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Acaricidal activities of some essential oils and their monoterpenoidal constituents against house dust mite, *Dermatophagoides pteronyssinus* (Acari: Pyroglyphidae)

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Abstract: The acaricidal activities of fourteen essential oils and fourteen of their major monoterpenoids were tested against house dust mites *Dermatophagoides pteronyssinus*. Five concentrations were used over two different time intervals 24 and 48 h under laboratory conditions. In general, it was noticed that the acaricidal effect based on LC_{50} of either essential oils or monoterpenoids against the mite was time dependant. The LC_{50} values were decreased by increasing of exposure time. Clove, matrecary, chenopodium, rosemary, eucalyptus and caraway oils were shown to have high activity. As for the monoterpenoids, cinnamaldehyde and chlorothymol were found to be the most effective followed by citronellol. This study suggests the use of the essential oils and their major constituents as ecofriendly biodegradable agents for the control of house dust mite, *D. pteronyssinus*.

Key words: Natural acaricides, Essential oils, Monoterpenoids, House dust mite, *Dermatophagoides pteronyssinus*
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INTRODUCTION

Among several species of the family Pyroglyphidae, *Dermatophagoides pteronyssinus* and *D. farinae*, have been found to be the predominant mites of household dust accounting for about 80%~90% of the total mite populations and are important sources of allergens worldwide inside homes in humid geographic areas (Hallas, 1991; Arlian *et al*., 1992). Both house dust mites are found in homes in various areas of Egypt (Rezk, 2004; Rezk *et al*., 1996). They are a major cause of respiratory allergies including asthma (Colloff *et al*., 1992; Solarz, 2001a; 2001b). They are also identified as etiologic agent of sensitization and asthma-triggering in children (Lau *et al*., 1989; Arshad and Hide, 1992).

Control of house dust mite is usually done by three different approaches: the first is a physical method such as exceptional cleaning standards (Adilah *et al*., 1997; Nishioka *et al*., 1998), the second is reducing the humidity levels (Bischoff *et al*., 1998) and the third reducing allergens by effective acaricides (Mitchell *et al*., 1985; Green *et al*., 1989). The latter option must only be employed after careful consideration. Although good control was obtained by the currently used synthetic acaricides (benzylbenzoate and tannic acid), the risk to human health would be a potential problem. This problem has led to research efforts to develop safer and efficient alternatives in controlling mites in the indoor environment. Thus, the use of plant-derived acaricides has drawn attention and has been considered as a promising alternative to chemical acaricides (Miyazaki *et al*., 1989; McDonald and Tovey, 1993; Tovey and McDonald, 1997; Kim, 2001; Kim *et al*., 2003; Rezk and Gadelhak, 2004; Rembold, 2005). Comparative data obtained by McDonald and Tovey (1993) showed that citronella oil and tee tree oil were as effective as 0.5% benzyl benzoate (synthetic acaricide). Recently, Lee (2004) compared the oil of *Foeniculum vulgare* and five of its commercial constituents (monoterpenoids) with benzyl benzoate against *D. farinae* and *D. pteronyssinus*. He found that both fenchone isomers and anisaldehyde were more effective than benzyl benzoate. However, thymol was as effective as benzyl benzoate as acaricide.

Therefore, the present work has been devoted to evaluating the miticidal activity of some commonly available essential oils and their major constituents against the house dust mite, *D. pteronyssinus*, the most important vector in asthma, in order to find new natural miticides.

MATERIALS AND METHODS

Stock culture of house dust mite

The house dust mite, *D. pteronyssinus*, was isolated from mattress dust and reared on a finely-ground mixture of dust, dried yeast and dried milk (1:1:0.5) in complete darkness (de Saint Georges-Gridelet, 1987). Stock jars were kept in an incubator at an average temperature of (25±2) °C and relative humidity of (80±5)%. Considerable numbers of different stages were available after five months for experimentation.

Plant essential oils

Fourteen essential oils were used in the present study namely: caraway, chenopodium, cinnamon, clove, eucalyptus, rosemary, garlic, geranium, lemon grass, matrecary, peppermint, rose, fennel and thyme. These oils were supplied by the Faculty of Pharmacy, University of Alexandria, Egypt.

Identification and determination of essential oils composition

The essential oil constituents (Table 1) were analyzed by gas chromatography (Hewlett-Packard 58590 II Plus) coupled with a HP-5989 B mass spectrometer detector and equipped with column HP-5 (30 m×0.25 mm×0.025 μm). Temperature program: initial temperature, 70 °C; hold 2 min; temperature rate 4 °C/min, final temperature 250 °C; hold 30 min; column flow rate 0.8 ml He/min constant, injection temperature 250 °C; injection volume 1 μl/min. The mass spectrometry was accomplished with Wiley library ID 275, and setting as: electron impact ionization mode with 70 eV electron energy. Scan mass range m/z 45~50, detector temperature 250 °C.

Table 1 Relative abundance of major constituents of the tested essential oils

Essential oil	Major constituents	Abundance (%)
Caraway	L-carvone	58.0
	Limonene	38.4
Chenopodium	*p*-cymene	33.3
	Pinene-2-ol	11.7
	Ascaridole	10.0
Cinnamon	Cinnamaldehyde der.	36.1
	Cinnamic aldehyde	12.3
Clove	Eugenol	62.3
	β-caryophyllene	19.2
Eucalyptus	1,8-cineole	45.1
	A-pinene	14.9
	A-terpineol	11.6
Garlic	Pentdecane	12.1
	Hexadecane	11.6
Geranium	Citronellol	31.0
	Citronellol formate	14.0
Lemon grass	Limonene	91.1
	Terpinene	2.2
Matrecary	Camphor	51.4
	p-cymene	20.1
	Thymol	9.5
Peppermint	Menthol	46.1
	p-menthone	18.3
Rose	Citronellol	34.4
	Geranoil	20.4
Rosemary	1,8-cineole	28.6
	Camphor	20.2
Thyme	*p*-cymene	31.5
	Thymol	18.5
Fennel	Anthol	47.6
	Pinene	11.7
	1,8-cineole	10.3

Monoterpenoids

Fourteen monoterpenoidal constituents of the tested essential oils, namely: eugenol, (S & R) carvone, borneol, carveol, benzyl alcohol, thymol, camphor, menthol, chlorothymol, cinnamaldehyde, carvacrol, cineol, and citronellol and geraniol were supplied by Aldrich Chemical Company. The structures of the tested monoterpenoidal constituents are shown in Fig.1.

Experimental treatments

Following preliminary experiments for either essential oil or monoterpenoid, five concentrations of

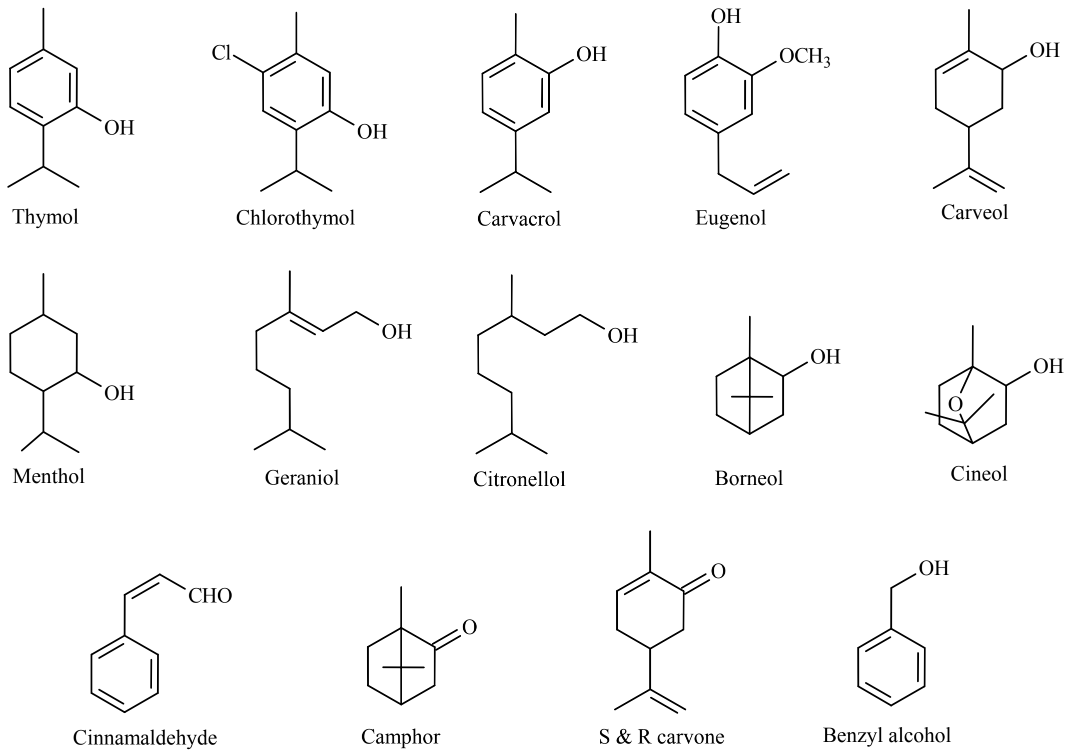

Fig.1 Structures of the tested monoterpenoidal constituents

25×10^{-6}, 50×10^{-6}, 100×10^{-6}, 250×10^{-6} and 500×10^{-6} per 0.5 g of dust and a control were selected. Each concentration was mixed with 0.5 g of house dust and placed on a microcell (3 cm×1.5 cm). Ten mites were introduced on top of the microcell with the aid a brush. Five replicates were maintained for each tested concentration. All experiments were conducted under laboratory conditions as the stock cultures were maintained. Mortality counts were done after 24 and 48 h of exposure. The LC_{50} values and their confidence limits (*CL*) were computed according to Finney (1971).

RESULTS

The effect of fourteen plant essential oils namely: caraway, chenopodium, clove, cinnamon, eucalyptus, fennel, garlic, geranium, lemon, matrecary, peppermint, rose, rosemary and thyme, on the mortality of house dust mite, *D. pteronyssinus* (Trouessart), is presented in Table 2. Five concentrations were used over time intervals 24 and 48 h under laboratory conditions. There was no mortality recorded in the controls. The 24 h-LC_{50} results showed that clove oil was the most effective (LC_{50}=29.78×10^{-6}) followed by matrecary (104.45×10^{-6}), chenopodium (117.53×10^{-6}), fennel (156.42×10^{-6}) and caraway (158.05×10^{-6}). According to the GC-MS analysis (Table 1), clove oil contains 62.3% of eugenol and 19.2% of caryophyllene. Matrecary oil containing camphor 51.4%, *p*-cymene 20.1%, and thymol 9.5%, showed good activity after 24 h of the treatment with LC_{50} value of 104.45×10^{-6}. The result of the tested oils after 48 h showed that clove oil was the most effective followed by matrecary, chenopodium, rosemary and eucalyptus. From the obtained results, clove was the most promising oil tested against the house dust mite with the LC_{50} values being 29.78×10^{-6} and 21.17×10^{-6} after 24 and 48 h, respectively.

The acaricidal activities of fourteen monoterpenoids against *D. pteronyssinus* were examined by direct application and are presented in Table 3. Responses varied according to the compound tested and the time exposure. On the basis of LC_{50} values, the compound most toxic against *D. pteronyssinus* was cinnamaldehyde (64.38×10^{-6}), followed by chloro-

Table 2 The acaricidal activities of some essential oils against house dust mites *Dermatophagoides pteronyssinus*

Essential oil	Time (h)	LC_{50} ($\times10^{-6}$)	95% fiducial limits	
			Lower	Upper
Caraway	24	158.05	179.66	192.89
	48	92.01	40.82	207.72
Chenopodium	24	117.53	97.23	142.19
	48	76.02	53.71	160.10
Cinnamon	24	390.51	260.52	586.83
	48	189.55	153.91	233.74
Clove	24	29.78	19.61	44.94
	48	21.17	15.97	27.97
Eucalyptus	24	275.42	183.93	414.46
	48	89.84	49.17	164.29
Fennel	24	156.42	122.40	185.09
	48	94.46	81.42	109.61
Garlic	24	736.30	94.01	6054.90
	48	571.31	271.73	1215.82
Geranium	24	181.53	146.07	225.93
	48	118.56	62.37	226.61
Lemon	24	300.66	213.32	425.03
	48	103.63	71.16	391.43
Matrecary	24	104.45	87.16	125.24
	48	71.64	14.52	349.55
Peppermint	24	274.27	175.57	431.16
	48	125.73	59.74	267.41
Rose	24	669.07	320.64	1405.82
	48	409.28	160.34	1064.27
Rosemary	24	375.71	238.48	594.90
	48	89.47	12.59	635.43
Thyme	24	488.65	259.86	919.04
	48	269.59	216.46	335.96

Table 3 The effects of some monoterpenoids against house dust mites *Dermatophagoides pteronyssinus*

Chemical	Time (h)	LC_{50} ($\times10^{-6}$)	95% fiducial limits	
			Lower	Upper
Carvacrol	24	318.33	157.69	652.63
	48	115.48	86.49	154.50
Camphor	24	1026.58	353.78	3018.48
	48	420.66	187.83	953.23
Citronellol	24	159.48	123.13	170.07
	48	64.89	72.81	207.01
Carvone (R)	24	669.07	320.64	1405.82
	48	253.13	118.33	547.46
Carvone (S)	24	643.89	72.81	6170.07
	48	159.48	123.13	207.01
Carveol	24	309.36	272.15	437.58
	48	85.37	72.01	101.15
Cineol	24	808.80	328.02	2023.69
	48	488.05	259.86	919.04
Cinnamaldehyde	24	64.38	56.48	73.36
	48	48.05	2.98	738.35
Chlorothymol	24	90.45	76.63	106.79
	48	45.81	37.21	56.32
Borneol	24	638.65	272.14	1506.23
	48	393.00	261.25	595.48
Benzyl alcohol	24	669.07	320.64	1405.82
	48	181.79	114.12	270.20
Geraniol	24	431.96	58.03	3329.67
	48	127.66	67.71	241.62
Menthol	24	253.68	73.06	907.36
	48	136.89	24.69	779.96
Thymol	24	345.93	127.45	957.04
	48	125.24	47.05	335.15

thymol (90.45×10^{-6}), citronellol (159.48×10^{-6}), and menthol (253.68×10^{-6}). After 48 h, the obtained results of cinnamaldehyde and chlorothymol are comparable to each other with LC_{50} values being 48.05×10^{-6} and 45.81×10^{-6} respectively.

DISCUSSION

This study showed the effectiveness of some essential oils and their monoterpenoidal constituents against house dust mite *D. pteronyssinus*. Because of the strong activities of clove oil and some others, GC-MS was used to identify the essential oils components and their essential oil activity. Thymol and the structurally related compounds like *p*-cymene are effective as acaricides (Lee, 2004). This observation is supported by the obtained results of chenopodium oil which also contains 33.3% of *p*-cymene. Matrecary oil exhibited high activity due to the presence of camphor 51.4%, *p*-cymene 20.1% and thymol 9.5%. Perhaps, there is a joint effect of those components leading to improved activity against mite. Rezk and Gadelhak (2004) studied the effect of thymol and eugenol against the adult stage of the house dust mite, *D. pteronyssinus.* They showed that thymol and eugenol had almost the same effect against the tested mite.

Interestingly, the similarity of the effect of rosemary and eucalyptus oils after 48 h may be due to 1,8-cineol as a major component which accounted for 28.6% and 45.1%, respectively (Table 1).

The chlorinated compound of thymol (chlorothymol) was much more effective than thymol itself indicating the important role of chlorine atom on the miticidal activity. Much more interesting results emerged from the potent effect of cinnamaldehyde. Therefore, cinnamaldehyde and chlorothymol merit further study as potential dust mite control agents or as lead compounds.

The public perception and acceptance of using washing product containing natural essential oils may be more positive. The most promising oil tested was clove which increased its potency over the test time. The toxic effect of these oils may be due to their fumigant and/or contact action. The vapor of some of these essential oils has been shown to kill pyroglyphidae mites (Watanabe *et al*., 1989). Further screening of these essential oils in solution is required. Criteria for choosing the most suitable oil would include its acaricidal activity with time and temperature, volatility, the acceptability of fragrance, and adverse effects on fabrics. Essential oils may also be particularly suitable for inclusion in carpet washing solutions to aid in maintaining mite control.

In conclusion, in our comprehensive screening, four essential oils, clove, matrecary, chenopodium and fennel and two monoterpenoids cinnamaldehyde and chlorothymol emerged as potential plant derived agents that provide promising results against house dust mites, *D. pteronyssinus*. The plant essential oils as a source for the control of the adult stage of house dust mite is a safe and effective approach, because many of them are selective to pests, with few if any harmful effects on non-target organisms and the environment (Susan and Ward, 1987; Kim *et al*., 2003; Kwon and Ahn, 2002). These studies suggest that essential oils and their major constituents are potentially effective, environmentally acceptable, inexpensive, simple and alternative approach for the control of house dust mites.

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