

Contact allergy to essential oils cannot always be predicted from allergy to fragrance markers in the baseline series

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Summary

Background. Essential oils are fragrance substances that are labelled on cosmetic products by their INCI names, potentially confusing consumers.

Objectives. To establish whether contact allergy to essential oils might be missed if not specifically tested for.

Methods. We tested 471 patients with 14 essential oils and 2104 patients with *Melaleuca alternifolia* oil between January 2008 and June 2014. All patients were tested with fragrance mix I, fragrance mix II, hydroxyisohexyl 3-cyclohexene carboxaldehyde, and *Myroxylon pereirae*. Three hundred and twenty-six patients were tested with hydroperoxides of limonene and linalool.

Results. Thirty-four patients had a +/++/+++ reaction to at least one essential oil. Eleven had no reaction to any of the six marker fragrance substances. Thus, 4 of 11 positive reactions to *M. alternifolia* oil, 2 of 7 reactions to *Cymbopogon flexuosus* oil, 1 of 5 reactions to *Cananga odorata* oil, 3 of 4 reactions to *Santalum album* oil and 2 of 3 reactions to *Mentha piperita* oil would have been missed without individual testing.

Conclusion. A small number of patients who are allergic to essential oils could be missed if these are not specifically tested. Labelling by INCI names means that exposure may not be obvious. Careful inspection of so-called 'natural' products and targeted testing is recommended.

Key words: contact allergic dermatitis; cosmetic reactions; essential oils; fragrances.

Contact allergy to fragrances is common. The prevalence of sensitization to fragrance mix I (FMI) in the Danish population aged 18–69 years was 1.6% in 2006 (1). Contact allergy to fragrances may affect up to 16% of patients patch tested for suspected allergic contact dermatitis in Europe (2).

The mandatory labelling scheme in Europe includes 26 fragrance substances, all of which are potential

contact allergens (3). These must be labelled if present at concentrations of more than 10 or 100 ppm in leave-on and rinse-off products, respectively. Essential oils may be found in cosmetic products as fragrance substances. They are labelled with the INCI (Linnaean) name of the plant from which they are derived. If any of the individual 26 fragrance substances are present in the essential oil, they must be listed separately on the ingredient label if present at concentrations higher than those listed above. Many products containing them are marketed as 'natural' products, which consumers associate with a sense of security, and so they may not spontaneously report their use when undergoing patch testing.

Essential oils are complex substances. They contain many different constituents, which can vary depending on which part of the plant is used, the season of harvesting, and the part of the world in which they are

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grown (4). There may also be varying degrees of oxidation of the constituents, which may alter their potential to produce an allergic response (5).

Some essential oils contain a high percentage of an allergen that is present in the fragrance markers in the baseline series and that is also available for separate testing. For example, *Syzygium aromaticum* (clove) oil contains 74–97% eugenol, *Cymbopogon flexuosus* (lemon grass) oil contains up to 85% citral, and *Citrus limon* (lemon) and *Citrus sinensis* (orange) oils contain, respectively, up to 80% and 95% limonene (4, 6). Thus, sensitization and/or contact allergy to these might be identified from reactions to markers in the baseline series [eugenol being in FMI, and citral being in fragrance mix II (FMII)]; we now routinely test with hydroperoxides of limonene] or by testing with individual fragrance substances in an extended fragrance series.

Some essential oils contain allergens available for testing, but in lower concentrations, such as *Cananga odorata* (ylang-ylang) oil (linalool up to 30% and benzyl benzoate up to 15%), or *Lavandula angustifolia* (lavender) oil (~40% linalool), which may still be detected by routine testing (4, 6).

However, other oils, such as *Santalum album* (sandalwood), *Juniperus virginiana* (cedarwood), *Mentha piperita* (peppermint) and *Melaleuca alternifolia* (tea tree) oils, contain a very low concentration, if any, of the fragrance substances that are routinely tested (4, 6). Therefore, contact allergy to these oils may well be missed if not specifically tested for.

We set out to investigate whether patients sensitized to essential oils would have been missed had they not been tested with the essential oils individually.

Patients and Methods

In this retrospective study, 471 patients were tested with 14 essential oils (including *Mentha piperita* oil, but not *Melaleuca alternifolia* oil) as part of the fragrance series between January 2008 and June 2014. An additional 23 patients were tested with *Mentha piperita* oil as part of the bakery/cheilitis series, so that a total of 494 patients were tested with *Mentha piperita* oil. A total of 2104 patients were tested with *Melaleuca alternifolia* oil, as part of the cosmetic series.

Patients were tested with the fragrance series, including the essential oils, if there was an indication from the history that fragrances might be a problem, if the patient was involved in hairdressing, beauty therapy, or aromatherapy, or if many fragranced or 'natural' or cosmetic products had been used. Patients were tested with a bakery series if they had cheilitis, dental symptoms, or oral

symptoms, including oral lichen planus, or if they worked in the catering industry.

All patients were tested with the baseline series used in Sheffield, which includes all of the allergens in the European baseline series and is regularly updated. Thus, all patients were tested with the four fragrance markers FMI [*Evernia prunastri* (oak moss absolute), isoeugenol, eugenol, geraniol, cinnamyl alcohol, cinnamal, hydroxycitronellal, and amyl cinnamal], FMII [coumarin, hydroxyisohexyl 3-cyclohexene carboxaldehyde (HICC), citral, hexyl cinnamal, citronellol, and farnesol], *Myroxylon pereirae*, and HICC. From January 2012, all patients (including 326 of the above 471 patients) were also tested with hydroperoxides of limonene and linalool.

All of the 471 patients tested with 14 essential oils were also tested with the rest of the full fragrance series, which includes 23 other individual fragrance substances. However, only 430 of the 2104 patients tested with *Melaleuca alternifolia* oil as part of the cosmetic series, and none of the 23 additional patients tested with *Mentha piperita* oil as part of the bakery/cheilitis series, were tested with the full fragrance series.

The full fragrance series was made up of 14 essential oils and the following 23 substances: cinnamyl alcohol, cinnamal, benzaldehyde, sodium benzoate, benzyl cinnamate, *Myroxylon toluiferum*, salicylaldehyde, coumarin, benzyl salicylate, benzoic acid, geraniol, hydroxycitronellal, citral, citronellol, hexyl cinnamal, amyl cinnamal, vanillin, eugenol, *Arnica* extract, jasmine synthetic, farnesol, isoeugenol, and *Evernia prunastri*.

Finn Chambers® were used. Readings were taken mostly on day (D)2 and D4, but in some patients on D3 and D5. Reactions were considered to be positive for allergy if they were +, ++, or +++ (ICDRG criteria). Allergens were obtained from Chemotechnique® (Vellinge, Sweden) or Trolab® (Reinbek, Germany) (see footnote of Table 1).

For the purposes of this analysis, relevance was mainly not considered.

Results

The positive allergic, equivocal and irritant reactions to the fragrance markers in the baseline series and to the essential oils are shown in Table 1.

Among the ~3500 patients tested with fragrance markers in the baseline series, positive allergic reactions were seen to FMI, FMII, *Myroxylon pereirae* and HICC in 5.5%, 1.4%, 3.7%, and 0.4%, respectively. Of the 1292 patients tested with hydroperoxides of limonene and linalool, 5.0% and 9.8% had positive allergic reactions.

Thirty-four patients had an allergic reaction to at least one essential oil. There were no positive reactions to

Table 1. Positive allergic, equivocal and irritant reactions to the fragrance markers in the baseline series and to the essential oils

Allergen	+ / ++ / +++ , n (%)	?+, n (%)	Irritant, n (%)	Total tested
Fragrance mix I	195 (5.5)	77 (2.2)	80 (2.3)	3541
Fragrance mix II	51 (1.4)	26 (0.7)	29 (0.8)	3537
<i>Myroxylon pereirae</i>	130 (3.7)	66 (1.9)	61 (1.7)	3502
HICC	14 (0.4)	17 (0.5)	7 (0.2)	3517
Hydroperoxides of limonene*	65 (5.0)	55 (4.3)	126 (9.8)	1292
Hydroperoxides of linalool*	126 (9.8)	85 (6.6)	176 (13.6)	1292
<i>Myroxylon toluiferum</i>	28 (5.9)	16 (3.4)	9 (1.9)	471
<i>Cananga odorata</i> (cananga) oil†	1 (0.2)	0	1 (0.2)	471
<i>Juniperus virginiana</i> (cedarwood) oil	1 (0.2)	0	1 (0.2)	471
<i>Syzygium aromaticum</i> (clove) oil	4 (0.8)	1 (0.2)	0	471
<i>Eucalyptus globulus</i> (eucalyptus) oil	0	1 (0.2)	0	471
<i>Lavandula angustifolia</i> (lavender absolute) oil	1 (0.2)	2 (0.4)	1 (0.2)	471
<i>Laurus nobilis</i> (laurel) oil	2 (0.4)	1 (0.2)	0	471
<i>Cymbopogon flexuosus</i> (lemon grass) oil	7 (1.5)	3 (0.6)	1 (0.2)	471
<i>Citrus limon</i> (lemon) oil	0	0	0	471
<i>Citrus aurantium</i> (neroli) oil	3 (0.6)	0	0	471
<i>Citrus sinensis</i> (orange) oil	0	1 (0.2)	1 (0.2)	471
<i>Mentha piperita</i> (peppermint) oil	3 (0.6)	2 (0.4)	0	494
<i>Rosa damascene</i> (rose) oil	3 (0.6)	1 (0.2)	0	472
<i>Santalum album</i> (sandalwood) oil	4 (0.8)	1 (0.2)	0	472
<i>Melaleuca alternifolia</i> (tea tree) oil	11 (0.5)	2 (0.1)	3 (0.1)	2104
<i>Cananga odorata</i> (ylang-ylang) oil†	5 (1.1)	8 (1.7)	0	475

HICC, hydroxyisohexyl 3-cyclohexene carboxaldehyde.

The following allergens were obtained from Chemotechnique: *Cananga odorata* (cananga oil), *Juniperus virginiana*, *Lavandula angustifolia*, *Rosa damascene*, *Santalum album* and *Melaleuca alternifolia* oils, fragrance mix II, hydroperoxides of limonene and linalool. Fragrance mix I, HICC, *Myroxylon pereirae* and all of the other essential oils were obtained from Trolab®. All allergens in the baseline series and full fragrance series were obtained from Chemotechnique or Trolab®.

All essential oils were tested in petrolatum, and all at 2%, except for *Juniperus virginiana* oil (10%) and *Melaleuca alternifolia* oil (5%).

*Only tested from January 2012 to June 2014. Hydroperoxides of limonene and linalool were always tested at 0.3% and 1%, respectively, but, for 1 year, they were also tested at 0.2% and 0.1%, and at 0.5% and 0.25%, respectively. The maximum reaction to any concentration on D4/5 is shown. This is therefore likely to be an overestimate of positive reactions.

†Ylang-ylang oil and cananga oil are derived from different varieties of the same species, *Cananga odorata*.

Eucalyptus globulus (eucalyptus), *Citrus sinensis* or *Citrus limon* oils. *Cymbopogon flexuosus* oil and *Cananga odorata* (ylang-ylang) oil gave the highest percentage of positive allergic reactions; 1.5% and 1.1%, respectively. Eleven of the 34 patients with positive reactions did not have an allergic reaction to any of the four fragrance substances in the baseline series (i.e. FMI, FMII, *Myroxylon pereirae*, and HICC) or to hydroperoxides of limonene or linalool (Table 2). Thus, 4 of 11 allergic reactions to *Melaleuca alternifolia* oil, 2 of 7 allergic reactions to *Cymbopogon flexuosus* oil, 1 of 5 allergic reactions to *Cananga odorata* (ylang-ylang) oil, 3 of 4 allergic reactions to *Santalum album* oil and 2 of 3 allergic reactions to *Mentha piperita* oil would have been missed if patients had been tested with the baseline series and hydroperoxides of limonene and linalool only.

The best indicator of a reaction to an essential oil was FMI, and the worst was HICC. Of the 34 patients who had a positive allergic reaction to an essential oil, 13 also reacted to FMI and none reacted to HICC.

Of the 11 patients with positive reactions to essential oils, but who did not react to any of the six fragrance substances in the baseline series, 1 patient who was allergic to *Cananga odorata* (ylang-ylang) reacted to *Myroxylon toluiferum*, isoeugenol, cinnamal, *Citrus aurantium* (neroli oil) and *Lavandula angustifolia* oil, a second patient who was allergic to *Cymbopogon flexuosus* oil reacted to geraniol, and a third patient who was allergic to *Santalum album* oil reacted to *Myroxylon toluiferum*. The others did not react to the 23 fragrance markers when these were tested separately.

Discussion

The percentages of positive reactions to the fragrance markers, FMI, FMII, *Myroxylon pereirae* and HICC, in the baseline series, were compatible with the lower end of the ranges reported in Europe (Table 3) (7), although contact allergy to HICC was less common in Sheffield than reported elsewhere. Similarly, a slightly lower percentage

Table 2. Patients with positive allergic reactions to one or more essential oil, in whom positive reactions would have been missed if they had been tested only with the four fragrance markers in the baseline series and hydroperoxides of limonene and linalool. None had an allergic contact reaction to hydroxyisohexyl 3-cyclohexene carboxaldehyde

	Total +/+/+/+	FMI	FMII	<i>Myroxylon pereirae</i>	(Oxidized) linalool	(Oxidized) limonene	None
<i>Melaleuca alternifolia</i> (tea tree) oil	11	2	0	2	1	6	4
<i>Cymbopogon flexuosus</i> (lemon grass) oil	7	4	2	1	1	1	1
<i>Cananga odorata</i> (ylang-ylang) oil	5	3	0	2	0	0	1
<i>Santalum album</i> (sandalwood) oil	4	0	0	0	0	1	3
<i>Mentha piperita</i> (peppermint) oil	3	0	0	0	1	0	2

FM, fragrance mix.

Table 3. Rates of positive allergic reactions to fragrance markers in the baseline series, in the current study and in Europe

Allergen	Current study +/+/+/+ %	Europe % Frosch et al. (7)
Fragrance mix I	5.5	3.79–13.63 (Lithuania–Austria)
Fragrance mix II	1.4	1.31–7.27 (Italy–Austria)
<i>Myroxylon pereirae</i>	3.7	3.63–11.63 (Italy–Austria)
Hydroxyisohexyl 3-cyclohexene carboxaldehyde	0.4	1.02–2.5 (Spain–Austria)

The highest and lowest reported rates in Europe are shown in column 3, the data being represented as the proportion of age-standardized and sex-standardized positive tests.

of patients in Sheffield had allergic reactions to the essential oils than described elsewhere in Europe (Table 4) (6). The latter may reflect different methods of testing, for example the source of the essential oil or the concentration used for testing. Indeed, in Sheffield, *Cananga odorata* (ylang-ylang) oil and *Santalum album* oil were both tested at 2% pet., whereas 10% was used in central Europe (6). Alternatively, the population of Sheffield may use fewer products containing essential oils.

We found no positive reactions to *Citrus limon*, *Citrus sinensis* or *Eucalyptus globulus* oils, so perhaps these could be omitted from future testing in Sheffield. The low rates of reaction to *Citrus limon* and *Citrus sinensis* oils are surprising, considering that 5% of patients tested had contact allergic reactions to hydroperoxides of limonene. This perhaps suggests a low concentration of the oxidized form of limonene in the patch test preparations of the essential oils.

Eleven of the 34 patients with positive reactions to essential oils did not react to any of the four fragrance substances in the baseline series or to hydroperoxides of limonene and linalool, with the potential for a missed allergen. Among these, the positive reaction was thought to be of current relevance in ~50%, and of unknown relevance in the rest (data not shown). The majority of missed reactions would have been to *Melaleuca alternifolia*

oil, *Mentha piperita* oil, and *Santalum album* oil, probably because these contain few and/or a low concentration of the fragrance substances found in the fragrance markers in the baseline series.

Cymbopogon flexuosus oil contains up to 85% citral (2:1 mixture of geranial and neral), and various other constituents, including small amounts of limonene and linalool (4, 6). It might be expected that sensitized patients would be identified with FMII, which contains citral, or with FMI, which contains geraniol [which auto-oxidizes to geranial and neral (10)]. However, only 2 of the 7 patients with positive reactions to *Cymbopogon flexuosus* oil reacted to FMII, and 4 to FMI. Two patients would have been missed if they had been tested only with the baseline series and hydroperoxides of limonene and linalool. One had a positive reaction to geraniol in the full fragrance series, but not FMI, but the other patient sensitized to *Cymbopogon flexuosus* oil did not react to any other fragrance substance, and so would have been missed.

Cananga odorata (ylang-ylang) oil contains up to 30% linalool and up to 15% benzyl benzoate, and ~15 other substances (4, 6). Thus, it might be expected that sensitized patients would react to hydroperoxides of linalool or *Myroxylon pereirae*, which contains benzyl benzoate. Two of the 5 patients who had positive reactions to *Cananga odorata* (ylang-ylang) oil reacted to *Myroxylon pereirae*, but none to hydroperoxides of linalool. One of the 5 patients would have been missed if testing had been performed only with the baseline series and hydroperoxides of limonene and linalool. This patient also had positive reactions to isoeugenol, *Citrus aurantium* oil, *Lavandula angustifolia* oil, cinnamal, and *Myroxylon toluiferum*, which also contains benzyl benzoate. Thus, this patient with multiple allergies to fragrance substances would have been missed if testing had been performed only with the fragrance markers in the baseline series and hydroperoxides of limonene and linalool.

Melaleuca alternifolia oil has at least 13 constituents, including terpinolene, ascardiol, α -terpinene, β -terpinene, 1,2,4-trihydroxy methane, and

Table 4. Rates of positive allergic reactions to essential oils in the current study and in Europe, the European data being the proportion of age-standardized and sex-standardized positive tests (tested along with baseline series; as special series)

Allergen	Current study +/++/+++ %	Europe % Uter et al. (6) (tested along with baseline series; as special series)
<i>Cananga odorata</i> (cananga) oil*	0.2	—
<i>Juniperus virginiana</i> (cedarwood) oil	0.2	NA; 0.83
<i>Syzygium aromaticum</i> (clove) oil	0.8	NA; 1.53
<i>Eucalyptus globulus</i> (eucalyptus) oil	0	NA; 0.24
<i>Lavandula angustifolia</i> (lavender absolute) oil	0.2	—
<i>Laurus nobilis</i> (laurel) oil	0.4	NA; 1.00
<i>Cymbopogon flexuosus</i> (lemon grass) oil	1.5	0.55; 2.25
<i>Citrus limon</i> (lemon) oil	0	NA; 0.27
<i>Citrus aurantium</i> (neroli) oil	0.6	NA; 0.71
<i>Citrus sinensis</i> (orange) oil	0	NA; 0.16
<i>Mentha piperita</i> (peppermint) oil	0.4	NA; 0.59
<i>Rosa damascene</i> (rose) oil	0.6	—
<i>Santalum album</i> (sandalwood) oil	0.8	1.26; 1.80
<i>Melaleuca alternifolia</i> (tea tree) oil	0.5	†
<i>Cananga odorata</i> (ylang-ylang) oil*	1.1	2.52; 3.93

NA, not available.

*Ylang-ylang oil and cananga oil are derived from different varieties of the same species, *Cananga odorata*.

†In other studies, the percentages of positive test reactions to tea tree oil were 0.5% in Denmark (8) and 1.8% in Australia (9).

α -phellandrene (4, 9, 11), of which ascardiol is thought to be an important allergen (12). It may contain a small amount, ~1%, of limonene (4), and, of interest, 6 of the 11 patients with positive reactions to *Melaleuca alternifolia* oil also reacted to hydroperoxides of limonene. However, 4 of the 11 patients would have been missed if only the four fragrance markers in the baseline series and hydroperoxides of limonene and linalool had been used (one was not tested with hydroperoxides of limonene). Only 3 of the 4 were tested with the full fragrance series.

Santalum album oil has at least 20 constituents, predominantly sesquiterpene alcohols, of which up to 55% are α -santalol, and up to 24% are β -santalol (4, 6). Only the tree grown in Western Australia contains an allergen that is routinely tested (up to 9% farnesol) (4). Three of 4 patients sensitized to *Santalum album* oil would have been missed if the essential oil series had not been used, and the other had a positive reaction to hydroperoxides of limonene (2 were not tested with hydroperoxides of limonene or linalool). Of the 3 patients, one had a reaction to *Myroxylon toluiferum*, but otherwise patients did not react to other allergens in the extended fragrance series, including farnesol. None of the 4 patients reacted to sesquiterpene lactone mix, but one had a reaction to propolis.

Mentha piperita oil contains ~16 constituents, including up to 49% menthol, up to 28% menthone, methyl acetate, menthofuran, isomenthone, and up to 3%

limonene and 0.4% linalool (4, 6, 13). Of the 3 patients with positive reactions to *Mentha piperita* oil, in 1 the relevance was not known, the patient being an old-age pensioner with female pattern hair loss, and, although the patient had had gum soreness after using a particular toothpaste, they could drink *Mentha piperita* tea without problems. In the other 2, the reactions were thought to be of current relevance, as 1 patient had perioral eczema, and the other worked as a baker or confectioner, and had eczema on the lower arms and generalized eczema. The latter patient also had a positive reaction to hydroperoxides of linalool, but the others did not react to other fragrance allergens in the baseline series. Thus, 2 of 3 patients would have been missed.

Mentha piperita oil, *Santalum album* oil and *Melaleuca alternifolia* oil contain few and/or a low percentage of allergens in the baseline and full fragrance series. This may explain why some patients with positive reactions to these three essential oils were missed.

However, some patients with reactions to essential oils containing a high percentage of fragrance substances contained in the fragrance markers in the baseline series were also missed. Failure to predict contact allergy with the baseline series fragrance markers has been described before (14). This may be because higher concentrations are used when the fragrances are tested alone than when they are tested as part of the fragrance mixes. Another problem may be the degree of air oxidation of the

fragrance substances. Indeed, recent work in the United Kingdom has shown the importance of testing with oxidized forms of limonene and linalool, for which such high rates of positivity have been shown that we incorporated them into our baseline series in January 2012 (15).

This study highlights a potential need to patch test with essential oils in addition to screening patients with the four fragrance markers in the baseline series and hydroperoxides of limonene and linalool, and to continue to test patients with the actual products that they use. Otherwise, a small number of patients with allergy to essential oils could be missed. This is perhaps particularly important when so-called 'natural' cosmetics have

been used, as well as when there has been contact with aromatherapy oils. It should be routine practice to quiz patients about the use of such products, and inspect labels for the INCI names of the plants from which the essential oils are derived (Table 4). Conversely, patients with contact allergy to fragrances should be warned to avoid products if the ingredients list includes such INCI names. This study supports the argument for the need to extend mandatory labelling schemes and to clearly label essential oils as fragrance substances. Indeed, a recent study suggested that 127 single fragrance substances and natural mixtures should be disclosed, owing to their skin-sensitizing potential (2).

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