Assessment of Integrated Personal Exposure to Particulate Emissions in Urban Microenvironments: A Pilot Study

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Abstract

City dwellers’ personal exposure to PM is influenced by numerous daily activities in multiple indoor and outdoor micro-environments (MEs). This study was designed to assess integrated personal exposure to PM across urban MEs together with the recording of individual time-activity patterns. We characterized simultaneously the personal exposure to PM₂.₅, black carbon (BC) and ultrafine particles (UFPs) in the Central Business District (CBD) area of Singapore. In addition, we quantified the Lung Deposited Surface Area (LDSA) concentration, which is an indicator of potential health impacts of UFPs. The field study was conducted over a 7-km walking route to identify air pollution hotspots. Subsequently, the personal exposure to PM₂.₅, BC and UFPs was measured at five selected hotspots for a duration of 1 hour, and across indoor and outdoor MEs under diverse daily human activities for 24 hours. The PM concentrations were found to vary considerably in both space and time at the CBD area. During the 1-hour personal exposure measurement, extremely high concentrations of PM₂.₅ (215 ± 129.5 µg m⁻³ and 36.4 ± 12.5 µg m⁻³) and BC (20.9 ± 10.4 µg m⁻³ and 18.1 ± 12.0 µg m⁻³) were observed at a temple and at a bus stop while elevated number concentrations of UFPs (320.8 ± 131.1 x 10³ # cm⁻³) and LDSA (564.6 ± 276.5 µg² cm⁻³) were measured at a food court. Potential health risk was estimated, which suggests that the continued inhalation of elevated levels of PM₂.₅ emitted from combustion sources is likely to lead to long-term adverse health effects among the exposed individuals. Overall, we provide insights into an individual's total exposure to PM based on time-activity patterns. The outcome of this work provides a scientific basis for development of air pollution control measures to mitigate personal exposure to PM at the city scale.

Keywords: Aerosol Sampling and Transport, Air Quality, Personal Exposure.

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INTRODUCTION

Many epidemiological and toxicological studies conducted worldwide indicated that human exposure to high levels of airborne particulate matter (PM), particularly those particles with aerodynamic diameters less than 2.5 µm (PM$_{2.5}$), can have adverse human health effects (Pope et al., 1995; WHO, 2013; Kim et al., 2015). The continued exposure to PM is ranked as one of the leading causes of increased morbidity and mortality worldwide (Gakidou et al., 2017). In recent years, the focus of several air pollution studies has shifted from PM$_{2.5}$ mass concentration to more specific metrics such as the mass concentration of black carbon (BC) and ultrafine particles (UFPs) due to their strong association with negative health effects (e.g., Heal et al., 2012; Wu et al., 2015). BC, a light-absorbing component of PM, is a good indicator of exposure to harmful particulate-bound chemical substances such as polycyclic aromatic hydrocarbons (PAHs) emitted from incomplete combustion of fossil fuels. In densely populated areas, the contribution of urban traffic to BC and UFPs is often considered to be more important than that of other fossil fuel combustion activities, including industry (Liu et al., 2005). UFPs (PM with aerodynamic diameter less than 0.1 µm) may be more toxic than PM$_{2.5}$ due to their enhanced pulmonary deposition efficiency, much higher surface area (in term of equal mass basis) and a greater proportion of volatile organic compounds in them (Delfino et al., 2005).

Fixed air quality monitoring stations, equipped with often expensive and bulky automated instruments, are widely used to measure PM levels with sufficient time-resolution to study the
temporal variability of air pollutant concentrations. Measured PM concentrations are required to be below the international air quality benchmarks such as WHO (the World Health Organization) guidelines to protect public health. However, recent studies have indicated that even at a concentration below current air quality regulations (e.g., Environmental Protection Agency-EPA regulations), health effects persist (Wellenius et al., 2012; Rice et al., 2013). Furthermore, the concentrations of air pollutants are subject to high variability in space and in time depending on variations of emission sources, meteorology, land use and topography (Wilson and Zawar-Reza, 2006). Therefore, fixed air quality monitoring stations do not provide spatially representative PM data for health risk assessment (Kaur et al., 2005). Mobile and cost-effective PM instruments have been recently developed to overcome these problems. Significant improvements in sensing techniques have enabled researchers to investigate personal exposures to a variety of airborne contaminants in a high temporal and spatial resolution measurement (Vincent, 2012; Koehler and Peters, 2015). Accurate assessment of human exposure to PM including PM$_{2.5}$, UFPs, and BC is, therefore, crucial for a proper determination of possible links between particulate air pollution in cities and human health effects.

On average, people spend less than 8% of their time in urban transport environments, but the urban transport contributes to over 20% of their daily integrated exposure, leading to many adverse health effects (Dons et al., 2012). Therefore, most of the PM studies using portable
devices have focused mainly on the human exposure while in transit and on the influence of different transport modes on exposure outcomes (e.g., Chan et al., 2002; Moreno et al., 2015; Good et al., 2016; Rivas et al., 2017; Shirmohammadi et al., 2017). Selected studies related to personal exposure to PM in transport micro-environments (MEs) using portable monitors are summarized and given in the SM-Table S3. In recent years, active modes of transport such as walking and cycling are receiving increased attention to reduce car dependence. Increased levels of physical activity associated with active modes of transport lead to potential public health benefits (Reynolds et al., 2010). However, some studies showed higher exposure to PM$_{2.5}$ during walking compared to other commuting modes because of emissions of PM in close proximity, higher inhalation parameters (such as respiratory rate and volume) and longer commuting time (Liu et al., 2015; Betancourt et al., 2017). Apart from transport, city dwellers’ personal exposure to air pollution is also influenced by other activities in multiple indoor and outdoor MEs on a daily basis. The characteristics and contributions of these MEs to the daily personal exposure in urban areas are not well understood. In addition, the personal exposure depends on physical health factors (e.g., gender, age, and body weight) and time-activity patterns of individuals. The integrated assessment of health risks associated with inhalation of PM across diverse MEs over 24-hours exposure needs to be examined on a day-to-day basis. In this regard, using a mobile
platform paired with real-time PM monitors and a global positioning system (GPS) tracking is a powerful approach to accurately characterize individuals’ exposure.

In this pilot field study, two university students (non-smoking individuals) assessed their personal exposure to PM$_{2.5}$, UFPs, and BC using portable devices and a GPS in the Central Business District (CBD) area of Singapore. In addition, they quantified the lung Deposited Surface Area (LDSA) concentration, which is considered to be an appropriate indicator of potential health impacts (Ntziachristos et al., 2007; Albuquerque et al., 2012). Elevated levels of LDSA are considered to be indicative of adverse health effects such as lung airway inflammation (Buonanno et al., 2013) or acute respiratory visits to an ambulatory medical facility (Sinclair and Tolsma, 2004). The students walked around a range of outdoor and indoor MEs as part of their daily routines. The specific aims of this work were: (1) to characterize the spatio-temporal patterns of the personal exposure to PM$_{2.5}$, UFPs, and BC and identify pollution hotspots; (2) to quantify the emission profiles of these pollutants in the identified hotspots; and (3) to estimate potential health risks associated with PM$_{2.5}$ inhalation over the 24-hour daily exposure. The city-state of Singapore was selected for this study as it is one of the most densely populated cities in the world and has a number of sources of airborne particles due to rapid urbanization and industrialization, large traffic volume, and frequent occurrence of transboundary smoke haze episodes (Kalaiarasan et al., 2009a, b; Huang et al., 2016; Sharma and Balasubramanian, 2017).
The Singapore government is currently promoting active mobility among commuters (i.e. “Walk Cycle Ride” modes) as part of its ongoing efforts towards sustainable development. The preliminary findings obtained can be useful for citizens to make informed choices to reduce their PM exposures while being involved in a range of outdoor and indoor activities on a daily basis. The results will also be useful for the local government to identify and implement effective mitigation measures to lower people’s exposure to PM at the city scale, contributing to improved urban air quality and enhanced health benefits.

**METHODS**

*Study location and design*

The personal exposure assessment to PM was conducted in the CBD area of Singapore between October 2017 to February 2018. This area attracts tourists and contains multiple street canyons amidst high-rise buildings representative of the urban landscape. The average traffic volume entering the CBD area is 300,400 vehicles per day, and the average speed of vehicles in the area is 28.9 km/h during peak hours (data obtained from Govtech-Singapore (2018)). Others than traffic, major sources of air pollution in this area comprise commercial cooking activities in restaurants and food courts, construction activities, smoking in the open area and incense or joss paper burning in temples. Fig. 1 provides an aerial overview of the study area.
The field study was divided into three separate components: the first component of the study was carried out over a 7-km walking route around the CBD area to study both temporal and spatial variations of PM concentrations. The second component quantified PM exposure levels in five pollution hotspots. The third one measured the 24-hour personal PM$_{2.5}$ exposure under a range of diverse daily human activities across indoor and outdoor MEs.

**Sub-study 1: 7-km walking route measurements**

A 7-km walking route around the CBD was chosen that covered many land uses and different road categories (highways, major arterial roads, minor arterial roads, primary access, and local access). The students walked over the pedestrian footpaths along the designated route across diverse indoor and outdoor MEs. Exposure pathways are shown in Figure 1. The study was conducted on typical weekdays (i.e., Tuesday-Thursday) and weekends (i.e., Saturday-Sunday) during the morning (7:30-10:00), afternoon (11:30-14:00) and evening (18:00-20:30). These time periods were selected to investigate the personal exposure under different levels of human activities in the CBD. In total, 19 personal exposure assessments were conducted over the 7-km route (~ 2.5-hour per walking trip) around the CBD area. A 7 hectares public park located to the east of the study site was selected as the background location (BG) which is relatively clean compared to the CBD due to the absence of combustion-related sources. Personal exposure data were also collected in the BG location with the same protocol as used in the CBD area.
Sub-study 2: 1-hour personal exposure measurements at selected air pollution hotspots

Five diverse outdoor and indoor MEs (including a smoking area, a temple, a food court (hawker center), a bus top, and a traffic intersection) were selected (based on the observations made in the sub-study 1) to better understand the typical emission profiles in response to the nature and intensity of human activities in the specific locations. The characteristics of these MEs are described in Table 1. Totally, 28 monitoring studies were conducted in the MEs over a duration of 1 hour on weekdays during morning, afternoon and evening time periods.

Sub-study 3: 24-hour personal exposure measurements

PM$_{2.5}$ mass concentrations were only considered in the 24-hour personal exposure assessment as they are routinely measured by the National Environment Agency (NEA) as one of the six criteria air pollutants at fixed monitoring stations throughout Singapore. To determine the inhaled dose of PM$_{2.5}$, 24-hour personal exposure studies were conducted for 4 days. For this work, a portable PM$_{2.5}$ monitor was used throughout the daily routine of the students for the entire day. During each sampling day, the student visited the common MEs as in the previous sub-studies (i.e., bus stops, traffic intersections, and the hawker center) and followed similar time activity patterns over the four days to maintain constancy. The students noted down their whereabouts in a time-activity diary, and a GPS receiver was used to identify the specific MEs visited at different times of the day.
Instrumentation and quality assurance

The PM measurements included (1) the mass concentration of PM$_{2.5}$ ($\mu$g m$^{-3}$), (2) the number concentration of UFPs (Np) (# cm$^{-3}$), (3) Lung Deposited Surface Area (LDSA) ($\mu$g$^2$ cm$^{-3}$), and (4) the mass concentration of black carbon (BC) ($\mu$g m$^{-3}$). The mass concentration of PM$_{2.5}$ was measured using a light-scattering laser photometer Sidepak (AM520, TSI, USA). Np and LDSA were measured using a handheld particle counter Discmini (Testo, Germany). The mass concentration of BC was measured by a Micro-Aethalometer (AE51, Aethlabs, USA). Also, relative humidity RH (%) and temperature T ($^o$C) were measured using temperature and humidity loggers (BG-Log-TempRH, Acez, Singapore). A GPS receiver was used to track the movement and exposure of the subjects across the MEs during the study period. Table S1 (Supplementary Material – 1) provides the characteristics of each device.

Sidepak (AM520) is a real-time laser photometer that detects PM$_{2.5}$ and quantifies its mass concentration based on the light scattering of aerosols. The device was operated with a flow rate of 1.7 L min$^{-1}$ which was verified before the sampling with a TSI flowmeter (4100, USA). The SidePak was factory-calibrated based on the standard test protocol (ISO12103 – A1 Arizona Test Dust). To get reliable PM$_{2.5}$ mass concentrations that are representative of the specific sampling locations, the Sidepak was further calibrated with a gravimetric sampler (MiniVol, Airmetrics, USA). Calibration factors of 0.22 and 0.19 were applied to the PM$_{2.5}$ concentrations obtained.
from the two Sidepak units to get the final PM$_{2.5}$ concentrations. Detailed steps involved in the calibration of the Sidepak are described in the SM- 2.

Discmini is a handheld particle counter that operates based on the diffusion charging principle. Particles are charged by a unipolar diffusion charger and the current is measured by an electrometer as the particles pass through the designed diffusion and filter stage in the instrument chamber. The current difference between the two stages is measured to obtain the total UFP particle number concentration ($N_p$) and the average particle size. The Discmini also measured alveolar lung deposited surface area (LDSA) which is defined as the particle surface area weighted by the deposition efficiency of airborne particles in the alveoli region based on a model proposed by the International Commission for Radiological Protection (ICRP, 1994). Very good agreements were observed when the performance of Discmini was compared to that of condensation particle counters in previous studies (Asbach et al., 2012; Meier et al., 2013). An impactor provided by the manufacturer was always equipped at the the inlet of the Discmini to prevent PM larger than 0.7 μm entering into the device.

The BC concentration was quantified by a micro-aethalometer (AE51), which measures the attenuation of light transmitted through airborne particles that are continuously collected onto a Teflon filter. In present study, filter strips were changed every 5-hour of air sampling to minimize filter loading effects. Before air sampling, the two microAeth units were calibrated side by side
using a TSI flowmeter (4100, USA), following the auto-flow calibration procedure recommended by the manufacturer. The micro-aethalometer was operated with the flow rate of 150 mL min\(^{-1}\). One known problem in AE51-based measurements is that at high logging intervals or very low BC values, the incremental change in light absorption may be affected by the instrumental noise, leading to negative or unchanging values in the recorded data (Hagler et al., 2011). To resolve this issue, the raw BC was corrected using the Optimized Noise reduction Averaging (ONA) algorithm tool (Hagler et al., 2011) with delta attenuation thresholds of 0.01-0.05. The second issue associated with the operation of AE-51 is the filter loading effect, which affects BC measurements by the decreasing instrumental sensitivity associated with the increased filter loading. To account for this issue, BC concentrations were adjusted using an empirical correction factor reported by Kirchstetter and Novakov (2007) based on the optical attenuation (ATN) measured by the instrument. The performance of AE51 was further checked by collocating it with Aethalometer AE33 (Magee Scientific, USA) and doing BC measurements over a period of 24 hours. A correction factor based upon the slope of the linear regression equation obtained from the inter-comparison study was finally applied to the data measured by the microAeth (the data are shown in Figure S2).

Sidepak and Discmini were synchronized and programmed to capture data with a time resolution of 1 s. The Micro-Aethalometer AE51 was programmed to capture BC data with a
time-averaging of 10 s, whereas temperature & relative humidity and GPS data were collected at
30 s and 1-minute readings, respectively as the highest logging intervals set up by the
manufacturers. All portable devices were carried in a backpack, and air sampling inlets were
placed within the breathing zone at approximately 1.5 m above the ground level. All the
measurements were conducted between October 2017 and February 2018. During this time
period, Singapore experienced strong winds with speeds in the range of 7 to 9 m/s and wet
conditions on several occasions (Meteorological Service Singapore) which help in the efficient
dispersion of air pollutants and their removal from ambient air through wet deposition. It should
be noted that all the personal exposure measurements were conducted on hot and sunny days with
clear skies to protect the operational integrity of sensors and to avoid the potential measurement
issues related to the PM concentrations, caused by high relative humidity and rainfall. A duplicate
set of Sidepak, Discmini, Micro-Aethalometer was operated at the background site during the
collection of the mobile measurements.

Before each day of air sampling, all instruments were clock synchronized using a laptop
connected to the internet to ensure that the time stamp was consistent across all instruments.
When arriving at the measurement site, instruments were zeroed with HEPA filtered air and left
to log data for about 5-10 minutes before the actual sampling. Collected data were imported,
analyzed mainly using R (R Core Team, 2018) and Microsoft Excel.
Reference fixed monitoring sites

1-hour average PM$_{2.5}$ data were obtained from the air quality monitoring station in the central part of Singapore, maintained by the National Environmental Agency (NEA) website (http://www.nea.gov.sg), and analyzed to determine the difference in the assessment of exposure to PM$_{2.5}$ based on measurements at a fixed location versus those across diverse MEs with mobile devices. In addition, as BC is not routinely monitored by NEA, a multiple-length Aethalometer (Model AE33, Magee Scientific, USA), located at the Atmospheric Research Laboratory in the National University of Singapore (NUS) campus, was used for the routine measurement of BC. A flow rate of 3 L min$^{-1}$ was used with a sampling time base of 60 seconds. The determination of the BC concentration is based on the measurement of light absorption on a filter loaded with aerosols. The Model AE33 Aethalometer is advantageous in the sense that it can reduce the filter loading effect by utilizing a ‘dual spot’ measurement method (Drinovec et al., 2015).

Exposure assessment

The integrated inhalation dose of PM$_{2.5}$ was calculated as the sum of exposure for all MEs (n) visited by the students over a specific period of time (t) with the inhalation rate (IR) (Eq. (1)). IR was assumed to be 1.3 x 10$^{-2}$ m$^3$. min$^{-1}$ (= 0.78 m$^3$ hour$^{-1}$ = 18.72 m$^3$ day$^{-1}$) selected from the US EPA’s Exposure Factor Handbook -Table 6.17 (Moya et al., 2011) for young adults aged 21-31 years with light activity levels.
The integrated inhaled dose of PM$_{2.5}$ based on measurements using the Sidepak was then compared to those obtained from fixed ambient air monitoring stations maintained by NEA for the respective days. Moreover, the toxicological health risk associated with chronic exposure to PM$_{2.5}$ was quantified as the risk quotient (RQ). The steps involved to estimate the Lifetime Average Daily Dose (LADD) and toxicological RQ to PM$_{2.5}$ are described in the SM – 4.

**RESULTS AND DISCUSSION**

*General characteristics of PM exposure around 7-km walking around Central Business District*

The observed personal PM exposure concentrations varied considerably in both space and time at the CBD area. Fig. 2 presents the variations in PM$_{2.5}$, BC, Np, and LDSA exposure concentrations observed during weekdays and weekends in the study area. The statistical data are summarized in the SM - Table S2. During weekdays, the mean PM$_{2.5}$ concentrations were 46.2, 35.3, and 22.2 µg m$^{-3}$ in the morning, afternoon, and evening hours, respectively; the overall
mean concentration was 32.5 µg m\(^{-3}\). Our study reveals that the PM\(_{2.5}\) concentrations were the lowest in the study area during the evening hours, which is attributed to changes in the traffic composition and in PM sources during the day. Specifically, there was a large volume of buses, taxis, and cars during morning and afternoon hours while a relatively high number of trucks and utility vehicles were during the afternoon hours. Additionally, the nature and intensity of other PM sources such as construction activities and incense burning were carried out only during the day. The changes in the tropical weather conditions during the day also played an important role in affecting the PM\(_{2.5}\) concentrations in the study area. No significant difference was found in the PM\(_{2.5}\) exposure between weekdays and weekends at the CBD area except in the morning session. Vehicular emissions contributed more to the ambient PM on weekdays in the morning (46.2 µg/m\(^3\)) as compared to the weekends (34.8 µg m\(^{-3}\)).

All the PM concentration values were much higher than those measured in the urban background due to the presence of many sources of air pollution along the measurement route. For example, the mean PM\(_{2.5}\) concentrations were 34.8, 33.2, and 24.7 µg m\(^{-3}\) on weekends in the morning, afternoon, and evening hours compared to 10.2, 9.9, and 11.9 µg m\(^{-3}\) at the background site, respectively. In the case of BC, the mean concentration observed in the studied area was 8.0 µg m\(^{-3}\) (about 24.5% contribution to PM\(_{2.5}\)) at the CBD area compared to 3.2 µg m\(^{-3}\) measured at the background location. High concentrations of BC indicate the dominant contributions of the
local traffic and incense burning activities at the study area. In present study, UFPs were quantified by particle number concentration Np and LDSA. In general, the measured values were 6.4 x 10^3 # cm^{-3} (ranging between 4.28 and 8.23 x 10^3 # cm^{-3}) and 125.5 µg^2 cm^{-3} (ranging between 71.6 and 179.7 µg^2 cm^{-3}) of Np and LDSA concentrations observed at the CBD area. These UFP concentrations were about 4 times higher than those measured at the background location. As the commercial areas (e.g., hawker centers) become more active towards weekends, higher UFP levels were observed during weekends compared to weekdays (as shown in Fig. 2 (c, d) and the SM -Table S2).

The distributions of PM concentrations are highly skewed, especially for UFPs and LDSA as shown in Fig. 2, suggesting a high spatiotemporal variability of PM during the field trips. Also, all mean values of particle metrics were higher than the median due to the presence of exceedances (i.e., outliers) that increased the mean but did not affect the median. These outliers were mainly associated with incense burning emissions and/or exhaust plumes of on-road vehicles under acceleration or deceleration. Elevated concentrations of PM (e.g., PM_{2.5} > 150 µg m^{-3}, BC > 10 µg m^{-3}, Np > 50 x10^3 # cm^{-3}, LDSA > 100 µg^2 cm^{-3}) were observed during most of the personal exposure measurements. Although these spike concentrations are temporal (i.e., short term exposure), they still contribute to personal pollution exposure to trigger the onset of
myocardial infarction (Peters et al., 2001; Peters et al., 2004) or systemic and pulmonary inflammatory responses (Salvi et al., 1999), so we retained these concentration values.

The representative exposure pathways within CBD are presented in Fig. 3 and the SM – Fig. S2 for the morning session. Both time series (Fig. 3) and spatial PM maps (the SM – Fig. S3) show the actual exposure to PM under different pollution conditions; the data were aggregated for each 1 min on representative days. PM$_{2.5}$ concentrations were generally homogeneous (Fig. 3(a)) compared to the heterogeneous spatial distributions of BC (Fig. 3(b)) and UFPs (Fig. 3(c, d)) over the length of the route. The lack of spatial variations on PM$_{2.5}$ can be explained by the relatively long atmospheric residence time of fine particles (Thai et al., 2008). In contrast, UFPs showed a heterogeneous spatial distribution due to the occurrence of nucleation and coagulation processes under ambient tropical weather conditions (e.g., high relative humidity and temperature). Their concentrations can drop to their background levels in open terrains approximately 300 m from a source (Klompmaker et al., 2015). BC with its small particle size (i.e., nano-diameter range) showed a spatial variation similar to that of UFPs, but it was much larger than that of PM$_{2.5}$ (WHO, 2012). Significant peaks in PM concentrations measured along the route are shown in Fig. 3. The highest concentrations of PM$_{2.5}$ and BC were observed at the temple and traffic-influenced locations, while the hawker center and the temple had the extremely high concentration of UFPs.
Ambient temperature and relative humidity were also continuously measured (Table.S2). The temperature ranged between 25.6°C to 35.3°C during the measurement days, and the mean temperature was 30.0 ± 2.0 °C. Among the five selected MEs, the marked temperature values were recorded at the temple and traffic-related locations with mean values of 33.2°C and 32.3°C (data not shown). The relative humidity generally ranged between 50% and 85% in the study area, and the mean value was 68.6 ± 7.9%.

**1-hour personal exposure measurements at five air pollution hotspots**

To better understand the typical emission profiles of PM in response to the nature and intensity of human activities in specific locations, 1-hour personal exposure measurements were conducted at five selected MEs three times a day (i.e., morning, afternoon and evening, Fig. 4). These measurements were repeated twice at each ME on different days. The measured exposure concentrations of PM$_{2.5}$, BC, Np, and LDSA were much higher than those measured at the fixed monitoring stations (the NEA air quality monitoring station and the NUS atmospheric research station). When Sidepak was used to measure the 1-hour PM$_{2.5}$ exposure at the bus stop (ME4), the measured concentrations were 3.3, 1.7 and 4.8 times higher than those observed at the nearby fixed monitoring station during the morning, afternoon and evening hours, respectively. The BC concentration observed at ME4 was generally 3.1 times higher than that measured by the AE33 instrument located at the NUS atmospheric research station over the entire measurement period.
Thus, the current results suggest that the personal exposure assessment has the potential to provide the public health-relevant data at the city scale through the participation of volunteers.

**Outdoor Smoking area – ME1**

While mainstream tobacco smoke is a direct concern for smokers, the exhaled and side-stream smoke (known as Environmental Tobacco Smoke, ETS) contributes to passive smoking. Exposure to ETS has been associated with adverse health outcomes in adults and children, such as asthma, respiratory infection, and lung cancer (Hill *et al.*, 2004; Yolton *et al.*, 2004). Smoking prohibition, first introduced in 1970, is part of the national effort to promote a smoke-free lifestyle in Singapore. However, there are still certain designated smoking areas in the country (around 900 smoking corners island-wide (Alfred, 2017)). To date, limited data have been published in the archival literature on the assessment of human exposure to outdoor tobacco smoke. Therefore, we chose ME1 as one of the monitoring locations to evaluate the personal exposure to PM in an urban area. The measurement was conducted at a distance within 1 to 2 meters from smokers. The PM$_{2.5}$ concentration observed in the outdoor smoking area varied from 14.5 to 140.8 µg m$^{-3}$ with a mean concentration of 55.14 ± 24.35 µg m$^{-3}$. This mean concentration was 5 times higher than the measured PM$_{2.5}$ at the background location (11.04 ± 2.71 µg m$^{-3}$; SM-Table S4). The measured PM concentration was the lowest in the afternoon compared to
other time periods probably due to a decrease in the number of smokers at this smoking area and favorable weather conditions for the dispersion of the cigarette smoke emissions.

The concentration of BC to PM$_{2.5}$ was about 18.1% on average (Fig. 5). The number concentration of UFPs ranged between $23.4 \times 10^3$ and $268.7 \times 10^3$ # cm$^{-3}$ with a mean concentration of $90.0 \times 10^3 \pm 38.65 \times 10^3$ # cm$^{-3}$. In general, the exposure of individuals to PM is highly dependent on the proximity of air pollution sources (Klepeis et al. (2007)). Average PM levels within a 0.5 m distance from a smoker were significant higher while the PM concentrations at a distance greater than 2 m were much lower and generally close to the background. This observation suggests that pedestrians should avoid spending time in smoking areas and try to move to a distance of greater than 2 m, if unavoidable, to reduce the likelihood of experiencing elevated particle exposure because of tobacco smoke.

_Temple – ME2_

Incense burning, or joss paper burning is a popular religious practice in Singapore, particularly at Hindu/Buddhist temples. This religious practice produces non-stop heavy smoke due to the nature of its long, slow and incomplete combustion process. Incense smoke contains PM$_{2.5}$ and UFPs in large quantities (See et al., 2007; Ji et al., 2010), along with gaseous pollutants including many organic compounds, which are mostly adsorbed onto PM due to their non-volatile or semi-volatile characteristics (Chuang et al., 2013). The air pollution in and around various temples has
been documented to have severe human health impacts such as respiratory dysfunction, allergy, dermatological effects and neoplasm (Lin et al., 2008). The mean PM$_{2.5}$ concentration (215 ± 129.5 µg m$^{-3}$) observed just outside the temple (ME2) was the highest, which makes this location as the leading pollution hotspot compared to the other MEs. This mean concentration was about 20 times higher than the measured PM$_{2.5}$ at the background location. At this ME, the contribution of BC to PM$_{2.5}$ was about 10.1% on average (shown in Fig. 5). For UFPs, the number concentration varied between $19 \times 10^3$ and $734 \times 10^3$ # cm$^{-3}$ with a mean value of $230 \times 10^3 \pm 112 \times 10^3$ # cm$^{-3}$. This variation is attributed to changes in the number of devotees visiting the temple and the number of incense sticks burnt in the temple. In general, the number of devotees was higher in the morning hours (about 9:00 to 10:00) compared to the afternoon hours (about 12:00 to 13:00). This pattern leads to higher particle levels measured inside the temple during the morning hours. The particle levels inside the temple were reported to be much higher (18 times based on the study of Lin et al. (2002)) than what was measured outside the temple. Exposure to incense smoke is of health concern and should be reduced as much as possible. The emission of air pollutants from incense smoke can be effectively reduced by using less-polluting incense sticks (e.g. nanoscale calcium carbonate (Yang et al., 2011)) and keeping sufficient indoor ventilation. Moreover, people should shorten the exposure time when they worship at the temple when the incense smoke is heavy to reduce the risk of exposure to toxic PM.
Hawker centers or food courts are an open-air complex that houses many food stalls selling a variety of ethnic food including Chinese, Malay, Indian and Western food. Hawker centers are a unique feature of Singaporean culture and lifestyle. There are currently more than 100 hawker centers with more than 6000 food stalls located all over Singapore (MEWR-Singapore, 2017). It is well known that the emission of PM from cooking activities is strongly influenced by the type of stoves used, cooking styles, lipid contents in the food and the quantities of food cooked and ingredients used (Lee et al., 2001; See and Balasubramanian, 2008). In Singapore, liquefied petroleum gas is used as the primary source of energy for cooking. Therefore, food cooking using gas stoves is considered to be a significant source of airborne particles, especially UFPs, in many commercial kitchens, located in different parts of Singapore. Also, stir frying and deep frying are the commonly used cooking methods in different food stalls in Singapore. This cooking approach is found to produce more PM than other cooking methods (See and Balasubramanian, 2006).

The mean PM$_{2.5}$, BC, and UFPs concentrations observed at the hawker center were $61.6 \pm 38.7 \mu g \ m^{-3}$, $6.9 \pm 2.9 \mu g \ m^{-3}$, and $320.8 \times 10^3 \pm 131.1 \times 10^3 \# \ cm^{-3}$, respectively. Higher PM concentrations were recorded in the morning and afternoon compared to the evening hours because intensive cooking takes place to prepare breakfast and lunch. The exposure to PM from cooking activities was not confined only to the kitchen, and airborne particles were quickly...
dispersed to other places including the eating areas as revealed by our personal exposure measurements. Even though the concentration of PM should be lower in these places than that in the kitchen corner, the measured PM$_{2.5}$ and UFPs concentrations at the dining table were 5.6 and 19.0 times higher than those in the background location, respectively (given in the IM-Table S4). These high particle concentrations, especially with UFPs, makes the hawker center a pollution hotspot compared to the other MEs. In a previous study conducted in Singapore (See and Balasubramanian, 2006), the levels of non-carcinogenic and carcinogenic risk were 1.54 and 111 times higher than the acceptable levels, respectively. Effective protective measures should, therefore, be undertaken to reduce cooking fumes and minimize human exposure to such particulate emissions. Using electric stoves instead of gas stoves, avoiding cooking at very high temperature, keeping exhaust fan on during cooking may prove to be effective in reducing the exposure to PM derived from commercial cooking. Moreover, the use of particulate control devices with a high removal efficiency of PM will also be useful to mitigate the health effects of PM released from cooking.

**Bus stop – ME4 and Traffic intersection – ME5**

The mean PM$_{2.5}$ concentrations measured at the bus stop (ME4) and at the traffic intersection (ME5) were 36.4 ± 12.5 and 21.5 ± 5.5 µg m$^{-3}$ during the study periods. However, the PM$_{2.5}$ concentrations can go up to 50 - 400 µg m$^{-3}$ during the acceleration or deceleration of on-road
vehicles approaching or leaving the bus stop or traffic intersection. The mean $N_p$ concentrations were $30.9 \times 10^3 \pm 17.3 \times 10^3$ # cm$^{-3}$, and $16.5 \times 10^3 \pm 6.7 \times 10^3$ # cm$^{-3}$ at the bus stop and the traffic intersection, respectively. These PM concentrations are comparable to those measured in similar studies in other high-populated cities such as Taipei- Taiwan (Lung et al., 2014), Florence - Italy (Fondelli et al., 2008) or Barcelona – Spain (Moreno et al., 2015) at traffic – related locations.

The ratio of the mean BC to the mean PM$_{2.5}$, an indicator of the contribution of local incomplete combustion sources to the PM$_{2.5}$ concentration at each MEs, is shown in Fig. 5. In Singapore, 99.7%, 81.5% and 2.5% of the buses, taxis, and passenger cars are powered by diesel (LTA-Singapore, 2017). As expected, BC is a significant component of PM$_{2.5}$ at the measured vehicular traffic-influenced locations as a product of incomplete diesel combustion. The mean BC concentrations were elevated to $18.1 \pm 12.0$ µg m$^{-3}$ (52.9% contribution of BC to PM$_{2.5}$) at the bus stop and $4.5 \pm 2.1$ µg m$^{-3}$ (23.2% contribution of BC to PM$_{2.5}$) at the traffic intersection whereas the mean BC to PM$_{2.5}$ ratio of other MEs was only 13.5%. The BC fractions observed at traffic-influenced locations in this study are similar to those reported in the previous studies on BC emission from diesel engines (Kirchstetter and Novakov, 2007; Minjares et al., 2014).

**Lung Deposited Surface Area concentrations**
LDSA is considered to be an indicator of the toxicological effects of UFPs (Wittmaack, 2006). In current study, the mean LDSA values were 213.8 ± 90.0, 834.2 ± 396.5, 564.6 ± 276.5 and 55.0 ± 29.4 µm² cm⁻³ at the outdoor smoking area, the temple, the hawker center, and the traffic-influenced locations, respectively. These values are significantly higher compared to 37.2 ± 4.2 µm² cm⁻³ measured at the background location. As expected, LDSA and Np concentrations followed essentially similar trends as these metrics are related to UFPs. Values of LDSA >200 µm² cm⁻³ were generally observed at the hawker center and the temple at any time. The increase in LDSA may be associated with particle-induced adverse health effects (Sinclair and Tolsma, 2004; Buonanno et al., 2013). The LDSA concentration was found to vary between 30 and 89 µm² cm⁻³ at the urban background measurement stations located in Los Angeles-USA (Ntziachristos et al., 2007), Barcelona- Spain (Reche et al., 2015) and Lisbon- Portugal (Gomes et al., 2012). Although not directly comparable, our present results had a similar order of magnitude at the urban background site (the mean LDSA of 37.2 ± 4.2 µm² cm⁻³). To the best of our knowledge, limited measurements of LDSA were conducted at different indoor and outdoor MEs in urban areas.

**24-hour exposure measurement-based risk assessment**

24-hour PM₂.₅ personal exposure monitoring was conducted by the two subjects over four days recording their time activity patterns as illustrated in Fig. 6. This figure shows a representative
integrated 24-hour exposure conducted on 3 Nov 2017. The PM$_{2.5}$ concentrations measured with the portable device were compared to those obtained from the fixed ambient air monitoring station maintained by the NEA for four days. For this comparison, an inhalation rate of $1.3 \times 10^{-2}$ m$^3$ min$^{-1}$ was assumed based on the recommendation made by Moya et al. (2011). To the best of our knowledge, integrated 24 hours assessment of personal exposure to PM$_{2.5}$ has been sparsely reported in the literature. We compared the results obtained from this study (Table 2) to those reported by Cao and Thompson (2017) and Assimakopoulos et al. (2018). Our present results indicate a higher particle dosage to the respiratory system as compared to the afore-mentioned studies which is attributed to the differences in time-activity patterns, physical health factors (e.g., gender, age, height and body weight), intensity of human activity levels (e.g., sitting, walking or cycling) and the time, duration, extent and severity of exposure to PM$_{2.5}$. Higher particle deposition fraction has been reported among children or the elderly than healthy adults with the same PM exposure level (Moya et al., 2011), and this can in turn lead to more adverse health impacts to the former due to their weak immune systems.

The toxicological health risk associated with exposure to PM$_{2.5}$ was estimated in the current study. The estimated risk quotient (RQ) value is 3.3 (Table 3) when the exposure duration (ED) was assumed as 70 years as the worst case scenario (Cal/EPA, 2003; Los Angeles International Airport 2014; Cal/EPA, 2015). It should be noted that the estimated potential non-carcinogenic
health risk would be lower (RQ = 1.4) if the exposure duration (ED) for adults is to be taken as 30 years at the MEIR (maximally exposed individual resident). A value of RQ greater than 1 is indicative of potential adverse health effects on the exposed human population. The estimated chronic non-carcinogenic health risk shows clearly that individuals exposed to PM$_{2.5}$ and the particulate-bound toxic components derived from diverse outdoor and indoor MEs in densely populated countries such as Singapore may experience adverse health outcomes over their lifetime, resulting in a reduction in their life expectancy. The RQ estimated for adults was at 1.15 in the urban Mexico City (Díaz and Domínguez, 2009), whereas that of children under the age of 8 was 2.23 in Brazilian Amazon region (de Oliveira et al., 2012). Our results, therefore, corroborate other studies conducted elsewhere which found that the long-term inhalation of elevated levels of PM was associated with lung hyperinflation, indicative of small airway, cardiovascular, and neurological diseases (Calderon-Garciduenas et al., 2000; Heal et al., 2012; Landrigan et al., 2018). For estimation of carcinogenic health risk, the chemicals that are adsorbed onto airborne particles such as polycyclic aromatic hydrocarbon (PAHs) and toxic metals such as Cr (VI), Zn and Pb should be considered. Inhalable PM containing such chemicals can be deposited deep in the interior part of human lungs (Zwozdzia et al., 2017).

The 24 h PM$_{2.5}$ concentrations obtained from the personal sampler and the ambient PM$_{2.5}$ monitor showed a poor correlation ($R^2 = 0.025$), indicating that the conventional PM$_{2.5}$
measurements made at fixed air quality monitoring stations are not representative of personal exposure to PM$_{2.5}$ and therefore cannot be used for health risk estimates. Conversely, short-term personal exposure measurements reveal that the health risk estimate obtained from the true exposure to PM$_{2.5}$ is higher than that predicted by outdoor ambient PM$_{2.5}$ data (i.e., the ratio of PM$_{2.5}$ from personal exposure/from fixed monitoring stations $\geq 1.5$ - shown in Table 2). This personal exposure quality and its potential health outcomes depend on various factors including existing health conditions of exposed individuals, the intensity of physical activities such as walking and cycling, the time, place, duration, and severity of exposure, the nature of PM emission sources to which the individual is exposed and the size, quantity and chemical compositions of PM.

**PRACTICAL IMPLICATIONS**

Elevated concentrations of PM$_{2.5}$, especially BC, were observed at transport-influenced MEs in Singapore, making them hotspots of PM emissions in the outdoor environment. The personal exposure to PM$_{2.5}$ in such MEs is, therefore, of health concern. Even a short-term exposure to PM$_{2.5}$ in such pollution hotspots can contribute substantially to the daily personal exposure and related health effects. A better design of bus stops (e.g., design bus shelters that minimize trapping of air pollutants) and/or relocation of bus stops away from busy traffic intersections may reduce the personal exposure to freshly emitted airborne particles of vehicular origin. In the long
term, significant mitigation of particulate emissions from automobiles, especially from diesel vehicles, substantial investments in electric mobility, and promotion of active modes of transport such as walking, and cycling can lead to enhancement of urban air quality in Singapore with health benefits. The assessment of personal exposure to UFPs in terms of the LDSA concentration indicated that the food court and the temple are the PM hotspots. Therefore, appropriate PM exposure mitigation measures should be implemented at these MEs. Overall, this study emphasizes the need of conducting integrated personal exposure assessment over 24 hours across urban microenvironments together with the recording of individual time-activity patterns. Such a comprehensive personal exposure assessment can indeed complement the fixed site PM measurements in the context of health risk assessment and long-term public health improvement.

CONCLUSIONS

The personal exposure concentrations of PM$_{2.5}$, BC, and UFPs were measured in the CBD area of Singapore together with the estimation of LDSA, an indicator of health impacts of ultrafine particles. The personal exposure assessment study was conducted over the 7-km walking route to
identify air pollution hotspots, and at five selected air pollution hotspots for a duration of 1 hour each. T, RH, and GPS data were also recorded during the personal exposure monitoring campaign. The PM$_{2.5}$ concentration varied from 22.2 to 46.2 $\mu$g m$^{-3}$ over the 7 km walking route with a mean concentration of 32.5 $\mu$g m$^{-3}$. On average, BC accounted for 24.5% of the mass concentration of PM$_{2.5}$. The measured mean values of Np and LDSA were $62.4 \times 10^3$ # cm$^{-3}$ and 125.5 $\mu$g$^2$ cm$^{-3}$ at the CBD area over the 7 km walking route. Extremely high concentrations of PM$_{2.5}$ and BC were observed at the temple and bus stop. The hawker center was identified as a pollution hotspot based on the number concentration of UFPs and LDSA compared to the other MEs.

In addition, to evaluate the potential human health risk associated with the personal exposure to PM$_{2.5}$, integrated personal exposure studies were conducted over 24-hours for a duration of 4 days. The estimated risk quotient (RQ) value of 3.3 indicates that there is a high probability of experiencing negative health effects due to the continued exposure to PM$_{2.5}$ in densely populated cities, which is in agreement with the findings reported in previous studies. Moreover, the study revealed that city dwellers are exposed to higher PM concentrations during their day-to-day activities in urban MEs than those suggested by the most commonly used fixed outdoor air quality monitoring stations.
Ambient PM$_{2.5}$ mass levels have been improved in many developed countries due to implementation of effective air pollution control policies. However, the recent PM exposure metrics such as BC and UFPs that are considered to be better indicators of potential health outcomes are seldom investigated in cities. By conducting time-integrated personal exposure measurements across urban diverse MEs together with the recording of individual time-activity patterns and other important factors, we anticipate that realistic health risk estimates can be estimated and communicated to city-dwellers. The outcome of such a comprehensive personal exposure assessment would help environmental policy makers identify and implement effective mitigation measures to reduce people’s exposure to particulate air pollution and provide the general public with improved health benefits at the city scale.

This study was mainly conducted in order to explore the feasibility of assessing integrated personal exposure to PM across diverse outdoor and indoor MEs and not to quantitatively conclude its levels at the measurement sites included in the study. Further work with participation of a large number of volunteers across a wide network of MEs would be required to provide more realistic exposure information on the city scale.

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REFERENCES


Table Captions

Table 1. Characteristics of micro-environments at the study location.

Table 2. 24-hour integrated exposure to PM$_{2.5}$ based on personal exposure and ambient outdoor air quality monitoring.

Table 3. Results of health risk assessment.
Table 1. Characteristics of micro-environments at the study location.

<table>
<thead>
<tr>
<th>Microenvironments</th>
<th>Characteristics</th>
</tr>
</thead>
<tbody>
<tr>
<td>ME1 Outdoor Smoking Area</td>
<td>Stay on the Nicoll Highway. There were about 6-10 smokers in the morning and 5-8 smokers in the afternoon and evening hours on observed days.</td>
</tr>
</tbody>
</table>
| ME2 Kwan Im Thong Hood Cho Temple | Buddhist Temple  
Prayer area with elevated incense smoke.  
Measurement location was at the courtyard of the temple, about 3m from the temple.  
Close at night |
| ME3 Hawker Center      | An open-air complex with selling a whole range of food. There are about 20 food stalls. |
| ME4 Opp Bugis Junction Bus Stop | 12 bus services  
Seven-lane road, with a bus lane. |
| ME5 Traffic Intersection | Four-way intersection crossing over of Beach Road and Ophir Road |
Table 2. 24-hour integrated inhaled dose of PM$_{2.5}$ based on personal exposure and ambient outdoor air quality monitoring.

<table>
<thead>
<tr>
<th>Monitoring day</th>
<th>24-hour integrated inhaled dose of PM$_{2.5}$ (µg)</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Personal Exposure PM$_{2.5}$ monitoring</td>
</tr>
<tr>
<td>Day 1</td>
<td>608.5</td>
</tr>
<tr>
<td>Day 2</td>
<td>725.3</td>
</tr>
<tr>
<td>Day 3</td>
<td>755.0</td>
</tr>
<tr>
<td>Day 4</td>
<td>586.2</td>
</tr>
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</table>
Table 3. Results of health risk assessment

<table>
<thead>
<tr>
<th>Factors</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Integrated Concentration of PM$_{2.5}$, CA</td>
<td>35.7</td>
<td>µg m$^{-3}$</td>
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<tr>
<td>Lifetime Average Daily Dose, LADD</td>
<td>4.4</td>
<td>µg kg$^{-1}$ day$^{-1}$</td>
</tr>
<tr>
<td>Reference Dose, RfD</td>
<td>1.3</td>
<td>µg kg$^{-1}$ day$^{-1}$</td>
</tr>
<tr>
<td>Risk Quotient, RQ</td>
<td>3.3</td>
<td>-</td>
</tr>
</tbody>
</table>
Figure Captions

**Fig. 1.** 7-km walking route around the CBD area with five selected micro-environments and background location. The map was generated from Google map.

**Fig. 2.** Personal exposure concentrations of: (a) PM$_{2.5}$, (b) BC, (c) Np, and (d) LDSA during weekdays and weekends at the CBD area compared to the background location.

**Fig. 3.** Time series of: (a) PM$_{2.5}$, (b) BC, (c) Np, and (d) LDSA. The measurements conducted on 2 Jan 2018 during morning hours.

**Fig. 4.** 1-hour exposure concentrations of: (a) PM$_{2.5}$, (b) BC, (c) Np, and (d) LDSA at selected MEs. These data were compared to those reported by NEA (for PM$_{2.5}$) and NUS laboratory (for BC).

**Fig. 5.** Ratios between mean BC and PM$_{2.5}$ concentrations (%) and mean PM$_{2.5}$ concentration (µg m$^{-3}$) at each ME. *(M-Morning, A-Afternoon, E-Evening).*

**Fig. 6.** Integrated 24-hour exposure to PM$_{2.5}$. The measurement was conducted on 3 Nov 2017.
Fig. 1
Fig. 2.
(a) 

(b)
Fig. 3
Fig. 4
Fig. 5.
Fig. 6