Queensland University of Technology
School of Physical and Chemical Sciences

AIRBORNE PARTICLES IN INDOOR RESIDENTIAL ENVIRONMENT: SOURCE CONTRIBUTION, CHARACTERISTICS, CONCENTRATION, AND TIME VARIABILITY

Congrong He

Bachelor of Science – Atmospheric Physics
(Nanjing University, P.R.China)
Master of Science – Environmental Science
(Murdoch University, Australia)

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KEYWORDS

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ABSTRACT

The understanding of human exposure to indoor particles of all sizes is important to enable exposure control and reduction, but especially for smaller particles since the smaller particles have a higher probability of penetration into the deeper parts of the respiratory tract and also contain higher levels of trace elements and toxins. Due to the limited understanding of the relationship between particle size and the health effects they cause, as well as instrument limitations, the available information on submicrometer (d < 1.0 µm) particles indoors, both in terms of mass and number concentrations, is still relatively limited.

This PhD project was conducted as part of the South-East Queensland Air Quality program and Queensland Housing Study aimed at providing a better understanding of ambient particle concentrations within the indoor environment with a focus on exposure assessment and control. This PhD project was designed to investigate comprehensively the sources and sinks of indoor aerosol particles and the relationship between indoor and outdoor aerosol particles, particle and gaseous pollutant, as well as the association between indoor air pollutants and house characteristics by using, analysing and interpreting existing experimental data which were collected before this project commenced, as well as data from additional experiments which were designed and conducted for the purpose of this project. The focus of this research was on submicrometer particles with a diameter between 0.007 – 0.808 µm. The main outcome of this project may be summarised as following:

- A comprehensive review of particle concentration levels and size distributions characteristics in the residential and non-industrial workplace environments was conducted. This review included only those studies in which more general
trends were investigated, or could be concluded based on information provided in the papers. This review included four parts: 1) outdoor particles and their effect on indoor environments; 2) the relationship between indoor and outdoor concentration levels in the absence of indoor sources for naturally ventilated buildings; 3) indoor sources of particles: contribution to indoor concentration levels and the effect on I/O ratios for naturally ventilated buildings; and 4) indoor/outdoor relationship in mechanically ventilated buildings.

- The relationship between indoor and outdoor airborne particles was investigated for sixteen residential houses in Brisbane, Australia, in the absence of operating indoor sources. 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 showed 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 and in the absence of operating indoor sources, outdoor particle concentrations could be used to predict instantaneous indoor particle concentrations but not for minimum ventilation, unless air exchange rate is known, thus allowing for estimation of the “delay constant”.

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Diurnal variation of indoor submicrometer particle number and particle mass (approximation of PM$_{2.5}$) concentrations was investigated in fifteen of the houses. The results show that there were clear diurnal variations in both particle number and approximation of PM$_{2.5}$ concentrations, for all the investigated houses. The pattern of diurnal variations varied from house to house, however, there was always a close relationship between the concentration and human indoor activities. The average number and mass concentrations during indoor activities were $(18.2\pm3.9) \times 10^3$ particles cm$^{-3}$ and $(15.5\pm7.9)$ µg m$^{-3}$ respectively, and under non-activity conditions, $(12.4\pm2.7) \times 10^3$ particles cm$^{-3}$ $(11.1\pm2.6)$ µg m$^{-3}$, respectively. In general, there was a poor correlation between mass and number concentrations and the correlation coefficients were highly variable from day to day and from house to house. This implies that conclusions cannot be drawn about either one of the number or mass concentration characteristics of indoor particles, based on measurement of the other. The study also showed that it is unlikely that particle concentrations indoors could be represented by measurements conducted at a fixed monitoring station due to the large impact of indoor and local sources.

Emission characteristics of indoor particle sources in fourteen residential houses were quantified. In addition, characterizations of particles resulting from cooking conducted in an identical way in all the houses were measured. All the events of elevated particle concentrations were linked to indoor activities using house occupants diary entries, and catalogued into 21 different types of indoor activities. This enabled quantification of the effect of indoor
sources on indoor particle concentrations as well as quantification of emission rates from the sources. For example, the study found that frying, grilling, stove use, toasting, cooking pizza, smoking, candle vaporizing eucalyptus oil and fan heater use, could elevate the indoor submicrometer particle number concentration levels by more than 5 times, while PM$_{2.5}$ concentrations could be up to 3, 30 and 90 times higher than the background levels during smoking, frying and grilling, respectively.

- Indoor particle deposition rates of size classified particles in the size range from 0.015 to 6 $\mu$m were quantified. Particle size distribution resulting from cooking, repeated under two different ventilation conditions in 14 houses, as well as changes to particle size distribution as a function of time, were measured using a scanning mobility particle sizer (SMPS), an aerodynamic particle sizer (APS), and a DustTrak. Deposition rates were determined by regression fitting of the measured size-resolved particle number and PM$_{2.5}$ concentration decay curves, and accounting for air exchange rate. The measured deposition rates were shown to be particle size dependent and they varied from house to house. The lowest deposition rates were found for particles in the size range from 0.2 to 0.3 $\mu$m for both minimum (air exchange rate: 0.61±0.45 h$^{-1}$) and normal (air exchange rate: 3.00±1.23 h$^{-1}$) ventilation conditions. The results of statistical analysis indicated that ventilation condition (measured in terms of air exchange rate) was an important factor affecting deposition rates for particles in the size range from 0.08 to 1.0 $\mu$m, but not for particles smaller than 0.08 $\mu$m or larger than 1.0 $\mu$m. Particle coagulation was assessed to be negligible compared to the two other processes
of removal: ventilation and deposition. This study of particle deposition rates, the largest conducted so far in terms of the number of residential houses investigated, demonstrated trends in deposition rates comparable with studies previously reported, usually for significantly smaller samples of houses (often only one). However, the results compare better with studies which, similarly to this study, investigated cooking as a source of particles (particle sources investigated in other studies included general activity, cleaning, artificial particles, etc).

- Residential indoor and outdoor 48 h average levels of nitrogen dioxide (NO₂), 48h indoor submicrometer particle number concentration and the approximation of PM₂.₅ concentrations were measured simultaneously for fourteen houses. Statistical analyses of the correlation between indoor and outdoor pollutants (NO₂ and particles) and the association between house characteristics and indoor pollutants were conducted. The average indoor and outdoor NO₂ levels were 13.8 ± 6.3 ppb and 16.7 ± 4.2 ppb, respectively. The indoor/outdoor NO₂ concentration ratio ranged from 0.4 to 2.3, with a median value of 0.82. Despite statistically significant correlations between outdoor and fixed site NO₂ monitoring station concentrations (p = 0.014, p = 0.008), there was no significant correlation between either indoor and outdoor NO₂ concentrations (p = 0.428), or between indoor and fixed site NO₂ monitoring station concentrations (p = 0.252, p = 0.465.). However, there was a significant correlation between indoor NO₂ concentration and indoor submicrometer aerosol particle number concentrations (p = 0.001), as well as between indoor PM₂.₅ and outdoor NO₂ (p = 0.004). These results imply that the outdoor or
fixed site monitoring concentration alone is a poor predictor of indoor NO₂ concentration.

- Analysis of variance indicated that there was no significant association between indoor PM₂.₅ and any of the house characteristics investigated ($p > 0.05$). However, associations between indoor submicrometer particle number concentration and some house characteristics (stove type, water heater type, number of cars and condition of paintwork) were significant at the 5% level. Associations between indoor NO₂ and some house characteristics (house age, stove type, heating system, water heater type and floor type) were also significant ($p < 0.05$). The results of these analyses thus strongly suggest that the gas stove, gas heating system and gas water heater system are main indoor sources of indoor submicrometer particle and NO₂ concentrations in the studied residential houses.

The significant contributions of this PhD project to the knowledge of indoor particle included: 1) improving an understanding of indoor particles behaviour in residential houses, especially for submicrometer particle; 2) improving an understanding of indoor particle source and indoor particle sink characteristics, as well as their effects on indoor particle concentration levels in residential houses; 3) improving an understanding of the relationship between indoor and outdoor particles, the relationship between particle mass and particle number, correlation between indoor NO₂ and indoor particles, as well as association between indoor particle, NO₂ and house characteristics.
LIST OF PUBLICATIONS


He, C., Morawska, L., Mengersen, K., Gilbert, D., 2004. The Effect of Indoor and Outdoor Sources and House Characteristics on Indoor Airborne Particles and NO₂. Submitted to *Environmental Science & Technology* (September 2004)
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STATEMENT OF ORIGINAL AUTHORSHIP

The work contained in this thesis has not been previously submitted for a degree or diploma at any other tertiary educational institution. To the best of my knowledge and belief, the thesis contains no material previously published or written by another person except where due reference is made.

Signed:____________________

Date:   ____________________
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CHAPTER 1. GENERAL INTRODUCTION

1.1 DESCRIPTION OF THE SCIENTIFIC PROBLEM INVESTIGATED

A number of epidemiological studies have linked daily mortality and morbidity statistics with increased particle concentrations measured outdoors (Pope et al., 1992; Dockery et al., 1993; Schwartz et al., 1996; Pope, 2000). Although studies have often included other outdoor gaseous pollutants, such as sulphur dioxide, sulphates, carbon monoxide, and nitrogen oxides, the strongest relationships have generally been found to be with particles (Chock and Winkler, 2000; Wallace, 2000; Schwartz, 2001).

Human exposure to aerosol particles takes place in outdoor and indoor environments. However, since people spend approximately 90% (95% in Australia, ABS 1996) of their time indoors (Fishbein and Henry, 1991; Jenkins et al., 1992; Byrne, 1998) and indoor particle concentrations often exceed outdoor concentrations (Yocom, 1982; Wallace, 1996; Monn, 2001), indoor exposure is the major contributor to total personal exposure (Janssen et al., 1998). Accordingly, concern over the health effects of indoor particles is increasing (Tuckett et al., 1998; Jones, 1999; Williams et al., 2000). Recent studies have also suggested that number concentration could be a better predictor than mass concentration for the health effects of particles (Oberdörster et al., 1992; Oberdörster, 1995; Oberdörster et. al., 1995, Wichmann and Peters 2000).

Studies found in existing literature provided information and background knowledge about indoor particles; however, due to the limited understanding of the relationship
between particle size and resultant health effects, as well as instrumental limitations, the majority of measurements of aerosol particles in previous studies were in terms of mass concentrations and coarse particles, such as total suspended particles (TSP), PM$_{10}$, PM$_{3.5}$ and PM$_{2.5}$ fraction. As a result, the available information for submicrometer (< 1.0 µm) particles indoors involving both mass concentration and number concentration is limited.

Characterisation of indoor particles is complex and requires consideration of their origin, sink, temporal and spatial variation, and dynamics. In addition, the indoor environment can be subdivided into different microenvironments with examples including: office, school, shopping centre, restaurant and residential houses. Within residential houses further distinction can be made between kitchen, living room, bedroom and bathroom areas. The characteristics of indoor particles can be indoor microenvironment specific; however, information on this subject is still relatively limited, especially for submicrometer particles in residential houses.

Based on the results of extensive literature review (see Chapter 2), it is hypothesised that there may be a relationship between indoor and outdoor particle characteristics in residential houses because the air exchange between indoor and outdoor. It is also hypothesised that indoor particle concentration may be diurnal variation and affected by indoor activity, indoor particle emission rate and deposition rate may be houses-specific because human indoor activities may be houses-specific. It is further postulated that there may be multiple correlation between indoor air pollutants, and association between indoor air pollutants and the house characteristics.
1.2 QUEENSLAND HOUSING PROGRAM

The South-East Queensland Air Quality program and Queensland Housing Study that started in July 1997, before this PhD project was undertaken, have generated:

- Determination of horizontal particle number concentration profiles in relation to a road,
- Determination of vertical particle number concentration profiles around building envelope,
- A large body of data from indoor/outdoor measurements of particle and gases characteristics as well as house characteristics.

This body of data and additional experimental data (including data collected from the EPA monitoring stations), were assessed, analysed, and interpreted, with the results forming an important component of this PhD study.

1.3 OVERALL OBJECTIVES OF THE STUDY

In order to investigate the above mentioned hypotheses and to enhance understanding of indoor particle characteristics, the overall objective of this PhD study was to comprehensively investigate indoor particle characteristics, with a particular focus on submicrometer particles and the relationship between indoor and outdoor particles. The intent of this investigation was to provide scientific understanding of indoor particle sources, sinks and dynamics, thus advancing knowledge related to exposure assessment and improving indoor air quality.
1.4 THE SPECIFIC AIMS OF THIS STUDY

The specific aims of this project were to:

- Comprehensively review literature data of indoor particle concentration levels and size distributions characteristics,
- Investigate and interpret the relationship between indoor and outdoor particle characteristics in residential houses,
- Investigate and interpret indoor particle variation behaviour,
- Investigate and interpret indoor particle source characteristics and their effect on indoor particle concentration levels,
- Investigate and interpret indoor particle deposition characteristics,
- Conduct multiple correlation analysis and interpret the analysis results.

1.5 ACCOUNT OF SCIENTIFIC PROGRESS LINKING THE SCIENTIFIC PAPERS

This thesis is presented as a collection of six papers that have been published or submitted for publication in refereed journals or edited books.

The results of a comprehensive review of literature concerning particle concentration levels and size distributions characteristics in the residential and non-industrial workplace environments were presented in the second paper (Chapter 3). This review highlighted four aspects of this body of work: outdoor particles and their effect on indoor environments, the relationship between indoor and outdoor concentration levels in the absence of indoor sources for naturally ventilated
buildings, indoor sources of particles in both their contribution to indoor concentration levels and effect on indoor to outdoor ratios for naturally ventilated buildings, and the indoor/outdoor relationship in mechanically ventilated buildings.

Residential houses do not normally use air filtration systems in Brisbane, Australia. This means that the principal factors governing the levels of airborne particles indoors are indoor sources, outdoor particle levels, the deposition rate of particles on indoor surfaces, and the air exchange rate. Thus, the following papers investigated these factors in residential houses.

The studies presented in the first paper (Chapter 4), concentrated on investigation of 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. This residential study was conducted for cases when there were no indoor activities performed that would result in particle generation (such as cooking, smoking, dusting and vacuuming) in the houses. The study also aimed to analyse trends in the relationship between indoor and outdoor particles for different ventilation conditions in the investigated houses, as well as to draw conclusions on the applicability of outdoor particle concentration data for indoor exposure assessment. The study revealed that while temporary values of the ratio vary over a broad range according to ventilation conditions, average values of the ratios were very close to unity regardless of ventilation conditions and particle size range when there were no indoor particle sources. The time series of indoor to outdoor particle concentrations showed a clear positive relationship existing for many houses under normal ventilation conditions, but not under minimum ventilation conditions. These results
suggest that for normal ventilation conditions, outdoor particle concentrations could be used to predict instantaneous indoor particle concentrations. This cannot be applied to the condition of minimum ventilation unless the air exchange rate is known, allowing for estimation of the “delay constant”.

In the third paper (Chapter 5), diurnal variation of indoor submicrometer (0.007 – 0.808 µm) particle number and particle mass (approximation of PM$_{2.5}$) concentrations were investigated during winter in 1999. The results indicated clear diurnal variations, in both particle number and approximation of PM$_{2.5}$ concentrations, for all the investigated houses. The pattern of diurnal variations was house-specific; however, there was always a close relationship between the concentration and human indoor activities. The average number and mass concentrations during periods of indoor activity were higher than those for non-activity conditions. In general, there was a poor correlation between mass and number concentrations and the correlation coefficients were highly variable from day to day and from house to house. This result implied that conclusions cannot be drawn about either one of the number or mass concentration characteristics of indoor particles, based on measurement of the other. The study also showed that it is unlikely that particle concentrations indoors could be inferred from measurements conducted at a fixed monitoring station due to the large impact of indoor and local sources.

To develop insight into indoor particle sources, the fourth paper (Chapter 6) quantified indoor particle emission characteristics. All the incidences of elevated particle concentrations were linked to indoor activities using house occupants’ diary entries, and catalogued into 21 different types of indoor activities. This enabled
quantification of the effect of indoor sources on indoor particle concentrations as well as quantification of emission rates from the sources. The study found that frying, grilling, stove use, toasting, cooking pizza, smoking, candle vaporizing eucalyptus oil and fan heater use, could elevate the indoor submicrometer particle number concentration levels by more than 5 times, while PM$_{2.5}$ concentrations could be up to 3, 30 and 90 times higher than the background levels during smoking, frying and grilling, respectively.

Indoor particle sink characteristics were quantified in the fifth paper (Chapter 7). Deposition loss rates were determined by regression fitting of the measured size-resolved (0.015 – 6 µm) concentration decay curves and PM$_{2.5}$ concentration decay curves, and then subtracting the air exchange rate. The lowest deposition rates were found for particles in the size range from 0.2 to 0.3 µm for both minimum and normal ventilation conditions. The results of statistical analysis indicated that ventilation condition (measured in terms of air exchange rate) was an important factor affecting deposition rates for particles in the size range from 0.08 to 1.0 µm, but not for particles smaller than 0.08 µm or larger than 1.0 µm. Particle coagulation was assessed and the resultant deposition rates were compared with literature data, with the details presented in this Chapter.

The final paper (Chapter 8) presented the results of multiple analyses of the indoor and outdoor NO$_2$ and particle concentrations. This study found that despite statistically significant correlations between outdoor and fixed site NO$_2$ monitoring station concentrations, there was no significant correlation between either indoor and outdoor NO$_2$ concentrations, or between indoor and fixed site NO$_2$ monitoring station concentrations. However, there was a significant correlation between indoor
NO₂ concentration and indoor submicrometer aerosol particle number concentrations, as well as between indoor PM$_{2.5}$ and outdoor NO₂ ($\rho = 0.004$). These results again suggested that the outdoor or fixed site monitoring concentration alone was a poor predictor of indoor NO₂ concentration. The results of analysis of variance strongly suggested that gas appliances were a more significant indoor source than smoking, and were the main indoor sources of indoor submicrometer particle and NO₂ concentrations in the residential houses studied.

Throughout this thesis, correlations and other measures of association are computed and interpreted as suggestive of physical pathways between variables of interest. It is acknowledged that a correlation coefficient only measures the degree of linear association between two variables, not a causal relationship. Further studies are required to confirm the suggested inferences.

REFERENCES


2.1. INTRODUCTION

Exposure to air pollutants contributes to human health risk and is an area in which a risk-reduction approach should be taken. Air pollutants can generally be classified into two types: gaseous pollutants, such as sulphur dioxide (SO₂), nitrogen dioxide (NO₂), ambient ozone (O₃), and volatile organic compounds (VOC); and particle pollutants.

Aerosol particles are defined as relatively stable suspensions of solid or liquid particles in a gas. Gaseous pollutants may be characterised by mass concentration alone; however, this is not the case for aerosol particles. Due to the complexity of aerosol particles and the importance of particle size in determining human exposure, there are multiple characteristics that must be considered. Aerosol particles are characterised not only by mass concentration but also number, surface area and volume concentrations. Furthermore, they could be sub-characterised by size, for example total suspended particles (TSP, mass concentration of particles with aerodynamic diameters between 0.1 and 100 µm), PM₁₀ fraction (mass concentration of particles with aerodynamic diameters smaller than 10 µm), or PM₂.₅ fraction (mass concentration of fine particles with aerodynamic diameters smaller than 2.5 µm). Similarly, aerosol particle number concentrations are also sub-characterised by size, such as supermicrometer particle number concentration (larger than 1 µm), submicrometer particle number concentration (smaller than 1 µm) and ultrafine particle number concentration (smaller than 0.1 µm). In addition, aerosol particles are also characterised by their chemical compositions and biological properties.
The fundamental properties of aerosols have been studied for many years. Physical investigations of aerosols were the domain of the early period of aerosol science. The results of early studies on aerosols provided the basic background to further developments. Greater practical interest in the properties of, and processes in, aerosol chemistry began during the 1950s (Spurny, 2000). However, only in the last decade has greater progress in environmental aerosol science become possible with the introduction of advanced new instrumental techniques.

The health effects of airborne particles have been studied since the 1950’s. A number of epidemiological studies have linked daily mortality and morbidity statistics with increased particle concentrations measured outdoors (Pope et al., 1992; Dockery et al., 1993; Schwartz et al., 1996; Schwartz, 2001). Although the studies have often included other outdoor gaseous pollutants, such as sulphur dioxide, sulphates, carbon monoxide, and nitrogen oxides, the strongest relationships have generally been found to be with particles (Chock and Winkler, 2000; Wallace, 2000). Smaller particles have a higher probability of penetration into the deeper parts of the respiratory tract (James et al., 1991; Owen et al., 1992; Berico et al., 1997) and also contain higher levels of trace elements and toxins such as polycyclic aromatic hydrocarbons and mutagens (Ando et al., 1996; Kiss et al., 1998; Phousongphouang and Arey, 2003). Accordingly, fine particle fractions (smaller than 2.5 µm) and submicrometer particle fractions (smaller than 1.0 µm) have been the subject of increasing concern. Schwartz et al. (1996) reported that daily mortality was more strongly associated with concentrations of fine particles (PM$_{2.5}$) than with concentrations of larger particles (coarse mass PM$_{10}$) in six U.S. cities. Klemm et al. (2000) validated the findings of Schwartz et al. (1996) by reconstruction and reanalyses of the original data from the 1996 study.
Like outdoor particles, characterisation of indoor particles is complex and requires consideration of their origin, sink, and both temporal and spatial variation. In addition, the indoor environment can be subdivided into different microenvironments, such as: office, school, shopping centre, restaurant and residential house. The characteristics of indoor particles can also be indoor microenvironment specific. However, since the residential house is the main indoor microenvironment in which people, especially aged people and young children, spend their time, the majority of studies focus on indoor aerosol particles in residential environments (Yocom, 1982; Thatcher and Layton, 1995; Wallace, 1996; Monn, 2001).

2.2 AIRBORNE PARTICLES IN INDOOR RESIDENTIAL ENVIRONMENTS

Residential houses do not normally use air filtration systems. This means that the principal factors governing the levels of airborne particles indoors are: indoor sources, outdoor particle levels, the deposition rate of particles on indoor surfaces, and the air exchange rate (Shair and Heitner, 1974; Kamens et al., 1991; Thatcher and Layton, 1995). Figure 2-1 illustrates the principal factors governing the concentration levels of airborne particles in residential houses. A mass conservation equation for figure 2-1 can be written as,

\[
\frac{dC_{in}}{dt} = P\alpha C_{out} + \frac{Q_s}{V} - (\alpha + k)C_{in}
\]

(2-1)

where \(C_{in}\) and \(C_{out}\) are the indoor and outdoor particle concentrations, respectively; \(P\) is the penetration efficiency; \(\alpha\) is the air exchange rate; \(k\) is the deposition rate; \(Q_s\) is the indoor particle generation rate; \(t\) is time; and \(V\) is the volume of the house.
This equation ignores the effects of condensation, evaporation, and coagulation since these are minor effects under the concentrations and conditions normally found in the residential environment (Thatcher and Layton, 1995).

Figure 2-1. Schematic diagram of the principal factors governing the levels of airborne particles in residential houses.

Although the relationship between indoor particle concentrations and their controlling factors could be expressed by the mass conservation equation, it is very difficult to obtain the assumed conditions (i.e. a high degree of mixing in the indoor air and steady controlling factors, such as $P$, $\alpha$, $k$ and $Q_s$) to apply the equation in real residential microenvironment situations. These factors are usually investigated by purpose designed studies and have been addressed individually or in groups by a variety of studies (Nazaroff and Cass, 1989; Fogh et al., 1997; Abt et al., 2000; Wallace et al., 2004). However, there is still a lack of experimental results to confirm the validity of the predictions of the models.
2.2.1 Outdoor sources

Outdoor particles can be an important source affecting indoor particle concentrations. Particles produced outdoors can enter residential houses through windows, doors, cracks or other ventilation systems. The level of the contribution of outdoor particles to indoor particle concentrations depends primarily on: the characteristics of outdoor particles (e.g. concentrations, size), the characteristics of the house and meteorologic conditions. The latter two affect the air exchange rate and penetration efficiencies, which will be discussed in section 2.2.4.

Outdoor aerosol particles have been studied for more than half a century and are considered a significant atmospheric air pollutant. As such, they have been regulated in most countries (Chow, 1995). Substantial data and scientific understanding in relation to the epidemiology, toxicology, physics and chemistry of coarse atmospheric particulate pollutants have been obtained to develop management and mitigation strategies for these particles. As a result, the concentration levels of outdoor coarse particles have been significantly reduced in many parts of the world today, compared with years ago. However, information about submicrometer particles, particularly ultrafine particles, is still limited.

Outdoor particle concentrations are strongly affected by outdoor particle emission sources and meteorological parameters. Outdoor aerosol is a complex mixture of particle size and type that originates from many sources. Outdoor particles can be generated by natural processes, such as wind action and sea spray, or they may be man made, as seen with industrial and motor vehicles emissions. In terms of size, ambient particles are generally distributed bimodally. It is generally agreed that fine particles (< 2.5 µm) are usually created as primary particles by the source, via
combustion or other high-temperature processes (Ohlström et al., 2000), or as secondary particles in the atmosphere formed from gaseous pollutants such as: condensable organic compounds, sulphur oxides, nitrogen oxides, and ammonia (Strader et al., 1999). Most of the mass of coarse particles (> 2.5 µm) is derived from materials that have been ground down by mechanical processes.

Uncertainty remains with respect to the origins of fugitive particle emissions, those particles that enter the atmosphere without first passing through a confined flow stream. Their emission rates are poorly quantified, but potentially large compared to the emission rates of combustion and other industrial sources (Tucker, 2000).

2.2.2 Indoor sources

Since indoor particle sources play an important role in indoor particle characteristics and affect indoor particle concentrations directly and immediately, it is significant that occupant exposure to indoor particles from indoor sources be quantified as an important step toward assessing their role in health risk.

A great number of indoor particle sources, such as: smoking, kerosene heaters, wood stoves, humidifiers, electric motors, cooking, resuspension of particles, people and pets, and showering, have been identified by previous indoor particle studies (Owen et al., 1992; Moriske et al., 1996; Tucker, 2000). Previous studies indicate that indoor sources could be short term, seasonal or continuous, with the most significant being environmental tobacco smoke and fumes from cooking, kerosene heating and wood burning stoves (Tuckett et al., 1998; Dennekamp et al., 2001). Other human activities such as dusting and vacuuming can also significantly contribute to elevated particle concentration levels indoors (Spengler et al., 1981; Monn et al., 1995; Ross et al., 1999).
Combustion sources are the main indoor generators of fine mode particles, which contain a range of organic and inorganic material (Maroni et al., 1995). Other indoor sources, such as gas-to-particle conversion, sprays and biological contaminants, can also contribute to the fine mode of indoor particles. Resuspension by human activity indoors can be the main contributor to the coarse mode of indoor particles. For example, in a study by Kleeman et al. (1999), size and composition distribution of fine particle sources from wood burning, meat charbroiling, and cigarettes were measured. It was found that wood burning and cigarettes both showed a single mode by mass with peaks at 0.1-0.2µm and 0.3-0.4µm particle diameters, respectively. The smoke from meat charbroiling shows a major peak in the particle mass distribution at 0.1-0.2µm, with some material present at larger particle sizes. In addition, these sources are largely composed of organic compounds.

Secondary particles in the indoor environment may also be formed from gaseous pollutants. Weschler and Shields (1999) reported the effects of reactions between ozone and selected terpenes on the concentrations and size distributions of airborne particles in a typical indoor setting (unoccupied offices). Their results demonstrated that ozone/terpene reactions could be a significant source of submicrometer particles in indoor settings.

Since it is very difficult to accurately measure indoor particle emission rates in real situations, information about emission rates is very limited. The emission rates associated with tobacco smoking have attracted attention and as a result, there is much information available on these rates in the literature. Brauer et al. (2000) measured the emission rate of tobacco smoking (1.67 mg/min) by a method involving nephelometer measurements and known air exchange rates. Their results
were comparable to the results (14-22 mg/cigarette) of Ott et al. (1996). The estimates for cooking ranged from a low of 1.5±0.6 mg/min to a high of 4.9±1.3 mg/min. The estimates for other indoor sources ranged from 2.5±0.9 mg/h to 12±4 mg/h (Wallace, 1996).

2.2.3 Particle sinks

In indoor environments, any surface in a room can act as a sink for particles. Particle deposition rate and air exchange rate are the two main components of overall particle removal rate from the air. The former represents the process whereby indoor particles are deposited on indoor surfaces by turbulent diffusion or other mechanisms (e.g. gravitation), while the latter represents the process by which indoor particles leave the house through exfiltration. In general, diffusive deposition is the predominant removal mechanism for submicrometer particles in the air, while gravitational deposition is prevalent for supermicrometer particles.

Particle deposition rates are extremely difficult to measure directly. As a result, only limited observational data exist on these rates and their variability within residential houses, particularly for short time periods and for discrete particle sizes (Long et al., 2001). Literature data showed several important points: it is clear that the deposition process is strongly dependent on particle size, reaching a minimum in the range of particle diameter between 0.1 to 0.3 µm (Lai, 2002); the trend in the deposition rate as a function of particle size is the same for all the studies, with a significant variation often by an order of magnitude or more, in the coefficient values reported by individual studies; while both modelling and experiments predict similar trends, the model estimates could be significantly different from those derived from the
experimental results, particularly for particles smaller than about 0.5 \( \mu \text{m} \) (Morawska and Salthammer, 2003).

### 2.2.4 Air exchange rate and penetration efficiency

Air exchange rates (AER) have been investigated by many researchers around the world and the literature data indicate that they are affected by house design and material characteristics as well as by meteorological conditions and building operational factors (Kvisgaard and Collet, 1990; Nantka, 1990; Nakai et al., 1993).

In the subtropical environment of Brisbane, under normal ventilation conditions, the air exchange rates range between about 2 h\(^{-1}\) and 5 h\(^{-1}\). Minimum ventilation conditions refer to periods when the AER was between about 0.5 h\(^{-1}\) and 1.0 h\(^{-1}\) (Morawska and Jamriska, 1994).

Penetration efficiency is the fraction of outdoor particles that penetrate the house shell. Thatcher and Layton (1995) used different modelling techniques to estimate penetration efficiencies. However, for both techniques employed, the estimated penetration efficiencies were found to be close to unity, independent of particle size. Furthermore, Wallace (1996) reported penetration efficiencies very close to unity for both fine and coarse particles. It should be noted that some studies reported inconsistent results. For example, Long et al., (2001) found that the penetration efficiencies were not a unit and showed strong size dependence.

### 2.2.5 Concentration and characteristics of indoor particles

The concentration of indoor particles is highly variable and house-specific. For example, in Bangkok, Thailand, Tsai et al. (2000) found that the mean 24h average indoor PM\(_{10}\) and PM\(_{2.5}\) concentrations measured in 12 homes ranged from 154 to
245 µg m$^{-3}$ and from 64 to 184 µg m$^{-3}$, respectively. The results of Tsai et al., (2000) are significantly higher than those of Jones et al., (2000), who found that in Birmingham, UK, the mean 24h average indoor PM$_{10}$ concentrations in 9 homes ranged from 20±30 to 88±97 µg m$^{-3}$ and PM$_{2.5}$ concentration was 7.9±5.2 µg m$^{-3}$. The variation pattern is affected by human activity, with the most important activities shown to be smoking and cooking. Homes with smokers have exhibited particle mass concentrations approximately three times higher than those observed in non-smoking residences (Jones, 1999). During cooking, Pandey et al. (1989) found that particle mass concentrations indoors were as high as 21,000 µg m$^{-3}$. This implies that short-term particle mass concentrations during periods of cooking would be significantly higher than background levels or during non-cooking periods of time.

As discussed before, there are several types of indoor particles that can be produced indoors and outdoors: combustion and mineral aerosols, plant and animal bioaerosols, and home/personal care aerosols. Owen et al., (1992) summarised the results of early literature research into the sources, sizes and concentrations of indoor particles.

**2.2.6 The relationship between particle characteristics and characteristics of other air pollutants**

Unlike outdoor aerosol research, studies on the relationship between indoor particle characteristics and the characteristics of other indoor air pollutants are scarce and available data are limited to measurements of particle mass and NO$_2$. Monn et al. (1997) measured indoor and outdoor PM$_{10}$, PM$_{2.5}$ and NO$_2$ in 17 homes in Switzerland. In 10 of these homes, the inhabitants also participated in conducting
personal measurements. They found that the correlation between NO$_2$ and PM$_{10}$ levels was lower for indoor and personal levels than that for outdoor levels. This indicated that NO$_2$ would not be a very good surrogate for PM$_{10}$ for a study based on individuals. Simoni et al. (1998) examined indoor PM$_{2.5}$ and NO$_2$ in 140 homes in the Po River Delta area of North Italy during summer and winter. A significant relationship between indoor NO$_2$ concentrations and PM$_{2.5}$ values in both seasons was found. However, there is no literature data available regarding the relationship between indoor submicrometer particles and any other indoor air pollutants.

2.3. GAPS IN KNOWLEDGE

2.3.1 Submicrometer particles

The first attempts to study indoor particles date back to prior to the 1900s, but the majority of studies on indoor particles were conducted after World War II (Spurny, 1999). Previous studies provided information and background knowledge about indoor particles; however, due to the limited understanding of the relationship between particle size and the health effects they cause, as well as instrumental limitations, the majority of measurements of aerosol particles in previous studies were in terms of mass concentrations and considered only coarse particles, such as total suspended particles (TSP), PM$_{10}$, and PM$_{3.5}$ and PM$_{2.5}$ fractions. As a result, the available information for submicrometer particles indoors involving both mass concentration and number concentration is very limited.

As mentioned above, there is mounting evidence indicating that the most harmful particles are likely to be fine particles with diameters smaller than 2.5 $\mu$m and submicrometer particles with diameters smaller than 1.0 $\mu$m (Schwartz et al., 1996;
Simpson et al, 2000). This is due to the fact that these particles usually contain high concentrations of trace elements and toxins, such as the polycyclic aromatic hydrocarbons and certain mutagens (Ando et al., 1996; Kiss et al., 1998; Goto et al., 1999), and have a high probability of deposition deeper in the respiratory tract (James et al., 1991; Owen et al., 1992; Berico et al., 1997). Ferin et al. (1992) found that ultrafine titanium dioxide aerosols had an adverse effect on the respiratory systems of rats; yet when the rats were exposed to the same mass concentration of micrometer particles there was no effect. In Brisbane, Australia, Simpson et al. (1997) found that there was a significant association between daily mortality and fine particle concentrations, and Petroeschevsky et al. (2001) found there was a significant association between daily hospital admissions and fine particles. Although the understanding of the health effects of fine and submicrometer particles is far from complete, and it can take some years to identify a precise mechanism, the results of existing toxicological studies suggest that both physical as well as chemical properties of the fine particulate fraction are involved in the observed health effects (Spurny, 1998).

2.3.2 Particle number concentrations

Previous health effects studies have also suggested that number concentration can be a better predictor of health effects than mass concentration (Oberdörster et al., 1992; Oberdörster, 1995; Oberdörster et. al., 1995; Wichmann and Peters, 2000). This is because submicrometer particles, particularly ultrafine particles, comprise the largest number of particles but do not represent the greatest mass, which is principally determined by supermicrometer particles. For example, Seaton et al. (1995)
indicated that ultrafine particles penetrate pulmonary interstitial spaces provoking inflammation. This response would depend less upon the mass of particles than their number, size and composition. This illustrates how fine particles, which can be high in number but contribute very little to particle mass (Jaenicke, 1993), can represent a risk to human health. Thus, it is clear that understanding of the behaviours of the number concentration of submicrometer and ultrafine particle indoors is of great importance in accurate exposure assessment and control strategies. Moreover, as mentioned above, studies on the number concentration of submicrometer and ultrafine particles indoors are limited.

2.3.3 Contribution of indoor particle sources to particle number concentration

Since indoor particle sources play a very important role in indoor particle concentrations, many studies on indoor particle sources have been conducted. As a result of these studies, many indoor particle sources have been identified qualitatively. However, due to the complexities involved in measuring emission rates of indoor particle sources, the database of particle emission rates from such sources is very limited. Furthermore, the existing database is restricted to indoor particle emission rates on a mass basis, and there are few measurements on a number basis.

2.3.4 The relationship between indoor and outdoor airborne particle concentrations

From equation 2-1, it can be seen that there is a relationship between indoor and outdoor particle concentrations. However, this relationship is affected by many other factors including: the air exchange rate, penetration efficiency, the deposition rate and the indoor particle generation rate.
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 particulate 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 concentrations 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; Monn, 2001; Liu et al., 2004). The results of these studies indicate that the ratio of the indoor to outdoor particle mass concentrations varies over a wide range, from 0.5 to 2, in the absence of indoor particle sources, and that indoor activities such as smoking or cooking can play an important role in affecting the relationship (Spengler et al., 1981; Monn et al., 1995; Ross et al., 1999, Monn 2001). While the focus of most of the reported studies has 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, as mentioned above, 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).

Understanding the relationship between 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 health risk. Current exposure assessment models often use outdoor pollutant concentration as the input parameter for predicting total human exposure (Colls and Micallef, 1997). However, as discussed above, indoor concentrations can
be different from outdoor ones, even in the absence of any significant indoor pollution sources.

2.3.5 Spatial and temporal variation of the submicrometer particle concentrations indoors

Although previous studies have provided information and knowledge about some aspects of indoor particles, the available information on fine and submicrometer particle indoors both in terms of mass concentration and number concentration is very limited.

Another clear weakness in the database of previous studies on indoor particles is the lack of information on the spatial and temporal, or short-term, variations of fine and submicrometer particle concentrations indoors. One of the main reasons for this is because the traditional filter sampling methods used in most of the previous studies provide only average data for indoor particle mass concentrations, over periods such as 2 hours, 24 hours or 48 hours. From these types of measurements, no information can be obtained about real-time particle concentration levels. It is difficult to accurately estimate the risk of particle exposure using this type of database as indoor particle levels can exhibit significant short-term variability (Kamens et al., 1991; Lyons and Morawska, 1996; Brauer et al., 1999) and the short-term peak can have a significant impact on human health (Michaels and Kleinman, 2000; Schwartz and Neas, 2000). Thus there is a need for time-series indoor monitoring of fine and submicrometer particles that would provide necessary information on the impact of time-varying sources of indoor fine and submicrometer particle concentrations. This information would help to improve understanding and assessment of human exposure.
In contrast to the large body of work relating to fine particle mass concentration indoors, there are few studies on the indoor fine and submicrometer particle number concentrations in the literature. Li et al. (1993) investigated real-time indoor submicrometer particle concentrations (size range: 0.017-0.886 µm) for 24 hours. Their results showed that the concentration varied from $1.4 \times 10^4$ to $1.5 \times 10^5$ particles cm$^{-3}$. There are also some studies that have simultaneously measured indoor fine and submicrometer particle number concentration and mass concentration for more than 24 hours. It was postulated that the pattern of diurnal variation of indoor fine and submicrometer particle concentrations is different to the pattern of diurnal variation of outdoor fine and submicrometer particle concentrations and indoor activities can significantly affect fine and submicrometer particle concentrations indoors.

2.3.6 The relationship between indoor and outdoor airborne particle morphology and elemental composition

Particle size is an important parameter in characterising the physical behaviour of aerosols. Particle size and shape can be quite complex and can affect the deposition of a particle in the respiratory tract and how it behaves after it has been deposited (Hickey, 1993; Vincent, 1995; Colbeck, 1998). From a toxicological standpoint, the particle chemical composition is of particular concern for health, since potentially toxic elements such as lead, arsenic, beryllium, cadmium, chromium, copper, selenium, zinc, acidic species, and certain organic compounds can cause both short and long-term medical problems. Therefore, these parameters are of fundamental importance in the understanding of the behaviour and potential health impact of an aerosol, and they are of current concern in terms of air quality and public health (Dye
et al., 2000). Moreover, such information is also important as a source signature for possible source identification (Figler et al., 1996; Lowenthal et al., 1992).

Sources of atmospheric aerosols, and size distributions of the chemical elements contained in aerosol particles, have been investigated by many authors using various methods for size segregation and elemental analysis for a variety of aerosols (Horvath et al., 1996; Davis and Guo, 2000; Teinila et al., 2000). These previous studies build up the basic knowledge of atmospheric aerosol sources and chemistry. For example, coarse particles (> 2 µm) are generally created by mechanical processes, such as weathering and wind erosion processes, and hence are rich in Ca, Fe, SiO₂, and other soil constituents. Fine particles are generally derived from processes such as combustion or gas to particle conversion, and are rich in C, Pb, sulphates, and ammonium and nitrate ions. Trace and often toxic species such as As, Cd, Cs, Sr, Zn, and Se are also mostly concentrated in the fine particles (Williams and Loyalka, 1991). Chan et al. (1997) examined the elemental and chemical composition of the PM₁₀ and PM₂.₅ aerosols. They found that on average, the major components of PM₁₀ were: crustal matter (25 % by mass), organic matter (17 %), sea salt (12 %), elemental carbon (10 %), and ammonium sulphate (7 %). The major components of PM₂.₅ from a site on a building located in a forest conservation area were: elemental carbon, organics and Pb/Br and sulphate. This confirms that the toxic elements in car exhaust exist in the finer fraction of particles, enhancing the risk of human exposure to these chemicals.

The vast majority of previous studies on the chemical composition of aerosol particles report only the chemical composition by bulk analysis techniques (e.g. Janssen et al., 1997; Hughes et al., 1999; Borbely-Kiss et al., 1999). The
disadvantage of using bulk analysis techniques is that they cannot reveal the morphological information of aerosol particles and chemical characterisation of single aerosol particles. Microanalysis of individual particles can reveal information on the chemical composition and size of the particle, the elemental lateral and depth distribution, and whether a specific element or compound is uniformly distributed over all particles. Based on these investigations, the particle's origin, formation processes, re-activity, transformation reaction, and environmental impact can be estimated (Bock and Grieken, 1999). In literature, there are relatively few cases in which an analysis of individual aerosol particles was performed. In addition, the majority of these previous studies on individual aerosol particles are focused on outdoor aerosol particles (e.g. Mukerjee et al, 1996; Ebert et al., 2000; Chabas and Lefèvre, 2000). For example, in a study by Chan et al. (1999), ten outdoor aerosol samples for PM$_{10}$ collected at five sites were examined by electron microprobe analysis of individual particle composition. They found that the electron microprobe technique was able to provide more detailed information on the specification and particle size characteristics of the mineral and carbonaceous particles than the other bulk elemental analysis techniques.

Similarly, most previous studies on the relationship between chemical composition of indoor and outdoor aerosol particles only report the chemical composition by bulk analysis techniques (e.g. Alzona et al., 1979; Sinclair et al., 1988; Kim et al., 1996). There are very few studies on the indoor/outdoor relationship by single particle analysis techniques. Ormstad et al. (1997) employed both a bulk analysis technique and a single particle analysis technique to study indoor and outdoor aerosol particles. They found that while the two techniques showed good agreement in the chemical composition analysis of aerosol particles, the single particle analysis technique
provided more detailed information that allowed for the conclusion that indoor air in Norwegian households is the same as outdoor air except for organic particles. The differences could be due to source characteristics, such as influence of sea salt aerosol on the total analysis.

2.3.7 The relationship between indoor and outdoor nitrogen dioxide and the relationship between submicrometer particles and nitrogen dioxide concentration levels

Fine and submicrometer particles are composed of a mixture of particles emitted directly into the air, and particles formed in the air from the chemical transformation of gaseous pollutants (secondary particles). One of the principal types of secondary particles is ammonium nitrate, formed from oxides of nitrogen (NOx) reacting with ammonia (NH3) (see Ohlström et al., 2000). Ultrafine particles are expected to behave in a similar manner to NO2. Thus, it is important to know the relationship between the fine particle and NO2 concentration levels and their trends.

Nitrogen dioxide (NO2) is a water-soluble red to brown gas with a pungent acrid odour. It is formed from the combination of nitrogen and oxygen from air during combustion at high temperatures (Maroni et al., 1995). It is an oxidising agent that is highly irritating to mucous membranes, and causes a wide variety of health effects (Jones 1999). As a major indoor pollutant, nitrogen dioxide (NO2) indoors and the relationship between indoor and outdoor concentrations of NO2 have been investigated by many researchers (Yocom, 1982; Monn, 2001). In the absence of indoor sources, indoor to outdoor ratios for NO2 are usually less than or near 1.0 (Yocom, 1982, Monn et al., 1998). In the presence of an indoor source, the NO2
levels are invariably higher than those outdoors. The order of magnitude of mean indoor/outdoor ratios for NO₂ is 2-5 with indoor NO₂ sources (Maroni et al., 1995).

Indoor NO₂ concentrations have been investigated in dwellings and schools in Australia (Brown, 1997). Ferrari et. al (1988) found that unflued gas heaters were a significant source of indoor NO₂. Lyall (1993) investigated NO₂ in 195 dwellings that used unflued gas heaters in Sydney, Adelaide and Perth and reported that 20% of the Sydney homes exceeded 300 ppb, but only 4-5% of Adelaide and Perth dwellings did so. Garrett et al. (1999) measured indoor and outdoor NO₂ in the Latrobe Valley, Victoria, by using passive samplers in 80 homes for over one year. They found that mean indoor NO₂ levels were higher than outdoor levels and seasonal variation was evident, with highest levels recorded in winter. The major indoor NO₂ sources were: gas stoves, vented gas heaters and smoking. Recently, a study by Lee et al. (2000) also demonstrated that the presence of gas ranges was the predominant factor affecting indoor NO₂ concentrations and personal exposures to NO₂ in Brisbane. However, there is still lack of indoor NO₂ data in Queensland residential houses.

2.3.8 Association between indoor air pollutants and house characteristics

Several published studies reported on the associations between indoor pollution and house characteristics. However, some of these studies investigated indoor NO₂ and other gaseous pollutants (e.g. Spengler et al., 1979; Lee, et al., 1996; Cyrlys et al. 2000; Sakai et al., 2004), some of them investigated indoor PM₁₀ or PM₂.₅ (e.g. Kingham et al., 2000; Fischer et al., 2000; Gotschi et al., 2002), but only one study investigates the associations with particle number concentration (Abt et al., 2000).
For example, a study by Fischer et al. (2000) indicated there were associations between house locations and indoor PM$_{10}$, PM$_{2.5}$, particulate components BaP, total polycyclic aromatic hydrocarbons (PAH), absorption coefficient (‘soot’) and the gas-phase components benzene, with the exception of the VOCs. In a study by Simoni et al. (1998), the associations between indoor PM$_{2.5}$, NO$_2$ and many house characteristics were analysed for both winter and summer. Cyrys et al. (2000) statistically analysed the associations between indoor NO$_2$ and house characteristics in 385 dwellings in Erfurt and Hamburg, Germany. Recently, Sakai et al. (2004) conducted statistical analysis of the associations between indoor gaseous pollutants and houses characteristics in 37 urban dwellings in Nagoya, Japan, and 27 urban dwellings in Uppsala, Sweden. Significant associations between indoor PM$_{10}$ concentration and several house characteristics including: house location (urban, suburban or rural), traffic volume (light or heavy), smoking (yes or no) and pet (yes or no), have recently been observed by BéruBé et al. (2004).

The results of previous studies highlighted some inconsistencies, such as in relation to smoking, or in the associations between indoor particle mass, NO$_2$ and house characteristics. It seems that the association between indoor air pollutants and several house characteristics could be case-specific. Abt et al. (2000) analysed the association between indoor particle number concentrations and house characteristics in three houses, such as gas appliances, house age, house volume, surface area, fraction of house carpeted, number of people. However, no association was found in their study. They mentioned it was possibly because of the small number of homes sampled.
As discussed above, there is no data available in the literature on simultaneous measurements of indoor and outdoor concentrations in residential houses of NO$_2$, submicrometer particle number concentration and PM$_{2.5}$. Thus, there is very limited data available for multiple association analysis between indoor air pollutants and house characteristics, especially for fine and submicrometer particles.

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CHAPTER 3

PARTICLE CONCENTRATION LEVELS AND SIZE DISTRIBUTION
CHARACTERISTICS IN THE RESIDENTIAL AND NON-INDUSTRIAL
WORKPLACE ENVIRONMENTS

L. Morawska, Australia, C. He, Australia

Centre for Medical and Health Physics, School of Physical & Chemical Sciences,
Queensland University of Technology,
Brisbane, QLD 4001, Australia

STATEMENT OF JOINT AUTHORSHIP

Title: Particle Concentration Levels and Size Distribution Characteristics in the Residential and non-Industrial Workplace Environments

Authors: Lidia Morawska and Congrong He

Lidia Morawska
Developed review design and scientific method; interpreted data and wrote manuscript

Congrong He (candidate)
Contributed to review design and scientific method; analysed and interpreted data; assisted with manuscript
3.1 Introduction

Particle characteristics in the indoor environment, including their concentration levels and size distributions, depend on a myriad of factors and conditions, and therefore vary considerably between houses in the same location and also between different geographic or socio-economic regions. Investigations of indoor particle characteristics encounter similar challenges to those of any studies of environmental properties where, due to the multiplicity of affecting factors, a large number of houses should be investigated to obtain valid and statistically significant results for the area under investigation. The area could be a suburb, city, region or country. The houses should be investigated for sufficiently long periods of time to account for temporal and seasonal variations caused by changes in meteorological conditions and in source strengths of human operated sources.

While the duration and spatial coverage of outdoor monitoring programs is constrained mainly by economic factors, indoor monitoring is far more complicated due to the varying degrees of intrusiveness of such monitoring to building occupants, and the effect various sampling techniques have on the indoor environment balance and thus on the parameters being measured. Economic factors affect the extent of the study, and, for example, the numbers of houses investigated, but the comprehensiveness of investigations in individual buildings, in terms of duration, continuity, or instrumentation used is also affected by what the occupants are prepared to accept. Therefore, studies reported in the literature vary considerably in terms of number of houses/buildings included (ranging from just one to over a thousand), duration of the measurements (ranging from a few hours in individual houses to months and years), and in the instrumentation which was used, and thus the parameters that were measured, which range from the simplest to the most
sophisticated. In addition, many studies had to be based on volunteer participation, as the response otherwise was poor, and therefore the results from such studies cannot be extrapolated to the whole population of the areas investigated.

Most of the earlier studies employed gravimetric methods for collection of particle mass, most commonly 24 hour average indoor concentrations of TSP, PM\textsubscript{10}, and PM\textsubscript{2.5} over a certain monitoring period ranging from hours to months (Dockery and Spengler, 1981 b; Sheldon et al., 1989; Pellizzari et al., 1993; Li, 1994 b). Such studies enabled the development of an understanding of the indoor mass concentration levels averaged over time (12- to 24-h time intervals). In addition, some of the studies provided information about relative contributions from indoor sources, and identified smoking and cooking as the most significant sources (Neas et al., 1994; Özkaynak et al., 1996). Other important contributions of these studies include:

- measurements of air exchange rates in the homes studied and use of air exchange rates to differentiate between indoor and outdoor sources and also for estimation of the relative importance of indoor and outdoor contributions;
- the development of a non-linear model using the measured air exchange rates to calculate an average penetration factor and deposition rate for PM\textsubscript{10} and PM\textsubscript{2.5};
- the use of a probability-selected population to be able to predict the exposures of an entire urban population (>100,000 persons) based on a sample of 178 participants;
- identification of the personal cloud (e.g., difference between personal and indoor concentrations) as an important contributor to exposure.
These studies, however, could not provide information about short-term variation in particle concentrations and their peak values, nor identify the effect of various house related and meteorological factors on the concentration levels. It is only since the 1990s that real time measurement methods have become more commonly available for indoor particle studies, and thus information on time resolved concentration has been attainable. However, a new complexity was introduced in relation to mass measurements, since some of these methods, as discussed in Chapter 2.2 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003), do not measure gravimetric mass, but derive a value based on other measured properties, for example light scattering (optical particle counters).

Since the 1980s, results of measurements of particle number concentration have started appearing, and previously unavailable information on number concentrations of particles in indoor environments have started emerging (e.g., McCusker et al., 1992). This information is still very limited, as the number of papers reporting particle number concentration is relatively small, and the scope of the reported studies, in terms of the number of houses investigated and/or duration of the studies, is not as extensive as the larger studies focussed on particle mass measurements.

The assessment and comparison of results from different studies is complicated by large differences in their design, including duration, number of houses investigated, instrumentation used, and thus the measured parameters. Nevertheless, a clearer picture of indoor particles, their concentration levels, trends in the concentrations and the factors affecting them, is emerging. In particular, there is a good understanding of the effect of the outdoor particle characteristics on those encountered indoors, and on the relative contributions from the most significant indoor sources to the indoor particle concentrations.
A comprehensive review of the studies conducted in the USA was undertaken by Wallace (1996). Other studies have been conducted since then, and in other parts of the world, but a detailed review of each individual study is beyond the scope of this chapter. Table 3.1 presents a summary of the major studies conducted to date on indoor particle characteristics, in terms of study duration and scope.

The number of houses investigated in the smallest of these studies was 140 (the Italian study); however, in the EXPOLIS study, while the overall number of houses was close to 200, the number of houses investigated in individual cities was lower (or well below 100). It can be seen from Table 3.1 that none of the larger studies measured particle number concentration or number size distribution. This is because the realization of the possible health effects of particles in the sub-micrometer or ultra-fine size ranges came later, and also because the instrumentation for particle number measurements is more expensive and complicated than for particle mass measurements, and there are no standardized methods for particle number measurements.

Not included in Table 3.1 are the studies that focussed mainly on personal exposure characteristics and less so on particle characteristics and house-related factors. For example, major personal exposures studies using probability-based sampling were conducted in Toronto and Indianapolis (Pellizzari et al., 1999, 2001). While in each case a large number of participants were included (close to 900 and 240 in Toronto and Indianapolis, respectively) and all the participants wore personal monitors, only a subset of the houses of the participants was investigated for indoor and outdoor particle concentrations. Some of the data from the study was provided in terms of the monitoring periods with usable PM data and not in terms of the number of houses for which the measurements were conducted.
<table>
<thead>
<tr>
<th>Name of the study and the country where it was conducted</th>
<th>Years study conducted and location</th>
<th>Number of houses and particle parameters measured</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Harvard Six-City study; USA</td>
<td>1979-1988 Cities: Portage, WI, Topeka, KN, Watertown, MA, Kingston, Harriman, TN, St Louis, MO, Steubenville, OH</td>
<td>More than 1400; PM3.5, PM2.5</td>
<td>Spengler et al., 1981; Dockery and Spengler 1981; Lebret et al., 1987; Neas et al., 1994</td>
</tr>
<tr>
<td>New York State ERDA study; USA</td>
<td>1986 (January-April) New York State Counties</td>
<td>433; PM2.5</td>
<td>Sheldon et. al., 1989, Leaderer and Hammond 1991</td>
</tr>
<tr>
<td>PTEAM Study; USA</td>
<td>1989 Riverside, California</td>
<td>178 PM10, PM2.5</td>
<td>Clayton et al., 1993; Wallace, 1996</td>
</tr>
<tr>
<td>Air Pollution Exposure Distribution within Adult Urban Populations in Europe EXPOLIS;</td>
<td>October 1996 to March 1998 Greece, Switzerland, French, Finland, Italy, Czech Republic</td>
<td>43 in Athens 41 in Basel 82 in Helsinki 20 in Prague PM2.5</td>
<td>Jantunen et al., 1998; Oglesby et al., 2000.</td>
</tr>
<tr>
<td>The Po River Delta indoor epidemiological study</td>
<td>August 1991-June 1992 A rural area of North Italy (Adria and Porto Tolle)</td>
<td>140 PM2.5</td>
<td>Simoni et al., 1998</td>
</tr>
</tbody>
</table>

The focus of this chapter is a review of the general trends in: particle characteristics, including mass and number concentration levels, and size distribution; the indoor/outdoor relationship, and the relative source contribution; the aim is not a discussion or comparison of absolute concentration levels between different studies. The factors discussed previously cause a large variation in absolute concentration.
levels and therefore it is generally more useful to understand the nature of the factors affecting the characteristics and the relative degree of the effect. This review therefore includes only those studies in which more general trends were investigated, or could be concluded based on information provided in the papers. However, for completeness of the discussion and interpretations, the concentration data for the discussed studies are also provided. Factors related to personal exposure assessment and thus personal exposure studies are not included in this chapter.

3.2 Outdoor Particles and Their Effect on Indoor Environments

There is a significant difference in the role of outdoor air, as a source of indoor particles, compared to the role of indoor sources. The indoor sources, while affecting outdoor particle characteristics to varying degrees, have a direct effect only on the house in which they operate. Since the characteristics of the sources and pattern of their usage differ from house to house, the resulting particle concentration levels and other characteristics will differ from house to house as well. Outdoor air, however, provides the same background for all the houses in the area, and even if the fraction of outdoor particles penetrating the buildings differs due to the differences in air exchange rate between the buildings or filters used, the time variation of this background remains the same (provided that the building operation parameters remain constant).

The relative role of this background concentration with respect to total indoor concentrations, which, in addition to the background, also include contributions from indoor sources, has been much discussed in the context of human exposure and health effects. In particular, as was concluded in the review conducted by Wallace
(2000a) (and already discussed in Chapter 1), despite significant variation in indoor particle concentration levels, a large number of epidemiological studies conducted in cities in different parts of the world have linked daily mortality statistics with increased particle concentrations measured outdoors. An increase of 1-8% in deaths for a 50 µg m⁻³ increases in outdoor air particle mass concentrations has been a common conclusion from these studies. This finding does not imply that indoor sources do not play a role in affecting particle concentrations and human exposures, as there have also been studies showing the link between concentrations elevated by indoor sources and health effects (Quackenboss et al., 1989; Naheer et al., 2000; Patterson and Eatough, 2000; Long et al., 2001b; Jetter et al., 2002). It indicates the importance of the outdoor air as a source of indoor particles, and the importance of understanding outdoor particle characteristics and their penetration of the buildings, in attempting to develop a general understanding of indoor particle characteristics and their variation. Indoor sources operate independently of the outdoor particle concentrations, so their effect on human exposure and thus health effects must be independent of the effects of the outdoor exposures to airborne particulate matter (Wilson et al., 2000).

The conclusion on the links between outdoor and indoor concentrations could be taken even further, and expanded to include the relationship between outdoor concentrations and personal exposure to outdoor-generated particulate matter, which as shown by Wilson et al. (2000), displays a strong temporal relationship. This is in contrast to much poorer correlations, or often a lack of correlation, between outdoor concentrations and personal exposure to non-outdoor particulate matter or total particulate matter. The authors concluded that outdoor concentrations might be used
in epidemiology as an appropriate surrogate for personal exposure to ambient-generated particles.

Information about outdoor particle characteristics, usually mass, is most commonly obtained from a limited number of central monitoring stations located in the study area, as it is not possible or practical to measure outdoor concentrations in the immediate proximity of each investigated house. It is thus of great importance to understand the spatial variability of particle concentration levels in the area under investigation. In the following, we give a brief overview of the trends in relation to spatial variability that have emerged from the data from central monitoring stations. Such stations are normally located away from known local sources, to provide data on the outdoor airshed characteristics of the pollutants measured. In the immediate vicinity of the sources, for example roads, the concentrations can be significantly elevated over that of the local background level, and the effect is more significant for particle number than for particle mass. The small-area variation, and, in particular, particle characteristics in proximity to roads, resulting from traffic emissions, and their impact on indoor particle characteristics, are discussed in chapter 3.7 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003).

In a number of studies, the spatial distribution of PM$_{10}$, PM$_{2.5}$ and other mass fractions in urban environments has been investigated. The results from the Six City study on the within-area variability of PM$_{3.5}$ showed that the spatial variation within the study sites was small, with the exception of one site in Steubenville, which was strongly affected by a single source (Spengler et al., 1981). A similar conclusion regarding homogeneity in spatial distribution was derived in the US-PTEAM study conducted in Riverside, CA (Clayton et al., 1993; Wallace, 1996). The study showed
that outdoor levels of PM$_{2.5}$ and PM$_{10}$ at different homes were in good agreement with the central monitoring site and that the correlations between outdoor (i.e. backyard) levels of PM$_{2.5}$ and levels at the central monitor were very good (0.96 overnight and 0.92 during the day). For PM$_{10}$, the correlations were also found to be high (0.93 overnight and 0.90 during the day).

Based on a comparison between an ambient monitoring site and the investigated residential sites, Williams et al. (2000) concluded that spatial homogeneity of PM$_{10}$ concentrations existed across the central region of Baltimore County, Maryland, USA. In addition, comparison of the concentrations from Baltimore County with those from another nearby State of Maryland monitoring site indicated close agreement (mean = 17.7 versus 18.8 µg m$^{-3}$), further supporting the conclusion of spatial homogeneity. Annual PM$_{2.5}$ mass concentration spatial homogeneity was also demonstrated across multiple measurement sites in Philadelphia and Washington (Bahadori, 1998). Studies of PM$_{2.5}$ concentrations conducted in the Minneapolis-St. Paul metropolitan area at three outdoor central community sites (Ramachandran et al., 2000) showed that the concentrations did not vary significantly by community or season, and that the day-to-day variability was the most important contributor to total variability. The 24-h PM$_{2.5}$ concentrations measured at the three sites tracked each other closely. Burton et al. (1996) also assessed the spatial variation within Philadelphia and concluded that the spatial variation was small for PM$_{2.5}$ but larger for PM$_{10}$ with spatial correlations near 0.9-1 and around 0.8 for PM$_{2.5}$ and PM$_{10}$, respectively. Similar results were obtained from a study by Ito et al. (1995) in Chicago and Los Angeles, with spatial correlations for PM$_{10}$ around 0.7-0.8. Blanchard et al. (1999) investigated the spatial variation of PM$_{10}$ concentrations within the San Joaquin Valley in California, and reported variation in PM$_{10}$ levels of
20% over distances of 4 to 14 km from the core sites. It was concluded that the local source affected sites over distances of less than 1 km, but depending on season, primary particulate emissions were transported over urban and sub-regional scales of approximately 10-30 km.

A number of studies have also been conducted in Europe on the spatial distribution of outdoor particulate matter. Oglesby et al. (2000) measured house indoor and outdoor PM$_{2.5}$ and outdoor PM$_{4.0}$ concentrations at the fixed monitoring site in the city of Basel, Switzerland, within the framework of the European EXPOLIS study. The study found that home outdoor PM$_{2.5}$ levels were highly correlated with corresponding, time-weighted outdoor PM$_{4.0}$ levels ($r_{sp} = 0.96$, $p < 0.0001$), and it was concluded that fixed site fine particle levels are valid proxies for population exposure to regional air pollution, predominantly consisting of secondary long-range particles, which include accumulated traffic-generated particles. Kingham et al., (2000) investigated small-area variations of pollutants within the area of Huddersfield, UK, and found that spatial variations of PM$_{10}$ and PM$_{2.5}$ were only modest and in most cases there was no significant association with distance from roads.

Spatial distribution of NO$_x$, ozone and PM$_{10}$ between three stations of the urban monitoring network of Brisbane, Australia, showed that spatial distribution of PM$_{10}$ was not as homogeneous as ozone or even NO$_x$, yet the degree of correlation was relatively high – with correlation coefficients in the range 0.34 to 0.54 (compared to correlation coefficients of ozone in the range 0.79-0.88) (Morawska et al., 2002).

In summary, the degree of homogeneity in PM$_{10}$ and PM$_{2.5}$ concentrations in the air is usually high or very high; nevertheless, complete homogeneity cannot be assumed. However, even in those cases where the reported values of correlation coefficients
were considerably less than unity, most of the correlations were statistically significant, which means that there was still a considerable degree of homogeneity, and therefore there could still be merit in applying the data from one station to nearby regions of investigation, if more detailed local information on spatial distribution is not available.

It is important to stress that homogeneity is seen when using monitoring data from stations not affected by local sources. As pointed out by Mage et al., (1999), data from a monitoring station intended to characterize the effect of the local sources of particulate matter should not be used in exposure and epidemiologic analysis of the wider community, and thus taken as a general background for indoor/outdoor investigations.

3.3 The Relationship Between Indoor and Outdoor Concentration Levels in the Absence of Indoor Sources for Naturally Ventilated Buildings

One of the major points of interest has been the particle concentration levels indoors in the absence of indoor sources, i.e. when outdoor concentrations and characteristics are the main contributing factors. It has been realized for a long time that infiltration of outdoor particles into houses is significant. For example, infiltration has been estimated to contribute about 70% in naturally ventilated homes and 30% in air-conditioned houses, to the indoor levels of respirable particles (Dockery and Spengler, 1981 a). The PTEAM study found that outdoor particles contributed about 75% of indoor levels for PM$_{2.5}$ and 66% to the indoor PM$_{10}$, in homes without apparent indoor sources (Özkaynak et al., 1996). Similar values were obtained by Janssen et al. (1998), who determined that approximately 70% of the variance in indoor levels in the Netherlands could be accounted for by changes in outdoor
concentrations. Abt et al. (2000) showed that at air exchange rates below 1 h⁻¹ and for particles in the size range 0.02-0.3 µm, 63-92% of indoor concentrations are from outdoor sources, while the remaining fraction is from indoor sources, in particular cooking. The outdoor contribution was found to be lower for larger particles and, in particular, for those in the size range from 2 to 10 µm, 57-80% of particles were estimated to be from indoor sources, including cooking, cleaning, and movement of people, with the remaining fraction being from outdoor sources.

Significant contribution of outdoor particle sources was also shown by Ott et al. (2000) who computed the PM₁₀ indoor proportion of outdoor particles for three large-scale field studies of residences by removing the impact of indoor sources on measured indoor PM levels with their random component superposition statistical model. The ratio of the 24 h PM₁₀ ambient indoor to outdoor components of concentration for the three studies were 0.54, 0.55, and 0.61, respectively, while the values ranged from 0.05 to more than 0.9.

As discussed in chapter 2.2, for naturally ventilated houses the main factors influencing particle concentration levels in the absence of indoor sources are: the penetration factor \( P \), deposition rate \( k \), and air exchange rate \( a \) (Eq. 1). A review of the numerical values of these parameters, factors affecting them, and their role on the indoor/outdoor relationship is also presented in Section 1.1.7 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003). For mechanically ventilated houses, as discussed in chapter 3.6 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003), the main factors are the type and efficiency of the filters used as well as the operating parameters of the HVAC system.
The relationship between indoor and outdoor concentrations is often considered in terms of indoor to outdoor concentration ratios (I/O). This section focuses on the discussion of the I/O in the absence of identified indoor sources, as reported in numerous studies, without, however, analysing the role of individual parameters affecting the ratio. As discussed in chapter 1.7 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003), this ratio is also termed infiltration factor or effective penetration efficiency.

First, the I/O ratio in naturally ventilated houses is discussed for PM$_{10}$ and PM$_{2.5}$ fractions. Secondly, the I/O for various other mass size ranges as well as particle number concentrations (in various size ranges) is examined.

### 3.3.1 PM$_{10}$ and PM$_{2.5}$

PM$_{10}$ PM$_{2.5}$ are the mass size fractions that predominantly investigated in studies assessing I/O ratios. The examples discussed below are studies conducted in Europe, Asia, USA and Australia.

Studies of indoor to outdoor ratios of PM$_{10}$ and PM$_{2.5}$ (as well as NO$_2$) for 17 naturally ventilated Swiss houses were conducted by Monn et al. (1997). In the study, the gravimetrical method of particle collection on Teflon filters was employed. The PM$_{10}$ concentration in the investigated houses ranged from 10.8 to 32.8 µg m$^{-3}$, while the outdoor concentrations ranged from 12.8 to 38.4 µg m$^{-3}$. The PM$_{2.5}$ concentration ranged from 18.3 to 26.0 µg m$^{-3}$ and from 21.0 to 33.9 µg m$^{-3}$, for indoor and outdoor, respectively. For houses with no indoor sources and no human activity the I/O ratios were 0.7 and 0.54 for PM$_{10}$ and PM$_{2.5}$, respectively (PM$_{2.5}$ without sources was measured only in one house). Under the same conditions, the NO$_2$ I/O ratios were <1.
Outdoor and simultaneous indoor measurements of PM$_{2.5}$ in an empty, fifth-floor lecture room with open windows were conducted in Chongju, Korea, during the summer season (Lee et al., 1997). The annular denuder system (ADS) was used to measure the indoor and outdoor air concentrations in this study. The I/O ratio was found to be 0.96, with a relatively high indoor-outdoor correlation ($R^2 = 0.785$).

Studies of nine non-smoking Boston-area homes were conducted for one or two periods during spring/summer and fall/winter (Long et al., 2001 a). PM$_{2.5}$ concentrations were measured using TEOM, and a seasonal-specific correction factor for loss of semi-volatile material due to the heating of the sample filter was applied. The mean outdoor hourly PM$_{2.5}$ concentration was 10.1 $\mu$g m$^{-3}$ (SD = 6.6 $\mu$g m$^{-3}$), while the mean indoor hourly PM$_{2.5}$ concentration was 7.1 $\mu$g m$^{-3}$ (SD = 5.2 $\mu$g m$^{-3}$). The I/O ratio for PM$_{2.5}$ under no indoor source conditions was found to be on average 0.74 (SD = 0.41). The results obtained in the same study for I/O ratios of size-classified samples are discussed later in this chapter.

Morawska et al. (2001) reported I/O ratios of PM$_{2.5}$ investigated in 14 residential houses in a suburb of Brisbane, Australia. The measurements of approximation of PM$_{2.5}$ mass were conducted using TSI DustTrak (using a light scattering technique, in which the amount of scattered light is proportional to the volume concentration of the aerosol). The results showed that the average ratio of indoor to outdoor PM$_{2.5}$ concentration was 1.01±0.14 and 1.08±0.22 under normal and minimum ventilation conditions, respectively. Normal ventilation meant that the windows, which are usually kept open by the residents, were also opened during the study, while minimum ventilation meant that all the windows and doors were closed.

The results from the representative studies listed above, which were conducted in different countries on different continents and using different techniques, as well as
those from other reported studies on I/O ratios of PM_{10} and PM_{2.5} concentrations, are compiled in Figures 3.1 A and 3.1 B, respectively. The conclusions that can be drawn from analysing the data presented in these figures are as follows.

**Figure 3.1 A.** Summary of the reported data on indoor/outdoor PM_{10} ratio in the absence of known indoor particle sources.

**Figure 3.1 B.** Summary of the reported data on indoor/outdoor PM_{2.5} ratio in the absence of known indoor particle sources.
For PM\textsubscript{10}, the reported of I/O ratios in the absence of known indoor sources have been ranged from 0.50 to 0.98, with a median value of 0.70. These values show that firstly, the contribution of outdoor air as a source of indoor particles is very significant, and secondly, that it is rather consistent across all the studies. Based on these results a reasonable prediction can be made about indoor concentration of PM\textsubscript{10} for a typical indoor environment, if outdoor concentrations are known and in the absence of indoor sources. More accurate information could be obtained by conducting comprehensive experimental investigations in the environment of interest. Modelling of this ratio, on the other hand, may not be a source of better information, given the need for a good knowledge of the parameters affecting the ratio (penetration factor, deposition rate and air exchange rate), and a high degree of uncertainty in estimating these parameters and decoupling one from the other, as discussed in chapter 1 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003).

Similar conclusions can be drawn in relation to the PM\textsubscript{2.5} ratio, although in this case the median value of the I/O ratio of 0.91 is somewhat larger than that for PM\textsubscript{10}, with the reported values ranging from 0.54 to 1.08. A higher ratio is expected for PM\textsubscript{2.5} compared to PM\textsubscript{10}, since as discussed in chapter 1 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003), penetration factor for larger particles in the supermicrometer range is lower than that for smaller particles in this range, and in addition the deposition loss rate of these larger particles is higher.
3.3.2 Mass, Volume or Number Size Distribution

Many studies of indoor/outdoor particle ratios have employed inlet cut-off points other than 2.5 or 10 µm for particle mass measurements or surrogate measures of this ratio. For example, Alzona et al. (1979) observed average infiltration factors of 0.24 and 0.42, respectively, for iron and lead. Iron is a known tracer for coarse particles and lead is more commonly associated with fine particles. Dockery and Spengler (1981 a) reported a mean infiltration factor for PM$_{3.5}$ of approximately 0.70 and a similar value of 0.75 for sulfate. Aerosol size distribution was measured by McMurry et al. (1985) indoors and outdoors at a sealed Minnesota home with an electrical aerosol analyser (EAA) for particle sizes between 0.01 and 0.56 µm and a single-particle optical counter (OPC) for particles between 0.56 and 1.0 µm. For the entire spectrum, the reported infiltration factors were consistently low, between 0.2 and 0.4.

Later studies started employing real time (or near real time) measurement methods, usually with the capability for particle size classification and for particle number measurements.

Abt et al. (2000) estimated effective penetration efficiencies for 0.02-0.5 µm and for 0.7-10 µm particles as ranging from 0.38 to 0.94 and from 0.12 to 0.53, respectively, in four houses in the Boston area. The measurements were conducted using a scanning mobility particle sizer (SMPS) and an aerodynamic mobility particle sizer (APS) for the smaller and larger particles, respectively. While the instruments measure particle number, the data from this study were analysed in terms of particle volume concentration (µm$^3$ cm$^{-3}$), which was recalculated from the number values. The authors showed that the results for 0.02-0.1, 0.1-0.2, 0.2-0.3, and 0.3-0.4 µm
particle size ranges were not statistically different from one another, with overlapping error bars for these ranges. However, for particles from 0.1 to 0.2 µm, for which the losses from diffusion and impaction are minimal, the estimated effective penetration efficiency was highest. For 0.7-10 µm particles, effective penetration efficiencies decreased with increasing particle size, which the authors partially attributed to the increased deposition rates associated with increasing particle size.

Similar results were obtained by Long et al. (2001a), who applied the same type of instruments, an SMPS and an APS, for measurements in 9 houses, also in the Boston area. The outdoor and indoor particle concentration ranges analysed were: 0.02-0.1, 0.1-0.5, 0.7-2.5, and 2.5-10 µm, and the results were presented as volume concentrations. The mean outdoor concentrations exceeded the mean indoor concentration in these size ranges by 0.15, 1.5, 1.3, and 1.9 µm³ cm⁻³, respectively. The mean infiltration factors for these size ranges were 0.75 (SD = 0.37), 0.74 (SD = 0.20), and 0.48 (SD = 0.18) and 0.27 (SD = 0.16), respectively. Thus, the variability in the infiltration factor was greater for fine particles than for coarse particles, and the factors were more broadly distributed across a larger range of values for the three lower size intervals.

The lowest infiltration factors were 0.52 for the smallest particle size range (for the 0.02-0.03 µm size interval) and 0.16 for the largest size range (for the 6-10 µm size interval), while the largest infiltration factors of 0.70-0.73 were observed for particles in the range from 0.08 and 0.5 µm (the accumulation mode). The lower I/O ratios in the ultra-fine and coarse-mode ranges can be explained by depositional losses due to diffusion and gravitational settling, during the transport of particles.
through the building envelope, while the peak in the accumulation mode coincides with the region where neither loss mechanism is significant.

Infiltration factors were higher in summertime, usually exceeding 0.7 when windows and doors were open more often, and lower in fall/wintertime periods when they were typically closed. For the closed windows condition, penetration efficiencies decrease, while deposition rates tend to decrease due to reduced indoor turbulence and thus decreased likelihood that particles will migrate through the boundary layer and deposit onto surfaces (Nazaroff and Cass, 1989). In the reported study, mean wintertime air exchange rates were 0.89 h\(^{-1}\), while the combined wintertime and summertime mean was 2.1 h\(^{-1}\). The authors concluded that there is a steady increase of infiltration factors with the increase in the air exchange rate, up to about two or more air changes per hour, when the infiltration factors level off. Such high air exchange rates are normally associated with open windows and doors.

Morawska et al. (2001) reported that under normal ventilation conditions the average ratio of indoor to outdoor concentrations of particles in the size range from 0.007 to 0.808 µm, measured with a condensation particle counter (CPC), was 0.89±0.14 and the average ratio of indoor to outdoor concentration of larger particles (measured by APS) was 0.97±0.14. Under minimum ventilation conditions, the average ratio of indoor to outdoor submicrometer particle concentrations was 0.78±0.49, and the ratio for larger particles was 0.95±0.18.

Since each of the studies used either different instruments or investigated different size ranges, or presented the results in a different format, direct comparison of the results, or attempts to present them in the same figure, would not be meaningful. A general conclusion that can be drawn from these results is that the scatter of the
reported results for size-classified particles is substantial, and more studies in this area should be conducted before more firm conclusions can be drawn.

3.4 Indoor Sources of Particles: Contribution to Indoor Concentration Levels and the Effect on I/O Ratios for Naturally Ventilated Buildings

When investigating the effect of indoor sources on particle characteristics in indoor environments, some of the main questions that could be asked include:

1. What are the emission rates or factors of the most important indoor sources?
2. What are the characteristics, including size distribution, of particles generated by these sources?
3. What is the absolute increase in particle concentration levels when the sources are in operation?
4. What are the I/O ratios in the presence of these sources?

This chapter will focus on the two later questions, as the source emission factors and size distribution, as well as other characteristics of particles generated by various sources, were discussed in chapter 19 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003). These questions are addressed first in relation to particle mass and mass size fractions, and then in relation to particle number.

The contribution of indoor sources to indoor particle concentration levels depends in the first instance on the characteristics of the sources, particularly their strength, but of equal importance are the operating parameters of the building, particularly the air exchange rate. Not all of the reported studies have involved the measurement of air exchange rate, and often, only general information about the buildings has been provided, for example, whether the building is naturally or mechanically ventilated.
This section is not concerned with air exchange rate and other parameters or their impact on the indoor concentration levels. Instead, we review the reported concentration levels as monitored in numerous studies, and their relationship with the indoor sources and with the outdoor air concentrations. For many of the studies, however, while it was stated the measurements were conducted in the presence of operating sources, the authors did not analyse the contribution or the effect of the individual sources.

In most of the studies reviewed, in which individual sources were investigated, it was concluded that the highest impact was from smoking, cooking and general activities, and therefore a brief review of the impact of these sources is provided in this section. This conclusion supports the earlier findings from the three major US studies on the impact of these sources, as summarised in the review by Wallace (1996). However, in the same review, the author concluded that a substantial portion of indoor particles were due to unexplained indoor sources. It was estimated through mass balance modelling that the contribution from unidentified sources was in the range of 25-30% of the particle matter present in the indoor environment.

Most of the studies reviewed in this section included only a relatively small number of houses, ranging from one to about forty houses, with the exception of the EXPOLIS study, which overall included more houses, although in the individual cities the numbers were small (higher in Helsinki).

3.4.1 Short-Term Versus Average Particle Concentrations

For many studies, particle concentrations have been reported only in terms of average values, particularly if the measurement techniques used were based on gravimetric sample collection. In some of the studies, average concentrations as well
as short-term variation have been reported. From these, it can be seen that short-term increases in particle concentration as a result of source operation could be hundreds of times higher than the average values.

For example, PM$_{2.5}$ indoor and outdoor concentrations were measured by Ramachandran et al. (2000) in 21 different residences in the Minneapolis-St. Paul metropolitan area, USA. While in general the measured concentrations did not vary significantly by community or season, short-term (15-min) indoor PM$_{2.5}$ concentrations routinely exceeded 24 h average outdoor values by a factor of three to four. The results showed that while the 24 h I/O ratios had a fairly narrow range (median = 1.17, minimum = 0.33, maximum = 4.02), the range of 15min ratios was relatively wider (median = 1.4, minimum = 0.36, maximum = 60.2).

Measurements of PM$_{10}$ and TSP concentrations were conducted in eight naturally ventilated residential flats in Hong Kong, China, and the study showed that very high concentrations were reached during some of the indoor activities (Chao et al., 1998). At higher air exchange rates, however, the peak concentrations were lower. Particle concentration resulting from incense burning reached a level of 1 850 µg m$^{-3}$ in one site, where air exchange rate was 1.09 h$^{-1}$; however, a level of only around 170 µgm$^{-3}$ at a site where the air exchange rate was three times higher. Removing of a thick layer of dust from the top of the cabinets and the floor using a broom resulted in concentrations of the order of 1600 µg m$^{-3}$. Frying of chicken wings, leading to the emission of hot oil and vapour mixture from the uncovered cooking pan, resulted in maximum particle levels in excess of 5500 µg m$^{-3}$. This was quite different from cooking steak, steamed fish and vegetables, which required a covering lid on top of the pan during cooking. Cigarette smoking led to concentration levels up to 300 µg m$^{-3}$ at the site with mean ventilation rate of 6.12 ACH.
Long et al. (2000) investigated nine non-smoking houses in the area of Boston, USA. The study showed that the mean values of indoor and outdoor concentrations were similar, with the outdoor concentrations somewhat lower than the indoor ones. There was, however, a clear short-term impact of the indoor sources, which for PM$_{2.5}$ was demonstrated both by larger standard deviations for the hourly data in comparison to 12 h measurements and in the greater maximum indoor PM$_{2.5}$ concentrations for the hourly data of 471.9 vs 85.5 µg m$^{-3}$ for the 12h samples. The mean indoor hourly and 12 h PM$_{2.5}$ concentrations found in this study were 11.8 µg m$^{-3}$ (SD = 20.7) and 11.9 µg m$^{-3}$ (SD = 9.6), respectively. The mean outdoor hourly and 12 h PM$_{2.5}$ concentration were 11.1 µg/m$^3$ (SD = 9.5) and 11.1 µg m$^{-3}$ (SD = 6.8), respectively. The mean and median values of the hourly air-exchange rates were 1.22 and 0.74 h$^{-1}$, with the range between 0.11 and 20.40 h$^{-1}$. The I/O ratios for daytime periods ranged from 0.03 to 257 with a mean of 2.4 ± 14, and for night-time non-source periods, from 0.03 to 3.7 with a mean of 0.74 ± 0.41. The five largest peak concentrations were observed during cooking activities – gas-fired oven baking (101 ± 185 µg m$^{-3}$), sautéing (66 ± 95 µg m$^{-3}$), toasting (55 ± 120 µg m$^{-3}$), frying (41± 43 µg m$^{-3}$) and stir-frying (37 ± 31 µg m$^{-3}$). For particles in the range from 2.5 to 10 µm, the most significant indoor events were dusting (82 ± 70 µm$^3$ cm$^{-3}$), vigorous walking (29 ± 19 µm$^3$ cm$^{-3}$) and sautéing (23 ± 23 µm$^3$ cm$^{-3}$). It is interesting to note that cleaning activities using a dilute ‘Pine Sol’ solution also contributed to the increase in the PM$_{2.5}$ mass concentration of approximately 3-32 µg m$^{-3}$.

A similar conclusion can be drawn from all of the above studies, namely that the short-term effect of the operation of indoor sources is an increase of the indoor particle concentrations by up to two orders of magnitude, with the actual level of
increase being dependent on source characteristics as well as on the air exchange rate. While there are some data available on indoor sources emission factors, the data is still very limited and variation in emission factors between sources of the same type is substantial; thus, model-based predictions of the level of increase in individual indoor environments are not very reliable.

Of interest with regard to human exposure assessment is not only the absolute increase in particle concentration as a result of source operation, but also for how long the concentration remain elevated after the sources cease to operate. This depends primarily on the air exchange rate and, under the range of conditions encountered in residential environment, can last from minutes to hours.

For example, Chao et al. (1998) showed that after conclusion of incense burning in a flat with an air exchange rate in the range from 1.09 to 3.24 h\(^{-1}\), smoke accumulation was sustained for half-an-hour or longer. At a site with a mean ventilation rate of 6.12 ACH, each peak resulting from cigarette smoking lasted for 20-30 minutes, although the smoker finished a cigarette within 4-5 min.

Number concentrations of submicrometer particles in four residential houses in Brisbane, Australia, were measured by Lyons and Morawska (1996). Sources such as cooking, clothes washing, vacuuming, lawn mowing, and smoking were investigated. It was concluded that concentrations from indoor sources can remain significantly higher than background levels for periods up to three to four hours under minimum ventilation conditions, when all the windows and doors were closed, and for around one to two hours for average conditions, i.e. when some windows were opened. In another study conducted in Brisbane (Morawska et al., 1997), a high concentration of environmental tobacco smoke (ETS) was recorded up to three hours after smoke generation, depending on the air exchange rate. In this study the air
exchange rates ranged from 0.55 to 0.79 h\(^{-1}\) for minimum ventilation and from 1.93 to 4.48 h\(^{-1}\) for normal ventilation.

An example of the duration of the impact of indoor activity such as cooking and smoking on indoor particle PM\(_{2.5}\) and number concentration is presented in Figure 3.2, based on a study conducted in Brisbane by Morawska et al. (2003). This figure illustrates the point discussed above, i.e. particle concentration indoors can remain elevated for a few hours after the sources generating the particles cease to operate. An implication of this is that nighttime particle concentrations do not necessarily represent the background concentrations (not affected by indoor sources). A similar conclusion was drawn by Long et al. (2001a) in relation to variability in assessed infiltration factors, despite the use of nightly average data. In particular, some of the infiltration factors, which were found to be greater than unity, were hypothesized to be due to the confounding effect of indoor-outdoor time lag.

![Figure 3.2: House 12, 21-23/May/1999, CPC and PM\(_{2.5}\) concentrations in kitchen, 48h (CS: cigarette smoking) (Morawska et al., 2003)](image-url)
3.4.2 The Effect of Indoor Sources on Particle Indoor Mass Concentration and Mass I/O Ratios

General

Many studies have been carried out to investigate the correlation between indoor and outdoor concentrations of particles, and while in some it was concluded that, in general, indoor concentrations followed the outdoor levels, in others the relationship was found to be weak, and in some cases no correlations could be identified. The presence or absence, or the strengths of correlations has also been shown to be dependent on the season, which is directly related to ventilation conditions; higher in summer and lower in winter.

In early studies, it was concluded that, in general, the correlation between indoor and outdoor levels of PM$_{10}$ and PM$_{2.5}$ was poor (Dockery and Spengler, 1981 b; Ju and Spengler, 1981; Sexton et al., 1984). However, the influence of season on indoor/outdoor relationships and reduced ventilation in winter was such that, for example, the differences between levels of both PM$_{10}$ and PM$_{2.5}$ in homes with smokers compared to homes without smokers were stronger during winter than summer (Spengler et al., 1981; Quackenboss et al., 1991).

Indoor and outdoor PM$_{2.5}$ concentration levels in winter and summer were measured within the framework of the European EXPOLIS study (Gotschi et al., 2002) at the homes of the study participants in Athens (Greece), Basel (Switzerland), Helsinki (Finland) and Prague (Czech Republic). A summary of the results from this study is presented in Tab 3.3-2. It can be concluded that PM$_{2.5}$ concentrations, apart from Helsinki, were higher in winter than in summer, both indoors and outdoors. Except in the case of Prague, a significant correlation was found between indoor and
outdoor concentrations of PM$_{2.5}$. Multivariate models for PM$_{2.5}$ attributed variance in the indoor concentrations to indoor sources (use of gas appliances, cigarettes) to extents of 47%, 54%, 51%, and 71% for Athens, Basel, Helsinki, and Prague, respectively. Thus, in Prague, the impact of outdoor on indoor concentrations was small and the variables for indoor sources were strong predictors of PM$_{2.5}$ indoor concentration.

### Table 3.2 PM$_{2.5}$ ($\mu$g m$^{-3}$) indoor and outdoor concentrations and the ratios of summer and winter, and smoking and non-smoking concentrations (adapted from (Gotschi et al., 2002))

<table>
<thead>
<tr>
<th>Center</th>
<th>$N$</th>
<th>Indoor (mean±sd)</th>
<th>Outdoor (mean±sd)</th>
<th>median of indoor/outdoor ratios (paired)$^a$</th>
<th>W/S ratio of means (unpaired)$^b$ Indoor</th>
<th>W/S ratio of means (unpaired)$^b$ Outdoor</th>
<th>SM/NSM ratio of means indoors (unpaired)$^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Athens</td>
<td>43</td>
<td>35.6 ± 29.4</td>
<td>37.3 ± 27.4</td>
<td>0.90</td>
<td>1.47</td>
<td>1.58$^b$</td>
<td>1.41 (15)</td>
</tr>
<tr>
<td>Basel</td>
<td>41</td>
<td>21.0 ± 16.7</td>
<td>19.3 ± 11.5</td>
<td>0.98</td>
<td>1.58$^b$</td>
<td>1.28</td>
<td>1.89$^b$ (7)</td>
</tr>
<tr>
<td>Helsinki</td>
<td>82</td>
<td>9.5 ± 6.1</td>
<td>10.5 ± 7.1</td>
<td>0.91</td>
<td>0.84</td>
<td>1.10</td>
<td>1.91 (2)</td>
</tr>
<tr>
<td>Prague</td>
<td>20</td>
<td>34.4 ± 28.7</td>
<td>27.3 ± 10.4</td>
<td>1.04</td>
<td>1.13</td>
<td>1.35$^b$</td>
<td>2.09$^b$ (8)</td>
</tr>
</tbody>
</table>

$^a$ No mean ratio of indoor-outdoor pairs significantly different from 1 at 5% level in paired $t$ test. Only median of indoor-outdoor ratios shown.

$^b$ Ratios of means significantly different from 1 at 5% level in unpaired $t$ test.

$^c$ $N$ of households with tobacco smoke exposure in parentheses. W/S: winter/summer; SM/NSM: smoke/no-smoke.

Measurements of PM$_{10}$, PM$_{2.5}$ and PM$_{1}$ were conducted by Jones et al. (2000) inside and outside seven homes in Birmingham, UK, and two homes in rural locations during a 12 month period, using TEOM. Two of the urban homes were on the 10$^{th}$ and 13$^{th}$ floors of a multi-storey block of flats in the city centre; others were at ground level. The results showed that indoor concentrations broadly followed outdoor concentrations, with additional indoor elevations being attributed to cooking.
(electric) and cleaning. A generally weak correlation between indoor and outdoor concentrations was found for all particle size fractions, especially for PM$_{10}$. The correlation improved somewhat during times of increased natural ventilation and at times of low indoor activity during the day. For PM$_{10}$, in addition to outdoor particles, indoor sources such as cooking, smoking, cleaning and general activity contributed substantially to the indoor concentrations and were the dominant sources of episodic peaks in this mass range. Cooking and smoking were determined to be the major indoor sources of PM$_{2.5}$ and PM$_{1}$, whilst cleaning and general activity had little influence on concentrations within this size range.

The few examples discussed above demonstrate the variability in the degree of correlation between indoor and outdoor particle concentrations found in different studies, and also the association of this dependence with the season of the year. The variability in the correlation can be attributed to differences in the air exchange rates for different buildings as well as to the differences in indoor source strengths. The seasonal dependence of the indoor to outdoor relationship between particle concentrations results from seasonal variation in the air exchange rate.

One other general comment that should be made is that, unlike the effect of outdoor on indoor particle characteristics, which is normally homogenous for the entire indoor naturally ventilated environment, provided that air exchange in all the indoor compartments is the same, operation of indoor sources or activities conducted indoors under most circumstances results in a heterogeneous distribution of particles and other pollutants indoors. It is logical to expect that, for example, particle concentration will be elevated during cooking in the kitchen, above the background levels present in the bedroom, or that concentration of larger particles resulting from resuspension will be higher near the ground. This would not be the case if air mixing
were very efficient, which in most indoor environments is rarely the case. For example, Micallef et al. (1998) showed that the vertical concentration profile for TSP changed with time and that human absence coincided with low concentrations at all levels from ground, while human activity induced higher concentration and a concentration gradient with peaks appearing over the height range 1.29-1.77 m. For TSP and PM$_{10}$, on average the highest concentration was measured at around 1.29 m. Similar, although less quantitative, were the results obtained by Luoma and Batterman (2001) on the dependence of concentrations of supermicrometer particles on height. In contrast, no significant differences were found in the concentrations of submicrometer particles with height. The latter can be attributed to different dynamics of smaller and larger particles in the air.

In conclusion, operation of indoor sources result in spatial heterogeneity of particle concentrations in indoor environments, which has to be taken into account when designing indoor measurements, and also when assessing human exposure indoors.

*Indoor Sources, no Speciation*

Many of the investigations of indoor particle concentrations have not included in their designs the gathering of detailed information on the presence, characteristics, or the effects of indoor sources. Instead, the investigations have focused on average particle concentrations under normal operating conditions of the studied indoor environments. Some of these studies had as an objective not only to assess particle concentration levels, but also to link these to the exposure characteristics of the occupants.

For example, Rojas-Bracho et al. (2000) performed a study on the personal, indoor, and outdoor PM$_{10}$, PM$_{2.5}$, and PM$_{2.5-10}$ exposures of 18 individuals with chronic
obstructive pulmonary disease living in Boston, MA, US. Monitoring was carried out on each participant for six consecutive days in winter and for six to twelve days in the summer. On each day, 12-hour personal, indoor, and outdoor samples of PM$_{10}$ and PM$_{2.5}$ were collected simultaneously. Their results showed that the geometric means for the I/O ratios were greater than unity, being 1.3 for PM$_{2.5}$ and 1.4 for PM$_{10}$. The study also showed that, in total, 61% and 69% of the measured indoor PM$_{2.5}$ and PM$_{10}$ concentrations were higher than the corresponding outdoor levels.

Another US study on PM$_{2.5}$ and PM$_{10}$ concentrations was focused on 13 residences in Coachella Valley, California, U.S. (Geller et al., 2002). The I/O ratios were 0.66 ±0.27 and 1.03 ±0.29 for coarse particles (2.5-10 µm) and PM$_{2.5}$, respectively. A relatively weak correlation was found between the outdoor and indoor concentrations ($R^2 = 0.35$) for coarse particles, and the authors concluded, that a substantial fraction of coarse particles are generated indoors by dusting, cleaning, washing, and resuspension, with the generation rate varying between the houses. However, outdoor concentrations could only account about 37% of the variation in indoor concentrations, indicating significant contributions from indoor sources to the overall PM$_{2.5}$ concentrations.

Indoor and outdoor PM$_{10}$ and PM$_{2.5}$ concentrations were measured over ten weeks for three houses in Taipei, Taiwan, China (Li et al., 1993a; Li, 1994a). The I/O ratios of PM$_{10}$ ranged from 0.58 to 0.71, of PM$_{2.5}$ from 0.54 to 0.91, and of TSP from 0.52 to 0.6. It was also reported that PM$_{10}$ concentrations ranged from 20 to 300 µg m$^{-3}$ indoors and from 40 to 350 µg m$^{-3}$ outdoors, while the PM$_{2.5}$ concentrations ranged from 20 to 150 µg.m$^{-3}$ indoors, and from 10 to 200 µg.m$^{-3}$ outdoors. In a later study, Li (1994a) investigated the indoor to outdoor ratio of PM$_{10}$ (and also of 20 elements present in the PM$_{10}$) of 15 homes in Taipei over an eight-week period. Mean PM$_{10}$
concentrations of 83 µg m\(^{-3}\) indoors and 107 µg m\(^{-3}\) outdoors were found, and the I/O ratio ranged from 0.10 to 2.30 with a median value of 0.83.

A study conducted in Brisbane, Australia during winter 1999 included PM\(_{2.5}\) and particle number measurements in 14 residential houses (Morawska et al., 2003). Diurnal variation of particle characteristics was monitored continuously for more than 48 hours in the kitchen of each house, and the residents kept diaries of the activities conducted. In addition, data from a central monitoring station was used to investigate indoor/outdoor relationships. The results showed that there were clear diurnal variations in particle number concentrations, for all the investigated houses. The pattern of diurnal variations varied from house to house, although there was always a close relationship between the concentration and human indoor activities. The average PM\(_{2.5}\) concentrations during indoor activities were \((15.5 \pm 7.9)\) µg m\(^{-3}\) and \((11.1 \pm 2.6)\) µg m\(^{-3}\) under non-activity conditions. The PM\(_{2.5}\) I/O ratio for all the houses ranged from 0.75 to 4.86, with the median of 1.44.

Despite somewhat different objectives of the five studies mentioned above, and the range of different outcomes, all of them included investigation of the I/O ratios under normal operating conditions of the house, i.e. in the presence of indoor sources. Due to the differences in the environments investigated (tropical versus temperate climate), the study designs, and the information provided on the characteristics of the houses investigated, direct comparison of the results from such studies might not always be possible. However, studies like these have contributed to the development of a general understanding of I/O ratios, and to the global database on the numerical values of this ratio. The latter can be used for identification of general trends in I/O ratios, and is further discussed below.
**Cigarette Smoking**

The impact of cigarette smoking on particle concentrations has been investigated in terms of: (1) the increase in particle concentrations in the houses of smokers compared to the houses of non-smokers for various averaging periods, (2) increase in particle concentration as a result of one cigarette smoked, and (3) I/O ratios for houses with and without smokers.

Increased concentrations of PM$_{2.5}$ as a result of cigarette smoking have been investigated in many places. Simoni et al. (1998) measured PM$_{2.5}$ concentrations in 140 homes in the Po River Delta area of Northern Italy (near Venice) during summer and winter, and showed that in the presence of ETS there was an increase of about 33 $\mu$g m$^{-3}$ in a 48-h average of PM$_{2.5}$ in winter. Similar levels of increase of 30 to 35 $\mu$g m$^{-3}$ in a 12-h average indoor PM$_{2.5}$ and of 25 to 32 $\mu$g m$^{-3}$ in the 24-h average were reported in the PTEAM study (Özkaynak et al., 1996) and the six cities study (Spengler et al., 1996), respectively. Larger increases of 45 to 47 $\mu$g m$^{-3}$ in a seven-day average were found by Sheldon et al. (1989) in a study conducted in N.Y. State. A study by Jones et al. (2000) showed that the I/O ratios in smoking homes were greater than unity (2.7±6.7) at almost all times of the day. A summary of the EXPOLIS study presented in Table 3.2 shows that the ratios of PM$_{2.5}$ concentrations in smoking and smoke-free houses were invariably greater than unity and ranged from 1.41 in Athens to 2.09 in Prague. However, Lee et al. (2002 b), who investigated PM$_{10}$ concentrations in six houses in Hong Kong, China showed that the average indoor levels of homes with and without smokers did not vary greatly, being 155 $\mu$g m$^{-3}$ and 148 $\mu$g m$^{-3}$, respectively.
In a number of studies, the contribution to the indoor PM$_{2.5}$ concentrations from the smoking of one cigarette has been considered. Simoni et al. (1998) found an increment due to each smoked cigarette of about 0.2 µg m$^{-3}$ in a 48-h average concentration, corresponding to an increase of 0.8 µg m$^{-3}$ in a 12-h average. Lebret et al. (1987) found a value of 0.08 in a 24-h average (based on the six-cities data), while Leaderer and Hammond (1991) (N.Y. State data) found an increment of 0.3 µg m$^{-3}$ in the weekly average, corresponding to 2.1 µg m$^{-3}$ in a 12-h average. The increase derived from the PTEAM data was 1.8 µg m$^{-3}$ in a 24-h average.

I/O PM$_{10}$ ratios of 17 houses in Zürich, Switzerland were investigated by Monn et al. (1997) for natural ventilation conditions. The study showed that of the indoor sources, smoking had the highest influence on I/O ratios, and, in the absence of sources ‘human activity’ was a significant source of particulate matter. For homes with indoor sources operating, the I/O ratio for PM$_{10}$ was 1.84 and 2.07 for two houses with smokers. Similar values were reported by Jones et al. (2000), who found that the I/O PM$_{10}$ ratios measured in a country home with smokers ranged from 1.8±0.9 (in May) to 2.9±3.0 (in September), and was 3.9±7.8 in Mach for an urban flat.

In conclusion, while all of the above-discussed studies showed an increase in absolute particle mass concentration levels as a result of smoking, or in the ratios of concentrations between houses with and without smokers, the actual level of increase has varied between the studies. The highest variation reported was in the contribution of one cigarette smoked to indoor concentration levels, with values differing by up to an order of magnitude. However, the variation in the overall level of increase was much smaller, in general not exceeding a factor of two, whether presented in terms of absolute concentrations or concentration ratios.
**Cooking**

Like smoking, the effect of cooking on indoor particle mass concentration levels has been investigated and expressed in a number of ways. In many studies, I/O ratios in the presence and absence of cooking activities have been determined, and the reported average values for this ratio most commonly ranged from somewhat above one to about two (with a small number of studies reporting higher than two). In the study by Monn et al. (1997), discussed above, the I/O ratio for PM$_{10}$ was 1.18 and 1.24 for two houses using gas cookers. Measurements of PM$_{10}$, PM$_{2.5}$ and PM$_{1}$ conducted by Jones et al. (2000) in the UK, also mentioned above, showed that in one of the houses elevated PM$_{10}$ concentrations were linked to meal preparation (often preparation of toast), while in another house, PM$_{2.5}$ concentrations were only slightly increased due to cooking, although PM$_{1}$ concentrations were elevated. Mean PM$_{10}$ I/O ratios were less than one during the early hours of the morning, close to or greater than one during the daytime, and generally much greater than one in the evening, as the occupants generated new particles by activities such as cooking. The mean daily PM$_{10}$ I/O ratios ranged from 1.0±1.6 to 3.9±7.8. Lee et al. (2002 b), who investigated PM$_{10}$ concentrations in six houses in Hong Kong, China, showed that the average levels in the kitchens exceeded those in the living rooms by 30% to 154%. The average I/O ratio was about 1.47 (±0.65). The average indoor (kitchen) to outdoor ratios was about 1.90 and the average indoor (living room) to outdoor ratios was about 1.03.

**Movement**

General human movement has frequently been shown to result in an increase in particle mass concentration. Activities such walking, cleaning, dressing, moving papers, etc., can significantly increase the concentration of supermicrometer particles
in the air. It has been demonstrated that even light activities could be a significant source of supermicrometer particles. However such physical activities do not contribute to submicrometer particles in the air, which are basically non-resuspendable under conditions present in residential or workplace environments (Thatcher and Layton, 1995).

In the study by Monn et al. (1997), the I/O ratio of PM$_{10}$ measured in 17 houses with ‘human activity’ was 1.40, and the PM$_{2.5}$ ratio (measured in only one house with general human activity) was 1.23. Jones et al. (2000) showed that in one of the investigated houses elevated average PM$_{10}$ concentrations were not linked to movement, although in another house they were, and increased from about 10 to about 40 $\mu$g m$^{-3}$ as a result of the movement of people. As in the case of other sources, the short-term increase in concentration as a result of movement is much higher than the effect of movement on longer term average values. For example, Wallace (2000 b) reported that walking on a carpet or waving one’s arms near the particle monitor could significantly affect the reading. Shirts of different fabric, such as flannel, cotton or polyester, could increase levels of coarse particles by factors of 5-10.

Physical movement always occurs in schools, where large numbers of children are very active. For example, Roorda-Knape et al. (1998) measured PM$_{10}$ concentrations in classrooms during school hours and found that the levels were highly variable and often much higher than those outdoors. The I/O PM$_{10}$ ratio ranged from 0.79 to 1.4, with an average of 1.14. Janssen et al. (1997) also found significantly higher PM$_{10}$ concentrations in classrooms compared to corresponding outdoor concentrations, and these were not correlated with distance of the school from a motorway, traffic intensity, or percentage of time downwind. The mean PM$_{10}$ indoor/outdoor ratios
ranged from 1.33 to 2.19 (ratios not actually provided in the paper, but assessed on the basis of the presented data).

The overall conclusion from these few studies, which attempted to quantify the effect of human activities and general movement on particle mass concentration levels, is that the effect is identifiable in most cases, although not always. In general, the I/O average ratios related to movement have been reported to be lower than those for smoking and cooking, those for PM$_{10}$ not exceeding 1.4. The reason for conflicting results as to the impact of movement on particle mass concentration levels indoors is most likely related to the relatively weaker strengths of this particle source, which, under conditions of higher exchange rates, may not be high enough to enable quantification.

**General Trends in I/O Ratio**

The results from all the studies discussed above, in which I/O ratios of PM$_{10}$ and PM$_{2.5}$ concentrations have been investigated in naturally ventilated buildings in the presence of operating indoor sources, as well as from other relevant reported studies on I/O ratios, are compiled in Figures 3.3A and B. The conclusions that can be drawn from analysing the data presented in these figures are as follows.

For PM$_{10}$, the reported range of average I/O ratios in the presence of indoor sources has been from 1.14 to 3.91 with median value of 1.47; for PM$_{2.5}$, the range has been from 1.00 to 2.40 with median value 1.21. As expected, almost all these values are greater than one and are higher than the values for the cases where no indoor source operated. The spread of the values is also much greater than in the absence of indoor sources. However, despite the differences in the environments investigated, differences in air exchange rates, and the dominant impact of different sources in
these environments, most of the reported I/O ratios are below two, with only a few significantly above this value. It is interesting to note that this range of average I/O ratios, while higher than in cases of no indoor sources, is still relatively narrow, considering the large variation in short-term concentrations of PM$_{10}$ and PM$_{2.5}$ resulting from emissions from the sources, and often increasing temporarily by one to two orders of magnitude. While this observation could be useful for conducting assessments of expected indoor/outdoor relationship in the absence of data for a particular environment, it should, however, be applied with caution, as under certain conditions the impact of indoor sources can be much higher than the general trend.

Figure 3.3 A: Summary of some reference data of Indoor/Outdoor PM$_{10}$ ratio under indoor particle source conditions
3.4.3 The Effect of Indoor Sources on Particle Indoor Number and Volume Size Distribution and the I/O Ratios

In recent years, an increased number of reported studies have focused on measurements of particle number concentration. Due to the differences in instrumentation used, such studies tend to employ somewhat different experimental designs than studies on PM$_{10}$ or PM$_{2.5}$ indoor characteristics. In particular, instrumentation for particle number or size distribution measurements enables collection of real-time data and, therefore, there has been a tendency to include more investigations on short-term variation in particle concentration, time series analyses, as well as investigations of the impact of individual sources. At the same time, such studies tend to generate fewer results on longer-term average concentration values.
For example, the number concentrations due to selected indoor combustion sources were determined in a first floor apartment in Taipei, Taiwan, China, by Li et al. (1992, 1993b). The particle sizer used was a high-resolution electrical mobility aerosol spectrometer, which including a differential mobility particle sizer (DMPS) and a condensation particle counter (CPC). The results showed that I/O ratios before active aerosol generation were about 0.75 and 0.95 for ultra-fine particles (0.01 – 0.1 \( \mu \text{m} \)) and submicrometer particles (0.01 – 1 \( \mu \text{m} \)), respectively. However, particle number concentrations for the background, smouldering cigarettes, burning mosquito coils, and joss sticks varied from \( 10^4 \) to \( 10^7 \) particles \( \text{cm}^{-3} \) for conditions where the windows and doors were kept closed. Thus, operation of the combustion sources resulted in a temporary increase in particle number concentration by up to three orders of magnitude.

Measurements of the effect of cigarette smoking on particle number concentration using SMPS was conducted in Brisbane, Australia by Morawska et al. (1995, 1997) in three residential houses and one naturally ventilated university club. In the residential houses, measurements commenced after one cigarette was smoked in the centre of a room selected for investigations, and were conducted (also in the centre of the room) until the ETS peak was no longer clearly distinguishable from the background indoor particle distribution. Measurements in the club were performed with varying numbers of smokers present. Background ambient aerosol condition was monitored during these measurements. The study showed that the initial concentration levels after one cigarette has been smoked in the residential houses were often above \( 10^5 \) particles \( \text{cm}^{-3} \) and still easily detectable up to two hours after smoking had ceased. The measurements in the club showed that, when the number of smokers was large, ETS concentration in the room was very high, up to \( 5 \times 10^4 \)
particles cm$^{-3}$, and characterised by a stable, well defined broad peak. However, when the number of smokers was small (<10), the smoke concentration decreased rapidly with time and the ETS peak was not detectable 10-15 min after a cigarette had been smoked.

Number concentration and size distribution of submicrometer particles were investigated in four residential houses in Brisbane, Australia, by Lyons and Morawska (1996), using SMPS. Sources such as cooking, clothes washing, vacuuming, lawn mowing, and smoking resulted in particle number concentrations of $(3.1-5.0)\times10^6$, $2.0\times10^5$, $4.4\times10^4$, $7.8\times10^7$, and $4.8\times10^4$ particles cm$^{-3}$, respectively.

No simultaneous measurements of outdoor concentrations were conducted, although these concentrations could be compared to the average background concentration for Brisbane of $7.4\times10^3$ cm$^{-3}$ (Morawska et al., 1998), indicating that the increase in concentrations due to these activities ranged up to four orders of magnitude.

A larger study conducted in 14 residential houses in Brisbane and discussed above in relation to PM$_{2.5}$ concentrations also included investigations of particle number concentration in the size range from 0.007 to 0.808 μm, as measured by a condensation particle counter (CPC) (Morawska et al., 2003). The average number concentrations were $(18.2\pm3.9)\times10^3$ particles cm$^{-3}$ during indoor activities and $(12.4\pm2.7)\times10^3$ particles cm$^{-3}$ under non-activity conditions. The maximum concentrations were in excess of $8\times10^5$ particles cm$^{-3}$. The median ratio of the 24-h indoor to outdoor concentrations was 1.03.

The study quoted above, conducted in nine non-smoking Boston-area homes by Long et al. (2000), size distribution of particles were also investigated using TEOM, SMPS and APS. Particle mean hourly volume concentrations in the size ranges
investigated of 0.02-0.1, 0.1-0.5, 0.7-2.5, and 2.5-10 µm were for indoor air: 0.71 µm³ cm⁻³ (SD = 1.8), 6.3 µm³ cm⁻³ (SD = 9.8), 2.3 µm³ cm⁻³ (SD = 9.2), and 2.4 µm³ cm⁻³ (SD = 3.8), respectively, and for outdoor air: 0.49 µm³ cm⁻³ (SD = 0.59), 6.3 µm³ cm⁻³ (SD = 5.1), 2.5 µm³ cm⁻³ (SD = 2.7), and 2.7 µm³ cm⁻³ (SD = 4.7), respectively. Again, for most of the size intervals, the standard deviations for the indoor data were larger than those for outdoor data, indicating higher short-term variability of indoor sources. Indoor and outdoor hourly concentrations (n > 2000) were found to be highly correlated (p ≤ 0.0001), with Spearman correlation coefficients of 0.69, 0.63, 0.82, 0.69, and 0.35, respectively, for the investigated size ranges. The authors suggested that the low correlation coefficient for the coarse particles was probably the result of both higher depositional losses and a greater contribution from indoor sources. As part of the study, a controlled experiment involving the mopping of hardwood and linoleum floors with a dilute ‘Pine Sol’ solution was conducted. This was in addition to instances of the homeowner using the ‘Pine Sol’ product to disinfect a bathroom toilet. It was shown that more than 50% of the particles (by volume) generated during these cleaning activities were ultra fine particles, the number concentrations of which increased by between 7 to 100-fold. These increases in particle number also resulted in an increase in the PM₂.₅ mass concentration as discussed above, revealing the strong impact of particle number on mass concentration.

Measurements of the relative contributions of outdoor and indoor particle sources to indoor concentrations were conducted by Abt et al. (2000) in four non-smoking homes for one- or six-day periods in the metropolitan Boston area, Massachusetts, USA. The measurements of particle number size distribution were conducted by means of SMPS and APS. It was shown that cooking, cleaning, and indoor work
significantly increased indoor concentrations for particles larger than about 1-2 µm. However, for particles smaller than 0.5 µm, the only variable significantly associated with the generation of indoor particles was the previous 20 min spent cooking. The I/O volume concentration ratios for different size ranges, calculated on the basis of data provided in the paper, showed that the ratios fluctuated between 1.24 for particles in the range from 0.02 to 0.1µm, to 0.95 for particles from 0.2 to 0.3 µm, and 1.14 for particles from 0.4 to 0.5 µm, and then showed a decreasing trend with the increased particle size – down to 0.60 for particles in the range from 6-10 µm.

A few conclusions can be drawn from the above review of the studies reporting indoor particle number and/or volume size distribution or total number concentration. Firstly, direct comparison of the results from the studies is not always possible as not only have different instruments been used to measure particle characteristics, operating in different size ranges, but the data have been reported differently. In particular, the data resulting from the measurements of particle number concentration has been reported either in terms of number or as particle volume distributions. Secondly, the short-term impact of indoor sources, particularly combustion sources, is even stronger on particle number than on particle mass, and the resulting concentrations can increase by up to four orders of magnitude. Thirdly, in the two studies in which I/O ratios were investigated, values somewhat above one for the smaller particles and below one for supermicrometer particles were reported. Again, direct comparison of the I/O ratios is not possible due to different particle size ranges investigated.
3.5 Indoor/outdoor relationship in mechanically ventilated buildings

The indoor/outdoor (I/O) relationship for mechanically ventilated buildings is even more complex than that for naturally ventilated ones and the factors affecting indoor concentrations for such buildings are discussed in Chapter 3.6 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003). The mechanically ventilated buildings investigated have, in most cases, been non-industrial workplaces or public buildings, such as offices, hospitals, restaurants, schools or shopping malls. Only a few mechanically ventilated residential houses have been investigated. A common characteristic of all such buildings is that the mechanical ventilation and filtration systems change the characteristics of particulate matter entering the buildings from outside in terms of concentration and size distribution, and also affect air circulation and thus particle characteristics inside the building. However, the buildings differ substantially in the presence and characteristics of the indoor sources, and therefore in the characteristics of indoor particulate matter. For example, in most office buildings there are no strong indoor particle sources such as tobacco smoking or cooking, whereas these are present in restaurants. I/O ratios investigated in a number of public buildings in several countries either in relation to particle mass or number characteristics are reviewed below.

Particle Mass

PM$_{2.5}$ concentrations were measured in 11 private air-conditioned residences in Birmingham, AL, USA, ranging from single- to two-storey dwellings with finished basements or basement garages (Lachenmyer and Hidy, 2000). In summer, mean outdoor PM$_{2.5}$ concentrations of 26.5±9.5 µg m$^{-3}$ were larger than the indoor concentrations of 16.1±5.7 µg m$^{-3}$, but the outdoor and indoor concentrations were
comparable in winter, at $12.2\pm5.1$ and $11.2\pm5.4 \, \mu g\, m^{-3}$, respectively. The results can be explained in terms of preferential use of air-conditioning in summer, as opposed to gas heating in winter and use of fireplaces in winter, and fireplace dampers being closed in summer and open in winter. The PM$_{2.5}$ I/O ratios ranged from 0.43 to 0.86 in summer, and from 0.61 to 1.86 in winter (the ratios were not provided in the paper, but have been calculated on the basis of the presented data).

In the South-eastern Brazil Indoor Air Quality Study (SEBIAQS), levels of gas and characteristic particles present in indoor and outdoor air were investigated in non-industrial office workplaces and in restaurants (Miguel et al., 1995). Twelve sites in the cities of São Paulo and Rio de Janeiro and in a rural area were analysed for the mass of particles with diameters smaller than 15 $\mu$m. Four of them were mechanically ventilated, one naturally, and seven both mechanically and naturally. The average I/O for the offices was found to be 1.31, and for the restaurant with an open pit charcoal stove, the ratio reached 2.8. In another Brazilian study, conducted in four mechanically ventilated offices in a building in Rio de Janeiro, Brickus et al. (1998) measured TSP and UV-RSP (ultraviolet respirable suspended particles, a surrogate for environmental tobacco smoke) in offices located at various levels of an office building. A summary of the results from this study is presented in Table 3.3.

**Table 3.3.** Average values of TSP and UV-RSP (ultraviolet respirable suspended particles) measured in offices Rio de Janeiro, Brazil ($\mu g/m^3$); (adapted from Brickus et al., 1998)

<table>
<thead>
<tr>
<th>Floor</th>
<th>TSP Indoor</th>
<th>TSP Outdoor</th>
<th>TSP I/O</th>
<th>UV-RSP Indoor</th>
<th>UV-RSP Outdoor</th>
<th>UV-RSP I/O</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st</td>
<td>91.4</td>
<td>141.4</td>
<td>0.7</td>
<td>10.4</td>
<td>5.8</td>
<td>1.8</td>
</tr>
<tr>
<td>9th</td>
<td>28.7$^a$</td>
<td>32.8$^a$</td>
<td>0.9</td>
<td>7.7</td>
<td>5.5</td>
<td>1.4</td>
</tr>
<tr>
<td>13th</td>
<td>53.5</td>
<td>58.8</td>
<td>0.9</td>
<td>6.6</td>
<td>5.0</td>
<td>1.3</td>
</tr>
<tr>
<td>25th</td>
<td>66.6</td>
<td>43.5</td>
<td>1.5</td>
<td>8.1</td>
<td>1.8</td>
<td>4.5</td>
</tr>
</tbody>
</table>

$^a$ Inhaled suspended particles.
According to the authors’ interpretation, as a consequence of the heavy street traffic, outdoor TSP values were very high, especially outside of the offices located at ground level. However, I/O ratios were in most of cases below unity, indicating a lack of significant indoor sources of TSP particle mass. At the same time, indoor UV-RSP concentrations were higher than those outdoors for offices on all floors, indicating a significant impact of human activities on particle mass concentration in this size range. This impact is further demonstrated by the I/O ratios, which are greater than one for all the offices, but do not exceed two, apart from the office on the 25th floor (in which case the outdoor concentration was significantly lower than on the other levels).

The average PM$_{10}$ concentrations investigated in domestic residences, restaurants, and shopping malls in Hong Kong, China, by Lee et al. (2002 a) were found to be two or three times higher than those measured in offices and air-conditioned classrooms. The mean PM$_{10}$ concentrations in outdoor air ranged from 30 to 182 $\mu$g/m$^3$. The I/O ratios of PM$_{10}$ were 0.98 for homes, 0.60 for offices, 0.82 for air-conditioned classrooms, 1.44 for shopping malls, and 3.77 for restaurants.

TSP and PM$_{10}$ concentration levels were investigated in a mechanically ventilated children’s day care centre in the city of Imatra, Finland, by Partti-Pellinen et al. (2000). The study showed that while the concentrations were very low during holidays and weekends, on weekdays indoor particle-generating and resuspending activities, as well as opening of doors and windows, increased indoor particle concentrations. Indoor particle concentrations remained well below outdoor concentrations under conditions of air recirculation combined with filtration; however, mechanical exhaust alone combined with a filter in the incoming duct was rather inefficient, and indoor particle levels could momentarily be even higher than
those outdoors. The indoor/outdoor TSP and PM$_{10}$ ratios varied from 0.06 to 0.61, with the average of 0.32, and from 0.11 to 2.12, with the average of 0.39, respectively (the ratios were not provided in the paper, but have been calculated on the basis of the presented data).

PM$_{2.5}$ concentrations were measured by Patterson and Eatough (2000) both indoor, in a carpeted classroom of a mechanically ventilated elementary school, and at an adjacent outdoor monitoring site in Lindon, UT, USA. The indoor PM$_{2.5}$ mass data displayed prominent peaks while students were in the classroom, and at such times there was a low indoors/outdoors correlation. The indoor/outdoor correlation was higher in the absence of the students and staff in the building, suggesting a substantial influence of human activity on PM$_{2.5}$. The PM$_{2.5}$ I/O ratios was 0.4 for all samples, 0.5 for day samples and 0.37 for night samples (the ratios were not provided in the paper, but have been calculated on the basis of the presented data).

Investigations conducted indoor and outdoor at three hospitals in Salt Lake City, Utah, USA, and including daily measurements of 24-hour PM$_{10}$ concentrations, showed that the average I/O ratio of PM$_{10}$ was about 0.58, with the range from 0.5 to 0.72. (Lillquist et al., 1998).

In summary, the results of the studies conducted in mechanically ventilated public buildings and non-industrial workplaces show a large variation in the TSP, PM$_{10}$ and PM$_{2.5}$ I/O ratios, which range from 0.06 to >2 (the value of 4.5 for UV-RSV was even higher), i.e. over two orders of magnitude. Large variations are expected for mechanically ventilated buildings, since the design characteristics and operating parameters of the buildings’ ventilation and filtration systems can differ substantially. While in the case of naturally ventilated buildings some general assessments of the expected relationship between indoor and outdoor average...
concentrations can be made based on a general understanding of the specific environment and the data available in the literature, such predictions would not be reliable for mechanically ventilated buildings, without modelling, and would require a number of input parameters relevant to the specific locations. For more information on modelling of particle indoor concentrations see chapter 3.8 of the book - Indoor Environment: Airborne Particles and Settled Dust (Morawska and Salthammer, 2003).

**Particle Number, Number and Volume Size Distribution**

A comprehensive investigation of particle size distribution has been reported in just one study conducted in a mechanically ventilated residential house. Wallace and Howard-Reed (2002) measured particle size distribution using a number of different instruments in a mechanically ventilated townhouse located in Reston, VA, USA. The instruments included SMPS, APS, optical scattering instruments (Climets), and passive nephelometers. No seasonal pattern for particle concentration was evident from the study; however, a strong diurnal pattern appeared for some size ranges. The ultra-fine particle concentrations showed a strong increase in both number and volume between 6:00 pm and 8:00 pm, corresponding to cooking with a gas stove, and a strong decrease between 12:00 am and 5:00 am. Levels of particles larger the 1 µm showed an increase during general personal activity. When no source was operating, the I/O ratios measured by the Climets ranged from 0.13 to 0.24. In the presence of operating indoor sources, the mean number concentration of ultra-fine particles was about eight times higher compared to the situation of no operating source, while volume concentrations for both the APS and the SMPS were higher by factors ranging from 2 to 20 for different size fractions. When a source was operating, the I/O ratios for 0.5 – 1 µm and 1 - 2.5 µm were 0.77 and 0.81,
respectively, and for 2.5 - 5 µm and 5 - 10 µm were 1.45 and 1.31, respectively. For all the data, the I/O ratio for the 0.3 - 2.5 µm ranged from 0.40 to 0.47, and for the 2.5 to >10 µm, from 0.79 to 1.42. Figure 3.4 presents the estimated relative contributions of indoor and outdoor sources to indoor concentration of particles in different size ranges.

![Figure 3.4](image)

**Figure 3.4**: Estimated relative contribution of indoor and outdoor sources to indoor concentrations of particles of different sizes: APS and SMPS (Wallace and Howard-Reed, 2002).

The distribution presented in Figure 3.4 can be explained in relation to the volume concentrations measured for different size ranges as follows. For ultra-fine particles, when indoor sources were operating, the particle volume averaged about 4-12 times the volume when no indoor sources are operating. For the accumulation mode, the ratio dropped to 2-3, and then, for particles above 1 µm, the ratio increased again to 5-20. The shape of this distribution implies a tendency for indoor sources to produce
either ultra-fine or coarse particles more readily than particles of intermediate size. Indoor sources were responsible for 50-80% of the ultra-fine particles, for approximately 40-50% of the particles from 100 nm to 1 µm, and for 75-90% for the particles greater than 1 µm.

In the above mentioned study of Patterson and Eatough (2000), conducted inside and outside an elementary school, while PM$_{2.5}$ correlated poorly indoors and outdoors, total particle number showed a strong indoor-outdoor relationship. With a background indoor particle number concentration of about 1.6×10$^3$ particles cm$^{-3}$, the total average daily concentration indoors was about 3.6×10$^3$ particles cm$^{-3}$ (total average day: 3.8×10$^3$ particles cm$^{-3}$; total average night: 3.5×10$^3$ particles cm$^{-3}$).

A few more studies on particle number size distributions in office buildings have been conducted. Fisk et al. (2000) measured particle number concentrations as a function of time on the second and fourth floors of a large office building with mechanical ventilation, air-conditioning, and sealed windows in St Louis, MO, USA. In this study, laser-based optical particle counters (Model 237B, Met One, Grants Pass, OR) were used to measure particle number concentration in six size ranges (0.3-0.5 µm, 0.5-0.7µm, 0.7-1.0 µm, 1.0-2.0 µm, 2.0-5.0 µm, >0.5 µm). The study showed that on the floor with normal filtration (of low efficiency for submicrometer particles), the smallest and the largest bins contained 41% and 45% of the particle mass, respectively, while on the floor with high efficiency filtration (of minimum efficiency of 95% for particles with a diameter of 0.3 µm), particles larger than 1.0 µm accounted for 89% of the particle mass. Large week-to-week variations in concentrations were found, up to a factor of approximately six between the highest and lowest concentrations. Under all the operating conditions of the ventilation and
filtration system of the building, outdoor particle number concentrations were substantially higher than the indoor concentrations for particles smaller than 2 µm. From this study it was concluded that indoor generation or resuspension of particles larger than 1 µm plays a significant role in large commercial buildings. In particular, it was discovered that at numerous times there were spikes in indoor particle concentrations at specific locations, and that these were more common for the larger particles, usually without a corresponding spike in the outdoor particle concentrations. For normal air filters, the measured average ratios of indoor to outdoor particle concentrations for workdays were 0.36, 0.21, 0.23, 0.31, and 0.54 for the first four size bins and for particles larger than 2 µm, respectively. Under high efficiency filtration, the indoor concentrations were, under certain conditions, 26 times lower than outdoor concentrations. The highest measured I/O ratios were for the largest particles (>2.0 µm). Indoor concentrations roughly tracked the outdoor ones in terms of number and mass for submicrometer particles and particles larger than 2 µm, respectively. From this study it was concluded that there was a high rate of removal of submicrometer indoor particles by processes other than ventilation or air filtration, and it was hypothesised that for these particles the deposition coefficients in this building may have greatly exceeded the commonly assumed coefficients.

Koponen et al. (2001) conducted studies on the first floor of an office building near downtown Helsinki, Finland. The measurements were conducted using two identical differential mobility particle sizer systems (DMPS) in the size range from 7 to 500 nm. The daytime mean concentrations were found to be in the range from $1 \times 10^4$ to $3 \times 10^4$ particle cm$^{-3}$ and the nighttime of the order of $5 \times 10^3$ particle cm$^{-3}$. I/O ratios were about 0.1, 0.15, and 0.3 for particles in the ranges 8 - 25 nm, 25 - 90 nm, and
90 - 500 nm, respectively. Indoor concentrations followed the outdoor values with about a 20 min delay (for ventilation ‘off’ and air exchange rate of 0.3 h⁻¹) or a 10 min delay for ventilation ‘on’ (air exchange rate of 3.7 h⁻¹). The effect of ventilation on particle size distribution was noticeable for particles between 20 and 80 nm and the I/O ratio was larger when ventilation was ‘off’ and smaller when it was ‘on’. The I/O ratios for the size range from 80 to 150 nm did not seem to be affected by ventilation rate, but for particles larger than 150 nm, the effect of ventilation on I/O ratio was found to be opposite to that seen for the smaller particles. There was no clear explanation for the trends observed and the role of the filter was hypothesised.

Measurements of particle number and mass were conducted in a non-smoking office building in Helsinki, Finland by Luoma and Batterman (2001). Particle number concentrations were measured using three optical particle counters, each of which counted particle in six size channels (>0.3, 0.5, 1, 5, 10, and 25 µm). Particle mass concentrations were determined gravimetrically using open-face filter cassettes. Particle concentration indoors averaged 13±3 µgm⁻³ indoors and 20±11 µgm⁻³ outdoors. A general conclusion of the study was that the daily trends varied for particles in different size ranges. For particles <1 µm diameter, concentrations increased indoors around coffee and lunchtimes, possibly due to smoke escaping from the smoking room, and decreased significantly in the afternoon. In contrast, outdoor trends differed from day to day. Concentrations of particles in the range from 5-25 µm appeared to vary largely randomly and were uncorrelated with known indoor and outdoor sources or activities. Particles in the size range from 1-5 µm showed characteristics of both fine and coarse particles, with concentrations unrelated to the time of day. Specifically, the particle concentrations that were highly correlated included: indoor 0.3-0.5 and 0.5-1 µm particles (r=0.88); 1-5 µm particles
with both 0.5-1 μm (r=0.38) and 5-10 μm (r=0.84) particles; and 5-10 with 10-25 μm particles (0.89). Indoor and outdoor concentrations for 0.3-0.5 μm and 0.5-1 μm particles were moderately correlated (r=0.72 and r=0.69 for), indicating a low efficiency of the filters in this building. The activities that were, however, not consistently associated with the increased concentrations of 0.3-0.5 and 0.5-1 μm particles included: (1) bioaerosol sampling, which took place every 30 min and involved installing and removing agar plates from the impactor; (2) handling papers near the monitoring site; (3) people leaving the room and coming back; (4) a person wearing a coat and standing near the monitoring site. Particles in the size range 1-25 μm generally increased as a whole, over the entire range, and were related to: (a) bioaerosol sampling; (b) frequent walks past and between the centre tables, in the vicinity of the monitoring site; (c) opening boxes (containing office supplies) at the centre tables; (d) an occupant putting on a sweater.

Based on these three studies conducted in office buildings, it can be concluded that there are still major difficulties in interpretation of the results of I/O ratios of particle size distributions in mechanically ventilated buildings and that much more research is needed to explain and quantify the role of different mechanisms contributing to particle concentration levels and size distribution characteristics in mechanically ventilated large buildings.

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CHAPTER 4

THE RELATIONSHIP BETWEEN INDOOR AND OUTDOOR AIRBORNE PARTICLES IN THE RESIDENTIAL ENVIRONMENT

Lidia Morawska, Congrong He, Jane Hitchins, Dale Gilbert* and Sandhya Parappukkaran

Centre for Medical and Health Physics, Queensland University of Technology, Brisbane, QLD 4001, Australia

*Built Environment Research Unit, Qld Dept. of Public Works and Housing, Brisbane, QLD 4001, Australia

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STATEMENT OF JOINT AUTHORSHIP

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

Authors: Lidia Morawska, Congrong He, Jane Hitchins, Dale Gilbert and Sandhya Parappukkaran

Lidia Morawska
Developed experimental design and scientific method; interpreted data and wrote manuscript

Congrong He (candidate)
Contributed to experimental design and scientific method; conducted part of measurements; analysed and interpreted data; assisted with manuscript

Jane Hitchins
Contributed to experimental design and scientific method; conducted part of measurements; assisted with manuscript

Dale Gilbert
Assisted with data interpretation and manuscript

Sandhya Parappukkaran
Assisted with part of measurements and manuscript
Abstract

The relationship between indoor and outdoor airborne particles was investigated for 16 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 minimum ventilation, unless air exchange rate is known, thus allowing for estimation of the “delay constant”.

Keywords: air pollution; indoor air quality; submicrometer particle; supermicrometer particles; PM$_{2.5}$; ventilation
4.1 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 health 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.
4.2 Experimental method

4.2.1 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.

4.2.2 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 concentration 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.
4.2.3 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 $\mu$m and particles with size less than 0.8 $\mu$m. 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.

4.2.4 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 minimum 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$^{-1}$ 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.
4.2.5 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).
4.3 Results and discussion

4.3.1 Non-simultaneous measurements

4.3.1.1. 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 4.1. 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 4.1). 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.
Table 4.1. 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\(^{-3}\)).

<table>
<thead>
<tr>
<th>House ID</th>
<th>Indoor Concentration</th>
<th>Indoor NMD</th>
<th>Outdoor NMD</th>
<th>I/O Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SMPS (×10(^3))</td>
<td>APS</td>
<td>SMPS (µm)</td>
<td>APS (µm)</td>
</tr>
<tr>
<td>H1</td>
<td>2.9±0.4</td>
<td>6.03±2.0</td>
<td>0.032</td>
<td>0.94</td>
</tr>
<tr>
<td>H3</td>
<td>4.3±0.6</td>
<td>1.47±0.1</td>
<td>0.034</td>
<td>0.87</td>
</tr>
<tr>
<td>H4</td>
<td>3.5±0.2</td>
<td>1.06±0.04</td>
<td>0.023</td>
<td>0.87</td>
</tr>
<tr>
<td>H5</td>
<td>10.5±0.2</td>
<td>5.2±0.15</td>
<td>0.038</td>
<td>0.9</td>
</tr>
<tr>
<td>H6</td>
<td>7.8±0.4</td>
<td>1.55±0.08</td>
<td>0.023</td>
<td>0.95</td>
</tr>
<tr>
<td>H7</td>
<td>0.7±0.09</td>
<td></td>
<td>0.028</td>
<td>0.92</td>
</tr>
<tr>
<td>H8</td>
<td>10.9±0.4</td>
<td>1.32±0.09</td>
<td>0.023</td>
<td>0.72</td>
</tr>
<tr>
<td>H9</td>
<td>8.0±0.4</td>
<td>1.43±0.06</td>
<td>0.025</td>
<td>0.96</td>
</tr>
<tr>
<td>H12</td>
<td>2.0±0.4</td>
<td>4.72±0.08</td>
<td>0.023</td>
<td>0.95</td>
</tr>
<tr>
<td>H13</td>
<td>12.9±1.9</td>
<td>2.81±0.13</td>
<td>0.023</td>
<td>0.92</td>
</tr>
<tr>
<td>H14</td>
<td>3.7±0.2</td>
<td>1.11±0.04</td>
<td>0.029</td>
<td>0.81</td>
</tr>
<tr>
<td>H15</td>
<td>7.7±0.3</td>
<td>4.15±0.05</td>
<td>0.033</td>
<td>0.84</td>
</tr>
<tr>
<td>H16</td>
<td>18.7±2.7</td>
<td>1.11±0.05</td>
<td>0.033</td>
<td>1.23</td>
</tr>
<tr>
<td>H17</td>
<td>8.4±0.4</td>
<td>1.80±0.05</td>
<td>0.024</td>
<td>0.91</td>
</tr>
<tr>
<td>H18</td>
<td>16.1±0.8</td>
<td>1.99±0.08</td>
<td>0.027</td>
<td>0.74</td>
</tr>
<tr>
<td>H19</td>
<td>0.8±0.09</td>
<td>1.77±0.05</td>
<td>0.024</td>
<td>0.99</td>
</tr>
<tr>
<td>Average</td>
<td>7.4±5.4</td>
<td>2.5±1.67</td>
<td>0.028</td>
<td>0.91</td>
</tr>
<tr>
<td>Max</td>
<td>18.7</td>
<td>6.03</td>
<td>0.038</td>
<td>1.23</td>
</tr>
<tr>
<td>Min</td>
<td>0.7</td>
<td>1.06</td>
<td>0.023</td>
<td>0.72</td>
</tr>
</tbody>
</table>

*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.*
4.3.1.2 Supermicrometer particle concentrations

Table 4.1 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.

4.3.1.3 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).

4.3.1.4 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 4.1). 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 4.1).

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.
4.3.2. 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.

4.3.2.1. 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 4.2. Figure 4.1 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 4.2 and Figure 4.1 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.
Table 4.2. 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$^a$

<table>
<thead>
<tr>
<th>House ID</th>
<th>Normal Ventilation Condition</th>
<th>Minimum Ventilation Condition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>APS Ratio</td>
<td>APS $^2$</td>
</tr>
<tr>
<td>H1</td>
<td>0.98</td>
<td>0.13</td>
</tr>
<tr>
<td>H3</td>
<td>1.42</td>
<td>0.33</td>
</tr>
<tr>
<td>H4</td>
<td>0.91</td>
<td>0.26</td>
</tr>
<tr>
<td>H5</td>
<td>0.91</td>
<td>0.31</td>
</tr>
<tr>
<td>H6</td>
<td>0.98</td>
<td>0.60**</td>
</tr>
<tr>
<td>H7</td>
<td>0.94</td>
<td>0.69**</td>
</tr>
<tr>
<td>H8</td>
<td>0.82</td>
<td>0.41*</td>
</tr>
<tr>
<td>H9</td>
<td>1.08</td>
<td>0.03</td>
</tr>
<tr>
<td>H12</td>
<td>0.91</td>
<td>0.72**</td>
</tr>
<tr>
<td>H13</td>
<td>0.97</td>
<td>0.94**</td>
</tr>
<tr>
<td>H14</td>
<td>0.79</td>
<td>0.71**</td>
</tr>
<tr>
<td>H15</td>
<td>0.91</td>
<td>0.67**</td>
</tr>
<tr>
<td>H16</td>
<td>0.93</td>
<td>0.64**</td>
</tr>
<tr>
<td>H17</td>
<td>0.88</td>
<td>0.81**</td>
</tr>
<tr>
<td>H18</td>
<td>0.89</td>
<td>0.90**</td>
</tr>
</tbody>
</table>

|          | 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    |

$^a$ 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.
Figure 4.1. 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.
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 4.2.
4.3.2.2. Minimum ventilation conditions

The results of CPC, APS and DustTrak measurements under minimum ventilation conditions are also presented in Table 4.2. The ranges of particle concentrations were (4.9 ~ 21.3)×10³ particles cm⁻³ indoor and (5.0 ~ 52.8)×10³ particles cm⁻³ outdoor, for the CPC; 0.46 ~ 3.74 particles cm⁻³ indoor and 0.47 ~3.98 particles cm⁻³ outdoor, for the APS; and 5.4 ~ 18.0 µg m⁻³ indoor and 6.2 ~ 18.0 µg m⁻³ outdoor, for the PM₂.₅.

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 PM₂.₅ concentrations was 1.08±0.22. It also can be seen from Table 4.2 that for many
Houses the indoor/outdoor ratios for larger 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 4.3. 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 4.3, it can be seen that introduction of a “delay” factor significantly improves the correlation between indoor and outdoor concentrations.
Figure 4.3. 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. $R_o^2$ and $R_s^2$ values are the correlation coefficient of indoor/outdoor and shifted indoor and outdoor concentrations, respectively.

\[ R_o^2 = 0.43 \quad R_s^2 = 0.53 \]

Delay: 6 minutes

\[ R_o^2 = 0.0 \quad R_s^2 = 0.46 \]

Delay: 10 minutes

\[ R_o^2 = 0.0 \quad R_s^2 = 0.79 \]

Delay: 24 minutes
4.4. 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 ($7.1 \times 10^3$ and $7.4 \times 10^3$ particles cm$^{-3}$, respectively), measured in this study, are very close to the value of $7.4 \times 10^3$ 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 minimum 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 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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CHAPTER 5

CHARACTERISTICS OF PARTICLE NUMBER AND MASS CONCENTRATIONS IN RESIDENTIAL HOUSES IN BRISBANE, AUSTRALIA

Lidia Morawska¹, Congrong He¹, Jane Hitchins¹, Kerrie Mengersen² and Dale Gilbert³

¹International Laboratory of Air Quality and Health, Queensland University of Technology, Brisbane, QLD 4001, Australia

²School of Mathematical and Physical Sciences, The University of Newcastle, University Drive, Callaghan, NSW 2308, Australia

³Built Environment Research Unit, Queensland Department of Public Works and Housing, Brisbane, QLD 4001, Australia

STATEMENT OF JOINT AUTHORSHIP

Title: Characteristics of particle number and mass concentrations in residential houses in Brisbane, Australia

Authors: Lidia Morawska, Congrong He, Jane Hitchins, Kerrie Mengersen and Dale Gilbert and

Lidia Morawska
Developed experimental design and scientific method; interpreted data and wrote manuscript

Congrong He (candidate)
Contributed to experimental design and scientific method; conducted part of measurements; analysed and interpreted data; assisted with manuscript

Jane Hitchins
Contributed to experimental design and scientific method; conducted part of measurements; assisted with manuscript

Kerrie Mengersen
Contributed to data analysed and interpreted data; assisted with manuscript

Dale Gilbert
Assisted with data interpretation and manuscript
Abstract

Diurnal variation of indoor submicrometer (0.007 – 0.808 µm) particle number and particle mass (approximation of PM$_{2.5}$) concentrations was investigated in fifteen houses in a residential suburb of Brisbane during winter in 1999. Continuous monitoring for more than 48 hours was conducted using a condensation particle counter (CPC) and a photometer (DustTrak) in the kitchen of each house, and the residents kept diaries of the activities conducted. In addition, data from a central monitoring station was used to investigate indoor/outdoor relationships. The results show that there were clear diurnal variations in both particle number and approximation of PM$_{2.5}$ concentrations, for all the investigated houses. The pattern of diurnal variations varied from house to house, however, there was always a close relationship between the concentration and human indoor activities. The average number and mass concentrations during indoor activities were $(18.2 \pm 3.9) \times 10^3$ particles cm$^{-3}$ and $(15.5 \pm 7.9)$ µg m$^{-3}$ respectively, and under non-activity conditions, $(12.4 \pm 2.7) \times 10^3$ particles cm$^{-3}$ $(11.1 \pm 2.6)$ µg m$^{-3}$, respectively. In general, there was a poor correlation between mass and number concentrations and the correlation coefficients were highly variable from day to day and from house to house. This implies that conclusions cannot be drawn about either one of the number or mass concentration characteristics of indoor particles, based on measurement of the other. The study also showed that it is unlikely that particle concentrations indoors could be represented by measurements conducted at a fixed monitoring station due to the large impact of indoor and local sources.

Keywords: indoor particles; indoor air quality; particle number concentration; submicrometer particles; indoor PM$_{2.5}$
5.1 Introduction

Understanding of human exposure to indoor particles is important to enable exposure control and reduction. The earlier studies on indoor particles provided information and background knowledge mainly on mass concentrations and on coarse particles, such as total suspended particles (TSP), \( \text{PM}_{10} \) and some also on \( \text{PM}_{3.5} \) fractions (mass concentration of particle matter with aerodynamic diameter smaller than 10 \( \mu \text{m} \), 3.5 \( \mu \text{m} \), respectively). This was due to the limited understanding of the relationship between particle size and the health effects they cause, as well as to instrumental limitations. More recently, a few studies on the indoor fine particle mass and number concentrations have been published, however, the available information on fine and ultra fine particles indoors, both in terms of mass and number concentration, and the relationship between mass and number concentration, is still relatively limited.

Another clear weakness in the databases provided by previous studies on indoor particles is the lack of information about the spatial and temporal or short-term variations of fine particle concentration indoors. One of the main reasons for this is that traditional filter sampling methods require sampling times in the range between 24 to 48 hours to collect a sufficient mass of particles. Such sampling methods are thus unsuitable for conducting real-time particle concentration studies. On the other hand, the TEOM (Tapered Element Oscillating Microbalance monitor) instrument commonly used for real-time outdoor measurements of particle mass (Soutar et.al. 1999), is not the most convenient instrument to be used indoors, due to its relatively large size, making it a nuisance for the occupants. Its high flow rate also affects the indoor air exchange rates and thus pollutant concentrations. In consequence, it is difficult to accurately estimate the risk due to particle exposure by using the existing
databases because indoor particle levels may exhibit significant short-term variability (e.g.; Brauer et al., 1999), which can potentially have significant health implications.

There are a few studies in the literature that have simultaneously and continually measured indoor fine particle number and mass concentration for periods up to 48 hours (Long et al., 2000; Wigzell et al., 2000). An emerging conclusion from these studies is that the pattern of diurnal variation of indoor fine particle concentrations is different from the pattern of diurnal variation of outdoor fine particle concentrations, and that indoor activities may significantly affect fine particle concentration indoors. Continuous real-time monitoring of indoor fine particles can provide the required information on the impact of the sources with time-varying intensity on indoor fine particle concentrations and thus lead to improvement in the understanding and assessment of exposure.

In order to address this need and to better understand exposure to indoor particles, the objectives of this study were to: 1) investigate the diurnal variation of particle number concentration and particle mass concentration (approximation of PM$_{2.5}$) in 15 residential houses; 2) quantify the impact of indoor activities on indoor particle concentration levels based on real-time measurements and time-activity information; and 3) compare the indoor particle concentrations with ambient particle concentrations measured at a central monitoring station. The present study is part of a larger program, some of the outcomes of which have already been published or submitted for publication (Morawska et al., 2001; Hargreaves et al, 2003; Ayoko et al., 2003).
5.2 Experimental methods

5.2.1 Sampling site and houses

A residential suburb on the south-eastern side of Brisbane, about 10km from the city centre and of reasonably flat topography was chosen as the measurement site. Fourteen houses in this suburb and one additional house for comparison, located to the east of the city were identified for the study. Ideally, houses for the study should be identified through a random selection process, however it was not feasible to use such a process for this study for two reasons; firstly, the houses had to be selected from one area to remove uncertainties related to additional factors, such as variation in topography, socioeconomic status, etc., and secondly, the studies were highly intrusive to the occupants, therefore acceptance rate was very low. The houses investigated in this study differed in age (from 2 to 100 years), construction material (timber, brick), stove type (electric, gas) and design (high set, low set). High set means that the house is elevated above ground on timber or brick stumps and low set, that the house is built directly on the ground. House design and material characteristics have an effect on the air exchange rate, but as this parameter is not included in the analyses presented in this paper, detailed house characteristics are not provided. In general, air exchange rate was estimated to range for all the houses in this study from 0.3 to 6.4 for normal ventilation conditions, and from 0.1 and 2.0, for minimum ventilation condition (all windows and doors closed). More details about the houses can be found in Morawska et al., 2001; Hargreaves, et al., 2003 and Ayoko et al., 2003. The majority of the houses were occupied by non-smokers, with Houses 5, 12 and 17 the only exceptions. However, on occasions there were visitors to some houses who smoked. The occupants of Houses 5 and 17 however never smoked inside. All the houses were naturally ventilated.
5.2.2 Instrumentation

The real-time total number concentrations of submicrometer particles in the range from 0.007 to 0.808 µm were measured using the TSI Model 3022A Condensation Particle Counter (CPC) (TSI Incorporated, St. Paul, MN, USA), with an inlet impactor (0.0508 cm nozzle) and aerosol flow rate of 0.3 L/min.

The TSI Model 8520 DustTrak aerosol monitor (TSI Incorporated, St. Paul, MN, USA) with a 2.5 µm inlet was used to measure the real-time approximation of PM$_{2.5}$ concentration. It should be noted that the DustTrak operates based on a light scattering technique where the amount of scattered light is proportional to the volume concentration of the aerosol. The approximation of PM$_{2.5}$ values obtained in this study using this instrument are not actual gravimetric values, as the instrument was not calibrated for each specific aerosol studied. It was, however, compared against TEOM (Tapered Element Oscillating Microbalance monitor) for a general type of indoor aerosol, from which correction factors were derived enabling better estimation of PM$_{2.5}$ than that based on the manufacturer’s calibration.

These instruments were chosen as the most suitable for an indoor study because of: 1) the low flow rates required thus having little impact on particle concentrations indoors; 2) their quiet operation causing less intrusion to the occupants; and 3) the short sampling times of 10 and 30 seconds respectively for the CPC and DustTrak, and thus providing real-time data.

Comparison of the indoor approximation of PM$_{2.5}$ and particle number concentrations with respective outdoor concentrations was conducted using the data collected from the Air Monitoring and Research Station (AMRS). The Station,
located in the Brisbane CBD is part of the South East Queensland monitoring network, and is operated by the Queensland Environmental Protection Agency. In addition to other parameters, PM$_{10}$ concentrations are monitored in the Station using a TEOM (50°C R&P 1400a) as well as particle number concentration using the TSI model 3934 Scanning Particle Sizer (SMPS) operating in the size range from 0.016 to 0.7 µm. Unfortunately PM$_{2.5}$ is not monitored in the Station.

5.2.3 Measurements

Particle number concentrations and the approximation of PM$_{2.5}$ concentrations were measured simultaneously for more than 48 hours in the kitchen of all the houses during March and July 1999, which is autumn/winter time in Brisbane. The CPC and DustTrak were placed side-by-side and positioned on average two metres from the stove in the kitchen. These measurements followed a few hours of intensive monitoring of particle number concentration and size distribution, approximation of PM$_{2.5}$, fungi, VOC’s and NO$_x$ concentrations under different ventilation conditions and with all the known indoor particle sources switched off. Some of the results of these investigations have been published elsewhere (Morawska et al., 2001). The occupants of the houses were required to fill in a diary, noting the time and duration of any activity conducted during the time of the measurements.

5.2.4 Data processing, correction and analysis

All statistical analyses (correlation, regression, t-test, one-way ANOVA) were conducted using a statistical analysis software package, SPSS for Windows version 10 (SPSS Inc.). Because the distribution of the concentration measures was not normal, a robust analysis in which the maximum and minimum quartiles were trimmed was employed. In addition, nonparametric tests were undertaken to confirm
the parametric results. That is, the corresponding nonparametric tests led to the same conclusions of significance/nonsignificance as the parametric tests.

5.2.4.1 Corrections for DustTrak and CPC
Since the DustTrak operates on the principle of light scattering, its response is highly dependent on the size distribution and refractive index of the sampled aerosol. This instrument was factory-calibrated with Arizona dust particles and it was reported that for finer aerosols such as commonly encountered in indoor air, the instrument’s response can be significantly higher than the true PM$_{2.5}$ value (Ramachandran et al., 2000). In order to obtain values closer to true PM$_{2.5}$ values from the data collected by the DustTrak, an additional experiment was conducted under laboratory conditions to compare the DustTrak indoor readings with the readings of a Tapered Element Oscillating Microbalance (TEOM) monitor (50ºC R&P 1400a with a URG PM$_{2.5}$ cyclone inlet). Ambient particles were used in the experiment. Based on the results of this additional experiment, the following linear regression equation was obtained:

$$PM_{2.5}^{(TEOM)} = 0.394 \times PM_{2.5}^{(DustTrak)} + 4.450$$

(with $R^2 = 0.83$)  \hspace{1cm} (1)

All the approximation of PM$_{2.5}$ data collected by the DustTrak in this study were corrected using this equation. The non-zero intercept of this equation is most likely the result of low sensitivity of the DustTrak to particles in the lower submicrometer range. While their contribution to PM$_{2.5}$ mass is normally not very high, yet it has a non-zero value. It should be noted that the values of PM$_{2.5}$ derived using this equation are still approximations of the true value, because different indoor activities may require somewhat different factors in the equation. Yet, as discussed above, it is expected that the values derived using equation 1 will differ from true values not
more than 17%, which is much less than it would be relying only on the factory calibration, when the difference sometimes is up to a factor of a few.

Recently, Yanosky et al., (2002) conducted a comparison study in an indoor environment (but without regular occupants) in which a DustTrak and a US EPA designated Federal Reference Method (FRM) PM$_{2.5}$ sampler (gravimetric method), the BGI, Inc. PQ200, were assessed to compare the two sampling methods. They found that the 24-h average DustTrak levels are well correlated with FRM levels ($R^2 = 0.859$) but show significant proportional bias. The correction equation obtained from their study was as follows:

$$PM_{2.5}^{(FRM)} = 0.33 \cdot PM_{2.5}^{(DustTrak)} + 2.25$$

(with $R^2 = 0.859$) \hspace{1cm} (1A)

Examining Equations 1 and 1A, it can be seen that the parameters in the two equations are comparable.

The CPC used in this study for the indoor measurements and the SMPS used for outdoor measurements had, due to instrumental constraints, slightly different ranges of operation. Due to this as well as to the fact that they are different systems, it was expected that they would yield somewhat different results when sampling the same aerosol. In order to provide a more accurate relationship between indoor and outdoor particle number concentrations, a comparison between the two instruments was conducted at the AMRS. Based on the results of this comparison, the following linear regression equation was obtained:

$$C_{SMPS} = 0.4213 \cdot C_{CPC} + 3019 \hspace{1cm} (with \hspace{0.5cm} R^2 = 0.67) \hspace{1cm} (2)$$
where $C_{\text{SMPS}}$ and $C_{\text{CPC}}$ are the total number concentrations measured by the SMPS and the CPC, respectively. When comparing indoor and outdoor particle number concentrations, the CPC data were adjusted using this equation.

It is acknowledged that transformation through the above equations induces uncertainty that is not appropriately accounted for in the ensuing analyses. Although the transformed values are ‘best estimates’ in that they are expected values and unbiased, any tests of significance made on their basis should be interpreted with caution.

### 5.2.4.2 Processing of particle data

The results from particle measurements were classified as follows and the average concentrations for each class were calculated:

- Based on the real-time concentration data, the total 24h average indoor concentrations were calculated for each house.

- Based on activity records, the situations indoors were classified into one of two categories: non-activity conditions and activity conditions, meaning lack or occurrence of occupant activities inside the house. As the next step, situations indoors under the non-activity conditions were further classified into one of two categories: daytime (about 06:30 - 20:00) and night-time (about 20:00 - 06:30). Under the activity conditions, the indoor situations were further classified into morning activities, day activities and evening activities. Average particle concentrations were calculated for each of these situations for each house. In addition, average concentrations were calculated for all non-activity situations that occurred during each 24 h period.
• Based on the real-time outdoor concentration data, outdoor total 24h average concentrations were calculated. Further, the outdoor concentrations were also classified into daytime (about 06:30 - 20:00) and night-time (about 20:00 - 06:30).

Finally, in order to compare the indoor and outdoor particle mass concentrations, the outdoor PM$_{10}$ were adjusted to PM$_{2.5}$ using the ratio of PM$_{2.5}$/PM$_{10}$ = 0.61. This value was provided by the Environmental Protection Agency (Neale D. and Wainwright D. 2000) based on the data from the monitoring stations in Brisbane for 1999, the year in which this project was conducted.

5.3. RESULTS

5.3.1 Diurnal variation

Real-time particle concentration data combined with the human activity information allow the sources of particle diurnal variations to be analysed. The results show that there were clear diurnal variations (one-way ANOVA, p<0.005) of particle number concentration and the approximation of PM$_{2.5}$ concentration for all the investigated houses with the pattern of the variations differing from house to house. In general, however, the particle concentrations in the morning (06:00 - 09:00) and evening (17:00 - 20:00) were higher than the concentrations during midnight to early morning, and daytime (09:00 - 17:00). This trend in particle concentrations correlates well with the pattern of human indoor activity where most cooking occurs in the evening or in the morning.

Typical diurnal variations of particle number and approximation of PM$_{2.5}$ concentrations are presented in Figures 5.1 and 5.2. The figures show how human
activities, especially cooking, can significantly affect the indoor particle concentration. There is a sharp increase in concentration at the start of each indoor activity and a slow decrease after the activity ceased. It can be seen from Figure 5.1 that both particle number and mass concentrations were very low in the absence of human activities (for example: CPC about \( 1.5 \sim 2.5 \times 10^3 \) particles cm\(^{-3}\) and approximation of PM\(_{2.5}\) about \( 6 \sim 8 \) µg m\(^{-3}\) for the period from 16:00 hour, 9 July to 19:00 hour, 10 July). However, particle concentrations were significantly higher during cooking activities (up to 100 times higher than the background level) and often remained high for a long period of time of up to fourteen hours, as presented in Figure 5.1 for the period from 19:00 hour, 11 July to 9:00 hour, 12 July. A more detailed discussion of the concentration levels and their variation for different times and activities is provided below.

![Graph showing CPC and PM\(_{2.5}\) concentrations over time with activities marked]

**Fig. 5.1.** House3, 9-12/July 1999, CPC and approximation of PM\(_{2.5}\) concentration in Kitchen, 48h.
Fig. 5.2. House 12, 21-23/May/1999, CPC and approximation of PM$_{2.5}$ concentrations in kitchen, 48h (CS: cigarette smoking).

5.3.2 Characteristics of variation in particle number concentrations

Table 5.1 presents a summary of concentrations for each situation under the classification developed for this study. The results presented in Table 5.1 show that the total 24h average particle number concentration is clearly higher (t-test, $p < 0.001$) than non-activity 24h average concentration for all houses with the ratio of the former to the latter ranging from 1.23 to 2.31, and with an average value of 1.49 ($\pm 0.3$). Furthermore, the average ratio of the 24h indoor to the 24h outdoor concentration (1.35$\pm 0.88$) is higher (t-test, $p = 0.001$) than that of the non-activity 24h indoor concentration to the outdoor 24h concentration (1.05$\pm 0.72$). (See Table 5.1)
Table 5.1. Summary of the diurnal variation of indoor and ambient particle number concentrations (particle number concentration: particles m\(^{-3}\) \(\times 1000\)) and the fine particle mass concentrations (PM\(_{2.5}\): µg m\(^{-3}\)), as well as the indoor and outdoor concentration ratios.

<table>
<thead>
<tr>
<th>Non-Activity</th>
<th>Activity</th>
<th>Min</th>
<th>Max</th>
<th>Ratio</th>
<th>Ratio</th>
<th>Day</th>
<th>Night</th>
<th>Total</th>
<th>Ratio</th>
<th>Ratio</th>
<th>Ratio</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Day</td>
<td>Night</td>
<td>Total</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Min</td>
<td>12.8</td>
<td>12.0</td>
<td>12.4</td>
<td>44.4</td>
<td>22.3</td>
<td>41.1</td>
<td>18.2</td>
<td>4.2</td>
<td>268.6</td>
<td>73.4</td>
<td>1.49</td>
<td>12.9</td>
</tr>
<tr>
<td>S.D</td>
<td>3.3</td>
<td>3.4</td>
<td>2.7</td>
<td>18.6</td>
<td>8.3</td>
<td>18.4</td>
<td>3.9</td>
<td>1.9</td>
<td>237.0</td>
<td>61.7</td>
<td>0.30</td>
<td>7.2</td>
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<td>19.1</td>
<td>85.9</td>
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<td>77.4</td>
<td>27.2</td>
<td>7.8</td>
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<tr>
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<td>8.5</td>
<td>9.0</td>
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<td>19.8</td>
<td>13.6</td>
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Min: Minimum; Max: Maximum; 24h: 24 hours average; NA24h: Non-Activity 24 hours average; A24h: total 24 hours average; 24h: 24 hours average; DI/DO: Day Indoor/Day Outdoor; NI/NO: Night Indoor/Night Outdoor; NAI24h: Non-Activity Indoor 24 hours average; I24h: total Indoor 24 hours average; O24h: Outdoor total 24 hours average.

*Due to instrument malfeasance data on particle number concentration was not available for one of the houses, therefore average values for number concentration were calculated for fourteen, not fifteen houses.
It can be seen from Table 5.1 that the daytime outdoor concentrations had an average of \((12.9 \pm 7.2) \times 10^3\) particles cm\(^{-3}\) and were always higher (t-test, \(p = 0.005\)) than the night-time concentrations, which averaged \((8.1 \pm 5.0) \times 10^3\) particles cm\(^{-3}\). There were no significant differences between the indoor and outdoor daytime concentrations under non-activity situations. The ratios of daytime indoor to outdoor concentrations under non-activity conditions averaged 0.93\(\pm\)0.67 and were statistically lower (t-test, \(p = 0.024\)) than the ratios of night-time concentrations, which averaged 1.33\(\pm\)0.74. This implies that the daytime indoor concentrations under these conditions were mainly influenced by outdoor concentrations, while the night-time indoor concentrations were affected by the activities that took place in the houses in the evening. During the winter, the residents of these houses normally keep windows open during the day and closed at night. This behaviour results in reduction of the air exchange rate at night, thus reducing the amount of outside air entering the house at night-time and reducing removal of particles generated indoors during evening activities.

The minimum particle number concentration ranged from \(1.3 \times 10^3\) to \(7.8 \times 10^3\) particles cm\(^{-3}\) with an average of \((4.2 \pm 1.9) \times 10^3\) particles cm\(^{-3}\). The times when the concentrations were at a minimum were between about 23:00 h and 08:00 h and between 14:00 h and 18:00 h in the absence of human activities. The maximum number concentration ranged from \(1.12 \times 10^5\) to \(8.17 \times 10^5\) particles cm\(^{-3}\) with an average of \((2.69 \pm 2.37) \times 10^5\) particles cm\(^{-3}\). The maximum number concentrations were observed between about 06:30 h and 08:50 h and between about 16:30 h and 20:00 h during cooking times. The maximum to minimum ratio of particle concentrations ranged from 15 to 238, with an average of 73\(\pm\)62. These large
standard deviations in the minimum, maximum and maximum/minimum ratio results from the high variability of the particle number concentration indoors. These results not only indicate that the diurnal variation of particle number concentration was house-specific and highly dependent on the resident's activities and lifestyle, but also quantify the ranges of concentrations and variation in the concentrations between the houses.

5.3.3 Characteristics of variation in the approximation of PM$_{2.5}$

Table 5.1 summarises also measured indoor and recalculated outdoor approximations in PM$_{2.5}$ concentrations. Under the non-activity conditions, the daytime and night-time approximation of PM$_{2.5}$ concentrations varied somewhat from house to house, with a daytime average for all the houses of 10.6±2.6 µg m$^{-3}$ and night-time average of 11.7±3.0 µg m$^{-3}$. Unlike particle number concentrations, there were no significant differences between the daytime and night-time approximation of PM$_{2.5}$ concentrations for most houses. The ratios of daytime approximation of PM$_{2.5}$ indoor concentrations for non-activity conditions to outdoor concentrations, varied only slightly from house to house, with an average of 1.06±0.2. This result is consistent with the findings of Morawska et al., (2001) that the ratios of indoor to outdoor approximation of PM$_{2.5}$ concentration for conditions of no indoor sources were close to one for most houses during the daytime (1.01 and 1.08 for normal and minimum ventilation conditions, respectively). The ratios of night-time indoor to outdoor concentrations varied from house to house and were greater than one for all houses, except House 1, with an average of 1.71±0.64. The outdoor night-time PM$_{2.5}$ concentrations averaged at 7.3±2.8 µg m$^{-3}$ and were significantly lower (t-test, $p<$
than the daytime outdoor concentration, most likely due to the decrease in
human activities outdoors during the night. Nine of the fifteen houses investigated in
this study displayed higher night-time than daytime indoor approximation of PM$_{2.5}$
concentrations. Further analysis revealed that the residents of the nine houses (except
for House 14) normally prepare their main meal in the evening. Although the
cooking ended in the evening, the indoor approximation of PM$_{2.5}$ concentrations
remained higher for prolonged periods of time during the night due to the low air
exchange rate at night during winter. In summary, the ratios of night-time indoor to
outdoor PM$_{2.5}$ concentrations are greater than one and are significantly greater (t-test,
$p = 0.001$) than the ratios of the daytime concentrations.

Both 24h non-activity average and the total indoor approximation of PM$_{2.5}$
concentrations varied from house to house, but the latter showed a more significant
variation with an average of 11.1±2.6 µg m$^{-3}$ for non-activity conditions and
15.5±7.9 µg m$^{-3}$ for total concentrations. In the absence of an Australian PM$_{2.5}$
standard, these average concentrations can be compared to the US EPA PM$_{2.5}$ 24h
and annual standards of 65 and 15 µg m$^{-3}$, respectively. While the 24h standard was
not exceeded in any house in this study, the concentration levels are close to the
value of the annual standard.

The ratios of 24h total to non-activity indoor approximation of PM$_{2.5}$ concentrations
were greater than one for all houses with an average of 1.36±0.49. For most of the
houses, however, the ratios were not much higher than one, with only a few houses
where the ratios were significantly elevated (t-test, $p < 0.001$).

The minimum approximation of PM$_{2.5}$ concentrations indoors normally occurred
over the period from 01:00 h to 07:00 h and were in the range from 5.2 µg m$^{-3}$ to 8.8
μg m⁻³ with an average of (7.3±1.2) μg m⁻³. The maximum concentrations ranged from 16 μg m⁻³ to 2.8×10³ μg m⁻³ with an average of (5.35±9.09)×10² μg m⁻³. The maximum concentrations normally coincided with cooking activities and were observed between 08:00 h and 14:00 h and between 17:30 h and 20:00 h. The ratio of maximum to minimum approximation of PM₂.₅ concentrations ranged from 1.8 to over 470. As for the number concentrations discussed above, these large standard deviations in the minimum, maximum and in the ratios of maximum to minimum concentrations result from the high variability of the approximation of PM₂.₅ concentration indoors. These results not only indicate that the diurnal variation of approximation of PM₂.₅ concentration was house-specific and highly dependent on the resident's activities and lifestyle, but also quantify the ranges of concentrations and variation in the concentrations between the houses.

The ratios of 24h total indoor to outdoor concentrations were greater than one for all houses except House 1, with the average for all the houses of 1.72±1.00. The ratios of 24h non-activity indoor to outdoor concentrations were lower than these with an average of 1.27±0.28. Comparison of the ratios not only demonstrates that the indoor activities increased the concentration of the indoor approximation of PM₂.₅ which became higher than the outdoor concentration, but also quantify the extent of this increase.

5.3.4 Comparison of number to mass concentration

The real-time paired data points of indoor particle number concentrations and approximation of PM₂.₅ concentrations were analysed to identify in the first instance the existence of linear correlations. The results showed that correlation coefficients (r) were highly variable from day to day, and from house to house. For example, the
correlation coefficient for House 14, varied from 0.39 (first day) to 0.69 (second day) and then 0.59 (third day). The correlation coefficient for House 4 changed from 0.24 (first day) to 0.67 (second day). The average correlation coefficient for each house ranged from 0.1 (House 6) to 0.73 (House 3), with an overall average value of 0.48±0.27. There were significant correlations (p < 0.01) between the indoor particle number concentrations and approximation of PM$_{2.5}$ concentrations on some days, but not on other days. Therefore in summary, there were no clear linear correlations between approximation of PM$_{2.5}$ and particle number concentration in the long-term. This could be due to a number of factors, the most important of them being that different sources could be the main contributors to particle number and approximation of PM$_{2.5}$ concentrations.

The results of further correlation analysis showed there were some other types of relationship (power, logarithmic and exponential) in some cases. However, in general, there was no long-term, consistent relationship between approximation of PM$_{2.5}$ concentration and number concentration. This implies that no conclusions can be made about one of these characteristics of indoor particles (number or mass concentration), based on a measurement of the other.

5.4. DISCUSSION

Figure 5.2 shows that both particle number concentrations and approximation of PM$_{2.5}$ concentrations clearly increased during the time when the residents were smoking in the room. The peak concentrations were up to two times, and five times higher than the background values, for number and mass concentrations, respectively. Many previous studies showed that cigarette smoking significantly
affects indoor particle concentration. For example, Brauer et al., (2000) found that the peak concentrations of PM$_{2.5}$ were up to 4.5 times higher than the background values in a bar during cigarette smoking. They also report the range of PM$_{3.5}$ concentration during cigarette smoking in a residential living room was 20 ~ 100 µg m$^{-3}$. Their results are comparable with the findings in this study (32 ~ 50 µg m$^{-3}$ for PM$_{2.5}$ concentration during cigarette smoking).

Investigations of diurnal variation in outdoor particle mass concentrations showed that the high outdoor concentrations were normally observed during the traffic peak hours. This result is consistent with previous findings that the patterns of outdoor diurnal variations were significantly affected by the diurnal variations in vehicle emissions and meteorological conditions in Brisbane city (Morawska et al., 1999; Jamriska et al., 1999).

When comparing indoor and outdoor diurnal variations, it was found that the patterns were quite different when there were significant indoor activities occurring. This finding is expected and consistent with, for example, the results reported by Patterson and Eatough (2000) who measured the diurnal variation of indoor and outdoor fine particle number and mass concentration (PM$_{2.5}$) in a school using a CNC and a TEOM. Their results showed clear diurnal variations in indoor particle concentration. However, the authors found a low correlation between indoor PM$_{2.5}$ concentrations and outdoor PM$_{2.5}$ concentrations because of the influence of student activities on the indoor concentrations.

A previous results from the same large study (Morawska et al., 2001), showed that the ratios of indoor to outdoor particle number concentrations were on average below one during the day when no indoor sources were in operation (0.89 and 0.78 under
normal and minimum ventilation, respectively). Those findings are consistent with the results presented here where the average ratio of daytime non-activity concentrations indoor and outdoor concentrations was 0.93±0.67. Unlike studies of particle mass concentration indoors, there are only a few studies reporting on indoor particle number concentrations. Li et al., (1993) investigated real-time indoor submicrometer particle concentrations in the size range: 0.017-0.886 µm for 24 hours. Their results showed that the concentration varied from 1.4×10^4 to 1.5×10^5 particles cm^-3. This is comparable with the results of this study and not only supports the finding that peak values indoors occur during significant indoor activities such as cooking, but also provides an estimate for the height of these peaks.

Several different measurement methods were used for monitoring of indoor PM_{2.5} concentrations in the studies reported in the literature, therefore comparisons of the results from these studies should be made with caution. Wiener, et al. (1990) performed a long-term study of PM_{2.5} concentrations in nine homes in the USA and the results showed that the average 24 hours PM_{2.5} concentration was (36.3±2.6) µg m^{-3}. Recently, Lachenmyer and Hidy (2000) examined the average mass concentration in 10 houses in Birmingham, AL, USA using filtration and gravimetric mass techniques. Their data showed that the average indoor 48 hours PM_{2.5} concentration was (16.1±5.7) µg.m^{-3} in summer, and (11.2 ± 5.4) µg.m^{-3} in winter. In a study conducted over two weeks in 28 homes in an urban area of Huddersfield, U.K., Kingham, et al., (2000) measured PM_{2.5} concentrations using active pump samplers running for 24h in each home. The mean values were (17.81±12.21) µg.m^{-3} for houses located within 50 m of the main road and (19.52±13.58) µg.m^{-3} for houses at a distance greater than 50 m of from the main road, respectively. In Perth, Australia, Stratifico and Dingle (1996) using a respirable dust cyclone found that the
average 24 hours PM$_{2.5}$ concentration of 48 houses was 34.4 ± 20.3 µg.m$^{-3}$. Monn et al., (1997) measured indoor PM$_{2.5}$ concentrations in Switzerland using Teflon filters. The sampling period was from 48 to 72 hours. The average daily PM$_{2.5}$ concentration was found to be 18.3 µg.m$^{-3}$ for non-activity situations and 26.0 µg.m$^{-3}$ for activity situations.

The results of the current study are comparable with the findings of Lachenmyer and Hidy (2000) and Kingham, et. al., (2000), are somewhat lower than those of Monn et al (1997) and are significantly lower than those of Wiener, et. al., (1990) and Stratico and Dingle (1996). The large difference between the results of these studies may be contributed to by the different measurement methods which were used for monitoring of indoor PM$_{2.5}$ concentrations, as well as differences in the indoor/outdoor environments investigated.

Comparing the indoor particle concentrations it was apparent that a low concentration of approximation of PM$_{2.5}$ does not necessarily imply a low particle number concentration and vice versa. These findings suggest that the daily correlations between particle number concentration and PM$_{2.5}$ concentration are complex and variable, and are highly affected by the number, type and strength of indoor activities and outdoor sources. It was clear that the average number concentrations including indoor activity conditions were higher than those for non-activity conditions for all houses, but not all the houses showed the same trend in relation to mass concentration (approximation of PM$_{2.5}$). This result suggests that number concentration is a better particle characteristic than PM$_{2.5}$ to determine the contribution from indoor sources to the total concentration (the most important of which are combustion sources).
5.5 CONCLUSIONS

In this study significant short-term variations in particle number concentrations and approximation of PM$_{2.5}$ concentrations were found in fifteen houses in Brisbane, Australia. The results show that there were clear diurnal variations in both particle number and approximation of PM$_{2.5}$ concentrations, for all the investigated houses. The pattern of diurnal variations varied from house to house, however, there was always a close relation between the variations and human indoor activities. The average number and mass concentrations during indoor activities were $(18.2\pm3.9) \times 10^3$ particles cm$^{-3}$ and $(15.5\pm7.9)$ µg m$^{-3}$, respectively, and under non-activity conditions, $(12.4\pm2.7) \times 10^3$ particles cm$^{-3}$ $(11.1\pm2.6)$ µg m$^{-3}$, respectively. The results of this study support the findings of previous studies that the major sources of particles indoors are cooking and environmental tobacco smoke (for smoking indoors). In general, there was a poor correlation between mass and number concentration and the correlation coefficients were highly variable from day to day, and from house to house. This implies that conclusions can not be drawn about either one of the number or mass concentration characteristics of indoor particles based on measurement of the other.

Another conclusion from this study is that due to the significant differences between the indoor and outdoor diurnal variations it would be very difficult to accurately estimate indoor particle mass or number concentrations by using the outdoor concentrations solely.
The limitations of the study were the relatively short time of measurement in each house, the relatively small number of houses investigated and using only an approximation of PM$_{2.5}$. The strengths were in the provision of real-time data with high resolution, enabling estimation of minimum and maximum concentration as well as the time series concentrations.

ACKNOWLEDGMENTS

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CHAPTER 6
CONTRIBUTION FROM INDOOR SOURCES TO PARTICLE NUMBER AND MASS CONCENTRATIONS IN RESIDENTIAL HOUSES

Congrong He\(^1\), Lidia Morawska\(^1\), Jane Hitchins\(^1\), and Dale Gilbert\(^2\)

\(^1\)International Laboratory of Air Quality and Health, Queensland University of Technology, Brisbane, QLD 4001, Australia

\(^2\)Built Environment Research Unit, Queensland Department of Public Works and Housing, Brisbane, QLD 4001, Australia

STATEMENT OF JOINT AUTHORSHIP

Title: Contribution from indoor sources to particle number and mass concentrations in residential houses

Authors: Congrong He, Lidia Morawska, Jane Hitchins, Kerrie Mengersen and Dale Gilbert and

Congrong He (candidate)
Developed experimental design and scientific method; analysed and interpreted data and wrote manuscript

Lidia Morawska
Contributed to experimental design and scientific method, interpreted data; assisted with manuscript

Jane Hitchins
Contributed to experimental design and scientific method; assisted with manuscript

Dale Gilbert
Assisted with data interpretation and manuscript
Abstract

As part of a large study investigating indoor air in residential houses in Brisbane, Australia, the purpose of this work was to quantify emission characteristics of indoor particle sources in 15 houses. Submicrometer particle number and approximation of PM$_{2.5}$ concentrations were measured simultaneously for more than 48 hours in the kitchen of all the houses using a condensation particle counter (CPC) and a photometer (DustTrak), respectively. In addition, characterizations of particles resulting from cooking conducted in an identical way in all the houses were measured using a scanning mobility particle sizer (SMPS), an aerodynamic particle sizer (APS) and a DustTrak. All the events of elevated particle concentrations were linked to indoor activities using house occupants diary entries, and catalogued into 21 different types of indoor activities. This enabled quantification of the effect of indoor sources on indoor particle concentrations as well as quantification of emission rates from the sources. For example, the study found that frying, grilling, stove use, toasting, cooking pizza, cooking, candle vaporizing eucalyptus oil and fan heater use, could elevate the indoor submicrometer particle number concentration levels by more than 5 times, while PM$_{2.5}$ concentrations could be up to 3, 30 and 90 times higher than the background levels during smoking, frying and grilling, respectively.

Keywords: Indoor air quality; Indoor emission sources; Ultrafine particles; Emission rates; Submicrometer particles
6.1 Introduction

Both indoor and outdoor sources contribute to and affect the concentration and composition of particles in indoor air. There is usually more information available on emission characteristics, such as emission factors or emission rates, of outdoor particle sources than of indoor sources. However, quantification of emissions from indoor sources is very important for assessment of total human exposure to particles. As a result of the operation of certain indoor sources, particle concentration levels indoors could be temporarily or even for prolonged periods of time elevated up to tenfold compared to the situation without the sources. This can have a significant effect on human health.

A large number of indoor particle sources have been identified and emissions from these sources investigated by many studies reported in the literature. The most significant sources include tobacco smoking, cooking, kerosene heating and wood burning (e.g. Tuckett et al., 1998; Long et al., 2000). Other sources or human activities contributing to elevated levels of indoor particles include re-suspension of particles by people and pets, dusting and vacuuming, showering, operation of humidifiers, electric motors, etc (e.g. Monn et al., 1995; Tucker, 2000).

Combustion processes are the main indoor sources of smaller particles, with the vast majority of them in the submicrometer range, containing a host of organic and inorganic material (Morawska and Zhang, 2002). Re-suspension of particles by human movement on the other hand, contributes to the coarse mode of indoor particles, usually in the size above one micrometer.

Secondary particles in the indoor environment are formed from gaseous pollutants through the process of gas-to-particle conversion. For example, Rohr et al. (2003)
reported the effect of reactions between ozone and selected terpenes on the concentration and size distributions of airborne particles in a chamber setting. Their results demonstrated that ozone/terpene reactions could be a significant source of submicrometer secondary particles in indoor settings.

Quantitative assessment of indoor source emission characteristics in real situations is a complex task, and therefore only qualitative information about the contribution of many indoor particle sources, or about indoor ranges of concentration levels as a result of operation of the sources is available. Emissions from tobacco smoking have attracted considerable attention and as a result there is more information available on emission rates from this, compared to other indoor sources (e.g. Brauer et al., 2000).

The existing database is limited to particle mass emission rates with only a few studies reported on particle number emission rates. However, since smaller particles, which can be high in number but contribute very little to particle mass, have a higher probability of penetration into the deeper parts of the respiratory tract (e.g. James et al., 1991), and also contain higher levels of trace elements and toxins such as the polycyclic aromatic hydrocarbons and mutagens (e.g. Kiss et al., 1998), they have been a subject of increasing concern. Recent health effects studies have also suggested that number concentration may be a more appropriate predictor of health effects than mass concentration (e.g. Penttinen et al., 2001; Stephenson et al., 2003).

Thus, it is clear that knowledge of particle number emission rates or factors is of importance for exposure assessment and for developing of appropriate control strategies.

As part of a large study investigating various aspects of indoor air in residential houses in Brisbane, Australia, the purpose of this work was to investigate the
emission characteristics of the most common indoor particle sources. The specific objectives of the study included: (1) identification of the main indoor sources contributing to the elevated number and mass (PM$_{2.5}$) concentration levels; (2) quantitative evaluation of the effect of these sources on indoor particle concentration levels; (3) estimation of the emission strength of the sources; (4) characterization and comparison of particle size distributions resulting from cooking conducted in an identical way in different types of residential houses under normal and minimum ventilation conditions. It was expected that cooking activities would be one of the major source of indoor particles and the hypothesis was that stove properties affect characteristics of emissions resulting from cooking.

6.2 Experimental method

6.2.1 Sampling site and house

Detailed information about the sampling site and the houses included in this study has been provided earlier (Morawska et al., 2001). Briefly, a residential suburb in Brisbane of a reasonably flat topography and with a good mix of houses, both in terms of age and of style i.e. newer and older houses, brick and timber, high set and low set, was chosen as the measurement site. Fourteen houses in the suburb were chosen for the study and one additional house was chosen from another suburb as a comparison site.

6.2.2 Instrumentation

Particle size and number concentration measurements were conducted using three different instruments: (1) the TSI Model 3320 Aerodynamic Particle Sizer (APS) (TSI Incorporated, St. Paul, MN, USA), which measures particle size distribution
and number concentration in real time, in the range from 0.5 to 20 µm and up to the order of $10^3$ or $10^4$ particle cm$^{-3}$ for coincidence errors of 1% and 5% respectively; (2) the TSI Model 3934 Scanning Mobility Particle Sizer (SMPS) (TSI Incorporated, St. Paul, MN, USA), which was set to measure particle size distribution and number concentration, in the range from 0.015 to 0.685 µm, and (3) the TSI Model 3022A Condensation Particle Counter (CPC) (TSI Incorporated, St. Paul, MN, USA), which measured the total number concentration of submicrometer particles in the size range from 0.007 to 0.808 µm.

For the purpose of this paper particles measured by the SMPS and CPC are called submicrometer particles, while the particles measured by the APS, are called supermicrometer particles.

Approximation of fine particle mass concentration (PM$_{2.5}$) was measured by the TSI Model 8520 DustTrak aerosol monitor (TSI Incorporated, St. Paul, MN, USA). It should be noted that the DustTrak operates based on a light scattering technique where the amount of scattered light is proportional to the volume concentration of the aerosol. The approximation of PM$_{2.5}$ values obtained in this study using this instrument are not actual gravimetric values, as the instrument was not calibrated for each specific aerosol studied. For simplification, all the DustTrak results discussed in this paper are referred to as PM$_{2.5}$, omitting the term ‘approximation’.

The TSI Model 8551 Qtrak (Q-Trak - TSI Incorporated, St. Paul, MN, USA) was employed to measure CO$_2$ concentrations, which was used to estimate air exchange rates (AER).

These instruments were chosen as the most suitable for indoor studies because of their low flow rates (and thus negligible impact on particle concentrations indoors),
quiet operation and their short sampling times (10 and 30 seconds for CPC and DustTrak, respectively).

6.2.3 Sampling protocol

The experiments consisted of three parts: air exchange rate (AER) measurement, controlled cooking test, and time series of indoor particle concentration measurement. All measurements (except House1) were conducted between May and July 1999, which is wintertime in Brisbane.

Knowledge of AER is important for determination of source emission rates measured in real residential houses. Before commencement of measurements in the houses, a comparison test between CO₂ and SF₆ methods for measuring AER was conducted in a room at the International Laboratory for Air Quality and Health, Queensland University of Technology. The results were comparable for the two methods. Under normal ventilation conditions the SF₆ method yielded 3.15 air changes per hour (ACH) and the CO₂ method 3.34 ACH; under minimum ventilation condition: SF₆ – 0.96 ACH, CO₂ – 0.76 ACH.). Since the method using CO₂ decay is cheaper (did not require renting of an instrument) and is widely used in AER measurement, it was chosen for the measurements in the houses. During the measurements of the AER the occupants were not present in the houses and CO₂ was released until its concentration was at least three times higher than the background level. The Q-Trak monitor was used to record the decay of the CO₂ concentration.

For the cooking test, the APS, SMPS and DustTrak were employed and measurements were conducted under both normal and minimum ventilation conditions for each house. Normal ventilation condition means all the door and windows of the house, which are normally opened, stayed opened during the
measurements. Minimum ventilation condition means that all the doors and windows of the house were closed.

The procedure for the cooking test was as follows: 10 minutes of background concentration measurements in the kitchen, followed by measurements during 10 minutes of cooking (half an onion was sliced and placed in a small frying pan containing 1 spoonful of vegetable oil, and the stove was switched onto high), and then by a further 20 minutes of measurements after the cooking ceased. Cooking tests were conducted first under minimum ventilation conditions, and then under normal ventilation conditions (in all houses with the exception of House7). These controlled cooking tests provided information about characteristics of particle size distribution and number median diameter resulting from one particular source (frying), which could be compared between the houses. The data obtained from the test was used for calculation of particle emission rates in conjunction with AER and other house data.

For indoor time series concentration measurements, a CPC and DustTrak were employed. PM$_{2.5}$ and particle number concentrations were measured simultaneously for more than 48 hours in the kitchen of all the houses. The CPC and DustTrak were placed side-by-side and positioned on average two metres from the stove in the kitchen. The occupants of the houses were asked to fill in a diary, noting the time and duration of any activity occurring throughout the house during the time of the measurements. From the time series concentration data and the information about indoor activities, contribution from the individual indoor sources or activities was qualitatively identified. Then in conjunction with AER and other house data this information was used to calculate sources emission rates according to the method described below.
6.2.4 Calculation of air exchange rate

The tracer gas technique involves injecting a tracer gas and mixing it through the house, then measuring its decay rate with an appropriate instrument. If exfiltration rates of the tracer gas are constant, mixing is uniform, the chemical reaction between the gas and other chemicals is negligible and no indoor source of the gas is operating, the air exchange rate, $\alpha$, can be calculated from the following equation (Nantka, 1990):

$$\alpha = \frac{1}{t} \ln \frac{C_t}{C_0}$$

where $t$ is time, $C_t$ and $C_0$ are concentrations of the gas at times $t$ and 0, respectively. Equation 1 was used for calculation of AER of the houses in this study based on measured CO$_2$ decay rates.

6.2.5 Estimation of source emission rates

Residential houses in Brisbane do not normally use air filtration systems. This means that the principal factors governing the levels of airborne particles indoors are the contributions from indoor and outdoor sources, the deposition rate of particles on indoor surfaces, and the air exchange rate (e.g. Thatcher and Layton, 1995). A formula for calculation of indoor particle concentration levels taking into consideration these factors can be written as follows (Koutrakis et al., 1992; Chen et al., 2000):

$$\frac{dC_{in}}{dt} = P\alpha C_{out} + \frac{Q}{V} - (\alpha + k)C_{in}$$

(2)
where $C_{in}$ and $C_{out}$ are the indoor and outdoor particle concentrations, respectively; $P$ is the penetration efficiency; $\alpha$ is the air exchange rate; $k$ is the deposition rate; $Q_s$ is the indoor particle generation rate; $t$ is time; and $V$ is the efficient volume of the house. A number of previous studies discussed the use of this equation for determination of source emissions and prediction of indoor air pollutant concentration levels (e.g. Dockery et al., 1981; Ott, 1999). All the factors in this equation, with the exception of the efficient volume of the house ($V$), are functions of some other factors and can vary in time (for example penetration efficiency is a function of particle size). In order to estimate the average emission rate ($\overline{Q}_s$), Equation 2 is simplified by using average values instead of functions and also by making some further assumptions about the experimental conditions. The penetration efficiency ($P$) is commonly assumed to be close to one for both fine and coarse particles (Wallace, 1996). However, it should be noted that a number of studies indicated that the penetration efficiency and its variability is higher for fine particles than for coarse ones (e.g. Abt et al., 2000a; Long et al., 2001). In addition, for conditions when no indoor source is in operation, the indoor particle concentration can be approximated by outdoor particle concentration (Morawska et al., 2001), and the initial indoor particle concentration ($C_{in0}$) could be used to replace outdoor particle concentrations. Thus, Equation 2 can be written as:

$$\overline{Q}_s = V \left[ \frac{C_{int} - C_{in0}}{\Delta T} + (\alpha + k) \times C_{in} - \alpha \times C_{in0} \right]$$ (3)
where \( \overline{Q} \) is average emission rate, \( C_{int} \) and \( C_{in0} \) are the peak and initial indoor particle concentrations, respectively; \( \overline{\alpha} \) is the average air exchange rate; \( \overline{\alpha + k} \) is the average total removal rate; \( \Delta T \) is time difference between initial and peak concentration. This equation ignores the effects of processes involving particles, such as condensation, evaporation or coagulation, since these are minor effects under particle concentrations and conditions normally encountered in residential environments (e.g. Thatcher and Layton, 1995). Equation (3) has most commonly been used for particle mass concentration and emission calculations (mass balance equation), but it can also be used for calculations of particle number concentration and emissions.

Using the average decay rate of indoor particle concentration after the indoor activity ceased (the source stopped operating), the average total removal rate \( \overline{\alpha + k} \) can be obtained. The average air exchange rate for each house was determined experimentally for two different ventilation conditions through CO\(_2\) decay test. Thus, Equation 3 can be used to determine indoor source particle emission rates, using the above data on the total removal rate, air exchange rate and background concentration (initial indoor concentration).

6.2.6 Data processing and analysis

All statistical analyses (correlation, regression, t-test, One-Way ANOVA) were conducted using a statistical analysis software package – SPSS for Windows version 10 (SPSS Inc.). A level of significance of \( p = 0.05 \) was used for all statistical procedures. When the distribution of the data was not a normal distribution, the robust analysis (trim off the maximum and minimum) was employed.
6.3 Results and discussion

6.3.1 Source identification and emission rate

Analyses of particles time series concentration data collected over 48 hours in each house and comparison of this data with the entries in the residents’ diaries enabled identification of the activities, which contributed to the elevated particle concentration levels. A total of 153 events were identified for which elevated concentrations were matched with a diary entry for specific activities. All the events linked to indoor activities were catalogued into 21 different types of indoor activities. One of the most common activities, cooking, was defined as any food preparation using stove not involving frying or grilling, without restricting, however, the number of dishes cooked simultaneously (e.g. boiling pasta and cooking sauce).

In addition to the events linked to indoor sources, there were a number of events when elevated indoor concentrations resulted from particles generated by outdoor sources, such as neighbour burning off rubbish, smoking outside or opening of the door. These events have also been included in the analyses to enable comparison between the effects of indoor and outdoor particle sources. Table 6.1 presents a list of the types of activities the identified together with the frequency of their occurrence, median peak concentration of PM$_{2.5}$ and submicrometer particle number resulting from the type of activity. Included in Table 6.1 are also the ratios of peak to background values and estimated particle PM$_{2.5}$ and submicrometer number emission rates for the specific activities.
Table 6.1. Summary the results from the 48h measurements for PM$_{2.5}$ and submicrometer particle numbers (measured by the Condensation Particle Counter): peak concentration values, the ratios of the peak to background concentration values and the emission rates

<table>
<thead>
<tr>
<th>Activity</th>
<th>N</th>
<th>Peak values (µg.m$^{-3}$)</th>
<th>Ratio (mass)</th>
<th>Emission rate (mg min$^{-1}$)</th>
<th>Peak values (particle cm$^{-3}$, ×10$^{3}$)</th>
<th>Ratio (number)</th>
<th>Emission rate (particle min$^{-1}$ × 10$^{11}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Median S.D.</td>
<td>Median S.D.</td>
<td>Median S.D.</td>
<td>Median S.D.</td>
<td>Median S.D.</td>
<td>Median S.D.</td>
</tr>
<tr>
<td>Food preparation</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cooking</td>
<td>24</td>
<td>37</td>
<td>194</td>
<td>2.89</td>
<td>12.6</td>
<td>0.11</td>
<td>0.99</td>
</tr>
<tr>
<td>Cooking Pizza$^a$</td>
<td>1</td>
<td>735</td>
<td>73.5</td>
<td>1.59</td>
<td>137.3</td>
<td>9.81</td>
<td>1.65</td>
</tr>
<tr>
<td>Frying</td>
<td>4</td>
<td>745</td>
<td>352</td>
<td>33.6</td>
<td>28.3</td>
<td>2.68</td>
<td>2.18</td>
</tr>
<tr>
<td>Grilling</td>
<td>6</td>
<td>718</td>
<td>3427</td>
<td>90.1</td>
<td>312</td>
<td>2.78</td>
<td>17.8</td>
</tr>
<tr>
<td>Kettle</td>
<td>25</td>
<td>13</td>
<td>20</td>
<td>1.13</td>
<td>0.67</td>
<td>0.03</td>
<td>0.31</td>
</tr>
<tr>
<td>Microwave</td>
<td>18</td>
<td>16</td>
<td>18</td>
<td>1.12</td>
<td>0.42</td>
<td>0.03</td>
<td>0.11</td>
</tr>
<tr>
<td>Oven</td>
<td>6</td>
<td>24</td>
<td>6</td>
<td>1.76</td>
<td>0.53</td>
<td>0.03</td>
<td>0.03</td>
</tr>
<tr>
<td>Stove</td>
<td>4</td>
<td>57</td>
<td>264</td>
<td>2.4</td>
<td>19.7</td>
<td>0.24</td>
<td>1.29</td>
</tr>
<tr>
<td>Toasting</td>
<td>18</td>
<td>35</td>
<td>32</td>
<td>2.08</td>
<td>8.31</td>
<td>0.11</td>
<td>0.37</td>
</tr>
<tr>
<td>Other activities</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Other activities</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Open door</td>
<td>9</td>
<td>21</td>
<td>9</td>
<td>1.23</td>
<td>0.37</td>
<td>22.0</td>
<td>14.6</td>
</tr>
<tr>
<td>Smoking</td>
<td>6</td>
<td>79</td>
<td>29</td>
<td>4.03</td>
<td>1.76</td>
<td>0.99</td>
<td>0.81</td>
</tr>
<tr>
<td>Sweep floor</td>
<td>3</td>
<td>35</td>
<td>4</td>
<td>2.04</td>
<td>1.3</td>
<td>0.05</td>
<td>0.01</td>
</tr>
<tr>
<td>Vacuuming</td>
<td>5</td>
<td>16</td>
<td>8</td>
<td>1.46</td>
<td>0.32</td>
<td>0.07</td>
<td>0.04</td>
</tr>
<tr>
<td>Washing</td>
<td>17</td>
<td>18</td>
<td>12</td>
<td>1.25</td>
<td>0.57</td>
<td>0.04</td>
<td>0.04</td>
</tr>
<tr>
<td>Other activities</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Other activities</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CVE oil$^a$</td>
<td>1</td>
<td>132</td>
<td>13.2</td>
<td>0.91</td>
<td>74.6</td>
<td>8.29</td>
<td>5.52</td>
</tr>
<tr>
<td>Dusting$^a$</td>
<td>1</td>
<td>22</td>
<td>1.69</td>
<td>0.09</td>
<td>14.1</td>
<td>1.00</td>
<td></td>
</tr>
<tr>
<td>Fan$^a$</td>
<td>1</td>
<td>20</td>
<td>1.67</td>
<td>11.0</td>
<td>1.00</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fan Heater$^a$</td>
<td>1</td>
<td>15</td>
<td>1.50</td>
<td>0.05</td>
<td>87.1</td>
<td>27.2</td>
<td>4.07</td>
</tr>
<tr>
<td>Hair Dryer$^a$</td>
<td>1</td>
<td>45</td>
<td>1.36</td>
<td>0.04</td>
<td>9.5</td>
<td>1.06</td>
<td>0.11</td>
</tr>
<tr>
<td>Shower$^a$</td>
<td>1</td>
<td>20</td>
<td>1.08</td>
<td>0.04</td>
<td>10.7</td>
<td>1.37</td>
<td>0.78</td>
</tr>
<tr>
<td>Washing M$^a$</td>
<td>1</td>
<td>43</td>
<td>2.05</td>
<td>0.12</td>
<td>11.1</td>
<td>1.18</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Note: N: sample number; CVE oil: Candle Vapour Eucalypt oil; Washing M: washing machine. $^a$no S.D.
6.3.1.1 \textit{PM}_{2.5} \textit{concentrations and emission rates}

While there are large errors associated with the concentration levels presented in Table 6.1 for all the activities, it can be seen that some activities result in particularly high indoor \textit{PM}_{2.5} concentrations. These include frying (median peak value: 745 \(\mu\text{g m}^{-3}\)), grilling (718 \(\mu\text{g m}^{-3}\)), candle vaporizing eucalyptus oil (132 \(\mu\text{g m}^{-3}\)) and smoking (79 \(\mu\text{g m}^{-3}\)). The high median peak concentrations caused by these activities may result in exceedence of the US EPA \textit{PM}_{2.5} 24h standards of 65 \(\mu\text{g m}^{-3}\) in the houses where such activities are conducted, provided that duration of the activity is sufficiently long. Indoor concentration levels are significantly elevated as a result of these activities, as can be concluded from the ratios of concentrations during the activity to the background level. There was an increase over the background by 89, 32 and 3 times during grilling, frying and smoking, respectively.

As could be expected from the above discussion, the estimated \textit{PM}_{2.5} emission rates from frying, grilling, cooking pizza, smoking and vaporizing eucalyptus oil were the highest compared with other indoor particle emission sources investigated in this study. The emission rates derived from this study can be compared with those presented in the literature, however, for some activities the comparison is not straightforward since the classification of the activity as described in the literature was not necessarily the same as in this study.

For example, Wallace (1996) concluded from a review of three major studies on particle concentrations in US homes, that cooking results in emission of 1.7±0.6 mg minute\(^{-1}\) of \textit{PM}_{2.5}, and sources other than cooking and smoking in emission of about 0.018±0.017 mg minute\(^{-1}\). Thus, the estimate of emission rates from cooking is higher than in this study (0.11 mg minute\(^{-1}\)), however it was not specified in the
review how the cooking was conducted or whether it included frying or grilling, which both have very high emission rates. This value is, however, within the range of values found in this study for emission rates related to various types of cooking activities and ranging from 0.03 mg minute\(^{-1}\) (microwave) to 2.78 mg minute\(^{-1}\) (grilling).

The emission rate of 0.99 mg minute\(^{-1}\) resulting from tobacco smoking found in this study is comparable to the results presented in the literature. For example, Klepeis et al. (1999) measured respirable suspended particle (PM\(_{3.5}\)) emitted in a residence where smoking took place. They found that the average particle emission rate of PM\(_{3.5}\) ranged from 0.98 mg minute\(^{-1}\) (cigar) to 1.9 mg minute\(^{-1}\) (Marlboro cigarette). Brauer et al. (2000) conducted a measurement of cigarette smoking by using a nephelometer in environmental chamber settings and found that the particle emission rate of PM\(_{2.5}\) was 1.67 mg minute\(^{-1}\). Other researchers characterised tobacco smoking in terms of emission factors, which could be compared to emission rates only if the duration of smoking was known. For example, Özkaynak et al. (1996) reported the emission factor for PM\(_{2.5}\) to be 13.8± 3.6 mg cigarette\(^{-1}\).

PM\(_{2.5}\) emission rates from candle burning ranging from 0.055 to 0.443 mg minute\(^{-1}\) depending on the burning rate were reported by Fine et al. (1999). The emission rate of candle vaporizing eucalyptus oil of 0.91 mg minute\(^{-1}\) found in this study was about twice as the high as the higher end of the range reported by Fine et al. (1999), which, however, it is not surprising considering that both candle burning and eucalyptus oil vapourising result in particle emissions.

Data presented in Table 6.1 also indicate how outdoor particle sources affect indoor particle concentration levels. For example PM\(_{2.5}\) concentration levels could be elevated by about 20%, due to opening the outside door.
6.3.1.2. Particle number concentrations and emission rates

The ratios of peak to background values for submicrometer particle number concentrations presented in Table 6.1 indicate that some types of indoor activities, such as cooking, frying, grilling, stove use, toasting, cooking pizza, smoking, candle vaporizing eucalyptus oil and fan heater use, can elevate the indoor particle number concentrations by a factor ranging from over one to over twenty seven. By comparison with the average outdoor submicrometer particle number concentration in Brisbane of $7.4 \times 10^3$ particle cm$^{-3}$ (Morawska et al., 1999), it could be seen that the median peak values of indoor concentrations were found to be fifteen times higher during cooking, frying, grilling, toasting, cooking pizza and stove use. Another conclusion from Table 6.1 is, that indoor particle number concentration levels were elevated by about 180% due to opening the outside door.

Table 6.1 also provides estimates of particle number emission rates for the source types investigated. It is difficult to compare this data with the literature, as there is very limited information available on particle number emission rates. For example, a study of particle number emission rates of a vacuum cleaner motor was conducted by Lioy et al. (1999) for particles in the size range from 0.3 to 0.5 µm. The authors found that the emission rates ranged from 0.079 to $2.83 \times 10^8$ particle minute$^{-1}$, which is much less than found in this study ($970 \times 10^8$ particle minute$^{-1}$). The difference is not surprising, taking that most particle emissions from the cleaner motor brushes during routine operation are below 0.3 µm in diameter (Helsper et al., 1993), and were thus not measured by Lioy et al. (1999), while they were measured in this study.
6.3.1.3. Discussion

A number of conclusions can be drawn from analysis of the results presented in Table 6.1. One of them is that some activities result in higher emissions in terms of particle number, but not in terms of mass, and vice versa, while some activities result in higher emissions in both (e.g. frying and grilling). This means that each type activity results of emission of particles in a specific size range. For example, a fan heater is a strong emitter in terms of particle number but not in terms of mass, while sweeping floors is a high contributing activity in terms of mass but not in terms of number. This finding supports the theory and the conclusions presented in the literature. For example, early studies by Lefcoe and Inculet (1975) found that household activities such as cleaning or children playing have a pronounced effect on indoor concentrations of particles with diameters greater than 1 µm and a smaller effect on particles with diameters less than 1 µm. The same conclusion can be drawn from this study, where PM$_{2.5}$ ratio of peak to background values for dusting is 1.69, but the number ratio is 1.00. Recently, Abt et al. (2000) reported that oven cooking and toasting contributed primarily to submicrometer particles, sautéing and cleaning to supermicrometer particles and frying contributed to both.

Another conclusion is that contribution from the same types of activities could significantly differ for different environments. An example of this is re-suspension of particles during indoor activities, which has been shown to be an important factor influencing the indoor particle concentration in occupied residential houses. For example, Kamens et al. (1991) found that vacuuming, causes a significant increase in the concentration of particles with diameters greater than 2.5 µm. In this study a range of different results were obtained in relation to vacuuming conducted in
different houses. For example, in House15 the PM$_{2.5}$ concentration during vacuuming increased from 15 to 31 µg m$^{-3}$, while the concentration of particle number did not increase significantly (from 2.06×10$^4$ to 2.38×10$^4$ particle cm$^{-3}$). However, for House16, the PM$_{2.5}$ concentration did not increase significantly (from 13 to 14 µg m$^{-3}$), while the concentration of particles number increased more significantly (from 5.3×10$^3$ to 5.88×10$^4$ particle cm$^{-3}$). One possible reason is that since different types of vacuum cleaners were used in the two houses the emission rates of particle numbers by the vacuum cleaner could have been different. Another possible reason is that the houses differed in general cleanliness levels (e.g. one house vacuumed regularly, while the other irregularly), which affects the reservoir of particles available for re-suspension.

Thirdly, the effect of outdoor air and the activities conducted outdoors on indoor particles can vary significantly. For example, during the neighbour burning off rubbish, both PM$_{2.5}$ concentration and the concentration of particle number increased significantly: from 13 to 90 µg m$^{-3}$ and from 14.3×10$^3$ to 45.2×10$^3$ particle cm$^{-3}$, respectively. However, opening the door after one of the houses was closed for several hours when no strong outdoor source was identified, resulted in a decrease in PM$_{2.5}$ concentration from 32 to 25 µg m$^{-3}$, but a sharp increase in the particle number concentration from 6.4×10$^3$ to 2.62×10$^4$ particle cm$^{-3}$. The changes in mass and number concentrations were rapid and occurred in nine and five minutes, respectively.
6.3.2 Cooking test

The APS, SMPS and DustTrak were employed for the cooking test, and the measurements were conducted under both normal and minimum ventilation conditions for each house. The ratios of peak to background concentrations for all the houses as well as median values of the ratios are presented in Figures 6.1 and 6.2; summary of particle emission rates and particle median diameter is presented in Table 6.2.

![Figure 6.1](image)

Figure 6.1. The ratios of peak to background values for submicrometer (SMPS) and supermicrometer (APS) particle number concentrations and PM$_{2.5}$ concentrations during cooking test under normal ventilation conditions.
Figure 6.2. The ratios of peak to background values for submicrometer (SMPS) and supermicrometer (APS) particle number concentrations and PM$_{2.5}$ concentrations during cooking test under minimum ventilation conditions.

Table 6.2. Summary of the results on emission factor from the cooking test conducted under normal and minimum ventilation conditions, including: submicrometer particle emission rates (particle min$^{-1}$ $\times$ 10$^{12}$) and number median diameters (NMD, $\mu$m) (measured by the SMPS), supermicrometer particle emission rates (particle min$^{-1}$ $\times$ 10$^{8}$) and number median diameters (NMD, $\mu$m) (measured by the APS), as well as approximation of PM$_{2.5}$ emission rates (mg min$^{-1}$) (measured by DustTrak).

<table>
<thead>
<tr>
<th></th>
<th>Normal Ventilation Conditions</th>
<th></th>
<th>Minimum Ventilation Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SMPS</td>
<td>APS</td>
<td>PM$_{2.5}$</td>
</tr>
<tr>
<td></td>
<td>ER</td>
<td>NMD</td>
<td>ER</td>
</tr>
<tr>
<td>Median</td>
<td>1.19</td>
<td>0.026</td>
<td>0.67</td>
</tr>
<tr>
<td>Average</td>
<td>1.36</td>
<td>0.031</td>
<td>3.06</td>
</tr>
<tr>
<td>S.D.</td>
<td>1.03</td>
<td>0.011</td>
<td>5.48</td>
</tr>
<tr>
<td>Min</td>
<td>0.20</td>
<td>0.022</td>
<td>0.12</td>
</tr>
<tr>
<td>Max</td>
<td>4.01</td>
<td>0.063</td>
<td>19.68</td>
</tr>
</tbody>
</table>

Min: Minimum; Max: Maximum; ER: emission rates.
Figures 6.1 and 6.2 show that the impact of cooking on indoor particle concentration levels varied from house to house, but it is clear that as a result of cooking, indoor particle concentration levels were elevated for both ventilation conditions (the ratio of the concentrations was always higher than one). From comparison of the two figures, it can be seen, that generally the impact of cooking on indoor particle number concentration levels are higher for minimum than that for normal ventilation conditions.

Inspection of the results presented in Table 6.2 indicates that even though the same procedure of cooking was carried and the same cooking material was used, the emission characterisations (emission rate and number median diameter) varied from house to house. This proves the hypothesis that stove properties and factors such as most likely stove temperature, affect characteristics of emissions resulting from cooking. Particle number size distribution resulting from cooking was always unimodal with the number median diameter ranging from 0.022 to 0.063 µm and from 0.64 to 0.94 µm for particles in the ranges 0.015 to 0.685 µm and 0.5 to 20 µm, respectively.

Statistical analysis (t-test) of the data obtained from the cooking tests demonstrated that indoor particle concentrations, including particle number and PM$_{2.5}$, under minimum or normal ventilation conditions, were significantly elevated as a result of cooking ($p < 0.05$), with the exception of particle number concentration in the range 0.5 to 20 µm under normal ventilation concentration. The K-S test results for cooking test confirmed that emissions from cooking can have significant impacts on indoor submicrometer size distributions, however, not on indoor supermicrometer size distributions, for many houses.
Statistical analysis (t-test) also demonstrated that there were no significant (p = 0.05) differences between the two ventilation conditions in any of the characteristics of indoor particles generated by cooking (e.g. particle number and PM$_{2.5}$ peak values, emission rates and number median), with the exception of the ratios for submicrometer particles (p = 0.018). This means that, as expected, the ventilation condition did not affect the emission characterisation in any visible way. However, ventilation conditions affect the decay behaviour of the generated aerosols by cooking. Statistical analysis of the results indicates that the decay rates of particle number and mass under normal ventilation conditions were significantly higher than those under minimum ventilation conditions (p = 0.05).

Among all these houses, only two of them (House13 and House15) were equipped with gas stoves, while all the other houses use electric stoves. It was expected that gas combustion might result in generation of higher numbers of particles in the submicrometer range. However, the results of statistical analysis (t-test) of all the characteristics (peak values, ratios, emission rates and number median diameter under both ventilation conditions) indicate that there were no significant (p=0.05) differences between the two types of stove, which is likely due to a small number of houses sampled.

Analysis of time series results for the cooking test indicates that in most cases there is a time delay in the increase in PM$_{2.5}$ and supermicrometer particle concentrations compared with the increase in concentration of submicrometer particles. This is likely to be due to coagulation of the aerosol, which results in the shift in particle size distribution towards larger sizes with time. While this process alone does not change the mass of airborne particles, however, larger particles are detected by the
DustTrack with higher efficiency, therefore there is an increase in the measured PM$_{2.5}$ recorded. Figures 6.3 and 6.4 present typical examples of this phenomenon under minimum and normal ventilation conditions, respectively. From these figures it is also evident that the decay rate under minimum ventilation condition is clearly lower than that under normal ventilation condition. The particle concentration levels decrease to background levels about 15 minutes after conclusion of cooking under normal ventilation condition (Figure 6.4), while they are still in high (two times higher than background levels) after 45 minutes after conclusion of cooking under minimum ventilation condition (Figure 6.3).

Figure 6.3. A typical example of changes in submicrometer (SMPS) and supermicrometer (APS) particle number concentration, as well as PM$_{2.5}$ concentration with time during the cooking test under minimum ventilation condition (House32, 9/07/1999)
Figure 6.4. A typical example of submicrometer particles concentration (SMPS), supermicrometer particle concentration and PM$_{2.5}$ concentration variations with time during the cooking test under normal ventilation condition (House12, 12/05/1999)

Literature review demonstrates that the peak values of particle concentration and number median diameter (NMD) during cooking varied between studies. For example, the NMD for submicrometer particles generated by cooking was 60 nm, 54 nm, and 30 nm as reported by Stoute et al. (1984), Tu and Knutson (1988) and Strong (1988), respectively. Li et al. (1993) investigated characteristics of submicrometer and ultrafine particles (small than 0.1 µm) resulting from cooking scrambled eggs and frying chicken in a residential apartment. They found that the peak values ranged from 1.12×10$^5$ to 1.80×10$^5$ particle cm$^{-3}$ with NMD ranging from 33 to 47 nm for scrambled eggs. For frying chicken, peak values ranged from 1.20×10$^5$ to 2.60×10$^5$ particle cm$^{-3}$ with average NMD of 60 nm. The average peak values in this study of 1.65~1.77×10$^5$ particle cm$^{-3}$ are comparable with the results of the above-mentioned studies. However, in this study the ultrafine particles
constituted about 95% of total submicrometer particles, which is higher than the 80-85% contribution reported by Li et al. (1993).

6.4 Conclusions

In this study particle number and mass concentration levels were measured in 15 residential houses for a period of 48 hours and the residents of the houses kept diaries of the activities conducted. Particle number concentration was measured in two size ranges: from 0.007 to 0.808 µm (called submicrometer particles for the purpose of this study) and from 0.5 to 20 µm (called supermicrometer particles). Particle mass concentration was measured as an approximation of PM$_{2.5}$ concentrations. In addition a cooking test was conducted in each house according to an identical procedure, in order to investigate the impact of stove and house ventilation conditions on the emission and concentration characteristics of particles. It should be noted that submicrometer particle number concentration measured during the cooking test was in the size range from 0.015 to 0.685 µm.

The most important achievement of this study is the quantification of the effect of indoor sources on indoor particle concentration level and emission rates from twenty types of indoor sources or activities. Such data in relation to particle number emissions has so far been largely unavailable. The study showed that indoor activities affect indoor particle concentration levels, with the degree of effect depending on the type of the source and on house characteristics.

Among the indoor activities recorded in this study, cooking, frying, grilling, stove use, toasting, making pizza, smoking, candle vapourising eucalyptus oil and fan heater use were showed to elevate the indoor particle number concentrations in the
range from 1.5 to over 27 times. Indoor approximation of PM$_{2.5}$ concentrations showed an increase over the background by 3, 30 and 90 times during smoking, grilling and frying respectively. The results of cooking tests give insight into the characterisations of cooking as a source of particles and indicate that even though the same procedure of cooking was carried and the same cooking material was used, the emission characterisations (emission rate and number median diameter) varied from house to house.

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CHAPTER 7

PARTICLE DEPOSITION RATES IN RESIDENTIAL HOUSES

Congrong He¹, Lidia Morawska¹, Dale Gilbert²

¹International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, QLD 4001, Australia

²Built Environment Research Unit, Queensland Department of Public Works, Brisbane, QLD 4001, Australia

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Title: A pilot study on particle deposition rates in residential houses

Authors: Congrong He, Lidia Morawska, and Dale Gilbert

Congrong He (candidate)
Developed experimental design and scientific method; analysed and interpreted data and wrote manuscript

Lidia Morawska
Contributed to experimental design and scientific method, interpreted data; assisted with manuscript

Dale Gilbert
Assisted with data interpretation and manuscript
Abstract

As part of a large study investigating indoor air in residential houses in Brisbane, Australia, the purpose of this work was to quantify the particle deposition rate of size classified particles in the size range from 0.015 to 6 \( \mu \text{m} \). Particle size distribution resulting from cooking, repeated under two different ventilation conditions in 14 houses, as well as changes to particle size distribution as a function of time, were measured using a scanning mobility particle sizer (SMPS), an aerodynamic particle sizer (APS), and a DustTrak. Deposition rates were determined by regression fitting of the measured size-resolved particle number and PM\(_{2.5}\) concentration decay curves, and accounting for air exchange rate.

The measured deposition rates were shown to be particle size dependent and they varied from house to house. The lowest deposition rates were found for particles in the size range from 0.2 to 0.3 \( \mu \text{m} \) for both minimum (air exchange rate: 0.61±0.45 h\(^{-1}\)) and normal (air exchange rate: 3.00±1.23 h\(^{-1}\)) ventilation conditions. The results of statistical analysis indicated that ventilation condition (measured in terms of air exchange rate) was an important factor affecting deposition rates for particles in the size range from 0.08 to 1.0 \( \mu \text{m} \), but not for particles smaller than 0.08 \( \mu \text{m} \) or larger than 1.0 \( \mu \text{m} \). Particle coagulation was assessed to be negligible compared to the two other processes of removal: ventilation and deposition. This study of particle deposition rates, the largest conducted so far in terms of the number of residential houses investigated, demonstrated trends in deposition rates comparable with studies previously reported, usually for significantly smaller samples of houses (often only one). However, the results compare better with studies which, similarly to this study,
investigated cooking as a source of particles (particle sources investigated in other studies included general activity, cleaning, artificial particles, etc).

**Keywords:** indoor air quality; indoor particle deposition; supermicrometer particles; submicrometer particles; PM$_{2.5}$
7.1 Introduction

It is well known that inhalation exposure to airborne particles, which occurs in outdoor and indoor environments, has adverse effects on human health. However, since people spend approximately 90% (95% in Australia, ABS 1996) of their time indoors (Fishbein and Henry, 1991; Jenkins et al., 1992; Byrne, 1998) and indoor particle concentrations often exceed outdoor concentrations (Yocom, 1982; Wallace, 1996; Monn, 2001), indoor exposures are major contributors to total personal exposures (Janssen et al., 1998).

As a result of a growing concern about the effects of particle exposure on human health, an increasing interest has been directed towards understanding and quantification of the mechanisms controlling particle dynamics in indoor environments. One of the significant mechanisms is particle deposition on indoor surfaces, which occurs when an aerosol particle adheres to a surface upon contact. In indoor environments, particle deposition rate and air exchange rate are the two main components of the overall particle removal rate from the air. Knowledge of particle deposition rates onto indoor surfaces, and the factors governing these rates, is therefore important to the understanding of particle fate in indoor air.

Indoor particle deposition rates have been investigated using the two available approaches, which are: theoretical modelling and experimental studies. The experimental investigations included both chamber and real house studies. However, owing to the large number of contributing factors, it is very difficult to measure particle deposition rates directly. As a result, the existing experimental data on the levels of deposition rates and their variability within residential houses, particularly for short time periods and for discrete particle sizes, are very limited (Howard-Reed
et al., 2003). In summary, previous studies have shown that: 1) it is clear that the deposition process is strongly dependent on particle size, reaching minimums at particle diameters between 0.1 to 0.3 µm (Lai, 2002); 2) the trend in deposition rates as a function of particle size is the same for all the studies, however, there is a significant variation, often by an order of magnitude or more, in the values reported by individual studies; 3) while both modelling and experimental approaches predict similar trends, model estimates are often significantly different from those derived from the experimental results, especially for particles smaller than about 0.5 µm (Morawska and Salthammer, 2003).

As part of a large study investigating various aspects of indoor air in residential houses in Brisbane, Australia, the purpose of this work was to investigate particle deposition characteristics in indoor environments. The specific objectives of the study included: 1) quantification of particle deposition rates indoors in terms of particle number and mass concentrations for normal and minimum ventilation conditions; and 2) comparison of the results with literature data.

7.2 Experimental Method

7.2.1. Test houses

The determination of deposition rates of particles resulting from cooking conducted in an identical way in all of the houses was a part of a larger indoor air quality study, with the other aspects of the study described elsewhere (Morawska et al 2001; Morawska et al 2003; Hargreaves et al 2003; He et al., 2004; Ayoko et al 2004). In summary, a residential suburb in Brisbane of reasonably flat topography and with a good mix of houses, both in terms of age and of style (i.e. newer and older houses,
brick and timber, high set and low set), was chosen as the measurement site. Fourteen houses from this suburb were selected for the study, as well as one additional house from another suburb as a comparison site.

7.2.2 Determination of particle size and number concentration

Particle size and number concentration measurements were conducted using two different instruments: 1) the TSI Model 3320 Aerodynamic Particle Sizer (APS) (TSI Incorporated, St. Paul, MN, USA), which measures particle size distribution and number concentration in real time, in the range from 0.5 to 20 $\mu$m and up to the order of $10^2$ or $10^3$ particle cm$^{-3}$ with coincidence errors of 1% and 5% respectively; 2) the TSI Model 3934 Scanning Mobility Particle Sizer (SMPS) (TSI Incorporated, St. Paul, MN, USA) linked to a Condensation Nucleus Counter (CPC) (TSI Model 3022A, St. Paul, MN, USA), which was set to measure particle size distribution and number concentration, in the range from 0.015 to 0.685 $\mu$m.

For the purpose of this paper, particles measured by the SMPS are referred to as submicrometer particles; while particles measured by the APS are considered supermicrometer particles.

7.2.3 $PM_{2.5}$ collection

An approximation of fine particle mass concentration ($PM_{2.5}$) was measured by the TSI Model 8520 DustTrak aerosol monitor (TSI Incorporated, St. Paul, MN, USA). It should be noted that the DustTrak operates based on a light scattering technique, where the amount of scattered light is proportional to the volume concentration of the aerosol. The approximation of $PM_{2.5}$ values obtained in this study using this instrument are not actual gravimetric values, as the instrument was not calibrated for each specific aerosol studied. However, in order to obtain results closer to the true
PM$_{2.5}$ values, data collected by the DustTrak were corrected using an equation which was obtained from a calibration experiment. The experiment was conducted under laboratory conditions, and compared the DustTrak indoor readings with the readings of a TEOM (50ºC R&P 1400a with a URG PM$_{2.5}$ cyclone inlet). For simplification, all the DustTrak results discussed in this paper are referred to as PM$_{2.5}$, omitting the term ‘approximation’.

7.2.4 Measurement of CO$_2$

The TSI Model 8551 Qtrak (Q-Trak - TSI Incorporated, St. Paul, MN, USA) was employed to measure CO$_2$ concentrations, which were used to estimate air exchange rates (AER).

These measurements were conducted under both normal and minimum ventilation conditions for each house. Normal ventilation condition means all the doors and windows of the house that are normally opened, stayed opened during the measurements. Minimum ventilation condition means that all the doors and windows of the house were closed.

The procedure for measurement was as follows: 10 minutes of background concentration measurements in the kitchen were made, followed by measurements during 10 minutes of cooking (half an onion was sliced and placed in a small frying pan containing 1 spoonful of vegetable oil, and the stove was switched onto high). A further 20 to 40 minutes of measurements were taken after the cooking ceased. The measurements were conducted first under minimum ventilation conditions, and then under normal ventilation conditions (in all houses with the exception of House 7). The data obtained from the test were used for estimation of particle deposition rates in conjunction with AER.
7.2.5 Calculation of air exchange rate

The tracer gas technique employed to calculate the air exchange rate involved injecting a tracer gas and mixing it through the house, then measuring its decay rate with an appropriate instrument. If exfiltration rates of the tracer gas are constant, mixing is uniform, the chemical reaction between the gas and other chemicals is negligible and no indoor source of the gas is operating, then the air exchange rate, $\alpha$, can be calculated from the following equation (Nantka, 1990):

$$\alpha = \frac{1}{t} \ln \frac{C_t}{C_0}$$

(1)

where $t$ is time, and $C_t$ and $C_0$ are concentrations of the gas at times $t$ and 0, respectively.

In this study, the gas used for AER measurements was CO$_2$. This was released into the house by a CO$_2$ cylinder until the indoor CO$_2$ level was more than 50% higher than the background level and a relatively stable concentration condition was established. Based on Equation 1, the value of AER was obtained by linear regression of the measured CO$_2$ concentration (decay rate) for each house. In order to reduce indoor sources of CO$_2$, there were no people in the houses during the CO$_2$ measurements.

7.2.6 Estimation of particle deposition rates

Residential houses in Brisbane do not normally use air filtration systems. This means that the principal factors governing the levels of airborne particles indoors are the contributions from indoor and outdoor sources, the deposition rate of particles on indoor surfaces, and the air exchange rate (Thatcher and Layton, 1995). Taking these factors into consideration and assuming well-mixed conditions, a formula for
calculation of indoor particle concentration levels can be written as follows (Koutrakis et al. 1992; Chen et al., 2000):

\[ \frac{dC_{\text{in}}}{dt} = P \alpha C_{\text{out}} + \frac{Q_{s}}{V} - (\alpha + \kappa)C_{\text{in}} \]  

(2)

where \( C_{\text{in}} \) and \( C_{\text{out}} \) are the indoor and outdoor particle concentrations, respectively; \( P \) is the penetration efficiency; \( \alpha \) is the air exchange rate; \( k \) is the deposition rate; \( Q_{s} \) is the indoor particle generation rate; \( t \) is time; and \( V \) is the efficient volume of the house. All the factors in this equation, with the exception of the efficient volume of the house (\( V \)), are functions of some other factors and can vary in time (for example penetration efficiency is a function of particle size). In the absence of indoor particle sources, Equation 2 can be written as:

\[ \frac{dC_{\text{in}}}{dt} = P \alpha C_{\text{out}} - (\alpha + \kappa)C_{\text{in}} \]  

(3)

In order to estimate the particle deposition rates (\( k \)), Equation 3 is simplified by assuming that \( \alpha \) and \( k \) are constants and \( P \) equals one. The penetration efficiency is commonly assumed to be close to one for both fine and coarse particles (Wallace 1996). This assumption simplifies Equation 3, however, it should be noted that a number of studies have indicated that the penetration efficiency is less than one and is a function of particle size (Abt et al., 2000; Long et al., 2001). It was considered that the uncertainty resulting from the assumption of \( P \) being equal to one in this study was negligible. This is because concentrations during the cooking period were significantly higher (more than 10 times) than background levels (\( C_{\text{in0}} >> C_{\text{out}} \)), thus the contribution of the outdoor source was comparatively small and could be ignored. Based on the above, the time-dependent solution to Equation 3 becomes:

\[ \ln\left(\frac{C_{\text{in}}}{C_{\text{in0}}}\right) = -(\alpha + \kappa)t \]  

(4)
where, $C_{in0}$ is the peak indoor particle concentration. Based on this equation, it is possible to determine $k$ by fitting a line to a plot of the log of $C_{in}/C_{in0}$ versus time and subtracting $\alpha$ from the slope. Several previous studies discuss the use of this equation for determination of particle deposition rate (Abt et al., 2000; Vette et al., 2001; Howard-Reed et al., 2003).

2.7. Data processing and analysis

The SMPS and APS provide a primary determination of the number of particles per unit volume. The volume of these particles was determined by assuming sphericity and multiplying the number of particles by $\pi d^3/6$. Diameter, $d$, as measured by the SMPS and APS was limited in precision by the dimensions of the bin boundaries as specified by the manufacturer. The diameter used for calculations was the logarithmic midpoint (geometric mean) of these bins boundaries. Thus, measured particle characteristics were expressed as number concentrations (particle cm$^{-3}$) and volume concentrations ($\mu$m$^3$ cm$^{-3}$), and divided into 18 size intervals: 0.015-0.02, 0.02-0.03, 0.03-0.04, 0.04-0.06, 0.06-0.08, 0.08-0.1, 0.1-0.15, 0.15-0.2, 0.2-0.3, 0.3-0.4, 0.4-0.5, 0.5-0.65, 0.7-1, 1-2, 2-3, 3-4, 4-5, 5-6 $\mu$m; for each house under both ventilation conditions. Data analysis showed that particle concentration levels for larger particles (> 6 $\mu$m) were very low and thus these particular results were associated with very large errors. Therefore these data were excluded from deposition rate quantification.

The statistical analyses conducted included regressions and t-tests, and were performed using Microsoft Excel and a statistical analysis software package, S-Plus for Windows version 6.0 (Insightful, 2001). A level of significance of $p = 0.05$ was used for all statistical procedures.
7.3. Results and Discussion

7.3.1 Deposition rate and its dependence on air exchange rate

An example of ln(C_{in}/C_{in0}) versus time plots generated by cooking under minimum ventilation conditions (AER = 0.1 h\(^{-1}\)) in one of the houses are presented in Figures 7.1 and 7.2. These two plots represent the 12 submicrometer and 6 supermicrometer particle size intervals respectively, with the results displayed on separate figures to enable easier visual inspection of the trends. As the linear correlation coefficient (R\(^2\)) for particles larger than 6 \(\mu m\) was less than 4, deposition rates for these particles were not considered in the further analysis.

Figure 7.1. An example of the relationship between ln(C_{in}/C_{in0}) and time for 12 submicrometer particle size intervals.
Figure 7.2. An example of the relationships between ln(C\textsubscript{in}/C\textsubscript{in0}) and time for 6 supermicrometer particle size intervals.

The measured deposition rates varied from house to house under both ventilation conditions, but especially for normal ventilation. These variations are expected to be due to the differences in the surface-to-volume ratio, turbulent mixing patterns, and the types of houses and internal surfaces, all of which affect the rate of particle deposition on surfaces (Abadie et al., 2001; Long et al., 2001; Thatcher et al., 2002).

The average particle deposition rates for the 18 size intervals under minimum and normal ventilation conditions are presented in Figures 7.3 and 7.4, respectively. It can be seen from these figures that the deposition rates are particle size specific. The polynomial fit line indicates that the lowest deposition rates were found for particles in the size range from 0.2 to 0.3 μm for both minimum (air exchange rate: 0.61±0.45 h\textsuperscript{-1}) and normal (air exchange rate: 3.00±1.23 h\textsuperscript{-1}) ventilation conditions. However, the correlation coefficient under minimum ventilation conditions is significant and much higher than that found under normal ventilation conditions. It can also be seen
from Figures 7.3 and 7.4 that the average deposition rates under normal ventilation conditions are higher than those under minimum ventilation conditions for all particle sizes studied. However, statistical analysis (t-test) showed that the differences were significant ($p < 0.05$) for particles within the size range from 0.08 $\mu$m to 1.0 $\mu$m, but not for particles of sizes smaller than 0.08 $\mu$m or larger than 1.0 $\mu$m. This implies that ventilation conditions not only affect the air exchange rate, but also significantly affect the particle deposition rate for particles within the size range from 0.08 $\mu$m to 1.0 $\mu$m.

Figure 7.3. The average of particle deposition rates (■) for the 18 particle size intervals under normal ventilation conditions (Error bars represent one standard deviation). The polynomial fit line with the correlation coefficient ($R^2 = 0.33$)
It has been recognized and confirmed that turbulence, influenced by the air exchange rate, is an important factor affecting particle deposition indoors (Xu et al., 1994; Mosley et al., 2001; Thatcher et al., 2002). Although the results of chamber studies (Nomura et al., 1997) have indicated that there is a positive correlation between air exchange rate and particle deposition rate; the real house studies showed inconsistent results. For example, data reported by Fogh et al., (1997), Abt et al., (2000) and Long et al., (2001) showed that correlation existed between these two factors; but the results reported by Howard-Reed et al., (2003) did not show any correlation. Furthermore, data from Thatcher and Layton (1995) showed deposition rates decreasing with higher air exchange rates. These inconsistent results may be due to a
number of factors including different study designs, different type and size range of particles investigated or different measurement techniques.

The average deposition rates of PM$_{2.5}$ were 2.01±1.11 h$^{-1}$ for minimum ventilation conditions and 3.61±2.6 h$^{-1}$ for normal ventilation conditions. These results obtained from DustTrak measurements are comparable to the results obtained from APS measurements for deposition rates for the particle size interval of 2-3 µm (for minimum ventilation conditions: 2.55±2.1 h$^{-1}$, for normal ventilation conditions: 3.79±2.50 h$^{-1}$).

Since some levels of particle deposition rates found in this study were higher than the levels of air exchange rates, the contribution of particle deposition rates to total particle removal rates could not be negligible and may be more important than AER in residential houses.

7.3.2 The effect of coagulation on particle dynamics

Equation 4, which was used for calculation of deposition rates, does not include coagulation as a factor affecting particle dynamics. In order to assess the effect of coagulation on particle dynamics in this study, calculations of deposition rates conducted using particle number concentrations were repeated for particle volume concentrations, and the two sets of results compared. The deposition rates using particle volume concentrations were estimated using the same method as for the deposition rates using particle number concentrations. If coagulation was a measurable factor, the deposition rate calculated from particle volume concentration would be lower than the deposition rate for particle number concentrations. This is because some of the particle loss accounted for as deposition in particle number calculations, would in fact be due to particle coagulation. Coagulation would
increase volume concentration and would therefore result in an increase of what has been calculated as the deposition rate. The results of comparison (t-test) indicated that there is no statistical difference between the two types of deposition rates (volume and number) for any of the particle size ranges studied under both ventilation conditions. This implies that overall, coagulation was not a significant factor for the particles investigated in this study, and therefore can be neglected.

7.3.3 Comparison with literature

As mentioned above, studies on particle deposition indoors can be classified into two categories: modelling and experimental studies. Experimental studies can be conducted in experimental chambers or controlled test houses (Xu et al., 1994; Byrne et al., 1995; Mosley et al., 2001; Thatcher et al., 2002; Lai et al., 2002), and in residential houses (uncontrolled real houses) (Thatcher and Layton 1995; Abt et al., 2000; Wallace et al., 2004). Recently, Thatcher et al. (2002) compared the results of nine experimental studies, including their own, and also one modelling study published in the literature on indoor particle deposition. Similarly, Lia (2002) compared the results of fifteen experimental studies, conducted using either chambers or controlled test houses, as well as in residential houses. Howard-Reed et al. (2004) compared four controlled test house experiments and seven residential house experiments, including their own. Two of the main conclusions from these comparisons were that the results from individual experimental studies were fairly scattered and that model results were about an order of magnitude below that of the experimental results, especially for particles smaller than about 0.5 \( \mu \text{m} \). The deposition rates measured in the controlled studies (chamber studies or test houses) were shown to be up to an order of magnitude smaller that those measured in the residential house studies, again particularly for smaller particles.
In order to compare the findings of this study with the results published in literature, nine experimental studies of particle deposition conducted by other investigators in residential houses were selected. A summary of the experimental conditions of the studies (including this study) is presented in Table 7.1, while Figure 7.5 presents a comparison of the size dependent particle deposition rates from the studies. Only under minimum ventilation conditions was the average AER (0.61 h\(^{-1}\)) in this study close to the AERs of the nine selected experimental studies. Due to this, only the deposition rates under minimum ventilation conditions from this study were used for comparison.

![Figure 7.5. A comparison of particle deposition rates measured in real houses reported in literature and determined in this study (for minimum ventilation).](image-url)
Table 7.1. A summary of the experimental conditions of the residential house studies on particle deposition rates

<table>
<thead>
<tr>
<th>Study</th>
<th>House</th>
<th>Particle source</th>
<th>Particle size range ((\mu m))</th>
<th>Particle monitor</th>
<th>AER ((h^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thatcher and Layton (1995)</td>
<td>a two-story house</td>
<td>vigorous housecleaning</td>
<td>0.5 – 6</td>
<td>OPC</td>
<td>0.3</td>
</tr>
<tr>
<td>Fogh et al. (1997)</td>
<td>4 houses (furnished)</td>
<td>labelled silica particle</td>
<td>0.5 – 5.5</td>
<td>APS, air filter sample</td>
<td>0.077-1.128</td>
</tr>
<tr>
<td>Abt et al. (2000)</td>
<td>4 houses</td>
<td>cooking</td>
<td>0.02 - 10</td>
<td>SMPS, APS</td>
<td>0.17-0.65</td>
</tr>
<tr>
<td>Long et al. (2001)</td>
<td>9 nonsmoking houses</td>
<td>ambient</td>
<td>0.02 - 10</td>
<td>SMPS, APS, TEOM</td>
<td>0.89 (Winter)</td>
</tr>
<tr>
<td>Vette et al. (2001)</td>
<td>a vacant residence</td>
<td>ambient</td>
<td>0.012 – 2.5</td>
<td>SMPS, LASX, PM$<em>{2.5}$ and PM$</em>{5}$ cyclone samplers, particle counter</td>
<td>2.1 (Summer)</td>
</tr>
<tr>
<td>Ferro et al. (2004)</td>
<td>1 house</td>
<td>non-cooking human activity</td>
<td>0.3 - &gt;5</td>
<td>APS, SMPS, DustTrak</td>
<td>0.46 ± 0.14</td>
</tr>
<tr>
<td>Howard-Reed et al. (2003)</td>
<td>a townhouse</td>
<td>cooking, citronella candle, kitty litter</td>
<td>0.3 - &gt;10</td>
<td>OPC</td>
<td>0.64 ± 0.56</td>
</tr>
<tr>
<td>Wallance et al. (2004)</td>
<td>a townhouse</td>
<td>Cooking only</td>
<td>0.0106 – 2.5</td>
<td>APS, SMPS</td>
<td>0.39± 0.26</td>
</tr>
<tr>
<td>Wallance et al. (2004)b</td>
<td>a townhouse</td>
<td>cooking, citronella candle, kitty litter</td>
<td>0.0106– 5.425</td>
<td>APS, SMPS, OPC</td>
<td>0.64± 0.56</td>
</tr>
<tr>
<td>This study</td>
<td>14 houses</td>
<td>Cooking only</td>
<td>0.014 - 10</td>
<td>APS, SMPS, DustTrak</td>
<td>0.61 ± 0.45</td>
</tr>
</tbody>
</table>

APS: aerodynamic particle sizer; LASX: laser aerosol spectrometer; OPC: optical particle counter; TEOM: tapered element oscillating microbalance monitor; SMPS: scanning mobility particle sizer
Several conclusions can be derived from inspection of the data presented in Figure 7.5. Firstly, although in general the pattern of the deposition rate as a function of particle size is similar for all the studies, deposition rates vary substantially between the studies. Secondly, particle type may be a factor influencing deposition rates. In particular, the deposition rates for particles with size $< 0.05 \mu m$ obtained from all the studies where cooking was a source of particles (Abt et al., 2000; Wallance et al., 2004a; 2004b; this study) are higher than those with non-cooking sources. The results of this study (under minimum ventilation conditions) compare better with the values of Wallace et al. 2004a, and Wallace et al. 2004b, in which cooking was a source. The third conclusion that may be drawn is that the method employed for the calculation of deposition rates may contribute to the differences encountered between the studies. The results reported by Long et al. (2001) are generally lower than those of the other studies. However, Long et al., (2001) estimated deposition rates by using regression of $C_{\text{out}}/C_{\text{in}}$, unlike in other studies where regression of $C_{\text{in}}/C_{\text{in0}}$ was used. Although both calculation methods use a physical-statistical model based on the indoor air mass balance equation, the concepts are different. The former one assumes that the indoor particle concentration is in a steady-state, which means that the quantity of particle infiltration from outdoor to indoor equates to the quantity of particles loss by deposition. The decay rate method (the latter one) assumes that the indoor particle concentration is not in a steady-state and that the contribution of particle infiltration from outdoor to indoor can be ignored. Long et al., (2001) noted that their resultant deposition rates were lower than most experimental results reported in the literature, but that there was better agreement between their data and the theoretical predications.
7.4 Conclusions

In summary, this study of particle deposition rates in indoor environments, the largest conducted so far in terms of the number of residential houses investigated, demonstrated deposition rates comparable with studies previously reported, usually for much smaller samples of houses (often only one). Furthermore, size-dependent particle deposition rates were quantified. The study also showed that the effect of ventilation conditions on deposition rates is particle size dependant. The results presented in this study and the comparative analysis of other studies have demonstrated the considerable difficulty in estimating particle deposition rate indoors. This is largely due to the process of particle deposition being complex and controlled by a large number of factors. The fact that deposition rates are associated with large standard divisions and vary from house to house, suggests that care must be taken when choosing values for exposure studies since particle deposition rate is a very significant parameter in determining the indoor particle concentration. These results also suggest that in order to improve the predictive capability of particle deposition models and exposure models, it would be beneficial to employ local experimental data to validate particle deposition models and include the experimental data in exposure models. Since it is impossible to perform measurements for every house, employing experiment results or empirical equations may be a first order solution.

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References


CHAPTER 8

THE EFFECT OF INDOOR AND OUTDOOR SOURCES AND HOUSE CHARACTERISTICS ON INDOOR AIRBORNE PARTICLES AND NO₂

Congrong Hea, Lidia Morawskaa, Kerrie Mengersenb, Dale Gilbertc

aInternational Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, QLD 4001, Australia. Phone: +61 7 3864 2616; Fax: +61 7 3864 9079; Email: l.morawska@qut.edu.au

bSchool of Mathematical Sciences, Queensland University of Technology, Brisbane, QLD 4001, Australia

cBuilt Environment Research Unit, Queensland Department of Public Works and Housing, Brisbane, QLD 4001, Australia

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Authors: Congrong He, Lidia Morawska, Kerrie Mengersen and Dale Gilbert

Congrong He (candidate)
Developed experimental design and scientific method; analysed and interpreted data and wrote manuscript

Lidia Morawska
Contributed to experimental design and scientific method, interpreted data; assisted with manuscript

Kerrie Mengersen
Contributed to data analysed and interpreted data; assisted with manuscript

Dale Gilbert
Assisted with data interpretation and manuscript
Abstract

As part of a larger indoor environmental study, residential indoor and outdoor 48h average levels of nitrogen dioxide (NO\textsubscript{2}), 48h indoor submicrometer particle number (0.007 – 0.808 µm) concentration and the approximation of PM\textsubscript{2.5} concentrations were measured simultaneously for 14 houses in a suburb of Brisbane, Queensland, Australia. These pollutants were also measured at two central monitoring stations, a few kilometres from the suburb. Statistical analyses of the correlation between indoor and outdoor pollutants, and the association between house characteristics and indoor pollutants were conducted.

The average submicrometer particle number concentration levels were 10.9 ± 6.1 (indoor) and 18.2 ± 3.9 (outdoor - QUT station) (particle. cm\textsuperscript{-3} × 10\textsuperscript{3}). The average PM\textsubscript{2.5} concentration levels were 15.5 ± 7.9 (indoor), 8.9 ± 3.1 (outdoor - QUT station) and 10.0 ± 4.0 (outdoor - EF station) (µg m\textsuperscript{-3}). The average NO\textsubscript{2} concentration levels were 13.8 ± 6.3 ppb (indoor) and 16.7 ± 4.2 ppb (house outdoor), respectively.

Analysis of variance showed that there was no significant association between indoor PM\textsubscript{2.5} and any of the house characteristics investigated (p > 0.05). However, associations between indoor submicrometer particle number concentration and some house characteristics (stove type, water heater type, number of cars and condition of paintwork) were significant at the 5% level. Associations between indoor NO\textsubscript{2} and some house characteristics (house age, stove type, heating system, water heater type and floor type) were also significant (p < 0.05). The results of these analyses suggest that gas stoves, gas heating systems and gas water heating systems are the main
indoor sources of indoor submicrometer particles and NO$_2$ concentrations in the investigated residential houses.

8.1 Introduction

Indoor air quality is a function of many contributing factors, the most important include indoor air sources and their characteristics, building characteristics, the use of household cleaning and maintenance products, cooking and air conditioning methods, thermal and moisture conditions, the presence and proximity of outdoor sources, air movement and general climatic conditions. Pollutants include particles, inorganic gases, volatile and semi-volatile organic compounds, heavy metals and other trace elements, biological agents, toxins and carcinogens. The relative importance of these factors or pollutants depends on the nature, location and other characteristics of the indoor and outdoor environments and also on the life style of the occupants. The multiplicity of these factors and their spatial and temporal variability, in addition to many intrinsic difficulties associated with studies of indoor environments (access to the building, interference with its operation, interference with occupants’ activities, etc) make quantification of all the necessary parameters, and interpretation of their relationships for the purpose of impact assessment and control, a very complex task.

Numerous studies conducted worldwide have helped to develop a good understanding of the sources and factors that have a significant impact on indoor air quality. Many studies have also provided quantitative information on individual source strength and their contribution to indoor concentrations of pollutants of interest. Not surprisingly, it has been shown that the results of studies conducted in different geographical locations vary, which may be attributed to the local
differences in the factors contributing to the overall characteristics of indoor air quality.

While general understanding of the key factors affecting indoor air quality has seen much improvement, there are still many local and global questions remaining, the answers to which are of critical importance for designing and maintaining healthier indoor environments.

Firstly, for many places in the world an important question is what is the relationship between indoor and outdoor (measured outside the house) concentrations of pollutants in terms of concentration levels and trend in concentrations? Knowledge of this relationship is significant as it enables quantification of the degree of impact of individual sources and factors. Secondly, also it is also important to have an understanding of the impact of housing characteristics on pollutant concentration levels. Answers to this question provide directions in future housing design, construction and operation to minimize possible negative impacts. Thirdly, another question of significance is what is the relationship between characteristics of pollutants indoors compared to the characteristics of the same pollutants monitored in central monitoring stations? If there is a relationship, and if it could be quantified, it would enable assessment to be made of indoor air quality based on the outdoor monitoring data, without costly indoor studies.

The aim of this work was multifold.

1. In the first instance, the study was conducted to provide information on particulate matter and nitrogen dioxide (NO₂) concentration levels and their trends in residential houses in Brisbane, the third largest city in Australia. To date, relatively little information has been collected about source apportionment, concentration
levels or their trends in Australia in general. This is particularly true for the indoor air of the relatively densely populated area of South East Queensland, and its major urban agglomeration, the city of Brisbane. A handful of reported studies includes a few focused on airborne particles (1-3) and biocontaminants (4). In the study presented here, particulate matter was measured in terms of particle number in the submicrometer range and PM$_{2.5}$. These pollutants were chosen for investigation because of their implications for human health and also the scarcity of data available, particularly in relation to particle number. The interest in these two pollutants also stems from the fact that they are generated by both indoor and outdoor sources and the generation rates are likely to be factors of indoor house characteristics and its operation.

Associations between airborne particles and health effects have been demonstrated through a large number of epidemiological studies, which linked daily mortality and morbidity statistics with increased particle concentrations measured outdoors (5). Since smaller particles have a higher probability of penetration into the deeper parts of the respiratory tract (6-8) and also contain higher levels of trace elements and toxins, such as the polycyclic aromatic hydrocarbons and mutagens (9-10), the fine particle fraction has been the subject of increasing concern. For example, Schwartz et al. (11) reported that daily mortality was more strongly associated with concentrations of fine particles (PM$_{2.5}$) than with concentrations of larger particles in six U.S. cities. Furthermore, health effects studies have also suggested that number concentration may be a better predictor of health effects than mass concentration (12-13). However, owing to the complexity and cost of instrumental techniques for measurements of particle number concentration, data on submicrometer or ultrafine (< 0.1 µm) particles indoors are still limited.
NO₂ is a secondary oxidation product of nitric oxide in the atmosphere and its health impacts include respiratory irritation, increased susceptibility to respiratory infection and lung development impairment (14). As a major indoor pollutant, NO₂ concentration indoors, and the relationship between the indoor and outdoor concentrations of NO₂ have been investigated for decades (15-16). These studies showed that gas stoves, outdoor air and the season (due to changes in ventilation) are the most important parameters influencing indoor NO₂ levels (16). However, these results also indicated that the relationship between indoor levels and personal exposure was associated with geographical conditions, indoor sources, and other demographic characteristics. Despite the extensive research of outdoor air pollutants carried out in Australian cities (17), the data for indoor NO₂ levels and sources in residential houses in Australia are relatively limited (18-19).

2. Secondly, the study aimed at investigating and interpreting the relationship between indoor and outdoor characteristics of the pollutants, with the outdoors measured in the vicinity of the houses as well as in central monitoring stations.

3. Thirdly, the study attempted to provide insight on the impact of house characteristics on the pollutant concentrations.

The work presented here is a part of a larger, multiparameter study program conducted in residential suburban houses in Brisbane, with a specific focus on fine and submicrometer particles (also included in the study were bioaerosols: fungi, bacteria, dust mite, and volatile organic compounds). Outcomes in relation to other investigated aspects of the program and on specific pollutants have been published elsewhere (1-4, 20).
8.2 Methods and techniques

**Sampling Site.** The residential suburb of Tingalpa, located on the eastern side of Brisbane and of reasonably flat topography to ensure that all houses were subject to similar meteorological conditions, was chosen as the measurement site. The distance from Tingalpa to the city centre is about 10 km. Two roads border the site, one a busy arterial road to the west of the site and a closer road of lower traffic density, to the south and southeast.

**Residential Houses included in the study.** As a result of a call for participation in the study distributed to the residents of the suburb, 14 families agreed to include their houses in the study program.

Information about factors relevant to the pollutant characteristics of the houses was obtained by means of a questionnaire, which was divided into three sections. The first part dealt with external features and included questions about the road-type on which dwellings were situated, dwelling structure category and building materials, and surrounding vegetation. Part two consisted of questions on internal features such as floor-type and wall materials, air-conditioning and renovations. The last section included questions about the residents’ household activities - cleaning, gardening, smoking, oven type, pets and health status.

The houses investigated in this study differed in age (from 2 to 100 years), construction material (timber, brick), stove type (electric, gas) and design (high set, low set). High set indicates that the house is elevated above ground on timber or brick stumps and low set describes a house built directly on the ground. House design and material characteristics have an effect on the air exchange rate.
All of the houses were naturally ventilated (one house was equipped with an air conditioner, however this did not operate during the course of this study). The majority of the houses were occupied by non-smokers, with only three of the houses containing occupants that smoked. Most houses used electric stoves and only two houses were equipped with gas stoves.

**Sampling Protocol.** The sampling protocol for indoor particles was described in detail by Morawska et al. (2). In brief, the real-time indoor submicrometer particle number concentrations and the approximation of PM$_{2.5}$ concentrations were measured simultaneously for more than 48 hours in the kitchen of all the houses. Passive samplers were used for indoor NO$_2$ concentration measurements and were placed in the living rooms. The residents of the houses kept detailed diaries of the activities conducted. In addition, outdoor concentration of submicrometer particles was measured at one central monitoring station, outdoor PM$_{10}$ concentrations were measured at two stations, and NO$_2$ concentrations of outside the houses as well as at the two central monitoring stations were measured.

The measurements were conducted between May and July 1999, which corresponds to winter in Brisbane. During this time the temperature ranged between 15 and 23°C (average 20°C), while relative humidity was between 36 and 92% (average 68%).

*Particle measurements*

Particle number concentration in the range from 0.007 to 0.808 µm (referred to for the purpose of this paper as submicrometer particles) was measured using the TSI Model 3022A Condensation Particle Counter (CPC) (TSI Incorporated, St. Paul, MN, USA) with an impactor and 0.0508 cm nozzle with aerosol flow rate of 0.3 L
min⁻¹ condition. Approximation of PM₂.₅ concentrations was measured with the TSI Model 8520 DustTrak aerosol monitor (TSI Incorporated, St. Paul, MN, USA) with a 2.5 µm inlet. The DustTrak operates on the basis of light scattering where the amount of scattered light is proportional to the volume concentration of the aerosol. The approximation of PM₂.₅ values obtained in this study using this instrument are not actual gravimetric mass, as the instrument was not calibrated for each specific aerosol studied. It was, however, compared against TEOM (Tapered Element Oscillating Microbalance monitor) for a general type of indoor aerosol, from which correction factors were derived, thus enabling better estimation of PM₂.₅ than those based on the manufacturer’s calibration.

The CPC and DustTrak were placed side-by-side and positioned on average two metres from the stove in the kitchen. These instruments were chosen as the most suitable for an indoor study for three reasons: 1) the low flow rates required and thus small impact on particle concentrations indoors; 2) their quiet operation, which means less noise and thus fewer nuisances causing less intrusion to the occupants; and 3) the short sampling times of 10 and 30 seconds respectively for the CPC and DustTrak, and the consequent provision of real-time data.

Outdoor particle concentration was measured at the QUT monitoring station in terms of PM₁₀, which was measured using a TEOM (50°C R&P 1400a), and submicrometer particle number concentration, measured using the TSI model 3934 Scanning Particle Sizer (SMPS) operating in the size range from 0.016 to 0.7 µm. In addition, the Eagle Farm (EF) monitoring station’s (EF) PM₁₀ concentration data, also collected using a TEOM (50°C R&P 1400a), were used in this study. The QUT station is situated within the Brisbane CBD, distant from the sampling site by 10 km,
while the EF station is located about 5 km north of the sampling site of Tingalpa. Figure 8.1 presents the location of the sampling site (Tingalpa) and ambient monitoring station sites (QUT and Eagle Farm). The two stations are the closest air quality monitoring stations in relation to the sampling site and thus the data from these two stations were considered to be the most relevant for comparison with the indoor and outdoor data for the investigated houses.

**NO$_2$ concentration measurements**

The passive sampler used for this study was a badge-type unit described by Yanagisawa and Nishimura (21). For indoor measurements, the sampler was placed in the living room more than 2m from the windows and 3m from the sources, facing upwards. For outdoor NO$_2$ measurements, the sampler was placed outside, more than 1m above the ground and protected from rain. An exposure time of 48 hours for all samplers was employed. The quality of NO$_2$ measurements was assured by blanks from each house (indoor and outdoor) to account for differences in sealing quality and lag time before analysis. Once sampling was completed, the NO$_2$ passive badges were analysed by a spectrophotometer (Beckman Model DU640).

Outdoor concentrations of NO$_2$ were also measured at QUT and EF stations using ML 9841B Oxides of Nitrogen Analysers (Ecotech, Melbourne, Australia). It should be noted that the NO$_2$ concentration difference may exist due to the use of two different measurement methods. However, literature data indicated that the accuracy of the badges (passive method) in comparison to the denuder technique (active method) expressed as percent relative error was found to be better than about ±10%, with a variation coefficient of 5.6% for NO$_2$ measurement (22).
**Data processing and analysis.** Based on the real-time particle concentration data, the total 48h average indoor concentrations were calculated for each house. Average concentrations were also calculated based on activity records. The situation indoors was classified into one of two categories: non-activity conditions and activity conditions, identifying a lack or occurrence of occupant activities inside the house. Furthermore, the non-activity 48h average indoor concentrations were calculated for each house.

The 18 house characteristics analysed in this study are: house age (years), house set (highset or lowset), wall materials (brick or timber), smoke (tobacco smoking in the house, yes or no), stove type (electric or gas), house heating (none, electric or gas), water heater type (electric, solar or gas), extractor fan (no, yes (to inside) or yes (to outside)), people (number), car (number), floor (timber or carpet), garage (attached or detached), distance from main road based on the dominant wind direction during the measurement period), pets (yes or no), distance from park, window open frequency (daily, weekly, monthly), bedroom number, and paintwork condition (good or poor). The (linear) correlation between each pair of pollutants was tested using the statistical analysis software package, SPSS for Windows version 10 (SPSS Inc.). In order to better meet the underlying statistical assumptions, log concentrations were used in the analyses of correlation and variance. Analysis of variance was used to test for associations between the pollutants and the house characteristics. Given the small number of houses and the relatively large number of variables to be tested, separate analyses of variance were conducted for each characteristic and pollutant, without account of the other variables. The hypothesis tested in each case was that the average level of the pollutant is the same for each category of the house characteristic under consideration. These analyses were
performed using S-Plus for Windows version 6.0 (Insightful Corp.). Due to the small sample size and the observation that the particle concentrations have skew distributions, the logarithm of the concentrations were used in the statistical analyses.

8.3 Results and Discussion

Data Correction for DustTrak and CPC. Details of the data correction for DustTrak and CPC have been described in Morawska et al. (2). Briefly, in order to obtain results more representative of true PM$_{2.5}$ values, data collected by the DustTrak in this study were corrected using an equation which was obtained from an additional experiment. The experiment was conducted under laboratory conditions to compare the DustTrak indoor readings with the readings of a TEOM (50ºC R&P 1400a with a URG PM$_{2.5}$ cyclone inlet).

The CPC used in this study for the indoor measurements and the SMPS used for ambient measurements at the QUT station had somewhat different ranges of operation. Due to this factor, (and also because they are different instruments), it was expected that they would yield different results when sampling the same aerosol. In order to provide a more accurate relationship between indoor and ambient particle number concentrations measured at QUT, the CPC data were adjusted using an equation based on the result of a comparison experiment between the two instruments (see Morawska et al. (2)).

Finally, in order to compare the indoor and outdoor particle mass concentrations, the ambient (QUT and EF) PM$_{2.5}$ were estimated from PM$_{10}$ values using the ratio of PM$_{2.5}$/PM$_{10}$ = 0.61. This value was provided by the Environmental Protection
Agency (23) based on the data from the monitoring stations in Brisbane for 1999, the year in which this project was conducted.

It is acknowledged that data transformation through the above equations induces uncertainty that is not fully accounted for in the ensuing analyses. Although the transformed values are ‘best estimates’ in that they are expected values and unbiased, any tests of significance made on their basis should be interpreted with caution.

**NO2 and particle concentration levels for all houses.** A summary of 48 hour average indoor, outdoor, and central site monitoring stations NO2, submicrometer particle number and the approximation of PM2.5 concentrations are provided in Table 8.1. 48 hour average indoor and outdoor NO2 concentration levels in 14 monitored houses is showed in Figure 8.2.

The average (±S.D) residential indoor and outdoor NO2 levels were 13.8 ± 6.3 ppb and 16.7 ± 4.2 ppb, with median values of 12.8 and 16.0, respectively. The results of further analysis showed that the indoor/outdoor NO2 concentration ratio was less than unity for all houses except one, ranging from 0.4 to 2.3 with a median value of 0.82. The median ratios of indoor concentrations to the two fixed station NO2 concentrations were higher than the indoor to outdoor ratios, with median values of 1.08 (indoor/QUT station) and 0.87 (indoor/EF station). Comparison between the house outdoor and the two fixed station concentrations show a trend with QUT being the lowest, Eagle Farm higher and house outdoor the highest in concentration. This was possibly due to the sampling site being close to heavy traffic roads and Eagle Farm’s location in an industrial area.
The NO$_2$ concentrations measured in this study can be compared with the data from existing literature. Indoor NO$_2$ concentration levels in Australian residential houses have been investigated in several studies. Dingle et al. (24) found that the indoor mean four-day average level was 28 ppb in Perth, Western Australia, which was slightly higher than the outdoor levels of 24 ppb. Garrett et al. (18) monitored indoor and outdoor NO$_2$ concentration levels using passive samplers in 80 homes in the Latrobe Valley, Victoria. They found that the overall indoor annual median was 5.7 ppb, with a mean of 7.8 ppb. They also found that mean indoor levels were higher than outdoor levels of 4.4 ppb, and that a seasonal variation was evident, with the highest levels both indoor and outdoor recorded in winter. In Brisbane, Queensland, Lee et al. (19) measured indoor and outdoor NO$_2$ concentrations and personal NO$_2$ exposures for 57 subjects in Brisbane. The authors reported that mean indoor and outdoor NO$_2$ concentration levels were 10.5 ppb and 14.5 ppb, respectively, with the indoor/outdoor ratio less than one. The outdoor NO$_2$ concentrations measured in the present study are slightly higher than those reported by Lee et al. (19), but are within the experimental error. However, the results are significantly lower and higher when compared to those in Perth (24) and the Latrobe Valley (18), respectively. These differences reflect the variation in the local outdoor NO$_2$ source strengths and thus concentrations between the three Australian cities. Comparisons between the indoor concentrations show a similar trend to that found outdoors, with the Latrobe Valley being the lowest, Brisbane as measured in this study higher, and Perth the highest. This suggests that outdoor air concentration of NO$_2$ plays a dominant role in contributing to indoor air concentration.

It should be noted, that the sampling location of indoor measurements has been shown to affect the concentrations measured and thus the indoor and outdoor ratios.
For example, Madany and Danish (26) measured the average concentrations of NO₂ inside and outside of 32 homes in Bahrain for 14 days using the passive diffusion tube sampler method. The authors found that NO₂ in the kitchen, hall, bedroom, and outdoors averaged 25.5, 10.8, 7.4, and 12.7 ppb, respectively. Similarly, Chao and Law (27) reported that NO₂ in the kitchen, living room, bedroom, and outdoors averaged 29.7, 26.2, 24.8, and 35.0 ppb, respectively, for two units in Hong Kong, China. It can be seen that the concentrations measured in the kitchen tend to be the highest. In this study the measurements were conducted in the living room and therefore are likely to reflect average house concentrations, rather than extreme concentrations.

The average (±S.D) residential indoor and outdoor (QUT) submicrometer particle number concentration levels were 18.2 ± 3.9 and 10.9 ± 6.1 (particle. cm⁻³ × 10³), with median values of 17.2 and 9.2 (particle. cm⁻³ × 10³), respectively. The average (±S.D) residential indoor submicrometer particle number concentration levels for non-activity situation was 12.4 ± 2.7 (particle. cm⁻³ × 10³). Thus, in general, submicrometer particle number concentration levels were somewhat higher indoor than outdoor during non-activity times, but significantly higher when activity time was included.

The average (±S.D) residential indoor, outdoor (QUT and EF) PM₂.₅ levels were 15.5 ± 7.9, 8.9 ± 3.1 and 10.0 ± 4.0 (µg m⁻³), with median values of 13.6, 7.5 and 9.6, respectively. The average (±S.D) residential indoor PM₂.₅ level for non-activity situation was 11.1 ± 2.6 (µg m⁻³). Thus similarly to submicrometer particle number concentrations, indoor PM₂.₅ levels were higher than the ambient levels.
Abt et al. (28) conducted a study on indoor and outdoor particle number concentrations (reported as particle volume) in four non-smoking households located in the metropolitan Boston area, US. Their results indicated that the mean indoor/outdoor ratio was 2.1 and 1.3 for particles in the size range from 0.02 to 0.1 µm and from 0.1 to 0.5 µm, respectively. Long et al. (29) also measured indoor and outdoor particle number concentrations (reported as particle volume) in nine non-smoking Boston-area homes, US. Their data showed that the mean indoor/outdoor ratio were 1.5 and 1.0 for particles in the size range from 0.02 to 0.1 µm and from 0.1 to 0.5 µm, respectively. The current study result of the ratio indoor/outdoor particle number concentration (about 1.7) is comparable with the results of Abt et al. (28).

**Impact of the type of gas appliances and smoking.** In this study, there were only two houses with gas appliances and three houses with smokers. In order to investigate the hypothesis that gas appliances and cigarette smoking may significantly affect indoor air qualities, the houses with gas appliances or smokers (the two indoor air pollutant sources expected to have the highest contribution) were classified into one category. All the other houses (without gas appliances and non-smoking) were classified into another category. The 48-hour average indoor NO₂ concentrations, indoor submicrometer particle number concentrations, indoor PM₂.₅, and the Indoor/Outdoor NO₂ concentration ratio (I/O) for the two groups are presented in Table 8.2.

It can be seen from Table 8.2 that both the average and the median of NO₂ and submicrometer particle number concentrations for the houses with gas appliances or smoking are significantly higher (p < 0.05) than those for the houses without these two sources. However, neither the mean PM₂.₅ nor the median PM₂.₅ is significantly
different for the two groups of houses. Thus smoking and gas appliances result in the overall increase of both indoor NO₂ and submicrometer number concentrations, but not PM₂.₅. The indoor/outdoor NO₂ ratios are higher for the houses with smoking and gas appliances, however, the difference is not significant \( (p = 0.059) \) for the two groups of houses.

Based on extensive literature review, Wallace (30) concluded that cigarette smoking was an important indoor source of fine and coarse particles, but that gas stove usage was not significant. This was further supported by a recent study of Simoni et al. (31), who reported that PM₂.₅ concentration levels were significantly higher in homes where tobacco smoking took place, and was significantly related to the number of cigarettes smoked. Gotschi et al. (32) analysed 186 indoor/outdoor PM₂.₅ data collected within the EXPLIS study in four cities. Their results indicated that cigarette smoking significantly affected indoor PM₂.₅ concentrations in two cities but not in the other cities. The study also showed that in addition to the outdoor concentrations, the main predictors for PM₂.₅ indoors are the number of cigarettes smoked per day and duration of the use of gas appliances. In the current study, the average number of cigarettes smoked per day in the houses with a smoker was less than 3, and this small number was concluded to be the reason that neither the mean PM₂.₅ nor the median PM₂.₅ is significantly different for the two groups of houses.

**Correlation between pollutants.** In order to examine the degree of linear correlation between the measured pollutants, individual t-tests were conducted for each pair of pollutants. A summary of the results from correlation analysis is provided in Table 8.3.

The results of these analyses indicated that there is no significant correlation between indoor and house outdoor NO₂ concentrations \( (p = 0.428) \), nor between
indoor and the two central monitoring station NO$_2$ concentrations ($p = 0.252$, 0.465 for QUT and EF respectively). However, there are significant correlations between house outdoor and the two central monitoring station NO$_2$ concentrations ($p = 0.014$ and $p = 0.008$, respectively), as well as between house outdoor NO$_2$ and indoor PM$_{2.5}$ concentrations ($p = 0.004$). There is also a significant correlation between indoor NO$_2$ and indoor average submicrometer particle number concentrations ($p = 0.001$), but there is no significant correlation between indoor NO$_2$ and non-activity indoor average submicrometer particle number concentrations ($p = 0.123$).

For particle number, no significant correlation between indoor and QUT submicrometer particle number concentrations was found ($p = 0.204$). Even given non-activity situations, the correlations between indoor and QUT submicrometer particle number concentrations is not improved. For PM$_{2.5}$, there is no significant correlation between indoor concentrations and the two fixed site monitoring stations ($p = 0.693$, $p = 0.201$), but there is significant correlation between indoor non-activity and the two central monitoring stations concentrations ($p = 0.009$, $p = 0.024$).

Several conclusions can be derived from the above analysis. First of all, lack of correlation between indoor and outdoor concentrations of NO$_2$ (neither house nor central monitoring stations) and particles (number and PM$_{2.5}$ mass) suggests that indoor activities significantly affect indoor concentration of these pollutants, and therefore data from outdoor or central monitoring are not representative of the concentrations of these pollutants indoors. However, presence of correlation between house outdoor and central monitoring stations NO$_2$ concentrations suggests that data from these stations are representative of the trends in the concentrations (but not necessarily absolute concentrations) in residential locations. Correlation between
PM$_{2.5}$ concentrations indoors during non-activity periods and those measured at the central monitoring stations suggests that in the absence of human activities, outdoor air is the main contributor to the indoor PM$_{2.5}$ concentrations. Correlation between indoor NO$_2$ and particle number concentrations may suggest a common source for both of these pollutants. However, lack of correlation between indoor particle number and PM$_{2.5}$ suggests different sources of these two particle characteristics, and therefore that no conclusions can be made about one of these characteristics (number or mass concentration), based on a measurement of the other.

In terms of the relationship between NO$_2$ and PM$_{2.5}$ concentrations, the above analysis shows the presence of correlation between house outdoor NO$_2$ and indoor PM$_{2.5}$ concentrations, but lack of correlation between indoor NO$_2$ and indoor PM$_{2.5}$. These results can be compared with the results of NO$_2$ and PM$_{2.5}$ in 140 homes in the Po River Delta area of North Italy, monitored during summer and winter by Simoni et al. (31). A significant relationship between indoor NO$_2$ (living room) and PM$_{2.5}$ concentrations was found during summer, but not in winter. Their study also found a significant relationship between house outdoor NO$_2$ and indoor PM$_{2.5}$ values in winter only. These results are similar to those found in this study conducted in winter. Chan & Hwang (33) examined the relationship between concentrations of several pollutants including NO$_2$, measured at an urban air monitoring station with those measured inside and outside of 22 homes, in a three week monitoring campaign. Similar to the conclusions drawn from this study, the authors found that the central station concentrations of NO$_2$ could be representative of the residential outdoor concentrations, but not of indoor concentrations.

Table 8.3 also presents correlations between the subset of houses without gas appliances, and all houses. One of the interesting observations from Table 8.3 is that
the correlation between indoor NO\textsubscript{2} and non-activity indoor submicrometer particle number concentrations is significantly improved by excluding the houses with gas appliances. In this case, the p value was improved from 0.123 to 0.012. This result implies that both NO\textsubscript{2} and submicrometer particles indoors originate externally and there is a correlation between concentrations of these two pollutants outside (34). It can also be seen that the correlations between indoor and house outdoor NO\textsubscript{2}, as well as indoor and QUT submicrometer particle number concentrations are improved by excluding the houses with gas appliances, but are still not significant (\(p = 0.098, p = 0.095\)). This result implies that data from central monitoring stations of NO\textsubscript{2} and submicrometer particle number concentrations may be a predictor of indoor NO\textsubscript{2} and submicrometer particle number concentrations for houses without gas appliances, but not for houses with gas appliances.

**Analysis of variance between house characteristics and indoor NO\textsubscript{2} and particles.** The results of the analyses of variance between 18 house characteristics and 48 hour average indoor log concentrations of NO\textsubscript{2} and particles (number and PM\textsubscript{2.5}) for activity and non-activity periods are presented in Table 8.4.

It can be seen from Table 8.4, that there are no significant associations between indoor PM\textsubscript{2.5} and any of the listed house characteristics. However, associations between indoor submicrometer particle number concentration and several house characteristics including stove type, water heater type, number of cars and paintwork conditions, are significant (\(p = 0.040, 0.035, 0.020\) and 0.020, respectively). Associations between indoor NO\textsubscript{2} and some house characteristics including house age, stove type, heating system, water heater system and floor type are also significant (\(p = 0.008, 0.030, 0.017, 0.007, 0.042\), respectively). There are no significant associations between indoor non-activity PM\textsubscript{2.5} or particle number
concentrations and any of the listed house characteristics. These results suggest that gas stove, gas heating system and gas water heater system are the main indoor sources of indoor submicrometer particle and NO₂ concentrations.

It should be noted that the Bonferroni correction for the multiple testing in Table 8.4 indicates that in order to maintain an overall 0.5% significance level, the individual p-values must be less than 0.005. None of the results meets this relatively strict criterion. Thus, caution should be exercised, particularly in relation to the results for which the p-values are close to 0.05.

The differences identified in Table 8.4 for heating systems and water heater types were further investigated by multiple comparison tests using Tukey’s method (using S-plus software). The results showed that the indoor NO₂ concentrations are significantly different \((p < 0.05)\) for houses with a gas heating system and those without a heating system or with an electric heating system, and between houses with gas water heater and those with an electric or solar water heater. No significant differences were found between houses without a heating system and with electric heating, or between houses with electric and solar water heaters. The same conclusions were reached for indoor submicrometer particle number as for indoor NO₂ concentrations in relation to the water heater type.

The multiple comparison analyses also indicated that the indoor submicrometer particle number concentrations of the houses with two cars are significantly higher \((p < 0.05)\) than those of the houses with one or three cars. It was expected that an increase in the number of cars used by the occupants would result in increased indoor particle concentrations. This was confirmed by the difference between the impact of one versus two cars, however, the expectation was not reflected in the results for three car households. Further investigation showed that the occupants of
one of the two houses with three cars never parked any of their cars in the garage, but rather on the street; the impact of a car parked on the street is lower than of car parked next to the house. Indoor submicrometer particle number concentrations of the houses with good paintwork conditions are significantly lower \((p = 0.020)\) than that of the houses with deteriorated paintwork conditions. There is no clear explanation of this finding, and similarly no explanation for the significant difference in NO\(_2\) concentrations between houses with timber (23.1 ppb) and carpet (12.3 ppb) floors.

Analysis of the associations between house age and other house characteristics showed that there are significant associations between average house age and stove type \((p = 0.0001)\), heating system \((p = 0.0005)\), and water heater system \((p = 0.0011)\). There are no significant associations between house age and other house characteristics. Further investigation showed that only older houses, aged over 40 years, are equipped with gas appliances. This was concluded to be the reason for the associations between house age and the above characteristics.

Several published studies reported on the associations between indoor pollution and house characteristics. However, some of these studies investigated indoor NO\(_2\) and other gaseous pollutants (e.g. 35-37), some of them investigated indoor PM\(_{10}\) or PM\(_{2.5}\) (e.g. 32, 38), but only one study investigates the associations with particle number concentration (28).

In the study by Simoni et al. (31), the associations between indoor NO\(_2\) and many house characteristics were analysed for both winter and summer. Their results indicated that significantly higher indoor NO\(_2\) levels were in houses with gas-furnace heating and/or with gas water heaters located inside the home \((p < 0.005)\). However, the associations between indoor NO\(_2\) and other house characteristics (such as type of
building, type of parking, presence of smoking, gas appliances, carpets, moquette, fireplaces wood-stove, cooking-air exhauster, new furniture and air conditioning) were not significant in both seasons. A positive association between indoor NO$_2$ and number of open window hours was found in summer ($p < 0.001$).

Cyrys et al. (39) measured indoor and outdoor concentrations of NO$_2$ in 385 dwellings in Erfurt and Hamburg, Germany, and statistically analysed the associations between indoor NO$_2$ and house characteristics, including the period when the house was built, housing material, home location, type of heating, type of stove, what was the windows facing (main road, side street, park or backyard), ventilation condition and smoking. They found that there was significant ($p < 0.05$) association between indoor NO$_2$ level and four house characteristics (gas stove, smoking, what the window was facing and ventilation conditions) for Hamburg, and significant association between indoor NO$_2$ and two house characteristics (gas stove, smoking) for Erfurt.

Recently, Sakai et al. (37) reported the results of indoor and outdoor NO$_2$ and other gaseous pollutants measurements in 37 urban dwellings in Nagoya, Japan, and 27 urban dwelling in Uppsala, Sweden and statistical analysis of the associations between indoor gaseous pollutants and houses characteristics. The house characteristics included type of dwelling (detached or apartment), structure (wooden or non-wooden), age ($\leq 10$ or $\geq 10$), floor area, smoking, and type of heater (clean or unclean). Clean heaters were those which did not generate combustion gases, or combustion heaters with proper ventilation ducting. An unclean heater was a combustion heater without ventilation that emitted combustion gases directly into the indoor living space. They found that in Nagoya, NO$_2$ concentration was significantly higher in modern concrete houses than in wooden houses ($p < 0.01$) and higher in
newer (less than 10 years) than in older dwellings ($p < 0.01$), possibly due to lower air exchange rate and more emission sources in modern buildings. Dwellings heated with unvented combustion sources had significantly higher indoor concentrations of NO$_2$ than those with clean heating ($p < 0.05$). There was no association between indoor NO$_2$ concentration and other house characteristics, such as smoking found in that study. In the study conducted in Uppsala, Sweden, there was no association found between indoor NO$_2$ concentration and any dwelling characteristics. In Australia, Garrett et al. (18) and Lee et al. (19) found significant associations between house characteristics (such as gas stove and smoking) and indoor NO$_2$ concentrations.

In summary, it can be seen that some inconsistencies exist between different studies reported in literature, such as in relation to smoking, or on the associations between indoor NO$_2$ and house characteristics. The results of this current study support findings from the previous study showing that there was significant association between indoor NO$_2$ concentration and indoor gas appliances, however, not for smoking.

With respect to particle mass, Thornton et al. (40) found that PM$_{10}$ levels were slightly elevated for houses in roadside locations. Recently, significant associations between indoor PM$_{10}$ concentration and several house characteristics including house location (urban, suburban or rural), traffic volume (light or heavy), smoking (yes or no) and pet (yes or no), had been found by BéruBé et al. (41) ($p = 0.04$, 0.0038, < 0.0001 and 0.001, respectively). The above mentioned study by Simoni et al. (31) showed that PM$_{2.5}$ was significantly higher in homes where tobacco smoking took place ($p < 0.001$), and was significantly related to the number of cigarettes smoked ($p = 0.01$). The average number of cigarettes per day reported in this study
was 8.7. However, the associations between indoor PM$_{2.5}$ and other house characteristics (such as type of building, type of parking, presence of gas appliances, carpets, moquette, fireplaces wood-stove, cooking-air exhauster, new furniture and air conditioning) were not significant for either of the seasons. Gotschi, et al. (32) observed that the association between indoor PM$_{2.5}$ and several house characteristics was city-specific. For example, cigarette smoking could significantly effect indoor PM$_{2.5}$ concentrations in two cities but not in another city. The average number of cigarettes per day in this study was 8.1. House location could significantly affect indoor PM$_{2.5}$ concentrations in one city but not in the other two cities. The current study shows no significant association between indoor PM$_{2.5}$ concentration and any listed house characteristics including gas appliances and smoking. However, the average number of cigarettes per day in this study was less than 3, which is significantly lower than the numbers in the above two studies. It is concluded that this is the main reason for the lack of significant association between indoor PM$_{2.5}$ and smoking in this study.

For particle number, Abt et al. (28) also analysed the association between indoor particle concentrations and house characteristics, such as gas appliances, house age, house volume, surface area, fraction of house carpeted, number of people. However, no association had been found in their study. They mentioned it was possibly because of the small number of homes sampled.

Comparison of associations between particle number concentration and other factors including house characteristics found in this study and other studies cannot be conducted, since, as discussed above, there is no data available in literature on simultaneous measurements of indoor and outdoor concentrations in residential houses of NO$_2$, submicrometer particle number concentration and PM$_{2.5}$. 
**Limitations of indoor particle number concentration studies.** There are several limitations of this study, which are of similar nature to the studies reported in literature.

Since different measurement methods were employed in this study for measurements conducted at the central monitoring stations and in the houses, systematic differences may exist between the results. However, although these differences could affect the results of the ratios of indoor to central monitoring stations concentrations, the results of correlation analysis will not be affected.

Although the results of this study demonstrated significant associations between several indoor pollutant concentrations and some house characteristics, the main limitation of the study is that it included only a relatively small number houses (or observations). The reason for this was that the measurements of particle characteristics are labour consuming, use relatively costly instruments, and are intrusive to the occupants (the instruments occupy substantial space). Shortage of costly instrumentation for particle number and PM$_{2.5}$ concentrations was also the reason for not conducting 48 hour house outdoor measurements. These are intrinsic difficulties of any indoor particle number concentration studies, and the reason for lack of published data on this topic. Therefore future studies will need to address these challenges by attracting more resources and thus including a larger number of houses. It also must be noted that correlations and other measures of association are computed and interpreted as suggestive of physical pathways between variables of interest. Thus, it is acknowledged that a correlation coefficient only measures the degree of linear association between two variables, not a causal relationship. Further studies are required to confirm the suggested inferences.
Acknowledgments

This project was funded by the Built Environment Research Unit, Queensland Department of Public Works, and Australian Research Council, through SPIRT grant No. C69804416. The assistance of Dr. Kiyoun Lee, Mike King, Keith Eigeland, Chris Greenaway and Gillian Isoardi is gratefully acknowledged. Members of the QUT ILAQH, in particular, Jane Hitchins-Loveday, Sandhya Parappukkaran, are appreciated for their discussions and assistance with this study. The authors would like to express their special gratitude again to the owners and occupants of the houses for their help and in assisting with this project. Without their help and assistance, this project could not have been conducted successfully.

Literature Cited


### TABLE 8.1. Summary of average indoor and outdoor concentration levels of NO\(_2\), submicrometer particle number and PM\(_{2.5}\)

<table>
<thead>
<tr>
<th></th>
<th>average</th>
<th>S.D(^a)</th>
<th>median</th>
<th>max(^b)</th>
<th>min(^c)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>NO(_2) (ppb)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Indoor</td>
<td>13.8</td>
<td>6.3</td>
<td>12.8</td>
<td>32.9</td>
<td>7.3</td>
</tr>
<tr>
<td>House outdoor</td>
<td>16.7</td>
<td>4.2</td>
<td>16.0</td>
<td>26.6</td>
<td>10.4</td>
</tr>
<tr>
<td>QUT</td>
<td>11.3</td>
<td>2.9</td>
<td>10.8</td>
<td>16.0</td>
<td>5.5</td>
</tr>
<tr>
<td>EF</td>
<td>14.4</td>
<td>4.8</td>
<td>14.8</td>
<td>22.5</td>
<td>7.3</td>
</tr>
</tbody>
</table>

| **Particle Number** |         |           |        |           |           |
| **(particle. cm\(^{-3}\) \times 1000)** |         |           |        |           |           |
| Indoor         | 18.2    | 3.9       | 17.2   | 27.2      | 13.6      |
| QUT            | 10.9    | 6.1       | 9.2    | 22.1      | 2.8       |

| **PM\(_{2.5}\) (µg m\(^{-3}\))** |         |           |        |           |           |
| Indoor         | 15.5    | 7.9       | 13.6   | 36.9      | 8.0       |
| QUT            | 8.9     | 3.1       | 7.5    | 15.3      | 5.6       |
| EF             | 10.0    | 4.0       | 9.6    | 19.5      | 4.4       |

| **Non-activity indoor** |         |           |        |           |           |
| **Particle number** |         |           |        |           |           |
| **(particle. cm\(^{-3}\) \times 1000)** |         |           |        |           |           |
| Indoor         | 12.4    | 2.7       | 11.9   | 19.1      | 9.0       |
| QUT            | 11.1    | 2.6       | 11.0   | 17.5      | 7.9       |

\(^a\) S.D: standard deviation. \(^b\) max: maximum. \(^c\) min: minimum.

### TABLE 8.2. The 48-hour average indoor NO\(_2\) concentrations (ppb), submicrometer particle number concentrations (particle. cm\(^{-3}\) \times 10\(^3\)), PM\(_{2.5}\) (µg m\(^{-3}\)), and the Indoor/Outdoor NO\(_2\) concentration ratio (I/O) for two groups of houses: (1) without gas appliances and non-smoking (NG & NS) the (2) gas appliances or smoking (G & S).

<table>
<thead>
<tr>
<th></th>
<th>NG &amp; NS</th>
<th>G or S</th>
<th>NG &amp; NS</th>
<th>G or S</th>
<th>NG &amp; NS</th>
<th>G or S</th>
<th>NG &amp; NS</th>
<th>G or S</th>
<th>NG &amp; NS</th>
<th>G or S</th>
<th>NO(_2) ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Average</strong></td>
<td>11.4</td>
<td>18.3(^b)</td>
<td>16.6</td>
<td>21.7(^b)</td>
<td>16.1</td>
<td>14.2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>S.D(^a)</strong></td>
<td>2.9</td>
<td>8.6</td>
<td>2.9</td>
<td>4.2</td>
<td>9.5</td>
<td>3.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Median</strong></td>
<td>10.8</td>
<td>15.0(^b)</td>
<td>16.2</td>
<td>21.1(^b)</td>
<td>12.9</td>
<td>14.9</td>
<td>0.74</td>
<td>0.86</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Maximum</strong></td>
<td>16.1</td>
<td>32.9</td>
<td>23.5</td>
<td>27.2</td>
<td>36.9</td>
<td>17.7</td>
<td>0.90</td>
<td>2.28</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Minimum</strong></td>
<td>7.3</td>
<td>11.1</td>
<td>13.6</td>
<td>17.4</td>
<td>8.0</td>
<td>9.5</td>
<td>0.38</td>
<td>0.77</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) S. D.: standard deviation, and \(^b\) significant difference (p < 0.05) between two group data.
### TABLE 8.3. A summary of the t-test correlation analysis (Log Concentration Data)

<table>
<thead>
<tr>
<th></th>
<th>All the houses</th>
<th>Houses without gas appliances</th>
</tr>
</thead>
<tbody>
<tr>
<td>Indoor NO₂ – house outdoor NO₂</td>
<td>14 0.231 0.428</td>
<td>12 0.499 0.098</td>
</tr>
<tr>
<td>Indoor NO₂ – QUT NO₂</td>
<td>12 0.359 0.252</td>
<td>12 0.393 0.232</td>
</tr>
<tr>
<td>Indoor NO₂ – EF NO₂</td>
<td>14 -0.213 0.465</td>
<td>12 0.203 0.527</td>
</tr>
<tr>
<td>Indoor NO₂ – indoor PN&lt;sup&gt;b&lt;/sup&gt;</td>
<td>13 0.796 <strong>0.001</strong></td>
<td>11 0.623 <strong>0.041</strong></td>
</tr>
<tr>
<td>Indoor NO₂ – indoor PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 0.120 0.684</td>
<td>12 0.056 0.864</td>
</tr>
<tr>
<td>House outdoor NO₂ – indoor PN</td>
<td>13 0.131 0.670</td>
<td>11 0.293 0.382</td>
</tr>
<tr>
<td>House outdoor NO₂ – indoor PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 0.712 <strong>0.004</strong></td>
<td>12 0.741 <strong>0.006</strong></td>
</tr>
<tr>
<td>House outdoor NO₂ – QUT NO₂</td>
<td>12 0.684 <strong>0.014</strong></td>
<td>11 0.690 <strong>0.019</strong></td>
</tr>
<tr>
<td>House outdoor NO₂ – EF NO₂</td>
<td>14 0.675 <strong>0.008</strong></td>
<td>12 0.695 <strong>0.012</strong></td>
</tr>
<tr>
<td>Indoor PN – indoor PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 0.164 0.576</td>
<td>12 0.114 0.725</td>
</tr>
<tr>
<td>Indoor PN – QUT PN</td>
<td>12 0.395 0.204</td>
<td>10 0.556 0.095</td>
</tr>
<tr>
<td>Indoor PM&lt;sub&gt;2.5&lt;/sub&gt; – QUT PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 0.116 0.693</td>
<td>12 0.159 0.621</td>
</tr>
<tr>
<td>Indoor PM&lt;sub&gt;2.5&lt;/sub&gt; – EF PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 0.363 0.201</td>
<td>12 0.385 0.217</td>
</tr>
<tr>
<td>QUT PM&lt;sub&gt;2.5&lt;/sub&gt; – EF PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>13 0.610 <strong>0.027</strong></td>
<td>11 0.628 <strong>0.038</strong></td>
</tr>
<tr>
<td>Non-activity indoor PN – indoor NO₂</td>
<td>13 0.449 0.123</td>
<td>11 0.725 <strong>0.012</strong></td>
</tr>
<tr>
<td>Non-activity indoor PN – non-activity Indoor PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 -0.152 0.603</td>
<td>12 -0.173 0.591</td>
</tr>
<tr>
<td>Non-activity indoor PN – QUT PN</td>
<td>12 0.315 0.319</td>
<td>10 0.321 0.367</td>
</tr>
<tr>
<td>Non-activity indoor PM&lt;sub&gt;2.5&lt;/sub&gt; – QUT</td>
<td>14 0.670 <strong>0.009</strong></td>
<td>12 0.675 <strong>0.016</strong></td>
</tr>
<tr>
<td>PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Non-activity indoor PM&lt;sub&gt;2.5&lt;/sub&gt; – EF PM&lt;sub&gt;2.5&lt;/sub&gt;</td>
<td>14 0.597 <strong>0.024</strong></td>
<td>12 0.602 <strong>0.038</strong></td>
</tr>
</tbody>
</table>

<sup>a</sup> N: paired sample number.  
<sup>b</sup> PN: submicrometer particle number concentration.  
<sup>c</sup> CR: correlation coefficient.  
<sup>d</sup> p: p value.
<table>
<thead>
<tr>
<th></th>
<th>DF&lt;sup&gt;a&lt;/sup&gt;</th>
<th>PM&lt;sub&gt;2.5&lt;/sub&gt; particle number</th>
<th>NO&lt;sub&gt;2&lt;/sub&gt;</th>
<th>non-activity PM&lt;sub&gt;2.5&lt;/sub&gt;</th>
<th>non-activity particle number</th>
</tr>
</thead>
<tbody>
<tr>
<td>Age</td>
<td>1</td>
<td>0.861</td>
<td>0.062</td>
<td>0.008</td>
<td>0.870</td>
</tr>
<tr>
<td>Height(D)</td>
<td>1</td>
<td>0.701</td>
<td>0.734</td>
<td>0.515</td>
<td>0.330</td>
</tr>
<tr>
<td>Walls</td>
<td>1</td>
<td>0.209</td>
<td>0.980</td>
<td>0.743</td>
<td>0.381</td>
</tr>
<tr>
<td>Smoke</td>
<td>1</td>
<td>0.604</td>
<td>0.472</td>
<td>0.552</td>
<td>0.972</td>
</tr>
<tr>
<td>Stove</td>
<td>1</td>
<td>0.680</td>
<td>0.040</td>
<td>0.030</td>
<td>0.918</td>
</tr>
<tr>
<td>Heating</td>
<td>2</td>
<td>0.532</td>
<td>0.069</td>
<td>0.017</td>
<td>0.314</td>
</tr>
<tr>
<td>Water heater</td>
<td>2</td>
<td>0.889</td>
<td>0.035</td>
<td>0.007</td>
<td>0.653</td>
</tr>
<tr>
<td>ExtractorFan</td>
<td>2</td>
<td>0.626</td>
<td>0.730</td>
<td>0.537</td>
<td>0.756</td>
</tr>
<tr>
<td>People</td>
<td>1</td>
<td>0.232</td>
<td>0.425</td>
<td>0.244</td>
<td>0.444</td>
</tr>
<tr>
<td>Car</td>
<td>2</td>
<td>0.527</td>
<td>0.020</td>
<td>0.126</td>
<td>0.552</td>
</tr>
<tr>
<td>Floor</td>
<td>1</td>
<td>0.436</td>
<td>0.388</td>
<td>0.042</td>
<td>0.352</td>
</tr>
<tr>
<td>Garage</td>
<td>1</td>
<td>0.072</td>
<td>0.205</td>
<td>0.125</td>
<td>0.159</td>
</tr>
<tr>
<td>Distance R&lt;sup&gt;b&lt;/sup&gt;</td>
<td>1</td>
<td>0.593</td>
<td>0.447</td>
<td>0.507</td>
<td>0.296</td>
</tr>
<tr>
<td>Pets</td>
<td>1</td>
<td>0.581</td>
<td>0.660</td>
<td>0.560</td>
<td>0.500</td>
</tr>
<tr>
<td>Distance P&lt;sup&gt;c&lt;/sup&gt;</td>
<td>1</td>
<td>0.241</td>
<td>0.290</td>
<td>0.384</td>
<td>0.563</td>
</tr>
<tr>
<td>Window</td>
<td>2</td>
<td>0.393</td>
<td>0.122</td>
<td>0.415</td>
<td>0.096</td>
</tr>
<tr>
<td>Bedroom</td>
<td>2</td>
<td>0.409</td>
<td>0.963</td>
<td>0.895</td>
<td>0.687</td>
</tr>
<tr>
<td>Paintwork</td>
<td>1</td>
<td>0.510</td>
<td>0.020</td>
<td>0.061</td>
<td>0.880</td>
</tr>
</tbody>
</table>

<sup>a</sup> DF: degrees of freedom. <sup>b</sup> Distance R: distance from the road. <sup>c</sup> Distance P: distance from the park.
Figure 8.1. Map presenting the locations of sampling site (Tingalpa) and two ambient monitoring station sites (QUT and Eagle Farm)

Figure 8.2. 48 hour average indoor and outdoor NO₂ concentration levels in 14 monitored houses.
Knowledge of particle sources, sinks, sizes, concentrations, phases and compositions in indoor air is important because of the potential health effects they cause. The behaviour of indoor particles is affected by many factors including indoor and outdoor sources, microenvironmental conditions, house characteristics as well as human lifestyle. Improving the understanding of indoor particle sources and sinks, and their multiple correlations and associations is critical for more accurate human exposure assessment and indoor air quality management.

This study was focussed on an investigation of particles, in particular in the submicrometer and ultrafine size ranges and their characteristics in residential houses. This led to the investigation of the multiple correlations between indoor and outdoor particles, between particles and NO$_2$, and between indoor air pollutants and house characteristics.

### 9.1 PRINCIPAL SIGNIFICANCE OF FINDINGS

In general, people spend the majority of their time indoors, and consequently, most of their exposure to particles occurs indoors. There are a number of different indoor environments; however, the residential house is the main one in which people, especially aged people and young children, spend their time. The study described in this thesis significantly advances the understanding of aerosol particle characteristics and dynamics, as well as important factors such as source emission rates and deposition rates in residential houses. While this study was conducted in Brisbane,
Australia, it is believed that the trends identified are considered to hold for other geographic settings as well. The main findings and conclusions of this study may be summarised as follows:

1. One of objectives of this study was to investigate the contribution of outdoor sources to indoor concentrations. In the absence of indoor particle sources, outdoor particle concentration is the main factor affecting indoor particle concentration. The strength of the effect of outdoor concentration on indoor particles is influenced by ventilation conditions or the air exchange rate (AER). For normal ventilation conditions (AER > 2 h⁻¹), outdoor particle concentrations can be used to predict instantaneous indoor particle concentrations, but for minimum ventilation (AER < 1 h⁻¹), the “delay constant” should be considered.

2. In the presence of indoor particle sources, however, indoor particle characteristics can be significantly affected by indoor particle sources. In this study, significant short-term variations and clear diurnal variations in particle number concentrations and approximation of PM₂.₅ concentrations were found in all the houses tested. The average number and mass concentrations during indoor activities were (18.2±3.9)×10³ particles cm⁻³ and (15.5±7.9) μg m⁻³ respectively, and under non-activity conditions, (12.4±2.7)×10³ particles cm⁻³ and (11.1±2.6) μg m⁻³, respectively. The pattern of diurnal variations varied from house to house; however, there was always a close relation between the variations and human indoor activities. Thus, human lifestyle can significantly affect indoor particle characteristics. It is, thus, possible to reduce particle exposures by reducing indoor particle emission and/or changing human lifestyle.
3. In general, there was a poor correlation between mass and number concentration. The correlation coefficients were highly variable from day to day, and from house to house. This implies that conclusions cannot be drawn about either one of the number or mass concentration characteristics of indoor particles based on a measurement of the other. This study also found that due to the significant differences between the indoor and outdoor diurnal variations it would be very difficult to accurately estimate indoor particle mass or number concentrations by using the outdoor concentrations solely.

4. One of the most important achievements of this study is the quantification of the effect of indoor sources on indoor particle concentration levels and emission rates from twenty types of indoor sources or activities. Such data in relation to particle number emissions have so far been largely unavailable. The study showed that indoor activities do affect indoor particle concentration levels, with the degree of effect dependent on the type of source and on house characteristics.

5. Among the indoor activities recorded in this study, the following were shown to elevate the indoor particle number concentrations in the range from 1.5 to over 27 times: cooking, frying, grilling, stove use, toasting, making pizza, smoking, candle vaporising eucalyptus oil and fan heater use. The indoor approximation of PM$_{2.5}$ concentrations showed an increase over the background concentration by 3, 30 and 90 times during smoking, grilling and frying respectively. The results of the cooking tests give an insight into the characterisations of cooking as a source of particles. They indicate that even though the same procedure of cooking was carried out, and the same cooking material was used, the emission
characterisations (emission rate and number median diameter) varied from house to house.

6. One of the most important parameters of indoor particle characteristics – the particle deposition rate in houses, has been determined. The results confirmed that the deposition rates are particle size specific and found the lowest deposition rates occurring in the particle size range 0.2-0.3 µm. The study also found that the measured deposition rate varied from house to house and showed that the average deposition rates under normal ventilation conditions were higher than those under minimum ventilation conditions for all particle sizes studied. However, the results of statistical analysis indicated that ventilation conditions could significantly affect particle deposition rates for particles in the size range from 0.08 µm to 1.0 µm, but not for particles of sizes smaller than 0.08 µm or larger than 1.0 µm. Since the levels of particle deposition rates found in this study were higher than the levels of air exchange rates, the contribution of particle deposition rates to total particle remove rates is not negligible and can be more important than AER in residential houses.

7. The multiple parameter analysis conducted in this study gave new insight into the correlation of indoor particles with other factors. This includes:

- There is no significant correlation between indoor and house outdoor NO$_2$ concentrations, nor between indoor and the two central monitoring station NO$_2$ concentrations.
- However, there are significant correlations between house outdoor and the two central monitoring station NO$_2$ concentrations, as well as between house outdoor NO$_2$ and indoor PM$_{2.5}$ concentrations.
• There is also a significant correlation between indoor NO₂ and indoor average submicrometer particle number concentrations, but there is no significant correlation between indoor NO₂ and non-activity indoor average submicrometer particle number concentrations.

• For particle number, no significant correlation between indoor and QUT submicrometer particle number concentration was found. Even under non-activity situations, the correlations between indoor and QUT submicrometer particle number concentrations are not significant.

• For PM₂.₅, there is no significant correlation between indoor concentrations and the two fixed site monitoring stations, but there is significant correlation between indoor non-activity and the two central monitoring stations concentrations.

• There are no significant associations between indoor PM₂.₅ and any of the listed house characteristics.

• However, associations between indoor submicrometer particle number concentration and several house characteristics including: stove type, water heater type, number of cars and paintwork conditions, are significant.

• Associations between indoor NO₂ and some house characteristics including: house age, stove type, heating system, water heater system and floor type are also significant.

• There are no significant associations between indoor non-activity PM₂.₅ or particle number concentrations, and any of the listed house characteristics.
Several conclusions can be derived from the above analysis. First of all, lack of correlation between indoor and outdoor concentrations of NO₂ (neither house nor central monitoring stations) and particles (number and PM₂.₅ mass) suggests that indoor activities significantly affect the indoor concentration of these pollutants. Therefore data from outdoor or central monitoring stations are not representative of the concentrations of these pollutants indoors. However, the presence of a correlation between house outdoor and the central monitoring stations NO₂ concentrations, suggest that data from these stations are representative of the trends in the concentrations (but not necessarily absolute concentrations) in residential locations. The correlation between PM₂.₅ concentrations indoors during non-activity periods and those measured at the central monitoring stations suggests that in the absence of human activities, outdoor air is the main contributor to the indoor PM₂.₅ concentrations. The correlation between indoor NO₂ and particle number concentrations suggests the same source for both these pollutants. However, the lack of correlation between indoor particle number and PM₂.₅ suggests they are from different sources. Non correlation of submicrometre particles and PM₂.₅ also suggests that there is an additional source of larger particles which dominates PM₂.₅. Therefore no conclusions can be made about one of these characteristics (number or mass concentration), based on a measurement of the other. These results also suggest that gas stoves, gas heating systems and gas water heater systems are the main indoor sources of indoor submicrometer particle and NO₂ concentrations.
In summary, this study experimentally investigated the characteristics of particle sources, sinks, sizes, concentrations and their variations, as well as the factors which effect particle sources and sinks in the indoor residential environment. This study also statistically analysed the multiple correlations and associations between air pollutants, as well as indoor air pollutants and the relative factors. The outcomes of this study are beneficial in improving the understating of indoor aerosol particle behaviour and their characteristics, as well as the factors influencing indoor aerosol particles.

9.2 FUTURE DIRECTION

This study was based on a relatively small number of houses, in a relatively short period (48 hrs), in the winter time. The high standard deviations of the measured data indicate the complexities of the measurement of indoor particles and the difficulties in investigating control factors. To improve the accuracy of the results of this study, more houses, a longer period (one or two weeks) and summer season measurements are suggested. Since different measurement methods were employed in this study for measurements conducted at the central monitoring stations and in the houses, systematic differences may exist between the results. Therefore, further study on indoor and outdoor particles using identical methods and equipment is also suggested. Additionally, a better understanding of air exchange rate will improve the accuracy of the calculated emission and deposition rates.

Throughout this thesis, correlations and other measures of association are computed and interpreted as suggestive of physical pathways between variables of interest. It is acknowledged that a correlation coefficient only measures the degree of linear
association between two variables, not a causal relationship. Further studies are required to confirm the suggested inferences.

The size and shape of the particles, as well as their chemical composition and properties determine the depth of inhalation, the extent of exhalation and the deposition rate in the human airways. Thus the improvement in understanding of indoor particle morphological and chemical properties is also very important. Hence further study on particle morphological and chemical properties is also strongly suggested.

Finally, since the characteristics of indoor particles can also be indoor microenvironment specific, collecting data from other main indoor microenvironments, such as office and school, is also important for future exposure assessments and epidemiological studies.