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RESEARCH ARTICLE

Pollution characteristics of particulate matters emitted from outdoor barbecue cooking in urban Jinan in eastern China

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HIGHLIGHTS

- Extremely high levels of particles were emitted from outdoor barbecue cooking.
- Barbecue particle numbers exhibited very large increase in super-micro particles.
- Barbecue cooking had influence on particle concentrations in surrounding areas.
- Range hood efficiently removed cooking particles and thus was recommended.

GRAPHIC ABSTRACT

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ABSTRACT

To understand the pollution characteristics of particulate matter emitted from outdoor barbecue cooking in eastern China, measurements of the $PM_{2.5}$ mass concentration, the number concentration of particles with a diameter of 0.01 to 1.0 μm, and the particle size distribution from 0.3 to 25 μm were carried out at seven barbecue restaurants in urban Jinan. The average $PM_{2.5}$ mass concentration and sub-micron particle number concentrations at a distance of 1 m from the grills were 250 to 1083 μg/m^3 and 0.90×10^5 to 2.23×10^5 cm^{-3}, respectively, which were much higher than those in the ambient air of the urban area. Compared to the ambient atmosphere, barbecue cooking emitted very high levels of particles with a larger increase in the concentrations of super-micron particles than that of sub-micron particles. The super-micron particle number concentrations at the barbecue restaurants were 10 to 100 times higher than those observed in the ambient urban atmosphere. The barbecue smoke had a significant effect on the particle concentrations in the surrounding region. Both mass and number concentrations of particles exhibited maximum values immediately near the barbecue grills and often reached a peak at a distance of 10 to 15 m. The removal efficiency of a range hood for the cooking particles was tested in an indoor kitchen. The range hood effectively cleaned the particulate matter pollution caused by cooking with a removal efficiency larger than 80%. Therefore, the use of a range hood is recommended for outdoor barbecue restaurants coupled with a smoke purifier to clean the emitted high concentrations of particles.

1 Introduction

Cooking smoke, a type of biomass burning smoke or its mixture with combustion smoke of fossil fuels, is an important emission source of atmospheric particulate matter and has a significant influence on air quality [1,2]. Household and restaurant cooking has been confirmed as one of the major anthropogenic activities responsible for high concentrations of particles indoors [3–9]. Previous studies have shown diverse emission characteristics of particles generated from different styles of cooking [10–

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13]. He et al. [14] found that the indoor $PM_{2.5}$ concentrations during grilling and frying processes in residences in Brisbane, Australia, reached 718 μg/m^3 and 735 μg/m^3, respectively, which represented increases of 30 and 90 times over ambient concentrations. In addition, the concentration of particulate matter emitted from Asian cooking was generally higher than that from Western cooking [14–19]. In particular, compared to other Chinese cooking styles, barbecue cooking emitted the highest levels of fine particulate matter with a maximum concentration of 1842 μg/m^3, which was dozens of times higher than the ambient $PM_{2.5}$ concentration [20]. Due to the significant contribution of cooking smoke to the indoor particle concentration, cleaning devices have been recommended for in-household kitchens and restaurants in recent years [21,22].

Only a few studies have been conducted to investigate the characteristics and influence of outdoor cooking smoke on air quality. In general, outdoor barbecue cooking can have a more significant influence on ambient air quality than indoor cooking due to the uncontrolled emissions and subsequent diffusion and transport [23–25]. Zhao and Lin [26] measured the air quality at four night markets where barbecue accounted for approximately 25% of the outdoor restaurants in Kaohsiung, Taiwan, China, from August to November 2008. They found that the hourly average $PM_{2.5}$ and PM_{10} concentrations ranged from 175 to 418 μg/m^3 and from 186 to 451 μg/m^3, respectively, and that the emitted particles contained high levels of polycyclic aromatic hydrocarbons [27]. Similarly, in Hefei in eastern China, the PM_{10} concentrations near barbecue grills were at levels of 168 to 426 μg/m^3, which were much higher than the surrounding areas (approximately 55 μg/m^3) [28]. To determine the chemical compositions of gaseous and particulate pollutants from barbecue cooking, combustion experiments were carried out in a combustor or kitchen [29,30]. Kabir et al. [31] measured the volatile organic compounds (VOCs) and carbonyl compounds emitted from barbecue cooking in East Asia and showed that toluene was the most abundant VOC and formaldehyde and acetaldehyde were the most prominent carbonyls. In addition, trace metals in the PM_{10} of barbecue smoke were at the level of several to 10^5 ng/m^3, with the highest levels found for Zn, Pb, and Mg [32]. Despite these efforts, there is still a lack of in situ measurements of particles emitted from normal outdoor barbecue cooking, which are essential to obtain a comprehensive understanding of the pollution characteristics.

Barbecue is very popular in China, and there are a large number of outdoor barbecue restaurants in urban areas, especially during summer. Thus far, the outdoor barbecue smoke has not been effectively controlled, which could potentially influence the local air quality. To understand the pollution characteristics of particulate matters from Chinese barbecue cooking, $PM_{2.5}$ mass and particle number concentrations of different size ranges were measured in situ at seven barbecue restaurants in different districts of Jinan in eastern China. We compared the $PM_{2.5}$ mass concentration, sub-micron particle number concentration, and size distribution for particles with a diameter of 0.3 to 25 μm at different distances from the grills of seven outdoor barbecue restaurants, with those in the ambient urban atmosphere to investigate the influence of outdoor barbecue cooking on the surrounding air quality. The removal efficiency of a range hood on cooking particles was also tested in an indoor kitchen.

2 Experimental methods

2.1 Sampling locations

Jinan is a populated city in eastern China with an urban population of 4.85 million in 2015 [33]. There are approximately 1500 barbecue restaurants distributed along the roadsides in urban Jinan (http://www.dzwww.com/shandong/sdnews/201605/t20160513_14283400.htm). Most of the barbecue restaurants are open from dusk to midnight, and some are even operated until early morning. In this study, seven moderately large-scale barbecue restaurants in five locations were selected for sampling and in situ measurements. They were distributed in different districts of urban Jinan. The locations of the sampling sites are shown in Fig. 1 and are marked as A, B, C, D, and E. Three barbecue restaurants (A1, A2, and A3) resided within location A and were very close to each other. The sampling sites were selected along minor roads with relatively little vehicle traffic in the evening (traffic volume was approximately 600–900 vehicles per hour) and that were more than 4 m from the motorway. Therefore, the traffic emissions had little influence on the

Fig. 1 Locations of the sampling sites in urban Jinan

measurement data. For comparison, an urban site in the Central Campus of Shandong University (SDU, approximately 20 m above the ground) was also used to examine the concentrations and characteristics in the ambient urban atmosphere.

2.2 Instruments and measurements

The $PM_{2.5}$ mass concentration was measured using a portable aerosol monitor (SidePak AM510, TSI, USA). The number concentration of sub-micron particles between 0.01 and 1 μm ($PM_{0.01–1}$) was monitored using a condensation particle counter (Model 3007, TSI, USA). The size distribution of the number concentration of particles between 0.3 and 25 μm ($PM_{0.3–25}$) was determined using an optical particle counter (Model 9306, TSI, USA). With the optical particle counter, the particle sizes were divided into six bins: 0.3 to 0.5, 0.5 to 1.0, 1.0 to 3.0, 3.0 to 5.0, 5.0 to 10.0, and 10.0 to 25.0 μm. The temperature and relative humidity were measured using a temperature and humidity sensor (HH314A, Omega, USA). The measurement data were automatically stored as averages in the instruments at an interval of 10 s.

The $PM_{2.5}$ mass concentration, particle number concentrations, temperature, and humidity were measured in real time at the barbecue restaurants and the SDU site. The portable instruments were placed 1.5 m above the ground. Table 1 shows the detailed information of the in situ measurements. The measurements usually started at dusk (18:00 to 19:00) and ended in mid- or late evening (after 20:00) with a duration of 1 to 4 h, a period during which the barbecue restaurants had the most customers and the cooking smoke was the most intense. During the sampling periods, the wind speeds were relatively low (generally below 3 m/s), and the wind direction was scattered due to the presence of high buildings close to the sampling sites. To assess the influence of the distance of the outdoor cooking smoke, the measurements were conducted one after another on the downwind side at distances of 1, 5, 10, 15, 25, and 35 m from the barbecue grills.

2.3 Test on the removal of cooking particles

To determine the effect of a range hood on the removal of cooking particles within a certain space, the $PM_{2.5}$ mass and particle number concentrations were measured in a kitchen. The same portable aerosol monitor and particle counters were deployed in the test, and the sample inlets were installed 1.5 m above the ground. The total area of the kitchen was 6 m^2, with a height of 2.8 m. The extraction rate of the range hood was 15 m^3/min.

At the beginning of the test, the doors and the window of the kitchen were completely closed. The particle mass and number concentrations were measured with the range hood inactive during cooking for 55 min (Period 1). After cessation of cooking, the particle mass and number concentrations were measured for an additional 50 min (Period 2). Finally, the range hood was activated and run at a constant extraction rate of 15 m^3/min with the door slightly open, and the changes of particle mass and number concentrations were measured for approximately 30 min (Period 3). The removal efficiency of the range hood during Period 3 was calculated based on the average particle concentration in the last minute of Periods 2 and 3.

3 Results and discussion

3.1 Mass and number concentrations of fine particles generated by barbecue cooking

Table 2 lists the $PM_{2.5}$ mass and sub-micron particle number concentrations, temperature, and relative humidity observed at different outdoor barbecue restaurants (1 m from the grills with a sampling duration of 0.5 h) and the SDU site in urban Jinan. During the measurement periods, the temperature was comparable to the ambient temperature at SDU site, but the relative humidity was a little higher than the ambient relative humidity. This discrepancy was caused by the release of water vapor during the burning process and the difference in the sampling times.

Table 1 Detailed information for the in situ measurements of particle mass and number concentrations at barbecue restaurants. The sampling sites included seven barbecue restaurants (A1-E) and one urban site (SDU). We measured $PM_{2.5}$ mass concentration and particle number concentration of $PM_{0.01–1}$ and $PM_{0.3–25}$. $PM_{0.3–25}$ was divided into six parts based on the size ranges

Site	Date	Time	Type	Parameters	Distances
A1	7/20/2015	19:00–22:00	Barbecue	T, RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	1 m, 5 m, 10 m
A2	7/21/2015	19:00–21:00	Barbecue	T, RH, $PM_{2.5}$, $PM_{0.3–25}$	5 m, 15 m, 25 m, 35 m
A3	7/22/2015	18:10–19:10	Barbecue	T, RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	1 m, 10 m
B	7/23/2015	19:00–20:30	Barbecue	T, RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	1 m, 5 m, 10 m
C	7/24/2015	19:10–20:40	Barbecue	T, RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	1 m, 10 m
D	7/27/2015	19:20–21:35	Barbecue	T,RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	1 m, 5 m, 10 m, 15 m
E	7/28/2015	18:20–20:25	Barbecue	T, RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	1 m, 5 m
SDU	7/31/2015	16:30–21:30	Urban site	T, RH, $PM_{2.5}$, $PM_{0.01–1}$, $PM_{0.3–25}$	

Table 2 Summary statistics of $PM_{2.5}$ mass and particle number concentrations at a 1-m distance from grills of seven barbecue restaurants and one urban site. Daily $PM_{2.5}$ concentration stands for the daily average $PM_{2.5}$ mass concentrations in Jinan city, which were published on the website of the Ministry of Environmental Protection of China

Site	T(°C)	RH(%)	Distance	$PM_{2.5}$(μg/m^3)				$PM_{0.01-1}$($\times 10^5$/cm^3)		
				Mean±SD	Min	Max	Daily	Mean±SD	Min	Max
A1	29.8	64.6	1 m	250±64	139	531	66	1.07±0.31	0.38	2.93
A3	29.8	79.9	1 m	1083±1124	126	6100	93			
B	29.4	71.6	1 m	415±311	102	1761	69	1.06±0.56	0.25	3.25
C	33.2	54.0	1 m					2.23±1.08	0.22	4.34
D	35.0	60.2	1 m	442±450	101	3026	65	1.74±1.08	2.57	4.28
E	33.5	61.4	1 m	331±807	44	7564	44	0.90±0.52	0.22	3.62
SDU	31.9	57.6	1 m	71±6	52	91	35	0.10±0.04	0.01	0.26

Both $PM_{2.5}$ mass and sub-micron particle number concentrations at the barbecue restaurants exhibited great variation and were much higher than those in the ambient urban atmosphere. The highest half-hour average $PM_{2.5}$ concentration, 1083 μg/m^3, was observed at the restaurant in A3, which was nearly 12 times higher than the daily average $PM_{2.5}$ concentration in urban Jinan. In contrast, the restaurant located in A1 had the lowest average $PM_{2.5}$ concentration (250 μg/m^3), which was 3.8 times higher than the daily average concentration. The sub-micron particle number concentration ranged from 0.90×10^5 to 2.23×10^5 cm^{-3} (i.e., particles/cm^3), which was 9.1 to 22.5 times higher than the average concentration at the SDU site (0.10×10^5 cm^{-3}). The large variation in particle concentrations among the seven barbecue restaurants was mainly attributed to the differences in charcoal fuel, food, cooking condition, surrounding buildings, and the wind direction and speed. The extremely high levels of $PM_{2.5}$ at the restaurant in location A3 were caused by the intensive barbecue cooking. Significantly higher particle concentrations at the barbecue restaurants than in the ambient urban atmosphere indicate the severe particulate matter pollution generated during barbecue cooking.

We further compared the frequency distributions of $PM_{2.5}$ mass and sub-micron number concentrations at the barbecue restaurants with those observed in the ambient atmosphere at the SDU site (see Fig. 2). As shown, the ambient $PM_{2.5}$ concentration presented one single peak at approximately 70 μg/m^3. However, the $PM_{2.5}$ concentration at the barbecue restaurants produced two peaks at 50 to 75 and 225 to 250 μg/m^3. Similar to the $PM_{2.5}$ mass concentration, the sub-micron particle number concentration at the barbecue restaurants was also bimodal, with two peaks at 0.50 and 0.90×10^5 cm^{-3} compared to a sole peak at 0.10×10^5 cm^{-3} in the ambient urban atmosphere. The additional second peak and much higher $PM_{2.5}$ mass and number concentrations at the barbecue restaurants indicate a significant influence of barbecue cooking on the fine particle concentrations in the surrounding air.

3.2 Size distribution of particles generated by barbecue cooking

To understand the size distribution of particles emitted from outdoor barbecue cooking, the number concentrations of particles in six size ranges from 0.3 to 25 μm were measured at a 1-m distance from grills of the six barbecue restaurants and the SDU site. As shown in Fig. 3(a), the overall size distribution of the particles at the barbecue restaurants was similar to that in the ambient urban atmosphere. In general, the particle number concentration between 0.3 and 25 μm decreased with rising particle size. However, apparent differences existed in the number concentrations within each size range. The particle number concentrations at the barbecue restaurants were all much higher than those in the ambient urban atmosphere. The average number concentrations of $PM_{0.3-0.5}$, $PM_{0.5-1}$, PM_{1-3}, PM_{3-5}, PM_{5-10}, and PM_{10-25} were 301, 121, 27, 0.78, 0.34, and 0.01 cm^{-3} at the six barbecue restaurants, respectively, and 135, 23, 0.77, 0.02, 0.01, and 0.001 cm^{-3} at the SDU urban site, respectively.

To clarify the distinct characteristics of the particles emitted from barbecue cooking, we calculated the ratios of the particle number concentration within six size ranges at the barbecue restaurants to those at the SDU urban site (see Fig. 3(b)). As shown, the ratios of sub-micron particles were all below 10 with a range of 1 to 8. Nevertheless, the ratios of super-micron particles were mostly above 10, and some ratios of the particles with the size range of 1 to 10 μm were close to 100, with a maximum of 135. These results suggest that barbecue cooking emitted a large number of particles and contributed more to the number of super-micron particles than to the number of sub-micron particles in the ambient urban atmosphere, which was primarily attributed to intensive emissions of fly ash and smoke dust from the complete and incomplete combustion of charcoals and meats. It should be noted that the relatively high altitude at the SDU site (20 m above the ground) might have influenced the difference in particle

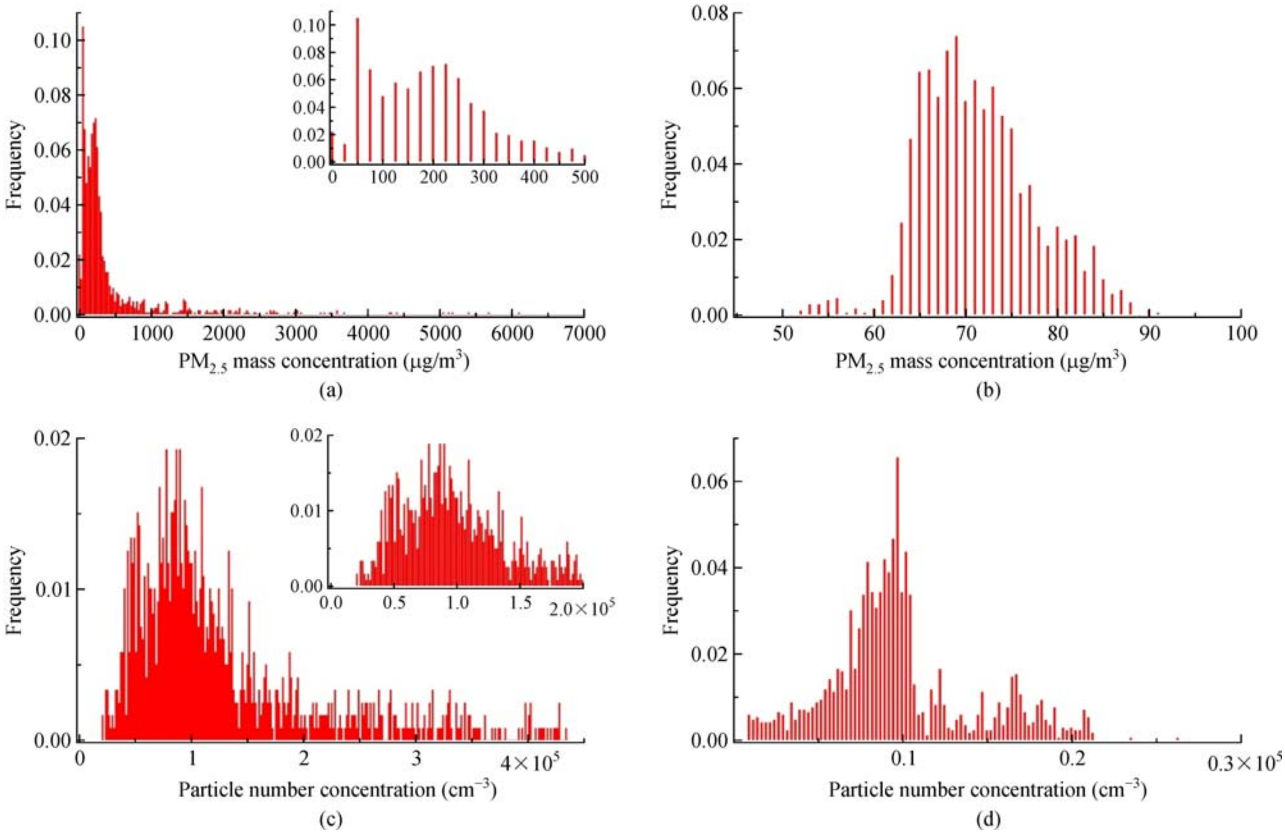

Fig. 2 Distribution of $PM_{2.5}$ mass concentrations at the barbecue restaurants (a) and the SDU site (b), and sub-micron particle number concentration at the barbecue restaurants (c) and the SDU site (d). The insets show the details within a small concentration range

pollution characteristics at the barbecue restaurants and the ambient urban atmosphere.

3.3 Influence of barbecue cooking on surrounding air quality

To determine the influence of barbecue cooking on the air quality of the surrounding areas, $PM_{2.5}$ mass and sub-micron particle number concentrations were measured at different distances (e.g., 1, 5, 10, 15, 25, and 35 m) from the barbecue grills at four restaurants (i.e., A1, A2, B, and D). The average $PM_{2.5}$ mass and sub-micron particle number concentrations at each distance for each restaurant are listed in Table 3. In general, both $PM_{2.5}$ mass and sub-micron particle number concentrations showed maximum values at 1 m. At restaurants A1, A2, and D, the $PM_{2.5}$ concentration slightly increased with distance from 5 m to 10 or 15 m and then gradually decreased with distance after a peak at 10 to 15 m. The sub-micron particle number concentration also exhibited a high value at 15 m at restaurant D. These results highlight the significant influence of barbecue cooking on the air quality in the surrounding region and suggest a possible smoke transport mechanism of uplift followed by dispersion with the wind, which declines on the downwind side.

3.4 Removal of cooking particles by range hood

The in situ measurements of the $PM_{2.5}$ mass and sub-micron particle number concentrations and the particle size distribution at the outdoor barbecue restaurants in urban Jinan demonstrate the severe particulate matter pollution in the surrounding region and its large influence on local air quality. A commonly used method to mitigate the air particulate matter pollution caused by barbecue cooking is to move the grills indoors and remove the cooking smoke with a range hood. An indoor test of the removal of cooking particles was conducted in this study, and the variations in the $PM_{2.5}$ concentration, the particle number concentration within different size ranges, and the temperature and relative humidity are shown in Fig. 4.

During Period 1, the temperature and relative humidity increased gradually once the cooking was started and a large number of particles were generated within the kitchen. The number concentration of sub-micron particles and super-micron particles rose rapidly with the concurrent dramatic increase in $PM_{2.5}$ mass concentration (maximum above 10 mg/m^3), indicating the accumulation of cooking particles in the closed kitchen. The number concentration of accumulation mode particles with sizes ranging from 0.3 to 1 μm exhibited a decrease during the period of 18:40 to

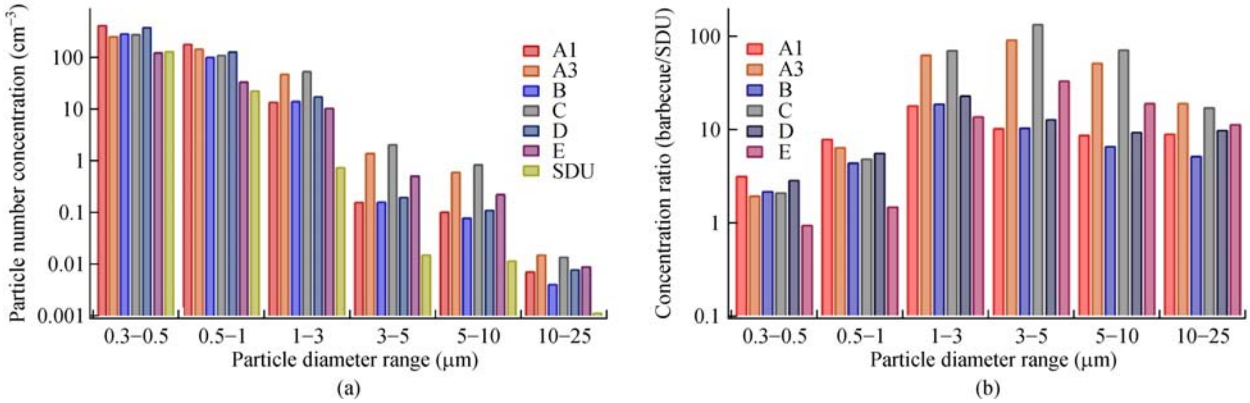

Fig. 3 Comparison of particle number concentrations in six size ranges at the barbecue restaurants and SDU site (a) and the ratios of particle number concentrations at the barbecue restaurants to those at the SDU site (b)

Table 3 $PM_{2.5}$ mass and sub-micron particle number concentrations at different distances from the grills of four barbecue restaurants

Site	Parameter	Distance(m)					
		1	5	10	15	25	35
A1	$PM_{2.5}$(μg/m^3)	250±64	274±194	485±518			
A2	$PM_{2.5}$(μg/m^3)		281±113		318±176	184±38	163±22
B	$PM_{2.5}$(μg/m^3)	415±311	159±170	100±41			
D	$PM_{2.5}$(μg/m^3)	442±451	96±25	125±160	105±86		
A1	$PM_{0.01-1}$(×10^5/cm^3)	1.07±3.10	0.89±0.28				
A2	$PM_{0.01-1}$(×10^5/cm^3)		1.19±0.39		1.04±0.21		
B	$PM_{0.01-1}$(×10^5/cm^3)	1.05±0.56	0.57±0.23	0.50±0.24			
D	$PM_{0.01-1}$(×10^5/cm^3)	1.74±1.08	0.40±0.21	0.50±0.23	0.59±0.34		

19:00, possibly due to condensation growth into larger particles or adsorption on the kitchen wall. During Period 2, when the cooking was completely stopped at 19:19, the particle concentration dropped slowly and remained at relatively high levels with a $PM_{2.5}$ mass concentration above 800 μg/m^3.

After the range hood was activated during Period 3, $PM_{2.5}$ mass and sub-micron and super-micron particle number concentrations in the kitchen all exhibited a rapid decrease. During the 28 min in which the range hood was active, $PM_{2.5}$ mass concentration dropped from 803 μg/m^3 to 113 μg/m^3 (i.e., close to the ambient $PM_{2.5}$ concentration) with a removal efficiency of 86%. The removal efficiency of sub-micron and super-micron particle number concentrations reached 80% and more than 90%, respectively. During the 5-min period immediately after the range hood was activated, the number concentrations of accumulation mode particles showed a sharp increase, which was attributed to particle formation in the accumulation mode due to the mixing of cooking smoke and ambient air, and the decrease in temperature and humidity. Overall, the range hood exhibited a high removal efficiency for most of the particles in a certain space within 30 min.

4 Summary and conclusions

To understand the pollution characteristics of particles emitted by outdoor barbecue cooking and the influence on air quality in surrounding areas, in situ measurements of $PM_{2.5}$ mass concentration and particle number concentrations in different size ranges were conducted at seven barbecue restaurants in urban Jinan, China. A very high loading of particles was observed in areas surrounding the barbecue grills. At a one-meter distance, the average $PM_{2.5}$ mass concentration ranged from 250 to 1083 μg/m^3. The sub-micron particle number concentration was in the range of 0.90×10^5 to 2.23×10^5 cm^{-3}. Compared to the ambient urban atmosphere, both mass and number concentrations of particles at the barbecue restaurants increased by several to one hundred times. In particular, the increase in the number concentrations of super-micron particles with a diameter of 1 to 10 μm was much larger than those of sub-micron particles. The barbecue smoke had a significant influence on the particle concentrations over a long distance. In general, $PM_{2.5}$ mass and particle number concentrations presented maximum values in areas very close to the grills and exhibited a concentration peak at a distance of 10 to 15 m. This indicates a possible smoke

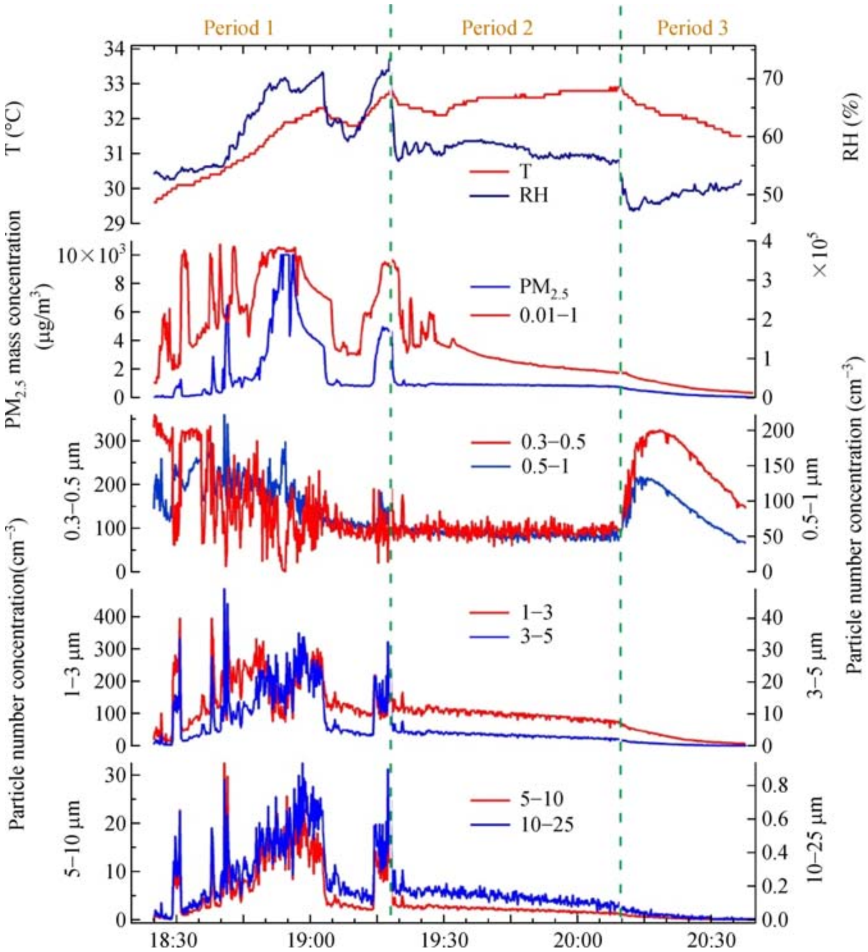

Fig. 4 Time series of $PM_{2.5}$ mass concentration and particle number concentrations in different size ranges during the cooking particle removal test in a kitchen. Three periods were separated by two dashed lines: Period 1 (cooking in the absence of an active range hood), Period 2 (no cooking in the absence of an active range hood), and Period 3 (no cooking in the presence of an active range hood)

transport mechanism of uplift at the grill followed by dispersion and a drop on the downwind side. In addition, the removal efficiency of particles in cooking smoke by a range hood was tested in an indoor kitchen. The range hood could effectively remove cooking particles with an efficiency larger than 80% within 30 min. Thus, it is recommended that barbecue restaurants use a range hood in combination with a smoke purifier. Future studies are required to measure the particle concentration and composition as well as the CO_2 concentration to understand the emission factors of barbecue cooking and further to evaluate its impact on regional air quality.

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