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Fumigation toxicity of monoterpenoids to several stored product insects

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Abstract

Twenty naturally occurring monoterpenoids were evaluated in a preliminary fumigation screening test on some important stored-product pest insects, including the rice weevil, *Sitophilus oryzae*, the red flour beetle, *Tribolium castaneum*, the sawtoothed grain beetle, *Oryzaephilus surinamensis*, the house fly, *Musca domestica*, and the German cockroach, *Blattella germanica.* Cineole, *l*-fenchone, and pulegone at 50 µg/ml air caused 100% mortality in all five species tested. Ketone compounds were generally more toxic than other monoterpenoids. Three monoterpenoids, the ketones pulegone, *l*-fenchone, and the aldehyde perillaldehyde, were selected for further study. They were effective against *T. castaneum* in the fumigation assay; however the toxicity was relatively low in comparison to dichlorvos. LC_{50} values of these three monoterpenoids tended to decrease at longer exposure times and higher temperatures. Inclusion of either maize kernels or house fly medium (HFM) increased LC_{50} values, HFM more so than maize kernels. Monoterpenoids may be suitable as fumigants or vapor-phase insecticides because of their high volatility, fumigation efficacy, and their safety.

Keywords: Monoterpenoid; Insecticide; Stored product pests; Fumigation toxicity

1. Introduction

Post-harvest insect pests can cause serious losses during product storage, reducing the quantity and/or quality of the stored products (Evans, 1987). Fumigants, which must be toxic in the gaseous state, have been used for many years for the control of these insects (Moffitt and Burditt,

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1989; Taylor, 1994). At least 16 chemicals have been registered as fumigants, but because of concern for human safety, methyl bromide and phosphine are the primary fumigants currently being used commercially for stored products (Bond, 1984; Evans, 1987; Taylor, 1994). In spite of the diverse biological activities of methyl bromide in agriculture, use of this ozone-depleting chemical is scheduled to be phased out in the United States by 2005 under the Montreal Protocol. Several alternatives for methyl bromide are being considered, including carbon dioxide (from compressed gas or a solid formulation) (White and Jayas, 1993), mixtures of contact insecticides and insect growth regulators, reduction of the oxygen content by using nitrogen and other gases, and storage temperature control (Donahaye et al., 1985; Moffitt and Burditt, 1989). Dichlorvos is a potent chemical insecticide known for many years for its broad spectrum activity and high efficacy, but is classed as a Class C human carcinogen.

Additional problems with controlling post-harvest insects include residues of applied chemicals on grain (Jessup and Sloggett, 1993), phytotoxicity to the grain, and the development of resistant strains of insect pests. There are several reports on resistance to organic chemicals applied against the red flour beetle *Tribolium castaneum* (Herbst) (Zettler and Cuperus, 1990; Zettler, 1991; Donahaye et al., 1992). Halliday et al. (1988) reported that 50% of field strains of *T. castaneum* in the southern United States were resistant to dichlorvos, and cross-resistance existed between dichlorvos and pirimiphos-methyl. Therefore, additional fumigants and control measures are required and have been studied by some researchers (Jilani et al., 1988; Talukder and Howse, 1993; Watters et al., 1983).

Monoterpenoid compounds have been considered as potential pest control agents because they are acutely toxic to insects and possess repellent (Watanabe et al., 1993) and antifeedant-properties (Hough-Goldstein, 1990). Previous laboratory evaluations of monoterpenoids on various insect pests have established their biological activity as ovicides, fumigants, and contact toxicants (Karr and Coats, 1988; Rice and Coats, 1994; Tsao et al., 1995). Pulegone, a constituent of some mint oils, has been considered effective as a defensive chemical in part because of repellency (Mason, 1990), but also because it interferes with insect development and reproduction (Gunderson et al., 1985). Acute toxicity of pulegone to various insects was also demonstrated by Harwood et al. (1990).

This paper reports the results of two studies. The first preliminary study examines the fumigation toxicity of 20 monoterpenoid compounds to three stored-product insect pests, the red flour beetle, *T. castaneum*, the rice weevil, *Sitophilus oryzae* (L.), and the sawtoothed grain beetle, *Oryzaephilus surinamensis* (L.), as well as two public health insects, the house fly, *Musca domestica* L. and the German cockroach, *Blattella germanica* (L.). In addition to being public health pests, house fly larvae can infest stored fish meal, and German cockroaches can be a pest of stored pet food. In the second study, we examined the fumigation activity of three monoterpenoids (pulegone, *l*-fenchone, and perillaldehyde) (Fig. 1) against the red flour beetle and compared their activities to dichlorvos. Among stored-products insect pests, the red flour beetle is one of the most prevalent (Zettler, 1991). We examined the effects of different exposure times and temperatures of these three compounds to the red flour beetle. Evans (1987) noted that temperatures of 35–40°C reduced oviposition and survivorship in most stored-product insects. In addition, we examined the effect of the presence of maize kernels or house fly larval medium (HFM) in the exposure vessels on toxicity of pulegone to the red flour beetle.

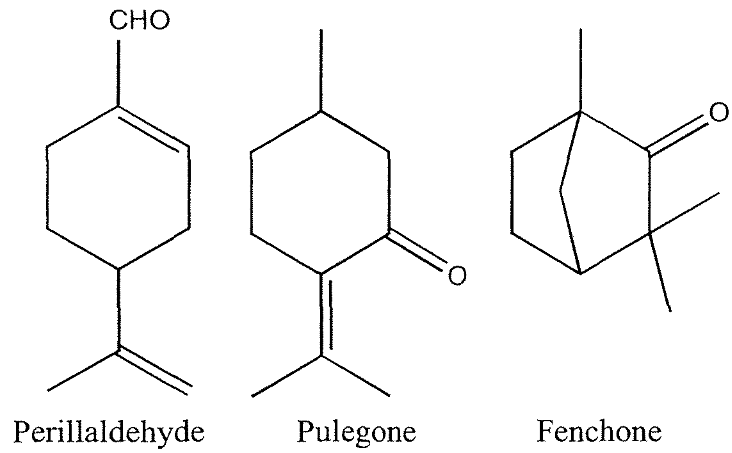

Fig. 1. Structures of perillaldehyde, pulegone and fenchone.

2. Materials and methods

2.1. Chemicals

The monoterpenoids were purchased from Aldrich (Milwaukee, WI), Sigma (St. Louis, MO), or Pfaltz and Bauer (Waterbury, CT). Dichlorvos (analytical grade) was purchased from Chemservice (West Chester, PA). All solvents were of certified grade and all chemicals were of over 95% purity.

2.2. Insects

All the test insects were supplied from laboratory colonies maintained under conditions of 25 ± 1°C, 40–60% relative humidity (r.h.), and a photoperiod of 14:10 h (L:D) in the Pesticide Toxicology Laboratory of the Department of Entomology, Iowa State University. House flies (Orlando Regular strain) were reared as larvae on HFM (Purina Labchow® Ralston Purina, St. Louis, MO), and as adults on a diet of powdered milk and sugar as described by Saito et al. (1992). German cockroaches were maintained in a 38-l glass aquarium on commercial dry cat chow. Red flour beetles were reared in 3-l glass jars on whole wheat flour and HFM. Sawtoothed grain beetles were maintained in 1-l glass jars on a whole wheat:oatmeal diet (1:1, v/v), and rice weevils were maintained in 1-l glass jars on untreated maize.

2.3. Preliminary fumigation test

The mortality caused by 50 μg/ml air of 20 monoterpenoids in the form of alcohols, phenols, ketones, and other miscellaneous monoterpenoids was evaluated on all five test species. This primary test was slightly modified from the method described by Rice and Coats (1994). Ten adult house flies, five male adult German cockroaches, 20 adult red flour beetles, 20 adult rice weevils, or 20 adult sawtoothed grain beetles were placed separately in small glass cylinders (6 × 2 cm) which were fitted on both ends with a fine stainless-steel screen secured with paraffin film, leaving an area open for gas exchange. The cylinders were then suspended by a wire in the center of a 2.78-l amber jar. Compounds were diluted in corn oil and applied to the jars to constitute 50 μg compound/ml of air in the fumigation jar. Doses were calculated based on nominal concentrations and assumed 100% volatilization of the compounds in the exposure vessel. Pure corn oil was used as a control. Each jar was equipped with a magnetic stir bar and magnetic stirring was continuous to ensure homogeneous distribution of the volatilized compounds. The jars were then sealed tightly with lids, and mortality observations were made after 14 h of exposure to the compounds. Each test was replicated three times, and the data are reported (Table 1) as the mean of the three replicates ±SEM ($\alpha = 0.05$).

Table 1
Percentage mortality (±SEM) of 20 monoterpenoids compounds to five species[a] of stored product insects

Monoterpenoids	% Mortality±SEM				
	HF	GC	RFB	RW	SGB
Alcohols and phenols					
Carvacrol	100	0	10±10	10±5.8	86±8.3
Carveol	90±6.7	50±5.8	0	0	100
Carvomenthenol	100	50±5.8	30±10	0	100
Citronellol	30±10	0	0	0	100
Linalool	100	100	10±5.8	0	100
Menthol	100	30±11.7	0	0	5±2.9
Terpineol	20±11.6	0	0	0	100
Verbenol	90±5.8	0	0	10±10	100
Isopulegol	100	0	20±10	50±5.8	100
Ketones					
d-carvone	100	80±5.8	50±5.8	10±5.8	100
l-carvone	100	80±5.8	50±0	20±0	100
l-fenchone	100	100	100	100	100
Menthone	100	100	100	90±6.7	100
Pulegone	100	100	100	100	100
Thujone	100	100	90±0	90±6.7	100
Verbenone	100	50±5.8	60±11.6	80±5.8	100
Miscellaneous					
Citronellal	100	0	0	0	100
Citral	80±5.8	0	0	10±0	90±5.8
Cineole	100	100	100	100	100
Limonene	100	100	60±10	100	100
Control	0	0	0	0	0

[a] HF = house fly, GC = German cockroach, RFB = red flour beetle, RW = rice weevil, SGB = sawtoothed grain beetle.

2.4. Fumigation test of three monoterpenoids

Approximately 15 adult red flour beetles were placed in a small glass cylinder that contained a small amount of a diet (brewer's dried yeast, dehydrated alfalfa meal, and wheat bran), and the cylinder was secured as described in the previous section. Red flour beetle adults were selected for the advanced tests because of convenience of rearing and their hardiness. Three or four small glass cylinders with insects were suspended in the center of a Mason jar (500-ml volume). These cylinders constituted sampling units within the replication. The ketones pulegone and *l*-fenchone and the aldehyde perillaldehyde were chosen for further study. Perillaldehyde, although not examined in our preliminary study, was found to be toxic to house flies and corn rootworm larvae in a previous study (Lee et al., 1997). The biological activities of pulegone, *l*-fenchone, and perillaldehyde have been reviewed by others (Gunderson et al., 1985; Mason, 1990; Harwood et al., 1990; Kang et al., 1992). The selected monoterpenoids and dichlorvos (used as a standard) were diluted with acetone to the appropriate concentrations. The appropriate dilution of monoterpenoid was administered in 100 μl portions on Whatman No. 1 filter paper (9 cm) with an Eppendorf pipette tip, and then the acetone was allowed to evaporate off the paper for 2 min outside of the jar. We had found previously that 2 min of drying time was sufficient to prevent solvent toxicity (data not shown). After the drying, the filter paper was placed in the bottom of the jar, the lid was put on, and the insects were held at 40–60% r.h. with a photoperiod of 14:10 h (L:D). The number of dead insects was counted after 24, 48, 72, and 96 h exposure to the compounds. Tests were replicated three times for each compound. As well, the treatments were subjected to three temperatures (24°C, 37°C, and 40°C) for each exposure period. Also, additional jars in the pulegone treatment were filled with HFM or maize. This was done to evaluate the effects of the inclusion of a stored product on pulegone toxicity. The treatment means were subjected to probit analysis to estimate LC_{50} values (Finney, 1971; SAS Institute, 1991).

3. Results and discussion

3.1. Preliminary fumigation test

Three compounds, pulegone, *l*-fenchone and cineole at 50 μg/ml air caused 100% mortality to all species tested. Among the five species tested, the house fly and the sawtoothed grain beetle were the most susceptible, having the highest mortality of the species tested for each compound, while the red flour beetle was the most tolerant, nearly always having the lowest mortality values for any particular compound (Table 1).

In the alcohol and phenol group, carvomenthenol, linalool, and isopulegol were the only compounds to show 100% mortality to more than one species. In the ketone group, pulegone and *l*-fenchone were the most effective, and mortalities of 100% were observed for all species tested. Menthone and thujone showed 100% mortality to four and three species, respectively. No mortality was observed in the control treatments. Rice and Coats (1994) reported that some ketones were more effective fumigants than structurally similar alcohols, and our observations support this (compare the toxicity of carveol to carvone, menthol to menthone, and verbenol to verbenone). These observations raise the possibility that the presence of a carbonyl group augments toxicity. However, cineole, an ether, was more toxic than some ketone compounds in this study, so this generalisation is not always true.

Cineole and limonene were the most active monoterpenoids in the miscellaneous group, cineole producing 100% mortality to all species tested, and limonene with 100% mortality to four of the five species tested. Karr and Coats (1988) noted that high concentrations of *d*-limonene vapors caused mortality in the German cockroach and the rice weevil and oral administration accelerated growth in German cockroach nymphs.

The results of the definitive fumigation assays of the red flour beetle are presented in Table 2. Local controls were conducted for most of the treatments, and in only two cases did control mortality exceed 10%. Control mortality averaged 3.3% over all treatments.

No treatment was found to be competitive (on a weight basis) with dichlorvos in these tests. The lowest LC_{50} value obtained for a monoterpenoid was 0.2 μg/ml (with pulegone at 37°C and 96 h). The corresponding treatment with dichlorvos had an LC_{50} value of 0.008 μg/ml.

It was found that increased exposure time decreased LC_{50} values. For example, at 37°C and 24 h, pulegone had an LC_{50} value of 23.2 μg/ml, which decreased to 1.0 and 0.2 μg/ml at 48 and 96 h, respectively. In some cases, LC_{50} values leveled off after 48 h, such as in the 24°C treatment with pulegone, where the LC_{50} value at 24 h was 195 μg/ml, which decreased to 4.8 and 5.9 μg/ml at 48 and 96 h, respectively. In only one case did increased exposure time increase the LC_{50} value (fenchone at 24°C). At 24°C and 24, 48, and 96 h exposure time, control mortality averaged 2.1%, 3.4%, and 3.6%, respectively. At 37°C and 24 and 48 h exposure time, control mortality averaged 3.3% and 3.2% respectively. At 40°C and 24 and 48 h exposure time, control mortality averaged 4.0% and 4.4%, respectively.

In all but two cases, an increase in temperature caused a decrease in LC_{50} values. The greatest difference was seen with the pulegone treatment at 24 h, where the 24 h LC_{50} value was equal to 195 μg/ml, and decreased to 23.2 and <0.1 μg/ml at 48 and 96 h, respectively. The two exceptions were cases where <50% mortality was observed at the highest dose tested for the three temperatures (observed for the treatment of pulegone with HFM inclusion at 24 and 48 h). At 24 h, control mortality equaled 2.0%, 3.3%, and 4.0% at 24°C, 37°C, and 40°C, respectively. At 48 h, control mortality equaled 4%, 3.2%, and 4.4% for 24°C, 37°C, and 40°C, respectively. At 96 h, control mortality equaled 3.6% at 24°C.

The inclusion of a stored product, HFM or maize, into the fumigation vessels in all cases increased LC_{50} values, except in cases where the highest doses tested did not cause more than 50% mortality. In all cases, HFM reduced these values to a greater extent than maize. The best example of this is in the 37°C treatment after 96 h exposure, where LC_{50} values of pulegone alone, pulegone with maize, and pulegone with HFM for 24 h were 0.2, 20.9, and 72.9 μg/ml, respectively. This is likely due to the size of the interstitial spaces between maize kernels or pieces of HFM. Presumably, the presence of a product prevented monoterpenoid vapors from diffusing freely through the container, or because of sorption of the compound to the product.

4. Conclusions

Although synthetic organic chemicals have been used as an effective means of stored-product pest control for many years, many compounds have been and will be phased out because of their

Table 2
LC_{50} values and 95% Fiducial Limits (FL) of three monoterpenoids and the standard dichlorvos to *Tribolium castaneum* at various temperatures after various time intervals

Treatments	°C	Hours	LC_{50} (μg/ml)	95% FL	Control mortality (% ±SEM)
Pulegone	24	24	195	(150–257)	1.7±1.7
		48	4.8	(3.7–6.1)	3.1±1.6
		96	5.9	(5.3–6.5)	5.0±2.9
	37	24	23.2	(18.1–31.3)	4.5±0.3
		48	1.0	(0.7–1.6)	10.2±0.6
		96	0.2	(0.1–0.3)	—
	40	24	<0.1	(—)[a]	5.0±5.0
		48	<0.1	(—)	0
		96	<0.1	(—)	—
Pulegone with house fly medium	24	24	>1000	(—)	0
		48	>1000	(—)	0
		96	>1000	(—)	1.2±1.2
	37	24	>1000	(—)	0
		48	>1000	(—)	0
		72	189	(128–291)	—
		96	72.9	(52–99)	—
	40	24	>1000	(—)	2.9±1.5
		48	>1000	(—)	8.8±4.5
		96	3.8	(1.4–6.3)	—
Pulegone with maize	24	24	>1000	(—)	7.9±7.9
		48	>1000	(—)	0
		72	>1000	(—)	—
		96	>1000	(—)	8.4±0.7
	37	24	151	(102–236)	0
		48	97.2	(78–120)	0
		72	43.9	(32–61)	—
		96	20.9	(17–27)	—
Perillaldehyde	24	24	>1000	(—)	0
		48	>1000	(—)	20.9±20.9
		96	>1000	(—)	4.6±4.6
	37	24	19	(14–25)	9.1±1.4
		48	28	(22–37)	8.8±8.8
		96	17	(13–22)	—
Fenchone	24	24	—	(—)	—
		48	104	(90–125)	0
		96	170	(136–260)	2.1±2.1
	37	24	—	(—)	—
		48	46	(41–53)	0
		96	7.8	(6.8–8.9)	—

Table 2 (*continued*)

Treatments	°C	Hours	LC_{50} (μg/ml)	95% FL	Control mortality (% ±SEM)
Dichlorvos	24	24	0.09	(0.07–0.1)	0
		48	0.06	(0.01–0.07)	0
		96	0.05	(0.04–0.06)	0
	37	24	0.03	(0.03–0.04)	2.8±2.8
		48	0.02	(0.01–0.03)	0
		96	0.008	(0.007–0.01)	—

[a] Column values marked with—, (—), or—for LC_{50}, 95% FL or % mortality, respectively, indicate that these values could not be calculated.

toxicity to humans, resistance problems in insects, and environmental concerns. Fumigation has played a significant role in controlling stored-product insect pests, and alternatives will be needed in the future as replacements, e.g., for methyl bromide. The effectiveness of many plant secondary metabolites as repellents, antifeedants, and insecticides against *T. castaneum* has been studied, and this beetle has shown susceptibility to plant-derived chemicals (Jilani et al., 1988; Talukder and Howse, 1993; Xie et al., 1995). Among them, monoterpenoids are typically volatile and rather lipophilic compounds that can penetrate into insects rapidly and interfere with their physiological functions. Their mechanism of action is not understood at this time.

Although none of the monoterpenoid compounds tested here had activities comparable to dichlorvos, it is possible that some monoterpenoids have sufficient potencies to replace the more problematic fumigants and insecticides. The large volume of literature devoted to the insecticidal properties of monoterpenoids and the attention paid by researchers to these compounds are indicative of current attitudes and of the desire to find potentially safer, yet effective, pest management strategies. Further investigations are needed to increase our understanding of the effective use of these technologies.

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