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# Measurements of fine and ultrafine particles in Helsinki: connection between outdoor and indoor air quality

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In this paper, we present aerosol number concentration and number size distribution measurements in the urban area of Helsinki, Finland. The outdoor air measurements were performed over several years between 1999 and 2003 giving a rather good overview of the general properties of fine aerosols as well as their daily, weekly and seasonal characteristics. The aerosol number concentrations in urban areas were significantly higher in winter as compared with those in summer. The morning rush hour during workdays was observed as an average peak concentration of more than $4 \times 10^4$ cm$^{-3}$, but no clear afternoon/evening peak was observed. Typical number size distributions were calculated for the winter and summer seasons separately. During the winter the morning peak in the size distribution in urban sites was found always between 10 and 50 nm in particle diameter. These particles were probably from local traffic emissions. The summer size distributions showed that the ultrafine (below 100 nm) size mode had often two separate peaks, one below 20 nm and the other at about 50 nm. The indoor air measurements have been done during several intensive field studies in various locations around Helsinki. The simultaneous indoor and outdoor aerosol size distribution measurements showed that a good quality filter can reduce the concentrations by a factor of about 10 and therefore influence the human exposure to urban particulate air pollution significantly.

## Introduction

In recent years a number of studies have focused on properties of ambient particulate matter due to its major public health concern. Elevated concentrations of aerosol particles have been associated with increases in mortality as well as adverse health effects (e.g. Dockery and Pope 1994, Donaldson *et al*. 2001, Pope *et al*. 2002, Metzger *et al*. 2004). Ultrafine particles (particles smaller than 100 nm in aerodynamic diameter) have only small contribution to total aerosol mass, but they often dominate the total number of particles. Ultrafine particles can easily be inhaled and deposited in the deeper regions of the respiratory tract and can therefore pose a health risk. During recent years, both toxicological and epidemiological studies have indicated that a high number concentration of ultrafine particles may cause serious health effects (e.g. Oberdörster *et al*. 1995, Pekkanen *et al*. 1997, Oberdörster 2000, Wichmann and Peters 2000). Therefore, an increasing number of papers focusing on the aerosol number concentrations and number size distribution in polluted urban areas have been published (e.g. Bukowiecki *et al*. 2003, Charron


**Fig. 1**. The map of Helsinki metropolitan area showing the measurement sites in the study: Siltavuori (UC1, Urban-city), Pasila (UR2, Urban-rooftop), Viikki (UBT3, Urban-background) and Friisilä (SU4, Suburban).

and Harrison 2003, Hussein *et al*. 2003, Wehner and Wiedensohler 2003).

Aerosol properties in urban areas are shown to be influenced strongly by traffic, but other anthropogenic sources such as domestic heating and local power and heating plants are also contributing significantly to the total aerosol (Wehner and Wiedensohler 2003). The relative importance of various sources depends strongly on the actual sampling site and especially its location in relation to major traffic lines. The number concentration that is dominated by local sources is shown to have strong gradients across urban areas (e.g. Buzorius *et al*. 1999). Therefore several studies focused specifically on either traffic sites (e.g. Shi *et al*. 2001, Molnar *et al*. 2002, Morawska *et al*. 2002, Zhu *et al*. 2002) or urban background sites located further away from nearby sources, such as traffic, and represent therefore better the overall situation in a specific city (e.g. Hussein *et al*. 2003, Wehner and Wiedensohler 2003).

One of the powerful tools in investigating the role of various aerosol sources in urban areas is the measurement of number size distributions. Traffic is known to produce primary particles with mean diameter of about 70–100 nm. In addition, traffic is responsible for producing high numbers of freshly nucleated particles with diameter close to 10 nm. These particles are formed after the tail pipe of cars in the atmosphere and their formation rate has been shown to depend on both the type of the vehicle and the meteorological conditions (e.g. Charron and Harrison 2003, Wehner and Wiedensohler 2003). On the contrary, the urban background aerosol has been shown to be rather uniform over relatively large areas forming the accumulation size mode between 100 and 1000 nm.

Estimating the exposure to ambient particulate matter requires information on the activity patterns of people within the urban area as well as the aerosol properties in all the microenvironments in which significant amounts of time are spent. It is known that a significant fraction of time is spent indoors, at home or at work. Therefore, it is important to study the transport of outdoor pollution indoors as well as the contribution of the indoor sources (Hussein *et al*. 2002, Koistinen *et al*. 2004).

In this paper, we present results of aerosol number concentration and size distribution measurements in the urban background of Helsinki (Finland). The outdoor air measurements were performed between 1999 and 2003 giving a rather good overview of the general properties of fine aerosols as well as their daily, weekly and seasonal characteristics. An overview of the whole data on our outdoor measurements is presented elsewhere by Hussein *et al*. (2003). The indoor air measurements were done during several intensive field studies in various locations around Helsinki. Therefore, the indoor studies have to be considered as case studies. Our aim in this paper is to characterise the main properties of fine and ultrafine aerosol particles in an urban area of Helsinki. This data is then connected with studies on outdoor–indoor relationship and consequences on human exposure are discussed.

## Experimental methods

The measurements were carried out in the Helsinki metropolitan area, which has a population of about 1 million people (Fig. 1). The measurements were performed at two downtown sites at elevated locations, at one urban background site (influenced by highway traffic) and at a suburban site. All of these locations (except the highway site) are distanced from local sources and they

are representative of city-wide background concentrations.

The investigations focus on general characteristics of the fine and ultrafine aerosol properties. Therefore the data were presented as average/typical values that are based on large data sets. In general, the concentrations and size distributions varied significantly as a function of time, indicating a clear influence of both variations in emissions and changes of meteorological parameters such as the local wind direction or temperature. When calculating the average concentrations and size distributions, representative time periods were always selected. In the following the basic information on the instrumentation as well as the measurement sites are given.

### Instrumentation

The number size distributions were measured in the particle size range between about 8 and 800 nm with a differential mobility particle sizer (DMPS). During different measurement campaigns the size range varied due to technical details, but the main number size modes were measured in all cases. The DMPS consists of a differential mobility analyser (DMA) that classifies aerosol particles according to their electrical mobility and a condensation particle counter (CPC) that detects particles of different mobilities. The mobility is then converted to a particle diameter. The actual models of the DMA and CPC varied between measurements, but all instruments were systematically checked and calibrated to ensure the comparability between the different measurements.

### Site descriptions and measurement periods

The urban sites were UC1 (urban-city: Siltavuori, on a hill some 100 m from the closest street) and UR2 (urban-rooftop: Pasila, at the roof top of a 30-m-high building surrounded by streets). At the urban-city site (UC1) the sampling was taken through a window pane at the ground floor. However, the window was facing in the direction of the sea and the traffic in nearby streets was not intensive. The measurements were done between May 1997 and March 2001. The urban-rooftop site (UR2) was located north of the urban-city site and it was surrounded by traffic from all sides. The measurements were performed in January 1999.

The UBT3 site (urban-background with traffic influence, Viikki) is some five km north-east of the city center. This site is located close to a highway and influenced by fresh traffic emissions during westerly winds. The measurements were carried out in May and June 2000.

As a reference site for the whole study, we had a suburban site in Friisilä (SU4) which is located some 15 km west from the city center and also west from all the other stations. The measurements were done in February 2001.

## Results

The DMPS data were calculated as average size distributions for the winter and summer seasons separately. For our analysis, we selected only periods representing clearly winter (January–February) or summer (mid May–August). The total number concentration was integrated over the size distribution and corrected for estimated diffusion losses in the sampling lines. Although the actual size range was slightly different during each measurement, the total number concentration is not expected to be significantly affected (*see* also Hussein *et al*. 2003).

The measurements were done in 1997–2001, but they were not simultaneous at all the locations. Therefore, direct comparison between different locations is not possible. However, it has been show earlier (Hussein *et al*. 2003) that the average behaviour from one year to another is relatively similar unless major meteorological differences occur during the measurement period.

### Total number concentrations and number size distributions of outdoor aerosols

A typical aerosol number concentration pattern during one day was investigated. This was done

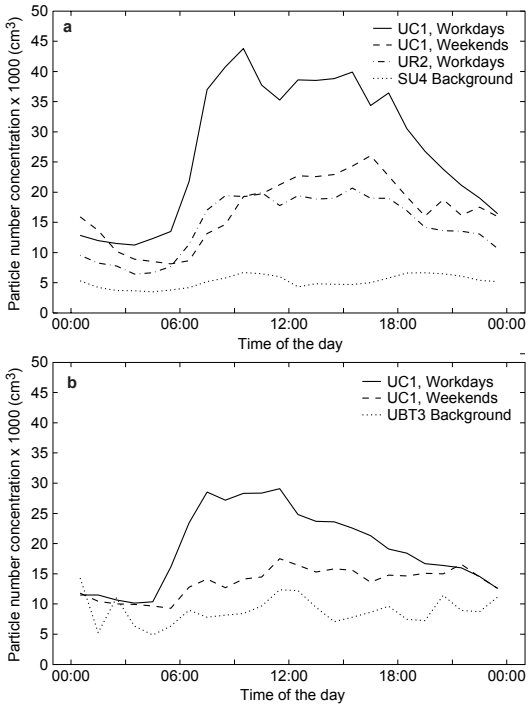


**Fig. 2**. Mean daily patterns of the total particle number concentrations (**a**) in January 1999 and February 2001 representing the wintertime, and (**b**) in May–June 2000 representing the summertime. The sites were UC1 (urban site Siltavuori), UR2 (urban rooftop, Pasila), SU4 (suburban, Friisilä), and UBT3 (urban background with traffic influence, Viikki).

by calculating the average concentration for all the workdays (Monday to Friday) and for the weekends (Saturday and Sunday) separately. The diurnal pattern varied also between the workdays as well as between Saturday and Sunday, but for simplicity this is not considered here. The diurnal behaviour of the concentration was studied separately for the winter and summer seasons (Fig. 2). Several general features are obvious. First of all, the aerosol number concentrations in the urban area of Helsinki were significantly higher during the winter. The morning rush hour during workdays was observed as an average peak concentration of more than $4 \times 10^4$ cm$^{-3}$. The concentration stayed relatively high until the late afternoon, but no clear afternoon/evening peak was observed. The reason for this is probably partly due to increased vertical mixing in the boundary layer. Other studies (Wåhlin *et al.* 2001, Wehner *et al.* 2002) have found a similar pattern for the total aerosol number concentration and related that to differences in the traffic patterns of petrol and diesel vehicles. In comparison, the weekends did not show the morning peak, since the traffic seemed to start later during the day. Also the early morning traffic was notable. The urban rooftop concentrations during the workdays were clearly lower than the downtown concentrations and, probably coincidentally, quite similar to those measured downtown during the weekends. A comparison with corresponding data from suburban site demonstrated clearly the significant difference between the city centre and more remote sites.

While the aerosol number concentrations in the city centre are clearly lower during the summer, the opposite seems to be the case for the suburban concentrations. This conclusion needs to be supported by further research in order to be generalized.

## Characteristics of the size distributions

In order to investigate the influence of the traffic as an important emission source for the aerosol number, the characteristics of the aerosol number size distributions were investigated. As with the diurnal behaviour of the number concentration, also the average size distributions showed clear differences between various sites and times (Fig. 3). These differences indicate the influence of the local emissions as well as dilution and dispersion and their fingerprints in different particle size ranges.

Typical number size distributions were calculated for the winter and summer periods separately (Fig. 3). During the winter the morning peak concentrations in urban sites were found always between 10 and 50 nm in particle diameter. These particles dominated the size spectrum during both workdays and weekends. However, the concentration of such freshly emitted particles varied, as might be expected between the workdays and weekends as well as between the urban-city site and urban-rooftop sites. These particles were probably from local traffic emissions and, therefore, the size distribution was much flatter at the suburban site. The peak value of the size distribution was also shifted to larger

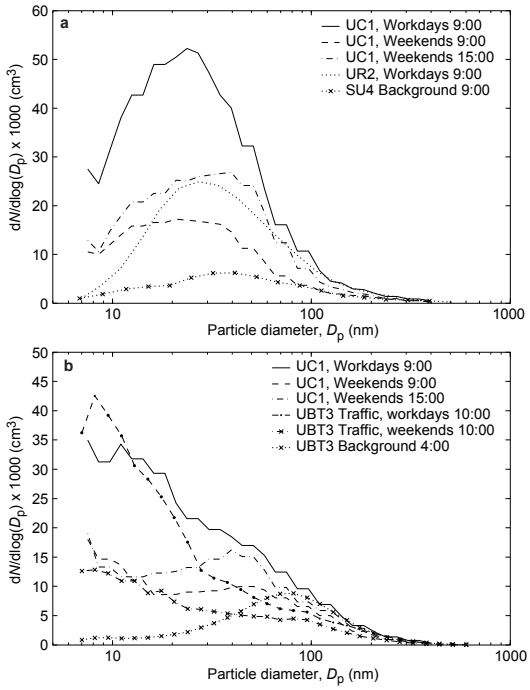


**Fig. 3**. Examples of mean particle number size distributions (**a**) in January 1999 and February 2001 representing the wintertime, and (**b**) in May–June 2000 representing the summertime. The sites were UC1 (urban site Siltavuori), UR2 (urban rooftop, Pasila), SU4 (suburban, Friisilä), and UBT3 (urban background with traffic influence, Viikki).

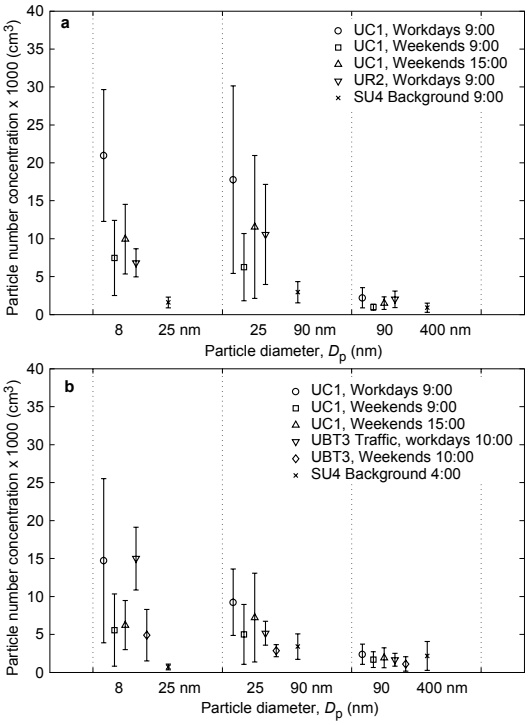


**Fig. 4**. Mean total particle number concentrations within different particle diameter ranges: smaller than 25 nm, between 25 and 90 nm, and larger than 90 nm (**a**) in January 1999 and February 2001 representing the wintertime, and (**b**) in May–June 2000 representing the summertime. The bars represent the standard deviation. The sites were UC1 (urban site Siltavuori), UR2 (urban rooftop, Pasila), SU4 (suburban, Friisilä), and UBT3 (urban background with traffic influence, Viikki).

sizes at the suburban site being indicative of a more aged aerosol. A similar shift was also found by Wehner *et al*. (2002) in their study in the city of Leipzig. In both urban and suburban sites, a more detailed analysis shows the accumulation mode at about 100–300 nm in particle diameter. This mode was relatively similar in all the sites and was probably related to long range transport of particulate pollution.

The summer size distributions were significantly different from the winter ones (Fig. 3). As compared with the winter peak, the ultrafine (below 100 nm) size mode had often two separate maxima. The first maximum was observed at diameter below 20 nm and its peak seemed to be quite often even below the measurement range (below 7–8 nm). The other mode was found at about 50 nm in diameter and it was somewhat larger than the corresponding winter maximum. The accumulation mode was again present in all the sites with relatively similar concentrations and sizes.

The contribution of different size modes to the total aerosol number was studied by integrating the size distribution in three classes: 8–25 nm, 25–90 nm and 90–400 nm. These size classes have been shown earlier to best represent the nucleation, Aitken and accumulation modes, respectively, in the urban air of Helsinki (Hämeri *et al*. 1996, Hussein *et al*. 2003). The high concentrations of the two smallest size classes were observed in all the urban sites (Fig. 4). The highest concentrations were observed during the morning rush hour. Interestingly, the second highest values were observed at 15:00 during weekend afternoons indicating probably the different daily pattern of traffic and its influence on these size classes. The lower values at 09:00

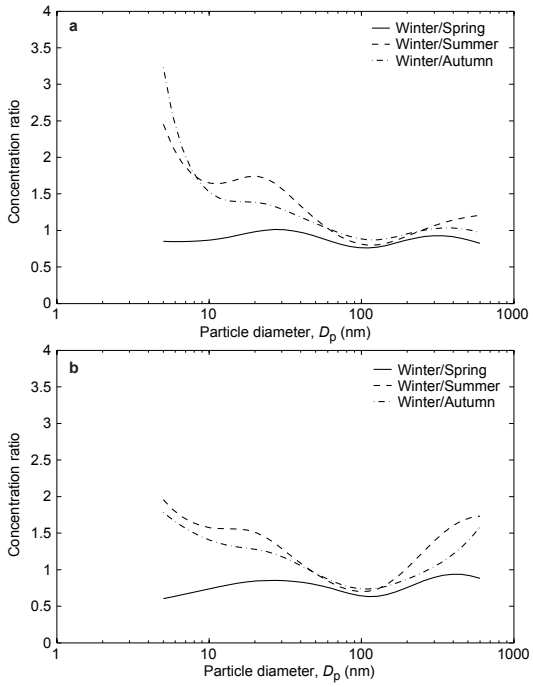


**Fig. 5**. The ratio of average winter size distribution to those of spring, summer and autumn for workdays (**a**) and weekends (**b**) in urban city centre (UC1).

during the weekends were probably partly due to less traffic and partly due to a better mixing of the boundary layer air.

The three size classes were investigated separately for the summer and winter periods (Fig. 4). The summer pattern was similar to the winter data but, as discussed earlier, significantly lower values were observed for both nucleation and Aitken mode. The summer data include also the urban background measurements (site UBT3), which were influenced by a highway. The data were selected for winds from the direction of the highway and highly elevated concentrations of the nucleation and Aitken modes were observed. The concentration levels were almost identical to those in Helsinki city centre.

The seasonal difference was investigated for the city centre site (UC1) by calculating the ratios between the average seasonal size distributions. The average winter size distribution was divided by the corresponding spring, summer and autumn size distributions for both workdays and weekends (Fig. 5). It appears that the winter and spring distributions were relatively similar while the contribution of ultrafine particles smaller than 100 nm in diameter was much smaller in the summer and autumn. In addition, the concentration of accumulation mode particles of about 300–400 nm in diameter was also smaller in the summer and autumn.

The reasons for the difference between the summer and winter data are unknown. Likely explanations include the meteorological parameters such as temperature (which influences atmospheric chemistry, saturation ratio of the vapours and nucleation rate of new particles) or atmospheric dilution (influences the concentration of the emitted vapours and nucleation rate). In addition, local and regional sources and their intensity may differ between the seasons.

## Indoor aerosols

Human exposure to atmospheric particulate matter takes place in all the environments in which people spend their time. These include both outdoor air and indoor air in several conditions (e.g. home, workplace, transportation). In western countries a significant fraction of time is spent inside buildings and therefore outdoor measurements need to be completed to include the transport of the pollutants indoors for different ventilation configurations. Our data set includes long term measurements of indoor/outdoor ratios for the particle number size distribution. The experiments were performed as simultaneous measurements of both indoor and outdoor aerosols. The experiments were done at sites UR2 (ventilation with EU7 class filter), UBT3 (ventilation with EU3 class filter) and SU4 (natural ventilation without a filter). The EU7 is a typical filter for new buildings with a mechanical ventilation system. Thus the experiments include good filtration (UR2), insufficient filtration (UBT3) and undefined filtration (SU4). Our aim was to characterise the general filtration patterns in various ventilation situations. The experiments have been performed in different outdoor situations, but this is expected not to cause significant problems as only indoor/outdoor concentration ratios were examined. The buildings included in our study did not have significant indoor sources for the total number

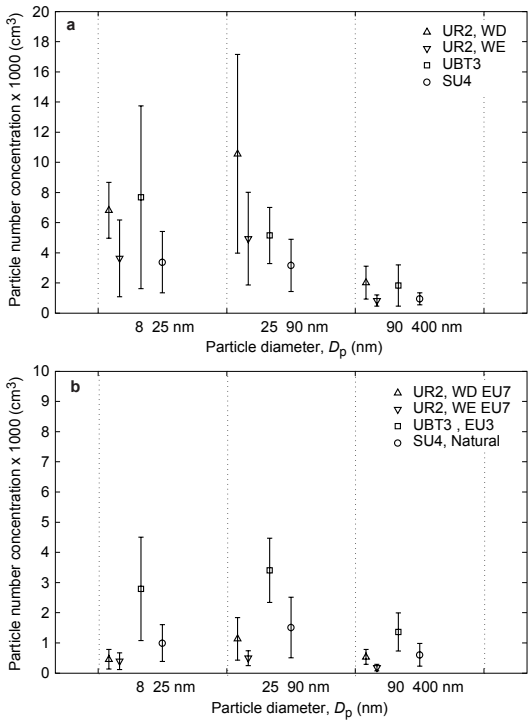


**Fig. 6**. Mean total particle number concentrations within different particle diameter ranges: smaller than 25 nm, between 25 and 90 nm, and larger than 90 nm (**a**) outdoors and (**b**) indoors. The sites were UR2 (urban rooftop, Pasila, during workdays, WD, with high ventilation, larger than 3 h$^{-1}$ and during weekends, WE, with low ventilation, smaller than 1 h$^{-1}$, filter EU7), SU4 (suburban, Friisilä, with a natural ventilation smaller than 1 h$^{-1}$), and UBT3 (urban background with traffic influence, Viikki, ventilation rate about 3 h$^{-1}$, filter EU3).

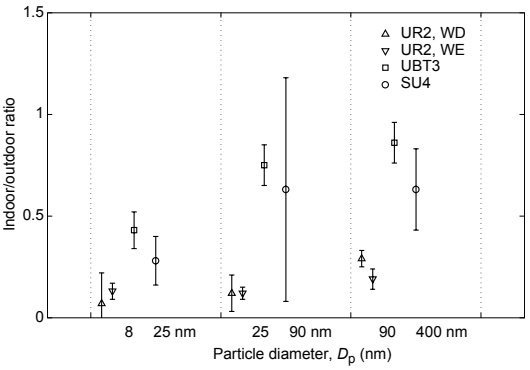


**Fig. 7**. The indoor/outdoor ratio for nucleation mode, Aitken mode and accumulation mode particles for the data plotted in Fig. 6.

concentration with the exception of the site SU4, which was a family house. For this site the situations with notable indoor sources were removed from the data before the analysis.

A summary of size classified concentrations at the study sites is shown in Fig. 6. The size distributions were used to calculate the concentrations in three size fractions: a nucleation mode (8–25 nm), Aitken mode (25–90 nm) and accumulation mode (90–400 nm). The urban site (UR2) was divided into workdays and weekends, while the concentrations for the other two sites were calculated for the whole time period. It is clearly seen that UR2 (good ventilation) showed good filtration efficiency for the two lowest sizes, the filtration efficiency being about 90% or more (Fig. 7). The accumulation mode, instead, showed a weaker filtration of about 70%–80%.

The traffic-influenced site (UBT3) displayed the highest concentrations for the nucleation mode outdoors (Fig. 6). With insufficient ventilation, the concentration indoors was rather high as well. This was the case for the other two modes as well, and the ratio was approaching the value of unity as the particle size increased (Fig. 7). The suburban site (SU4) showed the lowest outdoor concentrations for all the modes. However, the indoor concentrations were relatively high indicating the lack of any active filtration.

## Discussion and conclusions

This study shows that the aerosol particle number concentration in the urban area of Helsinki was dominated either by fine (smaller than 1.0 $\mu$m in diameter) or ultrafine (smaller than 0.1 $\mu$m in diameter) particles depending on the site and time. These particles have little mass due to their small total volume and therefore they are not detected efficiently by applying a traditional mass concentration monitoring.

The aerosol number concentrations in urban areas were significantly higher in winter as compared with those in summer. The highest concentrations were observed during high traffic situations. The morning rush hour during workdays was observed as an average peak concentration of more than $4 \times 10^4$ cm$^{-3}$. Traffic-generated particles dominated sizes below 50 nm. The peak

size could be below 10 nm during fresh traffic emissions. Particles of such small diameter are proposed to have significant health consequences and need advanced instrumentation for a precise determination. Their concentration, however, has been observed to vary significantly over short distances due to a strong contribution of local emissions.

Another important aspect of different particle size classes is their different penetration characteristics inside the human respiratory tract. The maximum penetration occurs in submicrometer size range with increasing deposition on the alveolar region towards ultrafine particle sizes.

The particulate exposure cannot be sufficiently estimated by applying data from a single measurement site only. A set of measurements is needed for characterising the spatial variation within an urban area. Also advanced modelling is needed for estimating the exposure to fine and ultrafine particles. In addition, the outdoor-to-indoor transport need to be studied and taken into account in estimating the total daily exposure. It can be estimated from the data of this paper that using a good filtration, the daily exposure can remain at about 10% of the simultaneous outdoor values. This level of filtration is realistic in buildings equipped with a modern mechanical ventilation system. These results have important consequences in designing ventilation details in heavily-polluted urban environments.

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