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A review of insecticidal effect of essential oils on stored grain pests

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Abstract

Grain production grows more every year, and stored grain pests have been a challenge for agriculture. The use of pesticides is the main solution to face this challenge; however, improper and extensive management leads to another difficulty: the resistance acquired by some pests, causing their ineffectiveness. Thus, essential oils and other natural products emerge as an alternative to overcome this situation. In this study, several essential oils with insecticidal/repellent properties were reviewed to evaluate the capacity to become an important method for protecting stored grains against the most recurrent pests. This is a literature review that analyzed articles in scientific database platforms, using different combinations of the keywords "Essential oils", "Insecticides", "Stored grains", "Repellents" and "Insects". Among the essential oils studied, the most outstanding were the essential oils of: *Artemisia argyi* and *Mentha haplocalyx* against *Lasioderma serricorne; Artemisia rupestri*s and *Ligusticum pteridophyllum* against *Liposcelis bostrychophila*; and *Artemisia anethoides, Elshotzia ciliata*, and *Amomum maximum* against *Tribolium castaneum*. Along with some essential oils, their main components and insecticidal/repellent capacity when isolated were also evaluated. The results were promising, although mechanisms of action have not yet been elucidated. Investing in research for alternative pest control can contribute to sustainable agriculture.

Keywords: essential oils; secondary metabolites; inseticide; insects; stored grains.

Practical Application: Essential oils with insecticidal potential against the main pests of stored grains.

1 Introduction

Agricultural grain production is an important pillar for the socioeconomic growth of many countries. The four largest producers are the United States, China, India, and Brazil, which account for 54% of all grains (Food and Agriculture Organization of the United Nations, 2020, 2022). In 2020, the total volume of grains produced by the world was more than 3 billion tons. The three main commodities are corn, representing 1.2 billion tons; rice, with 512 million; and soybean, with 371 million (Food and Agriculture Organization of the United Nations, 2020).

Currently, the agricultural sector has been suffering from the coronavirus (COVID-19) pandemic, which has led to an increase in commodity prices, input costs, and transportation costs (Food and Agriculture Organization of the United Nations, 2022), and, with the Ukrainian war, will also generate a significant reduction in wheat production (U.S. Department of Agriculture, 2022).

This scenario suggests that world agriculture needs to constantly invest in new technologies to optimize production and prolong the quality of stored products. Proper storage helps in monitoring the quality of seeds and grains and allows them to be marketed in better periods, avoiding market pressures (Magalhães et al., 2014). At this stage, the grains are prone to insect and fungal attacks, and the lack of investment in storage structure favors the action of these pests, leading to significant losses. Approximately one third of the world's production is destroyed by almost 20,000 species of pests in fields and stored grains (Ahmad et al., 2021).

Facing this problem is a daily challenge for grain producers, and proper management will ensure the profitability of their production. In addition to following proper storage practices, the application of pesticides has a favorable cost-benefit ratio. Nevertheless, indiscriminate use has been characterized by several negative impacts, such as resistance, toxicity, adulteration, erratic supplies and unavailability in critical periods (Fields & White, 2002).

An alternative solution to reduce pesticides, without losses in grain protection, would be the use of essential oils (EOs). EOs are substances derived from the secondary metabolism of plants. In some cases, they come from the skins of the fruits, which are generally waste to be discarded and can generate extra income with their extraction (Lü, 2017). These substances have volatile and non-fatty characteristics and are classified according to their biochemical activity and by their molecular structure (Lü, 2017).

EOs have a great potential in agriculture by methods of fumigation and contact with pests, causing repellency, mortality, inhibition of oviposition, decreased larval development, among other beneficial effects for the protection of stored grains (Ayvaz et al., 2010). However, their share in the world market is still very limited, as their use is restricted due to volatility,

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photosensitivity, and rapid oxidation. Therefore, their handling needs to be done carefully and under appropriate conditions (Rajkumar et al., 2020a).

In line with the context presented, this study sought to evaluate the effectiveness of EOs in combating stored grain pests and analyze their benefits compared to commonly used agrochemicals.

2 Methodology

To elaborate this study, a literature review was carried out, which had as a guiding question: are EOs really more or as effective as current chemical agents in controlling common pests in grain storage?

The exploratory study with a qualitative approach was based on a literature review with information collected by searches on the following scientific database platforms: PUBMED (National Center for Biotechnology Information – NCBI, U.S. National Library of Medicine), and SCiELO (Scientific Electronic Library Online), using different combinations of the keywords "Essential oils," "Insecticides," "Stored grains," "Repellents," and "Insects."

The inclusion criteria adopted were articles that match the predicted keywords and insects as pests in the stored grains. The exclusion criteria were publications prior to the year 2000. After selection, the articles were analyzed and organized in a spreadsheet in which the insect species, essential oils, and their effectiveness in pest control were highlighted.

3 Results and discussion

For the elaboration of the study, 196 articles on EOs and their insecticidal properties were found in all databases analyzed. 153 studies were excluded because they did not meet the inclusion and exclusion criteria, and 42 articles were analyzed. From these studies, numerous EOs were evaluated for their insecticidal potential against *Acanthoscelides obtectus, Callosobruchus maculatus, Callosobruchus chinensis, Lasioderma serricorne, Liposcelis bostrychophila, and Tribolium castaneum,* and they will be discussed below.

3.1 Essential oils in the control of Acanthoscelides obtectus

Acanthoscelides obtectus (*Coleoptera: Chrysomelidae: Bruchinae*) is a pest that consumes the common bean grain (*Phaseolus vulgaris L*). It attacks seeds in the field and in storage where, in unsuitable structures and with little care, it can cause a loss of up to 20 to 40%. Oviposition and growth are continuous, causing great losses in a few months (Fogang et al., 2012).

The control is done with insecticides such as pyrethroids, phosphine, and organophosphates, but these products are toxic to the health of workers, consumers, and the environment (Fogang et al., 2012; Rodríguez-González et al., 2019). The development of resistance has led to the search for natural alternatives (Fogang et al., 2012; Rodríguez-González et al., 2019).

The key to a good strategy is to find compounds that cause less damage to the environment and have different modes of action. Regarding the effects against *A. obtectus*, the EOs of basil (*Ocimum basilicum)* and citronella (*Cymbopogon winterianus)* show good results when applied directly to the insect in petri dishes. For *O. basilicum,* an application of 120 μL after 15 days showed a mortality rate of 74.94 ± 3.19% for adult insects. At a dose of 60 μL, a result of 70.08 ± 5.17% was obtained. For citronella, when using 120 μL of the oil, 70.25 ± 4.58% mortality was obtained after 15 days (Rodríguez-González et al., 2019).

O. basilicum is mainly composed of linalool and estragole, its toxicity can be explained by the action of the mixed activity of the enzyme acetylcholinesterase (AChE), especially when they are applied by fumigation. Namely, a study on corn grains showed that the increase in temperature (35 °C) and exposure to light during processing affected the stability of this EO (Moura et al., 2021).

The EOs of oregano *(Origanum onites L.),* Cretan savory *(Satureja thymbra L), and* common myrtle *(Myrtus communis L)* also presented increased mortality of adult insects with increasing concentration (Ayvaz et al., 2010). The EO of common myrtle was the most efficient in the repellency tests. Concerning the mortality of adult pests, 100% mortality was observed using a concentration of 195 μL/l of air after 144 hours for the EOs of oregano *(O. onites L.)* and Cretan savory *(S. thymbra L).* On the other hand, the EO of common myrtle *(M. communis L)* presented the same effect only 72 hours after exposure and using 65 μL/l of air. This higher toxicity of the common myrtle *(M. communis L)* can be attributed to its major constituent, linalool (31.3%), which is a strong acetylcholinesterase inhibitor (AChe) (Ayvaz et al., 2010; Wang et al., 2019).

The EO of oregano is an important aromatic plant rich in terpenoid components, researchers have demonstrated high contact toxicity in larvae of another storage product pest, *Tenebrio molitor.* Interestingly, in addition to the toxic effect, the study demonstrated that exposure to OEO affects the behavioral response and causes repellency in larvae and adults, suggesting a potent alternative to synthetic insecticides (Plata-Rueda et al., 2021).

In the study that evaluated the EO of thyme *(Thymus vulgaris)*, the researchers observed that females are more resistant, relating this to a greater resistance to the oxidative stress caused. It is believed that thymol, its main component, is responsible for this toxicity, since it is an acetylcholinesterase inhibitor, thus increasing oxidative damage to macromolecules and increasing mortality (Lazarević et al., 2020).

3.2 Essential oils in the control of Callosobruchus sp.

Callosobruchus maculatus and *Callosobruchus chinensis* are Coleoptera species of the subfamily *Bruchinae*. They present a qualitative and quantitative risk in the production and storage of beans *Vigna unguiculata Walp* (Balachandra et al., 2012), popularly known as black-eyed peas or string beans, and also a major threat to legume warehouses in general, even causing the loss of 32-64% of production in India (Chaubey, 2008). These grains have a significant importance for countries on the African, Asian, and European continents, mainly because they are an alternative source of proteins and essential nutrients (Balachandra et al., 2012).

The EO of *Citrus sinensis* and its components were tested addressing their inhibitory actions on AChE, Na+/K+ ATPase, and glutathione S-transferase (GST), with contact assays and dose-dependent fumigation, using as control a phosphine-based agrochemical in adult subjects (Oyedeji et al., 2020). In addition to the EO, nine of its most representative components, limonene, geraniol, L-carvone, linalool, citral, 3-Carene, terpineol, citronellol, and β-caryophyllene, were tested separately. The best results were the reduction of Na+/K+ ATPase activity, obtaining the values of 43.3% (CL_{25}), 70.2% (CL_{50}), and 85.3% (CL_{95}), and that of AChE inhibition, reaching the mark of 70.3% for linalool, 66.8% for limonene, and an inhibition of more than 75% for the EO of *C. sinensis*(Oyedeji et al., 2020). For the inhibition of GST, the results were not so satisfactory (Oyedeji et al., 2020).

According to Ya-Ali et al. (2020), one of the EOs with the highest pest management potential is the one of *Eucalyptus globulus Labill*, also known as Tasmanian eucalyptus and Tasmanian blue gum (TBG), for possessing a broad spectrum of action. In its assay, the toxicities by contact and fumigation with nanoformulations and bulk formulations were tested, as well as its ovicidal and repellent properties. Both the toxicity of the oil on insects and its greater stability in the nanoformulation have been proven (Ya-Ali et al., 2020).

The EO of *Artemisia annua* proved to be quite promising to deal with all stages of growth of the *C. maculatus*, including the elimination of eggs. However, further studies are needed to clarify whether it has non-target toxicity, in order to prevent poisoning of consumers and farmers (Tripathi et al., 2000).

The EO of *Coriandrum sativum* was efficient in repellency, achieving 100% mortality in 24 hours of exposure to oil vapor. The CL_{50} was 1.34 µL/L of air for this condition. The toxicity of this oil has been associated with linalool, an element that represents the largest percentage of its composition (Khani & Rahdari, 2012). The insects also showed symptoms such as hyperactivity, tremors, and convulsions, indicating neurotoxicity, due to the inhibitory action of the oil on the AChE enzyme in the central nervous system (CNS). This effect is similar to those caused by the pyrethroid class of agrochemicals (Khani & Rahdari, 2012).

3.3 Essential oils effective in controlling Lasioderma serricorne

Widely distributed in the world, mainly in subtropical and tropical regions (Wu et al., 2015), *Lasioderma serricorne* is one of the main pests of stored tobacco and grains (soy, cocoa), causing great losses that go beyond the consumption of the grains, but also an increase in temperature leading to the appearance of molds (Zhang et al., 2015b). As a control method, one still recommend the use of synthetic insecticides such as phosphine, but, insects have been showing resistance to this agrochemical and causing a risk of environmental pollution and resurgence of pests (Zhang et al., 2014, 2015a).

The composition of the EO of *Menta haplocalyx* varies greatly according to the region and also the conditions of this area, which can result in different biological effects (Zhang et al., 2015a). For contact toxicity tests, the three major components of the EO of *M. haplocalyx* (methyl acetate, menthol, and limonene) showed strong toxicity against adult insects (DL_{50} values for the components of 5.96, 7.91, and 13.7, respectively, *versus* 16.5 µg/adult of EO). Methyl acetate was the one that exhibited the highest contact toxicity; however, its result was 25x lower than that obtained by pyrethrins used as a positive control (Zhang et al., 2015a).

Repellent activity tests were performed with the EO of *M. haplocalyx* and its main constituents, and, as a result, menthol was the one that presented the highest repellency (2 to 4 hours after treatment at the lowest concentration tested 0.31 and 0.06 nl/cm^2), being even greater than the positive control DEET (N, N-Diethyl-3-methylbenzamide). The results obtained by menthol after 4 hours of exposure presented better levels of repellency, which indicates that menthol also has a good persistence; on the other hand, methyl acetate and limonene exhibited a lower repellent activity compared to the positive control (Zhang et al., 2015a).

The EO of *Alpinia kwangsiensis* showed contact toxicity against adults of *L. serricorne* however, when compared to the positive control phosphine and pyrethrins, the EO was much less toxic. The fumigant activity of the EO was 13x lower than that of the agrochemical. When compared to other EOs in the literature, *A. kwangsiensis* has been shown to be a strong option for both contact toxicity and fumigation. When testing its main components (camphor and eucalyptol), they showed better fumigant and contact effects than EO (DL_{50} for contact of 11.30 and 15.58 µg/adult and fumigation CL_{50} of 2.91 and 5.18 mg/L of air, respectively). The effects of the EO of *A. kwangsiensis* can be attributed to the synergistic effects of its components, and, in the case of fumigation, to their volatility (Wu et al., 2015).

When researching EOs of plants of different species of *Valerianaceae,* Feng et al. (2019) obtained satisfactory results regarding their toxicity by contact. The EO of *Valeriana khatamansi* was the one that obtained the highest toxicity, with a DL_{50} value of 17.6 µg/adult; moderate toxicity was observed in the EO of *Valeriana officinalis and V. officinalis latifolia,* with DL_{50} values of 23.2 and 24.6 µg/adult; in turn, *Nardostachys chinensis Bat. (NC)* was the least toxic among them, with a DL_{50} of 57.8 µg/adult (Feng et al., 2019).

The contact toxicity presented by the EO of Chinese sagebrush (*Arhemisia argyi)* was strong in relation to adults of *L. serricorne*, stronger than its four main isolated components (eucalyptol, β-pinene, β-caryophyllene, and camphor). The EO presented a value of DL_{50} of 6.42 µg/adult, while its isolated major components presented DL_{50} of 15.58; 65.55; 35.52; and 11.30 µg/adult, respectively. Compared to the positive control pyrethrins, the EO presented 10x less toxicity to the insect than this agrochemical (Zhang et al., 2014). Camphor presented excellent values in terms of fumigation toxicity against adult insects; its CL_{50} value was 2.91 mg/L of air, this value being better than the EO of *A. argyi* (CL_{50}= 8.04 mg/L of air). However, compared to the positive control phosphine (CL_{50}= 0.0923 mg/L air), camphor had a much lower fumigation toxicity than the agrochemical (Zhang et al., 2014).

The EO of *A. argyi* showed strong repellency against adult insects; in the concentration of 39.32 nL/cm^2, it can be considered a Class V repellent (which has repellency of 80.1-100%) at 2h and 4h after exposure (Zhang et al., 2014). At the lowest concentration tested 0.06 nL/cm^2, eucalyptol showed a very high repellency after 4 hours of exposure, being higher even than the DEET positive control. Compared to the positive control, the EO and eucalyptol showed strong repellency at concentrations of 1.57, 0.31, and 0.06 nL/cm^2 after 2 hours of exposure. Eucalyptol, EO, and β-pinene showed the same and higher repellency than the DEET control in the concentrations of 0.31 and 0.06 nL/cm^2 after 2 hours of exposure; the components β-caryophyllene and camphor did not show many repellent effects (Zhang et al., 2014).

Tests conducted by Pang et al. (2020) with the EO of *Peppermint (Mentha piperita)* and its two major components (menthol and L-menthone) brought good results. In the fumigation toxicity tests, the EO obtained a value of CL_{50} of 68.4 mg/L of air, while L-menthone achieved a result of CL_{50} of 14.8 mg/L of air; on the other hand, menthol did not produce good results(Pang et al., 2020). At the highest concentration tested, the contact toxicity of *M. piperita* and its two major components presented more than 90% mortality against target insects, including *L. serricorne;* the value of DL_{50} was 12.6 μg/adult for the EO. *L. serricorne* was also sensitive to the components tested: menthol presented DL_{50} value of 9.8 μg/adult and L-menthone, a DL_{50} value of 8.5 μg/adult (Pang et al., 2020).

The results of the repellency test of *M. piperita,* menthol, and L-menthone against adult insects were very satisfactory; at the maximum concentration tested (78.63 nL/cm^2), the EO and menthol showed the same level of repellency of the DEET positive control after 4h of exposure, both presenting as Class V repellents (80.1-100% repellency) (Pang et al., 2020).

When testing the repellent activity of the EO of *Ligusticum pteridophyllum* and its main component myristicin (90% of the EO), Qi et al. (2020) obtained good results. Compared to the DEET positive control, myristicin proved a strong repellent against adult insects at the concentration of 3.15 nL/cm^2 after 2 hours of exposure. Myristicin also showed repellent activity comparable with the EO of *L. pteridophyllum* in all concentrations after 4h of exposure; with the results obtained, it was also possible to observe that the repellent activity is dose-dependent (Qi et al., 2020). In the case of contact toxicity, when comparing the values obtained from the EO of *L. pteridophyllum* with those of the pyrethrin positive control, it is observed that the values obtained by the agrochemicals are more satisfactory. The values of DL_{50} for the EO in the treatment of adults of *L. serricorne* was 89.82 μg/adult (Qi et al., 2020).

The repellency data of the EO of *L. jeholense* have not been satisfactory when it comes to *L. serricorne*. The N-butylbenzene component did not exhibit any repellent effect; 3-butylidenephthalide showed weak activity; in addition, spathulenol and myristicin showed good repellency against this insect at concentrations of 78.63 and 15.73 nL/cm^2 after 2-4h of exposure (Luo et al., 2019).

The toxicity by contact and fumigation of the EO of *Artemisia anethoides* and its two main components (1,8-cineole and Terpinen-4-ol) was analyzed and, when compared to the positive control bromomethane, it was not satisfactory (Liang et al., 2017). In the repellency tests, the EO of *A. anethoides* was able to show the same level of repellency as the positive DEET control at concentrations of 78.63 and 15.73 nL/cm^2 after 2 hours of exposure against adults of *L. serricorne*; in other concentrations tested the result obtained was lower (Liang et al., 2017).

For the research of the EO of winged prickly ash *(Zanthoxylum planispinum)*, it was extracted from both leaves (EL) and fruit pericarp (EFP), and its main components identified were linalool (EF 71.33%; EPF 73.74%), silvestrene, and Terpinen-4-ol. The biggest difference observed was that EL also had the presence of a large amount of 2-dodecanone (11.52%), a component not present in EFP (Wang et al., 2019). Table 1 shows that the insects of *L. serricorne* were more susceptible to EFP in the case of fumigation; by contact, both EL and EFP had strong toxicity (Wang et al., 2019).

Lu et al. (2021) have obtained good results with EO of arbor (*Clerodendrum bungei*) and its main components (myristicin and linalool). At a concentration of 78.63 nL/cm^2, the EO and myristicin obtained good repellency values, but when the concentration decreased, it was found that the repellent activity decreased significantly; linalool showed repellent activity only at the concentration of 15.73 nL/cm^2 after 2 hours of exposure (Lu et al., 2021).

The EOs of *Ajania nitida* and *Achania nematoloba,* although being of the same genus, have main components that differ in some parts and, for this reason, their effects may be different. In fumigation tests, *A. nitida* did well, with CL_{50} of 11.23 mg/L air, whereas *A. nematoloba* presented no fumigant effect. Despite the good result, *A. nitida* was not as effective as the phosphine positive control (Li et al., 2018).

3.4 Essential oils in the control of Liposcelis bostrychophila

Lipocelis bostrychophila was a neglected pest that, due to its small size (~1 mm) and its resistance to the usual agrochemicals, began to gain ground in research (Liu et al., 2012b). In the articles analyzed, the EOs of *Illicium henryi Diels, Clinopodium chinense (Benth.) Kuntze, Kaempferia galanga*, *Artemisia rupestris L.,* and *Curcuma wenyujin* were tested in contact and fumigation tests, as well as their main isolated components. The EO of *I. Henry* obtained the CL_{50} of 96.83 μg/cm^2 and 380.39 μL/L of air, while its compounds, myristicin and safrole, showed results of CL_{50} of 18.74 μg/cm^2 and 69.28 μg/cm^2 and 121.95 μL/L of air

Table 1. Toxicity by contact and fumigation of the EO of *Z.planispinum*and its main constituents against adults of *Lasioderma serricorne*.

Treatment	Fumigation CL_{50} (mg/L air)	Contact DL_{50} (μg/adult)
EL	32.32	10.38
EFP	13.01	14.97
Linalool	46.93	27.41
Terpinen-4-ol	1.3	5.42
2-dodecanone	7.48	2.54
Pyrethrins	-	0.24
Phosphine	0.0923	-

Source: adapted from Wang et al. (2019).

and 322.54 µL/L of air for contact and fumigation, respectively (Liu & Liu, 2015).

For the EO of *Clinopodium chinense (Benth.) Kuntze* and its three compounds, bornyl acetate, caryophyllene, and piperitone, the results of CL_{50} were, in the contact test: 215.25, 321.45, 275.00, 139.74 µg/cm^2, and, in the fumigation tests: 423.39, 351.69, >10000, 311.12 µL/L of air, respectively (Li et al., 2015).

The EO of *K. galanga* presented four compounds with more significant results, namely 1,8-cineole, ethyl cinnamate, ethyl P-methoxycinnamate, and the trans isomer of cinnamaldehyde. The results of CL_{50} were, in the contact test: 68.58, 1049.41, 21.41, 44.61, 43.40 µg/cm^2, respectively. In the fumigation test, the crude EO and its components presented a CL_{50} of 1.47, 1.12, 10.21, 10.23, 1.29. Moreover, a repellency test was performed for this oil and its most relevant components, showing that cinnamaldehyde has the best result, which is a moderate repellency action of 77% after 4 hours of exposure (Liu et al., 2014).

For the oil of the aerial parts of *A. rupestris* L. and its four components, linalool, α-terpineol, α-terpinyl acetate, and 4-terpineol, the results of CL_{50} for the contact test were: 418.48, 393.16, 140.30, 92.59, 211.35 µg/cm^2; for the fumigation test: 6.67, 1.96, 1.12, 1.26, 0.34 µL/L of air, respectively. To them was also assigned the repellency test, in which α-terpinyl acetate and α-terpineol showed a strong repellency against *L. bostrychophila*, with the values of 91% and 85%, respectively, at the concentration of 13 nL/cm^2 after 2 hours of exposure; the values were close to those of the positive control, DEET (100%) (Liu et al., 2013).

It is suggested that the toxicity and repellency effects caused by EOs are due to their major components, since, when isolated, they maintained or even exceeded the effects of crude EOs. All toxicity values presented, both in contact tests and in fumigation tests, were lower than those of the positive controls. Nevertheless, the effectiveness of these EOs and their components cannot be disregarded, since, in addition to protecting stored grains, there are no studies that prove their non-target toxicities and contamination of grains with their residues (Li et al., 2015; Liu et al., 2012b, 2013, 2014; Liu & Liu, 2015).

3.5 Essential oils effective in controlling Tribolium castaneum

The red beetle or red flour beetle *(T. castaneum)* is a worldwide pest and one of the main insects of grain, flour, and cereal storage (Liang et al., 2020). For its control, synthetic insecticides such as organophosphates and carbamates are still used, which can cause multidrug resistance and damage to the environment and human health (Hashem et al., 2018; Mustapha et al., 2020).

One of the main studied EOs is *Artemisia anethoides* and its main constituents (1,8-cineole and Terpinen-4-ol). In the study by Liang et al. (2017), this oil and its components had good toxicity by fumigation and contact against adults of *T. castaneum*, but lower than the values obtained against *L. serricorne.* However, in the repellency tests, *A. anethoides* was more evident against *T. castaneum* than against *L. serricorne*. A percentage repellency of 100% was found at the tested concentrations of 78.63 and 15.73 nL/cm^2 after 2/4h of exposure, with a value similar to that of the DEET positive control (Liang et al., 2017).

For the EOs of *Artemisia absinthium, A. campestris L, and A. herba-alba,* fumigation tests resulted in increased toxicity by the EO of *A. herba-alba* after 24h of exposure. At a concentration of 200 µL/L, all EOs of *Artemisia sp* tested showed their greatest toxic effect (Chaieb et al., 2018). To try to increase the effectiveness of these oils, a combination of them was made to evaluate the interaction and a possible increase in the effect. An antagonistic effect was observed when mixing the oils, and only 60% mortality was obtained at the highest concentration used (200 µL/L) (Chaieb et al., 2018).

According to the repellency tests, *A. absinthium* was the one that repelled most quickly and effectively at all times of exposure. *A. herba-alba* showed better results only during 1h, and its effect was decaying over time. *A. campestris* showed better effect after 2h exposure (Chaieb et al., 2018).

When testing the EO of species of *Valerianaceae spp such as: V. officinalis L. (VO), V. officinalis L. var. latifolia Miq.(VOL), V. jatamansi Jones (VJ), and N. chinensis Bat. (NC)* against *T. castaneum,* it was observed that, the EO of *VO* was the one that obtained the best result regarding its contact toxicity (DL_{50}= 10.0 µg/adult), followed by *VOL* and *NC* (DL_{50}= 19.5 and 147.1 µg/adult, respectively). The EO of *VJ* was the least effective against *T. castaneum*; however, it is more effective against *L. serricorne* (Feng et al., 2020).

When analyzing the synergy obtained by mixing the main compounds (camphene and bornyl acetate) of these EOs, Feng et al. (2020) obtained results ranging from synergism, additive effects to antagonism, as can be seen in Table 2.

Repellency effects at concentrations of 78.63 and 15.83 nL/cm^2 have been observed, with *V. officinalis, V. jatamansi,* and *N. chinensis* standing out with effects comparable to or even greater than the DEET positive control. *V. khatamansi* was superior, because its repellency and long-lasting capacity surpassed the agrochemical (Feng et al., 2020). Tests revealed that the EO of *V. officinalis* has no fumigant activity against *T. castaneum*; nevertheless, its main components camphene and bornyl acetate have a certain fumigation toxicity (Feng et al., 2019).

Fumigation results for *M. piperita* and its main components (menthone and menthol) were satisfactory. At a concentration of

Table 2. Synergism of the two main components of the EO of *Valerianaceae* spp (camphene and bornyl acetate) in mixed groups against *Tribolium castaneum.*

Groups	% Mixture	DL_{50} observed (µg/adult)	
	Camphene	Bornyl acetate	
1	50	50	82.9
2	22.2	77.8	46.6[a]
3	18.2	81.8	30.3[b]
4	77.8	22.2	46.5
5	81.8	18.2	55.4

Source: adapted from Feng et al. (2020); [a] – Additive effect, same value of VO; [b] – synergistic effect.

100 µL/L, the EO and its components exhibited 100% mortality in 24 hours of exposure. Values of CL_{50} and CL_{90} were respectively 48.68, 78.87 µL/L for the EO; 51.95, 84.86 µL/L for menthone; and 54.49, 89.95 for menthol (Rajkumar et al., 2019). When testing the effects of fumigation with the EO of *M. piperita* encapsulated, the results were significantly better (Rajkumar et al., 2020b).

The bioactivity (fumigation toxicity) of a nanoemulsion of *Pimpinella anisum* was tested with great results against adults of *T. castaneum* and their progeny: the reduction in the production of these offspring was 70% and the value of CL_{50} obtained after 72h of exposure was 9.84%. Considering that the crude EO of *P. anisum* was used as positive control, the nanoemulsion obtained a better result (Hashem et al., 2018).

When testing the fumigant effects of the EO of *Piper nigrum*, pure and in nanoparticles, Rajkumar et al. (2020a) obtained values of CL_{50} and CL_{90} that were most effective for the EO in nanoparticles. Comparing values of CL_{50} and CL_{90} for *T. castaneum*, it is observed that the pure EO showed values of 55.77, 97.93 µL/L of air, while the nanoparticle EO showed values of 29.02, 59.13 µL/L of air. This more effective result for nanoparticles may be an indication that nanoformulations are an alternative for the development of potential insecticides (Rajkumar et al., 2020a).

For the EO of *Curcuma amada,* was obtained different results according to the extraction method used. For the fumigation tests, the best result was with ultrasound-assisted extraction. Regarding repellency and contact, the best results were obtained with microwave-assisted extraction and ultrasound-assisted extraction (Narayanankutty et al., 2021).

The EO of *Illicium pachyphyllum* and some of its components presented contact toxicity; however, when compared to the positive control, its effectiveness was 80x lower. The same goes for fumigation, where the EO was 22x less toxic than the bromomethane-based agrochemical (Liu et al., 2012a). Nevertheless, its trans-ρ-Mentha-1(7), 8-dien-2-ol component showed great potential to be developed as a possible natural fumigant/insecticide.

The repellency obtained by testing the EO of *Crithmum maritimum* was very satisfactory. A value of 93% repellency was obtained after 2h of treatment at a concentration of 0.04 µL/cm². When testing for contact toxicity, insect mortality increased according to the increase in EO concentration, and the value of DL_{50} obtained was 9%. For these two effects, it is believed that the high content of the Dillapiole component is responsible, due to its insecticidal effect (Mustapha et al., 2020).

The EO of *Elsholtzia ciliata* and its main components (limonene and carvone) have both contact and fumigation toxicity against *T. castaneum*. *E. cilliata* presented better results by contact against adult insects than larvae. Among its components, carvone was the one that obtained the highest toxicity by contact. In the case of fumigation, the effect was better observed in the larvae than in the adult insect (Liang et al., 2020). According to the authors, the results suggest that *E. ciliata* and its two monomers have a potential application value as a natural insecticide to eliminate *T. castaneum* (Table 3).

Table 3. Toxicity by fumigation and contact of the EO of *Elsholtzia ciliata* and its main components against adults and larvae of *Tribolium castaneum.*

	Treatment	DL_{50} (mg/adult)	CL_{50} (mg/L of air)
Adult	EO *of E. cilliata*	7.79	11.61
	Carvone	5.08	4.34
	Limonene	38.57	5.52
	Pyrethrin	0.09	–
	Bromomethane	–	1.83
Larva	EO *of E. cilliata*	24.87	8.73
	Carvone	33.03	28.71
	Limonene	49.68	20.64
	Pyrethrin	1.31	–
	Phoxim	–	1.05

Source: adapted from Liang et al. (2020).

The EO of *Juniperus formosana* and its main components (4-Terpineol, α-pinene, and D-limonene) presented contact toxicity. For the EO, the value of DL_{50} was 29.14 µg/adult, while its component 4-Terpineol showed better results, with a DL_{50} of 7.65 µg/adult, and more effective than the other components α-pinene and D-limonene. Compared to *L. bostrychophila,* the EO of *J. formosana* was more efficient for *T. castaneum* (Guo et al., 2016).

In the repellency tests, the EO of *J. formosana* and its compounds have shown good results, where the percentage of repellency exceeded 80% after 2h of exposure. Compared to the positive DEET control, the EO and its main components presented repellency equivalent to that of the agrochemical after 2h of exposure; nevertheless, at a concentration of 0.13 nL/cm², it showed an attractive property of insects (Guo et al., 2016).

Among the comparisons mentioned, *T. castaneum* was the insect most sensitive to the effect of EOs. In the case of repellency, the values obtained against other insects showed significant results, even compared to the agrochemical DEET, where, for *T. castaneum,* the values were often considered superior to this compound (Feng et al., 2020; Guo et al., 2015, 2016; Liang et al., 2017; Lu et al., 2021).

3.6 Commercialization and industrial uptake

Few practical studies with applicable results, legislation with many details for the use of EO as pesticides and maintenance difficulties after using the technology (Pavela & Benelli, 2016).

In order to grant the commercialization of EO, it is essential to simplify the authorization processes for the use of new botanical pesticides based on EO, together with new ways to avoid loss of efficiency when using stabilization processes such as encapsulation. Therefore, further research bringing together scientific centers and commercial manufacturers could help to increase practical results, facilitating the approach towards the development of an environmentally friendly, more efficient and stabilized pesticide that can be applied on an industrial scale (Pavela & Benelli, 2016).

Currently, only a few plant EO-based insecticides have had commercial success. Thus, there is room in the market for new products, but few go ahead due to the problems discussed earlier. In addition, these formulations are increasingly accepted, with sensitivity and safety for users, against flies, bedbugs and other insects mainly present in the hospitality industry and hospitals. In agriculture, even with lesser approval, there is a good commercialization of products for protected cultivation for greenhouses and row crops, with regard to the safety of field workers and the demand for residue-free pesticides. Opening opportunities for the application of these pesticides in mixing or rotation tanks in conventional large-scale food production systems (Isman & Tak, 2017).

4 Conclusions

EOs and their components have shown promising results against the storage pests that constantly threaten grain marketing around the world. *T. castaneum* was the most sensitive insect. For *L. serricorne,* the most effective EOs were *A. argyi*, by contact, its components camphor and eucalyptol, by fumigation and repellency, respectively. *L. bostrychophila* proved to be quite resistant; better results were found for *A. rupestris* by fumigation and *L. pteridophyllum* by repellency and contact.

Botanical active formulation of insecticides based on plant EO is a highly desirable property for commercialization because of better biodegradation into environmental conditions, elevated compliance with biocontrol agents and other natural forms of pesticides, along with bees and further pollinators. In this manner, nanotechnologies demonstrate better prospects for large-scale application of EO. Other concerns for commercial production are the maintenance of and availability of plant biomass for EO extraction. Sustainable production of EO for representative insecticide production may need a massive scale of widespread cultivation, which might be the reason that many potentially useful plants fail to exceed for extraction and, consequently, the commercialization process. At least, the expansion of EO commercialization considering industrial uptake is also related to the scientific investigation of non-explored plant sources associated with extended limits and interpretation of jurisdiction and regulatory regimes for EO utilization for producers of essential oil commodity markets.

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