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Murray B. Isman

University of British Columbia - Vancouver

Saber Miresmailli

University of British Columbia - Vancouver

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Commercial opportunities for pesticides based on plant essential oils in agriculture, industry and consumer products

Murray B. Isman · Saber Miresmailli · Cristina Machial

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Abstract In spite of intensive research on plant natural products and insect-plant chemical interactions over the past three decades, only two new types of botanical insecticides have been commercialized with any success in the past 15 years, those based on neem seed extracts (azadirachtin), and those based on plant essential oils. Certain plant essential oils, obtained through steam distillation and rich in mono- and sesquiterpenes and related phenols, are widely used in the flavouring and fragrance industries and in aromatherapy. Some aromatic plants have traditionally been used for stored product protection, but the potential for development of pesticides from plant essential oils for use in a wide range of pest management applications has only recently been realized. Many plant essential oils and their major terpenoid constituents are neurotoxic to insects and mites and behaviourally active at sublethal concentrations. Most plant essential oils are complex mixtures. In our laboratory we have demonstrated that individual constituents of oils rarely account for a major share of the respective oil's toxicity. Further, our results suggest synergy among constituents, including among those that appear non-toxic in isolation. Repellent effects may be particularly useful in applications against public health and domestic pests, but may be useful in specific agricultural applications as well. In all of these applications, there is a premium on human and animal safety that takes priority over absolute efficacy. In agriculture, the main market niche for essential oil-based pesticides is in organic food production, at least in developed countries, where there are fewer competing pest management products. There is also scope for mixing these oils with conventional insecticides and for enhancing their efficacy with natural synergists. Some examples of field efficacy against agricultural pests are discussed.

Keywords Plant essential oils · Natural pesticides · Insect repellents · Pest management

M. B. Isman (✉)
Faculty of Land and Food Systems, University of British Columbia, Vancouver, BC V6T 1Z4, Canada
e-mail: murray.isman@ubc.ca

Present Address:
S. Miresmailli
Department of Entomology, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA

Present Address:
C. Machial
Contech Inc., 7572 Progress Way, Delta, BC V4G 1E9, Canada

Introduction

Botanical insecticides, i.e., pest management products based on plant material, plant extracts, or natural products derived from plants, have long been touted as potential alternatives to conventional synthetic insecticides, presumably because the natural products would have lesser environmental and human health impacts than many of the older conventional pesticides that have had demonstrable adverse effects on nontarget organisms and ecosystems. Unfortunately this demarcation is not as clear cut as it is often portrayed in the scientific literature for two reasons: (1) some plant natural products used for pest management can be quite dangerous to humans (viz. nicotine, strychnine); and (2) the most recently introduced synthetic insecticides pose far less risk to nontarget species than there predecessors. And while the move to "reduced risk" pesticides has become a clarion call in the USA. and the European Union, there are other sources of insecticides (e.g., microbials such as *Bacillus thuringiensis*; microbial products such as the spinosyns; novel synthetic chemicals such as fipronil and indoxacarb) that meet these criteria as well as most botanicals products. Thus, in spite of the hundreds (or thousands) of research reports on the effects of plant extracts or plant allelochemicals to pest insects in the laboratory published over the past three decades, only two new botanical insecticides have been commercialized in the past 15 years (Isman 2006). These are the neem-based products, with the limonoid azadirachtin as their active ingredient (Schmutterer 2002), and those based on plant essential oils (Isman 2000).

It is difficult to obtain data on the use of botanical insecticides in most jurisdictions, but the State of California provides detailed reports of pesticide use and makes these freely available on the internet (Cal DPR 2007). Examination of the most recent report (for 2007) indicates modest use of botanical pesticides, with the majority of uses outside of agriculture (e.g. structural pest control, public health). The most heavily used botanical in agriculture remains pyrethrum (= pyrethrins), with approximately 4,500 kg used in agriculture (Table 1).

Table 1 Botanical insecticides used in California, 2007

Insecticide	Kg applied	Non-agricultural uses
D-limonene	31,340	89% used for structural pest control
Pyrethrins	7,790	51% for public health, 24% for structural pest control
Rotenone	3,350	98% for fish management
Azadirachtin (neem)	1,010	Primarily for agriculture
Sabadilla	95	Primarily for agriculture

Plant essential oils as pesticides

The volatile ("essential") oils obtained through steam distillation of aromatic plants have a long history of human use as flavouring agents in foods and beverages, and in the perfume industry. More recently they have become popular as agents for aromatherapy. Given that certain aromatic plants have seen traditional use for protection of stored products from pest infestation in many countries, it is not surprising that plant essential oils have been re-evaluated for use as pest management products. Facilitating the movement of plant essential oil-based pesticides from the laboratory bench to commercial practice in the USA in the late 1990s is the special status particular plant oils hold as "exempted active ingredients". A limited range of these oils can therefore be used in pesticides without the prerequisite of expensive and time-consuming toxicological and environmental tests normally required for registered products.

Apart from this commercial advantage in the USA, plant essential oils have other properties that make them suitable for use in insect management. These include:

1. large scale, worldwide production and trade in plant essential oils for the perfume and flavouring industries maintains low prices and abundant supplies for several of the oils;
2. many of the oils tested on insects to date appear to have multiple modes-of-action and sites-of-action in the insect nervous system and elsewhere (Enan 2001, 2005a, b; Kostyukovsky et al. 2002; Priestley et al. 2003). These may account for the wide range of pesticidal actions (viz., contact, knockdown, fumigant toxicity) and sublethal behavioural actions (viz., deterrence, repellence);
3. with rare exceptions, the essential oils and their major constituents are relatively nontoxic to mammals, with acute oral LD_{50} values in rodents ranging from 800 to 3,000 mg kg^{-1} for pure compounds and $\gg$5,000 mg kg^{-1} for formulated products;
4. owing to their volatility, the oils and their constituents are environmentally nonpersistent,

with outdoor halflives of <24 h on surfaces, in soil and in water.

Based on these attributes and the exemption from federal regulatory approval in the USA, EcoSMART Technologies introduced a range of professional, agricultural and consumer pesticides based on oils from clove leaf (*Syzygium aromaticum*, Myrtaceae) rosemary (*Rosmarinus officinalis*, Lamiaceae) peppermint (*Mentha* × *piperita*, Lamiaceae), cinnamon leaf (*Cinnamomum zeylandicum*, Lauraceae), lemongrass (*Cymbopogon nardus*, Poaceae) and thyme (*Thymus vulgaris*, Lamiaceae), beginning in 1998. Today, their professional products (for urban pest management including structural pest control and mosquito abatement) are marketed by Prentiss Inc. and their agricultural products are marketed by Brandt Consolidated Inc. EcoSMART is marketing their consumer pesticides for home and garden in the USA through major retailers such as Wal-Mart Stores Inc. A number of smaller companies have followed with similar products based on the EPA-exempt oils for agricultural applications and consumer uses.

Relationship between chemical composition and bioactivity in insects

The chemistry of plant essential oils is characterized by often complex mixtures (>50 constituents) of monoterpene and sesquiterpene alcohols, aldehydes and ketones, and the biogenetically related phenols, although it is not uncommon for an individual compound to constitute >20% by weight of an oil, or for 90+% by weight of an oil to consist of less than a dozen major constituents. Some of the major commercial sources of essential oils (e.g., thyme, *Thymus vulgaris*, rosemary, *Rosmarinus officinalis*, and lavender, *Lavendula* spp.) exist as chemically distinct races, ecotypes or chemotypes (e.g., Lahlou and Berrada 2003; Kaloustian et al. 2005).

A key question germane to the use of plant essential oils as active ingredients in formulated insecticides is that of the relationship between chemical composition and bioactivity in key target pest species. Investigating this question for plants of *Lavendula luisieri* (Lamiaceae) from Spain tested for antifeedant effects against the armyworm *Spodoptera littoralis* (Noctuidae), the Colorado potato beetle *Leptinotarsa decemlineata* (Chrysomelidae) and the green peach aphid *Myzus persicae* (Aphididae). Gonzalez-Coloma et al. (2006) found that no individual chemical constituent accounted for the observed bioactivities, suggesting synergisitic effects among constituents. In a similar type of study, Isman et al. (2008) determined chemical compositions of ten commercial samples of rosemary oil and compared these to topical LD_{50} values of the oils when applied to cabbage loopers (*Trichoplusia ni*, Noctuidae) and fall armyworms (*Pseudaletia unipuncta*, Noctuidae). For the loopers, toxicity was significantly correlated with D-limonene and with α-terpineol, but not correlated with any of the other seven compounds quantified in the oils, including the five major constituents. For the armyworms, none of the nine compounds were significantly correlated with the oils.

To explore this question further, a number of "synthetic" rosemary oils were prepared, based on the ten major constituents, admixed in proportions representing the average composition of thirty commercial rosemary oil samples. Each synthetic oil prepared was missing an individual constituent, and these oils were tested for contact toxicity against adult twospotted spider mites, *Tetranychus urticae* (Tetranychidae), on tomato, in comparison to the complete mixture and to a natural rosemary oil (Miresmailli et al. 2006). Tests with the artificial oils suggested some putative major active constituents (e.g., 1,8-cineole), some minor actives (e.g., *p*-cymene), and some inactive constituents (e.g., camphor). Unexpectedly, the mixture of four major constituents produced only 25% mortality, while the mixtures of minor constituents and of inactive constituents produced no mortality at all. The most interesting observations were that a combination of the major and minor active constituents produced 75% mortality, and the full mixture—including the putatively inactive constituents—raised mortality to more than 90%. These results confirm synergy among constituents of rosemary oil, including apparent synergy between apparently "inert" or inactive constituents and the "active" ones.

This conclusion received additional support from a further study investigating the relationship between the chemistry of *Litsea pungens* (Lauraceae) essential oil and topical toxicity to the cabbage looper (Jiang et al. 2009). In this latter study, the major actives alone again produced only 25% mortality, but

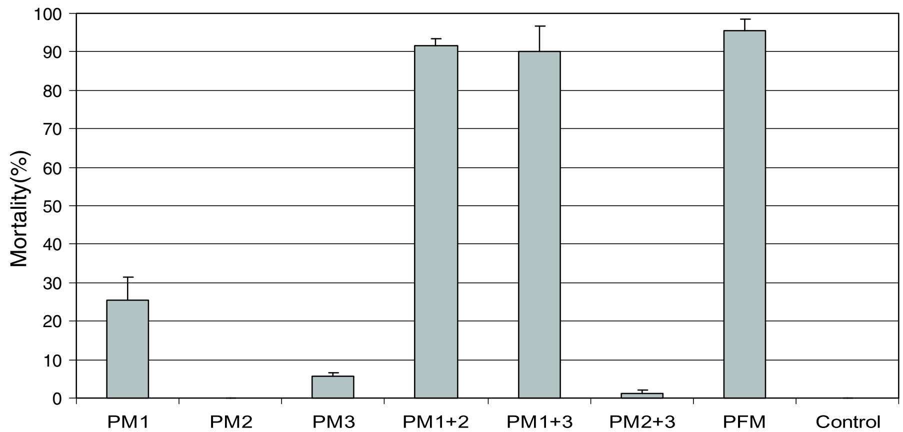


Fig. 1 Toxicity (24 h mortality) of mixtures of selected *Litsea pungens* essential oil constituents to 3rd instar cabbage looper, *Trichoplusia ni*, via topical administration. PM1 (major actives) = 1,8 cineole + carvone, PM2 (minor actives) = terpinen-4-ol + 3-carene + carene, PM3 (inactives) = D-limonene + β-caryophellene + linalool + camphene + geraniol + α-pinene, PFM = all constituents, i.e. PM1 + PM2 + PM3 (modified from Jiang et al. 2009)

combination of the major actives with either the minor actives or the inactive constituents raised mortality to > 90%, not significantly different from the full mixture, and consistent with synergy among constituents (Fig. 1).

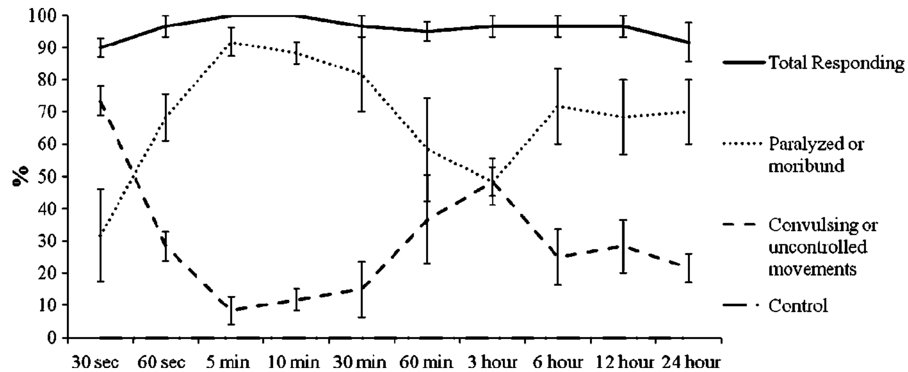
Another characteristic of plant essential oils is their marked interspecific differences in bioactivity among insects, even among closely related compounds (Isman 2000). Investigations of structure-activity relations have yielded little predictive value in this regard (Rice and Coats 1994; Tsao et al. 1995). We recently screened 17 commercially available plant essential oils for contact toxicity to the cabbage looper and to the obliquebanded leafroller (*Choristoneura rosaceana*, Tortricidae), and important fruit pest in North America (Machial et al. 2010). For the cabbage looper the most toxic oils were those from garlic (*Allium sativum*, Alliaceae), lemongrass (*Cymbopogon nardus*), and patchouli (*Pogostemon cablin*, Lamiaceae), whereas for the leafroller the most active oils were those from patchouli and thyme (*Thymus vulgaris*). Overall, patchouli oil was 1.7 times more toxic to neonate leafrollers than to neonate loopers when sprayed as an aqueous emulsion, and 3.2 times more toxic to the leafroller when applied topically to 3rd instar larvae of both species.

Observations of leafrollers treated topically with either thyme or patchouli oil indicate symptoms consistent with neurotoxicity but with an interesting time course. Within 30 s of administration, 70–80% of larvae displayed convulsions or other uncontrolled movements, but after 5 min, 90% of larvae were paralyzed or moribund. Interestingly, between 3 and 6 h, about half of all treated larvae recovered from the paralysis—possibly a consequence of enzymatic detoxification of the active principles in the oil—but by 24 h more than 90% of treated larvae succumbed to the treatment (Machial and Isman, unpublished data; Fig. 2).

Behavioural effects

In addition to the direct toxic effects of essential oils to insects via contact, ingestion or fumigation, essential oils or their chemical constituents can have potentially important behavioural effects on pests, especially as deterrents and repellents. The best known use of an essential oil in consumer products is that of lemongrass oil (also know as oil of citronella) in personal repellents applied to the skin for protection from mosquitoes and other biting insects. But these types of actions could be useful for crop protection from agricultural pests as well. For example, rosemary oil (1% emulsion) significantly inhibited oviposition by greenhouse whitefly, *Trialeurodes vaporariorum* (Aleyrodidae), on tomato plants for at least 72 h (Miresmailli and Isman, unpublished; Fig. 3). In a leaf disc choice test, adult twospotted spider mites were strongly deterred from colonizing

Fig. 2 Responses of 3rd instar obliquebanded leafrollers, *Choristoneura rosaceana*, treated topically with thyme oil (10 µg insect^{-1}) over a 24 h period of observation



tomato leaf discs treated with 1% rosemary oil for at least 12 h, and the difference in colonization on treated versus untreated discs remained significant after 48 h (Miresmailli et al. 2006). Most recently we have been investigating volatilization of individual constituents of essential oils from surfaces over time, using an ultrafast portable gas chromatograph, the zNose™ (Electronic Sensor Technology, Newbury Park, CA, USA). Key questions are: (1) do all components of an essential oil evaporate at the same rate; (2) if not, does deterrence/repellence of pests change over time as the composition of the volatile mixture changes? To address these questions we sprayed an essential oil mixture onto a glass plate, save for an area covered by a glass Petri dish, and determined the proportion of individual 1st instar obliquebanded leafrollers that moved from the unsprayed zone into the sprayed area. We tested naïve larvae ($n = 5$) every 5 min for 140 min, and simultaneously measured major constituents of the oil mixture every 5 min for the entire period of observation. We noted that larvae frequently approached the sprayed area but turned away; not a single larvae crossed into the sprayed area until 2 h had lapsed, at which point none of the larvae were deterred. The abrupt switch from deterrence to acceptance in this bioassay coincided with the loss of four of the five major monoterpenes detected by the portable GC (Miresmailli and Isman, in preparation)

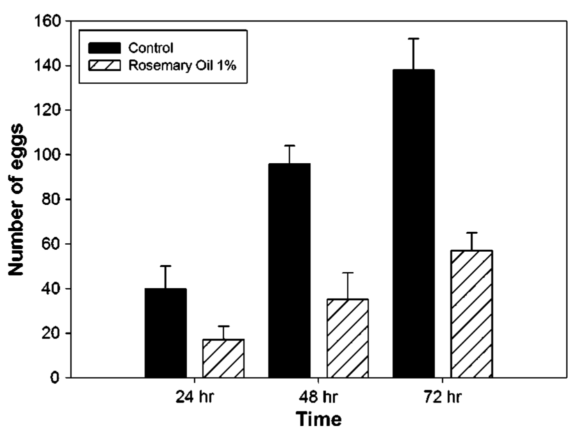


Fig. 3 Oviposition deterrence in greenhouse whitefly, *Trialeurodes vaporariorium*, on tomato by rosemary oil

A similar experiment using rosemary oil and adult twospotted spider mites more specifically assessed repellence of the volatiles emanating from the oil over time. In this case we measured directed movement of naïve mites away from the sprayed zone every 8 min ($n = 10$), and measured the volatile composition every 5 min. Over the course of 1 h we observed three distinct periods of maximum mite repellence: these coincided with peak emissions of camphor (at 25 min), 1,8-cineole and D-limonene (at 40 min) and β-pinene (at 55 min)(Miresmailli and Isman, in preparation). These experiments indicate that the mixture of volatiles emanating from a chemically complex essential oil changes over time, and more importantly, that pests can detect and respond to those changes.

Efficacy under field conditions

Toxicity and deterrence of plant essential oils to pests has been well established in the laboratory, but do these results translate into efficacy as crop protectants under true field conditions? Recent field trials in California, where essential oil-based insecticides are aimed at soft-bodied insects and mites, suggest that products of this type can be as effective as conventional insecticides, and are compatible with

Table 2 Control of thrips and aphids on lettuce and strawberries: field trials conducted in Salinas, California, August 2008

Treatment	WFT on head lettuce; thrips/sample at 3 weeks after treatment	GPA on head lettuce; aphids/sample at 3 weeks after treatment	WFT on strawberries; thrips/sample at 3 weeks after treatment
Ecotrol[a]	8.9a	1.9a	1.4a
Radiant (spintoram)	0.6b	4.1b	–
Ecotrol (0.5 rate) + Radiant (0.5 rate)	1.0b	1.8a	–
Entrust (spinosad)	–	–	3.3b
Ecotrol (0.5 rate) + Entrust (0.5 rate)	–	–	3.2b
Untreated control	14.6c	4.8b	9.1c

Different letters within a column indicate significantly different means, based on Duncan's multiple range test

Data from Pacific AgResearch and Brandt Consolidated

WFT western flower thrips, *Frankliniella occidentalis*, *GPA* green peach aphid, *Myzus persicae*

[a] Product is currently marketed under the tradename Ecotec™

conventional products in tank mixes. On a head lettuce crop, Ecotrol (an insecticide/miticide based on rosemary and peppermint oils) gave only modest control of western flower thrips (*Frankliniella occidentalis*, Thripidae), whereas a 1:1 mixture of Ecotrol with Radiant (a.i. = spintoram) each at one-half of the label rate, gave control equal to that of Radiant alone at the full label rate. For green peach aphids (*Myzus persicae*, Aphididae) in the same crop, Ecotrol provided significantly better control than Radiant, and a mixture of Ecotrol with Radiant at one-half of the label rate gave control equal to that of Ecotrol alone. On strawberries, Ecotrol gave significantly better control of western flower thrips than Entrust (spinosad); a mixture of the two products, each at one-half their label rates gave control comparable to that with Entrust alone. These results suggest that the essential oil-based insecticide can be effective as a stand alone product in certain pest/crop contexts, but may be even more effective as a tank mix with conventional insecticides (Pacific AgResearch, unpublished data; Table 2). What remains unresolved from these trials is the extent to which field efficacy is a consequence of contact toxicity, residual toxicity, deterrence or repellence (i.e., sublethal behavioural effects). Further laboratory, greenhouse and field experiments are needed that can specifically address this question.

Potential uses of essential oil-based insecticides in agriculture

Field trials conducted in the USA, Chile and elsewhere suggest that insecticides based on plant essential oils can be used as "stand alone" products for certain pests, especially soft-bodied and sucking insects and mites. They are likely to be particularly effective under low pest pressure, e.g., early in a growing season. More importantly, they can be used in rotation or in combination (tank-mixed) with other crop protectants, including conventional (synthetic or microbial) pesticide products. Advantages of using essential oil-based pesticides are the lack of harvest restrictions or worker re-entry restrictions for treated crops, owing to their low mammalian toxicity and environmental persistence. Their short residual half-lives on plants also enhance their compatibility with biological control agents and indigenous natural enemies of pests, and reduce risks to honeybees and other foraging pollinators. Disadvantages of these pesticides include their lack of persistence—when used as stand-alone products, two or more carefully-timed applications may be required to effect satisfactory management of pests. They also have limited efficacy against larger chewing insects (viz., lepidopterans, coleopterans), and can be phytotoxic if misused (e.g., applied at rates exceeding that recommended on the product label). Note however, that the disadvantages of limited persistence and phytotoxicity could be mitigated through microencapsulation of essential oils when formulated (Yang et al. 2009).

Other potentially valuable uses in non-crop agriculture are for fly control in dairy barns and for management of flies, tenebrionid beetles and ectoparasitic mites in intensive poultry production. Field trials for animal protection are underway.

Other potential uses of essential oil-based pesticides

There are numerous other opportunities for the use of essential oil-based pesticides, particularly ones where there is a premium on human and animal safety, and the relative efficacy-to-application cost ratio is less critical than it is for field and greenhouse crop protection. These products are useful for professional pest managers in the following situations:

1. for management of structural pests (viz., termites, carpenter ants), either as in situ treatments (i.e., in building foundations and wall voids), or as "perimeter" treatments;
2. for management of flies and cockroaches in commercial kitchens (restaurants, hospitals, schools), around waste containers, and in warehouses;
3. for mosquito management/abatement, either through large-scale urban fogging, or for individual property "perimeter" treatments, using controlled-release timers ("puffers").

Treatment of waterways and standing water using essential oil as larvicides is also feasible. The efficacies of essential oils as mosquito larvicides and repellents has been a very active field of investigation in recent years (e.g., Pavela 2009; Amer and Mehlhorn 2006).

Consumers (i.e., retail) products based on essential oils have several applications as well, including (1) home and garden use for flying and crawling insects and related pests; (2) management of turfgrass and landscape pests; (3) for ectoparasite (viz, flea and tick) control on dogs and cats; (4) as personal repellents for application to the skin and/or clothing to prevent/limit attack by blood-feeding flies and ticks.

Summary

Certain plant essential oils, or their chemical constituents, are toxic to a broad spectrum of economic insect pests, with some selectivity favouring biocontrol agents (Miresmailli et al. 2006). In addition to their acute toxicity to pests, deterrence and/or repellence may contribute to overall efficacy against some pests, both in agricultural contexts and in urban/domestic pest management. Most plant essential oils are chemically complex, which enhances their efficacy owing to synergy among constituents as recently demonstrated. For crop protection, products of this type can be used in rotation or in combination with other insecticides, potentially lessening the overall quantities applied and possibly mitigating or delaying the development of resistance in pest populations. Of utmost importance, products based on plant essential oils are in the vast majority of cases safe to the user, the consumer and the environment. As many conventional pesticide products fall into disfavour with the public, essential oil-based pesticides should become an increasingly popular choice for pest management.

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