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Characterization of hazardous and odorous volatiles emitted from scented candles before lighting and when lit



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HIGHLIGHTS

- Emission patterns of volatiles from scented candles are assessed in a number of respects.
- Compositional changes in volatiles are found considerably before lighting and when lit.
- Strikingly, exposure levels of some species are high enough to exceed guideline values.
- Scented candles can be significant emission sources of volatiles in indoor environment.

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ABSTRACT

Scented candles are known to release various volatile organic compounds (VOCs) including both pleasant aromas and toxic components both before lighting (off) and when lit (on). In this study, we explored the compositional changes of volatiles from scented candles under various settings to simulate indoor use. Carbonyl compounds and other VOCs emitted from six different candle types were analyzed under 'on/off conditions. The six candle types investigated were: (1) Clean cotton (CT), (2) Floral (FL), (3) Kiwi melon (KW), (4) Strawberry (SB), (5) Vanilla (VN), and (6) Plain (PL). Although a large number of chemicals were released both before lighting and when lit, their profiles were noticeably distinguishable. Before lighting, various esters (n = 30) showed the most dominant emissions. When lit, formaldehyde was found to have the highest emission concentration of 2098 ppb (SB), 1022 ppb (CT), and 925 ppb (PL). In most lit scented candles, there was a general tendency to show increased concentrations of low boiling point compounds. For some scented candle products, the emission of volatiles occurred strongly both before lighting and when lit. For instance, in terms of TVOC (ppbC), the highest concentrations were observed from the KW product with their values of 12,742 (on) and 2766 ppbC (off). As such, the results suggest that certain scented candle products should act as potent sources of VOC emission in indoor environment, regardless of conditions – whether being lit or not.

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1. Introduction

Various pollutants exist in indoor atmospheric environments due to indoor sources or inflow from outside areas. Many of these pollutants have deleterious effects or are potent enough to cause odor problems and a strong nuisance [1–4]. People increasingly suffer from psychological stress and insomnia due to stress of modern life (whether real or imagined). Hence, various treatment approaches including aromatherapy are used to provide psychological relief with a healing effect [5–8]. Among such treatments, the use of scented candles has gained a great deal of attention with

http://dx.doi.org/10.1016/j.jhazmat.2014.12.040 0304-3894/© 2014 Elsevier B.V. All rights reserved. the rapidly increasing demand for room décor and indoor air fresheners. Because of the growing demand for scented candles, the U.S. market for scented candles is estimated to be worth about 2 billion USD per year [9].

Numerous sources have been identified to contribute to indoor air pollution. For instance, the use of charcoals for cooking is found to release various pollutants including metals, VOC, polycyclic aromatic hydrocarbons (PAH), and odorants [10–14]. When scented candles are combusted in an interior space, various aromatic substances are also reportedly emitted and can persist indoors. These identified compounds include, but are not limited to, various aldehydes, hydrocarbons, and alcohols. A number of PAHs identified as carcinogens (e.g., naphthalene, anthrancene, and pyrene) were also observed [15–19]. Moreover, like many indoor activities facilitating the emissions of fine and ultrafine particulates, combustion of

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scented candles can play a key role as a source of particulate matter as well as various gaseous pollutants [19,20]. The number density of ultrafine particles were reported to be up to 241,000 particles/cm³ from the combustion of pure wax candles [21]. Scented candles were also found to release a characteristic odor and a large quantity of VOC because of additives included such as fragrance and aroma oil [22].

To describe the characteristics of VOCs emitted from scented candles before and during combustion, a series of laboratory experiments was conducted using six candle types (consisting of five scented candles and one plain candle as a reference). A small impinger type chamber system was employed to collect gaseous emission samples before and during combustion of scented candles. Samples were analyzed by liquid chromatography (LC) for aldehyde while gas chromatography (GC) was used for diverse hydrocarbons. Quantification of target compounds was basically made using their working standards. In addition, quantification was also extended for a number of non-target compounds by adopting a predictive response factor (RF) approach based on the effective carbon number (ECN) method [23–25]. The ECN method permits the quantification of compounds lacking authentic standard surrogates (CLASS). With the aid of the ECN and CLASS predictive method, we were able to compile a list of quantified pollutants released from scented candle products and their emission characteristics in several aspects. In summary, based on our study, we present a critical survey on scented candle emissions as a potent indoor pollution source.

2. Materials and methods

To comprehend the characteristics of important components released from scented candles, gaseous samples were collected by an impinger system modified as a small chamber (Fig. 1S). Most VOCs were analyzed by the TD–GC/MS, while carbonyl compounds were analyzed by the HPLC/UV. For quantitative analysis, a working standard containing a total of 24 compounds was prepared. Concentrations of 68 other compounds (i.e. CLASS) were also quantified by the ECN method.

2.1. Preparation of the working standard

Before collecting environmental samples, a working standard of 24 target compounds was prepared to conduct the basic QA/QC procedures. Liquid standards prepared at known analyte concentration were used for calibration to quantitate the target analytes (Table 1

). In the case of the 10 carbonyl compounds ((1) formaldehyde, (2) acetaldehyde, (3) propionaldehyde, (4) butyraldehyde, (5) isovaleraldehyde, (6) *n*-valeraldehyde, (7) acrolein, (8) acetone, (9) crotonaldehyde, and (10) benzaldehyde), HPLC/UV analysis was carried out using liquid-phase working standards prepared by diluting the TO11/IP-6A Aldehyde/Ketone-DNPH Mix (Supelco, USA) in a stepwise manner in acetonitrile. To obtain four-point calibration, the concentrations of the 10 carbonyl compounds were set in the range of $0.15-3.60 \text{ ng/}\mu\text{L}$. In the case of the other VOCs, a total of 14 compounds consisting of aromatic hydrocarbons, ketones, esters, an alcohol, and volatile fatty acids ((1) benzene, (2) toluene, (3) p-xylene, (4) m-xylene, (5) o-xylene, (6) styrene, (7) methyl ethyl ketone, (8) methyl isobutyl ketone, (9) butyl acetate, (10) isobutyl alcohol, (11) propanoic acid, (12) *n*-butyric acid, (13) *i*-valeric acid, and (14) *n*-valeric acid)) were selected. These VOCs were analyzed using thermal desorption (TD)–GC/MS. The standards were prepared by stepwise dilution of reagent grade chemicals (RGC) with purities of >95% (Sigma-Aldrich, USA) in methanol. The concentrations of the working standards for 14

compounds were 3.28-32.8 ng/ μ L (in the case of benzene) for a four-point TD-GC/MS calibration.

2.2. Collection of environmental samples from scented candles

To evaluate the emission characteristics of scented candles before and during combustion, a total of six candle samples (with one reference) were analyzed in this experiment. The scented candle type was identified by two letter codes: (1) clean cotton (CT), (2) floral (FL), (3) kiwi melon (KW), (4) strawberry (SB), (5) vanilla (VN), and (6) plain (PL: reference with no disclosed scent). In addition, we added the word off/on to each sample code to clearly indicate whether the candle had been lit (on) or not (off). In the preparatory stage of our study, the gross consumption rate (g/min) of each scented candle was calculated by taring the weight before and after a four minute lit period (Table 2). For the collection of the emission samples, an impinger system was used as shown in Fig. 1S. During experiments, the temperature and humidity in the laboratory were 25 °C and 40%, respectively. Each unused candle was lit soon after being placed inside the impinger. Pure air (used as the oxygen source), contained in a 20 L polyester aluminum (PEA) bag, was pulled through the impinger and supplied to the lit scented candle. This pure air was obtained from a purchased cylinder (Dongyang Industrial Gases Co., Ltd., Korea). In an ancillary experiment, we confirmed that the steady combustion was maintained at a pure air flow rate of 2 L.atm/min. A DNPH-cartridge was connected at the impinger exit to collect samples of carbonyl compounds for analysis at a flow rate of 2 L/min for 4 min. Concurrently, a three-bed sorbent tube packed with Tenax TA, Carbopack B, and Carbopack X (50 mg each) in a quartz tube was used to absorb the VOCs at a flow rate of 150 mL/min for 3 min.

2.3. Instrumental analysis setup

For the analysis of 10 carbonyl compounds in the working standard and the environmental samples, an HPLC/UV (Spectrasystem UV2000, Thermo scientific, USA) was installed with a C₁₈ column (5 μ m, 4.6 × 250 mm, ZORBAX Extend-C₁₈, Agilent, USA). A mixture of acetonitrile and distilled water (70:30 ratio), made as the mobile phase was flowed through the HPLC/UV at 1.5 mL/min for a total run time of 16 min; detection was by a UV detector at 360 nm (Table 1S).

The analysis of the 14 VOCs and reference compounds was done by thermal desorption (Unity 2, Markes International, Ltd., UK) and GC/MS (Shimadzu GCMS-QP2010, Japan). The cold trap (CT) in the TD unit was packed with Carbopack C and Carbopack B in a 1:1 volume ratio. Analytes in the ST were desorbed at 300 °C and transferred to the CT maintained at -10 °C. Then, the analytes in the CT were desorbed at 320 °C for 5 min to park the analytes on a CP-wax column (0.25 mm ID × 60 m L, 0.25 µm film thickness) for subsequent separation. The GC oven was initialized at 40 °C for 5 min and ramped to 220 °C at a 10 °C/min (Table 1S). In addition, for each sample, all compounds appearing in the chromatographic retention times between the hexane and hexadecane were summed and quantified as TVOC (ppbC) using the RF value of toluene.

2.4. WS calibration characteristics and quantification of the reference compounds

Prior to quantification of the target compounds from the scented candles, the basic calibration and QA conditions of our instrumental system were evaluated. The response factor (RF) values of the HPLC/UV system for all targets ranged from 6042 (BZA) to 16,732 (FA). The TD–GC/MS RF values ranged from 41,862 (PPA) to 324,630 (m-X). The R^2 values for the calibration linearity were satisfactory (>0.99) for all target compounds. The method detection limit

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