Emissions of soot, PAHs, ultrafine particles, NO\textsubscript{x}, and other health relevant compounds from stressed burning of candles in indoor air

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Abstract

Burning candles release a variety of pollutants to indoor air, some of which are of concern for human health. We studied emissions of particles and gases from the stressed burning of five types of pillar candles with different wax and wick compositions. The stressed burning was introduced by controlled fluctuating air velocities in a 21.6 m\textsuperscript{3} laboratory chamber. The aerosol physicochemical properties were measured both in well-mixed chamber air and directly above the candle flame with online and offline techniques. All candles showed different emission profiles over time with high repeatability among replicates. The particle mass emissions from stressed burning for all candle types were dominated by soot (black carbon; BC). The wax and wick composition strongly influenced emissions of BC, PM\textsubscript{2.5}, and particle-phase polycyclic aromatic hydrocarbons (PAHs), and to lower degree ultrafine particles, inorganic and organic carbon fraction of PM, but did not influence NO\textsubscript{x}, formaldehyde, and gas-phase PAHs. Measurements directly above the flame showed empirical evidence of short-lived strong emission peaks of soot particles. The results show the importance of including the entire burn time of candles in exposure assessments, as their emissions can vary strongly over time. Preventing stressed burning of candles can reduce exposure to pollutants in indoor air.

KEYWORDS
aerosol, airborne particles, black carbon, candle emissions, flickering candle flame, gaseous pollutants

1 | INTRODUCTION

Burning of candles is a common activity in many cultures around the world. It is associated with a cozy atmosphere and is commonly used in religious ceremonies. Christmas time in particular is a high season for candle burning. Burning candles emit particle and gas-phase pollutants. Examples of pollutants emitted by candle burning are aerosol soot particles (black carbon [BC]),\textsuperscript{1,3} ultrafine particles
Candle emissions have been associated with various adverse health effects in humans such as reduced cognitive abilities\textsuperscript{12} and negative changes in lung function.\textsuperscript{13} On the contrary, increased power of high-frequency spectral band of heart rate variability in healthy women has been reported after exposure to candle particles from stressed burn.\textsuperscript{14} Animal studies conducted on mice exposed to candle soot particles are associated with cytotoxicity and inflammation in the lungs\textsuperscript{15} as well as telomere shortening in the lung and spleen, a marker for the risk of developing cardiovascular diseases.\textsuperscript{16} In addition to being a major pollutant in indoor air, candle soot from various types of stressed burning has attracted a huge interest as a cheap emerging technology to produce functional nanomaterials with unique properties such as high hydrophobicity.\textsuperscript{17}

Candle emissions were found to be the primary source to the daily residential exposure to ultrafine particle number concentrations in a study of Danish homes.\textsuperscript{4} Wallace et al.\textsuperscript{18} measured personal exposure to ultrafine particles and found that for a typical nonsmoking suburban lifestyle, indoor exposure exceeds outdoor exposure to UFPs.

It is well-known that exposure to PAHs is of health relevance. The most toxic of the PAHs is benzo[a]pyrene (BaP), which is classified as a group 1 carcinogen by the International Agency for Research on Cancer.\textsuperscript{19} The United States Environmental Protection Agency has classified a group of 16 priority PAHs.\textsuperscript{20} Twelve of these have been assigned toxic equivalence factors based on their cancer potency relative to BaP.\textsuperscript{21} Alkylated PAHs are also important to include in emission studies since they reportedly are often more pervasive and more toxic than the parent PAHs.\textsuperscript{22} Thus, maximizing the number of PAHs included in studies is essential to increasing knowledge of emission and exposure scenarios as well as the associated risks. Moreover, the WHO has set an air quality guideline that estimates the unit risk of BaP level to be 0.12 ng m\textsuperscript{-3}, which will cause an excess lifetime cancer risk of 1/100 000.\textsuperscript{23}

Size distributions, total mass emission factors,\textsuperscript{24,25} and BC emission factors\textsuperscript{1,2} have been reported in a number of studies on candle emissions. Previous studies have also compared the candle soot morphology with other soot particle sources, and reported that candle soot particles showed a similar type of microstructure as the diesel exhaust particles, although the size of the soot aggregates was larger.\textsuperscript{26,27} Detailed soot formation inside candle flames has been studied.\textsuperscript{28,29} However, highly time-resolved emission data of soot, organic aerosol, PAHs, and inorganic aerosol in the emission zone near the candle are largely missing. The influence of the wax and wick composition on the formation of soot during stressed burn is not known.

It is important to estimate how candles contribute to soot levels in indoor environments, where we spend the majority of our time. This is especially important considering the health concerns related to the exposure to soot/BC.\textsuperscript{30} NO\textsubscript{x} is known to cause health effects in humans. NO\textsubscript{x} emissions from the steady burn of candles have previously been reported.\textsuperscript{9,11} However, the influence of the wax and wick composition on the NO\textsubscript{x} emissions has not been investigated.

The aim of this study is to investigate the pollutant emissions, including particles and gases from five types of pillar candles, during controlled stressed burn conditions in a chamber. Rasmussen et al.\textsuperscript{31} reported previously detailed descriptions of the wax and wick compositions of the same five types of pillar candles along with the emission characteristics under steady burn conditions. In this study, the emissions of the same five types of candles during controlled stressed burn conditions are compared, and further evaluated from the perspective of the potential impact that flickering candles may have on indoor air quality in specific emission scenarios.

### Practical implications

- The duration of burning and flame disturbance due to changing air velocities around the candle influence the level of emissions of airborne pollutants.
- Optimizing wax and wick composition can reduce emissions of soot (BC), PM\textsubscript{2.5}, and particle-phase polycyclic aromatic hydrocarbons (PAHs), but not emissions of NO\textsubscript{x}, formaldehyde, and gas-phase PAHs at stressed burn conditions.
- Preventing stressed burning of candles (flame disturbance) will reduce exposure to soot (BC), PM\textsubscript{2.5}, and particle-phase polycyclic aromatic hydrocarbons (PAHs).
- Introducing an international compliance standard for candle manufacturers that includes stressed burn emissions would enable users and retailers to make informed choices and would reduce exposure to airborne pollutants.

### 2 MATERIALS AND METHODS

#### 2.1 Experimental design

##### 2.1.1 Well-mixed emissions in a 21.6 m\textsuperscript{3} chamber

We measured the particle and gas emissions from five types of pillar candles (120 mm in height and 58–60 mm in diameter) that had different wax and wick combinations (denoted as candles 1–5) during stressed burning conditions (Table 1). All candles were white and non-scented. Three candles of one type were burned at a time for 3.5 h in each of the experiments in a 21.6 m\textsuperscript{3} stainless steel chamber with an air exchange rate (AER) of 2 h\textsuperscript{-1}.\textsuperscript{32} Two replicates were performed for each of the five candle types (ie, three candles of one type in each experiment, thus variability in six studied candles was captured). A rotating fan was applied to create a fluctuating air flow to make the candle flames flicker to create stressed burn conditions relevant for indoor environments. The length of the wicks was cut to...
1 cm before the experiments. The candles were lit with a gas lighter inside the chamber at the start of the experiment. After 3.5 h, the experiments ended and emissions during candle extinguishing were not included in these experiments. The extinguishing emissions were studied in complementary experiments “Above the flame” and are described below.

For comparison one steady burn experiment for candle type 1 was performed with an AER of 2 h⁻¹. In the steady burn experiment, the candle flames were not disturbed by fluctuating air velocities. Metal mesh cylinder shields (230 mm in diameter) were placed around each of the three candles. The fan was in this case positioned to mix the air in the chamber but not to disturb the flames. The air velocity in all experiments was measured with an air velocity meter (Swema-air 300), which was calibrated before the experiments.

The center of the rotating fan (AEG VL 5527 MS), in the stressed burn experiments, was placed at a distance of 160 cm from the candles with the neutral position in an angle of approximately 130 degrees. The three candles were placed on a table in a triangular shape with 10 cm between each of the candle edges. The velocimeter was placed behind the candles, Figure S1.

2.1.2 Measurements directly above the flame

Highly time-resolved measurements (1 s) above the flame of a single candle under similar stressed burn conditions were also performed. These were complementary measurements to gain mechanistic insight into the emission processes during stressed burn and during extinguishing the candles. In total 11 measurements were carried out: five of candle 1, three of candle 5, and one measurement each of candles 2, 3, and 4. A stainless steel ¼” sampling tube was placed approximately 4 cm above the flame. It was shifted approximately 1 cm to the side so as not to disturb the flame. An ejector dilutor (Model DI 100; Dekati; Finland) was used to dilute the emissions (1:8 times) before measurement. Highly time-resolved measurements above the flame were also carried out when the candles were extinguished with the snuffer. A snuffer is a device, that according to the candle manufacturer’s instructions, should be used to extinguish the candles by bending the wick and submerging it into the melted wax. In total, five measurements of the extinguishment were carried out: that is, one for each of the five candle types.

2.2 Emission characterization - equipment and analysis

2.2.1 Online characterization

A seven-wavelength aethalometer (model AE-33, Magee Sci.) was used for measurements of equivalent black carbon (eBC) mass concentrations. Equivalent black carbon is the terminology used for BC measurements with optical absorption methods. The eBC mass concentration was derived from the measured changes in attenuation through the filter at 880 nm using the default optical parameters used in the software.

Number concentration and particle size distributions were measured with a nano-Scanning Mobility Particle Sizer system (nanoSMPS) measuring in the range 2.4–79.1 nm (TSI, DMA 3085, CPC model 3776, TSI Inc.) and a long SMPS measuring in the range 18.8–914 nm (TSI, DMA 3081, CPC model 3775, TSI Inc.). Toward the end of the experiments, a catalytic stripper (CS) operating at 350°C and 1.5 lpm (Model CS 015, Catalytic Instruments GmbH) was introduced upstream the nano-SMPS to assess the volatile mass fraction of particles in the UFP size range. This allowed us to gather information on the composition of the UFPs emitted during stressed burn without the interference of the larger soot mode particles. Losses in the CS were accounted for using a size-dependent parameterization provided by the manufacturer. For larger particles (>60 nm), size-independent thermophoretic losses dominate (27% losses), as particle size decreases the losses increase due to the additional mechanism of diffusion. The combined losses from both mechanisms were as follows: 33% at 30 nm, 56% at 10 nm, and 86% at 5 nm.

A Soot Particle Aerosol Mass Spectrometer (SP-AMS) was used to measure the time-resolved emissions of refractory (dual vaporizer mode) and non-refractory (tungsten vaporizer mode) particles, previously described by Onasch et al. and DeCarlo et al. The SP-AMS measured 5 min in the dual vaporizer mode followed by 5 min in the tungsten vaporizer mode in the well-mixed chamber measurements. In the measurements above the flame, the dual vaporizer mode was used. In the above flame experiments, the SP-AMS was sampling in the FAST-MS mode, which allowed measurements with 1 s time resolution. Sampling was done in the closed mode (for background adjustments) for 5 s once per minute. The SP-AMS was calibrated using nebulized ammonium nitrate and carbon black (regal black, Cabbot Inc.) particles that were dried and then size selected to 300 nm using

<table>
<thead>
<tr>
<th>Candle No.</th>
<th>Wax material</th>
<th>Production method</th>
<th>Wick No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Animal stearin</td>
<td>Pressed</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>Paraffin (75%), Palm stearin (25%)</td>
<td>Fully refined paraffin, Pressed</td>
<td>2</td>
</tr>
<tr>
<td>3</td>
<td>Paraffin</td>
<td>Fully refined paraffin Molded, rustic looking</td>
<td>3</td>
</tr>
<tr>
<td>4</td>
<td>Palm stearin</td>
<td>Pressed</td>
<td>1</td>
</tr>
<tr>
<td>5</td>
<td>Paraffin</td>
<td>Fully refined paraffin Pressed</td>
<td>5</td>
</tr>
</tbody>
</table>
a DMA. Further details are provided in the Appendix S1. Phosphates were quantified as the sum of the phosphate-related ions PO$_4^{3-}$, PO$_2^{-}$, HPO$_4^{2-}$, HPO$_2^{-}$, PO$_3^{4-}$, PO$_3^{5-}$, and H$_2$PO$_4^{-}$.

NO and NO$_2$ were monitored with a chemiluminescence analyzer (CLD 700 AL, ECO PHYSICS AG). CO$_2$ was measured with a non-dispersive infrared CO$_2$ analyzer (LI-8020, LI-COR, Lincoln) and was below 1500 ppm. O$_3$ (OXROB3, FireStingO2, PyroScience) and temperature (Swema-air 300) in the chamber were also measured during all experiments. Sampling lines for online and offline measurements were situated in chamber air approximately 2 m from the burning candles.

All the above-listed instruments were used for particle characterization during the well-mixed emissions. For measurements above the flame, the SP-AMS and the NO$_x$ chemiluminescence analyzer were applied.

### 2.2.2 Offline characterization

Sampling for offline analysis was performed from 0.5 to 3.5 h in the experiment. Blank samples were collected in the chamber without burning candles.

Organic carbon (OC) and elemental carbon (EC) were determined on particles collected on quartz fiber filters with the use of a PM$_{2.5}$ pre-cyclone at a flow rate of 8.5 min$^{-1}$. Parallel sampling was used. This means that in one sampling line a quartz filter was used, while in the other line a quartz filter was placed behind a Teflon filter, which captured particles and allowed accounting for positive gas adsorption artifacts of OC on the quartz filter. The samples on the quartz filters were analyzed with a thermal optical analyzer (DRI Model 2001 OC/EC Carbon Analyzer, Atmoslytic Inc.) using the NIOSH NMAM 5040 diesel exhaust protocol. The EC limit of detection (LOD) was 0.06 µg C cm$^{-2}$.

Carbonyl compounds were sampled with 2,4-dinitrophenylhydrazine (DNPH) cartridges with a flow rate of 1 L min$^{-1}$ and subsequently analyzed with high-performance liquid chromatography (HPLC) for the following compounds: formaldehyde, acetaldehyde, acetone, acrolein, propionaldehyde, benzaldehyde, and butyraldehyde.

Samples were collected with a flow rate of 15 L min$^{-1}$ with two filters in parallel for subsequent analysis with ion chromatography for the following ions: Cl$^-$, NO$_3^{-}$, PO$_4^{3-}$, PO$_3^{4-}$, SO$_4^{2-}$, Na$^+$, NH$_4^{+}$, K$^+$, Ca$^{2+}$, and Mg$^{2+}$. The divalent cation Mg$^{2+}$ was not detected in the emissions from any of the candles. Further details are described in the Appendix S1.

The sampling equipment and analytical techniques used for PM$_{2.5}$, particle-associated PAHs, and gas-phase PAHs are thoroughly described elsewhere with some modifications.$^{36}$ In short, two sampling assemblies were used with a flow rate of 2 L min$^{-1}$. In the first assembly, PM$_{2.5}$ and associated PAHs were sampled on a filter (Teflo; Pall Corporation, 37 mm diameter, 2 µm pore size), behind a PM$_{2.5}$ cyclone pre-separator. In the second assembly, a solid adsorbent tube (XAD-II; 120 mg) collected gas-phase PAHs. A filter, the same type used for the PM$_{2.5}$ measurement, was placed in front of the adsorbent tube. This filter was not analyzed. After gravimetric analysis, the PM$_{2.5}$ filters were analyzed for their contents of particle PAHs, and the XAD-II tubes for gas-phase components. The concentrations of 33 PAHs, including 16 US EPA priority PAHs and 16 alkylated species, were determined. Further details are presented in the Appendix S1. All results were corrected for the blanks. The limit of detection (LOD) was calculated as three times the standard deviation of the values for the blanks or the background noise of these blanks. A certified reference material (SRM 1649a, urban dust) was used as quality control. The measured levels of 13 PAHs lie, for the most part, within 35% of the certified levels. Good precision was realized for the duplicates of both the gas-PAHs.

#### 2.2.3 Toxic equivalence factors for individual PAHs

The relative importance of individual PAH compounds for the total cancer risk of the total concentration was calculated with toxic equivalence factors (TEFs) obtained from Larsen and Larsen$^{26}$ of the 12 PAHs analyzed in this study. The compounds were ranked according to cancer potency relative to BaP (ie, BaP equivalents [BaP eqs]), with a TEF factor for BaP set to 1. Offline sampling and analyses were used for particle characterization only during the well-mixed emission experiments.

### 2.3 Data analysis

#### 2.3.1 Emission factor calculation

General equations describing how emission factors can be derived from the pollutant concentrations measured in indoor air, on the basis of mass balance equations for indoor environments, are given by Koutrakis et al.$^{37}$ and He et al.$^{38}$ Here, we used a simplified expression, Equation 1, where the emission factor per candle averaged over 3.5 or 3 h (see details in the Online and Offline characterization sections), $S_{av}$, was calculated as follows:

$$S_{av} = \frac{C_{av} \cdot V \cdot (a + k)}{n} \quad (1)$$

where $C_{av}$ is the average concentration over the time period (either number or mass concentration of the pollutant), $V$ is the chamber volume (m$^3$), $a$ is the air exchange rate (h$^{-1}$), $k$ is the loss rate due to deposition and coagulation (h$^{-1}$), and $n$ is the number of candles.

The air exchange rate in the chamber was controlled and set to 2 h$^{-1}$. The decay rate was determined on the basis of two decay experiments for candle types 4 and 5, following the stressed burn experiments after extinguishing the candles. The determined value of the loss rate ($k$) used for the calculation of the particle number emission factors was 2.4 h$^{-1}$ (strong coagulation effect), and for
the calculation of the particle mass emission factors 0.4 h\(^{-1}\) (mass is conserved upon coagulation so no coagulation effect). These values were used for calculating emission factors of all particle-phase characteristics. No loss rate was included to calculation of emission factors of gas-phase components.

3 | RESULTS AND DISCUSSION

3.1 | Time resolved measurement of well-mixed emissions in the chamber

The measured equivalent black carbon (eBC, measure of soot particle mass concentration) and the particle number concentrations (size interval 3–80 nm) over time are shown in Figure 1 and Figure S2. The presentation of the size interval 3–80 nm was motivated by the largest contribution within this size range to the total number concentration measured. The comparison of the five different candle types illustrates differing time emission profiles of eBC with relatively high repeatability of the profiles between the two replicates (three nominally identical candles in each experiment) for each candle type. The absolute values of the emitted mass concentrations of eBC over each of the two full experiments varied by on average \(\pm 20\%\) from the mean value for each candle type. This is a measure of the precision (repeatability) of the replicate experiments that includes both small variations in our ability to control the level of stress on the candle burning but also includes real differences caused by variations in emissions between candles of the same type.

A distinct variation between the initial first hour and the remaining time is evident for candles 1–4 (Figure 1 and Figure S2). Candles 1 and 2 show relatively high emissions of eBC in the initial half hour, followed by a decrease in the concentration. After burning the candles for 1.5–2 h, the concentrations again increase, particularly for candle 1. Candles 3 and 4 show opposite emission profiles, with candle 3 showing low eBC emissions in the start-phase, with increasing emissions over time, while candle 4 shows a peak emission of eBC in the start-up with strongly reduced emissions after the first 1–1.5 h. Candle 5 is the only one that shows an increasing eBC concentration over time, reaching "steady-state behavior". After 1–2 h, candle 5 reached by far highest eBC concentration measured in the chamber (21.6 m\(^3\), AER = 2 h\(^{-1}\) ~450 µg m\(^{-3}\)) (Figure 1), while the highest concentration resulting from burning of the remaining candles was ~80 µg m\(^{-3}\) (candle 2). The breaks in the eBC time trace for candle 5 are due to frequent filter changes in the instrument caused by the high chamber concentrations. The decreased concentration after each filter change in the later part of experiment 5.1 may be due to uncertainties in the filter-loading correction in the instrument caused by the high eBC concentrations.

There was no clear correlation between burn rate and eBC emissions (Table 2). The reproducible and strongly varying time trends in eBC emissions between the candles may be explained by variations in wick-length and flame height over time, properties that are partly controlled by the selected wax and wick combinations. In addition, a rim may develop near the candle edges which can affect the combustion conditions and the impacts of surrounding air currents on the flame stability. It is expected that soot emissions may vary more in the period early after igniting the candles when a fresh wick is ignited and the liquid fuel pool develops over time. To shed more light on the mechanisms of the soot emission, data measured directly over the flame with high time resolution are described below.

![Figure 1](https://via.placeholder.com/150)

**Figure 1** Examples of time traces of eBC in µg m\(^{-3}\) (black) and number concentration in particles cm\(^{-3}\) (gray) of particles in the size range from 3 to 80 nm for stressed burn experiments with candles 1, 3, 4, and 5. Three candles of each type were used in each experiment. All ten experiments including replicates can be seen in Figure S2. The breaks in the eBC time trace for candle 5 are due to frequent filter change in the instrument caused by the high chamber concentrations.
The number concentrations measured in the chamber also showed high repeatability between replicates both in emission patterns and in concentrations (particles/cm³) (Figure S2). All candles, except from candle 3, showed peak emissions when the candles were lit, whereas candle 3 showed more stable emissions, with a slight increase in particle number concentration over time.

It is worth noting that a relatively stable number concentration is reached even faster than expected from the air exchange rate (90% of the steady-state value reached after 30 min assuming constant emission level and $a + k = 2.4 \text{ h}^{-1}$). This could either be due to strongly increased emissions in the early part of the experiment, or to coagulation rapidly becoming a dominating loss mechanism of the particle number as the concentration reaches above $\sim 2 \times 10^6$ particles cm$^{-3}$, which was described by Wallace et al.\textsuperscript{5}

Candle 5 showed a peak in number concentration ($1-2 \times 10^6$ #/cm$^3$) in the initial half hour after igniting the candles, though with a lower number concentration than the other candles ($2-3 \times 10^5$ #/cm$^3$). This was followed by a strong decrease in number concentration and a simultaneous strong increase in soot particle (eBC) mass concentration. The increased emission of soot particles increased the probability of coagulation between soot particles and UFPs, which in turn reduced the measured particle number concentration.

A comparison of eBC concentrations with the AMS time traces (Figure S3) was carried out. It measured the time-resolved mass concentration of organic aerosol (OA) and inorganic aerosol particle mass concentration in tungsten vaporizer model and showed that the mass concentration for all candles during stressed burn was dominated by eBC, while the organic and inorganic mass concentrations were significantly lower. The organic mass concentration was 15–25 times lower than that for eBC. The time traces of eBC, OA, and inorganic components, measured with two different methods (aethalometer AE33 and HR-AMS), showed the same trend in emission profiles of eBC and OA when comparing each of the individual candle types. However, the inorganic particle components did not always follow the same emission profile (Figure 1 and Figure S3).

The above-described variations in concentrations over time illustrate the importance of including the entire burn time when

TABLE 2 Emission factors and burn rate per candle based on concentrations averaged over 3.5h (0–3.5 h of the experiments) based on online measurements

<table>
<thead>
<tr>
<th>PNC$^a$ (2.4–914 nm)</th>
<th>eBC</th>
<th>NO</th>
<th>NO$_2$</th>
<th>Burn rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>#/h</td>
<td>µg h$^{-1}$</td>
<td>mg h$^{-1}$</td>
<td>mg h$^{-1}$</td>
<td>g h$^{-1}$</td>
</tr>
<tr>
<td>Candle 1</td>
<td>9.3 $\times$ 10$^{13}$ (±0.2 $\times$ 10$^{13}$)</td>
<td>408 (±0.50)</td>
<td>2.8 (±0.2)</td>
<td>1.0 (±0.3)</td>
</tr>
<tr>
<td>Candle 2</td>
<td>9.0 $\times$ 10$^{13}$ (±0.4 $\times$ 10$^{13}$)</td>
<td>634 (±0.5)</td>
<td>4.1 (±0.2)</td>
<td>1.3 (±0.3)</td>
</tr>
<tr>
<td>Candle 3</td>
<td>6.3 $\times$ 10$^{13}$ (±0.3 $\times$ 10$^{13}$)</td>
<td>329 (±0.59)</td>
<td>3.4 (±0.2)</td>
<td>1.1 (±0.1)</td>
</tr>
<tr>
<td>Candle 4</td>
<td>8.5 $\times$ 10$^{13}$ (±0.35 $\times$ 10$^{13}$)</td>
<td>160 (±0.68)</td>
<td>3.2 (±0.3)</td>
<td>1.1 (±0.1)</td>
</tr>
<tr>
<td>Candle 5</td>
<td>1.5 $\times$ 10$^{13}$ (±0)</td>
<td>4346 (±874)</td>
<td>4.7 (±0)</td>
<td>1.5 (±0)</td>
</tr>
<tr>
<td>Steady$^b$</td>
<td>8.3 $\times$ 10$^{13}$</td>
<td>2.6</td>
<td>2.8</td>
<td>0.6</td>
</tr>
</tbody>
</table>

Average values are given together with range of two replicates (three candles each) in brackets. Values for burn rate represent average and standard deviation (given as ±) of 6 candles used in two replicate experiments.

$^a$Emission factors of PNC based on 3 h average concentrations (ie, 0–3 h of the experiments), due to use of the catalytic stripper in the last 30 min of the experiment.

$^b$One steady burn experiment for candle 1

TABLE 3 Emission factors of OC, EC, PM$_{2.5}$ (gravimetric analysis) and total particle inorganics

<table>
<thead>
<tr>
<th>OC</th>
<th>EC</th>
<th>PM$_{2.5}$</th>
<th>Inorg</th>
</tr>
</thead>
<tbody>
<tr>
<td>µg h$^{-1}$</td>
<td>µg h$^{-1}$</td>
<td>µg h$^{-1}$</td>
<td>µg h$^{-1}$</td>
</tr>
<tr>
<td>Candle 1</td>
<td>88 (±4)</td>
<td>328 (±53)</td>
<td>283 (±53)</td>
</tr>
<tr>
<td>Candle 2</td>
<td>106 (±16)</td>
<td>382 (±52)</td>
<td>510 (±61)</td>
</tr>
<tr>
<td>Candle 3</td>
<td>140 (±35)</td>
<td>252 (±67)</td>
<td>393 (±57)</td>
</tr>
<tr>
<td>Candle 4</td>
<td>46 (±9)</td>
<td>307 (±11)</td>
<td>330 (±15)</td>
</tr>
<tr>
<td>Candle 5</td>
<td>232 (±143)</td>
<td>3132 (±2145)</td>
<td>3038 (±1691)</td>
</tr>
<tr>
<td>Steady$^c$</td>
<td>134</td>
<td>&lt;DL</td>
<td>&lt;DL</td>
</tr>
</tbody>
</table>

All data are averaged over 3 h (0.5–3.5 h of the experiments) and reported per candle based on off-line analysis.

Average values are given together with range of two replicates (three candles each) in brackets.

<DL indicates that the concentration was below the detection limit

$^c$Possibly underestimated value, because the peak emissions occurring within the first half hour are not included (sampling 0.5–3 h).

$^d$Value based on single measurement.

$^e$One steady burn experiment for candle 1
evaluating the particle emissions from candles, and that the chosen burn time will influence the average emissions reported.

3.2 | Emission factors

Table 2 summarizes the calculated emission factors (EFs) based on the 3.5 h average concentrations of the parameters measured with online techniques for each of the candle types, presented as emissions per candle. The emission factors of OC, EC, PM$_{2.5}$, formaldehyde (CH$_2$O), and PAHs measured with offline techniques were averaged from 0.5 to 3.5 h and are presented in Tables 3 and 4.

Candle 5 shows the highest EFs of eBC, EC, PM$_{2.5}$, particle-phase PAHs, and NO$_x$ (see Tables 2, 3, and 4). The EF of UFP (2.4–100 nm) were of similar order of magnitude for all candles (ie, 10$^{12}$ particles per h); however, emissions for candle 5 (1.3 x 10$^{13}$ particles per h) were 4 to 6 times lower than for other candles (6.3 x 10$^{12}$–9.3 x 10$^{13}$ particles per h) (Table S1), which is in line with the result discussed above. It is important to mention that during steady burn candle 5, as reported by Rasmussen et al.31 also displayed the lowest UFP number emissions among candles 1–5. However, the same candle 5 has the by far highest eBC emission. This illustrates how important it is to evaluate candles at both steady and stressed burn conditions and assess different emission characteristics (ie, not only UFP). Candles 1–4 showed more similar magnitudes of EFs. Candle 4 showed slightly lower EFs for PM$_{2.5}$, eBC, and PAHs in the particle-phase, while it has the overall highest EF of gas-phase PAHs among all candle types, see Tables 2, 3, and 4.

3.3 | Soot measured as EC and eBC

Comparison of OC and EC results (Table 3) shows that the emissions from all candles were dominated by EC during flickering/stressed burn. The exception is candle 4, where OC and EC concentrations are low compared to all other candles. The initial half hour was not included in the OC/EC sampling. EC and eBC were highly correlated (Figure S4) suggesting that any of them can be used as proxies for each other when measuring stressed burn emissions from candles (slope 1.38, r$^2$ = 0.93; Figure S4). As shown in the time-resolved eBC measurement (Figure 1), soot emissions were almost exclusively taking place during the first hour for candle 4, which suggests that the lack of data for the first 30 min of the experiment caused substantial underestimation of the EC value. Consequently, when comparing soot emissions from different candle types, the values of eBC are used in this paper because they capture the entire burn period and all peak contributions during the experiments. For comparison, a steady burn experiment of candle 1 was included. The emissions of eBC were low, about 60 to 1700 times lower than at stressed burn of the five candle types.

The literature data on EC emissions during stressed burn of candles in laboratory studies range from 4.5 to 24.1 mg h$^{-1}$.32 The highest value in our study was for candle 5 (3.1 mg h$^{-1}$) and the lowest value for candle 4 (0.03 mg h$^{-1}$). Thus, the EC emission factors found at the degree of stress used in this study are lower than those found in the literature. However, the literature data are for tapered candles that may have higher EC emissions compared to pillar candles at a given stress level, for example, due to longer wicks. Clearly, more data are needed for candle emissions from stressed burn. Especially data for the same candle type used both during controlled stress levels in the laboratory and under real-world conditions would be useful.

Currently, there are no international standards regulating candles manufacturing and sales that would include the emissions of airborne pollutants at the stressed burn of candles. Within the European Union, there are several certification schemes for candle manufacturers, which use a sooting index of candles.39 However, these emission assessments are conducted at steady burn conditions with a metal mesh shield around the candle preventing disturbance of the candle flame. Including assessments of the stressed

<p>| TABLE 4 | Emission factors of formaldehyde (CH$_2$O), PAHs, and benzo[a]pyrene equivalent (BaP eq.) |
|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|</p>
<table>
<thead>
<tr>
<th></th>
<th>CH$_2$O</th>
<th>PAH particle</th>
<th>PAH gas</th>
<th>BaP eq. particle</th>
<th>BaP eq. gas</th>
<th>BaP eq. Gas + particle</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>µg h$^{-1}$</td>
<td>ng h$^{-1}$</td>
<td>ng h$^{-1}$</td>
<td>ng h$^{-1}$</td>
<td>ng h$^{-1}$</td>
<td>ng h$^{-1}$</td>
</tr>
<tr>
<td>Candle 1</td>
<td>54 (±4.5)</td>
<td>90 (±13.0)</td>
<td>416 (±289.0)</td>
<td>0.79 (±0.11)</td>
<td>0.11 (±0.025)</td>
<td>0.91 (±0.14)</td>
</tr>
<tr>
<td>Candle 2</td>
<td>46 (±6.5)</td>
<td>90 (±16.0)</td>
<td>600 (±30.0)</td>
<td>0.78 (±0.07)</td>
<td>0.13 (±0.025)</td>
<td>0.94 (±0.10)</td>
</tr>
<tr>
<td>Candle 3</td>
<td>43 (±0.5)</td>
<td>59 (±11.5)</td>
<td>591 (±29.5)</td>
<td>0.44 (±0.07)</td>
<td>0.23 (±0.075)</td>
<td>0.67 (±0.14)</td>
</tr>
<tr>
<td>Candle 4</td>
<td>49 (±3.0)</td>
<td>25 (±12.5)</td>
<td>1043$^b$</td>
<td>0.09 (±0.03)</td>
<td>0.16$^b$</td>
<td>0.27$^{ab}$</td>
</tr>
<tr>
<td>Candle 5</td>
<td>39 (±3.0)</td>
<td>578 (±254.5)</td>
<td>557 (±100.5)</td>
<td>3.86 (±1.95)</td>
<td>0.28 (±0.06)</td>
<td>4.13 (±2.01)</td>
</tr>
<tr>
<td>Steady$^c$</td>
<td>35</td>
<td>26</td>
<td>846</td>
<td>&lt;DL</td>
<td>0.11</td>
<td>0.11</td>
</tr>
</tbody>
</table>

PAHs and BaP eq. emission factors are given separately for the particle and gas-phase. All data are averaged over 3 h (0.5–3.5 h of the experiments) and reported per candle based on off-line analysis. Average values are given together with range of two replicates (three candles each) in brackets. <DL indicates that the concentration was below the detection limit.

Possibly underestimated value, because the peak emissions occurring within the first half hour are not included (sampling 0.5–3 h).

Value based on single measurement.

One steady burn experiment for candle 1.
burn emissions, which mimic real-life situations, would allow users to make informed choices, as well as reduce the emissions of airborne pollutants.

3.4 | PAHs and benzo[a]pyrene (BaP) equivalent (BaP eq)

The particle-phase PAH emissions were low in comparison with gas-phase PAH emissions, except from candle 5, which shows the highest particle-phase emissions and is comparable to gas-phase PAHs emissions (Table 4). However, it has to be noted that the particle-phase contribution to the BaP eq concentration is higher than from gas-phase PAHs.

During the steady burn experiment, the particle PAH emissions were low and comparable to the lowest emissions during stressed burn (candle 4). However, emissions of gas-phase PAHs during the steady burn were relatively high and comparable to the highest emissions of gas-phase PAHs during stressed burn for candle 4.

The benzo[a]pyrene equivalent (BaP eq) concentrations of the particle and gas-phase PAHs were calculated according to standard methods and are provided as EFs in Table 4. During stressed burn, the candles show total BaP eq EFs from 0.27 to 4.13 ng h$^{-1}$, with candle 5 having the highest EF and candle 4 the lowest. Candles 1–3 show similar BaP eq EFs of 0.67–0.94 ng h$^{-1}$. Particle-phase PAH emissions are the main contributors to the BaP eq emissions. All five candles show similar BaP eq EFs for the gas-phase PAH emissions. Gas-phase PAH emissions were not influenced to a big degree by wax and wick composition or type of burning (stressed or steady burn). It is important to include both particle- and gas-phase PAHs to assess the risks and toxicities of PAHs in emissions in environmental studies.

Polycyclic aromatic hydrocarbons were sampled from 0.5 to 3.5 h. Since candle 4 showed the highest eBC emission in the initial 0–1 h (Figure 1), the chosen sampling time will likely cause an underestimation of particle-phase PAH emissions from candle 4. Thus, the particle-phase PAH emissions are consistent with the trends observed for eBC EFs where candle 5 showed by far the highest emissions, candle 4 the lowest, and candles 1–3 intermediate emissions.

3.5 | NO$_x$

The EFs of NO$_x$ show similar levels for candles 1–4, while candle 5 shows slightly higher NO$_x$ EFs (Table 2). In contrast to the eBC emissions, NO$_x$ emissions for all candles during stressed burn showed increasing concentrations over the first hours and then reached a steady-state behavior (example for candle 3 showed in Figure S5). This is expected for the air exchange rate used and a roughly constant emission factor over time. The similarity in NO$_x$ emissions is consistent with previous reported NO$_x$ emissions from the steady burn of candles. Derudi et al$^9$ measured NO$_x$ emissions (1.7–2.5 mg h$^{-1}$) from the steady burn of paraffin candles of different wax refinements. Klosterköther et al$^{11}$ reported values of NO (2.7–3 mg h$^{-1}$) and NO$_2$ (1.8–2.1 mg h$^{-1}$) for the steady burn of paraffin containers candles and paraffin church candles. Thus, the slight variation in NO$_x$ emissions may be associated with increased flame height and a higher flame temperature.

The NO$_x$ EFs from the steady burn experiment of candle 1 are provided in Table 2. The steady burn NO and NO$_2$ EFs are similar to those of the stressed burn of candle 1. The NO values were 2.8 mg h$^{-1}$ for steady and stressed burn. The NO$_2$ values were 0.6 and 1.0 mg h$^{-1}$, respectively, for steady and stressed burn. Thus, NO$_x$ emissions did not show dependence on burning mode of the candles nor the wax and wick composition.

3.6 | Carbonyl compounds

The EFs of formaldehyde for the time period 0.5–3.5 h are presented in Table 4. The measured concentrations of formaldehyde and other carbonyl compounds (acetaldehyde, acetone, acrolein, propionaldehyde, benzaldehyde, and butyraldehyde) are shown in Table S2 for the same time period. Acrolein was not detected in the emissions from any of the candles. The five different candle types show very similar concentrations of emitted carbonyl compounds with their sum ranging from 7.18 ± 0.18 for µg m$^{-3}$ candle 2 to 9.32 ± 0.11 µg m$^{-3}$ for candle 1. The low emissions were expected, since aldehyde and carbonyl emissions primarily are related to the addition of fragrances or dyes in the candles.$^3$ These results indicate that emissions of VOCs from the stressed burning of candles are minor compared to the particle emissions.

3.7 | Inorganic particle emissions

The total inorganic particle EFs are presented in Table 3. The summarized inorganic emissions could explain between -2% (candle 5) and 31% (candle 1) of PM$_{2.5}$. This is substantially lower than for steady burn when PM emissions are commonly dominated by inorganic emissions.$^3$ The two paraffin candles, 3 and 5, show lower emitted mass concentrations of inorganic ions (Table 3) compared to candles 1, 2, and 4. This may be caused by less impregnation (eg, flame retardants) needed to protect the wick against the paraffin wax (candles 3 and 5) relative to stearin wax (candles 1, 2, and 4 [Table 1]). Notably, candle 5 is the only one, where the anions are dominated by nitrate and the cations by sodium (Table S3). All other candles show anion emissions dominated by phosphates. The inorganic PM from ion chromatography correlated relatively poorly with EC ($r^2 = 0.55$; Figure S4).

The sum of the inorganic emissions, OC, and EC is higher than PM$_{2.5}$ emissions for all candles except candle 4 where the opposite is observed. The observed differences can be explained by method uncertainties, possible evaporation of some of the OC before PM$_{2.5}$ analysis, and experimental uncertainties/difficulties with filter sampling.
The AMS has a limitation in measuring these particle sizes, because of soot mode being detected with much higher efficiency. On the mixing state, with inorganic components present in the larger range (assuming unity particle density of both treated and untreated emission). The particle mass fraction (assuming unity particle density of both treated and untreated emissions) surviving the CS after the loss correction was calculated in the size range 2.4–60 nm. 60 nm was chosen as the upper limit to reduce the influence of the soot mode on the heat-treated samples. The results from candles 1–4 showed that the majority of the particle mass was removed by the CS. The fraction of assessed UFP mass (2.4–60 nm) that survived the treatment was 10% for candle 1 (8% in steady burn experiment), 35% for candle 2, 23% for candle 3, and 14% for candle 4. This suggests that less than 10%–35% of the UFP mass consists of EC or other refractory species during stressed burn.

Ammonium phosphate salts (the dominating inorganic component of stressed burn emissions) and organic aerosol are readily removed by heating at this temperature. Compared to the inorganic composition (Table S3), candles 2 and 3 with the higher remaining mass fraction after the CS also contained K+ and Na+ cations in addition to NH4+. These may constitute alkali salts that are not volatile at 350°C (eg, potassium phosphate). For candle 5, the situation was different with 83% of the assessed mass surviving the CS. In this case, the soot mode also extended down below 60 nm which may explain the high mass fraction that survives the CS. Additionally, NaNO3, a major contributor to the inorganic emissions for candle 5, decomposed at 380°C and may not have been fully removed from the particles in the CS.

In summary, candles emit high number concentrations of particles in the UFP size range during both steady and stressed burn. Major components of this particle type are water-soluble inorganic components such as phosphates, sulfates, and nitrates. This particle type contains little, if any EC/eBC. The water-soluble nature of these particles will affect the lung deposition probability in the respiratory tract. Additionally, candles emit a second type of particles during stressed burn which have larger sizes and are dominated by soot detected as eBC/EC. This particle type dominated the mass emissions during stressed burn. This study showed that by varying the wax and wick combination, the composition and emission level under stressed burn conditions of both these particle types can be altered.
**Figure 2** Number and mass size distributions of candles 1, 3, and 5 during stressed burn and one experiment for candle 1 during steady burn. The two replicates for each case are shown. The results are based on measurements with nano-SMPS and regular SMPS (2.4–914 nm). Note the different y-axis scale in the case of candle 5 (particle number concentration) and for candle 5 and steady burn of candle 1 (particle mass concentration). Number and mass size distributions for candles 2 and 4 during stressed burn can be seen in Figure S6.

**Figure 3** AMS time trace of emissions of organic aerosol (OA), rBC, and polycyclic aromatic hydrocarbons (PAHs), as well as NOx and NO2 time traces. Example of candle 5 above the flame measurements. The graph shows two strong soot (rBC) peaks upon stressing of the candle flame. The graph supports the coemission of OA, and PAHs in the soot peaks, while NOx do not follow a similar pattern. The pie charts show the percentage distribution of mass concentrations of rBC, OA, and PAHs in each of the two main emission peaks. The upper pie chart represents the first peak (around 14:16 h) and the lower pie chart the second peak (around 14:17 h).
3.9 Above flame measurements and correlations between pollutants

Measurements with high time resolution were carried out a few cm above slightly to the side of the flame during both stressed burn and upon extinguishing of the candle. The aim was to gain improved understanding of the particle emission dynamics and pollutant formation mechanisms. Figure 3 shows measurements for candle 5 directly above the flame during stressed burn measured with the SP-AMS in dual vaporizer mode with 1 Hz time resolution. The time-resolved data show that the particle emissions were dominated by refractory black carbon (rBC). Organic aerosol (OA) and particle-phase PAHs were co-emitted with rBC in short-lived strong peaks from the flickering candle flame. Similar results were found for other candles (not shown). During stressed burn, rBC constituted about 94% of the sum of rBC, OA, and PAHs. NOx shows a completely different emission pattern, and no clear correlation with rBC, OA, and PAHs is evident.

Figure 4 shows a time-resolved AMS measurement above the flame when candle 2 was extinguished by use of a snuffer. Figure 4 shows a short-lived peak of strongly increased OA, PAHs, and rBC emissions. The particle composition was in this case completely dominated by OA, which contributed to 79% of the sum of OA, rBC, and PAHs. This was completely different compared to the emission peaks during stressed burn.

The correlations between rBC, OA, and PAH emissions measured above the flame from extinguish emissions are compared to stressed burn emissions in Figure 5. The graph indicates a correlation between OA, PAHs, and rBC both for extinguishing (OA vs. rBC slope = 3.44, \( r^2 = 0.86 \)) and for stressed burning (OA vs. rBC slope = 0.06 and \( r^2 = 0.88 \)). The co-emission of rBC, OA, and PAHs is further supported by the linear correlation between PAHs, OA, and \( C_{13}^+ \) (marker for rBC) in Figure S7 for candles 2–5 in well-mixed air in the large chamber. The correlation is in this case very high for rBC (\( C_{13}^+ \)) versus OA (slope = 9.2, \( r^2 = 0.97 \)) and high also for PAHs versus rBC (\( C_{13}^+ \); slope = 90.7, \( r^2 = 0.93 \)).

We interpret the observed emissions from stressed burn in the following way: During normal steady candle burning, the candle wax first evaporates and the volatile organics are then thermochemically converted in several reaction steps. At lower temperatures, the wax molecules are first degraded to smaller constituents. At increased temperatures, aromatic rings form that may grow to PAHs, for example, and eventually further contribute to soot (rBC) formation.\(^{42\text{a}}\) It may be that during steady burn almost all larger (particle-phase) PAHs are converted to rBC. rBC is later removed in the high-temperature oxidation zones at the outer edges of the candle flame where the hot flame meets oxygen from room air. In these regions, NOx can be expected to form. Thus, steady burn leads to very low emissions of particle-phase PAHs and rBC, but the emissions of NOx may be considerable.

When the candle flame is cooled down by the surrounding air flow (flickering) during stressed burn, the oxidation of the soot particles formed in the candle flame slows down, hence the residence time in the flame is not sufficient for full oxidation/removal, and the soot particles can instead be emitted from the flame to room air. In extreme cases, this phenomenon is visible as black smoke. OA and PAHs are also emitted as a consequence of the overall cooling of the flame, though in much lower quantities than rBC particles.

If the flame quenching during stressed burn is more extensive, it may reach a region where PAHs to rBC conversion is less progressed, thus resulting in a higher PAH/rBC ratio of the emissions. Our results are in line with Liang et al.\(^{43\text{a}}\) who reported a higher content of hydrocarbons in inner flame soot particles compared to flame tip soot, which constituted elemental carbon and ash components.

These results indicate, that in time-resolved measurements, above the flame and in the well-mixed chamber measurements, with use of AMS, the peak increases of one of the parameters—rBC, OA, or PAHs—may be used as a proxy for the emissions of the two other parameters for stressed burn.

The HR organic mass spectra measured above the flame for candle 2 during stressed burn and extinguishing are shown in Figure 6. Both spectra are dominated by CH fragments. The high contribution from CH fragments to the OA may be from aliphatic carbon chains from unburned- or thermochemically converted wax, in accordance with Fine et al.\(^{25\text{a}}\) The extinguishing smoke in Figure 6 shows clear signal at m/z 60 (\( C_6 H_{12} O_2^+ \)) and m/z 73 (\( C_7 H_{16} O_2^+ \)), which commonly are associated with anhydrous sugars, such as levoglucosan, emitted from the pyrolysis of cellulose in biomass and are marker ions for wood smoke.\(^{44\text{a}}\) The wicks are made of cotton, a biomass consisting of almost pure cellulose. These ions may also have contributions from carboxylic acids,\(^{44\text{a}}\) and have been found in the frying of meat with a content of fatty acids and other cooking events.\(^{45,46\text{a}}\)
Our results suggest that organic aerosol emissions from candle extinguishing may contain pyrolysis products from cotton in the wick in addition to the evaporated and pyrolyzed wax components that were previously reported by Fine et al.\textsuperscript{25}

3.10 | Estimated concentrations from stressed burning of candles in a 90 m\textsuperscript{2} apartment on the basis of EFs

Table 5 provides estimated concentrations of PM\textsubscript{2.5}, EC, NO\textsubscript{2}, and particle- and gas-phase PAHs after burning 5 candles for 3.5 h at stressed burn conditions in a 90 m\textsuperscript{2} apartment with all doors open and a ceiling height of 2.4 m. An air exchange rate of 0.5 h\textsuperscript{-1} was assumed, and a loss rate of 0.5 h\textsuperscript{-1} was used attributed to losses due to deposition onto surfaces in indoor environments. No loss rate was applied to the NO\textsubscript{2} concentrations and gas-phase PAH (PAH\textsubscript{gas}). The highest (candle 5) and lowest (candle 4) emitting candles with respect to eBC were used for the estimation as examples of high and low emission scenarios.

In the high emission scenario, the PM\textsubscript{2.5} and NO\textsubscript{2} concentrations would reach 70 and 38 µg m\textsuperscript{-3}, respectively, and in the low emissions scenario 8 and 26 µg m\textsuperscript{-3}, respectively. The WHO air quality guidelines for PM\textsubscript{2.5} are 25 µg m\textsuperscript{-3} as 24 h average and 10 µg m\textsuperscript{-3} as a yearly average. For NO\textsubscript{2}, the guidelines are 200 µg m\textsuperscript{-3} as an hourly average and 40 µg m\textsuperscript{-3} as a yearly average.\textsuperscript{47} The high emission scenario thereby exceeds the concentration of 25 µg m\textsuperscript{-3} of PM\textsubscript{2.5} in the

Table 5

<table>
<thead>
<tr>
<th></th>
<th>PM\textsubscript{2.5} (µg m\textsuperscript{-3})</th>
<th>EC (µg m\textsuperscript{-3})</th>
<th>NO\textsubscript{2} (µg m\textsuperscript{-3})</th>
<th>PAH\textsubscript{particle} (ng m\textsuperscript{-3})</th>
<th>PAH\textsubscript{gas} (ng m\textsuperscript{-3})</th>
<th>PAH\textsubscript{particle+gas} (ng m\textsuperscript{-3})</th>
<th>BaP eq\textsubscript{gas} (ng m\textsuperscript{-3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Low emissions</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(candle 4)</td>
<td>8</td>
<td>0.8</td>
<td>26</td>
<td>0.6</td>
<td>24.1</td>
<td>24.7</td>
<td>0.01</td>
</tr>
<tr>
<td>High emissions</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>(candle 5)</td>
<td>70</td>
<td>72</td>
<td>38</td>
<td>13.4</td>
<td>12.9</td>
<td>26.3</td>
<td>0.10</td>
</tr>
</tbody>
</table>

No losses were applied in the estimation of NO\textsubscript{2} and gas-phase PAH (PAH\textsubscript{gas}). The two scenarios represent the minimum (candle 4) and maximum (candle 5) emissions with respect to eBC.
hours of candle burning, but as a single source, it does not exceed the 24 h 25 µg m\(^{-3}\) average in the given scenario.

It should be pointed out that burning candles are not the only possible source of particles indoors. It is just one type of indoor source that contributes to the existing levels indoors, which depends on the infiltration of outdoor particles (which in turn depends on the outdoor concentrations, the building envelope, ventilation, and type of airing practices) and contributions from other indoor sources such as cooking, smoking, and use of cleaning products. Average indoor PM\(_{2.5}\) concentrations reported in Scandinavia range from 9.2 to 12.2 µg m\(^{-3}\).\(^{48-53}\) Therefore, burning candles especially in the case of the high emission scenario represent a significant contribution to exposure to particles indoors. Candle burning is an episodic event that generates high peak concentrations. How frequently these peaks occur (ie, how frequently candles are burned) depends on the occupants’ habits and vary to a high degree. However, the duration of elevated concentrations after the candle burning ceases, depends on the ventilation and airing practices, and in cases with low air exchange rates, the elevated concentrations were observed 12.5 h after the indoor source ceased.\(^{54}\)

The NO\(_2\) concentrations in both scenarios are well below the hourly average of 200 µg m\(^{-3}\). However, both scenarios would contribute to NO\(_x\) exposures.

Elemental carbon concentrations were estimated to reach 0.8 and 72 µg m\(^{-3}\) in the two scenarios. For comparison, the highest average personal BC exposures in Stockholm during transport have been measured to 2.7 µg m\(^{-3}\) during bus transportation.\(^{55}\) Thus, the high emission scenario would especially add to peak exposures of BC.

The highest air PAH concentrations, a total of 16 US EPA PAHs, obtained for the two scenarios (Table 5) are equal to those reported for urban outdoor and indoor air in Gothenburg, Sweden\(^{56,57}\) and indoor in a rural area where wood combustion for domestic heating is common.\(^{58}\) and moreover, in restaurant kitchens in Sweden.\(^{59}\) However, the calculated levels of BaP levels in both cases are below the level set by the WHO as an air quality guideline (0.1 ng m\(^{-3}\)).\(^{22}\) The European Community has suggested the BaP eq. on PM\(_{10}\) at an annual standard in ambient air of 1 ng m\(^{-3}\).\(^{60}\) The cancer potency values in this study were well below those stipulated values.

The concentrations provided in Table 5 were estimated on the basis of conditions that were considered possible to occur in Scandinavian homes. The estimated concentrations are both dependent on the number of candles burned, the volume of the apartment, the air exchange rate, and the level of stressed burn. Burning only one candle for 3.5 h at the conditions described above would thus result in five times lower emissions than those presented in Table 5. However, situations of burning more than five candles or burning candles in significantly smaller rooms (eg, in the bathroom) are also common and would generate higher concentrations than presented in Table 5. In restaurants, the burning of one or more candles per table in volumes that are comparable or smaller than the volumes described above are not unusual. Furthermore, the air movements by door opening, people passing by and talking in near proximity to the candles are likely to create stressed burning conditions.

### 3.11 Limitations

Offline sampling was performed from 0.5 h into the experiment and did not capture elevated emissions within the first 30 min in the case of some candle types. Because of this, the presented emission factors based on offline analysis are underestimated. We have studied only five different types of pillar candles, with specific wax and Wick composition provided by the manufacturers involved in the study. Thus, our study captures only a fraction of a vast variety of candle types available on the market regarding wax and Wick composition and shapes. The degree of stress candles are exposed to from ambient air movements will vary in different environments. We used one level of stressed burn in this study. This emission factors of EC in this study were in between those found for steady burn and stressed burn in previous studies. However, a direct comparison cannot be made because the emissions are also affected by the candle shape. Different candle shapes (eg, tapered candles) have been used in most of the previous studies. In addition, in the above flame measurements, only one measurement was made for candles 2, 3, and 4, making uncertainty estimation not possible.

### 4 CONCLUSIONS

The aerosol particle emissions during stressed burn were investigated for five types of pillar candles with different wax-wick composition. For all five candle types, the particle mass emissions were dominated by eBC (EC), with minor contributions from inorganic and organic carbon fraction of PM. The five types of candles showed variations in emission factors of PM\(_{2.5}\), eBC, and particle PAHs as well as strong variations in the emission time profiles, when burned at similar stressed burn conditions. This highlights the importance of the combination of wax and Wick composition to obtain the lowest possible emissions of eBC, PM\(_{2.5}\) and particle-phase PAHs. It also highlights the importance of including the entire burn time in the evaluation of candle particle emissions, since the average emissions are affected by the burn time.

Particle size distribution analysis over an extended size range showed two distinctly different particle types. UFPs below 100 nm dominated the number emissions in all experiments. The UFP mass showed strong correlations with the inorganic PM mass from chemical analysis. UFPs dominated mass emissions during steady burn. During stressed burn, a larger sized soot mode of eBC (EC) particles, emitted in addition to UFPs, dominated the mass emissions for all candles.

Measurements directly above the flame showed empirical evidence of short-lived very strong emission peaks of soot particles. This provided new fundamental knowledge on the emission mechanisms and dynamics during stressed candle burning, that can be used in the future formulation of candles with further reduced emissions during stressed burn. When the candles were extinguished by using a snuffer to intentionally minimize emissions (in-line with recommendations by candle manufacturers), a peak emission of OA was observed. The OA mass spectra from extinguishing showed markers...
of wick pyrolysis products in addition to the previously reported wax components.

The results reported in this research showed highly correlated co-emissions of the health-relevant particle components eBC, OA, and PAHs when the candle flame was stressed. This was observed for both chamber measurements and measurements directly above the flame. An implication from this is that the emissions of all these three components increase under stressed burn conditions, and that the emissions can be significantly reduced by limiting stressed burn conditions. For all candles, NO\textsubscript{x}, gas-phase PAHs, and formaldehyde did not show clear emission trends from the influence of fluctuating air velocities, wax and wick composition, or burning mode (stressed or steady burn). The results show that the burning of candles at stressed conditions in indoor environments is likely to significantly add to the exposure to soot (measured as eBC or EC), PM\textsubscript{2.5}, particle PAHs, and NO\textsubscript{x}.

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CONFLICT OF INTERESTS

The authors declare no competing interests.

AUTHOR CONTRIBUTIONS

CA, AW, JP, MB, MG, and MK responsible for conceptualization. CA, YO, and BS responsible for formal analysis. JP, AW, MB, MG, and MK responsible for funding acquisition. CA, YO, SNS, BBR, MNB, MK, and KW responsible for investigation. CA, AW, and JP responsible for methodology. AW responsible for project administration. AW, JP, MB, and MG responsible for resources. AW, JP, MB, and MG responsible for supervision. CA, AW, and JP responsible for validation. CA and YO responsible for visualization. CA wrote the original draft. All authors contributed to writing review and editing.

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SUPPORTING INFORMATION
Additional supporting information may be found online in the Supporting Information section.

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