



Determination of Size-Dependent Source Emission Rate of Cooking-Generated Aerosol Particles at the Oil-Heating Stage in an Experimental Kitchen

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ABSTRACT

Cooking-generated particles represent one major indoor particulate pollutant that significantly affect indoor air quality as well as pose a risk to human health. Prediction of the spatial-temporal distribution of fume particles and individual inhalation exposure is highly dependent upon the source characteristics. This study aims to determine the emission rate of cooking-generated particles in the range from 0.1 to 10 μm , because this size is of most concern when considering indoor particle dynamics. The mass concentration and volume-based size distribution of particles in the range from 0.1 to 10 μm are measured under controlled conditions in a laboratory kitchen. Based on a mass balance model, the total emission rates of $\text{PM}_{2.5}$ and PM_{10} are determined using the concentration decay rate derived from the measured concentration plus its changing curve with time. The size-dependent emission rate is further obtained by multiplying the total emission rate by the particle volume distribution. It is found that source strengths are highly sensitive to the oil type, whilst both the volume-distribution patterns and decay rate values exhibit less difference among the six types of vegetable oil examined in this work. The relative variance of source rate determined at time spans from 15 min to 2 h is less than 5.5%, and thus a short measuring period of 15 min is sufficient to derive a reliable emission rate. The results also show that the volume frequency of particles in the size range from 1.0 to 4.0 μm accounts for nearly 100% of the mass of PM_{10} in the oil-heating experiments. The size range and the associated size-based emission rates derived in this study can be applied as the source characteristics for further studies of indoor particle dynamics.

Keywords: Fume particle; Volume distribution; Decay rate; Emission rate; Indoor air quality.

INTRODUCTION

Cooking represents one of the most significant particle generating activities indoors (Kamens *et al.*, 1991; Ozkaynak *et al.*, 1996; Diapouli *et al.*, 2011; Cao *et al.*, 2012; Chowdhury *et al.*, 2012; Pervez *et al.*, 2012). Amount of epidemiological results have concluded the significant linkage between health effects and exposure to particulate matters from cooking (Loomis, 2000; Pope, 2000; Metayer *et al.*, 2002; Sze-To, *et al.*, 2012). However, epidemiology is the ‘macroscopic’ and statistical study of population-based distributions and patterns of health-events, health-characteristics and their causes. In regard to indoor air quality and individual exposure due to cooking-generated particles, it is more concerned about the spatial-temporal distribution of ‘microscopic’ particle indoors. Particle source

characteristics of cooking activities become one major factor for the well-mixing model (Chen *et al.*, 2000) and computational fluid dynamics (Lai and Chen, 2007) to achieve the spatial-temporal information.

Some studies (Lai and Chen, 2007; Lai and Ho, 2008) assumed the non-dimensional emission rate or single particle diameter for the simulation of spatial distribution of cooking-emitted particles. Such assumed source was insufficient to obtain the actual exposure concentration. A number of studies (Siegmann and Sattler, 1996; He *et al.*, 2004; Yeung and To, 2008; Wallace *et al.*, 2008; Buonanno *et al.*, 2009; Zhang *et al.*, 2010; Diapouli *et al.*, 2011) have attempted to measure the number concentration of indoor particles during cooking activities. Their measuring points were located a certain distance away from the source to represent the well-mixed concentration in the space. For example, all measurements were made at a distance of 2 m from the stove by He *et al.* (2004) and Buonanno *et al.* (2009), respectively, made at the height simulating the human breathing zone by See and Balasubramanian (2006), at 1 m away from the stove by Zhang *et al.* (2010), and at the exhaust outlet of ventilation system by Tan *et al.* (2012). In recent years, a few

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studies (Wallace *et al.*, 2004; He *et al.*, 2005; Wallace, 2006; Olson and Burke, 2006; Buonanno *et al.*, 2009; Buonanno *et al.*, 2010) realized to relate indoor particle concentration to the source emission factors of cooking activities. Typically, particle size and number concentration were measured in the space a certain distance away from the source using the scanning mobility particle sizer (SMPS, number-based size spectrum 0.015–0.685 μm) and condensation particle counter (CPC, total number concentration from 0.007 to 0.808 μm). Additionally, the aerodynamic particle sizer (APS) was also used to measure size distribution and number concentration of larger sized particles. They determined the source strengths by the concentration decay rate from the natural logarithm of measured concentration and its linear regression using the mass balance model. These studies mostly focused on ultrafine particle number concentration, and very few reported the $\text{PM}_{2.5}$ and PM_{10} mass emission factors in detail (Haryono *et al.*, 2011). It has been found that Chinese-style cooking contributes approximately 30% to indoor particle concentration in the range of 0.5 to 5 μm (Liao *et al.*, 2006). Grilling Chinese-style food is capable of elevating submicron particle and $\text{PM}_{2.5}$ concentrations up to 5 and 90 times higher than the normal, respectively (He *et al.*, 2004). Due to that accumulation or coarse particles behave very different from the ultrafine particles, and exposure to such size range also significantly impact on health (Loomis, 2000; Pope, 2000), therefore their mass concentration was regarded as the control index of indoor air quality, implying that it is of practical significance to carry out the detailed studies on the source characteristics of $\text{PM}_{2.5}$ and PM_{10} generated from cooking activities.

The aim of this work is to quantify source emission rates of particles ranging from 0.1 to 10 μm emitted during a specified oil-heating period for typical Chinese-style

cooking process in a laboratory kitchen. Volume-based size distribution of fume particles emitted from various types of vegetable oil is obtained using the real-time Malvern size analyzer at nearly the top surface of stove. Particle mass concentration of $\text{PM}_{2.5}$ and PM_{10} are measured using the TSI Model 8533 DustTrak aerosol monitor. Source strengths are determined via a mass balance model together with the measured data of concentration. Results of this work are statistically analyzed and expected to provide reasonable source characteristics for the assessment of indoor air quality and individual exposure due to cooking-generated particles.

METHODS

Experimental Methods

The study is carried out in a laboratory kitchen established in a large outer experimental chamber. The sizes of the kitchen and the outer space are 3.5 m (L) \times 1.8 m (W) \times 2.4 m (H) and 20 m \times 15 m \times 3.5 m, respectively. An electric griddle with 7 thermal control levels is used to heat the vegetable oil in a round-bottomed wok. An exhaust hood and the associated exhaust ductwork are installed. Fig. 1 presents the schematic of the kitchen room and the layout of wok, exhaust hood, and the measuring instrument. Six types of vegetable oil are used, which are rapeseed oil, soybean oil, peanut oil, sunflower oil, olive oil and blend oil. In order to figure out emission level of the oil, an oil-preheating stage of 2 min is considered in the present work, which is typical for the Chinese-style cooking such as frying. Fig. 2 shows the monitored temperature during the heating process for 6 types of vegetable oil which is heated from the environmental temperature level. The averaged highest temperature is 216°C, which is exactly near the smoke point of vegetable oil. Cooking fume is found denser

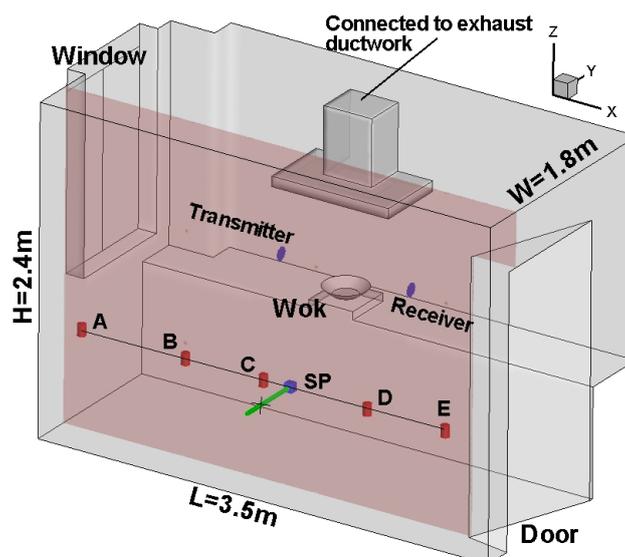


Fig. 1. Schematic of the kitchen room and the layout of measuring. Laser transmitter and receiver are at two opposite side of the wok when volume distribution is measured using the real-time Malvern Spraytec size analyzer. Points A-E denote the locations of wok center in the room when mass concentration is measured using the TSI Model 8533 DustTrak aerosol monitor at the specified sampling point SP. The distance between point A and B, Dis (A, B), is 0.8 m, Dis (B, C) = 0.6 m, Dis (C, SP) = 0.2 m, Dis (SP, D) = 0.6 m, Dis (D, E) = 0.6 m.

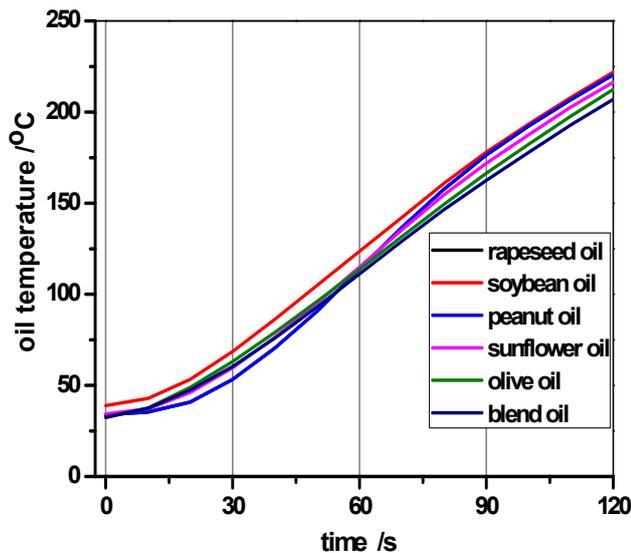


Fig. 2. Monitored oil temperature during the experimental cooking episode of 2-min heating. Oil temperature is controlled in the range of the smoke and flash point of vegetable oil.

with the increase of oil temperature in the experiment. However, oil temperature higher than 200°C is regarded as the necessary condition for Chinese-style medium- or high-temperature frying.

Particle mass concentration is measured using the TSI Model 8533 DustTrak aerosol monitor. Because the DustTrak operates based on a light scattering technique, the mass concentration of the aerosol is actually a reasonable approximation of the volume concentration. The measurements of mass concentration are conducted under nearly no-ventilation condition which means the door and window of the kitchen are closed and all the cracks are well sealed during the measuring. The procedure is as follows: 45 min of ventilation through exhaust hood before each measurement, followed by 120 min of mass concentration under closed condition during which the cooking process (2 min of oil heating) begins at 1.5 min and ceases at 3.5 min.

Volume distribution of the fume aerosols arising from the heated oil in the wok is measured using a real-time Malvern Spraytec size analyzer. This instrument is a laser-diffraction sizer system for the aerosol particle characterization based on the Fraunhofer approximation and Mie theory (Vargas-Ubera *et al.*, 2001). Measuring laser diffraction constructs the direct result as a volume distribution. The measurements are conducted under the dynamic emission process of the oil fume at nearly the top level of wok. Each measurement, using one type of oil, lasts 2 min. Sampling rate is 1 s. Before heating the oil, outdoor air is flushed through the window and door of the kitchen lasting for 20 min. During each measurement, the door and window are open to ensure the continuous arising of fume aerosols. Ventilation rate of 518.4 m³/h is maintained using the exhaust equipment.

Estimation of Particle Decay Rates and Emission Rates

To calculate the decay rates and emission rates of the

fume particles from cooking in an enclosure, a mass balance differential equation is employed as (Chen *et al.*, 2000):

$$\frac{dC_{in}}{dt} = P\alpha C_{out} - (\alpha + k)C_{in} + \frac{S}{V} \quad (1)$$

where, C_{in} and C_{out} are the indoor and outdoor particle concentration, respectively; P is the penetration efficiency; α is the air exchange rate; k is the deposition rate (natural decay rate); S is the emission rate; t is the time and V is the efficient volume of the space. The ventilation system is switched off and all cracks are sealed during the experiments, therefore, the first term on the right hand side of the equation does not exist and only the natural decay rate is considered.

The procedure of determining the decay rate and emission rate is as follows: (1) Present the measured data of concentration; (2) Find the peak concentration and the time t_p when it occurs; (3) Let $S=0$ and integrate Eq. (1) after t_p ; (4) Let k constant, integrate Eq. (1) before t_p .

$$C_{in}(t) = \frac{S}{\kappa V} (1 - e^{-\kappa t}) \quad t < t_p \quad (2)$$

$$C_{in}(t) = C_p e^{-\kappa(t-t_p)} \quad t \geq t_p \quad (3)$$

From the above Eq. (3), transform the measured data of C_{in} to natural logarithms and carry out a linear regression analysis over time, then the negative slope of the regressed line is the decay rate k . Substitute the calculated value of decay rate into Eq. (2), emission rate S is hereafter derived.

RESULTS AND DISCUSSION

Decay and Emission Rate of Fume Particles

The increase of mass concentration followed by a smooth decay within 900 s is presented in Fig. 3, where particle concentration is displayed as a function of time. Elevated mass concentration is found 2–3 orders of magnitude greater compared to the background concentration. As shown in Fig. 3, the peak concentration with the corresponding time t_p when it occurs is very significant. Five repeated measurements are carried out for each type of vegetable oil and their mean values are statistically analyzed. Regarding each measurement, wok is set at five different locations with different straight-line distances from the specified sampling point (See the locations A–E of the wok and the distance from the sampling point SP defined in Fig. 1). As the value of standard deviation shown in Fig. 3, the peak concentrations exhibit small differences among the repeated measurements. Therefore, well-mixing patterns of mass concentration are confirmed to some degree in this study. These criteria should be efficient to control and reduce the errors associated with applying the above Eqs. (2) and (3). Compared to the measurements conducted in previous published studies in the condition of ventilation, particle concentration measured in the closed space could be more uniform and less sensitive to the location of sampling point.

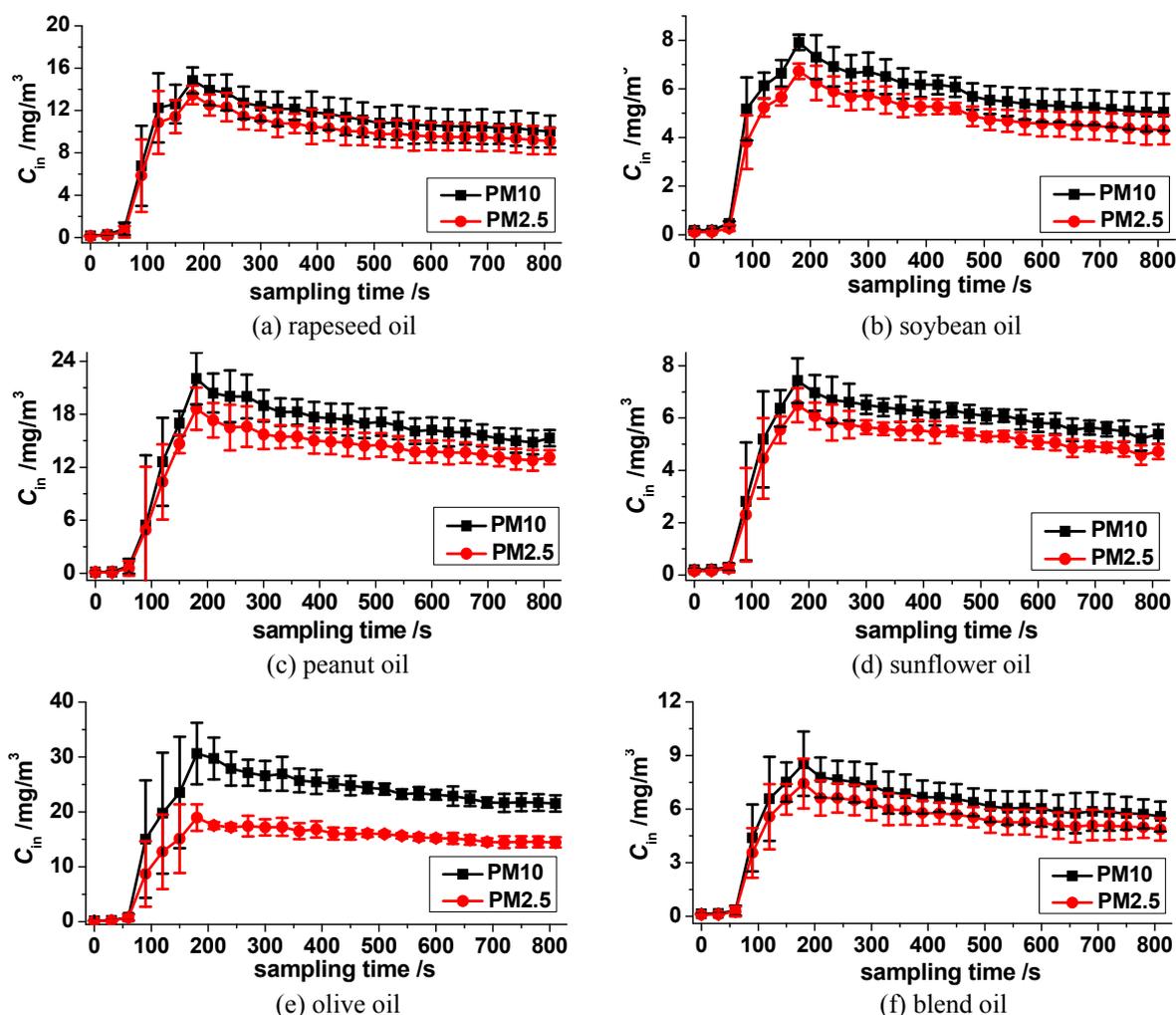


Fig. 3. Measured time-dependent mass concentration of cooking-generated $PM_{2.5}$ and PM_{10} for 6 types of vegetable oil applied. Error bar denotes standard deviation.

Fig. 4 shows the linear regression for the calculating of decay rate, where the negative slope of regression equation of $\ln(C_{in})$ is the decay rate k according to Eq. (3). The regression has R^2 values greater than 0.6 and are statistically significant. Decay rate of PM_{10} appears to be a little larger than that of $PM_{2.5}$ for all types of oil, resulting from the gravitation effect as well as the dominant mass fraction of particles in the range of 0.1–2.5 μm among PM_{10} .

Fig. 5 presents the calculated source emission rate of PM_{10} according to Eq. (2) and the corresponding values of decay rate. The derived source strengths for 6 types of oil under the same condition are found significantly different. Olive oil at the heating process is found to produce the highest rate compared to the lowest originated from sunflower oil. The highest one is 4.2 times of the lowest. In the experiments, we found out that much more amount of cooking fume emitted by heating olive and peanut oil than others, indicating that it should be careful to use the olive and peanut oil for the Chinese-style cooking. Buonanno *et al.* (2009) also found that sunflower oil generated the lowest mass emission rates while olive oil emitted the highest from frying 50 g of chips and 250 g of cheese with an electric frying pan at 190°C.

Meanwhile, values of decay rate among the 6 types of oil present the relatively insignificant differences, the mean value 0.00074 s^{-1} and standard deviation 0.00011 s^{-1} for PM_{10} which is equal to an air change rate of $(2.66 \pm 0.40) h^{-1}$ for the kitchen. This result implies that different types of vegetable oil features emitting similar size characteristics of cooking particles. It will be further verified by the volume distribution presented in latter section of this study. It should be mentioned that the results of this work are comparable to the results obtained by He *et al.* (2005) for natural decay rate of PM_5 derived under nearly non-ventilation condition in a residential house ($3.83 h^{-1}$ for 3–4 μm , $3.21 h^{-1}$ for 2–3 μm , $1.96 h^{-1}$ for 1–2 μm , and averaged $(2.28 \pm 0.89) h^{-1}$ for PM_5).

Sensitivity Analysis of Time Span

In order to verify the 15 min of time span is enough to be applied, the impacts of time span on the results of k and S are further analyzed. As the $C_{in}(t)$ of PM_{10} shown in Fig. 6, the peak concentration is at 180 s after which $\ln(C_{in})$ is calculated and linear regression is carried out with 8 different time spans. The linear correlation coefficient R^2 is

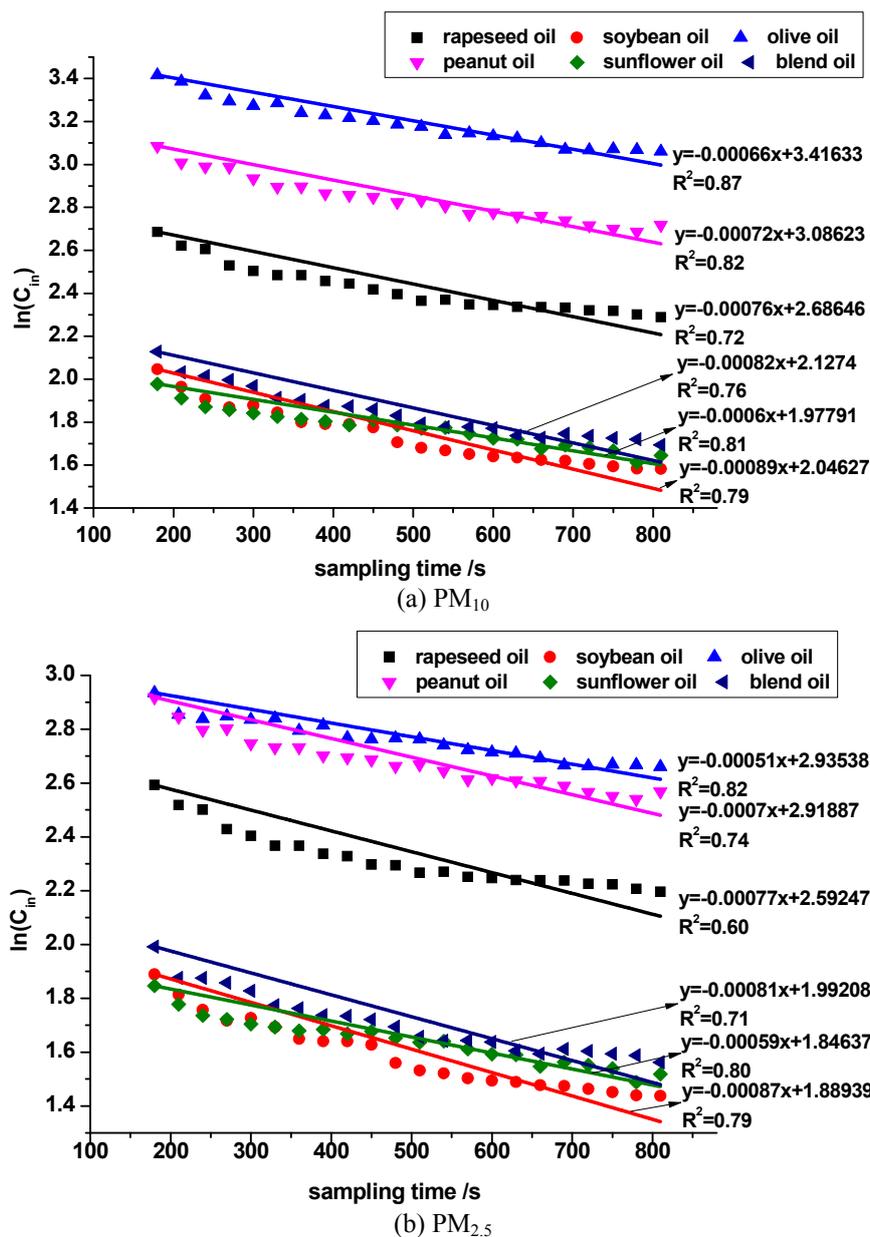


Fig. 4. Linear relationship between $\ln(C_{in})$ and time at the decay stage of mass concentration ($t > t_p$). The negative slope from linear regression denotes the decay rate k .

0.83 in relation to the span 900 s, which is the highest. The value of R^2 decreases to 0.36 for the span 7200 s. If the peak concentration C_p is not strictly fixed, i.e., the intercept of linear regression is allowed to be lower than $\ln(C_p)$, then decay rate for time span larger than 900 s could fall in between the highest 0.83 and its current value as shown in Fig. 6. However, a lower intercept means a reduced peak concentration and is unreasonable. Fig. 7 shows the values of decay rate k , derived as the negative slope of regressed lines in Fig. 6, and the associated values of emission rate S . It is found that the decay rate decreases obviously with the increase in time span. However, the emission rate appears to be not sensitive to the varied time spans. According to the following correlation, both the numerator and denominator decrease with the decrease of k , which makes emission rate

S change slightly with k in a wide range of time span.

$$S = \kappa V \Delta C / (1 - e^{-kt}) \quad (4)$$

As shown in Fig. 7, the largest relative difference of S is about 5.5% regarding the time span changing from 15 min to 2 h, whilst that of k reaches up to 73%. As if an intercept of linear regression lower than $\ln(C_p)$ is allowed as discussed above, measuring time as long as 2 h is not necessary for the derivation of emission rate. It is therefore verified that the time span of 15 min applied in this work is sufficient to derive the reliable results of emission rate. A much longer measurement for peanut oil is further tested, and the decay rate does not change anymore after 3 h and the emission rate keeps almost constant after 15 min.

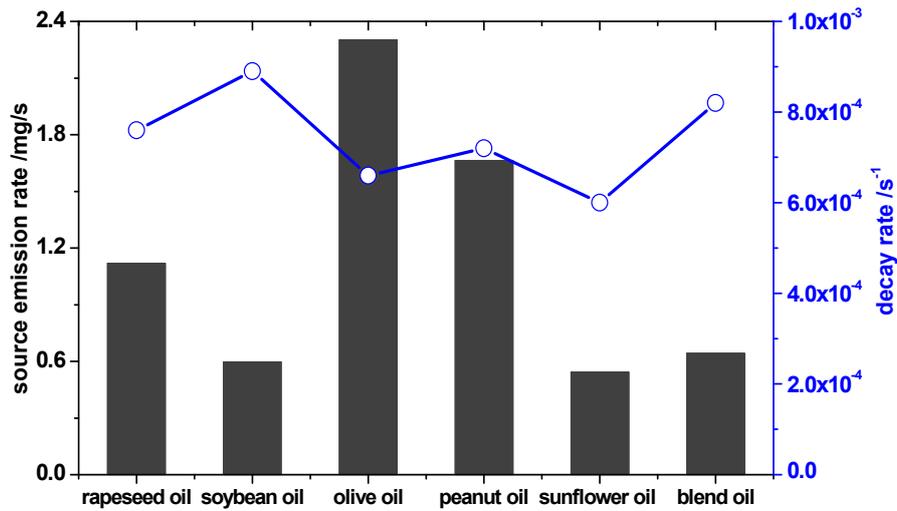


Fig. 5. Emission rate and decay rate of PM₁₀ obtained from the experimental cooking process using different types of oil.

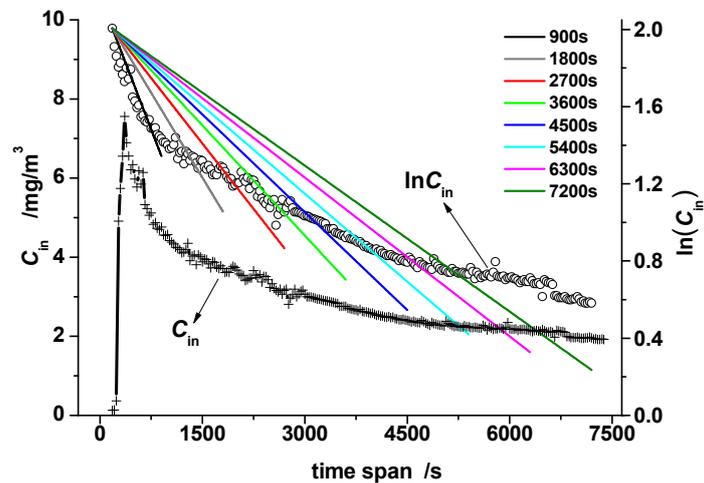


Fig. 6. The measured mass concentrations C_{in} , calculated $\ln(C_{in})$, and the linear regression with different time spans (an example of measured PM₁₀ for the soybean oil).

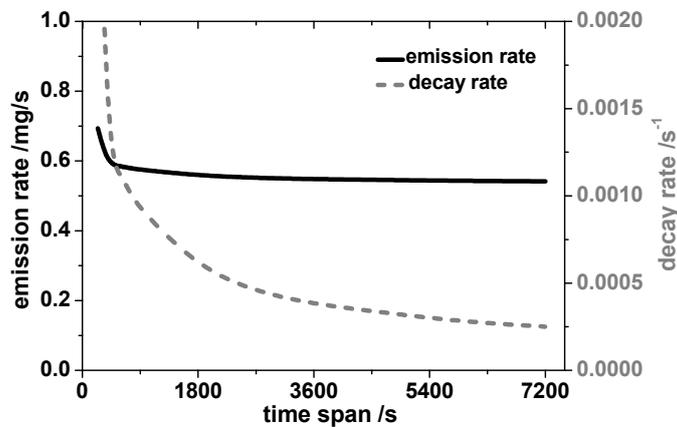


Fig. 7. The relationships between S , k and time span (an example of measured PM₁₀ for the soybean oil).

Volume Distribution of Fume Particles

Fig. 8 presents the measured volume-based size distribution of fume particles for 6 types of oil heated within 2 min. A

single modal distribution in the range from 1.0 to 5.0 μm is obtained for all types of vegetable oil. Mass concentration within this range accounts for nearly 100% of the total mass

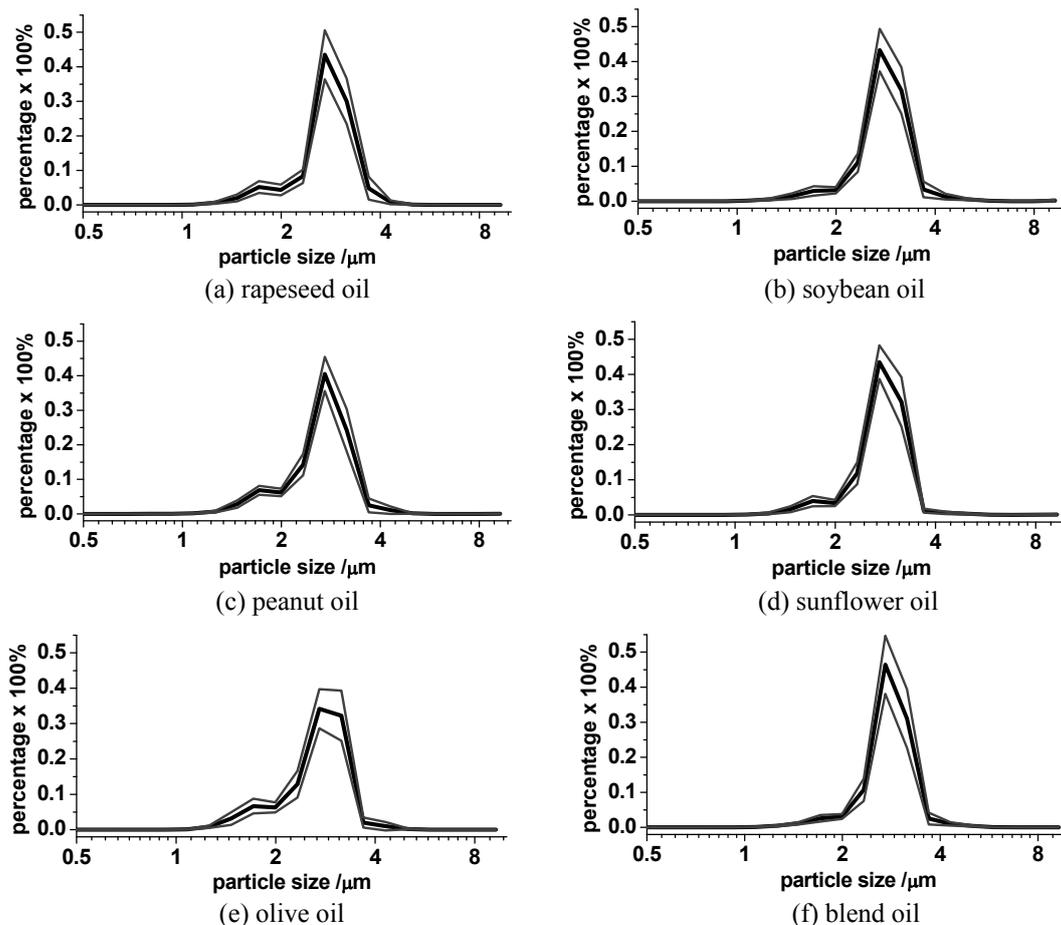


Fig. 8. Measured volume-based size distribution of particles emitted from the heated oil within 2 min. Measurements are repeated 15 times for each type of oil and the differences are statistically expressed by the 95% confidence intervals of mean. The size range is from 0.1 to 10 μm , however, the range of 0.5–10 μm is displayed according to the volume frequency observed.

of PM_{10} . To compare the results of volume distribution monitored by the Malvern sizer with the mass concentration obtained by the TSI Model 8533, we define $(\text{PM}_{2.5}/\text{PM}_{10})_{\text{TSI}} = (\text{PM}_v/\text{PM}_{10})_{\text{Malvern}}$ and then find the following values of x : 3.1 μm (rapeseed oil), 3.0 μm (soybean oil), 2.7 μm (olive oil), 2.9 μm (peanut oil), 3.0 μm (sunflower oil), and 3.0 μm (blend oil). Such comparison does mean there are some differences between the two measuring systems, although the instruments are calibrated every year. Fortunately, the relative differences found still demonstrate good consistency.

Fig. 8 also shows that volume-distribution pattern of cooking particles for olive oil is distinctly different from others and the ratio of cumulative volume frequency of $\text{PM}_{2.5}$ to that of PM_{10} is the lowest among 6 types of oil, corresponding to the largest difference between the mass of $\text{PM}_{2.5}$ and PM_{10} observed in Fig. 3.

Size-dependent Emission Rate

Size-dependent emission rate is calculated via multiplying the volume distribution by total mass rate of PM_{10} . Fig. 9 shows the emission rate as a function of particle size for 6 types of oil. It also presents the cumulative rate associated with the particle size. It is found that particles ranging from

1 to 4 μm account for 100% of the emission rate S and the highest mass rate is at the volume-based mode diameter 2.7 μm . There are significant differences among the peak values at the same mode diameter for 6 types of oil, in which olive oil is the highest in comparison with the lowest appeared in sunflower oil. Distribution of PM_{10} emission rate derived as Fig. 9 can be used as the initial and boundary condition for indoor particle dynamics as studied in (Chen *et al.*, 2000; Lai and Chen, 2007; Lai and Ho, 2008). Such distribution also helps to quantify the size-dependent inhalation intake factors due to cooking in the residential kitchen or flat. In addition, the distribution of emission rate estimated here is actually independent of such variables as room volume, air change rate, and decay rate. Therefore, for the modeling of indoor air quality, size-dependent emission rate is more practical than utilizing the volume or mass concentration for the source definition.

CONCLUSIONS

The decay rate as well as source emission rate of fume particles from the experimental cooking process is investigated in this study. Real-time mass concentration

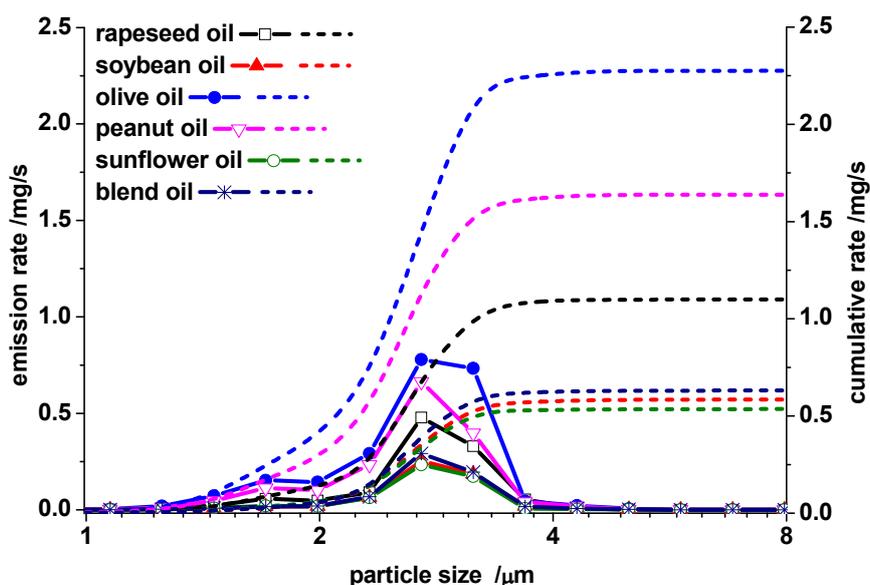


Fig. 9. Size-dependent emission rate of fume particles and the cumulative rate with the increase of particle size. Only PM_{1-8} is displayed due to the insignificant rate observed in other size ranges.

and volume distribution of fume particles are monitored, aiming to determine the size-dependent emission rate. A narrow size span of $PM_{1.0-4.0}$ is obtained within which the highest emission rate is found at $2.7 \mu\text{m}$, i.e., the volume-based mode diameter. A short sampling period of 15 min is proved to be sufficient to derive the reliable source emission rate, although the decay rate exhibits sensitive to the time span applied. Results also show that olive oil emits the highest rate of fume particles while sunflower the lowest during the heating process, the highest being 4.2 times of the lowest. However, the derived decay rate is not sensitive to the oil type, corresponding to the similar volume distribution measured for different types of oil. It is suggested that controlling of indoor air quality for Chinese-style residential kitchen at a specific stage should be carried out in two stages: mass control is the first stage when it is more efficient to reduce the fine particles ranging from 1 to $4 \mu\text{m}$ for decreasing the mass concentration of PM_{10} or $PM_{2.5}$; number control is the second stage when it is uppermost to remove the large number of PM_1 and $PM_{0.1}$ to ensure the low-level exposure to such particles. This present work determines the size-dependent source emission characteristics of a specific cooking stage for indoor particle dynamics. Further studies will be conducted to define the emission rates of different kinds of Chinese-style whole cooking process, and to find out the real individual intake factor through using an appropriate particle dynamical model.

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