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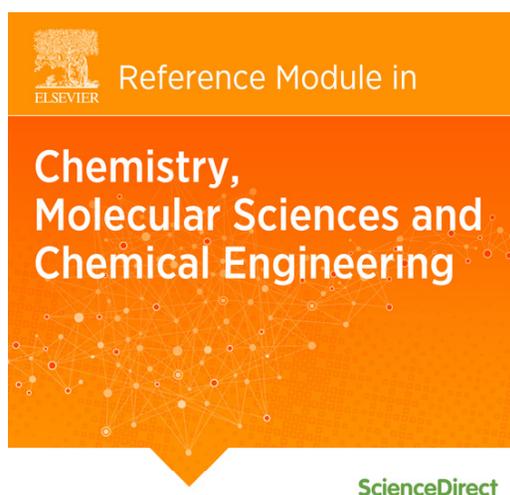
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Natural Product-Based Biopesticides for Insect Control[☆]

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Introduction

The direct use of natural products as pesticides or as leads for pesticides has been reviewed on previous occasions.¹⁻⁷

This short review will highlight methods and strategies and the rationale behind the use of natural products as insecticides with a more detailed discussion of new promising leads including a few examples from the authors' research.

The use of botanical insecticides dates back two millennia. The use of plant products in Europe goes back more than 150 years, until the discovery of synthetic insecticides (organochlorines, organophosphates, carbamates, pyrethroids), which replaced the botanical sort. Overuse of these synthetic insecticides has led to problems such as acute and chronic pollution, negative effects on wildlife (fish, birds), biological control and pollination, groundwater contamination, and resistance to pesticides.^{3,8}

[☆]*Change History:* June 2013. A Gonzalez, M Reina, CE Diaz, O Santana updated the text and further readings to this entire article.

Despite the concerted effort being made to breed or engineer plants with increased resistance to pests and disease, there will always be a need for crop protection; partly for mass-produced crops and partly for niche areas such as horticulture, greenhouses, organic farming, households, and gardens where biopesticides are particularly prevalent. There is a need for environmentally and consumer-friendly products which also preferably exhibit novel modes of action to mitigate resistance problems.

The development of crop protectants is similar to drug development and is presently based on synthesizing novel molecules which interact with well-defined targets found in the pest. The difference with drug development is that the compounds will be used on a large scale, must be free of all environmental toxicity, the products should be relatively stable and should be safe for human use (e.g. non-toxic, rapid breakdown). Toxicity is the major hurdle which needs to be overcome in the development of novel pesticides. Most compounds are eliminated due to adverse toxic effects.

Plant-Based Biopesticides

The documented use of plant extracts and powdered plant parts as insecticides goes back at least as far as the Roman Empire. There are reports of the use of pyrethrum (*Tanacetum cinerariaefolium*, Asteraceae) as early as 400 B.C. The first pure botanical insecticide used as such dates back to the 17th Century when it was shown that nicotine obtained from tobacco leaves was lethal to plum beetles. Around 1850 a new plant insecticide known as rotenone was introduced. Rotenone is a flavonoid derivative extracted from the roots of two different *Derris* spp. (Fabaceae) and *Lonchocarpus* spp. (Fabaceae). The ground seeds of Sabadilla, a plant of South American origin known as *Schoenocaulon officinale* (Liliaceae), are one of the plant insecticides exhibiting the least toxicity to mammals.⁹

The discovery of new natural crop protectants needs a systematic approach to finding new plant-derived products. Screening non-toxic plants for activity reduces the risk of discovering toxic biopesticides. Different sources can be considered including rare and traditionally used plants, fungal endophytes or agricultural wastes. The search for plant-based crop protectants depends to a great extent on the biodiversity and natural resources available locally or globally and involves International Cooperation in many cases. Therefore, the fair and equitable sharing of the benefits arising out of the utilization of genetic resources as described in the Nagoya Protocol on Access to Genetic Resources and the Convention on Biological Diversity should be followed.

The search for botanical biopesticides requires the screening of naturally occurring bioactive compounds in plants.³ The main groups of active compounds comprise phenylpropanoids and phenolics, terpenoids and steroids, alkaloids and nitrogenated compounds. The extracts from these plants need to be screened for activity and their active molecules characterized. Unlike conventional insecticides that are based on a single ingredient, plant-derived insecticides include an array of several compounds that decrease the chance of pests to develop resistance.¹⁰

Fungal Endophyte-Based Biopesticides

Endophytes live in the inter-cellular spaces of plants and produce bioactive substances involved in the endophyte-host relationship. The endophytes produce secondary plant metabolites which are used for plant defense by their host. They thus form a sustainable source of chemical diversity.¹¹ As such endophytes represent a potential biotechnological tool for production of extracts containing bioactive compounds, either different or similar to the ones in the host plant. Sometimes the same compound is detected in the plant extract and the endophyte. For example taxol is produced by *Taxus brevifolia*, *Taxus cuspidata* and *Taxus baccata* and it is detected in the endophytes associated with *Taxus*.^{12,13} Therefore the presence of bioactive compounds is an important factor in the selection of the host plant.^{14,15} There are several international patents on natural products of endophyte origin. The taxol producing strains, isolated from *Taxus* spp., are a well known example. However, no commercial products of endophyte origin are available for crop protection yet.

Waste-Based Biopesticides

Billions of metric tons of biomass are generated every year from the agricultural industry worldwide including liquid, solid and gaseous residues that may be considered one of the most abundant, cheap and renewable resources on earth. These residues can cause pollution and environmental problems when not properly managed. Worldwide between 10 and 60% of the agro-industrial production including crop- and processing-based residues are wasted yearly. In this regard, more than 250 MMT residues from the vegetable industry are disposed-off every year as reported by FAOSTAT¹⁶ on the production of the major crop commodities. For the industry, these organic wastes have been considered worthless substances requiring its management according to existing regulations, specifically Law 22/2011 of waste and contaminated soils of July 28 which transposes some of the provisions of Directive 2008/98/CE on waste. This management represents an estimated cost to the industry of 70 € per ton. Another problem is the CO₂ emission generated by their transport. For each 120 000 tonnes of waste produced an estimated 1 000 tons of CO₂ are emitted per year. There are currently no viable, nor sustainable, management solutions for centralization and valorization of biowastes in Europe.

A more efficient use of agricultural residues to yield a number of value added resources is highly attractive to ensure sustainable and cleaner production processes that are economically viable, environmentally sound and socially beneficial.^{17,18} Residues from crop production such as citrus peels are being used to obtain limonene or essential oils to control mosquitoes.⁶⁵³ Therefore,

agricultural wastes and specifically essential oil production wastes can be important, renewable and cheap sources of valuable compounds, however, little is known on their use as a source of natural crop protectants.¹⁷

The Challenge of Biopesticides

Natural Crop Protectants are needed in an increasingly restrictive pesticide market. The challenge of new and more stringent chemical pesticide regulations, combined with increasing demand for agriculture products with positive environmental and safety profiles, is boosting interest in natural crop protectants.¹⁹ A recent market analysis by Frost & Sullivan (<http://www.chemicals.frost.com>): North American & Western European Biopesticides Market (microbial, botanicals and biocontrol) showed \$594.2 millions of benefits in 2008, estimating \$1020.2 millions by 2015.

Cultivation and biomass production methods are needed. Environmentally-friendly extraction methods should be applied such as supercritical extraction²⁰ as well as efficient formulation processes.²¹ The successful development of biocides from discarded citrus peels in the United States is an excellent example of how such an approach can work.

The main markets for botanical pesticides are organic agriculture, horticulture, green houses, parks, gardens and households. Organic agriculture is a market with a high demand for biopesticides as organic growers cannot use conventional agrochemicals. This market is currently expanding owing to consumers' demand for improved Food Safety and the environmental problems associated with the use of synthetic pesticides. With an annual average growth of 30%, organic farming in the EU is one of the most dynamic agricultural sectors. Many more farmers have come on board since the enactment of Community Legislation regulating organic production (Council Regulation 2092/91/EEC of 24 June 1991). One of the overarching objectives of the Common Agricultural Policy (CAP) is the achievement of sustainable agricultural production in Europe which requires environmentally-friendly pest control measures.

Botanical pesticides also feature the advantage by being compatible with other low-risk options which are acceptable for insect management which include, inter alia, the use of pheromones, oils, detergents, entomopathogenic fungi, predators and parasitoids, significantly increase the likelihood of being integrated into IPM programs.

However, only a handful of botanical insecticides are in use today on commercially significant vegetable and fruit crops. In this chapter we will not review plant products currently in use (see recent reviews on this topic^{1,3}) but will rather discuss new sources and trends for future use and potential commercialization.

Commercial Insecticides of Plant Origin

4-Allyl-2-Methoxyphenol (Eugenol)

It is found in a wide range of plants including laurel (*Laurus* species) and in clove oil. Clove oil is predominantly composed of 4-allyl-2-methoxyphenol, but also contains a small amount of acetyl 4-allyl-2-methoxyphenol and other congeners such as methyl-eugenol. 4-Allyl-2-methoxyphenol and its derivatives are strong deterrent for most insect species, although in a few cases it can be an attractant.^{22,23} It is sold by a large number of different suppliers under many different trade names and is targeted at the home garden market. 4-Allyl-2-methoxyphenol is an irritant and should be used with care. Being a naturally occurring plant based phenolic, it is not expected to be hazardous to non-target organisms or to the environment.

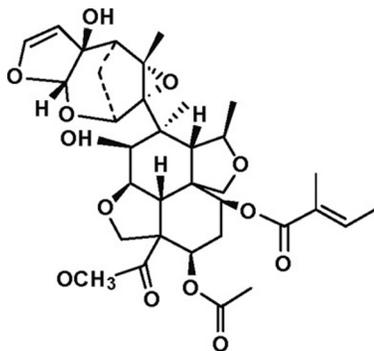
Azadirachtin/Dihydroazadirachtin

Azadirachtin is extracted from the neem tree (*Azadirachta indica* A. Juss). The tree is an attractive broad-leaved evergreen which is thought to have originated in Burma. It is now grown in the more arid subtropical and tropical zones of Southeast Asia, Africa, the Americas, Australia and the South Pacific Islands. The neem tree provides many useful compounds used as pesticides. The most significant neem limonoids are azadirachtin, salanin, meliantriol and nimbin (see Roy and Saraf).²⁴ Neem preparations containing azadirachtin are permitted in organic farming systems²⁵ and can be used in a wide range of crops, including vegetables (such as tomatoes, cabbage and potatoes), cotton, tea, tobacco, coffee, protected crops and ornamentals and in forestry. Azadirachtin acts as an Insect Growth Regulator and has several effects on phytophagous insects and is thought to disrupt insect moulting by antagonising the effects of ecdysteroids.²⁶ This effect is independent of feeding inhibition, which is another observed effect of the compound.^{1,27} The antifeedant/repellent effects are dramatic, with many insects avoiding treated crops, although other chemicals in the seed extract, such as salannin, have been shown to be responsible for these effects. Azadirachtin is sold by a large number of different companies as an EC under a wide range of trade names. Azadirachtin-based insecticides are widely used in India²⁸ and are increasingly popular in North America where they have found a place for garden use and in organic growing. Azadirachtin is considered to be non-toxic to mammals and is not expected to have any adverse effects on non-target organisms or the environment.^{1,26,29-32}

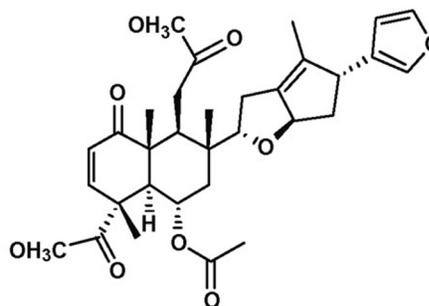
Dihydroazadirachtin is a reduced form of the naturally occurring azadirachtin obtained from the seed kernels of the neem tree. It is effective against a wide range of insect pests. The two compounds are functionally identical in their antipupation properties. Dihydroazadirachtin has both antifeedant and insect growth regulator properties. Products based on dihydroazadirachtin are not widely used outside the Indian subcontinent, although it is registered as a technical powder and an end-use product for indoor and outdoor use in the USA. Dihydroazadirachtin is of low mammalian toxicity, and risk to the environment is not expected because,

under approved use conditions, it is not persistent, is relatively short-lived in the environment and is metabolized by ubiquitous microorganisms in the soil and aquatic environments.¹

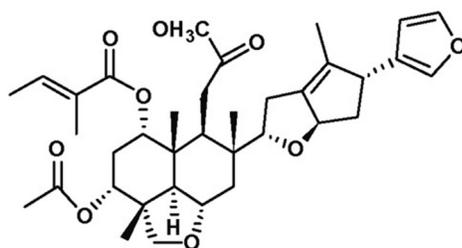
The toxicological data for neem-based preparations shows that the non-aqueous extracts appear to be the most toxic, the unprocessed materials, seed oil and the aqueous extracts being less toxic. For all preparations, a reversible effect on the reproductive capacity of both male and female mammals seems to be the most important toxic effect subsequent to sub-acute or chronic exposure.³³ This is the reason why an array of azadirachtin and neem extract-based insecticides and pesticides are available on the market today.



Azadirachtin



Nimbin

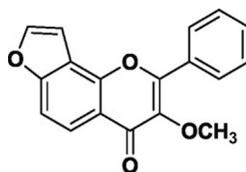


Salanin

Three commercial limonoid-based products (extracts of *A. indica*, *Melia toosendan*, and *M. azedarach*) have been approved in China for insect control in organic horticulture.³⁴

Karanjin

Karanjin is extracted from *Derris indica* (Lam.) Bennet [synonym *Pongamia pinnata* (L.) Pierre]. Karanjin is a potent deterrent to many different genera of insects and mites in a wide range of crops. Karanjin has a dramatic antifeedant/repellent effect, with many insects avoiding treated crops. It suppresses the effects of ecdysteroids and thereby acts as an insect growth regulator (IGR) and antifeedant. There are claims that it inhibits cytochrome P450 in susceptible insects and mites. Karanjin has not achieved wide acceptance as an insecticide. There is no evidence of allergic or other adverse effects and it is not expected that karanjin-based products will have any adverse effects on non-target organisms or on the environment.¹



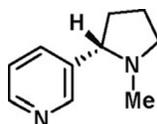
Karanjin

Ponneem

PONNEEM is a new biopesticidal formulation prepared using the combination of the seed oils of neem (*A. indica*) and karanj (*Pongamia glabra*) (Indian Patent No. 234081, <http://www.allindianpatents.com/patents/234081>). The combination of two oils at 1:1 ratio, containing bioactive molecules like azadirachtin and karanjin, gives synergistic effects in controlled lepidopteran pests.³⁵ PONNEEM is a strong antifeedant, larvicidal, ovicidal and growth inhibitor against polyphagous pest without affecting nontarget organism such as parasitoids.^{36,37}

Nicotine

Nicotine is the main bioactive component of the tobacco plants *Nicotiana tabacum* L., *N. glauca* Graham and, particularly, the species *N. rustica* L. It is also present in a number of other plants belonging to the families of Lycopodiaceae, Crassulaceae, Leguminosae, Chenopodiaceae and Compositae. The average nicotine content of the leaves of *N. tabacum* and *N. rustica* is 2–6% dry weight. It is used for the control of a wide range of insects, including aphids, thrips and whitefly, on protected ornamentals and field-grown crops, including orchard fruit, vines, vegetables and ornamentals.



Nicotine

It was once prepared from extracts of the tobacco plant but is now often obtained from waste of the tobacco industry, or it is synthesized. Nicotine is a non-systemic insecticide³⁸ that binds to the cholinergic acetylcholine nicotinic receptor (nACh) in the nerve cells of insects, leading to a continuous firing of this neuroreceptor.³⁹ Nicotine is one of the oldest known plant origin insecticides, which possesses the remarkable insecticidal activity⁴⁰ and has been used for many years as a fumigant for the control of many sucking insects. However, Nicotine is very toxic to humans by inhalation and by skin contact. It is toxic to birds, fish and other aquatic organisms, and is toxic to bees, but has a repellent effect. As a result, Nicotine is seldom used today in North America or Europe.²⁶ In the UK, nicotine is subject to regulation under the Poisons Act. The use of nicotine as a pesticide is banned in South Africa, severely restricted in Hungary and cancelled in Australia and New Zealand, as well as not being registered in numerous African, Asian and European countries.¹

Phenethyl Propionate

Phenethyl propionate is a component of peppermint (*Mentha piperita*) oil and is also used as an herbicide and as an insecticide/insect repellent and sold under a wide range of trade names in combination with other plant derived natural products (plus eugenol plus geraniol). The major use is in homes and gardens¹ and is thought to be very safe to the environment and to human health, as it is used in food flavorings.⁴¹

Plant-Derived Oils

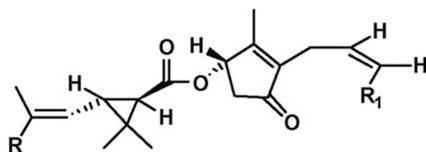
A wide range of plant oils are being sold for insect and mite control. Among these are canola oil, an edible refined vegetable oil obtained from the seeds of two species of rape plants (*Brassica napus* L. and *B. campestris* L.) of the family Cruciferae (mustard family), jojoba oil, derived from jojoba seeds, oleoresin, derived from *Capsicum* spp., oil of anise, soybean oil, eucalyptus oil (Bollcure®-India), rosemary oil distilled from wild and cultivated shrub *Rosmarinus officinalis*, thyme oil obtained by steam distillation of *Thymus* sp., clove oil from water distillation of dried flower buds of *Syzygium aromaticum* and capsaicin oil obtained from the genus *Capsicum*.^{1,41} More recently, hexa-hydroxyl, sold as a GR formulation containing 2.90% eugenol and 0.60% thyme oil as the active ingredients, and BugOil, made from the essential oils of three plant species, thyme (*Thymus vulgaris* L.), wintergreen (*Gaultheria procumbens* L.) and African marigold (*Tagetes erecta* L.), have been commercialized. Few of these oils have been fully characterized chemically. Various claims are made for the mode of action, including insect repellency caused by altering the outer layer of the leaf surface, acting as an insect irritant and preventing gas exchange (suffocation) and water loss by covering the insect's body.⁴² The potassium salts of plant oils (soft soaps) are also sold as insecticides under a wide range of trade names by many different manufacturers. Insecticidal soaps have not been chemically fully characterized and are contact insecticides, causing a breakdown of the target pest's cuticle, leading to dehydration and, ultimately, death. They cause the rapid knockdown of phytophagous insects, but, because they are broken down rapidly once sprayed, they will not prevent subsequent reinvasion. They are often used in conjunction with insect predators, being used to bring the populations down to manageable levels prior to release.¹

Plant-Derived Acids

A number of acids of plant origin are sold for insect control. These include citric acid, recommended for use against a wide range of insects, fatty acids (often oleic acid); and formic acid, used to control varroa (*Varroa destructor*) and tracheal mites in honey bees. The mode of action of citric acid is not identified with certainty. Formic acid is a severe irritant and acts by directly killing the mites while not disrupting bee behaviour or life span substantially. Oleic acid interferes with the cell membrane constituents of the target organism, leading to a breakdown of the integrity of the membrane and subsequent death.¹

Pyrethrins, Chrysanthemates and Pyrethrates

Pyrethrins, chrysanthemates and pyrethrates are extracted from the flower of *Tanacetum cinerariaefolium* (Trevisan). The extract is refined using methanol or supercritical carbon dioxide. The dried, powdered flower of *T. cinerariaefolium* has been used as an insecticide from ancient times. The species was identified in antiquity in China, and it spread to the west via Iran (Persia), probably via the Silk Routes in the Middle Age, known as 'Persian insect powder'.⁴³ Records of use date from the early nineteenth century when it was introduced to the Adriatic coastal regions of Croatia (Dalmatia) and some parts of the Caucasus. Subsequently, it was grown in France, the USA and Japan. Plants producing these compounds are now widely grown in East African countries, especially in Kenya (1930), in Ecuador, Papua New Guinea (1950) and in Australia (1980). The pyrethrins include pyrethrin I, cinerin I, jasmolin I, pyrethrin II, cinerin II and jasmolin II, and are recommended for control of insects and mites on fruit, vegetables, field crops, ornamentals, glasshouse crops and house plants.⁴¹ They have been shown to bind to and activate the voltage-sensitive sodium channels of nerve, heart and skeletal muscle cell membranes in insect nervous systems, prolonging their opening and thereby causing knockdown and death. They are non-systemic insecticides with contact action. Initial effects include paralysis, with death occurring later. They have some acaricidal activity.⁴⁴ They are approved for use as a broad-spectrum organic insecticide under many registered names.⁴¹ Pyrethrins have moderate mammalian toxicity, and there is no evidence that the addition of synergists increases this toxicity. The compounds show low toxicity to birds, but are highly toxic to fish and honey bees (although they exhibit a repellent effect to bees).¹ More recently, the synthetic derivatives of naturally occurring Pyrethrins (called Pyretroids) have been developed in order to improve the specificity and activity of Pyrethrins but most of them are more harmful to the environment and toxic to natural enemies, honey bees and fish.²⁶



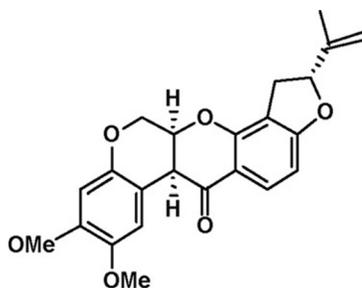
R = -CH₃ (chrysanthemates) or -CO₂CH₃ (pyrethrates)

R₁ = -CH=CH₂ (pyrethrin) or -CH₃ (cinerin) or -CH₂CH₃ (jasmolin)

Rotenone

Rotenone, also known as derris root, tuba-root, aker-tuba (for the plant extract) and barbasco, cube, haiari, nekoe and timbo (for the plants), is obtained from *Derris*, *Lonchocarpus* and *Tephrosia* species that were used originally in Asia and South America as fish poisons. The four major active ingredients are rotenone, deguelin, rotenolone, and tephrosin acting as inhibitors of NADH-ubiquinone oxidoreductase activity depending on the overall molecular configuration and the E-ring substituents.⁴⁵ Rotenone has a long history of use as a toxin for insect and a wide range of other arthropod pests and it is used to control aphids, suckers, thrips and other insects on fruit and vegetables.⁴⁶ It is an inhibitor of site I respiration within the electron-transport chain of susceptible insects and is a selective, non-systemic insecticide with contact and stomach action and secondary acaricidal activity.⁴⁷ Rotenone has been cleared for use in organic farming when insect pressure is very high. Rotenone has a high mammalian toxicity, with the estimated lethal dose for humans being 300–500mg kg⁻¹. It is more toxic when inhaled than when ingested and is very toxic to pigs. It is not toxic to bees, but combinations with pyrethrum are very toxic. It is very toxic to fish and must not be used near water courses.¹ Rotenone is commonly sold as a dust containing 1–5% active ingredients for home and garden use but liquid formulations used in organic agriculture can contain as much as 8% rotenone and 15% total rotenoids.³ Products containing rotenone are commercialized as broad-spectrum insecticides under many trade names.⁴¹

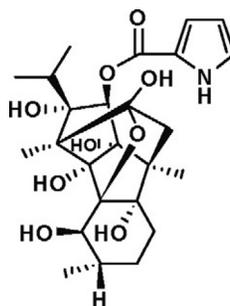
Rotenone is used in European organic agriculture nowadays, with a strong restriction regarding its environmental hazards. The use of rotenone is partially restricted in Austria, Italy, Spain, Switzerland, and the United Kingdom, but not in Denmark, Netherlands, Portugal, and Slovenia. In the United Kingdom, few private standard-setting organizations allow its use after preliminary permission, while others never permit its use. In Italy the use of rotenone formulations was permitted until April 30, 2011 on apples, peaches, pears and cherries.⁴⁶



Rotenone

Ryania Extract

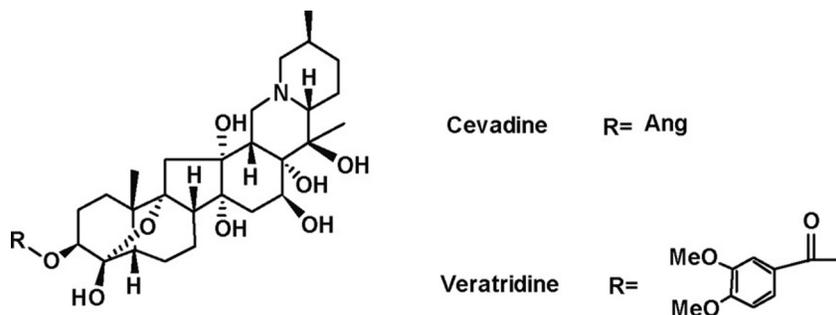
The alkaloids from the stem of *Ryania* species, particularly *R. speciosa* Vahl, represent the first successful discovery of a natural insecticide. The collaboration between Rutgers University and Merck in the early 1940s followed the lead from the use of *Ryania* species in South America for euthanasia and as rat poisons. This collaborative work revealed that *Ryania* alkaloid extracts were insecticidal. Ryanodine and 9,21-dehydroryanodine, are the main active ingredients of the botanical insecticide Ryania, and affect muscles by binding to the calcium channels in the sarcoplasmic reticulum. This causes calcium ion flow into the cells, and death follows very rapidly (see Sutko et al.⁴⁸). *Ryania* extracts have had limited use as insecticides, but they do give effective control of selected species. The size and complexity of the natural compound means that it can be used economically only to treat infested crops, and it has no systemic activity. The rapidity of its effect is an advantage in the control of boring insects. The ryanodine site is similarly sensitive in insects and mammals and a large series of analogs was prepared, but high insecticidal potency combined with low mammalian toxicity was not adequately achieved.⁴⁹ In this connection, more recently a new class of insecticides has been discovered that provides exceptional control through action on a novel target, the ryanodine receptor, e.g. Rynaxypyr™, anthranilic diamides and substituted phthalic acid diamides with potent insecticidal activity. These new class of insecticides (e.g. the diamides flubendiamide, chlorantraniliprole and cyantraniliprole) has been classified into new group as a ryanodine receptor modulators.^{49,50} These substances activate ryanodine-sensitive intracellular calcium release channels in insects.⁵¹⁻⁵⁴ *Ryania* extracts are moderately toxic to mammals, but very toxic to fish.



Ryanodine

Sabadilla

Sabadilla is an insecticidal preparation from the crushed seeds of the liliaceous plant, *Schoenocaulon officinale* Gray (formerly *Veratrum sabadilla* Retr.), which was used by native people of South and Central America as an insecticide for many years.⁴¹ Sabadilla has been used commercially since the 1970s. The seeds of *S. officinale* contain a mixture of alkaloids (veratrine) consisting of an approximately 2:1 mixture of cevadine and veratridine, in combination with many minor components, all of which are esters of the alkaline veracine. The product is produced by grinding the seeds of the plant and subsequent concentration. The seeds contain between 2 and 4% alkaloids. Cevadine, veratridine and related ceveratrum alkaloids have a mode of action that is similar to that of the pyrethrins in that they activate the voltage-sensitive sodium channels of nerve, heart and skeletal muscle cell membranes, although the binding site appears to be different from that of the pyrethroids.⁵⁵



They are non-systemic insecticides with contact action. Initial effects include paralysis, with death occurring later.¹ Sabadilla powder is not used widely in crop protection, but it is approved for use in organic farming systems and also has been approved for use in the USA as an organic insecticide, as well as for other uses, by the Organic Materials Review Institute (OMRI).⁵⁵ Sabadilla is considered among the least toxic of botanical insecticides, with an oral LD50 of 4000–5000 mg/kg. Sabadilla is effective by either contact or ingestion and has been effective against caterpillars, leaf hoppers, thrips, stink bugs and squash bugs.⁴¹ This powder has a low mammalian toxicity, but it is an irritant to mucous membranes. Sabadilla powder is not active against beneficial insects and may be used in insect control strategies that use them.⁵⁶

Starch Syrup

A new insecticide prepared from reduced starch syrup has just been made available by Kyoyu Agri. It is sold under the trade name YE-621 and works by obstructing the spiracles of insect pests, causing suffocation. YE-621 is potentially effective against insect pests that are resistant to chemical-based insecticides. It is nontoxic to humans and beneficial insects and/or natural predators. The main component of YE-621 is starch syrup mainly from corn and potatoes.^{1,57}

New Insecticide Sources

Plant Essential Oils

Plant essential oils (EOs) are produced commercially from cultivated plants mainly from the Lamiaceae family. EOs are complex mixtures of monoterpenes, sesquiterpenes and aromatic compounds. Steam distillation of aromatic plants yields EOs used in perfumery, traditional medicine, pharmaceutical preparations, herbal beverages and as natural flavorings.^{3,58}

Since the middle ages, essential oils have been widely used for bactericidal, virucidal, fungicidal, antiparasitical, insecticidal, medicinal and cosmetic applications and today are particularly vital to the pharmaceutical, health, cosmetic, agricultural, and food industries. While in-vitro physicochemical assays characterize most of these as antioxidants, recent work shows that in eukaryotic cells essential oils can act as prooxidants affecting inner cell membranes and organelles such as mitochondria. Depending on type and concentration, they exhibit cytotoxic effects on living cells but are usually non-genotoxic.⁵⁹ Plant EOs and their components have low mammalian toxicity but not all compounds found in plant essential oils are safe. Estragole and (+)-fenchone found in the essential oil of *Foeniculum vulgare* which are highly effective against *Sitophilus oryzae*, *Callosobruchus chinensis* and *Lasioderma serricorne* adults are known to be carcinogenic.⁶⁰ Similarly, safrole and β -asarone have been included on the list of carcinogenic compounds.

Plant essential oils play an important role in pest control of field, greenhouse and stored crops and have been used as alternative to many synthetic pesticides.^{61–64} EOs are considered minimum-risk biopesticides and are exempt from Environmental Protection Agency (EPA) registration under section 25(b) of the Federal Insecticide Fungicide and Rodenticide Act. (FIFRA, <http://www.epa.gov/agriculture/lfra.html>). In addition, their mammalian toxicity is low and environmental persistence is short.⁶⁵

Some aromatic plants have been traditionally used for the protection of stored commodities due to their fumigant and contact toxicity effects. Fumigant toxicity tests conducted with essential oils of plants (mainly belonging to Apiaceae, Lamiaceae, Lauraceae and Myrtaceae) and their components (cyanohydrins, monoterpenoids, sulphur compounds, thiocyanates and others) have largely focused on beetle pests such as *Tribolium castaneum*, *Rhyzopertha dominica*, *Sitophilus oryzae* and *Sitophilus zeamais*.⁶ Promising results have been obtained from a few EOs tested as repellents against head lice, *Pediculus humanus capitis* (Phthiraptera: Pediculidae), an ectoparasite preying on humans which causes pediculosis capitis, although in-vitro tests and clinical trials often produce contradictory results. A handful of fixed extracts and several EOs and their individual components have also been tested as contact pediculicides or fumigants.⁶⁶

There is also renewed interest in the use of EOs as anti-malarials in the form of biocidal (insect repellent) preparations against mosquitos and other arthropod vectors of human tropical diseases such as the common houseflies (*Musca domestica*), American and German cockroaches (*Periplaneta americana*, *Blattella germanica*), and oriental latrine/blowflies (*Chrysomya megacephala*) as well as biting, blood-sucking arthropods such as blackflies (*Simulium* spp.), fleas (*Xenopsylla cheopis*), kissing bugs (*Rhodnius* spp., *Triatoma infestans*), body and head lice (*Pediculus humanus humanus*, *P. humanus capitis*), sandflies (*Lutzomyia longipalpis*, *Phlebotomus* spp.), scabies mites (*Sarcoptes scabiei*, *S. scabiei* var *hominis*, *S. scabiei* var *canis*, *S. scabiei* var *suis*), and ticks (*Ixodes*, *Amblyomma*, *Dermacentor*, and *Rhipicephalus* spp.).^{67–70}

The swift results obtained from some of these oils suggest neurotoxic action. There is evidence of some common oil components such as thujone,⁷¹ thymol,⁷² menthol and borneol⁷³ interfering with the octopamine receptor^{74,75} and GABA-gated chloride channels.⁷⁶ Moreover, several reports indicate that monoterpenoids raise insect mortality by inhibiting acetylcholinesterase enzyme (AChE) activity (see Rajendran and Srinanjini⁶ and Houghton et al.,⁷⁷). However, it has been shown that the insecticidal effects of some EOs cannot be explained by the action of their major components suggesting that their insecticidal action is the result of a synergistic effect.^{78–80}

Variations in the composition of essential oils due to factors such as seasonal fluctuations, differences in the region of origin, extraction method used (steam or hydro distillation, solvent extraction and maceration, supercritical extraction with CO₂) and the plant part used for extraction have been reported.^{20,79–82} Therefore, careful attention should be paid to the presence of oil chemotypes for a given plant species.

Since EOs can often be extracted from cultivated plants, are readily available and do not require further purification, there is an increasing interest in the study of their insecticidal effects and other properties. **Table 1** shows the publications on this topic for the

Table 1 Insecticidal essential oils (EOs) for the period 2006–13

<i>Plant species</i>	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
<i>Acanthospermum hispidum</i>	<i>Spodoptera frugiperda</i>	Behavioral disorders	83
<i>Achillea biebersteinii</i>	<i>Sitophilus granarius</i> , <i>Tribolium confusum</i>	Fumigant toxicity	84
<i>A. wilhelmsii</i> ,	<i>Callosobruchus maculatus</i>	Fumigant toxicity	85
<i>Acorus gramineus</i>	<i>Lycoriella ingenua</i>	Toxic	86
<i>A. calamus</i>	<i>Spodoptera litura</i>	Toxic	87,88
	<i>Sitophilus oryzae</i>	Adulticidal	
<i>Aframomum latifolium</i>	<i>Bemisia tabaci</i>	Fumigant toxicity	89
<i>Ageratum conyzoides</i>	<i>Tribolium castaneum</i>	Toxic	90
<i>Aeollanthus pubescens</i>	<i>Hypothenemus hampei</i>	Adulticidal	91
<i>Allium cepa</i>	<i>Haematopinus tuberculatus</i>	Pediculicidal	92
<i>A. sativum</i>	<i>Lycoriella ingenua</i>	Toxic	93–95
	<i>Camptomyia corticalis</i>	Toxic	
	<i>Trichoplusia ni</i>		
<i>A. victoralis</i>	<i>Aedes aegypti</i>	Toxic	96
<i>Aloysia polystachya</i> , <i>A. citriodora</i>	<i>Rhizopertha dominica</i>	Fumigant toxicity, Repellent	97
<i>Amethystea caerulea</i>	<i>Drosophila melanogaster</i>	Contact toxicity	98
<i>Alpinia calcarata</i>	<i>Callosobruchus maculatus</i>	Fumigant toxicity and Repellent	99–101
<i>A. conchigera</i>	<i>Sitophilus zeamais</i> , <i>Tribolium castaneum</i>	Contact toxicity, Antifeedant, repellent	
<i>A. speciosa</i>			
	<i>Aedes aegypti</i>	Larvicidal	
<i>Anethum graveolens</i>	<i>Callosobruchus chinensis</i> , <i>Trachyspermum ammi</i>	Fumigant, oviposition Deterrence, antitermitic	102,103
<i>Apium graveolens</i>	<i>Aedes aegypti</i>	Adulticidal Oviposition deterrent, Ovicidal	104,105
<i>Armoracia rusticana</i>	<i>Lycoriella ingenua</i>	Toxic	95
<i>Artemisia annua</i>	<i>Tribolium castaneum</i>	Fumigant toxicity, Repellent	106
<i>A. herba-alba</i> , <i>A. monosperma</i>	<i>Bemisia tabaci</i> , <i>Aphis gossypii</i> , <i>Thrips tabaci</i> , <i>Acanthoscelides obtetus</i>	Toxic	107,108
<i>A. sieberi</i>	<i>Callosobruchus maculatus</i> , <i>Sitophilus oryzae</i> , <i>Tribolium castaneum</i>	Fumigant toxicity	109
<i>A. vulgaris</i>	<i>Thrips palmi</i> , <i>Tribolium castaneum</i> , <i>Aedes aegypti</i> , <i>Camptomyia corticalis</i>	Repellent, fumigant Toxicity, Larvicidal, toxic	93,110–112
<i>A. princeps</i>	<i>Sitophilus oryzae</i> , <i>Bruchus rugimanus</i> , <i>Aedes aegypti</i> , <i>Anopheles stephensi</i>	Fumigant toxicity, Larvicidal	113
<i>A. nilagirica</i>	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Larvicidal	114
<i>A. giraldii</i> , <i>A. subdigitata</i> , <i>A. eriopoda</i> , <i>A. lavandulaefolia</i> , <i>A. sieversiana</i> , <i>A. capillaries</i> , <i>A. mongolica</i> , <i>A. spicigera</i> , <i>A. herba-alba</i> , <i>A. pontica</i>	<i>Sitophilus zeamais</i>	Fumigant, contact Toxicity	115–118
<i>A. absinthium</i>	<i>Dendroctonus micans</i>	Toxic	107,119–121
	<i>Acanthoscelides obtectus</i>	Toxic	
	<i>Spodoptera littoralis</i> , <i>Myzus persicae</i> , <i>Rhopalosiphum padi</i>	Antifeedant	
<i>A. judaica</i>	<i>Spodoptera littoralis</i>	Antifeedant	
<i>Athamanta haynaldii</i>	Gypsy moth larvae	Larvicidal, antifeedant	122
<i>Azadirachta indica</i>	<i>Aedes aegypti</i> , <i>Culex quinquefasciatus</i> , <i>Anopheles stephensi</i>	Larvicidal	654
<i>Baccharis salicifolia</i>	<i>Spodoptera littoralis</i>	Post-ingestive toxicity	123
<i>Blumea mollis</i>	<i>Culex quinquefasciatus</i>	Toxic	124

(Continued)

Table 1 (Continued)

Plant species	Target insect	Action	Reference	
<i>Callistemon lanceolatus</i>	<i>Callosobruchus chinensis</i>	Repellent	125	
<i>Carum carvi</i>	<i>Aedes aegypti</i>	Adulticidal	104	
	<i>Lycoriella ingenua</i>	Fumigant toxicity	126	
	<i>Reticulitermes speratus</i>	Contact toxicity	103	
	<i>Sitophilus zeamais</i> , <i>Tribolium castaneum</i>	Repellency, toxicity	127	
	<i>Meligethes aeneus</i>	Toxicity	128	
	<i>Camptomyia corticalis</i>	Toxic	93	
	<i>Blattella germanica</i>	Fumigant, toxicity	103,129	
	<i>Trachyspermum ammi</i>	Fumigant, antitermitic		
	<i>Callosobruchus maculatus</i>	Toxicity	130	
	<i>Culex pipiens</i>	Larvicidal	131,132	
	<i>Plutella xylostella</i>	Larvicidal		
	<i>Aedes aegypti</i> , <i>A. albopictus</i>	Larvicidal	133	
	<i>Chenopodium ambrosioides</i>	<i>Lycoriella ingenua</i>	Toxic	86
<i>Pseudococcus longispinus</i> , <i>Bradysia</i> spp.		Toxic	134	
<i>Anopheles arabiensis</i> , <i>Aedes aegypti</i>		Larvicidal	135	
<i>Sitophilus zeamais</i>		Fumigant, contact Toxicity Fumigant	136,137	
<i>Blattella germanica</i>		Fumigant, contact Toxicity	138	
<i>Trogoderma granarium</i> , <i>Tribolium castaneum</i>		Toxic, F1 reduction	139,140	
<i>Callosobruchus chinensis</i> , <i>C. maculatus</i>		Ovicidal, antifeedant		
<i>Chloroxylon swietenia</i>		<i>Helicoverpa armigera</i>	Antifeedant	141,142
		<i>Anopheles gambiae</i> , <i>Culex quinquefasciatus</i> , <i>Aedes aegypti</i>	Fumigant toxicity	
		<i>Spodoptera litura</i>	Toxic	
<i>Cinnamomum camphora</i>		<i>Resseliella oculiperda</i>	Repellent	93,113,143,144,144–147
		<i>Sitophilus oryzae</i> , <i>Bruchus rugimanus</i>	Repellent	
<i>C. zeylanicum</i>		<i>Musca domestica</i>		
<i>C. osmophloeum</i>	<i>Camptomyia corticalis</i>	Knock down and Mortality		
	<i>Armigeres subalbatus</i>	Toxic		
<i>C. cassia</i>	<i>Chrysomya megacephala</i>	Larvicidal		
	<i>Trichoplusia ni</i>	Ovicidal		
<i>C. glanduliferum</i>	<i>Camptomyia corticalis</i>	Contact, residual, Fumigant toxicity		
	<i>Trichoplusia ni</i>	Toxic Contact, residual, Fumigant toxicity		
<i>Chrysanthemum coronarium</i>	<i>Culex pipiens</i>	Larvicidal	148	
<i>Citrus reticulata</i>	<i>Lycoriella ingenua</i>	Toxic	126	
	<i>Aedes aegypti</i>	Larvicidal	149	
	<i>Callosobruchus maculatus</i>	Fumigant	150	
<i>C. sinensis</i>	<i>Musca domestica</i>	Contact toxicity	151	
		Fumigant, contact Toxicity	152	
	<i>Aedes albopictus</i>	Larvicidal	153	
	<i>Sternechus subsignatus</i> , <i>Rhyssomatus subtilis</i>	Contact toxicity	154	
	<i>Bemisia tabaci</i>	Fumigant, oviposition Repellent	155	
		<i>Culex pipiens</i>	Larvicidal	156
<i>C. limon</i>	<i>Culex pipiens</i>	Larvicidal	156	
	<i>Aedes albopictus</i>	Larvicidal	153	
	<i>Sternechus subsignatus</i> , <i>Rhyssomatus subtilis</i>	Contact toxicity	154	
		<i>Callosobruchus maculatus</i>	Fumigant	150
<i>C. paradise</i>	<i>Aedes albopictus</i>	Larvicidal	153	

Table 1 (Continued)

Plant species	Target insect	Action	Reference
<i>C. aurantium</i>	<i>Callosobruchus maculatus</i>	Fumigant	150
	<i>Culex pipiens</i>	Larvicidal	156
	<i>Spodoptera frugiperda</i>	Antifeedant	157
	<i>Bemisia tabaci</i>	Fumigant, oviposition Repellent	155
	<i>Musca domestica</i>	Contact toxicity	151
<i>C. aurantium</i> subsp. <i>bergamia</i>	<i>Spodoptera frugiperda</i>	Toxic, antifeedant	157,158
	<i>Sitophilus zeamais</i>	Repellent	
	<i>Culex pipiens</i>	Larvicidal	159
<i>C. hystrix</i>	<i>Spodoptera litura</i>	Contact toxicity, Larvicidal	160
	<i>Aedes aegypti</i>	Larvicidal	149
<i>Clausena excavate</i> , <i>C. anisata</i>	<i>Aedes albopictus</i> , <i>Acanthoscelides obtectus</i>	Larvicidal, fumigant	140,161,162
<i>C. pentaphylla</i>	<i>Callosobruchus chinensis</i> , <i>C. maculatus</i>	Repellent	
<i>Coleus aromaticus</i>	<i>Tribolium castaneum</i>	Toxic	163
<i>Coriandrum sativum</i>	<i>Thrips palmi</i>	Fumigant toxicity	112
	<i>Aedes aegypti</i>	Larvicidal	164
	<i>Camptomyia corticalis</i>	Fumigant toxicity	93
<i>Croton nepetaefolius</i> , <i>C. argyrophyloides</i> , <i>C. sonderianus</i> , <i>C. zehntneri</i>	<i>Aedes aegypti</i>	Larvicidal	165–167
		Larvicidal, ovicidal, Pupicidal	
<i>C. pulegioidorus</i> , <i>C. heliotropiifolius</i>	<i>Aedes aegypti</i>	Larvicidal	168
<i>C. regelianus</i>	<i>Aedes aegypti</i>	Larvicidal	169
<i>Cryptomeria japonica</i>	<i>Lepisma saccharina</i>	Repellent and insecticide	170
	<i>Aedes aegypti</i> , <i>A. albopictus</i>	Larvicidal	171
<i>Cuminum cyminum</i>	<i>Coptotermes formosanus</i>	Antitermitic	172
	<i>Tribolium castaneum</i>	Fumigant toxicity	173
	<i>Callosobruchus chinensis</i>	Fumigant toxicity, Pupicidal, reduced egg Hatching, F1 progeny	102
	<i>Lycoriella ingenua</i>	Toxic	126
	<i>Blattella germanica</i>	Fumigant toxicity	129
<i>Cupressus sempervirens</i>	<i>Callosobruchus maculatus</i>	Fumigant toxicity	174
	<i>Aedes aegypti</i>	Adulticidal	104
	<i>Thrips palmi</i>	Fumigant toxicity	112
<i>C. arizonica</i>	<i>Anopheles stephensi</i>	Larvicidal	175
<i>Curcuma zedoaria</i>	<i>Aedes aegypti</i>	Adulticidal	104
		Larvicidal	176
	<i>Anopheles dirus</i>	Larvicidal	177
<i>C. longa</i>	<i>Sitophilus zeamais</i>	Contact toxicity	101
	wild mosquitoes, anthrophilic black flies	Repellent	178
	<i>Aedes aegypti</i>	Larvicidal	179
<i>C. wenyujin</i>	<i>Liposcelis bostrychophila</i>	Contact toxicity	180
<i>Cymbopogon citratus</i>	<i>Lycoriella ingenua</i>	Toxic	126
	<i>Aedes aegypti</i>	Larvicidal	100
	<i>Tribolium castaneum</i>	repellent	181
	<i>Sitophilus oryzae</i>	Contact toxicity	182
	<i>T.castaneum</i> , <i>S. oryzae</i>	Antifeedant and contact Toxicity (<i>S. oryzae</i>), Repellency, post- Ingestive toxicity	144,183
	<i>Trichoplusia ni</i>	Antifeedant	
	<i>A. aegypti</i> , <i>Anopheles dirus</i> , <i>Culex quinquefasciatus</i>	Oviposition deterrent, Ovicidal	184
<i>Trichoplusia ni</i>	Residual toxicity, Antifeedant	144	

(Continued)

Table 1 (Continued)

Plant species	Target insect	Action	Reference
	<i>Camptomyia corticalis</i>	Toxic	93
	<i>Zabrotes subfasciatus</i>	Repellent, ovicidal	185
	<i>Musca domestica</i>	Fumigant, contact toxicity	186
<i>C. martini</i>	<i>Callosobruchus chinensis</i> , <i>T. castaneum</i>	Repellent	187
	<i>Callosobruchus maculatus</i>	Toxic	188
	<i>Rhipicephalus microplus</i>	Ovicidal, larvicidal	189
<i>C. schoenanthus</i>	<i>C. maculatus</i>	Toxic	190
<i>C. nardus</i>	<i>Musca domestica</i>	Knock down and mortality	145
	<i>C. quinquefasciatus</i> , <i>Anopheles minimus</i>	Larvicidal, pupacidal	191
	<i>Sitophilus zeamais</i>	Repellency	192
	<i>A. aegypti</i>	Ovicidal	105
	<i>A. aegypti</i> , <i>Anopheles dirus</i> , <i>C. quinquefasciatus</i>	Oviposition deterrent, Ovicidal	184
	<i>Trichoplusia ni</i>	Residual toxicity, antifeedant	144
<i>C. spp.</i>	<i>Ixodes ricinus</i>	Repellent	193
<i>C. flexuosus</i>	<i>A. aegypti</i>	Larvicidal	110
<i>C. winterianus</i>	<i>Myzus persicae</i>	Contact toxicity	194
<i>Digitalis purpurea</i>	wild mosquitoes	Repellent	178
	anthropophilic black flies		
<i>Dorystoechas hastata</i>	<i>Rhipicephalus turanicus</i>	Toxic	195
<i>Eucalyptus grandis</i> ,	<i>Aedes aegypti</i> larvae, <i>Callosobruchus</i>	Larvicidal	63,196–198
<i>E. intertexta</i> , <i>E. sargentii</i> ,	<i>maculatus</i> , <i>Sitophilus oryzae</i> ,	Fumigant toxicity	
<i>E. camaldulensis</i>	<i>Tribolium castaneum</i>		
	<i>Anopheles stephensi</i>		
<i>E. camaldulensis</i>	<i>Aedes aegypti</i> , <i>A. albopictus</i>	Larvicidal	199
	<i>Anopheles stephensi</i>	Larvicidal	200
<i>E. tereticornis</i> <i>E. cinerea</i> , <i>E. viminalis</i>	<i>Pediculus humanus capitis</i>	Larvicidal, adulticidal	201
<i>E. viminalis</i>	<i>Aedes aegypti</i>	Knockdown	63
	<i>Alphitobius diaperinus</i>	Toxic	202
<i>E. smithii</i>	<i>Lycoriella ingenua</i>	Repellent	86
<i>E. dunni</i> , <i>E. saligna</i> , <i>E. benthamii</i>	<i>Sitophilus zeamais</i>	Toxic, repellent	203
<i>E. sideroxylon</i>	<i>Pediculus humanus capitis</i>	Knockdown	204
<i>E. globulus</i>	<i>Lycoriella ingenua</i>	Repellent	86
	<i>Pediculus humanus capitis</i>	Contact toxicity, knockdown	204,205
	<i>Lasioderma serricorne</i> , <i>Rhyzopertha dominica</i>	Fumigant	206
	<i>Aedes aegypti</i>	Larvicidal	207
	<i>Sternechus subsignatus</i> ,	Contact toxicity	154
	<i>Rhyssomatus subtilis</i>		
	<i>Musca domestica</i>	Repellent, larvicidal, pupicidal fumigant and contact	68
	<i>Camptomyia corticalis</i>	Larvicidal	93
	<i>Zabrotes subfasciatus</i>	Ovicidal, adulticidal, repellent	185
	<i>Bovicola ocellatus</i>	Toxic	208
<i>E. citriodora</i>	<i>Tribolium castaneum</i>	Repellent	181
	<i>Tetranychus urticae</i> , <i>Neoseiulus californicus</i>	Fumigant and contact toxicity	209
	<i>Zabrotes subfasciatus</i>	Ovicidal, adulticidal, repellent	185
<i>E. urograndis</i>	<i>Triatoma infestans</i>	Toxic	210
<i>E. polybractea</i>	<i>Haematobia irritans</i>	Knockdown	211
<i>E. staigeriana</i>	<i>Lutzomyia longipalpis</i>	Egg, larva and adult contact toxicity	212
<i>Eupatorium inulaefolium</i> , <i>E. viscidum</i>	<i>Myzus persicae</i>	Antifeedant	123

Table 1 (Continued)

Plant species	Target insect	Action	Reference
<i>Flourensia oolepis</i>	<i>Tribolium castaneum</i> <i>Myzus persicae</i> , <i>Leptinotarsa decemlineata</i>	Contact toxin antifeedant	655
<i>Foeniculum vulgare</i>	<i>Tribolium castaneum</i> <i>Aedes albopictus</i> , <i>A. aegypti</i> <i>Callosobruchus maculatus</i> <i>Sitophilus zeamais</i>	Fumigant toxicity Larvicidal Fumigant toxicity Contact toxicity	173 96,213 174 214
<i>Hyssopus officinalis</i> <i>Hyptis spicigera</i>	<i>Thrips palmi</i> <i>Callosobruchus maculatus</i>	Fumigant toxicity Fumigant and contact Toxicity, repellent,	112 215,216
	<i>Sitophilus zeamais</i> , <i>S. granarius</i>	Contact toxicity, repellent	213,217
<i>H. suaveolens</i>	<i>Sitophilus zeamais</i> , <i>S. granarius</i>	Contact toxicity, repellent	213,217
	<i>Tribolium castaneum</i> <i>Ceratitis capitata</i> <i>Aedes albopictus</i>	Fumigant toxicity Toxic Larvicidal, repellent	163 218 219
<i>H. pectinata</i> , <i>H. fruticosa</i> <i>Ilex purpurea</i> <i>Illicium verum</i>	<i>Aedes aegypti</i> <i>Trichoplusia ni</i> <i>Chrysomya megacephala</i> <i>Aedes aegypti</i> <i>Culex pipiens</i> <i>Alphitobius diaperinus</i>	Larvicidal Fumigant toxicity Ovicidal Adulticidal Larvicidal Antifeedant	220 144 104,146,221,222
<i>Juniperus oxycedrus</i> <i>J. sahuaria</i> , <i>J. squamata</i> <i>J. chinensis</i>	<i>Lycoriella ingenua</i> <i>Stephanitis pyrioides</i> <i>Dermatophagoides</i> spp., <i>Tyrophagus putrescentiae</i>	Toxic Toxic Toxic	126 223 224
<i>J. virginiana</i>	<i>Resseliella oculiperda</i> <i>Camptomyia corticalis</i>	Repellent Toxic	147 93
<i>J. drupacea</i> <i>J. sahuaria</i> , <i>J. squamata</i> <i>J. chinensis</i>	<i>Culex pipiens</i> <i>Stephanitis pyrioides</i> <i>Dermatophagoides farinae</i> , <i>D. pteronyssinus</i> , <i>Tyrophagus putrescentiae</i>	Larvicidal Toxic Contact toxicity	225 223 224
<i>Laurus novocanariensis</i> <i>L. nobilis</i>	<i>Myzus persicae</i> , <i>Rhopalosiphum padi</i> <i>Tribolium confusum</i> <i>Rhyzopertha dominica</i> and <i>Tribolium castaneum</i>	Antifeedant Fumigant toxicity Repellent, toxic	80 226 227
<i>Lavandula angustifolia</i>	<i>Resseliella oculiperda</i> <i>Ixodes ricinus</i> <i>Camptomyia corticalis</i> <i>Sitophilus zeamais</i> <i>Ceratitis capitata</i> <i>Tetranychus cinnabarinus</i>	Repellent Repellent Toxic Repellent Toxic Toxic	79,93,147,158,206,213,214,218,228–231
<i>L. stoechas</i>	<i>Lasioderma serricorne</i> , <i>Rhyzopertha dominica</i>	Fumigant, toxicity Antifeedant	
<i>L. luisieri</i>	<i>Leptinotarsa decemlineata</i> , <i>Myzus persicae</i> , <i>Rhopalosiphum padi</i> <i>Sitophilus zeamais</i>	Repellent Antifeedant	
Lavandin (<i>L. angustifolia</i> x <i>L. latifolia</i>) <i>L. latifolia</i>	<i>Leptinotarsa decemlineata</i> , <i>Myzus persicae</i> , <i>Spodoptera littoralis</i> , <i>Rhopalosiphum padi</i>		
<i>Leptospermum scoparium</i>	<i>Dermatophagoides farinae</i> , <i>D. pteronyssinus</i> , <i>Tyrophagus putrescentiae</i>	Adulticidal	232
<i>Lippia gracilis</i> <i>L. sidoides</i>	<i>Aedes aegypti</i> <i>Nasutitermes corniger</i> <i>Aedes aegypti</i>	Larvicidal, adulticidal, Termiticidal Ovicidal, pupicidal, oviposition repellent	220,233 234 166
<i>L. multiflora</i> <i>L. alba</i>	<i>Bemisia tabaci</i> <i>Callosobruchus chinensis</i>	Fumigant toxicity Toxic, oviposition repellent	89 125

(Continued)

Table 1 (Continued)

Plant species	Target insect	Action	Reference
<i>L. turbinata</i> , <i>L. polystachya</i> <i>Litsea pungens</i>	<i>Culex quinquefasciatus</i> <i>Trichoplusia ni</i>	Neurotoxic Contact, residual, fumigant toxicity	235 144,236
<i>L. salicifolia</i>	<i>Aedes aegypti</i> <i>Sitophilus zeamais</i> , <i>Tribolium castaneum</i>	Irritant, repellent Repellent	237 238
<i>L. cubeba</i>	<i>Aedes aegypti</i> <i>Trichoplusia ni</i> <i>Reticulitermes speratus</i>	Irritant, repellent Contact toxicity Fumigant toxicity	237 236 103
<i>Maclura pomifera</i>	<i>Culex pipiens</i>	Repellent	239
<i>Majorana hortensis</i>	<i>Spodoptera littoralis</i> , <i>Aphis fabae</i>	Toxic	240
<i>Marrubium vulgare</i>	<i>Culex pipiens</i>	Ovicidal	241
<i>Matthiola longipetala</i>	<i>Tribolium confusum</i>	Growth inhibitor	242
<i>Melaleuca viridiflora</i>	<i>Thrips palmi</i> , <i>Cadra cautella</i>	Fumigant toxicity Larvicidal, fumigant Toxicity	93,112,179,207,208,237,243,243–246
<i>M. quinquenervia</i>	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Repellent, larvicidal	
<i>M. leucadendron</i>	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Repellent, neurotoxic, Larvicidal	
<i>M. linariifolia</i> , <i>M. dissitiflora</i> <i>M. alternifolia</i>	<i>Aedes aegypti</i> <i>Lucilia cuprina</i> <i>Pediculus capitis</i> <i>Bovicola ocellatus</i>	Larvicidal Repellent, toxic Adulticidal, ovicidal Toxic	
<i>M. viridiflora</i>	<i>Camptomyia corticalis</i>	Toxic	
<i>Mentha piperita</i> , <i>M. spicata</i>	<i>Culex quinquefasciatus</i> , <i>Aedes aegypti</i> , <i>Anopheles tessellatus</i>	Fumigant toxicity Toxic	62,85,92,93,105,112,120,152,195,201, 231,247–258
<i>M. piperita</i>	<i>Leptinotarsa decemlineata</i> <i>Musa domestica</i> <i>Sitophilus oryzae</i> , <i>Corcyra cephalonica</i> <i>Aedes aegypti</i> <i>Haematopinus tuberculatus</i>	Larvicidal Toxic Ovicidal Ovicidal Repellent Fumigant toxicity	
<i>M. pulegium</i>	<i>Pediculus humanus capitis</i> <i>Thrips palmi</i> <i>Dermatophagoides farinae</i> , <i>D. pteronyssinus</i> <i>Musa domestica</i>	Toxic Fumigant Larvicidal Ovicidal Toxic, repellent	
<i>M. citrata</i> <i>M. arvensis</i>	<i>Musa domestica</i> <i>Callosobruchus chilensis</i> <i>Sitophilus oryzae</i> , <i>Tribolium castaneum</i>	Toxic Larvicidal Toxic	
<i>M. longifolia</i>	<i>Tribolium castaneum</i> , <i>Callosobruchus maculatus</i> <i>Dendroctonus micans</i> <i>Rhipicephalus turanicus</i>	Larvicidal Toxic Adulticidal Toxic	
<i>M. spicata</i>	<i>Camptomyia corticalis</i> <i>Culex quinquefasciatus</i> , <i>Aedes aegypti</i> , <i>Anopheles stephensi</i> <i>Tetranychus cinnabarinus</i> <i>Sitophilus oryzae</i> <i>Sitophilus granarius</i> <i>Sitophilus granaricus</i>	Repellent Larvicidal Larvicidal Larvicidal Larvicidal	
<i>M. viridis</i> <i>M. suaveolens</i>	<i>Culex pipiens</i>		
<i>Micromeria fruticosa</i>	<i>Tetranychus urticae</i> , <i>Bemisia tabaci</i>	Fumigant toxicity	259
<i>Myroxylon pereira</i>	<i>Daphnia magna</i> , <i>Aedes aegypti</i>	Toxic, larvicidal	260
<i>Momordica charantia</i>	<i>Anopheles stephensi</i>	Repellent	261
<i>Myristica fragrans</i>	<i>Culex quinquefasciatus</i> , <i>Aedes aegypti</i> , <i>Anopheles tessellatus</i> <i>Callosobruchus chinensis</i> <i>Lycoriella ingênue</i> Gypsy moth larvae	Fumigant toxicity Fumigant toxicity Toxic Larvicidal, antifeedant	102,122,126
<i>Myrtus communis</i>	<i>Phlebotomus papatasi</i> , <i>Thrips palmi</i> <i>Sitophilus zeamais</i>	Repellent Fumigant toxicity Toxic	112,213,214,262,263

Table 1 (Continued)

Plant species	Target insect	Action	Reference
<i>Nepeta cataria</i>	<i>Aedes albopictus</i>	Larvicidal	
	<i>Acanthoscelides obtectus</i>	Toxic	
	<i>Blattella germanica</i> , <i>Musca domestica</i> , <i>Aedes aegypti</i>	Repellent	239,243,259,264
	<i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Repellent	
	<i>Stomoxys calcitrans</i>	Repellent	
<i>N. racemosa</i>	<i>Tetranychus urticae</i> , <i>Bemisia tabaci</i>	Fumigant toxicity	
<i>Nigella sativa</i>	<i>Callosobruchus chinensis</i>	Fumigant, oviposition Deterrence	102
<i>Ocimum canum</i>	<i>Anopheles gambiae</i>	Toxic	112,140,265–267b
	<i>Callosobruchus chinensis</i> , <i>C. maculatus</i>	Repellent	105,114,135,139,268–274
<i>O. basilicum</i>	<i>Thrips palmi</i> , <i>Sitophilus oryzae</i>	Fumigant toxicity,	
	<i>Ceratitis capitata</i> , <i>Bactrocera dorsalis</i> , <i>B. cucurbitae</i>	Insecticidal	
	<i>Anopheles stephensi</i>	Toxic	
	<i>Lymantria dispar</i>	Larvicidal	
	<i>Aedes aegypti</i>	Antifeedant	
	<i>Trogoderma granarium</i>	Ovicidal	
	<i>Sitophilus zeamais</i>	Adulticidal	
	<i>Culex quinquefasciatus</i>	Toxic	
	<i>Culex quinquefasciatus</i>	Larvicidal	
<i>O. sanctum</i>	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Larvicidal	
	<i>Culex quinquefasciatus</i>	Toxic	
<i>O. gratissimum</i>	<i>Exorista sorbillans</i>	Toxic	
	<i>Sitophilus zeamais</i>	Repellent	
<i>O. lamiifolium</i>	<i>Anopheles gambiae</i>	Larvicidal	
	<i>Anopheles arabiensis</i> , <i>Aedes aegypti</i>	Larvicidal	
<i>Oenanthe pimpinelloides</i>	<i>Culex pipiens</i>	Larvicidal	275
<i>Origanum acutidens</i>	<i>Lasioderma serricorne</i> , <i>Sitophilus granarius</i> , <i>Ephestia kuehniella</i>	Fumigant toxicity	93,112,131,231,259,262,276,277, 277–279,279,279–282
	<i>Thaumatococcus wilkinsoni</i>	Larvicidal	315
<i>O. onites</i>	<i>Plodia interpunctella</i> , <i>Ephestia kuehniella</i>	Toxic	120,283–285
	<i>Tetranychus cinnabarinus</i>	Toxic	
<i>O. marjorana</i>	<i>Culex pipiens</i>	Fumigant toxicity	
	<i>Euproctis chrysorrhoea</i>	Toxic	
	<i>Camptomyia corticalis</i>	Toxic	
	<i>Thrips palmi</i>	Larvicidal	
	<i>Rhipicephalus turanicus</i>	Larvicidal	
<i>O. minutiflorum</i>	<i>Euproctis chrysorrhoea</i>	Toxic	
	<i>Culex pipiens</i>	Toxic	
<i>O. vulgare</i>	<i>Tetranychus urticae</i> , <i>Bemisia tabaci</i>	Fumigant toxicity	
	<i>Nezara viridula</i>	Fumigant	
	<i>Camptomyia corticalis</i>	Toxic	
	<i>Spodoptera littoralis</i>	Toxic	
	<i>Nezara viridula</i>	Repellent	
	<i>Tribolium confusum</i>	Toxic	
	<i>Rhizopertha dominica</i>	Fumigant toxicity	
<i>O. glandulosum</i>	<i>Dendroctonus micans</i>	Toxic	
	<i>Bruchus dentipes</i>	Toxic	
<i>O. acutides</i>	<i>Sitophilus granarius</i> , <i>Tribolium confusum</i>	Toxic	
	<i>Sitophilus zeamais</i>	Fumigant toxicity	286
<i>Ostericum grosseserratum</i>	<i>Sitophilus zeamais</i>	Contact toxicity	287
<i>Paederia scandens</i>	<i>Ixodes ricinus</i>	Repellent	103,229,288
<i>Pelargonium graveolens</i>	<i>Reticulitermes speratu</i>	Fumigant, antitermitic	
<i>P. roseum</i>	<i>Rhipicephalus (Boophilus) annulatus</i>	Repellent, toxic	
<i>Perovskia abrotanoides</i>	<i>Tribolium castaneum</i> , <i>Sitophilus oryzae</i>	Fumigant	289
	<i>Rhodnius prolixus</i>	Toxic	290
<i>Pilocarpus spicatus</i>	<i>Blattella germanica</i>	Toxic	93,103,280,291,292
	<i>Camptomyia corticalis</i>	Toxic	

(Continued)

Table 1 (Continued)

Plant species	Target insect	Action	Reference
<i>P. dioica</i>	<i>Spodoptera littoralis</i>	Toxic	
	<i>Aedes aegypti</i> , <i>Culex pipiens</i>	Larvicidal	
	<i>Trachyspermum ammi</i>	Fumigant, antitermitic	
	<i>Camptomyia corticalis</i>	Toxic	
<i>Pimpinella anisum</i>	<i>Reticulitermes speratus</i>	Fumigant, toxic	
	<i>Lycoriella ing�neue</i>	Toxic	95,139,221,293
	<i>Culex pipiens</i>	Larvicidal	
	<i>Trogoderma granarium</i> , <i>Tribolium castaneum</i>	Adulticidal Toxic	
<i>Pinus tropicalis</i> , <i>P. caribaea</i>	<i>Lucilia sericata</i>		
	<i>Aedes aegypti</i>	Ovicidal	656
<i>Piper betle</i>	<i>Musca domestica</i>	Fumigant-acute toxicity	102,104,110,188,251,271, 294–296,296–298,298–306
<i>P. nigrum</i>	<i>Callosobruchus maculatus</i> , <i>Sitophilus zeamais</i> , <i>Rhizopertha dominica</i> , <i>Tribolium castaneum</i>	Fumigant toxicity	
	<i>Sitophilus oryzae</i> , <i>Corcyra cephalonica</i>	Toxic	
<i>P. longum</i>	<i>Spodoptera litura</i>	Toxic	
	<i>Callosobruchus chinensis</i>	Fumigant toxicity	
	<i>Spodoptera littura</i>	Toxic, antifeedant	
	<i>Aedes aegypti</i>	Adulticidal	
<i>P. aduncum</i> , <i>P. hispidinervum</i>	<i>Culex quinquefasciatus</i>	Larvicidal	
	<i>Sitophilus zeamais</i> , <i>Anopheles marajoara</i> , <i>Aedes aegypti</i>	Insecticidal, larvicidal	
<i>P. hispidinervum</i>	<i>Sitophilus zeamais</i>	Fumigant toxicity	
	<i>Spodoptera frugiperda</i>	Toxic	
<i>P. aduncum</i>	<i>Callosobruchus maculatus</i>	Toxic	
	<i>Aedes aegypti</i> , <i>A. albopictus</i>	Fumigant toxicity	
	<i>Anopheles marajoara</i> , <i>Aedes aegypti</i>	Larvicidal	
<i>P. tuberculatum</i>	<i>Callosobruchus maculatus</i>	Toxic	
	<i>Aedes aegypti</i>	Larvicidal	
<i>P. marginatum</i>	<i>Solenopsis saevissima</i>	Toxic	
	<i>Aedes aegypti</i>	Larvicidal	
<i>P. guineense</i>	<i>Sitophilus zeamais</i>	Toxic	
<i>P. humaytanum</i> , <i>P. permucronatum</i> , <i>P. hostmanianum</i>	<i>Aedes aegypti</i>	Larvicidal	
<i>P. sarmentosum</i>	<i>Brontispa longissima</i>	Antifeedant, toxic	
<i>Plectranthus glandulosus</i>	<i>Anopheles gambiae</i>	Toxic	266
<i>Pogostemon cablin</i>	<i>Preris rapae</i> , <i>Plutella xylostella</i>	Insecticidal	94,234,255,307–309
	<i>Musa domestica</i>	Toxic	
<i>Prangos acaulis</i>	<i>Nasutitermes corniger</i>	Toxic	
	<i>Choristoneura rosaceana</i>	Toxic	
	<i>Bemisia tabaci</i>	Repellent	
	<i>Dermatophagoides farinae</i>	Toxic	
	<i>Tribolium castaneum</i> , <i>Sitophilus oryzae</i>	Repellent	310
<i>Premna latifolia</i>	<i>Spodoptera littura</i>	Antifeedant	68
<i>Protium confusum</i>	<i>Aedes aegypti</i>	Larvicidal	311
<i>Psidium spp.</i>	wild mosquitoes, anthropophilic black flies	Repellent	178,301
<i>P. guajava</i>	<i>Spodoptera frugiperda</i>		
<i>Psoralea corylifolia</i>	<i>Culex quinquefasciatus</i>	Larvicidal, Adulticidal	312
	<i>Aedes aegypti</i> , <i>Culex quinquefasciatus</i> , <i>Anopheles stephensi</i>	Larvicidal	654
<i>Pongamia glabra</i>		Larvicidal	280
<i>P. pinnata</i>	<i>Plutella xylostella</i>		
	<i>Rhizopertha dominica</i> , <i>Tribolium castaneum</i>	Adulticidal	313
<i>Pyrenacantha staudtii</i>			
<i>Rosmarinus officinalis</i>	<i>Thrips palmi</i>	Fumigant toxicity	93,100,112,120,214,218,226,246, 293,314–319
	<i>Tribolium confusum</i>	Larvicidal, fumigant	
	<i>Cadra cautella</i>	Toxicity	

Table 1 (Continued)

Plant species	Target insect	Action	Reference
	<i>Acanthoscelides obtectus</i> , <i>Tineola bisselliella</i>	Toxic	
	<i>Pseudaletia unipuncta</i> , <i>Trichoplusia ni</i>	Toxic	
	<i>Ceratitis capitata</i>	Fumigant toxicity	
	<i>Sitophilus zeamais</i>	Toxic	
	<i>Camptomyia corticalis</i>	Toxic	
	<i>Dendroctonus micans</i>	Larvicidal	
	<i>Lucilia sericata</i>	Toxic	
	<i>Culex tritaeniorhynchus</i> , <i>Anopheles subpictus</i>	Larvicidal	
	<i>Varroa destructor</i>		
	<i>Aedes aegypti</i>		
	<i>Tribolium confusum</i>		
<i>Salvia hydrangea</i>	<i>Sitophilus granarius</i> , <i>Tribolium confusum</i>	Toxic	320
		Toxic	635
<i>S. officinalis</i>	<i>Leptinotarsa decemlineata</i>	Fumigant toxicity	93,112,120,140,267,280,321,322
	<i>Thrips palmi</i>	Toxic	
	<i>Sitophilus oryzae</i>	Toxic	
	<i>Camptomyia corticalis</i>	Repellent	
<i>S. plebeian</i>	<i>Callosobruchus chinensis</i> , <i>C. Maculates</i>	Toxic	
		Toxic	
<i>S. tomentosa</i>	<i>Acanthoscelides obtectus</i> , <i>Tribolium castaneum</i>	Toxic	
		Larvicidal	
	<i>Camptomyia corticalis</i>	Larvicidal	
<i>S. sclarea</i>	<i>Spodoptera littoralis</i>		
	<i>Dendroctonus micans</i>		
<i>S. multicaulis</i>	<i>Aedes albopictus</i>		
<i>S. elegans</i> , <i>S. splendens</i>			
<i>Sarum heteropoides</i>	<i>Culex pipiens</i> , <i>Aedes aegypti</i>	larvicidal	323
<i>Schinus terebinthifolia</i>	<i>Anopheles gambiae</i>	toxic	324,325
<i>S. areira</i>	<i>Tribolium castaneum</i>	repellent	
<i>Satureja spinosa</i> , <i>S. parnassica</i> , <i>S. thymbra</i> , <i>S. montana</i>	<i>Culex pipiens</i>	larvicidal	93,174,257,285,326,327
<i>S. hortensis</i>	<i>Culex quinquefasciatus</i>	Larvicidal	
	<i>Leptinotarsa decemlineata</i>	Toxic	
	<i>Camptomyia corticalis</i>	Toxic	
	<i>Callosobruchus maculates</i>	Toxic	
	<i>Bruchus dentipes</i>	Toxic	
<i>Sabina vulgaris</i>	<i>Trichoplusia ni</i>	Fumigant toxicity	144
<i>Saussurea lappa</i>	<i>Aedes albopictus</i>	Larvicidal	328
<i>Schizonepeta tenuifolia</i>	<i>Lycoriella ingenua</i>	Toxic	86
<i>Syzygium aromaticum</i>	<i>Ixodes ricinus</i>	Repellent	144,149,184,193,329
	<i>Trichoplusia ni</i>	Toxic	
	<i>Aedes aegypti</i> , <i>Anopheles dirus</i> , <i>Culex quinquefasciatus</i>	Oviposition deterrent	
	<i>Aedes aegypti</i>	Larvicidal	
<i>Tagetes filifolia</i>	<i>Trialeurodes vaporariorum</i>	Repellent, ovicidal	183,330–332,332–335
<i>T. patula</i>	<i>Rhipicephalus sanguineus</i>	Ovicidal	
	<i>Sitophilus zeamais</i>	Repellent	
<i>T. terniflora</i>	<i>Sitophilus oryzae</i>	Feeding deterrent	
<i>T. erecta</i>	<i>Aedes aegypti</i>	Larvicidal	
<i>T. rupestris</i>	<i>Ceratitis capitata</i>	Toxic	
<i>T. minuta</i>	<i>Triatoma infestans</i>	Repellent	
	<i>Anopheles stephensi</i>	Toxic	
<i>Thuja occidentalis</i>	<i>Thrips palmi</i>	Fumigant toxicity	112,218
	<i>Ceratitis capitata</i>	Toxic	
<i>Thymus vulgaris</i>	<i>Musca domestica</i>	Fumigant toxicity, Adulticidal, larvicidal	93,94,126,128,195,230,241,248,257,280–282,308,327,336–338,338–340
	<i>Nezara viridula</i>	Fumigant toxicity	
	<i>Spodoptera littoralis</i> , <i>Musca domestica</i> , <i>Culex quinquefasciatus</i> , <i>Leptinotarsa decemlineata</i>	Toxic	

(Continued)

Table 1 (Continued)

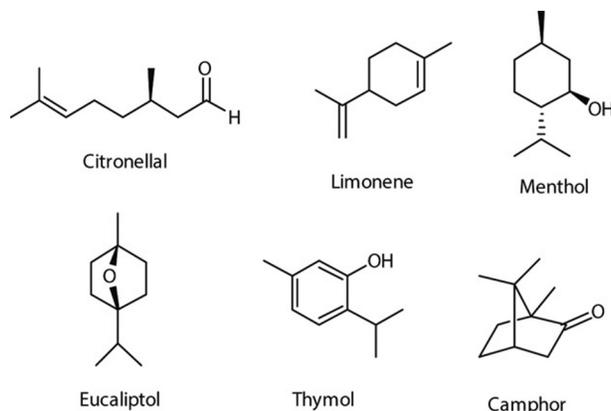
Plant species	Target insect	Action	Reference
	<i>Xanthogaleruca luteola</i>	Toxic	
	<i>Camptomyia corticalis</i>	Toxic	
	<i>Spodoptera littoralis</i>	Toxic	
	<i>Nezara viridula</i>	Ovicidal, repellent	
	<i>Choristoneura rosaceana</i>	Toxic	
	<i>Bemisia tabaci</i>	Contact toxicity	
	<i>Meligethes aeneus</i>	Repellent	
	<i>Culex quinquefasciatus</i>	Larvicidal	
	<i>Callosobruchus chilensis</i>	Toxic	
	<i>Leptinotarsa decemlineata</i> , <i>Myzus persicae</i> , <i>Spodoptera littoralis</i> , <i>Rhopalosiphum padi</i>		
<i>T. sipyleus</i>	<i>Rhipicephalus turanicus</i>	Antifeedant	
<i>T. capitatus</i>	<i>Culex pipiens</i>	Toxic	
<i>T. leucospermus</i>	<i>Culex pipiens</i>	Toxic	
<i>T. teucroides</i>	<i>Culex pipiens</i>	Repellent, larvicidal	
<i>T. persicus</i>	<i>Tribolium castaneum</i> , <i>Sitophilus oryzae</i>	Repellent Fumigant toxicity	
<i>T. spicata</i>	<i>Sitophilus granarius</i>	Fumigant toxicity	
<i>Trachyspermum ammi</i>	<i>Anopheles stephensi</i>	Repellent, repellent	103,129,341
	<i>Blattella germanica</i>	Contact toxicity	102
	<i>Daphnia magna</i> , <i>Aedes aegypti</i>	Toxic, larvicidal	
<i>Viola odorata</i>	<i>Callosobruchus chinensis</i>	Toxic	
	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Repellent	243
<i>Vitex pseudo-negundo</i>	<i>Tribolium castaneum</i> , <i>Sitophilus oryzae</i>	Fumigant toxicity	130,148,342
	<i>Callosobruchus maculatus</i>	Fumigant toxicity	
<i>V. agnus castus</i>	<i>Culex pipiens</i>	Larvicidal	
<i>Xylopiya aethiopica</i>	<i>Sitophilus zeamais</i>	Acute toxicity	343
<i>Zanthoxylum piperitum</i>	<i>Aedes gardnerii</i> , <i>Anopheles barbirostris</i> , <i>Armigeres subalbatus</i> , <i>Culex tritaeniorhynchus</i> , <i>C. gelidus</i> , <i>C. vishnui</i> group, <i>Mansonia uniformis</i> .	Repellent	86,344–349
	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>		
<i>Z. armatum</i>	<i>Lycoriella ing�neue</i>	Larvicidal	
<i>Z. piperitum</i>		Toxic	
<i>Z. xanthoxyloides</i>	<i>Acanthoscelides obtectus</i>	Repellent	
<i>Z. bungeanum</i>	<i>Stegobium paniceum</i>	Fumigant toxicity	
<i>Z. schinifolium</i>	<i>Sitophilus zeamais</i>	Fumigant toxicity	
<i>Z. beecheyamum</i>	<i>Culex pipens quinquefasciatus</i>	Repellent, toxic	
<i>Zingiber officinale</i>	<i>Culex tritaeniorhynchus</i> , <i>Anopheles subpictus</i>	Larvicidal	254,316
	<i>Musa domestica</i>	Larvicidal	
<i>Ziziphora clinopodioides</i>	<i>Callosobruchus maculatus</i>	Fumigant toxicity	350

years 2006–13) as proof of this extremely renewed interest. However, EOs of different origin shares a similar chemical profile and therefore their insecticidal effects are similar. We suggest a chemotype-based search for bioactive EOs rather than a plant species-based one to avoid data replication as shown in [Table 1](#).

Monoterpenes

Monoterpenes are the main components of plant EOs and, like these oils, have also been tested for their insecticidal effects. Some mosquito repellents include *p*-menthane-3,8-diol from mint as active ingredients and citronellal is also used in mosquito coils. A number of veterinary products for flea and tick control on domestic pets contain *d*-limonene from citrus peels as the active ingredient. Another important use of EO components is for the fumigation of beehives to control the honey bee parasite varroa (*Varroa Jacobson* and *V. destructor*) and the tracheal mite (*Acarapis woodi*). Thymol^{351–354} and menthol are used to control these mites.^{355,356} Other monoterpenes have also been tested: linalyl acetate, (*R*)-myrtenyl acetate, (*S*)-perillyl acetate, although thymyl

acetate exhibited high toxicity against *V. destructor* and significantly lower toxicity against *A. mellifera*.³⁵⁷ Camphor and eucalyptol are also used for this purpose.³⁵⁶ Several monoterpenoids exhibit toxicity against stored product and urban pests, are good spatial repellents and could be used in pest control.²³⁹



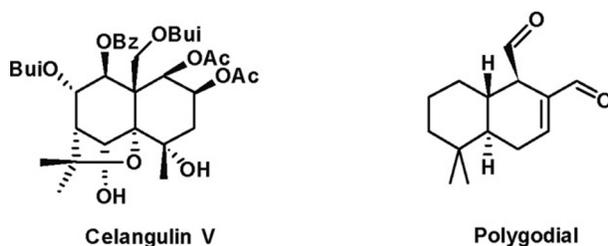
Although some monoterpenoid-based insecticides are used commercially, the mechanisms of action of these botanical insecticides have not been fully elucidated. Previous studies on modes of action of some monoterpenoids revealed several possible targets in the insect nervous system, including γ -aminobutyric acid (GABA) receptors,^{71,72,358} octopamine receptors,^{74,359} tyramine receptors,^{360,361} acetylcholinesterase (AChE)³⁶² and nicotinic acetylcholine receptors (nAChR).^{363,364} Among these targets, the GABA receptor may be involved in the response to monoterpenoids in both the central and the peripheral nervous system in insects. A recent study on the interaction between the chemical and structural properties of monoterpenoids and their binding activities at the housefly GABA receptor provided evidence that functional groups with oxygen atom(s) may be necessary for binding, and the hydrophobicity and total energy level of monoterpenoid molecules play key roles in the binding to the housefly GABA receptor.³⁶⁵

Table 2 provides an overview of the latest publications on insecticidal monoterpenes for the period 2006–12 (in part). Most of these compounds are known structures and have been studied as part of broader EO research.

Sesquiterpenes

Sesquiterpenes feature a different set of characteristics which also have an influence on insect activity, most effectively as contact irritants.²³⁹ Many species of the Celastraceae family such as the Chinese bittersweet (*Celastrus angulatus*) are widely distributed and used as traditional insecticides in China. These plants contain dihydro- β -agarofuran sesquiterpenoids based on a tricyclic 5,11-epoxy-5 β ,10 α -eudesman-4(14)-ene skeleton. The compact tricyclic scaffold seems to be a prerequisite for antifeedant or insecticidal activity as are the substitutions at C-1, C-6 and C-8. Nicotinic diacid substituent may also be involved in the antifeedant activity, possibly through neuronal action of nicotinic diacid.³⁹⁷ An emulsifiable mixture of celangulins has been developed for insect control.³⁹⁸ This functions as a digestive poison acting on the midgut tissue of the target insect larvae. Celangulins have structure-dependent effects on insect voltage-gated sodium channels³⁹⁹ and inhibit carboxylesterase activity.

A three-dimensional quantitative structure–activity relationship (3D-QSAR) study on the structural determinants that affect the narcotic and insecticidal activities of natural β -dihydroagarofuran sesquiterpene polyester have been performed.⁴⁰⁰ The relative contributions of the steric/electrostatic fields of the 3D-QSAR models show that the electronic effect governs the narcotic activities while a hybrid effect of the electrostatic and hydrophobic interactions is more influential in the insecticidal activities of these natural compounds.



Naturally occurring sesquiterpenoid dialdehydes of the drimane series such as polygodial, warburganal and muzigadial isolated from *Polygonum* and *Warburgia* spp. (Polygonaceae) have been thoroughly researched owing to their strong antifeedant activities and considerable attention has been devoted to the synthesis of these compounds.⁴⁰¹ The reactivity of the unsaturated dialdehyde functionality towards biological nucleophiles is considered to account for the antifeedant activity of these substances.⁴⁰¹ The

Table 2 Insecticidal monoterpenes for 2006–12 period (in part)

<i>M</i> Monoterpenes	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
Anethole	<i>Pediculus humanus capitis</i>	Ovicidal	366
Anisole	<i>Pediculus humanus capitis</i>	Ovicidal	366
Ascaridole	<i>Aedes aegypti</i>	Larvicidal	169
(Z)-ascaridole	<i>Sitophilus zeamais</i>	Fumigation toxicity	367
trans-anethole	<i>Aedes aegypti</i>	Larvicidal	368
Benzyl benzoate	<i>Tyrophagus putrescentiae</i>	Toxic	369
Bornyl acetate	<i>Dermatophagoides</i> spp., <i>Tyrophagus putrescentiae</i>	Toxic	224
Borneol	<i>Sitophilus oryzae</i>	Fumigant	370
Camphor	<i>Pseudaletia unipuncta</i> <i>Rhyzopertha dominica</i>	Toxic	317,370
3-Carene	<i>Aedes aegypti</i>	Larvicidal	371
Carvacrol	<i>Thaumetopoea wilkinsoni</i> <i>Aphis craccivora</i> , <i>Leucania separate</i> <i>Tyrophagus putrescentiae</i>	Larvicidal Toxic Toxic	278,372,373
1,2-epoxycarvone	<i>Aedes aegypti</i>	Larvicidal	374
R-S- Carvone	<i>Resseliella oculiperda</i>	Repellent action	129,147,250,366,374
Carvone	<i>Aedes aegypti</i> <i>Blattella germanica</i> <i>Anopheles stephensi</i> , <i>Aedes aegypti</i> <i>Pediculus humanus capitis</i>	Larvicidal Contact toxicity Larvicidal Ovicidal	
Carveol	<i>Blattella germanica</i>	Contact toxicity	129
cis-carveol	<i>Anopheles stephensi</i> , <i>Aedes aegypti</i>	Larvicidal	250
1,8-Cineole	<i>Pediculus humanus capitis</i> , <i>permethrin-resistant</i> , <i>Sitophilus oryzae</i> <i>Myzus persicae</i> , <i>Rhopalosiphum padi</i> <i>Callosobruchus maculatus</i> <i>Trichoplusia ni</i> <i>Musa domestica</i> <i>Musa domestica</i> <i>Pediculus humanus</i> <i>Callosobruchus chinensis</i> <i>Pediculus humanus capitis</i> <i>Pediculus humanus capitis</i>	Toxic, fumigant Toxicity Antifeedant Fumigant toxicity, Repellent Contact toxicity Fumigant Pupicidal Adulticidal Repellent, larvicidal Ache activity inhibitor Ovicidal	80,99,125,151,186,201,236,366,370,375,376
Citronellyl acetate	<i>Tetranychus urticae</i>	Fumigant toxicity	377
Citronellal	<i>Musca domestica</i> <i>Resseliella oculiperda</i> <i>Phaenicia sericata</i> <i>Aedes aegypti</i>	Toxic Repellent Toxic, flight disorders Larvicidal, repellent, Ovicidal	147,303,368,378
β-citronellol	<i>Tyrophagus putrescentiae</i> <i>Dermatophagoides farinae</i> , <i>D. pteronyssinus</i>	Toxic Toxic	369,379
Citronellol	<i>Ixodes ricinus</i> <i>Pediculus humanus</i> <i>Pediculus humanus capitis</i>	Repellent Toxic Toxic	193,380,381
Cuminaldehyde	<i>Blattella germanica</i> <i>Culex pipiens</i>	Contact toxicity Toxic	129,382
p-cymene	<i>Blattella germanica</i>	Contact toxicity	129
(R)-Fenchone	<i>Resseliella oculiperda</i>	Repellent action	147
Estragole	<i>Ceratitis capitata</i> , <i>Bactrocera dorsalis</i> , <i>Bactrocera cucurbitae</i>	Toxic	265
Eucalyptol	<i>Musa domestica</i> <i>Bhodnius prolixus</i>	Toxic Fumigant toxicity	383,384
Methyl.eugenol	<i>Ceratitis capitata</i> , <i>Bactrocera dorsalis</i> , <i>Bactrocera cucurbitae</i>	Toxic	265

Table 2 (Continued)

<i>M</i> Monoterpenes	Target insect	Action	Reference	
Eugenol	<i>Phaenicia sericata</i>	Toxic, flight disorders	368,378	
	<i>Aedes aegypti</i>	Larvicidal, repellent, Ovicidal		
Geraniol	<i>Ixodes ricinus</i>	Repellent	193,369,379–383,383	
	<i>Rhodnius prolixus</i>	Repellent		
	<i>Pediculus humanus</i>	Toxic		
	<i>Pediculus humanus capitis</i> (head lice)	Toxic		
	<i>Culex pipiens</i>	Toxic		
	<i>Tyrophagus putrescentiae</i>	Toxic		
	<i>Bhodnius prolixus</i>	Repellent		
Geranial	<i>Dermatophagoides farinae</i> , <i>D. pteronyssinus</i>	Toxic		
	<i>Callosobruchus chinensis</i>	Repellent	125	
Geranyl acetate	<i>Musca domestica</i>	Knock down and mortality	145	
R-, S-limonene	<i>Aedes aegypti</i>	Larvicidal	374	
	(4R)(+)-limonene	<i>Musca domestica</i>	Fumigant toxicity	385
Limonene oxide	<i>Rhipicephalus (Boophilus) microplus</i>	Larvicidal	386	
	Limonene	<i>Lycoriella ingenua</i>	Toxic	86,161,250,366,383,384,386,387
<i>Aedes albopictus</i>		Larvicidal		
<i>Anopheles stephensi</i> , <i>Aedes aegypti</i>		Larvicidal		
<i>Leptinotarsa decemlineata</i>		Toxic		
<i>Musa domestica</i>		Toxic		
<i>Bhodnius prolixus</i>		Fumigant toxicity		
<i>Rhipicephalus (Boophilus) microplus</i>		Larvicidal		
<i>Pediculus humanus capitis</i>		Ovicidal		
Myrcene		<i>Leptinotarsa decemlineata</i>	Toxic	387
		<i>Rhyzopertha dominica</i>	Fumigant	80,112,147,265,366,370,376,380,383,384
Linalool		<i>Myzus persicae</i> , <i>Rhopalosiphum padi</i>	Antifeedant	
	<i>Thrips palmi</i>	Fumigant toxicity		
	<i>Resseliella oculiperda</i>	Repellent		
	<i>Pediculus humanus</i>	Adulticidal		
	<i>Bactrocera dorsalis</i>	Toxic		
	<i>Pediculus humanus</i>	Toxic		
	<i>Musa domestica</i>	Toxic		
	<i>Bhodnius prolixus</i>	Fumigant toxicity		
	<i>Pediculus humanus capitis</i>	Adulticidal, ovicidal		
	<i>Ceratitis capitata</i> , <i>Bactrocera dorsalis</i> , <i>Bactrocera cucurbitae</i>	Toxic		
	<i>Pediculus humanus capitis</i>	Ovicidal		
	L-Menthol	<i>Culex quinquefasciatus</i> , <i>Aedes aegypti</i> , <i>Anopheles tessellatus</i>	Toxic	258
		Menthol	<i>Tetranychus urticae</i>	Fumigant toxicity
Menthone	<i>Lycoriella ingenua</i>		Toxic	86,383–385,388
	<i>Musca domestica</i>	Fumigant toxicity		
	<i>Sitophilus zeamais</i> , <i>Tribolium castaneum</i>	Contact-fumigant Toxicity		
	<i>Musa domestica</i>	Toxic		
Menthyl acetate	<i>Bhodnius prolixus</i>	Fumigant toxicity		
	<i>Bhodnius prolixus</i>	Repellent	383	
(Z,E)-Nepetalactone	<i>Musca domestica</i>	Toxic	239	
(E,Z)-Nepetalactone	<i>Blatella germanica</i>			
Nepetaparnone	<i>mosquito</i>	Larvicidal	389	
Nepetanudone				
Nerol	<i>Tyrophagus putrescentiae</i>	Toxic	369	
α -Pinene, β -Pinene	<i>Aedes aegypti</i>	Larvicidal	80,156,196,343,366	
	β -Pinene	<i>Myzus persicae</i> , <i>Rhopalosiphum padi</i>	Antifeedant	
<i>Sitophilus zeamais</i>		Acute toxicity		
<i>Culex pipiens</i>		Toxic		
	<i>Pediculus humanus capitis</i>	Ovicidal		

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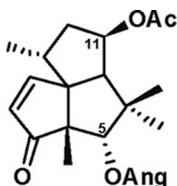
Table 2 (Continued)

<i>M</i> Monoterpenes	Target insect	Action	Reference
Piperitone	<i>Callosobruchus maculatus</i> <i>Artemisia judaica</i>	IGR ^a Antifeedant	119,190
(4R)(+)-pulegone	<i>Musca domestica</i>	Fumigant toxicity	385
Pulegone	<i>Lycoriella ingêneue</i> <i>Phaenicia sericata</i> <i>Sitophilus zeamais</i> , <i>Tribolium castaneum</i> <i>Aedes aegypti</i>	Toxic Toxic, flight disorders Contact-fumigant Toxicity Larvicidal, repellent, Ovicidal	86,368,378,388
4-Phenyl-2-butanone	<i>Sitophilus zeamais</i>	Contact toxicity	390
(-)-terpinen-4-ol	<i>Pediculus humanus capitis</i>	Adulticidal, ovicidal	376
(+)-Terpinen-4-ol	<i>Sitophilus zeamais</i> <i>Pediculus humanus</i>	Acute toxicity Adulticidal, ovicidal	343,391
□-Terpineol	<i>Trichoplusia ni</i>	Toxic	317
γ-terpinene, terpinen-4-ol	<i>Spodoptera littoralis</i> , <i>Aphis fabae</i> , <i>Mythimna separta</i>	Toxic	240,392
γ-Terpinene	<i>Leptinotarsa decemlineata</i> <i>Aedes aegypti</i>	Toxic Larvicidal	374,387
Terpinolene	<i>Aedes albopictus</i> <i>Ochlerotatus togoi</i>	Larvicidal Toxic	323,371
2-tridecanone	<i>Callosobruchus maculatus</i>	Fumigant toxicity	393
Thymol	<i>Sitophilus oryzae</i> <i>Thaumatococcus wilkinsoni</i> <i>Trichoplusia ni</i> <i>Chilo partellus</i> <i>Blattella germanica</i> <i>Aedes aegypti</i>	Fumigant Larvicidal Toxic Contact toxicity Larvicidal, repellent, Ovicidal	129,278,368,370,394,395
α-Terpineol	<i>Resseliella oculiperda</i> <i>Pediculus humanus capitis</i>	Repellent action Adulticidal, ovicidal	147,376
Geniposidic acid, 10-hydroxyloganin, deacetyl daphylloside, monotropein	<i>Kaloterms flavicollis</i> , <i>Crematogaster scutellaris</i>	Toxicity	396

^aIGR, Insect growth regulation effects.

antifeedant activity of polygodial acetal derivatives (propylene and ethylene) is consistent with the proposed adduct formation with amino groups.⁴⁰² However, the lack of correlation between reactivity towards nucleophiles and the antifeedant effects of polygodial and warburganal suggests that their insect antifeedant action may depend on other properties as indicated by the activity of keto-aldehydes and 3-hydroxydrimanones.⁴⁰³

The potential of the sesquiterpenes present in *Senecio* species as natural crop protectant lead have recently reported.⁴⁰⁴ These species are characterized by their content in sesquiterpenes with eremophilane, cacalol, bisabolane, silphinene, caryophyllane, humulane, germacrane and benzofurane skeletons. Eremophilanes, the most abundant sesquiterpene group in *Senecio*, are antifeedants and have post-ingestive effects, cacalolides are antifeedants and silphinenes exhibit remarkable insect antifeedant effects and act on insect GABA receptors. It is important to emphasize that silphinenes are uncommon skeletons in *Senecio*, only found in one species (*S. palmensis*) with a structural similarity to the known GABA antagonist, picrotoxinin.

**Silphinene derivative**

Mixtures that include both monoterpenes (acting as a good spatial repellent) and sesquiterpenes (good contact repellent) are extremely effective via both modes of action and show potential for residual repellent action from a natural product.²³⁹

Table 3 shows the latest publications on insecticidal sesquiterpenes. The number of compound hits is similar to that of monoterpenes, however new structures are described and these are mostly antifeedants in contrast to the monoterpenes shown in Table 2 which are all known and mostly toxic (fumigants). Therefore, sesquiterpenes can be considered as an interesting source of molecular models with potentially useful insect antifeedant properties.

Table 3 Insecticidal sesquiterpenes for 2006–12 period (in part)

<i>Sesquiterpenes</i>	<i>Type</i>	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
Nerolidol	Linear	<i>Pediculus humanus</i>	Adulticide, ovicidal	391
Polygodial derivatives	Drimane	<i>Spodoptera littoralis</i> , <i>Leptinotarsa decemlineata</i> , <i>Myzus persicae</i> , <i>Rhopalosiphum padi</i>	Antifeedant	402
(+)-Pterocarpol	Eudesmane	<i>Reticulitermes speratus</i> <i>Spodoptera litura</i>	Antifeedant	405,406
3 α -[2,3-Epoxy-2-methylbutyryloxy]-4 α -acetoxy-11-peroxyeudesmen-1-one		<i>Spodoptera frugiperda</i>	Antifeedant	
1 α -Tigloyloxy-8 β H,10 β H-eremophil-7(11)-en-8 α ,12-olide	Eremophilanolide	<i>Leptinotarsa decemlineata</i>	Antifeedant	407,408
10 α -Hydroxy-1-oxoeremophila-7(11),8(9)-dien-12,8-olide		<i>Spodoptera littoralis</i>	Antifeedant	
10 α -Hydroxy-1-oxoeremophila-7(11),8(9)-dien-12,8-olide		<i>Myzus persicae</i>	Antifeedant	
1 α ,10 β -Dihydroxy-8a-methoxyeremophil-7(11)-en-12,8 β -olide		<i>Spodoptera littoralis</i> , <i>Rhopalosiphum padi</i>		
1 β ,10 β -Epoxy-8a-methoxyeremophil-7(11)-en-12,8 β -olide				
6 β ,8 α -Dihydroxyeremophila-1(10),7(11)-dien-12,8 β -olide / Toluccanolide C				
6-Hydroxyeuryopsin, 6-acetyloxy-1(10)-epoxyeuryopsin	Furanoeremophilane	<i>Leptinotarsa decemlineata</i>	Antifeedant	409,410
6 β -angeloyloxy-1,10-dehydrofuranoeremophilan-9-one, 6 β -hydroxy-1,10-dehydrofuranoeremophilan-9-one, 6 β -propionyloxy-1,10-dehydrofuranoeremophilan-9-one		<i>Myzus persicae</i> , <i>Rhopalosiphum padi</i>		
Cacalol acetate	Cacalolide	<i>Leptinotarsa decemlineata</i> ,	Antifeedant	409
Aguerin B, Chlorojanerin, Janerin, Cynaropicrin	Guaianolide	<i>Sitophilus granarius</i> , <i>Trogoderma granarium</i> , <i>Tribolium confusum</i>	Antifeedant	411
Artesin, Taurin, Artemin	Eudesmanolide	<i>Spodoptera littoralis</i>	Antifeedant	412
Aureane	Bisabolane	<i>Aphicidal??</i>	Toxic	413,414
7-epizingiberene		<i>Bemisia tabaci</i>	Repellent	
R-curcumene				
Traginone	Norsesquiterpene	<i>Aphicidal</i>	Toxic	413
Pogostone	Norsesquiterpene	<i>Preris rapae</i> , <i>Plutella xylostella</i>	Toxic	309
Caryophyllene oxide	Caryophyllane	<i>Aedes aegypti</i> larvae <i>Leptinotarsa decemlineata</i> , <i>Spodoptera littoralis</i> .	Toxic Antifeedant	80,220
Celangulatin A and B Celangulins IV and V Celangulatin C-F Ejaponine A and B	Eudesmane	<i>Mythimna separate</i>	Toxic Insecticidal Stomach toxicity Insecticidal	415–420

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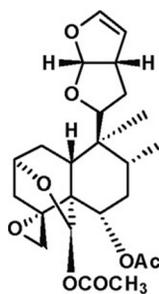
Table 3 (Continued)

<i>Sesquiterpenes</i>	<i>Type</i>	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
1 α ,2 α ,13-triacetoxy-8 β -isobutanoyloxy-9 α -benzoyloxy-4 β ,6 β -dihydroxy- β -dihydroagarofuran Celangulin I 1 β ,2 β ,6 α ,8 α ,12-pentaacetoxy-9 α -benzoyloxy-4 α -hydroxy- β -dihydroagarofuran, 1 β ,2 β ,6 α ,8 β -tetraacetoxy-9 β -benzoyloxy-12-isobutanoyloxy-4 α -hydroxy- β -dihydroagarofuran, Angulatueoid B 1 β -acetoxy-8 α ,13-di-isobutanoyloxy-2 β -(α -methyl)-butanoyloxy-4 α ,6 α -dihydroxy- β -dihydroagarofuran 1 β ,2 β ,6 α ,13-tetra-acetoxy-8 β -(α -methyl)-butanoyloxy-9 α -benzoyloxy- β -dihydroagarofuran 1 β ,2 β ,6 α ,8 β -tetra-acetoxy-9 α -benzoyloxy-13-butanoyloxy- β -dihydroagarofuran 1 β ,2 β ,6 α ,13-tetra-acetoxy-8 β -iso-butanoyloxy-9 α -furancarboxyloxy-4 α -hydroxy- β -dihydroagarofuran Kupiteng A-C Clavigerins A-C	Bergamotane	<i>Tineola bisselliella</i> <i>Anthrenocerus australis</i>	Antifeedant	421
Elemol Geijerene, Pregeijerene	Elemane Norsesquiterpene	<i>Culex pipiens</i> <i>Helicoverpa armigera</i> <i>Anopheles gambiae</i> <i>Culex quinquefasciatus</i> <i>Aedes aegypti</i> <i>Spodoptera litura</i>	Larvicidal Antifeedant and toxic Fumigant toxicity Antifeedant, oviposition deterrent	239 141,142,422
Germacrene D	Germacrane	<i>Anopheles gambiae</i> <i>Culex quinquefasciatus</i> <i>Aedes aegypti</i>	Fumigant toxicity	422
Hugonianene A (\pm), (+), (-)-Gossypol	Himachalene Cadinane	<i>Anopheles gambiae</i> <i>Helicoverpa zea</i> <i>Podisus nigrispinus</i>	Larvicidal Toxic, IGR ^a IGR ^a	423 424
Tavulin, Tanachin, Tamirin	Germacranolide	<i>Spodoptera littoralis</i>	Antifeedant	412
Tutin, 2- <i>iso</i> -Butenoyl-tutin Nepetaparnone, Nepetanudone megalanthine	Tutin group Iridoid Farnesane	<i>Mythimna separata</i> mosquito <i>Leptinotarsa decemlineata</i> , <i>Spodoptera littoralis</i>	Antifeedant Larvicidal Antifeedant	425 389 426
nootkatone	Valencane	<i>Ixodes scapularis</i> , <i>Amblyomma americanum</i>	Repellent	427

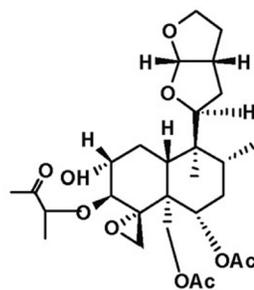
^aIGR, Insect growth regulation effects.

Diterpenes

Clerodane diterpenoids have been found in hundreds of species of plants from a number of different families. Several genera from the Verbenaceae and Lamiaceae families have been identified as rich sources of *neo*-clerodane diterpenoids. These metabolites have attracted considerable attention for their biological activity which includes piscicidal, trypanocidal and antibacterial properties. The insect antifeedant property of clerodane diterpenes is the most extensively studied bioactivity of these compounds.⁴²⁸ *Scutellaria* and *Ajuga* genera (Lamiaceae) produce some of the most potent clerodane antifeedants. In *Scutellaria*, jodrellin B (occurring in *S. albidia*, *S. galericulata*, *S. grossa*, *S. polyodon*, and *S. woronowii*) and scutecyprol B (found in *S. columnae*, *S. cypria*, *S. grossa* and *S. rubicunda*) exhibit the highest antifeedant index against *Spodoptera littoralis* (54; 483). From *Ajuga pseudoiva* leaves, 14,15-dihydro-ajugapitin displayed the highest activity.⁴²⁹ Furthermore, the genus *Ajuga* is one of the richest sources of clerodane diterpenes.⁴³⁰



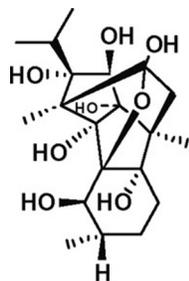
Jodrellin B



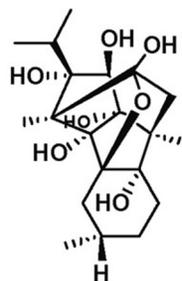
14,15-Dihydroajugapitin

Ryanodane diterpenes are compounds which are structurally related to the known insecticide ryanodine (see section 3.10). Several ryanodane diterpenes including ryanodol, cinnzeylanol, cinnzeylanone and cinnzeylanine have been isolated from the Macaronesian paleoendemism *Persea indica* (Lauraceae).^{431,432} Ryanodol and didehydroryanodol, in contrast to ryanodine and didehydroryanodine, have low toxicity to mice and scant activity at the mammalian ryanodine receptor but are potent knockdown agents for injected houseflies or cockroaches suggesting a possible difference in the target sites of mammals and insects.⁴³³ The antifeedant activity of these compounds has been evaluated showing the importance of the 11-hemiketal group for the antifeedant effects of ryanodane diterpenes. The comparative antifeedant effects of several non-alkaloidal and alkaloidal ryanoids supported the hypothesis of a ryanodol-specific mode of action in insects.^{432,434} The insect-selective insecticidal and antifeedant effects of ryanodanes hold a promising future for their use as biopesticides. However their availability is a problem which would need to be addressed prior to potential exploitation.

A recent review on labdane, halimane, and clerodate type diterpenes and their biological effects including antifeedant activities has been published.⁴³⁵ Table 4 shows the reported insecticidal diterpenes for the period 2006–11.



Ryanodol



Cinnzeylanol

Triterpenes

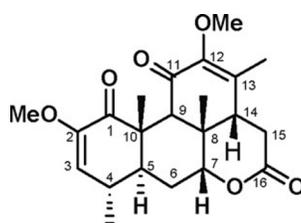
Triterpenes comprise a versatile group of bioactive compounds widely distributed in the plant kingdom with many pharmacological activities. The main groups of bioactive triterpenes and their glycosides (saponins) are represented by tetracyclic derivatives of protostane, cycloartane, dammarane; and pentacyclic derivatives of ursane lupane, hopane, oleanane and friedelane skeletons.

Quassinoids, the bitter compounds of the Simaroubaceae family, are a group of structurally complex and highly oxygenated degraded triterpenes. They are divided into five groups according to their basic skeleton, C-18, C-19, C-20, C-22 and C-25. In recent years, attention has been focused on quassinoids because several of them have shown promising biological activities and its extraction and cristalization from natural sources has been recently patented (Patent US 2013/0012726 A1). Some quassinoids such as bruceantin, isobrucein, simalikalactone and glaucarubinone present insecticidal and antifeedant effects in insects.⁴⁵⁴ Quassin was first used as an insecticide at the end of the 17th century with the application of plant extracts from *Quassia amara*. In contrast to other activities of quassinoids, quassin, which is inactive in most other biological activities, shows antifeedant activity against aphids,⁴⁵⁵ Mexican bean beetle and the southern armyworm.⁴⁵⁴ More recent studies also reveal this activity in other species and/or other quassinoids as in the case of Brutasol from *Brucea javanica* L. with toxic and antifeedant effects against *Spodoptera exigua* (Zhaug et al.,⁴⁵⁶ isobrucein and neosergeolide from *Picrolemma spruce* with larvicidal activity against *Aedes aegypti*⁴⁵⁷ and others.^{454,458}

Table 4 Insecticidal diterpenes for the period 2006–11 (in part)

Diterpenes	Class	Target insect	Action	Reference
Hugorosenone	Rosane	<i>Anopheles gambiae</i>	Larvicidal	436
4- <i>epi</i> -Abieta-7,13-dien-3-one	Abietane	<i>Mythimna separata</i> , <i>Pieris rapae</i>	Antifeedant	437,438
Abieta-7,13-dien-3-one			Insecticidal	
6,7-dehydroroleanone, taxodione, 14-deoxycoleon U, xanthoperol		<i>Reticulitermes speratus</i>	Terminicidal, Antifeedant	
6,10-(<i>E,E</i>)-Thymifodioic acid (2 <i>E,6E</i>)-2-(4-Methylpent-3-enyl)-6-[3-(2-oxo-2,5-dihydrofuran-3-yl)-propylidene]-hept-2-ene-dioic acid	Linear	<i>Tenebrio molitor</i>	IGR ^a	439
Neoclerodane derivatives	Neoclerodane	<i>Tribolium castaneum</i>	Antifeedant	440,441
Scuteceprol A		<i>Spodoptera exigua</i>	Antifeedant	
Parnapimarol	Pimarane	<i>mosquito</i>	Larvicidal	389
14- <i>O</i> -Methyl-ryanodanol	Ryanodane	<i>Aedes aegypti</i>	Larvicidal	442
13-deoxyitol A, itol A	Iso-ryanodane	<i>Nilaparvata lugens</i> , <i>Sogatella furcifera</i> , <i>Plutella xylostella</i> , <i>Spodoptera litura</i>	Antifeedant and contact toxicity	443
6 α -hydroxyvouacapan-7 β ,17 β -lactone (1), 6 α ,7 β -dihydroxyvouacapan-17 β -oic acid (2) and methyl 6 α ,7 β -dihydroxyvouacapan-17 β -oate	Furans	<i>Aedes aegypti</i>	Larvicidal	444
Ajuganipponin A, Bajugamarins A ₁ , B ₂ , A ₂ , F ₄	Neoclerodane	<i>Spodoptera littoralis</i>	Antifeedant	445
Bjugamacrin B, Ajugacumbin A, Ajugatakasin A, Ajugacumbin B, <i>ent</i> -3 β -(3-Methyl-2-butenoyl)oxy-beyer-15-en-19-oic acid	Beyerane	<i>Spodoptera littoralis</i>	Antifeedant	446
Mixt. (4 <i>R</i> ,19 <i>R</i>) and (4 <i>R</i> ,19 <i>S</i>) diastereoisomers of Coleon A	Abietane	<i>Spodoptera littoralis</i>	Antifeedant	446
Rhodojaponin-III	Grayanoid	<i>Pieris rapae</i>	Antifeedant, IGR ^a	447
14- <i>O</i> -methyl-ryanodanol	Ryanodane	<i>Aedes aegypti</i>	Larvicidal	442
grandifloric acid, 17-hydroxy-16 α - <i>ent</i> -kauran-19-oic acid, 15 β -hydroxy- <i>ent</i> -trachyloban-19-oic acid	<i>ent</i> -kaurene	<i>Cochylis hospes</i>	Oviposition stimulants	448
Anticopalic acid	Trachilobane			
3- <i>O</i> -(2,3-dimethylbutanoyl)-13- <i>O</i> -dodecanoylingenol, 3- <i>O</i> -(2' <i>E</i> ,4' <i>Z</i> -decadienoyl)-ingenol	Labdane	<i>Spodoptera frugiperda</i>	Antifeedant	449
Pierisoids A, B	Ingenane	<i>Tetranychus urticae</i> , <i>Nilaparvata lugens</i>	Acaricidal, insecticidal	450
	3,4- <i>seco</i> -grayanane	<i>Helicoverpa armigera</i>	Antifeedant	451
Jolkinolide B, 17-hydroxyjolkinolide A, B	<i>ent</i> -abietadienolide	<i>Tribolium castaneum</i> , <i>Sitophilus zeamais</i>	Feeding deterrent	452
Tagalsin A, B, H	Dolobrane	<i>Tribolium castaneum</i>	Feeding deterrent	453

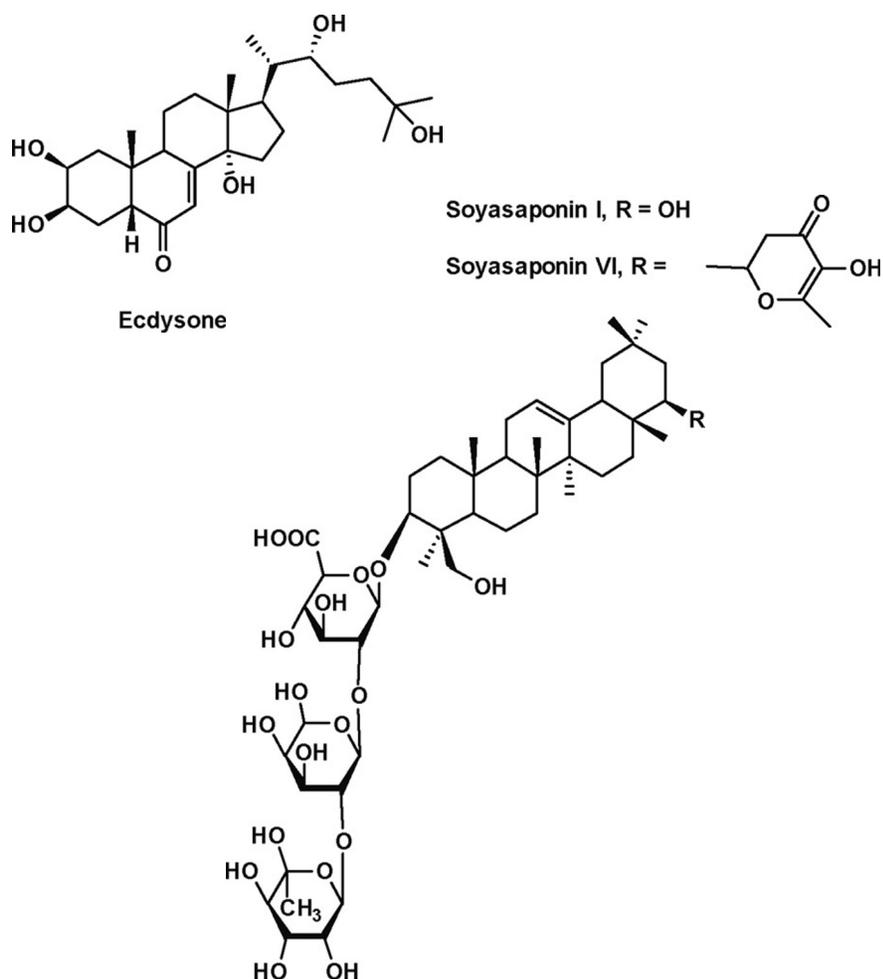
^aIGR, Insect growth regulation effects.

**Quassin**

Saponins are widely distributed among plants and have a wide range of biological properties and an increasing pesticidal potential. In fact, these substances are known by their toxicity to harmful insects (anti-feeding, disturbance of the moult, growth regulation, mortality, etc.). The insecticidal activity of saponins is due to their interaction with cholesterol, causing a disturbance of the synthesis of ecdysteroids. These substances are also protease inhibitors or cytotoxic to certain insects (see Chaieb).⁴⁵⁹

Cestrum parqui (Solanaceae) is a shrub from Chile and toxicity comes from the saponic fraction of the plant. *C. parqui* saponins, for example, are toxic to *Schistocerca gregaria*, *Spodoptera littoralis* and *Tribolium confusum*. This toxicity may also be the result of

interference with ecdysone metabolism by interfering with dietary cholesterol.^{460,461} Alfalfa saponins exhibited deterrent and toxic effects against the pea aphid *Acyrtosiphon pisum*.⁴⁶² The larvicidal effect of aqueous extracts of the African plants *Hemidesmus indicus* roots, *Gymnema sylvestre* and *Eclipta prostrata* against *Culex quinquefasciatus* larvae have been attributed to their high saponin content.⁴⁶³ Insecticidal soyasaponins have been isolated from field pea (*Pisum sativum*) extracts.⁴⁶⁴ The total saponins from the roots and shoots of three *Medicago* species (*M. arabica*, *M. hybrida* and *M. murex*) included in the diet of *Leptinotarsa decemlineata* larvae reduced their feeding and growth and survival rates.⁴⁶⁵ Saponins from the Indian trees *Diploknema butyracea* and *Sapindus mukorossi* were found to have antifeedant and IGR activity against *Spodoptera litura*.⁴⁶⁶ More recently, triterpene saponin with mosquitocidal activity against *Aedes aegypti* and *Culex quinquefasciatus* has been isolated from *Zygophyllum coccineum* growing in the Egyptian deserts.⁴⁶⁷ The Chilean tree *Quillaja saponaria* contains bioactive saponins showing strong aphicidal and deterrent activity against the pea aphid *Acyrtosiphon pisum*.⁴⁶⁸ Chinese mangrove associate, *Catunaregam spinosa*, contains many triterpenoid saponins showing strong antifeedant activity on *Plutella xylostella*.⁴⁶⁹ In the same way, saponins from *Microsechium helleri* and *Sicyos bulbosus* showed significant postingestive effects on *Spodoptera littoralis* larvae.⁴⁷⁰

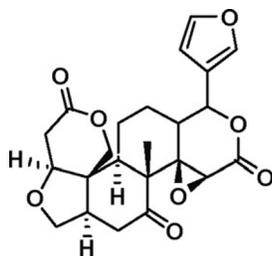


The search for limonoids started way back when scientists started looking for the factor responsible for bitterness in citrus which has a negative impact on citrus fruit and the juice industry worldwide. The term limonoids was derived from limonin, the first tetranortriterpenoid obtained from citrus bitter principles.

Although hundreds of limonoids have been isolated from several different plants, their occurrence in the plant kingdom is exclusively confined to plant families of the Rutales order, most abundant in Meliaceae and Rutaceae and less frequent in Cneoraceae and *Harrisonia* sp. of Simaroubaceae.

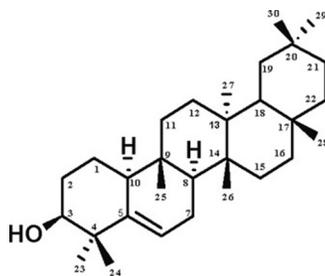
Limonoids are highly oxygenated modified triterpenoids with a prototypical structure derived from a precursor with a 4,4,8-trimethyl-17-furanylsteroid skeleton. All naturally occurring citrus limonoids contain a furan ring attached to the D-ring at C-17 as well as oxygenated functional groups at C-3, C-4, C-7, C-16 and C-17. There are fewer structural variations in limonoids found in Rutaceae as compared to Meliaceae and these are generally limited to the modification of A and B rings. The limonoids of Meliaceae are more complex with a very high degree of oxidation and rearrangement in structure.²⁴

Compounds from this group exhibit a wide range of biological activities (insecticidal, antifeedant and growth regulating) on insects as well as antibacterial, antifungal, antimalarial, anticancer and others. However, insect antifeedant activity is one of the most potent activity of these compounds. In this connection, an extensive review on antifeedancy and Insects Growth Regulatory Activity of Meliaceous Limonoids has been reported (see Tan and Luo).³⁴



Limonin

The insecticidal activity of a wide range of plant pentacyclic triterpenes from different chemical classes on several insect pests (*Spodoptera littoralis*, *Leptinotarsa decemlineata* and *Myzus persicae*) has been recently reviewed.⁴⁷¹ The presence of different substituents at C-3 and C-28 modulate these insecticidal effects.

Glutinol from *Echium wildpretii*

Other triterpene classes and derivatives including lanostanes, friedelanes and cyloartanes also exhibit insect growth regulation effects^{472–474,644} and therefore merit further investigation. Table 5 shows the reported insecticidal triterpenes for the period 2006–13.

Sterols

At least 100 different sterols have been identified in plants (e.g. sitosterol, stigmasterol and spinasterol) and these take part in plant growth processes. Most plant sterols have an alkyl group on the side chain and may have a double bond at position 5 (Δ^5 sterols) or 7 (Δ^7 sterols). Sitosterol is the most abundant and common plant sterol.

Sterols play a critical role in insects as components of cellular membranes, precursors of molting hormones (e.g. 20-OH ecdysone) and regulating genes involved in developmental processes. They are a required element in the diet of herbivorous insects since they lack the ability to biosynthesize cholesterol and have a dietary dependence on plant sterols that can be metabolized to cholesterol by the intervention of sterol carrier proteins (SCP). However, phytosterol have been reported to affect insects in different ways including antifeedant, cytotoxic and SCP inhibitor activities. This is why there is increased studies on insecticidal activities of phytosterols against major agricultural pests.^{12,17,480–483} Table 6 shows the reported insecticidal triterpenes for the period 2008–12.

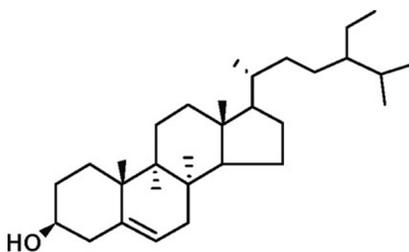
 β -sitosterol

Table 5 Insecticidal triterpenes for 2006–13 period (in part)

<i>Triterpenes</i>	<i>Class</i>	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
2 α -Hydroxyfriedel-3-one, 2,3- <i>seco</i> -Friedelan-2-al-3-oic acid, 3 β - and 3 α -Hydroxyfriedelane, 3 β - and 3 α -Hdroxyfriedelane, 3 α -Hydroxyfriedel-2-one, 4 β -Hydroxyfriedel-3-one, 3,4- <i>seco</i> -Friedelan-4-oxo-3-oic-acid, Friedelin-2,3-lactone, 3 α -Hydroxyfriedel-2-one	Friedelane	<i>Spodoptera littoralis</i>	Toxic, IGR ^a	474
β ,24,25-Trihydroxycycloartane beddomei lactone	Cycloartane	<i>Cnaphalocrocis medinalis</i>		198
Spirocaracolitones	Friedelin derivative	<i>Sitophilus oryzae</i>	Antifeedant	475
α -Euphol, α -Euphorbol, Obtusifoliol and 31-nor-Lanostenol derivatives	Lanostane	<i>Spodoptera littoralis</i>	IGR ^a	472
<i>iso</i> -Onoceratriene, 3-Keto-22-hydroxyonoceradiene, Onoceradienedione, Lansiolic acid, Lansiolic acid A, Humilinolides C and D, Gedunin	Limonoid	<i>Sitophilus oryzae</i>	Antifeedant	475
Musidunin, Musidulol	Limonoid	<i>Pectinophora gossypiella</i> <i>Spodoptera frugiperda</i> .	Antifeedant	476
Unidentifie saponin		<i>Schistocerca gregaria</i>	Toxic	461
Zanhic acid tridesmoside, Medicagenic acid glycosides	Oleanane	<i>Acyrtosiphon pisum</i>	Antifeedant	462
Dehydrosoyasaponin I	Oleanane	<i>Sitophilus oryzae</i>	Antifeedant and insecticidal	464
Soyasaponins	Quassinoid	<i>Spodoptera exigua</i>	Antifeedant, toxic	456
Brusatol	Quassinoid	<i>Spodoptera exigua</i>	Antifeedant, toxic	456
Isobrucein, neosergeolide	Quassinoid	<i>Aedes aegypti</i>	Larvicidal	457
3-O-[β -D-(2-O-sulphonyl) glucopyranosyl] quinovic acid	Saponin	<i>Aedes aegypti</i> , <i>Culex quinquefasciatus</i>	Insecticidal	467
3-O-[β -D-glucopyarnosyl- β -D-glucopyranosyl]-16-R-hydroxyprotobassic acid-28-O-[ara-glc-xyl]-ara (M1-I); 3-O- β -D-glucopyranosyl-glucopyranosyl-16-R-hydroxyprotobassic acid-28-O-[ara-xyl-ara]-apiose; 3-O-[β -D-xyl-(OAc) 3 β -D-arabinopyranosyl 3 β -D-rhamnopyranosyl] hederagenin-28-O-[β -D-glc 3 β -D-glc 3 β -D-rhamnopyranosyl] ester	Saponins	<i>Spodoptera litura</i>	Antifeedant, IGR ^a	466
Catanarosides, swartziatrioside, aralia-saponin, araliasaponin	Saponins	<i>Plutella xylostella</i>	Antifeedant	469
Bayogenin, heteropappussaponin 5, heteropappussaponin 7	Saponins	<i>Spodoptera littoralis</i>	Toxic	470
Nomilin, limonin	Limonoid	<i>Aedes albopictus</i>	Larvicidal	477
Epifriedelinol, β -amyrin, glutinol, epifriedelinol	Pentacyclic triterpenes	<i>Leptinotarsa decemlineata</i>	Antifeedant	471
Betulinic acid	Pentacyclic triterpenes	<i>Tribolium confusum</i>	IGR ^a	478
Fraxinellone	Limonoid	<i>Mythimna separata</i> , <i>Culex pipiens</i>	Antifeedant, toxic	479

^aIGR, Insect growth regulation effects.

Alkaloids

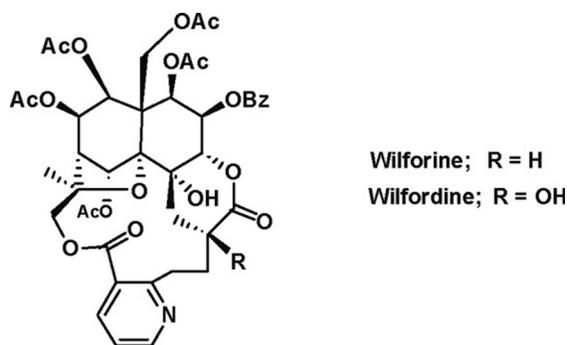
Alkaloids research contributes to our understanding of their ecological role and provides essential information on the structural requirements accounting for their insecticidal activity. While the direct use of these substances has recently diminished, they continue to serve as leads for synthetic analogs and are also indispensable biochemical tools in mode of action studies. However, the development of novel insecticides of commercial importance based on these prototypes is not readily predictable. Alkaloids are

Table 6 Insecticidal sterols for 2008–12 period (in part)

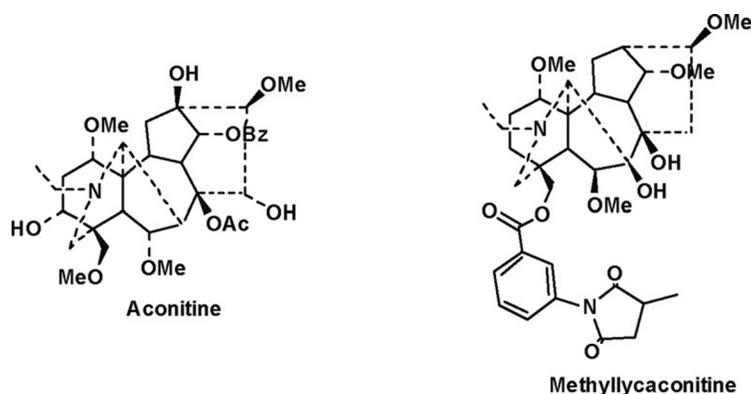
<i>Sterols</i>	<i>Class</i>	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
β -Sitosterol	Sterol	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Larvicidal	484
Stigmasterol	Sterol	<i>Spodoptera litura</i>	Citotoxic	481
β -Sitosterol -3-O- β -D-glucoside	Sterol glycoside	<i>Aedes aegypti</i> ,	Mosquitocidal	485
Ecdysterone, cyasterone	Ecdysteroids	<i>Oligonychus perseae</i> , <i>Bemisia tabaci</i>	Toxic	486
3 β -Sitosterol	Sterol	<i>Sitophilus zeamais</i>	Toxic	483
β -Sitosterol, stigmasterol	Sterol	<i>Tetranychus cinnabarinus</i>	Toxic	487,488
7 α -hydroxy- β -sitosterol, 7 α -methoxy- β -sitosterol	Sterol	<i>Leptinotarsa decemlineata</i>	Antifeedant	489

typically produced as a cocktail of metabolically related compounds and occasionally co-occur with other nonalkaloidal substances all modulating the toxicological properties of an individual component. Consequently, it would be fair to assume that a single natural compound is not optimized for a particular biological activity. Progress in the research on natural insecticides, botanicals in particular, has been surveyed from time to time.^{9,490,491} Specifically, Ujváry³⁹ has reviewed tobacco, lobeline, quinolizidine, unsaturated amides, veratrum, solanum, physostigmine (eserine), ryanodine, *Aconitum* and *Delphinium*, rocaglamide, cocaine, methylxanthines, isoquinoline-type, dioncophyllines, *Erythrina*, *Stemona*, *Trypterygium*, *Haplophyton* and polyhydroxy alkaloids, covering their insecticidal mode of action. There are recent reports on natural alkaloids from different chemical classes with insecticidal effects. For example, dihydroagarofuran alkaloids,^{264,492} didehydrostemofoline alkaloids from stemona plant,⁴⁹³ benzophenanthridine alkaloids,⁴⁹⁴ indoloquinazolin alkaloids,³²⁸ isubutilamide alkaloids,⁴⁹⁵ phenanthrene–indolizidine alkaloids,⁴⁹⁶ pyrrolizidine alkaloids⁴⁰⁴ and others.

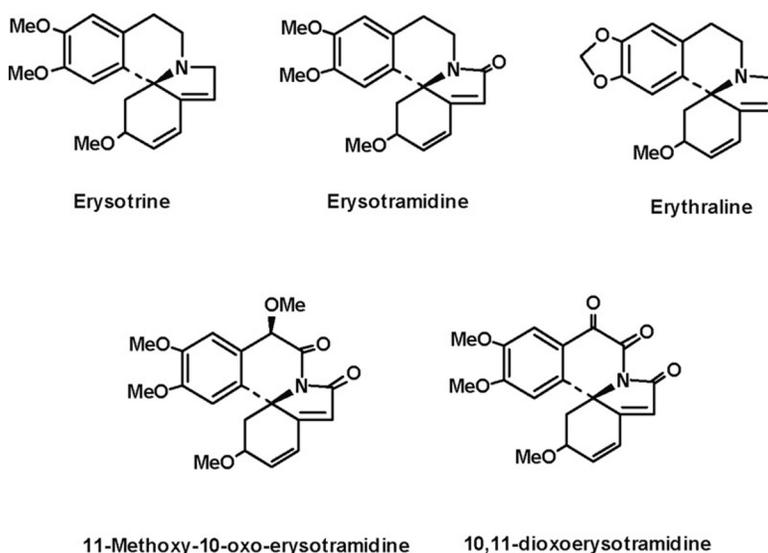
Dihydroagarofuran sesquiterpene esters and alkaloids are the main compounds exhibiting insect antifeedant and insecticidal activities which have already been isolated from species of Celastraceae. Insecticidal properties of *Trypterygium wilfordii* roots have been cited in the literature since 1931 and the sesquiterpene pyridine alkaloids wilforine and wilfordine were identified as its active components (reviewed by Liao⁴⁹⁷). Several macrolide pyridine alkaloids have recently been isolated from *Euonymus* spp. and *Maytenus* spp. (Celastraceae). The number and orientation of the ester groups and the existence of the pyridine alkaloids have a pronounced impact on the insecticidal activity of these dihydro- β -agarofuran sesquiterpene polyol alkaloids.^{397,498} Accordingly, the structure type of the nicotinic diacid and the components of the dihydro- β -agarofuran skeleton may affect the antifeedant potency of these macrolide alkaloids and could be involved in potential neuronal action of the nicotinic diacid.



Diterpenoid alkaloids are well known compounds of pharmacological interest. Aconitine, the major and one of the most toxic C19 norditerpene alkaloids isolated from *Aconitum napellus*, and methyllycaconitine, the principal toxic alkaloid of many *Delphinium* spp. but not found in *Aconitum* species are among the most toxic ones (see Ujváry³⁹). The insecticidal effects of C19 diterpene alkaloids and their effects on insect nicotine acetylcholine receptors (nAChR) were already known. Recent studies on the antifeedant effects of C19 norditerpenoid (NDAs) and C20 diterpenoid (DAs) alkaloids isolated from *Aconitum*, *Delphinium* and *Consolida* (Ranunculaceae) species showed that NDAs are better insect antifeedants and post-ingestive toxicants than the related DAs. Their antifeedant or insecticidal potencies did not coincide with their reported nAChR binding activity but did correlate with the agonist/antagonist insecticidal/ antifeedant model proposed for nicotinic insecticides. Among the most potent antifeedants are the NDAs 1,14 diacetylcardiopetaline, 18-hydroxy-14-O-methylgadesine and 14-O-acetyldelectinine and the DA 19-oxodihydroatisine (see Reina and González-Coloma).⁴⁹⁹



The *Erythrina* alkaloids isolated from flowers of *Erythrina latissima*, (+)-11 β -methoxy-10-oxoerysotramidine, (+)-10,11-dioxoerysotramidine, (+)-erysotrine, (+)-erysotramidine and (+)-erythraline, showed potent dose dependent antifeedant activity at concentrations ≥ 100 ppm against *Spodoptera littoralis* larvae (see Cornelius et al.).⁵⁰⁰



The piperamides are a class of compounds with strong insecticidal effects (see Scott⁵⁰¹). Many studies have shown the effectiveness of *Piper* spp. extracts for the control of stored products pests. Piperine, piperolein B, piperlonguminine, dihydropiperlonguminine, dihydropiperine, pipericide and guineensine, showed insecticidal activity against male *C. chinensis* and the cow pea weevil *Callosobruchus maculatus* (see Scott).⁵⁰¹

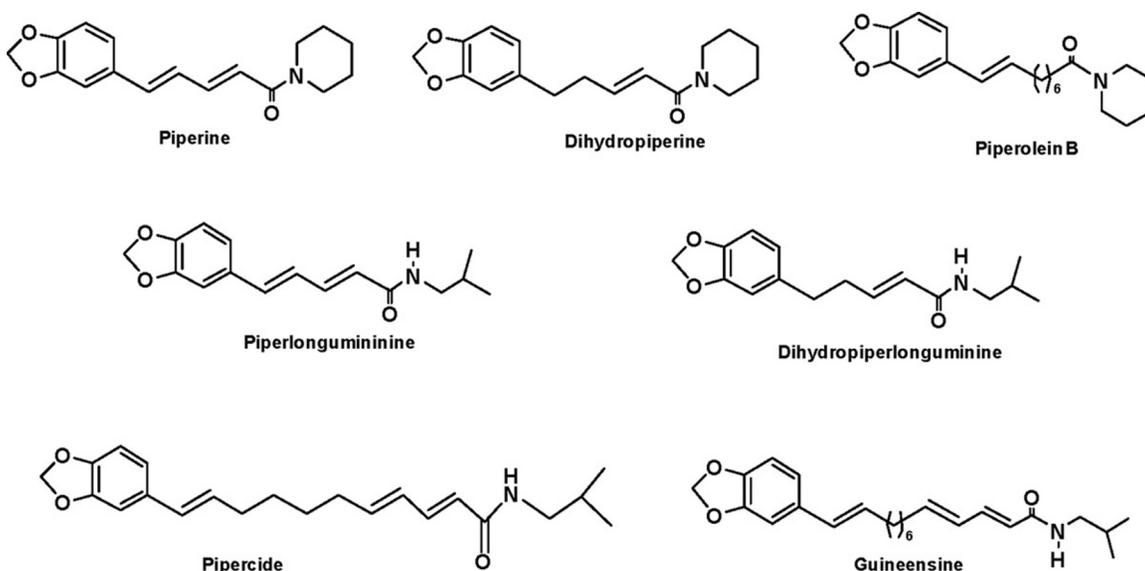
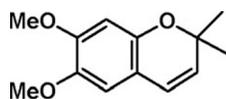


Table 7 shows the latest publications on the topic for the period 2006–13.

Elsevier Reference Module in Chemistry, Molecular Sciences and Chemical Engineering, (2013)

Flavones, Isoflavonoids, Chromenes, Coumarins, Iridoids, Lignans, Phenylpropanoids

Precocenes have notable effects on insect development and can specifically induce destruction of corpora allata cells thus preventing the synthesis of juvenile hormones. As juvenile hormones have wide-ranging physiological roles in insects from metamorphosis to reproduction, the effects of precocenes are also diverse. Precocene II and related compounds had morphogenetic, metabolic, toxic and antifeedant effects on several insect species.^{519–522}



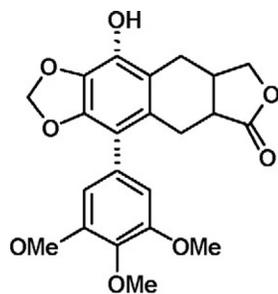
Precocene II

Lignans biogenetically-related secondary metabolites derived from phenylpropanoid precursors play a significant role in defending plants from insects. They mostly act as regulators of insect feeding but in a few cases they can also exert an influence on the specific physiological functions of insects. The mode of action of such compounds is mostly unknown.²⁶⁰ One possible mechanism might be interaction with and disruption of the endocrine system which is crucial for the proper development of insects and is dependent on the action of molting hormones (ecdysteroids).⁵²³ These compounds also affect feeding, excretion and *Trypanosoma cruzi* interactions with *Rhodnius prolixus*.⁵²⁴ More recent studies suggest that some lignans acted as tubulin polymerization inhibitors with activity against chewing pests⁵²⁵ and also could act on the excitatory receptor for acetylcholine.⁵²⁶ A structure-activity study revealed that natural lignan lactones with methoxy and/or methylenedioxy substituents showed significant activity that is strong enough to affect plant-insect interactions. Presence of polar substituents, especially hydroxyl or glycosyl groups, often reduces the activity. Non-polar substituents such as methoxy or methylenedioxy groups enhance the activity not only in lignans but also in simple phenylpropanoids.⁵²⁷ Wukisari et al.⁵²⁶ suggested that biological activities of lignans depend on the linkage of the phenyl propane, the oxidation patterns of the main structure, and the stereochemistry.

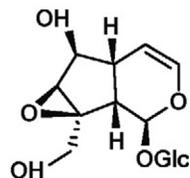
Coumarins are scantily studied insecticides and there is potential to exploit this chemically simple group of natural products.⁵²⁸ Iridoids are known to deter feeding or decrease the growth rate of many generalist insect herbivores. For example catalpol affected *Tribolium castaneum* growth probably related to the inhibitory activity of this iridoid against DNA polymerase.⁵²⁹ Phenylpropanoid derivatives accumulate in plants in response to insect herbivory and therefore are antiherbivore substances.⁵³⁰

Flavones and isoflavones can interfere with molting, reproduction, feeding, and behavior of many arthropods and their insecticidal and antifeedant activities have been documented.^{531–534}

Tables 8 and 9 show the latest reports on the insecticidal effects of the above mentioned type of compounds.



Podophyllotoxin



Catalpol

Glucosinolates and Isothiocyanates

Plants belonging to the order of *Capparales*, with particular reference to several species of the Brassicaceae family, possess an activated defence system consisting in two basic components: glucosinolates (non-toxic precursor compounds) and the activating myrosinases enzymes (glucosinolate-myrosinase system). When plant tissue damage (e.g. by chewing herbivore) abolishes the compartmentation, glucosinolates and myrosinases get mixed and glucosinolate activation is initiated resulting in the *in situ* formation of a number of hydrolytic compounds including isothiocyanates, nitriles, oxazolidinethione, ephthionitriles and organic thiocyanates.⁵⁵⁸ About more than 120 different glucosinolates are known to date. Their enzymatic hydrolysis products are responsible of many biocidal activities.^{559,560} The defensive function of the glucosinolate-myrosinase system has been mainly attributed to the isothiocyanates due to its toxic effects comparable to that of commercial insecticides.

Fatty Acids and Esters and Biogenetically Related Compounds

Essential for life, fatty acids are crucial in energy metabolism, cell and membrane structure, and physiological regulation to all organisms, including *Drosophila*.⁵⁶¹ The fatty acid compositions of all insect orders are fairly similar, in a qualitative way. Biochemical profiles include about eight components with chain lengths of 12 to 18 carbons, most of these saturated and

Table 8 Flavonoids, lignans, chromones, coumarins, etc. for 2006–13 period (in part)

Flavonoids, lignanes, etc	Type	Target insect	Action	Reference
Precocene II	Chromene	<i>Archips podana</i>	Modification of the insect sensory system	535
Isovitexin-2'-O- β -[6-O-E-p-coumaroylglucopyranoside]	Flavonoid	<i>Helicoverpa armigera</i>	Antifertility	536
(-)-Homopterocarpin	Isoflavonoid-	<i>Spodoptera litura</i>	Antifeedants	405
(-)-Methoxyhomopterocarpin	Pterocarpans	<i>Reticulitermes speratus</i>		
Quercetin glycoside	Flavonoid	<i>Spodoptera frugiperda</i>	Insecticidal	537
Tannins	Tannins		I GR ^a	
Kaempferol glycosides	Flavonoid	<i>Sitophilus oryzae</i>	Insecticidal	538
Tanetin (6-hydroxykaempferol 3,7,4'-trimethyl ether), 6-hydroxykaempferol 3,6-di-Me ether.	Flavone	<i>Spodoptera littoralis</i>	Antifeedant	412
Rutin	Flavone	<i>Anticarsia gemmatalis</i>	Antinutritional	539
Acacetin, galangustin	Flavone	<i>Rhopalosiphum padi</i> , <i>Myzus persicae</i>	Antifeedant	540
(-)-Kusunokinin	Lignan	<i>Anticarsia gemmatalis</i>	Toxic	541
Yangambin	Lignan	<i>Chrysomya megacephala</i>	Inhibition of postembryonic development, morphological alteration, and oviposition reduction	542
Leptostachyol acetate	Lignan	<i>Musca domestica</i>	Toxic	260
(-)-(8R,8'R)-3,3'-dimethoxy-9,9'-epoxylignane-4,4'-diol	Lignan	<i>Musca domestica</i>	Toxic	526
Dihydroguaiaretic acid, secoisolariciresinol	Lignan	<i>Culex pipiens</i>	Larvicidal	543
Nordihydroguaiaretic acid	Lignan	<i>Culex quinquefasciatus</i>	Larvicidal	544
Khellin, Visnagin, Ammiol	Chromone	<i>Spodoptera littoralis</i>	Antifeedant	545
2-Methyl-5,6,7-trimethoxychromone	Chromone	<i>Spodoptera litura</i>	Antifeedant	546
Sterigmatocystin	Xanthone	<i>Anopheles gambiae</i>	Larvicidal	547
Coumarin	Coumarin	<i>Rhyzopertha dominica</i> , <i>Sitophilus zeamais</i> , <i>Oryzaephilus surinamensis</i>	Insecticidal	548
Murraxocin	Coumarin	<i>Plecoptera reflexa</i> , <i>Clostera cupreata</i> , <i>Crypsiptya coclesalis</i>	Toxic	528
6-Hydroxy-7-isoprenyloxycoumarin, 6-Methoxy-7-isoprenyloxycoumarin, 6,7-Methylenedioxcoumarin, 5-Methoxy-6,7-methylenedioxcoumarin, 6-Methoxy-7-(2-hydroxyethoxy) coumarin	Coumarin	<i>Spodoptera frugiperda</i>	Antifeedant, toxic, I GR ^a	549
Scopoletin	Coumarin	<i>Spodoptera littoralis</i> , <i>Spilarcia obliqua</i>	Antifeedant	412
Imperatorin, osthole	Furanocoumarin coumarin	<i>Aedes aegypti</i> , <i>Culex pipiens pallens</i>	Antifeedant, I GR	550
Emodin	Antraquinone	<i>Anopheles gambiae</i> , <i>Bemisia tabaci</i>	Toxic, larvicidal	551
13-Hydroxyversicolorin B	Antraquinone	<i>Anopheles gambiae</i>	Larvicidal	547
1,4-naphthoquinone	Quinones	<i>Trichoplusia ni</i> .	Toxic	553
juglone, 2-methoxy-1,4-naphthoquinone, plumbagin, 2,3-dimethoxy-5-methyl-1,4-benzoquinone			Feeding deterrent	
Luteolin, genistein	Flavone, isoflavone	<i>Acyrtosiphon pisum</i>	Antifeedant	532
Santoflavone, eupatorin	Flavonoids	<i>Culex pipiens</i>	Larvicidal	531
Pinoembrin	Flavonoid	<i>Epilachna paenulata</i> , <i>Xanthogaleruca luteola</i> , <i>Spodoptera frugiperda</i>	Antifeedant	554

^aI GR, Insect growth regulation effects.

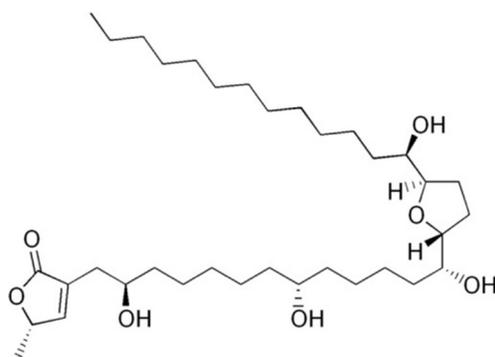
Table 9 Aromatic derivatives and organosulfur compounds for 2006, 2007 and 2008 (in part)

Aromatic derivatives, organosulfur	Type	Target insect	Action	Reference
[4-(Prop-2-enyl) phenyl angelate4-(3-methyloxiranyl) phenyl 2-methylbutyrate]	Phenylpropanoid	Aphicidal	Toxic	413
Anisole	Phenylpropanoid	<i>Pediculus humanus capitis</i> , <i>permethrin-resistant</i>	Toxic	201
<i>trans</i> -Anethole,	Phenylpropanoid	<i>Lycoriella ingenua</i> <i>Trichoplusia ni</i>	Toxic	95 395
Safrole	Phenylpropanoid	<i>Musca domestica</i>	Toxic	303
Methyl salicylate	Phenyl ester	<i>Trichoplusia ni</i>	Toxic	395
<i>p</i> -Anisaldehyde	Phenylpropanoid	<i>Lycoriella ingenua</i>	Toxic	95
Remirol	Dihydrobenzofurane	<i>Spodoptera litura</i>	Antifeedant	546
7-Acetyl-4,6-dimethoxy-2, 3-dihydrobenzofuran				
Syringin	Phenylpropanoid glucoside	<i>Sitophilus granarius</i> , <i>Trogoderma granarium</i> , <i>Tribolium confusum</i>	Antifeedants	411
Eugenol	Phenylpropanoid	<i>Ixodes ricinus</i>	Repellent	193
Eugenyl acetate		<i>Musca domestica</i>	Toxic, knock down	145,303
Cinnamaldehyde	Phenylpropanoid	<i>Musca domestica</i> <i>Chrysomya megacephala</i>	Knock down, toxic ovicidal	145,146
2-Phenylethanol	Phenethyl alcohol	<i>Ixodes ricinus</i> <i>Anopheles gambiae</i>	Repellent	193 555
Diallyl disulfide	Organosulfur	<i>Lycoriella ingenua</i>	Toxic	95
Diallyl trisulphide/tetrasulphide	Organosulfur	<i>Aedes aegypti</i>	Repellent	556
Dimethyl disulfide	Organosulfur	<i>Baltella germanica</i>	Toxic, fumigant	557

monounsaturated fatty acids (MUFAs) plus two polyunsaturated fatty acids (PUFAs) – Linoleic acid (LA, 18:2n-6) and α -linolenic acid (ALA, 18:3n-3).⁵⁶¹ The lipids of aquatic insects and some Antarctic beetles are abundant in long chain PUFAs with a chain length of 20 or 22 carbons. Certain C20 and C22 PUFAs play important regulatory roles in reproductive biology of some insect species. For example, arachidonic acid (AA, 20:4n-6) or certain structurally related C20 and C22 PUFAs are essential nutrients for several mosquito species,⁵⁶¹ which may be reflective of the mosquito diet compared to that of fruit flies. Fatty acids and their esters have been report as antifeedants against insect pests.^{489,562}

Oils may affect pests in several ways. Vegetable oils may block the insects air or breathing holes (spiracles), so the insect dies by suffocation. Oils prevent gas exchange through egg membranes, so eggs are often targets of control with oils. The fatty acids in oils may disrupt cell membranes and interfere with normal metabolism. Other oils may also have antifeedant properties or may clog stylets (stylet oils), which may help prevent insects, like aphids and leafhoppers, from transmitting viruses to plants. In general, oils are most effective against small, immobile or slow-moving, soft-bodied insects (e.g., aphids, scales, leafhopper nymphs, whiteflies) and mites that are thoroughly coated by an oil spray.

The Annonaceae family has drawn a lot of attention since the 1980's, due to the presence of acetogenins, of which their structural characteristics feature a variety of biological activities, where the insecticidal activity stands out. Acetogenins are a class of polyketide natural products. They are characterized by linear 32- or 34-carbon chains containing oxygenated functional groups including hydroxyls, ketones, epoxides, tetrahydrofurans and tetrahydropyrans. They are often terminated with a lactone or butenolide. Over 400 members of this family of compounds have been isolated from 51 different species of plants.⁵⁶³ Regarding their structure, the acetogenins comprise a series of natural products C-35/C-37 derived from C-32/C-34 fatty acids combined with a 2-propanol unit.



Chemical structure of annonacin

Acetogenins block the respiratory chain at NADH ubiquinone reductase (Complex I) and cause a decrease in ATP levels, affecting directly the electron transport in the mitochondria, causing apoptosis.⁵⁶² Acetogenins have been evaluated in several groups of insects of both medical and agricultural importance (Table 10).

Polyacetylene (IUPAC name: polyethyne) is an organic polymer with the repeating unit $(C_2H_2)_n$. Polyacetylenic natural products are a substantial class of often unstable compounds containing a unique carbon-carbon triple bond functionality, that

Table 10 Insecticidal fatty acids, esters and biogenetically related compounds for 2006–13 period (in part)

<i>Fatty acid/esters and related compounds</i>	<i>Class</i>	<i>Target insect</i>	<i>Action</i>	<i>Reference</i>
Spiroacetals	Acetylenic spiroacetal enolethers	<i>Spodoptera littoralis</i> , <i>Rhopalosiphum padi</i>	Antifeedant	564
Toosendanin	Alkyl/ alkenylacyloxy derivatives	<i>Mythimna separata</i>	Insecticidal	565
Allyl cinnamate, allyl 2-furoate, allyl heptanoate, allyl hexanoate	Allyl esters (fatty and aromatic acids)	<i>Tribolium castaneum</i>	Repellent, toxic	566
Hexadecanoic acid ethyl ester, octadecanoic acid ethyl ester, eicosane	Fatty acids esters	<i>Bemisia tabaci</i>	Toxic	567
Nonanoid acid, decanoid acid, dodecanoid acid,	Fatty acids	<i>Leptinotarsa decemlineata</i> , <i>Myzus persicae</i> , <i>Rhopalosiphum maidis</i>	Antifeedant	489
Methyl hexadecanoate, ethyl hexadecanoate	Fatty acids esters	<i>Myzus persicae</i>	Antifeedant	489
Ethyl esters (22:1)	Fatty acids esters	<i>Myzus persicae</i>	Toxic	568
Linolenic acid, linoleic acid	Fatty acids	<i>Spodoptera frugiperda</i>	Insectistatic	569
Oleic acid, palmitic acid, stearic acid	Fatty acids	<i>Spodoptera frugiperda</i>	Insectistatic	570
(2n-octylcycloprop-1-enyl)-octanoic acid	Cyclopropene fatty acid	<i>Sitophilus oryzae</i> , <i>Callosobruchus chinensis</i> , <i>Tribolium castaneum</i>	Contact and Fumigation Toxicity	571
Oleic acid, palmitic acid, linoleic acid	Fatty acids	<i>Helicoverpa armigera</i>	Toxic	572
Methyl palmitate	Fatty acid ester	<i>Tetranychus cinnabarinus</i>	Toxic, acaricidal	573
Octanoic acid	Fatty acid ester	<i>Varroa destructor</i>	Repellent	574
Octadecyl propanoate, heptadecyl butanoate, hexadecyl pentanoate, and tetradecyl heptanoate	Fatty acid esters	<i>Anopheles stephensi</i>	Oviposition Repellents	575
Oleic acid, linoleic acid	Fatty acid esters	<i>Aedes aegypti</i> , <i>Anopheles stephensi</i> , <i>Culex quinquefasciatus</i>	Larvicidal	576
Lauric acid, palmitic acid, stearic acid, oleic acid and linolenic acid	Fatty acid esters	<i>Culex quinquefasciatus</i>	Larvicidal	577
Itrabin, molvizarin, squamocin	Acetogenins	<i>Ceratitis capitata</i>	Oviposition disorders	578
Annonacin, cis-annonacin-10-one, densicomacin-1. Gigantetronenin, murihexocin-B (5), tucupentol	Acetogenins	<i>Spodoptera frugiperda</i>	Antifeedant, Toxic	579
Annonaceous acetogenin	Acetogenin	<i>Acalymma vittatum</i>	Feeding deterrent	580
Cyclic enyne (12E)-cis-maneonene-E	Acetogenin	<i>Tribolium confusum</i> , <i>Culex pipiens</i>	Larvicidal	581
Sylvaticin, rolliniastatin-1, rolliniastatin-2, motrilin, desacetyluvaricin	Acetogenins	<i>Spodoptera frugiperda</i>	Toxic	582
Rolliniastatin-1	Acetogenin	<i>Spodoptera frugiperda</i>	Larvicide, post ingestive Toxicity	583
Squamocin, molvizarin, itrabin, almuñequin, cherimolin-1, annonacin, annonacin-A, densicomacin-1, cis-annonacin-10-one and murihexocin-A	Acetogenins	<i>Oncopeltus fasciatus</i>	Toxic	584
Itrabin, asimicin, neoanonin, cherimolin-1, cherimolin-2, almuñequin, motrilin, and tucumanin	Acetogenins	<i>Spodoptera frugiperda</i>	Antifeedant	585
Echinopsacetylene A	Polyacetylene thiophene	<i>Formosoa subterranean</i>	Toxic	586
(E)-7-(2,4-hexadiynylidene)-1,6-dioxaspiro[4.4]nona-2,8-dien-4-ol	Spiroketal enol Polyacetylene	<i>Spodoptera littoralis</i>	Toxic	587

are intriguing for their wide variety of biochemical and ecological functions. The majority of these compounds are derived from fatty acid and polyketide precursors. The majority of reports on polyacetylene bioactivities include screens for anticancer agents. Also these compounds are toxic, antifeedants, allelochemicals, phytoalexins or antibiotics. A review focusing on the biological activities of naturally occurring acetylenes as insect divertants, fungicides, allelopathic species, insecticides, and pharmaceuticals has appeared.⁵⁸⁸ While evidence supports that acetylenic (and acetylenederived) thiophenes function as sensitizers for the production of singlet oxygen, most polyacetylenes have light-independent functions.⁵⁸⁹ Insecticidal polyacetylenes have been recently reported (Table 10).

Sustainable Production: Culture Methods

The main problem faced in the exploitation of natural compounds of plant origin as biopesticides is to ensure their sustainable supply at low cost. Biopesticides and botanicals tend to be more expensive than synthetics, and some are not produced in great quantity or are no longer commercially available (e.g. nicotine). Several sources of plant material may be used for botanical pesticide extraction. The simplest route is extraction from plants harvested from wild plant resources. However, wild plant resources may be limited and hence may not permit sustainable production. Moreover, some plants containing these compounds are endangered species due to over exploitation.

An alternative is plant cultivation using conventional agricultural methods. Traditional cultivation permits the sustainable production of plant material in the amount required for biopesticide production and the ongoing improvement of production levels through the breeding and selection of superior genotypes. The investment and the long periods of time required to establish plantations as well as environmental factors such as adverse weather conditions, pests and diseases are the main disadvantages. It may also be that plants with interesting activities only grow in certain regions and are difficult to cultivate outside of their local ecosystems (Bourgau, 2001). Additionally, some interesting compounds accumulate in specialized tissues such as pyrethrins in flower heads of crysanthemum resulting in high labor costs related to harvest and extraction.⁵⁹⁰

For plant species with interesting activity, sustainable and reproducible cultivation methods should be developed as a clear alternative to traditional agriculture or wild plant collection. In the last few decades, great progress has been made in plant cell cultivation for the production of botanical insecticides.^{590,591} Plant cell culture is not affected by changes in environmental conditions and the plant material can be maintained indefinitely in a defined production system. Strategies based on plant cell cultivations,⁵⁹² the use of elicitors^{593,594} and metabolic engineering techniques⁵⁹⁵ have been recently used for improving the yield and productivity of botanical insecticides in plant cell culture.

There are many studies in the literature involving technological, economical and scale-up aspects of large scale cultivation of plant cell suspension cultures under controlled conditions in bioreactors.⁶⁵² Despite of all considerable efforts, their exploitation commercial as an industrial process have not been effectively exploited due to problems associated to products yield, slow growth rates of plant cell cultures and low economical viability.

As an alternative to plant cell cultures, the use of organ cultures such as adventitious root cultures and fast growing hairy roots obtained after transformation with *Agrobacterium rhizogenes* offer new opportunities for a sustainable *in vitro* production of specific metabolites when the main location of metabolite biosynthesis is in the roots. These cultures are genetically stable for long periods of time in contrast to what has been observed in many plant cell cultures and can produce levels of metabolites comparable to those of intact plants. The potential of adventitious and hairy root cultures as a stable source of biologically active compounds indicate that the industrial exploitation of these culture systems may be possible through up scaling in bioreactors.^{596–600} Studies on the production of some commercially important botanical insecticides by means of hairy root cultures have been carried out. Some examples are azadirachtin (*Azadirachta indica*,^{601,602}), gossypol (*Gossypium hirsutum*,⁶⁰³), thiophenes (*Tagetes patula*,⁶⁰⁴), phytoecdysteroids (*Ajuga reptans* var. *atropurpurea*⁶⁰⁵ *Ajuga turkestanica*,⁶⁰⁶) and nicotine (*Nicotiana rustica*,^{607,608}). The production of Azadirachtin by hairy root culture of *Azadirachta indica* under optimized culture conditions has also been studied in different types of bioreactors.⁶⁰⁹

Additionally, this biotechnological technique can be used as a source for the discovery of new pesticides in roots of rare and endangered species that would otherwise be inaccessible. *Salvia broussonetii*, a Canarian endangered endemic species that produces triterpenes in the aerial parts^{610,611} is a good example. The phytochemical study of hairy culture has permitted the isolation of the dehydroabietane diterpenes 14-deoxycoleon U and demethylcryptojaponol with antifeedant effects against *Leptinotarsa decemlineata*. Additionally, these diterpenes showed strong selective cytotoxicity to insect Sf9 cells.⁶¹²

Another example is *Artemisia granatensis*, an endemic endangered plant species from Sierra Nevada (Spain). The cultivation under controlled conditions in artificial substrate of whole plants and *in vitro* transformed roots with *Agrobacterium rhizogenes*, have provided a biotechnological tool for the sustainable production of this plant and its metabolites. The study of its chemical composition showed the presence of monoterpenes, sesquiterpenic lactones and poliacetylenic spiroacetals with important antifeedant activities against aphids.⁵⁶⁴

Aeroponically grown plants in controlled environments can also be a sustainable source of metabolites from roots and aerial parts.⁶¹³ This artificial system allows the control of the root nutrient and water regimes, and also offers full access to the roots throughout the life of the plant. At the present time, aeroponic culture provides opportunity for biomass production on a commercial scale and it is being mainly applied to the obtaining of medicinal compounds.^{93,614–616}

As part of our ongoing studies on the sustainable production of natural biopesticides from endemic species, we have developed an aeroponic system for *Persea indica*. The aerial part and stems of this species are characterized by their content in insecticidal ryanodane and isoryanodane-type diterpenes.^{431,432,617} Aeroponic culture of this protected tree in a controlled environment has allowed to study the production of ryanodanes in aerial parts and roots.

Essential oils and major components from aerial parts of a Spanish population of *Artemisia absinthium* cultivated aeroponically and under controlled conditions in a growth chamber have been reported as having insect antifeedant properties. The main components produced in the organic extract were the sesquiterpene lactone hydroxypelenolide and the flavones artemetin and casticin.⁵³³ The oils were characterized by the presence of *cis*-epoxyocimene, chrysanthenol, and chrysanthenyl acetate and (Z)-2,6-Dimethyl-5,7-octadien-2,3-diol.⁷⁸

The New Biopesticide Market

The demand for nature-based biopesticides is rising steadily in all parts of the world. This is because of increased public awareness of the environment, and the pollution potential and health hazards related to many conventional pesticides. Extensive and systematic research has enhanced the effectiveness of biopesticides. Also, the techniques for their mass production, storage, transport and application have improved in recent years.

Biopesticides are safer than conventional pesticides that often are hazardous chemicals. They offer much more activity targeted to the desired pests as opposed to conventional pesticides that often affect a broad spectrum of pests as well as birds and mammalian species. Often they are effective in very small quantities, thereby offering lower exposure. They decompose quickly. Lastly, they can supplement conventional pesticides when used in integrated pest management (IPM) programs. Such programs offer high crop yields while dramatically reducing conventional pesticide use.

The natural crop protectants market is currently expanding in Western Europe and North America. Two important factors are the need for residue free crops and the expansion of organic farming. In addition, the challenge of new and more stringent chemical pesticide regulations, combined with increasing demand for agriculture products with positive environmental and safety profiles, is boosting interest in natural crop protectants.¹⁹ A recent market analysis by Frost & Sullivan (<http://www.chemicals.frost.com>): *North American & Western European Biopesticides Marke* (microbial, botanicals and biocontrol) showed \$594.2 millions of benefits in 2008, estimating \$1020.2 millions by 2015.

The European market represents 45% of the total demand. Due to environmental side effects and health concerns, many synthetic pesticides have been banned (Council Directive 91/414 EC) or are being under evaluation (Regulation 2009/1107/E and Directive 2009/128/EC). Additionally, the DIRECTIVE 2009/128/EC of the European Parliament and of the Council of the European Union establishing a framework for Community action to achieve the sustainable use of pesticides is now a National Normative (December 2011). Based on the Normative all Member States must adopt the necessary measures to minimize pesticide inputs giving priority to alternative control methods as part of an Integrated Pest Management (IPM) approach. This legal framework is based on the challenges and problems that modern agriculture is facing: the loss of productivity caused by pests and weeds which has been controlled by synthetic pesticides (SPs). The use of SPs allowed for increased crop yields but it has also generated environmental and health problems along with increased pest resistance and loss of biodiversity.⁶¹⁸

Furthermore, recent reports on the long-term negative effects of neonicotinoid insecticides on bees and the impact of these effects on pollination-dependent crops⁶¹⁹ has raised public concern on the use of such pesticides. The European Commission is proposing to suspend the use of imidacloprid, thiamethoxam and clothianidin on the basis of evidence from the European Food Safety Authority that the chemicals may be harmful to bees. The ban would apply to crops that attract bees, including sunflower, rapeseed and cotton, but this could have a major impact for many other crops in the near future. A BCC Research Corporation (<http://www.bccresearch.com/report/CHM029B.html>) report has estimated that horticultural production consumes 55% of the available biopesticides. Biopesticides are also used in public health (vector control) and forestry. Organic farming with an annual growth of 30% in the EU is one of the mayor consumers of natural crop protectants (Council Regulation No 2092/91/EEC). Therefore, the biopesticides market is expanding and there is an increasing need for new products.

Registration of Natural Products as Crop-Protection Agents

Requirements for the United States

Federal law requires that before selling or distributing a pesticide in the United States, a person or company must obtain a registration, or a license, from the Environmental Protection Agency (EPA). For registration, the EPA has currently separate pesticides into three categories: antimicrobials, biopesticides, and conventional.⁶²⁰ According to EPA (<http://www.epa.gov/pesticides/biopesticides/>), biopesticides include naturally occurring substances that control pests (biochemical pesticides), microorganisms that control pests (microbial pesticides or biocontrol agents), and pesticidal substances produced by plants containing added genetic material (plant-incorporated protectants, or PIPs).

Natural products generally fall into the second category, and the EPA has specified test requirements for registration in the USA in 'Guidelines for Biorational Pesticides' (Subdivision M of CFR 158).⁶²¹ Biochemical pesticides are distinguished from conventional chemical pesticides by their natural occurrence and non-toxic mode of action to the target pest. Thus insect pheromones and

plant-growth regulators, such as auxins and gibberelins, are defined as biochemical pesticides; active pesticide ingredients from common food sources such as garlic and cinnamon are also defined this way. However, plant extracted pesticidal materials, although of natural origin do not necessarily always have a non-toxic mode of action. In some cases the mode of action cannot be elucidated, and the best available scientific information and knowledge have then to be used to make the most appropriate decision. Semiochemicals (pheromones, either naturally occurring or synthetic) were also recognised by EPA as having low risks associated with their use. EPA has favoured biopesticides under the reduced-risk pesticide policy, has agreed to waivers to many of the study requirements and has agreed not to establish tolerances for many of the biopesticides.

During the period 1997–2010, EPA registered approximately 168 new active ingredient biopesticide registrations. Among these, 92 were categorized as natural products, 75 as biologicals, and one as synthetic natural product derived, while none were synthetic (See Cantrell et al.).⁶²⁰

Requirements for Europe

Europe uses the OECD definition of biopesticides, which includes pheromones, insect and plant growth regulators, plant extracts, transgenic plants and macro-organisms, as well as micro-organisms.⁶²² Regulatory control of biopesticides in Europe has been based on precedents and standards established in the same way as for chemicals. With the development of the European Registration Directive 91/414/EEC and the Biocidal Products Directive covering requirements for chemicals and micro-organisms, attempts have been made to harmonise the requirements and the interpretation of registration data throughout Europe.^{623,624} The Directive covers biopesticides, and data requirements are listed in Parts A (chemicals, pheromones, plant extracts) and Parts B (microorganisms-bacteria, fungi, protozoa, viruses and viroids) of Annexes II and III. The data requirements set out in these annexes appear to be very similar to those already agreed for chemicals, the requirements being fairly extensive to ensure that they cover all possible risk scenarios.

Conclusions

The main barriers to the commercialization of botanical insecticides are sustainability of the resource, standardization of chemically complex extracts, and regulatory approval. Additionally, finding new natural insecticides is not easy or currently being granted financial support as can be concluded from the lower number of publications on natural products with insecticidal properties (and mostly known ones) for the period 2006–08 (April) in contrast to the large number of publications on insecticidal essential oils.

Plant essential oils and/or their components have a broad spectrum of activity against insect and mite pests, plant pathogenic and other fungi, and nematodes. As such, they have considerable potential as crop protectants and for pest management in other situations (e.g. urban pest control). Current information indicates that they are safe to the user and the environment with few exceptions. However, the essential oils that are most effective against pests are often the most phytotoxic. This latter property requires serious attention when formulating products. Moreover, selectivity among invertebrates is not well documented.

Among new natural products with promising insecticidal properties we believe that, in addition to limonoids, attention should focus on the β -dihydroagarofuran sesquiterpenes and related pyridine alkaloids, silphinene type sesquiterpenes, drimanes, ryanodane diterpenes (more so than their pyrrole derivatives), lignans, flavonoids and phenylpropanoids, among others. However, new single compound-based natural insecticides are difficult to produce because compound isolation and identification takes time and effort, the alternative being the production of standardized extracts once the active compounds are identified. New extraction methods to produce standardized enriched extracts and biotechnological/traditional cultivation methods are needed to produce new botanical biopesticides with commercial potential.

Like other alternative pest management products, essential oil-based pesticides and enriched standardized extracts will not be a panacea for crop protection, but there should be substantial market niches, particularly certified organic farming and urban pest control.

Regulatory approval in industrial nations is costly and time consuming. However, there is a growing demand for organic production of food and the number of pest management products that can be used in this production is limited and it is here that botanical biopesticides can play an important role partially meeting such demand.

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