

Characteristics of Indoor Aerosols in Residential Homes in Urban Locations: A Case Study in Singapore

Rajasekhar Balasubramanian & Sheng Sheng Lee

To cite this article: Rajasekhar Balasubramanian & Sheng Sheng Lee (2007) Characteristics of Indoor Aerosols in Residential Homes in Urban Locations: A Case Study in Singapore, Journal of the Air & Waste Management Association, 57:8, 981-990, DOI: [10.3155/1047-3289.57.8.981](https://doi.org/10.3155/1047-3289.57.8.981)

To link to this article: <https://doi.org/10.3155/1047-3289.57.8.981>



Published online: 29 Feb 2012.



Submit your article to this journal [↗](#)



Article views: 1058



View related articles [↗](#)



Citing articles: 23 View citing articles [↗](#)

Characteristics of Indoor Aerosols in Residential Homes in Urban Locations: A Case Study in Singapore

Rajasekhar Balasubramanian

Division of Environmental Science and Engineering, National University of Singapore, Singapore

Sheng Sheng Lee

Department of Chemical and Biomolecular Engineering, National University of Singapore, Singapore

ABSTRACT

As part of a major study to investigate the indoor air quality in residential houses in Singapore, intensive aerosol measurements were made in an apartment in a multistory building for several consecutive days in 2004. The purpose of this work was to identify the major indoor sources of fine airborne particles and to assess their impact on indoor air quality for a typical residential home in an urban area in a densely populated country. Particle number and mass concentrations were measured in three rooms of the home using a real-time particle counter and a low-volume particulate sampler, respectively. Particle number concentrations were found to be elevated on several occasions during the measurements. All of the events of elevated particle concentrations were linked to indoor activities based on house occupant log entries. This enabled identification of the indoor sources that contributed to indoor particle concentrations. Activities such as cooking elevated particle number concentrations $\leq 2.05 \times 10^5$ particles/cm³. The fine particles collected on Teflon filter substrates were analyzed for selected ions, trace elements, and metals, as well as elemental and organic carbon (OC) contents. To compare the quality of air between the indoors of the home and the outdoors, measurements were also made outside the home to obtain outdoor samples. The chemical composition of both outdoor and indoor particles was determined. Indoor/outdoor (I/O) ratios suggest that certain chemical constituents of indoor particles, such as chloride, sodium, aluminum, cobalt, copper, iron, manganese, titanium,

vanadium, zinc, and elemental carbon, were derived through migration of outdoor particles (I/O < 1 or ≈ 1), whereas the levels of others, such as nitrite, nitrate, sulfate, ammonium, cadmium, chromium, nickel, lead, and OC, were largely influenced by the presence of indoor sources (I/O > 1).

INTRODUCTION

Over the last few decades, numerous epidemiological studies have independently demonstrated a significant correlation between daily variations in particulate air pollution levels and changes in mortality and morbidity.^{1,2} Although a majority of these time-series studies contended with more readily available data on pollutant concentrations in ambient air, personal exposures may be a relatively better indicator of adverse health effects from a dose-response relationship viewpoint.³ Given that people spend approximately 85% of their time indoors,⁴ a considerable portion of personal exposure to particulate matter (PM) could be derived from the indoor environment. As a result, monitoring and assessment of airborne particles in indoor air have been conducted by many researchers across the United States,⁵⁻⁷ Europe,⁸⁻¹⁰ Australia,^{11,12} and Asia.¹³⁻¹⁷ The key findings on the physical and chemical properties of indoor particles and their potential sources are summarized in the review article by Wallace.¹⁸

Earlier studies were mainly focused on the mass concentrations of total suspended particles or particles with a median aerodynamic diameter $\leq 10 \mu\text{m}$ (PM₁₀). However, in recent years, attention was directed toward particles with a median aerodynamic diameter $\leq 2.5 \mu\text{m}$ (PM_{2.5}) and number concentration, for which smaller particles contribute much more significantly than larger particles. These monitoring data are deemed to be important in view of the greater health hazards of smaller particles.^{19,20} Smaller particles are capable of penetrating more deeply into the human respiratory tract, near the gas exchange region,²¹ and translocate beyond to other target organs.²² In addition, the type of chemical components adsorbed on particles is also an important factor of toxicity, and small particles, with high surface area per unit mass, can contain more toxins.²³ Hence, a comprehensive knowledge of the particle number concentration and chemical speciation of PM_{2.5} in indoor environments is necessary.

IMPLICATIONS

Indoor particulate pollution has received considerable attention in Asia in view of rapid economic growth and urbanization. This work presents a case study regarding the number concentration of airborne particles and the chemical composition of fine particulate matter (PM_{2.5}) in a typical apartment in a multistory building in Singapore. The time-activity diary maintained by the occupants, together with the indoor- to-outdoor ratio of the chemical components of PM_{2.5}, is used to identify the possible sources of indoor aerosols. The data set can provide a scientific basis for assessment of indoor air quality in multistory buildings located in densely populated areas.

In residential homes, people are exposed both to indoor-generated particles and to particles that enter the building along with outside air. Significant indoor particle sources of $PM_{2.5}$ are combustion processes, such as tobacco smoking, cooking, candle burning, and incense burning,^{24,25} of which emissions contain a host of organic and inorganic material,^{26,27} although noncombustion sources, such as cleaning, also give rise to elevated levels of fine particles.^{28,29} Other than these primary sources, submicrometer-sized secondary particles can also be formed from gaseous pollutants through gas-to-particle conversion.³⁰ On the other hand, particles that are resuspended by human movement and crustal/soil matter are typically above $1\ \mu m$.³¹ The indoor particulate air pollution could even get worse in densely populated countries, for example Singapore, where buildings are also located in the proximity of other outdoor combustion sources, such as on-road vehicles. Depending on the level of infiltration of particles from outdoor sources, the indoor air quality can be very different from that of outdoors, even in the absence of any indoor sources.³² It is, therefore, essential to understand the relationship between indoor and outdoor concentrations, as well as the origin and transport of particles indoors.

This paper describes and discusses a short case study on the monitoring and assessment of particulate air pollution in a typical residential home in Singapore based on the analysis of the physical and chemical characteristics of particles found at three indoor and one residential outdoor environments. The physical properties investigated included the real-time number concentration of particles less than $1\ \mu m$ and the time-integrated mass concentration of $PM_{2.5}$, whereas the major chemical constituents, namely ions, metals, elemental carbon (EC),

and organic carbon (OC) were determined. Based on time activity logs maintained by the residential occupants, the main indoor sources of particles were identified, and their emission rates were evaluated and compared. Furthermore, indoor and outdoor air quality were compared, and the indoor/outdoor ratio (I/O) was calculated to provide an indication of the particle origins.

EXPERIMENTAL WORK

Description of Sampling Sites

The sampling site is a residential home on the eighth story of a 14-story building in the northwestern part of Singapore in Choa Chu Kang (Figure 1). The flat is within 50 m from a main road, where traffic volume averaged approximately 3000 vehicles per hour during the morning and evening peak hours, from 7:00 to 10:00 a.m. and 6:00 p.m. to 12:00 a.m., respectively, and approximately 800 vehicles per hour during nonpeak hours. The entire flat consists of two levels, and the layout and orientation of all the rooms are illustrated in the figure. Three of these rooms, namely, the living room ($90\ m^3$) on the first level and the master bedroom ($60\ m^3$) and a bedroom ($48\ m^3$) on the second level were selected for this study, because the five occupants spent most of their time there. The windows and/or doors of the living room and master bedroom face the road and were affected by particulate pollution from the on-road traffic as a result of their downwind positions during sampling. On the other hand, the bedroom on the opposite end is 30 m from other similar residential buildings and, thus, experienced only light winds because of street canyon effect. When air conditioning was switched on in the master bedroom and bedroom, the windows and doors in the rooms were shut. When air conditioning was not switched on, the windows

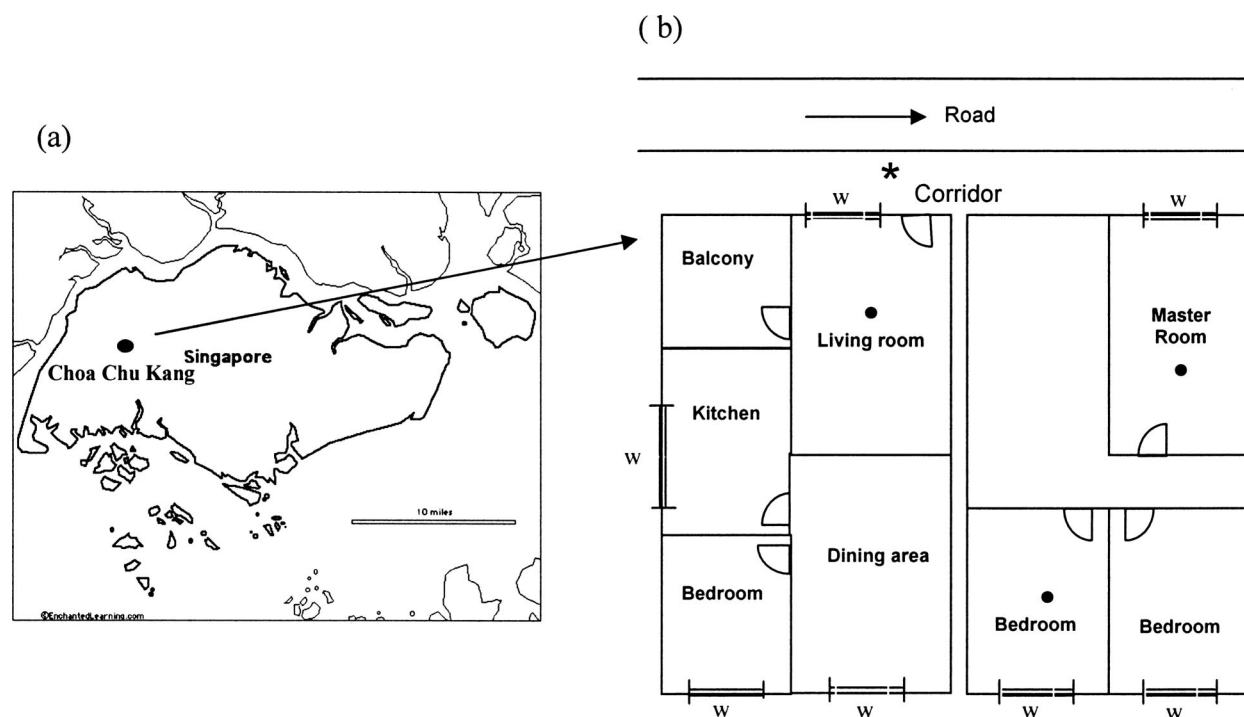


Figure 1. (a) Map of Singapore with (b) the floor plan of the sampling site at Choa Chu Kang. w = the windows in each of the rooms. Samples were taken from the rooms (living room, master bedroom, and bedroom), identified by ●, and outdoor sampling location identified by *.

were open, though the door remained shut. Outdoor sampling and measurements were carried out on the eighth floor along the corridor in front of the flat of which the position from the road is same as that of the living room.

Instrumentation

Three quiet and small instruments were chosen for this study to minimize disturbances to the occupants. The first is a Model 3007 Condensation Particle Counter (CPC; TSI, Inc.), which can measure the number concentration of 0.01–1.00- μm particles in real time. However, continuous operation was not possible, because the CPC had to be switched off every 5 hr to replenish the instrument with reagent-grade isopropyl alcohol required to grow the sub-micrometer-sized particles for detection, and this resulted in discontinuities in the temporal profiles of the particle counts over the 24-hr sampling period. The second and the third were MiniVol portable air samplers (Airmetrics), which can collect $\text{PM}_{2.5}$ by drawing air at a precalibrated flow rate of $0.005 \text{ m}^3 \text{ min}^{-1}$. One of them was equipped with a 47-mm Teflon filter of 2- μm pore size (Pall Corp.) and the other with 47-mm precombusted quartz filter. All of the filters were maintained in the desiccator for ≥ 24 hr before and after use before their weightings with a microbalance (Sartorius AG).

Measurements

The two MiniVols and the CPC were positioned as near to the center of the rooms as possible with the sampling nozzles approximately 1.5 m above the ground to simulate the breathing zone of human beings. Because the instruments were operated concurrently, they were placed in a triangular formation, approximately 1 m apart, so that any one instrument would not affect the airflow of the other. Particulate pollutant concentration was assumed to be homogeneous within the rooms. Measurements took place over 2 weeks, from May 12 to May 23, 2004, with 2 full days of sampling at each of the four sampling sites: living room, master bedroom, bedroom, and corridor. During the sampling period, the temperature and relative humidity (RH) at the sampling sites were read once per hour from 8:00 to 12:00 a.m. The temperature was fairly consistent at all of the sites on all of the sampling days, with a high of 32–34 °C in the daytime and a low of 27–28 °C at night, whereas RH ranged from 79% to 86% under naturally ventilated conditions (air change per hour ~ 0.5 in the living room and master bedroom and ~ 0.7 in the bedroom determined using the SF_6 decay method as described in the paper by See and Balasubramanian³³). When air conditioning was switched on in the master bedroom and bedroom, the temperature and RH dropped to a constant of 23 °C and 40%, respectively. To identify the possible activities affecting the number concentration of particles, the five occupants were requested to note down their movements within the house and the time at which they were involved in activities, for example, cooking in the kitchen, sweeping the floor in the living room, and so forth.

Chemical Analysis

The particles collected on Teflon filters were analyzed for six anions, that is, fluoride (F^-), chloride (Cl^-), nitrite

(NO_2^-), nitrate (NO_3^-), sulfate (SO_4^{2-}), and phosphate (PO_4^{3-}); six cations, that is, lithium (Li^+), sodium (Na^+), ammonium (NH_4^+), potassium (K^+), calcium (Ca^{2+}), and magnesium (Mg^{2+}); and 12 total metals, that is, aluminum (Al), cadmium (Cd), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), titanium (Ti), vanadium (V), and zinc (Zn). On the other hand, particles collected on quartz filters were analyzed for EC and OC content.

To extract water-soluble ions, one half of the Teflon filter was immersed in 20 mL of 18.2 M Ω ultrapure water and placed in an ultrasonic bath at 60 °C for 1 hr. The extracts were then filtered through an Autovial fitted with 0.45- μm membrane filter (Whatman plc) before they were analyzed for ions using the ion chromatography system (Metrohm AG). Detailed information on the analytical method and results are discussed by Karthikeyan and Balasubramanian.³⁴

The other half of Teflon filters was extracted for metals following the procedures described by Karthikeyan et al.³⁵ Briefly, extraction solvent consisting of reagent grade mixtures, 4 mL of 69.5% nitric acid (HNO_3), 2 mL of 30% hydrogen peroxide (H_2O_2), and 0.2 mL of 48% hydrofluoric acid (Hartree-Fock), was added to the filters and processed in the MLS1200 MEGA microwave digestion system (Milestone srl) for 5 min at 250 W, 5 min at 400 W, and 2 min at 600 W. The resulting extracts were analyzed in triplicates for total metals by means of an ELAN 6100 Inductive Coupled Plasma Mass Spectrometer (PerkinElmer, Inc). For the ions and metals analyses, a five-point calibration was performed at the beginning of the day for each analytical batch, and the regression coefficients were found to be above 0.99.

The quartz filters were cut into two halves, one for total carbon (TC) and the other for OC analysis, with EC being indirectly evaluated from the difference between TC and OC concentrations, that is, $\text{TC} = \text{EC} + \text{OC}$. The portion used for EC analysis was combusted at 350 °C for 2 hr to remove OC. A circular filter punch of 6-mm diameter, weighing between 1 and 2 mg, was then taken from both the noncombusted and combusted pieces and subsequently analyzed for carbon content in the 2400 Series II CHNS/O Analyzer (PerkinElmer). The analyzer was run in CHN (carbon, hydrogen, and nitrogen) mode and calibrated using acetanilide (71.09% carbon [C], 6.71% hydrogen [H], and 10.36% nitrogen [N]). Three standards weighing between 1 and 2 mg were run, and the K factor (the slope of the linear calibration plots) values were within the recommended range of 16.5 ± 3.5 for C, 50 ± 20 for H, and 6 ± 3 for N, respectively. For all of the analyses, appropriate field and laboratory blank filters were subjected to the same experimental procedures as the filter samples for each analytical batch, and their measured concentrations were subtracted from the concentration of the samples.

Limitations of the Study Design

Insufficient number of aerosol analyzers and samplers mainly contributed to the limitation of the study design followed in this work. Because both indoor and outdoor

measurements could not be made concurrently, an assumption was made that the outdoor pollutant concentrations did not vary significantly during the sampling days. This assumption is supported by the meteorological, as well as the visibility, data recorded during the sampling days (see Table 1). Both the ambient temperature and humidity, which can affect the particulate concentration, showed little variation on a day-to-day basis. Furthermore, there was no significant change in atmospheric visibility (8.5–9.8 km), suggesting that the skies were relatively clear during the sampling period. According to our previous investigations of ambient air quality in Singapore over a long period of time, PM_{2.5} concentrations in Singapore showed less than 20% deviation in the absence of the influence of external sources.³⁶ Based on these considerations, the average outdoor PM_{2.5} concentration measured on May 21–22, 2004, is taken to be representative of the outdoor particulate concentrations for other sampling days as well. Overall, the present study represents snap-short measurements of residential indoor air quality in multistory buildings, with the major aim being to identify the significant particle sources that people are exposed to in indoor environments before more extensive physical and chemical characterization studies could be conducted for specific sources of interest. As revealed by this study, an important source of indoor air pollution in Singapore is gas cooking, which we investigated recently in detail.^{33,37,38} Although the data available from the present study are insufficient to make major conclusions about the origin of particles indoors, the I/O ratios reported in this work could still be considered to be indicative of potential sources of different chemical components contained in particles.

RESULTS AND DISCUSSION

Physical Characteristics

Particle Number Concentration. Figures 2–4 illustrate the time series of the particle number concentrations measured in the living room, master bedroom, and bedroom, respectively, for each of the sampling days. The outdoor profile obtained on another separate day is also shown in the figures for the sake of comparison. Activities leading to sporadic increases in the particle counts are indicated in the figures. “Cleaning” refers to vacuuming, sweeping, mopping, and dusting, whereas “human activity” refers to the presence of human movements, for instance, walking and making up the bed.

From the temporal variations of the particle number concentration, it can be seen that, in general, higher

particle counts were found indoors than outdoors, suggesting that the indoor environments were more drastically affected by indoor particulate sources than outdoor sources. The exception usually occurred when air conditioning was switched on in the early mornings as observed in Figures 2a, 3a, 3b, and 4a. Particle air filters present in air conditioning units, the closed doors and windows, together with the lack of human activities, account for the stable indoor particle number concentration at approximately $8.5 \times 10^3 \text{ cm}^{-3}$.

An assessment of the time-activity logs provided by the occupants revealed that periods of elevated particle number concentration at the three indoor sampling sites were linked to three main activities, namely, smoking, cooking, and cleaning, although these activities might not have been carried out in the same room. For example, distinct cooking peaks were seen in the living room and the master bedroom during lunch and dinner preparation times despite the fact that the food was cooked in the kitchen. High peaks of $\leq 2 \times 10^5 \text{ cm}^{-3}$ could be attributed to cooking and smoking, whereas smaller peaks $< 1 \times 10^5 \text{ cm}^{-3}$ were because of cleaning and general human activities. The peak values of mostly approximately $1.1 \times 10^5 \text{ cm}^{-3}$ found for cooking in this study are rather comparable to the peak value of $1.26 \times 10^5 \text{ cm}^{-3}$ reported by He et al.¹¹ However, the peak values for smoking and cleaning in this study tend to be on the higher side compared with that study. This could be because of the difference in the location of sampling instruments relative to the sources; for example, air sampling was carried out in the kitchen in the latter study.

Particle Mass Concentration. The PM_{2.5} mass concentrations were calculated, and the values are reported in Table 2. The living room is reported to have the highest mass concentration among the four sampling sites, including the outdoor site, at an average value of $42.5 \mu\text{g}/\text{m}^3$. The elevated particle concentration measured there is conceivable, because tobacco smoking is present only in the living room, which is also exposed to cooking emissions from the kitchen. The average mass concentration in the master room was $22.4 \mu\text{g}/\text{m}^3$, which is lower than that of the bedroom ($31 \mu\text{g}/\text{m}^3$). One possible reason for this difference is that air conditioning was turned on during the night on both sampling days for the master room, as compared with one night only for the bedroom.³⁹ All three rooms have mass concentrations greater or equal to that of the outdoors, as can be seen from Table 2.

Mean I/O ratios based on PM_{2.5} mass concentrations were calculated and are summarized in Table 2. The ratio between indoor and outdoor concentrations of particles also gives an indication as to whether particles found indoors are a result of indoor generation. It is expected that, in the absence of indoor sources, the ratios would be ≤ 1 .^{40,41} The PM_{2.5} ratios were generally > 1 for all the rooms, with the exception of that for the master room on May 21, 2004 (I/O = 0.93). As can be seen from the 24-hr particle count trace in Figure 3b, on this particular day, the peaks occurred at values generally lower than those of the previous day, and this is reflected in the lower mass

Table 1. Meteorological conditions recorded during the sampling period.

Date in May 2004	Temperature (°C)	Humidity (%)	Visibility (km)
12	30 (27–32)	83 (66–94)	8.5
19	30 (28–32)	84 (70–94)	9.0
20	31 (28–34)	79 (59–94)	9.8
21	30 (28–33)	81 (62–94)	9.8
22	30 (28–32)	86 (70–94)	8.5
23	30 (27–33)	82 (63–100)	9.0

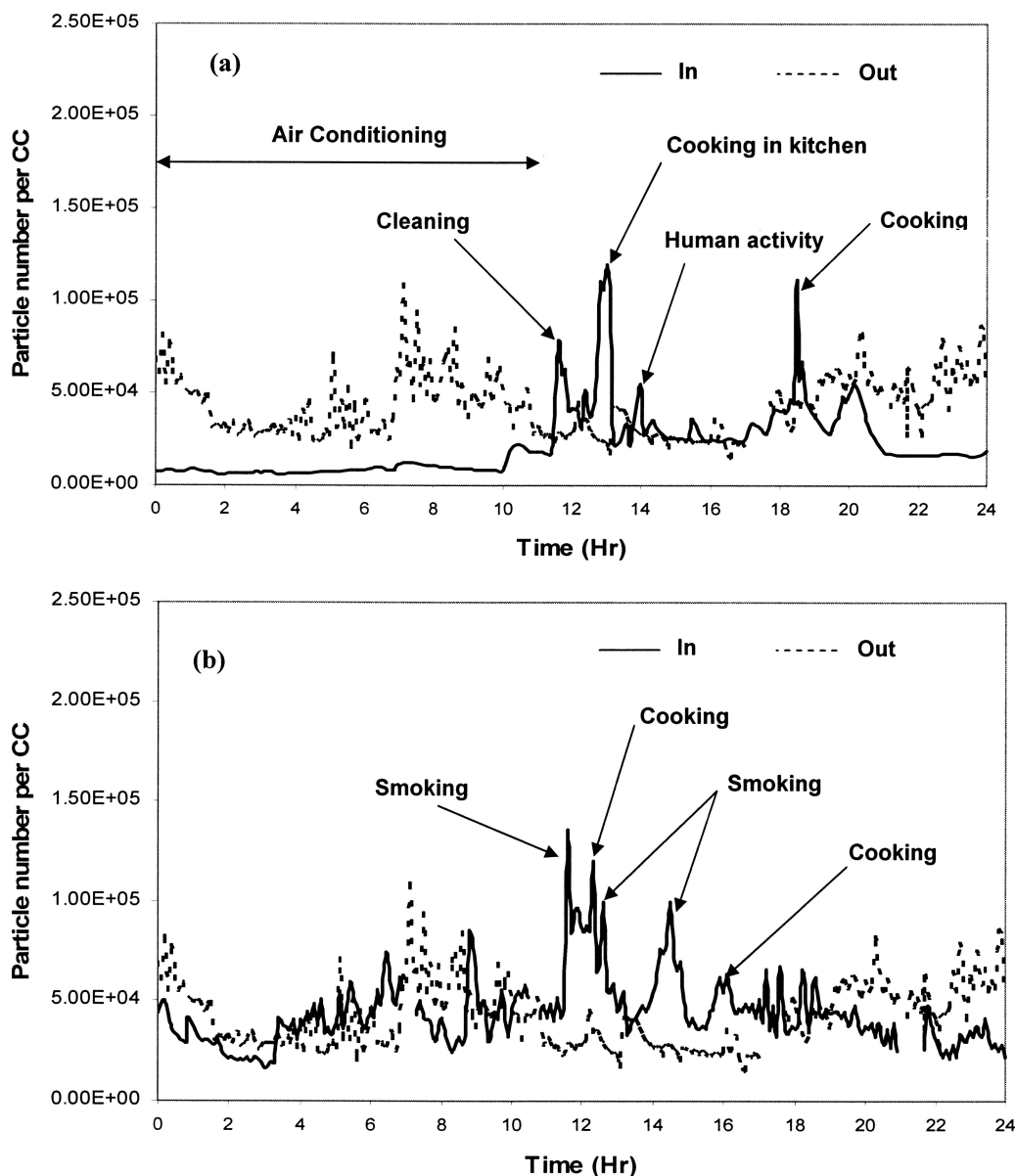


Figure 2. (a) 24-hr particle number concentration in the living room, May 12, 2004; and (b) 24-hr particle number concentration in the living room, May 19, 2004.

concentration, as summarized in Table 2. The lower particle concentration may be attributed to less intense indoor activities. Thus, the variability in I/O ratios is probably because of changes in the intensity of indoor activities on a day-to-day basis on some days than others. Dennekamp et al.⁴² reported similar findings that the ratios of indoor to outdoor approximation of $PM_{2.5}$ concentration for conditions of no indoor sources were close to 1 for most houses investigated in Australia, and the ratios were significantly higher than 1 in the presence of strong indoor sources.

Chemical Characteristics

Indoor aerosol particles are a complex mixture of water-soluble ions, metals, EC, and OC. To determine the relative proportions of the different chemical species in $PM_{2.5}$, their respective mass concentrations were determined, and the results are summarized in Table 3, along

with the mean I/O ratios. Li^+ , Ca^{2+} , and Mg^{2+} were below detection limits for all of the indoor and outdoor samples and, thus, were not reported in the table.

Water-Soluble Ions. Ions such as F^- , Cl^- , PO_4^{3-} , NO_2^- , and Na^+ were detected in trace amounts, as compared with those of SO_4^{2-} , NO_3^- , and NH_4^+ . The I/O ratios of NO_2^- , NO_3^- , SO_4^{2-} , and NH_4^+ are all >1 , whereas the ratios are <1 for all of the other components. The levels of both NO_2^- and NO_3^- were particularly elevated in the indoor environment. These secondary aerosols in the indoor environment were probably formed from the gaseous pollutants (the oxides of nitrogen) released from gas stoves and/or cooking⁴² through the process of gas-to-particle conversion.

The master room was observed to have the lowest I/O ratios for most of the ions as compared with the other two rooms. This is in line with the fact that the air conditioning was turned on in that room during the sampling time, thus,

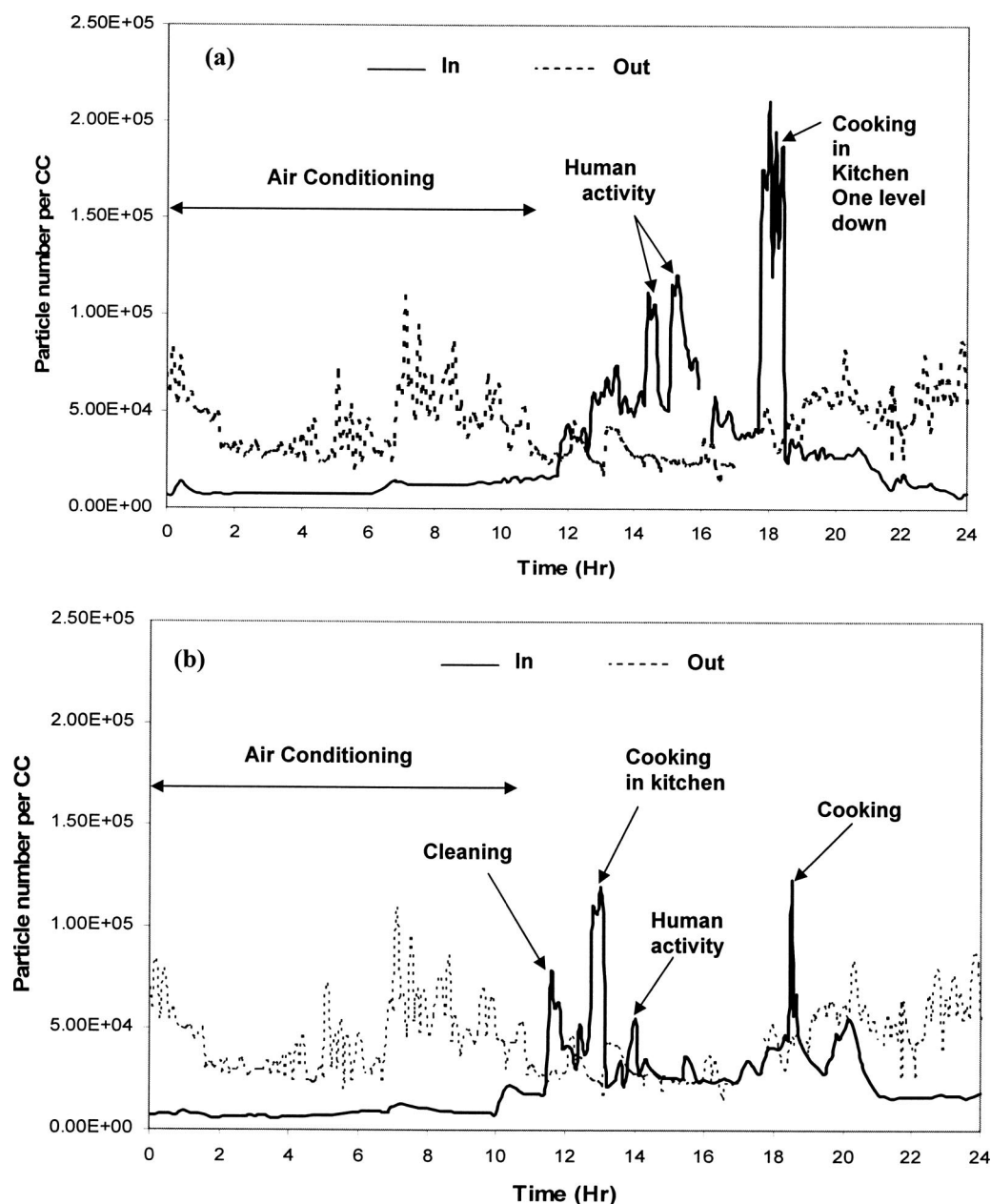


Figure 3. (a) 24-hr particle number concentration in the master bedroom, May 20, 2004; and (b) 24-hr particle number concentration in the master bedroom, May 21, 2004.

the penetration ability of outdoor particles was reduced. Although NO_3^- and NH_4^+ I/O ratios were highest in the bedroom, the I/O ratios of NO_2^- and SO_4^{2-} had their maximum values in the living room. There seems to be a strong association between the indoor and outdoor ion concentrations in this study for NO_3^- , NO_2^- , NH_4^+ , and SO_4^{2-} ions. These results suggest that a great proportion of outdoor particles that are transported into the home tend to accumulate within the home.⁴³ Apart from indoor sources, ventilation and ambient weather conditions are possible factors for the accumulation of these fine particles,⁴⁴ which should be further investigated to fully understand their impacts on the residence times of these particles.

Metals. In general, Cu, Ti, Al, Zn, and Fe were found to be the abundant trace elements both indoors and outdoors.

Whereas Ti, Al, and Fe predominantly originate from crustal materials,⁴⁵ Cu and Zn arise from fossil fuel combustion.⁴⁶ The I/O ratios of these elements are less than 1, suggesting that they originated mainly from the outdoors and that a significant fraction of their indoor concentrations could be attributed to infiltration of outdoor particles containing them. Similar conclusions can be drawn for Mn and Ni, which have lower concentrations indoors compared with outdoors. Both Mn and Ni are mainly derived from outdoor sources; for example, Mn can originate from methylcyclopentadienyl manganese tricarbonyl petroleum additive in automobiles,⁴⁷ whereas Ni is abundant in automobile dust.⁴⁸ The other metal levels in indoor air were higher than those outdoors, particularly Cr and Pb, with I/O ratios of 2.23 ± 0.8 and 1.73 ± 0.64 , respectively. This could be because of the contribution of indoor sources, in addition to

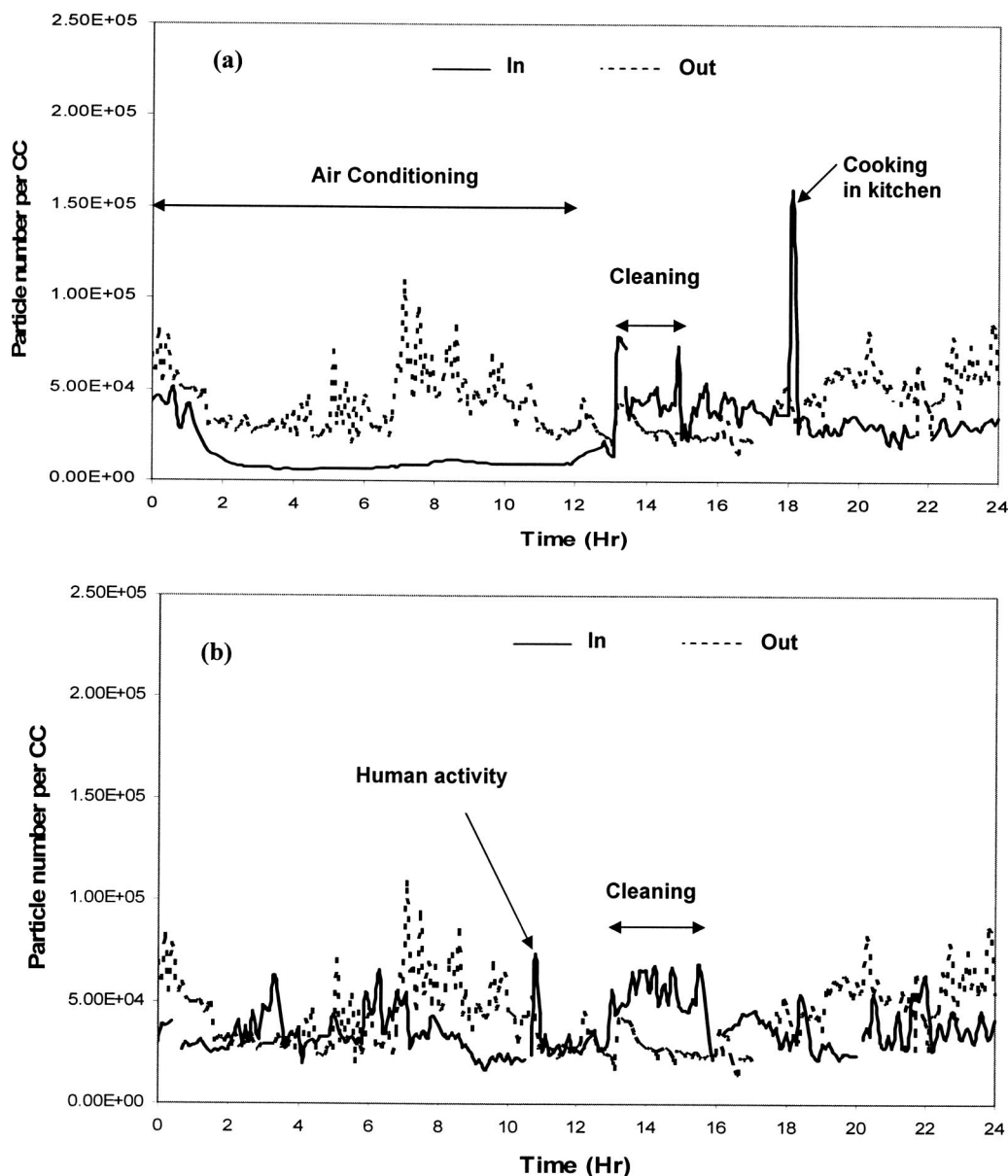


Figure 4. (a) 24-hr particle number concentration in the bedroom, May 22, 2004; (b) 24-hr particle number concentration in the bedroom, May 23, 2004.

outdoor sources, because of their accumulation indoors, or because of the limitation of the study design, discussed in a later section. For example, Cd, Cr, Ni, and Pb are reported in environmental tobacco smoke (ETS).⁴⁹ Wu et al.⁵⁰ reported that Cd was a marker for ETS in the presence of low background concentrations ($<1.5 \mu\text{g}/\text{m}^3$). Cd, Ni, and Pb had the maximum concentration in the living room in which the air quality was influenced by ETS as shown by the particle number concentration profiles in Figure 2. Cr may have additional sources within the house and possibly specific to the microenvironments in question.⁴⁸ These metals could have been released from the metallic cooking utensils because of a combination of corrosion, wear, and abrasion⁵¹ or emitted from the food during cooking.

EC and OC. Indoor EC concentrations are generally lower than those outdoors, with average I/O ratios ranging from

0.75 to 0.96. This finding is fairly consistent with results reported by Jones et al.⁵² and Geller et al.,⁵³ who found that EC has its primary sources outdoors, mainly vehicular emissions. EC (soot) is a byproduct of incomplete combustion emitted in exhaust from road traffic.⁵⁴ The I/O ratios for EC concentrations suggest a high penetration rate of the EC particles, because most of the EC is expected to originate outdoors.

Conversely, indoor OC concentrations were found to be relatively higher than those outdoors, with the I/O ratios within the range of 1.04 to 1.92 in this study. The higher indoor concentrations are because of the contribution of indoor sources, such as cooking⁵⁵ and smoking.⁵⁶ The highest I/O ratio of 1.92 was found to be in the living room, because cooking is the predominant activity that influences the air quality in the living room as compared with the other two rooms. Conversely, the lowest I/O

Table 2. Mass concentrations of PM_{2.5} on different days and I/O ratios.

Sampling Site	Sampling Day in May 2004	PM _{2.5} (μg/m ³)	I/O Ratio
Living room	12	41.1	1.77
Living room	19	44.0	1.90
Master bedroom	20	23.2	1.00
Master bedroom	21	21.5	0.93
Bedroom	22	25.7	1.11
Bedroom	23	36.3	1.56
Outdoor	21–22 ^a	23.2	NA ^b

Notes: ^a24-hr average mass concentration of particles collected over 2 consecutive days; ^bNA = not applicable.

ratio of 1.04 was found to be in the bedroom, because the bedroom has the lowest occurrence of indoor OC sources as compared with the living room. These observations suggest that, in addition to the infiltration of a substantial fraction of the particle-bound OC from outdoors, indoor sources, such as cooking and possibly smoking, make a considerable contribution to the overall OC in PM_{2.5}. These findings are consistent with those reported by Abt et al.⁵⁷ and Geller et al.⁵³ in that the I/O ratios for OC are considerably higher than 1 in the presence of strong indoor combustion sources.

Chemical Mass Balance. Figure 5 shows the chemical composition of the indoor fine particles collected from the three rooms, respectively. The percentages were found from average values of the component mass concentrations that made up the total PM collected. The major

components of air within the home are carbonaceous matter (EC and OC), SO₄²⁻, and NO₃⁻. Although appropriate conversion factors are normally used to convert OC concentrations to organic matter for outdoor aerosols based on the detailed knowledge of the organic chemical speciation of PM, this is not done so here, because this information is not currently available for indoor aerosols. Although the OC estimated contributed ≤30% of the mass concentration of particles, EC formed ≤19%, and SO₄²⁻ accounted for ≤17% of the mass. The total mass concentration of all of the trace elements in fine particles was calculated by assuming that they exist as oxides and multiplying the concentrations of the analyzed elements with corresponding factors to account for the oxygen mass.⁵⁸ The mass of aerosols that could not be accounted for through chemical measurements is shown in the figures as “others,” which also includes the measurement uncertainty.

CONCLUSIONS

This paper presents a case study on the physical and chemical characteristics of indoor aerosols to gain a better understanding of the potential indoor and outdoor sources affecting the indoor air quality of a typical residential home in Singapore. I/O ratios for mass concentrations of particles were generally >1 for all of the sampling sites, demonstrating the influence of indoor sources. From the time-activity logs maintained by the occupants, the main indoor sources that affected the indoor air quality included tobacco smoking, cooking, and, to a lesser extent, household cleaning and general human activities,

Table 3. Mass concentrations of chemical speciation and I/O ratios.

Species	Mass Concentration				I/O Ratio		
	Living Room	Master Bedroom	Bedroom	Outdoor	Living Room	Master Bedroom	Bedroom
F ⁻ (μg/m ³)	0.29	0.42	0.51	BDL	NA	NA	NA
Cl ⁻ (μg/m ³)	0.38	0.29	0.37	0.84	0.45	0.35	0.44
NO ₂ ⁻ (μg/m ³)	0.09	0.08	0.06	0.02	4.13	3.63	2.50
NO ₃ ⁻ (μg/m ³)	3.78	1.15	4.42	0.82	4.63	1.41	5.41
SO ₄ ²⁻ (μg/m ³)	5.48	3.91	5.24	3.22	1.70	1.21	1.63
PO ₄ ³⁻ (μg/m ³)	BDL	BDL	BDL	0.05	BDL	BDL	BDL
Na ⁺ (μg/m ³)	0.53	0.31	0.46	0.72	0.74	0.43	0.63
NH ₄ ⁺ (μg/m ³)	1.51	1.07	1.79	0.75	2.01	1.43	2.39
K ⁺ (μg/m ³)	BDL	BDL	BDL	0.73	BDL	BDL	BDL
EC (μg/m ³)	4.09	4.30	3.34	4.47	0.92	0.96	0.75
OC (μg/m ³)	10.47	6.64	5.66	5.45	1.92	1.22	1.04
Al (ng/m ³)	47.3	38.2	40.6	83.0	0.57	0.46	0.49
Co (ng/m ³)	0.10	0.07	0.07	0.08	1.32	0.90	0.86
Cr (ng/m ³)	0.79	1.44	1.71	0.59	1.35	2.43	2.90
Cu (ng/m ³)	57.9	69.3	50.4	102	0.57	0.69	0.50
Fe (ng/m ³)	16.9	23.8	24.1	56.3	0.30	0.42	0.42
Mn (ng/m ³)	0.87	0.71	0.47	2.07	0.42	0.35	0.23
Pb (ng/m ³)	2.88	5.34	0.87	2.17	1.33	2.47	0.40
Zn (ng/m ³)	41.7	43.7	31.8	54.2	0.77	0.81	0.59
Cd (ng/m ³)	0.70	0.60	0.03	0.40	1.75	1.50	0.74
Ni (ng/m ³)	1.34	0.72	0.75	1.02	1.32	0.71	0.74
Ti (ng/m ³)	57.5	58.3	61.7	78.8	0.73	0.74	0.78
V (ng/m ³)	1.60	1.53	1.58	1.52	1.05	1.01	1.04

Notes: BDL = below detection limit; NA = not applicable.

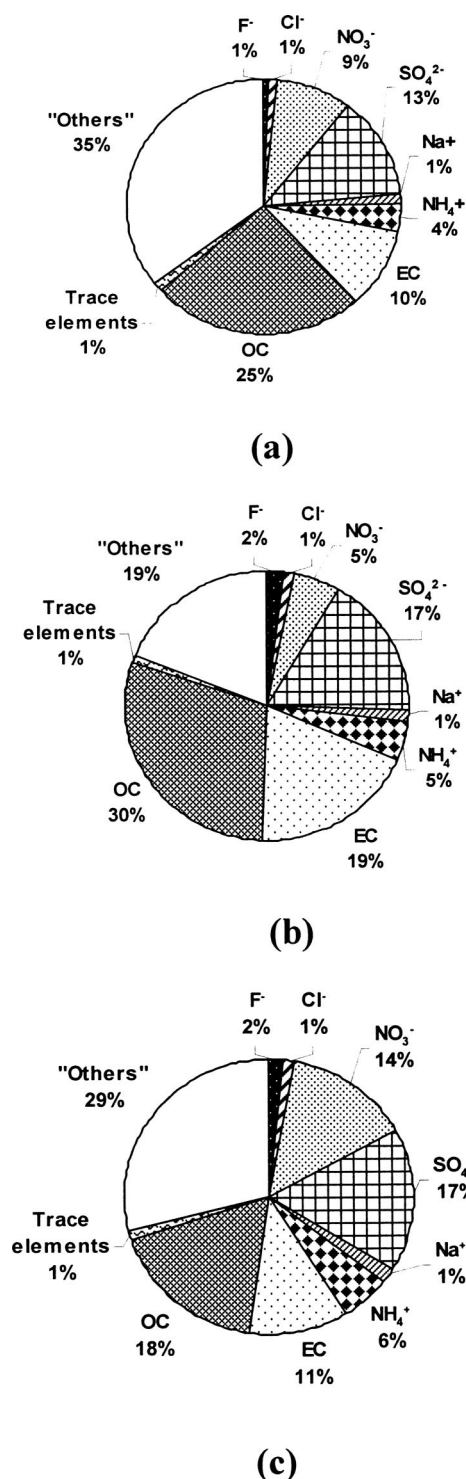


Figure 5. Chemical composition of indoor aerosols in living room (average $PM_{2.5} = 42.5 \mu\text{g}/\text{m}^3$; a), master bedroom (average $PM_{2.5} = 22.4 \mu\text{g}/\text{m}^3$; b), and bedroom (average $PM_{2.5} = 31 \mu\text{g}/\text{m}^3$; c).

such as movement within the indoor environment. However, the chemical composition of particles emitted from indoor and outdoor sources is not similar. The I/O ratios were estimated, which suggest that certain chemical constituents of indoor particles, such as Cl^- , Na^+ , Al, Co, Cu, Fe, Mn, Ti, V, Zn, and EC, were derived through migration of outdoor particles ($\text{I/O} < 1$ or ~ 1), whereas the levels of others, such as NO_2^- , NO_3^- , SO_4^{2-} , NH_4^+ , Cd, Cr, Ni, Pb,

and OC, were largely influenced by the presence of specific indoor sources ($\text{I/O} > 1$). Their specific sources are discussed.

ACKNOWLEDGMENTS

This study was funded by the National University of Singapore Academic Research Fund through grant RP-279-000-142-112. The authors thank Siao Wei See and S. Venkatesa Perumal for help with the paper.

REFERENCES

- Pope, C.A.; Dockery, D.W.; Schwartz, J. Review of Epidemiological Evidence of Health-Effects of Particulate Air-Pollution; *Inhal. Toxicol.* **1995**, *7*, 1-18.
- Brunekreef, B.; Holgate, S.T. Air Pollution and Health; *Lancet* **2002**, *360*, 1233-1242.
- Brauer, M.; Brumm, J.; Vedal, S.; Petkau, A.J. Exposure Misclassification and Threshold Concentrations in Time Series Analyses of Air Pollution Health Effects; *Risk Anal.* **2002**, *22*, 1183-1193.
- U.S. Environmental Protection Agency. *Air Quality Criteria for Particulate Matter, Volumes I-III*; EPA/600/P-95/001; U.S. Environmental Protection Agency: Washington, DC, 1996.
- Kamens, R.; Lee, C.T.; Wiener, R.; Leith, D. A Study to Characterize Indoor Particles in 3 Nonsmoking Homes; *Atmos. Environ.* **1991**, *25*, 939-948.
- Mukerjee, S.; Ellenson, W.D.; Lewis, R.G.; Stevens, R.K.; Somerville, M.C.; Shadwick, D.S.; Willis, R.D. An Environmental Scoping Study in the Lower Rio Grande Valley of Texas. 3. Residential Microenvironmental Monitoring for Air, House Dust, and Soil; *Environ. Int.* **1997**, *23*, 657-673.
- Wallace, L.; Howard-Reed, C. Continuous Monitoring of Ultrafine, Fine, and Coarse Particles in a Residence for 18 Months in 1999-2000; *J. Air & Waste Manage. Assoc.* **2002**, *52*, 828-844.
- Wiggzell, E.; Kendall, M.; Nieuwenhuijsen, M.J. The Spatial and Temporal Variation of Particulate Matter within the Home; *J. Expo. Anal. Environ. Epidemiol.* **2000**, *10*, 307-314.
- Fromme, H.; Lahrz, T.; Hainsch, A.; Oddoy, A.; Piloty, M.; Ruden, H. Elemental Carbon and Respirable Particulate Matter in the Indoor Air of Apartments and Nursery Schools and Ambient Air in Berlin (Germany); *Indoor Air* **2005**, *15*, 335-341.
- Hussein, T.; Hameri, K.; Heikkinen, M.S.A.; Kulmala, M. Indoor and Outdoor Particle Size Characterization at a Family House in Espoo-Finland; *Atmos. Environ.* **2005**, *39*, 3697-3709.
- He, C.R.; Morawska, L.; Hitchins, J.; Gilbert, D. Contribution from Indoor Sources to Particle Number and Mass Concentrations in Residential Houses; *Atmos. Environ.* **2004**, *38*, 3405-3415.
- Morawska, L.; He, C.; Hitchins, J.; Mengersen, K.; Gilbert, D. Characteristics of Particle Number and Mass Concentrations in Residential Houses in Brisbane, Australia; *Atmos. Environ.* **2003**, *37*, 4195-4203.
- Li, C.S. Elemental Composition of Residential Indoor PM_{10} in the Urban Atmosphere of Taipei; *Atmos. Environ.* **1994**, *28*, 3139-3144.
- Nitta, H.; Ichikawa, M.; Sato, M.; Konishi, S.; Ono, M. A New Approach Based on a Covariance Structure Model to Source Apportionment of Indoor Fine Particles in Tokyo; *Atmos. Environ.* **1994**, *28*, 631-636.
- Tung, T.C.W.; Chao, C.Y.H.; Burnett, J.; Pang, S.W.; Lee, R.Y.M. A Territory Wide Survey on Indoor Particulate Level in Hong Kong; *Build. Environ.* **1999**, *34*, 213-220.
- Chao, C.Y. Wong, K.K. Residential Indoor PM_{10} and $PM_{2.5}$ in Hong Kong and the Elemental Composition; *Atmos. Environ.* **2002**, *36*, 265-277.
- Lee, S.C.; Guo, H.; Li, W.M.; Chan, L.Y. Inter-Comparison of Air Pollutant Concentrations in Different Indoor Environments in Hong Kong; *Atmos. Environ.* **2002**, *36*, 1929-1940.
- Wallace, L. Indoor Particles: a Review; *J. Air & Waste Manage. Assoc.* **1996**, *46*, 98-126.
- Burnett, R.T.; Smith-Doiron, M.; Stieb, D.; Cakmak, S.; Brook, J.R. Effects of Particulate and Gaseous Air Pollution on Cardiorespiratory Hospitalizations; *Arch. Environ. Health* **1999**, *54*, 130-139.
- Goldberg, M.S.; Burnett, R.T.; Bailar, J.C.; Brook, J.; Bonvalot, Y.; Tamblin, R.; Singh, R.; Valois, M.F.; Vincent, R. The Association between Daily Mortality and Ambient Air Particle Pollution in Montreal, Quebec 2. Cause-Specific Mortality; *Environ. Res.* **2001**, *86*, 26-36.
- James, A.C.; Stahlhofen, W.; Rudolf, G.; Egan, M.J.; Nixon, W.; Gehr, P.; Briant, J.K. The Respiratory-Tract Deposition Model Proposed by the ICRP Task Group; *Radiat. Prot. Dosim.* **1991**, *38*, 159-165.
- Kreyling, W.G.; Semmler, M.; Erbe, F.; Mayer, P.; Takenaka, S.; Schulz, H.; Oberdorster, G.; Ziesenis, A. Translocation of Ultrafine Insoluble Iridium Particles from Lung Epithelium to Extrapulmonary Organs Is Size Dependent but Very Low; *J. Toxicol. Environ. Health* **2002**, *65*, 1513-1530.

23. Kiss, G.; Varga-Puchony, Z.; Rohrbacher, G.; Hlavey, J. Distribution of Polycyclic Aromatic Hydrocarbons on Atmospheric Aerosol Particles of Different Sizes; *Atmos. Res.* **1998**, *46*, 253-261.
24. Matson, U. Indoor and Outdoor Concentrations of Ultrafine Particles in Some Scandinavian Rural and Urban Areas; *Sci. Total Environ.* **2005**, *343*, 169-176.
25. Wallace, L. Indoor Sources of Ultrafine and Accumulation Mode Particles: Size Distributions, Size-Resolved Concentrations, and Source Strengths; *Aerosol Sci. Technol.* **2006**, *40*, 348-360.
26. Lighty, J.S.; Veranth, J.M.; Sarofim, A.F. Combustion Aerosols: Factors Governing Their Size and Composition and Implications to Human Health; *J. Air & Waste Manage. Assoc.* **2000**, *50*, 1565-1618.
27. Morawska, L.; Zhang, J.F. Combustion Sources of Particles. 1. Health Relevance and Source Signatures; *Chemosphere* **2002**, *49*, 1045-1058.
28. Monn, Ch.; Fuchs, A.; Kogelschatz, D.; Wanner, H.U. Comparison of Indoor and Outdoor Concentrations of PM-10 and PM-2.5; *J. Aerosol Sci.* **1995**, *26*, S515-S516.
29. Tucker, W.G. An Overview of PM_{2.5} Sources and Control Strategies; *Fuel Process. Technol.* **2000**, *65*, 379-392.
30. Koistinen, K.J.; Edwards, R.D.; Mathys, P.; Ruuskanen, J.; Kunzli, N.; Jantunen, M.J. Sources of Fine Particulate Matter in Personal Exposures and Residential Indoor, Residential Outdoor and Workplace Microenvironments in the Helsinki Phase of the EXPOLIS Study; *Scand. J. Work Environ. Health* **2004**, *30*, 36-46.
31. Thatcher, T.L.; Layton, D.W. Deposition, Resuspension, and Penetration of Particles within a Residence; *Atmos. Environ.* **1995**, *29*, 1487-1497.
32. Sarnat, S.E.; Coull, B.A.; Ruiz, P.A.; Koutrakis, P.; Suh, H.H. The Influences of Ambient Particle Composition and Size on Particle Infiltration in Los Angeles, CA, Residences; *J. Air & Waste Manage. Assoc.* **2006**, *56*, 186-196.
33. See, S.W.; Balasubramanian, R. Risk Assessment of Exposure to Indoor Aerosols Associated with Chinese Cooking; *Environ. Res.* **2006**, *102*, 197-204.
34. Karthikeyan, S.; Balasubramanian, R. Determination of Water-Soluble Inorganic and Organic Species in Atmospheric Fine Particulate Matter; *Microchem. J.* **2006**, *82*, 49-55.
35. Karthikeyan, S.; Joshi, U.M.; Balasubramanian, R. Microwave Assisted Sample Preparation for Determining Water-Soluble Fraction of Trace Elements in Urban Airborne Particulate Matter: Evaluation of Bioavailability; *Anal. Chim. Acta* **2006**, *576*, 9-16.
36. See, S.W.; Balasubramanian, R.; Wang, W. A Study of the Physical, Chemical and Optical Properties of Ambient Aerosol Particles in Southeast Asia during Hazy and Non-Hazy Days; *J. Geophys. Res.* **2006**, *111*, D10508.
37. See, S.W.; Balasubramanian, R. Emissions of Ultrafine Particles from Different Gas Cooking Methods; *Aerosol Air Qual. Res.* **2006**, *6*, 82-92.
38. See, S.W.; Karthikeyan, S.; Balasubramanian, R. Health Risk Assessment of Occupational Exposure to Particulate-Phase Polycyclic Aromatic Hydrocarbons Associated with Chinese, Malay, and Indian Cooking; *J. Environ. Monit.* **2006**, *8*, 369-376.
39. Wallace, L.; Williams, R. Use of Personal-Indoor-Outdoor Sulfur Concentrations to Estimate the Infiltration Factor and Outdoor Exposure Factor for Individual Homes and Persons; *Environ. Sci. Technol.* **2005**, *39*, 1707-1714.
40. Chao, C.Y.H.; Tung, T.C.W.; Burnett, J. Influence of Different Indoor Activities on the Indoor Particulate Levels in Residential Buildings; *Indoor Built Environ.* **1998**, *7*, 110-121.
41. Morawska, L.; He, C.R.; Hitchins, J.; Gilbert, D.; Parappukkaran, S. The Relationship between Indoor and Outdoor Airborne Particles in the Residential Environment; *Atmos. Environ.* **2001**, *35*, 3463-3473.
42. Dennekamp, M.; Howarth, S.; Dick, C.A.J.; Cherrie, J.W.; Donaldson, K.; Seaton, A. Ultrafine Particles and Nitrogen Oxides Generated by Gas and Electric Cooking; *Occup. Environ. Med.* **2001**, *58*, 511-516.
43. Adams, S.J.; Kibrya, R.; Brimblecombe, P. A Particle Accumulation Study during the Reconstruction of the Great Court, British Museum; *J. Cultural Heritage* **2002**, *3*, 283-287.
44. Nazaroff, W.W. Indoor Particle Dynamics; *Indoor Air* **2004**, *14*, 175-183.
45. Silva, P.J.; Carlin, R.A.; Prather, K.A. Single Particle Analysis of Suspended Soil Dust from Southern California; *Atmos. Environ.* **2000**, *34*, 1811-1820.
46. Lopez, J.M.; Callen, M.S.; Murillo, R.; Garcia, T.; Navarro, M.V.; de la Cruz, M.T.; Mastral, A.M. Levels of Selected Metals in Ambient Air PM10 in an Urban Site of Zaragoza (Spain); *Environ. Res.* **2005**, *99*, 58-67.
47. Pellizzari, E.D.; Clayton, C.A.; Rodes, C.E.; Mason, R.E.; Piper, L.L.; Fort, B.; Pfeifer, G.; Lynam, D. Particulate Matter and Manganese Exposures in Toronto, Canada; *Atmos. Environ.* **1999**, *33*, 721-734.
48. Madany, I.M.; Akhter, M.S.; Jowder, O.A.A. The Correlations between Heavy-Metals in Residential Indoor Dust and Outdoor Street Dust in Bahrain; *Environ. Int.* **1994**, *20*, 483-492.
49. Smith, C.J.; Lingington, D.; Doolittle, D.J. An International Literature Survey of "IARC Group I Carcinogens" Reported in Mainstream Cigarette Smoke; *Food Chem. Toxicol.* **1997**, *35*, 1107-1130.
50. Wu, D.; Landsberger, S.; Larson, S.M. Evaluation of Elemental Cadmium as a Marker for Environmental Tobacco-Smoke; *Environ. Sci. Technol.* **1995**, *29*, 2311-2316.
51. Jellesen, M.S.; Rasmussen, A.A.; Hilbert, L.R. A Review of Meal Release in the Food Industry; *Mater. Corros.* **2006**, *57*, 387-393.
52. Jones, N.C.; Thornton, C.A.; Mark, D.; Harrison, R.M. Indoor/Outdoor Relationships of Particulate Matter in Domestic Homes with Roadside, Urban and Rural Locations; *Atmos. Environ.* **2000**, *34*, 2603-2612.
53. Geller, M.D.; Chang, M.H.; Sioutas, C.; Ostro, B.D.; Lipsett, M.J. Indoor/Outdoor Relationship and Chemical Composition of Fine and Coarse Particles in the Southern California Deserts; *Atmos. Environ.* **2002**, *36*, 1099-1110.
54. *Urban Air Quality in the United Kingdom*. Quality of Urban Air Review Group, Department of Environment: London, U.K., 1993.
55. McDonald, J.D.; Zielinska, B.; Fujita, E.M.; Sagebiel, J.C.; Chow, J.C.; Watson, J.G. Emissions from Charbroiling and Grilling of Chicken and Beef. *J. Air & Waste Manage. Assoc.* **2003**, *53*, 185-194.
56. Smith, T.J.; Davis, M.E.; Reaser, P.; Natkin, J.; Hart, J.E.; Laden, F.; Heff, A.; Garshick, E. Overview of Particulate Exposures in the US Trucking Industry; *J. Environ. Monit.* **2006**, *8*, 711-720.
57. Abt, E.; Suh, H.H.; Catalano, P.; Koutrakis, P. Relative Contribution of Outdoor and Indoor Particle Sources to Indoor Concentrations; *Environ. Sci. Technol.* **2000**, *34*, 3579-3587.
58. Chow, J.C.; Watson, J.G.; Fujita, E.M.; Lu, Z.Q.; Lawson, D.R.; Ashbaugh, L.L. Temporal and Spatial Variations of PM_{2.5} and PM₁₀ Aerosol in the Southern California Air Quality Study; *Atmos. Environ.* **1994**, *28*, 2061-2080.

About the Authors

Rajasekhar Balasubramanian is an associate professor at the Division of Environmental Science and Engineering Program, National University of Singapore. Sheng Sheng Lee is an undergraduate student at the Department of Chemical and Biomolecular Engineering, National University of Singapore. Please address correspondence to: Rajasekhar Balasubramanian, Division of Environmental Science and Engineering, Block EA, #03-12 9 Engineering Dr. 1, National University of Singapore, Singapore 117576; phone: +65-6516-5135; fax: +65-6779-1936; e-mail: eserbala@nus.edu.sg.