Exposure to particulate matter and ozone of outdoor origin in Singapore

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ABSTRACT

Estimates of population exposure to ambient air pollution traditionally rely on concentrations measured at central-site monitors as a surrogate for concentrations to which people are exposed. In this study of Singapore, we estimate population-averaged exposure concentrations for $PM_{2.5}$, PM_{10}, and O_3 by applying a model and data that account for age and gender demographics, intraurban regional variability, and microenvironmental effects with age- and gender-stratified time-activity budgets. The study addresses exposure only to air pollutants of outdoor origin. Spatially averaged midpoint estimates of lifetime ambient exposure concentrations are 59%, 52%, and 47% of outdoor concentrations for $PM_{2.5}$, PM_{10}, and O_3, respectively. Utilizing ambient data for calendar year 2007, we estimate that intraurban variability in ambient concentration results in lifetime-integrated exposure concentrations in the respective ranges of 10–14 $\mu g\ m^{-3}$ for $PM_{2.5}$, 14–18 $\mu g\ m^{-3}$ for PM_{10}, and 7.5–15 $\mu g\ m^{-3}$ for O_3. Uncertainty in estimates of the indoor proportion of outdoor pollutants, which are input to the model, results in greater variability than do intraurban differences in ambient concentrations, resulting in respective ranges of 6.6–15 $\mu g\ m^{-3}$ for $PM_{2.5}$, 8.1–21 $\mu g\ m^{-3}$ for PM_{10} and 6.8–16 $\mu g\ m^{-3}$ for O_3. Estimates of time spent in naturally ventilated (NV) homes are in the range 10–13 h/d across the population and exposures in NV homes contribute 49%, 53%, and 56% of total exposure for $PM_{2.5}$, PM_{10} and O_3, of outdoor origin, respectively. Results illustrate the importance of accurately characterizing climate-specific indoor–outdoor pollutant relationships to better quantify human exposure to air pollutants.

1. Introduction

Epidemiological studies show that increases in outdoor particulate matter (PM) or ozone, at concentration increments of ~10 $\mu g\ m^{-3}$, are associated with increases in adverse health effects. Among the outcomes are increased rates of acute upper respiratory infections, asthma incidence, chronic obstructive pulmonary disease (COPD), and overall daily mortality [1–5]. Air quality indicators in Singapore are typically similar to those of major cities in Europe and the US, although per-capita emissions can be high when compared to cities of similar size [6]. Exposure to air pollution at ambient concentrations in Singapore raises health concerns that are similar to those in European and US cities. Quah and Boon [7] have assigned economic value to the health risks associated with elevated levels of PM in Singapore and report a central estimate of the total cost of ambient PM_{10} to be 4.3% of Singapore's gross domestic product. Sultan [8] has estimated health risk improvements and overall economic benefit from reducing PM_{10} exposures with varying building ventilation and filtration conditions for children and adults in Singapore. Other studies of air pollution in Singapore have investigated the relationship between outdoor air pollutants and traffic congestion [9], asthma exacerbation [10], and episodic haze events [11–14]. Rapid urbanization, penetration of household air-conditioning [15–17], and the potential for time-activity budgets to be affected by a year-round warm and humid climate, motivate research to better quantify population exposures to ambient air pollution in Singapore.

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As is common worldwide, published studies that have described economic costs and health impacts of outdoor air pollution in Singapore do not take careful account of personal exposures, but instead rely on a network of ambient air pollution monitors as surrogate indicators. In aggregate, people spend most of their time indoors [18]. As a result, exposure to air pollutants, even those of outdoor origin, largely occurs in indoor environments. A growing body of research highlights the importance of the built environment's impact on human exposure to air pollution [19,20]. For many pollutants, buildings attenuate indoor concentrations relative to those in the nearby outdoor air. Therefore, even in the absence of indoor pollutant sources, building operation and design, and patterns of human activity must be considered to accurately assess human exposure to air pollution of outdoor origin.

Several studies illustrate the indoor attenuation of outdoor pollutants by characterizing infiltration factors or the indoor proportion of outdoor pollutants (IPOP) for PM or ozone. For example, Riley et al. [21] report values of IPOP for $PM_{2.5}$ that vary considerably, from 0.2 to 0.75 for a modeled office building with two levels of particle filtration efficiency within the normal range of practice. Meng et al. [22] report infiltration factors for $PM_{2.5}$ estimated with a mass-balance model applied to experimental data from 114 homes to be 0.40 ± 0.19 (mean $\pm$ std. dev.). Meng et al. [22] also note that the exposure distribution bandwidth increases when models of indoor–outdoor transport account for the impact of building construction, ventilation, and particle properties on particle infiltration factors. With regard to ozone, in the absence of indoor sources, concentrations are supressed inside buildings as a result of reactive losses on building envelopes and indoor homogeneous and heterogeneous chemistry [23,24]. Indoor/outdoor ratios of ozone vary considerably, even among similar types of buildings. For example, Weschler [24] reports indoor/outdoor ratios of ozone that range from 0.3 to 0.7 for an office in Southern California. For both PM and ozone in occupied indoor environments, the degrees of attenuation of ambient air pollutants by buildings proportionately affect exposure concentrations, and so — given the large variability in these values — should be considered in estimating human exposures to ambient air pollution.

For Singapore, and, more broadly, for the large urban populations in tropical climates worldwide, there is a need to understand the implications of building operation and human time-activity budgets that are specific to tropical conditions, including climates and tropically-acclimatized populations. The investigation described here contributes in three ways to improving exposure assessments in tropical climates: 1) leveraging Singapore's extensive air-monitoring network of continuous air monitoring stations (CAMS) within or adjacent to centers of population [7]; 2) compiling new time-activity budgets from governmental surveys of the Singapore population and related scientific literature; and 3) quantifying the impact of indoor environments on human exposures through field monitoring as well as through a literature review.

2. Methods

A model was constructed to assess aggregate daily human exposure in Singapore to specific air pollutants: $PM_{2.5}$, PM_{10}, and ozone. In this evaluation, we are only considering the exposures that are attributable to these pollutants originating in outdoor air. For PM, it is well known that there are important indoor sources that also contribute to exposure. For ozone, in some special circumstances, there can be significant indoor sources. However, exposures resulting from indoor sources are not included in the analysis reported in this paper. The model was constructed to incorporate best available information, which includes regional variability in outdoor air quality, pollution attenuation in occupied microenvironments accounting for building type and mode of operation, and daily activities of age- and gender-stratified subpopulations.

The calculations utilized air pollution data from the year 2007. Where available, demographic and subpopulation time-budget data from 2007 were taken for six urban planning areas in Singapore, which comprise a total study population of 1,011,000 as of June 2007 [25]. Exposures are computed as the sum over all relevant microenvironments of the estimated microenvironmental pollutant concentration multiplied by the time spent in that microenvironment, as described by Burke et al. [26] and shown in equation (1):

$$Ex_k = \sum_{j=1}^{l} C_{k,j} t_j \tag{1}$$

where Ex_k is the average daily exposure for a member of a subpopulation to air pollutant k ($\mu g\ m^{-3}$ h), l is the number of microenvironments in which exposure to pollutant k occurs (−), $C_{k,j}$ is the time-averaged concentration of air pollutant k during the time spent in microenvironment j ($\mu g\ m^{-3}$) and t_j is the subpopulation-average time per day spent in microenvironment j (h).

Estimating population exposures to outdoor air pollution with equation (1) requires input data for concentrations of outdoor air pollutants, an estimate of the time spent in each microenvironment, and information on the indoor air pollutant level attributable to outdoor air pollutants in each microenvironment considered. In this investigation, we use a record of the time-series of hourly-averaged outdoor air pollutant concentrations from year 2007 as measured at six monitoring stations. Time-activity budgets are determined from governmental surveys of the population of Singapore supplemented where necessary with literature-based estimates. Values for the indoor proportion of outdoor pollutants (IPOP) for each pollutant and in each microenvironment are taken from a field study of homes in Singapore and from a review of relevant literature. The study population is further discussed in §2.1 and data sources are further described in §2.2–2.4.

2.1. Study population

The six regions included in this study were selected by balancing several criteria: availability of outdoor air quality data, proximity to a CAMS, preference for high population density, and distribution of locations across Singapore. Urban planning areas (UPAs) were chosen in the north, south, east, west, and central areas of the city. The geographic location of each monitoring station and UPA is shown in Fig. S1 of the Supporting Information. Data from each outdoor CAMS were assigned to one of the six Singapore zones based on the distance from the CAMS to the center of the respective UPA (see Table 1).

Input data were first structured for 5-year age increments across the population, resulting in estimates of exposure for an average individual belonging to a region, age, and gender subpopulation. Because the age-dependency of the estimated exposures was not strong, the results were then grouped and are reported here for four age clusters: 0–14 y, 15–24 y, 25–64 y, 65 + y. The overall output of the calculations is a yearly record of daily exposures to $PM_{2.5}$, PM_{10}, and O_3 for 8 subgroups: males and females in each of the four age subdivisions. Daily averaged exposures, with units of concentration $\times$ time, are then divided by the appropriate time increment to yield average exposure concentrations. For example, a daily exposure of 250 $\mu g\ m^{-3}$ h would be divided by the 24 h duration of the day and reported as an equivalent daily exposure

Table 1
Summary of selected monitoring stations and associated urban planning areas.

Monitoring station ID	Urban planning area, UPA	Distance, station to UPA center	UPA population, 2007 (1000s)	Data availability		
				O_3	$PM_{2.5}$	PM_{10}
P20	Ang Mo Kio	3.4 km	172.8	✓	✓	✓
P28	Bedok	2.4 km	284.1	✓	✓	✓
P31	Clementi	2.5 km	87.6	✓	✓	✓
P03	Novena	2.2 km	45.3	✓	✓	✓
P09	Jurong West	2.6 km	243.6	✓		✓
P24	Yishun	1.3 km	178.1	✓	✓	✓

concentration of 10.2 $\mu g\ m^{-3}$. When compared among the urban planning areas, exposure concentrations are reported as lifetime-integrated exposure concentrations to compress model output data and to facilitate clear comparisons across regions. Exposure concentrations were weighted for the years spent in each age subdivision for a hypothetical lifetime spent in each of the six study regions assuming average Singaporean life expectancies from available data (1980–2013). Life expectancies for males and females in the 0–14 y, 15–24 y, 25–64 y, and 65 + y age groups were taken as 78.1 y and 82.9 y, 74 y and 78 y, 70 y and 75 y, and 82 y and 86 y, respectively [27].

2.2. Ambient air pollutant concentrations

Hourly-average concentrations of $PM_{2.5}$, PM_{10}, and O_3 were obtained for CAMS in Singapore for the full year 2007. The hourly concentrations of $PM_{2.5}$, PM_{10}, and O_3 were processed to compute daily-average values and input into equation (1) as a constant value for each day. The daily average values of air pollutants input to the model are summarized and shown in Figs. 1 and 2 and in Fig. S2 of the Supporting Information. The daily average values of pollutant concentrations were used because surveys of time-activity budgets for Singapore only reflect the average amount of time per day spent in each microenvironment, with no information regarding the daily schedule.

As summarized by Velasco and Roth [6], the climate of Singapore, combined with environmental regulations, emission controls, and convective weather yield concentrations of criteria air pollutants that are generally within local and international standards. Daily average pollutant concentrations input to the model for the six regions considered in this study are summarized in the box and whisker plots shown in Fig. 1 and generally show that the year 2007 outdoor air pollutant concentrations were either below or only slightly above World Health Organization interim targets.

Regional variability in outdoor air pollution is small for PM and moderate for ozone across Singapore. Two of six monitoring stations (Bedok and Yishun) appear to have lower magnitude and smaller variability for both daily $PM_{2.5}$ and PM_{10} concentration, with 95th percentile values lower by ~5 $\mu g\ m^{-3}$ than in other reported regions. Ozone concentrations exhibit stronger regional variability than PM. Annual average concentrations vary by a factor of two across the six stations considered in this study.

Temporal variability, over diurnal and yearly time-scales, can influence exposure to ambient air pollution. The variability in daily average PM and O_3 concentrations is shown in Fig. 2. Daily average PM concentrations fluctuate but without strong trends across year 2007. Ozone concentrations are suppressed during the months of May–September, and exhibit high variability during the Feb–April and Oct–Dec periods.

Diurnal variability in ambient air pollution concentrations contributes to uncertainty in our model estimates of exposure. Because we lack data to track the population's time-activity through the course of the day, we assume that a single value of daily average concentration is applicable throughout. Diurnal variability for $PM_{2.5}$ and PM_{10} is small (see Fig. S2 in Supporting Information), so our approach does not introduce substantial uncertainty for these pollutants. However, O_3 exhibits the expected trend of elevated concentrations during afternoon hours (12:00–19:00 local time; Fig. S2). This feature may contribute to an underestimation of O_3 exposures for some, particularly for
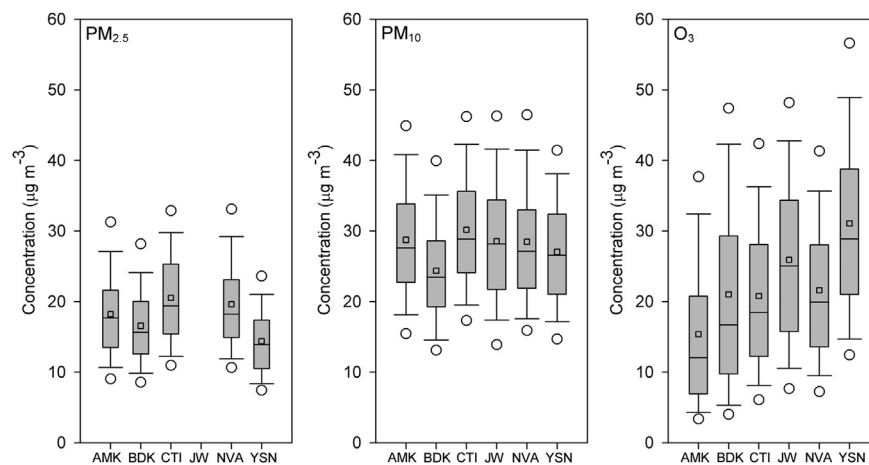

Fig. 1. Regional variability in daily-averages of outdoor air pollution concentrations for six urban planning areas in Singapore for the year 2007. Open circles are the 5th and 95th percentiles; whiskers correspond to the 10th and 90th percentiles; upper and lower bounds of the box represent 25th and 75th percentiles; solid line within box is the median; open squares are the annual averages. Acronyms refer to urban planning area: AMK is Ang Mo Kio, BDK is Bedok, CTI is Clementi, JW is Jurong West, NVA is Novena, and YSN is Yishun.
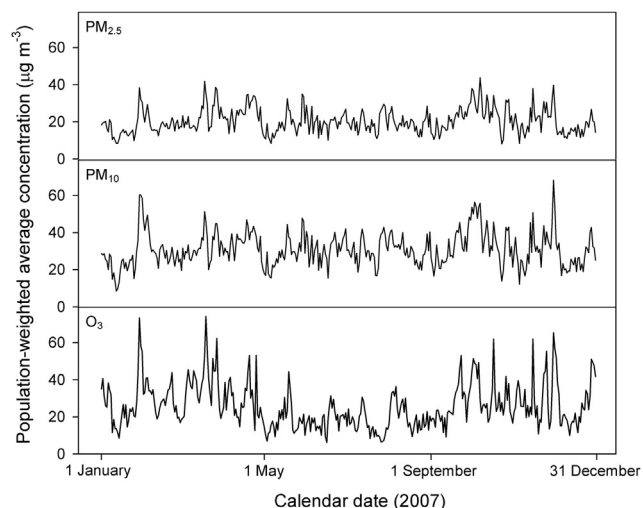
Fig. 2. Temporal variability in outdoor air pollution concentrations. Values shown are population-weighted daily averages across the six urban planning areas in Singapore considered in this study.

subpopulations that spend time during afternoon hours in less protective microenvironments when O_3 tends to be elevated (e.g., construction workers, elderly in naturally ventilated homes).

2.3. Description of microenvironments

Estimates of time-activity budgets were made for five microenvironment categories: work, commute, school, home, and outdoors. These five microenvironment categories were chosen based on two primary criteria: (a) data availability; and (b) microenvironments that individually and cumulatively contribute substantially to total daily activity. The five broad microenvironment categories were further classified into 11 specific microenvironments.

Time spent at work was assigned into one of four specific microenvironments: office, construction, manufacturing, or transportation based on a report of sector of employment [25]. Office microenvironments were assumed to have air-conditioning and ventilation systems that included some particle filtration. Construction work microenvironments were assumed to be open to the ambient atmosphere. With little specific data, manufacturing microenvironments were assumed to be enclosed and operate similar to office microenvironments. Transportation microenvironments were assumed to be enclosed vehicles.

Commute times were apportioned to two subcategories — 'open' or 'enclosed' — based on a survey of mode of transport to work or school [28]. Commuting modes of motorcycle, scooter, and walking were taken as 'open' to the ambient atmosphere, while commuting modes that included public bus, rail, taxi, private vehicle, and private chartered bus were taken as 'enclosed', i.e., not open to the ambient atmosphere.

Classrooms in Singapore traditionally rely on fan-assisted natural ventilation [29]. The World Bank reports that 6% of secondary enrollments are in private schools in Singapore [30]. Private schools were assumed to be air-conditioned and public schools were assumed to be served by fan-assisted natural ventilation.

Occupancy of homes was assumed to coincide with one of two ventilation/cooling modes: air-conditioned (AC) or naturally ventilated (NV). The time spent in homes was disaggregated according to a region-specific classification of dwelling type (e.g., 1- and 2-bedroom apartment, 3-bedroom apartment, detached house,

etc.), which was then multiplied by a distribution of the fraction of each dwelling type owning air-conditioning [28,31]. An average daily air-conditioning run-time of 8 h across all dwelling types, based on a survey of 46 Singaporeans [32], was assumed to apply for all dwellings in which occupants owned air-conditioners. Home AC runtime is explored in depth in §3.4.

2.4. Time-activity budgets

The average time spent in the 11 specific microenvironments across age- and gender-stratified subpopulations, shown in Table 2, was determined from publically available surveys of the Singapore population, augmented as necessary with data taken from the literature. A more detailed description and presentation of data sources (Figs. S3—S7 and Table S1) in estimating these time-activity budgets is provided in the Supporting Information. The working population in each age group for males and females was determined from a distribution of the fraction of employed residents aged 15 years and older across the Singapore resident population [25]. Age- and gender-subdivided population estimates were then multiplied by midpoint values of a distribution of the weekly working hours of employed residents ranging from "below 30 h" to "more than 65 h" in 5-h increments [28].

The contribution of commuting to daily time budgets was quantified from available data regarding typical commute durations of employed and student residents [28]. Daily commuting times of resident working persons aged 15 years and older are available classified in increments of 15 min, from a minimum one-way commute time of 7.5 min to a maximum of 67.5 min. A similar distribution describes the travel times to school of resident students aged 5 years and over [28]. The distributions for commute times are not disaggregated by gender, and were assumed to apply similarly to both male and female resident employees and students.

Time spent in school was determined from a distribution that defines the percentage of male and female students in 5-year subgroups from ages 5—24 [25]. Singapore guidelines recommend 5 and 6 h per day for primary and secondary schools, respectively [33]. An average value of 5.5 h/d was used for the time spent in school for the school-aged population.

Time spent outdoors for Singaporean children was taken from Rose et al. [34], who report that children between 6 and 7 years of age spent, on average, 3.05 h per week outdoors. This value was used for Singaporean residents aged 0—19. Estimates of time spent outdoors for other age subgroups were derived from Chau et al. [35] to be 4.95 h per week for age groups 20—65 and 10 h per week for the age subgroup 65 and older.

The amount of time spent in homes was estimated for each age and gender subgroup by summing the time at work, in commute, at school, and outdoors and subtracting from 24 h per day. This estimate of remaining time for each subgroup was multiplied by a scaling factor that was held constant across all subgroups. The value of the scaling factor was determined with a nonlinear solver constrained such that the time budget for the 'home' microenvironment across the population equaled 69% as reported for time spent in US residences by Klepeis et al. [18]. This procedure created a distribution across age and gender subgroups for time spent in the 'home' microenvironment that is inversely proportional to the amount of time spent in the sum of 'work,' 'commute,' 'school,' and 'outdoor' microenvironments. This approach was chosen with the assumption that individuals employed or in school are likely to spend relatively less time at home than those unemployed and not in school.

Averaged across all subpopulations, the 11 specific microenvironments were found to comprise ~21 h of daily time-activity. The unaccounted 3 h of daily time-activity was assigned to the 11

Table 2
Summary of time-activity budgets across age and gender subgroups for Case 1, where unaccounted time is apportioned to microenvironments other than 'home'.[a,b]

Age	Gender	Work (h d^{-1})				Commute (h d^{-1})		School (h d^{-1})		Home (h d^{-1})		Outdoors (h d^{-1})
		Office	Const.	Manuf.	Trans.	Encl.	Open	AC/MV	NV	AC	NV	
0–14	M	0.0	0.0	0.0	0.0	0.32	0.27	0.30	4.7	5.9	11.8	0.81
	F	0.0	0.0	0.0	0.0	0.33	0.26	0.30	4.7	5.9	11.8	0.81
15–24	M	1.6	0.03	0.15	0.08	0.66	0.12	0.22	3.4	5.6	11.1	1.1
	F	2.3	0.02	0.29	0.20	0.67	0.09	0.20	3.1	5.5	10.7	1.0
25–64	M	4.0	0.49	1.2	0.93	0.50	0.12	0.0	0.0	5.3	10.3	1.0
	F	4.2	0.12	0.85	0.28	0.49	0.06	0.0	0.0	5.7	11.2	1.2
65+	M	1.4	0.08	0.16	0.22	0.14	0.05	0.0	0.0	6.1	12.4	3.4
	F	0.62	0.01	0.06	0.01	0.06	0.01	0.0	0.0	6.3	12.7	4.2
LT avg.[c]	M	2.5	0.26	0.67	0.52	0.42	0.14	0.08	1.3	5.6	11.1	1.4
	F	2.4	0.06	0.45	0.16	0.38	0.09	0.08	1.2	5.8	11.6	1.8

[a] Abbreviations: Const. — construction, Manuf. — manufacturing, Trans. — transportation, Encl. — enclosed, AC — air-conditioning, MV — mechanical ventilation, NV — natural ventilation, and LT avg. — lifetime average.
[b] Total time per day may not sum to 24 h due to rounding.
[c] Lifetime average determined from the weighted average of the years spent in each age subdivision assuming a life expectancy typical of Singapore.

specific microenvironments in three different ways to create three case conditions of time-activity budget. In Case 1, unaccounted time was allocated to each microenvironment, except the home microenvironment, in proportion to the time previously quantified for that microenvironment. In Case 2, the unaccounted time was assigned in proportion to the time quantified for all microenvironments, including the home. In Case 3, all unaccounted time was assigned to the home microenvironment. For this study, Case 1 is considered the baseline time-activity budget input. We expect that Cases 2 and 3 overestimate the time spent in the home (See Tables S2 and S3 in Supporting Information for Cases 2 and 3). Nevertheless, Cases 2 and 3 are considered to assess the sensitivity of the model estimates of exposure to uncertainty in time-activity budgets.

Best estimates of time-activity budgets reported in Table 2 illustrate the importance of the home microenvironment for exposures to ambient air pollution, particularly given the predominance of natural ventilation in Singapore dwellings. For 25–64 year olds, the total average time spent in work microenvironments is 5.5 h/d for women and 6.6 h/d for men. These values might appear in conflict with previous exposure estimates made for Singapore that assume an 8-h work day [8]. However, as one important contributor to the difference, note that information in Table 2 represents annual averages including weekend and other days without work or school.

Time spent commuting is the only contributor to transit exposures in this assessment. We lack data to incorporate other time spent in transportation microenvironments. Data for commuting time is only available for the subsets of the population that are either in school or at work. It is certain that, averaged across the population, 1) working and student subpopulations spend additional time in transit in addition to commutes to work and school, and 2) non-working and non-student subpopulations also spend time in transit. However, the uncertainty introduced by this data limitation is likely to be small, given the small proportion of the full day that is spent in transit. For example, for Singaporeans in the age range 15–65, Table 2 reports time in transit in the range 33–47 min/d, corresponding to 2–3% of the full day. In the assessment of time-activity patterns in the United States, Klepeis et al. [18] reported that the average time spent in an enclosed transportation vehicle was 79 min/d, i.e. 5% of the total 24-h period.

2.5. Indoor proportion of outdoor pollutants

Estimates of microenvironment-specific exposure concentrations were made by multiplying the daily average outdoor concentration by an appropriate value of the indoor proportion of outdoor pollutants (IPOP) for each particular microenvironment. There is large variability in the level of indoor pollutants that is attributable to those in outdoor air [36]. We assess the impact of the IPOP on model estimates of exposure concentration by defining three IPOP values for each of the 11 specific microenvironments considered in this study and for each of the three pollutants: $PM_{2.5}$, PM_{10}, and O_3. The basis for these values combines information from a Singapore field study with information from the literature. Estimates for 'low', 'midpoint', and 'high' values of IPOP for each microenvironment are reported in Table 3 and briefly justified below. A detailed description of the rationale behind these estimates is provided in the Supporting Information.

Estimates of IPOP for PM in office environments were taken from Riley et al. [21], with the 'low' value of IPOP used here associated with offices with higher efficiency filtration, the 'high' value with lower efficiency filtration, and the 'midpoint' value as the arithmetic average of 'high' and 'low' values. Estimates of IPOP for O_3 in offices were taken from Spengler [37] for a 'low' value, Weschler et al. [38] for a 'high' value, and a 'midpoint' was taken as the arithmetic average of 'high' and 'low' values. The IPOP for construction workplaces was assumed to be unity. Values of IPOP for manufacturing work microenvironments were assumed to be similar to office microenvironments. Values of IPOP for $PM_{2.5}$ and PM_{10} for transportation work microenvironments were taken from Tsai et al. [39]: 'low' values correspond to those reported for rail transit; 'midpoint' values correspond to vehicle transit; and 'high' values correspond to bus transit. Values of IPOP for O_3 for transportation work microenvironments were taken from Hayes [40], with 'low', 'midpoint', and 'high' IPOP reported for three different vehicle speeds.

Open commuting microenvironments were assumed to have IPOP values of unity. Values of IPOP for enclosed commuting microenvironments were the same as for transportation working microenvironments. The 'midpoint' values of the IPOP for $PM_{2.5}$ and PM_{10} for air conditioned and mechanically ventilated (ACMV) school microenvironments were taken from the average values reported in Braniš et al. [41] for daytime weekend periods (no occupancy), while the 'high' and 'low' values are respectively increased and decreased by the standard deviation. Estimates of 'midpoint', 'low', and 'high' IPOP for O_3 for ACMV schools were taken from Blondeau et al. [42] as the average, minimum, and maximum value, respectively, for two schools with mechanical ventilation. The 'midpoint' IPOP for $PM_{2.5}$ and PM_{10} for NV school microenvironments was taken as the average value reported in the

Table 3
Summary of the estimates of indoor proportion of outdoor pollutants (IPOP) that were input to the model.[a,b]

	$PM_{2.5}$	PM_{10}	O_3
Work			
Office	0.46 (0.20–0.72)	0.35 (0.15–0.55)	0.44 (0.084–0.80)
Construction	1	1	1
Manufacturing	0.46 (0.20–0.72)	0.35 (0.15–0.55)	0.46 (0.084–0.84)
Transportation	0.82 (0.77–0.95)	0.74 (0.62–0.96)	0.33 (0.21–0.41)
Commute			
Open	1	1	1
Enclosed	0.82 (0.77–0.95)	0.74 (0.62–0.96)	0.33 (0.21–0.41)
School			
AC/MV	0.81 (0.64–0.98)	0.50 (0.31–0.69)	0.23 (0.15–0.28)
NV	0.88 (0.85–0.94)	0.85 (0.82–0.87)	0.28 (0.14–0.48)
Home			
AC	0.60 (0.37–0.91)	0.31 (0.12–0.55)	0.24 (0.20–0.31)
NV	0.74 (0.39–0.89)	0.67 (0.33–0.88)	0.57 (0.27–0.93)
Outdoor	1	1	1

[a] Midpoint values determined from the literature are reported outside parentheses, a range of maximum and minimum IPOP are reported inside parentheses.
[b] Abbreviations: AC — air conditioned, NV — naturally ventilated, MV — mechanically ventilated.

work of Chen and Chang [43] for unoccupied periods across six schools in Singapore, while the 'high' and 'low' values are the upper and lower limits of the range of reported values. These values are in general agreement with ratios of indoor and outdoor PM made during unoccupied periods in naturally ventilated classrooms in Thailand [44]. Lacking data specific to Singapore, estimates of 'midpoint', 'low', and 'high' IPOP for O_3 for NV schools were taken from Mi et al. [45] as the average, minimum, and maximum values, respectively, across five NV schools in central Shanghai. The IPOP in outdoor environments were assumed to be unity for all three pollutants.

Estimates of IPOP for home microenvironments were obtained experimentally from a field campaign that made measurements in five apartments in Singapore under two cooling modes: air conditioned (AC) and fan-assisted natural ventilation (NV). Indoor/outdoor pollutant concentrations for the residences recruited for the field study are shown in Fig. S8, a description of building characteristics in Table S4, and estimates of IPOP across field sites in Fig. S9 of the Supporting Information. Estimates of IPOP in home microenvironments reported here reflect the average and upper and lower ends of the range of IPOP values across the five residences in this field study.

The values of IPOP in Table 3 show that different microenvironments can afford a wide range in the degree of protection provided against exposure to outdoor pollutants. The ranges of IPOP values generally show that indoor and enclosed environments are more protective against PM_{10} than against $PM_{2.5}$, which is an expected result corresponding to the higher deposition loss-rate coefficients for the larger particles that constitute part of PM_{10} [46]. Midpoint estimates of IPOP for ozone are in the range of 0.23–0.57, in line with measurements summarized by Weschler [24]. Values of IPOP for O_3 are lower than those for PM, indicating that indoor spaces generally offer greater protection from O_3 than from PM in ambient air. Available information also indicates high degrees of variability in IPOP values for specific microenvironments. For example, the IPOP for naturally ventilated homes ranged from 0.27 to 0.93 for ozone and from 0.39 to 0.89 for $PM_{2.5}$. As we shall see, the corresponding large ratios between the highest to lowest values in these ranges — 3.4 for ozone and 2.3 for $PM_{2.5}$ — combined with the relatively high proportion of time spent in the residential microenvironment

contributes substantially to total uncertainty in the overall exposure estimates.

3. Results and discussion

Enclosed environments reduce exposure concentrations of ambient air pollutants. Midpoint estimates of lifetime-integrated exposure concentrations, averaged across the six regions considered in this study, are 59%, 52%, and 47% of the corresponding outdoor concentrations for $PM_{2.5}$, PM_{10}, and O_3, respectively. Based on year 2007 ambient air pollution data, regional-averaged lifetime-integrated ambient exposure concentrations would be 11.8 µg m^{-3}, 15.8 µg m^{-3}, and 11.5 µg m^{-3} for $PM_{2.5}$, PM_{10}, and O_3, respectively.

The estimates of exposure concentration attributable to ambient $PM_{2.5}$ made here are approximately 50% higher than those of Burke et al. [26] for a simulated population in Philadelphia, PA, USA. The difference is largely a consequence of the higher ambient $PM_{2.5}$ in this investigation. The population-weighted annual average outdoor $PM_{2.5}$ concentration in our investigation of Singapore was 20.5 µg m^{-3}. For the study of Philadelphia, mean outdoor $PM_{2.5}$ ranged from 10.4 µg m^{-3} to 19.3 µg m^{-3}, when averaged over nighttime winter and daytime summer periods, respectively.

Given the importance of the home microenvironment for exposure to ambient air pollution, substantial time spent in naturally ventilated homes contributes to a lesser cumulative protective effect of building envelopes when comparing exposures in tropical and subtropical climate zones. For example, in a study of asthma severity and personal ozone exposure in San Diego, CA, Delfino et al. [47] reported that the 12-h personal ozone concentrations for 12 subjects were 27% of mean outdoor O_3 concentrations. Higher exposure concentrations presented here are partially a result of substantial time spent by populations in naturally ventilated environments in which the degree of attenuation of outdoor ozone can be small.

3.1. Pollutant exposure in relation to age and gender

Daily average exposure concentrations to ambient $PM_{2.5}$, PM_{10}, and O_3 of the age- and gender-stratified subpopulations across all six regions considered in this investigation are reported in Table 4. Midpoint estimates of ambient exposure concentrations across age subgroups show small variability, with ranges of 11.5–12.6 µg m^{-3}, 14.8–16.6 µg m^{-3}, and 10.5–11.3 µg m^{-3} for $PM_{2.5}$, PM_{10}, and O_3, respectively. There is no meaningful difference in total exposure to ambient PM and O_3 between genders within an age subgroup, which is to be expected given the small differences in estimates of population averaged time-activity budgets between genders (Table 2). There is a total variation of ~10% in population weighted-average exposure concentration as a function of age subgroup, as illustrated in Table 4. Trends in exposure concentration with increasing age differ for PM and O_3. Exposure concentrations for PM are similar across 0–14, 15–24, and 65+ age groups, and are slightly suppressed for the 25–64 age group. This dip is a result of the low value of IPOP for $PM_{2.5}$ and PM_{10} in 'office' and 'manufacturing' work microenvironments. The midpoint estimated IPOP of 0.46 and 0.35 for $PM_{2.5}$ and PM_{10}, respectively, indicate that 'office' and 'manufacturing' microenvironments are relatively more protective than other settings for ambient PM. In the case of O_3, the exposure concentrations increase with increasing age subgroup. This result is a consequence of relatively low midpoint values of IPOP for O_3 in enclosed microenvironments: time spent outdoors influences the overall differences across age subgroups. Table 2 indicates that time spent outdoors is lowest for the 0–14 age subgroup and highest for the 65+ age subgroup.

Table 4
Population-weighted average exposure concentrations across age and gender subgroups and lifetime-integrated average exposure concentrations.[a,b]

Age	Gender	$PM_{2.5}$	PM_{10}	O_3
		Population-weighted average exposure concentration ($\mu g\ m^{-3}$)		
0—14	M	12.3 (8.0—15.3)	16.6 (10.0—21.6)	10.5 (5.9—16.3)
	F	12.3 (8.0—15.3)	16.3 (10.0—21.6)	10.5 (5.9—16.3)
15—24	M	12.3 (7.9—15.4)	16.3 (9.7—21.3)	10.5 (5.8—16.1)
	F	12.2 (7.8—15.3)	16.1 (9.6—21.2)	10.5 (5.7—16.2)
25—64	M	11.5 (6.8—15.0)	14.9 (8.1—20.5)	11.1 (5.8—17.1)
	F	11.5 (6.6—15.0)	14.8 (7.7—20.4)	11.0 (5.7—17.1)
65+	M	12.4 (7.6—15.6)	15.9 (9.0—21.3)	11.1 (6.8—16.4)
	F	12.6 (7.8—15.7)	16.2 (9.2—21.5)	11.3 (7.0—16.5)
Region		Lifetime-integrated average exposure concentration ($\mu g\ m^{-3}$)		
Ang Mo Kio		12.8 (8.1—16.1)	16.7 (10.0—22.1)	7.5 (4.3—11.4)
Bedok		11.6 (7.4—14.6)	14.2 (8.4—18.8)	10.3 (5.9—15.6)
Clementi		14.4 (9.1—18.1)	17.6 (10.4—23.2)	10.2 (5.8—15.4)
Jurong West		nd	16.6 (9.9—22.0)	12.7 (7.2—19.2)
Novena		13.8 (8.7—17.4)	16.6 (9.9—21.9)	10.6 (6.0—16.0)
Yishun		10.1 (6.4—12.7)	15.7 (9.4—20.8)	15.2 (8.7—23.0)

[a] Exposures determined from midpoint estimates of IPOP are reported outside parentheses, reported ranges are values of IPOP determined for minimum - maximum conditions.

[b] All exposures estimates are made for the Case 1 time-activity budget.

3.2. Variability in exposure concentrations for pollutants of ambient origin

Lifetime-integrated exposure concentrations are reported in Table 4 for the six regions (five for $PM_{2.5}$) included in this study. As described in §3.1, age and gender subgroups exhibit relatively minor variations in input parameters. Therefore, differences in population-average lifetime-integrated exposures across regions predominantly reflect intraurban variability in outdoor air quality. For $PM_{2.5}$, lifetime-integrated exposure concentrations differ by 40%, from 10 $\mu g\ m^{-3}$ to 14 $\mu g\ m^{-3}$, from the lowest to highest 'midpoint' exposure concentration across regions. Regional variability is slightly smaller for PM_{10}, contributing to 30%, from 14 $\mu g\ m^{-3}$ to 18 $\mu g\ m^{-3}$, from the lowest to highest 'midpoint' exposure concentration across regions. Ozone exposures show the highest variability across regions: the 'midpoint' exposure concentration in Yishun (15 $\mu g\ m^{-3}$) is 2× that at Ang Mo Kio (7.5 $\mu g\ m^{-3}$).

The contribution of ranges of IPOP to the variability in exposure concentration is reflected in the ranges reported in Table 4. For $PM_{2.5}$, the largest variability within a region occurs for Clementi, where the exposure concentration ranges from 9.1 $\mu g\ m^{-3}$ for 'low' IPOP to 18.1 $\mu g\ m^{-3}$ for 'high' IPOP, a factor of two change. The impact of the range of IPOP for PM_{10} is larger than for $PM_{2.5}$. Again, the Clementi UPA has the largest difference in magnitude from the 'low' to 'high' estimates of IPOP, ranging from 10.4 $\mu g\ m^{-3}$ to 23 $\mu g\ m^{-3}$, i.e., different by a factor of 2.2. Ozone exposure exhibits the greatest variability as a result of values of IPOP input to the model; the 'low' to 'high' estimates of IPOP produce average exposure concentration estimates that range from 9.4 $\mu g\ m^{-3}$ to 23 $\mu g\ m^{-3}$ in the Yishun UPA, i.e., different by a factor of 2.7. These comparisons indicate that — in Singapore — the characteristics of the built environment are of similar or perhaps even greater importance than intraurban variability in outdoor air pollution in influencing air pollutant exposure concentrations.

The three cases with different time-activity budgets, as described in §2.4, yield only minor variability in lifetime-integrated exposure concentration (data not shown). As an example, for the midpoint estimate of IPOP for $PM_{2.5}$, the lifetime-integrated exposure concentration in Ang Mo Kio only changes by 1% when changing the time-activity budget from Case 1 to Case 2. Results are similar for PM_{10} and O_3. This evidence indicates that — given the

current state of knowledge — accurately characterizing the variability in IPOP for a population's building stock is more important for improving exposure estimates than improving information on time-activity budgets, at least when viewed on a population-average basis.

The exposure analysis undertaken in this investigation has considered three pollutants, three cases of time-activity budget, and three conditions of IPOP. For brevity, not all comparisons across model conditions are reported in the main paper. A full-factorial comparison of the various analyses is shown in Tables S5—S7. This evidence supports the finding that differences in lifetime exposure are driven by variability in IPOP values within microenvironment categories to a much greater extent than by differences in the allocations of uncharacterized time-activity. For example, in the case of exposure to $PM_{2.5}$, nine distinct comparisons for constant IPOP with varying time-activity conditions yield total lifetime exposures that always vary by less than 10%. Conversely, comparisons across model conditions show that the largest impact to estimates of lifetime exposure concentration occur when comparing 'high' to 'low' estimates of IPOP. Estimates made with 'high' IPOP are a factor of 2.0—2.9 higher than 'low' IPOP for any particular time-activity budget.

3.3. Relative exposures among microenvironments

The values of IPOP input to the model affect the magnitude of population exposure and simultaneously impact the relative contributions of each microenvironment in which exposure occurs. Relative exposures across the sample population were calculated for each microenvironment by dividing the contribution to exposure calculated with equation (1) in each of the 11 specific microenvironments by the total exposure. The impact of changing the IPOP on relative exposures to $PM_{2.5}$, PM_{10}, and O_3 is shown in Fig. 3. For brevity, only Case 1 estimates of the bounding and central IPOP values are shown here. A full comparison that includes the effect of time-activity conditions on relative exposure in microenvironments is presented in Figs. S10—S12 of the Supporting Information.

The dominant microenvironment contributing to Singapore's population-averaged exposure to ozone and PM of outdoor origin is naturally ventilated homes, with proportional contributions to total exposure in the respective ranges 40%—49%, 43%—54%, and 47%—61%, for $PM_{2.5}$, PM_{10}, and O_3. The contribution of the naturally
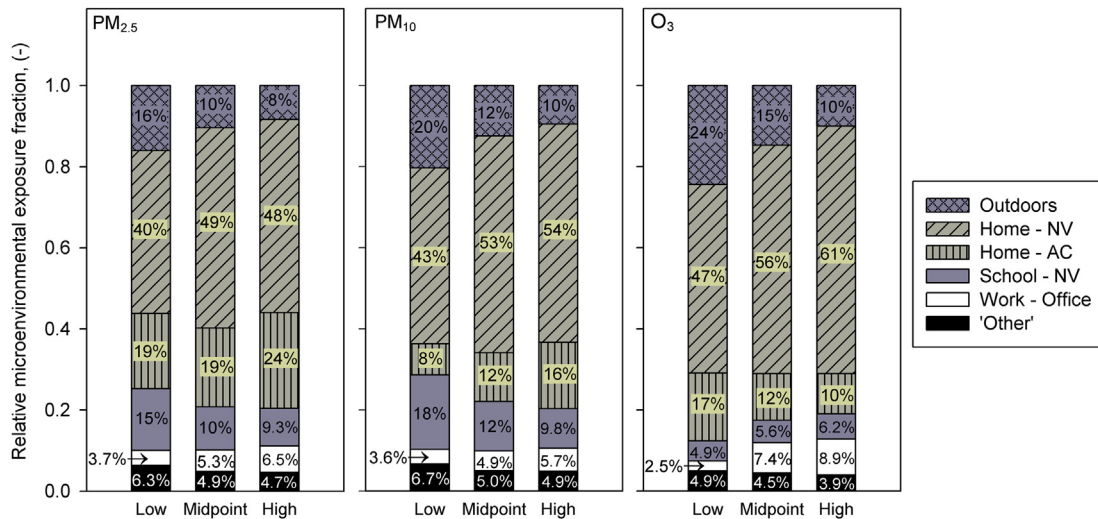
Fig. 3. Relative lifetime exposure contribution from specific microenvironments considered in this investigation for three conditions of indoor proportion of outdoor pollutants and one time-activity budget (Case 1). Microenvironments under the category of 'other' are the summed contribution of the microenvironments not explicitly shown in this figure but listed in Tables 2 and 3.

ventilated home microenvironment to cumulative population exposure corresponds approximately to the proportion of the time-budget spent in naturally ventilated homes. Time spent in NV homes is in the range 10.3–12.7 h/d across population subgroups (Table 2), or 43–53% of the respective subpopulation's total time-activity budget.

Cumulative population exposures to PM of outdoor origin in naturally ventilated schools are roughly comparable to exposures that occur outdoors, perhaps a surprising result given that only a subset of the population spends time in schools. The IPOP estimates for PM in NV schools are high, and a substantial amount of time is spent in schools by the 0–14 and 15–24 age subgroups. The IPOP for O_3 in naturally ventilated schools is suppressed compared to PM, and the relative contribution of NV schools to O_3 exposure is only 5–6%. Estimates of IPOP for O_3 in NV schools are from the findings of Blondeau et al. [42], in a study of schools in France. Future work should better characterize IPOP for O_3 in Singapore schools to strengthen the basis of exposure estimates.

Model runs employing 'low' values of the IPOP reflect more protective built and enclosed microenvironments and result in outdoor exposures constituting a greater fraction of total exposure than for 'midpoint' or 'high' IPOP values. This outcome is most evident in the case of O_3. Fig. 3 shows that under the 'low' IPOP condition, outdoor exposures to O_3 contribute 24% of population-wide exposure. This fraction decreases to 10% for the 'high' IPOP condition. Note that while the importance of outdoor exposures increases under the 'low' IPOP condition, as reported in Table 4, the magnitude of lifetime-integrated exposures is reduced with decreasing estimates of IPOP.

3.4. Effect of household ventilation condition

Homes are a predominant microenvironment contributing human exposure to ambient air pollution, as shown in Fig. 3 and discussed in §3.3. The indoor proportion of outdoor particulate matter and ozone is expected to vary in residential environments with, among other factors, the ventilation rate [48]. In turn, ventilation rates in Singapore residences are influenced by the prevalence and use patterns of air conditioning.

Air-conditioner ownership in homes has increased rapidly in Singapore. Based on year 2012 data, approximately 75% of households own at least one air-conditioner [49]. In Singapore, split air-conditioning systems are commonly used. These units have no dedicated outdoor air intake. Residences operating split air-conditioning systems typically rely on infiltration for air exchange. By contrast, naturally ventilated homes are typically characterized by large openings in the building façade (windows and doors) and often incorporate mechanical fans to promote air movement. Therefore, AC and NV homes have distinctive ventilation rates and interior airflow conditions that can influence the introduction of outdoor air pollutants into the indoor environment and their persistence therein.

While governmental surveys of household air-conditioner ownership exist for Singapore, data are limited regarding actual air-conditioner run-times. Considering the body of research which indicates that tropically acclimatized populations are comfortable at warmer temperatures and higher indoor air speeds than those in temperate climates [50,51], there is a need for more robust data regarding air-conditioning run-times, operating schedules, and associated population demographics. To estimate the impact of air-conditioner usage on estimates of lifetime- and regional-integrated population exposure, and to understand model sensitivity to this important characteristic of home microenvironments, we present in Fig. 4 the results of model runs where time spent in air-conditioned homes is varied from 10% to 90% of the overall, population-wide time spent in homes.

Fig. 4 illustrates that increasing air-conditioning run-times across the population has little effect on $PM_{2.5}$ exposure concentrations, a moderate impact on PM_{10} exposure concentrations, and a large impact on population exposure to O_3. These results incorporate results of the field study undertaken during this research to determine IPOP for AC and NV homes in Singapore. Split air-conditioners were observed to afford only modest protection from $PM_{2.5}$, as reflected in the similarity in estimates of IPOP for $PM_{2.5}$ in NV and AC homes shown in Table 3. As a result, across 'low,' 'midpoint,' and 'high' estimates of IPOP for $PM_{2.5}$, increasing AC run-time results in only small changes in population exposure to $PM_{2.5}$. Estimates of IPOP for PM_{10} were consistently lower in AC than in NV homes. As a result, population exposures to PM_{10} decrease by 45%, 46%, and 29% for 'low,' 'midpoint,' and 'high' estimates of IPOP, respectively, as AC run-time increases from 10% to 90% of time spent in the home. Estimates of IPOP for O_3 were highly
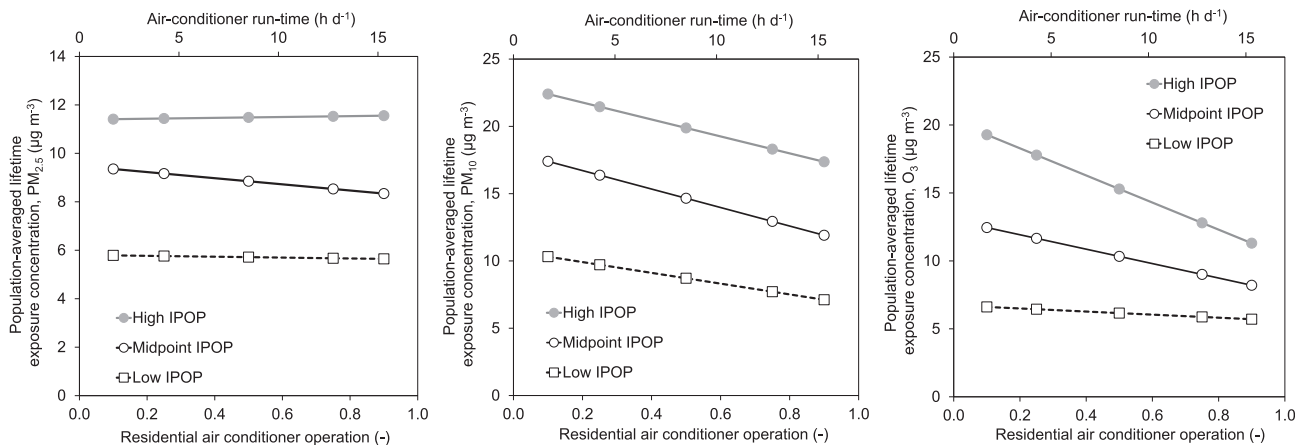
Fig. 4. Population weighted-average lifetime exposure concentrations as a function of air-conditioner run-time. Air-conditioner operation is the fraction of time spent in the home microenvironment. All model runs are calculated with the Case 1 time-activity budget.

variable for NV homes; Table 3 shows that IPOP values for O_3 span a broad range, 0.27—0.93. Nevertheless, IPOP for O_3 in AC homes are generally lower, ranging from 0.20 to 0.31. Consequently, population exposures to O_3 are reduced as home air-conditioning run-time increases. Lifetime- and region-integrated population exposures to O_3 decrease by 16%, 52%, and 71% for 'low', 'midpoint', and 'high' estimates of IPOP, respectively, as AC run-time increases from 10% to 90% of time spent at home.

Decreased air-exchange because of the use of AC rather than NV in homes contributes to a decoupling of residential indoor air from outdoor air. While such decoupling appears to yield some protective benefit for PM_{10} and O_3, using air-conditioning deliberately to provide protection from ambient air pollution should not be applied blindly. For example, risk factors for fine particles ($PM_{2.5}$) are greater than for coarse particles (PM_{10}) [52,53]. The absence of significant attenuation in $PM_{2.5}$ exposure concentration with increased time spent in air-conditioned home microenvironments (Fig. 4) implies that air-conditioning may not offer sufficient protection from fine ambient aerosol particles. In the case of O_3, lower air-exchange rates in air-conditioned homes produces lower indoor ozone concentrations because of the increased relative importance of ozone reactions on interior surfaces (and also, potentially, with gas-phase reactants) [24]. However, while reactive losses reduce indoor ozone concentrations, they also increase human exposure to oxidation products that may contribute to the associations observed between ambient ozone levels and human health impacts [54,55]. Furthermore, and perhaps most importantly, pollutants with indoor emission sources must be considered in homes with low air-exchange rates. As a regionally specific example, Wong and Huang [56] report considerable accumulation of carbon dioxide in air-conditioned Singaporean bedrooms, owing to human occupancy and low air-exchange rates, indicating a potential for the simultaneous accumulation of other air pollutants with indoor sources. Finally, increased household air-conditioning usage will increase building energy demand, an important consideration in Singapore, where the building stock accounts for approximately half of the country's total electricity use [57].

3.5. Study limitations

The model used in this investigation has several limitations. First, the calculations are based on population-averaged time-activity budgets and daily average pollutant concentrations, rather than actual or simulated diaries of individual's activities linked to time-varying ambient pollutant concentrations. Any influence on

exposure resulting from within-day correlations among time-activity, microenvironment characteristics, and outdoor air pollutant concentration is not captured in this approach. Outdoor air quality data from a specific central air monitoring station is assigned to a proximal region and is assumed to be representative throughout that region. Furthermore and potentially more important, the movement of Singaporeans within the city is not incorporated in the calculations. Future work should investigate more sophisticated approaches for incorporating the effects of population movement as well as spatial and temporal variability in ambient pollutant concentrations.

Time-activity budgets and estimates of IPOP were determined from available data that are specific to Singapore combined with most-appropriate estimates from literature when local data were not available. There are limited data regarding time-activity budgets of tropically acclimatized populations. Given the large and rapidly growing population in tropical regions, surveys of time-activity budgets of populations residing in Southeast Asia and other tropical climates are warranted. New approaches incorporating ubiquitous sensing in combination with models and/or measurements of air pollutants may improve exposure estimates through collection of real-time, individual data describing time-activity budgets as well as other relevant information such as building operation [58]. The approach employed here, which describes microenvironmental characteristics through estimates of IPOP, enables estimates to be made from a wide base of literature values; however, the impact of underlying parameters, for example the air-exchange rate, on population exposures is not explicitly incorporated. The use of IPOP estimates in the assessment does not support efforts to quantify the influence on exposure of indoor sources of PM or O_3. Estimates of IPOP specific to Singapore are reported here for residences and schools; measurements of IPOP across other building types and operation states described here would improve the applicability of the model.

4. Conclusions

The relationship between air pollutant concentrations measured at central monitoring stations and human exposure to air pollution of ambient origin depends on where people spend their time and on the proportion of outdoor pollution that penetrates and persists in occupied indoor microenvironments. Previous studies assessing population exposure to outdoor pollution have focused on temperate climates. Although the methods are suitable, the specific findings may be different in tropical climates, for

example because of differences in the way that buildings are thermally conditioned and ventilated.

The analysis reported here shows that intraurban variability in ambient concentrations as well as differences among built environments have important influence on human exposure to ambient air pollution. Lifetime-integrated exposure concentrations vary by up to a factor of two across regions in Singapore for the three pollutants considered in this investigation. Built environments attenuate to varying degrees indoor exposures to pollution of outdoor origin. Consequently, ambient air quality monitoring stations do not directly provide accurate measures of exposure concentrations. For Singapore, lifetime-integrated midpoint estimates averaged across six regions yield exposure concentrations that are 59%, 52%, and 47% of the corresponding outdoor concentrations for $PM_{2.5}$, PM_{10}, and O_3, respectively.

Human-building interactions (e.g., air-conditioning use, opening windows, etc.) in tropical climates should be better characterized. Modeling of human exposure to regionally homogeneous pollutants, like PM and O_3, may be improved by improved understanding of factors that affect residential concentrations [19]. Best estimates of building use reported here illustrate that despite the prevalence of air-conditioner ownership, considerable time is spent in naturally ventilated environments. This circumstance has important implications for exposure to ambient air pollutants, and may be especially important during episodic haze events when concentrations of outdoor air pollutants are elevated. Hypothetical calculations involving increased air-conditioner use in Singaporean residences show reductions in population-averaged PM_{10} and O_3 exposure concentrations. This finding should be considered in the context of accompanying increases in energy use; furthermore, lower air-exchange rates associated with split air-conditioner use would increase exposures to pollutants emitted from indoor sources. The predominance of exposure in the home microenvironment suggests that interventions targeting residential PM and O_3 might be effective components of an overall strategy to limit population exposures to ambient air pollution.

Acknowledgment

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.buildenv.2015.03.027.

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