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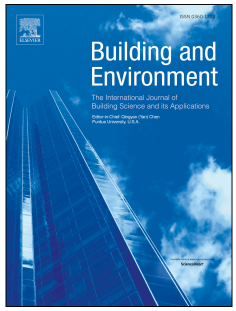

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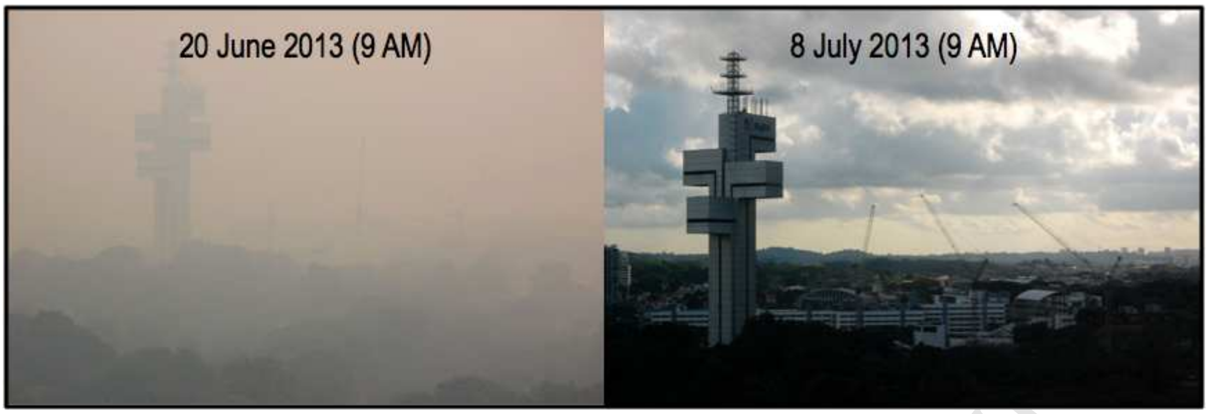
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Indoor and outdoor particles in an air-conditioned building during and after the 2013 haze in Singapore

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Abstract

Particles released from biomass burning can contribute to severe air pollution. We monitored indoor and outdoor particles in a mechanically ventilated and air-conditioned building during and after the 2013 haze event in Singapore. Continuous monitoring of time-and size-resolved particles in the diameter range 0.01-10 µm was conducted for two weeks in each sampling campaign. During the haze event, the averaged size-resolved outdoor particle volume concentrations ($dV/d(\log D_p)$) for diameters larger than 0.3 µm were considerably higher than those during the post-haze days (9-185 µm^3 cm^{-3} versus 1-35 µm^3 cm^{-3}). However, the average number concentration of particles with diameters in the range 10-200 nm was substantially lower on the hazy days than on the post-haze days (11,400 to 14,300 particles cm^{-3} for hazy days, versus an average of 23,700 particles cm^{-3} on post-haze days). The building mechanical ventilation system, equipped with MERV 7 filters, attenuated the penetration and persistence of outdoor particles into the monitored building. Indoor particle concentrations, in the diameter ranges 0.3-1.0 µm and 1.0-2.5 µm, closely tracked the corresponding patterns of outdoor particle concentrations. For particles in the size range 0.01-1.0 μm, the size-resolved mean indoor/outdoor (I/O) ratios were in the range 0.12-0.65 with the highest mean I/O ratio at 0.3 μm (0.59 in AC on mode and 0.64 in AC off mode). The air conditioning and mechanical ventilation system with MERV 7 filters provided low single-pass removal efficiency (less than ~ 30%) for particles with diameters of 0.01-1.0 μm. During the haze, for particles larger than ~ 0.2 μm, lower I/O ratios and higher removal efficiencies occurred with the air conditioning operating as compared to with mechanical ventilation only. This observation suggests the possibility of particle loss to air conditioning system surfaces, possibly enhanced by thermophoretic or diffusiophoretic effects.

Keywords: Indoor-outdoor relationship, Aerosol, Landscape fires, Pollutants, Particulate matter

1. Introduction

Two types of large, uncontrolled combustion can contribute to regional-scale air pollution episodes. Wildfires are common seasonal occurrences especially in semiarid regions such as the western United States and Australia. The use of large-scale biomass burning to clear land for agriculture is an important environmental issue in Southeast Asia. Such burning causes air quality problems because of the heavy emissions of combustion byproducts followed by atmospheric transport and dispersion plus photochemical transformation processes that create regional pollution episodes. Prior studies have investigated certain characteristics of airborne particulate matter associated with uncontrolled biomass burning, such as the organic and elemental carbon (OC/EC) composition of air [1, 2], biomass burning signatures of individual particles [3], trace elements in particulate matter [4, 5] and particle-bound polycyclic aromatic hydrocarbons (PAHs) [5, 6].

Particles originating from biomass burning might have significant impacts on human health. For example, such particles are demonstrated to be more toxic to lung macrophages than other ambient particles [7]. Particles from wildfires can induce pro-inflammatory responses [8] and contribute to oxidative stress [9]. A large wildfire in southern California was found to result in a “significant increase in hospital emergency room visits for asthma, respiratory problems, eye irritation, and smoke inhalation” [10]. Because of their potential contributions to the degradation of public health, it is worthwhile to pursue a deeper understanding of airborne particulate matter associated with uncontrolled biomass burning episodes.

Particle size is a key parameter, not only influencing dynamic behavior but also for assessing human health risks [11]. A few studies have documented that biomass burning activities can alter the airborne particle size distribution in the impacted area [12, 13]. Increases in particle mass concentrations are observed in the accumulation mode (0.1-2.0 μm). Particles in this size range contribute strongly to visibility impairment, a commonly observed adverse impact of large-scale biomass burning. Increases are also reported for the coarse mode (> 2.0 μm). However, decreases have been observed in the nucleation mode (diameter smaller than 0.1 μm). These findings highlight the importance of dynamic processes that influence the evolution of the particle size distribution. For example, growth induced by the condensation of semivolatile vapors would tend to shift nucleation mode particles toward the accumulation mode. It is important to better understand the size distributions of airborne particles associated with biomass burning events.

The penetration and persistence of particles from outdoor to indoor air is important with regard to health because people spend a large fraction of their time indoors [14]. When outdoor pollution levels are high, as during biomass burning episodes, people may be advised to curtail activities and remain indoors as a "shelter-in-place" strategy. For an office building, the major pathway connecting the indoor environment to outdoor air is the heating, ventilating and air-conditioning (HVAC) system [15]. For tropical climates such as in Singapore, heating is seldom or never needed, and so the analogous term, which we shall use in this paper, is the air-conditioning and mechanical ventilation (ACMV) system.

Several studies have reported that submicron particle number concentrations in office buildings closely follow the corresponding outdoor concentrations in the absence of a strong indoor source [16, 17, 18]. Among the factors that can affect the particle indoor/outdoor ratios (I/O) are particle size [19], air-exchange rate (AER) [19], and filter efficiency [16]. Indoor concentrations of particles originating outdoors can be reduced by improving filter efficiency [20]. Shi et al. [21] have reported laboratory tests that document the size-dependent particle removal efficiency of filters commonly used in ventilation systems. However, indoor-outdoor relationships have not been extensively reported for office buildings in relation to air pollution episodes caused by uncontrolled biomass burning. It is worthwhile to better understand the performance of normally used filters in office buildings for removing particles of outdoor origin, especially when the outdoor levels are episodically elevated, as during the 2013 haze in Singapore.

During the Southeast Asia haze episode of June 2013, the outdoor $PM_{2.5}$ concentrations rose to 250 μg m^{-3} on the most polluted days. This order-of-magnitude elevation above the normal ambient $PM_{2.5}$ concentration of 15-25 μg m^{-3} provided an opportunity to investigate the relationship between indoor and outdoor particle levels in a mechanically ventilated and air-conditioned building when the outdoor particle level was unusually high. The current study presents monitoring results and their interpretation considering size- and time-resolved indoor and outdoor particle concentrations both during the 2013 haze and on low-pollution days after the haze episode. The study aims to provide information and contribute new knowledge regarding four important features at the intersection of regional air pollution episodes, building environmental systems, and human exposure: 1) size-resolved outdoor particle volume and number concentrations measured in Singapore with and without episodic haze; 2) size-resolved indoor and outdoor particle relationships in a typical office building; 3) influence of ACMV operation modes on these relationships (i.e., with and without operating the air-conditioning cooling coil); and 4) performance of a typical ACMV system on reducing the penetration and persistence of outdoor particles indoors.

2. Material and Methods

2.1. Monitoring sites

Outdoor and indoor monitoring was undertaken on the campus of Nanyang Technological University (NTU). The NTU campus, located in western Singapore, is bordered by forested land to the north and west, by industrial areas to the south, and by residential areas to the east. On hazy days, the adjacent areas are not likely to have contributed substantially to the outdoor particle concentrations, as evidenced by the small variation in $PM_{2.5}$ concentrations across the five government-operated monitoring stations that span the city [22]. The sampling sites were on the western side of the campus, situated about 200 m from the forest. Vehicular traffic on the campus is small, consisting mainly of light-duty passenger cars for commuters. There are no other noteworthy particle sources on campus.

The present study reports results from two monitoring campaigns, with conditions that we will refer to as "hazy" and "clear sky," respectively. The hazy campaign spanned 14-29 June 2013 and the clear sky campaign took place 13-26 August 2013. Monitoring sites were the same for both campaigns. The outdoor monitoring station was sited on the balcony of a lecture theatre, with the air inlet positioned 12 m above the ground. The indoor station was 20 m away from the outdoor monitoring station and about 1.2 m above the floor. The fresh air intake of the ACMV system was situated at a height of 21 m above the ground and at 20 m horizontal distance from the outdoor monitoring station. Given the strong regional impact of the air pollution episode and the small contribution of local sources, we believe that the outdoor monitoring results reflect accurately the conditions prevailing in the ventilation air supplied to the indoor site. The room where the indoor station was placed had a hard-surface floor of area 19 m^2 and was part of a staff office. The office had an area of 300 m^2 and had been unoccupied for more than one year. Polyvinyl chloride flooring covered five-sixths of the office's floor surfaces and the remaining floor area was carpeted. The office also contained basic furniture such as tables, cabinets, and chairs. There were no obvious indoor particle sources. The room had casement windows and curtains; windows and doors were closed throughout both monitoring campaigns.

The air-handling unit (AHU) that served the office had an independent ventilation system (Figure 1), so the office was isolated from other rooms in the same building. When the mechanical ventilation was operating, make-up air accounted for ~ 10% of the volume flow rate of supply air. The make-up air mixed with the recirculated air first and then the air mixture passed through the filter and coil as shown in Figure 1. When the MV system was on, the office was slightly pressurized by the supplied air; such pressurization would have prevented outdoor air from substantially infiltrating into the office, making flow through the ACMV system the dominant pathway of fresh air supply and outdoor particle penetration. The filters in the AHU had a grade of MERV 7, which means its nominal removal efficiency is 25-35% for particles with diameters of 0.3-10.0 μm. In addition, its minimum removal efficiencies for particles with diameters of 0.3-1.0 μm, 1.0-3.0 μm and 3.0-10.0 μm are 17%, 46% and 50%, respectively [23].

When air conditioning was employed, chilled water circulating through the coil had a temperature of 7 °C. In normal practice at NTU, filters are replaced and cooling coils are cleaned concurrently at intervals of three months. From June to August 2013, the filters were not replaced and the cooling coils were not cleaned. When the mechanical ventilation was on, the air exchange rate of the office was 3.8 h^{-1}, whereas when the system was off, the average air exchange rate (owing to leakage) was 0.5 h^{-1}.

The ACMV system was operated in three different modes during the two monitoring campaigns. During weekdays of both the hazy and the clear-sky periods, the ACMV system was on (Mode 1: air conditioning and mechanical ventilation on) from 7:30 to 18:30. Overnight during the haze period, i.e. 18:30 to 7:30 on the next day, the AC was off but the MV system continued to operate (Mode 2: air conditioning off, mechanical ventilation on). During the weekday overnight intervals of the clear-sky period, the ACMV system was off (Mode 3: air conditioning and mechanical ventilation off). For weekends (both daytime and overnight), Mode 2 was applied during the haze period and Mode 3 was applied for the clear-sky days.

2.2. Instruments

During both campaigns, size- and time-resolved concentrations of both indoor and outdoor particles with diameters in the range 0.01 μm to 10 μm were concurrently monitored for multiple days. Particles with diameters of 0.01 μm to 0.2 μm were measured with TSI Nanoscan SMPS Nanoparticle Sizers (Model 3910, TSI Inc., Shoreview, USA). The SMPS uses isopropyl alcohol (purity ≥ 99.7%, Sigma-Aldrich) as the reagent and can measure particle number concentrations in the range 100-1,000,000 cm^{-3}. For larger particles, 0.3-10 µm in diameter, TSI optical particle sizers (OPS, Model 3330) were employed. These can measure particle concentrations up to 3,000 cm^{-3} and optically resolve particles into 16 size channels. Temperature and relative humidity were measured using TSI VelociCalc Air Velocity meters (model 9545-A). Monitoring was conducted continuously every day and measurement results were recorded at intervals of 1 min. However, with high water vapor content in Singapore's air, we found that the SMPSs only functioned properly (i.e. without reporting error) during some portions of each day. Consequently, we have relatively small datasets from the SMPSs in the current study.

An InfraRan Specific Vapor Analyzer (Wilkes Enterprise Inc., East Norwalk, USA) was used to measure the air exchange rates of the indoor environment based on the tracer gas decay method, using sulfur hexafluoride as the tracer.

2.3. Outdoor weather conditions

In accordance with expectations for Singapore's tropical climate, the outdoor weather conditions were similar during each sampling campaign. Table S1 presents a summary of selected outdoor atmospheric parameters and $PM_{2.5}$ mass concentrations for the two campaigns. The $PM_{2.5}$ mass concentration presented in Table S1 of each day was calculated based on outdoor sized-resolved particle number concentrations monitored by outdoor OPS and SMPS with assumed particle density of 1.0 g cm^{-3} [22]. During the hazy period, temperatures were between 25 and 35 °C and the relative humidity was 40-90%. The average daily wind speed was mainly in the range 5-8 km h^{-1} (except for 19-20 June) and from the southwest. Though it was during the monsoon season, there were only four precipitation events (June 16, 24, 25 and 26). During the clear-sky campaign, air temperatures were mainly between 27 and 32 °C and relative humidity was mainly between 50 and 90%. Mean wind speeds were mainly 4-8 km h^{-1} and were primarily from the south. There were six precipitation episodes during the clear-sky monitoring campaign. Given the similar weather conditions, the influence of meteorological conditions on outdoor particles is expected to be comparable for the two campaigns.

2.4. Data analysis and quality assurance

A clear difference is seen between the overall outdoor $PM_{2.5}$ concentrations of these two campaigns (Table S1). According to data reported by Singapore's National Environmental Agency (NEA), during the 2013 haze episode, the daily averaged outdoor $PM_{2.5}$ concentrations ranged from 38 to 268 μg m^{-3} and the average $PM_{2.5}$ concentration was 96 μg m^{-3}. Utilizing NEA data, and based on the daily-average outdoor $PM_{2.5}$ concentrations, we classified the hazy days into three categories: heavy haze ($PM_{2.5}$ > 150 μg m^{-3}), moderate haze (60-150 μg m^{-3}) and light haze (35-60 μg m^{-3}). During the clear-sky periods, the daily averaged outdoor $PM_{2.5}$ concentrations normally ranged from 10 to 30 μg m^{-3} and the average $PM_{2.5}$ concentration was approximately 20 μg m^{-3}.

Measured particle, temperature and RH results were first processed to exclude errors owing to instrument malfunction. Indoor and outdoor data were then paired as linked time series. For particles in the diameter range 0.01-10 μm, count concentrations were converted to volume concentrations based on the method reported in Zhou et al [22]. All data were arranged day-by-day and days that had complete data without evidence of error (i.e. owing to instrument malfunction) were chosen to compute outdoor size-resolved particle volume concentrations (dV/dlog D_p) and number concentrations (dN/dlog D_p). Size-resolved outdoor particle data for 19-22 June were averaged to represent the heavy haze days; data for 16-18 and 23 June were averaged to represent moderate haze conditions, data on 24-25 and 27-29 June were used to represent light-haze days, and measurements from 16-17 and 21-23 August were applied to represent the clear-sky conditions. In all, seventeen days were selected for further analysis, considering data availability as the major criterion. Days that had continuously valid data for less than three hours were excluded to limit errors in determining I/O ratios owing to lag time. In preliminary data processing, we only accepted data for which there was no error reported by either particle-monitoring instrument or otherwise recorded in our logbook.

Data records for the time period 10:00 to 18:00 on days with valid data were chosen for calculating I/O ratios for Mode 1. Data records from 20:00 to 6:00 of their next day were chosen for analysis for Mode 2 conditions. Data recorded close to the transition periods of the ACMV system (i.e., 6:00-10:00 each weekday morning and 18:00-20:00 each weekday evening) were excluded to avoid potential biases caused by time-varying indoor temperatures. One-way ANOVA tests were performed to compare size-resolved I/O ratios under different ACMV operation modes. We applied a probability of 0.05 as the threshold in testing for statistical significance (SPSS 22, IBM Inc., USA).

We conducted side-by-side tests for both the SMPSs and OPSs during light-haze and clear-sky periods with outdoor $PM_{2.5}$ concentrations between 20 μg m^{-3} and 60 μg m^{-3}. Adjustment factors based on these comparisons were applied to minimize the differences between individual instruments throughout the whole monitoring period. The side-by-side tests were carried out in the room where the indoor station was placed. The test duration for the SMPSs was 22.5 h and that for the OPSs was 21 h. In the tests, the monitors recorded data at 1-min intervals, which was consistent with the indoor and outdoor monitoring experiments. We calculated the adjustment factor for each channel using the average of readings in that channel from the paired monitors as a reference value. In each channel, the reference values were averaged over the whole test period and the average was divided by average of readings from each monitor. The calculated adjustment factors are listed in Table S2. The paired monitors were reasonably consistent with each other for both SMPSs and OPSs with most differences smaller than 15%.

2.5. Estimates of particle removal efficiencies of the ACMV system

We estimated size-resolved single-pass particle removal efficiencies of the ACMV system, which are believed to be mainly attributable to the MERV 7 filters. Various ACMV components, including filters, coils, and ducting, may contribute to particle removal when the system was operating; however, filters are believed to contribute the most to removal as other components should play limited roles, especially for fine particles [24, 25]. The filters remove the majority of coarse particles as they are the first layer of defense in the ACMV system (as shown in Figure 1) and they have much higher removal efficiency for coarse particles than for fine and ultrafine particles.

Equation 1, based on material balance, describes the time dependent indoor particle number concentration:

$$\frac{dN_{i,in}}{dt} = \lambda N_{i,out}(1-\eta_i) - \lambda_r N_{i,in}\eta_i - \beta_i N_{i,in} - \lambda N_{i,in} \qquad (1)$$

Here, $N_{i,in}$ is the indoor number concentration of particles in the i^{th} size bin (particles cm^{-3}); t is time (h); $N_{i,out}$ is the outdoor number concentration of particles in the i^{th} size bin (particles cm^{-3}); λ is the air-exchange rate (h^{-1}); η_i is single-pass removal efficiency of the ACMV system for particles in size bin i (unitless); λ_r is the recirculation rate of the indoor air in the ACMV system (h^{-1}); and β_i is the indoor deposition rate of particles in the i^{th} size bin (h^{-1}). In developing Equation 1, we assumed balanced volumetric flows (appropriate for near-isothermal conditions), no particle resuspension or generation indoors, no coagulation of particles, and no phase-change processes. We also assumed that during monitoring, when the mechanical ventilation was on, there was no particle infiltration from outdoors to indoors that would bypass the filter. In addition, we assumed that the air-exchange rate of the indoor environment was constant. We treated the filter efficiencies for particles of specific sizes to be identical for makeup and recirculated air, since there are no separate prefilters in the system.

To solve Equation 1, we apply time averaging, neglecting any change of particle number concentration in the indoor environment and assuming that $N_{i,in}$ and $N_{i,out}$ are not correlated in time with λ, η_i, λ_r or β_i. The result is Equation 2:

$$\frac{\overline{N_{i,in}}}{\overline{N_{i,out}}} = \frac{\lambda(1-\eta_i)}{\lambda_r\eta_i + \beta_i + \lambda} \qquad (2)$$

Here, $\overline{N_{i,in}}$ is the indoor time-averaged number concentration of particles in the i^{th} size bin (particles cm^{-3}) and $\overline{N_{i,out}}$ is the corresponding outdoor value (particles cm^{-3}). Considering $\overline{N_{i,in}} / \overline{N_{i,out}} = (I/O)_i$, we transformed Equation 2 to Equation 3 for calculating removal efficiency of the ACMV system for particles in each size bin:

$$\eta_i = \frac{\lambda - (\beta_i + \lambda)(I/O)_i}{\lambda + \lambda_r (I/O)_i} \qquad (3)$$

Here, $(I/O)_i$ is time-averaged ratio of indoor to outdoor particle concentrations in the i^{th} size bin (unitless). Before undertaking the calculations, we first estimated the size-resolved indoor particle deposition rates (β_i). In this study, the β_i values are based on the deposition model developed by Riley et al. [26]. Table S3 presents the calculated β_i value for each effective particle size. In the indoor environment, as shown in Figure 1, the air exchange rate was 3.8 h^{-1} and the recirculation rate was 34.2 h^{-1}. Size-resolved particle removal efficiencies of the ACMV system when it was operated in both Mode 1 (both AC on and MV on) and

Mode 2 (AC off and MV on) were computed based on the corresponding measured particle I/O ratios, utilizing Equation 3.

3. Results and Discussion

3.1. Summary of indoor and outdoor particle number concentrations

Table S4 summarizes the time-weighted and size-resolved indoor and outdoor particle number concentrations during and after the 2013 haze. For all haze levels, particles smaller than 0.37 μm account for most particles by number. In each size range, the indoor concentrations were always lower than the corresponding outdoor concentrations.

3.2. Size-resolved outdoor particle concentrations

Figure 2 illustrates time-averaged volume-weighted size distributions ($dV/d\log D_p$) measured outdoors for particles with diameters 0.01-10 μm for the four haze conditions. Overall, particle volume concentrations for the heavy haze days are approximately seven times higher than on clear-sky days, with ratios ranging from 4 to 60 across particle sizes. Compared with the clear-sky days, the total volume concentration is two times higher for light haze and five times higher for moderate haze. It is noteworthy that submicron particles account for approximately half (45-54%) of the total volume distribution for hazy days, whereas the percentage was smaller (35%) for clear-sky conditions. There is an evident shift in the peak of the submicron size distribution as the haze level increases. The peak diameter was 0.18 μm for clear-sky conditions and progressively increased to approximately 0.42 μm for the moderate and heavy haze days. This observation suggests the occurrence of substantial secondary growth of particles probably owing to a combination of condensation and coagulation during the haze episode.

Figure 3 illustrates the time-averaged and size-resolved particle number distribution (d*N*/dlog D_p) for particles with diameters of 0.01-10 μm, again sorted according to haze level. The striking feature of this figure is the prominence of a count-weighted peak, centered at a diameter of about 0.07 μm diameter, for clear-sky conditions. For hazy conditions, the peak shifts to a larger particle size of about 0.2 μm diameter, for which light scattering would be much more efficient.

Total number concentrations of ultrafine particles (0.01-0.2 μm) for hazy days were less than measured for clear-sky conditions. Specifically, levels were 13,100 ± 6,500, 11,400 ± 4,800, and 14,300 ± 10,800 particles cm^{-3} for heavy, moderate and light haze days, respectively, versus 23,700 ± 9,200 particles cm^{-3} for clear-sky conditions. Qualitatively similar observations have been reported by Betha et al. [27] and Mielonen et al. [28].

A plausible factor contributing to the shift in sizes is the different sources of ultrafine particles and the associated growth processes. On hazy days, the primary source of submicron particles over Singapore would be the agricultural fires in Sumatra, approximately 300 km to the west (as shown in Figure S1). It would take a day or two for pollutants emitted from this locale to travel to Singapore. The time scale would enable the ultrafine particles to grow to sizes larger than 0.10 μm in diameter [29]. Figure 3 shows that the count-weighted size distribution has a peak at approximately 0.17 μm for both the heavy and moderate haze days, whereas the peak occurs at 0.07 μm on the clear-sky days. For clear-sky conditions, probable sources of ultrafine particles measured in Singapore would be local emissions, including industrial and vehicular activities [30]. The proximity of these sources to the monitoring station offers much less time for ultrafine particle to grow through photochemically driven condensation.

Additional evidence about the importance of time for condensational growth of haze particles can be found in comparing the 2013 haze episode here to a 2009 haze event triggered by local biomass burning in Singapore [30]. During the 2009 haze, the mean hourly total particle number concentration was 37,800 particles cm^{-3} (5.6-560 nm), which was 3× that in the current study. During the 2009 haze, there was little time for newly generated ultrafine particles to grow to submicron particles given the close proximity between the monitoring and emissions sites. Differences in the peak diameters of the count-weighted size distribution (0.17 μm during the 2013 haze *versus* 0.06 μm during the 2009 episode) highlight the importance of reaction time as a factor influencing particle size distributions.

The findings shown in Figures 2 and 3 indicate that particles larger than 0.1 μm contributed the most to the outdoor particle pollution during the 2013 haze episode. The findings improve our understanding of the size distributions of particles originating from agricultural biomass burning upwind of Singapore. Because of the frequent recurrence of transboundary haze in Singapore, knowledge about particle size-distributions is useful for developing technology and policy to mitigate the adverse effects of haze particles. In Section 3.3, we evaluate indoor-outdoor relationships for particles in a mechanically ventilated building. Since the ultrafine particle concentrations were observed not to increase during the haze, we focus on particles with diameters of 0.3-10 μm and consider whether there are systematic differences among the four different outdoor pollution conditions.

3.3. Time-resolved outdoor and indoor particle concentrations

Figure 4 shows time-resolved indoor and outdoor particle volume concentrations ($\mu m^3\ cm^{-3}$) in three size bins (0.3-1.0 μm, 1.0-2.5 μm and 5.0-10.0 μm) for one typical day each for the heavy, moderate, and light haze conditions. In Figure 4, the ACMV was operating in Mode 1 (AC on + MV on) for 07:30-18:30 and in Mode 2 (AC off + MV on) for other times.

Figure 4 frames a, b, and c show that indoor particle concentrations in the size range 0.3-1.0 μm were always lower than the corresponding outdoor concentrations. Furthermore, concentrations of these smaller sized particles tracked the corresponding outdoor concentrations closely throughout the day. Temporal patterns of indoor concentrations were attenuated and delayed when compared with the corresponding outdoor concentrations. The indoor concentration was approximately half of the outdoor concentration. This attenuation is mainly attributable to the ACMV system's filtration effects on outdoor particles in the process of transporting air from outdoors to indoors and recirculating it; otherwise, the indoor environment is well isolated from the outdoors by the building envelope. The data also reveal a time lag of approximately 15 min between sudden changes in outdoor concentrations and corresponding changes indoors. That lag is consistent expectations: it is approximately the reciprocal of the measured air-exchange rate of 3.8 h^{-1}. For the three haze conditions, indoor average volume concentrations for particles sized 0.3-1.0 μm were 43.6 μm^3 cm^{-3}, 20.5 μm^3 cm^{-3}, and 5.5 μm^3 cm^{-3}, respectively; each of these values is higher than that for the clear-sky conditions (4.8 μm^3 cm^{-3}).

For particles with diameters in the range 1.0-2.5 μm, indoor concentrations were much lower than corresponding outdoor concentrations (Figure 4 frames d, e, and f). Impaction and interception control particle filtration efficiency in this size range and are much more efficient for these particles than for those in the 0.3-1 μm diameter range, for which the ACMV system exhibited a weaker attenuation effect [31]. For the 1.0-2.5 μm diameter range, indoor peak concentrations are approximately 20% of the corresponding outdoor peak concentrations. Despite attenuation, indoor concentrations were still notably higher when the outdoor concentrations were elevated during the haze. For heavy, moderate and light haze days, the indoor mean volume concentrations in this size bin were 8.1, 7.6 and 1.1 times the clear-sky values, respectively.

For particles in the diameter range 2.5-10.0 μm, there is no evident temporal covariation between indoor and outdoor concentrations (Figure 4 frames g, h, and i). The indoor volume concentrations of particles in the diameter range 2.5-10.0 μm were consistently lower than 5 μm^3 cm^{-3} and were comparable across the different haze intensities, even though the outdoor concentrations were markedly different for these days. These findings indicate that the ACMV system in this building effectively protects occupants against outdoor particles larger than 2.5 μm. The effectiveness of the ACMV system in limiting penetration and persistence of these coarse particles from outdoors results from the high proportion of recirculation flow (90%). Even though the single-pass efficiency of the MERV 7 filters is only moderate, the multiple passes of indoor air through the filters yields a high overall effectiveness in reducing airborne coarse particle concentrations.

In Section 3.2, we reported that particles larger than 0.1 μm dominated the particle volume or mass concentrations during the haze. Here, we have shown that the ACMV system was effective at removing particles larger than 2.5 μm under normal operation. Combining this information, we could state that, in the absence of important indoor particle sources, occupants of a building with a conventional ACMV system during the haze episode would mainly be exposed to particles in the diameter range 0.1-2.5 μm. Recognizing the importance of adverse human health effects associated with exposure to fine particles, it would be of scientific and public health value to develop improved strategies to mitigate indoor fine particle pollution from outdoor sources in this size range, especially during occasions of extreme outdoor pollution such as the Singapore 2013 haze. Such information might assist government agencies in setting policies to protect building occupants from excessive particle exposure during haze episodes.

3.4. Particle I/O ratios

Figure 5 shows the time averaged and size-resolved I/O ratios of particles with diameters of 0.01-6.0 μm for two ACMV operation modes (Mode 1: both AC and MV on; Mode 2: AC off and MV on). Table 1 reports the time, date and haze levels of the datasets for the I/O ratios calculation. The small number of entries in Table 1 occurs because we only used datasets when the both indoor and outdoor SMPSs were functioning properly. In both modes, the ACMV system is the major pathway by which outdoor particles migrate indoors.

The I/O ratios for all particle sizes are smaller than one, as expected given the absence of any notable indoor particle source. For particles in the size range 0.01-0.2 μm, the mean I/O ratios are in the range of 0.17-0.65 and there is a tendency for the I/O ratio to increase with increasing particle size. For particles of 0.1-1.0 μm, the size-resolved mean I/O ratios are in the range 0.12-0.65. The highest mean I/O ratios occur for particle diameters of approximately 0.3 μm. The mean I/O ratios decrease sharply when the particle size is larger than 0.3 μm. The trend for size-resolved I/O ratios of particles with diameters 0.3-5.0 μm generally agrees with the findings reported by Gupta and Cheong. [32] for ACMV-dominated indoor environments. These findings also align with theoretically predicted results that fibrous particle filters usually have minimum efficiencies for diameters in the range 0.05-0.5 μm [11].

Figure 5 suggests that, in addition to mechanical ventilation and active filtration, the operation of air conditioning influenced the indoor/outdoor particle ratio. There is a trend such that when the air conditioning was on, the I/O ratios for particles between 0.17 μm and 2.5 μm were lower than when the air conditioning was off. Conversely, for particles smaller than about 0.1 µm, there is a tendency for the I/O ratio to be higher when the air conditioning was on as compared to the air-conditioning off state.

We have compared the I/O ratios in these two modes using one-way ANOVA tests. The statistical analysis reveals that the differences are statistically significant ($p \leq 0.05$) for particles in all size bins between 0.17 µm and 2.5 µm, except for the size bin 0.3-0.374 µm (p = 0.29). These findings suggest that the ACMV system has higher removal efficiency for particles in this larger size range with active cooling by the air conditioning system.

In Singapore's tropical climate, whenever air conditioning is operating, the cooling coil would receive a flow of condensing water from the humid air stream passing over its cooled surfaces. The elevated removal efficiency suggests the possibility of enhanced removal of particles onto the wet surface of the cooling coil when air conditioning is on. The presence of a water film would narrow the gaps between fins. The process of condensation would also induce net transport of particles toward the condensing surfaces through the mechanism of diffusiophoresis. There may also be a thermophoretic influence inducing particle migration from the warmer air toward the cooler fins. It has been recognized that a cooling coil can contribute to removing particles from airstreams [24, 33, 34]. At present, the processes and mechanisms are not well understood and we know of no previously published data of the type presented in Figure 5.

For smaller particles, with diameters of 0.01-0.154 µm, we observe a trend of higher I/O ratios when the air conditioning is on compared to when it is off. However, one-way

ANOVA results reveal that the differences between the I/O ratios are statistically significant ($p < 0.05$) only for particles in a few size bins, 0.0205–0.0365 µm and 0.0649–0.154 µm. This trend contradicts the theoretically predicted results by Waring and Siegel [34]. In their study, higher deposition rates were predicted for ultrafine particles onto a wet surface than onto the dry surface of a cooling coil. We speculate that the higher I/O ratios that we observe for these smallest particles might be attributable to the growth of ultrafine particles owing to condensation as the air stream is cooled. The condensing species could include water and also semivolatile organic compounds in the air stream whose partitioning between the gas and particle phase is materially influenced by temperature.

The information in this study is insufficient to conclusively explain these observations. In future studies, laboratory tests with well-controlled operational parameters could serve to elucidate the influence of cooling coil operation on particle behavior across different size ranges.

It is conceivable that variations of outdoor particle concentrations might indirectly influence I/O ratios. However, our data indicate that the difference of time-averaged outdoor particle concentrations between the daytime (AC on) and nighttime (AC off) conditions is relatively small, i.e. less than a 10% difference. Consequently, we consider that variations in outdoor levels did not significantly affect the I/O ratios between the two ACMV operation modes in this investigation.

3.5. Particle removal efficiencies

Figure 6 depicts the time-averaged and size-resolved removal efficiencies of the ACMV system for particles with diameters of 0.01-6.0 μm for two ACMV operation modes (Mode 1: both AC and MV on; Mode 2: AC off and MV on). The single-pass removal efficiencies range from 5% to 80% in both ACMV operation modes, with the respective lowest and highest efficiencies occurring at 0.1 μm and 3.71 μm in Mode 1, and 0.33 μm and 6.0 μm in Mode 2. More specifically, the removal efficiencies are smaller than 30% for particles of diameter 0.01-1.0 μm in Mode 1 and for particles of diameter 0.015-1.12 μm in Mode 2.

The size–resolved particle removal efficiencies calculated in the current study have a similar profile with those reported by Azimi et al. [35] (Figure 5 of their paper), which were based on the measured single-pass sized-resolved removal efficiencies for particles of 0.03-10 μm by Hecker and Hofacre. [36].

When the mechanical ventilation system was on, indoor air passed through the filters and cooling coil an average of nine times before being replaced by outdoor air, and particle concentrations would diminish during each pass. Consequently, the overall effectiveness of the ACMV system with MERV 7 filters is much higher than the corresponding single-pass efficiency. However, the MERV 7 filters are still insufficient to protect indoor occupants from fine particles of outdoor origin during the haze episode when considering both the low single-pass particle removal efficiencies and the findings reported in Section 3.3. The low removal efficiency of the MERV 7 filters for ultrafine particle also indicates that the filters may fail to protect indoor occupants from ultrafine particles of outdoor origin even during clear-sky periods, when outdoor ultrafine particle number concentrations are elevated (Section 3.2). The high removal efficiencies for particles > 3 μm indicate that the MERV 7 filters work effectively to remove the coarse particles. The filters' improved removal efficiencies for coarse particles may be influenced by accumulated particles on the filters as the filters are used.

Comparisons of the particle removal efficiencies in the two ACMV operation modes reveal that the removal efficiencies for particles of 0.37- 3.74 μm are significantly higher in the AC on mode than in the AC off mode (one-way ANOVA test, $p < 0.05$). The wet cooling coil surface in the AC on mode results in increases of 4-25% in the removal efficiencies for particles with diameters of 0.37-3.74 μm when compared with the dry cooling coil surface in the AC off mode. The findings suggest that during the haze episode, air conditioning operation could contribute to the attenuation of outdoor particles in this size range in indoor air. However, it is also possible that enhanced particle deposition to wet cooling coil surfaces could contribute to fouling of those surfaces over the long term.

4. Conclusions

During the 2013 haze in Singapore, the outdoor mean size distribution of particles larger than 0.2 μm in diameter was remarkably higher than on clear days. Overall, particles of 0.1-1.0 μm accounted for large increases, with aggregate volume concentrations that were 5 to 60 times higher than during the clear-sky conditions that prevailed a few weeks after the haze episode. There was an evident size shift of the peak particle size to larger diameters within the accumulation mode. This phenomenon might be a consequence of secondary growth of organic aerosol induced by photochemical reactions during the haze.

In a mechanically ventilated and air conditioned room on the NTU campus, equipped with MERV 7 grade filters, indoor particles in the size range 0.3-1.0 µm followed the time pattern of outdoor particle concentrations, with some attenuation and a short lag time. The correlations between indoor and outdoor particles in the size range 1.0-2.5 µm were moderate and correlations were not observed for larger particles. Relative to the clear-sky conditions, indoor concentrations of particles in the size range 0.3-2.5 µm increased by factors of 2 to 14 during the haze. Any such increase for larger particles was marginal.

The mean I/O ratio and removal efficiency of the ACMV system of particles was observed to vary with particle size as would be expected. A conventional ACMV system with MERV 7 filters is insufficient to protect building occupants from high exposures to fine particles of outdoor origin under extraordinary circumstances such as the 2013 haze. More effective strategies to protect the public are needed for the recurring transboundary haze.

We observed that both I/O ratios and particle removal efficiencies of the ACMV system varied systematically depending on whether or not the air conditioning was on. Information in the current study is insufficient to fully explain these observations. As yet, there is limited scientific knowledge about how pollutants, such particles, semivolatile organic compounds, bioaerosols, and ozone, interact with cooling coil surfaces. More studies that advance our knowledge of these topics are necessary.

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Figure Captions

Figure 1. Schematic representation of the air-conditioning and mechanical ventilation system for the office, illustrating the flow rates (Q), fans (F), filter and coil in the system. For air-flow rates, the subscripts F, R and EX denote forced supply (make-up), recirculation and exfiltration, respectively.

Figure 2. Size-resolved time-averaged outdoor particle volume concentrations ($dV/d(\log D_p)$) sorted according to four particle pollution categories. The V value in the legend refers to total average particle volume concentration (0.01-10 µm) in each particle size category.

Figure 3. Size-resolved time-averaged outdoor particle number concentrations ($dN/d(\log D_p)$) sorted according to four particle pollution categories. The N value in the legend refers to total average particle number concentration (0.01-10 µm) in each particle size category.

Figure 4. Time-resolved indoor and outdoor particle volume concentrations (dV) in different particle size ranges and for different degrees of haziness.

Figure 5. Size-resolved particle indoor/outdoor (I/O) ratios in two different air-conditioning operation modes (AC on and AC off). Mechanical ventilation was provided at the same volumetric flow rate in both cases. The error bars refer to standard deviations.

Figure 6. Size−resolved particle removal efficiencies (η_i) of the ACMV system in two different air-conditioning operation modes (AC on and AC off). Mechanical ventilation was provided at the same volumetric flow rates in both cases. The error bars refer to standard deviations.

Table 1. Time, date and haze levels used for time-averaged, size-resolved I/O ratio calculations.

AC mode (MV on)	Time	Date	Haziness
AC on	11:21-15:35	14 June 2013	light
	9:00-18:00	17 June 2013	moderate
	13:56-16:59	20 June 2013	heavy
	13:06-17:00	27 June 2013	light
AC off	11:00-14:00	16 June 2013	moderate
	21:00-24:00	22 June 2013	heavy
	21:00-24:00	27 June 2013	light
	00:00-6:00	28 June 2013	light

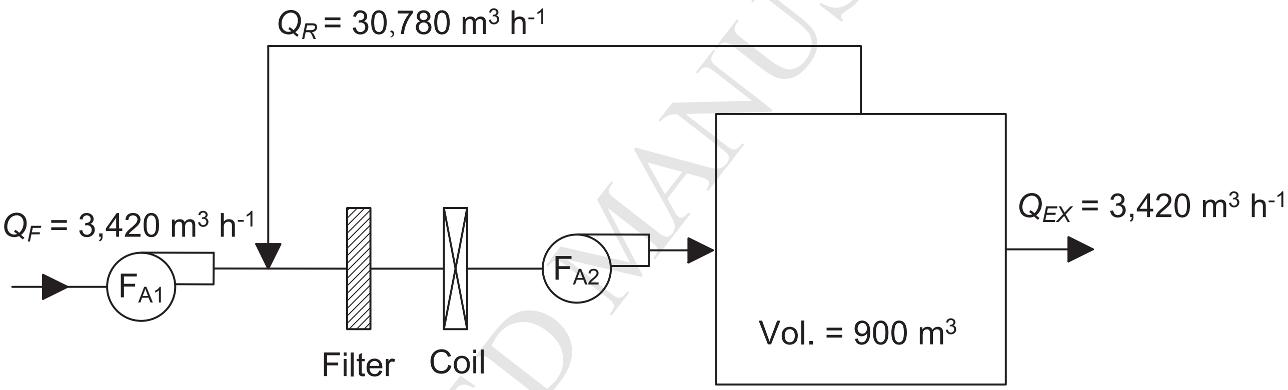

Figure 1. Schematic representation of the air-conditioning and mechanical ventilation system for the office, illustrating the flow rates (*Q*), fans (*F*), filter and coil in the system. For air-flow rates, subscripts *F*, *R* and *EX* denote forced supply (make-up), recirculation and exfiltration, respectively.

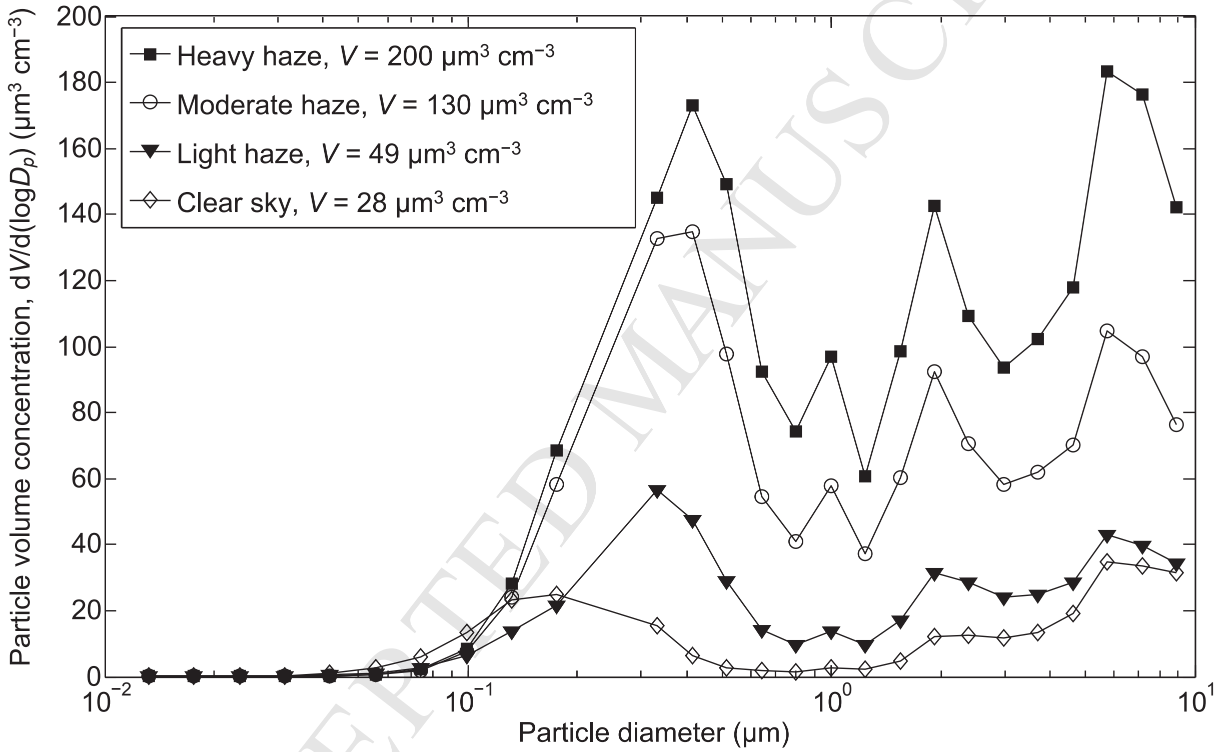

Figure 2. Size−resolved time-averaged outdoor particle volume concentrations (dV/d(log D_p)) sorted according to four particle pollution categories. The V value in the legend refers to total average particle volume concentration (0.01−10 μm) in each particle size category.

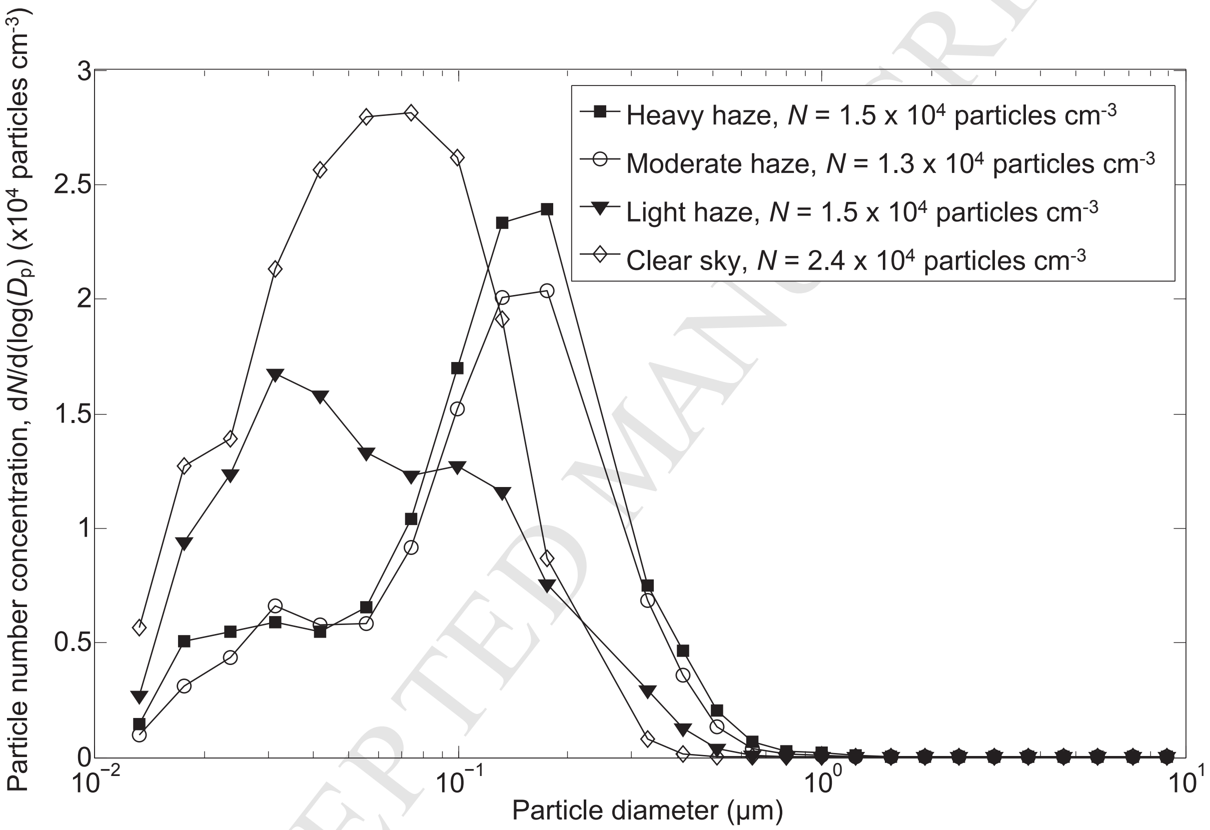

Figure 3. Size−resolved time-averaged outdoor particle number concentrations ($dN/d(\log D_p)$) sorted according to four particle pollution categories. The *N* value in the legend refers to total average particle number concentration (0.01−10 μm) in each particle size category.

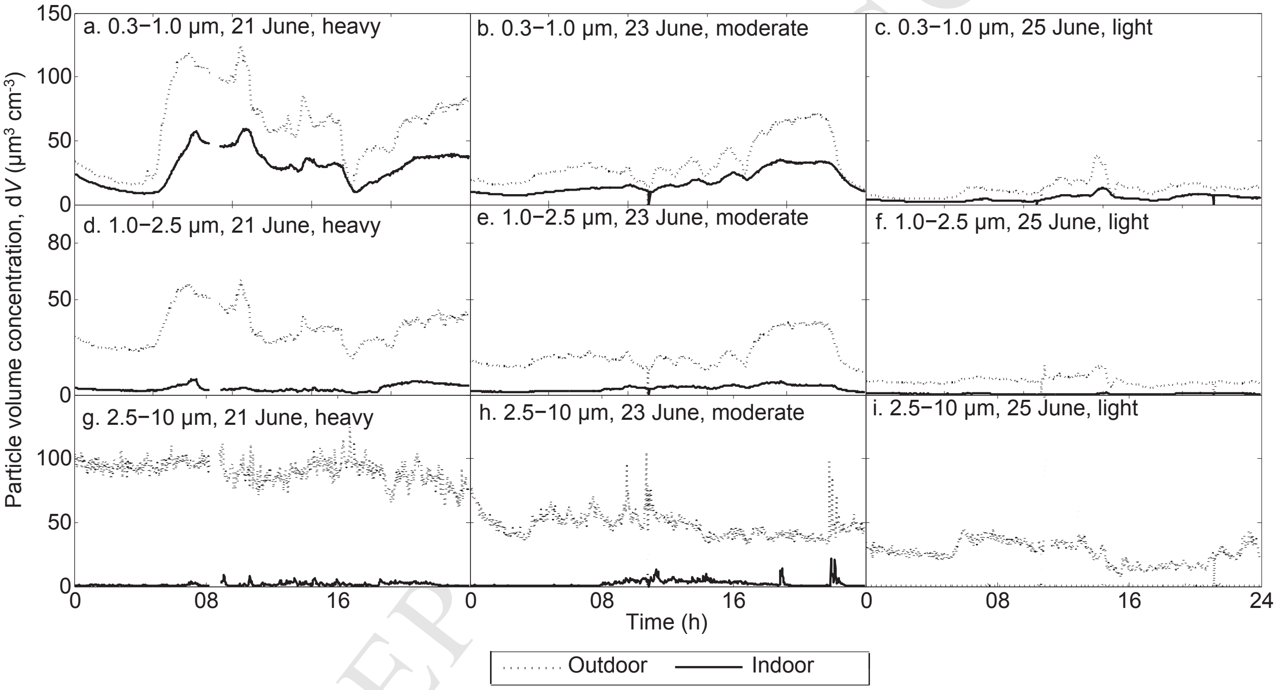

Figure 4. Time-resolved indoor and outdoor particle volume concentrations (dV) in different particle size ranges and for different degree of haziness.

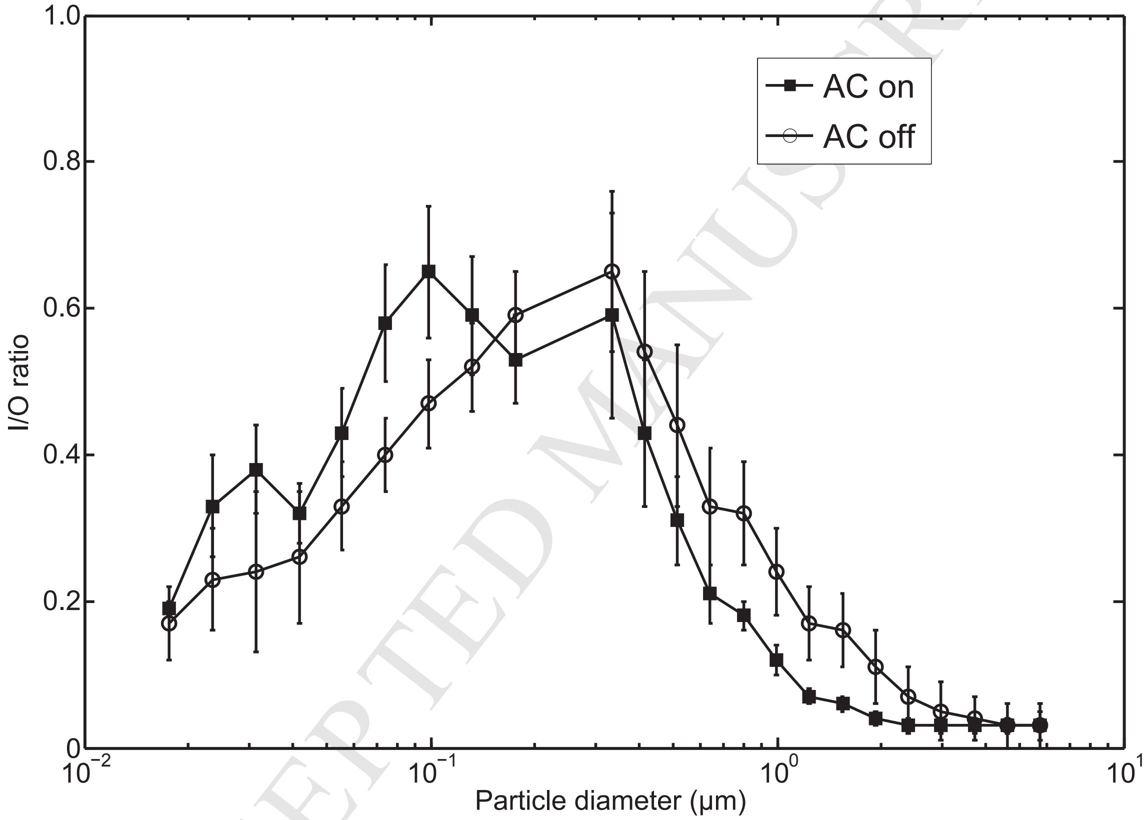

Figure 5. Size-resolved particle indoor/outdoor (I/O) ratios in two different air-conditioning operation modes (AC on and AC off). Mechanical ventilation was provided at the same volumetric flow rates in both cases. The error bars refer to standard deviations.

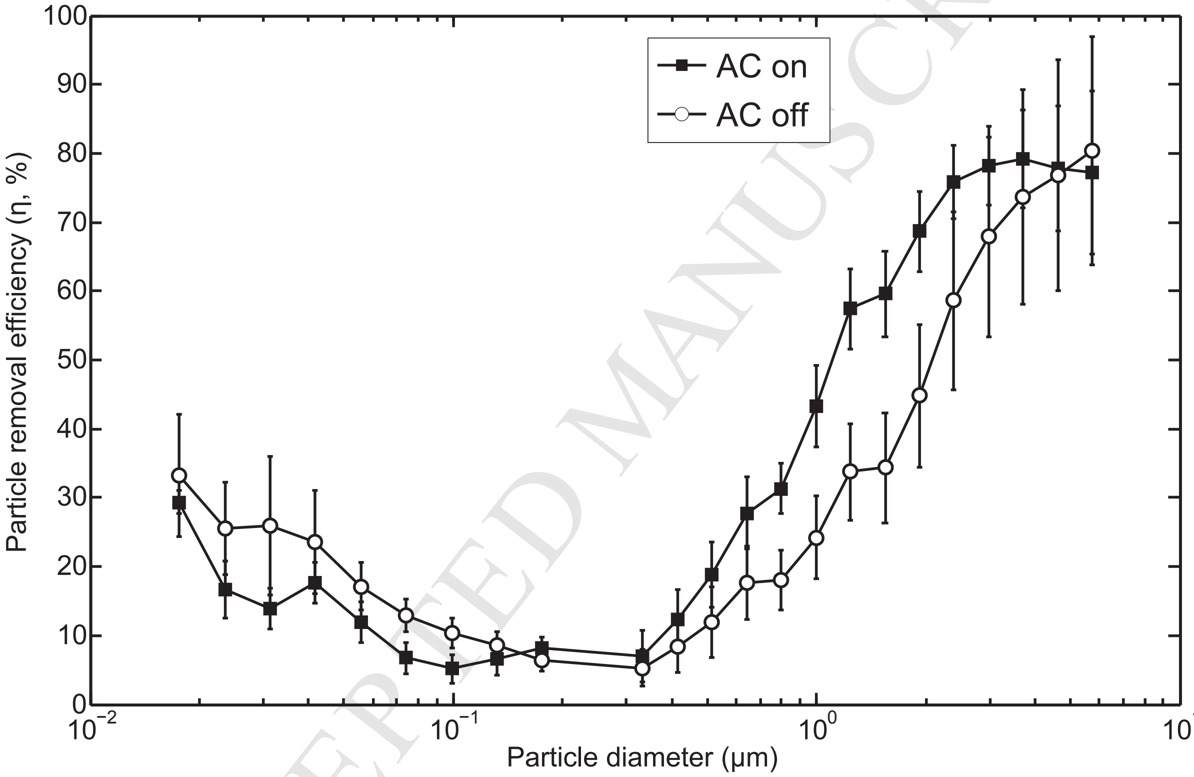

Figure 6. Size−resolved particle removal efficiencies (η_i) of the ACMV system in two different air-conditioning operation modes (AC on and AC off). Mechanical ventilation was provided at the same volumetric flow rates in both cases. The error bars refer to standard deviations.

HIGHLIGHTS (Chen et al., *Building and Environment*, 2015)

- Monitored indoor and outdoor particles during and after the 2013 haze in Singapore.
- Haze mainly causes increases in concentrations of particles larger than ~ 0.2 μm.
- ACMV system attenuated penetration and persistence of outdoor particles indoors.
- AC operation altered the indoor/outdoor concentration ratios of fine particles.
- MERV 7 filters provided < 30% removal efficiencies for particles of 0.01-1.0 μm.