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Characterization of particle number concentrations and $PM_{2.5}$ in a school: influence of outdoor air pollution on indoor air

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Abstract

Background, Aim and Scope The impact of air pollution on school children's health is currently one of the key foci of international and national agencies. Of particular concern are ultrafine particles which are emitted in large quantities, contain large concentrations of toxins and are deposited deeply in the respiratory tract.

Materials and methods In this study, an intensive sampling campaign of indoor and outdoor airborne particulate matter was carried out in a primary school in February 2006 to investigate indoor and outdoor particle number (PN) and mass concentrations ($PM_{2.5}$), and particle size distribution, and to evaluate the influence of outdoor air pollution on the indoor air.

Results For outdoor PN and $PM_{2.5}$, early morning and late afternoon peaks were observed on weekdays, which are consistent with traffic rush hours, indicating the predominant effect of vehicular emissions. However, the temporal variations of outdoor $PM_{2.5}$ and PN concentrations occasionally showed extremely high peaks, mainly due to human activities such as cigarette smoking and the operation of mower near the sampling site. The indoor $PM_{2.5}$ level was mainly affected by the outdoor $PM_{2.5}$ ($r = 0.68$, $p<0.01$), whereas the indoor PN concentration had some association with outdoor PN values ($r = 0.66$, $p<0.01$) even though the indoor PN concentration was occasionally influenced by indoor sources, such as cooking, cleaning and floor polishing activities. Correlation analysis indicated that the outdoor $PM_{2.5}$ was inversely correlated with the indoor to outdoor $PM_{2.5}$ ratio (I/O ratio) ($r = -0.49$, $p<0.01$), while the indoor PN had a weak correlation with the I/O ratio for PN ($r = 0.34$, $p<0.01$).

Discussion and Conclusions The results showed that occupancy did not cause any major changes to the modal structure of particle number and size distribution, even though the I/O ratio was different for different size classes. The I/O curves had a maximum value for particles with diameters of 100 – 400 nm under both occupied and unoccupied scenarios, whereas no significant difference in I/O ratio for $PM_{2.5}$ was observed between occupied and unoccupied conditions. Inspection of the size-resolved I/O ratios in the preschool centre and the classroom suggested that the I/O ratio in the preschool centre was the highest for accumulation mode particles at 600 nm after school hours, whereas the average I/O ratios of both nucleation mode and accumulation mode particles in the classroom were much lower than those of Aitken mode particles.

Recommendations and Perspectives The findings obtained in this study are useful for epidemiological studies to estimate the total personal exposure of children, and to develop appropriate control strategies for minimizing the adverse health effects on school children.

Key words: I/O ratios; Particle number concentration; $PM_{2.5}$; Aitken mode particles; School

1. Background, Aim and Scope

Indoor air quality in schools has attracted increasing public attention in recent years due to the fact that children spend up to ten hours per day at school (Leickly, 2003), and the health impact of air pollutants is much higher for small children than for adults in similar environments (WHO, 2005). There are many factors which affect indoor air quality, such as emissions from indoor sources, the operation of ventilation systems and the penetration of outdoor air pollutants indoors (Wallace *et al.*, 1996; Vette *et al.*, 2001; Morawska *et al.*, 2003; Godish, 2004; Guo *et al.*, 2008). In the absence of major indoor sources, indoor air quality is directly linked to the outdoor air quality (Godish, 2004). Previous studies consistently indicated that outdoor particulate matter (PM) was the most important source of particulate matter measured indoors (e.g. Jamriska *et al.*, 2000; Koponen *et al.*, 2001; Sawant *et al.*, 2004; Martuzievicius *et al.*, 2008). Studies have also shown that combustion sources such as cooking, fireplaces, kerosene heaters and cigarette smoke are the predominant sources of indoor particles (e.g. Wallace, 2000; Lee *et al.*, 2002; He *et al.*, 2004; Afshari *et al.*, 2005; Hussein *et al.*, 2006). In addition, emissions from consumer products and building materials were found to be significant aerosol sources (Sanchez *et al.*, 1987; Mathews., 1987; Afshari *et al.*, 2005; Hussein *et al.*, 2006).

A growing body of epidemiological data has indicated consistent and coherent associations between particulate matter and excess mortality and morbidity (e.g. Samet *et al.*, 1981; Pope III, 1991; Schwartz, 1991; Pope and Dockery, 1999; Weichenthal et al., 2007; Andersen et al., 2008).

In addition, accumulated evidence has supported the hypothesis that particle number (PN) concentration is an important determining factor affecting lung injury (e.g. Chen *et al.*, 1995; Oberdorster *et al.*, 1995; Penttinen et al., 2001; Weichenthal et al., 2007). More importantly, indoor concentrations of many pollutants, including airborne particles, are often higher than those typically encountered outdoors (Jones, 1999; He e*t al.*, 2004; Hussein *et al.*, 2006), and since people spend most of their time in different indoor micro-environments (Jenkins *et al.*, 1992; Robinson and Nelson, 1995; Kousa e*t al.*, 2002), the health impact of indoor air pollution may be significantly higher. In particular, the impact could be more serious on school children, since children are smaller and have a higher metabolic rate than adults, which means that they breathe in more air per unit of body weight and are generally more susceptible to the effects of indoor air pollutants (DEWHA, 2001). Previous studies indicate that school children aged between 5-12 years spend 8-10 hours per day at school, of which, only 2-3 hours are spent outdoors (Leickly, 2003; Xue *et al.*, 2004; Cleland *et al.*, 2009). Hence, the adverse effects of indoor air pollutants on school children, including particulate matter, cannot be underestimated.

A number of indoor studies on particulate air pollution have been conducted in schools, however most of them focused on large particles such as $PM_{2.5}$ (particles with aerodynamic diameter equals to and less than 2.5 µm), and PM_{10} (diameter ≤ 10 µm) (e.g. Gold *et al*., 1999; Lee *et al*., 2002; John *et al*., 2007; Annesi-Maesano *et al*., 2007; Oravisjarvi *et al*., 2008). These studies demonstrated that indoor PM levels are greatly affected by the presence of pupils and the intensity of their indoor activities, as well as outdoor PM levels. Furthermore, correlation analysis between particle mass concentrations ($PM_{2.5}$ and PM_{10}) and health effects in these studies showed contradictory results, implying that the number of submicrometer particles may be more health relevant than the mass of $PM_{2.5}$ and PM_{10} which is usually measured. Though some indoor studies on PN concentrations in non-school environments have been carried out (e.g. Franck *et al*., 2003, 2006; He *et al*., 2004; Wallace, 2006; Gehin *et al*., 2008), to date, only a handful of studies have been undertaken on PN concentrations in school environments. For instance, Blondeau *et al.* (2004) investigated the relationship between outdoor and indoor air quality in eight French schools. They found that the I/O ratio varied in from 0.03 to 1.79 for PN. Diapouli *et al*. (2007)

examined indoor and outdoor ultrafine particles concentrations in the schools of Athens in winter and found that the indoor-to-outdoor concentration (I/O) ratios were below 1.00 at all sites. Koponen *et al.* (2001) and Guo *et al.* (2008) indicated that ventilation had a strong influence on indoor particles and the I/O ratio was different in different particle size classes. Parker *et al.* (2008) reported that indoor concentrations of submicron particulate matter were about one-eighth of outdoor levels in an elementary school. A study on the relationship between PN concentration and hospital admissions in children in Copenhagen revealed that ultrafine and accumulation mode particles were relevant to paediatric asthma (Andersen *et al*., 2008). Scientific knowledge on the correlation between indoor and outdoor PN concentrations, the origins and transport of indoor submicron particles, and the factors affecting the I/O ratios for PN in schools located in warmer climates with higher ventilation rates remains limited. Therefore, this study focused on the I/O ratios of total PN concentration and size distribution for particles in the range 0.015 to 0.790 μm, and of $PM_{2.5}$ concentration. The indoor aerosol particle concentrations were monitored in two classrooms where different indoor activities were undertaken. A series of statistical analyses including univariate and multivariate analyses (i.e. correlation, regression, *t*-test, one-way ANOVA, and ANOVA with *post-hoc* multiple comparisons) were conducted to provide insight as to the impact of relevant parameters and pupils' activities on the I/O ratios. The statistical analyses were based on measurement data from the school under investigations. The influence of room occupancy and outdoor concentration level on the I/O ratios is also discussed.

2. Materials and Methods

2.1. Sampling site description

Situated on the fringe of the inner urban redevelopment zone, the subject school is located about 5 km south of the Brisbane Central Business District, Queensland, in subtropical eastern Australia (27.48°S, 153°E). Owing to the opposite directions of sea and land breezes, the prevailing wind in the morning is south-easterly towards the city and in the afternoon north-westerly wind brings urban air to the school. In February (the sampling period) in Brisbane, the temperature is between 21 and 29°C and the mean rainfall is about 158 mm. The mean daily sunshine is 6.6 hours and the mean number of cloudy days is 13.4 (http://www.bom.gov.au/climate/averages/tables/cw_040214.shtml). In this study, the outdoor sampling site was located at the school oval, which is a large outdoor grass playing field (Figure 1). The site was surrounded by a road which carried moderate-heavy traffic about 50 m to the west, a railway to the north and some buildings and a walking path to the south and east. The major outdoor pollution sources around the school were expected to be vehicle emissions.

The indoor sampling sites were located in a classroom (named C101) and a preschool centre (Figure 1). C101 is on level 2 of the building, while the preschool is located in the southern side of the school which is in the vicinity of the walking path. The classroom (C101) was predominately used for art classes including activities such as painting, gluing, drawing etc, by children aged 6-12 years. Therefore, a large number of bottles, tubes and liquids used for art activities were present in many locations within the classroom. Occasionally, it was also used for foreign language and religion lessons. The classroom was 50 m^2 and it could accommodate 40 pupils. The preschool centre was mainly for children under 6 years old. It was 60 m^2 with a capacity of 25-30 people. The children came to the centre on weekday mornings and conducted activities such as playing, drawing and studying,before leaving for home at 3:00pm. The children usually had a morning tea break between 10:00-11:00am and a lunch break at 12:30-13:30pm. There was a small open kitchen in the corner inside the preschool centre, 1 m away from the sampling spot and during these periods, a microwave oven and a small cooking heater were used for heating food and boiling water. In addition, the classroom and the preschool centre were subject to the same cleaning schedule as the rest of the school (e.g. early morning and late afternoon on weekdays). The 6-12 years old children spent 2 h of their 8 h school day outdoors for meal breaks, while the preschool children spent about a total of 3h on the playground outdoors.

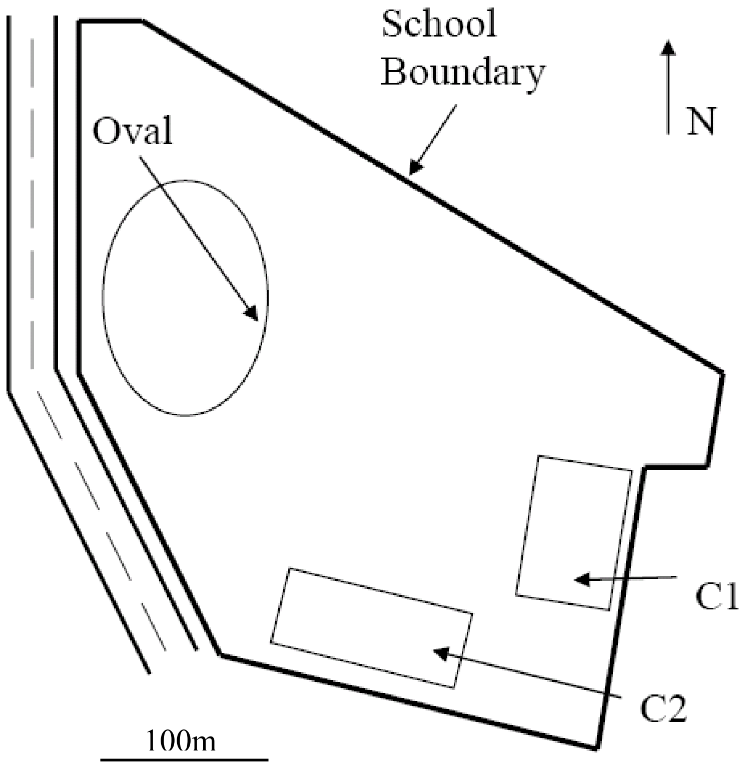


Figure 1 Schematic diagram on the sampling locations and surrounding area, including the outdoor (oval) and indoor sites (classroom - C1, preschool – C2).

2.2. Instrumentation and parameters measured

The parameters measured in this study included particle number concentration and size distribution, $PM_{2.5}$ mass concentration, temperature, relative humidity, wind direction and wind speed. Particle number concentration and size distribution in the size range of 0.015 to 0.790 µm were simultaneously measured indoors and outdoors using two Scanning Mobility Particle Sizers (SMPS), each comprising of an Electrostatic Classifier (EC) (TSI, Model 3071A) and Condensation Particle Counter (CPC) (TSI Models 3010 and 3025). All size bins of the SMPS (in total 109 bins) were considered and referred to "size classes" in the text. The SMPS operates on the principle of particle classification by the EC according to their electrical mobility, which is a function of their size; followed by particle counting by the CPC, which utilises laser light scattering. The whole process is automated and software controlled. A time resolution of 5 minutes was selected for the SMPS. Indoor and outdoor $PM_{2.5}$ concentrations were simultaneously measured by two DustTraks (TSI, Model 8520). The readings from the DustTraks were calibrated by a Tapered Element Oscillating Microbalance (TEOM) (Rupprecht & Patashnick, Model 1400A) using an equation which was obtained from a calibration experiment ($PM_{2.5(TEOM)}$ = 0.394 $PM_{2.5(DustTrak)}$ + 4.450, r = 0.91). The DustTraks were calibrated for ambient air dominated by traffic emissions by running it side by side with the TEOM at the school oval for a week. The logging interval used for the DustTrak/TEOM comparison assessment was 5 minutes. Both DustTrak units operated with a 2.5 μm impaction inlet (50% cut–off efficiency for particles larger than 2.5 μm), and a logging interval of 30 seconds. Wind direction, wind speed, temperature and relative humidity were continuously measured by a portable weather station (Davis Instruments Weather Monitor II), with a logging interval of 20 seconds.

2.3. Monitoring design

Outdoor measurements of $PM_{2.5}$ concentrations, together with meteorological conditions, were conducted in a fixed location at the school's oval, from 24 January to 17 February 2006 and particle number concentrations were measured from 8 – 17 February 2006. The instrumentation was installed and operated in an air-conditioned trailer where outdoor air was drawn into the instruments via conductive, plastic 1.5 m long tubes with an inner diameter of 8 mm, from an inlet on the roof of the trailer. An air splitter was applied for cases where several instruments sampled the air. Losses in the inlet tube were estimated based on laminar flow diffusion theory (Baron and Willeke, 2001).

Indoor $PM_{2.5}$ and particle number concentration measurements were conducted simultaneously with outdoor measurements. That is, $PM_{2.5}$ measurement started on 24 January whereas the monitoring of particle number concentration was initiated on 8 February 2006. The particle number concentrations were monitored at two locations in sequence: the preschool centre followed by the classroom C101. The sampling periods in the preschool centre and in the classroom were from 8 – 14 February 2006 and 14 – 17 February 2006, respectively. School hours were between 9 am and 3 pm on weekdays.

2.4. Survey

During the sampling period, whenever unusually high concentrations of air pollutants were observed, consultation with school teachers and staff was conducted immediately to explore the possible sources of those high peaks (identifying the potential sources and activities). All activities that could generate elevated air pollutant levels and took place near the sampling locations were also recorded.

2.5. Statistical analysis

All statistical analyses, including univariate and multivariate analyses (i.e. correlation, regression, *t*-test, one-way ANOVA and ANOVA with *post-hoc* multiple comparison) were performed using the SPSS statistical software package (SPSS Inc.). Non-parametric tests were undertaken to confirm the parametric results. That is, the corresponding non-parametric tests led to the same conclusions of significance/non-significance as the parametric tests.

3. Results and Discussion

3.1. Temporal trends of air pollutants

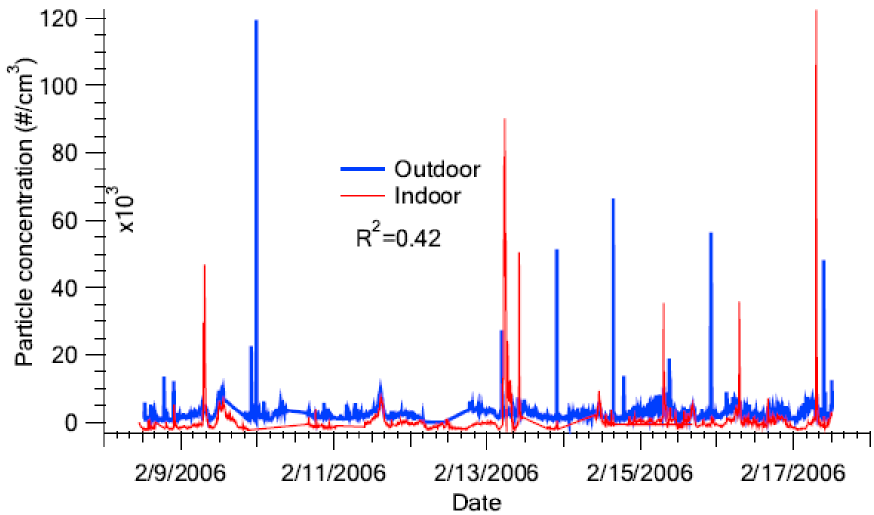
3.1.1. Temporal trends of particle number concentrations

Figure 2 shows the time series of indoor and outdoor particle number (PN) concentrations and $PM_{2.5}$ during the sampling period. It was found that the average outdoor PN concentration was $2.65 \times 10^3 \pm 1.52 \times 10^2$ cm^{-3} (mean ± 95% Confidence Interval). Very high PN concentrations ($5.0 \times 10^4 - 1.2 \times 10^5$ cm^{-3}) were often observed on the oval late at night (10pm – 0am), mainly due to local residents smoking cigarettes next to the sampling trailer whilst they were playing football at the oval during that time. It should be noted that local residents could easily access to the oval after school hours and on weekends, when the researchers were not always present on site. In addition, two peaks were observed in the morning on 13 February (~3.0×10^4 cm^{-3}) and 17 February (~4.5×10^4 cm^{-3}). These were caused by the operation of mower at the oval by the school staff. Small spikes in early morning at 6-8 am and late afternoon at 3-7 pm were also found for outdoor samples. This was consistent with traffic rush hours on weekdays, suggesting the effect of vehicular emissions. Nevertheless, the PN concentration levels due to vehicular emissions were much lower than those caused by nearby sources (i.e. smoking and operation of mower).

On the other hand, the average indoor PN concentration was $3.19 \times 10^3 \pm 2.63 \times 10^2$ cm^{-3}, which is statistically higher than average outdoor level. Very high indoor PN concentrations were often observed at 6-8 am and relatively low peaks were found at 3-6 pm in the classrooms. These elevated values were mainly caused by cleaning activities which were carried out twice per day. However, the morning peaks were generally 10 times higher than that in the afternoon, reflecting that source strength in the morning was much larger than that in the afternoon. Indeed, the highest indoor PN concentration (~1.1×10^5 cm^{-3}) observed at 7-8 am on 17 February was attributed to the operation of floor polishing machine, whereas the small spikes found at 3-5 pm on 15 February (~5- 8×10^3 cm^{-3}) were caused by wet cleaning with detergents. In addition, medium-size peaks (1-5×10^4 cm^{-3}) were sometimes observed at 10-11am in the morning in the preschool centre due to the cooking activities.

3.1.2. Temporal trends of particle mass concentrations

The temporal variations of indoor $PM_{2.5}$ generally followed the pattern of outdoor $PM_{2.5}$, but the fluctuation of indoor $PM_{2.5}$ was much lower than that of outdoor $PM_{2.5}$ during the sampling period (Figure 2). The average outdoor $PM_{2.5}$ concentration was 11.6 ± 0.8 μg/m^3 (mean ± 95% C.I.) whereas the mean indoor $PM_{2.5}$ level was 6.7 ± 0.2 μg/m^3. The highest outdoor $PM_{2.5}$ concentration (111 μg/m^3) was found in the morning on 5 February, which was attributed to the nearby smoking activities. In fact, the average $PM_{2.5}$ concentration on 4-5 February was 32 ± 5 μg/m^3, which was much higher than the average outdoor level for the whole period. The time series of outdoor $PM_{2.5}$ reflected the impact of vehicular emissions as morning (~7 am) and afternoon (~6 pm) peaks were regularly observed on weekdays, which were consistent with the traffic rush hours. In contrast to PN concentrations, the indoor $PM_{2.5}$ level was more likely to be affected by outdoor penetration rather than indoor sources related to the presence of pupils and the intensity of their indoor activities. This is consistent with the observations from other studies (e.g. Fromme *et al.*, 2008; Kingham *et al.*, 2008; Branis *et al.*, 2009). These studies revealed that there was a close relationship between indoor and outdoor $PM_{2.5}$; and the physical activity of pupils mainly resulted in the re-suspension of indoor coarse particles and greatly contributed to the increase of PM_{10} in classrooms. In addition, the influence of indoor cleaning and cooking activities was not as apparent as it was for PN concentrations (Figure 2), suggesting that these indoor activities might mainly generate submicrometer particulate matter, which contributes very little to $PM_{2.5}$ mass concentrations.



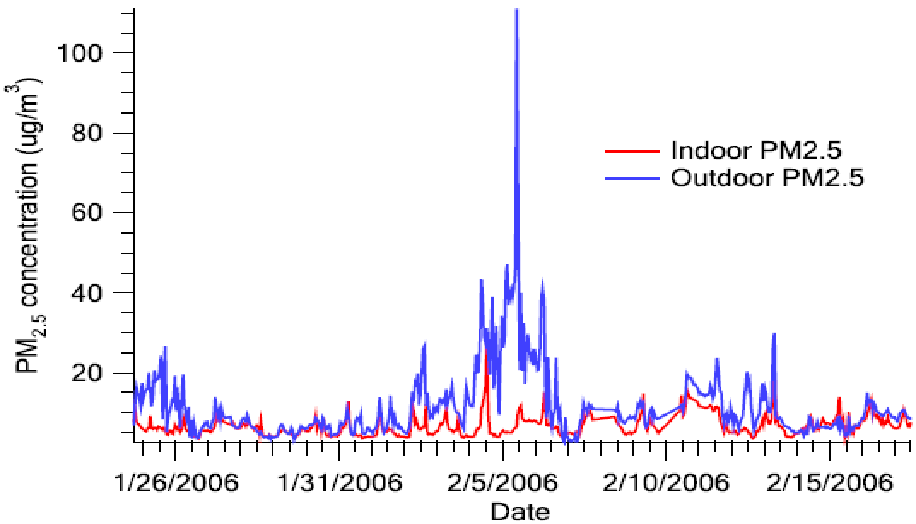


Figure 2 Time series of indoor and outdoor PN concentrations and $PM_{2.5}$ during the sampling period. The different time scales indicate the difference of sampling periods for PN and $PM_{2.5}$

3.2. Indoor and outdoor particulate matter correlations and size distribution

Table 1 shows the Spearman's rho correlations between indoor and outdoor PN concentrations, $PM_{2.5}$ and I/O ratios. It is noteworthy that in order to correctly explore the relationship between $PM_{2.5}$ and PN, the $PM_{2.5}$ data used for the correlation analysis were those collected during the same period as that for PN data, namely 144 hours of $PM_{2.5}$ and PN data collected from 12:30pm on 8 February to 11:30am on 17 February. It can be seen that outdoor $PM_{2.5}$ concentration was significantly correlated with indoor $PM_{2.5}$ (r = 0.68, p<0.01), suggesting that outdoor $PM_{2.5}$ had predominant impact on indoor $PM_{2.5}$ levels. It should be noted that the indoor to outdoor correlation coefficients for $PM_{2.5}$ (and PN) will presumably improve if the lag effect is considered. There is a well-known lag between the indoor and outdoor concentrations caused by the penetration time through the building envelope. Theoretically, during the occupancy period, when the windows are open, the I-O lag will be shorter and during non-occupancy (windows closed) it will be longer. It wasalso found that outdoor $PM_{2.5}$ had an inverse correlation with I/O ratios for $PM_{2.5}$ (r = -0.49, p<0.01), indicating that the I/O ratios for $PM_{2.5}$ concentrations would be reduced when outdoor $PM_{2.5}$ concentrations increased. It is not surprising that indoor $PM_{2.5}$ had a positive correlation with the I/O ratio for $PM_{2.5}$, as an increase in indoor $PM_{2.5}$ would lead to an increase in the I/O ratio. However, no correlation was observed between indoor $PM_{2.5}$ and the I/O ratio for PN concentration (r = -0.075, p = 0.37).

Good correlations were found between indoor and outdoor PN concentrations (r = 0.66, p<0.01), and between indoor PN concentration and the I/O ratio for PN concentration (r = 0.34, p<0.01). The results suggest that indoor PN concentration was affected by outdoor PN concentration. In contrast, a weak inverse correlation between outdoor PN concentration and the I/O ratio for PN concentration was observed (r = -0.39, p<0.01). In addition, the I/O ratio for $PM_{2.5}$ had rather weak correlation with the I/O ratio for PN concentration at the 0.05 level during the sampling period (r = 0.19, p = 0.025).

In order to better understand the source emissions/activities and interplay of indoor and outdoor PN concentrations, the particle size distribution spectra from outdoor and indoor measurements at four different times of the day on 16 February, 2006 are presented in Figure 3(a)-(d). Figure 4 shows the indoor and outdoor PN on that day. There were no indoor activities at 0:39 am (Figure 3(a)). Indoor cleaning activities started at about 7 am after the second spectrum (Figure 3(b)) and traffic began to increase at ~5 am. School hours were usually between 9 am and 3 pm. Hence, Figure 3(c) represents the time when pupils were in the classrooms and Figure 3(d) illustrates the time when pupils left the school and only cleaning staff were in the classrooms. It can clearly be seen that the modal structures of the particle number and size distributions, and the I/O ratios for different size classes at different times changed significantly, due to the differences in indoor and outdoor emission sources and activities. At midnight, the indoor and outdoor PN concentrations were low (peak value: ~2.5×10^{3} cm^{-3}) and the modal structure of indoor PN was similar to that of outdoor PN, dominated by Aitken mode particles (30-100 nm) (Figure 3(a)), suggesting the main impact of outdoor PN on indoor PN. However, the I/O ratio at midnight was equal to and even slightly higher than one for different size classes, probably owing to the weaker source emissions, easier deposition and greater dilution of outdoor PN at midnight, compared to the daytime.

Table 1 Spearman's rho correlations among indoor and outdoor PN concentrations and $PM_{2.5}$ (sample size N for all correlations = 144)

		Outdoor PM	Indoor PM	Indoor PN	Outdoor PN	I/O_PM	I/O_PN
Outdoor PM	Correlation Coefficient	1.00	0.68(**)	0.12	0.29(**)	-0.49(**)	-0.19(*)
	Sig. (2-tailed)		0.00	0.16	0.00	0.00	0.023
Indoor PM	Correlation Coefficient	0.68(**)	1.00	0.39(**)	0.43(**)	0.23(**)	-0.075
	Sig. (2-tailed)	0.00		0.00	0.00	0.005	0.37
Indoor PN	Correlation Coefficient	0.12	0.39(**)	1.00	0.66(**)	0.30(**)	0.34(**)
	Sig. (2-tailed)	0.16	.00		0.00	0.00	0.00
Outdoor PN	Correlation Coefficient	0.29(**)	0.43(**)	0.66(**)	1.00	0.11	-0.39(**)
	Sig. (2-tailed)	0.00	0.00	0.00		0.18	0.00
I/O_ PM	Correlation Coefficient	-0.49(**)	0.23(**)	0.30(**)	0.11	1.00	0.19(*)
	Sig. (2-tailed)	0.00	0.05	0.00	0.18		0.025
I/O_PN	Correlation Coefficient	-0.19(*)	-.0075	0.34(**)	-0.39(**)	0.19(*)	1.00
	Sig. (2-tailed)	0.023	0.37	0.00	0.00	0.025	

** Correlation is significant at the 0.01 level (2-tailed).
* Correlation is significant at the 0.05 level (2-tailed).

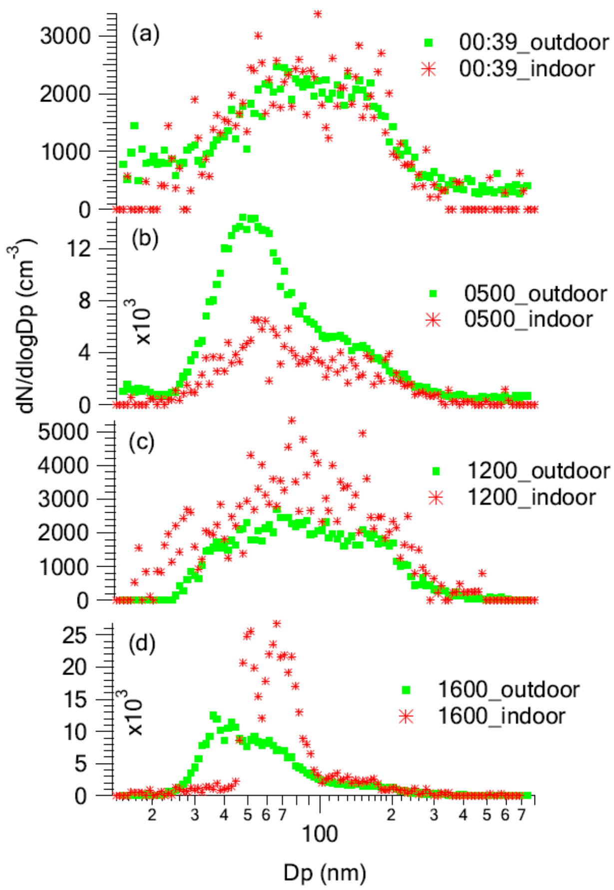


Figure 3. (a)-(d) Four different particle size distribution spectra at different times of the day.

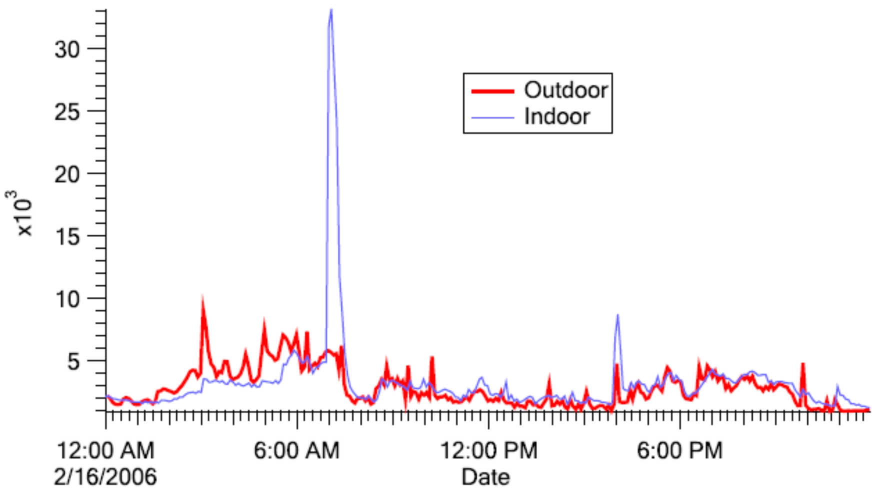


Figure 4. Total indoor and outdoor PN concentrations on 16 February 2006

In the early morning (5 am), indoor and outdoor PN concentrations increased significantly (peak values: ~7.0×10^3 cm^{-3} for indoor PN and ~1.5×10^4 cm^{-3} for outdoor PN). The modal structures of indoor and outdoor PN became more clearly defined and the count median diameter shifted to about 50 nm. The increase in outdoor PN concentration is in line with the traffic rush hours in the morning as shown in Figure 2. The count median diameter of 50 nm is coincident with that of gasoline engines (Ritowski *et al.*, 1998), perhaps suggesting the dominance of gasoline-fuelled vehicles. The average I/O ratio for different size classes was 0.46 ± 0.06, and 0.41 ± 0.04 for Aitken mode particles.

At noon, when the classroom was occupied, the indoor PN peak level ($\sim 5.0 \times 10^3$ cm^{-3}) was approximately twice the outdoor PN peak value. The modal structures broadened and the average I/O ratio for different size classes was 2.27 ± 0.46, suggesting significant indoor emissions. Further inspection found that particles with a size < 30 nm (nucleation mode) contributed most to the increased I/O ratios, perhaps implying the formation of new particles (Finlayson-Pitts and Finlayson-Pitts, 2000). As described in the section 2.1, the classroom was predominately used for art classes. Generally, the ambient hydroxyl radical (OH) and ozone (O_3) concentrations are high when the solar radiation is strong at noon. The volatile organic compounds (VOCs) emitted from the paints and glues in the art classes could react with OH and O_3 to form secondary organic aerosols. As such, a large number of particles in the lower nanometer size range would be present. Such a phenomenon was confirmed by follow-up measurements in the same classroom at noon when the art activities were carried out (Morawska *et al.*, 2009).

In the afternoon (4 pm), the cleaning activities in the classroom generated a sharp increase in indoor PN concentration for Aitken mode particles (maximum value: $\sim 2.5 \times 10^4$ cm^{-3} for particles with diameter of ~70 nm), whereas the modal structure of outdoor PN peaked at 40 nm with a value of 1.25×10^4 cm^{-3}, reflecting different indoor and outdoor sources. The highest I/O ratio was as high as 4.92 for Aitken mode particles.

3.3. Impact of occupancy on I/O ratios

Figure 5 illustrates the impact of occupancy (9am – 3pm weekdays) on the particle size distributions and the I/O ratios for different size classes. Unoccupancy was defined as the period between 10 pm – 4 am on weekdays, as well as all day on weekends. The remaining times (i.e. 3 -

10pm and 4 - 9am) were not included in either the occupancy or unoccupancy period, since the classrooms and preschool centre were often occupied by cleaners, teachers and/or parents during these hours. This kind of occupancy was different to the occupancy investigated in the study, which was the use of classrooms by children during school hours. It is noteworthy that the data points affected by indoor sources, in both the preschool centre and classroom, were removed based on questionnaires and measurement data analysis, in order to understand the interaction of indoor and outdoor PN. That is, the data points of total PN with an I/O ratio >1 were deleted. It was found that occupancy did not cause any major changes to the modal structure of the particle number and size distribution (Figure 5(1)-(2)) even though the I/O ratio was different for different size classes (Figure 5(3)-(4)). The size distributions at and after school hours were mainly dominated by Aitken mode particles, which are consistent with the results obtained by Koponen *et al.* (2001) and Hussein *et al.* (2006). Compared Figure 5(3) to Figure 5(4), it can be seen that the occupancy did not affect the relationship between I/O ratio and particle size. The I/O curves for the whole sampling period had a maximum value between particle diameters of 100 – 400 nm for both occupied and unoccupied scenarios, which suggested maximum penetration rate within the range. This observation is in line with the findings reported by Koponen *et al.* (2001) and Hussein *et al.* (2006), and agrees well with the fundamental penetration theory (Hinds, 1999). The average I/O ratios for nucleation mode particles (15 - 30 nm) were between 0.34 and 0.81 and the mean values for accumulation mode particles (102 - 737 nm) ranged from 0.90 to 0.93. The I/O ratios obtained in this study are comparable to those observed in other naturally ventilated dwellings (e.g. Thornburg *et al.*, 2001; Chao *et al.*, 2003; He *et al.*, 2005; Hussein *et al.*, 2005, 2006), but were much higher than those found in mechanically ventilated indoor environments (< 0.3, Koponen *et al.*, 2001).

In contrast, no significant difference in I/O ratio for $PM_{2.5}$ was observed between occupied (0.80 ± 0.06) and unoccupied (0.77 ± 0.04) conditions in the classrooms for the whole sampling period (Figure 6). This can be explained as follows: when the classrooms were occupied it was usually associated with the windows being opened and the air exchange rate was relatively high, leading to high I/O ratios. The elevated indoor $PM_{2.5}$ was mainly caused by infiltration of outdoor

$PM_{2.5}$ and to a lesser extent, re-suspension. When the classrooms were unoccupied, the windows were generally closed, it was night-time or very early morning and the air exchange rate was low. Under these conditions, infiltration by outdoor air was a less important process for indoor $PM_{2.5}$ and deposition was likely to be the main reason for the slow decay of indoor $PM_{2.5}$ concentrations. At the same time, outdoor $PM_{2.5}$ levels remarkably decreased at night time and in the very early morning, due to less traffic and human activities i.e. burning when compared to daytime and peak traffic hours, resulting in high I/O ratios as well. This result is consistent with other studies, which indicate that occupancy mainly leads to the increase of coarse particles in classrooms, while indoor $PM_{2.5}$ concentration is more closely associated with outdoor $PM_{2.5}$ (Branis *et al*., 2005, 2009; Fromme *et al*., 2007, 2008; Kingham *et al*., 2008).

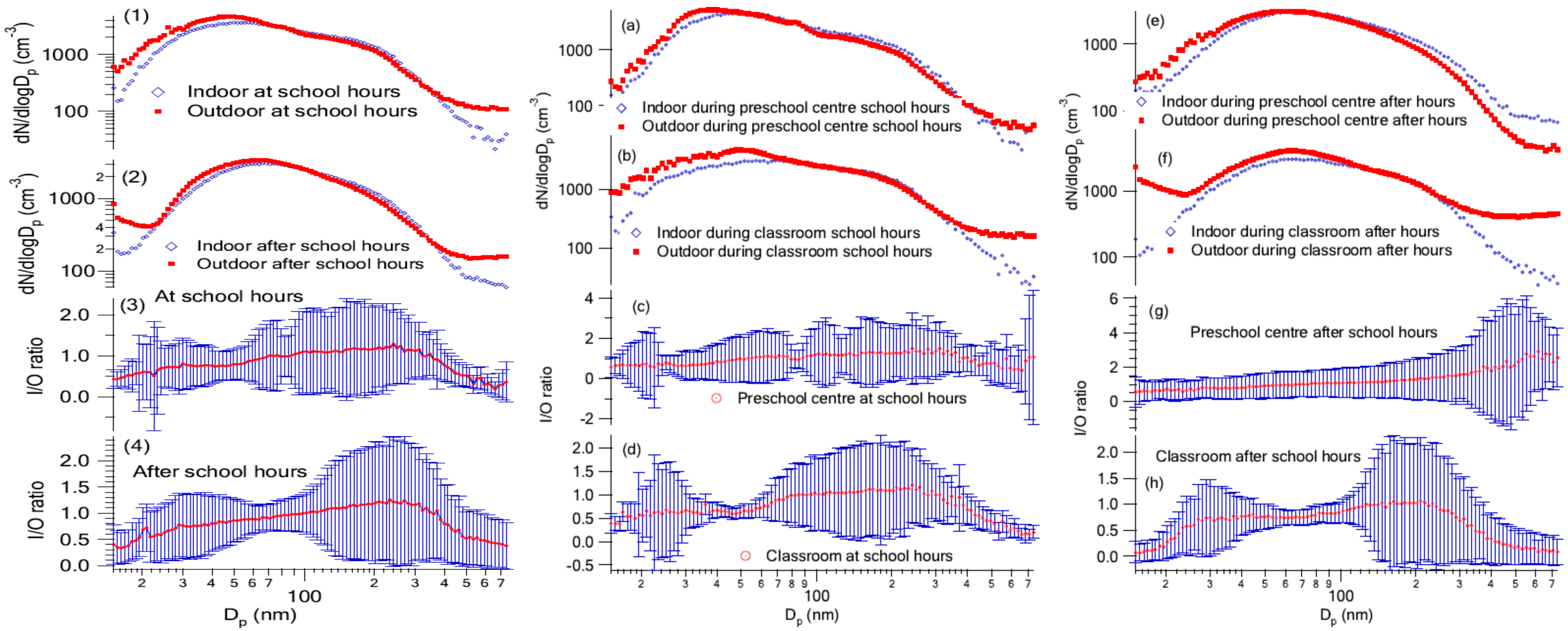


Figure 5. (1) and (2) are the average size distributions during and after school hours in both the preschool centre and classroom for the whole sampling period; (3) and (4) are the average I/O ratios at and after school hours for both the preschool centre and classroom for the whole sampling period. (a)-(d) Average size distributions and I/O ratios during the preschool centre school hours and during the classroom school hours, respectively; (e)-(h) average size distributions and I/O ratios during the preschool centre after school hours and during the classroom after school hours, respectively. The whiskers in the figures are standard deviation. The indoor sources at the two indoor sites have been removed.

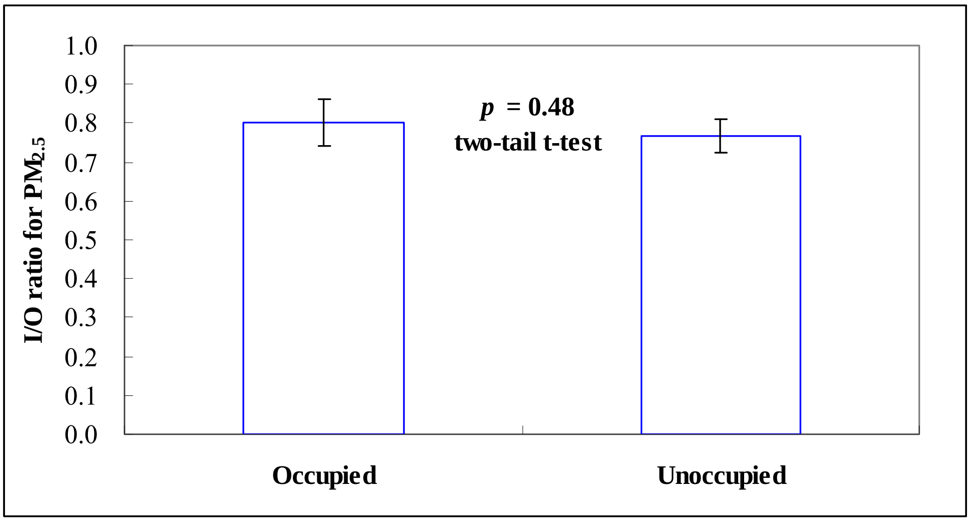


Figure 6 Effect of occupancy on I/O ratios for $PM_{2.5}$ for the whole sampling period

3.4. Spatial variation in size resolved I/O ratios

As the sampling was conducted in both a preschool centre and a classroom, it would be of interest to explore the relationship between outdoor and indoor PN concentrations in the different rooms. Figure 5 (a-h) shows the average size distributions and average size resolved I/O ratios in the preschool centre and the classroom. Figure 5(a)-(d) illustrates the situation when the students were present, and Figure 5(e)-(h) represents the situation when the buildings were unoccupied. Again, the data affected by indoor sources in both the preschool centre and classroom were removed. During school hours (i.e. 9 am – 3 pm), no obvious difference were found for size distributions between the preschool centre and the classroom (Figure 5(a)-(b)). The indoor size distributions followed the trends of outdoor particle size distributions. The modal structures were dominated by Aitken mode particles in both locations for both outdoor and indoor particles. The maximum I/O ratios in the two indoor environments were found between 100 – 400 nm (Figure 6(c)-(d)), which is consistent with previous studies (Koponen *et al.*, 2001; Chao *et al.*, 2003; Hussein *et al.*, 2006).

For after school hours (i.e. from 10 pm – 4 am on weekdays, as well as weekend days), the average size distributions in both buildings were similar to those found during school hours, however the highest particle diameter concentration shifted from 30-40 nm during school hours to 50-60 nm after school hours. Nevertheless, there was no difference in average size distribution between the preschool centre and the classroom after school hours. In contrast, the outdoor concentration of accumulation mode particles was lower than that in the preschool centre (Figure 5(e)), whereas the outdoor concentrations of nucleation mode and accumulation mode particles were much higher than those in the classroom after school hours (Figure 5(f)). The phenomena were better reflected by the size resolved I/O ratios (Figure 5(g)-(h)). In the preschool centre, the I/O ratio was the highest for accumulation mode particles at 600 nm (Figure 5(g)), which may be due to less traffic and outdoor human activities, i.e. combustion, when compared to daytime peak traffic hours, together with the fact that meteorological conditions during the pre-school measurements (8-14 February, 2006) were dramatically different from those during the classroom measurements (14-17 February, 2006). On-site observations and weather records indicated that, during field measurements at the preschool centre, it was raining on 8 February, as well asfrom 10-12 February and almost all of the rain events occurred during after school hours (i.e. very early morning and late evening) (http://www.wunderground.com/history/airport/YBBN/2006/2/8/DailyHistory.html?req_city=NA&req_state=NA&req_statename=NA). Hence, the weather conditions during the after school hours at the preschool centre were favourable for the dilution and deposition of outdoor particles, especially larger particles. In contrast, there was no rain during the sampling in the classroom, except on 15 February when it was raining during school hours (i.e. 9am – 3pm). On the other hand, a bimodal structure was observed for the I/O ratio in the classroom after school hours (Figure (h)). The average I/O ratios of both nucleation mode and accumulation mode particles were much lower than those of Aitken mode particles. The much lower than usual I/O ratios and larger variability in the smallest and the largest size ranges can be explained by increased depositional losses due to diffusion or gravitational settling during the transport of particles through the building envelope during that period. On the other hand, the highest I/O ratio and smaller variability for Aitken mode particles was due to the fact that losses from diffusion and impaction were minimal, which is consistent with the penetration theory (Hinds, 1999). These observations are in good agreement with previous findings in naturally ventilated residences (e.g. He *et al.*, 2005; Hussein *et al.*, 2005, 2006).

4. Conclusions

In this study, indoor and outdoor particle number (PN) and mass concentrations ($PM_{2.5}$) in a school were continuously monitored from 8 - 17 February and 24 January - 17 February 2006, respectively. The data indicated that indoor PN concentration was consistently affected by outdoor PN concentrations, and that the outdoor sources were likely to be attributable to vehicular emissions. However, the outdoor PN and $PM_{2.5}$ were also significantly affected by human activities i.e. cigarette smoking and the operation of a mower near the sampling site, meaning that vehicular emissions became a less significant contributor in this case. In addition, the particle size distribution spectra from outdoor and indoor measurements at different times suggested that particle size distribution was predominantly affected by emission sources and human activities such as vehicle emissions and classroom cleaning.

By analysing the I/O ratios for different size ranges of particles, it was found that particles with diameters of 100 - 400 nm had the highest I/O ratios, while particles with diameters below 30 nm or above 400 nm had lower I/O ratios, regardless of occupancy, suggesting a maximum penetration rate within the range. Although this phenomenon agrees with the fundamental penetration theory, occupancy had no influence on the I/O ratios of $PM_{2.5}$.

Inspection of the spatial impact on the I/O ratio of PN found that, in the preschool centre, the I/O ratio was the highest for accumulation mode particles at 600 nm after school hours, perhaps due to less traffic and outdoor human activities; and wet weather conditions during that period were favourable to the dilution (deposition) of outdoor particles, especially large particles. In the classroom, however, a bimodal structure was observed for the I/O ratio after school hours. The average I/O ratio of Aitken mode particles was much higher than that of nucleation mode and accumulation mode particles, suggesting that different mode particles had different depositional losses caused by diffusion or gravitational settling.

Nevertheless, the significance of the data is to some extent limited by the selection of only one site and a relatively short period of measurements. Therefore, further investigation is necessary to fully establish relevance to other settings, including different locations and seasons.

5. Recommendations and Perspectives

The findings obtained in this study are useful for epidemiological studies to estimate the total personal exposure of children, and to develop appropriate control strategies for minimizing the adverse health effects on school children.

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