Atmospheric Environment 106 (2015) 458–466

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Contribution of indoor-generated particles to residential exposure

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HIGHLIGHTS

- Several high time resolution instruments were operated for seven days in 22 homes.
- Concentrations above 10^4 cm^{-3} almost only occur during active periods of occupancy.
- Known and unknown indoor activities were 86% of the total integrated daily residential exposure.
- Source strengths of specific activities ranged from $1.6 \cdot 10^{12}$ to $4.5 \cdot 10^{12}$ min^{-1}.
- Correlation between UFP and mass conc of soot in total dust was on average 56%.

ARTICLE INFO

Article history:
Received 4 April 2014
Received in revised form
18 July 2014
Accepted 30 July 2014
Available online 1 August 2014

Keywords:
Ultrafine particles
Indoor sources
Exposure
Indoor measurements
Soot
Source strength

ABSTRACT

The majority of airborne particles in residences, when expressed as number concentrations, are generated by the residents themselves, through combustion/thermal related activities. These particles have a considerably smaller diameter than 2.5 μm and, due to the combination of their small size, chemical composition (e.g. soot) and intermittently very high concentrations, should be regarded as having potential to cause adverse health effects.

In this study, time resolved airborne particle measurements were conducted for seven consecutive days in 22 randomly selected homes in the urban area of Lund in southern Sweden. The main purpose of the study was to analyze the influence of human activities on the concentration of particles in indoor air. Focus was on number concentrations of particles with diameters <300 nm generated by indoor activities, and how these contribute to the integrated daily residential exposure. Correlations between these particles and soot mass concentration in total dust were also investigated.

It was found that candle burning and activities related to cooking (using a frying pan, oven, toaster, and their combinations) were the major particle sources.

The frequency of occurrence of a given concentration indoors and outdoors was compared for ultrafine particles. Indoor data was sorted into non-occupancy and occupancy time, and the occupancy time was further divided into non-activity and activity influenced time. It was found that high levels (above 10^4 cm^{-3}) indoors mainly occur during active periods of occupancy, while the concentration during non-activity influenced time differs very little from non-occupancy time.

Total integrated daily residential exposure of ultrafine particles was calculated for 22 homes, the contribution from known activities was 66%, from unknown activities 20%, and from background/non-activity 14%.

The collected data also allowed for estimates of particle source strengths for specific activities, and for some activities it was possible to estimate correlations between the number concentration of ultrafine particles and the mass concentration of soot in total dust in 10 homes. Particle source strengths (for 7 specific activities) ranged from $1.6 \cdot 10^{12}$ to $4.5 \cdot 10^{12}$ min^{-1}.

The correlation between ultrafine particles and mass concentration of soot in total dust varied between 0.37 and 0.85, with an average of 0.56 (Pearson correlation coefficient).

This study clearly shows that due to the importance of indoor sources, residential exposure to ultrafine particles cannot be characterized by ambient measurements alone.

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http://dx.doi.org/10.1016/j.atmosenv.2014.07.053

1. Introduction

The main aims of this study were to analyze the influence of human activities on particles in indoor air, with focus on determining the differences between residential and outdoor number concentrations of ultrafine particles; to estimate contribution of indoor sources to integrated daily residential exposure; and to check correlation between mass concentration of soot in total dust and number concentration of ultrafine particles in residences in general as well as during different activities.

In the industrialized part of the world, we spend approximately 65% of our lives in our homes (Leech et al., 2002; Brasche and Bischof, 2005). Hereby we are subjected to various indoor-generated airborne particles as well as to background particles originating from outdoors. It is widely known that outdoor particles contribute to the indoor aerosol concentration levels, and in epidemiology, exposure is often determined based on the outdoor particle concentration. However the outdoor contribution to the indoor aerosol size distribution is modified compared to the distribution outdoors, due to size-specific differences in penetration efficiency (Thatcher et al., 2003; Liu and Nazaroff, 2003; Nazaroff, 2004). Accumulation mode particles have the highest penetration (Nazaroff, 2004) since these particles have a rather low diffusivity and are too small to be significantly affected by sedimentation or impaction. To a far greater extent than outdoor generated particles, the particle concentration and size distribution indoors are dominated by particles generated by the residents' activities. For naturally ventilated buildings, Morawska and Salthammer (2003) summarized indoor/outdoor ratios (I/O) for PM_{10} and $PM_{2.5}$ (particulate matter smaller than 10 μm and 2.5 μm, respectively) from 0.5 to 0.98 and 0.54 to 1.08, respectively, in the absence of indoor sources. However, when indoor sources were present, I/O ratios for PM_{10} and $PM_{2.5}$ ranged from 1.14 to 3.91 and 1 to 2.4, respectively, which stress the significance of indoor source contributions (Morawska et al., 2013).

It has been shown (Hussein et al., 2006; Wallace, 2006; Turpin et al., 2007 Wierzbicka, 2008) that the major sources contributing to indoor air concentrations are combustion related or related to thermal processes (e.g. cooking, smoking and candle burning) and electric appliances. Peak number concentrations from cooking have been found to be higher than reported outdoor peak concentrations (Dennekamp et al., 2001; He et al., 2004; Wan et al., 2011), by at least an order of magnitude. It is well known that combustion generated particles generally are considerably smaller than 2.5 μm, often smaller than 300 nm (Géhin et al., 2008), which justifies that number concentration of ultrafine particles would be a more relevant metric than mass concentration to determine residential exposure to combustion-related particles. Ultrafine (<100 nm) particles are of special interest from a health perspective, since they, due to their small size, can penetrate deep into the respiratory system and cause inflammatory effects (Long et al., 2001). Particles <100 nm also have a higher deposition rate in both the upper airways and the alveolar tract, which in itself can cause adverse effects (Hirano, 2009; Araujo et al., 2008; Oberdörster et al., 2005). Studies (Wang et al., 2009; Kennedy, 2007) have shown that the number concentration of ultrafine particles induce more adverse health effects than the mass concentrations of PM_{10} and $PM_{2.5}$ (to which the ultrafine particles are a poor contributor).

Soot (black or elemental carbon), is a primary component in several emissions known to affect human health indoors, such as wood burning, cigarette smoke, cooking with poor ventilation but also e.g. diesel exhaust, which can infiltrate the building from outdoors. Soot particles are often carriers of elements such as polycyclic aromatic hydrocarbons (PAH) (Pagels et al., 2009), but are of health interest also due to their small size and because of the large surface area available for adsorption of gases and other particle phase toxins. Since the major sources of ultrafine particles indoors have been shown to be combustion/thermal related, it is likely that soot is a major component of this aerosol and it is therefore of interest to monitor the soot concentration indoors. The importance of the potential health effects of indoor generated soot has been pointed out by e.g. Buonanno et al. (2013). Monitoring soot concentrations in parallel to ultrafine particle number concentrations allows determination of their correlation, and, hypothetically, as a consequence gives a possibility of predicting the soot component on the basis of monitoring ultrafine particles.

2. Methods

For seven consecutive days, time resolved stationary air measurements were conducted in 22 randomly selected homes in the urban area of Lund in southern Sweden. The measurements were conducted during wintertime. Home characteristics, including ventilation type, air exchange rate, volume, indoor temperature and RH, and instruments used are summarized in Table 1. Initially measurements were conducted in 39 homes; however mainly due to technical reasons (see data processing for details) results from only 22 homes were included in this article.

The habitants filled in detailed activity log books specifying conducted activities and made notes of when they were present or absent in the dwelling.

2.1. Measurements and instrumentation

Number concentrations, and mean diameter, of ultrafine particles were monitored with Mini Diffusion Size Classifier (DiSC) (University of Applied Sciences, Windisch, CH) or Nanotracer (Philips Aerasense). These instruments are based on an electrical measurement technique, with a diffusion charger, by which the average acquired charge of a particle depends on its size, followed by a diffusion stage. This stage consists of a stainless steel grid, where the particles will deposit due to diffusion, connected to electrometers that detect the current carried by the deposited particles. The current is dependent on flow rate, number concentration and particle size. Both instruments have an upper size limit of 300 nm. Since over 80% of particles measured by these two instruments were below 120 nm, and the majority were below 100 nm, with a minimal contribution of particles between 100 and 300 nm, the measurements of the Nanotracer and DiSC can be considered as good approximations of ultrafine particles (number concentrations measured with Nanotracer and DiSC will in this article be entitled ultrafine particles). The accuracy of both the Nanotracer and DiSC are ±30% (Asbach et al., 2012).

The measurements of particles larger than 0.3 μm were performed by an optical particle counter, Indoor Air Quality monitor (IAQ3016, Lighthouse). Particles passing a laser beam inside the instrument scatter light. A photo detector converts the scattered light to an electrical pulse. For a single particle, the light intensity of the generated pulse depends on the size, refractive index and color of the particle. According to a reference aerosol of monodisperse PSL particles, the pulse height is transferred to six particle size intervals. The pulse intensity thresholds in the IAQ3016 correspond to particle sizes of 0.3, 0.5, 1.0, 2.5, 5.0 and 10 μm.

Black carbon (soot) levels were monitored by microAeth (AE51, Magee Scientific) in 10 of the homes, to investigate the correlation between soot measurements and indoor (particularly combustion

Table 1
Homes and measurement characteristics including measurement duration, temperature, RH and used instruments.

	Type of home	Measurement duration [days]	AER [h^{-1}]	Vent.	Volume [m^3]	Temp (°C) mean (std)	RH (%) mean (std)	Use of Nanotracer/DiSC[a]	Use of Aeth[b]	Use of IAQ[c]
01	House	6.12	0.4	A	293	22.0 (0.4)	47 (3)	Yes	Yes	No
02	House	6.10	0.7	C	350	22.0 (0.4)	47 (3)	Yes	No	No
03	Apartm.	7.26	0.4	D	250	23.6 (0.6)	35 (3)	Yes	Yes	No
04	House	6.95	N/A	A	N/A	22.4 (0.4)	47 (3)	Yes	No	No
05	Apartm.	2.42	1.6	B	168	21.9 (1.5)	37 (3)	Yes	Yes	No
06	Apartm.	7.14	1.5	A	96	21.8 (0.9)	38 (4)	Yes	Yes	No
07	Apartm.	6.59	1.4	C	227	23.4 (0.6)	36 (4)	Yes	No	No
08	House	6.50	2.3	C	327	23.5 (0.5)	37 (2)	Yes	No	No
09	Apartm.	6.30	1.0	B	202	22.9 (0.3)	34 (4)	Yes	No	Yes
10	Apartm.	6.64	2.9	A	N/A	N/A	N/A	Yes	No	No
11	Apartm.	6.82	1.1	D	118	20.7 (0.4)	43 (4)	Yes	No	Yes
12	House	6.61	1.0	C	288	22.8 (0.7)	35 (2)	Yes	Yes	No
13	House	6.55	1.0	N/A	N/A	22.3 (0.6)	43 (2)	Yes	Yes	No
14	Apartm.	7.10	0.3	A	195	22.3 (0.4)	38 (3)	Yes	No	Yes
15	Apartm.	5.69	0.4	B	132	N/A	N/A	Yes	No	No
16	Apartm.	8.95	1.4	A	208	23.4 (1.4)	25 (2)	Yes	Yes	Yes
17	House	6.33	N/A	A	442	23.3 (0.9)	26 (2)	Yes	Yes	Yes
18	House	6.25	N/A	C	N/A	22.3 (1.0)	26 (2)	Yes	No	Yes
19	Apartm.	6.92	N/A	A	N/A	25.2 (1.6)	28 (5)	Yes	Yes	No
20	Apartm.	7.01	N/A	C	N/A	23.0 (1.3)	30 (5)	Yes	No	Yes
21	House	7.00	N/A	A	328	21.8 (1.3)	35 (4)	Yes	No	No
22	House	6.62	N/A	N/A	N/A	N/A	N/A	Yes	Yes	No

A) Additional exhaust mechanical ventilation in kitchen only (kitchen exhaust hood).
B) Additional exhaust mechanical ventilation in kitchen and bathroom(s).
C) Mechanical exhaust air in kitchen and bathroom(s) and fresh air supply valve ventilation in other rooms.
D) Natural ventilation.
[a] Instruments measuring number concentration of particles <300 nm.
[b] Instrument measuring mass concentration of black carbon.
[c] Instrument measuring number (and mass) concentration of particles 0.3–10 μm.

related) activities. The soot monitor measures the transmission of infrared (IR) light (880 nm) through a filter, which is continuously loaded with airborne particles by the suction of an internal pump. The accumulation of particles on the filter over time increases the absorbance, which is calculated relative to a reference cell. The attenuation IR is then transferred to mass concentration of black carbon (described in detail by Cheng and Lin (2013)).

Temperature and relative humidity were logged (USB-500, Measurement Computing). All measurements were made during off-pollen season (October–April). The instruments were zero-calibrated before each home, and placed between 1 and 1.5 m above the floor centrally in the dwelling, but not in, or in direct connection to, the kitchen, on a rack of shelves. The rack was constructed for occupying a minimum of space so that the instruments could be placed at a central spot without being in the way of daily living.

Ambient (outdoor) number concentrations were monitored constantly by a scanning mobility particle sizer (SMPS, consisting of a TSI 3071 Differential Mobility Analyzer, DMA and a TSI 3010 Condensation Particle Counter, CPC) from a station in northern Lund. All homes were located within 5 km radius from the ambient monitoring station.

CO_2 data was collected in one bedroom in each residence. CO_2 concentration was measured with CARBOCAP CO_2 monitors (GMW22, Vaisala, Finland) connected to a HOBO12-012 data loggers (Onset Computer Corp., USA) to record the measured values. CO_2 levels were used for estimation of air exchange rates on the basis of occupant-generated CO_2 mass balance method as described by Bekö et al. (2010). The method is based on analyzing steady state concentrations or buildups and decays of CO_2 in bed rooms. In some cases the bedroom design was such that no obvious build-up occurred (e.g. no door, open window, bed placed in living room), and in a few cases it was not possible to estimate the bedroom volume (open space apartment). In total, it was possible to estimate air exchange rates in 15 homes.

A thorough examination of each home – e.g. construction year, floor and wall materials, and ventilation system – was conducted according to a structured protocol. In addition, the participants filled in a translated and modified version of the IUATLD (International Union Against Tuberculosis and Lung Disease) questionnaire (Burney and Chinn, 1987; Burney et al., 1989) covering both the characteristics of the dwelling and the residents and their everyday habits. The results of these examinations will be presented elsewhere.

2.2. Data processing

In total, data measured in 24 homes is presented here. Initially measurements were conducted in 39 homes, however three of these residences were student dormitories, which were not considered representative of common home settings and will be presented elsewhere. Further reduction of presented results to 24 homes is due to technical problems with the instruments.

Nanotracer or DiSC data, without reporting any problems, were collected in 22 homes and analyzed in this article (see details in Table 1).

A classification of the ultrafine particle number concentration measurements was carried out by sorting the data into two categories: *occupancy time* (at least one person present in the home) and *non-occupancy time*. The *known* and *unknown activity influenced concentrations* are identified as apparent concentration elevations from baseline concentration, which could, respectively could not, be identified by the residents' notes in the activity logs. The *activity influenced period* was defined as starting at the time when concentrations started to increase and ending at the time when concentrations had returned to baseline, thus it does not represent the duration of given activity but rather the periods of elevated concentrations being the effect of it. During *non-activity influenced periods*, it was assumed that the particles indoors originate mainly from outdoor sources.

Approximate source strengths, S, of ultrafine particles were estimated (Table 2) for 7 of the 13 activity types in Fig. 1, using a method described by Wallace and Ott (2011):

$$S = \frac{C_{max}\, V}{t}$$

where S is the source strength or emission rate (particles/min), C_{max} is the maximum concentration (cm^{-3}), V is the mixing volume (cm^3) and t is the time (min) during which a source is on. Activity logs did not provide information of when a source was turned off; instead t was approximated to the time from a start in concentration elevation to the corresponding peak concentration was reached. Everything which was elevated above the baseline, was possible to identify from the activity logs and did not overlap with other peaks, was counted as a peak. It was assumed that the entire volume of the house/apartment was a single well mixed volume.

From the particle number concentration data, the integrated residential exposure was calculated by integrating the concentration over time (particles/cm^3 h). The total integrated daily residential exposure was calculated by dividing the integrated residential exposure by the measurement period (in days) resulting in units of particles per $cm^3 \cdot$ h/day (Bhangar et al., 2011; Mullen et al., 2011; Wallace and Ott, 2011). The total daily integrated residential exposure was calculated for periods of activities and periods of non-activities.

Due to that the Indoor Air Quality monitor reported flow problems in several cases, data from only 9 homes (of which 7 matched the abovementioned 22 homes), where the instrument ran without complications for the whole measurement duration, could be evaluated. This data was sorted into occupancy time and non-occupancy time. The six channels were then compressed to $PM_{1-0.3}$ and PM_{10-1}, since these fractions match the accumulation mode and the coarse mode fairly well (Lundgren and Burton, 1995; Morawska et al., 2008).

In 10 of the 22 houses mentioned above, soot data from microAeth was analyzed. When the sampling time is short and/or the soot concentration is low, noise from the instrument can cause attenuation values to be unchanged or even decline, from one period to the next. This may result in an erroneously low value at one time followed by an erroneously high value at the next (Cheng and Lin, 2013). Noise obstructs the actual variations in soot concentrations and makes comparison of the microAeth data and the data from Nanotracer and DiSC difficult. A noise-reduction averaging (ONA) algorithm based on a user-specified minimum change in attenuation over a time interval, developed and described in detail by Hagler et al. (2011) and evaluated by Cheng and Lin (2013) was used to eliminate the negative values and reduce the noise, while maintaining high time resolution.

3. Results and discussion

3.1. Ultrafine particles and residents' activities

The data analysis was focused on the results from the ultrafine particle measurements by the Nanotracer and DiSC.

Occupancy time occurred during an average of 77% of the measurement period. The geometric mean of the concentration of ultrafine particles during occupancy time was 4500 cm^{-3}, with a lower quartile of 1200 and an upper quartile of 17 000 cm^{-3} (average ± standard deviation: 20 500 ± 99 000 cm^{-3}), and during non-occupancy time 2400 [1200, 4800] cm^{-3} (average ± standard deviation: 3200 ± 5100 cm^{-3}). The origins of elevated concentrations were identified using the activity logs from the 22 homes in Table 1. Activity influenced periods comprised on average 33% of the occupancy time.

Table 2
Source strengths, S, estimated for 7 activities.

Activity	Number of peaks	Peak concentration [#/cm^3] (std)	Mean S, [#/min] (std)
Boiling	11	$5.6 \cdot 10^4$ ($3.5 \cdot 10^4$)	$1.7 \cdot 10^{12}$ ($1.4 \cdot 10^{12}$)
Frying	8	$1.4 \cdot 10^4$ ($1.3 \cdot 10^4$)	$1.6 \cdot 10^{12}$ ($1.7 \cdot 10^{12}$)
Oven	10	$2.3 \cdot 10^5$ ($1.2 \cdot 10^5$)	$2.4 \cdot 10^{12}$ ($2.6 \cdot 10^{12}$)
Hairspray	3	$2.4 \cdot 10^4$ ($1.3 \cdot 10^4$)	$3.1 \cdot 10^{12}$ ($1.4 \cdot 10^{12}$)
Cleaning	3	$2.8 \cdot 10^5$ ($9.3 \cdot 10^4$)	$2.4 \cdot 10^{12}$ ($1.7 \cdot 10^{12}$)
Laundry	1	$2.5 \cdot 10^4$	$4.5 \cdot 10^{12}$
Toaster	3	$1.6 \cdot 10^5$ ($1.2 \cdot 10^5$)	$1.9 \cdot 10^{12}$ ($1.3 \cdot 10^{11}$)

Several of the periods with elevated concentration were results of a combination of two or more particle generating activities. 13 different single activity types, shown in Fig. 1, could however be identified as contributing significantly to the indoor air particle concentration.

Candle burning and activities related to cooking (using a frying pan, oven, toaster, and their combinations) were the major particle sources. The elevated concentrations of ultrafine particles when cleaning (i.e. dusting, vacuum cleaning and mopping) are likely to be originating from the motor of the vacuum cleaner (Lioy et al., 1999; Afshari et al., 2005). Increased indoor concentration due to opening a window cannot be considered as an indoor source, but is in this case given for comparative purposes.

The contribution of ultrafine particles due to activities was further studied by comparing the activity influenced time data to the outdoor (ambient) concentrations of ultrafine particles as extracted from the SMPS measurements. Due to that the SMPS and the Nanotracer/DiSC have very different time resolution, it is difficult to visualize them in the same figure (the number of measurement points from the SMPS is much lower from the Nanotracer/DiSC). This was taken care of by normalizing the data, i.e. the concentrations were divided by the number of concentration measurements done by the SMPS and the Nanotracers, respectively. Data was sorted into concentration bins and the frequency of a given concentration was studied. The resulting figure shows the normalized frequency of a given concentration to occur (y-axis) as a function of concentrations (x-axis). From Fig. 2a and b it is obvious that levels of 10^4 cm^{-3} indoors almost only occur during human presence, which illustrates the huge influence residents have on the air at home. When number concentration data is plotted separately for non-activity influenced periods and activity influenced periods (Fig. 3a and b) the impact of the residents' activities on number concentration of ultrafine particles is even more apparent. The indoor air particle concentration during non-activity influenced periods differs very little from non-occupancy time.

The particle source strengths ranged from $1.6 \cdot 10^{12}$ to $4.5 \cdot 10^{12}$ min^{-1}, which is in accordance with the higher range of emission rates reported by e.g. Wallace and Ott (2011). In some studies (e.g. He et al., 2004) lower source strengths were reported.

3.2. Comparison with number concentration of accumulation and coarse mode particles

The results of Indoor Air Quality monitor measurements of mass concentration of particles larger than 300 nm, together with the particle number concentrations measured with Nanotracer or DiSC devices are shown in Fig. 4. Mass concentrations were obtained with assumption of a particle density of 2500 g/cm^3 (proxy for ambient particles). Median PM_{10-1} (corresponding to coarse mode) mass concentration was 17.9 [4.8, 49.9] μg/m^3 during occupancy time and was about 6 times higher than during non-occupancy time 3.3 [2.0, 9.2] μg/m^3. The median $PM_{1-0.3}$ (accumulation

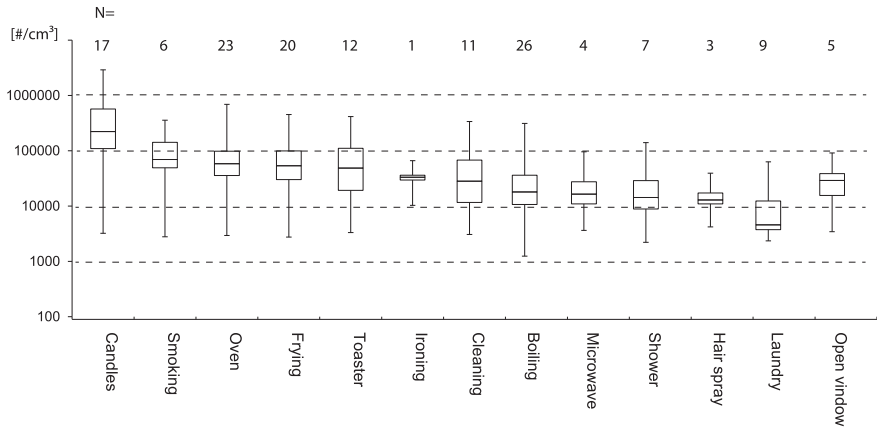

Fig. 1. Summary of ultrafine particle number concentrations from various activity influenced periods. The boxes denote the quartiles, the horizontal line the medians and the whiskers maximum and minimum concentrations. *N* is the number of identified periods the calculations are based on.

mode) mass concentration showed less differences: 8.0 [4.3, 14.3] μg/m^3 during occupancy time and 5.2 [2.9, 9.0] μg/m^3 during non-occupancy time. The number concentrations of $PM_{10-0.3}$ were very low with median 22 [12, 61] cm^{-3} during occupancy time and 14 [10, 33] cm^{-3} during non-occupancy time. However, despite their low number concentration they contribute considerably to the particle mass (due to their size) and this is the reason why authors decided to present the values as mass concentration in Fig. 4.

Generally, concentrations during occupancy time for accumulation mode, coarse mode and ultrafine were higher in comparison to non-occupancy time. Higher concentrations of coarse mode during occupancy time may reflect resuspension of settled dust

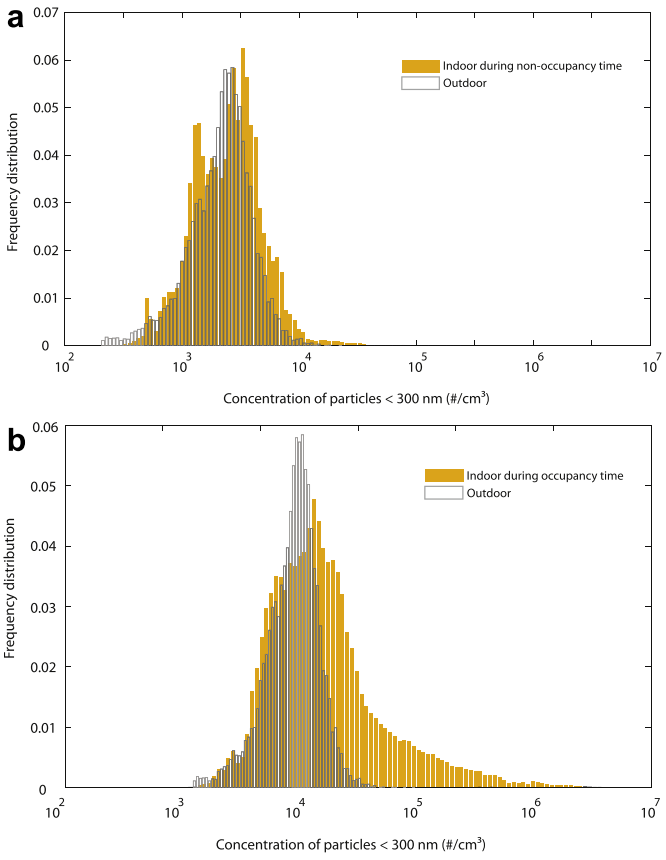

Fig. 2. Occurrence of ultrafine particle number concentrations outdoor (transparent) and indoors (orange) when no one is present in the residences (a) and when at least one person is present (b). Measurement data are unit normalized.

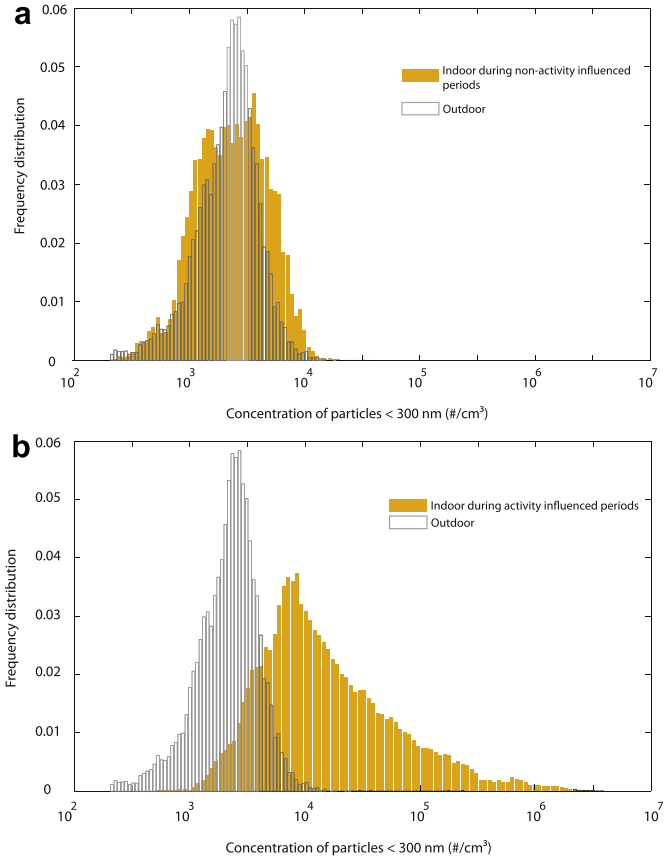

Fig. 3. Occurrence of ultrafine particle number concentrations outdoor (transparent) and indoors (orange) during non-activity influenced periods and activity influenced periods (i.e. times with elevated concentrations including decay of peaks after the activity ceased). Measurement data are unit normalized.

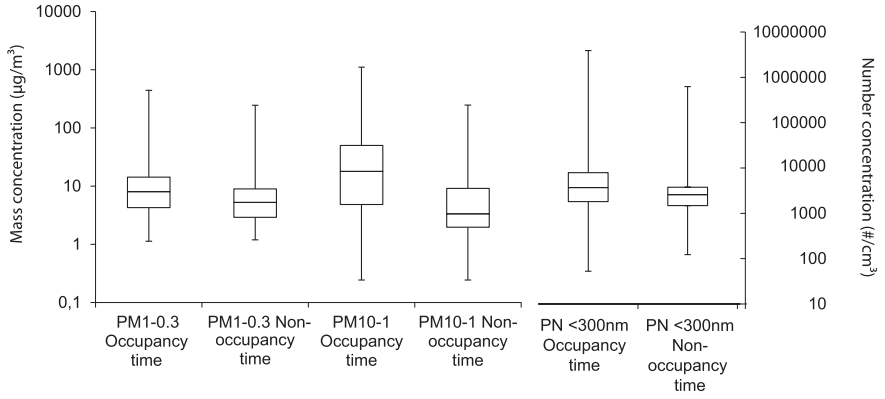

Fig. 4. Boxplots of particle mass concentrations ($PM_{1-0.3}$ and PM_{10-1}) (left) and particle <300 nm number concentration (right) during occupancy and non-occupancy time. The boxes denote the quartiles, the horizontal line the medians and the whiskers maximum and minimum concentrations.

indoors, while higher number concentrations of ultrafine particles reflect the (mainly combustion related) activities of the residents.

3.3. Total integrated daily residential exposure

Fig. 5 shows contribution of known and unknown activities and background/non-activities to the average total integrated daily exposure for 22 residences. The average total integrated daily exposure was $4.0 \cdot 10^5 \pm 3.5 \cdot 10^5$ cm^{-3} h/d, median total integrated daily exposure was $2.7 \cdot 10^5$ cm^{-3} h/d. Of this, the contribution from known activities was $2.7 \cdot 10^5 \pm 2.6 \cdot 10^5$ cm^{-3} h/d (66%) (median $1.8 \cdot 10^5$ cm^{-3} h/d), from unknown activities $1.1 \cdot 10^5 \pm 1.9 \cdot 10^5$ cm^{-3} h/d (20%) (median $2.9 \cdot 10^4$ cm^{-3} h/d), and from background/non-activity $3.6 \cdot 10^4 \pm 1.9 \cdot 10^{-4}$ cm^{-3} h/d (14%) (median $3.2 \cdot 10^4$ cm^{-3} h/d).

Total integrated daily residential exposure was calculated for the different known activities, to determine their specific contribution in each home. In the 22 homes overall, cooking related activities (using a frying pan, boiling oven, micro, toaster and their combinations) contributed to the total daily integrated exposure with 31% and candles with 26%. In 6 of the homes, the contribution from candles were more than 60%, and in two cases candles contribution reached 80%. Candles have been reported earlier as significant contributors to high particle levels indoors (Bekö et al., 2013), especially during winter time in northern Europe.

3.4. Correlation between soot concentration and number concentration of ultrafine particles

Soot indoors may originate from outdoors and/or from combustion related activities indoors. Contribution of indoor sources to soot levels indoors remains not fully understood. By correlating data from microAeth (measures mass concentration of soot in total dust) and ultrafine particle number concentration data from Nanotracer and DiSC, this was investigated.

Using the ONA algorithm (Hagler et al., 2011) to reduce the noise in the soot data, shown in Fig. 6a, enabled comparison between the two instruments, as seen in Fig. 6b.

As shown in Fig. 6b, a majority of the peaks identified in the Nanotracer data could be identified also in the microAeth data, suggesting that the major part of the soot in this home was actually generated indoors. Correlation factors were calculated for 10 homes by first calculating averages of the Nanotracer/DiSC data (data sampled every 16 s) to match the data of the microAeth (data sampled every 60 s). Correlation coefficients, r, between the two data sets were then calculated as:

$$r = \frac{1}{n-1}\sum_{i=1}^{n}\left(\frac{X_i - \overline{X}}{s_X}\right)\left(\frac{Y_i - \overline{Y}}{s_Y}\right)$$

where X_i and Y_i are the logarithms of the values from the Nanotracer/DiSC and microAeth respectively, n is the number of data points in one set, and $\overline{X}$ and $\overline{Y}$ (s_X and s_Y) are the corresponding means (standard deviations).

The correlation between ultrafine particles and mass concentration of soot in total dust in the 10 homes varied between 0.37 and 0.85, with an average of 0.56. In 3 homes, the correlations were higher than 0.60, in these homes a majority of the activities consisted of cooking, candle burning (in two of them) and cigarette smoking (in one).

The correlations of the instruments for identified activities were checked and are summarized in Table 3. The activities for which correlation is the highest coincide with activities which generate the highest number concentrations of ultrafine particles (Fig. 1). Soot mass concentration measurements correlate to measurements of ultrafine particles above 0.60 in the case of smoking cigarettes, burning candles, and cleaning. The relatively high correlation in the case of cleaning (0.64) is most likely due to the particles emitted from the engine of the vacuum cleaner. Correlation was also found for using microwave (0.61 ± 0.11) based on two events occurring in the same home. Due to that this may have resulted from a malfunctioning microwave it is not included in Table 3. Three other events (toaster, laundry and printer, with correlations of 0.56, 0.46 and 0.38, respectively) only occurred once, and are hence left out from Table 3.

The calculated correlations suggest that a significant part of the soot indoors originate from outdoors, however, a limitation that should be considered is the different size ranges covered by the two instruments.

3.5. Limitations

It is crucial to be able to identify and characterize the sources. In this study, this has been done by asking the residents to fill in detailed activity log books. These documents are not fully reliable (difficult to interpret, lack of consistency, parts not filled in etc.), thus they are rather a blunt means of gathering important information, and probably the major limitation of this study. For future studies, use of some other way of source identification, such as sensors giving information about operation of stove, oven, toaster, window, door opening, maybe in combination with GPS equipment worn by occupants allowing accurate location determination

(occupancy/non-occupancy time), could be recommended. If all activities (or at least the major part) could be known (see Fig. 5) the concentrations inside the residences could be modeled based on information of the resident's living habits, considering that the major contribution to observed concentrations in residences is influenced by the habitant's activities.

Another limitation of the study is the accuracy of used instruments. Asbach et al. reported that the Nanotracer overestimated particle number concentrations by a factor of ~7 for di(2-ethylhexyl) sebacate particles with a modal diameter of 180 nm, which is above the instrument's upper range for the mean diameter (120 nm). However, chamber experiments with candle smoke (resulting in the highest concentrations measured in this study) proved the opposite, i.e. underestimation of measured concentration by Nanotracer for the sizes between 20 and 120 nm (Bekö et al., 2013). Only 20% of measurement points in this study showed measured average diameter above 120 nm (mainly occurring during non-activity time), thus it is believed that presented results are not overestimated more than the assessed instrumental accuracy i.e. 30%.

One has to bear in mind that the comparison between the aethalometer and Nanotracer measures two different size ranges and two different particle properties. We know from outdoor studies that a majority of soot is in the ultrafine particle range, which was the basis of the assumption that the two instruments could be worthwhile comparing. The correlation showed that soot also, but not exclusively, originate from indoor sources.

4. Conclusions

Due to the importance of indoor sources, residential exposure to ultrafine particles cannot be characterized by ambient measurements alone, which has been clearly shown in this study. Indoor sources are of a transient nature. They quickly generate high peaks in concentration which decay at a rate mainly determined by the air exchange rate and deposition on interior surfaces (and, in cases of number concentrations higher than 10 000–20 000 #/cm^3, by coagulation). It is not fully understood whether it is the long term mean exposure or the events with elevated concentrations that has the greatest impact on human health. Until this has been further investigated and determined, it is of importance to gather exposure data based on high time resolution instruments. Comparison of ultrafine particle number concentration frequency distributions for 22 studied homes showed that outdoor concentrations can be used to describe concentrations in residences only during the time when occupants are not at home (which is not interesting from the exposure assessment perspective) or when they are not involved in any activities. Concentrations indoors during times influenced by activities, which comprise 33% of time spent at home, are

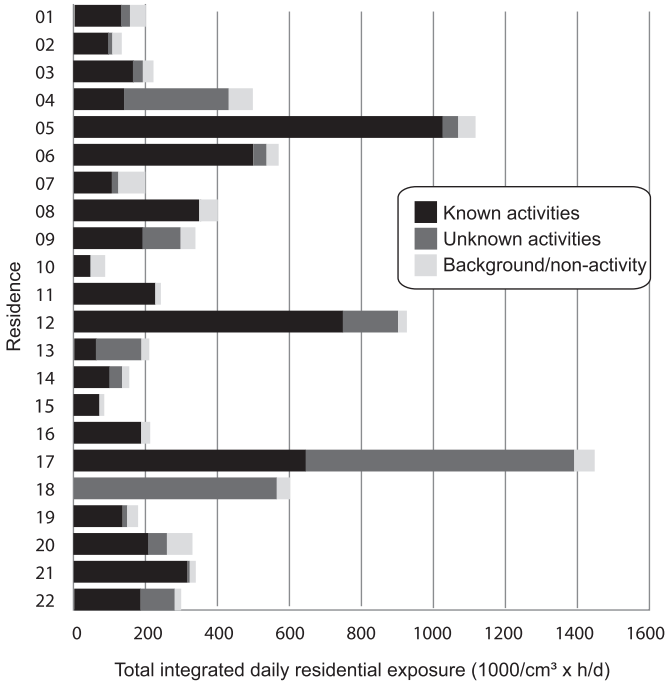

Fig. 5. Total integrated daily residential exposure for 22 residences and the contribution of time influenced by known and unknown activities and background/non-activity.

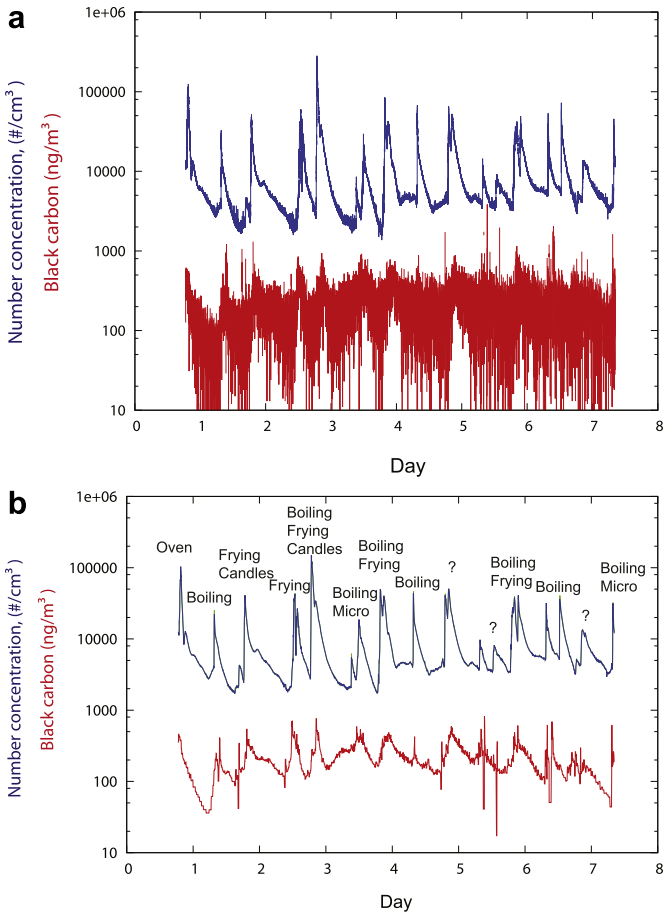

Fig. 6. Non-post-processed microAeth data (red) and Nanotracer data (blue) from one of the studied residences (a). Post-processed microAeth data (red) and Nanotracer data (blue) from the same residence (b).

Table 3
Correlation between number concentrations of ultrafine particles and mass concentration of soot in total dust during specific activities.

Activity	Number of events	Mean correlation ± standard deviation (%)
Cigarettes	2	0.74 ± 0.0 9
Candles	13	0.64 ± 0.26
Cleaning	6	0.64 ± 0.27
Frying	7	0.49 ± 0.49
Oven	6	0.46 ± 0.36
Open window	4	0.44 ± 0.38
Boiling	14	0.36 ± 0.35

significantly different ($p < 0.01$). In epidemiological studies, it is not uncommon to estimate the exposure of a population based on measurements from one outdoor station. If this approach would be used in the case of the homes studied here, the exposure estimation would only be approximately relevant when no activities at all are conducted at home (see Fig. 3a), i.e. mainly when people are asleep.

Integrated daily residential exposure of particles <300 nm was on average $4.0 \cdot 10^5 \pm 3.5 \cdot 10^5$ cm^{-3} h/d, where known indoor activities contributed with 66% to the integrated daily residential exposure, unknown activities 20% and outdoor particles 14%. These numbers are in agreement with results published by Bekö et al. (2013), Bhangar et al. (2011), and Wallace and Ott (2011). If unknown activities were not included as indoor sources, Bekö et al. (2013) found a 51% contribution from known activities, and a 65% contribution from known + unknown activities. The difference between known indoor contribution and unknown indoor contribution will depend on how unknown activities are classified in the different studies. We only included an indoor activity among the known if it was specifically stated in the activity log. Several of the peaks classified as "unknown" are however most likely originating from indoor activities. A typical example is a peak occurring at the same time every morning, which is only the first couple of days stated as "cooking breakfast". It should be noted that this study was performed in Northern Europe where outdoor air pollution is relatively low, and during winter time, when it is common to burn candles (which contribute significantly to the particle number concentration).

Based on the correlation calculations of soot and ultrafine particles, one cannot draw conclusions that measurements of ultrafine particles could be used for predicting soot levels indoors. If there would be only one source present, which generates a known fraction of soot, this approach can be relevant. However, in indoor settings, there is a variety of sources present, which can be operated under different conditions from time to time, and from household to household, resulting in very different ultrafine particles-to-soot ratios. Often elemental carbon is used as proxy of exposure to diesel exhaust particles. Interestingly enough, this study has shown that indoors a large part of the soot has indoor sources, and clearly elemental carbon should not be treated as a tracer for particles originating from outdoors.

Strictly speaking, the results of this study can only represent the 22 households studied during the time period in the winter when the measurements were conducted. The households were chosen from randomly selected addresses in and in the vicinity of Lund. However, only households where inhabitants accepted to be included in the studies became objects of measurements, and it may be possible that people willing to participate in scientific studies not necessarily represent an average of households in the community. There are, however, other studies that support the general conclusion that periods of various activities in dwellings dominate the sources which cause the major exposures of the inhabitants. More studies are needed in order to be able to generalize on a national, continental or global scale.

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