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Dynamic of Particulate Matter for Quotidian Aerosol Sources in Indoor Air

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Abstract: A correlation between the mass concentration of particulate matter (PM) and the occurrence of health-related problems or diseases has been confirmed by several studies. However, little is known about indoor PM concentrations, their associated risks or their impact on health. In this work, the PM_1, $PM_{2.5}$ and PM_{10} produced by different indoor aerosol sources (candles, cooking, electronic cigarettes, tobacco cigarettes, mosquito coils and incense) are studied. The purpose is to quantify the emission characteristics of different indoor particle sources. The mass concentration, the numerical concentration, and the size distribution of PM from various sources were determined in an examination room 65 m^3 in volume. Sub-micrometer particles and approximations of PM_1, $PM_{2.5}$ and PM_{10} concentrations were measured simultaneously using a diffusion aerosol spectrometer (DAS). The ultrafine particle concentration for the studied indoor aerosol sources was approximately 7×10^4 particles/cm^3 (incense, mosquito coils and electronic cigarettes), 1.2×10^5 particles/cm^3 for candles and cooking and 2.7×10^5 particles/cm^3 for tobacco cigarettes. The results indicate that electronic cigarettes can raise indoor $PM_{2.5}$ levels more than 100 times. PM_1 concentrations can be nearly 55 and 30 times higher than the background level during electronic cigarette usage and tobacco cigarette burning, respectively. It is necessary to study the evaluation of indoor PM, assess the toxic potential of internal molecules and develop and test strategies to ensure the improvement of indoor air quality.

Keywords: indoor sources; indoor air quality; particle number concentration; particle size; particulate matter

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1. Introduction

Over the past two decades, several epidemiological and clinical studies have been conducted that indicate a relationship between exposure to particulate matter (PM) and numerous health effects [1–17]. PM exposure mostly occurs indoors, as people spend a major part of their lives indoors at home or the workplace (approximately 90%) [16,18].

Increases in PM exposure predominantly led to increased hospitalization, especially in the elderly and individuals with heart and lung problems. Not only long-term, but also short-term exposure to fine PM air pollution increases the risk of myocardial infarctions [19–25], lung disease [26–28] and strokes [29–33].

With a confirmed negative impact on human health, scientists divided particles into fine (diameter >100 nm) and ultrafine particles (diameter <100 nm). Ultrafine particles can penetrate deep into the respiratory system and even cross biological barriers, leading to harmful health effects due to their small size [34–48]. Therefore, a precise characterization of multiple parameters for fine and ultrafine particles is required [42,49]. Indeed, it has been reported that number concentration may be a better indicator of exposure than mass concentration [46,50]. Moreover, it has been reported that the biological effects of

ultrafine particles are more clearly related to the surface area of the particles than to their mass [36,37]. Therefore, potential health effect assessments urgently need to evaluate the indoor exposure associated with a specific source emission using several complementary measures (number, mass, surface distribution, volume, and chemical composition). The dynamics of PM in the lungs and others tissues of the respiratory system are shown in Figure 1 [15].

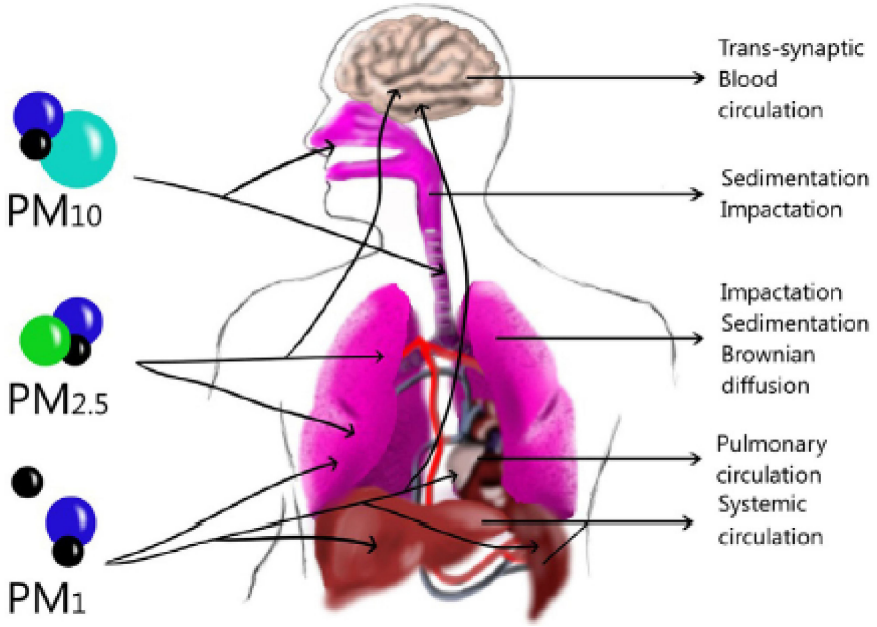

Figure 1. The size and dynamics of particulate matter (PM)in the lungs and others tissues [7].

In recent years, concerns over indoor air quality have significantly increased due to the recognition of different pollutants produced by various external and internal sources. These sources depend on the procedures operating within the environment [51]. The US Environmental Protection Agency (USEPA) put indoor air pollution onto its list of top five environmental hazards [52]. Singh et al. (2014) confirm that long-term exposure to resident pollutants in indoor air is the cause of diseases related to respiratory and cardiovascular issues, and even potentially carcinogenesis [53].

The characterization of particle size distribution is not only useful in determining the lung deposition of molecules on a regional level, but also for examining the distribution of the sources of internal aerosols via receptor modeling methods [28,54]. In an indoor environment, both indoor and outdoor sources contribute to PM levels. However, many PM measurements have been made outdoors: few data are available on indoor air pollution [55].

PM indoors is affected by ambient concentrations, air exchange rates, penetration factors and sedimentation and re-suspension mechanisms. There is a growing body of evidence linking ambient particulate air pollution from combustion sources to a harmful health outcome [1,56]. As people spend the majority of their time indoors, it is important to measure exposure to respirable particulates from indoor combustion sources, an important step towards assessing their role in health outcomes.

The main indoor sources are combustion processes producing smaller particles, the vast majority of which fall into the sub-micrometer zone and consist of organic and inorganic materials [57]. In this delicate and complex environment, activities such as cooking, cleaning, walking and especially smoking are the main sources of particles indoors. Numerous studies have identified a large number of indoor particle sources, and the emissions from these sources have been investigated. The most significant sources include cooking, smoking, heating and burning incense [28,54]. The smoking of cigarettes [58] causes PM

to form indoors [8]. Kankaria et al. (2014) examined ultrafine particle concentrations in seven residences in Northern California: their results indicate that cooking was the most significant internal activity contributing to the ultrafine particle level within the buildings. They also conclude that smoke from combustion causes the release of particulate matter, which poses a broader health risk when inhaled [13].

In general, exposure to aerosols through inhalation is of the utmost significance, as it is a major potential source of risk to human health. This depends on airborne concentrations, duration of exposure and chemical composition. Therefore, it is quite important to measure concentrations of PM in the size fractions known to affect human health [59]. The quantitative evaluation of the properties of indoor emissions in real situations is a complex task, and therefore, only qualitative information is available on the contribution of many sources of indoor particles to health problems or about the ranges of the PM levels resulting from such sources indoors.

Particulate Matter ($PM_{2.5/10}$) Health Effects

The main health impacts include premature deaths, respiratory and cardiovascular diseases and changes in lung function [7,13,60]. Inhaled particles that can reach the lower airways are classified into three categories by size: PM_{10} (diameter $\leq$ 10 μm), $PM_{2.5}$ (diameter $\leq$ 2.5 μm) and PM_1 (diameter $\leq$ 1 μm). Air particulates are generally classified by how deep they penetrate into the human respiratory system. Particles smaller than 2.5 microns ($PM_{2.5}$) are known as fine particles, and they pose the greatest health risks because they can enter the deepest parts of the human respiratory system and lead to harmful health effects. Particles larger than 2.5 microns but smaller than 10 microns (PM_{10}) in size are known as coarse particles; these are too large to enter the human respiratory system, but prolonged exposure can lead to respiratory disease. Short-term exposure has major implications for respiratory health, but, in terms of mortality, $PM_{2.5}$ is a greater risk than PM_{10}. Studies have shown that with a 10 $\mu g/m^3$ increase in PM_{10} concentration indoors, cardiovascular deaths increased by 0.36% and respiratory deaths by 0.42%. Likewise, with an increase of 10 $\mu g/m^3$ in the indoor $PM_{2.5}$ concentration, cardiovascular deaths increased by 0.63% and respiratory mortality by 0.75% [60–62].

In most cases, the concentration and size play a role in determining the response after inhalation, in addition to the particle number concentration itself. However, the interpretation of ultrafine particle data, in terms of the surface area of the particles, leads to a dose response independent of the diameter of the particles in many cases [37,63]. A similar trend has been observed in the diameters of larger particles [64], indicating that the low solubility particles that characterize exposure, in terms of surface area, will result in more appropriate exposure limits and evaluation methods [65,66].

Therefore, the aim in the present study is to simultaneously evaluate PM_1, $PM_{2.5}$ and PM_{10} suspended particles from different indoor aerosol sources (cooking, burning candles, regular and electronic cigarette smoking, incense and coils for protection against insects) in terms of their numerical concentration, mass concentration and surface area. This will deepen our knowledge of indoor air quality.

2. Materials and Methods

The sources were tested in a room with a volume of 65 m^3. The room temperature during the experiments ranged from 22 to 25 °C with a relative humidity of 40–50%. A diffusion aerosol spectrometer (DAS) was used to monitor mass concentrations continuously before the aerosol source was turned on (BG), during the time it was on and at intervals after it was switched off. The technical design and the description of the DAS (Model 2702 M) are presented in previous works [43–48].

Using the DAS, aerosol particle parameters such as numerical concentration, various PM concentrations (PM_1, $PM_{2.5}$ and PM_{10}), average particle surface area and metrological parameters (temperature, humidity, and pressure) were recorded and automatically saved every ~2 min. An electronic unit controlled the spectrometer operations, processing,

calculation, display of the measured results and storing data. The data was transferred from the DAS to another computer via a USB interface. The output files were EXCEL files containing the measured data. The specific surface area of aerosol particles was calculated from the numerical distribution of particles by diameter, under the assumption that the aerosol particle is spherical.

Aerosol Sources

Some of the most widely used indoor aerosol sources are presented in Table 1. Six sources were used (candles, cooking, electronic cigarettes, tobacco cigarettes, mosquito coils and incense). Throughout most of the experiments, the sources were active until the concentration seemed to stabilize. The source was then removed or deactivated, and the subsequent decrease in concentration was studied. Four candles were lit indoors until the concentration of aerosol particles was stable. After burning the candles for 40 min, they were extinguished with a damp cloth. After stopping burning, the candles were not removed from the room. The different aerosol sources were placed in the center of the room at 3 m from the DAS.

Table 1. Description of the six different types for indoor aerosol sources.

Indoor Sources	Burning Time	Amount of Source	Source Location
Candles	40 min	Four scented candles	In the centre of the room
Cooking	30 min	15 min for heating and 15 min for cooking	In the centre of the room
Electronic cigarette (Pone type)	10 min	One cigarette	In the centre of the room
Tobacco cigarette	10 min	One cigarette	In the centre of the room
Mosquito coil	60 min	One mosquito coil but not finished	In the centre of the room
Incense	20 min	Three popular and usable brands of incense	In the centre of the room

Figure 2 presents the level of particle concentration created from six different aerosol sources (cooking fish, electronic cigarettes, incense sticks, tobacco cigarettes, mosquito coils and candles) with different burning/functioning times. The sources were active until the concentration appeared to have stabilized. Cooking and burning candles produced the maximum concentration of ultrafine particles (<200 nm), approximately 1.2×10^5 particles/cm^3. The mosquito coil produced the minimum concentration, ~7×10^4 particles/cm^3. For 300 nm particles, the number concentration ranges from 50 to 800 particles/cm^3. The number concentrations for particles with diameters of 500 and 1000 nm ranged from 20 to 1400 particles/cm^3 and 5–700 particles/cm^3, respectively, for different sources. It was observed that the electronic cigarette produced the highest concentration of fine particles; the burning candles produced the lowest. For the incense sticks, tobacco cigarettes, and electronic cigarette, the fine and ultrafine particle fractions reached their maximum the moment they were extinguished. As for mosquito coils, this only happens with the fine particles. The situation for the candles was different, as the fine particles began to increase when they stopped burning. For cooking, there is no noticeable change in terms of the fine particles: only the ultrafine particles increased and slowly deceased after the stove was turned off.

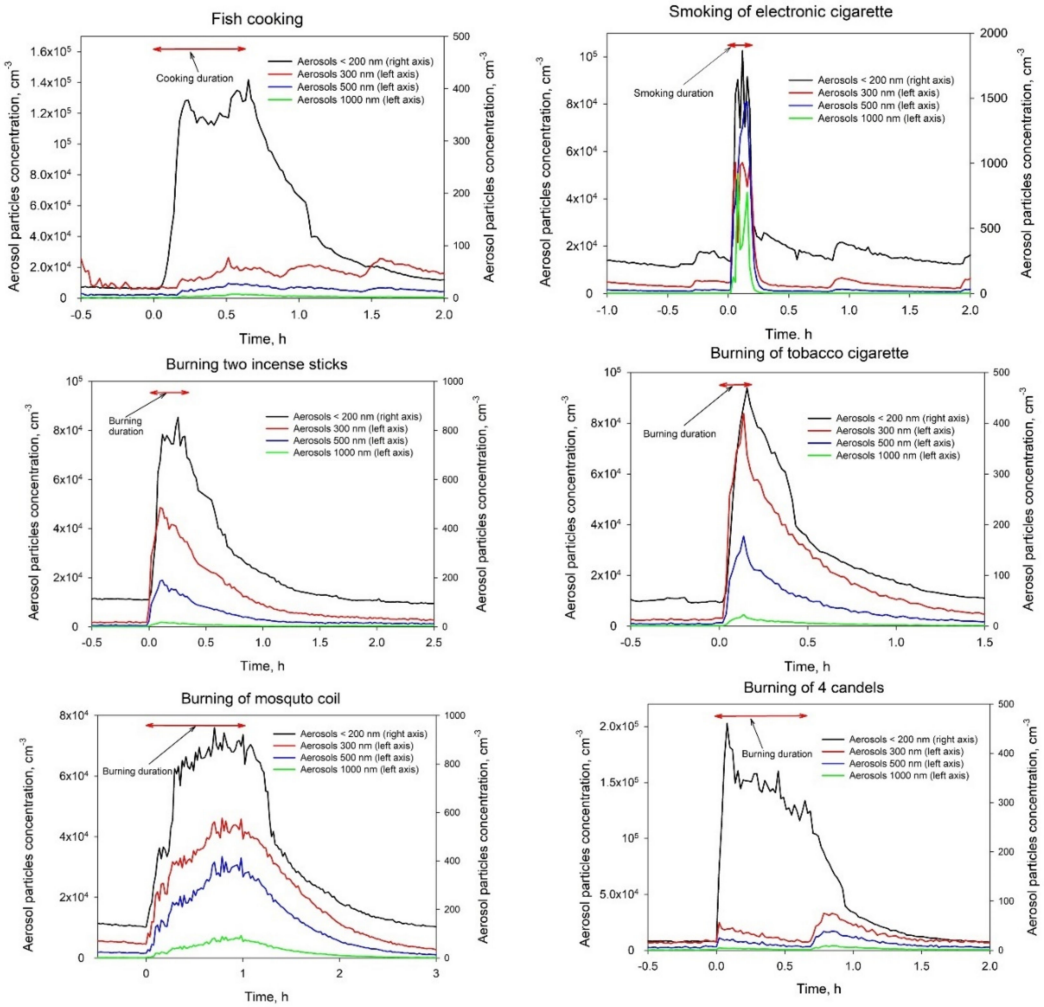

Figure 2. UFP (<200 nm) and fine particle concentration generated by six different aerosol sources (burning duration means when the aerosol source was on), as measured by the DAS. Left Y-axis for UFP (<200 nm) and right Y-axis for other sizes.

3. Results

The ultrafine particle (UFP) numerical concentrations of the six different indoor aerosol sources (candles, cooking, electronic cigarettes, tobacco cigarettes, mosquito coils and incense) are presented in Figure 3.

From Figure 3, it appears that the particulate matter concentration from incense, candles and tobacco cigarettes achieved the highest level at the end of the burning period, unlike the other sources (cooking, electronic cigarettes, and mosquito coils), which achieved this value five minutes after stopping the sources. The highest initial particle concentration was emitted by the combustion of the tobacco cigarette, but it also had a faster decline rate directly after combustion ceased. In general, the particle concentration decrease is due to various particle loss processes, including air exchange, particle filtering, deposition and coagulation [67–69]. Earlier published studies indicate that major indoor UFP sources include combustion due to cigarette smoking, candle burning and incense [7,8,70,71].

The particles from cooking achieved their maximum numerical concentration 5 min after the process was ended. The rate of particle removal was low: after 30 min, only half of the particles were removed. For other sources, the particle number concentration was nearly back to background levels after 30 min. This may be due to the nature of cooking particles, which are a combination of different materials evaporated with water vapor and have a low interaction cross section with surrounding particles in the air indoors [28,72].

The other sources emit particles, which quickly react with suspended aerosol particles when indoors.

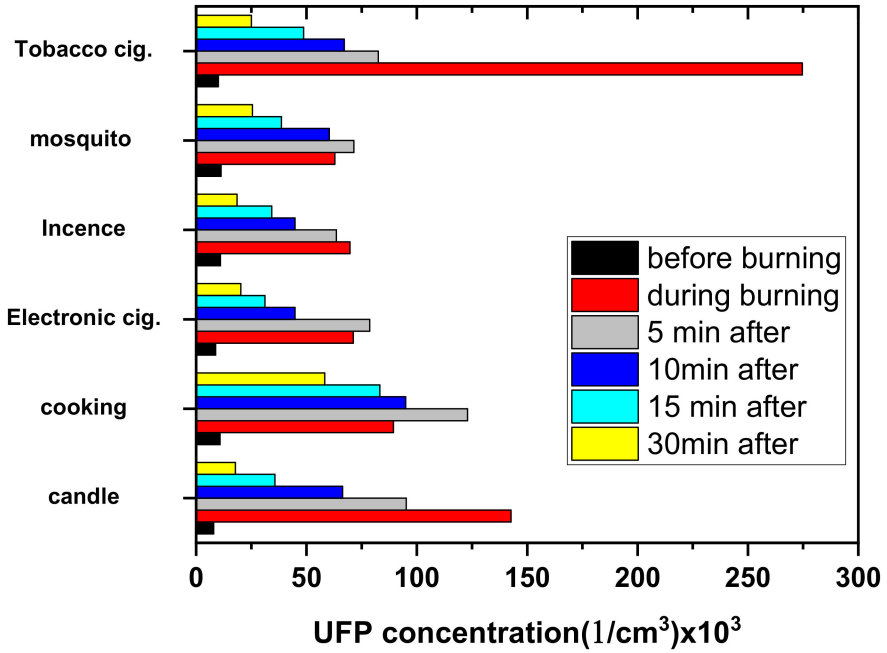

Figure 3. Ultrafine particle number concentration (<200 nm) for six indoor aerosol sources (candles, cooking, electronic cigarettes, tobacco cigarettes, mosquito coils and incense) at different times (before being turned on, during operation and after being stopped).

As shown in Figures 4–6, the highest mass concentrations for $PM_{1.0}$, $PM_{2.5}$ and PM_{10} were recorded for the electronic cigarettes (or, more precisely, for 5 min after switching off the source). These mass concentrations dispersed very quickly after switching off the source, within only 5 min (unlike cooking, which produced PM concentrations that remained for longer). In the present study, the electronic cigarettes raised the indoor $PM_{2.5}$ level more than 100 times, while the PM_1 and PM_{10} concentrations were nearly 55 times and 30 times higher than the background levels, respectively.

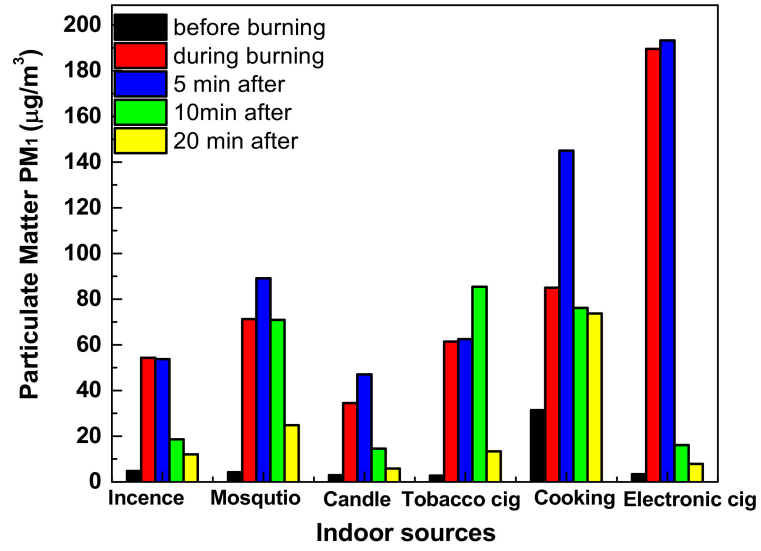

Figure 4. Particulate matter mass concentration (PM_1) for six indoor aerosol sources (incense, mosquito coils, candles, tobacco cigarettes, cooking and electronic cigarettes) at different times (before being turned on, during operation and after being stopped).

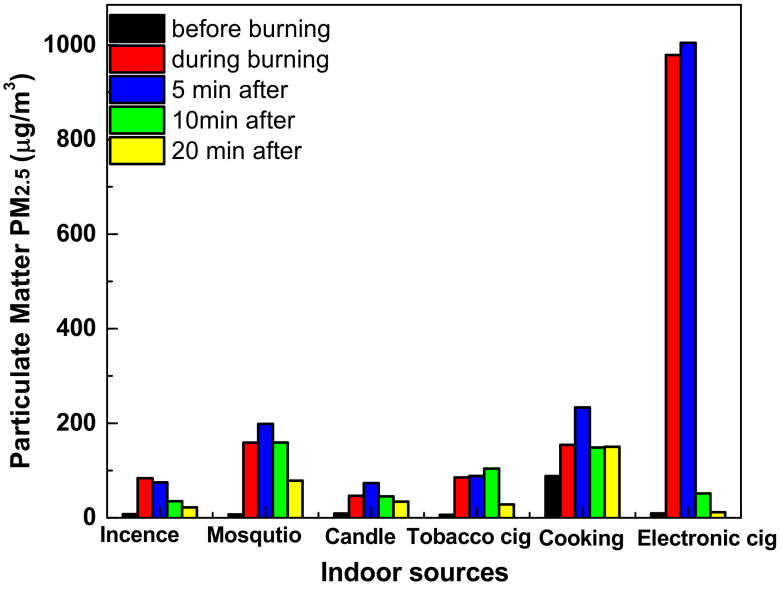

Figure 5. Particulate matter mass concentration ($PM_{2.5}$) for six indoor aerosol sources (incense, mosquito coils, candles, tobacco cigarettes, cooking and electronic cigarettes) at different times (before being turned on, during operation and after being stopped).

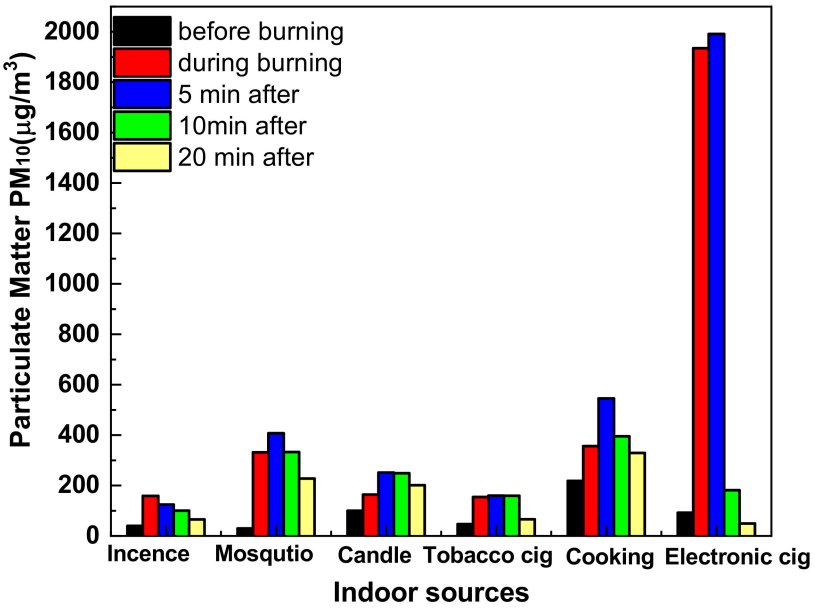

Figure 6. Particulate matter mass concentration (PM_{10}) for six indoor aerosol sources (incense, mosquito coils, candles, tobacco cigarettes, cooking and electronic cigarettes) at different times (before being turned on, during operation and after being stopped).

Comparing the present results regarding candles with other results in the literature, they agree. As shown in Figures 3 and 4, candles are identified as a source of ultrafine particles. This fact is confirmed by [7,55,73]. Long (2000) and Ogulei (2006) present candles as major emitters of particles [74,75]. LaRosa et al. (2002), Sørensen et al. (2005) and Afshari et al. (2005) consider them to sbe major contributors to $PM_{2.5}$ concentrations [76–78]. In the present study, the total numerical concentration for candles was 140,000 particles/cm^{-3} in a room 65 m^3 in volume; however, the maximum numerical concentration emitted from two scented candles in a 32 m^3 room was 69,000 particles/cm^{-3} [55].

4. Discussion

Two of the most significant indoor combustion activities and $PM_{2.5}$ sources are environmental tobacco smoke (ETS) and food cooking [34,79,80]. In the present study, the $PM_{2.5}$ indoor particle mass concentration was found to increase the background level 3 to 90 times during smoking and frying [81].

Incense burning, which is an incomplete combustion process, is known to emit fine and ultrafine particles in large quantities [42,82]. Incense burning leads to the production of particulate matter, out of which $PM_{2.5}$ is the most released pollutant: this is more hazardous and causes adverse health effects [62]. The present average total numerical concentration for incense was 140,000 particles/cm^{-3}; however, the maximum numerical concentration emitted from the three different types of incenses can range from 126 to 173 $\times$ 10^3 particles/cm^{-3} [83].

Since the electronic cigarettes emitted the highest numerical concentration and particulate matter concentration, it also achieved the highest result in terms of surface area, especially in the first five minutes after the device was turned off (see Figure 7). The difference in specific surface area is explained by the higher concentration of medium and small-sized aerosol particles in the smoke of an electronic cigarette (see Table 2).

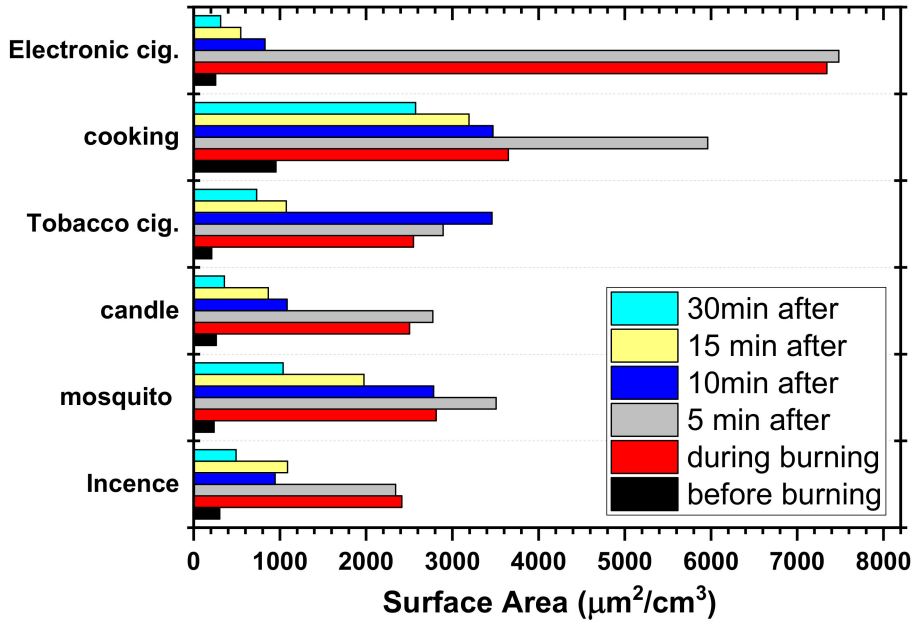

Figure 7. Surface area for six indoor aerosol sources (candles, cooking, electronic cigarettes, tobacco cigarettes, mosquito coils and incense) at different times (before being turned on, during operation and after being stopped).

Table 2. Number size distribution parameters of ultrafine particles for different aerosol sources, Adapted from [84].

Aerosol Sources	Background		During Source On		10 min after Stop		60 min after Stop	
	NMD (nm)	GSD	NMD (nm)	GSD	NMD (nm)	GSD	NMD (nm)	GSD
T-cig.	54.6	1.73	92.1	1.85	115.8	1.43	57.1	1.86
E-cig.	54.5	1.84	71.3	2.22	39.4	2.06	50.1	1.83

Regarding the toxic effects of UFPs, the relatively high concentration of particles and a large surface area allow for a greater proportion of absorbed or condensed particles to enter into tissues and the bloodstream [71,85–87].

For indoor aerosol sources utilizing high temperatures, the aerosol mass concentration of particles varies during the operation of the source and after nearly 10 min of its termina-

tion. The maximum mass concentration of aerosol particles formed during the operation of household aerosol sources corresponds to a particle diameter of 2–5 microns [87].

5. Conclusions

The study suggests that the combustion of six different indoor sources (candles, cooking, electronic cigarettes, tobacco cigarettes, mosquito coils and incense) in an indoor environment emits quite high levels of respirable PM, which may accumulate and lead to prolonged exposure. Households should have better ventilation in order to avoid a buildup of PM. Residing in a place with such a high particulate concentration may lead to serious respiratory health concerns.

1. The particulate matter concentration from incense, candles and tobacco cigarettes achieved the highest level at the end of the combustion period. The highest initial concentration of particles was emitted by the tobacco cigarettes.
2. The highest concentrations of PM_1, $PM_{2.5}$ and PM_{10} were recorded for the electronic cigarettes.
3. The study found that electronic cigarettes can raise indoor $PM_{2.5}$ levels more than 100 times, while PM_1 concentrations can be 55 and 30 times higher than the background level during the usage of electronic cigarettes and tobacco cigarettes, respectively.

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