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Analysis of transferred fragrance and its forensic implications

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ABSTRACT

Perfumes are widely used by many people in developed countries, and a large number of both men and women wear perfumes on a daily basis. Analysis of perfume trace materials from clothing is not commonly employed within forensic casework, yet as a form of trace evidence it has the potential to provide valuable intelligence. In order to appreciate the value of trace evidence there is a fundamental need for an evidence base that can both offer insight into how a trace material behaves under different scenarios and activities, and from which inferences can be made. With this purpose a gas chromatography-mass spectrometry method for trace analysis of perfumes was developed. This paper presents two different series of experiments that investigate the dynamics of perfume transfer as a factor of perfume ageing time, and as a factor of perfume contact time. Empirical data showed that both perfume ageing time, and perfume contact time play a key role in the number of perfume components transferred. These studies have implication for forensic protocols, specifically for perfume trace evidence collection, analysis, interpretation, and presentation, and there is potentially great value in analysing perfumes from clothing exhibits in forensic enquiries that involve close contact between individuals, such as sexual assaults.

1. Introduction

In many economies perfumes and fragrances are perceived as an indispensable part of life, and an increasing number of both men and women are wearing perfumes on a daily basis [1]. The British Market Research Bureau's annual Target Group Index survey identified that up to 89% of women wear fragrance on a regular basis [2], while the Men's Fragrance Track study [3] identified that 63% of adult males aged 18–64 wear fragrance periodically, and 23% use it daily.

Fragrances are volatile components by their nature, and skin and clothing can readily absorb fragrances by direct application, by contact with fragranced items, or by exposure to air containing fragrance chemicals [4]. Clothing is regularly recovered from crime scenes, or collected in the course of an investigation, and is often tested for a range of different forms of trace evidence [5]. However, as yet, there are no published studies aimed at obtaining empirical evidence concerning the behaviour of perfumes and the implications of fragrance identification for forensic enquiries. There has been limited research in the past decade on the analysis of cosmetic products for forensic applications. Research has been carried on the transfer of foundation smears [6] and on study of traces of lipstick to identify the lipstick brand [7]. Also, in a study from 2003 by Gniotek [8], an attempt was made to measure the odor firstly from a piece of cotton that was in direct contact with a fabric that had absorbed an odorant, and secondly from a piece of cotton that was in close proximity to the flavoured fabric. The headspace-gas chromatography results revealed that the first fabric absorbed 58 components of the odorant, and the second 32 components. The identification of components was not performed, as they were not present at high enough concentrations.

How significant or valuable trace material findings are depends not only on the type of trace evidence, but also on the amount of trace evidence, the location where the evidence is found, and the circumstances of the crime [9]. Understanding the persistence of different forms of trace evidence and identifying where that evidence is most likely to be deposited are a mean of developing an empirical evidence base that enables interpretation of the significance of the trace evidence that is identified, collected and analysed in the course of an investigation. Empirical evidence bases such as these have the potential to enable robust interpretation of the context of a particular form of trace evidence in a specific case, and therefore offer potentially valuable intelligence and evidence.

The evidential value of trace evidence is affected by two major factors: transference and persistence. Proof of work-studies for transfer of trace evidence, with a focus on textile fibres, first became available in 1975. In an early study by Pounds and Smalldon [10], looking at the transfer of fibres to wool and acrylic garments, it was identified that

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the force of contact and the nature of the recipient garment had a significant impact on the number of fibres being transferred. In a further study, Pounds and Smalldon [11] investigated the persistence of transferred fibres, finding an initial loss of approximately 80% after 4 h, which increased to around 95% after 24 h. Experimental work offers a means of understanding how perfume trace evidence behaves under certain circumstances and carrying out studies that mimic forensic reality is the first step towards assigning significance and weight to perfume trace evidence.

Gas chromatography coupled to mass spectrometry (GC-MS) is by far the most routinely employed analytical techniques in the perfume industry [12], but also an instrument commonly used in forensic laboratories. GC-MS has a long history and a wide application in forensic science. Its uses include but are not limited to analysis of drugs of abuse [13], of textile fibres [14], of automotive paint traces [15] and of fire debris [16]. GC-MS is particularly useful as samples are regularly composed of very complex matrices, and the results generated satisfy the requirement of high accuracy measurements with a high degree of certainty needed in court.

Multiple studies have shown that some volatile organic compounds (VOCs) are more common than others in fragranced items. In a study based on domestic and occupational products purchased on the European marked, Rastogi et al. [17] found that limonene was the most frequent component, present in 78% of the 59 products analysed, but it also had the highest mean concentration (9443 ppm). Linalool was the second most popular component, present in 61% of products. Other popular ingredients included geraniol (41%), eugenol (27%), and coumarin (25%). These five chemicals were selected to create a fragrance mix (FM) for the preliminary analyses in this study.

With a high number of people wearing perfume, and with a very reproducible analytical technique such as GC-MS, analysis of perfumes from garments can potentially aid in the investigation of crimes that involve close contact between individuals. A clear crime type that could benefit from perfume analysis as an additional piece of evidence is sexual assault, given the proximity of offender and victim during such a crime. Serious sexual assault is one of the most difficult crimes to prosecute as a great number of the offences are committed by a partner/ex-partner (52%), by a family member (6%) or by someone else known to the victim (39%), which can make inference of activity level from physical evidence difficult [18,19]. The nature of the encounter can be argued to have been consensual, and in such instances it is usually the word of one person against another, as in many cases there are no witnesses, contusions or conventional forensic evidence available [20]. In cases where the offender is a stranger to the victim, preserving the DNA is crucial evidence, however, even in these cases it has been identified that many victims bathe or shower before a forensic medical exam, making the recovery of a viable DNA sample difficult.

Due to the close contact between the victim and assailant during sexual assault, fragrance analysis has the potential to be an additional forensic tool that could be used to demonstrate a contact has taken place, and potentially indicate the type of contact made and the timeframe since the contact. However, this needs to be achieved through experimental studies to establish an evidence base for the dynamics of fragrance as a form of trace evidence, so that the identification and the interpretation of that fragrance can be transparent, reproducible and evidence based. This research addresses the current lack of published literature regarding perfume trace evidence. The main aim was to establish whether fragrance could be a viable form of trace evidence for crime reconstructions, achieved via the development of an analytical method that enables the analysis of perfumes from clothing, and also the development of a perfume transfer mechanism able to mimic the transfer of perfume between garments in order to assess the detectability of transferred fragrance.

Further, as perfume transfer is expected to be a dynamic process, this study sought to test the hypotheses that an increased perfume ageing time leads to the observation of predominately heavier molecular mass molecules onto a second piece of garment, and that the length of time two pieces of garment are in contact influences the number of perfume components transferred onto a second piece of garment.

2. Materials and methods

2.1. Chemicals and materials

Methanol (HPLC grade) was purchased from Fisher Scientific (UK). Reference standards of limonene, linalool, geraniol, eugenol, and coumarin with a purity of 96, 97, 98, 99, and 99% respectively were obtained from Sigma-Aldrich (UK).

This study focused upon a commercially available male perfume purchased from a mainstream retailer. Despite a moderate incidence of perfume use amongst men, this was chosen to reflect the high percentage of serious sexual assaults committed by men (up to 99%) [21].The fabric used was 100% cotton, and was selected because it is understood to be one of the most common garments encountered by forensic textile examiners [22]. For all perfume transfer experiments, cotton swatches of approximately 1 cm × 1 cm were used. To ensure that the fabric was not contaminated with any volatile organic compounds, a washing step was carried out ahead of any analysis. The washing was performed in a conventional washing machine without the use of detergent. After a drying step, the cotton was kept within a closed plastic bag to prevent contamination. All experiments involving transfer of perfume between cotton swatches were carried out in duplicates.

2.2. Instrumentation

GC-MS analyses were performed on a Thermo Scientific™ Trace™ 1310 Gas Chromatography system coupled to a Thermo Scientific™ ISQ™ Single Quadrupole Mass Spectrometer and connected to a Thermo Scientific™ TriPlus™ RSH Autosampler. Samples were injected on a Thermo Scientific™ TRACE TR-5MS column (30 m × 0.25 mm i.d., 0.25 μm film thickness) using liquid injection in splitless mode. Helium was employed as a carrier gas at a constant column flow of 1.2 mL/min.

The GC oven temperature was programmed from 40 °C (held for 1 min) to 270 °C (held for 3 min) at 40 °C/min rate. The injection volume was 0.2 μL and the injector temperature was set to 250 °C. The transfer line and the ion source temperatures of the ISQ mass spectrometer were set to 230 °C. The ISQ mass spectrometer was operated in Electron Impact (EI) ionisation mode at 70 eV. Data acquisition rate was of 5 spectra/s, and the mass range was scanned from m/z 10 to 400. EI mass spectrum of each component eluted from the GC column was compared with EI mass spectra from the National Institute of Standards and Technology (NIST) v 2.0 (2011) EI mass spectral library for identification.

During the experimental work, a portable thermometer-hygrometer (model 82021, VWR Scientific) was used to measure the ambient laboratory temperature and humidity. The temperature values ranged between 21 and 22 °C, and the humidity ranged between 23 and 46%.

2.3. Sample preparation for GC-MS analyses

Throughout the development of the GC-MS method, a fragrance mixture (FM) standard consisting of approximately 30 mM limonene, linalool, geraniol, eugenol and 15.2 mM of coumarin dissolved in methanol was used for the optimisation of the GC separation.

To build up a library of the components detected in the male perfume, a 1% perfume solution was prepared, by pipetting 10 μL perfume to a GC vial and adding methanol to a final volume of 1 mL.

For the analysis of fragranced fabrics, the cotton swatch was placed into a 1.5 mL glass vial following the addition of 1 mL of methanol for the extraction of the components from the fabric. The solvent was then transferred into a disposable 1.5 mL Eppendorf tube and