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Quantification of differences between occupancy and total monitoring periods for better assessment of exposure to particles in indoor environments



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HIGHLIGHTS

- Only the concentrations from occupancy periods should be used for the exposure assessment.
- Inclusion of non-occupancy data for exposure assessment underestimated the PM_{2.5} mass concentrations in residences and schools.
- PNC indoors varied to a great degree in residences and was influenced by indoor activities and site specificity.
- In an apartment, the median PNC was 58% higher during occupancy time than during total monitoring period.

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ABSTRACT

For the assessment of personal exposure, information about the concentration of pollutants when people are in given indoor environments (occupancy time) are of prime importance. However this kind of data frequently is not reported. The aim of this study was to assess differences in particle characteristics between occupancy time and the total monitoring period, with the latter being the most frequently used averaging time in the published data. Seven indoor environments were selected in Sweden and Finland: an apartment, two houses, two schools, a supermarket, and a restaurant. They were assessed for particle number and mass concentrations and number size distributions. The measurements using a Scanning Mobility Particle Sizer and two photometers were conducted for seven consecutive days during winter in each location. Particle concentrations in residences and schools were, as expected, the highest during occupancy time. In the apartment average and median PM_{2.5} mass concentrations during the occupancy time were 29% and 17% higher, respectively compared to total monitoring period. In both schools, the average and medium values of the PM_{2.5} mass concentrations were on average higher during teaching hours compared to the total monitoring period by 16% and 32%, respectively. When it comes to particle number concentrations (PNC), in the apartment during occupancy, the average and median values were 33% and 58% higher, respectively than during the total monitoring period. In both houses and schools the average and median PNC were similar for the occupancy and total monitoring periods. General conclusions on the basis of measurements in the limited number of indoor environments cannot be drawn. However the results confirm a strong dependence on type and frequency of indoor activities that generate particles and site specificity. The results also indicate that the exclusion of data series during non-occupancy periods can improve the estimates of particle concentrations and characteristics suitable for exposure assessment, which is crucial for estimating health effects in epidemiological and toxicological studies.

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1. Introduction

In numerous epidemiological studies, outdoor fine particulate matter PM_{2.5} (particles with diameter smaller than 2.5 µm) has been associated with cardiopulmonary diseases and increased mortality (Pope and Dockery, 2006). However, we spend the majority of our time indoors (about 90%) (Leech et al., 2002) where particles of both outdoor and indoor origin are found. Poor correlations have been found between outdoor PM_{2.5} and personal exposure, which initiated a debate whether the outdoor PM is a good surrogate for exposure to PM (Meng et al., 2005; Wilson and Brauer, 2006). This is because apart from outdoor PM sources, indoor, work related, in-vehicle sources and personal activities also contribute to personal exposure. Many indoor sources generate particles in amounts that exceed levels observed outdoors by far (He et al., 2004; Long et al., 2000). Recent studies show that about 60% of exposure to ultrafine particles (<100 nm) in residences comes from indoor sources (Isaxon et al., 2014; Bekö et al., 2013; Bhangar et al., 2011). Particles emitted from different sources have different physico-chemical characteristics. It may thus be that particles generated indoors have different health effects than those of outdoor origin. However epidemiological studies on population representative scale, so far have been exclusively based on outdoor particle characteristics. This is mainly due to lack of population representative data on indoor particle characteristics suitable for epidemiological studies.

Particles found indoors consist of: 1) outdoor particles that infiltrated indoors, 2) particles emitted indoors and 3) particles formed indoors through reactions of gas-phase precursors emitted both indoors and outdoors (Morawska and Salthammer, 2003). Concentration of particles in indoor environments are influenced by many factors such as building characteristics, geographical location, outdoor pollution, type of interiors, type of cooking appliances, human activities, hobbies, cleaning practices, and other practices (burning candles, incense etc.) which all vary to a great extent. Various indoor activities are known sources of fine particles such as tobacco smoking, cooking (frying, grilling, baking, etc.), use of gas and electric stoves, toasters, gas-powered clothes dryers, fireplaces, candle and incense burning, electronic cigarettes, decorative ethanol fireplaces, use of photocopiers, hair spray, cleaning products containing terpenes (which in presence of ozone form secondary aerosols) (Schrapp et al., 2014; Schober et al., 2013; Abt et al., 2000; Dennekamp et al., 2001; He et al., 2004; Hussein et al., 2006; Lee and Hsu, 2007; Long et al., 2000; Ogulei et al., 2006; Wainman et al., 2000; Wallace, 2006; Weschler, 2003). In general more information is available on outdoor particle characteristics while data on indoor particle characteristics remain limited.

Studies assessing particle concentrations indoors, outdoors and from personal monitoring have been reviewed by Morawska et al. (2013). In summary there are many studies based on integrated particle mass concentration measurements but studies investigating time-dependant changes in particle mass and number are limited. Averaging times in the existing studies vary to a great degree and comprise: hours (8, 24, 48 h), days, seasons, years, active sources, no sources and seldom occupancy (occupant present in a residence) or non-occupancy time (Morawska et al., 2013). The most frequently used averaging time is the total monitoring period. From the personal exposure perspective, information about particle concentrations when people are in given microenvironments are of prime importance, however this data is frequently not available.

Scattered information on particle number concentrations (PNC) and emission factors for indoor sources can be found in several publications, but the available data is far from being complete (Torkmahalleh et al., 2012; Buonanno et al., 2009; Ogulei et al.,

2006; Wallace, 2006; Dennekamp et al., 2001; Hussein et al., 2005; Morawska et al., 2003; Wallace et al., 2004).

The aim of the study was to assess differences in particle characteristics between occupancy and the total monitoring period, with the latter being the most frequently used averaging time in published studies on particle characteristics in indoor environments. Assessed particle characteristics comprise: number and mass concentrations as well as number size distributions. Additionally this study aims to identify indoor sources of particles and estimate their source strength to understand the difference between occupancy and the total monitoring period.

2. Materials and methods

2.1. Sampling sites

The sampling sites consisted of: an apartment, two houses (HA and HB), two schools (SA and SB), a supermarket, and a restaurant. The majority of the sampling sites are situated in southern Sweden, whereas the house HA is situated in southern Finland. Particle measurements were performed for at least seven consecutive days in each location. In Swedish locations measurements took place during the winter of 2006/2007, whereas the data from a Finnish house HA, were extracted from measurements conducted in February 2001. Table 1 summarises information about all sampling sites. Activities were identified using information from log books kept by occupants in the residences. In the schools, supermarket and restaurant, time schedules and interviews with employees were used to identify various activities.

2.1.1. Apartment

Apartment is situated on the third floor of a residential building, heated by district heating. Most of the time the windows were kept closed due to low outdoor temperature (daily mean between 0.5 and 3.5 °C). Two non-smoking, employed adults lived in the apartment during the measurements. On one occasion a dinner party took place with four invited guests. An electric stove was used for cooking. Even though an exhaust fan above the oven was installed, the occupants did not use it due to noise disturbance.

2.1.2. House HA

House HA is situated in a suburban area in Espoo, which is part of the Helsinki Metropolitan Area. It is a two-level detached house. The measurements were performed on the on a ground floor in a living room, while kitchen is on second floor where major activities took place. Two non-smoking adults and one child lived in the house. Due to low outdoor temperatures, windows were kept closed most of the time (daily mean outdoor temperatures between −22 and 2 °C). An electric stove was used for cooking. A boiler fired with wood logs was used for heating.

2.1.3. House HB

House HB is situated in a remote area in rural surroundings. It is a three-level detached house. The measurements were performed on the second floor close to a staircase. Two non-smoking adults lived in the house. Windows were also kept closed most of the time due to low outdoor temperatures (−5 to 1 °C). An electric stove was used for cooking. A boiler fired with wood logs was used for heating.

2.1.4. School SA

School SA is an elementary school. It is a one-level building, from the 1970s. The ventilation operates on weekdays between 5:00 and 18:00. Teaching hours on weekdays are between 8:00 and 16:00.

Table 1

Summary of sampling sites including: location, type of ventilation, filters used, measured air exchange rates (AER), sites' volume, sampling duration, and configuration of performed measurements indoor and outside.

| Sampling site | Location ^a | Type of ventilation and filters used | Occupancy ^b (%) | AER (h ⁻¹) | Site's volume (m ³) | Measurements | | | |
|---------------|-----------------------|--|-------------------------------|---------------------------|------------------------------------|--------------|----------------|----------------|--|
| | | | | | | Date | Number of days | Indoor | Simultaneous/alternate ^c measurements outside |
| Apartment | Malmö centre | Natural ^d | 70 | 0.3 ^e | ~280 | Feb '07 | 10 | SMPS, DustTrak | SMPS (monitoring station), DustTrak |
| House HA | Outskirts of Helsinki | Natural ^d | 85 | 0.75 | ~600 | Feb '01 | 7 | DMPS | DMPS ^c |
| House HB | Rural | Natural ^f | 78 | 0.12 ^e | ~320 | Feb '06 | 8 | SMPS | SMPS (monitoring station) |
| School SA | Outskirts of Lund | Mechanical, electrically charged synthetic F7 ^g | 33 | 2.4 ^h | ~900 | Oct '06 | 10 | SMPS, DustTrak | SMPS ^c |
| School SB | Outskirts of Lund | Natural | 33 | 0.4 ^h | ~5400 | Mar '07 | 11 | SMPS, DustTrak | SMPS ^c , DustTrak |
| Supermarket | Outskirts of Lund | Mechanical, glass fibre F7 ^g | 55 | 9.7 ⁱ | ~23 400 | Jan '07 | 7 | SMPS, DustTrak | SMPS ^{c,j} , DustTrak |
| Restaurant | Lund centre | Mechanical, glass fibre F7 ^g | 71 | 8.5 ⁱ | ~900 | Dec '06 | 7 | SMPS, DustTrak | SMPS ^{c,j} , DustTrak |

^a Lund (Sweden, 80 000 inhabitants), Malmö (Sweden, 240 000 inhabitants), Helsinki (610 000 inhabitants).

^b Percentage of occupancy time (people present at given location) out of total monitoring period.

^c Alternate measurements by SMPS system; when indicated data obtained from monitoring stations (see [Materials and Methods](#)). Simultaneous measurements by two DustTraks.

^d Natural ventilation with additional occupant controlled kitchen exhaust (kitchen hood).

^e Measured at low ventilation conditions, i.e. windows closed, doors between rooms opened.

^f Natural ventilation with additional occupant controlled kitchen and bathroom exhaust (both operate simultaneously upon switching).

^g Filter class according to EN 779: 2002.

^h Measured at low ventilation conditions, i.e. windows and doors closed.

ⁱ Measured during normal opening hours, doors opening not controlled.

^j Measured in supplied ventilation air.

2.1.5. School SB

School SB is a secondary school. It is a two-level building with natural ventilation, built in 1997. There are 8 classrooms. Teaching hours are on weekdays between 8:00 and 16:00.

2.1.6. Supermarket

The ventilation supplies heated and filtered air and operates on weekdays between 4:30 and 22:00 and on weekends between 6:30 and 20:30. Measurements were performed in the area with washing powders, cleaning products and air fresheners. The opening hours weekdays are 8:00–22:00, and on weekends 9:00–20:00.

2.1.7. Restaurant

Restaurant is a fast food restaurant. The ventilation operates 24 h a day, seven days a week. Measurements were performed in a dining area that seats ~100. The opening hours are: 9:00–24:00: Monday to Thursday and Sunday, 9:00–4:00 Friday and Saturday.

2.2. Instruments and measurements

Instruments, recording continuously, were placed in a specifically designed and insulated enclosure

2.2.1. SMPS

A Scanning Mobility Particle Sizer (SMPS 3934, TSI Inc., USA) consisting of a Differential Mobility Analyser (Long Column DMA, TSI Inc., USA) and Condensation Particle Counter (CPC 3010, TSI Inc., USA) forming. The SMPS was used in Swedish sites and measured particles between 15 and 700 nm, used sampling time 180 s. An automatic valve was used to switch between indoor and outdoor air, or between indoor and supplied ventilation air, see [Table 1](#). A correction for particle losses due to diffusion in the DMA and tubing ([Karlssohn and Martinsson, 2003](#)) was applied. As an example 45% of the 20 nm particles from ventilation/outdoor air were lost. The same procedure was used in the house HA in Finland using Differential Mobility Particle Sizer (DMPS). DMPS measured particle

number size distributions between 15 and 400 nm, and consisted of DMA (HAUKE 28.5 cm) and CPC (CPC 3010, TSI, Inc.).

2.2.2. DustTrak

A photometer (DustTrak 8520, TSI Inc., USA) was used for indirect mass concentration measurements of PM_{2.5} (down to ~0.1 µm), recorded values were one minute averages. In some locations a second DustTrak (the same model) was placed on the roof for simultaneous outdoor concentrations monitoring (see [Table 1](#)). The DustTrak's response is dependent on aerosol material (refractive index) and the size distribution and it was reported that it might significantly overestimate the true PM_{2.5} mass concentration ([Yanosky et al., 2002](#)). Laboratory comparison of DustTrak and Tapered Element Oscillating Microbalance (TEOM, 50 °C, model 1400a, R&P Inc.) of PM_{2.5} readings, using typical indoor sources such as candles, incense and frying onions were conducted (details in [Wierzbicka, 2008](#)). The derived and used for indoor data correction factor was 0.49 ($C_{PM_{2.5} TEOM}/C_{PM_{2.5} DustTrak}$). Outdoor DustTrak readings were compared to the TEOM by measuring at the monitoring station in Vavihill. The derived and used correction factor was 0.29. Nevertheless, all the DustTrak readings presented here, even after correction, remain only approximations of the PM_{2.5} mass concentration.

2.2.3. Air exchange rate (AER)

Air exchange rate (AER) was measured at each sampling site using a 1312 Photoacoustic Multi-gas Monitor together with a 1303 Multipoint Sampler and Doser (Innova AirTech Instruments). A tracer decay method was used with sulphur hexafluoride (SF₆) as the tracer gas.

2.2.4. Outdoor particle characteristics

Outdoor particle characteristics for the apartment were obtained from monitoring point at the roof top of the town hall in Malmö city centre ([Roldin et al., 2010](#)) situated about 2 km from the apartment, average values for winter 2006/2007 were used. For house HB outdoor concentrations were obtained from situated

nearby rural monitoring station in Vavihill (part of the European ACTRIS infrastructure network) (Kristensson et al., 2008).

2.3. Data presentation and analysis

PNC – denotes the particle number concentrations in the measured size range i.e. 15–700 nm, with the exception for house HA where the measured size range was 15–400 nm. The difference in measured size range should not affect the comparisons though, because PNC in the size range 400–700 nm is only a small fraction of particles smaller than 1000 nm. The data presented in this article were divided into various periods: *total monitoring period* or site average, defined as an average of the entire monitoring duration; *occupancy time*, defined as time when at least one person is present in given indoor environment, in case of schools the occupancy time reflects teaching hours and in the supermarket and restaurant, opening hours; *non-occupancy*, no one is present in given indoor environment; *active sources period*, time when an elevated PNC (a sharp increase) associated with indoor activity was observed until the concentration went down to average background (no active source) level.

2.4. Source strength estimations

Particle source strengths were calculated using the mass balance differential equation:

$$\frac{dC_{in}}{dt} = PaC_{out} + \frac{S}{V} - (a + k) C_{in} \quad (1)$$

where C_{in} and C_{out} are indoor and outdoor PNC (particles cm^{-3}), respectively; P is the penetration factor (h^{-1}); a is AER (h^{-1}); k is the particle deposition rate (h^{-1}); S is source strength (particles h^{-1}); and V is the volume of the apartment (m^3).

The simplified solution for S to the Equation (1) given by Wallace et al. (2004) was used:

$$S = \frac{V(a + k)\Delta C_{in}}{1 - e^{-(a+k)t}} \quad (2)$$

where the following assumptions were made: a) Equation (1) refers to a particular particle size, where P , k and S may all be functions of particle size, b) coagulation and condensation processes were neglected, c) constant outdoor concentration during the active indoor source episode and subsequent particle decay, d) constant values for P , a , k and S during the active indoor source time, e) apartment is a single well mixed zone with instantaneous mixing, f) the conditions, at the beginning of the active indoor source event, have held long enough before that to reach equilibrium.

Criteria for selecting an active indoor source episode for source strength calculations, adopted from Wallace et al. (2004) were: a) a sharp increase in PNC above average background (no active source) concentration level, b) no other reported activity at the time and a smooth decay (indicating no other significant active indoor source), c) a return to the initial (before the episode) concentration level indicating no change in outdoor concentration. These criteria limited active source episodes in the apartment, suitable for the source strength calculations to seven. For the ΔC_{in} calculation, the difference between peak concentration for the given active source episode and the initial background concentration were used. This determined the maximum concentration change and allowed estimation of average particle source strength for this period on the basis of Equation (2).

Decay rates were determined for given particle size intervals from the equation:

$$C_{(t)} = e^{-(a+k)t} C_{(t-1)} \quad (3)$$

where $C_{(t)}$ and $C_{(t-1)}$ are the indoor concentrations for given particle size interval at times t and $t-1$. Decay rates are determined by taking the natural logarithm of both sides of the Equation (3) and regressing over time. The negative slope of the regression is $a + k$, i.e. it is a total decay rate due to exfiltration and deposition.

3. Results and discussion

3.1. PNC during occupancy and total monitoring periods in residences

Characteristics of PNC in residences are summarised in Table 2. Compared values comprise total monitoring period, occupancy, non-occupancy time and outdoors. In the apartment the PNC during occupancy time is higher than during total monitoring, differences between these two averaging periods comprise 33% for average and 58% for median PNC. In the houses HA and HB total monitoring and occupancy are very similar, for both average and median PNC values. The observed differences between the apartment and both houses can be influenced by the following factors: a) in the apartment vigorous cooking activities took place without use of the kitchen extraction fan (due to noise disturbance) whereas in both houses cooking activities were limited and the extraction fans were used promptly; in case of house HA use of the kitchen extraction system automatically activated bathroom extraction system which further increased EAR and removal of the particles b) one level and open-space layout of the apartment (joined kitchen and living room) influenced the spread of the particles during cooking activities compared to enclosed kitchens in two- and three-level houses HA and HB, respectively; c) candle burning, known and significant contributor to particle loads indoors (Isaxon et al., 2014; Bekö et al., 2013) occurred only in the apartment. Recent studies, conducted in relatively large amount of residences in Denmark (56 residences, Bekö et al., 2013) and Sweden (22 residences, Isaxon et al., 2014) report higher concentrations of PNC during occupancy in comparison to total monitoring periods during winter, which agrees with results obtained in the apartment. Bekö et al. (2013) reports occupancy time average and median concentration 29% and 6% higher in comparison to total monitoring. Isaxon et al. (2014) reports occupancy average concentration 18% higher and median concentration 12% higher than total monitoring periods. Both studies (Isaxon et al., 2014; Bekö et al., 2013) identified candle burning and cooking activities as the main particle sources contributing to observed PNC. Lack of candle burning in houses HA and HB combined with less vigorous cooking activities compared to the apartment may explain why higher PNC is observed during occupancy time only in the apartment and not in houses HA and HB.

Non-occupancy time consistently is characterised with lower average and median values in comparison to occupancy periods in the residences. The greatest difference in PNC between occupancy and non-occupancy periods is seen in the apartment with non-occupancy average 3000 particles cm^{-3} in comparison to 12 000 particles cm^{-3} during occupancy time. Higher concentrations during occupancy in comparison to non-occupancy time are not surprising as occupants' presence and activities dictate occurrence of major indoor particle sources. PNC during total monitoring period in the apartment (in particles cm^{-3} : average 9000, STD 25 000, min 700 and max 183 000) are similar to the values reported by Ogulei et al. (2006) for particles between 10 and 450 nm, for a non-smoking townhouse in Boston as four months averages

Table 2

PNC (15–700 nm, SMPS measurements) characteristics for total monitoring period, occupancy, non-occupancy, and outdoors in studied residential indoor environments.

| | | Average | STD ^a | Median | Min | Max | Average number GMD ^b | Average GSD ^c |
|-----------|-----------------------|-------------------------------|------------------|--------|------|---------|---------------------------------|--------------------------|
| | | (particles cm ⁻³) | | | | | (nm) | |
| Apartment | Total monitoring | 9000 | 25 000 | 1200 | 700 | 246 000 | 82 | 2.1 |
| | Occupancy | 12 000 | 30 000 | 1900 | 700 | 246 000 | 84 | 2.0 |
| | Non-occupancy | 3000 | 3000 | 1900 | 700 | 28 000 | 79 | 2.1 |
| | Outdoors ^d | 3600 | 2500 | 3100 | 200 | 35 000 | 45 | 2.2 |
| House HA | Total monitoring | 3000 | 5200 | 2200 | 600 | 120 000 | 66 | 2.2 |
| | Occupancy | 3000 | 5500 | 2300 | 600 | 120 000 | 66 | 2.2 |
| | Non-occupancy | 2000 | 700 | 1900 | 1000 | 5100 | 62 | 2.3 |
| | Outdoors | 4100 | 4500 | 3900 | 1000 | 9800 | 60 | 2.2 |
| House HB | Total monitoring | 2500 | 2700 | 1600 | 260 | 39 000 | 71 | 1.9 |
| | Occupancy | 2600 | 2700 | 1500 | 260 | 39 000 | 72 | 1.9 |
| | Non-occupancy | 2300 | 1500 | 1900 | 500 | 7000 | 69 | 1.8 |
| | Outdoors ^e | 1500 | 600 | 1400 | 400 | 3300 | 82 | 2.5 |

^a Standard deviation.^b Geometric mean diameter.^c Geometric standard deviation.^d Average for winter 2006/2007 at the roof level in Malmö centre (Roldin et al., 2010).^e Average for house HB monitoring period from rural monitoring station in Vavihill (Kristensson et al., 2008), situated in the area of the house HB.

(in particles cm⁻³: average 8100, STD 17 250, min 180 and max 308 000), but higher in comparison to both houses.

Outdoor concentrations for the apartment and house HB, presented in Table 2, were lower than indoors during total monitoring periods and occupancy. Outdoor concentration for apartment and house HB were not measured directly outside the sites but taken from nearby monitoring stations (details in Materials and Methods). In house HA outdoor concentrations are slightly higher than during any other averaging period of indoor concentrations. This most probably can be explained by a few factors: a) lack of vigorous cooking activities with use of stove top or oven, as majority of cooking comprised food preparation with use of a microwave; b) measurements took place on a ground floor while kitchen is on second floor and major activities took place there; c) use of occupant controlled kitchen extraction system which automatically activated bathroom extraction system, thus increasing EAR and removal of the pollutants, d) in general higher AER (0.75 h⁻¹) in comparison to two other residences (below 0.3 h⁻¹) which enhances particle removal but also can contribute to increased particle deposition on different indoor surfaces e) higher volume of the house HA (by factor 2) in comparison to apartment and house HB, allowing dispersion.

3.2. Number size distribution during occupancy and total monitoring periods in residences

Average number size distribution in the apartment for occupancy and total monitoring are given in Fig. 1, additionally non-occupancy and periods with active and no sources are plotted. Average PNC during active sources in the apartment is about 13 times higher than during no source period (18 000 and 1300 particles cm⁻³, respectively). Number size distribution during total monitoring, occupancy and active sources is bi-modal with the modes about 20 and 60 nm. Number size distribution is uni-modal during non-occupancy and no source periods with modes about 80 and 100 nm, respectively. From Fig. 1 it can be seen that in the apartment PNC is dominated by particles smaller than 300 nm with significant contribution of ultrafine particles (<100 nm). The occupancy time in the apartment accounted for about 70% of the total sampling time. Out of the occupancy time 64% was during the active source period. During active source period particles smaller than 100 nm contributed to 77% of total PNC, while during no source period their contribution accounted for 50%. Contribution of

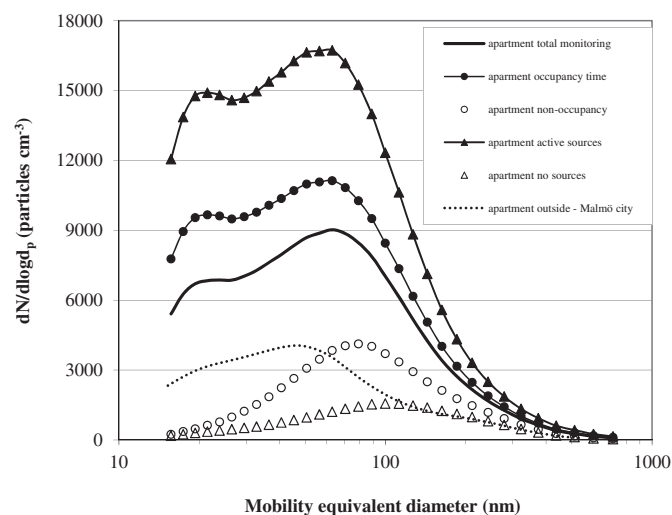


Fig. 1. Average number size distributions in the apartment for different occupancy periods, total monitoring and periods with active and no sources.

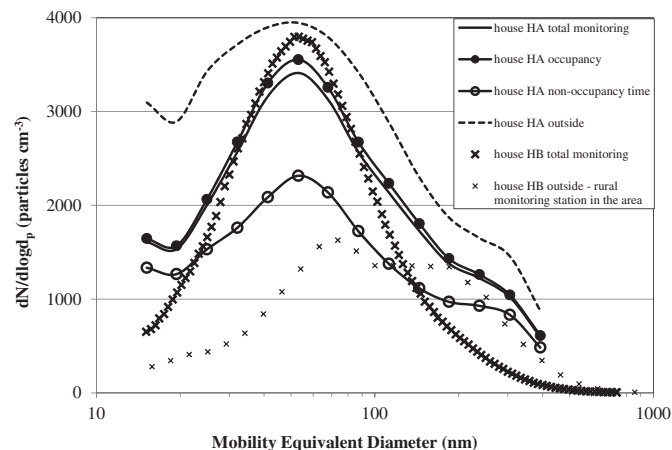


Fig. 2. Average number size distributions in the house HA for different occupancy periods, total monitoring and outside. Average number size distributions in the house HB for total monitoring period and outside are given for comparison.

particles smaller than 300 nm to total PNC for active and no source period was similar, accounting for 94 and 90%, respectively.

Morawska et al. (2003) measured average concentrations for 48 h in 15 houses in Brisbane. Their reported average concentration value for active source period is comparable to this study 18 200 and 18 000 particles cm^{-3} , respectively. However average outdoor concentrations in Brisbane (10 900 particles cm^{-3}) were higher in comparison to the concentrations in Malmö (7000 particles cm^{-3} , Roldin et al., 2010).

Average number size distribution in the houses HA and HB for occupancy, non-occupancy, total monitoring periods are given in Fig. 2. Number size distributions in house HA during occupancy and total monitoring periods are very similar. It can clearly be seen that all number size distributions in house HA display the same shape, the outdoors size distribution is somehow wider than indoors and the outdoors concentrations are higher. In all size distributions in house HA mainly one mode about 50 nm is visible, although the shapes of the curves indicate that there is a second mode below measured size range with this instrument. Average number size distribution in house HB is uni-modal with mode about 50 nm. Only total monitoring period in house HB is presented as occupancy averaging period number size distribution did not display any significant difference.

3.3. PNC during occupancy and total monitoring periods in non-residential indoor environments

In Table 3 PNC characteristics for studied non-residential indoor environments are summarised during occupancy periods i.e. teaching hours in both schools and opening hours in the supermarket and restaurant. The average PNC were relatively low in non-residential sites and ranged from 800 particles cm^{-3} in the restaurant to 2800 particles cm^{-3} in the supermarket. PNC characteristics during total measurements periods in the non-residential sites (both schools, the supermarket and restaurant) were very similar to presented occupancy periods, thus are not presented here. It has to be noted that in case of non-residential sites some misclassification of the data cannot be excluded, i.e. presence of staff outside teaching and opening hours could have occurred due to cleaning, food preparation (in the restaurant) or restocking (in the supermarket).

In both schools during teaching hours average PNC was between 1800 and 2000 (min 800, max 4000) particles cm^{-3} . Recent studies, summarised in a review by Morawska et al. (2013), report that in schools average PNC range from 2000 to 81 000 particles cm^{-3} . Thus the average PNC values obtained in this study belong to lower concentrations measured in schools.

Among the studied non-residential sites the highest average and median value of 2800 and 2300 particles cm^{-3} , respectively, was observed in the supermarket, despite high AER ($\sim 10 \text{ h}^{-1}$) and supply of ventilation air with low particle loads (median 850, min

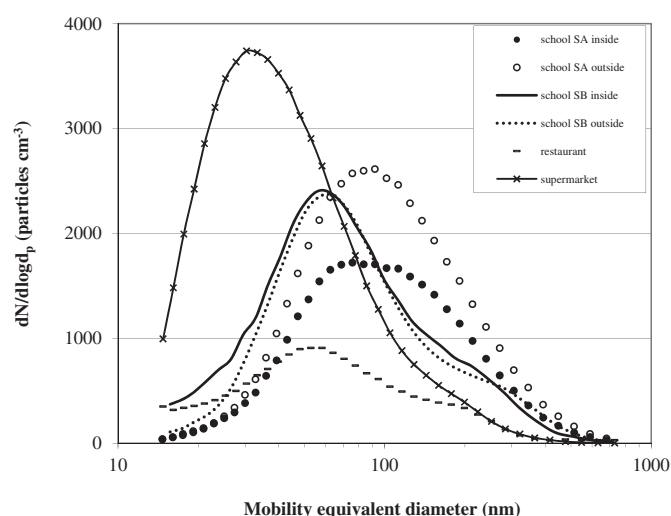


Fig. 3. Average number size distributions for studied non-residential indoor environments during teaching hours in both schools and opening hours in the supermarket and restaurant. Measured concentrations outside schools SA and SB are also plotted.

200, max 4000 particles cm^{-3}). Possible explanation for observed relatively high concentration is given in section “Number size distribution during occupancy and total monitoring periods in non-residential sites”.

In the fast food restaurant (Table 3) the lowest among non-residential sites, average and median PNC values (800 and 700 particles cm^{-3}) were observed. PNC found inside mirrored the concentrations measured in the supplied ventilation air with a median value of 600 particles cm^{-3} , ranging from 100 to 9000 particles cm^{-3} . These low concentrations in the restaurant can be explained by high AER in the dining section, ventilation system with good filtration efficiency (glass fibre F7) operating 24 h per day and separate extraction systems in the kitchen where grilling and frying took place. Levy et al. (2002) reported 140 000 particles cm^{-3} in a seating area of a mall food court in Boston (measured with TSI P-Trak, range 20–1000 nm); no information on AER or ventilation system was provided. These values are much higher in comparison to measured concentrations in the restaurant in this study. Higher AER and possibly lower intensity of cooking (in comparison to Boston mall food court) could contribute to it.

3.4. Number size distribution during occupancy and total monitoring periods in non-residential indoor environments

Average number size distribution for studied non-residential sites is presented in Fig. 3. In Fig. 3 apart from concentrations in schools SA and SB during teaching hours, the measured

Table 3
Total submicron PNC (15–700 nm, SMPS measurements) for studied non-residential indoor environments during teaching hours in both schools and opening hours in the supermarket and restaurant. The PNC during total monitoring periods, not presented here, were similar to given occupancy periods.

| | Average | STD ^a | Median | Min | Max | Average number GMD ^b | Average GSD ^c |
|-------------|-------------------------------|------------------|--------|-----|--------|---------------------------------|--------------------------|
| | (particles cm^{-3}) | | | | | (nm) | |
| School SA | 2000 | 600 | 1800 | 800 | 3400 | 96 | 2.0 |
| School SB | 1800 | 800 | 1700 | 800 | 4000 | 68 | 2.0 |
| Supermarket | 2800 | 1600 | 2300 | 700 | 11 000 | 41 | 1.9 |
| Restaurant | 800 | 500 | 700 | 200 | 9100 | 52 | 2.4 |

^a Standard deviation.

^b Geometric mean diameter.

^c Geometric standard deviation.

concentrations outdoors are also plotted. Number size distribution in both schools as well as outdoors is uni-modal with modes about 90 nm in school SA and 60 nm in school SB. In naturally ventilated school SB the average number size distribution as well as PNC mirrored the outdoor ones. Whereas in mechanically ventilated school SA the same shape of number size distribution is observed with lower concentrations, which represents effect of particles removal due to filtration in mechanical ventilation system. Similarity of number size distribution measured inside and outside in both schools could be explained due to: 1) lack of strong indoor sources of submicrometer particles in schools (no cooking, candle burning, tobacco smoking, etc.), and 2) a clear dependence of indoor PNC in schools on outdoor concentrations influenced by traffic related pollution as confirmed in published studies summarised by Morawska et al. (2013).

Average particle number size distribution in the supermarket is different in comparison to all non-residential sites presented in Fig. 3. The mode appears at lower diameter (about 30 nm) and the highest PNC (2800 particles cm^{-3}) are seen compared to other non-residential sites. In the supermarket AER was high ($\sim 10 \text{ h}^{-1}$) and particle concentrations in supplied ventilation air was low (median 850, min 200, max 4000 particles cm^{-3}). The median geometric mean diameter inside the supermarket was 41 nm which is about 20 nm smaller than in the supplied ventilation air, which indicates an indoor source of particles. Presence of nucleation mode particles (as seen in Fig. 3) is likely due to local emissions of terpenes (from washing powders, cleaning products and air fresheners), which in the presence of ozone from outdoors (supplied in ventilation air) form particles due to gas-to-particle conversions (Wainman et al., 2000; Weschler, 2003). Additional experiments were conducted in the laboratory chamber to investigate the possibility of formation of nucleation mode particles from cleaning products (in unopened packaging – mimicking supermarket conditions) at low ozone levels (16 ppb, matching the outdoor level during measurements in supermarket). The results confirmed such a possibility and are described in Wierzbicka et al. (2011).

3.5. Comparison of approximation of mass concentration (photometer reading) during occupancy and total monitoring periods

Photometer readings in the studied indoor environments are summarised in Table 4. The highest average $\text{PM}_{2.5}$ concentrations were observed in the apartment $28 \mu\text{g m}^{-3}$ and the lowest in the supermarket and restaurant 7 and $8 \mu\text{g m}^{-3}$, respectively. Consistently average and median concentrations during occupancy time

Table 4
Approximation of $\text{PM}_{2.5}$ mass concentrations (DustTrak readings) for different averaging periods in studied indoor environments.

| | | Average | STD ^a | Median | Min | Max |
|-------------|----------------------------|-----------------------|------------------|--------|-----|-----|
| | | (ug m^{-3}) | | | | |
| Apartment | Total monitoring | 28 | 41 | 18 | 5 | 403 |
| | Occupancy | 36 | 51 | 21 | 8 | 403 |
| | Non-occupancy | 17 | 18 | 13 | 5 | 171 |
| | Outdoors | 19 | 12 | 18 | 61 | 2 |
| School SA | Total monitoring | 21 | 10 | 19 | 7 | 47 |
| | Teaching hours | 24 | 11 | 25 | 8 | 43 |
| School SB | Total monitoring | 11 | 11 | 6 | 1 | 47 |
| | Teaching hours | 13 | 13 | 8 | 2 | 47 |
| | Outdoors | 9 | 6 | 9 | 1 | 34 |
| Supermarket | Opening hours ^b | 7 | 2 | 6 | 20 | 5 |
| Restaurant | Opening hours ^b | 8 | 1 | 8 | 14 | 2 |

^a STD – standard deviation.

^b Total monitoring periods were identical to opening hours values.

(teaching hours in schools) were higher than during total monitoring period in the apartment and both schools.

In the apartment the $\text{PM}_{2.5}$ mass concentration approximation was higher during occupancy time than during total monitoring period by 29% for average and 17% for median values. It is worth noting that relative comparison of photometer readings for considered averaging periods eliminates uncertainty of absolute reading values related to instrumental issues (described in Method section). In the apartment measured average values outdoors were lower than average concentration indoors both during occupancy and total monitoring. The median outdoor concentration was the same as total monitoring median ($18 \mu\text{g m}^{-3}$). Median $\text{PM}_{2.5}$ mass concentration $18 \mu\text{g m}^{-3}$ during total monitoring period in the apartment is within the range of values reported in other publications summarised in review by Morawska et al. (2013) i.e. between 8 and $33 \mu\text{g m}^{-3}$. When comparing reported $\text{PM}_{2.5}$ mass concentrations one has to bear in mind that frequently different averaging periods as well as instrumentation and analytical methods were used. Smoking residences were excluded as tobacco smoking is a known major source of fine particles, which has been reported to increase $\text{PM}_{2.5}$ mass concentration by 58–130% (Stranger et al., 2007; Breyse et al., 2005).

In Fig. 4 simultaneous indoor/outdoor $\text{PM}_{2.5}$ photometer readings in the apartment, are shown. Additionally plotted is outdoor $\text{PM}_{2.5}$, obtained from nearby monitoring point in city centre (as described in Materials and Methods). In Fig. 4, typically for indoor environments with active indoor sources, a sharp increase in particle concentration at the beginning of each indoor activity is observed. When the activity is ceased, a slow decrease follows. The decrease to background (no source) level in the apartment took up to 12.5 h, which is expected as the AER is about 0.3 h^{-1} . Similar patterns, i.e. sharp increases due to indoor activities with slower decrease, were observed in earlier studies (Afshari et al., 2005; Morawska et al., 2003; Wallace, 2006) and are expected for the low AER in naturally ventilated houses in winter time in northern Europe. These sharp increases in particle concentration (expressed both in mass and number concentration) are typical for occupancy time as occupant activities dictate their occurrence. These sharp increases, contributions from indoor sources, are clearly visible in Fig. 4 and reach values up to 20 times higher in comparison to the observed outdoor concentration. These short term peaks were identified, as different types of cooking and candle burning. Short term increases of $\text{PM}_{2.5}$ due to different type of cooking between 2 and 100 times greater than background or outdoor concentrations were reported by Abt et al. (2000), He et al. (2004), Morawska et al. (2003) and Wallace et al. (2004). The very high maximum concentration of $\text{PM}_{2.5}$ in the apartment in this study ($403 \mu\text{g m}^{-3}$) was due to cooking and can be compared to median peak concentration values of 745, 735, 718 $\mu\text{g m}^{-3}$ reported by He et al. (2004) due to frying, cooking pizza and grilling, respectively.

In both schools the $\text{PM}_{2.5}$ mass concentration approximation was higher during teaching hours than during total monitoring period (Table 4). On average the difference for both schools, between teaching hours and total monitoring, accounts to 16% for average and 32% for median values. In both schools the average values for total monitoring period (as presented in Table 4, i.e. 21 and $11 \mu\text{g m}^{-3}$ for SA and SB, respectively) are comparable to other reported measurements (summarised in review by Morawska et al., 2013) which range from 2 to $95 \mu\text{g m}^{-3}$. In school SB median outdoor concentration ($9 \mu\text{g m}^{-3}$) was higher than median during total monitoring and teaching hours (6 and $8 \mu\text{g m}^{-3}$, respectively).

In the fast food restaurant, the DustTrak inside measured PM_{10} mass concentration, with a median value of $8 \mu\text{g m}^{-3}$. The influence of outdoor concentrations was not noticed. This indicates high particle removal of the filters used in the ventilation. The authors of

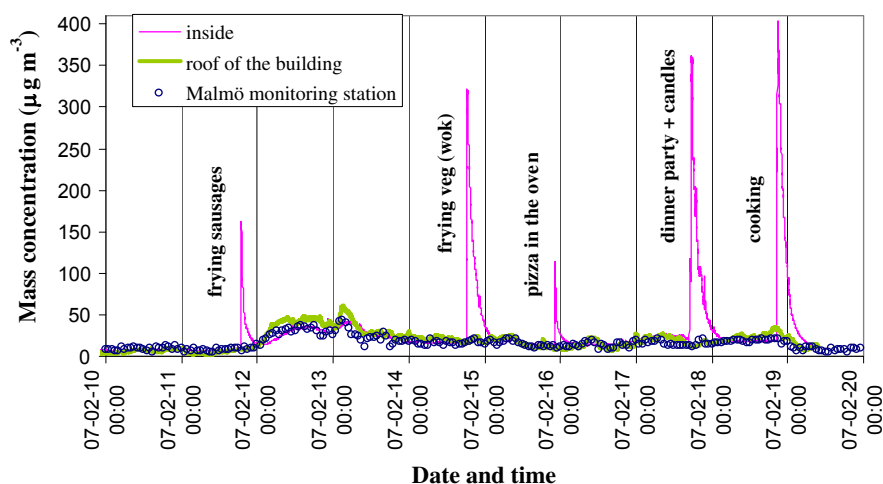


Fig. 4. Comparison of DustTrak readings indoors in the apartment and on the roof of the building together with PM_{2.5} mass concentration measured by TEOM at the monitoring point at the roof level in Malmö city centre (Roldin et al., 2010).

this study did not find any reference values for PM₁ concentrations measured in restaurants. PM_{2.5} values given by Travers et al. (2004) for 20 bars and restaurants after the smoking prohibition implemented in 2003 in the USA are higher ($24 \mu\text{g m}^{-3}$) than values obtained in the restaurant in this study. Many reference values were found for Chinese style of cooking but these were much higher e.g. $312 \mu\text{g m}^{-3}$ in See and Balasubramanian (2006) and $1167 \mu\text{g m}^{-3}$ in Lee et al. (2001).

The authors did not find any reference values for PM_{2.5} mass concentration measured in supermarkets at similar conditions.

3.6. Indoor sources

3.6.1. Identification of sources

In order to understand the difference between occupancy and total monitoring periods indoor sources of particles were identified from log books. The activities that significantly increased the PNC consisted of cooking (frying sausages, onions, vegetables, chicken, omelette, pizza in the oven), burning candles, burning incense, peeling oranges and tangerines, plugging in an air freshener. Fig. 5 presents the number size distribution at peak concentrations from different activities in the apartment. The highest peak contribution was observed at the dinner party, with vigorous cooking followed by seven candles burning. Observed peak concentrations are influenced by air exchange rate, site volume and deposition rates. This is why source strengths, which are independent of these variables, are more suitable for comparisons and are presented below.

3.6.2. Source strengths

Source strengths for some activities were estimated on the basis of measurements in the apartment and are summarised in Table 5, together with peak concentration and decay rates for given size intervals. Source strengths in house HA were reported in Hussein et al. (2005). In house HB overlapping activities did not allow identification of activity meeting the criteria described in the section “Source strengths estimation”. Table 5 comprises only activities meeting the selection criteria. Estimated total source strengths for cooking events range from 6.3×10^{11} to 1.7×10^{12} particles min^{-1} , lowest for frying an omelette (no oil, Teflon frying pan used) and highest for cooking (specified as “cooking in activity log”). The highest contribution is observed from ultrafine particles (15–100 nm), their source strengths range from 6.0×10^{11} to 1.3×10^{12} particles min^{-1} , which accounts on average

for 84% of total generated submicron particles. Peeling tangerines produced 3.9×10^{10} particles min^{-1} , and ultrafine particles accounted for 91%. Source strengths estimated by He et al. (2004) in 15 houses in Brisbane for cooking pizza, frying, cooking and grilling range from 1.7×10^{11} to 5.7×10^{11} particles min^{-1} . These are slightly lower but comparable with our results. Values reported by Wallace et al. (2004), 3.0×10^{12} particles min^{-1} from 44 cooking episodes agree with our upper limit of obtained source strengths. Wallace (2006) published source strengths per cooking event for 381 cooking events. Assuming average active cooking time lasting 20 min, recalculated source strengths for tea and toast, tortillas and broiled fish were 2.5×10^{11} , 2.0×10^{12} and 1.4×10^{12} particles min^{-1} , and are comparable to the values obtained in this study. Afshari et al. (2005) in a full-scale chamber study reported source strength of 8.3×10^{11} particles min^{-1} for frying meat, which agrees well with the values obtained here.

4. Conclusions

In the apartment and both schools PM_{2.5} mass concentration (photometer readings) consistently showed higher average and median concentrations during occupancy time (in schools teaching hours) in comparison to total monitoring period. In the apartment the occupancy time average PM_{2.5} mass concentration was 29% higher than during total monitoring periods, while median was 17% higher. On average the difference in both schools, between teaching hours and total monitoring, accounts to 16% for average and 32% for median values. The relative comparison of photometer readings presented here eliminates uncertainty of absolute reading values related to these instruments (described in Method section).

Results of the comparison of PNC in residences during occupancy and total monitoring period were not consistent. In the apartment average and median PNC were 33% and 58%, respectively, higher during occupancy in comparison to total monitoring period, which agrees with the results in recently published studies, conducted in larger amount of residences in Scandinavia. Whereas in houses HA and HB average and median PNC were very similar for the two averaging periods. General conclusions on the basis of measurements performed in three residences cannot be drawn, however the results confirm strong dependence on type and frequency of indoor activities generating particles and site specificity (e.g. building characteristics, volume, AER).

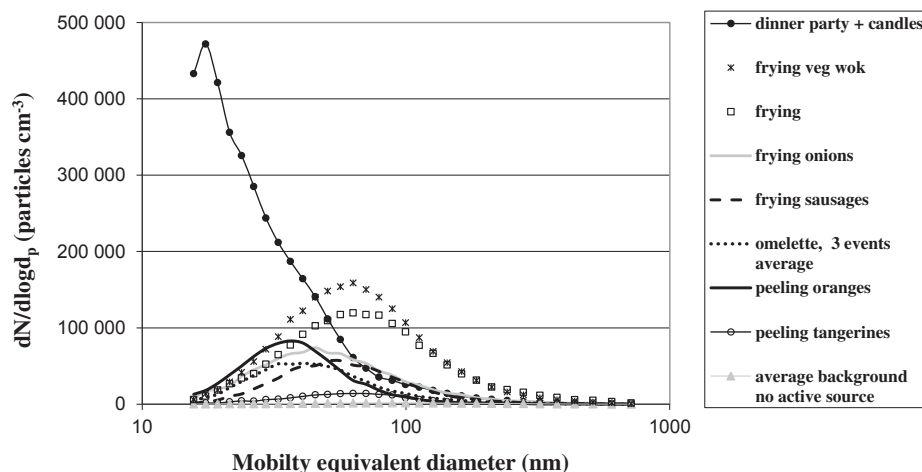


Fig. 5. Number size distribution at peak concentrations from different activities in the apartment.

In both schools PNC during teaching hours were very similar to values during total measurement periods and were mainly influenced by outdoor concentrations. This finding is consistent with previous studies i.e. that in indoor environments, without strong indoor sources of particles, observed indoor PNC values are mainly influenced by outdoor concentrations that can be modified by used filtration system and building characteristics.

The major differences observed between occupancy and total monitoring periods in residences were due to indoor sources, which can greatly elevate indoor particle levels for prolonged periods (up to 12.5 h after activity ceased), especially in residences with low AER. Due to different types of cooking and candle burning in the apartment short term peak mass concentrations (photometer readings) were observed up to $400 \mu\text{g m}^{-3}$ and peak PNC up to

Table 5
Source strengths estimations for some activities in the apartment.

| Activity | Size interval (nm) | Decay rate ^a ($a + k$) (h^{-1}) | Peak | | | Time ^d (min) | Source strength (particles min^{-1}) |
|---|--------------------|---|---|------------------------------|------------------|-------------------------|--|
| | | | Concentration (particles cm^{-3}) | Number GMD (nm) ^b | GSD ^c | | |
| Cooking | 15–50 | 1.1 | 24 200 | | | | 5.07E+11 |
| | 50–100 | 0.8 | 38 400 | | | | 7.70E+11 |
| | 100–300 | 0.5 | 18 200 | | | | 3.51E+11 |
| | 300–700 | 0.5 | 2500 | | | | 4.69E+10 |
| | total | | 83 300 | 69 | 1.9 | 15 | 1.67E+12 |
| Frying sausages | 15–50 | 1.2 | 13 100 | | | | 3.39E+11 |
| | 50–100 | 0.7 | 41 700 | | | | 3.58E+11 |
| | 100–300 | 0.4 | 4000 | | | | 8.37E+10 |
| | 300–700 | 0.1 | 300 | | | | 4.03E+09 |
| | total | | 59 100 | 58 | 1.8 | 12 | 7.85E+11 |
| Veg wok | 15–50 | 1.2 | 25 200 | | | | 3.07E+11 |
| | 50–100 | 0.8 | 54 000 | | | | 6.10E+11 |
| | 100–300 | 0.5 | 22 100 | | | | 2.30E+11 |
| | 300–700 | 0.5 | 2100 | | | | 2.16E+10 |
| | total | | 103 400 | 64 | 1.9 | 30 | 1.17E+12 |
| Peeling 1 kg of tangerines | 15–50 | 0.6 | 2600 | | | | 1.36E+10 |
| | 50–100 | 0.4 | 4300 | | | | 2.13E+10 |
| | 100–300 | 0.2 | 1100 | | | | 3.58E+09 |
| | 300–700 | 0.1 | 70 | | | | 6.55E+07 |
| | total | | 8070 | 61 | 1.8 | 59 | 3.85E+10 |
| Frying omelette (3 events average) | 15–50 | 0.9 | 17 100 | | | | 3.70E+11 |
| | STD ^e | (0.2) | (6900) | | | | (1.83E+11) |
| | 50–100 | 0.6 | 10 500 | | | | 2.24E+11 |
| | STD ^e | (0.1) | (6200) | | | | (1.59E+11) |
| | 100–300 | 0.3 | 2200 | | | | 3.56E+10 |
| | STD ^e | (0.2) | (1300) | | | | (2.95E+10) |
| | 300–700 | 0.1 | 200 | | | | 1.38E+09 |
| | STD ^e | (0.1) | (90) | | | | (1.32E+09) |
| | total | | 30 000 | 45 | 1.8 | 15 | 6.31E+11 |
| | STD ^e | | (14 490) | | | | (3.64E+11) |

^a Measured AER at low ventilation conditions equals 0.3 h^{-1} .

^b Geometric mean diameter.

^c Geometric standard deviation.

^d Time from initial background concentration (one measurement point preceding first observed concentration increase) until peak concentration was reached.

^e Standard deviation.

246 000 particles cm^{-3} (about 20 and 35 times higher than the average outdoor concentration, respectively).

In summary, to improve exposure assessment to particles, crucial for studying health effects, only data from occupancy periods should be used. Non-occupancy periods, while useful for studying influence of outdoor particles and establishing a baseline, should not be included in the exposure assessment. Data from total monitoring, as it includes non-occupancy periods, may underestimate the concentrations for exposure assessment. Detail characterisation of particles and better assessment of personal exposure indoors is of interest for epidemiological and toxicological studies. It can help in raising general awareness and guide development of control strategies. The number size distributions reported here could be also used for estimating lung deposited dose needed in studying health effects.

The importance of an adequate ventilation system, capable of removing and not allowing the accumulation of both particles and gaseous emissions (not studied here) generated by indoor sources, becomes apparent. This needs to be emphasised especially in the light of efforts to make residential houses energy efficient, which is frequently done at the expense of reduced AER. It is not possible to regulate activities in private homes to reduce particle exposure. However much can be done to enclose spaces and enhance extraction and ventilation in locations where strong indoor sources of particles occur, namely in the kitchen.

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