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Review Article
Green Pesticides: Essential Oils as Biopesticides in Insect-pest Management

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Abstract
The long-term applications of synthetic insecticides have resulted in residues accumulating in different environmental components. They have adverse effects on non-target organisms, ecosystems and human health. Therefore, bio-insecticides "Green pesticides" have posted as an alternative to synthetic insecticides in agriculture and public health sectors. The study presented here focuses on the prospects of essential oils (EOs) as bio-insecticides for insect pest management. In fact, many EOs have insecticidal, fumigant, antifeedant, attractive and repellent activities against a broad spectrum of insects with some selectivity. The EOs are a complex of chemical compounds with multiple modes of action that enhances their activity due to the synergistic action between constituents. Due to their volatility in nature, EOs are used as a fumigant against agriculture and storage food insects. Consequently, EO-based insecticides are very important for control stored insects because they are active against a variety of insects, fast penetrating and no toxic residues in the treated products. In contrast, some problems (e.g., volatility, solubility and oxidation) of EO-based insecticides were recorded, which plays an important role in the EOs activity, application and persistent. For this reason, new formulations with nanotechnology "Nanoformulation" can resolve these problems and offer numerous advantages. So, encapsulating the EOs has a considerable perspective as commercial insecticide products. Finally, EOs-based insecticides are low toxic, environmental persistence and eco-friendly. Therefore, they are not having worker re-entry and harvest restrictions after treating crops. They are compatible with biological control programs and indigenous natural enemies of pests.

Key words: Green pesticide, pest, essential oils, pest management, bioinsecticide

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INTRODUCTION

Essential oils (EOs) are known as essences, volatile oils, etheric oils or aetheroleum. They founded in aromatic plants as a mixture of volatile components produced as secondary metabolites[1]. They are extracted from plant through steam or water distillation. Generally, EOs have densities lower than those of water[2,3]. They contain hundreds of individual compounds and their mixtures that give physical, chemical and biological characteristics of them[4]. Additionally, aromatic plants could be extracted by organic solvents to give oleoresins and by carbon dioxide to yield high-quality extracts free of solvents. Nevertheless, the solvent extraction is more hard and complicated processes than steam distillation. The EOs extracted by organic solvent produces volatile EOs and non-volatile flavor components that used in wide applications in food, agriculture and pharmaceutical industries[4].

Recently, there has been an increasing interest in studying and evaluating the botanical insecticides (e.g., EOs) for pest management in both developing and developed countries as a result of insect resistant to the traditional insecticides[5]. Also, the long-term applications of synthetic insecticides have resulted in residues accumulating in different environmental components (e.g., water, food, air and soil)[6]. They have adverse effects on non-target organisms, ecosystems[7] and human health[8]. Therefore, bio-insecticides (e.g., EOs) have posted as an alternative to synthetic insecticides in agriculture and public health sectors[9]. The use of nature-oriented as bio-pesticide (e.g., plant extracts, EOs, etc.) to control insect are called "Green pesticides". The term "Green pesticides" include all natural materials that can reduce the pest population and increase food production. Consequently, EOs are playing an essential role of pest control in organic food production globally[10,11]. According to Rozman *et al.*[12] the botanical insecticide presented by 1% of global insecticide market. They have lesser extent of environmental and human health impacts than most synthetic pesticides.

It has been reported that about 17500 aromatic plant species growing worldwide in tropical environments and more than 3000 constituents have been identified[13]. Three hundred of these EOs are commercially used for pharmaceutical drugs, cosmetics and perfume industries[3] beside pesticides[14,15]. However, the highest plant families bear EOs is Apiaceae, Asteraceae, Combretaceae, Geraniaceae, Gramineae and Lamiaceae. In addition to Myrtaceae, Meliaceae, Piperaceae, Rutaceae, Verbenaceae and Zingiberaceae plant families[16]. The ability of angiosperms and gymnosperms plants to accumulate EOs is quite excessive in both plants. However, the most sources of important commercially EOs are related to angiosperms (flowering plants)[17,18].

In the past few decades, various studies have investigated the insecticidal activity of EOs and their potential uses as bio-insecticides against important insect pests[19-28].

The study presented here focuses on the prospects of essential oils as bio-insecticides for insect pest management.

CHEMISTRY OF ESSENTIAL OILS

Essential oils are naturally found in plants as secondary metabolites. They play an important role in plant defense system against microorganisms, insects, herbivores and allelopathic interactions[29-31]. Previous studies have reported that volatile compounds of EOs can be classified into four groups: (a) Terpenes, (b) Benzene derivatives, (c) Hydrocarbons and (d) Other miscellaneous compounds[32]. Based on the number of isoprene units in the chemical structures of terpenes, EOs are classified into (a) Hemiterpenes (1 unit, C_5), (b) Monoterpenes (2 units, C_{10}), (c) Sesquiterpenes (3 units, C_{15}), (d) Diterpenes (4 units, C_{20}) and (e) Consequently. It has been found that most terpenes in EOs are monoterpenes ($C_{10}H_{16}$) and sesquiterpenes ($C_{15}H_{24}$). However, monoterpenoids are the most terpenes and represented by 90% of essential oils[33-35]. Monoterpenes have a chemical structure varies greatly along with various functions. Monoterpenes and their related chemical structure compounds have 10-carbone hydrocarbon. These chemical structures divided into (1) Acyclic alcohols such as linalool, geraniol and citronellol, (2) Cyclic alcohols such as menthol, isopulegol and terpeniol, (3) Bicyclic alcohols such as borneol and verbenol, (4) Phenolic compounds such as thymol, carvacrol, (5) Ketones such as carvone, menthone and thujone, (6) Aldehydes such as citronellal, citral, (7) Acids such as chrysanthemic acid and (8) Oxides such as cineole[34].

ESSENTIAL OIL EXTRACTION TECHNIQUES

Essential oils are found in flowers and other plant parts associated with other substances, e.g., gums and resins. However, different methods have reported for extracting and preparation essential oils from different parts of the plant. The methods or isolating techniques used for EOs extraction were playing an important role in differentiating EOs chemical compositions. These changes in the EOs chemical profile contain changing in the number of chemical compounds and the stereochemical types of molecules extracted[34]. In laboratory scale techniques, steam distillation is the common methods used for EOs extraction. In this method, Cleavenger apparatus was used for EOs extraction. However, under certain conditions of distillation process, many reactions may occur. These reactions include isomerization, saponification and other chemical reactions. These changes lead to alteration in the composition of the isolated oil[36]. In addition, other methods are used for isolation and extraction of EOs such as extraction by solvent and simultaneous distillation. Other methods are also used such as extraction by supercritical carbon dioxide and microwave ovens. Infact, many factors can affect the quality and the quantity of the product and their chemical composition, e.g., plant age, part, vegetative cycle stage, soil, fertilizer and climate[37,38].

Steam distillation method is the most common and economical one used for the production of EOs. Nearly, most EOs from different sources are extracted by distillation techniques. There are three types of distillation operation, namely: (1) Water distillation that recommended mostly for dried plant material. Simultaneously, the plant material is in direct contact with boiling water, (2) Water and steam distillation that's used for both fresh and dried plant materials (the plant material is at the bottom of the still and the plant material is in contact with saturated steam only), (3) Direct steam distillation that used for treat quantities of fresh plant material[39,40]. However, during steam distillation, certain components of the volatile oil tend to hydrolyze, whereas, other constituents of EOs that sensitive to heat and prolonged action of steam in the stillness can cause degradation and decomposition with the high temperature. Also, polymerization and resinification might also take place. Some components of essential oils dissolved in distilled water can't be easily recovered[41]. Therefore, other methods are used to solve these components such as (1) Expression method used for *Citrus* peel sp., oil extraction, e.g., peel of the orange, lemon and bergamot, (2) Solvents extraction method used for oil extraction decomposed by the action of steam or oils are found in small quantities in the plant, (3) Carbon dioxide method, this new method for extraction of EOs using liquid and liquefied supercritical carbon dioxide gas[42]. (4) Continuous subcritical water extraction method, in this method, the EOs has been extracted by either steam distillation or solvent and (5) Microwave extraction method.

TECHNIQUES APPLIED FOR ANALYSIS OF ESSENTIAL OILS

It is well known that the composition of EOs is represented by terpenes as major constituents in addition to other compounds. These constituents contain monoterpenes, sesquiterpenes and their oxygenated derivatives. In addition to aliphatic aldehydes, alcohols, esters and phenol ethers. The most structurally varied classes of plant natural products are terpenes that derived from the repetitive fusion of branched five-carbon units (isoprene unit)[43]. The analytical methods applied in the characterization of EOs mainly depend on the number of molecular species. In addition, the essential oil, chemical profile is closely related to the extraction procedure employed. Therefore, the choice of the appropriate extraction method becomes crucial. Most of the methods applied in the analysis of essential oils rely on chromatographic procedures. These methods can enable the separation and identification of components[44]. The most frequent methods applied to EOs analysis is Gas Liquid Chromatography (GLC). In this technique, detectors such as a Flame Ionization Detector (FID), Thermal Conductivity Detector (TCD) and others can be used for EOs analysis. Another technique for EOs analysis and identification is gas chromatography-mass spectrometry (GC-MS). This is the most common, simple and easy for identification by comparison of the required unknown mass spectra with those identified in reference MS library[45].

INSECTICIDAL PROPERTIES OF ESSENTIAL OILS

The chemical composition of EOs is lipophilic, which can enter into insect and cause biochemical dysfunction and mortality[46]. The toxicity of EOs does not only depend on the chemical compounds that act as toxins but also on many other factors playing an important role in the toxicity. The point of entry of the toxin, molecular weights and the mechanisms of action are factors of EOs to induce toxicity. It has been reported that common essential oils with insecticidal activities can be inhaled, ingested or skin absorbed by insects[47]. Previous studies have confirmed that terpenoids such as monoterpenoids have insecticidal activity against different insects[19-28,46,48-50].

Literature survey on the potential use of EOs as bio-pesticides indicated that EOs obtained from the plant families, including Asteraceae, Myrtaceae, Apiaceae, Lamiaceae and Rutaceae have insecticidal activity. They are active as a repellent, fumigant, larvicidal and adulticidal against insects in Lepidoptera, Diptera, Coleopteran,

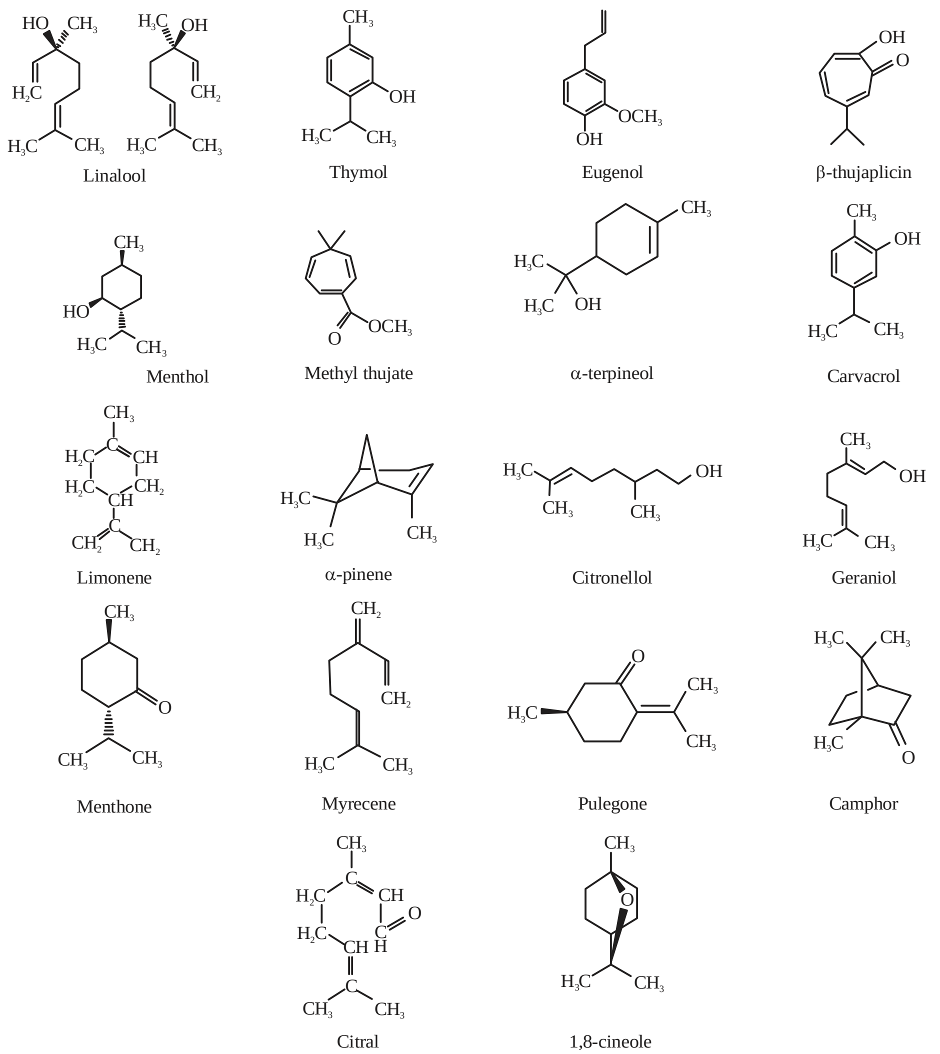


Fig. 1: Chemical structure of some monoterpenes found in essential oils with insecticidal activities

Hemiptera and Isopteran orders[34]. For example, EOs obtained from *Artemisia* species have insecticidal activity against some insects such as coleopteran beetles. They have vapor toxicity and repellent action against these insects[51-53]. Similarly, the numbers of monoterpenes were evaluated for their insecticidal activity. Monoterpenes (e.g., β-thujaplicin) obtained from the wood of Western crimson cedar (*Thuja plicata*) has insecticidal activity against larvae of the old house borer (*Hylotrupes bjulus*)[54]. In contrast, the low insecticidal activity from α and β-thujaplicin was observed against the termite, *Reticulothermes flavipes*. The (+)-3-thujone and (-)-3- iso-thujone accounted to 80-90% of the leaf EOs of this plant and have antifeedant activity against the white pine weevil[54]. Thymol has a contact toxicity to larva *Agriotes obscures* (L.) (Coleoptera: Elateridae)[55]. In this respect, several reports have shown insecticidal activity of terpenes such as citronellal[56,57], camphor[53,58,59], 1,8-cineole[60,61], a-terpineol[62] and α-pinene[63]. In addition, the insecticidal activities of EOs constituencies were evaluated. For example, pure geraniol, citronellol, citral and carvacrol have insecticidal activity. Also, cuminaldehyde, limonene, d-limonene, menthone, trans-anethole, thymol, 1,8-cineole and cinnamaldehyde have insecticidal activity against different insects[19,56,57,64,65,66]. Many compounds have been isolated, identified and evaluated for their insecticidal activities, e.g., cineole, geraniol and piperidine[67], linalool and limonene[68]. Some studies have reported insecticidal activity of piperitone against *Callosobruchus maculates*[69] and antifeedant activity of trans-ethyl cinnnamate[70]. Abdelgaleil *et al.*[71] has reported that both piperitone and trans-ethyl cinnamate obtained from *Artemisia judaica* L., EO showed antifeedant activity against *Spodoptera littoralis* (Boisd). However, Fig. 1 shows some chemical structure of terpenes found in some plant essential oil with insecticidal activities.

ESSENTIAL OILS AS BIOINSECTICIDES

Essential oils and other biopesticides are becoming a new trend for pest management in modern farming and organic agriculture. For 2 decades, EOs are used as insecticides to control insects, however, they have not reached their full potential because they are rapidly volatile and low residual activity. They are considered safe, environment-and-eco-friendly, compatible with biological control programs and have low mammals toxicity. Also, EOs are available worldwide for their low to moderate cost. The main active constituents with insecticidal activity are monoterpenes, sesquiterpenes and related phenylpropenes. The EOs can be applied as pesticides as they are or as their active components "Active ingredient" or as co-adjutants in pesticide formulations. However, they can be acted as a contact insecticide, causing changes in the pathways of biochemical metabolism of the insect, knockdown and rapid death[72], fumigants[73], repellents[74] and antifeedant[71].

Repellent activity: The repellent activity of EOs from numerous plants is well documented. This activity is related to major active compounds and other chemical constituents[7,75]. The repellent active compounds isolated from *Limnophila geoffrayi* and *Schizonepeta tenuifolia* are pulegone, linalool, eugenol, thymol and methyl chavicol[76,77]. While, other repellent compounds such as a-pinene, b-pinene, D-limonene, (E)-3, 7-dimethyl-, 2, 6-octadienal were isolated from *Armoracia rusticana*, *Pimpinella anisum*, *Allium sativum*, *Laurelia sempervirens* and *Drimys winteri*[78,79]. However, most of repellent activity studies were conducted on pests belonging to Coleoptera and Diptera species. Abd-Elhady[80] reported that EOs obtained from *Artemisia judaica* L., has repellent activity against the cowpea weevil, *Callosobruchus maculatus* (Fab.). It is concluded that EO at concentrations of 4.0, 8.0, 15.9, 31.9 and 63.7 μg cm^{-2} induced reduction in egg laying of *C. maculates* by 12.5, 42.7, 61.8, 86.0 and 92.5%, respectively. Garlic and mint EOs were evaluated against the cowpea aphid (*Aphis craccivora* Koch.)[81], which caused 100% repellent at a concentration of 4.0% on adult aphids, while the effect was decreased in a concentration-dependent manner. They also concluded that garlic oil has higher repellent than mint oil. This repellent action of EOs may be related to the major constituents, e.g., piperitone, camphor and (E)-ethyl cinnnamate.

An essential oil from *Citrus limonum* and *Litsea cubeba* have repellent activity against adults of *Tenebrio molitor*[82]. The authors reported that the repellent activity of these EOs may be due to the presence of D-limonene and 3,7-dimethyl-6-octenal of *C. limonum* EO and (E)-3, 7-dimethyl-2, 6-octadienal and (E)-cinnamaldehyde of *L. cubeba.* Zapata and Smagghe[79] showed that four EOs from the leaves and bark of *Laurelia sempervirens* and *Drimys winteri* have highly repellent activity against *Tribolium castaneum*. The repellent activity of *L. sempervirens* oil was 90% at low concentration 0.032 μL cm^{-2}, while the same effect was observed by higher concentration (3-10 times) of *D. winteri* oils after 4 h of exposure.

Pavela[83] evaluated 10 EOs for their repellent activity against *Meligethes aeneus*. They reported that EOs isolated from *Carum carvi* and *Thymus vulgaris* had the highest repellent activity and the repellent index accounted to 65.6 and 63.8%, respectively, while other EOs has a repellent index lower than 15%. Abdel-Sattar *et al.*[84] showed that EOs of *Schinus molle* (fruit and leaf) had insect repellent activity against *Trogoderma granarium* and *Tribolium castaneum*. They found that p-cymene was identified as a major component in fruits and leaf oils. In addition, GC-MS analysis demonstrate that monoterpenes presented the highest amounts and accounted for 80.43 and 74.84% in fruits and leaves, respectively. They also concluded that the repellent activity of the leaf and fruits of *S. molle* L. The EOs were 75.02 and 71.89%, respectively at a concentration of 1000 μL cm^{-2}. In this regard, the repellent activity of EO obtained from the leaves induced higher activity than that of the fruit.

The repellent activity of EOs obtained from *Mentha* longifolia and *Thymus kotschyanus* was evaluated against *Callosobruchus maculates*[85]. The repellent values at a concentration of 800 ppm were recorded 90 and 73.33% for *M. longifolia* and *T. kotschyanus* against *C. maculates*, respectively. Negahban and Moharamipour[53] found that EO of *Artemisia sieberi* at 1.5 ppm was more repellent to *Tribolium castaneum* than *Sitophilus oryzae* and *Callosobruchus maculatus*. The repellent values amounted 65.90, 59.70 and 55.80%, respectively. In contrast, *A. sieberi* EO showed a strong repellence activity against *T. castaneum*, *S. oryzae* and *C. maculatus*[86]. The repellent activity recorded 63.80, 62.01 and 48.57% for *T. castaneum*, *S. oryzae* and *C. maculatus*, respectively. Nikooei and Moharramipour[87] found that EO of *Salvia mirzayanii* at a concentration of 3 ppm had a strong repellent activity against *Tribolium confusum* (86.66%) than on *C. maculatus* (70.0%). In addition, EOs of *Anethum graveolens* L. (Apiaceae), *T. vulgaris* and *Rosmarinus officinalis* produced a strong repellent activity against *T. confusum* than on *C. maculatus*. The repellent values of these EOs were 100, 100 and 93.33%, respectively[88]. Saeidi *et al.*[89] showed that the EO of *Citrus reticulate* has higher repellent activity than *C. limon* and *C. aurantium*

Table 1: EOs of some plants with repellent activity

Plant			Insect		
Family	Name	Parts	Order	Name	References
Rutaceae	*Citrus limonum*	Leaves	Coleoptera	*Tenebrio molitor*	Wang *et al.*[82]
Lauraceae	*Litsea cubeba*				
Atherospermateceae	*Laurelia sempervirens*	Leaves and bark		*Tribolium castaneum*	Zapata and Smagghe[79]
	Drimys winteri				
Apiaceae	*Carum carvi*	Seeds		*Meligethes aeneus*	Pavela[83]
Lamiaceae	*Thymus vulgaris*	Aerial parts			
Anacardiaceae	*Schinus molle*	Fruit and leaf		*Trogoderma granarium*	Abdel-Sattar *et al.*[84]
				Tribolium castaneum	
Asteraceae	*Artemisia scoparia*	Leaves		*Callosobruchus maculatus*	Negahban *et al.*[86]
				Tribolium castaneum	
			Curculionidae	*Sitophilus oryzae*	
Lamiaceae	*Thymus kotschyanus*	Aerial parts	Coleoptera	*Callosobruchus maculatus*	Akrami *et al.*[85]
	Mentha longifolia				
Apiaceae	*Carum copticum*		Lepidoptera	*Plutella xylostella*	Jamal *et al.*[95]
Lamiaceae	*Perovskia abrotanoides*		Coleoptera	*Tribolium castaneum*	Arabi *et al.*[21]
			Curculionidae	*Sitophilus oryzae*	
Asteraceae	*Artemisia judaica*		Coleoptera	*Callosobruchus maculatus*	Abd-Elhady[80]
Lamiaceae	*Zhumeria majdae*			*Plodia interpunctella*	Karahroodi *et al.*[88]
Asteraceae	*Achillea wilhelmsii*		Lepidoptera		
Lamiaceae	*Hyssopus officinalis*				
Rutaceae	*Citrus reticulata*	Peel	Coleoptera	*Callosobruchus maculatus*	Saeidi *et al.*[89]
	Citrus limon				
	Citrus aurantium				
Apiaceae	*Carum copticum*	Fruits		*Tribolium castaneum*	Sahaf *et al.*[96]
Lamiaceae	*Salvia bracteata*	Aerial parts		*Callosobruchus maculatus*	Shakarami *et al.*[97]
			Curculionidae	*Sitophilus oryzae*	

against *Callosobruchus maculatus*. The repellent values of *C. reticulata* (peel essential oils) at a concentration of 1, 3, 5 and 7 ppm were 26.66, 33.33, 33.66 and 40.0%, respectively. The researchers confirmed that at concentration 7 ppm, the repellent activity of *C. reticulata* EO was higher than that of *C. aurantium*. The strong repellent activity of *Penstemon acaulis* EO was found against *S. oryzae*, *C. maculatus* and *T. castaneum*. The highest concentration 2 μL mL^{-1} of *P. acaulis* EO shows high repellence with values of 83.6, 71.6 and 63.6% for adult insects of *S. oryzae*, *C. maculatus* and *T. castaneum*, respectively[90].

The EOs extracted from plants with higher repellence included many active compounds. Examples of these active compounds are citronella, cedar, verbena, pennyroyal, geranium and lavender[76-78]. The repellent action was induced by pine, cinnamon, rosemary, basil, thyme, peppermint, p-cymene and pulegone. Also, linalool, eugenol, thymol, cymol and methyl chavicol have repellent activity against several insects[76-78,91]. Natural terpenoids found in crushed basil (*Ocimum basilicum* L.) leaves repels some thrips species[92]. These results show that linalool and eugenol have repellent activity. Also, the high repellent activity was shown by caryophyllene and 1,8-cineole. Some active compounds obtained from different EOs include methyl salicylate[93] and salicylaldehyde[94] have repellent activity. Table 1 illustrates examples of plants with repellent activity.

Insecticidal activity: The insecticidal activity of many EOs has been evaluated against a number of insects (Table 2). Previous studies have shown that the toxicity of EOs against a variety of insects is related to terpenes. They represent major of EOs components, especially monoterpenoids and sesquiterpenes[57,98-101]. Many EOs were analysis by GC/MS and the main active components were identified and tested on a variety of insects. For example, camphor (over 400 ppm) the major component of *Coriandrum sativum* and *Carum carvi* L. The EOs have high toxicity to *Rhyzopertha dominica* and *Cryptolestes pusillus*. While linalool (1617 ppm) found in coriander seed EO is toxic to *Sitophilus oryzae*. Carvone, limonene and (E)-anethole are the major active component found in caraway EO. Carvone (972 ppm) had high insecticidal activity against *Sitophilus oryzae*[57,98,99].

The insecticidal toxicity of many EOs and their chemical constituents were evaluated. For example, 1,8-cineole, carvacrol and eugenol have insecticidal activity[57,98-100]. In addition, limonene, α-pinene and thymol have a toxic effect against some insects[57,98-100]. The volatile toxicity of *Coriandrum sativum* and *Carum carvi* L., EOs were evaluated in the

Table 2: EOs of some plants with insecticidal activity

Plant			Insect		
Family	Name	Parts	Order	Name	References
Lamiaceae	*Ocimum basilicum*	Aerial parts	Hemiptera	*Aphis craccivora*	Sammour *et al.*[22]
Meliaceae	*Azadirachta indica*				
	Syzygium aromaticum	Flowers	Coleoptera	*Callosobruchus chinensis*	Tian *et al.*[106]
Apiaceae	*Coriander sativum*	Seeds		*Sitophilus oryzae*	Lopez *et al.*[107]
Umbelliferae	*Carum carvii*			*Rhyzopertha dominica*	
Lamiaceae	*Ocimum basilicum*	Leaves		*Cryptolestes pusillus*	
	Rosemarinus officinalis	Aerial parts	Hemiptera	*Aphis craccivora*	Abdel-Aziz *et al.*[26]
	Salvia officinalis				
Zingiberaceae	*Curcuma longa*	Leaves	Lepidoptera	*Agrotis ipsilon*	Abdelaziz *et al.*[101]
Lamiaceae	*Salvia offecinalis*				
Asteraceae	*Artemisia capillaris*	Aerial parts	Coleoptera	*Sitophilus zeamais*	Liu *et al.*[109]
	Artemisia mongolica				
Apiaceae	*Coriandrum sativum*	Seed		*Tribolium confusum*	Khani and Asghari[110]
				Callosobruchus maculatus	
Lamiaceae	*Origanum vulgare*	Leaves	Hemiptera	*Nezara viridula*	Gonzalez *et al.*[112]
	Thymus vulgaris				
Apiaceae	*Biforaradians bieberstein*	Aerial part		*Lipaphis pseudobrassicae*	Sampson *et al.*[113]
	Foeniculum vulgare	Fruit			
Lamiaceae	*Thymbra spicata.*	Aerial part			
Asteraceae	*Artemisia judaica*		Coleoptera	*Callosobruchus maculatus*	Abd-Elhady[80]
Lamiales	*Origanum onites*	Leaves	Lepidoptera	*Ephestia kuehniella*	Ayvaz *et al.*[5]
	Satureja thymbra			*Plodia interpunctella*	

laboratory. Where they show strong insecticidal activity against *S. oryzae*, *Rhyzopertha dominica* and *Cryptolestes pusillus*. The high activity of these oils can be due to the occurrence of linalool (1617 ppm of the oil) as a major compound[57,98-102]. Also, camphor-rich fractions (over 400 ppm) were very toxic to *Rhyzopertha dominica* and *Cryptolestes pusillus*[57,98-102]. The insecticidal activity of neemix® (4.5% EC, neem oil produced from *Azadirachta indica*) and basil oil (*Ocimum basilicum*) against *Aphis craccivora* Koch was evaluated[22]. They have shown insecticidal activity against *A. craccivora* Koch when the faba bean plants were treated systematically or by contact. They caused toxicity to adult stage of *A. craccivora* Koch and the accumulative mortality reached 100% after 7 and 8 days of treatment by basil oil and neemix oil, respectively. It has been reported that methyl chavicol, linalool and geranial are the major active component in basil oil. Therefore, the high insecticidal activity of this oil may be due to the presence of these active compounds[99]. In addition, many studyies have reported the insecticidal activity of essential oil against economic agriculture insects. Other studies reported toxicity of volatile methyl chavicol, linalool and geranial against insects, e.g., whitefly and cotton leaf worm[103,104].

In laboratory studies, EOs of rosemary (*Rosemarinus officinalis*), sage (*Salvia officinalis*) and curcuma (*Curcuma longa*) plants were formulated[26]. The formulated oils were applied as a soil treatment (systematically) or spray (contact) against sucking aphid (*Aphis craccivora* Koch) on *Vicia faba*. The accumulative mortality after 1 week of treatment by curcuma, sage and rosemary at a concentration of 1%, were 49.98, 63.33 and 93.33%, respectively[105]. Insecticidal activity of some terpenes such as carvone, linalool and terpeniol, which isolates from EOs (garlic and mint) were evaluated against *Agrotis ipsilon*. In addition to isolated phellandrine and citronellol were tested against *Agrotis ipsilon* (eggs, larva and pupae). Terpenes had different toxicity on eggs, larvae and pupae with different effects on growth and development. The high toxicity to egg and pupal stages were induced by phellandrine, while nerol acted as a stomach or contact poison to the second larval instar. They stated that combination exposure to citronellol plus garlic oil or mint oil increased their toxicity[105]. After 8 days of treatment with *Salvia officinalis* EO at concentration 4%, the mortality was 75% of *Agrotis ilpsilon* adult. The EO caused a high reduction in egg deposition (67.4%) and egg hatchability (69.4%) at sub-lethal concentration. Consequently, they induced more than 31.2% sterility in females[25]. It is reported that the insecticidal activity of *Salvia officinalis* might be due to the presence of terpenes and sesquiterpenes. The active components are camphene, α-pinene, β-pinene, myrcene and limonene. Moreover, 1,8-cinieole, α-thujone, β-thujone, camphor, linalool, bornylacetate and borneol have a high toxic effect[100].

The insecticidal activity of EO obtained from clove buds (*Syzygium aromaticum*) and their major constituents were evaluated against *Callosobruchus chinensis*[106]. At laboratory scale, their active components such as eugenol, eugenol acetate and β-caryophyllene were studied in the laboratory stage. The contact toxicity of EO and commercial eugenol was 99.00% and β-caryophyllene was 98.00%. The LD_{50} values for adults of *C. chinensis* found to be 0.730, 0.673 and 0.708 µg per adult, while that of nymphs were 1.795, 1.668 and 1.770 µg per nymph due to EOs, eugenol and β-caryophyllene, respectively. In comparison, LD_{50} values of commercial eugenol acetate (98%) were found[106] to be 92.66 and 99.42 µg per nymph. Lopez *et al.*[107] reported that *Coriandrum sativum* oil was very toxic to *S. oryzae*, *R. dominica* and *C. pusillus* and their main active ingredient linalool at a concentration of 1617 ppm. While, the fractions of camphor (over 400 ppm) were highly toxic to *R. dominica* and *C. pusillus*. The most effective component of caraway EO is carvone (972 ppm) a monoterpenoids that more toxic to *S. oryzae*. Sharaby *et al.*[108] evaluated the toxicity of essential oils of garlic, mint and *Eucalyptus* against the grasshopper (*Heteracris littoralis*). After 2 weeks of feeding on diet containing different concentrations of EOs, the LC_{50} values were estimated. The LC_{50} of these EOs were 0.067, 0.075 and 0.084 mL/100 mL diet of garlic, eucalyptus and mint, respectively. Liu *et al.*[109] reported contact toxicity of *Artemisia capillaris* and *A. mongolica* EOs against *Sitophilus zeamais*, which were 105.95 and 87.92 µg per adult insect, respectively. The insecticidal activity may be due to the main components found in the EO as 1,8-cineole, germacrene D and camphor of *A. capillaries* EO and α-pinene (12.68%), germacrene D and γ-terpinene of *A. Mongolic* EO[109]. Khani and Asghari[110] evaluated the EO of *Pulicaria gnaphalodes* against *T. confusum* and *C. maculatus* in the laboratory. They found that the major active components found by GC/MS of EO were linalool (57.57%) and geranyl acetate (15.09%). The toxicity of EO was studied by using the older adults (1-7 days) of insect. Mortality of insects was increased with increasing concentrations from 43-357 µL L^{-1} air along with exposure time from three to 24 h. The LC_{50} values of EO were 1.34 µL L^{-1} air on *C. maculatus* and 318.02 µL L^{-1} air of *T. confusum*. The toxicity of *P. gnaphalodes* EO can be due to their active ingredients, linalool[107] and camphor[111].

Origanum vulgare L. (oregano) and *Thymus vulgaris* L. (thyme) EOs were tested against *Nezara viridula*. Their active ingredients were p-cymene for oregano and thymol for thyme oil[112]. Their LC_{50} were 1.7 and 3.5 µg cm^{-2} for oregano and thyme for nymphs and 169.2 and 48.8 µg cm^{-2} for adults, respectively. Other studies confirmed that *Triaenops persicus* had insecticidal activity against adults of *T. castaneum* and *S. oryzae*[91]. However, the mechanism of toxicity of oregano and thyme may be due to their neurotoxic effect. In this regard, EO and their active compound thymol can interact with neuromodulator octopamine. They also can induce neurotoxicity via effects on gated chloride channels GABA. Thymol reported also to induce high toxicity to some insects such as *Lipaphis* pseudobrasicae[113], *Spodoptera litura*[56] and *S. oryzae*[12]. Another volatile oil of *Artemisia judaica* L., was specific against the cowpea weevil, *C. maculatus*. The seeds of the cowpea were treated with *A. judaica* EO with sub-lethal concentration, that has insecticidal activity against *C. maculatus* insect[80]. Their toxic effect was attributed to piperitone (32.4%), camphor (20.6%) and (E)-ethyl cinnnamate (8.2%). The previous compounds are monoterpenoids which are lipophilic have a fast penetration properties into insects which consequently interfere with biochemical and physiological functions[114]. Oils of *S. hortensis*, *T. serpyllum* and *O. creticum* at a dose of the 100 µg per larva were investigated. They induced more than 90% mortality in larval of tobacco cutworm, *S. litura* after 24 h of treatment. The LD_{50} values of *S. hortensis* and *T. ulgaris* were 48.4 and 46.9 µg per larva, respectively. The insecticidal action of *T. ulgaris* and *S. hortensis* species can be due to the presence of monoterpenoids, phenols, i.e., thymol and carvacrol[115]. However, lethal and sub-lethal doses of EOs can cause mortality and change in the fertility of insects[56,116].

The EOs of *Origanum onites*, *Satureja thymbra* at concentration 9 and 25 µL L^{-1} air was investigated against *E. kuehniella* and *P. Interpunctella*. After 24 h of treatment, the mortality was 100% for both insects[5]. Pavela[117] evaluated the toxicity of 34 EOs against *S. littoralis* larvae. The insecticidal activity of *Nepeta cataria* and *Thuja occidentalis* EOs against *S. littoralis* larvae were investigated by Wang *et al.*[82]. They found that the LC_{50} of these EOs were $\geq$10.0 mL m^{-3}. The EOs from other plants such as *Mentha* citrata, *Salvia sclarea*, *Origanum vulgare*, *Origanum compactum*, *M. officinalis*, *T. mastichina* and *L. angustifolia* have highly toxic effect against *S. littoralis* with $LC_{50}\leq$0.05 µL per larva[82].

Fumigants activity: Essential or volatile oils naturally are liquid at room temperature and get easy to change to vapors at room or with slightly higher temperature without any decomposition[91]. Therefore, the volatile oils are frequently used as a fumigant against insects in agriculture (greenhouse) and stored food[5,118]. Fumigants of EOs are very important for control-stored insects. The EOs have a broad activity spectrum against a variety of insects, by fast penetrating and

Table 3: EOs of some plants with fumigant activity

Plant			Insect		
Family	Name	Parts	Order	Name	References
Apiaceae	*Carum copticum*	Seed	Coleoptera	*Sitophilus oryzae*	Sahaf *et al.*[119]
Lamiaceae	*Zataria multiflora*	Leaves and stems	Hemiptera	*Brevicoryne brassicae*	Motazedian *et al.*[129]
Asteraceae	*Artemisia sieberi*				
	Tagetes minuta				
Apiaceae	*Anethum graveolens*	Fruits	Coleoptera	*Callosobruchus chinensis*	Chaubey[123]
	Cuminum cyminum				
Labiatae	*Schizonepeta tenuifolia*	Whole plant	Diptera	*Lycoriella ingenua*	Park *et al.*[77]
Illiciaceae	*Illicium verum*	Fruits	Blattoder	*Reticulitermes speratus*	Park and Shin[124]
Composite	*Cacalia roborowskii*	Whole plant			
Labiatae	*Schizonepeta tenuifolia*	Herba			
Liliaceae	*Allium cepa*	Bulb			
	Allium sativum				
Apiaceae	*Carum carvi*	Fruits	Coleoptera	*Sitophilus zeamais*	Fang *et al.*[125]
				Tribolium castaneum	
Lamiaceae	*Mentha microphylla*	Aerial parts		*Sitophilus oryzae*	Mohamed and Abdelgaleil[126]
Asteraceae	*Artemisia judaica*			*Tribolium castaneum*	
Rutaceae	*Citrus reticulata*	Fruits			
Lamiaceae	*Perovskia abrotanoides*	Flower		*Sitophilus oryzae*	Arabi *et al.*[127]
				Tribolium castaneum	
Atherospermateceae	*Laurelia sempervirens*	Leaves		*Tribolium castaneum*	Zapata and Smagghe[79]
Winteraceae	*Drimys winteri*				
Asteraceae	*Artemisia vestita*	Aerial parts		*Sitophilus zeamais*	Chu *et al.*[128]
Umbelliferae	*Carum carvi*	Fruits		*Sitophilus zeamais*	Fang *et al.*[125]
				Tribolium castaneum	
Rutaceae	*Citrus limonum*	Leaves		*Tenebrio molitor*	Wang *et al.*[82]
Poaceae	*Cymbopogon citratus*				
Lauraceae	*Litsea cubeba*				
Myristicaceae	*Muristica fragrans*				

non-toxic residues in the treated products[118]. For example, EO of *Carum copticum* has fumigant toxicity against *S. oryzae* and *T. castaneum*[119]. The high toxic effect of *C. copticum* EO was recorded at concentration 185.2 μL L^{-1}. They have fumigant toxicity against both insects and the LC_{50} were 0.91 μL L^{-1} for *S. oryzae* and 33.14 μL L^{-1} for *T. castaneum*. The mortality of *S. oryzae* reached 100% after 12 h of exposure time. The fumigant toxicity of *C. copticum* could be attributed to the presence of thymol and other monoterpenoids found as major constituents. Monoterpenoids are volatile and they induced toxic action quickly as fumigant due to their fast penetration into insects[61]. Some examples of plants EOs has fumigant activity are shown in Table 3.

It has been reported that pulegone, linalool and limonene have fumigant toxicity against *S. oryzae*. In addition, linalool and linalyl acetate, that found in *Mentha citrata* EO are fumigate toxicity against this insect[120]. However, Apiaceae family is one of the most promising fumigant agents. The fumigant activity of some EOs of this family has reported against several stored product insects. For example, Kim *et al.*[121] found that over 90% mortality of *S. oryzae* and *C. chinensis* was induced by *Foeniculum vulgare* after 3 or 4 days of treatment. Fumigant toxicity of 22 EOs against *Acanthoscelides obtectus* was tested[122]. They found that monoterpenoids such as thymol, carvacrol and terpineol are effective in inhibiting insect reproduction. Chaubey[123] evaluated the fumigant toxicity of EOs from *A. graveolens* and *C. cyminum* against *C. chinensis*. The LC_{50} values were 10.8 and 11.0 μL L^{-1} for adult of *C. chinensis* after 24 h of exposure. Park *et al.*[77] pointed out that the EO of *S. tenuifolia* at concentration 12.5 μg mL^{-1} air caused 96.6% mortality in larvae of *L. ingénue*. They reported that the fumigant toxicity of *S. tenuifolia* can be attributed to pulegone, menthone and limonene as a major active compound of volatile oil of *S. tenuifolia*. The LC_{50} values of these components were 1.21, 6.03 and 15.42 μg mL^{-1}, respectively. In addition, more than 90% mortality in *J. termite*, *R. speratus* Kolbe was recorded after 3 days of fumigant treatment with EO of *O. japonica* at concentration 3.5 μL L^{-1} air[124]. In contrast, during the first 2 days of treatment, 100% mortality was recorded of *I. verum*, *C. roborowskik*, *S. tenuifolia*, *A. cepa*, *A. sativum* and *E. caryophyllata* EO at concentration 2.0 μL L^{-1} air.

Various studies have reported strong fumigant activity of EOs against storage insects. Fang *et al.*[125] reported that EO of caraway had fumigant toxicity against *S. zeamais* and *T. castaneum*. The LC_{50} was 3.37 and 2.53 mg L^{-1} for

Table 4: EOs of some plants with antifeedant activity

Plant			Insect		
Family	Name	Parts	Order	Name	References
Asteraceae	*Artemisia judaica*	Aerial parts	Lepidoptera	*Spodoptera littoralis*	Abdelgaleil *et al.*[71]
Apiaceae	*Anethum graveolens*	Seed		*Pseudaletia unipuncta*	Sousa *et al.*[132]
	Petroselinum crispum				
	Foeniculum vulgare	Stems and leaves			
	Cuminum cyminum				
Poaceae	*Cymbopogon nardus*	Aerial parts		*Euprosterna elaeasa*	Hernandez-Lambrano *et al.*[133]
	Cymbopogon flexuosus			*Acharia fusca*	
	Cymbopogon martinii				
Asteraceae	*Eupatorium adenophorum*	Flowers and leaves	Coleoptera	*Rhynchophorus ferrugineus*	Shukia *et al.*[134]
	Artemisia nilagirica	Aerial parts			
Lamiaceae	*Satureja khuzistanica*	Leaves and flowers		*Leptinotarsa decemlineata*	Saroukolai *et al.*[137]
	Ocimum basilicum				
	Thymus daenensis				
	Mentha spicata				
Myrtaceae	*Myrtus communis*				
	Eugenia caryophyllus				
	Eucalyptus globulus	Leaves and flowers		*Tribolium castaneum*	Ebadollahi[138]
Lamiaceae	*Lavandula stoechas*				
	Thymus pseudopulegioides	Aerial parts		*Agelastica alni*	Bekircan *et al.*[139]
	Thymus transcaucasicus				
	Thymus leucotrichus				
	Teucrium polium				
	Satureja khuzistanica	Leaves and flowers		*Leptinotarsa decemlineata*	Saroukolai *et al.*[137]
	Ocimum basilicum				
	Thymus daenensis				
Myrtaceae	*Myrtus communis*				

S. zeamais and *T. castaneum*, respectively. Fumigate toxicity of some EOs against *T. castaneum* adults were studied. For example, the LC_{50} of EOs were 6.84 mg L^{-1} and LC_{50} 4.51, 19.47, 20.50, 11.39 and 9.0-10.5 µL L^{-1} of *M. exotica*[126], *T. polycephalum*[127], *L. sempervirens*, *D. winteri*[79] and *A. vestita*[128], respectively. Motazedian *et al.*[129] studied the fumigant toxicity of *Z. multiflora*, *N. cataria*, *T. minuta* and *A. sieberi* EOs against the adults of *B. brassicae*. All EOs were toxic in a concentration-dependent manner. After 24 h of treatment with *N. cataria* EO, the mortality were 24, 36, 46, 52, 76 and 94% of concentrations 3, 8, 16, 31, 63 and 126 µL L^{-1} air, respectively. The fumigant toxicity of *N. cataria* EO may be due to -α, 7-β, 7a-α-nepetalactone which accounted 76.8% of this EO as shown by GC/MS analysis. Also, Tayoub *et al.*[100] recorded the fumigant toxicity of EO obtained from *S. officinalis* against *T. granarium*. They reported that the activity of *S. officinalis* can be due to their occurrence of monoterpenes (57.3%) and sesquiterpenes (41.7%). The EOs of *P. gnaphalodes* and *A. wilhelmsii* were tested for fumigant toxicity against *C. maculates* and *T. castaneum*[110]. They found that the LC_{50} values ranged from 1.54-2.65 µL L^{-1} air against *C. maculates.* While, LC_{50} were 10.02 µL L^{-1} air of *A. wilhelmsii* and *M. longifolia* against *T. castaneum*, respectively.

Antifeedant activity: Antifeeding are compounds or substances that reduce insect feeding or consumption. This substance induces deter feeding by modification the insect behavior, through a direct action on peripheral sensilla of insects[130]. The antifeedant activity of many EOs has been tested against a number of insects as shown in Table 4. Huang *et al.*[131] reported that pogostone which isolated from *Pogostemon cablin* EO has highly antifeedant activity against *Spodoptera litura* and *Spodoptera exigua.* At concentration, 250 mg L^{-1} the time for feeding was shorter. Also, antifeedant activities were found 92.03 and 99.32% at concentration 4000 mg L^{-1} against *S. litura* and *S. exigua,* respectively. Abdelgaleil *et al.*[71] evaluated two antifeeding compounds (piperitone and trans-ethyl cinnamate) from EO of *A. judaica* against the third instar larvae of *S. littoralis*. Antifeedant activity (100% inhibition) was recorded at concentration 1000 µg mL mg L^{-1}. Sousa *et al.*[132] evaluated the antifeedant activity of EOs from *A. graveolens*, *P. crispum* and *C. cyminum* and 11 pure compounds against *Pseudaletia unipuncta*. They found that EOs from *P. crispum* and *A. graveolens* fruit and two pure compounds (trans-anethole and cuminaldehyde) have antifeedant activity and the inhibition of the larvae feeding account more than 70%. The antifeedant activity of

Cymbopogon nardus, *C. flexuosus* and *C. martini* was determined against larva of *A. fusca* and *E. elaeasa*. Strong antifeedant activity was observed at concentrations between 0.002-0.600 μL cm^{-2} of these EOs[133].

Shukia *et al.*[134] evaluated the antifeedant activity of *E. adenophorum* and *A. nilagirica* against adults of *R. ferrugineus*. After 96 h of treatment, both EOs shown significant antifeeding effects at concentrations 1000 ppm. The feeding prick-marks of adult insect were 60.13±11.31 and 60.13±7.94 for both EOs, respectively. The antifeedant activity of EOs can be due to its main constituents, camphor. Moreover, they reported that the minor constituents of EOs play an important role in changing the activity by synergistic effects. However, it has been reported that EOs plays an important role in plant defense systems. In general, the mixture of the chemical composition of EOs is more effective than that of individual pure compounds. Therefore, synergistic effects between EOs components are playing an essential role in the EOs activity[135,136]. Saroukolai *et al.*[137] evaluated the antifeedant activity of some EOs from *Satureja khuzistanica*, *O. basilicum* and *Myrtus communis* in addition to EOs from *T. daenensis*, *M. spicata* and *E. caryophyllus* against *L. decemlineata*. They found that the EO of *S. khuzistanica* was the most antifeeding activity compared to the other EOs against 4th instar larvae and adult insect. Ebdadollahi[138] studied the antifeedant activity of *Eucalyptus globulus* and *Lavandula stoechas* EOs against *T. castaneum*. All the tested EOs caused significant reductions in the feeding of insects. Also, either the oils or their vapors had antifeedant activity and the effect was increased by increasing EOs concentrations.

Bekircan *et al.*[139] found the strong antifeedant activity of EO obtained from *Thymus leucotrichus* against *Aphrophora alni* after 3 days of treatment. The EO at concentration 2000 ppm caused the highest antifeedant index (AFI). Also, the AFI value was 41.055 after 2 days of treatment. Another study by Rana *et al.*[140] showed the antifeedant activity of *Vitex negundo* EO against *C. chinensis* and *S. oryzae*. They found that EO at concentration 0.062-0.5% possess moderate antifeedant effects against *C. chinensis* and *S. oryzae*. While, at concentration 0.25% and upto 0.58 and 1.69% had strong antifeedant activity against both insects.

The highest antifeedant activity was observed after treatment with the *Vinca rosea* and C*allistemon lanceolatus* EOs and their combinations at ratio 1:1, 1:3 or 3:1 v/v against *Helicoverpa armigera*[141]. The percent of feeding inhibition (AI_{50}) was highest for *V. rosea* and *C. lanceolatus* EOs. The AI_{50} values were 5536 ppm and 6346 ppm for *V. rosea* and *C. lanceolatus* after 1 day of treatment. Strong antifeedant activity was recorded after treatment with the EOs combination at ratio 1:1. The AI_{50} was 4808 ppm of *C. lanceolatus* at concentration 10%, which induced 100% deterrence of feeding of *H. armigera*. After 4 days of exposure, the weight gain of the larva was decreased. Saroukolai *et al.*[137] determined the feeding index of EOs of *S. khuzistanica*, *O. basilicum*, *M. communis*, *Thymus daenensis*, *M. spicata* and *Eugenia caryophyllus* against *Leptinotarsa decemlineata*. All EOs shownantifeedant activity against 4th instar larvae and adults of insect. The EO of *S. khuzistanica* show highest antifeedant activity than other EOs. The antifeedant activity can be due to the main constituent, carvacrol, which has an insecticidal activity against important insects of agriculture and stored products[114,142]. It has been found that the eugenol obtained from *Laurus nobilis* (Lauraceae) EO has antifeedant activity against *Mythimna unipuncta*[143]. Moreover, some EOs such as *Artemisia tridentata*, *P. tridentate* and *Chrysothamnus nauseosus* shown antifeeding properties against *L. decemlineata*[144]. The EO of *S. officinalis* at concentration 4% had antifeeding activity (63.6%) against 2nd instar larvae of *A. ipsilon*[23].

Insect Growth Regulators (IGR) activity: Previous studies reported that several EOs and their constituents have properties similar to juvenile hormone and act as IGR (Table 5). They cause disruption in growth and affect the reproduction of insects[101,145,146]. For example, EOs obtained from several plants such as *A. graveolens*, *C. cyminum* and *I. verum* have reproductive activity against some insects. Also, EOs from *M. fragrans*, *N. sativa*, *P. nigrum* and *T. ammi* were induced changes in growth and reproduction of *C. chinensis*[123]. This disruption in growth of insects could be due to the inhibition of different biosynthetic processes of insects at different growth stages[145]. Abbas *et al.*[147] reported that EO of *C. reticulata* induced growth inhibition and decline of *R. domonica* insect population. Several essential oils are good inhibitors of pest's oviposition in that way they lead to disturbing the general growth of the populations. The EO of *E. cardamomum* has high activity on oviposition deterrence of *C. maculatus*. Therefore, treatment with this EO was reduced the numbers of insects in the treated grain[148]. In the same way, EO obtained from *Citrus* peels caused a high reduction in oviposition of insects e.g., *C. maculates*[135,149]. Both neemix and basil oil caused the prolongation of the nymphal duration and reduce the number of adult stage of *A. craccivora*[22]. Anshul *et al.*[150] studied the effect of EO of *A. annua* on the reproduction of larvae of *H. armigera*. The diet contains EO-induced 69.71% reduction in larva weight after feeding on the diet. Also, after treatment with *A. annua* EO, the larva stage was extended to 19.5 days. Maggi *et al.*[151]

Table 5: EOs of some plants with IGR activity

Plant			Insect		
Family	Name	Parts	Order	Name	References
Apiaceae	*Anethum graveolens*	Seed	Coleoptera	*Callosobruchus chinensis*	Chaubey[123]
	Cuminum cyminum				
Schisandraceae	*Illicium verum*				
Myristicaceae	*Myristica fragrans*				
Ranunculaceae	*Nigella sativa*				
Piperaceae	*Piper nigrum*				
Apiaceae	*Trachyspermum ammi*				
Rutaceae	*Citrus paradisi*	Fruit peels		*Rhyzopertha dominica*	Abbas *et al.*[147]
	Citrus reticulata				
Asteraceae	*Artemisia annua,*	Leaf and seed		*Helicoverpa armigera*	Anshul *et al.*[150]
Lamiaceae	*Ocimum basilicum*	Aerial parts	Hemiptera	*Aphis craccivora Koch*	Sammour *et al.*[22]
	Salvia offecinalis	Leaves	Epidoptera	*Agrotis ipsilon*	Abdelaziz *et al.*[101]
Apiaceae	*Carum copticum*	Fruits	Coleoptera	*Callosobruchus maculatus*	Sahaf and Moharamipour[154]
Apocynaceae	*Vinca rosea*	Leaves	Lepidoptera	*Helicoverpa armigera*	Halder *et al.*[141]
Myrtaceae	*Callistemon lanceolatus*				
	Eucalyptus globules		Lepidoptera	*Agrotis ipsilon*	Jeyasankar[155]
Ericaceae	*Gaultheria procumbens*				
Asteraceae	*Ageratum conyzoides*		Coleoptera	*Callosobrochus maculatus*	Aboua *et al.*[156]
Rutaceae	*Citrus aurantifolia*	Fruits			
Myrtaceae	*Melaleuca quinquenervia*	Leaves			
Apiaceae	*Trachyspermum ammi*	Fruits		*Tribolium castaneum*	Chaubey[157]
	Anethum graveolens				
Ranunculaceae	*Nigella sativa*				

reported that the reduction in growth and weight of larva might due to the disruption of insect hormone. In addition, EO of *P. interpunctella* inhibited the fertility of adult *A. annua*. The change in fecundity and fertility may be due to decrease in biochemical parameters. For example, protein and lipid were decreased after larva treatment with *M. azedarach*[152]. Basil oil caused changes in biochemical biomarkers (AST, ALT and ALP) in *A. craccivora*[22]. Several studies have been shown the oviposition deterrence of various EOs such as *T. kotschyanus* on *C. maculatus*[85]. In addition, *T. vulgaris* EO caused oviposition on *C. maculates*[153]. In the laboratory, oviposition deterrence was recorded of *C. copticum* and *V. pseudonegundo* EOs on *C. maculates*[154]. Jeyasankar[155] observed that larva of *Agrotis ipsilon* exposed to gaultheria oil shows the highest deformed development of larvae, pupae and adults.

Attractants activity: Essential oils are a complex mixture of different groups of chemical compounds. They contain monoterpenes, sesquiterpenes and phenylpropanoid compounds. The EOs have numerous activities against insects such as toxic, fumigant, repellent, ovicidal, larvicidal and antifeedant activities. Moreover, many volatile compounds in EOs such as monoterpenes and other are documented attracts activity which helpful for the control and monitoring the insect pests[121]. For example, Hernandez-Sanchez *et al.*[158] reported that p-cymene and limonene that isolated from *Mangifera indica* fruit had attractant activity. They have a high attractants activity to male and female *Ceratitis capitata*. Katerinopoulos *et al.*[159] found that 1,8-cineole is the main constituent of *Rosmarinus officinalis* essential oil. *Frankliniella occidentalis* is attracted by 1,8-cineole. Also, it is reported that 1,8-cineole, p-anisaldehyde, linalool and salicylaldehyde can be used for control or monitoring of *F. occidentalis* on greenhouse crops. It has been reported that essential oil from the peels of *Citrus limonum* that contains a number of terpenes like geraniol has attracted the insects. They attract thrips and *Japanese bettles*. Also, they attract several insects when the EOs adds to traps, for example, thrips, leafminer, aphid and whitefly[91]. In addition, cis-jasmone attracted adult Lepidoptera[160] and sandalwood oil, basil oil and grapefruit oil has shown a high attractiveness for whitefly[161].

MECHANISM OF ACTION OF ESSENTIAL OILS

The mechanism of toxicity and site of action of EOs and their chemical compounds as bio-insecticides were investigated[130,162]. The biological activity of EOs and their active components against insects were reported. They have insecticidal[57,98-102], repellent[75-81], antifeedant[130-133] and IGR[101,145,146] activities. This fact shows that the EOs can disrupt insect physiology in different ways. However, the activity of EOs as insecticides shows to be the action of their effect on

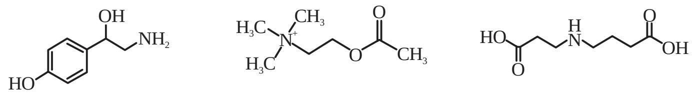


Fig. 2: Octopamine, acetylcholine and γ-aminobutyric acid neurotransmitter

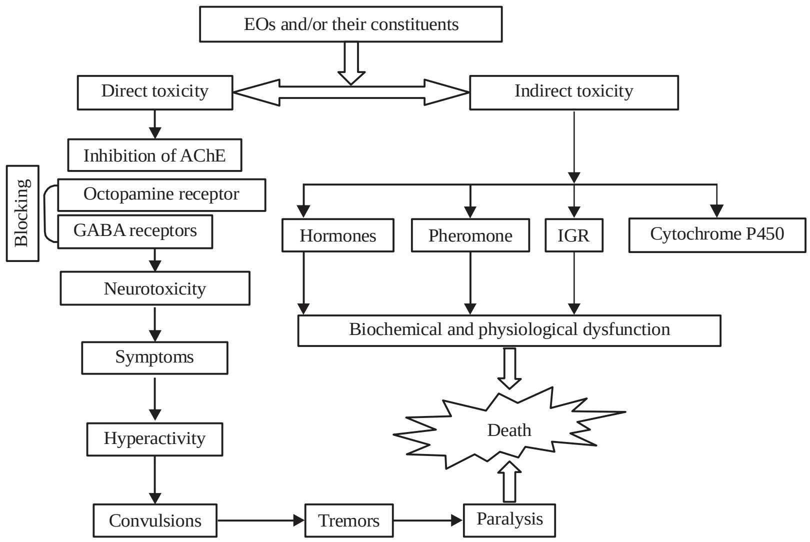


Fig. 3: Proposed mechanism of EOs-induced toxicity to insects

the insect nervous system. This effect, including the neurotoxic action of EOs by inhibition of acetylcholinesterase (AChE) or by blocking the octopamine receptors[163]. Also, γ-aminobutyric acid (GABA) receptor was suggested as one of the mechanisms of EOs induce toxicity in insects. They can interfere with GABA-gated chloride channels in insects[164]. For example, some monoterpenes such as thujone can induce the neurotoxic effects by acting GABA receptors in insects[165,166]. Besides, these modes of action of EOs on insects, some effects on the hormone and pheromone system and cytochrome P_{450} monooxygenase were reported[167]. However, some insect neurotransmitter is shown in Fig. 2.

Infact, octopamine is a neurotransmitter, neurohormone and circulating neurohormone-neuromodulator. Its disruption results in stop working of the nervous system in insects[163]. The octopamine receptor was not found in vertebrates. Therefore, EOs as bioinsecticides has selective toxicity to mammalian. Concerning the anti-cholinesterase effect of EOs, the mechanism of neurotoxic action and symptoms are similar to that induced by organophosphates and carbamates insecticides[130]. The symptoms show hyperactivity, convulsions, tremors and then paralysis[163].

Several studies point out that EOs and monoterpenoids induce mortality by inhibiting AChE activity in insects. The neurotoxic effect of EOs may be due to the inhibition of AChE at the hydrophobic site[168]. However, some monoterpenoids have anticholinesterasic e.g., pulegone[162], 1,8-cineole[169], fenchone, carvone and linalool[170]. In addition, the toxicity of EOs constitutions has resulted in the octopaminergic nervous system of insects[163]. In contrast, Emekci *et al.*[171] reported that monoterpenes can effects on many targets in insects with numerous mechanism of toxicity. However, proposed mechanism of EOs-induced toxicity to insects are illusterated in Fig. 3.

STRUCTURE-ACTIVITY RELATIONSHIPS

The relation between chemical structures of EOs constitutes and their biological activity were studied. It has been reported that slight change in molecular structure can make strong changes in their biological activity. For example, the modifications of the chemical structure of monoterpenoids from EOs can lead to enhanced the biological action of EOs and monoterpenoids that contain the functional groups. The activities of EOs and their constituents are dependent on the functional group (nature and position) and chemical properties such as volatility and molecular weights[172]. In the course of the importance of studying the

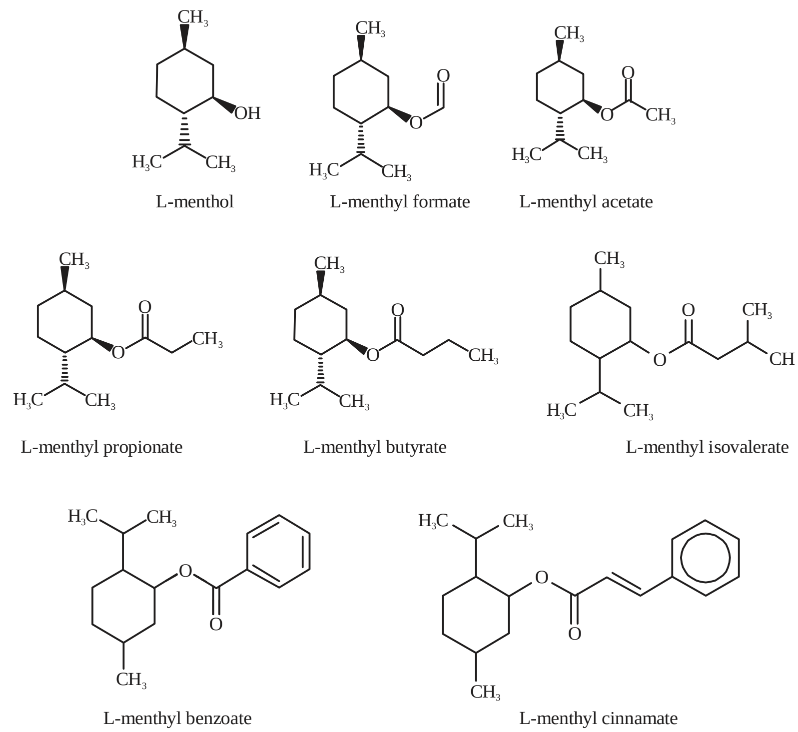


Fig. 4: Chemical structures of L-menthyl and its derivatives

relationships between chemical structure and biological activity, the knowledge of important functional groups and best chemical structure can be illustrated through biorational design of the derivatives according to the results obtained from bioassay experiments[172].

Some researchers have studied the correlation between the chemical structure of EOs constituents and their insecticidal activity. For example, EOs from *M. arvensis* has been reported to have insecticidal activity. The L-menthol isolated from this EO and seven of its acyl derivatives were evaluated against storage insects[173]. They reported that menthyl propionate and L-menthol were high insecticidal activity. The high activity of derivatives (menthyl propionate) as compared to L-menthol can be due to increasing the number of methyl groups in the side chain. Due to the nucleophilic properties of the methyl group (electron-donating), increasing the number of the methyl groups on the side chain cause decreasing in the positive charge (increase negative charge) on the carbon atom. Therefore, high activity of menthyl propionate may be due to increasing the negative charge on the carbon atom because of the presence of two methyl groups (Fig. 4). However, the increase of electrophilic groups such as methyl groups in the chain leads to increase the activity of a function carbon atom. For example, the low insecticidal activity of format derivative was found because it didn't contain the methyl group. Depending on the number of methyl groups, acetate derivative (-CH_3) have the slight activity while menthyl propionate (-$(CH_3)_2$) have high insecticidal activity. Low activity of menthyl benzoate was observed because they do not have methyl group and the benzene ring was attached directly to menthyl carbon. While, moderate activity was observed of menthyl cinnnamate because they have a double bond and benzene ring not directly attached to the carbon atom[34,173].

Nine benzene derivatives (eugenol, isoeugenol, methyl eugenol, safrole and isosafrole) and terpenes (cineole, limonene, p-cymine and α-pinene) were tested for insecticidal activity[174]. It is observed that derivatives of benzene have higher insecticidal activity than that of terpenes. This toxicity may be due to the active groups that found in benzene derivatives. For example, the insecticidal activity of eugenol and its chemical analogues isoeugenol and methyl eugenol. The presence of a double bond in the side chain of aromatic ring and the substitution of the methoxy group is playing an important role in the toxicity of these analogues. The knock down was increased in methyl-eugenol due to the further methoxy group. The order of contact toxicity (knock down) of these compounds was methyl-eugenol>isosafrole = eugenol>safrole. In contrast, when the double bond of the side chain is nearer to aromatic ring, the fumigant toxicity was decreased. Therefore, safrole is a more fumigant activity than isosafrole. However, the

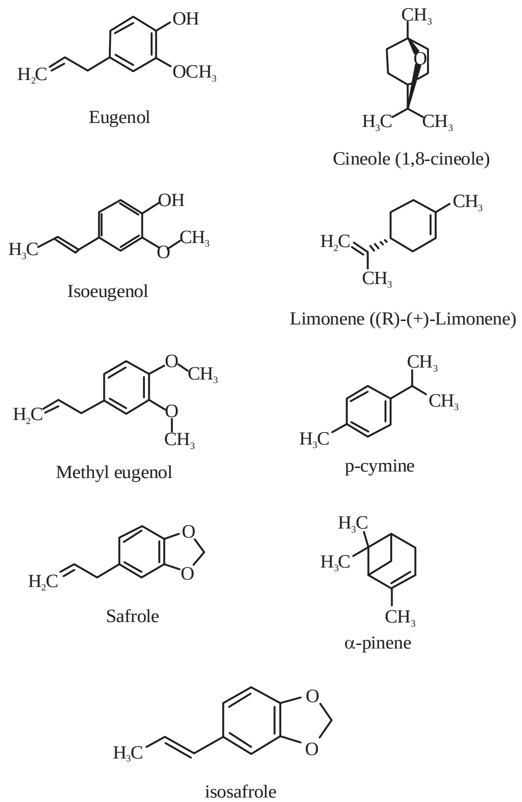
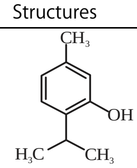
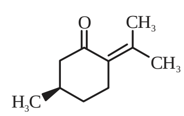
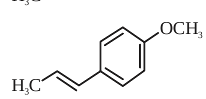
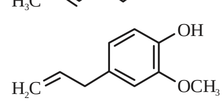


Fig. 5: Chemical structures of some benzene derivatives and terpenes

Table 6: Insecticidal activity of some essential oil constituents against *Spodoptera litura*

Compounds	Structures	LD_{50} µg per larva
Thymol		25.4
Pulegone		51.6
Trans-anethole		65.5
Eugenol		157.6

benzene derivatives have more insecticidal activity than monoterpenes[174]. In the mean time, chemical structures of some benzene derivatives and terpenes are shown in Fig. 5.

A good example of the correlation between the chemical structure of EOs constituents and their insecticidal activity was shown in Table 6. The insecticidal activity of thymol, pulegone, trans-anethole and eugenol were evaluated against *Spodoptera litura*[56]. Thymol has a higher toxicity than pulegone, trans-anethole and eugenol against *S. litura*. Their LD_{50} values were 25.4, 51.6, 65.5 and 157.6 µg per larva for thymol, pulegone, trans-anethole and eugenol, respectively. The order of toxicity of these compounds was thymol>pulegone>trans-anethole>eugenol. The high toxic effect of thymol as compared to other compounds attributed to the presence of methyl groups (-CH_3) in the side chain. This effect can be due to the electron-donating of methyl groups which decrease the positive charge on the carbon atom of the side chain. Also, some factors are effective on the toxicity the position of the double bond and the methoxy groups (Table 6).

SYNERGISTIC ACTION OF EOs

Various studies have investigated the synergistic effect of EOs, active components and their combinations against insects. Abbassy *et al.*[175] reported that terpinen-4-ol and c-terpinene isolated from *M. hortensis* EO has a synergistic effect. They increased the insecticidal activity of profenofos and methomyl by 2-4 folds against larva of *S. littoralis*. Another study by Regnault-Roger[176] showed that *M. hortensis* EO has a highly insecticidal activity than that of α-terpinene and terpinen-4-ol active compounds. They reported that the high toxic effect of EO against *S. littoralis* and *A. fabae* might be due to the synergistic effect of EO active compounds. Also, some studies have reported a synergistic effect of some EOs from *Ocimum* spp., *Conyza newii* and *Plectranthus marruboides* against some insects[177,178]. The synergistic effects of these EOs may be due to the different mechanism of action of their chemical constitutes. For example, the mechanism of toxicity of monoterpenes and some EOs is inhibiting AChE and some is neurotoxic[142]. In addition, monoterpenes can effects on many targets in insects with numerous mechanism of toxicity[171]. Jiang *et al.*[179] evaluated the insecticidal activity of EOs from *L. pungens*, *L. cubeba* and the mixture of major constituents against *Trichoplusia ni* larvae. The major active constituents of both EOs are 1,8-cineole and γ-terpinene from *L. pungens* and *L. cubeba*, respectively. The LD_{50} values of both EOs were 87.1 and 112.5 µg per larva, respectively. Also, the toxicity of individual major active constituents was lower than the toxicity of EOs. This result indicated that the synergistic effect of artificial mixtures of active and inactive compounds. Therefore, all active and inactive components are required to complete full toxicity. Faraone *et al.*[180] reported that the important role of second compounds and their synergistic effect with the major component in the toxicity of EOs. They found that the synergistic action of the mixtures of imidacloprid and EOs from *L. angustifolia* and *T. vulgaris* against insects increased the toxicity by 16-20 fold. The mixtures of imidacloprid and two active compounds linalool and thymol occurred dissimilarity and lower synergistic effect. Khalfi *et al.*[181] studied the insecticidal activity of spearmint EO and two active compounds 1,8 cineole and carvone against *Rhyzopertha dominica*. They found that the activity is due to the synergistic action of 1,8 cineole and carvone. It has been reported that EOs combination or in mixtures with botanical and synthetic insecticides has shown synergistic activity against variety insects[56,179].

ESSENTIAL OIL-BASED INSECTICIDES FROM RESEARCH TO MARKET

Aromatic plants have been used traditionally for control stored product insects worldwide. Consequently, it is normal to re-evaluate the EOs to be used against insect-pests. Some EOs and active substances with a special status where it can be used without the requirement of expensive and takes a long time for production. These requirements include toxicological and ecotoxicological tests that are necessary for registering commercial products. Dependent on the exception from federal regulatory approval in the USA. The EcoSMART technologies in the USA introduced some pesticides based on EOs[182]. They used EOs from rosemary, peppermint, cinnamon, lemongrass and thyme in 1998. Currently, Prentiss Inc. markets some qualified products, e.g., urban pest management such as pest controls. While, agricultural products are marketed by Brandt Consolidated Inc. In addition, Mycotech corporation produced an aphidicide/miticide/fungicide formulation. They use for greenhouse, horticultural crops and tree fruits. These formulations are based on cinnamon oil with cinnamaldehyde (30% EC). The EcoSMART technologies have introduced insecticides EcoPCO^R. They contain eugenol and 2-phenethyl propionate as active ingredients. They were used against crawling and flying insects and for pest control professionals. The EcoTrol^{TM} formulation is based on rosemary oil as the active ingredient. They used as an insecticide/miticide on horticultural crops. Also, garlic oil-based insecticide was produced in the US. These formulations contain mint oil as the active ingredient. They are used in the home and garden for pest control[183]. However, EOs have some properties that make them suitable for insect management[182]. These contain:

- EOs is produced in large global scale
- They have broad activity against many insects due to their multiple modes and site of action
- They have many activities such as insecticidal, repellent, fumigant, antifeedant and attractive
- EOs and their active constituents are nontoxic to mammals. The LD_{50} for rats were 800-3,000 mg kg^{-1} for pure compounds and $\geq$5,000 mg kg^{-1} for formulating insecticides
- Due to their volatility, the oils and active compounds are environmentally non-persistent
- They are effective under low pest pressure
- They mix with synthetic insecticides (tank-mixed) and are suitable for biological control programs
- They do not have to harvest or worker re-entry restrictions for treating crops
- They have a short residual half-life on plants

NANOINSECTICIDES BASED ON ESSENTIAL OILS

Although, the promising activities of EOs against many insects, some problems were registered. For example, EOs volatility, water solubility and oxidation, that playing an important role in the EOs activity, application and persistent. Therefore, these problems must be resolved before using the EOs as an alternative to synthetic pesticides for pest control[184]. New formulations with nanotechnology "Nanoformulation" can resolve these problems. The new trend for using nanoformulation lead to protect the EOs from degradation, increase their residue half-life by reducing the evaporation. They can achieve a controlled release of EOs and ease of application and handling[185]. These nanoformulations can enhance EOs activity due to the small particle size. It had a high surface area, solubility and mobility. Due to their solvent elimination, they have low toxicity to mammals[186-188]. However, polymeric nanoparticles are the most promising for EOs nanoformulations[188].

Anjali *et al.*[189] reported that the insecticidal activity of neem oil was increased in nanoemulision formulation. This effect can be due to the smallest droplet size of EO nanoemulision (31.03 nm). Yang *et al.*[190] showed that loaded nanoparticles with garlic EOs are active to control *Tribolium castaneum*. New nanotechnology methods were planned to control *H. armigera*[191]. As a result of this new method EO of *Artemisia arborescens* was stable. This stability can be due to built-in of EO with solid lipid nanoparticles and developed an emulsion. This emulsion has gotten better stability and insecticidal activity[192]. Nanoencapsulation shown high repellent activity than EOs[193]. For example, nanoencapsulation of *Artmisia* oil shown more activity than oil at concentration 1.9 ppm. The activity of nanocapsule was 80 and 62% for pure oil against *P. xylostella*.

SAFETY OF ESSENTIAL OILS

In general, the common use of plant essential oils in drugs and foods strength expectant the EOs are without significant toxicity to mammals. As shown by Isman *et al.*[182], most of the EOs and their active constituents are nontoxic to mammals. The LD_{50} for rats were 800-3,000 mg kg^{-1} for pure compounds and $\geq$5,000 mg kg^{-1} for formulating bioinsecticides. Moreover, due to the safety of some EOs and their active substances, they can be used without toxicological and ecotoxicological studies which are essential for registering commercial products[182]. On the other hand, some important exceptions were recorded. For example, pennyroyal oil was considered as poisonings to human and animal. The pulegone isolated from this oil is toxic to rats with LD_{50} 150 mg kg^{-1} i.p.[194]. Also, α-thujone from wormwood oil is very toxic to rats with LD_{50} = 45 mg kg^{-1} i.p.[165].

CONCLUSION

It can be concluded that many essential oils are toxic to a wide range of insect pests. They have insecticidal, fumigant, antifeedant, attractive and repellent activities against a broad spectrum of insects with some selectivity. The EOs are a complex of chemical compounds with multiple modes of action that enhances their activity due to the synergistic action between constituents. Due to their volatility in nature, EOs are used as a fumigant against agriculture and storage food insects. Consequently, essential oil-based insecticides are very important for control stored insects because they are active against a variety of insects, fast penetrating and non-toxic residues in the treated products. The EOs have some properties that made them suitable for insect management. Therefore, EcoSMART technologies in the USA introduced some insecticides based on EOs. In addition, some qualified products were used to control insect pests in urban pest management, greenhouse, horticultural crops and tree fruits. In contrast, some problems (e.g., volatility, solubility and oxidation) of essential oil-based insecticides were recorded, which plays an important role in the EOs activity, application and persistent. For this reason, new formulations with nanotechnology “Nanoformulation” such as the merging of the EOs in controlled release through nanocapsule formulations can re-solve these problems and offer numerous advantages. So, encapsulating the EOs has a considerable perspective as commercial insecticide products. Finally, EOs-based insecticides are low toxic, environmental persistence and eco-friendly. Therefore, they are not having worker re-entry and harvest restrictions after treating crops. So, they are compatible with biological control programs and indigenous natural enemies of pests.

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