



## Indoor PM<sub>2.5</sub> Characteristics and CO Concentration Related to Water-Based and Oil-Based Cooking Emissions Using a Gas Stove

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### ABSTRACT

Pollutant emissions from indoor cooking activities using clean fuels such as natural gas or LPG are strongly influenced by cooking ingredients and cooking methods. In this study, we explore the characterization of indoor fine particles (PM<sub>2.5</sub>) and CO that are produced by two distinctive cooking methods: frying and boiling. This characterization includes quantifying the presence of fine particles in a kitchen as well as in the adjoining room, analyzing size-segregated carbonaceous materials (EC and OC), and identifying variations in CO associated with the cooking method. Four monitoring devices—a UCB particle monitor, an optical particle counter, a cascade impactor, and a CO monitor—were simultaneously used to measure temporal variations in mass concentrations of fine particles (PM<sub>2.5</sub>), particle number concentrations, their size distributions, and CO concentrations in the two rooms, respectively. EC and OC analyses of the particles collected on a quartz filter by cascade impactor were conducted using the thermal optical method. Frying produced higher emissions of fine particles with a wider range of aerodynamic sizes than boiling. Particle spatial distribution was uniform across the rooms during boiling, because emissions were dominated by very fine particle size. It was observed that particle mass size distributions with cut size  $\leq 0.25 \mu\text{m}$  were predominant in all cooking methods. CO concentration was lowest in tofu boiling and about one-tenth of the stove background level. This is possibly due to the absorption of CO by steam cooking.

**Keywords:** Indoor air pollution; Fine particles; Size distribution; Temporal variation.

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### INTRODUCTION

It is believed that cooking emissions in controlled measurement experiments are influenced by the fuel used and the food being cooked. However, in actual cooking, emission measurement evolves into a complex situation influenced by many factors such as room arrangement, building materials, outdoor infiltration, other combustion devices, ventilation, and cooking methods.

Gas stoves (either using propane gas or natural gas) are widely used throughout the world, particularly in developed countries. In 2008, world gas consumption (all sectors) was around 3.14 trillion m<sup>3</sup>; the consumption increases by 3% per year (IEA, 2010). Compared to other cooking fuels (except for electricity), gas is located in the upper end of the energy ladder, which means that it produces relatively low air emissions than other fossil fuels. However, it should be used cautiously. Gas stoves emit large amounts

of ultrafine particles (UFP) with aerodynamic diameters of less than 100 nm. Wallace *et al.* (2008) found that gas stoves contribute to higher emissions of UFP whose size distribution has a peak around 5 nm in aerodynamic diameter. Meanwhile Li *et al.* (1993) reported that submicron-sized aerosols generated from cooking activities constitute about 60%–70% of UFP in indoor air.

Several recent studies have characterized gas stove emissions with respect to the cooking method that may have distinctive emission patterns, although the term “cooking method” itself still seems indefinite (depending on region, customs, and countries). For instance, on the basis of cooking temperature, Yeung and To (2008) found that aerosol peak number concentrations occurred in the 100–160 nm range and that at higher cooking temperatures, the aerosol mode diameter increased. Moreover, in their experiments, bimodal distributions of submicron-sized aerosols could be expected to be generated. According to Buonanno *et al.* (2009), temperature significantly affected cooking emissions, such that the mass emission factor at the maximum stove power could reach 29 times that of the minimum power, which was used as a baseline. Moreover, the emission factors for high-fat foods were substantially

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higher than those for low-fat foods. During cooking, temporal variability in emissions due to coagulation, condensation, and evaporation may occur.

Wallace *et al.* (2004) found that frying increased the total particle concentration by factors of 6–10 over other cooking methods, while Afshari *et al.* (2005) indicated that the maximum particle concentration reached 150,900 particles/cm<sup>3</sup> when frying meat on an electric stove in a full-scale test chamber. In fact, frying is not the only method for generating remarkable amounts of pollutants; grilling also emits large quantities of pollutants. For example, Lee *et al.* (2001) observed that different cooking styles in commercial restaurants had different emissions of PM<sub>2.5</sub> and CO, and hot pot barbecue emitted the most pollutants.

More straightforward studies, e.g., chemical characterization studies conducted by See and Balasubramanian (2008), revealed that frying emitted more than three times higher PM<sub>2.5</sub> than boiling and at least five times higher than that of the background levels, and organic carbon constituted more than 70% of PM<sub>2.5</sub> in the kitchen. These findings suggest that the risks of household residents' exposure to fine particles emitted from cooking activities vary with the cooking method. Their measurements, however, were carried out under the maximum condition (in an enclosed space and at a distance of around 20 cm from the stove). Yet, there is no information about the relationship between the cooking method and mass size distribution of generated PM<sub>2.5</sub> particles.

Indoor cooking is believed to increase the indoor/outdoor (I/O) pollutant ratio in the kitchen. Cao *et al.* (2005) reported that, in several houses (roadside, urban, rural) in Hong Kong, the average 24 h PM<sub>2.5</sub> I/O measurements had a narrower range (0.8–1.6) than the selected 20 min I/O sampling, which ranged from 0.5 to 6.7. Massey *et al.* (2009) showed that I/O ratios in rural areas were higher than those in roadside and urban areas in Agra, India, indicating more indoor sources of pollutants were present in those settings, particularly from cooking and smoking. Because cooking activities influence the I/O pollutant ratio, the ratio is expected to change according to the cooking method, even if the same fuel is used.

In general, CO emissions are associated with dirty fuels used in developing countries. As a result of low CO emissions from gas stoves, there have been only a few studies concerning this issue. In addition, many people in developed countries use electric stoves instead of gas stoves. Tian *et al.* (2008) reported that the 3 h mean CO concentration in the kitchen reached up to 1.96 mg/m<sup>3</sup> even by using natural gas. However, CO emissions related to the cooking method have not been studied sufficiently.

This study aims to investigate the characteristics of fine particles (PM<sub>2.5</sub>) associated with cooking, particularly temporal variations in the mass and number concentrations in a kitchen and the adjoining room for different cooking methods using a gas stove. Size distributions of carbonaceous particles were also determined for some samples with regard to different cooking methods. Furthermore, to consider human health impacts, we

measured CO concentration related to the cooking method used in the kitchen.

Although this study was conducted in Japan, we set up the measurement conditions to represent those in developing countries (i.e., natural ventilation and tropical weather conditions). Because a combination of cooking methods are used in most households, temporal variation patterns of the number and mass concentrations of fine particles in the kitchen and other rooms should be revealed with regard to the cooking method.

We used natural ventilation in cooking in order to emulate actual conditions in the field. The rate of natural ventilation is difficult to control; however, it is superior to mechanically driven ventilation in terms of energy savings and is the most appropriate in tropical areas where outdoor temperatures do not change considerably throughout the year. Hence, a sufficient natural ventilation system reduces the risk of exposure to fine particles for the persons in charge of cooking in households.

## METHODS

### *Sampling Site Characteristics*

The experiment was performed from July 15 to August 18, 2010 in a single apartment in Kyoto city comprising of a kitchen and an adjoining room. During this period, the weather was somewhat similar to that in a tropical environment.

This apartment had a living area of approximately 8.5 m<sup>2</sup> and a kitchen area of 3 m<sup>2</sup>. This condition is advantageous in terms of minimizing factors with respect to room arrangements. The layout of the sampling sites is depicted in Fig. 1. This apartment is on the first floor of a two-story house. The kitchen is close to the other building, while the adjoining room is adjacent to open space. A standard exhaust fan with a capacity of 550 m<sup>3</sup>/h (18 W) was installed in the kitchen. To emulate the ventilation conditions in developing countries, we used natural ventilation by partially opening windows in the kitchen and in the adjoining room. We set the window opening area in the kitchen at 0.13 m<sup>2</sup> and that in the adjoining room at 0.18 m<sup>2</sup>. During measurements, a common single gas stove for cooking was operated at medium setting instead of maximum or minimum to approximate common practices during cooking. This stove was cleaned with a wet cloth immediately after each cooking task.

We placed all measuring equipment in the spots marked S1 in Fig. 1. S1 was about 1.1 m from the stove. In the adjoining room, the measurement equipment were located near the center of the room. We monitored cooking temperature, ingredients including oil and water, cooking time, stove power, and cooking method. Stove power was controlled by marking the adjustment knob at a specified position, giving moderate power.

### *Measurement Setting*

During these measurements, outdoor meteorological data from the Japan Meteorological Agency showed the ranges of temperature, relative humidity, and wind speed

to be 24–35.6°C, 40%–87%, and 0.1–5.3 m/s, respectively. Background PM<sub>2.5</sub> concentrations were measured three times (on July 20, July 21, and August 17) to reveal the trend in concentration. There were no continuous emission sources nearby. To prevent disturbance from uncontrolled emission sources except from the stove, no other activities were conducted in the apartment. Simple distinctive cooking methods were selected as follows: background, stove firing without cooking, frying, and boiling (pot without lid). Soybean curd (tofu) and chicken were chosen to represent low- and high-fat foods. For each cooking cycle, we used approximately 400 g (for 1 serving) of each food item. It was expected that there were negligible variations of chemical compositions between the two food items because we chose all samples from the same type or brand. We did not use the deep frying method, which is commonly employed in commercial cooking. In our cooking, the recorded maximum temperature was around 163°C during frying and 100°C during boiling. We used a two-step frying method to ensure that all food was immersed in sunflower oil. Half of each ingredient was placed into the heated oil at a time, and the process was repeated again using the rest of ingredient.

Cooking time was determined on the basis of previous studies of household measurements in Indonesia (Huboyo *et al.*, 2009). We cooked on a daily schedule, i.e., in the morning 07.00–07.30 a.m., at midday 11.00–11.30 a.m., and in the afternoon 04.30–05.00 p.m. However, all measurement equipment were run for 12 h (from 6 a.m. to 6 p.m.) to capture indoor air pollution during the day, covering cooking and noncooking periods. Moreover, this time period assures that the indoor pollutant concentration returns to the initial background levels, which minimizes the collection of generated pollutants from the cooking task. For each cooking method, the measurement was replicated on different days.

### Measurement Instrumentation

Particle mass size distribution was measured using a Sioutas Cascade Impactor/CI (SKC Inc) with a Leland Legacy® sample pump at a constant flow rate of 9 L/min. The impactor separates and collects airborne particles into five 50% cut-off size ranges: larger than 2.5 µm, 1.0–2.5 µm, 0.50–1.0 µm, 0.25–0.50 µm, and less than 0.25 µm. As we were interested in PM<sub>2.5</sub>, particles with aerodynamic diameter larger than 2.5 µm were not included in the subsequent analysis. We used a prebaked (600°C, 4 h) quartz filter (Pallflex) with 25 mm diameter for each collection stage and a 37 mm filter for the backup stage. A quartz filter was selected for carbon analysis, as described below. The pump was secured in a semi-closed container to prevent air buoyancy from its outlet. Before weighing the sampling filters on a microbalance (Sartorius, MSP-F) with ± 1 µg accuracy, the filters were conditioned in a desiccator for at least 48 h followed by treatment with zerostat (Milty) to neutralize the static charge accumulated in the filters. Inexpensive UCB particle monitors (Barkeley Air Monitoring Group) were also installed for monitoring temporal variations in PM<sub>2.5</sub> concentration by the light

scattering principle. The outputs of the UCB monitors were calibrated using the impactor; details are specified elsewhere (Huboyo and Tohno, 2010). As part of each cooking task, the photoelectric chamber of the UCB monitors were cleaned.

To account for the uncertainties in handling filters during measurements and for carbon analysis, field blanks were provided. We used a USB-CO data logger (Lascar Co.) to measure indoor CO concentration, which has a measurement range of 0–1000 ppm and ± 6% reading accuracy. Span calibration was performed for the CO monitor using CO standard gas (Sumitomo Seika Co). In addition, daily average indoor temperatures and humidity data were monitored by a USB thermohygrometer (Lascar Co.). Because this study attempts to measure indoor PM<sub>2.5</sub> concentration from the viewpoint of its health impact, the samplers were set at a height of 1.5 m (roughly at the respiration height of a cook) and at a distance of 1 m from the edge of the stove. To minimize interference, the equipment were placed at least 1.5 m from the doors and windows.

Particle number concentrations were continuously measured using an optical particle counter (OPC; KC-01D, Rion Co. Ltd) in five size classes of 0.3–0.5 µm, 0.5–1 µm, 1–2 µm, 2–5 µm, and 5+ µm. Particles larger than 2 µm in diameter were omitted from the data because we are interested in fine particles only. The OPC measurements were set to 2 min cycles. Two OPCs were used simultaneously in the kitchen and the adjoining room; the OPCs were factory calibrated.

OC and EC analyses of particles collected on the quartz filters by the cascade impactor were conducted using a Lab OC-EC Aerosol Analyzer with National Institute for Occupational Safety and Health (NIOSH) Method 5040 (Sunset Laboratory Co.). The OC–EC split time was fixed at 420 s because the deposited particle area was a small slit and optical transmittance correction of pyrolysis did not work well in the automatic split time mode.

## RESULTS AND DISCUSSION

Throughout the measurement period, the average indoor temperature and the average relative humidity in the kitchen were 30.5°C and 74.6%, respectively. In the adjoining room, the temperature was slightly lower (29.9°C) and the humidity was slightly higher (76.1%). Because we did not measure the indoor airstream velocity, we used outdoor meteorological data to analyze the effect of the wind on indoor PM<sub>2.5</sub> concentration. Westerly wind likely contributed to much of the outdoor air infiltration because of the apartment's orientation. In general, the wind was stronger during the afternoon than in the morning and at midday.

Our site had natural cross ventilation (semi cross ventilation to be precise) because of nonsymmetrical opening locations; hence, the ventilation rate was generally high, particularly in the adjoining room, where the window directly faced an open park (Fig. 1). This nonsymmetrical cross ventilation provided well mixed indoor air (Stavrakakis

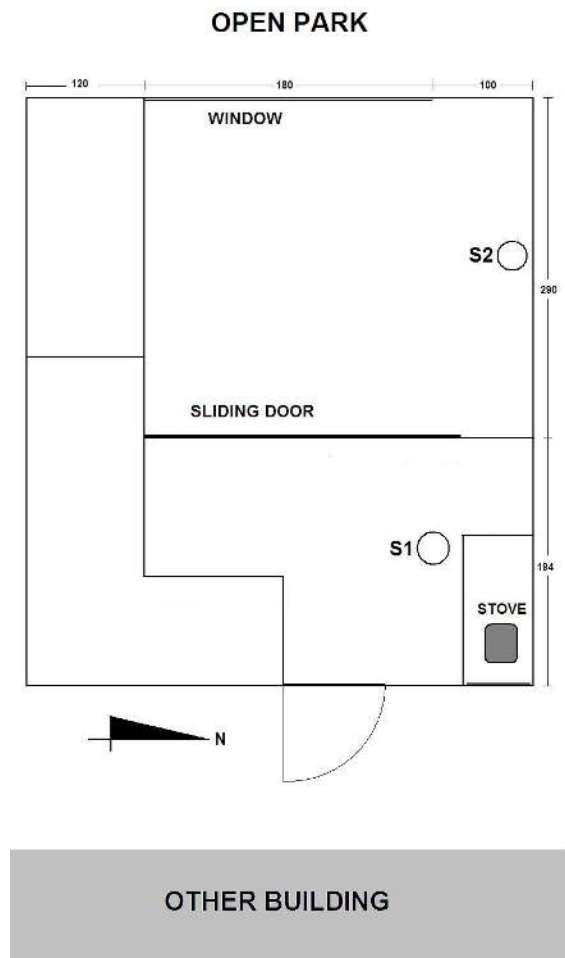


Fig. 1. Sampling sites in the apartment.

*et al.*, 2008). For a substantial analysis, we define “cooking effect” as the period during which indoor particles are highly affected by cooking activities. This simply facilitates comparative analysis of particle properties between the cooking methods and between the two rooms. The cooking effect is quite different for the mass and number concentrations of particles, because in most cases,

cooking effects lasted much longer on a quantity basis than that on a mass basis.

Approximately 30 min after each cooking period, the number and mass concentrations of fine particles returned to the initial conditions. Therefore, choosing a period of 1 h (from ignition of the stove) as the timeframe for comparison between the cooking methods and between the rooms is reasonable.

#### Outdoor Measurements

Typical diurnal variations in ambient  $PM_{2.5}$  concentration from continuous monitoring in Kyoto city (i.e., Fushimi ward in the south and Sakyo ward in the north) during this study are shown in Fig. 2. We used ambient  $PM_{2.5}$  monitoring data from Sakyo ward to approximate outdoor  $PM_{2.5}$  concentration because the sampling location is near our study site (about 1 km away).  $PM_{2.5}$  mass concentration in Sakyo ward (measured by the Energy and Environment Laboratory at Kyoto University) was monitored by TEOM1400 (Rupprecht & Patashnick), while in Fushimi ward (measured by the Kyoto Prefectural Institute of Public Health and Environment), the measurements were carried out by a  $PM_{2.5}$  monitor (Kimoto) based on beta-ray attenuation. From Fig. 2, it is clear that ambient  $PM_{2.5}$  concentrations tend to increase in the afternoon with small variations from day to day. Therefore, a westerly wind in the afternoon will strongly affect indoor air quality at our location. To quantify the contribution of outdoor air to the indoor environment, we calculated the I/O ratio. Average ratios of paired indoor to outdoor  $PM_{2.5}$  concentrations during the measurement period are presented in Table 1.

Table 1 shows that cooking activities resulted in high I/O ratios both in the kitchen and in the adjoining room. In the kitchen, I/O ratios during cooking were 3–4 fold higher than 12 h average I/O ratios, while in the adjoining room, the ratios were 1.1–2.9 fold higher. Our ratios are higher than those of Lee *et al.* (2001), who found that the mean I/O ratio of  $PM_{2.5}$  was around 15 for frying. Moreover, our outdoor  $PM_{2.5}$  concentrations were relatively lower than those of other studies in urban areas of other Asian countries (Tsai *et al.*, 2000; Zhao *et al.*, 2009) and were

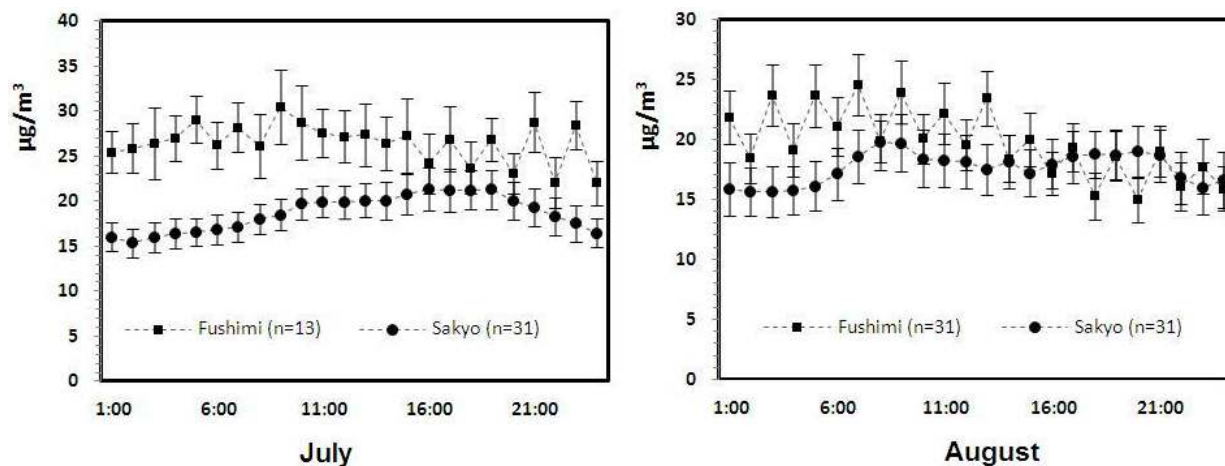


Fig. 2. Diurnal Variations of Ambient  $PM_{2.5}$  concentrations in Kyoto City in July and August, 2010.

**Table 1.** Average of paired indoor to outdoor ratios (I/O) of PM<sub>2.5</sub> concentrations.

Cooking method	Outdoor conc* μg/m <sup>3</sup>	Indoor conc‡ μg/m <sup>3</sup>	I/O ratio during cooking*		12 h I/O ratio†	
			K	A	K	A
Tofu boiling	0.3–18.1	1.21–294	8.92(1.93)	2.39(0.54)	2.84(0.88)	2.09(0.31)
Tofu frying	0.5–50.5	1.76–707	28.41(7.05)	4.49(0.86)	7.19(2.45)	1.78(0.32)
Chicken boiling	0.1–56.5	5.36–1,082	17.30(6.33)	0.97(0.19)	3.96(2.00)	0.62(0.09)
Chicken frying	0.2–51.9	1.67–1,366	27.13(6.09)	4.65(0.79)	6.18(1.82)	1.59(0.29)

\* averaged over 10 min interval; † averaged over 1 h interval; ‡ measured over 2 min interval, kitchen only; In parentheses are standard errors. Note: K = Kitchen, A = Adjoining Room.

comparable to other data measured in Japan (Khan *et al.*, 2010). Nonetheless, note that because the outdoor concentration during the measurements fluctuated widely (0.1–56.5 μg/m<sup>3</sup>), the I/O ratio also oscillated, particularly when 10 min average PM<sub>2.5</sub> concentration was used.

### Temporal Variation in Mass Concentration

Temporal variation patterns of mass concentrations in the kitchen and the adjoining room were similar for either tofu or chicken frying for the three consecutive cooking periods. The mass concentration in the kitchen was around six times higher than that in the adjoining room. This indicated that particle concentration was reduced during transportation to the adjoining room. This could be caused by air exchange between the rooms, deposition of the particles on surrounding surfaces, or other removal processes (Hussein *et al.*, 2006). In contrast, boiling showed different pattern profiles in the three consecutive cooking tasks. Only frying emitted a high load of fine particles in the kitchen and still substantially affected indoor air in the adjoining room. Temporal variations in PM<sub>2.5</sub> concentration in the kitchen showed a distinctive peak with narrow-base shape during frying (Fig. 3(a)), which means high concentration lasted for a short time. In contrast, in the adjoining room, the concentration peak was shorter with wider base (Fig. 3(b)). Spreading of cooking smoke from the kitchen to the adjoining room caused a time lag between concentration peaks in the two rooms and a broader peak in the adjoining room. Hussein *et al.* (2006) observed that PM concentration in a living room elevated to as much as 30% of the peak concentration in the kitchen, with a time lag of minutes to half an hour, during cooking at a home in Prague.

Fig. 3 also shows that particle contributions from the stove itself (from fuel burning) were relatively small because its background concentration profile oscillated only to a little extent during cooking periods. In the adjoining room, the concentration profile of the stove background was somewhat similar to the overall background concentration. According to the studies of Wallace *et al.* (2008) and Fan and Zhang (2001), higher concentrations of UFP (5–100 nm) are emitted from the burning of combustion devices alone, but it is difficult to detect them through the UCB particle monitor. This photoelectric monitor is sensitive to PM<sub>2.5</sub>; however, it is less sensitive to particles with aerodynamic diameters less than 1 μm (Chowdhury *et al.*, 2007). Our study also confirmed that, as compared to low-fat foods, high-fat

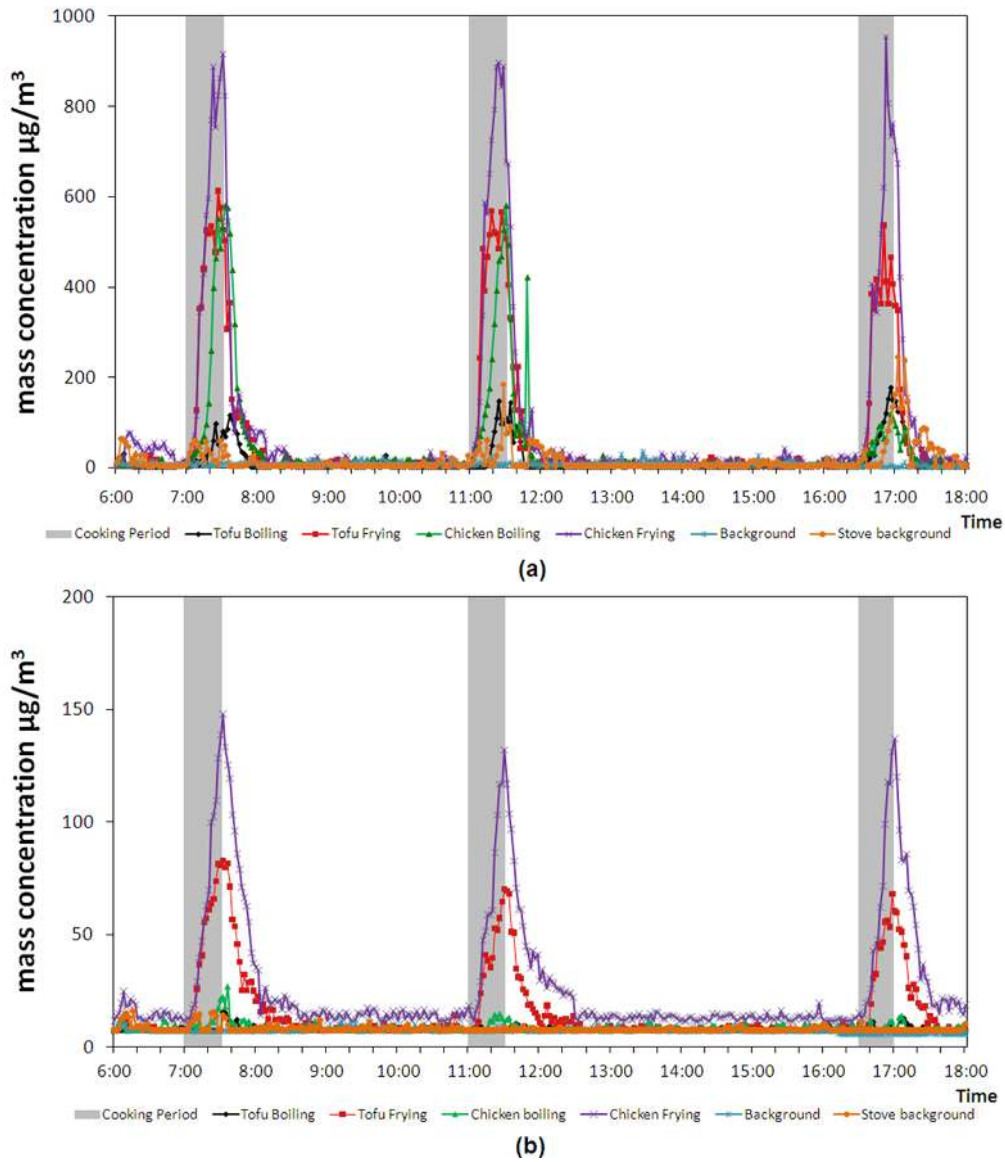
foods have higher PM mass emissions during cooking. In addition, frying generated more fine particle emissions than water-based cooking, as previously described by See and Balasubramanian (2008) and Buonanno *et al.* (2009).

If we assume negligible emission variations for each repetition of a cooking method, then natural ventilation helped to dissipate the pollutants in indoor air. As shown in Fig. 4, strong westerly prevailing winds reduced the average concentration of PM<sub>2.5</sub> both in the kitchen and in the adjoining room. These results were observed on the first day in the afternoon during tofu frying, on the first and second days in the afternoon during chicken boiling, and during the second day in the afternoon during chicken frying. Nikas *et al.* (2010) confirmed that beside the geometry of the openings of a building and the incidence angle of the wind, the magnitude of wind velocity has the most significant effect on the air exchange rate of a building because of its proportionality to the inlet volume and flow rate of the ventilation. Thus, wind speed considerably affects indoor PM<sub>2.5</sub> concentrations if the wind approaches the indoor opening from a fixed direction.

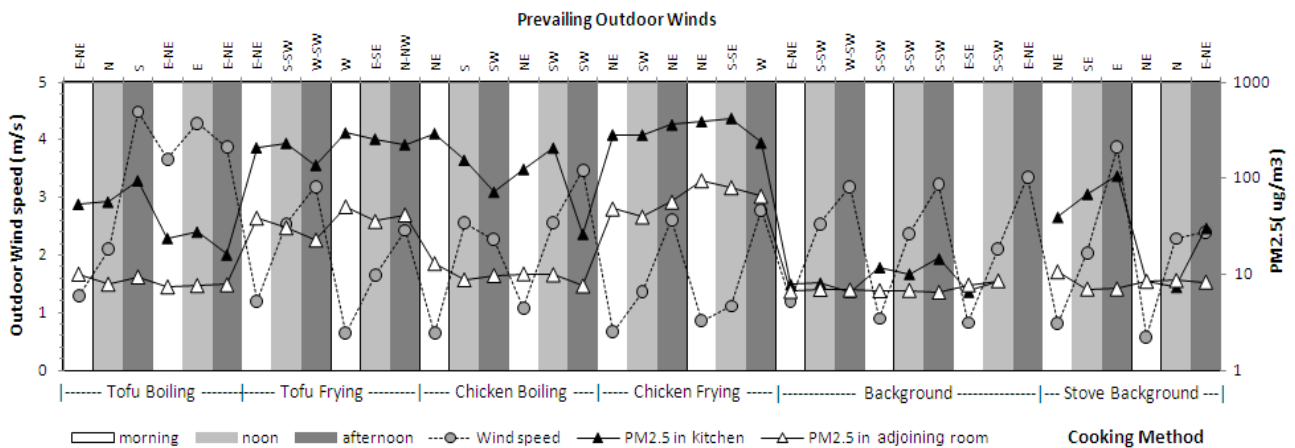
### Particle Number Concentration

In general, number concentration in submicrometer mode was significantly affected by cooking activity, irrespective of the cooking method. Temporal variations in the number concentrations (Fig. 5) showed particles with diameters of 0.3–5 μm fluctuated widely during frying, while during boiling, only those of 0.3–1 μm fluctuated. On average, during boiling (chicken and tofu), number concentrations of particles in the diameter range of 0.3–0.5 μm increased by 29%–48% over initial conditions. During frying, number concentrations of these particle sizes increased by 134%–247% over the initial values. Comparable results were obtained in the adjoining room, where the increases over the background levels were 20%–36% and 127%–237% for boiling and frying, respectively. Sjaastad *et al.* (2008) found that during frying a beefsteak, the highest number concentration of fine particles (in the size range of 0.3–0.5 μm) in the adjoining room was only 5.8% of that in the kitchen. Dominance of fine particles in the number concentration can be explained by the coagulation shifting of UFP, which are the most notable products of combustion. The shift of UFP to larger particles, related to stove combustion, has been suggested by Dennekamp *et al.* (2001) and Wallace *et al.* (2008).

Table 2 indicates that during the cooking effect, particle number concentrations in the kitchen and the adjoining



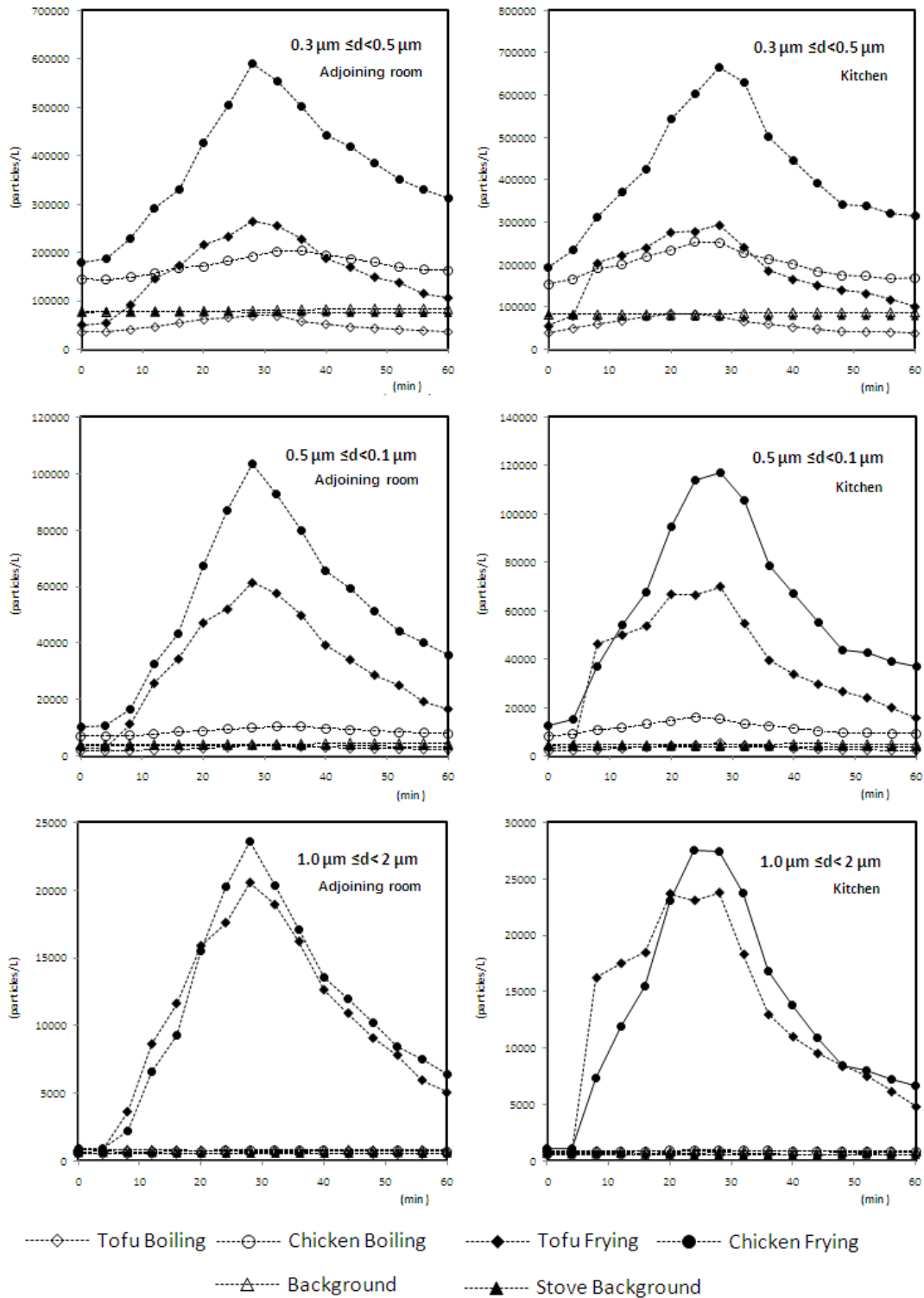
**Fig. 3.** Two-day average variations in fine mass concentrations in the kitchen (a) and the adjoining room (b). Background concentrations are three-day averages.



‡ All samples were duplicated in other day except for background (in triplicate)

**Fig. 4.** Prevailing Outdoor Wind directions and Wind Speed Related by Cooking Methods. PM<sub>2.5</sub> concentrations are also plotted.





Each cooking method: n = 6, except for background: n = 9

**Fig. 5.** Average temporal variations in the number concentrations of fine particles in the kitchen and the adjoining room during cooking effects.

room correlate better for boiling than for frying. During boiling, aerosols were homogenously distributed, unlike during frying. This might be due to the existence of water vapor droplets during boiling, as suggested by See and Balasubramanian (2006). In contrast, frying generated

more submicron-sized particles than boiling, and the emitted aerosols were less volatile (remaining in particle state) and more easily coagulated (Yeung and To, 2008). These growing particles settled during transportation to the adjoining room. Therefore, fine particle concentration

**Table 2.** Relationship between fine particle number concentrations in the kitchen and the adjoining room during cooking effects.

	Slope ( $\beta$ )	Intercept*	Std error ( $S_{Y_{ex}}$ )*	Correlation coefficient†
<b>0.3–0.5 <math>\mu\text{m}</math></b>				
Tofu Boiling	0.825	1305	9927	$r = 0.967$
Tofu Frying	0.728	29303	44193	$r = 0.833$
Chicken Boiling	0.765	21426	32718	$r = 0.956$
Chicken Frying	0.844	26517	61417	$r = 0.920$
Background	0.961	–964	1281	$r = 1.000$
Stove Background	1.038	–3894	4550	$r = 0.991$
<b>0.5–1.0 <math>\mu\text{m}</math></b>				
Tofu Boiling	0.710	257	488	$r = 0.962$
Tofu Frying	0.667	6574	12812	$r = 0.793$
Chicken Boiling	0.616	1615	2096	$r = 0.928$
Chicken Frying	0.801	3375	13128	$r = 0.911$
Background	0.817	354	218	$r = 0.996$
Stove Background	0.898	4	257	$r = 0.991$
<b>1.0–2.0 <math>\mu\text{m}</math></b>				
Tofu Boiling	0.919	2	42	$r = 0.991$
Tofu Frying	0.634	2321	4337	$r = 0.792$
Chicken Boiling	0.728	109	59	$r = 0.954$
Chicken Frying	0.774	753	3187	$r = 0.911$
Background	0.908	28	53	$r = 0.948$
Stove Background	0.870	28	39	$r = 0.972$

\* The units are in particles/L; † Pearson correlation coefficient at  $p < 0.01$  level (two-tailed).

decreased in the adjoining room. Gravitational settling is the major removal process of fine particles as they become larger because of coagulation (Afshari *et al.*, 2005). Correlation coefficients for the overall background as well as stove background were almost unity, indicating relatively no difference in the behavior of fine particles between the two rooms.

#### Size-fractionated Carbonaceous $\text{PM}_{2.5}$

As summarized in Table 3 and depicted in Fig. 6, a high percentage of OC was found in  $\text{PM}_{2.5}$  and very high OC concentration was observed during chicken frying. Because we did not characterize the sunflower oil used in frying or the compositions of the ingredients, it was hard to estimate conversion factors for OC to Organic Matter (OM). See and Balasubramanian (2008) found that the proportions of EC to  $\text{PM}_{2.5}$  were 9% in boiling and 8%–12% in frying; however, these were 3% and 2.6%, respectively, in our results (using the same ingredient, tofu). Our results for OC proportions were much lower, i.e., 39% compared to 44% in boiling and 39% compared to 52%–63% in frying. Note that our sample was taken about 1 m away from the stove (closely related to ambient indoor) while See and Balasubramanian (2008) sampled near the stove (about 20 cm away). Therefore, fewer fine particles were captured in our case. Moreover, lower frying temperatures in this study produced fewer oil mist emissions. Another possible reason for the variations was that outdoor EC and OC particles might penetrate indoors and alter the indoor carbonaceous  $\text{PM}_{2.5}$  concentration because we adopted natural ventilation and 12 h measurements.

**Table 3.** OC and EC mass concentrations in  $\text{PM}_{2.5}$ .

Samples	Concentration		
	$\text{PM}_{2.5}$ ( $\mu\text{g}/\text{m}^3$ )	OC ( $\mu\text{g C}/\text{m}^3$ )	EC ( $\mu\text{g C}/\text{m}^3$ )
Tofu boiling	22.88	8.93	0.78
Tofu frying	41.12	16.31	1.11
Chicken boiling	30.86	6.82	1.16
Chicken frying	101.64	43.27	4.09
Background	21.71	10.12	0.86
Stove background	23.27	9.66	1.33

Strangely, the background OC concentration in this study was higher (46% in  $\text{PM}_{2.5}$ ) than that of tofu boiling and chicken boiling. It originated from a nearby pollutant source because only at that particular daytime, the author suspected smoke from burning incense came up from a neighborhood building. Ambient ratio of OC to  $\text{PM}_{2.5}$  was only around 13% in previous measurements in Kyoto (He *et al.*, 2004). Outdoor  $\text{PM}_{2.5}$  might alter the composition of indoor  $\text{PM}_{2.5}$  if the concentrations are much different. On the basis of the total mass size distribution, the dominant size range was less than 0.25  $\mu\text{m}$ , followed by 0.25–0.5  $\mu\text{m}$ . Tofu boiling generated the fewest number of fine particles (compared to the background levels), particularly in the size range of 1–2.5  $\mu\text{m}$ . All samples exhibited unimodal distributions in the size range of less than 0.25  $\mu\text{m}$ . Our results are similar to those of Chao *et al.* (2002), who showed that the dominant size range was below 0.4  $\mu\text{m}$ . This measurement was conducted in only one sample per cooking method; thus, uncertainty may arise regarding the result.



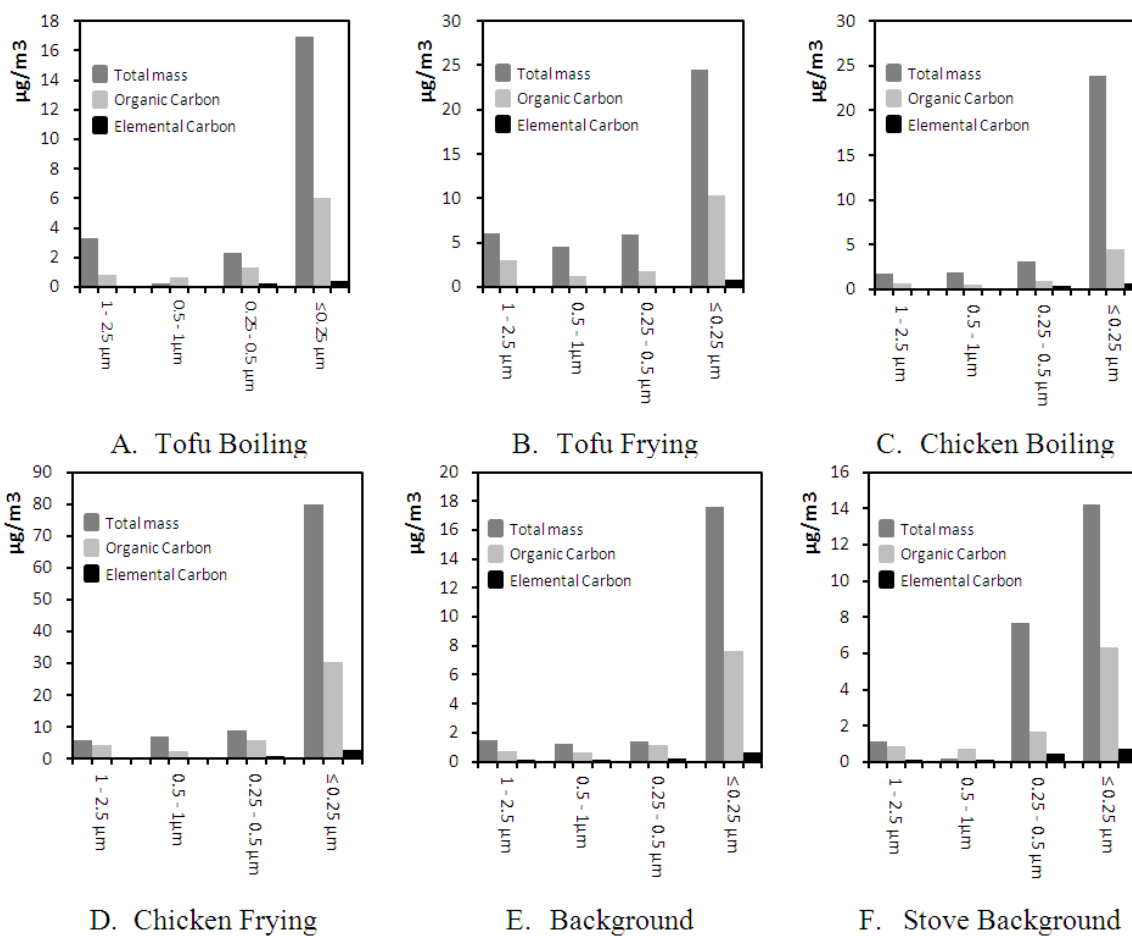


Fig. 6. Mass Size Distributions of Indoor Carbonaceous Particles Related to Cooking Methods.

### CO profiles Related to Cooking Method

We assume that there was no difference in energy (city gas) consumption for the cooking methods, as we followed the same procedure to treat the stove power for all measurements. Because we could not control the outdoor CO, relative CO concentration to the background was selected, as described in Fig. 7. In Japan, ambient outdoor CO is quiet low, i.e., below 1 ppm for a 24 h average (Ministry of the Environment, Government of Japan, 2005). Measured average background CO was below 0.5 ppm because our CO monitor has a resolution of 0.5 ppm.

The unique result is that stove background showed the highest concentration of CO during cooking effects, while boiling generated the least CO. This finding may be due to the fact that CO in the kitchen, which mainly originates from stove emissions, might not be easily absorbed by hot oil mists generated by frying. In contrast, boiling produces water mist, which can absorb CO gas because of comparable polarities. CO gas with a solubility of 0.024 g/kg of water (at 30°C) has a similar degree of polarity (1.95) compared with water (1.45) (Weast, 1988). In addition, we did not use a lid in this study; therefore, much water mist was generated during boiling. In full-scale experimental studies, Fang *et al.* (2006) used water mist to quench cooking oil fires and found that this mist could reduce both CO and CO<sub>2</sub> concentrations. However, further

study with fully controlled variables in test kitchens such as a chamber should be conducted to confirm whether CO is absorbed or dissipated from the room because of ventilation.

In general, using the same ingredients of food as a basis for comparison, frying increased the CO concentration over boiling by a factor of 1.4–2.5 (for chicken) and 3.8–4.5 (for tofu). During chicken frying, CO concentration increased by a factor of 1.2–1.4 over tofu frying. During chicken boiling, CO concentrations increased 2.3–3.8 times over tofu boiling. These results indicate that frying produces more CO than boiling if we use low-fat foods. Replacing low-fat foods with high-fat foods results in even higher CO generation; the difference in CO concentration between the foods was more significant for boiling than for frying. Our average indoor CO concentrations were much lower than WHO indoor CO guidelines of 30.55 ppm for 1 h average and 8.73 ppm for 8 h average (WHO, 2010). Thus, this scheme of cooking is still safe for people, even in households using natural ventilation.

### CONCLUSIONS

Frying emits higher amounts of PM<sub>2.5</sub> than boiling in terms of mass and number concentrations. There was a time lag between changes in concentration in the kitchen

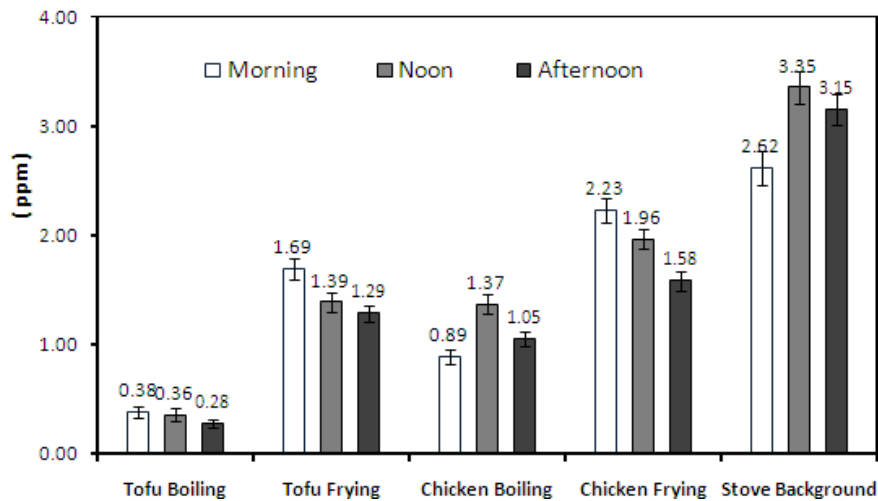


Fig. 7. Average CO Concentrations during Cooking Effects.

and in the adjoining room, and the particle size distribution in the adjoining room was lower and wider than that in the kitchen. With regard to the spatial distributions of fine particles in the kitchen and the adjoining room, frying exhibited less homogenous distribution of fine particles than boiling.

Frying resulted in high proportions of organic carbon in  $PM_{2.5}$ , while there was little difference in the ratio of elemental carbon between the cooking methods. Boiling produced lower emissions of CO in indoor air, possibly because of the absorption of the generated CO gas by steam.

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#### REFERENCES

- Afshari, A., Matson, U. and Ekberg, L.E. (2005). Characterization of Indoor Sources of Fine and Ultrafine Particles: A Study Conducted in a Full-scale Chamber. *Indoor Air* 15: 141–150.
- Buonanno, G., Morawska, L. and Stabile, L. (2009). Particle Emission Factors during Cooking Activities. *Atmos. Environ.* 43: 3235–3242.
- Cao, J.J., Lee, S.C., Chow, J.C., Cheng, Y., Ho, K.F., Fung, K., Liu, S.X. and Watson, J.G. (2005). Indoor/Outdoor Relationships for  $PM_{2.5}$  and Associated Carbonaceous Pollutants at Residential Homes in Hong Kong—Case Study. *Indoor Air* 15:197–204.
- Chao, C. Y., Wong, K.K. and Cheng, E.C. (2002). Size Distribution of Indoor Particulate Matter in 60 Homes in Hong Kong. *Indoor Built Environ.* 11: 18–26.
- Chowdhury, Z., Edwards, R.D., Johnson, M., Shields, K.N., Allen, T., Canuze, E. and Smith, K.R. (2007). An Inexpensive Light-scattering Particle Monitor: Field Validation. *J. Environ. Monit.* 9: 1099–1106.
- Dennekamp, M., Howarth, S., Dick, C.A.J, Cherrie, J.W., Donaldson, K. and Seaton, A. (2001). Ultrafine Particles and Nitrogen Oxides Generated by Gas and Electric Cooking. *Occup. Environ. Med.* 58: 511–516.
- Fan, C.W. and Zhang, J. (2001). Characterization of Emissions from Portable Household Combustion Devices: Particle Size Distributions, Emission Rates and Factors, and Potential Exposure. *Atmos. Environ.* 35: 1281–1290
- Fang, Y.D., Liu, J.H., Lin, L. and Liao, G.X. (2006). Full-scale Experimental Studies on Extinguishing Cooking Oil Fires with Water Mist. *Combust. Sci. Technol.* 12: 318–322
- He, Z., Kim, Y.J., Ogunjobi, K.O., Kim, J.E. and Ryu, S.Y. (2004). Carbonaceous Aerosol Characteristics of  $PM_{2.5}$  Particles in Northeastern Asia in Summer 2002. *Atmos. Environ.* 38: 1795–1800.
- Huboyo, H.S. and Tohno, S. (2010). Characteristic of Carbon Monoxide and Fine Particles in Indoor Air Related to Cooking Method, Poster in BAQ 2010, Singapore.
- Huboyo, H.S., Budihardjo, M.A. and Hardyanti, N. (2009). Black Carbon Concentration in Kitchens Using Fire-Wood and Kerosene Fuels. *J. Appl. Sci. Environ. Sanit.* 4: 55–62.
- Hussein, T., Glytsos, T., Ondracek, J., Dohanyosova, P., Zdimal, V., Hameri, K., Lazaridis, M., Smolik, J. and Kulmala, M. (2006). Particle Size Characterization and Emission Rates during Indoor Activities in a House. *Atmos. Environ.* 40: 4285–4307.
- IEA (2010). [http://www.ebook3000.com/Natural-Gas-Information-2010-with-2009-data\\_83365.html](http://www.ebook3000.com/Natural-Gas-Information-2010-with-2009-data_83365.html) accessed November 26, 2010.
- Khan, M.F., Shirasuna, Y., Hirano, K. and Masunaga, M. (2010). Characterization of  $PM_{2.5}$ ,  $PM_{2.5-10}$  and  $PM_{>10}$  in Ambient Air, Yokohama, Japan. *Atmos. Res.* 96: 159–172.

- Lee, S.C., Li, W.M. and Chan, L.Y. (2001). Indoor Air Quality at Restaurants with Different Styles of Cooking in Metropolitan Hong Kong. *Sci. Total Environ.* 279: 181–193.
- Li, C.S., Lin, W.H. and Jenq, F.T. (1993). Size Distributions of Submicrometer Aerosols from Cooking. *Environ. Int.* 19: 147–154.
- Massey, D., Masih, J., Kulshrestha, A., Habil, M. and Taneja, A. (2009). Indoor/Outdoor Relationship of Fine Particles less than 2.5  $\mu\text{m}$  ( $\text{PM}_{2.5}$ ) in Residential Homes Locations in Central Indian Region. *Build. Environ.* 44: 2037–2045
- Ministry of the Environment Government of Japan FY (2004). Status of Air Pollution, <http://www.env.go.jp/en/press/2005/0829c-10.pdf>, accessed February 4, 2011
- Nikas, K.S., Nikolopoulos, N. and Nikolopoulos, A. (2010). Numerical Study of a Naturally Cross-ventilated Building. *Energy Build.* 42: 422–434
- See, S.W. and Balasubramanian, R. (2006). Physical Characteristics of Ultrafine Particles Emitted from Different Gas Cooking Methods. *Aerosol Air Qual. Res.* 6: 82–92.
- See, S.W. and Balasubramanian, R. (2008). Chemical Characteristics of Fine Particles Emitted from Different Gas Cooking Methods. *Atmos. Environ.* 42: 8852–8862.
- Sjaastad, A.K., Svendsen, K. and Jorgensen, R.B. (2008). Sub-micrometer Particles: Their Level and How They Spread after Pan Frying of Beefsteak. *Indoor Built Environ.* 17: 230–236.
- Stavrakakis, G.M., Koukou, M.K., Vrachopoulos, M.Gr. and Markatos, N.C. (2008). Natural Cross-ventilation in Buildings: Building-scale Experiments, Numerical Simulation and Thermal Comfort Evaluation. *Energy Build.* 40: 1666–1681
- Tian, L., Zhang, G., Zhang, Q., Moschandreas, D.J., Hao, J., Lin, J. and Liu, Y. (2008). The Impact of Kitchen Activities on Indoor Pollutant Concentrations. *Indoor Built Environ.* 17: 377–383.
- Tsai, F.C., Smith, K.R., Vadakan, N.V., Ostro, B.D., Chesnut, L.G. and Kungskulniti, N. (2000). Indoor/Outdoor  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  in Bangkok Thailand. *J. Exposure Anal. Environ. Epidemiol.* 10: 15–26.
- Wallace, L.A., Emmerich, S.J. and Reed, C.H. (2004). Source Strengths of Ultrafine and Fine Particles Due to Cooking with a Gas Stove. *Environ. Sci. Technol.* 38: 2304–2311.
- Wallace, L.A., Fang, W., Reed, C.H. and Persily, A. (2008). Contribution of Gas and Electric Stoves to Residential Ultrafine Particle Concentrations between 2 nm and 64 nm: Size Distributions and Emission and Coagulation Rates. *Environ. Sci. Technol.* 42: 8641–8647.
- Weast, R.C. (1988). *Editor, (1st Student Edition ed.), CRC Handbook of Chemistry and Physics*, CRC Press, Inc., Boca Raton, Florida.
- WHO (2010). WHO Guidelines for Indoor Air Quality: Selected Pollutants. WHO Regional Office for Europe Scherfigsvej 8 DK-2100 Copenhagen Ø, Denmark.
- Yeung, L.L. and To, W.M. (2008). Size Distributions of the Aerosols Emitted from Commercial Cooking Processes. *Indoor Built Environ.* 17: 220–229.
- Zhao, X., Zhang, X., Xiaofeng, X., Xu, J., Meng, W. and Pu, W. (2009). Seasonal and Diurnal Variations of Ambient  $\text{PM}_{2.5}$  Concentration in Urban and Rural Environments in Beijing. *Atmos. Environ.* 43: 2893–2900.

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