HHS Public Access

Author manuscript

Aerosol Air Qual Res. Author manuscript; available in PMC 2018 December 19.

Published in final edited form as:

Aerosol Air Qual Res. 2017 September ; 17(9): 2235–2246. doi:10.4209/aaqr.2016.07.0301.

Comparison of $PM_{2.5}$ Exposure in Hazy and Non-Hazy Days in Nanjing, China

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Abstract

Fine particulate matter ($PM_{2.5}$), levels of which are about 6 times the 2014 WHO air quality guidelines for 190 cities in China, has been found to be associated with various adverse health outcomes. In this study, personal $PM_{2.5}$ exposures were monitored along a fixed routine that included 19 types of non-residential micro-environments (MEs) on 4 hazy days (ambient $PM_{2.5}$ 292 ± 70 μg m^{-3}) and 2 non-hazy days (55 ± 16 μg m^{-3}) in Nanjing, China using miniaturized real-time portable particulate sensors that also collect integrated filters of $PM_{2.5}$ (MicroPEMs, Research Triangle Institute (RTI), NC). Gravimetric correction is necessary for nephelometer devices in calculating real-time PM levels. During both hazy and non-hazy days, personal $PM_{2.5}$ levels were generally higher in MEs with noticeable $PM_{2.5}$ sources than MEs serving as receptor sites, higher in open MEs than indoor MEs, and higher in densely populated MEs than MEs with few people. Personal $PM_{2.5}$ levels measured during hazy and non-hazy days were 242 ± 91 μg m^{-3} and 103 ± 147 μg m^{-3}, respectively. The ratio of personal exposure to ambient $PM_{2.5}$ levels ($r_{p/a}$) was less than 1.0 and less variable on hazy days (0.85 ± 0.31); while it was larger than 1.0 and more variable on non-hazy days (1.71 ± 1.93), confirming the importance of local sources other than ambient during non-hazy days. Air handling methods (e.g., ventilation/filtration) impacted personal exposures in enclosed locations on both types of days. Street food vendors with cooking emissions were MEs with the highest personal $PM_{2.5}$ levels while subway cars in Nanjing were relatively clean due to good air filtration on both hazy and non-hazy days. In summary, on hazy days, personal exposure was mainly affected by the regional ambient levels, while on non-hazy days, local sources together with ambient levels determined personal exposure levels.

Keywords

$PM_{2.5}$; Micro-environment; Haze; Personal exposure; Subway

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SUPPLEMENTARY MATERIAL

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

INTRODUCTION

China has experienced severe air pollution over the past 10 years, as air pollutant emissions increased with rapid industrialization and relentless fossil fuel consumption (Zhang *et al.*, 2012; Guo *et al.*, 2014). $PM_{2.5}$, airborne particulate matter with aerodynamic diameter below 2.5 μm, frequently exceeds Chinese National Ambient Air Quality Standards of 35 μg m^{-3} per year over a large area including the Yangtze River Delta and Beijing-Tianjin-Heibei area (Chen *et al.*, 2013; Wang *et al.*, 2013). A one-year (2014–2015) survey of 190 Chinese cities revealed that the annual average $PM_{2.5}$ concentration in these cities was 61 μg m^{-3}, about six times the 2014 WHO air quality guideline (10 μg m^{-3}) (Zhang and Cao, 2015). Elevated levels of $PM_{2.5}$ obscures the sky and leads to haze, an atmospheric phenomenon defined as the concurrence of an average ambient concentration of $PM_{2.5}$ > 75 μg m^{-3} with visibility < 5 km for more than six consecutive hours (WMO, 2008; MEP, 2014). Days that exceed the standards are called “hazy days” in China. Otherwise they are called “non-hazy days”.

Numerous epidemiological studies have observed significant associations of chronic exposure to toxic components of $PM_{2.5}$, including heavy metals and polycyclic aromatic hydrocarbons (PAH), with adverse health effects such as respiratory or cardiovascular diseases and lung cancer (Cao *et al.*, 2013; Li *et al.*, 2013; Yang *et al.*, 2013). Even short-term exposure to high levels of $PM_{2.5}$ can lead to adjuvant effects in acute inflammatory and immunological responses (Salvi *et al.*, 1999; Svartengren *et al.*, 2000). Because of the high exposures to $PM_{2.5}$, studies have indicated that $PM_{2.5}$ has become the fourth leading health threat in China, accounting for enormous life expectancy loss and carcinogenic/non-carcinogenic diseases (Chen *et al.*, 2013; Yang *et al.*, 2013).

There is a need for more focused studies in China to characterize personal exposures and how the relative importance of different exposure pathways changes on hazy days vs. non-hazy days. The majority of the aforementioned health studies in China rely on data from central monitoring sites, which can underestimate (Wallace *et al.*, 2006; Rodes *et al.*, 2010) or overestimate (Du *et al.*, 2010; Jahn *et al.*, 2013) individual exposures, depending on the proximity to local $PM_{2.5}$ sources and usage of advanced air handling technology that includes filtration such as heating, ventilation and air conditioning (HVAC), which can substantially affect $PM_{2.5}$ levels in various microenvironments (MEs) where people routinely spend time. Personal exposure measurements are regarded as a gold standard for characterizing exposure (Delfino *et al.*, 2004; Morawska *et al.*, 2013). Even if exposures to elevated $PM_{2.5}$ in certain MEs are short in duration, it can contribute substantially to total daily exposure (Brown *et al.*, 2012). To date, personal exposure data in China have largely been obtained from 1) an individual ME (Baumgartner *et al.*, 2011) or multiple transportation MEs (Du *et al.*, 2010; Lu *et al.*, 2015); 2) daily average $PM_{2.5}$ concentrations without distinguishing contribution from various MEs (Jahn *et al.*, 2013; Chu *et al.*, 2015); or 3) using fixed-site monitoring data to substitute for personal exposure measurement. Furthermore, most of these studies were conducted on non-hazy days when ambient $PM_{2.5}$ concentration was below or around 75 μg m^{-3}. More integrated research on personal exposure in various MEs have been conducted in Europe and America (Adams *et al.*, 2001; Nieuwenhuijsen *et al.*, 2007; Hammond *et al.*, 2014), where annual ambient $PM_{2.5}$

concentrations were mostly < 45 μg m^{-3} or even < 20 μg m^{-3} (Liu *et al.*, 2004; Boldo *et al.*, 2006), not representative of the elevated ambient levels in China.

To illustrate the impacts of ambient $PM_{2.5}$ concentration in open and enclosed MEs, we measured $PM_{2.5}$ levels in various non-residential MEs during both hazy and non-hazy days. The goal of this study was to serve as reference for future epidemiological research on $PM_{2.5}$. In addition, approaches to minimizing $PM_{2.5}$ exposure under different pollution levels are discussed.

METHODS and MATERIALS

Sampling Area

Nanjing, the second largest city in eastern China with a population of ~8.2 million, has been undergoing rapid urbanization and industrialization. Average annual $PM_{2.5}$ concentrations in Nanjing in 2013 and 2014 were 78 μg m^{-3} and 74 μg m^{-3}, more than twice the national standard (35 μg m^{-3}) and over 7 times the WHO air quality guidelines. In 2013, Nanjing experienced high daily average $PM_{2.5}$ concentrations (> 75 μg m^{-3}) for about 242 days. Between Dec. 2nd and Dec. 14th 2013, a severe haze episode occurred with hourly $PM_{2.5}$ concentrations reaching up to 500 μg m^{-3} during the event. This event stretched over a distance of about 1,200 km from central to eastern China (Wang *et al.*, 2015) and attracted wide public concerns about air pollution.

Sampling Design

In this study, personal $PM_{2.5}$ exposure was monitored during part of this haze episode (from Dec. 4th to Dec. 8th, 2013), while exposures during non-hazy days were monitored in late Dec. 2013 and in March 2014. The sampling route was designed in consideration of 1) including microenvironments with local emission sources (e.g., motor vehicles, cooking) and common receptor sites like hospitals and shopping malls; and 2) proximity to nine of the Nanjing Municipal Environment Monitoring Center (NMEMC) monitoring sites where the ambient $PM_{2.5}$ levels are measured so that we could compare personal exposure data with ambient data from the nearest NMEMC site. Sampling started and ended in an office on the Xianlin campus of Nanjing University located in northeastern Nanjing (near XL site in Fig. 1).

The map of the chosen route is shown in Fig. 1, and details about selected MEs are listed in Table 1. Thirty five non-residential sub-MEs in downtown Nanjing were selected to characterize personal exposure patterns. In addition, both rush hour and non-rush hour samplings were included. MEs were classified into open MEs (e.g., road and park and open mall) and enclosed MEs (e.g., subway train and office) according to whether there was continuous natural ventilation or not. Based on whether there were emission sources, MEs were further classified into source MEs (e.g., road, bus station, subway station, street food vendor) and receptor MEs with relatively little $PM_{2.5}$ emission sources other than resuspension or personal clouds (Wallace *et al.*, 2006) such as shopping mall, lab, and office.

Depending on the type of MEs, the activity levels of the investigator who wore the monitor vary. The investigator sat in MEs such as taxi, restaurant, office and lab, sat/stood in bus or

subway cars and stations, and walked in the rest of the MEs such as parks. If sitting in a ME, the investigator would try to find a seat near its center. Sampling durations in each ME were at least 15 min except for taxi. Time spent in a taxi could be as low as 12 min, depending on traffic conditions.

$PM_{2.5}$ exposure was monitored from 8 am to 10 pm, for a total of about 14 hrs per day for four severe hazy (ambient $PM_{2.5}$ concentration > 280 μg m^{-3}) and two non-hazy days (i.e., ambient $PM_{2.5}$ concentration < 75 μg m^{-3}). Real-time $PM_{2.5}$ concentrations were measured by MicoPEM (v3.2, RTI International, NC, USA), which is a miniaturized personal $PM_{2.5}$ environment monitor that includes a two stage impactor with a $PM_{2.5}$ cut point (PM_{10} impactors are also available), a nephelometer for realtime measurements of particles with diameter > 300 nm, followed by a Teflo filter for an integrative gravimetric measurement of the entire size range of $PM_{2.5}$. The integrative filter allows the MicroPEMs nephelometer to be calibrated to the average sensitivity towards particulate matter during the sampling time that is represented by the filter. The sampling route was tracked using a portable GPSMAP 60CSx Navigator (Garmin, Taiwan). Both MicroPEM and GPS units were placed in the pocket of a jacket worn by a student-investigator, with the inlet of the MicroPEM extended to the student-investigator's breathing zone via a static dissipative Polyurethane tube (Freelin-Wade, OR, USA). Detailed time-activity diaries were also recorded.

Four fixed-site side-by-side deployments were conducted to explore the relationship between the MicroPEM response and one of the NMEMC $PM_{2.5}$ monitors. Among these comparisons, one run took place on a non-hazy day and three were on hazy days. Horizontal distance between MicroPEM and BAM-1020 was 1.5 m while inlets of these two sensors were at the same vertical height. NMEMC stations used BAM-1020 beta attenuation mass monitors (Met One Instruments, OR, USA) for $PM_{2.5}$ measurement. To test reproducibility, a side-by-side comparison of two MicroPEM units was also conducted. For this test, an extra MicroPEM unit was placed in another pocket. The inlet tubes of these two units were both extended to the breathing zone while keeping a distance of about 25 cm between them.

MicroPEMs were set to log data at a 10-second interval and were set to a target flow rate of 0.5 L min^{-1} calibrated with a TSI Flowmeter 4140 (TSI Incorporated, MN, USA). After passing an impactor with an aerodynamic cutoff of 2.5 μm, particles were collected onto a 25 mm Teflo filter (Pall Corporation, NY, USA) masked with a filter holder cassette to reduce the active sampling area to 78.5 mm^2 (active diameter of 10 mm). At the beginning and end of each sampling event, the nephelometer value for zero particles was obtained in the field by running the unit with a HEPA filter attached to the inlet for a 5 min period. Sample filters were kept in individual petri dishes in a sealed plastic bag and stored at −4°C. Filters were pre- and post-weighed in the automated MTL Weighing Chamber customized to be trace metal clean and installed at the Lamont-Doherty Earth Observatory (LDEO, Columbia University, USA) to calculate gravimetric mass (GM); filters were equilibrated in the chamber (long term stability of temperature at 22.5 ± 0.26°C; RH at 33.6 ± 1.6%) for 24 hrs and then weighed on a Mettler UMX2 microbalance, with 6 Po-210 strips for disappating static charge and with a final correction for buoyancy differences between pre and post weighing days.

Data Analysis

Data was analyzed using R software packages. Data from MicroPEMs were transformed to minute-averages for the analysis in this study. MicroPEM nephelometer readings were first adjusted for the interpolated HEPA readings collected at the beginning and end of each session. Personal exposure $PM_{2.5}$ concentrations were then gravimetrically corrected (GC). The GC ratio (r_{GC}) is the ratio of net grametric weight to the calculated nephelometer-based weight using the following equation:

$$r_{GC} = \frac{m_{post} - m_{pre} - m_{field\ blank}}{\sum_{i=1}^{N}(f_i \times c_i)} \quad (1)$$

where f_i and c_i, are the flow rate (m^3 per min) at the time internal i and its corresponding $PM_{2.5}$ concentration (μg m^{-3}) averaged in the interval i, respectively. In addition, m_{pre} and m_{post} are filter mass (μg) weighed before and after sampling and $m_{field\ blank}$ is the average weight change of field blank filters (FBF). FBF are filters that had been installed in MicroPEMs and brought to and from the field but not used for sampling. The weight change of each FBF was calculated by substracting pre-weight with post-weight. This was used to correct the possible filter weight changes and/or contamination during the transport, storage, and handling of the filters. Adjusted nephelometer $PM_{2.5}$ concentrations $C_{corrected}$ was then calculated using r_{GC}:

$$C_{Correct} = C_{raw} \times r_{GC} \quad (2)$$

where C_{raw} is the per-minute average concentration measured by MicroPEM.

Based on the time-activity diary, personal exposure data in vatious MEs were grouped. The ambient hourly data of these MEs reported by the nearest monitoring station were downloaded from the NMEMC website.

RESULTS

Side-by-Side Comparison and Gravimetric Correction

Fig. 2(a) shows the fixed site co-location results before gravimetric correction (GC) for MicroPEM and the NMEMC BAM-1020. The MicroPEM hourly averages of uncorrected values show a strong correlation with the NMEMC monitor results but with slopes less than 1 for non-hazy days and greater than one for hazy days.

Based on data collected both during the co-location and data from the personal deployments, the average gravimetric correction (GC) ratio r_{GC} for the 2 non-hazy days was 1.48 ± 0.43 while the average for 4 hazy days was 0.59 ± 0.07. These ratios are consistent with the trend in slopes seen in Fig. 2(a), and confirm that the Nanjing aerosols have different optical properties on non-hazy days vs. hazy days. In the co-location experiment, some filters had not been pre-weighed; for these filters, their data were corrected using these average $r_{GC.}$

Fig. 2(b) shows the MicroPEM data after gravimetric correction. Note the agreement to the NMEMC data are much better with the slopes now being within 10% of a value of 1.0 for both hazy and non-hazy days, where before correction they were quite different, off by 27% for non-hazy days and 56% for hazy days. Average $PM_{2.5}$ concentration of all monitoring hours is 186 ± 83 μg m^{-3} for MicroPEM data after GC and 195 ± 98 μg m^{-3} for NMEMC measured data.

As for duplicate deployment between two MicroPEM units for personal sampling, the 14-hr $PM_{2.5}$ averages were 34.3 μg m^{-3} for unit 472N and 80.7 μg m^{-3} for unit 474N before GC; and 228 and 227 μg m^{-3}, respectively, after gravimetric correction (Fig. S2). The percent difference between two units decreased from 135% to 0.3% (Fig. S2). The correction ratio, r_{GC}, was 6.65 and 2.82 for sampling from units 472N and 474N, respectively.

Hazy Days vs. Non-Hazy Days

As expected, total personal exposure in all MEs on hazy days was about 2.5 times of that on non-hazy days (Table 2) while ambient $PM_{2.5}$ levels were 292 ± 70 μg m^{-3} and 55 ± 16 μg m^{-3} on hazy and non-hazy days, respectively. On both hazy and non-hazy days, open MEs had higher $PM_{2.5}$ levels than enclosed MEs while $PM_{2.5}$ levels were higher in source MEs than in receptor MEs. $PM_{2.5}$ levels in enclosed receptor MEs (Table 2) were much lower than in the other three MEs (i.e., open source, open receptor and enclosed source MEs) on both hazy and non-hazy days, clearly due to removal of $PM_{2.5}$ by HVAC filtration that may be obscuring source terms within these enclosed “receptor” MEs. On hazy days, the open source MEs showed similar $PM_{2.5}$ levels to open receptor and enclosed source MEs, in contrast to the non-hazy days when the open source ME showed 30 to 40% higher levels to the open receptor and enclosed source MEs. Also, the variability as assessed by standard deviations were similar for open source, open receptor, enclosed source and enclosed receptor MEs in hazy days, but much higher on non-hazy days in open source MEs than the other three MEs on non-hazy days.

Thirty five sub-MEs in Table 1 were grouped into 19 classes of MEs as shown in Fig. 3. Among various source MEs, taxis had the lowest $PM_{2.5}$ personal exposure levels and street food vendors and restaurants showed the highest levels. As for receptor MEs, lab and underground/ground-level subway cars had the lowest levels while parks and school yards had relatively high $PM_{2.5}$ levels.

Personal vs. Ambient

Personal exposure was significantly correlated ($p < 0.001$) with simutaneously monitored NMEMC $PM_{2.5}$ level (hourly average of closest NMEMC site) in most MEs; exceptions include several enclosed receptor MEs (lab, underground mall, and mall) that had good HVAC filtration or MEs with cooking sources (Table S1). When separating this analysis into non-hazy days and hazy days, the correlation coefficients between minute personal exposed $PM_{2.5}$ concentration and the ambient $PM_{2.5}$ level were generally lower across the two non-hazy days than the four hazy days.

Fig. 4 shows a comparision between various MEs and ambient data. On non-hazy days, $PM_{2.5}$ average levels in most individual MEs were higher than ambient $PM_{2.5.}$ In contrast,

on hazy days, most open MEs have comparable $PM_{2.5}$ values (i.e., averaged over time in the ME) as to ambient hourly values.

Table 2 lists $r_{p/a}$, the ratio of personal to ambient $PM_{2.5}$, among different types of MEs. It was higher and more scattered on non-hazy days than the ratio on hazy days (1.71 ± 1.93 on non-hazy days vs. 0.85 ± 0.31 on hazy days). On both hazy and non-hazy days, the ratio was higher in open MEs than in enclosed MEs, and higher near sources than in receptor sites. The variabilty as measured by the standard error of $r_{p/a}$ is much higher on non-hazy days, confirming a greater variablity during relatively clean days. Enclosed receptor MEs with HVAC filtration (e.g., subway cars, lab and mall) had much smaller $r_{p/a}$ values (0.24–0.53) than other MEs over all 4 hazy days, while MEs with significant local sources, such as street food vendors, restaurants and subway stations (non-hazy days only) had the highest $r_{p/a}$.

DISCUSSION

Good Reproducibility of $PM_{2.5}$ Data after Gravimetric Correction (GC)

Airborne $PM_{2.5}$ concentration levels have been monitored worldwide using various optical based portable personal exposure sensors such as MicroPEM (EPA, 2014). The MicroPEM nephelometer measures $PM_{2.5}$ levels based on the intensity of the laser light reflected by the particles. Factors like particle photochemistry, geometric size, shape and relative humidity (RH) affect optical properties of particles and influence the performance of nephelometer (EPA, 1998; Fischer and Koshland, 2007). The RH values during our sampling period were always less than 90%, below which level the impact of RH is accounted for in the algorithms used by the MicroPEM (EPA, 2014). But the optical reflectivity of local PM sources can be very different from available standard reference particles (e.g., Arizona road dust) used for calibration by RTI, leading to the need for gravimetric correction (Jenkins *et al.*, 2004; Fischer and Koshland, 2007). The GC method allows real-time nephelometer values to be corrected for the mass weighted mean optical sensitivity during each deployment and also helps account for the mass of particles below 300 nm to which the MicroPEM nephelometer does not respond. After GC, MicroPEM measured $PM_{2.5}$ hourly concentrations are typically within 10% of NMEMC BAM-1020 hourly values for both non-hazy and hazy days, although individual data points can show larger differences. The BAM-1020, though recognized as equivalent to the U.S. federal gravimetric method for $PM_{2.5}$, can also have biases dependent on composition and weather conditions (Liu *et al.*, 2013), and as such the MicroPEM values when adjusted by the GC ratio determined during each deployment may be more reliable.

The much lower r_{GC} in hazy days than in non-hazy days indicates a difference in optical reflectivity, possibly due to variation in particle compostion between hazy and non-hazy days. The values of r_{GC} are also more variable for non-hazy days than for hazy days, consistent with the factor that many MEs were more easily impacted by nearby emission sources on non-hazy days. In contrast, extremely high ambient $PM_{2.5}$ levels on severe hazy days spreading over larger portions of the region, overwhelmed the influence from most local sources, and resulted in more similar composition across diverse MEs. The good agreement between corrected MicroPEM nephelometric and NMEMC monitored $PM_{2.5}$ concentrations, together with values and ranges of r_{GC} in hazy and non-hazy days, jointly

verify the ability of the GC to calibrate optical response during different deployments. In addition, side-by-side $PM_{2.5}$ data from two MicroPEM units also match better with each other after GC in both pattern and absolute value. Although the correction factor of 6.65 for unit 472N was abnormally high compared to our experience with many units suggesting an issue such as misalignment of optics, the excellent agreement with the second unit after gravimetric correction indicates the issue was corrected by the ratio and confirms the importance of having the filter weight for tying the average nephelometer response to reality.

Effect on $PM_{2.5}$ Level and $r_{p/a}$ from Nearby Emission Sources

Local $PM_{2.5}$ emission sources can be a significant factor influencing personal exposure in certain MEs, especially sources such as cooking and exhaust from motor vehicles (Wallace *et al.*, 1996; Kalaiarasan *et al.*, 2017). Among all MEs, street food vendors had the highest $PM_{2.5}$ levels (up to 1,100 μg m^{-3}) and highest $r_{p/a}$ values (up to 6.1 during non-hazy days). Gas-fueled open cooking (mainly stir frying, deep frying or teppanyaki grilling) in restaurants and the food-carts accounted for the extremely high personal $PM_{2.5}$ levels. Though the average personal $PM_{2.5}$ level in most enclosed MEs was lower than that in open MEs during both hazy and non-hazy episodes in this study, those enclosed MEs with known emission sources, like buses, restaurants and subway stations, have higher $PM_{2.5}$ than ambient levels at least during non-hazy days. For example, elevated $PM_{2.5}$ levels were observed during both hazy and non-hazy episodes in restaurant dining rooms with poor air handling. Many studies have reported elevated fine/ultrafine particle levels in restaurants and at home (Morawska *et al.*, 2013). There are two sources of fine particles from cooking: cooking fumes and fuel combustion smoke. The common Chinese cooking method of stir-frying results in heavy cooking fumes (Wang *et al.*, 2015), leading to a spike in $PM_{2.5}$ concentration from 27 to hundreds μg m^{-3} in a very short time (Zhao *et al.*, 2007), while pan-frying and deep-frying could generate even higher $PM_{2.5}$ concentrations (See and Balasubramanian, 2008).

Another prominent local source is vehicle emission, which resulted in high $PM_{2.5}$ levels in MEs like road, bus, and bus station. Vehicle exhaust contributed 29% and 24% to children's $PM_{2.5}$ exposure in the Chinese city of Tianjin in summer and winter, respectively (Zhang *et al.*, 2014). Among commuting methods influenced directly by vehicle emissions, personal exposure of $PM_{2.5}$ levels followed the trend: bus > walking along road > taxi > ambient in non-hazy days. This trend is similar to findings from previous studies in Beijing, Guangzhou and Aberdeen (Chan, 2002; Dennekamp, 2002; Huang *et al.*, 2012). The fact that $PM_{2.5}$ exposures were higher in buses than in taxis was partially due to the fuel type and vehicle type: diesel-fueled buses generated more fine particles than natural gas fueled taxies and frequent passenger movement in bus is associated with $PM_{2.5}$ re-suspension and concentration increase (Chan, 2002). Also, crankcases in certain buses have been shown to have direct emissions into the passenger cabin (Ireson, 2011). In non-hazy days, open MEs near busy roads like parks and schoolyards were also influenced by traffic sources.

Compared with other MEs without notable sources, personal $PM_{2.5}$ levels and $r_{p/a}$ in these two MEs were obviously higher. Construction could also contribute to $PM_{2.5}$ exposure due to increases in exposed soil and subsequent suspension, particle generation from

construction material, generator and truck emissions, and suspension during construction activities (Cao *et al.*, 2014). But the influence of construction on airborne $PM_{2.5}$ was not picked up by our short sampling periods near construction sites; more systematic sampling is probably needed that can take into effect different wind directions and speeds as well as catching episodic construction activities.

The ratio of personal exposure to ambient $PM_{2.5}$ levels ($r_{p/a}$) for open source MEs with emission sources is about 1 (0.98 in Table 2). MEs with emission sources would be expected to be higher than 1, but several possible reasons may complicate the result: 1) spatial variations in ambient $PM_{2.5}$ level exist within city, 2) local hourly EPA data are being compared to 1 min average data and thus there is error in this comparison, 3) r_{GC} for one filter/deployment is probably not enough to capture variable optical properties of the different sources of $PM_{2.5}$. The r_{GC} in different MEs could vary due to different PM sources, especially when a source has significant fraction of its mass below 300 nm, which the nephelometer does not respond to even though the gravimetric weight does include this fraction. It would be ideal to monitor each ME separately and get the r_{GC} for each of them. Unfortunately, our sampling design can only do one correction based on one filter for many MEs being visited/monitored. Therefore, some MEs could be over-/under-corrected to some extent.

Influential Factors in Enclosed MEs: Ventilation and the “Pig Pen” Effect

Ventilation could influence personal exposure in enclosed MEs under two situations: 1) limited natural ventilation inherent in the structure of certain MEs (e.g., MEs lying deep in underground with low ventilation rates) (Choi *et al.*, 2017); and 2) modern air handling systems with good filtration reduce particle levels in enclosed MEs.

Modern ventilation systems that include heating and air conditioning with filtration could further decrease the particle concentration in MEs (Hanninen *et al.*, 2005; Quang *et al.*, 2013; Ciuzas *et al.*, 2015). Aerosol levels in malls, labs and subway cars were comparatively low on hazy days compared with other enclosed MEs. The modern laboratory sampled in this study witnessed the lowest $PM_{2.5}$ exposure and lowest $r_{p/a}$ during both hazy and non-hazy days. This lab has 24/7 central air conditioning, no windows and the only door is well sealed. Furthermore, air conditioned subway cars in Nanjing are also low in $PM_{2.5}$. In contrast to subway cars in Seoul which lack mechanical ventilation/filtration systems and have high particle levels (Park and Ha, 2008), the Nanjing subway is relatively modern. Two subway lines in this study began service in 2005 and 2010 respectively, while the metro system in Helsinki started running in 1982, and the subway systems in London and New York have been in service for over 100 years.

The subway in Nanjing is powered by electricity, thus there is little to no combustion emissions during operation. However, wear and abrasion of steel wheels used in the subway have been shown to cause significant levels of $PM_{2.5}$ in other subway systems (Chillrud *et al.*, 2004; Nieuwenhuijsen *et al.*, 2007; Grass *et al.*, 2010; Martins *et al.*, 2016) as well as resuspension of particles due to passing trains (Qiao *et al.*, 2015), leading to higher levels of $PM_{2.5}$ in metro stations when compared with ambient levels (Praml and Schierl, 2000; Adams *et al.*, 2001). Consistent with this, the underground subway stations in Nanjing were

observed to have higher $PM_{2.5}$ levels than ambient levels during non-hazy days ($r_{p/a}$ = 2.49) but not in comparison to the high concentration seen outdoors on hazy days ($r_{p/a}$ = 0.80). The ground-level subway stations showed similar levels with ambient levels, confirming their high ventilation rate with outside air.

The levels in underground subway stations sampled during non-hazy days in this study are similar to those in Shanghai, Stockholm, and New York (Johansson and Johansson, 2003; Vilcassim *et al.*, 2014; Lu *et al.*, 2015), and they are all much higher than ambient levels on non-hazy days. The generation of particles by the friction of the subway cars on the subway rails plus the high population density in the subway stations could be partially responsible for the elevated $PM_{2.5}$ exposure levels due to resuspension of particles together with the combined "human cloud" of particles, which has been observed in several studies (Delfino *et al.*, 2004; Morawska *et al.*, 2013). Clothes shedding of fibers from dense population also contribute to short-term particle emission (You *et al.*, 2013). Similarly, high $PM_{2.5}$ exposure levels were observed in underground malls and hospitals, which were full of people during peak hours. When ambient particle concentrations are very low, the enrichment in exposure from resuspension and "human cloud" may even surpass the decreasing influence from ventilation. When background $PM_{2.5}$ concentration is lower than 25 μg m^{-3} in Foshan and Helsinki, $PM_{2.5}$ levels in subway cars and in ambient are almost identical despite the usage of ventilation systems (Aarnio *et al.*, 2005; Wu *et al.*, 2013).

Subway cars sampled in this study have the lowest $PM_{2.5}$ levels and $r_{p/a}$ among commuting MEs, demonstrating the good air handling in the subway cars that efficiently remove PM from the cars. Due to the same reason, personal $PM_{2.5}$ levels in subway cars are lower than personal levels in both types of subway stations (underground and ground level subway station).

Exposure in Hazy and Non-Hazy Days

Across multiple hazy and non-hazy days, personal $PM_{2.5}$ exposure levels in most MEs show significant correlation with ambient hourly $PM_{2.5}$ levels, in spite of influence from local sources and other characteristics of various MEs (e.g., ventilation). This demonstrates that ambient air has pronounced influence on various MEs, consistent with findings in other studies (Janssen *et al.*, 2000; Kinney *et al.*, 2011; Saraga *et al.*, 2017). Meanwhile, good air handling can help reduce $PM_{2.5}$ levels during both hazy and non-hazy days.

Despite the influence of ambient levels on the personal exposures observed in most MEs on both hazy and non-hazy days, it was clear that the ambient level has less pronounced influence on non-hazy days when local sources have a larger impact on overall personal exposures. The personal sampling strategy used in this study suggested that MEs have $PM_{2.5}$ exposure levels higher than the ambient levels on non-hazy days but lower on hazy days (Table 2, Fig. 4). MEs containing or exposed to local sources have more variable $PM_{2.5}$ exposure levels than receptor MEs on non-hazy days. Because of the dominant influence of ambient air on MEs during hazy days, their $PM_{2.5}$ levels and $r_{p/a}$ ratios are also less diverse on hazy days compared to non-hazy days. These observations jointly reflects that MEs, especially those with emission sources, are particle contributers to the personal $PM_{2.5}$

exposures on non-hazy days; in contrast, on hazy days, the ambient $PM_{2.5}$ levels overwhelm or mask many of the local sources for most but not all MEs.

When ambient $PM_{2.5}$ level is relatively low during non-hazy days, spending more time outside or in well-ventilated MEs can help decrease exposure compared to time spent near point sources. Meanwhile, because of the penetration of ambient PM into the majority of MEs on hazy days, $PM_{2.5}$ exposure levels in most enclosed MEs were increased, thus, there is no real good shelter during the haze episodes, except in those MEs with good air filtration (e.g., enclosed malls). Given most people spend the majority of their time indoors, a good air handling system with HEPA filter that can remove fine particles faster than air exchange with ambient air would reduce personal exposures.

This study had some limitations. Ideally, synchronous monitoring of various MEs over a longer period will help define the $PM_{2.5}$ pattern in these MEs and allow hourly averages to be compared to the hourly average data available from the NMEMC sites. This study was designed to investigate quite a few MEs in several field trips, thus investigator's activities and time spent in each activity can be different from those of a average person going about their normal routine. Furthermore, several enclosed MEs with air handling systems that can filter out particles had low $r_{p/a}$ ratios, but this simple approach of comparison to ambient values cannot rule out whether emission sources existed in these MEs. For example, the low $r_{p/a}$ ratios seen in the mall MEs hide the resuspension clouds in restauarants that exist in the mall areas.

CONCLUSIONS

The use of miniaturized optical sensors for $PM_{2.5}$ need regular calibration (e.g comparison to gravimetric filters or reference site data) to take into effect changes in optical properties of spatially and temporally varied PM sources. On both hazy and non-hazy days, $PM_{2.5}$ exposures in microenvironments (MEs) were affected by ambient $PM_{2.5}$ levels and strong local point sources such as street food vendors. However personal $PM_{2.5}$ exposures relative to ambient hourly levels showed markedly different relationships on hazy and non-hazy days. On hazy days, most MEs displayed lower personal exposure levels than ambient levels while on non-hazy days personal $PM_{2.5}$ exposures were higher than ambient levels in many MEs. Street food vendor MEs with cooking emissions had the highest $PM_{2.5}$ exposure levels on both hazy and non-hazy days and should be avoided by people sensitive to $PM_{2.5}$ exposure. The Nanjing subway stations are a location of increased exposure on non-hazy days and lowered exposure on hazy days as compared to ambient levels, while the filtration system in Nanjing's subway cars lowers $PM_{2.5}$ exposures appreciably below station levels on both hazy and non-hazy days.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

This study was supported by the National Natural Science Foundation of China (41230526). Support from the China Scholarship Council (201406190117) has made possible for graduate student Ting Zhang form Nanjing University to be trained in Columbia University. Additional support was provided by NIEHS (ES009089 and 1S10OD016219).

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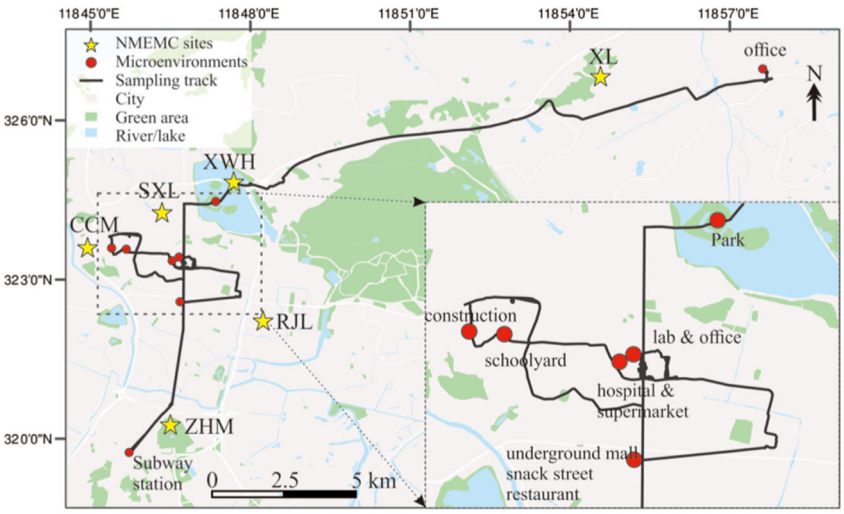

Fig. 1.

Sampling route for personal exposure in various MEs in Nanjing. Abbreviations (CCM, XL etc.) are the names of different NMEMC monitoring sites. NMEMC represents Nanjing Municipal Environment Monitoring Center.

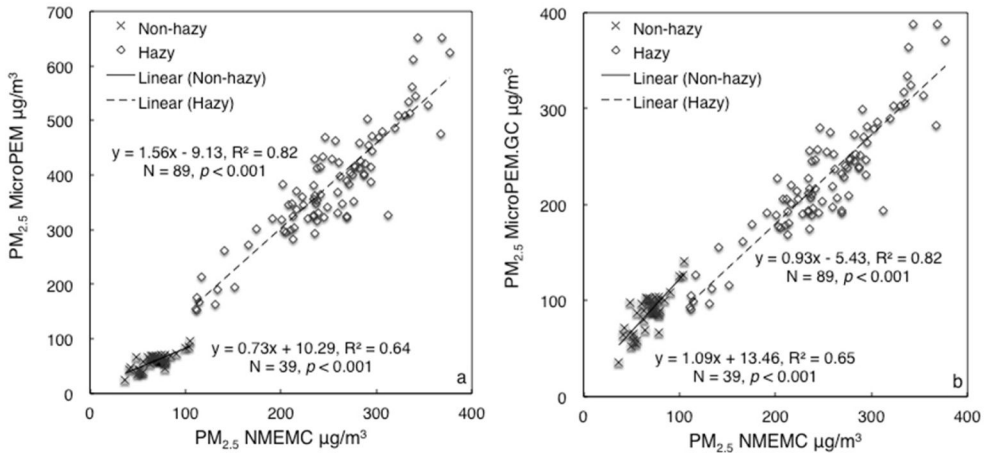

Fig. 2.

Fixed site side-by-side comparison between NMEMC and MicroPEM for hourly averages of $PM_{2.5}$ concentration before (Fig. 2(a)) and after (Fig. 2(b)) gravimetric correction (GC).

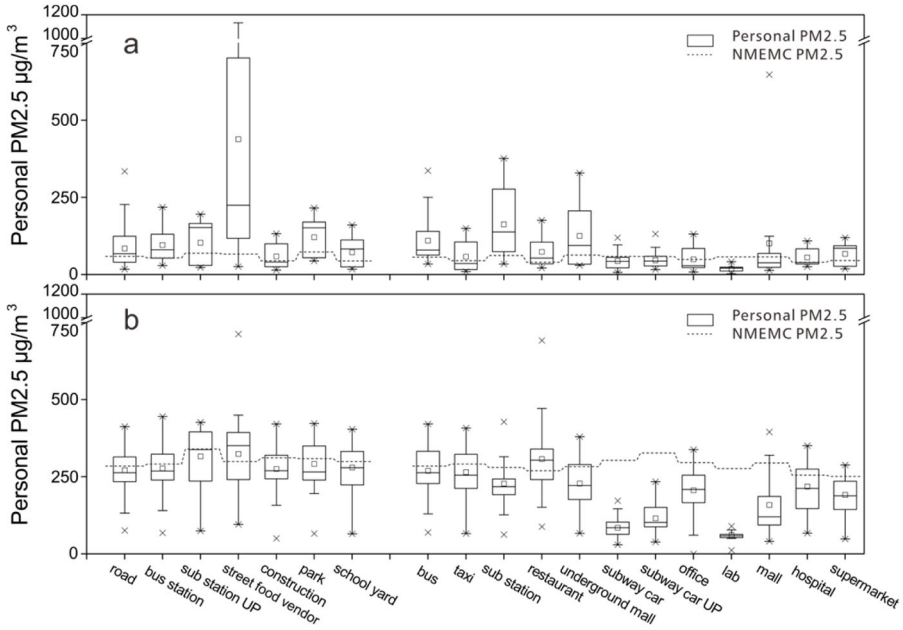

Fig. 3.

Personal exposure (minute average) levels in various MEs on non-hazy (Fig. 3(a)) and hazy (Fig. 3(b)) days and their comparison with EPA monitored $PM_{2.5}$. [a] Bars from top to bottom, respectively, represent 90%, 75%, 50%, 25% and 10% of $PM_{2.5}$ values, the stars are max/minimum $PM_{2.5}$ values, while the box in the middle represents average $PM_{2.5}$ concentrations in separate MEs. [b] Sub station and sub station UP represent underground and ground-level subway stations, respectively, while subway and subway UP represent underground and ground-level subway cars. [c] Similar sub-MEs have been goruped. Open MEs are grouped on the left while enclosed MEs are grouped on the right.

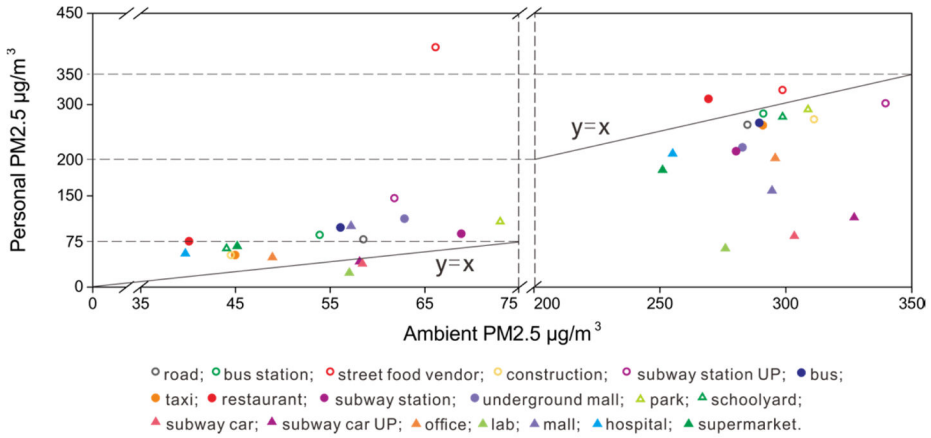

Fig. 4.

Comparison between personal exposure to $PM_{2.5}$ and NMEMC monitored ambient $PM_{2.5}$ concentration. [a] Solid and hollow dots represent enclosed and open MEs, respectively, while round and triangular dots represent source and receptor MEs, respectively. [b] The solid line is the 1:1 line. [c] Personal $PM_{2.5}$ data of each ME is the average of all the minute data measured in that ME over all hazy or non-hazy days.

Table 1

Detailed information of each sub-ME.

Sub-MEs	Classification	HVAC	Traffic	Comments
road 1	Open source	None	Heavy	In suburb; extends along a viaduct, morning
road 2	Open source	None	Light	Near park, not far from busy traffic, afternoon
road 3	Open source	None	Heavy	Between high buildings, afternoon
road 4	Open source	None	Light	Inside a hotel yard in downtown, night
bus 1	Closed source	On	Change	Traffic light to heavy; window closed, door open intermittently, morning
bus 2	Closed source	On	Heavy	Window closed, door open intermittently, afternoon
bus 3	Closed source	On	Light	Window closed, door open intermittently, night
bus station 1	Open source	None	Light	Very little traffic, morning
bus station 2	Open source	None	Heavy	Near a viaduct, morning
bus station 3	Open source	None	Heavy	Near crossroad, afternoon
bus station 4	Open source	None	Light	Near hotel, night
taxi 1	Closed source	On	Heavy	Downtown, noon
taxi 2	Closed source	On	Light	Downtown, afternoon
subway car 1	Closed receptor	On	Light	Downtown, morning, run on underground tunnel
subway car 2	Closed receptor	On	Heavy	Downtown, noon, run on underground tunnel
subway car 3	Closed receptor	On	Light	Downtown, night, run on underground tunnel
subway car UP 1	Closed receptor	On	Heavy	Downtown, noon, run on above ground bridge
subway car UP 2	Closed receptor	On	Light	Surburb, night, run on above ground bridge
subway station 1	Closed source	None	Light	Morning, underground station
subway station 2	Closed source	None	Heavy	Noon, underground station
subway station 3	Closed source	None	Light	Night, underground station
subway station UP	Open source	None	Heavy	Between surburb & downtown, noon, ground level
Street food vendor	Open source	None		With open cooking and small restaurants, noon
construction	Open source	None		Trucks fueling diesel; not far from heavy traffic, afternoon
park	Open receptor	None		Big lake and trees; few people; near busy roads, morning
underground mall	Closed receptor	None		Connects mall and subway station; dense population, small restaurants & clothes stores, noon
schoolyard 1	Open receptor	None		Near construction site and busy road; few people; trees, afternoon
schoolyard 2	Open receptor	None		Near busy-traffic road; dense population; trees, afternoon
restaurant	Closed source	None		Small restaurants near school, full of students
office 1/4	Closed receptor	On		Window closed and door open sometimes. Office 1 is early in morning with few people; while office 4 is at night with denser population
office (2&3 same)	Closed receptor	On		Big office with few people, window closed and door open, afternoon
lab	Closed receptor	On		Small well-sealed lab, with generally nobody inside, afternoon
mall	Closed receptor	On		Air conditioner only on during hazy day sampling; few people, afternoon
hospital	Closed receptor	On		Dense population, doors were always opened
supermarket	Closed receptor	None		Few people, not well isolated with outside

Table 2

Averages of personal exposure concentrations and average ratio of personal exposure to ambient $PM_{2.5}$ levels ($r_{p/a}$) in different kinds of micro-environments.

	Haze				Non-haze				Haze/Non-haze
	$PM_{2.5}$	sd.$PM_{2.5}$			$PM_{2.5}$	sd.$PM_{2.5}$			
ME classification	**μg m^{-3}**	**μg m^{-3}**	**$r_{p/a}$**	**sd.$r_{p/a}$**	**μg m^{-3}**	**μg m^{-3}**	**$r_{p/a}$**	**sd.$r_{p/a}$**	**$PM_{2.5}$**
ALL	243	91	0.85	0.31	103	147	1.7	1.9	2.4
Open	283	74	0.98	0.25	130	194	2.0	2.5	2.2
Closed	211	91	0.76	0.32	78	78	1.4	1.1	2.7
Source	275	77	0.97	0.27	127	178	2.1	2.3	2.2
Receptor	197	90	0.69	0.30	65	64	1.2	0.9	3.0
open_source	282	76	0.98	0.26	139	218	2.2	2.8	2.0
open_receptor	286	66	0.97	0.24	100	58	1.6	0.7	2.9
close_source	265	77	0.96	0.28	109	85	1.9	1.1	2.4
close_receptor	172	79	0.61	0.27	53	62	1.0	0.9	3.2