

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property

Organization

International Bureau

(43) International Publication Date

22 May 2020 (22.05.2020)



(10) International Publication Number

WO 2020/099404 A1

(51) International Patent Classification:

C07D 417/04 (2006.01) C07F 9/48 (2006.01)
C07F 9/38 (2006.01) A01N 43/82 (2006.01)

(21) International Application Number:

PCT/EP2019/081022

(22) International Filing Date:

12 November 2019 (12.11.2019)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

1818348.3 12 November 2018 (12.11.2018) GB

(71) Applicant: SYNGENTA CROP PROTECTION AG
[CH/CH]; Rosentalstrasse 67, 4058 Basel (CH).

(72) Inventors: SCUTT, James, Nicholas; Syngenta Limited Syngenta, Jealott's Hill International Research Centre, Bracknell Berkshire RG42 6EY (GB). WILLETTS, Nigel, James; Syngenta Limited Syngenta, Jealott's Hill International Research Centre, Bracknell Berkshire RG42 6EY (GB). NG, Sean; Syngenta Limited Syngenta, Jealott's Hill Int. Research Centre, Bracknell Berkshire RG42 6EY (GB).

(74) Agent: SYNGENTA IP; Rosentalstrasse 67, 4058 Basel (CH).

(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Declarations under Rule 4.17:

- as to applicant's entitlement to apply for and be granted a patent (Rule 4.17(ii))
- of inventorship (Rule 4.17(iv))

Published:

- with international search report (Art. 21(3))

(54) Title: HERBICIDAL COMPOUNDS

(57) Abstract: The present invention relates to herbicidally active thiadiazole derivatives, as well as to processes and intermediates used for the preparation of such derivatives. The invention further extends to herbicidal compositions comprising such derivatives, as well as to the use of such compounds and compositions in controlling undesirable plant growth: in particular the use in controlling weeds, in crops of useful plants.

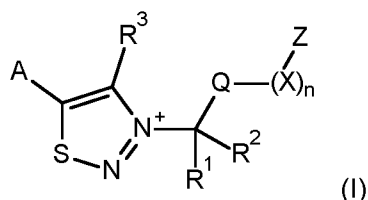


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Herbicidal Compounds

The present invention relates to herbicidally active thiadiazole derivatives, as well as to processes and intermediates used for the preparation of such derivatives. The invention further extends
 5 to herbicidal compositions comprising such derivatives, as well as to the use of such compounds and compositions in controlling undesirable plant growth: in particular the use in controlling weeds, in crops of useful plants.

The present invention is based on the finding that thiadiazole derivatives of formula (I) as defined herein, exhibit surprisingly good herbicidal activity. Thus, according to the present invention there is
 10 provided a compound of formula (I) or an agronomically acceptable salt or zwitterionic species thereof:



wherein: R^1 is selected from the group consisting of hydrogen, halogen, C_1 - C_6 alkyl, C_2 - C_6 alkenyl, C_2 - C_6 alkynyl, C_3 - C_6 cycloalkyl, C_1 - C_6 haloalkyl, $-OR^7$, $-OR^{15a}$, $-N(R^6)S(O)_2R^{15}$, $-N(R^6)C(O)R^{15}$, $-N(R^6)C(O)OR^{15}$, $-N(R^6)C(O)NR^{16}R^{17}$, $-N(R^6)CHO$, $-N(R^{7a})_2$ and $-S(O)_rR^{15}$;

15 R^2 is selected from the group consisting of hydrogen, halogen, C_1 - C_6 alkyl and C_1 - C_6 haloalkyl; or R^1 and R^2 together with the carbon atom to which they are attached form a C_3 - C_6 cycloalkyl ring or a 3- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O;

and wherein when R^1 is selected from the group consisting of $-OR^7$, $-OR^{15a}$, $-N(R^6)S(O)_2R^{15}$, $-N(R^6)C(O)R^{15}$, $-N(R^6)C(O)OR^{15}$, $-N(R^6)C(O)NR^{16}R^{17}$, $-N(R^6)CHO$, $-N(R^{7a})_2$ and $-S(O)_rR^{15}$, then R^2 is selected from the group consisting of hydrogen and C_1 - C_6 alkyl;

R^3 is selected from the group consisting of hydrogen, halogen, cyano, nitro, $-S(O)_rR^{15}$, C_1 - C_6 alkyl, C_1 - C_6 fluoroalkyl, C_1 - C_6 fluoroalkoxy, C_1 - C_6 alkoxy, C_3 - C_6 cycloalkyl and $-N(R^6)_2$;

Q is $(CR^{1a}R^{2b})_m$;

25 m is 0, 1, 2 or 3;

each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, halogen, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, $-OH$, $-OR^7$, $-OR^{15a}$, $-NH_2$, $-NHR^7$, $-NHR^{15a}$, $-N(R^6)CHO$, $-NR^{7b}R^{7c}$ and $-S(O)_rR^{15}$; or

each R^{1a} and R^{2b} together with the carbon atom to which they are attached form a C_3 - C_6 cycloalkyl ring or a 3- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O;

each R^6 is independently selected from hydrogen and C_1 - C_6 alkyl;

each R^7 is independently selected from the group consisting of C_1 - C_6 alkyl, $-S(O)_2R^{15}$, $-C(O)R^{15}$, $-C(O)OR^{15}$ and $-C(O)NR^{16}R^{17}$;

35 each R^{7a} is independently selected from the group consisting of $-S(O)_2R^{15}$, $-C(O)R^{15}$, $-C(O)OR^{15}$, $-C(O)NR^{16}R^{17}$ and $-C(O)NR^{6R^{15a}}$;

R^{7b} and R^{7c} are independently selected from the group consisting of C_1 - C_6 alkyl, $-S(O)_2R^{15}$, $-C(O)R^{15}$, $-C(O)OR^{15}$, $-C(O)NR^{16}R^{17}$ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different; or R^{7b} and R^{7c} together with the nitrogen atom to which they are attached form a 4- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N, O and S;

A is a 5-membered heteroaryl ring comprising 1, 2, 3 or 4 heteroatoms each independently selected from the group consisting of N, O and S, wherein said 5-membered heteroaryl ring is optionally substituted by 1, 2, or 3 R^8 substituents; and when A is a 5-membered heteroaryl ring substituted by R^8 on one or more ring carbon atoms, each of said R^8 substituents is independently selected from the group consisting of halogen, nitro, cyano, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OH$, $-OR^7$, $-S(O)_2R^{15}$, $-NR^6S(O)_2R^{15}$, $-C(O)OR^{10}$, $-C(O)R^{15}$, $-C(O)NR^{16}R^{17}$, $-S(O)_2NR^{16}R^{17}$, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 halocycloalkyl, C_3 - C_6 cycloalkoxy, C_2 - C_6 alkenyl, C_2 - C_6 haloalkenyl, C_2 - C_6 alkynyl, C_1 - C_3 alkoxy C_1 - C_3 alkyl-, hydroxy C_1 - C_6 alkyl-, C_1 - C_3 alkoxy C_1 - C_3 alkoxy-, C_1 - C_6 haloalkoxy, C_1 - C_3 haloalkoxy C_1 - C_3 alkyl-, C_3 - C_6 alkenyloxy, C_3 - C_6 alkynyloxy, $-C(R^6)=NOR^6$, phenyl and a 5- or 6-membered heteroaryl, which comprises 1, 2, 3 or 4 heteroatoms independently selected from N, O and S, and wherein said phenyl or heteroaryl are optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different;

and/or when A is a 5-membered heteroaryl ring substituted by an R^8 on a ring nitrogen atom, said R^8 is selected from the group consisting of $-OR^7$, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 halocycloalkyl, C_3 - C_6 cycloalkoxy, C_2 - C_6 alkenyl, C_2 - C_6 haloalkenyl, C_2 - C_6 alkynyl, C_1 - C_3 alkoxy C_1 - C_3 alkyl-, C_1 - C_3 alkoxy C_1 - C_3 alkoxy-, C_1 - C_6 haloalkoxy, C_1 - C_3 haloalkoxy C_1 - C_3 alkyl-, C_3 - C_6 alkenyloxy and C_3 - C_6 alkynyloxy;

each R^9 is independently selected from the group consisting of $-OH$, halogen, cyano, $-N(R^6)_2$, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkyl and C_1 - C_4 haloalkoxy;

X is independently selected from the group consisting of C_3 - C_6 cycloalkyl, phenyl, a 5- or 6-membered heteroaryl, which comprises 1, 2, 3 or 4 heteroatoms independently selected from N, O and S, and a 4- to 6-membered heterocyclyl, which comprises 1, 2 or 3 heteroatoms independently selected from N, O and S, and wherein said cycloalkyl, phenyl, heteroaryl or heterocyclyl moieties are optionally substituted by 1 or 2 R^9 substituents, which may be the same or different, and wherein the aforementioned CR^1R^2 Q and Z moieties may be attached at any position of said cycloalkyl, phenyl, heteroaryl or heterocyclyl moieties;

n is 0 or 1;

Z is selected from the group consisting of $-C(O)OR^{10}$, $-CH_2OH$, $-CHO$, $-C(O)NHOR^{11}$, $-C(O)NHCN$, $-OC(O)NHOR^{11}$, $-OC(O)NHCN$, $-NR^6C(O)NHOR^{11}$, $-NR^6C(O)NHCN$, $-C(O)NHS(O)_2R^{12}$, $-OC(O)NHS(O)_2R^{12}$, $-NR^6C(O)NHS(O)_2R^{12}$, $-S(O)_2OR^{10}$, $-OS(O)_2OR^{10}$, $-NR^6S(O)_2OR^{10}$, $-NR^6S(O)OR^{10}$, $-NHS(O)_2R^{14}$, $-S(O)OR^{10}$, $-OS(O)OR^{10}$, $-S(O)_2NHCN$, $-S(O)_2NHC(O)R^{18}$, $-S(O)_2NHS(O)_2R^{12}$, $-OS(O)_2NHCN$, $-OS(O)_2NHS(O)_2R^{12}$, $-OS(O)_2NHC(O)R^{18}$, $-NR^6S(O)_2NHCN$, $-NR^6S(O)_2NHC(O)R^{18}$, $-N(OH)C(O)R^{15}$, $-ONHC(O)R^{15}$, $-NR^6S(O)_2NHS(O)_2R^{12}$, $-P(O)(R^{13})(OR^{10})$, $-P(O)H(OR^{10})$, $-OP(O)(R^{13})(OR^{10})$, $-NR^6P(O)(R^{13})(OR^{10})$ and tetrazole;

R¹⁰ is selected from the group consisting of hydrogen, C₁-C₆alkyl, phenyl and benzyl, and wherein said phenyl or benzyl are optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

R¹¹ is selected from the group consisting of hydrogen, C₁-C₆alkyl and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

R¹² is selected from the group consisting of C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, -OH, -N(R⁶)₂ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

R¹³ is selected from the group consisting of -OH, C₁-C₆alkyl, C₁-C₆alkoxy and phenyl;

R¹⁴ is C₁-C₆haloalkyl;

R¹⁵ is selected from the group consisting of C₁-C₆alkyl and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

R^{15a} is phenyl, wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

R¹⁶ and R¹⁷ are independently selected from the group consisting of hydrogen and C₁-C₆alkyl; or R¹⁶ and R¹⁷ together with the nitrogen atom to which they are attached form a 4- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N, O and S;

R¹⁸ is selected from the group consisting of hydrogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, -N(R⁶)₂ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

and r is 0, 1 or 2.

According to a second aspect of the invention, there is provided an agrochemical composition comprising a herbicidally effective amount of a compound of formula (I). Such an agricultural composition may further comprise at least one additional active ingredient and/or an agrochemically acceptable diluent or carrier.

According to a third aspect of the invention, there is provided a method of controlling or preventing undesirable plant growth, wherein a herbicidally effective amount of a compound of formula (I), or a composition comprising this compound as active ingredient, is applied to the plants, to parts thereof or the locus thereof.

According to a fourth aspect of the invention, there is provided the use of a compound of formula (I) as a herbicide.

As used herein, the term "halogen" or "halo" refers to fluorine (fluoro), chlorine (chloro), bromine (bromo) or iodine (iodo), preferably fluorine, chlorine or bromine.

As used herein, cyano means a -CN group.

As used herein, hydroxy means an -OH group.

As used herein, nitro means an -NO₂ group.

As used herein, the term "C₁-C₆alkyl" refers to a straight or branched hydrocarbon chain radical consisting solely of carbon and hydrogen atoms, containing no unsaturation, having from one to six carbon atoms, and which is attached to the rest of the molecule by a single bond. C₁-C₄alkyl and C₁-

C₂alkyl are to be construed accordingly. Examples of C₁-C₆alkyl include, but are not limited to, methyl (Me), ethyl (Et), *n*-propyl, 1-methylethyl (iso-propyl), *n*-butyl, and 1-dimethylethyl (*t*-butyl).

As used herein, the term "C₁-C₆alkoxy" refers to a radical of the formula -OR_a where R_a is a C₁-C₆alkyl radical as generally defined above. C₁-C₄alkoxy is to be construed accordingly. Examples of C₁-C₄alkoxy include, but are not limited to, methoxy, ethoxy, propoxy, iso-propoxy and *t*-butoxy.

As used herein, the term "C₁-C₆haloalkyl" refers to a C₁-C₆alkyl radical as generally defined above substituted by one or more of the same or different halogen atoms. C₁-C₄haloalkyl is to be construed accordingly. Examples of C₁-C₆haloalkyl include, but are not limited to chloromethyl, fluoromethyl, fluoroethyl, difluoromethyl, trifluoromethyl and 2,2,2-trifluoroethyl.

As used herein, the term "C₂-C₆alkenyl" refers to a straight or branched hydrocarbon chain radical group consisting solely of carbon and hydrogen atoms, containing at least one double bond that can be of either the (*E*)- or (*Z*)-configuration, having from two to six carbon atoms, which is attached to the rest of the molecule by a single bond. C₂-C₄alkenyl is to be construed accordingly. Examples of C₂-C₆alkenyl include, but are not limited to, prop-1-enyl, allyl (prop-2-enyl) and but-1-enyl.

As used herein, the term "C₂-C₆haloalkenyl" refers to a C₂-C₆alkenyl radical as generally defined above substituted by one or more of the same or different halogen atoms. Examples of C₂-C₆haloalkenyl include, but are not limited to chloroethylene, fluoroethylene, 1,1-difluoroethylene, 1,1-dichloroethylene and 1,1,2-trichloroethylene.

As used herein, the term "C₂-C₆alkynyl" refers to a straight or branched hydrocarbon chain radical group consisting solely of carbon and hydrogen atoms, containing at least one triple bond, having from two to six carbon atoms, and which is attached to the rest of the molecule by a single bond. C₂-C₄alkynyl is to be construed accordingly. Examples of C₂-C₆alkynyl include, but are not limited to, prop-1-ynyl, propargyl (prop-2-ynyl) and but-1-ynyl.

As used herein, the term "C₁-C₆haloalkoxy" refers to a C₁-C₆alkoxy group as defined above substituted by one or more of the same or different halogen atoms. C₁-C₄haloalkoxy is to be construed accordingly. Examples of C₁-C₆haloalkoxy include, but are not limited to, fluoromethoxy, difluoromethoxy, fluoroethoxy, trifluoromethoxy and trifluoroethoxy.

As used herein, the term "C₁-C₃haloalkoxyC₁-C₃alkyl" refers to a radical of the formula R_b-O-R_a where R_b is a C₁-C₃haloalkyl radical as generally defined above, and R_a is a C₁-C₃alkylene radical as generally defined above.

As used herein, the term "C₁-C₃alkoxyC₁-C₃alkyl" refers to a radical of the formula R_b-O-R_a where R_b is a C₁-C₃alkyl radical as generally defined above, and R_a is a C₁-C₃alkylene radical as generally defined above.

As used herein, the term "C₁-C₃alkoxyC₁-C₃alkoxy-" refers to a radical of the formula R_b-O-R_a-O- where R_b is a C₁-C₃alkyl radical as generally defined above, and R_a is a C₁-C₃alkylene radical as generally defined above.

As used herein, the term "C₃-C₆alkenyloxy" refers to a radical of the formula -OR_a where R_a is a C₃-C₆alkenyl radical as generally defined above.

As used herein, the term "C₃-C₆alkynyloxy" refers to a radical of the formula -OR_a where R_a is a C₃-C₆alkynyl radical as generally defined above.

As used herein, the term "hydroxyC₁-C₆alkyl" refers to a C₁-C₆alkyl radical as generally defined above substituted by one or more hydroxy groups.

As used herein, the term "C₁-C₆alkylcarbonyl" refers to a radical of the formula -C(O)R_a where R_a is a C₁-C₆alkyl radical as generally defined above.

5 As used herein, the term "C₁-C₆alkoxycarbonyl" refers to a radical of the formula -C(O)OR_a where R_a is a C₁-C₆alkyl radical as generally defined above.

As used herein, the term "aminocarbonyl" refers to a radical of the formula -C(O)NH₂.

As used herein, the term "C₃-C₆cycloalkyl" refers to a stable, monocyclic ring radical which is saturated or partially unsaturated and contains 3 to 6 carbon atoms. C₃-C₄cycloalkyl is to be construed
10 accordingly. Examples of C₃-C₆cycloalkyl include, but are not limited to, cyclopropyl, cyclobutyl, cyclopentyl and cyclohexyl.

As used herein, the term "C₃-C₆halocycloalkyl" refers to a C₃-C₆cycloalkyl radical as generally defined above substituted by one or more of the same or different halogen atoms. C₃-C₄halocycloalkyl is to be construed accordingly.

15 As used herein, the term "C₃-C₆cycloalkoxy" refers to a radical of the formula -OR_a where R_a is a C₃-C₆cycloalkyl radical as generally defined above.

As used herein, the term "N-C₃-C₆cycloalkylamino" refers to a radical of the formula -NHR_a where R_a is a C₃-C₆cycloalkyl radical as generally defined above.

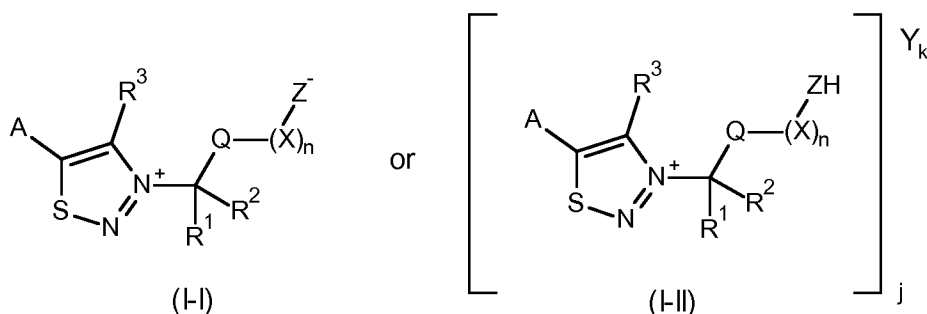
As used herein, except where explicitly stated otherwise, the term "heteroaryl" refers to a 5- or 6-
20 membered monocyclic aromatic ring which comprises 1, 2, 3 or 4 heteroatoms individually selected from nitrogen, oxygen and sulfur. The heteroaryl radical may be bonded to the rest of the molecule via a carbon atom or heteroatom. Examples of heteroaryl include, furyl, pyrrolyl, imidazolyl, thienyl, pyrazolyl, thiazolyl, isothiazolyl, oxazolyl, isoxazolyl, triazolyl, tetrazolyl, pyrazinyl, pyridazinyl, pyrimidyl or pyridyl.

As used herein, except where explicitly stated otherwise, the term "heterocyclyl" or "heterocyclic"
25 refers to a stable 4- to 6-membered non-aromatic monocyclic ring radical which comprises 1, 2, or 3 heteroatoms individually selected from nitrogen, oxygen and sulfur. The heterocyclyl radical may be bonded to the rest of the molecule via a carbon atom or heteroatom. Examples of heterocyclyl include, but are not limited to, pyrrolinyl, pyrrolidyl, tetrahydrofuryl, tetrahydrothienyl, tetrahydrothiopyranyl, piperidyl, piperazinyl, tetrahydropyranyl, dihydroisoxazolyl, dioxolanyl, morpholinyl or δ-lactamyl.

30 The presence of one or more possible asymmetric carbon atoms in a compound of formula (I) means that the compounds may occur in chiral isomeric forms, i.e., enantiomeric or diastereomeric forms. Also atropisomers may occur as a result of restricted rotation about a single bond. Formula (I) is intended to include all those possible isomeric forms and mixtures thereof. The present invention includes all those possible isomeric forms and mixtures thereof for a compound of formula (I). Likewise,
35 formula (I) is intended to include all possible tautomers (including lactam-lactim tautomerism and keto-enol tautomerism) where present. The present invention includes all possible tautomeric forms for a compound of formula (I). Similarly, where there are di-substituted alkenes, these may be present in *E* or *Z* form or as mixtures of both in any proportion. The present invention includes all these possible isomeric forms and mixtures thereof for a compound of formula (I).

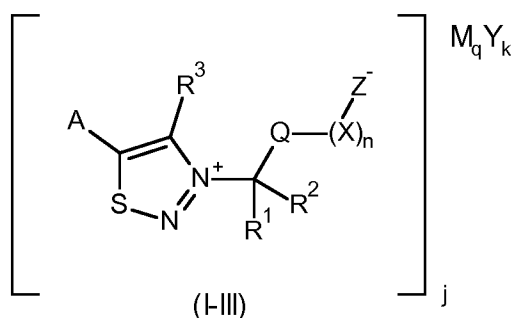
The compounds of formula (I) will typically be provided in the form of an agronomically acceptable salt, a zwitterion or an agronomically acceptable salt of a zwitterion. This invention covers all such agronomically acceptable salts, zwitterions and mixtures thereof in all proportions.

For example a compound of formula (I) wherein Z comprises an acidic proton, may exist as a zwitterion, a compound of formula (I-I), or as an agronomically acceptable salt, a compound of formula (I-II) as shown below:



wherein, Y represents an agronomically acceptable anion and j and k represent integers that may be selected from 1, 2 or 3, dependant upon the charge of the respective anion Y.

A compound of formula (I) may also exist as an agronomically acceptable salt of a zwitterion, a compound of formula (I-III) as shown below:



wherein, Y represents an agronomically acceptable anion, M represents an agronomically acceptable cation (in addition to the thiadiazolium cation) and the integers j, k and q may be selected from 1, 2 or 3, dependent upon the charge of the respective anion Y and respective cation M.

Thus where a compound of formula (I) is drawn in protonated form herein, the skilled person would appreciate that it could equally be represented in unprotonated or salt form with one or more relevant counter ions.

In one embodiment of the invention there is provided a compound of formula (I-II) wherein k is 2, j is 1 and Y is selected from the group consisting of halogen, trifluoroacetate and pentafluoropropionate. In this embodiment a nitrogen atom in ring A may be protonated or a nitrogen atom comprised in R¹, R², Q or X may be protonated. Preferably, in a compound of formula (I-II), k is 2, j is 1 and Y is chloride, wherein a nitrogen atom in ring A is protonated.

Suitable agronomically acceptable salts of the present invention, represented by an anion Y, include but are not limited chloride, bromide, iodide, fluoride, 2-naphthalenesulfonate, acetate, adipate, methoxide, ethoxide, propoxide, butoxide, aspartate, benzenesulfonate, benzoate, bicarbonate, bisulfate, bitartrate, butylsulfate, butylsulfonate, butyrate, camphorate, camsylate, caprate, caproate, caprylate, carbonate, citrate, diphosphate, edetate, edisylate, enanthate, ethanedisulfonate,

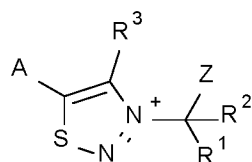
ethanesulfonate, ethylsulfate, formate, fumarate, gluceptate, gluconate, glucuronate, glutamate, glycerophosphate, heptadecanoate, hexadecanoate, hydrogen sulfate, hydroxide, hydroxynaphthoate, isethionate, lactate, lactobionate, laurate, malate, maleate, mandelate, mesylate, methanedisulfonate, methylsulfate, mucate, myristate, napsylate, nitrate, nonadecanoate, octadecanoate, oxalate, 5 pelargonate, pentadecanoate, pentafluoropropionate, perchlorate, phosphate, propionate, propylsulfate, propylsulfonate, succinate, sulfate, tartrate, tosylate, tridecylate, triflate, trifluoroacetate, undecylate and valerate.

Suitable cations represented by M include, but are not limited to, metals, conjugate acids of amines and organic cations. Examples of suitable metals include aluminium, calcium, cesium, copper, 10 lithium, magnesium, manganese, potassium, sodium, iron and zinc. Examples of suitable amines include allylamine, ammonia, amylamine, arginine, benethamine, benzathine, butenyl-2-amine, butylamine, butylethanolamine, cyclohexylamine, decylamine, diamylamine, dibutylamine, diethanolamine, diethylamine, diethylenetriamine, diheptylamine, dihexylamine, diisoamylamine, diisopropylamine, dimethylamine, dioctylamine, dipropanolamine, dipropargylamine, dipropylamine, 15 dodecylamine, ethanolamine, ethylamine, ethylbutylamine, ethylenediamine, ethylheptylamine, ethyloctylamine, ethylpropanolamine, heptadecylamine, heptylamine, hexadecylamine, hexenyl-2-amine, hexylamine, hexylheptylamine, hexyloctylamine, histidine, indoline, isoamylamine, isobutanolamine, isobutylamine, isopropanolamine, isopropylamine, lysine, meglumine, methoxyethylamine, methylamine, methylbutylamine, methylethylamine, methylhexylamine, 20 methylisopropylamine, methylnonylamine, methyloctadecylamine, methylpentadecylamine, morpholine, N,N-diethylethanolamine, N-methylpiperazine, nonylamine, octadecylamine, octylamine, oleylamine, pentadecylamine, pentenyl-2-amine, phenoxyethylamine, picoline, piperazine, piperidine, propanolamine, propylamine, propylenediamine, pyridine, pyrrolidine, sec-butylamine, stearylamine, tallowamine, tetradecylamine, tributylamine, tridecylamine, trimethylamine, triheptylamine, 25 trihexylamine, triisobutylamine, triisodecylamine, triisopropylamine, trimethylamine, tripentylamine, tripropylamine, tris(hydroxymethyl)aminomethane, and undecylamine. Examples of suitable organic cations include benzyltributylammonium, benzyltrimethylammonium, benzyltriphenylphosphonium, choline, tetrabutylammonium, tetrabutylphosphonium, tetraethylammonium, tetraethylphosphonium, tetramethylammonium, tetramethylphosphonium, tetrapropylammonium, tetrapropylphosphonium, 30 tributylsulfonium, tributylsulfoxonium, triethylsulfonium, triethylsulfoxonium, trimethylsulfonium, trimethylsulfoxonium, tripropylsulfonium and tripropylsulfoxonium.

Preferred compounds of formula (I), wherein Z comprises an acidic proton, can be represented as either (I-I) or (I-II). For compounds of formula (I-II) emphasis is given to salts when Y is chloride, bromide, iodide, hydroxide, bicarbonate, acetate, pentafluoropropionate, triflate, trifluoroacetate, 35 methylsulfate, tosylate and nitrate, wherein j and k are 1. Preferably, Y is chloride, bromide, iodide, hydroxide, bicarbonate, acetate, trifluoroacetate, methylsulfate, tosylate and nitrate, wherein j and k are 1. For compounds of formula (I-II) emphasis is also given to salts when Y is carbonate and sulfate, wherein j is 2 and k is 1, and when Y is phosphate, wherein j is 3 and k is 1.

Where appropriate compounds of formula (I) may also be in the form of (and/or be used as) an 40 N-oxide.

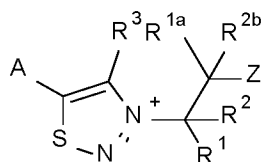
Compounds of formula (I) wherein m is 0 and n is 0 may be represented by a compound of formula (I-la) as shown below:



(I-la)

wherein R¹, R², R³, A and Z are as defined for compounds of formula (I).

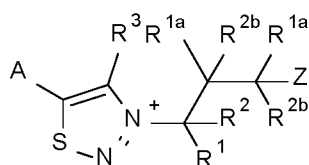
5 Compounds of formula (I) wherein m is 1 and n is 0 may be represented by a compound of formula (I-lb) as shown below:



(I-lb)

wherein R¹, R², R¹ᵃ, R²ᵇ, R³, A and Z are as defined for compounds of formula (I).

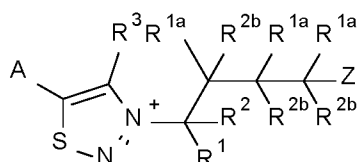
Compounds of formula (I) wherein m is 2 and n is 0 may be represented by a compound of formula (I-lc) as shown below:



(I-lc)

wherein R¹, R², R¹ᵃ, R²ᵇ, R³, A and Z are as defined for compounds of formula (I).

Compounds of formula (I) wherein m is 3 and n is 0 may be represented by a compound of formula (I-l) as shown below:



(I-l)

15 wherein R¹, R², R¹ᵃ, R²ᵇ, R³, A and Z are as defined for compounds of formula (I).

The following list provides definitions, including preferred definitions, for substituents n, m, r, A, Q, X, Z, R¹, R², R¹ᵃ, R²ᵇ, R³, R⁶, R⁷, R⁷ᵃ, R⁷ᵇ, R⁷ᶜ, R⁸, R⁹, R¹⁰, R¹¹, R¹², R¹³, R¹⁴, R¹⁵, R¹⁵ᵃ, R¹⁶, R¹⁷ and R¹⁸ with reference to the compounds of formula (I) according to the invention. For any one of these substituents, any of the definitions given below may be combined with any definition of any other substituent given below or elsewhere in this document.

R¹ is selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl, C₂-C₆alkenyl, C₂-C₆alkynyl, C₃-C₆cycloalkyl, C₁-C₆haloalkyl, -OR⁷, -OR^{15a}, -N(R⁶)S(O)₂R¹⁵, -N(R⁶)C(O)R¹⁵, -N(R⁶)C(O)OR¹⁵, -N(R⁶)C(O)NR¹⁶R¹⁷, -N(R⁶)CHO, -N(R^{7a})₂ and -S(O)_rR¹⁵. Preferably, R¹ is selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl, C₁-C₆fluoroalkyl, -OR⁷, -NHS(O)₂R¹⁵, -NHC(O)R¹⁵, -NHC(O)OR¹⁵, -NHC(O)NR¹⁶R¹⁷, -N(R^{7a})₂ and -S(O)_rR¹⁵. More preferably, R¹ is selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl, C₁-C₆fluoroalkyl, -OR⁷ and -N(R^{7a})₂. Even more preferably, R¹ is selected from the group consisting of hydrogen, C₁-C₆alkyl, -OR⁷ and -N(R^{7a})₂. Even more preferably still, R¹ is hydrogen or C₁-C₆alkyl. Yet even more preferably still, R¹ is hydrogen or methyl. Most preferably R¹ is hydrogen.

10 R² is selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl and C₁-C₆haloalkyl. Preferably, R² is selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl and C₁-C₆fluoroalkyl. More preferably, R² is hydrogen or C₁-C₆alkyl. Even more preferably, R² is hydrogen or methyl. Most preferably R² is hydrogen.

Wherein when R¹ is selected from the group consisting of -OR⁷, -OR^{15a}, -N(R⁶)S(O)₂R¹⁵, -N(R⁶)C(O)R¹⁵, -N(R⁶)C(O)OR¹⁵, -N(R⁶)C(O)NR¹⁶R¹⁷, -N(R⁶)CHO, -N(R^{7a})₂ and -S(O)_rR¹⁵, R² is selected from the group consisting of hydrogen and C₁-C₆alkyl. Preferably, when R¹ is selected from the group consisting of -OR⁷, -NHS(O)₂R¹⁵, -NHC(O)R¹⁵, -NHC(O)OR¹⁵, -NHC(O)NR¹⁶R¹⁷, -N(R^{7a})₂ and -S(O)_rR¹⁵, R² is selected from the group consisting of hydrogen and methyl.

Alternatively, R¹ and R² together with the carbon atom to which they are attached form a C₃-C₆cycloalkyl ring or a 3- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O. Preferably, R¹ and R² together with the carbon atom to which they are attached form a C₃-C₆cycloalkyl ring. More preferably, R¹ and R² together with the carbon atom to which they are attached form a cyclopropyl ring.

In one embodiment R¹ and R² are hydrogen.

25 In another embodiment R¹ is methyl and R² is hydrogen.

In another embodiment R¹ is methyl and R² is methyl.

Q is (CR^{1a}R^{2b})_m.

The value of m is 0, 1, 2 or 3. Preferably, m is 0, 1, or 2. More preferably, m is 1 or 2. Most preferably, m is 1.

30 Each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl, C₁-C₆haloalkyl, -OH, -OR⁷, -OR^{15a}, -NH₂, -NHR⁷, -NHR^{15a}, -N(R⁶)CHO, -NR^{7b}R^{7c} and -S(O)_rR¹⁵. Preferably, each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, halogen, C₁-C₆alkyl, C₁-C₆fluoroalkyl, -OH, -NH₂ and -NHR⁷. More preferably, each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, C₁-C₆alkyl, -OH and -NH₂. Even more preferably, each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, methyl, -OH and -NH₂. Even more preferably still, each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen and methyl. Most preferably R^{1a} and R^{2b} are hydrogen.

In another embodiment each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen and C₁-C₆alkyl.

40 Alternatively, each R^{1a} and R^{2b} together with the carbon atom to which they are attached form a C₃-C₆cycloalkyl ring or a 3- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms

individually selected from N and O. Preferably, each R^{1a} and R^{2b} together with the carbon atom to which they are attached form a C₃-C₆cycloalkyl ring. More preferably, each R^{1a} and R^{2b} together with the carbon atom to which they are attached form a cyclopropyl ring.

Preferably R³ is hydrogen, C₁-C₆alkyl, -C(O)NH₂, and -C(O)C₁-C₆alkyl; more preferably
5 hydrogen, C₁-C₆alkyl, or C₁-C₆alkoxy, and more preferably still, hydrogen or methyl. Most preferably R³ is hydrogen.

Each R⁶ is independently selected from hydrogen and C₁-C₆alkyl. Preferably, each R⁶ is independently selected from hydrogen and methyl.

Each R⁷ is independently selected from the group consisting of C₁-C₆alkyl, -S(O)₂R¹⁵, -C(O)R¹⁵,
10 -C(O)OR¹⁵ and -C(O)NR¹⁶R¹⁷. Preferably, each R⁷ is independently selected from the group consisting of C₁-C₆alkyl, -C(O)R¹⁵ and -C(O)NR¹⁶R¹⁷. More preferably, each R⁷ is C₁-C₆alkyl. Most preferably, each R⁷ is methyl.

Each R^{7a} is independently selected from the group consisting of -S(O)₂R¹⁵, -C(O)R¹⁵, -C(O)OR¹⁵,
-C(O)NR¹⁶R¹⁷ and -C(O)NR⁶R^{15a}. Preferably, each R^{7a} is independently -C(O)R¹⁵ or -C(O)NR¹⁶R¹⁷.

15 R^{7b} and R^{7c} are independently selected from the group consisting of C₁-C₆alkyl, -S(O)₂R¹⁵, -C(O)R¹⁵, -C(O)OR¹⁵, -C(O)NR¹⁶R¹⁷ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different. Preferably, R^{7b} and R^{7c} are independently selected from the group consisting of C₁-C₆alkyl, -C(O)R¹⁵ and -C(O)NR¹⁶R¹⁷. More preferably, R^{7b} and R^{7c} are C₁-C₆alkyl. Most preferably, R^{7b} and R^{7c} are methyl.

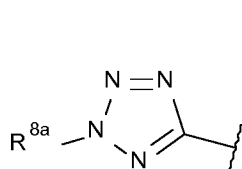
20 Alternatively, R^{7b} and R^{7c} together with the nitrogen atom to which they are attached form a 4- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N, O and S. Preferably, R^{7b} and R^{7c} together with the nitrogen atom to which they are attached form a 5- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N and O. More preferably, R^{7b} and R^{7c} together with the nitrogen
25 atom to which they are attached form an pyrrolidyl, oxazolidinyl, imidazolidinyl, piperidyl, piperazinyl or morpholinyl group.

A is a 5-membered heteroaryl attached to the rest of the molecule via a ring carbon atom, which comprises 1, 2, or 3 heteroatoms independently selected from the group consisting of N, O and S, and wherein the heteroaryl is optionally substituted (where feasible) by 1, 2 or 3 R⁸ substituents,
30 which may be the same or different.

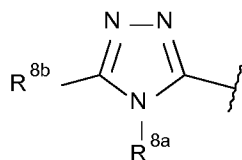
Preferably, A is a heteroaryl selected from the group consisting of 1,2,3,5-oxatriazol-4-yl, 1,2,3,5-thiatriazol-4-yl, 1,2,4-oxadiazol-3-yl, 1,2,4-oxadiazol-5-yl, 1,2,4-thiadiazol-3-yl, 1,2,4-thiadiazol-5-yl, 1,2,4-triazol-3-yl, 1,2,4-triazol-5-yl, 1,2,5-oxadiazol-3-yl, 1,2,5-thiadiazol-3-yl, 1,3,4-oxadiazol-2-yl, 1,3,4-thiadiazol-2-yl, 2-furyl, 2-thienyl, 3-furyl, 3-thienyl, imidazol-2-yl, imidazol-4-yl, imidazol-5-yl,
35 isothiazol-3-yl, isothiazol-4-yl, isothiazol-5-yl, isoxazol-3-yl, isoxazol-4-yl, isoxazol-5-yl, 1,2,3-oxadiazol-4-yl, 1,2,3-oxadiazol-5-yl, 1,2,3,4-oxatriazol-5-yl, oxazol-2-yl, oxazol-4-yl, oxazol-5-yl, pyrazol-3-yl, pyrazol-4-yl, pyrazol-5-yl, pyrrol-2-yl, pyrrol-3-yl, tetrazol-5-yl, 1,2,3-thiadiazol-4-yl, 1,2,3-thiadiazol-5-yl, 1,2,3,4-thiatriazol-5-yl, thiazol-2-yl, thiazol-4-yl, thiazol-5-yl, triazol-4-yl and triazol-5-yl wherein the heteroaryl is optionally substituted (where feasible) by 1, 2 or 3 R⁸ substituents, which
40 may be the same or different.

More preferably, A is a heteroaryl selected from the group consisting of tetrazol-5-yl, 1,2,4-triazol-3-yl, 1,2,4-triazol-5-yl, isoxazol-3-yl, oxazol-2-yl, thiazol-2-yl, 1,3,4-thiadiazol-2-yl, triazol-4-yl, triazol-5-yl, pyrazol-3-yl, pyrazol-5-yl, 1,3,4-oxadiazol-2-yl, 1,2,4-thiadiazol-5-yl, oxazol-4-yl, imidazol-2-yl, isothiazol-5-yl, 2-thienyl, 3-furyl, 2-furyl, isothiazol-4-yl, thiazol-4-yl, 3-thienyl, imidazol-5-yl, 5 isoxazol-5-yl and 1,2,4-oxadiazol-5-yl wherein the heteroaryl may, where feasible, be optionally substituted by 1, 2 or 3 R⁸ substituents, which may be the same or different.

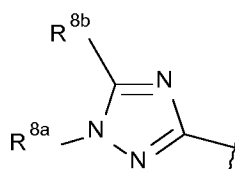
Even more preferably, A is selected from the group consisting of formula A-I to A-XXVIII below:



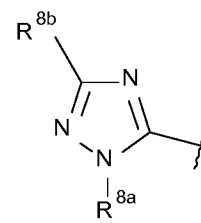
A-I



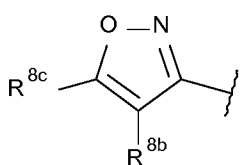
A-II



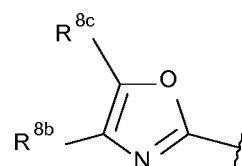
A-III



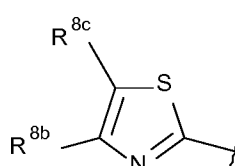
A-IV



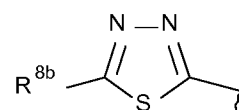
A-V



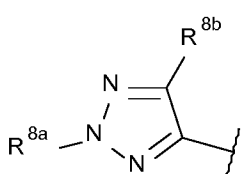
A-VI



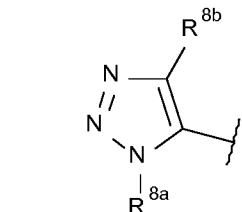
A-VII



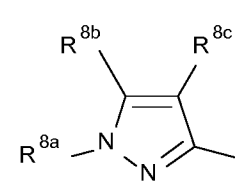
A-VIII



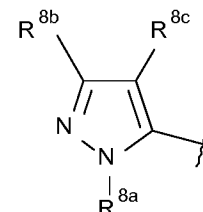
A-IX



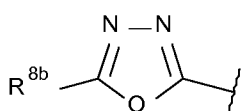
A-X



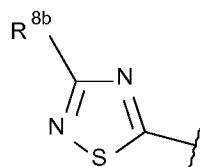
A-XI



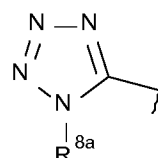
A-XII



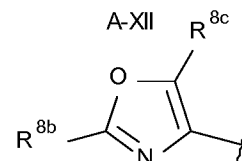
A-XIII



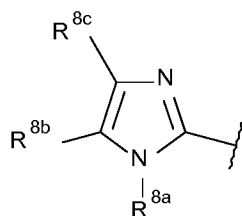
A-XIV



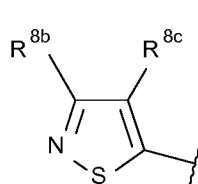
A-XV



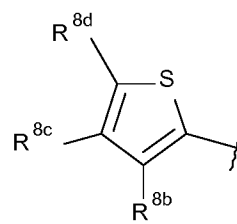
A-XVI



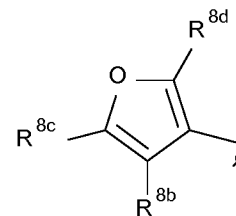
A-XVII



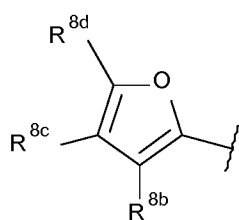
A-XVIII



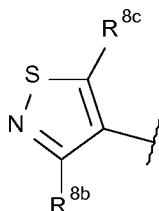
A-XIX



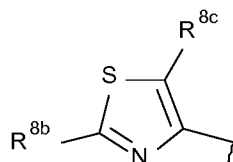
A-XX



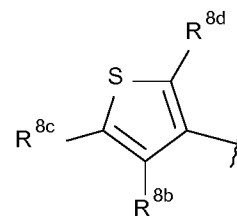
A-XXI



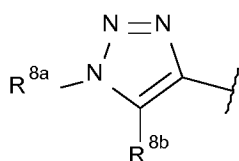
A-XXII



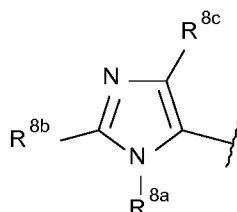
A-XXIII



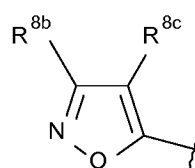
A-XXIV



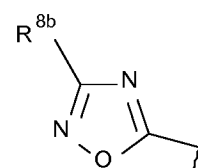
A-XXV



A-XXVI



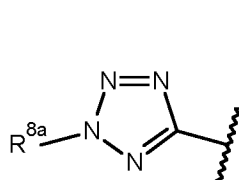
A-XXVII



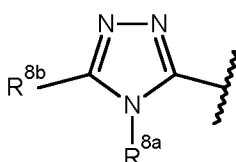
A-XXVIII

wherein the jagged line defines the point of attachment to the rest of the molecule and
 R^{8a} , R^{8b} , R^{8c} , R^{8d} , R^6 , R^7 , R^{10} , R^{15} , R^{16} and R^{17} are as defined herein. R^{8a} , R^{8b} , R^{8c} , R^{8d} are examples of
 5 R^8 wherein the subscript letter a, b, c and d are used to denote positions within individual heterocycles (A-I to A-XXVIII).

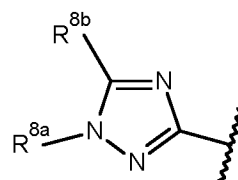
Even more preferably still, A is selected from the group consisting of formula A-I to A-VIII below



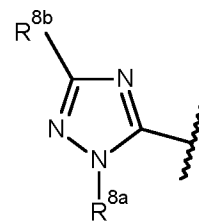
A-I



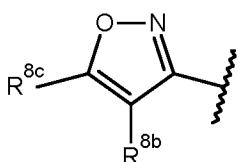
A-II



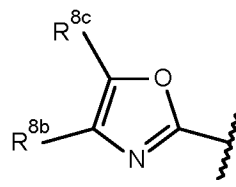
A-III



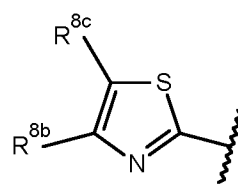
A-IV



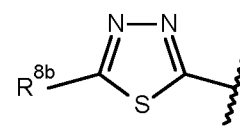
A-V



A-VI



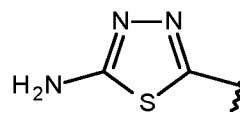
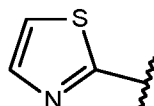
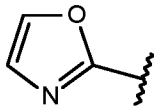
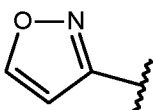
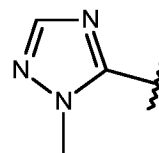
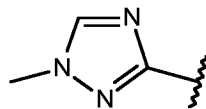
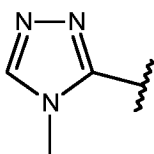
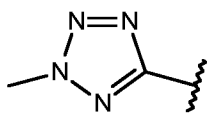
A-VII



A-VIII

10 wherein the jagged line defines the point of attachment to a compound of formula (I),
 R^{8a} , R^{8b} , R^{8c} , R^{8d} and R^7 is as defined herein.

Yet, even more preferably still, A is selected from the group consisting of formula A-Ia to A-VIIIa below



wherein the jagged line defines the point of attachment to the rest of the molecule. Yet even more
5 preferably, A is selected from A-IIa, A-VIa and A-VIIa as described above.

When A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of halogen, nitro, cyano, -NH₂, -NHR⁷, -N(R⁷)₂, -OH, -OR⁷, -S(O)_rR¹⁵, NR⁶S(O)₂R¹⁵, -C(O)OR¹⁰, -C(O)R¹⁵, -C(O)NR¹⁶R¹⁷, -S(O)₂NR¹⁶R¹⁷, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₆cycloalkyl, C₃-C₆halocycloalkyl, C₃-C₆cycloalkoxy, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₃alkoxyC₁-C₃alkyl-, hydroxyC₁-C₆alkyl-, C₁-C₃alkoxyC₁-C₃alkoxy-, C₁-C₆haloalkoxy, C₁-C₃haloalkoxyC₁-C₃alkyl-, C₃-C₆alkenyloxy, C₃-C₆alkynyloxy, -C(R⁶)=NOR⁶, phenyl and a 5- or 6-membered heteroaryl, which comprises 1, 2, 3 or 4 heteroatoms independently selected from N, O and S, and wherein said phenyl or heteroaryl are optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different.

15 Preferably, when A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of halogen, nitro, cyano, -NH₂, -NHR⁷, -N(R⁷)₂, -OH, -OR⁷, -S(O)_rR¹⁵, -NR⁶S(O)₂R¹⁵, -C(O)OR¹⁰, -C(O)R¹⁵, -C(O)NR¹⁶R¹⁷, -S(O)₂NR¹⁶R¹⁷, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₆cycloalkyl, C₁-C₃alkoxyC₁-C₃alkyl-, hydroxyC₁-C₆alkyl-, C₁-C₃alkoxyC₁-C₃alkoxy-, C₁-C₆haloalkoxy, phenyl and a 6- membered heteroaryl, which comprises 1 or 2 nitrogen atoms, and wherein said phenyl
20 or heteroaryl are optionally substituted by 1 or 2 R⁹ substituents, which may be the same or different.

More preferably, when A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of halogen, nitro, cyano, -NH₂, -NHR⁷, -N(R⁷)₂, -OH, -OR⁷, -S(O)_rR¹⁵, -NR⁶S(O)₂R¹⁵, -C(O)OR¹⁰, -C(O)R¹⁵, -C(O)NR¹⁶R¹⁷, -S(O)₂NR¹⁶R¹⁷, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₆cycloalkyl, hydroxyC₁-C₆alkyl-, C₁-C₆haloalkoxy and a 6- membered heteroaryl, which comprises 1 or 2 nitrogen atoms, and wherein said heteroaryl is optionally substituted by 1 R⁹ substituent.

Even more preferably, when A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of halogen, cyano, -NH₂, -NHR⁷, -N(R⁷)₂, -OH, -OR⁷, -S(O)₂R¹⁵, -C(O)OR¹⁰, -C(O)R¹⁵, -C(O)NR¹⁶R¹⁷, C₁-C₆alkyl and C₁-C₆haloalkyl.

30 Even more preferably still, when A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of chloro, fluoro, cyano, -NH₂, -NHMe, -NMe₂, -OH, -

OMe, $-S(O)_2Me$, $-C(O)OMe$, $-C(O)OH$, $-C(O)Me$, $-C(O)NH_2$, $-C(O)NHMe$, $-C(O)NMe_2$, methyl, *iso*-propyl and trifluoromethyl.

Most preferably, when A is substituted on one or more ring carbon atoms, each R^8 is independently selected from the group consisting of chloro, $-NH_2$, $-NHMe$, $-OMe$, methyl, *iso*-propyl and
5 trifluoromethyl.

When A is substituted on a ring nitrogen atom by R^8 , said R^8 substituent is selected from the group consisting of $-OR^7$, C_1-C_6alkyl , $C_1-C_6haloalkyl$, $C_3-C_6cycloalkyl$, $C_3-C_6halocycloalkyl$, $C_3-C_6cycloalkoxy$, $C_2-C_6alkenyl$, $C_2-C_6haloalkenyl$, $C_2-C_6alkynyl$, $C_1-C_3alkoxyC_1-C_3alkyl-$, hydroxy $C_1-C_6alkyl-$, $C_1-C_3alkoxyC_1-C_3alkoxy-$, $C_1-C_6haloalkoxy$, $C_1-C_3haloalkoxyC_1-C_3alkyl-$, $C_3-C_6alkenyloxy$ and
10 $C_3-C_6alkynyloxy$. Preferably, said R^8 is selected from the group consisting of $-OR^7$, C_1-C_6alkyl and $C_1-C_6haloalkyl$. More preferably, each of said R^8 is $-OR^7$ or C_1-C_6alkyl . Even more preferably still, each of said R^8 is C_1-C_6alkyl . Most preferably said R^8 is methyl.

When A is selected from the group consisting of formula A-I to A-XXVIII, R^{8a} (substituted on a ring nitrogen atom) is selected from the group consisting of hydrogen, C_1-C_6alkyl and $C_1-C_6haloalkyl$,
15 and each R^{8b} , R^{8c} and R^{8d} (substituted on a ring carbon atom) are independently selected from the group consisting of hydrogen, halogen, nitro, cyano, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OH$, $-OR^7$, $-S(O)_rR^{15}$, $-NR^6S(O)_2R^{15}$, $-C(O)OR^{10}$, $-C(O)R^{15}$, $-C(O)NR^{16}R^{17}$, $-S(O)_2NR^{16}R^{17}$, C_1-C_6alkyl and $C_1-C_6haloalkyl$. Preferably R^{8a} is hydrogen or C_1-C_6alkyl and each R^{8b} , R^{8c} and R^{8d} are independently selected from the group consisting of hydrogen, halogen, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OR^7$, C_1-C_6alkyl and $C_1-C_6haloalkyl$. More
20 preferably, R^{8a} is hydrogen or methyl and each R^{8b} , R^{8c} and R^{8d} are independently selected from the group consisting of hydrogen, chloro, $-NH_2$, $-NHMe$, $-OMe$, methyl, *iso*-propyl and trifluoromethyl.

When A is selected from the group consisting of formula A-I to A-VIII, R^{8a} (substituted on a ring nitrogen atom) is hydrogen or C_1-C_6alkyl , and each R^{8b} , R^{8c} and R^{8d} (substituted on a ring carbon atom) are independently selected from the group consisting of hydrogen, halogen, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OR^7$,
25 C_1-C_6alkyl and $C_1-C_6haloalkyl$. Preferably, R^{8a} is hydrogen or methyl and each R^{8b} , R^{8c} and R^{8d} are independently selected from the group consisting of hydrogen, chloro, $-NH_2$, $-NHMe$, $-OMe$, methyl, *iso*-propyl and trifluoromethyl.

Each R^9 is independently selected from the group consisting of halogen, cyano, $-N(R^6)_2$, C_1-C_4alkyl , $C_1-C_4alkoxy$, $C_1-C_4haloalkyl$ and $C_1-C_4haloalkoxy$. Preferably, each R^9 is independently
30 selected from the group consisting of halogen, C_1-C_4alkyl , $C_1-C_4alkoxy$ and $C_1-C_4haloalkyl$. More preferably, each R^9 is independently selected from the group consisting of halogen and C_1-C_4alkyl .

As defined herein, n is 0 or 1. When n is 1, X is selected from the group consisting of $C_3-C_6cycloalkyl$, phenyl, a 5- or 6- membered heteroaryl, which comprises 1, 2, 3 or 4 heteroatoms independently selected from N, O and S, and a 4- to 6- membered heterocyclyl, which comprises 1, 2
35 or 3 heteroatoms independently selected from N, O and S, and wherein said cycloalkyl, phenyl, heteroaryl or heterocyclyl moieties are optionally substituted by 1 or 2 substituents, which may be the same or different, selected from R^9 , and wherein the aforementioned CR^1R^2 and Z, or Q and Z, moieties may be attached at any position of said cycloalkyl, phenyl, heteroaryl or heterocyclyl moieties.

Preferably, X is selected from the group consisting of phenyl and a 4- to 6- membered
40 heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O, and wherein said phenyl or heterocyclyl moieties are optionally substituted by 1 or 2 substituents, which may be the same

or different, selected from R^9 , and wherein the aforementioned CR^1R^2 , Q and Z moieties may be attached at any position of said phenyl or heterocyclyl moieties.

More preferably, X is a 4- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O, and wherein said heterocyclyl moieties is optionally substituted by 1 or 2 substituents, which may be the same or different, selected from R^9 , and wherein the aforementioned CR^1R^2 , Q and Z moieties may be attached at any position of said heterocyclyl moiety.

In one embodiment, X is a 5-membered heterocyclyl, which comprises 1 heteroatom, wherein said heteroatom is N, and wherein the aforementioned CR^1R^2 , Q and Z moieties may be attached at any position of said heterocyclyl moiety. Preferably, X is a 5-membered heterocyclyl, which comprises 1 heteroatom, wherein said heteroatom is N, and wherein the aforementioned CR^1R^2 and Q moieties are attached adjacent to the N atom and the Z moiety is attached to the N atom.

In another embodiment, X is phenyl optionally substituted by 1 or 2 substituents, which may be the same or different, selected from R^9 , and wherein the aforementioned CR^1R^2 , Q and Z moieties may be attached at any position of said phenyl moiety. Preferably, X is phenyl and the aforementioned CR^1R^2 and Q moieties are attached in a position *para* to the Z moiety.

In further preferred embodiments, n is 0 and thus X is absent.

Z is selected from the group consisting of $-C(O)OR^{10}$, $-CH_2OH$, $-CHO$, $-C(O)NHOR^{11}$, $-C(O)NHCN$, $-OC(O)NHOR^{11}$, $-OC(O)NHCN$, $-NR^6C(O)NHOR^{11}$, $-NR^6C(O)NHCN$, $-C(O)NHS(O)_2R^{12}$, $-OC(O)NHS(O)_2R^{12}$, $-NR^6C(O)NHS(O)_2R^{12}$, $-S(O)_2OR^{10}$, $-OS(O)_2OR^{10}$, $-NR^6S(O)_2OR^{10}$, $-NR^6S(O)OR^{10}$, $-NHS(O)_2R^{14}$, $-S(O)OR^{10}$, $-OS(O)OR^{10}$, $-S(O)_2NHCN$, $-S(O)_2NHC(O)R^{18}$, $-S(O)_2NHS(O)_2R^{12}$, $-OS(O)_2NHCN$, $-OS(O)_2NHS(O)_2R^{12}$, $-OS(O)_2NHC(O)R^{18}$, $-NR^6S(O)_2NHCN$, $-NR^6S(O)_2NHC(O)R^{18}$, $-N(OH)C(O)R^{15}$, $-ONHC(O)R^{15}$, $-NR^6S(O)_2NHS(O)_2R^{12}$, $-P(O)(R^{13})(OR^{10})$, $-P(O)H(OR^{10})$, $-OP(O)(R^{13})(OR^{10})$, $-NR^6P(O)(R^{13})(OR^{10})$ and tetrazole.

Preferably, Z is selected from the group consisting of $-C(O)OR^{10}$, $-C(O)NHOR^{11}$, $-OC(O)NHOR^{11}$, $-NR^6C(O)NHOR^{11}$, $-C(O)NHS(O)_2R^{12}$, $-OC(O)NHS(O)_2R^{12}$, $-NR^6C(O)NHS(O)_2R^{12}$, $-S(O)_2OR^{10}$, $-OS(O)_2OR^{10}$, $-NR^6S(O)_2OR^{10}$, $-NR^6S(O)OR^{10}$, $-NHS(O)_2R^{14}$, $-S(O)OR^{10}$, $-OS(O)OR^{10}$, $-S(O)_2NHC(O)R^{18}$, $-S(O)_2NHS(O)_2R^{12}$, $-OS(O)_2NHS(O)_2R^{12}$, $-OS(O)_2NHC(O)R^{18}$, $-NR^6S(O)_2NHC(O)R^{18}$, $-N(OH)C(O)R^{15}$, $-ONHC(O)R^{15}$, $-NR^6S(O)_2NHS(O)_2R^{12}$, $-P(O)(R^{13})(OR^{10})$, $-P(O)H(OR^{10})$, $-OP(O)(R^{13})(OR^{10})$ and $-NR^6P(O)(R^{13})(OR^{10})$.

More preferably, Z is selected from the group consisting of $-C(O)OR^{10}$, $-C(O)NHOR^{11}$, $-C(O)NHS(O)_2R^{12}$, $-S(O)_2OR^{10}$, $-OS(O)_2OR^{10}$, $-NR^6S(O)_2OR^{10}$, $-NHS(O)_2R^{14}$, $-S(O)OR^{10}$ and $-P(O)(R^{13})(OR^{10})$.

Even more preferably Z is selected from the group consisting of $-C(O)OR^{10}$, $-C(O)NHS(O)_2R^{12}$, $-S(O)_2OR^{10}$, and $-P(O)(R^{13})(OR^{10})$.

Even more preferably still Z is selected from the group consisting of $-C(O)OH$, $-C(O)OCH_3$, $-C(O)OCH_2CH_3$, $-C(O)OCH(CH_3)_2$, $-C(O)OC(CH_3)_3$, $-C(O)OCH_2C_6H_5$, $-C(O)OC_6H_5$, $-C(O)NHS(O)_2CH_3$, $-S(O)_2OH$, $-P(O)(OH)(OCH_2CH_3)$ and $-P(O)(OCH_2CH_3)(OCH_2CH_3)$.

Most preferably Z is $-C(O)OH$ or $-S(O)_2OH$.

R^{10} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl, phenyl and benzyl, and wherein said phenyl or benzyl are optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different. Preferably, R^{10} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl, phenyl

and benzyl. More preferably, R^{10} is selected from the group consisting of hydrogen and C_1 - C_6 alkyl. Most preferably, R^{10} is hydrogen.

R^{11} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different.

5 Preferably, R^{11} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl and phenyl. More preferably, R^{11} is selected from the group consisting of hydrogen and C_1 - C_6 alkyl. Even more preferably, R^{11} is C_1 - C_6 alkyl. Most preferably, R^{11} is methyl.

R^{12} is selected from the group consisting of C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkoxy, -OH, - $N(R^6)_2$ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R^9 substituents, which
10 may be the same or different. Preferably, R^{12} is selected from the group consisting of C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkoxy, -OH, - $N(R^6)_2$ and phenyl. More preferably, R^{12} is selected from the group consisting of C_1 - C_6 alkyl, C_1 - C_6 haloalkyl and - $N(R^6)_2$. Even more preferably, R^{12} is selected from the group consisting of methyl, - $N(Me)_2$ and trifluoromethyl. Most preferably, R^{12} is methyl.

R^{13} is selected from the group consisting of -OH, C_1 - C_6 alkyl, C_1 - C_6 alkoxy and phenyl. Preferably
15 R^{13} is selected from the group consisting of -OH, C_1 - C_6 alkyl and C_1 - C_6 alkoxy. More preferably, R^{13} is selected from the group consisting of -OH and C_1 - C_6 alkoxy. Even more preferably, R^{13} is selected from the group consisting of -OH, methoxy and ethoxy. Most preferably, R^{13} is -OH.

R^{14} is C_1 - C_6 haloalkyl. Preferably, R^{14} is trifluoromethyl.

R^{15} is selected from the group consisting of C_1 - C_6 alkyl and phenyl, and wherein said phenyl is
20 optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different. Preferably, R^{15} is selected from the group consisting of C_1 - C_6 alkyl and phenyl. More preferably, R^{15} is C_1 - C_6 alkyl. Most preferably R^{15} is methyl.

R^{15a} is phenyl, wherein said phenyl is optionally substituted by 1, 2 or 3 R^9 substituents, which
25 may be the same or different. Preferably, R^{15a} is phenyl optionally substituted by 1 R^9 substituent. More preferably, R^{15a} is phenyl.

R^{16} and R^{17} are independently selected from the group consisting of hydrogen and C_1 - C_6 alkyl. Preferably, R^{16} and R^{17} are independently selected from the group consisting of hydrogen and methyl.

Alternatively, R^{16} and R^{17} together with the nitrogen atom to which they are attached form a 4-
30 to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N, O and S. Preferably, R^{16} and R^{17} together with the nitrogen atom to which they are attached form a 5- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N and O. More preferably, R^{16} and R^{17} together with the nitrogen atom to which they are attached form an pyrrolidyl, oxazolidinyl, imidazolidinyl, piperidyl, piperazinyl or morpholinyl group.

35 R^{18} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkoxy, - $N(R^6)_2$ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different. Preferably, R^{18} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkoxy, - $N(R^6)_2$ and phenyl. More preferably, R^{18} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl and C_1 - C_6 haloalkyl. Further more preferably, R^{18} is selected from the
40 group consisting of C_1 - C_6 alkyl and C_1 - C_6 haloalkyl. Most preferably, R^{18} is methyl or trifluoromethyl.

r is 0, 1 or 2. Preferably, r is 0 or 2.

In a set of preferred embodiments, in a compound according to formula (I) of the invention: R¹ is hydrogen or C₁-C₆alkyl; R² is hydrogen or methyl; Q is (CR^{1a}R^{2b})_m; m is 0, 1 or 2; R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, C₁-C₆alkyl, -OH and -NH₂; R³ is hydrogen or methyl; each R⁶ is independently selected from hydrogen and methyl; each R⁷ is C₁-C₆alkyl;

5 A is a 5-membered heteroaryl attached to the rest of the molecule via a ring carbon atom, said 5-membered heteroaryl heteroaryl comprising 1, 2, 3 or 4 heteroatoms independently selected from the group consisting of N, O and S, and wherein said heteroaryl is optionally substituted, where feasible, by 1, 2 or 3 R⁸ substituents, which may be the same or different; and when A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of halogen, nitro, cyano,

10 -NH₂, -NHR⁷, -N(R⁷)₂, -OH, -OR⁷, -S(O)_nR¹⁵, -NR⁶S(O)₂R¹⁵, -C(O)OR¹⁰, -C(O)R¹⁵, -C(O)NR¹⁶R¹⁷, -S(O)₂NR¹⁶R¹⁷, C₁-C₆alkyl and C₁-C₆haloalkyl; and/or when A is substituted on a ring nitrogen atom, each R⁸ is selected from the group consisting of -OR⁷, C₁-C₆alkyl and C₁-C₆haloalkyl; and n is 0; Z is selected from the group consisting of -C(O)OR¹⁰, -C(O)NHS(O)₂R¹², -S(O)₂OR¹⁰, -OS(O)₂OR¹⁰ and -P(O)(R¹³)(OR¹⁰); R¹⁰ is selected from the group consisting of hydrogen, C₁-C₆alkyl, phenyl and benzyl;

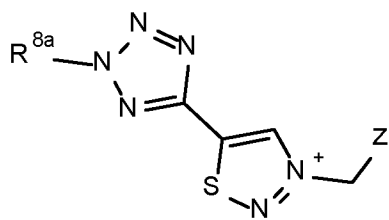
15 R¹² is selected from the group consisting of C₁-C₆alkyl, C₁-C₆haloalkyl and -N(R⁶)₂; R¹³ is selected from the group consisting of -OH and C₁-C₆alkoxy; R¹⁵ is C₁-C₆alkyl; R¹⁶ and R¹⁷ are independently selected from the group consisting of hydrogen and methyl; and r is 0 or 2.

More preferably in said preferred set of embodiments, R¹ is hydrogen or methyl; R² is hydrogen or methyl; Q is (CR^{1a}R^{2b})_m; m is 1 or 2; R^{1a} and R^{2b} are independently selected from the group consisting

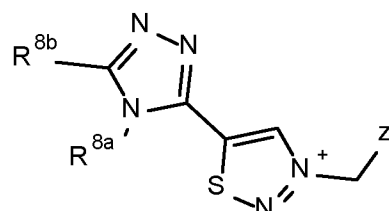
20 of hydrogen and methyl; R³ is hydrogen; A is a heteroaryl selected from the group consisting of tetrazol-5-yl, 1,2,4-triazol-3-yl, 1,2,4-triazol-5-yl, isoxazol-3-yl, oxazol-2-yl, thiazol-2-yl, 1,3,4-thiadiazol-2-yl, triazol-4-yl, triazol-5-yl, pyrazol-3-yl, pyrazol-5-yl, 1,3,4-oxadiazol-2-yl, 1,2,4-thiadiazol-5-yl, oxazol-4-yl, imidazol-2-yl, isothiazol-5-yl, 2-thienyl, 3-furyl, 2-furyl, isothiazol-4-yl, thiazol-4-yl, 3-thienyl, imidazol-5-yl, isoxazol-5-yl and 1,2,4-oxadiazol-5-yl wherein said heteroaryl is optionally substituted by 1, 2 or 3

25 R⁸ substituents, which may be the same or different; and when A is substituted on one or more ring carbon atoms, each R⁸ is independently selected from the group consisting of chloro, -NH₂, -NHMe, -OMe, methyl, iso-propyl and trifluoromethyl; and/or when A is substituted on a ring nitrogen atom, R⁸ is C₁-C₆alkyl; n is 0; Z is selected from the group consisting of -C(O)OR¹⁰, -S(O)₂OR¹⁰, and -OS(O)₂OR¹⁰; and R¹⁰ is hydrogen or C₁-C₆alkyl.

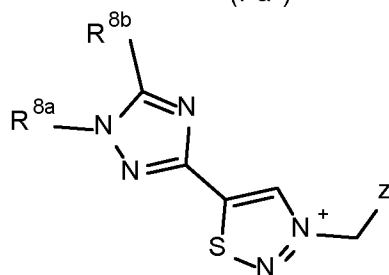
30 In a further set of preferred embodiments, the compound according to formula (I) is selected from the group consisting of a compound of formula (I-a*), (I-b*), (I-c*), (I-d*), (I-e*), (I-f*), (I-g*), (I-h*), (I-a), (I-b), (I-c), (I-d), (I-e), (I-f), (I-g), (I-h), (I-j), (I-k), (I-l), (I-m), (I-n), (I-p), (I-q), (I-r) and (I-s), as shown below:



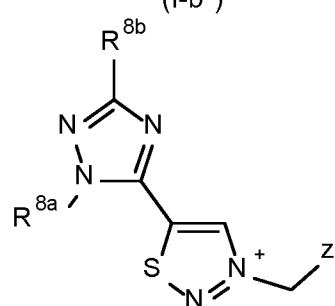
(I-a*)



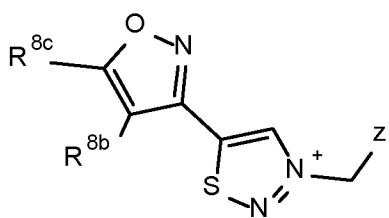
(I-b*)



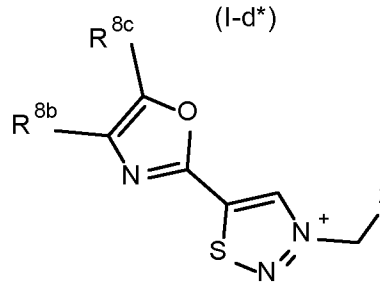
(I-c*)



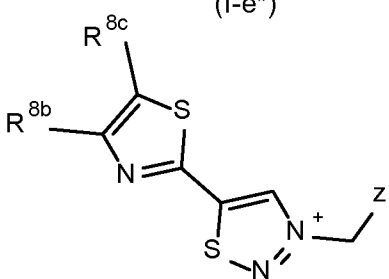
(I-d*)



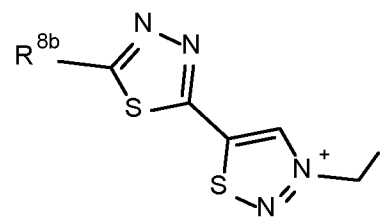
(I-e*)



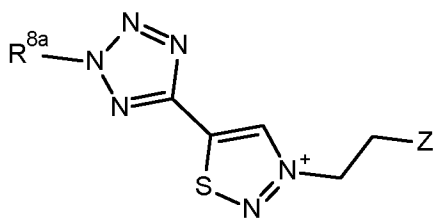
(I-f*)



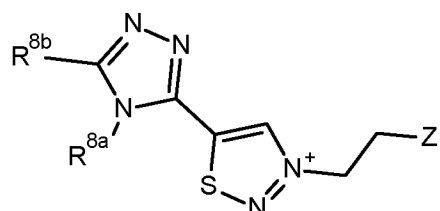
(I-g*)



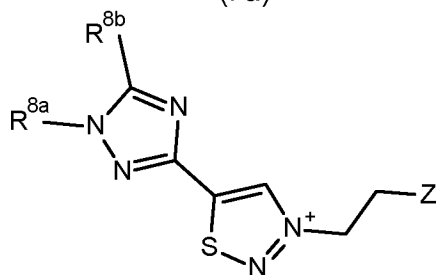
(I-h*)



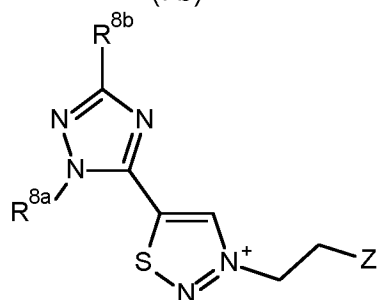
(I-a)



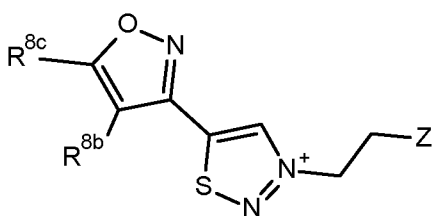
(I-b)



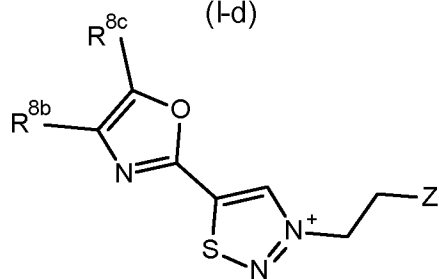
(I-c)



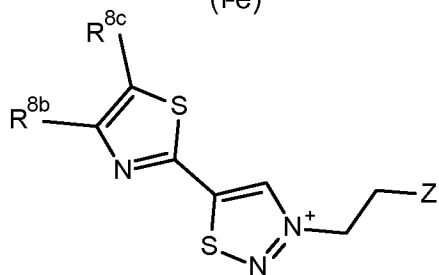
(I-d)



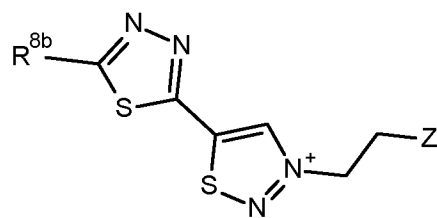
(I-e)



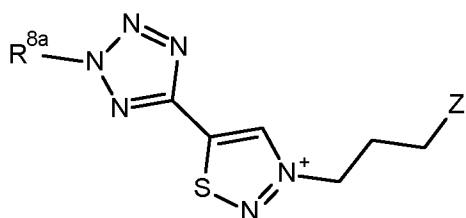
(I-f)



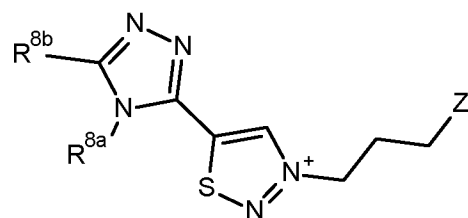
(I-g)



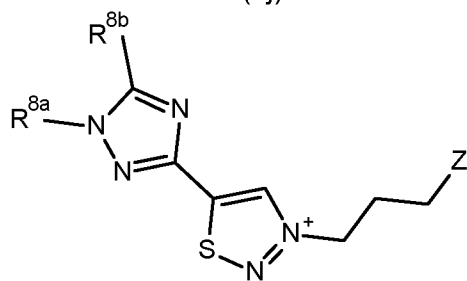
(I-h)



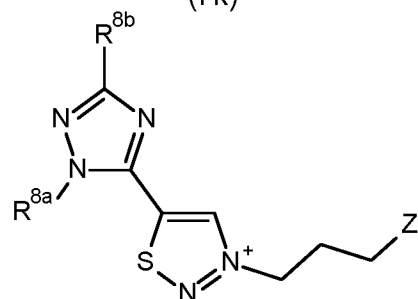
(I-j)



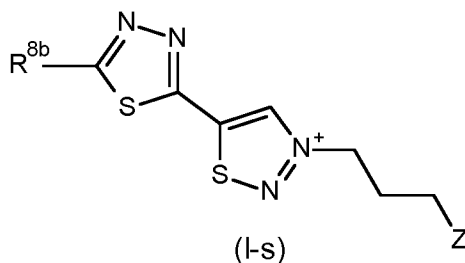
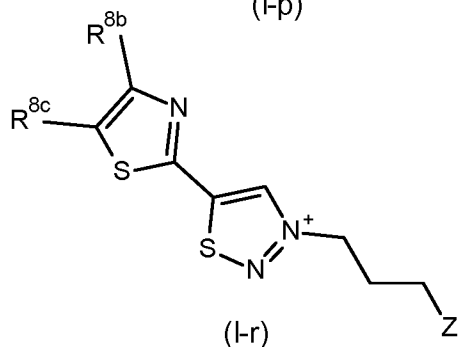
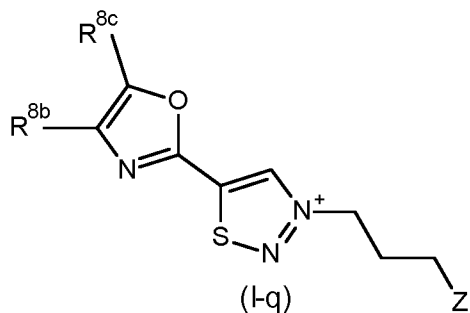
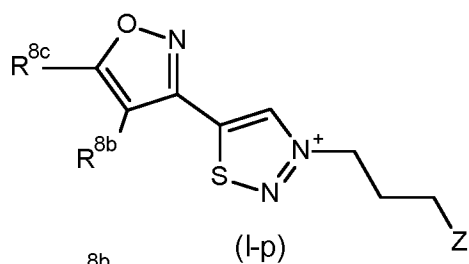
(I-k)



(I-m)



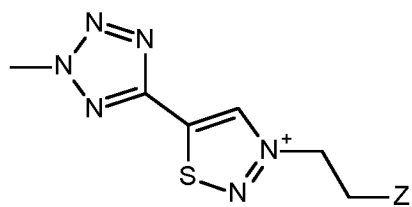
(I-n)



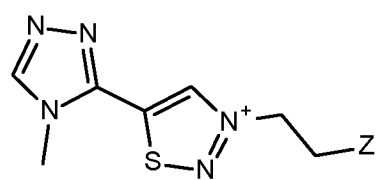
wherein in a compound of formula (I-a*), (I-b*), (I-c*), (I-d*), (I-e*), (I-f*), (I-g*), (I-h*), (I-a), (I-b), (I-c), (I-d), (I-e), (I-f), (I-g), (I-h), (I-j), (I-k), (I-m), (I-n), (I-p), (I-q), (I-r) and (I-s):

R^{8a} is hydrogen or C₁-C₆alkyl; each R^{8b}, R^{8c} and R^{8d} are independently selected from the group
 5 consisting of hydrogen, halogen, -NH₂, -NHR⁷, -N(R⁷)₂, -OR⁷, C₁-C₆alkyl and C₁-C₆haloalkyl; and Z is -C(O)OH or -S(O)₂OH. Within this set of embodiments, compounds of formula (I-d), (I-f), (I-g), (I-h), (I-n), (I-q) and (I-r) are particularly preferred.

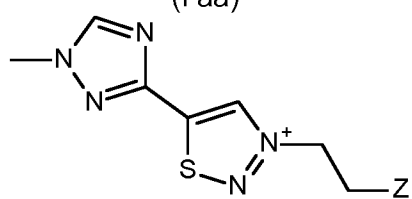
In a further more preferred set of embodiments, the compound according to formula (I) is
 selected from a compound of formula (I-aa), (I-bb), (I-cc), (I-dd), (I-ee), (I-ff), (I-gg), (I-hh), (I-jj), (I-kk), (I-
 10 mm), (I-nn), (I-pp), (I-qq), (I-rr) or (I-ss),



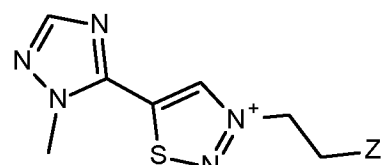
(I-aa)



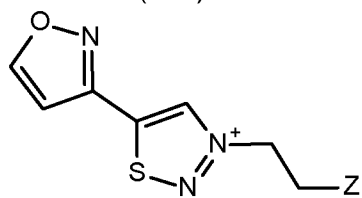
(I-bb)



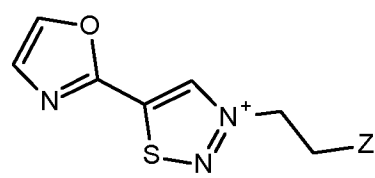
(I-cc)



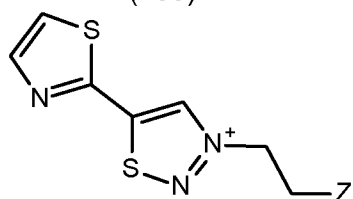
(I-dd)



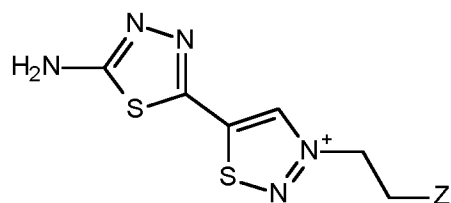
(I-ee)



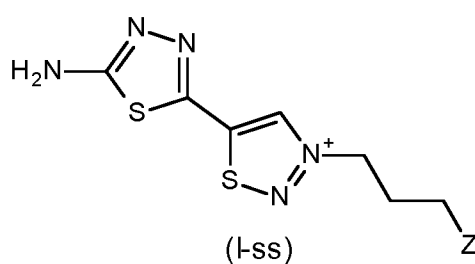
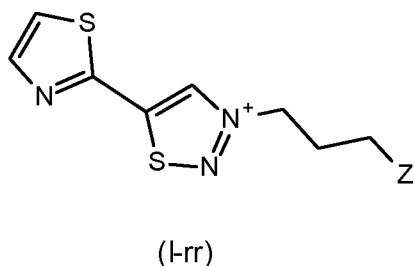
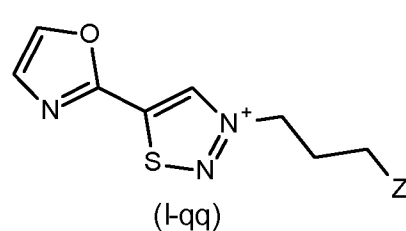
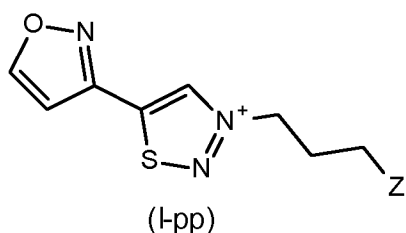
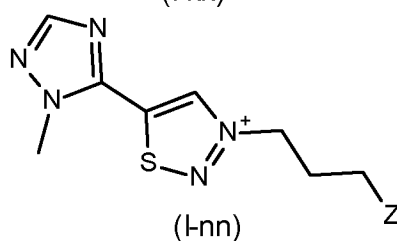
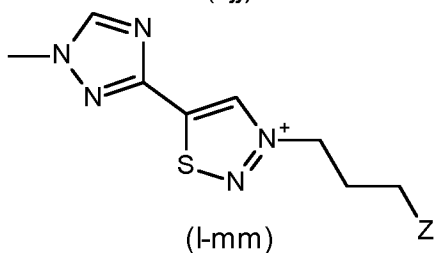
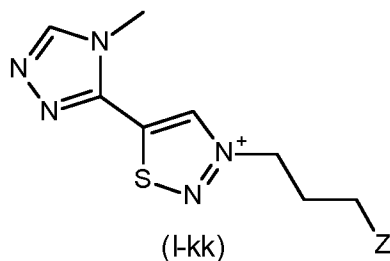
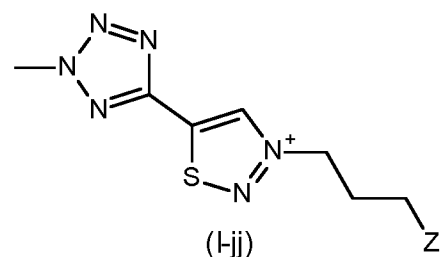
(I-ff)



(I-gg)



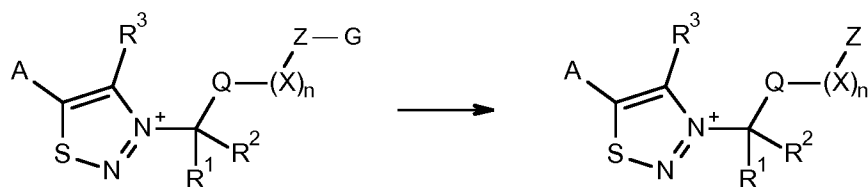
(I-hh)



wherein in a compound of formula (I-aa), (I-bb), (I-cc), (I-dd), (I-ee), (I-ff), (I-gg), (I-hh), (I-jj), (I-kk), (I-mm), (I-nn), (I-pp), (I-qq), (I-rr) or (I-ss), Z is -C(O)OH or -S(O)₂OH. Within this set of embodiments, 5 compounds of formula (I-dd), (I-ff), (I-gg), (I-hh), (I-nn), (I-qq), (I-rr) and (I-ss) are particularly preferred.

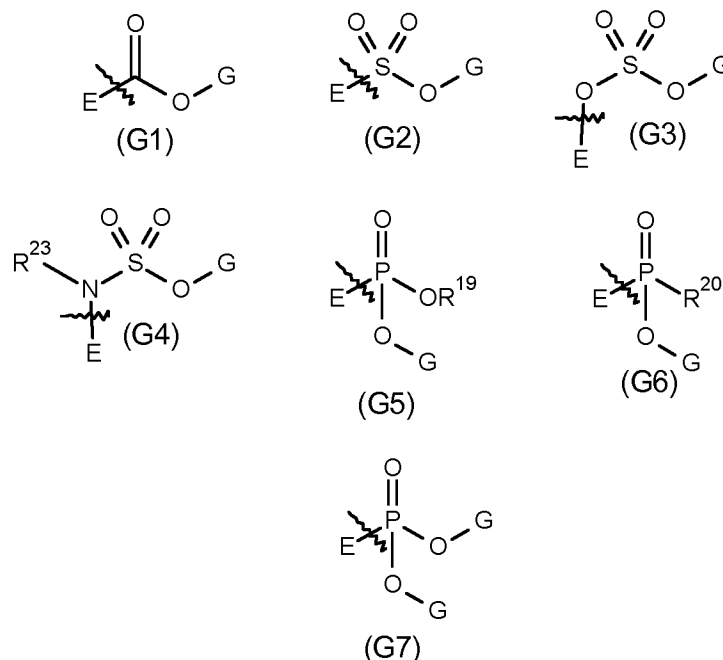
It should be understood that compounds of formula (I) may exist/be manufactured in 'procidal form', wherein they comprise a group 'G'. Such compounds are referred to herein as compounds of Formula (I-IV).

G is a group which may be removed in a plant by any appropriate mechanism including, but not 10 limited to, metabolism and chemical degradation to give a compound of Formula (I-I), (I-II) or (I-III) wherein Z contains an acidic proton, for example see the scheme below:



Whilst such G groups may be considered as 'procidal', and thus yield active herbicidal compounds 15 once removed, compounds comprising such groups may also exhibit herbicidal activity in their own right.

In such cases in a compound of Formula (I-IV), Z-G may include but is not limited to, any one of (G1) to (G7) below and E indicates the point of attachment to the remaining part of a compound of formula (I):



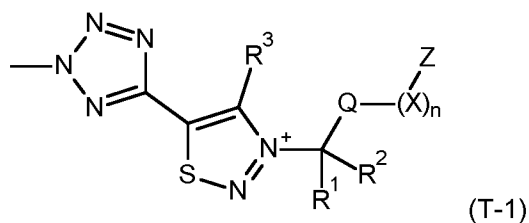
In embodiments where Z-G is (G1) to (G7), G, R¹⁹, R²⁰, R²¹, R²² and R²³ are defined as follows:

- 5 G is C₁-C₆alkyl, C₂-C₆alkenyl, C₂-C₆alkynyl, -C(R²¹R²²)OC(O)R¹⁹, phenyl or phenyl-C₁-C₄alkyl-, wherein said phenyl moiety is optionally substituted by 1 to 5 substituents independently selected from halo, cyano, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl or C₁-C₆alkoxy;
- R¹⁹ is C₁-C₆alkyl or phenyl;
- R²⁰ is hydroxy, C₁-C₆alkyl, C₁-C₆alkoxy or phenyl;
- 10 R²¹ is hydrogen or methyl;
- R²² is hydrogen or methyl;
- R²³ is hydrogen or C₁-C₆alkyl.

The compounds in Tables 1 to 39 below illustrate the compounds of the invention. The skilled person would understand that the compounds of formula (I) may exist as an agronomically acceptable salt, a zwitterion or an agronomically acceptable salt of a zwitterion as described hereinbefore.

Table 1:

This table discloses 53 specific compounds of the formula (T-1):



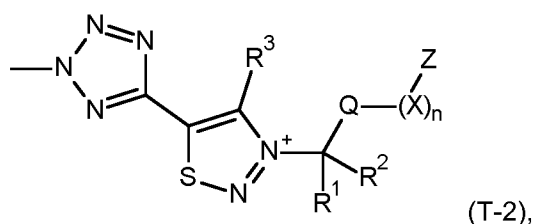
wherein m, Q, R³, and Z are as defined in Table 1, R¹ and R² are hydrogen and n is 0.

Table 1

Compound number	R ³	Z	m	Q
1.001	H	-C(O)OH	0	-
1.002	H	-C(O)OMe	0	-
1.003	H	-C(O)NHOMe	0	-
1.004	H	-OC(O)NHOMe	0	-
1.005	H	-NHC(O)NHOMe	0	-
1.006	H	-NMeC(O)NHOMe	0	-
1.007	H	-C(O)NHS(O) ₂ Me	0	-
1.008	H	-OC(O)NHS(O) ₂ Me	0	-
1.009	H	-NHC(O)NHS(O) ₂ Me	0	-
1.010	H	-NMeC(O)NHS(O) ₂ Me	0	-
1.011	H	-S(O) ₂ OH	0	-
1.012	H	-OS(O) ₂ OH	0	-
1.013	H	-NHS(O) ₂ OH	0	-
1.014	H	-NMeS(O) ₂ OH	0	-
1.015	H	-S(O)OH	0	-
1.016	H	-OS(O)OH	0	-
1.017	H	-NHS(O)OH	0	-
1.018	H	-NMeS(O)OH	0	-
1.019	H	-NHS(O) ₂ CF ₃	0	-
1.020	H	-S(O) ₂ NHC(O)Me	0	-
1.021	H	-OS(O) ₂ NHC(O)Me	0	-
1.022	H	-NHS(O) ₂ NHC(O)Me	0	-
1.023	H	-NMeS(O) ₂ NHC(O)Me	0	-
1.024	H	-P(O)(OH)(OMe)	0	-
1.025	H	-P(O)(OH)(OH)	0	-
1.026	H	-OP(O)(OH)(OMe)	0	-
1.027	H	-OP(O)(OH)(OH)	0	-
1.028	H	-NHP(O)(OH)(OMe)	0	-
1.029	H	-NHP(O)(OH)(OH)	0	-
1.030	H	-NMeP(O)(OH)(OMe)	0	-
1.031	H	-NMeP(O)(OH)(OH)	0	-
1.032	H	-tetrazole	0	-
1.033	H	-S(O) ₂ OH	1	CH(NH ₂)
1.033	H	-C(O)OH	1	CH(NH ₂)
1.035	H	-S(O) ₂ OH	2	CH(OH)CH ₂
1.036	H	-C(O)OH	2	CH(OH)CH ₂
1.037	H	-S(O) ₂ OH	1	CH(OH)
1.038	H	-C(O)OH	1	CH(OH)
1.039	H	-C(O)NHCN	0	-
1.040	H	-OC(O)NHCN	0	-
1.041	H	-NHC(O)NHCN	0	-
1.042	H	-NMeC(O)NHCN	0	-
1.043	H	-S(O) ₂ NHCN	0	-
1.044	H	-OS(O) ₂ NHCN	0	-
1.045	H	-NHS(O) ₂ NHCN	0	-
1.046	H	-NMeS(O) ₂ NHCN	0	-
1.047	H	-S(O) ₂ NHS(O) ₂ Me	0	-
1.048	H	-OS(O) ₂ NHS(O) ₂ Me	0	-
1.049	H	-NHS(O) ₂ NHS(O) ₂ Me	0	-
1.050	H	-NMeS(O) ₂ NHS(O) ₂ Me	0	-
1.051	H	-P(O)H(OH)	0	-
1.052	H	-N(OH)C(O)Me	0	-
1.053	H	-ONHC(O)Me	0	-

Table 2:

This table discloses 49 specific compounds of the formula (T-2):



5 wherein m, Q, R³, and Z are as defined in Table 2, R¹ and R² are hydrogen and n is 0.

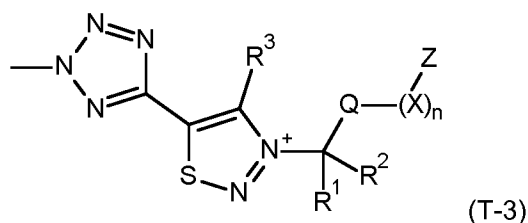
Table 2

Compound number	R ³	Z	m	Q
2.001	H	-C(O)OH	1	CH ₂
2.002	H	-C(O)OMe	1	CH ₂
2.003	H	-C(O)NHOMe	1	CH ₂
2.004	H	-OC(O)NHOMe	1	CH ₂
2.005	H	-NHC(O)NHOMe	1	CH ₂
2.006	H	-NMeC(O)NHOMe	1	CH ₂
2.007	H	-C(O)NHS(O) ₂ Me	1	CH ₂
2.008	H	-OC(O)NHS(O) ₂ Me	1	CH ₂
2.009	H	-NHC(O)NHS(O) ₂ Me	1	CH ₂
2.010	H	-NMeC(O)NHS(O) ₂ Me	1	CH ₂
2.011	H	-S(O) ₂ OH	1	CH ₂
2.012	H	-OS(O) ₂ OH	1	CH ₂
2.013	H	-NHS(O) ₂ OH	1	CH ₂
2.014	H	-NMeS(O) ₂ OH	1	CH ₂
2.015	H	-S(O)OH	1	CH ₂
2.016	H	-OS(O)OH	1	CH ₂
2.017	H	-NHS(O)OH	1	CH ₂
2.018	H	-NMeS(O)OH	1	CH ₂
2.019	H	-NHS(O) ₂ CF ₃	1	CH ₂
2.020	H	-S(O) ₂ NHC(O)Me	1	CH ₂
2.021	H	-OS(O) ₂ NHC(O)Me	1	CH ₂
2.022	H	-NHS(O) ₂ NHC(O)Me	1	CH ₂
2.023	H	-NMeS(O) ₂ NHC(O)Me	1	CH ₂
2.024	H	-P(O)(OH)(OMe)	1	CH ₂
2.025	H	-P(O)(OH)(OH)	1	CH ₂
2.026	H	-OP(O)(OH)(OMe)	1	CH ₂
2.027	H	-OP(O)(OH)(OH)	1	CH ₂
2.028	H	-NHP(O)(OH)(OMe)	1	CH ₂
2.029	H	-NHP(O)(OH)(OH)	1	CH ₂
2.030	H	-NMeP(O)(OH)(OMe)	1	CH ₂
2.031	H	-NMeP(O)(OH)(OH)	1	CH ₂
2.032	H	-tetrazole	1	CH ₂
2.033	H	-S(O) ₂ OH	2	CH ₂ CH(NH ₂)
2.034	H	-C(O)OH	2	CH ₂ CH(NH ₂)
2.035	H	-C(O)NHCN	1	CH ₂
2.036	H	-OC(O)NHCN	1	CH ₂
2.037	H	-NHC(O)NHCN	1	CH ₂
2.038	H	-NMeC(O)NHCN	1	CH ₂
2.039	H	-S(O) ₂ NHCN	1	CH ₂
2.040	H	-OS(O) ₂ NHCN	1	CH ₂
2.041	H	-NHS(O) ₂ NHCN	1	CH ₂
2.042	H	-NMeS(O) ₂ NHCN	1	CH ₂

Compound number	R ³	Z	m	Q
2.043	H	-S(O) ₂ NHS(O) ₂ Me	1	CH ₂
2.044	H	-OS(O) ₂ NHS(O) ₂ Me	1	CH ₂
2.045	H	-NHS(O) ₂ NHS(O) ₂ Me	1	CH ₂
2.046	H	-NMeS(O) ₂ NHS(O) ₂ Me	1	CH ₂
2.047	H	-P(O)H(OH)	1	CH ₂
2.048	H	-N(OH)C(O)Me	1	CH ₂
2.049	H	-ONHC(O)Me	1	CH ₂

Table 3:

This table discloses 49 specific compounds of the formula (T-3):



5 wherein m, Q, R³, and Z are as defined in Table 3, R¹ and R² are hydrogen and n is 0.

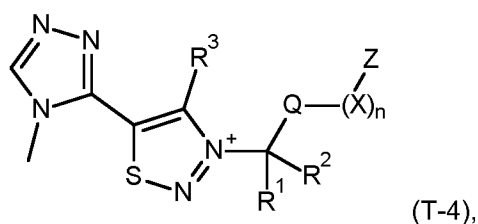
Table 3

Compound number	R ³	Z	m	Q
3.001	H	-C(O)OH	2	CH ₂ CH ₂
3.002	H	-C(O)OMe	2	CH ₂ CH ₂
3.003	H	-C(O)NHOMe	2	CH ₂ CH ₂
3.004	H	-OC(O)NHOMe	2	CH ₂ CH ₂
3.005	H	-NHC(O)NHOMe	2	CH ₂ CH ₂
3.006	H	-NMeC(O)NHOMe	2	CH ₂ CH ₂
3.007	H	-C(O)NHS(O) ₂ Me	2	CH ₂ CH ₂
3.008	H	-OC(O)NHS(O) ₂ Me	2	CH ₂ CH ₂
3.009	H	-NHC(O)NHS(O) ₂ Me	2	CH ₂ CH ₂
3.010	H	-NMeC(O)NHS(O) ₂ Me	2	CH ₂ CH ₂
3.011	H	-S(O) ₂ OH	2	CH ₂ CH ₂
3.012	H	-OS(O) ₂ OH	2	CH ₂ CH ₂
3.013	H	-NHS(O) ₂ OH	2	CH ₂ CH ₂
3.014	H	-NMeS(O) ₂ OH	2	CH ₂ CH ₂
3.015	H	-S(O)OH	2	CH ₂ CH ₂
3.016	H	-OS(O)OH	2	CH ₂ CH ₂
3.017	H	-NHS(O)OH	2	CH ₂ CH ₂
3.018	H	-NMeS(O)OH	2	CH ₂ CH ₂
3.019	H	-NHS(O) ₂ CF ₃	2	CH ₂ CH ₂
3.020	H	-S(O) ₂ NHC(O)Me	2	CH ₂ CH ₂
3.021	H	-OS(O) ₂ NHC(O)Me	2	CH ₂ CH ₂
3.022	H	-NHS(O) ₂ NHC(O)Me	2	CH ₂ CH ₂
3.023	H	-NMeS(O) ₂ NHC(O)Me	2	CH ₂ CH ₂
3.024	H	-P(O)(OH)(OMe)	2	CH ₂ CH ₂
3.025	H	-P(O)(OH)(OH)	2	CH ₂ CH ₂
3.026	H	-OP(O)(OH)(OMe)	2	CH ₂ CH ₂
3.027	H	-OP(O)(OH)(OH)	2	CH ₂ CH ₂
3.028	H	-NHP(O)(OH)(OMe)	2	CH ₂ CH ₂
3.029	H	-NHP(O)(OH)(OH)	2	CH ₂ CH ₂
3.030	H	-NMeP(O)(OH)(OMe)	2	CH ₂ CH ₂
3.031	H	-NMeP(O)(OH)(OH)	2	CH ₂ CH ₂
3.032	H	-tetrazole	2	CH ₂ CH ₂
3.033	H	-S(O) ₂ OH	3	CH ₂ CH ₂ CH(NH ₂)

Compound number	R ³	Z	m	Q
3.034	H	-C(O)OH	3	CH ₂ CH ₂ CH(NH ₂)
3.035	H	-C(O)NHCN	2	CH ₂ CH ₂
3.036	H	-OC(O)NHCN	2	CH ₂ CH ₂
3.037	H	-NHC(O)NHCN	2	CH ₂ CH ₂
3.038	H	-NMeC(O)NHCN	2	CH ₂ CH ₂
3.039	H	-S(O) ₂ NHCN	2	CH ₂ CH ₂
3.040	H	-OS(O) ₂ NHCN	2	CH ₂ CH ₂
3.041	H	-NHS(O) ₂ NHCN	2	CH ₂ CH ₂
3.042	H	-NMeS(O) ₂ NHCN	2	CH ₂ CH ₂
3.043	H	-S(O) ₂ NHS(O) ₂ Me	2	CH ₂ CH ₂
3.044	H	-OS(O) ₂ NHS(O) ₂ Me	2	CH ₂ CH ₂
3.045	H	-NHS(O) ₂ NHS(O) ₂ Me	2	CH ₂ CH ₂
3.046	H	-NMeS(O) ₂ NHS(O) ₂ Me	2	CH ₂ CH ₂
3.047	H	-P(O)H(OH)	2	CH ₂ CH ₂
3.048	H	-N(OH)C(O)Me	2	CH ₂ CH ₂
3.049	H	-ONHC(O)Me	2	CH ₂ CH ₂

Table 4:

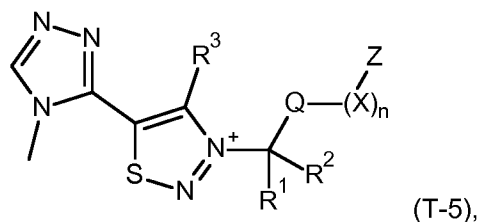
This table discloses 53 specific compounds of the formula (T-4):



5 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 5:

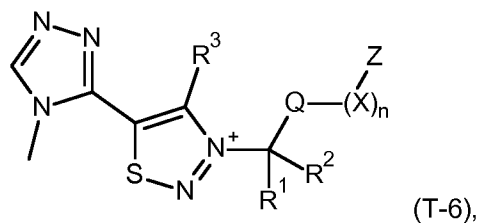
This table discloses 49 specific compounds of the formula (T-5):



10 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 6:

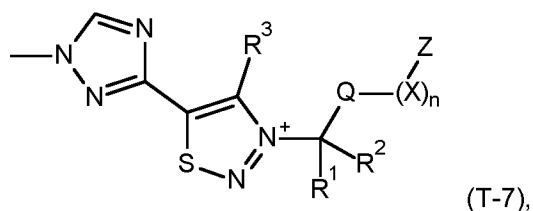
This table discloses 49 specific compounds of the formula (T-6):



15 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 7:

This table discloses 53 specific compounds of the formula (T-7):

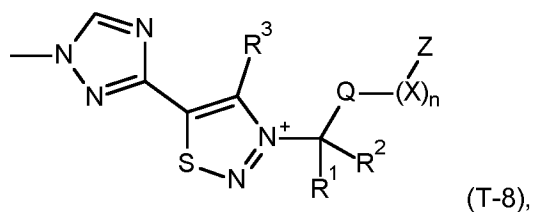


wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

5

Table 8:

This table discloses 49 specific compounds of the formula (T-8):

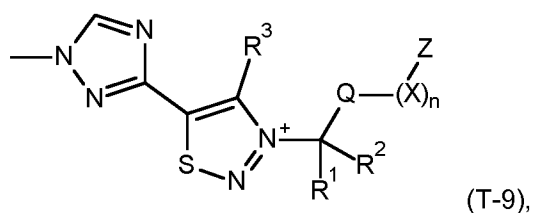


wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

10

Table 9:

This table discloses 49 specific compounds of the formula (T-9):

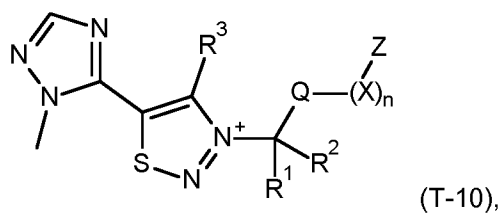


wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

15

Table 10:

This table discloses 53 specific compounds of the formula (T-10):



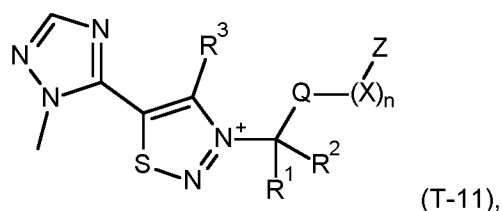
wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

20

25

Table 11:

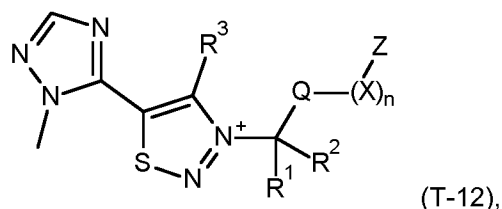
This table discloses 49 specific compounds of the formula (T-11):



wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

5 Table 12:

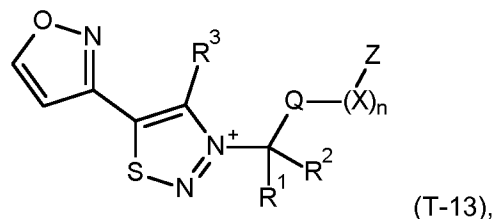
This table discloses 49 specific compounds of the formula (T-12):



wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

10 Table 13:

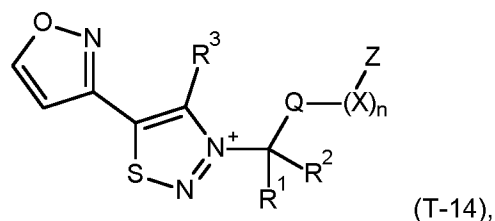
This table discloses 53 specific compounds of the formula (T-13):



wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

15 Table 14:

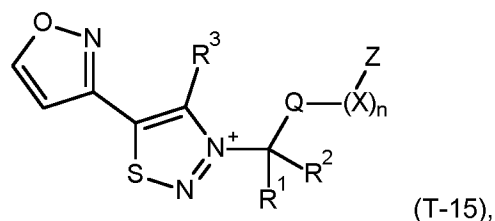
This table discloses 49 specific compounds of the formula (T-14):



wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 15:

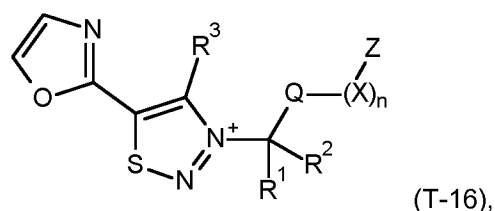
This table discloses 49 specific compounds of the formula (T-15):



5 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 16:

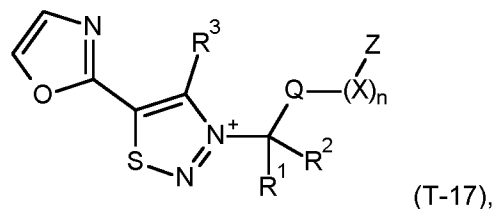
This table discloses 53 specific compounds of the formula (T-16):



10 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 17:

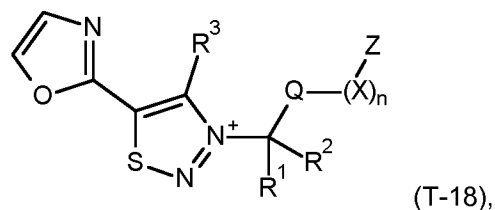
This table discloses 49 specific compounds of the formula (T-17):



15 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 18:

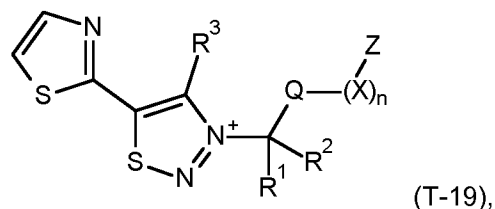
This table discloses 49 specific compounds of the formula (T-18):



20 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 19:

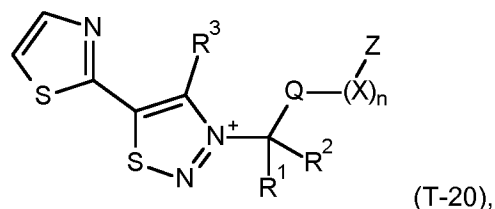
This table discloses 53 specific compounds of the formula (T-19):



5 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 20:

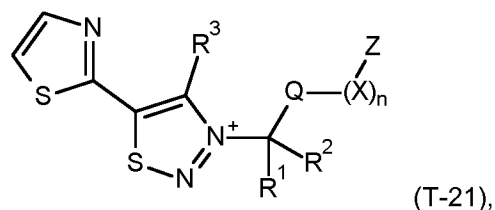
This table discloses 49 specific compounds of the formula (T-20):



10 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 21:

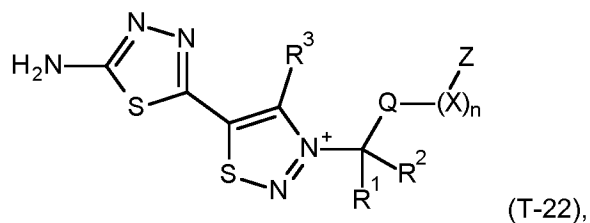
This table discloses 49 specific compounds of the formula (T-21):



15 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 22:

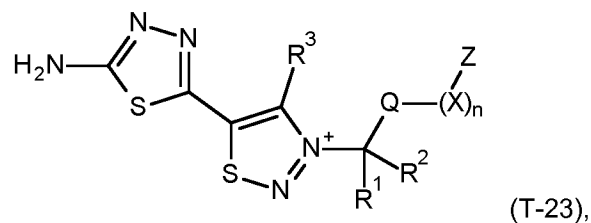
This table discloses 53 specific compounds of the formula (T-22):



20 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 23:

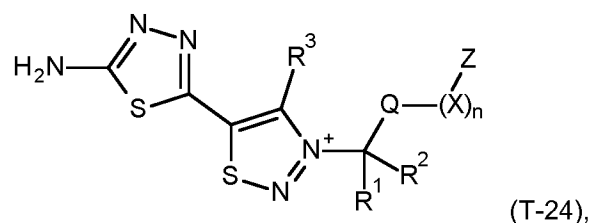
This table discloses 49 specific compounds of the formula (T-23):



5 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 24:

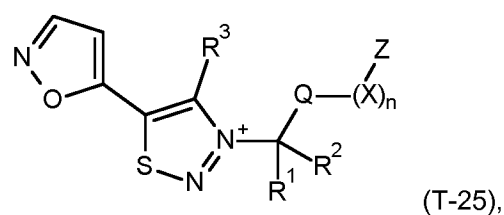
This table discloses 49 specific compounds of the formula (T-24):



10 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 25:

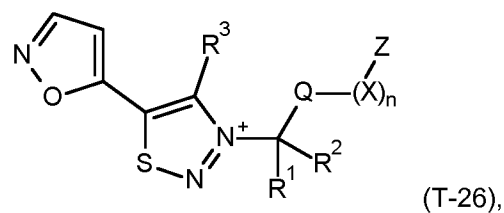
This table discloses 53 specific compounds of the formula (T-25):



15 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 26:

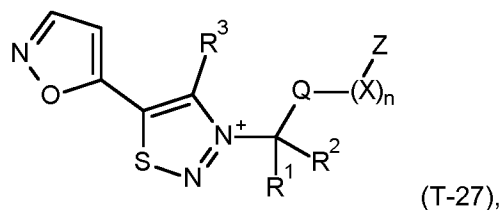
This table discloses 49 specific compounds of the formula (T-26):



20 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 27:

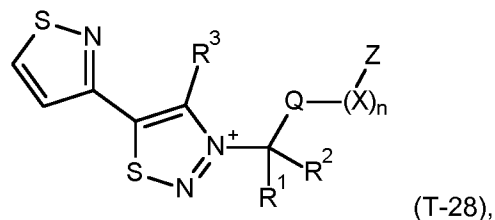
This table discloses 49 specific compounds of the formula (T-27):



- 5 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 28:

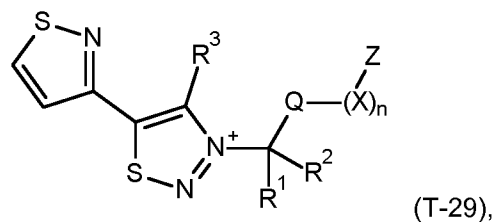
This table discloses 53 specific compounds of the formula (T-28):



- 10 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 29:

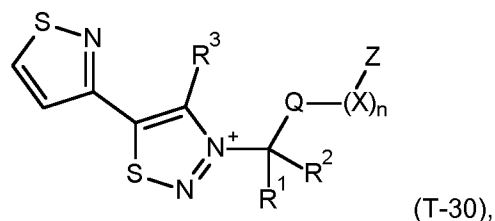
This table discloses 49 specific compounds of the formula (T-29):



- 15 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 30:

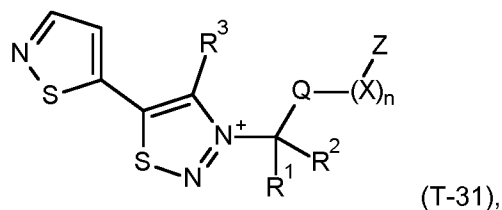
This table discloses 49 specific compounds of the formula (T-30):



- 20 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 31:

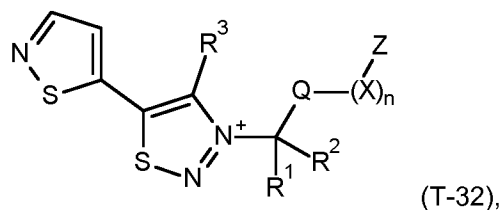
This table discloses 53 specific compounds of the formula (T-31):



5 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 32:

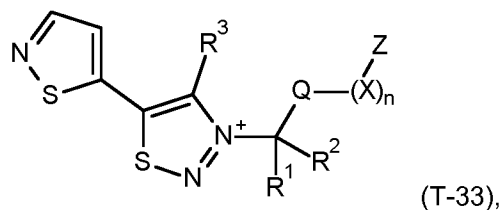
This table discloses 49 specific compounds of the formula (T-32):



10 wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

Table 33:

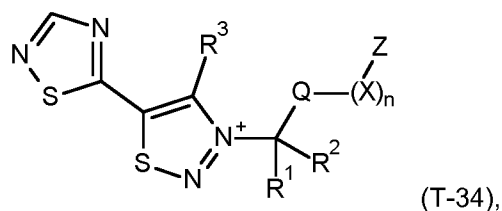
This table discloses 49 specific compounds of the formula (T-33):



15 wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

Table 34:

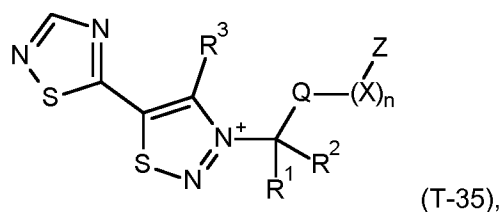
This table discloses 53 specific compounds of the formula (T-34):



20 wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

Table 35:

This table discloses 49 specific compounds of the formula (T-35):

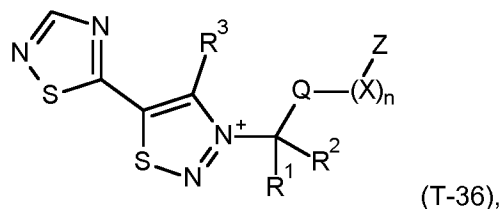


wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

5

Table 36:

This table discloses 49 specific compounds of the formula (T-36):

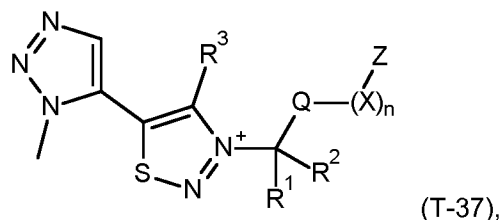


wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

10

Table 37:

This table discloses 53 specific compounds of the formula (T-37):

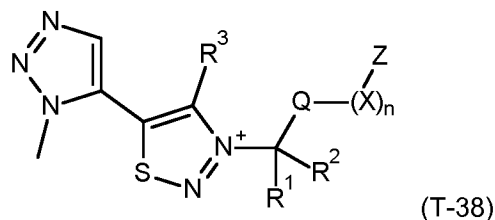


wherein m, Q, R³, and Z are as defined above in Table 1, R¹ and R² are hydrogen and n is 0.

15

Table 38:

This table discloses 49 specific compounds of the formula (T-38):

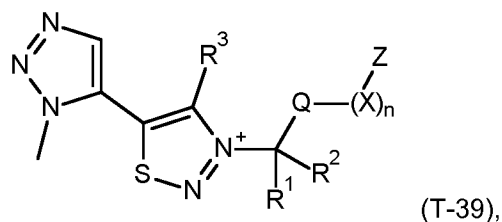


wherein m, Q, R³, and Z are as defined above in Table 2, R¹ and R² are hydrogen and n is 0.

20

Table 39:

This table discloses 49 specific compounds of the formula (T-39):



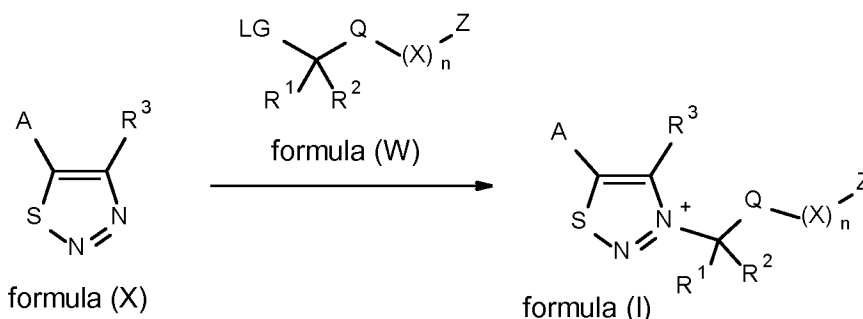
wherein m, Q, R³, and Z are as defined above in Table 3, R¹ and R² are hydrogen and n is 0.

The compounds of the present invention may be prepared according to the following schemes in which the substituents n, m, r, A, Q, X, Z, R¹, R², R^{1a}, R^{2b}, R², R³, R⁶, R⁷, R^{7a}, R^{7b}, R^{7c}, R⁸, R⁹, R¹⁰, R¹¹, R¹², R¹³, R¹⁴, R¹⁵, R^{15a}, R¹⁶, R¹⁷ and R¹⁸ are as defined hereinbefore unless explicitly stated

5 otherwise. The compounds of the preceeding Tables 1 to 39 may thus be obtained in an analogous manner.

The compounds of formula (I) may be prepared by the alkylation of compounds of formula (X), wherein R³ and A are as defined for compounds of formula (I), with a suitable alkylating agent of formula (W), wherein R¹, R², Q, X, n and Z are as defined for compounds of formula (I) and LG is a suitable
10 leaving group, for example, halide or pseudohalide such as triflate, mesylate or tosylate, in a suitable solvent at a suitable temperature, as described in reaction scheme 1.

Reaction scheme 1



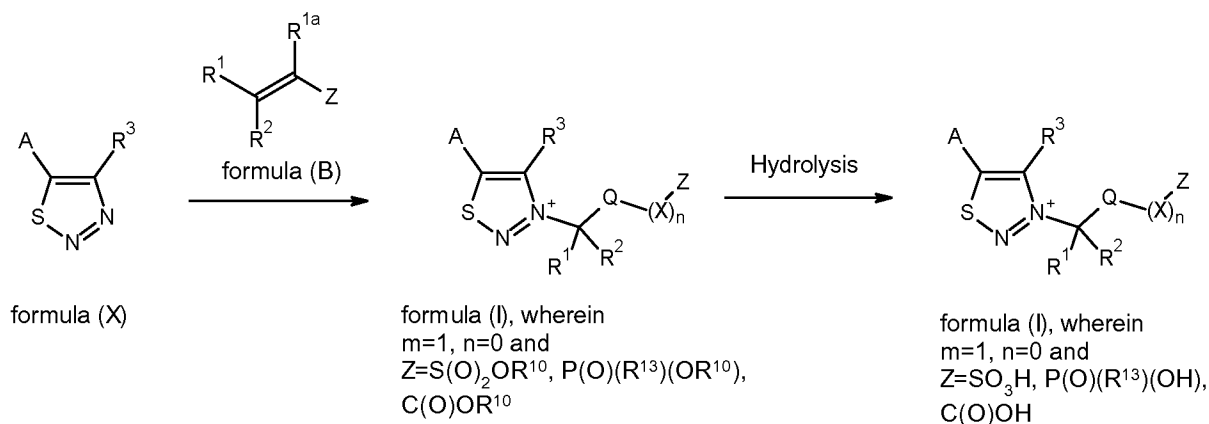
15 Example conditions include stirring a compound of formula (X) with an alkylating agent of formula (W) in a solvent, or mixture of solvents, such as acetone, dichloromethane, dichloroethane, *N,N*-dimethylformamide, acetonitrile, 1,4-dioxane, water, acetic acid or trifluoroacetic acid at a temperature between -78°C and 150°C. An alkylating agent of formula (W) may include, but is not limited to, bromoacetic acid, methyl bromoacetate, 3-bromopropionic acid, methyl 3-bromopropionate, 2-bromo-
20 *N*-methoxyacetamide, sodium 2-bromoethanesulphonate, 2,2-dimethylpropyl 2-(trifluoromethylsulfonyloxy)ethanesulfonate, 2-bromo-*N*-methanesulfonylacetamide, 3-bromo-*N*-methanesulfonylpropanamide, dimethoxyphosphorylmethyl trifluoromethanesulfonate, dimethyl 3-bromopropylphosphonate, 3-chloro-2,2-dimethyl-propanoic acid, diethyl 2-bromoethylphosphonate and ethyl 3-(trifluoromethylsulfonyloxy)propanoate. Such alkylating agents and related compounds are
25 either known in the literature or may be prepared by known literature methods. Compounds of formula (I) which may be described as esters of *N*-alkyl acids, which include, but are not limited to, esters of carboxylic acids, phosphonic acids, phosphinic acids, sulfonic acids and sulfinic acids, may be subsequently partially or fully hydrolysed by treatment with a suitable reagent, for example, aqueous hydrochloric acid or trimethylsilyl bromide, in a suitable solvent at a suitable temperature between 0°C
30 and 100°C.

Additionally, compounds of formula (I) may be prepared by reacting compounds of formula (X), wherein R³ and A are as defined for compounds of formula (I), with a suitably activated electrophilic alkene of formula (B), wherein Z is -S(O)₂OR¹⁰, -P(O)(R¹³)(OR¹⁰) or -C(O)OR¹⁰ and R¹, R², R^{1a}, R¹⁰ and R¹³ are as defined for compounds of formula (I), in a suitable solvent at a suitable temperature.

35 Compounds of formula (B) are known in the literature, or may be prepared by known methods. Example

reagents include, but are not limited to, acrylic acid, methacrylic acid, crotonic acid, 3,3-dimethylacrylic acid, methyl acrylate, ethene sulfonic acid, isopropyl ethylenesulfonate, 2,2-dimethylpropyl ethylenesulfonate and dimethyl vinylphosphonate. The direct products of these reactions, which may be described as esters of N-alkyl acids, which include, but are not limited to, esters of carboxylic acids, 5 phosphonic acids, phosphinic acids, sulfonic acids and sulfinic acids, may be subsequently partially or fully hydrolysed by treatment with a suitable reagent in a suitable solvent at a suitable temperature, as described in reaction scheme 2.

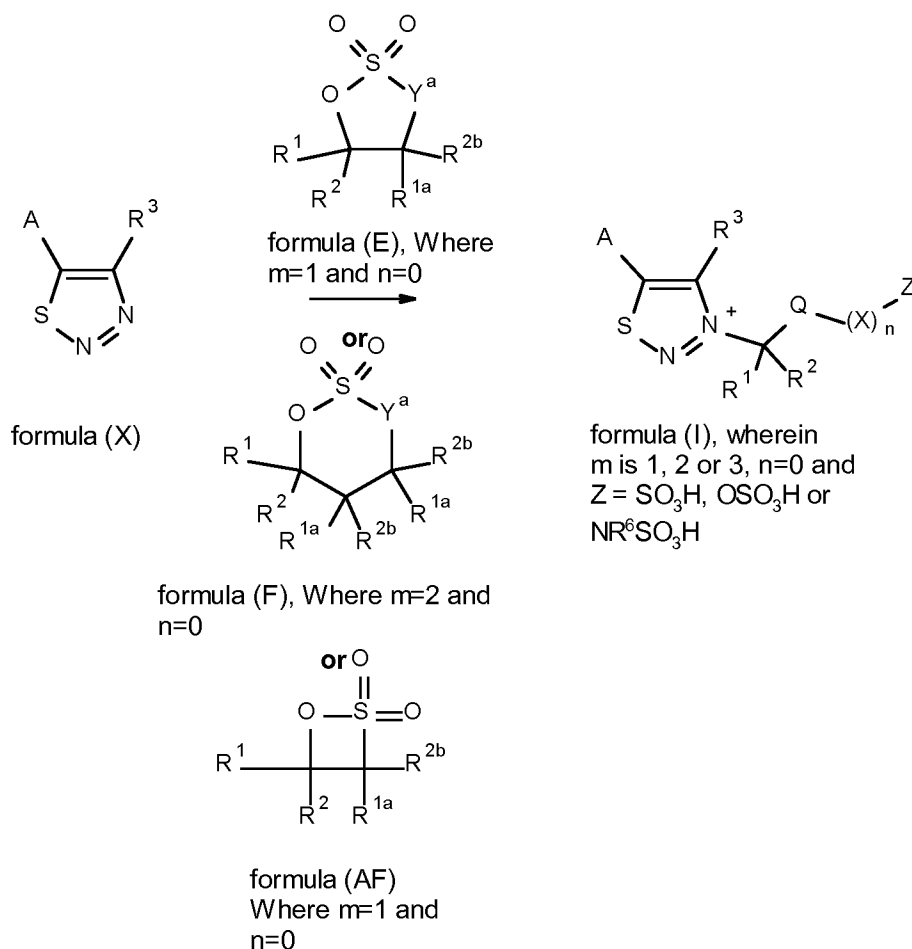
Reaction scheme 2



10

In a related reaction compounds of formula (I), wherein Q is $\text{C}(\text{R}^{1a}\text{R}^{2b})$, m is 1, 2 or 3, $n=0$ and Z is $-\text{S}(\text{O})_2\text{OH}$, $-\text{OS}(\text{O})_2\text{OH}$ or $-\text{NR}^6\text{S}(\text{O})_2\text{OH}$, may be prepared by the reaction of compounds of formula (X), wherein R³ and A are as defined for compounds of formula (I), with a cyclic alkylating agent of formula (E), (F) or (AF), wherein Y^a is $\text{C}(\text{R}^{1a}\text{R}^{2b})$, O or NR^6 and R¹, R², R^{1a} and R^{2b} are as defined for 15 compounds of formula (I), in a suitable solvent at a suitable temperature, as described in reaction scheme 3.

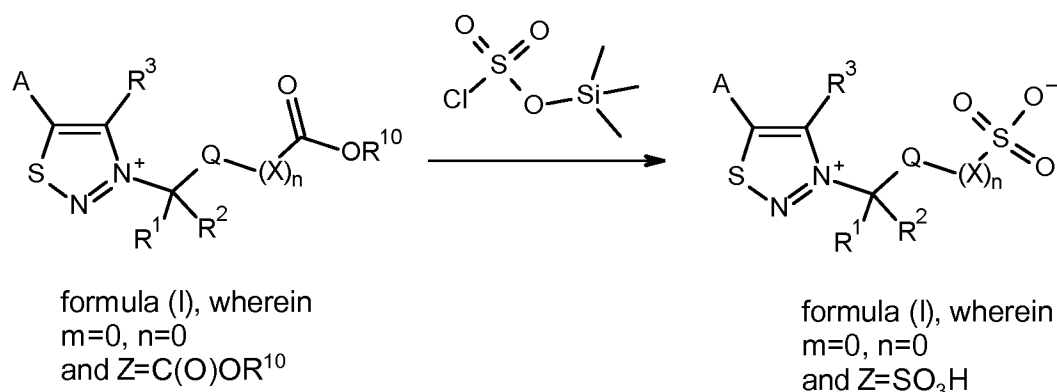
Reaction scheme 3



Suitable solvents and suitable temperatures are as previously described. An alkylating agent
 5 of formula (E) or (F) may include, but is not limited to, 1,3-propanesultone, 1,4-butanedisulfone, ethylenedisulfate, 1,3-propylene sulfide and 1,2,3-oxathiazolidine 2,2-dioxide. Such alkylating agents and related compounds are either known in the literature or may be prepared by known literature methods.

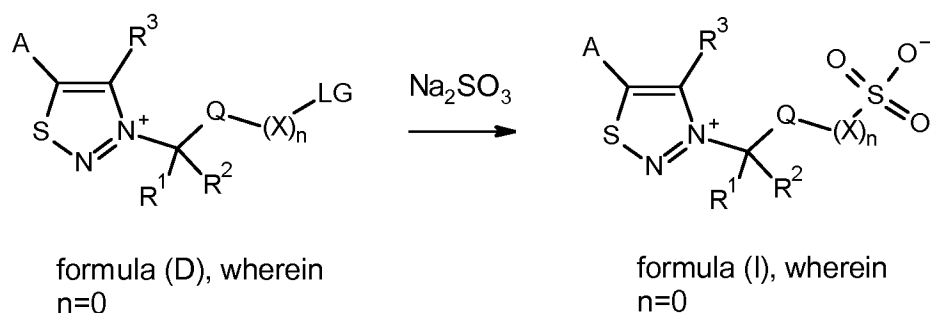
A compound of formula (I), wherein m is 0, n is 0 and Z is -S(O)₂OH, may be prepared from a
 compound of formula (I), wherein m is 0, n is 0 and Z is C(O)OR¹⁰, by treatment with trimethylsilylchloro
 10 sulfonate in a suitable solvent at a suitable temperature, as described in reaction scheme 4. Preferred conditions include heating the carboxylate precursor in neat trimethylsilylchlorosulfonate at a temperature between 25°C and 150°C.

Reaction scheme 4

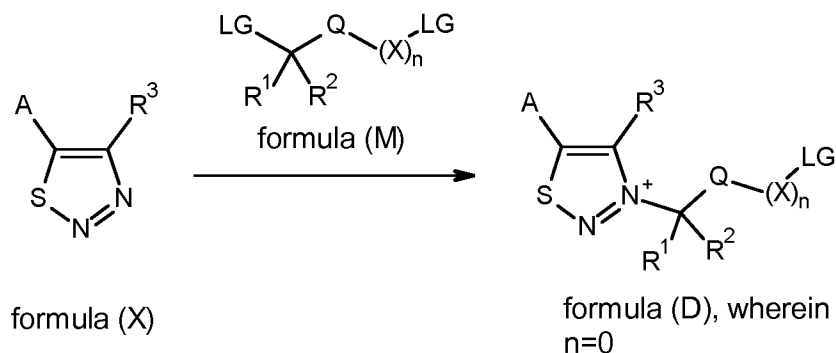


- 5 A compound of formula (I), wherein Z is $-S(O)_2OH$, may be further prepared from a compound of formula (D), wherein $n=0$ and R^1 , R^2 , R^3 , A, and Q are as defined for a compound of formula (I), and LG is a suitable leaving group, for example, but not limited to, halide or pseudohalide such as triflate, mesylate or tosylate, by treatment with sodium sulfite in a suitable solvent at a suitable temperature, as described in reaction scheme 5. Preferred conditions include heating the a compound of formula (D)
- 10 with sodium sulfite in water at a temperature between 30°C and 100°C.

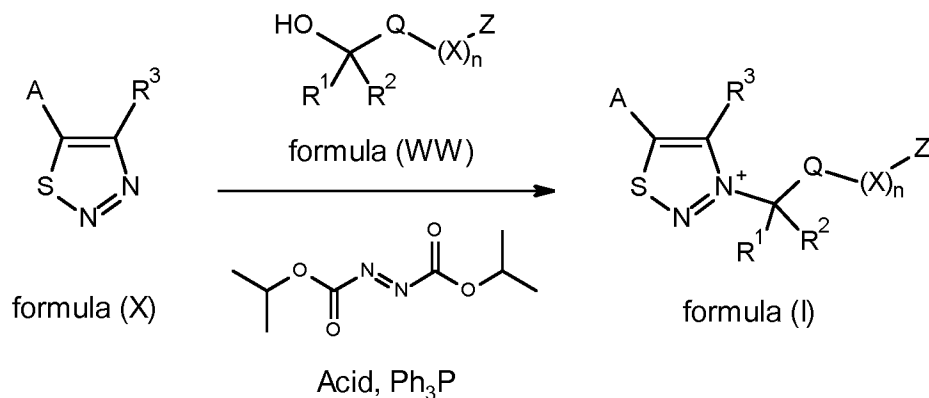
Reaction scheme 5



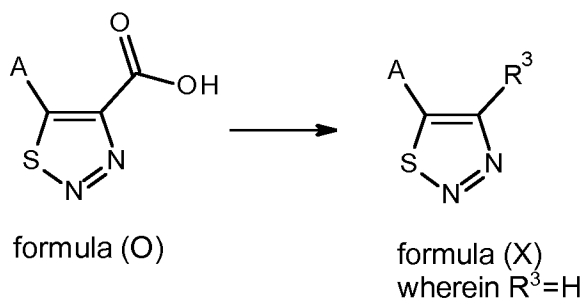
- A compound of formula (D) may be prepared by the alkylation of compounds of formula (X), wherein R^3 and A are as defined for compounds of formula (I), with a suitable alkylating agent of formula (M), wherein $n=0$ and R^1 , R^2 and Q are as defined for compounds of formula (I) and LG is a suitable leaving group, for example, but not limited to, halide or pseudohalide such as triflate, mesylate or tosylate, in a suitable solvent at a suitable temperature, as described in reaction scheme 6. An alkylating agent of formula (M) may include, but is not limited to, 2-chloroethyl trifluoromethanesulfonate.

Reaction scheme 6

- 5 Furthermore, compounds of formula (I) may be prepared by reacting compounds of formula (X), wherein R^3 and A are as defined for compounds of formula (I), with a suitable alcohol of formula (WW), wherein R^1 , R^2 , Q, X, n and Z are as defined for compounds of formula (I), under Mitsunobu-type conditions such as those reported by Petit et al, Tet. Lett. 2008, 49 (22), 3663. Suitable phosphines include triphenylphosphine, suitable azodicarboxylates include diisopropylazodicarboxylate and suitable
- 10 acids include fluoroboric acid, triflic acid and bis(trifluoromethylsulfonyl)amine, as described in reaction scheme 7. Such alcohols are either known in the literature or may be prepared by known literature methods.

Reaction scheme 7

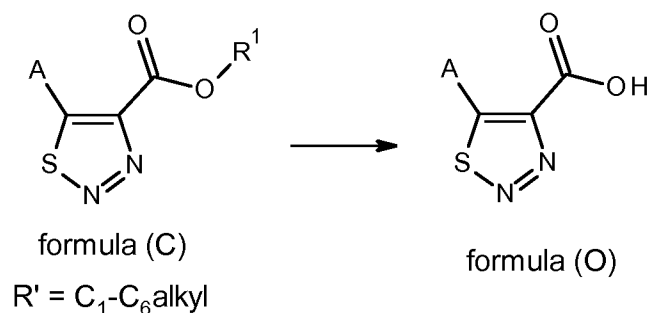
- 15 Compounds of formula (X), wherein A is as previously defined and R^3 is H, may be prepared through the decarboxylation of compounds of formula (O), in an appropriate solvent, at an appropriate temperature, as outlined in reaction scheme 8.

Reaction scheme 8

Examples of such a reaction are known in the literature, for example, Demaree et al., J. Heterocyclic Chem., 1978, 15, 1295.

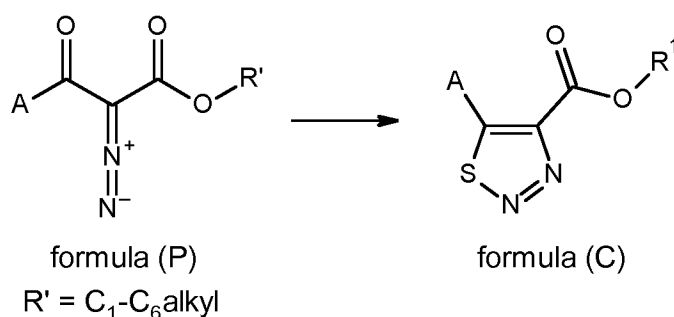
Compounds of formula (O), wherein A is as previously defined, may be prepared through the hydrolysis of compounds of formula (C), in the presence of a suitable base or acid, in an appropriate solvent, at an appropriate temperature, as outlined in reaction scheme 9. Related reactions are known in the literature.

Reaction scheme 9



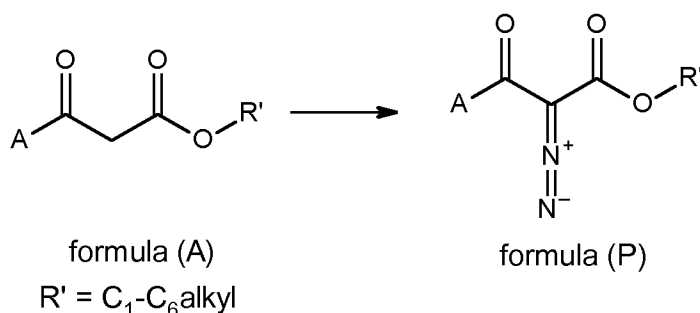
Compounds of formula (C), wherein A is as previously defined, may be prepared by the cyclisation of compounds of formula (P) using an appropriate reagent or a combination of reagents, example reagents include Lawesson's reagent or phosphorus pentasulfide, alternatively ammonium sulfite followed by bromine, in an appropriate solvent, at an appropriate temperature, as outlined in reaction scheme 10. Examples of such a reaction are known in the literature, for example, Gevorgyan et al., Org. Lett., 2016, 18, 1804.

Reaction scheme 10



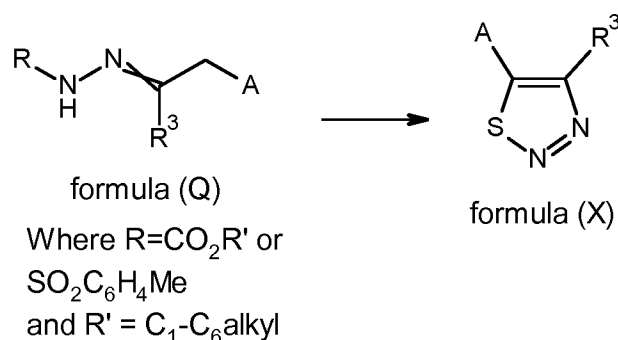
Compounds of formula (P), wherein A is as previously defined, may be prepared from a compound of formula (A) by reaction with an appropriate diazo transfer reagent, for example, but not limited to, 4-acetamidobenzenesulfonyl azide, in the presence of a suitable base, in an appropriate solvent, at an appropriate temperature, as outlined in reaction scheme 11. Examples of such a reaction are known in the literature, for example, Moody et al., Tetrahedron, 2004, 60, 3967. Compounds of formula (A), wherein A is as previously defined are either known in the literature or can be prepared by known methods.

Reaction scheme 11



Compounds of formula (X), wherein A and R³ are as previously defined, may also be prepared
 5 by a cyclisation of compounds of formula (Q) using an appropriate reagent, for example, but not limited to, thionyl chloride, in an appropriate solvent, at an appropriate temperature, as outlined in reaction scheme 12. Examples of such a reaction are known in the literature, for example, Gevorgyan et al., Org. Lett., 2016, 18, 1804 and Thomas et al., J. Med. Chem. 1985, 28, 442.

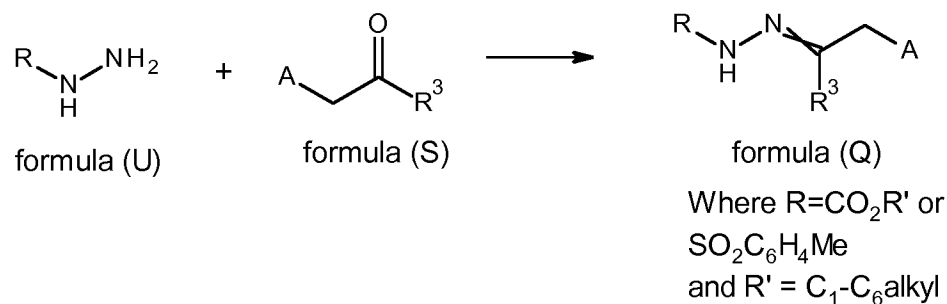
10 Reaction scheme 12



Compounds of formula (Q), wherein A and R³ are as previously defined, may be prepared from compounds of formula (S) and a hydrazide of formula (U), optionally in the presence of a suitable acid, for example acetic acid, in an appropriate solvent, at an appropriate temperature, as outlined in
 15 reaction scheme 13. Examples of such reactions are known in the literature, for example, Semakin et al., Beilstein J. Org. Chem., 2016, 12, 2471. Hydrazide compounds of formula (U) are either known in the literature or may be prepared by known methods. Compounds of formula (S), wherein A and R³ are as previously defined, are either known in the literature or may be prepared by known literature procedures, for example, Wolfe et al., J. Org. Chem., 1981, 46, 294.

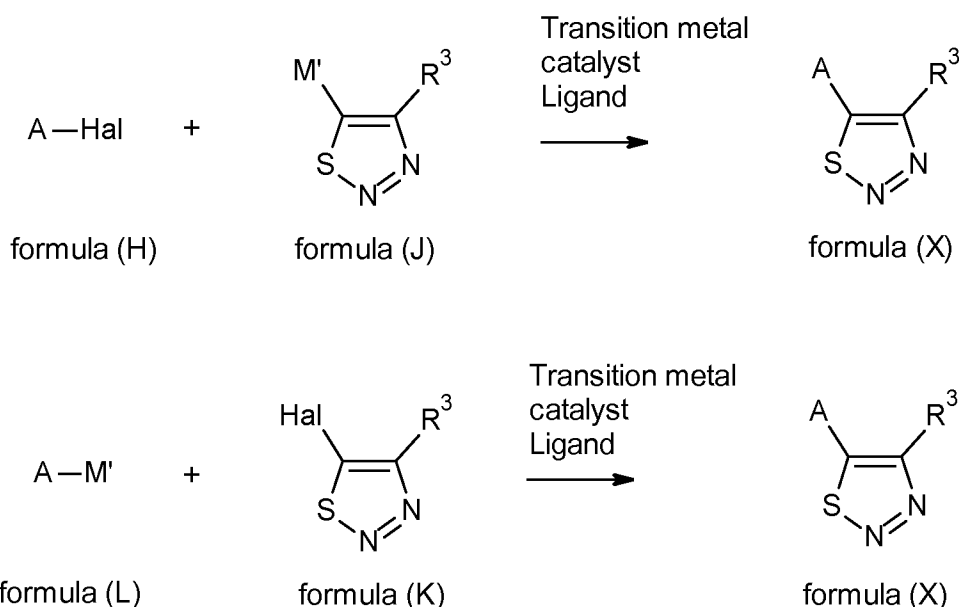
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Reaction scheme 13



In a further approach, compounds of formula (X) may be prepared by the transition metal cross-coupling of compounds of formula (H) and formula (J), or alternatively compounds of formula (K) and formula (L), in which compounds of formula (J) and formula (L) are either an organostannane, organoboronic acid or ester, organotrifluoroborate, organomagnesium, organocopper or organozinc (M'), as outlined in reaction scheme 14. Hal is defined as a halogen or pseudo halogen, for example triflate, mesylate or tosylate. Such cross-couplings include Stille (for example Stille, J. K. *Angew. Chem. Int. Ed. Engl.* 1986, 25, 508), Suzuki-Miyaura (for example Lindsley et al, WO2013063549A1), Negishi (for example King, A. O.; Okukado, N.; Negishi, E. *Chem. Comm.* 1977, 19, 683), and Kumada (for example Tamao, K.; Sumitani, K.; Kumada, M. *J. Am. Chem. Soc.* 1972, 94, 4374). The coupling partners may be selected with reference to the specific cross-coupling reaction and target product. Transition metal catalysts, ligands, bases, solvents and temperatures may be selected with reference to the desired cross-coupling and are known in the literature. Compounds of formula (H), formula (K) and formula (L) are known in the literature, or may be prepared by known literature methods.

15 Reaction scheme 14

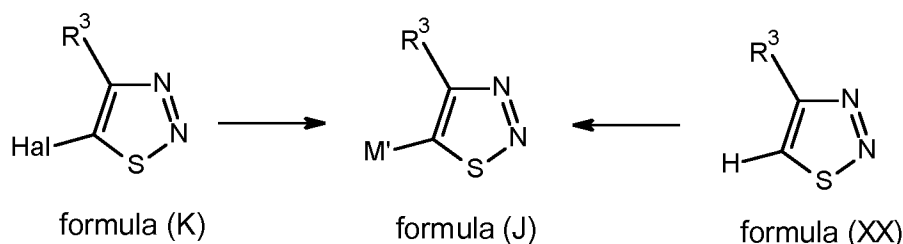


An organometallic of formula (J), which is either an organostannane, organoboronic acid or ester, organotrifluoroborate, organomagnesium, organolithium, organocopper or organozinc (M'), may be prepared from a compound of formula (XX), wherein R^3 is defined for compounds of formula (I), by metallation, as outlined in reaction scheme 16. Example conditions to prepare an organolithium of formula (J) include treatment of a compound of formula (XX) with methyl lithium in an appropriate solvent at an appropriate temperature, see, for example, Thomas, E. W.; Zimmerman, D. C. *Synthesis*, 1985, 10, 945). Alternatively, an organometallic of formula (J) may be prepared from a compound of formula (K), wherein R^3 is as defined for a compound of formula (I) and Hal is defined as a halogen or pseudo halogen, for example triflate, mesylate or tosylate, as described in scheme 16. Example conditions to prepare an organostannane of formula (J) include treatment of a compound of formula (K) with lithium tributyl tin in an appropriate solvent at an appropriate temperature, see, for example, WO 2010038465. Example conditions to prepare an organoboronic acid or ester of formula (J) include treatment of a

compound of formula (K) with bis(pinacolato)diboron, in the presence of an appropriate transition metal catalyst, appropriate ligand, appropriate base, in an appropriate solvent at an appropriate temperature, see, for example, KR 2015135626). Compounds of formula (K) and formula (XX) are either known in the literature or can be prepared by known methods.

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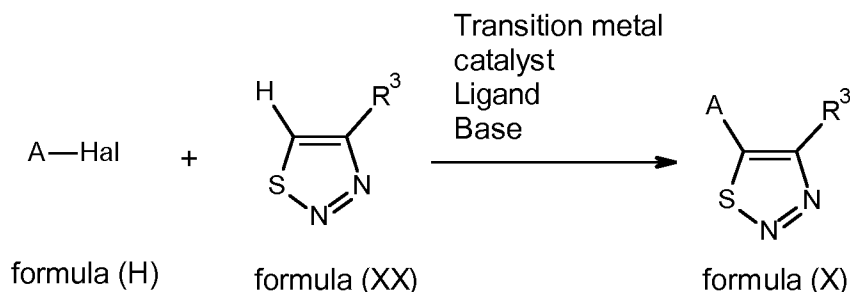
Reaction scheme 15



In a further approach, compounds of formula (X), wherein A and R³ are as previously defined, may be prepared by C-H activation cross-coupling of compounds of formula (XX) and formula (H), in the presence of an appropriate transition metal catalyst, ligand and base, in a suitable solvent at a suitable temperature, as outlined in reaction scheme 15. Hal is defined as a halogen or pseudo halogen, for example triflate, mesylate or tosylate. Examples of such reactions are known in the literature, for example, Chen et al, US20100152203. Compounds of formula (H) and formula (XX) are known in the literature, or may be prepared by known literature methods.

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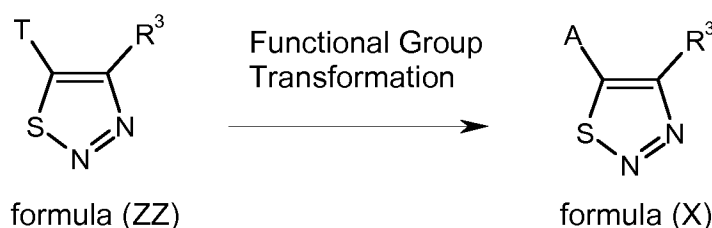
Reaction scheme 16



Finally, in an additional approach outlined in scheme 17, biaryl thiadiazoles of formula (X) may be prepared by classical ring synthesis approaches starting from a compound of formula (ZZ), wherein T is a functional group which can be converted through one or more chemical steps into a 5-membered heteroaryl A, wherein A is as defined for compounds of formula (I). Such functional groups include, but are not limited to, acid, ester, nitrile, amide, thioamide and ketone. Related transformations are known in the literature. Substituted thiadiazoles may be prepared using methodology outlined in the literature, for example, but are not limited to, Minkkiila, A. *et al*, Eur. J. Med. Chem. 2009, 44, 2994, Aoyama, Iwamoto and Shioiri Heterocycles, 1986, 24, 589 and Sakai, Hida and Kondo Bull. Chem. Soc. Jpn. 1986, 59, 179.

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Reaction scheme 17



5 The compounds according to the invention can be used as herbicidal agents in unmodified form, but they are generally formulated into compositions in various ways using formulation adjuvants, such as carriers, solvents and surface-active substances. The formulations can be in various physical forms, e.g. in the form of dusting powders, gels, wettable powders, water-dispersible granules, water-dispersible tablets, effervescent pellets, emulsifiable concentrates, microemulsifiable concentrates, oil-
10 in-water emulsions, oil-flowables, aqueous dispersions, oily dispersions, suspo-emulsions, capsule suspensions, emulsifiable granules, soluble liquids, water-soluble concentrates (with water or a water-miscible organic solvent as carrier), impregnated polymer films or in other forms known e.g. from the Manual on Development and Use of FAO and WHO Specifications for Pesticides, United Nations, First Edition, Second Revision (2010). Such formulations can either be used directly or diluted prior to use.
15 The dilutions can be made, for example, with water, liquid fertilisers, micronutrients, biological organisms, oil or solvents.

The formulations can be prepared e.g. by mixing the active ingredient with the formulation adjuvants in order to obtain compositions in the form of finely divided solids, granules, solutions, dispersions or emulsions. The active ingredients can also be formulated with other adjuvants, such as
20 finely divided solids, mineral oils, oils of vegetable or animal origin, modified oils of vegetable or animal origin, organic solvents, water, surface-active substances or combinations thereof.

The active ingredients can also be contained in very fine microcapsules. Microcapsules contain the active ingredients in a porous carrier. This enables the active ingredients to be released into the environment in controlled amounts (e.g. slow-release). Microcapsules usually have a diameter of from
25 0.1 to 500 microns. They contain active ingredients in an amount of about from 25 to 95 % by weight of the capsule weight. The active ingredients can be in the form of a monolithic solid, in the form of fine particles in solid or liquid dispersion or in the form of a suitable solution. The encapsulating membranes can comprise, for example, natural or synthetic rubbers, cellulose, styrene/butadiene copolymers, polyacrylonitrile, polyacrylate, polyesters, polyamides, polyureas, polyurethane or chemically modified
30 polymers and starch xanthates or other polymers that are known to the person skilled in the art. Alternatively, very fine microcapsules can be formed in which the active ingredient is contained in the form of finely divided particles in a solid matrix of base substance, but the microcapsules are not themselves encapsulated.

The formulation adjuvants that are suitable for the preparation of the compositions according to
35 the invention are known *per se*. As liquid carriers there may be used: water, toluene, xylene, petroleum ether, vegetable oils, acetone, methyl ethyl ketone, cyclohexanone, acid anhydrides, acetonitrile, acetophenone, amyl acetate, 2-butanone, butylene carbonate, chlorobenzene, cyclohexane,

cyclohexanol, alkyl esters of acetic acid, diacetone alcohol, 1,2-dichloropropane, diethanolamine, p-diethylbenzene, diethylene glycol, diethylene glycol abietate, diethylene glycol butyl ether, diethylene glycol ethyl ether, diethylene glycol methyl ether, *N,N*-dimethylformamide, dimethyl sulfoxide, 1,4-dioxane, dipropylene glycol, dipropylene glycol methyl ether, dipropylene glycol dibenzoate, diproxitol, 5 alkylpyrrolidone, ethyl acetate, 2-ethylhexanol, ethylene carbonate, 1,1,1-trichloroethane, 2-heptanone, alpha-pinene, d-limonene, ethyl lactate, ethylene glycol, ethylene glycol butyl ether, ethylene glycol methyl ether, gamma-butyrolactone, glycerol, glycerol acetate, glycerol diacetate, glycerol triacetate, hexadecane, hexylene glycol, isoamyl acetate, isobornyl acetate, isooctane, isophorone, isopropylbenzene, isopropyl myristate, lactic acid, laurylamine, mesityl oxide, methoxypropanol, methyl 10 isoamyl ketone, methyl isobutyl ketone, methyl laurate, methyl octanoate, methyl oleate, methylene chloride, m-xylene, *n*-hexane, *n*-octylamine, octadecanoic acid, octylamine acetate, oleic acid, oleylamine, o-xylene, phenol, polyethylene glycol, propionic acid, propyl lactate, propylene carbonate, propylene glycol, propylene glycol methyl ether, p-xylene, toluene, triethyl phosphate, triethylene glycol, xylenesulfonic acid, paraffin, mineral oil, trichloroethylene, perchloroethylene, ethyl acetate, amyl 15 acetate, butyl acetate, propylene glycol methyl ether, diethylene glycol methyl ether, methanol, ethanol, isopropanol, and alcohols of higher molecular weight, such as amyl alcohol, tetrahydrofurfuryl alcohol, hexanol, octanol, ethylene glycol, propylene glycol, glycerol, *N*-methyl-2-pyrrolidone and the like.

Suitable solid carriers are, for example, talc, titanium dioxide, pyrophyllite clay, silica, attapulgite clay, kieselguhr, limestone, calcium carbonate, bentonite, calcium montmorillonite, cottonseed husks, 20 wheat flour, soybean flour, pumice, wood flour, ground walnut shells, lignin and similar substances.

A large number of surface-active substances can advantageously be used in both solid and liquid formulations, especially in those formulations which can be diluted with a carrier prior to use. Surface-active substances may be anionic, cationic, non-ionic or polymeric and they can be used as emulsifiers, wetting agents or suspending agents or for other purposes. Typical surface-active 25 substances include, for example, salts of alkyl sulfates, such as diethanolammonium lauryl sulfate; salts of alkylarylsulfonates, such as calcium dodecylbenzenesulfonate; alkylphenol/alkylene oxide addition products, such as nonylphenol ethoxylate; alcohol/alkylene oxide addition products, such as tridecylalcohol ethoxylate; soaps, such as sodium stearate; salts of alkylnaphthalenesulfonates, such as sodium dibutylnaphthalenesulfonate; dialkyl esters of sulfosuccinate salts, such as sodium di(2- 30 ethylhexyl)sulfosuccinate; sorbitol esters, such as sorbitol oleate; quaternary amines, such as lauryltrimethylammonium chloride, polyethylene glycol esters of fatty acids, such as polyethylene glycol stearate; block copolymers of ethylene oxide and propylene oxide; and salts of mono- and di-alkylphosphate esters; and also further substances described e.g. in McCutcheon's Detergents and Emulsifiers Annual, MC Publishing Corp., Ridgewood New Jersey (1981).

35 Further adjuvants that can be used in pesticidal formulations include crystallisation inhibitors, viscosity modifiers, suspending agents, dyes, anti-oxidants, foaming agents, light absorbers, mixing auxiliaries, antifoams, complexing agents, neutralising or pH-modifying substances and buffers, corrosion inhibitors, fragrances, wetting agents, take-up enhancers, micronutrients, plasticisers, glidants, lubricants, dispersants, thickeners, antifreezes, microbicides, and liquid and solid fertilisers.

40 The compositions according to the invention can include an additive comprising an oil of vegetable or animal origin, a mineral oil, alkyl esters of such oils or mixtures of such oils and oil

derivatives. The amount of oil additive in the composition according to the invention is generally from 0.01 to 10 %, based on the mixture to be applied. For example, the oil additive can be added to a spray tank in the desired concentration after a spray mixture has been prepared. Preferred oil additives comprise mineral oils or an oil of vegetable origin, for example rapeseed oil, olive oil or sunflower oil, emulsified vegetable oil, alkyl esters of oils of vegetable origin, for example the methyl derivatives, or an oil of animal origin, such as fish oil or beef tallow. Preferred oil additives comprise alkyl esters of C₈-C₂₂ fatty acids, especially the methyl derivatives of C₁₂-C₁₈ fatty acids, for example the methyl esters of lauric acid, palmitic acid and oleic acid (methyl laurate, methyl palmitate and methyl oleate, respectively). Many oil derivatives are known from the Compendium of Herbicide Adjuvants, 10th Edition, Southern Illinois University, 2010.

The herbicidal compositions generally comprise from 0.1 to 99 % by weight, especially from 0.1 to 95 % by weight, compounds of formula (I) and from 1 to 99.9 % by weight of a formulation adjuvant which preferably includes from 0 to 25 % by weight of a surface-active substance. The inventive compositions generally comprise from 0.1 to 99 % by weight, especially from 0.1 to 95 % by weight, of compounds of the present invention and from 1 to 99.9 % by weight of a formulation adjuvant which preferably includes from 0 to 25 % by weight of a surface-active substance. Whereas commercial products may preferably be formulated as concentrates, the end user will normally employ dilute formulations.

The rates of application vary within wide limits and depend on the nature of the soil, the method of application, the crop plant, the pest to be controlled, the prevailing climatic conditions, and other factors governed by the method of application, the time of application and the target crop. As a general guideline compounds may be applied at a rate of from 1 to 2000 l/ha, especially from 10 to 1000 l/ha.

Preferred formulations can have the following compositions (weight %):

Emulsifiable concentrates:

active ingredient:	1 to 95 %, preferably 60 to 90 %
surface-active agent:	1 to 30 %, preferably 5 to 20 %
liquid carrier:	1 to 80 %, preferably 1 to 35 %

Dusts:

active ingredient:	0.1 to 10 %, preferably 0.1 to 5 %
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 %
surface-active agent:	1 to 40 %, preferably 2 to 30 %

Wettable powders:

active ingredient:	0.5 to 90 %, preferably 1 to 80 %
surface-active agent:	0.5 to 20 %, preferably 1 to 15 %
5 solid carrier:	5 to 95 %, preferably 15 to 90 %

Granules:

active ingredient:	0.1 to 30 %, preferably 0.1 to 15 %
solid carrier:	99.5 to 70 %, preferably 97 to 85 %

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The composition of the present may further comprise at least one additional pesticide. For example, the compounds according to the invention can also be used in combination with other herbicides or plant growth regulators. In a preferred embodiment the additional pesticide is a herbicide and/or herbicide safener.

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Thus, compounds of formula (I) can be used in combination with one or more other herbicides to provide various herbicidal mixtures. Specific examples of such mixtures include (wherein "I" represents a compound of formula (I)):- I + acetochlor; I + acifluorfen (including acifluorfen-sodium); I + aclonifen; I + alachlor; I + alloxymid; I + ametryn; I + amicarbazone; I + amidosulfuron; I + aminocyclopyrachlor; I + aminopyralid; I + amitrole; I + asulam; I + atrazine; I + bensulfuron (including

20 bensulfuron-methyl); I + bentazone; I + bicyclopyrone; I + bilanafos; I + bifenox; I + bispyribac-sodium; I + bixlozone; I + bromacil; I + bromoxynil; I + butachlor; I + butafenacil; I + cafenstrole; I + carfentrazone (including carfentrazone-ethyl); I + cloransulam (including cloransulam-methyl); I + chlorimuron (including chlorimuron-ethyl); I + chlorotoluron; I + cinosulfuron; I + chlorsulfuron; I + cinmethylin; I + clacyfos; I + clethodim; I + clodinafop (including clodinafop-propargyl); I + clomazone; I + clopyralid; I + cyclopyranil;

25 I + cyclopyrimorate; I + cyclosulfamuron; I + cyhalofop (including cyhalofop-butyl); I + 2,4-D (including the choline salt and 2-ethylhexyl ester thereof); I + 2,4-DB; I + daimuron; I + desmedipham; I + dicamba (including the aluminum, aminopropyl, bis-aminopropylmethyl, choline, dichloroprop, diglycolamine, dimethylamine, dimethylammonium, potassium and sodium salts thereof); I + diclofop-methyl; I + diclosulam; I + diflufenican; I + difenzoquat; I + diflufenican; I + diflufenzopyr; I + dimethachlor; I +

30 dimethenamid-P; I + diquat dibromide; I + diuron; I + esprocarb; I + ethalfluralin; I + ethofumesate; I + fenoxaprop (including fenoxaprop-P-ethyl); I + fenoxasulfone; I + fenquinotrine; I + fentrazamide; I + flazasulfuron; I + florasulam; I + florpyrauxifen; I + fluazifop (including fluazifop-P-butyl); I + flucarbazone (including flucarbazone-sodium); I + flufenacet; I + flumetralin; I + flumetsulam; I + flumioxazin; I + flupyrsulfuron (including flupyrsulfuron-methyl-sodium); I + fluroxypyr (including fluroxypyr-meptyl); I +

35 fluthiacet-methyl; I + fomesafen; I + foramsulfuron; I + glufosinate (including the ammonium salt thereof); I + glyphosate (including the diammonium, isopropylammonium and potassium salts thereof); I + halauxifen (including halauxifen-methyl); I + halosulfuron-methyl; I + haloxyfop (including haloxyfop-methyl); I + hexazinone; I + hydantocidin; I + imazamox; I + imazapic; I + imazapyr; I + imazaquin; I + imazethapyr; I + indaziflam; I + iodosulfuron (including iodosulfuron-methyl-sodium); I + iofensulfuron; I +

40 + iofensulfuron-sodium; I + ioxynil; I + ipfencarbazone; I + isoproturon; I + isoxaben; I + isoxaflutole; I + lactofen; I + lancotrione; I + linuron; I + MCPA; I + MCPB; I + mecoprop-P; I + mefenacet; I +

mesosulfuron; I + mesosulfuron-methyl; I + mesotrione; I + metamitron; I + metazachlor; I + methiozolin; I + metobromuron; I + metolachlor; I + metosulam; I + metoxuron; I + metribuzin; I + metsulfuron; I + molinate; I + napropamide; I + nicosulfuron; I + norflurazon; I + orthosulfamuron; I + oxadiargyl; I + oxadiazon; I + oxasulfuron; I + oxyfluorfen; I + paraquat dichloride; I + pendimethalin; I + penoxsulam; I + phenmedipham; I + picloram; I + picolinafen; I + pinoxaden; I + pretilachlor; I + primisulfuron-methyl; I + prodiamine; I + prometryn; I + propachlor; I + propanil; I + propaquizafop; I + propham; I + propyrisulfuron; I + propyzamide; I + prosulfocarb; I + prosulfuron; I + pyraclonil; I + pyraflufen (including pyraflufen-ethyl); I + pyrasulfotole; I + pyrazolynate; I + pyrazosulfuron-ethyl; I + pyribenzoxim; I + pyridate; I + pyriftalid; I + pyrimisulfan; I + pyriothiobac-sodium; I + pyroxasulfone; I + pyroxsulam; I + quinclorac; I + quinmerac; I + quizalofop (including quizalofop-P-ethyl and quizalofop-P-tefuryl); I + rimsulfuron; I + saflufenacil; I + sethoxydim; I + simazine; I + S-metolachlor; I + sulcotrione; I + sulfentrazone; I + sulfosulfuron; I + tebuthiuron; I + tefuryltrione; I + tembotrione; I + terbuthylazine; I + terbutryn; I + thiencarbazone; I + thifensulfuron; I + tiafenacil; I + tolypyralate; I + topramezone; I + tralkoxydim; I + triafamone; I + triallate; I + triasulfuron; I + tribenuron (including tribenuron-methyl); I + triclopyr; I + trifloxysulfuron (including trifloxysulfuron-sodium); I + trifludimoxazin; I + trifluralin; I + triflusulfuron; I + tritosulfuron; I + 4-hydroxy-1-methoxy-5-methyl-3-[4-(trifluoromethyl)-2-pyridyl]imidazolidin-2-one; I + 4-hydroxy-1,5-dimethyl-3-[4-(trifluoromethyl)-2-pyridyl]imidazolidin-2-one; I + 5-ethoxy-4-hydroxy-1-methyl-3-[4-(trifluoromethyl)-2-pyridyl]imidazolidin-2-one; I + 4-hydroxy-1-methyl-3-[4-(trifluoromethyl)-2-pyridyl]imidazolidin-2-one; I + 4-hydroxy-1,5-dimethyl-3-[1-methyl-5-(trifluoromethyl)pyrazol-3-yl]imidazolidin-2-one; I + (4R)-1-(5-tert-butylisoxazol-3-yl)-4-ethoxy-5-hydroxy-3-methyl-imidazolidin-2-one; I + 3-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]bicyclo[3.2.1]octane-2,4-dione; I + 2-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]-5-methyl-cyclohexane-1,3-dione; I + 2-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]cyclohexane-1,3-dione; I + 2-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]-5,5-dimethyl-cyclohexane-1,3-dione; I + 6-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]-2,2,4,4-tetramethyl-cyclohexane-1,3,5-trione; I + 2-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]-5-ethyl-cyclohexane-1,3-dione; I + 2-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]-4,4,6,6-tetramethyl-cyclohexane-1,3-dione; I + 2-[6-cyclopropyl-2-(3,4-dimethoxyphenyl)-3-oxo-pyridazine-4-carbonyl]-5-methyl-cyclohexane-1,3-dione; I + 3-[6-cyclopropyl-2-(3,4-dimethoxyphenyl)-3-oxo-pyridazine-4-carbonyl]bicyclo[3.2.1]octane-2,4-dione; I + 2-[6-cyclopropyl-2-(3,4-dimethoxyphenyl)-3-oxo-pyridazine-4-carbonyl]-5,5-dimethyl-cyclohexane-1,3-dione; I + 6-[6-cyclopropyl-2-(3,4-dimethoxyphenyl)-3-oxo-pyridazine-4-carbonyl]-2,2,4,4-tetramethyl-cyclohexane-1,3,5-trione; I + 2-[6-cyclopropyl-2-(3,4-dimethoxyphenyl)-3-oxo-pyridazine-4-carbonyl]cyclohexane-1,3-dione; I + 4-[2-(3,4-dimethoxyphenyl)-6-methyl-3-oxo-pyridazine-4-carbonyl]-2,2,6,6-tetramethyl-tetrahydropyran-3,5-dione and I + 4-[6-cyclopropyl-2-(3,4-dimethoxyphenyl)-3-oxo-pyridazine-4-carbonyl]-2,2,6,6-tetramethyl-tetrahydropyran-3,5-dione, I + 4-amino-3-chloro-5-fluoro-6-(7-fluoro-1H-indol-6-yl)pyridine-2-carboxylic acid as well as I + agrochemically acceptable esters thereof, for example, I + methyl 4-amino-3-chloro-5-fluoro-6-(7-fluoro-1H-indol-6-yl)pyridine-2-carboxylate).

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, Fourteenth Edition, British Crop Protection Council, 2006.

The compound of formula (I) can also be used in mixtures with other agrochemicals such as fungicides, nematicides or insecticides, examples of which are given in The Pesticide Manual.

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

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).

Compounds of formula (I) of the present invention may also be combined with herbicide
10 safeners. Preferred combinations (wherein "I" represents a compound of formula (I)) include:- I + benoxacor, I + cloquintocet (including cloquintocet-mexyl); I + cyprosulfamide; I + dichlormid; I + fenchlorazole (including fenchlorazole-ethyl); I + fenclorim; I + fluxofenim; I + furilazole I + isoxadifen (including isoxadifen-ethyl); I + mefenpyr (including mefenpyr-diethyl); I + metcamifen; I + N-(2-methoxybenzoyl)-4-[(methylaminocarbonyl)amino] benzenesulfonamide and I + oxabetrinil.

15 Particularly preferred are mixtures of a compound of formula (I) with cyprosulfamide, isoxadifen (including isoxadifen-ethyl), cloquintocet (including cloquintocet-mexyl) and/or N-(2-methoxybenzoyl)-4-[(methyl-aminocarbonyl)amino]benzenesulfonamide.

The safeners of the compound of formula (I) may also be in the form of esters or salts, as mentioned e.g. in The Pesticide Manual, 14th Edition (BCPC), 2006. The reference to cloquintocet-mexyl
20 also applies to a lithium, sodium, potassium, calcium, magnesium, aluminium, iron, ammonium, quaternary ammonium, sulfonium or phosphonium salt thereof as disclosed in WO 02/34048, and the reference to fenchlorazole-ethyl also applies to fenchlorazole, etc.

Preferably the mixing ratio of compound of formula (I) to safener is from 100:1 to 1:10, especially from 20:1 to 1:1.

25 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 safener).

The compounds of formula (I) of this invention are useful as herbicides. The present invention therefore further comprises a method for controlling unwanted plants comprising applying to the said plants or a locus comprising them, an effective amount of a compound of the invention or a herbicidal
30 composition containing said compound. 'Controlling' means killing, reducing or retarding growth or preventing or reducing germination. Generally the plants to be controlled are unwanted plants (weeds). 'Locus' means the area in which the plants are growing or will grow.

The rates of application of compounds of formula (I) may vary within wide limits and depend on the nature of the soil, the method of application (pre-emergence; post-emergence; application to the
35 seed furrow; no tillage application etc.), the crop plant, the weed(s) to be controlled, the prevailing climatic conditions, and other factors governed by the method of application, the time of application and the target crop. The compounds of formula (I) according to the invention are generally applied at a rate of from 10 to 2000 g/ha, especially from 50 to 1000 g/ha.

The application is generally made by spraying the composition, typically by tractor mounted
40 sprayer for large areas, but other methods such as dusting (for powders), drip or drench can also be used.

Useful plants in which the composition according to the invention can be used include crops such as cereals, for example barley and wheat, cotton, oilseed rape, sunflower, maize, rice, soybeans, sugar beet, sugar cane and turf.

Crop plants can also include trees, such as fruit trees, palm trees, coconut trees or other nuts.

5 Also included are vines such as grapes, fruit bushes, fruit plants and vegetables.

Crops are to be understood as also including those crops which have been rendered tolerant to herbicides or classes of herbicides (e.g. ALS-, GS-, EPSPS-, PPO-, ACCase- and HPPD-inhibitors) by conventional methods of breeding or by genetic engineering. An example of a crop that has been rendered tolerant to imidazolinones, e.g. imazamox, by conventional methods of breeding is Clearfield®
10 summer rape (canola). Examples of crops that have been rendered tolerant to herbicides by genetic engineering methods include e.g. glyphosate- and glufosinate-resistant maize varieties commercially available under the trade names RoundupReady® and LibertyLink®.

Crops are also to be understood as being those which have been rendered resistant to harmful insects by genetic engineering methods, for example Bt maize (resistant to European corn borer), Bt
15 cotton (resistant to cotton boll weevil) and also Bt potatoes (resistant to Colorado beetle). Examples of Bt maize are the Bt 176 maize hybrids of NK® (Syngenta Seeds). The Bt toxin is a protein that is formed naturally by *Bacillus thuringiensis* soil bacteria. Examples of toxins, or transgenic plants able to synthesise such toxins, are described in EP-A-451 878, EP-A-374 753, WO 93/07278, WO 95/34656, WO 03/052073 and EP-A-427 529. Examples of transgenic plants comprising one or more genes that
20 code for an insecticidal resistance and express one or more toxins are KnockOut® (maize), Yield Gard® (maize), NuCOTIN33B® (cotton), Bollgard® (cotton), NewLeaf® (potatoes), NatureGard® and Protexcta®. Plant crops or seed material thereof can be both resistant to herbicides and, at the same time, resistant to insect feeding ("stacked" transgenic events). For example, seed can have the ability to express an insecticidal Cry3 protein while at the same time being tolerant to glyphosate.

25 Crops are also to be understood to include those which are obtained by conventional methods of breeding or genetic engineering and contain so-called output traits (e.g. improved storage stability, higher nutritional value and improved flavour).

Other useful plants include turf grass for example in golf-courses, lawns, parks and roadsides, or grown commercially for sod, and ornamental plants such as flowers or bushes.

30 Compounds of formula (I) and compositions of the invention can typically be used to control a wide variety of monocotyledonous and dicotyledonous weed species. Examples of monocotyledonous species that can typically be controlled include *Alopecurus myosuroides*, *Avena fatua*, *Brachiaria plantaginea*, *Bromus tectorum*, *Cyperus esculentus*, *Digitaria sanguinalis*, *Echinochloa crus-galli*, *Lolium perenne*, *Lolium multiflorum*, *Panicum miliaceum*, *Poa annua*, *Setaria viridis*, *Setaria faberi* and
35 *Sorghum bicolor*. Examples of dicotyledonous species that can be controlled include *Abutilon theophrasti*, *Amaranthus retroflexus*, *Bidens pilosa*, *Chenopodium album*, *Euphorbia heterophylla*, *Galium aparine*, *Ipomoea hederacea*, *Kochia scoparia*, *Polygonum convolvulus*, *Sida spinosa*, *Sinapis arvensis*, *Solanum nigrum*, *Stellaria media*, *Veronica persica* and *Xanthium strumarium*.

Compounds/compositions of the invention are particularly useful in non-selective burn-down
40 applications, and as such may also be used to control volunteer or escape crop plants.

Furthermore, the compounds of formula (I) are also useful for pre-harvest desiccation in crops, for example, but not limited to, potatoes, soybean, sunflowers and cotton. Pre-harvest desiccation is used to desiccate crop foliage without significant damage to the crop itself to aid harvesting.

Various aspects and embodiments of the present invention will now be illustrated in more detail by way of example. It will be appreciated that modification of detail may be made without departing from the scope of the invention.

EXAMPLES

The Examples which follow serve to illustrate, but do not limit, the invention.

Formulation Examples

Wettable powders	a)	b)	c)
active ingredients	25 %	50 %	75 %
sodium lignosulfonate	5 %	5 %	-
sodium lauryl sulfate	3 %	-	5 %
sodium diisobutyl naphthalenesulfonate	-	6 %	10 %
phenol polyethylene glycol ether (7-8 mol of ethylene oxide)	-	2 %	-
highly dispersed silicic acid	5 %	10 %	10 %
Kaolin	62 %	27 %	-

The combination is thoroughly mixed with the adjuvants and the mixture is thoroughly ground in a suitable mill, affording wettable powders that can be diluted with water to give suspensions of the desired concentration.

Emulsifiable concentrate

active ingredients	10 %
octylphenol polyethylene glycol ether (4-5 mol of ethylene oxide)	3 %
calcium dodecylbenzenesulfonate	3 %
castor oil polyglycol ether (35 mol of ethylene oxide)	4 %
Cyclohexanone	30 %
xylene mixture	50 %

Emulsions of any required dilution, which can be used in plant protection, can be obtained from this concentrate by dilution with water.

Dusts	a)	b)	c)
Active ingredients	5 %	6 %	4 %
Talcum	95 %	-	-
Kaolin	-	94 %	-
mineral filler	-	-	96 %

Ready-for-use dusts are obtained by mixing the combination with the carrier and grinding the mixture in a suitable mill.

5

Extruder granules

Active ingredients	15 %
sodium lignosulfonate	2 %
carboxymethylcellulose	1 %
Kaolin	82 %

The combination is mixed and ground with the adjuvants, and the mixture is moistened with water. The mixture is extruded and then dried in a stream of air.

Coated granules

Active ingredients	8 %
polyethylene glycol (mol. wt. 200)	3 %
Kaolin	89 %

10 The finely ground combination is uniformly applied, in a mixer, to the kaolin moistened with polyethylene glycol. Non-dusty coated granules are obtained in this manner.

Suspension concentrate

active ingredients	40 %
propylene glycol	10 %
nonylphenol polyethylene glycol ether (15 mol of ethylene oxide)	6 %
Sodium lignosulfonate	10 %
carboxymethylcellulose	1 %
silicone oil (in the form of a 75 % emulsion in water)	1 %
Water	32 %

15 The finely ground combination is intimately mixed with the adjuvants, giving a suspension concentrate from which suspensions of any desired dilution can be obtained by dilution with water.

Slow Release Capsule Suspension

28 parts of the combination are mixed with 2 parts of an aromatic solvent and 7 parts of toluene diisocyanate/polymethylene-polyphenylisocyanate-mixture (8:1). This mixture is emulsified in a mixture
20 of 1.2 parts of polyvinylalcohol, 0.05 parts of a defoamer and 51.6 parts of water until the desired particle

size is achieved. To this emulsion a mixture of 2.8 parts 1,6-diaminohexane in 5.3 parts of water is added. The mixture is agitated until the polymerization reaction is completed.

The obtained capsule suspension is stabilized by adding 0.25 parts of a thickener and 3 parts of a dispersing agent. The capsule suspension formulation contains 28% of the active ingredients. The medium capsule diameter is 8-15 microns.

The resulting formulation is applied to seeds as an aqueous suspension in an apparatus suitable for that purpose.

List of Abbreviations:

10

Boc = *tert*-butyloxycarbonyl

br = broad

CDCl₃ = chloroform-d

CD₃OD = methanol-d

15 °C

= degrees Celsius

D₂O = water-d

DCM = dichloromethane

d = doublet

dd = double doublet

20 dt

= double triplet

DMSO = dimethylsulfoxide

EtOAc = ethyl acetate

h = hour(s)

HCl = hydrochloric acid

25 HPLC

= high-performance liquid chromatography (description of the apparatus and the methods used for HPLC are given below)

m = multiplet

M = molar

min = minutes

30 MHz

= mega hertz

mL = millilitre

mp = melting point

ppm = parts per million

q = quartet

35 quin

= quintet

rt = room temperature

s = singlet

t = triplet

THF = tetrahydrofuran

40 LC/MS

= Liquid Chromatography Mass Spectrometry

Preparative Reverse Phase HPLC Method:

Compounds purified by mass directed preparative HPLC using ES+/ES- on a Waters FractionLynx Autopurification system comprising a 2767 injector/collector with a 2545 gradient pump, two 515 isocratic pumps, SFO, 2998 photodiode array (Wavelength range (nm): 210 to 400), 2424 ELSD and 5 QDa mass spectrometer. A Waters Atlantis T3 5micron 19x10mm guard column was used with a Waters Atlantis T3 OBD, 5micron 30x100mm prep column.

Ionisation method: Electrospray positive and negative: Cone (V) 20.00, Source Temperature (°C) 120, Cone Gas Flow (L/Hr.) 50

10 Mass range (Da): positive 100 to 800, negative 115 to 800.

The preparative HPLC was conducted using an 11.4 minute run time (not using at column dilution, bypassed with the column selector), according to the following gradient table:

Time (mins)	Solvent A (%)	Solvent B (%)	Flow (ml / min)
0.00	100	0	35
2.00	100	0	35
2.01	100	0	35
7.0	90	10	35
7.3	0	100	35
9.2	0	100	35
9.8	99	1	35
11.35	99	1	35
11.40	99	1	35

515 pump 0ml/min Acetonitrile (ACD)

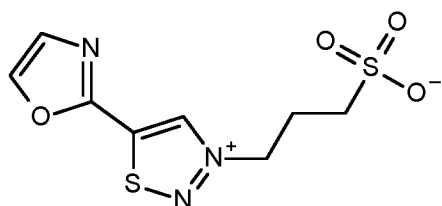
15 515 pump 1ml/min 90% Methanol/10% Water (make up pump)

Solvent A: Water with 0.05% Trifluoroacetic Acid

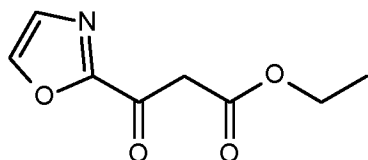
Solvent B: Acetonitrile with 0.05% Trifluoroacetic Acid

20 PREPARATION EXAMPLES

Example 1: Preparation of 3-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)propane-1-sulfonate: compound A1

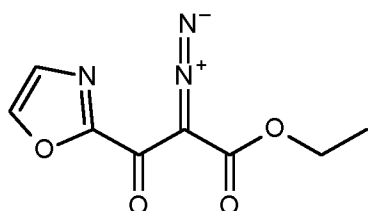


Step 1: Preparation of ethyl 3-oxazol-2-yl-3-oxo-propanoate



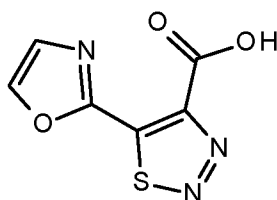
To a suspension of sodium hydride (1.88 g) in tetrahydrofuran (168 mL), under nitrogen atmosphere, was added ethyl oxazole-2-carboxylate (5 g) portionwise. The reaction mixture was heated at 75°C and ethyl acetate (4.47 g) was added dropwise and heating continued for 20 hours. The reaction mixture was quenched with saturated ammonium chloride and extracted with ethyl acetate. The organic layer was washed with brine, dried with magnesium sulfate, filtered and concentrated to give ethyl 3-oxazol-2-yl-3-oxo-propanoate as an orange oil. The product was used in the subsequent step without further purification.

10 Step 2: Preparation of ethyl 2-diazo-3-oxazol-2-yl-3-oxo-propanoate



To a solution of ethyl 3-oxazol-2-yl-3-oxo-propanoate (5.1 g) and 4-acetamidobenzenesulfonyl azide (7.59 g) in dichloromethane (167 mL) at 0°C was added triethylamine (8.54 g) dropwise. The reaction was slowly warmed to room temperature over 1 hour and stirred for an additional 1 hour. The reaction mixture was filtered through a plug of silica, washing through with further dichloromethane. The filtrate was concentrated to give ethyl 2-diazo-3-oxazol-2-yl-3-oxo-propanoate as a pale red solid. The product was used in the subsequent step without further purification.

Step 3: Preparation of 5-oxazol-2-ylthiadiazole-4-carboxylic acid

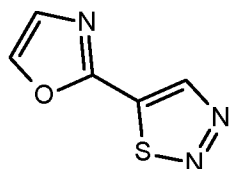


20

A solution of ethyl 2-diazo-3-oxazol-2-yl-3-oxo-propanoate (5.8 g) and Lawesson's reagent (13.88 g) in toluene (139 mL) was heated at 120°C for 2 hours. The cooled reaction mixture was filtered through a plug of silica, washing through with dichloromethane. The filtrate was concentrated to afford a crude brown gum. The crude gum was stirred in tetrahydrofuran (111 mL) and 2M aqueous sodium hydroxide (111 mL) at room temperature for 4 hours. The reaction mixture was partitioned with dichloromethane and the aqueous phase was carefully acidified to pH1 using concentrated hydrochloric acid. The resulting precipitate was filtered off and washed with diethyl ether to give 5-oxazol-2-ylthiadiazole-4-carboxylic acid as a beige solid.

¹H NMR (400MHz, DMSO-*d*₆) 8.50 (d, 1 H), 7.62 (d, 1 H) (one CO₂H proton missing).

Step 4: Preparation of 2-(thiadiazol-5-yl)oxazole



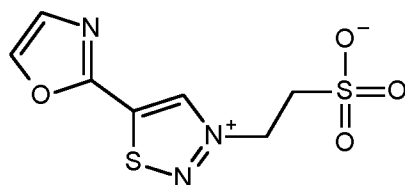
A mixture of 5-oxazol-2-ylthiadiazole-4-carboxylic acid (1.06 g) and 1,4-dioxane (54 mL) was heated at 100°C for 24 hours. The reaction mixture was concentrated to give 2-(thiadiazol-5-yl)oxazole as a beige solid.

¹H NMR (400MHz, CDCl₃) 9.18 (s, 1 H), 7.83 (d, 1 H), 7.37 (s, 1 H).

Step 5: Preparation of 3-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)propane-1-sulfonate (compound A1)

A solution of 2-(thiadiazol-5-yl)oxazole (0.08 g) and 1,3-propanesultone (0.097 g) was heated in 1,4-dioxane (3 mL) at 100°C for 5 days. The resulting precipitate was filtered off and washed with ethanol to give 3-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)propane-1-sulfonate as a beige solid.

¹H NMR (400 MHz, D₂O) 10.12 (s, 1 H), 8.15 (s, 1 H), 7.51 (s, 1 H), 5.17 (t, 2 H), 3.05 – 2.96 (m, 2 H), 2.64 – 2.51 (m, 2 H).

Example 2 Preparation of 2-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)ethanesulfonate: compound A2

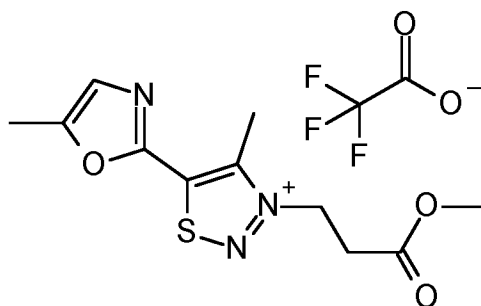
20

To a solution of 2-chloroethanol (0.08 g) and pyridine (0.094 g) in dichloromethane (7 mL) at -10°C was added trifluoromethanesulfonic anhydride (0.34 g) dropwise. The reaction was stirred at -10°C for 1 hour and a solution of 2-(thiadiazol-5-yl)oxazole (0.1 g) in dichloromethane (2 mL) was added. The reaction was allowed to warm to room temperature and stirred for 20 hours. The reaction mixture was concentrated and the residue was dissolved in water (3 mL). Sodium sulfite (0.125 g) was added in one portion and the mixture heated at 100°C for 2 hours. The cooled reaction mixture was extracted with dichloromethane and the aqueous phase was concentrated and purified by preparative reverse phase HPLC to give 2-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)ethanesulfonate as a colourless solid.

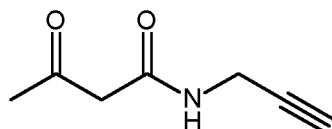
¹H NMR (400MHz, D₂O) 10.16 (s, 1 H), 8.13 (d, 1 H), 7.49 (d, 1 H), 5.41 – 5.33 (m, 2 H), 3.69 (dd, 2 H).

30

Example 3 Preparation of methyl 3-[4-methyl-5-(5-methyloxazol-2-yl)thiadiazol-3-ium-3-yl]propanoate 2,2,2-trifluoroacetate: compound A11

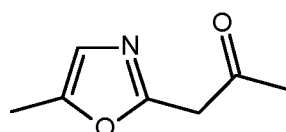


Step 1: Preparation of 3-oxo-*N*-prop-2-ynyl-butanamide



To a solution of tert-butyl 3-oxobutanoate (20 g) in toluene (160 mL) was added prop-2-yn-1-amine (6.27 g) drop wise at room temperature. The reaction mixture was heated at 110°C for 16 hours. The reaction mixture was concentrated and partitioned between water and ethyl acetate (300 mL). The organic layer was dried over anhydrous sodium sulfate and concentrated to afford 3-oxo-*N*-prop-2-ynyl-butanamide as a light brown solid, which was used crude in the next step.

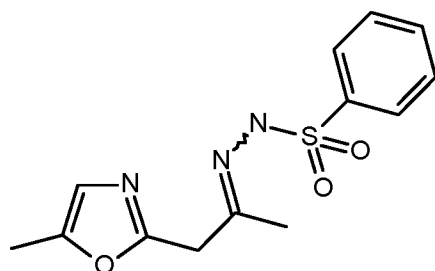
Step 2: Preparation of 1-(5-methyloxazol-2-yl)propan-2-one



To a solution of 3-oxo-*N*-prop-2-ynyl-butanamide (4 g) in acetonitrile (50 mL) was added trichlorogold (0.785 g) and the reaction mixture was stirred at room temperature for 16 hours. The reaction mixture was filtered through celite and partitioned between water and ethyl acetate (200 mL). The organic layer was dried over anhydrous sodium sulfate, concentrated and purified on silica eluting with 50% ethyl acetate in *n*-hexanes to afford 1-(5-methyloxazol-2-yl)propan-2-one as a violet liquid.

¹H NMR (400MHz, CDCl₃) 6.61 (s, 1H), 3.76 (s, 2H), 2.23 (s, 3H), 2.16 (s, 3H)

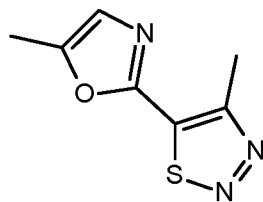
Step 3: Preparation of *N*-[1-methyl-2-(5-methyloxazol-2-yl)ethylidene]amino] benzenesulfonamide



A solution of 1-(5-methyloxazol-2-yl)propan-2-one (1.1 g) in chloroform (20 mL) was added to a mixture of benzenesulfonylhydrazide (1.09 g) and acetic acid (1.12 mL) at room temperature. The resulting mixture was heated at 70°C for 6 hours. The reaction mixture was concentrated under

reduced pressure to afford *N*-[1-methyl-2-(5-methyloxazol-2-yl)ethylidene]amino] benzenesulfonamide as a red solid, which was used crude in the next step.

Step 4: Preparation of 5-methyl-2-(4-methylthiadiazol-5-yl)oxazole



5

A mixture of *N*-[1-methyl-2-(5-methyloxazol-2-yl)ethylidene]amino] benzenesulfonamide (2.1 g) and thionyl chloride (20 mL) was stirred at room temperature for 16 hours. The reaction mixture was concentrated and purified by chromatography on silica eluting with 0–50% ethyl acetate in hexanes to afford 5-methyl-2-(4-methylthiadiazol-5-yl)oxazole as a light brown solid.

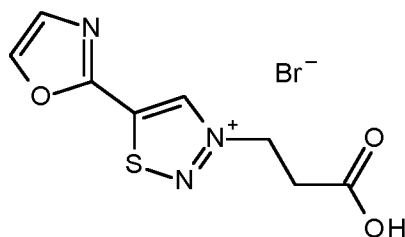
10 ¹H NMR (400MHz, CDCl₃) 6.90 (s, 1H), 2.97 (s, 3H), 2.36 (s, 3H)

Step 5: Preparation of methyl 3-[4-methyl-5-(5-methyloxazol-2-yl)thiadiazol-3-ium-3-yl]propanoate 2,2,2-trifluoroacetate A11

A mixture of 5-methyl-2-(4-methylthiadiazol-5-yl)oxazole (0.2 g) and methyl 3-bromopropoate (8.16 g) was heated at 85°C for 5 days. The reaction mixture was dissolved in water (25 mL) and washed with dichloromethane (2 × 25 mL). The aqueous layer was concentrated and purified by preparative reverse phase HPLC to give (trifluoroacetic acid was present in the eluent) to afford methyl 3-[4-methyl-5-(5-methyloxazol-2-yl)thiadiazol-3-ium-3-yl]propanoate 2,2,2-trifluoroacetate as light yellow liquid.

20 ¹H NMR (400 MHz, D₂O) 7.12 (s, 1H), 5.05 (t, 2H), 3.62 (s, 3H), 3.29 (t, 2H), 3.03 (s, 3H), 2.37 (s, 3H)

Example 4 Preparation of 3-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)propanoic acid bromide: compound A6

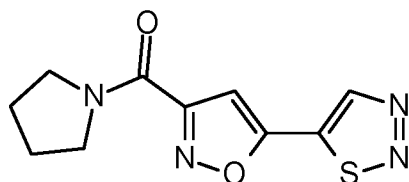
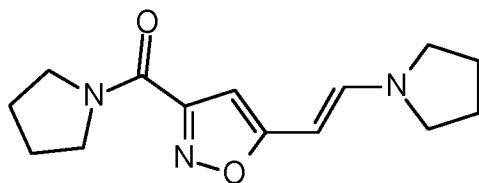


25

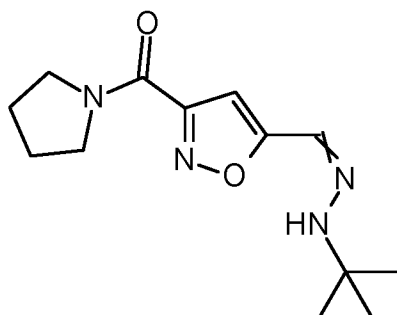
A mixture of 2-(thiadiazol-5-yl)oxazole (0.15 g) and methyl 3-bromopropoate (0.5 g) was heated at 70°C for 16 hours. The reaction mixture was dissolved in water (10.0 mL) and washed with dichloromethane (2 × 20 mL). The aqueous layer was concentrated and purified by preparative reverse phase HPLC (100% water) to afford 3-(5-oxazol-2-ylthiadiazol-3-ium-3-yl)propanoic acid

30 bromide as an off-white solid.

¹H NMR (400 MHz, DMSO-*d*₆) 10.55 (s, 1H), 8.66 (d, 1H), 7.77 (d, 1H), 5.19 (t, 2H), 3.21 (t, 2H) (CO₂H proton missing)

Example 5: Preparation of pyrrolidin-1-yl-[5-(thiadiazol-5-yl)isoxazol-3-yl]methanone**5 Step 1: Preparation of pyrrolidin-1-yl-[5-[(E)-2-pyrrolidin-1-ylvinyl]isoxazol-3-yl]methanone**

To a mixture of methyl 5-methylisoxazole-3-carboxylate (0.5 g) and pyrrolidine (0.591 mL), under nitrogen atmosphere, was added *N,N*-dimethylformamide dimethyl acetal (0.970 mL) and the resulting pale yellow solution was heated at 90°C for 2 hours, then left to cool overnight. No product
 10 was observed so the reaction mixture was heated at 90°C for a further 96 hours. The reaction was cooled and concentrated. Diethyl ether was added to the resulting residue and the mixture was stirred for 30 minutes. The resulting brown precipitate was collected by filtration, washed with diethyl ether and air dried to give pyrrolidin-1-yl-[5-[(E)-2-pyrrolidin-1-ylvinyl]isoxazol-3-yl]methanone.
¹H NMR (400MHz, CDCl₃) 7.30 (d, 1H), 6.03 (s, 1H), 4.95 (d, 1H), 3.85 (t, 2H), 3.63 (t, 2H), 3.33 - 3.22
 15 (m, 4H), 2.00 - 1.87 (m, 8H)

Step 2: Preparation of [5-[(tert-butylhydrazono)methyl]isoxazol-3-yl]-pyrrolidin-1-yl-methanone

To a solution of pyrrolidin-1-yl-[5-[(E)-2-pyrrolidin-1-ylvinyl]isoxazol-3-yl]methanone (0.4 g) in
 20 1,4-dioxane (2 mL) was added (tert-butylamino)ammonium chloride (0.191 g) and the reaction was heated at 80°C for 60 minutes. The reaction was allowed to cool and used in the next step without further purification.

Step 3: Preparation of pyrrolidin-1-yl-[5-(thiadiazol-5-yl)isoxazol-3-yl]methanone

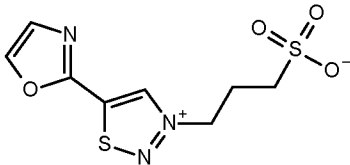
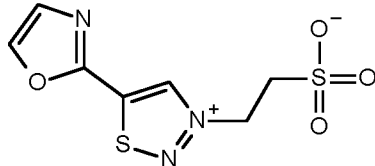
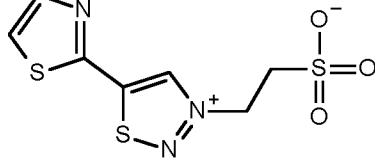
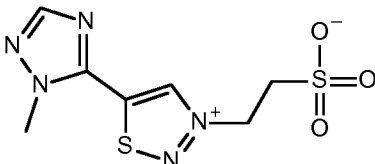
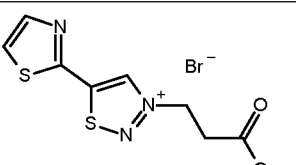
25 A solution of [5-[(tert-butylhydrazono)methyl]isoxazol-3-yl]-pyrrolidin-1-yl-methanone (0.2 g) in dichloromethane (3 mL) was cooled to -78°C and thionyl chloride (0.262 mL) was added drop wise. The reaction was allowed to warm slowly to room temperature. The mixture was partitioned between

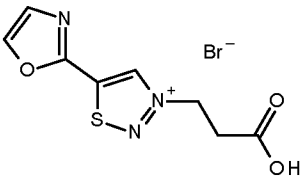
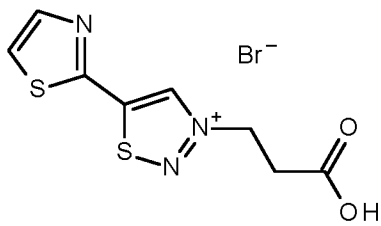
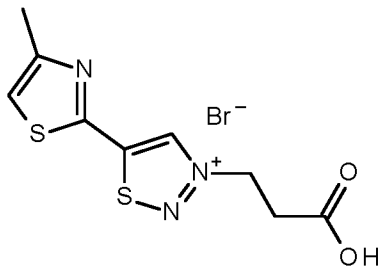
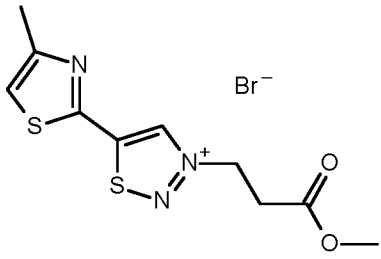
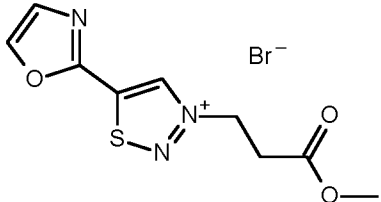
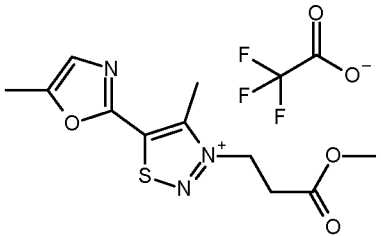
dichloromethane and water and the water layer was extracted with further dichloromethane. The combined organic layers were concentrated and purified by chromatography on silica eluting with a dichloromethane and methanol mixture to give pyrrolidin-1-yl-[5-(thiadiazol-5-yl)isoxazol-3-yl]methanone.

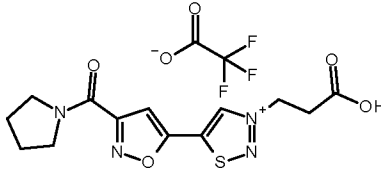
- 5 ¹H NMR (400MHz, CDCl₃) 9.41 (s, 1H), 8.24 (s, 1H), 3.96 - 3.80 (m, 3H), 3.37 - 3.27 (m, 2H), 2.09 - 1.95 (m, 4H)

Additional compounds in Table A were prepared by analogous procedures, from appropriate
10 starting materials.

Table A Physical data for compounds of the invention

Compound Number	Structure	¹ H NMR
A1		(400 MHz, D ₂ O) 10.12 (s, 1 H), 8.15 (s, 1 H), 7.51 (s, 1 H), 5.17 (t, 2 H), 3.05-2.96 (m, 2 H), 2.64-2.51 (m, 2 H)
A2		(400 MHz, D ₂ O) 10.16 (s, 1 H), 8.13 (d, 1 H), 7.49 (d, 1 H), 5.41-5.33 (m, 2 H), 3.69 (dd, 2 H)
A3		(400 MHz, D ₂ O) 10.13 (s, 1 H), 8.05 (d, 1 H), 8.00 (d, 1 H), 5.37-5.28 (m, 2 H), 3.74-3.63 (m, 2 H)
A4		(400 MHz, D ₂ O) 10.19 (s, 1 H), 8.13 (s, 1 H), 5.45-5.35 (m, 2 H), 4.07 (s, 3 H), 3.75-3.64 (m, 2 H)
A5		(400 MHz, DMSO-d ₆) 10.59 (s, 1H), 8.33 (d, 1H), 8.28-8.24 (m, 1H), 5.22-5.16 (m, 2H), 3.65 (s, 3H) (CH ₂ protons under DMSO peak @~2.5ppm)

Compound Number	Structure	¹ H NMR
A6		(400 MHz, DMSO-d ₆) 10.55 (s, 1H), 8.66 (d, 1H), 7.77 (d, 1H), 5.19 (t, 2H), 3.21 (t, 2H) (CO ₂ H proton missing)
A7		(400 MHz, DMSO-d ₆) 10.62 (s, 1H), 8.33 (d, 1H), 8.26 (d, 1H), 5.14 (t, 2H), 3.21 (t, 2H) (CO ₂ H proton missing)
A8		(400 MHz, D ₂ O) 10.01 (s, 1H), 7.61 (s, 1H), 5.18-5.11 (m, 2H), 3.13-3.06 (m, 2H), 2.47 (s, 3H) (CO ₂ H proton missing)
A9		(400 MHz, D ₂ O) 10.08 (s, 1H), 7.62 (s, 1H), 5.27-5.21 (m, 2H), 3.66 (s, 3H), 3.34-3.29 (m, 2H), 2.47 (s, 3H)
A10		(400 MHz, D ₂ O) 10.13 (s, 1H), 8.14 (s, 1H), 7.50 (s, 1H), 5.25 (t, 2H), 3.62 (s, 3H), 3.29 (t, 2H)
A11		(400 MHz, D ₂ O) 7.12 (s, 1H), 5.05 (t, 2H), 3.62 (s, 3H), 3.29 (t, 2H), 3.03 (s, 3H), 2.37 (s, 3H)

Compound Number	Structure	¹ H NMR
A12		(400 MHz, CD ₃ OD) 10.42 (s, 1H), 7.66 (s, 1H), 5.28 (t, 2H), 3.88 (t, 2H), 3.66 (t, 2H), 3.33 - 3.29 (m, 2H), 2.08 - 1.95 (m, 4H) (CO ₂ H proton missing)

BIOLOGICAL EXAMPLES

Post-emergence efficacy

- 5 Seeds of a variety of test species were sown in standard soil in pots. After cultivation for 14 days (post-emergence) under controlled conditions in a glasshouse (at 24/16 °C, day/night; 14 hours light; 65 % humidity), the plants were sprayed with an aqueous spray solution derived from the dissolution of the technical active ingredient formula (I) in a small amount of acetone and a special solvent and emulsifier mixture referred to as IF50 (11.12% Emulsogen EL360 TM + 44.44% N-methylpyrrolidone + 44.44%
- 10 Dowanol DPM glycol ether), to create a 50g/l solution which was then diluted to required concentration using 0.25% or 1% Empicol ESC70 (Sodium lauryl ether sulphate) + 1% ammonium sulphate as diluent. The test plants were then grown in a glasshouse under controlled conditions (at 24/16 °C, day/night; 14 hours light; 65 % humidity) and watered twice daily. After 13 days the test was evaluated (100 = total damage to plant; 0 = no damage to plant).
- 15 The results are shown in Table B (below). A value of n/a indicates that this combination of weed and test compound was not tested/assessed.

Test plants:

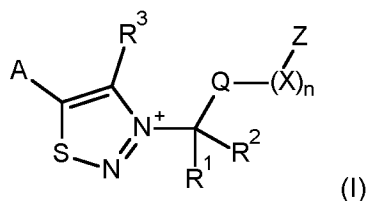
- Ipomoea hederacea* (IPOHE), *Euphorbia heterophylla* (EPHHL), *Chenopodium album* (CHEAL),
- 20 *Amaranthus palmeri* (AMAPA), *Lolium perenne* (LOLPE), *Digitaria sanguinalis* (DIGSA), *Eleusine indica* (ELEIN), *Echinochloa crus-galli* (ECHCG), *Setaria faberi* (SETFA)

Table B Control of weed species by compounds of formula (I) after post-emergence application

Compound Number	Application Rate g/Ha	AMAPA	CHEAL	EPHHL	IPHE	ELEIN	LOLPE	DIGSA	SETFA	ECHCG
A1	500	60	40	60	50	20	20	30	30	40
A2	500	100	70	-	70	70	70	70	80	40
A3	500	100	90	100	60	60	50	50	70	40
A4	500	100	80	100	80	90	70	70	70	60
A5	500	50	20	40	10	100	30	100	80	100
A6	500	100	80	100	20	90	80	90	100	100
A7	500	100	40	100	10	60	20	90	80	90
A8	500	100	30	60	10	90	30	100	80	90
A9	500	60	30	40	10	90	10	100	100	100
A10	500	100	50	50	10	90	50	80	100	60
A11	500	30	30	10	10	10	0	10	10	30
A12	500	50	40	40	30	60	40	100	90	80

CLAIMS:

1. A compound of formula (I) or an agronomically acceptable salt or zwitterionic species thereof:



wherein

10 R^1 is selected from the group consisting of hydrogen, halogen, C_1 - C_6 alkyl, C_2 - C_6 alkenyl, C_2 - C_6 alkynyl, C_3 - C_6 cycloalkyl, C_1 - C_6 haloalkyl, $-OR^7$, $-OR^{15a}$, $-N(R^6)S(O)_2R^{15}$, $-N(R^6)C(O)R^{15}$, $-N(R^6)C(O)OR^{15}$, $-N(R^6)C(O)NR^{16}R^{17}$, $-N(R^6)CHO$, $-N(R^{7a})_2$ and $-S(O)_rR^{15}$;

15 R^2 is selected from the group consisting of hydrogen, halogen, C_1 - C_6 alkyl and C_1 - C_6 haloalkyl; or R^1 and R^2 together with the carbon atom to which they are attached form a C_3 - C_6 cycloalkyl ring or a 3- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O;

20 and wherein when R^1 is selected from the group consisting of $-OR^7$, $-OR^{15a}$, $-N(R^6)S(O)_2R^{15}$, $-N(R^6)C(O)R^{15}$, $-N(R^6)C(O)OR^{15}$, $-N(R^6)C(O)NR^{16}R^{17}$, $-N(R^6)CHO$, $-N(R^{7a})_2$ and $-S(O)_rR^{15}$, then R^2 is selected from the group consisting of hydrogen and C_1 - C_6 alkyl;

25 R^3 is selected from the group consisting of hydrogen, halogen, cyano, nitro, $-S(O)_rR^{15}$, C_1 - C_6 alkyl, C_1 - C_6 fluoroalkyl, C_1 - C_6 fluoroalkoxy, C_1 - C_6 alkoxy, C_3 - C_6 cycloalkyl and $-N(R^6)_2$;

30 Q is $(CR^{1a}R^{2b})_m$;

m is 0, 1, 2 or 3;

35 each R^{1a} and R^{2b} are independently selected from the group consisting of hydrogen, halogen, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, $-OH$, $-OR^7$, $-OR^{15a}$, $-NH_2$, $-NHR^7$, $-NHR^{15a}$, $-N(R^6)CHO$, $-NR^{7b}R^{7c}$ and $-S(O)_rR^{15}$; or

each R^{1a} and R^{2b} together with the carbon atom to which they are attached form a C_3 - C_6 cycloalkyl ring or a 3- to 6- membered heterocyclyl, which comprises 1 or 2 heteroatoms individually selected from N and O;

35

each R^6 is independently selected from hydrogen and C_1 - C_6 alkyl;

each R^7 is independently selected from the group consisting of C_1 - C_6 alkyl, $-S(O)_2R^{15}$, $-C(O)R^{15}$, $-C(O)OR^{15}$ and $-C(O)NR^{16}R^{17}$;

5 each R^{7a} is independently selected from the group consisting of $-S(O)_2R^{15}$, $-C(O)R^{15}$, $-C(O)OR^{15}$, $-C(O)NR^{16}R^{17}$ and $-C(O)NR^{6}R^{15a}$;

10 R^{7b} and R^{7c} are independently selected from the group consisting of C_1 - C_6 alkyl, $-S(O)_2R^{15}$, $-C(O)R^{15}$, $-C(O)OR^{15}$, $-C(O)NR^{16}R^{17}$ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different; or R^{7b} and R^{7c} together with the nitrogen atom to which they are attached form a 4- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N, O and S;

15 A is a 5-membered heteroaryl ring comprising 1, 2, 3 or 4 heteroatoms each independently selected from the group consisting of N, O and S, wherein said 5-membered heteroaryl ring is optionally substituted by 1, 2, or 3 R^8 substituents;

20 and when A is a 5-membered heteroaryl ring substituted by R^8 on one or more ring carbon atoms, each of said R^8 substituents is independently selected from the group consisting of halogen, nitro, cyano, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OH$, $-OR^7$, $-S(O)_2R^{15}$, $-NR^6S(O)_2R^{15}$, $-C(O)OR^{10}$, $-C(O)R^{15}$, $-C(O)NR^{16}R^{17}$, $-S(O)_2NR^{16}R^{17}$, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 halocycloalkyl, C_3 - C_6 cycloalkoxy, C_2 - C_6 alkenyl, C_2 - C_6 haloalkenyl, C_2 - C_6 alkynyl, C_1 - C_3 alkoxy C_1 - C_3 alkyl-, hydroxy C_1 - C_6 alkyl-, C_1 - C_3 alkoxy C_1 - C_3 alkoxy-, C_1 - C_6 haloalkoxy, C_1 - C_3 haloalkoxy C_1 - C_3 alkyl-, C_3 - C_6 alkenyloxy, C_3 - C_6 alkynyloxy, $-C(R^6)=NOR^6$, phenyl and a 5- or 6- membered heteroaryl, which

25 comprises 1, 2, 3 or 4 heteroatoms independently selected from N, O and S, and wherein said phenyl or heteroaryl are optionally substituted by 1, 2 or 3 R^9 substituents, which may be the same or different;

30 and/or when A is a 5-membered heteroaryl ring substituted by an R^8 on a ring nitrogen atom, said R^8 is selected from the group consisting of $-OR^7$, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 halocycloalkyl, C_3 - C_6 cycloalkoxy, C_2 - C_6 alkenyl, C_2 - C_6 haloalkenyl, C_2 - C_6 alkynyl, C_1 - C_3 alkoxy C_1 - C_3 alkyl-, C_1 - C_3 alkoxy C_1 - C_3 alkoxy-, C_1 - C_6 haloalkoxy, C_1 - C_3 haloalkoxy C_1 - C_3 alkyl-, C_3 - C_6 alkenyloxy and C_3 - C_6 alkynyloxy;

35 each R^9 is independently selected from the group consisting of $-OH$, halogen, cyano, $-N(R^6)_2$, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkyl and C_1 - C_4 haloalkoxy;

40 X is independently selected from the group consisting of C_3 - C_6 cycloalkyl, phenyl, a 5- or 6-membered heteroaryl, which comprises 1, 2, 3 or 4 heteroatoms independently selected from N, O and S, and a 4- to 6- membered heterocyclyl, which comprises 1, 2 or 3 heteroatoms independently selected from N, O and S, and wherein said cycloalkyl, phenyl, heteroaryl or heterocyclyl moieties are optionally substituted by 1 or 2 R^9 substituents and wherein the

aforementioned CR¹R² Q and Z moieties may be attached at any position of said cycloalkyl, phenyl, heteroaryl or heterocyclyl moieties;

n is 0 or 1;

5

Z is selected from the group consisting of -C(O)OR¹⁰, -CH₂OH, -CHO, -C(O)NHOR¹¹, -C(O)NHCN, -OC(O)NHOR¹¹, -OC(O)NHCN, -NR⁶C(O)NHOR¹¹, -NR⁶C(O)NHCN, -C(O)NHS(O)₂R¹², -OC(O)NHS(O)₂R¹², -NR⁶C(O)NHS(O)₂R¹², -S(O)₂OR¹⁰, -OS(O)₂OR¹⁰, -NR⁶S(O)₂OR¹⁰, -NR⁶S(O)OR¹⁰, -NHS(O)₂R¹⁴, -S(O)OR¹⁰, -OS(O)OR¹⁰, -S(O)₂NHCN, -S(O)₂NHC(O)R¹⁸, -S(O)₂NHS(O)₂R¹², -OS(O)₂NHCN, -OS(O)₂NHS(O)₂R¹², -OS(O)₂NHC(O)R¹⁸, -NR⁶S(O)₂NHCN, -NR⁶S(O)₂NHC(O)R¹⁸, -N(OH)C(O)R¹⁵, -ONHC(O)R¹⁵, -NR⁶S(O)₂NHS(O)₂R¹², -P(O)(R¹³)(OR¹⁰), -P(O)H(OR¹⁰), -OP(O)(R¹³)(OR¹⁰), -NR⁶P(O)(R¹³)(OR¹⁰) and tetrazole;

10

15

R¹⁰ is selected from the group consisting of hydrogen, C₁-C₆alkyl, phenyl and benzyl, and wherein said phenyl or benzyl are optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

20

R¹¹ is selected from the group consisting of hydrogen, C₁-C₆alkyl and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

25

R¹² is selected from the group consisting of C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, -OH, -N(R⁶)₂ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

30

R¹³ is selected from the group consisting of -OH, C₁-C₆alkyl, C₁-C₆alkoxy and phenyl;

R¹⁴ is C₁-C₆haloalkyl;

R¹⁵ is selected from the group consisting of C₁-C₆alkyl and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

35

R^{15a} is phenyl, wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

40

R¹⁶ and R¹⁷ are independently selected from the group consisting of hydrogen and C₁-C₆alkyl;

or R¹⁶ and R¹⁷ together with the nitrogen atom to which they are attached form a 4- to 6-membered heterocyclyl ring which optionally comprises one additional heteroatom individually selected from N, O and S;

R¹⁸ is selected from the group consisting of hydrogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, -N(R⁶)₂ and phenyl, and wherein said phenyl is optionally substituted by 1, 2 or 3 R⁹ substituents, which may be the same or different;

5

and r is 0, 1 or 2.

2. A compound according to claim 1, wherein R¹ and R² are each independently hydrogen or C₁-C₆alkyl.

10

3. A compound according to claim 1 or claim 2, wherein m is 1 or 2.

4. A compound according to any one of claims 1 to 3, wherein R³, is selected from the group consisting of hydrogen, C₁-C₆alkyl and C₁-C₆alkoxy.

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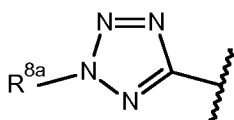
5. A compound according to any one of claims 1 to 4, wherein R³ is hydrogen.

6. A compound according to any one of claims 1 to 5, wherein A is selected from the group consisting of tetrazol-5-yl, 1,2,4-triazol-3-yl, 1,2,4-triazol-5-yl, isoxazol-3-yl, oxazol-2-yl, thiazol-2-yl, 1,3,4-thiadiazol-2-yl, triazol-4-yl, triazol-5-yl, pyrazol-3-yl, pyrazol-5-yl, 1,3,4-oxadiazol-2-yl, 1,2,4-thiadiazol-5-yl, oxazol-4-yl, imidazol-2-yl, isothiazol-5-yl, 2-thienyl, 3-furyl, 2-furyl, isothiazol-4-yl, thiazol-4-yl, 3-thienyl, imidazol-5-yl, isoxazol-5-yl and 1,2,4-oxadiazol-5-yl wherein the heteroaryl may, where feasible, be optionally substituted by 1, 2 or 3 R⁸ substituents, which may be the same or different and R⁸ is as defined in claim 1.

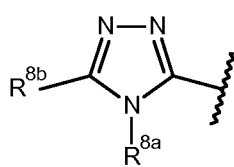
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7. A compound according to any one of claims 1 to 6, wherein A is selected from the group consisting of formula A-I to A-XXVIII below

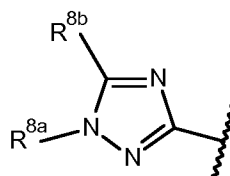
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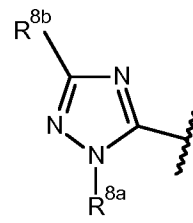
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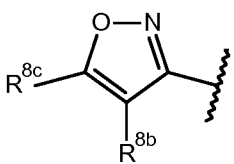
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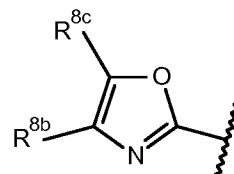
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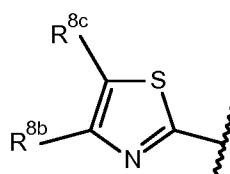
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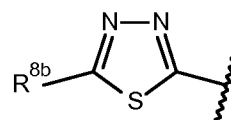
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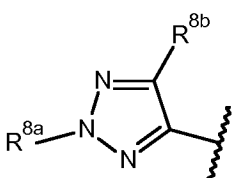
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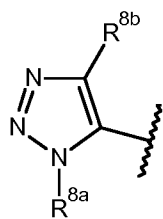
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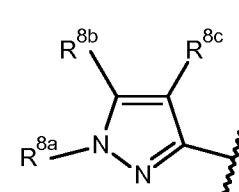
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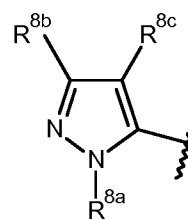
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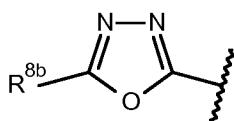
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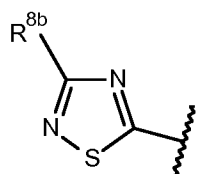
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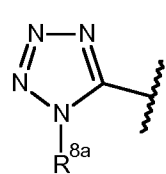
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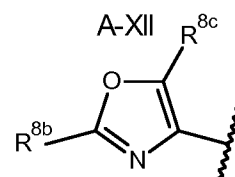
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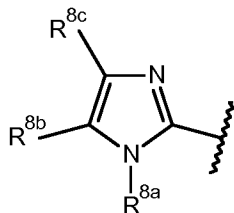
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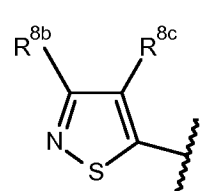
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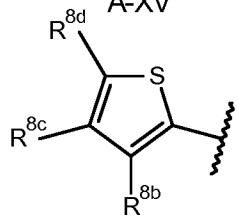
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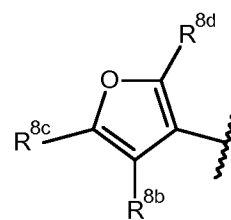
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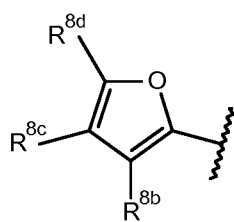
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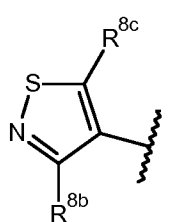
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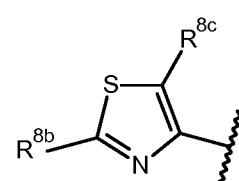
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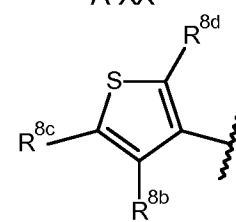
A-XXI



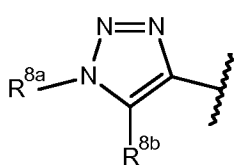
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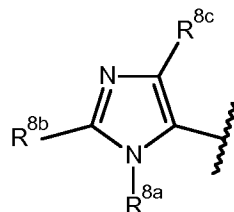
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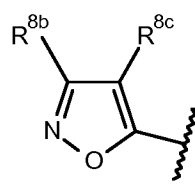
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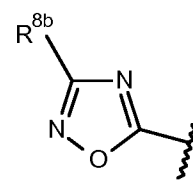
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A-XXVI



A-XXVII



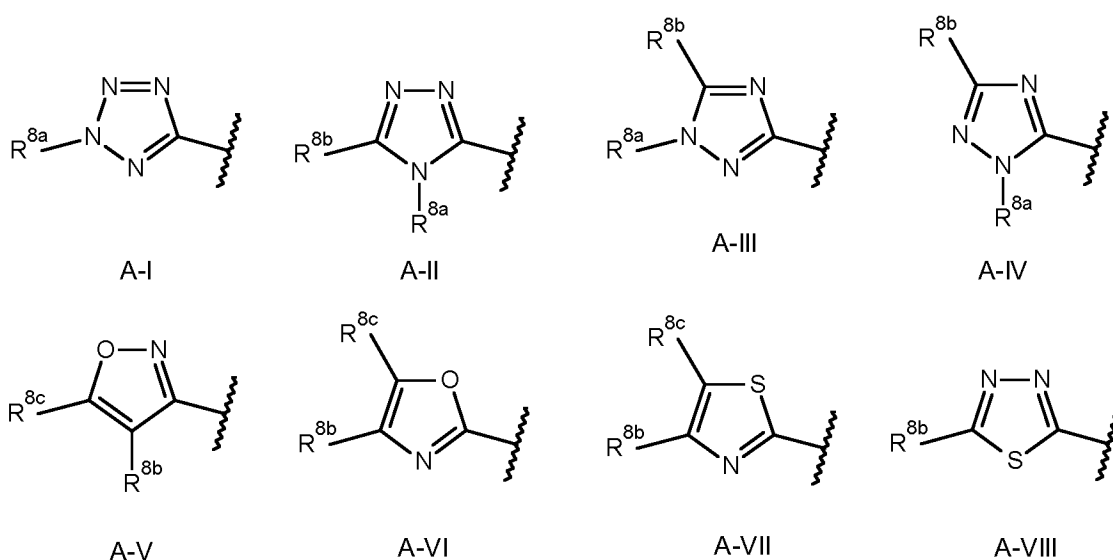
A-XXVIII

wherein the jagged line defines the point of attachment to the thiadiazole moiety of the compound;

R^{8a} is selected from the group consisting of hydrogen, C_1 - C_6 alkyl and C_1 - C_6 haloalkyl;

- 5 each R^{8b} , R^{8c} and R^{8d} are independently selected from the group consisting of hydrogen, halogen, nitro, cyano, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OH$, $-OR^7$, $-S(O)_rR^{15}$, $-NR^6S(O)_2R^{15}$, $-C(O)OR^{10}$, $-C(O)R^{15}$, $-C(O)NR^{16}R^{17}$, $-S(O)_2NR^{16}R^{17}$, C_1 - C_6 alkyl and C_1 - C_6 haloalkyl; and R^6 , R^7 , R^{10} , R^{15} , R^{16} , R^{17} and r are as defined in claim 1.

- 10 8. A compound according to any one of claims 1 to 7, wherein A is selected from the group consisting of formula A-I to A-VIII below



- 15 wherein the jagged line defines the point of attachment to the thiadiazole moiety of the compound ;

R^{8a} is hydrogen or C_1 - C_6 alkyl;

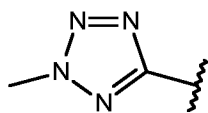
each R^{8b} , R^{8c} and R^{8d} are independently selected from the group consisting of hydrogen, halogen, $-NH_2$, $-NHR^7$, $-N(R^7)_2$, $-OR^7$, C_1 - C_6 alkyl and C_1 - C_6 haloalkyl;

- 20 and R^7 is as defined in claim 1.

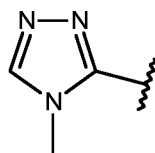
9. A compound according to claims 7 or 8, wherein R^{8a} is hydrogen or methyl and each R^{8b} , R^{8c} and R^{8d} are independently selected from the group consisting of hydrogen, chloro, $-NH_2$, $-NHMe$, $-OMe$, methyl, *iso*-propyl and trifluoromethyl.

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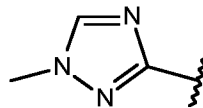
10. A compound according to any one of claims 1 to 9, wherein A is selected from the group consisting of formula A-Ia to A-VIIIa below



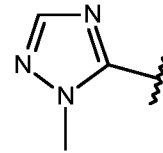
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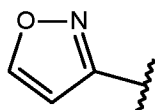
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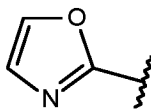
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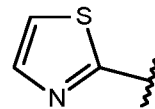
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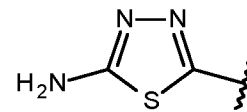
A-Va



A-VIa



A-VIIa



A-VIIIa

wherein the jagged line denotes the point of attachment to the thiadiazole moiety of the compound.

- 5 11. A compound according to any one of claims 1 to 10, wherein Z is selected from the group consisting of $-C(O)OR^{10}$, $-C(O)NHS(O)_2R^{12}$, $-S(O)_2OR^{10}$, $-OS(O)_2OR^{10}$ and $-P(O)(R^{13})(OR^{10})$.
12. A compound according to any one of claims 1 to 11, wherein Z is $-C(O)OH$ or $-S(O)_2OH$.
- 10 13. An agrochemical composition comprising a herbicidally effective amount of a compound of formula (I) as defined in any one of claims 1 to 12 and an agrochemically-acceptable diluent or carrier.
14. A method of controlling unwanted plant growth, comprising applying a compound of formula (I)
- 15 as defined in any one of claims 1 to 12, or a herbicidal composition according to claim 13 or claim 13, to the unwanted plants or to the locus thereof.
15. Use of a compound of formula (I) as defined in any one of claims 1 to 12, as a herbicide.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2019/081022

A. CLASSIFICATION OF SUBJECT MATTER INV. C07D417/04 C07F9/38 C07F9/48 A01N43/82 ADD.		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) C07D A01N		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) EPO-Internal, WPI Data, CHEM ABS Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 482 916 A (HANASAKI YASUAKI [JP] ET AL) 9 January 1996 (1996-01-09) -----	1-15
A	CN 103 896 934 A (YANCHENG INST TECHNOLOGY) 2 July 2014 (2014-07-02) the whole document -----	1-15
A	US 3 557 133 A (HOLTSCHMIDT HANS ET AL) 19 January 1971 (1971-01-19) the whole document -----	1-15
☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
* Special categories of cited documents :		
"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family	
Date of the actual completion of the international search 13 January 2020	Date of mailing of the international search report 24/01/2020	
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Bedel, Christian	

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2019/081022

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