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Essential oils as green pesticides of stored grain insects

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ABSTRACT: Essential oils are naturally occurring phytochemicals produced as secondary metabolite in plants. These are complex mixtures of volatile compounds and generally contain twenty to sixty individual compounds in different concentrations. They are lipophilic in nature and have density lower than water. These interfere with basic metabolic, biochemical, physiological and behavioral functions of insects. Several essential oils and its constituents have been established for their repellent, antifeedant, ovicidal, oviposition inhibitory and developmental inhibitory activities in insects. These insecticides probably interfere with the respiratory and nervous system of the insect to exert its actions. These essential oils provide an alternative source of insect control agents because they contain a range of bioactive chemicals, most of which are selective and have little or no harmful effect on the environment and the non-target organisms including human. Essential oils based formulations can be used as alternative tools in stored-grain insect management.

Keywords: Essential oils; Terpenes; Stored grain insects; Antifeedants; Insect growth regulators.

1. INTRODUCTION

Plant derived essential oils also known as volatile or ethereal oils are mixtures of odorous and volatile compounds. These are natural complex secondary metabolites characterized by a strong odour. These have generally lower density than that of water [1]. About 10% of the plant species are known to contain essential oils. There are 17,500 aromatic plant species among higher plants and approximately 3,000 essential oils are known out of which 300 oils are commercially used for pharmaceuticals, cosmetics and perfume industries apart from pesticidal application [1, 2]. Genera capable of producing essential oils are distributed in a limited number of families such as Apiaceae, Asteraceae, Compositae, Cupressaceae, Labiatae, Lauraceae, Myrtaceae, Piperaceae, Poaceae, Rutaceae and Zingiberaceae (Table 1). Essential oils are extracted from leaves, flowers, peels, seeds, wood, berries, resins, rhizomes and roots.

Essential oils are produced and accumulated in specialized structures as they are toxic to cells. There are several specialized secretory structures like oil cells, oil glands, ducts and trichomes that are discretely distributed within the plants from flowers to roots. According to Gottlieb and Salatino, essential oil production and secretory structures formation are closely connected [3]. For example, oil globules within membrane of secretory cell of rhizome in *Zingiber officinale*, peltate gland on leaves of *Lippia scaberrima* and secretory cavity in *Citrus* peel are responsible for biosynthesis and storage of essential oils [4-6]. Endogenous factors like development stage of whole plant and specific organs and exogenous factors both biotic and abiotic can alter essential oil production [7-9]. Sangwan et al. have indicated that ontogeny, photosynthetic rate, photoperiod, light quality, climatic and seasonal changes, nutrition, humidity, salinity, temperature, soil nature, storage structures and growth regulators are the factors that affect the production of essential oils both quantitatively and qualitatively [7].

Table 1. Some common essential oil producing plants.

Botanical name	Family
Acorus calamus	Acoraceae
Cananga odorata	Anonaceae
Anethum graveolens	Apiaceae
Angelica officinalis	
Apium graveolens	
Carum carvi	
Coriabdrum sativum	
Cuminum cyminum	
Ferula galbaniflua	
Foeniculum vulgare	
Levisticum officinale	
Petroselinum sativum	
Pimpinella anisum	
Trachyspermum ammi	
Acorus calamus	Araceae
Betula pendula	Betulaceae
Adnsonia digitata	Bombacaceae
Boswellia carteri	Burseraceae
Canarium luzonicum	
Commiphora myrrha	
Cassia fistula	Caesalpiniaceae
Abelia floribunda	Caprifoliaceae
Artemisia annua	Compositae
Artemisia dracunculus	
Calendula officinalis	
Chamaemelum nobile	
Chrysanthamum parthenium	
Tagetes erecta	
Brassica nigra	Cruciferae
Cochleria armoracia	
Cupressus sempervirens	Cupressaceae
Juniperus oxycedrus	
Thuja spp.	
Gaultheria procumbens	Ericaceae
Pelargonium spp.	Geraniaceae
Cymbopogon citratus	Graminae
Cymbopogon martini	
Cymbopogon nardus	

Botanical name	Family
Calamintha vulgaris	Labiatae
Hyssopus officinalis	
Lavandula augutifolia offinalis	
Lavandula spica	
Mellisa officinalis	
Mentha piperita	
Ocimum basilicum	
Ocimum sanctum	
Origanum majorana	
Origanum vulgare	
Orthodon punctulatum	
Pogostemon cablin	
Rosemarinus officinalis	
Salvia officinalis	
Satureja hortensis	
Thymus spp.	
Cinnamomum camphora	Lauraceae
Cinnamomum tamala	
Cinnamomum zeylanicum	
Laurus nibilis	
Sassafras albidum	
Myroxylon balsamum	Leguminosae
Myroxylon toluferum	
Trigonella foenum-graecum	
Allium cepa	Liliaceae
Allium sativum	
Illicium verum	Magnoliaceae
Acacia armata	Mimosaceae
Eucalyptus spp.	Myrtaceae
Eugenia earyophyllata	
Melaleuca alternifolia	
Melaleuca leucadendron	
Melaleuca viridiflora	
Myrtus communis	
Pimenta acris	
Pimenta officinalis	
Jasminum officinale	Oleaceae
Pandanus fasicularis	Pandanaceae
Piper nigrum	Piperaceae
Piper longum	
Piper cubeba	
Piper japonicum	
Rosa spp.	Rosaceae

Botanical name	Family
Aegle marmelos	Rutaceae
Citrus aurantifolia	
Citrus aurantium	
Citrus aurantium bergamia	
Citrus aurantium bigaradia	
Citrus aurantium sinensis	
Citrus limon	
Citrus reticulata	
Murraya koenigii	
Santalum album	Santalaceae
Cestrum nocturnum	Solanaceae
Stryrax benzoin	Stryraceae
Ageratum conyzoides	Umbelliferae
Daucus carota	
Lippia citriodora	Verbenaceae
Lantana camera	
Curcuma longa	Zingiberaceae
Elettaria cardamomum	
Zingiber officinale	

The total essential oil content of plants is generally very low and rarely exceeds 1%, but in some cases like clove (*Syzygium aromaticum*) and nutmeg (*Myristica fragrans*), it reaches up to 10% [10]. Due to presence of double bonds and functional groups like hydroxyl, aldehyde, ester in their molecular structures, essential oils are readily oxidizable by light, heat and air [11, 12]. These oils are stored as micro droplets in different glands of plants. After diffusing through glands, oil droplets spread over the surface of plant parts before evaporating and spreading in air. The most odoriferous plants are found in tropics where solar energy is greatest. There are numerous reports on differences of oil content and composition in aromatic plants due to seasonal variation. Microclimatic factors such as temperature, rainfall distribution and geographical features especially altitude also contribute to differences in chemotype of certain essential oil bearing plants. The type and nature of oil constituents and their individual concentration levels are important attributes particularly in terms of biological activities of the essential oils [13]. Vekiari et al. have reported seasonal variation in amounts of neryl acetate, geranyl acetate and citronellal in leaves and peel of certain lemon varieties [14]. Maximum values of compounds are obtained during spring season as compared to winter season [15].

2. CHEMICAL COMPOSITION OF ESSENTIAL OILS

Essential oils are highly complex mixtures of volatile compounds and may contain about 20-60 individual compounds in different concentrations. Each essential oil is characterized by two or three major components present in relatively high concentrations (20-70%) as compared to others components present in trace amounts. Generally, these major components determine the biological properties of essential oils. For example, carvacrol (30%) and thymol (27%) are the major components in *Origanum compactum* essential oil, linalol (68%) in *Coriandrum sativum* essential oil, 1,8-cineole (50%) in *Cinnamomum camphora* essential oil, phellandrene (36%) and limonene (31%) in *Anethum graveolens* leaf essential oil, carvone (58%) and limonene (37%) in *A. graveolens* seed essential oil, and menthol (59%) and menthone (19%) of *Mentha piperita* essential oil. The fragrance and chemical composition of essential oils can vary according to geoclimatic location and growing conditions (soil type, climate, altitude and amount of water available), season (before or after flowering), time of day when harvesting is achieved, genetic composition of the plant etc [7]. Therefore, all these factors influence the biochemical synthesis of essential oils in a given plant. Thus, same species of plant can produce a similar essential oil, however, with different chemical composition, resulting in different biological activities. Essential oil components can be classified into two groups, viz. volatile fraction and nonvolatile residue:

Volatile fractions: These constitute 90-95% of the oil. These include monoterpenes and sesquiterpenes as well as their oxygenated derivatives along with aliphatic aldehydes, alcohols and esters.

Nonvolatile residues: These comprise 1-10% of the oil. These include hydrocarbons, fatty acids, sterols, carotenoids, waxes and flavonoids.

Major volatile constituents are hydrocarbons (e.g. pinene, limonene, bisabolene), alcohols (e.g. linalol, santalol), acids (e.g. benzoic acid, geranic acid), aldehydes (e.g. citral), cyclic aldehydes (e.g. cuminal), ketones (e.g. camphor), lactones (e.g. bergaptene), phenols (e.g. eugenol), phenolic ethers (e.g. anethole), oxides (e.g. 1,8 cineole) and esters (e.g. geranyl acetate). Essential oil components can also be subdivided into two distinct groups of chemical constituents: hydrocarbons which are made up almost exclusively of terpenes (monoterpenes, sesquiterpenes, and diterpenes) and oxygenated compounds which are mainly esters, aldehydes, ketones, alcohols, phenols and oxides (Table 2).

Table 2. Major chemical components of plant derived essential oils.

Plant species	Main components	Reference
Commiphora molmol	Lindestrene, furanoeudesma-1,3-diene, Furanoeudesma-1,4-diene-6-one	[16]
Tagetes minuta	β-Phelandrene, Limonene, β-Ocimene, Dihydrotagetone, Tagetone, cis-Tagetenone, trans-Tagetenone	[17]
Cinnamomum zeylanicum	δ-Cadinene, γ-Cadinene, β-Caryophyllene	[18]
Commiphora guidottii	(E)-β-Ocimene	[19]
Boswellia rivae	Limonene	[19]
Boswellia pirotta	trans-Verbenol,Terpinen-4-ol	[19]
Boswellia neglecta	α-Thujene, α-Pinene, Terpinen-4-ol	[19]
Canarium luzonicum	α-Amyrin, β-Amyrin	[20]
Commiphora africana	Bisabolol, β-Sesquiphellandrene, α-Oxobisabolene, γ-Bisabolene	[21, 22]
Protium pilosum	α-Pinene, p-Cymene, α-Phellandrene	[23]
Commiphora myrrha	Isofuranogermacrene, Lindestrene, Furanoeudesma-1,3-diene, Furanodiene, Curzerene, Germacrone	[19, 24, 25]
Protium heptaphyllum	Terpinolene, β-Elemene, β-Caryophyllene, α-Pinene, Limonene, α-Phellandrene, β-Elemene	[23, 26, 27]
Ocimum basilicum	1,8-Cineole, Linalool, Estragole, *α*-Terpineol, *α*-Bergamotene	[28]
Laurus nobilis	1,8-Cineole, Linalool, *α*-Terpinyl acetate, Methyl eugenol, Eugenol	[28]
Coriandrum sativum	Linalool, Geraniol, *α*-Pinene	[28]
Myristica fragrans	Sabinene, *α*-Pinene, *β*-Pinene, Terpinen-4-ol, Safrole	[28]
Piper nigrum	Caryophyllene, Sabinene, Limonene, Germacrene B, *α*-Pinene, *α*-Humulene	[28]
Mentha piperita	Neomenthol, Isomenthone, 1,8-Cineole, Menth-8-ene, Neoisomenthol	[28]
Marjorana hortensis	Terpinen-4-ol, *γ*-Terpinene, *α*-Terpinene, Sabinene, *α*-Terpineol	[28]
Foeniculum vulgare	trans-Anethole, Fenchone, Estragole,	[28]
Canarium album	β-Pinene, α-Terpinene, γ-Terpinene, Terpinen-4-ol	[29]

Plant species	Main components	Reference
Thymus vulgaris	α-Pinene, p-Cymene, Limonene, γ-Terpinene, cis-Sabinene hydrate, Linalool, Terpinen-4-ol	[30]
Salvia officinalis	Camphor, 1,8-Cineole, α-Pinene, β-Pinene Camphene, α-Terpinyl acetate	[30]
Syzygium aromaticum	Eugenol, β-Caryophyllene, α-Humulene	[30]
Rosmarinus officinalis	α-Pinene, Camphene, 1,8-Cineole, Camphor	[30]
Cuminum cyminum	β-Pinene, p-Cymene, γ-Terpinene, Cuminal, 2-Caren-10-al	[30]
Origanum vulgare	Carvacrol, p-Cymene, Terpinolene	[30]
Artemisia haussknechtii	Camphor, 1,8-Cineole, cis-Davanone, 4-Terpineol, Linalool, β-Fenchyl alcohol, Borneol	[31]
Laurus nobilis	1.8-Cineole, Sabinene, α-Terpinyl acetate, α-Pinene, α-Phellandrene, trans-b-Osimen	[32]
Boswellia carterii	Isoincensole, Verticilla-4(20),7,11-triene, Isoincensole acetate	[33]
Boswellia papyrifera	Isoincensole, Isoincensole acetate, n-Octanol, n-Octyl acetate	[33]
Bursera graveolens	Limonene, α-Terpineol	[34]
Bursera simaruba	Limonene, β-Caryophyllene, α-Humulene, Germacrene	[35]
Boswellia serrata	Isoincensole, Isoincensole acetate, α-Thujene, α-Pinene, α-Thujene	[33, 36-38]
Trachyspermum ammi	Thymol, γ-Terpinene, p-Cymene, β-Pinene	[39]
Dacryodes edulis	Sabinene, Terpinene-4-ol, α-Pinene, p-Cymene, Myrcene, β-Caryophyllene, α-Thujene, α-Phellandrene, β-Pinene,	[40-42]
Boswellia sacra	E-β-Ocimene, Limonene, E-Caryophyllene	[43]
Boswellia ameero	(E)-2,3-Epoxycarene, 1,5-Isopropyl-2-methylbicyclo[3.1.0]hex-3-en-2-ol, α-Cymene, (3E,5E)-2,6-dimethyl-1,3,5,7-octatetraene, 1-(2,4-Dimethylphenyl) ethanol, 3,4-Dimethylstyrene, α-Campholenal, α-Terpineol	[44]
Coriandrum sativum	Linalool, trans-Anethol, c-Terpinene, Geranyl acetate	[45]
Artemisia absinthium	β-Pinene, β-Thujone	[46]
Fragaria vesca	Myrtenol, Citronellol, Linalool, Nonanal	[47]
Bursera microphylla	Caryophyllene, Myrcene	[48]
Commiphora habessinica	β-Elemene, α-Selinene, Cadina-1,4-diene, Germacrene B, α-Copaene, t-Muurolol, Caryophyllene oxide, α-Cadinol	[49]
Bunium persicum	p-Cuminaldehyde, γ-Terpinen-7-al, p-Cymene, γ-Terpinene	[50]
Cryptomeria japonica	A-Terpinene, γ-Terpinene, p-Cymine, 3-Carene, Terpinolene, β-Myrcene	[51]
Cinnamomum aromaticum	Cis-Cinnamaldehyde, Eugenol, α-Cadinol, Caryophyllene, Ledol	[52]
Haplopappus foliosus	Limonene, Bornyl acetate, Terpineol, p-Cymene, Agarospirol, α-Muurolene, δ-Cadinine, Caryophyllene	[53]
Thymus vulgaris	p-Cymene, γ-Terpinene, Thymol, Carvacrol, E-β-Caryophyllene, Germacrene D, β-Bisabolene	[54]
Santiria trimera	α-Humulene, β-Caryophyllene, α-Pinene, α-Terpineol, α-Pinene, β-Pinene	[55, 56]
Canarium schweinfurthii	Octyl acetate, Nerolidol, p-Cymene, Limonene, α-Terpineol	[57]
Artemisia gorgonum	Camphor, Chrysanthenone, Lavandulyl-*Z*-methylbutanoate, α-Phellandrene, Camphene, p-*C*ymene	[58]
Bahia ambrosoides	Lomonene, α-Pinene, Germacrene D, Sabinene, δ-Thujene, γ-Curcumene, α-Bergamotene	[53]
Carum carvi	(R)-Carvone, D-Limonene, α-Pinene, cis-Carveol	[59]
Cuminum cyminum	Caryophyllene oxide, α-Pinene, Geranyl acetate, α-Caryophyllene	[60]
Prangos acaulis	δ-3-Carene, α-Terpinolene, α-Pinene, Limonene	[61]
Foeniculum vulare	Methyl chavicol, α-Phellandrene, Fenchone	[62]
Carum copticum	Thymol, Terpinolene,o-Cymene	[63]
Zanthoxylum monophyllum	Sabinene, 1,8-Cineole, cis-4-Thujanol	[64]

Plant species	Main components	Reference
Zanthoxylum rhoifolium	β-Myrcene, β- Phellandrene, Germacrene D	[64]
Zanthoxylum fagara	Germacrene D-4-ol, Elemol, α-Cadinol	[64]
Artemisia annua	Camphor, 1,8-Cineole, Linalool, β-Caryophyllene, (E)-β-Farnese, Germacrene D	[64]
Schinus molle	α-Pinene, β-Pinene, Limonene, α-Ocimene, Germacrene D, γ-Cadinene, δ-Cadinene, Epi-bicyclosesquiphelandrene	[65]
Myristica fragrans	α-Pinene, Sabinene, β-Pinene, Myrcene, Limonene, Terpine-4-ol, Safrole, Myristicin	[66]
Boswellia socotrana	(E)-2,3-Epoxycarene, 1,5-Isopropyl-2-methylbicyclo[3.1.0]hex-3-en-2-ol, α-Cymene, (3E,5E)-2,6-dimethyl-1,3,5,7-octatetraene, 1-(2,4-Dimethylphenyl)ethanol,3,4-Dimethylstyrene, α-Campholenal, α-Terpineol, p-Cymene, 2-Hydroxy-5-methoxyacetophenone, Camphor	[44, 67]
Ammi visnaga	Isobutyrate, 2,2-Dimethylbutanoic acid, Croweacin, Linalool	[68]
Angelica dahurica	3-Carene, β-Elemene, β-Terpinene, β-Myrcene	[69]
Eucalyptus bicostata, E. cinerea, E. maidenii, E. odorata, E. sideroxylon, E. astringens, E. lahmannii	α-Pinene, p-Cymene 1.8-Cineole, Limonene, β-Eudesmol, α-Eudesmol, α-Terpineol, Pinocarveol, γ-Terpinene, Globulol	[70]
Smyrnium rotundifolium	Myrcene, Furanodiene, Germacrone, α-Selinene	[71]
Artemesia judaica	Piperitone, Camphor, Ethyl-cinnamate	[72]
Cupressus sempervirens	α-Pinene, δ-3-Carene, Limonene, α-Terpinolene	[73]
Angelica sylvestris	β-Phellandrene, α-Pinene, Myrcene, Germacrene D	[71]
Apium graveolens	(Z)-3-Butylidenephthalide, 3-Butyl-4,5-dihydrophthalide, α-Thujene	[74]
Azorella cryptantha	α-Pinene, α-Thujene, Sabinene, δ-Cadinene	[75]
Thamnosciadium junceum	Limonene,, cis-Ocimene, Terpinolene, trans-Isomirticisin	[71]
Protium icicariba	p-Cymene, α-Pinene, α-Terpinolene, Limonene, α-Copaene, γ-Elemene, δ-Cadinene	[76]
Cedrelopsis grevei	(E)-β-Farnesene, δ-Cadinene, α-Copaene, β-Elemene,	[77]
Pimpinella anisum	Camphène, Limonene, Fenchone, 4-Allylanisole, Anethole	[78]
Azadirachta indica	β-Elemene, γ- Elemene, Germacrene D, β-Caryophyllene, Bicyclogermacrene, Pentacosane, Tetracosane, β-Germacrene, Dodecene octadecanol, Verdiflorol, Farnesol, α–Terpineol	[79]
Protium crassipetalum	α-Copaene, Spathulenol, trans-Caryophyllene	[76]
Aucoumea klaineana	α-Pinene, β-Pinene, α-Phellandrene, β-Phellandrene, p-Cymene, 1,8-Cineole, p-Acetyl anisole, 3-Carene, p-Cymene, Limonene, Terpinolene, Terpineol, 3-Carene, α-Terpineol, Eucalyptol	[57, 80]
Schinus terebinthifolius	δ-3-Carene, Limonene, α-Phellandrene, α-Pinene	[81]
Parthenium ysterophorus	Germacrene-D, trans-β-Ocimene, β-Myrcene	[82]
Ambrosia polystachya	Germacrene-D, trans-β-Ocimene, β-Caryophyllene	[82]
Inula viscose	E-Foreseen Epoxide, Nerolidol B	[83]
Pelargonium graveolens	Citronellol, Geraniol, Caryophyllene oxide, Menthone, Linalool, β-Bourbonene, Iso-Menthone, Geranyl formate	[84]
Melissa officinalis	Neral, Geranial, Citronellal	[85]
Origanum vulgare	Thymol, γ-Terpinene, Carvacrol, Carvacrol methyl ether, cis-α-Bisabolene, Eucalyptol, p-Cymene, Elemol	[86]
Ruta graveolens	2-Nonanone, Undecanal, 2-Acetoxydodecane, 2-Decanone	[87]
Tithonia diversifolia	α-Pinene,(E,E)-α-Farnesene, β-Caryophyllene	[88]
Houttuynia cordata	α-Myrcene, 2-Undecanone, (Z)-β-Ocimene	[88]
Asarum glabrum	Safrole, Apiole	[88]
Hedychium forrestii	β-Pinene, β-Linalool, 1,8-Cineole, 4-Terpineol	[89]

Terpenes: Majority of essential oil components are terpenes. These contain hydrogen and carbon only. These are made of one or more 5-C unit, isoprene. These are classified into hemiterpenes (C_5), monoterpenes (C_{10}), sesquiterpenes (C_{15}), diterpenes (C_{20}), triterpenes (C_{30}) and tetraterpenes (C40). These hydrocarbons may be acyclic, alicyclic (monocyclic, bicyclic or tricyclic) or aromatic.

$$CH_2 = C(CH_3) - CH - CH_3$$

Isoperene unit

The monoterpenes (C_{10}) are formed by coupling of two isoprene units (C_5). They are the most representative molecules constituting 90% of essential oils. These have a great variety of structures. Limonene, myrcene, p-menthane, sabinene, cymene, phellandrene, thujane, fenchane, farnesene, azulene and cadinene are examples of this family (Fig. 1).

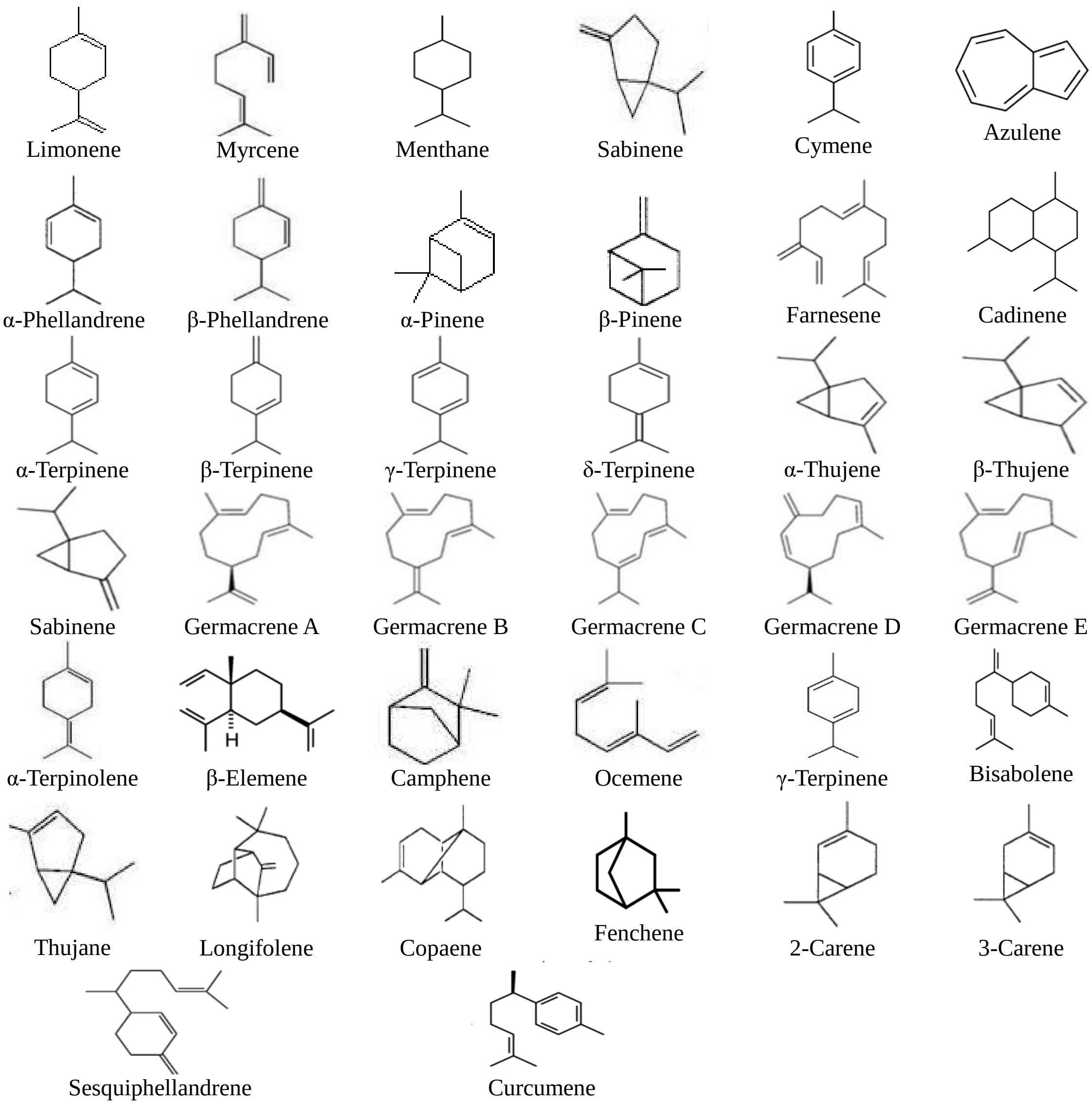

Figure 1. Common terpenes of essential oils.

Esters: These are sweet smelling and give a pleasant smell to the oils. These are formed by reaction of alcohol with acid. Esters are very common and are found in a large number of essential oils. These include linalyl acetate, geraniol acetate, eugenol acetate, bornyl acetate, citronellyl acetate, neryl acetate etc (Fig. 2).

Oxides: Oxides or cyclic ethers are the strongest odorants. The well known oxide is 1,8-cineole as it is the most omnipresent constituent in essential oils. Other examples of oxides are bisabolone oxide, linalool oxide, sclareol oxide, ascaridole, caryophyllene oxide, cis-piperitone oxide, aromadendrene oxide and bisabolene oxide (Fig. 3).

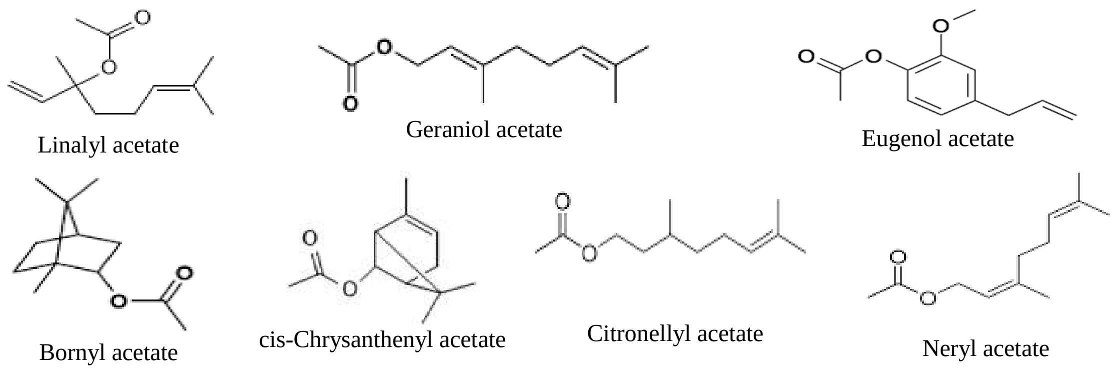

Figure 2. Common esters of essential oils.

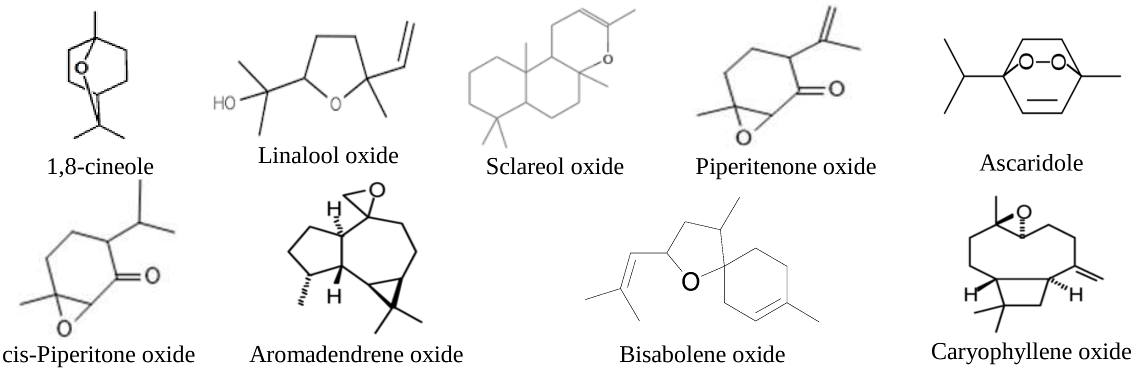

Figure 3. Common oxides of essential oils.

Lactones: These are of relatively high molecular weight components. Some common lactones are nepetalactone, bergaptene, costuslactone, dihydronepetalactone, alantolactone, citroptene and psoralen (Fig. 4).

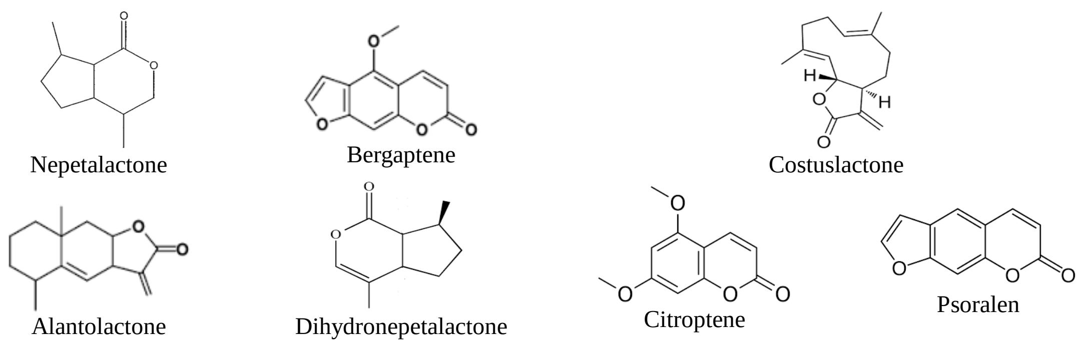

Figure 4. Common lactones of essential oils.

Alcohols: Linalol, menthol, borneol, santalol, nerol, citronellol, bisabolol, eucalyptol, trans-carveol, dihydrocarveol, α-cadinol, δ-cadoniol, τ-cadinol, elemeol, nerol and geraniol are common alcohols of essential oils (Fig. 5).

Phenols: These oxygen containing molecules are responsible for fragrance of oil. These aromatic components are among the most reactive components and are often found as crystals at room temperature. The most common phenols are thymol, eugenol, carvacrol, cumic alcohol, α-terpineol, safrole, verbenol and chavicol (Fig. 6).

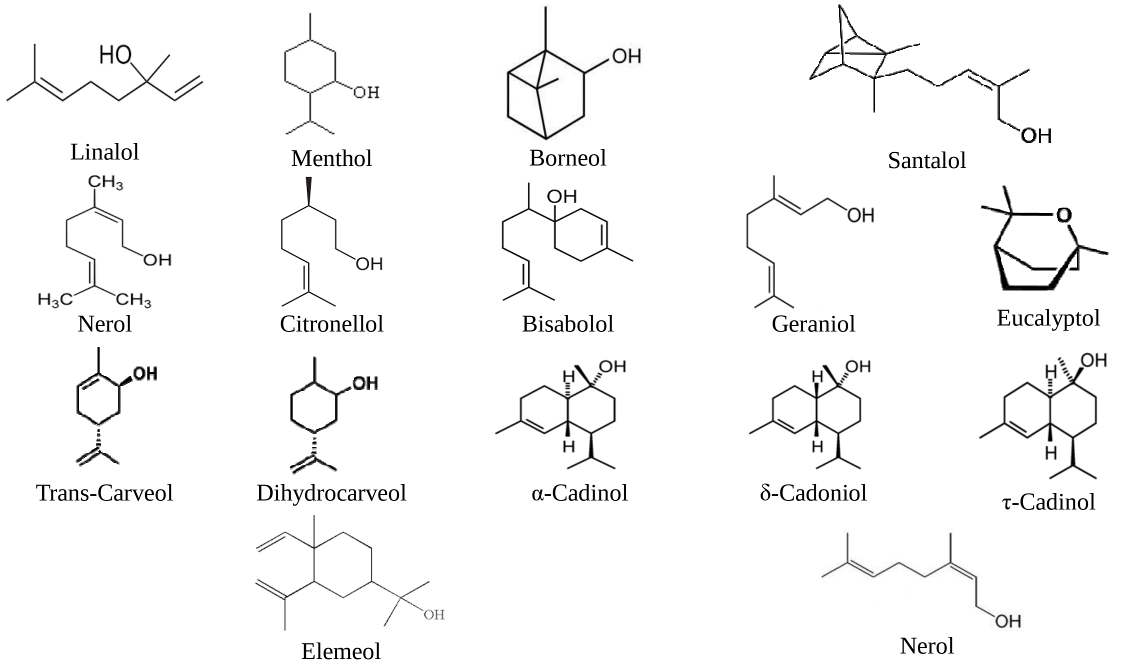

Figure 5. Common alcohols of essential oils.

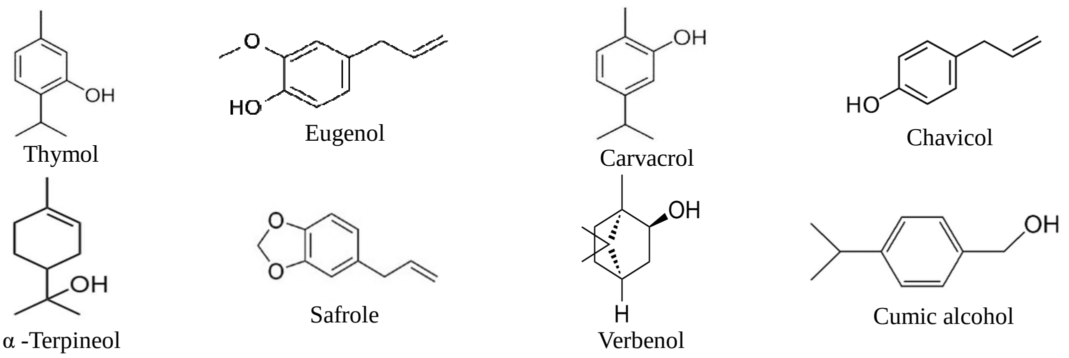

Figure 6. Common phenols of essential oils.

Aldehydes: These are highly reactive and characterized by -CHO group. These are common essential oil components that are unstable and oxidize easily. Geranial, neral, myrtenal, cuminaldehyde, citronellal, cinnamaldehyde, benzaldehyde and citral are common aldehydes of essential oils (Fig. 7).

Ketones: These are characterized by –C=O group. These are not very common in majority of essential oils. These are relatively stable molecules and are not important as fragrances or flavour substances. Carvone, menthone, pulegone, fenchone, camphor, jasmone, thujone, methyl nonyl ketone, pinocamphone and verbenone are common ketones of essential oils (Fig. 8).

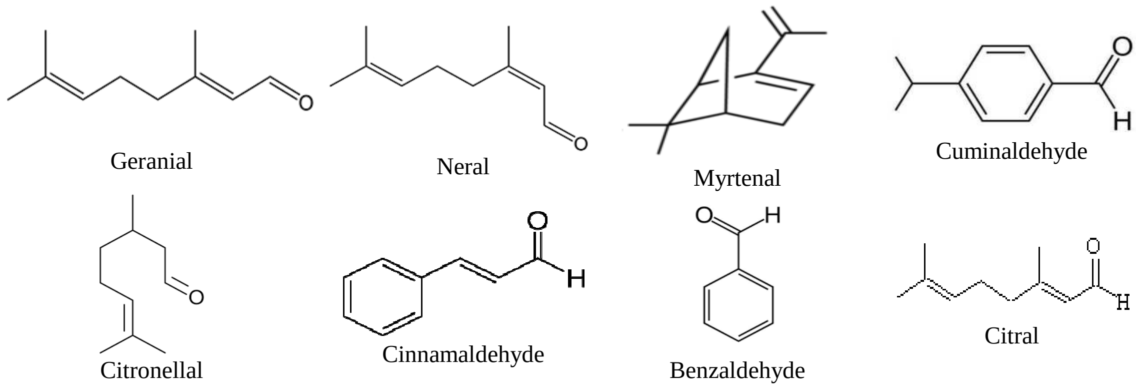

Figure 7. Common aldehydes of essential oils.

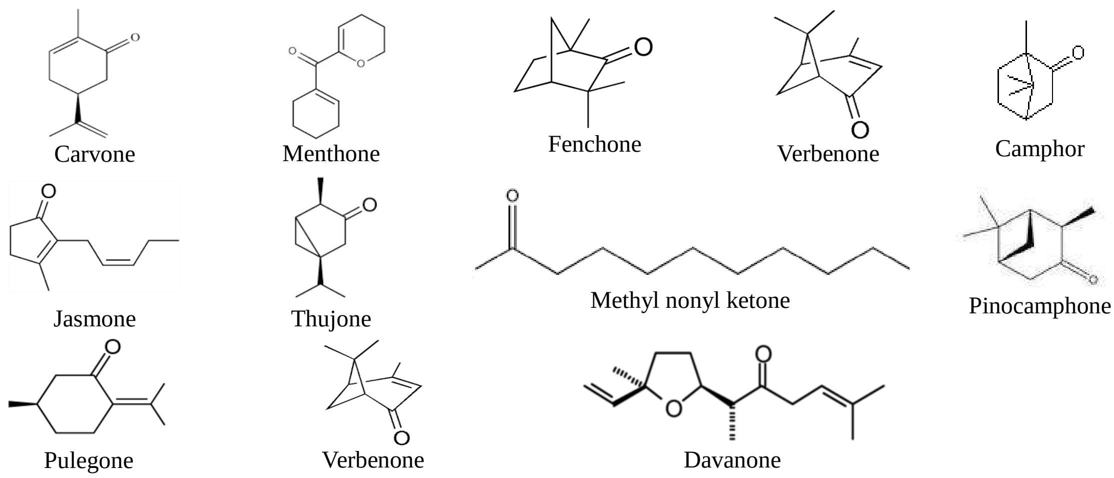

Figure 8. Common ketones of essential oils.

Physical properties of essential oils:

1. Essential oils are volatile and liquid at room temperature.
2. They are colourless or slightly yellowish.
3. They are less dense than water (sassafras and clove oils are exceptions).
4. They are always rotational and with a high refractory index.
5. They are soluble in alcohol and other organic solvents such as acetone, ether or chloroform.
6. They are lipid soluble and not very soluble in water.

3. BIOLOGICAL ACTIVITIES OF ESSENTIAL OILS AS INSECTICIDES

Conventional synthetic insecticides play important role in protecting stored grains from insect pest infestation in the 20th and 21st century. Use of these conventional insecticides is believed to be one of the major reasons for growth in agricultural productivity in the 20th century. However, more and more public concern all over the world about long-term health and environmental effects of uncontrolled and extensive use of synthetic insecticides increases. Many laboratory studies and clinical cases have shown that almost all the conventional synthetic insecticides have the potential to affect ecosystems adversely; and most of them have acute or chronic toxicity to humans or other non-target organisms. Keeping these problems in mind, there is an urgent demand to reduce use of conventional insecticides and develop alternatives with no or little harmful effects on the environment and with no toxicity to non-target organisms including human. From time immemorial with the beginning of human civilization, several plant parts have been used to protect stored grains from insect pests. Still several natural products such as nicotine from tobacco, pyrethrum from chrysanthemums, rotenone from Derris root, sabadilla from lilies, ryania from the ryania shrub, limonene from citrus peel and neem from the tropical neem tree have been used to protect grains from insect pest infestations in storage in several countries. In recent years, scientific communities have focussed attention towards volatile plant products as a replacement of the synthetic pesticides.

Essential oils are secondary metabolic products in plants whose functions are other than nutritions. These oils have strong aromatic components that give a plant its distinctive odour and flavour [90]. Essential oils are complex mixtures of a large number of constituents in variable ratios [91]. Their components and qualities vary with geographical distribution, harvesting time, growing conditions and extraction method [92]. These oils are liquid at room temperature and can be easily transformed from a liquid to a gaseous state at room temperature or a slightly higher temperature without decomposing [90]. Essential oils are very interesting natural plant products and they possess various biological properties. The term 'biological properties' include all activities that these volatile compounds exert on other organisms whether microbes, plants or animals. Knowledge of these essential oil producing plants, their chemistry and their biological properties is of prime importance not only to enable them to be utilized as natural pest control agents and replace commercial synthetic pesticides but also to enable us to understand the nature of their toxicity to non-target animals. Botanical insecticides degrade rapidly in air and moisture and are readily broken down by detoxification enzymes. This is very important because rapid breakdown suggests less persistence in the environment and reduced risks to non-target organisms. Plant derived essential oils are generally considered broad-spectrum and safe for the environment because variety of compounds they contain quickly biodegrade in soil.

Essential oils and their constituents are primarily lipophilic compounds that act as toxins, feeding deterrents, repellent, oviposition deterrents and even attractant to a wide variety of insect pests. Due to their volatility, essential oils have limited persistence under natural conditions. Recent findings suggest that plant derived volatiles are neurotoxic via octopaminergic mode of action [93]. Thus, these natural products are safe for human and other vertebrates due to lack of octopaminergic mode of nerve conduction.

3.1. Attractants

Presence of volatile essential oils and its components in plants provides an important defense strategy to plants especially against herbivorous insect pests. These plant derived essential oils also play a crucial role in plant-plant interactions and serve as attractants for some insects like pollinators. Attraction of insects by essential oils and its components have been studied. Petroski and Hammack [94] have reported that cinnamaldehyde, cinnamyl alcohol, 4-methoxy-cinnamaldehyde, geranylacetone and aterpineol attract adult corn rootworm beetles, *Diabrotica* sp. The cis-jasmone alone or in combination with linalool and/or phenylacetaldehyde has been reported to attract lepidopteran adults [95]. Geraniol and eugenol are effective attractants and are used to lure Japanese beetle, *Popillia japonica* [96]. Similarly, methyl-eugenol has been used to lure oriental fruit fly, *Dacus dorsalis* [96]. These attracted insects can then be killed by physical and/or chemical means. Katerinopoulos et al. [97] have demonstrated that 1,8-cineole from *Rosmarinus officinalis* attract grape berry moth, *Lobesia botrana* and flower thrips, *Frankliniella occidentalis*. Eugenol is a strong deterrent for most insect species, although in a few cases it can be an attractant [98]. They can act as internal messengers and as defensive substances against herbivores or as volatiles directing not only natural enemies to these herbivores but also attracting pollinating insects to their hosts [99]. Besides, attractive essential oil and its components can be used as bait in attracting parasitoids and predators for controlling their host insect species in biological insect pest management programme. These essential oils/components are useful for monitoring of these agricultural insect pests.

3.2. Repellents

Insect repellents are those substances that act locally or distally to deter insects. Several essential oils and their constituents are known to repel several insect species especially coleopterans (Table 3).

Table 3. Essential oils with repellent activity.

Plant species	Insect species	Reference
Carum copticum	*Callosobruchus maculatus*	[114]
Artemisia scoparia	*C. maculatus, T. castaneum, S. oryzae*	[115]
Melalecuca alternifolia	*C. maculatus, S. oryzae*	[116]
Cinnamomum zeylanicum	*C. maculatus, S. oryzae*	[116]
Syzygium aromaticum	*C. maculatus, S. oryzae*	[116]
Cymbopogon flexuosus	*C. maculatus, S. oryzae*	[116]
Thymus vulgaris	*C. maculatus, S. oryzae*	[116]
Eucalyptus globules	*C. maculatus, S. oryzae*	[116]
Simmondsia chinensis	*C. maculatus, S. oryzae*	[116]
Anethum graveolens	*T. castaneum*	[117, 118]
Nigella sativa	*T. castaneum*	[117, 118]
Trachyspermum ammi	*T. castaneum*	[117, 118]
Carum copticum	*T. castaneum, C. maculatus*	[119]
Perovskia abrotanoides	*T. castaneum*	[120]
Myristica fragrance	*T. castaneum*	[121]
Illicium verum	*T. castaneum*	[121]
Achillea wilhelmsii	*Plodia interpunctella*	[122]
Hyssopus officinalis	*P. interpunctella*	[122]
Zhumeria majdae	*P. interpunctella*	[122]
Drimys winteri	*T. castaneum*	[123]
Laurelia sempervirens	*T. castaneum*	[123]
Schinus molle	*Trogoderma granarium, T. castaneum, C. maculatus*	[124]
Carum carvi	*Meligethes aeneus*	[125]
Thymus vulgaris	*M. aeneus*	[125]
Piper cubeba	*T. castaneum*	[126]
Zingiber officinale	*T. castaneum*	[126]
Mentha longifolia	*C. maculates*	[127]
Thymus kotschyanus	*C. maculates*	[127]
Citrus reticulate	*C. maculatus, P. interpunctella*	[128]
Carum copticum	*Plutella xylostella, T. castaneum*	[129]
Perovskia abrotanoides	*P. xylostella, T. castaneum*	[129]
Artemisia judaica	*C. maculatus, P. interpunctella*	[72]
Achillea wilhelmsii	*C. maculatus, P. interpunctella*	[72]
Hyssopus officinalis	*C. maculatus, P. interpunctella*	[72]

Plant species	Insect species	Reference
Zhumeria majdae	*C. maculatus, P. interpunctella*	[72]
Piper cubeba	*T. castaneum, S. oryzae*	[130]
Zingiber officinale	*T. castaneum, S. oryzae*	[130]
Citrus aurantium	*T. castaneum*	[131]
Cinnamomum zeylanicum	*T. castaneum*	[131]
Gautheria fragrantissima	*T. castaneum*	[131]
Lavandula officinalis	*T. castaneum*	[131]
Oscimum sanctum	*T. castaneum*	[131]
Anethum graveolens	*S. oryzae*	[132]
Nigella sativa	*S. oryzae*	[132]
Trachyspermum ammi	*S. oryzae*	[132]
Citrus limonum	*Tenebrio molitor*	[133]
Litsea cubeba	*T. molitor*	[133]
Allium sativum	*S. oryzae*	[134]
Cinnamomum tamala	*S. oryzae*	[134]

Adhatoda vasica essential oil exhibited repellent activity against *Sitophilus oryzae* and *Bruchus chinensis* [100]. Ngoh et al. have investigated repellent activity of eugenol, isoeugenol, methyleugenol, safrole, isosafrole, α-pinene, limonene, 1,8-cineole and p-cymene against *Periplaneta americana* nymphs [101]. They proved that eugenol, methyl-eugenol, isoeugenol, safrole and isosafrole are better toxicants and repellents to insects than limonene, 1,8-cineole and p-cymene. Only α-pinene exhibited a considerable repellent effect on nymphs. Oils from *Ocimum suave* and *Lippia* repelled *S. zeamais* adults [102, 103]. *Acorus calamus* essential oil repelled *Tribolium castaneum* adults [31]. Essential oil from *Jupinerus communis* berries is a very good mosquito repellent [104]. Some essential oils repelled *S. granarius* and inhibited its feeding [105]. *Absinthium* essential oil exerted both toxic and repulsive effects on *S. granarius* [106]. Chahal et al. have reported that turmerone and dehydroturmerone, the major constituents of turmeric rhizome powder oil are strong repellents to stored grain pests [107]. Turmeric oil has been reported to provide protection to wheat grains against *T. castaneum* adults. Garcia et al. have shown repellent behaviour of *Baccharis salicifolia* essential oil against *T. castaneum* adults [108]. Essential oil isolated from *Tagetes terniflora* have been reported effective as repellent against fifth instar of *T. castaneum* [109]. Wang et al. have tested and established the repellent and fumigant activity of essential oil from *Artemisia vulgaris* against *T. castaneum* adults [110]. Repellent properties of *Eucalyptus* essential oil have also been well established. This oil presented high repellency against *Ixodes ricinus*, *Aedes albopictus* and *Pediculus humanus capitis* [111, 112]. *Triaenops persicus* oil has been reported for insecticidal activity against adults of *T. castaneum* and *S. oryzae* [90]. Garlic and mint essential oils have been evaluated against the cowpea aphid, *Aphis craccivora* and its absolute repellent activity was proved on adult aphids. Garlic oil had higher repellent than mint oil [113]. This repellent action of essential oils may be related to major constituents, e.g., piperitone, camphor and (E)-ethyl cinnnamate.

Essential oils from *Cuminum cyminum*, *Piper nigrum*, *Illicium verum*, *Myristica fragrans*, *Foeniculum vulgare*, *Trachyspermum ammi*, *A. graveolens* and *Nigella sativa* have been isolated and its repellent activity have been determined against *T. castaneum* adults [117, 118, 121]. Repellent activity of essential oils from *Afromomum melegueta* seeds and *Zingiber officinale* rhizomes has been evaluated against *R. dominica*. Both oils repelled adult beetles [135]. Lopez et al. have reported that *Coriandrum sativum* oil is very toxic to *S. oryzae*, *R. dominica* and *C. pusillus* while, fractions of camphor are highly toxic to *R. dominica* and *C. pusillus* [75]. Cosimi et al. have tested essential oils from *Laurus nobilis*, *Citrus bergamia* and *Lavandula hybrida* for repellent activities against *S. zeamais* and *Cryptolestes ferrugineus* adults [136]. *C. bergamia* essential oil exerted the highest repellency on *S. zeamais* adults. Repellent active compounds isolated from *Limnophila geoffrayi* and *Schizonepeta tenuifolia* are pulegone, linalool, eugenol, thymol and methyl chavicol [137]. While, other repellent compounds such as α-pinene, β-pinene, D-limonene, (E)-3,7-dimethyl-2,6-octadienal have been isolated from *Armoracia rusticana*, *Pimpinella anisum*, *Allium sativum*, *Laurelia sempervirens* and *Drimys winteri* [123, 138]. Kheradmand et al. have evaluated and established repellent activity of jojoba, *Simmondasia chinensis* seed oil on *Oryzaephilus surinamensis* and *C. maculatus* [139]. Kim et al. have evaluated repellent activity of origanum essential oil and its nine constituents against *T. castaneum* adults [140]. Among the nine constituents of origanum oil, caryophyllene oxide and α-pinene produced strong repellency. Thymol, carvacrol and myrcene which are hydrogenated monoterpenoids, have also shown strong repellency. Moderate and low repellency has been produced by terpinene and camphene. Abdel-Sattar et al. have shown that essential oils of *Schinus molle* (fruit and leaf) have repellent activity against *T. granarium* and *T. castaneum* [124]. They identified p-cymene as a major component in fruits and leaf oils. The repellent activity of essential oil obtained from leaves induced higher activity than that of fruit. Zapata and Smagghe have shown that essential oils from leaves and bark of *Laurelia sempervirens* and *Drimys winteri* have highly repellent activity against *T. castaneum* [123]. Liu et al. have reported contact toxicity of *Artemisia capillaris* and *A. mongolica* essential oils against *S. zeamais* [141]. Insecticidal activity may be due to main components, 1,8-cineole, germacrene D, α-pinene, germacrene D and γ-terpinene of *A. mongolic* essential oil. Pavela has evaluated ten essential oils for their repellent activity against *Meligethes aeneus* [125]. Oils isolated from *Carum carvi* and *Thymus vulgaris* have shown the highest repellent activity. Chaubey has evaluated *Z. officinale* and *Piper cubeba* essential oils for its repellent against *T. castaneum* [126]. *Z. officinale* and *P. cubeba* essential oils have repelled adults of *T. castaneum* significantly even at very low concentrations. Repellent activity of essential oils obtained from *Mentha longifolia* and *Thymus kotschyanus* has been evaluated against *C. maculatus* [127]. Ajayi and Olonisakin have studied and established repellent activities of *Syzgium aromaticum*, *Piper guineense* and *Xylopia aethiopica* essential oils against *T. castaneum* [142]. *Origanum vulgare* and *Thymus vulgaris* essential oils have been tested against *Nezara viridula* [143]. Khani and Asghari have evaluated and established insecticidal activity of *Pulicaria gnaphalodes* essential oil against *T. confusum* and *C. maculatus* [144]. Ben Jemba et al. have reported *Laurus nobilis* essential oils from Tunisia, Algeria and Morocco for their repellent and toxic activities against *R. dominica* and *T. castaneum*. *Artemisia judaica* essential oil has been reported specific against cowpea weevil, *C. maculatus* [145]. *A. judaica* essential oil has been reported for insecticidal activity against *C. maculatus* [72]. Their toxic effect has been attributed to piperitone, camphor and (E)-ethyl cinnnamate. These compounds are monoterpenoids and lipophilic. These have fast penetration properties into insects which consequently interfere with biochemical and physiological functions [146]. Abd-Elhady has reported *A. judaica* essential oil for its repellent activity against cowpea weevil, *C. maculatus* [72]. It reduced egg laying in *C. maculatus*. Essential oil of *A. sativum* has been isolated and evaluated for its repellent activities against *T. castaneum* and *S. oryzae*. *A. sativum* essential oil repelled *T. castaneum* and *S. oryzae* adults at very low concentration [134, 147]. Essential oils from *Citrus limonum* and *Litsea cubeba* have been reported for repellent activity against adults of *Tenebrio molitor* [133]. Repellent activity of these essential oils may be due to the presence of D-limonene and 3,7-dimethyl-6-octenal in *C. limonum* essential oil and

(E)-3,7-dimethyl-2,6-octadienal and (E)-cinnamaldehyde in *L. cubeba*.

3.3. Antifeedants

Antifeedants may be defined as substances that deter from feeding when come in contact made with insects. Plant derived essential oils and its compounds have been known to exhibit antifeedant properties against a number of insect pest species (Table 4). Paruch et al. have reported a terpenoid lactone exhibiting antifeeding activity against *S. granarium, T. granarium* and *T. confusum* [148]. Oils isolated from *Curcuma longa* and *Z. officinale* have been found effective as antifeedant and insect growth regulators [149]. Antifeedant activity of 1,8-cineole has been demonstrated against *T. castaneum* [150]. Tripathi et al. have reported feeding deterrence activity of *C. longa* leaf essential oil against adult and larvae of *R. domestica*, *S. oryzae* and *T. castaneum* which has been attributed to the presence of monoterpenes, carvone and dihydrocarvone [151].

Table 4. Essential oils with antifeedant activity.

Plant species	Insect species	Reference
Piper cubeba	*T. castaneum, S.oryzae*	[132]
Zingiber officinale	*T. castaneum, S.oryzae*	[132]
Allium sativum	*S. oryzae*	[134]
Cinnamomum tamala	*S. oryzae*	[134]
Curcuma longa	*R. dominica, S. oryzae, T. castaneum*	[151]
Schinus molle	*S. oryzae*	[156]
Eucalyptus globulus	*T. castaneum*	[157]
Lavandula stoechas	*T. castaneum*	[157]

Tripathi et al. have evaluated repellent and antifeedant activities of *Curcuma* leaf oil and d-limonene on *R. dominina*, *S. oryzae* and *T. castaneum*. Huamg and Ho established antifeedant activity of cinnamaldehyde against *T. castaneum* and *S. zeamais* [151, 153]. Perillyl alcohol, cisverbenol, cis-carveol, geraniol, citronellal, perillaldehyde, caryophyllene oxide, carvacrol, 4-isopropylbenzenemethanol, thymol, 3-carene and myrcene have been reported the most effective repelling chemicals. The toxicity of essential oil of *Piper aduncum* has been tested against *Cerotoma tingomarianus*. This oil causes physiological problems and reduces foliar consumption by beetles [154]. Rana and Rashmi have shown antifeedant activity of *Vitex negundo* essential oil against *C. chinensis* and *S. oryzae* [155]. Benzi et al. have evaluated nutritional indices and feeding deterrent activities of essential oil from leaves and fruits of the Brazilian pepper tree, *Schinus molle* on *S. oryzae* adults [156]. Oils from both plant parts have been found to alter nutritional indices. Fruit essential oil has a strong feeding deterrent activity while leaf oil has a slight effect. Ebdadollahi [157] has studied antifeedant activity of *Eucalyptus globulus* and *Lavandula stoechas* essential oils against *T. castaneum*. All the tested essential oils cause reductions in feeding of insects. Chaubey has evaluated antifeeding activities of essential oils from *Zingiber officinale* rhizomes and *Piper cubeba* berries as well as pure compounds, α-pinene and β-caryophyllene against *T. castaneum* and *S. oryzae* [132]. β-Caryophyllene has been shown highest toxicity followed by *P. cubeba*, *Z. officinale* and α-pinene against both insects. *S. oryzae* is more sensitive than *T. castaneum* to both essential oils and pure compounds. Feeding deterrency is maximum in both insects by *P. cubeba* essential oil followed by *Z. officinale* essential oil, β-caryophyllene and α-pinene. *Allium sativum* essential oil exhibits antifeedant activities against *T. castaneum* and *S. oryzae*

[134, 147]. The antifeedant activity of essential oils can be due to its major constituents. Moreover, the minor constituents of essential oils play an important role in changing the activity by synergistic effects. In general, the mixture of chemical constituents is more effective than that of individual pure compounds. Therefore, synergistic effects between essential oils components are playing an essential role in essential oils activity [158]. Exploration of the influence of chemical complexity of essential oils on feeding behaviour of insects can help in the development of new crop protection products for use in integrated pest management. However, all the products need to be tested for their effects on non-target organisms and their environmental impact and future. Understanding the role of each constituent in the efficacy of oil provides an opportunity to create artificial blends of different constituents on the basis of their activity and efficacy against different pests.

3.4. Ovicidal and oviposition deterrants

Essential oils are not only active against adults and larvae but also inhibit reproduction and egg hatching (Table 5). This action could be the result of female sensitivity resulting in reduction in fecundity. The inhibition of reproduction of *Acanthoscelides obtectus* by essential oils belonging to Labiatae, Umbelliferae, Lauraceae, Myristicaceae, Graminae, Rutacae, Myrtacae families has been observed. This beetle has been shown to be a convenient model to point out with accuracy which reproductive stage is targeted and speed of the activity of essential oils [159].

Table 5. Essential oils with ovicidal activity.

Plant species	Insect species	Reference
Allium sativum	*T. castaneum*	[166]
Myristica fragrance	*T. castaneum*	[167]
Cuminum cyminum	*T. confusum*	[168]
Cuminum cyminum	*E. kuehniella*	[168]
Pimpinella anisum	*T. confusum*	[168]
Pimpinella anisum	*E. kuehniella*	[168]
Ammi visnaga	*Mayetiola destructor*	[169]
Carum carvi	*Trialeurodes vaporariorum*	[170]
Anethum graveolens	*C. chinensis*	[163]
Cuminum cyminum	*C. chinensis*	[163]
Illicium verum	*C. chinensis*	[163]
Myristica fragrans	*C. chinensis*	[163]
Nigella sativa	*C. chinensis*	[163]
Piper nigrum	*C. chinensis*	[163]
Trachyspermum ammi	*C. chinensis*	[163]
Elletaria cardamomum	*T. castaneum*	[171]
Cinnamomum zeylanicum	*T. castaneum*	[171]
Syzygium aromaticum	*T. castaneum*	[171]
Eucalyptus spp.	*T. castaneum*	[171]
Azadirecta indica	*T. castaneum*	[171]
Piper cubeba	*C. chinensis*	[147]
Zingiber officinale	*C. chinensis*	[147]
Allium sativum	*C. chinensis*	[172]

Citrus peel oil causes a high reduction in oviposition of *C. maculatus* [158, 160]. *Acorus calamus* oil reduces oviposition in *C. maculatus* [161]. Carvone completely suppresses egg hatching of *T. castaneum* [151]. Brito et al. have studied the effects of *Eucalyptus citriodora, E. globulus* and *E. staigerana* essential oils on oviposition and number of emerged insects of *Zabrotes subfasciatus* and *C. maculatus* [162]. These essential oils reduce percentage of viable eggs and emerged insects of both coleopterous species. *A. graveolens*, *C. cyminum*, *I. verum*, *M. fragrans*, *N. sativa*, *P. nigrum* and *T. ammi* oils have been evaluated for oviposition inhibitory activities against *C. chinensis*. These essential oils reduce oviposition potential of the insect when fumigated with sublethal concentrations [163]. Waliwitiya et al. have evaluated and established oviposition deterrent activities of eugenol, citronellal, thymol, pulegone and cymene. [164] Nondenot et al. have tested essential oils of *Ageratum conyzoides*, *Citrus aurantifolia* and *Melaleuca quinquenervia* on *C. maculatus* [165]. These oils show insecticidal activity and reduce egg lying capacity in *C. maculatus*. Ajayi and Olonisakin have been evaluated ovicidal activity of *Syzgium aromaticum*, *Piper guineense* and *Xylopia aethiopica* essential oils against *T. castaneum* [142]. The three essential oils are able to reduce progeny emergence of *T. castaneum*. Higher number of adults emerged in *X. aethiopica* than in *S. aromaticum* and *Piper guineense*. Chaubey has shown oviposition inhibitory activity of α-pinene and β-caryophylene alone or in binary combination against *T. castaneum* by fumigation method [132]. Fumigation of *T. castaneum* adults with sublethal concentrations of α-pinene, β-caryophyllene and its binary combination reduce oviposition potential of insect. This study indicates that α-pinene and β-caryophylene in binary combination show synergism and reduce egg laying capacity in *T. castaneum*. *A. sativum essential* oil has been evaluated for oviposition inhibitory activities against *S. oryzae*. Exposure of *S. oryzae* adults to sublethal concentrations of *A. sativum* oil inhibits oviposition [134]. In all cases, essential oils and their components have strong effects on egg, oviposition and egg hatching of insect pests. Essential oils have been reported to have low vapour density than fatty oils; hence, they are readily volatilized. This could be the reason why most of the eggs that might have hatched could not survive the volatility effects of the essential oils especially as the concentration/dosage of oils increased. Results clearly indicate variations in the activity of essential oils regarding the stage of the insect, species of insect and plant origin of essential oil.

3.5. Toxicants

Several essential oils and their components have been evaluated for their toxic nature against diverse group of insect pests, and most of them have shown promising toxicity either by its fumigant or by contact action [117, 118, 121, 123, 124, 126, 130, 132, 134, 147, 151, 173-175] (Table 6).

Table 6. Essential oils toxic to stored grain insect pests.

Plant species	Insecticidal activity and tested insect	Reference
Anethum sowa	Fumigant activity against *T. castaneum*	[181]
Artemisia annua	Fumigant activity against *T. castaneum*	[182]
Elletaria cardomum	Fumigant activity against *T. castaneum*	[184]
Apium graveolens	Fumigant toxicity against *Acanthoscelides obtectus*	[192]
Foeniculum vulare	Fumigant toxicity against adults of *T. castaneum*	[193]
Pimpinella anisum	Fumigant toxicity against adults of *T. castaneum*	[193]
Foeniculum vulare	Contact and fumigant toxicity against adults of *Lasioderma serricorne*	[194]
Cnidium officinale	Contact and fumigant toxicity against adults of *L. serricorne*	[194]
Foeniculum vulare	Adulticidal on *S. oryzae* and *C. chinensis*	[195]

Plant species	Insecticidal activity and tested insect	Reference
Angelica dahurica	Contact and fumigant toxicity against adults of *L. serricorne*, *S. oryzae* and *C. chinensis*	[194, 195]
Carum copticum	Contact and fumigant toxicity against *S. oryzae* and *T. castaneum*	[196]
Trachyspermum ammi	Fumigant toxicity against *T. castaneum*	[117]
Anethum graveolens	Fumigant toxicity against *T. castaneum*	[117]
Nigella sativa	Fumigant toxicity against *T. castaneum*	[117]
Anethum graveolens	Fumigant toxicity against *C. chinensis*	[163]
Cuminum cyminum	Fumigant toxicity against adults of *C. chinensis*	[163]
Illicium verum	Fumigant toxicity against adults of *C. chinensis*	[163]
Myristica frangrans	Fumigant toxicity against adults of *C. chinensis*	[163]
Nigella sativa	Fumigant toxicity against adults of *C. chinensis*	[163]
Piper nigrum	Fumigant toxicity against adults of *C. chinensis*	[163]
Trachyspermum ammi	Fumigant toxicity against adults of *C. chinensis*	[163]
Coriandrum sativum	Fumigant toxicity against adults of *S. oryzae*, *R. dominica* and *Cryptolestes pusillus*	[75]
Coriandrum sativum	Toxicity against adults of *S. granarius*	[197]
Heracleum persicum	Fumigant toxicity on adults of *C. maculatus*	[198]
Prangos acaulis	Adulticidal and larvicidal against *C. maculatus*	[199]
Cinnamomum zetlanicum	Fumigant toxicity against *C. maculatus*, *S. oryzae* adults	[116]
Syzygium aromaticum	Fumigant toxicity against *C. maculatus*, *S. oryzae* adults	[116]
Cymbopogon flexuosus	Fumigant toxicity against *C. maculatus*, *S. oryzae* adults	[116]
Thymus vulgaris	Fumigant toxicity against *C. maculatus*, *S. oryzae* adults	[116]
Eucalyptus globules	Fumigant toxicity against *C. maculatus*, *S. oryzae* adults	[116]
Simmondsia chinensis	Fumigant toxicity against *C. maculatus*, *S. oryzae* adults	[116]
Trachyspermum ammi	Fumigant toxicity against adults of *S. oryzae*	[200]
Piper nigrum	Fumigant toxicity against adults of *S. oryzae*	[200]
Cuminum cyminum	Fumigant toxicity against adults of *S. oryzae*	[200]
Azilia eryngioides	Fumigant toxicity on adult of *S. granarius* and *T. castaneum*	[201]
Foeniculum vulare	Fumigant toxicity against *S. oryzae*, *S. granarius* adults	[202]
Ostericum sieboldii	Contact and fumigant toxicity against *T. castaneum*, *S. zeamais* adults	[203]
Cuminum cyminum	Fumigant toxicity against *C. maculatus* adults	[204]
Coriandrum sativum	Fumigant toxicity against *T. confusum* and *C. maculatus* adults	[205]
Coriander sativum	Fumigant toxicity against *C. maculatus*, *T. castaneum* adults	[205]
Heracleum persicum	Adulticidal against *C. maculates*	[206]
Citrus aurantium	Fumigant toxicity against *T. castaneum* adults	[131]
Cinnamomum zeylanicum	Fumigant toxicity against *T. castaneum* adults	[131]
Gautheria fragrantissima	Fumigant toxicity against *T. castaneum* adults	[131]
Lavandula officinalis	Fumigant toxicity against *T. castaneum* adults	[131]
Oscimum sanctum	Fumigant toxicity against *T. castaneum* adults	[131]
Trachyspermum ammi	Fumigant toxicity against adults of *S. oryzae*	[132]
Anethum graveolens	Fumigant toxicity against adults of *S. oryzae*	[132]
Nigella sativa	Fumigant toxicity against adults of *S. oryzae*	[132]
Piper cubeba	Fumigant toxicity against adults of *S. oryzae, T. castaneum*	[132]

Plant species	Insecticidal activity and tested insect	Reference
Zingiber officinale	Fumigant toxicity against adults of *S. oryzae*, *T. castaneum*	[132]
Syzygium aromaticum	Fumigant toxicity against *S. oryzae*, *Acanthoscelides obtectus* adults	[207]
Citrus reticulate	Fumigant toxicity against *T. castaneum* adults and larvae	[208]
Citrus sinensis	Fumigant toxicity against *T. castaneum* adults and larvae	[208]
Eucalyptus amaldulensis	Fumigant toxicity against *S. oryzae* adults	[189]
E. grandis	Fumigant toxicity against *S. oryzae* adults	[189]
E. viminalis	Fumigant toxicity against *S. oryzae* adults	[189]
E. microtheca	Fumigant toxicity against *S. oryzae* adults	[189]
E. sargentii	Fumigant toxicity against *S. oryzae* adults	[189]
Datura stramonium	Fumigant toxicity against *T. castaneum*, *Trogoderma granarium*, *Cryptolestes ferrugineus* adults	[209]
Eucalyptus camaldulensis	Fumigant toxicity against *T. castaneum*, *T. granarium*, *C. ferrugineus* adults	[209]
Moringa oleifera	Fumigant toxicity against *T. castaneum*, *T. granarium*, *C. ferrugineus* adults	[209]
Nigella sativa	Fumigant toxicity against *T. castaneum*, *T. granarium*, *C. ferrugineus* adults	[209]
Citrus limonum	Fumigant toxicity against *Tenebrio molitor* adults	[133]
Cymbopogon citrates	Fumigant toxicity against *T. molitor* adults	[133]
Litsea cubeba	Fumigant toxicity against *T. molitor* adults	[133]
Muristica fragrans	Fumigant toxicity against *T. molitor* adults	[133]
Allium sativa	Fumigant toxicity against *S. oryzae* adults	[134]
Cinnamomum tamala	Fumigant toxicity against *S. oryzae* adults	[210]

Gaultheria and *Eucalyptus* oils exhibit high toxicity on *S. oryzae* and *C. chinensis* [176]. Pulegone, linalool and limonene are known to cause fumigant toxicity against rice weevil, *S. oryzae*. *Mentha citrata* oil containing linalool and linalyl acetate exhibit significant fumigant toxicity to rice weevils [177]. Insects like *S. zeamais*, *T. castaneum* and *Prostephanus truncates* are very sensitive to topical applications of *Citrus* oil [178]. *Solidago canadensis* oil shows strong toxic action against *S. granarius* [179]. Eugenol is also toxic to *S. granaries* [180]. Essential oils of *Anethum sowa*, *Artemisia annua*, *Lippia alba* and *Elletaria cardomum* have been reported for their toxic behaviour against *T. castaneum* [181-184]. Carvone and menthol are the most effective as fumigant against *T. castaneum* and *C. maculatus.* Cineole exhibits both contact and fumigant toxicity against *T. castaneum* [150]. Lee et al. have reported toxicity of menthol, methonene, limonene, α-pipene, β-pipene and linalool against *S. oryzae* and proved that these oil components exert its toxicity by inhibiting acetylcholine esterase enzyme [185]. *Trans*-anethole, thymol, 1,8-cineole, carvacrol, terpineol and linalool have been evaluated as fumigants against *T. castaneum* but only trans-anethole shows significant effect [186]. Essential oils from seeds of *Coriandrum sativum* and *Carum carvi* have been evaluated for fumigant toxicity against *S. oryzae*, *R. dominica* and *Cryptolestes pusillus*. Coriander contains linalool as the main component and is active against the three pests. Camphor-rich fractions are very toxic to *R. dominica* and *C. pusillus*. Carvone is the most effective monoterpenoid against *S. oryzae*. (E)-Anethole is toxic to *R. dominica* while vapours of limonene kills adults of *C. pusillus* [75]. A comparative study has been conducted to assess contact and fumigant toxicities of monoterpenes viz. camphene, camphor, carvone, 1-8-cineole, cuminaldehyde, fenchone, geraniol, limonene, linalool, menthol and myrcene on *S. oryzae* and *T. castaneum*. In fumigant toxicity assays, 1-8-cineole has been found most effective against *S. oryzae* and *T. castaneum*. Structure-toxicity investigations reveal that carvone has the highest contact toxicity against both insects. *In vitro* inhibition studies of acetylcholine esterase from adults of *S. oryzae* show that cuminaldehyde inhibits enzyme activity most effectively followed by 1-8-cineole, limonene and fenchone. 1-8-Cineole is the most potent inhibitor of acetylcholine esterase activity from *T castaneum* larvae followed by carvone and limonene [187]. Essential oils of tea tree (*Melaleuca alternifolia*), cinnamon (*Cinnamomum zeylanicum*), cloves (*Syzygium aromaticum*), lemongrass (*Cymbopogon flexuosus*), thyme (*Thymus vulgaris*), eucalyptus (*Eucalyptus globulus*), and jojoba (*Simmondsia chinensis*) have been tested for their fumigant activity against *C. maculatus* and *S. oryzae* adults. Mortality increases with increasing concentration of oils and exposure period. The effect of volatile compounds of *Citrus reticulata* and *C. sinensis* oils have been studied on *T. castaneum* and indicated that essential oil of *C. reticulata* shows more toxic effects than that of *C. sinensis* against larvae and adult of *T. castaneum* [188]. Fumigant toxicity of essential oils from five species of *Eucalyptus viz. E. camaldulensis*, *E. grandis*, *E. viminalis*, *E. microtheca* and *E. sargentii* have been studied against *S. oryzae* adults [189]. Results have indicated that mortality in adults increases with increasing concentration and exposure time. Insecticidal activity of essential oils from *Datura stramonium*, *Eucalyptus camaldulensis*, *Moringa oleifera* and *Nigella sativa* against three major insect pest *viz.*, *T. castaneum*, *T. granarium* and *C. ferrugineus* has been determined [190]. Essential oils fumigation causes mortality at all levels of concentration and exposure periods tested. *D*. *stramonium* oil is found to be the most toxic against *T*. *granarium* and *C*. *ferrugineus* while *N. sativa* shows the highest fumigant mortality against *T*. *castaneum*. Essential oils naturally are liquid at room temperature and get easy to change to vapours at room or with slightly higher temperature without any decomposition [90]. Therefore, volatile oils are frequently used as a fumigant against insects of stored grain. The mechanism of toxicity of essential oils and its constituents may be due to their neurotoxic effect. Essential oil and their active compound thymol can interact with neuromodulator octopamine. They induce neurotoxicity via effects on gated chloride channels GABA. Thymol has been reported to induce high toxicity to some insects like *S. oryzae* [191].

3.6. Growth inhibitors

Many essential oils and its constituents have been investigated and established for their egg laying, growth inhibitory and progeny production inhibitory activities against different insect pests.

3.6.1. Progeny production inhibitors

Fumigant toxicity of *Cymbopogon flexuosus* leaf oil has been investigated on progeny production of *R. dominica*, *S. oryzae* and *T. castaneum*. This oil shows high effectiveness against *R. dominica* and *S. oryzae* [211]. Similarly, essential oil from *Clausena anisata* and a mixture of it with clay have been investigated for its insecticidal activity and effects on progeny production. The aromatized clay powder as well as essential oil reduces the F_1 progeny insect production [212]. The activity of *Cymbopogon martini*, *Piper aduncum*, *P. hispidinervium*, *Melaleuca sp.* and *Lippia gracilis* oils and fixed oils of *Helianthus annuus*, *Sesamum indicum*, *Gossypium hirsutum*, *Glycine max* and *Caryocar brasiliense* have been studied against *C. maculatus*. These oils except *Melaleuca* sp. reduce egg viability and adult emergence to approximately 100% [213]. *C. cyminum*, *P. nigrum*, *F. vulgare*, *T. ammi*, *A. graveolens*, *I. verum*, *M. fragrans* and *N. sativa* essential oils reduce egg hatching rate, pupation and adult emergence when fumigated with sublethal concentrations. The essential oil of *N. sativa* has been found most effective followed by *A. graveolens*, *C. cyminum*, *I. verum*, *P. nigrum*, *M. fragrans* and *T. ammi* oils [163]. Bachrouch et al. have tested fecundity and hatching rate of *Ectomyelois ceratoniae* and *Ephestia kuehniella* exposed to *Pistacia lentiscus* essential oil [214]. Fecundity and hatching rate of both insects decreases with increase in concentration or exposure time to oil. *P. lentiscus* oil is toxic to eggs of *E. kuehniella* and *E. ceratoniae*. Essential oils from rhizomes of *Z. officinale* and berries of *P. cubeba* have been evaluated for developmental inhibitory activities against *T. castaneum*. Fumigation with sublethal concentrations of these essential oils reduces oviposition potential of adults and inhibits development of larvae to pupae and the pupae to adults [126]. Essential oil of *A. sativum* has been evaluated for its oviposition inhibitory activities against *T. castaneum*. *A. sativum* reduces oviposition potential of adults when treated by fumigant and contact method both [147].

3.6.2. Development inhibitors

Several essential oils and their constituents have properties similar to juvenile hormone and act as insect growth regulators (Table 7). 1,8-Cineole isolated from *Artemisia annua* is also a potential insecticidal allelochemical that reduce growth rate, food consumption and food utilization in some post harvest pests [215, 216]. Essential oil from *C. schoenanthus* shows development inhibition in all stages of *C. maculatus* [217]. Essential oil from *Hyptis spicigera* has been evaluated for insecticidal activities on *C. maculatus*. Essential oil has dose-dependent insecticidal effect while sublethal doses are repellent to adults, reduces oviposition and eggs viability with increasing doses. Essential oil shows lethality in larvae developing within cowpea seeds; and younger instars are more susceptible [218]. The fumigant toxicity of *Laurus nobilis* and *R. officinalis* oils has been evaluated against all development stage of *T. confusum*. The major component of both oils is 1,8-cineole. The two oils are toxic to all stages of the insect [219]. *C. cyminum*, *P. nigrum*, *F. vulgare*, *T. ammi*, *A. graveolens*, *I. verum*, *M. fragrans* and *N. sativa* essential oils cause death of *T. castaneum* larvae and adults by fumigation. These essential oils reduce oviposition potential and increase developmental period of *T. castaneum*. Fumigation inhibits development of larvae to pupae and the pupae to adults and also result in the deformities in different developmental stages of insect [117, 118, 121]. They cause disruption in growth and affect reproduction of insects. *M. fragrans*, *N. sativa*, *P. nigrum* and *T. ammi* oils induce changes in growth and reproduction of *C. chinensis* [163]. Oviposition deterrence has been recorded when *C. copticum* and *Vitex pseudo-negundo* essential oils have been applied on *C. maculatus* [220]. *Elettaria cardamomum* oil has shown oviposition deterrence effect on *C. maculatus*. Therefore, treatment with this essential oil reduces numbers of insects in treated grain [221].

Table 7. Essential oils with insect growth regulatory (igr) activity.

Plant species	Insect species	Reference
Trachyspermum ammi	*T. castaneum*	[117]
Anethum graveolens	*T. castaneum*	[118]
Cuminum cyminum	*T. castaneum*	[118]
Piper nigrum	*T. castaneum*	[118]
Foeniculum vulgare	*T. castaneum*	[118]
Carum copticum	*C. maculatus*	[220]
Anethum graveolens	*C. chinensis*	[163]
Cuminum cyminum	*C. chinensis*	[163]
Illicium verum	*C. chinensis*	[163]
Myristica fragrans	*C. chinensis*	[163]
Nigella sativa	*C. chinensis*	[163]
Piper nigrum	*C. chinensis*	[163]
Trachyspermum ammi	*C. chinensis*	[163]

Plant species	Insect species	Reference
Ageratum conyzoides	*C. maculatus*	[224]
Citrus aurantifolia	*C. maculatus*	[224]
Melaleuca quinquenervia	*C. maculatus*	[224]
Citrus paradise	*R. dominica*	[222]
Citrus reticulate	*R. dominica*	[222]
Zingiber officinale	*T. castaneum*	[132]
Piper cubeba	*T. castaneum*	[132]
Allium sativum	*T. castaneum*	[223]

Developmental inhibitory activities of α-pinene and β-caryophyllene alone or in binary combination have been determined against 4th instars larvae of *T. castaneum*. Percentage of larvae transformed into pupae and percentage of pupae transformed into adult decreases when fumigated with sublethal concentrations of α-pinene and β-caryophyllene alone or in binary combination. Results indicate that α-pinene and β-caryophylene in binary combination show synergism and reduce pupation and adult emergence in *T. castaneum* [130]. Abbas et al. have reported *C. reticulata* oil inhibits growth and pupation in *R. domonica* [222]. Several essential oils are good inhibitors of pest's oviposition disturbing general growth of the populations. Essential oil of *A. sativum* has been evaluated for its developmental inhibitory activities against *T. castaneum*. *A. sativum* essential oil interferes with developmental processes and reduces transformation of larvae into pupae and adult emergence [223]. This disruption in growth of insects may be due to inhibition of different biosynthetic processes of insects at different growth stages. These studies revealed good results for utilizing sublethal concentrations/doses of essential oils and its constituents in reducing egg lying and hatchability, progeny production and growth inhibition.

3.7. Mode of action of essential oils

Several plant derived essential oils and its constituents have been described as insecticides [225]. Thus, the doses or concentrations of essential oils and its constituents needed to kill insect pests and their mechanism of actions are important for the safety of humans and other non-trget vertebrates. The toxicity of essential oils in insects appears to be the result of effects mainly on the insect's nervous system either by inhibition of acetylcholinesterase or by antagonism of the octopamine receptors [93]. The rapid action against some insects is indicative of a neurotoxic mode of action similar to that of conventional synthetic insecticides. Several studies indicate that essential oils and monoterpenoids cause insect mortality by inhibiting acetylcholinesterase enzyme activity. Ryan and Byrne have suggested that the toxic effect may be attributed to reversible competitive inhibition of acetylcholinesterase by binding to active site of the enzyme [226]. Chaubey has reported that *A. sativum* essential oil inhibits acetylcholinesterase enzyme activity in *T. castaneum* and *S. oryzae* adults [134, 210, 223]. A monoterpenoid, linalool has been demonstrated to act on the nervous system affecting ion transport and the release of acetylcholine esterase in insects [227]. Octopamine has a broad spectrum of biological roles in insects acting as a neurotransmitter, neurohormone and circulating neurohormone-neuromodulator [228, 229]. Octopamine exerts its effects through interaction with at least two classes of receptors. On the basis of pharmacological effects, these have been designated as octopamine-1 and octopamine-2 [230]. Interruption in the functioning of octopamine results in total break down of nervous system in insects. Therefore, octopaminergic system of insects represents a target for insect control. The lack of octopamine receptors in vertebrates accounts for the mammalian selectivity of essential oils as insecticides. A number of essential oil compounds have been demonstrated to act on octopaminergic system of insects [231]. Enan has suggested that toxicity of essential oil/constituents is related to the octopaminergic nervous system of insects [232]. Kostyukovsky et al. have shown the activity of two essential oil constituents, ZP-51 and SEM-76 on several insect species [93]. Both ZP-51 and SEM-76 show an inhibitory action on acetylcholinesterase, but only at the high dose. Essential oils can also disrupt communication in mating behaviour of insect by blocking the function of antennal sensilla. This lowers fecundity and ultimately the population of insect pest [233]. Fumigant toxicity of caryophyllene oxide may result from the inhibition of the mitochondrial electron transport system because changes in the concentration of oxygen or carbon dioxide may affect respiration rate of insect, thus, eliciting fumigant toxicity effects [234]. Thus, in conclusion plant derived essential oils and its constituents exert its toxicity in insects by interfering with the nervous co-ordination and respiratory system.

4. SYNERGISTIC ACTION OF ESSENTIAL OILS

Chemical control of insect pest involving synthetic insecticides has induced resistance in several insects. Thus, the current aim of pests control stratigies is to produce plant based formulations which reduce the risk of developing resistance against insecticide. None of the plant products alone provide adequate crop protection, but attemps in this direction have been started. Applications of botanical preparations increase the farmer's confidence in indigenous technology [235]. Although repellent activity of essential oils is generally attributed to some particular compounds, a synergistic phenomenon among these metabolites may result in a higher bioactivity compared to isolated components [236]. Omolo et al. have compared repellent activities between essential oil and synthetic oils formulated with its major constituents [152]. The activities of synthetic oils have been much smaller than those of corresponding natural essential oil. This indicates that minor constituents also contribute to repellent activity. This reflects the importance of compositional complexity in providing bioactivity to natural mixtures. It has been suggested that this is due to the fact that plant's defense system exists as a suite of compounds not as individual ones. Accordingly, minor constituents although found in low percentages may act as synergists enhancing the effectiveness of major constituents through a variety of mechanisms [237].

The components of synergistic combinations have diverse modes of actions and thus, efficacy of combined product is greater than sum total of known and unknown chemical components. Both positive and negative synergism can occur between essential oil and/or components. Essential oils combinations such as thyme, anise and saffron have been demonstrated for synergistic activity [238]. Hummelbrunner and Isman have reported that mixtures of different monoterpenes produce synergistic effect on mortality [180]. Chaubey has reported that mixture of *T. ammi*, *A. graveolens* and *N. sativa* essential oils cause reduction in oviposition and developmental inhibitory activities against *T. castaneum* at lower concentration than alone [200]. Terpenes, α-pinene and β-caryophyllene have been evaluated for their repellent, acute toxicity and developmental inhibitory activities alone and in binary combination against *T. castaneum*. Fumigation of larvae and adults of *T. castaneum* with these two compounds caused lethality in them. Median lethal concentration of α-pinene and β-caryophyllene binary combination against adults and larvae has been found lower than when used alone. Fumigation with two sublethal concentrations of these two compounds in binary combination reduce oviposition potential of adults and inhibited pupation and adult emergence in larvae more potently than used alone. This study concludes that these two volatile compounds in binary combination shows synergism and thus, can used as efficient insecticidal tool against *T. castaneum* [132]. Since essential oils are mixture of 20-60 chemical compounds of diverse nature, it will be difficult for the insects to develop resistance against them. This also suggests that essential oil constituents alone can be a weaker candidate than crude essential oil in insect pest management programme.

5. STRUCTURE-ACTIVITY RELATIONSHIPS IN ESSENTIAL OIL COMPONENTS

The relation between chemical structures of essential oil's constituents and their biological activity has been well documented. It has been reported that any change in molecular structure can change their biological activities. Modifications of chemical structure of monoterpenoids enhance biological properties of essential oils as a whole as well as monoterpenoids that contain functional groups. The activities of essential oils and their constituents depend on functional group and chemical properties such as volatility and molecular weights [239]. Scientists have studied correlation between chemical structure of essential oils constituents and their insecticidal activity. Essential oil from *M. arvensis* has been reported to have insecticidal activity. The L-menthol isolated from this oil and seven of its acyl derivatives have been evaluated against stored grain insect pests. It has been reported that menthyl propionate and L-menthol have high insecticidal activity. High activity of menthyl propionate as compared to L-menthol can be due to increasing number of methyl groups in side chain [173]. Due to nucleophilic properties of methyl group, increase in number of methyl groups on side chain cause decrease in positive charge (increase negative charge) on carbon atom. Therefore, high activity of menthyl propionate may be due to increasing negative charge on carbon atom because of the presence of two methyl groups. However, increase of electrophilic groups such as methyl groups in chain leads to increase the activity of a function carbon atom.

Depending on the number of methyl groups, acetate derivative have slight activity while menthyl propionate have high insecticidal activity. Low activity of menthyl benzoate has been observed because they do not have methyl group and the benzene ring is attached directly to menthyl carbon. Activity of menthyl cinnnamate has been found moderate as it has a double bond and benzene ring is not directly attached to carbon atom [173, 240]. Benzene derivatives (eugenol, isoeugenol, methyl eugenol, safrole and isosafrole) and terpenes (cineole, limonene, p-cymine and α-pinene) have been evaluated for insecticidal activity [101]. It has been observed that derivatives of benzene have higher insecticidal activity than that of terpenes. This toxicity may be due to the active groups in benzene derivatives. The presence of double bond in the side chain of aromatic ring and the substitution of methoxy group play an important role in the toxicity of these analogues. The knock down and contact activity has been found to increase in methyl eugenol due to further methoxy group. The order of contact toxicity of these compounds is methyl-eugenol > isosafrole = eugenol > safrole. In contrast, when double bond in side chain is nearer to aromatic ring, fumigant toxicity is decreased. Therefore, safrole shows more fumigant activity than isosafrole. However, benzene derivatives have more insecticidal activity than monoterpenes [101]. Insecticidal activity of thymol, pulegone, trans-anethole and eugenol has been evaluated against *S. litura* [180]. Thymol has higher toxicity than pulegone, trans-anethole and eugenol against *S. litura*. The order of toxicity of these compounds observed is thymol > pulegone > trans-anethole > eugenol. The high toxicity of thymol as compared to other compounds has been attributed to the presence of methyl groups in the side chain. This effect can be due to electron-donating property of methyl groups which decrease positive charge on carbon atom of side chain. Further researches involving strcture-activity are required to enhance insecticidal potency of essential oil. Further studies should be carried out to investigate whether the alteration in structure of essential oil constituents can modify mode of action.

6. MAMMALIAN TOXICITY OF ESSENTIAL OILS

Before using essential oils and/or their constituents as stored grain protectants, their probable toxicity in non-target animals including mammals must be acknowledged. The common use of plant essential oils in drugs and foods clearly indicate that essential oils show insignificant toxicity towards mammals. In general, most of the essential oils and their active constituents are nontoxic to mammals [241]. However, some essential oil/constituents are toxic to human and other mammals. The LD_{50} for toxic constituents against rats ranges from 800 to 3,000 mg/kg. Pulegone has been found toxic to rats with LD_{50} 150 mg/kg intraperitoneally [242]. Also, thujone oil is very toxic to rats with LD_{50} 45 mg/kg intraperitoneally [243].

Being a mixture of several constituents, essential oils seem to have no specific cellular targets [244]. Since essential oils are lipophilic in nature, they pass through cytoplasmic membrane and disrupt membrane structure leading to cytotoxicity. In eukaryotic cells, essential oils can induce depolarization of mitochondrial membranes by interfering with various ion channels [245, 246]. They change fluidity of membranes which become abnormally permeable resulting in leakage of radicals, cytochrome C, calcium ions and proteins similar to oxidative stress and bioenergetic failure. Permeability of outer and inner mitochondrial membranes leads to cell death by apoptosis and necrosis [247, 248]. This cytotoxic property is of great importance in the applications of essential oils in insect pest management.

Essential oils induce cytotoxicity in mammalian cells by inducing apoptosis and necrosis. Since most essential oils have been found to be cytotoxic without being mutagenic, it is likely that most of them are also devoid of carcinogenicity. However, some essential oils and their constituents are considered as secondary carcinogens after metabolic activation [249]. *Salvia sclarea* and *Melaleuca quinquenervia* oils induce estrogen secretions which can cause estrogen-dependent cancers. Some others contain photosensitizing molecules like flavins, cyanin, porphyrins, hydrocarbures which can cause skin cancer. Estragole, a constituent of *Ocimum basilicum* and *Artemisia dracunculus* oils has shown carcinogenic properties in rat and mouse [250, 251]. Psoralen, a photosensitizing molecule found in some essential oils like *Citrus aurantium* can induce skin cancer after formation of covalent DNA adducts under ultraviolet A or solar light [252]. Pulegone, a component of essential oils of mint species can induce carcinogenesis [253]. Safrole, the major constituent of *Sassafras albidum* and *Mespilodaphne pretiosa* oils induces carcinogenic metabolites in rats [254]. Methyl eugenol has also been shown to be carcinogenic in rats [254].

The most necessary aspect of using essential oils and/or their constituents for pest control is assessment of their toxicity in mammals because many essential oils and their constituents are commonly used as culinary herbs and spices. Many of the commercial products based on essential oils are included in the GRAS (Generally Recognized as Safe) list by FDA (Food and Drug Administration) and EPA (Environmental Protection Agency) in USA for food and brevarage consumption [255]. Some of essential oil terpenoids are moderately toxic to mammals, but, with few exceptions, oils themselves or products based on oils are mostly nontoxic tomammals, birds and fish. Due to their volatility, oils and their constituents are environmentally nonpersistent with outdoor half lives of 24 hours on surfaces, in soil and in water [256]. Because many conventional pesticides are harmful for public, botanical-based pesticides especially essential oils become a popular choice for insect pest management in storage.

7. ESSENTIAL OIL-BASED INSECTICIDES FROM RESEARCH TO MARKET

From time immemorial with the beginging of human civilization and storage of grain against poor agriculture production and famine, insect species have been damaging stored grain. To protect the grains in storage from insect infestation, aromatic plants have been used traditionally worldwide. Thus, scientific communities have started re-evaluating these plants and their volatile oils against insect pests of stored grains. Commonly used essential oils and active substances to be used as insecticide takes a long time as certain toxicological and ecotoxicological tests are required for registering commercial products. The EcoSMART technologies (USA) have introduced some pesticides based on essential oils [256]. These formulations are based on cinnamon oil with cinnamaldehyde. The EcoSMART technologies have introduced other plant volatile based insecticides EcoPCOR. They contain eugenol and 2-phenethyl propionate as active ingredients. They are used against crawling and flying insects. The EcoTrolTM formulation is based on rosemary oil and is used as an insecticide on horticultural crops. Garlic oil-based insecticides have also been produced in the US. These formulations contain mint oil as the active ingredient. They are used in home and garden for pest control [257]. However, essential oils must have following properties to be used in insect pest management programme [256]:

- Essential oil must be produced in large scale throughout the world.
- They must have broad activity against insects due to their multiple modes and sites of action.
- They must have a variety of actions such as insecticidal, attractive, repellent, fumigant and antifeedant.
- Essential oils and their active constituents must be nontoxic to mammals including human.
- Oils and active compounds must be environmentally non-persistent.
- They must be effective under low pest pressure.
- They must have a short residual half-life on plants.

Inspite of promising activities of essential oils against several insects, some problems have registered regarding its commercial application in the field. For example, essential oil’s volatility, water solubility and oxidation play important role in essential oils activity, application and persistent. Therefore, these problems must be resolved before using essential oils as alternative to synthetic pesticides for pest control [258]. New formulations with nanotechnology ‘Nanoformulation’ can resolve these problems. The new trend for using nanoformulation leads to protect essential oils from degradation and to increase their residue half-life by reducing evaporation. Nanoformulations have properties of controlled release of essential oils and ease of application and handling [259]. These nanoformulations can enhance essential oils activity due to small particle size. Yang et al. have shown that loaded nanoparticles with garlic essential oils are effective against *T. castaneum* [260]. Anjali et al. have reported that insecticidal activity of neem oil increases in nanoemulision formulation [261]. This effect can be due to the smallest droplet size of essential oil nanoemulsion. New nanotechnology methods have been planned to control *H. armigera* [262]. This new method stabilizes *Artemisia arborescens* oil with better insecticidal activity. This stability can be due to built-in of essential oil with solid lipid nanoparticles and developed an emulsion. Nanoencapsulation of essential oils have been shown high repellent activity than essential oils [115]. Some patents involving essential oils have shown that majority of the inventions focus on household insects. A cleaning solution including clove essential oil and pyrethroid destroy eggs and larvae, and leave a residue to prevent reinfestation by *Blattaria* [263]. Several essential oil based formulations have been proposed to control mosquitoes and flies [264]. Spearmint, bitter almond and birch, *Betula lenta* bark essential oils have been incorporated into a mixture showing insecticide and insect repellent properties [265].

A large number of patents have been assigned for preservation of clothes from moths and beetles including application of a solution containing clove essential oil on woollen cloth [266]. More recently, wash fast insect-resistant fabrics have been created with partially or wholly hollow porous fibres coated with encapsulated insecticidal agents such as *Eucalyptus* oil [267]. Beside these domestic uses, essential oils have got applications in agriculture and the food industry. Essential oils can also be incorporated with polymers into sheets. Attractant adhesive films with essential oils have been prepared to control insects in agriculture and horticulture. Coating materials, useful in agricultural structures, include pine essential oils to enhance their insecticidal properties and repel harmful insects [268]. Adhesives containing acrylic polymers and essential oils have been shown killing effect for *Blatella germanic*a [269]. Essential oils have also been shown some usefulness for building materials. A wood preservative solution mixed with eucalyptus oils pyrethroids and borax have common applications [270].

8. SUMMARY

Essential oils are produced as secondary metabolite in aromatic plants. These are complex mixtures of volatile compounds in different concentartions. These oils and its constituents have repellent, antifeedant, ovicidal, oviposition inhibitory and developmental inhibitory activities in insects. These interfere with the respiratory and nervous system of the insect. Thus, essential oils can be used as alternatives in insect management. Most of these oils are selective in their role with little or no harmful effect on the environment and the non-target organisms including human. The main aim of this review is to provide basic informations regarding essential oils, chemistry and their role in stored grain insect pest management.

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