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Physical Characteristics of Ultrafine Particles Emitted from Different Gas Cooking Methods

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ABSTRACT

Gas cooking is a significant source of airborne particles indoors. In order to assess health risk due to exposure from indoor particulate air pollution and to identify effective control strategies, data on size-differentiated aerosol particles released from different cooking methods are critically needed. In this study, controlled experiments were carried out in a domestic kitchen using a scanning mobility particle sizer (SMPS) to investigate the size distribution of ultrafine particles emitted from cooking. Five different cooking methods were studied: steaming, boiling, stir-frying, pan-frying, and deep-frying. During the course of these experiments, the amount and type of food, and the heat setting on the gas stove were kept constant. Results showed that deep-frying caused the largest increase (a 24-fold increase) in particle number concentration to 6.0 x 10^5 cm^{-3} compared to a background concentration of 2.5 x 10^4 cm^{-3} and contained the highest proportion of nanoparticles (90%). This increase was then followed by pan-frying (1.1 x 10^5 cm^{-3}, 78%), stir-frying (9.3 x 10^4 cm^{-3}, 69%), boiling (6.9 x 10^4 cm^{-3}, 62%), and steaming (5.4 x 10^4 cm^{-3}, 55%), implying that cooking with oil produced more particles than cooking with water. It was also observed that steaming and boiling produced a peak in the number concentration of particles at < 10 nm with a second peak at 70 to 80 nm which can be attributed to condensation of water vapor on pre-existing particles. Particle distribution profiles obtained during frying operations were less-distinct compared to steaming and boiling, and demonstrated a modal diameter between 10 and 25 nm. Overall, this study provided comprehensive data on the physical characteristics of particles emitted from cooking, and could be used to evaluate the potential health impacts resulting from exposure to particles indoors.

Keywords: Gas cooking, indoor air quality, number concentration, size distribution, ultrafine particles

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INTRODUCTION

Although several campaigns in the field of aerosol research have been carried out to measure submicron and ultrafine particles, especially in urban areas, an important knowledge gap persists with respect to indoor environments. Understanding of human exposure to indoor particles is important in order to control and reduce exposure. Many studies have investigated the health effects of fine particulate matter ($PM_{2.5}$, particulate matter with an aerodynamic diameter less than or equal to 2.5 μm) (Jamriska et al., 1999; Lee and Chang, 1999; Neas et al., 1993). However, few have paid special attention to the health effects of smaller particles, such as ultrafine and nanoparticles (particulate matter less than or equal to 100 nm and 50 nm, respectively). Both human and animal studies have revealed that ultrafine particles are deposited in all regions of the respiratory tract (Jaques and Kim, 2000; Yeh et al., 1997; Englert, 2004) and are even transported to extra-pulmonary organs, such as the liver, kidney and brain (Oberdorster et al., 2002; Oberdorster et al., 2004). Other than cardiorespiratory problems triggered by lung overload, harmful liquid and gaseous substances on the particle surface can induce inflammatory responses, resulting in more damage (Churg et al., 2005; Gavett et al., 1997).

In many countries, the majority of people spend most of their time (80–90%) indoors. Therefore, the sources of indoor particles, particularly ultrafine and nanoparticles, must be studied and taken into account in environmental studies concerning the health impact of airborne particulate matter. In indoor environments with non-smokers, gas cooking represents one of the most significant activities generating particles (Kamens et al., 1991; Ozkaynak et al., 1996). This particulate source has been specifically associated with respiratory ailments and lung cancer (Hoelscher et al., 2000; Ko et al., 2000), which is thought to be due to ultrafine particles emitted from gas cooking (Dennekamp et al., 2001; Wallace et al., 2004).

In order to gain a better understanding of the relationship between particulate air pollution and gas cooking, several studies have attempted to measure the concentration and size distribution of particles associated with gas cooking (Abt et al., 2000a; Abt et al., 2000b; Dennekamp et al., 2001; Li et al., 1993; Wallace et al., 2004). These studies provided valuable information on the characteristics of particles generated by different cooking methods. However, different types of food were also cooked during the investigations, for example, stir-frying vegetables, frying meat, baking potatoes and boiling soup. Therefore, the differences in particle concentrations and size distributions observed could have been due to a combination of both the cooking methods used and the type of food cooked.

Data on emissions of ultrafine particles from different cooking methods are needed for exposure assessment. The main goal of this work is to provide data on the concentration and size distribution of ultrafine particles from different gas-cooking methods, but with the same food type. The amount of food cooked and the gas-stove heat setting were kept constant during the course of the experiments in a domestic kitchen. The number, surface area, volume, and mass concentrations of size-differentiated particles generated by the different cooking methods were analyzed. In addition, the physical characteristics of particles in the background air in the kitchen were also monitored to assess the increase in particle concentration and the change in size distribution during gas cooking.

EXPERIMENTAL

This study was carried out during March 2005 in the kitchen of a residential house in Singapore. The tiled kitchen (~45m^3) had a two-burner gas stove connected to the local town gas system. Only one burner was used during the cooking experiments. The sampling instrument was placed on an elevated platform with its port facing the used burner. The sampling point was fixed at ~1.5 m above the kitchen floor and ~0.5 m above the gas stove, to simulate the human breathing zone. During the aerosol sampling, no particle sources existed in the kitchen except cooking. All the doors and windows in the kitchen were closed so as to prevent infiltration of particles from outdoor sources. 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 house.

The particle sizing and monitoring instrument used in this work was a TSI Model 3034 Scanning Mobility Particle Sizer (SMPS, TSI Incorporated, MN, USA). The SMPS measures particles at an inlet flow rate of 1 L min^{-1} in the range of 10 to 500 nm over 54 channels and determines the total particle number concentration up to 10^7 cm^{-3}. Number concentration is the primary parameter measured by the SMPS. The size distributions of surface area, volume, and mass concentrations of the particles were calculated based on the particle number distribution by assuming that all the particles are perfect spheres with a specific gravity of 1.0, typical of combustion particles which implies that a particle of volume 1 μm^3 cm^{-3} has a mass of 1 μg m^{-3} (Wallace et al., 2004). The sampling time was set to 3 min which corresponded to one scan per sample.

Five commonly used cooking methods were investigated in this study in terms of their association with indoor particulate air pollution in the kitchen: steaming (to cook over boiling water), boiling (to cook in a liquid heated to, or past, its boiling point), pan-frying (to fry in a small amount of oil), stir-frying (to fry quickly in a small amount of oil over high heat while stirring continuously), and deep-frying (to fry by immersing in hot oil). In all cases, a pack (150 g) of plain tofu (soybean curd), bought from a local supermarket, was cut into 10 circular pieces measuring ~3.0 cm in diameter and ~1.5 cm thick and cooked in the same Chinese wok. A steady heat of 3.0 kW was generated for a sufficient amount of time to obtain a representative number of samples per cooking method each time. Table 1 shows the experimental conditions during different cooking methods. The wok was washed thoroughly between each experiment. The next experiment commenced when particle levels returned to baseline, at which particle number concentration of all sizes fell to within ± 10% of the background concentration measured in the kitchen when there was no cooking.

Table 1. Experimental conditions for different cooking methods.

Method	Properties of water/oil used	Volume of water/oil used (cm^3)	Maximum temperature reached by water/oil (^oC)
Steaming	Tap water	1000	100
Boiling	Specific gravity = 1.0 Boiling point = 100 ^oC	1000	100
Pan-frying	Corn oil	15	190
Stir-frying	Specific gravity = 0.9	15	190
Deep-frying	Boiling point = 245 ^oC Smoking point = 235 ^oC	1000	190

RESULTS AND DISCUSSION

A summary of number, surface and volume/mass concentrations of nanoparticles (10 to 50 nm), ultrafine particles (50 to 100 nm) and accumulation mode particles (100 to 500 nm) is given in Table 2 for background (with no cooking) and the five different cooking methods, and their respective percentages. Volume and mass concentrations are reported together as a volume of 1 $\mu m^3\ cm^{-3}$ is equivalent to a mass of 1 $\mu g\ m^{-3}$. The size distributions of the particle concentration are illustrated in Figure 1 for different weight units. The horizontal axis represents the particle size (Dp) on a logarithmic scale and the vertical axis represents the normalized number concentration (dN/dlogDp) which allows the size distribution to be compared regardless of the channel resolution. The number of modes and the modal diameters can be determined by visual inspection of the individual figures.

From the mean number concentrations in Table 2, it can be seen that gas cooking emitted a substantial amount of particles. A 2- to 24-fold increase in the total counts over the background level was observed during gas cooking, especially during oil-based cooking methods such as pan-frying, stir-frying and deep-frying, compared to water-based cooking methods such as steaming and boiling. This trend in the particle number concentrations can be attributed to high-temperature heating of cooking oil (fatty acids) which presumably generated more particles than the boiling of water. In addition, water with a boiling point of

100 °C is much more volatile than corn oil which has a boiling point of 245 °C. Therefore, under the high temperature in enclosed the kitchen, water droplets are most likely to exist in the gaseous phase than in the particulate phase while less-volatile oil droplets tend to remain as particles.

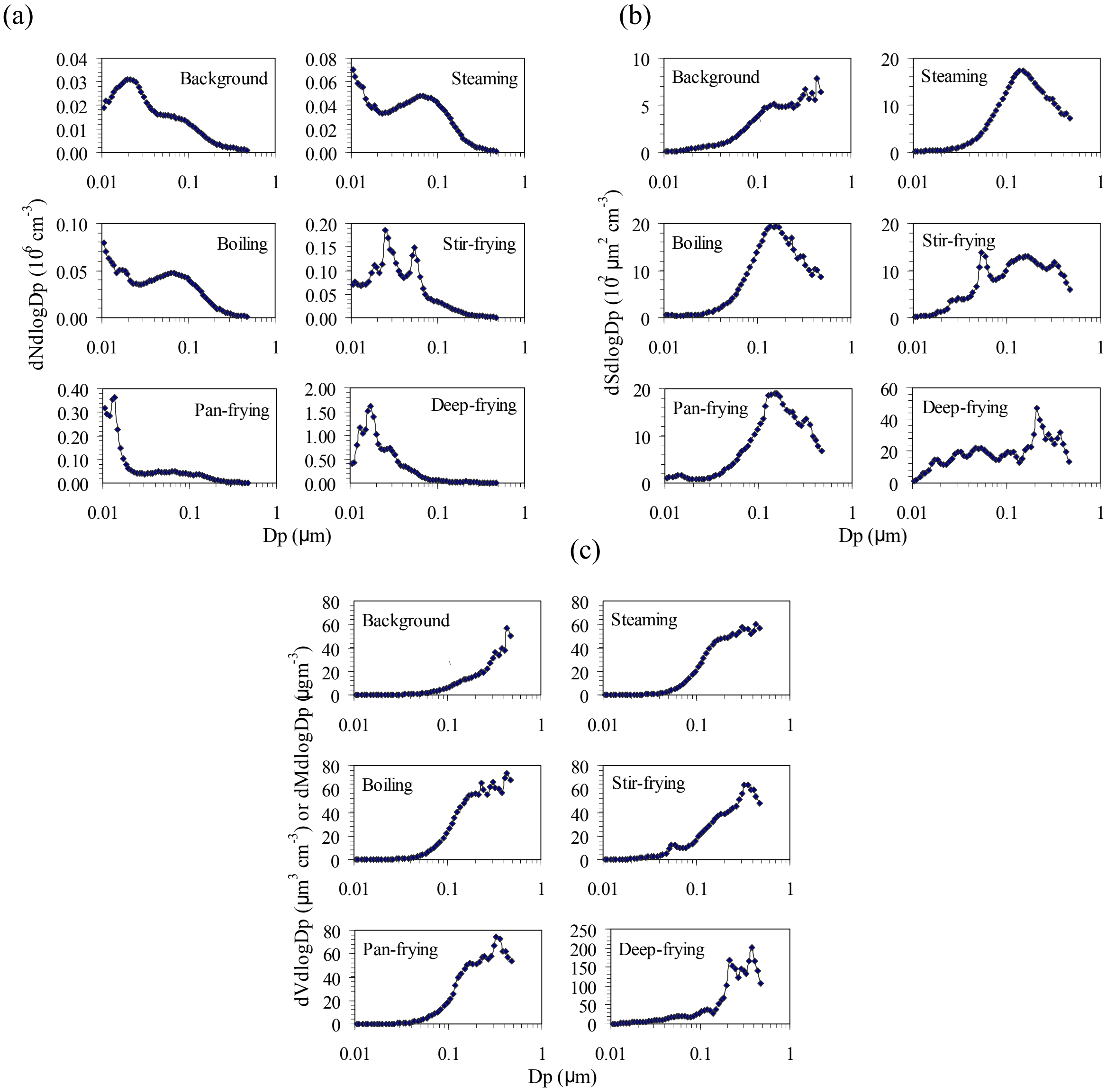

Figure 1. Representative mean size distributions of (a) number, (b) surface area, (c) volume or mass concentrations of particles associated with different cooking methods.

Table 2. Mean number, surface, volume and mass concentrations of size-differentiated particles associated with different cooking methods.

Weight	D_p (μm)	(Mean ± Standard Deviation) concentration and percentages (in parentheses)					
		Background	Steaming	Boiling	Stir-frying	Pan-frying	Deep-frying
Number	0.01–0.05	1.7±0.6 (68±7%)	3.0±0.6 (55±6%)	4.3±2.9 (62±11%)	6.4±3.3 (69±3%)	8.6±4.0 (78±3%)	53.8±26.5 (90±7%)
($10^4 cm^{-3}$)	0.05–0.1	0.5±0.2 (19±4%)	1.4±0.4 (27±4%)	1.5±0.4 (22±7%)	2.1±1.9 (23±6%)	1.4±0.4 (12±2%)	4.2±1.8 (7±4%)
	0.1–0.5	0.3±0.1 (13±3%)	1.0±0.3 (18±3%)	1.1±0.4 (16±4%)	0.8±0.1 (8±3%)	1.0±0.4 (9±1%)	1.4±0.5 (2±3%)
	Total	2.5±0.7	5.4±1.2	6.9±3.5	9.3±5.3	11.0±4.8	59.5±27.3
Surface	0.01–0.05	0.3±0.1 (7±2%)	0.6±0.1 (5±1%)	0.7±0.2 (5±1%)	1.6±0.9 (13±3%)	1.0±0.4 (8±1%)	8.9±4.6 (28±15%)
(10^2 $μm^2cm^{-3}$)	0.05–0.1	0.7±0.2 (16±3%)	2.4±0.7 (20±2%)	2.5±0.7 (19±2%)	3.1±2.1 (25±8%)	2.2±0.7 (17±1%)	5.7±1.6 (18±8%)
	0.1–0.5	3.7±1.2 (77±5%)	8.9±2.3 (75±2%)	10.2±3.3 (76±2%)	7.6±0.8 (61±11%)	9.7±2.4 (75±2%)	17.2±9.6 (54±21%)
	Total	4.8±1.4	11.9±3.0	13.4±4.1	12.4±3.7	12.9±3.5	31.7±9.5
Volume	0.01–0.05	0.2±0.1 (1±0%)	0.3±0.1 (1±0%)	0.4±0.1 (1±0%)	0.9±0.5 (3±1%)	0.5±0.2 (1±0%)	4.3±2.2 (5±3%)
$μm^3cm^{-3}$	0.05–0.1	0.9±0.3 (6±1%)	3.0±0.9 (8±1%)	3.2±1.0 (8±1%)	3.6±2.1 (11±4%)	2.8±0.9 (7±1%)	6.6±0.8 (8±3%)
Or Mass	0.1–0.5	15.9±14 (93±2%)	32.5±7.8 (91±1%)	37.3±10.9 (91±1%)	29.0±2.4 (87±5%)	35.7±6.2 (92±1%)	71.4±40.3 (87±5%)
$μg\ m^{-3}$	Total	17.0±5.8	35.9±8.6	40.9±11.8	33.6±4.9	39.0±7.3	82.3±40.8

As can be seen from the table, deep-frying produced the highest number of particles, followed by stir-frying, pan-frying, boiling, and finally steaming. As the same food was cooked under the same heat setting, the much-higher number concentration of particles observed during deep-frying, compared to stir-frying and pan-frying, can be attributed to the larger quantity of cooking oil used, rather than to oil temperature (Siegmann and Sattler, 1996). Oil temperature remained at ~190 °C before adding the tofu, which was kept at room temperature to reduce the drop in oil temperature. Since the same amount of oil was used for stir-frying and pan-frying, the resulting particle counts were comparable between these two methods. However, the number concentration during stir-frying was found to be slightly lower since the turbulence created by continuous stirring action could have caused a more effective dispersion of particles. Moreover, the temperature of the oil was observed to decrease slightly by ~5 °C during stirring which could partly explain the lower particle concentration. On the other hand, in contrast to the observations reported by Siegmann and Sattler (1996) who found the peak diameter to increase with temperature, the modal diameter did not decrease in the case of stir-frying in spite of the lower temperature. This could be a result of the different techniques employed in the two studies; there was no stirring process in the study by Siegmann and Sattler (1996). Steaming also released lower particle counts than boiling as the wok was covered during steaming and thus particles could have been trapped inside.

This association between particle counts and different cooking methods appeared to be similar to the observations reported in the literature previously. For example, Li et al. (1993) reported that the total number mean concentrations of particles in the size range of 17 to 886 nm were 1.9 x 10^5 cm^{-3}, 3.3 x 10^5 cm^{-3} and 4.0 x 10^5 cm^{-3} during cooking vegetable soup (considered to be boiling according to this study), scrambling eggs (stir-frying), and frying chicken (deep-frying), respectively. Dennekamp et al. (2001) reported a peak UFP concentration of 1.1 x 10^5 cm^{-3} when boiling water, 1.4 x 10^5 cm^{-3} while stir-frying vegetables, and 5.9 x 10^5 cm^{-3} when pan-frying bacon.

The findings from this study further reveal that cooking with oil (stir-frying, pan-frying and deep-frying) liberated more harmful submicron-sized particles than cooking with water (steaming and boiling). Nanoparticles accounted for 69% (stir-frying) to 90% (deep-frying) of all particles during oil-based cooking as compared to 55% during steaming and 62% during boiling. On the other hand, water-based cooking produced more ultrafine and accumulation mode particles presumably due to the higher humidity in the kitchen. The water vapor generated while boiling water could have condensed on pre-existing nanoparticles to form larger particles in the ultrafine and accumulation mode. This is confirmed by the shift in the modal diameter over the course of steaming and boiling experiments as illustrated by Figure 2. Three spectra representing the size distribution at the start, middle and end of the cooking experiment are shown. It can be seen that the peak shifted towards the right as the cooking progressed. The number of particles in the nano range decreased while that in the ultrafine and accumulation mode increased. Such a trend was not observed during the three different frying operations.

During steaming and boiling, the size distributions of the number concentration were clearly bimodal with a dominant peak < 10 nm, a smaller peak at 70 to 80 nm, and a local minimum at ~25 nm. Pan-frying also showed a somewhat similar profile although the mode in the ultrafine range was not so obvious. This is in good agreement with the study conducted by Wallace et al. (2004) despite the fact that the majority of the cooking episodes actually involved frying. The latter study revealed a bimodal distribution with a peak at or below 10 nm and a second peak at ~60 nm with an intervening minimum occurring at ~16 nm from 24 cooking episodes. However, it was found that stir-frying and deep-frying methods showed more complex and jagged profiles with at least three peaks, although all the modal diameters were confined to the ultrafine and nano range.

Dennekamp et al. (2001) noted a larger particle size at peak concentration during pan-frying bacon (69 nm) and stir-frying (41 nm) than during boiling water (17 nm). Wallace et al. (2004) observed a shift towards larger particle sizes during elaborate dinner cooking in comparison to simpler breakfast cooking (mainly boiling water and using an electric toaster). These observations are consistent with the findings reported in this communication. Steaming and boiling produced a peak at < 10 nm whereas the modal diameter was found to be 24.6 nm during stir-frying, 13.8 nm during pan-frying and 17.2 nm during deep-frying.

In contrast to particle counts, the total surface area, volume, and mass concentrations of particles during cooking did not increase significantly. The total particle surface area concentration increased by a factor of two to seven while the total particle volume and mass increased by a factor of two to five over the background levels. This is because small particles have a small surface area, volume, and mass. Hence, the accumulation mode particles which comprised < 20% of total number concentration accounted for 54% to 76% of the total surface concentration and as much as 93% of the total volume/mass concentration. Figure 1 (b and c) shows these trends with the occurrence of the peaks mainly in the accumulation mode. Similar observations were also reported by Wallace et al. (2004).

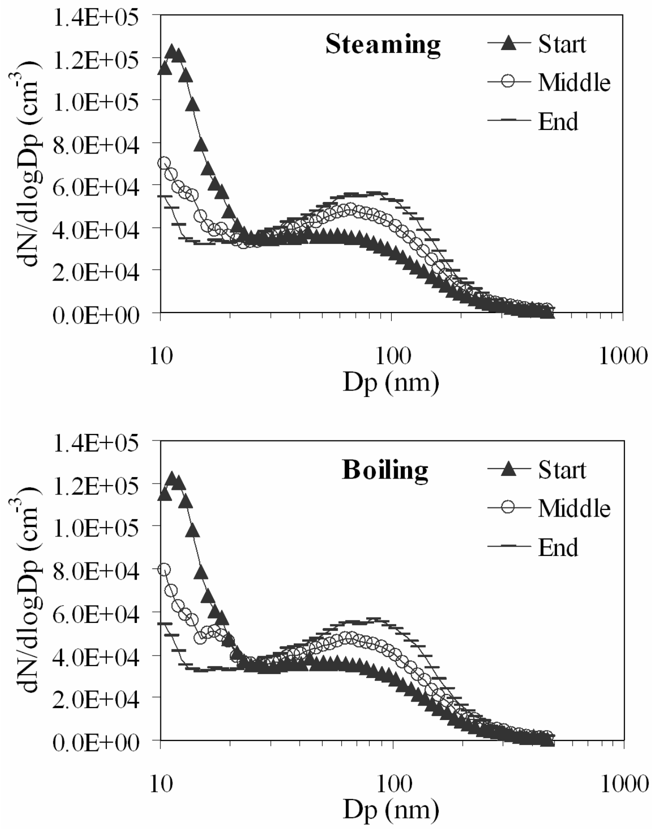

Figure 2. Representative size distributions of particle number concentrations at different times during steaming and boiling.

CONCLUSIONS

Cooking on a gas stove generated a significant amount of ultrafine and nanoparticles. The total number concentration increased by 2 to 24 times over the background levels, with more than 80% being in the nanometer size range. Likewise, total particle surface area, volume, and mass concentration also increased, but in different proportions. There were some variations in the particle concentration and size distribution when different cooking methods were used to cook the same quantity and type of food. It was observed that the deep-frying method emitted the most number of particles and the highest portion of nanoparticles, followed by pan-frying, stir-frying, boiling, and steaming. This trend between the particle number concentrations and different cooking methods implies that cooking with oil causes more particulate emissions than cooking with water. A larger proportion of ultrafine and accumulation mode particles was observed during water-based cooking which is thought to be due to the hygroscopic growth of freshly emitted particles in the presence of high humidity.

Although the overall percentage of nanoparticles found during steaming and boiling was lower than that during the use of oil-based cooking methods, the modal diameter was actually the smallest at < 10 nm. The other peak observed at 70 to 80 nm is attributed to water vapor condensation onto pre-existing particles. On the other hand, the particle size at peak concentration was shifted to between 10 and 25 nm during frying operations. Based on the data obtained from this study, it becomes clear that the people involved in cooking activities in kitchens and possibly other occupants in the same indoor environment can be exposed to an exceedingly large number of ultrafine and nanoparticles, and their health could be adversely affected depending on the chemical composition of the particles.

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