Particle Exposure during the 2013 Haze in Singapore: Importance of the Built Environment

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Abstract

The 2013 haze was the most serious air pollution event in Singapore's history. Individual exposures to particulate matter differ (a) according to time patterns of behaviour and (b) with the varying degrees of protection provided by buildings against penetration and persistence of outdoor particles. Utilizing real-time personal monitoring, we evaluated exposures to size segregated fine particulate matter (PM) of five office workers for six days during the latter portion of the 2013 haze event. The outdoor volume concentrations of particulate matter (0.3-2.5 µm diameter) during moderate and light haze days were in the ranges 15-21 µm^3/cm^3 and 7-10 µm^3/cm^3, respectively. More than 80% of total daily exposures occurred indoors in workplaces and residences. The daily-integrated personal exposures for the five subjects were 140-454 (µm^3/cm^3)-h and 66-239 (µm^3/cm^3)-h, respectively. Exposure factors for the five participants, quantifying the extent of exposure reduction associated with being indoors, had daily average values ranging from 0.32 to 0.75. The results of this study contribute toward deeper understanding the degree of protection provided by the buildings from pollution of outdoor origin. The work also contributes knowledge for estimating personal exposure to particulate matter during air pollution episodes especially in tropical climates.

1. Introduction

During the annual south-southwest monsoon, large amounts of airborne particulate matter (PM), generated from landscape fires, are transported across Southeast Asia (SEA), including Singapore [1-3]. The duration and intensity of the annual haze varies. Events that caused high levels of pollution in Singapore occurred in 1994, 1997, 2006, and 2013 [2]. The 2013 haze episode was the most serious [4, 5]. It was the first time that Singapore's Pollutant Standards Index (PSI) reached the "hazardous" range. The highest PSI reading, 401 (on a 3-h average basis), was recorded on 21 Jun 2013 [6].

Particulate pollution associated with landscape fires poses health risks for those exposed. Environmental health concerns owing to particulate matter exposure are well established [7]. More specifically, surges in outpatient admissions because of health outcomes that could be haze-related — such as rhinitis, asthma, and respiratory infections — were reported in Singapore during the 1997 haze [8] and again in 2013 [9].

Several studies have documented the ways in which landscape-fire induced smoke affects urban air quality. For example, studies in Singapore have found that haze events are associated with pronounced deterioration in the chemical, physical, radiative, and toxicological properties of outdoor air [3, 4, 10, 11].

Two studies of the 2013 haze event in Singapore included an exposure assessment for evaluating carcinogenic risk [4, 12]. Inhalation exposures were estimated directly from fixed-station outdoor measurements. Unresolved in such investigations is how well ambient measurements represent actual human exposure to haze-associated particulate matter.

It is well known that most people spend most of their time indoors. It is also well known that — for many pollutants — levels in indoor air do not match those in outdoor air. Specifically considering particulate matter, Kousa et al. [13] and Arhami et al. [14] investigated the correlation between outdoor, indoor, and personal exposure concentrations. Their results indicate that the outdoor concentration would not be a good predictor of personal exposures to particles. Studies that utilize direct method of personal monitoring could contribute important information to better understand exposure during serious air pollution events such as the episodic haze experienced in Southeast Asia.

The application of direct measurements of personal exposure to air pollutants utilizing real-time instruments is a relatively recent development. Wheeler et al. [15] reported one of the best examples in this class of investigation, acquiring an aggregate total of 325 days of real-time $PM_{2.5}$ monitoring data from approximately 45 asthmatic children. Real-time exposure monitoring also has been applied in recent years to traffic-associated particulate pollution, as in Apte et al. [16] in which about 180 hours of time-resolved data on $PM_{2.5}$, particle number concentration, and black carbon were acquired along a repeatedly travelled route in New Delhi.

Direct measurements of personal exposure to particulate matter during haze periods are scarce, in part because of the episodic nature of haze events and in part owing to historic instrumentation limitations. The only prior study of personal exposure to haze particles was reported by Muraleedharan and Radojevic [17]. Their work has the character of a preliminary investigation, presenting anecdotal and illustrative data from their $PM_{2.5}$ monitoring campaign.

We are aware of no prior studies that utilize personal monitoring of fine particles (<2.5 μm in diameter) with particle size resolution. The health risks associated with particulate matter exposure vary with particle size, an attribute that strongly influences penetration and regional deposition in the respiratory tract [18]. It is also well established that particle size influences substantially the extent to which particles of outdoor origin penetrate and persist indoors [19, 20]. Therefore, a better understanding of size-resolved exposure to particulate matter during haze periods would represent a useful contribution of knowledge as a basis for assessing associated health impacts and for developing effective interventions.

The present study emerged from the context summarized above, that no published studies provide data on particle size-resolved exposure during haze conditions utilizing direct personal monitoring. The work reported in this paper has three specific aims: (i) to assess daily-integrated personal exposures for a set of subjects to fine particles during the 2013 haze period in Singapore; (ii) to characterize the influence of indoor microenvironments on size-resolved particle exposures for those subjects; and (iii) to assess the overall level of protection provided by ordinary buildings in Singapore from particle exposure during the 2013 haze period.

2. Methods

2.1 Study design

Daily exposure measurements were conducted over a two-week period during June-July 2013. For the analysis in this paper, we consider two time blocks, each comprising three consecutive weekdays. During the first period (25-27 June 2013, with days labelled D1-D3), the haze level was moderate (24 h-$PM_{2.5}$ = 32-63 $\mu g/m^3$ as reported by Singapore's NEA). During the second period (2-4 July 2013, with days labelled D4-D6), the haze level was light (24 h-$PM_{2.5}$ = 18-34 µg/m^3). By focusing on ordinary workdays, the results reflect fairly consistent patterns of activities for each of the subjects. Because this project was devised and executed in response to the onset of the heavy haze, and because some time was required to mobilize the monitoring effort, sampling did not include the heaviest portion of the haze period, but did start shortly thereafter (see Figure 1).

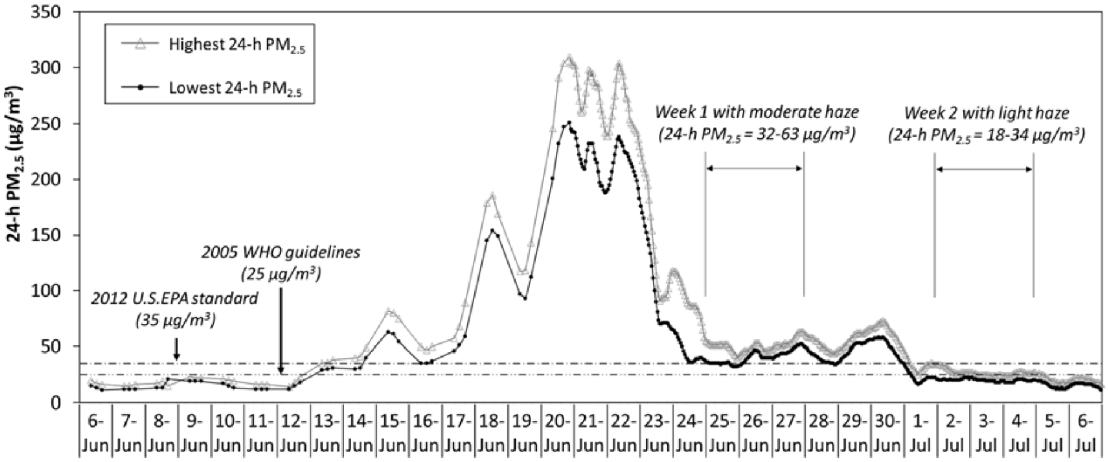


Figure 1. Time series plots of the regional 24-h $PM_{2.5}$ concentrations for 1-month (6 June-6 July) during 2013 haze episode in Singapore. Traces represent the highest and lowest running-average 24-h concentrations, as reported by Singapore National Environmental Agency [21]. The $PM_{2.5}$ concentrations were measured by beta attenuation mass monitor. The USEPA standard (35 µg/m^3, on a 24-h average basis) [22] and the WHO guideline (25 µg/m^3, on a 24-h average basis) [23] are noted.

Throughout the monitoring period, participants conducted regular daily activities. Each subject carried a portable optical particle counter (OPC), placing the device beside himself or herself while seated or sleeping. A log sheet was maintained by each subject to record the times and activities in primary microenvironments. We grouped the locations and activities into four categories: at work (W), in one's own residence (R), commuting (C), and other (O). The participants also were asked to note any activity or condition that might influence particle exposure, such as cooking or the extent to which doors and windows were open in their residence.

Twelve individuals participated in the sampling for most of a two-week period. After reviewing the activity log sheets and the particle sampling records for completeness, we identified five subjects (labelled S1-S5) who had sufficiently reliable and complete data to include in the analysis reported here. These participants comprised two males (S1 and S2) and three females (S3, S4, and S5). Ages were in the range 25 to 60 years. All participants worked in offices on a full-time basis during the monitoring period. We note that the direct monitoring approach employed here would be difficult to apply on a large scale in part because of instrument cost. However, availability of time-resolved monitoring results permits attribution of exposure to different microenvironments in a manner that cannot be realized with the more widely used time-integrated personal exposure sampling approach.

In addition to the instruments carried by subjects, two additional OPCs (labelled A and B) were operated at local outdoor monitoring sites to record the outdoor particle levels on a continuous basis. We used the results from A to represent outdoor conditions for two of the participants (S1 and S2) who spent their time within 5 km of that outdoor station. The results from B were applied to characterize outdoor conditions for the other three participants, who remained within 3 km of that outdoor station throughout the monitoring period.

2.2 Instrumentation and quality assurance

Temperature, relative humidity, and size-resolved particle number concentrations (PNCs) were recorded in real-time employing 1-min sampling intervals and utilizing portable OPCs (model Handheld 3016, Lighthouse Worldwide Solutions, Fremont, CA, USA; and model

Aerotrak 9000, TSI Inc., Shoreview, MN, USA). The instruments recorded particle number concentrations in six size bins (0.3-0.5, 0.5-1.0, 1.0-2.5, 2.5-5.0, 5.0-10.0, >10.0 μm optical diameter). To emphasize haze particles of greatest health concern and to minimize confounding by particle resuspension indoors, we focus here on the data recorded in the three smallest size channels, covering the diameter range 0.3-2.5 µm.

For quality assurance, side-by-side (SBS) tests of the OPCs were conducted in both indoor and outdoor environments to span a range of concentrations. The SBS procedure has been reported by Bhangar et al. [24]. Consistent with personal and outdoor monitoring experiments, the SBS tests employed 1-min sampling intervals and focused on the data recorded in the three smallest size bins (0.3-0.5 µm, 0.5-1.0 µm, 1.0-2.5 μm). We used two optical particle sizers (OPSs) (model 3330, TSI Inc., Shoreview, MN, USA) as the reference instruments. The factors summarized in Table S1 of the Supporting Information were used to adjust data from OPCs to match as closely as possible the mean response of the two OPSs, so as to minimize the bias in ratios of concentrations measured by various instruments during the study.

From the three channels of particle number concentration data, we estimated the aggregate particle volume concentration in the diameter range 0.3-2.5 µm. The procedure, described in the Supporting Information, assumes that the volume-weighted distribution, $dV/d(\log d_p)$, is uniform in relation to the log of particle diameter within each size section. Given the limitations of the instruments, this metric provides a reasonable aggregate measure of particles within the three size sections that best corresponds to the mass concentration of $PM_{0.3-2.5}$. For example, if the particle density were known to be 1 g/cm^3, then the numerical value of volume concentration in µm^3/cm^3 would correspond to a $PM_{0.3-2.5}$ mass concentration of the same numerical value in units of µg/m^3. As shown in the Supporting Information, the effective mean particle diameters for the three size sections (0.3-0.5 μm, 0.5-1.0 μm, 1.0-2.5 μm) are 0.38 μm, 0.67 μm, and 1.43 μm, respectively.

We report particle levels in volume concentration rather than mass concentration because we lack information on the average density of airborne particles. A check was made using gravimetric analysis of a 24-h filter sample collected with a $PM_{2.5}$ cyclone on 27 June (day = D3). The mass concentration of $PM_{2.5}$ would match the optically determined volume concentration of $PM_{0.3-2.5}$ if the density were 1.7 g/cm^3. Since the expected value of fine particle density would lie in the approximate range 1.0-2.5 g/cm^3, this comparison provides some reassurance about the comparability of our volume-concentration results with the more commonly reported $PM_{2.5}$ mass concentrations.

2.3 Monitoring and data analysis protocols

For each midnight-to-midnight 24-h period, we assessed as a time series, with 1-min resolution, each participant's exposure to particulate matter. We computed from the time series data the time-integrated daily exposure for each monitoring day. We expressed this exposure in units of volume concentration multiplied by time (μm^3-h/cm^3). For example, continuous exposure to 10 μg/m^3 of particulate matter with an average particle density of 1 g/cm^3 for 24 hours would produce a daily exposure of 240 μm^3-h/cm^3.

Each participant maintained a simple log of their activities and, based on the information in the log sheets (supported by the measured temperature, humidity, and particle concentration time series), we apportioned the daily exposure into the components that were attributable to four microenvironment/activity conditions: at work, at home, in commute, and other.

Equation (1) describes how the concentration data were converted into a daily particulate-matter volume exposure in units of μm^3-h/cm^3.

$$E_{PM0.3-2.5}^{daily} = \int_0^{24} C^{PM0.3-2.5}(t)dt \tag{1}$$

Where $E_{PM0.3-2.5}^{daily}$ is the daily integrated exposure, $C^{PM0.3-2.5}(t)$ is the instantaneous exposure concentration of particulate matter in the size range 0.3-2.5 µm, and *t* is the time of day, in units of hours. Total daily exposure and exposures in each of the four microenvironments were also decomposed into three particle size bins as provided by the optical particle counters: 0.3-0.5 µm, 0.5-1.0 µm, and 1.0-2.5 µm. Hence, for each of the five subjects and for each of the six days, the total exposure was represented as the sum of twelve components, which reflected three particle size bins in each of four microenvironments.

An important goal of this study is to quantify how much protection buildings provide from exposure to haze-associated particulate matter. We quantified the level of protection by defining and evaluating a microenvironmental exposure factor, EF_k^j, specific to each microenvironment j and particle size k. As shown in Equation (2), the exposure factor was evaluated as the ratio of the measured exposure that occurred in the given microenvironment, $Empirical\ E_k^j$, to the theoretical exposure that would have occurred if the person were exposed to the particle level measured at the outdoor monitoring station, $Hypothetical\ E_k^j$.

$$EF_k^j = \ Empirical\ E_k^j / Hypothetical\ E_k^j \tag{2}$$

An exposure factor of 1.0 indicates that the exposure determined from personal monitoring is the same as would have occurred at the outdoor station. An exposure factor of 0 implies that the particular microenvironment provides perfect protection against exposure to haze particles in the 0.3-2.5 µm size range.

A premise throughout the analysis is that indoor particle emission sources contribute little to exposure on the study days so that the exposure factor indicates the degree of protection afforded by being in a particular microenvironment against particles of outdoor origin. The information gathered in this study supports this view, although it does not prove it conclusively. All indoor environments were non-smoking. Log sheet data combined with particle monitoring results indicate that cooking would only have made a minor contribution to the measured exposures. The general consistency of exposure factors between monitoring week one (with moderate haze) and week two (with light haze) further supports the inference that particle exposures in indoor environments were primarily a consequence of the penetration and persistence of outdoor particles. The high time resolution of the monitoring data allows for these inferences to be made with confidence; such determinations would not be possible with time-integrated sampling.

3. Results and Discussion

3.1 Microenvironment characteristics and daily time activities

Table 1 shows the average apportionment of daily time for each participant. All participants spent significant amount of time at work (W) and in their residence (R), with the daily average value at 10.6 h (44% of the day) and 12.1 h (50% of the day), respectively. The daily time spent in commuting is comparable across all participants with an average value of 1.2 h per day (5% of the day). In comparison with cities in the United States, the subjects in our study spent typical amounts of time in commuting, but with a larger percentage using public transport.

Table 1. Description of participants (ID) and microenvironments (μE) for six study days. [a]

ID	μE	Time (h) Ave/SD	Location, configuration, cooling and ventilation conditions
S1	**W**	10.3/1.4	High-rise building near station A; a shared office room; two doors lead to internal hallway; windows on four walls; cooling by central air-condition system; mechanical ventilation with MERV 7 filter.
	R	12.6/1.4	High-rise building near station A; a single bedroom rented from a flat; one door leads to the sitting room; one window toward the traffic; no cooling; natural ventilation.
	C	1.1/0.2	By bus. The subject took bus twice daily (daily time in bus cabin: 20-30 min).
	O	NA	NA
S2	**W**	8.7/1.2	Same office with S1.
	R	13.9/1.1	High-rise building near station A; a double bedroom in a flat; one door leads to the sitting room; one window toward the traffic; cooling by central air-condition system; mechanical ventilation; filter grade unknown.
	C	1.4/0.3	On foot (including brief period walking outdoors during lunch). Part of the walking trail is alongside heavily traveled roadways.
	O	NA	NA
S3	**W**	12.3/1.0	High-rise building near station B; a shared office room; one door leads to outdoor hallway; windows on two walls; cooling by central air-condition system; mechanical ventilation with MERV 7 filter.
	R	10.9/0.9	High-rise building near station B; a twin shared bedroom rented from a flat; one door leads to the sitting room; one window toward the traffic; cooling by split-system air-conditioning unit; no mechanical ventilation.
	C	0.9/0.2	By bus. The subject took bus twice daily (daily time in bus cabin: 10-20 min) on D1 and D5, but once a day (daily time in bus cabin: 5-10 min) on other days.
	O	NA	NA
S4	**W**	9.5/3.2	High-rise building near station B; a shared office room; one door leads to outdoor hallway; windows on two walls; cooling by central air-condition system; mechanical ventilation with MERV 7 filter.
	R	12.4/2.2	High-rise building near station B; a single bedroom rented from a flat; one door leads to the sitting room; one window toward park; no cooling; natural ventilation.
	C	1.5/0.8	By private passenger car.
	O	1.5/NA	Site visit at a basement car park near station B on D1; no cooling; mechanical ventilation.
		1.8/NA	Site visit at a commercial building near station B on D4; entrance near bus station; no operable windows; cooling by central air-condition system; mechanical ventilation.
S5	**W**	12.0/2.8	High-rise building near station B; a shared office room; one door leads to outdoor hallway; no operable windows; cooling by central air-condition system; mechanical ventilation with MERV 13 filter.
	R	10.7/2.5	High-rise building near station B; a double bedroom from a flat; one door leads to the sitting room and one window toward traffic; cooling by split-system air-conditioning unit; no mechanical ventilation.
	C	0.9/0.1	By bus. The subject took bus twice daily (daily time in bus cabin: 20-30 min).
	O	1.0/NA	Dining at canteen near station B; semi-indoor; no cooling; no mechanical ventilation.
		0.4/NA	Shopping at supermarket near station B on D4; entrance near outdoor car park; no operable windows; cooling by central air-condition system; mechanical ventilation, filter grade unknown.
		0.8/NA	Dining at canteen near station B on D5; indoor; entrance leads to outdoor hallway; no operable windows; cooling by central air-condition system; mechanical ventilation.

[a] Microenvironment codes: W = work, R = residence, C = commute, O = other. NA = not applicable.

Each of the working environments in this study was equipped with a centralized air conditioning and ventilation system utilizing MERV 7 particle filters, except that the office where S5 works utilized a higher grade of filters (MERV 13). Among the residential environments, conditions varied broadly. As is common in Singapore, all five subjects lived in high-rise residential buildings. However, only the dwelling occupied by subject S2 was equipped with mechanical ventilation. The four other subjects lived in apartments with doors and operable windows to provide occupant-regulated natural ventilation. Residences S1 and S4 did not have air-conditioning, so the normal practice was to have windows open during occupancy. However, for protection against the high outdoor particle levels, windows were sometimes closed during the moderate haze period. Windows in these units were open when the rooms were occupied and the haze was light. The residences occupied by subjects S3 and S5 were equipped with split-system air-conditioning units that provide for cooling but not ventilation. As is common in Singapore's tropical climate, when these residential bedrooms were occupied, windows and doors were closed and the air conditioning units were on.

3.2 Particle concentrations

Figure 2 illustrates the time series plots of the size-resolved outdoor particle concentrations at station A and the exposure concentrations for participants S1 and S2 on the first monitoring day (D1), 25 June 2013. Analogous plots for each of the five participants and for each of the six monitoring days are presented as Figures S1-S12 in the online Supporting Information.

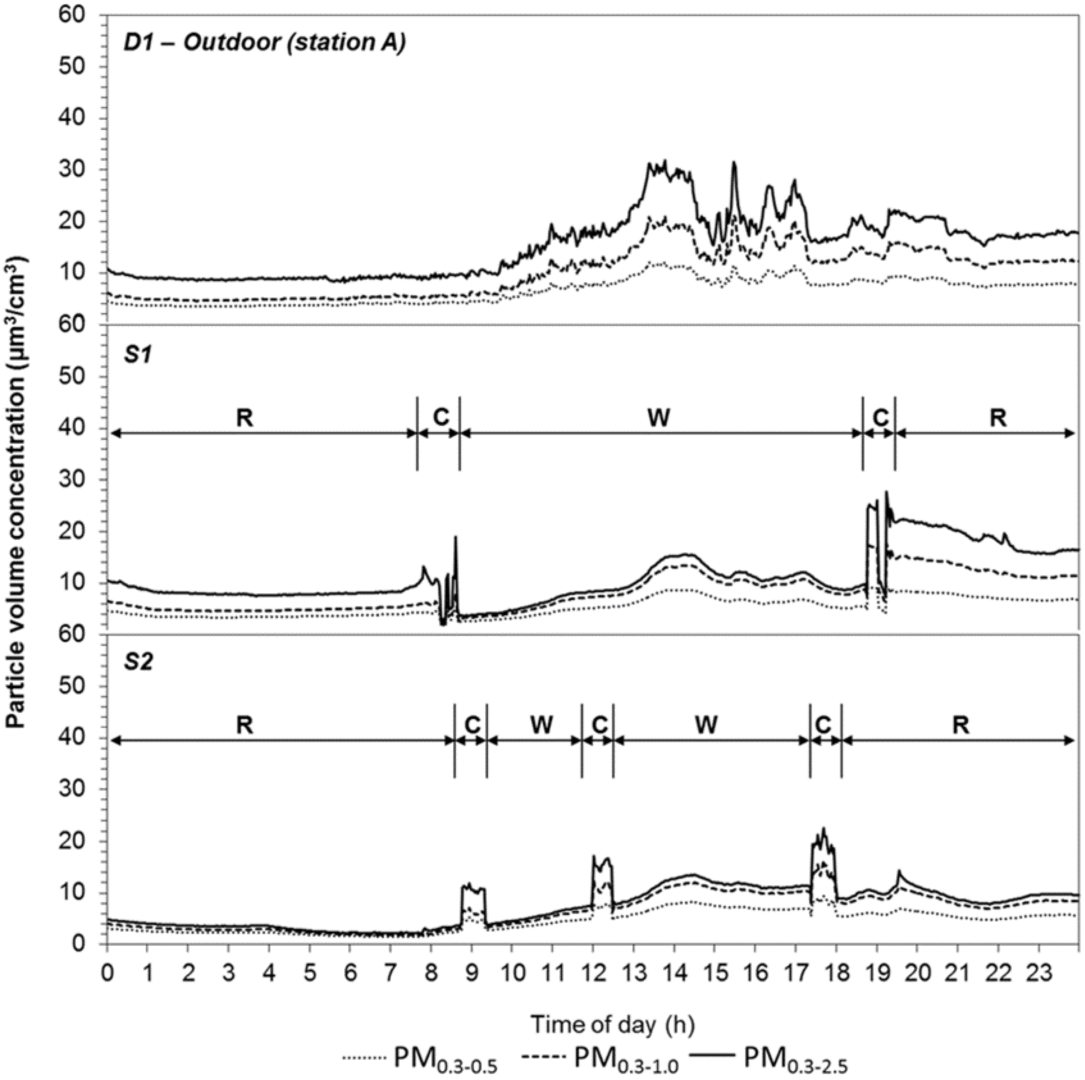


Figure 2. Time series plots of the size-resolved outdoor particle concentrations at station A and the exposure concentrations for participants S1 and S2 on the first monitoring day (D1, 25 Jun 2013). The daily activities are noted in bottom two plots, which present measured exposure concentrations (R = in residence, C = commuting, W = at work).

3.2.1. Outdoor particle concentrations

Consistent with the lower haze levels during the second week of monitoring as compared to the first (Figure 1), our measurements showed higher daily average particle concentrations at both outdoor monitoring stations during the first monitoring week (D1-D3) than during the second (D4-D6). Daily $PM_{0.3-2.5}$ concentrations during the moderate haze period ranged from 15 to 21 $\mu m^3/cm^3$, with an average across the three days of 18 $\mu m^3/cm^3$; the value during light haze days ranged from 7 to 10 $\mu m^3/cm^3$, with an average of 9 $\mu m^3/cm^3$. Our outdoor monitoring data corresponded well to the $PM_{2.5}$ data reported by Singapore NEA [21]. The NEA regional data from two stations close to the participants (South and West stations) indicated that the daily $PM_{2.5}$ mass concentrations during the first week (in the range 39 to 62 $\mu g/m^3$, with an average of 47 $\mu g/m^3$) were twice those during the second (in the range 19 to 27 $\mu g/m^3$, with an average of 23 $\mu g/m^3$). The regional haze maps [25] (Figure S13) also indicate that Singapore was affected by moderate haze during first monitored week, with a reduction to light haze during the second week of the monitoring period.

3.2.2. Exposure concentrations

Personal exposure concentrations varied primarily according to two factors: (a) what level of protection the buildings provided (b) what were the outdoor particle levels. Consider, for example, the concentrations to which participant S1 was exposed on D1 (see Figure 2, middle frame). When at home, the concentration profiles were similar to the outdoor levels. The concentration profile encountered in the working microenvironment followed the broad trends of the outdoor concentration, but with attenuation because of the protection provided by the central air-conditioning and mechanical ventilation system. Short-term fluctuations in the outdoor particle concentration trace are dampened in the exposure concentration plot, which reflects the capacity associated with the building volume.

One expects better filter efficiency as well as higher deposition rates for particles in the 1.0-2.5 μm diameter range than for 0.3-0.5 μm particles. This expectation is revealed in comparing the outdoor concentrations to the work microenvironment data in Figure 2. The figure shows a proportionately larger reduction in the indoor concentrations in the largest size section as compared with the smallest size section. For subject S1, the data also show a few sharp peaks in concentrations during commute when the subject was situated outside of the bus cabin and directly exposed to outdoor air.

Subject S2 was in air-conditioned and mechanically ventilated space both in the residence and at work, and the particle data show a consistent attenuation with respect to the outdoor concentrations during these times. Except for a brief period during lunch, the concentration trace is comparable to that observed for participant S1 during the working period. Elevated concentrations were seen for subject S2 during the walking-based commute, which included some time alongside a busy roadway.

Overall, during the working and residential periods in the six monitoring days, the concentration time series from personal monitoring followed the broad trends in the outdoor concentration, either at comparable or diminished level, depending on the effectiveness of particle removal processes in the occupied microenvironments. During the periods when subjects were at home, we observed only a few episodic deviations from the overall consistent pattern. These excursions were probably attributable to the effect of human activities on emissions or resuspension into the indoor air. Overall, these deviations had only minor impact on the time-integrated exposures of the study subjects.

Four of the participants (all except S2) traveled in public or private vehicles during commute between residence and work. We observed many short-term fluctuations in the time series of exposure concentrations during the commuting period. In part, these fluctuations appear to occur because of the frequent switching between being inside and outside of an air-conditioned automobile or bus passenger cabin.

There were a few instances when the exposure concentration trace was consistently higher than the corresponding outdoor profile for an interval. These included (i) S2 walking along the roadside on each of the six monitoring days; (ii and iii) S4 doing site visits at a basement car park on D1 and in two commercial buildings on D4; (iv and v) S5 having dinner at a semi-open canteen on D4 and at an air-conditioned canteen on D5. The higher values of exposure concentrations during these episodes likely reflect contributions of motor vehicle and cooking emissions superimposed on the haze particles. Overall, however, episodes such as these were evident in the time-series data less than 1% of the total time sampled and contributed only a few percent (< 5%) to total exposure.

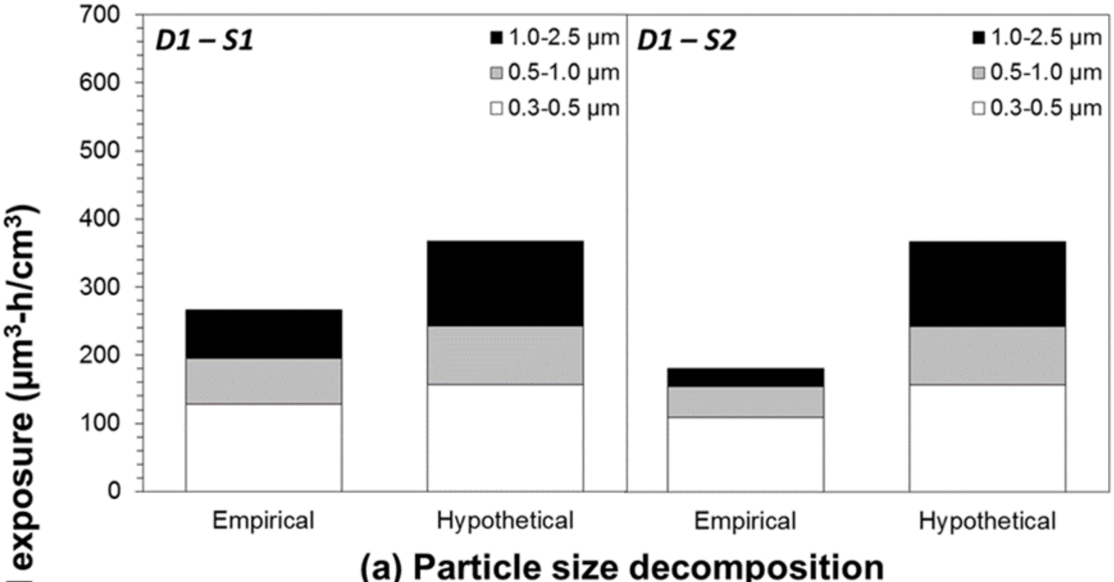
3.3 Exposures

Figure 3 displays empirical (left bar in each frame) and hypothetical (right bar in each frame) exposures for subjects S1 and S2 on day D1 (25 June 2013). The top two frames disaggregate exposures according to particle size, and the bottom two frames show how exposures decompose according to microenvironment. Analogous plots showing particle size and microenvironmental decomposition for each of the participants and for each of the monitoring days are presented in the Supporting Information (Figures S14-S24).

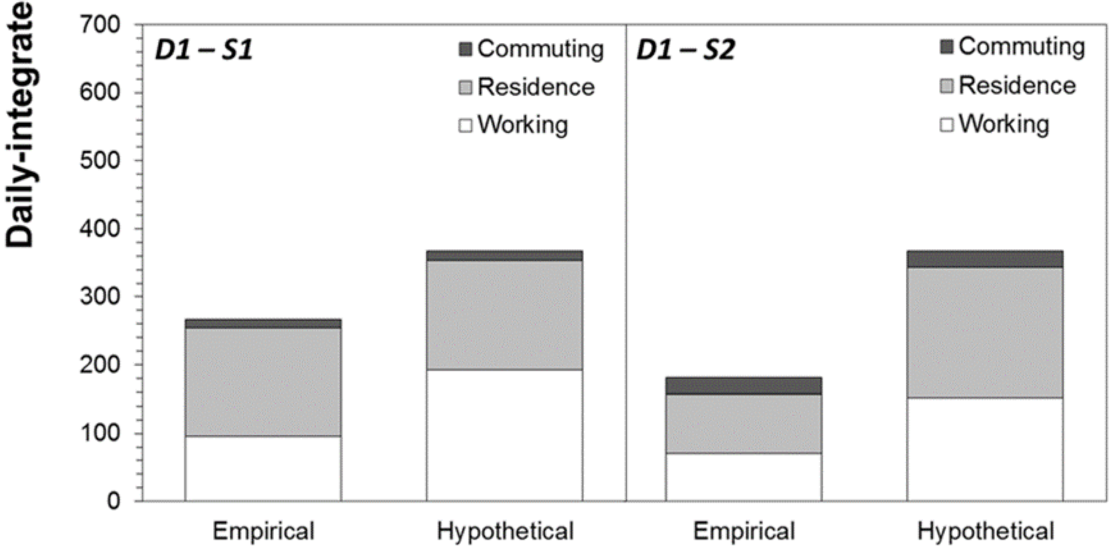
3.3.1. Hypothetical exposures

Outdoor measurements reveal that the hypothetical daily-integrated exposures to $PM_{0.3\text{-}2.5}$ during the moderate haze period (D1-D3) were almost twice those during light haze days (D4-D6). Specifically, during the moderate haze period, the average daily-integrated hypothetical exposures measured at stations A and B were 430 ± 67 μm^3-h/cm^3 and 543 ± 67 μm^3-h/cm^3, respectively; the corresponding values during light haze period were 206 ± 31 μm^3-h/cm^3 and 236 ± 31 μm^3-h/cm^3, respectively.

The particle size decomposition indicates that the size-fractionated exposure proportions did not change much among the six monitoring days. More than half (53 ± 7%) of the hypothetical exposure was contributed by particles of size in the range 0.3-0.5 µm, 22 ± 6% by particles in the size range 0.5-1.0 µm, and 25 ± 4% by particles in the size range 1.0-2.5 µm.



(a) Particle size decomposition



(b) Microenvironment decomposition

Figure 3. Empirical and hypothetical daily-integrated exposures for participants S1 and S2 on the first monitoring day (D1, 25 June 2013). The top two and bottom two frames decompose the exposures according to (a) particle size and (b) microenvironment, respectively.

3.3.2. Empirical exposures

Considering all six monitoring days for each of the five participants, we observed that the empirical daily-integrated exposure to $PM_{0.3\text{-}2.5}$ varied by about a factor of seven, from 66 to 454 μm^3-h/cm^3. Tables 2 and 3 present the average exposures for D1-D3 (week one, moderate haze) and D4-D6 (week two, light haze), respectively. Consistent with the trend observed in hypothetical exposures, for each participant, the average for the light haze days (D4-D6) was only about half of the value recorded for moderate haze days (D1-D3).

For each of the six monitoring days, the two highest empirical exposures occurred for participants S1 and S4. The microenvironmental decomposition reveals that it is the contribution from the residential period that elevated their daily-integrated exposures. As indicated in Table 1, participants S1 and S4 reside in naturally ventilated environments without air conditioning. The need to open windows to help regulate the air temperature and the lack of any active particle removal leads to indoor particle concentrations in these residences that are comparable to the corresponding outdoor levels. At the other limit, the lowest exposures always occurred for participant S5. The office where participant S5 works is outfitted with higher grade of filters, which leads to consistently lower particle concentrations to which S5 is exposed during the daytime.

Table 2. Average daily-integrated empirical exposures (in units of μm^3-h/cm^3) specific to different microenvironments (µE) and different particle sizes for five subjects during week one (D1-D3, 25-27 Jun 2013). [a]

µE	Particle size	S1	S2	S3	S4	S5
W	0.3-0.5 µm	64	48	105	73	24
	0.5-1.0 µm	37	24	47	34	9
	1.0-2.5 µm	14	8	18	9	4
	0.3-2.5 µm	**115**	**80**	**170**	**116**	**38**
R	0.3-0.5 µm	84	66	57	95	56
	0.5-1.0 µm	62	31	24	58	28
	1.0-2.5 µm	66	13	17	46	22
	0.3-2.5 µm	**212**	**110**	**98**	**199**	**106**
C	0.3-0.5 µm	6	13	7	8	7
	0.5-1.0 µm	4	10	4	4	5
	1.0-2.5 µm	5	10	2	2	4
	0.3-2.5 µm	**15**	**33**	**14**	**15**	**16**
Daily-integrated	0.3-0.5 µm	154	127	169	180	91
	0.5-1.0 µm	103	65	75	99	45
	1.0-2.5 µm	85	31	38	61	33
	0.3-2.5 µm	**342**	**223**	**282**	**340**	**169**

[a] The exposures in the "other" microenvironment (O) are not separately presented in this table due to their idiosyncratic characteristics and small contribution to totals. However, the "other" category is reflected in the "daily-integrated" results.

Table 3. Average daily-integrated empirical exposures (in units of μm^3-h/cm^3) specific to different microenvironments (µE) and different particle sizes for the five subjects during week two (D4-D6, 2-4 Jul 2013). [a]

µE	Particle size	S1	S2	S3	S4	S5
W	0.3-0.5 µm	32	32	47	25	11
	0.5-1.0 µm	13	12	17	7	3
	1.0-2.5 µm	7	7	11	5	2
	0.3-2.5 µm	**52**	**51**	**75**	**37**	**16.0**
R	0.3-0.5 µm	39	24	27	84	26
	0.5-1.0 µm	19	8	11	33	12
	1.0-2.5 µm	27	6	10	27	12
	0.3-2.5 µm	**86**	**38**	**48**	**144**	**50**
C	0.3-0.5 µm	5	5	5	4	3
	0.5-1.0 µm	2	3	2	2	2
	1.0-2.5 µm	3	4	2	1	2
	0.3-2.5 µm	**10**	**12**	**9**	**7**	**7**
Daily-integrated	0.3-0.5 µm	76	61	79	117	42
	0.5-1.0 µm	34	23	30	43	18
	1.0-2.5 µm	38	17	23	34	18
	0.3-2.5 µm	**148**	**101**	**132**	**194**	**78**

[a] The exposures in the "other" microenvironment (O) are not separately presented in this table due to their idiosyncratic characteristics and small contribution to totals. However the "other" category is reflected in the "daily-integrated" results.

It is noteworthy that the empirical exposures depend not only on the extent of protection that is provided by the building against outdoor particles but also on the time spent in each microenvironment. Compared to other microenvironments (such as during commute), participants were generally exposed to lower particle concentrations at work and at home. Nevertheless, their exposures in the work and home microenvironments amounted to more than 80% of the daily total because of the large proportion of time spent in these two settings. The results highlight that efforts to control daily exposure to particulate matter during haze events should appropriately focus on microenvironments in which subjects spend much of their time.

As indicated in the decomposition by particle size (Figures S14-S19), about half of the daily exposure is attributed to particles in the size range 0.3-0.5 µm, a result that is consistent with the proportion of haze particles found in this size range in outdoor air.

3.4 Exposure factors

Table 4 reports daily average exposure factors, specific to different microenvironments and different particle sizes, for each of the participants. The overall average value of the exposure factor to $PM_{0.3-2.5}$ ranged from 0.32 to 0.75 across the five participants. It is noteworthy that even in this relatively homogeneous sample of subjects, exposures could vary by a factor of 2.3 from highest to lowest, primarily because of differences in the levels of protection provided by different buildings that were occupied. The highest average exposure factors were observed for participants S1 and S4, whereas the lowest value was found for participant S5.

All of the workplace microenvironments investigated in this study were equipped with central air conditioning and mechanical ventilation systems, but only one (where S5 works) has air filters upgraded from the basic MERV 7 to the more efficient MERV 13. By comparing the exposure factors at work for S5 with the corresponding values for other subjects, we can see the strong effect that improved filter quality has on limiting exposures to haze particles. The overall average exposure factor for S5 at work across all particle sizes was 0.13, much lower than the values of 0.42-0.59 that were measured for the other four subjects.

Table 4. Averages (Avg) and standard deviations (SD) of the daily-averaged exposure factors specific to different microenvironments (µE) and different particle sizes for each of the five subjects. [a]

µE	Particle size	**S1**		**S2**		**S3**		**S4**		**S5**	
		Avg	*SD*	**Avg**	*SD*	**Avg**	*SD*	**Avg**	*SD*	**Avg**	*SD*
W	0.3-0.5 µm	**0.63**	*0.10*	**0.64**	*0.09*	**0.71**	*0.05*	**0.54**	*0.16*	**0.17**	*0.01*
	0.5-1.0 µm	**0.69**	*0.12*	**0.63**	*0.15*	**0.60**	*0.05*	**0.43**	*0.11*	**0.12**	*0.01*
	1.0-2.5 µm	**0.27**	*0.07*	**0.24**	*0.09*	**0.31**	*0.04*	**0.17**	*0.06*	**0.07**	*0.01*
	0.3-2.5 µm	**0.54**	*0.07*	**0.52**	*0.07*	**0.59**	*0.04*	**0.42**	*0.12*	**0.13**	*0.01*
R	0.3-0.5 µm	**0.75**	*0.19*	**0.48**	*0.16*	**0.49**	*0.06*	**1.00**	*0.27*	**0.48**	*0.07*
	0.5-1.0 µm	**1.37**	*0.28*	**0.56**	*0.10*	**0.44**	*0.08*	**1.03**	*0.15*	**0.51**	*0.10*
	1.0-2.5 µm	**1.27**	*0.29*	**0.22**	*0.07*	**0.37**	*0.07*	**0.87**	*0.15*	**0.38**	*0.10*
	0.3-2.5 µm	**0.94**	*0.12*	**0.41**	*0.09*	**0.45**	*0.06*	**0.98**	*0.17*	**0.49**	*0.07*
C	0.3-0.5 µm	**0.75**	*0.08*	**0.91**	*0.10*	**0.85**	*0.08*	**0.53**	*0.08*	**0.75**	*0.26*
	0.5-1.0 µm	**1.20**	*0.24*	**1.38**	*0.22*	**0.95**	*0.16*	**0.49**	*0.06*	**0.97**	*0.40*
	1.0-2.5 µm	**1.09**	*0.18*	**1.38**	*0.25*	**0.72**	*0.15*	**0.36**	*0.13*	**0.97**	*0.21*
	0.3-2.5 µm	**0.91**	*0.08*	**1.11**	*0.09*	**0.84**	*0.11*	**0.48**	*0.05*	**0.86**	*0.27*
Daily-integrated	0.3-0.5 µm	**0.69**	*0.13*	**0.57**	*0.13*	**0.62**	*0.05*	**0.79**	*0.20*	**0.33**	*0.05*
	0.5-1.0 µm	**1.00**	*0.17*	**0.64**	*0.10*	**0.54**	*0.06*	**0.75**	*0.11*	**0.33**	*0.05*
	1.0-2.5 µm	**0.76**	*0.14*	**0.30**	*0.06*	**0.35**	*0.05*	**0.54**	*0.11*	**0.28**	*0.05*
	0.3-2.5 µm	**0.75**	*0.09*	**0.50**	*0.05*	**0.54**	*0.04*	**0.72**	*0.13*	**0.32**	*0.05*

[a] The exposures in the “other” microenvironment (O) are not separately presented in this table due to their idiosyncratic characteristics and small contribution to totals. However, the “other” category is reflected in the “daily-integrated” results.

The residential microenvironments were of three types, governed by differences among cooling and ventilation systems: (i) natural ventilation without cooling (S1 and S4), (ii) natural ventilation with split-system air-conditioning (S3 and S5), and (iii) mechanical ventilation with filtration and central air-conditioning (S2). The residential exposure factor for S1 (0.94) and S4 (0.98) are close to unity, which indicates that a purely naturally ventilated space in a tropical climate offers little protection from outdoor particulate matter for its occupants. The reduced residential exposure factors for S3 (0.45) and S5 (0.49) with split-system air conditioning are comparable to those for the full mechanical ventilation system with air conditioning (0.41 for

S2). The moderate degree of protection provided by split-system air conditioning cases might reflect passive particle deposition to indoor surfaces combined with very low air exchange rates; however, the data available in this study do not allow us to confirm that explanation.

While commuting, the exposure factor varies among transportation modes. The commuting exposure factor for the walking subject (S2, 1.11) is higher than unity, probably because of full exposure not only to the haze particles in outdoor air but also to some contribution from the local emissions from vehicles. The commute exposure factors for the subject who used a private car (S4, 0.48) is considerably lower than the values for the other three subjects (0.84-0.91) who combined time spent outdoors (walking and waiting for a bus) with time in the bus cabin.

4. Conclusions

We adopted a personal monitoring approach to evaluate the daily-integrated PM exposures and associated exposure factors for five participants over a period of six weekdays during the 2013 haze episode in Singapore. Although limited to a small sample of subjects and moderate duration, the scale of this monitoring campaign is among the largest of those that utilize real-time monitoring equipment, having produced 720 hours of real-time, size-resolved fine-particle data. A distinctive feature is detailed coupling of this size- and time-resolved monitoring data with activity diaries that allow us to attribute the contributions to exposure to the different microenvironments occupied by the study subjects. Although there is a rich literature on particle indoor/outdoor relationships and penetration factors [19, 20], only a few studies have isolated the indoor/outdoor conditions during times of occupancy, which may be systematically different than the overall average conditions [26, 27].

Across the six monitored days, the daily-integrated $PM_{0.3-2.5}$ exposure for each participant spanned nearly an order of magnitude, ranging from 66 to 454 μm^3-h/cm^3. This high variability of exposures was substantially influenced by the change in outdoor particle concentrations, but also by differences among study subjects in the characteristics of the microenvironments that they occupied. Exposures experienced at work and in the home contributed more than 80% to the total daily exposure because participants spent a large majority of their daily time in these two microenvironments. The data from disaggregating exposure according to particle size indicated that the largest contribution came from the smallest of the three size bins, corresponding to diameters in the range 0.3-0.5 µm, and contributing about half to the total measured exposure.

We quantified an exposure factor to relate the measured exposure to the hypothetical value that would occur if a subject were continuously exposed to particles at the measured outdoor concentration. For the six days of monitoring in this study, the average exposure factor ranged from 0.32 to 0.75 across the five participants. The microenvironment-resolved results indicate that higher exposure factors were associated with (i) the lack (or low efficiency) of particle removal filters; (ii) high penetration rate of outdoor air (without filtration); and (iii) (to a limited extent) the occurrence of indoor source activities. For working and residential environments that are equipped with air conditioning systems, we observed relatively higher exposure factors for submicron sized particles ($PM_{0.3-0.5}$, $PM_{0.5-1.0}$) than for larger fine-mode particles ($PM_{1.0-2.5}$).

Personal masks, such as the N95, can contribute to respiratory protection from haze. However, as was learned during Singapore’s 2013 haze episode, there are limits to the effectiveness of this approach. Children cannot be protected effectively from exposure by the use of masks that are sized for adults. Adults who have respiratory impairment may not be able to manage the heavier breathing workload associated with the mask’s resistance to airflow. And, even among healthy adults, the use of N95 masks in a tropical environment enhances thermal discomfort. Consequently, there is an important role for the development and application of strategies to mitigate haze exposures through improved building design and operation. The results of this study suggest that the most effective ways to reduce the exposures during haze periods lie in improving particle removal efficiency in high occupancy environments such as in offices, residences, and schools. Using air-conditioners, using higher-grade air filters, and employing additional stand-alone air cleaners during haze periods are among possible choices for individuals and building operators. Future efforts are needed to develop and evaluate effective interventions for buildings that are naturally ventilated.

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Supporting Information

Additional Supporting Information includes:

Unit conversion. Details about converting particle number concentrations into volumetric concentrations.

Calibration factors. Details about side-by-side tests for optical particle counters.

Figure S1 – S12. Time-series plots of exposure concentrations and outdoor particle concentrations, along with daily activity information for each of the six monitoring days and five subjects.

Figure S13. Regional haze map for each of the six monitoring days.

Figure S14 – S19. Average empirical and hypothetical exposures for each of the six monitoring days and their decomposition according to particle size.

Figure S20 – S25. Average empirical and hypothetical exposures for each of the six monitoring days and their decomposition according to microenvironment.

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