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Determination of time- and size-dependent fine particle emission with varied oil heating in an experimental kitchen

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ABSTRACT

Particulate matter (PM) from cooking has caused seriously indoor air pollutant and aroused risk to human health. It is urged to get deep knowledge of their spatial–temporal distribution of source emission characteristics, especially ultrafine particles (UFP < 100 nm) and accumulation mode particles (AMP 100–665 nm). Four commercial cooking oils are auto dipped water to simulate cooking fume under heating to 265°C to investigate PM emission and decay features between 0.03 and 10 μm size dimension by electrical low pressure impactor (ELPI) without ventilation. Rapeseed and sunflower produced high $PM_{2.5}$ around 6.1 mg/m^3, in comparison with those of soybean and corn (5.87 and 4.65 mg/m^3, respectively) at peak emission time between 340 and 460 sec since heating oil, but with the same level of particle numbers 6–9 × 10^5/cm^3. Mean values of $PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$ at peak emission time are around 0.51–0.66 and 0.23–0.29. After 15 min naturally deposition, decay rates of $PM_{1.0}$, $PM_{2.5}$ and PM_{10} are 13.3%–29.8%, 20.1%–33.9% and 41.2%–54.7%, which manifest that $PM_{1.0}$ is quite hard to decay than larger particles, $PM_{2.5}$ and PM_{10}. The majority of the particle emission locates at 43 nm with the largest decay rate at 75%, and shifts to a larger size between 137 and 655 nm after 15 min decay. The decay rates of the particles are sensitive to the oil type.

Introduction

Particulate pollution from cooking emissions has been considered a serious environmental problem that is influencing indoor air quality, regional environments and human health (Buonanno et al., 2011; Huang et al., 2011; Kabir and Kim, 2011; Kim et al., 2011; Zhang et al., 2010). Cooking contributed 0.6%–1.6% to source apportionment of $PM_{2.5}$ collected during the high pollution events of 5–25 January 2013 at the urban sites of Beijing, Shanghai, Guangzhou and Xi'an (Huang et al., 2014). Cooking with gas and stove has been identified as one of the most significant particle generating activities indoors (Abdullahi et al., 2013), responsible for 43.2% of residential indoor PM concentrations (Zhao et al., 2006). The $PM_{2.5}$ concentrations in the cooking samples, meat roasting, cafeteria frying, fish roasting, snack-street boiling, and cafeteria boiling in Ya'an, China were 2.5–9.6 times higher than those in the corresponding backgrounds (Li et al., 2015). Cooking has been associated with lung cancer risk in retrospective case–control studies in Shanghai (Kim et al., 2015).

Many studies investigated the mass concentrations and size distribution of aerosol generated from cooking, which were

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proved to be affected by cooking oils, styles and temperature. Karimatu reviewed the emission range for particle number and mass concentration with different cooking conditions (Abdullahi et al., 2013). $PM_{2.5}$ mass concentrations ranged 0.4–1.8 mg/m^3 from cooking fume were 4.3 to 20.2 times higher than ambient air (Wang et al., 2015). Particle number concentrations increased to 1.4 × 10^6 particles/cm^3 during cooking hours from the background 1220 to 6200 particles/cm^3 (Yu et al., 2015). Torkmahalleh investigated emission rates of seven commercial cooking oils, soybean, safflower, canola, and peanut oils produced lower $PM_{2.5}$ emission fluxes around 10^5 μg/(min·m^2) than corn, coconut, and olive oils with 10^6 μg/(min·m^2) at 197°C (Torkmahalleh et al., 2012). Cooking styles, like frying, grilling, toasting could elevate indoor submicrometer particle number concentration levels by more than five times, while $PM_{2.5}$ concentrations could be up to 3, 30 and 90 times higher than the background levels during smoking, frying and grilling, respectively (He et al., 2004).

However, few have special attention to the time and size distribution for oil heating and their naturally decay character, especially for ultrathin particles. In recent years, there are some report on volume-based size distribution of accumulation and coarse particles ($PM_{0.1–10}$) from cooking fume during oil heating (Gao et al., 2013a, 2013b, 2013c). Wang reported that the average number concentrations of 14.6–100 nm and 100–661.2 nm particles elevated by 10 fold from the background level in the living room and by 20–40 fold in the kitchen, while $PM_{2.5}$ went up to about 160 mg/m^3 in the kitchen and about 60 mg/m^3 in the living room (Wan et al., 2011).

An electrical low pressure impactor (ELPI) manufactured by Dekati Ltd. (Tampere, Finland) was used to collect particles from 30 nm to 10 μm into 12 size fractions. ELPI is widely used for size distribution and density measurement of fine aerosol from wood combustion sources, urban/rural air, pharmaceutical aerosols or motor vehicle exhaust (Coudray et al., 2009; Glover and Chan, 2004; Held et al., 2008; Maricq et al., 2000). The particles pass through a unipolar corona charger, where particle surfaces are saturated with positive charges according to their Stokes diameter, and then impacted on different stages according to their inertia related to their aerodynamic diameter. Finally, the measured current values are inverted to yield particle number concentrations using transfer functions provided by the manufacturer. Mass concentration gives the total mass of all particles in each size range, which is formed by multiplying the current distribution by a vector formed from the masses of spheres having diameter equal to midpoint values of each stage. Due to the dependence of the particle charging efficiency on the Stokes diameter, the particle density must be known for accurate inversions. However, the value of the aerosol density is usually not precisely known so that the particle density estimated of 1.0 g/cm^3 is generally provided by the ELPI technique as a default value for particles measured with aerodynamic diameter (Held et al., 2008). Mass concentrations for cooking fine particles with a gas stove were measured by assuming a specific gravity of 1.0 g/cm^3, derived from typical of combustion particles (Wallace et al., 2004). More details about the ELPI measurement principle can be found elsewhere (Marjamaki et al., 2000).

The aim of this work is to quantify source emission character of particles ranging from 0.03 to 10 μm emitted during a specified oil-heating period for edible commercial oils. In order to gain a better understanding of the relationship between particulate mass and number concentration with heating time and particle size, four commercial oils (rapeseed, sunflower, soybean and corn oils) have been measured by ELPI when heated to 265°C by auto dipping water to simulate cooking fume. Results of this work are expected to provide information on time- and size-dependent fine particle emission dependent on oil style, which will assess the indoor air quality for Chinese style residential kitchen.

1. Experimental method

The size of the kitchen is 4.5 m (L) × 4.0 m (W) × 3.0 m (H) as shown in Fig. 1. Three hundred milliliter different oil (rapeseed, sunflower, soybean and corn oil) in a Supor nonstick pan was heated by liquid gas. When the oil temperature reaches up to 265°C with a real time temperature monitor (PT 1000 temperature sensor), auto dipping equipment will spray 5 ml water within 2 sec following 8 sec interval. After oil temperature reached 265°C again, auto dipping equipment will repeat spray water. Auto dipping equipment was designed to sprayed water twice for all the experiments. Afterwards, fire was shut to stop heating oil. It is observed that they will take within 24 sec to finish auto dipping. The measurements of mass concentration are conducted under nearly no ventilation condition with smoke sucker shutting and the door of the kitchen closed throughout the experimental process, which means the air exchange rate is near zero, and no infiltration of particles from outdoor sources. The kitchen was refreshed with fresh air

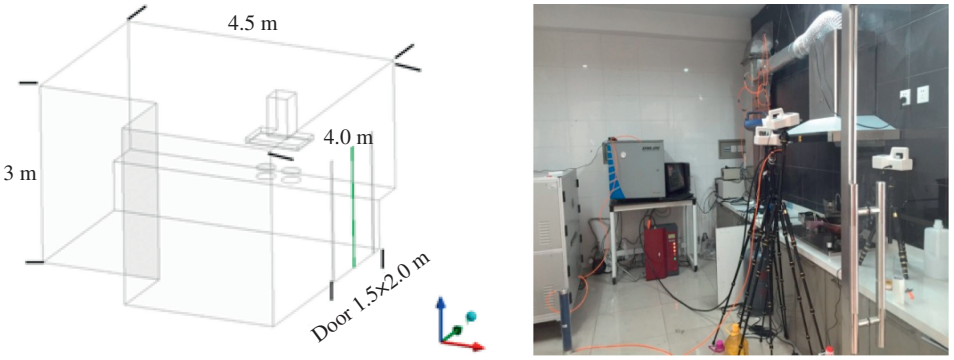

Fig. 1 – Schematic and real kitchen with the layout of measuring point using the TSI and ELPI monitors.

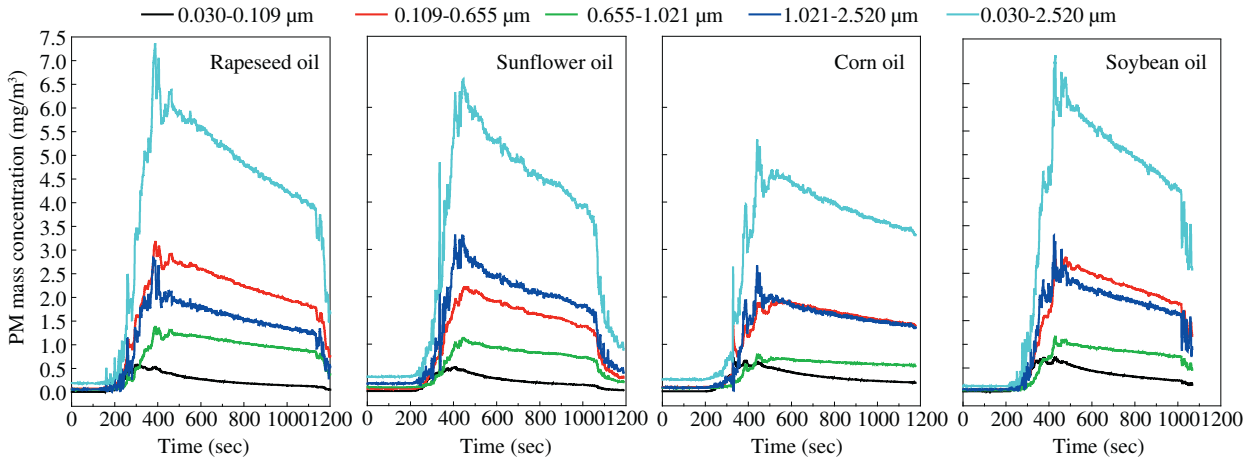

Fig. 2 – PM mass concentration of time-dependent for 5 kinds of particle size dimensions of rapeseed, sunflower, corn and soybean oil, respectively. PM: particulate matter.

thoroughly through the door and the ventilation between each experiment, till the kitchen air returned to background level when there was no cooking. During the course of experiments, no individuals were present in the house except the investigator, and no human activities other than cooking took place in the kitchen.

Particle number concentration was measured at the same time to that of starting heating oil by ELPI in the near real-time sampling system consisting of an aerosol charger and a cascade impactor. The ELPI has cutpoints at 0.030, 0.063, 0.109, 0.173, 0.267, 0.407, 0.655, 1.021, 1.655, 2.520, 4.085, 6.560 and 9.990 μm for stage 1 to stage 13 between 30 nm and 10 μm with operating flow rate of 10.0 L/min and a response time less than 5 s. Particle mass concentrations of size distribution were calculated by assuming that all the particles are perfect spheres with a specific gravity of 1.0 g/cm^3 with the time interval of survey setting to 1 sec. Buonanno characterized the particles collected during cooking, which showed that many spherical-like particles aggregate together (Buonanno et al., 2009). Cooking emitted PMs are quite complex in nature and could vary from one cooking episode to the other, it is impossible to calibrate ELPI charging efficiency using the cooking emitted PM. The results presented in this study should be considered as approximations. The real-time sampling monitor point was located 165 cm vertical to the floor, and 20 cm horizontal to the center of the facies lateralis of fume hood, roughly the breathing level of a standing adult. Particle mass concentration of $PM_{2.5}$ is measured using the 8532 DustTrak™ II (TSI, America) aerosol monitor based on a light scattering technique with 2.0 L/min measuring aerosol flow. The sampling corresponding time was set to 1 sec which corresponded to one scan record. Statistical analysis of the experimental data was performed using the ELPI software. All the collected data are the average value of twice parallel experiment.

2. Results and discussions

2.1. Time-dependent emission feathers

Fig. 2 records the time distribution of mass concentration for 5 kinds of different particle size distribution, 0.03–0.109, 0.109–0.655, 0.655–1.021, 1.021–2.520 and 0.030–2.520 μm, for rapeseed, sunflower, corn and soybean oil, respectively. Oil temperature higher than 200°C is regarded as the necessary condition for Chinese style cooking, which is a little lower to the smoke point of vegetable oil. Cooking fume was reported denser with the increase of oil temperature. For the sake of

Table 1 – Mean mass concentration of fume emission and decay rate of $PM_{1.0}$, $PM_{2.5}$ and PM_{10}, and the ratio of $PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$ at different heating oil time.

Mass concentration	Oil style	$PM_{1.0}$ (mg/m^3)	$PM_{2.5}$ (mg/m^3)	PM_{10} (mg/m^3)	$PM_{1.0}/PM_{2.5}$	$PM_{2.5}/PM_{10}$	Decay rate (%)		
							$PM_{1.0}$	$PM_{2.5}$	PM_{10}
Mean peak value between 400 and 460 sec	Rapeseed	4.09	6.19	21.23	0.66	0.29			
	Sunflower	3.12	6.13	25.20	0.51	0.24			
	Corn	2.53	4.65	18.33	0.54	0.25			
	Soybean	3.35	5.87	25.59	0.57	0.23			
Mean deposition value between 920 and 1000 sec	Rapeseed	3.00	4.41	11.69	0.68	0.38	26.60	28.79	44.96
	Sunflower	2.19	4.05	11.42	0.54	0.35	29.78	33.90	54.66
	Corn	2.14	3.72	10.79	0.59	0.34	13.30	20.12	41.17
	Soybean	2.72	4.42	13.30	0.61	0.33	18.79	24.66	48.05

PM: particulate matter.

producing more fume, the four kinds of oils in all experimental episodes were heated to 265°C. And their heating time to 265°C are counted as 220, 270, 299 and 212 sec, respectively, manifesting that rapeseed and soybean are easier heated than sunflower and corn. $PM_{2.5}$ of four oils showed very low emission level below 0.3 mg/m^3 at the beginning of heating (before 120 sec), similar with the background level. There exists time delay for reaching maximum mass concentration in comparison with the time of finishing auto dipping water. It was 50–71 sec delay for $PM_{0.030–0.109}$ and 130–165 sec delay for other three particle size dimension, 0.109–2.520 μm, It can be observed that $PM_{2.5}$ of four oils reached to 0.65, 0.81, 0.58 and 0.18 mg/m^3 before 265°C, then increased quickly to the maximum values 5.0–7.4 mg/m^3 around 385 s for rapeseed oil, and around 430 s for other oils. After nearly 15 min natural deposition, $PM_{2.5}$ remained around 1.5 mg/m^3 for all oils. The results are comparable to the previous report under non-ventilation condition (Gao et al., 2013c). The effect of particle size to mass concentration is observed as the following rank, $PM_{0.109–0.655}$, $PM_{1.021–2.520} > PM_{0.655–1.021} > PM_{0.030–0.109}$. To further evaluate the difference for mass concentration of particulate emission and decay rates of 4 kind oils, the $PM_{1.0}$ (0.030–1.021 μm), $PM_{2.5}$ (0.030–2.520 μm) and PM_{10} (0.030–9.990 μm) listed in Table 1, representing the mean peak value of the cooking fume for cooking time between 400 and 460 s, while $PM_{1.0}$, $PM_{2.5}$ and PM_{10} between 920 and 1000 sec (Table 1) representing the average PM level after nearly 15 min naturally deposition. As can be seen that the mass concentration emission from varied oils ranked like, rapeseed > sunflower, soybean > corn. Mass concentration ratio of $PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$, together with decay rate of $PM_{1.0}$, $PM_{2.5}$ and PM_{10} are counted (Table 1). Decay rate is calculated with the difference of mass concentration between mean peak value between 400 and 460 s and mean deposition values between 920 and 1000 sec divided by mean peak value listed in Table 1. $PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$ are around 0.51–0.66 and 0.23–0.29, manifested that $PM_{1.0}$ is beyond half $PM_{2.5}$, and $PM_{2.5}$ is nearly 1/4 PM_{10}. $PM_{2.5}/PM_{10}$ detected at the vent of exhausts in different Chinese restaurants usually above 0.6 (Zhao et al., 2007a, 2007b). The results here around 0.3 are quite lower than those of reported, which may derive from the different experiment condition. There existed amount of evaporated water vapor through artificially water dipping under highly heated oil temperature in our experiment, which maybe will increase the content of particle size above 2.5 μm. Furthermore, $PM_{2.5}/PM_{10}$ ratios before dipping water are detected as 0.54,

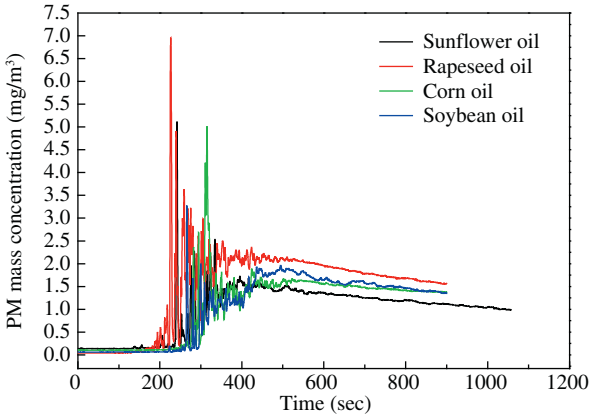

Fig. 4 – $PM_{2.5}$ mass concentration of time-dependent for rapeseed, sunflower, corn and soybean oil, respectively, observed by TSI.

0.48, 0.60, 0.64 for rapeseed, sunflower, corn and soybean oil, respectively, which are close to the reported results, manifesting water content may influence the $PM_{2.5}/PM_{10}$ ratios. Water-based cooking is reported to produce less ultra fine particles and $PM_{2.5}$ than oil-based cooking (Zhang et al., 2010). After 15 min naturally deposition, decay rates of $PM_{1.0}$, $PM_{2.5}$ and PM_{10} are 13.3%–29.8%, 20.1%–33.9% and 41.2%–54.7%. The results show that $PM_{1.0}$ is quite harder to decay than large particles, $PM_{2.5}$ and PM_{10}. Wan reported $PM_{2.5}$ mass concentration could decay to nearly the background level taking 90 min after cooking in the kitchen (Wan et al., 2011).

To evaluate the mass concentration distribution dependent on varied particulate size, their ratios of 4 kinds of particle dimensions to $PM_{2.5}$ for rapeseed, sunflower, corn and soybean oil, respectively based on the mean values at peak emission time between 400 and 460 s presented in Fig. 3. For the 4 kinds of oil, the mean $PM_{0.030–0.109}$ mass concentrations occupy only 6.88%–10.85% $PM_{2.5}$ (Fig. 3). $PM_{0.655–1.021}/PM_{2.5}$ around 13.37%–19.27% are a little larger than $PM_{0.030–0.109}/PM_{2.5}$. The mass concentration with particle sizes between 0.109–0.655 μm and 1.021–2.520 μm all occupy nearly 30–45% $PM_{2.5}$, respectively (Fig. 3).

In order to evaluate the performance of ELPI for monitoring cooking aerosol, TSI was incorporated for comparison to real-time measure $PM_{2.5}$ mass concentration for 4 kinds of oils (Fig. 4). The maximum of $PM_{2.5}$ mass concentration are around 3.0–7.0 mg/m^3, which are a little lower than those determined

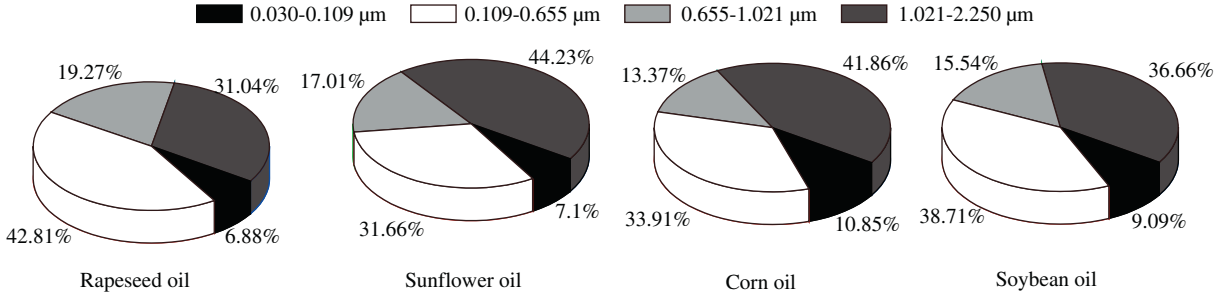

Fig. 3 – Mean mass concentration ratios of 4 kinds of particle dimensions to $PM_{2.5}$ for rapeseed, sunflower, corn and soybean oil, respectively, at peak emission time between 400 and 460 sec.

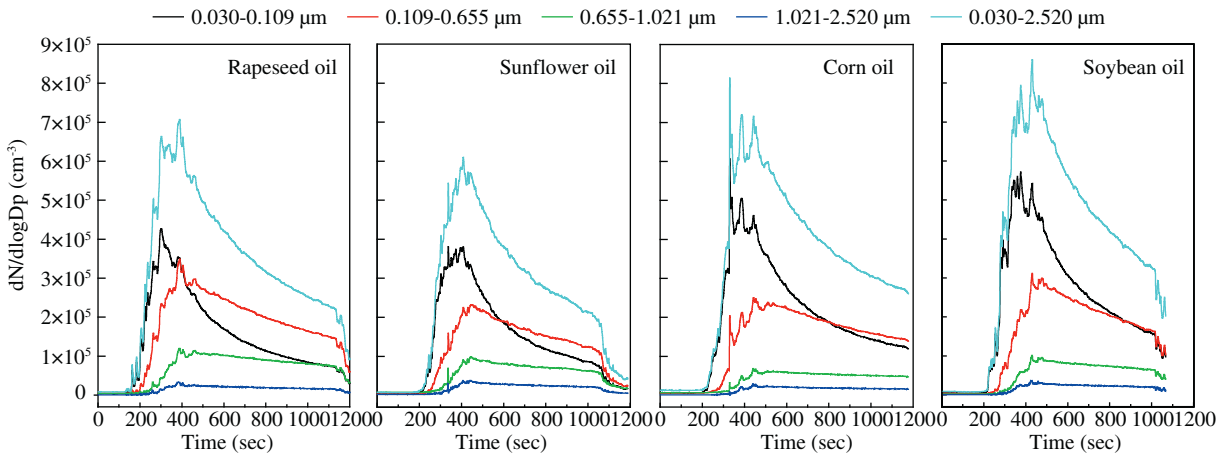

Fig. 5 – Number concentration of time-dependent particle distribution with 5 kinds of particle dimension of rapeseed, sunflower, corn and soybean oil, respectively.

from ELPI. After 15 min decay, $PM_{2.5}$ mass concentration maintained 1.08–1.54 mg/m^3, which are consistent with the results of ELPI. The results manifest that particle mass measurement using ELPI is compared to TSI from the perspective of characterizing $PM_{2.5}$ mass concentration. TSI were widely used for particle size measurements of cooking fume (Torkmahalleh et al., 2012).

Fig. 5 shows time-dependent number concentration distribution of cooking generated particle size dimensions of 0.03–0.109, 0.109–0.655, 0.655–1.021, 1.021–2.520 and 0.030–2.520 μm, for rapeseed, sunflower, corn and soybean oil, respectively. Auto dipping water to the heated oil lead to a remarkable increase in the number concentration of all particle size dimensions from the background level. The order of maximum number concentration of $PM_{2.5}$ are soybean (7.8×10^5) > corn (7.5×10^5) > rapeseed (6.8×10^5) > sunflower (5.7×10^5). The number concentration order of different particle size dimension for 4 kinds of oils are $PM_{0.030–0.109}$ > $PM_{0.109–0.655}$ > > $PM_{0.655–1.021}$ > $PM_{1.021–2.520}$. The number concentration is in the same order of magnitude as many other reports. See reported the number concentration of frying 1.1×10^5 cm^{-3} using a scanning mobility particle sizer (SMPS) (See and Balasubramanian, 2006). Wan investigated the average SMPS number concentration of the Chinese cooking in Hong Kong with $3–9 \times 10^5$ cm^{-3} (Wan et al., 2011). Fig. 6 shows the percentage of the maximum PM number concentration with 4 kinds of different particle dimension to $PM_{2.5}$ for rapeseed, sunflower, corn and soybean oil, respectively. The results revealed the sum of ultrafine particles 0.030–0.109 μm and accumulation particles 0.109–0.655 μm occupied 81.7% to 89.9% to $PM_{2.5}$ number concentration. The $PM_{0.030–0.109}/PM_{2.5}$ ratio for the rapeseed oil is 46.7%, while the ratio for corn oil is high to 59.8%. The number concentration percentage of $PM_{0.655–1.021}$ and $PM_{1.021–2.520}$ to total $PM_{2.5}$ particles present 2.91%–4.46% and 7.14%13.81%, respectively. The data presented in Fig. 6 for $PM_{0.030–0.109}/PM_{0.030/0.655}$ (55–60%) are lower than the previous report with 76–99% for PM (10–100 nm) to (10–500 nm) (Torkmahalleh et al., 2012).

The differences for mass and number concentration distribution among the above experimental results of the four kinds of commercial cooking oils were expected to be influenced by the fatty acid composition discrepancy, like the distinct content of oleic acid, linoleate, stearic acid and palmic acid. Researcher (Snyder et al., 1985) found the volatile compounds present in each stored vegetable oil sample related to the main fatty acid components of the oil. Sunflower seed and corn oil, with the highest amount of linoleate, tended to produce the greatest amount of volatiles, especially pentane and hexanal. Soybean oils, which contain linolenate, formed measurable amounts of 2,4-heptadienal. Katsuta also examine the relationship between the emission of volatile aldehydes with the oils with diverse fatty acid composition (Katsuta et al., 2008).

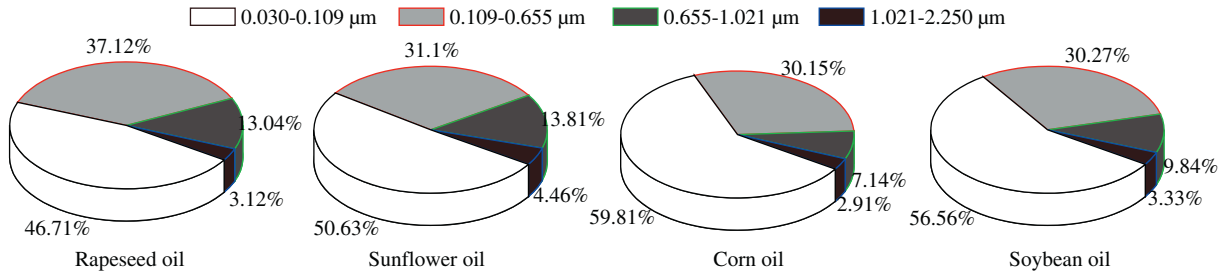

Fig. 6 – The percentage of the maximum number concentration with 4 kinds of different particle size dimension to that of <2.5 μm of rapeseed, sunflower, corn and soybean oil, respectively.

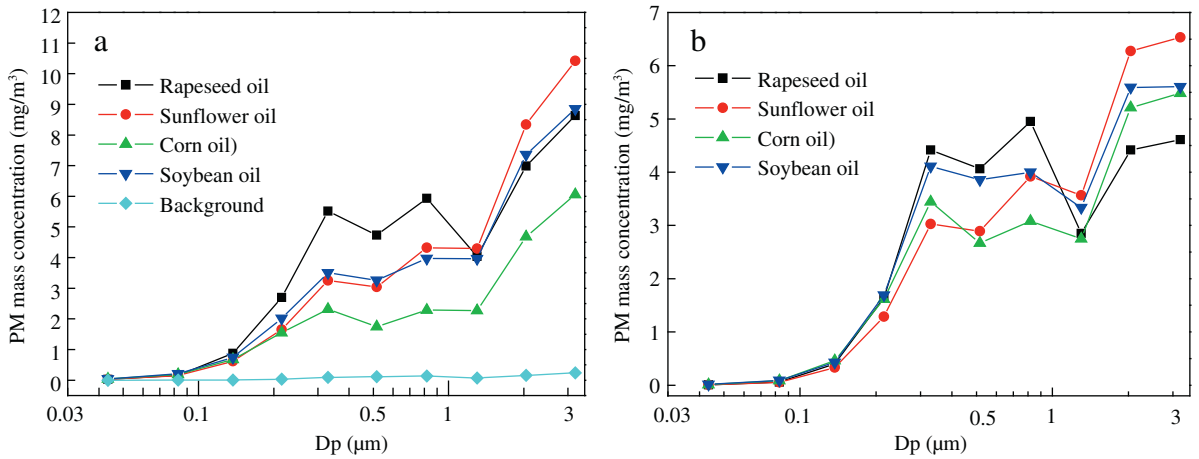

Fig. 7 – Mean particle size distribution of mass concentration for 4 kinds of oils with different cooking time. (a) Between 340 and 460 sec; (c) between 880 and 1000 sec.

2.2. Size-dependent emission feathers

Fig. 7 presents mean particle size distribution of mass concentration for 4 kinds of oils with different cooking time between 340–460 s and 880–1000 s. The horizontal axis represents the particle size (Dp) on a logarithmic scale. The background level of mass concentration is 0.03 mg/m^3. As can be seen that mass concentration of different oil increased quickly with enhanced particle size from 0.03 to 3.0 μm for different cooking time with peak concentration and 15 min decay (Fig. 7a, b). For different size channel, after 15 min decay, mass concentration all presented reduction in different degrees.

Furthermore, Fig. 8 represents mean particle size distribution of number concentration for 4 kinds of oils with different cooking time between 340–460 sec and 880–1000 sec. The decay rates also showed in Fig. 8c to evaluate the natural decay character of varied particle size. The background of number at different size is around 2084–9870/cm^3. The vertical axis represents the normalized number concentration (dN/dlogDp) which allows the size distribution to be compared regardless of the channel resolution. The size distribution of number concentration showed that the majority of emitted particles were more harmful ultrafine particles with the dominant peak at 43 nm for soybean (10.4×10^5), corn (9.4×10^5) and sunflower (6.9×10^5) oils. They all showed a somewhat similar profile for the exception of rapeseed oil with modal peak at 137 nm with 6.5×10^5. The dominant particle size is between 100 and 500 nm with peak modal 137 and 215 nm for all the oils after 15 min of natural deposition. Size with 43 nm owned the largest decay rate of 75%, then gradually decrease to 32%–53% for 137 nm particle size. Particle size of 0.2–1.0 μm exhibited the lowest decay with 15%, then decay rate had a little increase for the size of >1 μm. See noted different particle sizes for different gas cooking methods at peak concentration confined to the ultrafine and nano range, like steaming and boiling with bimodal peak <10 nm and 70–80 nm (See and Balasubramanian, 2006). The results revealed that coagulation of smaller particles to form large particles (300–1000 nm) during dispersion could be a major reason for the extreme decrease of <100 nm, accompanied with a faint decrease of 300–600 nm, even negative decay for corn and soybean oils. This is in good agreement with the study conducted by Wan who suggested that more small size particles could be lost during the dispersion process and coagulated to larger particles for the study of size distribution in kitchen and living room (Wan et al., 2011). The results stated that there existed a quite difference between the mass and number concentration with different particle size distributions. The giant number of ultrafine particle contributed the smallest mass concentration.

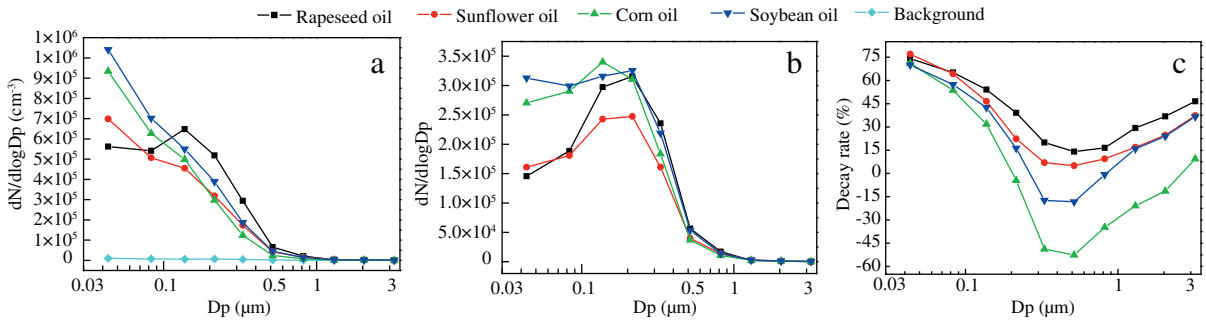

Fig. 8 – Mean particle size distribution of number concentration for 4 kinds of oils with different cooking time. (a) Between 340 and 460 sec; (b) between 880 and 1000 sec; (c) decay rates of size distribution with peak concentration to the decay number concentration between 880 and 1000 sec.

It's suggested that controlling of Chinese style kitchen air quality should be carried out number control, which will be uppermost to remove the large number of below 665 nm to ensure the low-level exposure to such particles, after then carrying out mass control, which will be more efficient to reduce the large particles >655 nm, especially for 1–10 μm for decreasing the mass concentration of $PM_{2.5}$ and PM_{10}.

3. Conclusions

Determination of time- and size-dependent fine particle emission with varied oil heating from the experimental cooking is investigated. Real-time mass and number concentration distribution of fume particles are monitored. The average $PM_{2.5}$ mass concentration at the higher fume emission time between 400 and 460 sec is 4.65–6.19 $\mu g/m^3$, ranked rapeseed > sunflower > soybean > corn oil. The order of maximum number concentration of $PM_{2.5}$ is soybean (7.8×10^5) > corn (7.5×10^5) > rapeseed (6.8×10^5) > sunflower (5.7×10^5). The mass concentration of $PM_{0.030–0.109}$ and $PM_{0.109–0.655}$ accounts for 6.88%–10.85% and 31.66%–42.80% of $PM_{2.5}$, however their number concentration accounts for 46.71%–59.81% and 30.155–37.12% of $PM_{2.5}$, respectively. Particles emitted from heating four kinds of oils could happen naturally deposition, the decay rate of $PM_{1.0}$ mass concentration is the slowest with 13.35–29.8%, followed by $PM_{2.5}$ with 20.15–339% while PM_{10} is the highest beyond 41.2%. The emitted particle size which owned the largest particle number concentration located at 43 nm, and shifted to 137–215 nm after 15 min decay. Results show that ultra fine particles will take place during coagulation to accumulate modal particles.

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