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Assessment and Mitigation of Personal Exposure to Particulate Air Pollution in Cities: An Exploratory Study

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KEYWORDS: Personal Exposure; Time-activity; Airborne Particles; Urban Air Quality; Health Risk Assessment

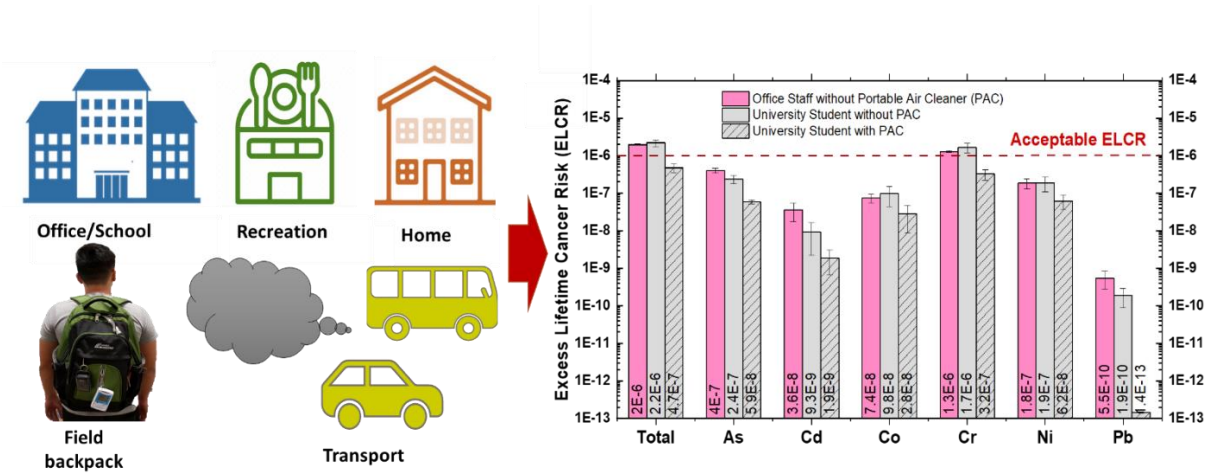
ABBREVIATIONS

PM: airborne particulate matter; BC: black carbon; UFP: ultrafine particle; PE: personal exposure; ME: micro-environment; MRT: mass rapid transport; HVAC: heating, ventilation and air conditioning; FMS: fixed monitoring stations; OS: office staff; US: University student; PAC: portable air cleaner; GPS: global positioning system; TAD: time-activity diary; NEA: National Environmental Agency; ATN: attenuation; NUS: National University of Singapore. MERV: Minimum Efficiency Reporting Value; ELCR: excess lifetime cancer risk; RQ: risk quotient; TE: trace element.

ABSTRACT

Assessment of integrated personal exposure (PE) to airborne particulate matter (PM) across diverse microenvironments (MEs) over 24 hours under different exposure scenarios is necessary to identify appropriate strategies to improve urban air quality and mitigate the health effects of PM. We carried out a collaborative study in a densely populated city-state (Singapore) to assess the integrated PE to fine particles ($PM_{2.5}$), ultrafine particles (UFPs) and black carbon (BC) across diverse indoor and outdoor urban MEs, estimate related health risk and make suitable recommendations for healthy living in cities. Two volunteers with different lifestyles participated in the study by tracking their PE to particulate air pollution and the time-activity patterns over 24 hours using portable PM monitoring devices and recording their whereabouts using GPS coordinates. Home, transport and recreation (i.e., food court) MEs represented pollution hotspots of $PM_{2.5}$ (21.0 $\mu g/m^3$), BC (3.4 $\mu g/m^3$) and UFP (33.0 $x10^3$ $\#/cm^3$), respectively. Among the different modes of transport used by the participants (walking, cycling, e-scooter, mass rapid transport (MRT), bus, car and taxi), the air pollutants had elevated concentrations while commuting by public transport (bus and MRT) as well as during active modes of transport (walking and cycling). Air-conditioned cars and taxis, equipped with air filtration systems, represented the lowest PE. The health risk assessment revealed that there are potential carcinogenic risks associated with the long-term exposure to elevated levels of $PM_{2.5}$-bound toxic trace elements. These risks can be mitigated with the introduction of low-carbon and active modes of transport in place of internal combustion engines and the use of indoor air pollution exposure mitigation devices.

Graphical Abstract



1. Introduction

Exposure to elevated levels of PM is known to be associated with increased mortality and hospital admissions due to respiratory and cardiovascular diseases around the globe (Kim et al., 2015; Kirrane et al., 2019; WHO, 2018). This association between particulate air pollution and health effects has been established through epidemiological and health impact assessment studies, providing a scientific basis for air quality management on the national and urban scale levels (Landrigan et al., 2018; Lelieveld et al., 2015; Martins and da Graça, 2018; Silva et al., 2013; WHO, 2019). Urban dwellers currently account for 55% of the world's population, which is projected to increase to 68% by 2050, particularly in Africa and Asia (UN, 2018). Consequently, a significant number of people will continue to be exposed to potentially dangerous levels of PM as urban areas are associated with high levels of particulate air pollution arising from local intense industrial activities and motorized traffic (Barzeghar et al., 2020; Karagulian et al., 2015). Residents in cities inhale PM of different sizes, shapes and chemical composition in both indoor and outdoor environments on a continual basis. This exposure to PM can vary substantially over time due to changes in the type and strength of PM (and/or their precursors) emissions. It is also necessary to consider the place, duration, time of PE, prevailing meteorological conditions, the characteristics of outdoor MEs, buildings and indoor MEs, and the type of vegetation in the outdoor environment (Abhijith et al., 2017; Challoner and Gill, 2014; Cyrys et al., 2004; Kumar et al., 2014; Morawska et al., 2001). Furthermore, the type of ACMV (air conditioning and mechanical ventilation)/HVAC (heating, ventilation and air conditioning), filtration systems, and the type of physicochemical transformations of PM influence the PE an individual may experience (Karner et al., 2010; Marshall et al., 2008; Tham et al., 2016). Fixed monitoring stations (FMS)

used as part of ambient air quality compliance requirements measure PM concentrations as a function of time at a particular location. However, these measurements do not account for spatial variations in PM exposure concentrations, limiting the ability of epidemiologists to identify vulnerable populations such as the elderly, children and individuals with pre-existing health conditions (Apte et al., 2017; Lei et al., 2020). As a consequence, public health concerns related to PM exposure are inadequately addressed due to lack of data on spatiotemporal variations of PM and true dose – effect relationships.

The advent of portable and reliable sensors in recent years has facilitated the characterization of $PM_{2.5}$, UFPs, and BC in several urban MEs, influenced by residential, outdoor, cultural, religious and recreational activities (Adam et al., 2020; Bekö et al., 2015; Jeong and Park, 2017a; Massey et al., 2009; Steinle et al., 2015; Sultan et al., 2020; Tran et al., 2020b). Motorized (car, bus, and train) and non-motorized (walking and cycling) modes of transport have been the focus of PE studies in North America, Europe and Asia, which showed large variability of short-term PM concentrations across diverse MEs (e.g., Ham et al., 2017; Li et al., 2015; Qiu et al., 2019; Rivas et al., 2017). However, the complex interaction between fast-changing space-time variations of air pollution and human movement leads to diverse exposure profiles and different levels of health risk (Park and Kwan, 2017). Hence, integrated daily 24-hour PE measurements of PM would provide insights into causes of its spatio-temporal variability and the contribution of diverse MEs to adverse health impacts of PM. Park and Kwan (2017) assessed individual integrated 24-hour exposures to O_3 (ozone) levels based on the spatiotemporal variability of air pollution and individual daily movement patterns. However, this study only considered the air quality with specific reference to O_3 in outdoor environments. A realistic methodology to assess the integrated

PE of individuals to PM in both outdoor and indoor MEs is needed. Previously reported 24-hour PE studies focused on the characterization of $PM_{2.5}$ (Assimakopoulos et al., 2018; Cao and Thompson, 2017; Gao et al., 2018), as well as BC (Dons et al., 2011; Dons et al., 2019), UFPs (Bekö et al., 2015), and a combination of these air pollutants by Koehler et al. (2019). These studies did not cover a wide range of outdoor and indoor MEs with varying levels of human activities. In addition, $PM_{2.5}$ is composed of several toxic chemical components including trace elements (TEs) which are known as a major contributor to potential human health risks (Lovett et al., 2018; Zhang et al., 2017). Several studies have shown that TEs play an important role in the generation of reactive oxygen species (ROS) in lung fluids which exhibit a high oxidative stress potential, leading to adverse health effects associated with exposure to PM (Feng et al., 2016; Karthikeyan et al., 2006; Lodovici and Bigagli, 2011; Pavagadhi et al., 2013; See et al., 2007). Estimating the water-soluble (bioavailable) fraction of PM-bound TEs is important to assess realistic human health risks (Huang et al., 2016; Karthikeyan et al., 2006). To the best of our knowledge, no systematic health risk assessment (carcinogenic or non-carcinogenic) based on bioavailable concentrations of TEs has been conducted in any of the integrated daily 24-hour PE studies reported in the literature. Also, several studies showed a reduction of indoor PM concentrations while using air cleaning devices (e.g., Sharma and Balasubramanian, 2017; Sharma and Balasubramanian, 2019; Tham et al., 2018; Tran et al., 2020c). It is therefore important account for integrated PE to PM under different exposure scenarios and assess health benefits associated with mitigation of PE to PM. To fill these knowledge gaps, we have initiated a Global Alliance project involving multi-institutional and multi-disciplinary collaboration with the aim of contributing to improved healthy living in cities based on the time-integrated assessment of PE to $PM_{2.5}$ across diverse MEs and related health risks.

Herein, we present the outcome of time-activity based PE studies conducted in the densely populated Southeast Asian city-state of Singapore (7,800 people per km^2). Two volunteers (an office staff (OS) and a University student (US)) participated in the study by tracking their PE to $PM_{2.5}$, BC, UFP and time-activity patterns over 24 hours using portable devices and a global positioning system (GPS). We conducted the integrated PE study for a duration of two months over diverse locations including residential apartments (with and without a portable air cleaner (PAC)), air-conditioned workplaces, transport MEs with active and/or passive mobility and recreational areas (either naturally or mechanically ventilated). We measured real-time concentrations of CO and CO_2 to differentiate between combustion and non-combustion sources of PM and to assess the sufficiency of ventilation levels in indoor environments. Based on the chemical speciation of $PM_{2.5}$, we quantified carcinogenic and non-carcinogenic related health risks. This work represents the first study of its kind for the assessment of integrated PE to PM in a densely populated city.

2. Materials and methods

2.1. Site description and experimental design

The PE measurements presented in this study were carried out in the city-state of Singapore, one of the wealthiest countries in the world based upon the gross domestic product per capita (100,345 USD; (IMF, 2018)). The air quality, and thus $PM_{2.5}$ concentrations at ground level, in Singapore are strongly impacted by on-road vehicular emissions, with the number of registered vehicles at nearly 1 million (LTA, 2018). Besides personal vehicles, public transport is a frequently used mode

of transport by commuters across the island. According to the land transport authority (LTA), the average daily bus ridership stands at 4,037,000 (53.6%) while that of mass rapid transport (MRT) is 3,302,000 (43.8%), with the remaining ridership from the use of light rail transit (LRT) at 199,000 (2.6%) (LTA, 2018). Because of Singapore's proximity to the equator (~1.2˚ N) and maritime exposure, the weather conditions year-round are characterized by relatively uniform air temperatures (21 – 33 ˚C), relative humidity (60 – 90 %) and low monthly average wind speeds (1.6 - 3 m/s) with monsoon rains leading to high annual total rainfall (2,340 mm) (Velasco and Roth, 2012). The southwest monsoon is active from June to October while the northeast monsoon prevails from December to March. Our study was conducted during the dry season, i.e., April to May 2018 on non-rainy days in the absence of regional smoke haze. Table S1 shows local meteorological conditions that prevailed in Singapore during the field study. Real-time PE observations were made relating to $PM_{2.5}$ (TSI SidePak520), BC (microAeth AE51), UFP (Testo DiscMini) and CO/CO_2 (TSI 7545 QTrak). Also, relative humidity RH (%) and temperature T (ºC) were measured using temperature and humidity loggers (BG-Log-TempRH). Table S2 provides the characteristics of each device. In addition, $PM_{2.5}$ samples were collected onto Teflon filters using a Personal Environmental Monitor (PEM, MSP Corporation, USA) and a Leland Legacy pump (SKC, USA) for speciation of trace elements. Further details are provided in the following sections.

The volunteers (OS and US), both belonging to the National University of Singapore (NUS), carried the portable PM measurement devices with them in a backpack (see Fig. S1) for 24 hours during their daily routine, which involved spending time in different MEs including their residential homes, transport MEs, offices, lecture halls and recreation areas (food courts). The OS'

apartment was located in the south-western part of Singapore on the 13th storey of a high-rise multi-storey building in close proximity (100 m) from a road connecting west-south Singapore (Fig. S2). The US' apartment was located in the eastern part of Singapore on the 11th storey of a high-rise multi-storey building close (100 m) to a road with relatively lower traffic volume compared to the OS (Fig. S2). As part of the study protocol, the US deployed a portable air cleaner (PAC) (City M, Camfil, Stockholm, Sweden, Particle Clean Air Delivery Rate (CADR) is 433 m^3/h and the average air purification area is about 75 m^2) at home in order to compare the PE to PM under two exposure scenarios: (i) natural ventilation (keeping all the windows of the bedroom fully open) without the PAC and (ii) keeping all the windows of the bedroom fully closed and using the PAC as the PM exposure mitigation device. The PAC was equipped with two filter-packages with each package containing a high-efficiency particulate air (HEPA) filter and a molecular gaseous contaminant removal filter. The use of the molecular filter may reduce the concentration of indoor gaseous pollutants such as volatile organic compounds (VOCs), O_3, and suppress odor. The daily commute of the OS and US involved travel from their residential locations to the NUS campus. NUS is located in the southwestern part of Singapore which is influenced by vehicular emissions from roads adjacent to the campus, in particular a busy expressway to the northern side of the campus leading to the Central Business District to the South-East and industrial emissions from petroleum, petrochemical and oil refinery industries located 5-10 km to the South-West from the University. In addition, sea spray and PM emissions from a seaport located about 1 km from the University influence the local air quality.

However, the traveling distances of the participants back and forth from their homes to the workplace differed considerably. The OS travelled approximately 3.5 km each way while the US

lived farther away (20 km). This resulted in different travel times and hence different levels of PE to air pollutants. Furthermore, each participant used a different combination of transport modes from their home to places of interest. The OS used a combination of active modes of transport (walking and cycling) and non-emitting motorized transport (e-scooter) with exposure to PM in ambient air, and passive modes of transport (bus and taxi) which were mechanically ventilated (air-conditioned). On the other hand, the US used only mechanically ventilated (air-conditioned) modes of transport (car, taxi, MRT, and bus). It should be noted that the taxis used by US and OS were diesel-driven while the car used was petrol-driven. Both participants visited different food courts on the NUS campus for lunch. The OS visited an air-conditioned food court located near the workplace, which serves food items prepared in different ethnic cooking styles (e.g., Indian, Chinese, and Malay). The US frequently visited a food court with food cooked with a range of cooking styles including western and characterized by natural ventilation. The participants noted down their whereabouts in a time-activity diary (TAD). The TAD together with a GPS receiver was used to identify the specific time periods and MEs for the PE assessment on a day-to-day basis. The description of all MEs is given in Table 1.

Table 1. Description of the microenvironments.

Participants	Microenvironment		Time (hour)	Location, configuration, cooling and ventilation conditions
Office staff	Home		16.2±0.4	13th storey of a tall multi-storey building in ≈100 m from a major road, in the south-western part of Singapore. Natural ventilation (Windows opened).
	Office		6.7±0.3	15th storey of a tall multi-storey building in the southwestern part of Singapore. Cooling by central air-conditioning system; mechanical ventilation with MERV-7 filter.
	Recreation		0.6±0.1	A range of cooking styles Cooling by central air-conditioning system; mechanical ventilation with MERV-7 filter.
	Transport	E-scooter	1.0±0.1	Ambient air conditions
		Walking	1.3±0.1	Ambient air conditions
		Cycling	1.1±0.1	Ambient air conditions
		Bus	1.1±0.1	Cooling by central air-conditioning system with recirculation mode.
		Taxi	0.8±0.1	Cooling by central air-conditioning system with recirculation mode. Diesel-drive.
University student	Home	Without PAC	13.6±1.6	11th storey of a tall multi-storey building in ≈100 m to a minor road in in the eastern part of Singapore. Natural ventilation (Windows opened).
		With PAC	13.0±0.8	Window closed, PAC on
	School (classroom)		6.8±1.6	6th storey of a tall multi-storey building in the southwestern part of Singapore. Cooling by central air-conditioning system; mechanical ventilation with MERV 7 filter.
	Recreation		0.8±0.2	A range of cooking styles. Natural ventilation
	Transport	Bus	3.3±0.2	Cooling by central air-conditioning system
		MRT + bus	2.4±0.1	Cooling by central air-conditioning system
		Car	0.9±0.1	Cooling by central air-conditioning system with recirculation mode. Petrol-driven
		Taxi	1.2±0.1	Cooling by central air-conditioning system with recirculation mode. Diesel-drive.

PAC: Portable air cleaner. MERV: Minimum Efficiency Reporting Value.

2.2. $PM_{2.5}$ measurements

Two portable nephelometers (SidePak AM520, TSI, USA) (the detection limit: 1μgm^{-3}) were used for real-time measurements of $PM_{2.5}$ concentrations. The measurement principle of the SidePak is based on light scattering by airborne particles. The scattered light is then transmitted to a photodetector, which processes the change in the light intensity to calculate the mass concentration of $PM_{2.5}$. The SidePak was factory calibrated based on a standard test protocol with ISO12103 – A1 Arizona Test Dust. Prior to field measurements, the two SidePaks were calibrated against a MiniVol (Airmetrics, USA) portable sampler (co-located with the instruments at the same sampling site) via gravimetric analysis to determine the calibration factor representative of Singapore's composition of light scattering airborne particles. The $PM_{2.5}$ concentration data obtained from the MiniVol were compared to those from the SidePaks and the calibration factors were found to be 0.26 and 0.28 of the factory default for the two units. The PM devices were operated at pre-calibrated flow rates of 1.7 l/min and fitted with a $PM_{2.5}$ impactor to remove PM with greater than 2.5 μm in diameter.

In addition, 1-hour average $PM_{2.5}$ data were obtained from the FMS in the central part of Singapore, maintained by the National Environmental Agency (NEA) website (http://www.nea.gov.sg). The ratio of PE to $PM_{2.5}$ measured across different MEs and $PM_{2.5}$ concentrations obtained from the FMS was used to assess the degree by which the PE and commonly used FMS $PM_{2.5}$ observations differ. This normalization of PM concentrations enables a realistic comparison of the PE to different MEs and urban ambient air.

2.3. Black carbon measurements

Two microAeth portable monitors (AethLabs, Model AE51, USA) were used by the two participants to measure BC concentrations in real-time during the 24-hour personal exposure study. The microAeth measures light absorption with wavelength of 880 nm on a T60 Teflon-coated glass fiber filter media to obtain attenuation coefficients. The sampled air of 150 mL/min is dried to less than 40% relative humidity using a portable aerosol dryer (Perma Pure, USA) prior to its entry into the device. The mass concentration of BC measured using AE51 can be underestimated as the filter BC mass increases, particularly when sampling highly light-absorbing particles (Jimenez et al., 2007; Kirchstetter and Novakov, 2007). We therefore made a suitable correction for this potential problem using the empirical relationship of Kirchstetter and Novakov (2007), which yielded a good agreement between thermal-optical analysis and aethalometer measurements of BC. The filter strip was also replaced after 12 to 14 hours of air sampling to minimize filter loading effects. Data with an optical attenuation (ATN) value greater than 70 were discarded to ensure the integrity of BC measurements. Before each sampling session, the flows of the two microAeths were calibrated with a flowmeter (TSI, model 4100, USA) by using the auto flow calibration procedure recommended by the manufacturer. The performance of AE51 was further checked by an inter-comparison with an Aethalometer (AE33, Magee Scientific, USA) over a 24-hour period of measurement. A good agreement was obtained from the inter-comparison study with $R^2 = 0.81$, slope = 0.998 (The data are shown in Fig. S3).

Moreover, fixed site measurements of ambient BC at the NUS atmospheric rooftop laboratory were carried out with the Aethalometer AE-33. The Aethalometer measures the ATN of light at

880 nm as the aerosols of 3 L/min are collected on a quartz-fiber filter, and BC is the exclusively absorbing element at this wavelength. Further details of the working principle and calibration steps of both Aethalometer AE33 and microAeth AE51 are provided in the Supporting Information (SI).

2.4. Ultrafine particle number concentration measurements

A Diffusion Size Classifier (DiSCmini, Testo, Germany), operated at 1 L/min and frequency of 1 Hz, was used to measure particle number concentrations in the range of 10 - 300 nm in real-time. Although the upper limit of the UFP is 300 nm, the particle number (PN) concentration is a reasonable proxy for PN less than 100 nm under the conditions encountered in our study. The DiSCmini operates based on the electrical charging of particles in a two-stage detection process. Positive ions are generated in a corona discharge and mixed with the aerosol sample. During the first detection process, small particles deposit on a pile of steel grids by diffusion and are detected as an electrical current. The remaining particles are detected in the second stage, also via electrical current. The ratio of these two currents is a measure of the average particle size. The number concentration and the average particle size (diameter) can then be determined from the total current together with the flow rate of the instrument as the charge per particle is a function of the diameter.

2.5. Mobile gas-phase (CO and CO_2) measurements

The TSI Model 7545 Q-Trak measures CO from 0 to 500 ppm and CO_2 from 0 to 5000 ppm. The mixing ratios are determined with the help of a non-dispersive infrared (NDIR) detector which measures the amount of IR radiation that is absorbed at a particular wavelength in the sample and

is proportional to the mixing ratio of CO or CO_2 contained in the sample. Note that the CO/CO_2 mixing ratios presented in Table 2 and Table 3 pertain exclusively to the measurements which the OS carried out as part of the study protocol.

2.6. Quality control, data processing and analysis

The integrated PE study was carried out for two months (April and May 2018) on 30-non-rainy days to avoid misrepresentation of urban air pollution conditions due to the wet scavenging of aerosols. Before each use of the SidePak, AE51, DiSCmini and Q-Trak, the impactors were cleaned, battery and memory checks were carried out, and clock synchronization was done. Noteworthy occurrences such as low airflow on the instruments were noted for the subsequent data analysis. The real-time data acquired by the mobile devices were downloaded, inspected and archived once a day. Other potential data quality issues (missing data, negative or otherwise spurious readings) were flagged for subsequent evaluation and resolution. If an instrument failure resulted in > 4 of 24 hours in a specific day, the data collected during those days were excluded. We used a code written in R studio (version 1.1.442) to import, synchronize, and combine datasets and to apply corrections, calibrations and perform statistical analysis. In this study, we consider the geometric mean (GM) rather than the arithmetic mean (AM) to discuss the variations of PM data as the GM fits the log normal-distribution of PM concentrations (Lee et al., 2015; Targino et al., 2016). GM is widely used in the literature to report the mean PM concentration (Adams et al., 2001; Hu et al., 2014; Lee et al., 2015; Paunescu et al., 2017). Nevertheless, all other descriptive statistics are also provided.

2.7. Filter samples

Together with the real-time PM measurements, 24-hour $PM_{2.5}$ gravimetric samples were collected on 37 mm Teflon filters using the Personal Environmental Monitor (PEM, MSP Corporation, USA) and the Leland Legacy pump (SKC, USA) operated at a flow rate of 10 L/min. Before and after sampling, filters were kept in a dry box at a constant temperature of 23.0 ± 0.5 ºC and relative humidity of 33 ± 3 % for at least 24 hours before weighing them with a microbalance (Sartorius AG, Göttingen, Germany) with an accuracy of ±1 µg and a precision of 0.1%. After sampling, the PM filters were stored in a refrigerator at -18 °C until extraction and chemical analyses.

All the Teflon filters were cut into two halves and analyzed for total and water-soluble trace elements (TEs). The measurement method and the QA/QC protocols used in this work were the same as reported in our previous studies (Betha et al., 2013; See and Balasubramanian, 2008; Sharma and Balasubramanian, 2017, 2019) and are described briefly in the SI. A total of 26 selected TEs (Li, B, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Cd, Sn, Cs, Ba, Ti and Pb) were analyzed to determine their total and water-soluble concentrations using an inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7700 Series, Agilent Technologies, USA). We selected these TEs based on their known or potential effects on human health. Also, these TEs are representative of specific emission sources of PM as discussed in the results and discussion section.

2.8. Data analyses/Inhaled dose calculation

The daily integrated inhaled dose (e.g., unit of μg/day for $PM_{2.5}$) was determined by integrating the concentrations (e.g., unit of μg/cm^3 for $PM_{2.5}$) obtained in each ME (i.e., office/school, home, recreation, transport) over the time spent (unit of hour/day) in the corresponding ME and the inhaled rate (unit of m^3/hour) (Eq. 1). Calculation of the inhalation rate in each ME *i* was done according to Eq. 2.

$$\text{Integrated inhaled dose} = \sum_{i=1}^{n}(\text{Concentration}_i \times \text{Inhalation rate}_i \times \text{Exposure time}_i) \quad (1)$$

$$\text{Inhalation rate}_i = \text{Tidal volume}_i \times \text{Breath frequency}_i \quad (2)$$

The tidal volumes chosen for different activities were 750, 1250, and 1920 cm^3 per breath during sitting (in homes, food courts and taxis or cars), light (e-scooter and bus rides and walking), and heavy (cycling) physical activities, respectively (Hinds, 2012). The typical breathing frequencies selected were 0.2, 0.33, and 0.43 breaths per second during passive (sitting) physical activities and active (light and heavy movement), respectively (Hinds, 2012). The tidal volumes and breathing frequencies were selected for male adults (the 21-65 age group). The method to estimate the inhalation rate based on tidal volume, breath frequency and physical activity level has been widely used in the literature (Gupta and Elumalai, 2019; Kumar et al., 2018; Tran et al., 2020a).

2.9. Health risk assessment

Potential carcinogenic and non-carcinogenic human health risk assessment associated with inhalation exposure to $PM_{2.5}$-bound water-soluble TEs and total $PM_{2.5}$ mass concentration was conducted for three exposure scenarios: OS and US remaining home with no air cleaner, and US

with a PAC operating in the home environment. The health risk analysis is based on the framework developed in previous studies (NRC, 1983; See and Balasubramanian, 2008; Sharma and Balasubramanian, 2019). There are four steps involved: (1) Hazard identification, (2) Exposure assessment, (3) Dose-response assessment and (4) Risk characterization. Details are given in the SI.

3. Results and discussion

3.1. Time-activity pattern of the two participants

The percentage of time the two participants spent in different MEs is shown in Fig. 1. Both participants spent most of their time at their homes, which varied on a day-to-day basis depending on the nature of their work, working schedule and the mode of transport taken to/from the work environment. The OS spent nearly the same amount of time during active (walking and cycling) and non-emitting motorized (e-scooter) (4.5%) and emitting-motorized (bus and taxi) (4.1%) modes of transport. Also, the percentage of time spent in the home environment is nearly equal while using active and non-emitting motorized (65.0%) and emitting motorized (63.9%) modes of transport.

However, due to longer travel distance, the US spent a significantly more time commuting to the University in public transport (11.1%) which resulted in relatively less time spent in the home environment (58.4%). In contrast, the faster mode of transport by car/taxi (4.2%) to the University resulted in a longer duration spent at home (66.0%). The time spent at home is quite similar to that reported by Jeong and Park (2017b) based on the time-activity pattern recorded for school going

children in Seoul, South Korea. The children spent, on average, 64% of their time at their home, 15% in school and 7% in transport (Jeong and Park, 2017b). Similar findings were also reported in a 2-year national probability telephone survey conducted in USA (Klepeis et al., 2001) with 87% of participants' time being spent in enclosed buildings and about 6% of their time in enclosed vehicles. In our study, the OS and US spent their remaining time in similar proportions at the office/University (~28%) and during recreational activities (2-3%) (see Fig. 1). The time-activity pattern presented in our study refers to weekdays only.

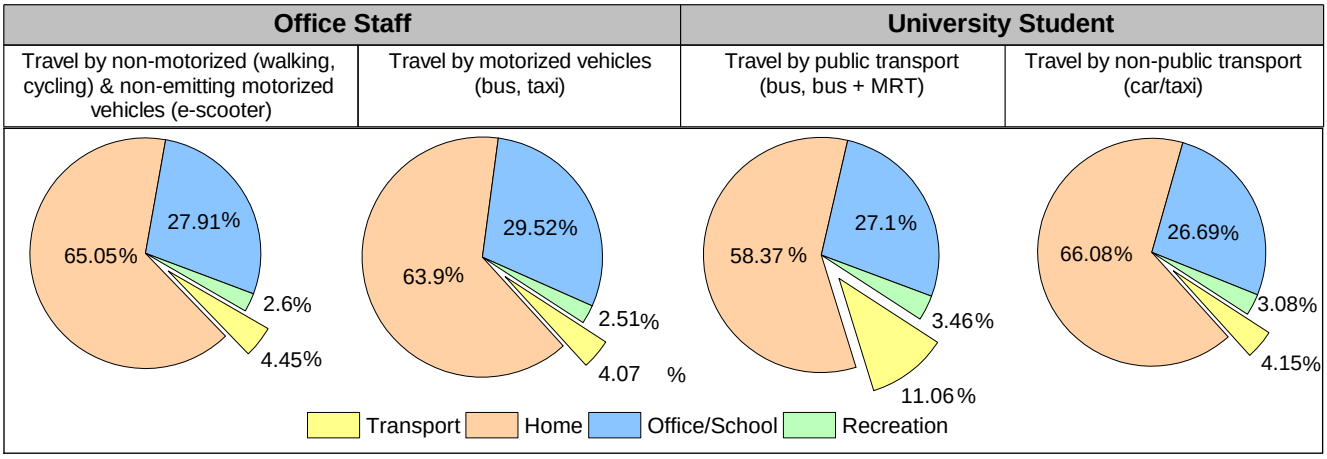


Fig. 1. Daily time-activity patterns of the study participants in diverse MEs.

3.2. Daily integrated inhaled dose of $PM_{2.5}$, BC and UFP by MEs

The PE to $PM_{2.5}$, BC and UFPs for the OS and the US is shown in Table 2, and illustrative examples of 24-hour time series showing the PE that the OS and US encountered in various MEs are shown in Fig. S4 and Fig. S5, respectively. The PE to PM assessed for both participants was significantly

different among types of time-activity, or types of MEs according to a multiple comparison test (p-value < 0.001). The recreation, home and transport MEs represented pollution hotspots. For the OS, the $PM_{2.5}$ (geometric mean (GM) = 21.92 ± 1.77 μg/m^3) and UFPs (GM = 32.91 x 10^3 ± 2.52 x 10^3 #/cm^3) concentrations were the highest in the food court, followed by home (GM = 18.16 ± 1.80 μg/m^3 & 14.68 x 10^3 ± 1.81 x 10^3 #/cm^3 for $PM_{2.5}$ and UFPs, respectively) and transport (GM = 13.74 ± 2.00 μg/m^3 & 15.45 x 10^3 ± 2.63 x 10^3 #/cm^3) MEs. However, the BC levels were the highest at home (GM = 4.54 ± 2.09 μg/m^3), followed by transport (GM = 3.32 ± 2.37 μg/m^3) and food court (GM = 1.72 ± 1.82 μg/m^3) MEs. CO concentrations experienced by the OS ranged from 0.17 ppm (parts per million by volume) in the office to 0.77 ppm in the food court, while CO_2 levels ranged from 395 ppm in the home microenvironment to 552 ppm in the food court. The observed high BC levels at home could be explained by the migration of traffic-related air pollution (TRAP) from an adjacent road with a high volume of on-road vehicles, especially diesel-powered vehicles (e.g., buses and trucks). BC is a good indicator of TRAP, since its emission is closely related to the combustion of carbonaceous fuels, a major component of traffic emissions (Targino et al., 2016; Tran et al., 2020a). Apart from increased BC emissions during the rush hours, the shallow boundary layer tends to accumulate air pollutants in ambient air due to poor dispersion, resulting in elevated BC concentrations in the morning compared to other time periods (see in Fig.S4).

Table 2. Personal $PM_{2.5}$, BC, UFP, CO and CO_2 exposure levels in different MEs.

Metric		Office Staff					University Student					
		Office	Home	Transport	Recreation	**Overall**	School	Home without PAC	Home with PAC	Transport	Recreation	**Overall**
$PM_{2.5}$ ($\mu g/m^3$)	AM ± SD	8.29 ± 4.82	21.25 ± 13.13	17.00 ± 11.04	25.97 ± 17.85	18.15 ± 13.02	10.93 ± 8.91	22.03 ± 8.65	3.19 ± 2.49	12.67 ± 10.48	23.35 ± 17.32	18.51 ± 10.84
	GM ± SD	6.90 ± 1.92	18.16 ± 1.80	13.74 ± 2.00	21.92 ± 1.77	14.38 ± 2.08	7.32 ± 2.89	20.37 ± 1.49	2.50 ± 2.26	9.15 ± 2.50	18.75 ± 1.93	14.70 ± 2.28
	Number	31406	94290	5965	3118	134779	23858	57448	28527	10516	5208	100384
	p-value	<0.001					<0.001					
		a	b	c	d		a	b	c	d	e	
BC ($\mu g/m^3$)	AM ± SD	1.42 ± 1.09	5.86 ± 4.96	4.80 ± 6.27	2.07 ± 1.33	4.64 ± 4.77	1.66 ± 1.54	3.60 ± 2.55	0.45 ± 0.36	5.12 ± 6.83	3.16 ± 2.71	3.32 ± 3.52
	GM ± SD	1.22 ± 1.70	4.54 ± 2.09	3.32 ± 2.37	1.72 ± 1.82	3.18 ± 2.45	1.12 ± 1.16	2.84 ± 2.00	0.37 ± 0.78	3.47 ± 2.46	2.35 ± 2.18	2.31 ± 2.05
	Number	31626	90468	6021	3105	130914	23838	54552	28523	10264	5208	97213
	p-value	<0.001					<0.001					
		a	b	c	d		a	b	c	d	e	
UFP x 10^3(#/cm^3)	AM ± SD	7.60 ± 4.26	21.83 ± 12.35	25.83 ± 19.36	54.28 ± 10.48	19.58 ± 10.24	10.62 ± 8.38	10.92 ± 5.52	3.05 ± 2.96	11.02 ± 8.84	68.53 ± 25.87	14.49 ± 11.78
	GM ± SD	6.49 ± 1.81	14.68 ± 1.81	15.45 ± 2.63	32.91 ± 2.52	12.48 ± 2.07	8.11 ± 2.16	9.71 ± 1.64	2.19 ± 2.34	8.43 ± 2.20	39.10 ± 3.18	10.23 ± 2.08
	Number	20446	62518	3943	2243	88997	11799	40859	28095	6658	3726	65761
	p-value	<0.001					<0.001					
		a	b	c	d		a	b	c	d	e	
CO_2 (ppm)	AM ± SD	485 ± 79	400 ± 72	743 ± 514	576 ± 157	433 ± 223						
	GM ± SD	481 ± 10	395 ± 13	494 ± 20	552 ± 14	417 ± 12						
	Number	11121	42617	2155	1270	57163						
	p-value	<0.001										
		a	b	b	c							
CO (ppm)	AM ± SD	0.00 ± 0.02	0.05 ± 0.30	0.17 ± 0.53	0.15 ± 0.16	0.05 ± 0.30						
	GM ± SD	0.17 ± 1.59	0.25 ± 2.16	0.43 ± 2.61	0.77 ± 1.04	0.26 ± 1.29						
	Number	11120	42614	2155	1270	57159						
	p-value	<0.001										
		a	b	c	b							

AM: arithmetic mean, GM: geometric mean, SD: standard deviation, PAC: portable air cleaner.

p-value: Non-parametric Kruskal-Wallis test for multiple comparisons and Mann-Whitney test for the post-hoc analyses. Different letters indicate different significance; p-value < 0.05 was considered statistically significant.

205

206 The US experienced the highest $PM_{2.5}$ (GM = 20.37 ± 1.49 μg/m^3) at home and UFPs (GM =
207 39.10 x 10^3 ± 3.18 x 10^3 #/cm^3) concentrations in the food court while the BC concentrations (GM
208 = 3.47 ± 2.46 μg/m^3) were most pronounced while commuting to the NUS campus and back. $PM_{2.5}$
209 levels were found to be elevated at home for both OS and US. However, as can be seen in Fig.
210 S4c, during cooking related activities at home or in the food court, a notable increase in the
211 concentrations of UFPs up to 600 x 10^3 #/cm^3 was observed with a decrease in the average PM
212 diameter to 20 nm as compared to about 60 nm in other MEs. Nevertheless, cooking activities at
213 home can also be a significant source of UFPs (Buonanno et al., 2009; See and Balasubramanian,
214 2008; Wan et al., 2011), affecting human health (Kim et al., 2011). The health risk involved with
215 domestic cooking is still poorly understood, and further investigations are warranted. The levels
216 of PM and BC emissions, however, differ depending on the cooking method used (Abdullahi et
217 al., 2013).
218

219 Among the different modes of transport that were used by the participants (e-scooter, walking,
220 cycling, MRT, bus, car, and taxi), exposure concentration to air pollutants (see Table 3) was the
221 highest while commuting by public transport (bus and MRT) and during active modes of transport
222 (walking and cycling) with $PM_{2.5}$ and BC concentrations (GM) being in the range of 12-19 μg/m^3
223 and ~5 μg/m^3, respectively. For public transport, the likely reasons for elevated concentrations of
224 $PM_{2.5}$ and BC were the frequent opening and closure of doors and the migration of outdoor air. In
225 the case of active mobility, the commuter was directly exposed to air pollutants in outdoor air.
226 Rides in cars and taxis with controlled ventilation (mechanical ventilation and filtration)
227 represented the lowest PE with mean $PM_{2.5}$ and BC concentrations being 5-10 and ~3 μg/m^3,

respectively. Our observation indicated that traveling by e-scooter exposed the OS to lower $PM_{2.5}$, BC and UFPs concentrations compared to walking and cycling. This observation could be explained by considering the relatively higher speed of e-scooters compared to the two active modes of transport (walking and cycling), resulting in less exposure time spent by the OS in air pollution hotspots (e.g., traffic intersections, roads with heavy traffic volume). While comparing the concentrations of PM encountered by the OS in motorized (bus, taxi) versus non-motorized (cycling, walking) modes of transport, no statistically significant difference was observed for $PM_{2.5}$ (p-value = 0.86) (see in Table S3). However, significantly higher UFP concentrations (p-value < 0.001) were observed during the non-motorized (GM = $17.14 \times 10^3 \pm 2.26 \times 10^3$ #/cm^3) transport compared to the motorized (GM = $12.85 \times 10^3 \pm 3.23 \times 10^3$ #/cm^3) mode of transport.

Table 3. Personal $PM_{2.5}$, BC and UFP exposure levels by different transport modes.

Metric		Office Staff					University Student			
		E-scooter	Walking	Cycling	Bus	Taxi	Bus	MRT	Car	Taxi
$PM_{2.5}$ ($\mu g/m^3$)	AM ± SD	13.66 ± 7.12	21.34 ± 10.45	19.23 ± 7.31	19.72 ± 6.72	14.27 ± 5.59	13.84 ± 8.05	19.99 ± 6.91	7.59 ± 4.63	10.78 ± 4.46
	GM ± SD	12.27 ± 1.58	16.06 ± 1.72	15.40 ± 1.86	15.39 ± 2.16	9.59 ± 2.60	8.89 ± 2.33	18.41 ± 1.50	4.54 ± 2.87	7.18 ± 2.28
	Number	1392	1280	1183	1236	874	5516	1868	1512	1608
	p-value	<0.001					<0.001			
		a	b	c	b	d	a	b	c	d
BC ($\mu g/m^3$)	AM ± SD	3.98 ± 2.91	4.21 ± 3.52	6.70 ± 2.38	5.52 ± 3.27	3.49 ± 2.81	5.14 ± 3.81	6.61 ± 2.62	3.34 ± 2.67	2.89 ± 1.08
	GM ± SD	2.70 ± 2.40	3.04 ± 2.27	4.16 ± 2.06	4.42 ± 1.94	2.59 ± 2.08	4.14 ± 2.26	4.85 ± 1.78	1.93 ± 1.83	2.17 ± 1.01
	Number	1404	1301	1191	1246	879	5412	1698	1523	1618
	p-value	<0.001					<0.001			
		a	b	c	c	a	a	b	c	d
UFP x 10^3(#/cm^3)	AM ± SD	20.92 ± 10.87	21.85 ± 11.01	25.56 ± 10.56	23.47 ± 16.03	15.29 ± 8.78	11.46 ± 9.50	13.27 ± 2.40	9.79 ± 8.10	9.91 ± 5.98
	GM ± SD	15.82 ± 2.35	18.47 ± 1.71	17.46 ± 2.63	14.17 ± 2.91	11.28 ± 3.63	8.58 ± 2.08	11.40 ± 1.21	6.54 ± 2.70	8.16 ± 2.14
	Number	919	792	816	807	609	3522	444	1100	1580
	p-value	<0.001					<0.001			
		a	b	c	d	e	a	b	c	a
CO_2 (ppm)	AM ± SD	370 ± 36	347 ± 33	390 ± 76	571 ± 212	1515 ± 603				
	GM ± SD	368 ± 11	346 ± 10	386 ± 12	482 ± 26	832 ± 29				
	Number	169	221	758	380	627				
	p-value	<0.001								
		a	b	c	d	e				
CO (ppm)	AM ± SD	0.17 ± 0.05	0.02 ± 0.07	0.11 ± 0.03	0.07 ± 0.09	0.36 ± 0.17				
	GM ± SD	0.48 ± 4.11	0.18 ± 1.64	0.31 ± 2.49	0.31 ± 2.19	0.60 ± 2.39				
	Number	169	221	758	380	627				
	p-value	<0.001								
		a	a	b	a	e				

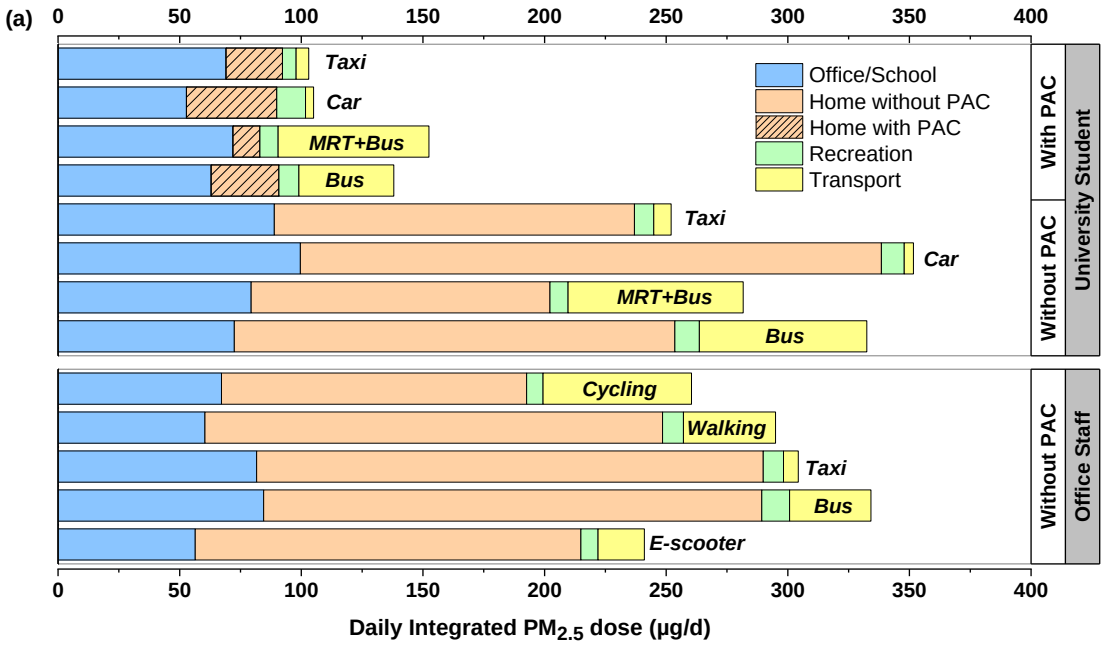
AM: arithmetic mean, GM: geometric mean, SD: standard deviation, p-value: ANOVA significance.

p-value: Non-parametric Kruskal-Wallis test for multiple comparisons and Mann-Whitney test for the post-hoc analyses. Different letters indicate different significance; p-value < 0.05 was considered statistically significant.

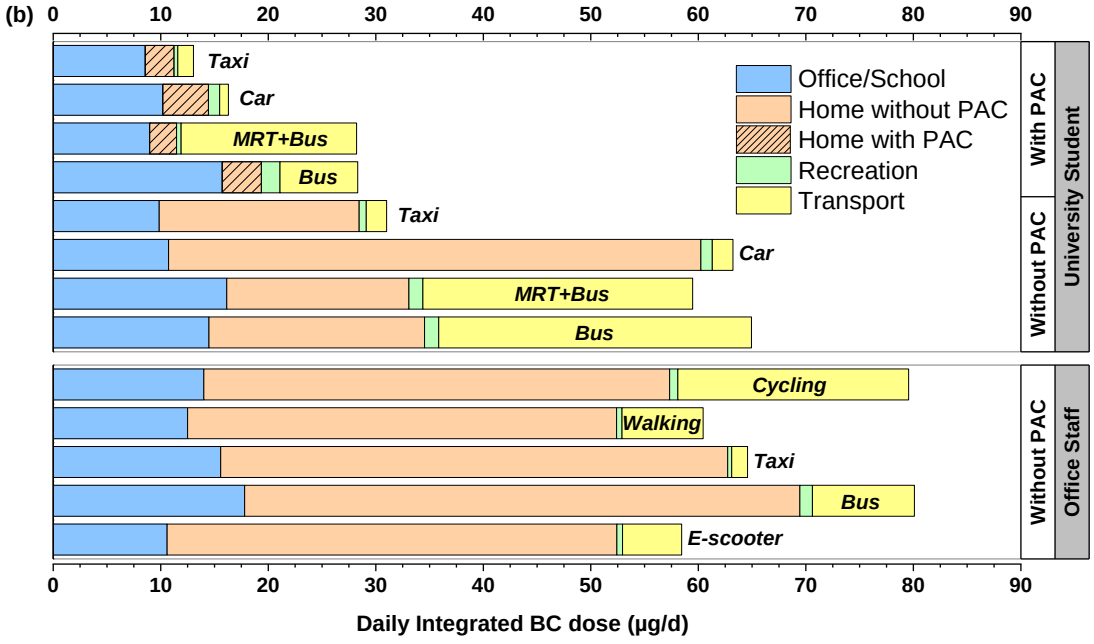
The strong correlation between $PM_{2.5}$ and BC (r = 0.75 and 0.69 for the US and OS, respectively, shown in Fig. S6) indicates the significant role that traffic emissions exert on the indoor air quality (IAQ) as BC is used as a tracer of vehicular emissions. This observation can be explained by the fact that the residential apartments of the OS and the US, and those of citizens in Singapore in general, are in close proximity to roads with high combustion and non-combustion emissions (e.g., tires, brakes and wear of vehicles) of PM from vehicles (Sharma and Balasubramanian, 2019). This observation is consistent with those from other IAQ studies in urban environments where a deterioration in air quality of naturally ventilated indoor spaces occurred due to the influence of outdoor air pollution sources (Challoner and Gill, 2014; Chatoutsidou et al., 2015; Rivas et al., 2014). In Singapore, 99.7%, 81.5% and 2.5% of the buses, taxis, and passenger cars are powered by diesel (LTA, 2018). Consequently, BC is a significant component of $PM_{2.5}$ at traffic-influenced locations as a product of incomplete diesel combustion (Adam et al., 2020; Zhang et al., 2017). Moreover, a moderate correlation between UFPs with CO (r = 0.48) was found, which is further indicative of airborne particles of fossil fuel combustion origin (Kaur et al., 2005; Spinazzè et al., 2013).

When the levels of PE to $PM_{2.5}$, BC and UFPs are considered with the amount of time spent in a particular ME and the type of human activities exerted, the resultant inhaled dose to $PM_{2.5}$, BC and UFPs for both OS and US can be calculated. The daily integrated inhaled dose values for $PM_{2.5}$, BC and UFP to which the OS and US were exposed to are shown in Fig. 2a-c. The corresponding values are given in Table S4. The inhaled dose in the home environment was dominant (up to ~70%) due to a combination of high $PM_{2.5}$ exposure concentrations and the long

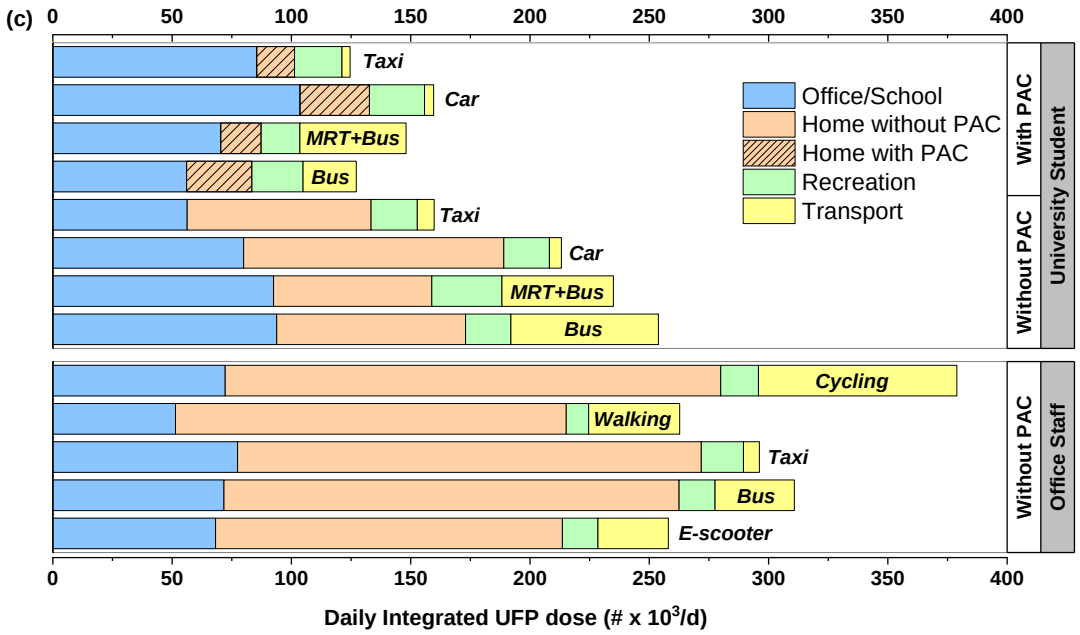
duration of time spent in this ME. Similarly, inhaled dose levels of BC (up to ~80%) and UFPs (up to ~65%) were also elevated in the home environment. The inhaled dose of the two participants varied significantly as a function of the mode of transport. The OS used active modes of transport such as walking and cycling of which cycling had a higher contribution to the daily integrated dose of $PM_{2.5}$ (~23%), UFP (~22%) and BC (~27%) compared to walking; the $PM_{2.5}$, BC and UFP inhaled dose was ~13% during walking. Higher physical activity levels associated with cycling lead to increased inhalation rates, resulting in an enhanced contribution to the daily integrated dose of PM during cycling.


(a)
0
50
100
150
200
250
300
350
400
Taxi
Car
MRT+Bus
Bus
Taxi
Car
MRT+Bus
Bus
Cycling
Walking
Taxi
Bus
E-scooter
Office/School
Home without PAC
Home with PAC
Recreation
Transport
With PAC
Without PAC
University Student
Without PAC
Office Staff
Daily Integrated PM2.5 dose (μg/d)

277


(b)
0
10
20
30
40
50
60
70
80
90
Taxi
Car
MRT+Bus
Bus
Taxi
Car
MRT+Bus
Bus
Cycling
Walking
Taxi
Bus
E-scooter
Office/School
Home without PAC
Home with PAC
Recreation
Transport
With PAC
Without PAC
University Student
Without PAC
Office Staff
Daily Integrated BC dose (μg/d)

278



279

280 **Fig. 2.** Contributions of various MEs to the total daily integrated exposure: (a) $PM_{2.5}$, (b) BC, and
281 (c) UFPs (PAC: portable air cleaner).

282 For the commute by bus, the daily contribution to the inhaled dose of $PM_{2.5}$, BC and UFPs was
283 roughly 10% for the OS. For the US who used either bus or a combination of MRT and bus, the
284 inhaled doses were higher due to longer travel time. The contribution to the daily inhaled dose in
285 terms of $PM_{2.5}$ was relatively less for bus (20%) than for MRT + bus (26%). However, it was
286 similar for both BC (~43%) and UFPs (~20%). In contrast, the commute by car and taxi was
287 associated with the lowest PE and daily integrated dose. The OS used taxis for the commute, and
288 the inhaled dose was clearly the lowest among all modes of transport with contributions of 2% or
289 less for all air pollutants. The US used a private car as well as taxis for the commute to the
290 University. It was observed that the car ride made up less than 3% of the daily dose for all three
291 air pollutants. However, during taxi trips the inhaled dose increased by up to two times for $PM_{2.5}$,
292 BC and UFPs. To standardize the inhaled dose amongst the different modes of transport, we
293 calculated the inhaled dose per unit time (min) and unit distance (km) (Table S5). In general, the

294 inhaled dose values for all modes of transport reflect the pattern described in previous paragraphs
295 due to similar distance and trip times. The travel distance for the OS varied between 3.4 and 3.5
296 km during different modes of transport while for the US it ranged from 20 to 25 km.
297 Correspondingly, the time to commute varied between 24 and 36 min for the OS and between 26
298 and 90 min for the US. The active modes of transport (cycling and walking) are responsible for
299 the highest $PM_{2.5}$ inhaled dose values for the OS with 9.20 and 5.52 μg/km while the taxi ride was
300 associated with the lowest inhaled $PM_{2.5}$ dose values per km (0.89). For the US, the highest inhaled
301 $PM_{2.5}$ dose values were observed for bus and bus+MRT with 1.25 and 1.42 μg/km, respectively.
302 The lowest inhaled $PM_{2.5}$ dose values were obtained for the US when car (0.09 μg/km) and taxi
303 (0.16 μg/km) were used as the preferred modes of transport.
304
305 The active modes of transport (walking and cycling) were associated with higher PE values and
306 inhaled doses of the air pollutants than the other modes of transport. This finding can be attributed
307 to the combination of proximity of the cycling path to the road and thus high emissions of PM
308 from on-road vehicles as well as increased inhalation rate of air pollutants due to the intense
309 physical activity and longer travel time compared to other modes of transport. Nonetheless, the
310 positive effects of physical activity during active commuting might outweigh the negative effects
311 of increased inhaled dose when compared to the passive modes of transport (e.g., car, train), except
312 for the urban environments with elevated concentrations of air pollutants (e.g., $PM_{2.5}$ >100 μgm^{-3})
313 (Cepeda et al. (2017) and references therein).
314
315 In motorized modes of transport, controlled ventilation settings act as physical barriers and
316 facilitate the filtration of airborne particles from the particular ME. However, we observed a

pronounced increase in CO and CO_2 (the products of combustion of fossil fuels) concentrations inside the taxi ME (see Table 3). CO is a known toxic air pollutant. While the accumulation of CO_2 has the potential to impact the mental state of drivers adversely (Allen et al., 2015; Kajtár and Herczeg, 2012; Satish et al., 2012), recent studies, however, have shown that there was no cognitive impairment of CO_2 exposure (Rodeheffer et al., 2018; Scully et al., 2019). The main reasons for the elevated levels of CO_2 inside the vehicle are presumably the exhalation of CO_2 by occupants and the intrusion of polluted air with products of incomplete combustion from surrounding vehicles. The increased exposure concentrations during the bus commute are likely due to high emissions of PM, frequent idling and longer trip time compared to the commute by car and taxi. Also, the opening and closing of the doors to allow passengers to enter and exit the bus at bus stops contributed to an increase in the PE to combustion-related air pollutants (Fig. S5). Similar to active modes of transport, we observed high levels of PE to PM in pollution hotspots on road such as intersections and traffic lights.

3.3. Potential health risks of PE to $PM_{2.5}$

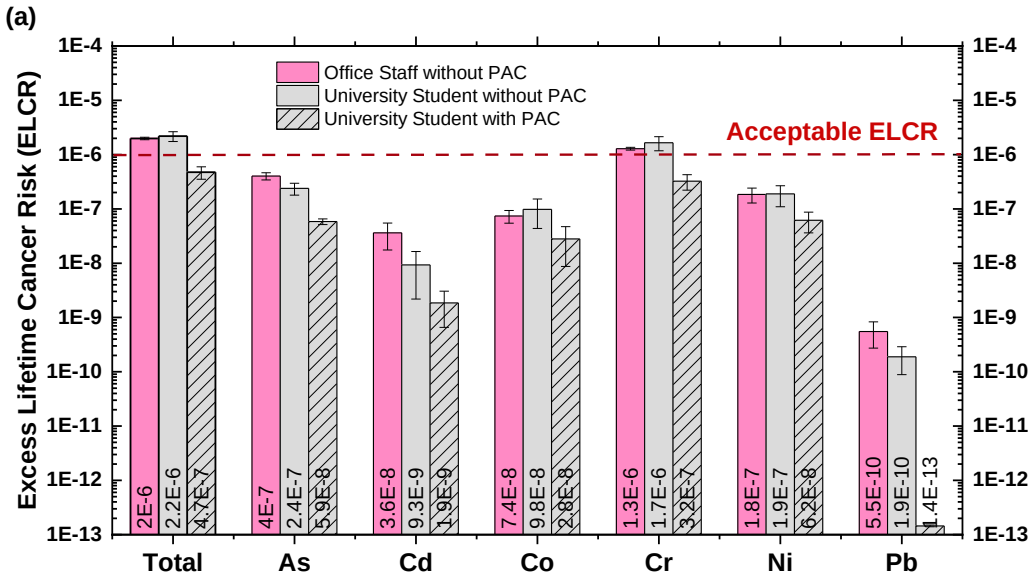
Table S6 shows the mean values and standard deviations of the measured concentrations of 26 PM-bound trace elements (TEs) for different PE scenarios experienced by the two participants. K, Fe, Al, and Zn were the major elements detected under all PE scenarios, accounting for 27%, 24.5%, 11.6% and 10.2% of the total mass of TEs, respectively. Overall, the mean concentrations of TEs over 24-hour exposure to $PM_{2.5}$ were found to be in the range of 0.1 to 2.1 $\mu g/m^3$, which account for 1.2% to 10.6% of the $PM_{2.5}$ mass. Toxic elements such as Fe, Cr, Cu and Zn play a major role in inducing adverse human health effects because of their contribution to oxidative

stress, caused by the formation of reactive oxygen species (Karthikeyan et al., 2006; See et al., 2007; Valavanidis et al., 2008; Velali et al., 2016).

High emission of several TEs appears to be associated with traffic-related sources such as tailpipe emissions from gasoline and diesel vehicles (Al, K) (Kleeman et al., 2000; Xu et al., 2017); re-suspension of road dust (Al, K, Fe, As) (Cheng et al., 2010); brakes and tires (Al, Fe, Ni, Mn, Cu, Cr and Zn) (Garg et al., 2000; Zhang et al., 2017). We observed the highest concentrations of TEs, particularly Fe (751.4 ± 64.8 ng/m^3), Al (173.7 ± 54.3 ng/m^3), Cr (26.0 ± 3.6 ng/m^3), Cu (25.8 ± 3.6 ng/m^3) and Mn (11.8 ± 3.0 ng/m^3), in PM samples collected during the US' ride on MRT + bus.

The potential carcinogenic (Excess Lifetime Cancer Risk - ELCR) and non-carcinogenic (Risk Quotient-RQ) health risk estimated in this study due to PE to $PM_{2.5}$ is shown in Fig. 3a and Fig. 3b, respectively. The health risk assessment outcome is summarized and categorized into three exposure scenarios: OS without PAC, US without PAC and US with PAC operating in home environments. For the carcinogenic risk assessment, the ELCR values of As, Be, Cd, Co, Cr, Ni and Pb were estimated and followed the order from high to low: Cr > As > Ni > Co > Cd > Pb. The total health risk of exposure to six PM-bound water-soluble TEs was generally higher than the acceptable upper limit of 1×10^{-6} for both OS and US without PAC, suggesting that there is a potential carcinogenic risk associated with long-term exposure to elevated $PM_{2.5}$. For a more realistic health risk assessment, other chemical species not determined in this study such as polycyclic aromatic hydrocarbons, quinones and hydroquinones should be considered. Additionally, health risk associated with PM should be evaluated in specific MEs.

We also calculated the potential non-carcinogenic health risk of exposure to $PM_{2.5}$ in terms of the risk quotient (RQ) associated with each participant and each ME as shown in Fig. 3b; the equations used for this calculation are given in the SI. The results indicate that the RQ estimated for both participants does not exceed the safe level (RQ = 1) and is slightly higher for the US compared to the OS. These results suggest that the non-carcinogenic health effects are unlikely to be of concern for both participants. We then assessed the effectiveness of using a PAC as a means to mitigate the PE to $PM_{2.5}$ and associated harmful TEs. The decreased carcinogenic and non-carcinogenic health risk shows the effectiveness of using the portable PM exposure mitigation device. Our results are in agreement with the recent findings reported by Sharma and Balasubramanian (2019); Tham et al. (2018); and Tran et al. (2020c) who found that air filtration by high-efficiency particulate air (HEPA) filters resulted in a substantial reduction in the migration of $PM_{2,5}$ and related TEs of outdoor origin into naturally ventilated indoor environments. This result demonstrates the effectiveness in employing a mitigation device to improve the well-being and health of urban dwellers in an indoor ME that is strongly influenced by emissions of PM from the local traffic. The lowering of ELCR and RQ (> 60 % reduction) is reflected in the decreased percentage of health risk at home.



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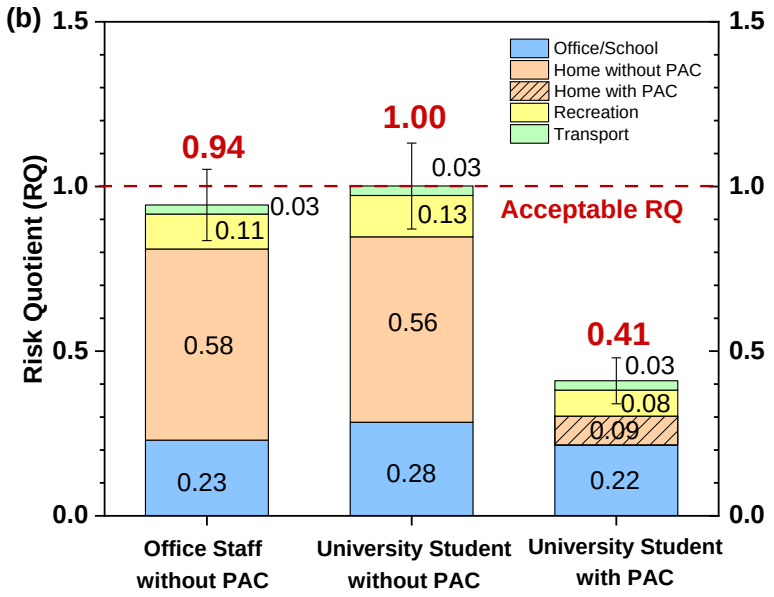


382

383 **Fig. 3.** Potential (a) carcinogenic and (b) non- carcinogenic health risk estimates of $PM_{2.5}$

384 (ELCR: excess lifetime cancer risk, PAC: portable air cleaner).

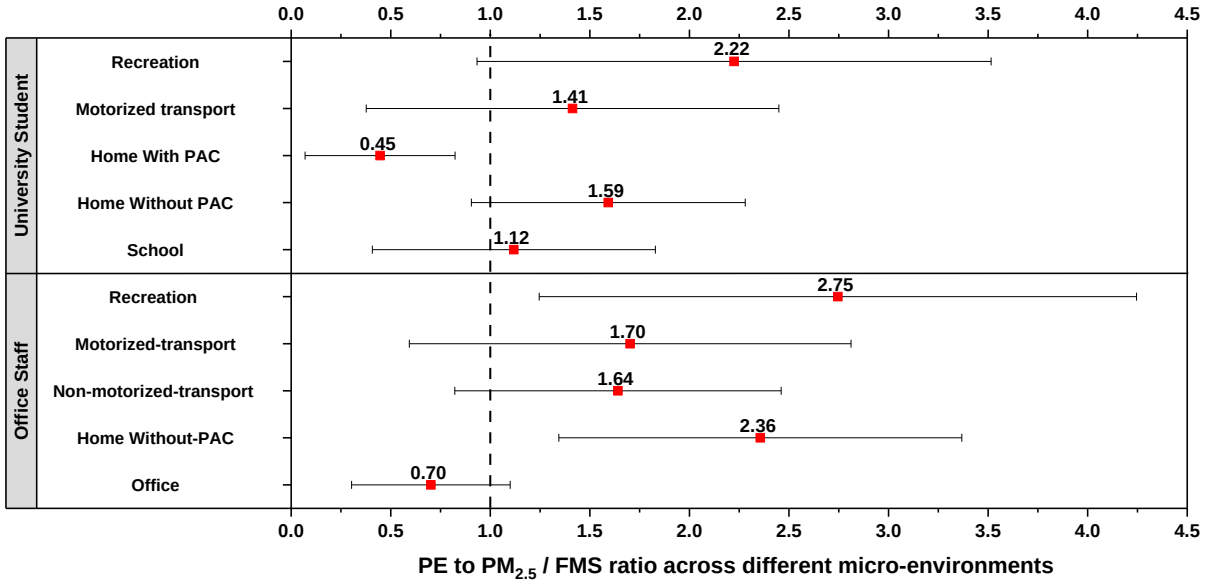
385

386

For both participants, the office/classroom (air-conditioned spaces) represented the cleanest MEs in terms of $PM_{2.5}$, BC and UFPs PE concentrations. This is mainly attributable to the air filtration of incoming outdoor air and the absence of major indoor air pollution sources (Clausen, 2004; Zhang et al., 2011). Thus, in densely populated cities where residential homes are built in close proximity to roads, an economically viable and relatively easy-to-implement approach to reducing PM exposure and associated health risks at home is to deploy a PAC. A major limitation of the carcinogenic health risk assessment is that the collection of PM took place over 24 hours and thus the health risk could not be apportioned to any particular ME. However, TEs that are representative of traffic-related emissions (Zhang et al., 2017) were found in all samples with ELCR values of water-soluble Cr exceeding the acceptable limit. As a consequence, the total carcinogenic health risk is well above the acceptable limit, indicating that the daily PE of city dwellers in the various MEs is associated with significant health risks and curtailing traffic emissions in urban environments is advisable.

$PM_{2.5}$ mass concentrations obtained from the FMS closest to the participants in Singapore (South) reported by NEA Singapore (NEA, 2019) and those from PE measurements correlate poorly ($r = 0.23$, shown in Fig. S7). The FMS $PM_{2.5}$ concentration data are consistently lower than the PE-derived data, which points to the underrepresentation of health risk associated with the inhalation of fine particles when health risk assessment is conducted with data from FMS stations. In order to gain insights into the protection provided by buildings and enclosed spaces in terms of reduced PE to PM, we computed the ratio of PE measured in this study to FMS-derived $PM_{2.5}$ concentrations (see Fig. 4). As can be seen in Fig. 4, the two study participants (OS and US) experienced the lowest PE in the MEs equipped with PE mitigation devices such as PAC and AC

(an air-conditioning system containing PM filter). This PE reduction to $PM_{2.5}$ is attributed to PM filters being used to remove airborne particles in indoor air. For example, the PAC was equipped with a HEPA filter on each side while the AC system contained a low-grade PM filter (Minimum Efficiency Reporting Value 7 (MERV-7) filter). As a consequence, the home ME has a lower ratio of PE-to-$PM_{2.5}$ to FMS-derived $PM_{2.5}$ (0.45) compared to the office ME (0.70). High PE ratios corresponding to low mitigation of PE to $PM_{2.5}$ are found in the recreation (2.22-2.75) and home environments without PAC (1.59-2.36) for the US and OS, indicating that building occupants are exposed to significantly high $PM_{2.5}$ levels. Overall, our findings highlight the significance of the assessment of PE to airborne particles in both indoor and outdoor MEs to obtain a comprehensive understanding of health risks associated with city dwellers during their daily routine. It should be noted that this pilot study involved the participation of only two subjects. Comprehensive studies with participation of more volunteers in different age groups with diverse lifestyles are warranted to provide epidemiologists and public health officials with extensive PE data and health risk estimates related to $PM_{2.5}$.



426

427 **Fig. 4.** Personal exposure to $PM_{2.5}$ and its variability across different microenvironments in relation
428 to $PM_{2.5}$ obtained from FMS. *PE: Personal Exposure; FMS: Fixed Monitoring Station.*

4. Summary and Conclusions

We investigated time-activity-based personal exposure to airborne particles in diverse indoor and outdoor MEs and its potential health impacts in Singapore, the densely populated city-state, for two months during the dry season with the participation of two volunteers (OS and US). Both OS and US used active (walking and cycling), non-emitting motorized (e-scooter), and motorized (MRT, bus, car, and taxi) modes of transport to commute from their home to the University and back. While the US assessed the PE to PM at home with and without indoor air filtration using a portable air cleaner, the OS did the exposure assessment under natural ventilation conditions. We studied the total PE to $PM_{2.5}$, UFPs, and BC continually using hand-held devices and GPS coordinates. We also collected $PM_{2.5}$ samples gravimetrically and estimated the potential carcinogenic (ELCR) and non-carcinogenic (RQ) health risk based on PE to $PM_{2.5}$-bound water-soluble toxic trace elements and total $PM_{2.5}$ concentrations, respectively.

The mode of transport, travel distance, travel route and the time spent in different MEs had a significant effect on the PE to $PM_{2.5}$, UFPs and BC and their inhaled doses. Among all MEs, the maximum PE occurred in home environments with emissions from on-road traffic-related sources that likely influenced indoor air quality and related PE. In outdoor MEs, urban road transport and food courts involved with intense cooking activities also exhibited elevated levels of $PM_{2.5}$, UFPs and BC. Cooking-related exposure to particulate pollution can be mitigated with the use of effective local exhaust. Fossil fuel combustion emissions of PM from the on-road transport deserve great attention from the PE mitigation viewpoint. The Singapore government has plans to phase

out all fossil fuel-driven automobiles by 2040. Until then, short-term measures need to be explored to mitigate the PE to TRAP. The use of a PAC with a suitable size and PM removal effectiveness, as defined by CADR, would reduce the exposure of indoor building occupants to both outdoor- and indoor-derived PM and thus related health risks. Other exposure mitigation strategies such as the use of ACMV equipped with suitable PM filters and fan filter units operated in an energy-efficient mode can also be explored. Finally, future city-scale air quality monitoring should focus on citizens-oriented research studies involving the assessment of PE to air pollution to enhance our scientific knowledge and contribute to healthy living in cities through effective air pollution control policy development. Such a program, if successfully implemented, will provide tangible environmental and health benefits as has been demonstrated in this exploratory study with the participation of two volunteers.

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