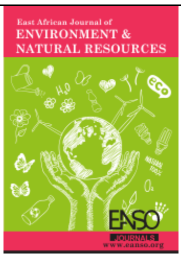

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Original Article

Selected Plant Essential and Seed Oils and their Secondary Metabolites Used as Insecticides: A Review

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ABSTRACT

Conscious consumers around the world have been getting organized to urge and encourage farmers to practice environmentally and human friendly organic agricultural production. Synthetic insecticides have been established to be toxic to the environment, non-target organisms and even to humans when used to control problematic insects in the homes/gardens and in agricultural establishments. On the other hand, plant-based insecticides have proven to be environmentally friendly and less toxic to non-target organisms and less detrimental to human health due to their short half-lives. This is because they do not possess 'unnatural' ring structures with relatively few halogen substituents, known to be closely related with disease states and genetic abnormalities, including cancer. Plant based oils such as cinnamon, garlic, peppermint, rosemary essential oils and castor, neem, sesame seed oils have been found to contain secondary metabolites with effective insecticidal activities. These secondary metabolites include cinnamaldehyde, which has been found to kill common house flies; ricin, which has been found to effectively control diamondback moth; dimethyl trisulphide and diallyl disulphide, which have been found to effectively kill red flour beetle; azadirachtin and menthol, which have strong insecticidal activity against malaria causing and dengue fever mosquitoes; α-Pinene, which has been found to have repellence effects on aedes mosquitoes; sesamin, with strong insecticidal activity against agricultural fungus-growing termites; and thymol, with strong larvicidal activities against malaria causing mosquitoes.

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INTRODUCTION

The United States of America's Environmental Protection Agency (US EPA) define an insecticide as a chemical used to control insects by killing them or preventing them from engaging in undesirable or destructive behaviours. Insecticides are classified based on their structure and mode of action. Many insecticides act upon the insect's nervous system, while others act as growth regulators or endotoxins (Smith et al., 2022). There are 859 chemical formulations registered for use as insecticides in India under the insecticide's act of 1968 (Pesticides and formulations registered for use in country under insecticides act of 1968) with tens of thousands of insecticides with different brand names produced from these formulations every year. This makes the insecticide business a multibillion-dollar industry not only in India but around the world.

Insecticides are not only used to control insects of agricultural importance, as some are used to control household insects such as mosquitoes, flies and cockroaches. In Botswana, insecticides are controlled under the Agrochemical Act of 1999, chapter 35:09. The act provides for the registration and licensing of agrochemicals; to control or regulate their importation, manufacture, distribution, use and disposal, so as to prevent pollution to the environment or harm to human, plant or animal life; and, to provide for matters incidental and connected to the foregoing (Agrochemical Act of 1999, chapter 35:09). The act, therefore, makes it mandatory only for importers and manufacturers of insecticides used as agrochemicals to be regulated leaving out those used for other purposes such as control of household insects. This loophole has seen an increase in the number of insecticides imported into the country, the majority of which are synthetic (chemical) insecticides whose environmental and human impacts are not well established.

Most chemical insecticides have been established to be toxic to other organisms living in the environment. Contamination from soil leaching, field runoff water, and spray drift, as well as adverse effects on wildlife, fish, plants, and other non-target organisms can potentially occur depending on the specific toxicity, quantity and potency of the applied insecticide (Ibanez et al., 2012). Once released into the environment, insecticides can be subjected to airborne and waterborne entry in aquatic ecosystems. Airborne processes encompass wind drift during insecticide spraying (spray drift) and volatilisation after application with subsequent atmospheric transport that may lead to the deposition of compounds in remote ecosystems (thousands of kilometres) from their initial application (Ralf et al., 2011). DDT (dichloro-diphenyl-trichloroethane) was developed as the first of the modern synthetic insecticides in the 1940s. It was initially used with great effect to combat malaria, typhus, and other insect-borne human diseases among both military and civilian populations. It was also effective for insect control in crop and livestock production, institutions, homes, and gardens (United States Environmental Protection Agency, 2023). In 1972, the United States Environmental Protection Agency issued a ban for DDT based on its adverse environmental effects on wildlife such as bees and small birds, as well as its potential human health risks (United States Environmental Protection Agency, 1975). The ban on DDT put a spotlight

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on synthetic insecticides as chemicals of environmental concern. The use of a wide range of synthetic insecticides has been restricted recently, due to their high cost, harmful environmental effects, their non-biodegradable nature, and increasing insecticidal resistance (Mina et al., 2016). This has, therefore, led scientists to put more focus and research on organic insecticides as alternatives to synthetic insecticides.

An important benefit of natural product-based pesticides is their relatively short environmental half-lives, because they do not possess 'unnatural' ring structures and many halogen substituents which are closely related with disease states and genetic abnormalities in humans and animals, including cancer (Anne et al., 2006, Yamawaki et al., 2021, & Hart et al., 2000). Conscious consumers in many countries, especially in the most developed countries, have been getting organized to urge and encourage farmers to practice environmentally and human friendly organic agricultural production (organic farming). This is because, this production system sustains the health of soils, ecosystems, and people. It depends on ecological processes, biodiversity and cycles adapted to local conditions, rather than the use of inputs such as synthetic insecticides with adverse environmental and health effects (Buragohain, 2020). This review is looking at plant secondary metabolites in selected essential and seed oils which have been found to be effective against insects of economic and public health importance as potential alternatives to synthetic insecticides.

SELECTED PLANT SEED AND ESSENTIAL OILS USED AS INSECTICIDES AND THEIR ACTIVE INSETICIDAL AGENTS

Herbal traditional medicines, including plant oils, have been used for thousands of years as sources of bioactive and therapeutic substances for industrial and agricultural purposes (Franck et al., 2009). One of these uses is as insecticides to control both agricultural and household insect pests. Different plant oils have also been used in the past to control insects in the home and in the fields. The following plant oils are some of the plant oils which have been studied and have been found to have insecticidal activities.

CINNAMON (*Cinnamomum Zeylanicum*) INSSENTIAL OIL

Cinnamomum zeylanicum (Cinnamon) is a perennial tree of medium size, (figure 1) native of South Asia belonging to the *Lauraceae* family, which is acclimatized in other tropical regions of the world (Liese et al., 2008). Boito et al, 2018, demonstrated that 10 % v/v of *C. zeylanicum* essential oil is able to kill 100% of *Musca domestica* (Common House Fly), in addition to its repellent effect against *Haematobia irritans* (Horn Fly) flies in cows. The five main components of *C. zeylanicum* essential oil are cinnamaldehyde (41.27 %), linalool (13.05 %), methyl eugenol (10.87 %), camphor (8.l12 %) and eugenol (7.03 %), representing 80.34% of the total essential oil composition. The flavonoid cinnamaldehyde ($C_6H_5CH{=}CHCHO$, ((2E)-3-phenylprop-2-enal)) at 41.27 % and noncyclic monoterpenoid Linalool $(CH_3)_2C{=}CH(CH_2)_2C(CH_3)(OH)CH{=}CH_2$, (2,6-dimethyl-2,7-octadien-6-ol)) at 13.05 % (figure 1) are the components reported to be responsible for insecticidal activity of *C. zeylanicum* (Boito et al., 2018 and Singha et al., 2007).

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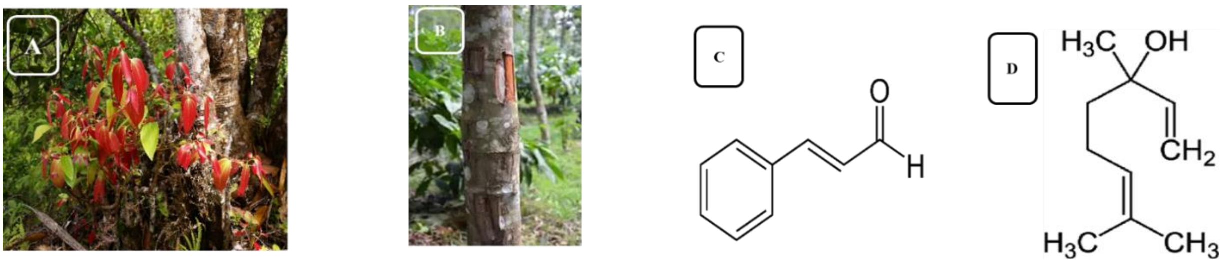
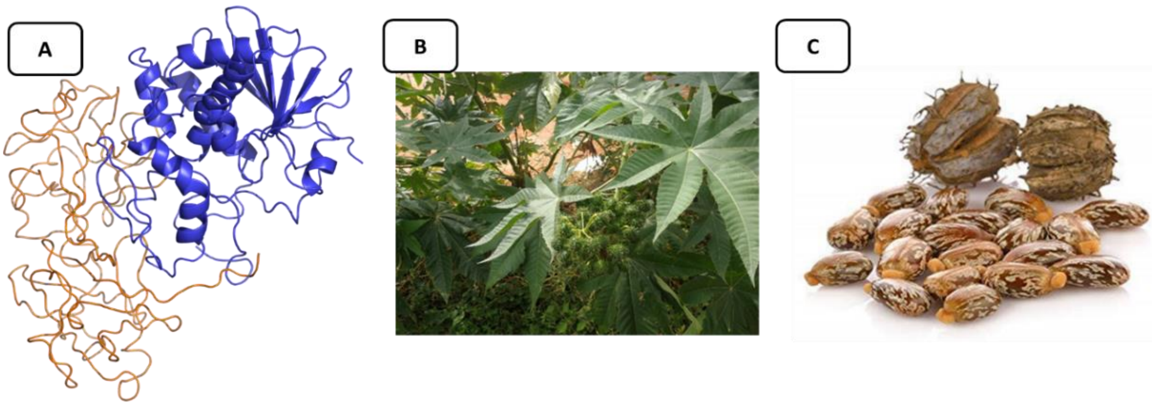
Figure 1: Cinnamon (C. zeylanicum) tree (A), the Cinnamon bark (B) used for making cinnamon spices and essential oils (Rawat et al., 2020), cinnamaldehyde (C) (Vina et al., 2022) and linalool (D) (Everton et al., 2022).



CASTOR (*Ricinus Communis L*) SEED OIL

Ricinus communis L (Castor oil) plant (figure 2) is a species of flowering plant in the spurge family, *Euphorbiaceae*, which contains a vast number of plants mostly native to the tropics. The plant is famous for its castor bean/seeds (figure 2) which produces castor oil which has many industrial uses ranging from cosmetics to coatings (Bolaji et al., 2014 and Paphane et al., 2021). The most abundant fatty acid within the *R communis* seed-oil is Ricinoleic Acid (Methyl-12-hydroxy-9-octadecenoate) at 81.51 % followed by Linoleic acid (9,12-Octadecadienoic acid (Z,Z)) at 6.74 %, Oleic acid (9-octadecanoic acid) at 3.43 %, Palmatic Acid (Hexadecanoic acid, methyl ester) at 2.32 %, 3-Eicosene at 2.06 %, 1,2-Octadecanediol at 1.54 %, 2-Ethylnon-1-en-3-ol at 1.44 % and lastly 1-Hexadecanol,2-methyl at 0.96 % (Paphane et al., 2021). Apart from the valuable oil, *R communis* seeds contain about 0.12 % w/w of proteinaceous toxin called ricin (figure 2) which remains within the de-oiled cake during oil extraction (Kumar et al., 2004). The treatment of Brassicaceae crops such as cabbage and cauliflower with *R communis* aqueous 20 % cake extracts and 10 % oil emulsion was found to effectively control diamondback moth (*Plutella xylostella*) with 100 % mortality recorded on 3rd instar larvae (Tounou et al., 2011). The toxicity of the plant is attributed to the presence of ricin, a water-soluble glycoprotein concentrated in the seed endosperm but present in lesser concentrations in other parts of the plant and reputed to be one of the most poisonous of the naturally occurring bio-compounds (El-Nikhely et al., 2007).

Figure 2: Ricin structure (A) (Lord and Roberts, 2005), *R communis* (Castor oil) plant (B) and *R communis* (Castor oil) seeds (C) (Paphane et al., 2021).



GARLIC (*Allium Sativum*) ESSENTIAL OIL

Allium sativum, also known as Clove Garlic, is part of the *Lilliceae* plant family (Figure 3). It is a bulbous perennial herb, closely related to the onion (Satyanand et al., 2013). The primary compounds of the garlic essential oil are dimethyl trisulfide (19.86%), diallyl disulfide (18.62%), diallyl sulfide (12.67%), diallyl tetrasulfide

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(11.34%), and 3-vinyl-[4 H]-1,2-dithiin (10.11%), followed by diallyl trisulfide (5.74%), allyl trisulfide (4.41%), 1,4-dimethyl tetrasulfide (4.06%), allyl disulfide (3.95%), methyl allyl disulfide (3.87%), and methyl allyl trisulfide (3.76%) (Satyanand et al., 2013). Plata-Rueda et al, 2017, noted that the mortality of *Tenebrio molitor* (mealworm) is obtained with 16 and 32 % (w/v) of the garlic essential oil. The toxicity of the oil to *T. molitor* is attributed to diallyl sulphide ($C_6H_{10}S$) and diallyl disulphide ($C_6H_{10}S_2$) (figure 3). Both diallyl trisulphide and methyl allyl disulphide isolated from garlic were found to be lethally toxic to *Tribolium castaneum* (Red Flour Beetle) adults and *Sitophilus zeamais* (Maize weevil) adults. Similar results were also obtained with contact toxicity of the essential oil of garlic to these two species using filter paper impregnation method showing that these compounds are responsible for insecticidal activity of garlic (Huag et al., 2000).

Figure 3: *Allium sativum* (Garlic) plant (A), Dimethyl trisulphide (B) and diallyl disulphide (C) structural formula (Huang et al., 2000).

NEEM (Azadirachta Indica) SEED OIL

Azadirachta indica, the neem tree (figure 4) belongs to family *Meliaceae*, is a tropical evergreen tree related to mahogany. *A. indica* oil is extracted from the seeds of the neem tree and has insecticidal properties to which it has been used in rice cultivations to control various pests (Roshan and Verma, 2015). Aremu et al. (2009) reported that a 10 % w/w neem oil body lotion/cream repelled *Anopheles gambiae* (African Malaria Mosquito) from the human skin for about 6 hours. Neem oil-based insecticides have been found to be effective against a wide range of insects of medical and veterinary importance, including mosquitoes, sparing the economically important ones like bees (Nicoletti et al., 2016). Azadirachtin ($C_{35}H_{44}O_{16}$), figure 4, is the most important component of a group of limonoids found in neem seeds including neem seed oil responsible for the insecticidal activity of the oil (Chatterjee et al., 2023).

Figure 4: *Azadirachta indica*, the neem tree (A), its ripe fruits (B), dried seeds (C) and the Azadirachtin structure (D) (Chatterjee et al., 2023).

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Mentha Piperita L. (PERPPERMINT) ESSENTIAL OIL

Mentha piperita (Peppermint) is a perennial herbal medicine and belongs to the mint family *Lamiaceaeas* (Figure 5) (Nayak et al., 2020). The peppermint plant is a cross hybrid between water mint (*Mentha aquatica L.*) and spear mint (*Mentha spicata L.*) plants (Ayman et al., 2015). The plant is used extensively in traditional remedies and alternative medicines for treatment of digestive disorders and nervous system actions because of its antitumor and antimicrobial activities (Ayman et al., 2015). The plant is cultivated mainly for its essential oil, which is obtained by steam distillation from the aerial parts of the plant. Menthol at 50% w/w is the main constituent of the peppermint essential oil followed by menthone at 10–30% w/w, menthyl esters around 10% w/w and monoterpene derivatives (pulegone, piperitone, menthofurane) at very low concentrations (Saeidnia et al., 2005). Kumar et al. (2011), investigative bioassay results showed that *M. piperita* possesses excellent larvicidal efficiency against dengue fever mosquito vector (*Aedes aegypti L)* with a LC_{50} value of 111.9 ppm and LC_{90} value of 295.18 ppm after 24 h of exposure. They also observed that the toxicity of the essential oil increased when the larvae were exposed to the oil for longer duration.

Figure 5: _Mentha piperita_ (Peppermint) herb (A) its essential oil (B) (Loolaie et al., 2017) and menthol structure (C) (Guy et al., 2013).

Rosmarinus Officinalis L. (ROSEMARY) ESSENTIAL OIL

Rosmarinus officinalis L. (Rosemary) is an evergreen brush belonging to the *Lamiaceae* family (Figure 6). In natural conditions, it can reach approximately 1.0 m to 2.5 m in height (Pawlowska et al., 2020). Rosemary plant is cultivated for its aromatic essential oil which is called "rosemary oil" and is obtained by steam distillation of the fresh leaves and flowering tops of the plant (Al-Sereti et al., 1999). The oil is an ingredient for Eau-de-cologne, hair tonics, hair lotion, cold cream etc. It has been used as an antispasmodic in renal colic and period pains and in relieving respiratory disorders and many other medical uses (Al-Sereti et al., 1999). *Rosmarinus officinalis L.* was reported to be a more potent inhibitor against Gram (+) bacteria due to the difference in the structure of the bacterial cell wall (Pawlowska et al., 2020). Rosemary oil profile is made up of α-Pinene at 25.62% - 29.42 % w/w, camphene at 9.61 - 24.62 % w/w, and camphor at 18.39 % - 20.95 % w/w (Adouane et al., 2022). Dhillon et al., 2020 noted that cotton fabrics treated with *Rosmarinus officinalis L.* essential oil showed 100 percent protection against Aedes mosquitoes (*Aedes aegypti*) till after 5 washings, after which this protection is reduced. Mahfuz et al, 2023's repellency bioassays of rosemary essential oil against red flour beetles (*Tribolium casteneum*), showed that the highest repulsion effect is observed after 2 hours of exposure in selected doses of the essential oil (Mahfuzl et al., 2023).

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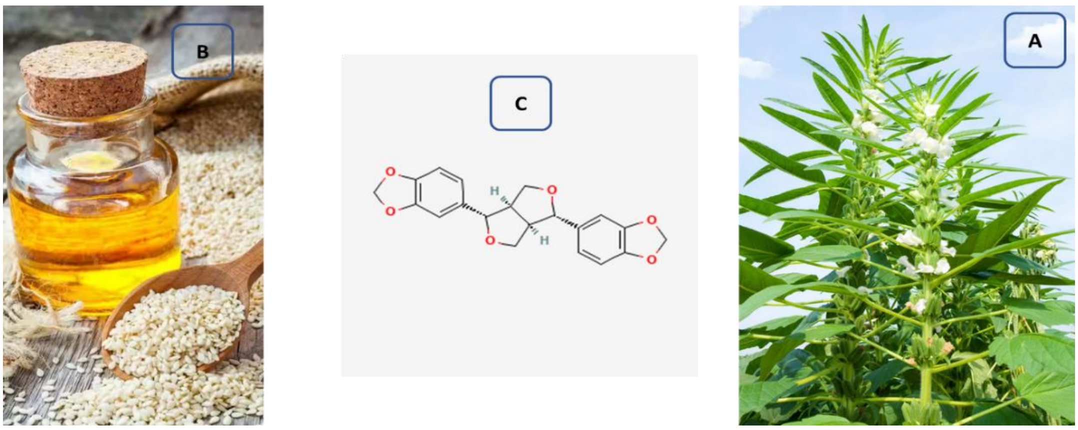
Figure 6: ***Rosmarinus officinalis L.*** **(Rosemary) herb (A) (Mersin and Iscan, 2022), its essential oil (B) and the its major compound α-Pinene (C) (Oliveira et al., 2019).**

Sesamum Indicum L. **(SESAME) SEED OIL**

Sesamum indicum L. (Sesame) herb is a member of *Pedaliaceae* family. It is an annual shrub with white bell-shaped flowers with a hint of blue, red or yellow with or without branches. It is grown around the world to produce seeds which are rich in high quality edible oil (Kandangath et al., 2010). Sesame seed oil is highly stable and rarely turns rancid in hot climates. It is rich in unsaturated fatty acids where the fatty acids composition is 14 % saturated, 39 % mono-unsaturated, and 46 % poly-unsaturated fatty acids (Kandangath et al., 2010). Sesame lignans are the main active ingredients in sesame seed oil and have strong antioxidant activity. Epidemiological studies have shown that sesame lignans have beneficial effects in regulating blood lipids and improving liver function. These properties are also responsible for the oxidative stability of sesame seed oil (Wei et al., 2022). Sesamin is the most abundant lignan in sesame seed oil accounting for 50% of the sesame lignans, with sesamolin, sesamol, and sesaminol accounting for a small proportion of the weight (Wu et al., 2017). Vasanthi and Rajavel, 2021 found out that 1000 ppm of sesamin registered about 97.59 % mortality in workers of the *Odontotermes obesus* (Agricultural fungus-growing termites) after 12 hours of treatment.

Figure 7: ***Sesamum indicum L.*** **(Sesame) herb (A), its seeds and oil (B) and sesamin structure (C) (Wei et al., 2022).**



THYMUS VULGARIS (THYME) ESSENTIAL OIL

Common thyme (*Thymus vulgaris L.*) is an aromatic perennial subshrub, native to the western Mediterranean region, belonging to *Lamiaceae* family (Marzec et al., 2010). The herb has received major attention as both a pharmaceutical and therapeutic agent across the globe. Reports indicate that the primary pharmacological effects

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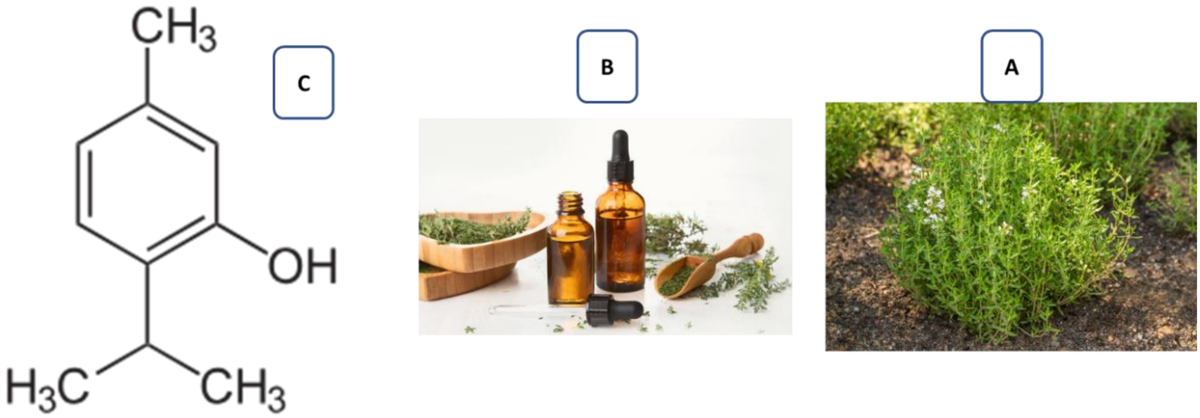
of thyme arise from thymol (5-Methyl-2-(propan-2-yl)phenol) at about 50 to 60 % w/w and carvacrol (2-Methyl-5-(propan-2-yl)phenol) at about 2 to 4 % w/w, which are the most important bioactive compounds that this plant contains in its essential oil (Gumus et al., 2017 and Szczepanik et al., 2012). *Thymus vulgaris* has been used for many centuries for its flavouring, culinary, and medicinal properties. In the Mediterranean region, it was used mainly as spice and then spread/sold all over the world (Dalal et al., 2022). Damtie and Mekonnen (2021), found out that 100 µL/L of *Thymus vulgaris* essential oil in acetone has 100 % Larvicidal Activity against the larvae of *Anopheles arabiensis* (Malaria causing) mosquito and that adult mosquitos fumigated with 50 µL/L *Thymus vulgaris* essential oil in acetone had 100 % mortality rate.

Figure 8: Common thyme (*Thymus vulgaris L.*) herb (A), its essential oil (B) and thymol structure (C) [53].



CONCLUSION

Synthetic insecticides have been established to be toxic to the environment, non-target organisms and even to human beings themselves when used to control problematic insects in the homes/gardens and in agricultural establishments. The use of a wide range of synthetic insecticides has been restricted around the world, due to the high cost, harmful environmental effects, their non-biodegradable nature, and increasing insecticidal resistance. This has led scientists to look more into the use of plant-based insecticides which have proven to be environmentally friendly and less toxic to non-target organisms and less detrimental to human health since they have short half-lives as they do not possess ‘unnatural’ ring structures and contain relatively few halogen substituents. Plant based oils such as Cinnamon, Garlic, Peppermint, Rosemary essential oils and Castor, Neem, Sesame seed oils have been found to contain secondary metabolites with effective insecticidal activities. These secondary metabolites include: cinnamaldehyde which has been found to kill common house flies; ricin which has been found to effectively control diamondback moth; dimethyl trisulphide and diallyl disulphide which have been found to effectively kill red flour beetle; azadirachtin and menthol which have strong insecticidal activity against malaria causing and dengue fever mosquitoes; α-Pinene which has been found to have repellence effects on aedes mosquitoes; sesamin with strong insecticidal activity against agricultural fungus-growing termites and thymol with strong larvicidal activities against malaria causing mosquitoes. This means that selected plant essential and seed oils or their derivatives can be used to control insects both in the homes and in agricultural environments. When these are used in agricultural environment, it means that farmers can produce high quality organic foods which are not only good for human health but does not destroy the natural environment when they are produced in.

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