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Pesticidal composition, in particular herbicidal composition comprising at least one essential oil

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Pesticidal composition, in particular herbicidal composition comprising at least one essential oil

5 **Field of the invention**

The invention relates to a pesticidal composition, in particular to a herbicidal composition comprising at least one essential oil and at least one surfactant. The invention also relates to the use of such a pesticidal
10 composition, in particular to the use of such a herbicidal composition.

Description of prior art

Among pests, weeds have been documented as serious plant
15 pests which can reduce crop yields and interfere with plant functions to stop their growth. They constantly compete with crops for water and nutrient resources reducing yields and quality and, consequently, causing huge economic losses which can reach 34% in major crops.

Currently, most reliable weed control methods include mechanical
20 weeding, hand weeding and application of chemical herbicides. These last chemical methods pose several problems with their negative impacts on environmental, animal and human health. Also, the systematic application of chemical herbicides increases the weed's resistance to the applied chemical molecules/substances. Moreover, the alternative methods (thermal, mechanical
25 ...) currently proposed are not very effective over time and the lack of persistence of the applied compositions forces the user to restart the operation at short intervals.

The current trend is to find a biological product to minimize the perceived impacts from synthetic herbicides in agriculture production. In this
30 context, essential oils have been identified as potential natural herbicidal active compounds.

An essential oil is a concentrated hydrophobic liquid containing volatile (defined as the tendency of a substance to vaporize) aroma compounds

from plants. Essential oils are also known as volatile oils, ethereal oils, aetherolea, or simply as the oil of the plant from which they were extracted. An oil is essential in the sense that it contains the essence of the plant's fragrance, i.e. the characteristic fragrance of the plant from which it is derived. Essential oils are generally extracted by distillation, often by using steam. Other processes include expression, solvent extraction, absolute oil extraction, resin tapping, and cold pressing.

Pesticidal compositions and in particular herbicidal compositions comprising at least one essential oil and at least one surfactant are already known and formulated in order to avoid the use of chemical pesticides/herbicides such as pelargonic acid. In this sense, the document WO 2008/141252 describes an organic herbicide comprising natural oils such as clove oil and cinnamon oil and a natural surfactant (casein). The document WO 99/46994 discloses the use of citronella oil as a herbicide in a herbicidal composition comprising a surfactant.

Unfortunately, with such pesticidal compositions, in particular with such herbicidal compositions, only poor results are obtained in term of efficiency, mainly because the spreading and the adhesion of these compositions are not always sufficient and adequate but also because these compositions are often volatile.

Thus, nowadays, there is a need to provide an efficient pesticidal composition, in particular to provide an efficient herbicidal composition, comprising at least one essential oil as an active compound, notably to provide an alternative to the chemical pesticides/herbicides such as pelargonic acid. In parallel, there is a need to provide a pesticidal composition, in particular to provide a herbicidal composition, presenting the right and adequate properties in terms of spreading, adhesion and volatility. More particularly, there is a need to provide a herbicidal composition comprising at least one essential oil as an active compound that presents a pre-emergence herbicidal activity or a post-emergence herbicidal activity, preferably that presents simultaneously a pre-emergence herbicidal activity and a post-emergence herbicidal activity.

Summary of the invention

It is an object of the invention to provide a pesticidal composition, in particular to provide a herbicidal composition as an alternative to the chemical pesticides/herbicides (such as pelargonic acid), comprising at least one essential oil as an active compound and at least one surfactant, such a composition presenting the right and adequate properties in terms of spreading, adhesion and volatility while being at the same time effective both on radical elongation and germination (pre-emergence herbicidal activity) of weeds but also on germinated and developed weeds (post-emergence herbicidal activity).

More particularly, it is an object of the invention to provide a pesticidal composition, in particular to provide a herbicidal composition, comprising at least one essential oil as an active compound, this composition remaining stable and "in place" over time once applied for example on weeds in the fields.

To this end, according to the invention, there is provided a pesticidal composition, in particular a herbicidal composition, comprising at least one essential oil as an active compound and at least one surfactant, said composition further comprising at least one fatty substance.

The terms "active compound" mean that the compound is biologically active, more particularly that the compound is at least active as a herbicide (herbicidal activity). The desired herbicidal activity may be the sort to inhibit seed germination, to prevent the resumption of underground or aerial meristem and bud development, or to inhibit the development and growth of the roots and also of the hypocotyls, the epicotyls and any aerial parts of the plants.

The invention is defined by the independent claims. The dependent claims define advantageous embodiments.

In the context of the present invention, it was surprisingly determined that a composition according to the invention comprising at least one essential oil as an active compound, at least one surfactant and at least one fatty substance, demonstrates an efficient pesticidal activity, in particular an efficient herbicidal activity on weeds. Indeed, it was shown that a composition according to the invention exhibits both a pre-emergence herbicidal activity and

a post-emergence herbicidal activity while being stable and remaining in place over time once applied.

It was also shown that the presence of at least one fatty substance in a composition according to the present invention confers the right and adequate properties in terms of spreading, adhesion and volatility to the composition. In other words, the presence of at least one fatty substance in a pesticidal composition according to the present invention, in particular in a herbicidal composition according to the present invention, was determined to improve the spreading and the adhesion of the composition on the target sites, for example on the leaves of weeds, while reducing the volatility of the composition. The reduction of the volatility of the composition allows to ensure that it remains on the target sites, for example on the leaves of weeds. It was also shown that the presence of a fatty substance in a herbicidal composition according to the present invention enhances at least its postemergence activity on weeds.

Preferably, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one fatty substance is a vegetable oil or a derivate of a vegetable oil or mixtures thereof.

Advantageously, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said vegetable oil is chosen from the group consisting of hazelnut oil, rapeseed oil, pine oil, soybean oil, sunflower oil, pistachio oil, olive oil, almond oil, sweet almond oil, peanut oil, canola oil, safflower oil, cottonseed oil, linseed oil, corn oil, coconut oil, palm oil, grape-seed oil, castor oil, sesame oil and mixtures thereof. This list is not exhaustive.

Preferably, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said derivate of a vegetable oil is chosen from the group consisting of the fatty esters obtained by esterification or transesterification of vegetable oils, fatty amides obtained by amidification or transamidification of vegetable oils, partly epoxidized vegetable oils and mixtures thereof.

By partly epoxidized vegetable oils is meant vegetable oils in which part of the unsaturations have been epoxidized and of which at least two unsaturations have not reacted, preferably between 20 and 80%, more preferably between 30 and 50% of the unsaturations are epoxidized.

5 In general, by (trans)esterification and (trans)amidification is meant reactions which take place on the terminal function of the constituent triglycerides of the vegetable oil, and by epoxidation is meant a reaction taking place on the unsaturations of the vegetable oil.

Advantageously, in a pesticidal composition according to the
10 invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one essential oil is chosen from the group consisting of cinnamon essential oil, citronella essential oil, lemon essential oil, eucalyptus essential oil and mixtures thereof. This list is not exhaustive.

Cinnamon essential oil essentially contains the following
15 molecules:

Cinnamomum cassia: cinnamaldehyde, cynamylacetate and benzaldehyde, eugenol, eugenol acetate, caryophyllene, linalool and phellandrene. This list is not exhaustive.

Citronella essential oil essentially contains the following:
20 molecules: citronellal, geraniol, citronellol, citral and limonene. This list is not exhaustive.

Eucalyptus essential oil essentially contains the following: eucalyptol, isopulegol, pinene (α et β) and p-cymene. This list is not exhaustive.

Preferably, in a pesticidal composition according to the invention
25 herbicidal, in particular in a herbicidal composition according to the invention, said cinnamon essential oil is obtained from the plant Cinnamomum zeylanicum or from the plant Cinnamomum cassia or from the plant Cinnamomum talama or from the plant Cinnamomum aromaticum or from the plant Cinnamomum glaucescens or from the plant Cinnamomum camphora. This list is not
30 exhaustive.

Preferably, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention,

said citronella essential oil is obtained from the plant *Cymbopogon winterianus* or from the plant *Cymbopogon flexuosus*.

Advantageously, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said eucalyptus essential oil is obtained from the plant *Eucalyptus citriodora* or from the plant *Eucalyptus globulus* or from the plant *Eucalyptus radiata*.

According to the invention, said at least one essential oil can be from synthetic origin or from natural origin, for example from an extract or from a fraction of an extract from the roots, the leaves, the stems, the fruits, the seeds and/or the flowers.

The extract used in the context of the invention to obtain an essential oil can be a cellular extract or an extracellular extract of the plant, which can be prepared according to any method known to those skilled in the art for extracting compounds from plant tissues. The extract can be obtained from the roots, the leaves, the stems, the fruits, the seeds and/or the flowers.

Advantageously, the extract is derived from the leaves, the fruits or the seeds, the use of which does not lead to the death of the plant from which they are extracted.

Advantageously, the extract used is obtained by extraction with a solvent selected from water, alcohols, alkanes, ketones, esters, ethers, polyols, chlorinated solvents and mixtures thereof. In particular, extraction may be carried out using a solvent, such as an alcoholic solvent selected from ethanol or methanol or else polyethylene glycol or chloroform.

The extract obtained can then be concentrated, or dried, for example by evaporation or lyophilization. The extract obtained can be incorporated as it is into the composition or else be purified, treated or fractionated, so as to enrich for desired active ingredient.

Advantageously, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one surfactant is a natural surfactant chosen from the group consisting of lecithin, casein, saponines and mixtures thereof.

Alternatively, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one surfactant is a synthetic surfactant chosen from the group consisting of sorbitan monododecanoate poly(oxy-1,2-ethanediyl), alkyl
 5 polyglucosides, sorbitan laurate, polyoxyethylene (20) sorbitan monolaurate, polyoxyethylene (20) sorbitan monopalmitate, polyoxyethylene (20) sorbitan monostearate, polyoxyethylene (20) sorbitan monooleate, polyoxyethylene glycol sorbitan laurate, hexaethylene glycol sorbitan monooleate, polyoxyethylene sorbitan stearate, decaglyceryl monooleate, decaglyceryl
 10 dioleate, polyoxyethylene sorbitan tristearate, monodehydrosorbitol monooleate, sorbitan monolaurate, sorbitan monopalmitate, sorbitan laurate and mixtures thereof. It is understood that any other appropriate surfactant is part of the present invention. This list is not exhaustive.

Commercial names of adequate non-ionic surfactants are for
 15 example: Tween 20®, Tween 80®, Span 80®, Atplus® UEP-100, ethoxylated castor oil, Span 85®, Brij® 93, Brij® S 100, IGEPAL® CA-210, IGEPAL® CO-890, MERPOL® HCS, SP Brij® C2 MBAL-SO-(SG), Triton™ x-405 reduced, Etocas® 5, glycerol monostearate, glyceryl stearate, Emulson® CO 9, Span 20® et Span 80® and mixtures thereof. This list is not exhaustive.

20 The presence of at least one surfactant in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, allows to obtain (1) a stable emulsion showing a good interaction between the hydrophilic and hydrophobic parts of the vegetable oil and the essential oil, (2) small particle size to improve the
 25 penetration of active molecules through epicuticular waxes of the weeds leaves and (3) an improvement on herbicidal activity of essential oils.

Preferably, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one surfactant presents a hydrophilic-lipophilic balance (HLB)
 30 value comprised between 5 and 10, preferably a HLB value comprised between 7 and 9, more preferably a HLB value equal to 8.

Such a HLB value of said at least one surfactant comprised between 5 and 10, preferably comprised between 6 and 8 and more preferably

equal to 7 was determined, in the context of the present invention, to be the adequate HLB value in order to obtain a composition under the form of an emulsion with the right droplet size allowing the composition to effectively penetrate the plant membranes. Preferably, according to the present invention, the composition under the form of an emulsion presents droplets having a droplet size comprised between 0.1 μm et 10 μm , preferably a droplet size comprised between 0.5 μm and 5 μm , more preferably a droplet size comprised between 3 μm and 4 μm .

It was also shown that the compositions according to the invention presenting such a HLB value were more effective in term of pre-emergence and post-emergence herbicidal activity. Also, it was determined that such a HLB value allows to obtain stable composition without the formations of precipitates or supernatants.

Within the meaning of the present invention, the term "HLB" is understood as being an empirical expression which expresses the hydrophilic and hydrophobic (or lipophilic) relationship of a surfactant. The hydrophilic-hydrophobic balance (HLB) of a surfactant expresses its properties: a surfactant has a greater affinity for water if the HLB balance is high (hydrophilic nature) and, conversely, a surfactant has a lower affinity for water (lipophilic or hydrophobic nature) when its HLB value is low.

The determination of the HLB value by calculation allows an arbitrary scale to be defined. The calculation method can be as follows: $\text{HLB} = 20 \times \text{Mh} / \text{M}$. This empirical formula allowing the HLB value to be calculated comprises the ratio between the molecular mass of the hydrophilic group of the surfactant in question (Mh) multiplied by 20 and the molecular mass of the surfactant in question (M).

Starting from this empirical formula, an arbitrary scale is therefore defined and has HLB values comprised between 0 and 20. An HLB value of zero corresponds to a completely lipophilic surfactant, and an HLB value of 20 corresponds to a completely hydrophilic surfactant. A distinction is made, therefore, between surfactants of low, medium and high HLB: a surfactant of low HLB has an HLB value of from 0 to 6; a surfactant of medium HLB has an

HLB value of from 6 to 14 and a surfactant of high HLB has an HLB value of from 14 to 20.

Finally, the HLB value of a surfactant allows the solubility of the surfactant in water or in oil to be indicated and therefore the direction of the emulsion (water-in-oil or oil-in-water) to be determined. For this reason, a surfactant of high HLB will have a greater affinity for water and will therefore be more soluble in water than in oil and vice versa for surfactants of low HLB, which will be more soluble in oil. A surfactant that is more soluble in water than in oil will influence the direction of the "oil-in-water" emulsion and vice versa for a surfactant that is more soluble in oil than in water.

In the sense of the present invention, said at least one surfactant can be a mixture of surfactants. For example, such a mixture of surfactants can be a mixture of a low HLB surfactant with a high HLB surfactant. As an example, a mixture of surfactants can be a mixture of a first surfactant having a HLB value comprised between 3 and 6 so allowing to disperse an aqueous phase in a lipophilic phase with a second surfactant having a HLB value comprised between 15 and 18 so allowing to disperse a lipophilic phase in an aqueous phase.

In the case of a binary mixture of surfactants, the HLB value of this mixture can be calculated as followed:

$$\text{binary mixture HBL} = [m1 / (m1 + m2) \cdot \text{HLB1}] + [m2 / (m1 + m2) \cdot \text{HLB2}]$$

where m1 = mass of the first surfactant

m2 = mass of the second surfactant

HLB1 = HLB of the first surfactant

HLB2 = HLB of the second surfactant

According to the invention, like for compositions only comprising one surfactant having a HLB value comprised between 5 and 10, the binary mixture HLB value is preferably comprised between 5 and 10, more preferably equal to 7. It is understood, that, according to the present invention, the composition may comprise more than two surfactants, for example mixtures of 3 or 4 surfactants.

Preferably, a pesticidal composition, in particular a herbicidal composition according to the invention further comprises an antioxidant agent

chosen from the group consisting of tocopherol, ascorbic acid, sodium ascorbate, calcium ascorbate, citric acid and mixtures thereof. This list is not exhaustive.

5 Advantageously, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one essential oil as an active compound is present in the composition in a proportion ranging from 1% to 20% by weight relative to the total weight of the composition, preferably ranging from 2% to 5% by weight relative to the total weight of the composition.

10 Preferably, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one surfactant is present in the composition in a proportion ranging from 0.1% to 20% by weight relative to the total weight of the composition, preferably ranging from 0.5% to 5% by weight relative to the total weight of the composition.

15 Advantageously, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the invention, said at least one fatty substance is present in the composition in a proportion ranging from 1% to 20% by weight relative to the total weight of the composition, preferably ranging from 2% to 5% by weight relative to the total weight of the composition.

20 Preferably, a pesticidal composition according to the invention, in particular a herbicidal composition according to the invention, further comprises at least one solvent chosen from the group consisting of water, alcohols, ketones, esters, ethers, polyols, ethyl acetate, propylene carbonate, acetonitrile and mixtures thereof.

25 Preferably, the pesticidal composition according to the invention, in particular the herbicidal composition according to the invention, further comprises at least one co-surfactant chosen from the group consisting of ethanol, low molecular weight alcohol from methanol to butanol and mixtures thereof.

30 Advantageously, in a pesticidal composition according to the invention herbicidal, in particular in a herbicidal composition according to the

invention, said at least one co-surfactant is present in the composition in a proportion ranging from 0.01% to 10% by weight relative to the total weight of the composition, preferably ranging from 0.1% to 5% by weight relative to the total weight of the composition, more preferably ranging from 0.5% to 2% by weight relative to the total weight of the composition.

Preferably, the pesticidal composition according to the invention, in particular the herbicidal composition according to the invention, further comprises at least one adjuvant chosen from the group consisting of paraffin wax, Trend® 90, Heliosol®, 1-octyl-2-pyrrolidone, Actirob® B, Atplus® 463, Actilandes™, Vogelux®, Emulsol® and mixtures thereof.

Preferably, the pesticidal composition according to the invention, in particular the herbicidal composition according to the invention, is under the form of microcapsulates, liquids, emulsions or concentrates. Any type of known herbicidal composition may be prepared.

The present invention also relates to the use of a composition according to the invention for obtaining phytotoxic activity comprising inhibiting seed germination, preventing the resumption of underground or aerial meristem and bud development, or inhibiting the development and growth of the roots, the hypocotyls, the epicotyls, the plantlets or aerial parts of the plant. In particular, use may be made of herbicidal compositions in the form of granules, liquids, emulsions or emulsifiable concentrates, as adjuvants for herbicidal mixture.

Short description of the drawings

These and further aspects of the invention will be explained in greater detail by way of examples and with reference to the accompanying figures in which:

Fig.1 shows the remaining quantity (% weight) on a support over time of a composition according to the invention comprising a fatty substance (with FS) in comparison with a composition not comprising a fatty substance (without FS);

- Fig.2 shows the germination rate (%) of different seeds (carrot, chamomile, clover and plantain) treated with two different compositions according to the invention (HE1 and HE2);
- Fig.3 shows the toxicity score measured on Bryophyte seven days after treatment with two different compositions according to the invention (HE1 and HE2);
- Fig.4 shows the toxicity score measured on Pteridophyte (*Equisetum arvense*) seven days after treatment with two different compositions according to the invention (HE1 and HE2);
- Fig.5 shows the lost percentage (%) measured on crimson clover 7 days after treatment with different compositions according to the invention;
- Fig.6 shows the lost percentage (%) measured on crimson clover 7 days after treatment with different compositions according to the invention;
- Fig.7 shows the lost percentage (%) measured on grass 7 days after treatment with different compositions according to the invention;

Detailed description of embodiments of the invention

Volatility of a composition according to the invention

In order to determine the volatility of a composition according to the invention comprising a fatty substance, in comparison with a composition not containing a fatty substance, the weight of 10 drops of each composition was measured every 5 minutes. In practice, 10 drops of 10 μL were put on a small piece of parafilm paper (5x5 cm). The volatility test was performed under a temperature of $20 \pm 2^\circ\text{C}$.

Figure 1 shows the obtained results with two different compositions (see Table 1), i.e. the remaining quantity (% weight) over time of a composition according to the invention comprising a fatty substance (with FS) in comparison with a composition not comprising a fatty substance (without FS).

Table 1 – compositions with and without a fatty substance (% w:w)

	Essential oil (1)	Fatty substance (2)	Surfactant (3)	Co-surfactant (4)	Water	
With FS	3%	3%	2%	0.5%	91.5%	n=3
Without FS	3%	0%	2%	0.5%	94.5%	n=3

(1) *Cinnamomum cassia* (from bark) (Pranarom)

(2) grape oil (Emile Noël)

(3) Tween 20® (Sigma Aldrich)

(4) ethanol 99.5% (Sigma Aldrich)

5 As it can be seen on Figure 1, a composition according to the invention comprising a fatty substance preserves the integrity of the composition once applied by decreasing its volatility. Indeed, higher weights were measured over time for the composition according to the invention (with FS) in comparison with a composition not comprising a fatty substance (without FS). Concretely, this means that a composition according to the invention presents a longer duration of action.

Biological tests

1. Pre-emergence activity

The pre-emergence activity of compositions according to the invention was evaluated on different seeds: carrot (*Daucus carota*), chamomile (*Chamaemelum nobile*), crimson clover (*Trifolium incarnatum*) and plantain (*Plantago lanceolata*).

Filter papers in petri dishes were moistened with two different compositions according to the invention (HE1 and HE2 – see Table 2) while other filter papers were moistened with water (control), before placing seeds in contact with these filter papers.

Table 2 – compositions according to the invention (% w:w)

	Essential oil	Fatty substance (3)	Surfactant (4)	Co-surfactant (5)	Water	
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HE1 (1)	0.75%	0.75%	2%	0.5%	96%	n=5
HE2 (2)	0.75%	0.75%	2%	0.5%	96%	n=5

(1) HE1 = *Cinnamomum cassia* (from bark) (Pranarom)

(2) HE2 = *Cinnamomum zeylanicum* (from leaves) (Pranarom)

(3) grape oil (Emile Noël)

(4) Tween 20® (Sigma Aldrich)

5 (5) ethanol 99.5% (Sigma Aldrich)

A first evaluation of the seeds germination rate on the filter paper was performed 8 days after the placement of the petri dishes in a growth chamber at a constant temperature of 23°C. The ungerminated seeds after 7 days were transposed on water agar in petri dishes placed in a growth chamber at a constant temperature of 23°C and a second assessment was performed 20 days later to complete the evaluation of the germination rate. A seed is considered germinated when the radicle pierces the integuments.

The obtained results are presented in Figure 2. As it can be seen, both the compositions according to the invention HE1 (*Cinnamomum cassia*) and HE2 (*Cinnamomum zeylanicum*) totally inhibit the germination of the tested seeds: compositions according to the invention presents a pre-emergence activity.

20 **2. Post-emergence activity**

Essay 1: post-emergence activity on Bryophyte

In order to assess the post-emergence activity of compositions according to the invention on Bryophyte (adult stage), different compositions according to the invention (HE1 and HE2 – see Table 3) were tested by spraying (100 ml/m²) on Bryophytes (adult stage). Pelargonic acid at a concentration of 30 g/L (PA) (Compo®) was used as a positive control and a composition without the active substance (essential oil) (SSA) was also tested.

30 Table 3 - compositions tested on Bryophyte (% w:w)

	Essential oil	Fatty substance (3)	Surfactant (4)	Co-surfactant (5)	Water	
HE1 (1)	3.4%	3.4%	2%	0.5%	90.7%	n=5
HE2 (2)	3.4%	3.4%	2%	0.5%	90.7%	n=5
SSA	0%	3.4%	2%	0.5%	94.1%	n=5

(1) HE1 = *Cinnamomum cassia* (from bark) (Pranarom)

(2) HE2 = *Cinnamomum zeylanicum* (from leaves) (Pranarom)

(3) grape oil (Emile Noël)

(4) Tween 20® (Sigma Aldrich)

5 (5) ethanol 99.5% (Sigma Aldrich)

The obtained results (toxicity score) are presented in Figure 3. For Bryophytes, it is recognized that the score of 0 represents a healthy and undamaged plant while a score of 6 represents a dead plant. These scores were determined 7 days after treatment with the different compositions.

10 As it can be seen, both the compositions according to the invention HE1 (*Cinnamomum cassia*) and HE2 (*Cinnamomum zeylanicum*) resulted in a toxicity score of 6 equivalent to the score noted with the positive control (pelargonic acid): compositions according to the invention presents a post-emergence activity on Bryophyte.

15

Essay 2: post-emergence activity on Pterydophyte

In order to assess the post-emergence activity of compositions according to the invention on Pterydophyte, the Pterydophyte *Equisetum arvense* (adult stage) was considered. Different compositions according to the invention (HE1 and HE2 – see Table 4) were tested by spraying (100 ml/m²). Pelargonic acid at a concentration of 30 g/L (PA) (Compo®) was used as a positive control and a composition without the active substance (essential oil) (SSA) was also tested.

25 Table 4 - compositions tested on Pterydophyte (% w:w)

	Essential oil	Fatty substance (3)	Surfactant (4)	Co-surfactant (5)	Water	
HE1 (1)	3.4%	3.4%	2%	0.5%	90.7%	n=10
HE2 (2)	3.4%	3.4%	2%	0.5%	90.7%	n=10
SSA	0%	3.4%	2%	0.5%	94.1%	n=10

(1) HE1 = *Cinnamomum cassia* (from bark) (Pranarom)

(2) HE2 = *Cinnamomum zeylanicum* (from leaves) (Pranarom)

(3) grape oil (Emile Noël)

(4) Tween 20® (Sigma Aldrich)

5 (5) ethanol 99.5% (Sigma Aldrich)

The obtained results (toxicity score) are presented in Figure 4. For Pteridophytes, it is recognized that the score of 4 represents a dead plant. These scores were determined 7 days after treatment with the different compositions.

10 As it can be seen, the compositions according to the invention HE1 (*Cinnamomum cassia*) and HE2 (*Cinnamomum zeylanicum*) respectively resulted in a toxicity score of 4 and 3 equivalent to the score noted with the positive control (pelargonic acid): compositions according to the invention presents a post-emergence activity on Pteridophyte.

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Essay 3: post-emergence activity on crimson clover

The post-emergence activity of compositions according to the invention was evaluated on crimson clover (*Trifolium incarnatum*) (2-3 leaves stage). By spraying (100 ml/m²), different compositions according to the invention were tested (see Table 5). Pelargonic acid at a concentration of 30 g/L (PA) (Compo®) was used as a positive control and compositions without the active substance (essential oil) (SSA) were also tested. Water was used as a negative control. After treatment with the different compositions, plants were maintained under the following conditions: temperature of 27 ± 3°C and RH (Relative

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Humidity) of $60 \pm 5\%$ and the post-emergence activity was evaluated 7 days after the treatment with the different compositions.

Table 5 - compositions tested on crimson clover (% w:w)

	Essential oil	Fatty substance	Surfactant (1)	Co-surfactant (2)	Water	
HE1, Ra	3%	Ra 3%	2%	0.5%	91.5%	n=3
HE2, Ra	3%	Ra 3%	2%	0.5%	91.5%	n=3
HE3, Ra	3%	Ra 3%	2%	0.5%	91.5%	n=3
SSA, Ra	0%	Ra 3%	2%	0.5%	94.5%	n=3
HE1, No	3%	No 3%	2%	0.5%	91.5%	n=3
SSA, No	0%	No 3%	2%	0.5%	94.5%	n=3
HE1, Li	3%	Li 3%	2%	0.5%	91.5%	n=3
SSA, Li	0%	Li 3%	2%	0.5%	94.5%	n=3

5 HE1 = *Cinnamomum cassia* (from bark) (Floressence)

HE2 = *Cinnamomum zeylanicum* (from leaves) (Floressence)

HE3 = *Cymbopogon winterianus* (Floressence)

Ra = grape oil; No = hazelnut oil ; Li = linseed oil (Emile Noël)

(1) Tween 20® (Sigma Aldrich)

10 (2) ethanol 99.5% (Sigma Aldrich)

The obtained results are presented in Figure 5. As it can be seen, the compositions HE1, RA; HE2, RA; HE1, No and HE1, Li all presented a high post-emergence activity. In particular, the obtained results allowed to highlight the fact that different oils (RA, No and Li) can be used since similar lost percentages were noted for the essential oil HE1 when mixed with these different oil (fatty substance). The presented results also demonstrated that the active substance, i.e. the essential oil, is essential for the post-emergence activity since the tests without essential oil (SSA) led to 0% lost percentage.

Essay 4: post-emergence activity on crimson clover

The post-emergence activity of compositions according to the invention was evaluated on crimson clover (*Trifolium incarnatum*) (2-3 leaves stage). By spraying (100 ml/m²), different compositions according to the invention were tested (see Table 6) were tested. Pelargonic acid at a concentration of 30 g/L (PA) (Belchim Crop Protection) was used as a positive control as well as two other positive controls (Beloukha® and Compo®) and compositions without the active substance (essential oil) (SSA) was also tested. Water was use as a negative control. After treatment with the different compositions, plants were maintained under the following conditions: temperature of 27 ± 3°C and RH (Relative Humidity) of 60 ± 5% and the post-emergence activity was evaluated 7 days after the treatment with the different compositions.

Table 6 - compositions tested on crimson clover (% w:w)

	Essential oil	Fatty substance (1)	Surfactant	Co-surfactant (4)	Water	
HE1 Bio	3%	3%	2% (2)	0.5%	91.5%	n=4
HE1 Bio-EtOh	3%	3%	2% (2)	0%	92%	n=4
HE1 Bio-HV	3%	0%	2% (2)	0.5%	94.5%	n=4
HE1 Bio-EtOh&HV	3%	0%	2% (2)	0%	95%	n=4
HE2 Bio	3%	3%	2% (2)	0.5%	91.5%	n=4
HE3 Bio	3%	3%	2% (2)	0.5%	91.5%	n=4
SSA Bio	0%	3%	2% (2)	0.5%	94.5%	n=4
HE1	3%	3%	2% (3)	0.5%	91.5%	n=4

HE1 = *Cinnamomum cassia* (from bark) (Floressence)

HE2 = *Cinnamomum zeylanicum* (from leaves) (Floressence)

HE3 = *Cymbopogon winterianus* (Floressence)

(1) = grape oil (Emile Noël)

(2) Soy lecithin (MP Biomedicals)

(3) Tween 20® (Sigma Aldrich)

5 (4) ethanol 99.5% (Sigma Aldrich)

The obtained results are presented in Figure 6. As it can be seen, all the compositions according to the present invention were effective with at least 50% lost percentage and most of these compositions allowed to reach lost percentages equivalent to those observed with the positive controls (PA, Compo, Bel.).

It is important to note the presence of a fatty substance (i.e., a vegetable oil) increased the post-emergence activity of a composition according to the present invention. Indeed, when comparing the results obtained for HE1 Bio-EtOh (essential oil + fatty substance + surfactant) to the results obtained for HE1 Bio-EtOh&HV (essential oil + surfactant), it can be concluded that the fatty substance enhances the post-emergence activity since the lost percentage is significantly increased with the composition Bio-EtOh (essential oil + fatty substance + surfactant).

Another point is that the use of a natural surfactant (from natural origin) or of a synthetic surfactant gave different results: the use of soy lecithin as natural surfactant (HE1 Bio) instead of Tween 20® (HE1) allowed to observe a significant higher lost percentage.

Essay 5: post-emergence activity on grass

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The post-emergence activity of compositions according to the invention was evaluated on grass (adult stage). By spraying (100 ml/m²), different compositions according to the invention were tested (see Table 7). Pelargonic acid at a concentration of 30 g/L (PA) (Belchim Crop Protection) was used as a positive control as well as two other positive controls (Beloukha® and Compo®) and compositions without the active substance (essential oil) (SSA) was also tested. Water was used as a negative control. After treatment with the different compositions, plants were maintained under the following conditions:

temperature of $27 \pm 3^\circ\text{C}$ and RH (Relative Humidity) of $60 \pm 5\%$ and the post-emergence activity was evaluated 7 days after the treatment with the different compositions.

5 Table 7 - compositions tested on crimson clover (% w:w)

	Essential oil	Fatty substance (1)	Surfactant	Co-surfactant (4)	Water	
HE1 Bio	3%	3%	2% (2)	0.5%	91.5%	n=4
HE2 Bio	3%	3%	2% (2)	0.5%	91.5%	n=4
HE3 Bio	3%	3%	2% (2)	0.5%	91.5%	n=4
SSA Bio	0%	3%	2% (2)	0.5%	94.5%	n=4
HE1	3%	3%	2% (3)	0.5%	91.5%	n=4

HE1 = *Cinnamomum cassia* (from bark) (Floressence)

HE2 = *Cinnamomum zeylanicum* (from leaves) (Floressence)

HE3 = *Cymbopogon winterianus* (Floressence)

(1) = grape oil (Emile Noël)

10 (2) Soy lecithin (MP Biomedicals)

(3) Tween 20® (Sigma Aldrich)

(4) ethanol 99.5% (Sigma Aldrich)

The obtained results are presented in Figure 7. As it can be seen, all the compositions according to the present invention showed a post-emergence activity similar to the one obtained with the positive controls (PA, Compo, Bel.).

The present invention has been described in terms of specific embodiments, which are illustrative of the invention and not to be construed as limiting. More generally, it will be appreciated by persons skilled in the art that the present invention is not limited by what has been particularly shown and/or described hereinabove.

Use of the verbs "to comprise", "to include", "to be composed of", or any other variant, as well as their respective conjugations, does not exclude the presence of elements other than those stated.

Use of the article "a", "an" or "the" preceding an element does not exclude the presence of a plurality of such elements.

Claims

1. Pesticidal composition, in particular herbicidal composition, comprising at least one essential oil as an active compound and at least one surfactant,
5 characterized in that it further comprises at least one fatty substance.
2. Pesticidal composition, in particular herbicidal composition, according to claim 1, characterized in that said at least one fatty substance is a vegetable oil or a derivate of a vegetable oil or mixtures thereof.
- 10 3. Pesticidal composition, in particular herbicidal composition, according to claim 2, characterized in that said vegetable oil is chosen from the group consisting of hazelnut oil, rapeseed oil, pine oil, soybean oil, sunflower oil, pistachio oil, olive oil, almond oil, sweet almond oil, peanut oil, canola oil, safflower oil, cottonseed oil, linseed oil, corn oil, coconut oil, palm oil, grape-
15 seed oil, castor oil, sesame oil and mixtures thereof.
4. Pesticidal composition, in particular herbicidal composition, according to claim 2, characterized in that said derivate of a vegetable oil is chosen from
20 the group consisting of the fatty esters obtained by esterification or transesterification of vegetable oils, fatty amides obtained by amidification or transamidification of vegetable oils, partly epoxidized vegetable oils and mixtures thereof.
- 25 5. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that said at least one essential oil is chosen from the group consisting of cinnamon essential oil, citronella essential oil, lemon essential oil, eucalyptus essential oil and mixtures thereof.
- 30 6. Pesticidal composition, in particular herbicidal composition, according to claim 5, characterized in that said cinnamon essential oil is obtained from the plant *Cinnamomun zeylanicum* or from the plant *Cinnamomun cassia* or from the plant *Cinnamomun talama* or from the plant *Cinnamomun*

aromaticum or from the plant *Cinnamomum glaucescens* or from the plant *Cinnamomum camphora*.

- 5 7. Pesticidal composition, in particular herbicidal composition, according to claim 5, characterized in that said citronella essential oil is obtained from the plant *Cymbopogon winterianus* or from the plant *Cymbopogon flexuosus*.
- 10 8. Pesticidal composition, in particular herbicidal composition, according to claim 5, characterized in that said eucalyptus essential oil is obtained from the plant *Eucalyptus citriodora* or from the plant *Eucalyptus globulus* or from the plant *Eucalyptus radiata*.
- 15 9. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that said at least one surfactant is a natural surfactant chosen from the group consisting of lecithin, casein, saponines and mixtures thereof.
- 20 10. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that said at least one surfactant presents a hydrophilic-lipophilic balance (HLB) value comprised between 5 and 10, preferably a HLB value comprised between 7 and 9, more preferably a HLB value equal to 8.
- 25 11. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that it further comprises an antioxidant agent chosen from the group consisting of tocopherol, ascorbic acid, sodium ascorbate, calcium ascorbate, citric acid and mixtures thereof.
- 30 12. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that the composition further comprises at least one solvent chosen from the group consisting of water, alcohols, ketones, esters, ethers, polyols, ethyl acetate, propylene carbonate, acetonitrile and mixtures thereof.

13. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that the composition further comprises at least one co-surfactant chosen from the group consisting of ethanol, low molecular weight alcohol from methanol to butanol and mixtures thereof.
14. Pesticidal composition, in particular herbicidal composition, according to any of preceding claims, characterized in that the composition is under the form of microcapsulates, liquids, emulsions or concentrates.
15. Use of a composition according to any claims 1 to 14 for obtaining phytotoxic activity comprising inhibiting seed germination, preventing the resumption of underground or aerial meristem and bud development, or inhibiting the development and growth of the roots, the hypocotyls, the epicotyls, the plantlets or aerial parts of the plant.

Abstract**Pesticidal composition, in particular herbicidal composition comprising at least one essential oil**

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The present invention relates to a herbicidal composition comprising at least one essential oil as an active compound and at least one surfactant, said composition further comprising at least one fatty substance.

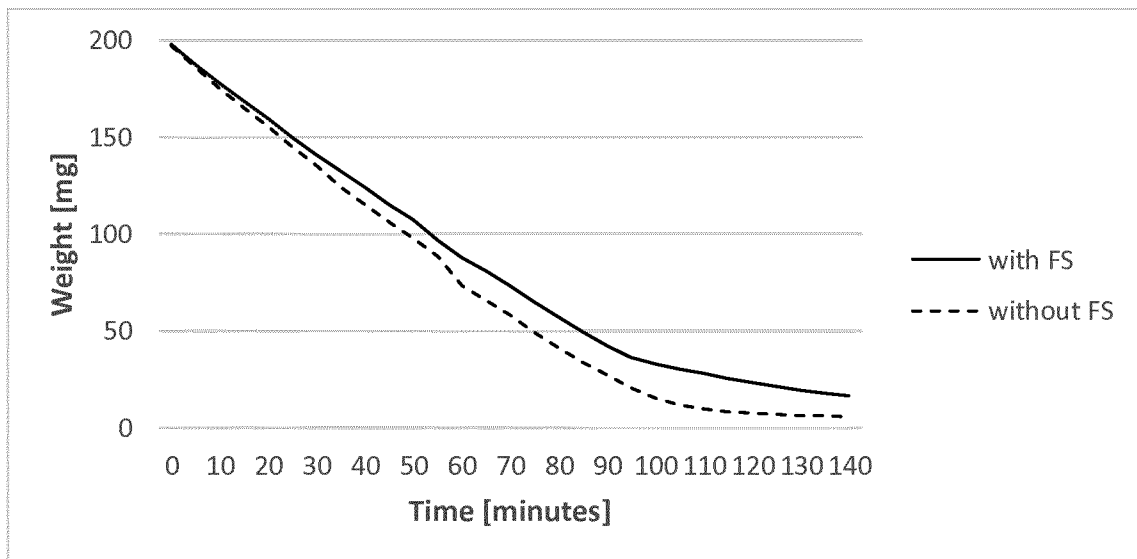


FIG. 1

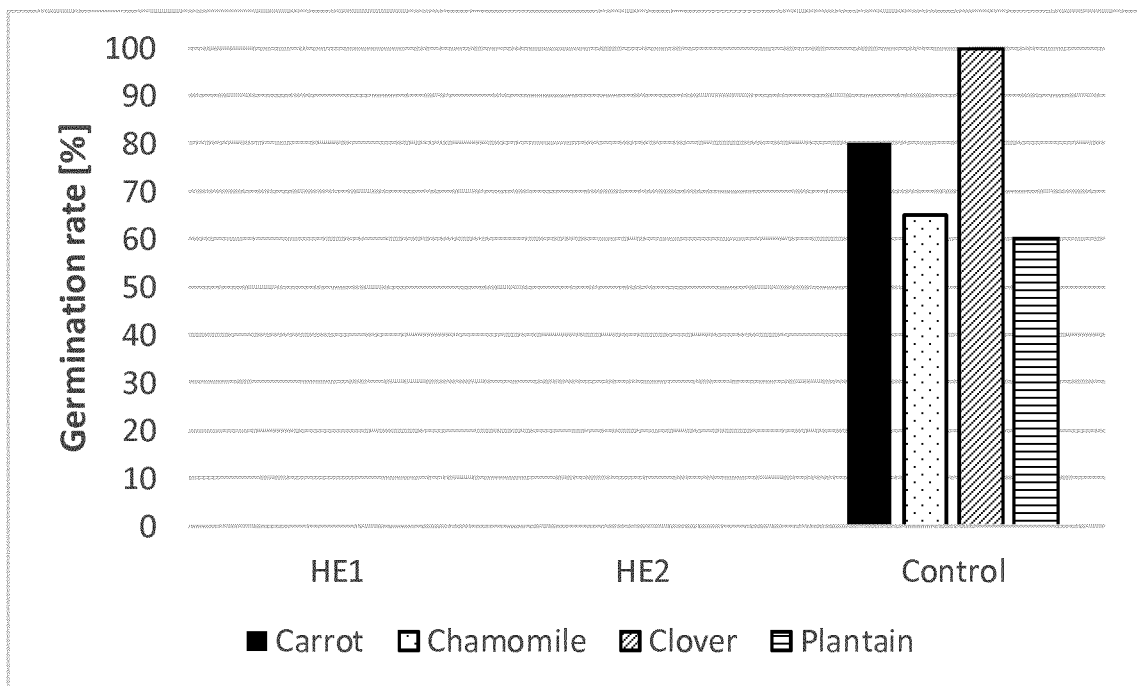
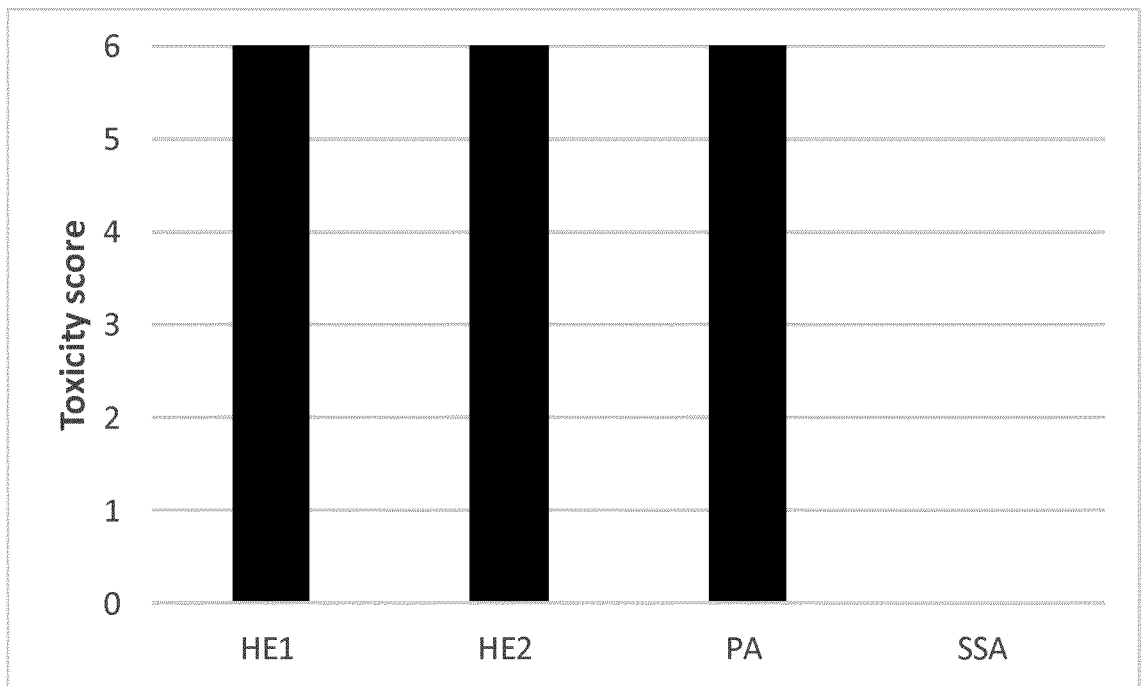
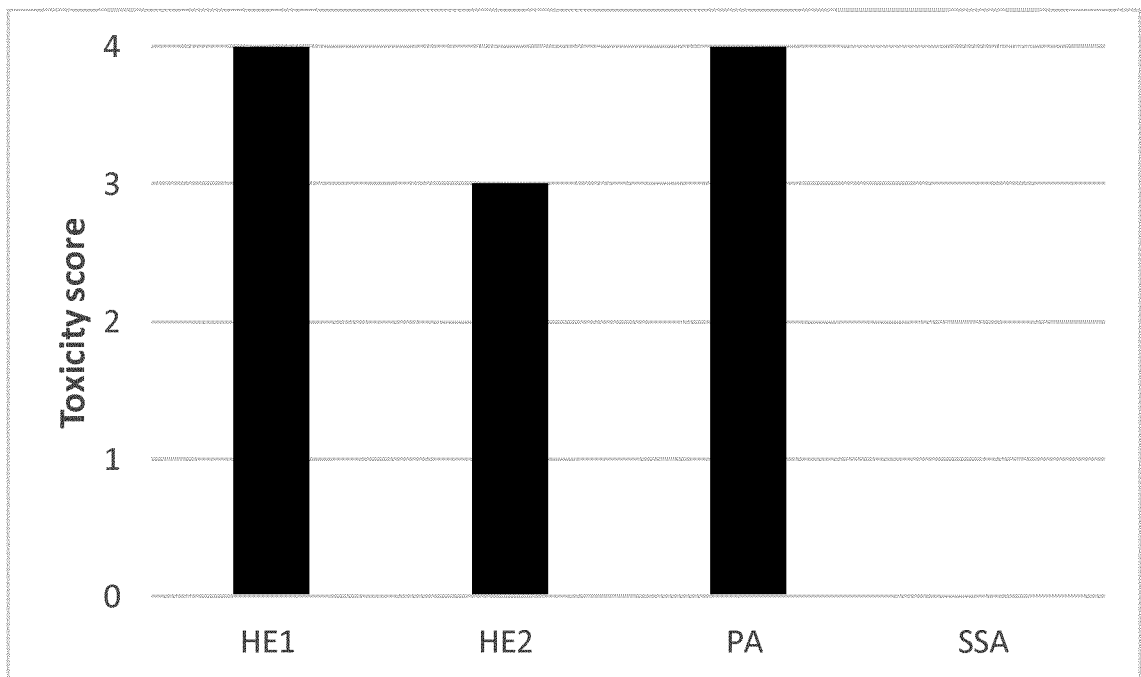


FIG. 2

**FIG. 3****FIG. 4**

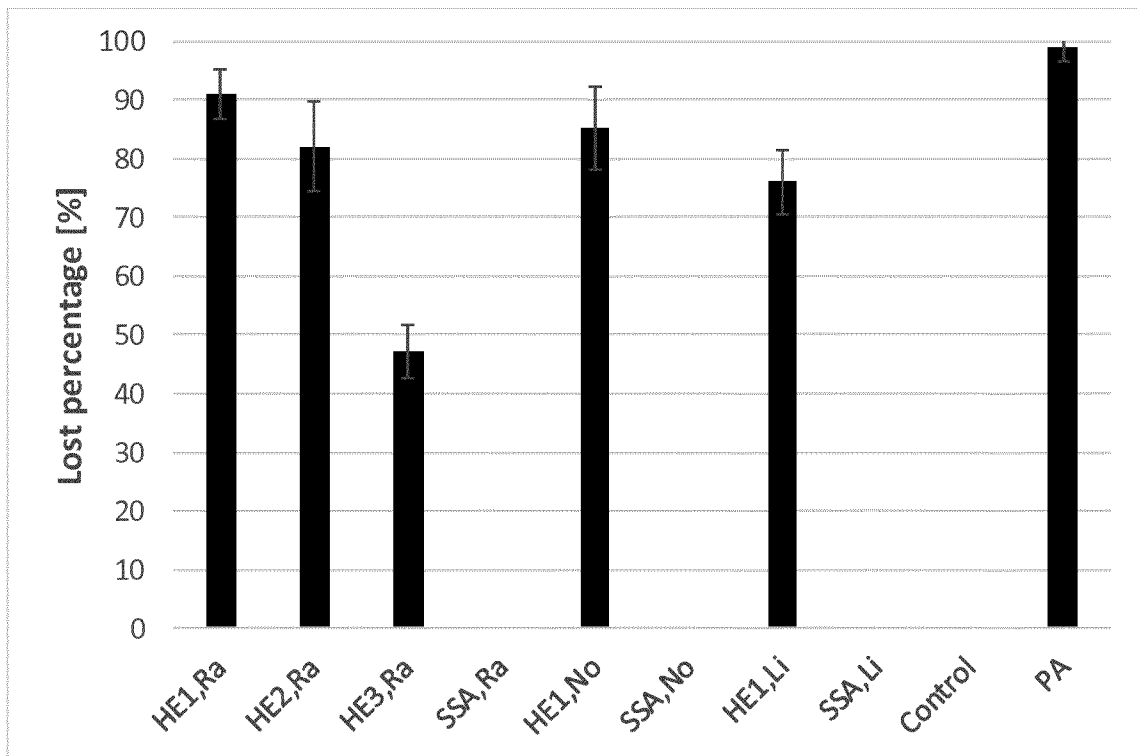


FIG. 5

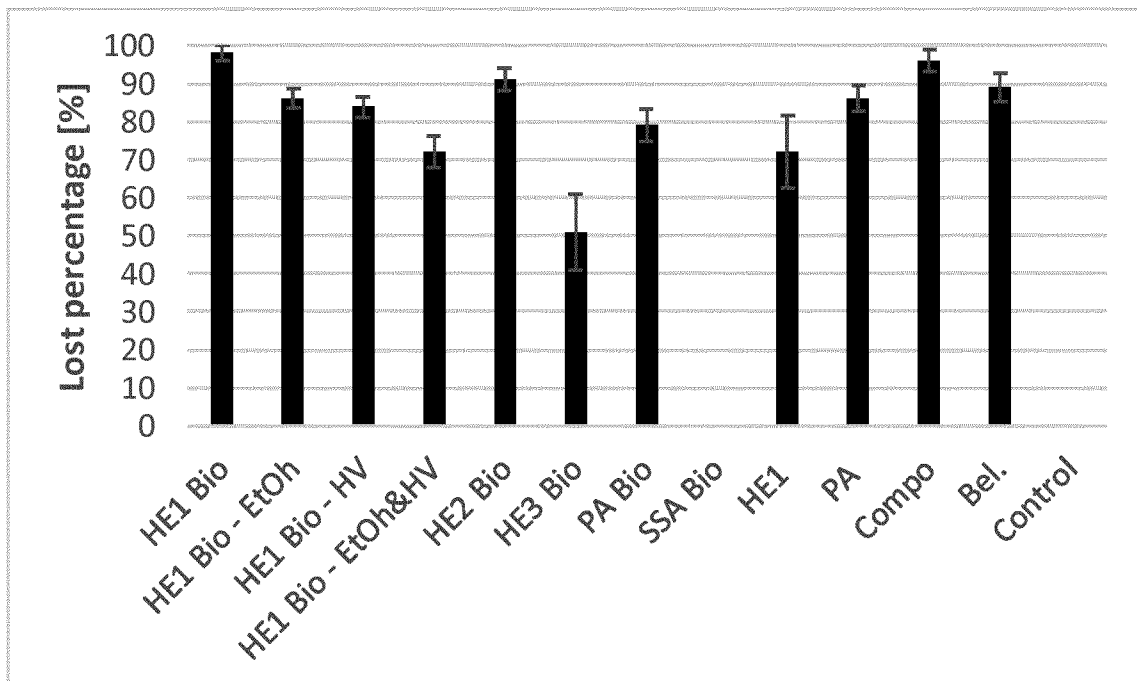


FIG. 6

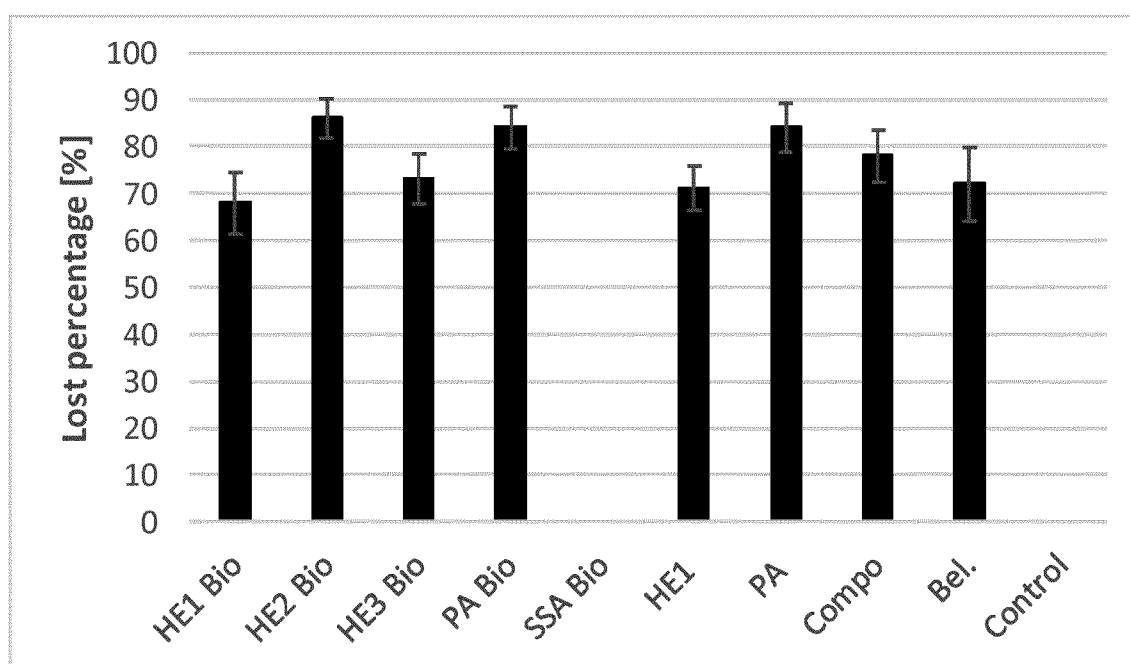


FIG. 7