

QUT Digital Repository:
<http://eprints.qut.edu.au/>



This is the accepted version of the following journal article:

[He, Congrong](#), [Morawska, Lidia](#), [Hitchins, Jane](#), & [Gilbert, Dale](#) (2004) Contribution from indoor sources to particle number and mass concentrations in residential houses. *Atmospheric Environment*, 38(21), pp. 3405-3415.

© Copyright 2004 Elsevier

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

Congrong He¹, Lidia Morawska¹, Jane Hitchins¹, 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

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 14 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 by 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 by 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, 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.

Keywords: indoor air quality, indoor emission sources, ultrafine particles, emission rates, submicrometer particles,

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 (eg 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 (eg 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 (eg 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 (eg James et al, 1991), and also contain higher levels of trace elements and toxins such as the polycyclic aromatic hydrocarbons and mutagens (eg 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 (eg 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

2. EXPERIMENTAL METHOD

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 ie. newer and older houses, brick and timber, high set and low set, was chosen as the measurement site. Thirteen houses in the suburb were chosen for the study and one additional house was chosen from another suburb as a comparison site.

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 10^2 or 10^3 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 ($\text{PM}_{2.5}$) 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$, 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).

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 DusTrak 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.

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

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

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.

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 (eg 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):

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

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. A number of previous studies discussed the use of this equation for

determination of source emissions and prediction of indoor air pollutant concentration levels (eg 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 (eg 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:

$$\overline{Q_s} = V \times \left[\frac{C_{int} - C_{in0}}{\Delta T} + (\overline{\alpha + \kappa}) \times \overline{C_{in}} - \alpha \times C_{in0} \right] \quad (3)$$

where ($\overline{Q_s}$) 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; $\overline{\alpha + \kappa}$ is the average total removal rate; ΔT 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 (eg 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 $\overline{\alpha + \kappa}$ can be obtained. The average air exchange rate for each house was determined experimentally for two different ventilation conditions through CO₂ 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).

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.

3. RESULTS AND DISCUSSION

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 1 presents a list of the types of activities 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 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.

3.1.1 *PM_{2.5} concentrations and emission rates*

While there are large errors associated with the concentration levels presented in Table 1 for all the activities, it can be seen that some activities result in particularly high indoor 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 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 during grilling, frying and smoking, respectively.

As could be expected from the above discussion, the estimated 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 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 vaporising result in particle emissions.

Data presented in Table 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.

3.1.2. Particle number concentrations and emission rates

The ratios of peak to background values for submicrometer particle number concentrations presented in Table 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 7.4×10^3 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 1 is, that indoor particle number concentration levels were elevated by about 180% due to opening the outside door.

Table 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 2.83×10^8 particle minute^{-1} , which is much less than found in this study (970×10^8 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.

3.1.3. Discussion

A number of conclusions can be drawn from analysis of the results presented in Table 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ concentration during vacuuming increased from 15 to 31 $\mu\text{g m}^{-3}$, 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 $\text{PM}_{2.5}$ concentration did not increase significantly (from 13 to 14 ($\mu\text{g m}^{-3}$)), 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 (eg 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 $90 \mu\text{g m}^{-3}$ 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 $25 \mu\text{g m}^{-3}$, 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.

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 Figure 1 and Figure 2; summary of particle emission rates and particle median diameter is presented in Table 2.

Figure 1 and Figure 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 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 $\text{PM}_{2.5}$, 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 $\text{PM}_{2.5}$ 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. Figure 3 and Figure 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 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 3).

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 $1.65 \sim 1.77 \times 10^5$ 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).

4. CONCLUSIONS

In this study particle number and mass concentration levels were measured in 14 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 $\text{PM}_{2.5}$ 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 vaporising 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 $\text{PM}_{2.5}$ 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.

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 Ray Duplock, Keith Eigeland and Chris Greenaway is gratefully acknowledged. Members of the ILAQH, QUT, in particular,

Milan Jamriska and Steve Thomas, are appreciated for their discussions and assistance with this study. The authors would like to express their special gratitude 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.

REFERENCES

- Abt E., Suh H.H., Allen G. Koutrakis P. 2000. Characterization of indoor particle sources: A study conducted in the metropolitan Boston area. *Environmental Health Perspectives*. 108(1), 35-44.
- Abt. E., Suh, H.H., Catalano, P., Koutrakis, P. 2000a. Relative contribution of outdoor and indoor particle sources to indoor concentrations. *Environmental Science & Technology* 34(17), 3579-3587.
- Brauer, M., Hirtle, R., Lang, B., Ott, W., 2000. Assessment of indoor fine aerosol contributions from environmental tobacco smoke and cooking with a portable nephelometer. *Journal of Exposure Analysis & Environmental Epidemiology*. 10(2), 136-144.
- Chen, Y.C., Zhang, Y.H., Barber, E.M., 2000. A dynamic method to estimate indoor dust sink and source. *Building & Environment*. 35(3), 215-221.
- Dockery, D., Spengler, J., 1981. Indoor-outdoor relationships of respirable sulfates and particles. *Atmospheric Environment*, 15, 335-343.
- Fine, P.M., Cass, G.R., Simoneit, B.R.T., 1999. Characterization of fine particle emissions from burning church candles. *Environmental Science & Technology*. 33(14), 2352-2362.

- Helsper, C., Mölter, W., Löffler, F., Wadenpohl, C., Kaufmann, S., Wenninger, G., 1993. Investigations of a new aerosol generator for the production of carbon aggregate particles, *Atmospheric Environment* 27 (A): 1271-1275.
- James, A.C., Stahlhofen, W., Rudolf, G., Egan, M.J., Nixon, W., Gehr, P., Briant, J.K., 1991. The respiratory tract deposition model proposed by the ICRP task group. *Radiation Protection Dosimetry*. 38, 159-165.
- Kamens, R., Lee, C.T., Weiner, R., Leith, D., 1991. A study to characterize indoor particles in three non-smoking homes. *Atmospheric Research*. 25A(5/6), 939-948.
- Kiss, G., Vargapuchony, Z., Rohrbacher, G., Hlavay, J., 1998. Distribution of polycyclic aromatic hydrocarbons on atmospheric aerosol particles of different sizes. *Atmospheric Research*. 46, (3-4 Special Issue SI), 253-261.
- Klepeis, N.E., Ott, W.R., Repace, J.L., 1999. The effect of cigar smoking on indoor levels and particles. *Journal of Exposure Analysis & Environmental Epidemiology*. 9(6), 622-635.
- Koutrakis, P., Briggs, S., Leaderer, B., 1992. Source apportionment of indoor aerosols in Suffolk and Onondaga Counties, New York. *Environmental Science & Technology*. 26, 521-527.
- Lefcoe, N.M., Inculet, I.I., 1975. Particulates in domestic premises II. Ambient levels and indoor-outdoor relationship. *Archives of Environmental Health*, 30, 565-570.
- Li, C.S., Lin, W.H., Jenq, F.T., 1993. Size distributions of submicrometer aerosols from cooking. *Environment International*. 19, 147-154.

- Lioy, P.J., Wainman, T., Zhang, J.F., Goldsmith, S., 1999. Typical household vacuum cleaners: The collection efficiency and emissions characteristics for fine particles. *Journal of the Air & Waste Management Association*. 49(2), 200-206.
- Long, C.M., Suh, H.H., Catalano, P.J., Koutrakis, P., 2001. Using time- and size-resolved particulate data to quantify indoor penetration and deposition behavior. *Environmental Science & Technology* 35(10), 2089-2099.
- Long, C.M., Suh, H.H. Koutrakis, P., 2000. Characterization of indoor particle sources using continuous mass and size monitors. *Journal of the Air & Waste Management Association*. 50(7), 1236-1250.
- Monn, Ch., Fuchs, A., Kogelschatz, D., Wanner, H.U., 1995. Comparison of indoor and outdoor concentrations of PM-10 and PM-2.5. *Journal of Aerosol Science*, 26, S515-S516
- Morawska, L., Thomas, S., Gilbert, D., Greenaway, C., Rijinders, E., 1999. A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road. *Atmospheric Environment* 33, 1261-1274
- Morawska, L., Zhang, J., 2002. Combustion sources of particles: Health relevance and source signatures. *Chemosphere*. 49(9), 1045-1058.
- Morawska, L., He, C., Hitchins, J., Gilbert, D., Parappukkaran, S., 2001. The relationship between indoor and outdoor airborne particles in the residential environment. *Atmospheric Environment*. 35, 3463-3473.
- Nantka, M., 1990. Comparison of Different Methods for Airtightness and Air Change Rate Determination. *Air Change Rate and Airtightness in Buildings, ASTM STP 1067, M.H. Sherman, Ed, American Society for Testing and Materials, Philadelphia, 267-282.*

- Ott, W.R., 1999. Mathematical models for predicting indoor air quality from smoking activity. *Environmental Health Perspectives*. 107(Suppl 2), 375-381.
- Özkaynak, H., Xue, J., Spengler, J., Wallace, L., Pellizzari, E., Jenkins, P., 1996. Personal Exposure to Airborne Particles and Metals - Results From The Particle Team Study In Riverside, California. *Journal of Exposure Analysis & Environmental Epidemiology*. 6(1), 57-78.
- Penttinen, P., Timonen, K.L., Tiittanen, P., Mirme, A., Ruuskanen, J., Pekkanen, J., 2001. Ultrafine particles in urban air and respiratory health among adult asthmatics. *European Respiratory Journal*, 17 (3), 428-435
- Rohr, A.C., Weschler, C.J., Koutrakis, P., Spengler, J.D., 2003. Generation and quantification of ultrafine particles through terpene/ozone reaction in a chamber setting. *Aerosol Science And Technology*, 37 (1), 65-78.
- Stephenson, D., Seshadri, G., Veranth, J.M., 2003. Workplace exposure to submicron particle mass and number concentrations from manual arc welding of carbon steel. *AIHA JOURNAL*, 64 (4), 516-521.
- Stoute, J.R.D, Groen, G.C.H., De Groot T.J.H., 1984. Characterization of indoor atmospheres. *Radiation protection dosimetry*. 7, 159-163.
- Strong, J.C., 1988. The size of attached and unattached radon daughters in room air. *Journal of Aerosol Sciences*. 19: 1327-1330.
- Thatcher, T.L., Layton, D.W., 1995. Deposition, resuspension, and penetration of particles within a residence. *Atmospheric Environment*, 29(13), 1487-1497.
- Tu, K.W., Knutson, E.O., 1988. Indoor outdoor aerosol measurements for two residential buildings in New Jersey. *Aerosol Science and Technology*. 9, 71-82.
- Tucker, W.G., 2000. An overview of PM_{2.5} sources and control strategies. *Fuel Processing Technology*. 65, 379-392.

Tuckett, C.J., Holmes, P., Harrison, P.T., 1998. Airborne particles in the home.

Journal of Aerosol Science, 29. Suppl. 1, S293-S294.

Wallace, L., 1996. Indoor particles: A review. *Journal of the Air & Waste*

Management Association. 46, 98-126.

Table 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.

Activity	N	Peak values (ug.m ⁻³)		Ratio (mass)		Emission rate (mg min ⁻¹)		Peak values (particle. cm ⁻³ , ×10 ³)		Ratio (number)		Emission rate (particle min ⁻¹ × 10 ¹¹)	
		Median	S.D	Median	S.D.	Median	S.D.	Median	S.D.	Median	S.D.	Median	S.D.
Food preparation													
Cooking	24	37	194	2.89	12.6	0.11	0.99	126	177	10.3	19.3	5.67	8.61
Cooking Pizza*	1	735		73.5		1.59		137.3		9.81		1.65	
Frying	4	745	352	33.6	28.3	2.68	2.18	154	21.3	10.0	6.1	4.75	2.45
Grilling	6	718	3427	90.1	312	2.78	17.8	161	69.9	8.69	5.27	7.34	5.06
Kettle	25	13	20	1.13	0.67	0.03	0.31	15.6	14.0	1.08	0.6	0.35	1.76
Microwave	18	16	18	1.12	0.42	0.03	0.11	16.3	28.6	1.12	1.55	0.55	1.94
Oven	6	24	6	1.76	0.53	0.03	0.03	61.5	31.9	2.96	0.78	1.27	2.10
Stove	4	57	264	2.4	19.7	0.24	1.29	179	287	12.5	10.5	7.33	51.4
Toasting	18	35	32	2.08	8.31	0.11	0.37	114	160	6.34	7.44	6.75	16.7
Other activities (multiple events)													
Open door	9	21	9	1.23	0.37			22.0	14.6	2.89	1.21		
Smoking	6	79	29	4.03	1.76	0.99	0.81	26.6	13.6	1.54	0.96	1.91	1.92
Sweep floor	3	35	4	2.04	1.3	0.05	0.01	34.9	5.86	1.05	0.01	0.12	0.02
Vacuuming	5	16	8	1.46	0.32	0.07	0.04	41.3	17.6	1.51	1.17	0.97	1.57
Washing	17	18	12	1.25	0.57	0.04	0.04	30.9	18.5	1.30	0.83	0.96	2.60
Other activities (single event)													
CVE oil*	1	132		13.2		0.91		74.6		8.29		5.52	
Dusting*	1	22		1.69		0.09		14.1		1.00			
Fan*	1	20		1.67				11.0		1.00			
Fan Heater*	1	15		1.50		0.05		87.1		27.2		4.07	
Hair Dryer*	1	45		1.36		0.04		9.5		1.06		0.11	
Shower*	1	20		1.08		0.04		10.7		1.37		0.78	
Washing M*	1	43		2.05		0.12		11.1		1.18		0.15	

Note: N: sample number; CVE oil: Candle Vapour Eucalypt oil; Washing M: washing machine; *: no S.D.

Table 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 $\text{min}^{-1} \times 10^{12}$) and number median diameters (NMD, μ) (measured by the SMPS), supermicrometer particle emission rates (particle $\text{min}^{-1} \times 10^8$) and number median diameters (NMD, μ) (measured by the APS), as well as approximation of $\text{PM}_{2.5}$ emission rates (mg min^{-1}) (measured by DustTrak).

	Normal Ventilation Conditions					Minimum Ventilation Conditions				
	SMPS		APS		PM _{2.5}	SMPS		APS		PM _{2.5}
	ER	NMD	ER	NMD	ER	ER	NMD	ER	NMD	ER
Median	1.19	0.026	0.67	0.80	0.84	0.72	0.033	0.64	0.82	0.30
Average	1.36	0.031	3.06	0.80	1.46	0.78	0.033	0.90	0.80	0.72
S.D.	1.03	0.011	5.48	0.09	2.12	0.42	0.010	0.88	0.08	0.93
Min	0.20	0.022	0.12	0.64	0.04	0.13	0.022	0.08	0.63	0.04
Max	4.01	0.063	19.68	0.94	8.02	1.60	0.060	3.05	0.89	3.06

Min: Minimum; Max: Maximum; ER: emission rates.

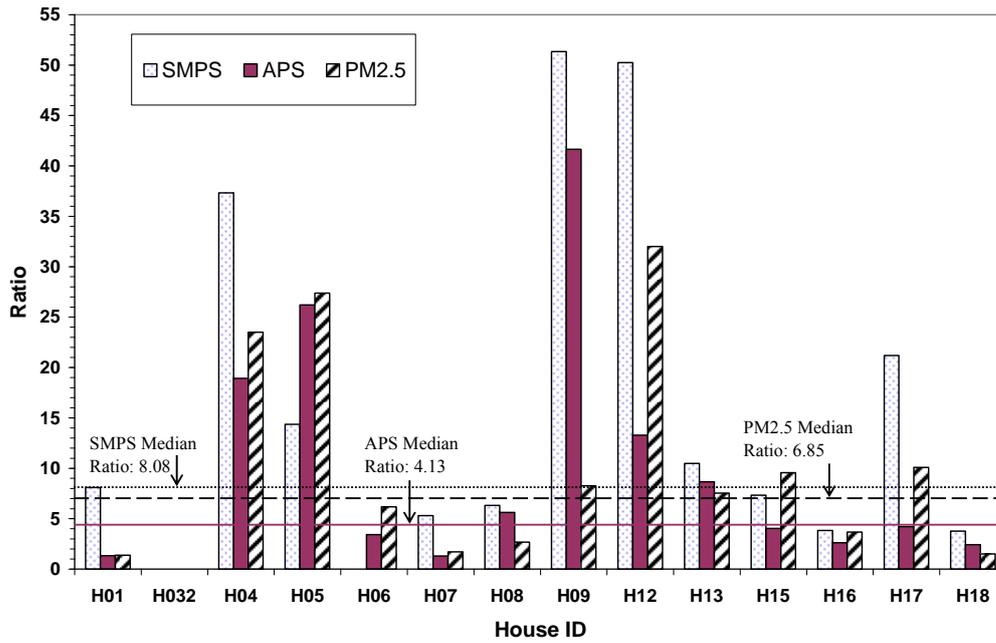


Figure 1. The ratios of peak to background values for submicrometer (SMPS) and supermicrometer (APS) particle number concentrations and PM_{2.5} concentrations during cooking test under **normal** ventilation conditions.

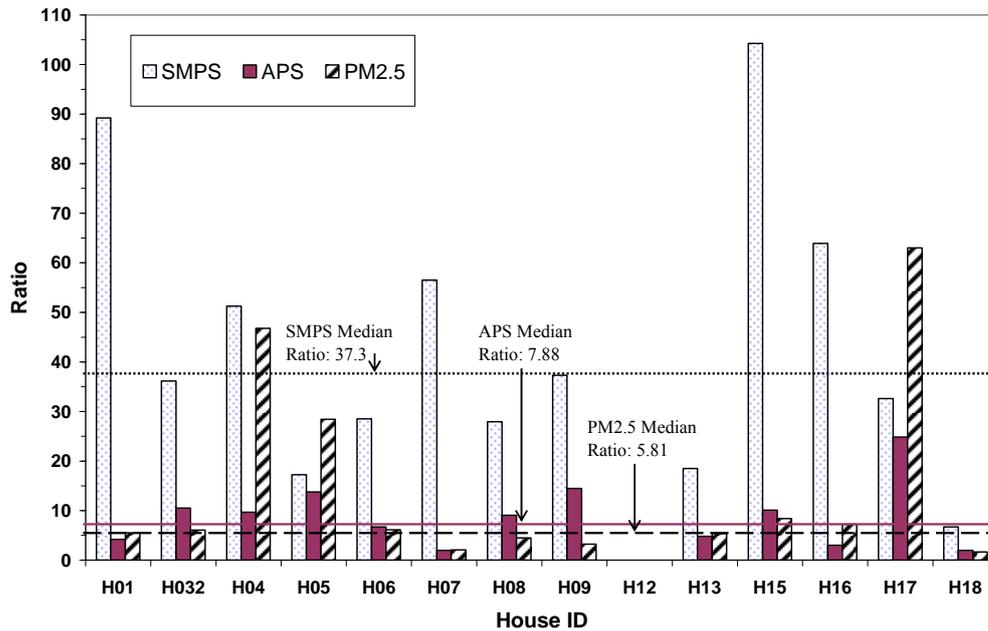


Figure 2. The ratios of peak to background values for submicrometer (SMPS) and supermicrometer (APS) particle number concentrations and PM_{2.5} concentrations during cooking test under **minimum** ventilation conditions.

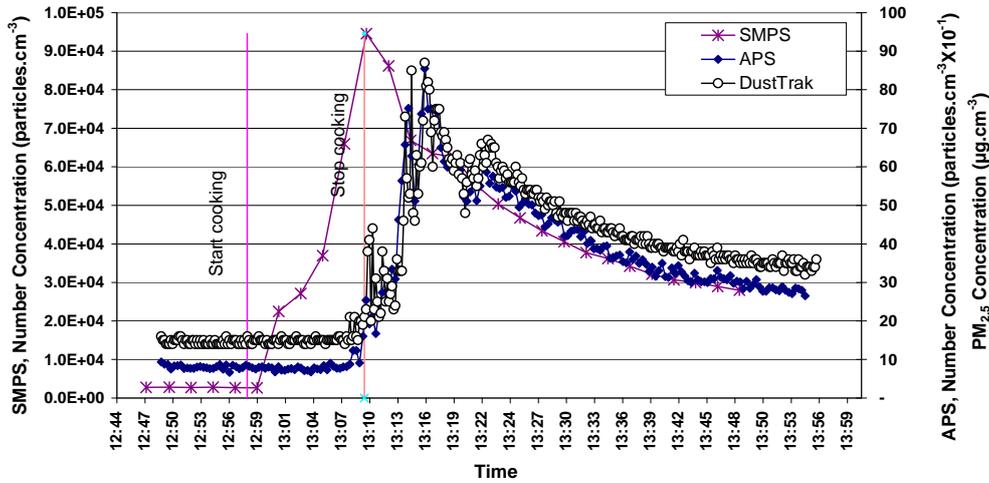


Figure 3. A typical example of changes in submicrometer (SMPS) and supermicrometer (APS) particle number concentration, as well as PM_{2.5} concentration with time during the cooking test under minimum ventilation condition (House32, 9/07/1999)

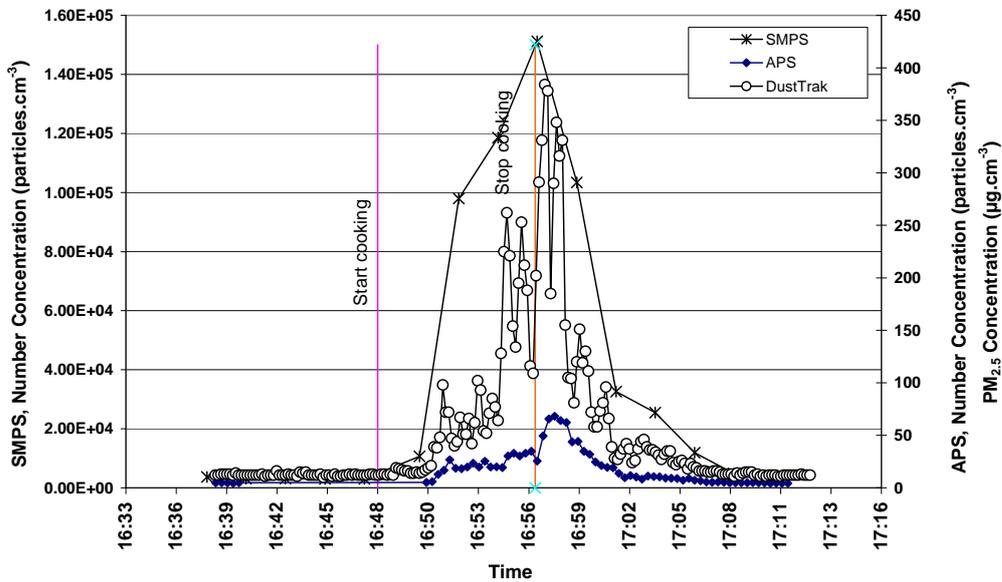


Figure 4. A typical example of submicrometer particles concentration (SMPS), supermicrometer particle concentration and PM_{2.5} concentration variations with time during the cooking test under **normal** ventilation condition (House12, 12/05/1999)