The Relationship between Indoor and Outdoor Airborne Particles in the Residential Environment

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Abstract - The relationship between indoor and outdoor airborne particles was investigated for sixteen residential houses located in a suburban area of Brisbane, Australia. The submicrometer particle numbers were measured using the Scanning Mobility Particle Sizer (SMPS), the larger particle numbers using the Aerodynamic Particle Sizer (APS) and an approximation of $PM_{2.5}$ was also measured using a DustTrak. The measurements were conducted for normal and minimum ventilation conditions using simultaneous and non-simultaneous measurement methods designed for the purpose of the study.

Comparison of the ratios of indoor to outdoor particle concentrations revealed that while temporary values of the ratio vary in a broad range from 0.2 to 2.5 for both lower and higher ventilation conditions, average values of the ratios were very close to one regardless of ventilation conditions and of particle size range. The ratios were in the range from 0.78 to 1.07 for submicrometer particles, from 0.95 to 1.0 for supermicrometer particles and from 1.01 to 1.08 for $PM_{2.5}$ fraction.

Comparison of the time series of indoor to outdoor particle concentrations shows a clear positive relationship existing for many houses under normal ventilation conditions (estimated to be about and above 2 h^{-1}), but not under minimum ventilation conditions (estimated to be about and below 1 h^{-1}). These results suggest that for normal ventilation conditions, outdoor particle concentrations could be used to predict instantaneous indoor particle concentrations but not for minium ventilation, unless air exchange rate is known, thus allowing for estimation of the "delay constant".

INTRODUCTION

Assessment of the risk to the community resulting from exposure to airborne pollutants should ideally include measurements of concentration levels of the pollutants in all microenvironments where people spend their time. Due to the multiplicity of different microenvironments, it is usually however, not possible to conduct measurements in all of them. The main consideration in designing exposure assessment studies is, which of the microenvironments should be studied to provide data allowing for most accurate assessments, while limiting the costs and efforts

relating to the studies. In many cases the subdivision is between the indoor and outdoor environment, with questions posed as to what extent indoor exposures could be predicted from measured concentrations of pollutants in outdoor air.

When considering human exposures to airborne pollutants, of particular importance is the exposure to airborne particles, and specifically to its finer fractions, classified as ultra fine particles (often defined as smaller than 0.1 µm), submicrometer particles (smaller than 1 µm) or $PM_{2.5}$ fraction (mass concentration of particles with aerodynamic diameter smaller that 2.5 µm). Smaller particles have a higher probability of penetration into the deeper parts of the respiratory tract (James *et al*, 1991; Owen and Ensor 1992, Berico *et al*., 1997) and also contain higher levels of trace elements and toxins, such as the polycyclic aromatic hydrocarbons and mutagens (Ando *et al*., 1996; Kiss *et al*., 1998).

It is also important to note that in the air smaller and larger particles behave differently, and in particular penetration of particles of different sizes through the building envelope is different.

The early studies on the relationship between indoor and outdoor particles conducted in the 1950s and summarised by Andersen (1972), showed that the ratio of indoor and outdoor total suspended particle matter varied from 0.20 to about 1.00. Benson *et al*. (1972) concluded in their review that, in general, the ratios of indoor and outdoor particle concentration are about one.

Since then a number of studies on the relationship between indoor and outdoor particles have been conducted (Spengler *et al*., 1981; Quackenboss *et al*., 1989; Wallace, 1996). The results of these studies indicate that the ratio of the indoor to outdoor particle mass concentrations varies in a wide range from 0.5 to 2 in the

absence of indoor particle sources, and that indoor activities such as smoking or cooking may play an important role in affecting the relationship (Spengler *et al*., 1981; Monn *et al*., 1995; Ross *et al*., 1999). While the focus of most of the studies reported have been on particle mass, there is still very little information available on the relationship between the numbers of particles in indoor and outdoor air. However, recent studies have indicated that particle number concentration could be a better indicator of health risk than particle mass (Oberdöster, *et al*., 1992; Oberdöster, 1995).

Theoretically, the indoor particle concentration is a function of a number of factors, the most important of which are the generation rate of particles indoors, the outdoor particle concentration, air exchange rate, particle penetration efficiency from the outdoor to the indoor environment, and the particle deposition rate on indoor surfaces (Shair and Heitner 1974; Kamens *et al*., 1991; Thatcher and Layton 1995). However, in practice, it is usually very difficult to assess the exposure due to the lack of data and information on the correlation between indoor and outdoor particles, which are house and environment specific.

Understanding the relationship of indoor and outdoor aerosol particles under different environmental conditions is of importance for improving exposure estimates and in turn for developing efficient control strategies to reduce human exposure and thus heath risk. Current exposure assessment models are often based on the outdoor pollutant concentration used as the input parameter for predicting total human exposure (Colls and Micallef, 1997). However, as discussed above, the indoor concentrations may be different than the outdoor ones even in the absence of any significant indoor pollution sources.

To address some of the deficiencies in the understanding of the relationship between indoor and outdoor particles, this study was undertaken with the following objectives: (1) to investigate experimentally the relationship between indoor and outdoor particle number distribution and concentration in the size range from 0.015 to 20 μm, as well as approximation of $PM_{2.5}$ concentrations in 16 residential houses located in a suburban area, for cases when there were no indoor activities conducted in the houses that would result in particle generation (such as cooking, smoking, dusting and vacuuming), (2) to analyse the relationship between, and trends of, indoor and outdoor particles for different ventilation conditions in the investigated houses, (3) to conclude on the applicability of outdoor particle concentration data for indoor exposure assessment.

2. EXPERIMENTAL METHOD

2.1. *The Sampling Site and House*

A residential suburb of Tingalpa, located on the eastern side of Brisbane was chosen as the measurement site. The distance from Tingalpa to the city centre is about 10 km. The site is relatively flat and represents a mix of house types, both in terms of age and design, including newer and older houses, made of brick and timber, as well as high and low set. High set means that the house is elevated above the ground on timber or brick stumps, while a low set house is built on ground level. Fourteen houses were chosen for the study from this suburb and additional two houses were chosen from other suburbs to provide a comparison. One of the two houses (House1), is also located east of the city but close to the ocean, and the second (House19) is located north of the city. The houses investigated in this study differed in age (2 ~ 100 years), construction material (timber, brick) and design with some being elevated above ground on stumps, and some set up on the ground. House design and material

characteristics have an effect the air exchange rate, but as this parameter is not included in the analyses presented in this paper, detailed house characteristics are not provided. The occupants of the houses were non-smokers, with the exception of an occupant of House 17 who, however, never smoked indoors. Only one house (House 13) was fitted with an air conditioning system, which was, however, not operating during the measurement conducted in this house.

The suburb of Tingalpa is located approximately 5 km from the Brisbane Airport site of the Bureau of Meteorology. It was considered that the meteorological conditions recorded at the Airport site would by representative of the conditions at the sampling site. The Bureau of Meteorology provided the data for the duration of the sampling period.

2.2. *Instrumentation*

Measurements of submicrometer particle number concentration and size distribution in the range from 0.015 – 0.685 μm were conducted using a TSI Model 3934 Scanning Mobility Particle Sizer (SMPS) (TSI Incorporated, St. Paul, MN, USA). The SMPS consists of an Electrostatic Classifier (EC) that size classifies the particles according to their ability to traverse an electrical field, and of a TSI Model 3022A Condensation Particle Counter (CPC) that counts the particles. During some of the measurements, particle size distribution was not measured, only the total number concentration using the CPC that measured in a similar size range to the SMPS.

Size distribution and concentartion of larger particles were measured using the TSI Model 3320 Aerodynamic Particle Sizer (APS). The APS measures particle size distribution in almost real time, and was set up for this study to measure in the size range from 0.54 to 19.81 μm.

The TSI Model 8520 DustTrak aerosol monitor (TSI Incorporated, St. Paul, MN, USA) was used to measure approximation of $PM_{2.5}$ concentrations. The instrument is a real time device that operates based on a light scattering technique where the amount of scattered light is proportional to the volume concentration of the aerosol. The $PM_{2.5}$ values obtained in this study using the DustTrak, are not actual gravimetric values, as the instrument was not calibrated for the specific aerosol studied, and would need to be re-calibrated for the ambient indoor and outdoor type aerosol. It was used in this study to provide relative readings.

2.3. *Design of sampling system for indoor/outdoor measurement*

As there was only one set of instrumentation available, an experimental design consisting of two different methods was developed to provide both indoor and outdoor measurements. One was called a non-simultaneous method, in which the SMPS or CPC and APS were used, and was based on conducting five to ten of outdoor measurements first, then shifting the instrumentation indoors and conducting five to ten of indoor measurements. All the measurements were conducted in the morning between about (9:00 – 12:00), except for two cases, when the measurements were conducted in the afternoon (13:30 – 14:00). This procedure was repeated during the course of each measuring run.

The second method used was called simultaneous and it employed an automatic indoor/outdoor sampling system, in which the CPC, APS and DustTrak were used. The sampling system allowed for sampling from indoor and outdoor air, switching from one to the other within a few seconds. The outdoor air was continuously pumped through a closed tubing system to the indoors and then out again. A valve installed in the system could switch between sampling from the tube (outdoor air), or directly from the indoor air. After the valve switched, there was a time delay of a few seconds

before the air from the sampled environment reached the instruments, which was the time the air travelled from the valve to the instruments. The system switched every 60-second between the indoor and outdoor measurements and six samples were taken during the 60-second. In order to avoid the possibility of mixing of the outdoor and indoor air streams, the samples taken at the beginning and the end of each 60-second period were disregarded, leaving the four intermediate samples to be averaged.

To decrease the loss of particles in the tube bringing outdoor air to the valve, a pipe of a diameter of 32 mm and length 5 m was used. A stainless steel 100 mm long sampling tube of 11 mm in diameter, with a bend of 135 ° drawing air to the instruments was inserted into the larger pipe. The flowrates in the whole system were set up such that the sampling conditions from the larger pipe were isokinetic which is a necessary condition for representative sampling of particles larger than about 4 μm (it is not necessary for smaller particles).

Automatic switching from indoor to outdoor measurements was required as manual switching would be too time consuming and inaccurate. A timer device was constructed which would switch a relay on and off at adjustable time intervals. An alternating current power supply was used for the timing circuit, where the time interval was obtained from the frequency of main's power. The timer accuracy was tested and found to be exact over a period of up to 1.5 hours.

It is known that particle deposition occurs in tubing used for sampling, and a test was done to determine the percentage loss of particles in the 5 m long pipe, used to bring the outside air into the sampling system. In this experiment, air from the laboratory was used and it was assumed that the characteristics of the particles present in the air were representative of the ambient air in a residential house with no indoor sources. The sampling system was set up to sample the laboratory air either directly or through

the pipe, at one-minute intervals for a period of five hours, and the percentage loss was calculated from the ratios of the concentrations measured and averaged over the time of the experiment.

The results show that 10.9% (SD 6.4%) of the particles in the CPC range are lost to the tube, 21.0% (SD 5.3%) of particles in the APS range, and that 0.5% (SD 2.9%) of the $PM_{2.5}$ mass is lost. Analysing the particle size distributions measured by the APS in this experiment, it was identified that the main losses in the APS size range occurred for particles over 2.5 μm and particles with size less than 0.8 μm. Under the normal particle concentration range in this study, there were no significant differences between the loss ratios under the high and low concentrations condition for APS, CPC and DustTrak. Therefore the average loss ratios were used in correction for the outdoor concentrations that were obtained in simultaneous measurements.

2.4. *Sampling protocol*

When the non-simultaneous sampling method was used, five measurements of outdoor particle concentration were conducted first, followed by five measurements of indoor air taken in the same indoor location, under the normal ventilation condition. Based on this method, indoor and outdoor aerosol particle number concentrations and particle size distributions were obtained.

Using the simultaneous method, one hour measurements were conducted with the instrumentation located in the living room, first under so called normal and then under minimum ventilation conditions. Normal ventilation was defined as occurring when those doors and windows were opened that are most commonly kept opened by the occupants, while minimum ventilation occurs when all the doors and windows were closed. In the subtropical environment of Brisbane, under normal ventilation

conditions, air exchange rate ranges between about 2 h^{-1} and 5 h^{-1}, and under minium ventilation condition, between about 0.5 h^{-1} and 1.0 h^{-1} (Morawska & Jamriska 1994). Based on this method, the variations of indoor and outdoor aerosol particle number concentrations and particle mass concentrations ($PM_{2.5}$) with time, as well as the relationship of indoor and outdoor aerosol particle concentrations were obtained.

The measurements in the houses were conducted between March and August 1999, which is wintertime in Brisbane. The ranges of meteorological parameters during that time were: 9 ~ 25 km/h for wind speed, 110 ~ 283 degree for wind direction, 15 ~ 23 °C for temperature and 54 ~ 92% for relative humidity. In general, it was attempted to conduct the measurement when the outside conditions were relatively stable in terms of both meteorological conditions (particularly in terms of wind speed and direction) and particle concentration. These were called steady state conditions that allowed for the best understanding to be achieved and the interpretation made of the indoor/outdoor relationship and that removed additional uncertainties that would relate to rapid changes of outdoor conditions. The criteria of a rapid change of outdoor condition used in this study were: wind speed was over 38 km/h, or wind direction changing over 120 degree or the ratio of any two consecutive concentration value over 2 during the measurement period. For cases when rapid changes of outdoor condition occurred during the measurements, the data was not used in the indoor/outdoor relationship analysis.

Since the focus of this work was on the relationship between indoor and outdoor particles in the absence of indoor sources, care was extended to avoid and to prevent any indoor activities that would result in the generation or resuspension of particles. Thus, the residents were generally absent during the measurements and the researchers reduced their movements to an absolute minium.

2.5. *Data processing and analysis*

For the simultaneous measurement method, the data collected is in the form of six indoor concentrations measured within one minute, followed by six outdoor concentrations measured in the next minute. In order to avoid any effects of air from previous measurements still being present in the system after it has switched to the next measurements, the first of the six measurements were not included in the data analyses. For a similar reason, in order to avoid any effects of air mixing when the system is switching, the last of the six samples were also not included in the data analyses. The remaining central samples were averaged providing one value of concentration for each minute of sampling, for indoor and outdoor alternately.

Comparisons of indoor and outdoor particle size distributions were performed using the Kolmogorov-Smirnov (K-S) test which can provide information on the level of similarity of two particle size distributions. Based on the results of the K-S test, the question as to whether two particle samples (e.g. indoor and outdoor) originate from the same source could be answered (Morawska, *et al*., 1999; Parat *et al*., 1999). If the statistic index D in the K-S test is less than 0.118 for two submicrometer particle size spectra in the SMPS range, then there are no significant differences between the two spectra (p = 0.1). For large particles in the APS range, two size spectra are statistically similar if the index D in K-S test is less than 0.171 (Hays and Winkler, 1970).

3. RESULTS AND DISCUSSION

3.1. *Non-simultaneous measurements*

3.1.1. *Submicrometer particle concentrations*

Results of the non-simultaneous measurements of the indoor and outdoor submicrometer particle number (0.015 – 0.685 µm) concentrations are shown in Table

1. It can be seen from this table that both indoor and outdoor levels of submicrometer particle concentrations varied widely. However, comparison of the average values for particle concentration in indoor and outdoor air, reveals that they are very similar. The indoor/outdoor ratio of submicrometer particle concentrations varied from house to house ranging from a minimum of 0.44 to a maximum of 2.46, but the total average ratio was 1.07 ± 0.44. This result indicated that in general under normal ventilation conditions, and in the absence of indoor sources, the concentrations of submicrometer particles indoors tend to closely follow the concentrations outdoors. For cases when the indoor/outdoor ratios reached the extreme values of 0.44 and 2.46, it was shown that the shapes of the indoor and outdoor spectra were statistically different, indicating different sources of particles (see Table 1). The reasons for the differences were not investigated in each individual case but could have resulted from either the change in the outdoor size distribution caused by a source of a short term effect (passing vehicle), or less likely, by unidentified indoor sources.

3.1.2. *Supermicrometer particle concentrations*

Table 1 also presents a summary of the indoor and outdoor concentrations of particles in the supermicrometer range obtained by the APS (diameter: 0.54 – 19.81 µm) during non-simultaneous measurements. It can be seen from the results presented in the table, that both indoor and outdoor concentrations varied significantly. However, the average concentrations were very similar. The ratio of indoor and outdoor particle concentrations varied from building to building with a minimum of 0.47 and a maximum of 1.96, but the average ratio for all the houses was 1.0 ± 0.3. This result indicated that in general under normal ventilation conditions, and in the absence of indoor sources of large particles, the concentrations of large particles indoors also tend to closely follow the concentrations outdoors.

3.1.3. *Comparison of submicrometer and supermicrometer particle concentrations*

Comparing the concentration ratios of indoor and outdoor submicrometer and supermicrometer particles, it can be seen that about 50% of houses show the same pattern, in which both ratios (of submicrometer and supermicrometer particles) were either higher than one or lower than one, while the other 50% house show a reverse pattern, in which if one ratio is higher than one, the other ratio is lower than one. An example of an extreme reverse pattern are the ratios measured in Houses 16 and House 3. For House 3 the indoor supermicrometer particle concentration was nearly two times higher than the outdoor concentration, while the indoor submicrometer particle concentration was less than half of the outdoor concentration. The situation was directly opposite for House16 with the indoor supermicrometer particle concentrations less than half of outdoor concentrations and the indoor submicrometer particle concentration nearly two and a half times higher than the outdoor concentration. These results indicate that in individual cases the ratios of indoor and outdoor submicrometer and supermicrometer particle concentration may vary significantly and caution should be exercised when attempting to predict one ratio based on the information from the other. Based on the non-simultaneous SMPS and APS indoor and outdoor measurement results there are no correlations between the ratio of indoor/outdoor particle concentration and building type (e.g. dwelling age, height, location and brick or timber), and meteorological parameters (wind direction, wind speed, temperature and relative humidity).

3.1.4. *Particle size distributions*

The size distributions of both submicrometer and supermicrometer particles obtained during the measurements varied between the measurements and within the same measurements, however, it was noticed that when the particle concentration levels

indoors and outdoors were similar, the indoor and outdoor size distributions were similar as well. One the other hand, when the ratios of the indoor and outdoor particle number concentrations differed significantly from one, the indoor and outdoor size distributions were visibly different. It should be stressed, that while the submicrometer and supermicrometer spectra are presented on one diagram, they were measured by two different instruments, operating on different physical principles and measuring different particle properties. A smooth transition is thus not expected between SMPS and APS spectra.

The analysis conducted using the K-S test showed that for submicrometer particles there were no statistically significant differences between indoor and outdoor size distributions for all houses, with the exception of three: Houses 1, 3 and 16 (see Table 1). The test showed that the difference between the indoor and outdoor submicrometer particle size distributions for House16 was very clear at the 99% significance level ($p < 0.01$). A hypothesis was formed that there was a source of submicrometer particles operating in this house that was not obvious to the researchers. Further investigations revealed the presence of an oil heater and bread maker operating in this house. However, the mechanism through which the operation of these devices might have contributed to generation submicrometer particles has not been investigated.

Analyses conducted for supermicrometer particles showed there were no statistically significant differences between indoor and outdoor size distributions for all houses except for two: House 1 ($p = 0.05$) and House 16 ($p = 0.1$), (see Table 1).

Particle number median diameters (NMD) were calculated for all the houses to show that except for Houses 1, 3 and 16, the differences of NMD for indoor and outdoor supermicrometer particles were less than 0.10 µm for all other houses. The average NMD of indoor and outdoor particles measured with the SMPS were 0.028 µm and

0.027 μm, respectively, and of those measured with the APS, 0.91 μm and 0.89 μm, respectively.

Comparing the K-S test results with the differences of NMDs, it can be seen that when the difference in NMD is larger than 0.005 μm for submicrometer particles, or larger than 0.1 μm for larger particles, the indoor and outdoor size spectra are statistically different. Furthermore, comparing the K-S test results with the indoor/outdoor concentration ratios, it can be seen that when the ratio is smaller than 0.65 or larger than 1.30, the indoor and outdoor size spectra are in most cases significantly different.

3.2. *Simultaneous measurements*

By using the automatic sampling system, which allowed automatic switching from sampling indoors to sampling outdoors, the ratios of indoor to outdoor particle concentrations were measured under normal and minimum ventilation conditions. Since the outdoor numbers had been corrected for particle losses, the uncertainties of the results of outdoor concentrations and the indoor and outdoor ratios also increased.

3.2.1. *Normal ventilation conditions*

A summary of the CPC, APS, and DustTrak results of indoor and outdoor measurements conducted under normal ventilation conditions is presented in Table 2. Figure 1 shows a typical time series variation of the indoor and outdoor concentrations. The ranges of average indoor and outdoor concentrations were $(5.2 \sim 40.0) \times 10^3$ particles cm^{-3} indoor and $(5.7 \sim 48.7) \times 10^3$ particles cm^{-3} outdoor, for CPC measurements, $0.51 \sim 3.86$ particles cm^{-3} indoor and $0.45 \sim 3.96$ particles cm^{-3} outdoor, for the APS, and $4.4 \sim 15.3$ μg m^{-3} indoor and $4.7 \sim 18.4$ μg m^{-3} outdoor, for PM$_{2.5}$.

Inspecting the results presented in Table 2 and Figure 1 it can be seen that positive correlations between indoor and outdoor concentrations measured by the three different instruments are displayed for many houses, indicating that indoor concentrations are clearly affected by outdoor sources under normal ventilation conditions.

The average ratio of indoor to outdoor submicrometer particle concentrations (measured with the CPC) was 0.89±0.14. The average ratio of indoor to outdoor concentration of larger particles (measured by APS) was 0.97±0.14. The average ratio of indoor to outdoor PM$_{2.5}$ concentration was 1.01±0.14. In summary, under normal ventilation conditions the ratios of indoor to outdoor particle number and mass are very close to one, however, the indoor submicrometer particle concentrations appear to be marginally lower than the outdoor concentrations. One possible explanation for the results that the indoor/outdoor ratios of supermicrometer were higher than the indoor/outdoor ratios of submicrometer could be re-suspension of particle indoors by the activities of the researchers and operation of the instruments. However, the further investigations for this would be needed.

In order to identify whether the somewhat lower average ratio of submicrometer particles measured with the CPC indicates lower indoor concentrations in this size range, or whether it is an artefact resulting from some aspects of the experimental design (for example variation in particle losses in the system) additional indoor and outdoor particle concentration measurements were conducted. The measurements were conducted with the CPC using the non-simultaneous method, in which one-hour indoor measurement and then one-hour outdoor measurement were repeated for twenty-four hours. In this case the ratio of the indoor and outdoor submicrometer

particle concentrations was found to be 1.0±0.49. The time series of particle concentrations measured during these measurements is provided in Figure 2.

3.2.2. *Minimum ventilation conditions*

The results of CPC, APS and DustTrak measurements under minimum ventilation conditions are also presented in Table 2. The ranges of particle concentrations were $(4.9 \sim 21.3)\times10^3$ particles cm^{-3} indoor and $(5.0 \sim 52.8)\times10^3$ particles cm^{-3} outdoor, for the CPC; $0.46 \sim 3.74$ particles cm^{-3} indoor and $0.47 \sim 3.98$ particles cm^{-3} outdoor, for the APS; and $5.4 \sim 18.0$ μg m^{-3} indoor and $6.2 \sim 18.0$ μg m^{-3} outdoor, for the PM$_{2.5}$.

The average ratio of indoor to outdoor submicrometer particle concentrations (measured with the CPC) under minimum ventilation conditions was 0.78±0.49, the ratio for larger particles (measured with the APS) was 0.95±0.18 and for PM$_{2.5}$ concentrations was 1.08±0.22. It also can be seen from Table 2 that for many Houses the indoor/outdoor ratios for lager particle concentrations were higher than the ratios of indoor/outdoor submicrometer particle concentrations. Similarly, one possible explanation for these results could be re-suspension of particle indoors by the activities of the researchers and operation of the instruments.

The time series analyses of the relationships between indoor and outdoor particle concentrations under minimum ventilation conditions show that, contrary to the situation for normal ventilation conditions, in most cases the correlation is poor. It suggests that under this ventilation condition, indoor concentrations are not immediately affected by outdoor concentration changes. In the case of low ventilation rate, it could be expected that the effect of outdoor air could be delayed and inversely proportional to the air exchange rate. An example of variation with time of the indoor

and outdoor CPC, APS, and $PM_{2.5}$ concentrations under minimum ventilation conditions, as well as the indoor to outdoor ratio calculated for an assumed delay in the effect of outdoor air on the indoor concentrations, are provided in Figure 3. The correlation coefficients for the calculations without a delay (Ro^2) and with a delay (Rs^2) are provided in the legend. Analysing the results presented in Figure 3, it can be seen that introduction of a "delay" factor significantly improves the correlation between indoor and outdoor concentrations.

3.3. *Discussion and conclusions*

The relationship between indoor and outdoor airborne particles was investigated for sixteen residential houses located in a suburban area of Brisbane, Australia. The submicrometer particles were measured using the Scanning Mobility Particle Sizer (SMPS) in the size range from 0.015 to 0.685 μm. The larger particles were measured using the Aerodynamic Particle Sizer (APS) in the range from 0.54 to 19.81 μm. An approximation of $PM_{2.5}$ was also measured using a DustTrak. The measurements were conducted for normal and minimum ventilation conditions using simultaneous and non-simultaneous measurement methods designed for the purpose of the study. The focus of this study was on the relationship between indoor and outdoor particle concentration in the absence of clear or obvious indoor sources, and thus care was taken to de-activate all indoor sources during the course of measurements. The study achieved all its objectives and in particular:

1. The average submicrometer particle concentrations of outdoor and indoor air (7.1×10^3 and 7.4×10^3 particles cm^{-3} , respectively), measured in this study, are very close to the value of 7.4×10^3 particles cm^{-3}, reported previously as the average for the city of Brisbane by Morawska *et al*., (1998). The average number median

diameter of 0.027 μm for outdoor air and of 0.028 μm for indoor air determined in this study is somewhat lower than the previously reported value of 0.040 μm (Morawska *et al*., 1998).

2. Analyses of submicrometer particle size distribution results show, that normally 85 –95% of submicrometer particles in indoor and outdoor air are smaller than 0.1 μm (ultrafine particles).

The relationship between indoor and outdoor particles was investigated using two methods: by calculation of the ratio of indoor to outdoor particles and comparing temporary and average values as well as a time series, and by using the Kolmogorov-Smirnov test to compare indoor and outdoor particle size spectra.

Comparison of the ratios of indoor to outdoor particle concentrations revealed that while temporary values of the ratio vary in a broad range from 0.2 to 2.5 for both lower and higher ventilation conditions average values of the ratios were very close to one regardless of ventilation conditions and of particle size range. It is estimated that under minium ventilation conditions in the houses investigated the air exchange rate varied between about 0.5 h^{-1} and 1.0 h^{-1} and under normal ventilation conditions, between about 2 h^{-1} and 5 h^{-1}. The ratios obtained by non-simultaneous measurement methods were 1.07 ± 0.44, and 1.0±0.49 (24 hours average) for submicrometer particles; 1.0 ± 0.3 for supermicrometer particles. The ratios obtained by simultaneous measurement methods were 0.78±0.49 (minimum ventilation condition) and 0.89±0.14 (normal ventilation conditions) for submicrometer particles; 0.95±0.18 (minimum ventilation condition) and 0.97±0.14 (normal ventilation conditions) for supermicrometer particles; and 1.08±0.22 (minimum ventilation condition) and 1.01±0.14 (normal ventilation conditions) for $PM_{2.5}$ fraction.

The Kolmogorov-Smirnov test results indicate that indoor and outdoor particle spectra are very similar for many houses in this study. Based on the results of the test, on the differences in number median diameters and the concentration ratios of indoor and outdoor particles, it was found that when the concentration ratio is close to one or the difference in number median diameters between indoor and outdoor distribution is small (0.005 μm < for submicrometer particles and 0.1 μm < for supermicrometer particles), the differences between the indoor and outdoor size spectra are not significant according to the K-S test. This implies that both indoor and outdoor particles originate from the same source.

Comparison of the time series of indoor to outdoor particle concentrations shows a clear positive relationship existing for many houses under normal ventilation conditions for both submicrometer and supermicrometer particles. These results suggest that for normal ventilation conditions, outdoor particle concentrations could be used to predict instantaneous indoor particle concentrations.

Under minimum ventilation conditions, for most houses there is no clear correlation between time series of indoor and outdoor particle concentrations. The correlation improves if a "delay constant" is introduced in calculations accounting for a delay in which the indoor air concentration follows the outdoor concentration for low ventilation conditions. In general, instantaneous outdoor particle concentration cannot be reliably used to predict indoor particle concentrations unless air exchange rate is known, thus allowing for estimation of the "delay constant".

3. A conclusion that can be drawn from this study for the purpose of exposure assessment is that the study design and the choice of parameter measured should depend on the expected relation between exposure and health effects considered. For cases when average concentrations are expected to be linked to health effects, average

20

outdoor concentrations can be used as a good approximation of indoor concentrations for air exchange rates above 0.5 h^{-1} in the absence of indoor particle sources, while the total human exposure results from particles generated by both indoor and outdoor sources. The objective of this study was to investigate contribution of outdoor sources to indoor concentrations. When, however, temporary values are required to investigate for example acute health effects, outdoor concentration cannot be reliably used as an approximation of indoor concentrations, particularly for air exchange rates of the order of 1 h^{-1} or lower.

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Table 1. Summary of the results from indoor and outdoor SMPS (0.015 ~ 0.685 µm) and APS (0.54 ~ 19.81 µm) non-simultaneous measurements (particle number concentration: particles cm^{-3})

House	Indoor Concentration		Indoor NMD		Oudoor NMD		I/O Concentration Ratio	
ID	SMPS (×10^3)	APS	SMPS (µ m)	APS (µ m)	SMPS (µ m)	APS (µ m)	SMPS	APS
H1	2.9±0.4	6.03±2.0	0.032	0.94	0.039*	0.83**	1.46	1.30
H3	4.3±0.6	1.47±0.10	0.034	0.87	0.026*	0.78	0.44	1.96
H4	3.5±0.2	1.06±0.04	0.023	0.87	0.023	0.88	0.68	0.83
H5	10.5±0.2	5.2±0.15	0.038	0.9	0.036	0.9	0.90	1.12
H6	7.8±0.4	1.55±0.08	0.023	0.95	0.024	0.96	0.97	0.89
H7	0.7±0.09		0.028		0.027	0.94	0.86	
H8	10.9±0.4	1.32±0.09	0.023	0.72	0.023	0.66	1.25	0.73
H9	8.0±0.4	1.43±0.06	0.025	0.96	0.024	0.89	1.03	1.00
H12	2.0±0.4	4.72±0.08	0.023	0.95	0.024	0.98	0.92	0.90
H13	12.9±1.9	2.81±0.13	0.023	0.92	0.023	0.91	0.91	0.93
H14	3.7±0.2	1.11±0.04	0.029	0.81	0.030	0.8	1.23	1.13
H15	7.7±0.3	4.15±0.05	0.033	0.84	0.032	0.83	0.90	0.89
H16	18.7±2.7	1.11±0.05	0.033	1.23	0.027**	1.42*	2.46	0.47
H17	8.4±0.4	1.80±0.05	0.024	0.91	0.024	0.91	0.88	1.02
H18	16.1±0.8	1.99±0.08	0.027	0.74	0.028	0.73	1.16	0.93
H19	0.8±0.09	1.77±0.05	0.024	0.99	0.023	0.92	1.04	0.87
Average.	**7.4±5.4**	**2.5±1.67**	**0.028**	**0.91**	**0.027**	**0.89**	**1.07**	**0.998**
Max	18.7	6.03	0.038	1.23	0.039	1.42	2.46	1.96
Min	0.7	1.06	0.023	0.72	0.023	0.66	0.44	0.47

NMD: number median diameter (µm). K-S Test: Kolmogorov-Smirnov Test Result, for SMPS D value, *: $p < 0.05$, **: $p < 0.01$,

for APS D value, *: $p < 0.05$, **: $p < 0.01$.

Table 2. Summary of the indoor to outdoor particle concentration ratios obtained for APS (0.54 ~ 19.81 µm), CPC (0.007 ~ 0.808 µm), and $PM_{2.5}$ simultaneous measurements conducted under normal ventilation condition and minimum ventilation condition.

House ID	Normal Ventilation Condition						Minimum Ventilation Condition					
	APS		CPC		$PM_{2.5}$		APS		CPC		$PM_{2.5}$	
	Ratio	R^2	Ratio	R^2	Ratio	R^2	Ratio	R^2	Ratio	R^2	Ratio	R^2
H1	0.98	0.13	1.32	0.01	1.05	0.34*	0.94	0.37	1.07	0.21	1.06	0.62**
H3	1.42	0.33			0.74	0.52**	0.99	0.07	0.21	0.00	0.76	0.05
H4	0.91	0.26	0.85	0.71**	0.94	0.85**	0.96	0.01	0.69	0.01	0.88	0.54**
H5	0.91	0.31	0.81	0.05	0.94	0.23	0.74	0.09	0.56	0.04	0.91	0.40*
H6	0.98	0.60**	0.93	0.84**	0.95	0.03	1.21	0.06	0.70	0.02	1.55	0.20
H7	0.94	0.69**	0.96	0.27	1.04	0.86**	1.00	0.25	1.69	0.14	1.08	0.31
H8	0.82	0.41*	0.83	0.00	1.01	0.44*	0.70	0.00	0.41	0.19	0.88	0.00
H9	1.08	0.03	0.86	0.35*	1.02	0.80**	0.99	0.41*	0.81	0.54*	1.17	0.03
H12	0.91	0.72**	0.87	0.29	0.93	0.92**	0.89	0.74**	0.55	0.00	1.03	0.14
H13	0.97	0.94**	1.00	0.85**	1.04	0.81	0.91	0.00	0.50	0.87**	0.97	0.00
H14			0.79	0.71**	0.97	0.58**	0.93	0.14	0.79	0.05	1.02	0.21
H15	0.91	0.67**	0.92	0.19	1.06	0.39*	0.87	0.80**	1.99	0.29	1.09	0.84**
H16	0.93	0.64**	0.63	0.45**	1.41	0.06	0.87	0.45*	0.37	0.01	1.21	0.02
H17	0.88	0.81**	0.87	0.53**	1.03	0.82**	0.78	0.00	0.44	0.21	1.04	0.20
H18	0.89	0.90**	0.86	0.02	1.02	0.63**	1.41	0.05	0.94	0.07	1.52	0.10
Average	**0.97**		**0.89**		**1.01**		**0.95**		**0.78**		**1.08**	
Max	1.42		1.32		1.41		1.41		1.99		1.55	
Min	0.82		0.63		0.74		0.70		0.21		0.76	

Notes: R^2: coefficient of indoor/outdoor correlation; *: $p < 0.05$, **: $p < 0.01$; Since the outdoor numbers had been corrected for particle losses, the uncertainties of the results of the indoor and outdoor ratios also increased.
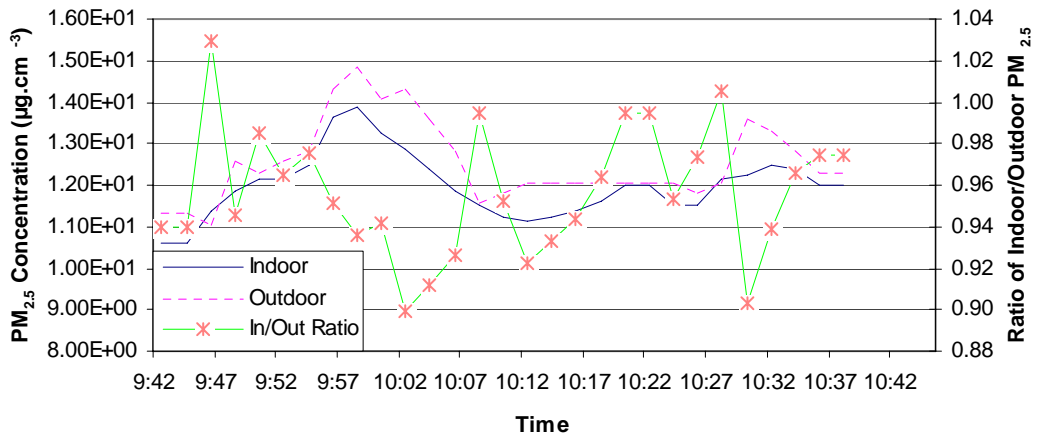

27
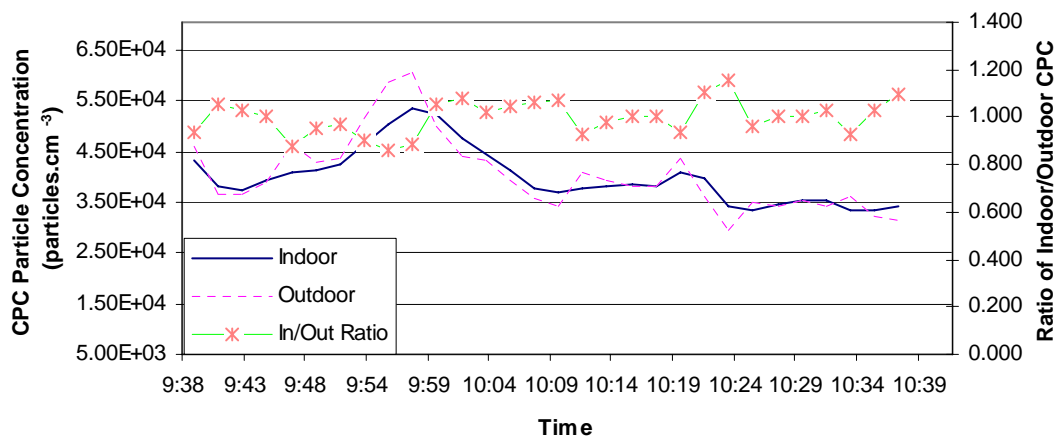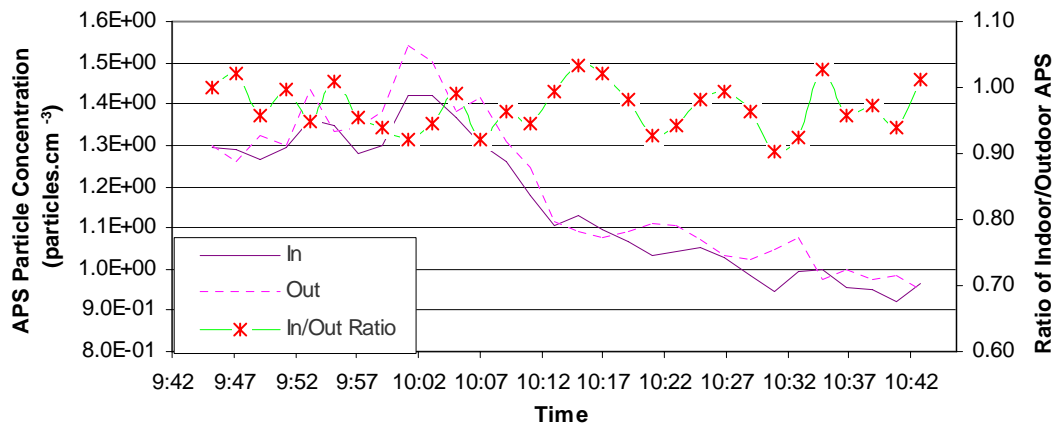
Figure 1. Time series of the indoor and outdoor particle concentrations in the APS (0.54 ~ 19.81 µm), CPC (0.007 ~ 0.808 µm) and $PM_{2.5}$ ranges as well as the variation of indoor to outdoor concentration ratios.
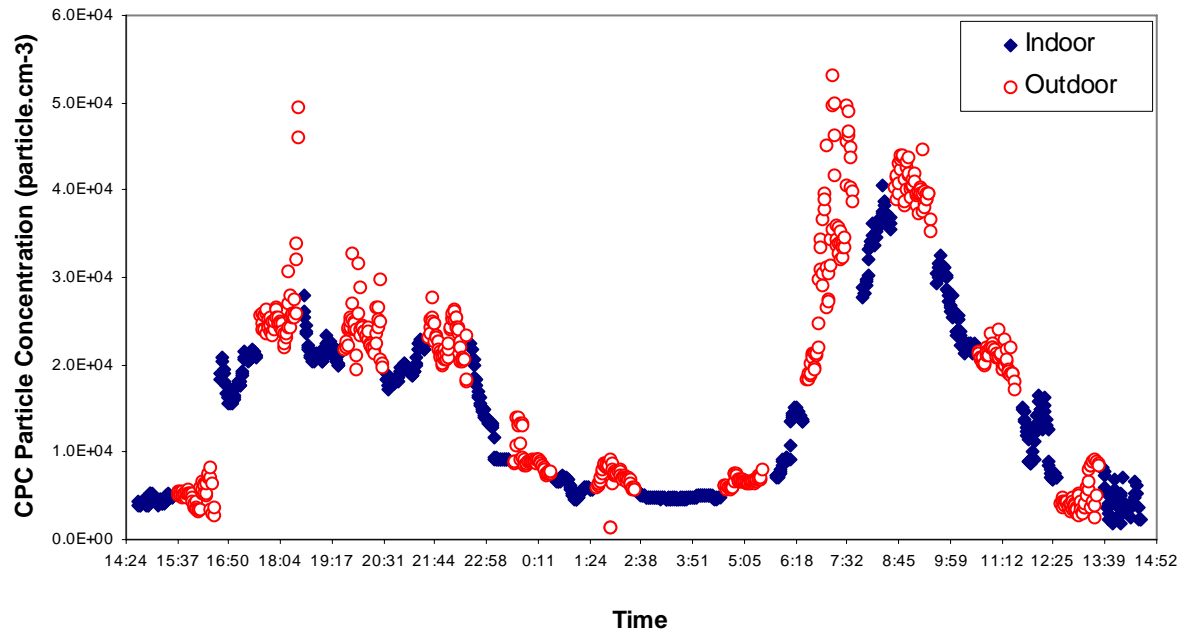
Figure 2. The variations of the indoor and outdoor submicrometer particle (0.007 ~ 0.808 µm) concentrations with time (21-22 June 2000), under normal ventilation condition in a residential house.
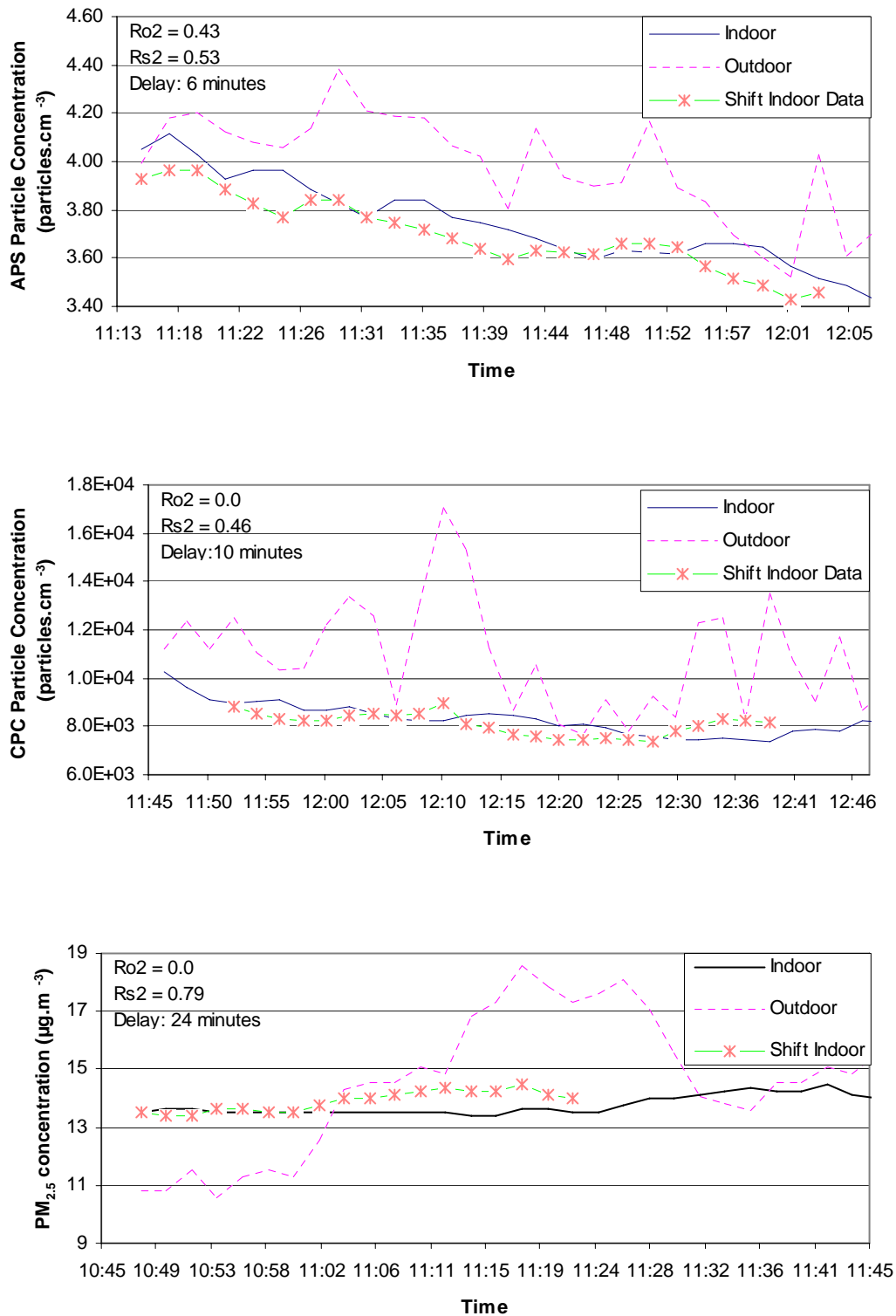
Figure 3. Time series of the indoor and outdoor particle concentrations in the APS (0.54 ~ 19.81 µm) and CPC (0.007 ~ 0.808 µm) ranges under minimum ventilation condition. Also provided are the indoor spectra re-calculated using "time delay" coefficient. Ro^2 and Rs^2 values are for indoor/outdoor and shifted indoor and outdoor concentration ratios, respectively.