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Analysis of odour compounds from scented consumer products using gas chromatography-mass spectrometry and gas chromatography-olfactometry

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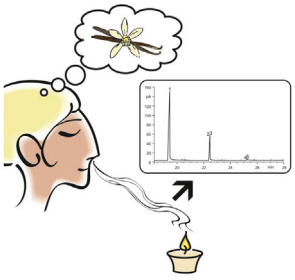
HIGHLIGHTS

- Emissions from various scented consumer products have been investigated.
- A combination of TD-GC-O and TD-GC-MS was used to detect odour-relevant substances.
- More than 300 potentially odorous compounds were identified across all products.
- Limonene and linalool were the most frequent EU-regulated fragrance allergens.
- Eugenol was one of the most frequently detected compounds in the candle emissions.

GRAPHICAL ABSTRACT



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ABSTRACT

Scented consumer products are being bought in increasing amounts and gaining more popularity. There is, however, relatively little information available about their ingredients, emissions and allergenic potential. Frequently, a mixture of different fragrance substances and not solely an individual substance contributes to the overall desired smell. The aim of this study was to investigate the odorous volatile organic compounds (OVOCs) in consumer products containing fragrances. Over 44 products were selected: various scented candles, printing products with different scent types and other products types particularly meant to be used indoors. Measurements were carried out in a desiccator. Air samples were collected on thermal desorption tubes to determine the released fragrance substances by means of gas chromatography-mass spectrometry (GC-MS). Moreover, gas chromatography-olfactometry (GC-O) was used to obtain sensory data and to ensure no important odorant was overlooked. Using both methods it was possible to distinguish between odour active and inactive compounds and subsequently to identify almost 300 different odorants across all scented products. Besides the advantage of differentiation, as the human nose is a very sensitive detector, GC-O was found to be a useful tool for detecting traces and chosen target compounds. One focus in this study lay on the 26 EU-regulated fragrance allergens to prove their relevance in scented consumer goods. In total, 18 of them were identified, with at least one substance being present in almost every product. Benzyl alcohol, cinnamaldehyde, citronellol, eugenol, linalool and limonene were the prevalently detected allergens. Particularly linalool and limonene were observed in over 50% of the products. In addition, eugenol appeared to be one of the most frequently detected compounds in trace-level concentrations in the candle emissions.

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1. Introduction

Odours play an important role in daily life. They can influence people significantly because the perception of odours is very closely linked with emotions and memories [1]. As people spend most part of their life indoors it is especially important to investigate the occurrence of odours and the perceived indoor air quality [2].

Dwellers are exposed to a variety of odours resulting from the emissions from furniture, construction materials and building products, but also from fragrance-containing products. Mostly, a complex mixture of odorous substances in different concentrations is emitted and the interaction is very often not predictable [1]. Odours of substances are reported, frequently before the estimated thresholds of irritation are reached [2,3]. Eye, nose and throat irritations and other health effects like headaches are occurring symptoms, but the prevalent complaint about odour is annoyance. Therefore, the absence of unpleasant smells is recommended to make people feel more comfortable [4]. It is important to find out the substances responsible for the odour and to reduce the emissions of products [2]. For that reason investigations are done to determine the relevant odorous compounds in interior rooms and offices [5].

Nowadays rooms and offices are often perfumed to mask malodours or to create a special atmosphere. Thus, this issue has to be taken into account when investigating indoor odours and the emissions of indoor materials and products. There are many different types of so-called air fresheners or scented products available on the market and they are gaining more popularity. One group of the most common air fresheners are scented candles which are used not only for lightning but for decoration purpose in interior rooms. Another trend, particularly used for advertising, are scented inks [6]. The possibilities for so-called Rub-and-Smell or Scratch 'N Sniff products like scented postcards, calendars or even paintings seem nearly endless. These products can release the usually encapsulated fragrance substances in a controlled way after breaking the capsules, e.g. through scratching or rubbing the printed surface [7].

Scented products may contain hundreds of individual ingredients but the formulations are often a carefully guarded secret [8]. Altogether several thousands of fragrance substances can be used for these formulations [9–11]. All these predominately attractive smelling products are supposed to give people a feeling of cleanliness and well-being [11]. Herein lies the inherent conflict: Fragrance containing products can be a strong additional source of volatile organic compounds (VOCs) and may increase the number and concentration of compounds in the indoor environment [12]. Therefore the released scent substances may even impair the indoor air hygiene. As a result the more frequent usage of fragranced home products and air fresheners also raises questions about the exposure to such compounds regarding possible upcoming health problems [13,14]. Estimates show that approximately 1–4% of the population are sensitised to fragrance substances (reported health effects or irritations from air fresheners can be even higher, e.g. Ref. [15]) and about 20% of the general population in Europe reacts allergenic to at least one contact allergen [16,17].

The exposure of consumers to the fragrances and the possible health effects are being controversially discussed. Skin reactions to fragrance chemicals have also been documented [18], but there is a lack of information about the effects of inhaling fragrances [19]. Referring to this, the question arises whether the inhalation exposure can potentially elicit allergic symptoms in already sensitised persons. In some studies no direct health effects associated with fragranced products are reported. Other studies, however, document that there is an association of asthma and other allergic effects when inhaling or having contact to these substances or their oxidation products [20–23]. Even if the substances have a weak sensitizing potency themselves, allergenic oxidation products could be formed via autoxidation [24].

Besides this, there is no declaration required for most of the consumer products. Existing regulations for fragrances apply to 24 allergenic substances plus two natural extracts of oakmoss and treemoss [18]. Their concentrations are limited in cosmetics and cleaning agents by the EU Cosmetics Directive [25]. Toys have to comply with the requirements of the Safety of Toys Directive 2009/48/EC, where the use of 55 substances is completely forbidden [26].

Still, there are relatively few publications investigating the range of VOC emissions from scented products or evaluating health risks [12–14,27]. Steinemann et al. [14], for example, found 133 different VOCs from 25 common scented products with an average of 17 VOCs per product which showed the wide range of VOCs emitted by common consumer products. Thereby, it was not yet differentiated between odour active and inactive substances. In general, for consumers who consciously choose a scented product, it may be helpful to know which odorous compounds are emitted or which substances were used for scenting.

This study describes the analysis of the emissions from certain scented consumer products. In addition to candles and other consumer goods scented printing products were selected. To our best knowledge no studies about the emissions of these printing products have been undertaken although there are a wide variety of scented inks on the market. Furthermore, they represent a group of products that are more or less completely unregulated.

As other studies about scented consumer products typically do not include sensory data the focus in this investigation was on the odorous volatile compounds (OVOCs) as they are the relevant substances when talking about fragrance usage. Particularly, the 26 EU-regulated fragrance allergens were on specific interest as they are recommended to be restricted elsewhere, for example, in candles [28,29]. The relation between measured VOCs and odour perception was also taken into account.

The emissions were determined by adsorption on Tenax® TA filled stainless steel tubes followed by thermal desorption (TD). The two chosen analysis methods were gas chromatography-mass spectrometry (GC-MS) and gas chromatography-olfactometry (GC-O). GC-O is a common method in food chemistry but it has a potential to be used in indoor science [1]. Many odorous substances are often present or added for fragrance in a very low concentration which makes it difficult to detect them by routine emission analysis using just GC-MS. They can be measured by the human nose as a very sensitive detector, for that reason GC-O was selected [1,30]. Occasionally, the combination of TD-GC-O with VOC measurements has been done before in the investigation of odour-active compounds derived from different unscented products and materials [4,30,31]. However, we couldn't find any other study using the proposed combination of chemical analysis and human-sensory testing for odorous compounds in scented consumer products.

2. Materials and methods

Different scented consumer products with a considerable odour were bought in local stores or supplied by manufacturers. Due to the wide range of scented products available for consumers and different product types a representative testing is hardly possible and a differentiation between the base material and the fragrance mixtures could not be made. Generally, products were selected to have a good mixture of quite different scents (typical scent types were preferred). The products were divided into three groups (candles (C), printing products (P), others (O)) because of their differences in product activation and treatment. The specifics for