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(54) Title: FORMULATION AND METHOD FOR CONTROLLING VARROA MITES

(57) Abstract: The present disclosure is directed to a formulation and method for preventing or reducing a Varroa mite infestation of a beehive and/or a bee colony. The present disclosure is also directed to a formulation and method for directing or repelling Varroa mites away from bees and/or a beehive. In particular, the method is directed to protecting bees and bee colonies from Varroa mites by utilizing a formulation containing a specific varroacide compound. The varroacide compound comprises a phenyl imidazole, a phenyl pyrrole, a phenyl pyrazole, or a combination thereof.



FORMULATION AND METHOD FOR CONTROLLING VARROA MITES**BACKGROUND**

[0001] Bees, such as honey bees, play a vital role in the agricultural ecosystem. For instance, bees are necessary for the pollination of certain crops and thus have a substantial impact on the agricultural economy. In addition, honey bees also produce honey and wax for consumption. However, bees are threatened worldwide due to various reasons, such as pathogens and parasites. These parasites include at least two species of mites, *Varroa* mites and tracheal mites, which are deadly and can cause the death of an entire colony. In particular, these mites can affect the health of bees and a colony as they can feed on the bees and are also a vector for diseases.

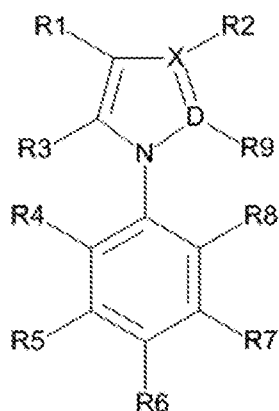
[0002] Various miticides, in particular varroacides, have been employed to eradicate these *Varroa* mites. However, some of these varroacides can be toxic to bees and/or to humans while others are simply not as effective. Meanwhile, varroicide resistance can limit the use of certain chemistries for controlling and/or reducing the risk of infestations in bee colonies. In addition, certain varroacides can only be employed during certain times of the year, for instance, when honey supers are not on the hives (typically before and after nectar flow periods) or when a colony is broodless (typically in the late fall through the mid to later winter).

[0003] As a result, there is a need for an improved formulation and/or method for controlling, directing, and/or repelling *Varroa* mites from beehives and preventing mite infestation in such hives. In particular, there is a need for a formulation and/or method with improved efficacy and selectivity and one that is less toxic, in particular to bees and beekeepers.

SUMMARY

[0004] In general, one embodiment of the present disclosure is directed to a method of preventing or a reducing a *Varroa* mite infestation of a beehive. The method comprises a step of exposing the *Varroa* mite to a formulation comprising a varroicide compound. The varroicide compound comprises a phenyl imidazole, a phenyl pyrrole, a phenyl pyrazole, or a combination thereof.

[0005] In general, according to another embodiment of the present disclosure, the present disclosure is directed to a formulation comprising a varroicide compound. The varroicide compound has the following structure:



[0006] wherein:

[0007] R1 is selected from the group consisting of hydrogen; a halogen; an alkyl group optionally substituted with 1 to 3 halogens; an alkenyl group optionally substituted with 1 to 3 halogens; $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1;

[0008] R12 is selected from the group consisting of a hydrogen and an alkyl group, wherein the alkyl group can be optionally substituted with 1 to 3 halogens;

[0009] X is selected from the group consisting of C and N, with the proviso (a) that when X is N, R2 does not exist and (b) that when X is C, R2 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkyl thio group, and a cyano group;

[00010] D is selected from the group consisting of C and N, with the proviso (a) that when D is N, R9 does not exist and (b) that when D is C, R9 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, and a cyano group;

[00011] R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkenyl group, an alkynyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, and an alkylsulfonyl group.

[00012] Other features and aspects of the present disclosure are discussed in greater detail below.

DETAILED DESCRIPTION

Definitions

[00013] “Alkyl” refers to monovalent saturated aliphatic hydrocarbyl groups having from 1 to 15 carbon atoms, such as from 1 to 10 carbon atoms, such as from 1 to 5 carbon atoms, such as from 1 to 4 carbon atoms, such as from 1 to 3 carbon atoms, such as from 1 to 2 carbon atoms. “C_{x-y} alkyl” refers to alkyl groups having from x to y carbon atoms wherein x and y may be any number from 1 to 15. This term includes, by way of example, linear and branched hydrocarbyl groups such as methyl, ethyl, *n*-propyl, isopropyl, *n*-butyl, isobutyl, *sec*-butyl, *t*-butyl, *n*-pentyl,

neopentyl, and so forth as well as cycloalkyl (e.g., cyclohexyl, cyclopentyl, cyclopropyl, etc.) and/or fused ring.

[00014] “Alkenyl” refers to a linear or branched hydrocarbyl group having from 2 to 15 carbon atoms, such as from 2 to 10 carbon atoms, such as from 2 to 5 carbon atoms and having at least 1 site of vinyl unsaturation ($>C=C<$). “C_{x-y} alkenyl” refers to alkenyl groups having from x to y carbon atoms wherein x and y may be any number from 2 to 15. This term includes, by way of example, ethenyl, propenyl, 1,3-butadienyl, and so forth.

[00015] “Alkynyl” refers to a linear or branched hydrocarbyl group having from 2 to 15 carbon atoms, such as from 2 to 10 carbon atoms, such as from 2 to 5 carbon atoms and having at least one carbon-carbon triple bond ($-C\equiv C-$). “C_{x-y} alkynyl” refers to alkynyl groups having from x to y carbon atoms wherein x and y may be any number from 2 to 15. This term includes, by way of example, ethynyl, propynyl, 2-butyne, 3-butyne, and so forth.

[00016] “Alkoxy” refers to an alkyl attached to a parent molecule through an oxygen atom. For instance, an alkoxy group may be an $-O-$ alkyl group. This term includes, by way of example, methoxy, ethoxy, propoxy, and so forth.

[00017] “Alkylthio” refers to an alkyl attached to a parent molecule through a sulfur atom. For instance, an alkylthio group may be an $-S-$ alkyl group. This term includes, by way of example, methylthio, ethylthio, propylthio, and so forth. This term also includes the various oxidation states of the sulfur atom.

[00018] “Alkylsulfinyl” refers to an alkyl attached to a parent molecule through a sulfinyl group (i.e., $-S(=O)-$). For instance, an alkylsulfinyl group may be an $-S(=O)-$ alkyl group. This term includes, by way of example, methylsulfinyl, ethylsulfinyl, propylsulfinyl, and so forth.

[00019] “Alkylsulfonyl” refers to an alkyl attached to a parent molecule through a sulfonyl group (i.e., $-S(=O)_2-$). For instance, an alkylsulfonyl group may be an $-S(=O)_2-$ alkyl group. This term includes, by way of example, methylsulfonyl, ethylsulfonyl, propylsulfonyl, and so forth.

[00020] “Halogen” refers to a fluoride (F), chloride (Cl), bromide (Br), or iodide (I).

[00021] It should be understood that the aforementioned definitions encompass unsubstituted groups, as well as groups substituted with one or more other functional groups as is known in the art. For example, an alkyl, alkenyl, alkynyl, alkoxy, alkyl thio group may be substituted (e.g., partially or fully) from substituents selected from the group consisting of alkyl, alkenyl, alkynyl, alkoxy, acyl, acylamino, acyloxy, amino, quaternary amino, amide, imino, amidino, aminocarbonylamino, amidinocarbonylamino, aminothiobonyl, aminocarbonylamino, aminothiobonylamino, aminocarbonyloxy, aminosulfonyl, aminosulfonyloxy, aminosulfonylamino, aryl, aryloxy, arylthio, azido, carboxyl, carboxyl ester, (carboxyl ester)amino,

(carboxyl ester)oxy, cyano, cycloalkyl, cycloalkyloxy, cycloalkylthio, guanidino, halo, haloalkyl, haloalkoxy, hydroxy, hydroxyamino, alkoxyamino, hydrazino, heteroaryl, heteroaryloxy, heteroarylthio, heterocyclyl, heterocyclyloxy, heterocyclylthio, nitro, oxo, thione, phosphate, phosphonate, phosphinate, phosphoramidate, phosphorodiamidate, phosphoramidate monoester, cyclic phosphoramidate, cyclic phosphorodiamidate, phosphoramidate diester, sulfate, sulfonate, sulfonyl, substituted sulfonyl, sulfonyloxy, thioacyl, thiocyanate, thiol, alkylthio, etc., as well as combinations of such substituents.

Detailed Description

[00022] Reference now will be made in detail to the embodiments of the invention, one or more examples of which are set forth below. Each example is provided by way of explanation of the invention, not limitation of the invention. In fact, it will be apparent to those skilled in the art that various modifications and variations can be made in the present invention without departing from the scope or spirit of the invention. For instance, features illustrated or described as part of one embodiment, can be used on another embodiment to yield a still further embodiment. Thus, it is intended that the present invention cover such modifications and variations.

[00023] In general, the present invention is directed to a formulation and method for controlling, directing, and/or repelling mites. In one aspect, the method is directed to preventing and reducing a mite infestation in a beehive. In another aspect, the method is directed to treating, preventing and/or reducing a disease, such as varroosis, of bees and bee colonies. In this regard, the present invention is directed to protecting bees, bee colonies, and/or beehives from mites.

[00024] In general, the formulation and methods disclosed herein can be used to protect bees of any genus and species affected by certain mites. For instance, these bees may include bumblebees (genus *Bombus*), mason bees (genus *Osmia*), alfalfa leafcutter bees (genus *Megachile*), honey bees (genus *Apis*), and wild bees.

[00025] In general, in one embodiment, the formulation and method can be used to protect bees from mites that are of the genus *Varroa*. In particular, the mites can be of the species *Varroa destructor*. In another embodiment, the formulation and method can be used to protect bees from tracheal mites (e.g., *Acarapis woodi*, etc.). In one embodiment, the formulation and method can be used to protect bees from tracheal mites and *Varroa* mites.

[00026] In general, *Varroa* mite reproduction begins when the adult female mite enters a brood cell, preferentially a drone brood cell but may also include worker and queen cells, shortly before it is capped. The pheromones emitted by the bee larvae can activate *Varroa* egg development, also referred to as oogenesis. After finalization of oogenesis, the female mite starts to lay eggs. The first egg develops into a male while eggs thereafter develop into female mite

offspring. The nymphs mature into adult female mites in about six days while males need about seven days to mature. Throughout the process, the developing eggs feed off of the bee pupa. The mites develop within the sealed cell and are released from the capped cell when the new bee emerges from the cell. These mites can also attach to the bees, which may serve as intermediate hosts and provide a means of transport to new sites of infestation. Thus, there is a need to prevent *Varroa* mites from being carried and transported by bees and to prevent *Varroa* mites from entering the beehive. By preventing mites from entering the hive, there can be a less likelihood of disease and death of the bee colony. In addition, there is also a need to reduce an infestation of *Varroa* mites once in a beehive and/or bee colony.

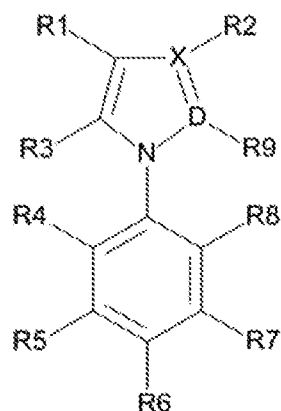
[00027] In this regard, the presently disclosed formulation and method employ a varroacide compound. For instance, in one embodiment, the varroacide compound comprises a phenyl group and a five-membered aromatic ring containing at least one heteroatom, such as nitrogen (e.g., imidazole, pyrrole, pyrazole). According to the present inventors, when employed in an effective amount, these varroacide compounds can control, direct, and/or repel a *Varroa* mite and/or reduce or inhibit a *Varroa* mite infestation. In this regard, the varroacide compound can alter the behavior of the *Varroa* mite.

[00028] Formulation

[00029] A. Varroacide Compound

[00030] In one embodiment, the varroacide compound comprises a phenyl imidazole, a phenyl pyrrole, a phenyl pyrazole, or a combination thereof. In one particular embodiment, the varroacide compound comprises a phenyl imidazole. In one particular embodiment, the varroacide compound comprises a phenyl pyrrole. In one particular embodiment, the varroacide compound comprises a phenyl pyrazole. It should be understood that the varroacide compound may contain any combination of the aforementioned compounds.

[00031] In one embodiment, the varroacide compound has a structure according to formula I



Formula I

[00032] wherein:

[00033] R1 is selected from the group consisting of hydrogen; a halogen; an alkyl group optionally substituted with 1 to 3 halogens; an alkenyl group optionally substituted with 1 to 3 halogens; $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1;

[00034] R12 is selected from the group consisting of a hydrogen and an alkyl group, wherein the alkyl group can be optionally substituted with 1 to 3 halogens;

[00035] X is selected from the group consisting of C and N, with the proviso (a) that when X is N, R2 does not exist and (b) that when X is C, R2 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkylthio group, and a cyano group;

[00036] D is selected from the group consisting of C and N, with the proviso (a) that when D is N, R9 does not exist and (b) that when D is C, R9 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, and a cyano group;

[00037] R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkenyl group, an alkynyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, and an alkylsulfonyl group.

[00038] It should be understood that the above mentioned groups may be optionally substituted. For instance, the hydrogen(s) of the substituent groups, such as the alkyl groups, alkoxy groups, alkylthio groups, etc., may be substituted. For instance, such hydrogen(s) may be substituted with a halogen.

[00039] As indicated above, R1 is selected from the group consisting of a hydrogen; a halogen; an alkyl group (e.g., C_{1-5} alkyl group, such as a C_{1-3} alkyl group, such as a C_{1-2} alkyl group, such as a C_1 alkyl group) optionally substituted with 1 to 3 halogens; an alkenyl group (e.g., C_{2-5} alkenyl group, such as a C_{2-4} alkenyl group, such as a C_{2-3} alkenyl group) optionally substituted with 1 to 3 halogens; $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1. In one embodiment, R1 is a halogen, an alkyl group optionally substituted with 1 to 3 halogens, or $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1. In one particular embodiment, R1 is a halogen or $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1. In one particular embodiment, R1 is a halogen or $-S(O)_nR_{12}$ where n is 0, 1, or 2. In a further embodiment, R1 is $-S(O)_nR_{12}$ where n is 0, 1, or 2. In one particular embodiment, R1 is $-S(O)_nR_{12}$ where n is 1.

[00040] As indicated above, R12 is selected from the group consisting of a hydrogen and an alkyl group, wherein the alkyl group can be optionally substituted with 1 to 3 halogens;. In one embodiment, R12 is an alkyl group. The alkyl group may be a C_{1-5} alkyl group, such as a C_{1-3} alkyl group, such as a C_{1-2} alkyl group, such as a C_1 alkyl group (i.e., methyl group). The alkyl group

may be optionally substituted, such as with at least one halogen. In one embodiment, the alkyl group is substituted, such as with at least one halogen. When substituted, the alkyl group may be partially substituted or fully substituted. In one embodiment, the alkyl group is fully substituted. In one embodiment, R12 is a methyl group substituted with at least 1 halogen, such as at least 2 halogens, such as with 3 halogens. R12 may be, but is not limited to, CH₃, CF₃, CCl₃, CF₂Cl, CFCl₂, CF₂Br, CHF₂, CHCl₂, or CHClF. It should be understood that R12 may be any of these groups. For instance, in one embodiment, R12 is CF₂Cl. For instance, in another embodiment, R12 is CFCl₂.

[00041] As indicated above, X is selected from the group consisting of C and N and D is selected from the group consisting of C and N. In this regard, in one embodiment, the varroacide compound can be a phenyl imidazole such that X is nitrogen (N) and D is carbon (C). Such compound may also be referred to as an aryl imidazole, such as a substituted aryl imidazole. In one embodiment, the varroacide compound can be a phenyl pyrrole such that X is carbon (C) and D is carbon (C). Such compound may also be referred to as an aryl pyrrole, such as a substituted aryl pyrrole. In one embodiment, the varroacide compound can be a phenyl pyrazole such that X is carbon (C) and D is nitrogen (N). Such compound may also be referred to as an aryl pyrazole, such as a substituted aryl pyrazole.

[00042] As indicated above, when X is N, R2 does not exist and when X is C, R2 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkyl thio group, and a cyano group. In one embodiment, X is N such that R2 does not exist. In another embodiment, X is C. In one embodiment, when X is C, R2 is a hydrogen, a halogen, an alkyl group, or a cyano group. In a further embodiment, when X is C, R2 is a halogen, an alkyl group, or a cyano group. In one particular embodiment, when X is C, R2 is a cyano group.

[00043] As indicated above, when D is N, R9 does not exist and when D is C, R9 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkyl thio group, an alkylsulfinyl group, an alkylsulfonyl group, and a cyano group. In one embodiment, D is N such that R9 does not exist. In another embodiment, D is C. In one embodiment, when D is C, R9 is a hydrogen, a halogen, an alkyl group, an alkylsulfinyl group, an alkylsulfonyl group, or a cyano group. In a further embodiment, when D is C, R9 is a hydrogen, a halogen, an alkyl group, or a cyano group. In a further embodiment, when D is C, R9 is a hydrogen, halogen, or an alkyl group. In one particular embodiment, when D is C, R9 is a hydrogen or an alkyl group. In one particular embodiment, when D is C, R9 is a hydrogen.

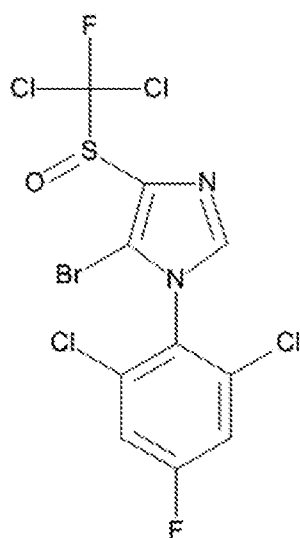
[00044] As indicated above, R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkenyl group, an alkynyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, and an

alkylsulfonyl group. In one embodiment, R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, an alkyl group, and an alkoxy group. In a further embodiment, R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, and an alkyl group. In an even further embodiment, R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen and a halogen.

[00045] In one embodiment, at least 1 of, such as at least 2 of, such as at least 3 of, such as at least 4 of R3, R4, R5, R6, R7, and R8 is a halogen. For instance, in one embodiment, R3, R4, R6, and/or R8 are a halogen. In one particular embodiment, R3, R4, R6, and R8 are a halogen. In one embodiment, R3 is Br. In one embodiment, R4 is Cl. In one embodiment, R8 is Cl. In one embodiment, R6 is F or Cl. In one particular embodiment, R6 is F while in another particular embodiment, R6 is Cl.

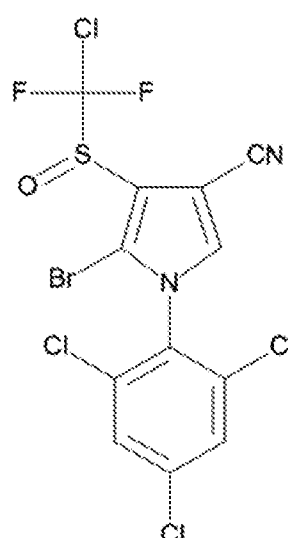
[00046] In one embodiment, at least 1 of, such as at least 2 of R5, R7, and R9 is a hydrogen. In one embodiment, R5, R7, and/or R9 are a hydrogen. In one particular embodiment, R5, R7, and R9 are a hydrogen.

[00047] In one particular embodiment, the varroacide compound is selected from a compound having the following structures:



Formula II

and



Formula III

[00048] In one embodiment, the varroacide compound comprises a phenyl imidazole (or aryl imidazole), such as one having the compound of Formula II. For instance, the varroacide compound may be 1H-imidazole,5-bromo-4-[(dichlorofluoromethyl)sulfinyl]-1-(2,6-dichloro-4-fluorophenyl).

[00049] In one embodiment, the varroacide compound comprises a phenyl pyrrole (or aryl pyrrole), such as one having the compound of Formula III. For instance, the varroacide compound

may be 1H-pyrrole-3-carbonitrile,5-bromo-4-[(chlorodifluoromethyl)sulfinyl]-1-(2,4,6-trichlorophenyl).

[00050] In addition to the compounds mentioned above, it should be understood that stereoisomers (e.g., diastereomers and optical isomers) should also be included.

[00051] These compounds can be prepared using any method known in the art or can be obtained commercially. For instance, methods of synthesis are disclosed in U.S. Patent Nos. 5,187,185 to Outcalt et al. and 5,223,525 to Wu et al., which are both incorporated herein by reference in their entirety.

[00052] Without intending to be limited by theory, it is believed that the varroacide compounds disclosed herein may employ a different mode of action than other conventional varroacides. In addition, it is also believed that the varroacide compounds exhibit unexpectedly improved miticide properties in comparison to other conventional varroacides.

[00053] The varroacide compounds disclosed herein can be employed in a formulation in an amount of 0.01 wt.% or more, such as 0.1 wt.% or more, such as 0.2 wt.% or more, such as 0.5 wt.% or more, such as 1 wt.% or more, such as 1.5 wt.% or more, such as 2 wt.% or less, based on the weight of the formulation. The varroacide compounds disclosed herein can be employed in a formulation in an amount of 50 wt.% or less, such as 40 wt.% or less, such as 30 wt.% or less, such as 20 wt.% or less, such as 15 wt.% or less, such as 10 wt.% or less, based on the weight of the formulation. For instance, the varroacide compounds disclosed herein can be employed in an amount of from 0.01 wt.% to 50 wt.%, such as from 0.1 wt.% to 50 wt.%, such as from 1 wt.% to 40 wt.%, such as from 1 wt.% to 30 wt.%, such as from 2 wt.% to 30 wt.%, such as from 2 wt.% to 20 wt.%, based on the weight of the formulation.

[00054] B. Carrier/Solvent

[00055] The formulation and method disclosed herein may also include an agrochemically acceptable carrier or solvent. These carriers and solvents may include any known in the art that can be combined with the aforementioned varroacide compounds and if any, optional additive components. In addition, these carriers and solvents should minimally detrimentally harm or affect the bees, bee colonies, and/or beehives.

[00056] In one embodiment, the carrier or solvent may be aqueous, non-aqueous, or a combination thereof. In one particular embodiment, the carrier or solvent may be aqueous such that it includes at least 50 wt.% water, such as at least 75 wt.% water, such as at least 90 wt.% water, such as at least 95 wt.% water. In one embodiment, the carrier or solvent may be a mixture of water and a non-aqueous solvent. For instance, when provided as a mixture, the water may be present in the aforementioned amounts, based on the total weight of the carrier or solvent.

[00057] In another embodiment, the carrier or solvent may be non-aqueous. For instance, the carrier or solvent may include an organic solvent or liquid. The carrier or solvent may be comprised of at least 50 wt.%, such as at least 75 wt.%, such as at least 90 wt.%, such as at least 95 wt.% of the organic solvent.

[00058] These solvents may include, but are not limited to, aromatic hydrocarbons (e.g., xylene, phthalates, etc.), aliphatic hydrocarbons (e.g., paraffins, etc.), alcohols (ethanol, isopropanol, etc.), glycols and their ethers and esters (e.g., ethylene glycol, ethylene glycol monomethyl ether, ethylene glycol monoethyl ether, etc.), ketones (e.g., ketones with a straight-chain or branched alkyl group containing from 3 to 20 carbon atoms, such as acetone, 2-hexanone, 2-decanone, 2-undecanone, etc.; cycloalkyl ketones containing from 4 to 9 carbon atoms such as cyclohexanone, cycloheptanone, or a combination thereof), polar aprotic solvents (e.g., dimethylformamide, dimethyl sulfoxide, N-methyl-2-pyrrolidone, etc.), dimethylamides (e.g., dimethylamides containing longer chain alkyls such as C8 (N,N-dimethyloctanamide) and/or C10 (N,N-dimethyldecanamide) alkyls), oils (e.g., vegetable oil such as coconut oil or soybean oil,), or a combination thereof. The solvents may be used alone or in combination.

[00059] In one embodiment, the solvent may include an alcohol, a polar aprotic solvent, a dimethylamide, or a combination thereof. For instance, the solvent may include ethanol, dimethylformamide, dimethyl sulfoxide, N-methyl-2-pyrrolidone, N,N-dimethyloctanamide, N,N-dimethyldecanamide, or a combination thereof. In one particular embodiment, the solvent comprises a polar aprotic solvent, a dimethylamide, or a combination thereof. For instance, the solvent comprises dimethyl sulfoxide, N-methyl-2-pyrrolidone, N,N-dimethyloctanamide, N,N-dimethyldecanamide, or a combination thereof.

[00060] In one embodiment, the agrochemically acceptable carrier may be a solid carrier. These solid carriers may include, but are not limited to, mineral fillers (e.g., talcum, montmorillonite, silica, etc.).

[00061] The solvent or carrier can be present in an amount of 50 wt.% or more, such as 55 wt.% or more, such as 60 wt.% or more, such as 65 wt.% or more, such as 70 wt.% or more, based on the weight of the formulation. The solvent or carrier can be present in an amount of 99.99 wt.% or less, such as 99.9 wt.% or less, such as 99 wt.% or less, such as 98 wt.% or less, such as 97 wt.% or less, such as 95 wt.% or less, such as 90 wt.% or less, based on the weight of the formulation. For instance, the solvent or carrier can be present in an amount of from 50 wt.% to 99.9 wt.%, such as 50 wt.% to 99 wt.%, such as from 50 wt.% to 98 wt.%, such as from 55 wt.% to 97 wt.%, such as from 60 wt.% to 95 wt.%, such as from 60 wt.% to 90 wt.%, based on the weight of the formulation.

[00062] C. Other Components

[00063] In addition, it should be understood that the formulations disclosed herein may contain other auxiliary agents generally known in the art. Such components may be employed to improve upon various properties of the formulations. These properties may include emulsifying properties, wetting properties, and/or dispersing properties.

[00064] For instance, these agents include antioxidants (e.g., ascorbic acid or a salt thereof such as sodium ascorbate), biocides, colorants, deodorizers/odor masking agents, antifreezes and evaporation inhibitors (e.g., glycerol, ethylene and/or propylene glycol), sorbitol, mineral oil, process oils, sodium lactate, penetrants, preservatives, fragrances/masking agents, fillers, carriers, colorants (e.g., pigments and/or dyes), pH modifiers (e.g., buffers, acids, bases), pH dispersants, salts (e.g., calcium, magnesium, ammonium, potassium, sodium, and/or iron chlorides), surfactants (e.g., wetting agents (such as sulfonic acids such as dodecylbenzenesulfonic acid), dispersing agents (such as sulfonates including lignin sulfonates), emulsifiers), suspension agents (such as minerals including silicates, silica), defoamers (such as silicones and polysiloxanes), thickeners (e.g., attapulgite, Xanthan gum, etc.), controlled release agents (e.g., oligosaccharides such as cyclic oligosaccharides such as beta-cyclodextrin), and fertilizers (e.g., ammonium sulfate, ammonium nitrate, urea). These additives can be employed in amounts generally employed for the preparation of such formulations and known to those skilled in the art.

[00065] In one embodiment, the formulation may include an antifreeze agent. The antifreeze agents include glycerol and/or a glycol. For instance, the glycol may be an ethylene glycol and/or a propylene glycol. The antifreeze agent can be employed in an amount of from 1 wt.% to 50 wt.%, such as from 5 wt.% to 40 wt.%, such as from 15 wt.% to 30 wt.%, based on the weight of the formulation. The aforementioned weight percentages may be for a concentrate and/or an end-use dispersion or low concentration formulation.

[00066] As indicated herein, the formulation may include a thickener. The thickener may be any thickener generally employed in an agricultural formulation. For instance, the thickener may be, but is not limited to, xanthan-based thickeners, silicas, and clays such as attapulgite. The thickener can be employed in an amount of from 0.01 wt.% to 10 wt.%, such as from 0.01 wt.% to 8 wt.%, such as from 0.01 wt.% to 5 wt.%, based on the weight of the formulation. The aforementioned weight percentages may be for a concentrate and/or an end-use dispersion or low concentration formulation.

[00067] As indicated herein, the formulation may include a surfactant. The surfactant may be any surfactant generally employed in an agricultural formulation. For instance, the surfactant may be an ionic surfactant, a non-ionic surfactant, or an amphoteric surfactant. Surfactants include, but are not limited to, polyethylene oxide ethers of fatty alcohols, alkyl arylpolyglycol ethers,

ethoxylated alcohols, ethoxylated/propoxylated block polymers, ethoxylated sorbitan or sugar, alkyl sulfate salts (e.g., sodium lauryl sulfate, sodium laureth sulfate, etc.), amine salts of sulfuric acid esters, alkyl sulfonate salts, arylsulfonate salts, sulfonic acids (e.g., dodecylbenzene sulfonic acid, etc.), sulfosuccinates (e.g., sodium dioctyl sulphosuccinate), phosphate esters, etc. In one embodiment, more than one surfactant may be employed within the dispersion. The surfactant can be employed in an amount of from 1 wt.% to 50 wt.%, such as from 5 wt.% to 40 wt.%, such as from 10 wt.% to 30 wt.%, based on the weight of the formulation. The aforementioned weight percentages may be for a concentrate and/or an end-use dispersion or low concentration formulation.

[00068] In one embodiment, the formulation may include a defoamer. The defoamer may be any defoamer generally employed in an agricultural formulation. The defoamer may be a silicone-based compound (e.g., a polysiloxane such as a polydimethylsiloxane), an ethylene oxide/propylene oxide copolymer, a water-based formulation containing oils (e.g., mineral oil and/or vegetable oil) and/or waxes (e.g., long chain fatty alcohol, long chain fatty acid soap or ester), long chain fatty alcohols, long chain fatty acids or esters, and/or a perfluoroalkyl phosphinic and phosphonic acid based compound. In one embodiment, the defoamer may be a silicone-based compound, such as a silicone oil. In one embodiment, the defoamer may be a polydimethylsiloxane, such as a linear polydimethylsiloxane. In general, a polydimethylsiloxane has a backbone with the following formula: $\text{HO}[\text{Si}(\text{CH}_3)_2\text{O}]_n\text{H}$. The end groups may be modified, for example by etherification, or may be attached to $-\text{Si}(\text{CH}_3)_3$ groups. In one embodiment, the defoamer may also contain silica. For instance, the defoamer may include a polydimethylsiloxane and silica. The silica may include polysilicic acids, meta-silicic acid, ortho-silicic acid, silica gel, silicic acid gels, kieselguhr, precipitated SiO_2 , and the like. The defoamer can be employed in an amount of from 0.001 wt.% to 10 wt.%, such as from 0.001 wt.% to 5 wt.%, such as from 0.01 wt.% to 1 wt.%, based on the weight of the formulation. The aforementioned weight percentages may be for a concentrate and/or an end-use dispersion or low concentration formulation.

[00069] The additional components may also include other active ingredients. These active ingredients may include, but are not limited to, another or second varroacide, an insecticide, a fungicide, a pesticide, an insect growth regulator or agent (e.g., those that can control hive beetles), a plant growth regulator, a plant growth enhancement agent, a miticide (e.g., spiromesfen, spirotetramet, etc.) other than a varroacide, etc.

[00070] In one embodiment, the varroacide compounds disclosed herein can be formulated with other varroacides known in the art. For instance, the other varroacides may employ a different mode of action or different chemistry class than the varroacide compounds disclosed

herein. These other varroacides may include, but are not limited to, sevin, coumaphos, amitraz, pyrethroids (e.g., tau-fluvalinate, flumethrin, pyrethrin, etc.), formic acid, thymol, etc.

[00071] Regardless of the components employed, the formulations may be formulated using any method generally known in the art. For instance, the varroacide compound disclosed herein can be combined with the other components as a mixture, such as a tank-mixture. In addition, the varroacide compound may be formulated as an emulsifiable concentrate, a sprayable solution, a dilutable solution (e.g., a concentrate), a dilute emulsion, a wettable powder, a soluble powder, a granulate, an encapsulate, a dust, etc.

[00072] Mite Control

[00073] As disclosed herein, in one embodiment, the present invention is directed to a method for controlling, directing, and/or repelling Varroa mites. In particular, the method is directed to controlling, directing, and/or repelling mites so that they do not attach to bees, will detach from bees, and/or will not enter a beehive containing bees. In one aspect, the method is directed to preventing and/or reducing a Varroa mite infestation in a beehive. In another aspect, the method is directed to preventing and/or reducing a disease, such as varroosis, of bees and bee colonies.

[00074] In one embodiment, a method of “repelling” refers to the ability of the formulation and/or varroacide compound described herein to influence or alter the behavior of a Varroa mite away from an area, location, structure, article, or substance of interest. In order to be classified as “repelling” according to an aspect of the disclosure, it is not necessary that 100% of the mites be directed away from an area, location, structure, article, or substance of interest. In one embodiment, a mite is “repelled” if at least 50%, at least 75%, at least 80%, at least 85%, at least 90%, at least 95%, or at least 98% of the mites are directed away from an area, location, structure, article, or substance of interest.

[00075] In one embodiment, a method of “directing” refers to the ability of the formulation and/or varroacide compound described herein to influence the behavior or movement of a Varroa mite in a given direction. In order to be classified as “directing” according to an aspect of the disclosure, it is not necessary that 100% of the mites be directed away from an area, location, structure, article, or substance of interest. In one embodiment, a mite is “directed away” if at least 50%, at least 75%, at least 80%, at least 85%, at least 90%, at least 95%, or at least 98% of the mites are directed away from an area, location, structure, article, or substance of interest.

[00076] In one embodiment, a method of “controlling” refers to the ability of the formulation and/or varroacide compound described herein to control or alter the behavior of a Varroa mite. In order to be classified as “controlling” according to an aspect of the disclosure, it is not necessary

that 100% of the mites are controlled relative to an area, location, structure, article, or substance of interest. In one embodiment, a mite is "controlled" if at least 50%, at least 75%, at least 80%, at least 85%, at least 90%, at least 95%, or at least 98% of the mite behavior is altered or controlled relative to an area, location, structure, article, or substance of interest.

[00077] In an aspect, a formulation or varroacide compound described herein has the ability to repel or direct a *Varroa* mite away from a structure. Without being limited, a "structure" is an area or locus capable of contacting or being infested by a *Varroa* mite. In another aspect, a "structure" can be man-made or naturally occurring. Without being limited, a "structure" can be a building, for example a commercial or residential building, the interior or exterior of a building, an outdoor space, including but not limited to a field, a park area, a golf course or the like, a car, a boat, a patio, a deck, a table, a chair, a window, a container, a container designed to hold a mite, a container designed to repel, direct, or control a mite, a trashcan or a trash receptacle, an indoor space, such as a room in a commercial or residential building, an outdoor space, or structures oftentimes present in an outdoor space, such as a tree, a bush, a clover, a shrub, a flowering plant, a crop, a wild flower, a wild grass pasture, a fruit tree and/or corresponding area, a lawn, a shed, or other such structure.

[00078] In an aspect, a formulation or varroacide compound described herein has the ability to repel or direct a *Varroa* mite away from an article. Without being limited, an "article" is an element that is capable of being contacted, influenced, or infested by a mite. In another aspect, without being limited, an "article" can be bait, a bait mechanism or holder, a device, garment, belt, air freshener, candle, other scented article, fiber, bedding, sheet, cloth, clothing, net, insect netting, fabric, paper, glass, paint, ink, clay, cotton, woods, furniture, carpet, sanitary good, plastic, polymer, biological or synthetic material, spray product, fogging system, fogger, foam, gel, evaporator product, evaporator system, granule, or dust.

[00079] Stated in another manner, the *Varroa* mites are controlled, directed, and/or repelled so that it changes trajectory, is prevented or inhibited from attaching to a bee and/or entering a beehive, and/or is caused to detach itself from a host bee. In one aspect, the *Varroa* mites are controlled, directed, and/or repelled so as to inhibit mite survival and/or reduce, slow, or stabilize the growth of a mite population.

[00080] The formulation and varroacide compound as described herein can be employed for preventing or reducing a mite infestation, such as a *Varroa* mite infestation, within a beehive. The formulation can be used to reduce, stabilize, or slow the growth of a *Varroa* mite population in a beehive or inhibit the growth, survival, reproduction, or other biological functions of such mites. In one embodiment, the method and formulation kill the *Varroa* mite.

[00081] In particular, the formulation and varroacide compound can provide a reduction in the number of *Varroa* mites (i.e., prevent or inhibit a mite infestation) in a beehive in comparison to a second beehive where mites were not exposed to the formulation and/or varroacide compound described herein. In particular, relative to the second beehive without the presence of or exposure to the formulation and/or varroacide compound, the beehive where mites are exposed to the formulation and/or varroacide compound will have at least about 10% less, such as at least about 25% less, such as at least about 50% less, such as at least about 75% less, such as at least about 90% less, such as at least about 95% less mites. Such second beehive may be within ten meters, such as within 20 meters, such as within 50 meters, such as within 100 meters, such as within 250 meters, such as within 500 meters of the first beehive.

[00082] In addition, the degree of mite infestation in a beehive and/or bee colony can be determined using various methods known in the art. For instance, the degree of infestation can be determined based on the number of mites present in a sample of bees from an infested beehive, the size (e.g., weight and/or size) of bees in an infested beehive in comparison to bees reared in a beehive not infested, amount of honey produced in an infected beehive compared to a healthy hive. However, while these methods may serve as an indirect indicator of the degree of mite infestation, direct indicators may also be employed. For instance, a mite drop test may be conducted to assess the degree of mite infestation wherein the mites present on an adhesive substrate can be counted.

[00083] As indicated herein, the method requires a step of exposing mites to the formulation and/or varroacide compound described herein. Thus, the varroacide compound should be present in an amount effective to control, direct, and/or repel mites and/or to prevent or reduce a mite infestation. For instance, the varroacide compound should be present in an amount to cause a mite to change trajectory, cause a mite to prevent or inhibit the mite from attaching to a bee and/or enter a beehive, cause a mite to detach itself from a host bee, and/or result in a reduction in the number of mites in a beehive. However, the varroacide compound should not be present in an amount that would also repel bees from entering a beehive and/or modifying their behavior.

[00084] According to the present method, the mites can be exposed to the formulation and/or varroacide compound as described herein using any method generally known in the art. In this regard, the formulation and/or varroacide compound should be applied so as to contact the mite and/or be sensed by the mite.

[00085] For instance, the mites can be exposed when the varroacide compound is applied to a desired area upon or through which the mites will traverse or travel.

[00086] For instance, in one embodiment, the formulation can be applied (e.g., by spraying, misting, and the like) on or around a beehive. In one embodiment, the formulation and/or

varroacide compound can be applied on at least a portion of a beehive. In one aspect, the formulation and/or varroacide compound can be applied to an entrance of a beehive, such as a portion of the entrance of a beehive or the entire entrance of a beehive. In one particular embodiment, the formulation and/or varroacide compound can be applied to the entrance of a beehive as illustrated in US Patent Application Publication No. 2008/0280528 to Mudd, which is incorporated herein by reference in its entirety. However, it should be understood that the entrance to the beehive may be configured in any manner that allows for bees to enter/exit the beehive.

[00087] In addition, in one embodiment, the formulation and/or varroacide compound may be ingested by the mites.

[00088] If applied around a beehive, the varroacide compound or formulation can be applied within about 0.1 meters, about 0.5 meters, about 1 meter, about 3 meters, about 5 meters, about 10 meters, about 25 meters, about 50 meters, about 100 meters, about 200 meters, or about 300 meters within the beehive, such as the entrance of a beehive.

[00089] In one embodiment, a dispensing device, such as a strip/substrate containing the formulation and/or varroacide compound can be placed at, on, or adjacent a beehive, such as the entrance of the beehive. The strip/substrate may be made of paper, wood, cardboard, plastic, or other absorbent material that can be moistened, soaked, or impregnated with the formulation and/or varroacide compound. In this regard, the strip/substrate can be placed within about 0.1 meters, about 0.5 meters, about 1 meter, about 3 meters, about 5 meters, about 10 meters, about 25 meters, about 50 meters, about 100 meters, about 200 meters, or about 300 meters within a beehive, such as the entrance of a beehive.

[00090] In one embodiment, the varroacide compound can be provided via a controlled release mechanism. For instance, the varroacide compound can be combined or surrounded by a permeable release rate controlling mechanism that permits the compound to permeate through at a desired rate. The varroacide compound can be provided with a complexation agent and/or with a chelating agent or the compound, can be encapsulated or provided as a host-guest complex. The varroacide compound can be released based on temperature or moisture controlling factors/methods. When provided in such manner, it can be placed within about 0.1 meters, about 0.5 meters, about 1 meter, about 3 meters, about 5 meters, about 10 meters, about 25 meters, about 50 meters, about 100 meters, about 200 meters, or about 300 meters within a beehive, such as the entrance of a beehive.

[00091] In one embodiment, the formulation and/or varroacide compound can be applied (e.g., sprayed) directly on a bee. For instance, the formulation and/or varroacide compound can be applied to the bee via a misting process. In such instance, the varroacide compound is applied

at a low concentration sufficient for controlling, directing, and/or repelling mites but not to modify the behavior of the bees.

[00092] When exposed to the varroacide compound, the mites are controlled, directed, and/or repelled so that they do not attach to bees, will detach from bees, and/or will not enter a beehive containing bees. Thus, as an example, when mites are on the bees and the bees enter the beehives, the mites can be exposed to the varroacide compound and detach from the bees and/or not enter a beehive. Alternatively, mites may not even attach to a bee when sensing the varroacide compound. For this to occur, the mites would have been exposed to the varroacide compound in any manner, such as when applied around the beehive, applied on the beehive (e.g., the entrance), applied on the bees, supplied through a delivery mechanism, etc. In turn, these methods can be employed for preventing and/or reducing a mite infestation in a bee hive.

[00093] As indicated herein, the varroacide compound should be applied or delivered in an amount effective to control, direct, and/or repel or disrupt a mite biological function such as a behavioral activity. For instance, the activity may include repelling a mite so that it does not enter a beehive with the bee, control a mite so that it does not attach to a bee or so that it does detach from a bee, etc. The varroacide compound can be applied or delivered in an amount effective to prevent or reduce a mite infestation in a beehive. However, the behavior of the bee should not be substantially modified. That is, bees should still enter/exit a beehive. In this regard, the varroacide compound is delivered at a concentration below a level that may have a detrimental effect on the bees.

[00094] By exposing the mites to the varroacide compound, without intending to be limited by theory, it should be understood that such exposure can come via any means so long as the mites are exposed to the compound. Thus, "exposing" may mean touching/contacting, associating with, or having proximity to. Without intending to be limited by theory, controlling, directing, and/or repelling activity or the prevention/reduction of a mite infestation may be the result of (1) physical transfer of the formulation and/or varroacide compound to the mite or (2) mites sensing the presence of the formulation and/or varroacide compound.

[00095] In addition, the formulation and/or varroacide compound can be delivered, whether applied/sprayed/misted, through a strip/substrate or other delivery mechanism, to the selected area once or more than once. The formulation and/or varroacide compound may be applied to a target area at least 1 time, such as at least 2 times, such as at least 3 times and 10 times or less, such as 7 times or less, such as 5 times or less. In particular, such applications may be at an interval of 1 day or more, such as 2 days or more, such as 4 days or more, such as 7 days or more and 21 days or less, such as 15 days or less, such as 10 days or less, such as 7 days or less. In order to prevent mites from entering the beehive, cause mites to detach from bees,

prevent mites from attaching to bees, and/or prevent/reduce a mite infestation, it may be desired to apply the formulation and/or varroacide compound two or three times over a complete brood cycle (21 days for workers or 28 days for drones).

[00096] The formulation and/or varroacide compound can be applied manually, such as by an individual (e.g., a beekeeper). Alternatively, a mechanism may be created that allows for remote applications/deliveries (e.g., remote sensing applications/deliveries). For instance, a remote unit containing a reservoir with the formulation and/or varroacide compound can be attached to a delivery device (e.g., sprayer, mister, etc.) and placed in, on or adjacent a beehive or selected area. The delivery device can be connected to a wireless network. A remote user at another location can then transfer a signal to the delivery device through the wireless network and cause the delivery device to deliver the formulation and/or varroacide compound at a desired rate/concentration from the reservoir. Such delivery mechanism is generally referred to as remote sensing and digital application technology.

[00097] In general, the beehives referenced herein may refer to man-made structures that contain a bee colony. The hives can include a bottom board, a cover, and one more boxes stacked one above the other. The boxes contain movable frames of comb or foundation. However, the beehives may have any sort of construction/configuration. Other beehive designs and constructions, including natural cavities inhabited by bees may also be included.

[00098] As indicated herein, the treatment can be applied directly to the bees, on a beehive in proximity to the bees, in a beehive (e.g., to coat the bars, frames, combs, and/or bees as well as the *Varroa* mites located therein), or a selected area adjacent to the beehive. Thus, in proximity or within a selected area adjacent to the beehive can mean within about 1 cm, about 5 cm, about 10 cm, about 100 cm, about 0.2 meters, about 0.5 meters, about 1 meter, about 2 meters, about 5 meters, about 10 meters, about 25 meters, about 50 meters, about 100 meters, about 200 meters, or about 300 meters.

[00099] The varroacide compound and formulations can also be delivered by applying directly on crops, on seeds (e.g., transgenic seeds), on trees (e.g., fruit trees, nut trees, other trees that flower), on crops (e.g., vegetable crops, other crops that flower), etc.

[000100] It should be understood that the quantity of mites present on bees can be determined using any method generally known in the art. These methods can be used to determine the amount of *Varroa* mites present on bees outside of a beehive and/or within a beehive.

[000101] In particular, one method is referred to as the alcohol wash method. Such method can provide the number of mites per a certain quantity of bees (e.g., per 100 bees). According to this method, each sample of bees is placed in a strainer basket and washed using a non-sudsing

or low-sudsing fluid (e.g., ethyl alcohol, low sudsing soap, windshield washer fluid). During the process, mites are dislodged from the bees. The number of mites washed off are counted and recorded. The number of bees that were washed are also counted and recorded. To calculate the number of mites per 100 bees, the total number of mites per sample is divided by the total number of bees in the sample and then multiplied by 100. The result provides the number of mites per 100 bees of the sample. In making such determination, a statistically significant number of bees should be sampled, such as at least about 50 bees, such as 100 bees, such as 150 bees, such as 200 bees, such as 250 bees, such as 300 bees. The bees can be sampled from the desired location (e.g., from within the beehive, immediately prior to the entrance to a beehive, etc.) based on the location upon which the mites are exposed to the formulation and/or varroacide compound.

[000102] As indicated herein, the method can be used to control, direct, and/or repel a *Varroa* mite. In particular, the method disclosed herein can effectively control, direct, and/or repel mites so that they do not attach to bees, will detach from bees, and/or will not enter a beehive containing bees.

[000103] In one embodiment, the formulation and/or varroacide compound described herein control, direct, and/or repel a *Varroa* mite for at least about 10 minutes, about 30 minutes, about 1 hour, about 4 hours, about 8 hours, about 12 hours, about 24 hours, about 2 days, about 3 days, about 7 days, about 14 days, about 21 days, about 1 month, about 2 months, about 3 months or more, or a designated time frame or growing or harvesting season. In one embodiment, the formulation and/or varroacide compound can control, direct, and/or repel a *Varroa* mite such that the mite is not in direct contact for at least the aforementioned times with a treated substance, such as a structure or article, including a beehive, or a treated area. In one embodiment, the formulation and/or varroacide compound can control, direct, and/or repel a mite away from a structure or article, including a beehive, for at least the aforementioned times.

[000104] In one embodiment, the formulation and/or varroacide compound described herein controls, directs, and/or repels *Varroa* mites from about 20% or more, about 30% or more, about 40% or more, about 50% or more, about 60% or more, about 75% or more, about 80% or more, about 90% or more, about 95% or more, about 98% or more, or about 99% or more to about 100% or less, such as about 90% or less, such as about 80% or less, such as about 70% or less relative to a formulation that does not contain an effective amount of the varroacide compound described herein. In one embodiment, the formulation and/or varroacide compound described herein controls, directs, and/or repels a *Varroa* mite by at least the aforementioned percentages and for at least about 5 minutes, about 10 minutes, about 30 minutes, about 1 hour, about 2 hours, about 3 hours, about 4 hours, about 8 hours, about 10 hours, about 15 hours, about 24 hours, about 48 hours, about 72 hours, about 7 days, about two weeks, about one month, about two months, about

three months or more, or during a complete or partial season relative to a formulation that does not contain an effective amount of the varroacide compound described herein.

[000105] In one embodiment, the quantity of *Varroa* mites in a beehive before and after providing the formulation and/or varroacide compound described herein can be determined. Relative to before the formulation and/or varroacide compound is provided, a beehive may have about 20% or less, such as about 30% or less, such as about 40% or less, such as about 50% or less, such as about 60% or less, such as about 70% or less, such as about 80% or less, such as about 90% or less, such as about 95 % or less, such as about 98% or less *Varroa* mites.

[000106] In one embodiment, the formulation and/or varroacide compound reduces mite interaction with a structure (e.g., beehive), article, and/or bees. For instance, the formulation and/or varroacide compound described herein is capable of reducing the interaction of a mite with a structure (e.g., beehive), article, and/or bees for at least about 5 minutes, about 10 minutes, about 30 minutes, about 1 hour, about 2 hours, about 3 hours, about 4 hours, about 8 hours, about 10 hours, about 15 hours, about 24 hours, about 48 hours, about 72 hours, about 7 days, about two weeks, about one month, about two months, about three months or more, or a designated time frame, for example, a growing or harvesting season. In one embodiment, the formulation and/or varroacide compound is capable of reducing the interaction by at least about 20% or more, about 30% or more, about 40% or more, about 50% or more, about 60% or more, about 75% or more, about 80% or more, about 90% or more, about 95% or more, about 98% or more, or about 99% or more relative to a formulation that does not contain an effective amount of the varroacide compound described herein.

[000107] In one embodiment, the formulation and/or varroacide compound is capable of controlling, directing, and/or repelling 20% or more, about 30% or more, about 40% or more, about 50% or more, about 60% or more, about 75% or more, about 80% or more, about 90% or more, about 95% or more, about 98% or more, or about 99% or more *Varroa* mites away from a structure, article, and/or bees for at least about 5 minutes, about 10 minutes, about 30 minutes, about 1 hour, about 2 hours, about 3 hours, about 4 hours, about 8 hours, about 10 hours, about 15 hours, about 24 hours, about 48 hours, about 72 hours, about 7 days, about two weeks, about one month, about two months, about three months or more, or a designated time frame, for example, a growing or harvesting season.

[000108] In addition, the method employed herein should not cause bees to be repelled or directed away from a beehive. In this regard, 75% or less, such as 50% or less, such as 40% or less, such as 30% or less, such as 20% or less, such as 10% or less, such as 5% or less bees are repelled or directed away from a structure or article, including a beehive. For instance, such percentages apply whether the structure or article is treated, the area adjacent the structure or

article is treated, the bee is treated, or a delivery mechanism is placed adjacent to such structure or article. In particular, such bees are not repelled or directed for at least about 10 minutes, about 30 minutes, about 1 hour, about 4 hours, about 8 hours, about 12 hours, about 24 hours, about 2 days, about 3 days, about 7 days, about 14 days, about 21 days, about 1 month, about 2 months, or about 3 months.

[000109] In addition, the varroacide compound and formulation disclosed herein may exhibit a high percent efficacy against mites, such as *Varroa* mites. The percent efficacy can be determined in accordance with the instructions provided in the examples. In this regard, the varroacide compound and formulation disclosed herein may have a percent efficacy against mites of 10% or greater, such as 20% or greater such as 30% or greater, such as 40% or greater, such as 50% or greater, such as 60% or greater, such as 70% or greater. Such percent efficacy can be observed after 3 hours and/or 6 hours. After 6 hours, the percent efficacy may be 30% or greater, such as 50% or greater, such as 60% or greater, such as 70% or greater. In particular, such percent efficacy may be achieved when employing a polar aprotic solvent.

EXAMPLES

Example 1

[000110] The field efficacy of the varroacide compounds of Formula II (1H-imidazole,5-bromo-4-[(dichlorofluoromethyl)sulfinyl]-1-(2,6-dichloro-4-fluorophenyl)) and Formula III (1H-pyrrole-3-carbonitrile,5-bromo-4-[(chlorodifluoromethyl) sulfinyl]-1-(2,4,6-trichlorophenyl)) to *Varroa* mite populations was determined. In the study, a 750-ml plastic container fastened to a wood platform with a removable sticky board was used as a bioassay arena. For the study, approximately 300 bees from the brood frames of a beehive were collected. A 3 cm x 3 cm filter paper suspended in the bioassay arena was treated to the point of saturation (approximately 3 mL of solution with 0.3 mL of the varroacide compound) with a 10% solution (v/v) of the varroacide compound in ethanol (EtOH), dimethyl formamide (DMF), dimethyl sulfoxide (DMSO), or 1-methyl-2-pyrrolidinone (1-MP). The bioassay arena was maintained in a dark environmental chamber at 34 °C to examine the time dependent activity of each chemistry against the *Varroa* mites.

[000111] The effective endpoint was measured as the number of *Varroa* mites collected on the sticky board per 100 bees in the bioassay arena following a 3-hour and a 6-hour exposure period. The bees were washed with an ethanol based solution for 3 minutes following the 6-h exposure period. After the ethanol wash, the number of bees and mites remaining on the bees was counted. The percent efficacy of the treatments was calculated as the number of mites collected on the sticky board divided by the sum of the number of mites collected on the sticky

board and the number of mites remaining on the bees following the 3- and 6-h exposure period. The data is presented in Table 1 below.

[000112] Table 1. % Efficacy after 3 hours and 6 hours of exposure time.

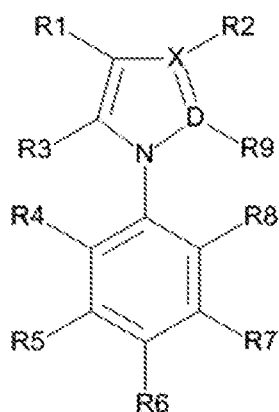
	Formula III		Compound of Formula II	
	% Efficacy after 3 hours	% Efficacy after 6 hours	% Efficacy after 3 hours	% Efficacy after 6 hours
EtOH	12	20	12	20
DMF	18	27	31	63
DMSO	13	31	37	54
NMP	26	36	37	76

[000113] These and other modifications and variations to the present invention may be practiced by those of ordinary skill in the art, without departing from the spirit and scope of the present invention, which is more particularly set forth in the appended claims. In addition, it should be understood that aspects of the various embodiments may be interchanged both in whole or in part.

[000114] Furthermore, those of ordinary skill in the art will appreciate that the foregoing description is by way of example only, and is not intended to limit the invention so further described in such appended claims.

CLAIMS

1. A method of preventing or a reducing a *Varroa* mite infestation of a beehive, the method comprising:
 exposing the *Varroa* mite to a formulation comprising a varroacide compound comprising a phenyl imidazole, a phenyl pyrrole, a phenyl pyrazole, or a combination thereof.
2. The method according to claim 1, wherein the *Varroa* mite is of the species *Varroa destructor*.
3. The method according to claim 1 or claim 2, wherein the varroacide compound is a phenyl imidazole.
4. The method according to claim 1 or claim 2, wherein the varroacide compound is a phenyl pyrrole.
5. The method according to any of the preceding claims, wherein the varroacide compound has the following structure:



wherein:

R1 is selected from the group consisting of hydrogen; a halogen; an alkyl group optionally substituted with 1 to 3 halogens; an alkenyl group optionally substituted with 1 to 3 halogens; $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1;

R12 is selected from the group consisting of a hydrogen and an alkyl group, wherein the alkyl group can be optionally substituted with 1 to 3 halogens;

X is selected from the group consisting of C and N, with the proviso (a) that when X is N, R2 does not exist and (b) that when X is C, R2 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkyl thio group, and a cyano group;

D is selected from the group consisting of C and N, with the proviso (a) that when D is N, R9 does not exist and (b) that when D is C, R9 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, and a cyano group; and

R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkenyl group, an alkynyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, and an alkylsulfonyl group.

6. The method according to claim 5, wherein R4, R6, and R8 are a halogen and R5 and R7 are hydrogen.

7. The method according to any one of claims 5-6, wherein R4 and R8 are Cl and R6 is F or Cl.

8. The method according to any one of claims 5-7, wherein R3 is a halogen.

9. The method according to any one of claims 5-8, wherein R3 is Br.

10. The method according to any one of claims 5-9, wherein X is C and R2 is a cyano group.

11. The method according to any one of claims 5-9, wherein X is N and R2 does not exist.

12. The method according to any one of claims 5-11, wherein R1 is $-S(O)_n C(R_{12})$ where n is 0, 1, or 2.

13. The method according to any one of claims 5-11, wherein R1 is $-S(=O)C(R_{12})$.

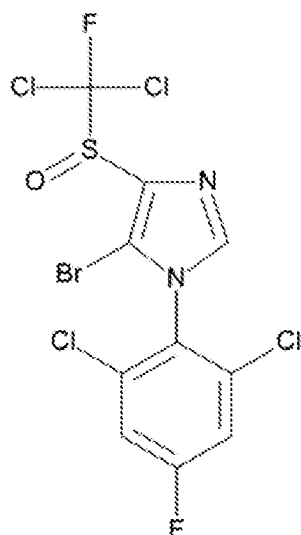
14. The method according to any one of claims 12-13, wherein R12 is an alkyl group.

15. The method according to claim 14, wherein alkyl group is a halogen substituted alkyl group.

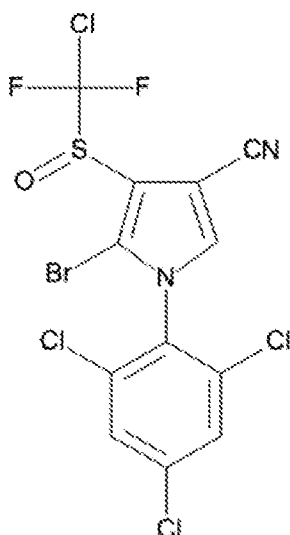
16. The method according to claim 15, wherein the alkyl group is $CFCl_2$.

17. The method according to claim 15, wherein the alkyl group is CF_2Cl .

18. The method according to claim 5, wherein the varroacide compound has the following structure:



19. The method according to claim 5, wherein the varroacide compound has the following structure:



20. The method according to any one of the preceding claims, wherein the varroacide compound is present in the formulation in an amount of 0.1 wt.% to 50 wt.%.

21. The method according to any one of the preceding claims, wherein the formulation further comprises a solvent.

22. The method according to claim 21, wherein the solvent comprises an organic solvent.

23. The method according to any one of claims 21-22, wherein the solvent comprises a polar aprotic solvent.

24. The method according to claim 23, wherein the polar aprotic solvent comprises dimethylformamide, dimethyl sulfoxide, N-methyl-2-pyrrolidone, or a combination thereof.

25. The method according to any one of the preceding claims, wherein the formulation further comprises a second varroacide, an insecticide, a fungicide, a pesticide, an insect growth regulator or agent, a plant growth regulator, a plant growth enhancement agent, a miticide other than a varroacide, or a combination thereof.

26. The method according to any one of the preceding claims, wherein the formulation is applied to at least a portion of a beehive.

27. The method according to any one of the preceding claims, wherein the varroacide compound is applied at a rate of 1 $\mu\text{g}/\text{cm}^2$ to 5 $\mu\text{g}/\text{cm}^2$.

28. The method according to any one of the preceding claims, wherein the formulation is applied to an area around a beehive.

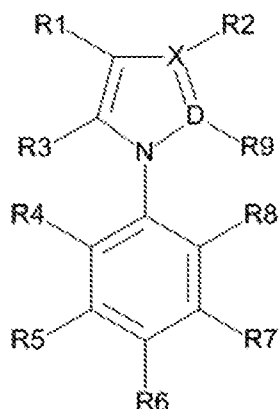
29. The method according to any one of the preceding claims, wherein the formulation is applied to a bee.

30. The method according to any one of the preceding claims, wherein the formulation is applied to a beehive.

31. The method according to any one of the preceding claims, wherein the formulation is applied in a beehive.

32. The method according to any one of the preceding claims, wherein the varroacide compound contacts the *Varroa* mite.

33. A formulation comprising a varroacide compound the following structure:



wherein:

R1 is selected from the group consisting of hydrogen; a halogen; an alkyl group optionally substituted with 1 to 3 halogens; an alkenyl group optionally substituted with 1 to 3 halogens; $-S(O)_nR_{12}$ where n is 0, 1, or 2; and $-C(O)_nR_{12}$ where n is 0 or 1;

R12 is selected from the group consisting of a hydrogen and an alkyl group, wherein the alkyl group can be optionally substituted with 1 to 3 halogens;

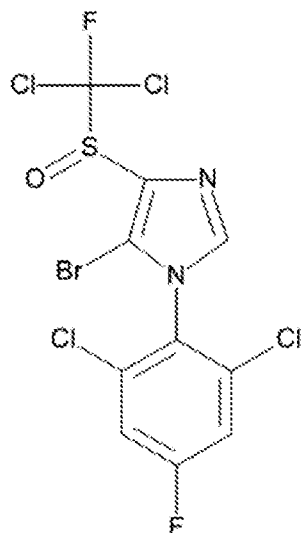
X is selected from the group consisting of C and N, with the proviso (a) that when X is N, R2 does not exist and (b) that when X is C, R2 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkyl thio group, and a cyano group;

D is selected from the group consisting of C and N, with the proviso (a) that when D is N, R9 does not exist and (b) that when D is C, R9 is selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, an alkylsulfonyl group, and a cyano group; and

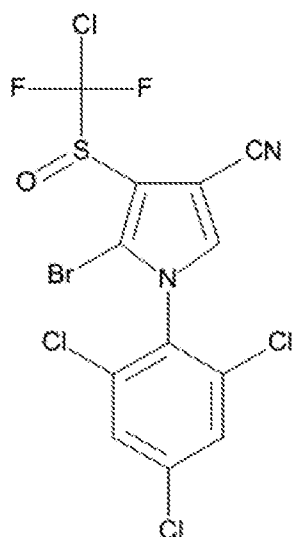
R3, R4, R5, R6, R7, and R8 are each independently of one another selected from the group consisting of a hydrogen, a halogen, an alkyl group, an alkenyl group, an alkynyl group, an alkoxy group, an alkylthio group, an alkylsulfinyl group, and an alkylsulfonyl group.

34. The formulation according to claim 33, wherein R4, R6, and R8 are a halogen and R5 and R7 are hydrogen.

35. The formulation according to any one of claims 33-34, wherein R4 and R8 are Cl and R6 is F or Cl.
36. The formulation according to any one of claims 33-35, wherein R3 is Br.
37. The formulation according to any one of claims 33-36, wherein X is C and R2 is a cyano group.
38. The formulation according to any one of claims 33-36, wherein X is N and R2 does not exist.
39. The formulation according to any one of claims 33-38, wherein R1 is $-S(=O)C(R_{12})$.
40. The formulation according to claim 39, wherein R12 is a halogen substituted alkyl group.
41. The formulation according to claim 40, wherein the alkyl group is $CFCl_2$.
42. The formulation according to claim 40, wherein the alkyl group is CF_2Cl .
43. The formulation according to claim 33, wherein the varroacide compound has the following structure:



44. The formulation according to claim 33, wherein the varroacide compound has the following structure:



45. The formulation according to any one of claims 33-44, wherein the varroacide compound is present in the formulation in an amount of from 0.1 wt.% to 50 wt.%.

46. The formulation according to any one of claims 33-45, wherein the formulation further comprises a solvent.

47. The formulation according to claim 46, wherein the solvent comprises a polar aprotic solvent.

48. The formulation according to claim 47, wherein the polar aprotic solvent comprises dimethylformamide, dimethyl sulfoxide, N-methyl-2-pyrrolidone, or a combination thereof.

49. The formulation according to any one of claims 33-48, wherein the formulation further comprises a second varroacide, an insecticide, a fungicide, a pesticide, an insect growth regulator or agent, a plant growth regulator, a plant growth enhancement agent, a miticide other than a varroacide, or a combination thereof.

50. The formulation according to any one of claims 33-49, wherein the formulation exhibits a percent efficacy of 30% or greater after 6 hours.

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2017/028768

A. CLASSIFICATION OF SUBJECT MATTER INV. A01N43/36 A01N43/50 A01N43/56 A01P7/02 ADD.		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) A01N		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) EPO-Internal, CHEM ABS Data, WPI Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	. ET AL: "ENVIRONMENTAL RISK MANAGEMENT AUTHORITY DECISION", 25 September 2006 (2006-09-25), pages 1-9, XP055374980, New Zealand Retrieved from the Internet: URL: https://www.google.de/url?sa=t&rct=j&q=&esrc=s&source=web&cd=30&cad=rja&uact=8&ved=0ahUKEwi7wMe3u4PUAhVKaFAKHXYyC2k4FBAWCEkwCQ&url=http%3A%2F%2Fwww.epa.govt.nz%2Fse-arch-databases%2FH5NO%2520Application%2520Register%2520Documents%2FH5E06001.doc&usg=AFQjCNGBnK1EE9-B5mpf7ZU4SjmArtmtAg [retrieved on 2017-05-22] the whole document ----- -/-	1-3,5-9, 11-18, 20-36, 38-43, 45-50
☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
* Special categories of cited documents : "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search		Date of mailing of the international search report
24 May 2017		31/07/2017
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Habermann, Jörg

INTERNATIONAL SEARCH REPORT

International application No

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	COURTNEE EDDINGTON ET AL: "Insecticide resistance management in the ectoparasitic mite, varroa destructor of honey bee, apis mellifera populations and susceptibility of this invasive alien pest to fungal pathogens", ESA 2014 SOUTHEASTERN BRANCH MEETING, 3 March 2014 (2014-03-03), pages 1-1, XP055375193, https://esa.confex.com/esa/2014seb/webprogram/Paper80561.html the whole document	1-3,5-9, 11-18, 20-36, 38-43, 45-50
X	----- EP 0 396 427 A1 (RHONE POULENC AGROCHIMIE [FR]) 7 November 1990 (1990-11-07)	33-36, 38-42, 45-50
Y	page 5, line 8 - line 12 page 74, line 15 - line 36 table 2 compounds 8, 18, 27, 30, 39, 40, 96, 106 claims 1-6, 13	1-3,5-9, 11-18, 20-36, 38-43, 45-50
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Y	page 4, line 9 - line 11 page 52, line 24 - line 44 table 1 compound 62 claims 1-3	1-3,5-9, 11-18, 20-36, 38-43, 45-50
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INTERNATIONAL SEARCH REPORT

International application No

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	DATABASE WPI Week 200627 Thomson Scientific, London, GB; AN 2006-263720 XP002770455, -& WO 2006/035808 A1 (NIPPON NOHYAKU CO LTD) 6 April 2006 (2006-04-06) abstract table 1 -----	1-3,5-9, 11-18, 20-36, 38-43, 45-50
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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2017/028768

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

3, 11, 18, 38, 43(completely); 1, 2, 5-9, 12-17, 20-36, 39-42
45-50(partially)

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 3, 11, 18, 38, 43(completely); 1, 2, 5-9, 12-17, 20-36, 39-42, 45-50(partially)

A method of preventing or reducing a Varroa mite infestation of a beehive, the method comprising: exposing the Varroa mite to a formulation comprising a varroacide compound comprising a phenyl imidazole.

2. claims: 4, 19, 44(completely); 1, 2, 5-10, 12-17, 20-37, 39-42, 45-50(partially)

A method of preventing or reducing a Varroa mite infestation of a beehive, the method comprising: exposing the Varroa mite to a formulation comprising a varroacide compound comprising a phenyl pyrrole.

3. claims: 1, 2, 5-10, 12-17, 20-37, 39-42, 45-50(all partially)

A method of preventing or reducing a Varroa mite infestation of a beehive, the method comprising: exposing the Varroa mite to a formulation comprising a varroacide compound comprising a phenyl pyrazole.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2017/028768

Patent document cited in search report		Publication date		Patent family member(s)		Publication date
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