

Novel heteroaryl-triazole compounds as pesticides

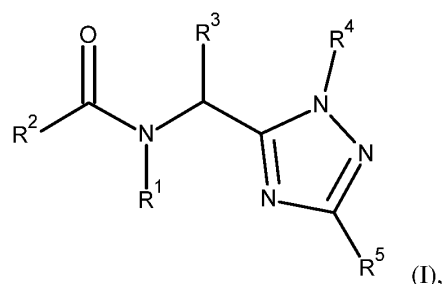
The present invention relates to novel heteroaryl-triazole compounds, to formulations and compositions comprising such compounds and to their use in the control of animal pests including arthropods and insects in plant protection and to their use for the control of ectoparasites on animals.

- 5 Certain heteroaryl-triazole compounds are disclosed for the use in controlling ectoparasites on animals in WO 2017/192385 and for the use in controlling animal pests including arthropods and insects in the field of plant protection in WO 2019/170626 and WO 2019/215198. Further, the patent applications WO 2019/197468, WO 2019/201835, WO 2019/202077 and WO 2019/206799 disclose certain heteroaryl-triazole compounds for the use in controlling ectoparasites on animals and for the control of animal pests
10 including arthropods and insects in the field of plant protection. WO 2020/002563, WO 2020/053364, WO 2020/053365, WO 2020/079198, WO 2020/094363 describe azole-amide compounds all of which can be used as insecticides.

- Modern plant protection products and veterinary ectoparasiticides have to meet many demands, for example in relation to efficacy, persistence, spectrum and resistance breaking properties. Questions of
15 toxicity, the combinability with other active compounds or formulation auxiliaries play a role, as well as the question of the expense that the synthesis of an active compound requires. Furthermore, resistances may occur. For all these reasons, the search for novel crop protection compositions or veterinary ectoparasiticides cannot be considered to be complete, and there is a constant need for novel compounds having properties which, compared to the known compounds, are improved at least in respect of individual
20 aspects.

It was an object of the present invention to provide compounds which widen the spectrum of the pesticides in various aspects.

The present invention therefore provides compounds of the general formula (I)



- 25 in which (Configuration 1-1):

R¹ is hydrogen;

- R² is phenyl wherein the phenyl is substituted with a total of one to two substituents, provided the substituent(s) are not on either carbon adjacent to the carbon bonded to the C=O group and one substituent is selected from group A consisting of
- 5 cyclopropyl, methylsulfonyl, ethylsulfonyl, isopropylsulfonyl, cyclopropylsulfonyl, difluoromethylsulfonyl, trifluoroethylsulfonyl, trifluoromethylsulfonyl, 1-cyanocyclopropyl, and -SF₅;
- and the other substituent is independently selected from group B consisting of
- chlorine, trifluoromethyl, and trifluoromethoxy; or
- R² is thiophene optionally substituted by one substituent selected from the group consisting of
- 10 fluorine, chlorine, bromine, and trifluoromethyl; or
- R² is pyrazol optionally substituted by one to two substituents selected from the group consisting of
- fluorine, chlorine, bromine, methyl, and trifluoromethyl;
- R³ is C₁-C₃alkyl;
- 15 R⁴ is pyridine, pyrimidine or pyrazine wherein the pyridine, pyrimidine or pyrazine is optionally substituted with one substituent selected from the group consisting of
- chlorine, and -CN;
- R⁵ is ethyl, n-propyl, iso-propyl, difluoromethyl, cyclopropyl, methoxy, ethoxy, iso-propoxy or halogen.
- 20 The compounds of the formula (I) likewise encompass any diastereomers or enantiomers and E/Z isomers which exist, and also salts and N-oxides of compounds of the formula (I), and the use thereof for control of animal pests.

Preferred radical definitions for the formulae specified above and hereinafter are given below.

The present invention furthermore provides compounds of the general formula (I)

- 25 in which (Configuration 1-2):

R¹ is hydrogen;

- R² is phenyl wherein the phenyl is substituted with a total of one to two substituents, provided the substituent(s) are not on either carbon adjacent to the carbon bonded to the C=O group and one substituent is selected from group A consisting of
- 5 methylsulfonyl, ethylsulfonyl, isopropylsulfonyl, cyclopropylsulfonyl, difluoromethylsulfonyl, trifluoroethylsulfonyl, trifluoromethylsulfonyl, and -SF₅ and cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl;
- and the other substituent is independently selected from group B consisting of
- 10 fluorine, chlorine, bromine, iodine, -CN, difluoromethyl, trifluoromethyl, difluoromethoxy, trifluoromethoxy and cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl; or
- R² is thiophene optionally substituted by one substituent selected from the group consisting of
- 15 fluorine, chlorine, bromine, and trifluoromethyl; or
- R² is pyrazol optionally substituted by one to two substituents selected from the group consisting of
- fluorine, chlorine, bromine, methyl, and trifluoromethyl;
- R³ is C₁-C₃alkyl;
- 20 R⁴ is pyridine, pyrimidine or pyrazine wherein the pyridine, pyrimidine or pyrazine is optionally substituted with one substituent selected from the group consisting of
- chlorine, and -CN;
- R⁵ is ethyl, n-propyl, iso-propyl, difluoromethyl, cyclopropyl, methoxy, ethoxy, iso-propoxy or halogen.
- 25 Preference (Configuration 2-1) is given to the compounds of the formula (I) in which
- R¹ is hydrogen;
- R² is 3-chloro-5-cyclopropylphenyl, 3-chloro-5-methylsulfonylphenyl, 5-chloro-3-thienyl, 3-chloro-5-(2,2,2-trifluoroethylsulfonyl)phenyl, 3-chloro-5-isopropylsulfonylphenyl, 3-cyclopropylsulfonyl-5-(trifluoromethyl)phenyl, 3-chloro-5-ethylsulfonylphenyl, 3-chloro-5-cyclopropylsulfonylphenyl, 3-ethylsulfonyl-5-(trifluoromethyl)phenyl, 3-cyclopropyl-5-
- 30

(trifluoromethoxy)phenyl, 3-chloro-5-(difluoromethylsulfonyl)phenyl, 3-methylsulfonyl-5-(trifluoromethyl)phenyl, 1-methyl-5-(trifluoromethyl)-1H-pyrazol-3-yl, 3-chloro-5-(pentafluoro- λ 6-sulfanyl)phenyl, 3-chloro-5-(1-cyanocyclopropyl)phenyl, or 3-chloro-5-(trifluoromethylsulfonyl)phenyl;

5 R³ is methyl;

R⁴ is 5-cyanopyridin-2-yl, pyrimidin-2-yl, 5-chloropyrimidin-2-yl or 5-chloropyridin-2-yl;

R⁵ is ethyl, n-propyl, iso-propyl, difluoromethyl, cyclopropyl, methoxy, ethoxy, iso-propoxy, iodine, bromine or chlorine.

Preference (Configuration 2-2) is also given to the compounds of the formula (I) in which

10 R¹ is hydrogen;

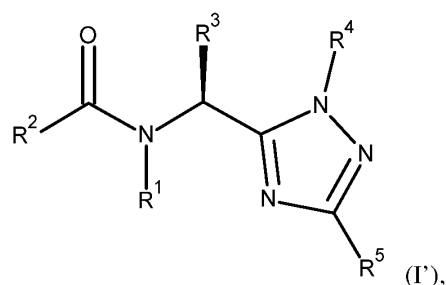
R² is 3-chloro-5-cyclopropylphenyl, 3-chloro-5-methylsulfonylphenyl, 5-chloro-3-thienyl, 3-chloro-5-(2,2,2-trifluoroethylsulfonyl)phenyl, 3-chloro-5-isopropylsulfonylphenyl, 3-cyclopropylsulfonyl-5-(trifluoromethyl)phenyl, 3-chloro-5-ethylsulfonylphenyl, 3-chloro-5-cyclopropylsulfonylphenyl, 3-ethylsulfonyl-5-(trifluoromethyl)phenyl, 3-cyclopropyl-5-(trifluoromethoxy)phenyl, 3-chloro-5-(difluoromethylsulfonyl)phenyl, 3-methylsulfonyl-5-(trifluoromethyl)phenyl, 1-methyl-5-(trifluoromethyl)-1H-pyrazol-3-yl, 3-chloro-5-(pentafluoro-λ6-sulfanyl)phenyl, 3-chloro-5-(1-cyanocyclopropyl)phenyl, 3-chloro-5-(trifluoromethylsulfonyl)phenyl, 3-bromo-5-[1-(trifluoromethyl)cyclopropyl]phenyl, 3-cyclopropyl-5-methylsulfonylphenyl, 3-(difluoromethyl)-5-methylsulfonylphenyl, 3-cyclopropyl-5-cyclopropylsulfonylphenyl, 3-fluoro-5-methylsulfonylphenyl, 3-(difluoromethoxy)-5-methylsulfonylphenyl, 3-methylsulfonyl-5-(trifluoromethoxy)phenyl, 3-cyclopropyl-5-(difluoromethyl)phenyl, 3-bromo-5-(1-fluorocyclopropyl)phenyl, 3-bromo-5-(2,2-difluorocyclopropyl)phenyl, 3-cyclopropyl-5-(difluoromethoxy)phenyl, 3-cyclopropylsulfonyl-5-(difluoromethoxy)phenyl, 3-cyclopropylsulfonyl-5-(trifluoromethoxy)phenyl, or 3-cyclopropylsulfonyl-5-(difluoromethyl)phenyl;

R^3 is methyl;

R⁴ is 5-cyanopyridin-2-yl, pyrimidin-2-yl, 5-chloropyrimidin-2-yl or 5-chloropyridin-2-yl;

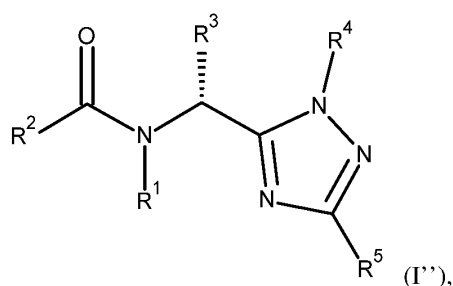
R⁵ is ethyl, n-propyl, iso-propyl, difluoromethyl, cyclopropyl, methoxy, ethoxy, iso-propoxy, iodine, bromine or chlorine.

30 In a further preferred embodiment, the invention relates to compounds of the formula (I') in which R³ is C₁-C₃alkyl, especially preferred Me, and



in which the structural elements R¹, R², R⁴ and R⁵ have the meanings given in Configuration (1-1) or in Configuration (2-1) or in Configuration (1-2) or in Configuration (2-2).

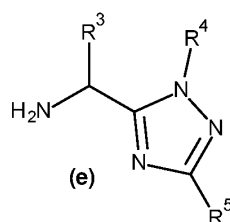
In a further preferred embodiment, the invention relates to compounds of the formula (I') in which R³ is
 5 C₁-C₃alkyl, especially preferred Me, and



in which the structural elements R², R⁴ and R⁵ have the meanings given in Configuration (1-1) or in Configuration (2-1) or in Configuration (1-2) or in Configuration (2-2).

In accordance with a further aspect, the present invention covers intermediate compounds which are
 10 useful for the preparation of the compounds of general formula (I), *supra*.

Particularly, the invention covers the intermediate compounds of general formula (e):

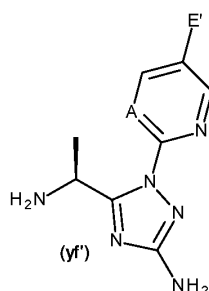


in which the structural elements R³, R⁴ and R⁵ have the meaning given in Configuration (1-1) or in Configuration (2-1) or in Configuration (1-2) or in Configuration (2-2), including the intermediate
 15 compounds INT-1 to INT-8, INT-10 to INT-12, INT-18, INT-20, INT-22, INT-24, INT-25 and INT-26, salts thereof and in case of amine hydrochlorides the free amines:

- INT-1: 6-[5-[(1S)-1-aminoethyl]-3-ethyl-1,2,4-triazol-1-yl]pyridine-3-carbonitrile hydrochloride
- INT-2: 6-[5-[(1S)-1-aminoethyl]-3-isopropyl-1,2,4-triazol-1-yl]pyridine-3-carbonitrile
- INT-3: 6-[5-[(1S)-1-aminoethyl]-3-cyclopropyl-1,2,4-triazol-1-yl]pyridine-3-carbonitrile hydrochloride
- INT-4: 6-[5-(1-aminoethyl)-3-(difluoromethyl)-1,2,4-triazol-1-yl]pyridine-3-carbonitrile
- 5 INT-5: 6-[5-[(1S)-1-aminoethyl]-3-methoxy-1,2,4-triazol-1-yl]pyridine-3-carbonitrile
- INT-6: 6-[5-[(1S)-1-aminoethyl]-3-ethoxy-1,2,4-triazol-1-yl]pyridine-3-carbonitrile
- INT-7: 6-[5-[(1S)-1-aminoethyl]-3-isopropoxy-1,2,4-triazol-1-yl]pyridine-3-carbonitrile
- INT-8: (1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethanamine hydrochloride
- 10 INT-10: 1-[3-chloro-1-(pyrimidin-2-yl)-1H-1,2,4-triazol-5-yl]ethanamine
- INT-11: 1-[3-bromo-1-(pyrimidin-2-yl)-1H-1,2,4-triazol-5-yl]ethanamine
- INT-12: 1-[3-iodo-1-(pyrimidin-2-yl)-1H-1,2,4-triazol-5-yl]ethanamine
- INT-18: (1S)-1-[1-(5-chloropyridin-2-yl)-3-methoxy-1H-1,2,4-triazol-5-yl]ethanamine
- INT-20: (1S)-1-[1-(5-chloropyridin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethanamine hydrochloride
- 15 INT-22: (1S)-1-[1-(5-chloropyrimidin-2-yl)-3-methoxy-1H-1,2,4-triazol-5-yl]ethanamine
- INT-24: 6-[5-(1-aminoethyl)-3-iodo-1H-1,2,4-triazol-1-yl]nicotinonitrile
- INT-25: 6-[5-(1-aminoethyl)-3-bromo-1,2,4-triazol-1-yl]pyridine-3-carbonitrile
- INT-26: 6-[5-(1-aminoethyl)-3-chloro-1,2,4-triazol-1-yl]pyridine-3-carbonitrile

- The invention also covers the intermediates 3-chloro-5-[(difluoromethyl)sulfanyl]benzoic acid (INT-9),
- 20 3-cyclopropyl-5-(difluoromethyl)benzoic acid (INT-13), 3-cyclopropyl-5-iodobenzoic acid (INT-14), 3-chloro-5-[(difluoromethyl)sulfonyl]benzoic acid (INT-16), 3-cyano-5-cyclopropylbenzoic acid (INT-15), 3-cyclopropyl-5-methylsulfonyl-benzoic acid (INT-17), 3-cyclopropyl-5-cyclopropylsulfonyl-benzoic acid (INT-19) acid, and 3-fluoro-5-methylsulfonyl-benzoic acid (INT-21) and salts thereof.

The invention also covers the intermediate compounds of general formula (y[¶]):



in which E' is hydrogen, chlorine or -CN and A is N or CH, including salts thereof. Preference is given to the hydrochloric acid salts, including:

INT-23: 5-[(1S)-1-aminoethyl]-1-(pyrimidin-2-yl)-1H-1,2,4-triazol-3-amine hydrochloride

- 5 The compounds of the formula (I) may possibly also, depending on the nature of the substituents, be in the form of stereoisomers, i.e. in the form of geometric and/or optical isomers or isomer mixtures of varying composition. This invention provides both the pure stereoisomers and any desired mixtures of these isomers, even though it is generally only compounds of the formula (I) that are discussed here.

- 10 However, preference is given in accordance with the invention to using the optically active, stereoisomeric forms of the compounds of the formula (I) and salts thereof.

The invention therefore relates both to the pure enantiomers and diastereomers and to mixtures thereof for controlling animal pests, including arthropods and particularly insects.

- 15 If appropriate, the compounds of the formula (I) may be present in various polymorphic forms or as a mixture of various polymorphic forms. Both the pure polymorphs and the polymorph mixtures are provided by the invention and can be used in accordance with the invention.

Definitions

The person skilled in the art is aware that, if not stated explicitly, the expressions "a" or "an" as used in the present application may, depending on the situation, mean "one (1)", "one (1) or more" or "at least one (1)".

- 20 For all the structures described herein, such as ring systems and groups, adjacent atoms must not be -O-O- or -O-S-.

- Structures having a variable number of possible carbon atoms (C atoms) may be referred to in the present application as C_{lower limit of carbon atoms}-C_{upper limit of carbon atoms} structures (C_{LL}-C_{UL} structures), in order thus to be stipulated more specifically. Example: an alkyl group may consist of 3 to 10 carbon atoms and in that case
25 corresponds to C₃-C₁₀alkyl. Ring structures composed of carbon atoms and heteroatoms may be referred

to as "LL- to UL-membered" structures. One example of a 6-membered ring structure is toluene (a 6-membered ring structure substituted by a methyl group).

If a collective term for a substituent, for example C_{LL}-C_{UL}alkyl, is at the end of a composite substituent, for example C_{LL}-C_{UL}cycloalkyl-C_{LL}-C_{UL}alkyl, the constituent at the start of the composite substituent, for example the C_{LL}-C_{UL}cycloalkyl, may be mono- or polysubstituted identically or differently and independently by the latter substituent, for example C_{LL}-C_{UL}alkyl. All the collective terms used in this application for chemical groups, cyclic systems and cyclic groups can be stipulated more specifically through the addition "C_{LL}-C_{UL}" or "LL- to UL-membered".

In the definitions of the symbols given in the above formulae, collective terms which are generally representative of the following substituents were used:

Halogen relates to elements of the 7th main group, preferably fluorine, chlorine, bromine and iodine, more preferably fluorine, chlorine and bromine, and even more preferably fluorine and chlorine.

Examples of heteroatom are N, O, S, P, B, Si. Preferably, the term "heteroatom" relates to N, S and O.

According to the invention, "alkyl" – on its own or as part of a chemical group – represents straight-chain or branched hydrocarbons preferably having 1 to 6 carbon atoms, for example methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, s-butyl, t-butyl, pentyl, 1-methylbutyl, 2-methylbutyl, 3-methylbutyl, 1,2-dimethylpropyl, 1,1-dimethylpropyl, 2,2-dimethylpropyl, 1-ethylpropyl, hexyl, 1-methylpentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl, 1,2-dimethylpropyl, 1,3-dimethylbutyl, 1,4-dimethylbutyl, 2,3-dimethylbutyl, 1,1-dimethylbutyl, 2,2-dimethylbutyl, 3,3-dimethylbutyl, 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethylbutyl and 2-ethylbutyl. Preference is also given to alkyls having 1 to 4 carbon atoms such as, inter alia, methyl, ethyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, s-butyl or t-butyl. The inventive alkyls may be substituted by one or more identical or different radicals.

According to the invention, "alkenyl" – on its own or as part of a chemical group – represents straight-chain or branched hydrocarbons preferably having 2 to 6 carbon atoms and at least one double bond, for example vinyl, 2-propenyl, 2-butenyl, 3-butenyl, 1-methyl-2-propenyl, 2-methyl-2-propenyl, 2-pentenyl, 3-pentenyl, 4-pentenyl, 1-methyl-2-butenyl, 2-methyl-2-butenyl, 3-methyl-2-butenyl, 1-methyl-3-butenyl, 2-methyl-3-butenyl, 3-methyl-3-butenyl, 1,1-dimethyl-2-propenyl, 1,2-dimethyl-2-propenyl, 1-ethyl-2-propenyl, 2-hexenyl, 3-hexenyl, 4-hexenyl, 5-hexenyl, 1-methyl-2-pentenyl, 2-methyl-2-pentenyl, 3-methyl-2-pentenyl, 4-methyl-2-pentenyl, 3-methyl-3-pentenyl, 4-methyl-3-pentenyl, 1-methyl-4-pentenyl, 2-methyl-4-pentenyl, 3-methyl-4-pentenyl, 4-methyl-4-pentenyl, 1,1-dimethyl-2-butenyl, 1,1-dimethyl-3-butenyl, 1,2-dimethyl-2-butenyl, 1,2-dimethyl-3-butenyl, 1,3-dimethyl-2-butenyl, 2,2-dimethyl-3-butenyl, 2,3-dimethyl-2-butenyl, 2,3-dimethyl-3-butenyl, 1-ethyl-2-butenyl, 1-ethyl-3-butenyl, 2-ethyl-2-butenyl, 2-ethyl-3-butenyl, 1,1,2-trimethyl-2-propenyl, 1-ethyl-1-methyl-2-propenyl and 1-ethyl-2-methyl-2-propenyl. Preference is also given to alkenyls having 2 to 4 carbon atoms

such as, inter alia, 2-propenyl, 2-butenyl or 1-methyl-2-propenyl. The inventive alkenyls may be substituted by one or more identical or different radicals.

According to the invention, "alkynyl" – on its own or as part of a chemical group – represents straight-chain or branched hydrocarbons preferably having 2 to 6 carbon atoms and at least one triple bond, for example 2-propynyl, 2-butylnyl, 3-butylnyl, 1-methyl-2-propynyl, 2-pentylnyl, 3-pentylnyl, 4-pentylnyl, 1-methyl-3-butylnyl, 2-methyl-3-butylnyl, 1-methyl-2-butylnyl, 1,1-dimethyl-2-propynyl, 1-ethyl-2-propynyl, 2-hexynyl, 3-hexynyl, 4-hexynyl, 5-hexynyl, 1-methyl-2-pentylnyl, 1-methyl-3-pentylnyl, 1-methyl-4-pentylnyl, 2-methyl-3-pentylnyl, 2-methyl-4-pentylnyl, 3-methyl-4-pentylnyl, 4-methyl-2-pentylnyl, 1,1-dimethyl-3-butylnyl, 1,2-dimethyl-3-butylnyl, 2,2-dimethyl-3-butylnyl, 1-ethyl-3-butylnyl, 2-ethyl-3-butylnyl, 1-ethyl-1-methyl-2-propynyl and 2,5-hexadiynyl. Preference is also given to alkynyls having 2 to 4 carbon atoms such as, inter alia, ethynyl, 2-propynyl or 2-butylnyl-2-propenyl. The inventive alkynyls may be substituted by one or more identical or different radicals.

According to the invention, "cycloalkyl" – on its own or as part of a chemical group – represents mono-, bi- or tricyclic hydrocarbons preferably having 3 to 10 carbons, for example cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl, bicyclo[2.2.1]heptyl, bicyclo[2.2.2]octyl or adamantyl. Preference is also given to cycloalkyls having 3, 4, 5, 6 or 7 carbon atoms such as, inter alia, cyclopropyl or cyclobutyl. The inventive cycloalkyls may be substituted by one or more identical or different radicals.

According to the invention, "alkylcycloalkyl" represents mono-, bi- or tricyclic alkylcycloalkyl preferably having 4 to 10 or 4 to 7 carbon atoms, for example methylcyclopropyl, ethylcyclopropyl, isopropylcyclobutyl, 3-methylcyclopentyl and 4-methylcyclohexyl. Preference is also given to alkylcycloalkyls having 4, 5 or 7 carbon atoms such as, inter alia, ethylcyclopropyl or 4-methylcyclohexyl. The inventive alkylcycloalkyls may be substituted by one or more identical or different radicals.

According to the invention, "cycloalkylalkyl" represents mono-, bi- or tricyclic cycloalkylalkyl preferably having 4 to 10 or 4 to 7 carbon atoms, for example cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl, cyclohexylmethyl and cyclopentylethyl. Preference is also given to cycloalkylalkyls having 4, 5 or 7 carbon atoms such as, inter alia, cyclopropylmethyl or cyclobutylmethyl. The inventive cycloalkylalkyls may be substituted by one or more identical or different radicals.

According to the invention, "hydroxyalkyl" represents a straight-chain or branched alcohol preferably having 1 to 6 carbon atoms, for example methanol, ethanol, n-propanol, isopropanol, n-butanol, isobutanol, s-butanol and t-butanol. Preference is also given to hydroxyalkyl groups having 1 to 4 carbon atoms. The inventive hydroxyalkyl groups may be substituted by one or more identical or different radicals.

According to the invention, "alkoxy" represents a straight-chain or branched O-alkyl preferably having 1 to 6 carbon atoms, for example methoxy, ethoxy, n-propoxy, isopropoxy, n-butoxy, isobutoxy, s-butoxy

and t-butoxy. Preference is also given to alkoxy groups having 1 to 4 carbon atoms. The inventive alkoxy groups may be substituted by one or more identical or different radicals.

According to the invention, "alkylthio", or "alkylsulfanyl" represents straight-chain or branched S-alkyl preferably having 1 to 6 carbon atoms, for example methylthio, ethylthio, n-propylthio, isopropylthio, n-butylthio, isobutylthio, s-butylthio and t-butylthio. Preference is also given to alkylthio groups having 1 to 4 carbon atoms. The inventive alkylthio groups may be substituted by one or more identical or different radicals.

According to the invention, "alkylsulfinyl" represents straight-chain or branched alkylsulfinyl preferably having 1 to 6 carbon atoms, for example methylsulfinyl, ethylsulfinyl, n-propylsulfinyl, isopropylsulfinyl, n-butylsulfinyl, isobutylsulfinyl, s-butylsulfinyl and t-butylsulfinyl. Preference is also given to alkylsulfinyl groups having 1 to 4 carbon atoms. The inventive alkylsulfinyl groups may be substituted by one or more identical or different radicals and embrace both enantiomers.

According to the invention, "alkylsulfonyl" represents straight-chain or branched alkylsulfonyl preferably having 1 to 6 carbon atoms, for example methylsulfonyl, ethylsulfonyl, n-propylsulfonyl, isopropylsulfonyl, n-butylsulfonyl, isobutylsulfonyl, s-butylsulfonyl and t-butylsulfonyl. Preference is also given to alkylsulfonyl groups having 1 to 4 carbon atoms. The inventive alkylsulfonyl groups may be substituted by one or more identical or different radicals.

According to the invention, "cycloalkylthio" or "cycloalkylsulfanyl" represents -S-cycloalkyl preferably having 3 to 6 carbon atoms, for example cyclopropylthio, cyclobutylthio, cyclopentylthio, cyclohexylthio. Preference is also given to cycloalkylthio groups having 3 to 5 carbon atoms. The inventive cycloalkylthio groups may be substituted by one or more identical or different radicals.

According to the invention, "cycloalkylsulfinyl" represents -S(O)-cycloalkyl preferably having 3 to 6 carbon atoms, for example cyclopropylsulfinyl, cyclobutylsulfinyl, cyclopentylsulfinyl, cyclohexylsulfinyl. Preference is also given to cycloalkylsulfinyl groups having 3 to 5 carbon atoms. The inventive cycloalkylsulfinyl groups may be substituted by one or more identical or different radicals and embrace both enantiomers.

According to the invention, "cycloalkylsulfonyl" represents -SO₂-cycloalkyl preferably having 3 to 6 carbon atoms, for example cyclopropylsulfonyl, cyclobutylsulfonyl, cyclopentylsulfonyl, cyclohexylsulfonyl. Preference is also given to cycloalkylsulfonyl groups having 3 to 5 carbon atoms. The inventive cycloalkylsulfonyl groups may be substituted by one or more identical or different radicals.

According to the invention, "phenylthio", or "phenylsulfanyl" represents -S-phenyl, for example phenylthio. The inventive phenylthio groups may be substituted by one or more identical or different radicals.

According to the invention, "phenylsulfinyl" represents -S(O)-phenyl, for example phenylsulfinyl. The inventive phenylsulfinyl groups may be substituted by one or more identical or different radicals and embrace both enantiomers.

According to the invention, "phenylsulfonyl" represents -SO₂-phenyl for example phenylsulfonyl. The
5 inventive phenylsulfonyl groups may be substituted by one or more identical or different radicals.

According to the invention, "alkylcarbonyl" represents straight-chain or branched alkyl-C(=O) preferably having 2 to 7 carbon atoms such as methylcarbonyl, ethylcarbonyl, n-propylcarbonyl, isopropylcarbonyl, s-butylcarbonyl and t-butylcarbonyl. Preference is also given to alkylcarbonyls having 1 to 4 carbon atoms. The inventive alkylcarbonyls may be substituted by one or more identical or different radicals.

10 According to the invention, "alkoxycarbonyl" - alone or as a constituent of a chemical group - represents straight-chain or branched alkoxycarbonyl, preferably having 1 to 6 carbon atoms or having 1 to 4 carbon atoms in the alkoxy moiety, for example methoxycarbonyl, ethoxycarbonyl, n-propoxycarbonyl, isopropoxycarbonyl, s-butoxycarbonyl and t-butoxycarbonyl. The inventive alkoxycarbonyl groups may be substituted by one or more identical or different radicals.

15 According to the invention, "alkylaminocarbonyl" represents straight-chain or branched alkylaminocarbonyl having preferably 1 to 6 carbon atoms or 1 to 4 carbon atoms in the alkyl moiety, for example methylaminocarbonyl, ethylaminocarbonyl, n-propylaminocarbonyl, isopropylaminocarbonyl, s-butylaminocarbonyl and t-butylaminocarbonyl. The inventive alkylaminocarbonyl groups may be substituted by one or more identical or different radicals.

20 According to the invention, "*N,N*-dialkylaminocarbonyl" represents straight-chain or branched *N,N*-dialkylaminocarbonyl having preferably 1 to 6 carbon atoms or 1 to 4 carbon atoms in the alkyl moiety, for example *N,N*-dimethylaminocarbonyl, *N,N*-diethylaminocarbonyl, *N,N*-di(n-propylamino)carbonyl, *N,N*-di(isopropylamino)carbonyl and *N,N*-di(s-butylamino)carbonyl. The inventive *N,N*-dialkylaminocarbonyl groups may be substituted by one or more identical or different radicals.

25 According to the invention, "aryl" represents a mono-, bi- or polycyclic aromatic system having preferably 6 to 14, especially 6 to 10, ring carbon atoms, for example phenyl, naphthyl, anthryl, phenanthrenyl, preferably phenyl. In addition, aryl also represents polycyclic systems such as tetrahydronaphthyl, indenyl, indanyl, fluorenyl, biphenyl, where the bonding site is on the aromatic system. The inventive aryl groups may be substituted by one or more identical or different radicals.

30 Examples of substituted aryls are the arylalkyls, which may likewise be substituted by one or more identical or different radicals in the C₁-C₄alkyl and/or C₆-C₁₄aryl moiety. Examples of such arylalkyls include benzyl and phenyl-1-ethyl.

According to the invention, "heterocycle", "heterocyclic ring" or "heterocyclic ring system" represents a carbocyclic ring system having at least one ring in which at least one carbon atom is replaced by a heteroatom, preferably by a heteroatom from the group consisting of N, O, S, P, B, Si, Se, and which is saturated, unsaturated or heteroaromatic and may be unsubstituted or substituted, where the bonding site
5 is on a ring atom. Unless defined differently, the heterocyclic ring contains preferably 3 to 9 ring atoms, especially 3 to 6 ring atoms, and one or more, preferably 1 to 4, especially 1, 2 or 3, heteroatoms in the heterocyclic ring, preferably from the group consisting of N, O, and S, although no two oxygen atoms should be directly adjacent. The heterocyclic rings usually contain not more than 4 nitrogen atoms and/or not more than 2 oxygen atoms and/or not more than 2 sulphur atoms. When the heterocyclyl radical or the
10 heterocyclic ring is optionally substituted, it may be fused to other carbocyclic or heterocyclic rings. In the case of optionally substituted heterocyclyl, the invention also embraces polycyclic systems, for example 8-azabicyclo[3.2.1]octanyl or 1-azabicyclo[2.2.1]heptyl. In the case of optionally substituted heterocyclyl, the invention also embraces spirocyclic systems, for example 1-oxa-5-azaspiro[2.3]hexyl.

Inventive heterocyclyl groups are, for example, piperidinyl, piperazinyl, morpholinyl, thiomorpholinyl,
15 dihydropyranyl, tetrahydropyranyl, dioxanyl, pyrrolinyl, pyrrolidinyl, imidazolynyl, imidazolidinyl, thiazolidinyl, oxazolidinyl, dioxolanyl, dioxolyl, pyrazolidinyl, tetrahydrofuranyl, dihydrofuranyl, oxetanyl, oxiranyl, azetidynyl, aziridinyl, oxazetidynyl, oxaziridinyl, oxazepanyl, oxazinanyl, azepanyl, oxopyrrolidinyl, dioxopyrrolidinyl, oxomorpholinyl, oxopiperazinyl and oxepanyl.

Of particular significance are heteroaryls, i.e. heteroaromatic systems. According to the invention, the
20 term heteroaryl represents heteroaromatic compounds, i.e. completely unsaturated aromatic heterocyclic compounds which fall under the above definition of heterocycles. Preference is given to 5- to 7-membered rings having 1 to 3, preferably 1 or 2, identical or different heteroatoms from the group above. Inventive heteroaryls are, for example, furyl, thienyl, pyrazolyl, imidazolyl, 1,2,3- and 1,2,4-triazolyl, isoxazolyl, thiazolyl, isothiazolyl, 1,2,3-, 1,3,4-, 1,2,4- and 1,2,5-oxadiazolyl, azepinyl, pyrrolyl, pyridyl, pyridazinyl,
25 pyrimidinyl, pyrazinyl, 1,3,5-, 1,2,4- and 1,2,3-triazinyl, 1,2,4-, 1,3,2-, 1,3,6- and 1,2,6-oxazinyl, oxepinyl, thiopinyl, 1,2,4-triazolonyl and 1,2,4-diazepinyl. The inventive heteroaryl groups may also be substituted by one or more identical or different radicals.

The term "in each case optionally substituted" means that a group/substituent, such as a alkyl, alkenyl, alkynyl, alkoxy, alkylthio, alkylsulfinyl, alkylsulfonyl, cycloalkyl, aryl, phenyl, benzyl, heterocyclyl and
30 heteroaryl radical, is substituted, meaning, for example, a substituted radical derived from the unsubstituted base structure, where the substituents, for example, one (1) substituent or a plurality of substituents, preferably 1, 2, 3, 4, 5, 6 or 7, are selected from a group consisting of amino, hydroxyl, halogen, nitro, cyano, isocyano, mercapto, isothiocyanato, C₁-C₄carboxyl, carbonamide, SF₅, aminosulphonyl, C₁-C₄alkyl, C₁-C₄haloalkyl, C₃-C₄cycloalkyl, C₂-C₄alkenyl, C₅-C₆cycloalkenyl, C₂-
35 C₄alkynyl, *N*-mono-C₁-C₄alkylamino, *N,N*-di-C₁-C₄alkylamino, *N*-C₁-C₄alkanoylamino, C₁-C₄alkoxy, C₁-C₄haloalkoxy, C₂-C₄alkenyloxy, C₂-C₄alkynyloxy, C₃-C₄cycloalkoxy, C₅-C₆cycloalkenyloxy, C₁-

C₄alkoxycarbonyl, C₂-C₄alkenyloxycarbonyl, C₂-C₄alkynyloxycarbonyl, C₆-,C₁₀-,C₁₄-aryloxycarbonyl, C₁-C₄alkanoyl, C₂-C₄alkenylcarbonyl, C₂-C₄alkynylcarbonyl, C₆-,C₁₀-,C₁₄-arylcarbonyl, C₁-C₄alkylthio, C₁-C₄haloalkylthio, C₃-C₄cycloalkylthio, C₂-C₄alkenylthio, C₅-C₆cycloalkenylthio, C₂-C₄alkynylthio, C₁-C₄alkylsulfinyl, including both enantiomers of the C₁-C₄alkylsulfinyl group, C₁-C₄haloalkylsulfinyl, including both enantiomers of the C₁-C₄haloalkylsulfinyl group, C₁-C₄alkylsulfonyl, C₁-C₄haloalkylsulfonyl, *N*-mono-C₁-C₄alkylaminosulfonyl, *N,N*-di-C₁-C₄alkylaminosulfonyl, C₁-C₄alkylphosphinyl, C₁-C₄alkylphosphonyl, including both enantiomers of C₁-C₄alkylphosphinyl and C₁-C₄alkylphosphonyl, *N*-C₁-C₄alkylaminocarbonyl, *N,N*-di-C₁-C₄alkylaminocarbonyl, *N*-C₁-C₄alkanoylaminocarbonyl, *N*-C₁-C₄alkanoyl-*N*-C₁-C₄alkylaminocarbonyl, C₆-,C₁₀-,C₁₄-aryl, C₆-,C₁₀-,C₁₄-aryloxy, benzyl, benzyloxy, benzylthio, C₆-,C₁₀-,C₁₄-arylthio, C₆-,C₁₀-,C₁₄-arylamino, benzylamino, heterocyclyl and trialkylsilyl, substituents bonded via a double bond, such as C₁-C₄alkylidene (e.g. methylenidene or ethylenidene), an oxo group, an imino group and a substituted imino group. When two or more radicals form one or more rings, these may be carbocyclic, heterocyclic, saturated, partly saturated, unsaturated, for example including aromatic rings and with further substitution. The substituents mentioned by way of example ("first substituent level") may, if they contain hydrocarbonaceous components, optionally have further substitution therein ("second substituent level"), for example by one or more of the substituents each independently selected from halogen, hydroxyl, amino, nitro, cyano, isocyano, azido, acylamino, an oxo group and an imino group. The term "(optionally) substituted" group preferably embraces just one or two substituent levels.

The inventive halogen-substituted chemical groups or halogenated groups (for example alkyl or alkoxy) are mono- or polysubstituted by halogen up to the maximum possible number of substituents. Such groups are also referred to as halo groups (for example haloalkyl). In the case of polysubstitution by halogen, the halogen atoms may be the same or different, and may all be bonded to one carbon atom or may be bonded to a plurality of carbon atoms. Halogen is especially fluorine, chlorine, bromine or iodine, preferably fluorine, chlorine or bromine and more preferably fluorine. More particularly, halogen-substituted groups are monohalocycloalkyl such as 1-fluorocyclopropyl, 2-fluorocyclopropyl or 1-fluorocyclobutyl, monohaloalkyl such as 2-chloroethyl, 2-fluoroethyl, 1-chloroethyl, 1-fluoroethyl, chloromethyl, or fluoromethyl; perhaloalkyl such as trichloromethyl or trifluoromethyl or CF₂CF₃, polyhaloalkyl such as difluoromethyl, 2-fluoro-2-chloroethyl, dichloromethyl, 1,1,2,2-tetrafluoroethyl or 2,2,2-trifluoroethyl.

Further examples of haloalkyls are trichloromethyl, chlorodifluoromethyl, dichlorofluoromethyl, chloromethyl, bromomethyl, 1-fluoroethyl, 2-fluoroethyl, 2,2-difluoroethyl, 2,2,2-trifluoroethyl, 2,2,2-trichloroethyl, 2-chloro-2,2-difluoroethyl, pentafluoroethyl, 3,3,3-trifluoropropyl and pentafluoro-*t*-butyl. Preference is given to haloalkyls having 1 to 4 carbon atoms and 1 to 9, preferably 1 to 5, identical or different halogen atoms selected from fluorine, chlorine and bromine. Particular preference is given to haloalkyls having 1 or 2 carbon atoms and 1 to 5 identical or different halogen atoms selected from fluorine and chlorine, such as, inter alia, difluoromethyl, trifluoromethyl or 2,2-difluoroethyl. Further examples of halogen-substituted compounds are haloalkoxy such as OCF₃, OCHF₂, OCH₂F, OCF₂CF₃, OCH₂CF₃, OCH₂CHF₂ und OCH₂CH₂Cl, haloalkylsulfanyls such as difluoromethylthio, trifluoromethylthio,

trichloromethylthio, chlorodifluoromethylthio, 1-fluoroethylthio, 2-fluoroethylthio, 2,2-difluoroethylthio, 1,1,2,2-tetrafluoroethylthio, 2,2,2-trifluoroethylthio or 2-chloro-1,1,2-trifluoroethylthio, haloalkylsulfinyls such as difluoromethylsulfinyl, trifluoromethylsulfinyl, trichloromethylsulfinyl, chlorodifluoromethylsulfinyl, 1-fluoroethylsulfinyl, 2-fluoroethylsulfinyl, 2,2-difluoroethylsulfinyl, 5 1,1,2,2-tetrafluoroethylsulfinyl, 2,2,2-trifluoroethylsulfinyl and 2-chloro-1,1,2-trifluoroethylsulfinyl, haloalkylsulfinyls such as difluoromethylsulfinyl, trifluoromethylsulfinyl, trichloromethylsulfinyl, chlorodifluoromethylsulfinyl, 1-fluoroethylsulfinyl, 2-fluoroethylsulfinyl, 2,2-difluoroethylsulfinyl, 1,1,2,2-tetrafluoroethylsulfinyl, 2,2,2-trifluoroethylsulfinyl and 2-chloro-1,1,2-trifluoroethylsulfinyl, haloalkylsulfonyl groups such as difluoromethylsulfonyl, trifluoromethylsulfonyl, 10 trichloromethylsulfonyl, chlorodifluoromethylsulfonyl, 1-fluoroethylsulfonyl, 2-fluoroethylsulfonyl, 2,2-difluoroethylsulfonyl, 1,1,2,2-tetrafluoroethylsulfonyl, 2,2,2-trifluoroethylsulfonyl and 2-chloro-1,1,2-trifluoroethylsulfonyl.

In the case of radicals having carbon atoms, preference is given to those having 1 to 4 carbon atoms, especially 1 or 2 carbon atoms. Preference is generally given to substituents from the group of halogen, 15 e.g. fluorine and chlorine, (C₁-C₄)alkyl, preferably methyl or ethyl, (C₁-C₄)haloalkyl, preferably trifluoromethyl, (C₁-C₄)alkoxy, preferably methoxy or ethoxy, (C₁-C₄)haloalkoxy, nitro and cyano. Particular preference is given here to the substituents methyl, methoxy, fluorine and chlorine.

Substituted amino such as mono- or disubstituted amino means a radical from the group of the substituted amino radicals which are *N*-substituted, for example, by one or two identical or different radicals from the 20 group of alkyl, hydroxy, amino, alkoxy, acyl and aryl; preferably *N*-mono- and *N,N*-dialkylamino, (for example methylamino, ethylamino, *N,N*-dimethylamino, *N,N*-diethylamino, *N,N*-di-*n*-propylamino, *N,N*-diisopropylamino or *N,N*-dibutylamino), *N*-mono- or *N,N*-dialkoxyalkylamino groups (for example *N*-methoxymethylamino, *N*-methoxyethylamino, *N,N*-di(methoxymethyl)amino or *N,N*-di(methoxyethyl)amino), *N*-mono- and *N,N*-diarylamino, such as optionally substituted anilines, 25 acylamino, *N,N*-diacylamino, *N*-alkyl-*N*-arylamino, *N*-alkyl-*N*-acylamino and also saturated *N*-heterocycles; preference is given here to alkyl radicals having 1 to 4 carbon atoms; here, aryl is preferably phenyl or substituted phenyl; for acyl, the definition given further below applies, preferably (C₁-C₄)-alkanoyl. The same applies to substituted hydroxylamino or hydrazino.

Substituted amino also includes quaternary ammonium compounds (salts) having four organic substituents 30 on the nitrogen atom.

Optionally substituted phenyl is preferably phenyl which is unsubstituted or mono- or polysubstituted, preferably up to trisubstituted, by identical or different radicals from the group of halogen, (C₁-C₄)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)alkoxy-(C₁-C₄)alkoxy, (C₁-C₄)alkoxy-(C₁-C₄)alkyl, (C₁-C₄)haloalkyl, (C₁-C₄)haloalkoxy, (C₁-C₄)alkylthio, (C₁-C₄)haloalkylthio, (C₁-C₄)alkylsulfinyl (C₁-C₄) haloalkylsulfinyl, 35 (C₁-C₄)alkylsulfonyl (C₁-C₄)haloalkylsulfonyl, cyano, isocyano and nitro, for example *o*-, *m*- and *p*-tolyl, dimethylphenyls, 2-, 3- and 4-chlorophenyl, 2-, 3- and 4-fluorophenyl, 2-, 3- and 4-trifluoromethyl- and

4-trichloromethylphenyl, 2,4-, 3,5-, 2,5- and 2,3-dichlorophenyl, o-, m- and p-methoxyphenyl, 4-heptafluorophenyl.

Optionally substituted cycloalkyl is preferably cycloalkyl which is unsubstituted or mono- or polysubstituted, preferably up to trisubstituted, by identical or different radicals from the group of halogen, cyano, (C₁-C₄)alkyl, (C₁-C₄)alkoxy, (C₁-C₄)alkoxy-(C₁-C₄)alkoxy, (C₁-C₄)alkoxy-(C₁-C₄)alkyl, (C₁-C₄)haloalkyl and (C₁-C₄)haloalkoxy, especially by one or two (C₁-C₄)alkyl radicals.

Inventive compounds may occur in preferred embodiments. Individual embodiments described herein may be combined with one another. Not included are combinations which contravene the laws of nature and which the person skilled in the art would therefore rule out on the basis of his/her expert knowledge. Ring structures having three or more adjacent oxygen atoms, for example, are excluded.

Isomers

Depending on the nature of the substituents, the compounds of the formula (I) may be in the form of geometric and/or optically active isomers or corresponding isomer mixtures in different compositions. These stereoisomers are, for example, enantiomers, diastereomers, atropisomers or geometric isomers. Accordingly, the invention encompasses both pure stereoisomers and any mixture of these isomers.

Methods and uses

The invention also relates to methods for controlling animal pests, in which compounds of the formula (I) are allowed to act on animal pests and/or their habitat. The control of the animal pests is preferably conducted in agriculture and forestry, and in material protection. Preferably excluded herefrom are methods for the surgical or therapeutic treatment of the human or animal body and diagnostic methods carried out on the human or animal body.

The invention furthermore relates to the use of the compounds of the formula (I) as pesticides, in particular crop protection agents.

In the context of the present application, the term "pesticide" in each case also always comprises the term "crop protection agent".

The compounds of the formula (I), having good plant tolerance, favourable homeotherm toxicity and good environmental compatibility, are suitable for protecting plants and plant organs against biotic and abiotic stressors, for increasing harvest yields, for improving the quality of the harvested material and for controlling animal pests, especially insects, arachnids, helminths, in particular nematodes, and molluscs, which are encountered in agriculture, in horticulture, in animal husbandry, in aquatic cultures, in forests, in gardens and leisure facilities, in the protection of stored products and of materials, and in the hygiene sector.

Within the context of the present patent application, the term “hygiene” is understood to mean any and all measures, procedures and practices which aim to prevent disease, in particular infectious disease, and which serve to protect the health of humans and animals and/or to protect the environment, and/or which maintain cleanliness. In accordance with the invention, this especially includes measures for cleaning, disinfection and sterilisation of, for example, textiles or hard surfaces, especially surfaces of glass, wood, concrete, porcelain, ceramics, plastic or also of metal(s), and for ensuring that these are kept free of hygiene pests and/or their excretions. Preferably excluded from the scope of the invention in this regard are surgical or therapeutic treatment procedures applicable to the human body or to the bodies of animals and diagnostic procedures which are carried out on the human body or on the bodies of animals.

- 10 The term “hygiene sector” thus covers all areas, technical fields and industrial applications in which these hygiene measures, procedures and practices are important, in relation for example to hygiene in kitchens, bakeries, airports, bathrooms, swimming pools, department stores, hotels, hospitals, stables, animal husbandries, etc.

- 15 The term “hygiene pest” is therefore understood to mean one or more animal pests whose presence in the hygiene sector is problematic, in particular for health reasons. It is therefore a primary objective to avoid or minimize the presence of hygiene pests, and/or exposure to them, in the hygiene sector. This can be achieved in particular through the application of a pesticide that can be used both to prevent infestation and to tackle an infestation which is already present. Preparations which avoid or reduce exposure to pests can also be used. Hygiene pests include, for example, the organisms mentioned below.

- 20 The term “hygiene protection” thus covers all actions to maintain and/or improve these hygiene measures, procedures and practices.

The compounds of the formula (I) can preferably be used as pesticides. They are active against normally sensitive and resistant species and against all or some stages of development. The abovementioned pests include:

- 25 pests from the phylum of the Arthropoda, in particular from the class of the Arachnida, for example *Acarus* spp., for example *Acarus siro*, *Aceria kuko*, *Aceria sheldoni*, *Aculops* spp., *Aculus* spp., for example *Aculus fockeui*, *Aculus schlechtendali*, *Amblyomma* spp., *Amphitetranychus viennensis*, *Argas* spp., *Boophilus* spp., *Brevipalpus* spp., for example *Brevipalpus phoenicis*, *Bryobia graminum*, *Bryobia praetiosa*, *Centruroides* spp., *Chorioptes* spp., *Dermanyssus gallinae*, *Dermatophagoides pteronyssinus*,
30 *Dermatophagoides farinae*, *Dermacentor* spp., *Eotetranychus* spp., for example *Eotetranychus hicoriae*, *Epitrimerus pyri*, *Eutetranychus* spp., for example *Eutetranychus banksi*, *Eriophyes* spp., for example *Eriophyes pyri*, *Glycyphagus domesticus*, *Halotydeus destructor*, *Hemitarsonemus* spp., for example *Hemitarsonemus latus* (=Polyphagotarsonemus latus), *Hyalomma* spp., *Ixodes* spp., *Latrodectus* spp., *Loxosceles* spp., *Neutrombicula autumnalis*, *Nuphessa* spp., *Oligonychus* spp., for example *Oligonychus*
35 *coffae*, *Oligonychus coniferarum*, *Oligonychus ilicis*, *Oligonychus indicus*, *Oligonychus mangiferus*,

- Oligonychus pratensis, Oligonychus punicae, Oligonychus yothersi, Ornithodorus spp., Ornithonyssus spp., Panonychus spp., for example Panonychus citri (=Metatetranychus citri), Panonychus ulmi (=Metatetranychus ulmi), Phyllocoptruta oleivora, Platytetranychus multidigituli, Polyphagotarsonemus latus, Psoroptes spp., Rhipicephalus spp., Rhizoglyphus spp., Sarcoptes spp., Scorpio maurus,
- 5 Steneotarsonemus spp., Steneotarsonemus spinki, Tarsonemus spp., for example Tarsonemus confusus, Tarsonemus pallidus, Tetranychus spp., for example Tetranychus canadensis, Tetranychus cinnabarinus, Tetranychus turkestani, Tetranychus urticae, Trombicula alfreddugesi, Vaejovis spp., Vasates lycopersici;
- from the class of the Chilopoda, for example Geophilus spp., Scutigera spp.;
- from the order or the class of the Collembola, for example Onychiurus armatus; Sminthurus viridis;
- 10 from the class of the Diplopoda, for example Blaniulus guttulatus;
- from the class of the Insecta, for example from the order of the Blattodea, for example Blatta orientalis, Blattella asahinai, Blattella germanica, Leucophaea maderae, Loboptera decipiens, Neostylopyga rhombifolia, Panchlora spp., Parcoblatta spp., Periplaneta spp., for example Periplaneta americana, Periplaneta australasiae, Pycnoscelus surinamensis, Supella longipalpa;
- 15 from the order of the Coleoptera, for example Acalymma vittatum, Acanthoscelides obtectus, Adoretus spp., Aethina tumida, Agelastica alni, Agrilus spp., for example Agrilus planipennis, Agrilus coxalis, Agrilus bilineatus, Agrilus anxius, Agriotes spp., for example Agriotes linneatus, Agriotes mancus, Agriotes obscurus, Alphitobius diaperinus, Amphimallon solstitialis, Anobium punctatum, Anomala dubia, Anoplophora spp., for example Anoplophora glabripennis, Anthonomus spp., for example
- 20 Anthonomus grandis, Anthrenus spp., Apion spp., Apogonia spp., Athous haemorrhoidales, Atomaria spp., for example Atomaria linearis, Attagenus spp., Baris caerulescens, Bruchidius obtectus, Bruchus spp., for example Bruchus pisorum, Bruchus rufimanus, Cassida spp., Cerotoma trifurcata, Ceutorrhynchus spp., for example Ceutorrhynchus assimilis, Ceutorrhynchus quadridens, Ceutorrhynchus rapae, Chaetocnema spp., for example Chaetocnema confinis, Chaetocnema denticulata, Chaetocnema
- 25 ectypa, Cleonus mendicus, Conoderus spp., Cosmopolites spp., for example Cosmopolites sordidus, Costelytra zealandica, Ctenicera spp., Curculio spp., for example Curculio caryae, Curculio caryatrypes, Curculio obtusus, Curculio sayi, Cryptolestes ferrugineus, Cryptolestes pusillus, Cryptorhynchus lapathi, Cryptorhynchus mangiferae, Cyllindrocopturus spp., Cyllindrocopturus adspersus, Cyllindrocopturus furnissi, Dendroctonus spp., for example Dendroctonus ponderosae, Dermestes spp.,
- 30 Diabrotica spp., for example Diabrotica balteata, Diabrotica barberi, Diabrotica undecimpunctata howardi, Diabrotica undecimpunctata undecimpunctata, Diabrotica virgifera virgifera, Diabrotica virgifera zea, Dichrocrocis spp., Dieladisa armigera, Diloboderus spp., Epicaerus spp., Epilachna spp., for example Epilachna borealis, Epilachna varivestis, Epitrix spp., for example Epitrix cucumeris, Epitrix fuscua, Epitrix hirtipennis, Epitrix subcrinita, Epitrix tuberis, Faustinus spp., Gibbium psylloides, Gnathocerus
- 35 cornutus, Hellula undalis, Heteronychus arator, Heteronyx spp., Hoplia argentea, Hylamorphia elegans,

Hylotrupes bajulus, Hypera postica, Hypomeces squamosus, Hypothenemus spp., for example Hypothenemus hampei, Hypothenemus obscurus, Hypothenemus pubescens, Lachnosterna consanguinea, Lasioderma serricorne, Latheticus oryzae, Lathridius spp., Lema spp., Leptinotarsa decemlineata, Leucoptera spp., for example Leucoptera coffeella, Limonius ectypus, Lissorhoptrus oryzophilus, 5 Lissorhoptrus (= Hyperodes) spp., Lixus spp., Luperodes spp., Luperomorpha xanthodera, Lyctus spp., Megacyllene spp., for example Megacyllene robiniae, Megascelis spp., Melanotus spp., for example Melanotus longulus oregonensis, Meligethes aeneus, Melolontha spp., for example Melolontha melolontha, Migdolus spp., Monochamus spp., Naupactus xanthographus, Necrobia spp., Neogalerucella spp., Niptus hololeucus, Oryctes rhinoceros, Oryzaephilus surinamensis, Oryzaphagus oryzae, 10 Otiorhynchus spp., for example Otiorhynchus cribricollis, Otiorhynchus ligustici, Otiorhynchus ovatus, Otiorhynchus rugosostriatus, Otiorhynchus sulcatus, Oulema spp., for example Oulema melanopus, Oulema oryzae, Oxycetonia jucunda, Phaedon cochleariae, Phyllophaga spp., Phyllophaga helleri, Phyllotreta spp., for example Phyllotreta armoraciae, Phyllotreta pusilla, Phyllotreta ramosa, Phyllotreta striolata, Popillia japonica, Premnotypes spp., Prostephanus truncatus, Psylliodes spp., for example 15 Psylliodes affinis, Psylliodes chrysocephala, Psylliodes punctulata, Ptinus spp., Rhizobius ventralis, Rhizophorthera dominica, Rhynchophorus spp., Rhynchophorus ferrugineus, Rhynchophorus palmarum, Scolytus spp., for example Scolytus multistriatus, Sinoxylon perforans, Sitophilus spp., for example Sitophilus granarius, Sitophilus linearis, Sitophilus oryzae, Sitophilus zeamais, Sphenophorus spp., Stegobium paniceum, Sternechus spp., for example Sternechus paludatus, Symphyletes spp., Tanyemecus spp., for example Tanyemecus dilaticollis, Tanyemecus indicus, Tanyemecus palliatus, Tenebrio molitor, 20 Tenebrioides mauretanicus, Tribolium spp., for example Tribolium audax, Tribolium castaneum, Tribolium confusum, Trogoderma spp., Tychius spp., Xylotrechus spp., Zabrus spp., for example Zabrus tenebrioides;

from the order of the Dermaptera, for example Anisolabis maritime, Forficula auricularia, Labidura 25 riparia;

from the order of the Diptera, for example Aedes spp., for example Aedes aegypti, Aedes albopictus, Aedes sticticus, Aedes vexans, Agromyza spp., for example Agromyza frontella, Agromyza parvicornis, Anastrepha spp., Anopheles spp., for example Anopheles quadrimaculatus, Anopheles gambiae, Asphondylia spp., Bactrocera spp., for example Bactrocera cucurbitae, Bactrocera dorsalis, Bactrocera 30 oleae, Bibio hortulanus, Calliphora erythrocephala, Calliphora vicina, Ceratitis capitata, Chironomus spp., Chrysomya spp., Chrysops spp., Chrysosona pluvialis, Cochliomya spp., Contarinia spp., for example Contarinia johnsoni, Contarinia nasturtii, Contarinia pyrivora, Contarinia schulzi, Contarinia sorghicola, Contarinia tritici, Cordylobia anthropophaga, Cricotopus sylvestris, Culex spp., for example Culex pipiens, Culex quinquefasciatus, Culicoides spp., Culiseta spp., Cuterebra spp., Dacus oleae, Dasineura spp., for 35 example Dasineura brassicae, Delia spp., for example Delia antiqua, Delia coarctata, Delia florilega, Delia platura, Delia radicum, Dermatobia hominis, Drosophila spp., for example Drosophila melanogaster, Drosophila suzukii, Echinocnemeus spp., Euleia heraclei, Fannia spp., Gasterophilus spp., Glossina spp.,

- Haematopota spp., Hydrellia spp., Hydrellia griseola, Hylemya spp., Hippobosca spp., Hypoderma spp., Liriomyza spp., for example Liriomyza brassicae, Liriomyza huidobrensis, Liriomyza sativae, Lucilia spp., for example Lucilia cuprina, Lutzomyia spp., Mansonia spp., Musca spp., for example Musca domestica, Musca domestica vicina, Oestrus spp., Oscinella frit, Paratanytarsus spp., Paralauterborniella subcincta, Pegomya or Pegomyia spp., for example Pegomya betae, Pegomya hyoscyami, Pegomya rubivora, Phlebotomus spp., Phorbia spp., Phormia spp., Piophilidae casei, Platyparea poeciloptera, Prodiptosis spp., Psila rosae, Rhagoletis spp., for example Rhagoletis cingulata, Rhagoletis completa, Rhagoletis fausta, Rhagoletis indifferens, Rhagoletis mendax, Rhagoletis pomonella, Sarcophaga spp., Simulium spp., for example Simulium meridionale, Stomoxys spp., Tabanus spp., Tetanops spp., Tipula spp., for example Tipula paludosa, Tipula simplex, Toxotrypana curvicauda;
- from the order of the Hemiptera, for example Acizzia acaciaebaileyanae, Acizzia dodonaeae, Acizzia uncatoides, Acrida turrita, Acyrthosiphon spp., for example Acyrthosiphon pisum, Acrogonia spp., Aeneolamia spp., Agonosceles spp., Aleurocanthus spp., Aleyrodes proletella, Aleurolobus barodensis, Aleurothrixus floccosus, Allocaridara malayensis, Amrasca spp., for example Amrasca biguttula, Amrasca devastans, Anuraphis cardui, Aonidiella spp., for example Aonidiella aurantii, Aonidiella citrina, Aonidiella inornata, Aphanostigma piri, Aphis spp., for example Aphis citricola, Aphis craccivora, Aphis fabae, Aphis forbesi, Aphis glycines, Aphis gossypii, Aphis hederae, Aphis illinoisensis, Aphis middletoni, Aphis nasturtii, Aphis nerii, Aphis pomi, Aphis spiraeicola, Aphis viburniphila, Arboridia apicalis, Arytainilla spp., Aspidiella spp., Aspidiotus spp., for example Aspidiotus nerii, Atanus spp., Aulacorthum solani, Bemisia tabaci, Blastopsylla occidentalis, Boreioglycaspis melaleuciae, Brachycaudus helichrysi, Brachycolus spp., Brevicoryne brassicae, Cacopsylla spp., for example Cacopsylla pyricola, Calligypona marginata, Capulinia spp., Carneiocephala fulgida, Ceratovacuna lanigera, Cercopidae, Ceroplastes spp., Chaetosiphon fragaefolii, Chionaspis tegalensis, Chlorita onukii, Chondracis rosea, Chromaphis juglandicola, Chrysomphalus aonidum, Chrysomphalus ficus, Cicadulina mbila, Cocomytilus halli, Coccus spp., for example Coccus hesperidum, Coccus longulus, Coccus pseudomagnoliarum, Coccus viridis, Cryptomyzus ribis, Cryptoneossa spp., Ctenarytaina spp., Dalbulus spp., Dialeurodes chittendeni, Dialeurodes citri, Diaphorina citri, Diaspis spp., Diuraphis spp., Doralis spp., Drosicha spp., Dysaphis spp., for example Dysaphis apiifolia, Dysaphis plantaginea, Dysaphis tulipae, Dysmicoccus spp., Empoasca spp., for example Empoasca abrupta, Empoasca fabae, Empoasca maligna, Empoasca solana, Empoasca stevensi, Eriosoma spp., for example Eriosoma americanum, Eriosoma lanigerum, Eriosoma pyricola, Erythroneura spp., Eucalyptolyma spp., Euphyllura spp., Euscelis bilobatus, Ferrisia spp., Fiorinia spp., Furcaspis oceanica, Geococcus coffeae, Glycaspis spp., Heteropsylla cubana, Heteropsylla spinulosa, Homalodisca coagulata, Hyalopterus arundinis, Hyalopterus pruni, Icerya spp., for example Icerya purchasi, Idiocerus spp., Idioscopus spp., Laodelphax striatellus, Lecanium spp., for example Lecanium corni (=Parthenolecanium corni), Lepidosaphes spp., for example Lepidosaphes ulmi, Lipaphis erysimi, Lopholeucaspis japonica, Lycorma delicatula, Macrosiphum spp., for example Macrosiphum euphorbiae, Macrosiphum lillii, Macrosiphum rosae, Macrosteles facifrons, Mahanarva spp., Melanaphis sacchari, Metcalfiella spp., Metcalfa pruinosa, Metopolophium dirhodum,

- Monellia costalis, Monelliopsis pecanis, Myzus spp., for example Myzus ascalonicus, Myzus cerasi, Myzus ligustri, Myzus ornatus, Myzus persicae, Myzus nicotianae, Nasonovia ribisnigri, Neomaskellia spp., Nephrotettix spp., for example Nephrotettix cincticeps, Nephrotettix nigropictus, Nettigoniella spectra, Nilaparvata lugens, Oncometopia spp., Orthezia praelonga, Oxya chinensis, Pachypsylla spp.,
- 5 Parabemisia myricae, Paratrioza spp., for example Paratrioza cockerelli, Parlatoria spp., Pemphigus spp., for example Pemphigus bursarius, Pemphigus populivenerae, Peregrinus maidis, Perkinsiella spp., Phenacoccus spp., for example Phenacoccus madeirensis, Phloeomyzus passerinii, Phorodon humuli, Phylloxera spp., for example Phylloxera devastatrix, Phylloxera notabilis, Pinaspis aspidistrae, Planococcus spp., for example Planococcus citri, Prosopidopsylla flava, Protopulvinaria pyriformis,
- 10 Pseudaulacaspis pentagona, Pseudococcus spp., for example Pseudococcus calceolariae, Pseudococcus comstocki, Pseudococcus longispinus, Pseudococcus maritimus, Pseudococcus viburni, Psyllopsis spp., Psylla spp., for example Psylla buxi, Psylla mali, Psylla pyri, Pteromalus spp., Pulvinaria spp., Pyrrilla spp., Quadraspidiotus spp., for example Quadraspidiotus juglansregiae, Quadraspidiotus ostreaeformis, Quadraspidiotus perniciosus, Quesada gigas, Rastrococcus spp., Rhopalosiphum spp., for example
- 15 Rhopalosiphum maidis, Rhopalosiphum oxyacanthae, Rhopalosiphum padi, Rhopalosiphum rufiabdominale, Saissetia spp., for example Saissetia coffeae, Saissetia miranda, Saissetia neglecta, Saissetia oleae, Scaphoideus titanus, Schizaphis graminum, Selenaspidus articulatus, Siphia flava, Sitobion avenae, Sogatella furcifera, Sogatodes spp., Stictocephala festina, Siphoninus phillyreae, Tenalaphara malayensis, Tetragnathella spp., Tinocallis caryaefoliae, Tomaspis spp., Toxoptera spp.,
- 20 for example Toxoptera aurantii, Toxoptera citricidus, Trialeurodes vaporariorum, Trioza spp., for example Trioza diospyri, Typhlocyba spp., Unaspis spp., Viteus vitifolii, Zygina spp.;

- from the suborder of the Heteroptera, for example Aelia spp., Anasa tristis, Antestiopsis spp., Boisea spp., Blissus spp., Calocoris spp., Campylomma livida, Cavalerius spp., Cimex spp., for example Cimex adjunctus, Cimex hemipterus, Cimex lectularius, Cimex pilosellus, Collaria spp., Creontiades dilutus,
- 25 Dasynus piperis, Dichelops furcatus, Diconocoris hewetti, Dysdercus spp., Euschistus spp., for example Euschistus heros, Euschistus servus, Euschistus tristigmus, Euschistus variolarius, Eurydema spp., Eurygaster spp., Halyomorpha halys, Heliopeltis spp., Horcias nobilellus, Leptocoris spp., Leptocoris varicornis, Leptoglossus occidentalis, Leptoglossus phyllopus, Lygocoris spp., for example Lygocoris pabulinus, Lygus spp., for example Lygus elisus, Lygus hesperus, Lygus lineolaris, Macropes excavatus,
- 30 Megacopta cribraria, Miridae, Monalonion atratum, Nezara spp., for example Nezara viridula, Nysius spp., Oebalus spp., Pentomidae, Piesma quadrata, Piezodorus spp., for example Piezodorus guildinii, Psallus spp., Pseudacysta persea, Rhodnius spp., Sahlbergella singularis, Scaevola castanea, Scotinophara spp., Stephanitis nashi, Tibraca spp., Triatoma spp.;

- from the order of the Hymenoptera, for example Acromyrmex spp., Athalia spp., for example Athalia rosae, Atta spp., Camponotus spp., Dolichovespula spp., Diprion spp., for example Diprion similis, Hoplocampa spp., for example Hoplocampa cookei, Hoplocampa testudinea, Lasius spp., Linepithema (Iridomyrmex) humile, Monomorium pharaonis, Paratrechina spp., Paravespula spp., Plagiolepis spp.,
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Sirex spp., for example *Sirex noctilio*, *Solenopsis invicta*, *Tapinoma* spp., *Technomyrmex albipes*, *Urocerus* spp., *Vespa* spp., for example *Vespa crabro*, *Wasmannia auropunctata*, *Xeris* spp.;

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

from the order of the Isoptera, for example *Coptotermes* spp., for example *Coptotermes formosanus*,
5 *Cornitermes cumulans*, *Cryptotermes* spp., *Incisitermes* spp., *Kaloterms* spp., *Microtermes obesi*,
Nasutitermes spp., *Odontotermes* spp., *Porotermes* spp., *Reticulitermes* spp., for example *Reticulitermes flavipes*, *Reticulitermes hesperus*;

from the order of the Lepidoptera, for example *Achroia grisella*, *Acronicta major*, *Adoxophyes* spp., for
example *Adoxophyes orana*, *Aedia leucomelas*, *Agrotis* spp., for example *Agrotis segetum*, *Agrotis*
10 *ipsilon*, *Alabama* spp., for example *Alabama argillacea*, *Amyeloides transitella*, *Anarsia* spp., *Anticarsia*
spp., for example *Anticarsia gemmatilis*, *Argyroplote* spp., *Autographa* spp., *Barathra brassicae*,
Blastodacna atra, *Borbo cinnara*, *Bucculatrix thurberiella*, *Bupalus piniarius*, *Busseola* spp., *Cacoecia* spp.,
Caloptilia theivora, *Capua reticulana*, *Carpocapsa pomonella*, *Carposina niponensis*, *Cheimatobia*
brumata, *Chilo* spp., for example *Chilo plejadellus*, *Chilo suppressalis*, *Choreutis pariana*, *Choristoneura*
15 spp., *Chrysodeixis chalcites*, *Clysia ambiguella*, *Cnaphalocerus* spp., *Cnaphalocrocis medinalis*,
Cnephasia spp., *Conopomorpha* spp., *Conotrachelus* spp., *Copitarsia* spp., *Cydia* spp., for example *Cydia*
nigricana, *Cydia pomonella*, *Dalaca noctuides*, *Diaphania* spp., *Diparopsis* spp., *Diatraea saccharalis*,
Dioryctria spp., for example *Dioryctria zimmermani*, *Earias* spp., *Ecdytoplopha aurantium*, *Elasmopalpus*
lignosellus, *Eldana saccharina*, *Ephestia* spp., for example *Ephestia elutella*, *Ephestia kuehniella*, *Epinotia*
20 spp., *Epiphyas postvittana*, *Erannis* spp., *Erschoviella musculana*, *Etiella* spp., *Eudocima* spp., *Eulia* spp.,
Eupoecilia ambiguella, *Euproctis* spp., for example *Euproctis chrysorrhoea*, *Euxoa* spp., *Feltia* spp.,
Galleria mellonella, *Gracillaria* spp., *Grapholitha* spp., for example *Grapholitha molesta*, *Grapholitha*
prunivora, *Hedylepta* spp., *Helicoverpa* spp., for example *Helicoverpa armigera*, *Helicoverpa zea*,
Heliopsis spp., for example *Heliopsis virescens*, *Hepialus* spp., for example *Hepialus humuli*,
25 *Hofmannophila pseudospretella*, *Homoeosoma* spp., *Homona* spp., *Hyponomeuta padella*, *Kakivoria*
flavofasciata, *Lampides* spp., *Laphygma* spp., *Laspeyresia molesta*, *Leucinodes orbonalis*, *Leucophaea*
spp., for example *Leucophaea coffeella*, *Lithocolletis* spp., for example *Lithocolletis blancardella*,
Lithophane antennata, *Lobesia* spp., for example *Lobesia botrana*, *Loxagrotis albicosta*, *Lymantria* spp.,
for example *Lymantria dispar*, *Lyonetia* spp., for example *Lyonetia clerkella*, *Malacosoma neustria*,
30 *Maruca testulalis*, *Mamestra brassicae*, *Melanitis leda*, *Mocis* spp., *Monopis obviella*, *Mythimna separata*,
Nemapogon cloacellus, *Nymphula* spp., *Oiketicus* spp., *Omphisa* spp., *Operophtera* spp., *Oria* spp.,
Orthaga spp., *Ostrinia* spp., for example *Ostrinia nubilalis*, *Panolis flammea*, *Parnara* spp., *Pectinophora*
spp., for example *Pectinophora gossypiella*, *Perileucoptera* spp., *Phthorimaea* spp., for example
Phthorimaea operculella, *Phyllocnistis citrella*, *Phyllonorycter* spp., for example *Phyllonorycter*
35 *blancardella*, *Phyllonorycter crataegella*, *Pieris* spp., for example *Pieris rapae*, *Platynota stultana*, *Plodia*
interpunctella, *Plusia* spp., *Plutella xylostella* (= *Plutella maculipennis*), *Podesia* spp., for example *Podesia*

- syringae, Prays spp., Prodenia spp., Protoparce spp., Pseudaletia spp., for example Pseudaletia unipuncta, Pseudoplusia includens, Pyrausta nubilalis, Rachiplusia nu, Schoenobius spp., for example Schoenobius bipunctifer, Scirpophaga spp., for example Scirpophaga innotata, Scotia segetum, Sesamia spp., for example Sesamia inferens, Sparganothis spp., Spodoptera spp., for example Spodoptera eradiana,
- 5 Spodoptera exigua, Spodoptera frugiperda, Spodoptera praefica, Stathmopoda spp., Stenoma spp., Stomopteryx subsecivella, Synanthedon spp., Tecia solanivora, Thaumetopoea spp., Thermesia gemmatilis, Tinea cloacella, Tinea pellionella, Tineola bisselliella, Tortrix spp., Trichophaga tapetzella, Trichoplusia spp., for example Trichoplusia ni, Tryporyza incertulas, Tuta absoluta, Virachola spp.;
- from the order of the Orthoptera or Saltatoria, for example Acheta domesticus, Dichroplus spp.,
- 10 Gryllotalpa spp., for example Gryllotalpa gryllotalpa, Hieroglyphus spp., Locusta spp., for example Locusta migratoria, Melanoplus spp., for example Melanoplus devastator, Paratlanticus ussuriensis, Schistocerca gregaria;
- from the order of the Phthiraptera, for example Damalinia spp., Haematopinus spp., Linognathus spp., Pediculus spp., Phylloxera vastatrix, Phthirus pubis, Trichodectes spp.;
- 15 from the order of the Psocoptera, for example Lepinotus spp., Liposcelis spp.;
- from the order of the Siphonaptera, for example, Ceratophyllus spp., Ctenocephalides spp., for example Ctenocephalides canis, Ctenocephalides felis, Pulex irritans, Tunga penetrans, Xenopsylla cheopis;
- from the order of the Thysanoptera, for example Anaphothrips obscurus, Baliothrips biformis, Chaetanaphothrips leeuweni, Drepanothrips reuteri, Enneothrips flavens, Frankliniella spp., for example
- 20 Frankliniella fusca, Frankliniella occidentalis, Frankliniella schultzei, Frankliniella tritici, Frankliniella vaccinii, Frankliniella williamsi, Haplothrips spp., Heliothrips spp., Hercinothrips femoralis, Kakothrips spp., Rhipiphorothrips cruentatus, Scirtothrips spp., Taeniothrips cardamomi, Thrips spp., for example Thrips palmi, Thrips tabaci;
- from the order of the Zygentoma (= Thysanura), for example Ctenolepisma spp., Lepisma saccharina,
- 25 Lepismodes inquilinus, Thermobia domestica;
- from the class of the Symphyla, for example Scutigera spp., for example Scutigera immaculata;
- pests from the phylum of the Mollusca, for example from the class of the Bivalvia, for example Dreissena spp.,
- and also from the class of the Gastropoda, for example Arion spp., for example Arion ater rufus,
- 30 Biomphalaria spp., Bulinus spp., Deroceras spp., for example Deroceras laeve, Galba spp., Lymnaea spp., Oncomelania spp., Pomacea spp., Succinea spp.;

plant pests from the phylum of the Nematoda, i.e. phytoparasitic nematodes, in particular *Aglenchus* spp., for example *Aglenchus agricola*, *Anguina* spp., for example *Anguina tritici*, *Aphelenchoides* spp., for example *Aphelenchoides arachidis*, *Aphelenchoides fragariae*, *Belonolaimus* spp., for example *Belonolaimus gracilis*, *Belonolaimus longicaudatus*, *Belonolaimus nortoni*, *Bursaphelenchus* spp., for example *Bursaphelenchus cocophilus*, *Bursaphelenchus eremus*, *Bursaphelenchus xylophilus*, *Cacopaurus* spp., for example *Cacopaurus pestis*, *Criconemella* spp., for example *Criconemella curvata*, *Criconemella onoensis*, *Criconemella ornata*, *Criconemella rusium*, *Criconemella xenoplax* (= *Mesocriconema xenoplax*), *Criconemoides* spp., for example *Criconemoides ferniae*, *Criconemoides onoense*, *Criconemoides ornatum*, *Ditylenchus* spp., for example *Ditylenchus dipsaci*, *Dolichodorus* spp., *Globodera* spp., for example *Globodera pallida*, *Globodera rostochiensis*, *Helicotylenchus* spp., for example *Helicotylenchus dihystra*, *Hemicriconemoides* spp., *Hemicycliophora* spp., *Heterodera* spp., for example *Heterodera avenae*, *Heterodera glycines*, *Heterodera schachtii*, *Hirschmaniella* spp., *Hoplolaimus* spp., *Longidorus* spp., for example *Longidorus africanus*, *Meloidogyne* spp., for example *Meloidogyne chitwoodi*, *Meloidogyne fallax*, *Meloidogyne hapla*, *Meloidogyne incognita*, *Meloinema* spp., *Nacobbus* spp., *Neotylenchus* spp., *Paralongidorus* spp., *Paraphelenchus* spp., *Paratrichodorus* spp., for example *Paratrichodorus minor*, *Paratylenchus* spp., *Pratylenchus* spp., for example *Pratylenchus penetrans*, *Pseudohalenchus* spp., *Psilenchus* spp., *Punctodera* spp., *Quinisulcius* spp., *Radopholus* spp., for example *Radopholus citrophilus*, *Radopholus similis*, *Rotylenchulus* spp., *Rotylenchus* spp., *Scutellonema* spp., *Subanguina* spp., *Trichodorus* spp., for example *Trichodorus obtusus*, *Trichodorus primitivus*, *Tylenchorhynchus* spp., for example *Tylenchorhynchus annulatus*, *Tylenchulus* spp., for example *Tylenchulus semipenetrans*, *Xiphinema* spp., for example *Xiphinema index*.

The compounds of the formula (I) can optionally, at certain concentrations or application rates, also be used as herbicides, safeners, growth regulators or agents to improve plant properties, as microbicides or gametocides, for example as fungicides, antimycotics, bactericides, viricides (including agents against viroids) or as agents against MLO (mycoplasma-like organisms) and RLO (rickettsia-like organisms). If appropriate, they can also be used as intermediates or precursors for the synthesis of other active compounds.

Formulations/Use forms

The present invention further relates to formulations, in particular formulations for controlling unwanted controlling animal pests. The formulation may be applied to the animal pest and/or in their habitat.

The formulation of the invention may be provided to the end user as “ready-for-use” use form, i.e. the formulations may be directly applied to the plants or seeds by a suitable device, such as a spraying or dusting device. Alternatively, the formulations may be provided to the end user in the form of concentrates which have to be diluted, preferably with water, prior to use. Unless otherwise indicated, the wording “formulation” therefore means such concentrate, whereas the wording “use form” means the end user as “ready-for-use” solution, i.e. usually such diluted formulation.

The formulation of the invention can be prepared in conventional manners, for example by mixing the compound of the invention with one or more suitable auxiliaries, such as disclosed herein.

The formulation comprises at least one compound of the invention and at least one agriculturally suitable auxiliary, e.g. carrier(s) and/or surfactant(s).

- 5 A carrier is a solid or liquid, natural or synthetic, organic or inorganic substance that is generally inert. The carrier generally improves the application of the compounds, for instance, to plants, plants parts or seeds. Examples of suitable solid carriers include, but are not limited to, ammonium salts, in particular ammonium sulfates, ammonium phosphates and ammonium nitrates, natural rock flours, such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite and diatomaceous earth, silica gel and synthetic
- 10 rock flours, such as finely divided silica, alumina and silicates. Examples of typically useful solid carriers for preparing granules include, but are not limited to crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, synthetic granules of inorganic and organic flours and granules of organic material such as paper, sawdust, coconut shells, maize cobs and tobacco stalks. Examples of suitable liquid carriers include, but are not limited to, water, organic solvents and combinations thereof.
- 15 Examples of suitable solvents include polar and nonpolar organic chemical liquids, for example from the classes of aromatic and nonaromatic hydrocarbons (such as cyclohexane, paraffins, alkylbenzenes, xylene, toluene, tetrahydronaphthalene, alkylnaphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride), alcohols and polyols (which may optionally also be substituted, etherified and/or esterified, such as ethanol, propanol, butanol,
- 20 benzylalcohol, cyclohexanol or glycol), ketones (such as acetone, methyl ethyl ketone, methyl isobutyl ketone, acetophenone, or cyclohexanone), esters (including fats and oils) and (poly)ethers, unsubstituted and substituted amines, amides (such as dimethylformamide or fatty acid amides) and esters thereof, lactams (such as N-alkylpyrrolidones, in particular N-methylpyrrolidone) and lactones, sulfones and sulfoxides (such as dimethyl sulfoxide), oils of vegetable or animal origin, nitriles (alkyl nitriles such as acetonitrile, propionitrile, butyronitrile, or aromatic nitriles, such as benzonitrile), carbonic acid esters
- 25 (cyclic carbonic acid esters, such as ethylene carbonate, propylene carbonate, butylene carbonate, or dialkyl carbonic acid esters, such as dimethyl carbonate, diethyl carbonate, dipropyl carbonate, dibutyl carbonate, dioctyl carbonate). The carrier may also be a liquefied gaseous extender, i.e. liquid which is gaseous at standard temperature and under standard pressure, for example aerosol propellants such as
- 30 halohydrocarbons, butane, propane, nitrogen and carbon dioxide.

Preferred solid carriers are selected from clays, talc and silica.

Preferred liquid carriers are selected from water, fatty acid amides and esters thereof, aromatic and nonaromatic hydrocarbons, lactams, lactones, carbonic acid esters, ketones, (poly)ethers.

- The amount of carrier typically ranges from 1 to 99.99%, preferably from 5 to 99.9%, more preferably
- 35 from 10 to 99.5%, and most preferably from 20 to 99% by weight of the formulation.

Liquid carriers are typically present in a range of from 20 to 90%, for example 30 to 80% by weight of the formulation.

Solid carriers are typically present in a range of from 0 to 50%, preferably 5 to 45%, for example 10 to 30% by weight of the formulation.

- 5 If the formulation comprises two or more carriers, the outlined ranges refer to the total amount of carriers.

The surfactant can be an ionic (cationic or anionic), amphoteric or non-ionic surfactant, such as ionic or non-ionic emulsifier(s), foam former(s), dispersant(s), wetting agent(s), penetration enhancer(s) and any mixtures thereof. Examples of suitable surfactants include, but are not limited to, salts of polyacrylic acid, ethoxylated poly(α -substituted)acrylate derivatives, salts of lignosulfonic acid (such as sodium
10 lignosulfonate), salts of phenolsulfonic acid or naphthalenesulfonic acid, polycondensates of ethylene oxide and/or propylene oxide with or without alcohols, fatty acids or fatty amines (for example, polyoxyethylene fatty acid esters such as castor oil ethoxylate, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers), substituted phenols (preferably alkylphenols or arylphenols), salts of sulfosuccinic esters, taurine derivatives (preferably alkyl taurates), phosphoric esters of polyethoxylated
15 alcohols or phenols, fatty esters of polyols (such as fatty acid esters of glycerol, sorbitol or sucrose), sulfates (such as alkyl sulfates and alkyl ether sulfates), sulfonates (for example, alkylsulfonates, arylsulfonates and alkylbenzene sulfonates), sulfonated polymers of naphthalene/formaldehyde, phosphate esters, protein hydrolysates, lignosulfite waste liquors and methylcellulose. Any reference to salts in this paragraph refers preferably to the respective alkali, alkaline earth and ammonium salts.

- 20 Preferred surfactants are selected from ethoxylated poly(α -substituted)acrylate derivatives, polycondensates of ethylene oxide and/or propylene oxide with alcohols, polyoxyethylene fatty acid esters, alkylbenzene sulfonates, sulfonated polymers of naphthalene/formaldehyde, polyoxyethylene fatty acid esters such as castor oil ethoxylate, sodium lignosulfonate and arylphenol ethoxylate.

The amount of surfactants typically ranges from 5 to 40%, for example 10 to 20%, by weight of the
25 formulation.

Further examples of suitable auxiliaries include water repellents, siccatives, binders (adhesive, tackifier, fixing agent, such as carboxymethylcellulose, natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, natural phospholipids such as cephalins and lecithins and synthetic phospholipids, polyvinylpyrrolidone and tylose), thickeners
30 and secondary thickeners (such as cellulose ethers, acrylic acid derivatives, xanthan gum, modified clays, e.g. the products available under the name Bentone, and finely divided silica), stabilizers (e.g. cold stabilizers, preservatives (e.g. dichlorophene, benzyl alcohol hemiformal, 1,2-Benzisothiazolin-3-on, 2-methyl-4-isothiazolin-3-one), antioxidants, light stabilizers, in particular UV stabilizers, or other agents which improve chemical and/or physical stability), dyes or pigments (such as inorganic pigments, e.g.
35 iron oxide, titanium oxide and Prussian Blue; organic dyes, e.g. alizarin, azo and metal phthalocyanine

dyes), antifoams (e.g. silicone antifoams and magnesium stearate), antifreezes, stickers, gibberellins and processing auxiliaries, mineral and vegetable oils, perfumes, waxes, nutrients (including trace nutrients, such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc), protective colloids, thixotropic substances, penetrants, sequestering agents and complex formers.

- 5 The choice of the auxiliaries depends on the intended mode of application of the compound of the invention and/or on the physical properties of the compound(s). Furthermore, the auxiliaries may be chosen to impart particular properties (technical, physical and/or biological properties) to the formulations or use forms prepared therefrom. The choice of auxiliaries may allow customizing the formulations to specific needs.
- 10 The formulation comprises an insecticidal/acaricidal/nematicidal effective amount of the compound(s) of the invention. The term "effective amount" denotes an amount, which is sufficient for controlling harmful insects/mites/nematodes on cultivated plants or in the protection of materials and which does not result in a substantial damage to the treated plants. Such an amount can vary in a broad range and is dependent on various factors, such as the insect/mite/nematode species to be controlled, the treated cultivated plant or
- 15 material, the climatic conditions and the specific compound of the invention used. Usually, the formulation according to the invention contains from 0.01 to 99% by weight, preferably from 0.05 to 98% by weight, more preferred from 0.1 to 95% by weight, even more preferably from 0.5 to 90% by weight, most preferably from 1 to 80% by weight of the compound of the invention. It is possible that a formulation comprises two or more compounds of the invention. In such case the outlined ranges refer to the total
- 20 amount of compounds of the present invention.

- The formulation of the invention may be in any customary formulation type, such as solutions (e.g. aqueous solutions), emulsions, water- and oil-based suspensions, powders (e.g. wettable powders, soluble powders), dusts, pastes, granules (e.g. soluble granules, granules for broadcasting), suspoemulsion concentrates, natural or synthetic products impregnated with the compound of the invention, fertilizers
- 25 and also microencapsulations in polymeric substances. The compound of the invention may be present in a suspended, emulsified or dissolved form. Examples of particular suitable formulation types are solutions, watersoluble concentrates (e.g. SL, LS), dispersible concentrates (DC), suspensions and suspension concentrates (e.g. SC, OD, OF, FS), emulsifiable concentrates (e.g. EC), emulsions (e.g. EW, EO, ES, ME, SE), capsules (e.g. CS, ZC), pastes, pastilles, wettable powders or dusts (e.g. WP, SP, WS, DP, DS),
- 30 pressings (e.g. BR, TB, DT), granules (e.g. WG, SG, GR, FG, GG, MG), insecticidal articles (e.g. LN), as well as gel formulations for the treatment of plant propagation materials such as seeds (e.g. GW, GF). These and further formulations types are defined by the Food and Agriculture Organization of the United Nations (FAO). An overview is given in the "Catalogue of pesticide formulation types and international coding system", Technical Monograph No. 2, 6th Ed. May 2008, Croplife International.
- 35 Preferably, the formulation of the invention is in form of one of the following types: EC, SC, FS, SE, OD, WG, WP, CS, more preferred EC, SC, OD, WG, CS.

Further details about examples of formulation types and their preparation are given below. If two or more compounds of the invention are present, the outlined amount of compound of the invention refers to the total amount of compounds of the present invention. This applies mutatis mutandis for any further component of the formulation, if two or more representatives of such component, e.g. wetting agent, binder, are present.

i) Water-soluble concentrates (SL, LS)

10-60 % by weight of at least one compound of the invention and 5-15 % by weight surfactant (e.g. polycondensates of ethylene oxide and/or propylene oxide with alcohols) are dissolved in such amount of water and/or water-soluble solvent (e.g. alcohols such as propylene glycol or carbonates such as propylene carbonate) to result in a total amount of 100 % by weight. Before application the concentrate is diluted with water.

ii) Dispersible concentrates (DC)

5-25 % by weight of at least one compound of the invention and 1-10 % by weight surfactant and/or binder (e.g. polyvinylpyrrolidone) are dissolved in such amount of organic solvent (e.g. cyclohexanone) to result in a total amount of 100 % by weight. Dilution with water gives a dispersion.

iii) Emulsifiable concentrates (EC)

15-70 % by weight of at least one compound of the invention and 5-10 % by weight surfactant (e.g. a mixture of calcium dodecylbenzenesulfonate and castor oil ethoxylate) are dissolved in such amount of water-insoluble organic solvent (e.g. aromatic hydrocarbon or fatty acid amide) and if needed additional water-soluble solvent to result in a total amount of 100 % by weight. Dilution with water gives an emulsion.

iv) Emulsions (EW, EO, ES)

5-40 % by weight of at least one compound of the invention and 1-10 % by weight surfactant (e.g. a mixture of calcium dodecylbenzenesulfonate and castor oil ethoxylate, or polycondensates of ethylene oxide and/or propylene oxide with or without alcohols) are dissolved in 20-40 % by weight water-insoluble organic solvent (e.g. aromatic hydrocarbon). This mixture is added to such amount of water by means of an emulsifying machine to result in a total amount of 100 % by weight. The resulting formulation is a homogeneous emulsion. Before application the emulsion may be further diluted with water.

v) Suspensions and suspension concentrates

v-1) Water-based (SC, FS)

In a suitable grinding equipment, e.g. an agitated ball mill, 20-60 % by weight of at least one compound of the invention are comminuted with addition of 2-10 % by weight surfactant (e.g. sodium lignosulfonate and polyoxyethylene fatty alcohol ether), 0.1-2 % by weight thickener (e.g. xanthan gum) and water to give a fine active substance suspension. The water is added in such amount to result in a total amount of 100 % by weight. Dilution with water gives a stable suspension of the active substance. For FS type formulations up to 40 % by weight binder (e.g. polyvinylalcohol) is added.

v-2) Oil-based(OD, OF)

In a suitable grinding equipment, e.g. an agitated ball mill, 20-60 % by weight of at least one compound of the invention are comminuted with addition of 2-10 % by weight surfactant (e.g. sodium lignosulfonate and polyoxyethylene fatty alcohol ether), 0.1-2 % by weight thickener (e.g. modified clay, in particular Bentone, or silica) and an organic carrier to give a fine active substance oil suspension. The organic carrier is added in such amount to result in a total amount of 100 % by weight. Dilution with water gives a stable dispersion of the active substance.

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

1-90 % by weight, preferably 20-80%, most preferably 50-80 % by weight of at least one compound of the invention are ground finely with addition of surfactant (e.g. sodium lignosulfonate and sodium alkylnaphthylsulfonates) and potentially carrier material and converted to water-dispersible or water-soluble granules by means of typical technical appliances like e. g. extrusion, spray drying, fluidized bed granulation. The surfactant and carrier material is used in such amount to result in a total amount of 100 % by weight. Dilution with water gives a stable dispersion or solution of the active substance.

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

50-80 % by weight of at least one compound of the invention are ground in a rotor-stator mill with addition of 1-20 % by weight surfactant (e.g. sodium lignosulfonate, sodium alkylnaphthylsulfonates) and such amount of solid carrier, e.g. silica gel, to result in a total amount of 100 % by weight. Dilution with water gives a stable dispersion or solution of the active substance.

viii) Gel (GW, GF)

In an agitated ball mill, 5-25 % by weight of at least one compound of the invention are comminuted with addition of 3-10 % by weight surfactant (e.g. sodium lignosulfonate), 1-5 % by weight binder (e.g. carboxymethylcellulose) and such amount of water to result in a total amount of 100 % by weight. This results in a fine suspension of the active substance. Dilution with water gives a stable suspension of the active substance.

ix) Microemulsion (ME)

5-20 % by weight of at least one compound of the invention are added to 5-30 % by weight organic solvent blend (e.g. fatty acid dimethylamide and cyclohexanone), 10-25 % by weight surfactant blend (e.g. polyoxyethylene fatty alcohol ether and arylphenol ethoxylate), and such amount of water to result in a total amount of 100 % by weight. This mixture is stirred for 1 h to produce spontaneously a thermodynamically stable microemulsion.

x) Microcapsules (CS)

An oil phase comprising 5-50 % by weight of at least one compound of the invention, 0-40 % by weight water-insoluble organic solvent (e.g. aromatic hydrocarbon), 2-15 % by weight acrylic monomers (e.g. methylmethacrylate, methacrylic acid and a di- or triacrylate) are dispersed into an aqueous solution of a protective colloid (e.g. polyvinyl alcohol). Radical polymerization initiated by a radical initiator results in the formation of poly(meth)acrylate microcapsules. Alternatively, an oil phase comprising 5-50 % by weight of at least one compound of the invention, 0-40 % by weight water-insoluble organic solvent (e.g. aromatic hydrocarbon), and an isocyanate monomer (e.g. diphenylmethene-4,4'-diisocyanate) are dispersed into an aqueous solution of a protective colloid (e.g. polyvinyl alcohol), this resulting in the formation of polyurea microcapsules. Optionally, the addition of a polyamine (e.g. hexamethylenediamine) is also used to result in the formation of polyurea microcapsules. The monomers amount to 1-10 % by weight of the total CS formulation.

xi) Dustable powders (DP, DS)

1-10 % by weight of at least one compound of the invention are ground finely and mixed intimately with such amount of solid carrier, e.g. finely divided kaolin, to result in a total amount of 100 % by weight.

xii) Granules (GR, FG)

0.5-30 % by weight of at least one compound of the invention are ground finely and associated with such amount of solid carrier (e.g. silicate) to result in a total amount of 100 % by weight.

xiii) Ultra-low volume liquids (UL)

1-50 % by weight of at least one compound of the invention are dissolved in such amount of organic solvent, e.g. aromatic hydrocarbon, to result in a total amount of 100 % by weight.

The formulations types i) to xiii) may optionally comprise further auxiliaries, such as 0.1-1 % by weight preservatives, 0.1-1 % by weight antifoams, 0.1-1 % by weight dyes and/or pigments, and 5-10% by weight antifreezes.

Mixtures

The compounds of the formula (I) may also be employed as a mixture with one or more suitable fungicides, bactericides, acaricides, molluscicides, nematocides, insecticides, microbiologicals, beneficial species, herbicides, fertilizers, bird repellents, phytotonics, sterilants, safeners, semiochemicals and/or plant growth regulators, in order thus, for example, to broaden the spectrum of action, to prolong the duration of action, to increase the rate of action, to prevent repulsion or prevent evolution of resistance. In addition, such active compound combinations may improve plant growth and/or tolerance to abiotic factors, for example high or low temperatures, to drought or to elevated water content or soil salinity. It is also possible to improve flowering and fruiting performance, optimize germination capacity and root development, facilitate harvesting and improve yields, influence maturation, improve the quality and/or the nutritional value of the harvested products, prolong storage life and/or improve the processability of the harvested products.

Furthermore, the compounds of the formula (I) can be present in a mixture with other active compounds or semiochemicals such as attractants and/or bird repellants and/or plant activators and/or growth regulators and/or fertilizers. Likewise, the compounds of the formula (I) can be used to improve plant properties such as, for example, growth, yield and quality of the harvested material.

In a particular embodiment according to the invention, the compounds of the formula (I) are present in formulations or the use forms prepared from these formulations in a mixture with further compounds, preferably those as described below.

If one of the compounds mentioned below can occur in different tautomeric forms, these forms are also included even if not explicitly mentioned in each case. Further, all named mixing partners can, if their functional groups enable this, optionally form salts with suitable bases or acids.

Insecticides/acaricides/nematicides

The active compounds identified here by their common names are known and are described, for example, in the pesticide handbook ("The Pesticide Manual" 16th Ed., British Crop Protection Council 2012) or can be found on the Internet (e.g. <http://www.alanwood.net/pesticides>). The classification is based on the current IRAC Mode of Action Classification Scheme at the time of filing of this patent application.

(1) Acetylcholinesterase (AChE) inhibitors, preferably carbamates selected from alanycarb, aldicarb, bendiocarb, benfuracarb, butocarboxim, butoxycarboxim, carbaryl, carbofuran, carbosulfan, ethiofencarb, fenobucarb, formetanate, furathiocarb, isoprocarb, methiocarb, methomyl, metolcarb, oxamyl, pirimicarb, propoxur, thiodicarb, thiofanox, triazamate, trimethacarb, XMC and xyllycarb, or organophosphates selected from acephate, azamethiphos, azinphos-ethyl, azinphos-methyl, cadusafos, chlorethoxyfos, chlorfenvinphos, chlormephos, chlorpyrifos-methyl, coumaphos, cyanophos, demeton-S-methyl, diazinon, dichlorvos/DDVP, dicotophos, dimethoate, dimethylvinphos, disulfoton, EPN, ethion,

- ethoprophos, famphur, fenamiphos, fenitrothion, fenthion, fosthiazate, heptenophos, imicyafos, isofenphos, isopropyl O-(methoxyaminothiophosphoryl) salicylate, isoxathion, malathion, mecarbam, methamidophos, methidathion, mevinphos, monocrotophos, naled, omethoate, oxydemeton-methyl, parathion-methyl, phenthoate, phorate, phosalone, phosmet, phosphamidon, phoxim, pirimiphos-methyl, profenofos, propetamphos, prothiofos, pyraclofos, pyridaphenthion, quinalphos, sulfotep, tebupirimfos, temephos, terbufos, tetrachlorvinphos, thiometon, triazophos, triclofon and vamidothion.
- (2) GABA-gated chloride channel blockers, preferably cyclodiene-organochlorines selected from chlordane and endosulfan, or phenylpyrazoles (fiproles) selected from ethiprole and fipronil.
- (3) Sodium channel modulators, preferably pyrethroids selected from acrinathrin, allethrin, d-cis-trans allethrin, d-trans allethrin, bifenthrin, bioallethrin, bioallethrin s-cyclopentenyl isomer, bioresmethrin, cycloprothrin, cyfluthrin, beta-cyfluthrin, cyhalothrin, lambda-cyhalothrin, gamma-cyhalothrin, cypermethrin, alpha-cypermethrin, beta-cypermethrin, theta-cypermethrin, zeta-cypermethrin, cyphenothrin [(1R)-trans-isomer], deltamethrin, emperthrin [(EZ)-(1R)-isomer], esfenvalerate, etofenprox, fenpropathrin, fenvalerate, flucythrinate, flumethrin, tau-fluvalinate, halfenprox, imiprothrin, kadethrin, momfluorothrin, permethrin, phenothrin [(1R)-trans-isomer], prallethrin, pyrethrins (pyrethrum), resmethrin, silafluofen, tefluthrin, tetramethrin, tetramethrin [(1R)- isomer)], tralomethrin and transfluthrin, or DDT or methoxychlor.
- (4) Nicotinic acetylcholine receptor (nAChR) competitive modulators, preferably neonicotinoids selected from acetamiprid, clothianidin, dinotefuran, imidacloprid, nitenpyram, thiacloprid and thiamethoxam, or nicotine, or sulfoximines selected from sulfoxaflor, or butenolids selected from flupyradifurone, or mesoionics selected from triflumezopyrim.
- (5) Nicotinic acetylcholine receptor (nAChR) allosteric modulators (Site I), preferably spinosyns selected from spinetoram and spinosad.
- (6) Glutamate-gated chloride channel (GluCl) allosteric modulators, preferably avermectins/milbemycins selected from abamectin, emamectin benzoate, lepimectin and milbemectin.
- (7) Juvenile hormone mimics, preferably juvenile hormone analogues selected from hydroprene, kinoprene and methoprene, or fenoxycarb or pyriproxyfen.
- (8) Miscellaneous non-specific (multi-site) inhibitors, preferably alkyl halides selected from methyl bromide and other alkyl halides, or chloropicrine or sulphuryl fluoride or borax or tartar emetic or methyl isocyanate generators selected from diazomet and metam.
- (9) Chordotonal organ TRPV channel modulators, preferably pyridine azomethanes selected from pymetrozine and pyrifluquinazone, or pyropenes selected from afidopyropen.

- (10) Mite growth inhibitors affecting CHS1 selected from clofentezine, hexythiazox, diflovidazin and etoxazole.
- (11) Microbial disruptors of the insect gut membranes selected from *Bacillus thuringiensis* subspecies *israelensis*, *Bacillus sphaericus*, *Bacillus thuringiensis* subspecies *aizawai*, *Bacillus thuringiensis* subspecies *kurstaki*, *Bacillus thuringiensis* subspecies *tenebrionis*, and *B.t.* plant proteins selected from Cry1Ab, Cry1Ac, Cry1Fa, Cry1A.105, Cry2Ab, Vip3A, mCry3A, Cry3Ab, Cry3Bb and Cry34Ab1/35Ab1.
- (12) Inhibitors of mitochondrial ATP synthase, preferably ATP disruptors selected from diafenthiuron, or organotin compounds selected from azocyclotin, cyhexatin and fenbutatin oxide, or propargite or tetradifon.
- (13) Uncouplers of oxidative phosphorylation via disruption of the proton gradient selected from chlorfenapyr, DNOC and sulfluramid.
- (14) Nicotinic acetylcholine receptor channel blockers selected from bensultap, cartap hydrochloride, thiocylam and thiosultap-sodium.
- (15) Inhibitors of chitin biosynthesis affecting CHS1, preferably benzoylureas selected from bistrifluron, chlorfluazuron, diflubenzuron, flucycloxuron, flufenoxuron, hexaflumuron, lufenuron, novaluron, noviflumuron, teflubenzuron and triflumuron.
- (16) Inhibitors of chitin biosynthesis, type 1 selected from buprofezin.
- (17) Moulting disruptor (in particular for Diptera, i.e. dipterans) selected from cyromazine.
- (18) Ecdysone receptor agonists, preferably diacylhydrazines selected from chromafenozide, halofenozide, methoxyfenozide and tebufenozide.
- (19) Octopamine receptor agonists selected from amitraz.
- (20) Mitochondrial complex III electron transport inhibitors selected from hydramethylnone, acequinocyl, fluacrypyrim and bifenazate.
- (21) Mitochondrial complex I electron transport inhibitors, preferably METI acaricides and insecticides selected from fenazaquin, fenpyroximate, pyrimidifen, pyridaben, tebufenpyrad and tolfenpyrad, or rotenone (Derris).
- (22) Voltage-dependent sodium channel blockers, preferably oxadiazines selected from indoxacarb, or semicarbazones selected from metaflumizone.

- (23) Inhibitors of acetyl CoA carboxylase, preferably tetronic and tetramic acid derivatives selected from spirodiclofen, spiromesifen, spiropidion and spirotetramat.
- (24) Mitochondrial complex IV electron transport inhibitors, preferably phosphides selected from aluminium phosphide, calcium phosphide, phosphine and zinc phosphide, or cyanides selected from calcium cyanide, potassium cyanide and sodium cyanide.
- (25) Mitochondrial complex II electron transport inhibitors, preferably *beta*-ketonitrile derivatives selected from cyenopyrafen and cyflumetofen, or carboxanilides selected from pyflubumide.
- (28) Ryanodine receptor modulators, preferably diamides selected from chlorantraniliprole, cyantraniliprole, cyclaniliprole, flubendiamide and tetraniliprole.
- (29) Chordotonal organ Modulators (with undefined target site) selected from flonicamid.
- (30) GABA-gated chlorid channel allosteric modulators, preferably *meta*-diamides selected from broflanilide, or isoxazoles selected from fluxametamide.
- (31) Baculoviruses, preferably Granuloviruses (GVs) selected from *Cydia pomonella* GV and *Thaumotobia leucotreta* (GV), or Nucleopolyhedroviruses (NPVs) selected from *Anticarsia gemmatilis* MNPV and *Helicoverpa armigera* NPV.
- (32) Nicotinic acetylcholine receptor allosteric modulators (Site II) selected from GS-omega/kappa HXTX-Hv1a peptide.
- (33) further active compounds selected from Acynonapyr, Afoxolaner, Azadirachtin, Benclothiaz, Benzoximate, Benzpyrimoxan, Bromopropylate, Chinomethionat, Chloroprallethrin, Cryolite, Cyclobutrifluram, Cycloxaprid, Cyetpyrafen, Cyhalodiamide, Cyproflanilide (CAS 2375110-88-4), Dicloromezotiaz, Dicofol, Dimpropyridaz, epsilon-Metofluthrin, epsilon-Momfluthrin, Flometoquin, Fluazaindoline, Flucypriprole (CAS 1771741-86-6), Fluensulfone, Flufenerim, Flufenoxystrobin, Flufiprole, Fluhexafon, Fluopyram, Flupyrimin, Fluralaner, Fufenozide, Flupentiofenox, Guadipyr, Heptafluthrin, Imidaclothiz, Iprodione, Isocycloseram, kappa-Bifenthrin, kappa-Tefluthrin, Lotilaner, Meperfluthrin, Nicofluprole (CAS 1771741-86-6), Oxazosulphyl, Paichongding, Pyridalyl, Pyrifluquinazon, Pyriminostrobin, Sarolaner, Spidoxamat, Spirobudiclofen, Tetramethylfluthrin, Tetrachlorantraniliprole, Tigolaner, Tioxazafen, Thiofluoximate, Tyclopyrazoflor, Iodomethane; furthermore preparations based on *Bacillus firmus* (I-1582, Votivo) and azadirachtin (BioNeem), and also the following compounds: 1-{2-fluoro-4-methyl-5-[(2,2,2-trifluoroethyl)sulphonyl]phenyl}-3-(trifluoromethyl)-1H-1,2,4-triazole-5-amine (known from WO2006/043635) (CAS 885026-50-6), 2-chloro-N-[2-{1-[(2E)-3-(4-chlorophenyl)prop-2-en-1-yl]piperidin-4-yl}-4-(trifluoromethyl)phenyl]isonicotinamide (known from WO2006/003494) (CAS 872999-66-1), 3-(4-chloro-2,6-dimethylphenyl)-4-hydroxy-8-methoxy-1,8-diazaspiro[4.5]dec-3-en-2-one (known from

WO 2010052161) (CAS 1225292-17-0), 3-(4-chloro-2,6-dimethylphenyl)-8-methoxy-2-oxo-1,8-diazaspiro[4.5]dec-3-en-4-yl ethyl carbonate (known from EP2647626) (CAS 1440516-42-6), PF1364 (known from JP2010/018586) (CAS 1204776-60-2), (3*E*)-3-[1-[(6-chloro-3-pyridyl)methyl]-2-pyridylidene]-1,1,1-trifluoro-propan-2-one (known from WO2013/144213) (CAS 1461743-15-6), *N*-[3-(benzylcarbamoyl)-4-chlorophenyl]-1-methyl-3-(pentafluoroethyl)-4-(trifluoromethyl)-1*H*-pyrazole-5-carboxamide (known from WO2010/051926) (CAS 1226889-14-0), 5-bromo-4-chloro-*N*-[4-chloro-2-methyl-6-(methylcarbamoyl)phenyl]-2-(3-chloro-2-pyridyl)pyrazole-3-carboxamide (known from CN103232431) (CAS 1449220-44-3), 4-[5-(3,5-dichlorophenyl)-4,5-dihydro-5-(trifluoromethyl)-3-isoxazolyl]-2-methyl-*N*-(*cis*-1-oxido-3-thietanyl)-benzamide, 4-[5-(3,5-dichlorophenyl)-4,5-dihydro-5-(trifluoromethyl)-3-isoxazolyl]-2-methyl-*N*-(*trans*-1-oxido-3-thietanyl)-benzamide and 4-[(5*S*)-5-(3,5-dichlorophenyl)-4,5-dihydro-5-(trifluoromethyl)-3-isoxazolyl]-2-methyl-*N*-(*cis*-1-oxido-3-thietanyl) benzamide (known from WO 2013/050317 A1) (CAS 1332628-83-7), *N*-[3-chloro-1-(3-pyridinyl)-1*H*-pyrazol-4-yl]-*N*-ethyl-3-[(3,3,3-trifluoropropyl)sulfinyl]-propanamide, (+)-*N*-[3-chloro-1-(3-pyridinyl)-1*H*-pyrazol-4-yl]-*N*-ethyl-3-[(3,3,3-trifluoropropyl)sulfinyl]-propanamide and (-)-*N*-[3-chloro-1-(3-pyridinyl)-1*H*-pyrazol-4-yl]-*N*-ethyl-3-[(3,3,3-trifluoropropyl)sulfinyl]-propanamide (known from WO 2013/162715 A2, WO 2013/162716 A2, US 2014/0213448 A1) (CAS 1477923-37-7), 5-[[*(2E)*-3-chloro-2-propen-1-yl]amino]-1-[2,6-dichloro-4-(trifluoromethyl)phenyl]-4-[(trifluoromethyl)sulfinyl]-1*H*-pyrazole-3-carbonitrile (known from CN 101337937 A) (CAS 1105672-77-2), 3-bromo-*N*-[4-chloro-2-methyl-6-[(methylamino)thioxomethyl]phenyl]-1-(3-chloro-2-pyridinyl)-1*H*-pyrazole-5-carboxamide, (Liudaibenjiaxuanan, known from CN 103109816 A) (CAS 1232543-85-9); *N*-[4-chloro-2-[[1,1-dimethylethyl]amino]carbonyl]-6-methylphenyl]-1-(3-chloro-2-pyridinyl)-3-(fluoromethoxy)-1*H*-pyrazole-5-carboxamide (known from WO 2012/034403 A1) (CAS 1268277-22-0), *N*-[2-(5-amino-1,3,4-thiadiazol-2-yl)-4-chloro-6-methylphenyl]-3-bromo-1-(3-chloro-2-pyridinyl)-1*H*-pyrazole-5-carboxamide (known from WO 2011/085575 A1) (CAS 1233882-22-8), 4-[3-[2,6-dichloro-4-[(3,3-dichloro-2-propen-1-yl)oxy]phenoxy]propoxy]-2-methoxy-6-(trifluoromethyl)-pyrimidine (known from CN 101337940 A) (CAS 1108184-52-6); (*2E*)- and 2(*Z*)-2-[2-(4-cyanophenyl)-1-[3-(trifluoromethyl)phenyl]ethylidene]-*N*-[4-(difluoromethoxy)phenyl]-hydrazinecarboxamide (known from CN 101715774 A) (CAS 1232543-85-9); 3-(2,2-dichloroethyl)-2,2-dimethyl-4-(1*H*-benzimidazol-2-yl)phenyl-cyclopropanecarboxylic acid ester (known from CN 103524422 A) (CAS 1542271-46-4); (4*aS*)-7-chloro-2,5-dihydro-2-[[methoxycarbonyl][4-[(trifluoromethyl)thio]phenyl]amino]carbonyl]-indeno[1,2-*e*][1,3,4]oxadiazine-4*a*(3*H*)-carboxylic acid methyl ester (known from CN 102391261 A) (CAS 1370358-69-2); 6-deoxy-3-*O*-ethyl-2,4-di-*O*-methyl-, 1-[*N*-[4-[1-[4-(1,1,2,2,2-pentafluoroethoxy)phenyl]-1*H*-1,2,4-triazol-3-yl]phenyl]carbamate]- α -L-mannopyranose (known from US 2014/0275503 A1) (CAS 1181213-14-8); 8-(2-cyclopropylmethoxy-4-trifluoromethyl-phenoxy)-3-(6-trifluoromethyl-pyridazin-3-yl)-3-aza-bicyclo[3.2.1]octane (CAS 1253850-56-4), (8-*anti*)-8-(2-cyclopropylmethoxy-4-trifluoromethyl-phenoxy)-3-(6-trifluoromethyl-pyridazin-3-yl)-3-aza-bicyclo[3.2.1]octane (CAS 933798-27-7), (8-*syn*)-8-(2-cyclopropylmethoxy-4-trifluoromethyl-phenoxy)-3-(6-trifluoromethyl-pyridazin-3-yl)-3-aza-bicyclo[3.2.1]octane (known from WO 2007040280 A1,

WO 2007040282 A1) (CAS 934001-66-8), N-[4-(aminothioxomethyl)-2-methyl-6-[(methylamino)carbonyl]phenyl]-3-bromo-1-(3-chloro-2-pyridinyl)-1*H*-pyrazole-5-carboxamide (known from CN 103265527 A) (CAS 1452877-50-7), 3-(4-chloro-2,6-dimethylphenyl)-8-methoxy-1-methyl-1,8-diazaspiro[4.5]decane-2,4-dione (known from WO 2014/187846 A1) (CAS 1638765-58-8), 3-(4-chloro-2,6-dimethylphenyl)-8-methoxy-1-methyl-2-oxo-1,8-diazaspiro[4.5]dec-3-en-4-yl-carbonic acid ethyl ester (known from WO 2010/066780 A1, WO 2011151146 A1) (CAS 1229023-00-0), *N*-[1-(2,6-difluorophenyl)-1*H*-pyrazol-3-yl]-2-(trifluoromethyl)benzamide (known from WO 2014/053450 A1) (CAS 1594624-87-9), *N*-[2-(2,6-difluorophenyl)-2*H*-1,2,3-triazol-4-yl]-2-(trifluoromethyl)benzamide (known from WO 2014/053450 A1) (CAS 1594637-65-6), *N*-[1-(3,5-difluoro-2-pyridinyl)-1*H*-pyrazol-3-yl]-2-(trifluoromethyl)benzamide (known from WO 2014/053450 A1) (CAS 1594626-19-3), (3*R*)-3-(2-chloro-5-thiazolyl)-2,3-dihydro-8-methyl-5,7-dioxo-6-phenyl-5*H*-thiazolo[3,2-*a*]pyrimidinium inner salt (known from WO 2018/177970 A1) (CAS 2246757-58-2); 3-(2-chloro-5-thiazolyl)-2,3-dihydro-8-methyl-5,7-dioxo-6-phenyl-5*H*-thiazolo[3,2-*a*]pyrimidinium inner salt (known from WO 2018/177970 A1) (CAS 2246757-56-0); *N*-[3-chloro-1-(3-pyridinyl)-1*H*-pyrazol-4-yl]-2-(methylsulfonyl)propanamide (known from WO 2019/236274 A1) (CAS 2396747-83-2), *N*-[2-bromo-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]-6-(trifluoromethyl)phenyl]-2-fluoro-3-[(4-fluorobenzoyl)amino]-benzamide (known from WO 2019059412 A1) (CAS 1207977-87-4).

Fungicides

The active ingredients specified herein by their Common Name are known and described, for example, in The Pesticide Manual (16th Ed. British Crop Protection Council) or can be searched in the internet (e.g. www.alanwood.net/pesticides).

All named fungicidal mixing partners of the classes (1) to (15) can, if their functional groups enable this, optionally form salts with suitable bases or acids. All named mixing partners of the classes (1) to (15) can include tautomeric forms, where applicable.

1) Inhibitors of the ergosterol biosynthesis, for example (1.001) cyproconazole, (1.002) difenoconazole, (1.003) epoxiconazole, (1.004) fenhexamid, (1.005) fenpropidin, (1.006) fenpropimorph, (1.007) fenpyrazamine, (1.008) fluquinconazole, (1.009) flutriafol, (1.010) imazalil, (1.011) imazalil sulfate, (1.012) ipconazole, (1.013) metconazole, (1.014) myclobutanil, (1.015) paclobutrazol, (1.016) prochloraz, (1.017) propiconazole, (1.018) prothioconazole, (1.019) pyrisoxazole, (1.020) spiroxamine, (1.021) tebuconazole, (1.022) tetraconazole, (1.023) triadimenol, (1.024) tridemorph, (1.025) triticonazole, (1.026) (1*R*,2*S*,5*S*)-5-(4-chlorobenzyl)-2-(chloromethyl)-2-methyl-1-(1*H*-1,2,4-triazol-1-ylmethyl)cyclopentanol, (1.027) (1*S*,2*R*,5*R*)-5-(4-chlorobenzyl)-2-(chloromethyl)-2-methyl-1-(1*H*-1,2,4-triazol-1-ylmethyl)cyclopentanol, (1.028) (2*R*)-2-(1-chlorocyclopropyl)-4-[(1*R*)-2,2-dichlorocyclopropyl]-1-(1*H*-1,2,4-triazol-1-yl)butan-2-ol, (1.029) (2*R*)-2-(1-chlorocyclopropyl)-4-[(1*S*)-2,2-dichlorocyclopropyl]-1-(1*H*-1,2,4-triazol-1-yl)butan-2-ol, (1.030) (2*R*)-2-[4-(4-chlorophenoxy)-2-(trifluoromethyl)phenyl]-1-(1*H*-1,2,4-triazol-1-yl)propan-2-ol, (1.031) (2*S*)-2-(1-chlorocyclopropyl)-4-

- [(1R)-2,2-dichlorocyclopropyl]-1-(1H-1,2,4-triazol-1-yl)butan-2-ol, (1.032) (2S)-2-(1-chlorocyclopropyl)-4-[(1S)-2,2-dichlorocyclopropyl]-1-(1H-1,2,4-triazol-1-yl)butan-2-ol, (1.033) (2S)-2-[4-(4-chlorophenoxy)-2-(trifluoromethyl)phenyl]-1-(1H-1,2,4-triazol-1-yl)propan-2-ol, (1.034) (R)-[3-(4-chloro-2-fluorophenyl)-5-(2,4-difluorophenyl)-1,2-oxazol-4-yl](pyridin-3-yl)methanol, (1.035) (S)-[3-(4-chloro-2-fluorophenyl)-5-(2,4-difluorophenyl)-1,2-oxazol-4-yl](pyridin-3-yl)methanol, (1.036) [3-(4-chloro-2-fluorophenyl)-5-(2,4-difluorophenyl)-1,2-oxazol-4-yl](pyridin-3-yl)methanol, (1.037) 1-({(2R,4S)-2-[2-chloro-4-(4-chlorophenoxy)phenyl]-4-methyl-1,3-dioxolan-2-yl}methyl)-1H-1,2,4-triazole, (1.038) 1-({(2S,4S)-2-[2-chloro-4-(4-chlorophenoxy)phenyl]-4-methyl-1,3-dioxolan-2-yl}methyl)-1H-1,2,4-triazole, (1.039) 1-[[3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-1H-1,2,4-triazol-5-yl thiocyanate, (1.040) 1-[[rel(2R,3R)-3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-1H-1,2,4-triazol-5-yl thiocyanate, (1.041) 1-[[rel(2R,3S)-3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-1H-1,2,4-triazol-5-yl thiocyanate, (1.042) 2-[(2R,4R,5R)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.043) 2-[(2R,4R,5S)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.044) 2-[(2R,4S,5R)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.045) 2-[(2R,4S,5S)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.046) 2-[(2S,4R,5R)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.047) 2-[(2S,4R,5S)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.048) 2-[(2S,4S,5R)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.049) 2-[(2S,4S,5S)-1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.050) 2-[1-(2,4-dichlorophenyl)-5-hydroxy-2,6,6-trimethylheptan-4-yl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.051) 2-[2-chloro-4-(2,4-dichlorophenoxy)phenyl]-1-(1H-1,2,4-triazol-1-yl)propan-2-ol, (1.052) 2-[2-chloro-4-(4-chlorophenoxy)phenyl]-1-(1H-1,2,4-triazol-1-yl)butan-2-ol, (1.053) 2-[4-(4-chlorophenoxy)-2-(trifluoromethyl)phenyl]-1-(1H-1,2,4-triazol-1-yl)butan-2-ol, (1.054) 2-[4-(4-chlorophenoxy)-2-(trifluoromethyl)phenyl]-1-(1H-1,2,4-triazol-1-yl)pentan-2-ol, (1.055) Mefentrifluconazole, (1.056) 2-[[3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.057) 2-[[rel(2R,3R)-3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.058) 2-[[rel(2R,3S)-3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-2,4-dihydro-3H-1,2,4-triazole-3-thione, (1.059) 5-(4-chlorobenzyl)-2-(chloromethyl)-2-methyl-1-(1H-1,2,4-triazol-1-ylmethyl)cyclopentanol, (1.060) 5-(allylsulfanyl)-1-[[3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-1H-1,2,4-triazole, (1.061) 5-(allylsulfanyl)-1-[[rel(2R,3R)-3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-1H-1,2,4-triazole, (1.062) 5-(allylsulfanyl)-1-[[rel(2R,3S)-3-(2-chlorophenyl)-2-(2,4-difluorophenyl)oxiran-2-yl]methyl]-1H-1,2,4-triazole, (1.063) N'-(2,5-dimethyl-4-[[3-(1,1,2,2-tetrafluoroethoxy)phenyl]sulfanyl]phenyl)-N-ethyl-N-methylimidoforamide, (1.064) N'-(2,5-dimethyl-4-[[3-(2,2,2-trifluoroethoxy)phenyl]sulfanyl]phenyl)-N-ethyl-N-methylimidoforamide, (1.065) N'-

(2,5-dimethyl-4-{{3-(2,2,3,3-tetrafluoropropoxy)phenyl}sulfanyl}phenyl)-N-ethyl-N-methylimidoforamide, (1.066) N'-(2,5-dimethyl-4-{{3-(pentafluoroethoxy)phenyl}sulfanyl}phenyl)-N-ethyl-N-methylimidoforamide, (1.067) N'-(2,5-dimethyl-4-{{3-[(1,1,2,2-tetrafluoroethyl)sulfanyl]phenoxy}phenyl)-N-ethyl-N-methylimidoforamide, (1.068) N'-(2,5-dimethyl-4-{{3-[(2,2,2-trifluoroethyl)sulfanyl]phenoxy}phenyl)-N-ethyl-N-methylimidoforamide, (1.069) N'-(2,5-dimethyl-4-{{3-[(2,2,3,3-tetrafluoropropyl)sulfanyl]phenoxy}phenyl)-N-ethyl-N-methylimidoforamide, (1.070) N'-(2,5-dimethyl-4-{{3-[(pentafluoroethyl)sulfanyl]phenoxy}phenyl)-N-ethyl-N-methylimidoforamide, (1.071) N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylimidoforamide, (1.072) N'-(4-{{3-(difluoromethoxy)phenyl}sulfanyl}-2,5-dimethylphenyl)-N-ethyl-N-methylimidoforamide, (1.073) N'-(4-{{3-[(difluoromethyl)sulfanyl]phenoxy}-2,5-dimethylphenyl)-N-ethyl-N-methylimidoforamide, (1.074) N'-[5-bromo-6-(2,3-dihydro-1H-inden-2-yloxy)-2-methylpyridin-3-yl]-N-ethyl-N-methylimidoforamide, (1.075) N'-[4-[(4,5-dichloro-1,3-thiazol-2-yl)oxy]-2,5-dimethylphenyl]-N-ethyl-N-methylimidoforamide, (1.076) N'-[5-bromo-6-[(1R)-1-(3,5-difluorophenyl)ethoxy]-2-methylpyridin-3-yl]-N-ethyl-N-methylimidoforamide, (1.077) N'-[5-bromo-6-[(1S)-1-(3,5-difluorophenyl)ethoxy]-2-methylpyridin-3-yl]-N-ethyl-N-methylimidoforamide, (1.078) N'-[5-bromo-6-[(cis-4-isopropylcyclohexyl)oxy]-2-methylpyridin-3-yl]-N-ethyl-N-methylimidoforamide, (1.079) N'-[5-bromo-6-[(trans-4-isopropylcyclohexyl)oxy]-2-methylpyridin-3-yl]-N-ethyl-N-methylimidoforamide, (1.080) N'-[5-bromo-6-[1-(3,5-difluorophenyl)ethoxy]-2-methylpyridin-3-yl]-N-ethyl-N-methylimidoforamide, (1.081) ipfentrifluconazole, (1.082) 2-[4-(4-chlorophenoxy)-2-(trifluoromethyl)phenyl]-1-(1H-1,2,4-triazol-1-yl)propan-2-ol, (1.083) 2-[6-(4-bromophenoxy)-2-(trifluoromethyl)-3-pyridyl]-1-(1,2,4-triazol-1-yl)propan-2-ol, (1.084) 2-[6-(4-chlorophenoxy)-2-(trifluoromethyl)-3-pyridyl]-1-(1,2,4-triazol-1-yl)propan-2-ol, (1.085) 3-[2-(1-chlorocyclopropyl)-3-(3-chloro-2-fluoro-phenyl)-2-hydroxypropyl]imidazole-4-carbonitrile and (1.086) 4-[[6-[rac-(2R)-2-(2,4-difluorophenyl)-1,1-difluoro-2-hydroxy-3-(5-thioxo-4H-1,2,4-triazol-1-yl)propyl]-3-pyridyl]oxy]benzonitrile.

2) Inhibitors of the respiratory chain at complex I or II, for example (2.001) benzovindiflupyr, (2.002) bixafen, (2.003) boscalid, (2.004) carboxin, (2.005) fluopyram, (2.006) flutolanil, (2.007) fluxapyroxad, (2.008) furametpyr, (2.009) Isofetamid, (2.010) isopyrazam (anti-epimeric enantiomer 1R,4S,9S), (2.011) isopyrazam (anti-epimeric enantiomer 1S,4R,9R), (2.012) isopyrazam (anti-epimeric racemate 1RS,4SR,9SR), (2.013) isopyrazam (mixture of syn-epimeric racemate 1RS,4SR,9RS and anti-epimeric racemate 1RS,4SR,9SR), (2.014) isopyrazam (syn-epimeric enantiomer 1R,4S,9R), (2.015) isopyrazam (syn-epimeric enantiomer 1S,4R,9S), (2.016) isopyrazam (syn-epimeric racemate 1RS,4SR,9RS), (2.017) penflufen, (2.018) penthiopyrad, (2.019) pydiflumetofen, (2.020) Pyraziflumid, (2.021) sedaxane, (2.022) 1,3-dimethyl-N-(1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl)-1H-pyrazole-4-carboxamide, (2.023) 1,3-dimethyl-N-[(3R)-1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl]-1H-pyrazole-4-carboxamide, (2.024) 1,3-dimethyl-N-[(3S)-1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl]-1H-pyrazole-4-carboxamide, (2.025) 1-methyl-3-(trifluoromethyl)-N-[2'-(trifluoromethyl)biphenyl-2-yl]-1H-pyrazole-4-carboxamide, (2.026) 2-fluoro-6-(trifluoromethyl)-N-(1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl)benzamide, (2.027) 3-

- (difluoromethyl)-1-methyl-N-(1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl)-1H-pyrazole-4-carboxamide, (2.028) inpyrfluxam, (2.029) 3-(difluoromethyl)-1-methyl-N-[(3S)-1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl]-1H-pyrazole-4-carboxamide, (2.030) fluindapyr, (2.031) 3-(difluoromethyl)-N-[(3R)-7-fluoro-1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl]-1-methyl-1H-pyrazole-4-carboxamide, (2.032) 3-(difluoromethyl)-N-[(3S)-7-fluoro-1,1,3-trimethyl-2,3-dihydro-1H-inden-4-yl]-1-methyl-1H-pyrazole-4-carboxamide, (2.033) 5,8-difluoro-N-[2-(2-fluoro-4-{[4-(trifluoromethyl)pyridin-2-yl]oxy}phenyl)ethyl]quinazolin-4-amine, (2.034) N-(2-cyclopentyl-5-fluorobenzyl)-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.035) N-(2-tert-butyl-5-methylbenzyl)-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.036) N-(2-tert-butylbenzyl)-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.037) N-(5-chloro-2-ethylbenzyl)-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.038) N-(5-chloro-2-isopropylbenzyl)-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.039) N-[(1R,4S)-9-(dichloromethylene)-1,2,3,4-tetrahydro-1,4-methanonaphthalen-5-yl]-3-(difluoromethyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.040) N-[(1S,4R)-9-(dichloromethylene)-1,2,3,4-tetrahydro-1,4-methanonaphthalen-5-yl]-3-(difluoromethyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.041) N-[1-(2,4-dichlorophenyl)-1-methoxypropan-2-yl]-3-(difluoromethyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.042) N-[2-chloro-6-(trifluoromethyl)benzyl]-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.043) N-[3-chloro-2-fluoro-6-(trifluoromethyl)benzyl]-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.044) N-[5-chloro-2-(trifluoromethyl)benzyl]-N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.045) N-cyclopropyl-3-(difluoromethyl)-5-fluoro-1-methyl-N-[5-methyl-2-(trifluoromethyl)benzyl]-1H-pyrazole-4-carboxamide, (2.046) N-cyclopropyl-3-(difluoromethyl)-5-fluoro-N-(2-fluoro-6-isopropylbenzyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.047) N-cyclopropyl-3-(difluoromethyl)-5-fluoro-N-(2-isopropyl-5-methylbenzyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.048) N-cyclopropyl-3-(difluoromethyl)-5-fluoro-N-(2-isopropylbenzyl)-1-methyl-1H-pyrazole-4-carbothioamide, (2.049) N-cyclopropyl-3-(difluoromethyl)-5-fluoro-N-(2-isopropylbenzyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.050) N-cyclopropyl-3-(difluoromethyl)-5-fluoro-N-(5-fluoro-2-isopropylbenzyl)-1-methyl-1H-pyrazole-4-carboxamide, (2.051) N-cyclopropyl-3-(difluoromethyl)-N-(2-ethyl-4,5-dimethylbenzyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.052) N-cyclopropyl-3-(difluoromethyl)-N-(2-ethyl-5-fluorobenzyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.053) N-cyclopropyl-3-(difluoromethyl)-N-(2-ethyl-5-methylbenzyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.054) N-cyclopropyl-N-(2-cyclopropyl-5-fluorobenzyl)-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.055) N-cyclopropyl-N-(2-cyclopropyl-5-methylbenzyl)-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.056) N-cyclopropyl-N-(2-cyclopropylbenzyl)-3-(difluoromethyl)-5-fluoro-1-methyl-1H-pyrazole-4-carboxamide, (2.057) pyrapropoyne.

- 3) Inhibitors of the respiratory chain at complex III, for example (3.001) ametocetradin, (3.002) amisulbrom, (3.003) azoxystrobin, (3.004) coumethoxystrobin, (3.005) coumoxystrobin, (3.006) cyazofamid, (3.007) dimoxystrobin, (3.008) enoxastrobin, (3.009) famoxadone, (3.010) fenamidone, (3.011) flufenoxystrobin, (3.012) fluoxastrobin, (3.013) kresoxim-methyl, (3.014) metominostrobin, (3.015) orysastrobin, (3.016) picoxystrobin, (3.017) pyraclostrobin, (3.018) pyrametostrobin, (3.019) pyraoxystrobin, (3.020) trifloxystrobin, (3.021) (2E)-2-{2-[[[(1E)-1-(3-[[[(E)-1-fluoro-2-phenylvinyl]oxy}phenyl]ethylidene]amino}oxy)methyl]phenyl]-2-(methoxyimino)-N-methylacetamide, (3.022) (2E,3Z)-5-[[1-(4-chlorophenyl)-1H-pyrazol-3-yl]oxy]-2-(methoxyimino)-N,3-dimethylpent-3-enamide, (3.023) (2R)-2-{2-[(2,5-dimethylphenoxy)methyl]phenyl}-2-methoxy-N-methylacetamide, (3.024) (2S)-2-{2-[(2,5-dimethylphenoxy)methyl]phenyl}-2-methoxy-N-methylacetamide, (3.025) fenpicoxamid, (3.026) mandestrobin, (3.027) N-(3-ethyl-3,5,5-trimethylcyclohexyl)-3-formamido-2-hydroxybenzamide, (3.028) (2E,3Z)-5-[[1-(4-chloro-2-fluorophenyl)-1H-pyrazol-3-yl]oxy]-2-(methoxyimino)-N,3-dimethylpent-3-enamide, (3.029) methyl {5-[3-(2,4-dimethylphenyl)-1H-pyrazol-1-yl]-2-methylbenzyl}carbamate, (3.030) metyltetraprole, (3.031) florylpicoxamid.
- 4) Inhibitors of the mitosis and cell division, for example (4.001) carbendazim, (4.002) diethofencarb, (4.003) ethaboxam, (4.004) fluopicolide, (4.005) pencycuron, (4.006) thiabendazole, (4.007) thiophanate-methyl, (4.008) zoxamide, (4.009) 3-chloro-4-(2,6-difluorophenyl)-6-methyl-5-phenylpyridazine, (4.010) 3-chloro-5-(4-chlorophenyl)-4-(2,6-difluorophenyl)-6-methylpyridazine, (4.011) 3-chloro-5-(6-chloropyridin-3-yl)-6-methyl-4-(2,4,6-trifluorophenyl)pyridazine, (4.012) 4-(2-bromo-4-fluorophenyl)-N-(2,6-difluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.013) 4-(2-bromo-4-fluorophenyl)-N-(2-bromo-6-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.014) 4-(2-bromo-4-fluorophenyl)-N-(2-bromophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.015) 4-(2-bromo-4-fluorophenyl)-N-(2-chloro-6-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.016) 4-(2-bromo-4-fluorophenyl)-N-(2-chlorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.017) 4-(2-bromo-4-fluorophenyl)-N-(2-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.018) 4-(2-chloro-4-fluorophenyl)-N-(2,6-difluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.019) 4-(2-chloro-4-fluorophenyl)-N-(2-chloro-6-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.020) 4-(2-chloro-4-fluorophenyl)-N-(2-chlorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.021) 4-(2-chloro-4-fluorophenyl)-N-(2-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.022) 4-(4-chlorophenyl)-5-(2,6-difluorophenyl)-3,6-dimethylpyridazine, (4.023) N-(2-bromo-6-fluorophenyl)-4-(2-chloro-4-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.024) N-(2-bromophenyl)-4-(2-chloro-4-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine, (4.025) N-(4-chloro-2,6-difluorophenyl)-4-(2-chloro-4-fluorophenyl)-1,3-dimethyl-1H-pyrazol-5-amine.
- 5) Compounds capable to have a multisite action, for example (5.001) bordeaux mixture, (5.002) captafol, (5.003) captan, (5.004) chlorothalonil, (5.005) copper hydroxide, (5.006) copper naphthenate, (5.007) copper oxide, (5.008) copper oxychloride, (5.009) copper(2+) sulfate, (5.010) dithianon, (5.011) dodine, (5.012) folpet, (5.013) mancozeb, (5.014) maneb, (5.015) metiram, (5.016) metiram zinc, (5.017) oxine-

copper, (5.018) propineb, (5.019) sulfur and sulfur preparations including calcium polysulfide, (5.020) thiram, (5.021) zineb, (5.022) ziram, (5.023) 6-ethyl-5,7-dioxo-6,7-dihydro-5H-pyrrolo[3',4':5,6][1,4]dithiino[2,3-c][1,2]thiazole-3-carbonitrile.

6) Compounds capable to induce a host defence, for example (6.001) acibenzolar-S-methyl, (6.002) isotianil, (6.003) probenazole, (6.004) tiadinil.

7) Inhibitors of the amino acid and/or protein biosynthesis, for example (7.001) cyprodinil, (7.002) kasugamycin, (7.003) kasugamycin hydrochloride hydrate, (7.004) oxytetracycline, (7.005) pyrimethanil, (7.006) 3-(5-fluoro-3,3,4,4-tetramethyl-3,4-dihydroisoquinolin-1-yl)quinoline.

8) Inhibitors of the ATP production, for example (8.001) silthiofam.

9) Inhibitors of the cell wall synthesis, for example (9.001) benthiavalicarb, (9.002) dimethomorph, (9.003) flumorph, (9.004) iprovalicarb, (9.005) mandipropamid, (9.006) pyrimorph, (9.007) valifenalate, (9.008) (2E)-3-(4-tert-butylphenyl)-3-(2-chloropyridin-4-yl)-1-(morpholin-4-yl)prop-2-en-1-one, (9.009) (2Z)-3-(4-tert-butylphenyl)-3-(2-chloropyridin-4-yl)-1-(morpholin-4-yl)prop-2-en-1-one.

10) Inhibitors of the lipid and membrane synthesis, for example (10.001) propamocarb, (10.002) propamocarb hydrochloride, (10.003) tolclofos-methyl.

11) Inhibitors of the melanin biosynthesis, for example (11.001) tricyclazole, (11.002) 2,2,2-trifluoroethyl {3-methyl-1-[(4-methylbenzoyl)amino]butan-2-yl} carbamate.

12) Inhibitors of the nucleic acid synthesis, for example (12.001) benalaxyl, (12.002) benalaxyl-M (kiralaxyl), (12.003) metalaxyl, (12.004) metalaxyl-M (mefenoxam).

13) Inhibitors of the signal transduction, for example (13.001) fludioxonil, (13.002) iprodione, (13.003) procymidone, (13.004) proquinazid, (13.005) quinoxifen, (13.006) vinclozolin.

14) Compounds capable to act as an uncoupler, for example (14.001) fluazinam, (14.002) meptyldinocap.

15) Further fungicides selected from the group consisting of (15.001) abscisic acid, (15.002) benthiazole, (15.003) bethoxazin, (15.004) capsimycin, (15.005) carvone, (15.006) chinomethionat, (15.007) cufraneb, (15.008) cyflufenamid, (15.009) cymoxanil, (15.010) cyprosulfamide, (15.011) flutianil, (15.012) fosetyl-aluminium, (15.013) fosetyl-calcium, (15.014) fosetyl-sodium, (15.015) methyl isothiocyanate, (15.016) metrafenone, (15.017) mildiomicin, (15.018) natamycin, (15.019) nickel dimethyldithiocarbamate, (15.020) nitrothal-isopropyl, (15.021) oxamocarb, (15.022) Oxathiapiprolin, (15.023) oxyfenthiin, (15.024) pentachlorophenol and salts, (15.025) phosphorous acid and its salts, (15.026) propamocarb-fosetate, (15.027) pyriofenone (chlazafenone), (15.028) tebufloquin, (15.029) tecloftalam, (15.030) tolnifanide, (15.031) 1-(4-{4-[(5R)-5-(2,6-difluorophenyl)-4,5-dihydro-1,2-oxazol-3-yl]-1,3-thiazol-2-yl}piperidin-1-yl)-2-[5-methyl-3-(trifluoromethyl)-1H-pyrazol-1-yl]ethanone, (15.032) 1-(4-{4-[(5S)-5-

(2,6-difluorophenyl)-4,5-dihydro-1,2-oxazol-3-yl]-1,3-thiazol-2-yl]piperidin-1-yl)-2-[5-methyl-3-(trifluoromethyl)-1H-pyrazol-1-yl]ethanone, (15.033) 2-(6-benzylpyridin-2-yl)quinazoline, (15.034) dipymetitron, (15.035) 2-[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]-1-[4-(4-{5-[2-(prop-2-yn-1-yloxy)phenyl]-4,5-dihydro-1,2-oxazol-3-yl]-1,3-thiazol-2-yl]piperidin-1-yl]ethanone, (15.036) 2-[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]-1-[4-(4-{5-[2-chloro-6-(prop-2-yn-1-yloxy)phenyl]-4,5-dihydro-1,2-oxazol-3-yl]-1,3-thiazol-2-yl]piperidin-1-yl]ethanone, (15.037) 2-[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]-1-[4-(4-{5-[2-fluoro-6-(prop-2-yn-1-yloxy)phenyl]-4,5-dihydro-1,2-oxazol-3-yl]-1,3-thiazol-2-yl]piperidin-1-yl]ethanone, (15.038) 2-[6-(3-fluoro-4-methoxyphenyl)-5-methylpyridin-2-yl]quinazoline, (15.039) 2-[(5R)-3-[2-(1-{[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]acetyl}piperidin-4-yl)-1,3-thiazol-4-yl]-4,5-dihydro-1,2-oxazol-5-yl]-3-chlorophenyl methanesulfonate, (15.040) 2-[(5S)-3-[2-(1-{[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]acetyl}piperidin-4-yl)-1,3-thiazol-4-yl]-4,5-dihydro-1,2-oxazol-5-yl]-3-chlorophenyl methanesulfonate, (15.041) Ipflufenoquin, (15.042) 2-[2-fluoro-6-[(8-fluoro-2-methylquinolin-3-yl)oxy]phenyl]propan-2-ol, (15.043) fluoxapiprolin, (15.044) 2-[3-[2-(1-{[3,5-bis(difluoromethyl)-1H-pyrazol-1-yl]acetyl}piperidin-4-yl)-1,3-thiazol-4-yl]-4,5-dihydro-1,2-oxazol-5-yl]phenyl methanesulfonate, (15.045) 2-phenylphenol and salts, (15.046) 3-(4,4,5-trifluoro-3,3-dimethyl-3,4-dihydroisoquinolin-1-yl)quinoline, (15.047) quinofumelin, (15.048) 4-amino-5-fluoropyrimidin-2-ol (tautomeric form: 4-amino-5-fluoropyrimidin-2(1H)-one), (15.049) 4-oxo-4-[(2-phenylethyl)amino]butanoic acid, (15.050) 5-amino-1,3,4-thiadiazole-2-thiol, (15.051) 5-chloro-N'-phenyl-N'-(prop-2-yn-1-yl)thiophene-2-sulfonohydrazide, (15.052) 5-fluoro-2-[(4-fluorobenzyl)oxy]pyrimidin-4-amine, (15.053) 5-fluoro-2-[(4-methylbenzyl)oxy]pyrimidin-4-amine, (15.054) 9-fluoro-2,2-dimethyl-5-(quinolin-3-yl)-2,3-dihydro-1,4-benzoxazepine, (15.055) but-3-yn-1-yl {6-[(Z)-(1-methyl-1H-tetrazol-5-yl)(phenyl)methylene]amino}oxy)methylpyridin-2-yl} carbamate, (15.056) ethyl (2Z)-3-amino-2-cyano-3-phenylacrylate, (15.057) phenazine-1-carboxylic acid, (15.058) propyl 3,4,5-trihydroxybenzoate, (15.059) quinolin-8-ol, (15.060) quinolin-8-ol sulfate (2:1), (15.061) tert-butyl {6-[(Z)-(1-methyl-1H-tetrazol-5-yl)(phenyl)methylene]amino}oxy)methylpyridin-2-yl} carbamate, (15.062) 5-fluoro-4-imino-3-methyl-1-[(4-methylphenyl)sulfonyl]-3,4-dihydropyrimidin-2(1H)-one, (15.063) aminopyrifin, (15.064) (N'-[2-chloro-4-(2-fluorophenoxy)-5-methylphenyl]-N-ethyl-N-methylimidoforamide), (15.065) (N'-(2-chloro-5-methyl-4-phenoxyphenyl)-N-ethyl-N-methylimido-formamide), (15.066) (2-[2-[(7,8-difluoro-2-methylquinolin-3-yl)oxy]-6-fluorophenyl]propan-2-ol), (15.067) (5-bromo-1-(5,6-dimethylpyridin-3-yl)-3,3-dimethyl-3,4-dihydroisoquinoline), (15.068) (3-(4,4-difluoro-5,5-dimethyl-4,5-dihydrothieno[2,3-c]pyridin-7-yl)quinoline), (15.069) (1-(4,5-dimethyl-1H-benzimidazol-1-yl)-4,4-difluoro-3,3-dimethyl-3,4-dihydroisoquinoline), (15.070) 8-fluoro-3-(5-fluoro-3,3-dimethyl-3,4-dihydroisoquinolin-1-yl)quinolone, (15.071) 8-fluoro-3-(5-fluoro-3,3,4,4-tetramethyl-3,4-dihydroisoquinolin-1-yl)quinolone, (15.072) 3-(4,4-difluoro-3,3-dimethyl-3,4-dihydroisoquinolin-1-yl)-8-fluoroquinoline, (15.073) (N-methyl-N-phenyl-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzamide), (15.074) (methyl[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]carbamate), (15.075) (N-{4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzyl}-cyclopropane-carboxamide), (15.076) N-methyl-4-(5-(trifluoromethyl)-1,2,4-

- oxadiazol-3-yl]benzamide, (15.077) N-[(E)-methoxyiminomethyl]-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzamide, (15.078) N-[(Z)-methoxyiminomethyl]-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzamide, (15.079) N-[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]-cyclopropane-carboxamide, (15.080) N-(2-fluorophenyl)-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzamide, (15.081) 2,2-difluoro-N-methyl-2-[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]-acetamide, (15.082) N-allyl-N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]acetamide, (15.083) N-[(E)-N-methoxy-C-methyl-carbonimidoyl]-4-(5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl)-benzamide, (15.084) N-[(Z)-N-methoxy-C-methyl-carbonimidoyl]-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzamide, (15.085) N-allyl-N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]-propanamide, (15.086) 4,4-dimethyl-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]-pyrrolidin-2-one, (15.087) N-methyl-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzenecarbothioamide, (15.088) 5-methyl-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]pyrrolidin-2-one, (15.089) N-((2,3-difluoro-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl)methyl)-3,3,3-trifluoro-propanamide, (15.090) 1-methoxy-1-methyl-3-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]-methyl]urea, (15.091) 1,1-diethyl-3-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]urea, (15.092) N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]propanamide, (15.093) N-methoxy-N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]cyclopropanecarboxamide, (15.094) 1-methoxy-3-methyl-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]urea, (15.095) N-methoxy-N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]-methyl]-cyclopropane-carboxamide, (15.096) N,2-dimethoxy-N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]-methyl]-propanamide, (15.097) N-ethyl-2-methyl-N-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]-propanamide, (15.098) 1-methoxy-3-methyl-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]-methyl]-urea, (15.099) 1,3-dimethoxy-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]urea, (15.100) 3-ethyl-1-methoxy-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]urea, (15.101) 1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]piperidin-2-one, (15.102) 4,4-dimethyl-2-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]isoxazolidin-3-one, (15.103) 5,5-dimethyl-2-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]phenyl]methyl]isoxazolidin-3-one, (15.104) 3,3-dimethyl-1-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]-phenyl]-methyl]-piperidin-2-one, (15.105) 1-[[3-fluoro-4-(5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl)-phenyl]-methyl]-azepan-2-one, (15.106) 4,4-dimethyl-2-[[4-(5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl)-phenyl]-methyl]isoxazolidin-3-one (15.107) 5,5-dimethyl-2-[[4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]-phenyl]methyl]isoxazolidin-3-one, (15.108) ethyl (1-{4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzyl}-1H-pyrazol-4-yl)acetate, (15.109) N,N-dimethyl-1-{4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzyl}-1H-1,2,4-triazol-3-amine and (15.110) N-{2,3-difluoro-4-[5-(trifluoromethyl)-1,2,4-oxadiazol-3-yl]benzyl}butanamide.

Biological pesticides as mixing components

The compounds of the formula (I) can be combined with biological pesticides.

Biological pesticides comprise in particular bacteria, fungi, yeasts, plant extracts and products formed by microorganisms, including proteins and secondary metabolites.

Biological pesticides comprise bacteria such as spore-forming bacteria, root-colonising bacteria and bacteria which act as biological insecticides, fungicides or nematocides.

- 5 Examples of such bacteria which are employed or can be used as biological pesticides are:

Bacillus amyloliquefaciens, strain FZB42 (DSM 231179), or *Bacillus cereus*, in particular *B. cereus* strain CNCM I-1562 or *Bacillus firmus*, strain I-1582 (Accession number CNCM I-1582) or *Bacillus pumilus*, in particular strain GB34 (Accession No. ATCC 700814) and strain QST2808 (Accession No. NRRL B-30087), or *Bacillus subtilis*, in particular strain GB03 (Accession No. ATCC SD-1397), or *Bacillus subtilis* strain QST713 (Accession No. NRRL B-21661) or *Bacillus subtilis* strain OST 30002 (Accession No. NRRL B-50421) *Bacillus thuringiensis*, in particular *B. thuringiensis* subspecies *israelensis* (serotype H-14), strain AM65-52 (Accession No. ATCC 1276), or *B. thuringiensis* subsp. *aizawai*, in particular strain ABTS-1857 (SD-1372), or *B. thuringiensis* subsp. *kurstaki* strain HD-1, or *B. thuringiensis* subsp. *tenebrionis* strain NB 176 (SD-5428), *Pasteuria penetrans*, *Pasteuria* spp. (Rotylenchulus reniformis nematode)-PR3 (Accession Number ATCC SD-5834), *Streptomyces microflavus* strain AQ6121 (= QRD 31.013, NRRL B-50550), *Streptomyces galbus* strain AQ 6047 (Accession Number NRRL 30232).

Examples of fungi and yeasts which are employed or can be used as biological pesticides are:

Beauveria bassiana, in particular strain ATCC 74040, *Coniothyrium minitans*, in particular strain CON/M/91-8 (Accession No. DSM-9660), *Lecanicillium* spp., in particular strain HRO LEC 12, *Lecanicillium lecanii*, (formerly known as *Verticillium lecanii*), in particular strain KV01, *Metarhizium anisopliae*, in particular strain F52 (DSM3884/ ATCC 90448), *Metschnikowia fructicola*, in particular strain NRRL Y-30752, *Paecilomyces fumosoroseus* (now: *Isaria fumosorosea*), in particular strain IFPC 200613, or strain Apopka 97 (Accession No. ATCC 20874), *Paecilomyces lilacinus*, in particular *P. lilacinus* strain 251 (AGAL 89/030550), *Talaromyces flavus*, in particular strain V117b, *Trichoderma atroviride*, in particular strain SC1 (Accession Number CBS 122089), *Trichoderma harzianum*, in particular *T. harzianum* rifai T39. (Accession Number CNCM I-952).

Examples of viruses which are employed or can be used as biological pesticides are:

Adoxophyes orana (summer fruit tortrix) granulosis virus (GV), *Cydia pomonella* (codling moth) granulosis virus (GV), *Helicoverpa armigera* (cotton bollworm) nuclear polyhedrosis virus (NPV), *Spodoptera exigua* (beet armyworm) mNPV, *Spodoptera frugiperda* (fall armyworm) mNPV, *Spodoptera littoralis* (African cotton leafworm) NPV.

Also included are bacteria and fungi which are added as 'inoculant' to plants or plant parts or plant organs and which, by virtue of their particular properties, promote plant growth and plant health. Examples which may be mentioned are:

5 *Agrobacterium* spp., *Azorhizobium caulinodans*, *Azospirillum* spp., *Azotobacter* spp., *Bradyrhizobium* spp., *Burkholderia* spp., in particular *Burkholderia cepacia* (formerly known as *Pseudomonas cepacia*), *Gigaspora* spp., or *Gigaspora monosporum*, *Glomus* spp., *Laccaria* spp., *Lactobacillus buchneri*, *Paraglomus* spp., *Pisolithus tinctorius*, *Pseudomonas* spp., *Rhizobium* spp., in particular *Rhizobium trifolii*, *Rhizopogon* spp., *Scleroderma* spp., *Suillus* spp., *Streptomyces* spp.

10 Examples of plant extracts and products formed by microorganisms including proteins and secondary metabolites which are employed or can be used as biological pesticides are:

Allium sativum, Artemisia absinthium, azadirachtin, Biokeeper WP, Cassia nigricans, Celastrus angulatus, Chenopodium anthelminticum, chitin, Armour-Zen, Dryopteris filix-mas, Equisetum arvense, Fortune Aza, Fungastop, Heads Up (Chenopodium quinoa saponin extract), Pyrethrum/Pyrethrins, Quassia amara, Quercus, Quillaja, Regalia, "RequiemTM Insecticide", rotenone, ryania/ryanodine, 15 Symphytum officinale, Tanacetum vulgare, thymol, Triact 70, TriCon, Tropaeolum majus, Urtica dioica, Veratrin, Viscum album, Brassicaceae extract, in particular oilseed rape powder or mustard powder, as well as bioinsecticidal / acaricidal active substances obtained from olive oil, in particular unsaturated fatty/carboxylic acids having carbon chain lengths C₁₆-C₂₀ as active ingredients, such as, for example, contained in the product with the trade name FLiPPER®.

20 **Safener as mixing components**

The compounds of the formula (I) can be combined with safeners such as, for example, benoxacor, cloquintocet (-mexyl), cyometrinil, cyprosulfamide, dichlormid, fenchlorazole (-ethyl), fencloirum, flurazole, fluxofenim, furilazole, isoxadifen (-ethyl), mefenpyr (-diethyl), naphthalic anhydride, oxabetrinil, 2-methoxy-N-({4-[(methylcarbamoyl)amino]phenyl}sulphonyl)benzamide (CAS 129531- 12-0), 4-(dichloroacetyl)-1-oxa-4-azaspiro[4.5]decane (CAS 71526-07-3), 2,2,5-trimethyl-3- 25 (dichloroacetyl)-1,3-oxazolidine (CAS 52836-31-4).

Plants and plant parts

All plants and plant parts can be treated in accordance with the invention. Here, plants are to be understood to mean all plants and plant parts such as wanted and unwanted wild plants or crop plants (including 30 naturally occurring crop plants), for example cereals (wheat, rice, triticale, barley, rye, oats), maize, soya bean, potato, sugar beet, sugar cane, tomatoes, pepper, cucumber, melon, carrot, watermelon, onion, lettuce, spinach, leek, beans, *Brassica oleracea* (e.g. cabbage) and other vegetable species, cotton, tobacco, oilseed rape, and also fruit plants (with the fruits apples, pears, citrus fruits and grapevines). Crop plants can be plants which can be obtained by conventional breeding and optimization methods or by

biotechnological and genetic engineering methods or combinations of these methods, including the transgenic plants and including the plant varieties which can or cannot be protected by varietal property rights. Plants should be understood to mean all developmental stages, such as seeds, seedlings, young (immature) plants up to mature plants. Plant parts should be understood to mean all parts and organs of the plants above and below ground, such as shoot, leaf, flower and root, examples given being leaves, needles, stalks, stems, flowers, fruit bodies, fruits and seeds, and also tubers, roots and rhizomes. Parts of plants also include harvested plants or harvested plant parts and vegetative and generative propagation material, for example seedlings, tubers, rhizomes, cuttings and seeds.

Treatment according to the invention of the plants and plant parts with the compounds of the formula (I) is carried out directly or by allowing the compounds to act on the surroundings, environment or storage space by the customary treatment methods, for example by immersion, spraying, evaporation, fogging, scattering, painting on, injection and, in the case of propagation material, in particular in the case of seeds, also by applying one or more coats.

As already mentioned above, it is possible to treat all plants and their parts according to the invention. In a preferred embodiment, wild plant species and plant cultivars, or those obtained by conventional biological breeding methods, such as crossing or protoplast fusion, and also parts thereof, are treated. In a further preferred embodiment, transgenic plants and plant cultivars obtained by genetic engineering methods, if appropriate in combination with conventional methods (genetically modified organisms), and parts thereof are treated. The term "parts" or "parts of plants" or "plant parts" has been explained above. The invention is used with particular preference to treat plants of the respective commercially customary cultivars or those that are in use. Plant cultivars are to be understood as meaning plants having new properties ("traits") and which have been obtained by conventional breeding, by mutagenesis or by recombinant DNA techniques. They can be cultivars, varieties, bio- or genotypes.

Transgenic plant, seed treatment and integration events

According to the invention, the compounds of formula (I) can be advantageously used to treat transgenic plants, plant cultivars or plant parts that received genetic material which imparts advantageous and/or useful properties (traits) to these plants, plant cultivars or plant parts. Therefore, it is contemplated that the present invention may be combined with one or more recombinant traits or transgenic event(s) or a combination thereof. For the purposes of this application, a transgenic event is created by the insertion of a specific recombinant DNA molecule into a specific position (locus) within the chromosome of the plant genome. The insertion creates a novel DNA sequence referred to as an "event" and is characterized by the inserted recombinant DNA molecule and some amount of genomic DNA immediately adjacent to/flanking both ends of the inserted DNA. Such trait(s) or transgenic event(s) include, but are not limited to, pest resistance, water use efficiency, yield performance, drought tolerance, seed quality, improved nutritional quality, hybrid seed production, and herbicide tolerance, in which the trait is measured with respect to a plant lacking such trait or transgenic event. Concrete examples of such advantageous and/or useful

properties (traits) are better plant growth, vigor, stress tolerance, standability, lodging resistance, nutrient uptake, plant nutrition, and/or yield, in particular improved growth, increased tolerance to high or low temperatures, increased tolerance to drought or to levels of water or soil salinity, enhanced flowering performance, easier harvesting, accelerated ripening, higher yields, higher quality and/or a higher nutritional value of the harvested products, better storage life and/or processability of the harvested products, and increased resistance or tolerance against animal and microbial pests, such as against insects, arachnids, nematodes, mites, slugs and snails.

Among DNA sequences encoding proteins which confer properties of resistance or tolerance to such animal and microbial pests, in particular insects, mention will particularly be made of the genetic material from *Bacillus thuringiensis* encoding the Bt proteins widely described in the literature and well known to those skilled in the art. Mention will also be made of proteins extracted from bacteria such as *Photorhabdus* (WO97/17432 and WO98/08932). In particular, mention will be made of the Bt Cry or VIP proteins which include the CryIA, CryIAb, CryIAc, CryIIA, CryIIIA, CryIIIB2, Cry9c Cry2Ab, Cry3Bb and CryIF proteins or toxic fragments thereof and also hybrids or combinations thereof, especially the CryIF protein or hybrids derived from a CryIF protein (e.g. hybrid CryIA-CryIF proteins or toxic fragments thereof), the CryIA-type proteins or toxic fragments thereof, preferably the CryIAc protein or hybrids derived from the CryIAc protein (e.g. hybrid CryIAb-CryIAc proteins) or the CryIAb or Bt2 protein or toxic fragments thereof, the Cry2Ae, Cry2Af or Cry2Ag proteins or toxic fragments thereof, the CryIA.105 protein or a toxic fragment thereof, the VIP3Aa19 protein, the VIP3Aa20 protein, the VIP3A proteins produced in the COT202 or COT203 cotton events, the VIP3Aa protein or a toxic fragment thereof as described in Estruch et al. (1996), Proc Natl Acad Sci US A. 28;93(11):5389-94, the Cry proteins as described in WO2001/47952, the insecticidal proteins from *Xenorhabdus* (as described in WO98/50427), *Serratia* (particularly from *S. entomophila*) or *Photorhabdus* species strains, such as Tc-proteins from *Photorhabdus* as described in WO98/08932. Also any variants or mutants of any one of these proteins differing in some amino acids (1-10, preferably 1-5) from any of the above named sequences, particularly the sequence of their toxic fragment, or which are fused to a transit peptide, such as a plastid transit peptide, or another protein or peptide, is included herein.

Another and particularly emphasized example of such properties is conferred tolerance to one or more herbicides, for example imidazolinones, sulphonylureas, glyphosate or phosphinothricin. Among DNA sequences encoding proteins which confer properties of tolerance to certain herbicides on the transformed plant cells and plants, mention will be particularly be made to the bar or PAT gene or the *Streptomyces coelicolor* gene described in WO2009/152359 which confers tolerance to glufosinate herbicides, a gene encoding a suitable EPSPS (5-Enolpyruvylshikimate-3-phosphate-synthase) which confers tolerance to herbicides having EPSPS as a target, especially herbicides such as glyphosate and its salts, a gene encoding glyphosate-n-acetyltransferase, or a gene encoding glyphosate oxidoreductase. Further suitable herbicide tolerance traits include at least one ALS (acetolactate synthase) inhibitor (e.g. WO2007/024782), a mutated *Arabidopsis* ALS/AHAS gene (e.g. U.S. Patent 6,855,533), genes encoding 2,4-D-

monooxygenases conferring tolerance to 2,4-D (2,4- dichlorophenoxyacetic acid) and genes encoding Dicamba monooxygenases conferring tolerance to dicamba (3,6-dichloro-2- methoxybenzoic acid).

Further and particularly emphasized examples of such properties are increased resistance against phytopathogenic fungi, bacteria and/or viruses owing, for example, to systemic acquired resistance (SAR), systemin, phytoalexins, elicitors and also resistance genes and correspondingly expressed proteins and toxins.

Particularly useful transgenic events in transgenic plants or plant cultivars which can be treated with preference in accordance with the invention include Event 531/ PV-GHBK04 (cotton, insect control, described in WO2002/040677), Event 1143-14A (cotton, insect control, not deposited, described in WO2006/128569); Event 1143-51B (cotton, insect control, not deposited, described in WO2006/128570); Event 1445 (cotton, herbicide tolerance, not deposited, described in US-A 2002-120964 or WO2002/034946); Event 17053 (rice, herbicide tolerance, deposited as PTA-9843, described in WO2010/117737); Event 17314 (rice, herbicide tolerance, deposited as PTA-9844, described in WO2010/117735); Event 281-24-236 (cotton, insect control - herbicide tolerance, deposited as PTA-6233, described in WO2005/103266 or US-A 2005-216969); Event 3006-210-23 (cotton, insect control - herbicide tolerance, deposited as PTA-6233, described in US-A 2007-143876 or WO2005/103266); Event 3272 (corn, quality trait, deposited as PTA-9972, described in WO2006/098952 or US-A 2006-230473); Event 33391 (wheat, herbicide tolerance, deposited as PTA-2347, described in WO2002/027004), Event 40416 (corn, insect control - herbicide tolerance, deposited as ATCC PTA-11508, described in WO 11/075593); Event 43A47 (corn, insect control - herbicide tolerance, deposited as ATCC PTA-11509, described in WO2011/075595); Event 5307 (corn, insect control, deposited as ATCC PTA-9561, described in WO2010/077816); Event ASR-368 (bent grass, herbicide tolerance, deposited as ATCC PTA-4816, described in US-A 2006-162007 or WO2004/053062); Event B16 (corn, herbicide tolerance, not deposited, described in US-A 2003-126634); Event BPS-CV127- 9 (soybean, herbicide tolerance, deposited as NCIMB No. 41603, described in WO2010/080829); Event BLRI (oilseed rape, restoration of male sterility, deposited as NCIMB 41193, described in WO2005/074671), Event CE43-67B (cotton, insect control, deposited as DSM ACC2724, described in US-A 2009-217423 or WO2006/128573); Event CE44-69D (cotton, insect control, not deposited, described in US-A 2010- 0024077); Event CE44-69D (cotton, insect control, not deposited, described in WO2006/128571); Event CE46-02A (cotton, insect control, not deposited, described in WO2006/128572); Event COT102 (cotton, insect control, not deposited, described in US-A 2006-130175 or WO2004/039986); Event COT202 (cotton, insect control, not deposited, described in US-A 2007-067868 or WO2005/054479); Event COT203 (cotton, insect control, not deposited, described in WO2005/054480);); Event DAS21606-3 / 1606 (soybean, herbicide tolerance, deposited as PTA-11028, described in WO2012/033794), Event DAS40278 (corn, herbicide tolerance, deposited as ATCC PTA-10244, described in WO2011/022469); Event DAS-44406-6 / pDAB8264.44.06.1 (soybean, herbicide tolerance, deposited as PTA-11336, described in WO2012/075426), Event DAS-14536-7 /pDAB8291.45.36.2 (soybean, herbicide tolerance, deposited as

PTA-11335, described in WO2012/075429), Event DAS-59122-7 (corn, insect control - herbicide tolerance, deposited as ATCC PTA 11384, described in US-A 2006-070139); Event DAS-59132 (corn, insect control - herbicide tolerance, not deposited, described in WO2009/100188); Event DAS68416 (soybean, herbicide tolerance, deposited as ATCC PTA-10442, described in WO2011/066384 or

5 WO2011/066360); Event DP-098140-6 (corn, herbicide tolerance, deposited as ATCC PTA-8296, described in US-A 2009- 137395 or WO 08/112019); Event DP-305423-1 (soybean, quality trait, not deposited, described in US-A 2008-312082 or WO2008/054747); Event DP-32138-1 (corn, hybridization system, deposited as ATCC PTA-9158, described in US-A 2009-0210970 or WO2009/103049); Event DP-356043-5 (soybean, herbicide tolerance, deposited as ATCC PTA-8287,

10 described in US-A 2010-0184079 or WO2008/002872); Event EE-I (brinjal, insect control, not deposited, described in WO 07/091277); Event Fil 17 (corn, herbicide tolerance, deposited as ATCC 209031, described in US-A 2006-059581 or WO 98/044140); Event FG72 (soybean, herbicide tolerance, deposited as PTA-11041, described in WO2011/063413), Event GA21 (corn, herbicide tolerance, deposited as ATCC 209033, described in US-A 2005-086719 or WO 98/044140); Event GG25 (corn, herbicide

15 tolerance, deposited as ATCC 209032, described in US-A 2005-188434 or WO98/044140); Event GHB119 (cotton, insect control - herbicide tolerance, deposited as ATCC PTA-8398, described in WO2008/151780); Event GHB614 (cotton, herbicide tolerance, deposited as ATCC PTA-6878, described in US-A 2010-050282 or WO2007/017186); Event GJ11 (corn, herbicide tolerance, deposited as ATCC 209030, described in US-A 2005-188434 or WO98/044140); Event GM RZ13 (sugar beet, virus

20 resistance, deposited as NCIMB-41601, described in WO2010/076212); Event H7-1 (sugar beet, herbicide tolerance, deposited as NCIMB 41158 or NCIMB 41159, described in US-A 2004-172669 or WO 2004/074492); Event JOPLINI (wheat, disease tolerance, not deposited, described in US-A 2008-064032); Event LL27 (soybean, herbicide tolerance, deposited as NCIMB41658, described in WO2006/108674 or US-A 2008-320616); Event LL55 (soybean, herbicide tolerance, deposited as NCIMB 41660, described

25 in WO 2006/108675 or US-A 2008-196127); Event LLcotton25 (cotton, herbicide tolerance, deposited as ATCC PTA-3343, described in WO2003/013224 or US- A 2003-097687); Event LLRICE06 (rice, herbicide tolerance, deposited as ATCC 203353, described in US 6,468,747 or WO2000/026345); Event LLRice62 (rice, herbicide tolerance, deposited as ATCC 203352, described in WO2000/026345), Event LLRICE601 (rice, herbicide tolerance, deposited as ATCC PTA-2600, described in US-A 2008-2289060

30 or WO2000/026356); Event LY038 (corn, quality trait, deposited as ATCC PTA-5623, described in US- A 2007-028322 or WO2005/061720); Event MIR162 (corn, insect control, deposited as PTA-8166, described in US-A 2009-300784 or WO2007/142840); Event MIR604 (corn, insect control, not deposited, described in US-A 2008-167456 or WO2005/103301); Event MON15985 (cotton, insect control, deposited as ATCC PTA-2516, described in US-A 2004-250317 or WO2002/100163); Event MON810

35 (corn, insect control, not deposited, described in US-A 2002-102582); Event MON863 (corn, insect control, deposited as ATCC PTA-2605, described in WO2004/011601 or US-A 2006-095986); Event MON87427 (corn, pollination control, deposited as ATCC PTA-7899, described in WO2011/062904); Event MON87460 (corn, stress tolerance, deposited as ATCC PTA-8910, described in WO2009/111263

or US-A 2011-0138504); Event MON87701 (soybean, insect control, deposited as ATCC PTA- 8194, described in US-A 2009-130071 or WO2009/064652); Event MON87705 (soybean, quality trait - herbicide tolerance, deposited as ATCC PTA-9241, described in US-A 2010-0080887 or WO2010/037016); Event MON87708 (soybean, herbicide tolerance, deposited as ATCC PTA-9670, described in WO2011/034704); Event MON87712 (soybean, yield, deposited as PTA-10296, described in WO2012/051199), Event MON87754 (soybean, quality trait, deposited as ATCC PTA-9385, described in WO2010/024976); Event MON87769 (soybean, quality trait, deposited as ATCC PTA- 8911, described in US-A 2011-0067141 or WO2009/102873); Event MON88017 (corn, insect control - herbicide tolerance, deposited as ATCC PTA-5582, described in US-A 2008-028482 or WO2005/059103); Event MON88913 (cotton, herbicide tolerance, deposited as ATCC PTA-4854, described in WO2004/072235 or US-A 2006-059590); Event MON88302 (oilseed rape, herbicide tolerance, deposited as PTA-10955, described in WO2011/153186), Event MON88701 (cotton, herbicide tolerance, deposited as PTA-11754, described in WO2012/134808), Event MON89034 (corn, insect control, deposited as ATCC PTA-7455, described in WO 07/140256 or US-A 2008-260932); Event MON89788 (soybean, herbicide tolerance, deposited as ATCC PTA-6708, described in US-A 2006-282915 or WO2006/130436); Event MS1 1 (oilseed rape, pollination control - herbicide tolerance, deposited as ATCC PTA-850 or PTA-2485, described in WO2001/031042); Event MS8 (oilseed rape, pollination control - herbicide tolerance, deposited as ATCC PTA-730, described in WO2001/041558 or US-A 2003-188347); Event NK603 (corn, herbicide tolerance, deposited as ATCC PTA-2478, described in US-A 2007-292854); Event PE-7 (rice, insect control, not deposited, described in WO2008/114282); Event RF3 (oilseed rape, pollination control - herbicide tolerance, deposited as ATCC PTA-730, described in WO2001/041558 or US-A 2003-188347); Event RT73 (oilseed rape, herbicide tolerance, not deposited, described in WO2002/036831 or US-A 2008-070260); Event SYHT0H2 / SYN-000H2-5 (soybean, herbicide tolerance, deposited as PTA-11226, described in WO2012/082548), Event T227-1 (sugar beet, herbicide tolerance, not deposited, described in WO2002/44407 or US-A 2009-265817); Event T25 (corn, herbicide tolerance, not deposited, described in US-A 2001-029014 or WO2001/051654); Event T304-40 (cotton, insect control - herbicide tolerance, deposited as ATCC PTA-8171, described in US-A 2010-077501 or WO2008/122406); Event T342-142 (cotton, insect control, not deposited, described in WO2006/128568); Event TC1507 (corn, insect control - herbicide tolerance, not deposited, described in US-A 2005-039226 or WO2004/099447); Event VIP1034 (corn, insect control - herbicide tolerance, deposited as ATCC PTA-3925, described in WO2003/052073), Event 32316 (corn, insect control-herbicide tolerance, deposited as PTA-11507, described in WO2011/084632), Event 4114 (corn, insect control-herbicide tolerance, deposited as PTA-11506, described in WO2011/084621), event EE-GM3 / FG72 (soybean, herbicide tolerance, ATCC Accession N° PTA-11041) optionally stacked with event EE-GM1/LL27 or event EE-GM2/LL55 (WO2011/063413A2), event DAS-68416-4 (soybean, herbicide tolerance, ATCC Accession N° PTA-10442, WO2011/066360A1), event DAS-68416-4 (soybean, herbicide tolerance, ATCC Accession N° PTA-10442, WO2011/066384A1), event DP-040416-8 (corn, insect control, ATCC Accession N° PTA-11508, WO2011/075593A1), event DP-043A47-3 (corn, insect control, ATCC Accession N° PTA-11509,

WO2011/075595A1), event DP- 004114-3 (corn, insect control, ATCC Accession N° PTA-11506, WO2011/084621A1), event DP-032316-8 (corn, insect control, ATCC Accession N° PTA-11507, WO2011/084632A1), event MON-88302-9 (oilseed rape, herbicide tolerance, ATCC Accession N° PTA-10955, WO2011/153186A1), event DAS-21606-3 (soybean, herbicide tolerance, ATCC Accession No. 5 PTA-11028, WO2012/033794A2), event MON-87712-4 (soybean, quality trait, ATCC Accession N° PTA-10296, WO2012/051199A2), event DAS-44406-6 (soybean, stacked herbicide tolerance, ATCC Accession N°. PTA-11336, WO2012/075426A1), event DAS-14536-7 (soybean, stacked herbicide tolerance, ATCC Accession N°. PTA-11335, WO2012/075429A1), event SYN-000H2-5 (soybean, herbicide tolerance, ATCC Accession N°. PTA-11226, WO2012/082548A2), event DP-061061-7 (oilseed 10 rape, herbicide tolerance, no deposit N° available, WO2012071039A1), event DP-073496-4 (oilseed rape, herbicide tolerance, no deposit N° available, US2012131692), event 8264.44.06.1 (soybean, stacked herbicide tolerance, Accession N° PTA-11336, WO2012075426A2), event 8291.45.36.2 (soybean, stacked herbicide tolerance, Accession N°. PTA-11335, WO2012075429A2), event SYHT0H2 (soybean, ATCC Accession N°. PTA-11226, WO2012/082548A2), event MON88701 (cotton, ATCC Accession N° 15 PTA-11754, WO2012/134808A1), event KK179-2 (alfalfa, ATCC Accession N° PTA-11833, WO2013/003558A1), event pDAB8264.42.32.1 (soybean, stacked herbicide tolerance, ATCC Accession N° PTA-11993, WO2013/010094A1), event MZDT09Y (corn, ATCC Accession N° PTA-13025, WO2013/012775A1).

Further, a list of such transgenic event(s) is provided by the United States Department of Agriculture's 20 (USDA) Animal and Plant Health Inspection Service (APHIS) and can be found on their website on the world wide web at aphis.usda.gov. For this application, the status of such list as it is/was on the filing date of this application, is relevant.

The genes/events which impart the desired traits in question may also be present in combinations with one another in the transgenic plants. Examples of transgenic plants which may be mentioned are the important 25 crop plants, such as cereals (wheat, rice, triticale, barley, rye, oats), maize, soya beans, potatoes, sugar beet, sugar cane, tomatoes, peas and other types of vegetable, cotton, tobacco, oilseed rape and also fruit plants (with the fruits apples, pears, citrus fruits and grapes), with particular emphasis being given to maize, soya beans, wheat, rice, potatoes, cotton, sugar cane, tobacco and oilseed rape. Traits which are particularly emphasized are the increased resistance of the plants to insects, arachnids, nematodes and 30 slugs and snails, as well as the increased resistance of the plants to one or more herbicides.

Commercially available examples of such plants, plant parts or plant seeds that may be treated with preference in accordance with the invention include commercial products, such as plant seeds, sold or distributed under the GENUITY®, DROUGHTGARD®, SMARTSTAX®, RIB COMPLETE®, ROUNDUP READY®, VT DOUBLE PRO®, VT TRIPLE PRO®, BOLLGARD II®, ROUNDUP 35 READY 2 YIELD®, YIELDGARD®, ROUNDUP READY® 2 XTEN^{DTM}, INTACTA RR2 PRO®, VISTIVE GOLD®, and/or XTENDFLEXTM trade names.

Crop protection – types of treatment

The treatment of the plants and plant parts with the compounds of the formula (I) is carried out directly or by action on their surroundings, habitat or storage space using customary treatment methods, for example by dipping, spraying, atomizing, irrigating, evaporating, dusting, fogging, broadcasting, foaming, painting, spreading-on, injecting, watering (drenching), drip irrigating and, in the case of propagation material, in particular in the case of seed, furthermore as a powder for dry seed treatment, a solution for liquid seed treatment, a water-soluble powder for slurry treatment, by incrusting, by coating with one or more coats, etc. It is furthermore possible to apply the compounds of the formula (I) by the ultra-low volume method or to inject the application form or the compound of the formula (I) itself into the soil.

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- 10 A preferred direct treatment of the plants is foliar application, i.e. the compounds of the formula (I) are applied to the foliage, where treatment frequency and the application rate should be adjusted according to the level of infestation with the pest in question.

In the case of systemically active compounds, the compounds of the formula (I) also access the plants via the root system. The plants are then treated by the action of the compounds of the formula (I) on the habitat of the plant. This may be done, for example, by drenching, or by mixing into the soil or the nutrient solution, i.e. the locus of the plant (e.g. soil or hydroponic systems) is impregnated with a liquid form of the compounds of the formula (I), or by soil application, i.e. the compounds of the formula (I) according to the invention are introduced in solid form (e.g. in the form of granules) into the locus of the plants, or by drip application (often also referred to as "chemigation"), i.e. the liquid application of the compounds of the formula (I) according to the invention from surface or sub-surface driplines over a certain period of time together with varying amounts of water at defined locations in the vicinity of the plants. In the case of paddy rice crops, this can also be done by metering the compound of the formula (I) in a solid application form (for example as granules) into a flooded paddy field.

Digital Technologies

- 25 The compounds of the invention can be used in combination with models e.g. embedded in computer programs for site specific crop management, satellite farming, precision farming or precision agriculture. Such models support the site specific management of agricultural sites with data from various sources such as soils, weather, crops (e.g. type, growth stage, plant health), weeds (e.g. type, growth stage), diseases, pests, nutrients, water, moisture, biomass, satellite data, yield etc. with the purpose to optimize profitability, sustainability and protection of the environment. In particular, such models can help to optimize agronomical decisions, control the precision of pesticide applications and record the work performed.
- 30

As an example, the compounds of the invention can be applied to a crop plant according to an appropriate dose regime if a model models the development of a pest and calculates that a threshold has been reached for which it is recommendable to apply the compound of the invention to the crop plant.

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Commercially available systems which include agronomic models are e.g. FieldScripts™ from The Climate Corporation, Xarvio™ from BASF, AGLogic™ from John Deere, etc.

The compounds of the invention can also be used in combination with smart spraying equipment such as e.g. spot spraying or precision spraying equipment attached to or housed within a farm vehicle such as a tractor, robot, helicopter, airplane, unmanned aerial vehicle (UAV) such as a drone, etc. Such an equipment usually includes input sensors (such as e.g. a camera) and a processing unit configured to analyze the input data and configured to provide a decision based on the analysis of the input data to apply the compound of the invention to the crop plants (respectively the weeds) in a specific and precise manner. The use of such smart spraying equipment usually also requires positions systems (e.g. GPS receivers) to localize recorded data and to guide or to control farm vehicles; geographic information systems (GIS) to represent the information on intelligible maps, and appropriate farm vehicles to perform the required farm action such as the spraying.

In an example, pests can be detected from imagery acquired by a camera. In an example the pests can be identified and/or classified based on that imagery. Such identification and/ classification can make use of image processing algorithms. Such image processing algorithms can utilize machine learning algorithms, such as trained neural networks, decision trees and utilize artificial intelligence algorithms. In this manner, the compounds described herein can be applied only where needed.

Treatment of seed

The control of animal pests by treating the seed of plants has been known for a long time and is the subject of continuous improvements. However, the treatment of seed entails a series of problems which cannot always be solved in a satisfactory manner. Thus, it is desirable to develop methods for protecting the seed and the germinating plant which dispense with, or at least reduce considerably, the additional application of pesticides during storage, after sowing or after emergence of the plants. It is furthermore desirable to optimize the amount of active compound employed in such a way as to provide optimum protection for the seed and the germinating plant from attack by animal pests, but without damaging the plant itself by the active compound employed. In particular, methods for the treatment of seed should also take into consideration the intrinsic insecticidal or nematicidal properties of pest-resistant or -tolerant transgenic plants in order to achieve optimum protection of the seed and also the germinating plant with a minimum of pesticides being employed.

The present invention therefore in particular also relates to a method for the protection of seed and germinating plants, from attack by pests, by treating the seed with one of the compounds of the formula (I). The method according to the invention for protecting seed and germinating plants against attack by pests furthermore comprises a method where the seed is treated simultaneously in one operation or sequentially with a compound of the formula (I) and a mixing component. It also comprises a method where the seed is treated at different times with a compound of the formula (I) and a mixing component.

The invention likewise relates to the use of the compounds of the formula (I) for the treatment of seed for protecting the seed and the resulting plant from animal pests.

Furthermore, the invention relates to seed which has been treated with a compound of the formula (I) according to the invention so as to afford protection from animal pests. The invention also relates to seed
5 which has been treated simultaneously with a compound of the formula (I) and a mixing component. The invention furthermore relates to seed which has been treated at different times with a compound of the formula (I) and a mixing component. In the case of seed which has been treated at different points in time with a compound of the formula (I) and a mixing component, the individual substances may be present on the seed in different layers. Here, the layers comprising a compound of the formula (I) and mixing
10 components may optionally be separated by an intermediate layer. The invention also relates to seed where a compound of the formula (I) and a mixing component have been applied as component of a coating or as a further layer or further layers in addition to a coating.

Furthermore, the invention relates to seed which, after the treatment with a compound of the formula (I), is subjected to a film-coating process to prevent dust abrasion on the seed.

15 One of the advantages encountered with a systemically acting compound of the formula (I) is the fact that, by treating the seed, not only the seed itself but also the plants resulting therefrom are, after emergence, protected against animal pests. In this manner, the immediate treatment of the crop at the time of sowing or shortly thereafter can be dispensed with.

It has to be considered a further advantage that by treatment of the seed with a compound of the formula
20 (I), germination and emergence of the treated seed may be enhanced.

It is likewise to be considered advantageous that compounds of the formula (I) can be used in particular also for transgenic seed.

Furthermore, compounds of the formula (I) can be employed in combination with compositions or compounds of signalling technology, leading to better colonization by symbionts such as, for example,
25 rhizobia, mycorrhizae and/or endophytic bacteria or fungi, and/or to optimized nitrogen fixation.

The compounds of the formula (I) are suitable for protection of seed of any plant variety which is used in agriculture, in the greenhouse, in forests or in horticulture. In particular, this takes the form of seed of cereals (for example wheat, barley, rye, millet and oats), corn, cotton, soya beans, rice, potatoes, sunflowers, coffee, tobacco, canola, oilseed rape, beets (for example sugarbeets and fodder beets), peanuts,
30 vegetables (for example tomatoes, cucumbers, bean, cruciferous vegetables, onions and lettuce), fruit plants, lawns and ornamental plants. The treatment of the seed of cereals (such as wheat, barley, rye and oats), maize, soya beans, cotton, canola, oilseed rape, vegetables and rice is of particular importance.

As already mentioned above, the treatment of transgenic seed with a compound of the formula (I) is also of particular importance. This takes the form of seed of plants which, as a rule, comprise at least one heterologous gene which governs the expression of a polypeptide with in particular insecticidal and/or nematocidal properties. The heterologous genes in transgenic seed can originate from microorganisms such as *Bacillus*, *Rhizobium*, *Pseudomonas*, *Serratia*, *Trichoderma*, *Clavibacter*, *Glomus* or *Gliocladium*. The present invention is particularly suitable for the treatment of transgenic seed which comprises at least one heterologous gene originating from *Bacillus* sp. It is particularly preferably a heterologous gene derived from *Bacillus thuringiensis*.

In the context of the present invention, the compound of the formula (I) is applied to the seed. Preferably, the seed is treated in a state in which it is stable enough to avoid damage during treatment. In general, the seed may be treated at any point in time between harvest and sowing. The seed usually used has been separated from the plant and freed from cobs, shells, stalks, coats, hairs or the flesh of the fruits. For example, it is possible to use seed which has been harvested, cleaned and dried down to a moisture content which allows storage. Alternatively, it is also possible to use seed which, after drying, has been treated with, for example, water and then dried again, for example priming. In the case of rice seed, it is also possible to use seed which has been soaked, for example in water to a certain stage of the rice embryo ('pigeon breast stage'), stimulating the germination and a more uniform emergence.

When treating the seed, care must generally be taken that the amount of the compound of the formula (I) applied to the seed and/or the amount of further additives is chosen in such a way that the germination of the seed is not adversely affected, or that the resulting plant is not damaged. This must be ensured particularly in the case of active compounds which can exhibit phytotoxic effects at certain application rates.

In general, the compounds of the formula (I) are applied to the seed in a suitable formulation. Suitable formulations and processes for seed treatment are known to the person skilled in the art.

The compounds of the formula (I) can be converted to the customary seed dressing formulations, such as solutions, emulsions, suspensions, powders, foams, slurries or other coating compositions for seed, and also ULV formulations.

These formulations are prepared in a known manner, by mixing the compounds of the formula (I) with customary additives such as, for example, customary extenders and also solvents or diluents, colorants, wetting agents, dispersants, emulsifiers, antifoams, preservatives, secondary thickeners, adhesives, gibberellins and also water.

Colorants which may be present in the seed-dressing formulations which can be used in accordance with the invention are all colorants which are customary for such purposes. It is possible to use either pigments, which are sparingly soluble in water, or dyes, which are soluble in water. Examples include the dyes known by the names Rhodamine B, C.I. Pigment Red 112 and C.I. Solvent Red 1.

Useful wetting agents which may be present in the seed dressing formulations usable in accordance with the invention are all substances which promote wetting and which are conventionally used for the formulation of agrochemically active compounds. Preference is given to using alkylnaphthalenesulphonates, such as diisopropyl- or diisobutylnaphthalenesulphonates.

- 5 Useful dispersants and/or emulsifiers which may be present in the seed dressing formulations usable in accordance with the invention are all nonionic, anionic and cationic dispersants conventionally used for the formulation of active agrochemical ingredients. Preference is given to using nonionic or anionic dispersants or mixtures of nonionic or anionic dispersants. Suitable nonionic dispersants include in particular ethylene oxide/propylene oxide block polymers, alkylphenol polyglycol ethers and
- 10 tristyrylphenol polyglycol ethers, and the phosphated or sulphated derivatives thereof. Suitable anionic dispersants are in particular lignosulphonates, polyacrylic acid salts and arylsulphonate/formaldehyde condensates.

- Antifoams which may be present in the seed dressing formulations usable in accordance with the invention are all foam-inhibiting substances conventionally used for the formulation of active agrochemical
- 15 ingredients. Preference is given to using silicone antifoams and magnesium stearate.

Preservatives which may be present in the seed dressing formulations usable in accordance with the invention are all substances usable for such purposes in agrochemical compositions. Examples include dichlorophene and benzyl alcohol hemiformal.

- Secondary thickeners which may be present in the seed dressing formulations usable in accordance with
- 20 the invention are all substances which can be used for such purposes in agrochemical compositions. Cellulose derivatives, acrylic acid derivatives, xanthan, modified clays and finely divided silica are preferred.

- Adhesives which may be present in the seed dressing formulations usable in accordance with the invention are all customary binders usable in seed dressing products. Polyvinylpyrrolidone, polyvinyl acetate,
- 25 polyvinyl alcohol and tylose may be mentioned as being preferred.

Gibberellins which can be present in the seed-dressing formulations which can be used in accordance with the invention are preferably the gibberellins A1, A3 (= gibberellic acid), A4 and A7; gibberellic acid is especially preferably used. The gibberellins are known (cf. R. Wegler "Chemie der Pflanzenschutz- und Schädlingsbekämpfungsmittel", vol. 2, Springer Verlag, 1970, pp. 401-412).

- 30 The seed dressing formulations usable in accordance with the invention can be used to treat a wide variety of different kinds of seed either directly or after prior dilution with water. For instance, the concentrates or the preparations obtainable therefrom by dilution with water can be used to dress the seed of cereals, such as wheat, barley, rye, oats, and triticale, and also the seed of maize, rice, oilseed rape, peas, beans, cotton, sunflowers, soya beans and beets, or else a wide variety of different vegetable seed. The seed

dressing formulations usable in accordance with the invention, or the dilute use forms thereof, can also be used to dress seed of transgenic plants.

For treatment of seed with the seed dressing formulations usable in accordance with the invention, or the use forms prepared therefrom by adding water, all mixing units usable customarily for the seed dressing
5 are useful. Specifically, the procedure in the seed dressing is to place the seed into a mixer, operated batch-wise or continuously, to add the particular desired amount of seed dressing formulations, either as such or after prior dilution with water, and to mix everything until the formulation is distributed homogeneously on the seed. If appropriate, this is followed by a drying operation.

The application rate of the seed dressing formulations usable in accordance with the invention can be
10 varied within a relatively wide range. It is guided by the particular content of the compounds of the formula (I) in the formulations and by the seed. The application rates of the compound of the formula (I) are generally between 0.001 and 50 g per kilogram of seed, preferably between 0.01 and 15 g per kilogram of seed.

Animal health

15 In the animal health field, i.e. in the field of veterinary medicine, the compounds of the formula (I) are active against animal parasites, in particular ectoparasites or endoparasites. The term endoparasite includes in particular helminths and protozoae, such as coccidia. Ectoparasites are typically and preferably arthropods, in particular insects or acarids.

In the field of veterinary medicine the compounds of the formula (I) are suitable, with favourable toxicity
20 in warm blooded animals, for controlling parasites which occur in animal breeding and animal husbandry in livestock, breeding, zoo, laboratory, experimental and domestic animals. They are active against all or specific stages of development of the parasites.

Agricultural livestock include, for example, mammals, such as, sheep, goats, horses, donkeys, camels, buffaloes, rabbits, reindeers, fallow deers, and in particular cattle and pigs; or poultry, such as turkeys,
25 ducks, geese, and in particular chickens; or fish or crustaceans, e.g. in aquaculture; or, as the case may be, insects such as bees.

Domestic animals include, for example, mammals, such as hamsters, guinea pigs, rats, mice, chinchillas, ferrets or in particular dogs, cats; cage birds; reptiles; amphibians or aquarium fish.

According to a particular embodiment, the compounds of the formula (I) are administered to mammals.

30 According to another particular embodiment, the compounds of the formula (I) are administered to birds, namely cage birds or in particular poultry.

By using the compounds of the formula (I) to control animal parasites, it is intended to reduce or prevent illness, cases of deaths and performance reductions (in the case of meat, milk, wool, hides, eggs, honey and the like), so that more economical and simpler animal keeping is made possible and better animal well-being is achievable.

- 5 The term "control" or "controlling", as used herein with regard to the animal health field, means that the compounds of the formula (I) are effective in reducing the incidence of the respective parasite in an animal infected with such parasites to innocuous levels. More specifically, "controlling", as used herein, means that the compounds of the formula (I) are effective in killing the respective parasite, inhibiting its growth, or inhibiting its proliferation.
- 10 Exemplary arthropods include, without any limitation
- from the order of the Anoplurida, for example, *Haematopinus* spp., *Linognathus* spp., *Pediculus* spp., *Phthirus* spp., *Solenopotes* spp.;
- from the order of the Mallophagida and the suborders Amblycerina and Ischnocerina, for example *Bovicola* spp., *Damalina* spp., *Felicola* spp., *Lepikentron* spp., *Menopon* spp., *Trichodectes* spp.,
- 15 *Trimenopon* spp., *Trinoton* spp., *Werneckiella* spp.;
- from the order of the Diptera and the suborders Nematocerina and Brachycerina, for example *Aedes* spp., *Anopheles* spp., *Atylotus* spp., *Braula* spp., *Calliphora* spp., *Chrysomyia* spp., *Chrysops* spp., *Culex* spp., *Culicoides* spp., *Eusimulium* spp., *Fannia* spp., *Gasterophilus* spp., *Glossina* spp., *Haematobia* spp., *Haematopota* spp., *Hippobosca* spp., *Hybomitra* spp., *Hydrotaea* spp., *Hypoderma* spp., *Lipoptena* spp.,
- 20 *Lucilia* spp., *Lutzomyia* spp., *Melophagus* spp., *Morellia* spp., *Musca* spp., *Odagmia* spp., *Oestrus* spp., *Philipomyia* spp., *Phlebotomus* spp., *Rhinoestrus* spp., *Sarcophaga* spp., *Simulium* spp., *Stomoxys* spp., *Tabanus* spp., *Tipula* spp., *Wilhelmia* spp., *Wohlfahrtia* spp.
- from the order of the Siphonapterida, for example *Ceratophyllus* spp.; *Ctenocephalides* spp., *Pulex* spp., *Tunga* spp., *Xenopsylla* spp.;
- 25 from the order of the Heteropterida, for example *Cimex* spp., *Panstrongylus* spp., *Rhodnius* spp., *Triatoma* spp.; as well as nuisance and hygiene pests from the order of the Blattarida.
- Further, among the arthropods, the following acari may be mentioned by way of example, without any limitation:
- from the subclass of the Acari (Acarina) and the order of the Metastigmata, for example, from the family
- 30 of argasidae like *Argas* spp., *Ornithodoros* spp., *Otobius* spp., from the family of Ixodidae like *Amblyomma* spp., *Dermacentor* spp., *Haemaphysalis* spp., *Hyalomma* spp., *Ixodes* spp., *Rhipicephalus* (*Boophilus*) spp., *Rhipicephalus* spp. (the original genus of multi host ticks); from the order of mesostigmata like *Dermanyssus* spp., *Ornithonyssus* spp., *Pneumonyssus* spp., *Raillietia* spp.,

Sternostoma spp., Tropilaelaps spp., Varroa spp.; from the order of the Actiniedida (Prostigmata), for example Acarapis spp., Cheyletiella spp., Demodex spp., Listrophorus spp., Myobia spp., Neotrombicula spp., Ornithocheyletia spp., Psorergates spp., Trombicula spp.; and from the order of the Acaridida (Astigmata), for example Acarus spp., Caloglyphus spp., Chorioptes spp., Cytodites spp., Hypodectes spp., Knemidocoptes spp., Laminosioptes spp., Notoedres spp., Otodectes spp., Psoroptes spp., Pterolichus spp., Sarcoptes spp., Trixacarus spp., Tyrophagus spp.

Exemplary parasitic protozoa include, without any limitation:

Mastigophora (Flagellata) such as:

Metamonada: from the order Diplomonadida, for example, Giardia spp., Spironucleus spp.

10 Parabasala: from the order Trichomonadida, for example, Histomonas spp., Pentatrichomonas spp., Tetratrichomonas spp., Trichomonas spp., Tritrichomonas spp.

Euglenozoa: from the order Trypanosomatida, for example, Leishmania spp., Trypanosoma spp.

Sarcomastigophora (Rhizopoda), such as Entamoebidae, for example, Entamoeba spp., Centramoebidae, for example, Acanthamoeba sp., Euamoebidae, e.g. Hartmanella sp.

15 Alveolata such as Apicomplexa (Sporozoa): e.g. Cryptosporidium spp.; from the order Eimeriida, for example, Besnoitia spp., Cystoisospora spp., Eimeria spp., Hammondia spp., Isospora spp., Neospora spp., Sarcocystis spp., Toxoplasma spp.; from the order Adeleida e.g. Hepatozoon spp., Klossiella spp.; from the order Haemosporida e.g. Leucocytozoon spp., Plasmodium spp.; from the order Piroplasmida e.g. Babesia spp., Ciliophora spp., Echinozoon spp., Theileria spp.; from the order Vesiculiferida e.g. Balantidium spp., Buxtonella spp.

20 Microspora such as Encephalitozoon spp., Enterocytozoon spp., Globidium spp., Nosema spp., and furthermore, e.g. Myxozoa spp.

Helminths pathogenic for humans or animals include, for example, acanthocephala, nematodes, pentastoma and platyhelmintha (e.g. monogenea, cestodes and trematodes).

25 Exemplary helminths include, without any limitation:

Monogenea: e.g.: Dactylogyrus spp., Gyrodactylus spp., Microbothrium spp., Polystoma spp., Troglocephalus spp.

Cestodes: from the order of the Pseudophyllidea, for example: Bothridium spp., Diphyllbothrium spp., Diplogonoporus spp., Ichthyobothrium spp., Ligula spp., Schistocephalus spp., Spirometra spp.

from the order of the Cyclophyllida, for example: *Andyra* spp., *Anoplocephala* spp., *Avitellina* spp., *Bertiella* spp., *Cittotaenia* spp., *Davainea* spp., *Diorchis* spp., *Diplopylidium* spp., *Dipylidium* spp., *Echinococcus* spp., *Echinocotyle* spp., *Echinolepis* spp., *Hydatigera* spp., *Hymenolepis* spp., *Joyeuxiella* spp., *Mesocestoides* spp., *Moniezia* spp., *Paranoplocephala* spp., *Raillietina* spp., *Stilesia* spp., *Taenia* spp., *Thysaniezia* spp., *Thysanosoma* spp.

Trematodes: from the class of the Digenea, for example: *Austrobilharzia* spp., *Brachylaima* spp., *Calicophoron* spp., *Catantropis* spp., *Clonorchis* spp., *Collyriclum* spp., *Cotylophoron* spp., *Cyclocoelum* spp., *Dicrocoelium* spp., *Diplostomum* spp., *Echinochasmus* spp., *Echinoparyphium* spp., *Echinostoma* spp., *Eurytrema* spp., *Fasciola* spp., *Fasciolides* spp., *Fasciolopsis* spp., *Fischoederius* spp., *Gastrothylacus* spp., *Gigantobilharzia* spp., *Gigantocotyle* spp., *Heterophyes* spp., *Hypoderaeum* spp., *Leucochloridium* spp., *Metagonimus* spp., *Metorchis* spp., *Nanophyetus* spp., *Notocotylus* spp., *Opisthorchis* spp., *Ornithobilharzia* spp., *Paragonimus* spp., *Paramphistomum* spp., *Plagiorchis* spp., *Posthodiplostomum* spp., *Prosthogonimus* spp., *Schistosoma* spp., *Trichobilharzia* spp., *Troglostrongylus* spp., *Typhlocoelum* spp.

Nematodes: from the order of the Trichinellida, for example: *Capillaria* spp., *Eucoleus* spp., *Paracapillaria* spp., *Trichinella* spp., *Trichomosoides* spp., *Trichuris* spp.

from the order of the Tylenchida, for example: *Micronema* spp., *Parastrongyloides* spp., *Strongyloides* spp.

from the order of the Rhabditina, for example: *Aelurostrongylus* spp., *Amidostomum* spp., *Ancylostoma* spp., *Angiostrongylus* spp., *Bronchonema* spp., *Bunostomum* spp., *Chabertia* spp., *Cooperia* spp., *Cooperioides* spp., *Crenosoma* spp., *Cyathostomum* spp., *Cyclococercus* spp., *Cyclodontostomum* spp., *Cylicocyclus* spp., *Cylicostephanus* spp., *Cylindropharynx* spp., *Cystocaulus* spp., *Dictyocaulus* spp., *Elaphostrongylus* spp., *Filaroides* spp., *Globocephalus* spp., *Graphidium* spp., *Gyaloccephalus* spp., *Haemonchus* spp., *Heligmosomoides* spp., *Hyostrongylus* spp., *Marshallagia* spp., *Metastrongylus* spp., *Muellerius* spp., *Necator* spp., *Nematodirus* spp., *Neostrongylus* spp., *Nippostrongylus* spp., *Obeliscoides* spp., *Oesophagodontus* spp., *Oesophagostomum* spp., *Ollulanus* spp., *Ornithostrongylus* spp., *Oslerus* spp., *Ostertagia* spp., *Paracooperia* spp., *Paracrenosoma* spp., *Parafilaroides* spp., *Parelaphostrongylus* spp., *Pneumocaulus* spp., *Pneumostrongylus* spp., *Poteriostomum* spp., *Protostrongylus* spp., *Spicocaulus* spp., *Stephanurus* spp., *Strongylus* spp., *Syngamus* spp., *Teladorsagia* spp., *Trichonema* spp., *Trichostrongylus* spp., *Triodontophorus* spp., *Troglostrongylus* spp., *Uncinaria* spp.

from the order of the Spirurida, for example: *Acanthocheilonema* spp., *Anisakis* spp., *Ascaridia* spp., *Ascaris* spp., *Ascarops* spp., *Aspiculuris* spp., *Baylisascaris* spp., *Brugia* spp., *Cercopithifilaria* spp., *Crassicauda* spp., *Dipetalonema* spp., *Dirofilaria* spp., *Dracunculus* spp., *Draschia* spp., *Enterobius* spp., *Filaria* spp., *Gnathostoma* spp., *Gongylonema* spp., *Habronema* spp., *Heterakis* spp., *Litomosoides* spp., *Loa* spp., *Onchocerca* spp., *Oxyuris* spp., *Parabronema* spp., *Parafilaria* spp., *Parascaris* spp., *Passalurus* spp., *Physaloptera* spp., *Probstmayria* spp., *Pseudofilaria* spp., *Setaria* spp., *Skjrabinema* spp., *Spirocerca*

spp., *Stephanofilaria* spp., *Strongyluris* spp., *Syphacia* spp., *Thelazia* spp., *Toxascaris* spp., *Toxocara* spp., *Wuchereria* spp.

Acantocephala: from the order of the *Oligacanthorhynchida*, for example: *Macracanthorhynchus* spp., *Prosthenorchis* spp.; from the order of the *Moniliformida*, for example: *Moniliformis* spp.

- 5 from the order of the *Polymorphida*, for example: *Filicollis* spp.; from the order of the *Echinorhynchida*, for example: *Acanthocephalus* spp., *Echinorhynchus* spp., *Leptorhynchoides* spp.

Pentastoma: from the order of the *Porocephalida*, for example: *Linguatula* spp.

- In the veterinary field and in animal keeping, the administration of the compounds of the formula (I) is carried out by methods generally known in the art, such as enterally, parenterally, dermally or nasally, in the form of suitable preparations. Administration can be carried out prophylactically, methaphylactically or therapeutically.
- 10

Thus, one embodiment of the present invention refers to the compounds of the formula (I) for use as a medicament.

Another aspect refers to the compounds of the formula (I) for use as an antiendoparasitical agent.

- 15 Another particular aspect refers to the compounds of the formula (I) for use as a anthelmintic agent, more particular for use as a nematicidal agent, a platyhelminthicidal agent, an acanthocephalidal agent, or a pentastomicidal agent.

Another particular aspect refers to the compounds of the formula (I) for use as an antiprotozoal agent.

- Another aspect refers to the compounds of the formula (I) for use as an antiectoparasitical agent, in particular an arthropodicidal agent, more particular an insecticidal agent or acaricidal agent.
- 20

- Further aspects of the invention are veterinary formulations, comprising an effective amount of at least one compound of the formula (I) and at least one of the following: pharmaceutically acceptable excipient (e.g. solid or liquid diluents), pharmaceutically acceptable auxiliary (e.g. surfactants), in particular a pharmaceutically acceptable excipient and/or pharmaceutically acceptable auxiliary which is normally used in veterinary formulations.
- 25

A related aspect of the invention is a method for preparing a veterinary formulation as described herein, comprising the step of mixing at least one compound of the formula (I) with pharmaceutically acceptable excipients and/or auxiliaries, in particular with pharmaceutically acceptable excipients and/or auxiliaries which are normally used in veterinary formulations.

- 30 Another particular aspect of the invention are veterinary formulations, selected from the group of ectoparasiticidal and endoparasiticidal formulations, more particular selected from the group of

anthelmintic, antiprotozoal, and arthropodicidal formulations, even more particular selected from the group of nematocidal, platyhelminthicidal, acanthocephalicidal, pentastomicidal, insecticidal, and acaricidal formulations, in accordance with the mentioned aspects, as well as their methods for preparation.

- 5 Another aspect refers to a method for treatment of a parasitic infection, in particular an infection by a parasite selected from the group of ectoparasites and endoparasites mentioned herein, by applying an effective amount of a compound of the formula (I) to an animal, in particular a non-human animal, in need thereof.

- 10 Another aspect refers to a method for treatment of a parasitic infection, in particular an infection by a parasite selected from the group of ectoparasites and endoparasites mentioned herein, by applying a veterinary formulation as defined herein to an animal, in particular a non-human animal, in need thereof.

Another aspect refers to the use of the compounds of the formula (I) in the treatment of a parasitic infection, in particular an infection by a parasite selected from the group of ectoparasites and endoparasites mentioned herein, in an animal, in particular a non-human animal.

- 15 In the present context of the animal health or veterinary field, the term "treatment" includes prophylactic, metaphylactic or therapeutical treatment.

In a particular embodiment, mixtures of at least one compound of the formula (I) with other active ingredients, particularly with endo- and ectoparasiticides, for the veterinary field are provided herewith.

- 20 In the field of animal health "mixture" not only means that two (or more) different active ingredients are formulated in a joint formulation and are accordingly applied together but also refers to products which comprise separate formulations for each active compound. Accordingly, if more than two active compounds are to be applied, all active compounds may be formulated in a joint formulation or all active compounds may be formulated in separate formulations; also feasible are mixed forms where some of the active compounds are formulated jointly and some of the active compounds are formulated separately.
- 25 Separate formulations allow the separate or successive application of the active compounds in question.

The active compounds specified herein by their common names are known and described, for example, in the Pesticide Manual (see above) or can be searched in the internet (e.g. <http://www.alanwood.net/pesticides>).

- 30 Exemplary active ingredients from the group of ectoparasiticides, as mixing partners, include, without limitation insecticides and acaricides listed in detail above. Further active ingredients which may be used are listed below following the aforementioned classification which is based on the current IRAC Mode of Action Classification Scheme: (1) Acetylcholinesterase (AChE) inhibitors; (2) GABA-gated chloride channel blockers; (3) Sodium channel modulators; (4) Nicotinic acetylcholine receptor (nAChR)

competitive modulators; (5) Nicotinic acetylcholine receptor (nAChR) allosteric modulators; (6) Glutamate-gated chloride channel (GluCl) allosteric modulators; (7) Juvenile hormone mimics; (8) Miscellaneous non-specific (multi-site) inhibitors; (9) Modulators of Chordotonal Organs; (10) Mite growth inhibitors; (12) Inhibitors of mitochondrial ATP synthase, such as, ATP disruptors; (13) 5 Uncouplers of oxidative phosphorylation via disruption of the proton gradient; (14) Nicotinic acetylcholine receptor channel blockers; (15) Inhibitors of chitin biosynthesis, type 0; (16) Inhibitors of chitin biosynthesis, type 1; (17) Moulting disruptor (in particular for Diptera, i.e. dipterans); (18) Ecdysone receptor agonists; (19) Octopamine receptor agonists; (21) Mitochondrial complex I electron transport inhibitors; (25) Mitochondrial complex II electron transport inhibitors; (20) Mitochondrial complex III 10 electron transport inhibitors; (22) Voltage-dependent sodium channel blockers; (23) Inhibitors of acetyl CoA carboxylase; (28) Ryanodine receptor modulators; (30) GABA-gated chloride channel allosteric modulators.

Active compounds with unknown or non-specific mode of action, e.g., fentrifanil, fenoxacrim, cycloprene, chlorobenzilate, chlordimeform, flubenzimine, dicyclanil, amidoflumet, quinomethionate, triarathene, 15 clothiazoben, tetrasul, potassium oleate, petroleum, metoxadiazone, gossyplure, flutenzin, bromopropylate, cryolite;

Compounds from other classes, e.g. butacarb, dimetilan, cloethocarb, phosphocarb, pirimiphos (-ethyl), parathion (-ethyl), methacrifos, isopropyl o-salicylate, trichlorfon, tigolaner, sulprofos, propaphos, sebufos, pyridathion, prothoate, dichlofenthion, demeton-S-methylsulphone, isazofos, cyanofenphos, 20 dialifos, carbophenothion, autathiofos, aromfenvinfos (-methyl), azinphos (-ethyl), chlorpyrifos (-ethyl), fosmethilan, iodofenphos, dioxabenzofos, formothion, fonofos, flupyrazofos, fensulfothion, etrimfos;

organochlorines, e.g. camphechlor, lindane, heptachlor; or phenylpyrazoles, e.g. acetoprole, pyrafluprole, pyriprole, vanilprole, sisapronil; or isoxazolines, e.g. sarolaner, afoxolaner, lotilaner, fluralaner;

pyrethroids, e.g. (cis-, trans-), metofluthrin, profluthrin, flufenprox, flubrocycrinate, fubfenprox, 25 fenfluthrin, protrifenbute, pyresmethrin, RU15525, tallethrin, cis-resmethrin, heptafluthrin, , bioethanomethrin, biopermethrin, fenpyrithrin, cis-cypermethrin, cis-permethrin, cloxythrin, cyhalothrin (lambda-), chlovaporthrin, or halogenated carbonhydrogen compounds (HCHs),

neonicotinoids, e.g. nithiazine

dicloromezotiaz, triflumezopyrim

30 macrocyclic lactones, e.g. nemadectin, ivermectin, latidectin, moxidectin, selamectin, eprinomectin, doramectin, emamectin benzoate; milbemycin oxime

triprene, epofenonane, diofenolan;

Biologicals, hormones or pheromones, for example natural products, e.g. thuringiensin, codlemone or neem components

dinitrophenols, e.g. dinocap, dinobuton, binapacryl;

benzoylureas, e.g. fluazuron, penfluron,

- 5 amidine derivatives, e.g. chlormebuform, cymiazole, demiditraz

Bee hive varroa acaricides, for example organic acids, e.g. formic acid, oxalic acid.

Exemplary active ingredients from the group of endoparasitocides, as mixing partners, include, without limitation, anthelmintically active compounds and antiprotozoal active compounds.

- 10 Anthelmintically active compounds, including, without limitation, the following nematocidally, trematocidally and/or cestocidally active compounds:

from the class of macrocyclic lactones, for example: eprinomectin, abamectin, nemadectin, moxidectin, doramectin, selamectin, lepipsectin, latidectin, milbemectin, ivermectin, emamectin, milbemycin;

- 15 from the class of benzimidazoles and probenzimidazoles, for example: oxibendazole, mebendazole, triclabendazole, thiophanate, parbendazole, oxfendazole, netobimin, fenbendazole, febantel, thiabendazole, cyclobendazole, cambendazole, albendazole-sulphoxide, albendazole, flubendazole;

from the class of depsipeptides, preferably cyclic depsipeptides, in particular 24-membered cyclic depsipeptides, for example: emodepside, PF1022A;

from the class of tetrahydropyrimidines, for example: morantel, pyrantel, oxantel;

from the class of imidazothiazoles, for example: butamisol, levamisole, tetramisol;

- 20 from the class of aminophenylamidines, for example: amidantel, deacylated amidantel (dAMD), tribendimidine;

from the class of aminoacetonitriles, for example: monepantel;

from the class of paraherquamides, for example: paraherquamide, derquantel;

- 25 from the class of salicylanilides, for example: tribromsalan, bromoxanide, brotlanide, clioxanide, closantel, niclosamide, oxyclozanide, rafoxanide;

from the class of substituted phenols, for example: nitroxylin, bithionol, disophenol, hexachlorophene, niclofolan, meniclopholan;

from the class of organophosphates, for example: trichlorfon, naphthalofos, dichlorvos/DDVP, crufomate, coumaphos, haloxon;

from the class of piperazinones / quinolines, for example: praziquantel, epsiprantel;

from the class of piperazines, for example: piperazine, hydroxyzine;

- 5 from the class of tetracyclines, for example: tetracyclin, chlorotetracycline, doxycyclin, oxytetracyclin, rolitetracyclin;

from diverse other classes, for example: bunamidine, niridazole, resorantel, omphalotin, oltipraz, nitroscanate, nitroxy Nile, oxamniquine, mirasan, miracil, lucanthone, hycanthone, hetolin, emetine, diethylcarbamazine, dichlorophen, diamfenetide, clonazepam, bephenium, amoscanate, clorsulon.

- 10 Antiprotozoal active compounds, including, without limitation, the following active compounds:

from the class of triazines, for example: diclazuril, ponazuril, letrazuril, toltrazuril;

from the class of polyether ionophore, for example: monensin, salinomycin, maduramicin, narasin;

from the class of macrocyclic lactones, for example: milbemycin, erythromycin;

from the class of quinolones, for example: enrofloxacin, pradofloxacin;

- 15 from the class of quinines, for example: chloroquine;

from the class of pyrimidines, for example: pyrimethamine;

from the class of sulfonamides, for example: sulfaquinoxaline, trimethoprim, sulfaclozin;

from the class of thiamines, for example: amprolium;

from the class of lincosamides, for example: clindamycin;

- 20 from the class of carbanilides, for example: imidocarb;

from the class of nitrofurans, for example: nifurtimox;

from the class of quinazolinone alkaloids, for example: halofuginon;

from diverse other classes, for example: oxamniquin, paromomycin;

- 25 from the class of vaccines or antigens from microorganisms, for example: Babesia canis rossi, Eimeria tenella, Eimeria praecox, Eimeria necatrix, Eimeria mitis, Eimeria maxima, Eimeria brunetti, Eimeria acervulina, Babesia canis vogeli, Leishmania infantum, Babesia canis canis, Dictyocaulus viviparus.

All named mixing partners can, if their functional groups enable this, optionally form salts with suitable bases or acids.

Vector control

5 The compounds of the formula (I) can also be used in vector control. For the purpose of the present invention, a vector is an arthropod, in particular an insect or arachnid, capable of transmitting pathogens such as, for example, viruses, worms, single-cell organisms and bacteria from a reservoir (plant, animal, human, etc.) to a host. The pathogens can be transmitted either mechanically (for example trachoma by non-stinging flies) to a host, or by injection (for example malaria parasites by mosquitoes) into a host.

Examples of vectors and the diseases or pathogens they transmit are:

10 1) Mosquitoes

- Anopheles: malaria, filariasis;

- Culex: Japanese encephalitis, other viral diseases, filariasis, transmission of other worms;

- Aedes: yellow fever, dengue fever, other viral diseases, filariasis;

- Simuliidae: transmission of worms, in particular *Onchocerca volvulus*;

15 - Psychodidae: transmission of leishmaniasis

2) Lice: skin infections, epidemic typhus;

3) Fleas: plague, endemic typhus, cestodes;

4) Flies: sleeping sickness (trypanosomiasis); cholera, other bacterial diseases;

20 5) Mites: acariosis, epidemic typhus, rickettsialpox, tularaemia, Saint Louis encephalitis, tick-borne encephalitis (TBE), Crimean–Congo haemorrhagic fever, borreliosis;

6) Ticks: borellioses such as *Borrelia burgdorferi sensu lato.*, *Borrelia duttoni*, tick-borne encephalitis, Q fever (*Coxiella burnetii*), babesioses (*Babesia canis canis*), ehrlichiosis.

25 Examples of vectors in the sense of the present invention are insects, for example aphids, flies, leafhoppers or thrips, which are capable of transmitting plant viruses to plants. Other vectors capable of transmitting plant viruses are spider mites, lice, beetles and nematodes.

Further examples of vectors in the sense of the present invention are insects and arachnids such as mosquitoes, in particular of the genera *Aedes*, *Anopheles*, for example *A. gambiae*, *A. arabiensis*, *A.*

funestus, A. dirus (malaria) and Culex, psychodids such as Phlebotomus, Lutzomyia, lice, fleas, flies, mites and ticks capable of transmitting pathogens to animals and/or humans.

Vector control is also possible if the compounds of the formula (I) are resistance-breaking.

- 5 Compounds of the formula (I) are suitable for use in the prevention of diseases and/or pathogens transmitted by vectors. Thus, a further aspect of the present invention is the use of compounds of the formula (I) for vector control, for example in agriculture, in horticulture, in gardens and in leisure facilities, and also in the protection of materials and stored products.

Protection of industrial materials

- 10 The compounds of the formula (I) are suitable for protecting industrial materials against attack or destruction by insects, for example from the orders Coleoptera, Hymenoptera, Isoptera, Lepidoptera, Psocoptera and Zygentoma.

Industrial materials in the present context are understood to mean inanimate materials, such as preferably plastics, adhesives, sizes, papers and cards, leather, wood, processed wood products and coating compositions. The use of the invention for protecting wood is particularly preferred.

- 15 In a further embodiment, the compounds of the formula (I) are used together with at least one further insecticide and/or at least one fungicide.

In a further embodiment, the compounds of the formula (I) are present as a ready-to-use pesticide, i.e. they can be applied to the material in question without further modifications. Suitable further insecticides or fungicides are in particular those mentioned above.

- 20 Surprisingly, it has also been found that the compounds of the formula (I) can be employed for protecting objects which come into contact with saltwater or brackish water, in particular hulls, screens, nets, buildings, moorings and signalling systems, against fouling. Likewise, the compounds of the formula (I), alone or in combinations with other active compounds, can be used as antifouling agents.

Control of animal pests in the hygiene sector

- 25 The compounds of the formula (I) are suitable for controlling animal pests in the hygiene sector. In particular, the invention can be applied in the domestic sector, in the hygiene sector and in the protection of stored products, especially for controlling insects, arachnids, ticks and mites encountered in enclosed spaces such as dwellings, factory halls, offices, vehicle cabins, animal husbandries. For controlling animal pests, the compounds of the formula (I) are used alone or in combination with other active compounds and/or auxiliaries. They are preferably used in domestic insecticide products. The compounds of the
30 formula (I) are effective against sensitive and resistant species, and against all developmental stages.

These pests include, for example, pests from the class Arachnida, from the orders Scorpiones, Araneae and Opiliones, from the classes Chilopoda and Diplopoda, from the class Insecta the order Blattodea, from the orders Coleoptera, Dermaptera, Diptera, Heteroptera, Hymenoptera, Isoptera, Lepidoptera, Phthiraptera, Psocoptera, Saltatoria or Orthoptera, Siphonaptera and Zygentoma and from the class
5 Malacostraca the order Isopoda.

They are used, for example, in aerosols, pressure-free spray products, for example pump and atomizer sprays, automatic fogging systems, foggers, foams, gels, evaporator products with evaporator tablets made of cellulose or plastic, liquid evaporators, gel and membrane evaporators, propeller-driven evaporators, energy-free, or passive, evaporation systems, moth papers, moth bags and moth gels, as granules or dusts,
10 in baits for spreading or in bait stations.

Abbreviations and Symbols

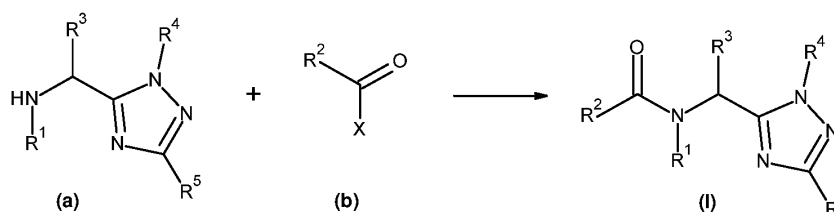
	AcOH:	acetic acid
	aq.:	aqueous
	br.:	broad
15	Boc:	<i>tert</i> -Butyloxycarbonyl
	d:	doublet
	DCC:	N,N'-dicyclohexylcarbodiimide
	DIPEA:	N,N-diisopropylethylamine
	DMF:	N,N-dimethylformamide
20	DMSO:	dimethylsulfoxide
	ee:	enantiomeric excess
	eq.:	equivalent
	ES:	electrospray ionization
	Et ₃ N	triethylamine
25	EtOAc:	ethyl acetate
	hr(s)	hour(s)
	HATU:	1-[bis(dimethylamino)methylene]-1H-1,2,3-triazolo[4,5-b]pyridinium-3-oxid hexafluorophosphate
	HOBt:	1-hydroxybenzotriazole hydrate
30	HPLC:	high performance liquid chromatography
	<i>i</i> PrOH:	isopropanol
	<i>J</i> :	coupling constant
	LCMS:	liquid chromatography-mass spectrometry
	<i>m/z</i> :	mass-to-charge ratio
35	M:	molarity
	m:	multiplet
	MeCN:	acetonitrile

	MeOH:	methanol
	NaH ₂ PO ₄ :	monosodium phosphate
	NaOH:	sodium hydroxide
	Na ₂ SO ₄ :	sodium sulfate
5	NH ₄ Cl:	ammonium chloride
	NMR:	nuclear magnetic resonance
	q:	quartet
	r. t.:	room temperature
	R _t :	retention time
10	s:	singlet
	sat.:	saturated
	T:	temperature
	t:	triplet
	T3P®:	propylphosphonic anhydride
15	THF:	tetrahydrofuran
	TMSOK	potassium trimethylsilanolate
	wt.:	weight
	δ:	chemical shift
	λ:	wavelength

Description of the processes and intermediates

Compounds of formula I and those shown in table 3 may be prepared as illustrated in the following scheme 1 where R¹, R², R³, R⁴, and R⁵ are as previously defined or stand for the corresponding fragments of the compounds shown in table 3. X stands for OH or Cl.

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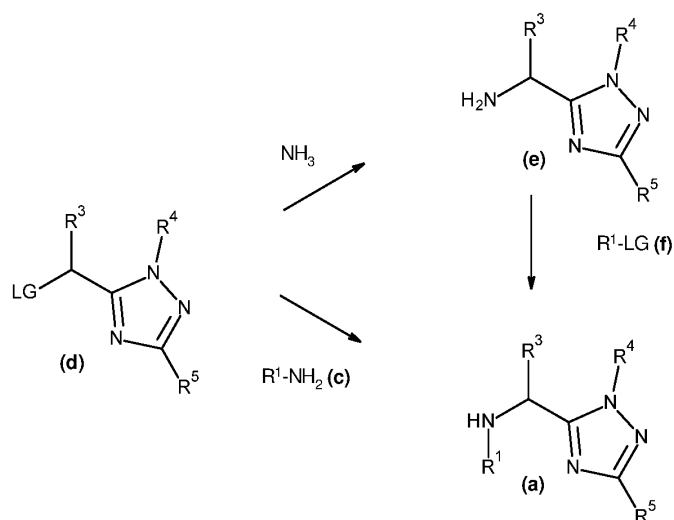
Scheme 1

X = OH: An triazole compound of formula (a) is reacted with a carboxylic acid of formula (b) (X = OH) to form compounds of formula I. For example, a mixture of a triazole of formula (a), a carboxylic acid of formula (b) (X = OH), a suitable coupling reagent, such as T3P[®], HATU or DCC/HOBt, a suitable base such as triethylamine or DIPEA, in a suitable solvent, such as ethyl acetate or DMF are mixed at temperatures ranging from around 0 to 100 °C to provide compounds of formula I which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

X = Cl: A triazole compound of formula (a) is reacted with a carboxylic acid chloride of formula (b) (X = Cl) to form compounds of formula I. For example, a mixture of a triazole of formula (a), a carboxylic acid chloride of formula (b) (X = Cl), a suitable base such as triethylamine or DIPEA, in a suitable solvent, such as dichloromethane or THF are mixed at temperatures ranging from around 0 to 100 °C to provide compounds of formula I which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

Carboxylic acids of formula (b) (X = OH) and carboxylic acid chlorides of formula (b) (X = Cl) are commercially available or may be synthesized by methods known to the skilled artisan. The synthesis of certain carboxylic acids of formula (b) (X = OH) has been described in WO 2019197468.

The requisite triazole compounds of formula (a) may be prepared as illustrated in the following scheme 2, where R³ and R⁴ are as previously described, R¹ is C₁-C₆alkyl and R⁵ is ethyl or n-propyl; or R¹, R³, R⁴ and R⁵ stand for the corresponding fragments of the compounds shown in table 3. LG is a suitable leaving group (see also WO 2017192385).

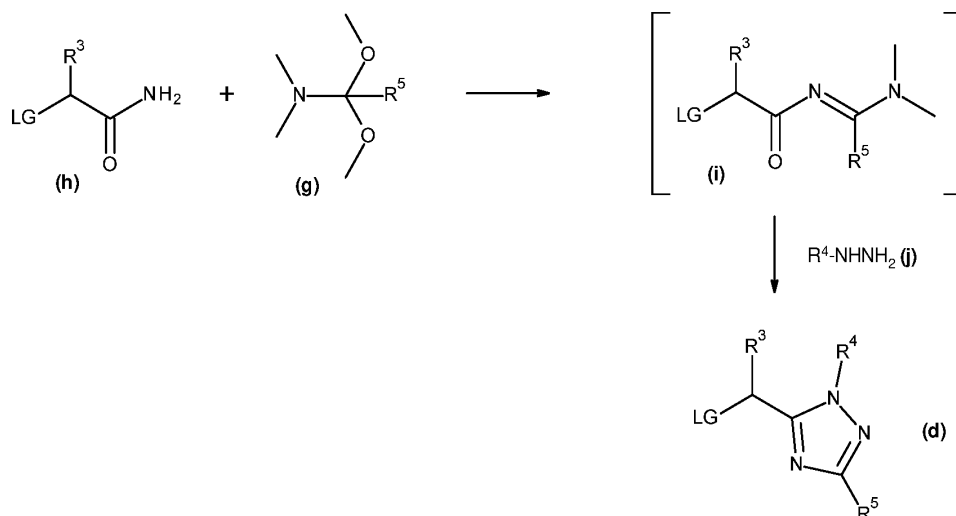
Scheme 2

An amine of formula (c) is reacted with a substituted triazole of formula (d) to form compounds of formula (a). For example, a mixture of a triazole of formula (d), an amine of formula (c), a suitable base, such as K₂CO₃, NaH or DIPEA in a suitable solvent, such as acetonitrile or DMF are mixed at temperatures ranging from around 20 to 120 °C to provide compounds of formula (a) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

Alternatively, a substituted triazole of formula (d) is reacted with ammonia to form compounds of formula (e). For example, a solution of ammonia in a suitable solvent, such as methanol, and a substituted triazole of formula (d) are mixed in a sealed tube at temperatures ranging from around 0 to 25 °C to provide compounds of formula (e) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as trituration. A substituted triazole of formula (e), a compound of formula (f), a suitable base, such as K₂CO₃ or DIPEA in a suitable solvent, such as acetonitrile or DMF are mixed at temperatures ranging from around 20 to 120 °C to provide compounds of formula (a) which may then be isolated and, if necessary and desired, purified using techniques well known in the art such as chromatography.

Amines of formula (c) and compounds of formula (f) are commercially available or may be synthesized by methods known to the skilled artisan.

The requisite triazole compounds of formula (d) may be prepared as illustrated in the following scheme 3, where R³ and R⁴ are as previously described and R⁵ is ethyl or n-propyl; or R³, R⁴ and R⁵ stand for the corresponding fragments of the compounds shown in table 3, LG is a suitable leaving group (see also WO 2017192385).

Scheme 3

An amide of formula (h) is reacted with an *N,N*-dimethylamide dimethyl acetal (g) to form compounds of formula (i) which are subsequently reacted with hydrazines (j) under acidic conditions to form compounds of formula (d). For example, a compound of formula (h) and an *N,N*-dimethylamide dimethyl acetal of formula (g) are reacted in a suitable solvent, such as CH_2Cl_2 at reflux to provide compounds of formula (i). Upon removal of the solvent, compounds of formula (i) are reacted with a substituted hydrazine (j) in a suitable solvent such as 1,4-dioxane, acetic acid or a mixture of such solvents at temperatures ranging from around 20 to 100 °C to provide compounds of formula (d) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

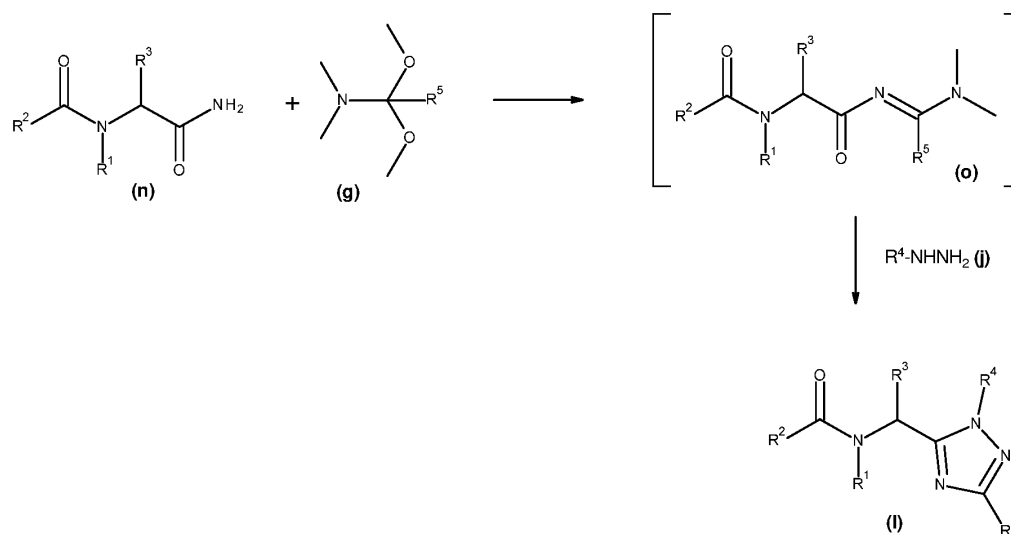
N,N-dimethylamide acetals of formula (g), amides of formula (h), and hydrazines of formula (j) are commercially available or may be synthesized by methods known to the skilled artisan.

For example:

For 5-bromo-2-hydrazinopyridine, see WO2013/038362

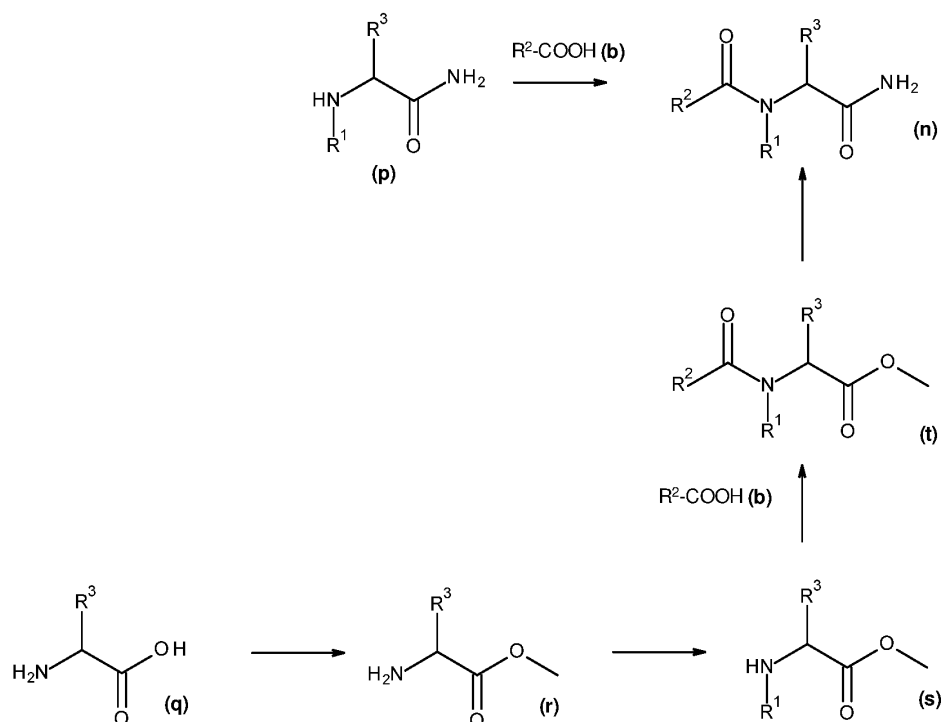
For 2-hydrazino-1,3-thiazoles, see US2008/0234327, WO2018/064119, WO2008/144767, WO2008/121861, WO2004/046120

Compounds of formula I and those shown in table 3 may be prepared as illustrated in the following scheme 4 where R^1 , R^2 , R^3 and R^4 are as previously defined and R^5 is ethyl or *n*-propyl; or R^1 , R^2 , R^3 , R^4 and R^5 stand for the corresponding fragments of the compounds shown in table 3.

Scheme 4

An amide of formula (n) is reacted with an *N,N*-dimethylamide dimethyl acetal of formula (g) to form compounds of formula (o) which are subsequently reacted with substituted hydrazines of formula (j) under acidic conditions to form compounds of formula I. For example, a compound of formula (n) and an *N,N*-dimethylamide dimethyl acetal of formula (g) are reacted in a suitable solvent, such as CH_2Cl_2 at reflux to provide compounds of formula (o). Upon removal of the solvent, compounds of formula (o) are reacted with a substituted hydrazine of formula (j) in a suitable solvent such as 1,4-dioxane, acetic acid or a mixture of such solvents at temperatures ranging from around 20 to 100 °C. The resulting compounds of formula I may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

The requisite amides of formula (n) may be prepared as illustrated in the following scheme 5, where R^2 and R^3 are as previously described and R^1 is $\text{C}_1\text{-C}_6$ alkyl or H (see also WO 2017192385); or R^1 , R^2 and R^3 stand for the corresponding fragments of the compounds shown in table 3.

Scheme 5

An amino amide of formula (p) is reacted with a carboxylic acid of formula (b) to form compounds of formula (n). For example, a mixture of an amino amide of formula (p), a carboxylic acid (b), a suitable coupling reagent, such as T3P[®], HATU or DCC/HOBt, a suitable base such as triethylamine or DIPEA, in a suitable solvent such as ethyl acetate or DMF are mixed at temperatures ranging from around 0 to 100 °C to provide compounds of formula (n) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

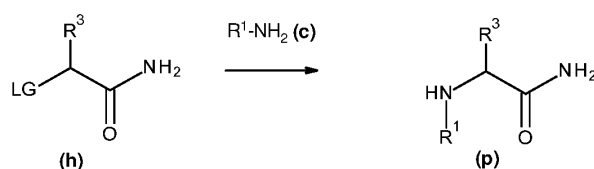
Alternatively, an amino acid of formula (q) is reacted with thionyl chloride in a suitable solvent, such as MeOH, at r.t. to provide amino esters of formula (r). The resulting amino esters (r) are reacted with an aldehyde or a ketone, a suitable reducing agent such as sodium triacetoxyborohydride, a dehydrating agent such as Na₂SO₄, in a suitable solvent such as acetic acid, at r.t. to provide compounds of formula (s). The resulting amino esters of formula (s) are then reacted with a carboxylic acid of formula (b), a suitable coupling reagent, such as T3P[®], a suitable base such as DIPEA, in a suitable solvent, such as ethyl acetate at about 90 °C to provide amido esters of formula (t) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography. The resulting amido esters of formula (t) are reacted with magnesium nitride in a suitable solvent, such as MeOH at about 80 °C in a sealed tube to provide compounds of formula (n) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography or extraction.

Compounds of formula (b) and (q) are commercially available or may be synthesized by methods known to the skilled artisan. The synthesis of certain carboxylic acids of formula (b) (X = OH) has been described in WO 2019197468. The requisite amino amide compounds of formula (p) are commercially available or may be prepared as illustrated in the following scheme 6, where R¹ and R³ are as previously described or stand for the corresponding fragments of the compounds shown in table 3 and LG is a suitable leaving group (see also WO 2017192385).

Compounds of formula (c) and (h) are commercially available.

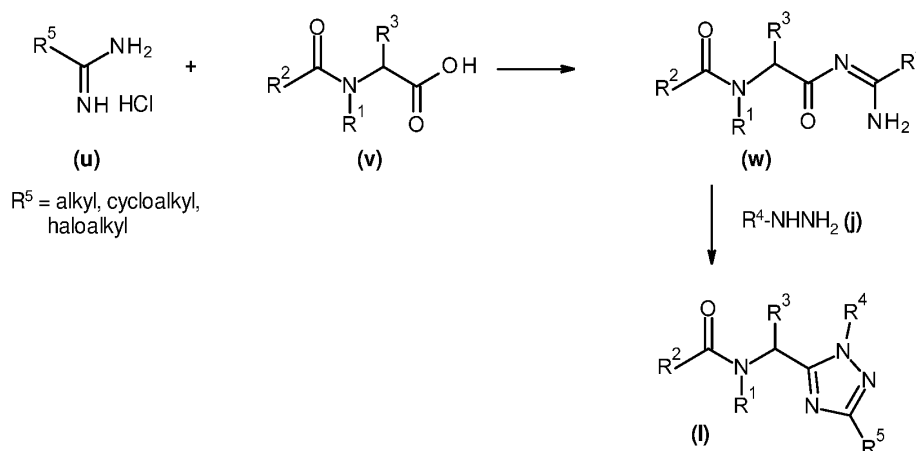
The requisite amines of formula (p) may be prepared as illustrated in the following scheme 6, where R¹ and R³, are as previously described (see also WO 2017192385) or stand for the corresponding fragments of the compounds shown in table 3.

Scheme 6



An amine of formula (c) is reacted with an amide of formula (h) to form compounds of formula (p). For example, a mixture of an amine of formula (c), an amide of formula (h), a suitable base, such as K₂CO₃ or DIPEA in a suitable solvent, such as acetonitrile or DMF are mixed at 25-80 °C to provide compounds of formula (p) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

In an alternative approach compounds of formula I may be prepared as illustrated in the following scheme 7 where R¹, R², R³, R⁴ are as previously defined and R⁵ is ethyl, n-propyl, iso-propyl, difluoromethyl or cyclopropyl; or R¹, R², R³, R⁴ and R⁵ stand for the corresponding fragments of the compounds shown in table 3.

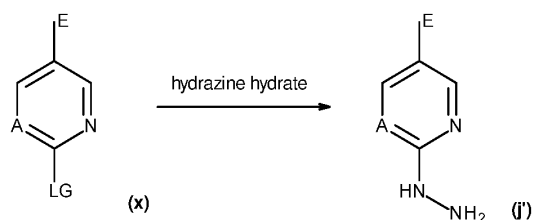
Scheme 7

An amidine hydrochloride of formula (u) is reacted with an acid of formula (v). For example, an amidine hydrochloride of formula (u), a carboxylic acid (v), a suitable coupling reagent, such as HATU, DCC or HOBt, a suitable base such as triethylamine or DIPEA, in a suitable solvent such as acetonitrile or DMF are mixed at temperatures ranging from around 0 to 100 °C, to form compounds of formula (w) which are subsequently reacted with substituted hydrazines of formula (j) under acidic conditions to form compounds of formula I which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

- 10 Amidine hydrochlorides of formula (u), carboxylic acid derivatives of formula (v) and hydrazines of formula (j) are commercially available or may be synthesized by methods known to the skilled artisan.

Compounds of formula (j') may be prepared as illustrated in the following scheme 8 where E is trifluoromethoxy, difluoromethoxy or trifluoromethylsulfanyl, LG is chlorine, fluorine, methylthio, methylsulfinyl or methylsulfonyl and A is N or CH.

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Scheme 8

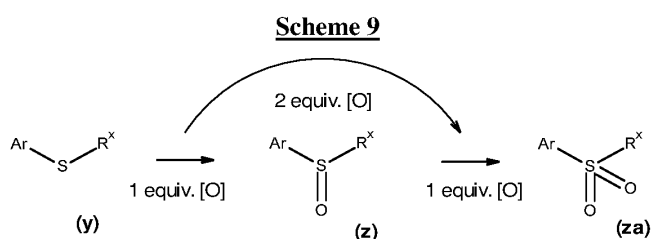
A compound of formula (x) containing a leaving group (LG) (WO2016/001266 for LG = methylsulfonyl) is reacted with hydrazine hydrate to form hydrazines of formula (j'). For example, a mixture of a leaving group containing compound (x) and hydrazine hydrate in a suitable solvent, such as methanol or ethanol is reacted at 0-80 °C to provide compounds of formula (j') or their hydrochloride, hydrobromide or

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methanesulfonate salts which may then be isolated and, if necessary and desired, purified using techniques well known in the art.

Compounds of formula (x) are either commercially available or may be synthesized by methods known to the skilled artisan.

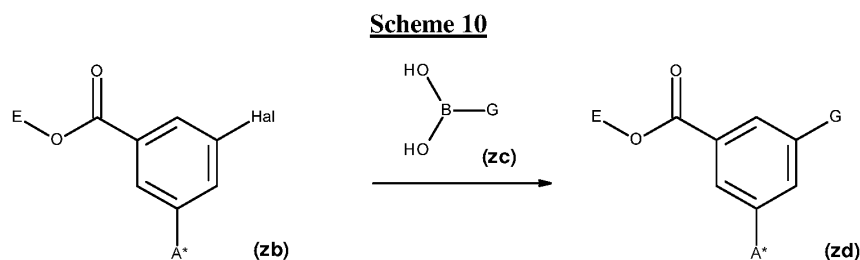
- 5 Sulfoxides (sulfines) of the general formula (z) and sulfones of the formula (za) may be prepared as illustrated in the following scheme 9 wherein Ar is phenyl or hetaryl and R^x is C₁-C₃alkyl, C₁-C₂haloalkyl or cyclopropyl.



- 10 A sulfanyl group containing compound of formula (y) is reacted with an oxidizing reagent (abbreviated as [O] in scheme 9) such as 3-chloroperoxybenzoic acid, ruthenium(III) chloride in combination with sodium periodate or a combination of formic acid and hydrogenperoxide to form compounds of formula (z) or (za), depending on the molar equivalents used of the oxidizing reagent. The use of one equivalent of the oxidizing reagent leads to sulfinyl compounds of formula (z) while the use of two equivalents leads to the isolation of sulfone compounds of formula (za). Sulfinyl compounds of the formula (z) are chiral compounds that form mixtures of optical isomers.
- 15

Compounds of formula (zd) may be prepared as illustrated in the following scheme 10 wherein E is H or C₁-C₆alkyl, Hal is bromine or iodine, G is cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl or trifluoromethyl. A* is fluorine, chlorine, bromine, iodine, -CN, difluoromethyl, trifluoromethyl, difluoromethoxy, trifluoromethoxy, methylsulfonyl, ethylsulfonyl, isopropylsulfonyl, cyclopropylsulfonyl, difluoromethylsulfonyl, trifluoroethylsulfonyl, trifluoromethylsulfonyl, -SF₅ and cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl.

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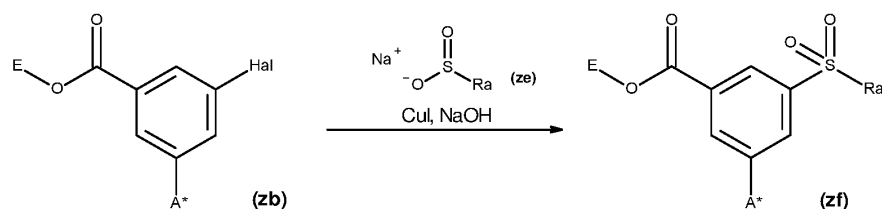


A halogen containing compound of formula (zb) is reacted with a boronic acid of formula (zc) to form compounds of formula (zd). For example, a mixture of a halogen containing compound of formula (zb), a boronic acid (zc), a suitable catalyst, such as palladium(II) acetate in combination with tricyclohexylphosphine, a suitable base such as tripotassium phosphate, in a suitable solvent or solvent mixture such as toluene and water are reacted at temperatures ranging from around 0 to 100 °C to provide compounds of formula (zd) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography. Compounds of formula (zd) in which E is C₁-C₆alkyl can be transformed to compounds of formula (zd) in which E is H by treatment with an alkali hydroxide in a suitable solvent or solvent mixture such as / containing tetrahydrofuran, ethanol or water at temperatures ranging from around 0 to 100 °C.

Compounds of formula (zb) are either commercially available or may be synthesized by methods known to the skilled artisan. The synthesis of compounds of formula (zb) for which A* equals methylsulfonyl, ethylsulfonyl, isopropylsulfonyl, cyclopropylsulfonyl is described in Scheme 11 and by Schemes 12 and 9.

Compounds of formula (zf) may be prepared as illustrated in the following scheme 11 wherein E is H or C₁-C₆alkyl, Hal is iodine or bromine, Ra is C₁-C₃alkyl or cyclopropyl; A* is fluorine, chlorine, difluoromethyl, trifluoromethyl, difluoromethoxy, trifluoromethoxy and cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl.

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Scheme 11

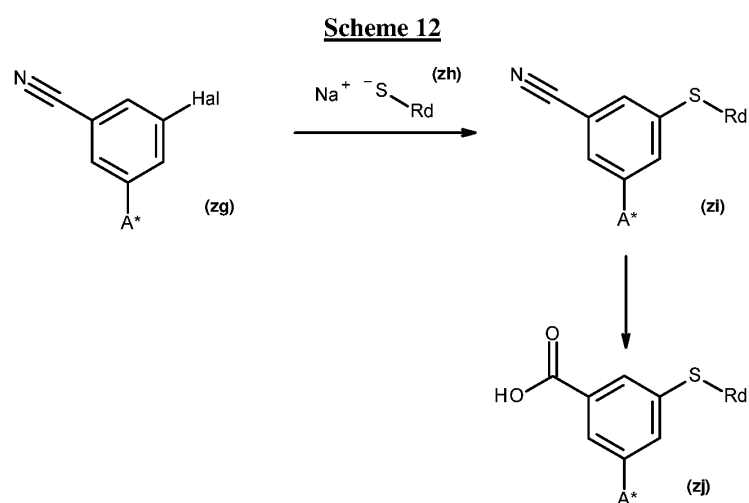
An aryl halide of formula (zb) is reacted with a sulfinate salt of formula (ze) under copper catalysis to form sulfones of formula (zf).

For example, a mixture of a compound of formula (zb), a sodium sulfinate salt of formula (ze), copper(I) iodide, proline and sodium hydroxide are reacted in a suitable solvent, such as dimethyl sulfoxide at temperatures ranging from 40 to 140 °C (compare WO 2019197468). In an alternative approach a mixture of a compound of formula (zb), a sodium sulfinate salt of formula(ze), copper(I) iodide, *trans*-*N,N*-dimethylcyclohexane-1-2-diamine and cesium carbonate are reacted in a suitable solvent, such as DMF at temperatures ranging from 40 to 140 °C (see for example 3-methylsulfonyl-5-(trifluoromethoxy)benzoic acid reported in this application).

The resulting compounds of formula (zf) may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography. Compounds of formula (zf) in which E is C₁-C₆alkyl can be transformed to compounds of formula (zf) in which E is H by treatment with an alkali hydroxide in a suitable solvent or solvent mixture such as / containing tetrahydrofuran, ethanol or water at temperatures ranging from around 0 to 100 °C. In case E is a tert-butyl group this ester can be cleaved under acidid conditions in a suitable solvent such as dichloromethane in the presence of a suitable acid such as trifluoroacetic acid at temperatures ranging from 0 – 40 °C (see also in this application the synthesis of 3-bromo-5-(1-fluorocyclopropyl)benzoic acid as an example of acidic cleavage of a tert-butyl ester).

- 10 The aryl halides (zb) and sulfinate salts of formula (zf) are commercially available or may be synthesized by methods known to the skilled artisan. The synthesis of compounds of formula (zb) for which A* is cyclopropyl optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl is described in Scheme 10 and Scheme 21.

Compounds of formula (zj) may be prepared as illustrated in the following scheme 12, Hal is fluorine or chlorine, Rd is C₁-C₃alkyl optionally substituted by fluorine or cyclopropyl; A* is chlorine, bromine, iodine, difluoromethyl, trifluoromethyl, difluoromethoxy, trifluoromethoxy and cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl.



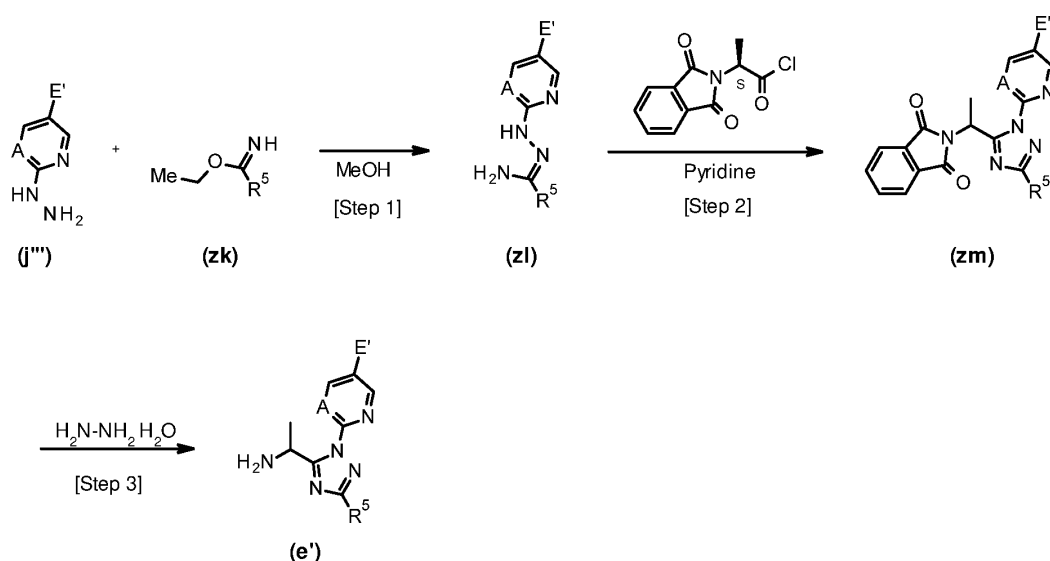
An aryl halide of formula (zg) is reacted with a thiolate salt of formula (zh) to form thioethers of formula (zi) which are then hydrolised to form carboxylic acids of formula (zj).

For example, a mixture of a halide of formula (zg) and a sodium thiolate of formula (zh), is reacted in a suitable solvent, such as N,N-dimethylformamide at temperatures ranging from – 20 to 50 °C. The resulting nitriles of formula (zi) are then hydrolized either under basic conditions, using for example

aqueous sodium hydroxide in a suitable solvent or solvent mixture, such as ethanol or methanol/THF at temperatures ranging from 40 to 100 °C or under acidic conditions in a suitable strong acid, such as sulfuric acid or hydrochloric acid either neat or diluted with a suitable dilutant such as water at temperatures ranging from 40 to 100 °C. The obtained carboxylic acids (zj) are then if necessary and desired, purified using techniques well known in the art, such as chromatography. The sulfanyl groups contained in the carboxylic acids (zj) may then be oxidized to sulfinyl or sulfonyl groups as described in Scheme 9.

The requisite aryl halides (zg) and thiolate salts of formula (zh) are commercially available or may be synthesized by methods known to the skilled artisan (e.g. WO 2013049250 for the synthesis of cyclopropanethiol). Thiolate salts may be synthesized from the corresponding thiols through deprotonation with sodium hydride in a suitable solvent such as N,N-dimethylformamide. The synthesis of compounds of formula (zg) for which A* is cyclopropyl optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl can be achieved in analogy to the procedures described in Scheme 10 and Scheme 21.

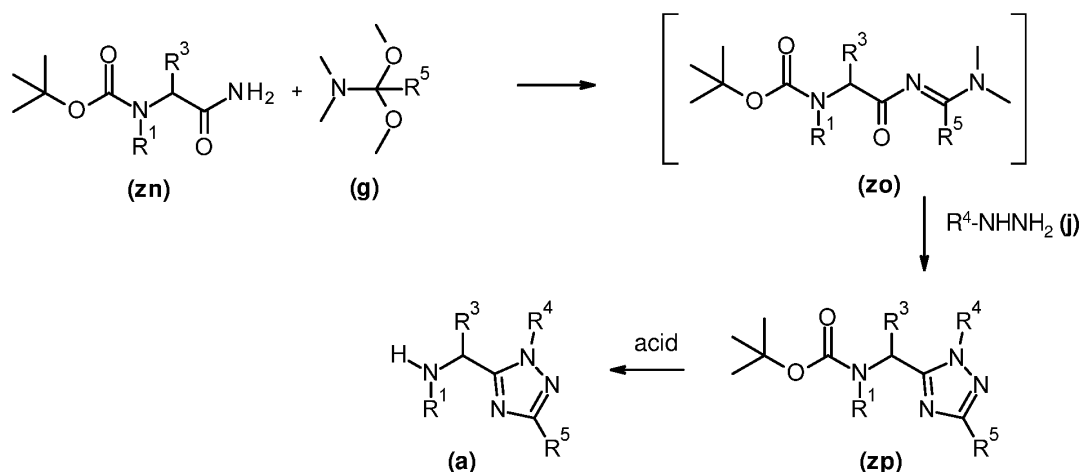
Scheme 13 illustrates the preparation of 3-haloalkyl triazoles containing amines (e') as used for the synthesis of e.g. example I-5. R⁵ is difluoromethyl and E' is hydrogen, chlorine or CN. A is N or CH.

Scheme 13

In a first step, a hydrazone amide (zl) is formed as described in EP 1099695. In a second step, (αS)-1,3-dihydro-α-methyl-1,3-dioxo-2H-isindole-2-acetyl chloride, prepared from (αS)-1,3-dihydro-α-methyl-1,3-dioxo-2H-isindole-2-acetic acid (Pht-Ala-OH purchased from ABCR) and oxalyl chloride according to *Tetrahedron: Asymmetry*, 21(8), 936-942, 2010, reacts with the hydrazone amide in the presence of a base, like pyridine, as described in EP 1099695 to form a triazole of formula (zm); a partial or full racemization is possible. In a third step, the phthalimide protecting group is removed by reaction with

hydrazine hydrate in a suitable solvent, like ethanol, as described in WO 2018086605. In a final step, the obtained amine is reacted with a carboxylic acid to form the example compounds, e.g. I-5 as described in scheme 1.

Compounds of formula (a) may also be prepared as illustrated in the following scheme 14 where R¹, R³ and R⁴ are as previously defined and R⁵ is hydrogen, methyl, ethyl or n-propyl; or R¹, R³ and R⁴ stand for the corresponding fragments of the compounds shown in table 3.

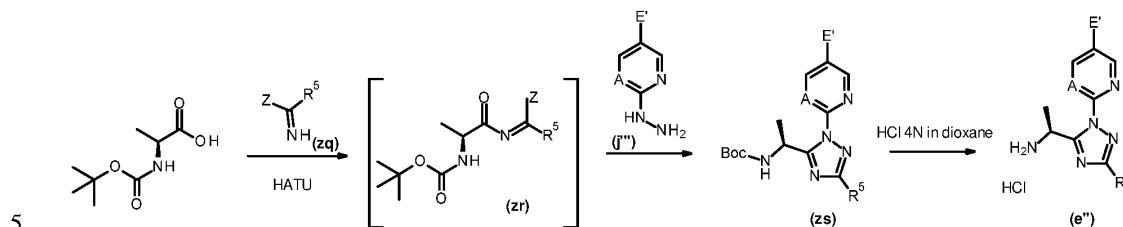
Scheme 14

An amide of formula (zn) is reacted with an *N,N*-dimethylamide dimethyl acetal of formula (g) to form compounds of formula (zo) which are subsequently reacted with substituted hydrazines of formula (j) under acidic conditions to form compounds of formula (zp). For example, a compound of formula (zn) and an *N,N*-dimethylamide dimethyl acetal of formula (g) are reacted in a suitable solvent, such as CH₂Cl₂ at reflux to provide compounds of formula (zo). After removal of the solvent, compounds of formula (zp) are reacted with a substituted hydrazine of formula (j) in a suitable solvent such as 1,4-dioxane, acetic acid or a mixture of such solvents at temperatures ranging from around 20 to 80 °C. The resulting compounds of formula (zp) may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

A carbamate of formula (zp) is treated with an acid to form amines of formula (a). For example, a carbamate of formula (zp) and a suitable acid, such as hydrogen chloride or trifluoroacetic acid, are reacted in a suitable solvent, such as dioxane or in the case of trifluoroacetic acid without an additional solvent at temperatures ranging from around 0 to 80 °C. The resulting amines of formula (a) may then be isolated as their acid salts or after base treatment as free amines and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

The requisite amides of formula (zn) and hydrazines of formula (j) are commercially available or may be synthesized by methods described in this application or methods known to the skilled artisan.

Scheme 15 illustrates the preparation of alkyltriazole containing amines (e'') as used for the synthesis of e.g. example I-1. R⁵ is ethyl, n-propyl, iso-propyl, cyclopropyl and E' is hydrogen, -CN or chlorine. Z is NH₂ or OC₁-C₆alkyl. A is N or CH.

Scheme 15

N-(tert-butoxycarbonyl)-L-alanine is reacted with an alkylamidine (for Z = NH₂) or an alkylimidate (for Z = OC₁-C₆alkyl) to form intermediates of formula (zr) which are subsequently reacted with substituted hydrazines of formula (j''') to form alkyltriazoles of formula (zs).

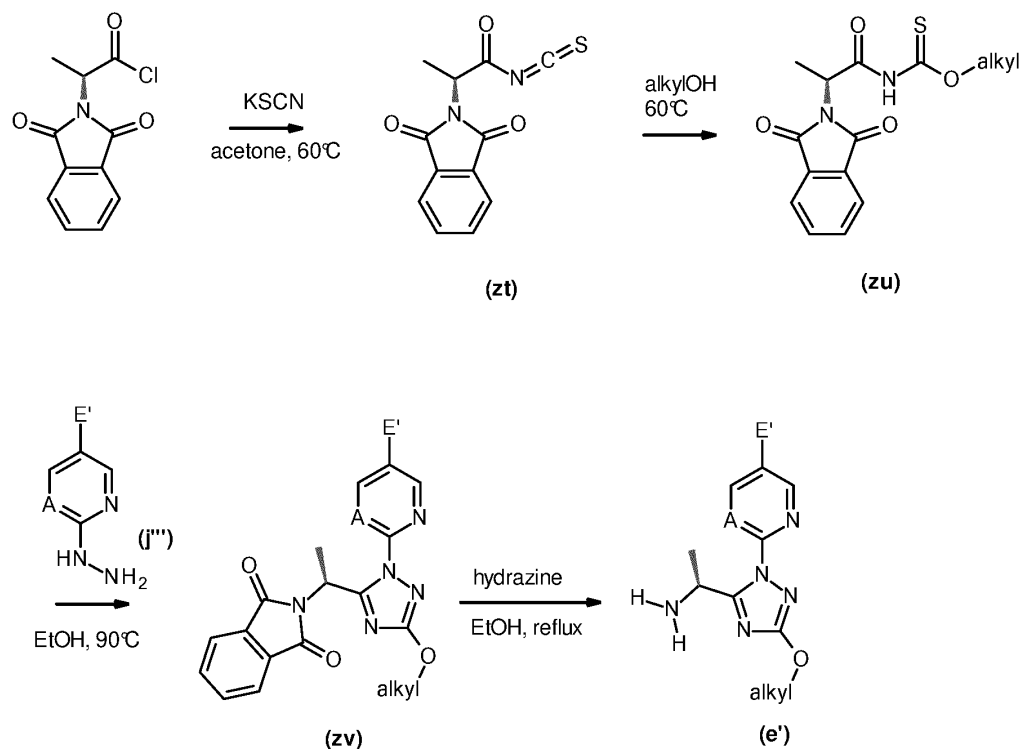
For example in the case of Z = NH₂ (compare *J. Org. Chem.* 2011, 76, 1177-1179) N-(tert-butoxycarbonyl)-L-alanine and an alkylamidine of formula (zq) are reacted in the presence of a suitable coupling reagent, such as HATU, a suitable base such as triethylamine or DIPEA, in a suitable solvent, such as DMF, at temperatures ranging from 0 to 50 °C to form an acylamidine intermediate of formula (zr). After removal of the solvent, the intermediates of formula (zr) are reacted with a substituted hydrazine of formula (j''') in a suitable solvent such as acetic acid at temperatures ranging from around 20 to 80 °C. The resulting alkyltriazoles of formula (zs) may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

In the case of Z = OC₁-C₆alkyl N-(tert-butoxycarbonyl)-L-alanine and an alkylimidate of formula (zq) or a suitable salt thereof are reacted in the presence of a suitable coupling reagent, such as HATU, a suitable base such as triethylamine or DIPEA, in a suitable solvent, such as THF at temperatures ranging from around 0 to 25 °C to form acyl imidate intermediates of formula (zr). Upon addition of a substituted hydrazine of formula (j''') the intermediate of formula (zr) reacts at temperatures ranging from around 20 to 80 °C to give alkyltriazoles of formula (zs) which may then be isolated and, if necessary and desired, purified using techniques well known in the art, such as chromatography.

Carbamates of formula (zs) are treated with an acid to form amines of formula (e'') as shown in scheme 14.

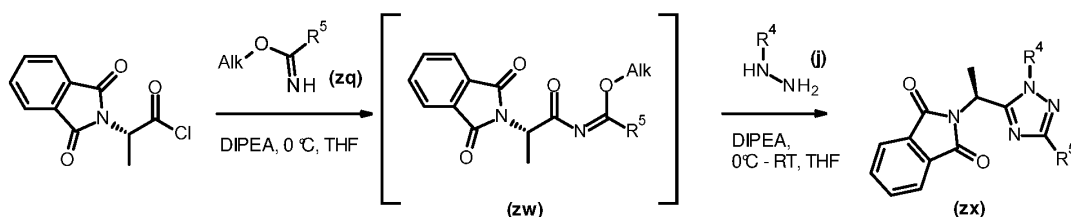
The requisite alkylamidines and alkylimidate or their suitable salts and hydrazines of formula (j''') are commercially available or may be synthesized by methods described in this application or methods known to the skilled artisan (see for example WO 2011133447 for the synthesis of methyl cyclopropanecarboximide hydrochloride).

Scheme 16 illustrates the preparation of alkoxytriazole containing amines (e') as used for the synthesis of e.g. example I-7. Alkyl is methyl, ethyl or iso-propyl and E' is hydrogen, chlorine or CN. A is N or CH.

Scheme 16

- 5 The synthesis starts with the reaction of (2S)-2-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)propanoyl chloride with potassium thiocyanate (KSCN) in acetone to yield the corresponding isocyanate intermediate (zt) which is treated in the next step with the corresponding alcohol to afford the O-alkyl [(2S)-2-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)propanoyl]carbamothioates (zu). The reaction between intermediate (zu) and a hydrazine of formula (j''') in ethanol affords cyclized products of formula (zv) as described in Bioorganic & Medicinal Chemistry 26 (2018) 3321–3344. The deprotection of the amino group with hydrazine hydrate yields primary amines of formula (e'). In a final step, the obtained amine is reacted with a carboxylic acid to form the example compounds, e.g. I-10 as described in scheme 1.
- 10

Compounds of formula (zx) may be prepared as illustrated in the following scheme 17 wherein Alk is C₁-C₆alkyl, R⁴ is 5-cyanopyridin-2-yl and R⁵ ethyl, n-propyl, iso-propyl or cyclopropyl.

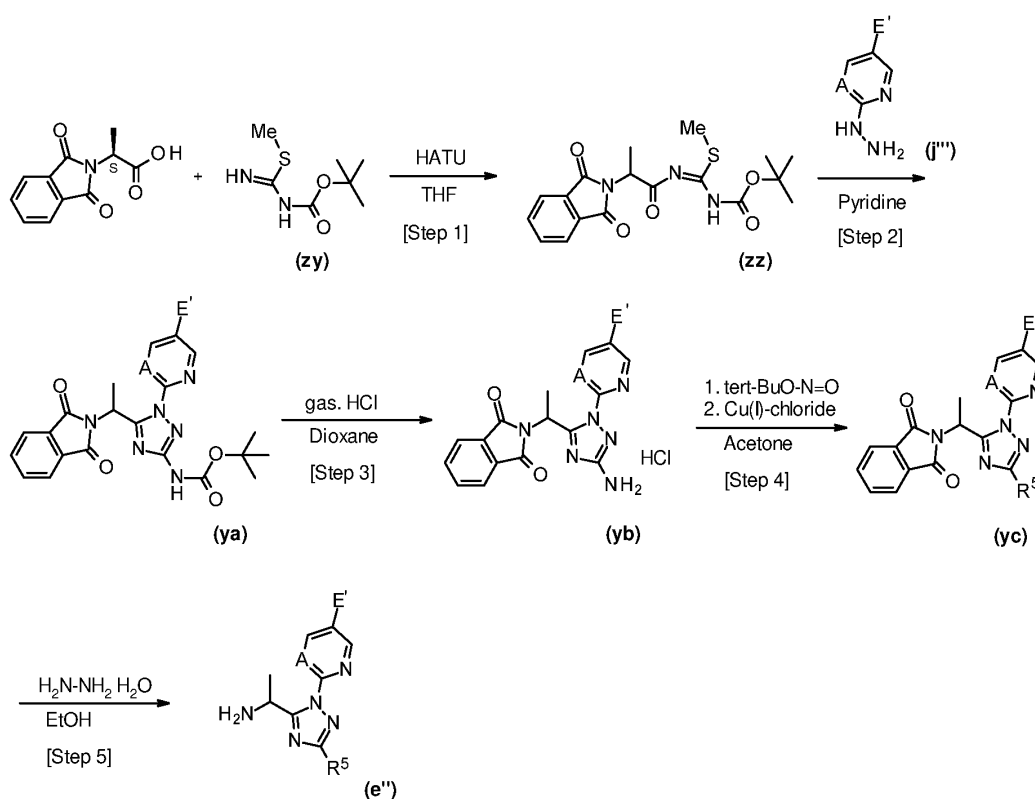
Scheme 17

(α S)-1,3-dihydro- α -methyl-1,3-dioxo-2H-isoindole-2-acetyl chloride is reacted with an imidate (zq) or a suitable salt thereof to form an acyl imidate intermediate of formula (zw) which then reacts with a hydrazine of formula (j) to yield triazoles of formula (zx). For example, a mixture of the acid chloride and a imidate of formula (zq), is reacted in a suitable solvent, such as THF at temperatures ranging from – 20 to 25 °C. The resulting intermediates of formula (zw) are then reacted with hydrazines of formula (j) in a suitable solvent, such as THF at temperatures ranging from 0 °C to 80 °C. The obtained triazoles of formula (zx) are then if necessary and desired, purified using techniques well known in the art, such as chromatography. The phthalimide protecting group may be removed using hydrazine as described in scheme 13 to yield the respective free amines.

The requisite acid chloride may be obtained as described in scheme 13 and imidates of formula (zq) or their salts are commercially available or may be synthesized by methods known to the skilled artisan.

Scheme 18 illustrates the preparation of 5-halogen triazole containing amines (e'') as used for the synthesis of e.g. example I-28. R⁵ is chlorine, bromine or iodine and E' is hydrogen, chlorine or -CN. A is N or CH.

15

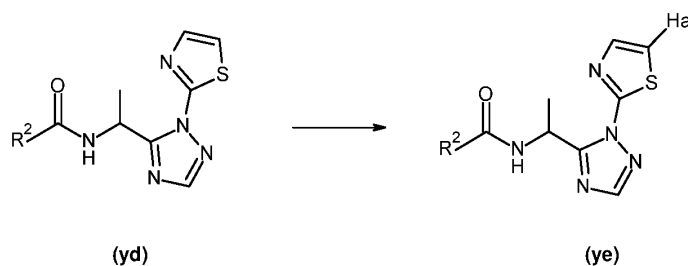
Scheme 18

In a first step, (α S)-1,3-dihydro- α -methyl-1,3-dioxo-2H-isoindole-2-acetic acid (Pht-Ala-OH purchased from ABCR) is reacted with 1-N-Boc-2-methyl-isothioureia (purchased from ABCR) in the presence of a base and the coupling reagent HATU to form the N-acylated 1-N-Boc-2-methyl-isothioureia (zz); a partial

or full racemization is possible. In a second step the cyclization occurs with hydrazines (e.g. A is nitrogen; E' is hydrogen) and in the presence of a base, like pyridine, as described in WO 2014009425 A1 to form the 1,2,4-triazoles of formula (ya), wherein R⁵ is NH-Boc. After N-Boc-deprotection in the third step under acidic conditions (HCl in dioxane) the 3-amino-1,2,4-triazole hydrochlorides (yb) (R⁵ = NH₂) are formed, which can be treated in the fourth step with at first with *tert*-butyl nitrite and afterwards with a copper halides as salts, like CuCl₂ (R⁵ = Cl) described by N. Desroy et al., *J. Med. Chem.* **2013**, 56, 1418-1430, CuBr₂ (R⁵ = Br) described in JP-Pat. 2010070503 A, CuI/I₂ mixture (R⁵ = I) as described by K. Pchalek and M. P. Hay *J. Org. Chem.* **2006**, 71, 6530-6535, or with 1,2-diiodomethane (R⁵ = I) as described by N. R. Norcross et al. *J. Med. Chem.*, **2016**, 59(13), 6101-6120, forming the 3-halogen-substituted 1,2,4-triazoles. Alternatively, fluorine (R⁵ = F) can be introduced e.g. using HF instead of copper halides as described for example by V. Krchnak and Z. Arnold *Coll. Czech. Chem. Commun.*, 1975, 40(5), 1390-1395. In a fifth step, the phthalimide protecting group is removed by reaction with hydrazine hydrate in a suitable solvent, like ethanol, as described in WO 2018086605. In a final step, the obtained amines (e'') are reacted with a carboxylic acid to form the example compounds, e.g. I-28 as described in scheme 1.

Compounds of formula (ye) may be prepared as illustrated in the following scheme 19 where R² is as previously defined. Hal is chlorine, bromine or iodine; or R² stands for the corresponding fragments of the compounds shown in table 3

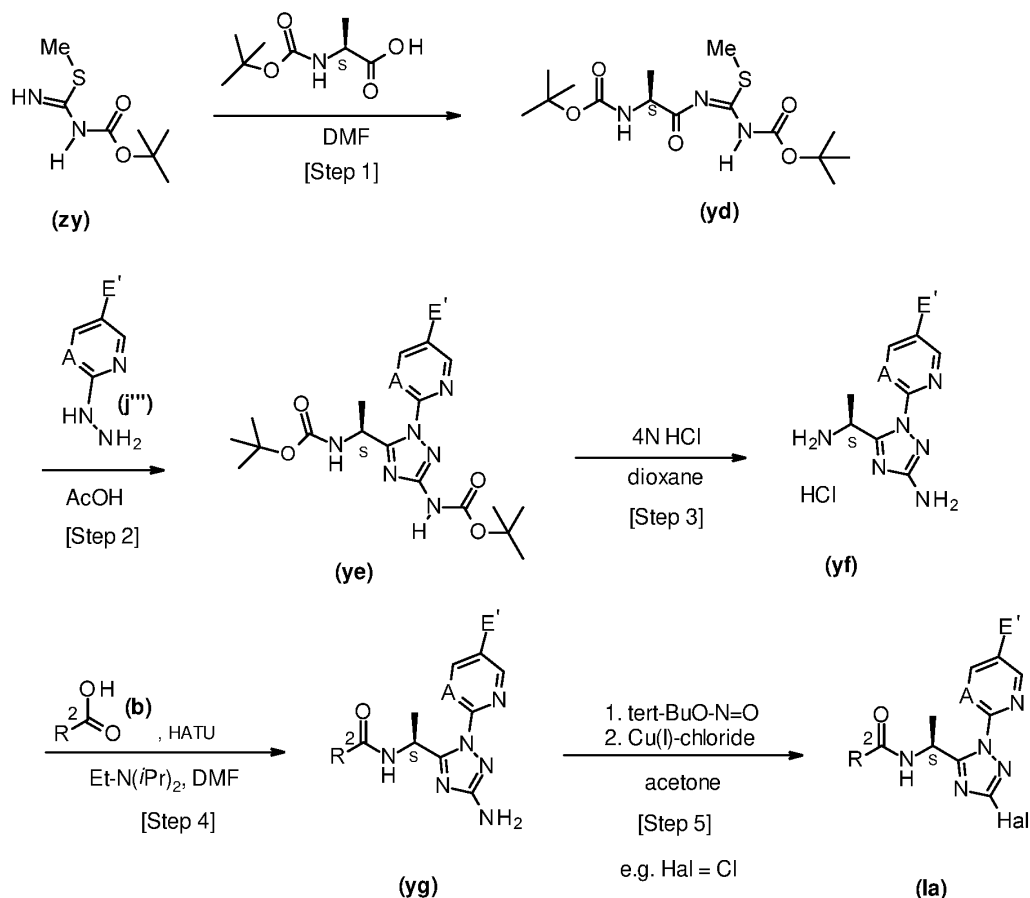
Scheme 19



Compounds of formula (yd) can be further derivatized by halogenation of the thiazole. The required methods are known to the skilled artisan. E.g. iodination is achieved with a halogenating agent such as N-iodo-succinimide in a suitable solvent such as DMF (synthesis of example II-51 described in this application).

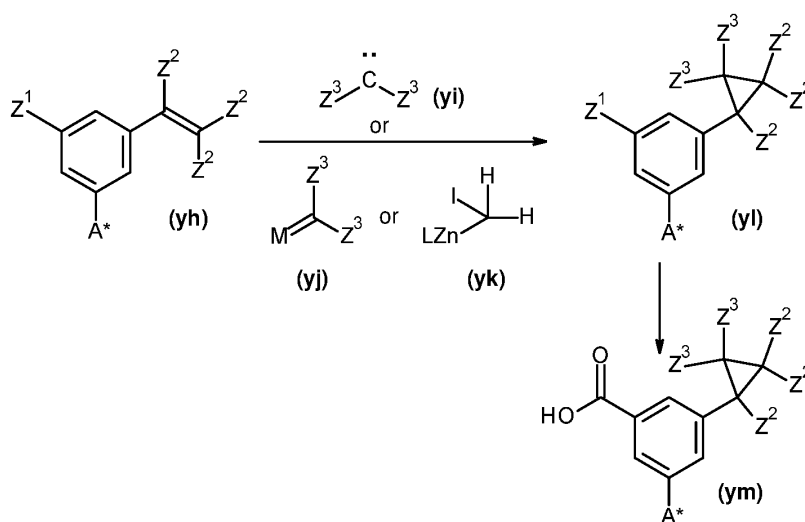
Scheme 20 illustrates the preparation of 3-halogen-1,2,4-triazole containing compounds (Ia) starting from 3-amino-1,2,4-triazole containing compounds (yg). Hal is chlorine, bromine or iodine and E' is hydrogen, chlorine or -CN. A is N or CH.

Scheme 20



In a first step, *N*-Boc-alanine reacts with 1-*N*-Boc-2-methyl-isothioureia (zy) (purchased from ABCR) (see scheme 18 and 20) in the presence of a base and the coupling reagent HATU to form the *tert*-butyl *N*-[(1*S*)-2-[[*tert*-butoxycarbonylamino)-methylsulfanyl-methylene]amino]-1-methyl-2-oxo-ethyl]carbamate (yd), which can be used without purification. In a second step the cyclization of (yd) occurs with hydrazines (j''') in the presence of an acid, like acetic acid to form the 1,2,4-triazoles of formula (ye). In the third step, both *N*-Boc protecting groups are removed from the 1,2,4-triazoles of formula (ye) by reaction with 4N HCl in dioxane to form the 3-amino-1,2,4-triazole containing amines as their hydrochloride salts (yf). In the fourth step, the obtained amine salts (yf) are reacted with a carboxylic acid to form the enantiomerically enriched compounds (yg), which can be treated in the fifth step at first with *tert*-butyl nitrite and afterwards with a copper halide salts, like CuCl₂ (R⁵ = Cl) described by N. Desroy et al., *J. Med. Chem.* **2013**, 56, 1418-1430, CuBr₂ (R⁵ = Br) described in JP-Pat. 2010070503 A, a CuI/I₂ mixture (R⁵ = I) as described by K. Pchalek and M. P. Hay *J. Org. Chem.* **2006**, 71, 6530-6535, or with 1,2-diiodoethane (R⁵ = I) as described by N. R. Norcross et al. *J. Med. Chem.*, **2016**, 59(13), 6101-6120, to form the 3-halogen-substituted 1,2,4-triazole containing compounds (Ia), e.g. I-95 and I-96. Acids of formula (ym) containing substituted cyclopropyl groups may also be prepared as illustrated in the following scheme 21 wherein A* is fluorine, chlorine, bromine, iodine, -CN, difluoromethyl,

trifluoromethyl, difluoromethoxy, trifluoromethoxy, -SF₅ and cyclopropyl wherein the cyclopropyl is optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl. Z¹ is either -CN or -CO₂C₁-C₆alkyl. Z² and Z³ are independently selected from the group of hydrogen, halogen, -CN, methyl, difluoromethyl or trifluoromethyl with the prerequisite that only up to two of the substituents Z² and Z³ are different from hydrogen. L is iodo or trifluoroacetate. M is a transition metal complex fragment containing iron, copper, palladium or rhodium and a suitable ligand substitution.

Scheme 21

Alkene containing compounds of formula (yh) react with free carbenes (yi), zinc carbenoids (yk) and certain transition metal carbene complexes (yj) to yield cyclopropyl containing compounds of formula (yl). These may then be transformed to acids of formula (ym) either by ester cleavage (in case Z¹ is -CO₂C₁-C₆alkyl) or by hydrolysis of a cyano group (in case Z¹ is -CN). Different cyclopropanation reactions are known to persons skilled in the art and have been reviewed in the literature (for example in Chem. Rev. 2017, 117, 11651 – 11679).

For a reaction with a zinc carbenoid (yk) the zinc carbenoid is generated upon first reacting Et₂Zn with trifluoroacetic acid in a suitable solvent such as absolute dichloromethane at 0 °C followed by the addition of CH₂I₂. Upon addition of the alkene (yh) the preformed zinc carbenoid reacts with the alkene to form the cyclopropane at temperature ranging from 20 – 40 °C (see also WO 2012139775).

Different transition metal carbene complexes (yj) have been found suitable for cyclopropanation reactions. Examples of suitable precursors for such complexes are CuBr, Pd(OAc)₂, Rh(OAc)₄ or iron(III)-5,10,15,20-tetraphenyl-porphyrin (Fe(TPP)Cl).

For a reaction via a palladium carbene complex, a solution of an alkene (yh) in a suitable solvent such as tetrahydrofuran or diethyl ether is treated with a solution of diazomethane in a suitable solvent such as

diethyl ether in the presence of a suitable palladium salt such as Pd(OAc)₂ at temperatures ranging from 0 °C – 20 °C (see also WO 2014023367 or the synthesis of tert-butyl 3-bromo-5-(1-fluorocyclopropyl)benzoate described in this application). A trifluoromethyl substituted cyclopropyl group can be obtained through reaction of an alkene (yh) with iron carbene complexes obtained from in situ generated trifluoromethyl diazomethane and Fe(TPP)Cl as described in Angew. Chem. Int. Ed. 2010, 49, 938-941.

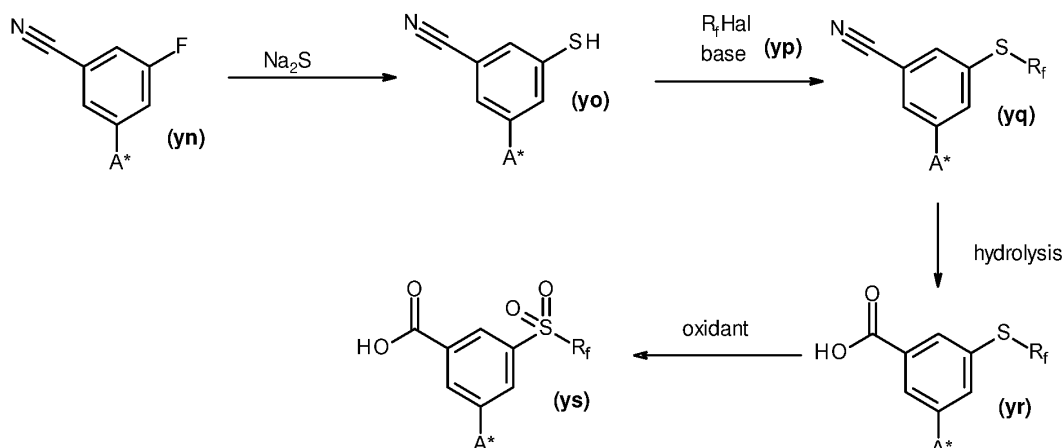
For a reaction with a free carbene (yi), a solution of an alkene (yh) in a suitable solvent is mixed with a carbene precursor from which the free carbene is generated in situ. For example a solution of an alkene (yh) in diglyme is heated in the presence of sodium bromo(difluoro)acetate at temperatures ranging from 60 – 80 °C (see the synthesis of 3-bromo-5-(2,2-difluorocyclopropyl)benzonitrile described in this application). An alternative carbene precursor is for example trimethyl(trifluoromethyl)silane which is used in combination with sodium iodide (as described in WO2017040742).

The final hydrolysis of the cyano groups to the corresponding acid (ym) may be conducted under basic or acidic conditions as described in scheme 12. The hydrolysis of esters may be conducted as described in scheme 11.

The requisite alkenes (yh) and reagents needed for the generation of free carbenes (yi), zinc carbenoids (yk) and certain transition metal carbene complexes (yj) are either commercially available or may be synthesized by methods known to the skilled artisan. For the synthesis of substituted alkenes (yh) via palladium catalyzed coupling reactions see for example WO 2013178362 (1-bromo-3-(1,1-dimethylethyl)-5-(1-methylethenyl)benzene), WO 2012035011 (1,5-dichloro-2-fluoro-3-(3,3,3-trifluoroprop-1-en-2-yl)benzene) and this application (1-bromo-3-(1,1-difluoroethyl)-5-vinylbenzene).

Compounds of formula (ym) in which A* is cyclopropyl optionally substituted with one to two substituent(s) selected from the group of halogen, -CN, methyl, difluoromethyl and trifluoromethyl may be obtained by subjecting the compounds (yl) (Z₁ is -CO₂C₁-C₆alkyl, A* is iodine or bromine) or acids (ym) in which A* is iodine or bromine to the reaction conditions described in scheme 10. In an alternative approach compounds of formula (yh) in which A* corresponds to the alkene fragment -(Z²C=CZ²Z²) may be reacted with 2 equivalents of the free carbenes (yi), zinc carbenoids (yk) and certain transition metal carbene complexes (yj) to give symmetrically substituted acids (ym). In another alternative approach compounds of formula (yl) in which A* is bromine or iodine may be converted to optionally substituted alkenes of formula (yh) via palladium catalyzed couplings reactions (see above) and then reacted to acids of formula (ym) as described in Scheme 21.

Compounds of formula (ys) may be prepared as illustrated in the following scheme 22. R_f is difluoromethyl and trifluoromethyl and A* is chlorine, bromine, iodine, difluoromethyl, trifluoromethyl, difluoromethoxy, trifluoromethoxy. Hal is iodine when R_f is trifluoromethyl and chlorine when R_f is difluoromethyl. If A* is iodine or bromine it can be converted to an optionally substituted cyclopropyl as described in scheme 10.

Scheme 22

An aryl fluoride of formula (yn) is reacted with sodium sulfide to form thiols of formula (yo) described for example in Tetrahedron Letters, 2012, 53(20), 2548-2551. Subsequently, a haloalkylthioether (yq) is formed under alkylation conditions using trifluoromethyl iodide or difluoromethyl chloride and a suitable base. In case of trifluoromethyl iodide an additional catalyst as described for example in WO 2015035223 is used. The nitrile function is then hydrolyzed to form carboxylic acids of formula (yr). In an additional step thioethers of formula (yr) are oxidized to sulfones of formula (ys).

For example, a mixture of an aryl fluoride of formula (yn) and sodium sulfide, is reacted in a suitable solvent, such as N,N-dimethylformamide at temperatures ranging from – 20 to 50 °C. The resulting thiols of formula (yo) are then alkylated with trifluoromethyl iodide in the presence of e.g. triethylamine and 1,1'-dimethyl-4,4'-bipyridinium dichloride in a suitable solvent, such as N,N-dimethylformamide at temperatures ranging from – 20 to 50 °C.

The obtained thioethers of formula (yq) are hydrolyzed either under basic conditions, using for example aqueous sodium hydroxide in a suitable solvent, such as methanol at temperatures ranging from 40 to 100 °C or under acidic conditions in a suitable strong acid, such as sulfuric acid or hydrochloric acid either neat or diluted with a suitable dilutant such as water at temperatures ranging from 40 to 100 °C. The obtained carboxylic acids (yr) are then if necessary and desired, purified using techniques well known in the art, such as chromatography.

A thioether containing compound of formula (yr) is reacted with an oxidizing reagent such as 3-chloroperoxybenzoic acid in a suitable solvent such as dichloromethane or a combination of acetic acid and hydrogen peroxide at temperatures ranging from 0 to 50 °C to form sulfones of formula (ys). The obtained sulfones of formula (ys) are then if necessary and desired, purified using techniques well known in the art, such as chromatography.

The requisite aryl fluorides (yn) are commercially available or may be synthesized by methods known to the skilled artisan.

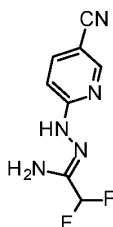
The preparation and use examples which follow illustrate the invention without limiting it.

Preparation examples

5 **Synthesis of 6-[5-(1-Aminoethyl)-3-(difluoromethyl)-1,2,4-triazol-1-yl]pyridine-3-carbonitrile (INT-4)**

Step 1

2-[6-Cyano-3-pyridinyl]hydrazide-2,2-difluoro-ethanimidic acid

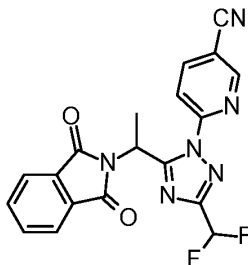


- 10 To 2.33 g (17.4 mmol) 5-hydrazinyl-2-pyridinecarbonitrile in methanol (30 mL) 3.15 g (24.3 mmol) ethyl 2,2-difluoroethanecarboximidate (purchased from Enamine Building Blocks) were added, and the reaction mixture was stirred at room temperature overnight. The solvent was evaporated and the residue was then stirred with *n*-hexane (30 mL) and ethyl acetate (3 mL). The brownish precipitate was separated and dried to obtain 3.38 g (purity: 90,4%; yield: 83.0 %) 2-[6-cyano-3-pyridinyl]hydrazide-2,2-difluoro-ethanimidic acid.
- 15

ESI mass [m/z]: 211.1 [M+H]⁺

Step 2

6-[3-(difluoromethyl)-5-[1-(1,3-dioxoisindolin-2-yl)ethyl]-1,2,4-triazol-1-yl]pyridine-3-carbonitrile



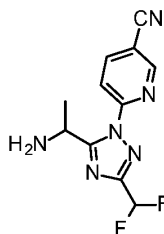
- 20 To 3.28 g (14.0 mmol) 2-[6-cyano-3-pyridinyl]hydrazide-2,2-difluoro-ethanimidic acid in pyridine (20 mL), 3.32 g (14.0 mmol) (αS)-1,3-dihydro-α-methyl-1,3-dioxo-2*H*-isindole-2-acetyl chloride (see

preparation from (αS)-1,3-dihydro- α -methyl-1,3-dioxo-2*H*-isoindole-2-acetic acid (Pht-Ala-OH purchased from ABCR) and oxalyl chloride: D. A. Gruzdev *et al.*, Tetrahedron: Asymmetry, 21(8), 936-942, 2010) were added, and the reaction mixture was stirred at room temperature overnight. Then water (200 mL) was added and the mixture was extracted with dichloromethane (200 mL). The organic phase was extracted twice with a saturated aqueous NaHCO₃ solution (100 mL), dried over Na₂SO₄, and evaporated under reduced pressure. The remaining solid residue was chromatographed with a cyclohexane/acetone gradient on silica gel to afford 1.09 g (purity: 95.7%; yield: 18.8 %) of the racemic title compound as a colorless solid.

ESI mass [m/z]: 395.2 [M+H]⁺

10 **Step 3**

6-[5-(1-aminoethyl)-3-(difluoromethyl)-1,2,4-triazol-1-yl]pyridine-3-carbonitrile (**INT-4**)



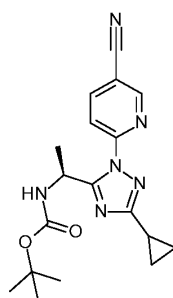
To 1.0 g (2.5 mmol) 6-[3-(difluoromethyl)-5-[1-(1,3-dioxoisoinolin-2-yl)ethyl]-1,2,4-triazol-1-yl]pyridine-3-carbonitrile in ethanol (20 mL), 577 mg (6.34 mmol) hydrazine-hydrate were added, and the reaction mixture was heated under reflux. After 30 minutes a colorless precipitate was formed. The reaction mixture was stirred and heated under reflux one additional hour, acetone (15 mL) was added and the heating was continued for further 30 minutes. The reaction mixture was concentrated and the solid residue was treated with ethanol. After filtration, the filtrate was evaporated under reduced pressure to afford 663 mg of the racemic 6-[5-(1-aminoethyl)-3-(difluoromethyl)-1,2,4-triazol-1-yl]pyridine-3-carbonitrile, which was used without further purification.

ESI mass [m/z]: 265.2 [M+H]⁺

Synthesis of 6-{5-[(1*S*)-1-aminoethyl]-3-cyclopropyl-1*H*-1,2,4-triazol-1-yl}nicotinonitrile hydrochloride (1:1) (INT-3**)**

Step 1

25 *tert*-butyl {(1*S*)-1-[1-(5-cyanopyridin-2-yl)-3-cyclopropyl-1*H*-1,2,4-triazol-5-yl]ethyl}carbamate



To a solution of 2.0 g (10.5 mmol) N-(tert-butoxycarbonyl)-L-alanine in N,N-dimethylformamide (37.5 ml) was added 1.91 g (15.9 mmol) cyclopropylamidin followed by 4.42 g (11.63 mmol) of HATU and 5.52 ml (31.7 mmol) of N,N-diisopropylethylamin and the reaction mixture was stirred at room temperature for 3 h. Afterwards 6.05 ml (105.7 mmol) of acetic acid and 2.13 g (15.8 mmol) of 6-hydrazinonicotinonitrile were added and the reaction mixture was stirred 5 h at 80 °C and then at room temperature overnight. The reaction mixture was cooled to room temperature, a saturated aqueous solution of Na₂CO₃ was added and then the mixture was extracted with EtOAc. The combined organic layers were washed with water, an aqueous solution of 5% NaH₂PO₄, brine and finally dried over Na₂SO₄. After filtration and evaporation of the solvent under vacuo the crude was purified by preparative HPLC (water/acetonitrile). The combined product fractions were evaporated to yield the title compound (0.77 g, 21%).

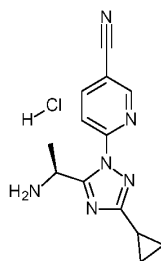
ESI mass [m/z]: 355.3 [M+H]⁺

¹H-NMR peaklist (400.2 MHz, CD₃CN):

15 δ= 8.8116 (6.3); 8.8100 (6.6); 8.8062 (6.7); 8.8046 (5.9); 8.2628 (4.8); 8.2573 (4.6); 8.2412 (5.5);
8.2357 (5.4); 7.9980 (7.0); 7.9964 (6.6); 7.9764 (5.9); 7.9747 (5.6); 5.8766 (0.8); 5.7388 (0.5); 5.7213
(1.4); 5.7031 (1.9); 5.6849 (1.3); 5.6682 (0.4); 2.1614 (41.0); 2.0585 (1.0); 2.0462 (2.0); 2.0378 (2.2);
2.0344 (1.4); 2.0255 (3.5); 2.0194 (1.3); 2.0132 (2.1); 2.0049 (2.2); 1.9926 (1.1); 1.9648 (4.6); 1.9528
(18.4); 1.9467 (34.9); 1.9405 (49.0); 1.9343 (33.6); 1.9281 (17.1); 1.4498 (14.5); 1.4328 (14.5); 1.3608
20 (16.0); 1.2685 (1.1); 1.2388 (0.7); 1.1974 (0.7); 1.0334 (0.4); 1.0281 (0.4); 1.0173 (1.7); 1.0106 (5.0);
1.0083 (4.1); 1.0049 (6.3); 0.9990 (1.5); 0.9901 (6.0); 0.9848 (6.4); 0.9754 (1.5); 0.9669 (2.7); 0.9546
(1.2); 0.9464 (3.2); 0.9446 (3.1); 0.9403 (2.8); 0.9385 (2.8); 0.9342 (3.2); 0.9324 (3.0); 0.9274 (4.6);
0.9204 (2.9); 0.9154 (3.3); 0.9084 (2.6); 0.9047 (1.3); 0.9014 (1.4); 0.8974 (1.0); 0.8927 (0.8); 0.8872
(0.7); 0.8837 (0.6); 0.1459 (0.8); 0.0080 (6.7); -0.0002 (166.9); -0.0086 (6.2); -0.0171 (0.6); -0.1495
25 (0.8)

Step 2

6-{5-[(1S)-1-aminoethyl]-3-cyclopropyl-1H-1,2,4-triazol-1-yl}nicotinonitrile hydrochloride (1:1) (**INT-3**)



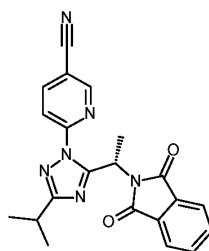
- 5 A solution of 830 mg (2.34 mmol) tert-butyl {(1S)-1-[1-(5-cyanopyridin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethyl}carbamate in dioxane (22 ml) was treated with HCl 4N in dioxane (10.9 ml). The reaction mixture was stirred at room temperature overnight. The resulting precipitate was separated by filtration and dried under air to yield the title compound (0.71, 100%).

ESI mass [m/z]: 255.1 [amine+H]⁺

- 10 **Synthesis of 3-chloro-N-[(1S)-1-[1-(5-cyanopyridin-2-yl)-3-isopropyl-1H-1,2,4-triazol-5-yl]ethyl]-5-[(2,2,2-trifluoroethyl)sulfonyl]benzamide (example I-21)**

Step 1

6-{5-[(1S)-1-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)ethyl]-3-isopropyl-1H-1,2,4-triazol-1-yl}nicotinonitrile



15

A solution of 5.00 g (95% purity, 21.6 mmol) (2S)-2-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)propanoic acid and 0.08 mL (1 mmol) DMF in 30 mL absolute CH₂Cl₂ was treated with 3.78 mL (43.3 mmol) oxalyl chloride at 0 °C. The reaction mixture was stirred for 2 d at ambient temperature. All volatiles were removed under reduced pressure and the residue used for the next step without further purification.

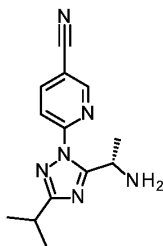
- 20 To a solution of 3.13 g (95% purity, 21.6 mmol) methyl 2-methylpropanimidate hydrochloride (1:1) in 40 mL absolute THF were added at 0 °C 15.1 mL (86.4 mmol) absolute DIPEA. The acid chloride prepared

in the first step was dissolved in 20 mL absolute THF and added dropwise within 25 min to the solution of the imidate. After 30 min stirring at 0 °C 3.19 g (23.7 mmol) 6-hydrazinonicotinonitrile and 10 mL absolute THF were added. The reaction mixture was stirred for 30 min at 0 °C and overnight at ambient temperature. All volatiles were removed under reduced pressure. To the residue were added 200 mL water and the mixture was extracted with 200 mL EtOAc. The phases were separated and the aqueous phase extracted several times with EtOAc. The combined organic phases were washed with brine and dried over Na₂SO₄. The solvent was removed under reduced pressure and the residue purified by chromatography on silica (cyclohexane / ethyl acetate) to provide 5.57 g of 6-{5-[(1S)-1-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)ethyl]-3-isopropyl-1H-1,2,4-triazol-1-yl}nicotinonitrile.

ESI mass [m/z]: 387.5 [M+H]⁺

Step 2

6-{5-[(1S)-1-aminoethyl]-3-isopropyl-1H-1,2,4-triazol-1-yl}nicotinonitrile (**INT-2**)

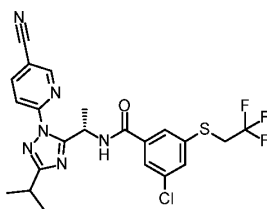


A solution of 2.00 g (5.17 mmol) 6-{5-[(1S)-1-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)ethyl]-3-isopropyl-1H-1,2,4-triazol-1-yl}nicotinonitrile and 0.38 mL hydrazine hydrate in 40 mL ethanol was heated for 2 h at 80 °C. The resulting suspension was stirred overnight at ambient temperature and then cooled to 10 °C. The mixture was filtered and the residue washed with ice-cold ethanol. The filtrate was concentrated under reduced pressure to yield 1.57 g (70% pure) of 6-{5-[(1S)-1-aminoethyl]-3-isopropyl-1H-1,2,4-triazol-1-yl}nicotinonitrile.

ESI mass [m/z]: 257.2 [M+H]⁺

Step 3

3-chloro-N-{(1S)-1-[1-(5-cyanopyridin-2-yl)-3-isopropyl-1H-1,2,4-triazol-5-yl]ethyl}-5-[(2,2,2-trifluoroethyl)sulfanyl]benzamide

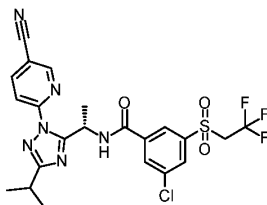


A mixture of 98 mg (0.36 mmol) 3-chloro-5-[(2,2,2-trifluoroethyl)sulfonyl]benzoic acid, 249 mg (0.65 mmol) HATU, 0.15 mL (1.1 mmol) *N*-ethyl-diisopropylamine and 2 mL DMF was stirred for 60 min at room temperature. 120 mg (70% purity, 0.32 mmol) 6-{5-[(1*S*)-1-aminoethyl]-3-isopropyl-1*H*-1,2,4-triazol-1-yl}nicotinonitrile were added and the mixture stirred overnight. Water was added and the mixture extracted repeatedly with EtOAc. The combined organic phases were washed with brine and dried over Na₂SO₄. The solvent was removed under reduced pressure and purified by reversed phase chromatography (H₂O / acetonitrile) to provide 166 mg 3-chloro-*N*-{[(1*S*)-1-[1-(5-cyanopyridin-2-yl)-3-isopropyl-1*H*-1,2,4-triazol-5-yl]ethyl]-5-[(2,2,2-trifluoroethyl)sulfonyl]benzamide.

ESI mass [m/z]: 509.3 [M+H]⁺

Step 4

3-chloro-*N*-{[(1*S*)-1-[1-(5-cyanopyridin-2-yl)-3-isopropyl-1*H*-1,2,4-triazol-5-yl]ethyl]-5-[(2,2,2-trifluoroethyl)sulfonyl]benzamide (**example I-21**)



A solution of 165 mg (0.32 mmol) 3-chloro-*N*-{[(1*S*)-1-[1-(5-cyanopyridin-2-yl)-3-isopropyl-1*H*-1,2,4-triazol-5-yl]ethyl]-5-[(2,2,2-trifluoroethyl)sulfonyl]benzamide in 2 mL CH₂Cl₂ was treated at 0 °C with 168 mg (70% purity, 0.68 mmol) *m*-chloroperoxybenzoic acid. The mixture was stirred for 3 h at 0 °C and then another 84 mg (70 % purity, 0.34 mmol) *m*-chloroperoxybenzoic acid were added. The reaction mixture was stirred further for 2 h at 0 °C and overnight at ambient temperature. 3 mL of a saturated aqueous NaHCO₃ solution were then added and the mixture stirred for 1 h at ambient temperature. The layers were separated and the aqueous phase repeatedly extracted with CH₂Cl₂. The combined organic phases were washed with brine and dried over Na₂SO₄. The solvent was removed under reduced pressure and purified by reversed phase chromatography (H₂O / acetonitrile) to provide 124 mg 3-chloro-*N*-{[(1*S*)-1-[1-(5-cyanopyridin-2-yl)-3-isopropyl-1*H*-1,2,4-triazol-5-yl]ethyl]-5-[(2,2,2-trifluoroethyl)sulfonyl]benzamide.

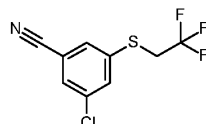
¹H-NMR (400 MHz, d₆-DMSO): see NMR peak list in table 1

ESI mass [m/z]: 541.4 [M+H]⁺

Synthesis of 3-chloro-5-[(2,2,2-trifluoroethyl)sulfanyl]benzoic acid

Step 1

5 3-chloro-5-[(2,2,2-trifluoroethyl)sulfanyl]benzonitrile



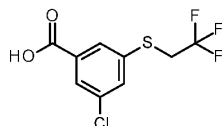
- To a suspension of 310 mg (7.1 mmol) sodium hydride (55% suspension in mineral oil) in 15 mL anhydrous *N,N*-dimethylformamide were added carefully 0.74 mL (8.35 mmol) 2,2,2-trifluoroethanethiol. After 30 min stirring at room temperature 1.00 g (6.42 mmol) 3-chloro-5-fluorobenzonitrile was added.
- 10 The mixture was stirred for 5 hrs at room temperature and then quenched by the addition of 1 mL H₂O and 1.1 mL glacial acetic acid. The volatiles were removed under reduced pressure. Water and ethyl acetate were added to the residue. The layers were separated and the aqueous layer repeatedly extracted with ethyl acetate. The combined organic layers were washed with brine and then dried with Na₂SO₄. The solvent was removed under reduced pressure and the residue purified by reversed phase chromatography
- 15 (H₂O/acetonitrile) to provide 1.40 g of 3-chloro-5-[(2,2,2-trifluoroethyl)sulfanyl]benzonitrile.

¹H-NMR (400 MHz, d₆-DMSO): 8.03 – 8.02 (m, 1H), 7.98 – 7.97 (m, 1H), 7.93 – 7.92 (m, 1H), 4.30 – 4.22 (q, 2H).

ESI mass [m/z]: 252.1 [M+H]⁺

Step 2

20 3-chloro-5-[(2,2,2-trifluoroethyl)sulfanyl]benzoic acid



- A mixture of 800 mg (3.17 mmol) 3-chloro-5-[(2,2,2-trifluoroethyl)sulfanyl]benzonitrile, 3.5 mL water and 3.6 mL concentrated sulfuric acid was heated 2 days at 100 °C. Water and ethyl acetate were added. The layers were separated and the aqueous layer repeatedly extracted with ethyl acetate. The combined
- 25 organic layers were washed with brine and then dried with Na₂SO₄. The solvent was removed under

reduced pressure and the residue purified by reversed phase chromatography (H₂O/acetonitrile) to provide 728 mg of 3-chloro-5-[(2,2,2-trifluoroethyl)sulfanyl]benzoic acid.

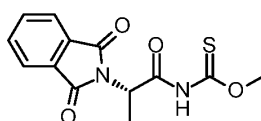
¹H-NMR (400 MHz, d₆-DMSO): 13.55 (s, 1H, COOH), 7.91 – 7.89 (m, 2H), 7.77 – 7.76 (m, 1H), 4.23 – 4.16 (q, 2H).

5 ESI mass [m/z]: 269.0 [M-H]⁻

Synthesis of 6-{5-[(1S)-1-aminoethyl]-3-methoxy-1H-1,2,4-triazol-1-yl}nicotinonitrile

Step 1

O-methyl [(2S)-2-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)propanoyl]carbamothioate

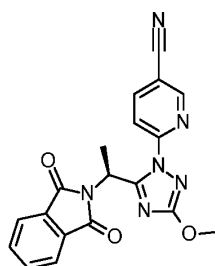


- 10 To a solution of 1.0 g (4.6 mmol) (2S)-2-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl) propanoic acid in toluene (15 ml) was added 0.80 ml (9.12 mmol) oxalyl chloride and one drop of N,N-dimethylformamide. The reaction mixture was stirred 3 h at room temperature and then hexane (15 ml) was added and the stirring was continued overnight. After this time additional oxalyl chloride (0.5 ml) was added again and the reaction mixture was stirred 3 h and finally was evaporated. The crude residue was dissolved in acetone
- 15 (15 ml) and then 0.44 g (4.56 mmol) KSCN were added as a solution in acetone (5 ml) and the mixture was stirred at 60 °C for 2 h. Then 0.46 ml (11.4 mmol) of methanol were added and the mixture was stirred at 60 °C overnight, cooled to room temperature and evaporated under reduced pressure. The resulting residue was dissolved in EtOAc, washed with water and brine respectively and finally the organic layer was dried over anhydrous Na₂SO₄ and then concentrated under reduced pressure. The crude product was
- 20 purified by silica gel chromatography to yield the tittle compound (0.82 g, 59%).

ESI mass [m/z]: 293.1 [M+H]⁺

Step 2

6-{5-[(1S)-1-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)ethyl]-3-methoxy-1H-1,2,4-triazol-1-yl}nicotinonitrile

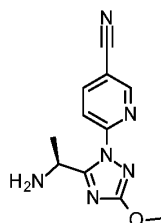


To a solution of 1.5 g (5.1 mmol) O-methyl [(2S)-2-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)propanoyl]carbamothioate in ethanol (30 ml) were added 0.69 g (5.1 mmol) 6-hydrazinonicotinonitrile and the reaction mixture was stirred at 90 °C overnight. The mixture was cooled to room temperature, evaporated under reduced pressure and the resulting residue was dissolved in EtOAc, washed with water and brine respectively. The organic layer was dried over anhydrous Na₂SO₄ and then concentrated under reduced pressure. The crude product was purified by silica gel chromatography to yield the title compound (1.23 g, 58%).

ESI mass [m/z]: 375.1 [M+H]⁺

Step 3

6-{5-[(1S)-1-aminoethyl]-3-methoxy-1H-1,2,4-triazol-1-yl}nicotinonitrile (INT-5)



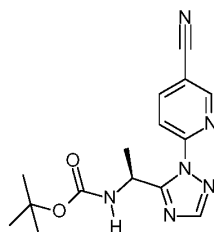
To a solution of 1.20 g (3.20 mmol) 6-{5-[(1S)-1-(1,3-dioxo-1,3-dihydro-2H-isoindol-2-yl)ethyl]-3-methoxy-1H-1,2,4-triazol-1-yl}nicotinonitrile in ethanol (30 ml) were added 0.39 ml (8.01 mmol) hydrazin hydrate and the reaction was heated to reflux temperature overnight. After cooling the mixture to room temperature, acetone (10 ml) was added and it was heated again to reflux temperature for 3 h. The resulting precipitate was filtered and the filtrate evaporated under reduced pressure to yield a residue which was used in the next step without further purification (1.05 g, 44 % purity, 59% yield).

ESI mass [m/z]: 245.1 [M+H]⁺

Following the above described procedure also (1S)-1-[1-(5-chloropyrimidin-2-yl)-3-methoxy-1H-1,2,4-triazol-5-yl]ethanamine (INT-22) and (1S)-1-[1-(5-chloropyridin-2-yl)-3-methoxy-1H-1,2,4-triazol-5-yl]ethanamine (INT-18) were obtained. In these cases, the crude amines obtained after removal of the phthalimide protecting group were purified by reversed phase chromatography (H₂O / acetonitrile / formic acid) and the purified amines were isolated as their formic acid salts. To provide the free amines the formic acid salts were mixed with sat. aq. NaHCO₃ solution. Repeated extraction of this mixture with ethyl acetate, drying of the combined organic layers with Na₂SO₄ and removal of the solvent under reduced pressure led to the isolation of the free amines (INT-22) and (INT-18) which were used for the synthesis of the example compounds.

Synthesis of 6-{5-[(1S)-1-aminoethyl]-1H-1,2,4-triazol-1-yl}nicotinonitrile hydrochloride**Step 1**

tert-butyl {(1S)-1-[1-(5-cyanopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}carbamate



- 5 To a solution of 2.00 g (10.6 mmol) N²-(tert-butoxycarbonyl)-L-alaninamide in 40 mL CH₂Cl₂ was added 2.1 mL (16 mmol) N,N-dimethylformamide dimethylacetal. The solution was heated at reflux for 2 h after which the solvent was removed under reduced pressure. The residue was dissolved in a mixture of 20 mL 1,4-dioxane and 20 mL glacial acetic acid. 1.7 g (13 mmol) 6-hydrazinonicotinonitrile was added and the mixture stirred at 50 °C for 60 min. The solvents were removed under reduced pressure, a saturated aqueous solution of NaHCO₃ was added and the mixture repeatedly extracted with ethyl acetate. The combined organic layers were washed with brine, dried with Na₂SO₄ and the solvent was removed under reduced pressure. The residue was purified by reversed phase chromatography (H₂O / acetonitrile) to provide 3.0 g of tert-butyl {(1S)-1-[1-(5-cyanopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}carbamate.

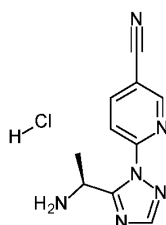
$[\alpha]_D^{20} = +89$ (c = 1.0; ethanol)

- 15 ¹H NMR (DMSO-d₆, 400 MHz): 9.10 (s, 1 H), 8.57 (dd, 1 H), 8.21 (s, 1H), 8.05 (d, 1H), 7.52 (d, 1H), 5.63 (m, 1 H), 1.43 (d, 3 H), 1.31 (s, 9 H).

ESI mass [m/z]: 259.2 [M-C₄H₈+H]⁺

Step 2

6-{5-[(1S)-1-aminoethyl]-1H-1,2,4-triazol-1-yl}nicotinonitrile hydrochloride



20

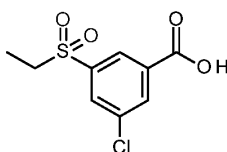
To a solution of 2.9 g (9.2 mmol) tert-butyl {(1S)-1-[1-(5-cyanopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}carbamate in 40 mL 1,4-dioxane were added 23 mL of a 4 M solution of HCl in 1,4-dioxane. The

mixture was stirred for 4 h at 50 °C and overnight at room temperature. The solvent was removed under reduced pressure to provide 2.81 g of a residue containing 6-{5-[(1S)-1-aminoethyl]-1H-1,2,4-triazol-1-yl}nicotinonitrile hydrochloride. This was used without further purification.

¹H NMR (DMSO-d₆, 400 MHz): 9.11 (d, 1H), 8.80 (*br* d, 3H), 8.61 (dd, 1H), 8.45 (s, 1H), 8.13 (d, 1H),
5 5.39 (m, 1H), 1.63 (d, 3H).

ESI mass [m/z]: 215.2 [amine+H]⁺

Synthesis of 3-chloro-5-(ethylsulfonyl)benzoic acid

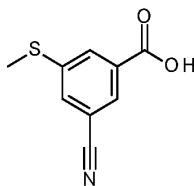


A mixture of 0.47 g (4.1 mmol) proline and 0.16 g (4.0 mmol) sodium hydroxide in 24 mL
10 dimethylsulfoxide was degassed for 30 min by purging with argon. 1.5 g (5.1 mmol) methyl 3-chloro-5-iodobenzoate, 4.7 g (40 mmol) sodium ethanesulfinate and 0.77 g (4.0 mmol) copper(I) iodide were added and the mixture further purged with argon for 5 min. The mixture was stirred at 120 °C for 3 h, cooled to room temperature and then treated with 4 mL of a 2 M aqueous sodium hydroxide solution. It was further stirred overnight at room temperature, cooled to 10 °C and acidified to pH 1 using concentrated
15 hydrochloric acid. Water was added to the mixture and it was extracted with ethyl acetate. The layers were separated and the aqueous layer repeatedly extracted with ethyl acetate. The combined organic layers were washed twice with water and once with brine and then dried with Na₂SO₄. The solvent was removed under reduced pressure and the residue purified by reversed phase chromatography (H₂O / acetonitrile) to provide 1.00 g of 3-chloro-5-(ethylsulfonyl)benzoic acid.

20 ¹H NMR (DMSO-d₆, 400 MHz): NMR peaklist: δ= 8.2775 (3.5); 8.2737 (6.8); 8.2698 (4.4); 8.2363 (3.4); 8.2325 (4.0); 8.2313 (5.4); 8.2277 (4.1); 8.2090 (4.8); 8.2046 (5.5); 8.1997 (3.0); 3.4891 (1.9); 3.4707 (6.7); 3.4523 (6.8); 3.4340 (2.1); 3.3298 (6.3); 2.5259 (0.6); 2.5212 (1.0); 2.5125 (17.2); 2.5080 (36.2); 2.5034 (48.0); 2.4988 (33.7); 2.4942 (15.6); 1.1402 (6.9); 1.1219 (16.0); 1.1034 (6.8); -0.0002 (5.1).

25 ESI mass [m/z]: 247.1 [M-H]⁻

Synthesis of 3-cyano-5-(methylsulfonyl)benzoic acid



To a solution of 1.00 g (6.06 mmol) 3-cyano-5-fluorobenzoic acid in 20 mL anhydrous *N,N*-dimethylformamide were added 363 mg (9.0 mmol) sodium hydride (55% suspension in mineral oil) and 849 mg (12.1 mmol) sodium methanethiolate. The mixture was stirred for 90 min at room temperature and overnight at 80 °C. The mixture was quenched by the addition of water. EtOAc was added and the layers were separated. The pH of the aqueous layer was adjusted to pH 1 by the addition of hydrochloric acid and the mixture repeatedly extracted with EtOAc. The combined organic layers were washed with brine and then dried with Na₂SO₄. The solvent was removed under reduced pressure and the residue purified by reversed phase chromatography (H₂O/acetonitrile) to provide 688 mg of 3-cyano-5-(methylsulfanyl)benzoic acid.

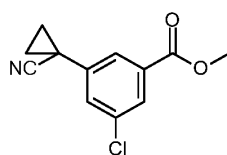
¹H-NMR (400 MHz, d₆-DMSO): 13.6 (brs, 1H), 8.03 – 7.95 (m, 3H), 2.58 (s, 3H).

ESI mass [m/z]: 192.1 [M-H]⁻

Synthesis of 3-chloro-5-(1-cyanocyclopropyl)benzoic acid

Step 1:

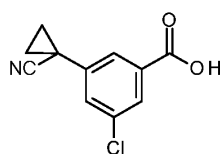
methyl 3-chloro-5-(1-cyanocyclopropyl)benzoate



To a solution of methyl 3-chloro-5-(cyanomethyl)benzoate (12 g, 57 mmol) in 1,2-dibromoethane (150 mL) was added NaOH (4.58 g, 114 mmol) and benzyl(trimethyl)ammonium chloride (10.32 g, 68.69 mmol) in one portion at 25 °C. The mixture was stirred at 65 °C for 12 h and afterwards diluted with a saturated aqueous solution of NH₄Cl (50 mL) and extracted with EtOAc (50 mL). The aqueous phase was extracted two times with EtOAc (30 mL). The combined organic layers were dried over Na₂SO₄ and filtered. The solvent was removed under reduced pressure. The residue was purified by column chromatography (silica gel, petrol ether/EtOAc = 100/1 to 20/1) to give methyl 3-chloro-5-(1-cyanocyclopropyl)benzoate (8.8 g, 65% yield) as yellow oil.

Step 2:

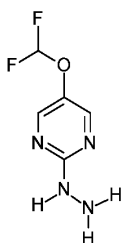
3-chloro-5-(1-cyanocyclopropyl)benzoic acid



To a solution of methyl 3-chloro-5-(1-cyanocyclopropyl)benzoate (8.8 g, 37 mmol) in THF (100 mL) was added TMSOK (6.71 g, 52.3 mmol) in one portion at 25 °C. The mixture was stirred at 25 °C for 12 h. The reaction suspension was adjusted to pH = 5 to 6 with 1 M hydrochloric acid. The color of the suspension turned to orange. The mixture was diluted with H₂O (15 mL). The water was extracted three times with EtOAc (50 mL). The combined organic layers were dried over Na₂SO₄, filtered and concentrated under reduce pressure to give 3-chloro-5-(1-cyanocyclopropyl)benzoic acid (5.05 g, 61% yield) as light yellow solid.

¹H-NMR (400 MHz, MeOD): δ, 7.85 – 7.93 (m, 2H), 7.55 (t, *J*=1.9 Hz, 1 H), 1.78 - 1.82 (m, 2 H), 1.55 - 1.59 (m, 2 H). Measured using a Varian 400MR NMR machine.

10 Synthesis of 5-(difluoromethoxy)-2-hydrazinopyrimidine



A solution of 500 mg (2.60 mmol) 5-(difluoromethoxy)-2-(methylsulfanyl)pyrimidine in 2 mL ethanol was treated with 0.52 mL (11 mmol) of hydrazine hydrate. The mixture was heated to reflux overnight. The reaction mixture was then cooled to 5 °C upon which a white precipitate formed. The suspension was filtered and the precipitate washed with ethanol. The residue was dried under reduced pressure to provide 125 mg of 5-(difluoromethoxy)-2-hydrazinopyrimidine.

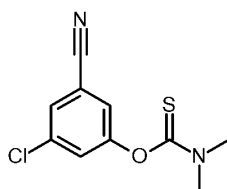
¹H NMR (DMSO-d₆, 400 MHz): 8.35 (s, 1 H), 8.28 (s, 2 H), 7.06 (t, *J* = 74 Hz, 1 H), 4.17 (*br s*, 2 H).

ESI mass [*m/z*]: 177.2 [*M*+H]⁺

Synthesis of 3-chloro-5-(difluoromethyl)benzoic acid (INT-09)

20 Step 1:

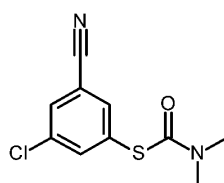
O-(3-chloro-5-cyanophenyl) dimethylcarbamothioate



- 38.9 mL (279 mmol) triethylamine, 1.14 g (9.3 mmol) N,N-dimethylpyridin-4-amine (DMAP) and 13.8 g (112 mmol) dimethylcarbamothioyl chloride were successively added to a vigorously stirred suspension of 14.3 g (93 mmol) 3-chloro-5-hydroxybenzonitrile in 450 mL anhydrous EtOAc. The reaction mixture was brought to 55-60°C and was stirred at this temperature for 24 h. After cooling down to room temperature the reaction mixture was washed with 450 mL water and 450 mL brine. The organic layer was separated, dried over Na₂SO₄ and filtered. The filtrate was concentrated in vacuo to a volume of about 50 mL. The concentrate was diluted with 150 mL n-hexane, the precipitate formed was filtered off, washed with 150 mL of a 1:1 mixture diethyl ether and n-hexane and vacuum dried at 60 °C (1 tor, 3 h) to give 9.3 g (86%) of O-(3-chloro-5-cyanophenyl) dimethylcarbamothioate as colorless crystals.
- ¹H NMR (400 MHz, CDCl₃) δ: 3.35 (s, 3H), 3.46 (s, 3H), 7.30 (s, 1H), 7.35 (s, 1H), 7.53 (s, 1H) (measured on a Varian Gemini 2000 machine).

Step 2:

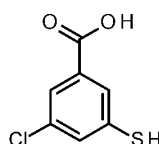
S-(3-chloro-5-cyanophenyl) dimethylcarbamothioate



- A solution of 2.41 g (10 mmol) O-(3-chloro-5-cyanophenyl) dimethylcarbamothioate in 20 mL anhydrous dimethyl acetamide was heated in a Biotage Initiator microwave for 35 min at 220 °C. The reaction mixture was brought to room temperature and diluted with water 40 mL. The precipitate formed was filtered off, washed with hot (ca. 70 °C) water and n-hexane and vacuum dried at 60 °C (1 tor, 3 h) to give 2.05 g (85%) of S-(3-chloro-5-cyanophenyl) dimethylcarbamothioate as a white powder.
- ¹H NMR (400 MHz, CDCl₃) δ: 3.05 (s, 3H), 3.10 (s, 3H), 7.64 (s, 1H), 7.69 (s, 1H), 7.73 (s, 1H) (measured on a Varian Gemini 2000 machine).

Step 3:

3-chloro-5-sulfanylbzoic acid

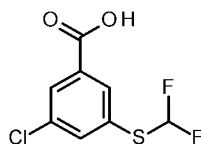


A hot (ca. 70 °C) solution of 68.5 g (1.71 mol) NaOH in 300 mL water was added to a suspension of 27.5 g (114 mmol) S-(3-chloro-5-cyanophenyl) dimethylcarbamothioate in 700 mL warm (ca. 40 °C) methanol. The reaction mixture was stirred under reflux (20 h). Methanol was removed in vacuo and the aqueous solution was washed with 2 x 200 mL diethyl ether. The aqueous layer was separated and added dropwise to a suspension of 300 g ice in concentrated aqueous HCl (under argon, cooling with ice bath). The solution formed was filtered off, washed with 2 x 50 mL water, 50 mL n-hexane and vacuum dried at 60 °C (1 tor, 3 h) to give 21.2 g (98%) of 3-chloro-5-sulfanylbenzoic acid as a white powder.

¹H NMR (400 MHz, CDCl₃) δ: 3.65 (s, 1H), 7.50 (s, 1H), 7.86 (s, 1H), 7.89 (s, 1H), 10.80 (brs, 1H) (measured on a Varian Gemini 2000 machine).

10 **Step 4:**

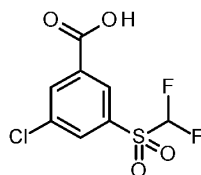
3-chloro-5-[(difluoromethyl)sulfanyl]benzoic acid (**INT-09**)



12.44 g (90 mmol) K₂CO₃ and 18.3 g (120 mmol) sodium chloro(difluoro)acetate were added successively to a solution of 11.32 g (60 mmol) 3-chloro-5-sulfanylbenzoic acid in anhydrous DMF under argon atmosphere. The reaction mixture was stirred at 95 - 100 °C for 3 h. Caution: At 90 - 95 °C CO₂ evolved vigorously! The volatiles were removed in vacuo and the residue was diluted with water to a volume of 500 ml. The product was extracted with diethyl ether. The aqueous layer was separated and added dropwise to the suspension of ca. 100 g ice in 200 mL 5% hydrochloric acid. The suspension was stirred at room temperature for 20 h, the precipitate was filtered off, washed with water 2 x 50 mL and a 1/1 mixture of n-hexane and diethylether. Vacuum drying 60 °C (1 tor, 3 h) gave 11 g of crude product (85% pure according to ¹H and ¹⁹F NMR). Sublimation at 90 - 95 °C/0.01 tor afforded 7.7 g (54%) of 3-chloro-5-[(difluoromethyl)sulfanyl]benzoic acid as white powder.

¹H NMR (400 MHz, CDCl₃) δ: 6.90 (t, 1H, J = 74.4 Hz), 7.83 (t, 1H, J = 2 Hz), 8.14 (t, 1H, J = 2 Hz), 8.20 (s, 1H), 10.50 (br s, 1H). (measured on a Varian Gemini 2000 machine).

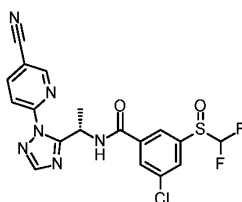
25 **Synthesis of 3-chloro-5-[(difluoromethyl)sulfonyl]benzoic acid (INT-16)**



Oxone (41.2 g, 67 mmol) was added in one portion to a stirred solution of 3-chloro-5-[(difluoromethyl)sulfinyl]benzoic acid (8.0 g, 33.5 mmol) in methanol (200 ml) and water (50 ml). The reaction mixture was stirred at 25°C for 48 h. It was filtered and the filter-cake was washed with methanol. The combined filtrates were concentrated *in vacuo* and diluted with water. A white precipitate formed which was filtered, washed with water and dried in oven at 100°C to give 8.95 g of a white powder which was purified on CombiFlash to give 6.9 g (76%) of the pure acid.

¹H-NMR (400 MHz, CD₃OD) δ = 8.46 (s, 1H), 8.39 (s, 1H), 8.07 (s, 1H), 6.85 (t, 1H, *J* = 52.7 Hz). (measured on a Varian Gemini 2000 machine).

Synthesis of 3-chloro-N-{(1S)-1-[1-(5-cyanopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}-5-[(difluoromethyl)sulfinyl]benzamide (example II-18)



A solution of 100 mg (0.20 mmol) 3-chloro-N-{(1S)-1-[1-(5-cyanopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}-5-[(difluoromethyl)sulfinyl]benzamide in 2 mL CH₂Cl₂ was treated at 0 °C with 62 mg (70% purity, 0.20 mmol) *m*-chloroperoxybenzoic acid. The mixture was stirred for 4 h at 0 °C and overnight at room temperature. 5 mL of a saturated aqueous NaHCO₃ solution were then added and the mixture stirred for 30 min at ambient temperature. The layers were separated and the aqueous phase repeatedly extracted with EtOAc. The combined organic phases were washed with brine and dried over Na₂SO₄. The solvent was removed under reduced pressure and the residue purified by reversed phase chromatography (H₂O / acetonitrile) to provide 88 mg 3-chloro-N-{(1S)-1-[1-(5-cyanopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}-5-[(difluoromethyl)sulfinyl]benzamide.

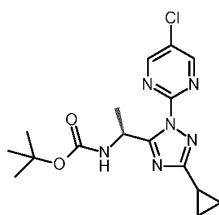
¹H-NMR (400 MHz, d₆-DMSO): see NMR peak list in table 3

ESI mass [*m/z*]: 451.1 [*M*+H]⁺

Synthesis of (1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethanamine hydrochloride (1:1) (INT-8)

Step 1:

tert-butyl {(1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethyl}carbamate



To a solution of 3.0 g (15.8 mmol) N-(tert-butoxycarbonyl)-L-alanine in THF (65 mL) were added 3.56 g (23.7 mmol) ethyl cyclopropanecarboximidate hydrochloride (1:1), 6.61 g (17.4 mmol) HATU and 10.25 ml (79.2 mmol) N,N-diisopropylethylamin. The reaction mixture was stirred at room temperature for 3 h.

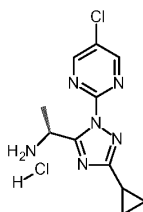
5 Afterwards 4.31 g (23.7 mmol) 5-chloro-2-hydrazinopyrimidine hydrochloride (1:1) were added and the reaction mixture was stirred overnight at room temperature. A saturated aqueous solution of NaHCO₃ was added and then the mixture was extracted with EtOAc. The combined organic layers were washed with water and brine. All volatiles were removed under reduced pressure and the residue was purified twice by reversed phase chromatography (H₂O / acetonitrile) to provide 1.1 g tert-butyl ((1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethyl)carbamate.

10

ESI mass [m/z]: 365.4 [M+H]⁺

Step 2:

(1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethanamine hydrochloride (1:1) (INT-8)



15

To a solution of 1.10 g (3.01 mmol) tert-butyl ((1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethyl)carbamate in 20 mL 1,4-dioxane were added 7.54 mL (30.1 mmol) of a 4 M solution of HCl in 1,4-dioxane. The mixture was stirred overnight at 50 °C. The solvent was removed under reduced pressure to provide 987 mg of a residue containing (1S)-1-[1-(5-chloropyrimidin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethanamine hydrochloride (1:1). This was used without further purification.

20

¹H NMR (DMSO-d₆, 400 MHz): 9.10 (s, 2H), 8.65 (*br s*, 3H), 5.18 – 5.28 (*br m*, 1H), 2.08 – 2.15 (m, 1H), 1.59 (d, 3H, J = 6.8 Hz), 0.89 – 1.08 (m, 4H).

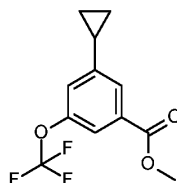
ESI mass [m/z]: 265.3 [amine+H]⁺

Following the above described procedure also (1S)-1-[1-(5-chloropyridin-2-yl)-3-cyclopropyl-1H-1,2,4-triazol-5-yl]ethanamine hydrochloride (1:1) (INT-20) was obtained.

Synthesis of 3-cyclopropyl-5-(trifluoromethoxy)benzoic acid

Step 1:

- 5 methyl 3-cyclopropyl-5-(trifluoromethoxy)benzoate

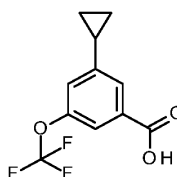


- A mixture of 1.89 g (6.30 mmol) methyl 3-bromo-5-(trifluoromethoxy)benzoate, 700 mg (8.15 mmol) cyclopropylboronic acid, 4.7 g (22 mmol) K_3PO_4 , 178 mg (0.64 mmol) tricyclohexylphosphine in 40 mL toluene and 2 mL H_2O was degassed by purging with argon. 72 mg (0.32 mmol) palladium(II) acetate were added. The mixture was stirred overnight at 100 °C. Water and ethyl acetate were added, the layers separated and the aqueous layer was extracted several times with ethyl acetate. The combined organic layers were dried with Na_2SO_4 , filtered and concentrated under reduced pressure to provide 2.00 g of a residue containing methyl 3-cyclopropyl-5-(trifluoromethoxy)benzoate which was used without further purification.

- 15 ESI mass [m/z]: 261.2 [M+H]⁺

Step 2:

3-cyclopropyl-5-(trifluoromethoxy)benzoic acid

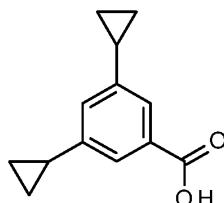


- A solution of the 2.00 g crude product obtained in the first step in 30 mL methanol was treated with 22 mL of a 1 M aqueous solution of sodium hydroxide. The mixture was stirred overnight at room temperature. The volatiles were then removed under reduced pressure. Water was added, the pH adjusted to pH 1 by the addition of 1 M hydrochloric acid and the mixture repeatedly extracted with ethyl acetate. The combined organic layers were dried with Na_2SO_4 and the solvent removed under reduced pressure to provide 1.55 g of 3-chloro-5-cyclopropylbenzoic acid.

¹H NMR (DMSO-d₆, 400 MHz): 13.4 (*br s*, 1H), 7.65 (t, 1H, *J* = 1.6 Hz), 7.56 (t, 1H, *J* = 1.2 Hz), 7.34 (s, 1H), 2.05 – 2.15 (m, 1H), 1.00 – 1.08 (m, 2H), 0.72 – 0.80 (m, 2H).

ESI mass [*m/z*]: 247.2 [*M*+H]⁺

Synthesis of 3,5-dicyclopropylbenzoic acid



5

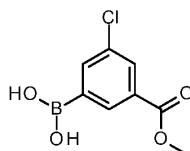
3,5-dicyclopropylbenzoic acid was synthesized analogously to the previous benzoic acid using methyl 3,5-dibromobenzoate and 2.4 equivalents of cyclopropylboronic acid as starting material.

¹H-NMR (400 MHz, DMSO-d₆): δ = 7.38 (d, *J* = 1.6 Hz, 2H), 7.03 (t, *J* = 1.6 Hz, 1H), 2.00 - 1.91 (m, 2H), 0.98 – 0.89 (m, 4H), 0.74 – 0.66 (m, 4H).

10 Synthesis of 3-chloro-5-[(trifluoromethyl)sulfanyl]benzoic acid

Step 1:

[3-chloro-5-(methoxycarbonyl)phenyl]boronic acid

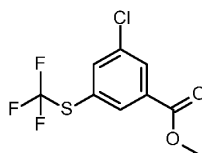


To a suspension of 12 g (40 mmol) methyl 3-chloro-5-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)benzoate in 30 mL acetone and 30 mL H₂O were added 17.3 g (80.9 mmol) sodium periodate and 6.24 g (80.9 mmol) ammonium acetate. The mixture was stirred at 25 °C for 2 h and then filtered through celite. The filtrate was evaporated. The residue was diluted with 200 mL ethyl acetate and washed with 100 mL H₂O. The organic phase was washed with brine, dried over anhydrous Na₂SO₄, filtered and concentrated in vacuum. The crude product was triturated with 10 mL petroleum ether at 15 °C for 20 min. The mixture was filtered and the residue dried under reduced pressure to obtain 7 g [3-Chloro-5-(methoxycarbonyl)phenyl]boronic acid as a white solid.

¹H-NMR (400 MHz, CDCl₃): δ = 8.72 (s, 1H), 8.35 (s, 1H), 8.27 (s, 1H), 4.02 (s, 3H). Referenced to the signal of trace CHCl₃ at 7.25 ppm. Measured using a Varian 400MR NMR machine.

Step 2:

methyl 3-chloro-5-[(trifluoromethyl)sulfanyl]benzoate



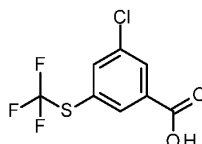
13 g (61 mmol) [3-Chloro-5-(methoxycarbonyl)phenyl]boronic acid, 43.1 g (303 mmol) trimethyl(trifluoromethyl)silane, 33.4 g (121 mmol) Ag_2CO_3 , 38.6 g (182 mmol) K_3PO_4 , 762 mg (6.06 mmol) CuSCN , 2.2 g (12 mmol) 1,10-Phenanthroline, 46.7 g (1.46 mol) sulfur and 13 g 4 Å molecular sieves in 500 mL DMF were stirred at 25 °C for 16 h under N_2 . The mixture was filtered through celite. The filtrate was diluted with 1.5 L methyl *tert*-butyl ether and washed with 2 x 500 mL H_2O . The organic phase was washed with brine, dried over anhydrous Na_2SO_4 , filtered and concentrated in vacuum. The crude product was purified by MPLC on silica gel (petroleum ether : ethyl acetate = 1 : 0 ~ 20 : 1) to obtain 5.5 g methyl 3-chloro-5-[(trifluoromethyl)sulfanyl]benzoate as a light yellow oil.

$^1\text{H-NMR}$ (400 MHz, CDCl_3): δ = 8.20 (s, 1H), 8.10 – 8.15 (m, 1H), 7.83 (s, 1H), 3.96 (s, 3H).

Referenced to the signal of trace CHCl_3 at 7.25 ppm. Measured using a Varian 400MR NMR machine.

Step 3:

15 3-chloro-5-[(trifluoromethyl)sulfanyl]benzoic acid



5.5 g (20 mmol) Methyl 3-chloro-5-[(trifluoromethyl)sulfanyl]benzoate were dissolved in a mixture of 12 mL tetrahydrofuran and 12 mL H_2O . 1.63 g (40.6 mmol) NaOH were added to the mixture which was then stirred at 25 °C for 2 h. The mixture was adjusted to pH 5 by addition of 40 mL 1 M HCl and extracted with 150 mL ethyl acetate. The organic phase was washed with brine, dried over anhydrous Na_2SO_4 , filtered and concentrated in vacuum. The crude product was triturated with 50 mL petroleum ether at 25 °C for 15 min. The mixture was filtered and the residue dried under reduced pressure to obtain 3.0 g 3-chloro-5-[(trifluoromethyl)sulfanyl]benzoic acid as a yellow solid.

$^1\text{H-NMR}$ (400 MHz, CDCl_3): δ = 11.28 (br s, 1H), 8.29 (s, 1H), 8.20 – 8.25 (m, 1H), 7.91 (s, 1H).

25 Referenced to the signal of trace CHCl_3 at 7.25 ppm. Measured using a Varian 400MR NMR machine.

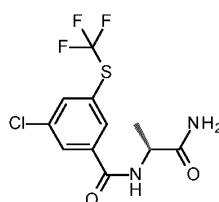
ESI mass $[\text{m/z}]$: 254.8 $[\text{M-H}]^-$

The determination by LC-MS was carried out using the mobile phases acetonitrile and 10 mM aqueous ammonium bicarbonate solution; linear gradient from 15% acetonitrile to 90% acetonitrile, flow rate 0.80 ml/min; instruments: Agilent 1200 & Agilent 6120. The column used for chromatography was a 2.1*50mm Xbridge Shield RPC18 column (5µm particles). Detection methods are diode array (DAD) and evaporative light scattering (ELSD) detection as well as negative electrospray ionization.

Synthesis of 3-chloro-N-[(1S)-1-[1-(5-cyanopyridin-2-yl)-3-methyl-1H-1,2,4-triazol-5-yl]ethyl]-5-[(trifluoromethyl)sulfonyl]benzamide (example II-15)

Step 1:

N-[(2S)-1-amino-1-oxopropan-2-yl]-3-chloro-5-[(trifluoromethyl)sulfonyl]benzamide



10

4.31 g (16.8 mmol) 3-chloro-5-[(trifluoromethyl)sulfonyl]benzoic acid were dissolved in dichloromethane and then two drops of DMF and 2.93 mL (34 mmol) oxalyl chloride were added. The mixture was stirred overnight room temperature. Dichloromethane and excess oxalyl chloride were removed under reduced pressure and the remaining residue was diluted with acetonitrile. This solution was added dropwise to a solution of 8.37 g (67.2 mmol) (2S)-2-aminopropanamide hydrochloride and 15.2 mL (109 mmol) triethylamine in acetonitrile. The mixture was stirred overnight at room temperature. Water was added upon which a precipitate formed. The mixture was filtered. The precipitate was dried to provide 3.59 g of the title compound as a white solid in 98% purity. The filtrate was extracted repeatedly with dichloromethane. The combined organic layers were washed with water, dried over anhydrous Na₂SO₄, filtered and concentrated *in vacuo*. This provided another batch of 4.14 g of the title compound in 70% purity.

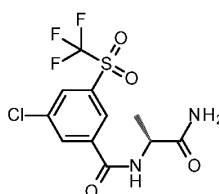
15

20

ESI mass [m/z]: 325.1 [M-H]⁺

Step 2:

N-[(2S)-1-amino-1-oxopropan-2-yl]-3-chloro-5-[(trifluoromethyl)sulfonyl]benzamide



25

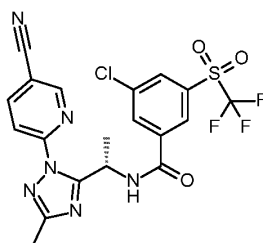
3.6 g (11 mmol) N-[(2S)-1-amino-1-oxopropan-2-yl]-3-chloro-5-[(trifluoromethyl)sulfonyl]benzamide

were suspended in a mixture of 57 mL dichloromethane, 56 mL acetonitrile and 113 mL of water. 7.07 g (33.1 mmol) sodium periodate and 2 mg (11 μ mol) ruthenium(III) chloride were added to the mixture which was then stirred overnight at room temperature. The mixture was diluted by the addition of water and extracted repeatedly with dichloromethane. The combined organic layers were washed with brine, dried over anhydrous Na₂SO₄, filtered and concentrated *in vacuo* to obtain 3.07 g of the title compound as a colorless solid. The crude product was used as such in the next step.

ESI mass [m/z]: 359.0 [M+H]⁺

Step 3:

- 10 3-chloro-N-{(1S)-1-[1-(5-cyanopyridin-2-yl)-3-methyl-1H-1,2,4-triazol-5-yl]ethyl}-5-[(trifluoromethyl)sulfonyl]benzamide (**example II-15**)



- 100 mg (279 μ mol) of N-[(2S)-1-amino-1-oxopropan-2-yl]-3-chloro-5-[(trifluoromethyl)sulfonyl]-benzamide were dissolved in 20 mL dichloromethane. 56 mg (0.42 mmol) *N,N*-dimethylformamide dimethyl acetal were added, and the reaction mixture was refluxed for 2 h. The solvents were then removed under reduced pressure, and the remaining residue was dissolved in a mixture of 5 mL dioxane and 0.5 mL acetic acid. 49 mg (0.36 mmol) 6-hydrazinonicotinonitrile were added. The reaction mixture was stirred for 2 h at 80 °C, concentrated *in vacuo* and the residue dissolved in CH₂Cl₂. This solution was washed with an aqueous sat. NaHCO₃ solution. The organic layer was then separated from the aqueous layer using a Chromabond™ PTS separation column and concentrated *in vacuo*. The remaining residue was purified by chromatography on silica (cyclohexane / ethyl acetate) to obtain 147 mg 3-chloro-N-{(1S)-1-[1-(5-cyanopyridin-2-yl)-3-methyl-1H-1,2,4-triazol-5-yl]ethyl}-5-[(trifluoromethyl)sulfonyl]-benzamide as a colorless solid.

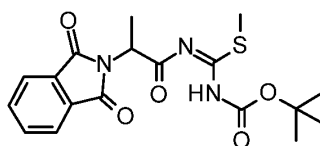
¹H-NMR (400 MHz, d₆-DMSO): see NMR peak list in table 3

- 25 ESI mass [m/z]: 499.1 [M+H]⁺

Synthesis of 3-chloro-N-{1-[3-chloro-1-pyrimidinyl-1H-1,2,4-triazol-5-yl]ethyl}-5-(methylsulfonyl)benzamide (example I-29)

Step 1

tert-butyl *N*-[(*E*)-*N*-[2-(1,3-dioxoisindolin-2-yl)propanoyl]-*C*-methylsulfanyl-carbonimidoyl]carbamate



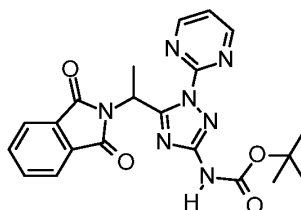
- 5 To 1.09 g (5.0 mmol) (αS)-1,3-dihydro- α -methyl-1,3-dioxo-2*H*-isoindole-2-acetic acid (Pht-Ala-OH purchased from ABCR) und 0.95 g (5.0 mmol) 1-*N*-Boc-2-methyl-isothiourea (purchased from ABCR) dissolved in tetrahydrofuran (30 ml), triethylamin (2.1 ml) and [O-(7-azabenzotriazol-1-yl)-*N,N,N',N'*-tetramethyluronium-hexafluoro-phosphate] (HATU) were added, and the reaction mixture was stirred 2 h at 80 °C temperature. Subsequently, water was added and the mixture was extracted with sodium
- 10 hydrogencarbonate solution and dichloromethane. The organic phase was separated, dried over Na₂SO₄, filtered and the solvent was evaporated. The remaining solid residue was purified by chromatography with a cyclohexane/acetone gradient on silica gel to afford 1.40 g (purity: 97.0 %; yield: 69.6 %) of the racemic title compound.

ESI mass [m/z]: 392.2 [M+H]⁺

- 15 ¹H-NMR (400 MHz, DMSO-d₆, ppm): NMR peaklist: δ = 11.8958 (0.5); 11.4353 (0.9); 7.9299 (0.5); 7.9221 (1.0); 7.9152 (1.2); 7.9102 (1.0); 7.9063 (1.2); 7.9000 (2.1); 7.8930 (1.1); 7.8847 (2.6); 7.8785 (1.3); 7.8744 (1.2); 7.8627 (0.8); 4.9976 (0.8); 4.9794 (0.8); 3.3230 (9.5); 2.5251 (0.4); 2.5204 (0.6); 2.5117 (8.2); 2.5072 (16.6); 2.5027 (21.9); 2.4981 (15.8); 2.4936 (7.6); 2.2949 (2.4); 1.9720 (6.0); 1.6029 (2.9); 1.5848 (3.0); 1.5719 (1.3); 1.5540 (1.1); 1.4430 (16.0); 1.3971 (11.0); 1.2665 (6.6); -0.0002 (0.5).

20 **Step 2**

2-[1-[3-(*N*-Boc-amino)-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl]-1*H*-isoindole-1,3(2*H*)-dione



- To a solution of 1.0 g (2.6 mmol) *tert*-butyl *N*-[(*E*)-*N*-[2-(1,3-dioxoisindolin-2-yl)propanoyl]-*C*-methylsulfanyl-carbonimidoyl]carbamate in pyridine (50 ml), were added at room temperature 338 mg
- 25 (3.06 mmol) 2-hydrazinopyrimidine. The reaction mixture was then stirred for 2 h at 80 °C. Afterwards

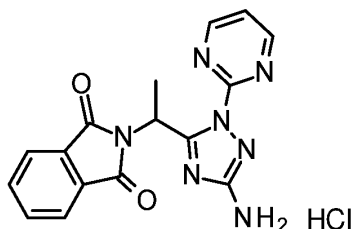
the solvent was evaporated under vacuo and the crude product was chromatographed with a cyclohexane/acetone gradient on silica gel to afford 780 mg (purity: 95.9 %; yield: 67.2 %) of the racemic title compound.

ESI mass [m/z]: 436 [M+H]⁺

- 5 ¹H-NMR (600 MHz, DMSO-d₆, ppm): NMR peaklist: δ = 9.9739 (1.2); 8.7635 (2.7); 8.7554 (2.7); 7.8169 (0.6); 7.8126 (0.8); 7.8097 (2.9); 7.8052 (3.0); 7.8020 (0.9); 7.7979 (0.6); 7.4395 (0.7); 7.4315 (1.4); 7.4234 (0.7); 6.0850 (0.8); 6.0732 (0.8); 5.7533 (0.3); 3.3088 (9.2); 2.5080 (3.8); 2.5050 (8.0); 2.5020 (11.1); 2.4989 (8.1); 2.4960 (3.8); 1.9448 (0.4); 1.9123 (0.4); 1.8008 (2.4); 1.7890 (2.4); 1.4365 (16.0); 1.3974 (6.1); -0.0001 (1.9).

10 **Step 3**

2-[1-(5-amino-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]isoindoline-1,3-dione -hydrochloride

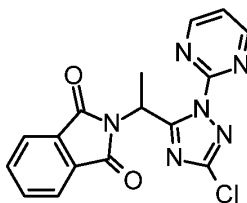


- 8.8 g (17.1 mmol) 2-[1-[3-(*N*-Boc-amino)-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl]-1*H*-isoindole-1,3(2*H*)-dione were treated with 4*N* solution of HCl in dioxane (150 ml) and the reaction mixture was stirred 18 h at room temperature. Then the reaction mixture was concentrated and the racemic solid residue was used for the halogen introduction (step 4) without further purification.

ESI mass [m/z]: 336.2 [M-Cl]⁺

Step 4

2-[1-[3-chloro-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl]-1*H*-isoindole-1,3(2*H*)-dione



20

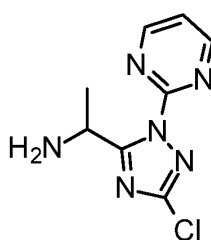
To 300 mg (0.89 mmol) 2-[1-(5-amino-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]isoindoline-1,3-dione -hydrochloride in acetonitrile (20 ml) were added 205 mg (1.52 mmol) Cu(II)-chloride, and then the

reaction mixture was treated dropwise at room temperature with 129 mg (1.25 mmol) *tert*-butyl nitrite. Then the reaction mixture was stirred 1 h at 70 °C temperature. The reaction mixture was treated with ethyl acetate and then extracted with a saturated NaHCO₃ solution and water. The organic phase was separated, dried and the solvent was evaporated to afford 178 mg (purity: 77 %; yield: 56.0 %) of the racemic title compound, which was used for the deprotection reaction (step 5) without further purification.

ESI mass [m/z]: 355.3 [M+H]⁺

Step 5

1-(5-chloro-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (**INT-10**)

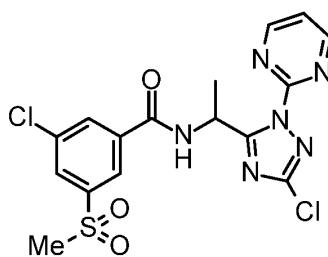


To 499 mg (2.5 mmol) 2-[1-[3-chloro-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl-1*H*-isoindole-1,3(2*H*)-dione in ethanol (20 mL) were added 320 mg (3.51 mmol) hydrazine-hydrate, and the reaction mixture was heated under reflux. After 30 minutes a colorless precipitate was formed. The reaction mixture was stirred and heated under reflux two additional hours then acetone (2 mL) was added and the heating was continued for further 30 minutes. The reaction mixture was concentrated and the solid residue was treated with ethanol. After filtration, the filtrate was evaporated under reduced pressure to afford 310 mg (purity: 70-80 %, yield: 98%) of the racemic 1-(5-chloro-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (**INT-10**), which was used in step 6 without further purification.

ESI mass [m/z]: 225.1 [M+H]⁺

Step 6

3-chloro-N-[1-(5-chloro-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-5-methylsulfonyl-benzamide (**example I-29**)



To 150 mg (0.66 mmol) 1-(5-chloro-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (INT-10), 160 mg (0.66 mmol) 3-chloro-5-(sulfonylmethyl)-benzoic acid, 120 mg (0.92 mmol) *N,N*-diisopropylethylamine (Hünig's Base) in *N,N*-dimethylformamide (DMF) (3 mL) were added 310 mg (0.81 mmol) [*O*-(7-azabenzotriazol-1-yl)-*N,N,N',N'*-tetramethyluronium-hexafluoro-phosphate] (HATU), and the reaction mixture was stirred at room temperature overnight. The reaction mixture was concentrated under reduced pressure and the solid residue was treated with dichloromethane and then extracted with a saturated aqueous NaHCO₃ solution and water. The organic phase was separated, dried over Na₂SO₄ and the solvent was evaporated under reduced pressure. The remaining solid residue was chromatographed with a cyclohexane/acetone gradient on silica gel to obtain 99 mg (purity: 100 %; yield: 34 %) of the racemic title compound.

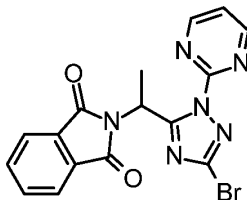
ESI mass [m/z]: 441.2 [M+H]⁺

¹H-NMR: see NMR peak list in table 1

Synthesis of 3-chloro-*N*-{1-[3-bromo-1-pyrimidinyl-1*H*-1,2,4-triazol-5-yl]ethyl}-5-(methylsulfonyl)benzamide (example I-35)

Step 4

2-[1-[3-bromo-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl]-1*H*-isoindole-1,3(2*H*)-dione

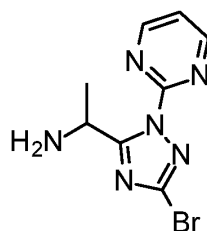


To 2.0 g (5.9 mmol) 2-[1-(5-amino-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]isoindoline-1,3-dione - hydrochloride in acetonitrile (133 ml) were added 2.7 g (12 mmol) Cu(II)-bromide, and then the reaction mixture was treated dropwise at room temperature with 1.0 g (9.7 mmol) *tert*-butyl nitrite. Then the reaction mixture was stirred for 1 h at 70 °C temperature. The reaction mixture was treated with ethyl acetate and then extracted with a saturated NaHCO₃ solution and water. The organic phase was separated, dried and the solvent was evaporated to afford 1.10 g (purity: 97%; yield: 45%) of the racemic title compound, which was used for the coupling reaction (step 5) without further purification.

ESI mass [m/z]: 399.2 [M+H]⁺

Step 5

1-(5-bromo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (INT-11)

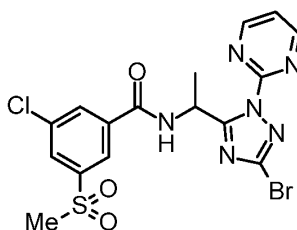


To 1.16 g (2.91 mmol) 2-[1-[3-bromo-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl-1*H*-isoindole-1,3(2*H*)-dione in ethanol (30 mL) were added 678.8 mg (7.45 mmol) hydrazine-hydrate and the reaction mixture was heated under reflux. After 30 minutes a colorless precipitate was formed. The reaction mixture was stirred and heated under reflux for two additional hours, the acetone (20 mL) was added and the heating was continued for further 30 minutes. The reaction mixture was concentrated and the solid residue was treated with ethanol. After filtration, the filtrate was evaporated under reduced pressure to afford 0.9 g (purity: 50-60 %) of the racemic 1-(5-bromo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (INT-11), which was used in step 6 without further purification.

ESI mass [m/z]: 271.1 [M+H]⁺

Step 6

N-[1-(5-bromo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-chloro-5-methylsulfonyl-benzamide (example I-35)



To 180 mg (0.66 mmol) 1-(5-bromo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (INT-011) were added 160 mg (0.66 mmol) 3-chloro-5-(sulfonylmethyl)-benzoic acid, 124 mg (0.95 mmol) *N,N*-diisopropylethylamine (Hünig's Base) in acetonitrile (5 mL) and 320 mg (0.84 mmol) [*O*-(7-azabenzotriazol-1-yl)-*N,N,N',N'*-tetramethyluronium-hexafluoro-phosphate] (HATU). Then the reaction mixture was stirred at room temperature overnight. The reaction mixture was concentrated under reduced pressure and the solid residue was treated with dichloromethane and then extracted with a saturated aqueous NaHCO₃ solution and water. The organic phase was separated, dried over Na₂SO₄ and the solvent was evaporated under reduced pressure. The remaining solid residue was purified by HPLC with a water/acetonitril gradient (neutral) to obtain 111 mg (purity: 98 %; yield: 32 %) of the racemic title compound.

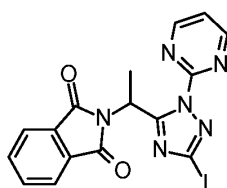
ESI mass [m/z]: 487.0 [M+H]⁺

¹H-NMR: see NMR peak list in table 1

Synthesis of 3-chloro-N-{1-[3-iodo-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl}ethyl}-5-(methylsulfonyl)benzamide (example I-43)

5 **Step 1**

2-[1-[3-Iodo-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl-1*H*-isoindole-1,3(2*H*)-dione

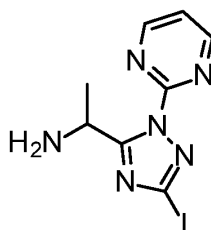


To 11.2 g (30.2 mmol) 2-[1-[3-(amino)-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl-1*H*-isoindole-1,3(2*H*)-dione-hydrochloride in acetonitrile (448 ml) (synthesized according to steps 1-3, example I-29), were added 181 g (676 mmol) diiodomethane (argon atmosphere), and then the reaction mixture was treated dropwise at room temperature with 14.3 g (139 mmol) *tert*-butyl nitrite. Then the reaction mixture was stirred for 3 h at 80 °C temperature. The reaction mixture was treated with ethyl acetate and then extracted with a saturated NaCl solution. Afterwards the solvent was evaporated under vacuo and the crude product was chromatographed with a cyclohexane/acetone gradient on silica gel to afford 8.1 g (yield: 60 %) of the racemic title compound.

ESI mass [m/z]: 447.0 [M+H]⁺

Step 2

1-(5-iodo-2-pyrimidin-2-yl)-1,2,4-triazol-3-yl)ethanamine (INT-12)



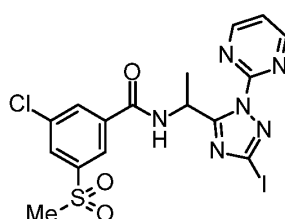
To 3.07 g (6.89 mmol) 2-[1-[3-iodo-1-pyrimidinyl]-1*H*-1,2,4-triazol-5-yl]ethyl-1*H*-isoindole-1,3(2*H*)-dione in ethanol (100 mL), 1.56 g (17.2 mmol) hydrazine-hydrate were added, and the reaction mixture was heated under reflux. After 30 minutes a colorless precipitate was formed. The reaction mixture was

stirred and heated under reflux for two additional hours, acetone (2 mL) was added and the heating was continued for further 30 minutes. The reaction mixture was concentrated and the solid residue was treated with ethanol. After filtration, the filtrate was evaporated under reduced pressure to afford the racemic 1-(5-iodo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (INT-12), which was used in step 3 without further purification.

ESI mass [m/z]: 317.0 [M+H]⁺

Step 3

3-Chloro-*N*-[1-(5-iodo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-5-methylsulfonyl-benzamide

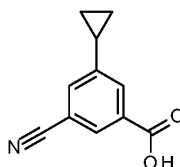


To 100 mg (0.41 mmol) 3-chloro-5-(sulfonylmethyl)-benzoic acid, were added 75 mg (0.58 mmol) *N,N*-diisopropylethylamine (Hünig's Base) in 3.6 g (50 mmol) *N,N*-dimethyl-formamid (DMF) and 194 mg (0.51 mmol) [*O*-(7-azabenzotriazol-1-yl)-*N,N,N',N'*-tetramethyluronium-hexafluoro-phosphate] (HATU). The mixture was stirred at room temperature for 1 h. Then 133 mg (0.37 mmol) 1-(5-iodo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethanamine (INT-12) were added and the reaction mixture was stirred at room temperature overnight. The reaction mixture was concentrated under reduced pressure and the solid residue was treated with dichloromethane and water. The organic phase was separated, dried over Mg₂SO₄ and the solvent was evaporated under reduced pressure. The remaining solid residue was purified by HPLC with a water/acetonitrile gradient (neutral) to obtain 18.6 mg (purity: 100 %; yield: 8 %) of the title compound.

ESI mass [m/z]: 532.9 [M+H]⁺

¹H-NMR: see NMR peak list in table 1

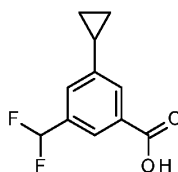
Synthesis of 3-cyano-5-cyclopropylbenzoic acid (INT-15)



A mixture of 500 mg (2.21 mmol) 3-bromo-5-cyanobenzoic acid, 285 mg (3.31 mmol) cyclopropylboronic acid, 1.64 g (7.72 mmol) K_3PO_4 , 63 mg (0.22 mmol) tricyclohexylphosphine in 10 mL toluene and 0.5 mL H_2O was degassed by purging with argon. 25 mg (0.11 mmol) palladium(II) acetate were added. The mixture was stirred overnight at 100 °C. Water and ethyl acetate were added, the layers separated and the aqueous layer was extracted several times with ethyl acetate. The aqueous layer was acidified with hydrochloric acid upon which a precipitate formed. The precipitate was filtered off and dried under reduced pressure to provide 215 mg of 3-cyano-5-cyclopropylbenzoic acid which was used without further purification.

ESI mass [m/z]: 186.1 [M-H]⁻

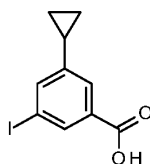
10 Synthesis of 3-cyclopropyl-5-(difluoromethyl)benzoic acid (INT-13)



3-Cyclopropyl-5-(difluoromethyl)benzoic acid was synthesized analogously to the previous benzoic acid using 3-bromo-5-(difluoromethyl)benzoic acid as starting material.

¹H-NMR (400 MHz, DMSO-d₆): δ = 13.12 - 13.40 (broad s, 1 H), 7.87 (s, 1 H), 7.78 (s, 1 H), 7.51 (s, 1 H), 7.20 - 7.06 (t, 1 H), 2.14 - 2.08 (m, 1 H), 1.07 - 1.00 (m, 2H), 0.76 - 0.74 (m, 2H).

Synthesis of 3-cyclopropyl-5-iodobenzoic acid (INT-14)

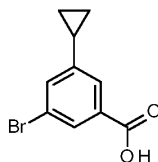


A mixture of 500 mg (1.33 mmol) 3,5-diiodobenzoic acid, 118 mg (1.33 mmol) cyclopropylboronic acid, 0.99 g (4.66 mmol) K_3PO_4 , 38 mg (0.13 mmol) tricyclohexylphosphine in 10 mL toluene and 0.5 mL H_2O was degassed by purging with argon. 15 mg (68 μ mol) palladium(II) acetate were added. The mixture was stirred overnight at 100 °C. Water and ethyl acetate were added, the layers separated and the aqueous layer was extracted several times with ethyl acetate. The aqueous layer was acidified with hydrochloric acid upon which a precipitate formed. The precipitate was filtered off and dried under reduced pressure. The remaining solid residue was purified by HPLC with a water/acetonitrile/HCOOH gradient to obtain 22 mg (purity: 81 %; yield: 5 %) of the title compound.

¹H-NMR (400 MHz, DMSO-d₆): δ = 13.3 (broad s, 1 H), 7.98 (s, 1 H), 7.68 (s, 1 H), 7.60 (s, 1 H), 2.08 - 1.96 (m, 1 H), 1.01 - 0.95 (m, 2H), 0.75 - 0.69 (m, 2H).

ESI mass [m/z]: 287.1 [M-H]⁻

Synthesis of 3-bromo-5-cyclopropylbenzoic acid



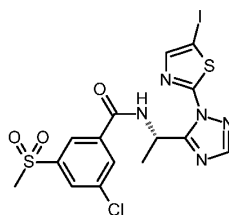
5

3-bromo-5-cyclopropylbenzoic acid was synthesized analogously to 3-cyclopropyl-5-iodobenzoic acid using 3-bromo-5-iodobenzoic acid as starting material.

ESI mass [m/z]: 241.1 [M-H]⁻

Synthesis 3-chloro-N-{(1S)-1-[1-(5-iodo-1,3-thiazol-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}-5-(methylsulfonyl)benzamide (example II-51)

10



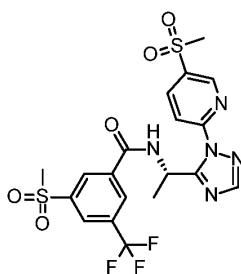
A solution of 0.83 g (2.0 mmol) of 3-chloro-5-(methylsulfonyl)-N-{(1S)-1-[1-(1,3-thiazol-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}benzamide in 60 ml DMF was stirred at 100 °C and treated with portions of N-iodo-succinimide until full conversion of the starting material was observed. A total of 4.5 g (20 mmol) N-iodo-succinimide was used and added within 96 h. Then an aq. solution of sodium bisulfite was added and the mixture repeatedly extracted with EtOAc. The combined organic layers were washed with water, aq. saturated NaHCO₃ solution and brine. All volatiles were removed under reduced pressure to obtain 3-chloro-N-{(1S)-1-[1-(5-iodo-1,3-thiazol-2-yl)-1H-1,2,4-triazol-5-yl]ethyl}-5-(methylsulfonyl)benzamide.

15

20 ESI mass [m/z]: 538.0 [M+H]⁺

¹H NMR peaklist see table 3

Synthesis of 3-(methylsulfonyl)-N-[(1S)-1-{1-[5-(methylsulfonyl)pyridin-2-yl]-1H-1,2,4-triazol-5-yl}ethyl]-5-(trifluoromethyl)benzamide (example II-56)



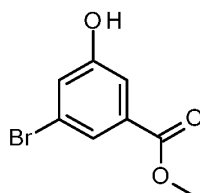
1.84 g (3 mmol) 3-iodo-N-((1S)-1-[1-(5-iodopyridin-2-yl)-1H-1,2,4-triazol-5-yl]ethyl)-5-(trifluoromethyl)benzamide (prepared according to the general description), 1.43 g (7.5 mmol) copper(I)iodide, 0.40 g (1.37 mmol) (E,E)-N,N'-cyclohexane-1,2-diylbis[1-(pyridin-2-yl)methanimine] and 3.6 g (30 mmol) sodium methanesulfinate were stirred in DMSO under Argon at 65 °C for 16 h. The reaction mixture was concentrated under reduced pressure, the residue was treated with ethyl acetate and water, and suction-filtered over diatomaceous earth. As the filtrate contained only traces of product, the diatomaceous earth was stirred with dichloromethane-methanol and suction-filtered. The filtrate was evaporated under reduced pressure and the residue was purified by reversed phase chromatography (RP18, acetonitrile-water, 0,1% HCOOH) to yield 25 mg (1.6%) of the title compound.

ESI mass [m/z]: 518.1 [M+H]⁺

Synthesis of 3-cyclopropyl-5-(difluoromethoxy)benzoic acid

Step 1

methyl 3-bromo-5-hydroxybenzoate



15

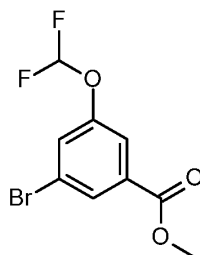
A solution of 3-bromo-5-hydroxybenzoic acid (49.9 g, 230 mmol) in MeOH (325 mL) was cooled by an ice bath to 7-8 °C. Then SOCl₂ (27.4 g, 16.79 mL, 230 mmol) was added dropwise to this solution over 25 min. The reaction mixture was warmed to room temperature, stirred under reflux for 3 h, cooled down to room temperature and then stirred for another 48 h at this temperature. All volatiles were removed in vacuo and the residue dissolved in ethyl acetate (400 mL). The solution was washed with NaHCO₃, brine, dried over Na₂SO₄ and the volatiles were removed under reduced pressure. The residue was triturated with hexanes (400 mL). The precipitate was filtered off, washed with hexanes/diethyl ether (1:1), dried at 110 °C to afford methyl 3-bromo-5-hydroxybenzoate (50.5 g) as a dark yellow powder.

20

^1H NMR (400 MHz, CDCl_3) δ = 7.73 (m, 1H), 7.51 (m, 1H), 7.26 (s, 0H), 7.23 (t, J = 2.1 Hz, 1H), 6.05 (br s, 1H), 3.92 (s, 3H). (recorded on a Varian Gemini 2000 machine)

Step 2

methyl 3-bromo-5-(difluoromethoxy)benzoate



5

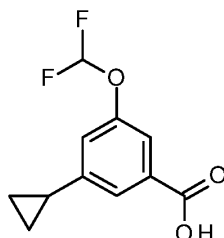
A mixture of methyl 3-bromo-5-hydroxybenzoate (23.1 g, 100 mmol), K_2CO_3 (41.5 g, 300 mmol) and $\text{ClF}_2\text{CCOONa}$ (45.7 g, 300 mmol) in DMF (350 mL) was stirred at 60-65°C for 2 h. The precipitate was then separated, washed with acetone and the filtrate was evaporated under reduced pressure. The residue was dissolved in diethyl ether (300 mL) and the solution was left to stand at rt for 12 h. A precipitate
10 formed which was filtered off and washed with water. The filtrate was washed with brine (300 mL) and the organic layer was evaporated under reduced pressure. The oily residue was dissolved in hexanes (250 mL) and kept at rt for 2 h. A precipitate formed which was removed by filtration and the filtrate was evaporated under reduced pressure. The residue was distilled under reduced pressure (3 tor) and the fraction with a boiling point between 80 and 85 °C was collected to afford 15.75 g methyl 3-bromo-5-
15 (difluoromethoxy)benzoate.

^1H NMR (400 MHz, CDCl_3) δ = 8.03 (t, J = 1.6 Hz, 1H), 7.75 – 7.70 (m, 1H), 7.49 (t, J = 2.1 Hz, 1H), 6.55 (t, J = 72.6 Hz, 1H), 3.93 (s, 3H). (recorded on a Varian Gemini 2000 machine)

^{19}F NMR (376 MHz, CDCl_3) δ = -84.89 (d, J = 72.7 Hz). (recorded on a Varian Gemini 2000 machine)

Step 3

20 3-cyclopropyl-5-(difluoromethoxy)benzoic acid

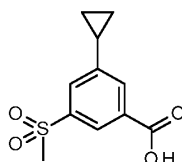


A mixture of methyl 3-bromo-5-(difluoromethoxy)benzoate (12.07 g, 52 mmol), K_3PO_4 (27.6 g, 130 mmol), Ph_3P (1.364 g, 5.2 mmol) and cyclopropylboronic acid (8.93 g, 104 mmol) in diglyme (250 mL) was heated to 100 °C and then $(Ph_3P)_2PdCl_2$ (1.825 g, 2.6 mmol) was added in one portion. The reaction mixture was stirred at 100 °C for 3 h, cooled down to rt and treated with a solution of NaOH (5 g) in water (250 mL). The reaction mixture was stirred for 48 h. Then the precipitate was filtered off and water 950 mL was added. The mixture was extracted with ethyl acetate (2x500 mL). The aqueous layer was acidified by concentrated hydrochloric acid to pH = 2 and kept at rt for 12 h. The precipitate was filtered off, washed with boiling water (4 x 200mL), dried at 110 °C to get 10.4 g of crude product. This material was dissolved in hot toluene (30 mL) and the solution was diluted by hexanes to 150 mL. The mixture was filtrated hot, the filtrate cooled down to room temperature and than left in the refrigerator for 3 h. A precipitate formed which was filtered off, washed with hexanes and dried at 110 °C to get 7.7 g of crude material. The mother liquor was evaporated and the residue triturated with hexanes:diethyl ether (1:1). The insoluble material was filtered off and combined with the 7.7 g crude material. The combined crude material was purified by sublimation (0.3 torr, 110 °C) to afford 9.7 g 3-cyclopropyl-5-(difluoromethoxy)benzoic acid.

1H NMR (400 MHz, $CDCl_3$) δ = 12.10 (s, 1H), 7.67 (d, J = 1.6 Hz, 1H), 7.62 (t, J = 1.9 Hz, 1H), 7.08 (t, J = 2.0 Hz, 1H), 6.55 (t, J = 73.5 Hz, 1H), 2.06 – 1.89 (m, 1H), 1.15 – 0.96 (m, 2H), 0.85 – 0.69 (m, 2H). (recorded on a Varian Gemini 2000 machine)

^{19}F NMR (376 MHz, $CDCl_3$) δ = -84.24 (d, J = 73.5 Hz). (recorded on a Varian Gemini 2000 machine)

Synthesis of 3-cyclopropyl-5-(methylsulfonyl)benzoic acid (INT-17)



20

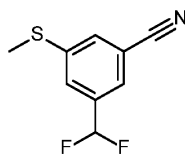
3-cyclopropyl-5-(methylsulfonyl)benzoic acid was synthesized analogously to 3-chloro-5-(ethylsulfonyl)benzoic acid using 3-bromo-5-cyclopropylbenzoic acid as starting material. For the synthesis of 3-cyclopropyl-5-(methylsulfonyl)benzoic acid the reaction mixture was stirred overnight at 120 °C.

25 ESI mass [m/z]: 239.0 [M-H]⁻

1H -NMR (400 MHz, DMSO- d_6): δ = 13.5 (broad s, 1 H), 8.16 (t, J = 1.6 Hz, 1H), 7.92 (t, J = 1.6 Hz, 1H), 7.85 (t, J = 1.6 Hz, 1H), 3.27 (s, 3H), 2.23 -2.15 (m, 1 H), 1.12 – 1.02 (m, 2H), 0.90 – 0.80 (m, 2H).

Synthesis of 3-(difluoromethyl)-5-(methylsulfonyl)benzoic acid

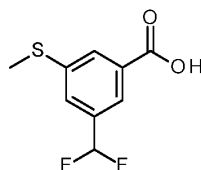
Step 1: 3-(difluoromethyl)-5-(methylsulfonyl)benzonitrile



To a mixture of 0.24 g (5.5 mmol) sodium hydride and 13.6 mL DMF were added at 20 °C 2.30 g (11.0 mmol) 3-chloro-5-(difluoromethyl)benzonitrile. The mixture was stirred for 15 min at 20 °C after which 1.01 g (14.3 mmol) sodium methanethiolate were added. The reaction mixture was then stirred for 3 h at 50 °C. The reaction was quenched by the careful addition of water and the reaction mixture acidified to pH 6 by the addition of acetic acid. All volatiles were then removed under reduced pressure. Water was added to the residue and the mixture repeatedly extracted with ethyl acetate. The combined organic layers were washed with brine and dried with Na₂SO₄. The solvent was removed under reduced pressure to provide a residue which was purified by reversed-phase chromatography (H₂O / acetonitrile) to yield the title compound (497 mg) and 3-(difluoromethyl)-5-(methylsulfanyl)benzamide (287 mg).

ESI mass [m/z]: 200.1 [M+H]⁺

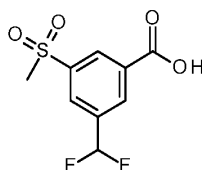
Step 2: 3-(difluoromethyl)-5-(methylsulfanyl)benzoic acid



497 mg (2.49 mmol) 3-(difluoromethyl)-5-(methylsulfanyl)benzonitrile were dissolved in 5.1 mL methanol and 10.1 mL THF. To this solution were added 1.98 mL of a 50% aqueous solution of sodium hydroxide and the reaction mixture was heated at reflux for 45 min. At this point 287 mg (1.32 mmol) 3-(difluoromethyl)-5-(methylsulfanyl)benzamide (obtained in the previous step) and further 1.98 mL of a 50% aqueous solution of sodium hydroxide solution were added. The mixture was heated at reflux for 1 h and stirred overnight at room temperature. All volatiles were removed under reduced pressure. Water was added. Then the mixture was acidified to pH 1-2 using conc. hydrochloric acid after which it was repeatedly extracted with ethyl acetate. The combined organic layers were washed with brine and dried with Na₂SO₄. The solvent was removed under reduced pressure to provide 811 mg of a residue containing 3-(difluoromethyl)-5-(methylsulfanyl)benzoic acid.

ESI mass [m/z]: 219.1 [M+H]⁺

Step 3: 3-(difluoromethyl)-5-(methylsulfonyl)benzoic acid



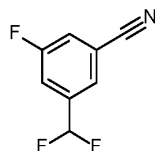
The crude material from the previous step containing 3-(difluoromethyl)-5-(methylsulfanyl)benzoic acid was dissolved in 40 mL CH₂Cl₂. Subsequently 0.7 mL formic acid and 2.7 mL of an aqueous 30 % hydrogen peroxide solution were added. The suspension was stirred overnight at room temperature after which further 0.7 mL formic acid and 2.7 mL 30 % hydrogen peroxide solution were added. The suspension was stirred for 1 h at room temperature and for 1 h at 30 °C. As the conversion of the starting material was still incomplete further 0.7 mL formic acid and 2.7 mL 30 % hydrogen peroxide solution were added and the reaction mixture was stirred for 3 d at room temperature. At this point a 513 mg (2.97 mmol) meta-chloroperoxybenzoic acid were added and the mixture stirred further overnight at room temperature. It was then quenched by the addition of 40% aqueous NaHSO₃ solution followed by stirring for 1 h. Water was added and the mixture repeatedly extracted with CH₂Cl₂. The combined organic layers were washed with brine and dried with Na₂SO₄. The solvent was removed under reduced pressure to provide a residue which was purified by reversed-phase chromatography (H₂O / acetonitrile) to yield 3-(difluoromethyl)-5-(methylsulfonyl)benzoic acid (244 mg) and some recovered starting material (79 mg).

ESI mass [m/z]: 251.0 [M+H]⁺

¹H NMR (DMSO-d₆, 400 MHz): δ = 14.0 (br s, 1H), 8.54 (s, 1H), 8.41 (s, 1H), 8.33 (s, 1H), 7.27 (t, J = 55 Hz, 1H). (Signal of CH₃-group is hidden under solvent signal)

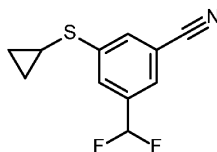
Synthesis of 3-(cyclopropylsulfanyl)-5-(difluoromethyl)benzonitrile

Step 1: 3-(difluoromethyl)-5-fluorobenzonitrile



In a plastic reaction vessel 10 g (67 mmol) 3-fluoro-5-formylbenzonitrile were dissolved in 300 mL CH₂Cl₂. At room temperature 10.6 mL (80.4 mmol) diethylaminosulfur trifluoride were added and the mixture was stirred overnight. To the reaction mixture was then carefully added a sat. aqueous solution of NaHCO₃. The mixture was stirred at room temperature until any remaining reagents had decomposed. The mixture was then extracted with CH₂Cl₂. The combined organic layers were washed with brine, dried with Na₂SO₄, filtered and concentrated under reduced pressure to provide 10.67 g 3-(difluoromethyl)-5-fluorobenzonitrile which was used for the next step without further purification.

¹H NMR (DMSO-d₆, 400 MHz): δ = 8.10 (d, J = 8 Hz, 1H), 7.99 (s, 1H), 7.89 (d, J = 8 Hz, 1H), 7.11 (t, J = 55 Hz, 1H).

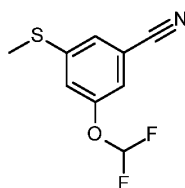
Step 2: 3-(cyclopropylsulfanyl)-5-(difluoromethyl)benzonitrile

To a solution of 3.50 g (20.4 mmol) 3-(difluoromethyl)-5-fluorobenzonitrile in 40 mL DMF were added at 0 °C 2.95 g (30.6 mmol) sodium cyclopropanethiolate. The mixture was stirred for 1 h at 0 °C and overnight at room temperature. The reaction mixture was then concentrated under reduced pressure and the residue purified by reversed-phase chromatography (H₂O / acetonitrile) to yield 3-(cyclopropylsulfanyl)-5-(difluoromethyl)benzonitrile (1.18 g) and some remaining 3-(difluoromethyl)-5-fluorobenzonitrile benzamide (582 mg).

¹H NMR (DMSO-d₆, 400 MHz): δ = 7.99 (s, 1H), 7.84 (s, 2H), 7.08 (t, *J* = 55 Hz, 1H), 2.45 - 2.38 (m, 1H), 1.22 - 1.15 (m, 2H), 0.67 - 0.60 (m, 2H).

ESI mass [m/z]: 226.0 [M+H]⁺

3-(cyclopropylsulfanyl)-5-(difluoromethyl)benzonitrile was then further converted to 3-(cyclopropylsulfonyl)-5-(difluoromethyl)benzoic acid via hydrolysis of the nitrile and oxidation of the thioether to the sulfone. These transformations were conducted in analogy to the conditions described above for the synthesis of 3-(difluoromethyl)-5-(methylsulfonyl)benzoic acid.

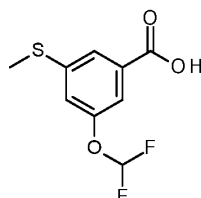
Synthesis of 3-(difluoromethoxy)-5-(methylsulfonyl)benzoic acid**Step 1: 3-(difluoromethoxy)-5-(methylsulfonyl)benzonitrile**

To a solution of 0.90 g (4.8 mmol) 3-(difluoromethoxy)-5-fluorobenzonitrile (obtained from FCH Group) in 10 mL DMF at 0 °C were added 0.34 g (4.8 mmol) sodium methanethiolate. The mixture was stirred for 2 h at 0 °C after which it was allowed to warm to room temperature. The reaction mixture was stirred for 50 h at room temperature and then recooled to 0 °C. Further 50 mg (0.7 mmol) sodium methanethiolate were added and the reaction mixture stirred for 1 h at 0 °C. As the conversion was still incomplete further 15 mg (0.2 mmol) sodium methanethiolate were added and the reaction mixture was stirred for 30 min at 0 °C. Water was then added and the reaction mixture acidified to pH 5 by the addition of acetic acid. All volatiles were removed under reduced pressure. Water was added to the residue and the mixture repeatedly

extracted with ethyl acetate. The combined organic layers were washed with brine and dried with Na_2SO_4 . The solvent was removed under reduced pressure to provide 1.41 g of a residue containing 3-(difluoromethoxy)-5-(methylsulfanyl)benzonitrile and residual DMF.

ESI mass $[\text{m/z}]$: 216.0 $[\text{M}+\text{H}]^+$

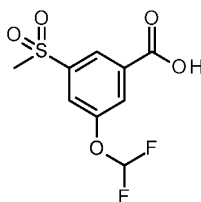
5 **Step 2: 3-(difluoromethoxy)-5-(methylsulfanyl)benzoic acid**



1.10 g of the residue obtained in step one containing 3-(difluoromethoxy)-5-(methylsulfanyl)benzonitrile were dissolved in 8.3 mL methanol and 16.6 mL THF. To this solution were added 3.25 mL of a 50% aqueous solution of sodium hydroxide and the reaction mixture was heated at reflux for 2 h. Water was added at room temperature. The mixture was then acidified to pH 1-2 using conc. hydrochloric acid and repeatedly extracted with ethyl acetate. The combined organic layers were washed with brine and dried with Na_2SO_4 . The solvent was removed under reduced pressure to provide a residue which was purified by reversed-phase chromatography (H_2O / acetonitrile) to yield 592 mg 3-(difluoromethoxy)-5-(methylsulfanyl)benzoic acid.

15 ESI mass $[\text{m/z}]$: 235.0 $[\text{M}+\text{H}]^+$

Step 3: 3-(difluoromethoxy)-5-(methylsulfonyl)benzoic acid

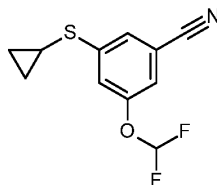


To a solution of 85 mg (0.36 mmol) 3-(difluoromethoxy)-5-(methylsulfanyl)benzoic acid dissolved in 4 mL CH_2Cl_2 were added 0.07 mL formic acid and 288 mg of an aqueous 30 % hydrogen peroxide solution. The reaction mixture was stirred overnight at room temperature. It was then quenched by the addition of 40% aqueous NaHSO_3 solution followed by stirring for 1 h. Water was added and the mixture extracted once with CH_2Cl_2 and repeatedly with ethyl acetate. The solvent was removed from the combined organic layers under reduced pressure to provide a residue which was purified twice by reversed-phase chromatography (H_2O / acetonitrile) to yield 43 mg 3-(difluoromethoxy)-5-(methylsulfonyl)benzoic acid.

¹H NMR (DMSO-d₆, 400 MHz): δ = 13.9 (br s, 1H), 8.26 (s, 1H), 7.97 (s, 2H), 7.49 (t, *J* = 73 Hz, 1H).
(Signal of CH₃-group is hidden under solvent signal)

ESI mass [m/z]: 267.0 [M+H]⁺

Synthesis of 3-(cyclopropylsulfanyl)-5-(difluoromethoxy)benzonitrile



5

To a solution of 0.90 g (4.8 mmol) 3-(difluoromethoxy)-5-fluorobenzonitrile (obtained from FCH Group) in 10 mL DMF at 0 °C were added 694 mg (7.21 mmol) sodium cyclopropanethiolate. The mixture was stirred for 1 h at 0 °C after which it was allowed to warm to room temperature. The reaction mixture was stirred overnight at room temperature. It was then directly purified by reversed-phase chromatography (H₂O / acetonitrile) to yield 283 mg of the title compound.

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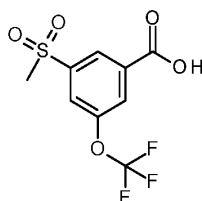
ESI mass [m/z]: 242.0 [M+H]⁺

¹H NMR (DMSO-d₆, 400 MHz): δ = 7.69 (s, 1H), 7.50 (s, 1H), 7.47 (s, 1H), 7.36 (t, *J* = 73 Hz, 1H), 2.43 - 2.36 (m, 1H), 1.22 - 1.15 (m, 2H), 0.61 - 0.66 (m, 2H).

3-(Cyclopropylsulfanyl)-5-(difluoromethoxy)benzonitrile was then further converted to 3-(cyclopropylsulfonyl)-5-(difluoromethoxy)benzoic acid via hydrolysis of the nitrile and oxidation of the thioether to the sulfone. These transformations were conducted in analogy to the conditions described above for the synthesis of 3-(difluoromethoxy)-5-(methylsulfonyl)benzoic acid.

15

Synthesis of 3-methylsulfonyl-5-(trifluoromethoxy)benzoic acid



A mixture of 2.95 g (17.5 mmol) trans-*N,N*-dimethylcyclohexane-1-2-diamine and 11.4 g (35 mmol) cesium carbonate in 60 mL DMF was degassed for 30 min by purging with argon. 5 g (17.5 mmol) 3-bromo-5-(trifluoromethoxy)benzoic acid, 3.58 g (35 mmol) sodium methanesulfinate and 3.34 g (17.5 mmol) copper(I) iodide were added and the mixture further purged with argon for 5 min. The mixture was stirred at 120 °C over night, cooled to room temperature and then three times extracted with

20

dichloromethane. The aqueous layer was acidified to pH 2 using concentrated hydrochloric acid and again extracted with dichloromethane. The dichloromethane phase was washed with brine several times. The layers were separated, and the combined organic layers were dried over anhydrous Na₂SO₄ and filtered. The solvent was removed under reduced pressure and the residue triturated with n-pentane, filtered-off and dried to provide 3.2 g of 3-methylsulfonyl-5-(trifluoromethoxy)benzoic acid.

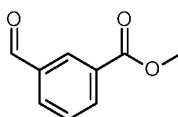
¹H NMR (DMSO-d₆, 400 MHz): δ= 14.00 (br s, 1H, COOH), 8.42 (s, 1H), 8.20 (s, 1H), 8.14 (s, 1H), 3.39 (s, 3H).

ESI mass [m/z]: 285.0 [M+H]⁺

In a similar way, the following intermediate was prepared: 3-(cyclopropylsulfonyl)-5-(trifluoromethoxy)benzoic acid

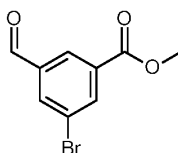
Synthesis of 3-bromo-5-(1-fluorocyclopropyl)benzoic acid

Step 1: methyl 3-formylbenzoate



3-formylbenzoic acid (95 g, 633 mmol) was dissolved in acetonitrile (1000 mL) and CDI (123 g, 759 mmol) was added in portions at room temperature. The mixture was stirred at room temperature 30 min and methanol (60.8 g, 1898 mmol) was added in one portion. The mixture was refluxed overnight, then cooled to room temperature, and evaporated in *vacuo* at 45 °C. The residue was dissolved in ethyl acetate (1000 mL), washed with 10 % aq. solution of NaHSO₄ (2×200 mL) and brine (1×100 mL), dried over Na₂SO₄, and evaporated in *vacuo* at 45 °C. Crude methyl 3-formylbenzoate (97 g, 93 % yield) as a colorless liquid was used in the next step without further purification.

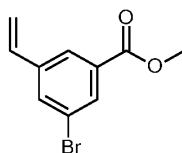
Step 2: methyl 3-bromo-5-formylbenzoate



Methyl 3-formylbenzoate (97 g, 591 mmol) was dissolved in 96% sulfuric acid (1000 mL), the solution was cooled to 0 °C and N-Bromosuccinimide (121 g, 680 mmol) was added in portions. The mixture was stirred at room temperature overnight then poured into ice and extract with dichloromethane (3×500 mL). The combined organic layers were washed with 10 % aq. solution of potassium carbonate (2×500 mL) and brine (1×100 mL), dried over Na₂SO₄, and evaporated in *vacuo* at 45 °C. The crude product was

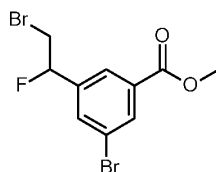
recrystallized from MTBE (100 mL) to obtain 130 g of methyl 3-bromo-5-formylbenzoate (91 % yield) as a white solid.

Step 3: methyl 3-bromo-5-vinylbenzoate



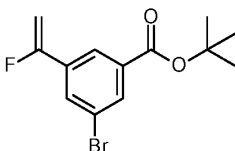
- 5 Methyltriphenylphosphonium iodide (249 g, 615 mmol) was suspended in THF (2500 mL) and the mixture was cooled to + 5 °C. Sodium *tert*-butoxide (68.4 g, 609 mmol) was then added in portions and the mixture was stirred at + 5 °C for 30 min. Methyl 3-bromo-5-formylbenzoate (130 g, 535 mmol) in THF (500 mL) was added dropwise to the mixture at + 5 °C and the mixture was stirred at room temperature overnight. Ethyl acetate (2500 mL) was added and the mixture was washed with brine
10 (3×500 mL), dried over Na₂SO₄, and evaporated in *vacuo* at 45 °C. The crude product was purified by column chromatography to obtain 49.9 g of methyl 3-bromo-5-vinylbenzoate (39 % yield) as a yellow oil.

Step 4: methyl 3-bromo-5-(2-bromo-1-fluoroethyl)benzoate



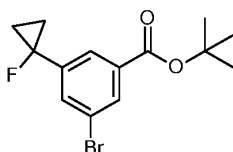
- Methyl 3-bromo-5-vinylbenzoate (44.9 g, 186 mmol) was dissolved in dichloromethane (450 mL) and the
15 mixture was cooled to + 5 °C, triethylamine trihydrofluoride (90.1 g, 559 mmol) and N-Bromosuccinimide (34.8 g, 196 mmol) were added to the mixture in one portion and the mixture was stirred at room temperature overnight. The mixture was washed with 10% aq. solution of potassium carbonate (2×200 mL) and brine (1×100 mL), dried over Na₂SO₄, and evaporated in *vacuo* at 45 °C. The
20 crude methyl 3-bromo-5-(2-bromo-1-fluoroethyl)benzoate was used in the next step without further purification. Yield 60 g (95%), brown oil.

Step 5: tert-butyl 3-bromo-5-(1-fluorovinyl)benzoate



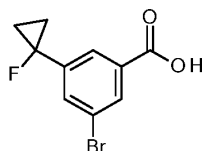
Potassium *tert*-butoxide (39.6 g, 353 mmol) was suspended in hexane (600 mL), the mixture was cooled to 0 °C and methyl 3-bromo-5-(2-bromo-1-fluoroethyl)benzoate (60 g, 176 mmol) in hexane (100 mL) was added dropwise to the mixture. The mixture was slowly heated up to room temperature and stirred at this temperature for 1 h. Ethyl acetate (300 mL) was added and the mixture was washed with brine (2×200 mL), dried over Na₂SO₄, and evaporated in *vacuo* at 45 °C. The crude product was purified by distillation in *vacuo* to obtain 12.4 g of *tert*-butyl 3-bromo-5-(1-fluorovinyl)benzoate (23% yield) as a colorless liquid; bp 110–112 °C / 1 mmHg.

Step 6: tert-butyl 3-bromo-5-(1-fluorocyclopropyl)benzoate



To a well stirred mixture of *tert*-butyl 3-bromo-5-(1-fluorovinyl)benzoate (16.5 g, 40 mmol) in diethyl ether (125 mL) in a liquid nitrogen bath under inert atmosphere, was added catalytic Pd(OAc)₂. Excess of diazomethane in diethyl ether was added by the help of a dropping funnel. The reaction temperature was gradually raised to room temperature and the mixture stirred for 1 h. After the completion of the reaction, the solvent was evaporated under reduced pressure. The crude mass was then purified by column chromatography (dichloromethane / hexane 0-50%) to give 2.4 g of *tert*-butyl 3-bromo-5-(1-fluorocyclopropyl)benzoate in 19% yield as yellow oil.

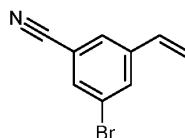
Step 7: 3-bromo-5-(1-fluorocyclopropyl)benzoic acid



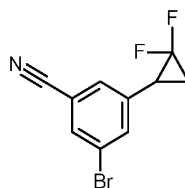
Tert-butyl 3-bromo-5-(1-fluorocyclopropyl)benzoate (2.4 g, 7.61 mmol) was dissolved in dichloromethane (11 mL) and trifluoroacetic acid (11 mL) was added. The mixture was stirred at room temperature for 2 h and evaporated in *vacuo* at 55 °C. The crude product was recrystallized from acetonitrile (3 mL) to obtain 1.73 g 3-bromo-5-(1-fluorocyclopropyl)benzoic acid (88% yield) as a white solid.

¹H NMR (500 MHz, DMSO-*d*₆) δ = 1.28 (m, 2H), 1.53 (m, 2H), 7.64 (s, 1H), 7.80 (s, 1H), 7.95 (s, 1H), 13.45 (s, 1H). Measured using a Bruker AVANCE DRX 500 MHz spectrometer.

ESI mass [m/z]: 256.9 [M-H]⁻

Synthesis 3-bromo-5-(2,2-difluorocyclopropyl)benzoic acid**Step 1: 3-bromo-5-vinylbenzonitrile**

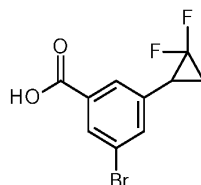
- [1,1'-Bis(diphenylphosphino)ferrocene]dichloropalladium(II) (PdCl₂dppf) (1.65 g, 2.22 mmol) was added to a refluxing solution (Ar atmosphere) of 3,5-dibromobenzonitrile (29 g, 111 mmol), potassium vinyltrifluoroborate (18.59 g, 139 mmol), Et₃N (28.1 g, 38.7 ml, 278 mmol) and p-methoxyphenol (2 mg) in *iso*-propanol (750 mL). The reaction mixture was stirred under reflux for 24 h. GCMS analysis of the reaction mixture showed incomplete conversion, therefore potassium vinyltrifluoroborate (1.5 g) and PdCl₂dppf (425 mg) were added to the reaction mixture which was then stirred under reflux for additional 4 h. After cooling down to room temperature the reaction mixture was evaporated in vacuo, and triturated with diethylether (750 mL) and water (750 mL). The organic layer was separated, filtered off, washed with brine (800 mL), separated and evaporated in vacuo to give 20 g of an oil containing (according to GCMS) 78% of 3-bromo-5-vinylbenzonitrile and 22% of 3,5-divinylbenzonitrile. This mixture was used in the next step.
- ¹H NMR (400 MHz, CDCl₃) δ= 7.75 (d, *J* = 1.8 Hz, 1H), 7.66 (d, *J* = 1.7 Hz, 1H), 7.59 (d, *J* = 1.5 Hz, 1H), 6.63 (dd, *J* = 17.6, 10.9 Hz, 1H), 5.88 – 5.79 (m, 1H), 5.46 (d, *J* = 10.9 Hz, 1H). Measured using a Varian Gemini 2000 spectrometer.

Step 2: 3-bromo-5-(2,2-difluorocyclopropyl)benzonitrile

- Sodium bromo(difluoro)acetate (56 g) was added in portions (over 20 - 15 min) to a solution of the crude product obtained in the previous step (11.44 g) in diglyme (25 mL) at 70 - 75 °C. After cooling down to room temperature, the reaction mixture was poured into water (600 mL) and the emulsion formed was extracted with diethylether (600 mL). The black tar was filtered off, the organic layer was separated, washed with brine (600 mL) and dried over MgSO₄. After filtration the filtrate was evaporated and dried *in vacuo* to give 17 g of a dark oil that contained 76 % of 3-bromo-5-(2,2-difluorocyclopropyl)benzonitrile and 24 % of 3,5-bis(2,2-difluorocyclopropyl)benzonitrile. Preparative flash column chromatography (hexane/EtOAc) afforded 6.9 g of pure 3-bromo-5-(2,2-difluorocyclopropyl)benzonitrile.

¹H NMR (400 MHz, CDCl₃): δ = 7.70 (t, *J* = 1.6 Hz, 1H), 7.61 (d, *J* = 1.8 Hz, 1H), 7.45 (d, *J* = 1.6 Hz, 1H), 2.75 (td, *J* = 12.0, 8.0 Hz, 1H), 1.95 (tdd, *J* = 11.7, 8.2, 5.3 Hz, 1H), 1.66 (dtd, *J* = 12.2, 8.2, 3.9 Hz, 1H). Measured using a Varian Gemini 2000 spectrometer.

Step 3: 3-bromo-5-(2,2-difluorocyclopropyl)benzoic acid



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A solution of 3-bromo-5-(2,2-difluorocyclopropyl)benzonitrile (3.87 g, 15 mmol) and sodium hydroxide (4.2 g, 105 mmol) in *iso*-propanol (60 mL) and water (25 mL) was stirred under reflux for 12 h. The reaction mixture was concentrated to 30-40 mL and the concentrate was added in portions to diluted HCl (10%, 200 mL). The precipitate formed was filtered off, washed with water and hexane and dried *in vacuo* (10 torr, 60 °C) for 3 h to obtain 3.74 g (90 %) 3-bromo-5-(2,2-difluorocyclopropyl)benzoic acid as a white solid.

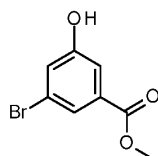
10

ESI mass [*m/z*]: 278.9 [*M*+*H*]⁺

¹H NMR (DMSO-*d*₆, 400 MHz): δ = 13.4 (br s, 1H), 7.94 (s, 1H), 7.83 (s, 1H), 7.77 (s, 1H), 3.22 – 3.12 (m, 1H), 2.20 – 1.90 (m, 2H).

15 Synthesis of 3-cyclopropyl-5-(difluoromethoxy)benzoic acid

Step 1: methyl 3-bromo-5-hydroxybenzoate

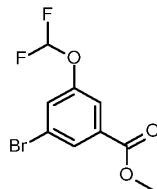


A solution of 3-bromo-5-hydroxybenzoic acid (49.9 g, 230 mmol) in MeOH (325 mL) was cooled by an ice bath to 7-8 °C. Then SOCl₂ (27.4 g, 16.79 mL, 230 mmol) was added dropwise to this solution over 25 min. The reaction mixture was warmed to room temperature, stirred under reflux for 3 h, cooled down to room temperature and then stirred for another 48 h at this temperature. All volatiles were removed in *vacuo* and the residue dissolved in ethyl acetate (400 mL). The solution was washed with NaHCO₃, brine, dried over Na₂SO₄ and the volatiles were removed under reduced pressure. The residue was triturated with hexanes (400 mL). The precipitate was filtered off, washed with hexanes/diethyl ether (1:1), dried at 110 °C to afford methyl 3-bromo-5-hydroxybenzoate (50.5 g) as a dark yellow powder.

25

^1H NMR (400 MHz, CDCl_3) δ = 7.73 (m, 1H), 7.51 (m, 1H), 7.26 (s, 1H), 7.23 (t, J = 2.1 Hz, 1H), 6.05 (br s, 1H), 3.92 (s, 3H). (recorded on a Varian Gemini 2000 machine)

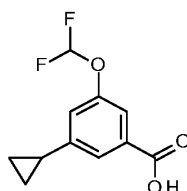
Step 2: methyl 3-bromo-5-(difluoromethoxy)benzoate



- 5 A mixture of methyl 3-bromo-5-hydroxybenzoate (23.1 g, 100 mmol), K_2CO_3 (41.5 g, 300 mmol) and $\text{ClF}_2\text{CCOONa}$ (45.7 g, 300 mmol) in DMF (350 mL) was stirred at 60-65°C for 2 h. The precipitate was then separated, washed with acetone and the filtrate was evaporated under reduced pressure. The residue was dissolved in diethyl ether (300 mL) and the solution was left to stand at rt for 12 h. A precipitate formed which was filtered off and washed with water. The filtrate was washed with brine (300 mL) and
- 10 the organic layer was evaporated under reduced pressure. The oily residue was dissolved in hexanes (250 mL) and kept at rt for 2 h. A precipitate formed which was removed by filtration and the filtrate was evaporated under reduced pressure. The residue was distilled under reduced pressure (3 tor) and the fraction with a boiling point between 80 and 85 °C was collected to afford 15.75 g methyl 3-bromo-5-(difluoromethoxy)benzoate.
- 15 ^1H NMR (400 MHz, CDCl_3) δ = 8.03 (t, J = 1.6 Hz, 1H), 7.75 – 7.70 (m, 1H), 7.49 (t, J = 2.1 Hz, 1H), 6.55 (t, J = 72.6 Hz, 1H), 3.93 (s, 3H). (recorded on a Varian Gemini 2000 machine)

^{19}F NMR (376 MHz, CDCl_3) δ = -84.89 (d, J = 72.7 Hz). (recorded on a Varian Gemini 2000 machine)

Step 3: 3-cyclopropyl-5-(difluoromethoxy)benzoic acid



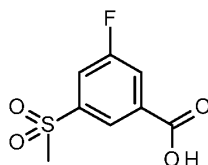
- 20 A mixture of methyl 3-bromo-5-(difluoromethoxy)benzoate (12.07 g, 52 mmol), K_3PO_4 (27.6 g, 130 mmol), Ph_3P (1.364 g, 5.2 mmol) and cyclopropylboronic acid (8.93 g, 104 mmol) in diglyme (250 mL) was heated to 100 °C and then $(\text{Ph}_3\text{P})_2\text{PdCl}_2$ (1.825g, 2.6 mmol) was added in one portion. The reaction mixture was stirred at 100 °C for 3 h, cooled down to rt and treated with a solution of NaOH (5 g) in water (250 mL). The reaction mixture was stirred for 48 h. Then the precipitate was filtered off and water 950
- 25 mL was added. The mixture was extracted with ethyl acetate (2x500 mL). The aqueous layer was acidified by concentrated hydrochloric acid to pH = 2 and kept at rt for 12 h. The precipitate was filtered off, washed with boiling water (4 x 200mL), dried at 110 °C to get 10.4 g of crude product. This material was dissolved

in hot toluene (30 mL) and the solution was diluted by hexanes to 150 mL. The mixture was filtrated hot, the filtrate cooled down to room temperature and than left in the refrigerator for 3 h. A precipitate formed which was filtered off, washed with hexanes and dried at 110 °C to get 7.7 g of crude material. The mother liquor was evaporated and the residue triturated with hexanes:diethyl ether (1:1). The insoluble material
5 was filtered off and combined with the 7.7 g crude material. The combined crude material was purified by sublimation (0.3 torr, 110 °C) to afford 9.7 g 3-cyclopropyl-5-(difluoromethoxy)benzoic acid.

^1H NMR (400 MHz, CDCl_3) δ = 12.10 (s, 1H), 7.67 (d, J = 1.6 Hz, 1H), 7.62 (t, J = 1.9 Hz, 1H), 7.08 (t, J = 2.0 Hz, 1H), 6.55 (t, J = 73.5 Hz, 1H), 2.06 – 1.89 (m, 1H), 1.15 – 0.96 (m, 2H), 0.85 – 0.69 (m, 2H). (recorded on a Varian Gemini 2000 machine)

10 ^{19}F NMR (376 MHz, CDCl_3) δ = -84.24 (d, J = 73.5 Hz). (recorded on a Varian Gemini 2000 machine)

Synthesis of 3-fluoro-5-(methylsulfonyl)benzoic acid (INT-21)



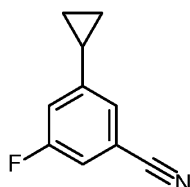
To a solution of 500 mg (2.68 mmol) 3-fluoro-5-(methylsulfonyl)benzoic acid dissolved in 40 mL CH_2Cl_2 were added 0.51 mL (13.4 mmol) formic acid and 1.92 mL (18.7 mmol) of an aqueous 30 % hydrogen
15 peroxide solution. The reaction mixture was stirred overnight at room temperature. It was then quenched by the addition of an aqueous $\text{Na}_2\text{S}_2\text{O}_3$ solution. The mixture was acidified using 10 M hydrochloric acid. The precipitate formed was removed by filtration and the filtrate was repeatedly extracted with ethyl acetate. The solvent was removed from the combined organic layers under reduced pressure to provide 677 mg of a residue containing 3-fluoro-5-(methylsulfonyl)benzoic acid which was used without further
20 purification.

^1H NMR ($\text{DMSO}-d_6$, 400 MHz): δ = 13.9 (br s, 1H), 8.25 (t, J = 1.4 Hz, 1H), 8.12 – 8.00 (m, 2H). (Signal of CH_3 -group is hidden under solvent signal)

ESI mass [m/z]: 217.0 [$\text{M}-\text{H}$] $^-$

Synthesis of 3-cyclopropyl-5-(cyclopropylsulfonyl)benzoic acid (INT-19)

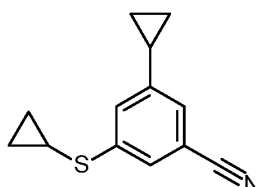
25 **Step 1: 3-cyclopropyl-5-fluorobenzonitrile**



Following the procedure described for 3-cyano-5-cyclopropylbenzoic acid (INT-15) 3-cyclopropyl-5-fluorobenzonitrile was obtained from 3-bromo-5-fluorobenzonitrile.

$^1\text{H-NMR}$ (400 MHz, DMSO- d_6): δ = 7.60 – 7.54 (m, 1H), 7.46 (s, 1H), 7.37 – 7.30 (m, 1H), 2.05 - 1.97 (m, 1H), 1.07 – 0.98 (m, 2H), 0.85 – 0.75 (m, 2H).

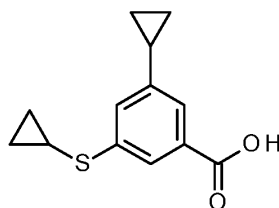
5 **Step 2: 3-cyclopropyl-5-(cyclopropylsulfanyl)benzonitrile**



3-cyclopropyl-5-(cyclopropylsulfanyl)benzonitrile was obtained following the procedure described for the synthesis of 3-(cyclopropylsulfanyl)-5-(difluoromethoxy)benzonitrile however with heating the reaction solution for 1 h at 100 °C.

10 ESI mass [m/z]: 216.1 [M+H]⁺

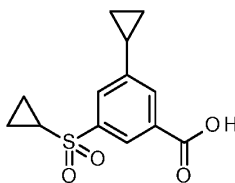
Step 3: 3-cyclopropyl-5-(cyclopropylsulfanyl)benzoic acid



3-cyclopropyl-5-(cyclopropylsulfanyl)benzoic acid was obtained following the procedure described for the synthesis 3-(difluoromethoxy)-5-(methylsulfanyl)benzoic acid however with heating the reaction solution for 14 h at 80 °C.

ESI mass [m/z]: 235.1 [M+H]⁺

Step 4: 3-cyclopropyl-5-(cyclopropylsulfonyl)benzoic acid (INT-19)



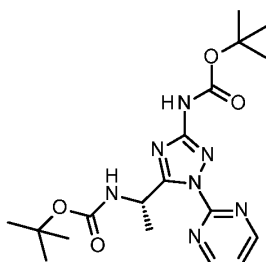
3-cyclopropyl-5-(cyclopropylsulfonyl)benzoic acid (INT-19) was obtained following the procedure described for 3-(difluoromethoxy)-5-(methylsulfonyl)benzoic acid.

¹H-NMR (400 MHz, DMSO-d₆): δ = 13.51 (s, 1H), 8.10 (t, *J* = 1.6 Hz, 1H), 7.90 (t, *J* = 1.6 Hz, 1H), 7.83 (t, *J* = 1.6 Hz, 1H), 3.02 - 2.95 (m, 1H), 2.25 - 2.15 (m, 1H), 1.18 - 1.02 (m, 6H), 0.85 - 0.78 (m, 2H).

ESI mass [*m/z*]: 267.1 [*M*+H]⁺

5 **Synthesis of *N*-[(1*S*)-1-(5-bromo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-methylsulfonyl-5-(trifluoromethyl)benzamide (example I-95)**

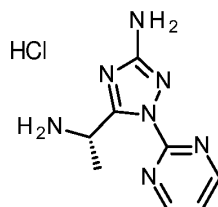
Step 1: *tert*-butyl *N*-[5-[(1*S*)-1-(*tert*-butoxycarbonylamino)ethyl]-1-pyrimidin-2-yl-1,2,4-triazol-3-yl]carbamate



- 10 To a solution of 8.00 g (42.2 mmol) *N*-Boc-alanine (Boc-Ala-OH purchased from Sigma-Aldrich) and 24.0 g (63.1 mmol) HATU in *N,N*-dimethylformamide (50 mL), 12.8 g (126.5 mmol) DIPEA (17.7 mL) and 8.0 g (42.0 mmol) 1-*N*-Boc-2-methyl-isothiourea (purchased from ABCR) were added, and the reaction mixture was stirred for 3 hours at room temperature. To the *in-situ* formed *tert*-butyl *N*-[(*Z*)-*N*-[(2*S*)-2-(*tert*-butoxycarbonylamino)propanoyl]-C-methylsulfonyl-carbonimidoyl]-*N*-carbamate were then added
- 15 acetic acid (2.8 mL) and 5.0 g (45.4 mmol) pyrimidin-2-yl-hydrazine. The reaction mixture was stirred for 2 hours at 50 °C. Afterwards, the solvent was removed under reduced pressure and the remaining crude product was chromatographed with an acetone/cyclohexane gradient to afford a product with 50-70% purity. Then, the product was purified by HPLC to afford 4.1 g (purity: 90%; yield: 23%) of the title compound. The enantiomeric excess of the chiral title compounds has been determined: *ee*-value > 100%.

20 ESI mass [*m/z*]: 406.2 [*M*+H]⁺

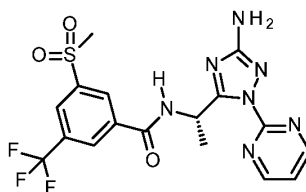
Step 2: 5-[(1*S*)-1-aminoethyl]-1-pyrimidin-2-yl-1,2,4-triazol-3-amine-hydrochloride (INT-23)



4.2 g (10.4 mmol) *tert*-butyl *N*-[5-[(1*S*)-1-(*tert*-butoxycarbonylamino)ethyl]-1-pyrimidin-2-yl-1,2,4-triazol-3-yl]carbamate were dissolved in 4N HCl-dioxane solution (40 mL) and the mixture was stirred 18 h at room temperature. Then the solvent was evaporated under reduced pressure to afford 4.0 g crude product of the title compound, which can be used for the amide coupling in step 3 without further purification.

ESI mass [m/z]: 206.1 [amine+H]⁺

Step 3: *N*-[(1*S*)-1-(5-amino-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-methylsulfonyl-5-(trifluoromethyl)benzamide

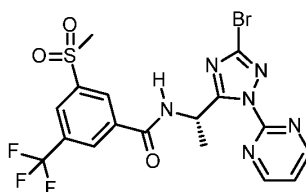


To 1.0 g (3.72 mmol) 3-chloro-5-methylsulfonyl-benzoic acid, 625 mg (4.83 mmol) DIPEA in acetonitrile (43.4 mL) and 1.55 g (4.07 mmol) HATU were added, and the reaction mixture was stirred for 10 minutes. Then 775.0 mg (3.77 mmol) 5-[(1*S*)-1-aminoethyl]-1-pyrimidin-2-yl-1,2,4-triazol-3-amine-hydrochloride in triethylamine (5 mL) were added and the reaction mixture was stirred at room temperature overnight. The reaction mixture was extracted with water and dichloromethane and the combined organic layers were dried over MgSO₄. After filtration the organic phase was concentrated under reduced pressure and the remaining crude product was chromatographed by preparative HPLC with a water/acetonitrile gradient to afford 291 mg (purity: 99%; yield: 17%) of the title compound.

¹H NMR (DMSO-d₆, 400 MHz): δ = 9.53 (d, *J* = 7.2 Hz, 1H), 8.86 (d, *J* = 4.8 Hz, 2H), 8.64 (s, 1H), 8.50 (s, 1H), 8.39 (s, 1H), 7.44 (t, *J* = 4.8 Hz, 1H), 6.10 – 6.00 (m, 1H), 5.75 (s, 2H), 3.38 (s, 3H), 1.61 (d, *J* = 6.8 Hz, 3H).

ESI mass [m/z]: 456.2 [M+H]⁺

Step 4: *N*-[(1*S*)-1-(5-bromo-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-methylsulfonyl-5-(trifluoromethyl)benzamide (example I-95)

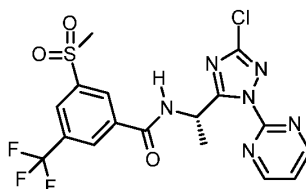


To 370 mg (0.81 mmol) *N*-[(1*S*)-1-(5-amino-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-methylsulfonyl-5-(trifluoromethyl)benzamide in acetonitrile (15 mL), 300 mg (1.34 mmol) Cu(II)-bromide was added, and then the reaction mixture was treated drop by drop at room temperature with 120 mg (1.16 mmol) *tert*-butyl nitrite. Then the reaction mixture was stirred 1 h at 70 °C temperature. The reaction mixture was treated with acetic acid ethyl ester and then washed with a brine and water. The organic phase was separated, dried and the solvent was evaporated. The remaining crude product was chromatographed by preparative HPLC to afford 146 mg (purity: 100 %; yield: 35 %) of the title compound. The enantiomeric excess of the chiral title compounds has been determined: *ee*-value > 99%.

ESI mass [m/z]: 521.1 [M+H]⁺

¹H-NMR (400 MHz, d₆-DMSO): see NMR peak list in table 1

Synthesis of *N*-[(1*S*)-1-(5-chloro-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-methylsulfonyl-5-(trifluoromethyl)benzamide (example I-96)



To 300 mg (0.69 mmol) *N*-[(1*S*)-1-(5-amino-2-pyrimidin-2-yl-1,2,4-triazol-3-yl)ethyl]-3-methylsulfonyl-5-(trifluoromethyl)benzamide in acetonitrile (15 ml), 150.5 mg (1.12 mmol) Cu(II)-chloride was added, and then the reaction mixture was treated drop by drop at room temperature with 95.1 mg (0.92 mmol) *tert*-butyl nitrite. Then the reaction mixture was stirred 1 h 70 °C temperature. The reaction mixture was treated with acetic acid ethyl ester and then washed with a saturated NaCl solution and water. The organic phase was separated, dried and the solvent was evaporated. The remaining crude product was chromatographed by MPLC with a cyclohexane/acetone gradient to afford 118 mg (purity: 97.5%; yield: 37 %) of the title compound. The enantiomeric excess of the chiral title compounds has been determined: *ee*-value > 100%.

ESI mass [m/z]: 475.0 [M+H]⁺

¹H-NMR (400 MHz, d₆-DMSO): see NMR peak list in table 1

Analytical data of the compounds

The determination of [M+H]⁺ or [M-H]⁻ by LC-MS under acidic chromatographic conditions was done with 1 ml formic acid per liter acetonitrile and 0.9 ml formic acid per liter Millipore water as eluents. The column Zorbax Eclipse Plus C18 50 mm * 2.1 mm was used. The temperature of the column oven was 55 °C.

Instruments:

LC-MS3: Waters UPLC with SQD2 mass spectrometer and SampleManager autosampler. Linear gradient 0.0 to 1.70 minutes from 10 % acetonitrile to 95 % acetonitrile, from 1.70 to 2.40 minutes constant 95 % acetonitrile, flow 0.85 ml/min.

- 5 **LC-MS6 and LC-MS7:** Agilent 1290 LC, Agilent MSD, HTS PAL autosampler. Linear gradient 0.0 to 1.80 minutes from 10 % acetonitrile to 95 % acetonitrile, from 1.80 to 2.50 minutes constant 95 % acetonitrile, flow 1.0 ml/min.

The determination of $[M+H]^+$ by LC-MS under neutral chromatographic conditions was done with acetonitrile and Millipore water containing 79 mg/l ammonia carbonate as eluents.

- 10 Instruments:

LC-MS4: Waters IClass Acquity with QDA mass spectrometer and FTN autosampler (column Waters Acquity 1.7 μ m 50 mm * 2.1 mm, oven temperature 45°C). Linear gradient 0.0 to 2.10 minutes from 10 % acetonitrile to 95 % acetonitrile, from 2.10 to 3.00 minutes constant 95 % acetonitrile, flow 0.7 ml/min.

- 15 **LC-MS5:** Agilent 1100 LC system with MSD mass spectrometer and HTS PAL autosampler (column: Zorbax XDB C18 1.8 μ m 50 mm * 4.6 mm, oven temperature 55°C). Linear gradient 0.0 to 4.25 minutes from 10 % acetonitrile to 95 % acetonitrile, from 4.25 to 5.80 minutes constant 95 % acetonitrile, flow 2.0 ml/min.

- 20 Optical rotations were measured using a Perkin Elmer model 341 polarimeter at a wavelength of 589 nm, a pathlength of 10 cm and a temperature of 20 °C. They are reported as specific rotations including the concentration "c" of the measured compound (in g / 100 mL) and the solvent used.

- 25 The enantiomeric excesses of the intermediate compound *tert*-butyl *N*-[5-[(1*S*)-1-(*tert*-butoxycarbonylamino)ethyl]-1-pyrimidin-2-yl]-1,2,4-triazol-3-yl]carbamate and the two examples I-95 and I-96 were determined using chiral HPLC: Chiralcel OD-RH column (4.6 mm x 150 mm x 5 μ m), room temperature, eluting with 0.1% phosphoric acid (A) and acetonitrile (B), gradient A:B 95/5 to 10/90, detecting at 210 nm.

- 30 The determination of the ^1H NMR data was effected with a Bruker Avance III 400 Mhz equipped with a 1.7 mm TCI cryo probe, a Bruker Avance III 600 Mhz equipped with a 5 mm multi-nuclear cryo probe or a Bruker Avance NEO 600 Mhz equipped with a 5 mm TCI cryo probe with tetramethylsilane as reference (0.0) and the solvents CD_3CN , CDCl_3 or $\text{D}_6\text{-DMSO}$.

The NMR data of selected examples are listed either in conventional form (δ values, multiplet splitting, number of hydrogen atoms) or as NMR peak lists.

NMR peak list method

5 The ^1H NMR data of selected examples are stated in the form of ^1H NMR peak lists. For each signal peak, first the δ value in ppm and then the signal intensity in round brackets are listed. The pairs of δ value–signal intensity numbers for different signal peaks are listed with separation from one another by semicolons.

The peak list for one example therefore takes the form of:

δ_1 (intensity₁); δ_2 (intensity₂);.....; δ_i (intensity_i);.....; δ_n (intensity_n)

10 The intensity of sharp signals correlates with the height of the signals in a printed example of an NMR spectrum in cm and shows the true ratios of the signal intensities. In the case of broad signals, several peaks or the middle of the signal and the relative intensity thereof may be shown in comparison to the most intense signal in the spectrum.

For calibration of the chemical shift of ^1H NMR spectra, we use tetramethylsilane and/or the chemical shift of the solvent, particularly in the case of spectra which are measured in DMSO. Therefore, the tetramethylsilane peak may but need not occur in NMR peak lists.

15 The lists of the ^1H NMR peaks are similar to the conventional ^1H NMR printouts and thus usually contain all peaks listed in a conventional NMR interpretation.

In addition, like conventional ^1H NMR printouts, they may show solvent signals, signals of stereoisomers of the target compounds which are likewise provided by the invention, and/or peaks of impurities.

20 In the reporting of compound signals within the delta range of solvents and/or water, our lists of ^1H NMR peaks show the standard solvent peaks, for example peaks of DMSO in DMSO- D_6 and the peak of water, which usually have a high intensity on average.

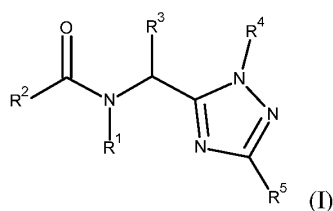
The peaks of stereoisomers of the target compounds and/or peaks of impurities usually have a lower intensity on average than the peaks of the target compounds (for example with a purity of > 90%).

25 Such stereoisomers and/or impurities may be typical of the particular preparation process. Their peaks can thus help in identifying reproduction of our preparation process with reference to "by-product fingerprints".

30 A person skilled in the art calculating the peaks of the target compounds by known methods (MestreC, ACD simulation, but also with empirically evaluated expected values) can, if required, isolate the peaks of the target compounds, optionally using additional intensity filters. This isolation would be similar to the peak picking in question in conventional ^1H NMR interpretation.

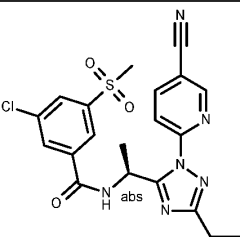
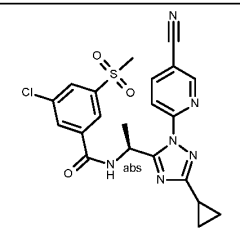
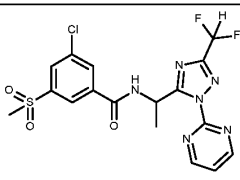
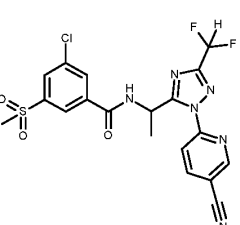
Further details of ^1H NMR peak lists can be found in the Research Disclosure Database Number 564025.

The compounds according to the invention described in table 1 below are likewise preferred compounds of the formula (I) according to the invention which are obtained according to or analogously to the preparation examples described above.

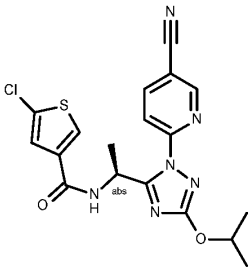
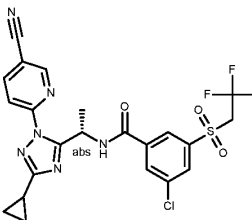
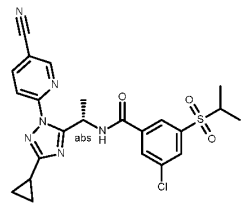


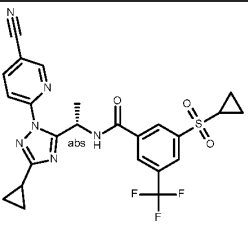
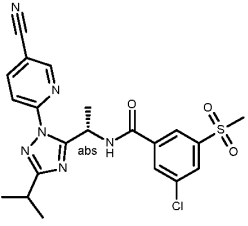
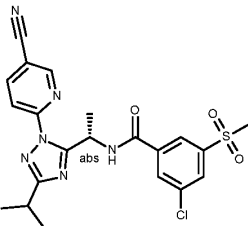
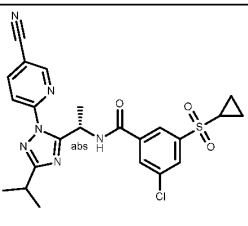
5 Table 1

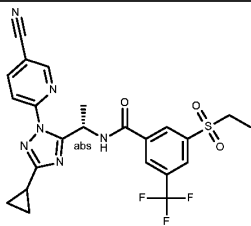
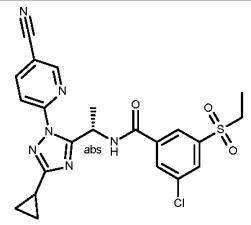
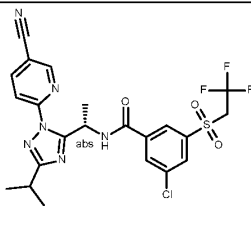
Example	Structure ²⁾	NMR Peaklist ¹⁾	ESI mass [m/z] ³⁾
I-1		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.1141 (2.3); 9.0962 (2.4); 9.0264 (3.8); 9.0247 (4.3); 9.0209 (4.2); 9.0191 (4.1); 8.5369 (3.5); 8.5313 (3.3); 8.5154 (3.7); 8.5098 (3.7); 8.3143 (0.4); 8.0202 (4.2); 8.0185 (4.4); 7.9987 (3.9); 7.9970 (4.1); 7.5844 (3.1); 7.5800 (4.9); 7.5758 (3.4); 7.4074 (2.8); 7.4038 (5.2); 7.4002 (3.1); 7.3268 (3.0); 7.3224 (5.0); 7.3181 (2.8); 6.0704 (0.4); 6.0533 (1.8); 6.0357 (2.8); 6.0181 (1.8); 6.0010 (0.4); 3.3458 (0.4); 3.3239 (121.1); 2.6710 (4.7); 2.6622 (0.7); 2.6528 (6.2); 2.6338 (4.3); 2.5250 (2.5); 2.5203 (3.4); 2.5115 (45.2); 2.5070 (93.9); 2.5024 (129.4); 2.4978 (97.6); 2.4934 (48.2); 2.3340 (0.6); 2.3293 (0.8); 2.3248 (0.6); 2.0742 (0.7); 2.0136 (0.5); 2.0010 (1.0); 1.9925 (1.0); 1.9888 (0.8); 1.9801 (2.1); 1.9675 (1.1); 1.9591 (1.1); 1.9466 (0.5); 1.7580 (0.4); 1.7389 (2.0); 1.7204 (4.0); 1.7021 (4.0); 1.6836 (2.0); 1.6644 (0.5); 1.6089 (9.5); 1.5915 (9.4); 1.2334 (0.5); 1.0185 (1.0); 1.0077 (3.4); 1.0021 (3.6); 0.9919 (1.6); 0.9867 (3.5); 0.9811 (3.5); 0.9711 (1.3); 0.9552 (7.8); 0.9369 (16.0); 0.9184 (7.1); 0.7682 (1.5); 0.7580 (3.4); 0.7554 (3.0); 0.7523 (3.2); 0.7456 (3.4); 0.7410 (3.2); 0.7296 (1.2); 0.1460 (0.4); 0.0080 (2.8); -0.0002 (96.9); -0.0085 (3.3); -0.1495 (0.4)	
I-2		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.4815 (1.1); 9.4637 (1.1); 9.0388 (1.9); 9.0370 (2.1); 9.0333 (2.1); 9.0314 (2.0); 8.5418 (1.8); 8.5362 (1.7); 8.5203 (1.9); 8.5147 (1.9); 8.3132 (0.7); 8.2989 (1.7); 8.2951 (3.3); 8.2913 (1.9); 8.2009 (1.6); 8.1963 (2.5); 8.1923 (1.8); 8.1488 (1.9); 8.1444 (2.7); 8.1399 (1.5); 8.0348 (2.2); 8.0329 (2.2); 8.0132 (2.0); 8.0114 (2.0); 6.1201 (0.8); 6.1026 (1.3); 6.0850 (0.8); 5.7533 (0.4); 3.3795 (0.3); 3.3337 (1.0); 3.3223 (15.8); 3.3157 (71.3); 2.6901 (16.0); 2.6802 (2.3); 2.6751 (1.1); 2.6704 (1.4); 2.6622 (3.5); 2.6431 (2.1); 2.5240 (3.7); 2.5193 (5.3); 2.5106 (69.0); 2.5061 (140.7); 2.5015 (186.2); 2.4968 (133.5); 2.4922 (63.0); 2.3376 (0.4); 2.3329 (0.8); 2.3283 (1.2); 2.3237 (0.8); 2.3191 (0.4); 2.1293 (0.4); 1.9883 (0.5); 1.7451 (1.0); 1.7267 (2.0); 1.7083 (2.0); 1.6899 (1.0); 1.6338 (4.6); 1.6164 (4.5); 1.2457 (0.4); 1.2300 (0.6); 1.2139 (0.3); 1.1753 (0.3); 0.9589 (4.0); 0.9405 (8.3); 0.9220 (3.6); 0.1459 (0.5); 0.0080 (3.8); -0.0002 (126.5); -0.0085 (3.8); -0.1495 (0.5)	

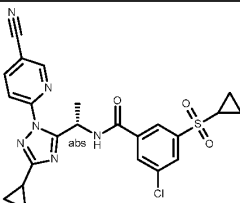
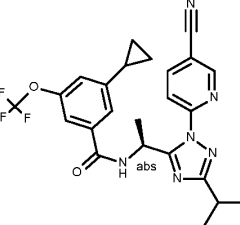
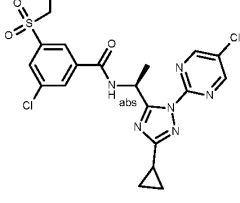
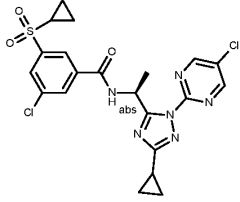
I-3		¹ H-NMR(600.1 MHz, CD ₃ CN lowT): δ= 8.8690 (2.8); 8.8655 (2.9); 8.4296 (0.5); 8.3283 (1.5); 8.3249 (1.6); 8.3140 (1.7); 8.3105 (1.8); 8.1959 (3.5); 8.0692 (3.0); 8.0545 (4.0); 8.0512 (3.4); 8.0296 (2.9); 6.2231 (1.1); 6.2114 (1.6); 6.1997 (1.1); 3.1200 (0.3); 3.1020 (16.0); 2.7776 (0.8); 2.7733 (0.7); 2.7648 (2.0); 2.7607 (2.1); 2.7521 (2.0); 2.7482 (2.1); 2.7394 (0.7); 2.7354 (0.8); 2.3047 (5.6); 1.9867 (1.5); 1.9785 (0.9); 1.9706 (8.2); 1.9666 (14.9); 1.9625 (21.9); 1.9584 (15.4); 1.9544 (8.0); 1.6592 (6.6); 1.6476 (6.6); 1.3115 (4.4); 1.2989 (8.8); 1.2863 (4.4); -0.0001 (4.0)	
I-4		¹ H-NMR(600.1 MHz, CD ₃ CN lowT): δ= 8.8478 (2.7); 8.8460 (2.7); 8.8445 (2.9); 8.3068 (1.4); 8.3038 (1.5); 8.2924 (1.6); 8.2894 (1.6); 8.1832 (3.4); 8.1816 (3.4); 8.0889 (3.4); 8.0865 (3.6); 8.0836 (3.5); 8.0342 (2.9); 8.0198 (2.6); 7.9840 (1.1); 7.9720 (1.2); 6.1526 (1.1); 6.1409 (1.7); 6.1292 (1.1); 3.1204 (16.0); 2.2948 (31.1); 2.2616 (0.5); 2.0799 (0.3); 2.0760 (0.5); 2.0717 (0.5); 2.0675 (0.5); 2.0635 (0.5); 2.0400 (0.6); 2.0334 (0.8); 2.0262 (1.3); 2.0189 (0.8); 2.0125 (0.7); 2.0048 (0.4); 1.9858 (3.4); 1.9774 (2.8); 1.9695 (29.0); 1.9656 (53.5); 1.9615 (79.4); 1.9577 (57.2); 1.9537 (30.4); 1.8505 (0.3); 1.8465 (0.5); 1.8427 (0.4); 1.6232 (6.7); 1.6117 (6.7); 1.0174 (1.0); 1.0118 (2.4); 1.0053 (1.1); 0.9979 (2.4); 0.9775 (0.3); 0.9531 (0.4); 0.9474 (0.5); 0.9360 (1.2); 0.9341 (1.1); 0.9280 (1.2); 0.9180 (1.0); 0.9104 (1.2); 0.9055 (1.0); 0.8959 (0.6); 0.0044 (0.5); -0.0001 (15.6)	
I-5		¹ H-NMR (400.2 MHz, d ₆ -DMSO): δ = 9.5334 (2.3); 9.5158 (2.3); 9.0292 (15.6); 9.0171 (16.0); 8.3149 (0.4); 8.2409 (3.4); 8.2371 (6.9); 8.2332 (4.0); 8.1424 (14.8); 8.1385 (13.9); 7.7041 (4.1); 7.6919 (7.9); 7.6798 (4.0); 7.3510 (2.1); 7.2191 (5.6); 7.0873 (2.6); 6.0668 (0.4); 6.0494 (1.8); 6.0319 (2.9); 6.0144 (1.8); 5.9972 (0.4); 3.3761 (0.7); 3.3210 (29.2); 3.3134 (29.9); 2.6902 (1.3); 2.6759 (0.6); 2.6714 (0.8); 2.6668 (0.6); 2.5249 (2.3); 2.5202 (3.4); 2.5115 (47.0); 2.5070 (98.2); 2.5024 (131.0); 2.4978 (93.4); 2.4932 (43.8); 2.3338 (0.6); 2.3293 (0.8); 2.3247 (0.6); 2.0864 (14.9); 1.6881 (9.9); 1.6707 (9.9); 1.3978 (3.2); 0.1460 (0.4); 0.0080 (3.6); -0.0002 (110.8); -0.0085 (3.2); -0.1495 (0.4)	
I-6		¹ H-NMR (400.2 MHz, d ₆ -DMSO): δ = 9.5650 (3.0); 9.5478 (3.0); 9.1052 (4.9); 9.1037 (5.2); 9.0999 (5.3); 9.0983 (4.8); 8.6283 (4.0); 8.6228 (3.8); 8.6069 (4.3); 8.6014 (4.2); 8.3330 (0.6); 8.3292 (1.3); 8.3254 (0.8); 8.3155 (0.4); 8.3032 (0.7); 8.2984 (1.2); 8.2940 (0.8); 8.2837 (4.1); 8.2799 (7.6); 8.2762 (4.5); 8.2522 (0.8); 8.2483 (1.0); 8.2435 (0.6); 8.1877 (3.7); 8.1832 (6.6); 8.1792 (4.6); 8.1593 (4.9); 8.1550 (6.8); 8.1505 (3.3); 8.0985 (5.4); 8.0971 (5.2); 8.0772 (5.0); 8.0756 (4.9); 7.3652 (2.4); 7.2334 (6.1); 7.1018 (2.9); 6.1232 (0.4); 6.1058 (2.2); 6.0885 (3.4); 6.0712 (2.2); 6.0541 (0.4); 4.4152 (0.5); 4.3974 (1.7); 4.3797 (1.7); 4.3619 (0.5); 3.4207 (0.4); 3.3673 (6.0); 3.3219 (100.7); 2.8914 (0.7); 2.7324 (0.6); 2.6758 (0.9); 2.6713 (1.3); 2.6667 (0.9); 2.5248 (3.9); 2.5200 (5.8); 2.5113 (77.9); 2.5069 (157.2); 2.5023 (204.6); 2.4978 (145.7); 2.4933 (69.7); 2.3382 (0.4); 2.3338 (0.9); 2.3291 (1.2); 2.3246 (0.9); 2.0864 (13.8); 1.9806 (0.6); 1.9532 (0.7); 1.6812 (11.7); 1.6638 (11.7); 1.3977 (16.0); 1.3837 (0.3); 1.3746 (1.8); 1.3568 (3.8); 1.3391 (1.7); 0.1460 (1.0); 0.0178 (0.4); 0.0080 (7.8); -0.0001 (231.5); -0.0085 (8.2); -0.1495 (1.0)	

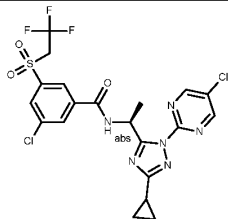
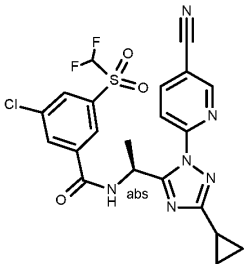
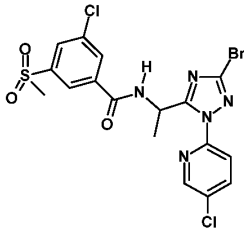
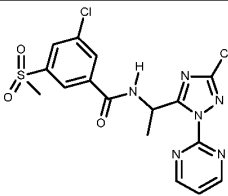
I-7		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8219 (1.9); 8.8175 (1.9); 8.2806 (1.3); 8.2751 (1.2); 8.2590 (1.5); 8.2535 (1.4); 8.1833 (1.6); 8.1797 (2.8); 8.1761 (1.7); 8.0839 (1.4); 8.0796 (2.4); 8.0754 (1.7); 8.0596 (1.9); 8.0555 (2.5); 8.0512 (1.2); 7.9913 (2.0); 7.9697 (1.7); 7.8582 (0.6); 7.8486 (0.4); 7.8398 (0.6); 6.1805 (1.0); 6.1628 (1.4); 6.1450 (1.0); 3.9984 (16.0); 3.1060 (15.2); 2.7269 (13.6); 2.2475 (0.3); 2.1317 (29.9); 2.1124 (0.5); 2.1064 (0.7); 2.0999 (0.5); 1.9712 (1.2); 1.9632 (2.5); 1.9514 (33.4); 1.9452 (62.5); 1.9391 (85.4); 1.9329 (58.8); 1.9268 (30.2); 1.7676 (0.5); 1.7614 (0.3); 1.6535 (6.4); 1.6362 (6.3); 1.2033 (0.5); 0.1455 (1.6); 0.0230 (0.5); 0.0074 (14.8); -0.0006 (319.8); -0.0085 (15.8); -0.1499 (1.6)
I-8		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8084 (2.1); 8.8031 (2.0); 8.2688 (1.4); 8.2633 (1.4); 8.2472 (1.6); 8.2417 (1.6); 7.9704 (2.2); 7.9488 (2.0); 7.7701 (2.9); 7.7659 (3.0); 7.4242 (0.5); 7.4074 (0.6); 7.2972 (2.9); 7.2930 (2.9); 6.1087 (1.0); 6.0906 (1.5); 6.0729 (1.1); 5.4461 (1.8); 4.0676 (0.6); 4.0498 (0.6); 3.9904 (16.0); 2.1334 (16.0); 1.9715 (2.6); 1.9638 (1.1); 1.9518 (14.1); 1.9457 (26.8); 1.9395 (37.5); 1.9334 (25.9); 1.9272 (13.4); 1.6092 (6.9); 1.5919 (6.9); 1.4365 (4.9); 1.2215 (0.6); 1.2037 (1.3); 1.1859 (0.6); 0.1456 (0.7); 0.0078 (5.8); -0.0002 (142.1); -0.0084 (6.8); -0.1498 (0.7)
I-9		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8191 (1.8); 8.8173 (2.0); 8.8136 (2.0); 8.8119 (1.9); 8.2725 (1.4); 8.2669 (1.4); 8.2508 (1.6); 8.2453 (1.6); 8.1928 (1.6); 8.1890 (3.0); 8.1853 (1.9); 8.0920 (1.5); 8.0875 (2.4); 8.0834 (1.8); 8.0603 (1.8); 8.0561 (2.5); 8.0515 (1.4); 7.9763 (2.0); 7.9745 (2.1); 7.9546 (1.8); 7.9529 (1.8); 7.9133 (0.6); 7.8956 (0.6); 6.1803 (1.0); 6.1623 (1.4); 6.1444 (1.0); 5.4473 (2.1); 4.3851 (1.3); 4.3674 (4.0); 4.3497 (4.1); 4.3321 (1.3); 4.0677 (0.3); 4.0501 (0.4); 3.1088 (16.0); 2.4639 (0.4); 2.1579 (102.2); 2.1135 (0.3); 2.1074 (0.4); 1.9718 (1.7); 1.9643 (2.0); 1.9581 (3.2); 1.9524 (26.7); 1.9462 (50.2); 1.9401 (69.2); 1.9339 (47.6); 1.9277 (24.4); 1.7686 (0.4); 1.6507 (6.6); 1.6334 (6.6); 1.4368 (0.6); 1.4113 (4.2); 1.3936 (8.7); 1.3760 (4.2); 1.2766 (0.4); 1.2706 (0.3); 1.2215 (0.4); 1.2037 (0.8); 1.1859 (0.4); 0.1459 (1.2); 0.0079 (12.4); -0.0002 (265.5); -0.0085 (11.6); -0.1496 (1.2)
I-10		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8058 (2.1); 8.8043 (2.1); 8.8005 (2.2); 8.2602 (1.6); 8.2547 (1.5); 8.2386 (1.8); 8.2330 (1.7); 7.9575 (2.3); 7.9559 (2.2); 7.9359 (2.0); 7.9343 (1.9); 7.7719 (3.2); 7.7676 (3.1); 7.4218 (0.6); 7.4054 (0.6); 7.2998 (3.4); 7.2956 (3.2); 6.1077 (1.1); 6.0899 (1.5); 6.0716 (1.1); 4.3760 (1.3); 4.3584 (4.2); 4.3407 (4.3); 4.3231 (1.4); 2.1340 (19.6); 2.1068 (0.4); 1.9716 (0.6); 1.9638 (1.5); 1.9519 (19.9); 1.9457 (37.0); 1.9396 (50.4); 1.9334 (34.6); 1.9273 (17.6); 1.6066 (7.6); 1.5893 (7.6); 1.4368 (16.0); 1.4077 (4.6); 1.3901 (9.4); 1.3725 (4.5); 0.1459 (0.9); 0.0078 (11.2); -0.0002 (194.3); -0.0086 (8.7); -0.0190 (0.4); -0.1495 (0.9)
I-11		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8161 (1.9); 8.8145 (2.1); 8.8107 (2.2); 8.8090 (2.0); 8.2656 (1.5); 8.2600 (1.4); 8.2439 (1.6); 8.2384 (1.6); 8.1888 (1.7); 8.1853 (3.0); 8.1818 (1.9); 8.0896 (1.5); 8.0854 (2.5); 8.0815 (1.7); 8.0598 (1.9); 8.0556 (2.6); 8.0511 (1.4); 7.9729 (2.2); 7.9527 (1.8); 7.9512 (1.9); 7.8732 (0.6); 7.8544 (0.6); 6.1811 (1.0); 6.1628 (1.4); 6.1452 (1.0); 5.4460 (1.9); 5.0130 (0.4); 4.9976 (1.1); 4.9822 (1.6); 4.9668 (1.2); 4.9515 (0.5); 3.1211 (0.3); 3.1077 (16.0); 2.1357 (24.8); 2.1129 (0.3); 2.1067 (0.4); 1.9636 (1.1); 1.9517 (16.8); 1.9456 (32.6); 1.9394 (46.3); 1.9332 (32.0); 1.9271 (16.5); 1.9052 (0.5); 1.8475 (0.5); 1.6476 (7.0); 1.6303 (6.9); 1.5546 (0.3); 1.3936

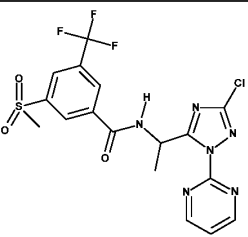
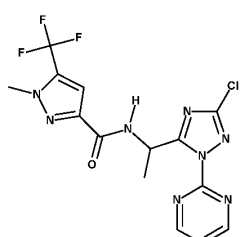
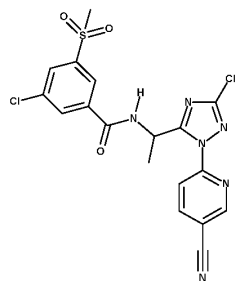
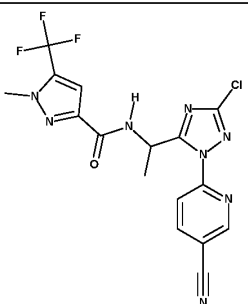
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I-12		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 8.9978 (1.8); 8.9930 (1.8); 8.8968 (1.1); 8.8793 (1.1); 8.5234 (1.2); 8.5179 (1.1); 8.5018 (1.2); 8.4964 (1.2); 8.0525 (2.5); 8.0484 (2.4); 7.9445 (1.8); 7.9229 (1.7); 7.4967 (2.5); 7.4927 (2.4); 5.9971 (0.7); 5.9798 (1.2); 5.9624 (0.7); 4.9282 (0.8); 4.9128 (1.1); 4.8974 (0.8); 3.3234 (65.7); 2.6710 (0.7); 2.5063 (92.6); 2.5022 (113.7); 2.4980 (82.3); 2.3290 (0.7); 1.5744 (4.1); 1.5570 (4.1); 1.3980 (16.0); 1.3576 (5.2); 1.3413 (8.7); 1.3248 (5.1); -0.0001 (14.5)	
I-13		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4864 (4.2); 9.4689 (4.2); 9.0087 (7.5); 9.0046 (7.4); 8.5331 (5.4); 8.5276 (5.1); 8.5116 (5.7); 8.5060 (5.6); 8.3146 (0.8); 8.3044 (4.6); 8.3008 (9.9); 8.2970 (7.5); 8.2908 (6.9); 8.2862 (9.2); 8.2825 (4.9); 8.1765 (5.8); 8.1721 (9.6); 8.1677 (5.1); 8.0155 (7.6); 7.9939 (7.0); 6.0988 (0.6); 6.0816 (3.0); 6.0642 (4.7); 6.0468 (3.0); 6.0295 (0.6); 5.1431 (2.2); 5.1186 (7.2); 5.0939 (7.6); 5.0692 (2.6); 3.3267 (443.2); 3.2527 (0.3); 2.6760 (1.5); 2.6716 (2.2); 2.6672 (1.6); 2.5249 (6.2); 2.5114 (140.1); 2.5071 (284.6); 2.5027 (373.4); 2.4982 (267.7); 2.4938 (128.9); 2.3339 (1.6); 2.3294 (2.2); 2.3250 (1.6); 2.0903 (0.8); 2.0779 (1.9); 2.0693 (2.0); 2.0571 (3.8); 2.0451 (2.3); 2.0365 (2.0); 2.0244 (1.0); 1.6180 (16.0); 1.6006 (16.0); 1.0321 (0.4); 1.0220 (0.5); 1.0025 (5.0); 0.9981 (6.7); 0.9816 (5.0); 0.9772 (6.8); 0.9597 (0.9); 0.9503 (0.7); 0.9174 (1.0); 0.9042 (1.2); 0.8935 (3.9); 0.8876 (2.4); 0.8812 (4.4); 0.8757 (3.8); 0.8685 (3.3); 0.8563 (2.8); 0.8414 (1.1); 0.8332 (0.7); -0.0001 (5.5)	
I-14		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4667 (2.7); 9.4492 (2.8); 9.0080 (4.7); 9.0065 (5.1); 9.0026 (5.0); 9.0011 (4.8); 8.5311 (3.9); 8.5256 (3.7); 8.5096 (4.2); 8.5040 (4.2); 8.3138 (0.3); 8.2280 (3.6); 8.2238 (6.1); 8.2195 (4.6); 8.1968 (4.3); 8.1931 (7.4); 8.1894 (3.8); 8.0537 (4.2); 8.0493 (6.6); 8.0449 (3.8); 8.0118 (5.0); 8.0102 (5.1); 7.9903 (4.7); 7.9886 (4.8); 6.0924 (0.4); 6.0752 (2.1); 6.0577 (3.3); 6.0403 (2.1); 6.0231 (0.4); 3.6141 (0.7); 3.5971 (2.1); 3.5801 (3.0); 3.5630 (2.2); 3.5461 (0.8); 3.3244 (232.4); 2.6809 (0.4); 2.6765 (0.8); 2.6719 (1.1); 2.6674 (0.8); 2.6629 (0.4); 2.5254 (3.1); 2.5207 (4.6); 2.5120 (66.4); 2.5075 (139.6); 2.5030 (186.0); 2.4984 (131.2); 2.4938 (61.4); 2.3343 (0.8); 2.3298 (1.0); 2.3252 (0.8); 2.3207 (0.3); 2.0912 (0.6); 2.0791 (1.3); 2.0705 (1.4); 2.0673 (1.1); 2.0584 (2.7); 2.0463 (1.6); 2.0377 (1.4); 2.0254 (0.7); 1.6170 (11.1); 1.5996 (11.1); 1.1864 (14.6); 1.1745 (16.0); 1.1694 (16.0); 1.1576 (14.3); 1.0038 (3.3); 0.9988 (4.6); 0.9829 (3.1); 0.9779 (4.8); 0.9592 (0.5); 0.9514 (0.5); 0.9191 (0.7); 0.9070 (0.8); 0.8950 (2.8); 0.8897 (1.5); 0.8827 (3.0); 0.8767 (2.2); 0.8690 (1.9); 0.8638 (1.5); 0.8590 (1.7); 0.8567 (1.7); 0.8412 (0.8); 0.8331 (0.5); -0.0001 (3.1)	
I-15		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6124 (3.8); 9.5949 (3.9); 9.0196 (6.7); 9.0178 (7.4); 9.0141 (7.4); 9.0122 (7.1); 8.5735 (7.7); 8.5344 (5.6); 8.5289 (5.4); 8.5129 (6.0); 8.5073 (6.1); 8.4809 (6.8); 8.3566 (6.9); 8.3121 (0.9); 8.0192 (7.2); 8.0175 (7.4); 7.9977 (6.8); 7.9959 (6.9); 6.1335 (0.6); 6.1162 (2.9); 6.0987 (4.6); 6.0812 (3.0); 6.0641 (0.6); 4.0567 (0.3); 4.0390 (1.0); 4.0212 (1.0); 4.0035 (0.3); 3.3131 (99.0); 3.2892 (0.3); 3.1258 (0.8); 3.1140 (1.8); 3.1062 (2.0); 3.1021 (1.3); 3.0945 (3.8); 3.0864 (1.4); 3.0824 (2.0); 3.0748 (1.9); 3.0626 (0.9); 2.6801 (0.4); 2.6758 (1.0); 2.6713 (1.3); 2.6667 (1.0); 2.5247 (3.9); 2.5200 (5.8); 2.5114 (81.7); 2.5069 (171.5); 2.5023 (229.2); 2.4977 (162.6); 2.4932	

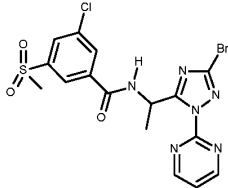
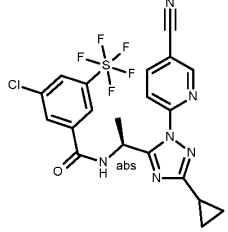
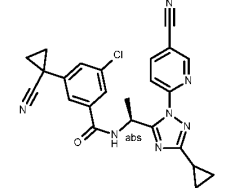
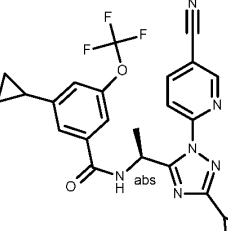
		(76.9); 2.3337 (0.9); 2.3292 (1.3); 2.3246 (0.9); 2.3201 (0.5); 2.0934 (0.8); 2.0812 (1.8); 2.0726 (2.0); 2.0694 (1.5); 2.0604 (3.8); 2.0482 (2.2); 2.0397 (2.0); 2.0274 (1.0); 1.9886 (4.5); 1.6394 (16.0); 1.6220 (15.9); 1.2531 (0.5); 1.2414 (0.7); 1.2294 (2.8); 1.2169 (4.9); 1.2088 (5.2); 1.1978 (3.4); 1.1938 (2.7); 1.1876 (1.0); 1.1758 (3.4); 1.1580 (1.5); 1.1495 (0.5); 1.1403 (0.6); 1.1318 (1.4); 1.1180 (5.2); 1.1103 (4.4); 1.0984 (5.0); 1.0902 (4.1); 1.0762 (0.7); 1.0328 (0.4); 1.0239 (0.5); 1.0045 (4.8); 0.9998 (6.7); 0.9836 (4.7); 0.9789 (6.8); 0.9615 (0.8); 0.9525 (0.8); 0.9225 (0.9); 0.9101 (1.1); 0.8989 (3.9); 0.8929 (2.2); 0.8866 (4.3); 0.8812 (3.5); 0.8739 (3.1); 0.8700 (2.4); 0.8617 (2.6); 0.8470 (1.1); 0.8384 (0.6); 0.1460 (0.7); 0.0080 (5.7); -0.0002 (187.1); -0.0085 (6.6); -0.1497 (0.7)	
I-16		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4805 (2.1); 9.4626 (2.2); 9.0294 (4.0); 9.0253 (3.9); 9.0240 (3.9); 8.5400 (2.8); 8.5345 (2.7); 8.5185 (3.0); 8.5130 (3.1); 8.3121 (0.4); 8.2905 (3.0); 8.2868 (5.6); 8.2831 (3.4); 8.1922 (2.8); 8.1877 (4.8); 8.1838 (3.3); 8.1463 (3.4); 8.1419 (5.1); 8.1375 (2.7); 8.0363 (3.8); 8.0349 (4.1); 8.0149 (3.5); 8.0133 (3.8); 6.1364 (0.3); 6.1193 (1.6); 6.1017 (2.6); 6.0841 (1.6); 6.0664 (0.3); 5.7521 (3.0); 3.3646 (0.6); 3.3332 (0.7); 3.3209 (26.6); 3.3132 (31.0); 3.0589 (0.8); 3.0416 (2.1); 3.0242 (3.0); 3.0069 (2.3); 2.9896 (0.9); 2.6756 (0.6); 2.6710 (0.8); 2.6666 (0.6); 2.5246 (2.0); 2.5198 (3.1); 2.5110 (49.2); 2.5066 (102.6); 2.5021 (136.2); 2.4976 (97.4); 2.4931 (46.6); 2.3336 (0.6); 2.3290 (0.8); 2.3245 (0.6); 1.6393 (9.1); 1.6220 (9.1); 1.3659 (0.7); 1.3503 (0.7); 1.3020 (15.7); 1.2952 (16.0); 1.2847 (15.6); 1.2779 (15.6); 1.2306 (0.4); 0.1459 (0.4); 0.0080 (3.4); -0.0001 (110.6); -0.0085 (3.9); -0.1496 (0.4)	
I-17		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4896 (2.1); 9.4718 (2.1); 9.0256 (3.6); 9.0240 (3.9); 9.0202 (3.9); 9.0185 (3.7); 8.5383 (3.0); 8.5327 (2.9); 8.5168 (3.3); 8.5112 (3.3); 8.2370 (2.7); 8.2333 (5.6); 8.2295 (3.6); 8.2099 (3.3); 8.2053 (4.8); 8.2013 (2.8); 8.0933 (3.3); 8.0888 (5.1); 8.0845 (2.9); 8.0338 (3.9); 8.0322 (4.0); 8.0123 (3.6); 8.0106 (3.8); 6.1303 (0.3); 6.1132 (1.6); 6.0956 (2.5); 6.0780 (1.6); 6.0606 (0.3); 5.7535 (0.9); 3.4451 (1.7); 3.4267 (5.8); 3.4083 (5.9); 3.3900 (1.7); 3.3159 (62.8); 3.0599 (0.8); 3.0425 (2.2); 3.0252 (3.0); 3.0079 (2.3); 2.9906 (0.9); 2.6756 (0.6); 2.6711 (0.9); 2.6665 (0.7); 2.6621 (0.5); 2.5246 (2.4); 2.5199 (3.7); 2.5112 (54.3); 2.5068 (113.7); 2.5022 (151.2); 2.4976 (105.8); 2.4930 (49.3); 2.3336 (0.6); 2.3290 (0.9); 2.3244 (0.6); 1.6406 (8.8); 1.6232 (8.8); 1.3014 (15.9); 1.2949 (16.0); 1.2841 (15.8); 1.2776 (15.6); 1.2446 (0.5); 1.2287 (0.9); 1.2131 (0.5); 1.1323 (6.4); 1.1232 (1.0); 1.1140 (14.6); 1.0955 (6.2); 0.1459 (0.5); 0.0080 (4.0); -0.0001 (125.9); -0.0085 (3.9); -0.1495 (0.5)	
I-18		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4990 (2.3); 9.4812 (2.3); 9.0274 (4.2); 9.0233 (4.2); 8.5400 (2.9); 8.5345 (2.8); 8.5185 (3.1); 8.5130 (3.2); 8.2510 (3.1); 8.2474 (5.7); 8.2437 (3.6); 8.2012 (3.2); 8.1967 (5.0); 8.1928 (3.2); 8.1180 (3.5); 8.1136 (5.3); 8.1093 (3.0); 8.0356 (4.1); 8.0140 (3.8); 6.1346 (0.4); 6.1176 (1.6); 6.1000 (2.6); 6.0824 (1.7); 6.0650 (0.4); 5.7522 (5.7); 3.3153 (102.9); 3.0603 (0.8); 3.0430 (2.1); 3.0351 (0.7); 3.0256 (3.4); 3.0153 (1.4); 3.0084 (2.8); 3.0038 (2.4); 2.9914 (2.0); 2.9839 (1.2); 2.9722 (0.6); 2.6758 (0.5); 2.6713 (0.7); 2.6666 (0.6); 2.5247 (2.2); 2.5111 (46.2); 2.5069 (93.9); 2.5024 (123.6); 2.4979 (89.1); 2.4935 (43.5); 2.3337 (0.5); 2.3291 (0.7); 2.3246 (0.5); 1.6416 (9.2); 1.6241 (9.2); 1.3022 (15.7); 1.2959 (16.0); 1.2849 (15.7); 1.2786 (15.5); 1.2458 (0.4); 1.2299 (0.7); 1.2226 (0.5); 1.2138 (0.6); 1.2000 (1.7); 1.1873 (3.1); 1.1778 (3.1); 1.1684 (2.0); 1.1579 (0.8); 1.1456 (0.6); 1.1393 (0.4); 1.1336 (0.5); 1.1151 (1.0); 1.1022 (3.0); 1.0954 (2.5); 1.0824 (2.9); 1.0773 (2.0); 1.0735 (2.2); 1.0610 (0.4); 0.1459 (0.3); 0.0079 (2.8); -0.0002 (79.8); -0.0083 (3.2); -0.1496 (0.4)	

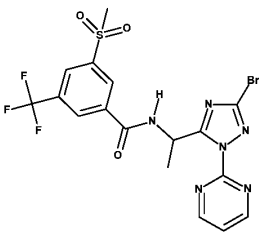
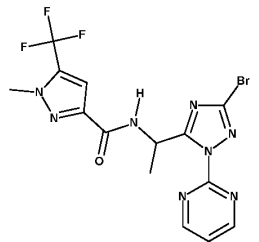
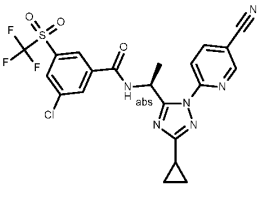
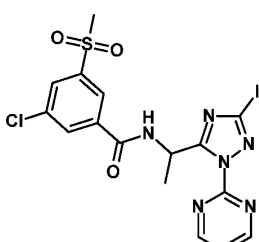
I-19		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6047 (2.4); 9.5871 (2.5); 9.0167 (4.2); 9.0149 (4.8); 9.0112 (4.7); 9.0093 (4.7); 8.5635 (4.8); 8.5329 (3.6); 8.5273 (3.5); 8.5113 (3.9); 8.5058 (4.0); 8.4906 (4.3); 8.3361 (4.4); 8.3129 (0.3); 8.0166 (4.4); 8.0148 (4.8); 7.9950 (4.2); 7.9932 (4.6); 6.1306 (0.4); 6.1133 (1.8); 6.0959 (2.9); 6.0784 (1.9); 6.0611 (0.4); 3.5079 (1.8); 3.4895 (6.4); 3.4711 (6.5); 3.4528 (1.9); 3.3181 (136.9); 2.6761 (0.7); 2.6715 (1.0); 2.6670 (0.7); 2.6625 (0.3); 2.5251 (2.7); 2.5203 (4.3); 2.5116 (59.4); 2.5072 (124.3); 2.5026 (165.2); 2.4980 (117.4); 2.4935 (55.5); 2.3339 (0.7); 2.3294 (1.0); 2.3247 (0.7); 2.0928 (0.5); 2.0807 (1.2); 2.0720 (1.2); 2.0690 (1.0); 2.0599 (2.4); 2.0477 (1.4); 2.0392 (1.3); 2.0269 (0.6); 1.9889 (0.6); 1.6383 (10.0); 1.6209 (9.9); 1.1759 (0.4); 1.1483 (7.0); 1.1300 (16.0); 1.1115 (6.8); 1.0040 (3.0); 0.9995 (4.1); 0.9831 (3.0); 0.9786 (4.2); 0.9613 (0.6); 0.9518 (0.5); 0.9217 (0.6); 0.9093 (0.7); 0.8982 (2.3); 0.8922 (1.4); 0.8858 (2.6); 0.8804 (2.2); 0.8730 (2.0); 0.8694 (1.5); 0.8651 (1.3); 0.8608 (1.7); 0.8502 (0.5); 0.8464 (0.7); 0.8381 (0.4); 0.1459 (0.5); 0.0080 (4.2); -0.0001 (131.4); -0.0084 (4.6); -0.1496 (0.5)	
I-20		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4504 (2.3); 9.4329 (2.4); 9.0107 (4.4); 9.0067 (4.2); 9.0052 (4.2); 8.5312 (3.2); 8.5257 (3.1); 8.5097 (3.4); 8.5041 (3.4); 8.3126 (0.4); 8.2307 (3.0); 8.2270 (6.2); 8.2232 (4.0); 8.2076 (3.7); 8.2030 (5.3); 8.1991 (3.1); 8.0923 (3.6); 8.0878 (5.6); 8.0835 (3.2); 8.0119 (4.2); 8.0104 (4.5); 7.9904 (4.0); 7.9888 (4.2); 6.0956 (0.4); 6.0784 (1.8); 6.0609 (2.9); 6.0434 (1.8); 6.0263 (0.4); 4.0385 (0.7); 4.0207 (0.8); 3.4458 (1.8); 3.4274 (6.3); 3.4090 (6.4); 3.3906 (1.9); 3.3131 (70.3); 2.6753 (0.9); 2.6706 (1.3); 2.6660 (1.0); 2.6616 (0.4); 2.5241 (3.7); 2.5193 (5.5); 2.5107 (80.4); 2.5062 (167.0); 2.5017 (222.0); 2.4971 (156.6); 2.4926 (73.7); 2.3330 (0.9); 2.3285 (1.3); 2.3238 (0.9); 2.3194 (0.4); 2.0902 (0.5); 2.0779 (1.1); 2.0692 (1.2); 2.0572 (2.3); 2.0450 (1.4); 2.0365 (1.2); 2.0242 (0.6); 1.9884 (3.2); 1.6158 (9.9); 1.5984 (9.8); 1.3980 (0.5); 1.2346 (0.3); 1.1932 (0.9); 1.1754 (1.8); 1.1577 (0.9); 1.1338 (7.0); 1.1154 (16.0); 1.0970 (6.8); 1.0220 (0.3); 1.0033 (3.0); 0.9983 (4.2); 0.9823 (3.0); 0.9774 (4.2); 0.9606 (0.5); 0.9510 (0.6); 0.9191 (0.6); 0.9067 (0.7); 0.8957 (2.5); 0.8894 (1.5); 0.8833 (2.8); 0.8778 (2.0); 0.8717 (2.2); 0.8676 (1.4); 0.8594 (1.7); 0.8446 (0.6); 0.8365 (0.4); 0.1459 (0.7); 0.0080 (5.2); -0.0001 (170.7); -0.0085 (5.6); -0.1497 (0.7)	
I-21		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5231 (2.1); 9.5054 (2.2); 9.0228 (3.7); 9.0211 (4.2); 9.0174 (4.1); 9.0156 (3.9); 8.5394 (3.2); 8.5338 (3.0); 8.5179 (3.4); 8.5123 (3.4); 8.3150 (0.7); 8.3107 (2.5); 8.3070 (5.3); 8.3031 (3.8); 8.2939 (3.6); 8.2892 (4.9); 8.2854 (2.7); 8.1785 (3.3); 8.1740 (5.3); 8.1696 (2.9); 8.0383 (4.0); 8.0365 (4.1); 8.0167 (3.7); 8.0149 (3.9); 6.1336 (0.3); 6.1169 (1.6); 6.0993 (2.5); 6.0817 (1.6); 6.0644 (0.3); 5.1416 (1.2); 5.1172 (3.8); 5.0926 (4.0); 5.0680 (1.4); 3.3256 (129.3); 3.0588 (0.8); 3.0415 (2.1); 3.0242 (3.0); 3.0068 (2.3); 2.9896 (0.9); 2.6808 (0.4); 2.6763 (0.7); 2.6717 (1.0); 2.6672 (0.8); 2.6627 (0.4); 2.5252 (3.2); 2.5205 (5.1); 2.5118 (65.8); 2.5073 (135.1); 2.5028 (177.1); 2.4982 (125.2); 2.4937 (58.6); 2.3386 (0.3); 2.3342 (0.8); 2.3296 (1.0); 2.3250 (0.7); 2.3204 (0.4); 1.6429 (8.6); 1.6255 (8.5); 1.2996 (15.8); 1.2936 (16.0); 1.2823 (15.8); 1.2763 (15.6); 0.1459 (0.5); 0.0081 (5.0); -0.0001 (143.5); -0.0085 (4.7); -0.1495 (0.6)	
I-22		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4713 (3.9); 9.4538 (4.0); 9.0190 (6.8); 9.0174 (7.4); 9.0136 (7.4); 9.0119 (7.2); 8.5362 (5.6); 8.5306 (5.4); 8.5146 (6.0); 8.5091 (6.0); 8.3148 (1.0); 8.3094 (1.1); 8.2566 (1.4); 8.2531 (1.1); 8.2463 (5.4); 8.2426 (10.6); 8.2388 (6.5); 8.2014 (5.8); 8.1968 (8.9); 8.1928 (5.7); 8.1212 (6.4); 8.1168 (9.6); 8.1124 (5.4); 8.0335 (1.0); 8.0289 (1.7); 8.0244 (1.1); 8.0166	

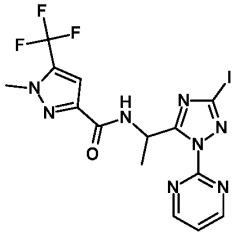
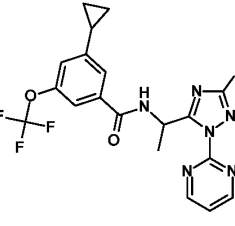
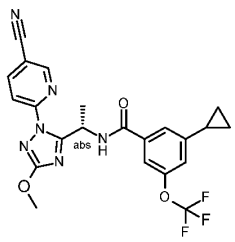
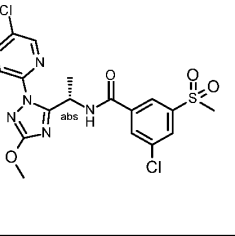
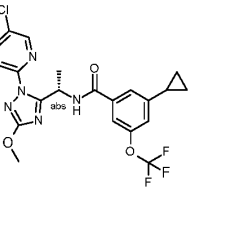
		(7.2); 8.0149 (7.6); 7.9950 (6.9); 7.9934 (7.3); 7.9117 (0.6); 7.8812 (0.6); 6.1901 (0.6); 6.1597 (0.6); 6.0998 (0.6); 6.0826 (3.0); 6.0652 (4.7); 6.0477 (3.0); 6.0304 (0.6); 4.2822 (0.4); 3.8017 (0.4); 3.7850 (0.3); 3.3256 (225.7); 3.0612 (0.4); 3.0494 (0.7); 3.0411 (1.1); 3.0291 (2.2); 3.0212 (2.0); 3.0170 (1.4); 3.0096 (3.8); 3.0013 (1.4); 2.9975 (2.1); 2.9898 (2.0); 2.9776 (1.0); 2.6809 (0.6); 2.6762 (1.3); 2.6716 (1.8); 2.6669 (1.7); 2.5252 (4.8); 2.5205 (7.0); 2.5117 (107.5); 2.5073 (226.8); 2.5027 (301.4); 2.4981 (213.6); 2.4936 (100.5); 2.3386 (0.6); 2.3341 (1.3); 2.3295 (1.7); 2.3249 (1.2); 2.3204 (0.6); 2.0910 (0.8); 2.0788 (2.0); 2.0749 (6.0); 2.0703 (2.2); 2.0670 (1.6); 2.0581 (3.9); 2.0460 (2.3); 2.0374 (2.0); 2.0251 (1.0); 1.6166 (16.0); 1.5992 (16.0); 1.5736 (0.5); 1.4604 (0.3); 1.4425 (0.4); 1.2453 (4.7); 1.2289 (9.2); 1.2125 (5.5); 1.2020 (3.4); 1.1885 (6.6); 1.1797 (6.2); 1.1690 (4.7); 1.1593 (1.9); 1.1474 (1.0); 1.1398 (0.7); 1.1345 (0.7); 1.1221 (1.2); 1.1163 (1.6); 1.1032 (5.5); 1.0967 (5.0); 1.0836 (5.2); 1.0781 (4.4); 1.0614 (0.9); 1.0319 (0.4); 1.0224 (0.5); 1.0044 (5.1); 0.9992 (7.1); 0.9835 (5.2); 0.9783 (7.1); 0.9618 (0.9); 0.9526 (0.7); 0.9357 (0.3); 0.9195 (1.0); 0.9072 (1.1); 0.8959 (4.2); 0.8899 (2.4); 0.8835 (4.7); 0.8779 (3.3); 0.8722 (3.4); 0.8682 (2.4); 0.8601 (2.9); 0.8452 (1.0); 0.8364 (0.7); 0.1458 (0.9); 0.0079 (6.7); -0.0002 (228.9); -0.0086 (7.8); -0.1496 (0.9)	
I-23		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8288 (2.5); 8.8244 (2.6); 8.2775 (1.7); 8.2719 (1.6); 8.2559 (2.0); 8.2504 (1.9); 8.0294 (2.8); 8.0078 (2.4); 7.6891 (0.8); 7.6703 (0.8); 7.4047 (5.4); 7.4012 (5.7); 7.1506 (2.3); 6.1584 (0.4); 6.1410 (1.3); 6.1231 (1.9); 6.1051 (1.3); 6.0877 (0.3); 3.0718 (0.5); 3.0545 (1.4); 3.0372 (1.9); 3.0198 (1.5); 3.0025 (0.6); 2.1536 (45.3); 2.0314 (0.4); 2.0187 (0.7); 2.0104 (0.8); 1.9979 (1.5); 1.9855 (0.9); 1.9769 (0.9); 1.9650 (1.4); 1.9532 (10.1); 1.9471 (18.9); 1.9409 (26.6); 1.9347 (18.5); 1.9285 (9.6); 1.6397 (8.6); 1.6224 (8.6); 1.4362 (1.4); 1.3263 (16.0); 1.3090 (15.7); 1.2646 (0.9); 1.0666 (0.6); 1.0571 (2.3); 1.0518 (2.5); 1.0431 (0.9); 1.0360 (2.4); 1.0309 (2.3); 1.0215 (0.7); 0.7793 (1.0); 0.7665 (2.4); 0.7631 (2.1); 0.7564 (2.2); 0.7533 (2.2); 0.7410 (0.8); -0.0002 (2.5)	
I-24		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4398 (1.8); 9.4219 (1.9); 9.0672 (16.0); 8.2033 (4.3); 8.1998 (3.1); 8.1872 (2.6); 8.1828 (3.9); 8.1791 (2.3); 8.0893 (2.5); 8.0852 (3.9); 8.0809 (2.2); 5.9610 (1.3); 5.9436 (2.0); 5.9260 (1.3); 3.4972 (0.6); 3.4428 (1.3); 3.4245 (4.4); 3.4062 (4.5); 3.3878 (1.4); 3.3248 (60.2); 2.6756 (0.7); 2.6714 (0.9); 2.6670 (0.7); 2.5244 (3.6); 2.5108 (56.2); 2.5068 (108.6); 2.5024 (142.5); 2.4980 (108.6); 2.3338 (0.7); 2.3293 (0.9); 2.3247 (0.7); 2.0774 (0.4); 2.0653 (0.8); 2.0566 (0.9); 2.0445 (1.7); 2.0324 (1.0); 2.0238 (0.9); 2.0116 (0.4); 1.7810 (0.5); 1.6125 (7.1); 1.5951 (7.1); 1.1284 (4.8); 1.1101 (10.6); 1.0917 (4.7); 1.0036 (0.4); 0.9902 (2.1); 0.9844 (3.2); 0.9695 (2.1); 0.9637 (3.1); 0.9527 (0.6); 0.9010 (0.3); 0.8833 (1.0); 0.8779 (1.8); 0.8719 (2.4); 0.8658 (2.8); 0.8602 (2.3); 0.0078 (1.8); -0.0001 (46.8); -0.0082 (1.9)	
I-25		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4515 (1.5); 9.4336 (1.5); 9.0706 (16.0); 9.0644 (0.6); 8.2226 (1.8); 8.2190 (3.6); 8.2153 (2.3); 8.1799 (2.0); 8.1753 (3.2); 8.1713 (2.0); 8.1172 (2.2); 8.1128 (3.3); 8.1083 (1.9); 5.9657 (1.0); 5.9482 (1.6); 5.9306 (1.0); 3.3331 (55.1); 3.0249 (0.7); 3.0172 (0.7); 3.0127 (0.4); 3.0055 (1.4); 2.9971 (0.5); 2.9934 (0.8); 2.9857 (0.7); 2.9737 (0.4); 2.5268 (0.7); 2.5220 (1.0); 2.5133 (12.7); 2.5089 (25.6); 2.5044 (33.9); 2.4998 (25.3); 2.4954 (12.7); 2.0670 (0.7); 2.0583 (0.7); 2.0462 (1.4); 2.0341 (0.8); 2.0255 (0.7); 2.0132 (0.4); 1.6151 (5.6); 1.5977 (5.6); 1.1967 (0.8); 1.1880 (1.9); 1.1847 (1.8); 1.1793 (1.6); 1.1765 (1.9); 1.1679 (1.2); 1.1353 (0.3); 1.1134 (0.5); 1.1012 (1.7); 1.0936 (1.5); 1.0840 (1.4); 1.0812 (1.6); 1.0762 (1.2); 1.0735 (1.1); 0.9917 (1.6); 0.9858 (2.6); 0.9741 (1.4); 0.9709	

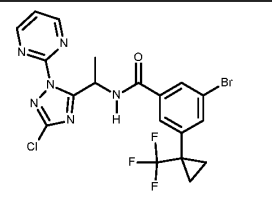
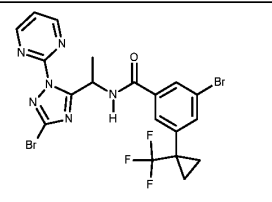
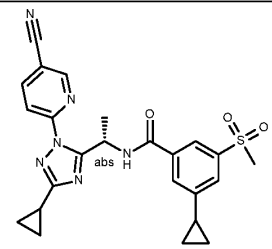
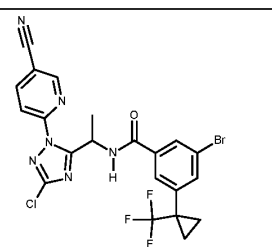
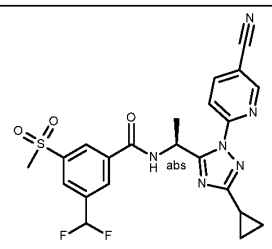
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I-26		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4719 (1.6); 9.4541 (1.7); 9.0680 (16.0); 8.2901 (2.0); 8.2867 (3.8); 8.2830 (2.7); 8.2724 (2.5); 8.2680 (3.5); 8.2642 (2.0); 8.1717 (2.3); 8.1673 (3.7); 8.1630 (2.0); 5.9708 (1.1); 5.9533 (1.8); 5.9357 (1.2); 5.7577 (3.0); 5.1413 (0.9); 5.1168 (2.8); 5.0922 (3.0); 5.0675 (1.0); 3.3300 (14.7); 2.5274 (0.6); 2.5139 (11.2); 2.5096 (21.9); 2.5051 (28.4); 2.5006 (21.2); 2.4963 (10.7); 2.0794 (0.3); 2.0671 (0.7); 2.0586 (0.8); 2.0464 (1.5); 2.0342 (0.9); 2.0257 (0.8); 2.0133 (0.4); 1.9907 (1.1); 1.6190 (6.1); 1.6016 (6.1); 1.1765 (0.6); 1.0051 (0.3); 0.9910 (1.9); 0.9856 (2.9); 0.9702 (1.9); 0.9648 (2.8); 0.9532 (0.6); 0.8849 (0.9); 0.8795 (1.6); 0.8733 (2.1); 0.8674 (2.4); 0.8616 (2.1); 0.8529 (1.0); 0.0079 (0.4); -0.0002 (11.4); -0.0085 (0.5)	
I-27		¹ H-NMR(400.2 MHz, CD ₃ CN): δ= 8.8165 (4.7); 8.8128 (4.8); 8.8111 (4.4); 8.2739 (3.4); 8.2684 (3.4); 8.2523 (4.0); 8.2468 (4.0); 8.2254 (3.4); 8.2213 (6.1); 8.2169 (4.9); 8.2073 (4.3); 8.2038 (6.5); 8.1095 (3.5); 8.1054 (5.5); 8.1012 (3.1); 8.0210 (4.9); 8.0195 (5.1); 7.9995 (4.2); 7.9979 (4.3); 7.9054 (1.5); 7.8881 (1.5); 6.7326 (3.1); 6.6011 (6.5); 6.4696 (3.3); 6.1726 (0.6); 6.1552 (2.4); 6.1372 (3.5); 6.1195 (2.4); 6.1020 (0.6); 2.1397 (41.4); 2.1140 (0.4); 2.1078 (0.5); 2.1018 (0.4); 2.0576 (0.8); 2.0452 (1.6); 2.0369 (1.7); 2.0334 (1.2); 2.0245 (2.6); 2.0212 (1.7); 2.0121 (1.6); 2.0040 (1.7); 1.9915 (1.0); 1.9721 (1.3); 1.9648 (3.1); 1.9584 (3.7); 1.9529 (28.4); 1.9467 (53.7); 1.9405 (74.7); 1.9344 (51.9); 1.9282 (26.7); 1.7752 (0.4); 1.7691 (0.5); 1.6319 (16.0); 1.6146 (15.9); 1.4363 (12.7); 1.2817 (0.4); 1.2655 (0.7); 1.2495 (0.3); 1.2323 (0.4); 1.2216 (0.4); 1.2039 (0.6); 1.1859 (0.4); 1.0357 (0.3); 1.0291 (0.4); 1.0215 (0.8); 1.0114 (3.8); 1.0064 (5.6); 0.9984 (1.2); 0.9909 (3.9); 0.9856 (6.3); 0.9714 (2.2); 0.9603 (1.0); 0.9512 (3.3); 0.9450 (2.1); 0.9388 (4.8); 0.9330 (2.7); 0.9267 (2.5); 0.9157 (1.3); 0.9087 (1.1); 0.8948 (0.4); -0.0002 (7.2)	
I-28		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4850 (2.5); 9.4678 (2.6); 8.6522 (4.6); 8.6512 (5.0); 8.6460 (4.9); 8.6448 (4.7); 8.2707 (3.7); 8.2670 (6.7); 8.2633 (4.0); 8.2327 (3.5); 8.2263 (3.2); 8.2109 (3.9); 8.2044 (3.7); 8.1643 (2.4); 8.1596 (6.5); 8.1546 (7.9); 8.1503 (6.4); 8.1455 (2.2); 7.8800 (5.0); 7.8788 (5.1); 7.8582 (4.4); 7.8570 (4.5); 5.9720 (0.4); 5.9548 (1.8); 5.9375 (2.9); 5.9202 (1.8); 5.9028 (0.4); 5.7570 (16.0); 3.3300 (145.0); 3.3212 (33.0); 3.1755 (0.8); 3.1624 (0.7); 2.6808 (0.4); 2.6764 (0.8); 2.6719 (1.1); 2.6674 (0.8); 2.5254 (4.2); 2.5119 (73.4); 2.5075 (144.2); 2.5030 (184.8); 2.4984 (132.0); 2.4940 (63.4); 2.3344 (0.8); 2.3298 (1.0); 2.3252 (0.8); 1.6403 (10.0); 1.6229 (9.9); -0.0002 (2.2)	520.0
I-29		¹ H-NMR(600.1 MHz, CD ₃ CN): δ= 8.8962 (0.9); 8.8881 (0.9); 8.1424 (0.5); 8.0510 (0.5); 8.0484 (0.7); 8.0459 (0.4); 7.5033 (0.5); 3.1005 (3.0); 2.1303 (16.0); 1.9635 (0.4); 1.9554 (0.4); 1.9511 (0.6); 1.9474 (7.9); 1.9433 (15.0); 1.9392 (22.1); 1.9350 (15.2); 1.9309 (7.7); 1.6917 (1.2); 1.6801 (1.2); -0.0001 (0.9)	

I-30		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6564 (1.9); 9.6393 (2.0); 9.0147 (9.2); 9.0025 (9.4); 8.5892 (4.0); 8.4504 (3.6); 8.3992 (3.6); 7.6832 (2.4); 7.6711 (4.6); 7.6589 (2.3); 6.0507 (1.5); 6.0335 (2.3); 6.0162 (1.5); 5.7567 (16.0); 3.3730 (21.0); 3.3348 (131.5); 3.1761 (1.0); 3.1629 (0.9); 2.6771 (0.5); 2.6727 (0.7); 2.6681 (0.5); 2.5259 (2.5); 2.5082 (92.6); 2.5038 (117.7); 2.4993 (85.6); 2.3350 (0.5); 2.3306 (0.7); 2.3260 (0.5); 1.6876 (8.2); 1.6702 (8.1); -0.0002 (1.0)	475.1
I-31		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0035 (8.1); 8.9913 (8.2); 8.9442 (1.6); 8.9261 (1.7); 8.0416 (0.4); 8.0223 (0.8); 8.0029 (0.5); 7.9274 (1.8); 7.7483 (0.6); 7.7286 (1.4); 7.6769 (2.2); 7.6647 (4.1); 7.6526 (2.4); 7.6365 (0.7); 7.5574 (0.7); 7.5380 (0.6); 7.5078 (0.9); 7.4981 (0.5); 7.4906 (0.7); 7.4839 (0.6); 7.4225 (0.4); 7.4005 (0.4); 7.3153 (0.5); 7.3044 (1.1); 7.2945 (0.8); 7.2893 (0.9); 7.2808 (1.0); 7.2676 (0.4); 7.2156 (5.1); 7.1888 (0.3); 7.1802 (0.3); 7.1752 (0.4); 7.1675 (0.4); 7.1614 (0.4); 7.1529 (0.7); 7.1456 (0.6); 7.1390 (0.8); 7.1295 (0.4); 7.0456 (1.0); 7.0392 (0.6); 7.0332 (0.4); 7.0115 (0.4); 6.9482 (0.4); 6.9290 (0.4); 5.9878 (0.4); 5.9706 (1.3); 5.9529 (2.0); 5.9352 (1.4); 5.7570 (5.6); 5.5080 (2.3); 5.0545 (0.5); 4.9534 (0.6); 4.0099 (12.9); 3.4918 (15.2); 3.3206 (0.4); 3.2876 (0.4); 3.2586 (3.7); 3.2332 (0.5); 3.1804 (0.4); 3.1696 (0.3); 3.1190 (0.4); 3.1020 (0.7); 3.0461 (4.7); 2.6771 (0.4); 2.6729 (0.5); 2.6683 (0.4); 2.5082 (75.2); 2.5038 (96.1); 2.4994 (71.3); 2.3351 (0.4); 2.3307 (0.6); 2.3261 (0.4); 1.6113 (7.4); 1.5939 (7.4); 1.4235 (0.5); 1.4094 (1.2); 1.4031 (0.7); 1.3804 (0.8); 1.3765 (0.7); 1.3532 (1.7); 1.3495 (1.6); 1.3363 (1.0); 1.3208 (1.0); 1.3131 (2.8); 1.3082 (1.7); 1.2958 (16.0); 1.2822 (2.1); 1.2743 (8.8); 1.2543 (1.2); -0.0002 (1.0)	401.4
I-32		¹ H-NMR(600.4 MHz, d ₆ -DMSO): δ= 9.5125 (3.9); 9.5013 (4.0); 9.0764 (6.0); 9.0753 (6.3); 9.0728 (6.2); 9.0716 (5.7); 8.6009 (5.2); 8.5972 (4.9); 8.5866 (5.3); 8.5829 (5.2); 8.2865 (5.6); 8.2840 (9.8); 8.2815 (5.6); 8.1960 (5.2); 8.1930 (8.0); 8.1903 (5.3); 8.1570 (5.7); 8.1541 (8.1); 8.1511 (4.4); 8.0510 (6.6); 8.0498 (6.4); 8.0367 (6.2); 8.0355 (6.1); 6.0783 (0.6); 6.0668 (3.0); 6.0553 (4.5); 6.0438 (3.0); 6.0322 (0.6); 4.0796 (0.3); 4.0708 (0.4); 3.3714 (1.1); 3.3430 (0.4); 3.3324 (2.3); 3.3216 (44.4); 3.3054 (338.1); 3.2817 (0.6); 3.1733 (1.5); 3.1645 (1.5); 2.6897 (0.5); 2.6183 (1.2); 2.6153 (2.4); 2.6123 (3.2); 2.6093 (2.3); 2.6062 (1.1); 2.5214 (8.9); 2.5183 (11.4); 2.5151 (13.2); 2.5064 (196.3); 2.5034 (390.8); 2.5003 (523.4); 2.4973 (379.6); 2.4943 (178.4); 2.3904 (1.2); 2.3873 (2.4); 2.3843 (3.2); 2.3812 (2.3); 2.3783 (1.0); 2.0719 (2.0); 1.6714 (0.4); 1.6547 (16.0); 1.6431 (15.9); 1.2424 (0.9); 1.2312 (1.2); 1.2168 (0.8); 0.0053 (2.3); -0.0001 (56.2); -0.0057 (1.8)	467.3
I-33		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0719 (3.7); 9.0702 (4.1); 9.0666 (4.1); 9.0648 (3.8); 8.9616 (2.1); 8.9438 (2.2); 8.5973 (3.0); 8.5917 (2.9); 8.5758 (3.2); 8.5703 (3.2); 8.0360 (3.9); 8.0343 (4.0); 8.0146 (3.6); 8.0128 (3.8); 7.2360 (6.3); 6.0323 (0.4); 6.0150 (1.8); 5.9975 (2.8); 5.9799 (1.8); 5.9622 (0.3); 5.7564 (13.1); 4.0229 (16.0); 3.3346 (274.5); 3.1755 (0.6); 3.1624 (0.6); 2.6766 (0.8); 2.6721 (1.0); 2.6676 (0.8); 2.5256 (3.6); 2.5207 (6.0); 2.5121 (68.0); 2.5077 (135.8); 2.5032 (176.0); 2.4986 (126.9); 2.4942 (61.7); 2.3390 (0.4); 2.3344 (0.7); 2.3301 (1.0); 2.3255 (0.8); 1.6077 (9.4); 1.5903 (9.4); -0.0001 (1.7)	425.3

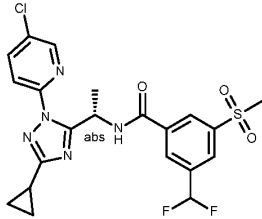
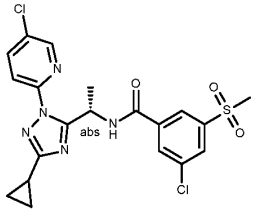
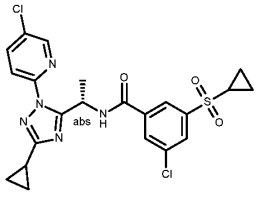
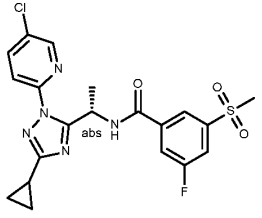
I-34		¹ H-NMR (600.1 MHz, CD ₃ CN): δ = 8.8958 (0.8); 8.8877 (0.8); 8.1416 (0.4); 8.0490 (0.5); 8.0463 (0.7); 8.0437 (0.4); 7.5029 (0.4); 3.1006 (2.7); 2.1456 (16.0); 1.9482 (2.0); 1.9441 (3.8); 1.9400 (5.5); 1.9359 (3.8); 1.9317 (1.9); 1.6882 (1.0); 1.6766 (1.0)	
I-35		¹ H-NMR(400.2 MHz, CD ₃ CN): δ = 8.8129 (4.4); 8.8113 (4.5); 8.8075 (4.6); 8.2726 (3.5); 8.2670 (3.5); 8.2510 (4.1); 8.2454 (3.9); 8.0817 (5.0); 8.0777 (3.7); 8.0341 (3.2); 8.0293 (6.2); 8.0244 (3.6); 8.0166 (5.2); 8.0149 (5.2); 8.0048 (5.0); 7.9949 (4.2); 7.9933 (4.0); 7.8333 (1.2); 7.5872 (0.6); 6.1635 (0.6); 6.1459 (2.5); 6.1282 (3.4); 6.1102 (2.4); 6.0929 (0.5); 2.4692 (0.6); 2.4646 (0.8); 2.4595 (0.5); 2.2302 (0.3); 2.1576 (192.2); 2.1552 (193.7); 2.1202 (0.9); 2.1141 (0.9); 2.1077 (1.3); 2.1017 (0.9); 2.0957 (0.4); 2.0579 (0.8); 2.0458 (1.7); 2.0375 (1.7); 2.0340 (1.2); 2.0251 (2.6); 2.0214 (1.6); 2.0128 (1.7); 2.0045 (1.8); 1.9923 (1.1); 1.9721 (1.4); 1.9648 (9.4); 1.9586 (10.8); 1.9529 (86.8); 1.9467 (161.3); 1.9406 (224.3); 1.9344 (154.3); 1.9282 (79.0); 1.9156 (1.1); 1.7815 (0.5); 1.7751 (0.9); 1.7691 (1.3); 1.7629 (0.9); 1.7566 (0.5); 1.6284 (16.0); 1.6111 (16.0); 1.4369 (12.4); 1.2715 (0.4); 1.2605 (0.6); 1.2436 (0.4); 1.2253 (0.4); 1.2039 (0.4); 1.0342 (0.4); 1.0212 (0.9); 1.0117 (3.2); 1.0064 (5.9); 0.9986 (1.2); 0.9907 (3.0); 0.9855 (6.7); 0.9717 (1.5); 0.9680 (2.0); 0.9606 (1.0); 0.9555 (1.0); 0.9501 (3.4); 0.9444 (1.6); 0.9381 (3.3); 0.9320 (2.7); 0.9278 (1.9); 0.9240 (1.8); 0.9197 (2.0); 0.9155 (1.6); 0.9109 (1.2); 0.9020 (1.0); 0.8977 (0.8); 0.8886 (0.5); 0.0077 (0.6); -0.0002 (15.5); -0.0082 (0.6)	
I-36		¹ H-NMR(400.2 MHz, CD ₃ CN): δ = 8.8245 (4.4); 8.8229 (4.7); 8.8191 (4.8); 8.8174 (4.4); 8.2716 (3.5); 8.2660 (3.4); 8.2500 (4.1); 8.2444 (4.0); 8.0136 (4.9); 8.0120 (4.9); 7.9920 (4.2); 7.9904 (4.2); 7.7730 (1.3); 7.7552 (1.4); 7.6841 (3.9); 7.6801 (6.2); 7.6757 (4.2); 7.5989 (4.2); 7.5949 (6.9); 7.5909 (4.1); 7.4670 (4.1); 7.4624 (6.9); 7.4578 (3.6); 6.1503 (0.6); 6.1329 (2.4); 6.1151 (3.3); 6.0970 (2.4); 6.0796 (0.6); 5.4481 (5.7); 2.1716 (38.6); 2.0873 (0.7); 2.0593 (0.7); 2.0469 (1.5); 2.0387 (1.7); 2.0351 (1.1); 2.0262 (2.6); 2.0224 (1.8); 2.0139 (1.6); 2.0058 (1.7); 1.9933 (0.9); 1.9657 (1.8); 1.9595 (1.9); 1.9538 (15.7); 1.9477 (29.3); 1.9415 (40.8); 1.9353 (28.3); 1.9291 (14.6); 1.9170 (0.4); 1.7767 (2.6); 1.7637 (7.6); 1.7570 (7.7); 1.7444 (3.2); 1.7052 (0.4); 1.6170 (16.0); 1.5996 (16.0); 1.5657 (0.4); 1.5273 (2.9); 1.5140 (7.5); 1.5074 (7.5); 1.4944 (2.2); 1.2669 (0.4); 1.0285 (0.4); 1.0215 (0.6); 1.0104 (3.2); 1.0048 (6.1); 0.9996 (1.4); 0.9897 (3.8); 0.9839 (6.5); 0.9770 (1.3); 0.9698 (2.2); 0.9635 (1.1); 0.9543 (3.7); 0.9479 (2.0); 0.9420 (3.6); 0.9351 (2.5); 0.9301 (2.1); 0.9276 (2.0); 0.9226 (1.7); 0.9179 (1.3); 0.9112 (0.6); 0.9077 (0.9); 0.8956 (0.4); -0.0002 (3.3)	
I-37		¹ H-NMR(400.2 MHz, CD ₃ CN): δ = 8.8147 (4.3); 8.8105 (4.4); 8.2690 (3.0); 8.2636 (3.0); 8.2475 (3.5); 8.2420 (3.5); 8.0090 (4.9); 7.9888 (4.0); 7.9874 (4.1); 7.6695 (1.2); 7.6519 (1.2); 7.3991 (8.2); 7.3957 (9.0); 7.1520 (3.8); 6.1389 (0.6); 6.1217 (2.2); 6.1038 (3.2); 6.0859 (2.2); 6.0681 (0.6); 3.8007 (0.8); 2.4698 (0.4); 2.4652 (0.6); 2.4600 (0.4); 2.1694 (115.6); 2.1206 (0.4); 2.1143 (0.5); 2.1080 (0.7); 2.1019 (0.5); 2.0580 (0.8); 2.0457 (1.6); 2.0375 (1.7); 2.0336 (1.4); 2.0251 (2.6); 2.0205 (2.5); 2.0123 (2.6); 2.0048 (2.2); 1.9986 (2.8); 1.9919 (1.4); 1.9861 (1.5); 1.9775 (1.5); 1.9651 (5.1); 1.9588 (5.1); 1.9531 (42.1); 1.9470 (79.2); 1.9408 (110.4); 1.9346 (76.0); 1.9285 (39.1); 1.9156 (0.8); 1.7752 (0.5); 1.7693 (0.6); 1.7632 (0.4); 1.6153 (16.0); 1.5980 (15.9); 1.4365 (2.7); 1.0679 (1.0); 1.0584 (4.2); 1.0532 (4.4); 1.0421 (1.6); 1.0373 (4.5); 1.0323 (4.3); 1.0210 (1.8); 1.0144 (1.5); 1.0103 (2.8); 1.0047 (6.1); 0.9984 (1.3); 0.9938 (1.4);	

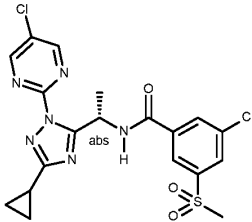
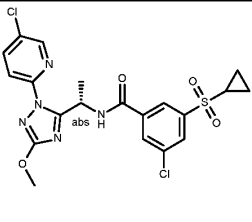
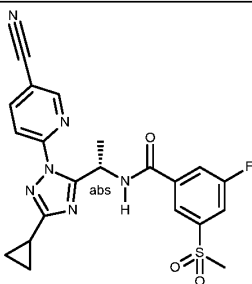
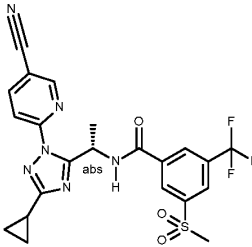
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I-38		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6537 (1.7); 9.6364 (1.7); 9.0145 (2.7); 9.0110 (7.8); 9.0024 (2.8); 8.9988 (7.9); 8.5879 (3.5); 8.4487 (3.2); 8.3984 (3.2); 7.6832 (0.6); 7.6789 (2.1); 7.6710 (1.3); 7.6667 (3.9); 7.6590 (0.7); 7.6546 (2.0); 6.0547 (1.1); 6.0373 (1.8); 6.0200 (1.2); 5.7570 (16.0); 3.3725 (18.3); 3.3310 (96.4); 3.1759 (0.6); 3.1628 (0.6); 2.6767 (0.5); 2.6724 (0.7); 2.6680 (0.5); 2.5255 (2.4); 2.5079 (95.0); 2.5035 (119.5); 2.4990 (88.0); 2.3347 (0.5); 2.3303 (0.7); 2.3261 (0.5); 1.6819 (6.1); 1.6645 (6.1); -0.0001 (1.2)	520.0
I-39		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 10.4070 (0.5); 9.0014 (7.2); 8.9893 (7.3); 8.9347 (1.4); 8.9164 (1.3); 7.6741 (1.8); 7.6619 (3.5); 7.6497 (1.8); 7.3521 (0.6); 7.2139 (4.8); 5.9737 (1.1); 5.9560 (1.7); 5.9382 (1.1); 5.7566 (16.0); 4.0677 (1.6); 4.0350 (0.3); 4.0106 (12.4); 3.3332 (113.6); 3.1757 (0.4); 3.1626 (0.4); 2.6769 (0.5); 2.6723 (0.7); 2.6680 (0.5); 2.5255 (2.9); 2.5120 (49.6); 2.5078 (95.7); 2.5033 (121.9); 2.4988 (88.6); 2.4946 (44.0); 2.3346 (0.5); 2.3301 (0.7); 2.3257 (0.5); 1.6045 (6.0); 1.5928 (2.4); 1.5872 (5.9); -0.0002 (1.3)	446.0
I-40		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6282 (4.1); 9.6108 (4.1); 9.0056 (7.2); 9.0017 (7.2); 9.0002 (6.8); 8.5610 (0.4); 8.5342 (5.3); 8.5286 (5.2); 8.5126 (5.7); 8.5071 (6.0); 8.5004 (5.5); 8.4961 (8.9); 8.4920 (6.2); 8.4255 (8.4); 8.4094 (5.4); 8.4054 (7.3); 8.3792 (0.4); 8.3168 (0.7); 8.0152 (7.1); 8.0139 (7.2); 7.9937 (6.6); 7.9923 (6.7); 6.1050 (0.6); 6.0877 (2.9); 6.0703 (4.6); 6.0529 (3.0); 6.0351 (0.6); 4.4139 (0.6); 4.3960 (0.7); 3.3283 (281.8); 2.6764 (1.7); 2.6719 (2.4); 2.6675 (1.7); 2.5252 (6.9); 2.5117 (143.2); 2.5074 (284.7); 2.5029 (371.1); 2.4984 (269.7); 2.4940 (132.4); 2.3343 (1.7); 2.3298 (2.3); 2.3252 (1.7); 2.0927 (0.8); 2.0803 (1.9); 2.0716 (2.1); 2.0597 (3.8); 2.0476 (2.2); 2.0390 (2.0); 2.0267 (1.0); 1.6233 (16.0); 1.6059 (16.0); 1.3789 (0.7); 1.3611 (1.5); 1.3434 (0.7); 1.2483 (0.8); 1.2312 (1.5); 1.2140 (0.8); 1.0342 (0.3); 1.0254 (0.4); 1.0055 (5.0); 1.0008 (6.6); 0.9844 (4.8); 0.9799 (6.8); 0.9617 (0.9); 0.9535 (0.7); 0.9183 (1.0); 0.9065 (1.1); 0.8947 (4.0); 0.8890 (2.3); 0.8824 (4.4); 0.8768 (3.7); 0.8694 (3.2); 0.8649 (2.4); 0.8572 (2.8); 0.8458 (0.9); 0.8424 (1.1); 0.8337 (0.7); 0.1460 (0.5); 0.0079 (4.2); -0.0002 (114.3); -0.0085 (4.3); -0.1496 (0.5)	
I-41		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4827 (2.8); 9.4650 (2.8); 8.9925 (14.2); 8.9803 (14.5); 8.3151 (0.3); 8.2480 (4.1); 8.2443 (7.2); 8.2405 (4.3); 8.1545 (2.8); 8.1498 (6.8); 8.1458 (6.7); 8.1435 (7.2); 8.1394 (6.8); 8.1347 (2.6); 7.6571 (3.7); 7.6450 (7.1); 7.6328 (3.7); 6.0289 (0.4); 6.0118 (2.0); 5.9943 (3.1); 5.9768 (2.0); 5.9593 (0.4); 5.7560 (16.0); 3.3395 (283.1); 3.3156 (32.6); 3.2861 (0.9); 3.1692 (2.2); 2.6770 (0.9); 2.6724 (1.2); 2.6680 (0.9); 2.5259 (4.5); 2.5123 (86.2); 2.5080 (167.7); 2.5035 (213.7); 2.4990 (155.4); 2.4948 (77.4); 2.3348 (0.9); 2.3303 (1.2); 2.3259 (0.9); 1.6403 (10.6); 1.6230 (10.6); -0.0002 (1.5)	532.9

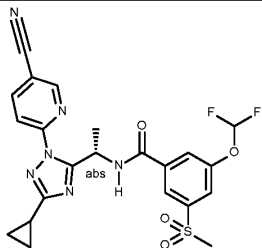
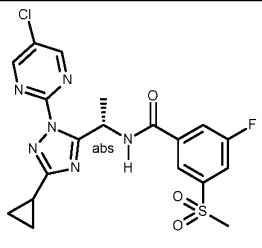
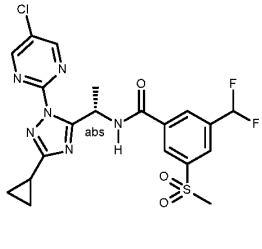
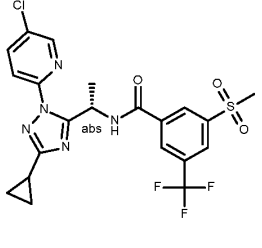
I-42		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0031 (0.5); 8.9909 (12.2); 8.9787 (12.1); 8.8887 (2.1); 8.8702 (2.1); 7.6566 (3.1); 7.6445 (5.9); 7.6323 (3.1); 7.2128 (6.3); 5.9881 (0.4); 5.9710 (1.7); 5.9532 (2.5); 5.9352 (1.7); 5.9180 (0.4); 5.7570 (13.0); 4.0119 (16.0); 3.3316 (120.1); 3.1757 (0.6); 3.1625 (0.5); 2.6767 (0.6); 2.6722 (0.8); 2.6675 (0.6); 2.5256 (2.7); 2.5121 (55.0); 2.5077 (107.4); 2.5032 (136.6); 2.4987 (97.9); 2.4943 (47.4); 2.3346 (0.6); 2.3300 (0.8); 2.3254 (0.6); 1.6106 (0.4); 1.5842 (8.8); 1.5668 (8.8); -0.0002 (1.7)	493.0
I-43		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1735 (1.9); 9.1558 (1.9); 8.9993 (0.4); 8.9802 (9.1); 8.9681 (9.3); 7.6458 (2.4); 7.6336 (4.5); 7.6215 (2.3); 7.4320 (6.8); 7.4286 (6.8); 7.2715 (3.0); 5.9581 (1.4); 5.9404 (2.2); 5.9229 (1.4); 5.7570 (16.0); 3.3325 (98.4); 3.1762 (0.7); 3.1631 (0.7); 2.6769 (0.6); 2.6725 (0.8); 2.6682 (0.6); 2.5258 (2.8); 2.5080 (99.7); 2.5036 (125.9); 2.4991 (92.0); 2.3347 (0.5); 2.3303 (0.7); 2.3261 (0.5); 2.0572 (0.4); 2.0444 (0.8); 2.0358 (0.9); 2.0235 (1.6); 2.0111 (1.0); 2.0027 (0.9); 1.9904 (0.4); 1.6495 (0.4); 1.6256 (7.4); 1.6082 (7.3); 1.0431 (1.0); 1.0324 (2.6); 1.0269 (2.9); 1.0165 (1.7); 1.0116 (2.7); 1.0060 (2.7); 0.9959 (1.1); 0.7839 (1.2); 0.7733 (3.2); 0.7684 (3.1); 0.7611 (3.0); 0.7562 (3.1); 0.7450 (0.9); -0.0002 (1.4)	545.0
I-44		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2008 (1.3); 9.1835 (1.3); 9.0098 (2.4); 9.0047 (2.4); 8.5423 (1.6); 8.5367 (1.5); 8.5207 (1.7); 8.5152 (1.7); 7.9760 (2.4); 7.9543 (2.2); 7.5214 (2.9); 7.5040 (2.0); 7.2840 (2.0); 6.0521 (1.0); 6.0348 (1.6); 6.0174 (1.0); 3.9618 (16.0); 3.3312 (19.3); 2.5270 (0.6); 2.5131 (13.4); 2.5090 (26.8); 2.5045 (35.0); 2.5000 (25.6); 2.4958 (12.8); 2.0632 (0.6); 2.0548 (0.6); 2.0425 (1.2); 2.0300 (0.7); 2.0216 (0.6); 1.6181 (5.4); 1.6007 (5.4); 1.0528 (0.6); 1.0416 (1.9); 1.0361 (2.1); 1.0256 (1.0); 1.0207 (2.0); 1.0152 (2.0); 1.0050 (0.8); 0.8033 (0.8); 0.7927 (2.2); 0.7877 (2.2); 0.7805 (2.1); 0.7755 (2.3); 0.7641 (0.7); -0.0002 (0.9)	
I-45		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4571 (1.1); 9.4397 (1.2); 8.6060 (2.2); 8.6050 (2.2); 8.5997 (2.3); 8.3013 (1.7); 8.2976 (3.1); 8.2938 (1.9); 8.1994 (1.6); 8.1948 (2.7); 8.1905 (3.3); 8.1838 (1.6); 8.1684 (1.8); 8.1618 (1.8); 8.1512 (1.9); 8.1469 (2.8); 8.1424 (1.6); 7.8283 (2.4); 7.8272 (2.4); 7.8065 (2.2); 7.8053 (2.2); 5.9950 (0.9); 5.9776 (1.4); 5.9602 (0.9); 3.9427 (16.0); 3.3288 (19.3); 3.3245 (15.0); 2.5261 (0.7); 2.5213 (1.1); 2.5127 (14.2); 2.5082 (29.2); 2.5037 (38.3); 2.4991 (27.5); 2.4946 (13.2); 2.0763 (2.7); 1.6198 (4.9); 1.6023 (4.9); -0.0002 (1.3)	
I-46		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1465 (1.2); 9.1290 (1.2); 8.5848 (2.3); 8.5835 (2.3); 8.5784 (2.4); 8.5770 (2.2); 8.1787 (1.7); 8.1722 (1.6); 8.1568 (1.9); 8.1503 (1.8); 7.8141 (2.5); 7.8128 (2.4); 7.7922 (2.2); 7.7908 (2.1); 7.4852 (3.8); 7.4819 (3.7); 7.2777 (1.8); 5.9381 (0.9); 5.9206 (1.4); 5.9032 (0.9); 3.9367 (16.0); 3.3323 (68.0); 2.6725 (0.4); 2.5260 (1.1); 2.5211 (1.7); 2.5125 (23.3); 2.5081 (46.4); 2.5036 (59.4); 2.4990 (42.1); 2.4945 (20.0); 2.3304 (0.3); 2.0558 (0.5); 2.0472 (0.6); 2.0348 (1.1); 2.0223 (0.6); 2.0139 (0.6); 1.6064 (5.0); 1.5889 (5.0); 1.0491 (0.6); 1.0379 (1.8); 1.0324 (2.0); 1.0218 (1.0); 1.0170 (1.8); 1.0114 (1.8); 1.0011 (0.7); 0.7974 (0.8); 0.7867 (2.1); 0.7815 (2.0); 0.7744 (2.0); 0.7693 (2.2); 0.7577 (0.6); -0.0002 (1.7)	

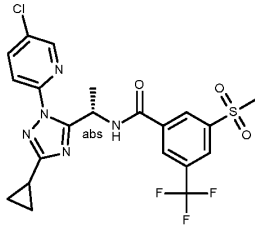
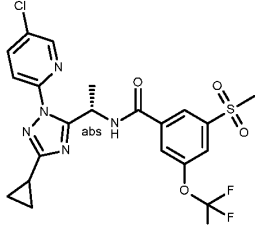
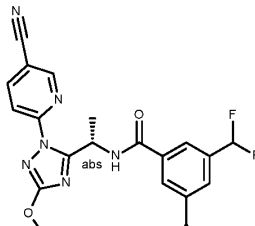
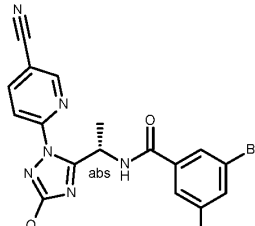
I-47			515.0
I-48			561.00
I-49		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2891 (1.4); 9.2714 (1.4); 9.0184 (2.4); 9.0169 (2.5); 9.0131 (2.6); 8.5352 (1.9); 8.5296 (1.8); 8.5136 (2.0); 8.5081 (2.0); 8.0723 (2.0); 8.0684 (3.4); 8.0645 (2.0); 8.0098 (2.6); 8.0082 (2.5); 7.9883 (2.4); 7.9867 (2.3); 7.7828 (1.8); 7.7788 (3.2); 7.7748 (1.9); 7.7337 (2.0); 7.7300 (3.1); 7.7262 (1.7); 6.0652 (1.0); 6.0476 (1.6); 6.0301 (1.0); 3.3286 (114.7); 3.2798 (0.8); 3.2341 (16.0); 2.6760 (0.7); 2.6714 (0.9); 2.6669 (0.7); 2.5249 (3.1); 2.5114 (59.6); 2.5070 (117.8); 2.5025 (152.1); 2.4979 (109.2); 2.4934 (52.4); 2.3338 (0.7); 2.3293 (0.9); 2.3247 (0.7); 2.1421 (0.6); 2.1339 (0.7); 2.1213 (1.2); 2.1089 (0.7); 2.1004 (0.7); 2.0866 (0.6); 2.0740 (0.8); 2.0650 (0.7); 2.0531 (1.4); 2.0409 (0.8); 2.0324 (0.7); 2.0199 (0.4); 1.6145 (5.6); 1.5970 (5.6); 1.3462 (0.5); 1.0877 (0.8); 1.0764 (2.1); 1.0710 (2.2); 1.0603 (1.2); 1.0555 (2.1); 1.0500 (2.1); 1.0396 (0.9); 1.0003 (1.8); 0.9948 (2.6); 0.9795 (1.7); 0.9740 (2.6); 0.9149 (0.4); 0.9021 (0.4); 0.8910 (1.4); 0.8856 (0.8); 0.8788 (1.5); 0.8728 (1.3); 0.8663 (1.2); 0.8604 (0.9); 0.8546 (1.2); 0.8401 (1.3); 0.8292 (2.7); 0.8244 (2.5); 0.8171 (2.3); 0.8121 (2.6); 0.8004 (0.7); 0.0080 (1.7); -0.0002 (46.4); -0.0084 (1.7)	
I-50			541.0
I-51		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5422 (3.8); 9.5247 (3.9); 9.0233 (6.6); 9.0180 (7.2); 8.5377 (3.8); 8.5330 (3.9); 8.5159 (5.7); 8.5103 (9.6); 8.4126 (0.8); 8.3786 (0.5); 8.3363 (6.8); 8.3166 (0.5); 8.2641 (6.9); 8.2075 (0.9); 8.1698 (1.4); 8.0510 (0.6); 8.0199 (6.8); 7.9983 (6.3); 7.9345 (0.5); 7.9032 (1.0); 7.3802 (0.5); 7.3698 (2.8); 7.3545 (0.6); 7.2425 (1.0); 7.2317 (6.1); 7.2166 (1.2); 7.1043 (0.5); 7.0937 (3.0); 7.0788 (0.6); 6.1956 (0.4); 6.1652 (0.4); 6.1256 (0.6); 6.1081 (2.5); 6.0907 (3.9); 6.0733 (2.5); 6.0556 (0.6); 5.7576 (1.9); 5.3245 (0.4); 3.8101 (0.4); 3.7937 (0.4); 3.4480 (0.3); 3.4313 (0.4); 3.3922 (0.5); 3.3757 (1.0); 3.3585 (7.6); 3.3494 (6.8); 3.3307 (290.0); 3.1531 (0.3); 2.6794 (2.6); 2.6726 (2.2); 2.5073 (249.5); 2.5036 (326.1); 2.4998 (265.5); 2.3304 (2.0); 2.0902 (0.7); 2.0781 (1.6); 2.0691 (1.8); 2.0574 (3.2); 2.0456 (2.0); 2.0367 (1.8); 2.0255 (1.2); 2.0095 (1.3); 1.9903 (1.5); 1.9724 (0.5); 1.6303 (13.9); 1.6129 (13.9); 1.4569 (0.6); 1.4087 (0.8); 1.3716 (0.6); 1.3422 (0.4); 1.2984 (1.2); 1.2518 (6.5); 1.2352 (16.0); 1.2136 (3.4); 1.1968 (3.3); 1.1801 (1.1); 1.1418 (1.5); 1.0307 (0.5); 1.0209 (0.6); 0.9974 (6.4); 0.9764 (6.5); 0.9593 (1.0); 0.9492 (0.9); 0.9355 (0.4);	

		0.9186 (1.0); 0.9062 (1.2); 0.8947 (3.6); 0.8894 (2.8); 0.8821 (4.3); 0.8713 (4.4); 0.8558 (3.7); 0.8445 (1.4); 0.8363 (1.5); 0.1460 (0.7); 0.0011 (136.3); -0.0002 (137.6); -0.1492 (0.7)	
I-52		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6286 (1.2); 9.6114 (1.3); 8.5897 (2.3); 8.5885 (2.4); 8.5832 (2.5); 8.5821 (2.3); 8.5135 (1.8); 8.5092 (2.8); 8.5051 (2.0); 8.4545 (2.6); 8.4093 (2.3); 8.1842 (1.7); 8.1777 (1.6); 8.1623 (1.9); 8.1557 (1.9); 7.8252 (2.5); 7.8240 (2.5); 7.8033 (2.3); 7.8020 (2.2); 6.0056 (0.9); 5.9883 (1.5); 5.9709 (1.0); 5.7579 (1.4); 3.9457 (16.0); 3.3300 (23.2); 2.5271 (0.8); 2.5135 (17.2); 2.5092 (34.0); 2.5047 (43.5); 2.5001 (31.2); 2.4957 (15.1); 1.6303 (5.2); 1.6129 (5.1); -0.0002 (0.6)	
I-53		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5811 (1.2); 9.5639 (1.3); 8.5943 (2.4); 8.5882 (2.5); 8.4272 (1.7); 8.4232 (2.8); 8.4190 (1.9); 8.3439 (1.8); 8.3404 (3.0); 8.3370 (1.8); 8.1851 (2.5); 8.1794 (4.2); 8.1754 (1.8); 8.1638 (1.8); 8.1573 (1.8); 7.8269 (2.6); 7.8050 (2.3); 7.5440 (1.0); 7.4140 (2.2); 7.2842 (1.1); 5.9992 (1.0); 5.9818 (1.5); 5.9645 (1.0); 5.7581 (2.9); 3.9446 (16.0); 3.3315 (20.6); 2.5273 (0.6); 2.5136 (14.0); 2.5094 (28.0); 2.5049 (36.3); 2.5004 (26.4); 2.4961 (13.1); 1.6248 (5.2); 1.6074 (5.2); -0.0002 (0.5)	
I-54		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6763 (1.3); 9.6593 (1.3); 9.0163 (2.2); 9.0146 (2.4); 9.0108 (2.4); 9.0092 (2.2); 8.5481 (1.8); 8.5425 (2.1); 8.5397 (2.0); 8.5354 (2.9); 8.5311 (2.2); 8.5265 (2.0); 8.5210 (1.9); 8.4748 (2.6); 8.4165 (2.3); 7.9870 (2.3); 7.9854 (2.3); 7.9655 (2.2); 7.9637 (2.2); 6.1151 (1.0); 6.0979 (1.5); 6.0806 (1.0); 5.7585 (2.0); 3.9707 (16.0); 3.3310 (13.2); 2.5282 (0.7); 2.5147 (12.2); 2.5104 (23.8); 2.5059 (30.4); 2.5013 (21.8); 2.4969 (10.4); 1.6417 (5.2); 1.6243 (5.1); -0.0002 (0.4)	
I-55		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6276 (1.3); 9.6106 (1.4); 9.0166 (2.4); 9.0124 (2.4); 8.5473 (1.6); 8.5418 (1.6); 8.5257 (1.7); 8.5202 (1.7); 8.4545 (1.8); 8.4505 (2.9); 8.4464 (1.9); 8.3552 (3.1); 8.1905 (1.8); 8.1863 (2.8); 8.1822 (1.6); 7.9863 (2.4); 7.9646 (2.2); 7.5465 (1.0); 7.4165 (2.3); 7.2866 (1.2); 6.1073 (1.0); 6.0900 (1.5); 6.0728 (1.0); 5.7573 (3.5); 3.9684 (16.0); 3.3302 (23.0); 2.5128 (17.6); 2.5088 (33.8); 2.5044 (43.0); 2.4999 (31.2); 1.6348 (5.4); 1.6174 (5.4); -0.0002 (0.6)	
I-56		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.3066 (4.4); 9.2890 (4.5); 9.0150 (7.7); 9.0110 (7.6); 8.5357 (5.0); 8.5302 (4.7); 8.5142 (5.3); 8.5087 (5.2); 8.3161 (0.4); 8.0278 (9.7); 8.0116 (7.4); 7.9900 (6.8); 7.7643 (9.2); 7.7174 (8.8); 6.0815 (0.7); 6.0641 (3.0); 6.0467 (4.6); 6.0292 (2.9); 6.0114 (0.6); 3.3271 (72.6); 2.9319 (0.9); 2.9199 (1.9); 2.9122 (2.1); 2.9006 (3.7); 2.8885 (2.2); 2.8808 (2.0); 2.8687 (0.9); 2.6761 (1.1); 2.6717 (1.4); 2.6672 (1.1); 2.5549 (0.5); 2.5071 (175.7); 2.5027 (223.0); 2.4984 (167.5); 2.3339 (1.0); 2.3296 (1.4); 2.3252 (1.0); 2.1673 (0.9); 2.1548 (1.8); 2.1464 (2.0); 2.1340 (3.5); 2.1216 (2.2); 2.1131 (1.8); 2.1006 (0.9); 2.0871 (0.9); 2.0753 (7.5); 2.0666 (2.2); 2.0545 (3.7); 2.0424 (2.3); 2.0339 (2.0); 2.0216 (1.0); 1.6163 (16.0); 1.5989 (15.9); 1.2311 (0.9); 1.2184 (0.8); 1.1730 (0.6); 1.1613 (0.9); 1.1513 (3.0); 1.1386 (5.9); 1.1291 (6.0); 1.1190 (4.1); 1.0913 (2.7); 1.0797 (6.7); 1.0743 (7.9); 1.0584 (11.7); 1.0536 (10.5); 1.0385 (6.3); 1.0324 (4.6); 1.0297 (4.6); 1.0165 (1.2); 1.0012 (5.5); 0.9956 (7.2); 0.9805 (5.1); 0.9748 (7.4); 0.9551 (0.9); 0.9494 (0.8); 0.9323 (0.4); 0.9167 (1.2); 0.9041 (1.5); 0.8922 (3.8); 0.8803 (3.9); 0.8714 (2.9); 0.8659 (3.5); 0.8590 (2.8); 0.8538 (3.4); 0.8376 (3.9); 0.8265 (8.2); 0.8222 (7.6); 0.8146 (6.9); 0.8098 (7.5); 0.7980 (2.0); -0.0002 (2.1)	

I-57		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4943 (4.0); 9.4763 (4.1); 8.5931 (7.4); 8.5871 (7.6); 8.4866 (7.3); 8.3151 (7.5); 8.2549 (7.2); 8.1708 (4.8); 8.1643 (4.5); 8.1489 (5.4); 8.1424 (5.2); 7.8396 (8.0); 7.8177 (7.0); 7.3635 (2.9); 7.2253 (6.7); 7.0873 (3.2); 6.0153 (0.6); 5.9978 (2.9); 5.9802 (4.6); 5.9627 (2.9); 5.9454 (0.6); 3.3711 (0.4); 3.3270 (147.8); 3.3215 (52.3); 2.6761 (1.2); 2.6717 (1.5); 2.6673 (1.1); 2.5072 (189.7); 2.5027 (241.6); 2.4983 (178.1); 2.3339 (1.1); 2.3296 (1.5); 2.3252 (1.1); 2.0748 (0.4); 2.0695 (0.8); 2.0572 (1.9); 2.0486 (2.1); 2.0364 (3.8); 2.0243 (2.2); 2.0158 (2.0); 2.0035 (1.0); 1.6177 (16.0); 1.6003 (15.9); 1.0081 (0.4); 0.9964 (0.7); 0.9792 (6.0); 0.9749 (6.9); 0.9586 (6.0); 0.9539 (6.5); 0.9396 (1.1); 0.9287 (0.9); 0.9153 (0.4); 0.9006 (1.0); 0.8879 (1.2); 0.8765 (3.6); 0.8646 (5.6); 0.8587 (4.3); 0.8488 (2.4); 0.8431 (2.9); 0.8305 (0.9); 0.8209 (0.6); 0.1459 (0.4); 0.0078 (3.8); -0.0002 (82.6); -0.0083 (3.7); -0.1494 (0.4)
I-58		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4095 (4.1); 9.3918 (4.2); 8.5934 (7.7); 8.5870 (7.8); 8.3161 (0.5); 8.2607 (9.6); 8.1727 (4.5); 8.1662 (4.6); 8.1506 (11.4); 8.1445 (6.1); 8.1393 (6.9); 8.1352 (8.8); 7.8347 (8.2); 7.8128 (7.2); 5.9882 (0.7); 5.9714 (2.9); 5.9539 (4.6); 5.9363 (2.9); 5.9186 (0.6); 5.7565 (1.4); 3.4004 (0.6); 3.3915 (0.7); 3.3841 (0.6); 3.3678 (0.6); 3.3256 (114.0); 3.3188 (47.1); 3.1411 (0.4); 2.6754 (1.3); 2.6714 (1.7); 2.6672 (1.3); 2.5068 (206.2); 2.5025 (269.3); 2.4982 (204.1); 2.3334 (1.2); 2.3292 (1.6); 2.3250 (1.2); 2.0684 (0.8); 2.0560 (1.8); 2.0471 (2.0); 2.0352 (3.7); 2.0232 (2.2); 2.0145 (1.9); 2.0023 (1.0); 1.6029 (16.0); 1.5855 (15.9); 1.3652 (0.5); 1.3494 (0.4); 1.0086 (0.4); 0.9962 (0.8); 0.9804 (5.6); 0.9759 (7.0); 0.9597 (5.6); 0.9550 (6.5); 0.9420 (1.2); 0.9301 (0.7); 0.9148 (0.5); 0.8993 (1.0); 0.8871 (1.2); 0.8756 (3.7); 0.8631 (5.4); 0.8571 (4.6); 0.8495 (2.5); 0.8437 (2.7); 0.8321 (0.9); 0.8213 (0.6); 0.1458 (0.4); 0.0075 (3.7); -0.0002 (82.8); -0.1498 (0.4)
I-59		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4243 (4.1); 9.4064 (4.2); 8.5878 (7.8); 8.5814 (7.7); 8.3159 (0.7); 8.2213 (9.7); 8.2179 (6.5); 8.1684 (7.4); 8.1635 (13.0); 8.1484 (5.2); 8.1420 (5.0); 8.1131 (5.8); 8.1090 (8.8); 8.1050 (4.9); 7.8341 (8.3); 7.8122 (7.4); 5.9861 (0.7); 5.9690 (2.9); 5.9515 (4.6); 5.9340 (2.9); 5.9162 (0.6); 3.3264 (259.1); 3.0352 (0.9); 3.0232 (1.9); 3.0156 (2.0); 3.0039 (3.7); 2.9920 (2.1); 2.9842 (2.0); 2.9721 (0.9); 2.6754 (1.8); 2.6712 (2.4); 2.6672 (1.9); 2.5066 (285.2); 2.5023 (371.8); 2.4980 (280.8); 2.3335 (1.7); 2.3292 (2.3); 2.3248 (1.7); 2.0695 (0.8); 2.0575 (1.8); 2.0487 (2.0); 2.0368 (3.7); 2.0247 (2.2); 2.0160 (2.0); 2.0037 (1.0); 1.6043 (16.0); 1.5869 (15.9); 1.5615 (0.4); 1.2448 (0.3); 1.2285 (0.6); 1.2189 (0.7); 1.1955 (2.8); 1.1855 (5.9); 1.1745 (5.7); 1.1657 (3.8); 1.1446 (1.0); 1.1330 (0.9); 1.1197 (1.2); 1.1129 (1.8); 1.1006 (5.4); 1.0930 (4.7); 1.0806 (5.2); 1.0756 (3.9); 1.0595 (0.9); 1.0093 (0.4); 0.9980 (0.8); 0.9811 (5.8); 0.9768 (7.1); 0.9606 (6.0); 0.9559 (6.6); 0.9418 (1.2); 0.9310 (0.8); 0.9148 (0.5); 0.9007 (1.0); 0.8884 (1.3); 0.8772 (3.8); 0.8650 (5.8); 0.8594 (4.4); 0.8487 (2.4); 0.8435 (3.1); 0.8319 (0.9); 0.8205 (0.7); 0.1457 (0.6); 0.0074 (5.4); -0.0004 (112.9); -0.0083 (5.0); -0.1495 (0.6)
I-60		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.3898 (4.4); 9.3721 (4.5); 8.5942 (8.1); 8.5930 (6.9); 8.5878 (8.3); 8.3164 (0.4); 8.1948 (9.6); 8.1721 (5.2); 8.1656 (4.8); 8.1502 (5.6); 8.1437 (5.3); 7.9711 (4.1); 7.9676 (4.0); 7.9575 (6.3); 7.9538 (6.0); 7.9373 (4.2); 7.9341 (4.1); 7.8366 (8.3); 7.8147 (7.6); 5.9936 (0.7); 5.9760 (2.9); 5.9585 (4.5); 5.9410 (3.0); 5.9236 (0.8); 5.7566 (1.2); 3.3290 (192.5); 3.3080 (46.9); 2.6760 (1.3); 2.6717 (1.6); 2.5071 (210.8); 2.5028 (254.9); 2.4992 (191.4); 2.3338 (1.3); 2.3295 (1.6); 2.0684 (0.8); 2.0558 (1.9); 2.0470 (2.2); 2.0351 (3.8); 2.0230 (2.4); 2.0145 (2.1); 2.0023 (1.1); 1.6063 (15.9); 1.5889 (16.0); 1.0082 (0.4); 0.9955 (0.9); 0.9801 (6.2); 0.9757 (7.0); 0.9595 (6.4); 0.9550 (6.6); 0.9419 (1.7); 0.9300 (1.0); 0.9146 (0.6);

		0.8995 (1.1); 0.8750 (4.0); 0.8627 (6.0); 0.8568 (5.4); 0.8442 (3.2); 0.8220 (0.9); 0.1465 (0.3); 0.0012 (56.6); -0.0002 (72.4); -0.0085 (5.5); -0.1497 (0.3)	
I-61		¹ H-NMR(600.1 MHz, d ₆ -DMSO): δ= 9.4239 (1.6); 9.4119 (1.6); 9.0696 (16.0); 8.2566 (2.1); 8.2540 (4.1); 8.2515 (2.3); 8.1673 (2.0); 8.1643 (3.3); 8.1616 (2.4); 8.1388 (2.3); 8.1358 (3.3); 8.1329 (1.8); 5.9612 (1.2); 5.9495 (1.9); 5.9377 (1.2); 3.3217 (23.7); 3.3172 (18.4); 2.5217 (0.4); 2.5186 (0.4); 2.5099 (7.7); 2.5068 (17.2); 2.5038 (24.2); 2.5007 (17.3); 2.4977 (7.8); 2.0754 (1.3); 2.0663 (0.3); 2.0582 (0.7); 2.0524 (0.8); 2.0502 (0.6); 2.0443 (1.5); 2.0385 (0.5); 2.0361 (0.8); 2.0304 (0.8); 2.0222 (0.4); 1.6115 (6.3); 1.5999 (6.3); 0.9854 (2.0); 0.9822 (2.5); 0.9715 (2.0); 0.9683 (2.4); 0.9593 (0.4); 0.8813 (0.8); 0.8776 (1.2); 0.8729 (1.6); 0.8694 (2.0); 0.8646 (1.5); 0.8612 (0.9); 0.8595 (0.8); 0.8566 (0.7); -0.0001 (2.6)	
I-62		¹ H-NMR(600.1 MHz, d ₆ -DMSO): δ= 9.4568 (1.2); 9.4455 (1.2); 8.5973 (2.0); 8.5965 (2.0); 8.5930 (2.3); 8.5889 (0.3); 8.3128 (0.3); 8.2618 (1.7); 8.2593 (3.2); 8.2567 (1.8); 8.2071 (1.7); 8.2041 (2.4); 8.2013 (1.6); 8.1814 (1.6); 8.1770 (1.8); 8.1668 (1.8); 8.1624 (2.0); 8.1177 (1.7); 8.1148 (2.6); 8.1117 (1.5); 7.8539 (0.3); 7.8225 (2.3); 7.8160 (0.4); 7.8080 (2.1); 7.8071 (2.0); 7.8009 (0.3); 7.4804 (0.4); 5.9867 (0.9); 5.9751 (1.4); 5.9635 (0.9); 5.7531 (3.1); 3.9429 (16.0); 3.9352 (2.4); 3.3169 (142.7); 3.0221 (0.6); 3.0168 (0.6); 3.0089 (1.1); 3.0036 (0.4); 3.0010 (0.6); 2.9957 (0.6); 2.6167 (0.6); 2.6137 (0.8); 2.6107 (0.6); 2.5227 (1.7); 2.5196 (2.1); 2.5166 (2.0); 2.5078 (42.0); 2.5047 (93.2); 2.5017 (131.0); 2.4986 (93.4); 2.4956 (42.1); 2.3886 (0.6); 2.3856 (0.8); 2.3824 (0.6); 1.6177 (4.9); 1.6061 (4.9); 1.5923 (0.8); 1.5807 (0.8); 1.1922 (0.9); 1.1843 (1.5); 1.1788 (1.5); 1.1715 (0.9); 1.1057 (0.4); 1.0964 (1.4); 1.0912 (1.4); 1.0832 (1.4); 1.0782 (1.3); -0.0001 (11.3); -0.0057 (0.4)	
I-63		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4376 (1.6); 9.4201 (1.6); 9.0201 (2.5); 9.0186 (2.7); 9.0148 (2.7); 9.0131 (2.6); 8.5349 (2.0); 8.5293 (1.9); 8.5133 (2.2); 8.5078 (2.2); 8.2236 (8.2); 8.2181 (2.4); 8.2145 (3.9); 8.2110 (2.3); 8.0162 (2.7); 8.0148 (2.7); 7.9947 (2.8); 7.9932 (3.0); 7.9835 (3.0); 7.9635 (3.4); 7.9602 (3.2); 6.0869 (1.1); 6.0695 (1.7); 6.0520 (1.1); 3.3130 (17.3); 3.2640 (0.4); 3.2485 (0.6); 3.2324 (0.8); 3.2159 (0.6); 3.2003 (0.3); 2.7221 (0.7); 2.7043 (0.7); 2.6771 (0.3); 2.6724 (0.4); 2.5257 (0.8); 2.5121 (19.5); 2.5077 (39.6); 2.5032 (54.4); 2.4987 (41.8); 2.4944 (21.1); 2.3302 (0.4); 2.0752 (1.6); 2.0681 (0.8); 2.0560 (1.4); 2.0439 (0.8); 2.0353 (0.7); 2.0229 (0.4); 1.6180 (5.8); 1.6006 (5.8); 1.0841 (14.5); 1.0674 (16.0); 1.0488 (1.9); 1.0026 (2.0); 0.9978 (2.5); 0.9819 (2.2); 0.9768 (2.4); 0.9619 (0.4); 0.9176 (0.4); 0.9055 (0.4); 0.8943 (1.4); 0.8879 (1.0); 0.8817 (2.0); 0.8760 (1.5); 0.8723 (1.4); 0.8642 (0.8); 0.8600 (1.1); 0.8465 (0.4); -0.0002 (1.1)	
I-64		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6066 (4.3); 9.5890 (4.4); 9.0250 (7.1); 9.0233 (7.4); 9.0195 (7.5); 9.0178 (6.8); 8.6166 (8.2); 8.5372 (5.8); 8.5317 (5.5); 8.5157 (6.2); 8.5101 (6.1); 8.4729 (7.4); 8.3977 (7.3); 8.3152 (0.5); 8.0198 (7.6); 8.0182 (7.3); 7.9983 (7.1); 7.9966 (6.8); 6.1354 (0.6); 6.1180 (3.0); 6.1006 (4.8); 6.0831 (3.1); 6.0657 (0.6); 4.4338 (0.4); 4.4161 (0.4); 3.4265 (1.3); 3.3783 (45.0); 3.3260 (207.4); 2.6812 (0.5); 2.6767 (1.0); 2.6721 (1.3); 2.6675 (1.0); 2.5254 (4.4); 2.5120 (76.2); 2.5076 (148.6); 2.5030 (198.1); 2.4985 (146.6); 2.4940 (70.5); 2.3390 (0.5); 2.3345 (0.9); 2.3299 (1.2); 2.3254 (0.9); 2.0918 (0.8); 2.0795 (1.9); 2.0749 (1.2); 2.0709 (2.1); 2.0678 (1.6); 2.0588 (3.9); 2.0466 (2.2); 2.0381 (2.0); 2.0258 (1.0); 1.6376 (16.0); 1.6202 (15.9); 1.3941 (0.4); 1.3764 (0.8); 1.3586 (0.4); 1.0326 (0.4); 1.0219 (0.6); 1.0035 (5.3); 0.9992 (6.7); 0.9828 (5.6); 0.9782 (6.6); 0.9620 (1.0); 0.9515 (0.7); 0.9201 (0.9); 0.9074 (1.1); 0.8971 (3.5); 0.8904 (2.6); 0.8836 (4.5); 0.8783 (4.0);	

		0.8734 (3.4); 0.8661 (2.2); 0.8612 (2.8); 0.8508 (0.8); 0.8475 (1.0); 0.8388 (0.6); 0.1459 (0.7); 0.0081 (7.8); -0.0001 (179.0); -0.0084 (7.0); -0.1495 (0.8)	
I-65		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.4522 (4.5); 9.4346 (4.6); 9.0195 (7.5); 9.0156 (7.3); 9.0140 (7.0); 8.5364 (5.8); 8.5309 (5.5); 8.5149 (6.1); 8.5093 (6.1); 8.3156 (0.5); 8.2377 (6.1); 8.2342 (10.6); 8.2307 (6.2); 8.0174 (7.5); 8.0161 (7.6); 7.9960 (6.9); 7.9945 (7.1); 7.9868 (0.4); 7.9061 (6.8); 7.8629 (5.8); 7.8587 (7.1); 7.8538 (4.4); 7.6078 (4.7); 7.4253 (10.0); 7.2426 (4.9); 6.1089 (0.6); 6.0916 (3.0); 6.0742 (4.8); 6.0567 (3.0); 6.0394 (0.6); 5.7559 (2.0); 3.3246 (236.5); 3.3115 (49.2); 2.6758 (1.3); 2.6713 (1.8); 2.6668 (1.3); 2.6622 (0.6); 2.5246 (5.8); 2.5112 (103.1); 2.5068 (204.6); 2.5023 (275.8); 2.4977 (206.6); 2.4933 (101.0); 2.3379 (0.6); 2.3337 (1.3); 2.3292 (1.8); 2.3247 (1.3); 2.0884 (0.8); 2.0762 (1.9); 2.0675 (2.0); 2.0645 (1.6); 2.0555 (3.9); 2.0433 (2.3); 2.0347 (2.0); 2.0225 (1.0); 1.8144 (0.4); 1.7965 (0.4); 1.6217 (16.0); 1.6043 (16.0); 1.2348 (0.5); 1.2056 (0.4); 1.1888 (0.5); 1.1735 (0.5); 1.0303 (0.4); 1.0201 (0.6); 1.0024 (5.4); 0.9975 (7.0); 0.9816 (5.7); 0.9765 (6.8); 0.9605 (1.0); 0.9509 (0.7); 0.9325 (0.3); 0.9170 (1.0); 0.9048 (1.1); 0.8935 (4.0); 0.8871 (2.6); 0.8809 (4.9); 0.8746 (3.9); 0.8702 (3.6); 0.8581 (3.1); 0.8439 (1.0); 0.8351 (0.7); 0.1459 (0.9); 0.0080 (8.8); -0.0002 (201.9); -0.0084 (7.7); -0.1495 (0.9)	
I-66		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.4105 (1.6); 9.3926 (1.6); 9.0698 (16.0); 8.1864 (2.2); 8.1829 (3.8); 8.1794 (2.2); 7.9723 (1.7); 7.9682 (2.6); 7.9645 (1.8); 7.9587 (1.2); 7.9527 (1.5); 7.9490 (1.5); 7.9447 (1.5); 7.9412 (1.5); 5.9674 (1.1); 5.9499 (1.8); 5.9323 (1.1); 3.3238 (57.4); 3.3053 (17.7); 2.6757 (0.4); 2.6713 (0.6); 2.6668 (0.4); 2.5246 (1.8); 2.5112 (34.6); 2.5068 (67.8); 2.5022 (90.4); 2.4977 (67.7); 2.4933 (33.1); 2.3337 (0.4); 2.3292 (0.6); 2.3247 (0.4); 2.0746 (2.4); 2.0632 (0.7); 2.0546 (0.8); 2.0513 (0.6); 2.0425 (1.4); 2.0302 (0.8); 2.0217 (0.7); 2.0094 (0.4); 1.6147 (5.9); 1.5973 (5.9); 1.0017 (0.4); 0.9887 (1.7); 0.9829 (2.6); 0.9719 (1.4); 0.9678 (1.7); 0.9621 (2.4); 0.9516 (0.6); 0.8832 (0.8); 0.8764 (1.6); 0.8708 (1.9); 0.8644 (2.3); 0.8587 (1.6); 0.0079 (2.2); -0.0002 (48.2); -0.0085 (1.8)	
I-67		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.5153 (1.5); 9.4973 (1.6); 9.0707 (16.0); 8.5353 (0.4); 8.4776 (2.6); 8.3813 (0.4); 8.3131 (2.5); 8.2560 (2.8); 7.3630 (1.1); 7.2484 (0.4); 7.2249 (2.6); 7.0868 (1.2); 5.9918 (1.0); 5.9742 (1.7); 5.9566 (1.1); 3.3251 (39.8); 3.3200 (19.8); 3.3032 (0.6); 2.6718 (0.4); 2.5250 (1.3); 2.5117 (22.4); 2.5073 (44.6); 2.5027 (60.5); 2.4982 (45.9); 2.4938 (22.7); 2.3295 (0.4); 2.0654 (0.7); 2.0568 (0.7); 2.0535 (0.6); 2.0446 (1.4); 2.0325 (0.8); 2.0239 (0.7); 2.0116 (0.4); 1.6276 (5.6); 1.6102 (5.6); 1.5385 (0.3); 1.0022 (0.3); 0.9885 (1.7); 0.9829 (2.6); 0.9711 (1.4); 0.9677 (1.6); 0.9621 (2.5); 0.9510 (0.5); 0.9318 (0.8); 0.9136 (1.6); 0.8951 (0.8); 0.8843 (0.8); 0.8785 (1.5); 0.8725 (1.9); 0.8662 (2.2); 0.8608 (1.8); 0.8523 (0.8); 0.0080 (1.9); -0.0002 (42.6); -0.0084 (1.6)	
I-68		¹ H-NMR(600.4 MHz, d ₆ -DMSO): δ = 9.5354 (3.9); 9.5234 (4.1); 8.5934 (7.4); 8.5853 (7.0); 8.5810 (7.0); 8.4337 (6.6); 8.3821 (6.5); 8.3088 (0.5); 8.1592 (4.7); 8.1549 (4.6); 8.1446 (5.1); 8.1403 (5.1); 7.8313 (7.7); 7.8168 (7.0); 6.0081 (0.6); 5.9968 (3.0); 5.9851 (4.6); 5.9734 (3.0); 5.9616 (0.7); 3.3682 (43.3); 3.3083 (171.1); 3.3058 (249.0); 2.6161 (1.1); 2.6131 (1.7); 2.6103 (1.2); 2.5221 (2.8); 2.5191 (3.4); 2.5160 (3.2); 2.5071 (85.9); 2.5042 (186.4); 2.5012 (259.2); 2.4982 (188.8); 2.4953 (90.4); 2.3883 (1.1); 2.3853 (1.6); 2.3819 (1.2); 2.0590 (0.8); 2.0510 (1.8); 2.0451 (1.9); 2.0372 (3.6); 2.0291 (2.1); 2.0233 (1.9); 2.0151 (1.0); 1.6225 (15.8); 1.6109 (16.0); 0.9967 (0.4); 0.9914 (0.4); 0.9799 (2.6); 0.9745 (5.2); 0.9661 (2.5); 0.9606 (5.3); 0.9463	

		(0.5); 0.9398 (0.6); 0.8972 (0.6); 0.8940 (0.9); 0.8895 (1.0); 0.8857 (0.9); 0.8786 (2.8); 0.8706 (3.1); 0.8644 (2.6); 0.8595 (2.5); 0.8564 (2.4); 0.8512 (2.5); 0.8414 (1.1); 0.8363 (0.7); 0.8328 (0.6); 0.0967 (0.7); 0.0052 (3.9); -0.0002 (140.3); -0.0056 (5.4); -0.1003 (0.6)	
I-68		¹ H-NMR(600.4 MHz, d ₆ -DMSO): δ= 9.5358 (4.4); 9.5238 (3.3); 8.5937 (8.2); 8.5856 (5.9); 8.5813 (5.5); 8.4342 (5.7); 8.3945 (2.2); 8.3826 (5.4); 8.1592 (4.6); 8.1549 (4.6); 8.1447 (4.4); 8.1404 (4.0); 7.8442 (1.6); 7.8315 (6.6); 7.8170 (5.5); 6.0090 (1.2); 5.9971 (3.3); 5.9853 (4.2); 5.9735 (2.5); 3.3812 (9.4); 3.3683 (35.6); 3.3191 (49.9); 3.3058 (230.9); 2.6132 (3.2); 2.5142 (77.1); 2.5111 (83.4); 2.5073 (125.8); 2.5042 (183.7); 2.5012 (223.0); 2.4982 (156.6); 2.4952 (75.3); 2.3852 (2.2); 2.0509 (2.3); 2.0452 (2.2); 2.0371 (3.5); 2.0290 (2.0); 2.0233 (1.7); 2.0151 (0.9); 1.6353 (3.8); 1.6226 (16.0); 1.6109 (13.3); 0.9744 (6.0); 0.9662 (3.3); 0.9605 (5.0); 0.8787 (3.7); 0.8706 (3.8); 0.0968 (0.7); 0.0129 (23.2); 0.0053 (11.0); -0.0002 (118.8); -0.0057 (6.2)	
I-69		¹ H-NMR(600.4 MHz, d ₆ -DMSO): δ= 9.4601 (4.2); 9.4482 (4.3); 8.5800 (6.6); 8.5758 (6.8); 8.3811 (9.0); 8.3093 (0.4); 8.1585 (4.2); 8.1543 (4.2); 8.1440 (4.5); 8.1397 (4.6); 8.0564 (11.3); 7.8303 (7.4); 7.8157 (6.7); 5.9890 (0.6); 5.9777 (2.8); 5.9661 (4.5); 5.9544 (2.9); 5.9425 (0.7); 5.7493 (0.8); 3.3442 (40.2); 3.3059 (237.6); 2.6132 (1.3); 2.5221 (1.7); 2.5192 (2.4); 2.5162 (2.2); 2.5041 (149.1); 2.5012 (222.7); 2.4984 (186.0); 2.3852 (1.6); 2.0582 (0.7); 2.0501 (1.6); 2.0442 (1.8); 2.0362 (3.5); 2.0282 (2.2); 2.0224 (1.9); 2.0142 (1.0); 1.6121 (15.6); 1.6005 (16.0); 0.9743 (5.7); 0.9602 (5.9); 0.8930 (0.8); 0.8881 (1.0); 0.8764 (3.0); 0.8682 (3.3); 0.8583 (3.0); 0.8534 (2.7); 0.8501 (2.9); 0.8398 (1.3); 0.0965 (0.4); 0.0053 (2.1); -0.0002 (107.4); -0.1004 (0.6)	
I-70		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1973 (1.3); 9.1802 (1.3); 9.0161 (2.1); 9.0145 (2.3); 9.0106 (2.2); 9.0090 (2.1); 8.5424 (1.8); 8.5368 (1.6); 8.5208 (1.9); 8.5152 (1.8); 7.9784 (2.3); 7.9768 (2.2); 7.9568 (2.1); 7.9551 (2.1); 7.7756 (2.1); 7.6275 (2.1); 7.4508 (2.1); 7.1527 (1.0); 7.0133 (2.2); 6.8739 (1.1); 6.0600 (1.0); 6.0426 (1.5); 6.0252 (1.0); 5.7558 (1.4); 3.9587 (16.0); 3.3245 (40.0); 2.6715 (0.4); 2.5248 (1.2); 2.5115 (24.9); 2.5071 (48.0); 2.5025 (61.3); 2.4979 (43.8); 2.4935 (21.2); 2.3294 (0.4); 2.0677 (0.6); 2.0593 (0.6); 2.0555 (0.5); 2.0469 (1.1); 2.0344 (0.7); 2.0260 (0.6); 1.6174 (5.3); 1.5999 (5.2); 1.2271 (0.4); 1.0430 (0.6); 1.0323 (2.0); 1.0268 (2.1); 1.0163 (1.0); 1.0113 (2.1); 1.0058 (2.0); 0.9958 (0.6); 0.7813 (0.9); 0.7713 (1.9); 0.7682 (2.0); 0.7655 (1.9); 0.7588 (2.0); 0.7552 (1.9); 0.7434 (0.7); -0.0002 (3.3)	
I-71		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2630 (1.4); 9.2457 (1.5); 9.0154 (2.4); 9.0102 (2.4); 8.5425 (1.7); 8.5370 (1.6); 8.5209 (1.8); 8.5154 (1.7); 7.9769 (2.6); 7.9554 (2.4); 7.9404 (1.9); 7.9366 (3.0); 7.9328 (1.8); 7.6857 (2.9); 7.5885 (2.8); 6.0593 (1.0); 6.0419 (1.6); 6.0246 (1.0); 5.7551 (2.1); 3.9604 (16.0); 3.3229 (44.6); 2.6758 (0.4); 2.6714 (0.6); 2.6667 (0.4); 2.5110 (37.0); 2.5070 (69.7); 2.5026 (88.2); 2.4981 (63.5); 2.4940 (31.3); 2.3337 (0.4); 2.3295 (0.6); 2.3249 (0.4); 1.6117 (5.7); 1.5942 (5.6); 1.5690 (0.5); 1.5532 (1.6); 1.5487 (1.6); 1.5340 (0.6); 1.5201 (0.5); 1.5046 (1.5); 1.5000 (1.6); 1.4855 (0.7); 1.2938 (0.6); 1.2779 (1.8); 1.2724 (2.0); 1.2550 (2.1); 1.2502 (1.6); 1.2334 (0.8); -0.0002 (6.7)	

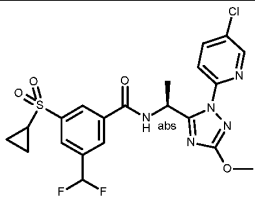
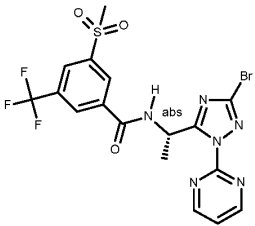
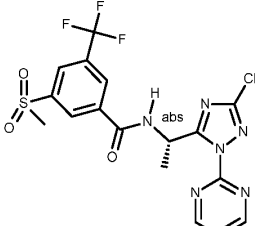
I-72		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2141 (0.9); 9.2047 (1.0); 9.1968 (1.0); 9.1878 (0.9); 9.0149 (2.7); 9.0095 (2.6); 8.5429 (2.2); 8.5373 (2.1); 8.5213 (2.4); 8.5158 (2.3); 7.9777 (2.6); 7.9558 (2.5); 7.9275 (1.2); 7.9234 (2.6); 7.9192 (2.6); 7.9151 (1.1); 7.7094 (1.6); 7.6969 (1.7); 7.6856 (2.6); 6.0527 (0.8); 6.0372 (1.3); 6.0199 (0.9); 5.7557 (2.9); 3.9595 (16.0); 3.3231 (76.0); 3.1446 (0.3); 3.1128 (0.7); 3.0921 (0.7); 3.0838 (0.4); 3.0623 (0.4); 2.6757 (0.6); 2.6711 (0.8); 2.6666 (0.6); 2.5245 (2.6); 2.5111 (47.9); 2.5067 (92.9); 2.5022 (119.5); 2.4976 (85.6); 2.4931 (41.4); 2.3335 (0.6); 2.3289 (0.8); 2.3244 (0.6); 2.1490 (0.4); 2.1397 (0.6); 2.1315 (0.5); 2.1185 (0.6); 2.1074 (0.6); 2.0979 (0.4); 2.0547 (0.3); 2.0398 (0.5); 2.0240 (0.6); 2.0088 (0.5); 1.9984 (0.3); 1.9943 (0.4); 1.6100 (6.2); 1.5926 (6.2); -0.0002 (7.4)	
I-73		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4831 (1.3); 9.4660 (1.4); 9.0288 (2.3); 9.0246 (2.3); 8.5518 (1.7); 8.5463 (1.6); 8.5302 (1.8); 8.5247 (1.8); 8.2541 (1.9); 8.2508 (3.2); 8.2473 (1.9); 8.0253 (0.7); 8.0192 (1.1); 8.0159 (1.0); 7.9991 (1.8); 7.9959 (2.4); 7.9909 (3.5); 7.9802 (1.2); 7.9765 (1.3); 7.9693 (2.6); 6.1064 (1.0); 6.0891 (1.5); 6.0718 (1.0); 3.9690 (16.0); 3.3327 (127.0); 3.3202 (17.3); 2.6793 (0.5); 2.6748 (0.7); 2.6702 (0.5); 2.5452 (3.9); 2.5281 (2.3); 2.5147 (45.0); 2.5104 (87.3); 2.5059 (111.6); 2.5013 (80.0); 2.4970 (39.1); 2.3371 (0.5); 2.3327 (0.7); 2.3282 (0.5); 2.0786 (5.6); 1.6355 (5.4); 1.6180 (5.4)	
I-74		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1090 (1.3); 9.0918 (1.3); 9.0136 (2.3); 9.0094 (2.2); 8.5429 (1.7); 8.5373 (1.6); 8.5213 (1.8); 8.5158 (1.8); 7.9744 (2.3); 7.9541 (2.1); 7.9528 (2.2); 7.4326 (1.4); 7.3746 (2.8); 7.3714 (1.9); 7.3448 (2.0); 7.2477 (2.9); 7.0663 (2.3); 7.0627 (2.9); 6.0442 (1.0); 6.0268 (1.5); 6.0094 (1.0); 3.9588 (16.0); 3.3303 (68.1); 2.5419 (0.8); 2.5247 (1.0); 2.5114 (21.1); 2.5071 (41.1); 2.5026 (52.8); 2.4980 (37.7); 2.4936 (18.1); 2.0754 (2.0); 2.0095 (0.6); 2.0013 (0.6); 1.9975 (0.5); 1.9888 (1.1); 1.9762 (0.7); 1.9679 (0.6); 1.6115 (5.3); 1.5941 (5.2); 1.0252 (0.5); 1.0148 (1.9); 1.0092 (2.0); 0.9989 (0.9); 0.9939 (1.9); 0.9883 (2.0); 0.9783 (0.6); 0.7766 (0.8); 0.7665 (1.8); 0.7632 (1.9); 0.7543 (1.9); 0.7502 (1.8); 0.7385 (0.6)	
I-75		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5801 (1.3); 9.5629 (1.4); 9.0269 (2.3); 9.0230 (2.3); 9.0215 (2.2); 8.5523 (1.7); 8.5467 (1.6); 8.5307 (1.8); 8.5251 (1.8); 8.4482 (1.8); 8.4447 (3.2); 8.4412 (1.8); 8.1277 (1.9); 8.0911 (1.9); 7.9907 (2.3); 7.9707 (2.1); 7.9691 (2.2); 6.1178 (0.9); 6.1005 (1.5); 6.0832 (1.0); 3.9708 (16.0); 3.3622 (15.2); 3.3345 (117.4); 2.6798 (0.4); 2.6752 (0.5); 2.6707 (0.4); 2.5457 (2.5); 2.5285 (1.7); 2.5152 (34.4); 2.5108 (67.3); 2.5063 (86.6); 2.5017 (62.0); 2.4972 (30.0); 2.3376 (0.4); 2.3331 (0.5); 2.3284 (0.4); 2.0789 (3.7); 1.6454 (5.2); 1.6280 (5.2)	
I-76		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0864 (1.4); 9.0671 (15.0); 7.4332 (1.4); 7.3492 (2.8); 7.3459 (2.0); 7.3311 (1.7); 7.3263 (2.1); 7.2482 (3.0); 7.0635 (3.1); 5.9468 (1.0); 5.9293 (1.5); 5.9119 (1.0); 3.9395 (16.0); 3.3337 (85.5); 2.6751 (0.4); 2.5455 (2.1); 2.5283 (1.4); 2.5151 (25.8); 2.5107 (49.7); 2.5062 (63.6); 2.5016 (45.5); 2.4971 (22.1); 2.3329 (0.4); 2.0790 (2.3); 2.0076 (0.6); 1.9992 (0.6); 1.9868 (1.1); 1.9743 (0.6); 1.9659 (0.6); 1.6075 (5.3); 1.5900 (5.3); 1.0261 (0.6); 1.0152 (2.0); 1.0096 (2.1); 0.9993 (1.0); 0.9943 (2.0); 0.9888 (2.0); 0.9788 (0.7); 0.7763 (0.8); 0.7660 (2.1); 0.7607 (2.1); 0.7537 (2.1); 0.7486 (2.1); 0.7375 (0.7)	

I-77		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5582 (1.3); 9.5409 (1.4); 9.0758 (13.9); 8.4273 (1.9); 8.4238 (3.2); 8.4203 (1.9); 8.1160 (2.0); 8.0853 (2.0); 6.0177 (1.0); 6.0003 (1.5); 5.9829 (1.0); 3.9474 (16.0); 3.3865 (0.4); 3.3577 (15.6); 3.3328 (129.1); 2.6796 (0.6); 2.6753 (0.8); 2.6707 (0.6); 2.5455 (2.9); 2.5284 (2.6); 2.5150 (51.3); 2.5107 (99.9); 2.5062 (128.5); 2.5016 (92.9); 2.4973 (45.7); 2.3375 (0.6); 2.3328 (0.8); 2.3284 (0.6); 2.0789 (2.6); 1.6377 (5.3); 1.6203 (5.3)	
I-78		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4810 (1.3); 9.4634 (1.3); 9.0768 (13.6); 8.2974 (1.7); 8.2937 (3.2); 8.2900 (2.0); 8.2124 (1.6); 8.2079 (2.8); 8.2039 (1.9); 8.1572 (1.9); 8.1528 (3.0); 8.1484 (1.7); 6.0025 (0.9); 5.9850 (1.5); 5.9676 (1.0); 3.9460 (16.0); 3.3325 (233.0); 2.6796 (1.0); 2.6751 (1.3); 2.6706 (1.0); 2.5455 (2.3); 2.5282 (4.9); 2.5150 (87.1); 2.5107 (166.7); 2.5061 (212.7); 2.5016 (151.0); 2.4971 (72.4); 2.3373 (1.0); 2.3330 (1.3); 2.3284 (0.9); 2.0791 (6.0); 1.6252 (5.3); 1.6078 (5.3); 0.0043 (0.3)	
I-79		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4962 (1.3); 9.4789 (1.3); 9.0756 (14.4); 8.2661 (1.7); 8.2624 (3.3); 8.2586 (2.1); 8.2247 (1.9); 8.2202 (2.8); 8.2161 (1.8); 8.1310 (2.0); 8.1266 (2.9); 8.1223 (1.7); 6.0028 (0.9); 5.9854 (1.5); 5.9680 (1.0); 3.9468 (16.0); 3.3337 (170.1); 3.0380 (0.6); 3.0302 (0.7); 3.0252 (0.4); 3.0185 (1.2); 3.0103 (0.5); 3.0064 (0.7); 2.9987 (0.6); 2.6795 (0.8); 2.6749 (1.0); 2.6704 (0.7); 2.5453 (4.3); 2.5283 (3.4); 2.5149 (65.0); 2.5105 (126.2); 2.5060 (161.6); 2.5014 (116.1); 2.4970 (56.6); 2.3373 (0.7); 2.3328 (1.0); 2.3283 (0.7); 2.0789 (6.7); 1.6270 (5.2); 1.6096 (5.1); 1.1989 (0.8); 1.1890 (1.8); 1.1803 (1.7); 1.1715 (0.9); 1.1162 (0.5); 1.1040 (1.6); 1.0968 (1.4); 1.0841 (1.5); 1.0788 (1.2)	
I-80		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5304 (1.5); 9.5130 (1.5); 8.6016 (2.7); 8.5960 (2.6); 8.4276 (2.1); 8.4242 (3.4); 8.4208 (2.0); 8.1894 (1.8); 8.1829 (1.7); 8.1675 (2.0); 8.1610 (1.9); 8.1005 (2.2); 8.0803 (2.2); 7.8283 (2.8); 7.8063 (2.4); 6.0065 (1.0); 5.9891 (1.6); 5.9717 (1.0); 3.9433 (16.0); 3.3546 (15.9); 3.3280 (114.1); 2.6759 (0.5); 2.6715 (0.6); 2.6669 (0.4); 2.5419 (1.8); 2.5070 (77.8); 2.5026 (97.6); 2.4981 (70.0); 2.4941 (34.2); 2.3336 (0.4); 2.3294 (0.6); 2.3247 (0.4); 2.0755 (2.4); 1.6312 (5.5); 1.6138 (5.5)	
I-81		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5286 (4.2); 9.5111 (4.2); 9.0175 (7.5); 9.0121 (7.2); 8.5357 (5.1); 8.5302 (4.8); 8.5142 (5.4); 8.5086 (5.3); 8.4005 (10.9); 8.3973 (6.4); 8.2579 (0.6); 8.2103 (1.1); 8.0808 (12.9); 8.0377 (0.4); 8.0156 (7.5); 7.9940 (8.1); 7.9610 (0.4); 7.9287 (0.6); 7.8983 (0.7); 7.8788 (0.4); 6.2010 (0.7); 6.1705 (0.7); 6.1102 (0.6); 6.0929 (2.9); 6.0755 (4.6); 6.0580 (2.9); 6.0406 (0.6); 4.2903 (0.4); 4.2738 (0.5); 4.2573 (0.3); 3.8273 (0.4); 3.8105 (0.5); 3.7935 (0.4); 3.4569 (0.4); 3.4384 (0.4); 3.3985 (0.9); 3.3823 (2.4); 3.3732 (9.5); 3.3558 (48.6); 3.3282 (208.9); 2.6716 (3.5); 2.5419 (2.1); 2.5244 (4.4); 2.5112 (92.7); 2.5069 (180.4); 2.5025 (233.0); 2.4979 (168.6); 2.4936 (82.9); 2.3381 (0.5); 2.3338 (1.0); 2.3292 (1.4); 2.3248 (1.0); 2.0915 (0.8); 2.0789 (2.2); 2.0756 (6.6); 2.0707 (2.3); 2.0585 (3.8); 2.0464 (2.3); 2.0379 (2.0); 2.0256 (1.0); 1.6264 (16.0); 1.6090 (16.0); 1.4432 (0.3); 1.3617 (0.4); 1.2700 (0.4); 1.2472 (6.2); 1.2347 (8.0); 1.2312 (7.8); 1.2186 (6.2); 1.2071 (2.2); 1.1904 (2.0); 1.1854 (1.8); 1.1676 (0.7); 1.0324 (0.4); 1.0226 (0.6); 1.0044 (5.3); 0.9996 (7.0); 0.9836 (5.6); 0.9787 (6.9); 0.9623 (1.0); 0.9526 (0.7); 0.9336 (0.4); 0.9183 (1.0); 0.9057 (1.2); 0.8946 (4.0); 0.8884 (2.6); 0.8822 (4.7); 0.8760 (3.8); 0.8712 (3.6); 0.8590 (3.0); 0.8447 (1.0); 0.8359 (0.7); -0.0004 (0.7)	

I-82		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.5081 (1.6); 9.4904 (1.6); 9.0697 (16.0); 8.3764 (2.1); 8.3729 (3.7); 8.3693 (2.2); 8.0693 (6.0); 5.9756 (1.1); 5.9581 (1.7); 5.9405 (1.1); 3.3821 (2.8); 3.3722 (1.4); 3.3481 (19.4); 3.3268 (423.9); 2.6751 (1.6); 2.6706 (2.3); 2.6660 (1.7); 2.5409 (1.2); 2.5238 (6.9); 2.5104 (145.0); 2.5061 (285.5); 2.5016 (369.9); 2.4971 (266.5); 2.4926 (130.0); 2.3329 (1.6); 2.3284 (2.2); 2.3239 (1.6); 2.0747 (5.0); 2.0650 (0.8); 2.0564 (0.8); 2.0443 (1.5); 2.0321 (0.9); 2.0235 (0.8); 2.0113 (0.4); 1.6220 (6.1); 1.6046 (6.1); 1.2715 (1.4); 1.2570 (2.1); 1.2415 (2.2); 1.2357 (1.0); 1.2223 (0.7); 1.2183 (0.8); 1.2090 (0.4); 1.1929 (0.3); 1.1345 (0.5); 1.1204 (0.4); 1.0036 (0.4); 0.9904 (1.8); 0.9846 (2.8); 0.9729 (1.6); 0.9696 (1.8); 0.9638 (2.7); 0.9528 (0.6); 0.8824 (0.8); 0.8762 (1.6); 0.8704 (2.0); 0.8640 (2.4); 0.8586 (1.9); -0.0002 (1.0)	
I-83		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 10.7265 (2.2); 9.2168 (0.4); 9.1983 (4.4); 9.1809 (4.3); 9.0595 (0.6); 9.0558 (0.6); 9.0540 (0.6); 9.0420 (6.6); 9.0404 (7.2); 9.0366 (7.1); 9.0349 (6.8); 8.6005 (0.4); 8.5950 (0.4); 8.5790 (0.6); 8.5713 (5.8); 8.5658 (5.4); 8.5498 (6.1); 8.5443 (6.0); 8.3158 (0.8); 8.0432 (0.9); 8.0381 (7.1); 8.0365 (7.3); 8.0218 (0.8); 8.0167 (6.6); 8.0150 (6.9); 7.6174 (2.4); 7.5904 (1.7); 7.4792 (9.3); 7.4754 (7.8); 7.4691 (6.0); 7.4665 (6.3); 7.3816 (1.5); 7.2813 (6.2); 6.0400 (0.6); 6.0228 (3.0); 6.0055 (4.7); 5.9880 (3.0); 5.9709 (0.6); 3.3241 (307.8); 2.6805 (0.9); 2.6759 (1.8); 2.6713 (2.5); 2.6668 (1.8); 2.6623 (0.9); 2.5247 (8.2); 2.5113 (147.1); 2.5069 (293.7); 2.5024 (382.8); 2.4978 (272.6); 2.4933 (129.9); 2.3380 (0.8); 2.3338 (1.7); 2.3292 (2.4); 2.3246 (1.7); 2.3203 (0.8); 2.1189 (0.5); 2.1106 (0.5); 2.0981 (1.0); 2.0858 (0.6); 2.0746 (4.0); 2.0689 (1.0); 2.0562 (1.9); 2.0478 (2.0); 2.0354 (3.7); 2.0229 (2.2); 2.0145 (2.1); 2.0019 (1.0); 1.6456 (1.4); 1.6221 (16.0); 1.6046 (15.8); 1.0901 (0.6); 1.0788 (1.6); 1.0733 (1.8); 1.0628 (1.1); 1.0579 (1.8); 1.0507 (3.2); 1.0391 (6.6); 1.0335 (6.9); 1.0230 (3.6); 1.0181 (6.6); 1.0126 (6.6); 1.0023 (2.7); 0.8518 (0.8); 0.8409 (2.0); 0.8359 (2.0); 0.8289 (1.8); 0.8236 (2.1); 0.8116 (0.7); 0.7943 (2.8); 0.7836 (7.4); 0.7784 (7.4); 0.7713 (7.0); 0.7662 (7.9); 0.7547 (2.2); 0.1458 (2.6); 0.0294 (0.4); 0.0079 (24.9); -0.0001 (595.9); -0.0085 (22.3); -0.1497 (2.6)	
I-84		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 10.7266 (3.6); 9.2140 (4.1); 9.1970 (4.1); 9.0551 (7.0); 9.0505 (6.6); 8.5931 (4.9); 8.5876 (4.6); 8.5717 (5.2); 8.5662 (5.0); 8.3152 (1.2); 8.0481 (6.9); 8.0265 (6.4); 7.6487 (1.0); 7.6175 (3.2); 7.5909 (2.2); 7.5547 (0.6); 7.4819 (9.1); 7.4677 (6.3); 7.3832 (2.3); 7.3415 (0.6); 7.2813 (6.1); 6.0361 (0.6); 6.0197 (2.8); 6.0024 (4.5); 5.9853 (2.9); 5.9685 (0.6); 3.3712 (0.3); 3.3223 (246.8); 2.6754 (3.2); 2.6709 (4.3); 2.6665 (3.2); 2.5240 (16.4); 2.5108 (269.1); 2.5065 (518.5); 2.5021 (666.3); 2.4975 (481.2); 2.4932 (236.5); 2.4235 (0.4); 2.3334 (3.1); 2.3289 (4.2); 2.3244 (3.0); 2.1318 (0.4); 2.1189 (0.8); 2.1106 (0.9); 2.0983 (1.5); 2.0867 (1.0); 2.0746 (1.1); 2.0699 (1.0); 2.0571 (1.9); 2.0490 (2.0); 2.0364 (3.6); 2.0240 (2.1); 2.0154 (2.0); 2.0031 (1.0); 1.6397 (16.0); 1.6222 (16.0); 1.0899 (0.8); 1.0789 (2.2); 1.0735 (2.4); 1.0627 (1.5); 1.0580 (2.5); 1.0514 (4.1); 1.0393 (6.6); 1.0336 (7.2); 1.0233 (3.8); 1.0184 (6.4); 1.0129 (6.4); 1.0026 (2.6); 0.9861 (0.3); 0.8520 (1.0); 0.8414 (2.6); 0.8364 (2.6); 0.8293 (2.5); 0.8242 (2.8); 0.8122 (0.9); 0.7938 (3.0); 0.7829 (8.0); 0.7780 (7.9); 0.7708 (7.4); 0.7657 (8.5); 0.7540 (2.3); 0.1459 (1.2); 0.0078 (11.8); -0.0001 (262.6); -0.0083 (10.9); -0.1496 (1.2)	

I-85		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 10.7243 (0.4); 9.2173 (4.0); 9.2006 (4.1); 9.0592 (6.7); 9.0552 (6.7); 8.6002 (4.8); 8.5947 (4.6); 8.5787 (5.1); 8.5733 (5.0); 8.3153 (1.1); 8.0447 (6.8); 8.0232 (6.3); 7.6154 (0.6); 7.5871 (0.5); 7.4844 (8.8); 7.4689 (6.1); 7.3782 (0.3); 7.2819 (6.0); 6.0320 (0.6); 6.0146 (2.8); 5.9974 (4.4); 5.9802 (2.9); 5.9626 (0.6); 3.3920 (0.4); 3.3241 (557.1); 2.6754 (3.2); 2.6710 (4.2); 2.6667 (3.1); 2.5994 (0.4); 2.5065 (500.2); 2.5021 (633.8); 2.4976 (460.4); 2.3332 (3.0); 2.3290 (4.0); 2.3245 (3.0); 2.0739 (1.0); 2.0709 (0.9); 2.0580 (1.7); 2.0496 (1.9); 2.0371 (3.4); 2.0249 (2.1); 2.0163 (1.9); 2.0039 (0.9); 1.6453 (16.0); 1.6279 (15.9); 1.0730 (0.6); 1.0510 (2.5); 1.0396 (6.0); 1.0341 (6.3); 1.0235 (3.4); 1.0187 (6.0); 1.0133 (5.9); 1.0028 (2.4); 0.8338 (0.7); 0.8226 (0.6); 0.7936 (2.4); 0.7827 (7.0); 0.7777 (6.8); 0.7705 (6.5); 0.7656 (7.2); 0.7539 (2.0); 0.1459 (1.8); 0.0078 (19.5); -0.0002 (416.4); -0.0083 (19.7); -0.1496 (1.9)	
I-86		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4742 (4.0); 9.4567 (4.1); 9.0167 (7.0); 9.0128 (6.9); 9.0113 (6.7); 8.5367 (5.4); 8.5312 (5.2); 8.5152 (5.6); 8.5096 (5.6); 8.3153 (0.7); 8.2020 (5.5); 8.1986 (9.8); 8.1952 (6.1); 8.1351 (0.6); 8.0186 (7.0); 8.0172 (7.3); 8.0087 (0.6); 7.9971 (6.4); 7.9957 (6.8); 7.9881 (0.5); 7.9527 (0.4); 7.9152 (6.3); 7.8262 (5.2); 7.8219 (6.9); 7.8172 (4.7); 7.6201 (4.3); 7.4375 (9.2); 7.2548 (4.6); 6.1072 (0.6); 6.0901 (3.0); 6.0727 (4.6); 6.0552 (2.9); 6.0377 (0.6); 5.7552 (4.0); 3.3237 (213.7); 3.0059 (0.9); 2.9944 (2.3); 2.9862 (2.1); 2.9814 (1.4); 2.9746 (3.9); 2.9663 (1.6); 2.9625 (2.2); 2.9548 (2.0); 2.9428 (1.0); 2.8911 (2.7); 2.7314 (2.3); 2.6756 (1.6); 2.6711 (2.2); 2.6666 (1.7); 2.5244 (7.0); 2.5110 (125.9); 2.5067 (253.5); 2.5022 (332.5); 2.4976 (241.1); 2.4932 (118.0); 2.3335 (1.5); 2.3290 (2.1); 2.3245 (1.6); 2.0898 (0.8); 2.0775 (1.9); 2.0688 (2.0); 2.0567 (3.8); 2.0446 (2.3); 2.0360 (2.0); 2.0238 (1.0); 1.8150 (0.7); 1.7970 (0.7); 1.6232 (16.0); 1.6058 (15.9); 1.2344 (0.6); 1.2238 (0.8); 1.2123 (0.9); 1.2000 (3.2); 1.1876 (4.9); 1.1797 (5.5); 1.1683 (4.2); 1.1565 (1.6); 1.1435 (1.6); 1.1305 (0.9); 1.1169 (1.9); 1.1016 (5.7); 1.0968 (5.0); 1.0818 (5.4); 1.0772 (4.2); 1.0602 (0.8); 1.0529 (0.4); 1.0303 (0.5); 1.0224 (0.5); 1.0030 (5.1); 0.9978 (7.2); 0.9822 (5.0); 0.9770 (7.4); 0.9594 (0.9); 0.9508 (0.7); 0.9338 (0.3); 0.9181 (1.1); 0.9060 (1.2); 0.8945 (4.2); 0.8887 (2.4); 0.8821 (4.5); 0.8766 (3.8); 0.8699 (3.3); 0.8651 (2.4); 0.8578 (2.9); 0.8424 (1.1); 0.8337 (0.8); 0.1457 (2.3); 0.0196 (1.1); 0.0079 (21.3); -0.0002 (530.1); -0.0085 (21.6); -0.1497 (2.3)	
I-87		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5163 (1.4); 9.4991 (1.5); 9.0227 (2.5); 9.0185 (2.4); 8.5484 (1.7); 8.5428 (1.6); 8.5268 (1.8); 8.5213 (1.8); 8.2808 (3.4); 8.2773 (2.2); 8.2389 (1.9); 8.2345 (3.0); 8.2306 (1.9); 8.1295 (1.9); 8.1252 (3.0); 8.1210 (1.8); 7.9866 (2.5); 7.9649 (2.3); 6.1029 (1.0); 6.0855 (1.6); 6.0683 (1.0); 3.9667 (16.0); 3.3241 (125.1); 3.0354 (0.6); 3.0278 (0.7); 3.0160 (1.3); 3.0041 (0.7); 2.9962 (0.7); 2.9841 (0.3); 2.6756 (0.7); 2.6712 (1.0); 2.6667 (0.7); 2.5066 (117.5); 2.5022 (152.4); 2.4978 (111.4); 2.3334 (0.7); 2.3290 (1.0); 2.3247 (0.7); 1.6314 (5.6); 1.6140 (5.6); 1.1987 (0.9); 1.1902 (1.9); 1.1871 (1.9); 1.1792 (1.9); 1.1703 (1.2); 1.1368 (0.4); 1.1237 (0.3); 1.1146 (0.5); 1.1022 (1.8); 1.0951 (1.6); 1.0825 (1.7); 1.0769 (1.3); 0.1459 (0.9); 0.0079 (8.5); -0.0001 (196.1); -0.0085 (8.4); -0.1497 (0.9)	
I-88		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5609 (1.4); 9.5434 (1.4); 9.0726 (13.1); 8.5194 (2.3); 8.3525 (2.3); 8.2658 (2.2); 7.3699 (1.0); 7.2317 (2.2); 7.0937 (1.1); 6.0269 (1.0); 6.0094 (1.6); 5.9920 (1.0); 3.9436 (16.0); 3.9316 (0.5); 3.3389 (61.7); 3.3257 (22.3); 2.6723 (0.4); 2.5254 (1.4); 2.5122 (23.7); 2.5079 (46.4); 2.5034 (60.1); 2.4989 (43.3); 2.4944 (21.2); 2.3302 (0.4); 2.0748 (0.4); 1.6374 (5.4); 1.6200 (5.4); 0.0078 (1.6); -0.0002 (36.2); -0.0085 (1.4)	

I-89		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5861 (1.4); 9.5689 (1.4); 9.0287 (2.4); 9.0239 (2.4); 8.5493 (3.2); 8.5443 (4.0); 8.5281 (1.9); 8.5225 (1.8); 8.3726 (2.4); 8.2743 (2.4); 7.9916 (2.5); 7.9702 (2.3); 7.3759 (1.0); 7.2377 (2.4); 7.0997 (1.1); 6.1296 (1.0); 6.1123 (1.6); 6.0949 (1.0); 3.9675 (16.0); 3.3318 (19.0); 3.3281 (23.2); 2.5125 (13.0); 2.5083 (25.0); 2.5038 (31.9); 2.4993 (22.9); 2.4950 (11.2); 1.6463 (5.5); 1.6289 (5.5); 0.0077 (0.7); -0.0002 (17.6); -0.0084 (0.8)
I-90		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5523 (4.2); 9.5349 (4.3); 9.0144 (6.9); 9.0099 (7.1); 8.5357 (4.8); 8.5302 (4.6); 8.5141 (5.2); 8.5086 (5.1); 8.3667 (9.4); 8.3157 (0.4); 8.0960 (6.3); 8.0526 (6.2); 8.0181 (7.3); 7.9966 (6.7); 6.1108 (0.6); 6.0937 (2.9); 6.0762 (4.6); 6.0588 (2.9); 6.0417 (0.6); 5.7558 (1.2); 3.3237 (69.2); 3.0592 (0.8); 3.0472 (1.8); 3.0394 (2.0); 3.0278 (3.7); 3.0158 (2.1); 3.0081 (1.9); 2.9961 (0.9); 2.6760 (1.4); 2.6718 (1.8); 2.6674 (1.4); 2.5072 (210.0); 2.5028 (266.1); 2.4985 (193.0); 2.3340 (1.3); 2.3295 (1.7); 2.3254 (1.3); 2.0931 (0.8); 2.0808 (1.9); 2.0720 (2.1); 2.0600 (3.7); 2.0480 (2.2); 2.0394 (2.0); 2.0271 (1.0); 1.6285 (16.0); 1.6111 (15.9); 1.2444 (0.7); 1.2324 (0.9); 1.2200 (3.3); 1.2076 (4.8); 1.1996 (5.3); 1.1884 (3.7); 1.1761 (1.4); 1.1637 (1.0); 1.1507 (0.7); 1.1383 (1.1); 1.1315 (1.4); 1.1164 (5.6); 1.1110 (4.9); 1.0966 (5.3); 1.0912 (4.3); 1.0746 (0.7); 1.0659 (0.3); 1.0332 (0.4); 1.0243 (0.5); 1.0052 (5.2); 1.0003 (7.1); 0.9842 (5.2); 0.9794 (7.1); 0.9620 (0.9); 0.9532 (0.8); 0.9374 (0.4); 0.9209 (1.1); 0.9086 (1.3); 0.8971 (4.2); 0.8911 (2.5); 0.8847 (4.7); 0.8793 (3.9); 0.8727 (3.5); 0.8607 (3.0); 0.8451 (1.1); 0.8360 (0.8); 0.0077 (2.1); -0.0002 (43.0); -0.0083 (1.8)
I-91		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5755 (1.6); 9.5582 (1.6); 9.0701 (12.9); 8.3940 (3.4); 8.1269 (2.4); 8.0560 (2.5); 6.0158 (1.0); 5.9986 (1.6); 5.9811 (1.1); 3.9452 (16.0); 3.3224 (24.3); 3.0620 (0.3); 3.0501 (0.7); 3.0423 (0.8); 3.0308 (1.3); 3.0188 (0.8); 3.0111 (0.7); 2.9994 (0.3); 2.6713 (0.9); 2.5066 (108.6); 2.5024 (138.2); 2.4982 (104.4); 2.3293 (0.9); 1.6362 (5.9); 1.6188 (5.9); 1.2155 (1.1); 1.2058 (2.3); 1.1955 (2.3); 1.1866 (1.4); 1.1651 (0.4); 1.1503 (0.3); 1.1398 (0.4); 1.1262 (0.7); 1.1137 (2.1); 1.1062 (1.9); 1.0940 (2.0); 1.0886 (1.6); -0.0001 (23.5); -0.0082 (1.2)
I-92		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5980 (1.5); 9.5810 (1.5); 9.0204 (2.6); 9.0165 (2.6); 8.5488 (1.7); 8.5432 (1.7); 8.5272 (1.8); 8.5216 (1.8); 8.4133 (3.4); 8.1388 (2.2); 8.0612 (2.3); 7.9895 (2.4); 7.9883 (2.5); 7.9680 (2.3); 7.9666 (2.3); 6.1163 (1.0); 6.0991 (1.6); 6.0818 (1.0); 3.9683 (16.0); 3.3243 (61.3); 3.0655 (0.3); 3.0539 (0.7); 3.0462 (0.8); 3.0346 (1.3); 3.0228 (0.8); 3.0149 (0.7); 3.0029 (0.3); 2.6715 (0.9); 2.6672 (0.7); 2.5069 (103.6); 2.5025 (130.7); 2.4981 (96.0); 2.3336 (0.6); 2.3293 (0.8); 2.3248 (0.6); 2.0748 (0.8); 1.6443 (5.6); 1.6269 (5.6); 1.2317 (0.5); 1.2186 (1.2); 1.2084 (2.1); 1.1999 (2.1); 1.1888 (1.5); 1.1673 (0.3); 1.1535 (0.4); 1.1383 (0.4); 1.1297 (0.6); 1.1165 (2.1); 1.1097 (1.9); 1.0968 (2.0); 1.0913 (1.5); 0.0080 (1.0); -0.0002 (21.5); -0.0083 (1.1)
I-93		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5321 (1.8); 9.5144 (1.9); 9.0679 (16.0); 8.3435 (4.3); 8.0819 (2.8); 8.0478 (2.9); 5.9780 (1.3); 5.9604 (2.1); 5.9429 (1.4); 3.3244 (29.4); 3.0543 (0.4); 3.0420 (0.8); 3.0341 (0.9); 3.0226 (1.7); 3.0107 (1.0); 3.0028 (0.9); 2.9909 (0.4); 2.6764 (0.4); 2.6719 (0.6); 2.6676 (0.4); 2.5075 (75.1); 2.5031 (95.4); 2.4986 (69.6); 2.3343 (0.5); 2.3297 (0.6); 2.3254 (0.5); 2.0755 (4.1); 2.0680 (1.0); 2.0592 (1.0); 2.0472 (1.8); 2.0350 (1.1); 2.0264 (0.9); 2.0141 (0.5); 1.6250 (7.4); 1.6076 (7.4); 1.2484 (0.4); 1.2321 (0.8); 1.2163 (1.8); 1.2039 (2.9); 1.1949 (2.8); 1.1853 (1.8); 1.1744 (0.7); 1.1625 (0.5); 1.1497 (0.6); 1.1364 (0.7); 1.1266 (1.0); 1.1138 (2.8); 1.1068 (2.3); 1.0941 (2.6); 1.0890 (1.9); 1.0854 (2.0); 1.0727 (0.4); 1.0053 (0.4); 0.9919 (2.3); 0.9862 (3.5); 0.9712 (2.3); 0.9654 (3.3); 0.9542 (0.7);

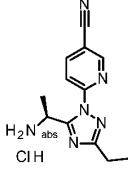
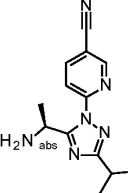
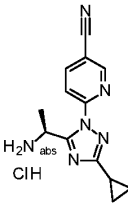
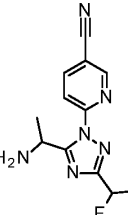
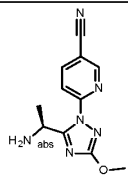
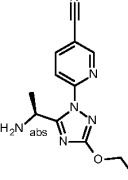
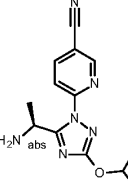
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I-94		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5627 (1.4); 9.5454 (1.4); 8.6004 (2.5); 8.5951 (2.5); 8.5941 (2.4); 8.4913 (2.3); 8.3696 (2.2); 8.2293 (2.2); 8.1878 (1.8); 8.1813 (1.7); 8.1659 (2.0); 8.1594 (1.9); 7.8311 (2.6); 7.8091 (2.4); 7.3734 (1.0); 7.2352 (2.2); 7.0972 (1.1); 6.0191 (1.0); 6.0019 (1.5); 5.9843 (1.0); 5.7558 (3.7); 3.9431 (16.0); 3.3260 (73.5); 3.0132 (0.6); 3.0055 (0.7); 3.0007 (0.4); 2.9939 (1.3); 2.9817 (0.7); 2.9741 (0.7); 2.6761 (0.4); 2.6717 (0.6); 2.6672 (0.4); 2.5249 (2.2); 2.5115 (34.8); 2.5072 (68.7); 2.5027 (89.2); 2.4981 (64.3); 2.4937 (31.8); 2.3341 (0.4); 2.3295 (0.6); 2.3249 (0.4); 1.6348 (5.3); 1.6174 (5.2); 1.2025 (0.8); 1.1902 (1.8); 1.1795 (1.7); 1.1737 (1.0); 1.1700 (1.2); 1.1618 (0.5); 1.1488 (0.3); 1.1199 (0.6); 1.1074 (1.7); 1.1001 (1.4); 1.0877 (1.6); 1.0820 (1.2); 0.0079 (0.6); -0.0002 (14.9); -0.0085 (0.6)	
I-95		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6518 (3.9); 9.6343 (4.0); 9.0101 (15.6); 8.9980 (16.0); 8.5867 (7.2); 8.4469 (6.7); 8.3964 (6.8); 8.3160 (0.6); 7.6781 (4.3); 7.6660 (8.1); 7.6538 (4.0); 6.0714 (0.6); 6.0539 (2.5); 6.0366 (4.0); 6.0193 (2.5); 6.0017 (0.6); 5.7562 (0.7); 3.3710 (37.9); 3.3271 (350.6); 2.6753 (1.8); 2.6713 (2.4); 2.5066 (292.6); 2.5025 (378.8); 2.4982 (281.7); 2.3294 (2.2); 2.0748 (1.3); 1.6808 (14.2); 1.6634 (14.2); 0.1457 (1.2); -0.0001 (252.7); -0.0082 (12.0); -0.1498 (1.2)	
I-96		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6558 (3.5); 9.6385 (3.5); 9.0145 (15.8); 9.0023 (16.0); 8.5901 (6.8); 8.4508 (6.3); 8.3986 (6.2); 8.3160 (0.3); 7.6831 (4.3); 7.6709 (8.1); 7.6587 (4.1); 6.0687 (0.6); 6.0516 (2.5); 6.0343 (4.0); 6.0170 (2.6); 6.0000 (0.5); 3.3725 (36.6); 3.3292 (177.5); 2.6767 (0.9); 2.6723 (1.2); 2.6681 (0.9); 2.5253 (4.3); 2.5119 (79.1); 2.5078 (152.1); 2.5034 (193.6); 2.4990 (139.6); 2.3348 (0.9); 2.3302 (1.2); 2.3258 (0.8); 2.0753 (2.8); 1.6878 (14.1); 1.6704 (14.0); 0.1459 (0.7); 0.0075 (6.5); -0.0004 (135.5); -0.0077 (5.9); -0.1495 (0.7)	

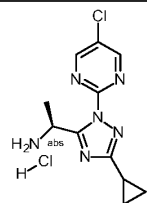
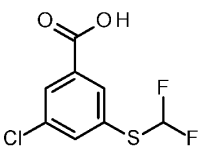
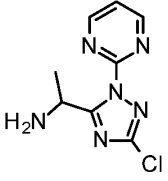
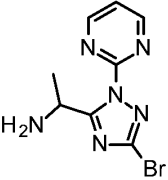
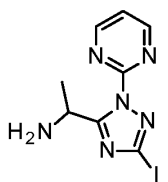
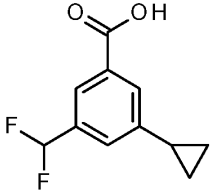
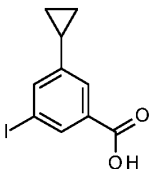
¹⁾ 'lowT' denotes that the measurement was conducted at a temperature of 260 Kelvin.

²⁾ 'abs' denotes that the compound was obtained in an enantiomerically enriched or pure form with the major stereoisomer having the absolute configuration depicted in the drawing.

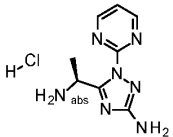
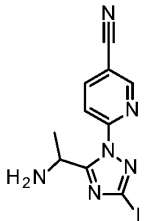
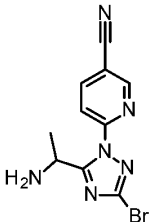
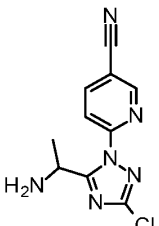
5 ³⁾ The stated mass corresponds to the peak from the isotope pattern of the [M+H]⁺ ion with the highest intensity. # denotes that the [M-H]⁻ ion was recorded.

Table 2 (Intermediates)

Example	Structure ²⁾	NMR data ¹⁾	ESI Mass (m/z) ³⁾
INT-1			243.2 [amine+H] ⁺
INT-2			257.2 [M+H] ⁺
INT-3			255.1 [amine+H] ⁺
INT-4			265.2 [M+H] ⁺
INT-5			245.1 [M+H] ⁺
INT-6			259.3 [M+H] ⁺
INT-7			273.1 [M+H] ⁺

INT-8			265.3 [amine+H] ⁺
INT-9		¹ H NMR (400 MHz, CDCl ₃) δ = 6.90 (t, 1H, J = 74.4 Hz), 7.83 (t, 1H, J = 2 Hz), 8.14 (t, 1H, J = 2 Hz), 8.20 (s, 1H), 10.50 (brs, 1H). (measured on a Varian Gemini 2000 machine).	
INT-10			225.1 [M+H] ⁺
INT-11			271.1 [M+H] ⁺
INT-12			271.1 [M+H] ⁺
INT-13		¹ H-NMR (400 MHz, DMSO-d ₆): δ = 13.12 - 13.40 (broad s, 1 H), 7.87 (s, 1 H), 7.78 (s, 1 H), 7.51 (s, 1 H), 7.20 - 7.06 (t, 1 H), 2.14 - 2.08 (m, 1 H), 1.07 - 1.00 (m, 2H), 0.76 - 0.74 (m, 2H).	
INT-14		¹ H-NMR (400 MHz, DMSO-d ₆): δ = 13.3 (broad s, 1 H), 7.98 (s, 1 H), 7.68 (s, 1 H), 7.60 (s, 1 H), 2.08 - 1.96 (m, 1 H), 1.01 - 0.95 (m, 2H), 0.75 - 0.69 (m, 2H).	287.1 [M-H] ⁻

INT-15		¹ H-NMR (400 MHz, DMSO-d ₆): δ = 13.5 (broad s, 1 H), 8.03 (t, <i>J</i> = 1.6 Hz), 7.93 (t, <i>J</i> = 1.6 Hz), 7.79 (t, <i>J</i> = 1.6 Hz), 2.15 - 2.05 (m, 1 H), 1.10 - 1.00 (m, 2H), 0.85 - 0.75 (m, 2H).	186.1 [M-H] ⁻
INT-16		¹ H-NMR (400 MHz, CD ₃ OD) δ = 8.46 (s, 1H), 8.39 (s, 1H), 8.07 (s, 1H), 6.85 (t, 1H, <i>J</i> = 52.7 Hz). (measured on a Varian Gemini 2000 machine).	
INT-17		¹ H-NMR (400 MHz, DMSO-d ₆): δ = 13.5 (broad s, 1 H), 8.16 (t, <i>J</i> = 1.6 Hz, 1H), 7.92 (t, <i>J</i> = 1.6 Hz, 1H), 7.85 (t, <i>J</i> = 1.6 Hz, 1H), 3.27 (s, 3H), 2.23 - 2.15 (m, 1 H), 1.12 - 1.02 (m, 2H), 0.90 - 0.80 (m, 2H).	239.0 [M-H] ⁻
INT-18		¹ H NMR (DMSO-d ₆ , 400 MHz): δ = 8.61 (d, <i>J</i> = 2.8 Hz, 1H), 8.15 (d, <i>J</i> = 2.8, 8.8 Hz, 1H), 7.78 (d, <i>J</i> = 8.8 Hz, 1H), 4.69 - 4.60 (m, 1H), 3.94 (s, 3H), 2.07 (br s, 2H), 1.36 (d, <i>J</i> = 6.8 Hz, 3H).	254.1 [M+H] ⁺
INT-19		¹ H-NMR (400 MHz, DMSO-d ₆): δ = 13.51 (s, 1H), 8.10 (t, <i>J</i> = 1.6 Hz, 1H), 7.90 (t, <i>J</i> = 1.6 Hz, 1H), 7.83 (t, <i>J</i> = 1.6 Hz, 1H), 3.02 - 2.95 (m, 1H), 2.25 - 2.15 (m, 1H), 1.18 - 1.02 (m, 6H), 0.85 - 0.78 (m, 2H).	267.1 [M+H] ⁺
INT-20		¹ H NMR (methanol-d ₄ , 400 MHz): δ = 8.55 - 8.50 (m, 1H), 8.09 (dd, <i>J</i> = 2.6, 8.8 Hz, 1H), 7.97 (d, <i>J</i> = 8.8 Hz, 1H), 5.41 (q, <i>J</i> = 6.8 Hz, 1H), 2.16 - 2.07 (m, 1H), 1.72 (d, <i>J</i> = 6.8 Hz, 3H), 1.07 - 1.00 (m, 4H). Measured using a Varian 400MR NMR machine.	264.1 [amine+H] ⁺
INT-21		¹ H NMR (DMSO-d ₆ , 400 MHz): δ = 13.9 (br s, 1H), 8.25 (t, <i>J</i> = 1.4 Hz, 1H), 8.12 - 8.00 (m, 2H). (Signal of CH ₃ -group is hidden under solvent signal)	217.0 [M-H] ⁻
INT-22		¹ H NMR (DMSO-d ₆ , 600 MHz): δ = 9.05 (s, 2H), 4.67 - 4.63 (m, 1H), 3.94 (s, 3H), 2.03 (br s, 2H), 1.37 (d, <i>J</i> = 6.6 Hz, 3H).	255.1 [M+H] ⁺

INT-23			206.1 [amine+H] ⁺
INT-24			341.1 [M+H] ⁺
INT-25			295.0 [M+H] ⁺
INT-26			249.2 [M+H] ⁺

¹⁾ 'lowT' denotes that the measurement was conducted at a temperature of 260 Kelvin.

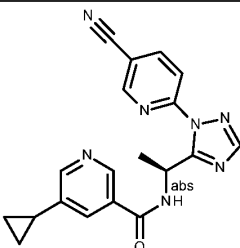
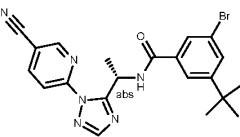
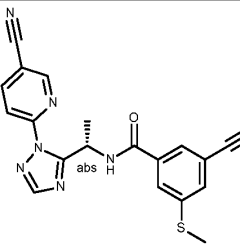
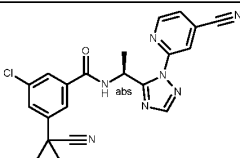
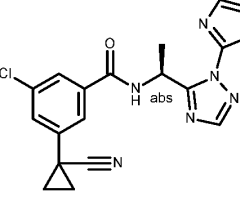
²⁾ 'abs' denotes that the compound was obtained in an enantiomerically enriched or pure form with the major stereoisomer having the absolute configuration depicted in the drawing.

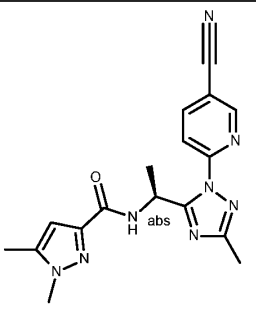
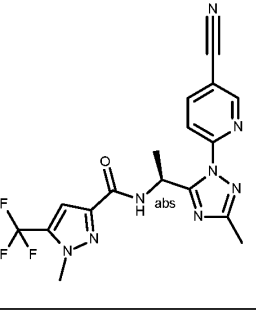
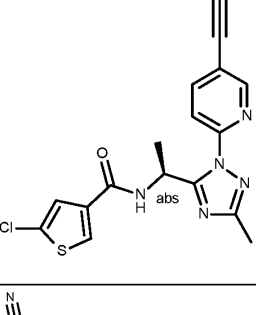
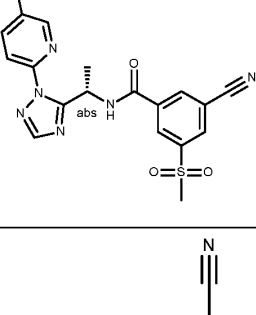
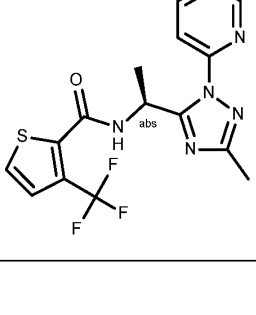
5 ³⁾ The stated mass corresponds to the peak from the isotope pattern of the [M+H]⁺ ion with the highest intensity. # denotes that the [M-H]⁻ ion was recorded.

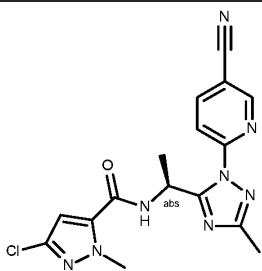
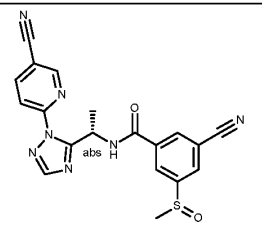
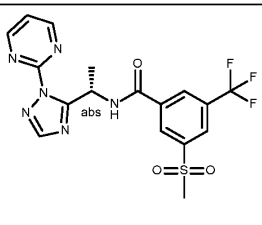
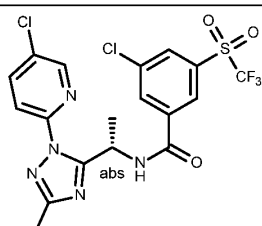
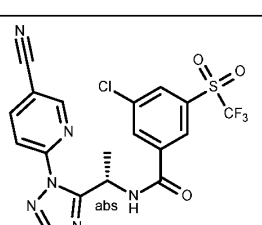
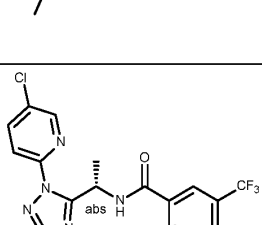
Additionally, the present invention provides the following compounds:

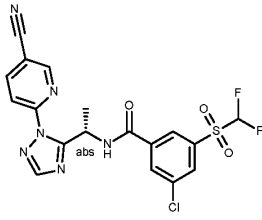
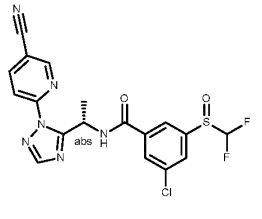
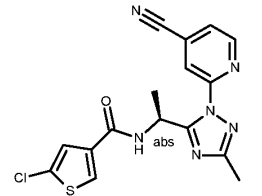
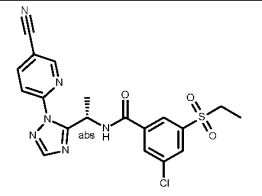
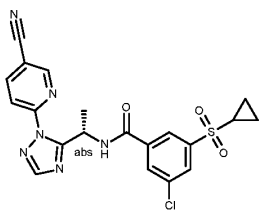
Table 3

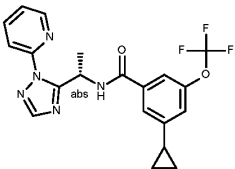
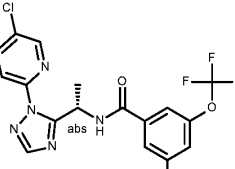
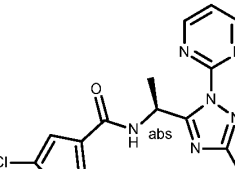
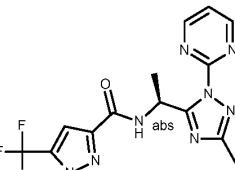
Example	Structure ²⁾	NMR Peaklist ¹⁾
II-1		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.4234 (0.6); 9.4048 (0.6); 9.2061 (3.3); 9.1887 (3.4); 9.1514 (1.2); 9.1459 (1.3); 9.0845 (1.1); 9.0791 (1.1); 9.0662 (5.8); 9.0646 (6.2); 9.0608 (6.2); 9.0592 (5.9); 8.9925 (1.2); 8.9875 (1.3); 8.6876 (8.2); 8.6825 (8.3); 8.5857 (5.7); 8.5802 (5.5); 8.5643 (6.1); 8.5587 (6.1);

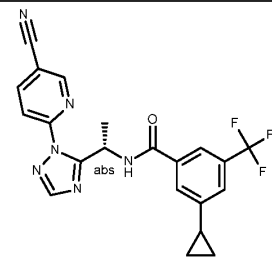
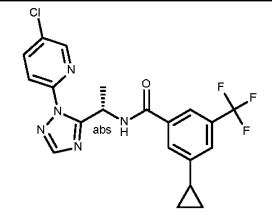
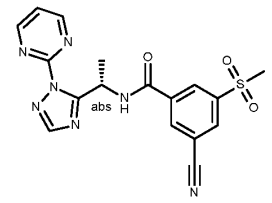
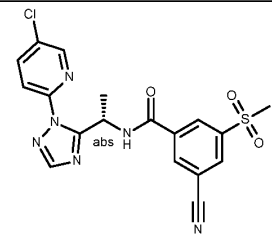
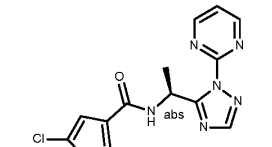
		8.5523 (0.9); 8.5469 (1.4); 8.5389 (7.7); 8.5335 (7.6); 8.3157 (1.7); 8.2625 (2.7); 8.2392 (16.0); 8.0991 (1.1); 8.0809 (6.7); 8.0793 (7.4); 8.0595 (6.0); 8.0579 (6.0); 7.7031 (4.1); 7.6978 (7.6); 7.6925 (4.1); 6.1649 (0.4); 6.1476 (0.7); 6.1304 (0.4); 6.1062 (0.6); 6.0890 (2.6); 6.0717 (4.1); 6.0542 (2.6); 6.0367 (0.5); 3.3236 (302.9); 2.6798 (1.4); 2.6754 (3.0); 2.6708 (4.2); 2.6663 (3.0); 2.6617 (1.4); 2.5243 (11.6); 2.5196 (17.6); 2.5109 (248.1); 2.5064 (509.7); 2.5019 (677.7); 2.4973 (494.4); 2.4928 (241.7); 2.3958 (0.4); 2.3377 (1.6); 2.3332 (3.2); 2.3287 (4.4); 2.3241 (3.2); 2.3197 (1.6); 2.0389 (0.8); 2.0265 (1.6); 2.0182 (1.7); 2.0056 (3.3); 1.9931 (1.9); 1.9849 (1.8); 1.9719 (0.9); 1.6776 (2.4); 1.6602 (2.5); 1.6378 (15.8); 1.6204 (15.8); 1.4562 (0.3); 1.0549 (2.0); 1.0441 (4.9); 1.0385 (5.5); 1.0342 (2.9); 1.0282 (3.0); 1.0230 (5.2); 1.0175 (5.3); 1.0073 (2.3); 0.9905 (0.4); 0.8022 (2.4); 0.7917 (5.8); 0.7866 (5.9); 0.7793 (5.5); 0.7741 (6.2); 0.7629 (1.9); -0.0002 (5.8)
II-2		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1841 (0.4); 9.1669 (0.5); 9.0574 (0.8); 9.0521 (0.8); 8.5813 (0.6); 8.5757 (0.6); 8.5599 (0.6); 8.5543 (0.6); 8.2349 (1.9); 8.0742 (0.8); 8.0527 (0.8); 7.8119 (0.6); 7.8079 (1.0); 7.8041 (0.7); 7.7556 (0.6); 7.7518 (1.0); 7.7482 (0.7); 7.6872 (0.6); 7.6829 (1.0); 7.6787 (0.6); 6.0668 (0.3); 6.0496 (0.5); 3.3187 (43.7); 2.5239 (1.2); 2.5105 (19.4); 2.5062 (37.6); 2.5016 (49.2); 2.4971 (36.7); 2.4928 (18.7); 1.6399 (1.9); 1.6224 (1.8); 1.3978 (8.4); 1.2749 (16.0); 0.0079 (1.1); -0.0002 (23.9); -0.0084 (1.0)
II-3		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.3035 (1.2); 9.2861 (1.2); 9.0680 (2.1); 9.0663 (2.2); 9.0626 (2.3); 9.0608 (2.1); 8.5863 (1.8); 8.5807 (1.7); 8.5648 (1.9); 8.5592 (1.9); 8.2424 (5.6); 8.0866 (2.3); 8.0849 (2.2); 8.0652 (2.1); 8.0634 (2.1); 7.9733 (1.8); 7.9698 (3.3); 7.9662 (2.0); 7.9102 (1.6); 7.9058 (2.9); 7.9018 (1.9); 7.8799 (2.1); 7.8761 (2.6); 7.8719 (1.5); 6.0943 (0.9); 6.0770 (1.4); 6.0595 (0.9); 5.7555 (1.3); 3.3243 (9.9); 2.5642 (16.0); 2.5264 (0.6); 2.5216 (0.9); 2.5129 (11.3); 2.5085 (22.4); 2.5040 (29.0); 2.4994 (21.0); 2.4949 (10.1); 1.6368 (5.2); 1.6194 (5.2); 0.0079 (0.9); -0.0002 (25.2); -0.0086 (0.8)
II-4		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2475 (1.2); 9.2298 (1.3); 8.8095 (2.2); 8.8079 (2.2); 8.7969 (2.2); 8.7953 (2.3); 8.3130 (3.2); 8.2305 (6.0); 7.9681 (1.9); 7.9648 (1.8); 7.9555 (1.8); 7.9521 (1.8); 7.7684 (1.8); 7.7643 (2.9); 7.7602 (1.9); 7.6689 (1.8); 7.6650 (3.1); 7.6611 (1.8); 7.5194 (1.9); 7.5148 (3.4); 7.5102 (1.7); 6.0161 (0.9); 5.9987 (1.5); 5.9813 (1.0); 5.7582 (16.0); 4.0579 (0.4); 4.0401 (1.2); 4.0223 (1.2); 4.0045 (0.4); 3.5702 (0.4); 3.3290 (7.0); 3.1795 (0.4); 3.1664 (0.3); 2.5231 (0.4); 2.5147 (5.7); 2.5103 (11.6); 2.5057 (15.3); 2.5011 (11.0); 2.4966 (5.2); 2.0784 (0.5); 1.9913 (4.9); 1.8167 (1.0); 1.8041 (3.0); 1.7966 (3.2); 1.7851 (1.4); 1.6397 (5.6); 1.6295 (2.2); 1.6222 (6.2); 1.6102 (3.2); 1.5969 (1.1); 1.3961 (1.2); 1.1944 (1.4); 1.1767 (2.7); 1.1589 (1.3)
II-5		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.3333 (2.3); 9.3164 (2.3); 8.1981 (10.7); 7.8639 (3.2); 7.8599 (5.3); 7.8558 (3.5); 7.7998 (5.2); 7.7910 (8.2); 7.7621 (8.3); 7.7533 (5.6); 7.7491 (3.6); 7.7451 (5.7); 7.7413 (3.4); 7.5425 (3.3); 7.5379 (6.0); 7.5334 (3.2); 6.0601 (0.4); 6.0432 (1.7); 6.0261 (2.7); 6.0088 (1.7); 5.9913 (0.4); 4.0557 (0.8); 4.0379 (2.4); 4.0201 (2.5); 4.0023 (0.8); 3.3215 (87.6); 2.6794 (0.4); 2.6752 (0.9); 2.6707 (1.3); 2.6662 (1.0); 2.6618 (0.5); 2.5241 (3.5); 2.5194 (5.2); 2.5106 (77.7); 2.5062 (160.3); 2.5017 (213.1); 2.4971 (155.6); 2.4927 (76.1); 2.3330 (0.9); 2.3285 (1.3); 2.3240 (0.9); 2.0748 (0.4); 1.9890 (10.8); 1.8230 (1.9); 1.8099 (5.6); 1.8025 (6.0); 1.7909 (2.6); 1.7520 (0.4); 1.6856 (0.4); 1.6468 (2.7); 1.6345 (6.4); 1.6267 (16.0); 1.6090 (10.8); 1.3975 (15.3); 1.1924 (2.9); 1.1746 (5.8); 1.1568 (2.8); -0.0002 (3.7)

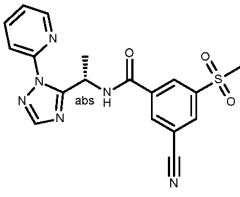
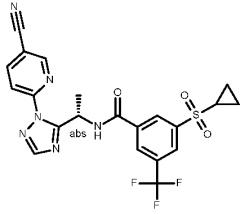
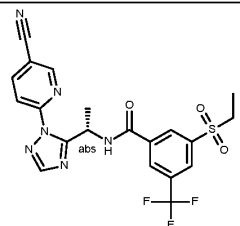
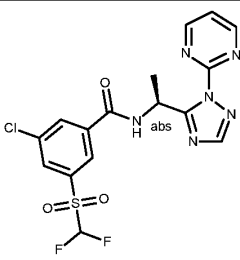
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II-7		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0545 (2.2); 9.0527 (2.4); 9.0490 (2.4); 9.0471 (2.2); 8.7683 (1.2); 8.7496 (1.2); 8.5427 (2.0); 8.5371 (1.9); 8.5212 (2.1); 8.5156 (2.1); 8.0207 (2.4); 8.0189 (2.4); 7.9991 (2.3); 7.9973 (2.2); 7.2552 (3.7); 6.0736 (1.0); 6.0557 (1.4); 6.0377 (1.0); 4.0321 (9.4); 4.0198 (0.6); 3.3217 (157.4); 2.6794 (0.6); 2.6748 (1.3); 2.6703 (1.8); 2.6657 (1.3); 2.6611 (0.6); 2.5238 (5.0); 2.5191 (7.6); 2.5104 (107.9); 2.5059 (220.8); 2.5013 (289.7); 2.4967 (204.7); 2.4922 (95.4); 2.3431 (16.0); 2.3328 (1.5); 2.3282 (1.8); 2.3236 (1.3); 2.3192 (0.6); 1.9887 (2.2); 1.5718 (6.0); 1.5545 (5.8); 1.3976 (4.4); 1.1922 (0.6); 1.1744 (1.2); 1.1566 (0.6); 0.0080 (0.6); -0.0002 (19.8); -0.0085 (0.6)
II-8		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0300 (2.2); 9.0282 (2.4); 9.0245 (2.4); 9.0227 (2.2); 8.9118 (1.2); 8.8938 (1.2); 8.5391 (1.9); 8.5336 (1.8); 8.5176 (2.0); 8.5120 (2.0); 8.0475 (3.8); 8.0431 (3.9); 8.0109 (2.4); 8.0091 (2.4); 7.9894 (2.2); 7.9876 (2.2); 7.4945 (3.9); 7.4902 (3.8); 6.0327 (1.0); 6.0151 (1.6); 5.9974 (1.0); 4.0556 (0.4); 4.0378 (1.4); 4.0200 (1.4); 4.0022 (0.5); 3.3217 (40.4); 2.6751 (0.5); 2.6706 (0.6); 2.6661 (0.5); 2.5241 (2.0); 2.5193 (3.1); 2.5107 (37.8); 2.5062 (76.2); 2.5016 (99.6); 2.4970 (71.1); 2.4925 (33.6); 2.3508 (0.6); 2.3357 (16.0); 1.9889 (6.3); 1.5783 (5.6); 1.5608 (5.6); 1.3975 (2.1); 1.1923 (1.7); 1.1745 (3.4); 1.1568 (1.7); 0.0080 (0.5); -0.0002 (15.5); -0.0085 (0.4)
II-9		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5704 (1.8); 9.5530 (1.9); 9.0794 (3.2); 9.0777 (3.4); 9.0739 (3.5); 9.0722 (3.2); 8.5906 (4.3); 8.5844 (11.9); 8.5826 (16.0); 8.5691 (3.0); 8.5636 (3.0); 8.2610 (8.8); 8.0971 (3.6); 8.0954 (3.6); 8.0757 (3.3); 8.0738 (3.3); 6.1479 (1.3); 6.1305 (2.1); 6.1133 (1.3); 3.3452 (22.3); 3.3232 (45.2); 2.6762 (0.4); 2.6717 (0.5); 2.6671 (0.3); 2.5253 (1.6); 2.5205 (2.4); 2.5118 (30.3); 2.5073 (61.2); 2.5027 (79.9); 2.4981 (56.7); 2.4936 (26.7); 2.3341 (0.4); 2.3296 (0.5); 2.3249 (0.4); 1.6610 (7.9); 1.6437 (7.8); -0.0002 (5.5)
II-10		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2836 (1.3); 9.2655 (1.3); 9.0072 (2.3); 9.0031 (2.2); 8.5531 (1.7); 8.5475 (1.6); 8.5315 (1.8); 8.5260 (1.8); 8.3149 (0.4); 8.0248 (2.4); 8.0033 (2.2); 7.8126 (1.9); 7.7998 (2.0); 7.3601 (3.2); 7.3471 (3.0); 6.0148 (1.0); 5.9973 (1.6); 5.9797 (1.0); 4.0379 (0.4); 4.0201 (0.4); 3.3198 (61.4); 2.6795 (0.4); 2.6750 (1.0); 2.6706 (1.3); 2.6661 (1.0); 2.6615 (0.5); 2.5240 (3.9); 2.5192 (5.8); 2.5105 (79.1); 2.5061 (161.7); 2.5016 (213.3); 2.4970 (156.1); 2.4927 (77.3); 2.3577 (16.0); 2.3374 (0.5); 2.3329 (1.0); 2.3284 (1.4); 2.3238 (1.0); 1.9886 (1.7); 1.5643 (5.9); 1.5470 (5.9); 1.1927 (0.5); 1.1750 (0.9); 1.1572 (0.4); -0.0001 (9.4); -0.0084 (0.3)

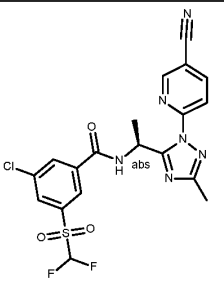
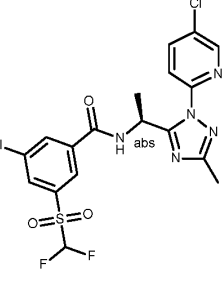
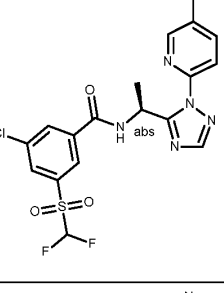
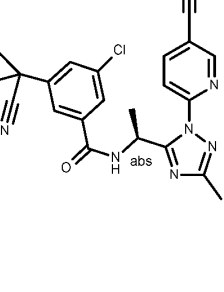
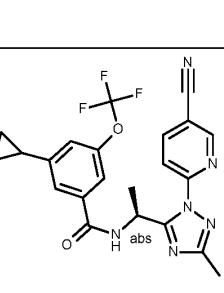
II-11		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1259 (1.2); 9.1079 (1.2); 9.0290 (1.9); 9.0274 (2.1); 9.0236 (2.1); 9.0220 (2.0); 8.5433 (1.7); 8.5378 (1.6); 8.5218 (1.8); 8.5162 (1.8); 8.0271 (2.2); 8.0054 (2.0); 6.9764 (6.6); 6.0616 (0.9); 6.0440 (1.4); 6.0263 (0.9); 3.9303 (16.0); 3.3243 (134.9); 2.6755 (0.5); 2.6711 (0.7); 2.6665 (0.6); 2.5412 (0.4); 2.5245 (2.4); 2.5197 (3.5); 2.5110 (44.3); 2.5066 (90.4); 2.5021 (119.5); 2.4975 (87.2); 2.4931 (43.1); 2.3489 (13.8); 2.3336 (0.6); 2.3289 (0.8); 2.3245 (0.6); 1.9887 (0.4); 1.5764 (5.2); 1.5590 (5.2); -0.0002 (5.0)
II-12		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4717 (0.9); 9.4558 (1.0); 9.0718 (2.7); 9.0668 (2.8); 8.5866 (2.0); 8.5811 (2.0); 8.5651 (2.1); 8.5596 (2.1); 8.3951 (5.0); 8.3914 (6.2); 8.3788 (1.4); 8.3747 (1.9); 8.3710 (1.1); 8.3149 (9.3); 8.3041 (2.5); 8.3006 (3.8); 8.2545 (6.1); 8.0891 (2.8); 8.0676 (2.6); 6.1290 (0.7); 6.1133 (1.1); 6.0960 (0.8); 3.3214 (13.4); 3.2975 (3.4); 2.8410 (16.0); 2.6755 (0.5); 2.6713 (0.6); 2.6665 (0.5); 2.5108 (38.5); 2.5067 (73.3); 2.5023 (95.9); 2.4978 (74.4); 2.3334 (0.4); 2.3290 (0.6); 2.3246 (0.4); 1.6535 (6.6); 1.6362 (6.6); -0.0002 (5.4)
II-13		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5894 (2.7); 9.5715 (2.7); 8.9957 (15.8); 8.9836 (16.0); 8.5693 (5.6); 8.4259 (5.0); 8.3789 (4.9); 8.3091 (0.7); 8.1777 (12.6); 7.6373 (4.3); 7.6252 (8.2); 7.6130 (4.2); 6.0783 (0.5); 6.0611 (2.1); 6.0436 (3.3); 6.0259 (2.1); 6.0085 (0.5); 3.3956 (0.4); 3.3633 (34.0); 3.3029 (142.0); 2.6741 (1.5); 2.6695 (2.1); 2.6650 (1.5); 2.6604 (0.7); 2.5229 (6.7); 2.5181 (10.6); 2.5096 (125.4); 2.5051 (250.0); 2.5006 (327.6); 2.4960 (235.5); 2.4915 (114.4); 2.3362 (0.7); 2.3320 (1.5); 2.3274 (2.0); 2.3229 (1.5); 2.3184 (0.7); 1.6764 (12.9); 1.6590 (12.8); -0.0002 (5.2)
II-14		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6061 (1.2); 9.5884 (1.2); 8.5926 (2.3); 8.5912 (2.4); 8.5862 (2.5); 8.5847 (2.3); 8.5026 (1.7); 8.4984 (2.7); 8.4940 (2.0); 8.4428 (2.4); 8.4006 (1.4); 8.3965 (2.2); 8.1731 (1.9); 8.1666 (1.8); 8.1512 (2.2); 8.1447 (2.2); 7.8461 (2.6); 7.8448 (2.5); 7.8243 (2.4); 7.8229 (2.3); 6.0232 (0.9); 6.0056 (1.5); 5.9881 (1.0); 3.3187 (52.4); 2.6717 (0.4); 2.5253 (1.2); 2.5205 (1.8); 2.5119 (24.3); 2.5074 (49.7); 2.5028 (65.3); 2.4981 (46.1); 2.4935 (21.6); 2.3326 (16.0); 1.6295 (5.2); 1.6121 (5.2); 1.3978 (10.8); 0.0081 (1.2); -0.0002 (39.5); -0.0085 (1.2)
II-15		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6573 (1.3); 9.6399 (1.3); 9.0264 (2.3); 9.0246 (2.5); 9.0209 (2.6); 9.0190 (2.3); 8.5431 (2.1); 8.5375 (2.1); 8.5336 (2.0); 8.5293 (3.0); 8.5250 (2.3); 8.5216 (2.4); 8.5160 (2.2); 8.4658 (2.6); 8.4085 (1.5); 8.4045 (2.4); 8.0289 (2.5); 8.0271 (2.5); 8.0074 (2.4); 8.0055 (2.3); 6.1350 (1.0); 6.1176 (1.6); 6.1001 (1.0); 4.0567 (0.4); 4.0389 (1.1); 4.0211 (1.1); 4.0033 (0.4); 3.3196 (34.5); 2.6720 (0.4); 2.5255 (1.2); 2.5208 (1.7); 2.5122 (22.8); 2.5077 (46.6); 2.5031 (61.4); 2.4985 (44.2); 2.4939 (21.2); 2.3515 (16.0); 2.3347 (0.4); 2.3300 (0.4); 1.9893 (5.2); 1.6408 (5.5); 1.6235 (5.5); 1.3976 (1.3); 1.1937 (1.4); 1.1759 (2.8); 1.1581 (1.4); 0.0080 (1.2); -0.0002 (36.1); -0.0085 (1.2)
II-16		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6054 (2.7); 9.5876 (2.8); 8.6439 (5.4); 8.6378 (5.6); 8.6054 (5.8); 8.4494 (5.2); 8.3914 (5.1); 8.3150 (0.4); 8.2172 (3.7); 8.2107 (3.6); 8.1953 (4.6); 8.1896 (16.0); 7.9007 (6.0); 7.8789 (5.2); 6.0631 (0.4); 6.0457 (2.0); 6.0283 (3.2); 6.0109 (2.1); 5.9936 (0.4); 3.4001 (0.4); 3.3729 (32.4); 3.3211 (34.4); 2.6803 (0.4); 2.6758 (0.9); 2.6713 (1.2); 2.6668 (0.9); 2.5248 (3.6); 2.5200 (5.7); 2.5113 (74.6); 2.5069 (151.2); 2.5024 (198.4); 2.4978 (142.9); 2.4934 (69.1); 2.3337 (0.8); 2.3292 (1.2); 2.3246 (0.9); 1.9890 (0.8); 1.6633 (12.4); 1.6459 (12.4); 1.2356 (0.3); 1.1755 (0.5); 0.1459 (0.4); 0.0080 (3.4); -0.0002 (100.9); -0.0084 (3.5); -0.1495 (0.4)

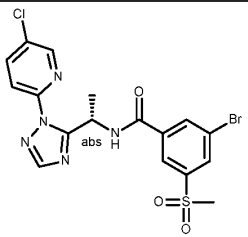
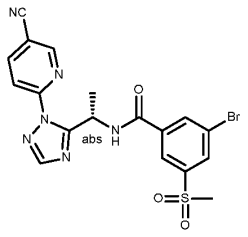
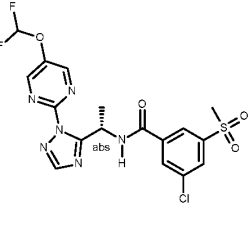
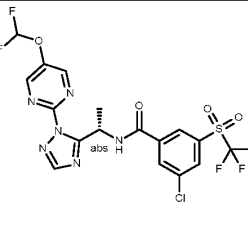
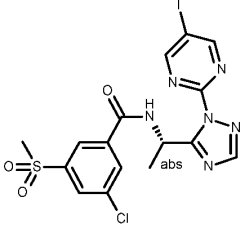
II-17		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6271 (3.3); 9.6099 (3.4); 9.0657 (5.7); 9.0639 (6.4); 9.0603 (6.3); 9.0584 (6.1); 8.5866 (5.2); 8.5810 (5.0); 8.5651 (5.5); 8.5595 (5.5); 8.4298 (4.8); 8.4253 (7.4); 8.4212 (5.3); 8.3310 (4.6); 8.3273 (8.3); 8.3235 (4.7); 8.3156 (0.9); 8.2587 (16.0); 8.1824 (4.4); 8.1780 (7.4); 8.1737 (4.2); 8.0919 (6.4); 8.0901 (6.6); 8.0705 (6.0); 8.0686 (6.2); 7.5412 (2.8); 7.4113 (6.6); 7.2814 (3.4); 6.1441 (0.5); 6.1269 (2.4); 6.1096 (3.8); 6.0922 (2.4); 6.0751 (0.5); 5.7560 (9.3); 3.3219 (55.2); 2.6804 (0.4); 2.6760 (0.9); 2.6714 (1.2); 2.6668 (0.9); 2.6622 (0.4); 2.5250 (3.5); 2.5202 (5.2); 2.5116 (70.9); 2.5071 (146.8); 2.5025 (195.0); 2.4979 (139.9); 2.4933 (66.6); 2.3386 (0.4); 2.3339 (0.8); 2.3293 (1.2); 2.3247 (0.8); 2.3201 (0.4); 1.6562 (14.3); 1.6389 (14.3); 1.2345 (0.6); 0.1458 (0.4); 0.0079 (2.9); -0.0003 (97.6); -0.0086 (3.2); -0.1498 (0.4)
II-18		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4542 (3.0); 9.4368 (3.1); 9.0632 (5.5); 9.0583 (5.5); 8.5851 (5.3); 8.5795 (5.0); 8.5636 (5.7); 8.5580 (5.7); 8.3148 (0.5); 8.2495 (16.0); 8.1572 (2.2); 8.1536 (5.5); 8.1490 (6.4); 8.1457 (6.3); 8.1418 (5.0); 8.1359 (4.4); 8.0872 (6.4); 8.0854 (6.4); 8.0658 (5.9); 8.0639 (6.0); 7.9844 (5.3); 7.9808 (5.0); 7.1913 (2.5); 7.0582 (3.9); 6.9236 (3.0); 6.1271 (0.5); 6.1101 (2.3); 6.0928 (3.7); 6.0754 (2.3); 6.0579 (0.5); 5.7552 (3.6); 3.3237 (138.0); 2.6806 (0.4); 2.6761 (0.8); 2.6715 (1.1); 2.6669 (0.8); 2.6625 (0.4); 2.5250 (3.1); 2.5203 (4.6); 2.5116 (64.8); 2.5071 (134.4); 2.5025 (178.0); 2.4979 (126.4); 2.4933 (59.0); 2.3384 (0.3); 2.3340 (0.8); 2.3293 (1.1); 2.3248 (0.8); 2.3203 (0.3); 1.6466 (13.5); 1.6292 (13.4); 1.2343 (0.7); 0.1460 (0.3); 0.0080 (2.8); -0.0002 (95.3); -0.0085 (3.0); -0.1497 (0.3)
II-19		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 8.8838 (1.3); 8.8657 (1.3); 8.7827 (2.2); 8.7809 (2.3); 8.7701 (2.3); 8.7683 (2.4); 8.2275 (2.4); 8.2246 (3.3); 8.2223 (2.5); 8.0270 (3.8); 8.0227 (3.9); 7.9243 (2.2); 7.9208 (2.1); 7.9116 (2.1); 7.9082 (2.0); 7.4729 (3.9); 7.4686 (3.8); 5.9633 (1.0); 5.9456 (1.6); 5.9278 (1.0); 5.7555 (0.4); 4.0382 (0.6); 4.0204 (0.6); 3.3214 (14.2); 2.6713 (0.4); 2.5247 (1.4); 2.5113 (22.6); 2.5069 (43.7); 2.5024 (56.1); 2.4978 (40.3); 2.4933 (19.4); 2.3541 (0.3); 2.3374 (16.0); 1.9890 (2.7); 1.5774 (5.9); 1.5600 (5.8); 1.3976 (3.9); 1.1931 (0.7); 1.1753 (1.4); 1.1575 (0.7); 0.0079 (2.1); -0.0002 (46.7); -0.0085 (1.6)
II-20		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5106 (2.3); 9.4933 (2.3); 9.0698 (4.1); 9.0657 (4.1); 9.0644 (3.9); 8.5875 (3.2); 8.5820 (3.1); 8.5660 (3.5); 8.5605 (3.5); 8.2563 (11.3); 8.2451 (3.0); 8.2414 (6.3); 8.2376 (4.0); 8.2211 (3.7); 8.2166 (5.3); 8.2126 (3.0); 8.0986 (3.6); 8.0941 (5.7); 8.0897 (7.3); 8.0681 (4.0); 8.0666 (4.0); 6.1389 (0.4); 6.1217 (1.7); 6.1045 (2.7); 6.0871 (1.7); 6.0694 (0.4); 5.7559 (4.6); 3.4496 (1.8); 3.4312 (6.3); 3.4128 (6.4); 3.3945 (1.9); 3.3214 (30.7); 2.6760 (0.7); 2.6714 (0.9); 2.6668 (0.6); 2.5249 (2.8); 2.5201 (4.2); 2.5115 (52.6); 2.5070 (107.2); 2.5024 (140.8); 2.4978 (100.5); 2.4933 (47.6); 2.3338 (0.6); 2.3293 (0.8); 2.3247 (0.6); 1.6535 (10.2); 1.6362 (10.1); 1.2342 (0.5); 1.1320 (7.0); 1.1137 (16.0); 1.0953 (6.8); 0.1459 (0.5); 0.0080 (4.1); -0.0002 (123.1); -0.0085 (3.9); -0.1496 (0.5)
II-21		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5200 (2.6); 9.5028 (2.6); 9.0726 (4.6); 9.0685 (4.8); 9.0672 (4.6); 8.5895 (3.4); 8.5840 (3.5); 8.5680 (3.6); 8.5625 (3.8); 8.2561 (15.6); 8.2511 (4.4); 8.2317 (0.9); 8.2131 (3.6); 8.2087 (5.6); 8.2046 (3.5); 8.1233 (3.8); 8.1190 (6.0); 8.1146 (3.4); 8.0923 (4.7); 8.0838 (0.5); 8.0709 (4.3); 8.0697 (4.3); 8.0622 (0.4); 7.8541 (0.4); 7.4797 (0.4); 6.1416 (0.4); 6.1245 (1.8); 6.1072 (2.8); 6.0898 (1.8); 6.0727 (0.4); 5.7556 (16.0); 3.3245 (105.7); 3.0445 (0.6); 3.0326 (1.2); 3.0248 (1.3); 3.0131 (2.5); 3.0011 (1.3); 2.9934 (1.3); 2.9813 (0.6); 2.6760 (0.7); 2.6715 (0.9); 2.6672 (0.7); 2.5250 (2.9); 2.5202 (4.4); 2.5114 (57.0); 2.5071 (114.8); 2.5027 (150.1); 2.4981 (108.6); 2.4938 (53.2); 2.3340 (0.6); 2.3295 (0.9); 2.3251 (0.6); 1.6545 (10.8); 1.6371 (10.8); 1.6102 (0.9); 1.2348 (0.5); 1.2210 (0.3); 1.2094 (0.4); 1.1957 (1.6); 1.1877 (3.4); 1.1843 (3.4); 1.1766 (3.5); 1.1677 (2.0); 1.1477 (0.5); 1.1351 (0.6); 1.1217 (0.7); 1.1139 (1.0); 1.1014 (3.3); 1.0943 (2.9); 1.0817

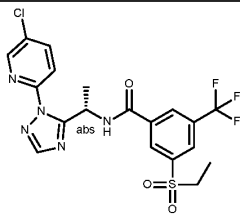
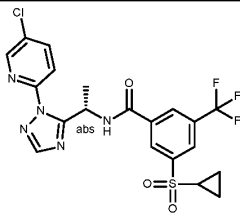
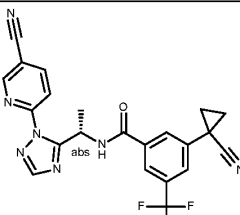
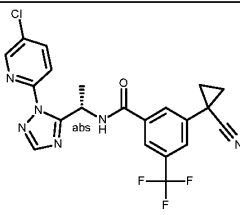
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II-22		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.1336 (3.1); 9.1157 (3.2); 8.5627 (2.9); 8.5608 (3.3); 8.5581 (3.5); 8.5561 (3.1); 8.5506 (3.1); 8.5486 (3.5); 8.5460 (3.4); 8.5440 (3.0); 8.3144 (0.4); 8.1321 (16.0); 8.0967 (2.4); 8.0920 (2.4); 8.0778 (3.2); 8.0763 (3.5); 8.0732 (3.2); 8.0716 (3.4); 8.0575 (3.1); 8.0528 (3.0); 7.8559 (3.5); 7.8538 (6.2); 7.8518 (3.8); 7.8355 (3.1); 7.8334 (5.3); 7.8312 (3.1); 7.4997 (3.0); 7.4973 (3.1); 7.4875 (2.9); 7.4851 (3.2); 7.4810 (3.2); 7.4786 (2.9); 7.4689 (3.2); 7.4664 (3.2); 7.4579 (11.2); 7.4541 (10.4); 7.2591 (4.8); 6.0357 (0.5); 6.0185 (2.5); 6.0009 (3.9); 5.9833 (2.5); 5.9660 (0.5); 3.3180 (69.5); 2.6804 (0.4); 2.6758 (0.8); 2.6712 (1.1); 2.6667 (0.8); 2.6621 (0.4); 2.5248 (3.5); 2.5201 (5.1); 2.5114 (65.6); 2.5069 (134.7); 2.5023 (177.4); 2.4977 (125.8); 2.4931 (59.4); 2.3383 (0.3); 2.3337 (0.8); 2.3291 (1.1); 2.3245 (0.8); 2.3203 (0.3); 2.0585 (0.7); 2.0459 (1.5); 2.0375 (1.6); 2.0336 (1.1); 2.0251 (3.1); 2.0165 (1.1); 2.0124 (1.8); 2.0041 (1.6); 1.9914 (0.9); 1.6350 (15.6); 1.6175 (15.6); 1.2347 (0.5); 1.0423 (2.0); 1.0316 (4.7); 1.0260 (5.2); 1.0216 (2.8); 1.0157 (2.8); 1.0106 (5.0); 1.0050 (4.9); 0.9949 (2.2); 0.7875 (2.4); 0.7771 (5.4); 0.7718 (5.2); 0.7647 (5.2); 0.7597 (5.3); 0.7485 (1.8); 0.0080 (2.4); -0.0002 (76.7); -0.0085 (2.3)
II-23		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.1368 (3.4); 9.1192 (3.5); 8.6193 (6.8); 8.6131 (6.8); 8.2056 (4.6); 8.1991 (4.4); 8.1838 (5.2); 8.1773 (5.2); 8.1637 (16.0); 7.8785 (7.4); 7.8566 (6.5); 7.4511 (11.8); 7.4475 (11.3); 7.2653 (5.2); 5.9764 (0.5); 5.9594 (2.6); 5.9419 (4.0); 5.9244 (2.6); 5.9070 (0.6); 3.3233 (62.4); 2.6805 (0.3); 2.6763 (0.7); 2.6718 (0.9); 2.6672 (0.7); 2.6628 (0.3); 2.5253 (2.6); 2.5204 (4.0); 2.5118 (55.9); 2.5074 (113.7); 2.5029 (149.8); 2.4983 (107.8); 2.4939 (52.1); 2.3343 (0.6); 2.3298 (0.9); 2.3251 (0.7); 2.0606 (0.7); 2.0482 (1.6); 2.0398 (1.7); 2.0274 (3.2); 2.0149 (1.8); 2.0065 (1.7); 1.9938 (0.9); 1.6289 (15.5); 1.6115 (15.4); 1.3977 (4.6); 1.2343 (1.0); 1.2240 (1.0); 1.2172 (1.0); 1.2058 (1.0); 1.1909 (0.3); 1.0455 (2.0); 1.0345 (5.3); 1.0289 (5.7); 1.0185 (3.0); 1.0136 (5.4); 1.0080 (5.4); 0.9977 (2.3); 0.7884 (2.5); 0.7777 (6.0); 0.7727 (6.0); 0.7656 (5.6); 0.7604 (6.3); 0.7491 (1.8); 0.1459 (0.9); 0.0080 (6.9); -0.0002 (199.0); -0.0085 (7.6); -0.1496 (0.9)
II-24		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 8.9540 (6.1); 8.9419 (6.1); 8.8647 (1.4); 8.8462 (1.4); 8.0080 (3.3); 8.0040 (3.3); 7.5943 (1.7); 7.5822 (3.1); 7.5700 (1.6); 7.4564 (3.4); 7.4525 (3.4); 5.9496 (1.1); 5.9319 (1.7); 5.9141 (1.1); 5.7559 (6.8); 4.0395 (0.3); 4.0217 (0.3); 3.3259 (4.8); 2.6914 (1.7); 2.5087 (15.0); 2.5046 (19.2); 2.5004 (14.3); 2.3393 (0.5); 2.3258 (16.0); 1.9901 (1.4); 1.5847 (6.5); 1.5673 (6.4); 1.1941 (0.4); 1.1763 (0.7); 1.1585 (0.4); 0.0078 (0.6); -0.0002 (14.3)
II-25		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 8.9675 (6.5); 8.9554 (6.6); 8.7218 (1.2); 8.7024 (1.2); 7.6035 (1.8); 7.5914 (3.4); 7.5792 (1.7); 7.2269 (3.8); 6.0018 (1.0); 5.9836 (1.4); 5.9655 (1.0); 4.0174 (10.0); 3.3202 (57.9); 2.6901 (1.3); 2.6755 (0.6); 2.6711 (0.8); 2.6666 (0.6); 2.5243 (2.5); 2.5109 (47.5); 2.5066 (95.0); 2.5022 (124.0); 2.4977 (89.3); 2.4934 (43.5); 2.3295 (16.0); 1.9890 (0.5); 1.5689 (5.9); 1.5517 (5.8); 0.1462 (0.4); 0.0082 (3.1); 0.0001 (79.5); -0.0081 (2.9); -0.1492 (0.4)

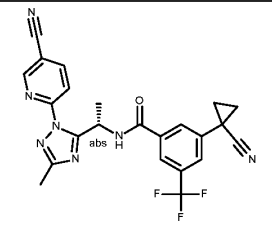
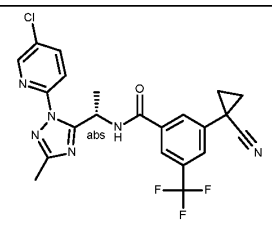
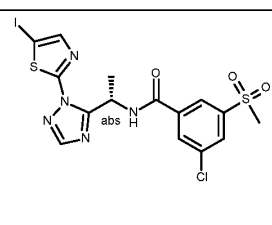
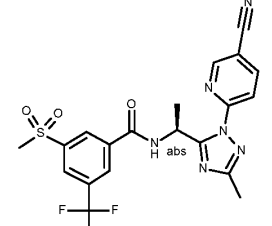
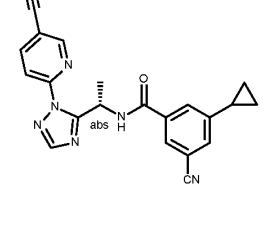
II-26		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2840 (3.7); 9.2666 (3.8); 9.0599 (6.6); 9.0558 (6.4); 9.0545 (6.4); 8.5814 (4.7); 8.5758 (4.5); 8.5599 (5.0); 8.5543 (5.0); 8.2389 (16.0); 8.0756 (6.7); 8.0541 (6.2); 7.8629 (6.4); 7.7024 (6.6); 7.6185 (6.4); 6.1071 (0.6); 6.0900 (2.5); 6.0726 (4.0); 6.0551 (2.6); 6.0379 (0.6); 5.7555 (2.6); 3.3211 (59.9); 2.6759 (0.7); 2.6715 (1.0); 2.6671 (0.8); 2.5246 (3.2); 2.5112 (63.2); 2.5071 (125.9); 2.5026 (165.3); 2.4981 (120.6); 2.4938 (60.8); 2.3339 (0.7); 2.3295 (1.0); 2.3249 (0.7); 2.1354 (0.7); 2.1228 (1.6); 2.1144 (1.7); 2.1019 (3.2); 2.0894 (1.8); 2.0810 (1.7); 2.0749 (1.0); 2.0684 (0.9); 1.6484 (15.1); 1.6310 (15.1); 1.2342 (0.6); 1.0618 (1.8); 1.0505 (5.3); 1.0450 (5.7); 1.0345 (2.9); 1.0296 (5.4); 1.0241 (5.4); 1.0138 (2.2); 0.8229 (2.3); 0.8120 (6.2); 0.8072 (6.1); 0.7999 (5.8); 0.7949 (6.6); 0.7833 (1.9); 0.1458 (0.6); 0.0077 (4.8); -0.0002 (123.2); -0.0085 (5.8); -0.1498 (0.6)
II-27		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2260 (3.8); 9.2084 (3.8); 8.6222 (7.2); 8.6158 (7.3); 8.2047 (4.3); 8.1982 (4.2); 8.1829 (5.0); 8.1764 (5.1); 8.1665 (15.8); 7.8778 (7.9); 7.8560 (7.0); 7.8312 (6.9); 7.6630 (7.1); 7.6121 (6.8); 5.9933 (0.6); 5.9761 (2.7); 5.9586 (4.2); 5.9411 (2.7); 5.9239 (0.6); 3.3236 (112.7); 2.6761 (0.8); 2.6717 (1.0); 2.6675 (0.8); 2.5248 (3.1); 2.5072 (132.8); 2.5028 (175.2); 2.4984 (129.8); 2.3340 (0.7); 2.3296 (1.0); 2.3251 (0.8); 2.1285 (0.7); 2.1160 (1.6); 2.1075 (1.8); 2.0951 (3.2); 2.0827 (1.9); 2.0742 (1.7); 2.0615 (0.8); 1.6370 (16.0); 1.6196 (15.9); 1.2337 (1.2); 1.0586 (1.9); 1.0473 (5.4); 1.0418 (5.8); 1.0314 (3.0); 1.0264 (5.5); 1.0210 (5.6); 1.0107 (2.2); 0.8542 (0.4); 0.8169 (2.3); 0.8056 (6.5); 0.8013 (6.5); 0.7938 (5.9); 0.7888 (6.9); 0.7773 (1.8); 0.1458 (0.4); 0.0078 (3.0); -0.0002 (79.8); -0.0083 (3.4); -0.1499 (0.4)
II-28		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5237 (2.8); 9.5058 (2.9); 9.0022 (15.7); 8.9900 (16.0); 8.5707 (3.5); 8.5669 (7.5); 8.5630 (5.0); 8.5318 (12.8); 8.5283 (10.2); 8.3154 (0.4); 8.1803 (13.2); 7.6464 (4.2); 7.6342 (8.0); 7.6221 (4.0); 6.0689 (0.5); 6.0519 (2.1); 6.0344 (3.3); 6.0169 (2.1); 5.9995 (0.4); 5.7558 (2.2); 3.3618 (1.0); 3.3558 (0.8); 3.3352 (35.2); 3.3231 (124.6); 2.9093 (0.3); 2.6757 (1.0); 2.6711 (1.4); 2.6666 (1.1); 2.5245 (4.4); 2.5197 (6.8); 2.5111 (88.0); 2.5067 (179.8); 2.5022 (236.9); 2.4976 (170.3); 2.4932 (82.2); 2.3334 (1.0); 2.3291 (1.4); 2.3244 (1.0); 1.6644 (12.7); 1.6470 (12.6); 1.2348 (1.0); 0.1460 (0.5); 0.0080 (4.5); -0.0001 (127.1); -0.0084 (4.6); -0.0190 (0.3); -0.1496 (0.6)
II-29		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5214 (3.6); 9.5038 (3.7); 8.6504 (7.1); 8.6441 (7.1); 8.5748 (8.3); 8.5713 (8.1); 8.5653 (4.6); 8.5613 (11.1); 8.5568 (13.1); 8.5533 (9.4); 8.3143 (0.4); 8.2221 (4.5); 8.2157 (4.4); 8.2003 (5.2); 8.1938 (5.5); 8.1867 (16.0); 7.9038 (7.6); 7.8819 (6.7); 6.0547 (0.6); 6.0378 (2.6); 6.0204 (4.1); 6.0029 (2.6); 5.9854 (0.6); 5.7545 (2.2); 3.3713 (0.9); 3.3402 (41.4); 3.3210 (90.1); 2.6760 (0.8); 2.6713 (1.0); 2.6669 (0.8); 2.5414 (0.6); 2.5247 (3.2); 2.5111 (68.4); 2.5069 (137.7); 2.5024 (181.6); 2.4980 (133.6); 2.4938 (67.3); 2.3338 (0.8); 2.3291 (1.1); 2.3248 (0.8); 1.6496 (15.5); 1.6323 (15.5); 1.2345 (0.9); 0.1458 (0.4); 0.0079 (3.1); -0.0001 (84.2); -0.0083 (3.7); -0.1496 (0.4)
II-30		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 8.9884 (15.7); 8.9762 (16.0); 8.8777 (2.5); 8.8596 (2.6); 8.3152 (1.9); 8.1429 (12.9); 7.9858 (8.0); 7.9815 (8.2); 7.6399 (4.4); 7.6277 (8.1); 7.6156 (4.1); 7.4345 (8.2); 7.4302 (8.2); 5.9575 (0.6); 5.9395 (2.2); 5.9219 (3.4); 5.9040 (2.2); 5.8871 (0.5); 4.0376 (1.0); 4.0198 (1.0); 3.3858 (0.3); 3.3627 (0.5); 3.3214 (451.2); 2.6893 (0.5); 2.6750 (4.5); 2.6704 (6.2); 2.6660 (4.5); 2.6166 (0.4); 2.5240 (17.0); 2.5192 (25.9); 2.5105 (379.2); 2.5061 (784.9); 2.5015 (1038.9); 2.4970 (745.6); 2.4925 (358.8); 2.3372 (2.0); 2.3329 (4.3); 2.3283 (6.1); 2.3238 (4.5); 1.9885 (4.4); 1.6047 (13.8); 1.5873 (13.9); 1.2376 (0.4); 1.1927 (1.3); 1.1749 (2.5); 1.1572 (1.3); 0.1459 (2.2); 0.0080 (16.9); -0.0002 (540.1); -0.0085 (20.5); -0.1497 (2.2)

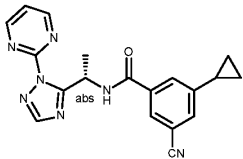
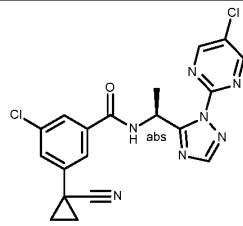
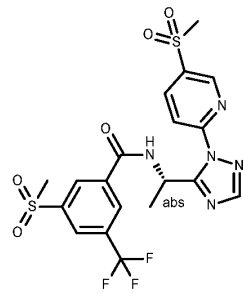
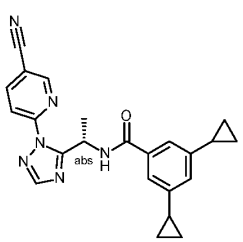
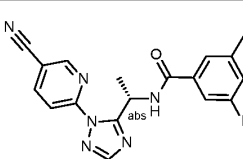
II-31		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.5151 (3.6); 9.4973 (3.7); 8.5805 (3.9); 8.5764 (4.4); 8.5687 (12.3); 8.5653 (11.6); 8.5600 (6.0); 8.5559 (10.1); 8.5518 (5.2); 8.5454 (7.4); 8.5420 (8.7); 8.3154 (0.3); 8.1586 (15.4); 8.1086 (2.0); 8.1041 (2.0); 8.0885 (3.8); 8.0843 (3.7); 8.0694 (2.6); 8.0648 (2.5); 7.8751 (6.2); 7.8546 (5.2); 7.5112 (3.1); 7.4989 (3.2); 7.4942 (3.0); 7.4818 (2.7); 7.4803 (2.8); 6.1091 (0.6); 6.0920 (2.7); 6.0745 (4.2); 6.0569 (2.7); 6.0393 (0.6); 3.3398 (41.2); 3.3232 (70.5); 2.6755 (1.0); 2.6712 (1.3); 2.6669 (1.0); 2.5067 (164.5); 2.5024 (211.5); 2.4980 (154.5); 2.3337 (0.9); 2.3293 (1.2); 2.3248 (0.9); 1.9890 (0.9); 1.6584 (16.0); 1.6411 (15.9); 1.2348 (0.7); 1.1754 (0.4); 0.1459 (0.4); 0.0075 (4.2); -0.0002 (95.8); -0.0082 (4.3); -0.1497 (0.5)
II-32		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.6737 (3.4); 9.6564 (3.2); 9.0778 (7.1); 9.0759 (7.3); 9.0723 (6.6); 9.0703 (6.0); 8.5902 (12.4); 8.5847 (8.0); 8.5687 (6.4); 8.5632 (5.9); 8.5003 (5.8); 8.3652 (5.2); 8.3632 (6.0); 8.3617 (5.6); 8.3152 (0.9); 8.2650 (16.0); 8.0965 (6.8); 8.0946 (6.4); 8.0751 (6.3); 8.0731 (6.0); 6.1764 (0.6); 6.1592 (2.4); 6.1419 (3.7); 6.1245 (2.3); 6.1069 (0.5); 5.7553 (1.8); 3.3218 (63.9); 3.1316 (0.9); 3.1197 (1.9); 3.1119 (2.0); 3.1075 (1.6); 3.1002 (3.5); 3.0922 (1.5); 3.0882 (2.0); 3.0804 (1.7); 3.0685 (0.8); 2.6808 (0.8); 2.6762 (1.3); 2.6716 (1.5); 2.6670 (1.0); 2.6623 (0.5); 2.5117 (135.6); 2.5072 (214.7); 2.5026 (246.7); 2.4980 (163.0); 2.4934 (70.7); 2.3387 (0.7); 2.3340 (1.2); 2.3295 (1.5); 2.3248 (1.0); 2.3204 (0.4); 2.0748 (2.2); 1.6776 (14.4); 1.6602 (13.8); 1.6328 (0.4); 1.2500 (0.6); 1.2303 (2.5); 1.2246 (3.2); 1.2169 (5.3); 1.2128 (5.3); 1.2053 (4.8); 1.2010 (2.8); 1.1972 (2.4); 1.1876 (0.8); 1.1752 (0.7); 1.1612 (0.5); 1.1504 (0.6); 1.1376 (0.8); 1.1289 (1.8); 1.1159 (4.8); 1.1091 (4.4); 1.0982 (4.3); 1.0961 (4.4); 1.0908 (3.2); 1.0748 (0.7); 0.1459 (0.8); 0.0079 (29.1); 0.0038 (27.3); -0.0002 (237.7); -0.0086 (7.3); -0.1496 (0.8)
II-33		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.6614 (2.4); 9.6440 (2.5); 9.0739 (4.2); 9.0724 (4.4); 9.0685 (4.6); 9.0669 (4.2); 8.5874 (3.7); 8.5818 (6.1); 8.5784 (5.2); 8.5660 (3.8); 8.5604 (3.7); 8.5071 (4.3); 8.3411 (4.4); 8.3143 (0.4); 8.2640 (11.6); 8.0929 (4.6); 8.0914 (4.5); 8.0715 (4.2); 8.0699 (4.2); 6.1735 (0.4); 6.1563 (1.7); 6.1390 (2.8); 6.1216 (1.8); 6.1041 (0.4); 5.7545 (1.2); 3.5097 (1.9); 3.4913 (6.4); 3.4730 (6.6); 3.4546 (2.0); 3.3194 (97.2); 2.6757 (1.0); 2.6712 (1.4); 2.6665 (1.0); 2.6623 (0.5); 2.5247 (4.1); 2.5199 (6.2); 2.5112 (84.6); 2.5068 (173.6); 2.5022 (228.1); 2.4977 (162.0); 2.4931 (77.6); 2.3382 (0.4); 2.3336 (1.0); 2.3291 (1.4); 2.3245 (1.0); 1.6762 (10.5); 1.6589 (10.4); 1.1459 (7.0); 1.1276 (16.0); 1.1092 (6.8); 0.1460 (0.8); 0.0080 (6.4); -0.0001 (190.9); -0.0085 (6.6); -0.1494 (0.8)
II-34		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.5855 (1.4); 9.5678 (1.4); 8.9967 (6.4); 8.9845 (6.5); 8.3764 (2.9); 8.3728 (2.0); 8.2869 (3.0); 8.1817 (6.1); 8.1698 (1.9); 8.1656 (3.1); 8.1616 (1.8); 7.6404 (1.7); 7.6282 (3.3); 7.6161 (1.7); 7.5374 (1.1); 7.4075 (2.5); 7.2776 (1.3); 6.0381 (1.0); 6.0206 (1.5); 6.0032 (1.0); 5.7576 (16.0); 4.0405 (0.9); 4.0227 (0.9); 3.3297 (6.1); 3.1801 (0.8); 3.1669 (0.8); 2.5108 (16.0); 2.5065 (21.3); 2.5021 (16.0); 1.9915 (4.0); 1.6632 (5.8); 1.6459 (5.8); 1.1950 (1.0); 1.1773 (2.1); 1.1594 (1.0); -0.0002 (3.0)

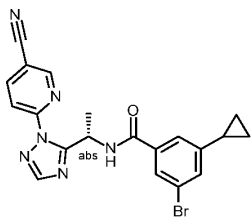
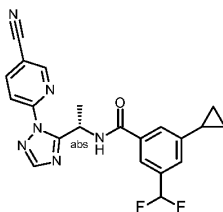
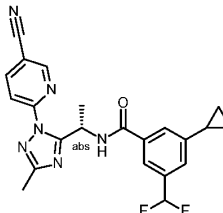
II-35		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6113 (1.7); 9.5941 (1.7); 9.0295 (3.1); 9.0256 (3.2); 8.5440 (1.8); 8.5387 (1.8); 8.5226 (1.9); 8.5172 (2.0); 8.4465 (3.6); 8.3502 (3.8); 8.1782 (3.5); 8.0292 (2.8); 8.0076 (2.6); 7.5416 (1.2); 7.4118 (2.5); 7.2818 (1.3); 6.1298 (1.2); 6.1124 (1.8); 6.0948 (1.2); 4.0385 (0.9); 4.0206 (0.9); 3.3219 (16.9); 2.6721 (0.6); 2.5068 (82.8); 2.5029 (101.4); 2.4989 (78.4); 2.3494 (16.0); 2.3302 (0.8); 1.9893 (3.8); 1.6346 (6.6); 1.6172 (6.5); 1.1934 (1.0); 1.1757 (2.0); 1.1579 (1.0); -0.0002 (5.5)
II-36		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5617 (1.7); 9.5441 (1.6); 8.5994 (2.8); 8.5940 (2.9); 8.4122 (3.4); 8.3309 (3.6); 8.1704 (4.9); 8.1548 (1.8); 8.1483 (1.8); 7.8483 (2.8); 7.8262 (2.4); 7.5377 (1.1); 7.4080 (2.4); 7.2781 (1.2); 6.0170 (1.1); 5.9996 (1.7); 5.9820 (1.1); 4.0381 (0.7); 4.0203 (0.7); 3.3227 (68.6); 2.6711 (1.2); 2.5023 (196.4); 2.3299 (16.0); 1.9889 (3.0); 1.6223 (6.1); 1.6050 (6.1); 1.3978 (1.2); 1.1931 (0.8); 1.1753 (1.6); 1.1575 (0.8); -0.0002 (11.6)
II-37		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.5749 (2.6); 9.5574 (2.7); 8.6308 (4.9); 8.6255 (5.0); 8.6245 (4.8); 8.3958 (3.6); 8.3918 (5.6); 8.3876 (4.0); 8.3119 (6.1); 8.2127 (3.2); 8.2062 (3.0); 8.1908 (4.4); 8.1850 (13.1); 8.1745 (3.8); 8.1703 (5.8); 7.8971 (5.0); 7.8960 (5.1); 7.8753 (4.4); 7.8742 (4.5); 7.5380 (2.0); 7.4080 (4.5); 7.2781 (2.3); 6.0340 (0.4); 6.0169 (1.8); 5.9996 (2.8); 5.9821 (1.8); 5.9648 (0.4); 3.3222 (33.2); 2.6726 (0.5); 2.5081 (69.4); 2.5038 (87.1); 2.4995 (64.0); 2.3306 (0.5); 1.9898 (0.7); 1.6465 (10.7); 1.6292 (10.6); 1.3974 (16.0); 1.1762 (0.4); -0.0002 (5.0)
II-38		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2525 (1.3); 9.2347 (1.3); 9.0293 (2.3); 9.0276 (2.4); 9.0238 (2.5); 9.0221 (2.3); 8.5410 (1.9); 8.5354 (1.8); 8.5194 (2.0); 8.5139 (2.0); 8.0215 (2.4); 8.0198 (2.5); 8.0000 (2.2); 7.9982 (2.3); 7.8278 (1.8); 7.8236 (2.8); 7.8194 (2.0); 7.7205 (1.8); 7.7165 (3.1); 7.7125 (1.9); 7.5286 (2.0); 7.5240 (3.5); 7.5194 (1.9); 6.0847 (1.0); 6.0672 (1.6); 6.0497 (1.0); 4.0385 (0.8); 4.0207 (0.8); 3.3165 (111.8); 2.6903 (12.6); 2.6797 (0.4); 2.6752 (0.7); 2.6707 (1.0); 2.6661 (0.7); 2.5242 (2.7); 2.5194 (4.1); 2.5108 (57.2); 2.5063 (119.6); 2.5017 (159.7); 2.4971 (113.2); 2.4926 (53.6); 2.3422 (16.0); 2.3333 (1.1); 2.3286 (1.1); 2.3240 (0.8); 1.9883 (3.8); 1.8155 (1.3); 1.8033 (3.1); 1.7957 (3.2); 1.7844 (1.4); 1.6456 (0.4); 1.6353 (1.6); 1.6226 (4.0); 1.6172 (7.8); 1.6003 (6.0); 1.3981 (3.5); 1.2351 (0.5); 1.1932 (1.0); 1.1754 (2.0); 1.1576 (1.0); 0.1460 (0.5); 0.0081 (4.0); -0.0001 (127.9); -0.0084 (4.5); -0.1495 (0.5)
II-39		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1619 (1.4); 9.1442 (1.4); 9.0161 (2.7); 9.0109 (2.6); 8.5355 (1.7); 8.5299 (1.6); 8.5140 (1.8); 8.5085 (1.8); 8.3117 (0.4); 8.0132 (2.7); 7.9916 (2.5); 7.5091 (3.2); 7.4914 (2.3); 7.2717 (2.3); 6.0705 (1.1); 6.0529 (1.7); 6.0352 (1.1); 3.3138 (158.0); 2.6897 (4.6); 2.6743 (1.4); 2.6699 (1.8); 2.6655 (1.4); 2.5054 (236.8); 2.5010 (306.0); 2.4966 (220.2); 2.3391 (16.0); 2.3280 (2.1); 2.3234 (1.4); 2.0691 (0.3); 2.0565 (0.6); 2.0480 (0.7); 2.0357 (1.3); 2.0229 (0.8); 2.0150 (0.7); 2.0020 (0.3); 1.9878 (0.3); 1.6171 (6.0); 1.5997 (6.0); 1.3978 (2.1); 1.2351 (0.8); 1.0485 (0.8); 1.0373 (2.3); 1.0319 (2.4); 1.0166 (2.3); 1.0111 (2.3); 1.0008 (0.8); 0.7955 (0.9); 0.7847 (2.5); 0.7800 (2.5); 0.7727 (2.4); 0.7676 (2.6); 0.7562 (0.8); 0.1454 (0.9); -0.0005 (185.7); -0.0084 (7.0); -0.1498 (0.9)

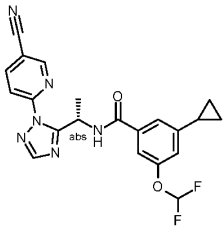
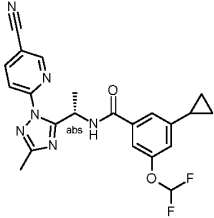
II-40		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4391 (3.2); 9.4218 (3.3); 8.6411 (6.7); 8.6348 (6.8); 8.3781 (0.4); 8.3734 (0.4); 8.3121 (0.7); 8.2951 (9.5); 8.2916 (13.2); 8.2875 (9.4); 8.2487 (5.7); 8.2445 (8.5); 8.2403 (4.0); 8.2179 (4.4); 8.2115 (4.2); 8.1961 (5.1); 8.1896 (5.0); 8.1770 (16.0); 7.8952 (7.4); 7.8734 (6.5); 6.0252 (0.5); 6.0081 (2.5); 5.9906 (3.9); 5.9731 (2.5); 5.9557 (0.5); 4.0382 (0.3); 4.0204 (0.4); 3.3259 (2.0); 3.3120 (88.2); 2.6789 (0.7); 2.6747 (1.5); 2.6702 (2.1); 2.6657 (1.5); 2.5237 (6.1); 2.5189 (9.0); 2.5102 (128.5); 2.5058 (266.0); 2.5013 (352.8); 2.4967 (248.7); 2.4922 (117.4); 2.3372 (0.7); 2.3326 (1.5); 2.3281 (2.0); 2.3235 (1.5); 2.3191 (0.7); 1.9880 (1.5); 1.6377 (15.2); 1.6203 (15.1); 1.2980 (0.4); 1.2592 (0.5); 1.2352 (0.9); 1.1930 (0.4); 1.1753 (0.9); 1.1575 (0.4); 0.1458 (1.1); 0.0079 (8.2); -0.0002 (257.5); -0.0085 (8.7); -0.1498 (1.1)
II-41		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4914 (2.6); 9.4739 (2.6); 9.0735 (4.6); 9.0718 (5.0); 9.0681 (5.0); 9.0663 (4.7); 8.5873 (3.9); 8.5817 (3.7); 8.5658 (4.2); 8.5602 (4.2); 8.3309 (2.6); 8.3270 (6.6); 8.3222 (8.0); 8.3174 (7.7); 8.3137 (2.9); 8.2587 (4.6); 8.2543 (8.0); 8.2504 (16.0); 8.0907 (5.0); 8.0889 (5.0); 8.0692 (4.6); 8.0674 (4.7); 6.1391 (0.4); 6.1220 (1.9); 6.1046 (3.0); 6.0873 (1.9); 6.0696 (0.4); 4.0568 (0.5); 4.0390 (1.5); 4.0212 (1.5); 4.0034 (0.5); 3.3207 (32.5); 3.3134 (27.8); 2.6761 (0.4); 2.6715 (0.6); 2.6669 (0.4); 2.5250 (1.8); 2.5202 (2.7); 2.5116 (36.1); 2.5071 (74.6); 2.5025 (98.7); 2.4979 (69.4); 2.4934 (32.4); 2.3339 (0.4); 2.3292 (0.6); 2.3248 (0.4); 2.0113 (1.0); 1.9997 (0.3); 1.9888 (6.6); 1.6510 (11.3); 1.6336 (11.2); 1.1936 (1.8); 1.1759 (3.6); 1.1581 (1.7); 0.8890 (1.1); 0.8722 (1.0); 0.0080 (2.7); -0.0002 (80.2); -0.0085 (2.5)
II-42		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.4692 (2.1); 9.4514 (2.1); 8.9731 (16.0); 8.3157 (1.2); 8.2525 (2.8); 8.2488 (5.5); 8.2450 (3.3); 8.1859 (9.2); 8.1585 (2.3); 8.1538 (4.8); 8.1499 (3.6); 8.1400 (3.8); 8.1358 (4.8); 8.1312 (2.2); 7.6275 (2.4); 7.4464 (5.2); 7.2654 (2.6); 5.9642 (1.4); 5.9467 (2.3); 5.9292 (1.4); 3.3271 (274.2); 3.3104 (25.5); 2.6801 (0.6); 2.6756 (1.2); 2.6711 (1.7); 2.6665 (1.2); 2.5245 (5.7); 2.5198 (8.5); 2.5111 (91.5); 2.5066 (184.5); 2.5020 (249.0); 2.4974 (189.3); 2.4930 (96.0); 2.3378 (0.5); 2.3334 (1.1); 2.3289 (1.6); 2.3244 (1.1); 2.3199 (0.5); 2.0861 (0.8); 1.6521 (8.4); 1.6347 (8.4); 0.1460 (0.4); 0.0080 (3.3); -0.0002 (100.1); -0.0085 (3.7); -0.1496 (0.4)
II-43		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6317 (2.0); 9.6140 (2.1); 8.9555 (16.0); 8.4677 (2.6); 8.4634 (4.7); 8.4592 (3.5); 8.4118 (4.3); 8.4022 (3.0); 8.3979 (3.7); 8.1992 (9.6); 7.6158 (2.4); 7.4348 (5.3); 7.2539 (2.6); 5.9726 (1.4); 5.9552 (2.3); 5.9377 (1.5); 5.7567 (11.5); 3.3264 (56.3); 2.6767 (0.4); 2.6723 (0.6); 2.6677 (0.4); 2.5258 (2.3); 2.5210 (3.5); 2.5124 (34.5); 2.5079 (69.2); 2.5033 (93.4); 2.4987 (71.2); 2.4942 (36.1); 2.3348 (0.4); 2.3301 (0.6); 2.3254 (0.4); 2.0121 (0.5); 1.9896 (1.1); 1.6632 (8.5); 1.6458 (8.4); 1.1757 (0.6); 0.8888 (0.6); 0.8720 (0.6); 0.0080 (1.5); -0.0002 (42.3); -0.0085 (1.6)
II-44		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.7611 (0.6); 9.7578 (0.7); 9.4550 (1.4); 9.4371 (1.4); 9.2209 (16.0); 9.0607 (0.7); 8.5905 (0.4); 8.5292 (1.4); 8.5242 (4.3); 8.2498 (1.9); 8.2460 (3.7); 8.2423 (2.3); 8.1876 (6.4); 8.1497 (1.0); 8.1445 (4.2); 8.1422 (5.2); 8.1387 (4.3); 8.1338 (0.9); 5.9817 (1.0); 5.9641 (1.5); 5.9466 (1.0); 3.3357 (2.2); 3.3259 (47.3); 3.3179 (16.8); 2.6716 (0.4); 2.5252 (1.3); 2.5205 (1.9); 2.5117 (25.0); 2.5073 (51.6); 2.5027 (69.1); 2.4981 (51.2); 2.4936 (25.3); 2.3295 (0.4); 2.0866 (1.1); 1.8616 (4.8); 1.7798 (0.6); 1.6365 (5.6); 1.6191 (5.6); 1.4380 (0.5); 1.4200 (0.6); 1.0869 (0.7); 1.0373 (0.4); 0.0080 (1.6); -0.0002 (47.9); -0.0084 (1.6)

II-45		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.6149 (2.5); 9.5973 (2.5); 8.6375 (4.5); 8.6364 (4.7); 8.6311 (4.8); 8.6300 (4.6); 8.5584 (5.0); 8.4753 (4.4); 8.3352 (4.4); 8.3161 (0.4); 8.2130 (3.5); 8.2065 (3.4); 8.1936 (11.8); 8.1847 (4.0); 7.8980 (5.1); 7.8969 (5.2); 7.8763 (4.5); 7.8750 (4.6); 6.0603 (0.4); 6.0432 (1.8); 6.0258 (2.8); 6.0083 (1.8); 5.9908 (0.4); 4.0558 (0.5); 4.0381 (1.6); 4.0203 (1.6); 4.0026 (0.6); 3.5066 (1.9); 3.4882 (6.5); 3.4698 (6.6); 3.4515 (2.0); 3.3281 (212.5); 2.6763 (0.8); 2.6717 (1.2); 2.6672 (0.9); 2.5252 (3.4); 2.5205 (5.2); 2.5117 (67.2); 2.5073 (136.6); 2.5027 (181.4); 2.4982 (133.5); 2.4937 (65.4); 2.3388 (0.4); 2.3342 (0.8); 2.3296 (1.1); 2.3251 (0.8); 1.9893 (7.0); 1.6645 (10.6); 1.6471 (10.5); 1.1931 (1.9); 1.1754 (3.9); 1.1575 (2.0); 1.1420 (7.0); 1.1237 (16.0); 1.1052 (6.8); 0.1460 (0.4); 0.0080 (3.0); -0.0001 (94.9); -0.0085 (3.3); -0.1496 (0.4)
II-46		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.6242 (3.0); 9.6065 (3.1); 8.6387 (5.9); 8.6324 (6.0); 8.5693 (6.1); 8.4673 (5.5); 8.3569 (5.5); 8.2147 (4.1); 8.2082 (3.9); 8.1941 (16.0); 8.1864 (4.8); 7.9007 (6.5); 7.8788 (5.7); 6.0640 (0.4); 6.0467 (2.2); 6.0293 (3.4); 6.0118 (2.2); 5.9941 (0.4); 4.0388 (0.7); 4.0210 (0.7); 3.6631 (0.5); 3.3299 (106.2); 3.1283 (0.6); 3.1164 (1.4); 3.1087 (1.6); 3.1045 (1.0); 3.0970 (2.9); 3.0887 (1.1); 3.0850 (1.6); 3.0772 (1.5); 3.0652 (0.7); 2.6774 (0.4); 2.6729 (0.6); 2.6684 (0.4); 2.5263 (1.9); 2.5129 (35.2); 2.5085 (71.3); 2.5039 (94.7); 2.4994 (70.0); 2.4949 (34.7); 2.3353 (0.4); 2.3307 (0.6); 2.3263 (0.4); 1.9900 (3.0); 1.6672 (12.9); 1.6498 (12.8); 1.2476 (0.3); 1.2290 (1.3); 1.2225 (2.2); 1.2115 (4.1); 1.2038 (4.0); 1.1938 (2.7); 1.1759 (2.0); 1.1693 (0.4); 1.1617 (0.4); 1.1582 (0.9); 1.1499 (0.6); 1.1364 (0.4); 1.1282 (1.1); 1.1156 (3.7); 1.1086 (3.7); 1.0957 (3.6); 1.0899 (2.9); 1.0750 (0.6); 0.0078 (1.5); -0.0002 (44.2); -0.0085 (1.7)
II-47		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.4409 (3.4); 9.4235 (3.5); 9.0690 (5.7); 9.0672 (6.2); 9.0635 (6.3); 9.0617 (5.9); 8.5859 (5.1); 8.5804 (4.9); 8.5645 (5.4); 8.5589 (5.5); 8.3161 (0.5); 8.2504 (16.0); 8.0864 (12.2); 8.0847 (11.0); 8.0649 (6.0); 8.0631 (6.2); 8.0131 (6.2); 7.7837 (5.7); 6.1343 (0.5); 6.1171 (2.4); 6.0997 (3.8); 6.0823 (2.4); 6.0651 (0.5); 5.7563 (10.3); 4.0561 (0.6); 4.0382 (1.9); 4.0204 (1.9); 4.0026 (0.6); 3.3254 (108.5); 2.6809 (0.5); 2.6764 (1.0); 2.6718 (1.4); 2.6673 (1.0); 2.6628 (0.5); 2.5254 (4.1); 2.5207 (6.0); 2.5119 (77.5); 2.5074 (159.3); 2.5029 (211.4); 2.4983 (154.8); 2.4937 (75.1); 2.3387 (0.4); 2.3343 (0.9); 2.3297 (1.3); 2.3251 (1.0); 2.3206 (0.4); 1.9894 (8.6); 1.8615 (2.6); 1.8493 (7.2); 1.8415 (8.1); 1.8302 (3.7); 1.7906 (0.5); 1.7378 (0.5); 1.6991 (4.2); 1.6868 (7.9); 1.6792 (7.3); 1.6624 (14.8); 1.6450 (14.4); 1.1934 (2.4); 1.1756 (4.8); 1.1578 (2.4); 0.8719 (0.3); 0.0080 (2.5); -0.0001 (85.2); -0.0085 (2.8)
II-48		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.3826 (3.2); 9.3650 (3.3); 8.6301 (6.2); 8.6239 (6.4); 8.2100 (4.4); 8.2036 (4.2); 8.1882 (5.2); 8.1795 (16.0); 8.0524 (5.9); 7.9792 (6.2); 7.8881 (7.0); 7.8774 (0.6); 7.8663 (6.2); 7.7780 (5.7); 7.4167 (0.3); 7.3945 (0.4); 7.3879 (0.3); 6.4579 (0.6); 6.4360 (0.5); 6.1222 (0.4); 6.0205 (0.5); 6.0033 (2.4); 5.9858 (3.7); 5.9683 (2.4); 5.9507 (0.5); 5.7574 (11.8); 4.0566 (0.4); 4.0388 (1.3); 4.0210 (1.3); 4.0033 (0.4); 3.3269 (36.9); 2.6773 (0.5); 2.6727 (0.7); 2.6682 (0.5); 2.5262 (2.1); 2.5213 (3.3); 2.5127 (42.7); 2.5083 (86.1); 2.5038 (113.3); 2.4992 (82.9); 2.4948 (40.4); 2.3351 (0.5); 2.3306 (0.7); 2.3262 (0.5); 1.9900 (5.7); 1.9102 (0.5); 1.8611 (2.6); 1.8484 (7.2); 1.8409 (7.9); 1.8293 (3.5); 1.7900 (0.5); 1.7310 (0.5); 1.6924 (3.9); 1.6801 (7.6); 1.6728 (7.3); 1.6592 (4.0); 1.6524 (14.3); 1.6350 (14.1); 1.1937 (1.5); 1.1759 (3.0); 1.1581 (1.5); 0.0080 (1.4); -0.0002 (45.6); -0.0085 (1.5)

II-49		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.4228 (1.6); 9.4050 (1.6); 9.0331 (2.6); 9.0278 (2.7); 8.5437 (1.9); 8.5381 (1.9); 8.5221 (2.0); 8.5166 (2.1); 8.1143 (2.8); 8.0338 (2.9); 8.0252 (3.1); 8.0035 (2.6); 7.7865 (2.7); 6.1211 (1.1); 6.1035 (1.7); 6.0859 (1.1); 5.7561 (3.0); 4.0383 (0.6); 4.0205 (0.6); 3.3272 (54.8); 2.6765 (0.3); 2.6720 (0.4); 2.6680 (0.3); 2.5256 (1.4); 2.5119 (26.6); 2.5077 (53.8); 2.5032 (71.6); 2.4987 (53.9); 2.4945 (27.7); 2.3448 (16.0); 2.3304 (0.6); 2.3256 (0.4); 1.9895 (2.8); 1.8630 (1.1); 1.8501 (3.1); 1.8426 (3.6); 1.8312 (1.6); 1.7017 (1.7); 1.6897 (3.4); 1.6823 (3.2); 1.6691 (1.2); 1.6403 (6.0); 1.6229 (6.0); 1.1935 (0.7); 1.1758 (1.4); 1.1580 (0.7); 0.0078 (0.7); -0.0002 (22.5); -0.0083 (0.9)
II-50		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.3646 (1.3); 9.3466 (1.4); 8.5969 (2.6); 8.5905 (2.6); 8.1731 (1.7); 8.1666 (1.6); 8.1512 (1.9); 8.1447 (1.9); 8.0777 (2.5); 7.9980 (2.5); 7.8411 (2.9); 7.8192 (2.6); 7.7793 (2.4); 6.0060 (1.0); 5.9884 (1.6); 5.9708 (1.0); 5.7565 (2.3); 4.0385 (0.8); 4.0207 (0.8); 3.3264 (27.0); 2.6723 (0.4); 2.5257 (1.1); 2.5208 (1.7); 2.5122 (21.8); 2.5078 (43.6); 2.5033 (57.3); 2.4988 (42.0); 2.4943 (20.6); 2.3271 (16.0); 1.9896 (3.6); 1.8608 (1.0); 1.8481 (2.9); 1.8406 (3.2); 1.8291 (1.4); 1.6941 (1.6); 1.6820 (3.1); 1.6747 (3.0); 1.6613 (1.1); 1.6292 (5.6); 1.6118 (5.6); 1.1936 (1.0); 1.1758 (1.9); 1.1580 (0.9); 0.0078 (0.7); -0.0002 (20.5); -0.0085 (0.7)
II-51		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.5415 (1.3); 9.5249 (1.3); 8.3344 (2.0); 8.3308 (3.3); 8.3271 (2.0); 8.2308 (8.0); 8.1645 (2.1); 8.1602 (3.0); 8.1559 (1.7); 7.9519 (6.4); 6.0004 (0.9); 5.9834 (1.3); 5.9662 (0.9); 4.3785 (2.4); 3.3302 (15.2); 3.3204 (35.9); 2.6753 (0.6); 2.6710 (0.7); 2.6665 (0.5); 2.5854 (5.7); 2.5651 (16.0); 2.5240 (2.9); 2.5064 (88.4); 2.5020 (110.0); 2.4975 (79.4); 2.3332 (0.5); 2.3288 (0.7); 2.3244 (0.5); 1.9886 (0.7); 1.6223 (5.1); 1.6048 (5.0); 1.1751 (0.4); 0.0076 (1.8); -0.0003 (37.9); -0.0085 (1.8)
II-52		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.6427 (1.3); 9.6250 (1.4); 9.0467 (2.2); 9.0449 (2.4); 9.0412 (2.4); 9.0393 (2.2); 8.6483 (2.6); 8.5487 (2.0); 8.5431 (1.9); 8.5272 (2.1); 8.5216 (2.2); 8.5082 (2.3); 8.4002 (2.3); 8.3157 (0.3); 8.0348 (2.5); 8.0331 (2.5); 8.0133 (2.3); 8.0115 (2.4); 6.1625 (1.0); 6.1449 (1.6); 6.1274 (1.0); 3.9616 (0.3); 3.3802 (15.4); 3.3262 (111.4); 2.6804 (0.3); 2.6760 (0.7); 2.6713 (1.0); 2.6667 (0.7); 2.6622 (0.3); 2.5417 (1.4); 2.5248 (2.7); 2.5201 (4.1); 2.5114 (54.5); 2.5069 (111.8); 2.5023 (148.4); 2.4977 (108.1); 2.4932 (52.4); 2.3496 (16.0); 2.3389 (0.6); 2.3338 (0.8); 2.3292 (1.0); 2.3246 (0.7); 2.3202 (0.4); 1.6518 (5.4); 1.6344 (5.4); 0.0080 (1.6); -0.0002 (53.7); -0.0085 (1.7)
II-53		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.2370 (3.5); 9.2196 (3.6); 9.0673 (5.9); 9.0656 (6.3); 9.0618 (6.4); 9.0601 (6.0); 8.5862 (5.0); 8.5807 (4.8); 8.5647 (5.4); 8.5592 (5.4); 8.3174 (0.4); 8.2406 (16.0); 8.0827 (6.4); 8.0810 (6.6); 8.0612 (6.0); 8.0594 (6.2); 7.9817 (4.9); 7.9780 (8.9); 7.9742 (5.2); 7.7594 (3.9); 7.7554 (7.8); 7.7514 (5.0); 7.7308 (5.3); 7.7270 (8.0); 7.7232 (4.0); 6.1031 (0.5); 6.0862 (2.5); 6.0688 (3.9); 6.0514 (2.5); 6.0342 (0.5); 3.3325 (189.5); 2.6767 (1.0); 2.6722 (1.4); 2.6676 (1.0); 2.6633 (0.5); 2.5256 (4.6); 2.5209 (7.3); 2.5121 (82.4); 2.5077 (165.1); 2.5032 (215.8); 2.4986 (157.9); 2.4941 (77.2); 2.3345 (1.0); 2.3300 (1.3); 2.3255 (1.0); 2.3211 (0.5); 2.0758 (9.7); 2.0610 (1.6); 2.0526 (1.7); 2.0488 (1.2); 2.0402 (3.1); 2.0277 (1.8); 2.0193 (1.6); 2.0068 (0.8); 1.6362 (14.8); 1.6189 (14.8); 1.0536 (1.8); 1.0424 (5.2); 1.0368 (5.6); 1.0332 (3.0); 1.0263 (2.9); 1.0214 (5.4); 1.0158 (5.3); 1.0055 (2.2); 0.8246 (2.3); 0.8139 (6.0); 0.8087 (5.9); 0.8016 (5.6); 0.7964 (6.4); 0.7848 (1.8); 0.1459 (1.0); 0.0080 (9.4); -0.0001 (254.6); -0.0085 (9.3); -0.1496 (1.0)

II-54		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1903 (2.7); 9.1724 (2.8); 8.9927 (15.7); 8.9805 (16.0); 8.1615 (12.8); 7.9213 (3.9); 7.9176 (7.2); 7.9138 (4.2); 7.7175 (2.9); 7.7137 (6.6); 7.7100 (5.0); 7.7043 (4.9); 7.7002 (6.3); 7.6962 (2.8); 7.6392 (4.3); 7.6271 (8.1); 7.6149 (4.1); 6.0106 (0.4); 5.9935 (2.0); 5.9759 (3.2); 5.9583 (2.1); 5.9409 (0.4); 3.3375 (32.8); 2.6775 (0.5); 2.6729 (0.6); 2.6683 (0.5); 2.5264 (2.2); 2.5216 (3.5); 2.5129 (39.1); 2.5085 (78.2); 2.5039 (102.2); 2.4994 (75.2); 2.4949 (37.1); 2.3354 (0.5); 2.3308 (0.6); 2.3263 (0.5); 2.0764 (1.1); 2.0607 (0.6); 2.0481 (1.2); 2.0397 (1.3); 2.0273 (2.5); 2.0147 (1.4); 2.0064 (1.3); 1.9938 (0.7); 1.6397 (12.4); 1.6222 (12.3); 1.0464 (1.5); 1.0355 (4.0); 1.0298 (4.3); 1.0259 (2.4); 1.0195 (2.4); 1.0145 (4.2); 1.0088 (4.1); 0.9986 (1.8); 0.8126 (1.9); 0.8020 (4.6); 0.7968 (4.5); 0.7897 (4.3); 0.7847 (4.7); 0.7733 (1.4); 0.1459 (0.5); 0.0079 (4.4); -0.0002 (117.6); -0.0085 (4.6); -0.1496 (0.5)
II-55		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.2501 (1.4); 9.2323 (1.5); 9.1197 (16.0); 8.1904 (6.7); 7.7653 (2.0); 7.7611 (3.2); 7.7570 (2.2); 7.6667 (2.1); 7.6628 (3.5); 7.6589 (2.1); 7.5225 (2.2); 7.5179 (3.7); 7.5133 (1.9); 5.9580 (1.0); 5.9405 (1.6); 5.9229 (1.0); 5.7562 (5.8); 3.6535 (0.5); 3.3300 (108.0); 2.6764 (0.4); 2.6717 (0.6); 2.6673 (0.4); 2.5422 (1.0); 2.5252 (1.8); 2.5203 (3.0); 2.5118 (36.4); 2.5074 (73.3); 2.5029 (95.8); 2.4983 (68.9); 2.4938 (33.0); 2.3342 (0.4); 2.3296 (0.6); 2.3252 (0.4); 1.8135 (1.2); 1.8021 (3.0); 1.7943 (3.4); 1.7833 (1.6); 1.6360 (6.4); 1.6275 (2.7); 1.6186 (8.1); 1.6103 (2.7); 1.6076 (2.9); 1.5963 (1.4); 0.0079 (2.6); -0.0002 (67.8); -0.0085 (2.4)
II-56		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.6734 (3.4); 9.6559 (3.5); 9.0628 (5.9); 9.0614 (6.3); 9.0568 (6.3); 9.0553 (6.2); 8.6328 (6.6); 8.5885 (4.8); 8.5824 (4.6); 8.5669 (5.2); 8.5608 (5.2); 8.5016 (5.9); 8.4009 (5.8); 8.2703 (16.0); 8.1645 (6.4); 8.1631 (6.4); 8.1430 (5.8); 8.1415 (6.0); 6.2014 (0.5); 6.1843 (2.3); 6.1669 (3.7); 6.1495 (2.4); 6.1319 (0.5); 3.4175 (0.4); 3.3894 (40.8); 3.3771 (38.1); 3.3326 (69.4); 2.6772 (0.5); 2.6728 (0.7); 2.6682 (0.5); 2.5262 (2.1); 2.5214 (3.5); 2.5128 (42.3); 2.5083 (85.5); 2.5038 (112.6); 2.4992 (81.6); 2.4947 (39.4); 2.3351 (0.5); 2.3306 (0.7); 2.3260 (0.5); 2.0766 (0.4); 1.6942 (13.6); 1.6768 (13.5); 0.0080 (0.5); -0.0002 (15.8); -0.0085 (0.5)
II-57		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.0589 (5.8); 9.0572 (6.3); 9.0535 (6.4); 9.0517 (5.9); 8.9326 (3.4); 8.9150 (3.6); 8.5810 (5.1); 8.5755 (5.0); 8.5596 (5.5); 8.5540 (5.5); 8.3162 (0.4); 8.2209 (16.0); 8.0656 (6.3); 8.0639 (6.4); 8.0442 (5.9); 8.0424 (6.1); 7.3810 (0.5); 7.3766 (0.6); 7.1731 (15.0); 7.1690 (16.0); 6.9735 (4.2); 6.9696 (7.2); 6.9659 (4.1); 6.0573 (0.5); 6.0401 (2.4); 6.0227 (3.8); 6.0052 (2.4); 5.9877 (0.5); 3.3259 (137.7); 2.6804 (0.4); 2.6759 (0.9); 2.6713 (1.2); 2.6668 (0.9); 2.6621 (0.4); 2.5248 (3.4); 2.5201 (5.2); 2.5114 (69.2); 2.5069 (142.5); 2.5023 (189.5); 2.4978 (138.1); 2.4933 (66.8); 2.3382 (0.4); 2.3337 (0.8); 2.3292 (1.2); 2.3246 (0.8); 2.3203 (0.4); 2.0748 (3.2); 1.9227 (1.4); 1.9100 (3.0); 1.9017 (3.2); 1.8977 (2.3); 1.8891 (6.1); 1.8806 (2.2); 1.8765 (3.5); 1.8682 (3.3); 1.8555 (1.6); 1.6255 (14.5); 1.6081 (14.5); 0.9653 (0.5); 0.9598 (0.6); 0.9528 (3.8); 0.9420 (10.8); 0.9366 (11.6); 0.9320 (5.3); 0.9265 (5.4); 0.9210 (11.0); 0.9156 (11.0); 0.9056 (4.3); 0.8885 (0.4); 0.8676 (0.3); 0.7332 (0.4); 0.7198 (0.4); 0.6948 (4.6); 0.6846 (11.6); 0.6821 (9.6); 0.6794 (12.2); 0.6721 (11.5); 0.6669 (12.0); 0.6560 (3.8); 0.1459 (0.8); 0.0080 (6.3); -0.0001 (187.8); -0.0085 (6.7); -0.0165 (0.4); -0.1496 (0.8)
II-58		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1145 (3.6); 9.0973 (3.7); 9.0608 (6.4); 9.0591 (6.8); 9.0554 (7.0); 9.0536 (6.3); 8.5847 (5.2); 8.5792 (5.0); 8.5633 (5.6); 8.5577 (5.6); 8.3171 (2.1); 8.2462 (0.8); 8.2422 (0.6); 8.2351 (1.7); 8.2288 (16.0); 8.1245 (1.0); 8.1205 (1.0); 8.0731 (6.5); 8.0715 (6.7); 8.0516 (6.0); 8.0499 (6.2); 7.8789 (5.0); 7.8753 (8.9); 7.8715 (5.2); 7.8206 (0.4); 7.6227 (4.8); 7.6191 (8.2); 7.6155 (4.8); 7.4100 (8.0); 6.0630 (0.6);

		6.0454 (2.6); 6.0279 (4.0); 6.0105 (2.5); 5.9924 (0.6); 3.3994 (0.3); 3.3282 (488.3); 2.6756 (4.8); 2.6711 (6.6); 2.6666 (4.9); 2.6349 (0.4); 2.6181 (0.5); 2.5245 (22.1); 2.5110 (394.8); 2.5067 (786.9); 2.5021 (1028.3); 2.4976 (752.1); 2.4931 (370.2); 2.4329 (0.5); 2.3335 (4.6); 2.3289 (6.3); 2.3245 (4.7); 2.0750 (2.7); 1.9658 (0.8); 1.9536 (1.5); 1.9451 (1.7); 1.9323 (3.1); 1.9201 (1.8); 1.9116 (1.7); 1.8991 (0.8); 1.6535 (0.5); 1.6181 (15.2); 1.6007 (15.2); 0.9920 (1.9); 0.9808 (5.4); 0.9754 (5.9); 0.9714 (3.1); 0.9651 (3.0); 0.9599 (5.6); 0.9545 (5.6); 0.9443 (2.2); 0.7331 (2.2); 0.7225 (6.0); 0.7174 (6.1); 0.7101 (5.8); 0.7050 (6.4); 0.6938 (1.9); 0.1458 (3.7); 0.0079 (33.6); -0.0002 (895.2); -0.0085 (35.4); -0.0667 (0.3); -0.1497 (3.9)
II-59		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1401 (3.9); 9.1228 (4.0); 9.0921 (0.5); 9.0617 (7.2); 9.0567 (6.6); 8.5845 (4.5); 8.5790 (4.2); 8.5629 (5.1); 8.5575 (4.6); 8.5454 (0.7); 8.5404 (0.6); 8.3165 (1.6); 8.2312 (14.4); 8.1341 (0.4); 8.1249 (0.4); 8.1050 (0.4); 8.0745 (6.7); 8.0530 (6.1); 7.7703 (0.8); 7.7108 (8.2); 7.5397 (0.8); 7.4576 (8.0); 7.4251 (8.2); 7.4061 (0.9); 6.0913 (0.4); 6.0725 (0.9); 6.0548 (2.9); 6.0373 (4.1); 6.0200 (2.6); 6.0024 (0.6); 3.5682 (0.9); 3.3874 (0.4); 3.3265 (491.9); 2.6752 (5.0); 2.6710 (6.4); 2.6668 (4.7); 2.6509 (0.4); 2.6316 (0.4); 2.5064 (840.1); 2.5021 (1044.8); 2.4977 (756.2); 2.3332 (4.9); 2.3289 (6.3); 2.3245 (4.5); 2.0743 (0.5); 2.0481 (0.4); 2.0059 (0.8); 1.9931 (1.6); 1.9846 (1.8); 1.9721 (3.3); 1.9600 (1.9); 1.9513 (1.9); 1.9436 (0.5); 1.9392 (0.9); 1.6561 (1.9); 1.6386 (2.0); 1.6229 (16.0); 1.6055 (15.9); 1.5694 (0.7); 1.5511 (0.6); 1.1045 (0.5); 1.0115 (2.2); 1.0003 (6.2); 0.9950 (6.6); 0.9844 (3.4); 0.9794 (5.9); 0.9742 (5.5); 0.9638 (2.2); 0.9470 (0.4); 0.7998 (0.8); 0.7865 (0.7); 0.7609 (2.3); 0.7499 (6.7); 0.7453 (6.4); 0.7380 (6.2); 0.7331 (6.6); 0.7214 (1.8); 0.1459 (3.4); 0.0077 (34.1); -0.0002 (744.7); -0.0084 (31.8); -0.0681 (0.4); -0.1496 (3.5)
II-60		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1970 (3.6); 9.1795 (3.6); 9.0636 (6.2); 9.0622 (6.4); 9.0583 (6.7); 8.5831 (4.7); 8.5776 (4.6); 8.5616 (5.0); 8.5561 (5.0); 8.3165 (0.4); 8.2330 (16.0); 8.0778 (6.6); 8.0765 (6.4); 8.0564 (6.1); 8.0549 (5.9); 7.7424 (6.3); 7.5957 (6.1); 7.4415 (6.2); 7.3347 (0.3); 7.1458 (2.8); 7.0064 (6.4); 6.8670 (3.1); 6.0984 (0.6); 6.0810 (2.6); 6.0636 (4.1); 6.0461 (2.6); 6.0294 (0.6); 4.0379 (0.6); 4.0202 (0.6); 3.3271 (80.0); 2.9806 (0.5); 2.8774 (0.5); 2.6761 (0.9); 2.6717 (1.3); 2.6672 (1.0); 2.5251 (3.8); 2.5201 (6.0); 2.5115 (80.3); 2.5072 (161.4); 2.5027 (209.2); 2.4982 (151.9); 2.4939 (74.7); 2.3339 (0.9); 2.3295 (1.2); 2.3251 (0.9); 2.0732 (0.8); 2.0605 (1.7); 2.0520 (1.8); 2.0482 (1.4); 2.0396 (3.2); 2.0271 (2.0); 2.0187 (1.7); 2.0118 (0.5); 2.0061 (0.9); 1.9893 (2.6); 1.6399 (15.4); 1.6225 (15.4); 1.2264 (0.5); 1.2095 (0.5); 1.1930 (0.7); 1.1752 (1.4); 1.1574 (0.7); 1.0404 (1.8); 1.0295 (5.6); 1.0241 (6.2); 1.0198 (3.2); 1.0137 (2.9); 1.0085 (5.9); 1.0032 (5.9); 0.9929 (2.2); 0.7745 (2.3); 0.7639 (6.2); 0.7590 (6.4); 0.7516 (6.1); 0.7468 (6.5); 0.7354 (2.0); 0.0079 (1.4); -0.0002 (40.5); -0.0085 (1.5)
II-61		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ= 9.1771 (1.4); 9.1593 (1.4); 9.0281 (2.6); 9.0242 (2.6); 8.5413 (1.7); 8.5357 (1.6); 8.5197 (1.9); 8.5142 (1.8); 8.3162 (0.4); 8.0191 (2.5); 7.9975 (2.3); 7.7700 (2.5); 7.6194 (2.4); 7.4451 (2.5); 7.1487 (1.0); 7.0091 (2.4); 6.8697 (1.1); 6.0831 (1.0); 6.0656 (1.6); 6.0482 (1.0); 3.3256 (63.1); 2.6755 (1.1); 2.6711 (1.4); 2.6668 (1.1); 2.5244 (4.5); 2.5107 (96.6); 2.5067 (183.9); 2.5022 (231.2); 2.4978 (167.4); 2.3372 (16.0); 2.0747 (1.6); 2.0630 (0.6); 2.0544 (0.7); 2.0422 (1.2); 2.0297 (0.7); 2.0211 (0.6); 2.0089 (0.3); 1.6181 (5.8); 1.6007 (5.8); 1.0415 (0.6); 1.0306 (2.2); 1.0252 (2.3); 1.0097 (2.2); 1.0044 (2.1); 0.9954 (0.6); 0.7772 (0.9); 0.7670 (2.1); 0.7639 (2.2); 0.7548 (2.2); 0.7515 (2.0); 0.7394 (0.7); -0.0002 (8.7); -0.0085 (0.4)

II-62		¹ H-NMR(400.2 MHz, d ₆ -DMSO): δ = 9.1098 (3.4); 9.0924 (3.5); 9.0633 (5.9); 9.0617 (6.4); 9.0579 (6.5); 9.0562 (6.1); 8.5841 (4.9); 8.5786 (4.7); 8.5627 (5.2); 8.5571 (5.2); 8.2327 (16.0); 8.0771 (6.4); 8.0753 (6.5); 8.0557 (6.0); 8.0538 (6.1); 7.4260 (3.9); 7.3433 (7.8); 7.3399 (5.3); 7.3233 (4.4); 7.3186 (5.6); 7.2619 (0.3); 7.2411 (8.2); 7.0562 (9.7); 6.0850 (0.5); 6.0678 (2.4); 6.0504 (3.8); 6.0330 (2.4); 6.0157 (0.5); 5.7571 (0.4); 3.3301 (69.3); 2.6768 (0.5); 2.6723 (0.8); 2.6678 (0.6); 2.5258 (2.3); 2.5211 (3.5); 2.5123 (44.8); 2.5079 (91.2); 2.5034 (119.2); 2.4988 (86.1); 2.4943 (41.8); 2.3348 (0.5); 2.3301 (0.7); 2.3257 (0.5); 2.0146 (0.7); 2.0020 (1.5); 1.9935 (1.7); 1.9899 (1.2); 1.9811 (3.1); 1.9722 (1.2); 1.9686 (1.8); 1.9602 (1.7); 1.9476 (0.8); 1.6354 (14.7); 1.6180 (14.6); 1.0231 (1.8); 1.0122 (5.5); 1.0067 (5.8); 1.0025 (3.0); 0.9963 (3.0); 0.9912 (5.7); 0.9857 (5.5); 0.9755 (2.1); 0.9182 (0.4); 0.9000 (0.9); 0.8817 (0.4); 0.7705 (2.3); 0.7600 (6.1); 0.7547 (5.8); 0.7476 (5.7); 0.7425 (6.0); 0.7313 (1.9); 0.0080 (1.1); -0.0002 (35.2); -0.0085 (1.3)
II-63		II-63: ¹ H-NMR(600.1 MHz, d ₆ -DMSO): δ = 9.0735 (1.5); 9.0617 (1.5); 9.0242 (2.3); 9.0215 (2.3); 8.5352 (1.8); 8.5315 (1.7); 8.5208 (1.8); 8.5171 (1.8); 8.0115 (2.4); 7.9972 (2.3); 7.9964 (2.3); 7.3885 (0.6); 7.3638 (4.2); 7.3395 (2.2); 7.2597 (0.5); 7.2412 (2.8); 7.1179 (1.4); 7.0591 (2.3); 7.0411 (0.4); 6.0612 (1.1); 6.0495 (1.7); 6.0378 (1.1); 5.7560 (0.5); 3.3206 (111.6); 3.2472 (0.4); 3.2375 (0.4); 2.6166 (0.4); 2.6136 (0.6); 2.6107 (0.4); 2.5226 (1.4); 2.5196 (1.8); 2.5165 (1.8); 2.5075 (31.4); 2.5046 (65.2); 2.5016 (90.4); 2.4985 (67.8); 2.4956 (32.4); 2.3885 (0.4); 2.3855 (0.6); 2.3825 (0.4); 2.3372 (16.0); 2.0055 (0.4); 1.9971 (0.8); 1.9915 (0.7); 1.9890 (0.6); 1.9832 (1.3); 1.9749 (0.8); 1.9693 (0.6); 1.6096 (6.0); 1.5980 (6.0); 1.4958 (0.4); 1.3243 (0.4); 1.3119 (0.4); 1.0126 (0.8); 1.0077 (2.4); 1.0040 (2.3); 0.9987 (1.2); 0.9938 (2.3); 0.9901 (2.2); 0.9848 (0.5); 0.9830 (0.5); 0.9117 (0.8); 0.8995 (1.6); 0.8872 (0.7); 0.7739 (0.5); 0.7704 (0.5); 0.7649 (1.1); 0.7561 (2.1); 0.7543 (1.9); 0.7489 (1.9); 0.7458 (1.5); 0.7399 (0.7); -0.0001 (1.2)

¹⁾ 'lowT' denotes that the measurement was conducted at a temperature of 260 Kelvin.

²⁾ 'abs' denotes that the compound was obtained in an enantiomerically enriched or pure form with the major stereoisomer having the absolute configuration depicted in the drawing.

Biological examples**Rhipicephalus (Boophilus) microplus – in-vitro contact tests larval cattle tick (Strain Parkhurst, resistant against synthetic pyrethroids)**

9 mg compound is solved in 1 mL acetone and diluted with acetone to the desired concentration. 250 µL
5 of the test solution is filled in 25 mL glass test tubes and homogeneously distributed on the inner walls by rotation and tilting on a shaking device (2 h at 30 rpm). With a compound concentration of 900 ppm, an inner surface of 44.7 cm² and a homogeneous distribution, a dose of 5 µg/cm² is achieved.

After the solvent has evaporated, each test tube is filled with 20-50 cattle tick larvae (*Rhipicephalus microplus*), closed with a perforated lid and incubated in a horizontal position at 85 % relative humidity
10 and 27 °C in an incubator. After 48 hours efficacy is determined. The larvae are patted on the ground of the tubes and negative geotactic behaviour is recorded. Larvae that climb back to the top of the vial in a manner comparable to untreated control larvae are marked as alive, larvae not climbing back up comparable to untreated control larvae but are moving uncoordinatedly or only twitching their legs are marked as moribund, tick larvae remaining on the bottom and not moving at all are counted as dead.

15 A compound shows a good efficacy against *Rhipicephalus microplus*, if at a compound concentration of 5 µg/cm² an efficacy of at least 80 % is monitored. An efficacy of 100 % means all larvae are dead or moribund; 0 % means no larvae are dead or moribund.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 5 µg/cm² (= 500 g/ha): I-1, I-2, I-3, I-6, I-7, I-9, I-14, I-15, I-16, I-19, I-
20 20, I-22, I-23, I-24, I-25, I-27, I-28, I-30, I-31, I-32, I-33, I-35, I-36, I-37, I-39, I-40, I-42, I-43, I-44, I-49, I-51, I-54, I-55, I-56, I-57, I-58, I-59, I-60, I-61, I-70, I-71, I-74, I-76, I-83, II-7, II-8, II-11, II-12, II-15, II-16, II-17, II-18, II-20, II-21, II-22, II-23, II-25, II-26, II-27, II-29, II-30, II-32, II-33, II-35, II-36, II-37, II-38, II-39, II-40, II-41, II-42, II-43, II-44, II-45, II-46, II-47, II-48, II-49, II-50, II-51, II-52, II-55, II-57, II-58, II-59, II-60, II-61, II-62, II-63.

25 In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 5 µg/cm² (= 500 g/ha): I-18, I-26, I-45, I-47, I-48, I-50, I-72, I-85, II-3, II-10, II-34.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 5 µg/cm² (= 500 g/ha): I-11, I-17, II-9, II-13, II-14, II-53, II-54.

30 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 1 µg/cm² (= 100 g/ha): I-1, I-2, I-3, I-6, I-7, I-9, I-14, I-15, I-16, I-19, I-20, I-22, I-23, I-24, I-25, I-27, I-28, I-30, I-31, I-32, I-33, I-35, I-36, I-37, I-39, I-40, I-42, I-43, I-44, I-49, I-51, I-54, I-55, I-56, I-57, I-58, I-59, I-60, I-61, I-70, I-71, I-72, I-74, I-76, I-83, II-8, II-11, II-15, II-16,

II-17, II-18, II-20, II-21, II-22, II-23, II-25, II-26, II-27, II-29, II-30, II-32, II-33, II-34, II-35, II-36, II-37, II-38, II-39, II-40, II-41, II-42, II-43, II-44, II-45, II-46, II-47, II-48, II-49, II-50, II-51, II-52, II-55, II-57, II-58, II-59, II-60, II-61, II-62, II-63.

5 In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 1 $\mu\text{g}/\text{cm}^2$ (= 100 g/ha): I-4, I-11, I-18, I-26, I-45, I-47, I-48, I-85, II-3, II-54.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 1 $\mu\text{g}/\text{cm}^2$ (= 100 g/ha): I-17, I-50, II-9, II-12, II-53.

Rhipicephalus (Boophilus) microplus – dip test

10 Test animal: cattle ticks (*Rhipicephalus microplus*) strain Parkhurst, SP-resistant

Solvent: dimethyl sulfoxide

To produce a suitable preparation of active compound, 10 mg of active compound are dissolved in 0.5 mL solvent, and the concentrate is diluted with water to the desired concentration.

15 This compound solution is pipetted into tubes. 8-10 engorged, adult, female cattle ticks (*Rhipicephalus microplus*) are placed in perforated tubes. These tubes are immersed in the aqueous compound solution until the ticks are completely moistened. After the liquid has drained off, the ticks are transferred to a filter paper in a plastic tray and stored in a climate chamber.

20 After 7 days egg deposition of fertile eggs is monitored. Eggs where fertility is not visible are stored in a climate chamber till hatching after about 42 days. An efficacy of 100 % means all eggs are infertile; 0 % means all eggs are fertile.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 ppm: I-4, II-13, II-39, II-42, II-47, II-48, II-49.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 100 ppm: I-15.

Rhipicephalus (Boophilus) microplus – Injectiontest

Solvent: dimethyl sulfoxide

To produce a suitable preparation of active compound, 10 mg of active compound are dissolved in 0.5 mL solvent, and the concentrate is diluted with solvent to the desired concentration.

Five adult engorged female ticks (*Rhipicephalus microplus*) are injected with 1 µL compound solution into the abdomen. The ticks are transferred into replica plates and incubated in a climate chamber.

After 7 days egg deposition of fertile eggs is monitored. Eggs where fertility is not visible are stored in a climate chamber till hatching after about 42 days. An efficacy of 100 % means all eggs are infertile; 0 % means all eggs are fertile.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 20 µg/animal: I-1, I-2, I-3, I-4, I-5, I-6, I-7, I-8, I-9, I-11, I-13, I-14, I-15, I-16, I-17, I-18, I-19, I-20, I-21, I-22, I-23, I-24, I-25, I-26, I-28, I-30, I-31, I-32, I-47, II-2, II-3, II-4, II-5, II-6, II-7, II-8, II-9, II-11, II-12, II-13, II-14, II-15, II-16, II-17, II-18, II-19, II-20, II-21, II-22, II-23, II-24, II-25, II-26, II-27, II-28, II-29, II-30, II-31, II-32, II-33, II-34, II-35, II-36, II-37, II-38, II-39, II-40, II-41, II-42, II-43, II-44, II-45, II-46, II-47, II-48, II-49, II-50, II-51.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 20 µg/animal: I-10, I-12, I-33.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 4 µg/animal: I-1, I-2, I-4, I-5, I-6, I-7, I-9, I-11, I-13, I-14, I-15, I-16, I-17, I-18, I-19, I-20, I-21, I-22, I-23, I-24, I-25, I-26, I-27, I-28, I-30, I-31, I-32, I-34, I-35, I-36, I-37, I-39, I-40, I-44, I-45, I-47, I-48, I-51, II-2, II-3, II-4, II-5, II-6, II-7, II-9, II-11, II-12, II-13, II-15, II-16, II-17, II-18, II-19, II-20, II-21, II-22, II-23, II-24, II-25, II-26, II-27, II-28, II-29, II-30, II-31, II-32, II-33, II-34, II-35, II-36, II-37, II-38, II-39, II-40, II-41, II-42, II-43, II-44, II-45, II-46, II-47, II-48, II-49, II-50, II-51, II-52, II-55, II-56, II-57, II-58, II-60, II-61, II-62, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 4 µg/animal: I-8, I-33, II-8, II-14, II-59.

Ctenocephalides felis – in-vitro contact tests adult cat flea

9 mg compound is solved in 1 mL acetone and diluted with acetone to the desired concentration. 250 µL of the test solution is filled in 25 mL glass test tubes and homogeneously distributed on the inner walls by rotation and tilting on a shaking device (2 h at 30 rpm). With a compound concentration of 900 ppm, an inner surface of 44.7 cm² and a homogeneous distribution, a dose of 5 µg/cm² is achieved.

After the solvent has evaporated, each test tube is filled with 5-10 adult cat fleas (*Ctenocephalides felis*), closed with a perforated lid and incubated in a lying position at room temperature and relative humidity. After 48 hours efficacy is determined. The fleas are patted on the ground of the tubes and are incubated on a heating plate at 45-50 °C for at most 5 minutes. Immobile or uncoordinated moving fleas, which are not able to escape the heat by climbing upwards, are marked as dead or moribund.

A compound shows a good efficacy against *Ctenocephalides felis*, if at a compound concentration of 5 µg/cm² an efficacy of at least 80 % is monitored. An efficacy of 100 % means all fleas are dead or moribund; 0 % means no fleas are dead or moribund.

5 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 5 µg/cm² (= 500 g/ha): I-7, I-23, I-25, I-30, I-32, I-45, I-61, II-3, II-6, II-7, II-8, II-11, II-12, II-13, II-16, II-17, II-18, II-20, II-21, II-22, II-23, II-24, II-25, II-27, II-28, II-29, II-31, II-34, II-35, II-36, II-38, II-39, II-40, II-41, II-42, II-43, II-44, II-47, II-49, II-50, II-52, II-53, II-54, II-55, II-59, II-60, II-61, II-63.

10 In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 5 µg/cm² (= 500 g/ha): I-3, I-4, I-34, I-51, I-52, II-9, II-14, II-15, II-26, II-30, II-48.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 5 µg/cm² (= 500 g/ha): I-5, I-24, I-70, II-4, II-45, II-58.

Ctenocephalides felis – oral test

15 Solvent: dimethyl sulfoxide

To produce a suitable preparation of active compound, 10 mg of active compound are dissolved in 0.5 mL solvent, and the concentrate is diluted with cattle blood to the desired concentration.

20 Approximately 20 adult unfed cat fleas (*Ctenocephalides felis*) are placed in flea chambers. The blood chamber, sealed with parafilm on the bottom, are filled with cattle blood supplied with compound solution and placed on the gauze covered top of the flea chamber, so that the fleas are able to suck the blood. The blood chamber is heated to 37 °C whereas the flea chamber is kept at room temperature.

After 2 days mortality in % is determined. 100 % means all the fleas have been killed; 0 % means none of the fleas have been killed.

25 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 ppm: I-1, I-2, I-3, I-4, I-5, I-6, I-7, I-9, I-15, I-16, I-20, I-22, I-23, I-25, I-26, I-27, I-28, I-30, I-32, I-33, I-34, I-35, I-36, I-39, I-48, I-51, II-3, II-4, II-5, II-6, II-7, II-8, II-9, II-11, II-15, II-16, II-17, II-18, II-19, II-22, II-27, II-29, II-30, II-31, II-32, II-33, II-34, II-35, II-36, II-37, II-38, II-39, II-40, II-41, II-42, II-43, II-44, II-45, II-47, II-48, II-49, II-50, II-51, II-52, II-53, II-54, II-55, II-56, II-57, II-58.

30 In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 100 ppm: I-13, I-17, I-19, I-24, I-31, II-46, II-60.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 100 ppm: I-8, I-18, I-47.

Rhipicephalus sanguineus - in-vitro contact tests with adult brown dog ticks

9 mg compound is solved in 1 mL acetone and diluted with acetone to the desired concentration. 250 µL of the test solution is filled in 25 mL glass test tubes and homogeneously distributed on the inner walls by rotation and tilting on a shaking device (2 h at 30 rpm). With a compound concentration of 900 ppm, an inner surface of 44.7 cm² and a homogeneous distribution, a dose of 5 µg/cm² is achieved.

After the solvent has evaporated, each test tube is filled with 5-10 adult brown dog ticks (*Rhipicephalus sanguineus*), closed with a perforated lid and incubated in a lying position at room temperature and relative humidity. After 48 hours efficacy is determined. The ticks are patted on the ground of the tubes and are incubated on a heating plate at 45-50°C for at most 5 minutes. Immotile or uncoordinated moving ticks, which are not able to escape the heat by climbing upwards, are marked as dead or moribund.

A compound shows a good efficacy against *Rhipicephalus sanguineus*, if at a compound concentration of 5 µg/cm² an efficacy of at least 80 % is monitored. An efficacy of 100 % means all ticks are dead or moribund; 0 % means no ticks are dead or moribund.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 5 µg/cm² (= 500 g/ha): I-15, I-16, I-18, I-23, I-30, I-43, I-44, I-60, I-61, II-6, II-23, II-24, II-25, II-26, II-27, II-28, II-29, II-30, II-32, II-34, II-38, II-39, II-40, II-41, II-42, II-47, II-48, II-49, II-50, II-52, II-55, II-57, II-58, II-59, II-60, II-61, II-62, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 5 µg/cm² (= 500 g/ha): I-14, I-19, I-22, I-24, I-33, I-36, I-39, I-57, I-58, I-70, I-83, II-11, II-35, II-36, II-46.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 1 µg/cm² (= 100 g/ha): I-15, I-16, I-23, I-24, I-30, I-36, I-43, I-44, I-57, I-58, I-60, II-23, II-24, II-25, II-26, II-27, II-28, II-30, II-31, II-32, II-34, II-38, II-39, II-40, II-41, II-42, II-46, II-47, II-48, II-49, II-50, II-52, II-54, II-55, II-58, II-59, II-60, II-61, II-62, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 1 µg/cm² (= 100 g/ha): I-14, I-39, I-61, II-6, II-11, II-29, II-33, II-51, II-57.

Diabrotica balteata – spray test

Solvent: 78.0 parts by weight of acetone
1.5 parts by weight of dimethylformamide

Emulsifier: alkylarylpolyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, and the concentrate is diluted with water, containing an emulsifier concentration of 1000 ppm, to the desired concentration. Further test concentrations are prepared by dilution with emulsifier containing water.

Soaked wheat seeds (*Triticum aestivum*) are placed in a multiple well plate filled with agar and some water and are incubated for 1 day to germinate (5 seeds per well). The germinated wheat seeds are sprayed with a test solution containing the desired concentration of the active ingredient. Afterwards each unit is infected with 10-20 larvae of the banded cucumber beetle (*Diabrotica balteata*).

After 7 days efficacy in % is determined. 100 % means all the seedlings have grown up like in the untreated, uninfected control; 0 % means none of the seedlings have grown.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 500 g/ha (= 160 µg/well): I-1, I-3, I-4, I-5, I-7, I-8, I-9, I-10, I-28, II-2, II-3, II-4, II-5, II-6, II-7, II-8, II-9, II-11, II-12, II-13, II-14, II-15, II-16, II-17, II-18, II-19, II-22, II-23, II-25, II-26, II-27, II-28, II-29, II-30, II-31, II-32, II-33, II-34, II-35, II-36, II-37.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 500 g/ha (= 160 µg/well): I-11, II-24.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 g/ha (= 32 µg/well): I-1, I-4, I-5, I-7, I-18, I-22, I-25, I-26, I-27, I-28, I-29, I-30, I-34, I-35, I-36, I-37, I-38, I-40, I-41, I-42, I-43, I-44, I-45, I-46, I-47, I-51, I-53, I-55, I-57, I-58, I-60, I-61, I-62, I-63, I-64, I-65, I-66, I-67, I-68, I-69, I-71, I-73, I-76, I-77, I-78, I-80, I-81, I-82, I-83, I-84, I-85, I-86, I-88, I-90, I-91, I-93, I-94, I-95, I-96, II-3, II-4, II-7, II-8, II-9, II-11, II-12, II-13, II-14, II-15, II-16, II-17, II-18, II-22, II-23, II-26, II-27, II-31, II-32, II-33, II-34, II-35, II-36, II-37, II-38, II-39, II-41, II-43, II-45, II-46, II-47, II-48, II-49, II-50, II-51, II-52, II-54, II-55, II-57, II-58, II-59, II-60, II-61, II-62, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 80 % at an application rate of 100 g/ha (= 32 µg/well): I-8, I-16, I-19, I-20, I-23, I-59, I-74, I-75, II-42.

Meloidogyne incognita - test

Solvent: 125.0 parts by weight of acetone

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

Vessels are filled with sand, a solution of the active ingredient, a suspension containing eggs and larvae of the southern root-knot nematode (*Meloidogyne incognita*) and salad seeds. The salad seeds germinate and the seedlings grow. Galls develop in the roots.

After 14 days the nematicidal activity is determined based on the percentage of gall formation. 100 % means no galls were found and 0 % means the number of galls found on the roots of the treated plants was equal to that in untreated control plants.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 20 ppm: I-18, I-22, I-56, I-83, I-84.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 20 ppm: I-17, I-21, II-20, II-21, II-22, II-60, II-63.

Myzus persicae –oral test

Solvent: 100 parts by weight acetone

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

50 µL compound solution is filled in microtiter plates and 150 µL IPL41 insect medium (33 % + 15 % sugar) is added to obtain a total volume of 200 µL per well. Afterwards the plates are sealed with parafilm through which a mixed population of the green peach aphid (*Myzus persicae*) can suck on the compound preparation.

After 5 days mortality in % is determined. 100 % means all aphids have been killed and 0 % means none of the aphids have been killed.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 4 ppm: I-3, I-4, I-5, I-6, I-7, I-9, I-15, I-16, I-18, I-19, I-22, I-24, I-25, I-29, I-30, I-34, I-38, I-41, I-43, I-45, I-47, I-48, I-49, I-51, I-53, I-54, I-55, I-58, I-59, I-60, I-61, I-62, I-63, I-64, I-65, I-66, I-67, I-68, I-69, I-71, I-74, I-75, I-76, I-79, I-80, I-81, I-82, II-3, II-4, II-9, II-12, II-13, II-15, II-16, II-17, II-18, II-20, II-21, II-22, II-26, II-28, II-29, II-31, II-32, II-33, II-34, II-35, II-36, II-38, II-39, II-41, II-42, II-43, II-44, II-47, II-48, II-49, II-50, II-54, II-55, II-57, II-59, II-60, II-61, II-62, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 4 ppm: I-13, I-26, I-28, I-31, I-44.

Myzus persicae – spray test

Solvent: 78.0 parts by weight acetone
1.5 parts by weight dimethylformamide
Emulsifier: alkylarylpolyglycol ether

- 5 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvents and is diluted with water, containing an emulsifier concentration of 1000 ppm, to the desired concentration. Further test concentrations are prepared by dilution with emulsifier containing water.

Chinese cabbage (*Brassica pekinensis*) leaf disks infected with all instars of the green peach aphid (*Myzus persicae*), are sprayed with a preparation of the active ingredient of the desired concentration.

After 5 days mortality in % is determined. 100 % means all aphids have been killed and 0 % means none of the aphids have been killed.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 500 g/ha: I-3, I-66, I-67, II-3, II-13, II-23, II-26, II-29, II-34, II-35.

- 15 In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 500 g/ha: I-7, I-61, II-4, II-6, II-9, II-12, II-15, II-22, II-24, II-25, II-30, II-32, II-36.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 g/ha: I-3, I-7, I-25, I-41, I-43, I-64, I-66, I-77, I-93, II-3, II-23, II-34, II-35, II-41, II-47, II-48, II-49, II-50, II-54, II-55, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 100 g/ha: I-21, I-23, I-29, I-30, I-38, I-45, I-61, I-63, I-67, I-79, I-80, I-81, I-82, I-91, I-94, I-95, II-6, II-9, II-12, II-13, II-26, II-29, II-38, II-42, II-52, II-61.

Nezara viridula – spray test

- 25 Solvent: 78.0 parts by weight of acetone
1.5 parts by weight of dimethylformamide
Emulsifier: alkylarylpolyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, and the concentrate is diluted with water, containing an emulsifier concentration of 1000 ppm, to the desired concentration. Further test concentrations are prepared by dilution with emulsifier containing water.

Barley plants (*Hordeum vulgare*) infested with larvae of the southern green stink bug (*Nezara viridula*) are sprayed with a test solution containing the desired concentration of the active ingredient.

After 4 days mortality in % is determined. 100 % means all the stink bugs have been killed; 0 % means none of the stink bugs have been killed.

- 5 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 500 g/ha: I-3, I-4, I-7, I-28, I-58, I-59, I-60, I-61, I-61, I-65, I-66, I-67, I-77, I-79, I-80, I-81, I-82, I-86, I-87, I-88, I-89, I-90, I-91, I-93, I-94, I-96, II-3, II-4, II-9, II-12, II-13, II-14, II-15, II-16, II-17, II-18, II-20, II-21, II-22, II-23, II-25, II-26, II-27, II-28, II-29, II-31, II-32, II-33, II-34, II-35, II-36.
- 10 In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 500 g/ha: I-5, I-57, I-72, I-75, I-92, II-7.

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 g/ha: I-4, I-7, I-24, I-25, I-30, I-34, I-38, I-41, I-43, I-57, I-58, I-59, I-60, I-61, I-63, I-64, I-65, I-66, I-67, I-75, I-80, I-81, I-82, II-3, II-4, II-9, II-12, II-13, II-16, II-18, II-20, II-23, II-25, II-26, II-27, II-28, II-29, II-31, II-32, II-33, II-34, II-35, II-36, II-40, II-41, II-42, II-43, II-44, II-45, II-47, II-48, II-49, II-50, II-52, II-54, II-55, II-61.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 100 g/ha: I-5, I-29, I-69, I-79, II-21, II-60.

Nilaparvata lugens – spray test

- 20 Solvent: 78.0 parts by weight of acetone
1.5 parts by weight of dimethylformamide
Emulsifier: alkylaryl polyglycol ether

- To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvents and is diluted with water, containing an emulsifier concentration of 1000 ppm, to the desired concentration. Further test concentrations are prepared by dilution with emulsifier containing water.
- 25

Rice plants (*Oryza sativa*) are sprayed with a preparation of the active ingredient of the desired concentration and the plants are infested with the brown planthopper (*Nilaparvata lugens*).

- After 4 days mortality in % is determined. 100 % means all planthoppers have been killed and 0 % means none of the planthoppers have been killed.
- 30

In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 500 g/ha: I-3, I-77, I-81, I-88, I-95, I-96, II-12, II-13, II-28, II-34.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 500 g/ha: I-7, I-82, I-93, II-22, II-25.

- 5 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 g/ha: I-30, I-38, I-64, II-12, II-13, II-28, II-34, II-49, II-52, II-54.

Spodoptera frugiperda – spray test

- Solvent: 78.0 parts by weight acetone
1.5 parts by weight dimethylformamide
10 Emulsifier: alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvents and is diluted with water, containing an emulsifier concentration of 1000 ppm, to the desired concentration. Further test concentrations are prepared by dilution with emulsifier containing water.

- 15 Maize (*Zea mays*) leaf sections are sprayed with a preparation of the active ingredient of the desired concentration. Once dry, the leaf sections are infested with fall armyworm larvae (*Spodoptera frugiperda*).

After 7 days mortality in % is determined. 100 % means all caterpillars have been killed and 0 % means none of the caterpillars have been killed.

- 20 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 500 g/ha: I-1, I-2, I-3, I-4, I-6, I-7, I-9, I-28, II-1, II-2, II-3, II-5, II-6, II-7, II-8, II-9, II-11, II-12, II-13, II-14, II-15, II-16, II-17, II-18, II-20, II-21, II-22, II-23, II-26, II-27, II-29, II-32, II-34, II-36, II-37.

In this test, for example, the following compounds from the preparation examples showed good activity of 83 % at an application rate of 500 g/ha: I-11, II-31, II-33.

- 25 In this test, for example, the following compounds from the preparation examples showed good activity of 100 % at an application rate of 100 g/ha: I-3, I-4, I-7, I-9, I-11, I-15, I-16, I-19, I-20, I-22, I-23, I-24, I-25, I-27, I-28, I-32, I-35, I-36, I-40, I-43, I-44, I-45, I-47, I-48, I-49, I-51, I-53, I-54, I-55, I-56, I-57, I-58, I-59, I-60, I-61, I-62, I-63, I-64, I-65, I-66, I-67, I-68, I-69, I-71, I-72, I-73, I-74, I-75, I-76, I-77, I-78, I-79, I-80, I-81, I-82, I-83, I-84, I-86, I-87, I-89, I-90, I-91, I-92, I-93, I-94, I-95, I-96, II-7, II-14, II-15, II-16, II-18, II-21, II-22, II-23, II-26, II-27, II-32, II-33, II-36, II-37, II-38, II-39, II-40, II-41, II-43, II-44, II-46, II-47, II-48, II-49, II-50, II-51, II-52, II-55, II-57, II-58, II-59, II-60, II-62, II-63.

In this test, for example, the following compounds from the preparation examples showed good activity of 83 % at an application rate of 100 g/ha: I-2, I-6, I-52, II-4, II-8, II-17, II-20, II-29, II-34, II-53, II-54, II-61.

Tetranychus urticae – spray test OP-resistant

- 5 Solvent: 78.0 parts by weight acetone
1.5 parts by weight dimethylformamide
Emulsifier: alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvents and is diluted with water, containing an emulsifier concentration of 1000
10 ppm, to the desired concentration. Further test concentrations are prepared by dilution with emulsifier containing water.

French bean (*Phaseolus vulgaris*) leaf disks infected with all instars of the two spotted spidermite (*Tetranychus urticae*), are sprayed with a preparation of the active ingredient of the desired concentration.

After 6 days mortality in % is determined. 100 % means all spider mites have been killed and 0 % means
15 none of the spider mites have been killed.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 500 g/ha: II-24.

In this test, for example, the following compounds from the preparation examples showed good activity of 90 % at an application rate of 100 g/ha: II-26, II-33, II-47.

20 **Aedes aegypti test (AEDSAE surface treatment & contact assay)**

Solvent: Aceton + 2000 ppm rapeseed oil methyl ester (RME)

In order to produce a sufficient, active ingredient containing solution it is necessary to solve the test compound in the solvent-mix (acetone at 2 mg/ml / RME 2000 ppm). This solution is pipetted onto a glazed tile and after evaporation of the acetone, adult mosquitoes of the species *Aedes aegypti* strain
25 MONHEIM are placed onto the dried surface. The exposure time is 30 minutes. Mortality in percent (%) is determined 24 hours after contact of the insects to the treated surface. 100% mortality means that all tested insects are dead, whereas 0% means that no insect died.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 20 mg/m²:
I-3, I-4, I-8, I-15, I-23, I-25, I-29, I-30, I-34, I-36, I-47, I-64, I-82, II-3, II-4, II-6, II-7, II-8, II-11, II-12,
30 II-13, II-16, II-17, II-18, II-20, II-21, II-22, II-23, II-24, II-25, II-26, II-27, II-28, II-29, II-30, II-32, II-33, II-34, II-35, II-36, II-37, II-39, II-42, II-43, II-46, II-47, II-48, II-49, II-50, II-52, II-53, II-54, II-57.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 4 mg/m²: I-1, I-4, I-8, I-23, I-25, I-29, I-30, I-34, I-36, I-47, I-81, II-1, II-3, II-4, II-6, II-7, II-8, II-9, II-11, II-12, II-13, II-16, II-17, II-18, II-19, II-20, II-21, II-22, II-23, II-24, II-25, II-26, II-27, II-28, II-29, II-30, II-32, II-33, II-34, II-35, II-36, II-37, II-38, II-39, II-40, II-42, II-43, II-45, II-46, II-47, II-48, II-49, II-50, II-52, II-53, II-54, II-57.

Culex quinquefasciatus test (CULXFA surface treatment & contact assay)

Solvent: Aceton + 2000 ppm rapeseed oil methyl ester (RME)

In order to produce a sufficient, active ingredient containing solution it is necessary to solve the test compound in the solvent-mix (acetone at 2 mg/ml / RME 2 000ppm). This solution is pipetted onto a glazed tile and after evaporation of the acetone, adult mosquitoes of the species *Culex quinquefasciatus* strain P00 are placed onto the dried surface. The exposure time is 30 minutes. Mortality in percent (%) is determined 24 hours after contact of the insects to the treated surface. 100% mortality means that all tested insects are dead, whereas 0% means that no insect died.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 20 mg/m²: I-25, II-3, II-6, II-7, II-12, II-17, II-18, II-20, II-22, II-23, II-24, II-25, II-27, II-32, II-34, II-37, II-39, II-42.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 4 mg/m²: I-15, I-25, II-3, II-6, II-11, II-12, II-13, II-16, II-17, II-18, II-23, II-24, II-25, II-27, II-29, II-32, II-34, II-37, II-42, II-43, II-46.

Anopheles funestus test (ANPHFU surface treatment & contact assay)

Solvent: Aceton + 2000 ppm rapeseed oil methyl ester (RME)

In order to produce a sufficient, active ingredient containing solution it is necessary to solve the test compound in the solvent-mix (acetone at 2 mg/ml / RME 2000 ppm). This solution is pipetted onto a glazed tile and after evaporation of the acetone, adult mosquitoes of the species *Anopheles funestus* strain FUM0Z-R (Hunt et al., Med. Vet. Entomol. 2005 Sep; 19(3): 271-275) are placed onto the dried surface. The exposure time is 30 minutes. Mortality in percent (%) is determined 24 hours after contact of the insects to the treated surface. 100% mortality means that all tested insects are dead, whereas 0% means that no insect died.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 20 mg/m²: I-23, I-29, I-30, I-34, II-3, II-8, II-11, II-12, II-23, II-24, II-25, II-28, II-34, II-42, II-47, II-48, II-49, II-54.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 4 mg/m²: I-23, I-29, I-30, I-34, II-1, II-7, II-11, II-12, II-13, II-16, II-17, II-24, II-25, II-27, II-28, II-30, II-34, II-42, II-47, II-48.

Musca domestica test (MUSCDO surface treatment & contact assay)

- 5 Solvent: Aceton + 2000 ppm rapeseed oil methyl ester (RME)

In order to produce a sufficient, active ingredient containing solution it is necessary to solve the test compound in the solvent-mix (acetone at 2 mg/ml / RME 2000 ppm). This solution is pipetted onto a glazed tile and after evaporation of the acetone, adult flies of the species *Musca domestica* strain WHO-N are placed onto the dried surface. The exposure time is 30 minutes. Mortality in percent (%) is determined 24 hours after contact of the insects to the treated surface. 100% mortality means that all tested insects are dead, whereas 0% means that no insect died.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 20 mg/m²: I-13, I-19, I-23, I-25, I-30, I-36, I-64, I-80, I-81, I-82, II-7, II-14, II-18, II-22, II-23, II-25, II-26, II-27, II-32, II-34, II-35, II-37, II-38, II-39, II-46, II-47, II-48, II-49, II-50, II-52.

- 15 The following examples showed in this test efficacy of 80-100% at a surface concentration of 4 mg/m²: I-19, I-25, I-36, I-64, I-80, I-81, I-82, II-11, II-13, II-22, II-23, II-25, II-26, II-27, II-32, II-35, II-36, II-38, II-39, II-47, II-48, II-49, II-52, II-57.

Blattella germanica test (BLTTGE surface treatment & contact assay)

Solvent: Aceton + 2000 ppm rapeseed oil methyl ester (RME)

- 20 In order to produce a sufficient, active ingredient containing solution it is necessary to solve the test compound in the solvent-mix (acetone at 2 mg/ml / RME 2000 ppm). This solution is pipetted onto a glazed tile and after evaporation of the acetone, adult animals of the species *Blattella germanica* strain PAULINIA are placed onto the dried surface. The exposure time is 30 minutes.

- 25 Mortality in percent (%) is determined 24 hours after contact of the insects to the treated surface. 100% mortality means that all tested insects are dead, whereas 0% means that no insect died.

The following examples showed in this test efficacy of 80-100% at a surface concentration of 20 mg/m²: I-34.