



# Measurement and evaluation of gaseous and particulate emissions from burning scented and unscented candles

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## ABSTRACT

It has been known for a long time that incomplete combustion processes produce by-products that are harmful to human health. Particularly high concentrations of such by-products can arise in indoor environments when operating open flames without venting. The emission behavior of many combustion sources, including candles, has already been examined in detail. However, to date there are no studies in which the chemical composition of the candles is known exactly or where the candles were specifically manufactured for comparative measurements. In this respect, the study presented here, which was designed in collaboration with candle manufacturers and fragrance houses, demonstrates new insights into the emissions of burning candles depending on their composition.

All investigations were carried out under controlled climatic conditions in an 8 m<sup>3</sup> stainless steel chamber. Combinations of four different fuels (waxes) and five different fragrances in addition to one set of unscented control candles were examined. This resulted in 24 experiments, 20 with scented candles and four with unscented candles. The typical combustion gases carbon monoxide, carbon dioxide and NO<sub>x</sub>, organic compounds, such as formaldehyde, benzene, and polycyclic aromatic hydrocarbons, PM<sub>2.5</sub> and ultrafine particles were monitored in the chamber air and the emission rates were determined. The data were statistically evaluated using parametric and non-parametric methods as well as hierarchical cluster analysis. Exposure scenarios typical for indoor environments were calculated from the emission rates and the results were compared with indoor guidance and reference values.

As expected, a multitude of gaseous and particulate emissions were detected. These were typical combustion products as well as evaporated constituents of the fragrance mixtures. In most cases, the calculated indoor concentrations were well below the respective guidance and reference values. The exceptions observed in some cases for nitrogen dioxide, acrolein and benzo[a]pyrene are discussed critically.

## 1. Introduction

After mankind discovered the benefits of fire in terms of warmth and light, it was not long before combustion processes found their way into the cultural customs of societies. Over the centuries, smoking, the use of water pipes, incense sticks and especially candles have become firmly established human habits. The paramount importance of the candle in social and religious rituals inspired Michael Faraday to give his famous lecture on “*The Chemical History of a Candle*” (Faraday, 1848). Nevertheless, since the publication of John Evelyn’s “*Fumifugium*” (Evelyn, 1661) at the latest, it has been known that combustion processes can

produce gases and particles that are harmful to human health. Various combustion sources related to indoor air quality were identified (Singer et al., 2003; Géhin et al., 2008; Lee and Wang, 2006; Glytsos et al., 2010; Wallace and Ott, 2011; Stabile et al., 2012; Salthammer et al., 2014; Schripp et al., 2014; Fromme and Schober, 2015; Mullen et al., 2016; Posis et al., 2019). In some cases, particularly in churches, increased levels of indoor pollutants have been attributed to candle burning (Huynh et al., 1991; Fine et al., 1999; Chuang et al., 2012; Wallace et al., 2019).

In principle, the chemistry of flames (Gardiner, 1982) and hydrocarbon combustion (Hucknell, 1985) are well studied and understood.

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However, the combustion processes that occur in a candle in particular can be of a complex nature, since fragrances (Bartsch et al., 2016) and colorants are often added in addition to the basic components wax and wick. The gaseous and particulate substances that are formed and released during the combustion process can be selectively analyzed and quantified in an emission chamber under controlled climatic conditions (Salthammer, 2009; 2018).

Zai et al. (2006) studied the particulate emissions from candles in dependence of steady burn, unsteady burn and smoldering. Derudi et al. (2012; 2014) used a specially designed chamber geometry to study the chemical components of candle emissions. Klosterkötter et al. (2021) combined candle emission studies in a flow reactor with model room measurements. A number of other studies dealt with the emissions from burning candles under different conditions (Maupetit and Squinazi, 2009; Pagels et al., 2009; Orecchio, 2011; Stabile et al., 2012; Manoukian et al., 2013; Petry et al., 2014; Ahn et al., 2015; Rasmussen et al., 2021). Furthermore, Api et al. (2007) examined the transfer of fragrances from candles upon skin contact.

In recent years, the chemical composition of candles has changed depending on research results, environmental standards and consumer preferences. It therefore makes sense to examine the emission behavior of current products in detail. Consequently, the objective of this work was to determine and evaluate the release of gaseous compounds and airborne particles during the burning of candles with different fuel/fragrance combinations. The experimental setup and the composition of the test candles were designed in close collaboration with candle manufacturers, fragrance manufacturers and candle associations. This was done to best represent the candles currently on the market and determine the typical use patterns of consumers. For each type of candle the chemical composition was known and unit specific emission rates were calculated from the combustion product concentrations in a ventilated and well-mixed 8 m<sup>3</sup> stainless-steel emission test chamber. Furthermore, the burn rates (mass losses over time) were measured during each test and the burning behavior was documented. In total, 24 tests were performed with the four types of wax most commonly used as candle fuels (palm, paraffin, wax, soy, stearin), each with one of five different types of fragrances in addition to one set made without fragrance (unscented candles). From the results conclusions can be drawn as to whether the emissions from individual fuels differ significantly or whether there are differences between the scented and unscented candles. Furthermore, the data were converted to Reference Room concentrations, which allows a realistic estimate of the exposure level of consumers when burning candles in their home environment. The calculated concentrations of the gaseous and particulate pollutants can be assessed using guide and reference values.

## 2. Methods

The study methodology was based on the standard EN 16738 (2015), which describes a test procedure for measuring emissions from various combustible air fresheners, including scented candles during steady burn conditions. The experimental protocol involves a test chamber procedure for the determination of benzene, naphthalene, other VOCs (Volatile Organic Compounds) and formaldehyde, which is a VVOC (Very Volatile Organic Compound). In deviation from EN 16738 (2015), which demands a 1 m<sup>3</sup> test chamber, an emission test chamber with a volume of 8 m<sup>3</sup> was used in this study. This allowed the additional determination of particles and polycyclic aromatic hydrocarbons (PAHs). For these parameters a large sampling volume is necessary to reach a sufficiently low limit of quantification. The duration of the individual burn tests was also extended to 7 h. Sampling of the discontinuously determined VOCs and carbonyl compounds started 4 h after ignition of the candles when equilibrium (steady-state) conditions were reached. The data from the continuously recording measuring instruments were evaluated for the same period. The calculation of the emission rates for the parameters particles and polycyclic aromatic hydrocarbons (PAHs) took place over a period of 4.5 h (see Table 1). For each experiment, four candles were burned in the chamber simultaneously, thus a mean value was therefore measured for each individual candle type by averaging the differing emission and burning behaviors. The complete test protocol is compiled in Table 1.

### 2.1. Tested candles

#### 2.1.1. General

There is a wide variety of scented candles on the market that consumers can choose from. The most common differences relate to the type of wax (fuel), the fragrance ingredients and fragrance load, the wick, and the shape of candle (e.g. filled glass or pillar candle). This study explicitly aimed at examining and assessing scented candles in general and comparing them with unscented candles. An advisory committee of members of the European Candles Association, the National Candle Association and representatives of seven major fragrance houses supplying the global candle industry with fragrances (see Acknowledgement) developed the criteria for the manufacturing of candles. Once the criteria were fixed, candle manufacturers in Europe and the United States of America were assigned to find the proper wicks for acceptable burning performance for the different combinations and to produce the candles for this study.

**Table 1**

Test protocol. The conditions in the test chamber at t = -1h were AER = 2.0 h<sup>-1</sup>, T = 23 °C, RH = 50%.

| Time [hour] | Activity                                                                                                                                                               |
|-------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Before test | Sampling of VOCs and carbonyls (DNPH)                                                                                                                                  |
| -1          | Start of continuous measurements: CO, CO <sub>2</sub> , NO <sub>x</sub> , HCHO, O <sub>2</sub> and UFP in the empty chamber. Weighing of candles before the test cycle |
| 0           | 4 candles placed in the test chamber                                                                                                                                   |
|             | The 4 candles are lit (gas lighter); continuous weight loss monitoring for one candle with an electronic balance                                                       |
| 0           | Start of video recording                                                                                                                                               |
| 2.5         | Start sampling of PM <sub>2.5</sub>                                                                                                                                    |
|             | Start sampling of PAHs                                                                                                                                                 |
| 4           | Sampling of VOCs (60 min) and carbonyls (DNPH) (60 min)                                                                                                                |
| 6           | Sampling of VOCs (60 min) and carbonyls (DNPH) (60 min) *)                                                                                                             |
| 7           | End of continuous measurements CO, CO <sub>2</sub> , NO <sub>x</sub> , HCHO, O <sub>2</sub> , UFP                                                                      |
|             | End of sampling PM <sub>2.5</sub> , PAHs                                                                                                                               |
|             | End of video recording                                                                                                                                                 |
| 7           | Entering the test chamber and extinguishing of candles                                                                                                                 |
|             | Weighing of candles after test cycle                                                                                                                                   |
| After test  | Heating (thermal cleaning) the chamber for 15 h and preparation for the next test cycle                                                                                |

\*Analysis only in case of problems with samples from hour 4.

### 2.1.2. Fragrances and fuels

The relevant and representative fragrance mixtures and their ingredients were identified in a multistage process. As a first step, the common candle fragrances were divided into five families: floral, fresh, fruit, oriental and spice & edibles. The involved fragrance houses provided lists of all ingredients that are frequently used for each fragrance family. All ingredients mentioned by at least two fragrance houses plus three of the most common diluents that are necessary to dissolve the ingredients were added to a master ingredient list for each fragrance family. Subsequently, the fragrance houses were asked to assign common industry usage-level ranges (minimum to maximum) representative of overall industry practices for each fragrance material in each formula. The individual arithmetic means were used to create a representative concentration value for each fragrance material in each fragrance family. The final fragrance formulations with all ingredients and their concentrations are provided in the Supplementary Material (Tables S1 to S5).

The purpose of selecting the fragrance ingredients in this manner was to best represent an “average” candle that is on the market rather than to represent any one specific candle. By selecting the most common fragrance ingredients used in each of the five fragrance families at their typical use levels, an average candle that is more representative of candles sold to consumers was produced.

The fragrance load, i.e. how much fragrance is added to the fuel, varies a lot depending on consumer preferences, the country in which the candle is sold, the type of fragrance and the type of candle, etc. The advisory committee agreed to use a fragrance load of 5% by weight as this was considered to represent best the general European and North American market. Candles were produced containing 95% by weight of fuel mixed homogeneously with 5% by weight of fragrance. The most widely used fuels nowadays in Europe and Northern America are palm, paraffin wax, soy and stearin, either pure or blended with each other. The advisory committee decided to use pure fuels instead of blends.

### 2.1.3. Candle types

Scented candles come in different shapes and sizes, with or without containers, with one or several wicks. However, the most popular scented candle types both in Europe and North America are filled glasses with a single wick. This study used a slightly conical glass container with a diameter of approx. 76 mm (3.0 in.) at the top, a height of approx. 92 mm (3.6 in.) and a filling capacity of approx. 240 ml (8.1 Fl. Oz) considered representative for a scented candle with one wick (see Fig. 1A).

### 2.1.4. Wicks

Two candle manufacturers were assigned to find proper wicks for acceptable burning performance of the 24 different combinations (five different fragrance mixtures  $\times$  four different fuels in addition to one set

of the unscented fuels). It was not possible to use the same wick for all different combinations. One reason is that the viscosity of the fuel and fragrance mixtures was different, i.e. using the same wick size would have resulted in very different burn rates. The burn rate can considerably affect the performance of the candle including the combustion efficiency and the production of combustion by-products and particulates. Therefore, it was decided to use the individually chosen wicks for each fuel/fragrance combination that provided a consistent burn rate to allow comparisons of the results.

Another important reason is that the wicks require different chemical treatment for different fuels. The wick selection criteria for proper and comparable burning performance were: a) no visible emission of soot; b) average burn rate of 3.5–4.5 g/h for all combinations; c) burn rates of individual candles of one combination ideally in a range of  $\pm 10\%$  from the average burn rate of this combination; d) average burn rates of the different combinations ideally in a range of  $\pm 10\%$  from the overall average burn rate of all combinations.

It was possible to find proper wicks to comply with the first, most important criterion a) for all the combinations. Most combinations also complied with criterion b), but it was impossible to find wicks for the combinations paraffin/oriental and stearin/oriental to exceed a burn rate of 3.5 g/h without having visible emission of soot. Most combinations complied with criterion c) ( $\pm 10\%$ ) or did only exceed it slightly. Only the palm/fruit, palm/spice & edibles, paraffin/spice & edibles and stearin/spice & edibles combinations exceeded a range of  $\pm 15\%$ . Criterion d) was also met by most combinations. The palm/fresh and soy/spice & edibles combinations exceeded it slightly, the paraffin/oriental and stearin/oriental combinations exceeded it clearly ( $-19\%$ ) due to the lower burn rate that had to be chosen. The advisory committee accepted these deviations. If necessary, the wick was trimmed to a length of approx. 10 mm before the test began.

### 2.2. Emission test chamber

The experiments were performed in a 8 m<sup>3</sup> stainless steel emission test chamber (WEISS GmbH, Gießen, Germany) in accordance with EN 16738 (2015), ISO 16000-9 (2006) and EN 16516 (2017). Four candles were placed in wire mesh cylinders type 2 according to EN 15426 (2018) to protect the flames from draught caused by the higher than normal air exchange and enable calm burning conditions as during normal consumer use. The conditions in the test chamber during ignition were as follows: air exchange rate (AER) = 2.0 h<sup>-1</sup>, loading factor (L) = 4 candles/8 m<sup>3</sup>, temperature (T) = 23  $\pm$  2 °C, relative humidity (RH) = 50  $\pm$  5%. The air exchange rate corresponds to the continuous air flow into the chamber, which is determined by means of a calibrated mass flow controller. Continuous monitoring and sampling were performed before and during the burning phase. Regarding sink effects (Uhde and Salthammer, 2006) the chamber fulfills the requirements of ISO 16000-9

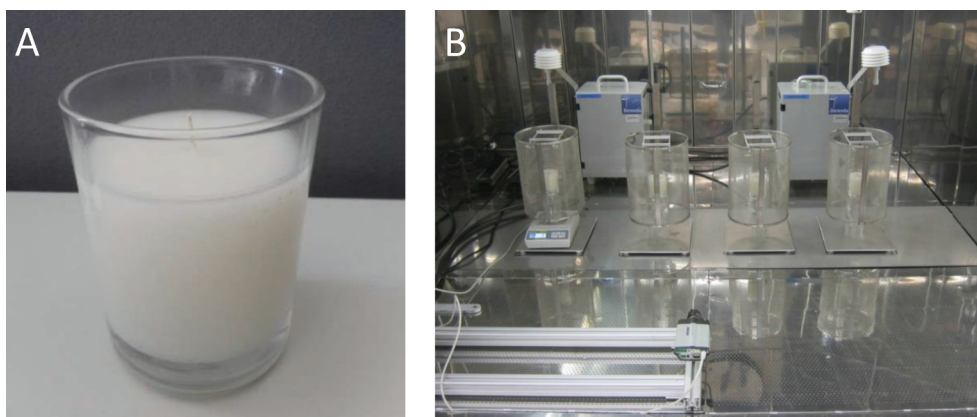


Fig. 1. Representative picture of a tested candle (A) and the experimental design in the 8 m<sup>3</sup> stainless-steel chamber.

(2006). As required by the standard, the recovery rates for the VOCs toluene (b.p. 110° C) and dodecane (b.p. 216° C) are higher than 80%. The concentration of VVOCs such as formaldehyde in the chamber is not influenced in the absence of sorptive materials (Gunschera et al., 2013). For nitrogen dioxide, the deposition rate on surfaces such as glass and metal can be neglected in relation to the air exchange (Grøntoft and Raychaudhuri, 2004). However, in the case of the high-boiling PAHs, sinks might cause an underestimation of emission rates. The experimental design is shown in Fig. 1B and the test protocol is provided in Table 1.

### 2.3. Analytical methods

The candles were weighed before and after the test (7 h total burning time), and the burn rate, i.e. the consumption of candle mass per hour, was calculated. In each experiment, the weight loss for one candle was followed online with an electronic analytical balance (Kern 440-35A).

Temperature and relative humidity were recorded by using a Rotronic Hygrolog-D data logger at intervals of 10 min.

The oxygen concentration was measured continuously using a Testo 330-1, which records the concentration with a resolution of 0.1 vol% and an accuracy of  $\pm 0.2$  vol%.

Carbon monoxide (CO) and carbon dioxide (CO<sub>2</sub>) were measured simultaneously by use of photo acoustic detector (INNOVA 1312) at wavelengths (wavenumbers) of 4.4  $\mu\text{m}$  (2270  $\text{cm}^{-1}$ ) and 4.7  $\mu\text{m}$  (2150  $\text{cm}^{-1}$ ) with a flow of 140 ml per sample and a time resolution of 60 s (flow rate 175  $\text{ml min}^{-1}$ ). The detection limit of the device was 0.2 ppm (0.2  $\mu\text{g/m}^3$ ) for carbon monoxide and 1.5 ppm (2.7  $\text{mg/m}^3$ ) for carbon dioxide, respectively.

Nitrogen oxides (NO and NO<sub>2</sub>) were analyzed using a chemiluminescence technique (Horiba, APNA-370) with a flow rate of 0.8 l/min and a time resolution of 60 s. A dual measurement principle was applied to determine nitrogen monoxide NO and NO<sub>x</sub> experimentally. Nitrogen dioxide (NO<sub>2</sub>) was calculated from the difference of the NO<sub>x</sub> and NO signals. The detection limits for NO and NO<sub>2</sub> were 0.5 ppb (0.66  $\mu\text{g/m}^3$ ; 0.95  $\mu\text{g/m}^3$ ).

The AL4021 (Aero-Laser GmbH) was used for online monitoring of the formaldehyde concentration. The system works on the basis of the Hantzsch reaction with fluorometric detection (Belman, 1963). Air is continuously sampled into the device and the formaldehyde is stripped in water. In a flow reactor, the solution is mixed with acetyl acetone and ammonium acetate and heated up to 70 °C. Under these conditions, formaldehyde is derivatized to 3,5-diacetyl-1,4-dihydrolutidine (DDL) which is quantified via fluorescence spectroscopy at 412 nm. The time to complete the reaction to DDL is 90 s and the selected data acquisition time was 2 s. The detection limit was 0.5 ppb (0.6  $\mu\text{g/m}^3$ ).

Volatile carbonyl compounds were trapped on DNPH-coated cartridges and analyzed after elution with acetonitrile by HPLC-UV according to ISO 16000-3 (2011). The samples were taken before loading the chamber; during the tests sampling started 4 h and 6 h after igniting the candles. The sampling time was 60 min and the total volume was 60 l (see Table 1).

Very volatile, volatile and semi volatile organic compounds (VVOCs, VOCs and SVOCs) were collected on Tenax TA® tubes spiked with internal standards. The component benzene was additionally collected on tubes filled with Carbotrap. All Tenax TA® samples were checked for possible degradation products (Klenø et al., 2002). The samples were taken before loading the chamber; during the tests sampling started 4 h and 6 h after igniting the candles. The sampling time was 60 min and the total volume was 5 l (see Table 1). Before using, the sorbent tubes were preconditioned with nitrogen. After sampling, the analysis was carried out on a J&W Scientific DB-5MS column (I = 60 m; I.D. = 0.25 mm; film = 0.25  $\mu\text{m}$ ) using a GC/MS system (Agilent 7890A/5975C) equipped with a thermal desorption system (Markes TD100). Identification was based on a probability-based matching (PBM) library search (McLafferty and Turecek, 1993) and analysis of retention data versus internal

standards. Moreover, mass spectra were compared with those of reference compounds. The detection limit was 1  $\mu\text{g/m}^3$ . The analytical procedure is in accordance with ISO 16000-6 (2011).

For sampling of PM<sub>2.5</sub> on quartz filters a low volume sampler (Derenda) was used. The total sampling time was 4.5 h with a flow rate of 2.3  $\text{m}^3/\text{h}$  (10.4  $\text{m}^3$  total volume). The mass on the filter was then determined gravimetrically with an electronic balance (Mettler Toledo, MX 5).

The number concentration and size distribution of particles in the size range between 5.6 nm and 560 nm were measured by using a TSI Model 3091 Fast Mobility Particle Sizer (FMPS). Because the vast majority of measured particle numbers were < 100 nm, the particles measured by FMPS were named as ultrafine particles (UFP).

The concentration of the 16 EPA polycyclic aromatic hydrocarbons (PAHs) (gas phase + particle phase) was analyzed according to VDI 3874 (2006) respectively ISO 12884 (2000) by an external accredited laboratory. Each sampling cartridge contained a PU foam spiked with internal standards and a glass fiber filter. The sampling time was 4.5 h with a flow rate of 2.3  $\text{m}^3 \text{h}^{-1}$  (10.4  $\text{m}^3$  total volume). After sampling, the PU foam was Soxhlet extracted with cyclohexane/toluene. The extract was cleaned up by column, measured via GC/MS and quantified with internal and external standards. The detection limit was 1  $\text{ng/m}^3$ .

Statistical analyses were carried out with the Origin Pro 18G (OriginLab Corp.) software package. Pearson's R was used as the fitting criterion for the linear regression. The hierarchical cluster analysis is based on the calculation of the Euclidean distance. Before the analysis, the applied data sets were normalized between 0 and 1 (Einax et al., 1997).

### 2.4. Calculation of unit specific emission rates $SER_{it}$ and exposure scenarios

The time resolved emission rate  $SER_t$  can be calculated from equation (1) (Salthammer, 2018).

$$SER_{u(t)} = \frac{\frac{\Delta C}{\Delta t} + AER \cdot C_t}{L} \quad (1)$$

$SER_{u(t)}$  ( $\mu\text{g/h}$ ) is the unit specific emission rate (related to the burning of a single candle) at time  $t$  (h),  $\Delta C/\Delta t$  is the change of concentration within the time interval of  $\Delta t$ ,  $AER$  is the chamber air exchange rate ( $2 \text{ h}^{-1}$ ),  $C_t$  ( $\mu\text{g/m}^3$ ) is the concentration of the target compound at time  $t$ , and  $L$  is the loading factor (0.5 candle/ $\text{m}^3$ ). For the steady-state ( $\Delta C/\Delta t = 0$ ), to which all calculated emission rates refer, equation (2) is obtained.

$$SER_u = \frac{AER \cdot C_t}{L} \quad (2)$$

The standards EN 16738 (2015) and EN 16739 (2015) describe a procedure for the calculation of short-term peak concentrations STPC ( $\mu\text{g/m}^3$ ) (see equation (3)) and long-term average concentrations TWA ( $\mu\text{g/m}^3$ ) (see equation (4)) in indoor environments. The calculated TWA is based on the assumption that the frequency of use of a scented candle is 4 times per week and that the candle burns 4 h per day with a constant emission rate (EN 16739, 2015; IPSOS, 2006).

$$STPC = \frac{SER_u}{V \cdot AER} \quad (3)$$

$$TWA = \frac{SER_u \cdot AUF}{V \cdot AER} \quad (4)$$

$V$  = room volume (here 30  $\text{m}^3$ ),  $AER$  = air exchange rate (here  $0.5 \text{ h}^{-1}$ ) and  $AUF$  = exposure time per day \* frequency of use days per week (here  $(4 \text{ h} / 24 \text{ h}) \times (4 \text{ days} / 7 \text{ days}) = 0.09523$ ). A volume of  $V = 30 \text{ m}^3$  and an air exchange rate of  $AER = 0.5 \text{ h}^{-1}$  are the standard conditions of the European Reference Room as defined in EN 16516 (2017). The calculated air concentrations (STPC or TWA) can then be compared with

**Table 2**

Guide values and reference values for indoor air (RfC = Reference Concentration for inhalation exposure as defined by the US EPA).

| Parameter                                  | Guide value or reference value         | Comment                      | Reference                          |
|--------------------------------------------|----------------------------------------|------------------------------|------------------------------------|
| Formaldehyde<br>(HCHO)                     | 0.1 mg/m <sup>3</sup>                  | 30 min                       | WHO (2010)                         |
|                                            | 0.10 mg/m <sup>3</sup>                 | 30 min                       | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 30 µg/m <sup>3</sup>                   | 30 min                       | EU (2005)                          |
| Acetaldehyde<br>(CH <sub>3</sub> CHO)      | 0.009 mg/m <sup>3</sup>                | lifetime RfC                 | US EPA (1991)                      |
|                                            | 200 µg/m <sup>3</sup>                  | long-term                    | EU (2005)                          |
|                                            | 0.10 mg/m <sup>3</sup>                 | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
| Acrolein                                   | 1.0 mg/m <sup>3</sup>                  | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 2 · 10 <sup>-5</sup> mg/m <sup>3</sup> | lifetime RfC                 | US EPA (2003b)                     |
|                                            | 8 · 10 <sup>-3</sup> mg/m <sup>3</sup> | lifetime RfC                 | US EPA (2008)                      |
| Propanal                                   | 0.10 mg/m <sup>3</sup>                 | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
| ΣC4-C9 Aldehydes<br>(aliphatic, saturated) | 2.0 mg/m <sup>3</sup>                  | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
| Benzene                                    | 0.17 µg/m <sup>3</sup>                 | lifetime                     | WHO (2010)                         |
|                                            | 0.03 mg/m <sup>3</sup>                 | lifetime RfC                 | US EPA (2003a)                     |
|                                            | 4.5 µg/m <sup>3</sup>                  | long-term                    | AIR (2020a)                        |
| Toluene                                    | 5 mg/m <sup>3</sup>                    | lifetime RfC                 | US EPA (2005)                      |
|                                            | 300 µg/m <sup>3</sup>                  | long-term                    | EU (2005)                          |
|                                            | 15,000 µg/m <sup>3</sup>               | short-term                   | EU (2005)                          |
| ΣXylenes<br>(o, m, p)                      | 0.30 mg/m <sup>3</sup>                 | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 3.0 mg/m <sup>3</sup>                  | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 200 µg/m <sup>3</sup>                  | long-term                    | EU (2005)                          |
| Styrene                                    | 20 mg/m <sup>3</sup>                   | short-term                   | EU (2005)                          |
|                                            | 0.10 mg/m <sup>3</sup>                 | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 0.80 mg/m <sup>3</sup>                 | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
| Limonene                                   | 250 µg/m <sup>3</sup>                  | long-term                    | EU (2005)                          |
|                                            | 2000 µg/m <sup>3</sup>                 | short-term                   | EU (2005)                          |
|                                            | 0.030 mg/m <sup>3</sup>                | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
| Naphthalene                                | 0.30 mg/m <sup>3</sup>                 | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 1.0 mg/m <sup>3</sup>                  | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 10.0 mg/m <sup>3</sup>                 | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
| Benzo[a]pyrene (BaP)                       | 10 µg/m <sup>3</sup>                   | long-term                    | EU 2005                            |
|                                            | 0.1 mg/m <sup>3</sup>                  | annual mean                  | WHO (2010)                         |
|                                            | 0.003 mg/m <sup>3</sup>                | lifetime RfC                 | US EPA (1998)                      |
| Carbon dioxide (CO <sub>2</sub> )          | 10 µg/m <sup>3</sup>                   | GV I                         | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 30 µg/m <sup>3</sup>                   | GV II                        | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 2 · 10 <sup>-6</sup> mg/m <sup>3</sup> | lifetime RfC                 | US EPA (2017)                      |
| Carbon monoxide (CO)                       | 0.012 ng/m <sup>3</sup>                | lifetime                     | WHO (2010)                         |
|                                            | 1000 ppm                               | harmless (hygiene based)     | Fromme et al. (2019)               |
|                                            | 2000 ppm                               | Inacceptable (hygiene based) | Fromme et al. (2019)               |
| Nitrogen dioxide (NO <sub>2</sub> )        | 35 mg/m <sup>3</sup>                   | 1 h                          | WHO (2010)                         |
|                                            | 10 mg/m <sup>3</sup>                   | 8 h                          | WHO (2010)                         |
|                                            | 7 mg/m <sup>3</sup>                    | 24 h                         | WHO (2010)                         |
| PM <sub>2.5</sub>                          | 40 µg/m <sup>3</sup>                   | annual mean                  | WHO (2010)                         |
|                                            | 200 µg/m <sup>3</sup>                  | 1 h                          | WHO (2010)                         |
|                                            | 40 µg/m <sup>3</sup>                   | 1 week                       | EU (2005)                          |
|                                            | 200 µg/m <sup>3</sup>                  | 1 h                          | EU (2005)                          |
|                                            | 0.080 mg/m <sup>3</sup>                | GV I (1 h)                   | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 0.25 mg/m <sup>3</sup>                 | GV II (1 h)                  | Fromme et al. (2019) <sup>1)</sup> |
|                                            | 10 µg/m <sup>3</sup>                   | annual mean                  | WHO (2005)                         |
|                                            | 25 µg/m <sup>3</sup>                   | 24 h mean                    | WHO (2005)                         |
|                                            | 35 µg/m <sup>3</sup>                   | 24 h mean                    | US EPA (2020)                      |

1) In the publication by Fromme et al. (2019) all guide values are provided in "mg/m<sup>3</sup>". Moreover, the German Committee on Indoor Guide Values (AIR) recently updated the number of significant decimals (AIR, 2020b). The current values are available on the UBA website <https://www.umweltbundesamt.de>.

short-term and long-term guide values for indoor air (see Table 2). The compliance factor (CF) according to EN 16739 (2015) describes the ratio of the calculated concentration (STPC or TWA) to the evaluation criterion. If the criterion is complied with, CF is < 1. For carbon dioxide and carbon monoxide, the background values measured in the chambers (CO<sub>2</sub> ≈ 400 ppm, CO ≈ 0.44 ppm) were added to the STPC and TWA.

### 3. Guide values for the evaluation of exposure scenarios

Depending on the concentration, exposure time and toxicological property, air pollutants can have negative impact on human health and welfare. In indoor areas, the precautionary principle is applied in order to provide all population groups with the best-possible protection against the negative health effects of air pollutants, whilst also excluding nuisances and irritants (Salthammer, 2011). The World Health Organization (WHO, 2005; 2010) and the EU (2005) have published guide values for selected indoor air pollutants, through which the general

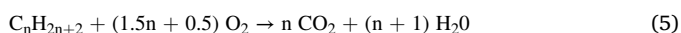
population should be protected against adverse health effects. The US EPA (United States Environmental Protection Agency) has published the so-called chronic Reference Concentrations for inhalation (RfC), which can also be used for the assessment of indoor areas. The RfC is an estimate of a continuous inhalation exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime. In Germany guide values are derived by the Committee on Indoor Guide Values (AIR) at the Federal Environment Agency. A distinction is made between Guide Value I and II (Fromme et al., 2019). Guide Value II (GV II) is an effect-related value based on current toxicological and epidemiological knowledge of a substance's effect threshold that takes uncertainty factors into account. Guide Value I (GV I) represents the concentration of a substance in indoor air for which, when considered individually, there is no evidence at present that even life-long exposure is expected to bear any adverse health impacts. The publication by Fromme et al. (2019) summarizes the available original works (in German) for individual

substances and substance groups. Table 2 lists all guide values that were used in this study for the health-related assessment. The selection (WHO, EU, US EPA, AIR) represents the international state-of-the-art. Depending on the exposure scenario for which the specific value applies, either the short-term or the long-term value was used for the evaluation.

## 4. Results and discussion

### 4.1. Candle burning rate, carbon dioxide and nitrogen oxides formation

The main fuels used for candles are hydrocarbons with chain lengths between C20 and C40, long-chain fatty acids and their esters. In the ideal case, as shown in the balance equation (5) using the example of hydrocarbons, optimum combustion takes place with the formation of carbon dioxide and water.



If the combustion is incomplete, carbon monoxide, elemental carbon and other hydrocarbons are also formed. The thermal conditions in a candle are very complex. The flame is divided into four zones with temperatures from 600 – 800 °C to 1200 – 1400 °C (Luerßen et al., 2015). Depending on the respective zone, pyrolytic, oxidative or recombination processes take place. In principle, high temperatures favor complete oxidation to carbon dioxide, but the then thermal formation of nitrogen oxides from nitrogen and oxygen must also be taken into account. The primary cause of the formation of NO<sub>2</sub> during the burning of candles is the high temperature of the candle flame of over 1000 °C, at which nitrogen and oxygen present in the ambient air react to form NO<sub>2</sub> (Miller and Bowman, 1989; Glarborg et al., 2018). As a result, the effective prevention of this NO<sub>2</sub> formation when burning a scented or unscented candle is virtually impossible. However, this general problem of NO<sub>2</sub> formation also affects all other sources in indoor air for which combustion processes play a role, such as open fireplaces, ethanol combustion, gas heaters, gas stoves, etc. Burning candles are also a source of other nitrogen compounds, e.g. nitrous acid (HONO) (Gligorovski, 2016). In a recent flow reactor study Klosterkötter et al. (2021) measured the ratio of HONO in NO<sub>x</sub> to be about 6.6%. Since the emission rate of HONO could not be determined in the context of this study, it is possible that HONO interferes with the NO<sub>x</sub> data.

The background values for carbon dioxide in the chamber correspond to the respective ambient air value, as these substances are not filtered by the air cleaning system. Consequently, the background values were subtracted to calculate the emission rates. The mean burning rates (mass losses over time) of the candles examined here, averaged from four candles, were between 2.89 g/h (stearin/oriental) and 4.57 g/h (paraffin/fresh) (see Table 3). Rasmussen et al. (2021), who conducted a similar study, measured higher burn rates between 4.7 g/h and 7.1 g/h. This is due to the fact that they used a different candle type (cylindrical pillar candles) made of different unscented fuels. These candles typically show a higher fuel consumption. In Fig. 2 the burning rate is plotted against the carbon dioxide emission rate SER<sub>CO<sub>2</sub></sub>. The relationship is linear to a good approximation and the calculated slope shows that 1 g of fuel

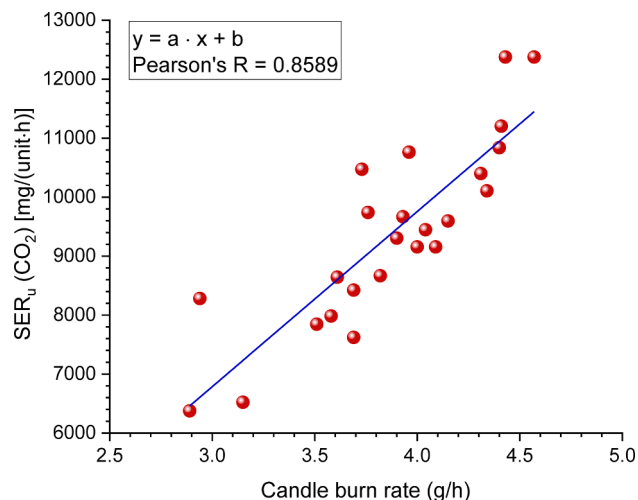


Fig. 2. Plot of the candle burn rate versus unit specific emission rate (SER<sub>CO<sub>2</sub></sub>) of carbon dioxide (CO<sub>2</sub>). The linear regression parameters are  $a = 2971 \pm 377$  mg/(unit g) and  $b = -1225 \pm 1471$  mg/(unit h).

produces 2971 mg of CO<sub>2</sub>. Under the model assumption that the fuel consists of C<sub>24</sub>H<sub>50</sub> (MW = 338 g/mol) only, 23 mol of CO<sub>2</sub> (MW = 44 g/mol) are formed from 1 mol of fuel, which is in very good agreement with the value of 24 mol of CO<sub>2</sub> per mole C<sub>24</sub>H<sub>50</sub> from the theoretical equation (5) for complete combustion. According to Hamins et al. (2005) the average carbon number of paraffin candle wax is 23 – 25.

### 4.2. Chamber concentrations and emission rates

In all experiments the temperature (23 °C), the relative humidity (50%) and the oxygen concentration (21%) varied only slightly over the test period of seven hours. A representative example is shown in the Supplementary Material (Figure S2).

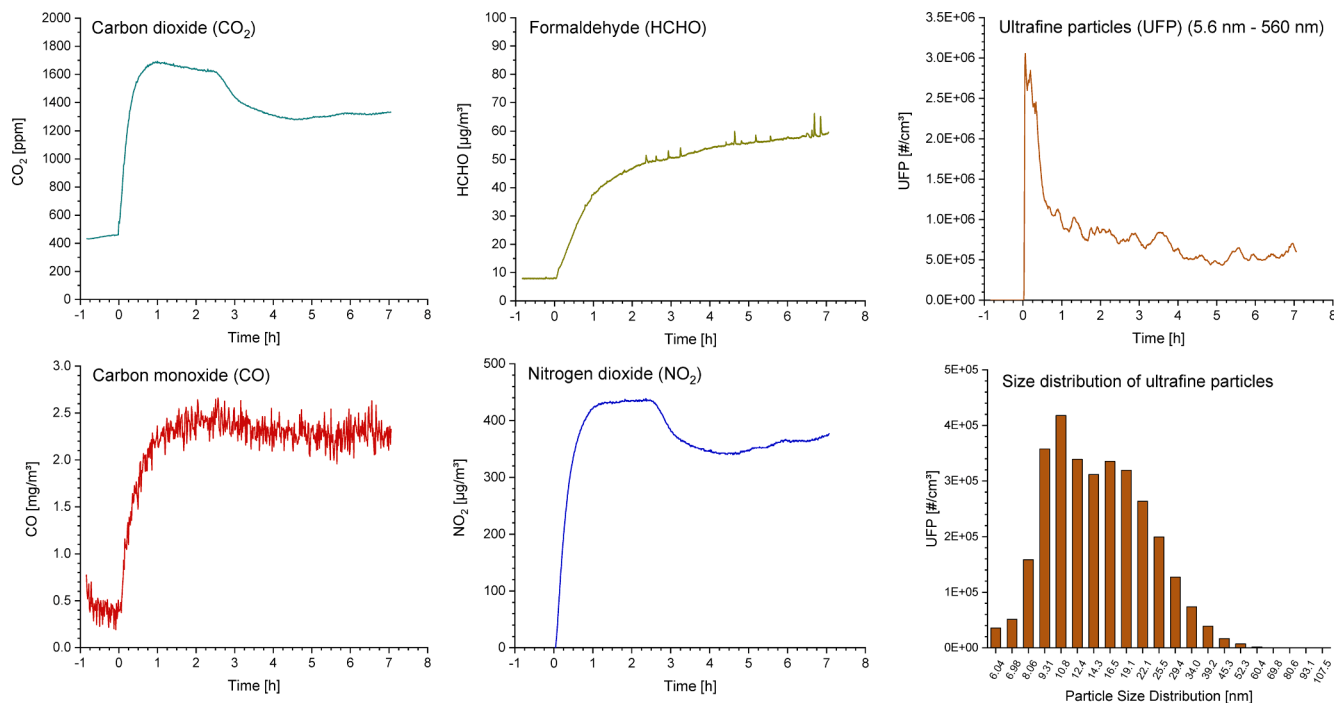
The concentration versus time behavior for various online measured parameters is shown in Fig. 3 using the data from the candle type “stearin/floral” over the entire measurement period of seven hours. Please note that the measured chamber concentrations refer to four burning candles in the 8 m<sup>3</sup> chamber and do not represent normal use conditions. In all cases, the respective concentration rises steeply immediately after lighting the candles. The background values for carbon dioxide, carbon monoxide and formaldehyde correspond to the air concentrations outside the chamber. The concentrations for carbon dioxide, carbon monoxide and nitrogen dioxide remain at a largely constant level with a slight decrease after about 2.5 h of burning time. For formaldehyde, on the other hand, the chamber concentration increases steadily, which indicates a constant slight increase in the emission rate. With a constant emission rate and an air change of 2 h<sup>-1</sup>, more than 99.9% of the steady-state concentration should be reached after 3 h of burning. In Fig. 3, the slight decrease in the concentration–time curves for carbon dioxide and nitrogen dioxide is significant for the combination stearin/floral, but not necessarily typical for all 24 experiments. In some cases the concentration is almost constant after reaching steady-state, in other cases small fluctuations also occurred. Such burning behavior is quite typical for real candles and may be related to small changes in the wick and in the fuel.

The concentration of ultrafine particles (UFPs) in the chamber rises sharply immediately after lighting and reaches its maximum within a few minutes (see also Fig. 3). The particle concentration then drops to a largely constant level within 1 h. The particle sizes are in the range from 6 nm to 60 nm with a count median diameter (CMD) of 18.6 nm. It takes a certain time for the temperature profile in the flame to develop completely. As a result, the soot particles formed from the fuel can initially only be partially oxidized to carbon dioxide. This behavior is

Table 3

The mean burn rates and standard deviations (mass loss calculated from 7 h burning time, n = 4) of the studied candles.

| Fragrance       | Palm        | Paraffin    | Soy         | Stearin     |
|-----------------|-------------|-------------|-------------|-------------|
|                 | [g/h]       |             |             |             |
| Floral          | 3.51 ± 0.09 | 3.96 ± 0.18 | 4.34 ± 0.29 | 3.15 ± 0.16 |
| Fresh           | 3.69 ± 0.37 | 4.57 ± 0.35 | 4.31 ± 0.17 | 3.69 ± 0.30 |
| Fruit           | 4.15 ± 0.43 | 3.73 ± 0.27 | 4.40 ± 0.21 | 3.93 ± 0.17 |
| Oriental        | 3.82 ± 0.21 | 2.94 ± 0.26 | 4.04 ± 0.43 | 2.89 ± 0.12 |
| Spice & Edibles | 3.90 ± 0.43 | 3.58 ± 0.53 | 4.41 ± 0.42 | 3.61 ± 0.35 |
| no fragrance    | 4.09 ± 0.43 | 4.43 ± 0.27 | 3.76 ± 0.53 | 4.00 ± 0.17 |



**Fig. 3.** Concentration versus time behavior of the online measured parameters carbon dioxide, carbon monoxide, nitrogen dioxide, formaldehyde and ultrafine particles for the four burning candles of type “stearin/floral” in the 8 m<sup>3</sup> stainless steel chamber with an air exchange of 2 h<sup>-1</sup>. The size distribution of the particles 6 min after lighting the candles is also shown. From the FMPS channel of 69.8 nm the particle number concentration was negligible and is therefore not shown. The short-term concentration peaks in the formaldehyde curve are caused by instrumental interference.

**Table 4**

Unit specific emission rates under steady-state conditions of inorganic gases, sum of VVOCs, VOCs, SVOCs, PAHs and particles from candle burning.

| Fuel     | Fragrance       | CO <sub>2</sub> <sup>1)</sup><br>[mg/(unit*h)] | CO <sup>1)</sup><br>[mg/(unit*h)] | NO   | NO <sub>2</sub> | ΣVVOC<br>[µg/(unit*h)] | ΣVOC<br>[µg/(unit*h)] | ΣSVOC<br>[µg/(unit*h)] | ΣPAH<br>[ng/(unit*h)] | PM <sub>2.5</sub><br>[µg/(unit*h)] | UFP<br>[#/(unit*h)] |
|----------|-----------------|------------------------------------------------|-----------------------------------|------|-----------------|------------------------|-----------------------|------------------------|-----------------------|------------------------------------|---------------------|
| Palm     | Floral          | 7844                                           | 8.5                               | 1.28 | 1.76            | 232                    | 4490                  | 282                    | 526                   | 94                                 | 1.4E + 12           |
|          | Fresh           | 7620                                           | 7.9                               | 1.28 | 1.92            | 40                     | 1644                  | 38                     | 316                   | 67                                 | 1.2E + 12           |
|          | Fruit           | 9596                                           | 10.6                              | 1.80 | 2.68            | 176                    | 5782                  | 14                     | 400                   | 80                                 | 9.7E + 10           |
|          | Oriental        | 8668                                           | 8.4                               | 1.44 | 1.92            | 348                    | 2014                  | < 4                    | 755                   | 62                                 | 2.6E + 12           |
|          | Spice & Edibles | 9304                                           | 8.4                               | 2.00 | 1.76            | 28                     | 1444                  | < 4                    | 160                   | 119                                | 3.3E + 11           |
| Paraffin | no fragrance    | 9156                                           | 6.1                               | 2.36 | 1.60            | < 4                    | 108                   | < 4                    | 79                    | 46                                 | 1.2E + 12           |
|          | Floral          | 10,764                                         | 11.6                              | 2.44 | 2.92            | 392                    | 10,218                | 98                     | 236                   | 122                                | 1.6E + 11           |
|          | Fresh           | 12,376                                         | 13.5                              | 2.20 | 3.96            | 528                    | 11,738                | 22                     | 172                   | 299                                | 1.1E + 11           |
|          | Fruit           | 10,476                                         | 10.7                              | 2.88 | 3.20            | 460                    | 14,388                | 68                     | 218                   | 121                                | 1.1E + 11           |
|          | Oriental        | 8280                                           | 9.4                               | 1.60 | 2.16            | 20                     | 2066                  | 132                    | 515                   | 88                                 | 4.3E + 11           |
| Soy      | Spice & Edibles | 7984                                           | 10.6                              | 1.80 | 1.68            | 126                    | 8202                  | 428                    | 1286                  | 221                                | 4.7E + 11           |
|          | no fragrance    | 12,376                                         | 7.2                               | 3.76 | 2.28            | 50                     | 70                    | < 4                    | 118                   | 124                                | 5.9E + 10           |
|          | Floral          | 10,108                                         | 12.1                              | 1.80 | 2.36            | 258                    | 5973                  | 20                     | 1018                  | 80                                 | 1.9E + 12           |
|          | Fresh           | 10,400                                         | 15.3                              | 1.28 | 2.76            | 448                    | 9408                  | < 4                    | 479                   | 102                                | 1.9E + 12           |
|          | Fruit           | 10,840                                         | 14.4                              | 2.04 | 3.00            | 226                    | 7856                  | 8                      | 434                   | 142                                | 1.7E + 12           |
| Stearin  | Oriental        | 9448                                           | 15.6                              | 1.36 | 2.08            | 296                    | 2059                  | 286                    | 703                   | 64                                 | 3.2E + 12           |
|          | Spice & Edibles | 11,204                                         | 15.2                              | 2.00 | 2.16            | 88                     | 4344                  | 132                    | 556                   | 379                                | 1.5E + 12           |
|          | no fragrance    | 9740                                           | 9.3                               | 2.04 | 1.60            | 38                     | 204                   | < 4                    | 194                   | 38                                 | 9.9E + 11           |
|          | Floral          | 6524                                           | 7.5                               | 1.16 | 1.36            | 274                    | 2756                  | < 4                    | 563                   | 64                                 | 1.7E + 12           |
|          | Fresh           | 8424                                           | 9.5                               | 1.28 | 2.16            | 1044                   | 8,446                 | < 4                    | 342                   | 70                                 | 8.2E + 11           |
|          | Fruit           | 9668                                           | 10.3                              | 1.96 | 2.60            | 600                    | 13,430                | < 4                    | 349                   | 74                                 | 6.2E + 11           |
|          | Oriental        | 6376                                           | 7.7                               | 0.96 | 1.44            | 16                     | 1244                  | 150                    | 365                   | 44                                 | 9.6E + 11           |
|          | Spice & Edibles | 8644                                           | 9.0                               | 1.84 | 1.44            | 58                     | 2855                  | < 4                    | 285                   | 127                                | 6.4E + 10           |
|          | no fragrance    | 9156                                           | 7.5                               | 2.08 | 1.52            | 32                     | 116                   | < 4                    | 192                   | 16                                 | 9.1E + 11           |

1) data corrected for chamber background concentrations.

typical when burning a candle and has been observed earlier (Zai et al., 2006; Rasmussen et al., 2021). Here, no shift to higher CMD is observed over time. Instead, the CMD drops to 11.8 nm after 4 h, indicating optimal combustion conditions.

The unit specific emission rates (average emission rates per candle) of the measured inorganic gases, individual compounds, sum values and particles are shown in Tables 4 and 5. The calculation was made with the equations (1) and (2) from mean chamber concentrations for the period

of 4–5 h (except PM<sub>2.5</sub> and PAHs, see Supplementary Material). With regard to the organic compounds, the sampling on Tenax TA® ensures an almost complete retention monitoring of the airborne VOCs in the gas chromatographic retention gap between n-hexane (C6) and n-hexadecane (C16) on a non-polar capillary column, but VVOCs (<C6) and SVOCs (>C16) are only partly recorded (Salthammer et al., 2018; Schieweck et al., 2018). For formaldehyde (HCHO), the results of the online measurement agree very well with the results of the discontinuous DNPH

**Table 5**

Unit specific emission rates under steady-state conditions of selected individual VVOCs and VOCs from candle burning (n.a. = not available, device not correctly calibrated).

| Fuel     | Fragrance       | HCHO<br>(online <sup>1</sup> )<br>[μg/(unit*h)] | HCHO | CH <sub>3</sub> CHO | Acrolein | Propanal | Butanal | Benzene | Toluene | Styrene | Naphthalene | Xylenes | Limonene | BaP<br>[ng/(unit*h)] |
|----------|-----------------|-------------------------------------------------|------|---------------------|----------|----------|---------|---------|---------|---------|-------------|---------|----------|----------------------|
| Palm     | Floral          | 110                                             | 109  | 29                  | < 8      | < 16     | < 16    | 16      | 22      | 38      | < 4         | 8       | 14       | < 4                  |
|          | Fresh           | 159                                             | 154  | 33                  | < 8      | < 16     | < 16    | < 4     | 8       | < 4     | < 4         | < 4     | 136      | < 1.2                |
|          | Fruit           | 257                                             | 260  | 80                  | 12       | < 16     | 24      | 24      | 28      | < 4     | < 4         | 4       | 442      | < 1.2                |
|          | Oriental        | n.a.                                            | 131  | 19                  | < 8      | < 16     | 22      | < 4     | 38      | < 4     | < 4         | 8       | 704      | < 4                  |
|          | Spice & Edibles | 74                                              | 69   | < 16                | < 8      | < 16     | < 16    | 8       | 8       | 10      | < 4         | < 4     | 8        | < 1.2                |
|          | no fragrance    | 17                                              | 16   | < 16                | < 8      | < 16     | < 16    | < 4     | 8       | < 4     | < 4         | < 4     | < 4      | < 1.2                |
| Paraffin | Floral          | 179                                             | 178  | 28                  | < 8      | < 16     | < 16    | 12      | 14      | 26      | < 4         | < 4     | 18       | < 1.2                |
|          | Fresh           | 360                                             | 363  | 82                  | 13       | 24       | 37      | 12      | 28      | 4       | < 4         | 20      | 1184     | < 1.2                |
|          | Fruit           | 235                                             | 246  | 64                  | 12       | < 16     | < 16    | 8       | 16      | < 4     | < 4         | < 4     | 996      | < 1.2                |
|          | Oriental        | 124                                             | 137  | 18                  | < 8      | < 16     | 27      | < 4     | 12      | < 4     | < 4         | < 4     | 716      | 6                    |
|          | Spice & Edibles | 97                                              | 97   | 18                  | 20       | < 16     | < 16    | 8       | 16      | 24      | < 4         | < 4     | 30       | 9                    |
|          | no fragrance    | 18                                              | 16   | < 16                | < 8      | < 16     | < 16    | < 4     | 10      | < 4     | < 4         | < 4     | < 4      | < 4                  |
| Soy      | Floral          | 145                                             | 140  | 24                  | < 8      | < 16     | < 16    | 5       | 18      | 14      | < 4         | < 4     | 66       | 8                    |
|          | Fresh           | 256                                             | 265  | 57                  | 10       | < 16     | 34      | 10      | 20      | < 4     | < 4         | 12      | 1006     | < 4                  |
|          | Fruit           | 230                                             | 221  | 56                  | 8        | < 16     | < 16    | 8       | 14      | < 4     | < 4         | < 4     | 696      | < 1.2                |
|          | Oriental        | n.a.                                            | 116  | 16                  | < 8      | < 16     | < 16    | 5       | 20      | < 4     | < 4         | 4       | 652      | 4                    |
|          | Spice & Edibles | 128                                             | 126  | 27                  | 14       | < 16     | < 16    | 6       | 10      | 6       | < 4         | < 4     | 64       | < 4                  |
|          | no fragrance    | 23                                              | 20   | < 16                | < 8      | < 16     | < 16    | < 4     | 8       | < 4     | < 4         | < 4     | 26       | < 1.2                |
| Stearin  | Floral          | n.a.                                            | 86   | < 16                | < 8      | < 16     | < 16    | 10      | 16      | 24      | < 4         | < 4     | 4        | < 1.2                |
|          | Fresh           | n.a.                                            | 194  | 41                  | < 8      | 17       | 22      | 15      | 40      | < 4     | < 4         | 18      | 912      | 12                   |
|          | Fruit           | n.a.                                            | 240  | 69                  | 10       | < 16     | 25      | 10      | 44      | 6       | < 4         | 8       | 960      | < 4                  |
|          | Oriental        | 100                                             | 102  | < 16                | < 8      | < 16     | 20      | < 4     | 10      | < 4     | < 4         | 4       | 494      | < 4                  |
|          | Spice & Edibles | 56                                              | 58   | < 16                | < 8      | < 16     | < 16    | 5       | 16      | 24      | < 4         | 10      | 8        | < 4                  |
|          | no fragrance    | n.a.                                            | 24   | < 16                | < 8      | < 16     | < 16    | < 4     | 4       | < 4     | < 4         | < 4     | < 4      | < 4                  |

1) Continuously measured by use of an AL4021 (Aero-Laser GmbH, acac method), data corrected for experimental baseline if necessary.

measurement (see Table 5). Szulejko and Kim (2016) compared formaldehyde emission rates for different combustible products. For candles, emission rates between 3 µg/h and 310 µg/h are given. The authors state that these values are at the lower end compared to the other combustion sources studied (cigarettes, unvented decorative fire places, mosquito coils, unvented kerosene heaters).

Interesting connections and results arise for the organic compounds. First of all, it should be noted that the group of SVOCs (except PAHs, see below) does not play a major role and is therefore not considered further. The highest emission rate of 428 µg/(unit h) (paraffin/spice & edibles) is mainly caused by the not clearly identified ester of a higher carboxylic acid.

All candles without the addition of fragrances show by far the lowest emissions of organic substances. Only traces of typical combustion reaction products (benzene, toluene, acetophenone) are detectable. The increased value of 204 µg/(unit h) for soy/no fragrance may be due to slight contamination with terpenes. If the fragrances are compared directly, floral, fresh and fruit tend to cause higher emissions than oriental and spice & edibles. The reason for this lies in the different volatility of the fragrance components in different fragrance families, as these substances evaporate directly from the hot liquid fuel at different rates. The essential components of the fragrances used here are also known from other products (Nazaroff and Weschler, 2004; Liu et al., 2004; Singer et al., 2006; Su et al., 2007; Uhde and Schulz, 2015). Substances such as benzaldehyde (a main component of the fragrance “fruit”), limonene, linalool, etc. have high vapor pressures and are therefore predominant in the chromatograms of the air samples. However, the combination paraffin/spice & edibles also has a comparatively high ΣVOC value, which is due to the components 4-methoxy-benzaldehyde (anisaldehyde), cinnamaldehyde, methylcinnamaldehyde and eugenol. Benzaldehyde, limonene, various acetates and the esters of higher carboxylic acids are mainly responsible for the high ΣVOC value of 14388 µg/(unit h) for paraffin/fruit. If this emission rate is converted to the conditions of the Reference Room ( $V = 30 \text{ m}^3$ ,  $\text{AER} = 0.5 \text{ h}^{-1}$ ),  $\Sigma\text{VOC} = 959 \text{ µg/m}^3$  results for the short-term exposure. This value, which can only be assessed from a hygienic point of view, corresponds to Level 2 in the concept of the German Committee for Indoor Guide Values (AIR) (Fromme et al., 2019): “no relevant consequences required, increased

ventilation is recommended”.

As far as ultrafine particles (UFPs) are concerned, the release is between  $5.9 \cdot 10^{10} \text{ \#/(unit h)}$  and  $3.2 \cdot 10^{12} \text{ \#/(unit h)}$ . This is in the range of other emission sources for UFP indoors (Weschler and Shields, 1999; Wallace and Ott, 2011; Salthammer et al., 2012; Isaxon et al., 2015; Gu et al., 2019). By converting the median value (see Table 4) from  $9.4 \cdot 10^{11} \text{ \#/(unit h)}$  to the conditions of the Reference Room using equation (3) for short-term exposure, a concentration of  $6.3 \cdot 10^4 \text{ \#/cm}^3$  is obtained. This is in the range of the median determined by Isaxon et al. (2015) from 17 experiments for the particle number concentration caused by candles in a living space. However, the conversion of particle concentrations and emissions is usually associated with great uncertainties, as particles are subject to high dynamic processes such as diffusion, settling, condensation and evaporation, depending on their chemical composition, concentration and environment (Hinds, 1999). It must also be noted that the initial exposure to particles is underestimated here. During the ignition phase of approximately 30 min, the particle concentration is on average a factor of 3–5 higher than under steady burn conditions.

The ΣPAH emission rates range between 79 ng/(unit h) for palm/no fragrance and 1286 ng/(unit h) for paraffin/spice & edibles. In all cases, the ΣPAH emissions of the candles without fragrance were considerably lower than those of the scented candles. A comparison with Derudi et al. (2012) and Orecchio (2011) (both also measured the sum of gas phase and particle phase PAHs) is only possible to a limited extent, as the data from these authors refer to the mass of the candle material burned. Assuming an average burning rate of 4 g/h (see Fig. 2 and Table 3), the maximum value of 49.8 µg/kg published by Orecchio (2011) results in an emission rate of 199 ng/(unit h), which is at the lower end of the range measured in this work. For BaP, with the maximum value from Orecchio (2011) of 1.4 µg/kg, an emission rate of 5.6 ng/(unit h) is obtained, which is in the order of magnitude with the value of 12 ng/(unit h) measured for stearin/fresh (see Table 5).

When the scents are compared, systematic differences can be observed for formaldehyde and acetaldehyde emissions. The fragrances floral, oriental and spice & edibles consistently show significantly lower emissions than the fresh and fruit fragrances. For the parameters benzene and PM<sub>2.5</sub>, the oriental fragrance consistently shows the lowest

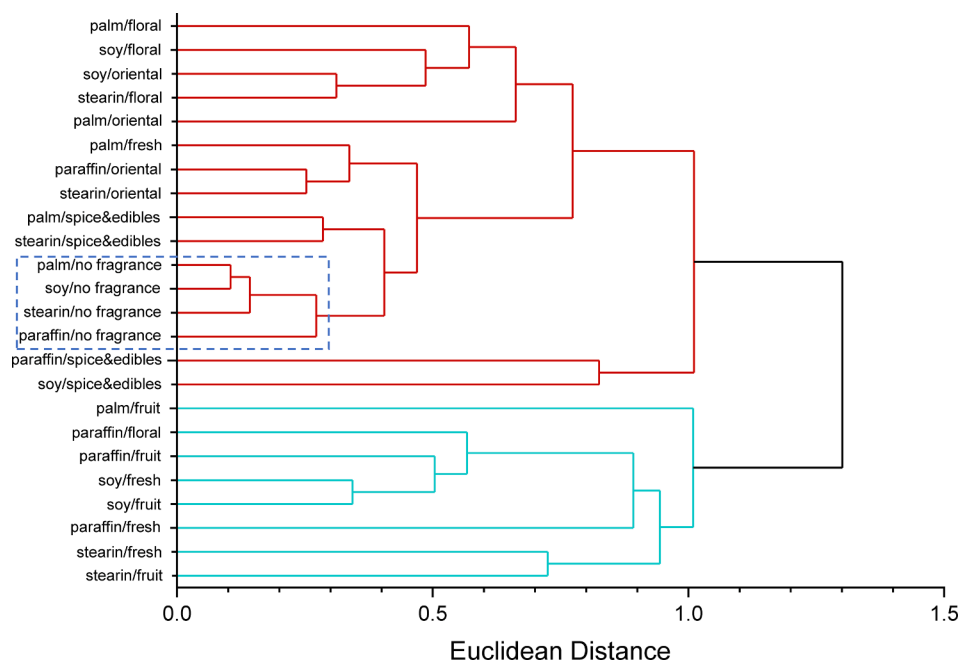


Fig. 4. Hierarchical cluster analysis of fuel/fragrance combinations. Emission rate data sets of the following parameters were used as variables: formaldehyde (HCHO), acetaldehyde (CH<sub>3</sub>CHO), benzene, toluene, ΣVOC, ΣVOC, ΣPAH, PM<sub>2.5</sub>.

emissions. The fragrance spice & edibles, on the other hand, almost always shows the highest emission for PM<sub>2.5</sub>.

Fig. 4 shows the results of a hierarchical cluster analysis. This statistical method is useful for making visible structures within observed and given data (Einax et al., 1997; see Supplementary Material for more detailed information). The following parameters, which were dependent on the fuel/fragrance combination, were used for the analysis: formaldehyde, acetaldehyde, benzene, toluene,  $\Sigma$ VOC,  $\Sigma$ VOC,  $\Sigma$ PAH PM<sub>2.5</sub>. As expected, all four fuels without fragrance are clustered in the dendrogram (see dashed box in Fig. 4). A total of eight combinations with high emissions form a separate cluster (green). Seven of them belong to the fragrances fruit and fresh. With one exception, the fragrances floral, oriental, spice & edibles are grouped in the other cluster (red). Interestingly, these combinations are more similar to the fuels without fragrances than to the green cluster with the high emissions. No further significant similarities or differences between the fuel/fragrance combinations can be derived from the dendrogram.

All results of this study relate to steady burning conditions, with the exception of the initial phase after lighting the candle. No smoldering or excessive soot generation was observed. The release of pollutants during different burning modes (steady, soot development, smoldering) was investigated by Zai et al. (2006) and Pagels et al. (2009). In the case of particle emissions, a shift to larger diameters and higher masses was observed under non-optimal conditions in both studies. In the case of organic and inorganic compounds, the emission rates increased significantly.

#### 4.3. Evaluation of compliance factors with respect to guide values

To assess the exposure, realistic assumptions must be made about its duration. For short-term exposure (STPC), emission rates can be converted directly into expected air concentrations. For long-term exposure (TWA), factors may have to be taken into account. It was assumed that in an average household the burning time of a candle is 4 h per day and 4 days per week. From an analytical point of view, the TWA corresponds to the result of a passive sampler measurement over a period of one week. Bekö et al. (2013) investigated consumer behavior in relation to

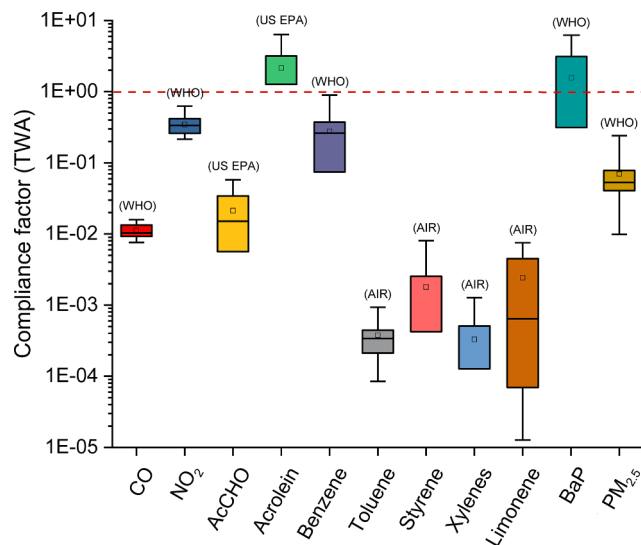


Fig. 6. Box-whisker plots (minimum, maximum 25- and 75-percentile, mean, median) of compliance factors (CF) for time-weighted average (TWA) concentrations. The unit specific emissions rates (SER<sub>u</sub>) were taken from Tables 4 and 5 and converted to Reference Chamber concentrations ( $V = 30 \text{ m}^3$ ,  $AER = 0.5 \text{ h}^{-1}$ ,  $AUF = 0.09523$ ) using Equation (4). The following values were used for calculating compliance factors: CO:  $7 \text{ mg/m}^3$  (WHO, 2010); NO<sub>2</sub>:  $40 \text{ }\mu\text{g/m}^3$  (WHO, 2010); acetaldehyde (CH<sub>3</sub>CHO):  $0.009 \text{ mg/m}^3$  (US EPA, 1991); acrolein:  $2 \cdot 10^{-5} \text{ mg/m}^3$  (US EPA, 2003b); benzene:  $0.17 \text{ }\mu\text{g/m}^3$  (WHO, 2010); toluene:  $0.30 \text{ mg/m}^3$  (Fromme et al., 2019); styrene:  $0.030 \text{ mg/m}^3$  (Fromme et al., 2019); xylenes:  $0.10 \text{ mg/m}^3$  (Fromme et al., 2019); limonene:  $1.0 \text{ mg/m}^3$  (Fromme et al., 2019); benzo[a]pyrene (BaP):  $0.012 \text{ ng/m}^3$  (WHO, 2010); PM<sub>2.5</sub>:  $10 \text{ }\mu\text{g/m}^3$  (WHO, 2005). Note: The paper by Fromme et al. (2019) summarizes the work of German Committee on Indoor Guide Values (AIR).

various activities and calculated an average daily source duration of 142 min/d (2.37 h/day) for burning a candle. Their weekly burning time of 16.5 h is in very good agreement with the 16 h used here. It is clear that the use of candles varies over a wide range in terms of number and time and that the 16 h is consequently a convention value, which allows the comparison of different candle types. When calculating STPC and TWA, however, the phase immediately after lighting a candle is not taken into account and constant emission rates are assumed. For the gaseous emissions, it takes some time to establish equilibrium, depending on the air exchange. For the ultrafine particles, on the other hand, the total exposure due to exclusion of the initial peak is underestimated.

In Fig. 5 the box-whisker plots of the compliance factors (CF) for short term peak concentrations (STPC) of carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>), formaldehyde (HCHO) and PM<sub>2.5</sub> are displayed. For NO<sub>2</sub>, two guide values from different organizations are compared. The US EPA (2020) defined a 24-hour PM<sub>2.5</sub> standard of  $35 \text{ }\mu\text{g/m}^3$  for ambient air. In case of PM<sub>2.5</sub> a 24 h mean of  $25 \text{ }\mu\text{g/m}^3$  as recommended by the WHO (2005) for outdoor air and the German Committee on Indoor Guide Values for indoor air (Fromme et al., 2019) is applied. All individual CF values for CO, CO<sub>2</sub>, formaldehyde and PM<sub>2.5</sub> are  $< 1$ , so the STPCs are lower than the corresponding guide values. For NO<sub>2</sub>, the use of the WHO guide value of  $200 \text{ }\mu\text{g/m}^3$  leads to two cases with CF values greater than 1. Using the GV I value of  $0.080 \text{ mg/m}^3$  results in increased CF values in the range between 1.1 and 3.1 with a median of 1.8. GV I is a precautionary value and contains numerous additional safety factors for particularly sensitive population groups (AIR, 2019).

Fig. 6 shows box-whisker plots of the CF values for the time weighted averages (TWA) of carbon monoxide (CO), nitrogen dioxide (NO<sub>2</sub>), acetaldehyde, various VOCs, BaP and PM<sub>2.5</sub>. For NO<sub>2</sub>, the long-term values of the WHO and the EU ( $40 \text{ }\mu\text{g/m}^3$ ) were complied with by all the candle types examined. For CO, acetaldehyde, toluene, styrene,

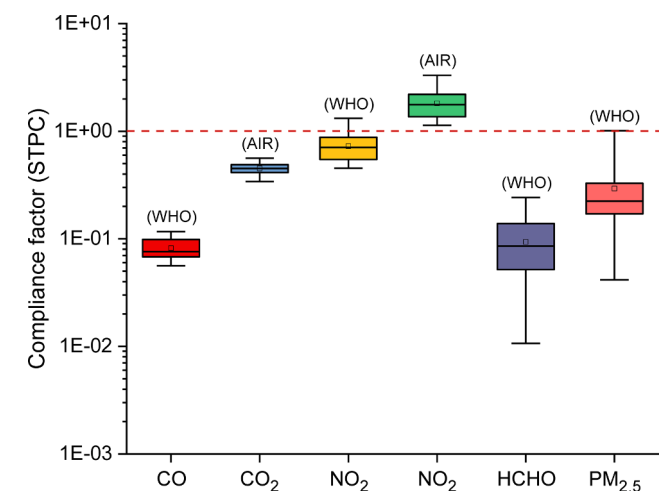


Fig. 5. Box-whisker plots (minimum, maximum 25- and 75-percentile, mean, median) of compliance factors (CF) for short-term peak concentrations (STPC). The unit specific emissions rates (SER<sub>u</sub>) were taken from Tables 4 and 5 and converted to Reference Chamber concentrations ( $V = 30 \text{ m}^3$ ,  $AER = 0.5 \text{ h}^{-1}$ ) using equation (3). The following values were used for calculating compliance factors: CO:  $10 \text{ mg/m}^3$  (WHO, 2010); CO<sub>2</sub>: 2000 ppm (Fromme et al., 2019); NO<sub>2</sub>:  $200 \text{ }\mu\text{g/m}^3$  (WHO, 2010); NO<sub>2</sub>:  $0.080 \text{ mg/m}^3$  (Fromme et al., 2019); formaldehyde (HCHO):  $0.1 \text{ mg/m}^3$  (WHO, 2010); PM<sub>2.5</sub>:  $25 \text{ }\mu\text{g/m}^3$  (WHO, 2005). Note: The paper by Fromme et al. (2019) summarizes the work of German Committee on Indoor Guide Values (AIR).

xylene, limonene and PM<sub>2.5</sub> the CF values are in the range of 0.1 or lower. According to the WHO (2010) guide value of 0.17 µg/m<sup>3</sup>, the median CF for benzene is 0.26. The German Committee on Indoor Guide Values (AIR, 2020a) recently published a preliminary indicator value of 4.5 µg/m<sup>3</sup>. BaP was found in 5 out of 24 experiments. Using the US EPA guide value of 2·10<sup>-6</sup> mg/m<sup>3</sup> leads to low CF values between 0.002 and 0.04 (not shown), while using the WHO guide value of 0.012 ng/m<sup>3</sup> results in considerably higher CF values between 0.32 and 6.35 (see Fig. 6). It must be mentioned that measurements in the range of the limit of determination (as in the case of BaP) are subject to greater uncertainties. In this case it is also difficult to evaluate the reliability of the calculated CF values.

Acrolein was detected in eight experiments. As shown in Fig. 6, all CF values are higher than 1 (range between 1.3 and 6.3) if the US EPA RfC of 2·10<sup>-5</sup> mg/m<sup>3</sup> is applied. Due to the lack of reliable long-term toxicity data on this compound the US EPA used a safety factor of 1000 in setting this standard. This raises the problem that the resulting RfC is far below the limit of analytical detection and quantification (Schieweck et al., 2021). The impracticability of non-accessible guide values was discussed in detail by Salthammer (2011). The OEHHA gives the following Reference Exposure Limits (REL) for the inhalation path of acrolein: acute REL = 2.5 µg/m<sup>3</sup>; 8 h REL = 0.7 µg/m<sup>3</sup>; chronic REL = 0.35 µg/m<sup>3</sup> (Office of Environmental Health Hazard Assessment, 2015). Logue et al. (2011) have evaluated several studies and calculate a median of 0.84 µg/m<sup>3</sup> for acrolein in indoor air. Similar to NO<sub>2</sub>, acrolein is a by-product that cannot be avoided in thermal processes of organic substances.

However, it is also clear from Table 2 and especially from the acrolein example, that even toxicologically based guide values might differ by several orders of magnitude. This complicates the health-related assessment of the measured emission rates, which sometimes leads to contradicting results.

## 5. Conclusions

As expected, a multitude of gaseous and particulate emissions were detected during the burning of the tested candles. These were typical combustion products as well as evaporated constituents of the fragrance mixtures. In the initial phase, which lasts 30 min – 60 min (see also Rasmussen et al., 2021), significantly increased concentrations of ultrafine particles could be observed. In contrast, the concentrations of carbon monoxide, carbon dioxide, nitrogen dioxide and formaldehyde were initially not increased. The formation of NO<sub>2</sub> is a characteristic of combustion chemistry in air and cannot be avoided. During the safety assessment of the emissions, it was determined that for the vast majority of measured parameters for which toxicologically justified indoor air guide values are available, the corresponding short-term and long-term concentrations for indoor air are safely met and, in many cases, are well below the established safety levels. For benzo[a]pyrene (BaP), an indicator for incomplete combustion processes, and acrolein the long-term concentrations set by WHO and US EPA were exceeded or partly exceeded.

The results of the 24 experiments provide insights into the burning behavior of various combinations of fuel and fragrance. For the unscented candles it can be determined that the fuels palm, paraffin, soy and stearin behave very similarly in terms of emissions (see Tables 4 and 5). No individual wax was shown to have a consistently better emission profile than the others. This group also forms a cluster in the statistical analysis (see dashed box in Fig. 4). Fundamentally, it can be stated that candles without fragrance additives produce lower emissions of combustion by-products than candles with fragrance. This can be attributed to the fact that many fragrance constituents exhibit a considerably more complex combustion chemistry than the fuels. Consequently, scented candles generate more VOCs than unscented candles. The individual VOCs measured during the course of this study like benzaldehyde, limonene, etc. are mostly fragrance ingredients that evaporate from the molten wax pool of the burning candle. Fragrance molecules that are

drawn up into the wick and exposed to the flame are decomposed by the combustion process, which leads to the production of carbon dioxide as well as combustion by-products and particles.

The great advantage of this study is that the contents of the candles are precisely known (see Supplementary Material). Although the exact precursor substances responsible for the formation of combustion by-products and particles could not be determined due to the large number of substances in the formulations, the experimental results and statistical analyses provide important information on the influence of the individual components on the emission behavior. These can be used to better describe and further optimize this cultural asset, which is of great importance for the social life of humans.

## CRedit authorship contribution statement

**Tunga Salthammer:** Conceptualization, Writing - original draft, Formal analysis, Validation, Visualization. **Jianwei Gu:** Writing - original draft, Methodology, Formal analysis. **Sebastian Wientzek:** Methodology, Validation, Investigation. **Rob Harrington:** Conceptualization, Writing - review & editing. **Stefan Thomann:** Conceptualization, Writing - review & editing, Investigation.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2021.106590>.

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