Using a Mobile Measurement to Characterize Number, Surface Area, and Mass Concentrations of Ambient Fine Particles with Spatial Variability during and after a PM Episode

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ABSTRACT

Fine particles play a key role in regional air quality deterioration. Commonly used central-site monitoring data, which offer rough determinations of spatial particulate matter (PM) distributions, is insufficient to estimate potential local emissions or population exposure levels. This study characterizes the spatial variability of fine particles in suburban and rural regions during and after a winter episode of elevated PM (PM episode). Commercial instruments of high time resolution in a mobile laboratory platform were deployed to measure the distribution, number, surface area, and mass concentrations of fine particles. Spatial variations of those particle properties were mainly affected by regional feature, PM episode, local primary source and wind speed. Particle concentrations and size distributions were found to differ considerably during and after PM episode. The PM episode was found to exhibit a lower degree of spatial concentration contrast with respect to particle number, surface area and mass, where similar particle size patterns were distributed across all study regions with decreased particle number under nucleation and Aitken modes and increased number under the accumulation mode. The mobile measurement may supplement information on spatial particle distributions for estimating levels of population exposure and for characterizing detailed physical properties of short-term, high-exposed scenarios.

Keywords: Fine particle; Mobile laboratory platform; Episode; Spatial variation.

INTRODUCTION

Episodes of air quality deterioration are associated with the proliferation of respiratory and heart disease (Chang et al., 2013; Pope, 2000; Pope and Dockery, 2006; Rückerl et al., 2011), and fine particulate matter (PM₂.₅, particles ≤ 2.5 μm in aerodynamic diameter) plays a key role in regional air quality deterioration (Sloane et al., 1991; Deshmukh et al., 2011). PM₂.₅ that contains ultrafine particles (≤ 0.1 μm in aerodynamic diameter) can be more indicative of potential threats to human health than other size fractions (Cascio et al., 2009; Knol et al., 2009; Hoek et al., 2010) because they can easily penetrate into deeper regions of the lung. Recently, more frequent episodes of air quality deterioration have occurred in particular locations and during particular seasons in Taiwan (e.g., the winter episode of elevated PM (PM episode)). While numerous studies have examined ambient mass concentrations and chemical constituents of PM₂.₅ that emerge during episodes of evaluated PM (Niemi et al., 2004; Niemi et al., 2005; Katzman et al., 2010; Chang et al., 2011; Chen et al., 2012; Kuo et al., 2012; Tsai et al., 2012; Wang et al., 2013), physical properties of PM₂.₅ such as number, surface area and size distributions have rarely been investigated. The particle number and surface area in the small size fractions (such as ultrafine particles) can induce significant health impacts (Donaldson et al., 1998; Tran et al., 2000; Elder et al., 2005; Stoeger et al., 2006).

Ambient PM₂.₅ levels are typically obtained via time-integrated, filter-based measurements carried out in air quality network and stationary monitoring sites. This common approach may reveal characteristics of ambient PM₂.₅ under time and space resolution restrictions. In addition, the limited number of air quality networks and stationary monitoring sites established, especially in suburban and rural regions,
results in the formation of vague particle distribution determinations that is neither sufficient to estimate air quality and population exposure levels for a given area (Drewnick et al., 2012) nor adequately identify local primary sources. Typically, such stationary monitoring data that is used directly as a surrogate of exposure with averaged spatial and/or temporal distributions may overestimate or underestimate potential health impacts of local outdoor emissions.

Mobile laboratories equipped with trace gas and/or particle properties generate geographic and meteorological data that are used to measure on-road ambient concentrations of air pollutants (Weijers et al., 2004; Pirjola et al., 2009; Wang et al., 2009; Drewnick et al., 2012; Von der Weiden-Reinmuller et al., 2014). Mobile laboratories have often been used to examine regional pollutants maps, mobile sources and other applications with rapid-response observations of both spatial and temporal variations in urban and traffic-dominated environments (Canagaratna et al., 2004; Schneider et al., 2005; Schneider et al., 2008; Wang et al., 2009). These data cannot be obtained from currently existing stationary monitoring sites. To our knowledge, no study has used a mobile laboratory platform to characterize PM$_{2.5}$ number, surface area and mass concentrations and size distribution during a PM episode and to investigate the spatial variation of those particle properties within a combination of suburban and rural regions affected by a short-term event.

This study characterizes real-time physical properties of ambient fine particles in suburban and rural regions during and after a PM episode using a mobile laboratory platform of high spatial and temporal flexibility.

**METHODS**

**Study Design and Sampling Method**

We conducted a mobile laboratory platform to monitor on-road particles along a selected route (Fig. 1) across rural and suburban regions of Yunlin County, central Taiwan both during (December 9th and 10th, 2013) and after PM episode (December 11th, 12th and 13th, 2013) over five consecutive days in the winter. An elevated PM air quality deterioration alert was announced by the Taiwan EPA when local hourly averaged concentrations of PM$_{10}$ exceeded 125 µg m$^{-3}$. Though it was impossible to specifically determine the episode’s duration, we arbitrarily examined the period running from the day (December 9th) that the PM episode was announced to the following day (December 10th), which exhibited an average PM$_{10}$ level of > 125 µg m$^{-3}$, as the PM episode period. Two Taiwan EPA air quality monitoring stations in the study area (MS1: Mailiao site and MS2: Lunbei site, see Fig. 1) provided hourly meteorological parameters and mass concentrations of PM$_{10}$ and PM$_{2.5}$ for the measurement period.

![Fig. 1. Overview of the sampling map and route (thick colorful line) across the suburban (S1: Siluo, S2: Erlun, S3: Lunbei, and S4: Mailiao) and rural (R1 to R6) regions and two air quality monitoring stations (MS1: Mailiao site and MS2: Lunbei site).](image-url)
The sampling route is away ~4 km south of the Choshui River and ~8 to 24 km east of the Mailiao petrochemical complex, crossing four townships (23°47.070’N, 120°12.207’E, at 4 m above sea level) (Fig. 1). Mobile measurements on the selected sampling route (70 km in total) were conducted during rush hour (7–9 AM and 5–7 PM) twice a day, given our interest in local primary source effects (such as effects of traffic, households, and restaurants) on PM air quality in the study area during and after PM episode. Source conditions likely remained consistent over the sampling days (weekdays). Here, suburban areas (S1–S4) across the township are characterized by high traffic and population density levels, and rural sections (R1–R4) are positioned along agricultural areas with low traffic and population density levels (see Fig. 1).

Mobile Laboratory Platform
A 2012 Luxgen7 MPV gasoline vehicle (L = 4.8 m, W = 1.9 m, H = 1.8 m; net weight = 1.9 metric tons) was used as the mobile laboratory platform for performing on-road measurements (Fig. 2). To ensure the continuous operation of the complete suite of particle instruments, eight 12 V-110AH DC lead-calcium alloy car-batteries with two sets of DC/AC inverters (2500 W-12-110) were installed on the vehicle to support all equipment operations without interruption for up to two hours. An isokinetic sampling system was designed to minimize particle aerodynamic losses (inlet velocity = vehicle speed). The inlet was positioned in the front of the vehicle and 2 m above ground to avoid self-contamination from engine exhausts. When all instruments (as described in detail below) were in operation, airflows containing particles entered the inlet nozzle designed for isokinetic sampling at normal driving speeds. The nozzle was connected to a stainless steel tube (total length of 220 cm, with an inlet of 6.40 mm ID and body of 19.0 mm). The sample flow was decelerated to 1.5 ± 0.1 m s⁻¹, and the flow was then split into four lines of conductive tubes. It should be noted that the flow within the sampling line was at laminar flow (Reynolds number Re < 2000). While we set up other instruments, data generated from these instruments were not used in this study.

Sampling Instruments
The commercial instruments with a high time resolution of one second were deployed in the mobile laboratory. The DustTrak particle monitor (model 8530, TSI Inc., St. Paul, MN, USA) with a flow rate of 3 L min⁻¹ was used to measure mass concentrations of PM₂.₅. The DustTrak mechanism illuminates particles with a laser as particles are drawn into the sensing chamber. Here, a gold-coated spherical mirror captures a significant fraction of light scattered by the particles and redirects this light to a photo detector. The voltage across the photo detector is proportional to the mass concentration of particles over a wide range of concentrations. The Nanoparticle Surface Area Monitor (NSAM, model 3550, TSI Inc., St. Paul, MN, USA) with a flow rate of 2.5 L min⁻¹ was adapted to measure particle surface area deposition concentrations (sizes ranging from 10 to 1000 nm). The NSAM can be programmed to measure deposition levels in the tracheobronchial and alveolar regions of the lung by switching two values of ion trap voltage (100 V and 200 V). The ion trap voltage can be adjusted to manipulate particle size distributions by selecting two voltages that best reflect deposition levels in the two regions of interest in accordance with the deposition fraction of particles as a function of their size and as tabulated by the International Commission on Radiological Protection (Fissan et al., 2007). In the present study, the NSAM was only used to determine

![Fig. 2. Sketch of the mobile laboratory platform (GPS = Global position system; FMPS = Fast Mobility Particle Sizer; DustTrak = DustTrak monitor; APS = Aerodynamic Particle Sizer; NSAM = Nanoparticle Surface Area Monitor).](image-url)
surface area concentrations of deposited particles in the alveolar region of the lung. A Fast Mobility Particle Sizer Spectrometer (FMPS, Model 3091, TSI Inc., St. Paul, MN, USA) with a flow rate of 10 L min$^{-1}$ was used to measure concentrations of particles number in various sizes. The FMPS is composed of two concentric cylinders (classification columns), a corona diffusion charger and 32 electrometers, covering a particle size range of 5.6 to 560 nm. When particles are drawn into the sensing chamber, they are positively charged to a determined level using a corona charger. A particle with high electrical mobility strikes an electrometer positioned near the top, whereas a particle of lower electrical mobility strikes an electrometer positioned lower in the stack. This arrangement, which utilizes highly sensitive electrometers, allows for the simultaneous execution of concentration measurements of various particle sizes. An Aerodynamic Particle Sizer (APS, model 3321, TSI Inc., St. Paul, MN, USA) with a flow rate of 5 L min$^{-1}$ was used to monitor concentrations of particle number ranging from 0.5 to 20 µm in size. The APS is designed to accelerate sampled particles as a carrier gas flows through the converging nozzle. Due to inertial forces, particles accelerate slower than carrier gas, and hence the velocity of the particles lags behind that of the gas. Particle sizes relative to velocity lag are measured at the nozzle opening as particles pass through two closely positioned laser beams. The aerodynamic diameter is calculated based on the time of particle movement between two laser beams (Wang and John, 1987; Rader et al., 1990). The Global Positioning System (GPS, FlexPak6™ Triple-Frequency + L Band GNSS Receiver) and video camera were used to record sampling location, sampling time and vehicle speed data and traffic levels in front of the mobile laboratory platform.

### Quality Assurance and Control

In examining loss mechanism on gravitational settling, impaction, and diffusion, the particle loss resulting from bent section and abrupt contractions in circular tubes of the sampling system were evaluated using the aerosol calculator (Excel 2007–2013 version, http://aerosols.wustl.edu/AAAR workshop08/html/calculator.htm) (Baron and Willeke, 2001), where particle sizes of 2.5–0.1 µm, 0.1–0.01 µm and 0.01–0.005 µm accounted for <2%, <1% and 6–10% of particle loss, respectively. In addition, all particle instruments were set to zero using particle-free air with a HEPA filter before and after each measurement. As self-contamination by engine exhaust is necessary to consider, we maintained a mobile laboratory platform speed of 50 ± 5 km h$^{-1}$ (=13.9 m s$^{-1}$) for isokinetic sampling purpose to avoid this potential bias of measurements, when the wind was from the behind of the vehicle with the speed (= 3.9 m s$^{-1}$, range = 1.0–7.3 m s$^{-1}$, observed in Yunlin County shown in Table 1) during the sampling period. In the presence of 10% less than 50 km h$^{-1}$ vehicle speed and <20% backwind on-road data (based on wind directions measured in MS1 and MS2, Table 1) yielded a net self-contamination probability of <2%. Additionally, throughout the sampling period, the driver of the mobile laboratory deliberately drove far behind any vehicle driving directly in front of the sampling vehicle. To ensure the reliability and precision of the FMPS, a Scanning Mobility Particle Sizer (SMPS) was used on three occasions (chamber with 0.5% (NH4)2SO4 nanoparticles; suburban indoor and outdoor environments, see Fig. S1 in supplementary information) for inter-instrument comparison, as SMPS particle size distributions with longer time (1 min) periods are recommended for studying atmospheric aerosols. This detailed procedure of inter-instrument comparison was employed in a previous study (Jeong and Evans, 2009).

### RESULTS AND DISCUSSION

#### Episode Type Identification Based on Taiwan EPA Air Quality Data

Fig. 3 presents the wind rose diagram obtained from Lunbei (MS2) air quality monitoring stations (data for Mailiao (MS1) is not shown) during and after PM episode for the

<table>
<thead>
<tr>
<th>Site</th>
<th>Episode</th>
<th>Date</th>
<th>PM$_{10}$ (µg m$^{-3}$)</th>
<th>PM$_{2.5}$ (µg m$^{-3}$)</th>
<th>PM$<em>{2.5}$/PM$</em>{10}$ ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mailiao</td>
<td>during</td>
<td>Dec. 09$^{th}$</td>
<td>81.6 ± 26.4</td>
<td>52.1 ± 22.3</td>
<td>0.63 ± 0.14</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dec. 10$^{th}$</td>
<td>143.2 ± 25.0</td>
<td>86.6 ± 20.2</td>
<td>0.61 ± 0.12</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dec. 11$^{th}$</td>
<td>81.2 ± 16.9</td>
<td>44.8 ± 10.3</td>
<td>0.56 ± 0.07</td>
</tr>
<tr>
<td></td>
<td>after</td>
<td>Dec. 12$^{th}$</td>
<td>70.7 ± 8.6</td>
<td>44.7 ± 7.9</td>
<td>0.63 ± 0.06</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dec. 13$^{th}$</td>
<td>58.7 ± 14.1</td>
<td>33.4 ± 11.4</td>
<td>0.56 ± 0.07</td>
</tr>
<tr>
<td>Lunbei</td>
<td>during</td>
<td>Dec. 09$^{th}$</td>
<td>76.3 ± 24.8</td>
<td>49.4 ± 20.1</td>
<td>0.64 ± 0.15</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dec. 10$^{th}$</td>
<td>134.4 ± 24.5</td>
<td>95.0 ± 19.4</td>
<td>0.71 ± 0.09</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dec. 11$^{th}$</td>
<td>86.4 ± 16.8</td>
<td>49.7 ± 14.1</td>
<td>0.57 ± 0.10</td>
</tr>
<tr>
<td></td>
<td>after</td>
<td>Dec. 12$^{th}$</td>
<td>85.8 ± 21.4</td>
<td>57.2 ± 14.5</td>
<td>0.67 ± 0.11</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dec. 13$^{th}$</td>
<td>68.0 ± 10.0</td>
<td>36.7 ± 7.1</td>
<td>0.54 ± 0.07</td>
</tr>
</tbody>
</table>
Fig. 3. Wind rose diagram obtained from Lunbei (MS2) monitoring station (a) during and (b) after PM episode for study region.

Sampling region. High wind speeds were found during PM episode. The predominant wind direction was northeast for both periods. Table 1 shows 24-hour average concentrations of PM$_{10}$, PM$_{2.5}$, and PM$_{2.5}$/PM$_{10}$ ratios for the study period obtained from Mailiao (MS1) and Lunbei (MS2). On average, elevated mass concentrations of PM$_{10}$ (Mailiao site = 112.3 µg m$^{-3}$ and Lunbei site = 105.2 µg m$^{-3}$) and PM$_{2.5}$ (Mailiao site = 69.4 µg m$^{-3}$ and Lunbei site = 72.2 µg m$^{-3}$) were observed during PM episode, while concentrations of PM$_{10}$ (Mailiao site = 70.2 µg m$^{-3}$ and Lunbei site = 80.1 µg m$^{-3}$) and PM$_{2.5}$ (Mailiao site = 41.0 µg m$^{-3}$ and Lunbei site = 47.9 µg m$^{-3}$) declined after PM episode. Here, we found that the mean ratio of PM$_{2.5}$/PM$_{10}$ during PM episode (Mailiao site = 0.62 and Lunbei site = 0.68) was relatively higher than that after PM episode (Mailiao site = 0.59 and Lunbei site = 0.60), suggesting that particle size distributions (for small particle sizes especially) during transport from sources to ambient monitors likely differed on both occasions.

In Taiwan, PM episodes are usually caused by long-range dust storm and frontal pollution transport and local emissions resulting from river dust and stagnant weather (Kuo et al., 2013). PM$_{2.5}$/PM$_{10}$ ratios of less than 0.50 dominated by coarse particles have been reported in central Taiwan for the river dust episode period (Kuo et al., 2010); additionally, stagnant weather of low wind speeds (< 2.0 m s$^{-1}$) can spur a local pollution episode (Kuo et al., 2013). The PM episode examined in this study was unlikely attributable to local pollution characteristic of Taiwan, such as river dust and stagnant weather patterns, as high wind speeds (~4.0 m s$^{-1}$) and PM$_{2.5}$/PM$_{10}$ ratios (> 0.60) were observed for the study region (see Table 1). This PM episode was instead exclusively caused by long-range transported pollution. In addition, the episode was unlikely attributable to emissions generated by the nearby Mailiao petrochemical complex, as wind directions were mainly oriented northeast during the study period (Table 1).

Particle Size Distributions

Fig. 4 shows average particle number size distributions (5.6 nm to 2,500 nm) generated from on-road measurements for the sampling route covering suburban (S1 – S4) and rural (R1 – R6) regions for periods during and after PM episode. Overall, particle concentrations and size distributions changed considerably after PM episode. The PM episode caused similar size distribution patterns across the entire study region even though particle concentrations varied regionally. The constant presence of a 10 nm peak is likely attributable to local traffic emissions and nucleation events that occurred during the sampling period. However, this FMPS result may be partly related to a positive (or negative) artifact resulting from FMPS electrometer or data inversion algorithm calibration (Jeong and Evans, 2009). Thus, a bimodal distribution dominated by the highest 20 nm (nucleation mode) peak (where particles have the highest deposition efficiency in the alveolar region (~50%) and greater health impacts (Fissan et al., 2007)), was considered during PM episode. In addition, the accumulation mode (0.1–1 µm) was dramatically increased for all regions relative to that of the period following the PM episode, indicating that this PM episode may have generated larger particles, in addition to promoting the formation of small-particle aggregations. Here, an accumulation mode to Aitken mode concentration ratio of less than one was obtained for the PM episode.

After the PM episode occurred, regional particle size distributions varied somewhat, and particle concentrations for each size range varied regionally. For instance, R1 and R2 sections with similar distribution patterns may solely be affected by on-road traffic emissions and limited residential emissions (see Fig. 1). The R3 to R6 section covers main roads which connect each township (S1–S4), where traffic,
Fig. 4. Particle number size distribution (5.6 nm to 2.5 µm), on average, with on-road measurements in the selected suburban (S1: Siluo, S2: Erlun, S3: Lunbei, and S4: Mailiao) and rural (R1 to R6 corresponding to Fig. 1.) regions during and after PM episode.
residential and restaurant emissions may contribute to on-road particle measurements. As a result, particle number and size distributions of dissimilar patterns and concentration levels were obtained from R3 to R6 sections (Table 2). Number concentration in size distributions with peaks at 10, 20, or 40 nm (Aitken mode) were found for all of the studied regions during the PM episode period, complementing previous studies on particle features found during the PM episode of Nanjing, China (Kang et al., 2013).

While the reliability and precision of the FMPS for atmospheric aerosol monitoring has been questioned, it is still frequently used to quantify particle number and size distributions via on-road measurements given its high time resolution capabilities. Via particle monitoring, we found that SMPS and FMPS inter-comparison results correlated well ($r^2 > 0.80$) for number concentrations of 6 to 100 nm measured on three occasions. Only the size distribution with a mode diameter of 10 nm for suburban outdoor aerosols presented an inconsistent pattern (Fig. S1 in supplementary information). Similar number and size distribution results were found by Jeong and Evans (2009).

**Spatial Patterns of Particle Concentration**

As shown in Fig. 3, strong temporal and spatial variations in concentrations of on-road particle mass, surface area (particles deposited in the alveolar region of the lung) and number for each run during and after PM episode were measured from December 9th to 13th in rural and suburban regions of Yunlin County. Overall, spatial patterns of particle concentrations changed following the PM episode which could generate relatively lower spatial differences in particle mass, surface area, and number concentrations (Fig. 5). The lower day-to-day variation in particle concentrations during PM episode was observed in comparison with the period after PM episode. We can see lower concentrations of particle mass, surface area, and number for each run during and after PM episode. We can observe that suburban areas (with the exception of S2) did not show high particle concentrations than rural areas during PM episode. Unsurprisingly, we found the relatively higher particle mass, surface area, and number in suburban regions after PM episode (Table 2). For instance, we observed lower number, surface area and mass concentrations in the upwind rural areas (e.g., R1: number = 2.41 ± 2.99 10^4 # cm^{-3}, surface area = 13.64 ± 6.65 µm^2 cm^{-3} and mass = 34.52 ± 6.57 µg m^{-3}) after PM episode, in comparison with those in the downwind suburban vicinity of driving vehicles on the road.

We also summarized particle concentrations performing an average of all runs for each selected region (S1–S4 and R1–R6) during and after episode (as shown in Table 2). On average, the higher mass concentrations (during = 72.72 ± 43.91 µg m^{-3}, after = 49.92 ± 24.16 µg m^{-3}), but lower surface area (during = 20.92 ± 18.03 µm^2 cm^{-3}, after = 22.63 ± 19.18 µm^2 cm^{-3}) and number (during = 2.80 ± 6.00 × 10^4 # cm^{-3}, after = 3.82 ± 7.98 × 10^4 # cm^{-3}) concentrations were observed for all runs during the PM episode. Suburban areas (with the exception of S2) did not show high particle concentrations than rural areas during PM episode. Unsurprisingly, we found the relatively higher particle mass, surface area, and number in suburban regions after PM episode (Table 2). For instance, we observed lower number, surface area and mass concentrations in the upwind rural areas (e.g., R1: number = 2.41 ± 2.99 10^4 # cm^{-3}, surface area = 13.64 ± 6.65 µm^2 cm^{-3} and mass = 34.52 ± 6.57 µg m^{-3}) after PM episode, in comparison with those in the downwind suburban vicinity of driving vehicles on the road.

**Table 2.** The mean (± standard deviation) of number, surface area and mass concentrations of PM2.5 obtained from the mobile laboratory platform for study areas during and after PM episode.

<table>
<thead>
<tr>
<th>Section</th>
<th>No. of measurements</th>
<th>Number (10^4 # cm^{-3})</th>
<th>Surface area (µm^2 cm^{-3})</th>
<th>Mass (µg m^{-3})</th>
<th>p-value</th>
<th>p-value</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>During</td>
<td>SI</td>
<td>279</td>
<td>2.11 ± 1.03</td>
<td>5.45 ± 9.02</td>
<td>0.011</td>
<td>&lt; 0.01</td>
<td>&lt; 0.01</td>
</tr>
<tr>
<td></td>
<td>S2</td>
<td>322</td>
<td>5.24 ± 11.02</td>
<td>3.67 ± 2.62</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>S3</td>
<td>427</td>
<td>1.08 ± 2.43</td>
<td>0.67 ± 2.62</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>S4</td>
<td>1411</td>
<td>2.99 ± 1.96</td>
<td>4.06 ± 3.43</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R1</td>
<td>2439</td>
<td>5.760 ± 7.76</td>
<td>5.44 ± 8.76</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R2</td>
<td>1503</td>
<td>5.08 ± 7.49</td>
<td>5.54 ± 7.99</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R3</td>
<td>1457</td>
<td>2.990 ± 2.43</td>
<td>3.98 ± 2.75</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R4</td>
<td>278</td>
<td>1.067 ± 1.96</td>
<td>2.22 ± 1.29</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
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<tr>
<td></td>
<td>R5</td>
<td>416</td>
<td>1.580 ± 3.34</td>
<td>3.58 ± 5.59</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
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<tr>
<td></td>
<td>R6</td>
<td>1797</td>
<td>1.706 ± 2.43</td>
<td>1.52 ± 1.52</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td>After</td>
<td>SI</td>
<td>279</td>
<td>3.54 ± 9.02</td>
<td>39.32 ± 43.83</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
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</tr>
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<td></td>
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<td>3.98 ± 2.75</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R4</td>
<td>278</td>
<td>1.067 ± 1.96</td>
<td>2.22 ± 1.29</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R5</td>
<td>416</td>
<td>1.580 ± 3.34</td>
<td>3.58 ± 5.59</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td></td>
<td>R6</td>
<td>1797</td>
<td>1.706 ± 2.43</td>
<td>1.52 ± 1.52</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td>Mean (SI–S4)</td>
<td>990</td>
<td>2.61 ± 1.03</td>
<td>5.45 ± 9.02</td>
<td>39.32 ± 43.83</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
<tr>
<td>Mean (R1–R4)</td>
<td>8703</td>
<td>2.80 ± 6.00</td>
<td>3.82 ± 7.98</td>
<td>49.92 ± 24.16</td>
<td>&lt; 0.01</td>
<td>&gt; 0.05</td>
<td>&gt; 0.05</td>
</tr>
</tbody>
</table>
The similar pattern for R3 (upwind rural areas with lower particle levels) versus S3 (downwind suburban area with higher particle levels) was also obtained. As a result, particle concentrations in suburban regions were mainly affected by local primary sources (such as traffic or household emission) without episode effects. We found average number concentrations of PM$_{2.5}$ during PM episode ($2.80 \times 10^4$ # cm$^{-3}$) to be slightly lower than those after PM episode ($3.82 \times 10^4$ # cm$^{-3}$), and ultrafine particles accounted for 83.0% and 92.0% of total number concentrations of PM$_{2.5}$ during and after PM episode, respectively. We further compared particle number concentration for nucleation ($< 0.025$ µm), Aitken (0.01–0.1 µm) and accumulation (0.1–1 µm) modes for two periods. Particle number concentration, on average, increased by a factor of 1.8 in the accumulation mode and decreased in the Aitken and nucleation modes by factors of 0.6 and 0.8, respectively, during PM episode. Such findings are consistent with those of previous studies (Hinds, 1999; Mönkkönen et al., 2004; Niemi et al., 2004; Niemi et al., 2005; Jayaratne et al., 2011; Kang et al., 2013). Hinds (1999) found that greater differences in particle size correlate with more significant polydisperse coagulation effects. Thus, small molecular clusters and particles are efficiently scavenged by high concentrations of larger particles. In addition, Mönkkönen et al. (2004) and Salma et al. (2011) found that a higher surface area of larger particles results in the consumption of condensable vapors, thus preventing particle formation and growth. Consequently, the total number of particles with diameters of less than 500 nm changed little, as the decline in nucleation and Aitken mode concentrations was balanced by an increase in accumulation mode concentrations (Mönkkönen et al., 2004). Hence, the decrease in particle number and surface areas (deposited particles in the alveolar region of the lung) found under the nucleation and Aitken modes for PM episode period was foreseeable. However, the decrease in surface area and number concentrations and the increase in mass concentrations during episode were not found in all of the selected sampling sections (e.g., S2, R1, and R2 sections in Table 2). The cause of this inconsistent result remains unknown.

**Study Limitations**

Mobile on-road measurements are limited in that processes occurring in the immediate vicinity of the mobile laboratory; e.g., vehicles driving on the road or sources next to the road can affect measurements and therefore bias local particle results. Short-term events can also change significantly over the course of a measurement period, potentially limiting the accuracy of study results. Additionally, temporal variability can be easily interpreted as spatial variability and vice versa. However, this was not the case for the PM episode period examined, as it covered two days. While the mobile laboratory platform generates on-road particle measurements of high temporal and spatial resolution, we note that the results presented here only apply to the measured period and route and may not represent the particle distributions for other times of day or for other areas. Thus, our two-hour-

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**Fig. 5.** (a) Mass, (b) surface area and (c) number concentrations of particles in the selected sections for each run during and after PM episode.
long measurements conducted in the study area at rush hour periods twice a day (morning and night) may overestimate daily particle concentrations if these measurement data are used in population-based health studies. Although the mobile measurement with high time resolution allows us to differentiate the spatial variation within a sampling route across rural and suburban areas for episode and non-episode periods, we are not able to identify the specific sources of particle resulting from local traffic, household or restaurant contributions.

CONCLUSIONS

While stationary air quality monitoring stations generate reliable mass concentration and meteorological data with temporal variations (hourly scale) for a particular site where an assumption of evenly dispersed particles is applied, spatial distributions of on-road particle parameters (particle number, surface area, mass and size distributions) could aid the effort on sufficiently estimating air quality and population exposure levels in a given area. We characterized spatial and temporal variations of the above physical properties for on-road fine particles in suburban and rural regions by conducting mobile laboratory platform experiments during episodes and after PM episode. The PM episode observed in this study, which is likely attributable to long-range frontal pollution transport, presented unique particle number size distribution wherein similar distributions evenly covered entire regions of the selected route in relation to those present after PM episode. Particle size distributions revealed through mobile measurements may help identify the origins of particles found in suburban and rural areas. While the PM episode was expected to increase particle mass concentrations; the particle number and surface area in small particle sizes, decreased under nucleation (<0.025 µm) and Aitken (0.01–0.1 µm) modes in most cases. This phenomenon was confirmed through our findings of dramatic increases of the particle number under the accumulation mode due to polydisperse coagulation. Our results show that particle trends were mainly shaped by episode, by local primary sources and by meteorological conditions. Thus, spatial and temporal particle variations in suburban and rural regions are vital to characterize.

ACKNOWLEDGEMENTS

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aqar.org.

REFERENCE


