

Characterization of Pollutants Emissions from Burning Candles

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1. INTRODUCTION

Both indoor and outdoor sources contribute to and affect the concentration and composition of particles in indoor air. There is usually more information available on emission characteristics, such as emission factors or emission rates, of outdoor pollutants sources than of indoor sources. However, quantification of emissions from indoor sources is very important for assessment of total human exposure to pollutants. Due to several indoor activities different pollutants, gaseous species and particles, are emitted and concentration levels indoors could be temporarily or even for prolonged periods of time elevated up to tenfold compared to the situation without the sources. This can have a significant effect on human health. The most significant sources, for indoor environments, include tobacco smoking, cooking, kerosene heating and wood burning [1,2]. Combustion processes are the main indoor sources of smaller particles, with the vast majority of them in the submicrometer range, containing a host of organic and inorganic material [3]. Gaseous pollutants play also an important role because they can affect directly the human health or they can act as precursors of secondary particles in the indoor environment, through the process of gas-to-particle conversion [4]. Emissions from tobacco smoking have attracted considerable attention and as a result there is more information available on emission rates from this, compared to other indoor sources [5].

Scented candles have gained popularity over the past 30 years resulting in the current abundance of candle shops and aromatherapy candle products. U.S. candle sales reached \$2.1 billion in 1998 and are growing at 10 to 15 percent annually [6]. With the increase in candle use, comes public concern about potential health effects due to the exposure to candle emissions and to fragrances. For the sake of example, the production of candle soot has been implicated as a possible source of a phenomenon called black soot deposition. Soot is a product of incomplete combustion of carbon-containing material and can appear as a gray to black colored substance. The number and type of wicks and the candle scent may also play a major role in the type of particulates that are emitted in the soot. The composition of the candle and the fragrance will also determine the number and the amount of emissions produced. It is a common opinion that scented candles, jar candles, and oil candles appear to emit a higher soot output than standard wax candles. Other emissions of concern produced from burning candles include carbon monoxide (CO), carbon dioxide (CO₂), nitrogen oxides (NO_x), aldehydes, unburned/partially burned hydrocarbons, specifically polycyclic aromatic hydrocarbons (PAH) which are well known for their cancer-causing potency.

Only few studies are available in literature concerning the emissions from burning candles; in particular emissions of lead [7,8], zinc [9] or soot [10-12], produced by the heating of candles wick cores or by the wax combustion/evaporation were evaluated, while less researches have been focused on the presence of volatile organic compounds in candle emissions [13-15].

The purpose of this work was to determine the emissions of selected organic compounds, such as PAHs, BTEX and some aldehydes, as well as the concentrations of main gaseous combustion products in the exhaust fumes of five scented candles.

2. EXPERIMENTAL SECTION

2.1. Test conditions

Since a crucial point in the determination of the emissions concentration was the simulation of realistic conditions for burning candles, a special enclosed chamber was developed to ensure defined burning conditions and the possibility to perform a gas sampling of the exhausts without turbulences. The test chamber exhibit a volume of about 170 l, and it is equipped with an air sparger, set on the bottom of the equipment, to supply the inlet-air to the chamber with minimum turbulence and spatial velocity. For all experiments pre-cleaned air, passed through a charcoal trap, was used; moreover, to ensure the filter effectiveness during each run a blank measure has been also made on the air fed to the chamber. The air flow rate in the test chamber was adjusted in order to reach realistic burning conditions; in particular, the total flow at the air-outlet has been set at appr. 15 Nl/min. Furthermore, equipment to monitor temperatures, into the candle body, at the chamber wall and roof and within the stack, respectively, as well as an equipment for monitoring main combustion products (O₂, NO_x, CO and CO₂) of the exhaust fumes in the air-outlet completes the test system.

The candles are to be placed upright, spaced adequately from one another in keeping with the candle format, so that the candles do not transmit heat to each other, thus avoiding an undesirable thermal influence. The locations of the individual candles inside the chamber were fixed and recorded in detail. So, for every fragrance tested, candles were located at the same position in the chamber. To determine the wax consumption every candle was weighted before and after the burning experiment. The corresponding burning time was recorded. To control comparable burning conditions between real room situation and the test chamber, the wax consumption in a real room was determined for every individual candle, too, as summarized in figure 1.

Coloured and scented commercially available candles made of a mixture of paraffin (78%), stearin (20%) and a fragrance (2%), provided by a cotton wick (without a sustainer), have been tested. The candles with a length of 100 mm, a diameter of 50 mm and an approximate weight of 155 g were finished, in the external part, with about 7 g of microcrystalline wax and 8-50 mg dye. In particular, scented candles containing the fragrances of aloe, bark, opium, frangipani and rhubarb were studied.

2.2. Analytical methods

Several preliminary TGA/DTA analyses were performed on both raw materials and candles (taking samples in different positions) in order to highlight the materials properties once they are heated up by the flame.

For the chamber experiments four candles were burnt simultaneously; this allows to control the temperatures within the test equipment and to reduce possible heterogeneity and differences among candles that can burn in a slightly different way. Resulting exhaust gases were sampled and analyzed to evaluate the concentrations of polycyclic aromatic hydrocarbons (PAHs), BTEX, as well as short chain aldehydes such as formaldehyde, acetaldehyde, propionaldehyde and acroleine. In particular, for sampling and analysis of the short chain aldehydes the DNPH method (Dinitrophenylhydrazine) was applied. By means of a gas sampling pump with an included gas meter a defined volume of exhaust gas was passed through a cartridge containing the DNPH sorbent. The aldehydes are reacting quantitatively with the 2,4-dinitrophenylhydrazine to the corresponding hydrazone compounds. The hydrazone-derivates could be measured by HPLC. According to the limited sampling capacity of the DNPH cartridge a partial flow of about 0.5 l/min and a total volume of about 15 liters was collected. For analysis the DNPH cartridge was desorbed using acetonitrile, aldehydes were analyzed using HPLC and a diode array detector at a wavelength of 365 nm. The limit of quantification for the individual aldehyde was in the range of 0.1 µg/g wax.

To evaluate the amounts of benzene, toluene, ethyl-benzene and xylenes (BTEX), a defined volume of exhaust gas was passed during an adsorption cartridge including a charcoal sorbent (ORBO TM32). The sorbent cartridges were completely desorbed by means of CS₂ and fluorotoluene was added as internal standard. For sampling a flow rate of appr. 1 l/min was used and a total flow of approximately 240 l were collected. Then, the sample has been analyzed in a gas chromatography provided by a FID detector. Limits of quantification for individual compounds were in the range of 0.01 µg/g wax.

Lastly, PAHs have been sampled with a combined particle/gas phase system constituted by a PTFE filter (diameter of 47 mm, pore size of 0.2 µm) followed by an adsorption cartridge filled with a XAD2 adsorbent; particles are collected at the PTFE filter surface while gas phase constituents are collected within the XAD2 adsorbent. The sampling line was located in the air-outlet and was connected to an exhaust pump and gas volume detector. A flow rate of about 5 l/min was used and a total flow of at least 3.6 m³ were collected.

Samples were preliminary extracted with a sonication treatment of 30 min, using acetonitrile to desorb the pollutants from both cartridge and filter; then, after a vacuum concentration, the sample has been analyzed in a gas chromatography coupled with a mass-spectrometry (GC/MS). For PAHs, the limits of quantification for individual compounds were in the range of 0.01 ng/g wax.

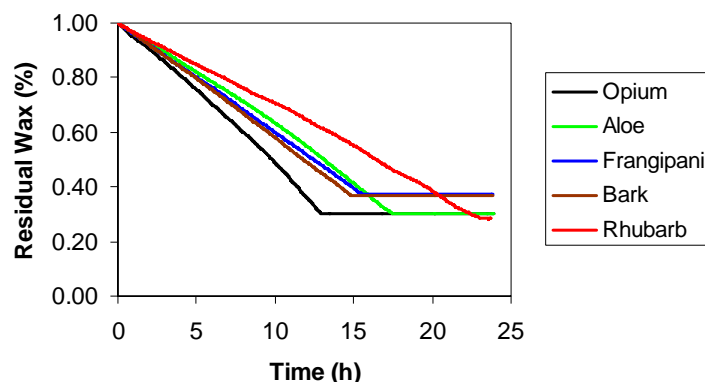


Fig.1 Wax consumption in real room conditions for the investigated scented candles.

3. RESULTS AND DISCUSSION

First, candles were burnt in a real room in order to evaluate the wax consumption at room conditions, as reported in figure 1; apart from the beginning of the test, when a calm flame gradually formed a cup rim surrounding the so-called burn bowl, the flame burned without visible release of smoke and the candles did not drip. In particular, scented candles were burnt from the lighting until the blow out, recording the weight change, but also in different burning cycles of about 4 hours, followed by a 1 hour stop between the cycles. No significant differences were found for the two procedures, however the second one led to a slightly lower wax consumption. As reported in figure 1, the investigated candles burn with different velocities and the wax consumption usually increases during the test; two candles showed a particular behaviour: rhubarb and opium scented candles evidenced the lower and the higher wax consumption rates, respectively. These differences were reduced using 4 candles, in particular when they were burnt within the test chamber, in a well-controlled environment; in this case the average wax consumption was in the range 4.3-4.9 g/h for all the investigated candles.

In order to evaluate the proper conditions that should be used for the experiments with the test chamber, several preliminary TGA/DTA analyses have been performed on both candles and raw materials (from 30 up to 900°C, heating rate of 10°C/min), using air or nitrogen as carrier gas. It was found that light compounds, say those contained in the fragrances, begin to evaporate at about 50°C, the paraffin-stearin blend begin to melt at about 55-58°C while the external microcrystalline wax starts to soften at 65°C. These issues suggested to carefully monitor the temperatures inside the enclosed combustion chamber used for the experiments in order to avoid an excessive wax consumption and dripping, that is burning mass running out of the burning bowl of the candle, but also to prevent the candles loss of shape and resistance. For this reason, four thermocouples allowed for monitoring the temperatures into the candle body, at the chamber wall, at the roof of the chamber and within the stack; figure 2 reports typical temperatures profiles measured during an experimental run performed burning four candles simultaneously.

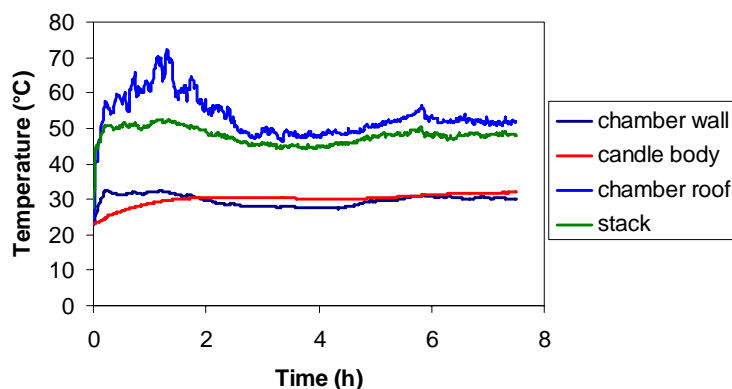


Fig. 2 Temperature profiles for a test performed with four scented candles.

Moreover TGA analyses carried out both with nitrogen or air as carrier gas evidenced that a marked weight loss of the wax blend starts at about 120-130°C, thus indicating an overlapping of different phenomena, such as evaporation/decomposition and combustion. The process was obviously faster with air and it was always characterized by two main steps; at about 520°C only the 0.3% of the initial sample was found as a residue.

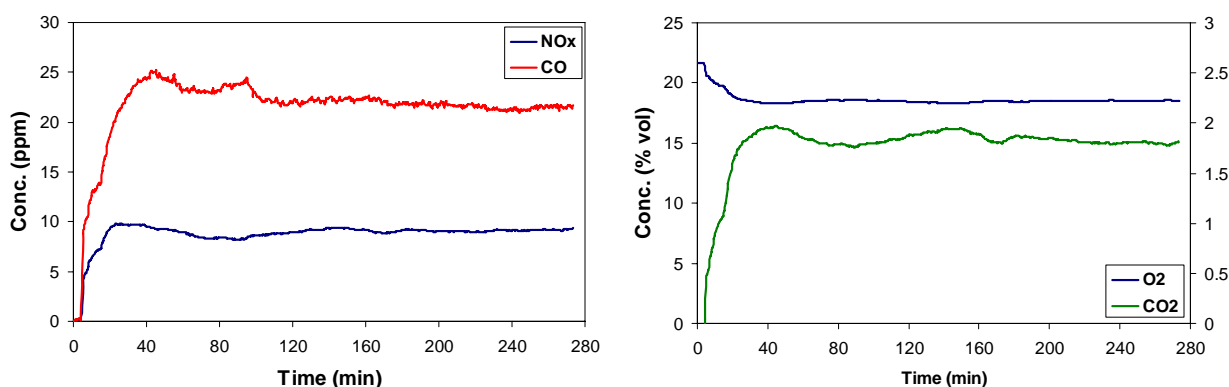


Fig. 3 Main pollutants concentrations detected in a test carried out burning four aloe scented candles.

As the temperature measurements helped to choose the maximum number of scented candles that can be burnt simultaneously, the detection of main combustion by-products allowed to determine the proper inlet air flow-rate that must be used to ensure a combustion environment similar to the real room conditions and also the duration of the initial transitory of the

experiments, in which progressively steady conditions in terms of pollutants production and wax consumption are reached within the test chamber (figure 3).

Measured emission were calculated as emission factors adjusted to the amount of substance released when 1 g of wax is being burnt. These values allow a direct comparison between different types of candles; as shown in tables 1 to 3, emission factors vary with the type of fragrance of the scented candle.

	Bark	Frangipani	Opium	Rhubarb	Aloe
Benzene	0.125	0.086	0.092	0.084	0.01
Toluene	0.113	0.025	0.093	0.23	0.05
Ethylbenzene	0.127	0.14	0.132	0.174	0.05
Xylenes	=	=	=	=	=

Tab. 1 Emission factors, in $\mu\text{g/g}$ of burnt wax, of BTEX for the investigated scented candles.

	Bark	Frangipani	Opium	Rhubarb	Aloe
Formaldehyde	2.49	2.91	2.28	2.54	2.32
Acetaldehyde	0.73	1.11	0.62	0.83	1.09
Acroleine	2.54	2.02	1.78	0.95	1.05
Butyraldehyde	0.44	0.3	0.14	=	0.38
Benzaldehyde	0.41	0.55	0.31	=	0.3
Tot. aldehydes	6.6	6.87	5.12	4.32	5.14

Tab. 2 Emission factors, in $\mu\text{g/g}$ of burnt wax, of aldehydes for the investigated scented candles.

	Bark	Frangipani	Opium	Rhubarb	Aloe
Fluoranthene	0.032	<0.01	0.062	0.074	0.015
Pyrene	0.032	<0.01	0.094	0.097	<0.01
Phenanthrene	0.068	0.02	0.135	0.059	0.03
Anthracene	0.016	0.01	0.007	0.022	<0.01
Benzo(a)anthracene	0.032	0.02	<0.01	<0.01	<0.01
Naphtalene	<0.01	0.01	0.031	0.037	<0.01
Fluorene	<0.01	<0.01	0.02	<0.01	0.04
Chrysene	<0.01	<0.01	<0.01	0.74	<0.01
Benzo(a)pyrene	<0.01	<0.01	<0.01	0.34	<0.01

Tab. 3 Emission factors, in ng/g of burnt wax, of PAHs for the investigated scented candles.

As it could be notice from the estimated emission factors the general pattern of the individual pollutants tested is also quite comparable for all materials, indicating no significant different pollutants formation conditions. Moreover, high emissions of e.g. benzene are not necessarily connected with high levels of PAHs, and even within the BTEX no clear relation exists. In particular, among the tested candles, aloe containing ones showed very low levels of BTEX and PAHs while they evidenced emission factors for the aldehydes similar to the other fragrances. This could be ascribed, probably to the lower amount of colour that is contained within this kind of scented candles; in fact, the aloe candles are characterized by a green pale colour, while the other scented candles are all characterized by a larger amount of a dark dye (blue for frangipani, black for opium, red for rhubarb and brown for bark). Therefore, the colour amount seems to influence mainly the formation of BTEX but not the production of aldehydes. On the other hand, aldehydes emissions, as found also by Lau et al. [14], probably depends on the amount of paraffin within the wax mixture; this explains because they are

similar in all the candles. Concerning PAHs, it is possible to notice that only frangipani and aloe scented candles show emission factors close to the lower detection limit of the analytical procedures.

Emissions of several pollutants were then compared to limit values for working places; on basis of the determined emission rates of the candles, a simple indoor air scenario was used to determine resulting indoor air levels from a simultaneously burning of four scented candles for 4h in a 50 m³ room. The resulting indoor air levels were evaluated by a comparison to available air quality standards - toxicologically based as well as descriptive - both including work place, indoor air, and ambient air references. The vast majority of the resulting air levels from the scented candles utilized the compared air quality standard to less than 0.5 % for work place related standards, less than 10% for indoor air and ambient air related standards. Therefore, no applied air quality standard was exceeded or even touched by the resulting air levels for non sooting candles. All levels are lower than the most sensitive standard used. From this point of view, no indication could be identified for a possible significantly elevated indoor air level due to a 4 h burning of four scented candles tested in this study and considering the applied worst case model conditions.

4. References

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