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Environmentally friendly candles with reduced particle emissions

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Preface

The project “Environmentally friendly candles with reduced particle emissions” is co-financed by the Danish Environmental Protection Agency (EPA) under the subsidy scheme MUDP 2016.

This report describes the project results and methodology used in achieving these results. The purpose of the project was the development of environmentally friendly candles with reduced particle emissions.

The project was carried out in a partnership between ASP-Holmblad A/S, Liljeholmens Stearinfabriks AB, Promol – Industria de vesla S.A., European Candle Association ASBL and Danish Technological Institute.

All pictures and figures shown in the report are produced within the project group unless stated otherwise.

Danish Technological Institute

Aarhus, August 2018

Summary and Conclusion

Measurements of the emissions and their chemical composition
To support the development of low emission candles, measurements of emissions from five different types of candles were conducted in triplicates. These measurements where done partly to characterize the emission from candles and to balance possible differences in the performance of individual candles.

Prior to the measurements, it was decided to protect the candles from draught during burning because this is what candle manufacturers strongly recommend consumers to do. A steady burn setup was also chosen as it was a more reproducible method and covers the major time of candle burn in practice.

Measurements of ultrafine particles (4-166 nm) showed room concentrations between approx. 104,000 and 430,000 particles/cm^3. The candles with the highest fuel consumption per hour and with the strongest wick protection showed the highest emission. Water-soluble salts, such as ammonium phosphates and sulphates are used to protect the wick from corrosion. Analyses of the collected particles showed that virtually the entire emitted particle mass of some candles consisted of these salts. The total particle mass ranged between 29 µg/m^3 and 59 µg/m^3 and its black carbon content ranged at a low percentage, between 0.46 % and 1.5 %. Black carbon is an important indicator of soot and critical substances such as polycyclic aromatic hydrocarbons which are typical constituents of soot. Previous studies have shown that the levels of emitted black carbon increases dramatically under non-optimal burn conditions (sooting burn), and this knowledge underline the importance of development of candles towards a stable burn under sub-optimal conditions

Measurements of metals, such as lead and nickel, showed no emissions above the detection limit, and also the measurement of emitted (semi) volatile organic compounds, (S)VOC, was unremarkable. Some typical combustion products from this group of substances, e.g. acetone, isopropyl alcohol and toluene were detected in the emissions of all candles, but at low levels. Modifying the composition of the reference candles resulted in a change of the emission, but it was not possible to reduce all different kinds of emission with one of the modified candles.

Assessment and conclusions
The study confirmed that candles that are commonly used in Denmark are a significant source of fine particles to indoor air. The number of particles and the total particle mass that are emitted depend on the burn rate and the amount of salts necessary for wick protection. It must be noted however, that if candles are protected from draught, the level of black carbon, which is an indicator of soot, is very low and virtually all emitted particles of some candles at least consist of the salts for wick protection.

This is an important finding because the salts are, in contrast to particles such as diesel soot or dust, not persistent but can dissolve easily when they get in contact to the humid surfaces in the human respiratory tract. One could therefore assume that these salts can be excreted much easier by the body. Exposure studies of human cells and preferably of human panellists should follow up this angle to confirm the assumption of course. But based on these new findings, direct comparison of particles from candle burning under optimal conditions with those from other sources, such as traffic, does not seem to be appropriate.

Future research should also provide more knowledge which type of emission can be reduced with which modification of the candles design or composition. If it is not possible to reduce all types of emission in general, priority should be given to those that pose a potential health risk. Until then, consumers should take care to use high-quality candles, to trim the wick if it gets too long and to protect the candles from air flows that make the candle flame flicker so that the candles can burn calmly and without sooting.

Sammendrag og konklusion

Måling af emissioner og deres kemiske sammensætning
For at understøtte udviklingen af lavemissions levende lys, er der foretaget målinger på tre eksemplarer af fem forskellige slags levende lys. Disse målinger er foretaget dels for at karakterisere emissioner fra levende lys og dels for at balancere mulige forskelle på præstationerne for disse.

Inden målingerne blev udført, blev det besluttet, at beskytte de levende lys mod træk under afbrænding, da det er dette producenterne anbefaler forbrugerne. En opsætning, hvor lysene brænder med rolig flamme blev valgt, da den metode er mere reproducerbar og dækker lysenes brændetid i praksis.

Målinger af ultrafine partikler (4-166 nm) viste koncentrationer i rummet på ca. 104.000 og 430.000 partikler/cm3. Lysene med den største forbrænding pr. time og den stærkeste vægebeskyttelse udviste den største emission. Vandopløselige salte som fx ammoniumfosfat og sulfat bruges til at beskytte vægen mod korrosion. Analyser af de opsamlede partikler viste, at for nogle lys bestod stort set hele den afgivne partikelmasse af disse salte. Hele partikelmassen var på omkring 29 µg/m3 og 59 µg/m3, og indholdet af carbon black var lavt – mellem 0,46 % og 1,5 %. Carbon black er en vigtig indikator for sod og kritiske komponenter som fx polyaromatiske kulbrinter, som er typiske bestanddele i sod. Tidligere studier har vist at mængden af black carbon stiger væsentligt ved ikke optimal forbrænding (sodende afbrænding), og denne viden understreger vigtigheden af at udvikle lys imod en stabil forbrænding under ikke optimale forhold

Målinger af metaller, som fx bly og nikkel, viste ingen emissioner over detektionsgrænsen og der var heller ingen bemærkninger til målingen af afgivne (halv) flygtige organiske komponenter (S)VOC. Dog blev nogle typiske forbrændingsprodukter fra denne gruppe af bestanddele fx acetone, isopropylalkohol og toluen påvist i lave mængder. Modificeringen af referencelysenes komposition resulterede i en ændring i emissionen, men alle former for emission kunne ikke reduceres samtidigt med de valgte modifikationer.

Vurdering og konklusioner
Undersøgelsen bekræftede, at de levende lys, som oftest bruges i Danmark, er en væsentlig kilde til fine partikel i indendørsluften. Antallet af partikler og den totale partikelmasse, som afgives, afhænger af hastigheden, hvormed lysene brænder, og mængden af salt, som er nødvendig for at beskytte vægen. Det bør dog bemærkes, at hvis lysene beskyttes mod træk, så er carbon black-niveauet, som er en indikator for sod, meget lavt, og mere eller mindre alle emitterede partikler fra de levende lys består af salte benyttet til vægebeskyttelse.

Dette er en vigtig iagttagelse, da salte i modsætning til partikler som fx dieselsod eller støv ikke er bestandige, men de opløses nemt, når de kommer i kontakt med fugtige overflader i luftvejene hos mennesker. Derfor kan man antage, at kroppen meget lettere kan udskille disse salte. Studier af humanceller og studier på mennesker bør forfølge denne vinkel for at bekræfte antagelsen. På baggrund af de nye iagttagelser, er det ikke hensigtsmæssigt, at partikler fra afbrænding af levende lys under optimale forbrændingsforhold sammenlignes direkte med partikler fra andre kilder som fx meget trafikerede gader.

Fremtidig forskning bør også bidrage med mere viden om, hvilke typer af emission der kan reduceres, og med hvilke modifikationer af lys mht. design eller komposition. Hvis det generelt ikke er muligt at reducere alle former for emission, så bør man prioritere de former for emission, der medfører potentielt sundhedsmæssige effekter. Indtil da bør forbrugerne vælge levende lys af høj kvalitet, og de bør klippe vægen, hvis den bliver for lang. Lysene bør beskyttes mod luftstrømme, som får dem til at blafre, og man bør også sørge for, at de brænder med en rolig flamme og uden at sode.

Definition of Words

Abbreviation	Explanation
AAS	Flame atom absorption spectrometry
CPC	Condensing Particle Counter
GC-MS	Gas chromatography – mass spectrometry
ICP-AES	Inductively coupled plasma atomic emission spectroscopy
ICP-MS	Inductively coupled plasma mass spectrometry
ICP-OES	Inductively coupled plasma optical emission spectrometry
NOx	Sum of NO and NO_2
POA	Primary organic aerosol
SOA	Secondary organic aerosol
SMPS	Scanning mobility particle sizer
PAH	Poly aromatic hydrocarbon
PM	Particulate matter
PTR-MS	Proton transfer reaction – mass spectrometer
VOC	Volatile organic compound
XAD2	Chemical adsorption material

1. Introduction

Denmark is the country in the EU with the highest per head consumption of candles. 39 % of the Danes use candles daily or almost daily in their homes, but there has been a significant drop in candle use since 2010. The major reason for that is probably the numerous media reports in recent years that reported about high emissions of fine particles from burning candles and that this could be a significant health risk for people who use a lot of candles, based on comparisons with particles emitted by other sources, such as traffic or dust for example.

Little is known about the health effects of particles from indoor sources, such as cooking, vacuuming or burning candles however. Apart from the number and size of the particles, the chemical composition might have a significant influence on the assessment of health effects. For this reason, the present project was initiated in order to develop new candles with lower emissions of health-relevant substances. This developement will be based on a better understanding of what particles and substances are emitted during candle burning and at what levels.

As a first step, two of the most common candle types used in Denmark were picked as reference candles and tested for the most relevant emissions: crown top candles and pillar candles made of animal stearin. Then the pillar candle type was modified by using a different wicks and/or fuels to investigate the effects on the emissions.

The measurements according to scientific standards and with state-of-the-art equipment were performed by the Danish Technological Institute, DTI, in a climate room of 20 m^3 in order to simulate realistic consumer use. Background knowledge about candle performance and the candles were provided by partners from candle industry.

1.1 Objectives of the project

The project had two objectives. The first objective was to develop a new type of candle with similar performance as existing candles but with lower emission of (health-relevant) particles. The second objective was to measure the emission of particles and other substances from burning candles with state-of-the-art measuring equipment to obtain a better understanding of emitted substances under normal candle use – both qualitatively and quantitatively.

2. Background

Danes have a reputation for enjoying life and making themselves comfortable during the long hours they spend indoors during the dark times in winter. They even created their own word for it, *hygge*. For many Danes, warm and cozy candle light is something that is inseparably linked to *hygge*, and so it does not come as a surprise that Denmark has the highest per capita consumption of candles in Europe.

In recent years, there have been a number of reports that warned people of health risks when they use a lot of candles. The focus has been on ultrafine particles that are emitted during candle burning and can get into the pulmonary alveoli if inhaled. The reports often compared the ultrafine particles from candle emissions with particles known to be emitted from other sources, such as diesel cars or wood stove fires.

Measurements in rooms in which candles were burning have shown comparably high concentrations of particles which are small enough to get into the pulmonary system and concluded that burning candles are a major source of particles in indoor air. Other indoor sources are, for example, ethanol fire places, cooking and vacuuming. At this point in time however, there is very little knowledge about the particle composition and the concrete health risks of ultrafine particles, especially in indoor air, and if the particles emitted by different sources are equally critical.

This lacking knowledge has been seen in some headlines in press reports that suggested breathing the air in a room with burning candles was even more dangerous than the air beside a major road. These reports have had an effect on the population and unsettled consumers by the warnings and fear for their health. This has caused a significant decrease in the use of candles which can be seen in a massive drop in the candle consumption per capita over the past years.

The reported measurements typically focused on the particle size and number concentration. These kinds of measurements can be performed using advanced equipment but have also in some cases been measured with handheld measurement devices. It is crucial to evaluate the health risks, however. The chemical composition of the particles, which would have been the prerequisite of such an assessment, has been a lacking part of these measurements. Suggestively because it can be a complex task and requires both additional equipment and scientific expertise. This is very important however, because even though small particles behave differently than larger ones or gases, it is assumed that the chemical composition of the particles has a huge effect on the health risk.

A scientific study by Pagels et al. from 2009[1] investigated the composition of candle particles. It found that the majority of particles emitted by well-burning candles, i.e. by candles that are not visibly sooting, primarily consist of the salts used to treat the candle wicks. These types of salts are easily dissolved in water. One assumption is therefore, that when such salt particles are inhaled, they dissolve when they get in contact with the humid tissue in the lungs and can be excreted by the body. This would be a major difference compared to other, persistent particles and might also lead to an entirely different evaluation of possible health risks. The situation is however different during a sooting burn, where relatively high levels of elemental carbon is observed, these studies underline the importance of focusing the development of low emission candles towards a stabile non-sooting burn.

[1] https://doi.org/10.1016/j.jaerosci.2008.10.005

As a reaction to the increasing number of concerned consumers, a joint research group was established with the objective of getting a better understanding of the emissions from burning candles and, based on these findings, reducing the emission of health-relevant particles by development of a new candle. The Danish Eco-Innovation Program MUDP with its objective of improving the environment granted funding. The project partners are the Danish Technological Institute (DTI), several candle manufacturers and the European Candle Association (ECA). DTI was in charge of measuring the emissions with state-of-the-art equipment. The results were evaluated by the project group. The industry partners provide knowledge of candle performance and suggestions to design modifications in order to reduce health-relevant emissions, e.g. by using different fuels or wicks.

The long-term objective after understanding the emission of health-relevant particles is having low emission candles that can be recognised as such by consumers and help them creating a healthy indoor environment. The most commonly found quality labels for candles in today´s market are the Swan label and the RAL Quality Mark for Candles. Both contain requirements on the raw materials and performance including a limit for the emission of soot, but do not cover the emission of fine particles due to lack of knowledge today.

2.1 Candles and Culture

There is a long tradition of burning candles on many different occasions in Denmark. They are used at home for enjoying and relaxing, on the table in restaurants or during a meal with family and friends, in the living room while watching TV, and one often sees candles in windows or in corners to create a nice atmosphere.

Candles are also used in churches for religious ceremonies, especially during Easter and Christmas celebrations. It is common to have candles on the birthday cake and it is a tradition to celebrate the liberation day on 4th May with burning candles in the windows. During Christmas time, candles are used for advent decoration and on Christmas wreaths, or people use calendar candles to count down the days until 24th December. On Christmas eve, many people have candles on their Christmas tree.

Generally speaking, candles have been present for centuries when people come together and celebrate or simply enjoy live, and they have become part of the Danish culture and tradition.

2.2 Facts and figures

During the last 7 years, there has been a trend of decreased candle use in Denmark while the use of candles in the EU as a total increased at the same time. Nevertheless, Denmark is still the country in the EU with the highest consumption per capita.

A recent study[2] showed that 39 % of the Danes light candles daily or almost daily in their homes and that 41 % of the daily users have 3-4 candles burning at a time. Older people use more candles than younger.

Market research data from A.C Nielsen that measure candle sales out of retail stores suggest a declining trend, too. Since 2010, the market has been going down from 641.8 mill DKK to 420.8 mill DKK by the end of 2017, so there has been a drop in the retail trade of 34.5 % in value.

There are many possible explanations for this development, but the biggest impact is suggested to come from negative press reports. Unsettled consumers have started to replace candle light with products such as LED lights, as one example. A positive effect of this is that LED

[2] Made in November 2014 by YOUGOV for Bolius; https://www.bolius.dk/derfor-elsker-danskerne-stearinlys-24773/

lights do not emit particles. From an environmental perspective, though, use of plastic and battery is a possible concern.

A window of opportunity thus exists for achieving improved knowledge about candle emissions and for using this information in development of new, improved, low-emission candles which has been the main objective in this project.

TABLE 1. Consumption of candles in Denmark. Source: Eurostat 2017.

Year	Candle consumption [kg]
2007	6.8
2008	5.9
2009	6.3
2010	7.2
2011	6.1
2012	5.7
2013	6.0
2014	5.8
2015	5.1
2016	4.3

2.3 About the tested candles - background

Candle manufacturing is nowadays a strongly regulated industrial activity due to the complexity of the process and the materials used. Historically candles were used to overcome the absence of light for indoor activities, mainly during the night. Nowadays there is a culture behind the utilization of candles, in order to create a warm and cosy ambience and tune the senses into a positive and relaxed mood. But the user requires a candle to behave within a certain pattern and quality is a must. While the main functionality is to generate light and heat, the candle “system” must be balanced all the way: it must stand a tall flame, show a consistent burn rate, not drip off liquid wax, it must not produce soot (unless the flame is disturbed by massive air movement), the flame must not flicker, and it must be odour free unless this is intended, e.g. with scented candles. To achieve these standards, it must be assured that the raw materials are adequate and the industrial process is controlled.

The candles chosen for these measurements are representative of the most widely used candles in Denmark:

- Crown top candles:
  cylindrically shaped candles in several heights and diameters, to be used in a candle holder for stability against toppling over. Their name “crown top” comes from the shape of the head. Depending on the weight, such candles burn typically for 6-9 hours.
  Crown top candles are typically produced using moulding technology. Raw material in liquid form is poured into pre-heated metal moulds and the raw material is allowed to slowly solidify. In the centre of the mould a wick of suitable size is fixed. When the candle has cooled down and is solid the candle can be removed from the mould and the process repeated. A typical production line consists of rotary moulding machines with up to 3,195 moulds. Stearin is the most widely used raw material for crown top candles.

- Pillar candles:
  cylindrical candles not needing to be put on a candle holder as they are normally wide enough to stay in the vertical position without additional support. Pillar candles with one wick

typically have diameters between 40 mm and 100 mm. Depending on the weight of the candle, they can burn for 50 hours or longer.
For the production of this type of candle, stearin or paraffin is used as solid flakes, being compressed in a metallic mold using the force of a hydraulic press, with the wick inserted. The candle is then over-dipped with an outer material (normally paraffin) to create an outer protection against mechanical stress and heat deformation.

There are two critical components for producing well-performing candles:

- Fuel:
  often called wax or base material; this is the material that is burned in order to create light and heat. The selection is based on quality criteria and environmental criteria. While the quality criteria define the technical specifications that should be controlled in order to guarantee a good and controlled behaviour of the candle while burning (melting point, penetration, oil content, etc.), the environmental criteria is based on the sustainability of the overall sourcing system, meaning how severe the impact is on the utilization of a certain fuel.

  In Denmark, the most widely used fuel is animal based stearin, but vegetable stearin and paraffin can be used as well. Stearin is a mixture of fatty acids that is obtained from fats and oils of animal or vegetable base, by the saponification of the triglycerides using hot water. Commercial stearin is predominantly a mixture of stearic and palmitic acid. Paraffin is a side product of mineral oil refinement that is typically cleaned to a white, odourless wax with hydrotreatment.

- Wick:
  without a wick it would not be possible to sustain the flame for a long time. It feeds the heat source with enough fuel to keep the combustion going and produce light and heat, creating a loop effect and keeping the system stable. It is normally made of pure cotton and braided with a certain number of thin threads that transport the melted fuel with capillary effect to the flame.

Decades ago, lead was used as a core to stabilize the wick of certain candle types, mostly outdoor candles. But due to the volatility of lead and the risk that it is emitted to the air during burning, the use of such lead core wicks was stopped in the 1980s in Europe. Some studies made around the turn of the millennium found such lead cores in candles on the Australian and US market however, and emission measurements showed significant lead emission to the indoor air coming from these candles while burning. As a consequence, lead core wicks were also banned in both Australia and the USA soon after. More recent studies have, if at all, only shown extremely low lead contents in candle base materials coming from ubiquitous contamination.

Nickel compounds are not actively added to candles either but can be present as contamination at very low levels. This is because, nickel can be used as a catalyst in the process of hydrotreating paraffin wax, vegetable fats and animal fats and residues can be found in the resulting materials. In the first case, problematic substances that are natural constituents of crude oil, such as polycyclic aromatic hydrocarbons and aromatics, are removed. In the second case, liquid oils or soft fats are hardened so that they can be used as a candle fuel. The catalysts are removed after the hydrotreatment so that possible residues are only contained at very low levels. Nickel compounds are also much less volatile than lead.

High-quality candles are designed to burn with a calm, non-flickering flame that facilitates a nearly complete combustion of the fuel. In case of low-quality candles or if the combustion is disturbed, e.g. by draught, there is a risk that the combustion is incomplete and soot is emitted that mostly consists of carbon enriched, incompletely burned fuel. Since soot can also contain problematic substances like polycyclic aromatic hydrocarbons, the emission should be as low as possible.

2.4 Performance of Quality Control

Before a new product is introduced in the market for the first time and continuously during and after its production, the performance is assessed and compared with project specifications. The most important protocols of Quality Control are burning performance tests and soot tests.

2.5 Methodology of the performance tests

Performance tests are made according to RAL-GZ 041 (burning tests) and EN 15426 (Candles - Specification for sooting behaviour).

To perform these tests, the air temperature has got to range between 20-25°C and candles are burned in different cycles depending on the candle type and size.

In burning tests after around 5 min, the candle has to show a calm flame and gradually form a cup rim surrounding the so-called burning bowl. Furthermore, the wick must have a medium curvature while the candle is burning, the candle must not drip and the flame must burn without visible release of soot.

In sooting tests, the soot which is emitted from a candle is collected on a glass plate throughout a defined period depending on the candle type and size. Then the light intensity of the clean glass and of the glass after burning is measured. The attenuation of light intensity caused by soot precipitation is quantified as the hourly soot index. An hourly soot index below 1,0/h indicates that the candle is not sooting visibly. Typically, the hourly soot index is much lower than this limit.

FIGURE 1. Example of sooting test equipment (diverse diameters for different diameter candles).

3. Methodology and limitations in this study

Two of the most commonly used candle types in Denmark were chosen as reference candles - uncoloured and unscented crown top candles with a diameter of 23 mm and a height of 200 mm and pillar candles with a diameter of 58 mm and a height of 120 mm, both made of 100 % animal stearin (i.e. animal based fatty acids). Another very popular candle type - uncoloured and unscented tea lights – had to be disregarded due to limited financial resources. Tea lights have a rather small flame however and are presumed to have lower emissions than other candle types.

The idea was to take the emission of the two candle types as a reference and starting point for developing candles with a lower emission by changing the wick and fuel. Thin candles like crown top candles are limited to certain fuels because otherwise they would not be stable enough. For this reason, it was decided to take pillar candles for the modifications because they can be made of more different fuels and blends of fuels.

In earlier studies, DTI made measurements of the source concentration (measurement point 20 cm above the burning candle). This measurement method allows direct comparison of the emissions of different candles. The drawback of this method is that it can be difficult to relate to realistic human exposure. The emitted particles can agglomerate to larger particles, especially in humid air, and there are also sink effects that are not considered when measuring too close to the source. Based on these considerations, it was decided to burn the candles in a test room and measure the room concentration in adequate distance to the source, simulating a more realistic use and exposure situation.

The room concentration was measured using a climate room with 20 m^3 (3.17x2.17x2.88 m^3) in size. The climate room is placed inside an existing laboratory room away from outer walls and windows and consists of laboratory walls on two of the sides and two plastic walls. There is an entrance to the room via a double layer of plastic which covers the outside and inside of the room so as to guarantee that the room is airtight. Inside the room ventilation is regulated by means of an air change of 0.5 times per hour.

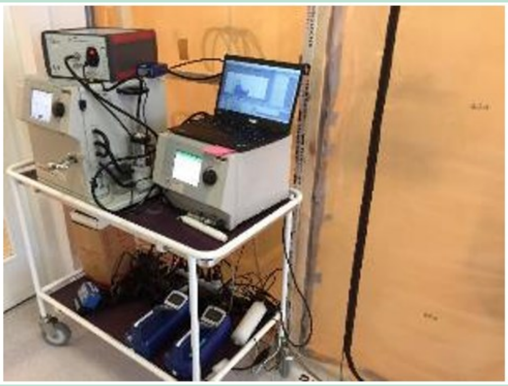

FIGURE 2. LEFT: Sampling position 2 with DustTrak and pumps for filter collection on the roller table to the left. A burning candle can be seen on the roller table to the right.
RIGHT: Roller table with SMPS equipment outside the climate chamber.

Two monitoring stations were utilized for the measurement of particles in the climate room. The two monitoring stations were placed in diagonally opposite corners, 40 x 60 cm from the walls and 125 cm above the floor (around head height for a seated person). One lit candle was placed at a distance of approx. 150 cm from both monitoring stations in a wire mesh cylinder (230 mm in diameter, 300 mm in height, air permeability 60 ± 5 %) in order to protect the flame from possible draught caused by the ventilation (see FIGURE 3).

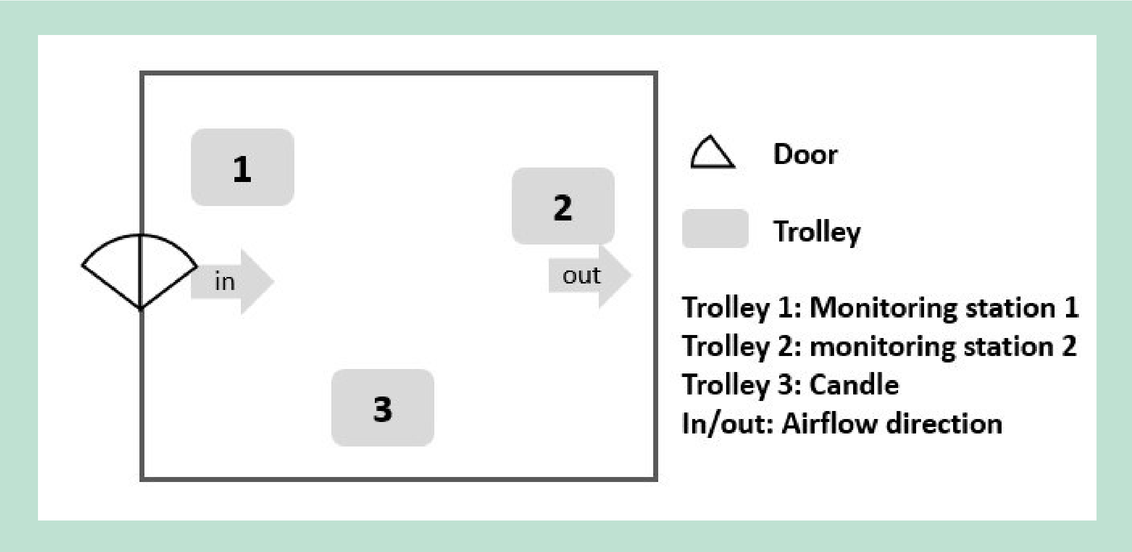

FIGURE 3. Illustration of the climate room and the placement of measurement stations.

To ensure a proper mixing of the air in the room, the air inlet was placed just above floor height, and the air outlet was placed under the roof. This was to ensure the mixing of the air, not only from “side to side” in the room, but also from top to bottom. Both temperature (22-27°C) and air humidity (RH = 40-60 %) were logged over the measuring period. A data point per 5 second was recorded.

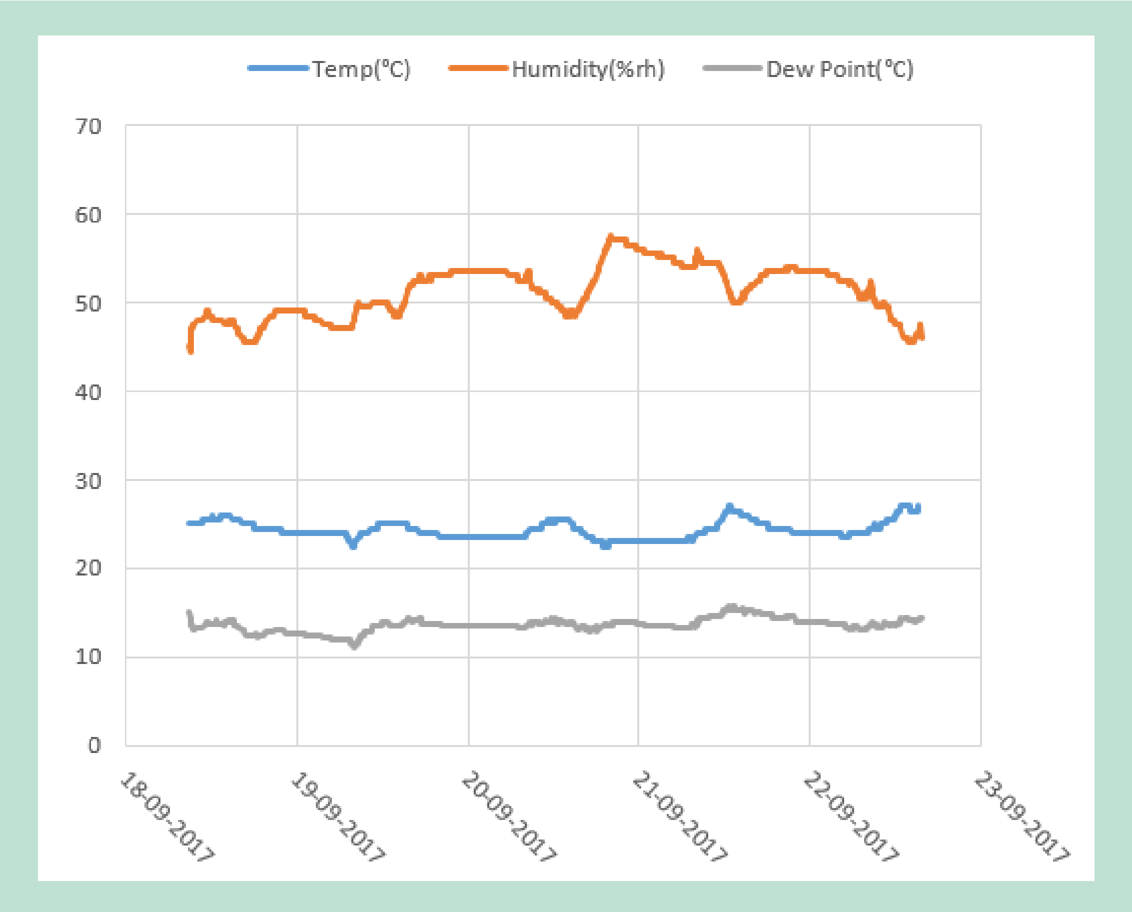

FIGURE 4. Example of logged temperature, humidity and dew point data from inside the climate chamber from a measuring campaign period over a working week. Average temperature was 24.3°C, average humidity was 51.3 %RH.

3.1 Description of measurement methods

3.1.1 SMPS – number concentration and size distribution

SMPS (Scanning Mobility Particle Sizer, model 3080) with nanoDMA (Differential Mobility Analyzer, model 3085) from TSI was used for all measurements. NanoDMA size distributes the particles in up to 167 size fractions. Subsequently, the particles are counted in a CPC (Condensation Particle Counter, model 3776, TSI), so it is possible to read the number concentration (number/cm³) as well as the size distribution. With SMPS, measurements were carried out in the size interval of 4.4-166 nm, which according to experience covers the particle emissions from candles the best.

The scanning time per measurement is 180 seconds with subsequent 30 seconds of down scan (”preparation” for next scan), which gives a classification in app. 100 size fractions (scanning time is of importance to number of possible fractionations).
Particle size distributions of the different types of candles will be treated. Measurements have been performed with SMPS in the size range ~4 – 160 µm distributed on approx. 100 size bins. For each bin size, the number of particles have been counted. This gives rise to a particle size distribution which can be seen in paragraph 4.3.

3.1.2 DustTrak – particle mass emission

DustTrak DRX (model 8533 from TSI) was used to measure the particle mass emission from burning candles. The particles are measured simultaneously in five size fractions (PM1, PM2.5, PM4, PM10 and total PM, where PM stands for Particulate Matter). PM2.5 covers the mass of particles with an optical diameter less than 2.5µm, measured in the unit mg/m³. The time solution is set to 5 seconds, which makes it possible to follow the development of the particle mass emission over time during the measurement period. DustTrak detects particle mass for particles larger than approx. 0.3µm and up to approx. 15µm in the concentration area of 0.001-150mg/m³. The measurement with DustTrak was started and finalized so the measurement was carried out in the same test period as the measurement with SMPS.

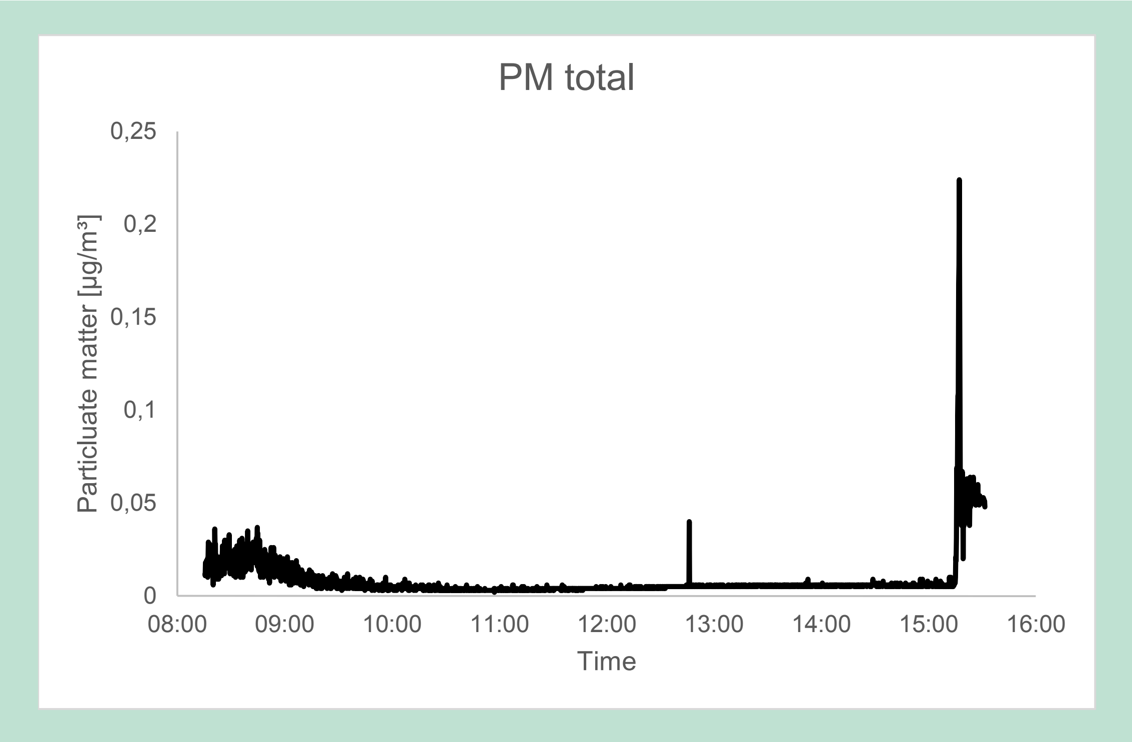

FIGURE 5. A typical graph showing the measurement of particle mass during burning of a candle. When igniting the candle, a relatively larger particle mass is seen emitted than when the candle burns stable. In the end (after 15:15), the candle is extinguished and the particle mass rises due to smoke typically seen. At 12:47 a disturbance is registered.

3.1.3 Aethalometer – black carbon measurement

Black carbon (BC) was measured using Aethlabs microAeth, model AE51. The aethalometer performs continuously absorbance measurements on an accumulating filter, i.e. it measures the change in absorption of transmitted light due to continuous collection of aerosol deposit on the filter. If the concentration of BC is very low, electronic noise from the instrument will dominate. This will be shown as very low mass concentrations that varies around zero where both positive and negative values for the mass will fluctuate. The measurement resolution for the instrument is 1ng BC/m^3. Measurement range is 0-1 mg BC/m³.

3.1.4 Analysis of total dust content

The samples are analysed according to method UA206 at Danish Technological Institute. Weighing is performed according to method AMI L15. The detection limit is estimated to 2.0 µg/m³. In practice, this means that the filters are acclimatised 48 hours in an accredited climate chamber before weighing and use. After use, the filters are again acclimatised 48 hours before final weighing. The total dust content is then calculated as the difference between the two weightings.

3.1.5 Analysis of soluble salts

Particles are collected on membrane filters with a pore size of 0.8 µm. The filters are extracted with 30 mL demineralized water for 30 minutes. The nitrate and sulphate content in the extract is determined by ion chromatography. The content of ammonium and phosphate are analysed in the same extract using spectrophotometric on Rohasys Hach Lange DR3800 robot with analysis kit Hach APC304 and APC349.

3.1.6 Analysis of metals

Quantitative analysis by ICP-MS

Particles are collected on membrane filters with a pore size of 0.8 µm. The filters were digested with concentrated HNO_3 diluted to a total volume of 50mL in a closed Teflon tube using microwave induced heating. The blind sample was handled accordingly. Single determination was performed.

An aliquot of digestion solution and a blind were analysed for the content of the following elements: Pb, Ni, Cu, Cr, Zn, Sn, Sb, Fe and Co using ICP-MS with CCT in KED-mode using He as collision gas, and Ge, Rh and Re as internal standards.

Quantification by ICP-MS was performed against traceable external standards of the same elements. Calibrations were verified with independent traceable control solutions.

Quantitative analysis by AAS

Particles are collected on membrane filters with a pore size of 0.8 µm. The filters were digested by means of microwave induced heating prepared with 5ml conc. HNO_3 (from sub boiling quality). The resulting solution was diluted to 25ml with Milli-Q water. Blanks were made correspondingly.

Samples and blanks were analysed for content of K and Na by PE300 Atom absorption spectrometer (AAS). Quantification by AAS was carried out with traceable external standards of the elements. The calibrations were verified with independent traceable control samples.

3.1.7 Analysis of VOC

Approx. 40 and partly 20 litres of air are pumped through ATD-tubes (tenax/carbograph 5TD). The tubes are subsequently desorbed thermally and analysed with gas chromatography using GC/MS.

Identification of detected components is made by comparison of measured mass spectra with NIST MS-library. The content of the detected components is quantified against toluene-d_8. The detection limit is estimated to be 2.0µg/m³. The uncertainty of the method is estimated to be 25 % relative standard deviation.

Apart from the use of ATD-tubes for VOC analysis, a proton-transfer-reaction quadrupole mass-spectroscopy (QMS300 from Ionicon) was used for measuring the absolute VOC concentration in a time-depend measurement. The instrument uses soft ionization by H_3O^+ which leads to a low level of fragmentation, hence no conditioning or sample preparation is needed. The measurement is setup to scan all masses (m/z) from 21-180 amu (atomic mass units), which include most relevant VOCs. The drift tube pressure is 2.3mbar, drift tube voltage is 600V (ca. 140 Td), a low level (<4 %) of parasitic ions (O_2^+, NO^+, $H_2OH_3O^+$) was ensured, and a reference spectrum was taken with activated carbon filtered air. The instrument was calibrated using a VOC gas mix at regular intervals. A scan time is about 90s and typical lower detection limit is 0.1ppb (or ca. 0.3-1µg/m^3), but depends on the actual VOC.

The QMS300 drew the sampling air directly from the candle burning chamber about 20cm from the wall at a height of 200cm from the floor. The sampling line of PEEK is partly heated, but experiments have not shown problems with condensation of the VOCs in the sampling system. The concentrations have been calculated based on a fixed reaction rate (k=2), and without further calibration leads to an error of about 25 %. The instrument background level was found by measuring sampling air from the climate room and subtracting filtered air from an activated carbon filter.

3.2 Measurement set-up

The crown top candle was put into an appropriate candle holder, the pillar candle was used without a candle holder. The candle was placed in the centre of a wire mesh cylinder according to DS EN 15426 (type 1 with a diameter of 230 mm, a height of 300 mm and a permeability of 60 ± 5 %) in order to protect the flame from possible draught caused by the ventilation system of the climate room. One candle and its cylinder was placed on a table in the middle of the room at a distance of approx. 150 cm from the monitoring stations. The burning performance of two candles can vary even if they come from the same batch. All measurements were made as triplicates for this reason.

3.3 Timeline for measurements

During start-up, zero measurement was carried out on the instruments to ensure that the systems were air tight. In addition, background measurements were carried out on the air in the room in order to be able to correct the subsequently measured results on emissions from candles.

The candles were prepared for measurement and were lit. Then the candles burned for approx. four to seven hours during which the measurements were made. Afterwards, the candles were extinguished and the air in the room was changed before the next measurements.

4. Results and discussion

In the following, tables and graphs showing measurement results will be displayed. Common to them all is that the following nomenclature will be used:

CTA:	Crown top candle, 23x200 mm, 100 % animal stearin (animal based fatty acids)
PA:	Pillar candle, 58x120 mm, 100 % animal stearin (animal based fatty acids)
PANW:	Pillar candle with an alternative wick, 58x120 mm, 100 % animal stearin (animal based fatty acids)
PPAL:	Pillar candle, 58x120 mm, 100 % palm stearin (vegetable fatty acids from palm oil)
PPAR:	Pillar candle, 58x120 mm, 100 % fully-refined paraffin wax

CTA and PA are the reference candles representing two of the most commonly used candle types in Denmark. PANW has the same dimensions and fuel as the reference pillar candle PA, but a different wick. PPAL and PPAR have the same dimensions as the reference pillar candle PA, but a different wick and fuel.

CTA and PA were the first candles measured. Based on these findings, a new pillar candle with a new wick was made and measured. After evaluating these results, two new pillar candles were made with other fuel (and wick) types, with the objective to obtain lower emissions.

4.1 Burn rate

All results below are based on 4.4-6.9 hours of candle burning time (see also Table 2)

All uncertainties displayed on the graphs are calculated as one standard deviation of the measurements.

Numeration refers to the actual test number of the specific candles of a certain candle type. For instance, CTA-5 is the fifth test on crown top candles. Previous CTA measurements (e.g. CTA-1) were removed from the tables and graphs because the measurement procedure had been modified based on findings during the project. The results displayed in the tables and graphs were all measured under identical conditions.

The burn rate, or the fuel consumption per hour, is calculated from the total burned mass and the total burn time in each measurement. The crown top candles have the highest burn rate. These thin candles are more prone to dripping after lighting than the thicker pillar candles, and so a comparably thick wick is used to avoid dripping.

TABLE 2. In the table, data of measurement time, burned mass and burning rate can be read.

Sample	Start time	End time	Duration [hours]	Start mass [g]	End mass [g]	Burned mass [g]	Burn rate [g/h]
CTA-5	09:35	15:21	5.8	84.47	35.90	48.6	8.42
CTA-6	08:22	14:58	6.6	84.69	30.22	54.5	8.25
CTA-7	08:05	15:00	6.9	84.89	26.39	58.5	8.46
PA-5	08:40	15:14	6.6	255.6	200.3	55.2	8.41
PA-6	08:49	15:00	6.2	251.17	203.50	47.7	7.71
PA-7	09:02	14:54	5.9	254.14	208.23	45.9	7.83
PANW-2	09:01	14:10	5.2	248.5	211.9	36.7	7.12
PANW-3	08:29	15:02	6.6	249.5	201.0	48.5	7.40
PANW-4	08:29	14:38	6.2	254.7	209.9	44.9	7.30
PPAL1	08:18	14:52	6.6	266.6	222.0	44.6	6.79
PPAL2	08:32	14:58	6.4	268.7	225.1	43.6	6.78
PPAL3	08:56	15:00	6.1	268.4	226.8	41.6	6.86
PPAR1	11:22	15:43	4.4	230.8	203.7	27.1	6.23
PPAR2	08:10	14:34	6.4	221.8	181.5	40.3	6.30
PPAR3	08:29	15:12	6.7	233.0	193.1	39.9	5.94

4.2 Particle number concentration of ultrafine particles

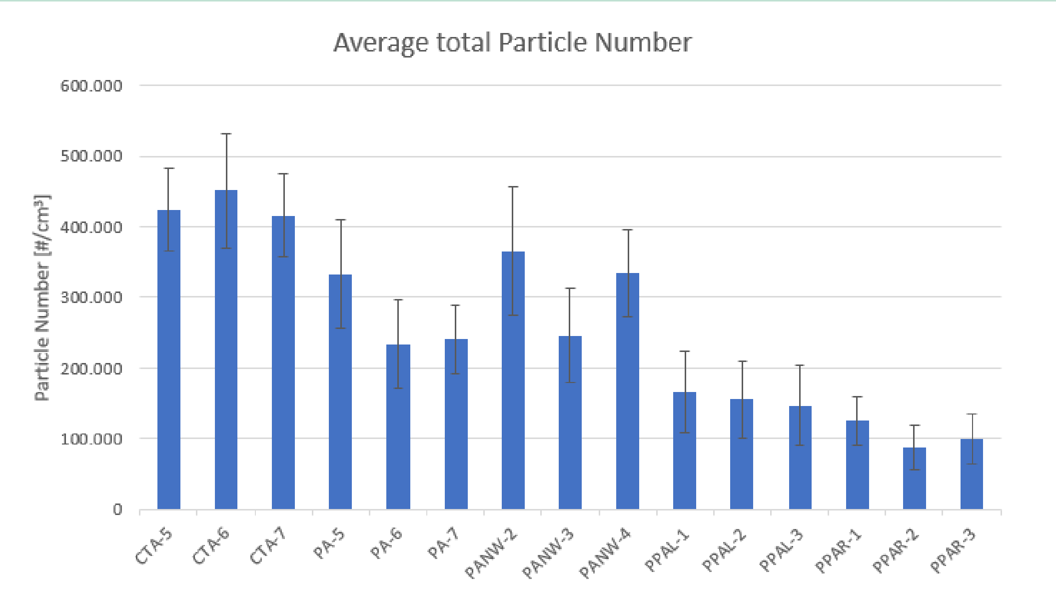

FIGURE 6. The average number concentration of ultrafine particles measured by SMPS.

FIGURE 6 shows the measured average number concentration of ultrafine particles (size range approx. 4.4-166 nm). The highest concentration was measured for crown top candles (CTA), around 430.000 particles/cm³. The reference pillar candles (PA) and the pillar candles with an alternative wick (PANW) resulted in a concentration of around 269.000 particles/cm³ and 315.000 particles/cm³ respectively, showing significant differences between the single measurements however. The pillar candles made of palm stearin (PPAL) and paraffin wax

(PPAR) caused the lowest concentration of ultrafine particles, around 156.000 particles/cm³ and 104.000 particles/cm³.

The measured values can be explained as follows. For one, CTA have the highest burn rate, approx. 8.4 g/h on average. The burn rates of PA and PANW are rather high as well, at 8.0 g/h and 7.3 g/h, and higher than the burn rates of PPAL (6.8 g/h) and PPAR (6.2 g/h). If exactly the same burning conditions are assumed, one can expect that the more ultrafine particles are emitted the more fuel is burned.

Another explanation is that modern candle wicks are braids of very thin cotton threads, and these threads are very sensitive to corrosion, e.g. by the hot, melted fuel. The suction effect of the wick decreases over time and the flame gets smaller and smaller if the threads are not protected. Wick manufacturers therefore treat the wicks with certain water-soluble salts that form a protective layer around the threads. The more corrosive the melted fuel is, the more protective salt is needed. And if one assumes that these salts are not destroyed by the flame but emitted in form of particles, one could expect the highest ultrafine particle emission from the candles with the strongest wick treatment.

CTA, PA and PANW are made of 100 % animal stearin which is a mixture of different animal based fatty acids. The melted fatty acids are more corrosive than other fuels, such as paraffin wax for example, and so more salt is required to protect the wick.

4.3 Particle size distribution of ultrafine particles

FIGURE 7 to FIGURE 11 show the size distribution of the ultrafine particles UFP (size range approx. 4.4-166 nm) for the different candle types. The x-axis shows the size of the measured particles. The y-axis shows the normalized particle number concentration (dN/dlogDp). The graph shows the average distribution during the stable burning period. The average results are calculated based on at least 50 single data sets.

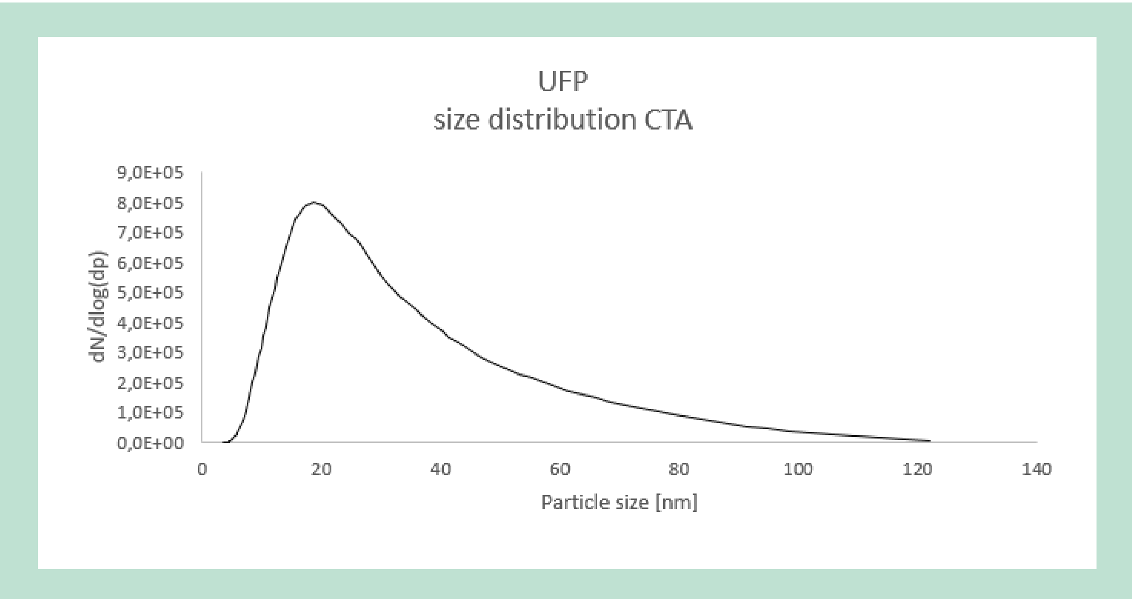

FIGURE 7. Distribution of particles emitted from the reference crown top candle CTA.

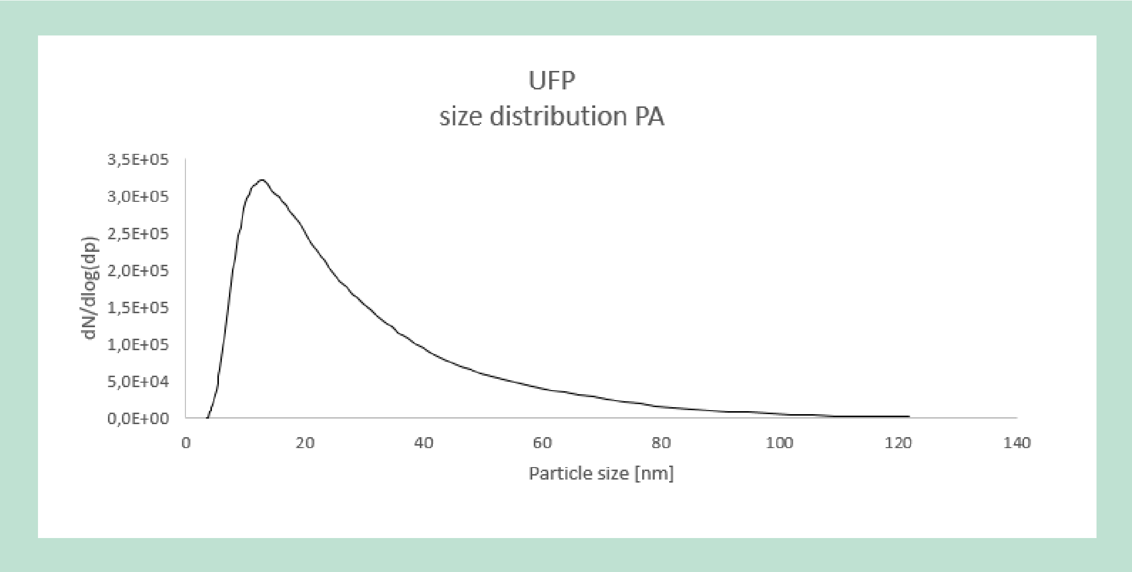

FIGURE 8. Particle size distribution of emitted particles from the reference pillar candle PA.

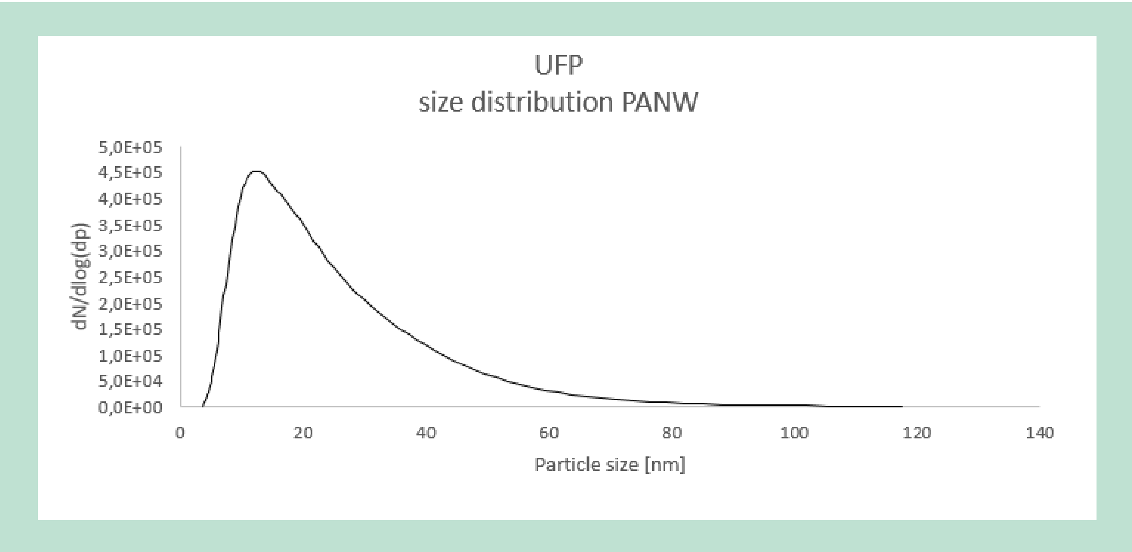

FIGURE 9. Particle size distribution of emitted particles from pillar candle PANW.

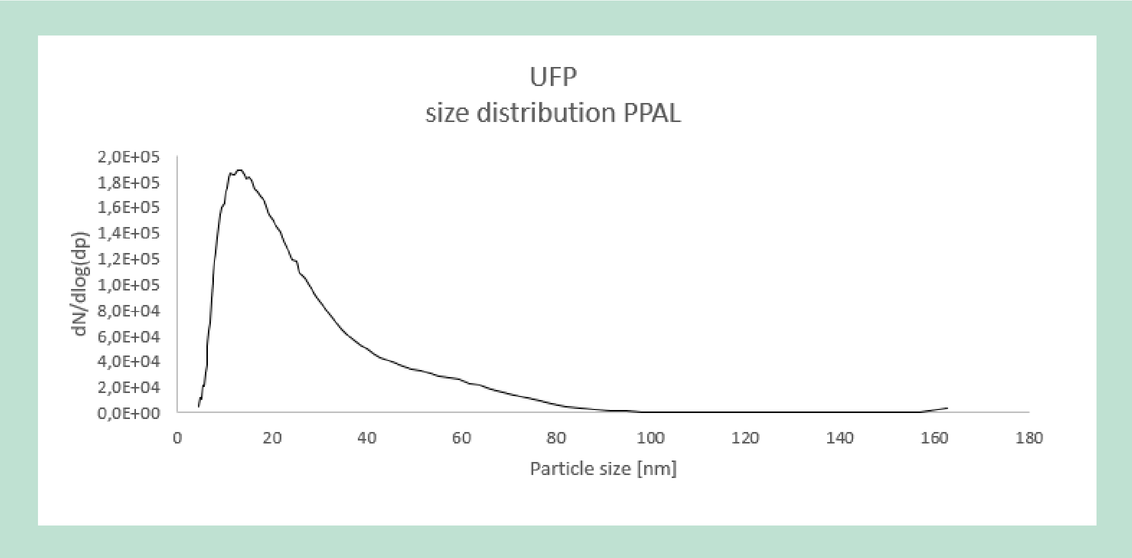

FIGURE 10. Size distribution of emitted particles from pillar candle PPAL.

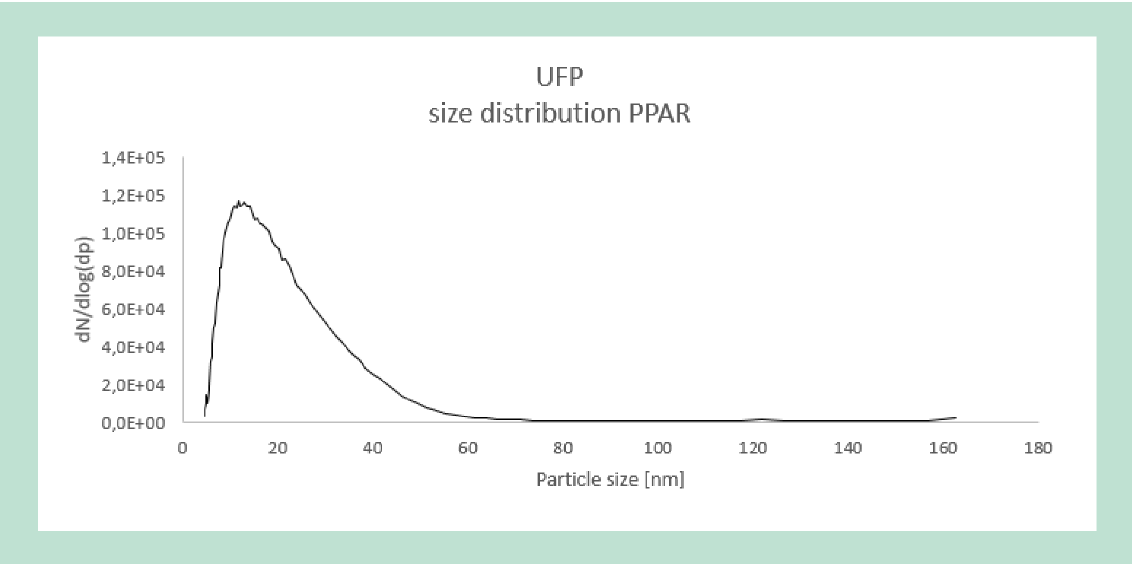

FIGURE 11. Particle size distribution of emitted particles from pillar candle PPAR.

The ultrafine particle distribution looks similar for all candle types. The crown top candles (CTA) emit particles in a size range between approx. 5 and 120 nm with a peak at approx. 20 nm. The different pillar candle types emit particles in a size range between approx. 5 and 80-120 nm with a peak at approx. 10-15 nm. Based on these results, it seems that the type of wick or fuel does not have a significant effect on the size distribution of the emitted ultrafine particles, only on the number of emitted particles. This fits well with the fact that from a burn process point of view, it is similar candle types burning, and the dimensions of the candles are alike, therefore giving rise to same size of particles.

4.4 Particle mass concentration

4.4.1 Total particle mass concentration measured by DustTrak

Two DustTraks were used for online measurement inside the climate room. The two DustTraks were located on each side of the room in order to compare results and thereby beside measure the mass concentration also control the air mixing in the room.

The following mass concentrations measured by DustTrak are all shown as total mass concentration in order to have the full picture of the mass emissions. In all the cases, the total mass concentrations PMtot are very low. PM2.5, which is often used for health-related studies, can in all these cases be considered the same as PMtot, as the particle size of the measured candles are more or less all below 1 µm in size.

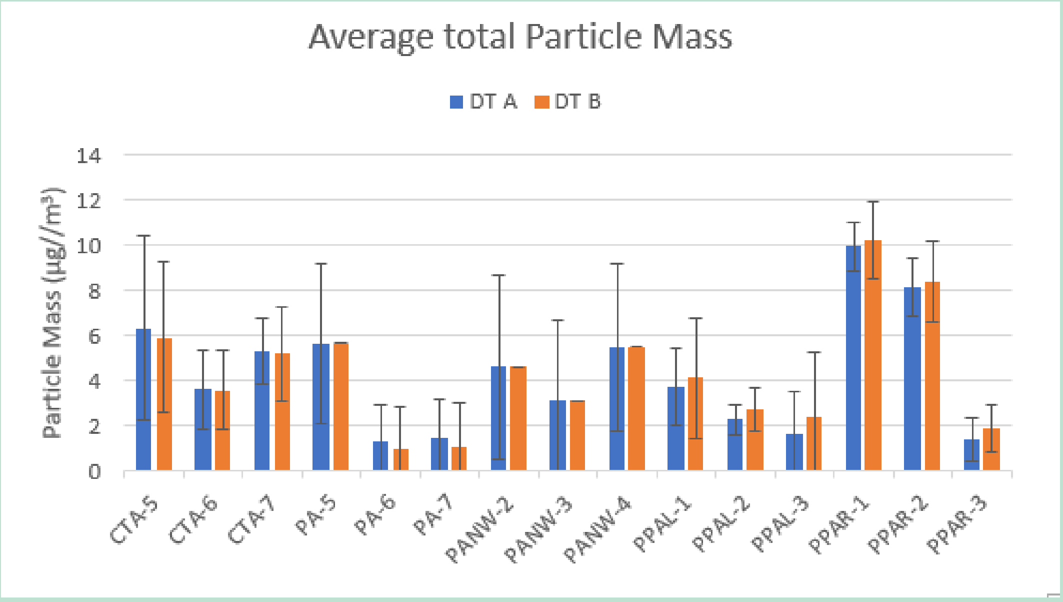

FIGURE 12. The mass emission of particles measured by DustTrak (DT) is illustrated in this graph.

FIGURE 12 shows the total particle mass of particles in the size range from approx. 0.3 to 15µm measured by DustTrak A (DT A) and DustTrak (DT B) in parallel but at two different positions in the climate room. The results of DT A and DT B were very similar which shows that the particle concentration in the room was very homogenous.

TABLE 3. The table shows the average amount of total particulate matter measured by DustTrak.

Candle type	CA	PA	PANW	PPAL	PPAR
Avg. particle mass [µg/m³]	4.9	2.6	4.4	3.1	6.8

Firstly, it should be noticed that the results measured by the DustTrak are very low and near the detection limit of the instrument (1 µg/m³).

Secondly, it is difficult to see a clear trend because there was some rather high variation between the triplicate measurements of the same candle types, especially for PA and PPAR.
In general, the results show that the amount of total particulate matter is below 10 µg/m³ in all cases.

4.4.2 Particle mass concentration measured by filter collection

Particle mass was collected on filters and the filters were weighed to determine the amount of collected total particulate matter PMtot.

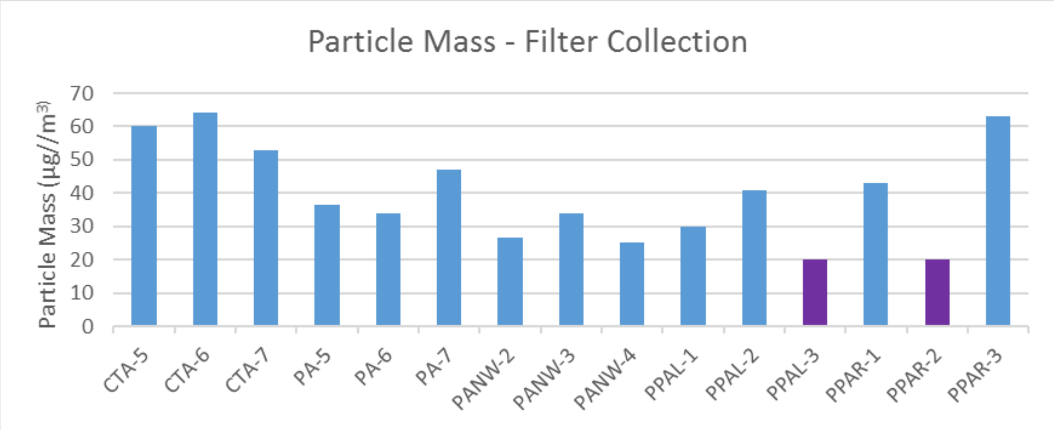

FIGURE 13. The mass emission of particles collected on filters is illustrated in this graph. For PPAL-3 and PPAR-2, the measured values were under the detection limit of 20µg/m^3.

The total amount of collected mass in the PPAL-3 and PPAR-2 filters were below the detection limit of 20µg/m³ the method, hence the other colour of the bars in **Fejl! Henvisningskilde ikke fundet.**.

TABLE 4. The table shows the average amounts of total particulate matter collected on filters.

Candle type	CA	PA	PANW	PPAL	PPAR
Avg. particle mass [µg/m³]	59	39	29	36	53

Looking at the filter collection of particulate matter, the highest concentrations were measured for the crown top candles, followed by the pillar candles made of paraffin wax (PPAR), the reference pillar candles (PA) and the pillar candles made of palm stearin (PPAL) (leaving the filters below the detection limit out of account). The lowest particulate matter concentration was measured for the pillar candles with an alternative wick (PANW). It must be noted however that there has been some significant difference between single measurements for the same candle type.

The values seen in TABLE 4 found by filter collection are higher than the measured amount of particle mass found by the online instrument DustTrak and showed in TABLE 3. It should be noticed that two different measuring systems often give different results. Also, it should be kept in mind that the filter collection method is the 'official' method for measuring particle mass in ambient air.

The advantage of the filter collection is that it collects all mass, typically with the lowest efficiency towards particles between 100-400 nm in size. Particles larger and smaller than this size are almost 100 % certain for being caught. The efficiency depends primarily on particle size and face velocity, which is the velocity of the air flow passing the filter. The pore size of the used membrane filters is defined to 0.8 µm.

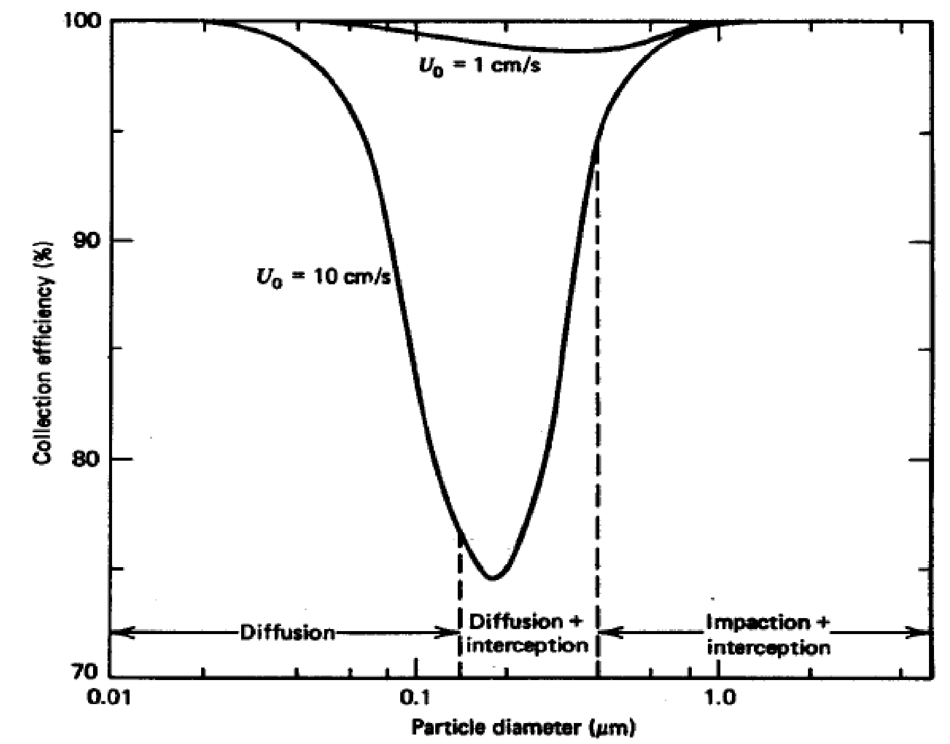

FIGURE 14. Typical efficiency of filters towards particles. It can be seen that almost all particles below 100 nm and above 400 nm are caught in the filter. (Figure is from presentation about filters from Training Course ATM319).

4.5 Salt concentration

As explained in section 4.2, a possible explanation of the different particle number concentrations of ultrafine particles that were measured could be the wick treatment with various water-soluble salts. In order to verify this possibility, the particle matter collected on the filters was tested for different anions and cations by ion chromatography and spectrophotometry respectively.

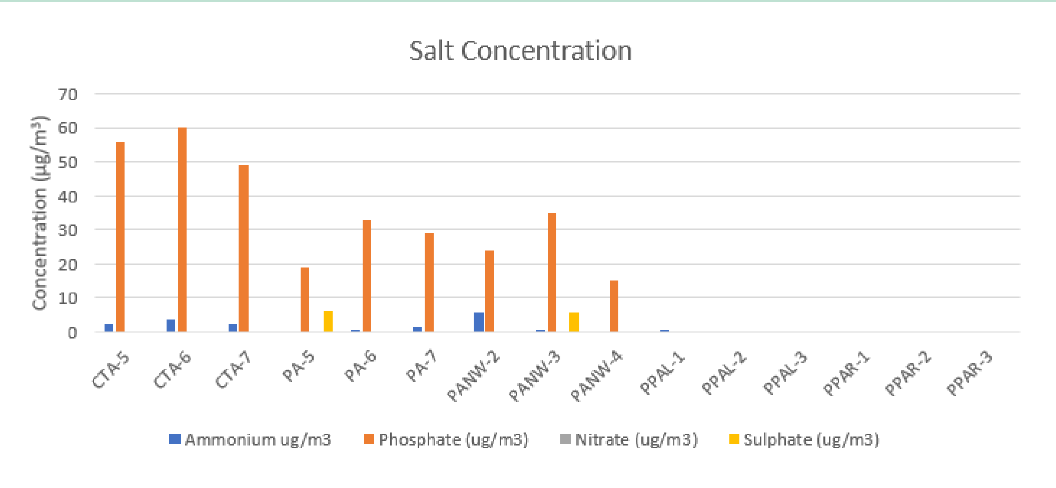

FIGURE 15. Salt concentration measured by analysing the particulate matter collected on filters.

Most of the collected material on the filters is phosphate. Small amounts of ammonium and sulphate can be seen, too. No nitrate is found within the detection limit of the method (approx. 3 µg/m³ for nitrate). A very small amount of ammonium is found on one of the filters collecting particle mass from the PPAL pillar candles, i.e. <1 µg/m³ (detection limit is 0.6µg/m³ for ammonium). Beside this, no soluble salt material was found on the filters emitted from PPAL or PPAR pillar candles. This is somewhat strange however because supposedly the same salts were used to treat the wicks, although in a lower concentration.

TABLE 5. The table shows the average concentration of the salt anions and cations. ~ indicates that it is an estimated sum. * indicates that a sum cannot be given.

Candle type	CTA	PA	PANW	PPAL	PPAR
Ammonium NH_4+ [µg/m³]	2.8	1.1	3.3	<1	<0.6
Phosphate PO_4^{3-} [µg/m³]	55	27	25	<3	<3
Nitrate NO_3- [µg/m³]	<3	<3	<3	<3	<3
Sulphate SO_4^{2-} [µg/m³]	<3	6.1	5.7	<3	<3
Sum [µg/m³]	~58	~34	~34	~0.7	*

TABLE 5 comprises all the average concentrations of salt anions and cations emissions collected on the filters for the different candle types.

Toxicological data for ammonium phosphates and ammonium sulphates established during REACH registration and published by the European Chemicals Agency, ECHA, suggest a comparably low health risk when these substances are inhaled. The Derived No Effect Levels, DNEL, which indicate the levels that should not be exceeded to avoid health effects, for hazards via inhalation route of the general population after long term exposure can be found in table 6.

TABEL 6. DNELs for the general population after long term exposure via inhalation route as provided by ECHA.

Salt	EC no.	DNEL [µg/m^3]
Diammonium hydrogenorthophosphate[3]	231-987-8	1,450
Ammonium dihydrogenorthophosphate[4]	231-764-5	1,800
Ammonium sulphate[5]	231-984-1	1,667

The DNELs established during the REACH registrations are many times higher than the measured salt concentrations emitted by candles.

[3] https://echa.europa.eu/de/registration-dossier/-/registered-dossier/15555/7/1

[4] https://echa.europa.eu/de/registration-dossier/-/registered-dossier/15548/7/1

[5] https://echa.europa.eu/de/registration-dossier/-/registered-dossier/15571/7/1

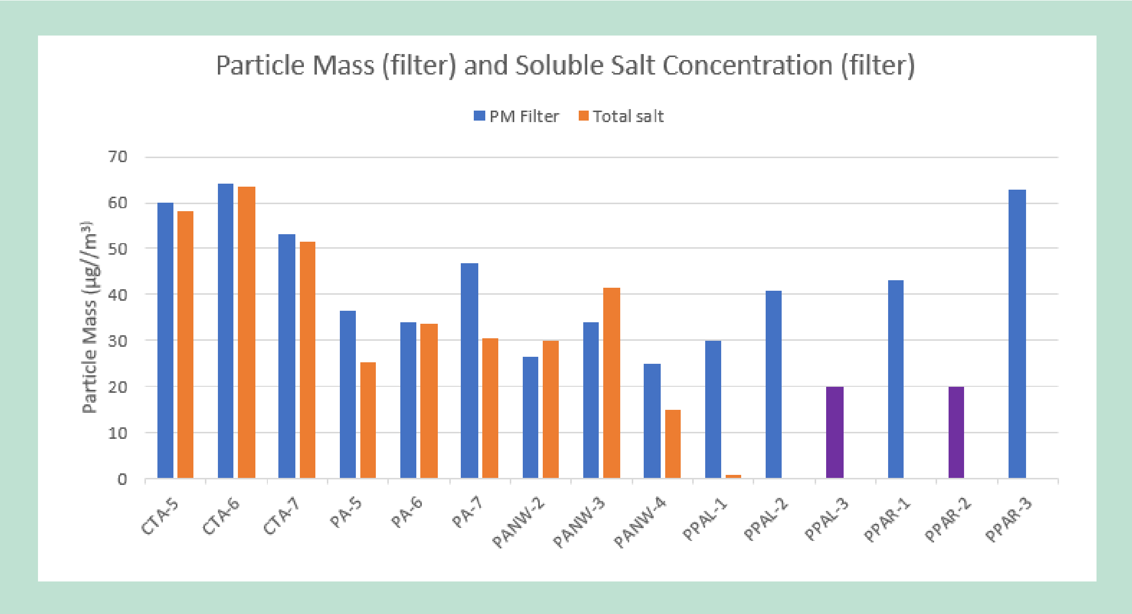

FIGURE 16. Comparison of Particle Mass Concentration and Salt Concentration on filters. For PPAL-3 and PPAR-2, the measured Particle Mass values were under the detection limit of 20µg/m3 and hence the bars are in a different colour.

The collected salt anions and cations on the filters are mostly phosphate and smaller amounts of ammonium and sulphate. When the measured salts are added and compared to the total particulate matter collected on the filters (see FIGURE 16), it can be seen that basically all collected particulate matter consists of the water-soluble salts used for wick treatment indeed, at least for those candles with the strongest wick treatment and the highest ultrafine particle emission (CTA, PA and PANW). The analyses of PANW gives a higher content of salts than total mass collected on the filters which obviously cannot be the case. The results are within the uncertainty of the methods though.

Only very small content of soluble salts has been found in the collected filter mass for the palm stearin pillar candle and the paraffin pillar candle. This is somewhat odd however because supposedly the same salts were used to treat the wicks, although in a lower concentration.

4.6 Black carbon concentration

Black carbon is a good indicator for the amount of soot that is emitted. Black carbon is a term used for combustion particles consisting of only carbon in its pure form. Black carbon is formed when combustion is incomplete and it is part of the emitted soot. Soot is composed of a complex mixture of organic compounds and is a result of incomplete combustion of hydrocarbons, hereunder for instance various candle waxes such as stearin and paraffin. Black carbon is on the WHO's list of IARC Group 2B Carcinogens ("*Possible carcinogenic to* humans") due to its short-term changes in health[6].

The black carbon content of the emitted particles from the burning candles was measured using a microAeth.

[6] http://www.euro.who.int/__data/assets/pdf_file/0004/162535/e96541.pdf

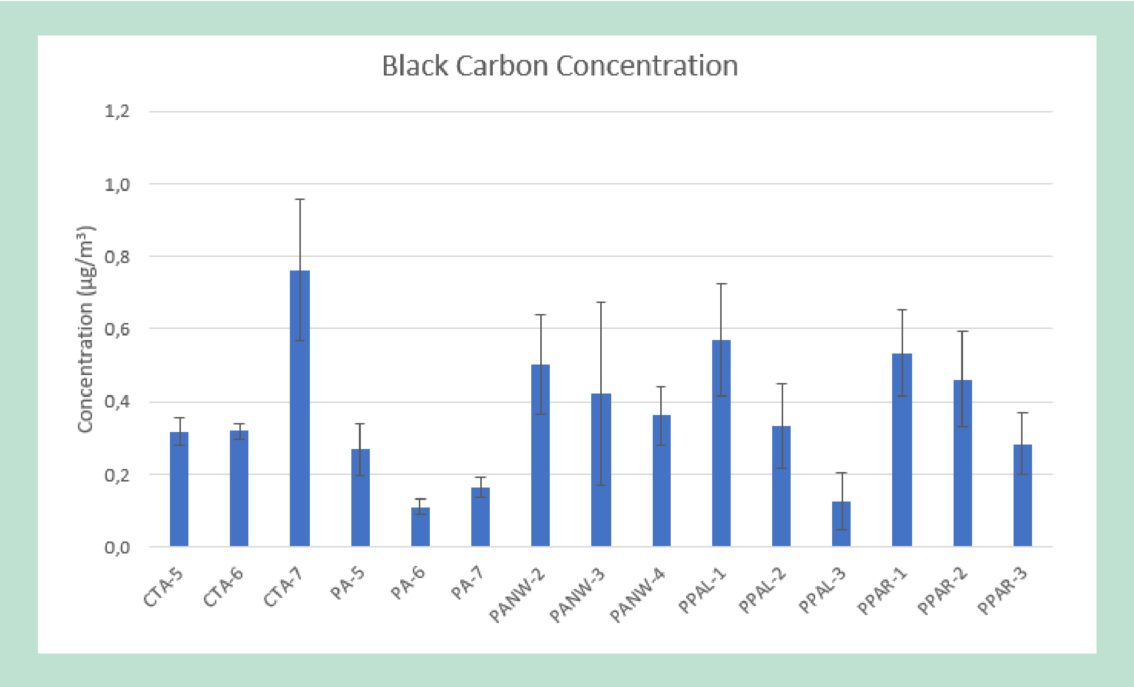

FIGURE 17. The amount of black carbon emitted from each of the measured candles are shown with appertaining uncertainties.

TABLE 7. The table shows the measured average values of black carbon concentration for each candle type.

Candle type	CTA	PA	PANW	PPAL	PPAR
Average black carbon concentration [µg/m3]	0.47	0.18	0.43	0.34	0.43

It is difficult to see a clear trend of black carbon emitted from the different candle types, especially since there was partly significant variation between the individual measurements of one candle type. It can be seen that the reference pillar candle (PA) emits only half the amount of black carbon than the rest of the candles. Substituting the wick and/or fuel in the reference pillar candle PA did not result in a lower emission with regards to black carbon emission.

Comparison of Particle Mass Concentration (DustTrak) and Black Carbon Concentration

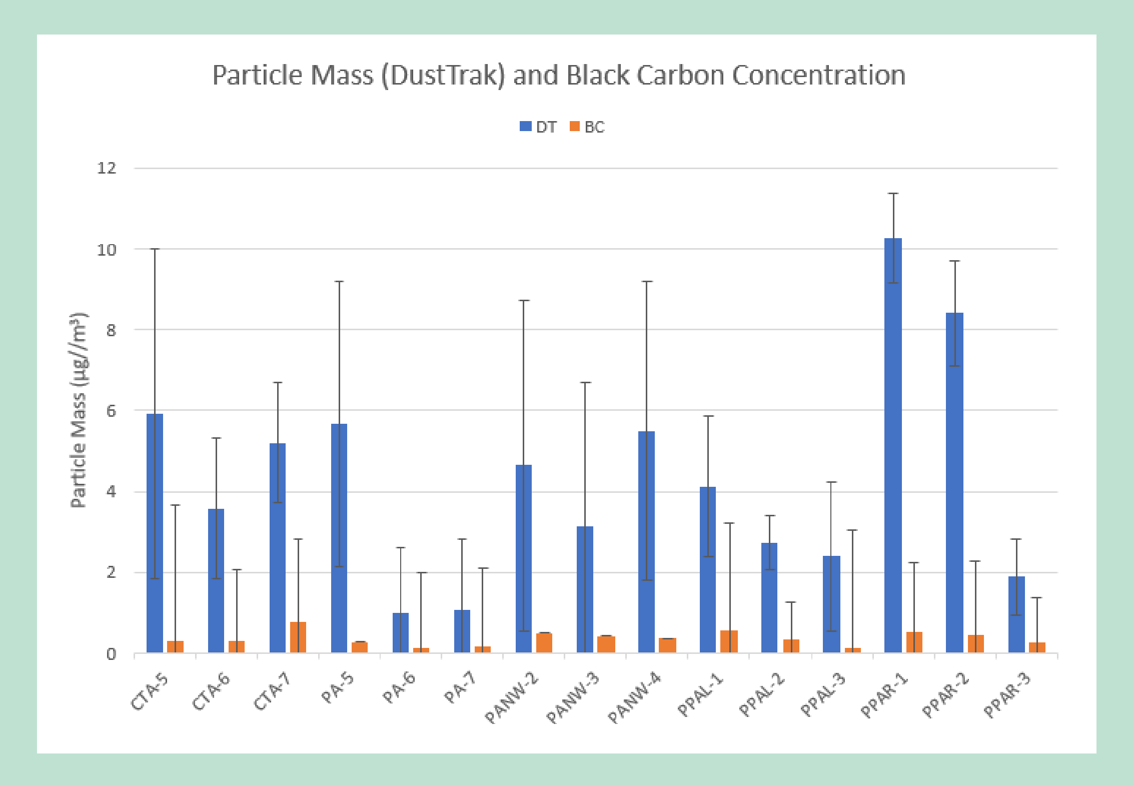

FIGURE 18. Particle mass measured by DustTrak (DT) compared to black carbon mass measured by microAeth (BC). Both are online measurement methods.

Comparison of Particle Mass Concentration (Filter) and Black Carbon Concentration

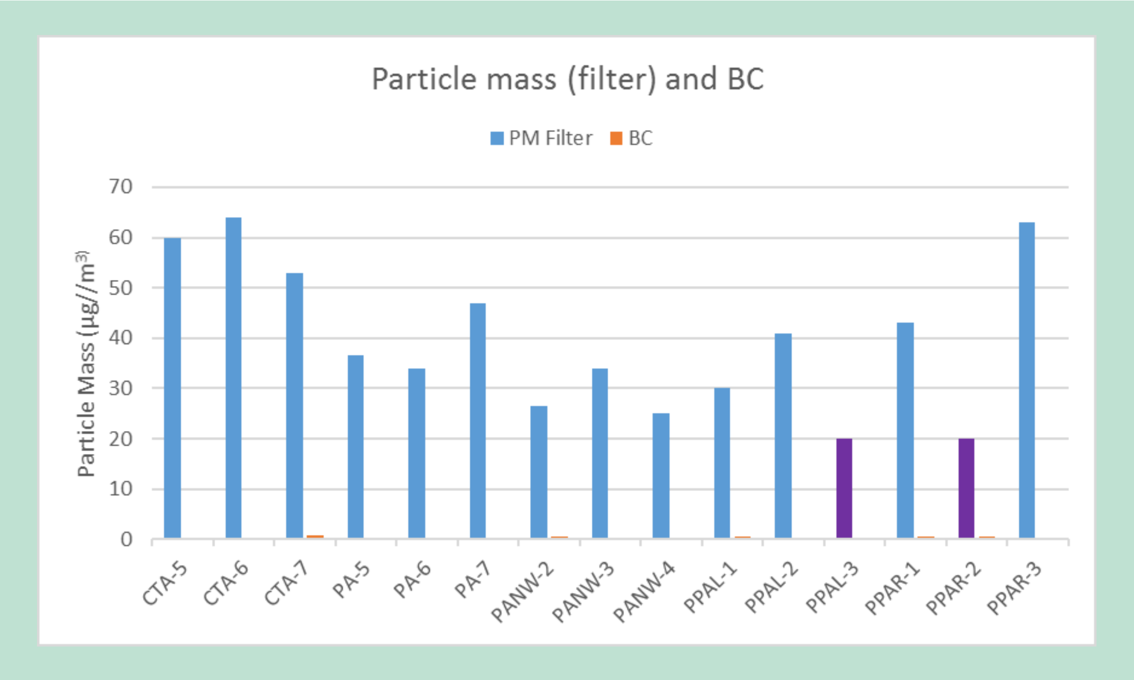

FIGURE 19. Only very low percentage of the total mass collected seems to consist of black carbon.

TABLE 8. The table shows the average black carbon concentration compared to the particulate mass concentration measured by DustTrak and collected on filters.

Candle type	CTA	PA	PANW	PPAL	PPAR
Average black carbon concentration [µg/m3]	0.47	0.18	0.43	0.34	0.43
Average particle mass concentration DustTrak [µg/m3]	4.9	2.6	4.4	3.1	6.8
Percentage black carbon / particle mass concentration DustTrak [%]	9.5	7.0	9.7	11.2	6.2
Average particle mass concentration filter [µg/m3]	59	39	29	36	53
Percentage black carbon / particle mass concentration filter [%]	0.79	0.46	1.5	0.97	0.80

The measured black carbon concentration of all candles is low which indicates a low emission of soot. If the black carbon measurements are compared with the particulate mass measured by DustTrak, the highest black carbon percentage is found for the pillar candles with palm stearin (PPAL 11.2 %) and the lowest for the pillar candles with paraffin wax (PPAR 6.2 %). If the black carbon measurements are compared with the particulate mass collected on filters, the highest black carbon percentage is found for the pillar candles with the alternative wick (PANW 1.5 %) and the lowest for the reference pillar candles (PA 0.46 %).

4.7 Metal concentration

The particulate matter collected on filters was tested for metals for two reasons. Firstly, the salts used for wick treatment could contain metals as cations, such as sodium or potassium for example. And secondly, earlier studies showed certain heavy metals, mostly lead and nickel, in the candle base materials.

TABLE 9. All analyses for metals were under the detection limit, except for iron (Fe) in PPAR-2, which was just above the detection limit.

Metal [µg/m³]	Pb	Sn	Sb	Na	K	Cr	Fe	Co	Ni	Cu	Zn
CTA-5	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
CTA-6	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
CTA-7	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PA-5	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PA-6	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PA-7	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PANW-2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PANW-3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PANW-4	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PPAL-1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PPAL-2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PPAL-3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PPAR-1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PPAR-2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	0.22	<0.2	<0.2	<0.2	<0.2
PPAR-3	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2

No metals, neither possible salt contents such as sodium or potassium nor possible heavy metal contaminants such as lead or nickel were measured in the particulate matter collected on the filters above the detection limit. Only one pillar candle of paraffin (PPAR-2) had a low content of iron just above the detection limit of the analysis.

5. SVOCs and VOCs: semi- and volatile organic compounds

In the following, the analyses of semi-volatile and volatile organic compounds are covered. The VOCs were collected using tubes with Tenax/Carbograph material as adsorbent. After thermal desorption analysis using GC/MS was performed. The 20 most commonly found VOCs were looked for in the GCMS analyses.

For each of the three candles PPAL-1, PPAL-2, and PPAR-3, double sampling using two tubes and two pumps was used in parallel. The sample flow was different in the two pumps in order to secure correct collection of the VOC (e.g. half sample flow should collect the half amount of VOC). This was performed to ensure that the collection was performed correctly and no bias was seen in the results. In total VOCs from 20 L and 40 L air were collected in each tube, respectively. The 40 L of air was the amount of air that was used for collection of VOCs as standard in all the samplings.

The following figures show only the VOCs that exceeded the detection limit.

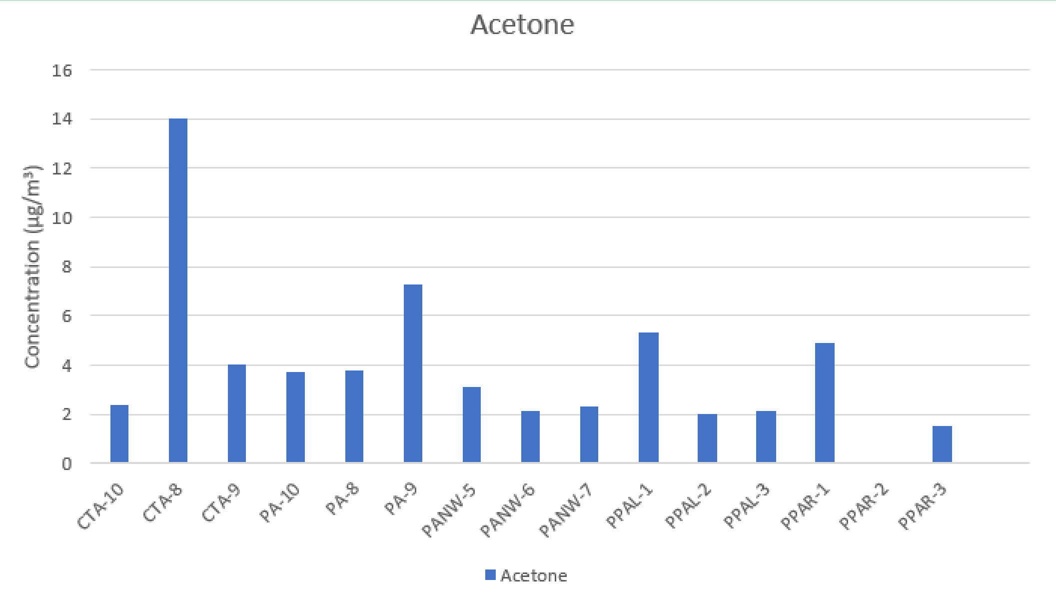

FIGURE 20. The emitted amount of acetone from the different candles.

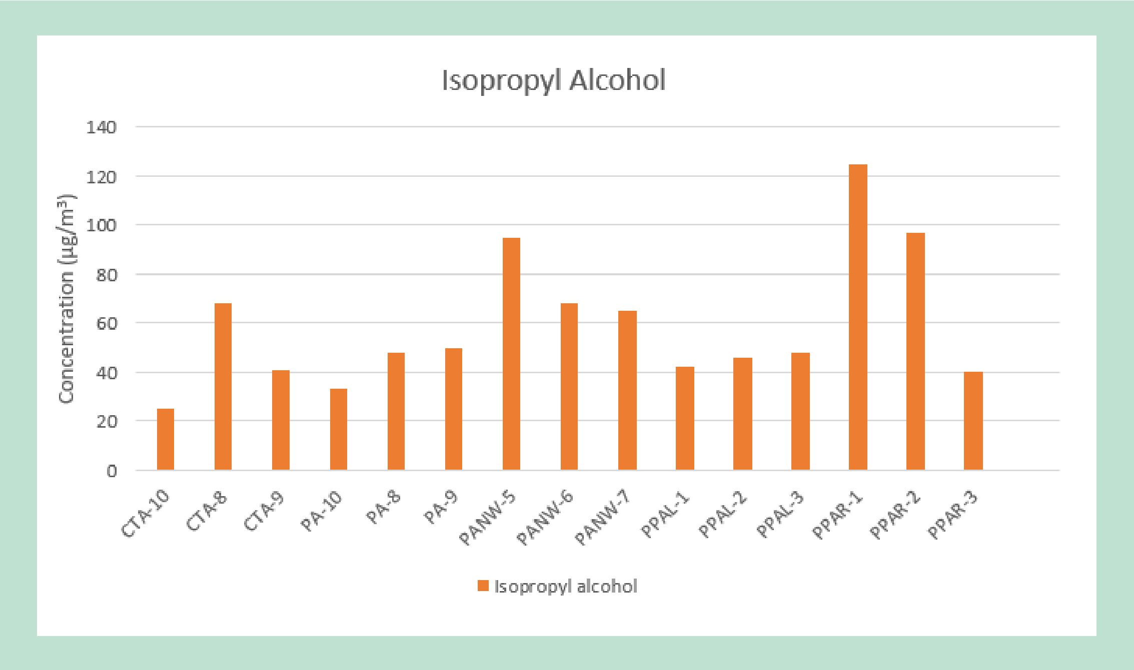

FIGURE 21. The emitted amount of isopropyl alcohol from the different candles.

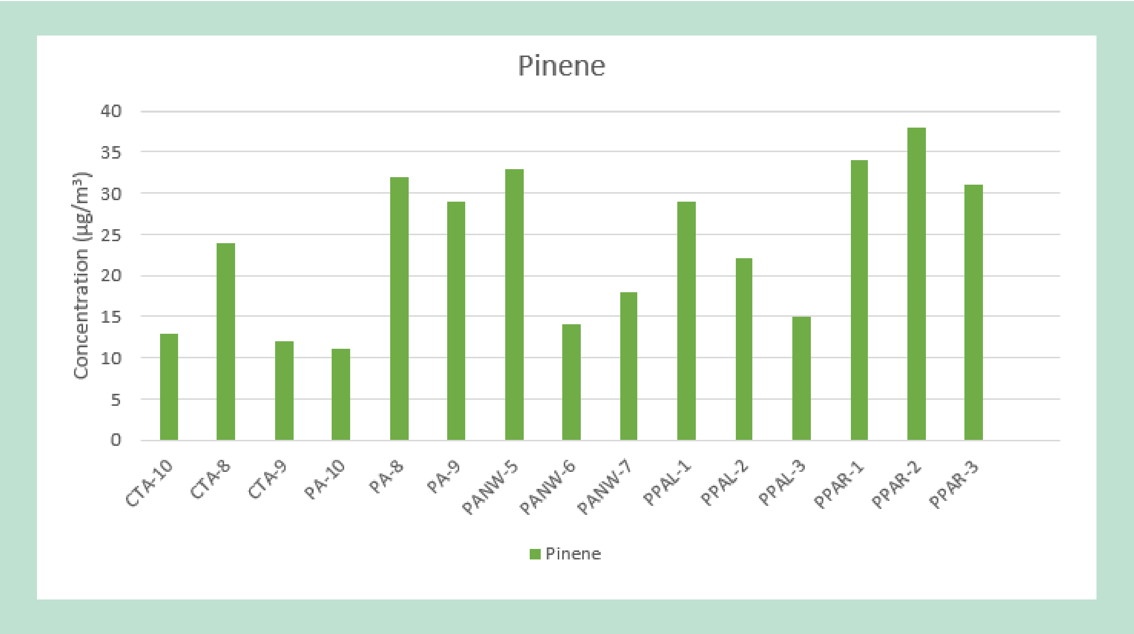

FIGURE 22. The emitted amount of pinene from the different candles. See the comment below for explanation.

Concentrations of Pinene in the emissions from the burning candles have been found in all of the GC/MS analyses. The concentrations are relatively large, since no Pinene was expected. No explanation for this has been found, as Pinene is a typical VOC found in scented candles, and all the analysed candles are un-scented candles. A possible but not confirmed explanation is contamination from outside the climate room.

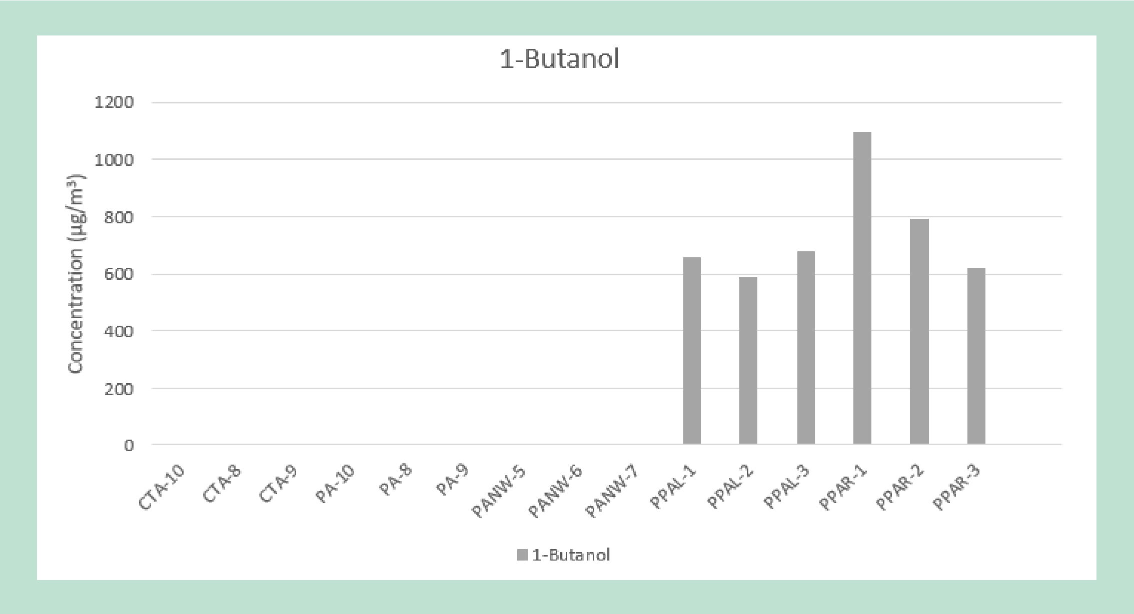

FIGURE 23. The emitted amount of 1-butanol from the different candles.

It can be seen in the graph that large amounts of 1-butanol have been registered from the paraffin and palm stearin pillar candles PPAR and PPAL. This is in striking contrast with the measured values from CTA, PA and PANW candles which are all below 5μg/m³. No further explanation can be giving why these values are so high for the PPAL and PPAR candles, just that the triple determination, as can be seen in the graph, support within a factor of two that the results are correct.

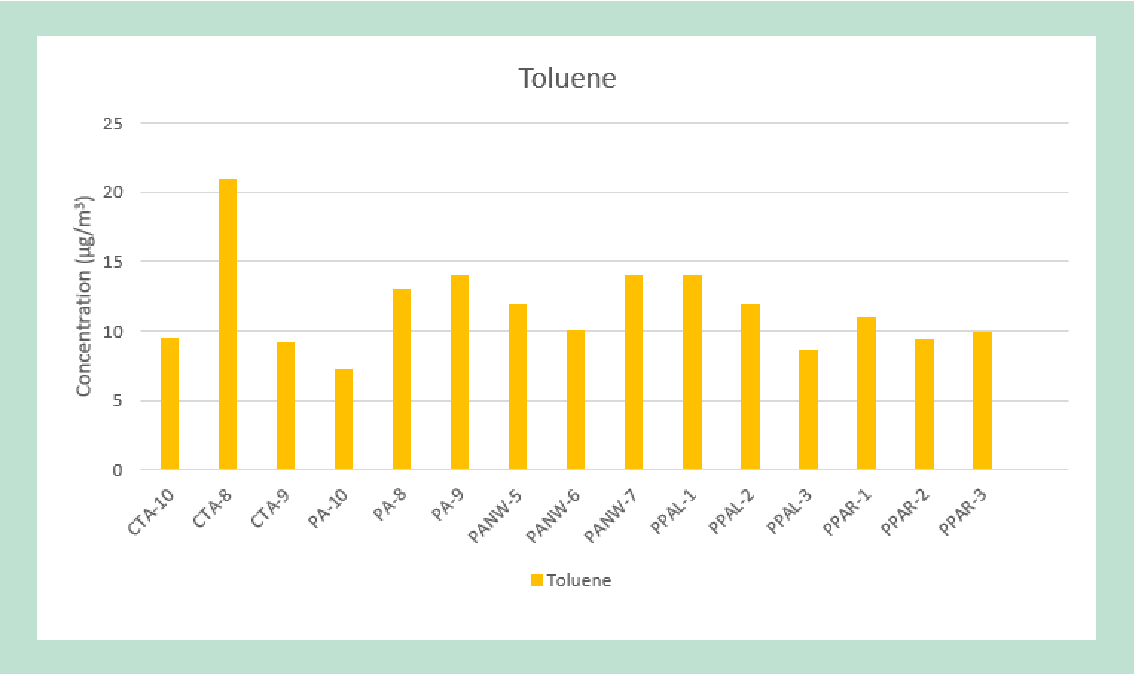

FIGURE 24. The emitted amount of toluene from the different candles.

More or less the same amount of toluene is collected from all of the candles. The amount is similar to what has been seen in earlier studies.

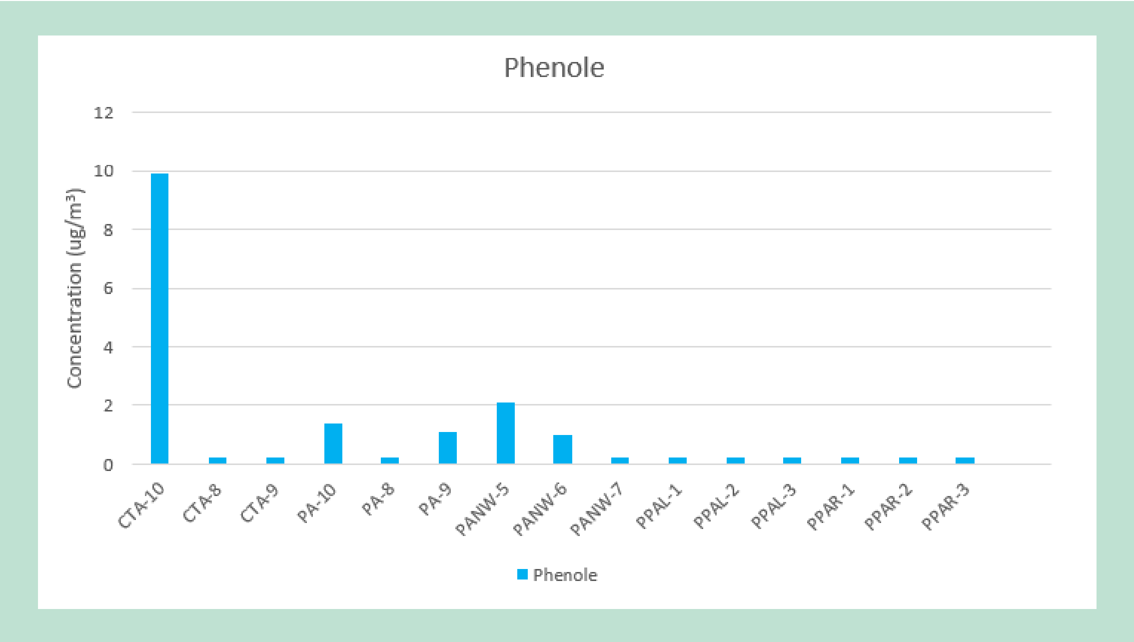

FIGURE 25. The emitted amount of phenole from the different candles.

Only five candles emitted larger amount of phenole than could be detected by the analysis. The remaining candles are shown with the detection limit of 0.2 µg/m³.

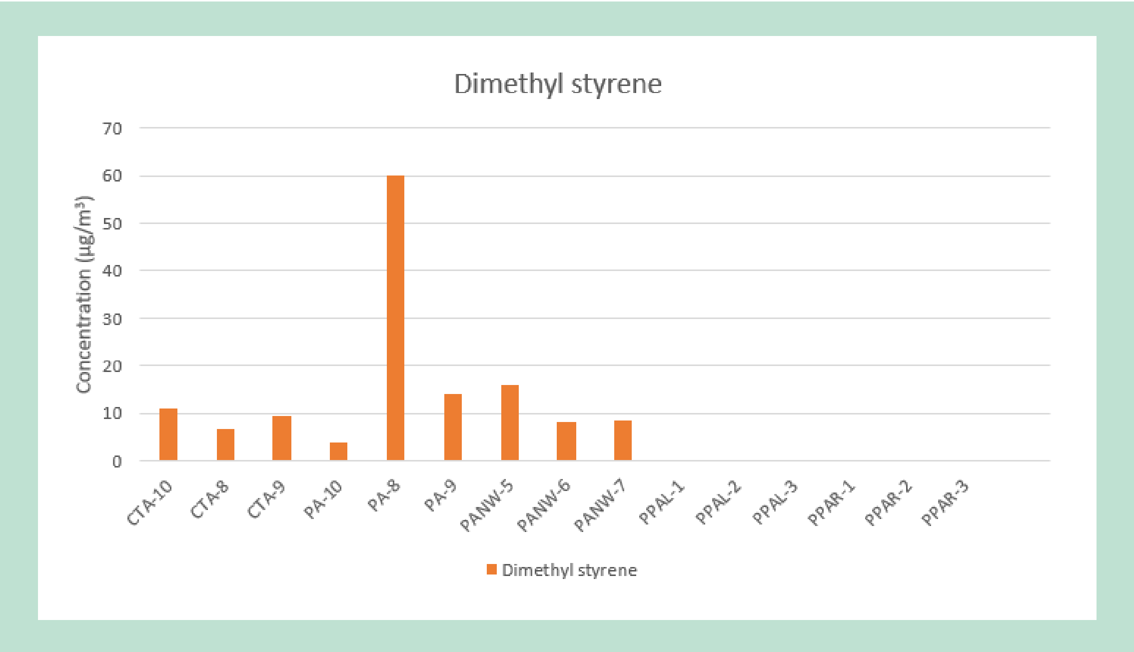

FIGURE 26. The emitted amount of dimethyl styrene from the different candles.

Acetone, iso-propanol, 1-butanol, toluene and phenol are typical combustion products that have been found in the emissions of burning candles before.[7]

[7] https://doi.org/10.1016/j.atmosenv.2012.03.027 ; https://doi.org/10.1016/j.jhazmat.2014.12.040

Pinene was found in the emissions of all candles which is somewhat surprising. Pinene is a frequently used substance in fragrances and scented candles, but the candles tested in this project are all unscented candles. A possible explanation could be contamination with a cleaning agent used for cleaning the lab for example, but this needs to be confirmed.

Dimethyl styrene was found in the emission of the three types of candles made of animal stearin. This was rather surprising since it has never been reported in candle emissions. Generally speaking, the measured concentrations of all the identified substances were low.

It has been attempted to perform time-resolved measurements and VOCs in the air containing emissions from the burning candles using PTR-MS. The advantage of using the PTR-MS to measure the VOC content is to have a method for screening the emission for various substances and to have the resolution in time at the same instant, respectively. However, during the project, it proved very challenging to interpret PTR-MS data obtained from burning candle emissions. Therefore, it is not possible to conclude on the VOC emissions based on the PTR-MS measurements with high enough certainty to include the PTRMS results in the report.

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6. Assessment and Discussion

6.1 Discussion

In recent years, there have been a number of reports about comparably high concentrations of fine particles measured in the air of rooms in which candles were burning. The size of these particles was small enough that the particles can get into the pulmonary alveoli of the lungs if they are inhaled. It is known that persistent particles, i.e. insoluble particles that cannot be easily excreted by the body, from outdoor sources, such as diesel cars, tyre abrasion or dust, can have a negative effect on human health. Although little is known about the effects of particles from indoor sources, such as cooking, vacuuming or candle use, particle emissions of candles are often compared with those of massive traffic.[8]

Denmark is the country with the highest per head consumption of candles in the European Union. Danish consumers seem unsettled by the negative press coverage and have used less candles in the past years.

This project was initiated in order to develop candles with a lower emission of particles, starting with a systematic and scientific investigation of the candle emissions and the potential health risk involved in using candles. As a first step in the project, the project partners were supposed to measure the emission of particles and other substances from commonly used burning candles with state-of-the-art test equipment to get a better understanding of what substances are emitted and at which levels. Starting from this basis, the partners were supposed to develop a new type of candle with similar performance but lower emission of (health-relevant) particles.

Two of the most common candle types in Denmark - uncoloured and unscented crown top candles with a diameter of 23 mm and a height of 200 mm and pillar candles with a diameter of 58 mm and a height of 120 mm, both made of 100 % animal stearin – were picked as reference candles. The most relevant emissions were measured in order to establish the state of the art with regards to candle emissions. The combustion process of candles, and therefore the emission, is complex however with a lot of potential influencing factors. Even two individual candles coming from the same batch sometimes show a significantly different burning performance. Hence, after seeing the first measurement results, it was decided that triplicate measurements for each candle type were necessary to balance differences between individual candles in order to allow a general statement for a certain candle type.
It is a very comprehensive work to do all measurements and analyses on every candle to see the big picture of emissions, chemical composition, etc., wherefore one candle type was chosen to be investigated for influence of using a different wick and/or fuel on the emission. This is why pillar candles were chosen as they have better use for more different fuels and blends. A total of consecutively three modified pillar candle types were tested, all with the same dimensions as the reference pillar candles.

[8] https://videnskab.dk/krop-sundhed/nye-maalinger-partikler-fra-stearinlys-kan-vaere-lige-saa-farlige-som-dieseludstoedning
https://www.bt.dk/forbrug/undgaa-at-levende-lys-forurener-din-stue

The first modified candle type was made with the same fuel as the reference candle, but a different wick. The second modified candle type used a different fuel – stearin from palm oil instead of animal stearin – and a different wick which is necessary because of the different fuel. The third modified candle type used paraffin wax instead of animal stearin and again a different wick. It is important to underline that the wick type is highly dependent on the choice of fuel type, as mentioned earlier. Therefore, it is not possible to create a candle with a completely new fuel type using the same wick type. This would have a negative influence on the performance and possibly safety.

6.1.1 Measurement results

The main focus of this project was on measuring particles, both ultrafine particles (number concentration in size range approx. 4-166 nm) and larger particles (mass concentration). The size distribution of the ultrafine particles showed no significant differences between the reference crown top candles and the different pillar candles. All candles emitted particles in the range between 5 and approx. 120 nm, with a peak at approx. 20 nm for the crown top candles and at approx. 10-15 nm for the four different pillar candles. Based on these results, neither the shape nor the wick or fuel seem to have a significant influence on the size distribution.

There are differences in the number of emitted ultrafine particles measured as room concentration, however. The crown top candles showed the highest particle number concentration (approx. 430,000 particles/cm^3), and the modified pillar candles made of paraffin showed the lowest particle number concentration (approx. 104,000 particles/cm^3). One probable explanation is the different burn rate of the candle types, i.e. the mass of fuel burned per hour. A general trend is that more particles are emitted the more fuel is burned. The pillar candles made of animal stearin but with a modified wick do not follow this trend however. The second probable explanation is that the more salts are used to protect the wick from corrosion the more ultrafine particles are emitted. This will be covered in more detail later.

The total mass of the emitted particles was measured with two different methods, with an online test device that works in the size range of approx. 0.3 to 15 µm and with filter collection that is the official method for measuring particles in ambient air and covers all particle sizes (particles larger than 15 µm is not expected from a steady burning candle, though, and particles less than 0.3 µm in size have a comparably little mass).

The online measurements showed comparably low total particle masses between 3 µg/m^3 for PA (the reference pillar candles) and 7 µg/m^3 for PPAR (the modified pillar candles made of paraffin). It must be noted however, that the low mass emissions measured are all close to the detection limit of the method which gives rise to higher uncertainty of the individual results.

The filter measurements showed higher total particle masses between 59 µg/m^3 for CTA (the reference crown top candles) and 29 µg/m^3 for PANW (the pillar candles made of animal stearin with a modified wick). The reference pillar candles (PA 39 µg/m^3), the pillar candles made of vegetable stearin (PPAL 36 µg/m^3) and the pillar candles made of paraffin (PPAR 53 µg/m^3) ranged in between.

As mentioned before, this study aimed not only at measuring the number, size and mass, but also at working out the composition of the emitted particles. For this reason, the particles collected on the filters were tested for soluble salts, black carbon and content of metals.

The measurements showed that in case of CTA, PA and PANW – these were the candle types with the highest emission of ultrafine particle - almost the entire particle mass consisted of the water-soluble salts (ammonium, phosphate, sulphate) that are typically used for wick protection. This strongly indicates that salts are the largest fraction of the emitted particles and the

results are consistent with the findings of Pagels et al[9]. It is also an important finding for assessing the potential health risks of burning candles.
The emission of black carbon, which is an indicator of soot and therefore of incomplete combustion and which is also relevant for the assessment of health risks due to its possible content of critical substances like polycyclic aromatic hydrocarbons (PAH), was relatively low. Between 0.46 % (PA) and 1.5 % (PANW) of the particle mass collected on filters consisted of black carbon, and between 6.2 % (PPAR) and 11.2 % (PPAL) of the particle mass measured online. Since black carbon is a known carcinogen, the emission should be as low as possible. Here, the reference pillar candle PA distinguishes itself.

The measurements of metals such as sodium and potassium, which could be components of the salts used for wick treatment, and lead and nickel, which could be contaminants in the fuel and wicks, showed no contents above the detection limits. Neither did any of the other metals which were analysed for (Sn, Sb, Cr, Co, Cu, Zn). Only a very small amount of iron was found in one analysis of emitted particles from one of the paraffin pillar candles (0.22 µg/m³ in PPAR).

The last group of substances measured in the emissions of the candles were VOCs and SVOCs. Typical combustion products, such as acetone, isopropyl alcohol and toluene were found in the emissions of all candles at low levels. Other combustion products, such as n-butanol and phenol were only found in the emissions of some candles, again at low levels. A low content of pinene was found in all samples, but the content of this in unscented candles cannot be explained at this point in time. Furthermore, content of a styrene derivate was found in the analyses of the emissions from PPAL and PPAR candles.

Unfortunately, it was not possible to modify the reference candles in a way that all emissions decreased simultaneously. Some candle types showed lower ultrafine particle emission, others a lower emission of particle mass or black carbon and yet others lower VOC emissions. This underlines that it is not a trivial task to develop new low emission candles when looking at the emissions in a broader perspective. Within the limited financial resources and timeline of this project, it has not been possible to conclude this objective.

A more thorough investigation with a more specific focus on identifying and measuring health relevant emissions, would be a future perspective for achieving improved candles based on health considerations. Fundamental knowledge about the effect of the various candle components on the combustion process, and if or how these affect the human health, is yet largely unknown. Improved scientific understanding of candle combustion could thus pave the way for improved candles and form the basis for a classification of candles. This work forms the basis for further exploration of these effects and development of low emission candles in a large project funded by Innovation Fund Denmark: CANDLE[10]

6.1.2 Health aspects

A comprehensive assessment of possible health risks of candle use is not possible within the limitations of this project giving relatively limited measurement data. On the one hand, the emission data of a much larger number of candles would be required for a systematic assessment. And on the other hand, a human exposure study would help to find out if the emissions of particles and other substances from different types of candles have different effects or any effects at all. Both was not possible within the scope of this study.

[9] https://doi.org/10.1016/j.jaerosci.2008.10.005

[10] https://innovationsfonden.dk/en/node/1720

Some important new knowledge was established with this project however which should be taken into account for assessing possible adverse effects of candle burning on human health:

The most interesting finding is that the particles emitted by the candles with the highest emission of ultrafine particles (CTA, PA, PANW) consist mostly of the water-soluble salts used for wick protection (ammonium phosphates and sulphates). Ultrafine particles can get into the pulmonary alveoli of the lungs when inhaled, and it is these that experts are normally most concerned about from a health point of view. If these particles are persistent, i.e. if the body cannot excrete them easily, they can cause severe health problems, such as respiratory or cardiovascular diseases, especially with people who suffer from health issues already. The measurements have showed however that the majority of particles from candles are water-soluble and therefore non-persistent, i.e. they dissolve when they get in contact to humid surfaces and can supposedly be excreted easily from the body. This has to be verified by human exposure studies, though.

Another important finding is that the particles emitted by the candles in this study have a relatively low content of black carbon which is an indicator of soot and also a health concern. The percentage of black carbon in the particle emissions from other sources, such as diesel exhaust[11], is much higher.

6.2 Uncertainties and limitations

The study was performed with a limited number of different candle types that burned without disturbance by draught. It is therefore unclear if the obtained data for these candles is representative for other candles, e.g. coloured candles, scented candles or candles with other dimensions or composition. It is expected that burning candles in draughty conditions would lead to a different particle emission and composition, as observed in Pagels *et. al.*[12]

However, the study investigated the emissions of two of the most commonly used candle types in Denmark. The measurements have been performed in a climate room to simulate normal exposure levels of particles in a small room. Under the assumption of candle use in private homes protected from draught, e.g. by open windows or heat sources such as convectors close by, one can expect similar emissions in reality.

6.3 Improving knowledge

The following aspects are considered essential in order to achieve increased knowledge with regards to the emissions of candles, the health effects of the emissions and the possibilities to reduce the (health-relevant) emissions:

Emission

- Testing more different candle types for their emission and to increase representativeness. The chemical composition of the emitted particles is a very important aspect.
- Influence of colour, scent, etc. on the emissions.
- Influence of draughty conditions (non-optimal burning) on the emissions.

Health effects

- Performing *in vitro* tests with human blood and tissue and *in vivo* tests with human panellists exposed to candle emissions to see if there are any health effects. Both long-term effects and acute effects are of interest.
- Toxicological evaluation of the different groups of emissions and weighting of the possible effects on health.

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[11] https://doi.org/10.5194/acp-14-7585-2014

[12] https://doi.org/10.1016/j.jaerosci.2008.10.005

Possibilities to reduce (health-relevant) emissions

- Further knowledge regarding which modifications (dimensions, wick, fuel, etc.) cause which changes in the emissions. Testing more different candles with systematic modifications will be required for this.

6.4 Conclusion

Thorough measurements of the emissions of two very common candle types in Denmark, crown top candles and pillar candles made of animal stearin, and three modifications of the pillar candles with regards to the wick and/or fuel have provided some important new findings for a better understanding of the emissions and their possible effect on consumers´ health. The size distribution of ultrafine particles seems to be very similar for all candles, regardless of type, wick or fuel. Compared to other particle sources, the particles emitted by burning candles are rather small, in a size range of approx. 5-120 nm. The number of emitted particles seems to be higher the more fuel is burned per hour and the more salts are used to protect the wick from corrosion, and therefore indirectly what kind of fuel is used. Most ultrafine particles are emitted from the three types of candles made of animal stearin, the least from pillar candles made of paraffin.

The highest particle mass is emitted by the crown top candles again, followed by the pillar candles made of paraffin and the reference candle made of animal stearin.

The examination of the composition of the particles shows two important things. Firstly, the content of black carbon is relatively low, indicating a low emission of soot by the candles. This comprises with the steady burning circumstances under which the tests have been performed. Consumers can support this by using high-quality candles, trimming the wick if it gets too long and protecting the candles from air flows that make the candle flame flicker. Secondly, the particulate matter emitted by the candles made of animal stearin almost completely consists of the water-soluble salts used to protect the wick from corrosion. It can be assumed that the particles dissolve easily when they are inhaled and get in contact with the humid surfaces of the respiratory tract. This property distinguishes them from the persistent particles emitted by other sources, e.g. diesel engines, tyre abrasion or dust. Additional measurements of further candle types should be made to confirm these findings, preferably supplemented by toxicological exposure studies of human cells and human panellists. But based on these new findings, direct comparison of particles from candle burning with those from other sources, such as traffic, does not seem to be appropriate.

The tested candles do not emit any metals. Certain VOCs are emitted but at low levels consistent with finding in other studies.

Appendix 1. Sooting test

Example of sooting test report

Promol — Sample ☐ Production ☒ Test ☐

Sooting Test

Standard: EN 15426:2007
Report nº:
Date:

Press ☒ Dipping ☐ Filling line ☐ Tealights ☐ Extruder ☐ Maxis ☐

Mixture: 100% Stearin Pavio: PW48XXL Massa de Banh 14 Côr: White Perfume / Serigrafia/Verniz: NA

Candle: 100% Stearin, ASP-HOLMBLAD

Sample	Height mm	ø mm	Weight g start	Weight g end	mass consumed g	Measuring Period 1 start time		Measuring Period 1 end time		Measuring Period 2 start time		Measuring Period 2 end time		Measuring Period 3 start time		Measuring Period 3 end time		Tempo de Queima Total (min)	Massa Consumida g/time + 5 min por periodo de estabilização	Ev, sooted glass plate	Ev, cleaned glass	Soot index	Average Soot index per hour
1	120	58	251,6	184,0	67,6	9	30	13	30	8	30	12	30					480	8,2	14920	14991	0,47	0,06
2	120	58	251,4	185,1	66,3	9	30	15	30	8	30	12	30					480	8,0	14942	14990	0,32	0,04
3	120	58	251,6	184,9	66,7	9	30	13	30	8	30	12	30					480	8,1	14931	14985	0,36	0,05
																							0,05

Tolerance acording to EN 15426:2007

Average soot index from three tests shall be < 1.0/h
Every sample shall be < 2.0/h

Appendix 2. Burning test

Example of burning test report

Relatorio de Teste
Test Report / Delivery control

Report Nº: PROMOLLABID030
Date: 22/11/2017

Data recepção *Date Good receipt*	16/11/2017	Photos
Vela / *Candle*	Cilindrical, 58x120	
Mistura / *Misture*	100% 2468	
Pré-banho / *Pre-dip*	58/60	
Banho / *Dip*	14	
Coto / *Pressed core*		
Pavio / *Wick*	PW 48 XXL	
Côr / *Colour*	White / Red / Black	
Fragância / *Fragrance*		
Fornecedor / *Supplier*	Promol	
Objectivo / Objective	Burning Performance with 100% 2468 with wick PW 48 XXL	

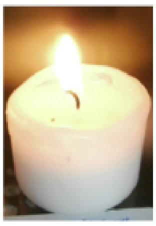
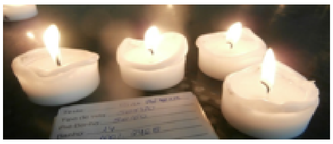
Ciclos de Queima RAL	X	Sim *yes*		Não *no*		Sem requisito *not requested*

Resultado do teste / Test results

Cor *Colour / design*	Peso Inicial *Weight* (g)	T. Queima *burning time* (h)	Peso final *end weight* (g)	Parafina residual *residual* (%)	ok	nok	Consumo *consumption* (gr/h)	Altura Chama *flame height* (mm)	Emissão Fumo *sooting* S	N	Escorrimento *dripping* S	N
White	245,01	33,15	24,76	10,11	X		6,64	38-45		X		X
White	245,03	33,15	23,84	9,73	X		6,67	38-45		X		X
White	244,38	32,15	21,05	8,61	X		6,95	38-45		X		X
White	244,99	32,15	23,95	9,78	X		6,88	38-45		X		X
White	245,35	32,05	26,17	10,67	X		6,84	38-45		X		X
White	244,50	29,55	26,28	10,75	X		7,38	38-45		X		X
Red	246,88	31,15	42,21	17,10	X		6,57	35-43		X		X
Red	247,66	31,15	39,61	15,99	X		6,68	35-43		X		X
Red	247,22	31,15	26,49	10,72	X		7,09	35-43		X		X
Red	246,81	31,15	37,41	15,16	X		6,72	35-43		X		X
Red	248,71	31,15	35,42	14,24	X		6,85	35-43		X		X
Black	247,31	33,15	19,35	7,82	X		6,88	33-42		X		X
Black	248,51	33,15	20,76	8,35	X		6,87	33-42		X		X
Black	248,48	33,15	22,66	9,12	X		6,81	33-42		X		X
Black	247,23	33,15	14,53	5,88	X		7,02	33-42		X		X
Black	249,00	33,15	19,80	7,95	X		6,91	33-42		X		X
Average	246,69	32,11	26,52	10,75	X		6,86	33-45		X		X

Photos / Comment: Good performance. No oscillations in flame between cycles.




X	Aprovado *approved*		Não Aprovado *not approved*		Standby

Environmentally friendly candles with reduced particle emissions

To support the development of low emission candles a series of emission measurements were conducted on a number of candles commonly used in Denmark. Here, it was observed, that the number of emitted particles and total particle mass depends on the burn rate and the amount of salts necessary for wick protection. The studies also show, that when candles are protected from draught, the level of black carbon is very low and a large part of the emitted particle mass consist of the salts used for wick protection. Measurements of metals, such as lead and nickel under similar conditions, showed no emissions above the detection limit, and the measurement of emitted (semi) volatile organic compounds, (S)VOC, was unremarkable. Modifying the composition of these candles resulted in a change of the observed emission, but a candle composition exhibiting reduction of all emissions was not achieved.

For at understøtte udviklingen af et lavemissions levende lys, er der foretaget emissionsmålinger på en række lys typer der ofte benyttes i Danmark. Her er der observeret at emissionen af partikler primært afhænger af hastigheden hvormed lyset brænder og af den imprægnering der er anvendt til vægebeskyttelse. Undersøgelserne viser også, at hvis lysene beskyttes mod træk, så er carbon black-niveauet, som er en indika-tor for sod, meget lavt, og mere eller mindre alle emitterede partikler fra de levende lys består af salte benyttet til vægebeskyttelse. Målinger af metaller, som fx bly og nikkel uden samme forudsætninger, viste ingen emissioner over detektionsgrænsen og der var heller ingen bemærkninger til målingen af afgivne (halv) flygtige organiske komponenter (S)VOC. Modificering af kompositionen af disse lys resulterede i ændringer i den observerede emission, men et lys med en komposition der reducerer emissionerne på alle parametre blev ikke fundet i dette projekt.

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