

Ultrafine particles, and PM_{2.5} generated from cooking in homes

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ABSTRACT

Exposure to airborne particulate matters (PM) emitted during cooking can lead to adverse health effects. An understanding of the exposure to PM during cooking at home provides a foundation for the quantification of possible health risks. The concentrations of airborne particles covering the ultrafine (14.6–100 nm) and accumulation mode (100–661.2 nm) size ranges and PM_{2.5} (airborne particulate matters smaller than 2.5 µm in diameter) during and after cooking activities were measured in 12 naturally ventilated, non-smoking homes in Hong Kong, covering a total of 33 cooking episodes. The monitored homes all practiced Chinese-style cooking. Cooking elevated the average number concentrations of ultrafine particles (UFPs) and accumulation mode particles (AMPs) by 10 fold from the background level in the living room and by 20–40 fold in the kitchen. PM_{2.5} mass concentrations went up to the maximum average of about 160 µg m⁻³ in the kitchen and about 60 µg m⁻³ in the living room. Cooking emitted particles dispersed quickly from the kitchen to the living room indicating that the health impact is not limited to occupants in the kitchen. Particle number and mass concentrations remained elevated for 90 min in the kitchen and for 60 min in the living room after cooking. Particles in cooking emissions were mainly in the ultrafine size range in terms of the number count while AMPs contributed to at least 60% of the surface area concentrations in the kitchen and 73% in the living room. This suggests that AMPs could still be a major health concern since the particle surface area concentration is suggested to have a more direct relationship with inhalation toxicity than with number concentration. Particle number concentration (14.6–661.2 nm) in the living room was about 2.7 times that in the outdoor environment, suggesting that better ventilation could help reduce exposure.

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1. Introduction

Exposure to PM can induce adverse health effects, especially on the human respiratory system (Kennedy, 2007). PM can be inhaled into the human body, become deposited in the respiratory airway, and react with the organs. Some studies have suggested that exposure to PM can cause serious respiratory diseases, such as lung cancer (Ko et al., 2000; Moshammer et al., 2006; Hung et al., 2007). As modern people spend most of their time indoors, much research has been conducted to study the indoor coarse particles (PM₁₀, airborne particulate matters of ≤10 µm in aerodynamic diameter) and fine particles (PM_{2.5}). In the broad size spectrum of PM, UFPs, defined as particles with an aerodynamic diameter of less than 100 nm, have been shown to have the potential to induce more severe adverse health effects than coarser particles (WHO, 2000). Due to their small size, UFPs play a weak role in mass

concentration, but in many cases, dominate the number concentration and contribute significantly to the surface area concentration. Many previous studies have used mass concentration to assess the health effect of particle exposure, which may not be able to fully reflect the possible health impacts of UFPs.

Common indoor human activities including cooking, smoking, vacuuming cleaning and incense burning are major sources of indoor PM (e.g., Balasubramanian and Lee, 2007 and Wallace et al., 2008). Among those, gas cooking was found to be the most important source of UFPs in non-smoking homes in developed countries (See and Balasubramanian, 2006a). A number of laboratory-based (e.g., Dennekamp et al., 2001; Wallace et al., 2008) and site measurement (He et al., 2004a,b) studies have shown that cooking contributes greatly to the concentration of UFPs in residential houses. During cooking processes, a range of inorganic and organic compounds, including species that are identified possible carcinogens (See et al., 2006), are emitted. Chiang et al. (1999) indicated that exposure to fumes from cooking was an important health risk factor for women in Taiwan. However, the number of studies investigating UFP generation from Chinese-style cooking and the

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corresponding exposure in homes is still limited. See and Balasubramanian (2006a) conducted a study at a commercial Chinese food stall in Singapore. Their results revealed that about 80% of the particles was attributed to UFPs. See and Balasubramanian (2006b) further compared the UFP emission from different gas cooking methods and found that deep-frying generated the largest number of particles and contained the highest proportion of nanoparticles (about 90%). Zhang et al. (2010) indicated that that oil-based cooking (e.g., frying) emitted more UFPs and PM_{2.5} than water-based cooking (e.g., boiling). Chen et al. (2007) compared the cooking emissions from Chinese, western and western fast-food restaurants in Hong Kong and reported that particulate phase percentage contribution to total PAH for Chinese restaurants was the highest.

Hong Kong has a predominantly Chinese population of about 7 million people. Chinese-style cooking, which often involves a mix of cooking methods including frying, boiling, steaming, etc., is a common daily practice in most Hong Kong households. A site sampling campaign is currently being conducted in Hong Kong covering a wide variety of building types, and locations, smoking/non-smoking households, etc. This study aims at investigating the physical characteristics of UFPs, AMPs and PM_{2.5} in Hong Kong homes and to quantify the exposure of the occupants. Due to the complexity and the large number of influencing parameters in the whole sampling database, the sampling results obtained in 12 homes sharing some common features were selected from this on-going study for discussion in this paper. These homes are naturally ventilated, non-smoking homes with range hoods fitted in the kitchens. Natural ventilation and using range hood in the kitchen are some very popular features in residential apartments in Hong Kong. In these 12 homes a total of 33 cooking episodes (one cooking episode represents the cooking activities of preparing one meal for all the occupants in a home) were measured. Site measurements conducted in smoking homes, in homes with air-conditioning during cooking or in homes without range hoods fitted in kitchens are excluded in this paper.

2. Experimental method

2.1. Sampling sites

The 12 selected homes represent a mix of some of the most common types of residential buildings (2 were apartments in village houses, 10 were apartments in high-rise residential buildings) and unit sizes (floor areas of 40–150 m²) in Hong Kong. PM sampling was conducted between February and June, 2010. The sampling sites were located in Kowloon peninsula and the New

Territories. None of them was located in the outlying islands. All the sampled buildings were constructed out of reinforced concrete and ranged from 2 to 25 years old. The number of occupants living in these selected homes ranged from 2 to 6. All occupants were Hong Kong locals. The characteristics and locations of the sampling sites are summarized in Table 1. In all of these homes, gas stoves were used for cooking and range hoods were used for exhausting cooking fume. The range hoods were switched off after cooking. Unitary air conditioners were installed in all of these homes but they were not operated during the sampling period (1 h before and 3 h after cooking). Instead, these homes were under natural ventilation during the sampling period with windows opened. None of these homes had an open-kitchen configuration, i.e., the kitchens were in separate rooms. The kitchen doors were open during the sampling. No other combustion-related activity, e.g., incense or candle burning, was recorded at these sites during the sampling period.

2.2. Instrumentation and sampling protocol

To capture the exposure of occupants in their normal living, the occupants were told to prepare food in their usual way. No special instruction on the type of dish or method of cooking was given to the occupants but the dishes cooked and the time of cooking was recorded. Chinese-style cooking was practiced in all of these homes. Continuous, real-time monitoring of UFPs and AMPs number concentrations as well as PM_{2.5} mass concentration were conducted in the kitchen and the living room in each of the sampling sites. Custom-made mounting racks were fabricated to hold the particle instruments so that the sampling inlets were 1.7 m above the floor, roughly the breathing level of a standing adult. A scanning mobility particle sizer (SMPS, TSI Model 3936, USA) was used to monitor the number concentration and size distribution in the size range of 14.6–661.2 nm. PM in the size range of 14.6–100 nm are regarded as UFPs while those in the size range of 100–661.2 nm are regarded as AMPs in subsequent discussions. The SMPS consisted of an electrostatic classifier (EC, TSI Model 3080, USA) and a condensation particle counter (CPC, TSI Model 3776, USA). The sampling flow rate was set to 0.3 L min⁻¹. The duration of each SMPS sample scan was 135 s. The sampling inlet of the SMPS was connected to an auto-switching gas multiplexer for taking air samples from the two sampling points (the kitchen and the living room) alternately. The interval between each switch of sampling point was set to 10 min to allow enough time for the residual air sample in the tubing to be purged. Measurements were repeated in three of the 12 sites with the two sampling points being the living room and just outside the apartment to obtain the indoor–outdoor (I/O) ratio. The sampling inlet of the outdoor

Table 1
Summary of sampling site information.

Site	Floor area (m ²)	Floor	No. of occupants	No. of cooking episodes sampled	Duration of each cooking episode (min)				Horizontal distance to the nearest main road (m)	District of location	Building age (years)
1 ^b	56	25th	6	3 + 1 (I/O)	50	50	80	[50]	50	Tuen Mun	20
2	42	10th	4	2	30	50			200	Tseung Kwan O	20
3 ^{a,b}	80	G	2	3 + 1 (I/O)	20	20	20	[40]	300	Sai Kung	20
4 ^b	115	36th	3	3 + 1 (I/O)	50	10	10	[70]	100	Ma On Shan	10
5	40	22th	4	3	50	40	40		100	Tuen Mun	24
6	85	33th	3	2	40	30			10	Tsuen Wan	23
7	42	11th	2	3	30	20	20		10	Hung Hom	25
8	50	10th	3	3	20	50	50		20	Hung Hom	13
9	70	3rd	3	4	40	130	50	30	50	Hung Hom	3
10	70	22nd	3	1	40				30	Tseung Kwan O	2
11 ^a	80	G	3	2	20	20			400	Tai Po	20
12	150	5th	5	1	20				10	Tai Po	10

^a Apartments in village houses. Other sites were apartments in high-rise residential buildings.

^b I/O ratio measurement results available. The cooking time in [] is the cooking time for I/O ratio measurement.

sampling point was held at about 1 m away from the window of the living room and about 1.2 m above the floor of the apartment.

To account for the loss of particles in the sampling system due to deposition, a test according to the procedures of Morawska et al. (2001) was conducted. In the laboratory, which was found to have a stable particle concentration in the SMPS size range in a pre-test measurement, the SMPS was setup to sample air either directly from the laboratory or through the auto-switching sampling system alternately. The entire test lasted for 5 h and the average differences in particle concentrations measured with and without the auto-switching sampling system were used to calculate the mean percentage loss, which was 9.0% from the test. This was used subsequently to correct all readings from the SMPS.

The mass concentrations of $PM_{2.5}$ were measured at 1-min intervals by two optical-scattering monitors (DustTrak, TSI Model 8520, USA), one in the kitchen and the other in the living room. The readings of the DustTrak monitors were calibrated against a gravimetric sampler (MiniVol Portable Air Sampler, Airmetrics, USA) in a separate test conducted in the same laboratory where the above-mentioned SMPS particle loss test was conducted. In the calibration test, a Teflon filter was fitted into the MiniVol sampler which was set to sample at an airflow rate of 5 L min^{-1} . Sampling for the calibration test was repeated six times (lasting 38 h each time) and the results were averaged. The Teflon filters were stored in a dry box at $25 \pm 1^\circ\text{C}$ and $60 \pm 5\%$ relative humidity (corresponding to the conditions of the laboratory) for more than 1 month to equilibrate. The filters were weighed before and after the sampling using a high-precision electronic balance (Mettler-Toledo AT261, USA). The mean $PM_{2.5}$ mass concentration was determined as the ratio of mass difference to the airflow rate through the instrument during the whole sampling period. Two calibration functions ($0.24D_1$ with $R^2 = 0.87$ and $0.21D_2 - 0.38$ with $R^2 = 0.90$, where D_1 and D_2 were the readings from the two DustTraks in $\mu\text{g m}^{-3}$) were obtained by linear regression analysis of mean mass concentrations over the sampling period measured by the two DustTrak monitors and the MiniVol sampler. The calibration functions were used to adjust the readings from the DustTrak monitors in the subsequent on-site sampling. It should be noted that the optical-scattering method employed by the DustTrak monitors depends on the optical-scattering properties of the PM. Considering the fact that cooking emitted PM are highly

complex in nature and could vary significantly from one cooking episode to the other (caused by different food cooked and different cooking method used), it was highly possible that the above calibration procedure using the PM sampled in the laboratory did not fully calibrate for the range of optical properties of cooking emitted PM encountered in this study. The $PM_{2.5}$ results presented in this paper should be considered as approximations of the mass concentrations. The above calibration procedure was an effort to produce better approximations by the DustTrak monitors than using the factory calibrations.

Air change rate (ACR) was measured using the sulphur hexafluoride (SF_6) decay method reported by Chao and Wong (2002). A photo-acoustic multi-gas monitor (INNOVA 1312, Denmark) was used to measure the SF_6 level during the ACR measurements. The SF_6 measurement point was located at the center of the living room, 1.7 m above the floor.

2.3. Data analysis

Statistical analysis of the experimental data was performed using the data mining package SPSS (version 12). The normality of the data was checked using the Shapiro–Wilk test. Transformations (square root or log) were applied to some data to achieve normality before hypothesis tests were conducted.

3. Results

3.1. Particle concentrations

Fig. 1 shows the average SMPS total number and surface area concentrations of the 30 cooking episodes (not including the three for I/O measurements) in the 12 sampling sites. The (geometric) surface area concentrations were estimated based on the SMPS number concentration readings by assuming that the particles were spherical. Results shown in this figure are for the entire size range measured by the SMPS. The data at time 0 (the 1st '0' on the x-axis) is the background concentration obtained from 30 min of PM sampling before the start of cooking. Results for cooking were shown up to 50 min since most cooking episodes were 50 min or shorter (28 out of 30, see Table 1). The vertical dotted line denoted

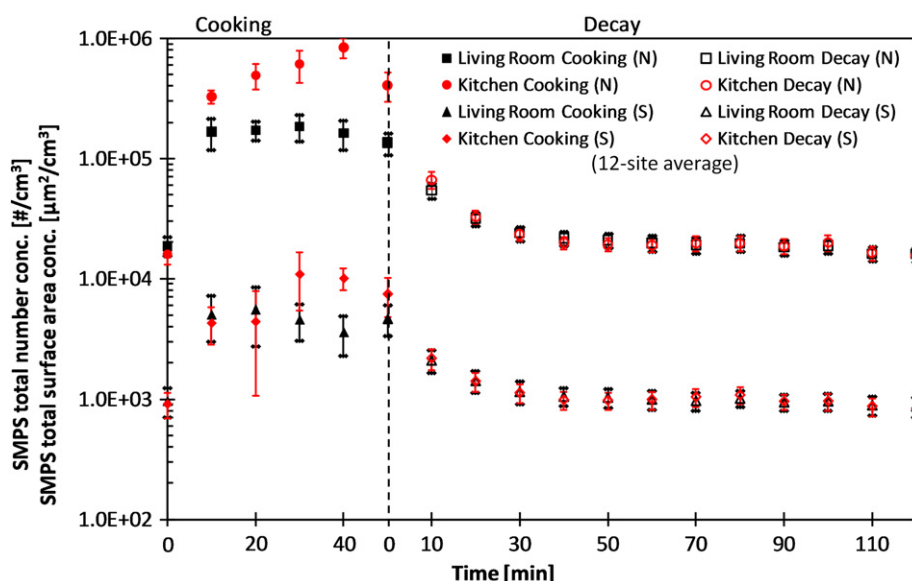


Fig. 1. Total number and surface area concentrations in the SMPS size range for 12 sampling sites. (N) – number concentration; (S) – surface area concentration.

by another '0' on the x-axis indicates the start of the decay period. Therefore, the first point of data for the decay period was taken 10 min after cooking was finished.

Fig. 1 shows that cooking led to an order of magnitude increase in the average number concentration from the background level in the living room. The average number concentrations in the kitchen during cooking went even higher resulting in an increase of about 20–40 fold the background level. The average number concentrations in these homes during cooking are in the same order of magnitude as measured in a Chinese food stall in Singapore ($7.7 \times 10^5 \text{ cm}^{-3}$, See and Balasubramanian, 2006a). He et al. (2004b) sampled in the kitchens in 14 Australian homes and reported a range of median peak number concentrations of 1.6×10^4 – $1.8 \times 10^5 \text{ cm}^{-3}$. Li et al. (1993) reported a maximum sub-micrometre aerosol number concentration of $2.6 \times 10^5 \text{ cm}^{-3}$ in a Taiwanese home. In the current study the median peak number concentration in kitchens was $6.3 \times 10^5 \text{ cm}^{-3}$ with a standard deviation of $1.7 \times 10^6 \text{ cm}^{-3}$. Comparing the peak number concentration in each kitchen of the current study with those reported by He et al. (2004b) and Li et al. (1993), the peak number concentrations in Hong Kong homes are significantly higher than the maximum median peak number concentration in Australian homes ($p = 0.014$) and that in Taiwanese home ($p = 0.019$) during cooking. Fig. 1 also shows that the particle number concentrations dropped quickly after cooking, suggesting that cooking was the major contributor to the high number concentrations in kitchens and living rooms during cooking. The number concentrations in kitchens remained significantly higher than the background level ($p < 0.05$) until 90 min after cooking. In living rooms, the number concentrations were significantly higher than the background level until 60 min ($p = 0.100$) after cooking.

The increase in surface area concentration due to cooking is noticeable but to a less extent compared to number concentrations. Average surface area concentrations in the living room during cooking were about 3.6–5.6 times the background level. In the kitchen, the average surface area concentrations were in the range of 5.4 – $14.0 \times 10^4 \mu\text{m}^2 \text{ cm}^{-3}$, about 5.4–14.0 times the background level. The smaller increase in surface area concentration compared to that in number concentration may suggest that the particles emitted from cooking were mainly in the small size end. This will be further analyzed in later sections of the paper. Buonanno et al. (2010) reported a range of maximum surface area concentrations of 6.8×10^2 – $1.9 \times 10^4 \mu\text{m}^2 \text{ cm}^{-3}$ during pizza making with wood-burning ovens in 15 pizzerias in central Italy. Picking the higher 12 surface area concentrations out of these 15 pizzerias and comparing them to the 12 homes in the current study using a two-sample *t*-test, the maximum average surface area concentrations in the 12 homes during cooking are significantly ($p = 0.020$) higher than those in the Italian pizzerias.

Fig. 2 shows the average $\text{PM}_{2.5}$ mass concentrations of the 30 cooking episodes (not including the three for I/O measurements). Again, cooking led to an increase in the average $\text{PM}_{2.5}$ concentration, as shown in Fig. 3, but the extent of increase was smaller compared to number and surface area concentrations. Average $\text{PM}_{2.5}$ concentrations increased from the background level of about $40 \mu\text{g m}^{-3}$ to about $60 \mu\text{g m}^{-3}$ in the living room (about 1.5 times the background level) and about $160 \mu\text{g m}^{-3}$ in the kitchen (about 4 times the background level) during cooking. The differences in the extent of increase between number and mass concentrations suggest that the particles generated from cooking were mainly UFPs. Another run of hypothesis test shows that the $\text{PM}_{2.5}$ mass concentration decayed to a level that was no longer significantly higher than the background level 90 min ($p = 0.059$) after cooking in the kitchen and 60 min ($p = 0.054$) after cooking in the living room.

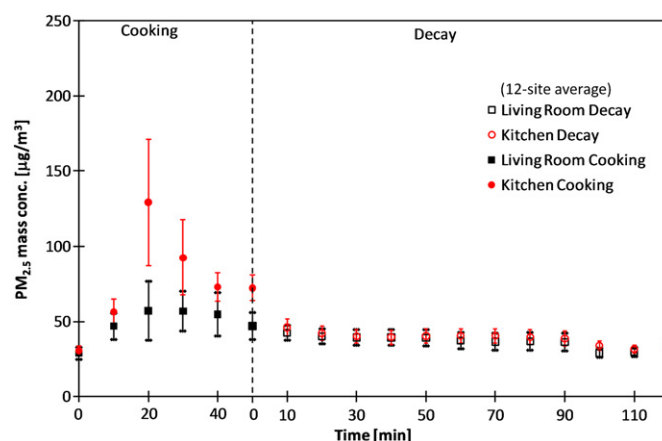


Fig. 2. $\text{PM}_{2.5}$ mass concentrations for 12 sampling sites.

3.2. Living room–kitchen (L/K) ratio

The exposure to cooking emitted particles in the kitchen is compared to that in the living room in Fig. 3, which shows the average ratios of particle concentrations (number, surface area and $\text{PM}_{2.5}$ mass) between the living room and the kitchen (or L/K ratio) of the 30 cooking episodes (not including the three for I/O measurements). The error bars show 1 standard error. Only the minus side of the error bars is shown for number concentrations and only the plus side is shown for surface area and $\text{PM}_{2.5}$ mass concentrations to minimize overlapping. Fig. 3 shows that the averages of the three types of particle concentrations (number, surface area, $\text{PM}_{2.5}$ mass) dropped once cooking had been started and remained at values below one during cooking. The L/K ratio for number concentrations dropped to the lowest of about 0.3 after 30 min of cooking and remained at similar levels for the next 20 min, meaning that exposure in terms of number concentrations in the kitchen was about 3 times that in the living room during this 50-min period. The average L/K ratio for surface area concentration was higher than that for number concentration during cooking. This could be due to the fact that the larger particles contributed a higher portion to the total number concentration in the living room than that in the kitchen.

The number concentrations are indicating that exposure in the living room is a small fraction of that in the kitchen during cooking, it may not be the case for surface area concentration. For instance,

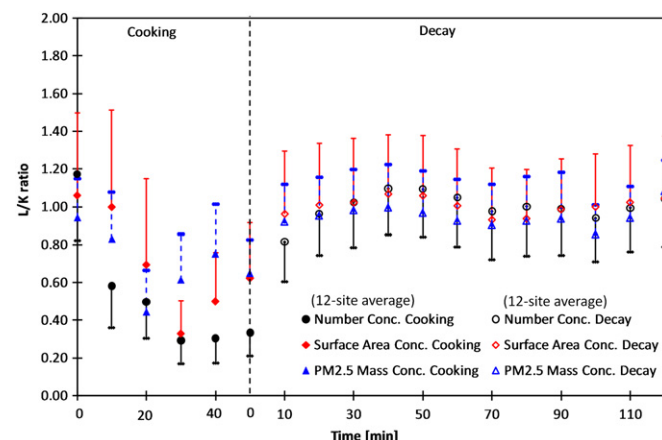


Fig. 3. L/K ratios for 12 sampling sites.

the average L/K ratio for number concentration was about 0.6, 10 min into cooking, meaning that the number concentration in the living room was about 60% of that in the kitchen on average. However, at the same time, the average L/K ratio for surface area concentration was about 1, i.e., surface area concentration in the living room was about the same as that in the kitchen. Using number concentrations to estimate the relative exposures at different parts of a building may not be able to truly reflect the possible health impact if the surface area concentration plays a role in the toxicity of the pollutant. The average L/K ratio for $PM_{2.5}$ mass concentration was lower than 1 during cooking, showing that exposure to $PM_{2.5}$ in living room was about 45–80% of that in the kitchen during cooking. The average L/K ratio for $PM_{2.5}$ mass concentration was mostly higher than the average L/K ratio for number concentration during cooking. The higher L/K ratio for mass concentration could also be due to the shift of the size spectrum towards the larger end due to coagulation.

The significance of the L/K results here is to give an indication on how the exposure by the occupants in the living room, who were not involved in the cooking activity, compares to the exposure by the person who did the cooking in the kitchen. The information could shed some new insight onto the potential health risk of cooking to occupants not only in the kitchen but in other parts of the apartment. L/K results could be affected by inhomogeneous spatial distribution of PM concentration (Lai and Chen, 2007). The variations in L/K under 12 different room geometries and settings were sampled and presented in this paper statically with the error range. Detailed investigation of the spatial distribution of PM concentration in each compartment involves complex fluid and aerosol dynamics, which is beyond the scope and the capability of the experimental setup and protocol of the current study. Measurements in the current study were all conducted with the kitchen door opened, which is usually the case as expressed by the owners. Closing the kitchen door may lead to substantial difference in the dynamics of L/K ratio and size distributions as different door positions are shown to have substantial impact on the transport of particles between rooms (Chuah et al., 2000). Further investigations on the spatial distribution of PM concentration and the closed door effect could provide a more comprehensive understanding on the potential health risk of occupants in other parts of the apartments.

3.3. Particle size distributions

The results presented in this section are based on SMPS measurements. The discussions in this section are limited to the size range of SMPS, i.e., 14.6–661.2 nm.

3.3.1. Number size distributions

Figs. 4 and 5 show the particle size distributions (number) in the kitchen and in the living room, respectively. The number concentrations shown are normalized, according to $dN/d\log d_p$, where N is the number concentration (cm^{-3}) and d_p is the particle size (nm), and are averaged from the 30 cooking episodes (not including the three for I/O measurements). The error bars show 1 standard error. Only the plus side of the error bars is shown to minimize overlapping. Fig. 4 shows that the majority of cooking emitted particles were UFPs and mainly in the smaller size end. During cooking, the normalized number concentrations of small particles (<50 nm) rose by about 2 orders of magnitude from the background level in the kitchen. The extent of this rise in normalized number concentrations became smaller for larger particles. Cooking led to a rise of about an order of magnitude for AMPs. UFPs contributed to more than 80% of the total number concentrations in the SMPS size range during cooking while the background level of UFP was only about

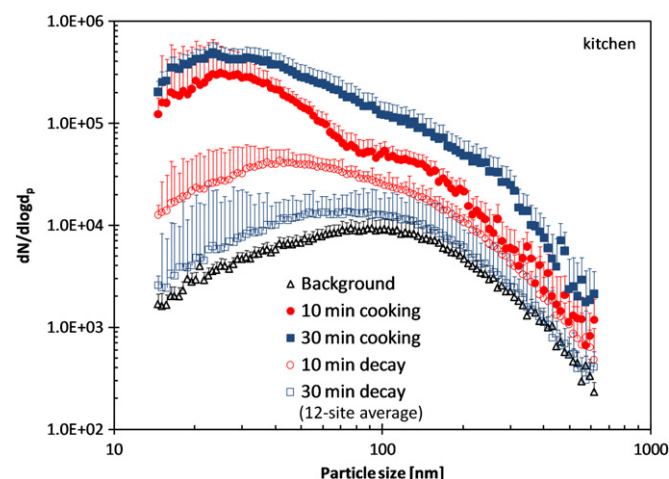


Fig. 4. Particle size distributions (in normalized number concentration) in the kitchens of 12 sampling sites.

60%. The mode size shifted from 73.7 nm in the background to 20.2 nm 10 min into cooking.

The rise of normalized number concentrations in the living room (Fig. 5) 10 min into cooking suggests that particles generated from cooking could disperse quickly from the kitchen to the living room. Similarly, in the living room, the normalized number concentrations rose by a greater extent in the UFP size range compared to the larger sizes. However, the size distributions in the living room were different from them in the kitchen during cooking. The mode in the living room shifted towards the larger sizes compared to the mode in the kitchen. The mode was at 37.2 nm in the living room 10 min into cooking and was at 55.2 nm 10 min into the decay period.

The dynamics of average particle number mean diameters (NMDs) are shown in Fig. 6. The NMDs shown in the figure are average values of the 30 cooking episodes (excluding the three for I/O measurements). The average NMD in the kitchen and the living room dropped rapidly from about 100 nm before cooking to about 65 nm and 75 nm, respectively, 10 min into cooking. The average NMD in the living room was about 10 nm larger than that in the kitchen throughout the cooking period. This suggests that more small size particles could be lost during the dispersion process from

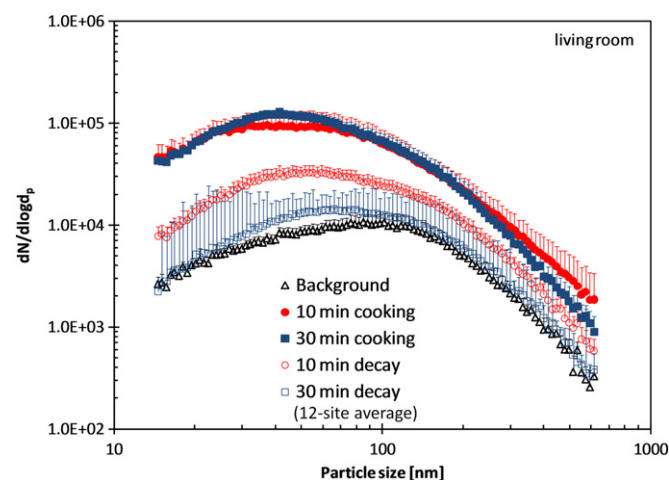


Fig. 5. Particle size distributions (in normalized number concentration) in the living rooms of 12 sampling sites.

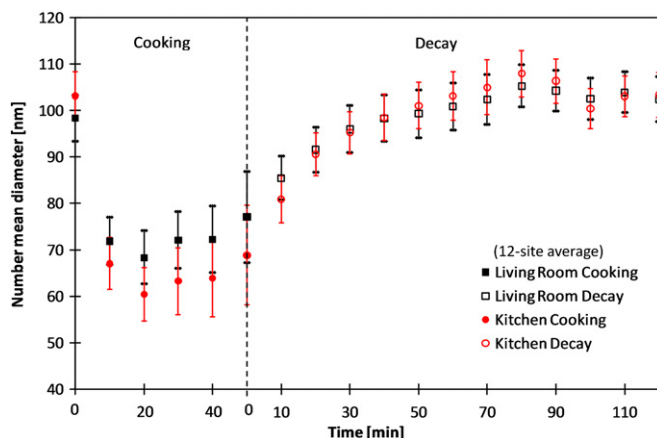


Fig. 6. Number mean diameters in the SMPS size range for 12 sampling sites.

the kitchen to the living room than large size particles. Coagulation of smaller particles to form larger particles during dispersion could be a major reason for the differences in NMDs between the kitchen and the living room. To investigate the impact of coagulation on this size spectrum shift, polydispersed coagulation rates were estimated using the average number size spectrum for kitchen at 10 min of cooking (which should give a conservative estimate since the average number concentration at 10 min was the lowest during cooking, see Fig. 1). The model of Wallace et al. (2008) was used with the assumptions of air temperature = 298 K, air viscosity = $1.81 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$, particle density = 1000 kg m^{-3} . The estimated coagulation rates for up to 100 nm are shown in Fig. 7. It shows that particles smaller than about 45 nm were lost while particles larger than about 45 nm were gained due to coagulation. The loss of smaller particles (<45 nm) could also be accelerated by deposition. Using the model of Lai and Nazaroff (2000) with a friction velocity of 0.1 m s^{-1} , and a surface-to-volume ratio of 3.0 m^{-1} , the deposition loss rate was found to be about 1 h^{-1} for 20 nm particles (in addition to the 4 h^{-1} coagulation loss rate for this size). ACR measured in the 12 sites averaged 1.8 h^{-1} (range $0.8\text{--}3.6 \text{ h}^{-1}$, SD = 0.8 h^{-1}), which was also relatively minor to the estimated coagulation loss rate for 20 nm particles. For 100 nm particles, the deposition loss rate decreases to about 0.1 h^{-1} , which is minor compared to the coagulation gain rate of about 4 h^{-1} for particles of this size. High coagulation rates (compared to deposition and air change rates) of cooking emitted

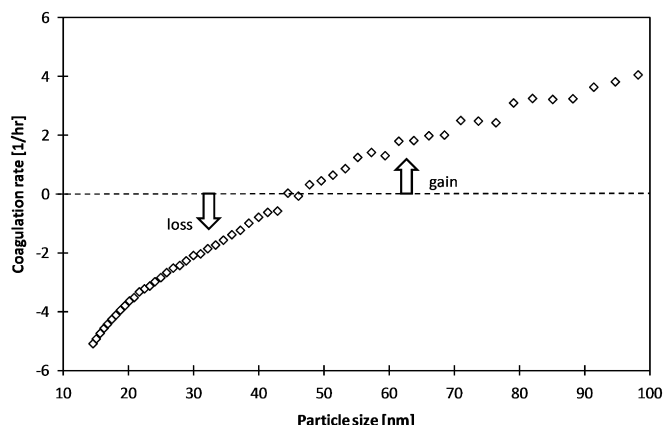


Fig. 7. Estimated polydispersed coagulation rates for 10 min of cooking.

PM were also reported by Wallace et al. (2008). They estimated a range of $+13 \text{ h}^{-1}$ (loss) to -7 h^{-1} (gain) for the size range of 2–64 nm. The NMDs in both the kitchen and the living room kept increasing after cooking had stopped until they were once again around 100 nm.

3.3.2. Surface area size distributions

The contributions from particles of different sizes to the total surface area concentration are depicted in Figs. 8 and 9. The two figures show the average particle size distributions (surface area concentration) of the 12 sampling sites measured by the SMPS in the kitchen and in the living room. The surface area concentrations shown were normalized, $dS/d\log d_p$, where S is the surface area concentration ($\mu\text{m}^2 \text{ cm}^{-3}$). Figs. 8 and 9 reveal that the major contributors to the surface area concentration are very different from those to the number concentration. Despite the fact that UFPs (especially those <50 nm in size) were the major contributors to total number concentrations, AMPs were the major contributors to total surface area concentrations. This could be because surface area is a function of d_p^2 . In the background, only around 12% of the total surface area concentrations were attributed to UFPs. Despite the large number of UFPs emitted during cooking, contributions by UFPs to total surface area concentrations were only about 39% in the kitchen and 27% in the living room at maximum. This suggests that particles larger than 100 nm in size were the major contributors to surface area concentrations, even during cooking.

Research increasingly suggests that UFPs are more toxic than their larger counterparts because of their large surface areas (due to their large numbers) for condensation or adsorption of toxic substances. This claim could be valid when the UFPs are compared with larger particles under equal mass, i.e., a unit mass of UFPs has a larger total surface area than a unit mass of larger particles. This could be the reason for the higher responses resulting from UFP exposure than from AMP exposure in some toxicology studies (e.g., Fig. 3 in Brown et al., 2001 and Fig. 3a in Oberdörster, 2001) on a mass-based dose–response relationship. However, when a surface area-based dose–response relationship is used, responses due to UFP exposure and AMP exposure fall on the same dose–response curve (Fig. 4 in Brown et al., 2001 and Fig. 3b in Oberdörster, 2001). This suggests that exposure to particle surface area is an important factor that could affect toxicity while the difference in particle size could play a relatively minor role. The

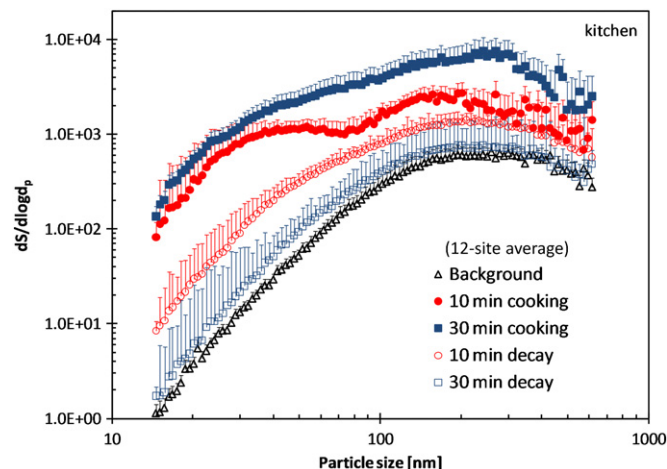


Fig. 8. Particle size distributions (in normalized surface area concentration) in the kitchens of 12 sampling sites.

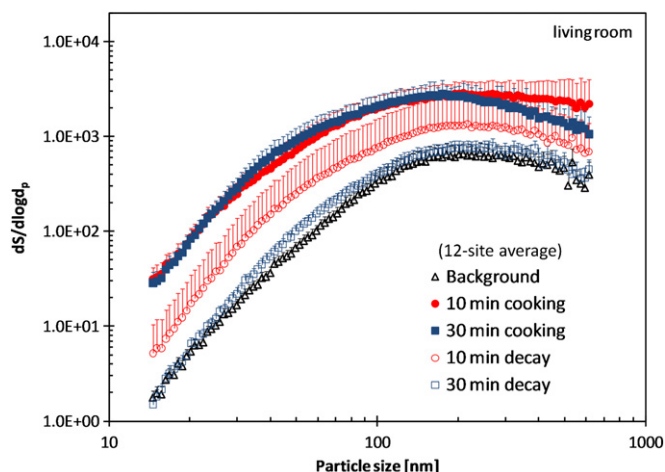


Fig. 9. Particle size distributions (in normalized surface area concentration) in the living rooms of 12 sampling sites.

results in Figs. 8 and 9 show that, in the sub-micron range, AMPs were still the major contributors to total surface area concentration during and after cooking. Contributions to total surface area concentration by UFPs were modest but certainly not negligible (from 12% in the background to a maximum of 40% during cooking in the kitchen). However, this by no means implies that the toxicity from the UFP portion is less than that from the AMP portion in cooking scenarios based on their relative contributions to surface area concentration alone. There are other parameters, such as the particle's ability to penetrate interstitial sites or blood streams, lung retention, chemical compositions (also affected if the particles are generated from food heating or combustion), that could affect the toxicity.

3.4. I/O ratio

Fig. 10 shows the average I/O ratios (in terms of total number concentration measured by the SMPS) for the three extra cooking episodes. The minus side of the error bars is shown and the error bars indicate 1 standard error. The I/O ratio during cooking was significantly ($p < 0.001$) higher than 1, indicating that the number concentration in the living room was highly influenced by the cooking emission during cooking. The average I/O ratio during

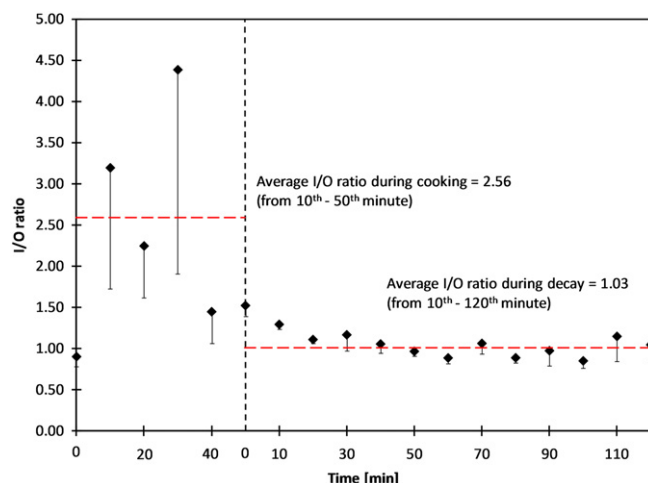


Fig. 10. Average I/O ratios for three sampling sites.

cooking was 2.56. This suggests that improving the ventilation in the apartments could help reduce the particle exposure in the living room during cooking. However, the ACR was not the only factor that affected I/O ratio. The ACR measured in the 3 sites were Site 1 = 2.0 h^{-1} , Site 3 = 2.6 h^{-1} and Site 4 = 1.5 h^{-1} but the average I/O ratios during cooking were Site 1 = 3.71, Site 3 = 1.23 and Site 4 = 3.01. The highest I/O ratio during cooking was in Site 1 but its ACR was not the lowest among the 3 sites. The outdoor concentration, PM emission rate and spatial variations in air exchange between indoor and outdoor air caused by variations in pressure differentials and wind direction (Liao et al., 2003) could also affect the I/O ratio. The influence of cooking emission on number concentration in the living room is further evidenced by the trend that the I/O ratio quickly resumed to nearly 1 after cooking was finished. The average I/O ratio during the decay period was 1.01 which might suggest that particle generated (in the SMPS size range) from indoor activities other than cooking was minor during the measurement period. It should be noted that I/O ratio measurements were made only in 3 out of the 12 homes. Considering that the I/O ratio could be affected by many factors, the I/O ratio results may not be fully representative of all the homes in the current study. The I/O ratio results could only give some indications on the extent of indoor PM concentration elevation due to cooking compared to outdoor concentration.

4. Conclusion

Cooking emissions led to significant increases in particle concentrations both in the kitchen and in the living room. During cooking the average number concentrations of UFPs and AMPs in the kitchen were about 20–40 times the background level and were about 10 times the background level in the living room. The average surface area concentrations were 5.4–14 times the background level in the kitchen and were about 3.6–5.6 times of background level in the living room. For $\text{PM}_{2.5}$ mass concentrations, the maximum average was about 4 times the background level in the kitchen (about) and about 1.5 times the background level in the living room during cooking.

PM in cooking emissions was mainly in the UFP ($< 100 \text{ nm}$) size range, especially the lower end of that size range. UFPs contributed to more than 80% of the total number concentrations during cooking while the portion of UFPs in the background was only about 60%. However, in terms of surface area concentrations, AMPs were the major contributors. During cooking, AMPs still contributed to a minimum of 60% of the surface area concentration in the kitchen and 73% in the living room.

Particles emitted from cooking could disperse quickly to the living room. Coagulation took place during the dispersion process. The average number mean diameter of UFPs and AMPs in the living room was about 10 nm larger than that in the kitchen during cooking. Particle number concentration remained elevated until 90 min after cooking in the kitchen and until 60 min after cooking in the living room.

This study reveals the exposure to and size characteristics of UFPs, AMPs and $\text{PM}_{2.5}$ during uncontrolled cooking in Hong Kong homes (occupants cooked in their usual ways). A mix of cooking methods, ingredients, and amounts of food were seen during the sampled cooking episodes. Analysis of detailed correlations between cooking methods, food, oil and PM exposure using the current results would be difficult and subjected to many uncertainties. The current sampling protocol was designed to study the PM concentrations under normal daily cooking situations. More in-depth investigations on the impacts of cooking methods and cooking oil on PM exposures using controlled cooking sampling protocol is covered in another parallel paper (Wu et al., 2011).

Comments on the potential health risk associated with the exposure to cooking emitted PM in this paper are made based on the number/surface area/mass concentrations only. However, to get a more comprehensive understanding of the health impact, further chemical analysis would be necessary to reveal the toxicity of the PM. The impacts of operating the air-conditioning, switching off the range hood, opening or closing the kitchen door were also not investigated. An understanding of their impacts would be useful for recommending proper ventilation methods in home kitchens.

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