULA I, flocoumafen (357) + COMPOUND OF FORMULA I, fluoroacetamide (379) + COMPOUND OF FORMULA I, flupropadine (1183) + COMPOUND OF FORMULA I, flupropadine hydrochloride (1183) + COMPOUND OF FORMULA I, gamma-HCH (430) + COMPOUND OF FORMULA I, HCH (430) + COMPOUND OF FORMULA I, hydrogen cyanide (444) + COMPOUND OF FORMULA I, iodomethane (IUPAC name) (542) + COMPOUND OF FORMULA I, lindane (430) + COMPOUND OF FORMULA I, magnesium phosphide (IUPAC name) (640) + COMPOUND OF FORMULA I, methyl bromide (537) + COMPOUND OF FORMULA I, norbormide (1318) + COMPOUND OF FORMULA I, phosacetim (1336) + COMPOUND OF FORMULA I, phosphine (IUPAC name) (640) + COMPOUND OF FORMULA I, phosphorus [CCN] + COMPOUND OF FORMULA I, pindone (1341) + COMPOUND OF FORMULA I, potassium arsenite [CCN] + COMPOUND OF FORMULA I, pyrinuron (1371) + COMPOUND OF FORMULA I, scilliroside (1390) + COMPOUND OF FORMULA I, sodium arsenite [CCN] + COMPOUND OF FORMULA I, sodium cyanide (444) + COMPOUND OF FORMULA I, sodium fluoroacetate (735) + COMPOUND OF FORMULA I, strychnine (745) + COMPOUND OF FORMULA I, thallium sulfate [CCN] + COMPOUND OF FORMULA I, warfarin (851) and zinc phosphide (640) + COMPOUND OF FORMULA I,

a synergist selected from the group of substances consisting of 2-(2-butoxyethoxy)-ethyl piperonylate (IUPAC name) (934) + COMPOUND OF FORMULA I, 5-(1,3-benzodioxol-5-yl)-3-hexylcyclohex-2-enone (IUPAC name) (903) + COMPOUND OF FORMULA I, farnesol with nerolidol (alternative name) (324) + COMPOUND OF FORMULA I, MB-599 (development code) (498) + COMPOUND OF FORMULA I, MGK 264 (development code) (296) + COMPOUND OF FORMULA I, piperonyl butoxide (649) + COMPOUND OF FORMULA I, piprotal (1343) + COMPOUND OF FORMULA I, propyl isomer (1358) + COMPOUND OF FORMULA I, S421 (development code) (724) + COMPOUND OF FORMULA I, sesamex (1393) + COMPOUND OF FORMULA I, sesasmolin (1394) and sulfoxide (1406) + COMPOUND OF FORMULA I,

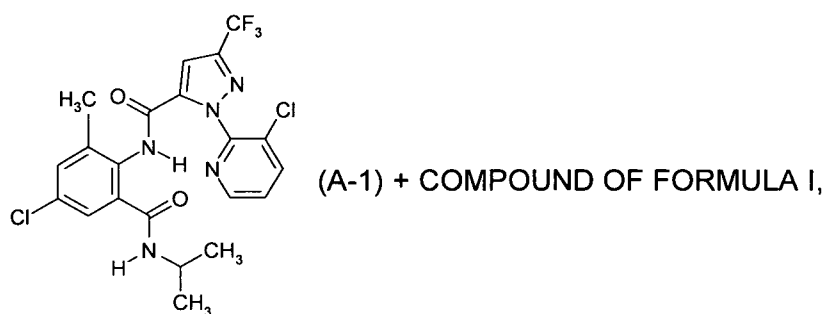
an animal repellent selected from the group of substances consisting of anthraquinone (32) + COMPOUND OF FORMULA I, chloralose (127) + COMPOUND OF FORMULA I, copper naphthenate [CCN] + COMPOUND OF FORMULA I, copper oxychloride (171) + COMPOUND OF FORMULA I, diazinon (227) + COMPOUND OF FORMULA I, dicyclopentadiene (chemical name) (1069) + COMPOUND OF FORMULA I, guazatine (422) + COMPOUND OF FORMULA I, guazatine acetates (422) + COMPOUND OF FORMULA I, methiocarb (530) + COMPOUND OF FORMULA I, pyridin-4-amine (IUPAC name) (23) + COMPOUND OF FORMULA I, thiram (804) + COMPOUND OF

FORMULA I, trimethacarb (840) + COMPOUND OF FORMULA I, zinc naphthenate [CCN] and ziram (856) + COMPOUND OF FORMULA I,

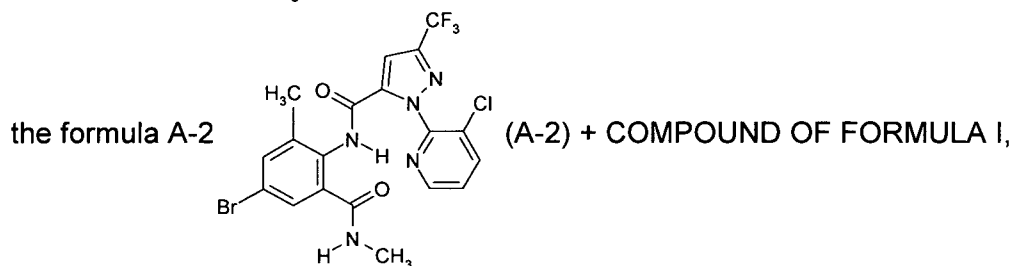
a virucide selected from the group of substances consisting of imanin (alternative name) [CCN] and ribavirin (alternative name) [CCN] + COMPOUND OF FORMULA I,

5 a wound protectant selected from the group of substances consisting of mercuric oxide (512) + COMPOUND OF FORMULA I, octhilinone (590) and thiophanate-methyl (802) + COMPOUND OF FORMULA I,

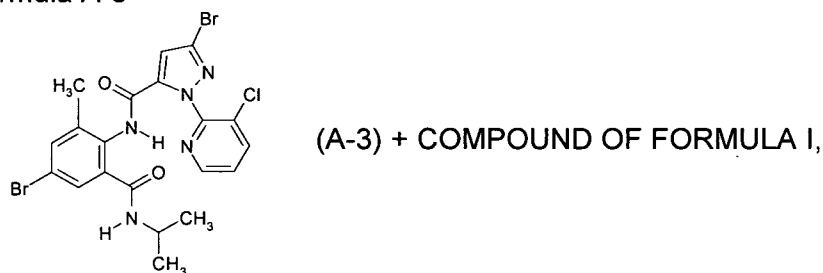
an insecticide selected from the group consisting of the compound of the formula A-1



10

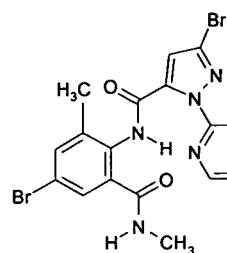


the formula A-3



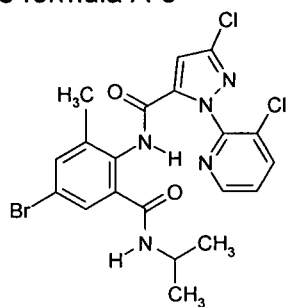
15

the formula A-4



(A-4) + COMPOUND OF FORMULA I,

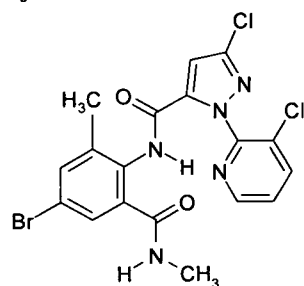
the formula A-5



(A-5) + COMPOUND OF FORMULA I,

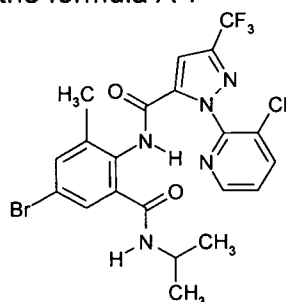
5

the formula A-6



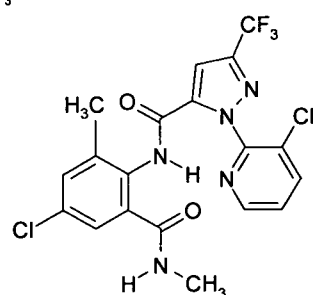
(A-6) + COMPOUND OF FORMULA I,

the formula A-7



(A-7) + COMPOUND OF FORMULA I,

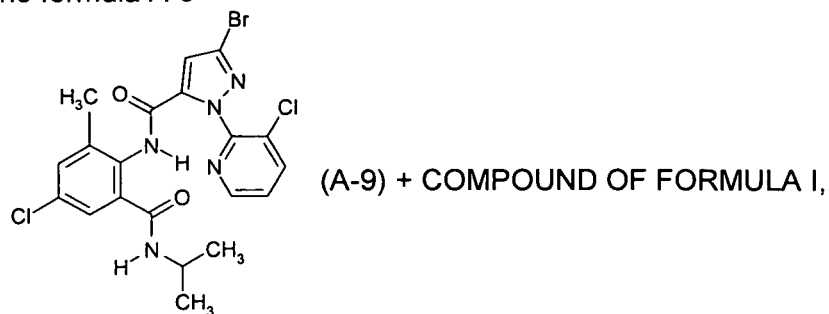
the formula A-8



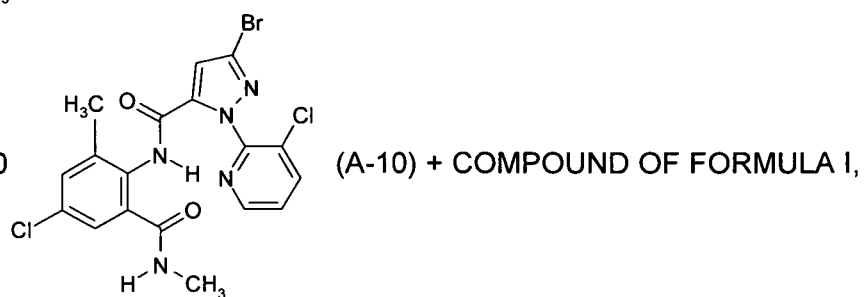
(A-8) + COMPOUND OF FORMULA I,

10

the formula A-9

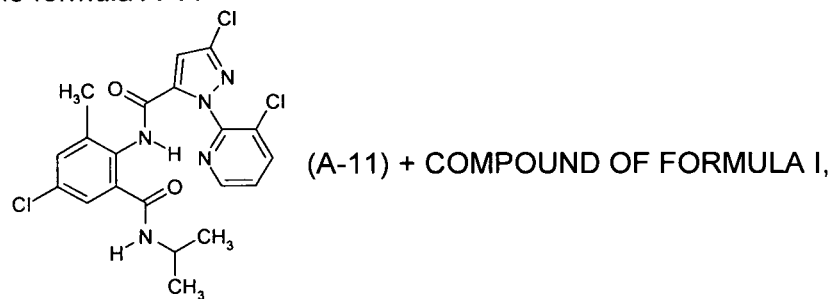


the formula A-10

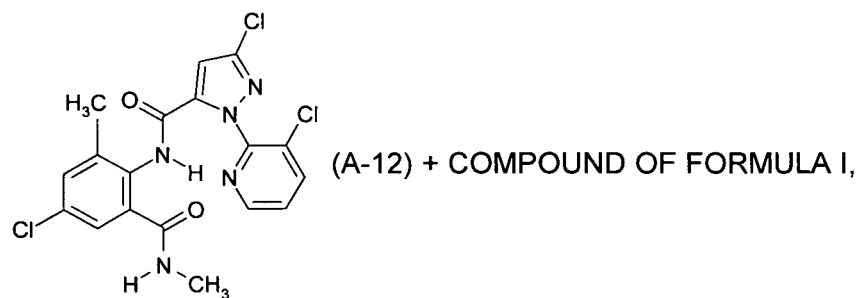


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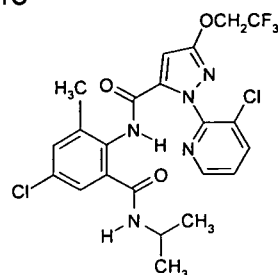
the formula A-11



the formula A-12

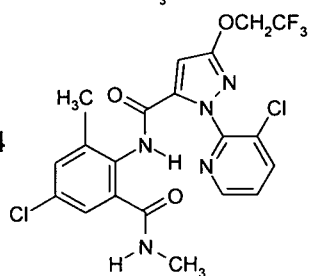


the formula A-13



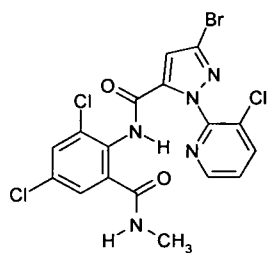
(A-13) + COMPOUND OF FORMULA I,

the formula A-14



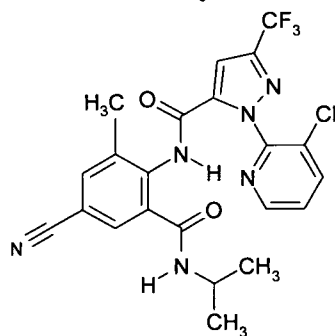
(A-14) + COMPOUND OF FORMULA I,

5 the formula A-15



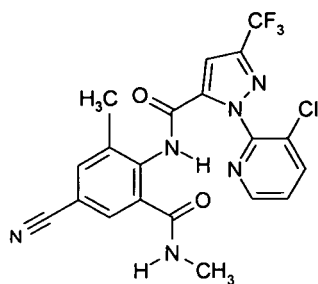
(A-15) + COMPOUND OF FORMULA I,

the formula A-16



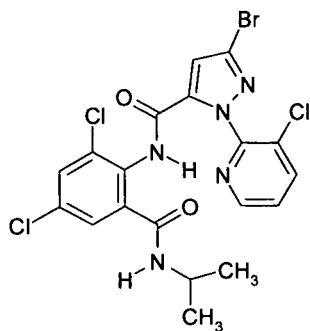
(A-16) + COMPOUND OF FORMULA I,

- 71 -



(A-17) + COMPOUND OF FORMULA I,

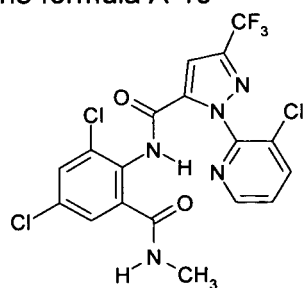
the formula A-17



(A-18) + COMPOUND OF FORMULA I,

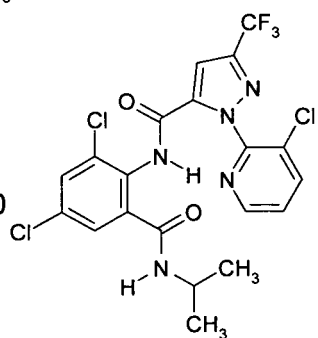
the formula A-18

the formula A-19

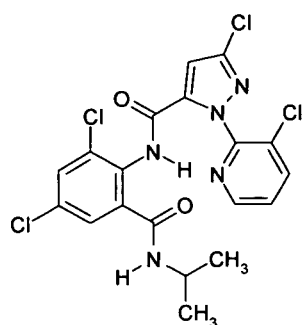


(A-19) + COMPOUND OF FORMULA I,

the formula A-20

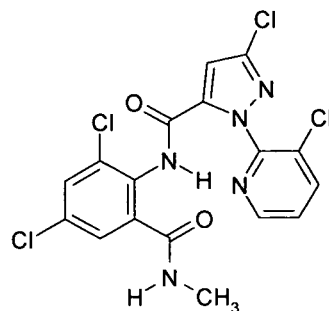


(A-20) + COMPOUND OF FORMULA I,



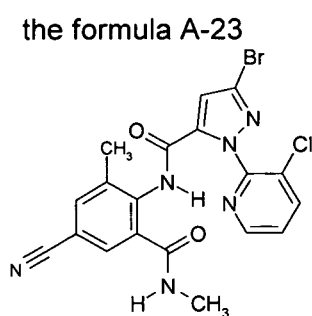
(A-21) + COMPOUND OF FORMULA I,

the formula A-21



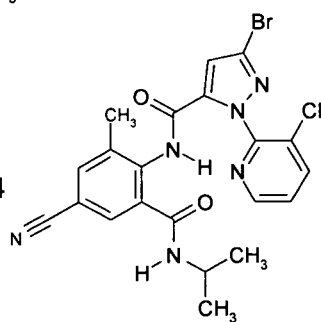
(A-22) + COMPOUND OF FORMULA I,

the formula A-22



(A-23) + COMPOUND OF FORMULA I,

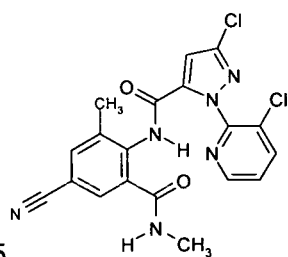
the formula A-23



(A-24) + COMPOUND OF FORMULA I,

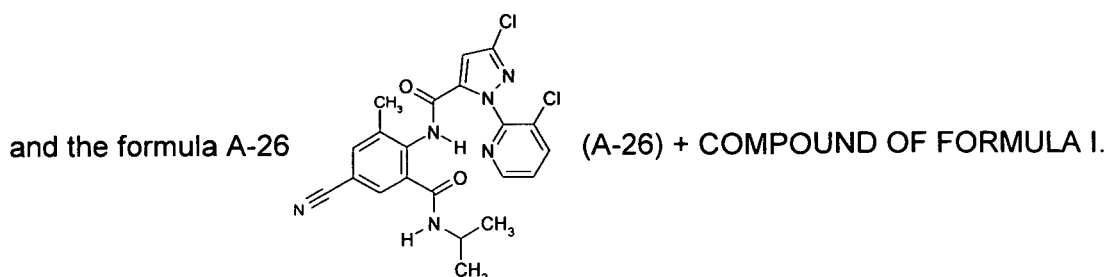
the formula A-24

5



(A-25) + COMPOUND OF FORMULA I,

the formula A-25



The references in brackets behind the active ingredients, e.g. [3878-19-1] refer to the Chemical Abstracts Registry number. The compounds of the formula A-1 to A-26 are described in WO 03/015518 or in WO 04/067528. The above described mixing partners are known. Where the active ingredients are included in "The Pesticide Manual" [The Pesticide Manual - A World Compendium; Thirteenth Edition; Editor: C. D. S. Tomlin; The British Crop Protection Council], they are described therein under the entry number given in round brackets hereinabove for the particular compound; for example, the compound "abamectin" is described under entry number (1). Where "[CCN]" is added hereinabove to the particular compound, the compound in question is included in the "Compendium of Pesticide Common Names", which is accessible on the internet [A. Wood; Compendium of Pesticide Common Names, Copyright © 1995-2004]; for example, the compound "acetoprole" is described under the internet address <http://www.alanwood.net/pesticides/acetoprole.html>.

Most of the active ingredients described above are referred to hereinabove by a so-called "common name", the relevant "ISO common name" or another "common name" being used in individual cases. If the designation is not a "common name", the nature of the designation used instead is given in round brackets for the particular compound; in that case, the IUPAC name, the IUPAC/Chemical Abstracts name, a "chemical name", a "traditional name", a "compound name" or a "development code" is used or, if neither one of those designations nor a "common name" is used, an "alternative name" is employed. "CAS Reg. No" means the Chemical Abstracts Registry Number.

Examples of fungicidal compounds which may be included in the composition of the invention are (*E*)-*N*-methyl-2-[2-(2,5-dimethylphenoxy)methyl]phenyl]-2-methoxyiminoacetamide (SSF-129), 4-bromo-2-cyano-*N,N*-dimethyl-6-trifluoromethylbenzimidazole-1-sulphonamide, α -[*N*-(3-chloro-2,6-xylyl)-2-methoxyacetamido]- γ -butyrolactone, 4-chloro-2-cyano-*N,N*-dimethyl-5-*p*-tolylimidazole-1-sulfonamide (IKF-916, cyamidazosulfamid),

3-5-dichloro-*N*-(3-chloro-1-ethyl-1-methyl-2-oxopropyl)-4-methylbenzamide (RH-7281, zoxamide), *N*-allyl-4,5,-dimethyl-2-trimethylsilylthiophene-3-carboxamide (MON65500), *N*-(1-cyano-1,2-dimethylpropyl)-2-(2,4-dichlorophenoxy)propionamide (AC382042), *N*-(2-methoxy-5-pyridyl)-cyclopropane carboxamide, acibenzolar, alanycarb, aldimorph, 5 anilazine, azaconazole, azoxystrobin, benalaxyl, benomyl, biloxazol, bitertanol, blasticidin S, bromuconazole, bupirimate, captafol, captan, carbendazim, carbendazim chlorhydrate, carboxin, carpropamid, carvone, CGA41396, CGA41397, chinomethionate, chlorothalonil, chlorozolate, clozylacon, copper containing compounds such as copper oxychloride, copper oxyquinolate, copper sulphate, copper tallate and Bordeaux mixture, cymoxanil, 10 cyproconazole, cyprodinil, debacarb, di-2-pyridyl disulphide 1,1'-dioxide, dichlofluanid, diclomezine, dicloran, diethofencarb, difenoconazole, difenzoquat, diflumetorim, *O,O*-di-*iso*-propyl-*S*-benzyl thiophosphate, dimefluzole, dimetconazole, dimethomorph, dimethirimol, diniconazole, dinocap, dithianon, dodecyl dimethyl ammonium chloride, dodemorph, dodine, doguadine, edifenphos, epoxiconazole, ethirimol, ethyl(*Z*)-*N*-benzyl-*N*- 15 ([methyl(methyl-thioethylideneaminooxycarbonyl)amino]thio)- β -alaninate, etridiazole, famoxadone, fenamidone (RPA407213), fenarimol, fenbuconazole, fenfuram, fenhexamid (KBR2738), fencpiclonil, fenpropidin, fenpropimorph, fentin acetate, fentin hydroxide, ferbam, ferimzone, fluazinam, fludioxonil, flumetover, fluoroimide, fluquinconazole, flusilazole, flutolanil, flutriafol, folpet, fuberidazole, furalaxyl, furametpyr, guazatine, hexaconazole, 20 hydroxyisoxazole, hymexazole, imazalil, imibenconazole, iminoctadine, iminoctadine triacetate, ipconazole, iprobenfos, iprodione, iprovalicarb (SZX0722), isopropanyl butyl carbamate, isoprothiolane, kasugamycin, kresoxim-methyl, LY186054, LY211795, LY248908, mancozeb, maneb, mefenoxam, mepanipyrim, mepronil, metalaxyl, metconazole, metiram, metiram-zinc, metominostrobin, myclobutanil, neoasozin, nickel 25 dimethyldithiocarbamate, nitrothal-*isopropyl*, nuarimol, ofurace, organomercury compounds, oxadixyl, oxasulfuron, oxolinic acid, oxpoconazole, oxycarboxin, pefurazoate, penconazole, pencycuron, phenazin oxide, phosetyl-Al, phosphorus acids, phthalide, picoxystrobin (ZA1963), polyoxin D, polyram, probenazole, prochloraz, procymidone, propamocarb, propiconazole, propineb, propionic acid, pyrazophos, pyrifenoxy, pyrimethanil, pyroquilon, pyr- 30 oxyfur, pyrrolnitrin, quaternary ammonium compounds, quinomethionate, quinoxifen, quintozone, sipconazole (F-155), sodium pentachlorophenate, spiroxamine, streptomycin, sulphur, tebuconazole, tecloftalam, tecnazene, tetraconazole, thiabendazole, thifluzamid, 2-(thiocyanomethylthio)benzothiazole, thiophanate-methyl, thiram, timibenconazole, tolclofos-methyl, tolylfluanid, triadimefon, triadimenol, triazbutil, triazoxide, tricyclazole, 35 tridemorph, trifloxystrobin, triforine, triflumizole, triticonazole, validamycin A, vapam, vinclozolin, zineb and ziram.

The compounds of formula I may be mixed with soil, peat or other rooting media for the protection of plants against seed-borne, soil-borne or foliar fungal diseases.

- 5 Examples of suitable synergists for use in the compositions include piperonyl butoxide, sesamex, safroxan and dodecyl imidazole.

The compounds of formula I according to the invention can also be used in combination with one or more other herbicides. In particular, the following mixtures of the compound of
10 formula I, preferably selected from one of the following Tables 1 to 22 are important:
compound of formula I + acetochlor, compound of formula I + acifluorfen, compound of formula I + acifluorfen-sodium, compound of formula I + aclonifen, compound of formula I + acrolein, compound of formula I + alachlor, compound of formula I + alloxydim, compound of formula I + allyl alcohol, compound of formula I + ametryn, compound of formula I +
15 amicarbazone, compound of formula I + amidosulfuron, compound of formula I + aminopyralid, compound of formula I + amitrole, compound of formula I + ammonium sulfamate, compound of formula I + anilofos, compound of formula I + asulam, compound of formula I + atraton, compound of formula I + atrazine, compound of formula I + azimsulfuron, compound of formula I + BCPC, compound of formula I + beflubutamid,
20 compound of formula I + benazolin, compound of formula I + benfluralin, compound of formula I + benfuresate, compound of formula I + bensulfuron, compound of formula I + bensulfuron-methyl, compound of formula I + bensulide, compound of formula I + bentazone, compound of formula I + benzfendizone, compound of formula I + benzobicyclon, compound of formula I + benzofenap, compound of formula I + bifenox,
25 compound of formula I + bilanafos, compound of formula I + bispyribac, compound of formula I + bispyribac-sodium, compound of formula I + borax, compound of formula I + bromacil, compound of formula I + bromobutide, compound of formula I + bromoxynil, compound of formula I + butachlor, compound of formula I + butafenacil, compound of formula I + butamifos, compound of formula I + butralin, compound of formula I +
30 butroxydim, compound of formula I + butylate, compound of formula I + cacodylic acid, compound of formula I + calcium chlorate, compound of formula I + cafenstrole, compound of formula I + carbetamide, compound of formula I + carfentrazone, compound of formula I + carfentrazone-ethyl, compound of formula I + CDEA, compound of formula I + CEPC, compound of formula I + chlorflurenol, compound of formula I + chlorflurenol-methyl,
35 compound of formula I + chloridazon, compound of formula I + chlorimuron, compound of formula I + chlorimuron-ethyl, compound of formula I + chloroacetic acid, compound of

formula I + chlorotoluron, compound of formula I + chlorpropham, compound of formula I + chlorsulfuron, compound of formula I + chlorthal, compound of formula I + chlorthal-dimethyl, compound of formula I + cinidon-ethyl, compound of formula I + cinmethylin, compound of formula I + cinosulfuron, compound of formula I + cisanilide, compound of
5 formula I + clethodim, compound of formula I + clodinafop, compound of formula I + clodinafop-propargyl, compound of formula I + clomazone, compound of formula I + clomeprop, compound of formula I + clopyralid, compound of formula I + cloransulam, compound of formula I + cloransulam-methyl, compound of formula I + CMA, compound of formula I + 4-CPB, compound of formula I + CPMF, compound of formula I + 4-CPP,
10 compound of formula I + CPPC, compound of formula I + cresol, compound of formula I + cumyluron, compound of formula I + cyanamide, compound of formula I + cyanazine, compound of formula I + cycloate, compound of formula I + cyclosulfamuron, compound of formula I + cycloxydim, compound of formula I + cyhalofop, compound of formula I + cyhalofop-butyl, compound of formula I + 2,4-D, compound of formula I + 3,4-DA,
15 compound of formula I + daimuron, compound of formula I + dalapon, compound of formula I + dazomet, compound of formula I + 2,4-DB, compound of formula I + 3,4-DB, compound of formula I + 2,4-DEB, compound of formula I + desmedipham, compound of formula I + dicamba, compound of formula I + dichlobenil, compound of formula I + ortho-dichlorobenzene, compound of formula I + para-dichlorobenzene, compound of formula I +
20 dichlorprop, compound of formula I + dichlorprop-P, compound of formula I + diclofop, compound of formula I + diclofop-methyl, compound of formula I + diclosulam, compound of formula I + difenzoquat, compound of formula I + difenzoquat metilsulfate, compound of formula I + diflufenican, compound of formula I + diflufenzopyr, compound of formula I + dimefuron, compound of formula I + dimepiperate, compound of formula I + dimethachlor,
25 compound of formula I + dimethametryn, compound of formula I + dimethenamid, compound of formula I + dimethenamid-P, compound of formula I + dimethipin, compound of formula I + dimethylarsinic acid, compound of formula I + dinitramine, compound of formula I + dinoterb, compound of formula I + diphenamid, compound of formula I + diquat, compound of formula I + diquat dibromide, compound of formula I + dithiopyr, compound of
30 formula I + diuron, compound of formula I + DNOC, compound of formula I + 3,4-DP, compound of formula I + DSMA, compound of formula I + EBEP, compound of formula I + endothal, compound of formula I + EPTC, compound of formula I + esprocarb, compound of formula I + ethalfluralin, compound of formula I + ethametsulfuron, compound of formula I + ethametsulfuron-methyl, compound of formula I + ethofumesate, compound of formula I
35 + ethoxyfen, compound of formula I + ethoxysulfuron, compound of formula I + etobenzanid, compound of formula I + fenoxaprop-P, compound of formula I + fenoxaprop-

P-ethyl, compound of formula I + fentrazamide, compound of formula I + ferrous sulfate, compound of formula I + flamprop-M, compound of formula I + flazasulfuron, compound of formula I + florasulam, compound of formula I + fluazifop, compound of formula I + fluazifop-butyl, compound of formula I + fluazifop-P, compound of formula I + fluazifop-P-butyl, compound of formula I + flucarbazone, compound of formula I + flucarbazone-sodium, compound of formula I + flucetosulfuron, compound of formula I + fluchloralin, compound of formula I + flufenacet, compound of formula I + flufenpyr, compound of formula I + flufenpyr-ethyl, compound of formula I + flumetsulam, compound of formula I + flumiclorac, compound of formula I + flumiclorac-pentyl, compound of formula I + flumioxazin, compound of formula I + fluometuron, compound of formula I + fluoroglycofen, compound of formula I + fluoroglycofen-ethyl, compound of formula I + flupropanate, compound of formula I + flupyrsulfuron, compound of formula I + flupyrsulfuron-methyl-sodium, compound of formula I + flurenol, compound of formula I + fluridone, compound of formula I + flurochloridone, compound of formula I + fluroxypyr, compound of formula I + flurtamone, compound of formula I + fluthiacet, compound of formula I + fluthiacet-methyl, compound of formula I + fomesafen, compound of formula I + foramsulfuron, compound of formula I + fosamine, compound of formula I + glufosinate, compound of formula I + glufosinate-ammonium, compound of formula I + glyphosate, compound of formula I + halosulfuron, compound of formula I + halosulfuron-methyl, compound of formula I + haloxyfop, compound of formula I + haloxyfop-P, compound of formula I + HC-252, compound of formula I + hexazinone, compound of formula I + imazamethabenz, compound of formula I + imazamethabenz-methyl, compound of formula I + imazamox, compound of formula I + imazapic, compound of formula I + imazapyr, compound of formula I + imazaquin, compound of formula I + imazethapyr, compound of formula I + imazosulfuron, compound of formula I + indanofan, compound of formula I + iodomethane, compound of formula I + iodosulfuron, compound of formula I + iodosulfuron-methyl-sodium, compound of formula I + ioxynil, compound of formula I + isoproturon, compound of formula I + isouron, compound of formula I + isoxaben, compound of formula I + isoxachlortole, compound of formula I + isoxaflutole, compound of formula I + karbutilate, compound of formula I + lactofen, compound of formula I + lenacil, compound of formula I + linuron, compound of formula I + MAA, compound of formula I + MAMA, compound of formula I + MCPA, compound of formula I + MCPA-thioethyl, compound of formula I + MCPB, compound of formula I + mecoprop, compound of formula I + mecoprop-P, compound of formula I + mefenacet, compound of formula I + mefluidide, compound of formula I + mesosulfuron, compound of formula I + mesosulfuron-methyl, compound of formula I + mesotrione, compound of formula I + metam, compound of formula I +

metamifop, compound of formula I + metamitron, compound of formula I + metazachlor, compound of formula I + methabenzthiazuron, compound of formula I + methylarsonic acid, compound of formula I + methyldymron, compound of formula I + methyl isothiocyanate, compound of formula I + metobenzuron, compound of formula I + metolachlor, compound of formula I + S-metolachlor, compound of formula I + metosulam, compound of formula I + metoxuron, compound of formula I + metribuzin, compound of formula I + metsulfuron, compound of formula I + metsulfuron-methyl, compound of formula I + MK-616, compound of formula I + molinate, compound of formula I + monolinuron, compound of formula I + MSMA, compound of formula I + naproanilide, compound of formula I + napropamide, compound of formula I + naptalam, compound of formula I + neburon, compound of formula I + nicosulfuron, compound of formula I + nonanoic acid, compound of formula I + norflurazon, compound of formula I + oleic acid (fatty acids), compound of formula I + orbencarb, compound of formula I + orthosulfamuron, compound of formula I + oryzalin, compound of formula I + oxadiargyl, compound of formula I + oxadiazon, compound of formula I + oxasulfuron, compound of formula I + oxaziclomefone, compound of formula I + oxyfluorfen, compound of formula I + paraquat, compound of formula I + paraquat dichloride, compound of formula I + pebulate, compound of formula I + pendimethalin, compound of formula I + penoxsulam, compound of formula I + pentachlorophenol, compound of formula I + pentanochlor, compound of formula I + pentoxazone, compound of formula I + pethoxamid, compound of formula I + petroleum oils, compound of formula I + phenmedipham, compound of formula I + phenmedipham-ethyl, compound of formula I + picloram, compound of formula I + picolinafen, compound of formula I + pinoxaden, compound of formula I + piperophos, compound of formula I + potassium arsenite, compound of formula I + potassium azide, compound of formula I + pretilachlor, compound of formula I + primisulfuron, compound of formula I + primisulfuron-methyl, compound of formula I + prodiamine, compound of formula I + profluazol, compound of formula I + profoxydim, compound of formula I + prometon, compound of formula I + prometryn, compound of formula I + propachlor, compound of formula I + propanil, compound of formula I + propaquizafop, compound of formula I + propazine, compound of formula I + propham, compound of formula I + propisochlor, compound of formula I + propoxycarbazone, compound of formula I + propoxycarbazone-sodium, compound of formula I + propyzamide, compound of formula I + prosulfocarb, compound of formula I + prosulfuron, compound of formula I + pyraclonil, compound of formula I + pyraflufen, compound of formula I + pyraflufen-ethyl, compound of formula I + pyrazolynate, compound of formula I + pyrazosulfuron, compound of formula I + pyrazosulfuron-ethyl, compound of formula I + pyrazoxyfen, compound of formula I + pyribenzoxim, compound of formula I +

pyributicarb, compound of formula I + pyridafol, compound of formula I + pyridate,
compound of formula I + pyriftalid, compound of formula I + pyriminobac, compound of
formula I + pyriminobac-methyl, compound of formula I + pyrimisulfan, compound of
formula I + pyrithiobac, compound of formula I + pyrithiobac-sodium, compound of formula I
5 + quinclorac, compound of formula I + quinmerac, compound of formula I + quinoclamine,
compound of formula I + quizalofop, compound of formula I + quizalofop-P, compound of
formula I + rimsulfuron, compound of formula I + sethoxydim, compound of formula I +
siduron, compound of formula I + simazine, compound of formula I + simetryn, compound
of formula I + SMA, compound of formula I + sodium arsenite, compound of formula I +
10 sodium azide, compound of formula I + sodium chlorate, compound of formula I +
sulcotrione, compound of formula I + sulfentrazone, compound of formula I + sulfometuron,
compound of formula I + sulfometuron-methyl, compound of formula I + sulfosate,
compound of formula I + sulfosulfuron, compound of formula I + sulfuric acid, compound of
formula I + tar oils, compound of formula I + 2,3,6-TBA, compound of formula I + TCA,
15 compound of formula I + TCA-sodium, compound of formula I + tebuthiuron, compound of
formula I + tepraloxym, compound of formula I + terbacil, compound of formula I +
terbumeton, compound of formula I + terbuthylazine, compound of formula I + terbutryn,
compound of formula I + thenylchlor, compound of formula I + thiazopyr, compound of
formula I + thifensulfuron, compound of formula I + thifensulfuron-methyl, compound of
20 formula I + thiobencarb, compound of formula I + tiocarbazil, compound of formula I +
topramezone, compound of formula I + tralkoxydim, compound of formula I + tri-allate,
compound of formula I + triasulfuron, compound of formula I + triaziflam, compound of
formula I + tribenuron, compound of formula I + tribenuron-methyl, compound of formula I +
tricamba, compound of formula I + triclopyr, compound of formula I + trietazine, compound
25 of formula I + trifloxysulfuron, compound of formula I + trifloxysulfuron-sodium, compound
of formula I + trifluralin, compound of formula I + triflusulfuron, compound of formula I +
triflusulfuron-methyl, compound of formula I + trihydroxytriazine, compound of formula I +
tritosulfuron, compound of formula I + [3-[2-chloro-4-fluoro-5-(1-methyl-6-trifluoromethyl-
2,4-dioxo-1,2,3,4-tetrahydropyrimidin-3-yl)phenoxy]-2-pyridyloxy]acetic acid ethyl ester
30 (CAS RN 353292-31-6), compound of formula I + 4-[(4,5-dihydro-3-methoxy-4-methyl-5-
oxo)-1H-1,2,4-triazol-1-ylcarbonylsulfamoyl]-5-methylthiophene-3-carboxylic acid (BAY636),
compound of formula I + BAY747 (CAS RN 335104-84-2), compound of formula I +
topramezone (CAS RN 210631-68-8), compound of formula I + 4-hydroxy-3-[[2-[(2-
methoxyethoxy)methyl]-6-(trifluoromethyl)-3-pyridinyl]carbonyl]-bicyclo[3.2.1]oct-3-en-2-one
35 (CAS RN 352010-68-5), and compound of formula I + 4-hydroxy-3-[[2-(3-methoxypropyl)-6-
(difluoromethyl)-3-pyridinyl]carbonyl]-bicyclo[3.2.1]oct-3-en-2-one.

The mixing partners of the compound of formula I may also be in the form of esters or salts, as mentioned e.g. in The Pesticide Manual, 12th Edition (BCPC), 2000.

- 5 In the above list, the compound of the formula I is preferably a compound of Tables 1 to 22, and more preferably, a compound of Tables A to F.

The mixing ratio of the compound of formula I to the mixing partner is preferably from 1: 100 to 1000:1.

10

The mixtures can advantageously be used in the above-mentioned formulations (in which case "active ingredient" relates to the respective mixture of compound of formula I with the mixing partner).

- 15 Some mixtures may comprise active ingredients which have significantly different physical, chemical or biological properties such that they do not easily lend themselves to the same conventional formulation type. In these circumstances other formulation types may be prepared. For example, where one active ingredient is a water insoluble solid and the other a water insoluble liquid, it may nevertheless be possible to disperse each active ingredient
- 20 in the same continuous aqueous phase by dispersing the solid active ingredient as a suspension (using a preparation analogous to that of an SC) but dispersing the liquid active ingredient as an emulsion (using a preparation analogous to that of an EW). The resultant composition is a suspoemulsion (SE) formulation.

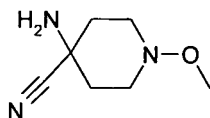
- 25 The mixtures comprising a compound of formula I selected from Tables 1 to 22 and one or more active ingredients as described above can be applied, for example, in a single "ready-mix" form, in a combined spray mixture composed from separate formulations of the single active ingredient components, such as a "tank-mix", and in a combined use of the single active ingredients when applied in a sequential manner, i.e. one after the other with a
- 30 reasonably short period, such as a few hours or days. The order of applying the compounds of formula I selected from Tables 1 to 22 and the active ingredients as described above is not essential for working the present invention.

The invention is illustrated by the following Examples:

EXAMPLE 1

5 Example P1: Preparation of carbonic acid 3-(2,5-dimethyl-phenyl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester methyl ester (compound A1)

Step P1.1: 4-Amino-1-methoxy-piperidine-4-carbonitrile



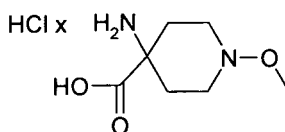
10

To 10g of N-methoxy-4-piperidone (Journal of Organic Chemistry (1961), 26, 1867-74) in 240ml of ammonium hydroxide (25% in water) was added 6.2g of ammonium chloride and 4.6g of sodium cyanide. After stirring for 18 hours at 25°C the reaction mixture was diluted with 200ml of water and extracted with ethyl acetate. The organic phase was separated,
15 dried over sodium sulfate and evaporated. 8.25g of 4-amino-1-methoxy-piperidine-4-carbonitrile was obtained as a light brown oil, which was used without further purification in the next step.

¹H-NMR (CDCl₃): δ 1.61-2.22 (br signals, total 6H), 2.61-3.43 (br signals, total 4H), 3.51 (s, 3H).

20

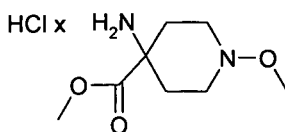
Step P1.2: 4-Amino-1-methoxy-piperidine-4-carboxylic acid hydrochloride salt



25 A mixture of 8.25g of 4-amino-1-methoxy-piperidine-4-carbonitrile and 30ml of hydrochloric acid 32% was heated to 100°C. After 16 hours the reaction mixture was evaporated. The solid residue was suspended in ethanol, filtered and dried to obtain 12.5g of 4-amino-1-methoxy-piperidine-4-carboxylic acid as the hydrochloride salt.

30 Step P1.3: 4-Amino-1-methoxy-piperidine-4-carboxylic acid methyl ester hydrochloride salt

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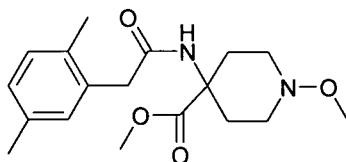


25.7g of thionyl chloride was added at a temperature of 0-10°C within 40 minutes to a suspension of 12.5g of 4-amino-1-methoxy-piperidine-4-carboxylic acid in 100ml of methanol. The reaction mixture was then heated up to 60°C for 48h. After cooling to 20°C, the solids were filtered and the filtrate was evaporated to give 13.2g of 4-amino-1-methoxy-piperidine-4-carboxylic acid methyl ester hydrochloride salt as a brown crystalline solid, mp: 198°C.

¹H-NMR (CDCl₃, free base): δ 1.40-1.72 (br signals, total 2H), 1.58 (s, 2H), 2.02-2.37 (br signals, total 2H), 2.58-2.90 (br signals, total 2H), 3.04-3.32 (br signals, total 2H), 3.52 (s, 3H), 3.73 (s, 3H).

LC-MS (EI, ES⁺): 189 (M+H)⁺ of the free base.

Step P1.4: 4-[2-(2,5-dimethyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester

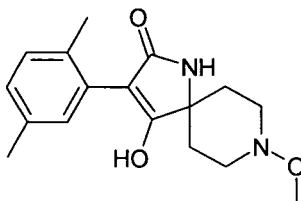


To 5.4g of potassium carbonate and 2g of 4-amino-1-methoxy-piperidine-4-carboxylic acid methyl ester hydrochloride in 10ml of acetonitrile was added 1.94g of (2,5-dimethyl-phenyl)-acetyl chloride in 5ml of acetonitrile at a temperature of 0-5°C. After stirring for 22 hours at room temperature, the reaction mixture was poured on ice water and extracted with ethyl acetate. The organic phase was dried over sodium sulfate, filtered and evaporated to yield 2.13g of 4-[2-(2,5-dimethyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester as a beige crystalline solid, mp: 91-93°C.

¹H-NMR (CDCl₃): δ 1.95-2.30 (br signals, total 4H), 2.27 (s, 3H), 2.33 (s, 3H), 2.77-3.23 (br signals, total 4H), 3.48 (s, 3H), 3.54 (s, 2H), 3.71 (s, 3H), 5.40 (br s, 1H), 7.02 (s, 1H), 7.04 (d, 1H), 7.11 (d, 1H).

LC-MS (EI, ES⁺): 335 (M+H)⁺

Step P1.5: 3-(2,5-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C1)

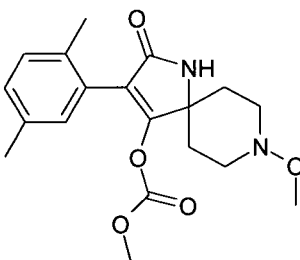


5

To 0.82g of sodium methylate in 20ml of dimethylformamide was added a solution of 4-[2-(2,5-dimethyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester in 10ml of dimethylformamide at a temperature of 60°C. After stirring for 3 hours at 60°C, the reaction mixture was evaporated. The residue was diluted with 10ml of water, neutralized with 10% hydrochloric acid and extracted with ethyl acetate. The organic phase was dried over sodium sulfate, filtered and evaporated to yield 1.23g of 3-(2,5-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C1) as a light brown resin. This material was triturated with diethyl ether/hexane, filtered and dried to afford a solid, mp: 176-177°C.

10
15 LC-MS (EI, ES⁺): 303 (M+H)⁺

Step P1.6: Carbonic acid 3-(2,5-dimethyl-phenyl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester methyl ester (compound A1)



20

To a solution of 200mg of 3-(2,5-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one and 0.14ml of ethyl diisopropyl amine in 2ml of chlorobenzene was added a solution of 66mg of methyl chloroformate in 0.5ml of chlorobenzene at 50°C. After stirring for 1 hour at 50°C, the reaction mixture was cooled to room temperature, diluted with 5 ml of chlorobenzene and washed with cold 5% aqueous sodium hydroxide and water. The organic phase was separated, dried over sodium sulfate, filtered and evaporated. The residue was purified by flash chromatography on silica gel. Yield: 120mg

25

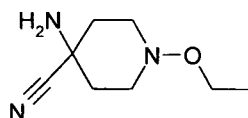
of carbonic acid 3-(2,5-dimethyl-phenyl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester methyl ester (title compound A1). This material was triturated with ethyl acetate/hexane, filtered and dried to afford a solid, mp: 186-188°C.

¹H-NMR (CDCl₃): δ 1.73 (m, 2H), 2.21 (s, 3H), 2.24 (m, 2H), 2.30 (s, 3H), 2.52 (m, 2H),
5 3.46 (m, 2H), 3.56 (br s, 3H), 3.63 (s, 3H), 6.78 (br s, 1H), 6.98 (s, 1H), 7.05 (d, 1H), 7.11 (d, 1H).

LC-MS (EI, ES⁺): 361 (M+H)⁺

10 Example P2: Preparation of 2,2-dimethyl-propionic acid 8-ethoxy-2-oxo-3-(2,4,6-trimethyl-phenyl)-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester (compound A54)

Step P2.1: 4-Amino-1-ethoxy-piperidine-4-carbonitrile

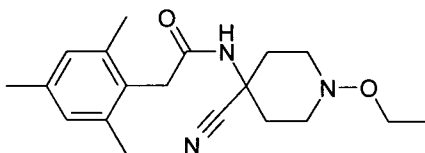


15 4-Amino-1-ethoxy-piperidine-4-carbonitrile was prepared analogously to the synthesis of 4-amino-1-methoxy-piperidine-4-carbonitrile (preparation example P1, step P1.1) starting from N-ethoxy-4-piperidone (Journal of Organic Chemistry (1961), 26, 1867-74).

¹H-NMR (d₆-DMSO, 88°C): δ 1.08 (t, 3H), 1.71 (m, 2H), 1.93 (m, 2H), 2.38 (br s, 2H), 2.67
20 (m, 2H), 3.09 (m, 2H), 3.63 (q, 2H).

LC-MS (EI, ES⁺): 170 (M+H)⁺

Step P2.2: N-(4-Cyano-1-ethoxy-piperidin-4-yl)-2-(2,4,6-trimethyl-phenyl)-acetamide

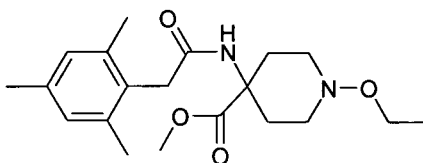


25 3.0 g of (2,4,6-trimethyl-phenyl)-acetyl chloride and 10 g of potassium carbonate in 90 ml of acetonitrile were treated at 0°C with a solution of 3.6 g of 4-amino-1-ethoxy-piperidine-4-carbonitrile in 30 ml of acetonitrile. After stirring for 18 hours at room temperature the
30 reaction mixture was poured into 100 ml of ice water and extracted with ethyl acetate. The organic phase was washed with brine and concentrated. Chromatography (heptane/ethyl

acetate 5:1) yielded 4.5 g of N-(4-cyano-1-ethoxy-piperidin-4-yl)-2-(2,4,6-trimethyl-phenyl)-acetamide as a solid, mp: 194-195°C.

LC-MS (EI, ES⁺): 330 (M+H)⁺

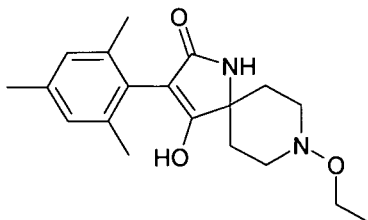
- 5 Step P2.3: 1-Ethoxy-4-[2-(2,4,6-trimethyl-phenyl)-acetyl-amino]-piperidine-4-carboxylic acid methyl ester



- 10 1.4 ml of concentrated sulfuric acid was slowly added to a solution of 4.3 g of N-(4-cyano-1-ethoxy-piperidin-4-yl)-2-(2,4,6-trimethyl-phenyl)-acetamide in 11 ml of methanol. After stirring 20 hours under reflux, the reaction mixture was allowed to cool down to room temperature and diluted with ice water. Sodium carbonate was added and the aqueous phase was extracted with ethyl acetate. The organic phase was dried over sodium sulfate,
- 15 filtered and concentrated. Chromatography (dichloromethane + 1% of ethanol) gave 3.2 g of 1-ethoxy-4-[2-(2,4,6-trimethyl-phenyl)-acetyl-amino]-piperidine-4-carboxylic acid methyl ester as a solid, mp: 131-132°C.

LC-MS (EI, ES⁺): 363 (M+H)⁺

- 20 Step P2.4: 8-Ethoxy-4-hydroxy-3-(2,4,6-trimethyl-phenyl)-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C8)



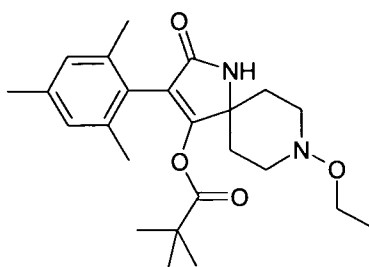
- 25 To a solution of 2.3 g of 1-ethoxy-4-[2-(2,4,6-trimethyl-phenyl)-acetyl-amino]-piperidine-4-carboxylic acid methyl ester in 26 ml of dimethylformamide was added 3 g of sodium methoxide. The reaction mixture was heated to 65°C and stirred for 5 hours. The reaction mixture was poured into brine, neutralized and extracted with ethyl acetate. The organic phase was washed with brine, dried with sodium sulfate, filtered and concentrated. The

residue was subjected to column chromatography (heptane/ethyl acetate 2:1) to yield 510 mg of 8-ethoxy-4-hydroxy-3-(2,4,6-trimethyl-phenyl)-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C8) as a solid, mp: >250°C.

LC-MS (EI, ES⁺): 331 (M+H)⁺

5

Step P2.5: 2,2-Dimethyl-propionic acid 8-ethoxy-2-oxo-3-(2,4,6-trimethyl-phenyl)-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester (compound A54)



10

To 143 mg of 8-ethoxy-4-hydroxy-3-(2,4,6-trimethyl-phenyl)-1,8-diaza-spiro[4.5]dec-3-en-2-one in 1 ml of tetrahydrofuran were added 36 µl of pyridine and 53 µl of pivaloyl chloride. After stirring at room temperature for 20 hours, water and ethyl acetate were added and the phases separated. The aqueous phase was extracted with ethyl acetate, the combined organic phases dried with sodium sulfate, filtered and concentrated. Chromatography (heptane/ethyl acetate 2:1) yielded 117 mg of 2,2-dimethyl-propionic acid 8-ethoxy-2-oxo-3-(2,4,6-trimethyl-phenyl)-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester (title compound A54) as a solid, mp: 230-231°C.

15

¹H-NMR (CDCl₃): δ 1.05 (s, 9H), 1.18 (t, 3H), 1.70 (m, 2H), 2.12 (m, 2H), 2.14 (s, 6H), 2.22 (s, 3H), 2.51 (m, 2H), 3.38 (m, 2H), 3.75 (q, 2H), 6.42 (br s, 1H), 6.81 (s, 2H).

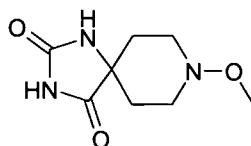
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LC-MS (EI, ES⁺): 415 (M+H)⁺

Example P3: Preparation of 4-amino-1-methoxy-piperidine-4-carboxylic acid hydrochloride salt

25

Step P3.1: 8-Methoxy-1,3,8-triaza-spiro[4.5]decane-2,4-dione

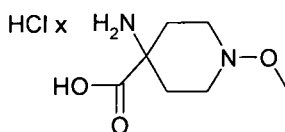


N-methoxy-4-piperidone (Journal of Organic Chemistry (1961), 26, 1867-74) (12.9 g) was added to a solution of ammonium carbonate (14.41 g) and potassium cyanide (13.02 g) in water (200 ml). The reaction mixture was stirred for 30 minutes at room temperature, then for 16 hours at 55-60°C and partially concentrated. The aqueous residue was treated with
5 brine and extracted with ethyl acetate (8x). The combined organic phases were dried over sodium sulfate and concentrated. The crude product was triturated with diethyl ether, filtered and dried. Yield: 11.5 g of 8-methoxy-1,3,8-triaza-spiro[4.5]decane-2,4-dione as a solid, mp: 230-234°C.

¹H-NMR (d₆-DMSO): δ 1.26-1.71 (br signals, total 2H), 1.71-2.12 (br signals, total 2H),
10 2.50-2.68 and 2.91-3.26 (br signals, total 4H), 3.40 (s, 3H), 8.34 and 8.54 (each br s, total 1H), 10.64 (br s, 1H).

LC-MS (EI, ES⁺): 200 (M+H)⁺

Step P3.2: 4-Amino-1-methoxy-piperidine-4-carboxylic acid hydrochloride salt



A solution of 8-methoxy-1,3,8-triaza-spiro[4.5]decane-2,4-dione (4.0 g) and sodium hydroxide (0.8 g) in water (25 ml) was heated at 160°C in an autoclave for 18 hours. The
20 reaction mixture was concentrated, the white solid residue taken up in hot methanol, filtered and the filtrate evaporated. The residue was treated with toluol several times to remove water azeotropically until constant weight. Yield: 4.19 g of 4-amino-1-methoxy-piperidine-4-carboxylic acid hydrochloride salt as a solid. This material was identical to the compound described above under preparation example P1, step P1.2.

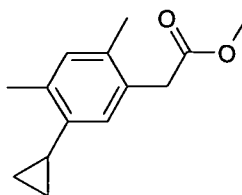
¹H-NMR (d₄-MeOH): δ 2.13-2.64 (br signals, total 4H), 3.39-3.82 (br signals, total 4H), 3.85 (s, 3H).

LC-MS (EI, ES⁺): 175 (M+H)⁺ of the free base.

Example P4: Preparation of 3-(5-cyclopropyl-2,4-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C16)

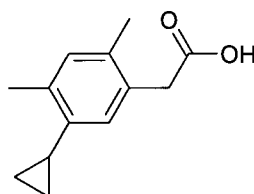
Step P4.1: (5-Cyclopropyl-2,4-dimethyl-phenyl)-acetic acid methyl ester

- 88 -



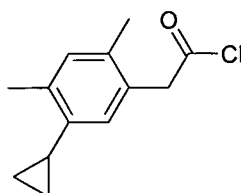
To a solution of 6.0 g of (5-bromo-2,4-dimethyl-phenyl)-acetic acid methyl ester (WO99/48869) in 100 ml of toluene was added 2.2 g of cyclopropylboronic acid and 20 g of potassium phosphate tribasic trihydrate at room temperature. The reaction mixture was stirred for 5 minutes under nitrogen atmosphere, followed by further addition of 1.2 g of tetrakis(triphenylphosphine)palladium(0). After heating and stirring for 16 hours at 110°C, the reaction mixture was filtered, the solvent removed *in vacuo* and the residue was subjected to silica gel chromatography (isohexane/diethyl ether 2:1) to yield 2.5 g of (5-cyclopropyl-2,4-dimethyl-phenyl)-acetic acid methyl ester.

Step P4.2: (5-Cyclopropyl-2,4-dimethyl-phenyl)-acetic acid



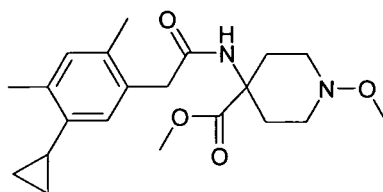
2.5 g of (5-cyclopropyl-2,4-dimethyl-phenyl)-acetic acid methyl ester in 30 ml of methanol was kept at ice bath temperature and treated with 0.5 g of sodium hydroxide in 5 portions. The reaction mixture was stirred at room temperature for 1.5 hours. The solvent was removed *in vacuo*, the residue poured into water and extracted with diethyl ether. The combined organic phases were dried with sodium sulfate, filtered and concentrated to afford 2.3 g of (5-cyclopropyl-2,4-dimethyl-phenyl)-acetic acid.

Step P4.3: (5-Cyclopropyl-2,4-dimethyl-phenyl)-acetyl chloride



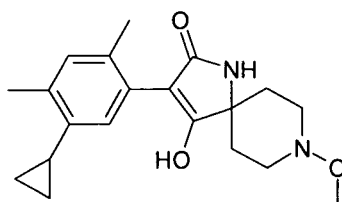
2.3 g of (5-cyclopropyl-2,4-dimethyl-phenyl)-acetic acid in 20 ml of dichloromethane were treated with 2.1 g of oxalyl chloride and a catalytic amount of dimethylformamide. The reaction mixture was stirred at room temperature for 2 hours. The solvent was removed *in vacuo* and the crude residue of (5-cyclopropyl-2,4-dimethyl-phenyl)-acetyl chloride (2.5 g) was used for the next step.

Step P4.4: 4-[2-(5-Cyclopropyl-2,4-dimethyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester



To 4 g of potassium carbonate and 2.5 g of 4-amino-1-methoxy-piperidine-4-carboxylic acid methyl ester hydrochloride (preparation example P1, step P1.3) in 10 ml of acetonitrile was added at 0-5°C a solution of 2.1 g of crude (5-cyclopropyl-2,4-dimethyl-phenyl)-acetyl chloride in 5 ml of acetonitrile. After stirring for 18 hours at room temperature, the solvent was removed *in vacuo* and the crude residue of 4-[2-(5-cyclopropyl-2,4-dimethyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester (3.2 g) was used without further purification in the next step.

Step P4.5: 3-(5-Cyclopropyl-2,4-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C16)



To 1.3 g of sodium methoxide in 10 ml of dimethylformamide was added a solution of 3.2 g of crude 4-[2-(5-cyclopropyl-2,4-dimethyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester in 10 ml of dimethylformamide and the reaction mixture was heated to 65°C for 2 hours. The mixture was poured into ice water, extracted with dichloromethane, the combined organic phases dried with sodium sulfate, filtered and

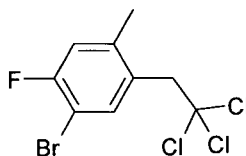
concentrated. The residue was subjected to column chromatography (dichloromethane/methanol 95:5) to yield 700 mg of 3-(5-cyclopropyl-2,4-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (title compound C16) as a wax.

LC-MS (EI, ES⁺): 343 (M+H)⁺

5

Example P5: Preparation of 3-(5-bromo-4-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C19) and 3-(4'-chloro-6-fluoro-4-methyl-biphenyl-3-yl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound A82)

10 Step P5.1: 1-Bromo-2-fluoro-4-methyl-5-(2,2,2-trichloro-ethyl)-benzene



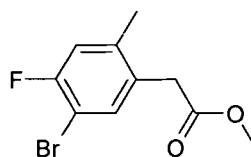
To a solution of vinylidene chloride (127 ml), *tert*-butyl nitrite (19 ml) and copper(II)chloride (18.4 g) in acetonitrile (150 ml) was added a solution of 5-bromo-4-fluoro-2-methyl-phenylamine (Bioorganic & Medicinal Chemistry Letters (2006), 16(2), 457-460) (21.5 g) in acetonitrile (100 ml) dropwise below 20°C. The reaction mixture was stirred at room temperature for 48 hours, poured on diluted HCl and extracted with *tert*-butyl methyl ether (3x). The combined organic layers were washed with brine, dried over sodium sulfate and concentrated. The residue was purified by chromatography on silica gel (ethyl acetate/cyclohexane 1:4). Yield: 29.20 g of 1-bromo-2-fluoro-4-methyl-5-(2,2,2-trichloro-ethyl)-benzene as an oil.

20

¹H-NMR (CDCl₃): δ 2.42 (s, 3H), 3.93 (s, 2H), 7.00 (d, ³J(H,F)=9.2Hz, 1H), 7.69 (d, ⁴J(H,F)=7.0Hz, 1H).

25

Step P5.2: (5-Bromo-4-fluoro-2-methyl-phenyl)-acetic acid methyl ester

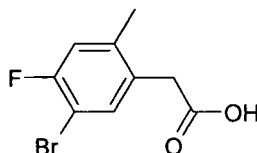


30 To a solution of 1-bromo-2-fluoro-4-methyl-5-(2,2,2-trichloro-ethyl)-benzene (29.2g) in methanol (100 ml) was added a sodium methoxide solution (30% in methanol, 78.2 ml)

dropwise. The reaction mixture was stirred at reflux for 24 hours, cooled to 5°C and treated with concentrated sulfuric acid (13.2 ml) dropwise. After further warming to reflux for 21 hours, the mixture was concentrated and the residue diluted with water/ethyl acetate. The aqueous layer was extracted with ethyl acetate, the combined organic layers were washed with brine, dried over sodium sulfate and concentrated. The residue was purified by chromatography on silica gel (ethyl acetate/cyclohexane 1:3). Yield: 18.76 g of (5-bromo-4-fluoro-2-methyl-phenyl)-acetic acid methyl ester as an oil.

$^1\text{H-NMR}$ (CDCl_3): δ 2.24 (s, 3H), 3.56 (s, 2H), 3.69 (s, 3H), 6.93 (d, $^3\text{J}(\text{H},\text{F})=9.3\text{Hz}$, 1H), 7.35 (d, $^4\text{J}(\text{H},\text{F})=7.0\text{Hz}$, 1H).

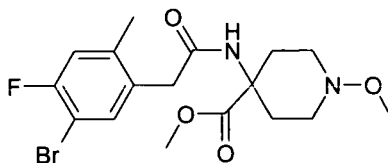
Step P5.3: (5-Bromo-4-fluoro-2-methyl-phenyl)-acetic acid



To a solution of (5-bromo-4-fluoro-2-methyl-phenyl)-acetic acid methyl ester (10.3 g) in methanol (50 ml) was added 1N aqueous sodium hydroxide (47.4 ml) and the reaction mixture was stirred at room temperature for 18 hours. The mixture was concentrated, the residue treated with 1N hydrochloric acid, the resulting precipitate filtered off, washed with ice-water and dried. Yield: 8.60 g of (5-bromo-4-fluoro-2-methyl-phenyl)-acetic acid as a solid, mp: 100-101°C.

$^1\text{H-NMR}$ (CDCl_3): δ 2.26 (s, 3H), 3.60 (s, 2H), 6.95 (d, $^3\text{J}(\text{H},\text{F})=9.3\text{Hz}$, 1H), 7.36 (d, $^4\text{J}(\text{H},\text{F})=7.0\text{Hz}$, 1H), 8.6 (br s, 1H).

Step P5.4: 4-[2-(5-Bromo-4-fluoro-2-methyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester



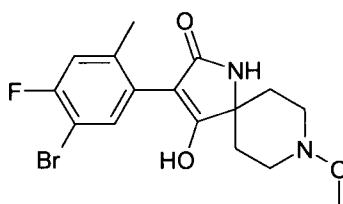
A suspension of (5-bromo-4-fluoro-2-methyl-phenyl)-acetic acid (preparation example P5.3) (8.0 g) and 1,1'-carbonyldiimidazole (5.8 g) in tetrahydrofuran (150 ml) was heated at reflux

for 30 minutes. Upon cooling to room temperature, triethylamine (9.0 ml) and 4-amino-1-methoxy-piperidine-4-carboxylic acid methyl ester hydrochloride salt (preparation example P1, step P1.3) (13.8 g) were added and warming at reflux was continued for 3 hours. The cold reaction mixture was poured on water/ethyl acetate, the layers separated, the organic phase washed with brine, dried over sodium sulfate and concentrated. The residue was solubilised in ethyl acetate/cyclohexane 3:1 and purified by filtration on alumina. Yield: 6.83 g of 4-[2-(5-bromo-4-fluoro-2-methyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester as a solid, mp: 192-193°C.

¹H-NMR (CDCl₃): δ 2.04-2.54 (br signals, total 4H), 2.26 (s, 3H), 2.79-3.27 (br signals, total 4H), 3.49 (br s, 5H), 3.71 (s, 3H), 5.40 (br s, 1H), 6.99 (d, ³J(H,F)=9.3Hz, 1H), 7.38 (d, ⁴J(H,F)=6.9Hz, 1H).

MS (FIMS-EI, ES+): 417/419 (M+H)⁺

Step P5.5: 3-(5-Bromo-4-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diazaspiro[4.5]dec-3-en-2-one (compound C19)

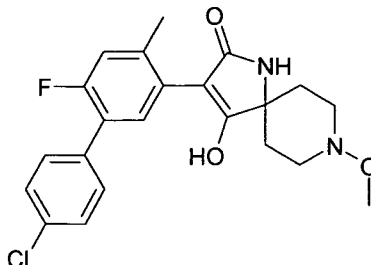


To a solution of 4-[2-(5-bromo-4-fluoro-2-methyl-phenyl)-acetylamino]-1-methoxy-piperidine-4-carboxylic acid methyl ester (6.0 g) in dimethylformamide (20 ml) at 100°C was added potassium *tert*-butoxide (3.23 g) and stirring continued at 100°C for 10 minutes. The reaction mixture was quenched at room temperature by addition of acetic acid (1.64 ml), diluted with water (20 ml) and extracted with *tert*-butyl methyl ether (3x). The combined organic layers were washed with brine, dried over sodium sulfate and concentrated. The residue was triturated with acetonitrile, filtered and dried. Yield: 3.69 g of 3-(5-bromo-4-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diazaspiro[4.5]dec-3-en-2-one (title compound C19) as a solid, mp: 229-230°C.

¹H-NMR (d₆-DMSO): δ 1.45 (m, 2H), 2.09 (s, 3H), 2.15 (m, 2H), 2.62 (m, 2H), 3.25 (m, 2H), 3.42 (s, 3H), 6.93 (d, ³J(H,F)=9.8Hz, 1H), 7.27 (d, ⁴J(H,F)=7.3Hz, 1H), 7.92 (br s, 1H), 10.64 (br s, 1H).

MS (FIMS-EI, ES-): 383/385 (M-H)⁻

Step P5.6: 3-(4'-Chloro-6-fluoro-4-methyl-biphenyl-3-yl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C24)



5

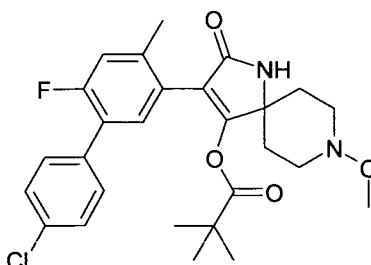
To a suspension of 3-(5-bromo-4-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (230 mg) in dimethoxyethane (10 ml) under nitrogen atmosphere was added tetrakis(triphenylphosphine)palladium(0) (35 mg) and the mixture stirred at room temperature for 15 minutes. After further addition of water (2 ml), 4-chlorophenylboronic acid (112 mg) and sodium carbonate (250 mg), the mixture was heated at reflux for 8 hours. The reaction mixture was acidified at room temperature with 1N hydrochloric acid and extracted with ethyl acetate (3x). The combined organic layers were washed with brine, dried over sodium sulfate and concentrated. The residue was purified by chromatography on silica gel (ethyl acetate/cyclohexane 5:1). Yield: 170 mg of 3-(4'-chloro-6-fluoro-4-methyl-biphenyl-3-yl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C24) as a solid.

15

MS (FIMS-EI, ES⁺): 417/419 (M+H)⁺; MS (FIMS-EI, ES⁻): 415/417 (M-H)⁻

20

Step P5.7: 2,2-Dimethyl-propionic acid 3-(4'-chloro-6-fluoro-4-methyl-biphenyl-3-yl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester (compound A82)



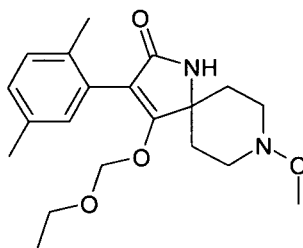
25

To a solution of 3-(4'-chloro-6-fluoro-4-methyl-biphenyl-3-yl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (170 mg) and pivaloyl chloride (98 mg) in acetonitrile (5 ml) was added pyridine (64 mg) and the reaction mixture stirred at room temperature for 8

hours. The mixture was poured on diluted HCl and extracted with ethyl acetate (3x). The combined organic layers were washed with brine, dried over sodium sulfate and concentrated. The residue was triturated with heptane, filtered and dried. Yield: 100 mg of 2,2-dimethyl-propionic acid 3-(4'-chloro-6-fluoro-4-methyl-biphenyl-3-yl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester (title compound A82) as a solid, mp: 150-151°C.

¹H-NMR (CDCl₃): δ 1.09 (s, 9H), 1.72 (m, 2H), 2.10 (m, 2H), 2.29 (s, 3H), 2.49 (m, 2H), 3.44 (m, 2H), 3.55 (s, 3H), 6.59 (s, 1H), 7.02 (d, ³J(H,F)=11.5Hz, 1H), 7.10 (d, ⁴J(H,F)=8.0Hz, 1H), 7.36 (d, 2H), 7.41 (d, 2H).

10 Example P6: Preparation of 3-(2,5-dimethyl-phenyl)-4-ethoxymethoxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound A61)



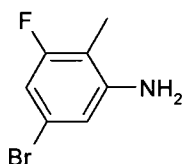
- 15 To 250 mg of 3-(2,5-dimethyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound C1, preparation example P1, step P1.5) in 2.5 ml of tetrahydrofuran were added 160 µl of Hünig's base and 85 µl of chloromethyl ethyl ether. After stirring for 20 hours at room temperature, water and ethyl acetate were added and the layers were separated. The aqueous phase was extracted with ethyl acetate, the combined organic
- 20 phases dried with sodium sulfate, filtered and concentrated. Chromatography (heptane/acetone 4:1) yielded 72 mg of 3-(2,5-dimethyl-phenyl)-4-ethoxymethoxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (title compound A61) as a solid, mp: 150-152°C.
- ¹H-NMR (CDCl₃): δ 1.15 (t, 3H), 1.65 (m, 2H), 2.16 (s, 3H), 2.28 (s, 3H), 2.31 (m, 2H), 2.45 (m, 2H), 3.46 (m, 2H), 3.56 (s, 3H), 3.58 (q, 2H), 4.80 (s, 2H), 6.05 (br s, 1H), 6.97 (s, 1H), 7.03 (d, 1H), 7.08 (d, 1H).
- LC-MS (EI, ES⁺): 361 (M+H)⁺

30 Example P7: Preparation of 3-(5-bromo-3-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound F3) and carbonic acid 3-(5-bromo-3-fluoro-

2-methyl-phenyl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester ethyl ester
(compound E4)

Step P7.1: 5-Bromo-3-fluoro-2-methyl-phenylamine

5

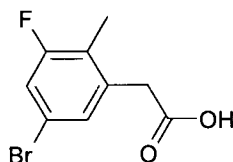


A mixture of 5-bromo-1-fluoro-2-methyl-3-nitro-benzene (9.96 g) and iron powder (11.9 g) in ethanol (100 ml), water (20 ml) and concentrated hydrochloric acid (2 ml) was heated at reflux for one hour. After cooling, the reaction mixture was filtered through hyflo (calcined diatomaceous earth) and concentrated. The residue was purified by chromatography on silica gel (ethyl acetate/cyclohexane 1:9 to 1:4). Yield: 7.06 g of 5-bromo-3-fluoro-2-methyl-phenylamine as an oil.

$^1\text{H-NMR}$ (CDCl_3): δ 1.99 (s, 3H), 3.78 (br s, 2H), 6.60 (s, 1H), 6.63 (d, $^3\text{J}(\text{H},\text{F})=9.0\text{Hz}$, 1H).

15

Step P7.2: (5-Bromo-3-fluoro-2-methyl-phenyl)-acetic acid

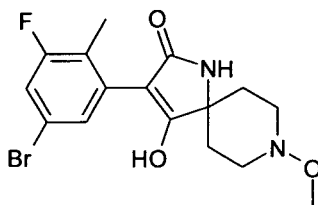


(5-Bromo-3-fluoro-2-methyl-phenyl)-acetic acid was prepared analogously to the synthesis of (5-bromo-4-fluoro-2-methyl-phenyl)-acetic acid (preparation example P5, step P5.3) starting from 5-bromo-3-fluoro-2-methyl-phenylamine (preparation example P7, step P7.1) by making use of the procedures described under step P5.1, step P5.2 and step P5.3. The title acid was obtained as a solid, mp: 147°C.

$^1\text{H-NMR}$ ($\text{d}_6\text{-DMSO}$): δ 2.07 (d, $^4\text{J}(\text{H},\text{F})=2.1\text{Hz}$, 3H), 3.67 (s, 2H), 7.30 (s, 1H), 7.37 (dd, $^3\text{J}(\text{H},\text{F})=9.2\text{Hz}$, $\text{J}=1.9\text{Hz}$, 1H), 12.51 (br s, 1H).

Step P7.3: 3-(5-Bromo-3-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (compound F3)

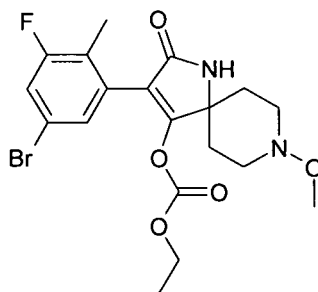
30



3-(5-bromo-3-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (title compound F3) was prepared analogously to the synthesis of 3-(5-bromo-4-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (preparation example P5, step P5.5) starting from (5-bromo-3-fluoro-2-methyl-phenyl)-acetic acid (preparation example P7, step P7.2) by making use of the procedures described under step P5.4 and step P5.5. The title compound F3 was obtained as a solid.

$^1\text{H-NMR}$ (CDCl_3): δ 1.58 (m, 2H), 1.99 (d, $^4J(\text{H},\text{F})=2.0\text{Hz}$, 3H), 2.24 (m, 2H), 2.49 (m, 2H), 3.40 (m, 2H), 3.53 (s, 3H), 6.62 (br s, 1H), 7.06 (dd, 1H), 7.07 (s, 1H).

Step P7.4: Carbonic acid 3-(5-bromo-3-fluoro-2-methyl-phenyl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester ethyl ester (compound E4)



To a solution of 3-(5-bromo-3-fluoro-2-methyl-phenyl)-4-hydroxy-8-methoxy-1,8-diaza-spiro[4.5]dec-3-en-2-one (350 mg) and ethyl chloroformate (77 μl) in acetonitrile (2 ml) was added pyridine (80 μl) and the reaction mixture stirred at room temperature for one hour. The mixture was poured on diluted HCl and extracted with ethyl acetate (3x). The combined organic layers were washed with brine, dried over sodium sulfate and concentrated. The residue was purified by chromatography on silica gel (ethyl acetate/cyclohexane 3:1). Yield: 310 mg of carbonic acid 3-(5-bromo-3-fluoro-2-methyl-phenyl)-8-methoxy-2-oxo-1,8-diaza-spiro[4.5]dec-3-en-4-yl ester ethyl ester (title compound E4) as a solid.

$^1\text{H-NMR}$ (CDCl_3): δ 1.17 (t, 3H), 1.70 (m, 2H), 2.11 (d, $^4J(\text{H},\text{F})=2.1\text{Hz}$, 3H), 2.25 (m, 2H), 2.51 (m, 2H), 3.45 (m, 2H), 3.55 (s, 3H), 4.09 (q, 2H), 6.83 (br s, 1H), 7.11 (s, 1H), 7.18 (dd, $^3J(\text{H},\text{F})=9.0\text{Hz}$, $J=1.9\text{Hz}$, 1H).

Compounds of the formula (I-b) from Tables A, B, C and D can be prepared by analogous procedures.

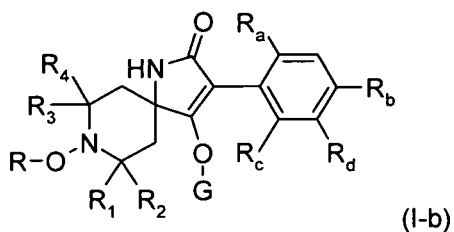


Table A

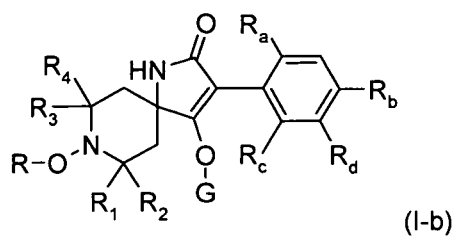
Compounds of formula I-b, wherein R₁, R₂, R₃, and R₄ are H. Cyclo-C3 means cyclopropyl.

	R	G	R _a	R _b	R _c	R _d	Mp.
A1	CH ₃	C(=O)O-CH ₃	CH ₃	H	H	CH ₃	186-188°C
A2	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	CH ₃	170-173°C
A3	CH ₃	C(=O)O-n-C ₃ H ₇	CH ₃	H	H	CH ₃	133-136°C
A4	CH ₃	C(=O)O-i-C ₃ H ₇	CH ₃	H	H	CH ₃	207-209°C
A5	CH ₃	C(=O)O-allyl	CH ₃	H	H	CH ₃	140-142°C
A6	CH ₃	C(=O)O-vinyl	CH ₃	H	H	CH ₃	162-164°C
A7	CH ₃	C(=O)O-propargyl	CH ₃	H	H	CH ₃	151-153°C
A8	CH ₃	C(=O)O-t-C ₄ H ₉	CH ₃	H	H	CH ₃	
A9	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	H	H	CH ₃	158-159°C
A10	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	CH ₃	H	H	CH ₃	
A11	CH ₃	C(=O)NH(C ₂ H ₅)	CH ₃	H	H	CH ₃	
A12	CH ₃	C(=O)N(C ₂ H ₅) ₂	CH ₃	H	H	CH ₃	129-131°C
A13	CH ₃	SO ₂ -i-C ₃ H ₇	CH ₃	H	H	CH ₃	168-170°C
A14	CH ₃	CH ₃	CH ₃	H	H	CH ₃	203-205°C
A15	CH ₃	CH ₂ O-CH ₃	CH ₃	H	H	CH ₃	183-185°C
A16	CH ₃	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	CH ₃	H	H	CH ₃	180-183°C
A17	CH ₃	CH ₂ -(3,4,5-{CH ₃ O} ₃ -C ₆ H ₂)	CH ₃	H	H	CH ₃	128-131°C
A18	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	CH ₃	H	193-195°C
A19	CH ₃	C(=O)O-allyl	CH ₃	CH ₃	CH ₃	H	174-176°C
A20	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	CH ₃	CH ₃	CH ₃	H	220-224°C
A21	CH ₃	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	CH ₃	CH ₃	CH ₃	H	
A22	CH ₃	C(=O)O-C ₂ H ₅	Cl	Cl	H	H	187-189°C
A23	CH ₃	C(=O)O-allyl	Cl	Cl	H	H	

	R	G	R _a	R _b	R _c	R _d	Mp.
A24	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	Cl	Cl	H	H	171-172°C
A25	CH ₃	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	Cl	Cl	H	H	111-113°C
A26	CH ₃	C(=O)O-C ₂ H ₅	Cl	H	Cl	H	229-232°C
A27	CH ₃	C(=O)O-allyl	Cl	H	Cl	H	203-205°C
A28	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	Cl	H	Cl	H	239-242°C
A29	CH ₃	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	Cl	H	Cl	H	
A30	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	Br	205-209°C
A31	CH ₃	C(=O)O-allyl	CH ₃	H	H	Br	157-158°C
A32	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	CH ₃	H	H	Br	197-200°C
A33	CH ₃	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	CH ₃	H	H	Br	
A34	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	4-Cl-C ₆ H ₄	209-211°C
A35	CH ₃	C(=O)O-allyl	CH ₃	H	H	4-Cl-C ₆ H ₄	
A36	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	CH ₃	H	H	4-Cl-C ₆ H ₄	
A37	CH ₃	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	CH ₃	H	H	4-Cl-C ₆ H ₄	
A38	CH ₃ CH ₂	C(=O)O-C ₂ H ₅	CH ₃	H	H	CH ₃	167-168°C
A39	CH ₃ CH ₂	C(=O)O-allyl	CH ₃	H	H	CH ₃	
A40	CH ₃ CH ₂	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	CH ₃	H	H	CH ₃	
A41	CH ₃ CH ₂	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	CH ₃	H	H	CH ₃	
A42	CH ₃ CH ₂	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	CH ₃	H	174-175°C
A43	CH ₃ CH ₂	C(=O)O-allyl	CH ₃	CH ₃	CH ₃	H	
A44	CH ₃ CH ₂	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	CH ₃	CH ₃	CH ₃	H	
A45	CH ₃ CH ₂	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	CH ₃	CH ₃	CH ₃	H	
A46	CH ₃ CH ₂	C(=O)O-C ₂ H ₅	Cl	Cl	H	H	183-184°C
A47	CH ₃ CH ₂	C(=O)O-allyl	Cl	Cl	H	H	
A48	CH ₃ CH ₂	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	Cl	Cl	H	H	
A49	CH ₃ CH ₂	CH ₂ -(4-CH ₃ O-C ₆ H ₄)	Cl	Cl	H	H	
A50	CH ₃	C(=O)O-CH ₂ C(CH ₃) ₃	CH ₃	H	H	CH ₃	196-197°C
A51	CH ₃	C(=O)-CH ₃	CH ₃	H	H	CH ₃	233-236°C
A52	CH ₃	C(=O)-CH ₂ CH ₃	CH ₃	H	H	CH ₃	202-204°C
A53	CH ₃ CH ₂	C(=O)-t-C ₄ H ₉	CH ₃	H	H	CH ₃	169-170°C
A54	CH ₃ CH ₂	C(=O)-t-C ₄ H ₉	CH ₃	CH ₃	CH ₃	H	230-231°C
A55	CH ₃ CH ₂	C(=O)-t-C ₄ H ₉	Cl	Cl	H	H	165-166°C
A56	CH ₃	C(=O)O-C ₂ H ₅	H	Cl	H	Cl	222-224°C
A57	CH ₃	C(=O)O-C ₂ H ₅	OCH ₃	H	H	OCH ₃	192-193°C
A58	CH ₃	CH ₂ CH ₂ O-CH ₃	CH ₃	H	H	CH ₃	161-164°C
A59	CH ₃	C(=O)-i-C ₃ H ₇	CH ₃	H	H	CH ₃	201-203°C

	R	G	R _a	R _b	R _c	R _d	Mp.
A60	CH ₃	CH ₂ CN	CH ₃	H	H	CH ₃	104-106°C
A61	CH ₃	CH ₂ O-CH ₂ CH ₃	CH ₃	H	H	CH ₃	150-152°C
A62	CH ₃	allyl	CH ₃	H	H	CH ₃	173-174°C
A63	CH ₃	propargyl	CH ₃	H	H	CH ₃	184-185°C
A64	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	H	Cl	H	Cl	56-59°C
A65	CH ₃	C(=O)N(CH ₃)(CH ₂ CH ₂ CN)	OCH ₃	H	H	OCH ₃	187-189°C
A66	CH ₃	C(=O)O-allyl	H	Cl	H	Cl	210-213°C
A67	CH ₃	C(=O)O-allyl	OCH ₃	H	H	OCH ₃	156-157°C
A68	CH ₃	C(=O)O-allyl	CH ₃	Br	CH ₃	H	207-211°C
A69	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	Br	CH ₃	H	185-187°C
A70	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	cyclo-C3	CH ₃	H	215-216°C
A71	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	H	Br	206-209°C
A72	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	CH ₃	H	Br	225-226°C
A73	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	H	4-Cl-C ₆ H ₄	211-213°C
A74	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	CH ₃	H	4-Cl-C ₆ H ₄	229-230°C
A75	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	H	cyclo-C3	174-176°C
A76	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	CH ₃	H	cyclo-C3	218-219°C
A77	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	cyclo-C3	CH ₃	H	>200°C
A78	CH ₃	C(=O)O-C ₂ H ₅	Cl	cyclo-C3	H	H	190-192°C
A79	CH ₃	C(=O)-t-C ₄ H ₉	Cl	cyclo-C3	H	H	195-197°C
A80	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	cyclo-C3	H	H	193-195°C
A81	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	cyclo-C3	H	H	244-245°C
A82	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	F	H	4-Cl-C ₆ H ₄	150-151°C
A83	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	F	H	Br	216-217°C
A84	CH ₃	C(=O)O-C ₂ H ₅	Cl	Br	H	H	199-201°C
A85	CH ₃	C(=O)-t-C ₄ H ₉	Cl	Br	H	H	223-225°C
A86	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	Br	H	H	201-203°C
A87	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	Br	H	H	239-240°C
A88	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	4-Cl-C ₆ H ₄	CH ₃	H	217-219°C
A89	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	4-Cl-C ₆ H ₄	CH ₃	H	>250°C
A90	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	F	H	4-F-C ₆ H ₄	101-104°C

Table BCompounds of formula I-b, wherein R₁, R₂, R₃, and R₄ are CH₃.

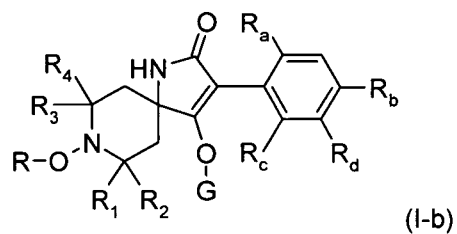


	R	G	R _a	R _b	R _c	R _d	Phys. data
B1	CH ₃	C(=O)O-CH ₃	CH ₃	H	H	CH ₃	solid, LC-MS: 417 (M+H) ⁺
B2	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	CH ₃	
B3	CH ₃	C(=O)O-n-C ₃ H ₇	CH ₃	H	H	CH ₃	
B4	CH ₃	C(=O)O-i-C ₃ H ₇	CH ₃	H	H	CH ₃	
B5	CH ₃	C(=O)O-allyl	CH ₃	H	H	CH ₃	solid, LC-MS: 443 (M+H) ⁺
B6	CH ₃	C(=O)O-vinyl	CH ₃	H	H	CH ₃	
B7	CH ₃	C(=O)O-propargyl	CH ₃	H	H	CH ₃	
B8	CH ₃	C(=O)O-t-C ₄ H ₉	CH ₃	H	H	CH ₃	
B9	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	H	H	CH ₃	
B10	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	CH ₃	H	
B11	CH ₃	C(=O)O-allyl	CH ₃	CH ₃	CH ₃	H	
B12	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	CH ₃	CH ₃	H	
B13	CH ₃ CH ₂	C(=O)O-C ₂ H ₅	CH ₃	H	H	CH ₃	
B14	CH ₃ CH ₂	C(=O)O-C ₂ H ₅	CH ₃	CH ₃	CH ₃	H	

5 Table C

Compounds of formula I-b, wherein R₁, R₂, R₃, and R₄ are H, G is hydrogen.

(Cyclo-C3 means cyclopropyl.)

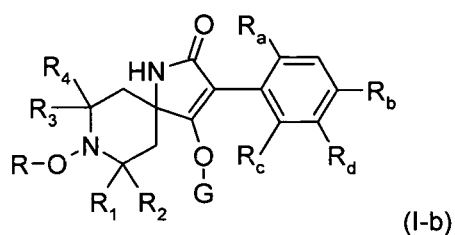


	R	G	R _a	R _b	R _c	R _d	Phys. data
C1	CH ₃	H	CH ₃	H	H	CH ₃	176-177°C
C2	CH ₃	H	CH ₃	CH ₃	CH ₃	H	253-255°C
C3	CH ₃	H	Cl	Cl	H	H	221-223°C

	R	G	R _a	R _b	R _c	R _d	Phys. data
C4	CH ₃	H	Cl	H	Cl	H	257-260°C
C5	CH ₃	H	CH ₃	H	H	Br	194-196°C
C6	CH ₃	H	CH ₃	H	H	4-Cl-C ₆ H ₄	solid, LC-MS: 399/401 (M+H) ⁺
C7	CH ₃ CH ₂	H	CH ₃	H	H	CH ₃	166-167°C
C8	CH ₃ CH ₂	H	CH ₃	CH ₃	CH ₃	H	>250°C
C9	CH ₃ CH ₂	H	Cl	Cl	H	H	244-245°C
C10	CH ₃	H	H	Cl	H	Cl	239-242°C
C11	CH ₃	H	OCH ₃	H	H	OCH ₃	194-196°C
C12	CH ₃	H	CH ₃	Br	CH ₃	H	260-264°C
C13	CH ₃	H	CH ₃	cyclo-C3	CH ₃	H	>250°C
C14	CH ₃	H	CH ₃	CH ₃	H	Br	solid, LC-MS: 381/383 (M+H) ⁺
C15	CH ₃	H	CH ₃	CH ₃	H	4-Cl-C ₆ H ₄	168-169°C
C16	CH ₃	H	CH ₃	CH ₃	H	cyclo-C3	wax, LC-MS: 343 (M+H) ⁺
C17	CH ₃	H	Cl	cyclo-C3	H	H	206-207°C
C18	CH ₃	H	CH ₃	cyclo-C3	H	H	191-194°C
C19	CH ₃	H	CH ₃	F	H	Br	229-230°C
C20	CH ₃	H	CH ₃	CH ₃	CH ₃	Cl	>200°C
C21	CH ₃	H	Cl	Br	H	H	167-169°C
C22	CH ₃	H	CH ₃	Br	H	H	187-189°C
C23	CH ₃	H	CH ₃	4-Cl-C ₆ H ₄	CH ₃	H	255-257°C
C24	CH ₃	H	CH ₃	F	H	4-Cl-C ₆ H ₄	MS: 417/419 (M+H) ⁺
C25	CH ₃	H	CH ₃	F	H	4-F-C ₆ H ₄	

Table D

Compounds of formula I-b, wherein R₁, R₂, R₃, and R₄ are CH₃, G is hydrogen.



	R	G	R _a	R _b	R _c	R _d	Phys. data
D1	CH ₃	H	CH ₃	H	H	CH ₃	223-225°C

	R	G	R _a	R _b	R _c	R _d	Phys. data
D2	CH ₃ CH ₂	H	CH ₃	H	H	CH ₃	
D3	CH ₃	H	CH ₃	CH ₃	CH ₃	H	
D4	CH ₃ CH ₂	H	CH ₃	CH ₃	CH ₃	H	

Compounds of the formula (I-c) from Tables E and F can be prepared by analogous procedures.

5

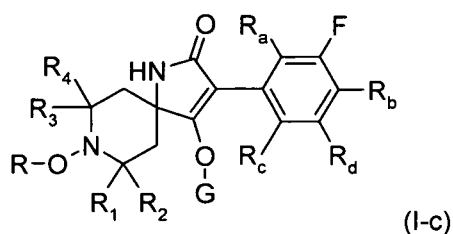


Table E

Compounds of formula I-c, wherein R₁, R₂, R₃, and R₄ are H.

10

(Cyclo-C3 means cyclopropyl.)

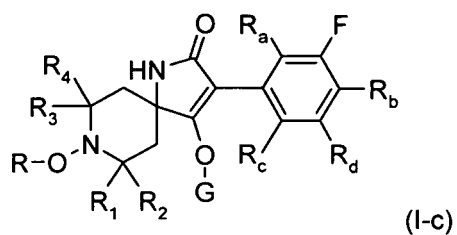
	R	G	R _a	R _b	R _c	R _d	Phys. data
E1	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	4-F-C ₆ H ₄	gum
E2	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	H	H	cyclo-C3	gum
E3	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	H	H	4-F-C ₆ H ₄	gum
E4	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	Br	solid, 1H-NMR see preparation example P7, step P7.4
E5	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	CH ₃	
E6	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	4-Cl-C ₆ H ₄	
E7	CH ₃	C(=O)O-C ₂ H ₅	CH ₃	H	H	cyclo-C3	
E8	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	H	H	4-Cl-C ₆ H ₄	
E9	CH ₃	C(=O)-t-C ₄ H ₉	CH ₃	H	H	Br	

Table F

15

Compounds of formula I-c, wherein R₁, R₂, R₃, and R₄ are H, G is hydrogen.

(Cyclo-C3 means cyclopropyl.)

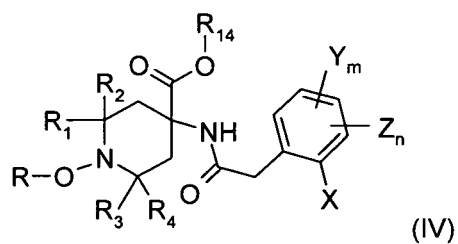


	R	G	R _a	R _b	R _c	R _d	Phys. Data
F1	CH ₃	H	CH ₃	H	H	4-Cl-C ₆ H ₄	solid
F2	CH ₃	H	CH ₃	H	H	cyclo-C3	solid
F3	CH ₃	H	CH ₃	H	H	Br	solid, ¹ H-NMR see preparation example P7, step P7.3
F4	CH ₃	H	CH ₃	H	H	CH ₃	
F5	CH ₃	H	CH ₃	H	H	4-F-C ₆ H ₄	solid
F6	CH ₃ CH ₂	H	CH ₃	H	H	4-Cl-C ₆ H ₄	

- 5 Intermediates: compounds of the formula (IV), (V), (VII), (VIII) and (XI) from Tables G, H and J can be prepared by analogous procedures.

Table G

Compounds of formula IV



10

Structure	Mp. or MS / NMR
	Preparation example P1, step P1.4
	Preparation example P2, step P2.3

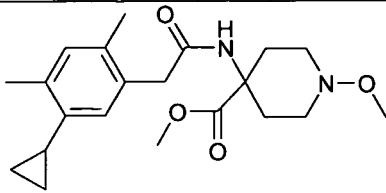
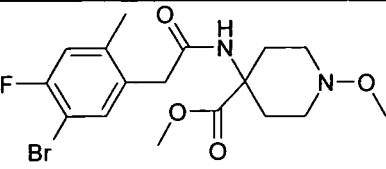
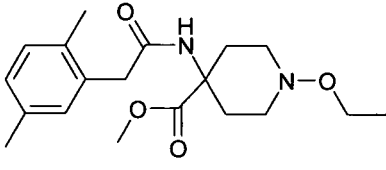
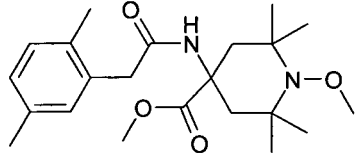
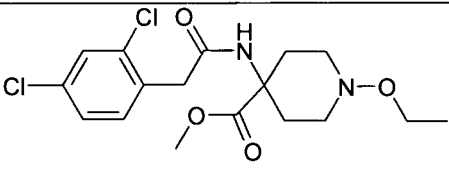
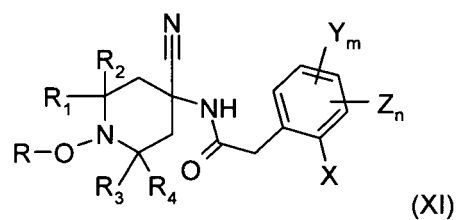
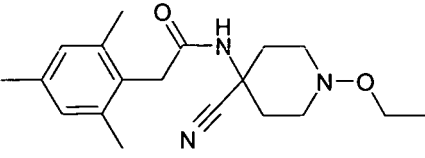
Structure	Mp. or MS / NMR
	Preparation example P4, step P4.4
	Preparation example P5, step P5.4
	132-133°C
	solid, LC-MS: 391 (M+H) ⁺
	166-167°C

Table H

Compounds of formula XI



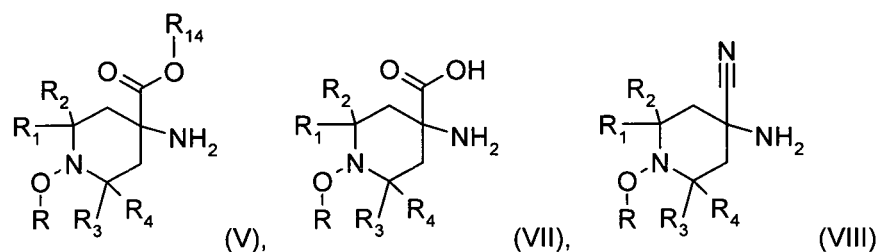
5

Structure	Mp. or MS / NMR
	Preparation example P2, step P2.2

Structure	Mp. or MS / NMR
	219-220°C
	208-210°C
	159-160°C
	169-170°C

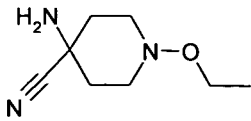
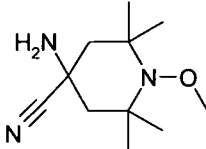
Table J

Compounds of formula V, VII or VIII



5

Structure	Mp. or MS / NMR
	Preparation example P1, step P1.1
	Preparation example P1, step P1.2 and preparation example P3, step P3.2
	Preparation example P1, step P1.3

Structure	Mp. or MS / NMR
	Preparation example P2, step P2.1
	¹ H-NMR (d ₆ -DMSO, major isomer): 1.14 (s, 6H), 1.29 (s, 6H), 1.51 (d, 2H), 2.00 (d, 2H), 2.64 (br s, 2H), 3.55 (s, 3H); LC-MS (EI, ES ⁺): 212 (M+H) ⁺

EXAMPLE 2

5 Formulation examples (% = percent by weight)

Example F1: Emulsion concentrates

	a)	b)	c)
Active ingredient	25 %	40 %	50 %
Calcium dodecylbenzenesulfonate	5 %	8 %	6 %
Castor oil polyethylene glycol ether (36 mol of EO)	5 %	-	-
10 Tributylphenoxypolyethylene glycol ether (30 mol of EO)	-	12 %	4 %
Cyclohexanone	-	15 %	20 %
Xylene mixture	65 %	25 %	20 %

Emulsions of any desired concentration can be prepared from such concentrates by dilution
 15 with water.

Example F2: Solutions

	a)	b)	c)	d)
Active ingredient	80 %	10 %	5 %	95 %
Ethylene glycol monomethyl ether	20 %	-	-	-
20 Polyethylene glycol MW 400	-	70 %	-	-
N-Methylpyrrolid-2-one	-	20 %	-	-
Epoxidized coconut oil	-	-	1 %	5 %
Petroleum ether (boiling range: 160-190°)	-	-	94 %	-

25 The solutions are suitable for use in the form of microdrops.

Example F3: Granules

a)	b)	c)	d)
----	----	----	----

Active ingredient	5 %	10 %	8 %	21 %
Kaolin	94 %	-	79 %	54 %
Highly disperse silica	1 %	-	13 %	7 %
Attapulgate	-	90 %	-	18 %

5

The active ingredient is dissolved in dichloromethane, the solution is sprayed onto the carrier(s), and the solvent is subsequently evaporated in vacuo.

Example F4: Dusts

	a)	b)
10 Active ingredient	2 %	5 %
Highly disperse silica	1 %	5 %
Talc	97 %	-
Kaolin	-	90 %

15 Ready-to-use dusts are obtained by intimately mixing the carriers and the active ingredient.

Example F5: Wettable powders

	a)	b)	c)
Active ingredient	25 %	50 %	75 %
Sodium lignosulfonate	5 %	5 %	-
20 Sodium lauryl sulfate	3 %	-	5 %
Sodium diisobutylphenathalenesulfonate	-	6 %	10 %
Octylphenoxypolyethylene glycol ether (7-8 mol of EO)	-	2 %	-
Highly disperse silica	5 %	10 %	10 %
25 Kaolin	62 %	27 %	-

The active ingredient is mixed with the additives and the mixture is ground thoroughly in a suitable mill. This gives wettable powders, which can be diluted with water to give suspensions of any desired concentration.

30

Example F6: Extruder granules

Active ingredient	10 %
Sodium lignosulfonate	2 %
Carboxymethylcellulose	1 %
35 Kaolin	87 %

The active ingredient is mixed with the additives, and the mixture is ground, moistened with water, extruded, granulated and dried in a stream of air.

Example F7: Coated granules

5	Active ingredient	3 %
	Polyethylene glycol (MW 200)	3 %
	Kaolin	94 %

10 In a mixer, the finely ground active ingredient is applied uniformly to the kaolin, which has been moistened with the polyethylene glycol. This gives dust-free coated granules.

Example F8: Suspension concentrate

	Active ingredient	40 %
	Ethylene glycol	10 %
15	Nonylphenoxypolyethylene glycol ether (15 mol of EO)	6 %
	Sodium lignosulfonate	10 %
	Carboxymethylcellulose	1 %
	37 % aqueous formaldehyde solution	0.2 %
	Silicone oil (75 % aqueous emulsion)	0.8 %
20	Water	32 %

The finely ground active ingredient is mixed intimately with the additives. Suspensions of any desired concentration can be prepared from the thus resulting suspension concentrate by dilution with water.

25

Example F9: Powders for dry seed treatment

	a)	b)	c)
active ingredient	25 %	50 %	75 %
light mineral oil	5 %	5 %	5 %
highly dispersed silicic acid	5 %	5 %	-
Kaolin	65 %	40 %	-
Talcum	-		20 %

The active ingredient is thoroughly mixed with the adjuvants and the mixture is thoroughly ground in a suitable mill, affording powders that can be used directly for seed treatment.

Example F10: Flowable concentrate for seed treatment

active ingredient	40 %
propylene glycol	5 %
copolymer butanol PO/EO	2 %
tristyrenephenol with 10-20 moles EO	2 %
1,2-benzisothiazolin-3-one (in the form of a 20% solution in water)	0.5 %
monoazo-pigment calcium salt	5 %
Silicone oil (in the form of a 75 % emulsion in water)	0.2 %
Water	45.3 %

The finely ground active ingredient is intimately mixed with the adjuvants, giving a suspension concentrate from which suspensions of any desired dilution can be obtained by dilution with water. Using such dilutions, living plants as well as plant propagation material can be treated and protected against infestation by microorganisms, by spraying, pouring or immersion.

EXAMPLE 3

This Example illustrates the pesticidal/insecticidal properties of compounds of formula I.

Example B1: Activity against *Myzus persicae* (green peach aphid)

(mixed population, feeding/residual contact activity, preventive)

Sunflower leaf discs are placed on agar in a 24-well microtiter plate and sprayed with test solutions. After drying, the leaf discs are infested with an aphid population of mixed ages. After an incubation period of 6 days, samples are checked for mortality and special effects (e.g. phytotoxicity).

In this test, compounds listed in the Tables A, B, C, D, E and F above show good activity. In particular compounds F1, F2, C1, C2, C3, C5, C6, C7, C8, C9, C12, C13, C14, C15, C16, C17, C18, C20, C21, C22, C23, A1, A2, A3, A4, A5, A6, A7, A9, A12, A15, A18, A19, A22, A30, A31, A34, A38, A42, A46, A50, A51, A52, A55, A59, A61, A68, A69, A70, A71, A75, A78, A86 and E1 show an activity of over 80% at a concentration of 400ppm.

Example B2: Activity against *Myzus persicae* (green peach aphid)

(mixed population, systemic/feeding activity, curative)

Roots of pea seedlings, infested with an aphid population of mixed ages, are placed directly in the test solutions. 6 days after introduction, samples are checked for mortality and special effects on the plant.

In this test, compounds listed in the Tables A, B, C, D, E and F above show good activity. In particular compounds F1, C1, C2, C3, C7, C8, C9, C12, C13, C20, C21, A1, A2, A3, A4, A5, A7, A9, A18, A20, A24, A22, A38, A42, A46, A51, A52, A53, A54, A55, A59, A61, A68, A69, A70, A77, A78, A80, A84 and A86 show an activity of over 80% at a concentration of 400ppm.

10 Example B3: Activity against *Tetranychus urticae* (two-spotted spider mite)

(mixed population, feeding/residual contact activity, preventive)

Bean leaf discs on agar in 24-well microtiter plates are sprayed with test solutions. After drying, the leaf discs are infested with mite populations of mixed ages. 8 days later, discs are checked for egg mortality, larval mortality, and adult mortality.

15 In this test, compounds listed in the Tables A, B, C, D, E and F above show good activity. In particular compounds C1, C2, C12, C15, C16, C20, C21, C23, A1, A2, A3, A4, A5, A6, A7, A9, A15, A19, A50, A58, A59, A69, A80, A82, A84, A85, A86, A87, A90, E1, E2 and E3 show an activity of over 80% at a concentration of 400ppm.

20 Example B4: Activity against *Thrips tabaci* (onion Thrips)

(mixed population, feeding/residual contact activity, preventive)

Sunflower leaf discs are placed on agar in a 24-well microtiter plate and sprayed with test solutions. After drying, the leaf discs are infested with a thrips population of mixed ages. After an incubation period of 6 days, samples are checked for mortality and special effects (e.g. phytotoxicity).

25 In this test, compounds listed in the Tables A, B, C, D, E and F above show good activity. In particular compounds F1, C1, C2, C3, C6, C8, C9, C14, C15, C17, C20, C21, C22, A3, A6, A7, A12, A16, A17, A18, A19, A22, A25, A26, A34, A38, A42, A46, A51, A61, A73, A75, A84, A90, E1 and E3 show an activity of over 80% at a concentration of 400ppm.

30

Example B5: Activity against *Plutella xylostella* (Diamond back moth)

(larvicide, feeding/residual contact activity, preventive)

24-well microtiter plate (MTP) with artificial diet is treated with test solutions by pipetting. After drying, the MTP's are infested with larvae (L2)(10-15 per well). After an incubation period of 5 days, samples are checked for larval mortality, antifeedant and growth regulation.

35

In this test, compounds listed in the Tables A, B, C, D, E and F above show good activity. In particular compounds C2, C8, A6, A7, A19, A34, A42, A50 and A52 show an activity of over 80% at a concentration of 400ppm.

5 Example B6: Activity against *Diabrotica balteata* (Corn root worm)

(larvicide, feeding/residual contact activity, preventive)

24-well microtiter plate (MTP) with artificial diet is treated with test solutions by pipetting.

After drying, the MTP's are infested with larvae (L2)(6-10 per well). After an incubation period of 5 days, samples are checked for larval mortality, antifeedant and growth

10 regulation.

In this test, compounds listed in the Tables A, B, C, D, E and F above show good activity. In particular compounds C6, C8, A34 and A52 show an activity of over 80% at a concentration of 400ppm.

15 Example B7: Activity against *Frankliniella occidentalis* (western flower thrips)

Bean leaf discs on agar in petri dishes or bean plants in a spray chamber are treated with diluted test solutions. After drying leaf discs are cut and placed in plastic cups on the surface of an agar layer and infested with mixed population. 6 days (leaf discs) or 14 days (plants) after the infestation, samples are checked for reduction of treated population and

20 compared to the non treated population.

In this test, compounds listed in Table A, B, C, D, E and F above show good activity. For example compounds C1, C6, A2, A3, A4, A5, A42, A70, A80 and E1 show an activity of over 80% at a concentration of 400 ppm.

25 Example B8: Activity against *Bemisia tabaci* (tobacco white fly)

(larvicide, contact/feeding)

Bean plants are infested with 20-30 adults that were removed after a 4 day egg-laying period. After another 7 days, bean plants with hatched nymphs (N-2) are treated (2 replicates) with the test solutions in a spray chamber. Three weeks later, samples are

30 checked for number of emerged adults. Efficacy was calculated by comparing number of emerged adults in treated and non treated samples.

In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C1, A1, A2, A3, A4, A5, A78, A80 and A81 show an activity of over 80% at a concentration of 400 ppm.

35

Example B9: Activity against *Nilaparvata lugens* (brown rice planthopper)

(larvicide, feeding/contact)

Rice seedlings are treated with the diluted test solutions in a spray chamber. After drying, they are infested with 20 N₃ nymphs (2 replicates). 6-12 days after the treatment samples are checked for mortality, growth regulation, and effects on the F₁ generation.

In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C2, C3, C6, C8, A1, A2, A3, A4, A5, A7, A9, A12, A18, A22, A34, A42, A70, A77, A78, A80, A81 and E1 show an activity of over 80% at a concentration of 400 ppm.

Example B10: Activity against *Aphis craccivora* (pea aphid)

(mixed population, contact/feeding)

Pea seedlings, infested with an aphid population of mixed ages, are treated (2 replicates) with diluted test solutions in a spray chamber. 6 days after treatment, samples are checked for mortality.

In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C2, C8, A2, A5, A15 and A42, show an activity of over 80% at a concentration of 400 ppm.

Example B11: Activity against *Aphis craccivora* (pea aphid)

(mixed population, systemic/feeding)

Roots of pea seedlings, infested with an aphid population of mixed ages, are placed (2 replicates) directly in the test solution. 6 days later, samples are checked for mortality.

In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C1, C2, A1, A2, A3, A4, A5, A6, A7, A9, A18, A22 and A42 show an activity of over 80% at a concentration of 400 ppm.

Example B12: Activity against *Aphis gossypii* (cotton aphid)

(mixed population, contact/feeding)

Pea seedlings, infested with an aphid population of mixed ages, are treated (2 replicates) with diluted test solutions in a spray chamber. 6 days after treatment, samples are checked for mortality.

In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C2, C6, C8, A2, A5 and A18 show an activity of over 80% at a concentration of 400 ppm.

Example B13: Activity against *Aphis gossypii* (cotton aphid)

(mixed population, systemic/feeding)

Roots of pea seedlings, infested with an aphid population of mixed ages, are placed (2 replicates) directly in the test solution. 6 days later, samples are checked for mortality.

- 5 In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C1, C2, C3, C8, A1, A2, A3, A4, A5, A7, A9, A18, A22, A42, A50, A70 and A80 show an activity of over 80% at a concentration of 400 ppm.

Example B14: Translaminar activity against *Aphis craccivora* (ground nut aphid)

- 10 French bean leaves (*Phaseolus vulgaris*) are infested with about 20 mixed age individuals on the lower leaf side using clip cages. 1 day after the infestation, the upper side of the leaves is treated with the test solution by painting. 5 days later, samples are checked for mortality.

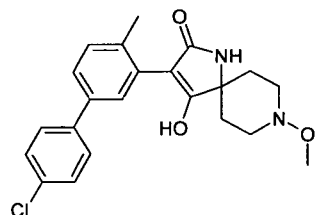
- 15 In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C2, C6, C8, A2, A3, A5, A7, A18 and E1 show an activity of over 80% at a concentration of 400 ppm.

Example B15: Activity against *Aonidiella aurantii* (red scale)

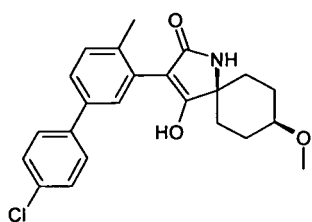
- 20 Treatment of potato tubers by dipping the in the test solution. One day later, tubers are infested with about 50 crawlers. 6-8 weeks after application samples are checked for the number of crawlers of the next generation (compared to the non treated samples).

In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C1, A1, A2, A3, A4 and A5 show an activity of over 80% at a concentration of 400 ppm.

- 25 Example B16 to B17: Comparison of the insecticidal activity of compounds according to the invention with the structurally most closely comparable compound from the state of the art (compound No. I-1-a-16 described on page 157 of WO1999/48869):



(Compound No. C6 according to the invention)



(Compound No. I-1-a-16 according to state of the art)

Example B16: Activity against *Myzus persicae* (green peach aphid)

(mixed population, feeding/residual contact activity, preventive)

- 5 Sunflower leaf discs are placed on agar in a 24-well microtiter plate and sprayed with test solutions. After drying, the leaf discs are infested with an aphid population of mixed ages. After an incubation period of 6 days, samples are checked for mortality.

Results are shown in Table B16:

10

Table B16: Activity against *Myzus persicae* (green peach aphid)

Compound:	Concentration (ppm)	Death rate (%) after 6 days
Comp. I-1-a-16 (state of the art)	50	50
Comp. I-1-a-16 (state of the art)	12.5	0
Comp. C6 (invention)	50	90
Comp. C6 (invention)	12.5	25

- 15 Table B16 shows that compound No. C6 according to the invention exerts a substantially better insecticidal action on *Myzus persicae* than the compound from the state of the art. This enhanced effect was not to be expected on the basis of the structural similarity of these compounds.

Example B17: Activity against *Diabrotica balteata* (Corn root worm)

- 20 (larvicide, feeding/residual contact activity, preventive)

24-well microtiter plate (MTP) with artificial diet is treated with test solutions by pipetting. After drying, the MTP's are infested with larvae (L2)(6-10 per well). After an incubation period of 5 days, samples are checked for growth regulation.

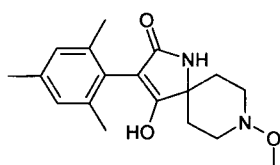
- 25 Results are shown in Table B17:

Table B17: Activity against *Diabrotica balteata* (Corn root worm)

Compound:	Concentration (ppm)	Inhibition of growth (%) after 5 days
Comp. I-1-a-16 (state of the art)	200	0
Comp. I-1-a-16 (state of the art)	50	0
Comp. C6 (invention)	200	75
Comp. C6 (invention)	50	50

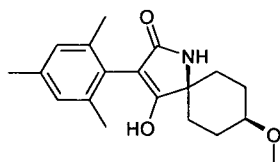
5 Table B17 shows that compound C6 according to the invention exerts a substantially better insecticidal action on *Diabrotica balteata* than the compound from the state of the art. This enhanced effect was not to be expected on the basis of the structural similarity of these compounds.

10 Example B18: Comparison of the insecticidal activity of compounds according to the invention with the structurally most closely comparable compound from the state of the art (compound No. Ia-32 described on page 66 of EP596298):



(Compound No. C2 according to the invention)

15



(Compound No. Ia-32 according to state of the art)

Example B18: Activity against *Thrips tabaci* (onion Thrips)

(mixed population, feeding/residual contact activity, preventive)

20 Sunflower leaf discs are placed on agar in a 24-well microtiter plate and sprayed with test solutions. After drying, the leaf discs are infested with a thrips population of mixed ages. After an incubation period of 6 days, samples are checked for mortality.

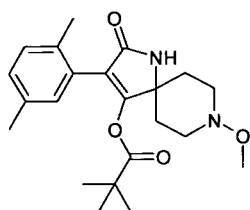
Results are shown in Table B18:

Table B18: Activity against *Thrips tabaci* (onion Thrips)

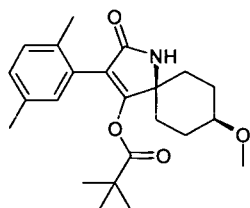
Compound:	Concentration (ppm)	Death rate (%) after 6 days
Comp. Ia-32 (state of the art)	200	25
Comp. Ia-32 (state of the art)	50	25
Comp. Ia-32 (state of the art)	12.5	25
Comp. C2 (invention)	200	100
Comp. C2 (invention)	50	100
Comp. C2 (invention)	12.5	90

Table B18 shows that compound No. C2 according to the invention exerts a substantially better insecticidal action on *Thrips tabaci* than the compound from the state of the art. This enhanced effect was not to be expected on the basis of the structural similarity of these compounds.

Example B19 to B20: Comparison of the insecticidal activity of compounds according to the invention with the structurally most closely comparable compound from the state of the art (compound I-1-b-20 described on page 94 of WO1998/05638):



(Compound No. A9 according to the invention)



(Compound No. I-1-b-20 according to state of the art)

Example B19: Activity against *Myzus persicae* (green peach aphid)

(mixed population, systemic/feeding activity, curative)

Roots of pea seedlings, infested with an aphid population of mixed ages, are placed directly in the test solutions. 6 days after introduction, samples are checked for mortality.

Results are shown in Table B19:

5

Table B19: Activity against *Myzus persicae* (green peach aphid)

Compound:	Concentration (ppm)	Death rate (%) after 6 days (systemic action)
Comp. I-1-b-20 (state of the art)	200	0
Comp. I-1-b-20 (state of the art)	50	not tested
Comp. A9 (invention)	200	80
Comp. A9 (invention)	50	50

Table B19 shows that compound No. A9 according to the invention exerts a substantially better insecticidal action on *Myzus persicae* than the compound from the state of the art. This enhanced effect was not to be expected on the basis of the structural similarity of these compounds.

Example B20: Activity against *Tetranychus urticae* (two-spotted spider mite)

(mixed population, feeding/residual contact activity, preventive)

Bean leaf discs on agar in 24-well microtiter plates are sprayed with test solutions. After drying, the leaf discs are infested with mite populations of mixed ages. 8 days later, discs are checked for egg mortality, larval mortality, and adult mortality.

Results are shown in Table B20:

Table B20: Activity against *Tetranychus urticae* (two-spotted spider mite)

Compound:	Concentration (ppm)	Death rate (%) after 8 days
Comp. I-1-b-20 (state of the art)	200	50
Comp. I-1-b-20 (state of the art)	50	0
Comp. I-1-b-20 (state of the art)	12.5	0

Comp. A9 (invention)	200	80
Comp. A9 (invention)	50	80
Comp. A9 (invention)	12.5	50

Table B20 shows that compound A9 according to the invention exerts a substantially better insecticidal action on *Tetranychus urticae* than the compound from the state of the art. Especially at low application rates (50 and 12.5 ppm) the compound according to the invention is far superior to the compound of the state of the art. This enhanced effect was not to be expected on the basis of the structural similarity of these compounds.

Example B21: Insecticide seed treatment

Sugarbeet seeds are treated with test compounds at various rates (mg a.i. /seed). The seeds are sown into pots filled with soil. After placing the seeds on the soil surface they are further covered with soil. Before and during the bioassays, the plants are grown under optimal greenhouse conditions. After 14 days, the plants are infested with African bean aphid (*Aphis craccivora*) by placing one infested pea seedling containing approximately 150 aphids of a population of mixed developmental stages onto each plant. Seven days after infestation, the number of aphids is counted per plant. Each treatment group is replicated 10 times. Each replicate contains 1 plant. In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. The table below shows as an example the reduction of aphids compared to the untreated control plants.

Table B21: Reduction of African bean aphid exposed to sugarbeet plants grown from seeds treated with compounds C1 or A5

mg a.i. /seed	Efficacy, % Comp. C1	Efficacy, % Comp. A5
0.03	65	37
0.1	96	97
0.3	99	99
0.6	99	100

Example B22: Insecticide seed treatment

Chinese cabbage seeds are treated with test compounds at various rates (mg a.i. /seed). The seeds are sown into pots filled with soil. After placing the seeds on the soil surface they

- are further covered with soil. Before and during the bioassays, the plants are grown under optimal greenhouse conditions. After 14 days, the plants are infested with cabbage aphid (*Myzus persicae*) by placing one infested pea seedling containing approximately 150 aphids of a population of mixed developmental stages onto each plant. Seven days after
- 5 infestation, the number of aphids is counted per plant. Each treatment group is replicated 10 times. Each replicate contains 1 plant. In this test, compounds listed in Tables A, B, C, D, E and F above show good activity.

Example B23: Activity against *Meloidogyne incognita*

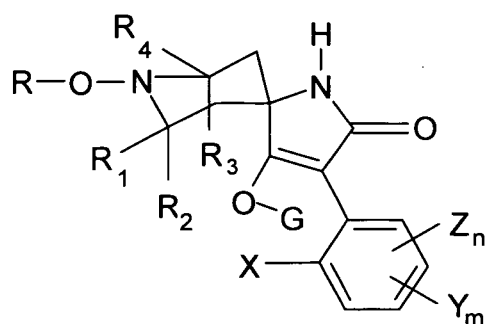
- 10 (pouch test, contact)

Cucumber seeds are sown into each pouch which is treated at the same time with test solutions and nematode egg solutions. 12 days after treatment, samples are checked for gall reduction in the root mass. Efficacy was calculated by comparing formation of galls in treated and untreated samples.

- 15 In this test, compounds listed in Tables A, B, C, D, E and F above show good activity. For example compounds C1, A4, A5, A6, A15 and A80 show an activity of over 80% at a concentration of 400 ppm.

CLAIMS

1. Compounds of the formula I



(I) ,

wherein

X, Y and Z, independently of each other, are C₁₋₄alkyl, C₃₋₆cycloalkyl, C₁₋₄haloalkyl, C₁₋₄alkoxy, halogen, phenyl or phenyl substituted by C₁₋₄alkyl or halogen, C₁₋₄haloalkyl, halogen or cyano,

m and n, independently of each other, are 0, 1, 2 or 3, where m+n is 0, 1, 2 or 3,

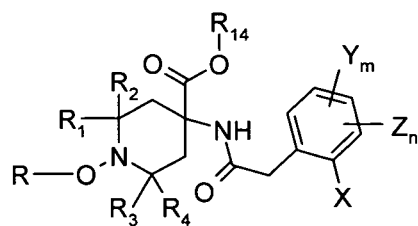
G is hydrogen, a metal, ammonium, sulfonium or latentating group,

R is hydrogen, C₁₋₆alkyl, C₁₋₆haloalkyl, C₁₋₆cyanoalkyl, benzyl, C₁₋₄alkoxy(C₁₋₄)alkyl, C₁₋₄alkoxy(C₁₋₄)alkoxy(C₁₋₄)alkyl or a group selected from G, and

R₁, R₂, R₃ and R₄, independently of each other, are hydrogen or methyl, or

an agrochemically acceptable salt or an N-oxide thereof.

2. A process for the preparation of the compounds of the formula I according to claim 1, wherein G is hydrogen, which comprises cyclisation of the compound of formula IV



(IV),

wherein X, Y, Z, m, n, R, R₁, R₂, R₃, and R₄ have the meanings assigned to them in claim 1, and R₁₄ is C₁₋₄alkyl, under basic conditions.

3. A pesticidal composition comprising a pesticidal effective amount of at least one compound of formula I according to claim 1.

4. A pesticidal composition according to claim 3, which, in addition to comprising the compound of formula I, comprises formulation adjuvants.

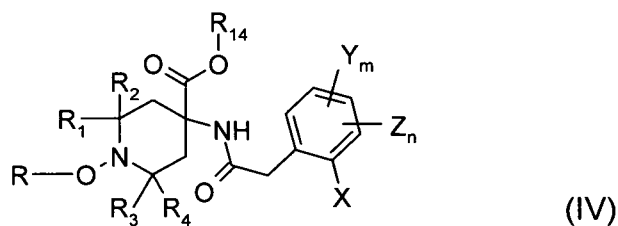
5. A pesticidal composition according to claim 3, which, in addition to comprising the compound of formula I, comprises at least one additional insecticide, acaricide, nematocide or molluscicide.

6. A pesticidal composition according to claim 3, which, in addition to comprising the compound of formula I, comprises at least one additional fungicide, herbicide or plant growth regulator.

7. A method of combating and controlling pests which comprises applying to a pest, to a locus of a pest, or to a plant susceptible to attack by a pest a pesticidally effective amount of a compound of formula I.

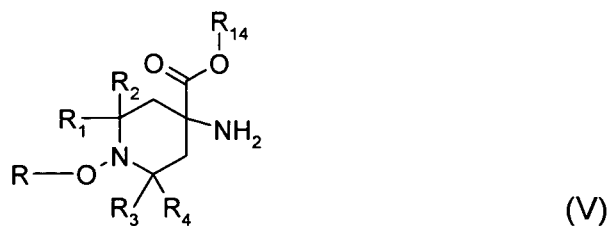
8. A method of combating and controlling pests which comprises applying to a pest, to a locus of a pest, or to a plant susceptible to attack by a pest a pesticidal composition according to claim 3.

9. Compounds of the formula IV



wherein X, Y, Z, m, n, R, R₁, R₂, R₃ and R₄ have the meanings assigned to them in claim 1, and R₁₄ is C₁₋₄alkyl.

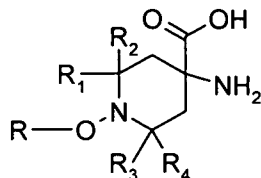
10. Compounds of the formula V



or salts thereof,

wherein R, R₁, R₂, R₃ and R₄ have the meanings assigned to them in claim 1, and R₁₄ is C₁₋₄alkyl.

11. Compounds of the formula VII

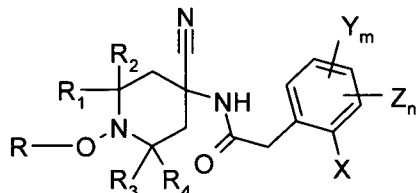


(VII)

or salts thereof,

wherein R₁, R₂, R₃ and R₄ have the meanings assigned to them in claim 1 and R is C₁-C₆alkyl, C₂-C₆alkenyl, C₃-C₆alkynyl or benzyl.

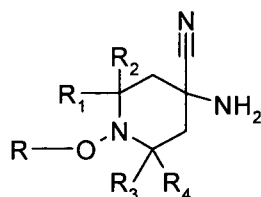
12. Compounds of the formula XI



(XI)

wherein X, Y, Z, m, n, R, R₁, R₂, R₃, and R₄ have the meanings assigned to them in claim 1.

13. Compounds of the formula VIII

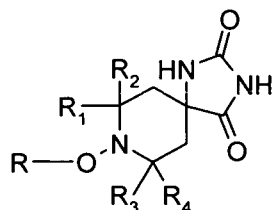


(VIII)

or salts thereof,

wherein R, R₁, R₂, R₃ and R₄ have the meanings assigned to them in claim 1.

14. Compounds of the formula IX



(IX)

wherein R₁, R₂, R₃ and R₄ have the meanings assigned to them in claim 1 and R is C₁-C₆alkyl, C₂-C₆alkenyl, C₃-C₆alkynyl or benzyl.