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Human health risk evaluation of selected VOC, SVOC and particulate emissions from scented candles

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ABSTRACT

Airborne compounds in the indoor environment arise from a wide variety of sources such as environmental tobacco smoke, heating and cooking, construction materials as well as outdoor sources. To understand the contribution of scented candles to the indoor load of airborne substances and particulate matter, candle emission testing was undertaken in environmentally controlled small and large emission chambers. Candle emission rates, calculated on the basis of measured chamber concentrations of volatile and semi-volatile organic compounds (VOC, SVOC) and particulate matter (PM), were used to predict their respective indoor air concentrations in a standard EU-based dwelling using 2 models: the widely accepted ConsExpo 1-box inhalation model and the recently developed RIFM 2-box indoor air dispersion model. The output from both models has been used to estimate more realistic consumer exposure concentrations of specific chemicals and PM in candle emissions. Potential consumer health risks associated with the candle emissions were characterized by comparing the exposure concentrations with existing indoor or ambient air quality guidelines or, where not existent, to established toxicity thresholds. On the basis of this investigation it was concluded that under normal conditions of use scented candles do not pose known health risks to the consumer.

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1. Introduction

People spent up to 90% of their time indoors (EU, 2005). Consequently, factors impacting the indoor air quality such as dampness, presence of molds and chemical exposures can have a considerable influence on human health. Concerns about possible health effects of indoor pollution with regard to carcinogenic and non-carcinogenic effects like asthma, allergies or non-specific symptoms from eyes, upper airways and facial skin are increasing. Such non-specific symptoms are occasionally referred to as 'sick building syndrome' (SBS) (Sahlberg, 2012). Most indoor pollutants consist of chemicals stemming from various sources including outdoor, indoor combus-

tion sources such as tobacco smoke, heating or cooking, and burning of candles. Other sources may include emissions from furniture and construction materials, the use of cleaning products, air fresheners, indoor pesticides, and other consumer products. In addition, microbiological contaminants which may induce allergies and asthma require careful consideration as potential indoor air pollutants (SCHER, 2007).

Although there is general acceptance that fragranced products can play an important role in enhancing the odor quality of the indoor environment, the increased use of scented candle use has also raised queries about the exposure to candle emissions and their possible impact on indoor air quality. Fragranced candles, however, are only one source of volatile organic compounds (VOCs), semi-volatile organic compounds (SVOCs) or particulate matter (PM) in the indoor environment. In fact VOCs, SVOCs and PM are ubiquitous in indoor air and their presence is due to a multitude of sources, both anthropogenic and natural (e.g., EU, 2005; HEI, 2005, 2007; SCHER,

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2007; Kotzias et al., 2009; Massolo et al., 2010; Geiss et al., 2011; Sarigiannis et al., 2011; Isaacs et al., 2013).

To date, few studies have been undertaken that investigated the candle emissions of both intentionally added fragrance materials as well as VOCs and/or SVOCs resulting from incomplete candle combustion (e.g., short chain aldehydes, benzene, polycyclic aromatic hydrocarbons (PAH), polychlorinated dibenzodioxins PCDD and polychlorinated dibenzofurans PCDF) (Schwind and Hosseinpour, 1994; Lau et al., 1997; Oekometric GmbH, 1997, 1999; VITO, 2006). Even fewer studies have examined the particulate matter (PM) emissions using various analytical techniques to determine different particulate size fractions (i.e., PM_{2.5}, PM₁₀) emitted by candles (Fine et al., 1999; Guo et al., 2000; Krause et al., 1999). However, as no validated standardized protocol for the measurement of candle emissions is available, the aforementioned analytical investigations differ in conditions and test set-up, making it difficult to compare and estimate consumer exposure on the basis of the presented data (Petry et al., 2013).

Only Lau et al. (1997), VITO (2006, 2008) and Petry et al. (2013) have attempted to estimate actual human exposure to candle emissions and characterize the associated human health risks. Using a simple inhalation uptake model, Lau et al. (1997) compared estimated human indoor exposures to formaldehyde, acetaldehyde, acrolein, benzo(a)pyrene and PCDD/PCDF to existing German Maximum Workplace Concentrations (MAK-value) and Technical Guidance Concentrations (TRK value). VITO (2006, 2008) estimated consumer indoor exposure to volatile short-chain aldehydes, benzene, naphthalene or xylenes, and particulate matter (i.e., PM_{2.5}, PM₁₀) released from unscented and scented candles using the modified CONTAM model. The CONTAM model takes into account chemical specific candle emission rates, indoor VOC deposition and adsorption rates, size and building characteristics, ventilation rates as well as actual consumer candle use practices and time-activity patterns. Modeled VOC and particulate exposures were compared to existing short term and long term indoor air quality guideline values. Petry et al. (2013) calculated consumer time-weighted average exposures to candle emissions of benzene and formaldehyde using the ConsExpo 1-box model² and compared those to existing indoor/outdoor guideline values. All three investigators concluded that on the basis of their investigations, emissions from candles are unlikely to pose long term health effects to people. VITO (2008) pointed out that short term peak exposures may trigger or aggravate symptoms in sensitive individuals.

Understanding the release and subsequent exposure to VOCs, SVOCs and PM emitted from scented candles is crucial to assessing potential consumer health risks. The overall objective of this paper is to present an approach in evaluating consumers' health risk from exposure to chemicals released from candles used in the indoor environment. Against the background of the afore mentioned obstacles of differing candle emission testing protocols and related uncertainties in interpreting the results, the aim of this investigation is to first report the results of a comprehensive set of studies conducted to measure the emissions of VOC, SVOC and PM from scented candles in a comparable manner under environmentally controlled conditions and thereby to substantially increase the understanding of candle emissions. Analytical data obtained in the form of chamber concentrations were back-calculated to candle emission rates on which basic consumer exposures were estimated for different candle use scenarios using a default 1-box as well as a newly developed higher tiered 2-box computational model. The health risks associated with consumer exposure estimates were assessed by comparison to existing or proposed regulatory air guideline values or, where not existent, to 'derived no effect

levels' (DNELs) for longer term exposure, as required under the European REACH Regulation (EC) No. 1907/2006.

2. Materials and methods

Volatile and semi-volatile organic compounds as well as particulate emissions of candles were investigated in a total of 3 independent studies. A first screening study, conducted at the laboratories of TNO Quality of Life, the Netherlands, aimed at establishing a candle emission testing protocol in a small emission test chamber with a volume of 2.2 m³ ("small chamber study"). Two subsequent studies were conducted at the laboratory of Air Quality Sciences (AQS), an ISO 9001:2000 registered Indoor Air Quality Laboratory, in normal room size emission chambers with a volume of approximately 26 m³. The first study at AQS ('LCS1') investigated the same compounds emitted from candles in the 'small chamber study' ('SCS') allowing a direct comparison of the results obtained. The second study at AQS ('LCS2') investigated different VOCs including semi-volatile PAH, PCDD and PCDF. The large chamber investigations also determined candle emissions of particulates characterized as PM₁₀ and PM_{2.5}. The contract laboratories were chosen on the basis of their substantial experience with the measurement of VOC, SVOC and PM in chamber emission studies using existing ISO- and ASTM standards (ISO, 2006, ASTM, 2001). The methodology to measure candle emissions largely followed the frame protocol described later by Petry et al. (2013) with regard to the experimental set-up, study protocol, meteorological conditions with regard to chamber temperature and relative humidity at candle ignition and analytical methods used to measure VOC emissions from candles according to existing ISO- and/or ASTM standards.

2.1. Determination of candle emissions: chamber emission studies

2.1.1. Small chamber study

2.1.1.1. Test products. Six fragranced jar candles (i.e., FC1, FC2, FC3, FC4, FC5, FC6), consisting of paraffin wax, a non-lead wick, as well as a fragrance mix, and one unfragranced (i.e., UFC1) jar candle were evaluated in the small chamber experiments. The individual total candle weight contained in the glass jar was approximately 113 g. In the case of the fragranced candles, the fragrance load was approximately 6% of total candle weight. The candles were received as packaged and shipped by the manufacturer. Upon receipt at the laboratories, the candles were visually inspected and stored in an environmentally controlled space, immediately following sample check-in until the moment of testing.

2.1.1.2. Study design. The VOC emissions from burning candles were investigated in two 2.2 m³ stainless steel chambers with steel framed glass doors. The chambers were process-controlled and equipped with a continuous data acquisition system that verified operating conditions of airflow, temperature and humidity. During each candle test, the air supply to the chamber was maintained at a temperature of 23 °C ± 2 °C and the relative humidity was kept at 50% ± 5%. The air exchange rate was set at 0.5 air changes/h.

The VOC background concentrations in the testing chambers were below the detection limit of total carbon using a flame ionization detector with a detection limit of 0.2 ppm based on propane. At study initiation, the candles were lit using a gas lighter outside and subsequently placed into the chamber. Candles were allowed to burn for 4–6 h, after which they were extinguished using a remotely controlled small jet of water, thereby allowing the chamber to remain closed throughout the whole experiment. While this procedure may have led to some dampening of the PM emissions, it was not expected to impact the VOC measurements. Particulate matter emissions were not investigated in the SCS study.

² <http://www.rivm.nl/en/Topics/Topics/C/ConsExpo>.

Air samples were taken at 2, 6, and 23 h after ignition in the chamber via the ingoing and outgoing air stream. At the 2 h sampling point, the candles were assumed to be at a steady emission rate and the VOC-chamber concentration in a build-up phase. The chamber concentration measured at the 6 h time point was expected to be close to or at the highest conceivable chamber concentration (see Fig. 2a). The 23 h sampling point served to verify that VOC values returned to background levels as a result of about 9–10 air exchanges in the chamber following extinction of the candle. After each experiment, the testing chambers were cleaned by flushing with HEPA filtered ambient air for at least 6 h. Before starting the next experiment, the test chambers were ventilated over night with humidified and purified ambient air to ensure 'normal' background levels. Fig. 1 presents a schematic diagram of the experimental set-up. Table 1 summarizes the measurements taken in both the small and large chamber studies.

2.1.1.3. Analytical measurements. Volatile organic compounds were measured in the outgoing airstream of the test chamber at various sampling time points throughout the experiment.

Samples were obtained by low volume air sampling through Tenax tubes (Tenax TA[®] 60/80, Sigma-Aldrich) at a flow rate of 0.1 L/min for 20 min to measure the aromatic VOCs and through silica-DNPH (2,4-dinitrophenylhydrazine) cartridges, prepared by the laboratory, at a flow rate of 1.5 L/min for 30 min for determining carbonyl compounds. Prior to each use, the Tenax tubes were conditioned for five minutes at 250 °C. The conditioned tubes were routinely analyzed by the contract laboratory to ensure low blank levels of the analytes under investigation. Although analysis revealed that the cleaned Tenax tubes could be stored for at least 7 days without altering the background levels of the analytes, cleaned tubes were re-used within a maximum of 2.5 days after cleaning.

The Tenax tubes, spiked with the internal standard dodecane (99% purity), were thermally desorbed using a Gerstel TDS unit.

The VOCs were intermediately trapped at –50 °C and analyzed on the basis of gas chromatography with mass spectrometric detection. The selected compounds were identified and quantified on the basis of total ion current recordings. The column used was a DB-5MS capillary column and the following GC-oven program was run: 0 °C (3 min), then 10 °C/min to 320 °C (5 min). The internal standard dodecane was solely used to demonstrate complete desorption of the VOCs from Tenax tubes. The peak areas of the selected compounds were not corrected with the internal standard. Quantification was conducted on the basis of the results obtained upon GC-MS analysis of air in a Tedlar bag filled with a known amount of air and spiked with known amounts of the reference substances (i.e., formaldehyde, acetaldehyde, benzene, naphthalene, toluene, styrene, o/m/p-xylene; Sigma-Aldrich) with a purity of ≥99%. The reference substances were brought into the Tedlar bag by injecting solutions of the substances. Subsequently, the air in the Tedlar bag was sampled with the same procedure as the sampling procedure for Hazleton chambers.

In the case of the silica-DNPH cartridges, the hydrazine derivatives were eluted with acetonitrile and analyzed by High Performance Liquid Chromatography (HPLC) with UV detection. The aldehyde-DNPH-derivative concentrations were measured by HPLC on a Lichrocart 125/4.0 mm, Lichrosphere 100 RP-18 column with UV detection at 365 nm. The eluent was an ammonium acetate solution in demi-water and tetrahydrofurane. The HPLC-UV system was calibrated with standard solutions of formaldehyde- and acetaldehyde-DNPH in acetonitrile. The purity of these standards was >99.9%. Before and after sampling cartridges were stored at a temperature of ≤18 °C.

2.1.2. Large chamber studies

2.1.2.1. Test products. Fragranced candles (FC1 and FC2) investigated in the small chamber studies were also analyzed for VOCs emissions in the first large chamber study ("LCS 1"), indicated as

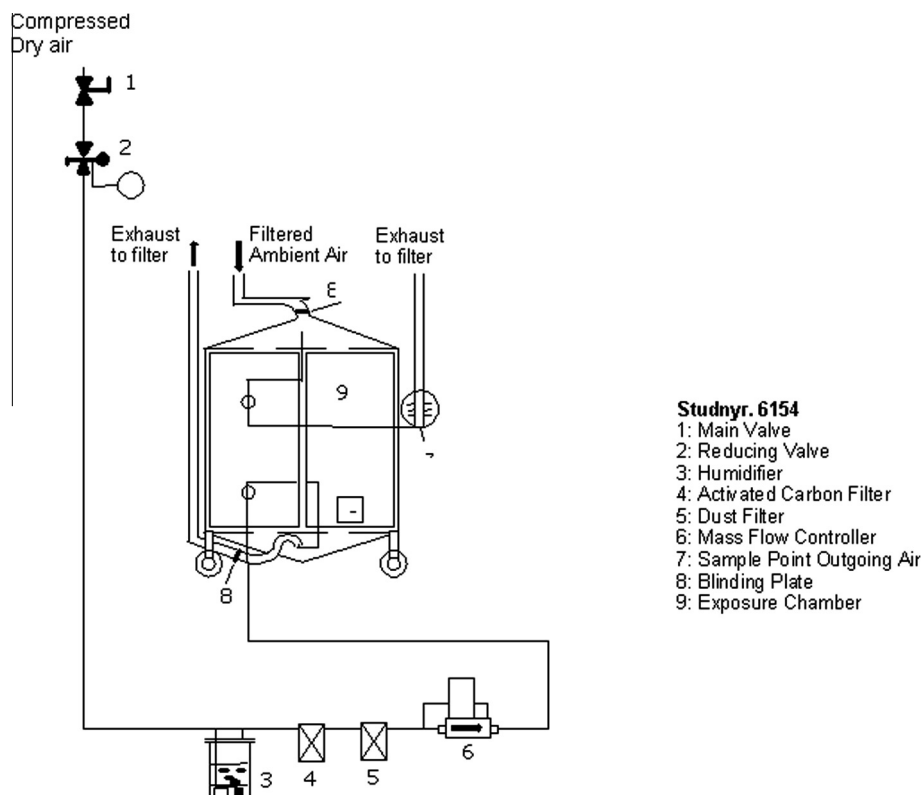


Fig. 1. Experimental set-up of the small chamber candle emission study as used by TNO Quality of Life.

Table 1
Air sample collection schedule and measurements taken in small chamber study (SCS) and large chamber studies 1 and 2 (LCS 1; LCS 2).

Study	Measured parameter	State of Combustion						
		Empty chamber (background)	Loaded chamber (background)	Burn		Post-burn		
				0–2 h	2–4 h	0–2 h	4–6 h	16–20 h
SCS	VOCs	✓	✓	✓	✓	✓	✓	✓
	Aldehydes	✓	✓	✓	✓	✓	✓	✓
	SVOC (PAH, PCDD/F)	-	-	-	-	-	-	-
	PM ₁₀	-	-	-	-	-	-	-
	PM _{2.5}	-	-	-	-	-	-	-
LCS 1	VOCs	✓	✓	✓	✓	✓	✓	✓
	Aldehydes	✓	✓	✓	✓	✓	✓	✓
	SVOC (PAH, PCDD/F)	-	-	-	-	-	-	-
	PM ₁₀	✓	✓	✓	✓	✓	✓	✓
	PM _{2.5}	✓	✓	✓	✓	✓	✓	✓
LCS 2	VOCs	✓	✓	✓	✓	✓	✓	-
	Aldehydes	✓	✓	✓	✓	✓	✓	-
	SVOC (PAH, PCDD/F)	✓	✓	✓	✓	✓	✓	-
	PM ₁₀	-	-	-	-	-	-	-
	PM _{2.5}	✓	✓	✓	✓	✓	✓	-

FC7 and FC8. The second large chamber study (“LCS 2”) examined the VOC, semi-volatile compounds and particulate emissions of one fragranced candle performed in triplicate (i.e., FC9) as well as the unfragranced base candle (i.e., UFC2).

The candles were made of paraffin wax, containing a non-lead cotton wick and a fragrance. The total candle weight for candles used FC1, FC2, FC7 and FC8 was approximately 113 g and that of FC9 and UFC2 400 g.

2.1.2.2. Study design. Burning candle emissions of VOC, SVOC and PM were measured according to the operating requirements of ASTM Standard D 6670 in an environmentally controlled emission chamber of approximately 26 m³. The chambers used were manufactured from stainless steel and aluminum with polished interior to minimize sink effects due to contaminant surface absorption. The chamber supply air was stripped of background formaldehyde, ozone, VOC’s PMs and other contaminants prior to study initiation by means of HEPA and charcoal filtration. The chambers were process-controlled and equipped with a continuous data acquisition system to monitor airflow, temperature and humidity. During the testing of each candle, air supply to the chamber was maintained at 23 ± 2 °C and relative humidity at 50 ± 5%. The air exchange rate was set at 0.5 air changes/hour in LCS 1 and 1.0 air changes/hour in LCS 2.

Prior to loading a candle in the testing chamber and starting the experiment, background chamber air was collected and analyzed for the target VOCs (LCS 1 and 2), particle concentrations (LCS 1 and 2), PAHs and PCDDs/PCDFs (LCS 2 only). After background sampling was conducted, the candle was placed inside the chamber at a central location on a stainless steel table approximately 76 cm off the floor. The candle in the chamber was allowed to equilibrate for a minimum of 4–8 h prior to ignition. After the equilibration period, chamber air was collected and analyzed for target compounds and particulates before the candle was ignited. To avoid any disturbances in the burning conditions, no mixing of air in addition to the normal air exchange was conducted. The equilibrium and measurement cycle was based on internal data and experience of the contract laboratory indicating that the concentration of emitted compounds in the air across the test chamber usually balances out after a time corresponding to 4–5 air exchange of the test chamber volume. A similar consideration is found in the forthcoming CEN construction product standard Fpr-CEN/TS 16516 which considers the requirement of 5 air exchanges to ensure proper mixing in the chamber.

The actual ignition of the candle was done by remote ignition without opening the chamber door. Following ignition, the candles were allowed to burn continuously for either 4 h (LCS 2) or 16 h (LCS 1). After the burning time, the candles were remotely extinguished with a puff of air. Candles remained in the chamber, following flame extinction, for an additional 1 (LCS 1) or 4 h (LCS 2) ‘post burn’ period, respectively, before being removed. Air samples were taken in the human breathing zone at a height of 1 m and within 1 m of the candle at time points 0, 1 and 4 h after candle ignition and 1 or 4 h after candle extinction. Given the vicinity of the sampling point to the candle it can be assumed that the measured chamber concentrations provide a conservative over-estimate of the actual chamber air concentration.

2.1.2.3. Analytical measurements. Air sampling was performed on sorbents followed by analysis of VOCs via thermal desorption and GC/MS or, in the case of carbonyl compounds (e.g., formaldehyde, acetaldehyde, acrolein) which were adsorbed on DNPH impregnated solid sorbent cartridges, followed by elution with acetonitrile and analysis by HPLC-UV. The method used for VOC determination was compliant with ASTM D 6169 (ASTM, 2001), with a limit of quantification of 1 µg/m³ and, for carbonyl compounds, with standard ASTM D5197 (ASTM, 2003) and a limit of quantification of 1–2 µg/m³.

Quality control data on TVOC measurements conducted over a period of 12 months at AQS laboratories which conducted LCS 1 and LCS 2 studies reveal an average precision of 6% RSD (relative standard deviation) for VOC and 4.5% RSD for aldehydes. However, a more specific method validation study was conducted as part of study LCS-2 for fragrance-related and target VOCs. In this context the analytical performance of specific fragrance compounds including vanillin, benyl benzoate, eugenol F, cinnamic aldehyde, linalyl acetate, methyl cinnamic aldehyde, coumarine, citral (pure) and damascene alpha was evaluated. The validation work included standard VOC analytes including acetaldehyde, acrolein, formaldehyde, benzene, styrene, toluene and xylenes. Sorbent tubes were prepared with known amounts of the target chemicals using a flash vaporization technique and then analyzed. For the aldehyde analyses, the corresponding DNPH derivative were directly injected into the HPLC and analyzed. Quantitation was performed using authentic multipoint calibration. Recoveries for each of the compounds were between 82% and 118% of the known amount. VOC detection limits ranged from 0.01 µg/m³ for toluene and xylenes to 1.3 µg/m³ for vanillin. Aldehyde detection limits were 0.5 µg/m³ or less as expected for this methodology.

Breakthrough studies for target chemicals were conducted in LCS 2 to ensure that all collected VOC was retained on the collection media at the flow rates used and at concentration levels expected. Flash vaporization was used to spike a Tenax sorbent tube (Tenax TA[®] 60/80, Sigma–Aldrich) with an authentic standard at the 1000 ng level. A second sorbent tube was connected in series with the first tube, and a total of 24 L of air pulled across both tubes at a rate of 200 mL/minute. Both tubes were then analyzed and the area response compared. There was no breakthrough observed for any of the target VOCs. Overall the analytical validation study indicated that analyses of the volatile target chemicals were conducted accurately and precisely within acceptable quality objectives.

Semi-volatile PAHs and PCDD/PCDF investigated in LCS 2 were collected in accordance to US EPA methods TO-9A (US EPA, 1999a) and TO-13A (US EPA, 1999b) in an adsorbent cartridge assembly containing two polyurethane foam (PUF) plugs, with an internal layer of polymeric XAD resin in between, as supplied by a specialized PAH and PCDD/PCDF testing laboratory (i.e., Alta Analytical Laboratory, Inc., El Dorado Hills, CA). This assembly allows for the collection of PAHs/PCDD/PCDF in both the particulate as well as in the gas phase. In line with methods TO-9A and TO-13A, the PUF plugs as well as the XAD resin were cleaned by Soxhlet extraction using appropriate solvents followed by vacuum and room drying prior to use. Alta Analytical Laboratories also performed the analytical work using high resolution gas chromatography mass spectrometry (HRMS), following CARB Method 429 for the identification and quantification of PAHs (CARB, 1989), and CARB Method 428 for the identification and quantification of PCDD and PCDF (CARB, 1988). To improve the detection limit, PUF plugs and XAD resin were combined prior to extraction and analyses. Based on the air sample collection volumes utilized, the quantification limits of the methods were 8 ng/m³ for PAHs and 1 pg/m³ for PCDD/PCDF. Depending on congeners the overall collection and analytical recover efficiency of the method ranged from 85% to 110% for PAHs and 90% to 110% for PCDD/PCDF when corrected for pre-spiked surrogate standards. Isotopically-labeled standards were added to the PUF plugs prior to field sampling and account for any loss occurring during sample collection, shipment and analysis.

Continuous particle concentration monitoring was performed using TSI Model 8520 Aerosol Monitors. The analytical range of this instrument is 0.001 to 100 mg/m³, with the measurement of particles ranging from 0.1 to 10 µm in size. While there are some limitations of real-time measurements over gravimetric methods, gravimetric analyses, as typically recommended for the work place by NIOSH or ACGIH, were too insensitive under the experimental set-up. Two instruments were set up for the collection of particles less than 2.5 µm (PM_{2.5}) and two instruments were arranged for the collection of particles less than 10 µm (PM₁₀). A calibration factor for fly ash was determined and applied subsequent to the original data collection to closely align measurements with a presumably similar combustion particle mix.

Particle counts were measured using Pacific Scientific Met One 2100 Particle Counters. Particle counts were provided for the following size channels: 0.1, 0.2, 0.3, 0.5, 0.7 and 1.0 µm. Ultrafine particle counts were measured using a TSI P-track Ultrafine Particle Counter Model 8525. The minimum particle size measurable by this instrument is 20 nm with an overall particle range of 20 to 100 nm.

2.2. Estimating human indoor exposures to candle emissions

Two models have been chosen to estimate human indoor exposures to candle emissions on the basis of the candle emission rates obtained from the environmental chamber studies. These are the ConsExpo 1-box model (RIVM, 2007) as well as the 2-Box Indoor

Air Dispersion Model developed by the Research Institute for Fragrance Materials (2010).

The ConsExpo ('exposure to vapors – constant release') 1-box model² was adopted to conservatively estimate consumer exposure concentrations to VOCs, SVOCs and particulates released from candles at maximum emission rates. The model assumes that the removal of volatile compounds and particles from the indoor environment is solely a function of the air exchange. Although removal of VOCs, SVOCs or particulates due to adsorption or deposition ('sinks') is known to readily occur in common living spaces on carpets, walls or other household items, with environmental tobacco smoke as the prime example (e.g., EU, 2005; SCHER, 2007; VITO, 2008), such removal is not considered in the ConsExpo model increasing the conservatism of the exposure estimates.

In addition to running the ConsExpo model, a refinement was adopted by modeling the highest measured emission rates in RIFM's 2-Box Indoor Air Dispersion Model. This model is a slightly simplified version of the US EPA's Multi Chamber Concentration and Exposure Model (US EPA, 2001). This indoor exposure calculation model is considered to provide a more realistic view on potential human inhalation exposure concentrations to chemicals released into indoor air from consumer products. The 2-box model incorporates a more complex understanding of air flow/ventilation within an indoor environment, modeling the dispersion of VOCs, SVOCs or particulates between two inter-connected, enclosed zones, such as the room of interest and the rest of the house. In this model, the emission source (i.e., the candle) is located, for example, in a living room with a defined volume (Zone 1) and air exchange is modeled between this room and Zone 2 (i.e., the rest of the residence). The model incorporates the basic assumptions that concentrations in Zones 1 and 2 disperse in a homogenous fashion and that all chemical losses can be treated through a single degradation term. The term, which is assumed to follow a first order loss kinetic, is represented by a degradation half-life. This model also considers human activity patterns in the house throughout the day, and uses these data along with airborne concentration to derive human exposures. Like the ConsExpo model, the RIFM's 2-box model does not consider sink effects that are likely to occur in common living spaces.

Both models have been used to determine peak and time-weighted average consumer exposure concentrations at maximum emission rates during, and following, the use of the candles. Due to the infrequent usage of candles (i.e., 8.3 times/month), the short burning time (i.e., 1.9–2.4 h/day) (IPSOS, 2006; VITO, 2006) and the rapid decline of the airborne pollutant concentration, no accumulation of pollutants needs to be considered.

Table 2 presents the default parameters for room volumes, burn duration, residency time and air flow rates that have been used for calculating consumer exposure to candle emissions.

2.2.1. Candle or indoor factors impacting human exposures to candle emissions

The ConsExpo 1-box and RIFM 2-box models both require certain input parameters and consideration of building characteristics to calculate human indoor exposures to candle emissions on the basis of candle emissions measurements in environmental test chambers.

To calculate the candle emissions in a standard indoor room based on the concentrations determined in experimental chamber studies, the candle is assumed to be a point source emitting VOC, SVOC and particulate matter at a constant emission rate. Under normal candle burning conditions, the concentration of the desired fragrance, or the pollutant in the case of combustion product, reaches an equilibrium that depends only on the candle's specific fragrance or pollutant emission rate and the ventilation rate. The

Table 2
Default parameters for calculation of consumer exposures to candle emissions depending on candle use location.

Scenario	Living room		Kitchen and dining		Bedroom		Bathroom/toilet	
	1-Box model	2-Box model	1-Box model	2-Box model	1-Box model	2-Box model	1-Box model	2-Box model
Candle burning time in room (h)	4	4	4	4	4	4	4	4
Volume: Room (m ³)	58.00	58.00	15.00	15.00	27.00	27.00	10.00	10.00
Volume: Rest of house (m ³)	N/A	140.50	N/A	183.50	N/A	171.50	N/A	188.50
Air exchange rate: Room (h ⁻¹)	0.50	N/A	2.50	N/A	1.00	N/A	2.00	N/A
Air exchange rate: House (h ⁻¹)	N/A	0.60	N/A	0.60	NR	0.60	N/A	0.60
Air flow: Room → Outdoor (m ³ /min)	N/A	0.58	N/A	0.15	N/A	0.06	N/A	0.10
Air flow: House → Outdoor (m ³ /min)	N/A	1.41	N/A	1.84	N/A	1.72	N/A	1.89
Air flow: Room → House (m ³ /min)	N/A	0.93	N/A	0.93	N/A	0.39	N/A	0.87

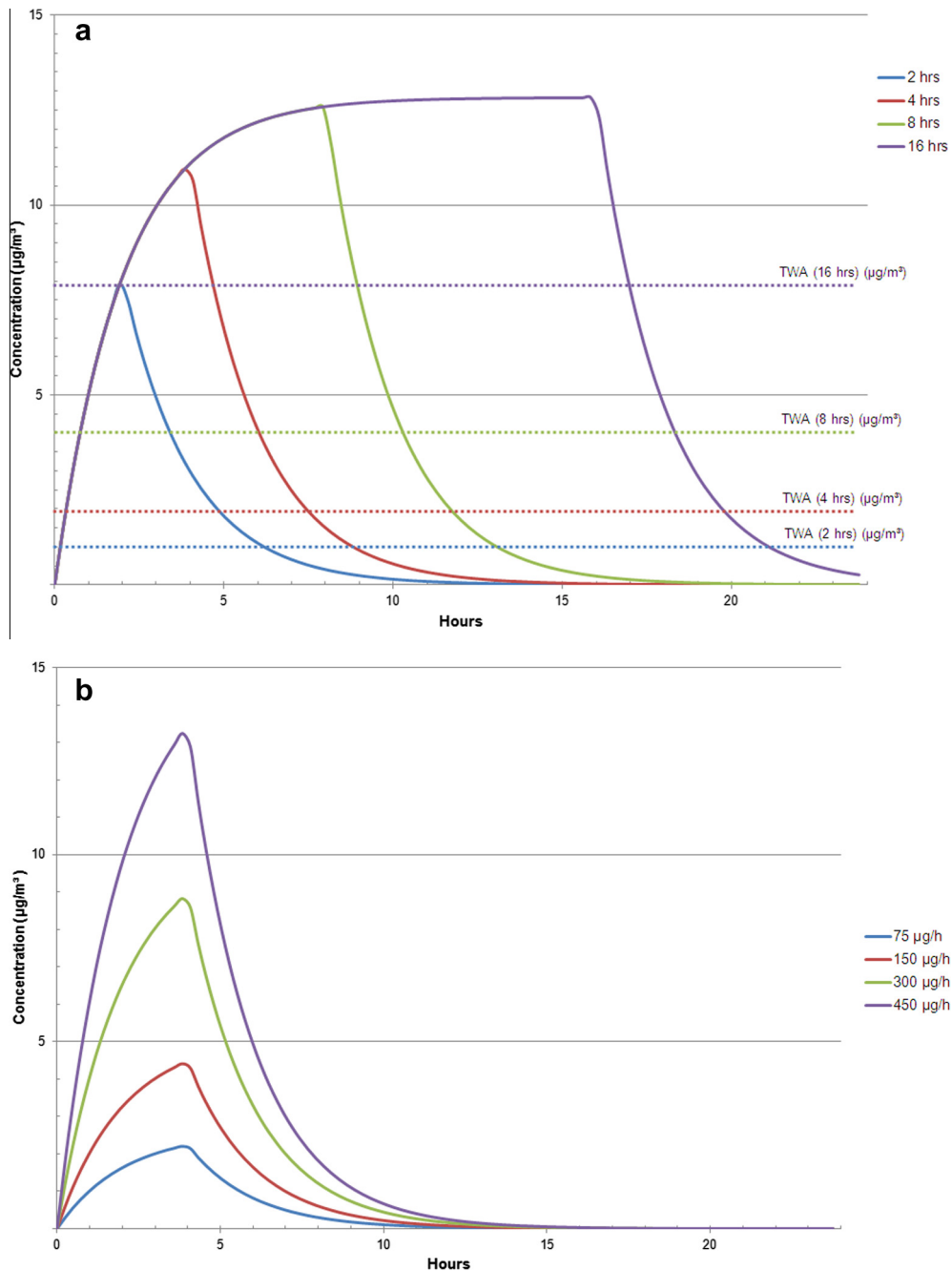


Fig. 2. (a) Candle emission profiles modeled at different burn times and constant emission rate (ER = 372 µg/h). (b) Candle emission profiles modeled at different emission rates and constant burn time (4 h).

room volume and the ventilation rate determine the time it takes to reach equilibrium. This relationship is described in Eq. (1).

$$\text{Air concentration} = \text{BG} + \text{ER} \times (1 - e^{-T \times \text{VR}}) / (\text{RV} \times \text{VR}) \quad (1)$$

BG = Background ($\mu\text{g}/\text{m}^3$); ER = emission rate ($\mu\text{g}/\text{h}$); T = Time (h); RV = room volume (m^3); VR = ventilation rate (h^{-1}); AC = Air concentration ($\mu\text{g}/\text{m}^3$).

Accordingly, the emission rate is the scaling factor that can be used to calculate the equilibrium air concentrations in different room sizes at different air exchange rates. The emission rate can be calculated according to the Eq. (2):

$$\text{Emission Rate} = (\text{AC} - \text{BG}) / (1 - e^{-T \times \text{VR}}) / (\text{RV} \times \text{VR}) \quad (2)$$

AC = Air concentration ($\mu\text{g}/\text{m}^3$); BG = background ($\mu\text{g}/\text{m}^3$); ER = emission rate ($\mu\text{g}/\text{h}$); T = Time (h); RV = room volume (m^3); VR = ventilation rate (h^{-1}).

In cases where the emission rate is constant, an asymptotic increase of the concentration can be seen until equilibrium is theoretically reached. Therefore, calculating an equilibrium concentration at the highest measured emission rate represents the worst case scenario for the exposure assessment. In reality, indoor air concentrations in homes are however lower as a result of sink effects or higher air exchange rates. Fig. 2a and b illustrate the impact of varying burn time or emission rates on peak exposures as well as the 24 h time-weighted average (TWA) concentrations at constant room volume, and air exchange. Both figures have been modeled using the ConsExpo 1-box model.

As can be seen in Eqs. (1) and (2), the key variables determining the inhalable air concentration to candle emissions are room volume and ventilation rate. Average room sizes, building characteristics, as well as ventilation rates, vary significantly from region to region and country to country.

The European Chemicals Agency's (ECHA) technical guidance documents for consumer exposure assessments lists standard room volumes to be used for exposure and risk assessment purposes indicating, for example, median room volumes in the Netherlands of 10 m^3 for a bathroom and 58 m^3 for a living room. In Germany, median room volumes are reported to be 64 m^3 . The ECHA technical guidance documents for consumer exposure assessments suggest ventilation rates to be 0.5–2.5 per hour (ECHA, 2012a). Air exchange rates and room sizes from RIVM's Technical Guidance Document were used to model exposure in the one-box model.

Likewise, depending on the chemical nature of the fragrance ingredient or the pollutant, the building materials and the presence of furniture in rooms/dwellings, there is also a great variation of VOC adsorption and particle deposition rates. VITO (2008) reviewed studies available on the adsorption of VOCs on surfaces, and deposition rates of particles, as well as consideration of these

measurements in indoor air exposure assessments. On the basis of this review, and for the purpose of this human health assessment, the conservative assumption was made that neither VOC/SVOC adsorption, nor particle deposition, occurs in addition to that occurring in the environmental chambers. Particle deposition rates largely depend on the particle size distribution and the ventilation rate in the room. This assumption increases the conservatism of the consumer exposure estimates.

2.2.2. Consumers candle use practices

Consumer exposure to candle emissions is determined by the candle emission itself and consumer candle use patterns. A significant market research investigation on consumer air freshener shopping and use behavior was commissioned by the Belgian Ministry of Health to have a better basis for conducting consumer exposure assessments to chemicals released from air freshener including scented candles. This research was conducted by the market research firm IPSOS and completed in 2006 (IPSOS, 2006). Under the European Union Health Programme 2008–2013, the European Commission launched the EPHECT project which is a European collaborative action focusing on surveying use as well as identifying and assessing risks associated with indoor air pollutants emitted by a selection of consumer products including scented candles (VITO, 2006; EU, 2011). This project is expected to be completed in 2013, but results of this survey were not yet publicly available at the time of this publication.

The IPSOS consumer research was performed via the computer assisted personal interview (CAPI) method on the basis of 646 interviews of randomly selected and representative individuals living in Belgium with purchasing responsibility in their families. Specifically with regard to scented candles, the following consumers' habits and practices were identified by IPSOS (2006):

- 92% of the consumers use scented candles in the living room, 9% in the kitchen and dining area and 6% in adults' bedroom.
- On average scented candles are used 8.3 times/month in the living room, 9.8 times/month in the kitchen and dining area and 7.1 times/month in adults' bedroom.
- The average use of scented candles is 2.4 h in the living room, 2 h in the adult bedroom and 1.9 h in the kitchen and dining area.
- In terms of 'intensity' [defined as (use) \times (frequency)], 88% of the purchased scented candles are used in the living room, 7% in the kitchen and dining area and 5% in adult bedrooms.

3. Results

3.1. Chamber emission studies

Tables 3–6 summarize the results obtained in the small and large chamber emission studies. The tables present the maximum

Table 3
Measured maximum chamber concentrations and calculated emission rates of volatile organic compounds.

Study	Candle code	Formaldehyde		Acetaldehyde		Benzene		Naphthalene		Toluene		Styrene		Total xylenes	
		$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$
SCS	FC1	121.20	137.90	34.75	41.20	18.62	21.10	6.68	7.70	8.88	11.40	17.85	20.70	8.80	10.20
	FC2	136.98	235.30	34.81	58.60	13.52	15.50	4.48	5.20	9.24	14.60	16.21	18.80	10.26	14.50
	FC3	59.49	73.00	12.93	13.60	2.66	2.70	2.89	0.50	2.37	2.60	1.55	1.80	2.89	3.30
	FC4	221.72	283.90	47.48	62.80	1.58	1.80	0.99	1.70	3.79	6.50	1.51	1.80	1.37	2.40
	FC5	323.50	372.20	74.95	85.70	28.49	32.60	1.60	1.90	14.15	16.30	69.63	80.60	6.68	7.70
	FC6	275.27	316.50	73.66	84.00	27.30	29.70	3.75	2.80	5.65	7.70	12.51	16.20	12.58	19.20
	UFC1	18.58	19.60	2.61	3.00	0.88	0.90	0.10	0.20	0.28	0.30	0.09	0.20	0.31	0.50
LCS1	FC7	18.20	234.00	8.20	105.00	2.30	29.60	4.40	56.50	3.30	42.40	3.40	43.70	2.30	29.60
	FC8	22.50	289.00	13.00	167.00	1.40	18.00	0.40	5.10	1.00	12.90	2.00	25.70	0.50	6.40
LCS2	FC9	10.90	280.00	2.40	61.70	2.80	72.00	0.13	3.24	3.30	84.80	2.20	56.50	<1.00	<25.70
	UFC2	<1.00	<25.70	<1.00	<25.70	<1.00	<25.70	0.03	0.72	<1.00	<25.70	<1.00	<25.70	<1.00	<25.70

Table 4
Measured maximum chamber concentrations and calculated emission rates of volatile fragrance materials.

Study	Candle Code	Cinnamic aldehyde		Citral Pure		Coumarin		Eugenol F		Limonene		Lilial		Linalool		Linalyl acetate	
		$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$
LCS1	FC7	–	–	–	–	–	–	–	–	9.5	122.0	33.6	432.0	137	1760.0	–	–
	FC8	–	–	–	–	–	–	–	–	3.5	45.0	<0.1	<1.3	11.4	146	–	–
LCS2	FC9	12.4	319	22.6	581	3.0	77.1	18.2	468	–	–	–	–	–	–	3.9	100
	UFC2	<1.0	<25.7	<1.0	<25.7	<1.0	<25.7	<1.0	<25.7	–	–	–	–	–	–	<1.0	<25.7

Table 5
Measured maximum chamber concentrations and calculated emission rates of polycyclic aromatic hydrocarbons (PAH) and polychlorinated dibenzodioxins/furans (PCDD/PCDF).

Study	Candle code	Benzo(a) anthracene		Benzo(k,f) fluoranthene		Benzo(a) pyrene		Dibenz(a,h) anthracene		Indeno(1,2,3-cd)pyrene		Benzo(g,h,i) perylene		2,3,7,8-TCDD (TEF = 1)		1,2,3,7,8-PeCDD (TEF = 1)		2,3,4,7,8-PeCDF (TEF = 0.3)	
		$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	pg/m ³	pg/h	pg/m ³	pg/h	pg/m ³	pg/h
LCS2	FC9	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<1.43	<36.80	<0.96	<24.70	<1.22	<31.40
	UFC2	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<0.01	<0.21	<1.36	<35.00	<0.87	<22.40	<1.11	<28.50

Table 6
Measured burn and post-burn particulate chamber concentrations and calculated emission rates.

Study	Candle code	Burn				Post-burn			
		PM _{2.5}		PM ₁₀		PM _{2.5}		PM ₁₀	
		$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$	$\mu\text{g}/\text{m}^3$	$\mu\text{g}/\text{h}$
LCS1	FC7	1	13	4	51	844	10,800	1060	13,600
	FC8	1	13	6	77	267	3430	350	4500
LCS2	FC9	8	206	–	–	17	437	–	–
	UFC2	9	231	–	–	55	1410	–	–

measured chamber concentrations and emission rates of non-fragrance VOCs (Table 3), volatile fragrance compounds (Table 4), semi-volatile PAHs and PCDD/PCDF (Table 5) and particulate matter (Table 6). Candle emissions of volatile fragrance compounds, SVOCs and particulate matter were only investigated in the large chamber studies.

While VOC and fragrance-VOC emissions from scented candles were clearly measurable and discernible from the chamber background concentrations, semi-volatile PAHs and PCDD/PCDF emissions were below the detection limit (i.e., <0.01 $\mu\text{g}/\text{m}^3$ for PAHs; <1.5 pg/m^3 for PCDD/PCDF) and therefore not identifiable. The particulate matter concentrations and emission rates for PM_{2.5} and PM₁₀ are presented for the burn and the post-burn phase, taking into account that, following extinction of the candle, there is a sharp increase in PM-emissions by the candle.

3.2. Estimation of room-specific human indoor air exposure concentrations

Peak as well as 24 h time-weighted average human indoor air exposure concentrations have been calculated for each measured emission at highest emission rate using the conservative ConsExpo 1-box model as well as the more refined RIFM 2-Box indoor air dispersion model. The results obtained for volatile and semi-volatile as well as for particulate matters are presented in Table 7.

4. Discussion

In this investigation, the emissions of VOC, SVOC and/or PM of a total of nine scented candles were investigated in 3 independent studies in external quality-controlled contract research laboratories. The methodology to measure candle emissions largely

followed the frame protocol described later by Petry et al. (2013) with regard to the experimental set-up, study protocol, meteorological conditions and analytical methods used to measure VOC emissions from candles according to existing ISO- and/or ASTM standards.

Similar to other candle emission studies published previously (e.g., Vito, 2006), some uncertainties relate to the lack of monitoring of possible changes of chamber temperature, relative humidity, surface velocity and the degree of oxygen depletion during the experiments. These factors have been identified to impact the quality of the candle burn and subsequently the candle emission pattern (Petry et al., 2013). On the basis of their investigations on formaldehyde and benzene emissions from scented candles in environmental test chambers, Petry et al. (2013) obtained reproducible results when the chamber temperature and relative humidity (RH) did not exceed 30° and 75% RH throughout the experiments. Likewise, the oxygen concentration should not decrease by more than 2% throughout the experiment. Moreover, Petry et al. (2013) considered it important to include a reference candle group outside the chamber which should be burned in parallel to those inside the chamber to monitor the chamber candle's burn rate. A minimal difference in burn rates of $\leq 15\%$ between inside and outside the testing chamber is considered to reflect stable burn conditions within the chamber. Despite the limitations of not fully controlling these parameters during the experiments underlying this investigation, the emission rates that have taken forward to model consumer exposures are considered suitable for health risk evaluation purposes as they are higher, hence more conservative than those that can be derived from other comparable candle emission investigations (Vito, 2006, 2008; Petry et al., 2013).

Consumer risks associated with the inhalation exposure to candle emissions can be characterized by comparing estimated human

Table 7
Estimated highest peak and 24 h TWA concentrations using the ConsExpo 1-box as well as the RIFM 2-box model.

	Living room				Kitchen and dining				Bedroom				Bathroom/toilet			
	1-Box model ($\mu\text{g}/\text{m}^3$)		2-Box model ($\mu\text{g}/\text{m}^3$)		1-Box model ($\mu\text{g}/\text{m}^3$)		2-Box model ($\mu\text{g}/\text{m}^3$)		1-Box model ($\mu\text{g}/\text{m}^3$)		2-Box model ($\mu\text{g}/\text{m}^3$)		1-Box model ($\mu\text{g}/\text{m}^3$)		2-Box model ($\mu\text{g}/\text{m}^3$)	
	Peak	TWA	Peak	TWA	Peak	TWA	Peak	TWA	Peak	TWA	Peak	TWA	Peak	TWA	Peak	TWA
<i>Volatile combustion products</i>																
Formaldehyde	10.94	2.14	5.16	0.84	42.43	8.19	7.80	1.30	13.47	2.31	2.40	2.14	63.85	12.28	29.41	1.44
Acetaldehyde	4.91	0.96	2.32	0.38	19.04	3.67	3.50	0.58	6.05	1.04	1.07	0.96	28.56	5.51	13.20	0.65
Benzene	2.12	0.41	1.00	0.16	8.21	1.58	1.51	0.25	2.61	0.45	0.21	0.19	12.31	2.38	5.69	0.28
Naphthalene	1.66	0.32	0.04	0.01	6.44	1.24	1.18	0.20	2.05	0.35	0.46	0.41	9.66	1.86	0.25	0.22
Toluene	2.49	0.49	1.18	0.19	9.67	1.87	1.78	0.30	3.07	0.53	0.02	0.02	14.50	2.80	6.70	0.33
Styrene	2.37	0.46	1.12	0.18	9.19	1.77	1.69	0.28	2.92	0.50	0.55	0.49	13.78	2.66	6.37	0.28
Total xylene	0.87	0.17	0.41	0.07	3.37	0.65	0.41	0.07	1.07	0.18	0.52	0.46	5.06	0.98	2.34	0.11
<i>Volatile fragrance compounds</i>																
Cinnamic aldehyde	9.38	1.83	4.42	0.72	36.37	7.02	6.69	1.11	11.55	1.98	13.01	1.84	54.55	10.53	7.07	1.19
Citral Pure	317.10	3.34	8.06	1.30	66.23	12.78	12.18	2.02	21.03	3.60	23.69	3.35	99.35	19.17	12.88	2.16
Coumarine	2.27	0.44	1.07	0.17	8.79	1.70	1.62	0.27	2.79	0.48	3.14	0.44	13.18	2.54	1.71	0.29
Eugenol F	13.76	2.69	6.49	1.05	53.35	10.30	9.81	1.63	16.94	2.90	19.08	2.69	80.03	15.44	10.38	1.74
Limonene	3.59	0.70	1.69	0.27	13.91	2.68	2.56	0.43	4.42	0.76	4.97	0.70	20.86	4.03	2.71	0.45
Lilial	12.70	2.48	5.99	0.97	49.25	9.50	9.06	1.51	15.64	2.68	17.61	2.49	73.87	14.26	9.58	1.61
Linalool	51.74	10.12	24.40	3.95	200.60	38.72	36.89	6.13	63.71	10.91	71.75	10.13	300.90	58.08	39.02	6.54
Linalyl acetate	2.94	0.58	1.39	0.22	11.40	2.20	2.10	0.35	3.62	0.62	4.08	0.58	17.10	3.30	2.00	0.37
<i>Semi-volatile organic compounds</i>																
PAH	–	$<0.10 \times 10^{-2}$	–	$<0.03 \times 10^{-2}$	–	$<0.40 \times 10^{-2}$	–	$<0.05 \times 10^{-2}$	–	$<0.13 \times 10^{-2}$	–	$<0.12 \times 10^{-2}$	–	$<0.70 \times 10^{-2}$	–	$<0.05 \times 10^{-2}$
PCDD/PCDF	–	$<0.20 \times 10^{-6}$	–	$<5.30 \times 10^{-8}$	–	$<0.80 \times 10^{-6}$	–	$<7.90 \times 10^{-8}$	–	$<1.70 \times 10^{-6}$	–	$<21.00 \times 10^{-8}$	–	$<1.20 \times 10^{-6}$	–	$<8.70 \times 10^{-8}$
<i>Particulate matters</i>																
PM _{2.5}	–	15.34	–	5.48	–	60.46	–	6.23	–	17.16	–	9.73	–	90.24	–	6.36
PM ₁₀	–	19.44	–	0.17	–	76.58	–	7.21	–	21.77	–	11.19	–	114.27	–	7.29

Table 8
Existing or proposed indoor air quality guidelines and typical indoor air concentrations in the European Union as summarized by EU (2005) or WHO (2010).

	WHO indoor air (WHO, 2010) ($\mu\text{g}/\text{m}^3$)	EU indoor air (EU, 2005) ($\mu\text{g}/\text{m}^3$)	US EPA ^a ($\mu\text{g}/\text{m}^3$)	Typical EU indoor air concentrations (EU, 2005; WHO, 2010) ($\mu\text{g}/\text{m}^3$)
<i>Non-fragrance VOC</i>				
Formaldehyde	100	30	N/A	33–79
Acetaldehyde	N/A	200	9 ^g	10
Benzene ^b	0.17	5 ^e	30 ^h	2–13
Naphthalene	10	10	3 ⁱ	1–3
Toluene	N/A	300	5000 ^j	20–74
Styrene	N/A	250	1000 ^k	1–6
Xylenes	N/A	200	100 ^l	8–37
<i>Semi-VOC</i>				
Total PAH	12×10^{-6} (BaP) ^c	N/A	N/A	N/P
Benzo(a)pyrene	12×10^{-6c}	N/A	N/A	<0.001
PCDD/PCDF	N/A	N/A	N/A	N/P
<i>Particulate matters</i> (no indoor air guidelines available; ambient air guidelines temporarily considered to be used as exposure reference)				
PM _{2.5}	10 ^d (WHO, 2005)	20 (EU, 2008)	12 ^d (US EPA, 2013)	N/P
PM ₁₀	20 ^d (WHO, 2005)	50 ^f (EU, 2008)	150 ^f (US EPA, 2013)	N/P

N/A = not available; N/P = not provided.

^a No indoor air guideline values have been published by the US EPA; instead RfC values as published by the US EPA and ACGIH are shown for comparison.

^b Benzene is generally considered a genotoxic carcinogen for which a safe exposure threshold cannot be established. Lifetime exposure to the presented concentration is estimated to produce an excess lifetime cancer risk of 1/1,000,000.

^c Mixtures of polycyclic aromatic hydrocarbons (PAH) are generally considered genotoxic carcinogens. Lifetime exposure to the PAH represented by its marker compound Benzo(a)pyrene (BaP) represents the carcinogenic activity of the PAH mixture to which coke oven workers have been exposed to and is estimated to produce an excess lifetime cancer risk of 1/1,000,000.

^d Annual guideline value (arithmetic mean).

^e No specific indoor air guideline value proposed for benzene in the EU Index report. The EU Index report presents the EU ambient air guideline limit for benzene (EU, 2008) for orientation.

^f 24-h guideline value.

^g US EPA (1991).

^h US EPA (2003).

ⁱ US EPA (1998).

^j US EPA (2005).

^k US EPA (1993).

^l US EPA (1987).

exposures to air quality guideline values as published or proposed by national or international regulatory bodies. For emitted chemicals for which such air guideline values do not exist, like for most fragrance materials, consumer risks can be assessed by comparing the toxicity thresholds of chemical substances as determined in suitable toxicology studies to estimated exposures.

The results of the standardized exposure scenarios using the ConsExpo 1-box and the RIFM 2-box model are presented in Table 7. While VOC and PM emissions from scented candles were clearly measurable and discernible from the chamber background concentrations, semi-volatile PAHs and PCDD/PCDF emissions were below the detection limit and therefore not identified. Depending on room of candle use, the estimated peak or 24 h time-weighted average (TWA) consumer exposure concentration can vary by a factor of up to 6 when using the 1-box model and 3 when using the more dynamic 2-box model. For example, when burning a candle for four hours, the estimated TWA concentration for formaldehyde ranged from 2.14 $\mu\text{g}/\text{m}^3$ in the living room to 12.28 $\mu\text{g}/\text{m}^3$ in the bathroom according to the 1-box model and from 0.84 $\mu\text{g}/\text{m}^3$ in the living room to 2.14 $\mu\text{g}/\text{m}^3$ in the bedroom according to the 2-box model. The respective short-term peak values are from 10.9 $\mu\text{g}/\text{m}^3$ in the living room to 63.9 $\mu\text{g}/\text{m}^3$ in the bathroom according to the 1-box model and from 5.2 $\mu\text{g}/\text{m}^3$ in the living room to 29.4 $\mu\text{g}/\text{m}^3$ in the bedroom according to the 2-box model.

Table 8 presents existing (i.e., WHO) or proposed (i.e., EU index values) indoor air quality guideline values as well as VOC and PM reference concentrations (RfC) published by the US EPA. Moreover, to put any estimated consumer exposures to candle emissions into perspective, Table 8 further lists typical air concentrations that have been reported for the EU in the EU Index Report (EU, 2005)

and the WHO Guidelines for Indoor Air (2010) for indoor environments.

Comparing the modeled short-term peak and TWA human exposure concentrations for the volatile non-fragrance combustion products (e.g., benzene, formaldehyde, styrene) to the identified regulatory guideline levels (Table 8) reveals that even under worst case conditions (i.e., 1-box model – bathroom/toilet scenario), the estimated TWA exposure concentration is at least a factor of 2 (i.e., benzene) below the regulatory guidance level which can be considered an acceptable exposure level for chronic exposure. For the same scenario, the more realistic 2-box model indicates that the estimated consumer TWA exposure to benzene (i.e., 0.28 $\mu\text{g}/\text{m}^3$) is approximately 18 times below the air quality guideline value which has been established for lifetime exposure to benzene (i.e., 5 $\mu\text{g}/\text{m}^3$). For perspective, typical residency time in a bathroom/toilet is estimated to be at maximum 30–60 min per day and market research revealed that candles are only used on average 7–9 times per month in indoor premises. But nevertheless, even under the assumption of a lifetime exposure to candle emissions of the volatile non-fragrance combustion products are not assessed to pose a hazard to consumers.

Regulatory air guideline values are not available for the volatile fragrance materials considered in this investigation. Therefore, to establish safe consumer exposure levels for each fragrance material, a REACH compliant derived no effects level (DNEL) for long term exposure was determined. This was done by applying the assessment factors recommended by the European Chemicals Agency (ECHA, 2012a) for the purpose of the REACH Regulation to the substances no observed adverse effect level (NOAEL) or concentration (NOAEC) as identified in inhalation or oral toxicology studies. In the cases where suitable inhalation toxicology studies

Table 9
Consumers safe exposure levels ('DNELs') of fragrance ingredients.

	Study type	NOAEL (mg/kg/d)	NOAEC (mg/m ³)	NOAEC Starting Point (mg/m ³)	Assessment factor ^c	DNEL (mg/m ³)
Cinnamic Aldehyde (US NTP, 2004)	Chronic oral	200	–	86 ^a	25	3.4
Citral Pure (Hagan, et al., 1967)	Subchronic oral	500	–	215 ^a	50	4.3
Coumarin (Evans, et al. 1979)	Chronic oral	22.5	–	9.7 ^a	25	0.4
Eugenol F (Hagan, et al. 1967)	Subchronic oral	500	–	215 ^a	50	4.3
Limonene (Kirkpatrick, 2013)	Subacute Inhalation	–	543	543	150	3.6
Lillial (RIFM, 1984)	Subchronic oral	44.6	–	19 ¹	50	0.4
Linalool (Randazzo, 2012)	Subacute Inhalation	–	63	63	150	0.4
Linalyl acetate		–	–	63 ^b	150	0.4

^a Route to route calculation according to ECHA REACH guidance documents (ECHA, 2012a): Inhalation N(L)OAEC = oral NOAEL × (1/sRVrat) × (ABSoral-rat/ABSinh-rat) × (ABSinh-rat/ABSinh-human) = oral N(L)OAEL * (1/1.15 m³/kg/d) * (50%/100%).

^b Linalyl acetate is structurally closely related to linalool. In the absence of suitable long term repeated dose toxicity study on linalyl acetate, available data on linalool has been used on a read-across basis.

^c Default assessment factors according to ECHA REACH Guidance documents (ECHA, 2012a): 2.5 for Interspecies (remaining difference; no allometric scaling for derivation of inhalation DNEL); 10 for Intraspecies; 2 or 6 for exposure duration (2 – subchronic to chronic; 6 – subacute to chronic).

were not available for the fragrance material of interest, a NOAEC was estimated on the basis of an established no observed adverse effect level (NOAEL) by route-to-route extrapolation using the conversion algorithm recommended by ECHA (ECHA, 2012a). When doing this, it has to be taken into account that inhalation DNELs established by means of route-to-route extrapolation are considered suitable to address systemic endpoints, but may not sufficiently address local effects. The underlying NOAECs and NOAELs as well as the established DNELs are presented for each fragrance material considered in this investigation in Table 9.

Comparing the estimated consumer levels as presented in Table 7 to the DNELs presented in Table 9 reveals that the estimated exposures to the volatile fragrance materials are well below the established safe exposure levels (i.e., DNELs). For example, the smallest difference between the DNEL and an exposure scenario is that determined for linalool in the bathroom/toilet scenario as determined in the 1-box model: here the estimated TWA exposure (i.e., 58.1 µg/m³) is approximately seven times below the DNEL of 400 µg/m³. Considering a more realistic exposure by comparing to the exposure estimate with the 2-box model reveals already a difference of more than a factor 60. Additionally, all estimated peak values are below the respective long term safe exposure levels.

It would go beyond the scope and objective of this investigation to review the toxicology of each measured compound in full detail. However, from a toxicological point of view it is considered to be most appropriate to examine the assessment of potential consumer exposures to volatile formaldehyde, benzene and particulate matters in more detail. None of these materials is intentionally added to scented candles. They are formed as a result of incomplete combustion of the organic material present in the candle and released into indoor air.

Formaldehyde has been classified as a Group 1 carcinogen ('Carcinogenic to humans') by the International Agency for Research on Cancer (IARC, 2012), a Category 1B carcinogen ('Presumed human carcinogen') by the Risk Assessment Committee of the European Chemicals Agency (ECHA, 2012b) and is listed as a 'known to be a human carcinogen' in the 12th edition of the Report of Carcinogens published by the National Toxicology Program (US NTP, 2011). While IARC (2012) and the US NTP (2011) concluded that there is sufficient evidence from studies in humans paired with supporting evidence from experimental studies in animals that formaldehyde causes cancer of the nasopharynx and leukemia in

humans, the Risk Assessment Committee of ECHA did not consider that a causal relationship between exposure to formaldehyde and human nasopharyngeal cancer has been established with sufficient confidence. It based its Category 1B carcinogen classification '...largely on the animal data rather than the extensive but difficult to interpret epidemiological studies' (ECHA, 2012b). The Risk Assessment Committee of ECHA further stated that in the absence of convincing evidence for a biologically plausible mechanism and considering the discrepancy of results in epidemiological studies, a causal relationship between exposure to formaldehyde and induction of leukemia in humans cannot be concluded (ECHA, 2012b).

The evaluation of the cancer risks associated with inhalation exposures to formaldehyde requires a careful consideration of the likely mechanism of tumor initiation and promotion. While some genotoxicity has been observed for formaldehyde, it is the pre-dominant view of the academic and regulatory community that there is an association between cytotoxic, genotoxic and carcinogenic effects of formaldehyde. Hence, formaldehyde is widely considered as a carcinogen for which a toxicity threshold and hence a safe exposure level can be defined (EU, 2005; BfR, 2006; WHO, 2010). At concentrations below levels causing cytotoxic damage in the nasal mucosa (e.g., by irritation), the risk for humans to develop upper respiratory tract cancer is generally considered negligible. The German committee for setting occupational exposure limits ('MAK commission') has therefore classified formaldehyde as a Category 4 carcinogen, indicating that the carcinogenic effect of formaldehyde is secondary to cytotoxicity and that, under non-cytotoxic conditions, no relevant risk of cancer to humans is to be expected (MAK, 2002).

In terms of setting an acceptable indoor air exposure guideline value, the WHO considered sensory irritation as the critical outcome for guideline definition and has accordingly established the indoor air guideline value of 100 µg/m³ as the 30 min average concentration. The WHO considered the use of this short term guideline also as preventive of long-term health effects including cancer (WHO, 2010). Specifically with regard to protecting consumers against carcinogenic effects of formaldehyde, the WHO indoor air guideline value of 100 µg/m³ can be considered as conservative. Other modeled cancer risk-based exposure values have been proposed which actually support higher consumer exposures to formaldehyde. Conolly et al. (2004) predicted on the basis of a

biologically motivated computational modeling of the combined rodent and human dataset an additional risk of upper respiratory tract cancer of 10^{-6} or less for non-smokers exposed continuously to 200 ppm (i.e., $246 \mu\text{g}/\text{m}^3$) formaldehyde. Using a similar modeling approach, the German Federal Institute for Risk Assessment (BfR, 2006) concluded that a level of 100 ppm (i.e., $123 \mu\text{g}/\text{m}^3$) formaldehyde is 'safe' for the general population.

Fig. 3 presents formaldehyde exposure concentrations, as estimated with conservative standard assumptions using the ConsExpo 1-box and the RIFM 2-box models, in comparison to the WHO indoor air guideline value of $100 \mu\text{g}/\text{m}^3$. It further presents the comparison of the modeled exposure values in the 4 different scenarios to the range of formaldehyde indoor concentrations, typically found in homes as determined in a survey by the European Union (EU, 2005; see also Table 8).

Benzene is generally considered as a human carcinogen by regulatory bodies and has accordingly been classified as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC, 2012). For the purpose of the current investigation, it was considered suitable to compare the estimated human exposures to benzene from candle emissions to existing air quality guideline values as well as to benzene levels typically found in indoor as well ambient air. While so far no indoor air guideline value exist for benzene, ambient air quality levels or reference concentrations for non-cancer endpoints have been established by the European Union and the US EPA (see Table 8). The lowest existing ambient air limit value has been established in the EU by Directive 2000/69/EEC at $5 \mu\text{g}/\text{m}^3$. The highest 1-box benzene TWA consumer exposure estimates (i.e., $2.4 \mu\text{g}/\text{m}^3$; see Table 7) is approximately twice below this ambient air limit value. The highest estimated exposure according to the more refined, more realistic, 2-box model (i.e., $0.7 \mu\text{g}/\text{m}^3$; see Table 7) is already 7 times below this ambient air guideline value for benzene. However, as stated before in context of formaldehyde, it should be born in mind that these exposure estimates basically reflect a lifelong exposure to the estimated TWA concentration on the basis of default use assumptions which do not reflect a typical use and exposure pattern of candle users.

Fig. 4 presents benzene exposure concentrations as estimated with conservative standard assumptions using the ConsExpo

1-box and the RIFM 2-box models in comparison to the EU ambient air guideline value of $5 \mu\text{g}/\text{m}^3$ and the range of benzene indoor concentrations typically found in homes as determined in a survey by the European Union (EU, 2005; see also Table 8).

To manage risks associated with benzene exposures, various national and international regulatory bodies, such as the World Health Organization (WHO), have published exposure values for benzene associated with excess lifetime cancer risk. These values allow a more quantitative estimation of excess lifetime cancer risk under the assumption of 24 h exposure to the same concentration for 70 or more years. According to the WHO, the concentrations of airborne benzene exposures associated with an excess lifetime risk of 1/10,000, 1/100,000 and 1/1,000,000 are 17, 1.7 and $0.17 \mu\text{g}/\text{m}^3$ respectively (WHO, 2010). According to the US EPA, the concentrations of airborne benzene exposures associated with an excess lifetime risk of 1/10,000 and 1/1,000,000 are 13–45 $\mu\text{g}/\text{m}^3$ and 0.13–0.45 $\mu\text{g}/\text{m}^3$ respectively (US EPA, 2012).

Only a few institutions, however, published excess risk levels which are considered acceptable as this is, according to the WHO, a socio-political decision which should be defined at a national level. The EPA of California considers an excess lifetime risk of 1 in 100,000, calculated on the basis of realistic exposure scenarios, to be acceptable (OEHHA, 2001). The Dutch risk assessment institute, RIVM, bases its maximum permissible risk level of $20 \mu\text{g}/\text{m}^3$ on an acceptable risk of 1 in 10,000 (RIVM, 2001).

The estimated benzene exposure concentrations, as presented in Table 7, are, however, not considered to be suitable for a quantitative cancer risk assessment due to the underlying overly conservative standard use assumptions. Firstly, as was shown by the IPSOS data, consumers don't use scented candles on a daily basis for an entire life. Moreover, humans neither spend 24 h in a specific room, nor in a house. Various regulatory agencies such as the US EPA or the industry research institution ECETOC published exposure handbooks which provide estimations on the total time spent indoors (e.g., home, work, public buildings) or outdoors. It has been estimated that humans spend, on average, 21.7 h of the day indoors and only 2.4% of their time outdoors. Of the indoor time, humans spend on average 16.9 h at home (ECETOC, 2001; US EPA, 1997).

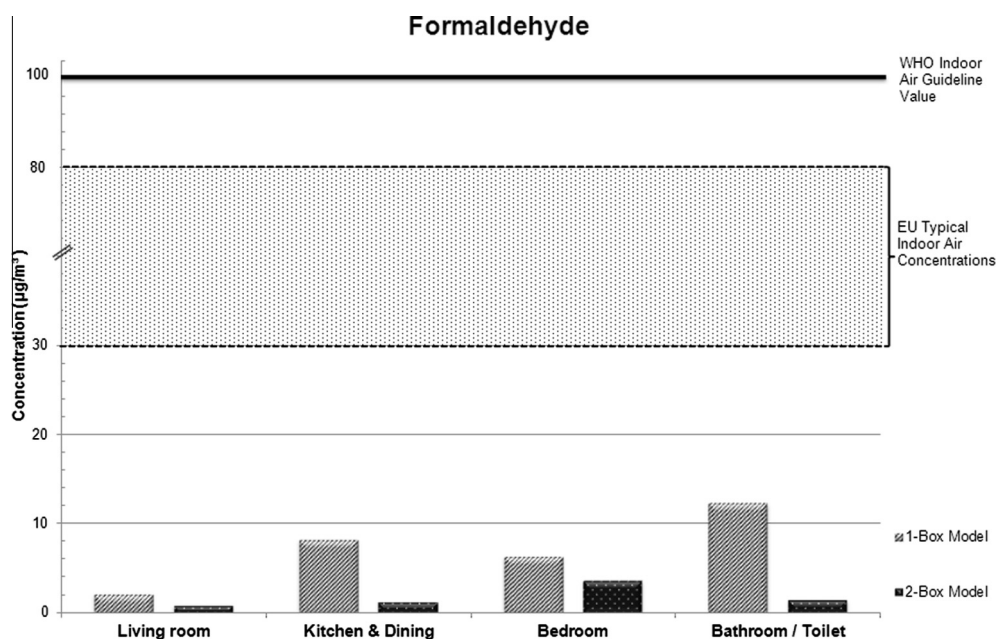


Fig. 3. Modeled time-weighted average (TWA) formaldehyde exposure concentrations in comparison to WHO indoor air guideline value of $100 \mu\text{g}/\text{m}^3$ and range of typical formaldehyde EU indoor air concentrations (EU, 2005).

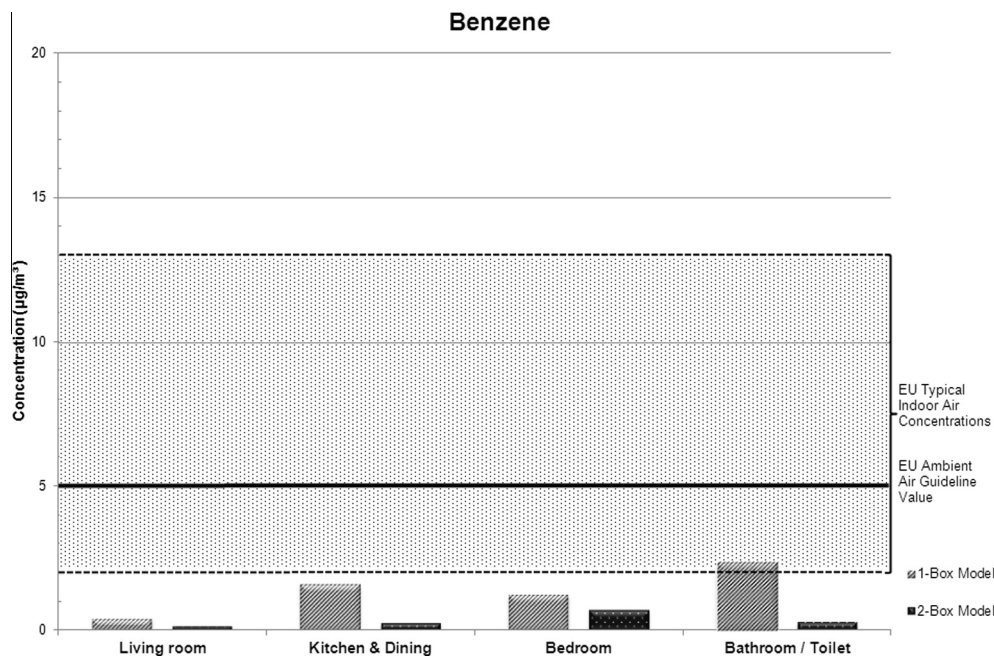


Fig. 4. Modeled benzene exposure concentrations in comparison to proposed EU ambient air guideline value of $5 \mu\text{g}/\text{m}^3$ and range of typical EU benzene indoor air concentrations (EU, 2005).

Table 10 presents monthly average exposures to a selected group of candle emission surrogates, i.e., formaldehyde, benzene, limonene and particulate matter, that have been calculated according to 1-box and 2-box models based on more realistic consumer habits and practice information that can be derived from the IPSOS study (IPSOS, 2006) and the time humans spend on average at home according to the US EPA (1997). For simplicity reasons, consumer exposures have been calculated for those rooms in which scented candles are known to be most frequently used (IPSOS, 2006).

On the basis of a more refined exposure assessment, the potential monthly average consumer exposure to benzene released from scented candles following use in the living room amounts to $0.05 \mu\text{g}/\text{m}^3$ according to the 1-box model or $0.02 \mu\text{g}/\text{m}^3$ according to the 2-box model. Using scented candles in the bedroom may result in average exposures of 0.02 or $0.04 \mu\text{g}/\text{m}^3$. In this context it is also worth noting that the IPSOS study revealed that 92% of the consumers purchasing scented candles use them in the living room and only 6% in the bedroom. Comparing these exposure estimates to the quantitative risk values provided by

the WHO (2010) or the US EPA (2012), suggests that cancer risks associated with inhaled benzene released from candles are *de minimis* (i.e., 10^{-6} or less).

With regard to potential health effects of particulate matter (PM), a large body of animal and epidemiological data has been published. Reviewing and discussing the entirety of the existing data would go beyond the scope of this investigation. It can be summarized that the critical effects, underlined in the available epidemiological literature, point to significant associations between long-term exposure to particulate matter and increases in mortality related to cardiovascular diseases. The WHO (2005) established the lowest annual exposure value of $20 \mu\text{g}/\text{m}^3$ for PM_{10} and $10 \mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ as air quality guidelines for particulate matters. PM_{10} is an indicator that is commonly relied on in the majority of published epidemiological studies and for which extensive measurement data are available worldwide. The WHO guideline values were established based on data for $\text{PM}_{2.5}$ and is the basis of the PM_{10} guideline value. The $\text{PM}_{2.5}$ guideline value of $10 \mu\text{g}/\text{m}^3$ as an annual mean for long term exposure was selected based on the available scientific literature that indicated it was below the mean concentration for

Table 10
Monthly average consumer exposure concentrations to candle emission surrogates benzene, formaldehyde, limonene and particulate matters.

Exposure Reference Value ^a ($\mu\text{g}/\text{m}^3$)	Living room <i>Habits & practice assumptions</i>		Bedroom <i>Habits & practice assumptions</i>	
	1-box model ($\mu\text{g}/\text{m}^3$)	2-box model ($\mu\text{g}/\text{m}^3$)	1-box model ($\mu\text{g}/\text{m}^3$)	2-box model ($\mu\text{g}/\text{m}^3$)
<i>Volatile organic compounds</i>				
Formaldehyde 100	0.25	0.09	0.19	0.17
Benzene 5	0.05	0.02	0.04	0.02
Limonene 3.6	0.08	0.03	0.06	0.05
<i>Particulate matters</i>				
$\text{PM}_{2.5}$ 10	1.78	0.64	1.42	0.81
PM_{10} 20	2.25		1.80	0.09

^a See discussion and Tables 8 and 9.

most health effects. Statistically, significant differences in morbidity and mortality have been reported at concentrations slightly greater than the guideline value. Typically, in urban areas of developing countries, the ratios $PM_{2.5}/PM_{10}$ falls within the range 0.5–0.8. Using $PM_{2.5}$ indicator studies and the lower end of the ratio, i.e., 0.5, the PM_{10} guideline value has been derived (WHO, 2005). It is important to note that these guidelines have not been established for indoor air quality. However, given the lack of a specific criterion for indoor air, it is considered suitable for the purpose of this health risk assessment.

Similarly to the benzene assessment, a more refined exposure assessment is required to allow an appropriate health assessment of consumer exposure to PM emissions. Comparing the potential PM_{10} and $PM_{2.5}$ exposure concentrations which have been calculated for the four different rooms with the exposure reference value of $20 \mu\text{g}/\text{m}^3$ for PM_{10} and $10 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ reveals that almost all estimates using the basic ConsExpo 1-box model exceed the exposure reference values for PM_{10} (i.e., calculated TWA ranging from 19.44 to $114.27 \mu\text{g}/\text{m}^3$) and $PM_{2.5}$ (i.e., calculated TWA ranging from 15.34 to $90.24 \mu\text{g}/\text{m}^3$). Calculating the consumer exposure to particulate matters, by using the conservative standards assumptions of all estimations, regardless of the model chosen, are above established exposure reference values. Refining consumer exposure concentration estimate to the particulate matter released from candles further with the more realistic consumers' habits and practice information that can be derived from the IPSOS study (IPSOS, 2006) and the time consumers spend on average at home according to the US EPA (1997) results in concentrations that are at 5–10 times below the established air guideline values (Table 8).

In this context it should be noted, that there are two distinct phases in terms of particle exposure during and after use of candles: as can be seen in Table 6, the actual emission of particulate matter during the burn phase is relatively low. However, following extinction of the candle there is, depending on the candle, a sharp increase of PM emissions ('sooting') which drive and determine the potential TWA PM concentrations to which consumers may be exposed. Product development efforts are targeted at generally producing low sooting, or clean burning, candles.

5. Conclusion

The present paper presents an approach for evaluating in a standardized way consumer health risks to emissions from candles used in indoor environments on the basis of environmental chamber studies. Measured analyte chamber concentrations were back-calculated to determine candle emission rates which can, depending on a specific candle use scenario, be modeled to obtain scenario-specific consumer exposure estimates to candle emissions. Consumer health risks can subsequently be assessed by comparing estimated consumer exposures to either regulatory guideline values or direct toxicological data, as available or deemed most appropriate.

In this investigation, emissions of volatile and semi-volatile organic compounds as well as particulate matter from scented candles have been investigated in independent and ISO accredited contract research laboratories according to adjusted ISO or ASTM emission measurement protocols in a comprehensive set of small and large chamber emission studies. This included substances that were intentionally added to the candles to give them their characteristic smell (i.e., fragrance ingredients such as limonene, linalool or cinnamic aldehyde) or that were formed as a result of incomplete combustion of organic material (e.g., short chain aldehydes including formaldehyde and acetaldehyde, benzene, styrene, polycyclic aromatic hydrocarbons PAH, polychlorinated dibenzodioxins PCDD and polychlorinated dibenzofurans PCDF).

While VOC and PM emissions from scented candles were clearly measurable and discernible from the chamber background

concentrations, semi-volatile PAHs and PCDD/PCDF emissions were below the detection limit and therefore not identified. Two models, the RIFM ConsExpo 1-box and the RIVM 2-box indoor air dispersion model were chosen to model consumer indoor exposures to candle emissions on the basis of highest measured compound-specific emission rates obtained from the environmental chamber studies. The process depends on using initially conservative exposure assumptions as well as more refined consumer research based habits & practice assumptions. In the European Union, the ConsExpo 1-box model is the tool recommended by regulators to estimate human inhalation exposures to chemicals released from consumer products. RIFM's 2-box indoor air dispersion model provides a more realistic view on consumer inhalation exposure to indoor chemicals by incorporating the understanding of air flow within an indoor environment, modeling the dispersion between two inter-connected zones. In this model, the emission source (i.e., the candle) is located in a living room with a defined volume and air flow is modeled between the room in which the candle sits and Zone 2 (i.e., the rest of the residence).

Potential consumer health risks associated with the candle emissions were characterized by comparing the potential human exposure concentrations with established indoor or, in the case of PM ambient air quality guidelines, where not existent, to established toxicity thresholds. With the exception of the PM emissions, all estimated peak and time weighted average concentrations were, despite the underlying conservative first tier standard assumptions (i.e., 4 h burning time, daily use of scented candles; 24 h consumer exposure to candle emissions), below established exposure reference values and are therefore not considered to be cause for safety concern.

Despite this overall favorable first tier assessment, it remains important to provide a more realistic understanding of potential consumer exposures. Typically, candles are not used by consumers on a daily basis. Neither are consumers exposed to candle emissions for 24 h each day. Modeling of potential consumer exposures to a group of surrogate compounds including formaldehyde, benzene, limonene and particulate matter using consumer research based habits and practice information revealed consumer exposures that were approximately 10 times below those values derived under the overly conservative standard assumptions which were used in the first Tier. Accordingly, all measured compound emissions including those of particulate matter fractions PM_{10} and $PM_{2.5}$ were clearly below existing indoor or ambient air quality guideline values or established toxicity thresholds (see Table 10).

On the basis of this investigation, it was concluded that under normal and foreseeable use conditions, the use of scented candles does not pose a safety concern to the consumer.

Conflict of Interest

Financial support for this work has been provided by a consortium of companies organized by the Research Institute for Fragrance Materials Inc. (RIFM).

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