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He, C and Morawska, L and Gilbert, DL (2005) Particle deposition rates in residential houses. *Atmospheric Environment* 39(21):pp. 3891-3899.

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Particle deposition rates in residential houses

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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 μ 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 μ 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 µm, but not for particles smaller than 0.08 µm or larger than 1.0 µm. 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}

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, is 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.

2. Experimental Method

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.

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 μ m and up to the order of 10² or 10³ 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), which was set to measure particle size distribution and number concentration, in the range from 0.015 to 0.685 μ m; and 3) the TSI Model 3022A Condensation Nucleus Counter (CPC).

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.

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 was 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 (50°C 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'.

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 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 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 was used for estimation of particle deposition rates in conjunction with AER.

2.1. 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, α , can be calculated from the following equation (Nantka, 1990):

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

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.

2.2. 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):

$$\frac{dC_{in}}{dt} = P\alpha C_{out} + \frac{Q_s}{V} - (\alpha + \kappa)C_{in}$$
(2)

where C_{in} and C_{out} are the indoor and outdoor particle concentrations, respectively; *P* is the penetration efficiency; α 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:

$$\frac{dC_{in}}{dt} = P\alpha C_{out} - (\alpha + \kappa)C_{in}$$
(3)

In order to estimate the particle deposition rates (k), Equation 3 is simplified by assuming that α 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 then one and is a function of particle size (Abt et al., 2000; Long et al., 2001). It was considered that the uncertainly 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 ($C_{in0} >> C_{out}$), 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:

$$\ln(\frac{C_{in}}{C_{in0}}) = -(\alpha + \kappa)t \tag{4}$$

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 α from the slope. Several previous studies discussed the use of this equation for determination of particle deposition rate (Abt et al., 2000; Vette et al., 2001; Howard-Reed et al., 2003).

2.3. Data processing and analysis

Measured particle characteristics were expressed as number concentrations (particle cm⁻³) and volume concentrations (μ m³ 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 μ m; for each house under both ventilation conditions. Data analysis showed that particle concentration levels for larger particles (> 6 μ m) were very low and thus these particular results were associated with very large errors. Therefore this data was 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 Corp.). A level of significance of p = 0.05 was used for all statistical procedures.

3. Results and Discussion

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 1 and 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 (R²) for particles larger than 6 µm was less than 4, deposition rates for these particles were not considered in the further analysis.

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 3 and 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 μ 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. 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 3 and 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 µm to 1.0 µm, but not for particles of sizes smaller than 0.08 µm or larger than 1.0 µm. 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 µm to 1.0 µm.

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 remove rates could not be negligible and may be more important than AER in residential houses.

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 (recalculated from particle numbers assuming particle sphericity), 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 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 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. However, it should be noted that the count median diameter (CMD) of submicrometer particles increased by 50% in three cases during the measurements. This indicates that in these specific cases, coagulation played a more important role.

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 µm. 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 in residential houses were selected. A summary of the experimental conditions of the studies (including this study) is presented in Table 1, while Figure 5 presents a comparison of the size

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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.

Several conclusions can be derived from inspection of the data presented in Figure 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 Wallance et al. 2004a, and Wallance 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_{out}/C_{in} , unlike in other studies where regression of C_{in}/C_{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.

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.

Acknowledgments

This project was funded by the Built Environment Research Unit, Queensland Department of Public Works, and the Australian Research Council, through SPIRT grant No. C69804416. The assistance of Keith Eigeland, Chris Greenaway and Gillian

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Isoardi is gratefully acknowledged. Members of the QUT ILAQH, in particular, Jane Hitchins-Loveday and 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.

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

Table 1. A summary of the experimental conditions of the residential house studies on particle deposition rates



Figure 1. An example of the relationship between $ln(C_{in}/C_{in0})$ and time for 12 submicrometer particle size intervals.



Figure 2. . An example of the relationships between $ln(C_{in}/C_{in0})$ and time for 6 supermicrometer particle size intervals.



Figure 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 ($R^2 = 0.33$)



Figure 4. The average of particle deposition rates for the 18 particle size intervals under minimum ventilation conditions (Error bars represent one standard deviation). The polynomial fit line with the correlation coefficient ($R^2 = 0.84$)



Figure 5. A comparison of particle deposition rates measured in real houses reported in literature and determined in this study (for minimum ventilation).