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Review

Controlling Stored Products' Pests with Plant Secondary Metabolites: A Review

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Abstract: To date, only a handful of pesticides have been authorized by the European Council for the protection of stored grains. Resistance issues and ecotoxicity concerns necessitate the development of ecofriendly tools in that direction. In this review, we refer to the recent findings on plant extracts and pure plant-derived substances with promising biological activity and the potential to be used as biopesticides for stored products. The main aim of biopesticides is to be effective against target pests, without harming humans and the environment. Many plant species, among those reported herein, are part of the human diet, and are thus not harmful to humans. Edible plant extracts produced with inorganic solvents represent safe candidates for use as repellants, fumigants or contact pesticides. Cinnamon, rosemary, parsley, garlic, oregano and basil are found in products destined for human consumption but also display significant biological activities. Interestingly, cinnamon is one of the most widely tested botanical matrixes, exhibiting the best lethal effects on almost all insect and mite taxa reported herein (Acaroidea, Coleoptera and Lepidoptera), followed by basil and garlic. *Prunus persica*, *Azadirachta indica* A. Juss and *Carum* sp. seem to be very promising too as miticides and/or insecticides, with *A. indica* already being represented commercially by a plant-derived acaricidal formulation.

Keywords: biopesticides; plant extracts; stored grain pests; insects; mites; grains; commodities

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1. Introduction

Over 800 million people worldwide suffer from malnutrition, and one-third of global food production is lost or wasted annually [1]. Pesticides are needed to control weeds, insect infestations and various pests and disease carriers (e.g., mosquitoes, ticks, rats and mice) in houses, offices, malls and streets [2]. Worldwide, approximately 2 million tons of pesticides are utilized annually [3]. However, the modes of action of pesticides are not always species-specific and exposure at even very low levels may have adverse health effects on humans. Additionally, concerns have been raised about environmental risks associated with exposure to these products through various routes (e.g., residues in food and drinking water) [2]. As a result, many pesticides have been withdrawn from the market. Interestingly, in an eight-year period (2001–2008), 26% of insecticides were banned from the European Union due to unintentional impacts [4], extending as far as unbalancing/damaging an entire ecosystem [5].

Due to the constant increase of the population, larger quantities of cereals are required to cover the needs of the growing human population, thus making it important to find ways to minimize the loss of stored grains. Cereal grains are in fact the most basic ingredients of the human diet. According to the FAO, 'Cereals continue to be by far the most important source (in terms of calories) of total food consumption'. The food use of cereals has continued to increase, albeit at a decelerating rate. In developing countries, the per capita average is now 173 kg, providing 56 percent of total calories [6]. On the other hand, insects feeding on stored grains can cause losses of up to 420 million tons annually [1]. There are two classes of arthropods—arachnids (Arachnida) and insects (Insecta)—infesting grains and food products. Within the first class, the subclass mites (Acari) contains some species of great economic importance. Among insects, species belonging to the orders of beetles (Coleoptera), moths and butterflies (Lepidoptera) are significant pests. Stored products may be damaged either through contamination by primary pests or through secondary contamination due to previously damaged grains; either caused by other pests (primary pests) or by being poorly threshed, dried and handled. Flour and milled rice might exhibit a reduction in weight and a decrease in quality due to an infestation of pests. Insects can encourage mold germination, increase the fatty acid percentage in the grain and can cause grain rancidity due to the uric acid they release, as well as causing grain pollution through their exuviae or feces. This, in consequence, can cause price discounts and shipping restrictions [1]. Traditionally, stored product insects have been controlled with synthetic insecticides, most of which are now out of the market due to ecotoxicological concerns (EC 1107/2006), as well as resistance issues [7,8]. To date, only a few of their active ingredients are still registered in the EU for stored product pest control. In particular, these include the fumigants magnesium phosphide and aluminum phosphide; the synthetic pyrethroids cypermethrine and deltamerthine, the phosphorothioate pirimiphos methyl and piperonyl butoxide applied by dusting [7]. Natural molecules of botanical origin have attracted international research interest in recent years as ecofriendly alternatives to their synthetic pesticidal ancestors (in commercial terms) [9]. Recent reviews on the use of the secondary metabolites of plants against stored product insects are those of Rajendran and co-workers, reporting the fumigant toxicity results conducted with essential oils of plants (mainly belonging to Apiaceae, Lamiaceae, Lauraceae and Myrtaceae) and their components (cyanohydrins, monoterpenoids, sulphur compounds, thiocyanates and others) [10], as well as that of Stejskal and co-workers, reporting on gas, liquid, gel and solid formulations of natural pesticides for stored-product applications [11].

In the present review, we report on the most important arthropod pests affecting grains and food products, along with the plant-derived substances reported to exhibit significant activity in the last 10 years. We categorized pests based on their taxonomic class and order, exhibiting similar habits, life cycles and metabolism. We aim to pinpoint the natural plant-derived substances that could be developed to combat stored-product pest infestations.

2. Economically Important Groups of Stored-Product Pests and Plant-Derived Tools with Reported Activity

2.1. Mites (Acari)

There are several mite species that cause severe losses to grains and stored products, mostly because they reproduce in large numbers, tolerate lower temperatures than insects and are not readily seen, causing great impact on storage facilities. Among the mites that are regarded as the most important pests of stored grains and food products are the species *Gohieria fusca* and *Lepidoglyphus destructor* (Glycyphagidae); *Blomia freeman* (Echimypodiae) and *Chortoglyphus arcuatus* (Chortoglyphidae), as well as *Aleuroglyphus ovatus*, *Tyrophagus longior*, *Tyrophagus putrescentiae*, *Tyroborus lini* and *Acarus farris* and *Acarus siro* of the Acaridae [12–15].

Traditionally, mites have been eradicated using synthetic miticides such as organophosphates, pyrethroids, pyridazines, juvenile hormone analogs and chitin synthesis inhibitors [13,16,17]. In addition, elevated CO_2 concentrations are applied [18]. However, benzyl-benzoate, a substance that is produced and used industrially but which is also naturally produced by a range of plants—cinnamon and cassia (*Cinnamomum* spp.), carnation (*Dianthus caryophyllus* L.), hyacinths (*Hyacinthus* spp.), tuberose (*Agave amica* Medik.), common jasmine (*Jasminum officinale* L.) and Santos mahogany (*Myroxylon balsamum* L. Harms)—is often used as a commercial acaricide or as a reference substance in acaricidal tests. However, this substance is regarded as an allergen [19].

Azadirachtin (neem), a commercially available plant-derived pesticide obtained from the plant species *Azadirachta indica* A. Juss, is one of the most commonly used natural substances that causes lethal and sublethal insecticidal and miticidal effects. As reviewed by Collins (2006), it limits the growth of populations and causes mortality of *T. putrescentiae*, *A. siro* and *L. destructor* [13] but it proved to be less effective against five species of stored product mite pests than synthetic commercial miticides [20].

The components of clove bud (*Eugenia caryophyllata* Thumb.) oil—methyleugenol (median lethal dose (LD_{50}) = 1.18 μg/cm^2, isoeugenol, ß-caryophyllene, eugenol and α-humulene (LD_{50} = 12.90 μg/cm^2)—showed lethal toxicity against *T. putrescentiae*, with methyleugenol and isoeugenol being more toxic than benzyl benzoate [21]. Furthermore, *Rosmarinus officinalis* L. essential oil (EO) and its constituent compounds were active against this mite, both as a fumigant (LD_{50} = 8.24 μg/cm^3) and in contact toxicity (LD_{50} = 5.49 μg/cm^2). The constituent camphor appeared to be successful in combating the pest (fumigant toxicity LD_{50} = 2.25 μg/cm^3, contact toxicity LD_{50} = 1.34 μg/cm^2) more effectively than benzyl benzoate (LD_{50} = 12.56 μg/cm^3, and 9.03 μg/cm^2) [22]. Other constituent substances in rosemary oil are α-pinene, 1,8-cineole and camphene, all exhibiting miticidal activity [23]. Lee (2015) proved the fumigant and contact toxicity of the essential oil of *Ligustrum japonicum* leaves against *T. putrescentiae* and calculated the respective LD_{50} values to be 16.48 μg/cm^3 and 8.02 μg/cm^2, respectively. α-pinene is one of the compounds found in *L. japonicum* oil, showing the highest percentage [24]. These data indicate that camphor may be effective, as well as other compounds. However, camphor seems to be more toxic to mites than α-pinene [21]. Ottoboni et al. (1992) reported that caraway (*Carum carvi* L.) essential oil was a significant candidate to combat *L. destructor*, *G. fusca*, *A. siro* and *T. putrescentiae* [25,26]. Furthermore 3,4-methylenedioxybenzene and its derivatives were described as successful miticides against, among others, *T. putrescentiae* [27]. Interestingly, apiol, which naturally occurs in the seeds of parsley (*Petroselinum sativum* Hoffm), was not toxic against *T. putrescentiae*, although it has been proven to be active against Dermatophagoides species. Essential oils obtained from both the aerial parts and seeds of the forget-me-not plant (*Myosotis arvensis* (L.)) or ingredient compounds used individually, namely 2,4,5-trimethylbenzaldehyde, 2,4-methylbenzaldehyde, 2,5-dimethylbenzaldehyde, 2-methylbenzaldehyde, 2,3-dimethylbenzaldehyde, 3-methylbenzaldehyde, 4-methylbenzaldehyde, 3-octanone, butyl isothiocyanate and nonanal, showed significantly greater contact and vapor toxicity against *T. putrescentiae* than benzyl benzoate [28]. *T. putrescentiae* was proven to be susceptible to the essential oil of garlic (*Allium sativum* L.), basil (*Ocimum basilicum* L.) and fenugreek (*Trigonella foenum-graecum* L.), in descending order, when exposed for one to three days [29]. However, the sulfide-rich garlic essential oil was toxic to Cheyletus malaccensis, a predatory mite and natural enemy of *A. siro*, *T. putrescentiae* and *L. destructor* [30]. Nonetheless, garlic essential oil and its active compounds can be used as possible miticides against a range of mites [31]. Unfortunately, some botanical extracts have been proven to be lethal for beneficial insects, such as citronella, eucalyptus, garlic, pyrethrum and neem. Sometimes their effects may be non-lethal, such as inhibiting natural enemies from utilizing prey, reducing prey availability, decreasing reproduction, inhibiting the ability of natural enemies to recognize prey, influencing the sex ratio (females:males) and reducing mobility. Nonetheless, detailed knowledge of the lethal or non-lethal effects of botanical pesticides on beneficial insects is essential for the sustainable control of insect pests and pollination activities for improved and sustainable agricultural production [32].

De Assis and co-authors (2011) tested the fumigant toxicity of eugenol and essential oils from cinnamon (*Cinnamomum zeylanicum* Blume), Surinam cherry (Eugenia uniflora L.), uvalha (*Eugenia uvalha* Cambess.), weeping paperbark (*Melaleuca leucadendra* (L.)), cake bush (*Piper marginatum* Jacq.) and Brazilian peppertree (*Schinus terebinthifolia* G.Raddi) against *T. putrescentiae*. The lowest median lethal concentration (LC_{50}) values were obtained for eugenol and *C. zeylanicum* essential oil. These data are in agreement, since eugenol is the major component of *C. zeylanicum* essential oil and its content may exceed $\frac{3}{4}$ of all the volatile ingredients of this oil [33]. The next most abundant component—linalool—is present in about nine times lower amounts [34]. Although it is not as important, linalool may increase toxic effects since it appeared to be toxic against *T. longior* in contact and fumigant toxicity studies. Similarly the toxicity of menthol, menthone, fenchone, linalyl acetate and eucalyptol, the most abundant substances in the essential oils of lavender (*Lavandula angustifolia* Mill, *Lavandula stoechas* L.), peppermint (*Mentha x piperita* L.) and eucalyptus (*Eucaliptus globulus*), should be assessed for their lethality [35].

The observation of the variable susceptibility of various species of mites to natural substances is also seen in the case of natural aldehydes used as miticides. This phenomenon was described for three natural aldehydes, namely, (2E)-hexenal, (2E, 6Z)-nonadienal and (2E)-nonenal, produced in plants from organic acids, when tested in feeding tests from 36 to 314 mg/g against *A. siro, A. ovatus* and *T. putrescentiae*. Specifically, the susceptibility of A. siro was similar for all three aldehydes, whereas *T. putrescentiae* was about eight times more susceptible to nonadienal than to hexenal, and was not significantly affected by nonenal [36]. In another study, benzaldehyde (LD_{50} = 4.23 μg/cm^2) isolated from the peach *Prunus persica*, as well as salicylaldehyde (LD_{50} = 1.02 μg/cm^2), cinnamaldehyde (LD_{50} = 1.66 μg/cm^2) and phthaldialdehyde (LD_{50} = 5.16 μg/cm^2), were tested against *T. putrescentiae*. All aldehydes exhibited better efficacies than benzyl benzoate (LD_{50} = 9.75 μg/cm^2). However, the values calculated for *P. persica* essential oil (LD_{50} = 11.23 μg/cm^2) were higher than those for benzyl benzoate [37]. This research shows that aldehydes and essential oils can be applied in grain and food protection. However, their application must be carefully adjusted to the tested species.

There are suggestions that jasmonic acid (JA) can be used as a miticide against mite-pests of grains and stored food. This compound may affect mite reproduction and limit losses. The tomato mutants that were unable to accumulate JA were characterized by a higher rate of egg-hatching of mite-pests compared to the wild type. Therefore, JA was suggested as a substance of ovicidal activity [38]. Most interestingly, JA additionally attracts predatory mites and therefore it may decrease the level of pests [39]. These data seem to open a field of interesting further research on the species of interest.

Angiosperms also deliver bioactive substances that may be useful in limiting losses caused by mites. For instance, essential oils obtained from gymnosperm plants such as *Pinus pinea, Pinus halepensis, Pinus pinaster* and *Pinus nigra* were described as toxic for *T. putrescentiae* [40], with *P. pinea* being the most effective. Moreover, 1,8-cineole and limonene showed miticidal activity when tested at 8 or 6 μL on 6 cm of filter paper. Likewise, *Juniperus chinensis* essential oil and its respective components were applied in impregnated disc biotests against *T. putrescentiae*, with the LD_{50} values calculated at 38.1, 15.33, and 42.85μg/cm^2 for the essential oil, bornyl acetate and α-eudesmol, respectively [41]. Based on the effect and the content of the substances in the oil, the authors suggested that bornyl acetate is the major substance responsible for this acaricidal activity. The authors also reported that sabinene and α–thujene were not toxic to the mites.

The abovementioned results prove that plant-derived extracts and single compounds may become interesting alternatives to commercial miticides. In many cases, they are obtained from plants that are nontoxic to humans, since they are part of the human diet, for example, garlic or parsley (Table 1). Therefore, they can be used in food stores, being relatively safe to humans, of course depending on the concentration used.

Table 1. Plant species reported to exhibit significant activity against stored-product pests in recent years.

Mites			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Allium sativum	essential oil essential oil	*Tyrophagus putrescentiae* *Cheyletus malaccensis*	[29,31] [30]
Azadirachta indica	Commercial product of neem (Fortune AZA)	*Tyrophagus putrescentiae* *Acarus siro* *Lepidoglyphus destructor*	[13,20]
Carum carvi	essential oil	*Gohieria fusca* *Lepidoglyphus destructor* *Acarus siro* *Tyrophagus putrescentiae*	[25]
Cinnamomum zeylanicum	essential oil	*Tyrophagus putrescentiae*	[33]
Eucaliptus globulus	essential oil	*Tyrophagus longior*	[35]
Eugenia caryophyllata	essential oil	*Tyrophagus putrescentiae*	[21]
Eugenia uniflora	essential oil	*Tyrophagus putrescentiae*	[33]
Eugenia uvalha	essential oil	*Tyrophagus putrescentiae*	[33]
Juniperus chinensis	essential oil	*Tyrophagus putrescentiae*	[41]
Lavandula angustifolia	essential oil	*Tyrophagus longior*	[35]
Lavandula stoechas	essential oil	*Tyrophagus longior*	[35]
Ligustrum japonicum	essential oil	*Tyrophagus putrescentiae*	[24]
Melaleuca leucadendra	essential oil	*Tyrophagus putrescentiae*	[33]
Mentha piperita	essential oil	*Tyrophagus longior*	[35]
Myosotis arvensis	essential oil	*Tyrophagus putrescentiae*	[28]
Ocimum basilicum	essential oil	*Tyrophagus putrescentiae*	[29]
Petroselinum sativum	active constituents	*Tyrophagus putrescentiae*	[27]
Pinus pinea	essential oil	*Tyrophagus putrescentiae*	[40]
Pinus halepensis	essential oil	*Tyrophagus putrescentiae*	[40]
Pinus pinaster	essential oil	*Tyrophagus putrescentiae*	[40]
Pinus nigra	essential oil	*Tyrophagus putrescentiae*	[40]
Piper marginatum	essential oil	*Tyrophagus putrescentiae*	[33]
Prunus persica	essential oil	*Tyrophagus putrescentiae*	[37]
Rosmarinus officinalis	essential oil	*Tyrophagus putrescentiae*	[22]
Schinus terebinthifolius	essential oil	*Tyrophagus putrescentiae*	[33]
Trigonella foenum-graecum	essential oil	*Tyrophagus putrescentiae*	[29]
Coleoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Achillea wilhelmsii	essential oil	*Tribolium castaneum*	[42]
Achyranthus aspera	essential oil	*Cryptolestes ferrugineus*	[43]
Acisanthera.	ethanolic extract	*Tenebrio molitor*	[44]
Acorus calamus	essential oil	*Sitophilus oryzae* *Tribolium castaneum*	[45]
Adenocalymma nodosum	ethanolic extract	*Tenebrio molitor*	[44]
Agastache rugosa	plant extract	*Tribolium castaneum*	[46]
Allium sativum	powder	*Oryzaephilus surinamensis*	[47]
	essential oil	*Tenebrio molitor*	[48]
	essential oil	*Sitophilus oryzae* *Tribolium castaneum*	[49]
Alpinia blepharocalyx	essential oil	*Lasioderma serricorne*	[50]
Amomum maximum	essential oil	*Tribolium castaneum*	[51]
Amomum tsaoko	essential oil	*Lasioderma serricorne* *Tribolium castaneum*	[52]

Table 1. *Cont.*

Coleoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Anethum graveolens	essential oil	*Sitophilus zeamais*	[53]
Armoracia rusticana	essential oil	*Sitophilus zeamais*	[54]
Artemisia absinthium	powder	*Oryzaephilus surinamensis*	[47]
Artemisia anethoides	essential oil	*Lasioderma serricorne*	[55]
		Tribolium castaneum	
Artemisia herba-alba	essential oil	*Oryzaephilus surinamensis*	[56]
		Tribolium castaneum	
Artemisia judaica	essential oil	*Sitophilus oryzae*	[57]
Artemisia monosperma			
Artemisia vulgaris	essential oil	*Sitophilus zeamais*	[58]
Artemisia stolonifera	essential oil	*Lasioderma serricorne*	[59]
		Tribolium castaneum	
Aster ageratoides	essential oil	*Sitophilus zeamais*	[60]
		Tribolium confusum	
Astoma seselifolium	essential oil	*Sitophilus oryzae*	[57]
Atalantia guillauminii	essential oil	*Lasioderma serricorne*	[61]
		Tribolium castaneum	
Azadirachta indica	essential oil	*Tribolium castaneum*	[62]
	seed oil		
Bauhinia purpurea	methanol extract	*Trogoderma granarium*	[63]
Bidens sulphurea	ethanolic extract	*Tenebrio molitor*	[44]
Caesalpinia gilliesii	methanol extract	*Trogoderma granarium*	[63]
Carum carvi	essential oil	*Tribolium castaneum*	[64]
		Sitophilus oryzae	
		Rhizopertha dominica	
Carum copticum	essential oil	*Sitophilus granarius*	[65]
		Tribolium confusum	
Caryopteris incana	essential oil	*Sitophilus zeamais*	[66]
Calendula officinalis	essential oil	*Sitophilus granarius*	[67]
Callistemon viminals	essential oil	*Sitophilus oryzae*	[57]
Capsicum annuum	plant extract	*Tribolium castaneum*	[46]
Cassia fistula	methanol extract	*Trogoderma granarium*	[63]
Cassia occidentalis	water extract	*Oryzaephilus surinamensis*	[68]
	ethanol extract		
	acetone extract		
Cassia senna	methanol extract	*Trogoderma granarium*	[63]
Cayratia japonica	essential oil	*Sitophilus zeamais*	[69]
		Tribolium castaneum	
Chenopodium album	ether extract	*Oryzaephilus surinamensis*	[70]
Chrysanthemum frutescens	methanol extract	*Trogoderma granarium*	[63]

Table 1. *Cont.*

Coleoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Cinnamomum verum	essential oil	*Tenebrio molitor*	[71]
		Tribolium castaneum	[64]
		Sitophilus oryzae	
		Rhizopertha dominica	
Citrus aurantifolia	essential oil	*Sitophilus oryzae*	[57]
Citrus lemon	essential oil	*Sitophilus oryzae*	[57]
Citrus medica	essential oil	*Tribolium castaneum*	[72]
Citrus paradisi	essential oil	*Rhizopertha dominica*	[73]
Citrus reticulata	essential oil	*Tribolium confusum*	[74]
	powder	*Tribolium castaneum*	[46,75]
	ethanol extract		
	essential oil		[76]
	essential oil	*Cryptolestes ferrugineus*	[77]
	essential oil	*Rhyzopertha dominica*	[73]
Citrus sinensis	essential oil	*Tribolium castaneum*	[76]
	essential oil	*Rhizopertha dominica*	[78]
	essential oil	*Sitophilus oryzae*	[57]
	essential oil	*Sitophilus zeamais*	[79]
Calamintha glandulosa	essential oil	*Tribolium castaneum*	[80]
Clausena anisum-olens	essential oil	*Lasioderma serricorne*	[81]
Cleome viscosa	water extract	*Oryzaephilus surinamensis*	[68]
	ethanol extract		
	acetone extract		
Coriandrum sativum	essential oil	*Sitophilus oryzae*	[82]
Crithmum maritimum	essential oil	*Oryzaephilus surinamensis*	[83]
		Sitophilus granarius	
		Sitophilus oryzae	
		Tribolium castaneum	
		Tribolium confusum	
Cuminum cyminum	essential oil	*Sitophilus zeamais*	[53]
Cupressus lusitanica	essential oil	*Sitophilus zeamais*	[84]
		Tribolium castaneum	
Cupressus macrocarpa	essential oil	*Sitophilus oryzae*	[57]
Cupressus sempervirens	essential oil	*Sitophilus oryzae*	[57]
Cymbopogon citratus	essential oil	*Sitophilus oryzae*	[85]
	essential oil	*Tribolium castaneum*	[86]
Cymbopogon giganteus	essential oil	*Tribolium castaneum*	[86]
Cymbopogon schoenanthus	essential oil	*Tribolium castaneum*	[86]
Cymbopogon winterianus	essential oil	*Tribolium castaneum*	[64]
		Sitophilus oryzae	
		Rhizopertha dominica	

Table 1. *Cont.*

Coleoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Dahlia pinnata	essential oil	*Sitophilus oryzae*	[50]
		Sitophilus zeamais	
Dennettia tripetala	essential oil	*Sitophilus oryzae*	[87]
Dimorphandra mollis	ethanolic extract	*Tenebrio molitor*	[44]
Dracocephalum moldavica	essential oil	*Sitophilus zeamais*	[88]
		Tribolium confusum	
Drimys winteri	essential oil	*Tribolium castaneum*	[89]
Dictamnus dasycarpus	essential oil	*Lasioderma serricorne*	[90]
Eruca sativa	essential oil	*Tribolium confusum*	[74]
Etlingera yunnanensis	essential oil	*Tribolium castaneum*	[91]
Eucalyptus camaldulensis	essential oil	*Sitophilus oryzae*	[92,93]
		Tribolium castaneum	
Eucalyptus citriodora	essential oil	*Tribolium castaneum*	[86]
Eucalyptus floribundi	essential oil	*Oryzaephilus surinamensis*	[94]
		Rhizopertha dominica	
Eucalyptus globulus	essential oil	*Tribolium confusum*	[74]
	essential oil	*Lasioderma serricorne*	[95]
		Rhizopertha dominica	
Eucalyptus intertexta	essential oil	*Sitophilus oryzae*	[93]
		Tribolium castaneum	
Eucalyptus leucoxylon	essential oil	*Sitophilus oryzae*	[96]
		Tribolium castaneum	
Eucalyptus obliqua	essential oil	*Sitophilus oryzae*	[82]
Eucalyptus procera	essential oil	*Tribolium castaneum*	[97]
Eucalyptus saligna	essential oil	*Sitophilus zeamais*	[84]
		Tribolium castaneum	
Eucalyptus sargentii	essential oil	*Sitophilus oryzae*	[93]
		Tribolium castaneum	
Euonymus japonicus	methanol extract	*Trogoderma granarium*	[63]
Ferula narthex	essential oil	*Cryptolestes ferrugineus*	[43]
Foeniculum vulgare	essential oil	*Sitophilus zeamais*	[53]
	essential oil	*Tribolium castaneum*	[64]
		Sitophilus oryzae	
		Rhizopertha dominica	
Ginkgo biloba	plant extract	*Tribolium castaneum*	[46]
Hyptis suaveolens	essential oil	*Rhizopertha dominica*	[98]
		Sitophilus oryzae	
		Tribolium castaneum	
Juniperus formosana	essential oil	*Tribolium castaneum*	[99]

Table 1. *Cont.*

Coleoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Juniperus polycarpus	essential oil	*Tribolium confusum*	[100]
Juniperus sabina	essential oil	*Tribolium confusum*	
Kadsura heteroclita	essential oil	*Sitophilus zeamais*	[101]
Laurelia sempervirens	essential oil	*Sitophilus zeamais*	[102]
	essential oil	*Tribolium castaneum*	[89]
Laurus nobilis	essential oil	*Lasioderma serricorne*	[103]
		Rhizopertha dominica	[104]
		Tribolium castaneum	
Laggera pterodonta	essential oil	*Lasioderma serricorne*	[105]
Lavandula angustifolia	essential oil	*Sitophilus granarius*	[106]
Lavandula officinalis	essential oil	*Sitophilus oryzae*	[92]
		Tribolium castaneum	
Lavandula stoechas	essential oil	*Lasioderma serricorne*	[95]
		Rhizopertha dominica	
		Tribolium castaneum	
Lepidoploa aurea	ethanolic extract	*Tenebrio molitor*	[44]
Litsea cubeba	essential oil	*Lasioderma serricorne*	[107]
Litsea salicifolia	essential oil	*Sitophilus zeamais*	[108]
		Tribolium castaneum	
Linium usitatissium	essential oil	*Cryptolestes ferrugineus*	[43]
Lippia javanica	essential oil	*Sitophilus zeamais*	[109]
Lippia sidoides	essential oil	*Tenebrio molitor*	[110]
Liriope muscari	essential oil	*Lasioderma serricorne*	[111]
		Tribolium castaneum	
Maytenus emarginata	ether extract	*Oryzaephilus surinamensis*	[70]
Melia azedarach	essential oil	*Cryptolestes ferrugineus*	[43]
Mentha piperita	powder	*Oryzaephilus surinamensis*	[47]
	essential oil	*Tribolium castaneum*	[112]
		Lasioderma serricorne	
	essential oil	*Sitophilus oryzae*	[113]
Mentha longifolia	essential oil	*Tribolium castaneum*	[42]
	essential oil	*Sitophilus zeamais*	[114]
Mentha pulegium	essential oil	*Sitophilus granarius*	[115]
	essential oil	*Tribolium castaneum*	[116]
		Lasioderma serricorne	
Mentha .	essential oil	*Sitophilus oryzae*	[85]
Mesua ferrea	water extract	*Oryzaephilus surinamensis*	[68]
	ethanol extract		
	acetone extract		
Micromeria fruticosa	essential oil	*Sitophilus granarius*	[117]
Minthostachys verticillata	essential oil	*Sitophilus zeamais*	[118]

Table 1. *Cont.*

Plant Species	Formulation	Coleoptera Pest Species	Reference
Mosla soochowensis	essential oil	*Sitophilus zeamais*	[119]
		Tribolium confusum	
Myristica fragrans	essential oil	*Tribolium castaneum*	[64]
		Sitophilus oryzae	
		Rhizopertha dominica	
Myrtus communis	essential oil	*Sitophilus oryzae*	[57]
Nardostachys chinensis	essential oil	*Tribolium castaneum Lasioderma serricorne*	[120]
	supercritical CO_2 fluid extract		
Nigella sativa	essential oil	*Tribolium castaneum*	[64]
		Sitophilus oryzae	
		Rhizopertha dominica	
Ocimum basilicum	essential oil	*Sitophilus oryzae*	[121]
	essential oil	*Sitophilus zeamais*	[79]
		Tribolium castaneum	[122]
		Tribolium confusum	
		Trogoderma granarium	
Ocimum gratissimum	essential oil	*Oryzaephilus surinamensis*	[123]
		Rhizopertha dominica	
		Sitophilus oryzae	
		Tribolium castaneum	
Origanum acutidens	essential oil	*Lasioderma serricorne*	[124]
		Sitophilus granarius	
Origanum majorana	essential oil	*Tribolium confusum*	[125]
Origanum minutiflorum	essential oil	*Tribolium confusum*	
Origanum onites	essential oil	*Tribolium confusum*	
Origanum syriacum	essential oil	*Tribolium confusum*	
Origanum vulgare	essential oil	*Sitophilus oryzae*	[57]
	essential oil	*Tribolium castaneum*	[46,125]
Ostericum viridiflorum	essential oil	*Tribolium castaneum*	[126]
Petroselinum crispum	essential oil	*Sitophilus zeamais*	[53]
Perilla frutescens	essential oil	*Lasioderma serricorne*	[127]
Pimenta dioica	powder	*Oryzaephilus surinamensis*	[47]
Pimpinella anisum	essential oil	*Tribolium castaneum*	[46]
Pinus longifolia	essential oil	*Sitophilus oryzae*	[82]
Pituranthos tortuosus	essential oil	*Sitophilus oryzae*	[57]
Platycladus orientalis	essential oil	*Sitophilus oryzae*	[128]
		Tribolium castaneum	
Pongamia pinnata	water extract	*Oryzaephilus surinamensis*	[68]
	ethanol extract		
	acetone extract		

Table 1. *Cont.*

		Coleoptera	
Plant Species	**Formulation**	**Pest Species**	**Reference**
Psidium guajava	powder	*Tribolium castaneum*	[75]
	ethanol extract		
Pulicaria gnaphalodes	essential oil	*Tribolium castaneum*	[42]
Punica gronatum	ether extract	*Oryzaephilus surinamensis*	[70]
Ricinus communis	essential oil	*Tribolium castaneum*	[116]
		Lasioderma serricorne	
	essential oil	*Tribolium confusum*	[74]
Rosmarinus officinalis	essential oil	*Tribolium confusum*	[74]
Salvia officinalis	powder	*Oryzaephilus surinamensis*	[47]
Salvertia convallariaeodora	ether extract	*Oryzaephilus surinamensis*	[70]
Sasurrea costus	essential oil	*Cryptolestes ferrugineus*	[43]
Satureja hortensis	essential oil	*Tribolium castaneum*	[129]
Schinus molle	essential oil	*Sitophilus oryzae*	[57]
Schinus terebinthifolius			
Solanum nigrum	glycoalkaloid extract	*Tenebrio molitor*	[130]
Syzygium aromaticum	essential oil	*Tenebrio molitor*	[71]
		Sitophilus oryzae	[92]
		Tribolium castaneum	
Syzygium cumini	essential oil	*Tribolium confusum*	[74]
	essential oil	*Sitophilus oryzae*	[57]
Tanacetum cinerariifolium	essential oil	*Tribolium castaneum*	[46]
Tagetes erecta	essential oil	*Sitophilus oryzae*	[131]
		Tribolium castaneum	
Tagetes minuta	essential oil	*Sitophilus oryzae*	[131]
		Tribolium castaneum	
Tagetes patula	essential oil	*Sitophilus oryzae*	[131]
		Tribolium castaneum	
Teucrium polium	essential oil	*Tribolium castaneum*	[132]
Thespesia populnea var. acutiloba	methanol extract	*Trogoderma granarium*	[63]
Thuja occidentalis	essential oil	*Sitophilus oryzae*	[57]
Trewia nudiflora	water extract	*Oryzaephilus surinamensis*	[68]
	ethanol extract		
	acetone extract		
Typhonium trilobatum	water extract	*Oryzaephilus surinamensis*	[68]
	ethanol extract		
	acetone extract		
Valeriana jatamansi	essential oil	*Tribolium castaneum*	[120]
	supercritical CO_2 fluid extract	*Lasioderma serricorne*	
Valeriana officinalis	essential oil	*Tribolium castaneum Lasioderma serricorne*	[120]
	supercritical CO_2 fluid extract		
Vepris heterophylla	essential oil	*Sitophilus oryzae*	[133]

Table 1. *Cont.*

Plant Species	Formulation	Pest Species	Reference
		Coleoptera	
Verbascum cheiranthifolium	ethanol extract	*Sitophilus oryzae*	[134]
Verbascum speciosum	ethanol extract	*Sitophilus oryzae*	[134]
Viola odorata	essential oil	*Cryptolestes ferrugineus*	[43]
Vitex negundo	ether extract	*Oryzaephilus surinamensis*	[70]
Xylopia aethiopica	essential oil	*Sitophilus oryzae*	[87]
Zanthoxylum armatum	essential oil	*Lasioderma serricorne*	[135]
		Tribolium castaneum	
Zanthoxylum dissitum	essential oil	*Lasioderma serricorne*	[135]
		Tribolium castaneum	
Zanthoxylum planispinum var. dintanensis	essential oil	*Tribolium castaneum Lasioderma serricorne*	[136]
Zingiber officinale	essential oil	*Sitophilus oryzae*	[85]
Zingiber purpureum	essential oil	*Lasioderma serricorne*	[137]
	essential oil	*Tribolium castaneum*	
		Lepidoptera	
Plant Species	**Formulation**	**Pest Species**	**Reference**
Acacia nilotica	acetone and pet ether extracts	*Corcyra cephalonica*	[138]
Acorus calamus	petroleum ether extract acetone extract	*Sitotroga cerealella*	[139]
Aframomum melegueta	powder extract	*Sitotroga cerealella*	[140]
Agastache rugosa	plant extract	*Plodia interpunctella*	[46]
Ajuga iva	methanol extract	*Plodia interpunctella*	[141]
Allium cepa	essential oil	*Plodia interpunctella*	[142]
	essential oil	*Sitotroga cerealella*	[143]
Allium sativum	major component: diallyl disulfide and diallyl trisulfide	*Sitotroga cerealella*	[143]
	major component: diallyl trisulfide	*Sitotroga cerealella*	[144–146]
	essential oil	*Ephestia kuehniella*	[147–149]
	essential oil	*Ephestia cautella*	[149]
	essential oil	*Plodia interpunctella*	[147]
Anacardium occidentale	essential oil	*Ephestia cautella*	[150]
	ethanolic oil extract and petroleum ether extract	*Ephestia cautella*	[151]
Anethum graveolens	essential oil	*Plodia interpunctella*	[152]
	oils applied as a 30% aqueous solution	*Ephestia kuehniella*	[153]
Arachis hypogaea	acetone extract	*Corcyra cephalonica*	[154]
Armoracia rusticana	essential oil	*Plodia interpunctella*	[54]
Artemisia haussknechtii	essential oil	*Ephestia kuehniella*	[155]
Artemisia herba alba	essential oil	*Ephestia kuehniella*	[156]
Artemisia khorassanica	essential oil	*Sitotroga cerealella*	[157]
	essential oil	*Plodia interpunctella*	[158]
Artemisia sieberi Bess	essential oil	*Sitotroga cerealella*	[157]
Artemisia vulgaris	essential oil	*Cadra cautella*	[159]
	essential oil	*Sitotroga cerealella*	[58]
Azadirachta indica	essential oil	*Ephestia cautella*	[160]
Azadirachta indica + Bacilus thuringiensis		*Ephestia cautella*	[160]

Table 1. *Cont.*

Lepidoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Betula lenta	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[147]
Brassica alba	essential oil	*Ephestia cautella*	[148]
Callistemon viminalis	essential oil	*Ephestia kuehniella*	[161]
Calotripis procera roots	extract	*Cadra cautella*	[162]
Capsicum annuum	plant extract	*Plodia interpunctella*	[46]
Capsicum frutescens	essential oil	*Ephestia cautella*	[150]
	ethanolic oil extract and petroleum ether extract	*Ephestia cautella*	[151]
Carum copticum	essential oil	*Ephestia kuehniella* *Plodia interpunctella*	[163]
Cassia angustifolia	protease inhibitor	*Plodia interpunctella*	[164]
Centaurium erythraea	methanol extract	*Plodia interpunctella*	[141]
Cinnamomum camphora	essential oil	*Ephestia kuehniella*	[165]
Cinnamomum.	oil encapsulated with different types of alcohol	*Plodia interpunctella*	[166]
	polyethylene-laminated polypropylene films	*Plodia interpunctella*	[167]
Cinnamomum zeylanicum	essential oil	*Plodia interpunctella*	[168,169]
	essential oil	*Plodia interpunctella*	[147]
	essential oil	*Ephestia kuehniella*	
Cistus ladanifer	methanol extract	*Plodia interpunctella*	[141]
Cistus monspeliensis	methanol extract	*Plodia interpunctella*	
Cistus salviaefolius	methanol extract	*Plodia interpunctella*	
Citrus aurantium	essential oil	*Sitotroga cerealella*	[170]
Citrus bergamia	essential oil	*Plodia interpunctella*	[171]
Citrus limon	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[172]
	essential oil	*Ephestia kuehniella*	[173]
Citrus reticulata	plant extract	*Plodia interpunctella*	[46]
Citrus vulgaris	essential oil	*Ephestia kuehniella*	[155]
Clausena anisata	essential oil	*Sitotroga cerealella*	[174]
Cleisthopholis patens	root bark powder	*Plodia interpunctella*	[175]
	stem bark powder	*Plodia interpunctella*	
	leaf powder	*Plodia interpunctella*	
Conocarpus lancifolius (leaves)	aqueous and ethanolic extract	*Ephestia cautella*	[176]
Coriandrum sativum	essential oil	*Sitotroga cerealella*	[177]
	essential oil	*Ephestia kuehniella*	[178,179]
	essential oil	*Plodia interpunctella*	[178]
	essential oil	*Corcyra cephalonica*	[82]
Cuminum cyminum	essential oil	*Ephestia kuehniella*	[180]
Cupressus lusitanica	leaf essential oils	*Sitotroga cerealella*	[181]
Cupressus sempervirens	essential oil	*Cadra cautella*	[159]
Cupressus sempervirens L. horizontalis	resin essential oil	*Ephestia kuehniella*	[182]
Cymbopogon martinii	essential oil	*Plodia interpunctella*	[171]
Cyrtocymura cincta	sesquiterpene lactones isolated from leaves and flowers	*Sitotroga cerealella*	[183]
Dalbergia sissoo	acetone extract	*Corcyra cephalonica*	[138]
Dimorphandra mollis	flowers	*Sitotroga cerealella*	[184]
Dryopteris filix (mas root and rhizome)	ethanolic extract	*Corcyra cephalonica*	[185]
Elaeagnus angustifolia	ethyl acetate aqueous	*Ephestia cautella* *Ephestia cautella*	[186]
Elettaria cardamomum	essential oil	*Ephestia kuehniella*	[187,188]

Table 1. *Cont.*

Lepidoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Eucalyptus astringens	essential oil	*Ephestia cautella* *Ephestia kuehniella*	[189]
Eucalyptus camaldulensis	essential oil	*Ephestia kuehniella*	[180]
	essential oil	*Ephestia cautella* *Ephestia kuehniella* *Cadra cautella*	[189]
Eucalyptus dives	oil and constituents	*Plodia interpunctella*	[190]
Eucalyptus globulus	essential oil	*Ephestia cautella* *Ephestia kuehniella*	[149]
	essential oil	*Plodia interpunctella*	[171]
Eucalyptus lehmannii	essential oil	*Ephestia cautella* *Ephestia kuehniella*	[189]
Eucalyptus leucoxylon	essential oil	*Ephestia cautella* *Ephestia kuehniella*	[189]
Eucalyptus obliqua	essential oil	*Corcyra cephalonica*	[82]
Eucalyptus platyphylla	essential oil	*Ephestia cautella*	[191]
Eucalyptus rudis	essential oil	*Ephestia cautella* *Ephestia kuehniella*	[189]
Eucalyptus saligna	leaf essential oils	*Sitotroga cerealella*	[181]
Ferula galbaniflua	essential oil	*Cadra cautella*	[159]
Ferula gummosa	essential oil	*Ephestia kuehniella*	[161,188]
Geranium maculatum	essential oil	*Plodia interpunctella*	[171,192]
Ginkgo biloba	plant extract	*Plodia interpunctella*	[46]
Glossocardia bosvallia	hexane extracts	*Corcyra cephalonica*	[193]
Hyossopus officinalis	essential oil	*Plodia interpunctella*	[152]
Hypericum scabrum	essential oil	*Ephestia kuehniella*	[117]
Hyssoppus officinalis	essential oil	*Ephestia kuehniella*	[117]
Lantana camara	essential oil	*Cadra cautella*	[194]
Launaea arborescens	methanol extract	*Plodia interpunctella*	[141]
Laurus nobilis	essential oil	*Ephestia kuehniella*	[195]
	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[172]
Lavandula angustifolia	essential oil	*Ephestia kuehniella*	[196]
Lavandula angustifolia	extract	*Ephestia kuehniella*	[197]
Lavandula angustifolia	essential oil	*Plodia interpunctella*	[171]
Lavandula dentata	methanol extract	*Plodia interpunctella*	[141]
Lawsonia inermis	pet ether extract	*Corcyra cephalonica*	[138]
Lippia turbinata	essential oil	*Plodia interpunctella*	[198]
Maxillaria tenuifolia	ethanolic oil extract and petroleum ether extract	*Ephestia cautella*	[151]
Melaleuca viridiflora	essential oil	*Cadra cautella*	[159]
Mentha longifolia	essential oil	*Ephestia kuehniella*	[199]
Mentha piperita	essential oil	*Corcyra cephalonica*	[113]
	polylactic acid solution	*Plodia interpunctella*	[200]
	essential oil	*Plodia interpunctella*	[171,192]
	essential oil	*Ephestia cautella*	[191]
Mentha pulegium	essential oil	*Ephestia kuehniella*	[201]
Mentha spicata	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[202]
Micromere fruticosa	essential oil	*Ephestia kuehniella*	[117,203]
Monodora tenuifolia	essential oil	*Ephestia cautella*	[150]
Myristica fragrans	essential oil	*Cadra cautella*	[159]
	essential oil	*Plodia interpunctella*	[204]
Myrtus commonis	essential oil	*Ephestia kuehniella*	[195,205]
	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[172]
Nepata racemosa	essential oil	*Ephestia kuehniella*	[203]
Neroli birgard	essential oil	*Sitotroga cerealella*	[170]

Table 1. *Cont.*

Lepidoptera			
Plant Species	**Formulation**	**Pest Species**	**Reference**
Ocimum basilicum	oils applied as a 30% aqueous solution	*Ephestia kuehniella*	[153]
	essential oil	*Ephestia kuehniella*	[178,201,202]
	essential oil	*Plodia interpunctella*	[178,202]
Ocimum kilimandscharicum	ground leaves	*Sitotroga cerealella*	[206]
	essential oil	*Sitotroga cerealella*	
Ocimum suave	essential oil	*Sitotroga cerealella*	[207]
	dry or ground leaves	*Sitotroga cerealella*	
Origanum acutidens	essential oil	*Ephestia kuehniella*	[117,124]
Origanum majorana	essential oil	*Corcyra cephalonica*	[208]
	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[172]
	essential oil	*Ephestia kuehniella*	[173]
Origanum onites	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[205]
Origanum syriacum var. bevanii	essential oil	*Ephestia kuehniella*	[180]
Origanum vulgare	essential oil	*Plodia interpunctella*	[46]
	essential oil	*Ephestia kuehniella*	[156,203]
Origanum vulgaris	essential oil	*Ephestia kuehniella*	[209]
Parthenium hysterophorus	acetone extract	*Corcyra cephalonica*	[138]
Peganum harmala	methanol extract	*Plodia interpunctella*	[141]
Petroselinum crispum	essential oil	*Ephestia kuehniella*	[179]
Petroselinum sativum	essential oil	*Plodia interpunctella*	[152]
Pimpinella anisum	essential oil	*Ephestia cautella*	[149]
	essential oil	*Ephestia kuehniella*	[147,149,180]
	essential oil	*Plodia interpunctella*	[46,147]
Pinus brutia	essential oil	*Ephestia kuehniella*	[210]
Pinus densiflora	diterpene resin acids (DRAs)	*Plodia interpunctella*	[211]
Pinus longifolia	essential oil	*Corcyra cephalonica*	[82]
Pinus pinea	essential oil	*Ephestia kuehniella*	[210]
Piper nigrum	essential oil	*Corcyra cephalonica*	[113]
Piper sarmentosum	hexane extracts	*Plodia interpunctella*	[212]
Pistacia lentiscus	essential oil	*Ephestia kuehniella*	[213]
Polyalthia longifolia	essential oil	*Sitotroga cerealella*	[174]
Portulaca oleracea	essential oil	*Ephestia cautella*	[214]
	essential oil	*Ephestia kuehniella*	[215]
Prunus	grain coating with amygdalin	*Plodia interpunctella*	[216]
Psychotria prunifolia	leaves	*Sitotroga cerealella*	[184]
Ricinus communis	essential oil	*Ephestia cautella*	[150,214]
	ethanolic oil extract and petroleum ether extract	*Ephestia cautella*	[151]
Rosemarinus officinalis	essential oil	*Ephestia kuehniella*	[149,180,196]
	essential oil	*Ephestia cautella*	[149]
	essential oil	*Cadra cautella*	[159]
	essential oil	*Plodia interpunctella*	[152]
	methanol extract	*Plodia interpunctella*	[141]
Ruta graveolens	essential oil	*Ephestia kuehniella*	[201]
Ruta montana	essential oil	*Ephestia kuehniella*	[156]
Salvia limbata	essential oil	*Ephestia kuehniella*	[117]
Salvia nemorosa	essential oil	*Ephestia kuehniella*	[117]
Salvia officinalis	essential oil	*Ephestia kuehniella*	[188]
	polylactic acid solution	*Plodia interpunctella*	[200]
Satureja hortensis	essential oil	*Plodia interpunctella*	[129]
		Ephestia kuehniella	[129]
	essential oil	*Cadra cautella*	[159]
Satureja thymbra	essential oil	*Plodia interpunctella*	[205]
		Ephestia kuehniella	[172,205]
Satureje hortensis	essential oil	*Ephestia kuehniella*	[117]
Syzygium aromaticum	essential oil	*Ephestia kuehniella*	[165]

Table 1. *Cont.*

Plant Species	Formulation	Pest Species	Reference
		Lepidoptera	
Tanacetum cinerariifolium	essential oil	*Plodia interpunctella*	[46]
Thymus daenensis	essential oil	*Plodia interpunctella* *Ephestia kuehniella*	[217]
Thymus vulgaris	essential oil	*Plodia interpunctella*	[152]
	essential oil	*Ephestia kuehniella*	[117]
Tithonia diversifolia	leaves	*Sitotroga cerealella*	[184]
Trachyspermum ammi	essential oil	*Plodia interpunctella*	[204]
Trigonella foenum-graecum	protease inhibitor	*Plodia interpunctella*	[164]
Vitex pseudo-negundo	essential oil	*Plodia interpunctella*	[158]
Xylopia aethiopica	essential oil	*Ephestia cautella*	[150]
	ethanolic oil extract and petroleum ether extract	*Ephestia cautella*	[151]
Zingiber officinale	essential oil	*Ephestia kuehniella*	[153]
	essential oil	*Ephestia kuehniella*	[218]
	essential oil	*Plodia interpunctella*	[218]
Ziziphora clinopodioides Lam.	essential oil	*Ephestia kuehniella*	[219]

2.2. Insects (Insecta)

Pests affecting grains can be found in various orders of insects. Some of these belong to the major pests affecting stored crops and food, whereas some are of minor importance. Among the most important ones are beetles (Coleoptera), true bugs (Hemiptera), butterflies and moths (Lepidoptera). Therefore, we will focus on these three orders.

2.2.1. Beetles (Coleoptera)

Coleopterans form the largest order within the animal kingdom, with more than 400,000 species and about 200 families. It is not surprising that within this order we may find pests affecting grains, which are of economic importance. One of the largest families of Coleoptera is Tenebrionidae, or as they are commonly known, darkling beetles. Within this family there are some very important grain pests. Firstly, there is the confused flour beetle *Tribolium confusum* Jacq. Du Val, a pest that feeds on all kinds of grains and flour. *T. confusum* can have as many as five generations per year and every female can lay as many as 600 eggs on the stored product. In the case of *T. confusum*, very few botanical insecticides have been tested. Firstly, *Carum copticum* and *Cuminum cyminum* were effective in their contact toxicity effects against *T. confusum* with LD_{50} values of 0.037 µg/mg and 0.039 µg/mg, respectively [65]. Secondly, *Aster ageratoides* [60], *Juniperus polycarpos* and *Juniperus sabina* [100] acted as fumigants in the form of EO, whereas *Crithmum maritimum* did not cause any mortality [83].

Tribolium castaneum Herbst, commonly known as the rust-red flour beetle, belongs to the same genus as *T. confusum*, with which it has physical similarities, life cycle and feeding habits, meaning it is also a secondary pest. *T. castaneum*, as a model beetle species, has been the most tested beetle in regard to the effects of botanical insecticides. The tested natural substances have been mainly evaluated for their fumigant toxicity, contact toxicity or repellant properties. *Tagetes minuta* [131], *Tagetes patula* [131], *Platycladus orientalis* (fruits) [128], *A. sativum* [49] and *M. piperita* [112] have been proven to be the most effective fumigants against the adults of this species. *Eucalyptus procera* [97], basil [79], orange [79] and *Satureja hortensis* oils were toxic after contact with *T. confusum* adults [129], whereas *Citrus reticulata* and *Citrus sinensis* essential oils were effective against larvae [76]. Finally, many plant extracts were proven to be effective as repellents; the repellency abilities of the botanical substances were assessed based on the area test, as described by McDonald et al. (1970) [220]—namely, *Litsea salicifolia* [108], *Artemisia anethoides* [55], *Zanthoxylum planispinum* [136] and *Hyptis suaveolens* exhibited repellency rates of more than 90% [98]. *C. reticulata* was also effective as a repellent in the form of powder of ethanol extract.

The yellow mealworm beetle, *Tenebrio molitor* L., is also a secondary pest, feeding on flour, grains and plant-based products. Garlic essential oil caused necrosis in larvae, pupae and adults of *T. molitor* L. after 20–40 h of exposure [48], whereas *Bidens sulphurea* and *Adenocalymma nodosum* were more effective against the pupae [44]. Cinnamon oil was most toxic to *T. molitor* L. larvae, whereas clove oil was most effective on adults. Cinnamon oil mainly yielded eugenol (10.19%), trans-3-caren-2-ol (9.92%) and benzyl benzoate (9.68%); whereas clove oil yielded eugenol (26.64%), caryophyllene (23.73%) and caryophyllene oxide (17.74%) [71]. Interestingly, some research on *T. molitor* L. has focused on the sublethal effects of glycoalkaloids, describing malformations of organelles, chromatin condensation and altered contractility of the heart and oviduct [130].

Another large family of Coleoptera is the Curculionidae, or as they are commonly known, "snout beetles". In this family there are 83,000 species described worldwide. There are some important grain pests in this family. *Sitophilus granarius* L. is a frequent pest of wheat and barley, but also can attack other cereals, such as maize, sorghum and rice. Lower developmental stages (eggs, larvae and pupae) of *S. granarius* complete their development inside a seed kernel or products, which is common for all other primary insects. Every female of the species can produce as many as 400 eggs, which are placed separately inside the seeds. It is a primary pest which feeds on all Poaceae grains. Many plant extracts have been tested for acute toxicity against *S. granarius* with *Calendula officinalis* being amongst the most promising contact toxicant extracts [67], whereas *Origanum acutidens* [124], *C. maritimum* [83] and *L. angustifolia* were the best fumigants [106]. Furthermore, *L. angustifolia* [106], *C. copticum* and *Cuminum cyminum* were very powerful in terms of their contact toxicity [65]; *L. angustifolia* was also effective as a repellent [106].

The rice weevil (*Sitophilus oryzae* L.) is morphologically very similar to *S. granarius*, but it is more commonly found in stored rice. It feeds mainly on cereal seeds and is less likely to feed on grain products. It is also more resistant to lower temperatures than *S. granarius*. Among the natural substances trialed for their fumigant properties, the most effective were the essential oils obtained from *Cymbopogon citratus* and *Zingiber officinale* [85], *O. basilicum* [121], *Origanum vulgare* and *Citrus* lemon oils [57]. Essentials oils of fennel (*Foeniculum vulgare*), caraway (*C. carvi*), cinnamon (*Cinnamomum verum*), citronella (*Cymbopogon winterianus*), nutmeg (*Myristica fragrans*) and black cumin (*Nigella sativa*) were all proven to be highly competent as contact toxicity insecticides [64]. Additionally, *Artemisia judaica, Callistemon viminals* and *O. vulgare* caused high-contact toxicity to *S. oryzae* with LC_{50} values of 0.08, 0.09 and 0.11 mg/cm^2, respectively [57]. *Ocimum gratissimum* (EO) and its constituents [123], as well as *H. suaveolens* (EO), seemed to be very effective as repellents [98]. Finally, some botanical extracts (*Acorus calamus* + *Corchorus capsularis* seed petroleum ether extract, *A. calamus* + *Thevetia neriifolia* seed petroleum ether extract and *A. calamus* + *Zingiber cassumunar Roxb.* rhizome petroleum ether extract) were assessed in terms of the synergism of their toxic effects [45] or repellent effects (*Hyptis spicigera*, *Vepris heterophylla*) [133].

The maize weevil (*Sitophilus zeamais* Motsch) is another important pest, of an appearance similar the previously mentioned beetles; it is mainly found in stored maize but can sometimes be found in wheat and barley. *S. zeamais* Motsch has the ability to fly; hence, the infestation can start in the ripening crop in the field and can carry on in storage. *Chenopodium ambrosioides* [221] has been a very effective toxic fumigant, as has *Blumea balsamifera* [222], as well as orange and basil oil [79]. *Kadsura heteroclite* stems (EO) were the most effective in terms of contact toxicity, followed by *Cayratia japonica* (EO) [101]. Furthermore, *L. salicifolia* [108] and *Anethum graveolens* (EO), *Petroselinum crispum* (EO), *F. vulgare* (EO) and *C. cyminum* (EO) were effective repellents against the maize weevil [53].

There are also some important pests of grains within other families. For example, *Lasioderma serricorne* (Fabricius) (Anobiidae), commonly known as the "cigarette beetle", is a secondary pest, which feeds on a great variety of products, including grain-based products, herbs, and legumes. It can have up to four generations per year and it overwinters as larvae. Extracts of the botanical species *O. acutidens* seemed to be the most effective in terms of fumigant activities against this pest [124], followed by *Artemisia mongolica* [81], *Alpinia blepharocalyx* [50] and *Perilla frutescens* [127]. *P. frutescens* (L.) [127], *Z. planispinum* [127] and *Artemisia stolonifera* [59] extracts and ingredient components were also very promising as alternative and greener fumigants. Finally, *M. piperita* menthol extract showed very strong repellent properties, with 80% repellency after 4 h at the test concentration of 3.15 μL/cm^2 [112], which was much greater than *Z. planispinum* essential oil, exhibiting 62% repellency after 4 h of exposure at 78.63 nL/cm^2 [136], and *Laurus nobilis*, exhibiting 60% repellency at a concentration of 0.2 μL/cm^2 after 24 h of exposure [103]. *Cryptolestes ferrugineus* (Stephens) and other Cryptolestes species (Cucujidae) are important secondary pests of food grains [223]. EO from *C. reticulata* peel achieved 100% fumigant mortality, contact toxicity that was fully effective (99.0% after 48 h of exposure at the dosage of 2.0 μL/cm^2) and finally had a very good repellency rate (achieved >80.0% after 60 h of exposure at 0.4 μL/cm^2) [77]. Unfortunately, according to other studies carried out with extracts of *Melia azedarach, Linium usitatissium, Ferula narthex, Sasurrea costus, Viola odorata* and *Achyranthus aspera*, they were not very effective, although M. azedarach was able to decrease larval and pupal emergence [122].

The saw-toothed grain beetle (*Oryzaephilus surinamensis* L. (Silvanidae)) feeds on grains, as well as bran and grain products. It is a secondary pest that lays eggs in flour and causes further qualitative and quantitative losses. Regarding the fumigant activity of *O. gratissimum* essential oil [123], which showed the best results at a concentration of 1 mL/L air, the oil caused a mortality rate of 99% 24 h after treatment, respectively, followed by *C. maritimum* [83], *Eucalyptus globules* and *L. stoechas* [95]. The essential oils of *H. suaveolens* leaves [98], *C. maritimum* [83] and *Artemisia herba-alba* and *Artemisia absinthium* caused necrosis to adults [56]. Powdered *Salvia officinalis* also caused a high mortality rate, reaching 80% (the powdered plant was added to culture containers at concentrations of 5.88%) [47]. Finally, *Pongamia pinnata* has the potential for use as a mediocre repellent [68]. *Rhyzopertha dominica* Fab. can be considered the main insect pest species in grains stored around the world [224,225]. It is a primary pest of stored products, feeding on all cereals and grain products. According to a study published in 2017 by Tawfeek et al., essential oils of fennel (*F. vulgare*), caraway (*C. carvi*), cinnamon (*C. verum*), citronella (*C. winterianus*), nutmeg (*M. fragrans*) and black cumin (*N. sativa*) seemed to be very active, with fennel being the most effective [64]. The essential oil of *H. suaveolens* is reported to have potential as a contact insecticide [98]. Lastly, *L. nobilis* and *Eucalyptus floribundi* are two essential oils with repellent and fumigant properties [94,189].

Khapra beetle (*Trogoderma granarium* Everts.) is the only genus from the family Dermestidae that feeds on plant-based products, especially stored grains. For many countries it is considered a quarantine concern because its spread occurs mainly through international trade. According to Derbalah and his team, in a research study in 2011, seven plant methanol extracts were tested for their insecticidal properties and the results were positive, with *Cassia senna* being the most effective (100 mg/L achieved 86.7% mortality after one week) [63]. Interestingly, the plant extracts of *Bauhinia purpurea, C. senna, Caesalpinia gilliesii, Cassia fistula, Chrysanthemum frutescens, Euonymus japonicus* and *Thespesia populnea var. acutiloba* were also effective towards the beetle and displayed no side effects to rats, indicating friendliness to mammals. Finally, the essential oil of *O. basilicum* was found to cause mortality at 54.33% after 72 h [122].

As one can see, there are not enough data concerning the sublethal action of plant-derived substances on beetles, such as the research on the sublethal effects of glycoalkaloids on *T. molitor* L. [130], as well as their mode of action (Table 1), compared to the research on other groups, especially moths and butterflies (see below). We think that basic research on these sublethal effects, including the analysis of both behavioral and physiological changes, may provide plenty of important data, which can be applied in studies leading to novel natural insecticides.

2.2.2. Butterflies and Moths (Lepidoptera)

The range of lepidopteran species representing pests of grains and the research on plant-derived substances tested against them is wide (Table 1). The most important pests affecting grains are found within the Pyralidae family, although *Sitotroga cerealella* Oliv., belonging to Gelechiidae, is also considered a serious pest affecting grains.

S. cerealella Oliv. burrows in grains and makes them unfit for human consumption. The families Asteraceae, Bignoniaceae, Fabaceae and Rubiaceae contain species with insecticidal and repellent properties with potential applications against *S. cerealella* in stored grains. The dried ground leaves and essential oil of *Ocimum kilimandscharicum* exhibited significant activity against this species in maize and sorghum grains in the laboratory. In particular, there was no adult survival or progeny production in grains treated with each of the two materials at doses of 25.0 g (dried ground leaves) and 0.3 g (essential oil) per 250 g of grain, respectively. In addition, ground leaves and the essential oil protected the grains against feeding, thus resulting in lower weight losses and numbers of damaged seeds compared with untreated grains [206]. When *Ocimum suave* as dry or ground leaves and essential oil was applied to *S. cerealella* in maize and sorghum, all treatments evoked higher mortalities in the moths, as well as significant reductions in the progeny produced by the insects [207]. Furthermore, individual plant secondary metabolites exhibit significant biological properties affecting S. cerealella. For instance, when two sesquiterpene lactones, isolated from the surface of leaves and flowers of *Cyrtocymura cincta* (Griseb.), were tested at 250 and 500 ppm in the diet of *S. cerealella* they lowered the percentage of adult emergence and produced malformations in adults, altered the oviposition capacity and viability of eggs laid and ceased the production of viable offspring [183]. In another study, the fumigant activity of *A. sativum* essential oil and its two major components, diallyl disulfide and diallyl trisulfide, expressed as a 50% lethal concentration for the adult moths, was calculated at 1.33, 0.99 and 1.02 μL/L air space, respectively. Additionally, behavioral deterrent activities were noticed, along with reduced adult longevity and inhibited oviposition by more than 70% at a concentration of 1.5 μL/25 g [143]. Considering the mode of action of diallyl trisulfide treatment, it was found to provoke a decrease in the cuticular chitin content of *S. cerealella* and to reduce the thermal stability and crystallinity of chitin [144]. Diallyl trisulfide was also found to accelerate the rate of metabolism in males at LC_{10}, leading to the accumulation of greater levels of total soluble sugar to support life activities and to the increased synthesis of proteins to resist an adverse environment [145]. Furthermore, female circadian mating rhythms and calling periodicity changed significantly after diallyl trisulfide treatment, whereas mating frequency and mating duration declined [146]. A recent study proved the ovicidal effect of extracts of *Tithonia diversifolia* (Asteraceae) flowers and *Psychotria prunifolia* (Rubiaceae) leaves, as well as astilbin from *Dimorphandra mollis* (Fabaceae) flowers if applied at 1% (w/w or m/m) to eggs of *S. cerealella*, on grains of *Triticum aestivum* (Poaceae) and on the surfaces of Petri dishes [184]. When Adeyemo and co-authors applied the powders and extracts of *Framomum melegueta* on paddies at 0.1 to 0.8 g and 1% to 5%, respectively, they observed the significant reduction or prevention of *S. cerealella* adult emergence and an increase in the developmental period as well as the reduction or prevention of paddy seed weight loss [140]. In another study, the petroleum ether extract of *A. calamus* at the application rates of 1000, 500 and 250 μg/g and the acetone extract at 1000 and 500 μg/g completely inhibited the emergence of S. cerealella adults [139]. In a fumigant bioassay the essential oil of Coriandrum sativum was tested against *S. cerealella* and the LD_{50} was calculated at 18.76 μg/cm^3. Camphor was considered the most active ingredient, with an LD_{50} of 19.31 μg/cm^3 [177]. Furthermore, Nazeri and co-authors studied the fumigant toxicity and sublethal effects of essential oils from *Artemisia khorassanica* Podl. and *Artemisia sieberi* Besser on adults of S. cerealella. They found that *A. khorassanica* (LC_{50} = 7.38 μL/liter air) was a more active fumigant than *A. sieberi* (LC_{50} = 9.26 μL/liter air), and that the insecticidal effect of *A. khorassanica* (LT_{50}: 9.01 h) was faster than that of *A. sieberi* (LT_{50} = 14.37 h). Lastly, the fecundity of *S. cerealella* was reduced by 25.29% and 35.78% following exposure to sublethal concentrations of *A. sieberi* and *A. khorassanica*, respectively [157]. The essential oil of Clausena anisata completely inhibited the viability of the larvae and the emergence of adult butterflies from the dose of 0.5 µL/mL, contrary to *Polyalthia longifolia* EO, which respectively recorded 10.0% and 50.0% at the dose of 3 µL/mL [174]. The leaf essential oils from *Cupressus lusitanica* and *Eucalyptus saligna* were proven to exhibit contact toxicity and air-in-space fumigation against *S. cerealella*, with LC_{50} values of 0.11% v/w and 7.02 µL/L, respectively [181]. In another study, *Neroli bigarad* oil (1.70 µg/cm^3) was the most toxic against *S. cerealella*, followed by *Citrus aurantium* (1.80 µg/cm^3) and *Artemisia vulgaris* (1.81 µg/cm^3) [58,170].

The Indian meal moth (*Plodia interpunctella* Hubner Pyralidae) is an economically important pest affecting various food products, including cereals, grains and various dry food products. For the most part, the references on plant extracts used to combat *P. interpunctella* report on essential oils. In this context, when the fumigant toxicity of the *C. copticum* EO was assessed against growth stages of *P. interpunctella*, it was concluded that the adults were about 500 times (LC_{50} = 257.83 µL/m^3 air and LC_{90} = 598.94 µL/m^3 air) more susceptible than other growth stages. Furthermore, last-instar larvae (LC_{50} = 91.36 µL/L air and LC_{90} = 213.79 µL/L air) and pupae (LC_{50} = 105.69 µL/L air and LC_{90} = 203.24 µL/L air) were significantly more susceptible than eggs (LC_{50} = 184.61 µL/L air and LC_{90} = 435.32 µL/L air) [226]. In a choice test of *P. interpunctella* using an olfactometer, the strongest repellency was exhibited by the essential oil of *A. graveolens* (100%), *Thymus vulgaris* (100%) and *R. officinalis* L. (93.33%) and the weakest repellency by *Hyossopus officinalis* (7.69%) and *P. sativum* (9.48%) [152]. In another study, the essential oils of myrtle, laurel, marjoram and lemon provoked mortality at 41.66%, 50.83%, 57.50% and 26.66% for *P. interpunctella* eggs, respectively. At the moderate dose (50 µL/L air) the LT_{99} values of the most effective essential oil (savory) was 81.88 h for the eggs of the moth [205]. The eggs of *P. interpunctella* were the most tolerant to the essential oils of garlic, birch (*Betula lenta*), cinnamon (*C. zeylanicum*) and aniseed (*Pimpinella anisum*), with LC_{90} values ranging from 22.02 to 72.42 µL/L air [147]. The essential oils of oregano and savory were highly effective against *P. interpunctella*, with 100% mortality obtained after 24 h at 9 µL/L air [205]. The essential oil of *Armoracia rusticana* was assessed against *P. interpunctella* and adults were found to be much more susceptible than pupae; its relatively low fumigant effect on pupae might be due to the gas vapor being unable to permeate through the thick wall of the pupal case [54]. After 9 h of exposure, the LC_{50} values of the essential oil from *S. hortensis* and *Z. officinale* for *P. interpunctella* were 139.8 µL/L and 69.05 µL/L air, respectively [129,218]. In another study, insect-resistant films and anti-insect polymer strips containing cinnamon oil were developed to protect food products from the Indian meal moth [168,169]. When *Thymus daenensis* essential oil was tested for its fumigant toxicity against first- and third-instar larvae and adults the LC_{50} values were calculated at 25.32, 34.80 and 0.27 µL/L, respectively [217]. The fumigant activity of essential oil vapors distilled from sweet basil *O. basilicum* and spearmint *Mentha spicata* were tested against *P. interpunctella* at 0.5 to 1.500 µL/L air. Adult moths were the most sensitive, with a notable mortality (>80%) recorded after exposure to low doses such as 2.5 µL/L, but other than that, basil and spearmint oils did not show satisfactory overall insecticidal activity against the moth [202]. In another study, beads of encapsulated coriander and basil EO proved to be efficacious in funnel traps in stores of almonds and pet foods against *P. interpunctella* [178]. When two Indian spices, namely, *Trachyspermum ammi* and *M. fragrans*, were studied for the fumigant activity of their essential oil at 10 µL/L air against *P. interpunctella*, *T. ammi* was found to be comparatively more effective [204]. Similarly, the lethal and sublethal effects of essential oils of *A. khorassanica* and *Vitex pseudo-negundo* were studied on *P. interpunctella*. The fumigant toxicity of *A. khorassanica* (LC_{50} = 9.60 µL/L air) was higher than that of *V. pseudo-negundo* (LC_{50} = 23.05 µL/L air) and the exposure to sublethal concentrations of *A. khorassanica* negatively affected the protein, lipid and glycogen contents of the larvae coming from treated adults [158]. In another study, the toxicity of the essential oils isolated from parsley, *P. crispum* and coriander, *C. sativum*, was studied, and the LC_{50} values were calculated to be 55.197 and 50.956 µ/L air for *P. interpunctella* larvae, respectively. In another study, *M. piperita* and *S. officinalis* were incorporated into polylactic acid solution to test for contact toxicity on *P. interpunctella*. The product showed higher contact toxicity than the pure essential oil because the ploylactic acid nanofibers cause surface tension and longer efficiency times due to the slow release. Moreover, *M. piperita* showed higher toxicity than *S. officinalis* [165]. A multilayered insect-proof film preventing contamination with *P. interpunctella* was developed based on garlic and onion EOs and their compounds chosen as efficient anti-insect agents [142]. When lavender (*L. angustifolia*), peppermint (*M. piperita*), geranium (*Geranium maculatum*), palmarosa (*Cymbopogon martini (Roxb.) Wats*), eucalyptus (*E. globulus*) and bergamot (*Citrus bergamia Risso*) were used against adult moths, the contact toxicity assay showed that the EO from palmarosa was the most toxic, with an LD_{50} value of 22.8 μg/cm^2, and the greatest fumigant toxicity was found with the EO from eucalyptus, with a KT_{50} value of 8.34 min [171]. In another study, low-density polyethylene-laminated polypropylene films printed with ink incorporating microencapsulated cinnamon oil using a large-scale film production system effectively repelled Indian meal moth larvae [167]. *Eucalyptus dives* oil and constituent 3-carvomenthenone, cyclohexanone (LD_{50} against, 2.45 and 3.63 μg/cm^3), methylcyclohexanone (2.95 and 4.24 μg/cm^3) and seudenone (3.02 and 4.44 μg/cm^3) were proven to have fumigant activity against larvae and adults of *Plodia interpunctella* [190]. In a recent study, geranium EO (*G. maculatum*) was used to develop micro and nanoemulsions, adding Tween 80 as surfactant which was stable at 25 °C for 60 days. This formulation can increase the insecticidal efficacy of EO twofold [192]. The essential oil distilled from *Lippia turbinata* ("poleo") was insecticidal on *P. interpunctell*a larvae [198]. In a recent study, the essential oils (*O. vulgare, P. anisum* and *Tanacetum cinerariifolium*) and four plant extracts (*Agastache rugosa, Capsicum annuum, C. reticulata* and *Ginkgo biloba*) were proven to be repellent against *P. interpunctella. Additionally, O. vulgare* and *T. cinerariifolium* had greatest repellent efficacy against the moth larvae [46]. In another study, insect-resistant adhesives were developed for application to a cardboard packaging system for preventing *P. interpunctella* larvae infestation. Cinnamon EO was used as an , encapsulated with maltodextrin, β-cyclodextrin and polyvinyl alcohol, in corn starch paste that was able to control insect penetration in the distribution and storage steps [166].

There are fewer studies referring to other essential oil plant extracts and powders used for controlling *P. interpunctella*. In a study conducted by Akkineye and co-authors the root bark powder of *Cleisthopholis patens* at 1.0, 2.0 and 3.0 g/20 g of the maize evoked 100% adult moth mortality within 72 h of application, whereas the stem bark powder of *C. patens* at 1.0, 2.0 and 3.0 g/20 g of the maize produced 78%–100% moth mortality within 72 h of treatment. The leaf powder was moderately effective against the adult moth at 3.0 g/20 g of the maize grain, evoking 70%–80% adult mortality within 96 h of treatment [175]. The methanol extracts of *Peganum harmala, Ajuga iva, R. officinalis L., L. stoechas, Lavandula dentata, Cistus ladanifer, Cistus salviaefolius, Cistus monspeliensis, Centaurium erythraea* and *Launaea arborescens* were tested at 500 ppm on post-embryonic development parameters of *P. interpunctella*. Most plant extracts provoked a notable decrease in larval weight, causing significant alterations on pupation and adult emergence. When applied at 500, 750 and 1000 ppm, they also affected physiological parameters such as larval reserve substances and the midgut activities of hydrolytic and detoxification enzymes [141]. Plant protease inhibitors regulate proteolytic processes in insects and are thus considered to be a potential safe weapon against insect pests, either through their direct application or via their expression in transgenic plants. In this regard, medicinal legume plants, namely, senna (*Cassia angustifolia*) and fenugreek (*T. foenum-graecum*), could be used for their total proteolytic inhibitory activity against the Indian meal moth larval midgut [164]. Another study investigated a bioinspired cyanogenic grain coating with amygdalin as a cyanogenic precursor, mimicking the feeding-triggered release of hydrogen cyanide found, for example, in bitter almonds. Upon feeding of coated cyanogenic wheat grains to *T. molitor* L., *R. dominica* Steph. (lesser grain borer) and *P. interpunctella*, their reproduction, as well as consumption rate, were significantly reduced, whereas their germination ability increased compared to non-coated grains [216]. Diterpene resin acids are important components of oleoresin and greatly contribute to the defense strategies of conifers against herbivorous insects and also function as insect juvenile hormone antagonists that interfere with the juvenile hormone-mediated binding of the JH receptor methoprene-tolerant and steroid receptor coactivator protein [211]. Hexane extracts of the roots of *Piper sarmentosum* Roxb. and constituent asaricin 1, isoasarone 2 and trans-asarone 3 were toxic to *P. interpunctella* [212]. Plant juvenile hormone disruptor activity is concentrated in certain plant groups, families and their plant metabolites, which have insect group-specific activity. Reciprocal diversification has occurred between plants and insects through the evolution of secondary plant metabolism and juvenile hormone receptors, respectively, and that plant metabolites could be developed into insect group-specific pesticides with limited effects on non-target species [227].

The rice moth (*Corcyra cephalonica* Stainton, Pyralidae) feeds on both seeds and flour. Most of the literature on natural substances against *C. cephalonica* investigate essential oils, although constraints including a lack of data for single or multiple components of essential oils in terms of their sorption, tainting and residues in food commodities, and registration protocols are yet to be highlighted [10]. *M. piperita* and *Piper nigrum* EO LC_{50} values against the larvae were calculated at 343.9 and 530.5 µL/L of air 72 h after commencement [113]. In a fumigation assay, coriander and eucalyptus oils at 130µg/cm^2, caused 100% toxicity to *C. cephalonica* within 24 h of treatment, whereas pine oil revealed 90% mortality at the same concentration after 72 h of treatment. In a contact assay, the test oils were effective against adults producing about 90% toxicity only after 72 h of treatment [82]. The fumigant toxicity of *Origanum majorana* essential oil against *C. cephalonica* adults and larvae was calculated, with LC_{50} values of 11.31 and 49.83 µL/L air, respectively. The contact toxicity against adult, pupa, larvae and eggs was observed, with LC_{50} values of 2.54, 0.95, 2.78 and 0.49 µL/L, respectively. The acetylesterase inhibition activity of *O. majorana* EO was observed against adults and larvae, with LC_{50} values of 35.89 and 118.54 µL/mL, respectively [208].

Plant extracts of *Lawsonia inermis*, petroleum ether (pet ether) extract, produced a complete inhibition of the moths, and were thus recorded as the best repellants (96.70%), whereas *Parthenium hysterophorus, Dalbergia sissoo* (acetone extracts) and *Acacia nilotica* (acetone and pet ether extracts) showed 70% repellency. However, a minimum of 30.81% repellency was exhibited by *Eucalyptus rudis* (pet ether extract) [138]. Larvicidal and pupicidal effects of *Dryopteris filix-mas* root and rhizome ethanolic extracts were studied against the third-instar larvae of *C. cephalonica. D. filix-mas* extract 0.20% (v/w) caused 100% larval mortality. The plant extracts reduced the pupation percentage, pupal death and adult emergence, indicating absolute toxicity to the pest [185]. Essential oils isolated from pine (*Pinus longifolia*), eucalyptus (*Eucalyptus obliqua*) and coriander (*C. sativum*) were screened for contact and fumigant activities. In a fumigation assay, coriander and eucalyptus oils at 130µg/cm^2 caused 100% toxicity within 24 h of treatment, whereas pine oil revealed 90% mortality at the same concentration after 72 h of treatment. In a contact assay, the test oils were effective against adults, producing about 90% toxicity only after 72 h of treatment [82]. High larvicidal, adulticidal, antifeedant and oviposition deterrent activities were exhibited by hexane extracts of *Glossocardia bosvallia* 30 and 60 mg/mL in wheat grains [193]. Acetone extracts of young leaves, old leaves, flowers and stems of groundnut plant with *Trichogramma chilonis* (Ishii) and *C. zastrowi sillemi* (Esben-Petersen) revealed their kairomonal activities under in vitro conditions against *C. cephalonica* [154].

Fumigation with essential oils of *Eucalyptus platyphylla* and *M. piperita*, at different concentrations, on adults of the almond moth, *Ephestia cautella* Walker (Pyralidae), showed that those of *M. piperita* caused the highest mortality rates [191]. The almond moth, *E. cautella* Walker, comes from the family Pyralidae. In a filter paper contact toxicity bioassay, potent toxicity was produced from buchu leaf, niaouli and rosemary oils at 2.4 mg/cm and armoise, cypress, galbanum and mace oils at 4.7 mg/cm against *E. cautella* Walker. In vapor phase toxicity bioassays with larvae, cypress, galbanum, niaouli and rosemary oils were much more effective in closed containers than in open containers, indicating that the lethal effects of these oils were largely because of action in the vapor phase [159].

In another study, early eggs of *E. cautella* Walker showed a lower LC_{50} value of 48.56 μL/L, compared to 77.75 μL/L with late eggs, when the essential oil of *Lantana camara* was used to treat them with fumigation [194]. Finally, in a recent study it was found that extracts of *Calotripis procera* roots displayed the most potent activity, with 50% of *E. cautella* Walker eggs not hatching at 10.000 ppm (1%). The chemical composition analysis of the extracts demonstrated a high presence of cardenolides, including calactin, uscharidin, 15β-hydroxy-calactin, 16β-hydroxy-calactin and 12β-hydroxy-calactin [162]. When five *Eucalyptus* species, namely, *E. camaldulensis, E. astringens, E. leucoxylon*, *E. lehmannii* and *E. rudis*, were assessed for their fumigant activity on *Ephestia* spp., the *E. camaldulensis* essential oil was more toxic against *E. cautella* and *Ephestia kuehniella* Zeller and the LC_{50} values were 11.07 and 26.73 μL/L air, respectively. In all cases the fumigant activity was strongest for the summer season oils and *E. cautella* was the most sensitive species [189]. In a choice test, rosemary and eucalyptus revealed high percentages of repellency after 5 days, at 72% and 50%, respectively, in the case of *E. kuehniella* and 71% and 54%, respectively, in case of *E. cautella*.

The most effective oil in enhancing the potency of an entomopathogenic fungus, *Metarhisium anisopliae*, against insect species was rosemary and it decreased the LC_{50} for *E. cautella* and *E. kuehniella*, respectively [148]. When the oils of *Capsicum frutescens, Anacardium occidentale, Monodora tenuifolia, Xylopia aethiopica* and *Ricinus communis* were studied at 0.5 mL and 1.0 mL dosages against the egg, larvae and adults of *E. cautella, A. occidentale* was found to be the most effective [150]. *E. angustifolia* ethyl acetate and aqueous extracts caused mortality of *E. cautella*, observed after 24 and 48 h of treatment [186]. The ethanolic oil extract of *M. tennuifolia* achieved 100% *E. cautella* moth mortality and its effect was significantly ($p < 0.05$) different from that of all other extracts. However, petroleum ether increased the effect of the other extracts on the survival of the moth, as reflected by their LD_{50} values. Regardless of the solvent used, all the oils reduced the hatchability of the eggs of the insect, and the adult emergence of the insect was prevented by all the oil extracts [151]. Another study was conducted to evaluate the activity of the neem oil of *Azadirachta indica* and *Bacillus thuringiensis* alone and in combination in controlling *E. cautella*. The *B. thuringiensis* LC_{50} was at 0.1% concentration after one day of treatment, whereas the effect of the combination of *B. thuringiensis* and neem oil showed an increase in the mortality rate of larvae treated with the combination, as compared with the treatment with each separately [160].

The Mediterranean flour moth (also known as the mill moth, *Ephestia kuehniella*) is another species from the same family and genus as *E. cautella*, and is similarly an important pest affecting grains and flour. *E. camaldulensis* essential oil was toxic against E. cautella and the LC_{50} value was 11.07 μL/L air, whereas the median lethal time (LT_{50}) value was 13.49h [189]. Exposure to vapors of essential oils from anise and cumin resulted in 100% mortality of *E. kuehniella* eggs. Oregano achieved mortality as high as 89%, whereas eucalyptus and rosemary achieved 45% and 65%, respectively [180]. In another study, the average mortality rates with a dose of 10 μL of essential oils of *Origanum acutidens, Satureje hortensis, Hypericum scabrum, T. vulgaris, Micromere fruticosa, Salvia limbata C. A. Meyer, S. nemerosa* and *Hyssoppus officinalis* were approximately 74%, 66%, 73%, 4%, 12%, 7%, 10% and 14% for *S. granarius*, and 79%, 62%, 72%, 24%, 24%, 6%, 0% and 14% for *E. kuehniella*, respectively [117]. Essential oil vapors of Micromeria fruticosa, Nepata racemosa and *O. vulgare* tested at 8 μL/L air provoked 100% mortality of the larvae (third instar) of *E. kuehniella* after 120 h of treatment [203]. Essential oil vapors from the plant species *O. acutidens* achieved 100% mortality with a dose of 2 μL/L air within 96 h in third-instar larvae of *E. kuehniella* [124]. Assessing the ovicidal activity of five essential oil vapors distilled from savory *Satureja thymbra*, laurel *L. nobilis*, myrtle *Myrtus communis*, lemon *Citrus limon* and marjoram *O. majorana*, the essential oil obtained from savory produced 100% mortality for the eggs of *E. kuehniella* and at the moderate dose (50 μL/L air) the LT_{99} values of the most effective essential oil, savory, were 158.50 and 81.88 h for the eggs of *E. kuehniella* [172]. The effects of vapors of essential oils from garlic, birch (*B.*

lenta), cinnamon (*C. zeylanicum*) and aniseed (*Pimpinella anisum*) were studied on eggs of *E. kuehniella*. Generally, garlic and birch essential oils were more toxic to the eggs of the tested insect species than cinnamon and aniseed essential oils. Garlic and birch essential oils were found to be very promising since they also showed high fumigant toxicity on eggs of *E. kuehniella* [147]. When the essential oils from oregano, *Origanum onites*; savory, *Satureja thymbra*; and myrtle, *Myrtus communis* were tested on *E. kuehniella* adults, oregano and savory achieved 100% mortality, obtained after 24 h at 25 µL/L air [205]. *Pistacia lentiscus* oil was toxic to *E. kuehniella* with LC_{50} = 1.84 µL/L and LC_{95} = 5.14 µL/L, respectively. At 136 µL/L air, fecundities and hatching rates were 78 eggs/female and 29.49% [213]. The preventative properties of *Cinnamomum camphora* and *Syzygium aromaticum* to the *E. kuehniella* 5th-instar larvae penetration to packaged cereals were tested by Allahvaisi and co-workers. Four days after the initiation of the experiment, the results showed that *S. aromaticum* had more of a repellency effect than *C. camphora* [165]. In another study, the toxicity of *S. hortensis* essential oil was investigated against 12-to-14-day-old larvae of the Mediterranean flour moth. LC_{50} values were calculated as 80.9 µL/L after 9 h for *E. kuhniella*, and additional repellency was evident [129]. *Origanum majorana* and *Citrus limon* were the most effective essential oils against *E. kuehniella* and the LC_{50} and LC_{99} values were estimated to be 3.27 and 5.13 µL/L of air for marjoram and 4.05 and 5.57 µ/L of air for lemon essential oils at the longest exposure time [173]. *O. basilicum* and *Z. officinale* oils produced 100% mortality within 24 h with doses of 32 µL/L air for *E. kuehniella* adults, whereas *A. graveolens* oil was the most active against *E. kuehniella* larvae. The oils were selected as having a possible application as a 30% aqueous solution for the protection of wheat flour [153]. In another study, the insecticidal activity of the essential oil from cardamom, *Elettaria cardamomum*, on adults of *E. kuehniella* was investigated, and the highest mortality of these insects was seen after 12 h [187]. The fumigant toxicity varies with season, essential oil concentration and exposure time, as shown for five *Eucalyptus* species, as tested by Ben Jemâa and co-authors against *E. kuehniella* [189]. In another study, the LC_{50} value for *E. kuehniella* after 9 h was calculated as 258.95 µL/L air. In a contact toxicity assay, *Z. officinale* essential oil's LC_{50} value for *E. kuehniella* was determined to be 0.61 µL/cm^2 and its repellency was significant [218]. The toxicity of vapors of the essential oil of *Ferula gummosa, Elettaria cardamomum* and *S. officinalis* was tested on the adults and larvae of *E. kuehniella* and the results indicated that the effect of the essential oil of *F. gummosa* was the strongest [188]. The insecticidal effect of *Cupressus sempervirens horizontalis* resin essential oil exposure (48 h) on *E. kuehniella* eggs showed 20.83% mortality at a 100 µL/L air concentration [182]. In another study, the repellent effects of essential oils from *L. nobilis* and *M. communis* against adults of *E. kuehniella* were studied. Around 20.4% and 10.6% repellency rates were observed at the lowest concentration (0.50 µL/L) of *L. nobilis* and *Myrtus commonis*, respectively. Furthermore, at the highest-concentration (2.00 µL/L air) repellency rates reached 84.2%, obtained by *L. nobilis*, and 61.3%, obtained by *M. commonis* [195]. Hematological observations showed that in comparison to fumigation, the topical application of *Callistemon viminalis* and *Ferula gummosa* oils caused a drastic reduction in the total hemocyte counts of treated larvae in a dose-dependent manner at all time intervals [161]. When *T. daenensis* essential oil and its toxicity was investigated against the first- and third-instar larvae and adults of *E. kuehniella* in terms of fumigant toxicity, the adult stage was more sensitive, whereas the sensitivity of larvae decreased from the first to the third instar [217]. Essential oils of the resins of *Pinus brutia* and *P. pinea* were found to be insecticidal on *E. kuehniella* eggs [182]. When essential oils from sweet basil *O. basilicum* and spearmint *Mentha spicata* L. were tested against on *E. kuehniella* no significant differences were found in insecticidal action and both oils produced a notable level of mortality (>80%) after exposure to low doses, such as 2.5 µL/liter. Larvae and pupae were the most tolerant stages in all cases [202]. In another study, the fumigant toxicity potential of *L. angustifolia* oil (LC_{50} = 19 µL/L air) was greater than that of *R. officinalis* oil (LC_{50} = 28 µL/L air) as regards *E. kuehniella* larvae [196]. Essential oils of *Artemisia haussknechtii* and *Citrus vulgaris* exhibited LC_{50} values of 479.13 and 110.98 µL/L air *E. kuehniella* [155].

The LC_{50} value of *Ziziphora clinopodioides Lam*. oil was estimated to be 54.61 µL/L air for *E. kuehniella* larvae and 1.39 µL/L air for adults [219]. Beads of encapsulated coriander and basil EO were tested in funnel traps in stores of almonds and pet foods for 2 months. The number of adult moth *E. kuehniella* dead captures was similar with either coriander or basil EO beads and with vapona tablets, although there were more insects alive in the control [178]. The LC_{50} values of *C. sativum* and *P. crispum* essential oils were 62.633 and 52.412 µL/L air against *E. kuehniella* [179]. Purslane oil (bulk) exhibited mortality against larvae of *E. kuehniella*, with 66.64%, 55.21%, 45.32% and 18.61% mortality rates gained at 3%, 1.5%, 0.5% and 0.2% concentrations, respectively. However, the mortality values amounted to 96.64%, 88.68%, 78.79% and 60.61% when the larvae were exposed to 0.1%, 0.5%, 0.05% and 0.005% concentrations of nano-purslane, respectively [215]. Concentrations of 5.42 and 6.81 µL/ml were obtained as LC_{50} values for *C. copticum* essential oils and thymol, and these significantly influenced the enzymatic activities of *E. kuehniella* [163]. As for *E. cautella*, as well as for *E. kuehniella*, the potential activities of four essential oils (anise, eucalyptus, garlic and rosemary) and the microbial agent *M. anisopliae* and their combinations were evaluated. The most effective oil in enhancing the potency of *M. anisopliae* was rosemary, against both insect species [149]. The LC_{50} value for *M. longifolia* essential oil in regard to *E. kuehniella* was 21,352 ppm, whereas this value for the nanoemulsion was 14,068 ppm. In addition, *M. longifolia* oil had lower durability and the 50% persistent time (PT_{50}) was 2.39 days, compared to the nanoemulsion's value of PT_{50} = 17.13 days, in the highest concentration of essential oil [199]. In another study, the effect of the essential oil of *Origanum vulgaris* was tested on the reproduction and mortality of the flour moth *E. kuehniella*. It was found to affect the pupal development. It also disrupted the reproduction of exuviated adults by extending the preoviposition period and reducing the period of egg-laying and fecundity because fecundated females could not live more than four days compared to the control group [209]. In a recent study, wormwood (*A. herba-alba*), *O. vulgare* and rue (*Ruta montana*) were evaluated for their repellent and fumigant toxic potential against larvae of the flour moth *E. kuehniella*. *Origanum* oil was the most repellent, with a 67% repellency rate, followed by *Artemisia* oil (46%) at 120 µL/mL after 2 h of exposure. The oil of *R. montana* showed attractant activity against the larvae and was the most toxic, with 56.7% of larval mortality in the first 24 h. The median lethal concentrations (LC_{50}) recorded were 11.6, 175.4 and 1100.0 µL/L air for the plant oils *R. montana, O. vulgare* and *A. herba-alba*, respectively. *R. montana* and *O. vulgare* essential oils were shown to be efficient, with high toxic and repellent properties against E. kuehniella larvae [156]. The lethal concentration values of *O. basilicum*, *Mentha pulegium* and R*uta graveolens* were, respectively, 0.96, 0.3 and 1.02 µL/L air on *E. kuehniella* [201]. In a recent study, highly synergistic effects, considering antifeedance, relative growth rate and relative consumption rate, were observed in 1.14 and 1.7 µL/L air of *L. angustifolia* in combination with 200 Gy of gamma radiation [197].

3. Practical Concerns

In recent years, environmental contamination, pesticide resistance and the destruction of nontarget organisms have led to increased support for environmentally safe pesticide alternatives to the use of synthetics. The research on botanical pesticides for pest management practices has intensified because they demonstrate a wide range of bioactivities and display contact and fumigant toxicity, as well as repellent activity, effects on oviposition and feeding deterrence. Additionally, many botanicals display low mammalian toxicity and rapid degradation. In this regard, EOs may prove to be reasonable alternatives to the more persistent synthetic pesticides as tools to control stored-product insects, even combined with gamma radiation or diatoms [228]. However, the number of commercial biopesticides based on EOs remains low and the key challenges include the stabilization processes (e.g., microencapsulation) and authorization requirements, as well as plant growing conditions and extraction processes [229]. Loading EOs in nanostructured systems represents a potential solution to their sensitivity to pH, oxygen, light and moderate temperatures. The use of biopolymers as nanocarriers provides the controlled release of the active ingredient, also ameliorating the issues of biodegradability, biocompatibility, environmental safety, low toxicity and competitive production costs [230]. Other than EO plant-based products (powders, extracts, essential oils, etc.) are also cost-effective alternatives to synthetic insecticides, and these are of great importance regarding insect pest control strategies for stored food commodities. To date, there have been serious bottlenecks in regard to their commercialization and standardization, including their low priority in agricultural policy, as well as biosafety and intellectual property rights issues [231]. The commercialization of new botanical insecticides and the market expansion of existing botanicals has lagged considerably, considering the academic interest demonstrated in the past 20 years. Some countries (such as Turkey, Uruguay, the United Arab Emirates and Australia) have relaxed regulatory requirements for specific plant extracts and oils, whereas in North America and the European Union, stricter requirements have slowed progress toward the commercialization of new products. Thus, botanicals are likely to remain niche products for some countries, while having the greatest impact in places where the source plants are readily available and where conventional products are both expensive and dangerous to users [9].

4. Conclusions

Plant extracts have been tested mainly in the form of essential oils against stored grain pests and, in some cases, the research has extended as far as including the main components of these oils.

Regarding mites, studies have addressed the effects on pests, as well as predatory species. There are three substances that have shown the best results when tested in the lowest of concentrations, even lower than benzyl-benzoate (an already industrially available product). These substances are components of the clove bud (*E. caryophyllata*) essential oil, methyleugenol, forget-me-not (*M. arvensis*) essential oil and benzaldehyde of *P. persica*. Furthermore, cinnamon (naturally containing benzyl-benzoate) has been proven to be effective against *T. putrescentiae*, *T. molitor, S. oryzae, R. dominica, P. interpunctella* and *E. kuhniella* as a contact toxicant to larvae or adults, as a repellant or as a fumigant. However, detailed knowledge of the lethal or non-lethal effects of botanical pesticides on beneficial insects is important for integrated pest management (IPM).

Regarding the impact of pesticides on Coleoptera and Lepidoptera, many plant species have been studied for their pesticidal properties. *C. copticum* was found to be active against *T. confusum*, *S. granarius, P. interpunctella* and *E. kuehniella*, whereas basil was effective as a fumigant for *S. zeamais*, *O. surinamensis* and *E. kuehniella*. Most interestingly various *Citrus* species were effective as fumigants, contact toxicants and as repellants, for both Coleopterans (*T. castaneum*, *S. oryzae* and *C. ferrugineus*) and Lepidoptera (*P. interpunctella*, *S. cerealella* and *E. kuehniella*). In addition, in regard to Lepidoptera, many substances have been evaluated for sublethal effects (*C. cincta* lowered the percentage of adult emergence, and *A. sativum* reduced adult longevity and inhibited oviposition), whereas for Coleoptera only a few studies of sublethal effects are available.

T. minuta and *T. patula* extracts were very effective fumigants against *T. castaneum* Herbst, whereas *Eucalyptus* spp. extracts were very effective fumigants against the adults of *R. dominica*, *T. castaneum* and *O. surinamensis*, but were also very effective against Lepidoptera (*E. kuehniella*, *E. cautella*, *C. cephalonica* and *P. interpunctella*) as fumigants or repellents. *C. reticulata* and *C. sinensis* essential oils were among the few substances found to be effective against Coleoptera (*T. castaneum*) larvae, as was the garlic essential oil, causing necrosis to the larvae, pupae and adults of *T. molitor* L. The fumigant properties of *C. citratus* and *Z. officinale* were the most effective fumigants against the rice weevil. Furthermore, *A. judaica* and *C. viminals* had two of the lowest LC_{50} values observed in this entire review. *L. salicifolia*, *L. nobilis* and *E. floribundi* were significantly effective repellents, whereas *O. acutidens* and *C. reticulata* were effective fumigants against *L. serricorne* and *C. ferrugineus.*

Finally, regarding Lepidoptera, the essential oil of *C. anisata* was found to be completely toxic to the larval stages, and also inhibited the emergence of adults. *M. piperita* and *S. officinalis* incorporated into polylactic acid solution showed significant contact toxicity on *P. interpunctella*, whereas *M. piperita* showed higher toxicity than *S. officinalis*. The essential oils exhibiting contact toxicity showed higher toxic effects when they were loaded on polymeric nanoparticles, as in the case of palmarosa (*C. martinii*), geranium (*G. maculatum*), and peppermint (*M. piperita*) against *P. interpunctella*. *Eucalyptus* spp. was the most tested substance in Lepidoptera, achieving 100% mortality, and it was effective as a fumigant and exhibited contact toxicity. Additionally, purslane essential oil was very effective against *E. cautella* in terms of fumigant toxicity, oviposition deterrence and persistence against larvae and adults. *M. tennuifolia* achieved 100% *E. cautella* moth mortality and reduced the hatchability of the eggs, whereas adult emergence was prevented, making it a very useful tool against *E. cautella*; as was birch essential oil, which displayed high fumigant toxicity on the eggs of *E. kuehniella*. Furthermore, *A. graveolens* oil was found to be the most active against *E. kuehniella* larvae, the life stage of the Lepidoptera species that causes the most damage. Finally, the essential oil of *O. vulgaris* was found to affect pupal development and disrupt the reproduction of exuviated adults by extending the preoviposition period and reducing the period of egg-laying and fecundity.

In general, there is a shortage of tools for combatting stored-product pests, as an alternative to the use of synthetics. With this review, we refer to the botanical substances reported in the literature of recent years to exhibit pesticidal properties of potential utility in the management of stored products.

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